The diode-pumped continuous-wave Raman laser: classical, Quantum, and thermo-optic fundamentals
by Peter Aaron Roos

A thesis submitted in partial fulfillment of the requirements of the degree Masters of Science
Montana State University
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Abstract:
This thesis describes the creation and development of far-off-resonance diode-pumped cw Raman lasers using high-finesse cavity enhancement from both theoretical and experimental standpoints. Comprehensive theoretical models are developed for the cw Raman system in the classical and quantum mechanical domains. The classical treatments include two separate derivations of the time-dependent cw Raman equations (one as an appendix), derivations of the analytical steady-state threshold power, impedance-matching power, all emitted powers, and conversion efficiency, as well as the system optimization with respect to every parameter available, and a useful frequency tuning picture (as an appendix). An entire chapter is devoted to the classical treatment of thermo-optic effects in this laser system including mode pulling and thermal lensing.

The quantum mechanical treatment uses the Heisenberg/Langevin approach to derive the Stokes output noise spectra analytically for this system. Subsequent analysis reveals the possibility of approaching perfect photon statistics transfer from the pump to the Stokes and generating nonclassical photon number statistics. An interesting and useful connection to the nondegenerate optical parametric oscillator is uncovered. The quantum mechanical approach is also used to show that a similar system can generate efficient cw anti-Stokes emission.

The process of constructing these laser systems and the subsequent measured behavior comprise the experimental component of this thesis. Diode laser frequency locking for this system is treated thoroughly as an exercise in precision frequency stabilization and control theory. The optimal designs of the diode laser pump source, the high-finesse Raman laser cavity, and the electronic servo are discussed. The use of injection locking and passive optical frequency stabilization are also addressed.

Using these construction and locking techniques, the results from diode-pumped cw Raman laser experiments in diatomic hydrogen gas are provided and are shown to compare favorably with the theoretical predictions. The results include steady-state data obtained from the first diode-pumped cw Raman laser, the first high efficiency cw Raman laser, a cw Raman laser pumped by an injection-locked diode laser, and the first cw Raman ring laser. Data confirming the effects of thermal mode pulling and lensing are also provided.
THE DIODE-PUMPED CONTINUOUS-WAVE RAMAN LASER: CLASSICAL, QUANTUM, AND THERMO-OPTIC FUNDAMENTALS

by

Peter Aaron Roos

A thesis submitted in partial fulfillment of the requirements of the degree Masters of Science

MONTANA STATE UNIVERSITY
Bozeman, Montana

October 2002
APPROVAL

of a dissertation submitted by

Peter Aaron Roos

This dissertation has been read by each member of the dissertation committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies

John L. Carlsten  

(Signature)

10-14-02  

Date

Approved for the Department of Physics

John C. Hermanson  

(Signature)

10-14-02  

Date

Approved for the College of Graduate Studies

Bruce R. McLeod  

(Signature)

10-16-02  

Date
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ACKNOWLEDGEMENTS

I must first thank my partner, Jennifer, my sister, Bonnie, and my parents, Kathy and Roger for their unwavering love and encouragement throughout my graduate work and especially while I was writing this thesis. They have been incredibly supportive during one of the most difficult, but rewarding times of my life.

I also thank Sally O’Neill for her critical guidance and assistance during the Fulbright application process. As a direct result of her efforts, I can also thank Tim Ralph, Andrew White, Rodney Polkinghorne, Michael Nielsen, Joe Altepeter, and all the folks at the University of Queensland physics for helping to make my experience in Australia one of the best of my life. This experience (the guidance of Tim and Andrew in particular) also led to the findings presented in Chapter 3.

I am grateful to Norm in the shop for teaching me basically everything I know about machining, as well as Margaret, Jeannie, Rose and Jeremy in the office for their assistance with my day-to-day activities. These people have kept me and the whole department running smoothly.

Thanks to Tom and Pete for their friendship and advice with frequency locking. These are two very talented physicists who have thankfully not lost their appreciation nor their devotion to the wonderful outdoors.
Thanks go to Sytil, Tina, Brian, and Jennifer for making the past year so enjoyable in the lab. Our lunchtime discussions will be sorely missed. Furthermore, Sytil was kind enough to edit this entire thesis. But more importantly, she played a significant role in the findings of Chapter 5. I had a wonderful time working with her this summer.

I thank Monty for his friendship and for keeping me sane with daily climbing or bouldering excursions. We shared many good times and laughs together. We’ve never discussed physics with each other, and I’m sure we never will. True lifelong dirtbag friends like Monty are impossible to replace.

I am also thankful to Randy for his friendship, climbing, and physics discussions during our graduate years. I greatly admire Randy’s ability to balance his roles as a caring father and husband with his amazing skills and productivity as a physicist.

I give special thanks to my predecessor Jay not only for his true friendship, but also for giving me such an incredible head start in the field of cw Raman lasers. Jay has an extraordinary gift as an experimentalist in the lab. He is the most natural person in that environment that I’ve ever seen. I can only hope that some of his talent rubbed off on me. I’ll always enjoy working with Jay in the lab, not to mention shooting pool and drinking beer with him at the bars. He’s an all-round friend.

I am grateful to Josh Bienfang for some of the most stimulating physics discussions that I’ve had as a graduate student. Josh has an ability blend rock-solid mathematics and theory with superb experimental skills to yield unparalleled understandings of
physics. Of my contemporaries, he is the most talented all-round physicist that I know of (even though we have yet to actually meet in person). Josh played a *major* role in the development of Appendix B and Chapter 4. I’ve learned a great deal from our interactions and I truly hope they continue in the future.

I also thank Lei for his wonderful companionship and friendship during our graduate work. We shared both lab space and living space while writing our theses. During my year in Australia, I realized how valuable our day-to-day discussions on physics really were. Those interactions will be missed in the future. Almost every experiment in Chapter 6 was conducted with Lei. His dedication and attention to detail make him a superb physicist. His thoughtfulness and sense of humor make him an genuine friend.

Last, but definitely not least, I thank my advisor, John. This guy has undoubtedly had the single most profound influence on my development as a physicist. But the wonderful thing about John is that his teachings transcend physics. He has dramatically influenced my thoughts and philosophies about life as well. John magically combines knowledge, experience, skill, and compassion like no one I’ve ever met. He has provided guidance beyond anything I could have asked for. He will remain a cherished colleague and friend throughout my life.
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The process of constructing these laser systems and the subsequent measured behavior comprise the experimental component of this thesis. Diode laser frequency locking for this system is treated thoroughly as an exercise in precision frequency stabilization and control theory. The optimal designs of the diode laser pump source, the high-finesse Raman laser cavity, and the electronic servo are discussed. The use of injection locking and passive optical frequency stabilization are also addressed.

Using these construction and locking techniques, the results from diode-pumped cw Raman laser experiments in diatomic hydrogen gas are provided and are shown to compare favorably with the theoretical predictions. The results include steady-state data obtained from the first diode-pumped cw Raman laser, the first high efficiency cw Raman laser, a cw Raman laser pumped by an injection-locked diode laser, and the first cw Raman ring laser. Data confirming the effects of thermal mode pulling and lensing are also provided.
Stimulated Raman Scattering: A Brief Review

The stimulated Raman effect was discovered in 1962 [1], shortly after the advent of the laser. One can view this two-photon process most simply as an exchange of "pump" photons for lower energy "Stokes" photons through a stimulated interaction with a Raman-active medium as illustrated in Fig. 1. The energy difference between the incident pump and stimulated Stokes photons resides within the Raman medium after the interaction, often in the form of a molecular vibration.

Figure 1. Simple picture of the stimulated Raman process. An incident pump photon is converted to a lower energy (longer wavelength) stimulated Stokes photon and the leftover energy remains in the Raman medium.
This process can result in exponential growth (amplification) of input Stokes light at the expense of the input pump light. But even when there is no input Stokes, the process can be initiated by spontaneous emission alone. In other words, by simply directing an intense pump laser through a Raman medium, some of the pump light will be converted to the Stokes frequency, as shown in Fig. 2. Stimulated Raman scattering (SRS) therefore provides, among other things, a valuable method of down-shifting the frequency of laser light. However, this stimulated process differs from parametric conversion processes in that the phase of the generated Stokes light doesn’t directly depend on that of the pump. Due to this phase-insensitive amplification, the system shown in Fig. 2 is often considered to be an optically-pumped laser; more specifically, a Raman laser.

Figure 2. Frequency downconversion using the stimulated Raman process. Because this involves phase insensitive amplification of the Stokes light, the system shown is often called a Raman laser. The laser action is initiated by spontaneously emitted Stokes photons.
However, due to its third-order nonlinear nature, the stimulated Raman process requires substantial pump power to exceed the laser threshold and therefore to efficiently convert energy from the pump to the Stokes frequency. This obstacle is exacerbated by the fact that the frequency of the pump laser often does not match an atomic resonance of the Raman medium, as will be discussed. These two factors have traditionally relegated Raman conversion to the pulsed (as opposed to continuous-wave) laser regime where the required high peak powers can be achieved.

Given its importance in understanding the Raman process, a more detailed analysis of the Raman medium is now given. The nonlinear Raman medium, represented by ovals in Figs. 1 and 2, is often modeled quantum mechanically as a collection of three-level atoms or molecules, each in a lambda (Λ) configuration as depicted in Fig. 3. As the figure suggests, it is useful to juxtapose two different Raman transition regimes in order to place the focus of this thesis into perspective.

Figure 3. Simplified energy level diagrams for the (a) near-resonance and (b) far-off-resonance Raman processes. The large detuning (Δ) causes the gain for the far-off-resonance process to be much lower than for the near-resonance process.
Near-resonance SRS

Figure 3(a) shows the near-resonance case where both the pump and Stokes field frequencies nearly match the single-photon atomic transition frequencies (i.e. the single-photon detuning $\Delta$ is small). Gain for the Raman process is maximized in this near-resonance regime. Through the process shown, energy can be converted from the field at the pump frequency ($\omega_p$) to the Stokes frequency ($\omega_s$). Energy conservation requires that the atomic system be promoted to an excited state (level 2 in the figure) during the interaction. The quantum structure of the atom therefore determines the Raman frequency shift between the input pump and generated Stokes light.

By capitalizing on the enhanced gain in this near-resonance regime, cw Raman emission has been achieved experimentally in ammonia [2, 3], neon [4, 5, 6, 7], and atomic vapor [8, 9]. However, there are two significant drawbacks to operating in this regime. First, these systems depend critically on the pump laser’s spectral proximity to the single-photon transition frequency. As evident from the schematic in the center of Fig. 3, small changes in the pump laser’s frequency alter the gain significantly. This limitation substantially restricts the suitable pump sources as well as the spectral coverage and tunability of the Stokes output. Secondly, additional processes (other than Raman) based on the population of level 3 complicate the behavior of these systems both theoretically and experimentally when the single-photon detuning is small.
Far-off-resonance SRS

In sharp contrast to the near-resonance systems, Fig. 3(b) depicts the far-off-resonance Raman regime, where $\Delta$ is very large. This means that the field frequencies are not even remotely close to the single-photon atomic transition frequencies. Note, however, that the two-photon Raman transition is assumed resonant $[\omega_p - \omega_s = \omega_{21} = (E_2 - E_1)/\hbar]$, as it was for the near-resonance case. The relative energy level spacings shown, including the virtual level (represented by a dashed line in the figure), are to-scale for the molecular system analyzed in this thesis (diatomic hydrogen gas). Again, energy is transferred from the pump to the Stokes frequency, with the difference remaining in the medium. But this time, as will be shown in later chapters, the extreme single-photon detuning isolates the Raman process from population-based effects. Furthermore, as can be seen from the center schematic in Fig. 3, tuning the pump laser frequency (which therefore tunes the Stokes output frequency) results in very little change in the Raman gain when $\Delta$ is this large. For hydrogen, this means that all visible and near infrared pump lasers will experience very similar gain and allows for the possibility of broadly tunable Stokes output. The primary drawback of this regime is the abysmal gain realized for such large single-photon detuning. In fact, esteemed Raman researcher and Nobel laureate Nicolaas Bloembergen stated in 1967: "The light intensity required to obtain an appreciable gain is so high that pulsed solid-state lasers are essential [10]."
Since that time, two techniques have led to the realization of cw Raman amplification and laser oscillation in the far-off-resonance regime. Both substantially increase the effective interaction length/time for Raman conversion in order to compensate for the reduced gain far from resonance. The first of these techniques involves the propagation of both the pump and the Stokes fields in Raman-active single-mode optical fiber [11, Chapter 8]. The glass core of the fiber itself serves as the Raman medium [12]. Due to the amorphous nature of glass, Raman gain occurs for vibrational transitions over a very broad spectral region with a peak at 13.2THz (440cm⁻¹). The process will likely play a significant role in telecommunications where multiple Raman shifts in optical fiber are used to convert existing cw lasers to wavelengths needed for telecommunications amplifiers [13]. However, in spite of the increased interaction length within the fiber, multi-watt pump lasers are typically required for these devices and cascaded Stokes shifts are often necessary to reach the target wavelength because a single shift is too small [14, 15, 16]. Furthermore, other nonlinear processes can compete with the Raman conversion [11].

The second technique currently used for far-off-resonance cw Raman conversion is that of high-finesse cavity enhancement. Instead of propagating long distances in a fiber, this method capitalizes on the prolonged photon storage time and the corresponding optical power enhancement within a high-finesse Fabry-Perot cavity [17, Chapter 11] to dramatically improve the effective interaction length for cw Raman conversion. This technique provides the foundation for the work presented in this
thesis and I'll therefore now narrow the focus of this introductory discussion accordingly.

Cavity-Enhanced CW Raman Lasers

Due to relatively recent improvements in mirror coating technology, Fabry-Perot optical cavities, or resonators, can now exhibit optical power enhancement factors exceeding $10^5$ [18]. Furthermore, the high-finesse cavity (HFC) mirrors can be coated for two wavelengths, rather than just one. As a result, by enhancing both the pump and the Stokes fields within a HFC containing a gaseous Raman medium, the third-order nonlinear Raman transition can be enhanced by factors exceeding $10^9$. This can reduce the typical far-off-resonance Raman laser threshold from hundreds of kilowatts to hundreds of microwatts. Figure 4 illustrates the benefit of double cavity enhancement by comparing the Raman laser threshold for three different cavity styles: a multi-pass cell, a HFC that is resonant just for the Stokes, and a HFC that is resonant for both the pump and the Stokes. As a result of the HFC enhancement at both wavelengths, the stimulated Raman laser threshold can be reached using low-power ($<100\text{mW}$) cw pump sources.
Figure 4. Diagram depicting the Stokes laser threshold for three different enhancement techniques. The diagram emphasizes the usefulness of high-finesse cavity (HFC) enhancement of both the pump and the Stokes fields. Specifically, the far-off-resonance Raman transition can be enhanced by nearly nine orders of magnitude.

However, this substantial cavity enhancement is not without cost. Because the cavity finesse is so high, its linewidth (or spectral bandpass) is necessarily very narrow. This means that if one wishes to build optical power within the HFC for Raman conversion, the pump laser’s frequency/phase must be stabilized to that of a cavity resonance. Fortunately, precision laser stabilization techniques have received considerable attention in recent years in such fields as coherent communications [19, and references therein], atomic physics [20, and references therein], high-resolution spectroscopy [21, and references therein] and precision metrology [22, and references therein], in addition to other forms of nonlinear optical frequency conversion. In fact, HFCs (with no gas between the mirrors) are themselves often used as phase/frequency references to which lasers are stabilized [23, 24, 25].
It is therefore not surprising that the basic physical design for a cw Raman system is similar in concept to those of laser stabilization schemes as well as other cavity-enhanced nonlinear processes. As shown in Fig. 5, at its most basic level, the Raman laser setup can perhaps be viewed as containing three essential elements: The pump laser, the stabilization system, and the HFC. The pump laser provides the input energy for the process, the HFC containing hydrogen gas provides the conditions necessary for nonlinear conversion of the pump light into Stokes light, and the stabilization system ensures that the pump laser remains resonant with the HFC.

Using these concepts, far-detuned cw Raman lasing was first achieved in 1998 using a stable, frequency-doubled Nd:YAG laser as the pump source at 532nm [26, 27]. The system generated several milliwatts of Stokes power at 683nm with a peak photon conversion efficiency of 27%. The output was characterized with respect to its time dependent behavior [27], continuous tuning [28], and linewidth [27]. This system also provided the first evidence of thermally-induced intensity dependent refractive
index [29] and coherent anti-Stokes generation [30]. But perhaps most importantly, it provided experimental confirmation of a Raman laser threshold below 1mW with measured cavity finesses near 13,000 for each of the two wavelengths. This pioneering work therefore confirmed the possibility of using diode lasers as pump sources.

Diode Pumping and Applications

Diode lasers are ideal pump sources for cavity-enhanced cw Raman lasers from an applications standpoint owing to their relative low cost, small physical size, broad spectral coverage, and tunability [20, 31]. It can be seen from Fig. 3 that if the input pump frequency is tunable, then the output Stokes frequency can tune as well (i.e. the virtual level will shift). In fact, Fig. 6 shows that by Raman shifting existing room temperature diode lasers in just two gases, gap-free spectral coverage from the visible past 5μm can be obtained. This diode laser frequency conversion technique becomes even more attractive when one considers the modest pump power requirements [32], high output power possibilities [33], high conversion efficiencies [34], as well as the exceptional spatial and spectral purity of the output [27]. The primary drawback of using diode lasers as pump sources is the limited single-mode output power currently available from commercial devices.
Applications for relatively inexpensive, high quality, moderate power, near-infrared laser sources are widespread, yet there is currently a dearth of such sources in much of the spectral region. As a research tool, the GHz-range continuous frequency tuning and kHz-range linewidth of cw Raman lasers lend themselves well to the field of high resolution spectroscopy. In related fields, the possibility exists to create compact diode-pumped systems for remote sensing and on-site pollutant detection. Furthermore, frequency doubling the output of a Raman laser near 1178nm could generate a source for conducting atomic physics experiments and for adaptive optics using the sodium vapor transition near 589nm [35]. This would provide a relatively inexpensive alternative to the cumbersome cw dye laser which is predominantly used at this time.
In the field of precision metrology, cw Raman lasers may also provide a new method of frequency comb generation for laser stabilization [22]. Additional applications will undoubtedly also be found in other scientific fields and industries.

Clearly, the possibility exists for widespread use of cw Raman lasers. However, their niche will develop amongst higher power cw optical parametric oscillators, simpler diode pumped fiber lasers, less expensive communications wavelength diode lasers, as well as the cryogenically-cooled quantum cascade and lead salt lasers, just to name a few. The precise role that cw Raman lasers will play in the near-infrared arena remains to be seen.

Ring-Cavity Geometry

Most of the applications for cw Raman lasers rely on their long and short-term amplitude and frequency stabilities. Several factors, including stability, favor a ring-cavity geometry [17, pages 532-538] over a linear (two-mirror) geometry for cavity-enhanced cw Raman conversion. Examples of these two cavity geometries are shown schematically in Fig. 7. Furthermore, the traditional challenge of obtaining unidirectional output from a ring laser is elegantly averted in the Raman system, as discussed below.
Perhaps the most important advantage of the ring cavity over its linear counterpart for diode-pumped Raman lasers is the elimination of optical feedback from the input mirror. For the linear cavity, the reflected-plus-transmitted pump field from the input mirror returns directly along the input path towards the pump laser, as shown in Fig. 7. Such optical feedback can seriously debilitate the stability of the pump source, particularly for diode lasers, [20, and references therein] and can cause severe locking instabilities. Substantial optical isolation of the pump laser is therefore a necessity with linear cavity setups. But even with 60-70 dB of isolation, the effects of optical feedback can still be observed. On the other hand, the reflected-plus-transmitted field does not return to the pump laser when a ring cavity is employed. This allows one to minimize or even omit the often-expensive optical isolators.

A second advantage of the ring laser geometry is the increased interaction volume within the Raman active medium. This dilutes the deleterious effects of heat
generation [29, 36] and also offers additional gain with each pass. In contrast to most applications, it is not necessary to focus tightly since the gaseous gain medium fills the entire intracavity region. Additionally, even though it has not been an issue for the linear cavity Raman lasers, the traveling wave nature of the circulating field in the ring cavity also prohibits standing wave effects such as spatial hole burning [17, pages 316–323]. This is an enormous advantage for other ring-cavity laser systems.

Perhaps the most intriguing aspect of the ring-cavity Raman laser is its inherent unidirectional operation. In typical ring lasers, symmetry in the gain between the forward and backward directions can cause non-stable, bi-stable, or dual-mode operation. [17, pages 534–535] Intracavity optical elements are therefore often required to coerce the laser to oscillate stably in a single direction. However, due to preferential Doppler broadening of the Raman transition in the backward direction, the ring-cavity cw Raman laser exhibits inherent stability in the forward direction when tuned to the Raman transition line-center.

The drawbacks of using a ring cavity for Raman conversion include the added complexity in design, the cost of additional mirrors, and a slightly lower cavity finesse. As will be shown in Chapter 2, any transmission through the extra mirrors enters as a loss in the Raman equations, and will therefore degrade the conversion efficiency from pump to Stokes power. However, with high-quality and properly chosen mirrors, this drawback is insignificant.
Overview of this Thesis

This thesis describes the classical, quantum, and thermo-optic fundamentals of the diode-pumped cw Raman laser from both theoretical and experimental standpoints. It is intended as an educational tool rather than a reproduction and collection of publications. I attempt throughout to either describe each conceptual jump in detail through the extensive use of appendices, or to refer the reader explicitly to adequate resources. In light of this, the reader who finds the material overly explanatory can alternatively find the essential results and information in the pertinent journal publications.

The remainder of the thesis can be partitioned into two segments. The theoretical segment includes classical, quantum, and thermo-optic treatments of the cw Raman laser system. The experimental segment describes the construction, operation, and measured results of diode-pumped cw Raman lasers.

The classical treatment in Chapter 2 is used to derive analytical equations to describe the basic cw Raman processes. I make a concerted effort to retain as much simplicity and clarity as possible in this treatment. The chapter focuses on the ring-geometry Raman laser cavity, but the methods can be applied equally well to the linear cavity. In fact, a comparison is made between the two cavity styles as part of Appendix B. Also included in the classical analysis is a mathematically and practically oriented discussion of the optimal construction and operation parameters.
for the Raman system.

The quantum optical treatment provided in Chapter 3 is used to derive the time-dependent operator equations for the cw Raman laser in the Heisenberg picture starting from the system Hamiltonian. The Heisenberg picture facilitates physical understanding of the system. As a result, interesting connections are drawn between the cw Raman laser and the nondegenerate optical parametric oscillator as well as the traditional (population-based) laser. The quantum optical treatment also predicts the possibility of nonclassical behavior from the cw Raman system and generates analytical expressions for the output intensity noise (which include quantum noise).

Important thermo-optic effects are addressed theoretically in Chapter 4. The treatment deals with both the thermally-induced mode pulling and the thermal lensing that occur within the Raman laser cavity as functions of the generated Stokes power. The resulting modifications to the basic cw Raman laser equations are derived and discussed. The thermo-optic effects present the largest fundamental obstacle to be overcome in creating stable, high-power cw Raman systems.

Chapter 5 focuses on the frequency stabilization of the diode pump source to the HFC for Raman conversion. This is perhaps the most important and challenging experimental task involved in the construction of these systems. Both passive and active stabilization techniques are discussed, but a detailed description of only the active electronic locking procedure is given. Specific descriptions of the external-cavity pump laser, the high-finesse Raman laser cavity, and the high-bandwidth locking servo
are given. The chapter also describes the implementation of the injection locking technique to enhance the often-meager power of the external-cavity diode laser pump source.

Some experimental results that have been measured when the pump laser is stabilized to the high-finesse Raman laser cavity are provided in Chapter 6. Specifically, the chapter documents the first diode-pumped cw Raman laser, the first high-efficiency Raman laser, the implementation of injection locking in the Raman laser system, the thermal mode pulling and lensing, and the unidirectional operation of the first cw Raman ring laser. All of the experimental results are compared to the theoretical models developed in Chapters 2-4. The final chapter is used to review the primary achievements and findings presented in this thesis and to discuss possible future research directions.
The goal of this chapter is to provide the most straightforward time-dependent theoretical description of the far-off-resonance cw Raman laser possible. I therefore strongly favor simplicity at the expense of rigor. Although many subtleties are lost in the process and are left to be discovered in later chapters, it is my hope that the results presented here allow insight into the basic physical processes that are involved in cw Raman lasers. When appropriate, I try to mention the shortcomings or limitations of the results. I also use four appendices to cover some important topics in more depth. In particular, Appendix B is used to derive the Raman laser equations from first principles using the Lamb approach. Consequently, some may find this appendix more useful than the cursory treatment given in the main chapter. I find myself referring back to this appendix perhaps more than any other part of the thesis. Also of particular interest is Appendix D, which documents the development of a useful physical tuning picture for the cw Raman system.

The treatment in this chapter is purely classical. Here, the quantum structure of the atom is conveniently avoided by connecting the Raman gain to an empirically-based parameter (the plane-wave gain coefficient). Indeed, this is how the Raman gain is typically obtained in practice when theory is compared to experiment. Also
note that an adequate physical description of the Raman gain can be established using strictly classical considerations [37]. The chapter following this provides the opposite extreme where both the fields and the atoms are treated quantum mechanically.

This chapter begins with a review of previous theoretical studies of cw Raman systems. In the second section, rate equations for both the pump and the Stokes intracavity optical powers are derived. Along the way, the gain constant for the Raman medium in the focused geometry is obtained simply by connecting to the results of existing pulsed Raman laser work. In the third section the steady-state limits of the intracavity Raman power equations are taken in order to generate the laser threshold condition. Equations describing the output versus input powers in the steady state also follow directly from the intracavity powers. The fourth section combines mathematical with practical considerations in determining the optimum construction and operation parameters for cw Raman systems. The last section consolidates the findings of the chapter and connects with the material to follow.

The appendices that are associated with this chapter are as follows: Appendix A provides a simple method of obtaining a very useful general laser cavity field equation. The equation can be applied to both the pump and the Stokes fields for the system at hand. Appendix B provides a rigorous derivation of the Raman laser equations in the focused geometry starting from the Maxwell equations. This appendix can be viewed as an alternative to the treatment given in the main chapter. Appendix C shows that the fundamental eigenmode of a single pass through the bow-tie ring cavity
can be approximated by that of a linear cavity when the reflection angles are small. Appendix D provides a very useful physical picture of the frequency tuning behavior for the cw Raman laser. Throughout the chapter and the appendices, I attempt to follow Siegman's notation in *Lasers* as closely as possible [17].

**Previous Work**

The origins of cw Raman laser theory can be traced to the mid-1960's where amplifier theory (based in the spatial domain rather than the time domain) was the dominant approach [38, 39]. Most of this early work was devoted to predicting and optimizing the laser threshold in the plane-wave limit, but subsequent treatments incorporated beam focusing. In particular, Boyd and coworkers developed very useful analytic relations between the often-measured plane-wave gain coefficient and the Raman gain in the focused geometry [40].

The experimental realization of far-off-resonance cw Raman lasing in 1998 was followed by several theoretical treatments. Repasky and coworkers developed a steady-state interferometric model, predicting a peak photon conversion efficiency of 50% for symmetric mirrors (meaning the front and back mirrors have identical reflectivities) and 100% for asymmetric mirrors (meaning the front and back mirrors have different reflectivities) [41, 42]. The researchers discovered that the peak conversion efficiency occurs at a pump rate of four times the threshold value. Peterson and coworkers used an amplifier theory approach in another steady-state treatment [43]. They showed
that under certain circumstances, a reduction in the mirror reflectivities could lead to
greater power extraction, thereby reinforcing the findings of Repasky et al. Brasseur
and coworkers developed the first time-dependent cw Raman laser theory [44] based
on a general laser derivation that was introduced by Lamb long ago [45]. In the
appropriate limits, all the treatments mentioned above can be shown to generate
equivalent results in the steady state (aside from a factor of two error in the Repasky
treatments). Quantum optical treatments that are related to cw Raman lasers are
discussed in the next chapter.

Raman Power Rate Equations

Starting from the general laser cavity field equation derived in Appendix A, this
section derives time-dependent equations for both the pump and Stokes optical pow­
ers. The Raman gain is obtained by connecting to previous pulsed Raman work.

The treatment given here is based upon the bow-tie ring cavity depicted in Fig. 8,
but it can be applied equally well to the linear cavity geometry as discussed at the end
of Appendix B. In the figure, the $E'$s represent one-way traveling wave electric field
amplitudes, while the $r$'s represent amplitude reflectivities. The cavity is assumed to
exhibit a high finesse so that all the reflectivities are very nearly unity. The mirror
with reflectivity $r_0$ serves as both the input (for the pump) and output (for the Stokes)
coupler. The other three mirrors exhibit identical reflectivities ($r_1$). It will be shown
that $r_1$ should be chosen as high as possible. All the mirrors have equal curvature so
that a focus occurs midway between each pair and there exists a symmetry axis about the center of the cavity in both the horizontal and vertical dimensions as can be seen in Fig. 8. The gain medium occupies the entire intracavity volume. The beam path within the cavity is composed of four passes; two short passes of length $L$ (horizontal in the figure), and two long passes of length $L' = L/c\cos(2\theta)$ (diagonal in the figure). In this work, the angle $\theta$ is small so that $L \approx L'$.

Figure 8. Illustration showing the ring-cavity geometry with the relevant fields. The laser medium occupies the entire intracavity volume.

From Eq. (A.5) in Appendix A, the amplitudes of both the pump and the Stokes fields within the laser cavity obey the equation

$$\frac{dE(t)}{dt} = -\frac{\gamma}{2}E(t) + c\alpha_mE(t) + \frac{1}{\tau_{rt}}\sqrt{T_0}E_{inc}(t),$$

(2.1)

where $E(t)$ represents the real one-way circulating field amplitude (the subscript \textquotedblleft circ\textquotedblright{} has been dropped from the notation used in Appendix A), $E_{inc}(t)$ is the incident
field amplitude (also real), $c$ is the speed of light, $\tau_{rt} = p/c$ is the round-trip time in the cavity, $p \approx 4L$ is the round-trip cavity length, $\alpha_m$ is Siegman's gain (or loss) coefficient that is due to interaction with the atomic medium [17, page 272], $T_0$ is the power transmissivity of the input coupler$^1$, and

$$\gamma \equiv \tau_{rt}^{-1} (1 - R_{rt})$$

(2.2)

is the cold-cavity power decay rate, where $R_{rt} = R_0 R_1^3$ is the round-trip power reflectivity for the ring cavity ($R_{rt} \equiv r_{rt}^2$). Eq. (2.1) simply indicates that three factors can affect the circulating field dynamics. Specifically, the first term on the right represents decay due to transmission and absorption of the mirrors (loss), the second term represents the effect of the intracavity medium (loss or gain), and last term is due to an external input field (gain).

Because the optical power is proportional to the field magnitude squared ($P = \text{const.} \times E^2$ for real $E$) differentiation of this power leads to the equation

$$\frac{dP(t)}{dt} = \text{const.} \times 2E(t)\frac{dE(t)}{dt}.$$ (2.3)

Using this expression, Eq. (2.1) can be converted to a power rate equation to yield

$$\frac{dP(t)}{dt} = -\gamma P(t) + 2c \alpha_m P(t) + 2\tau_{rt}^{-1} \sqrt{T_0} \sqrt{P(t)} \sqrt{P_{inc}(t)}.$$ (2.4)

This expression is also quite general and applies to a wide variety of laser systems. All the parameters in the equation can be directly measured with the possible exception

$^1$ The variable $T_0$ used here for the input coupler transmissivity is also used for temperature in Chapter 4.
of the laser gain coefficient, $\alpha_m$. Attention will now be focused on this parameter for the specific Raman gain medium at hand.

**Raman Gain Coefficient**

This subsection is devoted to relating Siegman’s laser gain coefficient, $\alpha_m$ (units 1/m), in Eq. (2.4) to the commonly-measured Raman plane-wave gain coefficient, denoted as $\alpha$ (units m/W). This is accomplished somewhat superficially by simply connecting to previous Raman research in the pulsed laser regime. A more thorough treatment of the laser gain beginning with the Maxwell equations is given in Appendix B, but the results are identical. That appendix also compares the results to that of a standing-wave cavity.

Boyd and coworkers contributed substantially to the field of nonlinear optics through (among other things) their analyses of parametric and stimulated interactions in the focused geometry [40, 46]. Of primary interest here is their expression for the fractional increment in Stokes power per pass given by Eq. (3.8) of Ref. [40]. For the case when the confocal parameters for the two wavelengths are the same (this is true when a single cavity is used to resonate both wavelengths), Rabinowitz and coworkers simplified Boyd’s expression to [47]

$$\frac{\Delta P_s}{P_s} = \frac{4\alpha P_p}{\lambda_p + \lambda_s} \tan^{-1}(L/b), \quad \text{(single pass, traveling wave)} \quad (2.5)$$

where the pump and Stokes are distinguished by the subscripts $p$ and $s$, respectively, the circulating pump power $P_p$ is assumed to be constant over a single pass, $\alpha$ is the
plane-wave gain coefficient (not Siegman’s gain coefficient), λ’s are wavelengths, b is the confocal parameter, and L is the length of one focused pass in the cavity (not the round-trip length).

There are several obstacles presented with applying Eq. (2.5) to the ring cavity. First off, due to the non-zero reflection angle from each mirror, the stable fundamental resonator mode exhibits elliptical beam profiles at the four waists. As a result, the confocal parameters for the two radial dimensions are not the same. Second, the lengths and confocal parameters for the short passes are different from those of the long passes. However, Appendix C shows that when the reflection angle is small, one can assume that each of the four passes in the bow-tie cavity is effectively identical to that of a linear cavity, but with a traveling- rather than a standing-wave axial dependence.

To connect Eq. (2.5) with the power rate equation derived above, one can consider just the medium gain term in Eq. (2.4) for the moment (the second term on the right). When the terms involving the mirrors are ignored, the resulting differential equation is separable and can be written

\[
\frac{dP_s(t)}{P_s(t)} = 2c \alpha_m, s dt.
\]  

(2.6)

This equation can be directly integrated over the round-trip time \((\tau_t = p/c)\). The integration is particularly easy when one notes that \(P_s(t)\) can be assumed nearly
constant over the round-trip time. The result can then be written

\[
\frac{\Delta P_s}{P_s} = 2p \alpha_{m,s}, \quad \text{(round trip)} \tag{2.7}
\]

where \(\Delta P_s = P_s(\tau_{rt}) - P_s(0)\). Note that this last step is incompatible with systems for which the single-pass gain is significant. The total fractional change in Stokes power given by Eq. (2.7) is just the sum of the contributions from each of the four single passes from Eq. (2.5). That is, when the angle \(\theta\) is small (see Fig. 8), the four passes yield a total fractional Stokes power change of

\[
\frac{\Delta P_s}{P_s} = 4 \times \frac{4\alpha P_p}{\lambda_p + \lambda_s} \tan^{-1}(L/b). \quad \text{(round trip, traveling wave)} \tag{2.8}
\]

Equating Eqs. (2.7) and (2.8), and solving for Siegman’s gain coefficient for the Stokes gives

\[
\alpha_{m,s} = \frac{P_p}{2pP_1}, \tag{2.9}
\]

where the constant

\[
P_1 = \frac{\lambda_p + \lambda_s}{16 \alpha \tan^{-1}(L/b)} \quad \text{(bow-tie cavity unity gain constant)} \tag{2.10}
\]

has units of power and can be interpreted as the circulating pump power required to achieve unity gain over a distance of one round trip. As indicated in Appendix C, \(P_1\) can be modified to accommodate larger reflection angles in the bow-tie cavity.

To obtain an expression analogous to Eq. (2.9) for the pump gain coefficient, one can notice that energy conservation requires the power gained by the Stokes per pass
to be related to the power lost by the pump through

$$\Delta P_p = -\frac{\lambda_s}{\lambda_p} \Delta P_s. \quad (2.11)$$

With this, and using expression (2.7) as well as an identical equation for $\Delta P_p/P_p$, one can quickly identify the pump gain coefficient as

$$\alpha_{m,p} = -\frac{\lambda_s}{\lambda_p} \frac{P_s}{2P_1}. \quad (2.12)$$

With the gain constants given by Eqs. (2.9) and (2.12), Eq. (2.4) can now be used to generate the time-dependent equations for the pump and Stokes circulating powers

$$\frac{dP_p}{dt} = -\gamma_p P_p + \frac{\lambda_s}{\lambda_p} \tau_{rt}^{-1} \frac{P_s P_P}{P_1} + 2\tau_{rt}^{-1} \sqrt{T_{P,0}} \sqrt{P_p} \sqrt{P_{inc}} \quad (2.13)$$

$$\frac{dP_s}{dt} = -\gamma_s P_s + \tau_{rt}^{-1} \frac{P_s P_P}{P_1}. \quad (2.14)$$

The explicit time dependence was omitted here for simplicity and it was assumed that no incident Stokes beam exists (no "seed"). These two equations are the primary results of this section. They give the dynamic behavior of the circulating powers in the two active cavity modes. All coefficients are given in terms of measurable or tabulated quantities. To find the laser system's intensity noise, the equations can be solved numerically or they can be linearized about the steady-state values and solved analytically. Such a treatment will be postponed until the next chapter so that quantum noise can be included.

**Steady-State Powers**

The steady-state limit of Eqs. (2.13) and (2.14) can be taken by setting the time
derivatives to zero. In this limit, and using Eq. (2.2), the circulating pump and Stokes powers above threshold become

\[ P_p = P_1(1 - R_{s,rt}), \quad (2.15) \]
\[ P_s = \frac{\lambda_p}{\lambda_s} \left[ \left( \frac{4 T_{p,0} P_1 P_{\text{inc}}}{1 - R_{s,rt}} \right)^{1/2} - P_1(1 - R_{p,rt}) \right]. \quad (2.16) \]

It should be noted that a second set of solutions is also possible from Eqs. (2.13) and (2.14). The second set corresponds to zero Stokes power generation and represents stable solutions only below threshold.

Note that the circulating pump power given by Eq. (2.15) is clamped (independent of incident power) above threshold. The system therefore acts as an optical power limiter for the incident pump light. As discussed in Chapter 3, similar clamping behavior is also observed in both the nondegenerate optical parametric oscillator and the traditional (population-based) laser. Also, the intracavity Stokes power is predicted to grow as the square root of the incident pump power. However, it should be mentioned here that the intracavity pump power ceases to clamp and the Stokes power departs from a strictly square-root dependence on the incident pump power when significant Stokes power is generated (typically \( \gtrsim 10\text{mW} \) for the ring cavity). These effects and the associated modifications to Eqs. (2.15) and (2.16) are due to thermal lensing within the laser cavity and are discussed at length in Chapter ??.

**Threshold Power**

The incident pump power necessary to reach the Stokes laser oscillation threshold...
can be obtained by setting Eq. (2.16) to zero. This yields

\[ P_{\text{th}} = \frac{P_1 (1 - R_{p,rt})^2}{4 T_{p,0}} (1 - R_{s,rt}) \]  

(2.17)

\[ \approx \pi^2 \frac{P_1}{\mathcal{F}_p \mathcal{F}_s}, \]  

(2.18)

where the approximate equality holds when the input coupler transmissivity dominates the other cavity losses for the pump. The symbols \( \mathcal{F}_p \) and \( \mathcal{F}_s \) are the cold-cavity (i.e. no intracavity medium) finesses of the resonator at the pump and the Stokes wavelengths, respectively. They are of the form [17, page 436]

\[ \mathcal{F} \equiv \frac{\pi \sqrt{T_{rt}}}{1 - r_{rt}} \approx \frac{2\pi}{1 - R_{rt}}. \]  

(2.19)

Approximation (2.18) highlights the usefulness of enhancing both the pump and Stokes fields in the cavity since the threshold pump power scales inversely with the product of the two finesses.

**Emitted Powers**

The optical powers outside the cavity can be easily obtained by multiplying Eqs. (2.15) and (2.16) by the appropriate power transmission coefficients, with the exception of the pump power reflected from the input coupler. In this case, the total reflected field is a coherent sum of the transmitted portion of the circulating pump field and the (out of phase) reflected portion of the incident field

\[ E_{\text{refl}} = t_{p,0} E_p - r_{p,0} E_{\text{inc}}. \]  

(2.20)
Converting this equation to optical power yields

\[
P_{\text{refl}} = \left( \sqrt{T_{p,0}P_p} - \sqrt{R_{p,0}P_{\text{inc}}} \right)^2.
\]  \hspace{1cm} (2.21)

Using this result and Eqs. (2.15) and (2.16), the output Stokes power, the reflected pump power, and the transmitted pump power are found to be

\[
P_{\text{out}} = \frac{\lambda_p}{\lambda_s} T_{s,\text{tot}} \left[ \left( \frac{4 T_{p,0} P_1 P_{\text{inc}}}{1 - R_{s,rt}} \right)^{1/2} - P_1 (1 - R_{p,rt}) \right], \hspace{1cm} (2.22)
\]

\[
P_{\text{refl}} = \left\{ \left[ P_1 (1 - R_{s,rt}) T_{p,0} \right]^{1/2} - \sqrt{P_{\text{inc}}} \right\}^2, \hspace{1cm} (2.23)
\]

\[
P_{\text{trans}} = P_1 (1 - R_{s,rt}) T_{p,1}, \hspace{1cm} (2.24)
\]

where \( T_{s,\text{tot}} = T_{s,0} + 3T_{s,1} \) for the ring cavity. The overwhelming similarities between these equations and those of the non-degenerate parametric oscillator are discussed in Chapter 3. One can also derive the previous three expressions for the emitted powers from the work of Repasky (aside from a factor of 2 error) \[41\], Peterson \[43\], and Brasseur \[27\] when modified for the ring cavity and in the appropriate limits.

Interestingly, it is shown at the end of Appendix B that Eqs. (2.22) - (2.24) can also be directly applied to the two-mirror linear cavity with \( T_{s,\text{tot}} = T_{s,0} + T_{s,1} \) and \( R_{rt} = R_0 R_1 \).

Using Eq. (2.17) for the threshold pump power, a handy dimensionless pumping ratio can be defined as

\[
r_p \equiv \frac{P_{\text{inc}}}{P_{\text{th}}}.
\]  \hspace{1cm} (2.25)

So \( r_p \), called the pump rate, represents the incident pump power normalized to the
threshold power. In terms of pump rate, Eqs. (2.22) and (2.23) can be recast as

\[ P_{\text{out}} = \frac{\lambda_p}{\lambda_s} P_s T_{s,\text{tot}} (1 - R_{p,\text{rt}}) (\sqrt{r_p} - 1) \]  
(2.26)

\[ P_{\text{refl}} = P_{\text{th}} \left( \frac{2T_{p,0}}{1 - R_{p,\text{rt}}} - \sqrt{r_p} \right)^2. \]  
(2.27)

The transmitted pump and output Stokes power from Eqs. (2.24) and (2.26) are plotted in Fig. 9(a) as functions of pump rate. Note the clamping of the transmitted pump power and the square-root dependence of the Stokes power on pump rate above threshold. This dependence is strictly a result of the cavity. The emitted Stokes power is linear when plotted versus the pump power that actually enters the cavity (i.e. that which is not reflected off the input coupler).

Figure 9. (a) Emitted and (b) reflected powers from the cw Raman laser cavity versus pump rate. The transmitted pump power is clamped above threshold \( r_p = 1 \), while the emitted Stokes power exhibits a square-root dependence on the pump rate. By increasing the input coupler transmissivity \( T_{p,0} \) relative to the other intracavity losses at the pump wavelength \( (A_{p,\text{gen}}) \) impedance matching can occur above threshold, but not greater than four times threshold. The parameters used to generate these curves are: \( P_1 = 24\text{kW}, R_{p,0} = R_{s,0}^3 = 0.9998, T_{p,0} = 200\text{ppm}, \) and \( R_{p,1} = R_{s,1}^3 = 0.9998. \)
The first curve in Fig. 9(b) shows the reflected pump power from Eq. (2.27) for the same parameters. The other two curves in Fig. 9(b) will be discussed in the next subsection.

**Impedance-Matching Power**

Eq. (2.23) can be used to determine the pump power for which the active cavity is impedance matched; that is, the input pump power for which the reflected pump power vanishes [17, page 423]. Setting Eq. (2.23) to zero gives

\[ P_{\text{match}} = P_1 (1 - R_{s,rt}) T_{p,0} \]

\[ = 4P_{th} \left( \frac{T_{p,0}}{1 - R_{p,rt}} \right)^2 \]

\[ \approx \frac{4P_{th}}{(1 + A_{p,\text{gen}}/T_{p,0})^2}, \]

where

\[ R_{p,rt} = R_{p,0} R_{p,1}^3 \]

\[ = (1 - T_{p,0} - A_{p,0})(1 - T_{p,1} - A_{p,1})^3 \]

\[ \approx 1 - T_{p,0} - A_{p,0} - 3T_{p,1} - 3A_{p,1} \]

\[ = 1 - T_{p,0} - A_{p,\text{gen}}. \]

Here, the \( A \)'s are mirror power absorption coefficients and the generalized absorption is defined as

\[ A_{p,\text{gen}} \equiv A_{p,0} + 3T_{p,1} + 3A_{p,1}. \]
Approximation (2.29) shows that the pump power required to reach the impedance-matching condition approaches $4 \times P_{th}$ ($r_p = 4$) in the limit of small generalized absorptions. Fig. 9(b) shows the reflected power for three different choices of the ratio $A_{p, gen}/T_{p, 0}$. As the ratio decreases, impedance matching occurs for higher input pump rates, meaning more and more pump light can enter the cavity for Raman conversion. However, the impedance-matching pump rate can never exceed $r_p = 4$. As will soon be shown, this particular pump rate is also (not coincidentally) significant for the system's conversion efficiency.

The impedance-matching phenomenon can be explained from a physical standpoint. For an empty cavity, no optical power is reflected when the input coupler losses exactly equal the other mirror losses [17, page 423]. This is because the portion of the circulating intracavity field that is transmitted back through the input coupler exactly cancels the portion of the incident field that is reflected from this mirror (they are out of phase). All the light is transmitted through an impedance-matched empty cavity with no absorptions.

However, for the active laser cavity currently under investigation, the circulating pump power is clamped by the Raman process above threshold. This means that the transmitted portion of the circulating field no longer perfectly cancels the reflected portion of the incident field. As a result, some optical power is reflected off the input coupler above threshold and is therefore not available for Raman conversion.
This problem is not unique to the cw Raman laser and is faced in every cavity-enhanced nonlinear process. The solution is to increase the transmissivity of the input coupler relative to the other intracavity losses at the pump wavelength. This means that incomplete cancellation of the two fields occurs below threshold (the transmitted intracavity portion is initially larger than the reflected incident portion), but that perfect cancellation will occur for one specific incident pump power value above threshold, as shown in the lower two curves of Fig. 9(b).

Raman Laser Optimization

This section is devoted to finding the optimal operation and construction parameters for the cw Raman laser. Note that the results of this section will not hold when thermo-optic effects are significant. The derivation process simply consists of taking derivatives with respect to the parameter of interest, setting to zero, and solving for the parameter. A useful equation for this purpose will be that of the photon conversion, or extraction efficiency, which is related to the ratio of the output to input powers through

\[
\eta_{\text{ph}} = \frac{\lambda_s P_{\text{out}}}{\lambda_p P_{\text{inc}}} = \frac{T_{s,\text{tot}}}{P_{\text{inc}}} \left[ \left( \frac{4 T_{p,0} P_{\text{inc}}}{1 - R_s, rt} \right)^{1/2} - P_1 (1 - R_{p, rt}) \right]
\]

(2.32)

As a function of pump rate, this equation can be written

\[
\eta_{\text{ph}} = \left( \frac{T_{p,0}}{1 - R_{p, rt}} \right) \left( \frac{T_{s,\text{tot}}}{1 - R_{s, rt}} \right) \left( \frac{1}{\sqrt{r_p}} - \frac{1}{r_p} \right)
\]

(2.33)
With these equations, the photon extraction efficiency can now be optimized with respect to the pump rate.

**Optimum Pump Rate**

Differentiating Eq. (2.33) with respect to the pump rate, setting to zero, and solving for the optimum pump rate quickly yields

\[ r_{p, \text{opt}} = 4, \quad \text{or equivalently} \quad P_{\text{opt}} = 4 \times P_{\text{th}}. \quad (2.34) \]

This indicates that, regardless of mirror absorptions, transmissions, Raman gain, or cavity geometry, the optimal pump rate is always \( r_{p, \text{opt}} = 4 \). It is interesting that the impedance-matched condition approaches this optimal pump rate for high \( T_{p,0} \).

Inserting \( r_{p, \text{opt}} = 4 \) into Eq. (2.33) and only considering Stokes power emitted from the output coupler gives the optimum usable photon conversion efficiency

\[ \eta_{\text{ph}, \text{opt}} \equiv \frac{\lambda_s P_{\text{out}}}{\lambda_p 4P_{\text{th}}} \approx \left( 1 + \frac{A_{p,\text{gen}}}{T_{p,0}} \right)^{-1} \left( 1 + \frac{A_{s,\text{gen}}}{T_{s,0}} \right)^{-1}, \]  

(2.35)

which approaches unity in the limit of small generalized absorptions, where the generalized absorption terms \( A_{p,\text{gen}} \) and \( A_{s,\text{gen}} \) are of the form given in Eq. (2.31). This is an important result because it states that the only parameters that limit the conversion efficiency for cw Raman lasers are the ratios of the generalized absorptions to the input/output coupler transmissions at the two wavelengths. Eq. (2.35) as well as the results to follow can be modified to include all the output Stokes power (not just that emitted out the input coupler) by setting \( A_{s,\text{gen}} \rightarrow A_{s,\text{tot}} \) and \( T_{s,0} \rightarrow T_{s,\text{tot}} \).
The choice of mirror transmissions for optimum power extraction is now discussed. In this case, it is assumed that the maximum amount of incident pump power available is a fixed quantity ($P_{\text{max}}$) and the input and output coupling coefficients are determined in order to optimize the Stokes power extraction. Because $T_{p,1}$ and $T_{s,1}$ enter equivalent to absorptions in Eq. (2.35), these transmissivities should clearly be chosen as low as possible (this can be proven more rigorously). This allows $T_{p,0}$ and $T_{s,0}$ to be considered as the only optimization parameters. It is now possible to determine whether there is any advantage to be gained by choosing the mirror transmissivities higher for one wavelength than the other and what the values must be to optimize the Stokes power extraction. Specifically, by differentiating Eq. (2.32) once with respect to $T_{p,0}$ and again with respect to $T_{s,0}$, the resulting two equations can be set to zero and solved simultaneously for the transmissions to yield

$$T_{p,0,\text{opt}} = \sqrt{\frac{A_{p,\text{gen}}}{A_{s,\text{gen}}} \frac{P_{\text{max}}}{P_1}}$$  \hspace{1cm} (2.36)

$$T_{s,0,\text{opt}} = \sqrt{\frac{A_{s,\text{gen}}}{A_{p,\text{gen}}} \frac{P_{\text{max}}}{P_1}} - A_{s,\text{gen}}.$$  \hspace{1cm} (2.37)

Interestingly, these equations indicate that, to extract the most power from the system, the transmission coefficients for the input/output coupler will in general be different for the two wavelengths. One might be tempted to assume that these transmission coefficients should reproduce the condition $P_{\text{max}} = 4 \times P_{\text{th}}$ (i.e. satisfy the four-times-threshold condition). However, when Eqs. (2.36) and (2.37) are inserted
into Eq. (2.17) the result is

\[ P_{\text{th, opt}} = \frac{P_{\text{max}}}{4} \left[ 1 + \left( \frac{4 P_{1}}{P_{\text{max}}} A_{p,\text{gen}} A_{s,\text{gen}} \right)^{1/2} \right], \] (2.38)

which indicates that the optimum threshold is slightly larger than \( P_{\text{max}}/4 \). In other words, the optimum input/output coupling is such that the system will run slightly under the four-times-threshold condition when the maximum incident pump power is used. This seems to contradict the findings of the previous subsection. However, one must remember that the four-times-threshold condition optimizes the power extraction efficiency with respect to pump rate rather than mirror transmissions.

Figure 10 helps to further clarify this issue. The figure shows the photon conversion efficiencies for three different choices of transmission coefficients, as functions of the pump rate for the curve labeled A. In other words, the incident pump powers for all the curves are normalized to the threshold of curve A. The vertical dashed line represents the maximum available pump power. Pump powers greater than this \( (\tau_{p}^{(A)} > 4) \) are therefore inaccessible to the system. Curve A represents a choice for mirror transmissions such that \( P_{\text{max}} = 4 \times P_{\text{th}} \). Note that the conversion efficiency peaks at \( P_{\text{max}} \) as predicted by Eq. (2.34). However, by increasing the output coupling coefficient (and thereby raising the threshold) while holding all other parameters constant, one can increase the conversion efficiency from the previous situation even though this means that the conversion efficiency does not peak with respect to incident pump power. Curve B represents the optimal mirror transmissions calculated.
using Eqs. (2.36) and (2.37). Curve C shows that any further increase in the output transmission beyond the optimal values again results in a reduced conversion efficiency.

![Diagram showing photon conversion efficiency versus pump rate relative to curve A threshold](image)

Figure 10. Photon conversion efficiency versus the pump rate relative to the curve A threshold. The mirror transmissivities for curve A are chosen such that the threshold occurs at 1/4 of the maximum available pump power. The transmissivities for curve B are chosen according to Eqs. (2.36) and (2.37) and maximize the conversion efficiency for the available pump power.

In practice, more pressing concerns usually dominate the choice of mirror reflectivities. First off, it must be emphasized that the effect just described is only significant when the mirror absorptions are comparable or larger than the input/output transmission coefficient. Also, obtaining mirrors that are coated with the required accuracy for two wavelengths can be difficult and expensive. Nevertheless, Research
Electro-Optics (REO) in Boulder, Colorado can coat mirrors with transmission accuracy better than 95%. But perhaps more importantly, as discussed in the next two chapters, the input/output transmission coefficients should be chosen larger for the pump than for the Stokes for the purposes of stability, regardless of power extraction efficiency.

**Optimum Detuning**

A very useful physical picture of cw Raman laser frequency tuning is given in Appendix D. To this point in the chapter, it has been assumed that the pump and Stokes cavity modes differ in frequency by precisely the Raman shift. In other words, the HFC length is adjusted such that the Stokes cavity mode falls directly on the line center of the Raman transition (see Fig. 63 in Appendix D). Frequency tuning of the Raman laser away from this situation can yield some interesting behavior and was first addressed in Ref. [28]. For optimization purposes, detuning of the Stokes cavity mode from the Raman gain line center can be incorporated into all of the previous equations using the following simple transformation:

\[ P_1 \rightarrow \frac{P_1}{\mathcal{L}(\Delta_{12})}, \]

where

\[ \mathcal{L}(\Delta_{12}) = \frac{\gamma_{21}^2}{(2\Delta_{12})^2 + \gamma_{21}^2} \]

is the Lorentzian lineshape of the Raman transition in the high-pressure limit [48]. Here, \( \Delta_{12} \equiv \omega_p - \omega_s - \omega_{21} \) is the detuning from the transition line center, where
\( \omega_p, \omega_s, \) and \( \omega_{21} \) are the pump, Stokes, and vibrational frequencies, respectively. The constant \( \gamma_{21} \) represents the full width at the half maximum of the Raman gain. This treatment assumes that no mode hops (spatial, longitudinal or directional) occur during the tuning.

With Eq. (2.39) included, Eq. (2.32) can be differentiated with respect to the detuning \( \Delta_{12} \). The result can be set to zero and solved for the optimal detuning to yield

\[
\Delta_{12, \text{opt}} = 0 \quad (r_p \leq 4) \tag{2.41}
\]
\[
= \pm \frac{\gamma_{21}}{2} \sqrt{\frac{P_{\text{inc}}}{4P_{\text{th}}} - 1} \quad (r_p > 4) \tag{2.42}
\]

Figure 11(a) shows the output powers versus detuning for four different choices of pump rate (2, 4, 6, and 8). For a pump rate of \( r_p = 8 \), the Stokes power peaks at \( \pm \gamma_{21}/2 \) as predicted by Eq. (2.42). Figure 11(b) illustrates the bifurcation predicted by Eqs. (2.41) and (2.42) with increasing pump rate. Note also that for pump rates between \( r_p \approx 4 \) and \( r_p \approx 6 \), the Stokes power output is nearly constant with detuning out to about \( \pm \gamma_{21}/2 \). This feature can be used to determine the maximum constant power detuning as discussed in Appendix D.
Although the tuning behavior for $r_p > 4$ resembles a Lamb dip, the cause is physically unrelated. The Lamb dip occurs due to Doppler broadened gain media in standing-wave laser cavities, as discussed in Ref. [17, pages 1199-1212]. The dip in the Raman output power is due to the fact that the peak in the conversion efficiency occurs at $r_p = 4$. By tuning away from the Raman line center, the Raman gain drops, thus raising the effective threshold. Therefore, when $r_p > 4$ on the line center, detuning takes the system closer to $r_p = 4$, thereby increasing the conversion efficiency.

**Optimum Cavity Geometry**

Because the optimum power extraction efficiency is independent of $P_1$, it is therefore independent of the cavity geometry. This ceases to be true when the medium is
depleted or nonlinear refractive index effects (such as heating) are significant. Regardless, $P_1$ does play a role in determining the laser threshold. Using the trigonometric relation [49, page 467]

$$\csc^{-1}(x) = \tan^{-1}\left(\frac{1}{\sqrt{x^2 - 1}}\right),$$

Eq. (2.10) can be recast as

$$P_1 = \frac{\lambda_p + \lambda_s}{16\alpha \csc^{-1}\sqrt{2R/L}}$$

(2.43)

where $R$ is the mirror radius of curvature and the following relation for the confocal parameter of a cavity has been used [50, page 140]

$$b = L\sqrt{\frac{2R}{L} - 1}.$$  

(2.44)

To most effectively utilize the Raman gain, one can therefore maximize the inverse cosecant in Eq. (2.43). This function is plotted versus $R/L$ in Fig. 12. Two well-known cavity geometries (the concentric and the confocal) are indicated on the plot by solid circles and the open squares indicate $R/L$ ratios that have been used successfully.
The figure suggests that the concentric geometry should minimize the threshold, but practical concerns again dominate the choice of $R/L$. Specifically, as discussed by Boyd in Ref. [40], $R/L$ values near unity "place a premium on the quality of the mirrors" because the beam spot size on the mirrors is so large. Nonuniformities in the mirror surfaces over large areas can degrade the cavity finesse. I have not investigated such deleterious effects, but other researchers have used a 1mm spot size and retained a cavity finesse of 150,000 using mirrors from REO [51]. Boyd also points out that "in the degenerate case the spatial nonuniformity of Stokes gain results in a parametric
coupling of the simultaneously resonant transverse modes." Thus one may want to avoid rational fractions for $R/L$ such as 1 (confocal), 2/3, 3/2, etc.

Chapter Summary

This chapter has been devoted to developing the cw Raman laser theory in a manner that hopefully facilitates basic understanding of the system. Equations (2.13) and (2.14) gave the time dependent, coupled behavior of the circulating pump and Stokes powers. The conditions for the laser threshold and impedance matching were given in Eqs. (2.17) and (2.28). Perhaps most useful were Eqs. (2.22) - (2.24), which gave the steady-state output powers. It was also shown that the optimal pump rate is four times the threshold value and that the peak photon conversion efficiency can approach 100% in the limit of small generalized absorptions (which include all transmissions other than the input/output coupler). Optimization was also performed with respect to the input/output coupling, detuning from the Raman line-center, and cavity geometry. In the next chapter, the emphasis shifts from simplicity toward rigor as the quantum mechanical aspects of the cw Raman laser are addressed.
CHAPTER 3

QUANTUM THEORY

The primary goal of this chapter is to provide detailed insight into the quantum mechanical processes involved with cw Raman lasing. The work presented here is the result of the better part of a wonderful year spent in Australia interacting with Tim Ralph and Andrew White, both of whom were at the University of Queensland.

Rather than simplifying matters by assuming a real, non-transient, macroscopic polarization that only includes the Raman process as I did in Appendix B, I now start with a more fundamental microscopic picture of the underlying atomic structure and allow the physical system itself to determine any simplifications. And instead of treating the optical modes as fields that can assume any value of energy, I start with quantized cavity modes so that their energies are discretized and exchanges of energy must involve the creation or annihilation of a photon. By the end, this treatment allows for determination of all the populations, both the fields, and the intensity noise behavior of the emitted light with quantum noise included. The system is eventually shown to be capable of exhibiting intensity noise below the standard quantum limit (amplitude squeezing) for certain operating conditions. Along the way, an interesting and useful connection is uncovered between the far-off-resonance cw Raman laser and
the nondegenerate optical parametric oscillator (NDOPO). Additionally, the analysis given here lends itself naturally to investigation of parametric anti-Stokes and second-Stokes generation. A streamlined treatment of anti-Stokes generation in the semiclassical limit is provided in Appendix G.

This chapter begins with a review of past quantum mechanical treatments of related systems to provide context. The second section is used to assemble the appropriate components of the Hamiltonian for the far-off-resonance cw Raman laser system at hand. Due to the extraordinarily large detunings present in this system, the rotating wave approximation (RWA) is not valid, and is therefore not invoked. The quantum Langevin equations for the system operators are generated in the third section following the work of Gardiner and Collett [52]. In the fourth section, the large single-photon detuning is exploited to simplify the equations of motion significantly. In the fifth section, the semiclassical limit of the quantum Langevin equations is taken and the results are compared to the previous chapter. Returning to the quantum domain in the sixth section, direct connections are drawn between the cw Raman system and the NDOPO. In the seventh section, the simplified quantum Langevin equations for the two fields are linearized and the noise spectra of the emitted Stokes light is determined analytically. The eighth section is used to analyze and explore the parameter space of these equations. The final section consolidates the work of this chapter. Throughout this chapter, I attempt to follow the notation of Gardiner and Zoller [53].

The appendices associated with this chapter are provided primarily to supplement
large mathematical gaps in the main chapter. However, Appendix G documents the development of the semiclassical anti-Stokes theory when cavity enhancement of the anti-Stokes field is included. It is shown that the photon conversion efficiency can approach the quantum limit of 50% under ideal experimental conditions.

Previous Work

The literature addressing the quantum mechanical nature of pulsed Raman systems is prodigious, [54, and references therein] but little attention was devoted to the cw regime until the late 1980's and the 1990's, when three-level atoms interacting with quantized field modes were analyzed in the process of identifying and characterizing nonclassical sources of light. For such atoms in the A-configuration, many competing processes (including Raman) often contribute to the overall dynamics of the system. The dominance of certain processes over others is determined by the relative strengths of the rates involved (decay rates, detunings, Rabi frequencies, etc.) and the pumping scheme (coherent versus incoherent).

In particular, three level A-systems can exhibit optical bistability [55, and references therein] traditional population-based lasing (see, for example, Ref. [56], lasing without inversion [57, 58, 59], electromagnetically induced transparency [60, and references therein] in addition to Raman lasing [61, 62, 63, 64]. General treatments of three level systems have also been performed, which can accommodate many of these processes [65, 66, 67, 68]. All of the specifically Raman laser studies mentioned
above address the case where no decay path to the ground state is included to close the pump cycle. In these systems, the middle level population returns to the ground state via coherent interactions with the cavity fields (anti-Stokes process). On the other hand, Ritsch, et al. [65, 66] as well as others [67, 68] accommodate decay to the ground state as part of the pump cycle. These works provide useful general treatments of three level systems interacting with two cavity modes in the Schrödinger picture. Presumably none of them dealt with detunings large enough to become incompatible with the rotating wave approximation (RWA) since it was invoked in all.

The present treatment is directly based on the far-off-resonance cw Raman laser systems that have been experimentally realized [69, and references therein]. Specifically, this treatment assumes a collection of diatomic hydrogen molecules (approximated by three-level systems in the $\Lambda$-configuration) interacting with two strongly cavity-enhanced field operators. The field operators are very far detuned in frequency from the single-photon transitions, so the RWA is not invoked. Decay of all the level populations and coherences is allowed. In this treatment, the single-photon detunings are by far the largest rates in the system. The energy level diagram and specific descriptions of the interactions are included in the next section.

Another closely related work is that of Poizat, et al. [70] who examine two field modes interacting with a system of three-level atoms in a ladder or cascade configuration. It is pointed out by Olsen et al. [71] that this system can behave very similar to the corresponding $\Lambda$-configuration under similar conditions.
In the interest of retaining as much clarity as possible, attention will be focused on the temporal aspects of the system in this work and only a nonrigorous treatment of the spatial dependence will be provided. Also for simplicity, the cavity is assumed to be lossless and single-ended (i.e. only the input/output coupler has a reflectivity less than unity), but other mirror transmissions and losses can easily be included using the same formalism [72, and references therein]. It should be noted that thermal population of the pertinent excited states is negligible for the system at hand because they differ in energy from the ground state by more than $20 \times k_B T$. The generation of anti-Stokes radiation is neglected in the main text because it is very small in practice [30] and does not appreciably affect the dynamics of the other fields when it is not cavity enhanced. The effect of cavity enhancing the anti-Stokes field is treated in Appendix G. The complicating effects of heat generation are diverted until the next chapter [29, 36].

The atom-photon interactions that are included in this treatment, and the corresponding energy level diagram, are somewhat unconventional due to the extreme single-photon detuning that exists. As shown in Fig. 13, the hydrogen molecule is approximated by a three-level system in the $\Lambda$-configuration where level 1 is the ground state, level 2 is the first excited vibrational state, and level 3 is the first excited electronic state. The figure shows these three levels and all the pertinent fields to-scale.
in order to emphasize the large single-photon detunings. As a result of the large detunings, note that both the pump and Stokes fields are permitted to drive each of the two allowed single-photon transitions (1–3 and 2–3). Single-photon 1–2 transitions are forbidden by selection rules. Also note that the counter-rotating portions of each field are explicitly included and are represented by arrows below the energy levels. Detunings are represented by $\Delta$'s, the population decay rates by $\gamma$'s and the collisional dephasing rates by $\tilde{\gamma}$'s (not shown in the figure). Due to the large single-photon detunings, the fields will interact with more upper levels than just the one shown, but it is assumed that these will all contribute in a similar manner.

Figure 13. To-scale energy level diagram for the diatomic hydrogen molecule showing the pertinent levels and the far-off-resonance fields. The arrows below the energy levels represent counter-rotating terms that must be included due to the large single-photon detunings ($\Delta$'s).
The total Hamiltonian describing the atoms, the fields, the baths (to incorporate decay and noise), and their mutual interactions is

\[ H = H_1 + H_2 + H_3 + H_4 + H_5 + H_{\text{baths}}, \tag{3.1} \]

where the components of \( H \) are given as follows.

**Free Energy**

\( H_1 \) represents the free energy of the atoms and fields in the absence of any interactions and is given by

\[ H_1 = \sum_{i=1}^{3} \hbar \omega_i S_{ii} + \sum_{q=p,s} \hbar \omega_q^c a_q^\dagger a_q, \tag{3.2} \]

where \( \hbar \omega_i \) and \( S_{ii} \) are the energy and collective population operator for the \( i \)-th atomic level, respectively. Throughout this work, the subscript \( p \) refers to the pump while \( s \) refers to the Stokes. In this way, the operators \( a_q \) and \( a_q^\dagger \) refer to the annihilation and creation of pump \( (q = p) \) and Stokes \( (q = s) \) photons, respectively, while \( \omega_p^c \) and \( \omega_s^c \) are the frequencies of the cold cavity modes nearest to the pump and Stokes laser frequencies, respectively.

**Reversible Interaction Energy**

\( H_2 \) represents the reversible interaction energy associated with the atom-field couplings in the electric dipole approximation and is given by

\[ H_2 = i\hbar \sum_{q=p,s} (a_q^\dagger - a_q) \left( g_{q,13} S_{13} + g_{q,23} S_{23} + \text{H.c.} \right), \tag{3.3} \]
where H.c. denotes the Hermitian conjugate, the collective coherence operator between levels $i$ and $j$ is given by $S_{ij}$, and $g_{q,ij}$ represents atom-field coupling constant for the traveling wave field mode $q$ driving the $i-j$ atomic transition. This coupling constant is given by

$$g_{q,ij} = \frac{\mu_{ij}}{\hbar} \sqrt{\frac{\hbar \omega_q}{2 \epsilon_0 V_q}},$$

(3.4)

where $\mu_{ij}$ is the dipole matrix element between levels $i$ and $j$, and $V_q$ is the effective volume of the $q$-th fundamental cavity mode. Note that Eq. (3.3) includes the interaction energy associated with both fields driving each of the two allowed transitions. Moreover, it includes the counter-rotating terms associated with each field. This is in contrast to the case of near-resonance interactions where the counter-rotating terms, which do not “conserve energy” (so to speak), are neglected in the RWA as discussed by Scully and Zubairy [73, page 196] as well as Orszag [74, page 84]. In the present case, all of the terms are horrendously detuned and are therefore similarly abysmal in their (lack of) energy conservation. This severely decreases the probability for the transitions involved and also dictates that no single field component will dominate the interaction. They shall all, therefore, be retained.

**Irreversible Interaction Energy**

$H_3$ represents the coupling between the two active cavity modes and the external
field baths for decay and noise purposes and is given by

\[ H_3 = i\hbar \sum_{q=p,s} \int_{-\infty}^{\infty} d\omega \sqrt{\kappa_q / \pi} \left[ b_q(\omega) a_q - a_q^\dagger b_q(\omega) \right], \]

(3.5)

where the external field bath operators, \( b_q \) and \( b_q^\dagger \), are coupled to the \( q \)-th internal mode of the cavity through the (amplitude) cavity decay rate \( \kappa_q \). The decay rate, and therefore the mirror reflectivity for each mode is assumed to be constant over a large frequency bandwidth compared to the cavity resonance (the first Markoff approximation), which is easily achieved in practice. Mirror absorptions can be included in direct analogy with Eq. (3.5) [72].

\( H_4 \) represents the coupling between atomic coherences and atomic bath operators to generate damping and noise in the atoms and is given by

\[ H_4 = i\hbar \int_{-\infty}^{\infty} d\omega \left\{ \sqrt{\frac{\gamma_{21}}{2\pi}} \left[ B_{12}^\dagger(\omega) S_{12} - S_{12}^\dagger B_{12}(\omega) \right] \right. \]

\[ + \sqrt{\frac{\gamma_{31}}{2\pi}} \left[ B_{13}^\dagger(\omega) S_{13} - S_{13}^\dagger B_{13}(\omega) \right] \]

\[ + \sqrt{\frac{\gamma_{32}}{2\pi}} \left[ B_{23}^\dagger(\omega) S_{23} - S_{23}^\dagger B_{23}(\omega) \right] \right\}, \]  

(3.6)

where the the decay rates of the level populations, given by the \( \gamma_{ji} \)'s, can be interpreted as coupling constants between the system operators and the atomic bath operators, which are given by \( B_{ij} \) and \( B_{ij}^\dagger \). It has again been assumed that the decay rates are constant over a large frequency bandwidth compared to the atomic resonances (the first Markoff approximation). Population decay downward from levels 2 and 3 is assumed to be due to inelastic molecular collisions and spontaneous emission, respectively.
Similarly, $H_5$ represents the coupling between atomic populations and atomic bath operators to generate decay and noise of the atomic coherences through dephasing and is given by

$$H_5 = i\hbar \sum_{i=1}^{3} \int_{-\infty}^{\infty} d\omega \frac{\gamma_{ii}}{2\pi} \left[ B_{ii}^\dagger(\omega) S_{ii} - S_{ii} B_{ii}(\omega) \right],$$

(3.7)

where the $\gamma_{ii}$'s are the dephasing rates associated with each level due to elastic collisions, while $B_{ii}^\dagger$ and $B_{ii}$ are the corresponding atomic bath operators. This interaction is modeled after Gardiner and Zoller [53] and Eschmann et al. [68].

**Bath Energy**

Finally, $H_{\text{baths}}$ represents the free energy of the external bath or reservoir modes (one operator for each decay path available to the system) and is given by

$$H_{\text{baths}} = \sum_{q=p, s} \int_{-\infty}^{\infty} d\omega \hbar \omega b_q^\dagger(\omega)b_q(\omega) + \int_{-\infty}^{\infty} d\omega \hbar \omega \left[ B_{12}^\dagger(\omega)B_{12}(\omega) + B_{13}^\dagger(\omega)B_{13}(\omega) + B_{23}^\dagger(\omega)B_{23}(\omega) \right] + \sum_{i=1}^{3} \int_{-\infty}^{\infty} d\omega \hbar \omega B_{ii}^\dagger(\omega)B_{ii}(\omega).$$

(3.8)

The above components of $H$ do not deviate drastically from previous treatments aside from the reversible interaction energy $H_2$, which includes several additional terms due to the large single-photon detunings.

**Multiplications and Commutations**

The following sum will be referred to as the “system” Hamiltonian

$$H_{\text{sys}} = H_1 + H_2.$$
This is the portion of the total Hamiltonian that does not include the reservoirs or their interactions with the atoms and fields inside the cavity. The system operators are therefore the cavity field operators as well as the population and coherence operators. They obey the following standard equal-time commutation relations and multiplication rules

\[ [a_q, a_{q'}^+] = \delta_{qq'}, \quad S_{ij}S_{kl} = S_{il} \delta_{jk}, \quad [S_{ij}, S_{kl}] = S_{il} \delta_{jk} - S_{jk}^\dagger \delta_{il}, \]

(3.10)

where it should be noted that \( S_{jk}^\dagger = S_{kj} \). The reservoir operators obey standard boson commutation relations.

In the Heisenberg picture, one can now use the above Hamiltonian and commutation relations to generate the quantum Langevin equations for the system operators. An alternative approach is to proceed in the Schrödinger picture and generate a master equation for the time dependence of the density matrix. The benefits and drawbacks of these two approaches are discussed in Ref. [75].

**Quantum Langevin Equations**

In this section, the Heisenberg equation of motion is used to generate Langevin equations for the system operators. Unfortunately, as will become evident, the choice not to use the RWA complicates matters when converting to rotating coordinate frames. The crux of the problem lies in the fact that the (non-RWA) coherences are composed of multiple Fourier frequencies rather than just one. This means that there is no single coordinate frame to which one can transform in order to produce the
desired slowly varying coherence coefficients (one Fourier component will be slowly varying but the others will not). Fortunately, the Fourier components are spectrally separated by optical frequencies so that one can assume there is no mutual interaction. This allows a Langevin equation to be generated for each independent Fourier component of the $S_{13}$ and $S_{23}$ coherence operators. For reasons discussed below, this is not an issue for the two-photon coherence ($S_{12}$), the populations, or the fields, and they are readily converted to rotating frames. It must also be stated here that the equations generated in this section are undeniably nasty, but they can be interpreted from physical standpoints and are greatly simplified in the following section.

**Heisenberg Equation of Motion**

Using the Hamiltonian and commutation relations from the previous section, one can now calculate the quantum Langevin equations using the Heisenberg equation of motion. This procedure is greatly facilitated by the work of Gardiner and Collett [52] who showed that the Heisenberg equation of motion can take the form

$$\dot{a}_p = -\frac{i}{\hbar} [a_p, H_{sys}] - \sum_{j=1}^{8} \{ a_p, c_j \} \left\{ \frac{\gamma_j}{2} c_j - \sqrt{\gamma_j} b_{j_{in}}^\dagger(t) \right\}$$

for the pump field operator, for instance. The equations for the other system operators are obtained by simply replacing $a_p$ with the desired system operator. The first term on the right side accounts for the free evolution and reversible interactions of the system operators, whereas the last two terms generate decay and noise of the system.
operators through irreversible interactions with the reservoirs from $H_4$ and $H_5$ of the previous section.

The elements of the array $c$, which are indexed with the subscript $j$, include the system operators that are coupled to external reservoir operators, with the array given by

$$c = (a_p, a_s, S_{12}, S_{13}, S_{23}, S_{11}, S_{22}, S_{33}).$$

(3.12)

This array includes all of the system operators for the system considered here because each of the eight system operators is coupled to one reservoir as described in the previous section. The $\gamma_j$'s in Eq. (3.11) are the constants with which the system operators of $c$ are coupled to the reservoir operators from Eqs. (3.5) - (3.7). The $\gamma_j$'s also compose an array, which is given by

$$\gamma = (2\kappa_p, 2\kappa_s, \tilde{\gamma}_{21}, \tilde{\gamma}_{31}, \tilde{\gamma}_{32}, \tilde{\gamma}_{11}, \tilde{\gamma}_{22}, \tilde{\gamma}_{33}).$$

(3.13)

As described in Ref. [52], the $b_j^{\text{in}}$'s are reservoir input operators that result from the system-reservoir coupling. The array of input operators is given by

$$b^{\text{in}} = (a_p^{\text{in}}, a_s^{\text{in}}, B_{12}^{\text{in}}, B_{13}^{\text{in}}, B_{23}^{\text{in}}, B_{11}^{\text{in}}, B_{22}^{\text{in}}, B_{33}^{\text{in}}),$$

(3.14)

each component of which is defined in the following manner [76]

$$b_j^{\text{in}}(t) \equiv -\frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} dw e^{-i\omega(t-t_0)} b_j^0(\omega)$$

(t > t_0),

(3.15)

where $b_j^0(\omega)$ is the value of $b_j(\omega)$ at $t = t_0$ with the corresponding reservoir operators
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\( b_j(\omega) \) given by the array

\[
b = (b_p, b_s, B_{12}, B_{13}, B_{23}, B_{11}, B_{22}, B_{33}).
\]  

(3.16)

Note that the input operators given in Eqs. (3.14) and (3.15) have units of \( \sqrt{Hz} \) as opposed to the system operators, which are unitless. This is so the square of an input operator gives a photon flux (s\(^{-1}\)). It should also be pointed out that all of the input operators except for that of the pump represent stochastic noise inputs with zero expectation values (for instance, \( \langle a_s^{in} \rangle = 0 \)). The pump field input operator is a bit different because it is assumed that there exists an input pump beam into the cavity. For this case, \( \langle a_p^{in} \rangle = a_p^{in} \sqrt{\tau rt} \), where \( \tau rt \) is the round-trip time in the cavity and \( a_p^{in} \) is the unitless classical input pump field (a complex, or c-number; not an operator).

The input operators obey the same mutual commutation relations as their reservoir counterparts, given by

\[
[\hat{b}_j^{in}(t), \hat{b}_j^{in\dagger}(t')] = \delta_{jj'} \delta(t - t').
\]

(3.17)

However, it should be noted that although the reservoir operators (elements of \( b \)) commute with the system operators at equal times, this is not necessarily true for the input operators (elements of \( b^{in} \)), whose commutation relations are given in reference [52]. With the knowledge of the total Hamiltonian and all the pertinent commutation relations, Eq. (3.11) can be used to obtain a complete set of first order coupled differential equations for the system operators; the so-called quantum Langevin equations.
Rotating Coordinate Transformations

The standard next step is to use Eq. (3.11) to generate quantum Langevin equations of motion for the system operators. The operators are then transformed to rotating coordinate systems in order to extract the slowly varying behavior from the rapidly oscillating carrier frequencies.

In the present case, this procedure is straightforward for both of the field operators and for the 1–2 coherence operator. This is because the pump and Stokes field operators are very nearly resonant with the associated cavity modes \((\omega_p \approx \omega_p^c\) and \(\omega_s \approx \omega_s^c\)) and the two-photon Raman transition \((\omega_p - \omega_s \approx \omega_2 - \omega_1\)). Therefore, each of the three operators will exhibit slowly varying behavior around a single carrier frequency that is close to the corresponding resonance. Fourier components that are detuned from the resonances by optical frequencies \((\sim 100\text{THz})\) will be heavily attenuated because the cavity and Raman linewidths are \(\sim 1\text{MHz}\) and \(\sim 1\text{GHz}\), respectively. These components can be ignored when compared to the resonant components. The validity of this assumption can be proven rigorously. One can therefore perform the simple transformations

\[
a_p = a_p' e^{-i\omega_p t}, \quad a_s = a_s' e^{-i\omega_s t}, \quad \text{and} \quad S_{12} = S_{12}' e^{-i(\omega_p - \omega_s)t} \quad (3.18)
\]

to extract the slowly varying behavior (now contained in the primed operators) from the rapidly oscillating carrier frequencies. The associated noise operators are transformed similarly.

The situation is not so straightforward for the 1–3 and 2–3 coherence operators.
This is because the pump and Stokes field operators are not remotely resonant with the single-photon transitions \((\omega_p \neq \omega_3 - \omega_1 \text{ and } \omega_s \neq \omega_3 - \omega_2)\) as shown in Fig. 13. As a result, no carrier Fourier component dominates the interaction and all contribute significantly to the coherence. This means that there is no single rotating coordinate system to which one can transform \(S_{13}\) and \(S_{23}\) in order to extract the slowly varying behavior from the rapid carrier oscillations. A transformation directly following Eq. (3.18) will not produce a slowly varying coefficient because multiple carrier frequencies exist.

However, because the carrier frequencies differ from one another by optical frequencies, it is possible to treat the slowly varying behavior associated with each carrier frequency as a separate degree of freedom. As will become evident later in this section both \(S_{13}\) and \(S_{23}\) exhibit six carrier frequencies. One can therefore perform the transformations

\[
S_{13} = S_{13}^{(-\omega_p)} e^{-i\omega_p t} + S_{13}^{(+\omega_p)} e^{+i\omega_p t} + S_{13}^{(-\omega_s)} e^{-i\omega_s t} + S_{13}^{(+\omega_s)} e^{+i\omega_s t} \\
+ S_{13}^{(-\omega_{as})} e^{-i\omega_{as} t} + S_{13}^{(+\omega_{as})} e^{+i\omega_{as} t}, \tag{3.19}
\]

\[
S_{23} = S_{23}^{(-\omega_p)} e^{-i\omega_p t} + S_{23}^{(+\omega_p)} e^{+i\omega_p t} + S_{23}^{(-\omega_s)} e^{-i\omega_s t} + S_{23}^{(+\omega_s)} e^{+i\omega_s t} \\
+ S_{23}^{(-\omega_{ss})} e^{-i\omega_{ss} t} + S_{23}^{(+\omega_{ss})} e^{+i\omega_{ss} t}, \tag{3.20}
\]

where \(\omega_{as} \equiv 2\omega_p - \omega_s\) is the anti-Stokes frequency and \(\omega_{ss} \equiv 2\omega_s - \omega_p\) is the second Stokes frequency. All the coefficients in Eqs. (3.19) and (3.20) are slowly varying in time. Using the coordinate transformations of this subsection, it is now possible to
generate quantum Langevin equations for the slowly varying system operators.

Field Equations

Using Eqs. (3.11) and (3.18), the Langevin equations for the slowly varying field operators are found to be

\[
\dot{a}_p = - (\kappa_p + i\Delta_p) a_p + g_{p,13} S_{13}^{(-\omega_p)} + g_{p,13}^* \left( S_{13}^{(+\omega_p)} \right)^\dagger + g_{p,23} S_{23}^{(-\omega_p)} + g_{p,23}^* \left( S_{23}^{(+\omega_p)} \right)^\dagger + \sqrt{2\kappa_p} a_p^{in} 
\]

(3.21)

for the pump and

\[
\dot{a}_s = - (\kappa_s + i\Delta_s) a_s + g_{s,13} S_{13}^{(-\omega_s)} + g_{s,13}^* \left( S_{13}^{(+\omega_s)} \right)^\dagger + g_{s,23} S_{23}^{(-\omega_s)} + g_{s,23}^* \left( S_{23}^{(+\omega_s)} \right)^\dagger + \sqrt{2\kappa_s} a_s^{in} 
\]

(3.22)

for the Stokes, where the \( \Delta_q \equiv \omega_q^c - \omega_q \) represent the detunings of the driving frequencies from the cold cavity resonances. Note that the primes from Eq. (3.18) have been dropped and it should be understood that \( a_p \) and \( a_s \) now refer to slowly varying operators. Also note that only the components of \( S_{13} \) and \( S_{23} \) that produce oscillations near the cavity resonances have been retained. The nonresonant components have been discarded as discussed in the previous subsection. A sample derivation of Eq. (3.21) is provided in Appendix E.

Two-Photon Coherence Equation

The Langevin equation for the 1–2 coherence is obtained in a similar fashion with
the result

\[ \dot{S}_{12} = -(\gamma_{21} + i\Delta_{12})S_{12} \]

\[ + \alpha_p^* \left[ g_{p,23} S_{13}^{(-\omega_{as})} - g_{p,13}^* (S_{23}^{(+\omega_{as})})^\dagger \right] - \alpha_p \left[ g_{p,23} S_{13}^{(+\omega_{as})} - g_{p,13}^* (S_{23}^{(-\omega_{as})})^\dagger \right] \]

\[ + \alpha_s^* \left[ g_{s,23} S_{13}^{(-\omega_{sp})} - g_{s,13}^* (S_{23}^{(+\omega_{sp})})^\dagger \right] - \alpha_s \left[ g_{s,23} S_{13}^{(+\omega_{sp})} - g_{s,13}^* (S_{23}^{(-\omega_{sp})})^\dagger \right] \]

\[ + F_{12}, \quad (3.23) \]

where \( \Delta_{12} \equiv (\omega_2 - \omega_1) - (\omega_p - \omega_s) \) is the two-photon Raman detuning and again, only the near-resonant terms have been included. The nonresonant Fourier components are detuned from the Raman transition by optical frequencies and are therefore neglected.

The overall coherence decay constant is defined as

\[ \gamma_{21} \equiv \frac{1}{2} (\tilde{\gamma}_{21} + \tilde{\gamma}_{11} + \tilde{\gamma}_{22}) \], \quad (3.24) \]

and the noise term is

\[ F_{12} \equiv + \sqrt{\gamma_{21}} (S_{11} - S_{22}) B_{12}^{in} - \sqrt{\gamma_{31}} (S_{23}^{\dagger} E_{13}^{in})^{(-\omega_{21})} - \sqrt{\gamma_{22}} (B_{23}^{\dagger} S_{13}^{(-\omega_{21})}) \]

\[ - \sqrt{\gamma_{11}} (S_{12} B_{11}^{in} - B_{11}^{in\dagger} S_{12}^{\dagger}) + \sqrt{\gamma_{22}} (S_{12} B_{22}^{in} - B_{22}^{in\dagger} S_{12}^{\dagger}) \], \quad (3.25) \]

respectively, where the superscript \((-\omega_{21})\) indicates that only the products that oscillate near the Raman transition frequency are retained.

**Single-Photon Coherence Equations**

The 1–3 and 2–3 coherences are a bit more complicated. As alluded to earlier in this section, it is not possible to generate slowly varying Langevin equations for the
$S_{13}$ or $S_{23}$ operators themselves. Instead, one can generate slowly varying Langevin equations for the separate Fourier components of the two operators. To see this, one can first use Eq. (3.11) again to generate Langevin equations that are not slowly varying for $S_{13}$ and $S_{23}$. With the use of Eq. (3.18), this yields

$$\dot{S}_{13} = -[\gamma_{31} + i(\omega_3 - \omega_1)] S_{13}$$

$$- \left[ g_{s,13}^* a_p (S_{11} - S_{33}) + g_{s,23}^* a_s S_{12} \right] \times e^{-i\omega_p t}$$

$$+ \left[ g_{s,13}^* a_p^\dagger (S_{11} - S_{33}) + g_{s,23}^* a_p^\dagger S_{12} \right] \times e^{+i\omega_p t}$$

$$+ g_{s,13}^* a_p^\dagger (S_{11} - S_{33}) \times e^{+i\omega_p t} - g_{s,13}^* a_s (S_{11} - S_{33}) \times e^{-i\omega_p t}$$

$$- g_{s,23}^* a_p S_{12} \times e^{-i(2\omega_p - \omega_3)t} + g_{s,23}^* a_s S_{12} \times e^{+i(2\omega_p - \omega_3)t} + F_{13}, \quad (3.26)$$

$$\dot{S}_{23} = -[\gamma_{32} + i(\omega_3 - \omega_2)] S_{23}$$

$$- \left[ g_{s,23}^* a_s (S_{22} - S_{33}) + g_{s,13}^* a_p S_{12}^\dagger \right] \times e^{-i\omega_p t}$$

$$+ \left[ g_{s,23}^* a_p^\dagger (S_{22} - S_{33}) + g_{s,13}^* a_p^\dagger S_{12}^\dagger \right] \times e^{+i\omega_p t}$$

$$+ g_{s,23}^* a_p^\dagger (S_{22} - S_{33}) \times e^{+i\omega_p t} - g_{s,23}^* a_p (S_{22} - S_{33}) \times e^{-i\omega_p t}$$

$$- g_{s,13}^* a_s S_{12}^\dagger \times e^{-i(2\omega_p - \omega_2)t} + g_{s,13}^* a_p S_{12}^\dagger \times e^{+i(2\omega_p - \omega_2)t} + F_{23}, \quad (3.27)$$

where all the coefficients of the exponentials are slowly varying in time. The primes on the slowly varying field operators have again been dropped. A sample derivation of Eq. (3.26) is provided in Appendix F. The decay constants in these equations are

$$\gamma_{31} = \frac{1}{2} \left( \bar{\gamma}_{31} + \bar{\gamma}_{32} + \bar{\gamma}_{11} + \bar{\gamma}_{33} \right), \quad (3.28)$$
and

\[ \gamma_{32} \equiv \frac{1}{2} (\gamma_{32} + \gamma_{31} + \gamma_{21} + \gamma_{33} + \gamma_{22}), \]  

(3.29)

and the noise terms are

\[ F_{13} \equiv \sqrt{\gamma_{31}} (S_{11} - S_{33}) B_{13}^{\text{in}} + (\sqrt{\gamma_{32}} S_{12} B_{23}^{\text{in}} - \sqrt{\gamma_{21}} S_{23} B_{12}^{\text{in}}) \times e^{-i(\omega_p - \omega_s)t} \]

\[ - \sqrt{\gamma_{11}} (S_{13} B_{11}^{\text{in}} - B_{11}^{\dagger \text{in}} S_{13}) + \sqrt{\gamma_{33}} (S_{13} B_{23}^{\text{in}} - B_{23}^{\dagger \text{in}} S_{13}), \]  

(3.30)

and

\[ F_{23} \equiv \sqrt{\gamma_{33}} (S_{22} - S_{33}) B_{23}^{\text{in}} + (\sqrt{\gamma_{31}} S_{12}^{\dagger} B_{13}^{\text{in}} + \sqrt{\gamma_{21}} B_{12}^{\text{in}} S_{13}) \times e^{+i(\omega_p - \omega_s)t} \]

\[ - \sqrt{\gamma_{22}} (S_{23} B_{22}^{\text{in}} - B_{22}^{\dagger \text{in}} S_{23}) + \sqrt{\gamma_{33}} (S_{23} B_{33}^{\text{in}} - B_{33}^{\dagger \text{in}} S_{23}). \]  

(3.31)

The forms of Eqs. (3.26) and (3.27) make clear the fact that the 1–3 and 2–3 coherences each exhibit Fourier components at six different carrier frequencies. But because none of them are nearly resonant with the atomic transition, one must assume that they all contribute in a non-negligible manner to the coherences. Note the appearance of carrier Fourier components at the anti-Stokes \((\omega_{as} \equiv 2\omega_p - \omega_s)\) and second Stokes \((\omega_{ss} \equiv 2\omega_s - \omega_p)\) frequencies. Generation of light at the anti-Stokes frequency is addressed in Appendix G.

To generate slowly varying operators for the 1–3 and 2–3 coherences, one can now substitute Eqs. (3.19) and (3.20) into Eqs. (3.26) and (3.27). Because the carrier frequencies differ from one another by optical frequencies, while the coefficients are all slowly varying in time, the terms with different carrier frequencies will not interact.
This can be proven rigorously. Therefore, the terms with like carrier frequencies can be equated to generate six separate slowly varying first-order differential equations; one for each carrier frequency. For the 1–3 coherence, these are given by

\[
\begin{align*}
\dot{S}_{13}^{(-\omega_p)} &= -[\gamma_{31} + i\Delta] S_{13}^{(-\omega_p)} - g_{p,13}^* a_p (S_{11} - S_{33}) - g_{s,23}^* a_s S_{12} + F_{13}^{(-\omega_p)}, \\
\dot{S}_{13}^{(+\omega_p)} &= -[\gamma_{31} + i\Delta] S_{13}^{(+\omega_p)} + g_{p,13}^* a_p^\dagger (S_{11} - S_{33}) + F_{13}^{(+\omega_p)}, \\
\dot{S}_{13}^{(-\omega_s)} &= -[\gamma_{31} + i\Delta_s] S_{13}^{(-\omega_s)} - g_{s,13}^* a_s (S_{11} - S_{33}) + F_{13}^{(-\omega_s)}, \\
\dot{S}_{13}^{(+\omega_s)} &= -[\gamma_{31} + i\Delta] S_{13}^{(+\omega_s)} + g_{s,13}^* a_s^\dagger (S_{11} - S_{33}) + g_{p,23}^* a_p^\dagger S_{12} + F_{13}^{(+\omega_s)}, \\
\dot{S}_{13}^{(-\omega_as)} &= -[\gamma_{31} + i\Delta_s] S_{13}^{(-\omega_as)} - g_{s,13}^* a_p S_{12} + F_{13}^{(-\omega_as)}, \\
\dot{S}_{13}^{(+\omega_as)} &= -[\gamma_{31} + i\Delta_as] S_{13}^{(+\omega_as)} + g_{s,23}^* a_s^\dagger S_{12} + F_{13}^{(+\omega_as)}.
\end{align*}
\]

This last step is only possible because the slowly varying portions corresponding to each carrier frequency do not overlap spectrally. The new slowly varying operators represent separate degrees of freedom (they commute). The detunings in these equations are defined in Table 1 and are shown in Fig. 13.

<table>
<thead>
<tr>
<th>Rotating</th>
<th>Counter-Rotating</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta = (\omega_3 - \omega_1) - \omega_p \approx (\omega_3 - \omega_2) - \omega_s$</td>
<td>$\Delta' = (\omega_3 - \omega_1) + \omega_s \approx (\omega_3 - \omega_2) + \omega_p$</td>
</tr>
<tr>
<td>$\Delta_{as} = (\omega_3 - \omega_2) - \omega_p$</td>
<td>$\Delta'_{as} = (\omega_3 - \omega_1) + \omega_p$</td>
</tr>
<tr>
<td>$\Delta_{ss} = (\omega_3 - \omega_1) - \omega_s$</td>
<td>$\Delta'_{ss} = (\omega_3 - \omega_2) + \omega_s$</td>
</tr>
</tbody>
</table>
The analogous slowly varying equations corresponding to the Fourier components of $S_{23}$ are the same as Eqs. (3.32a) - (3.32f) but with the transformations $p = s$, $s = ss$ and $1 \Rightarrow 2$. The symmetry that allows these transformations is also evident from Fig. 13.

**Population Equations**

The populations are also obtained using Eq. (3.11) and are given by

\[
\dot{S}_{11} = \gamma_{21} S_{22} + \gamma_{31} S_{33} + \left[ g_{p,13} a_p^\dagger S_{13}^{(-\omega_p)} - g_{p,13} a_p S_{13}^{(+\omega_p)} + g_{s,13} a_s^\dagger S_{13}^{(-\omega_s)} - g_{s,13} a_s S_{13}^{(+\omega_s)} + \text{H.c.} \right] + F_{11}, \tag{3.33}
\]

\[
\dot{S}_{22} = \gamma_{32} S_{33} - \gamma_{21} S_{22} + \left[ g_{p,23} a_p^\dagger S_{23}^{(-\omega_p)} - g_{p,23} a_p S_{23}^{(+\omega_p)} + g_{s,23} a_s^\dagger S_{23}^{(-\omega_s)} - g_{s,23} a_s S_{23}^{(+\omega_s)} + \text{H.c.} \right] + F_{22}, \tag{3.34}
\]

\[
\dot{S}_{33} = -\dot{S}_{11} - \dot{S}_{22}, \tag{3.35}
\]

where atom conservation ($S_{11} + S_{22} + S_{33} = N$) has been used in obtaining Eq. (3.35), and the noise terms are given by

\[
F_{11} \equiv -\sqrt{\gamma_{21}} (S_{12}^\dagger B_{12}^{\text{in}} + B_{12}^{\text{in}\dagger} S_{12}) - \sqrt{\gamma_{31}} (S_{13}^\dagger B_{13}^{\text{in}} + B_{13}^{\text{in}\dagger} S_{13})^{(\text{dc})}, \tag{3.36}
\]

and

\[
F_{22} \equiv -\sqrt{\gamma_{32}} (S_{23}^\dagger B_{23}^{\text{in}} + B_{23}^{\text{in}\dagger} S_{23})^{(\text{dc})} + \sqrt{\gamma_{21}} (S_{12}^\dagger B_{12}^{\text{in}} + B_{12}^{\text{in}\dagger} S_{12}^{(\text{dc})}), \tag{3.37}
\]

where the superscript (dc) indicates that only the dc Fourier components are retained.

The next section is devoted to simplifying the rather unwieldy quantum Langevin equations that were developed in this section.
Simplifications Based on Large Detunings

Up to this point, the large single-photon detunings have only complicated matters by adding additional Fourier components to the single-photon coherences. In this section, the large detunings are used to our advantage by simplifying the quantum Langevin equations. For reference purposes, estimated values of the pertinent rates in this system are provided in Table 2.

Table 2. Parameters used for simplifying the quantum Langevin equations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effective Atom/Field Coupling Constants</td>
<td>$g_{q,ij}/2\pi$</td>
<td>$\sim 10^4 - 10^5$ Hz</td>
</tr>
<tr>
<td>Level 2 Population Decay rate</td>
<td>$\gamma_{21}/2\pi$</td>
<td>$\sim 10^4 - 10^5$ Hz</td>
</tr>
<tr>
<td>Cavity Amplitude Decay Rates</td>
<td>$\kappa_{q}/2\pi$</td>
<td>$\sim 10^5 - 10^6$ Hz</td>
</tr>
<tr>
<td>Level 2 Coherence Decay rate</td>
<td>$\gamma_{21}/2\pi$</td>
<td>$\sim 10^8 - 10^9$ Hz</td>
</tr>
<tr>
<td>Level 3 Coherence Decay Rates</td>
<td>$\gamma_{31}/2\pi$, $\gamma_{32}/2\pi$</td>
<td>$\sim 10^9 - 10^{10}$ Hz</td>
</tr>
<tr>
<td>Single-Photon Rabi Frequencies</td>
<td>$\Omega_{q,ij}/2\pi$</td>
<td>$\lesssim 10^{10}$ Hz</td>
</tr>
<tr>
<td>Single-Photon Detunings</td>
<td>$\Delta/2\pi$</td>
<td>$\sim 10^{15} - 10^{16}$ Hz</td>
</tr>
</tbody>
</table>

The extreme single-photon detunings allow one to make the following simplifications:

(i) Adiabatically eliminate the upper level coherences. As shown by Raymer et al. for a single frequency component [77], all of the 1–3 and 2–3 coherence Fourier components can be adiabatically eliminated when the single-photon detunings are much larger than the other rates in the system. One can therefore solve for the
steady-state Fourier components from Eqs. (3.32a)-(3.32f) and their $S_{32}$ counterparts and insert these into the remaining six equations.

(ii) Disregard the single-photon absorption/gain and mode pulling. Terms arise in the two field equations that represent linear absorption/gain (real parts) as well as dispersion (imaginary parts). For near-resonance systems, the real portions can supply the population inversion necessary for laser gain. In the present system, they effectively broaden and pull the frequency of the cavity resonances due to single-photon interactions with the medium. In the limit $\Delta^2 \kappa_q \gg \gamma_{ij} |g_{q,ij}|^2$, where the subscript $q$ denotes an optical mode and $ij$ refers to a single-photon transition, the absorption terms can be disregarded. The mode pulling is small and is assumed here to be nullified by active electronic stabilization of the cavity length to the pump laser frequency (i.e. the cavity length is changed to compensate for the refractive index change). It is also assumed that the Stokes field will build on the active cavity resonance line-center. Appendix H describes this simplification in more detail.

(iii) Ignore power broadening and AC Stark shifts. Terms arise in the 1–2 coherence equation that are quadratic in the field operators (linear in optical power), and linear in the coherence. In direct analogy with simplification (ii), these terms cause power broadening (real part) and AC Stark shifts (imaginary parts) of the two-photon (1–2) atomic transition. In the limit $\Delta^2 \gamma_{21} \gg \gamma_{ij} |\Omega_{q,ij}|^2$, where $\Omega_{q,ij} \equiv g_{q,ij} a_q$ is the Rabi frequency for the optical field $q$ driving the $ij$ single-photon atomic transition, the power broadening can be ignored. The AC Stark shift is predicted to be very
mild ($\lesssim 10 \text{MHz}$) compared to the two-photon resonance width, $\gamma_{21}$, for the optical powers considered, and can be compensated easily in practice by tuning the pump laser (with the cavity following). The AC Stark shift is much larger and plays a critical role in electromagnetically induced transparency [78, and references therein]. Appendix I describes this simplification in more detail.

(iv) Neglect spontaneous emission. In the limit of large single-photon detuning relative to the level 3 decay rate, one can make the approximation $(\gamma_{31} + i\Delta)^{-1} \approx (i\Delta)^{-1}$ and likewise for similar terms. As one might expect from the fluctuation/dissipation theorem [73], because the upper level decay can be neglected, it can also be shown that that the associated noise terms entering from the 1–3 and 2–3 coherences are severely diminished by the detuning as well. In the above limits, spontaneous emission will therefore contribute negligibly to the noise of the field and remaining coherence operators, and is neglected (i.e. ignore $F_{13}$ and $F_{23}$).

With these simplifications, the field and remaining coherence equations become

\begin{align}
\dot{a}_p &= -\kappa_p a_p + i \left( \frac{1}{\Delta} + \frac{1}{\Delta_r} \right) g_{p,13} g_{s,23}^* a_s S_{12} + \sqrt{2\kappa_p} a_p^\text{in}, \\
\dot{a}_s &= -\kappa_s a_s + i \left( \frac{1}{\Delta} + \frac{1}{\Delta_r} \right) g_{p,13}^* g_{s,23} a_p S_{12}^\dagger + \sqrt{2\kappa_s} a_s^\text{in}, \\
\dot{S}_{12} &= - (\gamma_{21} + i\Delta_{12}) S_{12} + i \left( \frac{1}{\Delta} + \frac{1}{\Delta_r} \right) g_{p,13}^* g_{s,23} (S_{11} - S_{22}) a_p a_s^\dagger + F_{12}.
\end{align}

These equations are significantly more manageable than those of the previous section. In essence, the system initially described by Fig. 13 can now be described by the simpler energy level diagram given in Fig. 14. The large single-photon detunings
have allowed the elimination of all the interactions that are not Raman resonant. Furthermore, the only vestige of *not* invoking the RWA is the $\Delta'$ in the gain term of each equation. This extra term can be viewed as the contribution from the counter-rotating version of the Raman process, as can be seen in Fig. 14. Clearly, when the fields are nearly resonant with the single-photon transitions ($\Delta' \gg \Delta$) this contribution becomes negligible. Neglecting the $\Delta'$ contribution in this limit is precisely the RWA.

![Figure 14. To-scale energy level diagram for the diatomic hydrogen molecule after simplifications based on large single-photon detunings.](image)
Similarly, after adiabatic elimination of the upper level coherences, the populations become

\[ \dot{S}_{11} = \bar{\gamma}_{31} S_{33} + \bar{\gamma}_{21} S_{22} \]
\[ - \left[ \frac{1}{\Delta^2} + \frac{1}{(\Delta_{ss})^2} \right] 2\gamma_{31}|g_{p,13}|^2 (S_{11} - S_{33})|a_p|^2 \]
\[ - \left[ \frac{1}{(\Delta')^2} + \frac{1}{(\Delta_{ss})^2} \right] 2\gamma_{33}|g_{s,13}|^2 (S_{11} - S_{33})|a_s|^2 \]
\[ + \left[ i \left( \frac{1}{\Delta} + \frac{1}{\Delta'} \right) g_{p,13}^* g_{s,23} a_p^\dagger a_s S_{12} + \text{H.c.} \right] + F_{11}, \quad (3.41) \]

\[ \dot{S}_{22} = \bar{\gamma}_{32} S_{33} - \bar{\gamma}_{21} S_{22} \]
\[ - \left[ \frac{1}{\Delta^2} + \frac{1}{(\Delta_{ss})^2} \right] 2\gamma_{32}|g_{s,23}|^2 (S_{22} - S_{33})|a_s|^2 \]
\[ - \left[ \frac{1}{(\Delta')^2} + \frac{1}{(\Delta_{ss})^2} \right] 2\gamma_{33}|g_{p,23}|^2 (S_{22} - S_{33})|a_p|^2 \]
\[ + \left[ i \left( \frac{1}{\Delta} + \frac{1}{\Delta'} \right) g_{p,13}^* g_{s,23} a_p a_s^\dagger S_{12} + \text{H.c.} \right] + F_{22}, \quad (3.42) \]

\[ \dot{S}_{33} = - \dot{S}_{11} - \dot{S}_{22}, \quad (3.43) \]

where the terms arising from single-photon interactions have been retained here so that the populations of the upper level can be estimated later.

**Semiclassical Analysis**

At this point, one can take the semiclassical limit of the remaining field and atomic equations of motion. This simply entails taking the expectation values of Eqs. (3.38) - (3.43). As noted previously, this eliminates the contributions from the
input operators (except the input pump operator) because the expectation values of their stochastic noise contributions are zero. The following definitions are therefore assigned to distinguish the semiclassical complex variables (c-numbers) from the quantum operators

\[ \alpha_q \equiv \langle a_q \rangle, \quad \alpha_p^{in} \equiv \sqrt{\tau_{rt}} \langle a_p^{in} \rangle, \quad \text{and} \quad J_{ij} \equiv \frac{1}{N} \langle S_{ij} \rangle. \] (3.44)

All of the newly assigned variables are unitless. It can be noted here that \(|\alpha_p^{in}|^2\) now represents the unitless semiclassical incident pump power, which is valid for large photon number. Also note that the total population variables have been renormalized to unity so that \(J_{11} + J_{22} + J_{33} = 1\).

Furthermore, because the atomic decay rates \(\gamma_{21}, \gamma_{31}, \) and \(\gamma_{32}\) are all much larger than the cavity decay rates \(\kappa_p\) and \(\kappa_s\), one can also adiabatically eliminate the 1–2 coherence and the level 3 population. One can therefore use the steady-state values of these two variables in the remaining four equations of motion.

**Simplified Population Equations**

Solving for the steady-state 1–2 coherence and plugging it in to the steady state-level 3 population yields

\[ J_{33} = \frac{\Gamma_{13}}{(\gamma_{31} + \gamma_{32})} J_{11} + \frac{\Gamma_{23}}{(\gamma_{31} + \gamma_{32})} J_{22}, \] (3.45)

where the single-photon absorption (upward transition) rates of population from level
1 to 3 and from level 2 to 3 are defined as

\[
\Gamma_{13} = \left( \frac{\Omega_{p,13}^2}{\Delta^2} + \frac{|\Omega_{s,13}|^2}{(\Delta')^2} + \frac{|\Omega_{s,13}|^2}{(\Delta_{ss})^2} \right)^2 \gamma_{s1}, \tag{3.46}
\]

\[
\Gamma_{23} = \left[ \frac{|\Omega_{s,23}|^2}{\Delta^2} + \frac{|\Omega_{s,23}|^2}{(\Delta_{ss})^2} + \frac{|\Omega_{p,23}|^2}{(\Delta')^2} + \frac{|\Omega_{p,23}|^2}{(\Delta_{ss})^2} \right] 2\gamma_{s2}, \tag{3.47}
\]

respectively. Because the single-photon detunings (\(\Delta's\)) are all much larger than any of the Rabi frequencies (\(\Omega's\)), one can conclude

\[
J_{33} \ll 1. \tag{3.48}
\]

This simply verifies what may have been suspected from the start; the two-photon Raman process contributes nothing to the upper level population and this population is much less than unity when the single-photon detunings are much greater than the Rabi frequencies.

Similarly, one can define the upward transition rate that is due to the two-photon Raman process as

\[
\Gamma_{12} = \left( \frac{\gamma_{21}}{\gamma_{21} + \Delta_{12}} \right) \left( \frac{1}{\Delta} + \frac{1}{\Delta'} \right)^2 |\Omega_{p,13}|^2 |\Omega_{s,23}|^2, \tag{3.49}
\]

which has typical value of \(\Gamma_{12}/2\pi \lesssim 1\text{Hz} \) for the fields and detunings of this system. This means that each atom in the beam path is excited by the two-photon process on average less than once per second. The remaining two populations, which are still time dependent, can now be simplified to

\[
\dot{J}_{22} = -\gamma_{21} J_{22} + \Gamma_{12}, \tag{3.50}
\]

\[
\dot{J}_{11} = -\dot{J}_{22}, \tag{3.51}
\]
where the fact that $\bar{\gamma}_{21} \gg \Gamma_{12}, \Gamma_{23}, \Gamma_{13}$ has been used. These represent simple first order differential equations except for the fact that there is actually field dependence hidden within $\Gamma_{12}$ in the form of Rabi frequencies. To approximate the behavior of the populations analytically, one can therefore assume that the fields decay rapidly compared to level 2 population. This is typically a very reasonable assumption. In that case, all coefficients are constant and the differential equations can be solved analytically to give

$$J_{22}(t) = \frac{\Gamma_{12}}{\bar{\gamma}_{21}} [1 - \exp (-\bar{\gamma}_{21} t)], \quad (3.52)$$

$$J_{11}(t) = 1 - J_{22}(t), \quad (3.53)$$

where it has been assumed that all the population is initially in the ground state. Equation (3.52) is plotted in Fig. 15, where it can be seen that the level two population simply rises monotonically to its steady-state value of $\Gamma_{12}/\bar{\gamma}_{21}$. The time it takes to reach steady state depends only on the level 2 decay rate $\bar{\gamma}_{21}$ for the conditions considered here.
Because the two-photon upward transition rate $\Gamma_{12}$ is much smaller than the level 2 decay rate $\gamma_{21}$, one can conclude that for all times

\[ J_{22} \ll 1, \quad \text{and} \quad J_{11} \approx 1. \tag{3.54} \]

In other words, one can assume that all the population remains in the ground state at all times. This is a critical finding that will simplify future calculations and that will also cause this Raman system to behave like a parametric system.
Simplified Field Equations

Using this knowledge, one can obtain the following simplified Raman field equations

\[ \dot{\alpha}_p = -\kappa_p \alpha_p - G_1 |\alpha_s|^2 \alpha_p + \tau_{rt}^{-1} \sqrt{T_{p,0}} \alpha_p^{in}, \quad (3.55) \]

\[ \dot{\alpha}_s = -\kappa_s \alpha_s + G_1 |\alpha_p|^2 \alpha_s, \quad (3.56) \]

where the Raman gain (rad/s) is defined as

\[ G_1 = \left( \frac{\gamma_{21} N}{\gamma_{21}^2 + \Delta_{12}^2} \right) \left( \frac{1}{\Delta} + \frac{1}{\Delta'} \right)^2 |g_{p,13}|^2 |g_{s,23}|^2, \quad (3.57) \]

and dispersion due to the Raman resonance (imaginary contribution to \( G_1 \)) has been neglected because it is small (\( \lesssim 1 \text{MHz} \) mode pulling relative to a \( \sim 500 \text{MHz} \) Raman linewidth). Note that the field equations decouple from those of the populations because the amount of population in the upper two states is not sufficient to affect the dynamics of the fields.\(^1\) These equations exactly match the forms of Eqns. (B.36) and (B.37) from Appendix B (note, however, that \( G_1 \neq G \)). This consistency between the classical and quantum mechanical derivations is reassuring. Of course, the steady-state limits of Eqs (3.55) and (3.56) also match the classical results.

Steady-State Behavior

Setting Eqs. (3.55) and (3.56) to zero gives a stable steady-state solution of zero for the (unitless) intracavity Stokes power below threshold. Above threshold, the

\(^1\) Strictly speaking, when thermal effects are included, the fields once again become coupled to the populations.
solutions for the intracavity pump and Stokes unitless powers are

\[ |\alpha_p|^2 = \frac{\kappa_s}{G_1}, \quad (3.58) \]
\[ |\alpha_s|^2 = \frac{\kappa_p}{G_1}(\sqrt{r_p} - 1), \quad (3.59) \]

where the cavity field decay rates are given by \( \kappa \approx (1 - R_n)/2\tau_n \) and the pump rate, defined by the relation

\[ r_p \equiv \frac{|\alpha_p^{in}|^2}{|\alpha_p^{th}|^2}, \quad (3.60) \]

is the incident unitless pump power normalized to threshold. The unitless input threshold pump power is given by

\[ |\alpha_p^{th}|^2 = \frac{\kappa_p^2\kappa_s^2}{G_1T_{p,0}}. \quad (3.61) \]

Note that in the process of obtaining these equations from Eqs. (3.55) - (3.56), determination of the Stokes field phase is not permitted. This is a characteristic common to phase insensitive amplification processes and indicates phase diffusion of the Stokes field [79]. Note also that the association of quantities like |\( \alpha_p \)|\(^2\) with an optical power is a semiclassical approximation that is only strictly valid for photon numbers much greater than one.

The unitless powers from above can be converted to optical powers with standard units [J/s] using the relation

\[ P_q = \frac{\hbar\omega_0}{\tau_{rt}}|\alpha_q|^2. \quad (3.62) \]
Equations (3.58) - (3.59) can then be directly compared to Eqs. (2.15) - (2.17) from Chapter 2. This allows one to solve for $G_1$ in terms of measurable quantities, with the result

$$G_1 = \frac{8\hbar \omega_p \alpha \tan^{-1}(L/b)}{\tau_{rt}^2 \lambda_p + \lambda_s},$$

(3.63)

where $\alpha$ is the plane wave gain coefficient and the fact that $\tau_{rt} = 4L/c$ has been used. By solving Eq. (3.63) for $\alpha$ and using Eq. (3.57), one might be tempted to interpret the resulting relation as a definition of $\alpha$ in terms of fundamental parameters of the medium. This would be reasonable if the fields were nearly resonant with the single-photon transitions. For the present far-off-resonance case, however, one must remember that only one upper level (level 3) was considered in this work, whereas all the upper levels (including the continuum) must be included for an accurate estimation of $\alpha$ based on the fundamental parameters of the medium.

Even though the fields no longer depend on the populations, the reverse is definitely not true. The simplified steady-state populations are given by

$$J_{22} = \frac{\Gamma_{12}}{\bar{\gamma}_{21}} \lesssim 10^{-4},$$

(3.64)

$$J_{33} = \frac{\Gamma_{13}}{(\bar{\gamma}_{31} + \bar{\gamma}_{32})} \lesssim 10^{-10},$$

(3.65)

$$J_{11} = 1 - J_{33} - J_{22} \approx 1,$$

(3.66)

where the fields are embedded as Rabi frequencies in the $\Gamma$'s. It should also be noted that the thermal population of level 2 is $\sim 10^{-9}$ for the system at hand. Despite the meager amount of upper state population, the atoms decaying from level 2 can
still dramatically affect the behavior of the fields via heat deposition, as discussed in Chapter 4.

Spatial Considerations

As a side note, the dependence of the gain $G_1$ from Eq. (3.57) on the spatial characteristics of the pump and Stokes cavity modes can be incorporated by simply defining the number of atoms as that contained in the effective interaction volume of the two cavity modes. That is

$$N = \rho V_{\text{int}}, \quad (3.67)$$

where $\rho$ is the gas density in the Raman cell and the effective volume of the interaction region is given by

$$V_{\text{int}} \equiv \int \int \int_{\text{cavity}} |\bar{u}_p(r)|^2 |\bar{u}_s(r)|^2 dr, \quad (3.68)$$

where $\bar{u}_p(r)$ and $\bar{u}_s(r)$ are the pump and Stokes spatial modes of the cavity. This is effectively equivalent to including the Boyd-Klienman factor, which was performed in the classical regime for Raman interactions in Ref. [40]. It can be shown rigorously within a quantum mechanical formalism using methods similar to those of Xiao, et al. [80].

Connection to the NDOPO

Returning to the fully quantum formalism, but with the knowledge that the vast majority of the population always remains in the ground state, Eqs. (3.38) - (3.40)
can be simplified by setting $S_{11} - S_{22} = N$, where $N$ is the total population in the interaction volume. Furthermore, although it may not seem natural at the moment, one can also make the enlightening variable changes

$$S'_{12} = \frac{S_{12}}{\sqrt{N}} \quad \text{and} \quad S^\text{in}_{12} = \frac{1}{\sqrt{2\gamma_21 N}} F_{12},$$

where the noise term $F_{12}$ is given by Eq. (3.25). Substitution of these into Eqs. (3.38) - (3.40) and dropping the prime for notational simplicity yields

$$\dot{a}_p = -\kappa_p a_p + ig a_s S_{12} + \sqrt{2\kappa_p} a^\text{in}_p,$$

$$\dot{a}_s = -\kappa_s a_s + ig^* a_p S^\dagger_{12} + \sqrt{2\kappa_s} a^\text{in}_s,$$

$$\dot{S}_{12} = -(\gamma_{21} + i\Delta_{12}) S_{12} + ig^* a_p a_s^\dagger + \sqrt{2\gamma_{21}} S^\text{in}_{12},$$

where

$$g = \sqrt{N} \left( \frac{1}{\Delta} + \frac{1}{\Delta^*} \right) g_{p,13} g_{s,23}^*.$$  

A clear symmetry now begins to emerge between Eqs. (3.71) and (3.72). Indeed, the $S_{12}$ operator plays a very similarly to the Stokes field operator in Eqs. (3.70) - (3.72). Moreover, the similarities between $S_{12}$ and an optical mode annihilation operator extend beyond the semiclassical domain as shown in the next subsection.

**Operator Correlation Functions**

To determine the intensity noise spectra of the emitted light from this system, one must calculate the second order correlation functions of the input operators. For the
field input operators, this is not too tricky, but the coherence input operator requires
a bit more thought. Here it is shown that all of the input operators, including $S_{12}^{\text{in}}$

exhibit identical second order correlation functions.

The nonvanishing second order correlation functions of the field operators are

$$\langle a_p^{\text{in}}(t)a_p^{\text{in} \dagger}(t') \rangle = \langle a_q^{\text{in}}(t)a_q^{\text{in} \dagger}(t') \rangle = \delta(t - t'), \quad (3.74)$$

where the commutation relations from Eq. (3.17) have been used and it has been
assumed that the input fluctuations are ordinary vacuum so that $\langle a_q^{\text{in} \dagger} \rightarrow 0$ and
$a_q^{\text{in}} \rightarrow 0$.

To calculate the correlation functions for the coherence input operator, one has
two choices. First one can continue to use the normal rules of calculus and apply the
commutation rules between the system and input operators given in Ref. [52]. For
this path, some extra terms show up in the second order correlations that eventually
integrate to zero when the noise spectra is calculated. An easier path is to convert
to Ito calculus, where the rules of calculus are different, but the system operators
commute with the input operators. In this case, no extra terms appear in the second-
order correlations. Identical results are obtained using either approach.

Following the latter path, one can verify that the 1–2 input operator is delta
correlated $[\langle S_{12}^{\text{in}}(t)S_{12}^{\text{in} \dagger}(t') \rangle = \delta(t - t')]$ when the input fluctuations are vacuum, just
like the field operators. That is, using the commutation relations from Eq. (3.17),
the definitions given by Eqs. (3.69) and (3.25), and the fact that the input operators
commute with the system operators for Ito calculus, one can show

\[ \langle S_{12}^{\text{in}}(t)S_{12}^{\text{in}}(t') \rangle = \langle S_{12}^{\text{in} \dagger}(t)S_{12}^{\text{in} \dagger}(t') \rangle = 0 \]  
(3.75)

\[ \langle S_{12}^{\text{in} \dagger}(t)S_{12}^{\text{in}}(t') \rangle = \frac{\tilde{\gamma}_{32}}{N^2 \gamma_{21}} \langle S_{33}(t) \rangle \delta(t - t') \approx 0 \]  
(3.76)

\[ \langle S_{12}^{\text{in}}(t)S_{12}^{\text{in} \dagger}(t') \rangle = \frac{1}{N^2 \gamma_{21}} \left[ \bar{\gamma}_{21} \langle S_{11}(t) + S_{33}(t) \rangle + \bar{\gamma}_{31} \langle S_{33}(t) \rangle \right] \delta(t - t') \]  

\[ \approx \frac{1}{2 \gamma_{21}} (\bar{\gamma}_{21} + \bar{\gamma}_{11} + \bar{\gamma}_{22}) \delta(t - t') \]  

\[ = \delta(t - t'), \]  
(3.77)

just as for the input operator of an optical mode. Note that this is a luxury afforded by the dearth of population in the upper two states.

**System Comparison**

With the equivalence of the input operator correlation functions established, one can now say that the quantum cw Raman laser equations defined by Eqs. (3.70) - (3.72) are formally identical to those of the nondegenerate optical parametric oscillator (NDOPO) [81] with the following associations: Pump $\leftrightarrow$ Pump, Stokes $\leftrightarrow$ Signal, 1–2 Coherence $\leftrightarrow$ Idler.

The connection between the stimulated Raman and optical parametric processes was actually established from a theoretical standpoint shortly after the discovery of both [82]. At that time, both of the processes were only experimentally achievable in the pulsed regime, making precise experimental comparisons difficult. Since then cw NDOPOs have been experimentally realized and have received a good deal of
theoretical attention [83, and references therein]. But not until recently has the cw Raman process been so isolated from the complicating effects of the single-photon transitions. Indeed, the experimental realization of the far-off-resonance cw Raman laser now allows for detailed comparison with the cw NDOPO and it is fascinating that such a precise correspondence resurfaces after a 35-year hiatus.

The predicted steady-state behaviors of the two systems are identical and have been experimentally verified. They exhibit pump clamping (power limiting) behavior above threshold, which was first identified theoretically for the NDOPO by Siegman [84]. Furthermore, the output modes of both systems exhibit square root dependences on the input pump power, and peak photon conversion efficiencies approaching 100% at four times threshold for single-ended cavities.

All of the population for both systems effectively remains in the ground state. And both systems exhibit phase insensitive amplification when only one output mode is observed. There is actually phase sensitivity hidden between the two output modes for the NDOPO case and between the Stokes and 1–2 coherence for the Raman case. This can be observed by expressing the field operators in terms of amplitude and phase operators (for example $a_s = |a_s| \exp(-i\phi_s)$). It is then straightforward to show from the steady-state solutions of Eq. (3.71) that the following phase relationship holds between the pump, the Stokes (signal) and the 1–2 coherence (idler)

$$\phi_p = \phi_s + \phi_{12} + \pi/2$$

This relation is consistent with the findings of Giordmaine [82] for both the Raman
and NDOPO cases. It is also consistent with other amplification processes for which the driving polarization is 90° out of phase with the amplified light.

Both systems also exhibit the attractive feature of frequency insensitive gain. In other words, the gain doesn’t change much if one uses a visible versus a near-infrared pump laser. This is afforded by the large detuning from any single-photon transitions in both the Raman and NDOPO cases and allows for large tuning ranges and spectral coverage of the emitted light.

The only formal difference between these systems lies in what has become the fundamental difference between parametric and stimulated processes [85]. That is, the NDOPO must cope with phase matching difficulties, while the cw Raman laser must deal with challenges associated with heat deposition. There are also differences on a more practical level that can cause their behaviors to deviate from Eqs. (3.70) - (3.72) and from one another. [36, 86]

It is also noted briefly here that there exist less direct connections between the cw Raman laser and the standard (based on population inversion) laser. For instance, when the atomic variables can be eliminated, one can associate the population inversion of the normal laser with the intracavity pump photon number of the NDOPO and Raman systems. Consequently, it is the population inversion that is clamped in the normal laser rather than the intracavity pump power [17, page 514]. Other connections also exist between these systems.

This entire discussion therefore begs the question: “Is the Raman system a laser
or a parametric amplifier?” As unsatisfying as it may be, the answer to this question lies entirely in semantics. Indeed, the word “laser” is a human construction and interestingly, there exist an astounding number of different definitions. Most definitions would probably consider this Raman system a laser, but there is an undeniable link between it and the NDOPO.

Alternative Forms of the Hamiltonian

Due to similarities between the Raman and NDOPO systems, it is natural to think that an alternative form of the Raman Hamiltonian, similar to that of the NDOPO, may be valid. Indeed, such an alternative Hamiltonian does exist and can significantly simplify the quantum mechanical treatment of the cw Raman system. For reference, the original system Hamiltonian from the second section of this chapter is given again here as [see Eqs. (3.2), (3.3), and (3.9)]

\[
H_{sys} = \hbar \omega_1 S_{11} + \hbar \omega_2 S_{22} + \hbar \omega_3 S_{33} + \hbar \omega_p a_p^\dagger a_p + \hbar \omega_s a_s^\dagger a_s \\
+ i\hbar (a_p^\dagger - a_p)(g_{p,13} S_{13} + g_{p,23} S_{23} + \text{H.c.}) \\
+ i\hbar (a_s^\dagger - a_s)(g_{s,13} S_{13} + g_{s,23} S_{23} + \text{H.c.}).
\]

(Original Form) (3.79)

Also for reference, the system Hamiltonian for the NDOPO is

\[
H_{sys} = \hbar \omega_p a_p^\dagger a_p + \hbar \omega_s a_s^\dagger a_s + \hbar \omega_i a_i^\dagger a_i \\
- \hbar \left(g_{OPO} a_p a_s^\dagger a_i^\dagger + \text{H.c.}\right).
\]

(NDOPO) (3.80)

One can obtain a simpler Hamiltonian for the Raman system simply by analogy
with that of the NDOPO. Alternatively, Gerry and Eberly [63] utilized the RWA to model two optical modes interacting with a single atom in a \(\Lambda\) configuration, with no decay paths. The optical modes were assumed to be far detuned from the single-photon atomic transitions, but exactly resonant with the two-photon transition. The researchers showed that when the upper level could be adiabatically removed, the single-photon Jaynes-Cummings model for atom-photon interactions [examples of which are shown in Eq. (3.79) without the RWA] could be simplified to a two-photon version. The effective interaction Hamiltonian that they generate is very similar to that of the NDOPO.

I have not assessed the validity of the Gerry and Eberly method. So instead, using the analogy between the far-off-resonance cw Raman system and the NDOPO that was established above, one can obtain the two-photon version of the Raman Hamiltonian

\[
H_{\text{sys}} = \hbar \omega_1 S_{11} + \hbar \omega_2 S_{22} + \hbar \omega_p a_p^\dagger a_p + \hbar \omega_s a_s^\dagger a_s
- \hbar \left( g^* a_p a_s^\dagger S_{12} + \text{H.c.} \right),
\]

(Alternate Form 1) \(3.81\)

where \(g\) is given by Eq. (3.73). One can quickly verify that applying the Heisenberg equation of motion to this Hamiltonian yields equations that are equivalent to Eqs. (3.38) - (3.40) and therefore averts a good deal of mathematical complexity that was carried out earlier in this chapter. To be clear, the system Hamiltonian given in Eq. (3.81) neglects all single-photon interactions, level 3 population, Stark shifts,
and power broadening. The decay and noise processes associated with level 3 are also implicitly ignored with this system Hamiltonian. It describes only the effects of the two-photon Raman process.

The Hamiltonian given in Eq. (3.81) isn’t precisely analogous to that of the NDOPO due to the presence of the level 2 population. However, when the rate of level 2 Raman excitation [see Eq. (3.49)] is small compared to the level 2 decay rate, one can additionally assume that that there is negligible population in level 2. This is the situation that exists for the Raman system that is currently under consideration. Interestingly, in this limit, the 1–2 coherence operator behaves suspiciously like a boson operator (note the commutation relation $[S_{12}, S_{12}^\dagger] = 1$ when $S_{22} \to 0$).

The coherence can then be treated in the same manner as a field operator (including reservoir interactions) so that the system Hamiltonian becomes

$$H_{\text{sys}} = \hbar \omega_p a_p^\dagger a_p + \hbar \omega_s a_s^\dagger a_s + \hbar \omega_{21} S_{12}^\dagger S_{12}$$

$$- \hbar \left( g^\ast a_p a_s^\dagger S_{12} + \text{H.c.} \right).$$

(Alternate Form 2) (3.82)

This form of the Raman system Hamiltonian is truly analogous to that of the NDOPO given by Eq. (3.80). It neglects the same processes as the first alternate form, but now none of the free energy of the system resides in the populations. Instead, all of it resides in the fields, as for a parametric process. Here, the coherence “field” operator between the lower two levels can be viewed as the annihilation operator of a molecular vibration or an optical phonon.
It now becomes clear why some authors have successfully applied Hamiltonians similar to Eq. (3.82) but with $S_{12}$ replaced by a phonon operator to Raman systems (see, for instance Ref. [54]). One can now accurately estimate the limits of validity for such an approximate Hamiltonian. Applying the Heisenberg equation of motion to this the second alternate Hamiltonian form yields Eqs. (3.70) - (3.72). As long as one does not need information about the level 2 population (which can be important for thermo-optic considerations), the system Hamiltonian given by Eq. (3.82) accurately describes the behavior of the cw Raman system when the appropriate reservoir interactions are included.

Output Intensity Noise Spectra

The output intensity noise spectra of the generated Stokes light is calculated in this section using the linearization procedure developed by Yamamoto [87] and others [88] and the input/output formalism developed by Collet and Gardiner [89]. The technique has proven to be a valuable tool for analyzing the spectra of parametric systems [81, 90, 91] as well as systems that explicitly involve atomic degrees of freedom [70, 58, 72].

First, the 1–2 coherence can be adiabatically eliminated from Eqs. (3.70) - (3.72) above as for the semiclassical case. However, now quantum noise from the 1–2 coherence finds its way into the remaining two field equations explicitly. Specifically, the
equations become

\[ \dot{a}_p = -\kappa_p a_p - G_1 a_s a_p^\dagger a_p + i\sqrt{2G_1} a_s S_{12} + \sqrt{2\kappa_p} a_p^{in}, \]  
\[ \dot{a}_s = -\kappa_s a_s + G_1 a_p a_p^\dagger a_s + i\sqrt{2G_1} a_p (S_{12}^{in})^\dagger + \sqrt{2\kappa_s} a_s^{in}, \]

where \( G_1 \) is given by Eq. (3.57). These serve as the starting equations for the following derivation of the Stokes laser output intensity noise. They are identical to the semiclassical equations of motion except that the noise contributions are now included.

The procedure is now described. A change of variables is first introduced to decouple the amplitude and phase portions of the field operators. Only the two amplitude equations are of interest for the intensity noise calculation given here. Each amplitude operator is then separated into two portions; the semiclassical mean value, and the small time-dependent fluctuations about this mean value. Such a distinction allows one to linearize the two amplitude equations for small fluctuations about the stable semiclassical mean amplitudes. This linearization dramatically simplifies the subsequent analysis by enabling the use of linear algebra. Accordingly, the equations for the amplitude fluctuations are recast in matrix form and are transformed to the Fourier domain in preparation for an eventual intensity spectra calculation. Using the cavity boundary conditions, the intracavity amplitude fluctuation equations are then transformed to equations for the amplitude fluctuations outside the cavity. From the these fluctuation equations, the intensity noise of the emitted light can be
determined.

Amplitude/Phase Decoupling

As will be shown later, direct detection of a laser's intensity noise only involves the field amplitude; not the phase. It may therefore seem natural to separate the field amplitudes in Eqs. (3.83) and (3.84) from the phases. One can perform the following variable transformation to amplitude and phase operators [87]

\[ a_q = \tilde{a}_q e^{-i\phi_q}, \quad \text{for } q = p, s \]  

(3.85)

and similarly for all the input operators, where

\[ \tilde{a}_q = \sqrt{a_q a_q^\dagger} \quad \text{and} \quad \phi_q = \frac{i}{2} \ln \left( \frac{a_q}{a_q^\dagger} \right). \]  

(3.86)

The transformation given above is only valid when the photon number is much larger than unity [87, and references therein]. This condition is easily satisfied for the system at hand. It should also be noted here that a field quadrature expansion yields identical results to this amplitude/phase expansion [81]. Inserting the transformations given by Eq. (3.85) into Eqs. (3.83) and (3.84) produces the following two amplitude equations:

\[ \frac{d}{dt} \tilde{a}_p = -\kappa_p \tilde{a}_p - G_1 \tilde{a}_s^2 \tilde{a}_p \]

\[ + i \sqrt{2G} \tilde{a}_s S_{12}^{in} e^{-i(\phi_s + \phi_{12} - \phi_p)} + \sqrt{2\kappa_p} \tilde{a}_p^{in}, \]  

(3.87)

\[ \frac{d}{dt} \tilde{a}_s = -\kappa_s \tilde{a}_s + G_1 \tilde{a}_p^2 \tilde{a}_s \]

\[ + i \sqrt{2G} \tilde{a}_p S_{12}^{in} e^{-i(\phi_p - \phi_{12} - \phi_s)} + \sqrt{2\kappa_s} \tilde{a}_s^{in}. \]  

(3.88)
Using the phase relationship between the fields given by Eq. (3.78), the amplitude operators completely decouple from the phase operators. This is quite expected for phase insensitive amplification processes like Raman lasing. Because only the amplitude equations are of interest for the intensity noise, this decoupling therefore conveniently reduces the number of pertinent equations from four ($a_p$, $a_p^\dagger$, $a_s$, and $a_s^\dagger$) to two ($\bar{a}_p$ and $\bar{a}_s$).

**Linearization**

The two remaining equations are still nonlinear, thereby prohibiting straightforward analytical solutions. To alleviate this difficulty, one can first make an explicit distinction between the semiclassical steady-state field amplitude, and the small amplitude fluctuations about this mean value as is often done (see, for instance Ref. [92, page 34]). Accordingly, the following field fluctuation *operator* is defined

$$\delta \tilde{a}_q(t) = 2 (\bar{a}_q(t) - |\alpha_q|), \quad (3.89)$$

so that

$$\bar{a}_q(t) = |\alpha_q| + \frac{1}{2} \delta \tilde{a}_q(t), \quad (3.90)$$

where the factor of two is included for later mathematical convenience. Now all the time dependence (and all the quantum nature) of each field amplitude is contained within the fluctuation operator $\delta \tilde{a}_q(t)$, while $|\alpha_q|$ represents the real-valued steady-state semiclassical field amplitude, which is independent of time. The input
fluctuation operators are represented in a similar manner. However, only the input pump operator will have a nonzero mean deterministic value. The other two input operators represent purely stochastic noise and fluctuate about zero mean values (it is assumed that there is no Stokes seed).

These newly-defined field and input fluctuation operators can be inserted into Eqs. (3.87) and (3.88). Doing this generates both the behavior of the semiclassical steady-state amplitudes as well as that of the amplitude fluctuations. For the steady-state behavior above threshold, one recovers the previous semiclassical results (Eqs. 3.58 and 3.59), which are written here as

\[ |\alpha_p| = \sqrt{\frac{\kappa_p}{G_1}}, \quad \text{and} \quad |\alpha_s| = \sqrt{\frac{s \kappa_p}{G_1}}, \]  
(3.91)

with

\[ s = \sqrt{r_p} - 1, \]  
(3.92)

where the pump rate \( r_p \) is defined in Eq. (3.60). And for the time-dependent amplitude fluctuation operators, one obtains the two linear equations

\[ \frac{d}{dt} \delta \tilde{a}_p = - \left( \kappa_p + G_1 |\alpha_s|^2 \right) \delta \tilde{a}_p - 2G|\alpha_p||\alpha_s| \delta \tilde{a}_s 
- \sqrt{2G} \delta \tilde{S}_{12}^{\text{in}} + \sqrt{2\kappa_p} \delta \tilde{a}^{\text{in}}_p, \]  
(3.93)

\[ \frac{d}{dt} \delta \tilde{a}_s = - \left( \kappa_s - G_1 |\alpha_p|^2 \right) \delta \tilde{a}_s + 2G|\alpha_p||\alpha_s| \delta \tilde{a}_p 
+ \sqrt{2G} \delta \tilde{S}_{12}^{\text{in}} + \sqrt{2\kappa_s} \delta \tilde{a}^{\text{in}}_s, \]  
(3.94)

where it is understood that all the fluctuation operators are functions of time. In obtaining these fluctuation equations it has been assumed that the steady-state field
amplitudes are very large compared to the associated fluctuations (\(|\alpha_q| \gg |\delta a_q(t)|\)) so that second order fluctuation terms can be neglected. In practice, this linear approximation is incredibly good.

The fact that the above amplitude fluctuation equations are now linear significantly simplifies subsequent analysis by enabling the use of linear algebra. Specifically, Eqs. (3.93) and (3.94) can be written in the compact form

\[
\frac{d}{dt} \delta a(t) = -\mathbf{A} \delta a(t) + \mathbf{B} \delta S^{\text{in}}(t) + \mathbf{C} \delta a^{\text{in}}(t),
\]

(3.95)

where the following vectors and matrices have been defined

\[
\delta a = \left( \begin{array}{c} \delta \bar{a}_p \\ \delta \bar{a}_s \end{array} \right), \quad \delta S^{\text{in}} = \delta \bar{S}^{\text{in}}_{12} \left( \begin{array}{c} 1 \\ 1 \end{array} \right), \quad \delta a^{\text{in}} = \left( \begin{array}{c} \delta \bar{a}_p^{\text{in}} \\ \delta \bar{a}_s^{\text{in}} \end{array} \right),
\]

(3.96)

\[
\mathbf{A} = \left( \begin{array}{cc} \kappa_p + G_1|\alpha_s|^2 & 2G_1|\alpha_p||\alpha_s| \\ -2G_1|\alpha_p||\alpha_s| & \kappa_s - G_1|\alpha_p|^2 \end{array} \right) = \left( \begin{array}{cc} (1 + s)\kappa_p & 2\sqrt{s} \kappa_p \kappa_s \\ -2\sqrt{s} \kappa_p \kappa_s & 0 \end{array} \right),
\]

(3.97)

\[
\mathbf{B} = \left( \begin{array}{cc} -\sqrt{2G_1}|\alpha_s| & 0 \\ 0 & \sqrt{2G_1}|\alpha_p| \end{array} \right) = \left( \begin{array}{cc} -\sqrt{2s} \kappa_p & 0 \\ 0 & \sqrt{2s} \kappa_p \end{array} \right),
\]

(3.98)

\[
\mathbf{C} = \text{diag} \left( \sqrt{2\kappa_p}, \sqrt{2\kappa_s} \right).
\]

(3.99)

Note that the relations given by Eqs. (3.91) and (3.92) have been used.

**Input/Output Formalism**

The previous analysis has generated a set of linear equations describing the amplitude fluctuations in time for the fields within the cavity. However, the noise spectra in the Fourier domain for the field fluctuations outside the cavity are sought. One can
therefore next take the Fourier transform of Eq. (3.95) and solve for the intracavity field fluctuations (in the frequency domain) to give

$$\delta \tilde{a}(\omega) = (i\omega + A)^{-1} \left[ B \delta \tilde{S}^{\text{in}}(\omega) + C \delta \tilde{a}^{\text{in}}(\omega) \right],$$  

(3.100)

where the fact has been used that the Fourier transform of the derivative of an operator can be expressed by multiplying the transform of the operator by $i\omega$. The operators in Fourier space are denoted with tilde's and are defined by

$$\delta \tilde{a}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} dt \delta a(t)e^{i\omega t},$$  

(3.101)

and similarly for the input operators.

One can now use the cavity boundary conditions [52]

$$\delta \tilde{a}^{\text{out}}(\omega) = C \delta \tilde{a}(\omega) - \delta \tilde{a}^{\text{in}}(\omega),$$  

(3.102)

to find the output amplitude fluctuations as functions of the input amplitude fluctuations. The vector $\delta a^{\text{out}}$ is defined analogously to its input counterpart. Inserting Eq. (3.100) into Eq. (3.102) yields

$$\delta \tilde{a}^{\text{out}}(\omega) = C (i\omega + A)^{-1} B \delta \tilde{S}^{\text{in}}(\omega) + [C (i\omega + A)^{-1} C - 1] \delta \tilde{a}^{\text{in}}(\omega),$$  

(3.103)

where $\mathbf{1}$ is the identity matrix. One can now relate these output amplitude fluctuations to the measured intensity noise of the laser.

**Intensity Noise**

One can define a (unitless) output intensity operator for the field through

$$I_q^{\text{out}} = a_q^{\text{out}} a_q^{\text{out} \dagger}.$$  

for $q = p, s$  

(3.104)
The expectation value of this operator gives the mean photon flux (photons/second) exiting the cavity when the photon number is much larger than one. Like the input operators, the output field operators are defined with units $\sqrt{Hz}$. Inserting the expression for the field operator given by Eq. (3.85), and using the fluctuation operator definition given by Eq. (3.90) this equation becomes

$$I_q^{\text{out}}(t) = \left[\bar{a}_q^{\text{out}}(t)^2\right]$$

$$= \left[|\alpha_q^{\text{out}}| + \frac{1}{2} \delta\bar{a}_q^{\text{out}}(t)\right]^2$$

$$\approx |\alpha_q^{\text{out}}|^2 + |\alpha_q^{\text{out}}| \delta\bar{a}_q^{\text{out}}(t), \quad (3.105)$$

where the second order fluctuations have again been neglected in the last step. This relation shows that direct detection of a laser's intensity noise is proportional to the fluctuations in the field amplitude, and has nothing to do with the field phase. One can now define the fluctuation operator for the intensity as

$$\delta I_q^{\text{out}}(t) = |\alpha_q^{\text{out}}| \delta\bar{a}_q^{\text{out}}(t), \quad (3.106)$$

In practice, the intensity noise spectra of the laser output is obtained using a power spectrum analyzer which measures the power spectral density as a function of rf
frequency. Mathematically, this measurement is described by

\[
S_q^{\text{out}}(\omega) = \int_{-\infty}^{\infty} \langle \delta I_q^{\text{out}}(t + \tau) \delta I_q^{\text{out}}(t) \rangle e^{-i\omega \tau} d\tau
\]

\[
= |\alpha_q^{\text{out}}|^2 \int_{-\infty}^{\infty} \langle \delta \tilde{a}_q^{\text{out}}(t + \tau) \delta \tilde{a}_q^{\text{out}}(t) \rangle e^{-i\omega \tau} d\tau
\]

\[
= |\alpha_q^{\text{out}}|^2 \langle \delta \tilde{a}_q^{\text{out}}(\omega) (\delta \tilde{a}_q^{\text{out}}(\omega))^\dagger \rangle
\]

\[
= |\alpha_q^{\text{out}}|^2 \langle |\delta \tilde{a}_q^{\text{out}}(\omega)|^2 \rangle, \tag{3.107}
\]

where the Wiener-Khintchin theorem has been used to get from the second to the third step [81]. This valuable result shows that the output intensity noise is simply related to the variance of the amplitude operator.

The noise spectra are often normalized to that of a coherent state light source of the same average power. This universal noise reference is called the standard quantum limit (SQL) and is often referred to as shot noise. Many sources of light can closely approximate a coherent state, making this an incredibly useful normalization from an experimental standpoint.

To normalize Eq. (3.107) to the SQL, it can be divided by a power spectral density of the same average intensity, but with the output fluctuation operators replaced by vacuum field fluctuation operators (i.e. a coherent state). As shown in Appendix J, the power spectral density for a coherent state is simply \(S_q^{\text{coh}}(\omega) = |\alpha_q^{\text{out}}|^2\). Using the symbol \(V_q^{\text{out}}(\omega)\) to represent the power spectral density normalized to the SQL, one obtains

\[
V_q^{\text{out}}(\omega) = \frac{S_q^{\text{out}}(\omega)}{S_q^{\text{coh}}(\omega)} = \langle |\delta \tilde{a}_q^{\text{out}}(\omega)|^2 \rangle. \tag{3.108}
\]
One can now use Eq. (3.109) to obtain the output intensity noise as a function of the input intensity noise. Specifically, the matrix defined by the vector product

\[
\textbf{V}^{\text{out}}(\omega) = \langle \delta \tilde{a}^{\text{out}}(\omega) \rangle^\dagger \delta \tilde{a}^{\text{out}}(\omega) \rangle
\]

exhibits the desired quantity in the bottom right diagonal element, \(V^{\text{out}}_{22}(\omega)\). It should be noted that a dagger superscript of a vector quantity represents the conjugate transpose of that vector. Using a computer to calculate the elements of \(V^{\text{out}}_{22}(\omega)\) analytically yields

\[
V^{\text{out}}_{s}(\omega) = V^{\text{out}}_{22}(\omega)
\]

\[
= \frac{4\kappa_2^2 \left[ \omega^2 + \kappa_2^2(s - 1)^2 \right]}{(\omega^2 - 4\kappa_2^2\kappa_0 s)^2 + \kappa_2^2(s + 1)^2 \omega^2} \times V^{\text{in}}_{\text{atoms}}(\omega)
\]

\[
+ \frac{16\kappa_2^2\kappa_0 s}{(\omega^2 - 4\kappa_2^2\kappa_0 s)^2 + \kappa_2^2(s + 1)^2 \omega^2} \times V^{\text{in}}_p(\omega)
\]

\[
\omega^2 + \left[ \kappa_2^2(1 + s)^2 + 4\kappa_2^2 - 8\kappa_2\kappa_0 s \right] \omega^2 + 4\kappa_2^2\kappa_0 s(s - 1)^2
\]

\[
(\omega^2 - 4\kappa_2^2\kappa_0 s)^2 + \kappa_2^2(s + 1)^2 \omega^2
\]

\[
\times V^{\text{in}}_{s}(\omega),
\]

(3.110)

where the input variances are defined as

\[
V^{\text{in}}_{\text{atoms}}(\omega) = \langle |\delta \tilde{S}_{12}^{\text{in}}(\omega)|^2 \rangle,
\]

(3.111)

\[
V^{\text{in}}_p(\omega) = \langle |\delta \tilde{a}^{\text{in}}_p(\omega)|^2 \rangle
\]

(3.112)

\[
V^{\text{in}}_{s}(\omega) = \langle |\delta \tilde{a}^{\text{in}}_s(\omega)|^2 \rangle
\]

(3.113)

The first term on the right side of Eq. (3.110) represents the noise contribution from
the atoms while the second and third terms are the contributions from pump and Stokes input fields, respectively.

Taking the input variances of the atoms and the Stokes field to be ordinary vacuum \( V_{\text{atoms}}^\text{in}(\omega) = V_s^\text{in}(\omega) = 1 \), but leaving the input pump variance as a variable parameter gives the following analytical result for the output Stokes intensity noise

\[
V_{s}^{\text{out}}(\omega) = \frac{\omega^4 + \left[\kappa_p^2(1+s)^2 + 8\kappa_s^2 - 8\kappa_p\kappa_s s\right]\omega^2 + 8\kappa_p^2\kappa_s^2 [(s - 1)^2 + 2sV_p^\text{in}(\omega)]}{(\omega^2 - 4\kappa_p\kappa_s s)^2 + \kappa_p^2(s + 1)^2}\omega^2
\]

(3.114)

This analytical solution for the Stokes laser intensity noise spectra is the primary result of this section. An analogous result for the emitted pump light intensity noise can be calculated in the same manner. Furthermore, using the formalism of this section, one can also calculate the intensity correlations between the output pump and Stokes. A thorough analysis of the Stokes intensity noise is given in the next subsection.

**Stokes Intensity Noise Analysis**

Although it may seem difficult to extract much insight from Eq. (3.114), several limiting cases can shed some light on the underlying physics involved. In particular, one can analyze the Stokes intensity noise behavior as a function of pump rate for very low and very high Fourier frequencies. Figure 16 illustrates the pump rate behavior of the intensity noise for Fourier frequencies below the cavity linewidths \( \omega \ll \kappa_p, \kappa_s \) and for several different input pump noise levels. A coherent state input pump noise...
corresponds to $V_p^{in}(\omega) = 1$. The behavior is plotted on both linear [Fig. 16(a)] and a log-log [Fig. 16(b)] scales. Also, Eq. (3.114) becomes

$$V_{s}^{\text{out}}(\omega) = \frac{[(s - 1)^2 + 2sV_p^{\text{in}}(\omega)]}{2s^2} \quad (\omega \ll \kappa_p, \kappa_s)$$

(3.115)

in the limit of low Fourier frequency. From the figures and from Eq. (3.115), first note that the Stokes intensity noise is well above the SQL near threshold ($s \approx 0, \gamma_p \approx 1$) for all curves. This system is therefore inherently noisy near threshold, a trait that is shared with other laser systems. However, it is very important to note that the output Stokes intensity noise is equal to the input, pump noise [$V_{s}^{\text{out}}(\omega) = V_p^{\text{in}}(\omega)$] at four times threshold ($s = 1, \gamma_p = 4$) for all curves. Vertical lines at $\gamma_p = 4$ have been provided on both plots to emphasize this point. This feature can be confirmed using Eq. (3.115) with $s = 1$. The system therefore displays perfect photon statistics transfer from the input pump to the output Stokes for a pump rate of four times threshold and frequencies below the cavity linewidths. This is also consistent with the steady-state results of Chapter 2 indicating a photon conversion efficiency of 100% occurring at $\gamma_p = 4$. 
Figure 16. (a) Linear and (b) log-log plots of the Stokes intensity noise relative to the SQL as a function of pump rate for frequencies well below the cavity linewidths. Curves for several different values of the input pump intensity noise $[V_p^{in}(\omega)]=1$ are shown. The system exhibits perfect photon statistics transfer from pump to Stokes at four times threshold and the intensity noise approaches 50% below the SQL in the limit of large pump rate.

It is also interesting to note that the output intensity noise can drop below the shot noise level $[V_s^{out}(\omega)=1]$ for higher pump rates and asymptotically approaches $V_s^{out}(\omega)=1/2$ in the limit of large pump rate ($r_p \gg 1$) even when the input pump noise is greater than vacuum. This feature is observed most easily in the log-log plot and can also be confirmed by simply taking the limit $s \to \infty$ in Eq. (3.115). This system is therefore predicted to approach 50% intensity noise reduction below the SQL for frequencies within both of the cavity bandwidths and for high pump rates.

Up to now, only the low frequency limit of Eq. (3.114) has been considered. On the other hand, for frequencies much larger than the cavity bandwidths, the intensity noise approaches the SQL for all pump rates and for all input pump noise levels. This simply expresses the fact that the cavity acts as a low-pass filter and that the only
contribution to the output Stokes intensity noise far above the cavity bandwidths is the reflected vacuum noise. This can be confirmed by taking the limit $\omega \to \infty$ in Eq. (3.114). Using Eq. (3.110), it is also possible to determine the particular contributions to the total intensity noise from each of the three noise sources (input pump noise, input Stokes noise, and input coherence noise).

Although the low and high frequency limits are simpler from a mathematical standpoint, much of the interesting behavior can occur for frequencies between the extremely low and extremely high limits. This behavior depends critically on the ratio of the two cavity decay rates. Figures 17 and 18 help to illustrate the issues involved.

Figure 17(a) shows the Stokes intensity noise as a function of Fourier frequency for a pump rate of four times threshold ($s = 1$, $r_p = 4$) and an input pump noise 10 times greater than shot noise [$V_p^{\text{in}}(\omega) = 10$]. The Fourier frequency on the horizontal axis is given relative to $\sqrt{\kappa_p \kappa_s}$ so that the curves can be compared on even ground. In the figure several curves are given corresponding to several different values of the cavity decay rate ratio ($\kappa_s / \kappa_p$). As this ratio increases, relaxation oscillations become undamped as evidenced by the increasing noise peak. In general, decreasing the cavity decay rate ratio (i.e. making the Stokes cavity finesse greater than that of the pump) suppresses relaxation oscillations. This is consistent with previous findings in the time domain [69].
Figure 17. Stokes intensity noise relative to the SQL as a function of normalized Fourier frequency for several different ratios of the cavity decay rates \((\kappa_s / \kappa_p)\) and for a pump rate of (a) 4 times threshold and (b) 25 times threshold. The product \(\kappa_s \times \kappa_p\) is held constant for all curves so that they correspond to the same threshold. Decreasing the cavity decay rate of the Stokes relative to that of the pump suppresses relaxation oscillations. Increasing the pump rate from (a) \(r_p = 4\) to (b) \(r_p = 25\) in produces broadband suppression of the intensity noise and pushes the relaxation oscillations to slightly higher frequencies.

Figure 17(b) shows the same thing, but for a pump rate 25 times threshold \((r_p = 25)\). One can first note that the relaxation oscillations are pushed to slightly higher frequencies as the pump rate is increased from \(r_p = 4\) in Fig. 17(a) to \(r_p = 25\) in Fig. 17(b). Also evident is the fact that the noise levels, including the relaxation peaks diminish when the pump rate is increased [from Fig. 17(a) to Fig. 17(b)].

The pump rate behavior is more clearly demonstrated in Fig 18. Fig 18(a) shows the normalized intensity noise as a function of Fourier frequency for the symmetric case when \(\kappa_p = \kappa_s\) and for an input pump noise of 10 times shot noise. Several curves are provided corresponding to different pump rates. Indeed the relaxation
oscillations are both diminished and pushed to higher frequencies as the pump rate is increased. Also note that $V_{s,\text{out}}(\omega) = V_{p,\text{in}}^{\text{out}}(\omega)$ for the four-times-threshold curve at low frequencies. Fig 18(b) shows the same thing, but for a coherent state input. As a general rule, increasing the pump rate yields broadband noise suppression.

Figure 18. Stokes intensity noise relative to the SQL as a function of normalized Fourier frequency for several different pump rates, equal cavity decay rates ($\kappa_s = \kappa_p$) and for an input pump noise of (a) $V_{p,\text{in}}^{\text{in}}(\omega) = 10$ and (b) $V_{p,\text{in}}^{\text{in}}(\omega) = 1$ (coherent state). Again, the product $\kappa_s \times \kappa_p$ is held constant for all curves. The relaxation oscillations are diminished and pushed to higher frequencies as the pump rate is increased.

From Fig. 18(b) one can again observe squeezing behavior for frequencies below the cavity bandwidth. The spectral region over which intensity squeezing is observed is maximized when the two cavity linewidths are equal and as large as possible. Unfortunately, the desire for large cavity bandwidths directly competes with the desire for large pump rates because an increase in the cavity bandwidths necessarily results in an increase in the threshold. This is a common dilemma found in many other
nonlinear optical systems and poses the most significant obstacle to experimentally generating intensity noise levels below the SQL for this system.

To summarize the findings of this section, this ideal cw Raman system is predicted to exhibit perfect photon transfer from the input pump to the output Stokes for a pump rate of four times threshold and below the cavity linewidths. Also, the system is predicted to exhibit 50% noise reduction below the SQL for very high pump rates and very low Fourier frequencies. The region of Fourier frequencies over which such squeezing can be observed is largest when the cavity decay rates are equal and as large as possible.

It was also shown that operation at very high pump rates and with very small decay rate ratios is desirable from a classical intensity noise standpoint. In other words, the cavity finesse for the Stokes should be greater than that of the pump and the threshold should be as low as possible. This was also the conclusion towards the end of Chapter 2. As discussed in Chapter 4, intensity fluctuations have further reaching ramifications because they couple to Stokes laser frequency fluctuations (mode pulling) through a thermo-optic nonlinearity of the Raman gas.

Chapter Summary

This rather involved chapter was devoted to developing the quantum theory of the far-off-resonance cw Raman laser using the Heisenberg/Langevin approach. The large single-photon detunings present in the system initially complicated matters by
disallowing the use of the RWA, but later provided the means for significant simplifications. The only effect of not invoking the RWA was to scale the Raman gain; it did not affect the qualitative behavior of the system. Equations (3.83) and (3.84) were identified as the quantum mechanical cw Raman laser equations. The semiclassical limit of these equations were shown to be consistent with the classical results obtained in Chapter 2. A strong connection between this cw Raman system and the cw NDOPO was also revealed. Critical to this connection was the fact that all of the population effectively remains in the ground state. Appendix G was devoted to a streamlined treatment of enhanced anti-Stokes generation, and showed that conversion efficiencies from the pump to the anti-Stokes can approach 50% in the limit of small mirror absorption and perfect phase matching.

To analyze the intensity noise of the emitted light, an amplitude/phase expansion was performed on the field operators. This decoupled the phase and amplitude behaviors as expected for this phase-independent amplification process and allowed the resulting two amplitude equations to be linearized about stable mean values. Using the input/output formalism developed by Collet and Gardiner, the intensity noise of the Stokes output was then calculated analytically as a function of Fourier frequency [see Eqs. (3.114) and (3.115)]. It was shown that perfect photon statistics transfer from the pump to the Stokes occurs at a pump rate of four times the threshold value and low Fourier frequencies. For higher pump rates, the system can exhibit up to 50% intensity squeezing within the cavity bandwidths. In order to suppress relaxation
oscillations in the system, the most favorable operational conditions were shown to be a high pump rate \( (r_p \gg 1) \) and a low ratio of cavity decay rates \( (\kappa_s \ll \kappa_p) \). For the sake of simplicity, cavity mirror absorptions were neglected, but can be included in a straightforward manner.

The effects of heat deposition in the Raman gas were also neglected in this chapter. Such effects can couple the amplitude and frequency behaviors of the system and can therefore cause frequency locking difficulties. The heating also substantially alters the steady-state behavior of the cw Raman system. These thermo-optic effects are the subject of the next chapter.
CHAPTER 4

THERMO-OPTIC THEORY

This chapter is devoted to theoretically describing the effects of thermally-induced nonlinearities observed in the refractive index of the Raman medium. The mode pulling work was conducted with Jay Brasseur just before he graduated, while the thermal lensing was the result of a wonderful long-distance interaction with Josh Bienfang who was at the University of New Mexico. To some degree, the procedures covered in this chapter can be extended to other contributions to the nonlinear refractive index as well (two-photon dispersion, gain guiding, etc.). However, as with other nonlinear optical systems, the thermal contribution is by far the largest. The primary measurable thermo-optic effects in the cw Raman system are intensity dependent mode pulling and lensing within the Raman laser cavity. The first of these couples the laser’s amplitude behavior to its frequency behavior, and is therefore a serious concern for frequency locking and absolute frequency stability. The second effect is the source of the most significant discrepancies between the steady-state theories previously presented and the experimental results obtained to date.

The refractive index can also be affected by the two-photon dispersion associated with the Raman process, and single-photon dispersion associated with the weak single-photon transitions. Both of these effects are small and the mode pulling that results
from each was explicitly neglected in Chapter 3. Other sources of nonlinear refractive
index, such as electrostriction, are discussed in the self-focusing chapter of Shen's
book *Nonlinear Optics* [93, Chapter 17]. Boyd provides a nice discussion and gives
an order-of-magnitude comparison of several contributions to the intensity-dependent
refractive index in his book of the same title [85, Chapter 4].

The heating process in the Raman system occurs as follows: An atomic excitation
to level 2 (see Fig. 13) inherently accompanies the generation of each Stokes photon
that is produced via the Raman process. For the case at hand, this excitation is a
molecular vibration. Collectively, the molecules are excited with a spatial distribution
that is related to that of the pump and Stokes optical modes in the cavity. After
randomly diffusing for a bit, these molecules decay (nonradiatively) to the ground
state. This means that the energy stored as molecular vibrations is converted to
translational energy of the molecules, causing a rise in the temperature of the gas that
is greatest near the optical beam axis. This temperature increase causes a decrease
in the density of the gas and therefore a decrease in its refractive index. This can
change the effective optical length of the cavity (mode pulling) and it can act as a
diverging lens within the cavity, similar to a gradient index (GRIN) lens [17, pages
589-590]. The theoretical treatment given here is entirely steady state.

This chapter begins with a discussion of the previous pertinent treatments of
thermo-optic effects in lasers. In the second section, the refractive index of the heated
gas is derived as a function of radial distance from the beam axis. In the third
section, the refractive index along the axis of the optical beam is calculated and used to estimate the magnitude of the resulting mode pulling. In the fourth section, the stable cavity eigenmode is calculated in the presence of the thermal lens. The manner in which this intensity-dependent lens affects the steady-state behavior of the Raman laser is analyzed in the final section.

**Previous Work**

Considerable attention has been focused on deleterious thermal effects in lasers. This is primarily because the laser process is inherently inelastic (i.e. it leaves energy in the medium). To my knowledge, the first evidence and treatment of thermal effects in lasers was given by Gordon, et al. [94]. That work analyzed the transient behavior that occurred when absorbing liquid samples were placed within the cavity of a He-Ne laser. The work also derived the steady-state temperature profile within the sample using the diffusion equation.

Subsequently, researchers used this foundation to study the resulting thermal lensing [95, 96, 97, 98, 99] and beam distortion [100, 101, 102] in various materials. In the Raman laser, the on-axis frequency pulling is of equal importance. This is because the pump laser must be phase/frequency stabilized to the Raman laser cavity in order to build optical power. Therefore, any instability in the cavity's optical length (caused by heating, for instance) is a serious concern for locking and ultimately the laser's absolute frequency stability. The first qualitative experimental evidence
of such frequency instability was reported with the first demonstration of both the Nd:YAG-pumped Raman laser and the diode-pumped Raman laser above the Stokes laser threshold. It was later shown that these instabilities were indeed caused by thermo-optic mode pulling of the laser cavity resonances. Researchers addressed these concerns theoretically and experimentally for the cw Raman systems [29, 103]. The amount of mode pulling was quantified and it was shown that increased locking bandwidth is required to achieve stable frequency locking of the pump laser above the Stokes laser threshold.

Unfortunately, the thermal mode pulling was not the only effect resulting from the thermo-optic nonlinearity. The initial low-power Raman laser systems displayed striking agreement with the basic theoretical models (see Chapter 2). However, when the Stokes powers exceeded ~10mW, it became evident that the basic cw Raman laser equations were insufficient. Josh Bienfang was the first to identify the thermal effects as a likely cause of the observed discrepancies between the measured data and the theoretical predictions. These effects were then thoroughly studied both theoretically and experimentally in Ref. [36]. It was found that the generated Stokes power increased the confocal parameter of the stable cavity modes in the laser cavity. This consequently decreased the laser gain and altered the coupling efficiency of the incident pump laser into the Raman laser cavity mode. As a result, the transmitted pump power "unclamped" above threshold (i.e. the transmitted power became dependent on the incident pump power), and the emitted Stokes power departed from
a purely square-root dependence on the incident pump power. This chapter analyzes the steady-state mode pulling and thermal lensing in the bow-tie Raman laser system. The procedure can easily be applied to the linear cavity geometry as well.

**Thermal Refractive Index Profile**

In calculating the refractive index as a function of position within the laser cavity, one can start with the spatial distribution over which the molecular excitations are created (i.e. the spatial profile of the level 2 population). The heat deposition spatial profile is then obtained by allowing these molecular excitations to diffuse by random walk during the excited-state decay time. The diffusion equation can then be used to find the resulting temperature distribution within the cavity, from which the refractive index profile follows directly.

**Heat Deposition Profile**

Because the level 2 population depends on the product of the pump and Stokes optical powers [see Eq. (3.64) and note that $T^r$ contains the product of the two Rabi frequencies squared], its distribution is given by the Gaussian profile

$$
|\tilde{u}_p(r)^{(sp)}|^2 |\tilde{u}_s(r)^{(sp)}|^2 = \frac{1}{[1 + (2\pi/b)^2]^2} \exp \left[ -\frac{2\pi^2}{1 + (2\pi/b)^2} \left( \frac{1}{\omega_{0,p}} + \frac{1}{\omega_{0,s}} \right) \right] \quad (4.1)
$$

$$
\approx \exp \left( -\frac{2\pi^2}{\omega_{0,\text{int}}^2} \right) \quad (b \gg L) \quad (4.2)
$$

for a single pass in the bow-tie cavity, where the $\tilde{u}_q(r)$ are the fundamental field modes of the cavity at the pump ($q = p$) and Stokes ($q = s$) wavelengths, the $\omega_{0,q}$
are the field minimum waists (i.e. the 1/e amplitude points), and \( b \) is the confocal parameter. To simplify the thermo-optic calculation, it is assumed that the confocal parameter is much greater than the cavity length, which results in the approximate equality given by Eq. (4.2). This amounts to neglecting longitudinal variations of the field in the cavity, which is a good approximation when \( b > L \). The effective field waist of the molecular excitation profile is given by the relation

\[
\frac{1}{\omega_{0,\text{int}}^2} = \frac{1}{\omega_{0,p}^2} + \frac{1}{\omega_{0,s}^2}.
\]  

(4.3)

The subscript "int" is used because this waist also (not coincidentally) corresponds to that of the interaction volume discussed in Chapter 3. Optical power is exchanged from the pump to the Stokes over a volume characterized by the waist \( \omega_{0,\text{int}}/\sqrt{2} \). It is therefore natural that this should also be the waist that describes the generated level 2 population profile.

If the Raman medium were solid, then Eq. (4.3) would also describe the waist for the heat deposition. However, for gaseous Raman media, the molecules diffuse between the time of their excitation and the time of their decay to the ground state. As shown in Appendix K, the thermal waist, which accounts for this diffusion, is given by

\[
\omega_{0,\text{th}}^2 = \omega_{0,\text{int}}^2 \left( 1 + \frac{8D\tau_{\text{vib}}}{\omega_{0,\text{int}}^2} \right),
\]  

(4.4)

where \( D \) is the molecular diffusion constant and \( \tau_{\text{vib}} \) is the decay time of the vibrational state (level 2). Diffusion typically adds ~10-20% to the waist for hydrogen gas at
\( \sim 10 \text{ atmospheres.} \)

To clarify the various waists that have been discussed, Fig. 19 shows the radial optical power and population profiles that are involved with this system. Each of the profiles is a Gaussian centered about the beam axis. The relative waists of the profiles can be qualitatively ascertained from the plot. The population profile is just the product of the pump power and Stokes power profiles and gives the distribution of excited molecules within the gas before diffusion. The thermal profile is slightly broadened compared to the population profile due to diffusion of the molecules.

![Figure 19. Plot showing the the radial pump power, Stokes power, population, and](image-url)
thermal profiles within the cavity for comparison of the relative waists. All of the Gaussians are normalized to a peak of unity.

Now that the distribution of excited and diffused molecules is known, the profile of the heat deposition (thermal power density) can be written as

\[ Q(r) = Q_0 \exp \left( -\frac{2r^2}{\omega_{0,th}^2} \right) \]  

(4.5)

in analogy with Eq. (4.2), where \( Q_0 \) is the (as yet undetermined) thermal power density (in W/m²) on axis.

**On-Axis Thermal Power Density**

The on-axis thermal power density can be determined by the requirement that the volume integration of Eq. (4.5) times four (for the four passes present in the bow-tie cavity) yield the total energy deposited in the gas per unit time (i.e. the thermal power generated). Mathematically, this is represented by

\[ 4 \times \int \int \int_{\text{single pass}} Q(r) \, dr = \frac{\nu_{21}}{\nu_s} P_{\text{gen}}, \]  

(4.6)

where \( P_{\text{gen}} \) is the total Stokes optical power generated. Note that the generated thermal power is just the total Stokes power generated multiplied by the frequency ratio \( \nu_{21}/\nu_s \), where \( \nu_{21} \) is the frequency separation between the ground and first excited vibrational states. This is the "leftover" energy from the Raman transitions.

Performing the integrals in Eq. (4.6), using Eq. (4.5), and solving for \( Q_0 \) yields

\[ Q_0 = \frac{P_{\text{gen}}}{2\pi L \omega_{0,th}^2} \frac{\nu_{21}}{\nu_s}, \]  

(4.7)
where $L$ is the single-pass length. For later convenience, one can express $P_{\text{gen}}$ in terms of the intracavity powers and also in terms of the output Stokes power. This can be accomplished by integrating the gain term of Eq. (2.4) over the round-trip time ($\tau_{\text{rt}} = p/c$). It is also given by the steady-state Stokes output power when the mirror absorptions are set to zero. In either case,

$$P_{\text{gen}} = \frac{P_p P_s}{P_1}, \quad (4.8)$$

or

$$P_{\text{gen}} = \frac{1 - R_s,\tau_{\text{rt}}}{T_s,\text{tot}} P_{\text{out}}, \quad (4.9)$$

where $P_p, P_s,$ and $P_1$ are the intracavity pump, Stokes, and unity gain powers defined by Eqs. (2.15), (2.16) and (2.10), and $P_{\text{out}}$ is the total output Stokes power given by Eq. (2.22) from Chapter 2. Both forms of $P_{\text{gen}}$ are useful.

Near-Axis Refractive Index Profile

With $Q_0$ known, one can now follow the work of Gordon, et al. [94] to find the resulting refractive index profile in the gas. The procedure entails using $Q(r)$ from Eq. (4.5) as the heat source and solving the diffusion equation to find the temperature distribution within the laser cavity. The refractive index profile is then readily determined from this temperature profile. The details of this derivation are given in Appendix L. From that appendix, the refractive index profile due to heat
deposition is given by

\[
\Delta n(r) = n(r) - n_0 = -\frac{Q_0 \omega_{0,th}^2 (n_0 - 1)}{8 K_{th} T_0} \left[ \text{Ei} \left( -\frac{2r^2}{\omega_{0,th}^2} \right) - \text{Ei} \left( -\frac{2r_c^2}{\omega_{0,th}^2} \right) - \ln \frac{r^2}{r_c^2} \right], \tag{4.10}
\]

where \( K_{th} \) is the thermal conductivity, \( T_0 \) and \( n_0 \) are the temperature and refractive index in the absence of optical power\(^1\), \( \text{Ei}(x) \) is the exponential integral function [49, page 504], and \( r_c \) is the radial distance to a constant-temperature surface. In some laser systems this constant-temperature surface is well defined because the outer surface of the laser rod is actively cooled or temperature controlled. In the present system, this parameter is somewhat arbitrary because the Raman laser cavity is not actively temperature controlled. Fortunately, the results of this chapter do not depend drastically on this parameter, and it is usually safe to assume that its value is close to the inner radius of the cylindrical surface that contains the Raman gas.

In any case, this radius is much larger than the beam spot size for all the systems that have been studied to date. When \( r_c \gg \omega_{0,th} \), the second term in the square brackets in Eq. (4.10) can be neglected because \( \text{Ei}(-x) \to 0 \) for \( x \gg 1 \). Furthermore, the first term in the square brackets can be expanded using the following series expansion for the exponential integral function

\[
\text{Ei}(-x) = \gamma_E + \ln x + \sum_{n=1}^{\infty} \frac{(-x)^n}{n \cdot n!}, \quad (x > 0) \tag{4.11}
\]

---

\(^1\) \( T_0 \) used in this chapter should not be confused with the input coupler transmissivities, \( T_{p,0} \) and \( T_{s,0} \)
where $\gamma_E = 0.5772 \ldots$ is the Euler constant. Near the beam axis, Eq. (4.10) can therefore be simplified to

$$\Delta n(r) = n(r) - n_0 \approx -\frac{Q_0 \omega_{0,th}^2 (n_0 - 1)}{8K_{th}} \left[ \gamma_E + \ln \left( \frac{2r_c^2}{\omega_{0,th}^2} \right) - \frac{2r_c^2}{\omega_{0,th}^2} \right]. \quad (r \ll \omega_{0,th}) \quad (4.12)$$

This equation is one of the primary results of this chapter. Figure 20 compares the exact solution given by Eq. (4.10) to the solutions when the infinite sum given in Eq. (4.11) is truncated after the first (up to $r^2$) and after the fifth (up to $r^{10}$) terms. The curves are plotted as functions of the radial distance from the beam axis, relative to the pump amplitude waist ($\omega_{0,p}$). The zeroth order term (no radial dependence) gives the on-axis change in refractive index, which will be used to calculate the thermal mode pulling. The second order term will be used to calculate the thermal lensing using the ABCD matrix formalism. Because the refractive index increases with radial distance, this lens is diverging.
As pointed out by Bienfang, the second order approximation does not fit the exact behavior particularly well out to the pump beam waist [36]. As a result, the authors of Ref. [36] perform a best fit of a quadratic curve to the exact profile to obtain a broadened thermal waist. This reduces the strength of the lens a bit and gives better agreement with experimental data. The figure shows that terms up to the \( \sim 10 \)th order term must be retained to accurately approximate the exact function out to the pump field waist. The modified version of Eq. (4.12) with the broadened
waist is given by

\[ \Delta n(r) = -\frac{Q_0 \omega_0^{2,th} (n_0 - 1)}{8K_{th}} \left[ \gamma E + \ln \left( \frac{2r_c^2}{\omega_0^2} \right) - (0.518) \frac{2r_c^2}{\omega_0^2} \right] \]  \hspace{1cm} (4.13)

Terms higher than second order represent spherical aberrations caused by the (non-quadratic) thermal lens. Such spherical aberrations will cause the eigenmodes of the Raman cavity to deviate from purely Hermite Gaussian. The higher order terms will therefore cause coupling losses into the cavity and beam distortion when the amount of generated Stokes power is very large. These spherical aberrations are not considered here because their effects have not yet been observed experimentally. Instead, attention will now be focused on the mode pulling and thermal lensing in the next two sections.

**Thermal Mode Pulling**

The zeroth order contribution to Eq. (4.13) gives the on-axis change in the refractive index that is due to Raman-generated heat deposition. The index decrease causes the optical length of the cavity to decrease slightly, and therefore causes a blue-shift of the cavity resonance frequencies. One can solve for the frequency pulling by using the fundamental relationship between optical frequency and wavelength. Specifically, one can derive the relation

\[ \Delta \nu_p \approx -\nu_p \Delta n^{(0)} \]  \hspace{1cm} (4.14)

for the frequency pulling of the pump cavity mode, where \( \Delta n^{(0)} \) is the refractive index...
change given by Eq. (4.13) when \( r = 0 \). This gives the following expression for the pump mode pulling for the four-pass ring cavity:

\[
\Delta \nu_p = P_{\text{gen}} \frac{\nu_p \nu_{21}}{\nu_s} \frac{(n_0 - 1)}{16\pi L K_{th} T_0} \left[ \gamma_E + \ln \left( \frac{2\pi c^2}{\omega_{0,th}^2} \right) \right], \tag{4.15}
\]

where Eq. (4.7) was used for \( Q_0 \), and Eq. (4.8) or Eq. (4.9) can be used to cast \( P_{\text{gen}} \) in terms of the intracavity powers or the output Stokes power, respectively. This equation is very useful in estimating the frequency pulling, but it should be noted that \( \omega_{0,th} \) actually depends on the generated Stokes power as will soon be shown. Therefore, the frequency pulling is not strictly linear with the generated Stokes power. Nevertheless, a good estimate of the mode pulling is obtained by assuming \( \omega_{0,th} \) is independent of optical power (use the cold-cavity value); especially for low powers (typically <10mW Stokes power). Alternatively, it will be shown that one can numerically solve for \( \omega_{0,th} \) in terms of \( P_{\text{gen}} \) to obtain a more precise value of the mode pulling for a given amount of generated Stokes power.

The frequency pulling predicted by Eq. (4.15) is typically very significant (tens of MHz per mW of generated Stokes power) compared to the typical cavity linewidth of \( \lesssim 1 \)MHz. The positive value of the pulling indicates that it is a blue-shift (increasing frequency with Stokes power). The result given in Eq. (4.15) can be multiplied by the factor 4 for the linear cavity.
Practical Considerations

The easiest way to observe this mode pulling behavior is by detecting the transmitted pump (or Stokes) light while scanning the length of the cavity without locking. This only works when the laser's inherent linewidth is much narrower than the cavity resonance. When the scan rate is slow enough and when the injected pump power exceeds the laser threshold, one observes hysteresis between the forward and backward scans and oscillations in the transmitted power for forward scans. These effects are shown and discussed in depth in Chapter 6.

A time-dependent treatment of the heating, coupled to the Raman laser equations is necessary to quantitatively describe these effects. Such a treatment is not included in this thesis. However, related effects were covered by Brasseur, et al. [103]. To summarize that work, the generation of Stokes light was shown to degrade the discriminator slope for locking and also establish a minimum feedback loop bandwidth due to thermally-induced cavity length instabilities. One may therefore need to consider the thermal mode pulling in the design of the locking servo and the frequency actuators from the standpoint of bandwidth, slew rate, and dynamic range.

Thermal Lensing

The on-axis (zeroth order) index change was just used to calculate the mode pulling effect. To calculate the effect of the thermal lens, one can use the second order radial dependence of the refractive index profile within the ABCD matrix formalism.
and calculate the beam parameters of the modified stable cavity mode. This lens affects the Stokes production, which in turn affects the lens. An iterative procedure is therefore required to calculate the stable lens and Stokes power that result. This section treats the thermal lens effect from start to finish.

Cavity with Quadratic Duct

The second order radial contribution to Eq. (4.13) causes thermal lensing within the laser cavity. To quantify this, one can first recast the radially-dependent refractive index into the notation of Siegman in the following manner

\[ n(r) = n_0 + \Delta n(r) \quad \text{(current notation)} \]
\[ = n_1 + \frac{1}{2} n_2 r^2, \quad \text{(Siegman's notation)} \]  

(4.16)

where \( n_0 \) is the refractive index of the gas in the absence of optical power and \( \Delta n(r) \) comes from Eq. (4.13). Using this equation, the new variables \( n_1 \) and \( n_2 \) can be identified as

\[ n_1 \equiv n_0 + \Delta n^{(0)} \quad \text{and} \quad n_2 \equiv \frac{Q_0 (n_0 - 1)(0.518)}{2K_{th}T_0}, \]  

(4.17)

where \( \Delta n^{(0)} \) is \( \Delta n(r) \) from Eq. (4.13) when \( r = 0 \). In this form, the effect of the quadratic index dependence can be analyzed using the ABCD matrix formalism given by Siegman [17, Chapter 15]. Most authors refer to a second order radial refractive index dependence as a “quadratic duct”. Following Siegman’s treatment, one can
obtain the ABCD matrix

\[
M_{\text{duct}} = \begin{bmatrix}
\cosh \sqrt{n_2} z & n_2^{-1/2} \sinh \sqrt{n_2} z \\
\frac{1}{n_2} \sinh \sqrt{n_2} z & \cosh \sqrt{n_2} z
\end{bmatrix}
\]  

(4.18)

for propagation through an unstable (defocusing) quadratic duct, where \( z \) is the distance of propagation, the fact has been used that \( n_0 \approx 1 \), and it should be noted that there is an important sign error in Eq. (18) of Chapter 15 in Siegman's first edition of *Lasers*.

The full cavity matrix is composed of four passes (assumed identical) through the duct and four end-mirror reflections. However, when the cavity is symmetric, one only needs to consider one full pass and one reflection. Choosing the reference plane to be the center of one pass, the effective ABCD matrix is given by: (1) duct propagation of distance \( L/2 \), (2) reflection from a mirror of curvature \( R \), and (3) another duct propagation of distance \( L/2 \). After some matrix algebra, one can show that the resulting effective matrix is given by

\[
M = \begin{bmatrix}
A & B \\
C & D
\end{bmatrix}
\]  

(4.19)

where

\[
A = D = \cosh (2\xi) - \frac{2}{R \sqrt{n_2}} \cosh (\xi) \sinh (\xi)
\]  

(4.20)

\[
B = \frac{2}{\sqrt{n_2}} \cosh (\xi) \sinh (\xi) - \frac{2}{R n_2} \sinh^2 (\xi)
\]  

(4.21)

\[
C = 2 \sqrt{n_2} \cosh (\xi) \sinh (\xi) - \frac{2}{R} \cosh^2 (\xi)
\]  

(4.22)
and

\[ \xi \equiv \frac{L\sqrt{n_2}}{2}. \quad (4.23) \]

One can obtain all of the beam parameters for the stable cavity mode from these matrix elements. In particular, the confocal parameter is given by [17, page 820]

\[ b = \frac{2\pi\omega_0^2}{\lambda_q} \]

\[ = 2|B|\sqrt{\frac{1}{1-A^2}}. \quad (4.24) \]

Plugging in \( A \) and \( B \) from Eqs. (4.20) and (4.21) yields a nasty transcendental result for the confocal parameter. When the generated Stokes power is very large, one must therefore solve for the confocal parameter numerically. Alternatively, when the heating is not so significant so that \( \xi \ll 1 \) and when \( R \gtrsim L \), one can simplify the result.

**Simplified Confocal Parameter**

The confocal parameter obtained through Eq. (4.24) can be simplified to

\[ b \approx b_0 \left[ 1 - \frac{L}{6} \left( \frac{2R - L)^2}{2R - L} \right) n_2 \right]^{-1/2}, \quad \xi \ll 1 \text{ and } R \gtrsim L \quad (4.25) \]

when the heating is not too great and when the field is almost a plane wave in the cavity. In this equation,

\[ b_0 = L \sqrt{\frac{2R}{L}} - 1 \]

(4.26)
is the confocal parameter in the absence of optical power (i.e. the cold-cavity value).

Inserting Eq. (4.17) into Eq. (4.25) and using Eq. (4.7) gives

\[
b = b_0 \left\{ 1 - \left[ \frac{(2R - L)^2 + 2R^2}{(2R - L)} \right] \left[ \frac{P_{\text{gen}}(n_0 - 1)(0.518) \nu_{21}}{12K_{\text{th}}T_0 b \lambda_{\text{int}} \nu_s} \right] \left( 1 + \frac{16\pi D\tau_{\text{vib}}}{b \lambda_{\text{int}}} \right)^{-1} \right\}^{-1/2},
\]

(4.27)

where

\[
\lambda_{\text{int}} = \frac{\lambda_p \lambda_s}{\lambda_p + \lambda_s}
\]

(4.28)

Equation (4.27) can be used to obtain the thermal waist numerically through

\[
\omega_{0,\text{th}}^2 = \frac{\lambda_{\text{int}} b}{2\pi} \left( 1 + \frac{16\pi D\tau_{\text{vib}}}{b \lambda_{\text{int}}} \right)^{-1}.
\]

(4.29)

With this, Eq. (4.15) can be used to determine the thermal mode pulling more accurately as a function of the generated Stokes power.

Of more use for the present discussion, is Eq. (4.27) in terms of the intracavity optical powers. Using Eq. (4.8) from above and the expression for \(P_1\) [Eq. (2.10)] from Chapter 2, one can express Eq. (4.27) in the analytical form

\[
b \approx b_0 \left\{ 1 + \left[ \frac{(2R - L)^2 + 2R^2}{(2R/L - 1)} \right] \left[ \frac{4P_p P_s(n_0 - 1)(0.518) \alpha \nu_{21}}{3K_{\text{th}}T_0 b_0^2 \lambda_p \lambda_s \nu_s} \right] \left( 1 + \frac{16\pi D\tau_{\text{vib}}}{b_0 \lambda_{\text{int}}} \right)^{-1} \right\}^{1/2}
\]

(4.30)

where only small errors have been introduce by using the cold cavity value of the confocal parameter, \(b_0\), rather than \(b\) in the last term. This allows the confocal parameter to be determined analytically as a function of the intracavity powers, as given by Eq. (4.30). The second term in curly brackets can be multiplied by the factor 4 for the linear cavity.
The important point from this section is that the confocal parameter (and therefore all the waists) for the stable cavity modes now depends on the generated Stokes power (or equivalently, the intracavity powers). This is the consequence of the thermal lens.

Modified Steady-State Behavior

The dependence of the confocal parameter on the generated Stokes power modifies the steady-state behavior of the Raman laser in two ways. First, the power constant $P_1$ from Chapter 2 depends on the confocal parameter, and therefore depends on the generated Stokes power. Second, because the stable cavity mode changes, the coupling efficiency into the cavity will also change with generated Stokes power. It is therefore possible to describe the system with the following three coupled nonlinear steady-state equations [see Eqs. (2.15), (2.16), and (2.10) for comparison]

\[
P_p(b) = P_1(b) [1 - R_{s,rt}],
\]

\[
P_s(b) = \frac{\lambda_p}{\lambda_s} \left\{ \left[ \frac{4T_{p,0} P_1(b) P_{inc}}{1 - R_{s,rt}} \right]^{1/2} \right\} \eta_{inc}(b) - P_1(b) [1 - R_{p,rt}]\right\},
\]

\[
b(P_p, P_s) = b_0 \left[1 + C_{th} P_p P_s \right]^{1/2}.
\]

where the equations are coupled through the unity gain constant (which isn't really constant anymore)

\[
P_1(b) = \frac{\lambda_p + \lambda_s}{16 \alpha \tan^{-1} (L/b)},
\]
and the coupling efficiency into the active cavity mode is given by

\[
\eta_{\text{inc}}(b) \equiv \frac{1}{\sqrt{V_p V_{\text{inc}}}} \int \int \int_{\text{cavity}} \tilde{u}_p(r) \tilde{u}_{\text{inc}}^*(r) \, d\mathbf{r}
\]

\[
= \frac{2\sqrt{b b_{\text{inc}}}}{b + b_{\text{inc}}}.
\]

(4.35)

Also, the constant

\[
C_{\text{th}} = \left( \frac{(2R - L)^2 + 2R^2}{(2R/L - 1)} \right) \left[ \frac{4(n_0 - 1)(0.518) \alpha \nu_2}{3 K_{\text{th}} T_0 b_0^2 \lambda_p \lambda_s \nu_s} \right] \left( 1 + \frac{16\pi D_{\nu \text{th}}}{b_0 \lambda_{\text{int}}} \right)^{-1}
\]

(4.36)

has been defined. In these equations, \(b_{\text{inc}}\) is the confocal parameter of the incident pump beam.

One can solve for the steady-state behavior including thermal lensing using an iterative approach. Specifically, one can first solve for the unperturbed intracavity powers \(P_p(b_0)\) and \(P_s(b_0)\) calculated using Eqs. (4.31), (4.32), and (4.26). These powers can then be used to solve for the first-iteration confocal parameter, which will be slightly larger than \(b_0\). This modified confocal parameter can then be used to calculate the modified intracavity powers, again using Eqs. (4.31) and (4.32). These revised powers are then used to obtain the second-iteration confocal parameter and so forth. The procedure can be repeated until the desired accuracy is obtained. For typical powers, usually only one or a few iterations are sufficient. Note also that if significant optical power is generated, then one can perform the same procedure, but calculate the confocal parameter numerically for each iteration using Eq. (4.24).

Figure 21 shows the most recognizable effect of the thermal lensing on the steady-state behavior. The figure shows the transmitted pump power for a linear cavity
and a ring cavity as functions of the pump rate. For comparison, the steady-state prediction from the earlier chapters is also included as a solid line. Clearly, the transmitted pump power no longer remains clamped above threshold when thermal effects are significant. The parameters used to generate these curves were:

\[ \lambda_p = 792\text{nm}, \lambda_s = 1180\text{nm}, P_1 = 114\text{kW}, R_{p,t} = 0.99695, R_{s,t} = 0.99926, T_{p,0} = 3000\text{ppm}, T_{p,1} = 25\text{ppm}, T_{s,0} + T_{s,1} = 625\text{ppm}, L = 7.6\text{cm}, R = 50\text{cm}, K_{th} = 2.18\text{mW/cm} \cdot K, \]

\[ n_0 = 1.001276, \tau = 38\mu s, D = 0.1443\text{cm}^2/\text{s}. \]

---

**Figure 21.** Plot showing transmitted pump powers as functions of the pump rate. The solid line represents the predicted behavior when thermal effects are ignored. The dotted line shows the behavior including thermal effects for the ring cavity and the dashed line shows the same for the linear cavity.
Figure 21 shows the thermal effects on the output Stokes power and the photon conversion efficiency. The conversion efficiency no longer necessarily peaks at four times the threshold value \( (r_p = 4) \) and the peak conversion efficiency drops. The drop in the peak conversion is only due to poorer coupling into the cavity and is therefore not a fundamental loss mechanism. In other words, one can recover the optimum peak conversion efficiency by adjusting the incident beam parameters. This practice of “thermal loading” the cavity is commonly encountered with solid state lasers.

Figure 22. Plot showing the output Stokes pump power and the photon conversion efficiency as functions of the pump rate.
To mitigate the effect of the lens, one can distribute the heat over a larger volume by increasing the length of a pass or by incorporating additional passes. This is a clear advantage of the bow-tie ring cavity over the two-mirror linear cavity.

Chapter Summary

The deleterious effects of the thermal contribution to the nonlinear refractive index in cw Raman laser systems were addressed in this chapter. The thermally-induced refractive index spatial profile was calculated and the on-axis contribution was used to estimate the blue-shift of the cavity mode resonance frequencies relative to their cold-cavity values [see Eq. (4.15)]. The ramifications of this mode pulling were discussed. Using the ABCD matrix formalism, the second order contribution to the refractive index profile was shown to cause an increase in the confocal parameter for the stable cavity modes. In other words, the heat deposited by the Raman gas was shown to effectively generate a diverging lens within the cavity.

The resulting modifications to the steady-state emitted powers from the previous two chapters were derived and discussed [see Eqs. (4.31) - (4.33)]. Due to the fact that the confocal parameter increases with the generated Stokes power, the transmitted pump power was shown to depend on the incident pump power above threshold (i.e. the transmitted pump power no longer clamped). Also, the emitted Stokes power departed from a strictly square-root dependence on the incident pump power and the peak conversion efficiency did not necessarily occur at four time the threshold value.
The next chapter marks the beginning of the experimental portion of this thesis. It discusses the construction and operation of a diode-pumped Raman laser with a strong emphasis on the task of frequency locking the pump laser to the Raman laser cavity. Using the techniques described in the next chapter, one can begin analyzing the behavior of the Raman laser in comparison with the theoretical predictions of this and the previous chapters.
CHAPTER 5

FREQUENCY LOCKING

This chapter describes the physical realization of diode-pumped cw Raman lasers. At its core, construction of a cw Raman laser is essentially an exercise in precision frequency stabilization. As discussed briefly in the introduction, the pump laser must be phase/frequency stabilized, or locked, to a resonance of the hydrogen-filled high-finesse cavity (HFC) so that Raman conversion can occur (see Fig. 5). In control theory terminology, the pump laser is the “plant” and the Raman laser cavity (the HFC) is the “discriminator” [104, 19]. For the cw Raman system, the HFC takes on the additional responsibility of generating Stokes light. Diode laser frequency locking is not a trivial task and requires a fair bit of expertise. Nevertheless, controlling the pump laser’s behavior can be rewarding and perhaps addictive after some experience is gained.

Fortunately, laser frequency stabilization has received a good deal of attention in the past few decades in such fields as coherent optical communications [19], frequency metrology [105], atomic physics [106], and high-resolution spectroscopy [107]. Even though many of the same techniques can be applied to cw Raman lasers, the goals and emphases can be different. In particular, for Raman laser locking one is typically only concerned with the relative stability between the pump laser source and the
HFC. At this point, the absolute frequency stability of the Raman laser output is not a significant concern.

Also, the degree of relative stability need not break any records. One typically only desires to efficiently and stably couple the pump light into the HFC for Raman conversion. A tangible goal might therefore be to achieve a relative frequency stability between the pump laser and the HFC of a few percent of the cavity linewidth. This ensures that the pump laser’s frequency noise will cause less than a few percent intensity noise on the pump light that is transmitted through the cavity. Because the typical Raman laser cavity linewidth is larger than 50kHz, a relative stability on the order of 1kHz or better will therefore suffice most of the time. Other researchers have achieved diode laser stabilities that are three orders of magnitude better than this [108].

Another difference between standard frequency stabilization schemes and the cw Raman laser system is that optical power is a much more valuable commodity. Although the locking itself can be performed with several tens of μW of light, cw Raman lasers require a good deal more than this just to reach the laser threshold. The primary limiting factor for several of the diode-pumped Raman experiments to date is insufficient pump light. For this reason, optical amplification is often an integral part

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1 One can envision using the Raman transition itself as an absolute frequency reference for slow time scales if this is important.
of the feedback loop. The technique of injection locking is investigated for amplification in this thesis. A passive optical locking technique is also shown to permit higher pump powers and eliminate the need for a high-bandwidth electronic servo.

It is my pleasure to point out that Jay Brasseur was instrumental in the early development of my frequency locking skills with the Nd:YAG-pumped cw Raman laser [44]. More generally, it was Jay who brought the techniques of precision frequency locking to MSU physics. To my knowledge, his locking servos and/or low noise PZT drivers (adapted from those of Jan Hall) are currently used in every precision frequency stabilization scheme in MSU physics. As will be discussed in this chapter, some aspects of frequency locking are made easier with diode lasers (the fast current feedback path) and some are made more difficult (as much as two orders of magnitude more frequency noise to start with).

This chapter begins with a review of some pertinent diode laser phase/frequency noise reduction and stabilization references. The second section gives a flexible and useful experimental layout for the cw Raman laser system. The third, fourth, and fifth sections describe the three essential elements of the actively locked cw Raman system: the diode laser pump source (the plant), the high-finesse Raman cavity (the frequency discriminator), and the electronic feedback amplification/filter system (the servo). The sixth section describes the injection locking technique (the amplifier), which is an optional element in the feedback loop. The seventh section describes the advantages and drawbacks of the passive optical stabilization technique. The final
section reviews the findings of this chapter and provides some concluding thoughts.

### Previous Work

As shown in Fig. 23, one might view diode laser stabilization on a scale with one end completely passive and the other end completely active. Most diode laser noise reduction and stabilization schemes fall somewhere in the middle.

![Scale of possible diode laser stabilization schemes. Most feedback configurations, including ECDL locking, use some passive and some active feedback for frequency noise reduction.](image)

The passive end of the scale is represented by techniques such as resonant optical feedback. This technique was introduced by Dahmani, et al. in 1987 [109]. The researchers demonstrated that the phase noise of a free-running diode laser can be substantially reduced by spectrally filtering and returning a small fraction of its emitted light back into the diode. Specifically, the researchers coupled a small portion of the light from a free-running diode laser into a confocal HFC in an off-axis geometry so as to prevent the reflected portion of the input light from returning to the diode source. The field that was resonant with the cavity, however, was allowed to return to the diode. Such resonant optical feedback was shown to dramatically narrow the
laser's spectral width and provide center frequency stabilization to a cavity resonance.

Many related experimental and theoretical studies of this locking technique followed (see, for instance, Refs. [110, 111]). The advantages of the method are its simplicity and the fact that nearly all of the diode laser's free running optical power can be delivered to the experiment. The method does not necessarily require any modulation or wide-bandwidth electronics and it can be implemented without the use of optical isolators. Its primary drawback is that the locking depends critically on the phase of the optical feedback so that the overall distance between the diode and the feedback element (the HFC) must either be inherently stable, or it must be actively stabilized (albeit with low bandwidth electronics). A further shortcoming is that an additional servo loop is required to enable frequency tuning [112]. The tuning must be performed using the laser's injection current. Nevertheless, when tuning is not an issue, the optical feedback alone can lock the diode laser's frequency for time scale of minutes to hours.

The active end of the scale shown in Fig. 23 is represented by extremely fast electronic techniques. Telle [113, 107] and also Ohtsu [114] showed that one can significantly reduce the frequency noise of free-running diode lasers with electronic feedback alone. However, these efforts required diodes with exceptional modulation characteristics and ultra-wideband electronic servos. Of course, the frequency tunability of the stabilized system is limited to that of the free-running diode laser (i.e. temperature and current tuning).
External-cavity diode laser (ECDL) stabilization represents a compromise between the completely passive and the completely active techniques. The external cavity provides some optical noise reduction relative to the free-running diode laser, and electronic feedback of reasonable bandwidth (typically $\lesssim 2$MHz) is also employed. ECDL stabilization therefore falls somewhere in the middle of the scale shown in Fig. 23, with the exact location depending on the particular ECDL design used. This hybrid stabilization method has been researched for quite some time now [115, 116, 106, 108, 117, and references therein].

Many of the stabilization results are impressive, but it is often difficult to extract practical information, such as full circuit diagrams, from the mainstream literature. One notable exception is the very recent work of Fox, et al., which provides an excellent practical guide for actively frequency locking an ECDL to a high finesse optical cavity [117]. This reference is highly recommended and would have saved me a great deal of time and effort if it had been printed earlier. To some extent, the section of this chapter covering the electronic servo design can be viewed as a supplement to the work of Fox, et al. I am aware of only two other examples of diode laser locking servo circuit diagrams in the literature [115, 116].

The primary advantage of the hybrid active locking technique is the high degree of control achieved with a single feedback loop. It will be shown that frequency tuning can be achieved without difficulty through PZT control of the external cavity length. Also, the overall distance to the HFC need not remain stable from an interferometric
The primary drawbacks of the active locking are the complexity involved with generating, detecting, amplifying, and mixing the error signal, not to mention the challenges associated with constructing a sufficiently wideband electronic servo. Furthermore, the output powers from most ECDLs are substantially lower than their free-running powers due to optical feedback from the grating element. As a result, less optical power can be delivered to the HFC for Raman conversion.

One can therefore incorporate the technique of injection locking to amplify the output from the ECDL for actively-locked cw Raman laser systems. Injection locking has been widely researched [17, Chapter 29] and for diode lasers involves injecting a small amount of spectrally pure light into a free-running diode [118, 119, and references therein]. The free running laser retains its ability to safely produce high output power, but it acquires the superior spectral characteristics of the injected light. When combined with active stabilization, injection locking has proven invaluable for producing well-behaved, high power diode-pumped Raman lasers. A similar, and perhaps preferred alternative is to use a high-power tapered amplifier instead [120]. However, these amplifiers have been very difficult to obtain commercially for several years now and have not been used in the work of this thesis.

**Experimental Layout**

A particularly flexible and useful experimental layout for the diode-pumped cw
Raman laser system is shown in Fig. 24. The optical portion of the layout is separated into subsystems: the pump laser, the injection locking, and the Raman cavity. Detailed descriptions and experimental setups for these subsystems are provided later in the chapter. The electronic servo portion of the feedback loop is also shown. The essential elements of the feedback loop are therefore the pump laser plant, the Raman cavity discriminator, and the electronic servo. The injection locking subsystem is optional and is used only for amplification purposes.

Figure 24. Flexible and useful experimental layout for the diode-pumped Raman laser system. The layout is divided into three optical subsystems and an electronic servo. The use of optical fibers facilitates beam alignments and adjustments.

The three optical subsystems can be connected by polarization-maintaining (PM) single-mode (SM) fibers. To initiate and optimize the frequency lock, one can first
connect the pump laser subsystem directly to the Raman cavity subsystem via the connection labeled A (B and C are not connected). If the pump laser subsystem alone can deliver sufficient optical power to the HFC for Raman conversion, then the injection locking subsystem can be ignored. However, if additional optical power is required, and the lock has been optimized, one can then direct the pump laser light to the injection locking subsystem via connection B. The output from this subsystem is then used for Raman conversion via connection C. Regardless of whether injection locking is used, it is often useful to sample the pump laser light using a separate fiber for diagnostic purposes. Light from this fiber can be sent to an optical spectrum analyzer (OSA) or a flat-plate Fabry-Perot to monitor the spectral characteristics of the pump laser.

The optical fibers serve more significant purposes than just setup flexibility. First, adjustments to the optical alignment of any one of the three subsystems will not affect alignment in the other two. Second, because the pump laser's mechanical stability is of utmost importance, one can spend some time vibrationally isolating the entire pump laser subsystem. Lei Meng found that placing the components on a 1/2" slab of aluminum, cushioned on the bottom side with bubble wrap (the packaging material) works well for this purpose. Others have used much thicker slabs for the same purpose with improved results. The pump laser subsystem is therefore capable of movement relative to the rest of the optical table, and the fiber preserves alignment with the rest of the experiment. Also, as described later in this chapter, the fiber path through
connection B is used to automatically align the light from the pump laser into the slave laser for injection locking. Use of the fiber here also allows one to estimate the amount of master light that is coupled into the slave laser’s spatial mode. Finally, because the output from the fiber is in a high-quality spatial mode, the process of optimizing the light coupling into the HFC is significantly simplified. As a note, the fiber lengths should be kept as short as possible to reduce time delays (resulting in phase lag) in the locking feedback loop.

**ECDL Pump Laser Design**

The pump laser serves as the plant and is the first essential element in the feedback loop [19]. Its frequency must be stabilized to a resonance of the HFC (the discriminator). Unfortunately, a free-running laser’s linewidth is often greater than several MHz. Extraordinarily wide electronic servo bandwidths and diode lasers with exemplary modulation characteristics are required to achieve sub-kHz relative stability between a free running diode laser and a HFC [114].

It is therefore advantageous to employ some passive reduction of the laser’s frequency noise first so that an electronic servo of reasonable bandwidth can be employed to achieve the desired level of stability. The most common method of accomplishing this is by placing the free-running diode laser in an external cavity. The back facet of the diode laser is highly reflective and forms one end of the external cavity, while the first order reflection from a dispersive diffraction grating often forms the other. The
front facet of the diode can also form a cavity with the the back facet. It is therefore advantageous to anti-reflection (AR) coat the front facet to minimize competition between the two cavities. Most commercial diodes already have a reasonable ($\leq 5\%$) AR coating. For the ECDLs presented in this thesis, only commercial free-running diode lasers were used with nothing more than the manufacturer’s standard AR coatings.

There are two dominant external cavity configurations: the Littman-Metcalf [121, 122, and references therein] and the Littrow [123, 124, 125, and references therein]. Simple diagrams of these cavity configurations showing the optical beam paths are provided in Fig. 25. Both of these configurations have been used as pump sources for cw Raman lasers. Each has its advantages and drawbacks.

![Littman-Metcalf](image1)

![Littrow](image2)

Figure 25. Simple diagram showing the Littman-Metcalf and Littrow configuration external-cavity diode lasers (ECDLs). There are advantages and drawbacks for each configuration.

The desired qualities of the ECDL source for pumping a cw Raman laser are as follows:
1. A reasonable continuous frequency tuning range. This depends on the free spectral range (FSR) of the HFC and the Raman shift, but a tuning range \( \geq 4\text{GHz} \) (2 FSRs) is typically sufficient for the work presented in this thesis. Larger tuning ranges are preferred, but may not improve the continuous tuning range of the Stokes laser output. The Stokes continuous tuning range is fundamentally limited by the double resonance requirement, as discussed in Appendix D.

2. A coarse frequency tuning range that is sufficient to generate the desired Stokes output frequency. A free running diode laser should be chosen such that the center of its gain profile will Raman shift as close to the desired Stokes frequency as possible.

3. A reasonably narrow linewidth (\( \leq 1\text{MHz} \)). The narrower the inherent linewidth of the ECDL, the less work the electronics will have to do. A narrow linewidth will save many headaches when locking.

4. Low long-term drift (\( \leq 50\text{MHz/hr} \)). As it turns out, the absolute frequency stability of the Stokes laser output at slow Fourier frequencies is determined by that of the ECDL pump source. It is therefore desirable for the ECDL to drift much less than the Raman transition linewidth for times on the order of the experiment. This is typically not a difficult condition to satisfy.

5. High output power. Clearly, it is advantageous to deliver as much optical power to the HFC as possible for Raman conversion. Most commercial free-running single-mode diode lasers exhibit powers below \( \sim 100\text{mW} \). Placing the laser within an external cavity decreases this maximum output power due to optical feedback from
the grating element.

6. Insensitivity to acoustic and mechanical perturbations. Because one often wishes to either use the Stokes output in some other experiment or measure the characteristic of the Stokes output itself over time period of up to an hour, it is necessary for the ECDL frequency lock to withstand ordinary lab conditions such as shutting doors, moving chairs and adjustment of elements on the optical table.

With these qualities in mind, the advantages and drawbacks of the two different ECDL configuration are now discussed.

Littman-Metcalf Configuration

The Littman-Metcalf cavity configuration was the first ECDL to be used as the pump source for cw Raman lasers. This ECDL was designed and constructed by Gregg Switzer originally for the purpose of water vapor spectroscopy [126]. Pictures of this cavity design are shown in Figs. 26(a) and (b).
Because the ECDL was designed to interrogate broad absorption spectra, it can exhibit an extraordinarily large continuous tuning range. In fact, when properly aligned and when using an anti-reflection coated diode, the ECDL can tune continuously over 20-30nm without a mode hop. Even when standard commercial diodes are used, it exhibits far more than adequate continuous tuning (>20GHz). The large tuning range is made possible by simultaneously tuning the ECDL cavity length and reflection angle. This requires very precise positioning of the mechanical tuning pivot point.

Because the diode light hits the grating at grazing incidence for the Littman configuration (see Fig. 25), the majority of the 4cm grating is illuminated. Also, the light bounces twice off the grating during the round trip in the external cavity. These two features dramatically enhance the dispersion (proportional to the number
of grating lines illuminated) of the grating, and therefore substantially suppress the growth of any external cavity modes other than the lasing mode. With the commercial diode, the ECDL can exhibit a mode suppression ratio slightly under $50\text{dB}$ (greater than $55\text{dB}$ with an AR coated diode). This is a good first indication that the fast frequency noise of the diode laser (which comes partly from mode competition noise) will also be significantly reduced. A further indication of the frequency noise reduction is that the laser’s linewidth has been estimated using a self-homodyne beat technique [127] to be less than $200\text{kHz}$. This Littman cavity length was approximately $6.5\text{cm}$. Other researchers have noted that the longer cavity lengths tend to yield narrower linewidths [117]. The highly dispersive feedback and the meticulous design of this Littman-style ECDL therefore result in superb high frequency noise suppression and tuning characteristics, as will be substantiated.

The primary drawbacks of the laser design for use as a pump source for Raman lasers are that it is highly susceptible to lower frequency acoustic perturbations and it can only output $\leq 1/3$ of its free running optical power. Shutting a door or moving a chair anywhere in the lab, let alone adjusting elements on the optical table, often causes large frequency excursions of the laser and results in disruption of the Raman laser lock. Another downside of this design is that the machining, assembly, and alignment of the ECDL are nontrivial tasks; together they typically require a few months.
Littrow Configuration

In an attempt to produce a more robust lock to the Raman laser cavity, a Littrow-style ECDL was also investigated. This laser design was similar to that of Arnold, et al. [125], except that the entire mount was machined from scratch instead of using a commercially available mount. Also, the output beam was bounced off another reflector as shown in Fig. 25 to prevent angular beam steering during tuning. This simple technique was also recently demonstrated by Hawthorn, et al. [128]. Pictures of the Littrow cavity design are shown in Fig. 27.

Figure 27. (a) Back and (b) front images of the Littrow-style ECDL designed to produce a more robust lock to the high-finesse Raman laser cavity.

High tension springs hold together two monolithic aluminum pieces that were each more than 1.25cm (1/2") thick. One of the aluminum pieces houses the laser and the other holds the grating and reflection mirror. The laser block is temperature
controlled using a thermoelectric cooler. The total machining and alignment of two of
these lasers took less than three weeks. A PZT stack allows fine tuning of the laser’s
frequency by simultaneously adjusting the grating feedback angle and the external
cavity length. The length of the cavity is nominally \( \sim 1.5 \text{cm} \), but can be adjusted
to over \( \sim 2.5 \text{cm} \) (adjustment of the cavity length requires realignment of the grating
feedback).

The Littrow ECDL can emit between 60% and 85% of its free running optical
power, depending on the grating used. The particular grating suggested by Arnold,
et al. [125] was found to cause polarization problems for some diode lasers but not
for others. The continuous tuning range is typically \( \sim 4 \text{GHz} \) (about half of the FSR)
which is adequate for the Raman laser purposes. Enhancing the continuous tuning
range by adjusting the external cavity pivot point was not investigated. Other re­
searchers have shown that such adjustments can increase the continuous tuning range
to \( \geq 10 \text{GHz} \) [128]. A heterodyne beat note of \( \sim 2 \text{MHz} \) was measured between two of
these lasers. Other researchers using Littrow cavity designs have reported linewidths
below 400kHz [123, 125]. The linewidth as well as the continuous tuning behaviour
were therefore worse than other Littrow designs and the Littman design. Neverthe­
less, the laser typically exhibited \( \sim 40 \text{dB} \) mode suppression and was substantially less
sensitive to acoustic perturbations than the Littman design discussed above. Fu­
ture designers are encouraged to save some headaches and follow the designs in the
literature more closely.
Assessing the Pump Laser Frequency Noise

It is useful to quantify the laser’s frequency noise in order to compare the two cavity designs and to estimate the ease with which the electronic feedback will be able to achieve the desired frequency stability for each design. The linewidth measurement via heterodyne beat note can be used to quantify the laser’s frequency noise level, but the linewidth depends on the measurement integration time, and is therefore somewhat arbitrary. It is also difficult to identify individual frequency noise contributions to the linewidth. Allan variance measurements also provide information about the laser’s frequency stability. This technique is usually best suited for long term (>1s) stability measurements.

A third measure of the laser’s noise is the frequency noise spectral density (NSD) [129]. This is the preferred assessment method here, primarily because it offers a spectral map of the laser’s various frequency noise contributions. In general, ECDLs exhibit noise contributions from a variety of different sources at different Fourier frequencies. These are discussed at length elsewhere [108, 129, 117, and references therein]. The frequency noise assessment method can therefore be used to identify (and hence to minimize) these contributions. Another advantage of using the NSD for frequency noise assessment is that it can be used to directly calculate both the laser’s linewidth and the Allan variance [117, 129, and references therein].

Although other methods exist, the NSD can be measured by simply parking the laser on the side of a Fabry-Perot (FP) peak and measuring the transmission using a
fast detector and an RF spectrum analyzer. The FP converts frequency fluctuations of the laser into intensity fluctuations of the FP transmission. A low bandwidth active lock can be used to hold the laser to the half maximum point of the FP transmission peak. The choice of FP linewidth represents a trade-off between measurement sensitivity (determined by the maximum slope of the cavity transmission peak) and measurement bandwidth (determined by the half-width of the cavity resonance). It should be realized that such a measurement is also sensitive to the laser’s amplitude noise. With a reasonably narrow cavity linewidth (≤10MHz), the frequency noise typically dominates the amplitude noise, but this should be verified.

A typical setup is shown in Fig. 28. Note that the laser source should be optically isolated (preferably >60dB) from the FP because small amounts of optical feedback will affect the laser’s frequency noise. The single-mode optical fiber is used simply for ease of alignment of the FP. The cavity converts the frequency noise into intensity noise, which is then measured on two detectors. The output of the fast detector (D2) is sent to the RF spectrum analyzer for analysis, while a small amount of light is sent to another detector (D1) for low-bandwidth locking purposes. The cavity can be bypassed in this setup to measure the amplitude noise of the laser for the same light intensity that is incident on the detectors during the frequency noise measurement. Sometimes, additional wideband amplification is required to adequately surpass the background noise level of the spectrum analyzer. The resulting NSD is often expressed in the units of Hz²/Hz.
Figure 28. Setup used to measure the frequency noise spectral density of the ECDLs. The Fabry-Perot (FP) length is adjusted via low bandwidth feedback and a PZT to ensure that the laser remains parked on the side of the FP transmission peak.

One can convert the signal measured on the spectrum analyzer (in watts of electrical power) into NSD using

\[ NSD = \frac{SA \times R_{in}}{BW \times Amp \times Slope^2} \]  

where \( SA \) is the measured spectrum analyzer signal minus noise (watts), \( R_{in} \) is the spectrum analyzer’s input impedance (ohms), \( BW \) is the resolution bandwidth used for the measurement (Hz), \( Amp \) is the power amplification of the detected signal (unitless), and \( Slope \) is the slope of the FP transmission peak (volts/Hz) at the point of the lock (typically at half of the peak maximum). This slope can be obtained by scanning the FP cavity length and is calibrated using the known free spectral range of the cavity. One should take care to properly deal with the various impedances of
the detector, amplifier, and spectrum analyzer in this conversion.

Figure 29 shows some experimental results from the NSD setup. Trace A shows a NSD obtained from the Littman-style ECDL, while trace B shows that from Littrow-style ECDL. The peaks observed around 8kHz are due to PZT resonances excited by excessive bandwidth of the locking servo and are not representative of the laser noise. The peaks at higher frequencies in trace A were determined to be due to a noisy electronic component operating near the diode laser. These were later eliminated, but the identification of this noise source highlights the utility of the NSD as a measure of the frequency noise. It should also be noted that the behavior of both of these lasers depends critically on several parameters including the particular free-running diode laser used (manufacturer, maximum output power, etc.), the drive current supplied to the laser, the alignment of the grating feedback, and the laser's spectral proximity to a mode hop. Nevertheless, some characteristics are consistently observed for each laser style.

The Littman ECDL consistently exhibits greater noise at lower Fourier frequencies than the Littrow ECDL. On the other hand, the high frequency noise level of the Littman ECDL consistently drops below that of the Littrow ECDL. Both lasers exhibit plateaus in their noise spectra at high frequencies (this is not clear for trace A in Fig. 29), but the Littman plateau level ($\sim 10^3\text{Hz}^2/\text{Hz}$) is typically an order of magnitude below that of the Littrow ($\sim 10^4\text{Hz}^2/\text{Hz}$).
Figure 29. Data showing the frequency noise spectral density as a function of Fourier frequency for the Littman (trace A) and Littrow (trace B) ECDL configurations.

These characteristics emphasize some of the strengths and weaknesses of each design as discussed in the previous two subsections. Namely, the rigid, monolithic design of the Littrow laser suppresses low frequency noise quite well, while the highly dispersive grating reflection of the Littman design suppresses noise at higher frequencies. When it comes to stabilizing these two lasers to the high-finesse Raman laser cavity, both of the cavity styles lock successfully, but it is considerably easier to reduce intensity fluctuations that are transmitted through the HFC with the Littman ECDL design (meaning that the linewidth is narrower). Retaining the lock, on the
other hand, is considerably easier with the Littrow ECDL.

Based on the diode-pumped Raman lasers that have been constructed thus far (50kHz < HFC linewidths (FWHM) < 1MHz), the characteristic high frequency noise plateau in the ECDL’s frequency noise spectra should be reduced via optical means (an external cavity) to at least $\sim 10^4 \text{Hz}^2/\text{Hz}$ if possible, preferably below. Although it will be shown that active electronic feedback will also reduce the frequency noise, it is well worth spending some extra time reducing the noise by optical means. This will substantially simplify and improve the subsequent electronic stabilization effort.

**Pump Laser Transfer Function**

In order to properly design a servo for electronic locking, one must measure the frequency responses, or transfer functions, of the various elements in the loop. For the present system, it turns out that neither the pump laser, nor the HFC respond uniformly at all Fourier frequencies. Measurements of the pump laser transfer function are provided in this subsection.

The first element of the feedback loop that displays nonuniformity as a function of Fourier frequency is the response of the diode laser itself to current modulation. This is important because the current is used as the feedback path to correct the laser's frequency. The precise response of different diode lasers can vary significantly, but the transfer function is sometimes modeled as a single pole (20dB/decade roll off in magnitude) beginning somewhere between 100kHz and 1MHz. One would therefore...
expect that frequency corrections will be attenuated and will lag the current corrections in phase by $-90^\circ$ for Fourier frequencies above the pole in the diode's response. The physical nature of the pole is discussed in Ref. [107]. Basically, the frequency response of diode lasers to current modulation is dominated by thermal effects at low frequencies, but carrier density effects at high frequencies. Unfortunately, one of the effects generates a blue-shift and the other a red-shift of the laser's frequency. This can result in nasty phase characteristics of the response.

Because this behavior often varies between diodes and because the response is not truly single-pole in character, it should be measured. Fortunately, the transfer function measurement can be easily performed with the setup shown in Fig. 28. While dc-locked to the side of a FP peak, one can simply modulate the laser's injection current using the source output of a network analyzer and monitor the resulting frequency (converted to amplitude) modulations measured on the fast detector through the network analyzer's input port. This method can also conveniently take into account the response of any electronics used to add the modulation to the laser's current.

The curves labeled A in Fig. 30 show the results from one such measurement. The curves labeled B show the predicted behavior from a single pole at 400kHz. The data has been corrected for round-trip time delays introduced by optical fiber length and for the poles introduced by the FP measurement cavity. From the figure, one can see that the response is often not strictly single-pole in character. The magnitude roll off is reasonably well approximated by the 20dB/decade decline of the single pole.
But perhaps more importantly, the phase lag is more severe than a single pole and falls off sharply at high frequencies. This effect is seriously detrimental to any hopes of an ultra-wide locking bandwidth.

Figure 30. The curves labeled A show the measured magnitude and phase of the diode laser modulation transfer function. The curves labeled B show the predicted behavior assuming a single pole at 400kHz. The excessive phase lag exhibited in the diode laser’s modulation response limits the servo bandwidth.

The excess phase lag means that, without the mitigating effects of the servo, corrections made at high Fourier frequency eventually represent positive rather than negative feedback. This leads to closed loop instability issues. Even though the servo
can help push these instabilities to higher frequencies, the excess phase lag induced by the diode modulation response ultimately limits the achievable locking bandwidth of this system. The transfer function measurement is therefore critical in estimating and optimizing the servo bandwidth, as will be shown later in this chapter.

**Pump Laser Subsystem**

With sufficient passive noise reduction of the ECDL achieved and its transfer function measured, one can assemble the remaining elements of the pump laser subsystem (see Fig. 24). A diagram of the experimental setup for the pump laser subsystem is shown in Fig. 31.

![Diagram of Pump Laser Subsystem](image)

**Figure 31.** Setup for the pump laser subsystem. The output from this subsystem can either go to the injection locking subsystem for amplification or directly to the Raman cavity subsystem for conversion to Stokes.

Although it is not shown in the figure, a low-noise current supply following the design of Libbrecht, *et al.* [130] is used to drive the ECDLs. The current supply
is equipped with a buffered modulation input for electronic feedback to the laser injection current. After the construction and initial operation of this current supply, an undergraduate in the lab, Ross Dermott, confirmed that its noise performance compared favorably to several commercial supplies and initiated future circuitboard layouts.

The laser's output is first directed through at least one optical isolator (>60dB isolation is recommended when using a linear HFC). An anamorphic prism pair can be used to shape the diode's beam profile before the isolators if so desired. A half-wave plate and polarizing beam splitter serve as a variable attenuator. One output from the polarizer is fiber coupled and used for diagnostic purposes (see Fig. 24). The other output from the polarizer is sent through an EOM to place phase modulation sidebands on the optical carrier for locking purposes. This beam is then fiber coupled and sent to either the Raman cavity subsystem or the injection locking subsystem (see Fig. 24). The half-wave plate can be used to adjust the amount of light sent to each fiber.

Raman Laser Cavity Design

The second essential element of the feedback loop is the HFC (the discriminator). The discriminator is used to convert optical frequency fluctuations of the pump laser into voltage fluctuations. The section following this describes how these voltage fluctuations are then fed back to the laser to stabilize its frequency. The Raman laser
cavity also provides the necessary power enhancement to convert the pump light into Stokes. Four different cavity designs were investigated over the duration of this thesis work. Two were linear standing-wave cavities, one was a standing-wave V-shaped cavity and the last was a traveling-wave bow-tie ring cavity. Only the better of the two linear cavities and the ring cavity will be described in this section. The V-cavity reduced the need for optical isolators, but it suffered from inherently poor impedance matching.

Most often, high-finesse reference cavities for precision frequency stabilization are designed with ultra-low expansion materials, are evacuated, and are acoustically isolated to obtain the highest possible level of absolute stability [25]. For Raman laser HFCs, only vibrational isolation is recommended. Obviously, the Raman gas must occupy the region between the mirrors so evacuation is not possible. The presence of the gas and the fact that PZTs must control of the cavity length to counteract thermal mode pulling render ultra-low expansion materials virtually useless. The cavities were therefore constructed out of aluminum. The PZTs were tube-style with one-inch outer diameters. The light enters and exits each of the cavities through 3/8” AR coated windows. These windows must withstand the gas pressure, which is usually less than 12 atmospheres. The high-finesse cavity mirrors, on the other hand, are within the hermetically-sealed aluminum containers and are never exposed to pressure differences. The mirrors were all purchased from Research Electro-Optics in Boulder, CO. The back surface of the mirror substrate should be AR coated if
possible. The two different cavity styles are now described.

Linear Cavity

For the linear cavity, a design with two nested aluminum concentric cylinders has performed very well for the cw Raman work thus far. A schematic cross-section of the cavity is provided in Fig. 32. The schematic is for conceptual purposes only: it is not exactly to scale and does not show all the components of the apparatus. Also, a digital image of the cavity is shown in Fig. 33 (the inner cylinder is not visible in the image).

![Diagram of linear cavity design](image)

Figure 32. Schematic cross-section of the linear cavity design. The schematic is not exactly to scale and does not show all the components of the apparatus.

The outer cylinder provides containment of the Raman gas while the inner cylinder holds the cavity mirrors. The space separating the two cylinders is filled with an
acoustically and thermally isolating material. Three one-inch PZT tubes separate the mirrors. The endcaps of the inner aluminum cylinder clamp the mirrors and PZTs together. O-rings between the back surfaces of the mirrors and the endcaps preload the PZTs slightly. For the linear cavity, a wedged back surface of the mirror is recommended to avoid interference effects, even if it is also AR coated. An undergraduate in the lab, Anna Hagenston discovered from ring-down measurements that interference from the (uncoated) back surface of the mirror can alter the mirror transmissions by as much as ±15%. The outer cylinder was machined to fit in a 3” optical tilt mount, so only one external mirror is required for coupling. The total length of the outer cylinder is about 6”.
Ring Cavity

The bow-tie style ring cavity does not use a nested cylinder approach. A schematic diagram of this cavity is provided in Fig. 34. Again, the schematic is not exactly to scale. A picture of the ring cavity is also provided in Fig. 35. It was machined out of one solid block of aluminum. Four entrance holes are at slight angles towards the center of the rectangular aluminum block. The cavity reflection angles are kept small ($< 5\,^\circ$) so that the mirrors' reflectivities are not altered and to minimize birefringence effects in the mirrors. Two of the mirrors are flush up against PZT tubes to provide cavity length control. Air paths are provided so that no pressure difference exists
between the inner and outer mirror surfaces. Again, the gas pressure is exerted on
the entrance/exit windows. The PZTs are again pre-loaded using o-rings. This cavity
was not machined to fit in an optical tilt mount, so two external mirrors are needed
for coupling. The total length of the apparatus is again about 6".

Figure 34. Schematic cross-section of the bow-tie style ring cavity design. The
schematic is not exactly to scale and does not show all the components of the appa­
ratus.

Without a nested structure similar to the linear cavity, the ring was noticeably
more susceptible to thermal perturbations. By grasping the cavity with a warm hand,
one could relatively quickly change the cavity length by several wavelengths. However,
this did not prove to be a problem. In fact, one could envision using thermal control
of the cavity length in the feedback loop if the PZT range were insufficient. Wedged
mirror back surfaces are not needed for the ring cavity.
Raman Cavity Transfer Function

In the last section, the frequency response of the diode laser to current modulation was measured. Similarly, one must also assess the transfer function of the second essential element in the feedback loop (the HFC or Raman cavity). Indeed, the cavity also exhibits a nonuniform response as a function of Fourier frequency. However, the transfer function of the cavity is well known and predictable, and therefore need not be measured for Raman laser stabilization as long as the cavity linewidth is known.

In reflection mode, the HFC exhibits a well-behaved single pole at half its linewidth [23,
114]. This means that the error signal derived from the cavity is in phase with the input noise below the cavity half width, but is attenuated (20dB/decade) and is \(-90^\circ\) out of phase above the cavity half width. The curves in Fig. 36 show the expected magnitude and phase behavior of the HFC transfer function. The cavity pole was chosen at 40kHz. The magnitude and phase behavior of this second element of the feedback loop can and will be compensated by the servo.

Figure 36. Theoretical transfer function of the high-finesse Raman cavity. This response need not be measured because it is very predictable.
Raman Cavity Subsystem

With the Raman HFC machined, one can assemble the remaining elements of the pump laser subsystem. A detailed diagram of this subsystem is shown in Fig. 37 for the ring cavity. A PM, SM fiber delivers the pump light either directly from the pump laser subsystem or from the injection locking subsystem (see Fig. 24). A single lens can be used to focus the output from the fiber to the appropriate spot size. The minimum beam waist is determined by the mirror spacing and curvature through [50, page 140]

$$\omega_{0,p}^2 = \frac{\lambda_p L}{2\pi} \sqrt{\frac{2R}{L}} - 1. \quad (5.2)$$

Two mirrors are used to couple light into the ring cavity because there is no adjustment on the cavity itself. The center of one cavity pass is places at the location of the minimum beam waist. Aligning the HFC takes some practice, but is significantly facilitated by the fact that the spatial mode exiting the fiber is nearly perfect. During alignment, one first scans the cavity and observes the transmitted pattern using an IR viewer and a white card. The coupling mirrors are then adjusted to minimize the transmitted spot size. This spot can then be directed onto a detector and the coupling can be optimized by viewing the transmitted signal on an oscilloscope.
Figure 37. Setup for the Raman cavity subsystem. The input pump light can come from either the pump laser subsystem or the injection locking subsystem.

A small portion of the input light is sampled for data-taking purposes (D1). One can also use the small reflection from the back surface of the input coupler mirror for this purpose. The main reflection from the input coupler comes off at an oblique angle and is attenuated and focused onto a fast, low-noise detector (D2) for locking purposes. The dc reflected pump power level can also be monitored (D3). The cavity transmission is separated using a Pellin-Broca prism or a dichroic mirror and both the transmitted pump and the emitted Stokes powers are monitored (D4 and D5). Sometimes additional narrow band filters are used in front of these detectors to ensure the purity of the measured light. Although only one output port is shown, Stokes and pump light will exit all four ports. Aluminum pieces are clamped up
against the cavity so that one can remove it, fill it with hydrogen, and replace it without significantly degrading the cavity coupling. When the linear cavity is used, a polarizer and quarter-wave plate can separate the incident light from the reflected light for error signal detection. With the pump laser and HFC well understood, one can now concentrate on the last essential element in the feedback loop.

Electronic Servo Design

The third element in the feedback loop is the electronic servo. This element processes the information from the discriminator (the HFC), and provides the necessary negative feedback to the plant (the pump laser) in order to lock the laser’s frequency. Several steps are involved in the process of designing and implementing the servo. First, a discussion is given on generating the error signal from the discriminator cavity. This is brief because the topic has been thoroughly covered by other authors. The transfer functions measured in the previous two sections are then used to design the servo electronics so that the feedback bandwidth is optimized. Finally, one can close the feedback loop and start locking the laser. The manners in which the Raman gas in the cavity influence the process of initiating and optimizing the lock are discussed.

Generating the Error Signal

The error signal is derived from the reflected optical field from the HFC (the discriminator). By measuring the reflected field, one can convert optical frequency
fluctuations of the pump laser into voltage fluctuations, which are then used to stabilize the laser. In other stabilization schemes, the cavity transmission can be used to generate the error signal. Raman lasing within the cavity prohibits this approach for the current system. It is possible to lock to the side of the HFC transmission peak as was performed for the pump laser noise assessment, but this is also unappealing for the Raman laser system because much of the light does not enter the cavity and is therefore wasted.

There are several methods of generating an error signal that will produce a lock to the center of a cavity resonance [131, 132, 23, 133]. Only the Pound-Drever-Hall (PDH) method has been investigated for the work presented in this thesis. A great deal of attention has been focused on this method in the literature [23, 24, 19, 105, 25, 117]. In particular, Ref. [117] gives a thorough and applicable discussion of the method. The discussion includes some tips on coupling light into the HFC, generating modulation sidebands, detecting the reflected field, and demodulating the error signal. These topics will therefore not be discussed here. It is noted here that the EOM in the pump laser subsystem and the fast detector (D2) in the Raman cavity subsystem are both integral to the generation of a PDH error signal.

As an aside, future researchers in this field are encouraged to investigate the “tilt locking” method demonstrated by Shaddock, et al, because it eliminates the need for an EOM and for rf modulation altogether [133]. With an error signal ready for processing, one should next identify the salient features of the open-loop transfer
Identifying the Loop Response

Feedback loops are discussed at length in texts [134, 104] and have been applied to laser linewidth reduction by several authors (see, for example, Refs. [19, 105, 108, 117]). Of critical importance for the current discussion is the loop transfer function in the absence of the servo. It was already shown that neither the plant nor the discriminator responds uniformly at all Fourier frequencies. The transfer functions of these loop elements were either measured or estimated (see Figs. 30 and 36).

One can combine these two frequency responses to generate the open-loop transfer function in the absence of the servo. The magnitudes multiply and the phases add. Performing this yields the amplitude and phase behavior shown in Fig. 38.
Figure 38. Composite transfer functions (magnitude and phase) of the diode and cavity frequency responses. The purpose of the servo is to compensate for the excess phase lag introduced by these two loop elements.

From the figure, the composite phase of the cavity and diode exhibits a lag of $180^\circ$ by 1MHz. The locking bandwidth is typically limited to the frequency at which the closed loop phase lag exceeds $\sim 135 - 145^\circ$. The function of the servo is therefore to compensate for as much of this phase lag as possible in order to extend the locking bandwidth. This allows for more frequency noise reduction for all frequencies below the locking bandwidth. It should also be noted here that time delays such as excess cable length and optical path length also cause phase lag, but these sources are insignificant in comparison with the diode and cavity poles for Fourier frequencies.
Designing the Servo Electronics

With the open loop transfer function known, one can now properly design an electronic servo. The purpose of the servo is to compensate for the phase lags introduced by the cavity and the diode modulation. This will allow one to maximize the closed loop locking bandwidth. In general a wider locking bandwidth will yield a tighter lock to the HFC and better stability.

Perhaps the most straightforward servo one can envision would be simply an integrator circuit, which gives high gain to low frequencies (where most of the frequency noise exists) and whose gain falls at 20dB/decade for higher frequencies due to a single pole. Also, the phase for frequencies above the pole lags the input phase by $-90^\circ$ [104, page 101]. The simplicity of this servo is appealing, but when combined with the pole of the cavity, excessive closed loop phase lag already occurs for Fourier frequencies greater than the cavity half-width. The locking bandwidth is therefore limited to roughly the cavity half-width and this consequently limits the gain at lower frequencies as well. Any attempt to increase the gain will lead to closed loop instabilities.

In order to push the closed loop bandwidth beyond the cavity half-width, one must compensate for the cavity pole with a zero, or corner, in the servo response. With this added feature, the servo gain is again highest at low frequencies and falls
at 20dB/decade like the pure integrator, but this time the gain eventually levels off. Part A of Fig. 39 shows the measured frequency response of an integrator with a zero. Above the zero, the magnitude of the transfer function is flat and the phase eventually recovers by $+90^\circ$. The fall off of the phase around 10MHz is due to the finite bandwidth of the operational amplifiers used to construct the circuit. The frequency at which the zero occurs is often chosen higher than the cavity pole to optimize gain at lower frequencies [23, 114]. An integrator with such a zero will yield a locking bandwidth that exceeds the cavity half-width.

However, as one might expect, similar limitations are encountered when attempting to push the locking bandwidth past the pole imposed by the diode laser's modulation response. Indeed, when using a servo with a single zero, the locking bandwidths for the ECDLs analyzed in this thesis are typically limited to around 600kHz. This may be sufficient if the linewidth of the ECDL is significantly smaller than this frequency. To increase the bandwidth beyond this point, one must compensate for the second pole that is caused by the diode response. This involves placing yet another zero in the servo's response, leading to a $+20\text{dB/decade}$ slope and another phase lead of $+90^\circ$ at high frequencies. The measured response of such a phase-lead compensator is shown in part B of Fig. 39. The phase boost given by this stage is limited by the bandwidth of the chips.
Figure 39. The curves in part A show the measured response of just an integrator with a zero. Part B shows the that of a phase lead compensator. Part C shows the composite measured response of the servo.

The composite response of the integrator with a zero and a phase lead compensator is shown in part C of Fig. 39. The positions of the corners as shown in the figure are not optimized. In control theory, the behavior of this servo is known as proportional-integral-differential (PID) compensation [104, page 436]. When the corners are optimized, one can combine the closed loop responses of the diode, the cavity, and the servo to estimate the closed-loop transfer function.

A plot of the closed-loop phase as a function of Fourier frequency is shown in
Fig. 40. The benefit of the servo is immediately clear from this figure. The phase boost that was designed into the servo preserves the overall closed loop phase above the critical $\sim 145^\circ$ until beyond 1MHz. Consequently, a locking bandwidth that exceeds 1MHz is expected as well.

![Phase Response Graph](image)

Figure 40. The closed-loop phase response including the diode, the cavity, and the servo. The servo preserves the phase past 1MHz.

The closed-loop magnitude corresponding to this phase plot is shown in Fig. 41. The vertical scale has been adjusted to reflect the expected unity gain frequency (0 dB) of between 1 and 2 MHz. The area under the magnitude curve, but above unity
gain represents servo gain that will reduce the frequency noise of the diode laser. If the two corners in the servo would have been placed precisely at the cavity pole and the diode pole, respectively, then the magnitude would fall at 20dB/decade for all frequencies. However, by pushing the corners out to higher frequencies, the slope of the magnitude is steeper after the cavity pole, enabling increased gain at lower frequencies.

Figure 41. The closed loop magnitude response including the diode, the cavity, and the servo.
One very rarely finds actual servo circuit diagrams in the literature on frequency locking. In fact, the *only* full circuit diagram (including chips) that I know of in the mainstream literature on diode laser stabilization is that of Hilico, *et al* [115]. This is truly unfortunate because constructing a wide bandwidth servo circuit that performs as desired is very often the most challenging and time consuming aspect of frequency locking. The circuit that was used to generate the transfer functions that were shown in this subsection and that was for Raman locking is therefore given in Appendix M. I do not profess to be an expert at wideband servo circuit design and construction, but this circuit has worked well in practice. The corners were initially placed to generate a peak in the phase at the expected bandwidth of ~2MHz and were later adjusted slightly to optimize the locking.

**Slow Frequency Corrections**

Up to this point, only the fast frequency corrections via the injection current have been considered. The slow frequency corrections are performed using PZT elements that control the Raman cavity length. For this feedback path, a double integrator configuration is used, which enables higher gain at dc. OP27 op-amps are used for this feedback channel because they are reasonably quiet and exhibit large open-loop gains. A very low-noise, high-voltage amplifier (adapted from the Jan Hall group by Jay Brasseur) is also a critical element of this feedback channel.

The slow corrections are performed on the Raman cavity length rather than the
ECDL cavity length in order to counteract dc and low frequency fluctuations in the Raman cavity length due to thermo-optic effects (mode pulling) during lasing [29]. At dc, the Raman laser’s absolute stability is therefore determined by that of the ECDL, whereas at high frequencies, it is determined by the Raman laser cavity. Ideally, one would prefer to perform corrections on the Raman cavity for all frequencies for which heating-induced fluctuations occur (typically up to ~20kHz), but resonances in the PZT elements prohibit this. The PZT’s primary roles are therefore to compensate for heat-induced HFC length changes below ~2kHz and to keep the fast correction servo in the middle of its dynamic range. A nice discussion of using PZT elements in feedback loops is given in Ref. [108]. With the servo operating properly, one can now close the feedback loop and stabilize the ECDL to the Raman laser cavity.

Closing the Feedback Loop

With the essential elements in place, one can now close the feedback loop and lock the laser. One initiates the lock by scanning the HFC (or laser) and watching for the cavity transmission peak to broaden. This process is thoroughly described in Ref. [117], but a few unique aspects arise when locking to the Raman-active cavity.

First, when the HFC is filled with the Raman-active gas, the possibility exists for the system to lase. This will dramatically affect the cavity transmission and the error signal derived from the reflection when attempting to initiate and optimize the lock. One should therefore first establish and optimize the pump laser locking without gas
in the cavity.

With gas in the cavity, one should still initiate the lock with a pump power level that is below the laser threshold. Often, when the lock is initiated above the laser threshold, the sudden generation of Stokes light results in a sudden deposition of heat, and thus a sudden change in the cavity length. This can create unwanted and severe dynamics or it can prevent locking from occurring altogether. The heat-induced cavity resonance shift is typically several tens of MHz per mW of generated Stokes power (compared to a HFC linewidth below 1MHz).

When increasing the locked pump power from below threshold, the transmitted pump power clamps at threshold and the reflected power (from which the error signal is derived) no longer increases linearly with the incident pump power. Furthermore, the discriminator slope of the error signal is also modified above threshold [103]. As a result, the servo gain must sometimes be adjusted in order to maintain optimal locking above threshold.

One can envision avoiding such difficulties if the ring cavity is used. Specifically, one could use a small amount of light to lock to the HFC in, say, the clockwise direction of the ring. The majority of the light could then be used to Raman shift in the counter-clockwise direction of the ring. In this way, the pump power could be adjusted without changing the pump power used for locking. Also, one could envision this setup to perform interesting tests of the time dependent theory of the laser. With the cavity locked with the low power beam, the high power beam can be
quickly turned on (or off) to observe the start-up behavior.

When properly locked, the frequency noise of the ECDL is substantially reduced. Figure 42 shows a set of NSD measurements. Trace A shows the typical frequency noise spectra of a free-running diode laser, trace B shows that of a typical unstabilized Littrow-style ECDL, and trace C shows the same ECDL when it is actively stabilized to the Raman laser cavity using the servo circuit discussed above and given in Appendix M. This plot therefore highlights the benefits of frequency noise reduction via both passive (the external cavity) and active (electronic stabilization) means. The external cavity alone is seen to reduce the fast frequency fluctuations by about two orders of magnitude over the free-running diode. The roll-off in the noise spectra of all the traces above a few MHz is due to the measurement bandwidth and is not representative of the actual frequency noise.
Figure 42. Data showing the frequency noise spectral density as a function of Fourier frequency for (trace A) a typical free-running diode laser, (trace B) a Littrow ECDL, and (trace C) the same Littrow ECDL that is electronically stabilized to the HFC.

The reduction between trace B and trace C is due to the electronic feedback. This noise reduction is greater than two orders of magnitude out to several hundred kHz. The degradation of the signal fidelity at lower frequencies is due to the reduction of the laser’s frequency noise down to the background level of the rf spectrum analyzer. The locking bandwidth is defined by the Fourier frequency range over which the active locking reduces the frequency noise. This can be identified as the point at which trace B crosses trace C in Fig. 42. The measured locking bandwidth of ~ 1.5MHz matches the predictions from earlier in this section. The slight increase in the frequency noise
of the stabilized laser above the locking bandwidth is common and represents the beginnings of closed loop instability. If the servo gain is increased, this instability will grow.

**Injection Locking**

For cw Raman lasers, increased optical pump power is often desired beyond that which can be supplied by the ECDL alone, particularly for Littman-style ECDLs. This power enhancement can be achieved through the technique of injection locking.

Several concepts of injection locking can be implemented in diode-pumped Raman lasers. Most importantly, the optical and electronic frequency noise reduction performed on the ECDL (master) can be imparted to a free-running diode laser (slave) by simply injecting a small amount of the master's light back into the slave's output mode. Because the level of the injected light is small, the slave can still safely operate very near its maximum free running optical power level. The slave basically therefore operates as an amplifier for the master's output. Typical amplification factors are on the order of 20dB.

Another aspect of injection locking that can be used in cw Raman lasers is the fact that the amplification of the injected signal is phase sensitive. Therefore, when modulated within the injection locking bandwidth (as in the present case), phase modulation of the master signal can be imparted to the slave laser output. This was essentially first demonstrated in Ref. [118], although for dc phase changes. The
primary advantage of phase modulating the master signal rather than directly modulating the slave beam is to prevent optical damage of the EOM from a high power slave laser (if this is a concern) but also to save the optical power lost due to the imperfect EOM transmission. Furthermore, any unwanted amplitude modulations produced by the EOM (or from other sources for that matter) can be servoed out of the master signal without reducing the usable output power of the slave.

Injection of the high quality, phase modulated master output into the lasing mode of the slave can be accomplished in several ways. A particularly useful technique is diagramed in Fig. 43 and warrants discussed here. During normal operation, the combination of a polarizing beam splitter (PBS), a half-wave plate \((\lambda/2)\), a Faraday rotator, and another PBS can serve as a Faraday isolator as shown in the figure. This configuration allows all the light from the slave to be directed towards the Raman cavity subsystem, but it also allows light from the pump laser subsystem to be injected into the discarded port of the isolator (the second PBS) and sent to the FRDL for injection locking.
Figure 43. Setup for the injection locking subsystem.

To align the injected beam, one can rotate the half-wave plate so that all the light from the slave is alternatively directed back towards the pump laser subsystem (against the arrows shown in Fig 43). This light can be coupled into the PM fiber end from which the pump laser light is emitted. With the master laser optimally coupled into one end of the fiber and the slave into the other, the injected light is automatically aligned during normal operation (when the half-wave plate angle is returned to its original position). Furthermore, the coupling efficiency of the slave laser into this fiber during alignment can be used to estimate the efficiency with which the master’s output is injected into the slave diode’s spatial mode during normal operation. Other methods of determining the injected signal coupling efficiency have been demonstrated (see, for instance, Ref. [118]), but were not employed here. The use of the optical fiber
removes the question of spatial mode matching during the injection locking process, leaving only the matter of spectral mode matching.

Spectral adjustment of either the slave or the master must be performed in order to achieve injection locking. In theory, one must adjust the frequency difference between the master and the slave to within the injection locking bandwidth. This is typically several GHz, but depends on many factors for diode lasers [119]. In practice, as shown in Fig. 44, it is possible to achieve injection locking when the two frequencies differ by much more than the injection locking bandwidth. Traces A and B in the figure show the spectral output from a free-running slave laser with no injected signal, while trace C shows the output of the same laser when light is injected several nanometers below its free-running wavelength. The >40dB mode suppression ratio observed for the injection locked trace is the same as that of the master laser. An injected power ratio of less than 1% was used to generate the injection locking shown in trace C.
Figure 44. Traces A and B in the figure show the spectral output from a free-running slave laser with no injected signal, while trace C shows the output of the same laser with an injected signal.

To find a suitable injection locking condition, one can simply tune the master laser's wavelength while viewing the output from the slave laser on an optical spectrum analyzer. One will observe a sharp drop in the mode suppression ratio (to the master's level) when the injection locking is achieved. If one wishes to injection lock very near the free-running diode's frequency, then both the master and the slave frequencies may need to be adjusted and monitored using a spectrometer with better than 1GHz resolution, such as a Burleigh Wavemeter.

Once the slave laser is injection locked, one can direct the slave's output to the
Raman laser cavity. One can verify that the phase modulation sidebands on the master are effectively transferred to the slave by scanning the Raman cavity length. From this point, the active locking procedure is performed as if only the ECDL were being locked. That is, fast corrections are performed on the master laser injection current. The resulting frequency noise reduction is efficiently imparted to the slave. In this sense, the master laser itself can be viewed as a frequency actuator in the control loop. Fortunately, the transfer function for this portion of the control loop is typically very flat over the frequencies of interest.

It is also possible to use current modulation of the slave laser as an alternative to the EOM. Under free running conditions (without an injected signal), modulation of a diode laser's injection current results in frequency (and amplitude) modulation of the output for low modulation frequencies. However, Kobayashi et al. demonstrated that current modulation within the locking bandwidth of an injection locked slave laser results in phase modulation of the output [118]. The sidebands resulting from such modulation can be used in place of those generated by an EOM. The price paid is an amplitude modulation coupled to any current modulation of the diode. These cannot be removed without reducing the slave output power.

Passive Optical Stabilization

This section describes several different schemes that were investigated for optically locking diode-pumped cw Raman lasers. The concepts behind optical locking were
discussed in the first section of this chapter.

The ideas of Dahmani, et al. [109] are implemented for Raman lasers by using resonant optical feedback from the Raman HFC itself for the pump laser stabilization. In this way, the HFC serves two functions; it acts as a spectral filter for the optical feedback, and it provides the optical power build-up for cw Raman conversion. In contrast to the work of Dahmani, et al., only non-confocal HFCs are used for cw Raman generation. The Raman configurations therefore differ slightly, but the basic ideas are the same. Furthermore, rather than sending a small amount of the diode’s emitted light to the cavity for locking, as much of the diode laser’s light as possible is coupled into the HFC in order to maximize the pump power available for Raman conversion. Even though most of the light entering the HFC is typically reflected off of the input coupler of the cavity or converted to Stokes, a small portion of the pump light is still transmitted through the HFC and can be fed back to the diode laser source for locking. Feedback levels less than 1 part in $10^5$ of the emitted diode power are sufficient for optical locking. Due to the miniscule feedback levels, essentially all of the diode laser’s free-running optical power can be devoted to generating Stokes light.

The first configuration employs the transmitted field of a linear cavity. A conceptual schematic showing only the essential elements is provided in Fig. 45. More detailed experimental setups are shown in the next chapter. A similar configuration was previously demonstrated for second harmonic generation [135]. When the
transmitted optical field is injected into the discarded output port of the Faraday isolator (the second PBS), it returns to the diode laser source for passive locking. The injection of this filtered light narrows the diode laser’s linewidth substantially and provides center frequency stabilization to an HFC resonance. Although such systems will operate for short periods of time (several minutes) with no active locking whatsoever, additional low-bandwidth electronic stabilization is required for long-term operation. This is necessary because of the system’s sensitivity to thermal, acoustic, and barometric perturbations to the overall path length.

Figure 45. Possible setup for the passively-locked cw Raman laser.

The half-wave plate within the Faraday isolator serves essentially the same role as
in the injection locking setup. During normal operation, light propagates in the clockwise direction around the loop as indicated by the arrows. However, for alignment purposes, the half-wave plate can be rotated to direct the output from the second isolator in the opposite direction. This light can be coupled into the HFC, which results in automatic alignment of the optical feedback during normal operation. When compared to the confocal cavity locking introduced by Dahmani, the on-axis linear resonator unfortunately requires optical isolation to reduce non-resonant optical feedback that returns from the front of the HFC. On the other hand, the non-confocal configuration allows for one to adjust the feedback level independent of the input light.

Configurations more similar to that of the confocal technique were also investigated for Raman conversion. For instance one can passively lock the pump laser to higher-order spatial modes of the linear cavity. This method can be achieved by tilting the HFC optic axis with respect to the input beam. The input beam is reflected at a small angle, so isolators can be eliminated, but the resonant higher-order spatial mode transmits some light back along the beam path to the diode laser for locking. A three-mirror “V” cavity geometry can also be used. This is very similar to the confocal cavity locking. Again, only the light that resonates within the cavity returns to the diode laser. Both of these configurations have been used to generate Stokes laser light in the fundamental spatial mode of the HFC. However, although these two methods boast even further simplicity, they are inherently inefficient for producing
Stokes because of the poor impedance matching.

The last configuration that has been used for optical locking was discovered unintentionally. When actively locking the bow-tie ring style HFC, resonant optical feedback from the ring back to the pump laser along the beam path was observed where none was expected to exist. This feedback was used to efficiently generate Stokes light with a setup composed of only a free-running diode laser, a pair of coupling lenses, and a pair of mirrors for mode matching. The ring cavity geometry need not suffer from poor impedance matching. The source of the unexpected resonant feedback was not investigated, but may be due to four-wave-mixing within the cavity, which could generate a counter-propagating field.

Despite some advantages in available optical power and simplicity, the optical locking techniques were only used to generate Stokes light for relatively short periods of time (several minutes) without additional active electronic stabilization. Furthermore, continuous frequency tuning, which is crucial for these systems, was difficult for the optically-locked system. On top of these drawbacks, it was also discovered that nonlinear thermo-optic processes within the Raman cavity interacted with the transmitted pump light and led to system instabilities for the narrow linewidth HFCs ($<200$kHz FWHM). Nevertheless, the passive locking method was used to generate the first cw Raman laser and the first high-efficiency Raman laser.
Chapter Summary

This chapter has been devoted to describing the practical aspects involved with frequency locking a diode laser to an active high-finesse Raman cavity. The three essential elements of the active feedback loop, the pump laser, the Raman HFC, and the servo electronics, were discussed in some depth. As a fourth, optional element, it was shown that the injection locking technique can be implemented to boost the amount of optical power that can be delivered to the HFC for Raman conversion. Finally, a method of passive optical locking was also discussed. While this chapter discussed the intricacies of constructing a cw Raman laser, the next chapter documents experimental Raman laser results that can be obtained using the techniques discussed above.
CHAPTER 6

EXPERIMENTAL RESULTS

This chapter is devoted to documenting the experimental results associated with the creation and development of diode-pumped Raman lasers. Much of this work was conducted with my labmate Lei Meng. I began with the optical locking technique and Lei began with the electronic locking technique. However, we soon combined forces to the benefit of both. The injection locking experiments highlight this alliance. I quickly grew to appreciate and prefer the electronic locking due to the superior level of control that can achieved through a single feedback loop. Thus, the earlier experiments presented in this chapter were performed using optical locking and the later ones using electronic locking. This is also the reason that the lion’s share of the previous chapter was devoted to describing the electronic locking procedure.

This chapter begins with a review of the previous cw Raman laser experimental work that was conducted prior to the advent of diode pumping. This work was based entirely on a frequency-doubled Nd:YAG-pumped system [44]. In the second section, results from the first experimental demonstration of diode-pumped cw Raman lasing are presented. The basic theory that was developed in Chapter 2 or the steady-state limit of the quantum results that were derived in Chapter 3 are shown to accurately describe the behavior of this initial low-power system. In the third section, results
from a similar system are used to demonstrate the first extraction of greater than 50% photon conversion efficiency and high output power from a cw Raman lasers by using an impedance matched laser cavity. However, the behavior of this high-power system is shown to deviate from the steady-state theories presented in Chapters 2 and 3. In the fourth section, the technique of injection locking is shown to substantially increase the pump power available for Raman conversion. The value of the technique is demonstrated by pumping a low-power Raman laser to more than 90 times threshold. In the fifth and sixth sections, measurements of thermal mode pulling and lensing in the Raman cavity are compared with the predictions of the thermal models presented in Chapter 4. The deviations observed in the behavior of the high-power system are shown to be caused by thermal lensing. In the seventh section, results from the first cw Raman ring laser are given. The ring system is shown to not only exhibit stable operation in the forward direction, but also to reduce the deleterious effects of thermal lensing and reduce optical feedback to the pump laser source.

**Previous Work**

Although this thesis documents the creation and development of diode-pumped cw Raman lasers, many of the general characteristics of cw Raman lasers were previously studied using a frequency-doubled Nd:YAG laser. In particular, Brasseur, *et al.* demonstrated the first cw Raman laser in 1998 [26, 27]. The researchers used a highly-stabilized, 200 mW frequency-doubled cw Nd:YAG laser at 532nm as the
pump source to produce Stokes laser output at 683nm. The cavity finesse was over 13,000 for both the pump and Stokes wavelengths. This initial system exhibited a Stokes laser threshold below 1 mW as predicted, thereby demonstrating the possibility of pumping with low-power diode lasers. A peak conversion efficiency of 27% and a peak Stokes output power of 2.5mW were also demonstrated. The peak in the conversion efficiency occurred for pump rate of four times threshold, as predicted. However, the peak value was fundamentally limited to under 50% by poor impedance matching because identical front and back mirrors were used in the Raman laser cavity (see Chapter 2). The system's time-dependent behavior was measured and compared to theory [27]. Furthermore, measurements of the continuous tuning [28] and linewidth [27] were also performed. The continuous tuning was compared to theory and it was suggested the Stokes linewidth of 10kHz was due to the mechanical stability of the Raman laser cavity. Finally, a very small amount of axial coherent anti-Stokes emission at 436nm was observed for the first time using this system and was compared to theory [30]. The Raman processes are no different when pumping with diode lasers, but the frequency locking differences are substantial, as discussed in the previous Chapter.

Diode-Pumped Raman Lasing

The first diode-pumped Raman laser was created using the passive optical feedback frequency locking method and a linear high-finesse Raman laser cavity (HFC) [32].
This system not only confirmed that it was possible to Raman shift low-cost, tunable diode lasers, but it also demonstrated to my knowledge the lowest threshold of any Raman lasing process at that time. Microsphere resonators have since exhibited lower thresholds [136]. The optically-locked system also exhibited a small amount of coherent anti-Stokes radiation.

The experimental setup is shown in Fig. 46. The hydrogen-filled HFC was pumped by a free-running diode laser at 790nm. This laser source was a temperature stabilized, 100mW Fabry-Perot type device with <5% front facet reflectivity. A Faraday isolator attenuated non-resonant reflected light by ~35dB. A half-wave plate and polarizer were used in tandem as a variable attenuator for the pump. Mirror M1 is coated to reflect only the pump wavelength, thereby allowing transmission of Stokes (1178nm) and anti-Stokes (595nm). The HFC was described in Chapter 5 and consisted of two curved mirrors of radii +50cm, separated by 7.62cm in a linear-cavity geometry. The mirrors were coated for very high reflection and low loss at both the pump and Stokes wavelengths. Using the manufacturer-quoted reflectivities, the finesse at each wavelength was about 60,000. The mirrors were enclosed within a hermetically sealed container filled with 10 atmospheres of diatomic hydrogen gas.
Figure 46. Passively locked experimental setup used to demonstrate the first diode-pumped cw Raman laser. LD = laser diode, APP = anamorphic prism pair, GP = Glan polarizer, FR = Faraday rotator, λ/2 = half-wave plate, PBS = polarizing beam splitter, EOM = electrooptic modulator, BS = beam splitter, MML = mode matching lenses, M = mirror, PBP = Pellin-Broca prism, and HFC = high finesse cavity. Mirror M1 is reflective coated only for the pump wavelength.

The cavity transmission was fed back to the diode laser through the discarded port of the output polarizer in the Faraday isolator. The feedback power could be adjusted using another half-wave plate in the transmission leg before this polarizer. Assuming lossless coupling back into the diode’s spatial mode, the ratio of optical feedback power to the laser diode emission power was ∼10⁻³ for the data presented in this section. Later versions of this optical feedback setup used a half-wave plate within the Faraday isolator as shown in Fig. 45 so that automatic alignment of the feedback could be achieved.
Stable locking could be obtained from this setup for short (≤30s) durations with no further stabilization. The limited lock duration was due to the dependence of the optical locking on the (interferometrically unstable) round-trip optical phase of the feedback. To provide further stabilization, the HFC mirror separation was electronically servoed via the Pound-Drever-Hall (PDH) method [23]. Stable locking could also be achieved by controlling mirror M3 with a PZT. For the electronic locking, an electrooptic modulator (EOM) placed 10 MHz sidebands on the carrier frequency to produce an error signal. A 75/25 beam splitter (BS2) was used to divert a portion of the reflected beam from the front of the HFC in order to detect the error signal for locking.

Figure 47 shows the results from this initial optical locking setup. In the figure, the transmitted pump and output Stokes optical powers are plotted as functions of the coupled input pump optical power. The solid lines represent the theoretical steady-state predictions from either Chapter 2 or 3 (see the end of Appendix B for discussion of the linear cavity) using the following parameters: \( \lambda_p=790\text{nm}, \lambda_s=1178\text{nm}, P_1 = 28.8\text{kW}, R_p,rt=0.99986, R_s,rt=0.99990, T_{p,0} = T_{p,1}=58\text{ppm}, T_{s,0} = T_{s,1} = 22\text{ppm}. \) The manufacturer-quoted mirror reflectivities are 0.99995 for both the pump and Stokes wavelengths. The plane-wave gain coefficient, \( \alpha \), that was needed to calculate \( P_1 \) was obtained from Ref. [137].
From Fig. 47, one can see that the transmitted pump power increased linearly until the Stokes laser threshold at 240±19mW. For input pump powers greater than this, the transmitted pump power abruptly clamped as predicted. The observation of lasing with this low threshold confirmed the possibility of Raman-shifting low-power (<10mW) diode lasers. The error in the reported threshold value is not stochastic, but instead comes from uncertainty in the coupling efficiency into the fundamental cavity mode.

The Stokes emission behaved similar to that observed in the Nd:YAG-pumped
system. Namely, it increased monotonically with the characteristic $\sqrt{P_{\text{inc}}}$ dependence after threshold. The Stokes polarization matched that of the input pump. With reasonable HFC mode matching, optical feedback assured locking to only the fundamental HFC Gaussian mode for the pump (the transmission of the higher-order spatial modes was very low). As expected from the HFC, high spatial quality was observed in both the transmitted pump and the output Stokes beams. Stokes emission was observed over a discontinuous wavelength range of ~10nm by changing the resonator cavity length while holding the diode laser drive current and temperature fixed.

At higher pump powers, coherent anti-Stokes emission at 595nm could easily be observed with the naked eye. However, intensity dependent phase instabilities caused output oscillations that were beyond the locking bandwidth at these higher powers. The oscillations were later discovered to be due to thermo-optic effects associated with the Raman lasing. The instabilities were only observed when the HFC resonance was narrow (<50kHz FWHM), suggesting that the resulting narrow-bandwidth optical feedback played a role. As discussed in the next section, stable high-power operation was observed for cavities with larger linewidths (~1MHz).
Figure 48 shows the measured and predicted photon conversion efficiencies from the input pump to the output Stokes beam as a function of the input pump power. The maximum conversion efficiency observed was 15%, but the system was predicted to reach a peak value over 17% at four times the threshold pump power. An actively-locked ECDL was later used to confirm the behavior of this low-power system [138]. The ECDL could be locked stably to the HFC for any input pump power level and demonstrated broad discontinuous tunability of the Stokes output.
The ECDL-pumped system also exhibited a broader discontinuous tuning range. The reflectivities of the mirrors used in these initial experiments were selected to minimize the Stokes laser threshold rather than to optimize the photon conversion efficiency. In general, poor impedance matching fundamentally limits a symmetric-mirror cavity to a maximum of 50%.

High-Efficiency Raman Lasing

The optically-locked system was next used to demonstrate the first high-efficiency cw Raman laser [139]. It was shown theoretically in Chapter 2 that the conversion efficiency of a Raman laser can only exceed 50% if the HFC impedance matching is improved. The challenge of impedance matching is not unique to the cw Raman laser; it is confronted in every form of cavity-enhanced frequency conversion. To improve impedance matching, one can simply reduce the reflectivity of the HFC input coupler relative to the other cavity losses.

The setup for this experiment is shown in Fig. 49. It is similar to the previous setup except that the reflectivity of the front (input coupler) mirror at the pump wavelength was reduced. This not only made the Raman process more efficient, it also made the optical locking significantly more stable because the optical feedback bandwidth was greater with the broader cavity linewidth. An additional Faraday
isolator was included, which allowed the error signal to be measured without attenuating the beam while retaining linear polarization. Also, a single-mode, polarization-maintaining optical fiber was used as a spatial filter. This made coupling into the HFC easier and it allowed a more accurate determination of the conversion efficiency because almost all of the light from the fiber was coupled into the cavity mode.

Figure 49. Passive locking experimental setup used to demonstrate the first high-efficiency cw Raman laser. See the caption of Fig. 46 for symbol definitions.

The experimental results obtained from this setup are shown in Figs. 50 and 51. Fig. 50 shows the generated Stokes power (right axis) and the corresponding photon conversion efficiency (left axis) as functions of the coupled pump power. The threshold was significantly higher (~17mW) than for the data shown in the last section because of the reduction in the input coupler reflectivity. However, due to the related improvement in the impedance matching, the system exhibited a peak output Stokes
power of ~14mW and a peak photon conversion efficiency of ~61%. This system therefore provided the first experimental confirmation that the conversion efficiencies of cw Raman lasers could surpass 50% through the use of an impedance-matched laser cavity.

The system also provided the first indications that the basic steady-state cw Raman laser theories were insufficient for high-power systems. The theoretical predictions for the system are shown as lines in the figure. The system’s behavior departed from the strictly square root dependence on the input pump power. It produced less Stokes, and therefore it exhibited poorer photon conversion than predicted. The fit parameters used were: $\lambda_p=792\text{nm}$, $\lambda_s=1180\text{nm}$, $P_1 = 28.5\text{kW}$, $R_{p,rt}=0.9969$, $R_{s,rt}=0.9993$, $T_{p,0}=2950\text{ppm}$, $T_{p,1}=26.5\text{ppm}$, $T_{s,0} + T_{s,1} = 690\text{ppm}$. 
Figure 50. Plot showing the output Stokes power and photon conversion efficiency from the impedance-matched Raman high-finesse cavity as functions of the coupled pump power. This system exhibited \( \sim 14\text{mW} \) of output Stokes power, \( \sim 61\% \) photon conversion efficiency, and it provided the first indication that the basic Raman laser theory was insufficient for high-power systems.

More convincing data showing the inadequacy of the basic cw Raman theory is shown in Fig. 51. This figure shows the reflected (left axis) and transmitted (right axis) pump powers from the HFC. The reflected pump power displayed a characteristic decline after threshold, indicating that more light entered the HFC for Raman conversion due to the impedance matched mirrors. However, the decline is not as severe as expected. It should be noted that the non-unity cavity coupling efficiency was included in the calculation this curve. Furthermore, the transmitted pump power
did not clamp as it had for the lower power system. Instead, the transmitted pump power continued to rise after threshold as a function of incident pump power.

![Graph](image)

**Figure 51.** Reflected and transmitted pump powers from the impedance-matched Raman laser cavity as functions of the coupled pump power. The behavior of this system provided the first clear indication that the basic Raman laser theory was insufficient for high-power systems.

These departures from the predicted behavior will be shown later in this chapter to be caused by thermal lensing within the Raman laser cavity. This system was therefore not only instrumental in demonstrating that high-efficiency cw Raman lasing was possible, it also revealed an inadequacy of the basic cw Raman laser theory for
Injection-Locked Raman Lasing

At the time when the high-power data was first measured, it was questioned whether the departures of the measured data from the basic theory were in some way related to the optical locking. Unfortunately, the high-efficiency Raman laser threshold was too high to be accessed by an actively-locked ECDL. After isolation, modulation, and fiber coupling, the Littman ECDL that was used at that time could deliver only about 10mW to the front of the HFC. Only the optical locking technique could reach the required optical powers for the high-power system. Therefore, the technique of injection locking was used to amplify the optical power of the ECDL [140].

Details of the injection locking technique for Raman lasers are discussed in Chapter 5. Figure 52 shows the experimental injection locking setup used. The master laser was a temperature-controlled ECDL in Littman configuration operating at 792nm with ~30mW maximum output power. A commercially available 100mW diode laser with a standard 3-5% anti-reflection coating was used inside the external cavity. The ECDL was discretely tunable over a range of nearly 20nm and continuously tunable over a 20GHz range. A nearly 50dB side mode suppression ratio was observed from the ECDL output.
The elliptical beam shape from the ECDL was corrected by an anamorphic prism pair, the beam was isolated (~35dB), and sent through an electrooptic modulator (EOM) to place 12 MHz phase modulation sidebands on the carrier for locking purposes. The phase modulation sidebands of the master signal were imparted to the slave laser output, as discussed in Chapter 5. A half-wave plate and polarizing beam splitter were used to adjust the optical power sent to the free-running slave laser via a single-mode polarization-maintaining optical fiber. The unused optical power from the beam splitter was sent to an optical spectrum analyzer (OSA) to monitor the spectral characteristics of the master laser.

The slave laser was the same laser used for the optical locking technique described above. The slave was operated slightly below its maximum output power capability to account for the injected light. The combination of a half-wave plate, a Faraday
rotator, and two polarizing beam splitters served as an optical isolator. However, the configuration also permitted the alignment of the injected signal as discussed in Chapter 5. When the slave’s output was directed towards the HFC, the injected master signal could sneak through the isolator unattenuated. The injected power was \(~1\text{mW} ~(\sim1\%\ of\ the\ slave’s\ output\ power)\), which gave a typical injection locking bandwidth of greater than the 5GHz. After this isolator configuration, a half-wave plate was placed before a final isolator and used as a variable attenuator for the pump power. The discarded beam was sent to the OSA for spectral analysis.

As a first test of the injection locking technique, identical front and back mirrors were used in a linear HFC. Thus the threshold was expected to be low as with the first diode-pumped system, but the conversion efficiency was expected to be poor. The laser’s frequency was stabilized to a HFC resonance using the PDH method. The phase sidebands on the slave were used to generate an error signal that was reflected off the input coupler of the HFC. This signal was mixed down to DC and fed to the servo for amplification and filtering. To maintain resonance within the cavity, slow (DC to 1.5 kHz) frequency corrections were performed on the cavity resonance frequency by changing the HFC mirror spacing via the PZT tube spacers while fast (DC to \(~600\text{kHz})\) corrections were performed on the master laser’s injection current (thereby correcting the slave laser’s frequency).

Figure 53 shows the experimental results from the injection-locked Raman laser setup. The figure shows the output Stokes power at 1182nm (circles), the transmitted
pump power (diamonds), and the photon conversion efficiency (squares) as functions of coupled input pump power. Theoretical predictions from Chapter 2 or 3 are also shown as solid lines in the figure. The Stokes laser threshold was measured to be $330 \pm 20$ mW. Excellent agreement with the basic theory was observed for pump powers well above threshold for this low-power system. In fact, the figure shows that a pump rate of greater than 90 times threshold was achieved using this injection locking technique. This represented more than a three-fold increase in the input and output optical powers relative the case when the ECDL is used alone. The thermal effects were not evident because the amount of Stokes power generated was still small. The fitting parameters are given in Ref. [140].

The injection locking technique therefore succeeded in substantially increasing the pump power available for Raman conversion. I am unaware of any other laser system that has exceeded a pump rate of 90 times threshold. The system also demonstrated that the basic Raman laser theory continued to accurately describe the laser's behavior at these high pump rates when the Stokes power remained low (below several mW).
Figure 53. Plot showing the output Stokes power (circles), transmitted pump power (diamonds), and photon conversion efficiency (squares) as functions of the coupled input pump power. The injection locking technique enabled a pump rate of 90 times threshold to be achieved.

Lei Meng took the injection locking technique one step further by using a broad-area diode laser as the slave. Coupling the light into this slave is substantially more involved and is discussed in Ref. [141]. The use of a tapered diode amplifier as the slave laser is also suggested for higher power applications, but these can be very difficult to obtain commercially.

Using the exact setup shown in Fig. 52, but with an impedance matched cavity, the aberrant measurements shown in Figs. 50 and 51 were confirmed [34]. This ensured that the deviations from the basic theories were not due to the optical locking
Indeed, the next section shows that the cause of the deviations was of a more fundamental nature.

**Raman Laser Thermal Mode Pulling**

As discussed in Chapter 5, cw Raman lasers rely heavily on the relative phase and frequency stability between the pump source and the Raman laser cavity. However, frequency locking instabilities were observed above the Stokes laser threshold for both the first Nd:YAG-pumped system [27] and the first diode-pumped system [32]. Because the instabilities were qualitatively correlated with the existence of Stokes power, an intensity dependent refractive index within the Raman laser cavity was a probably culprit.

An experiment was therefore devised to measure the refractive index of the hydrogen gas as a function of the input and generated optical powers. The concept behind the measurement is quite simple and takes advantage of the fact that the active locking is extremely sensitive to changes in the HFC optical path length. When the refractive index within the HFC changes, the optical path length between the mirrors also changes. As a result, the resonant frequencies of the cavity are shifted. When the Raman laser is locked to a resonance of the HFC, this mode pulling is compensated by the PZT elements that control the physical cavity length. Therefore, changes in the *optical* path length between the HFC mirrors due to refractive index shifts are manifest as changes in applied PZT voltage as the system adjusts the
physical path length between the mirrors in order to maintain resonance within the optical cavity.

The experimental setup is shown in Fig. 54. For this experiment, a frequency doubled Nd:YAG laser operating at 532nm was used because of its superb long-term frequency stability. The pump source could be temperature tuned over a range of roughly 10 GHz. Two optical isolators were used to minimize optical feedback resulting in ~60dB attenuation of light returning along the beam path. The EOM placed 12 MHz sidebands on the carrier frequency for locking purposes. A half-wave plate and polarizer were used to produce a variable attenuator for the pump. The laser’s frequency was stabilized to a HFC resonance using the Pound-Drever-Hall method. To maintain resonance within the cavity, fast ($\lesssim 200$kHz) frequency corrections were performed on the Nd:YAG laser output by double-passing an AOM. Slow ($\lesssim 1$kHz) corrections were again performed on the cavity resonance frequency by changing the HFC mirror spacing via the PZT tubes.
Figure 54. Experimental setup used to measure the intensity-dependent thermal mode pulling. Thermally-induced shifts in the Raman laser cavity length (HFC) were manifest as voltage applied to the PZT elements.

During the measurement, the Stokes output power and PZT voltage applied by the servo were monitored as functions of the input pump optical power both below and above the Stokes laser threshold. In order to minimize the effects of long term PZT and temperature drifts that were not associated with the optical power, reference data points were taken between each of the signal data points at a fixed pump power level well below threshold. The slow background drifts evident in the reference data were removed from the signal data leaving only the effects associated with the optical power. The time between successive measurements was ~12s so steady-state conditions were expected to prevail [36]. To ensure that the PZT movement was not in response to DC frequency shifts generated by the AOM, the pump laser’s frequency
was monitored with 10MHz resolution after the AOM as shown in Fig. 54.

Figure 55 shows the resulting voltage applied to the PZT as a function of the pump optical power coupled into the HFC when the laser frequency was tuned to the line center of the \( \sim 610 \text{ MHz (12 atm)} \) Raman resonance. A clearly defined change in behavior can be observed around \( \sim 850 \text{mW} \), which corresponds to the Stokes laser threshold. Note that above this threshold, the intra-cavity pump power remains nearly constant. This established that the dominant cause of the index change was related the Stokes optical power.

![Figure 55. Plot showing the measured PZT voltage versus the pump optical power. A clear behavioral change is observed at the Stokes laser threshold (840\text{\mu W}).](image)
Therefore, Fig. 56 shows the resulting cavity resonance shift as a function of the generated Stokes power for the cases when the cavity was tuned to the line center ($\Delta_{12}=0$) and to the positive half-width ($\Delta_{12}=305$MHz) of the Raman resonance. The solid lines represent linear fits to the measured data for Stokes power greater than zero. From these slopes, the index change with Stokes power is observed to be independent of detuning to within the measurement uncertainty. Detuning the cavity to the negative half-width of the Raman resonance yields statistically indistinguishable results from that on the positive frequency side. This indicates that the observed effect is dominated by thermally induced index changes rather than by dispersion associated with the Raman resonance. For the latter case, the sign of the index change for the two detuned data sets would be reversed with respect to one another.
Figure 56. Experimental data showing the mode pulling as a function of the generated Stokes optical power.

The slopes of the linear fits to this data indicate mode pulling toward the blue of ~35MHz per mW of generated Stokes power. This is significant when compared to the typical HFC linewidth of less than 1MHz. If the frequency locking performs slow corrections on the laser’s frequency, then this mode pulling will tune both the pump and the Stokes emission frequencies with Stokes power. For this reason, one often chooses to use PZT’s within the HFC itself for the slow portion of the feedback loop in order to compensate for the mode pulling.

Also included in Fig. 56 are the theoretical predictions (dashed lines) based on the
thermo-optic model presented in Chapter 4. Two curves are given for two different values of the constant temperature surface radius, \( r_c \). The two radii reflect the actual HFC geometry, which consists of two concentric aluminum cylinders with inner radii of 1.2cm and 2.4cm respectively, neither of which are temperature stabilized. The theoretical model neglects convection in the gas and heat transfer between the gas and the mirrors.

A more subtle index shift that was associated only with the pump power is also evident in Fig. 56. Specifically, the linear fits to the measured data do not include the origin within measurement uncertainty. This characteristic was common to all data taken regardless of location on the Raman resonance and indicates that some mode pulling occurs before the Stokes laser threshold, due only to the input pump power. The detuned case displays a higher vertical axis intercept. This can be explained by noting that the Stokes laser threshold for the detuned case requires twice the input pump power as that for the line center case due to decreased Raman gain. For input pump powers greater than the Stokes laser threshold value, the intracavity pump power remains nearly constant. Figure 57 shows the mode pulling as a function of input pump power below the Stokes laser threshold for the two different locations on the Raman resonance. Again, the linear slope of the induced index change does not depend on detuning within measurement uncertainty. Crude estimates indicate that the source of this pulling may be heating of the high-finesse mirrors. The two lines represent linear fits to the measured data.
Figure 57. Experimental data showing the subtle mode pulling below threshold as a function of the coupled pump optical power.

Another effect of the heating can be observed by simply scanning the cavity length without frequency locking if the laser's inherent linewidth is much narrower than that of the cavity. Figure 58 shows the transmitted pump power for backward and forward scans of the cavity when the input pump power level is above threshold. Hysteresis is clearly evident between the two scan directions. When the cavity length is decreased during a backward scan ($\leq 1.7\text{ms}$), Stokes light is generated, which pulls the cavity mode towards the blue (i.e. decreases the optical cavity length). This effectively
accelerates the scan rate and results in a narrow (in time) transmission from the cavity. On the other hand, when the physical cavity length is increased (>1.7ms), the optical path length still decreases due to the drop in refractive index. This time, the cavity scan and the mode pulling partially counteract one another allowing the injected laser light to stay in resonance with the cavity longer. Depending on the scan rate and the Stokes power generated, one can observe oscillations in the transmitted power due to interplay between the changing index and the Lorentzian bandpass of the cavity, as shown in Fig. 57. Similar thermally-induced hysteresis has been previously observed in other nonlinear systems.

![Graph showing cavity length, pump, transmission vs. time](image)

Figure 58. Plot showing the measured transmitted pump power and the Raman laser cavity length while scanning the cavity and not locking.
As one might expect, the thermo-optic nonlinearities also affect the frequency locking of Raman lasers. This is discussed in Ref. [103]. The cavity length instabilities caused by the thermal mode pulling alter the discriminator slope and typically necessitate a minimum locking bandwidth of at least 100kHz, even when the inherent frequency noise of the pump laser requires less. One should also consider the dynamic range and slew rate requirements imposed by the mode pulling on the frequency actuators for locking.

Raman Laser Thermal Lensing

The thermal mode pulling that was just discussed is a significant concern for frequency locking the pump laser source and for the absolute frequency stability of the Raman system. However, there is also another significant effect of the Raman heating. As discussed in Chapter 4, the heating profile within the gas is radially dependent as a result of the radial dependence of the optical beams. As a result, the refractive index varies radially from the beam axis as well. This effectively generates a diverging thermal lens within the cavity, whose focal length depends on the generated Stokes optical power. This can dramatically affect the steady-state behavior of the Raman system when significant Stokes power is generated.

Figures 59 and 60 shows high-power data taken using the injection locking setup shown in Fig. 52 with an impedance-matched cavity. The dashed lines show the
steady-state predictions of the basic theory presented in Chapters 2 and 3. As observed with the optically-locked data, the reflected pump power was greater than predicted, the transmitted pump power no longer clamped above threshold, and the Stokes generation was poorer than predicted.

![Graph showing transmitted and reflected pump powers](image)

Figure 59. Transmitted (circles) and reflected (squares) pump powers from a high-power Raman system. The thermo-optic theory significantly improves the agreement with the measured data.

The solid lines show the modified steady-state theory when the thermal effects are included. Significant improvement in the agreement between the data and theory can be observed. The thermo-optic modifications should be included when greater than 5-10mW of Stokes power is generated for the 3” Raman cavity. To mitigate the
thermal lensing, one can either increase the cavity length or provide multiple passes (as with the ring cavity) to distribute the heat over a greater volume within the laser cavity. The rise in the transmitted pump power provides an excellent gauge for the significance of the thermal lensing. For the data shown here, the transmitted pump power rises ~1% for every mW of Stokes power generated. The fitting parameters used were: $\lambda_p=792\text{nm}$, $\lambda_s=1180\text{nm}$, $P_1 = 28.6\text{kW}$, $R_{p,rt}=0.99697$, $R_{s,rt}=0.99926$, $T_{p,0}=2910\text{ppm}$, $T_{p,1}=25\text{ppm}$, $T_{s,0} + T_{s,1} = 620\text{ppm}$, $L = 7.6\text{cm}$, $R = 50\text{cm}$, $K_{th} = 2.18\text{mW/cm-K}$, $n_0 = 1.001276$, $\tau = 38\mu\text{s}$, $D = 0.1443\text{cm}^2/\text{s}$.

![Graph](image.png)

Figure 60. Plot showing the output Stokes power (circles) and photon conversion efficiency (squares) as functions of the incident pump power. The reduction in the Stokes output is due to the thermal lensing.
Although the improved fitting with the steady-state power data was convincing, the thermal lensing was more directly verified by experimentally measuring the emitted beam radius as a function of the generated Stokes power in the far field. The setup for this measurement is very similar to Fig. 52. The impedance-matched HFC was used with the injection locked diode laser setup in order to maximize the generated Stokes power. A chopper was inserted at the exit of the HFC, several Rayleigh ranges away. Using the chopper, the beam radius was determined by the fall time as measured on a fast detector and an averaging oscilloscope.

The experimental results for the relative change in the beam radius are shown in Fig. 59. An average of 100 consecutive fall times were accumulated on the oscilloscope. Each data point with error bars in the figure represents the mean and standard deviation of 10 such accumulations. Therefore, a total of 1000 fall times were used to generate each data point.
Figure 61. Plot showing the relative far-field beam waist as a function of the incident pump power. The squares are were taken when no Stokes was generated (detuned from the Raman line center), whereas the circles were taken with Stokes.

Two sets of data are shown in the figure. The circles show the measured relative transmitted pump beam radius as a function of incident pump power. For reference, the squares show the the same measurements when the pump frequency was detuned from the Raman transition so that no Stokes light was generated. The far-field beam radius was observed to decrease with the Stokes intensity, corresponding to an increase in the effective mode waist within the HFC. This was direct evidence that the resonant mode waist within the Raman laser cavity was growing as a result of...
the Stokes power generation. The solid line is the theoretical prediction for the beam radius based on the thermo-optic model presented in Chapter 4.

**Unidirectional Raman Ring Lasing**

Motivated by mitigating the thermal lensing problem and reducing or eliminating the need for optical isolators, the first ring-geometry cw Raman laser cavity was created. The reflectivity of the cavity's input coupler was chosen much lower than the other three mirror reflectivities at the pump wavelength to optimize impedance matching. The resulting relatively high threshold of the impedance-matched configuration necessitated the use of the injection locking setup for pumping. The setup was therefore identical to figure 52 except that the last isolator after the slave was eliminated and the error signal was obtained by simply detecting the reflected signal from the cavity (no beam separator was necessary).

Interestingly, most ring lasers do not exhibit stable unidirectional operation because the gain for the forward and reverse directions are identical. However, stable operation was observed for this system in the forward direction without the use of intracavity elements to imbalance the forward and reverse gains. The gain for the Raman laser is inherently larger in the forward direction due to preferential Doppler broadening of the Raman transition in the reverse direction. However, a switch in the direction of the laser operation is predicted for large detunings because the reverse gain is broader than the forward. This means that at some detuning (estimated at
about the Raman half-width), the reverse gain should exceed the forward gain. However, insufficient pump power was available to observe this effect. This system was detuned ~50MHz to either side of the Raman line center and no switch was observed.

Figure 62 shows the steady-state results of the ring laser. When the cavity coupling efficiencies were considered, the threshold was observed to be very close to that of the comparable impedance-matched linear cavity, as predicted by the theory in Chapter 2. However, the deleterious effects of the thermal lensing were reduced roughly by a factor of four simply because the heat generation in the ring laser is deposited over four times the volume as in the comparable linear cavity. The solid curves shown in Fig. 62 represent the theoretical models from Chapter 4, while the dashed curves show the cw Raman theory without thermo-optic modifications. The fitting parameters used were: $\lambda_p=792\text{nm}, \lambda_s=1180\text{nm}, P_1 = 23.8\text{kW}, R_{p,rt}=0.99683, R_{s,rt}=0.99918, T_{p,0}=2920\text{ppm}, T_{p,1}=23.4\text{ppm}, T_{s,0} + T_{s,1} = 660\text{ppm}, L = 7.6\text{cm}, R = 50\text{cm}, K_{th} = 2.18\text{mW/cm-K}, n_0 = 1.001276, \tau = 38\mu\text{s}, D = 0.1443\text{cm}^2/\text{s}.$
Figure 62. Data showing stable operation in the forward direction of the first cw Raman ring laser. The dashed lines show the basic theory and the solid lines show the theory when the thermo-optic modifications are included.

In addition to its improved thermal behavior, the ring cavity allowed for the reduction in the number of isolators used. This was because the initial reflection from the cavity no longer returned along the input beam path. However, as discussed in Chapter 5, some unexpected resonant optical feedback from the cavity was still directed back along the input beam path. As a result, optical isolators could not be completely eliminated for active locking setups. On the other hand, the resonant optical feedback was used to passively lock the pump laser to the ring cavity. With this configuration, the ring cavity was made to lase using only two mirrors for alignment,
and two lenses for mode matching.

The first Raman ring laser therefore provided improved thermal and feedback characteristics. It also exhibited stable unidirectional operation in the forward direction due to preferential Doppler broadening of the Raman gain. Future researchers are encouraged to investigate the switching region that can be accessed by detuning the laser from the Raman gain line center.

Chapter Summary

This chapter has been devoted to documenting the experimental results from several diode-pumped cw Raman laser systems. Steady-state results from the first diode-pumped and the first high-efficiency cw Raman lasers were given. Both of the systems used the passive optical feedback method of frequency stabilization. The lowest threshold and the highest output power and conversion efficiency (at that time) were observed from these systems. The technique of injection locking was then implemented to increase the usable output power from an ECDL when active electronic locking was used. With the injection locking, a pump rate of 90 times threshold was achieved.

Results from the low-power systems were shown to agree well with the standard Raman laser theories. However, the results from the higher power systems deviated from the standard theory. The deviations were determined to be caused by thermo-optic effects inherent in the Raman process and experiments were conducted
to measure the resulting mode pulling and thermal lensing. With the thermal effects included, the theoretical models more accurately described the observed behavior.

The first Raman ring laser was then constructed and observed to operate stably in the forward direction. The laser was shown to be less sensitive to thermal lensing and the optical feedback along the beam path was significantly reduced, but not eliminated. The final chapter of this thesis provides a summary of the primary findings and suggests future research paths.
CHAPTER 7

SUMMARY

This thesis has documented the creation and development of diode-pumped cw Raman lasers. Extensive theoretical treatments of the classical, quantum, and thermo-optic aspects of cw Raman lasers was given. The classical treatment given in Chapter 2 was initiated by simply Taylor expanding Siegman's steady-state cavity equations with an intracavity gain medium. The results of the chapter included self-contained analytical expressions for the steady-state optical powers emitted from the cw Raman laser as well as the system's optimal construction and operation parameters. These equations are very useful in practice. Two appendices associated with this chapter are also worth mentioning. Appendix B gave a thorough treatment of the cw Raman laser equations starting from the Maxwell equations. Appendix D described the interaction between the Raman gain and the cavity modes. This appendix is very useful in developing an intuitive picture of the cw Raman laser tuning.

The quantum treatment that was given in Chapter 3 explored the quantum structure of the Raman medium interacting the two (quantized) cavity modes in detail. The very large single-photon detunings present in the system prohibited the use of the rotating wave approximation. This complicated matters when the quantum Langevin equations were derived, but later in the chapter the large detunings enabled dramatic
simplifications. In the semiclassical limit, the quantum results were shown to agree with the corresponding results from Chapter 2. An interesting and useful connection between this cw Raman system and the nondegenerate optical parametric oscillator was also revealed in this chapter. Finally, by linearizing the simplified quantum Langevin equations, an analytical expression for the output Stokes intensity noise spectra was derived.

By exploring this equation, it was shown that 100% photon statistics transfer could theoretically be obtained from the input pump to the output Stokes beam when the system is operated at four times the threshold and for Fourier frequencies below both of the cavity linewidths. Nonclassical behavior (50% intensity squeezing) was also predicted below the cavity linewidths, but for higher pump rates. The optimal operation conditions from an intensity noise standpoint were also determined and discussed. Appendix G gave a streamlined treatment of cw anti-Stokes generation. It was shown that photon conversion efficiencies to the anti-Stokes that approach the quantum limit of 50% are theoretically possible.

Thermo-optic effects were treated in Chapter 4. It was shown that inherent heat deposition from the Raman process caused frequency pulling of the Raman laser cavity modes and thermal lensing. The mode pulling was shown to be due to the thermally-induced refractive index change of the Raman gas along the beam axis. It was also discussed as a significant concern for frequency locking and the absolute frequency stability of the Stokes output. The mode pulling was shown to be significant
for even the most moderate Stokes output powers. The thermal lensing was shown to be caused by the thermally-induced refractive index gradient in the radial dimension away from the beam axis. The lensing was shown to modify the steady-state behavior of the cw Raman system from the treatments given in Chapters 2 and 3. The thermal lensing was shown to be a dominant concern only for the higher power systems.

The experimental work presented in this thesis began in Chapter 5. In this chapter the construction of a cw Raman laser was treated as an exercise in precision frequency stabilization and control theory. The three essential elements of the feedback loop (the plant, the discriminator, and the servo) were each discussed in detail. Optimization of the locking bandwidth was a central focus. It was also shown that the technique of injection locking could be used to amplify the pump power available for Raman conversion. A passive optical method of frequency locking was also discussed.

The experimental results obtained from properly locked diode-pumped cw Raman laser systems were discussed in Chapter 5. The results provided in this chapter document the first demonstration of diode-pumped Raman lasing, the first high efficiency Stokes generation through the use of an impedance-matched cavity, the achievement of a 90-times-threshold pump rate through the use of injection locking, the measurement of thermal mode pulling and thermal lensing, the measurement of the deleterious thermo-optic effects on the steady-state cw Raman laser behavior, and the demonstration of the first cw Raman ring laser. All of the experimental results compared favorably to the theories developed in Chapters 2 - 4.
The work presented in this thesis also opens many exciting future research paths. One can experimentally verify the time-dependent noise spectra predictions that were covered theoretically in Chapter 3. One should also be able to probe the predicted directional switching behavior of the ring laser with tuning. Along a similar line, one might also consider seeding the ring cavity with Stokes light in the backward direction to coerce a directional switch. Using the ring cavity, one can also examine the start-up behavior of the cw Raman process. This can be accomplished by performing the active cavity lock in one direction with a small amount of laser power. The majority of light could then be injected abruptly in the opposite direction to observe the start-up dynamics. Theoretical predictions in Chapter 3 also suggest the possibility of high-efficiency anti-Stokes generation. An experiment to demonstrate this would be well worth the effort. These are just a few of the many possible paths available to researchers in the future. The cw Raman laser system will undoubtedly provide fertile research ground for years to come.
APPENDICES
APPENDIX A

LASER-CAVITY FIELD EQUATION
This appendix is devoted to deriving a time-dependent laser-cavity equation based on a perturbative approach. Specifically, a standard steady-state laser-cavity equation is expanded in a Taylor series to first order in time to obtain a differential equation for the circulating laser field within a low-loss laser cavity. A variation of this method is posed as a problem in Siegman's *Lasers* [17, page 940].

A simplistic diagram of the cavity on which this treatment is based is given in Fig. 8. In chapter 11 of *Lasers*, Siegman performs a steady state analysis of a single-mode circulating field within a similar optical cavity. His treatment includes the effects of cavity mirrors, internal losses, a laser medium and an incident field. Because it deals with only round-trip effects and pertains only to the *one-way* circulating fields, the treatment applies equally well to standing-wave and traveling-wave cavities. Siegman's initial equation [17, page 413] can be written in the form

\[ E_{\text{circ}}(t) = g_{rt} E_{\text{circ}}(t - \tau_{rt}) + t_0 E_{\text{inc}}(t), \]  

where \( \tau_{rt} \equiv \frac{p}{c} \) is the round-trip time, and in contrast to Siegman, the explicit time dependence has been retained and the boundary conditions have been chosen such that there is no phase shift imparted to the field upon transmission through a mirror. This choice allows all the field amplitudes to be real. Also, the net round-trip gain, \( g_{rt} \), has been chosen to impart no net phase shift to the field upon round-trip propagation. The circulating field is therefore assumed to be resonant with the active cavity and mode pulling effects are ignored. As shown in Fig. 8, the circulating field amplitude, \( E_{\text{circ}}(t) \), is assumed to be measured just inside the input-coupling mirror,
while the incident field amplitude, $E_{\text{inc}}(t)$, is assumed to be measured just outside the same mirror. Equation (A.1) therefore implies that the total circulating field inside the input mirror is the sum of the circulating field which has traversed once around the cavity (and therefore left the input mirror one round-trip time earlier); plus the portion of the incident field that is transmitted through the input coupling mirror.

The net round-trip gain is defined as the fractional change in the field amplitude for a wave making one complete transit around the interior of the resonant cavity, and is given by

$$g_{\text{rt}} = r_{\text{rt}} \exp(\alpha_m p - \alpha_0 p), \quad (A.2)$$

where $r_{\text{rt}}$ is the effective round-trip amplitude mirror reflection coefficient ($r_{\text{rt}} = r_0 r_1^3$ for the case at hand). The net round-trip gain given by Eq. (A.2) therefore includes the effects of the mirrors, laser gain or absorption ($\alpha_m$ is the amplitude gain coefficient), and internal losses ($\alpha_0$ is the amplitude loss coefficient) due to Rayleigh, diffraction or other competing scattering processes. Note that the gain coefficient may be positive (gain) or negative (absorption). Since the argument of the exponential in Eq. (A.2) is very small compared to unity (one expects very small gain and loss per round-trip time), the term can be linearized to yield

$$g_{\text{rt}} \approx r_{\text{rt}} (1 + \alpha_m p - \alpha_0 p). \quad (A.3)$$

From this point on, it is assumed that $\alpha_0 = 0$ (lossless medium). This is a valid assumption for the laser medium at hand since the gas is very pure and other competing
effects are negligible. The term can be easily included if needed.

To examine the time dependence of the circulating field amplitude, the first term on the right side of Eq. (A.1) can be expanded in a Taylor series to yield

$$E_{\text{circ}}(t - \tau_{\text{rt}}) = E_{\text{circ}}(t) - \tau_{\text{rt}} \frac{dE_{\text{circ}}(t)}{dt} + \cdots$$  \hspace{1cm} (A.4)

So long as the circulating field changes little in one round-trip time of the cavity (i.e. low-loss cavity and low-gain active medium ($g_{\text{rt}} \approx 1$) one can safely retain only up to the first order term in the expansion. Equation (A.1) then becomes

$$\frac{dE_{\text{circ}}(t)}{dt} = -\tau_{\text{rt}}^{-1} (1 - g_{\text{rt}}) E_{\text{circ}}(t) + \tau_{\text{rt}}^{-1} t_0 E_{\text{inc}}(t)$$

$$= -\gamma E_{\text{circ}}(t) + \tau_{\text{rt}}^{-1} p \alpha_m E_{\text{circ}}(t) + \tau_{\text{rt}}^{-1} t_0 E_{\text{inc}}(t),$$  \hspace{1cm} (A.5)

where

$$\gamma \equiv \frac{2}{\tau_{\text{rt}}} (1 - \tau_{\text{rt}})$$

$$\approx \frac{1}{\tau_{\text{rt}}} (1 - R_{\text{rt}})$$  \hspace{1cm} (A.6)

is the cold-cavity power decay rate. Here, $R_{\text{rt}} = r_{\text{rt}}^2$ is the effective round-trip power reflection coefficient. Result (A.5), is therefore an expression for the first order dynamics within cavity for either the pump or Stokes field. This result is identical to that generated by the more rigorous approach when the slowly varying envelope approximation is invoked.
APPENDIX B

RAMAN LASER EQUATIONS: LAMB APPROACH
This extended appendix is devoted to deriving the Raman laser amplitude equations from first principles. The derivation came about through a wonderful interaction with Josh Bienfang in the spring of 2001. Together, we sketched out what has become my favorite of all the theoretical cw Raman treatments. The treatment integrates the Raman polarization smoothly and the necessary spatial overlap integrals emerge naturally. The material covered here follows closely the general laser treatments given in Chapter 24 of Siegman’s *Lasers* [17] and Chapter 8 of Sargent, Scully, and Lamb’s *Laser Physics* [142]. To my knowledge this approach was first introduced by Lamb [45] in 1964 and has since become somewhat of a standard for deriving general time-dependent laser-cavity equations.

Three contributions influence the dynamics of the intracavity laser field: the interactions with the atomic medium, the cavity input, and the cavity output. Lamb’s approach handles the field interactions with the atomic medium remarkably well, but resorts to an association with ohmic losses in order to incorporate decay of the intracavity field due to the cavity mirrors (output). Similar difficulties exist when incorporating an injected (pump) beam into the cavity (Siegman uses a lumped circuit analogy in his treatment). Nevertheless, the approach is widely used and very successful when compared to experimental observations. More rigorous treatments exist for dealing with the cavity input/output formalism [17, page 939].

Because the cavity input/output terms do not depend on the particular laser system, and because the derivation of these terms has been reproduced many times
(in varying degrees of rigor), this appendix will only concentrate on the Raman gain terms in the laser equations (i.e. the interactions with the medium). Everything that is unique to the cw Raman laser is contained in these terms. The cavity input/output terms are tacked on at the end for completeness.

**Vector Wave Equation**

Assuming a medium with no losses and neglecting contributions from the magnetic-dipole, the Maxwell equations for the (real) electric \([\mathcal{E}(r,t)]\) and magnetic \([h(r,t)]\) fields inside the laser cavity are (in mks units)

\[
\nabla \times \mathcal{E}(r,t) = -\frac{1}{\mu_0} \frac{\partial h(r,t)}{\partial t} \quad \text{and} \quad \nabla \times h(r,t) = \varepsilon_0 \frac{\partial \mathcal{E}(r,t)}{\partial t} + \frac{\partial p_a(r,t)}{\partial t},
\]

where the fields are functions of space \((r = x,y,z)\) and time. The electric-dipole atomic polarization density is represented by \(p_a(r,t)\). Plugging the second of these equations into the curl of the first, one can generate the vector wave equation.

\[
\frac{\partial^2 \mathcal{E}(r,t)}{\partial t^2} - \frac{1}{\mu_0 \varepsilon_0} \nabla^2 \mathcal{E}(r,t) = -\frac{1}{\varepsilon_0} \frac{\partial^2 p_a(r,t)}{\partial t^2},
\]

where the vector identity \(\nabla \times \nabla \times \mathcal{E}(r,t) \equiv \nabla(\nabla \mathcal{E}(r,t)) - \nabla^2 \mathcal{E}(r,t)\) has been used. From this point, it will be assumed that all fields are linearly polarized in the \(\hat{x}\)-direction and their vector characters can therefore be neglected.

**Transformation to Rotating Coordinates**

One can separate the rapidly oscillating portions of the field and polarization
from the slowly varying portion through the following transformations

\[ E(r, t) = E^{(\omega_p)}(r, t) + E^{(\omega_s)}(r, t) \]

\[ = \frac{1}{2} [\tilde{E}^{(\omega_p)}(r, t)e^{-i\omega_p t} + \text{c.c.}] + \frac{1}{2} [\tilde{E}^{(\omega_s)}(r, t)e^{-i\omega_s t} + \text{c.c.}] \]  \hspace{1cm} (B.3)

\[ \tilde{p}_a(r, t) = \tilde{p}_{a}^{(\omega_p)}(r, t) + \tilde{p}_{a}^{(\omega_s)}(r, t) \]

\[ = \frac{1}{2} [\tilde{p}_{a}^{(\omega_p)}(r, t)e^{-i\omega_p t} + \text{c.c.}] + \frac{1}{2} [\tilde{p}_{a}^{(\omega_s)}(r, t)e^{-i\omega_s t} + \text{c.c.}] \]  \hspace{1cm} (B.4)

where \( \tilde{E}^{(\omega_q)}(r, t) \) and \( \tilde{p}_{a}^{(\omega_q)}(r, t) \) are the slowly varying portions of the complex electric field and atomic polarization density that oscillate near the frequency \( \omega_q \).

Plugging Eqs. (B.3) and (B.4) into Eq. (B.2) and realizing that Fourier components at different carrier frequencies do not interact yields

\[ \frac{\partial^2 \tilde{E}^{(\omega_q)}(r, t)}{\partial t^2} - 2i\omega_q \frac{\partial \tilde{E}^{(\omega_q)}(r, t)}{\partial t} - \omega^2_q \tilde{E}^{(\omega_q)}(r, t) - c^2 \nabla^2 \tilde{E}^{(\omega_q)}(r, t) \]

\[ = -\frac{1}{\varepsilon_0} \left[ \frac{\partial^2 \tilde{p}_{a}^{(\omega_q)}(r, t)}{\partial t^2} - 2i\omega_q \frac{\partial \tilde{p}_{a}^{(\omega_q)}(r, t)}{\partial t} - \omega^2_q \tilde{p}_{a}^{(\omega_q)}(r, t) \right] \]  \hspace{1cm} (B.5)

for the negative frequency components at either the pump \((q = p)\) or Stokes \((q = s)\) frequency, where \( c^2 \equiv 1/\mu_0 \varepsilon_0 \).

**Slowly Varying Envelope Approximation**

One can now make the slowly varying envelope approximation (SVEA) by assuming that the time variations in \( \tilde{E}^{(\omega_q)}(r, t) \) and \( \tilde{p}_{a}^{(\omega_q)}(r, t) \) are slow compared to the optical carrier frequency \( \omega_q \). This allows the second order derivatives in Eq. (B.5) to be neglected. One can also assume that the atomic polarization density adiabatically follows any changes in the fields because it decays very rapidly compared to
the fields. This allows one to assume \( d\tilde{p}_{a}(\omega_{q})(r, t)/dt = 0 \). With these assumptions, Eq. (B.5) simplifies to

\[
2i\omega_{q} \frac{\partial \tilde{E}(\omega_{q})(r, t)}{\partial t} + \omega_{q}^{2} \tilde{E}(\omega_{q})(r, t) + c^{2} \nabla^{2} \tilde{E}(\omega_{q})(r, t) = -\frac{\omega_{q}^{2}}{\varepsilon_{0}} \tilde{p}_{a}(\omega_{q})(r, t).
\]  

(B.6)

This equation describes the evolution of the slowly-varying field \( \tilde{E}(\omega_{q})(r, t) \) at any given time and point in space driven by the slowly-varying polarization density \( \tilde{p}_{a}(\omega_{q})(r, t) \).

The next task will be to find out how the field and polarization interact within a high-finesse cavity.

**Expansion into Cavity Mode Basis**

One can expand both the field and the polarization density into the basis of cavity modes as

\[
\tilde{E}(\omega_{q})(r, t) = \sum_{n} \tilde{E}_{q,n}(t) \tilde{u}_{q,n}(r) \quad \text{and} \quad \tilde{p}_{a}(\omega_{q})(r, t) = \sum_{n} \tilde{P}_{q,n}(t) \tilde{u}_{q,n}(r),
\]  

(B.7)

where the \( \tilde{u}_{q,n}(r) \) represent the eigenmodes of the cavity in the spatial domain. Note that the subscript \( q \) refers to a longitudinal or axial mode \( (q = p, s) \) while \( n \) refers to a transverse or spatial mode of the cavity. The transverse modes are assumed to compose a complete, orthogonal set and are normalized such that

\[
\int \int \int_{\text{cavity}} \tilde{u}_{q,n}(r, t) \tilde{u}_{q,m}^{*}(r, t) d\mathbf{r} = V_{q,n} \times \delta_{nm},
\]  

(B.8)

where \( d\mathbf{r} \equiv dx \, dy \, dz \), \( \delta_{nm} \) is the Kronecker delta function and \( V_{q,n} \) is the effective volume of the \( n \)-th spatial mode corresponding to the \( q \)-th axial mode. This volume
is given by

\[ V_{q,n} = \int \int \int_{\text{cavity}} |\tilde{u}_{q,n}(r)|^2 dr. \] (B.9)

This normalization makes the mode functions \( u_{q,n}(r) \) dimensionless.

Because the cavity is high finesse and nonconfocal, one can safely assume that the pump and Stokes fields within the cavity can each be decomposed into a single spatial mode. Therefore, the infinite field sum given by the first of Eqs. (B.7) can be reduced to just a single term for each field

\[ \tilde{E}^{(\omega_q)}(r,t) = \tilde{E}_{q,n}(t)\tilde{u}_{q,n}(r). \] (B.10)

Moreover, in practice, it is almost always desirable and possible for the laser to operate in the fundamental (lowest order) spatial mode. Accordingly, from this point on only the fundamental spatial modes of the fields will be considered. The subscript \( n \) will therefore be omitted, but it should be understood that quantities like \( V_q \), \( \tilde{E}_q \), and \( \tilde{P}_q \) refer to the effective volume, the complex field, and the complex polarization, respectively, for the fundamental transverse mode of the \( q \)-th axial mode.

The spatial distribution of the polarization density, on the other hand, cannot be similarly decomposed into a single eigenmode in the cavity basis. The infinite sum from Eq. (B.7) is therefore retained for the polarization density. The resulting expansion coefficients in the cavity basis are then given by

\[ \tilde{P}_q(t) = \frac{1}{V_q} \int \int \int_{\text{atoms}} \tilde{\rho}^{(\omega_q)}(r,t) \tilde{u}_q^*(r) dr. \] (B.11)
This is just the spatial overlap of the atomic polarization density with the \( q \)-th fundamental cavity mode. Plugging the field from Eq. (B.10) and the polarization from Eq. (B.7) into Eq. (B.6) now allows a slowly-varying wave equation to be written for each mode \( q \) as

\[
2i\omega_q \tilde{u}_q(\mathbf{r}) \frac{\partial \tilde{E}_q(t)}{\partial t} + \omega_q^2 \tilde{E}_q(t) \tilde{u}_q(\mathbf{r}) + c^2 \tilde{E}_q(t) \nabla^2 \tilde{u}_q(\mathbf{r}) = -\frac{\omega_q^2}{\varepsilon_0} \tilde{P}_q(t) \tilde{u}_q(\mathbf{r}).
\] (B.12)

One can now separate this partial differential equation into a pair of ordinary differential equations through the separation-of-variables technique.

**Separation of Variables**

At this point, it is fruitful to separate time and space dependencies by first dividing both sides of Eq. (B.12) by \( c^2 \tilde{E}_q(t) \tilde{u}_q(\mathbf{r}) \). One can then reorganize terms to give

\[
\frac{2i\omega_q}{c^2 \tilde{E}_q(t)} \frac{\partial \tilde{E}_q(t)}{\partial t} + \frac{\omega_q^2}{c^2} + \frac{\omega_q^2}{c^2 \varepsilon_0 \tilde{E}_q(t)} \tilde{P}_q(t) = -\frac{1}{\tilde{u}_q(\mathbf{r})} \nabla^2 \tilde{u}_q(\mathbf{r}).
\] (B.13)

Because the left side is only a function of time while the right is only a function of space, each side can be set equal to a "separation constant", \( +k_{c,q}^2 \). This decouples the time and space dependencies and generates the following two differential equations

\[
(\nabla^2 + k_{c,q}^2) \tilde{u}_q(\mathbf{r}) = 0 \quad \text{(space)} \tag{B.14}
\]

\[
2i\omega_q \frac{d \tilde{E}_q(t)}{dt} + (\omega_q^2 - \omega_c^2) \tilde{E}_q(t) = -\frac{\omega_q^2}{\varepsilon_0} \tilde{P}_q(t) \quad \text{(time)} \tag{B.15}
\]

where \( \omega_c \equiv k_{c,q} c \). Eq. (B.14) is the Helmholtz equation, the eigenmodes of which can be determined given the boundary conditions of the cavity. The current discussion has
already been limited to just the fundamental spatial modes. The precise mathematical forms of these fundamental modes will be shown later. Solving the Helmholtz equation also allows identification of $k_{c,q}$ as the wave number, and therefore $\omega_{c,q}$ as the temporal (circular) frequency of the $q$-th cavity mode.

Choosing the phase such that the field is entirely real and equating the imaginary contributions in Eq. (B.15) gives

$$\frac{dE_q(t)}{dt} = -\frac{\omega_q}{2\varepsilon_0} \text{Im} \tilde{P}_q(t). \quad (B.16)$$

This important result matches Eq. (8.11) from Laser Physics and also Eq. (24.61) from Lasers when the input and output terms are ignored and when one realizes that

$$\tilde{P}_q(t) = \tilde{P}_{\text{Sargent}}(t) = \tilde{P}_{\text{Siegman}}(t).$$

This is due to the fact that Siegman associates his polarization with the positive frequency component when transforming to rotating coordinates while the present treatment follows Sargent, et al. and associates the polarization with the negative frequency component. This therefore also means that the quadrature component of the polarization relative to the field is $S_q(t) = \text{Im}\tilde{P}_q(t)$ for this treatment and $S_q(t) = -\text{Im}\tilde{P}_{\text{Siegman}}(t)$ for Siegman’s treatment.

**Raman Polarization**

The next task is therefore to determine the complex Raman polarization driving each mode $[\tilde{P}_q(t)]$. The imaginary part of this polarization (with respect to the field) can then be inserted into Eq. (B.16). Yariv [50, pages 469-473], and others [37, 85],
have used purely classical arguments to derive the complex Raman polarization density. This exercise will not be repeated here; only the results will be used. Yariv shows that the induced Raman polarization density at the Stokes (the pump counterpart is also given here) temporal Fourier frequency is given (in the notation of this thesis) by

$$\tilde{p}(\omega_s)(r, t) = \varepsilon_0 (\chi'_R - i \chi''_R) |\tilde{E}(\omega_p)(r, t)|^2 \tilde{E}(\omega_s)(r, t), \quad \text{(Stokes)} \tag{B.17}$$

$$\tilde{p}(\omega_p)(r, t) = -\varepsilon_0 (\chi'_R - i \chi''_R) |\tilde{E}(\omega_p)(r, t)|^2 \tilde{E}(\omega_p)(r, t), \quad \text{(pump)} \tag{B.18}$$

where $\chi'_R$ and $\chi''_R$ are the reactive (real) and dissipative (imaginary) portions of the third-order Raman nonlinear susceptibility as defined in Section 18.4 of *Quantum Electronics* [143]. In Eqs. (B.17) and (B.18), the time dependence of the polarization density has been included and the result has been generalized to three spatial dimensions. Because the phases of the two fields are not coupled to one another in either equation, one is free to choose them both arbitrarily. For simplicity, and to be consistent with Eq. (B.16), the phases are chosen such that the field amplitudes are real [i.e. $\tilde{E}(\omega)(r, t) = E_q(t)\tilde{u}_q(r)$].

One can now simply expand these polarization densities into the cavity basis as described previously in this appendix. Explicitly, applying Eqs. (B.9) and (B.11) to the Stokes polarization density given by Eq. (B.17) yields

$$\tilde{P}_s(t) = \frac{\varepsilon_0 (\chi'_R - i \chi''_R)}{V_s} \int \int \int_{\text{atoms}} |\tilde{E}(\omega_p)(r, t)|^2 \tilde{E}(\omega_s)(r, t) \tilde{u}_s^*(r) \, dr$$

$$= \varepsilon_0 (\chi'_R - i \chi''_R) |E_p(t)|^2 E_s(t) \eta_s. \tag{B.19}$$
where the unitless mode filling factor is defined by

\[ \eta_s = \frac{1}{V_s} \int \int |\tilde{u}_p(r)|^2 |\tilde{u}_s(r)|^2 dr, \]  

(B.20)

which represents the overlap between the atomic polarization and the Stokes cavity mode. The mode volume is given by

\[ V_s = \int \int \int_{cavity} |\tilde{u}_s(r)|^2 dr. \]  

(B.21)

The imaginary part of Eq. (B.19), which is the quadrature component with respect to the field, is then given by

\[ \text{Im} \tilde{P}_s(t) = -\varepsilon_0 \chi'' R |E_p(t)|^2 E_s(t) \eta_s. \]  

(B.22)

Plugging this into Eq. (B.16) gives

\[ \frac{dE_s(t)}{dt} = \frac{\omega_s}{2} \chi'' R |E_p(t)|^2 E_s(t) \eta_s, \]  

(B.23)

This important result and its pump field counterpart (exchange the s’s and the p’s and flip the sign) are the gain terms in the Raman field rate equations. The two remaining tasks are to solve for \( \eta_s \) and \( \chi'' R \) in terms of measurable quantities.

**Mode Filling Factor**

In order to perform the integrations in Eqs. (B.20) and (B.21), the expressions for the fundamental cavity modes [the \( \tilde{u}(r) \)'s] are needed. The bow-tie cavity mode (with small reflection angles) is assumed to be comprised of four identical segments (one for each pass), which do not overlap spatially. Appendix C shows that each of
these segments can be approximated by a radially symmetric fundamental TEM$_{00}$
Gaussians in the transverse dimensions (as for a linear cavity) and traveling waves in
the axial dimension (as opposed to standing waves for a linear cavity). The spatial
dependence of each cavity mode for a single pass [denoted by the superscript (sp)]
then takes the form [40]

\[
\tilde{u}^{(sp)}_q(r) = \frac{1}{1 + i2z/b} \exp \left[ -\frac{r^2 k_q}{b(1 + i2z/b)} \right] \times e^{-i k_n z}, \quad \text{(traveling waves, single pass)}
\]  (B.24)

where $b$ is confocal parameter (this is defined by the cavity mirror separation and
curvature), $k_q = 2\pi/\lambda_q$ is the wavenumber, and the origin is assumed to be at the
focus. The overall mode function $\tilde{u}_q(r)$ will be a sum of four of these single-pass
mode functions, each of which occupies a different volume in the cavity. However,
since the four segments are identical and do not overlap, only a single pass needs to
be considered. From Eq. (B.20), the mode filling factor for the Stokes becomes

\[
\eta_s = \frac{V^{(sp)}_{int}}{V^{(sp)}_s} \quad \text{(B.25)}
\]

where the single-pass interaction volume is defined by

\[
V^{(sp)}_{int} \equiv \int \int \int_{\text{single pass}} |\tilde{u}_p^{(sp)}(r)|^2 |\tilde{u}_s^{(sp)}(r)|^2 dr
= \frac{b^2 \lambda_p \lambda_s}{4(\lambda_p + \lambda_s)} \tan^{-1}(L/b), \quad \text{(B.26)}
\]

and the Stokes mode volume for a single pass is given by

\[
V^{(sp)}_s = \int \int \int_{\text{cavity}} |\tilde{u}_s^{(sp)}(r)|^2 dr
= \frac{\lambda_s bL}{4}, \quad \text{(B.27)}
\]
where $L$ is the length of the pass. Combining the last two expressions gives

$$\eta_s = \frac{b}{L \lambda_p + \lambda_s} \tan^{-1}(L/b), \quad \text{(traveling waves)} \quad (B.28)$$

which has a typical value between $1/2$ and $1$. Note that because $\eta_s$ represents the fractional overlap, its value doesn’t depend on the number of passes that are made as long as they are identical and don’t overlap. The mode filling factor for the pump can be obtained by exchanging the $p$’s and the $s$’s. The only remaining task is to find the Raman susceptibility in terms of measurable quantities.

**Plane-Wave Gain Coefficient**

The plane wave gain coefficient, $\alpha$, is often used to characterize the Raman gain rather than the susceptibility because it can be measured experimentally. It is defined through the exponential intensity growth equation

$$I_s(z) = \exp[\alpha I_p z]. \quad (B.29)$$

Therefore, $\alpha$ can be interpreted as the exponential growth rate of the Stokes intensity per unit length, per unit pump intensity, for plane wave fields (uniform in the transverse dimensions). This assumes that the pump intensity remains constant (undepleted pump approximation).

To connect $\chi''_R$ from Eq. (B.23) with $\alpha$, one can use the relation between intensity and a real field along with its derivative [144, page 171]

$$I_s(t) = \frac{c\epsilon_0}{2} E_s^2(t) \quad \text{and} \quad \frac{dI_s(t)}{dt} = c\epsilon_0 E_s(t) \frac{dE_s(t)}{dt}. \quad (B.30)$$
Plugging Eq. (B.23) into the second equation and using the first to convert the fields to intensities, the Stokes intensity growth in time is

\[ \frac{dI_s(t)}{dt} = \frac{2\omega_s \chi''_R}{c \varepsilon_0} I_p I_s(t). \]  

(B.31)

One can either integrate this over a time \( t' = z/c \), where \( z \) is distance or equivalently, use the transformation

\[ \frac{d}{dz} \rightarrow \frac{1}{c} \frac{d}{dt} \]  

(B.32)

to give

\[ \frac{dI_s(z)}{dz} = \frac{2\omega_s \chi''_R}{c^2 \varepsilon_0} I_p I_s(z). \]  

(B.33)

This equation is separable with a solution

\[ I_s(z) = \exp \left[ \frac{2\omega_s \chi''_R}{c^2 \varepsilon_0} I_p z \right]. \]  

(B.34)

Comparison with Eq. (B.29) quickly yields

\[ \chi''_R = \frac{c^2 \varepsilon_0 \alpha}{2\omega_s}. \]  

(B.35)

This gives the relationship between the Raman susceptibility and the Raman plane-wave gain coefficient.

**Raman Field Equations**

Combining Eqs. (B.23), (B.28), and (B.35) gives the following two time-dependent
Raman field equations

\[ \frac{dE_p}{dt} = -\frac{\gamma_p}{2} E_p - \frac{\omega_p^2}{\omega_s^2} G|E_s|^2 E_p + \tau_{it}^{-1}\sqrt{T_{p,0}} E_p^{\text{inc}}, \]  
\( (B.36) \)

\[ \frac{dE_s}{dt} = -\frac{\gamma_s}{2} E_s + G|E_p|^2 E_s, \]  
\( (B.37) \)

where the cavity decay and input terms have now been included for completeness and

\[ G = \frac{c^2 \varepsilon_0 \alpha b}{4L} \frac{\lambda_p}{\lambda_p + \lambda_s} \tan^{-1}(L/b). \]  
\( (B.38) \)

Note that the ratio of optical frequencies is squared in Eq. (B.36) in contrast to the treatment of Brasseur, et al. [26]. This apparent discrepancy was pointed out by Lei Meng. However, the difference is due to the fact that Brasseur effectively normalizes all of his mode filling factors to the pump mode volume. For consistency, he also uses the pump volume when converting to optical power. In this work, the volume of the mode in question is used for normalization and when converting to optical power. It can easily be verified that the two methods yield equivalent results after converting to optical power.

**Conversion to Optical Power**

One can convert Eqs. (B.36) and (B.37) to optical power using the relation \( P = I \times A \), where \( A \) is the cross-sectional area of the beam. For radially symmetric Gaussian beams \( A = \pi \omega_0^2 \), where \( \omega_0 \) is the minimum beam radius. Using this and \( I \) from Eqs. (B.30), the optical power and its derivative can be written

\[ P_q = \frac{\pi \omega_0^2 c \varepsilon_0}{4} E_q^2 \quad \text{and} \quad \frac{dP_q}{dt} = \frac{\pi \omega_0^2 c \varepsilon_0}{2} E_q \frac{dE_q}{dt}. \]  
\( (B.39) \)
This allows Eqs. (B.36) and (B.37) to be written in terms of optical powers as

\[
\frac{dP_p}{dt} = -\gamma_p P_p - \frac{\omega_p^2}{\omega_s^2} \frac{16G}{\lambda_s b c e_0} P_s P_p + \frac{2c}{p} \sqrt{T_{p,0}} \sqrt{P_p} \sqrt{P_{\text{inc}}},
\]

(B.40)

\[
\frac{dP_s}{dt} = -\gamma_s P_s + \frac{16G}{\lambda_p b c e_0} P_p P_s.
\]

(B.41)

It can be readily verified that these two equations match the results given in the main chapter by Eqs. (2.10), (2.13) and (2.14).

**Standing Wave Cavity**

The derivation given in this appendix, with the exception of the mode filling factors, is independent of cavity geometry. The methods presented can be applied equally well to a standing wave cavity. To do this, one should take \(E_p(t)\) and \(E_s(t)\) as the one-way circulating fields in the cavity. With this, the only modifications for standing wave cavities are the mode filling factors.

Take, for instance, a two-mirror standing-wave cavity (i.e. a linear cavity) whose round trip is composed of two passes that can be described by Eq. (B.24). However, it is important to note that these two passes now overlap in space. This means that the round-trip mode function (not just a single pass) for the standing-wave cavity is given by

\[
\tilde{u}_q(r) = \frac{1}{1 + i2z/b} \exp \left[ \frac{-k_q r^2}{b(1 + i2z/b)} \right] \times 2 \cos k_q z. \quad \text{(standing waves, round trip)}
\]

(B.42)

Using this form of the cavity mode function, the resulting mode filling factors can be
calculated exactly as above. Performing the necessary integrations yields

\[ \eta_s = \frac{2b}{L} \frac{\lambda_p}{\lambda_p + \lambda_s} \tan^{-1}(L/b), \quad \text{(standing waves)} \]  

(B.43)

which is a factor of two greater than the traveling wave case [see Eq. (B.28)]. Again, the pump mode filling factor for standing waves can be obtained by simply exchanging the s's and p's.

Using the standing wave mode filling factors and a round-trip length of \( p = 2L \), one can quickly verify that in the steady state, the laser threshold, the transmitted pump power, the reflected pump power, and the emitted Stokes power are all identical for the linear cavity and the bow-tie ring cavity of equal single-pass length, mirror curvature, and mirror reflectivities. In other words, for the linear cavity, one can simply use Eqs. (2.10), (2.17), and (2.22)-(2.24) with \( R_{rt} = R_0R_1 \) and \( T_{s, tot} = T_{s,0} + T_{s,1} \). Interestingly, the larger mode filling factor of the linear cavity just balances the fact that it only has two passes per round trip rather than four in the ring cavity.
APPENDIX C

BOW-TIE CAVITY EIGENMODE
This appendix is devoted to calculating the lowest order (fundamental) eigenmode of the bow-tie cavity starting from the Helmholtz equation \[ \text{given by Eq. (B.14) from Appendix B}. \] It will be shown that when the reflection angles are small, each pass in the bow-tie cavity can be approximated by a circular Gaussian beam as for the linear cavity, but with a traveling wave character in the axial dimension rather than a standing wave character.

**Paraxial Helmholtz Equation**

Assuming traveling wave propagation in the \( \hat{z} \)-direction, one can extract the rapidly oscillating portion of \( \tilde{u}_q(r) \) using the transformation

\[
\tilde{u}_q(r) = \tilde{U}_q(r)e^{-ik_qz}
\]

where now \( \tilde{U}_q(r) \) is the (slowly-varying) complex field envelope for the \( q \)-th axial mode. This basically extracts the axial traveling-wave character. Inserting this into the Helmholtz equation [Eq. (B.14)] from Appendix B yields

\[
\nabla^2 \tilde{U}_q(r) + \frac{\partial^2 \tilde{U}_q(r)}{\partial z^2} - 2ik_q \frac{\partial \tilde{U}_q(r)}{\partial z} = 0,
\]

where two terms have canceled and \( \nabla^2 T = \partial^2 / \partial x^2 + \partial^2 / \partial y^2 \) is the transverse Laplacian operator. Also, the difference between the cavity and field wavenumbers has been neglected \( (k_{c,q} = k_q) \).

Now one can apply the spatial version of the slowly varying envelope approximation. Since the field varies slowly over the distance an optical wavelength, one
can neglect the second order derivative with respect to $z$ compared to the first order derivative. This yields the paraxial Helmholtz equation

$$\nabla^2 \tilde{U}_q(r) - 2i k_q \frac{\partial \tilde{U}_q(r)}{\partial z} = 0. \quad (C.3)$$

The fundamental mode solution to this differential equation describes a Gaussian beam, which takes the mathematical form [17, page 639]

$$\tilde{U}_q(r) = \frac{\tilde{q}_0}{\tilde{q}(z)} \exp \left[ -ik_q \frac{x^2 + y^2}{2\tilde{q}(z)} \right], \quad \text{(Circular Gaussian)} \quad (C.4)$$

when the beam cross-section is circular. In this expression, the complex beam radius as a function of the axial displacement $z$ from the focus is given by

$$\tilde{q}(z) = \tilde{q}_0 + z, \quad (C.5)$$

where $\tilde{q}_0 = -iz_R$, with $z_R$ representing the Rayleigh range. Noting that the confocal parameter and the Rayleigh range are related by $b = 2z_R$, one can verify that Eq. (C.4) is consistent with the slowly Eq. (B.24) from Appendix B, which came from the work of Boyd [40].

When an elliptical beam cross-section can exist (as in all cavities except the linear cavity), Eq. (C.4) can be generalized to give the elliptic Gaussian beam function

$$\tilde{U}_q(r) = \sqrt{\tilde{q}_0 x \tilde{q}_0 y} \frac{\tilde{q}_0 x}{\tilde{q}_x(z) \tilde{q}_y(z)} \exp \left[ -ik_q \left( \frac{x^2}{2\tilde{q}_x(z)} + \frac{y^2}{2\tilde{q}_y(z)} \right) \right]. \quad \text{(Elliptic Gaussian)} \quad (C.6)$$

Now the complex beam radius is divided up into two components; one for each transverse dimension. For the $x$-dimension, this gives

$$\tilde{q}_x(z) = \tilde{q}_0 x + z, \quad (C.7)$$
where \( \tilde{q}_{0,x} = -iz_{R,x} \) and similarly for the y-dimension. Like Eq. (C.4), the elliptic Gaussian function given by Eq. (C.6) is normalized to unity at the center of the focus \((z = x = y = 0)\), and depends on the cavity geometry only through the Rayleigh range. Therefore, if the Rayleigh range (or confocal parameter) is known along each path of the ring cavity, then all information about the beam can be obtained using Eq. (C.6). Attention is therefore now focused on calculating the confocal parameter.

**Confocal Parameter**

The ABCD matrix formalism developed by Kogelnik and Li [145] can be used to determine the confocal parameter for any path within a stable cavity. Consult Chapters 14-23 of *Lasers* [17] for a complete discussion of this method.

Since the bow-tie cavity under consideration is symmetric, the only ray matrices required are [17, page 585]

\[
M_L = \begin{bmatrix} 1 & L \\ 0 & 1 \end{bmatrix}, \quad \text{(propagation through free space distance } L) \quad (C.8)
\]

\[
M_R = \begin{bmatrix} 1 & 0 \\ \frac{-2}{R_e} & 1 \end{bmatrix}, \quad \text{(Reflection by curved mirror radius } R_e) \quad (C.9)
\]

\[
M_{L'} = \begin{bmatrix} 1 & L' \\ 0 & 1 \end{bmatrix}, \quad \text{(propagation through free space distance } L') \quad (C.10)
\]

where \( L' = L / \cos 2\theta \), the effective mirror curvature \( R_e = R \cos \theta \) in the plane of incidence ("tangential") and \( R_e = R / \cos \theta \) perpendicular to the plane of incidence ("sagittal").

Taking the reference plane to be just inside the input mirror (along the \( L \) path),
and exploiting the cavity symmetries, the overall cavity matrix is

\[ M' = (M_R \cdot M_{L'} \cdot M_R \cdot M_L)^2. \]  

(C.11)

Also due to the cavity symmetry, one can drop the square and pretend there’s a flat mirror bisecting the cavity horizontally, perpendicular to the plane containing the beams. This simplification gives

\[ M = M_R \cdot M_{L'} \cdot M_R \cdot M_L = \begin{bmatrix} 1 - \frac{2L'}{R_e} & L + L' \left(1 - \frac{2L}{R_e}\right) \\ -\frac{2}{R_e} \left(2 - \frac{2L'}{R_e}\right) \left(1 - \frac{2L}{R_e}\right) - \frac{2L}{R_e} \end{bmatrix} \]  

(C.12)

The requirement for a stable resonator mode to exist is [17, 818-820]

\[ \left| \frac{A + D}{2} \right| \leq 1. \]  

(C.13)

This places a limit on the effective mirror radii of curvature relative to the lengths \( L \) and \( L' \). The beam’s radius of curvature at the reference plane is given by [17]

\[ R(-L/2) = \frac{2B}{D - A} = \frac{2}{R_e} \left( \frac{L + L' - \frac{2L'L'}{R_e}}{L - \frac{2L'}{R_e}} \right), \]  

(C.14)

where the origin \((z = 0)\) is midway between the mirrors by symmetry. Note that in the limit that \( L = L' \), the beam’s radius of curvature equals that of the mirror (as it does for the linear cavity).
The expression

\[ R(z) = z \left( 1 + \frac{z^2}{b^2} \right) \]

relates the beam radius of curvature at any distance, \( z \), to the confocal parameter (or the Rayleigh range) [17, page 665]. Solving this equation for \( b \) at \( z = -L/2 \) and inserting Eq. (C.14) yields

\[ b = L \left[ \frac{R_e}{L} \left( \frac{L + L' - \frac{2LL'}{R_e}}{L - \frac{LL'}{R_e}} \right) - 1 \right]^{1/2} \]  

(C.16)

This expression gives the confocal parameter for either of the two transverse dimensions. For the dimension in the plane of the beams (call this \( b_x \), \( R_e = R \cos \theta \)), and for the other transverse dimension (\( b_y \), \( R_e = R/ \cos \theta \)). Also, using the symmetry properties of the cavity, the confocal parameter along the length \( L' \) is obtained by simply making the transformation \( L \leftrightarrow L' \) in Eq. (C.16). Finally, note that this result is independent of the wavelength of light, so it is equally valid for both the pump and Stokes cavity modes. With the confocal parameters determined, Eq. (C.6) completely describes the spatial properties of the fields within the cavity.

**Mode Filling Factor**

The mode filling factor, \( \eta_s \), in Eq. (B.20) of Appendix B can now be calculated. As in Appendix B, the critical assumption is again made that the overall cavity mode is a sum of four passes that do not overlap. Mathematically, this means that cross
terms that occur in the calculation of $\eta_s$ can be neglected. The mode filling factor from Eq. (B.20) can then be written as

$$\eta_s = \frac{\psi_{int}(L) + \psi_{int}(L')} {\psi_s(L) + \psi_s(L')}$$  \hspace{1cm} (C.17)

where the superscripts $L$ and $L'$ are associated with the single passes of lengths $L$ and $L'$, respectively, and

$$\psi_{int}^{(L)} = \int \int \int_{L_{pass}} |\hat{u}_{p}^{(L)}(r)|^2 |\hat{u}_{s}^{(L)}(r)|^2 dr,$$

with a similar expression for the $L'$ pass. Eq. (C.17) leaves four volume integrals (two interaction volumes and two mode volumes) to be calculated. However, due to the cavity symmetry, only the integrals for one pass need to be calculated and then $L$ and $L'$ can be interchanged to give the remaining solutions. It is handy to note that the magnitude squared of Eq. (C.6) is the same as that of Eq. (C.1) and can be written

$$|\hat{u}_q(r)|^2 = |\hat{U}_q(r)|^2$$

$$= \frac{1}{\sqrt{1 + \tau_x^2}} \frac{1}{\sqrt{1 + \tau_y^2}} \exp \left[ -2k_q \left( \frac{x^2}{b_x(1 + \tau_x^2)} + \frac{y^2}{b_y(1 + \tau_y^2)} \right) \right]$$  \hspace{1cm} (C.19)

where

$$\tau_x = 2z/b_x \quad \text{and} \quad \tau_y = 2z/b_y$$  \hspace{1cm} (C.20)

are scaled propagation distances from the focus.

One of the mode volume integrals in Eq. (C.17) will be tackled first. For a single
$L$ pass, the Stokes mode volume is given by

$$V_s^{(L)} = \int \int \int_{L \text{ pass}} |\tilde{u}_s^{(L)}(r)|^2 dr$$

$$= \int \int \int_{L \text{ Pass}} \frac{1}{\sqrt{1 + \tau_x^2}} \frac{1}{\sqrt{1 + \tau_y^2}}$$

$$\times \exp \left[ -2k_s \left( \frac{x^2}{b_x(1 + \tau_x^2)} + \frac{y^2}{b_y(1 + \tau_y^2)} \right) \right] dr. \quad (C.21)$$

This looks a bit nasty, but the situation improves dramatically with the substitution $v_x^2 = 2k_s x^2 / b_x(1 + \tau_x^2)$ and analogously for the $y$-dimension. The integral then simplifies to

$$V_s^{(L)} = \frac{\sqrt{b_x b_y}}{2k_s} \int_{-L/2}^{L/2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{-v_x^2} e^{-v_y^2} dv_x dv_y dz, \quad (C.22)$$

which can be easily integrated to yield

$$V_s^{(L)} = \frac{\pi L \sqrt{b_x b_y}}{2k_s} = \frac{\lambda_s \sqrt{b_x b_y} L}{4}. \quad (C.23)$$

With the mode volume nailed down, the overlap integral in Eq. (C.17) can now be calculated. This integral is not as pleasant as the one just performed. In this case, noting that the confocal parameter is independent of wavelength, one can write

$$V_{\text{int}}^{(L)} = \int \int \int_{L \text{ pass}} |\tilde{u}_p^{(L)}(r)|^2 |\tilde{u}_s^{(L)}(r)|^2 dr$$

$$= \int \int \int_{L \text{ Pass}} \frac{1}{(1 + \tau_x^2)} \frac{1}{(1 + \tau_y^2)}$$

$$\times \exp \left[ -2(k_p + k_s) \left( \frac{x^2}{b_x(1 + \tau_x^2)} + \frac{y^2}{b_y(1 + \tau_y^2)} \right) \right] dr. \quad (C.24)$$

Again, with a similar substitution of $v_x^2 = 2(k_p + k_s) x^2 / b_x(1 + \tau_x^2)$ and analogously
for the $y$-direction, the $x$- and $y$-dimensions can be easily integrated. This leaves

$$V_{\text{int}}^{(L)} = \frac{\pi \sqrt{b_x b_y}}{2(k_p + k_z)} \int_{-L/2}^{L/2} \frac{1}{\sqrt{1 + \tau_x^2}} \frac{1}{\sqrt{1 + \tau_y^2}} \, dz. \quad (C.25)$$

The remaining integral over the $z$-dimension is an elliptic integral of the first kind, which requires numerical integration to solve. However, there are two possible options for obtaining an analytic result. First, if the beam remains nearly planar within the cavity, one can assume $b \gg L$, directly expand the integrand for small $\tau$, and integrate term by term. Alternatively, when the reflection angle $\theta$ is reasonably small, $\tau_x \approx \tau_y$, and it is worthwhile expressing the integrand of Eq. (C.25) as

$$\frac{1}{\sqrt{1 + \tau_x^2}} \frac{1}{\sqrt{1 + \tau_y^2}} = \frac{1}{\sqrt{1 + (2 - \epsilon)\tau^2 + \tau^4}}, \quad (C.26)$$

where

$$\tau^2 = \tau_x \tau_y = \frac{4x^2}{b_x b_y} \quad \text{and} \quad \epsilon = 2 - \left( \frac{\tau_x}{\tau_y} + \frac{\tau_y}{\tau_x} \right) = 2 - \left( \frac{b_x}{b_y} + \frac{b_y}{b_x} \right). \quad (C.27)$$

The integrand can then be expanded about $\epsilon = 0$ to yield

$$\frac{1}{\sqrt{1 + (2 - \epsilon)\tau^2 + \tau^4}} = \frac{1}{1 + \tau^2} + \frac{\tau^2 \epsilon}{2(1 + \tau^2)^3} + \cdots. \quad (C.28)$$

This result can be used in Eq. (C.25) and integration can be performed term by term to generate an analytic result. Keeping only the first term in the expansion and
integrating gives

\[ V_{\text{int}}^{(L)} = \frac{\pi \sqrt{b_x b_y}}{2(k_p + k_s)} \int_{-L/2}^{L/2} \frac{1}{1 + \tau^2} d\tau \]

\[ = \frac{\pi b_x b_y}{2(k_p + k_s)} \tan^{-1}(L/\sqrt{b_x b_y}) \]

\[ = \frac{b^2}{4} \frac{\lambda_p \lambda_s}{(\lambda_p + \lambda_s)} \tan^{-1}(L/b), \quad (C.29) \]

where

\[ b \equiv \sqrt{b_x b_y}. \quad (C.30) \]

Keeping the only the first term in the expansion turns out to be an amazingly good approximation. It producing less than 0.1% error for all angles \( \theta < 10 \) degrees when \( L/R > 1/2 \). This is due to the fact that the confocal parameters are very mild functions of \( \theta \) in Eq. (C.16). With the mode volume and the overlap now known, the mode filling factor for the bow-tie cavity can now be calculated from Eq. (C.17) to give

\[ \eta_s = \frac{\lambda_p}{(\lambda_p + \lambda_s)} \frac{b^2 \tan^{-1}(L/b) + b^2 \tan^{-1}(L'/b')}{L b + L'b'} \]

\[ \approx \frac{b}{L} \frac{\lambda_p}{\lambda_p + \lambda_s} \tan^{-1}(L/b) \quad \text{(for small } \theta), \quad (C.31) \]

which matches Eq. (B.28) from Appendix B. Therefore, when the reflection angle (relative to the normal) is less than about ten degrees, one using the mode filling factor for the linear traveling-wave cavity results in errors \( \lesssim 5\% \).
APPENDIX D

FREQUENCY TUNING DESCRIPTION
This short appendix is devoted to describing a very useful physical picture of the interactions between the Raman gain profile and the HFC cavity modes. Although it seems elementary now, the thought of viewing the pump and the Stokes cavity modes as members (albeit distant) of the same frequency comb that represents the possible HFC resonances had not occurred. However, in the process of preparing and applying for an upcoming post-doctoral position in femtosecond modelocked lasers, I found that the frequency comb ideas that are so prevalent in that field could be directly adapted to the doubly-resonant cw Raman system. The frequency comb interpretation lends itself to a very useful picture of the frequency tuning behavior for a cw Raman lasers. A simple schematic of this picture is shown in Fig. 63 and will now be discussed.
Figure 63. Frequency tuning picture for a doubly-resonant cw Raman laser. The picture is not to scale. Part (a) shows the HFC (high-finesse cavity) modes for a round-trip cavity length of $p$, whereas part (b) shows the modes for a longer round-trip cavity length of $p'$. The picture illustrates how changing the cavity length will tune the Stokes cavity mode across the Raman gain profile by an amount $\Delta_{12}$.

Parts (a) and (b) of the figure show the cavity modes for two different HFC round-trip lengths. In practice, the cavity length can be changed using a piezoelectric transducer. The picture is not to scale to make the mechanisms more clear. Also, all the frequencies shown in the figure are given as circular frequencies (rad/s) to maintain consistency with the rest of this thesis.

Part (a) of the figure shows the cavity modes of the HFC for a round trip length $p$ ($p = 2L$ for the linear cavity). This means that the cavity mode spacing, or free spectral range (FSR) of the cavity is $c/p$ in Hz (the extra $2\pi$ comes from the conversion
to circular frequency). One can imagine injecting pump light into one of these cavity modes (marked $\omega_p$ in the figure). Due to the Raman process, this pump light will produce gain that is centered at a frequency $\omega_{21}$ (the Raman shift) below the pump frequency. This gain has a width of $\gamma_{21}$ and its profile is represented by a dotted curve in the figure. For the case of diatomic hydrogen gas, the gain is centered $\omega_{21}/2\pi \approx 125\text{THz}$ ($4155\text{cm}^{-1}$) below the pump laser frequency. Stokes laser light can then build in the cavity mode nearest to the peak of the gain profile (marked $\omega_s$ in the figure). In part (a) of the figure, the Stokes cavity mode is shown tuned (by changing the length of the cavity) to the Raman gain line center.

Part (b) shows what happens when the cavity length is increased to $l'$. The HFC mode spacing decreases, so the cavity modes all compress towards $\omega = 0$ like an accordion. It is important to note that the modes do not tune the same amount for a given change in the cavity length. Instead, the fractional change in each frequency is the same for all modes. Using this fact, one can relate the absolute frequency tuning of Stokes mode ($\delta_s$) to that of the pump mode ($\delta_p$) through

$$\delta_s = \omega_s \frac{\delta_p}{\omega_p}.$$  \hspace{1cm} (D.1)

Even though the cavity modes compress like an accordion when the length is changed, the Raman gain profile always remains centered $\omega_{21}$ below the pump frequency. In other words, the gain profile tunes rigidly with the pump cavity mode as shown in the figure. As a result, the Stokes cavity mode is “dragged” through the Raman gain profile.
In Chapter 2, frequency tuning of the Stokes cavity mode relative to the Raman gain line-center ($\Delta_{12}$) is discussed and the Stokes output curves are shown in Fig. 11. However, this relative tuning should not be confused with the absolute tuning of the emitted Stokes optical frequency. The relative tuning of the Stokes mode can be identified as $\Delta_{12}$ in Fig 63. From this figure, one can relate the relative frequency tuning of the Stokes mode to the absolute frequency tunings of the modes through

$$\Delta_{12} = \delta_p - \delta_s$$

$$= \delta_p \left(1 - \frac{\omega_s}{\omega_p}\right), \tag{D.2}$$

where Eq. (D.1) has been used. This gives the tuning of the Stokes mode relative to the Raman gain center as a function of the absolute tuning of the pump laser. Using this expression, one can convert the horizontal axis of Fig 11 to absolute pump tuning.

One can estimate the maximum absolute tuning range of the Stokes output light by replacing $\Delta_{12}$ in Eq. (D.2) with $\gamma_{21}$ and using Eq. (D.1) to solve for $\delta_s$. This is the absolute Stokes mode tuning that corresponds to a relative tuning of a full width across the Raman gain profile and is given by

$$\delta_{s,\text{max}} = \frac{\gamma_{21}}{\left(\frac{\omega_p}{\omega_s} - 1\right)}$$

$$= \gamma_{21} \frac{\omega_s}{\omega_{21}}. \tag{D.3}$$

This is more of a guideline than an ultimate maximum. The result is interesting because it shows that if the Raman shift ($\omega_{21}$) is smaller than the Stokes frequency,
then the maximum Stokes tuning range will be greater than the Raman gain linewidth. This is the regime shown in Fig. 63. However, if the Raman shift is greater than the Stokes frequency, then the maximum tuning range will be smaller than the gain linewidth. In Fig. 63, this would mean that the Raman gain profile would be centered at less than half the pump frequency. For vibrational Raman shifts in diatomic hydrogen gas, this effect significantly decreases the tuning range for mid-infrared relative to visible or near-infrared Raman lasers.
APPENDIX E

PUMP LANGEVIN EQUATION
This appendix is devoted to deriving the quantum Langevin equation for the pump annihilation operator using the Heisenberg equation of motion. Derivation of the Stokes equation follows analogously.

Choosing the first element \( a_p \) in the a array given in Eq. (??), the Heisenberg equation of motion in the form of Eq. (3.11) becomes

\[
\dot{a}_p = -\frac{i}{\hbar}[a_p, H_{\text{sys}}] - \sum_{j=1}^{s} [a_p, c_j^\dagger] \left\{ \frac{1}{2} \gamma_j c_j - \sqrt{\gamma_j} b_j^{\text{in}} \right\} + \sum_{j=1}^{s} \left\{ \frac{1}{2} \gamma_j c_j^\dagger - \sqrt{\gamma_j} b_j^{\text{in}} \right\}[a_p, c_j],
\]  

(E.1)

with the \( c_j \)'s, \( \gamma_j \)'s and \( b_j \)'s defined by Eqs. (3.12) - (3.15). Using Eqs. (3.2) and (3.3), the commutation in the first term on the right side of Eq. (E.1) can be written

\[
[a_p, H_{\text{sys}}] = \hbar \omega_1 [a_p, S_{11}] + \hbar \omega_2 [a_p, S_{22}] + \hbar \omega_3 [a_p, S_{33}]
\]

\[
+ \hbar \omega_p^c [a_p, a_p^\dagger a_p] + \hbar \omega_p^s [a_p, a_s^\dagger a_s]
\]

\[
+ i\hbar \left( [a_p, a_p^\dagger] - [a_p, a_p] \right)
\]

\[
\times \left( g_{p,13} S_{13} + g_{p,13}^* S_{13}^\dagger + g_{p,23} S_{23} + g_{p,23}^* S_{23}^\dagger \right)
\]

\[
+ i\hbar \left( [a_p, a_s^\dagger] - [a_p, a_s] \right)
\]

\[
\times \left( g_{s,13} S_{13} + g_{s,13}^* S_{13}^\dagger + g_{s,23} S_{23} + g_{s,23}^* S_{23}^\dagger \right).
\]

(E.2)

Fortunately, \( a_p \) commutes with all the system operators except \( a_p^\dagger \). Therefore, only two of the commutation terms in this equation are non-zero. Using the fact that \( [a_p, a_p^\dagger] = 1 \), the first term on the right of Eq. (E.1) is therefore

\[
-\frac{i}{\hbar}[a_p, H_{\text{sys}}] = -i\omega_p a_p + g_{p,13} S_{13} + g_{p,13}^* S_{13}^\dagger + g_{p,23} S_{23} + g_{p,23}^* S_{23}^\dagger.
\]

(E.3)
The first of the terms in this equation represents harmonic oscillation of the $a_p$ operator in time. The rest of the terms represent the ability of this operator to exchange energy with the atoms via either the 1–3 or 2–3 transitions. If the RWA had been used, then only the first two terms on the right would be present.

Using Eqs. (3.12) – (3.15), the second term on the right side of Eq. (E.1) can also be written out explicitly to give

\[
- \sum_{j=1}^{8} [a_p, c_j^\dagger] \left\{ \frac{1}{2} \gamma_j c_j - \sqrt{\gamma_j} b_j^{\text{in}} \right\} = -[a_p, a_p^\dagger] \{ \kappa_p a_p - \sqrt{2\kappa_p} a_p^{\text{in}} \} -[a_p, a_s^\dagger] \{ \kappa_s a_s - \sqrt{2\kappa_s} a_s^{\text{in}} \} -[a_p, S_{12}^t] \left\{ \frac{1}{2} \gamma_{21} S_{12} - \sqrt{\gamma_{21}} B_{12}^{\text{in}} \right\} -[a_p, S_{13}^t] \left\{ \frac{1}{2} \gamma_{31} S_{13} - \sqrt{\gamma_{31}} B_{13}^{\text{in}} \right\} -[a_p, S_{23}^t] \left\{ \frac{1}{2} \gamma_{32} S_{23} - \sqrt{\gamma_{32}} B_{23}^{\text{in}} \right\} -[a_p, S_{11}] \left\{ \frac{1}{2} \gamma_{11} S_{11} - \sqrt{\gamma_{11}} B_{11}^{\text{in}} \right\} -[a_p, S_{22}] \left\{ \frac{1}{2} \gamma_{22} S_{22} - \sqrt{\gamma_{22}} B_{22}^{\text{in}} \right\} -[a_p, S_{33}] \left\{ \frac{1}{2} \gamma_{33} S_{33} - \sqrt{\gamma_{33}} B_{33}^{\text{in}} \right\}. \tag{E.4}
\]

Again, since the annihilation operator commutes with all other operators except the creation operator of the same mode, only the first term on the right side of this equation is non-zero. In fact, the entire third term of Eq. (E.1) is zero for the same reason. Using this and Eqs. (E.3) and (E.4), the quantum Langevin equation for the pump annihilation operator is then given by

\[
\dot{a}_p = -\kappa_p a_p - i\omega_p a_p + g_{p,13} S_{13} + g_{p,13}^* S_{13}^\dagger + g_{p,23} S_{23} + g_{p,23}^* S_{23}^\dagger + \sqrt{2\kappa_p} a_p^{\text{in}}, \tag{E.5}
\]
In its current state, $a_p$ will exhibit slow variations in time near the cavity mode resonance at $-\omega_c^p$. One typically transforms to a rotating coordinate system in order to extract this slowly varying behavior from the rapidly oscillating carrier frequency.

Using the pump coordinate transformation given in Eq. (3.18) and a similar expression for the input operator, Eq. (E.5) becomes

$$\dot{a}_p' e^{-i\omega_p t} = -(\kappa_p + i\Delta_p) a_p' e^{-i\omega_p t}$$

$$+ g_{p,13} S_{13} + g_{p,23}^* S_{23}^\dagger + g_{p,23} S_{23}^\dagger + \sqrt{2\kappa_p} a_p'_{\text{in}} e^{-i\omega_p t}, \quad (E.6)$$

where $\Delta_p \equiv \omega_c^p - \omega_p$ is the detuning of the field operator frequency from the cavity mode resonance. For the reasons discussed in the second section of Chapter 3, only the components of $S_{13}$ and $S_{23}$ that produce field oscillations near the cavity mode resonance at $-\omega_c^p$ are significant. The other components will be detuned by optical frequencies from the cavity resonance and will therefore be highly suppressed. With this understanding, Eq. E.6 becomes

$$\dot{a}_p' e^{-i\omega_p t} = -(\kappa_p + i\Delta_p) a_p' e^{-i\omega_p t}$$

$$+ g_{p,13} S_{13} (-\omega_p) e^{-i\omega_p t} + g_{p,13}^* \left(S_{13}^{(+\omega_p)}\right)^\dagger e^{-i\omega_p t}$$

$$+ g_{p,23} S_{23} (-\omega_p) e^{-i\omega_p t} + g_{p,23}^* \left(S_{13}^{(+\omega_p)}\right)^\dagger e^{-i\omega_p t} + \sqrt{2\kappa_p} a_p'_{\text{in}} e^{-i\omega_p t}, \quad (E.7)$$

All the coefficients are now slowly varying in time. The exponential factor can be divided out of each term and $a_p'$ can be replaced by $a_p$ for simplicity of notation. This leaves the result given in Eq. (3.21).
APPENDIX F

1-3 COHERENCE LANGEVIN EQUATION
This appendix is devoted to deriving the quantum Langevin equation for the 1–3 coherence operator using the Heisenberg equation of motion. Derivation of the $S_{23}$ equation follows analogously.

Choosing the fourth element $(S_{13})$ in the array given in Eq. (??), the Heisenberg equation of motion in the form of Eq. (3.11) becomes

$$\dot{S}_{13} = -\frac{i}{\hbar} [S_{13}, H_{\text{sys}}] - \sum_{j=1}^{8} [S_{13}, c_j] \left\{ \frac{1}{2} \gamma_j c_j - \sqrt{\gamma_j} b_j^{\text{in}} \right\}$$

$$+ \sum_{j=1}^{8} \left\{ \frac{1}{2} \gamma_j c_j^\dagger - \sqrt{\gamma_j} b_j^{\text{in}} \right\} [S_{13}, c_j],$$

(F.1)

with the $c_j$'s, $\gamma$'s and $b_j$'s defined by Eqs. (3.12) - (3.15). Using Eqs. (3.2) and (3.3), the commutation in the first term on the right side of Eq. (F.1) can be written

$$[S_{13}, H_{\text{sys}}] = \hbar \omega_1 [S_{13}, S_{11}] + \hbar \omega_2 [S_{13}, S_{22}] + \hbar \omega_3 [S_{13}, S_{33}]$$

$$+ \hbar \omega_p^c [S_{13}, a_p^\dagger a_p] + \hbar \omega_p^e [S_{13}, a_s^\dagger a_s]$$

$$+ i \hbar \left( a_p^\dagger - a_p \right) \left( g_{p,13} [S_{13}, S_{13}] + g_{p,13}^* [S_{13}, S_{13}^\dagger] \right)$$

$$+ \hbar \omega_p^c [S_{13}, a_p^\dagger a_p] + \hbar \omega_p^e [S_{13}, a_s^\dagger a_s]$$

$$+ i \hbar \left( a_s^\dagger - a_s \right) \left( g_{s,13} [S_{13}, S_{13}] + g_{s,13}^* [S_{13}, S_{13}^\dagger] \right)$$

$$+ \left( g_{p,23} [S_{13}, S_{23}] + g_{p,23}^* [S_{13}, S_{23}^\dagger] \right).$$

(F.2)

Noting that $S_{13}$ commutes with all field operators, and using the commutation relations for the atomic operators given in Eq. (3.10), the first term on the right of
Eq. (F.1) is therefore

\[
-\frac{i}{\hbar} [S_{13}, H_{\text{sys}}] = -i (\omega_3 - \omega_1) S_{13} + (a_p^\dagger - a_p) \left[ g^*_{p,13} (S_{11} - S_{33}) + g^*_{p,23} S_{12} \right]
+ (a_s^\dagger - a_s) \left[ g^*_{s,13} (S_{11} - S_{33}) + g^*_{s,23} S_{12} \right].
\] (F.3)

Using Eqs. (3.12) - (3.15), the second term on the right side of Eq. (F.1) can also be written out explicitly to give

\[
- \sum_{j=1}^{8} [S_{13}, c_j^\dagger] \left\{ \frac{1}{2} \gamma_j c_j - \sqrt{\gamma_j} b_j^{\text{in}} \right\} = -[S_{13}, a_p^\dagger] \left\{ \kappa_p a_p - \sqrt{2\kappa_p} a_p^{\text{in}} \right\} \\
- [S_{13}, a_s^\dagger] \left\{ \kappa_s a_s - \sqrt{2\kappa_s} a_s^{\text{in}} \right\} \\
- [S_{13}, S^\dagger_{12}] \left\{ \frac{1}{2} \gamma_{21} S_{12} - \sqrt{\gamma_{21}} B_{12}^{\text{in}} \right\} \\
- [S_{13}, S^\dagger_{13}] \left\{ \frac{1}{2} \gamma_{31} S_{13} - \sqrt{\gamma_{31}} B_{13}^{\text{in}} \right\} \\
- [S_{13}, S^\dagger_{23}] \left\{ \frac{1}{2} \gamma_{32} S_{23} - \sqrt{\gamma_{32}} B_{23}^{\text{in}} \right\} \\
- [S_{13}, S_{11}] \left\{ \frac{1}{2} \gamma_{11} S_{11} - \sqrt{\gamma_{11}} B_{11}^{\text{in}} \right\} \\
- [S_{13}, S_{22}] \left\{ \frac{1}{2} \gamma_{22} S_{22} - \sqrt{\gamma_{22}} B_{22}^{\text{in}} \right\} \\
- [S_{13}, S_{33}] \left\{ \frac{1}{2} \gamma_{33} S_{33} - \sqrt{\gamma_{33}} B_{33}^{\text{in}} \right\}.
\] (F.4)

Again using the atomic commutation relations and multiplication rules, the last expression can be simplified to

\[
- \sum_{j=1}^{8} [S_{13}, c_j^\dagger] \left\{ \frac{1}{2} \gamma_j c_j - \sqrt{\gamma_j} b_j^{\text{in}} \right\} = -\sqrt{\gamma_{21}} S_{23} B_{12}^{\text{in}} - \frac{1}{2} \gamma_{31} S_{13} + \sqrt{\gamma_{31}} (S_{11} - S_{33}) B_{13}^{\text{in}} \\
- \frac{1}{2} \gamma_{32} S_{13} + \sqrt{\gamma_{32}} S_{12} B_{23}^{\text{in}} - \sqrt{\gamma_{11}} S_{13} B_{11}^{\text{in}} \\
- \frac{1}{2} \gamma_{33} S_{13} + \sqrt{\gamma_{33}} S_{13} B_{33}^{\text{in}}.
\] (F.5)
The final term on the right side of Eq. (F.1) can be treated in the same manner, with
the result
\[ \sum_{j=1}^{8} \left\{ \frac{1}{2} \gamma_j c_j^* - \sqrt{\gamma_j} b_j^{\text{int}} \right\} [S_{13}, c_j] = -\frac{1}{2} \tilde{\gamma}_{11} + \sqrt{\gamma_{11}} B_{11}^{\text{int}} S_{13} - \sqrt{\gamma_{33}} B_{33}^{\text{int}} S_{13} \]  \hspace{1cm} (F.6)

Combining Eqs. (F.3), (F.5), and (F.6) finally gives

\[ \dot{S}_{13} = -[\gamma_{31} + i(\omega_3 - \omega_1)] S_{13} + (a_p^\dagger - a_p) \left[ g_{p,13}^* (S_{11} - S_{33}) + g_{p,23}^* S_{12} \right] \\
\hspace{1cm} + (a_s^\dagger - a_s) \left[ g_{s,13}^* (S_{11} - S_{33}) + g_{s,23}^* S_{12} \right] \\
\hspace{1cm} - \sqrt{\gamma_{21}} S_{23} B_{12}^{\text{in}} + \sqrt{\gamma_{31}} (S_{11} - S_{33}) B_{13}^{\text{in}} + \sqrt{\gamma_{32}} S_{12} B_{23}^{\text{in}} \\
\hspace{1cm} - \sqrt{\gamma_{11}} S_{13} B_{11}^{\text{in}} + \sqrt{\gamma_{33}} S_{13} B_{33}^{\text{in}} + \sqrt{\gamma_{11}} B_{11}^{\text{int}} S_{13} - \sqrt{\gamma_{33}} B_{33}^{\text{int}} S_{13}, \]  \hspace{1cm} (F.7)

where the decay constants have been consolidated as given in Eq. (3.29). Inserting
the rotating versions of \( a_p, a_s, \) and \( S_{12} \) from Eq. (3.18) yields the result given by
Eq. (3.26), as advertised.
APPENDIX G

ANTI-STOKES THEORY
The goal of this appendix is to give a streamlined treatment of the effect of cavity enhancing the anti-Stokes field. It is shown that the anti-Stokes generation suppresses the Stokes gain. However, it is also shown that this Stokes gain suppression, which has traditionally been avoided in the past, actually provides the means for efficient conversion to anti-Stokes. In fact, it is shown that efficiencies approaching the fundamental quantum limit (50%) are possible in the cw regime. The treatment given here begins from a fully quantum mechanical foundation, but quickly reverts to the semiclassical limit. The quantum optical aspects of this process are therefore left for someone else to iron out.

Previous Work

For quite some time there has been a concerted effort to improve the efficiency with which a pump beam can be frequency up-converted into an anti-Stokes beam through Raman-assisted four-wave-mixing (FWM). In the pulsed laser regime this effort has involved techniques such as temporal pulse shaping, spatial focusing, and off-axis Stokes seeding [146, 147, and references therein]. Alternatively, researchers have significantly improved the up-conversion efficiency of a probe pulse by initially preparing the atomic medium with a near-maximal coherence [148, 149, 150]. Although the maximal-coherence method (a form of electromagnetically-induced transparency) has exhibited unity conversion efficiency for near-resonance systems [148, 149] and boasts the very attractive feature of automatic phase matching, it often requires additional
strong laser fields to establish the maximal atomic coherence. The net conversion efficiency (including all beams) reported in these studies was lower than that achieved through the more conventional methods (i.e. those methods that did not prepare the atomic medium). I am aware of one previous continuous-wave (cw) anti-Stokes conversion study, which exploited self-oscillation to generate a maximum combined power conversion efficiency into Stokes and anti-Stokes of 4% [151]. There is also a very small amount of anti-Stokes light that accompanies the Stokes generation in a cw Raman laser [30]. In these systems, the HFC mirrors were not highly reflective at the anti-Stokes wavelength, so the light was not resonantly enhanced.

This appendix describes how it is possible to use the cavity-enhancement techniques to approach the fundamental quantum limit of Raman-resonant anti-Stokes conversion through the use of cw cavity-enhanced four-wave mixing (FWM). More specifically it is shown that a photon conversion efficiency approaching 50% is possible from a cw pump beam into a single cw anti-Stokes order using a gaseous Raman medium within a triply-resonant high-finesse cavity. In a related work, Shinzen and coworkers have numerically studied the possibility of using cavity enhancement of anti-Stokes orders to generate highly repetitive optical pulses [152].

**Hamiltonian**

The system considered here is composed of three strongly-enhanced field modes interacting with a collection of three-level atoms, each in a Λ configuration as shown
in Fig. 64(a). All the fields are detuned very far from the single-photon transitions, but here it is assumed that both of the two-photon Raman transitions are resonant.

Figure 64. To-scale energy level diagram showing (a) the pertinent atomic levels, fields (α’s) and single-photon detunings (Δ’s), and (b) the energy flow for the overall anti-Stokes generation process.

Higher orders of Stokes and anti-Stokes are assumed to fall outside the bandwidth of the mirror reflectivities, and are therefore neglected. The mathematical description corresponding to the atoms, fields, and the interactions shown is given by the total Hamiltonian is (in the electric dipole and rotating wave approximations)

\[ H = H_{\text{sys}} + H_{\text{irrev}} + H_{\text{baths}}, \]  

where the components of \( H \) are given as follows.

\( H_{\text{sys}} \) represents the energy associated with the free energy of the system operators
and their mutual interactions, and is given by

\[ H_{\text{sys}} = \sum_{i=1}^{3} \hbar \omega_i S_{ii} + \sum_{q=p,s,as} \hbar \omega_q a_q ^\dagger a_q + i \hbar \left( g_{p,13} a_1 ^\dagger S_{13} - \text{H.c.} \right) + i \hbar \left( g_{s,23} a_2 ^\dagger S_{23} - \text{H.c.} \right) + i \hbar \left( g_{as,13} a_{as} ^\dagger S_{13} - \text{H.c.} \right) , \]

where \( \hbar \omega_i \) and \( S_{ii} \) are the energy and collective population operator for the \( i \)-th atomic level, respectively, while \( S_{ij} \) and \( g_{q,ij} \) represent the collective coherence operator and the atom-field coupling constant for the traveling wave field mode \( q \) driving the \( i-j \) atomic transition. The \( a \)'s refer to annihilation operators while \( \omega_c \)'s refer to empty cavity mode frequencies. Throughout this work, the subscript \( \text{"p"} \) refers to the pump, \( \text{"s"} \) refers to the Stokes, and \( \text{"as"} \) refers to the anti-Stokes. Note that the rotating wave approximation (RWA) is used here, but is not valid. As shown in the main chapter, not invoking the RWA only serves to scale the gain; it will not change the qualitative behavior of the system. Also, there will be single photon interactions due to the Stokes field interacting with the 1-3 transition and the anti-Stokes interacting with the 2-3 transition, but these are neglected because they are far-off resonance, just as in the main chapter.

For brevity, \( H_{\text{irrev}} \) and \( H_{\text{baths}} \) are not shown explicitly, but include the standard interactions between reservoir operators and each of the system operators as in the main chapter. These interactions generate decay and noise. In the following, the semiclassical limit is taken, which eliminates the quantum noise terms.
Time-Dependent Semiclassical Equations

Following the same procedure as in the derivation of the Raman equations in the main text, one can use the Hamiltonian and the commutation relations to generate a set of quantum Langevin equations; one for each system operator. As with the Raman derivation, the large single-photon detunings present in the system allow one to perform significant simplifications to the resulting Langevin equations. With these, and taking the semiclassical limit of the remaining system operator Langevin equations (ignore the noise terms with zero mean), one obtains the following time-dependent equations for the slowly-varying complex fields:

\[
\begin{align*}
\dot{\alpha}_p &= -\kappa_p \alpha_p - G_1 |\alpha_s|^2 \alpha_p + G_2 |\alpha_{as}|^2 \alpha_p + \tau_{rt}^{-1} \sqrt{T_{p,0}} \alpha_p^{in}, \\
\dot{\alpha}_s &= -\kappa_s \alpha_s + G_1 |\alpha_p|^2 \alpha_s + G_{12} \alpha_p^2 \alpha_{as}^* \\
\dot{\alpha}_{as} &= -\kappa_{as} \alpha_{as} - G_2 |\alpha_p|^2 \alpha_{as} - G_{12} \alpha_p^2 \alpha_s^*,
\end{align*}
\]

where the \( \alpha \)'s are unitless complex field amplitudes, \( \kappa \)'s are cavity decay rates, \( T_{p,0} \) is the power transmission of the input coupler mirror at the pump wavelength, \( \tau_{rt} \) is the round-trip time in the cavity, and the \( G \)'s are gains; \( G_1 \) for the pump \( \leftrightarrow \) Stokes Raman process, \( G_2 \) for the anti-Stokes \( \leftrightarrow \) pump Raman process and \( G_{12} \) for the FWM process. Because exact Raman resonance is assumed, \( G_1 \) and \( G_2 \) are real, but \( G_{12} \) can still be complex. The large single-photon detunings [see Fig. 64(a)] cause these gains to be insensitive to the pump laser frequency. Detunings of the fields from the cavity resonances are ignored.
It is also instructive to include the two lower level populations, even though they are so small that they don’t appreciably affect the fields for this system. These are given by

\[ \dot{J}_{11} = \gamma_{21} J_{22} - 2 G_1 |\alpha_s|^2 |\alpha_p|^2 - 2 G_2 |\alpha_p|^2 |\alpha_{as}|^2 \]

\[ - [2 G_{12} \alpha_p^2 \alpha_s^* \alpha_{as}^* + c.c.] , \quad (G.6) \]

\[ \dot{J}_{22} = -\dot{J}_{11}. \quad (G.7) \]

where \( \gamma_{21} \) is the population decay rate from level 2.

It is worth noting here that only Raman-type gain terms influence the pump field dynamics given by Eq. (H.1). Also note that there is a FWM contribution to the populations given by Eqs. (G.6) and (G.7). This evidence supports the assertion by Bobbs and Warner [153] that Raman resonant FWM involves energy exchange with the medium. Therefore, the more detailed energy level diagram given in Fig. 64(b) that shows the direction of energy flow does not correctly describe the FWM contributions alone. However, it will soon be shown that when all terms (FWM and Raman) are included, one can correctly interpret the energy flow as shown in Fig. 64(b).

One can show that the \( G \)'s depend on the spatial overlaps of the cavity modes involved with each process. The spatial overlap in \( G_{12} \) generates a phase-matching term, as expected. However, in order to extract as much physical insight as possible, from these equations, perfect phase matching and spatial overlap are assumed for the moment. Mathematically, this means that \( G_1 G_2 = |G_{12}|^2 \). Departures from this
idealization are considered later.

Steady-State Solutions

With this simplification, and by setting Eqs. (H.1) - (G.5) to zero, the steady-state unitless powers above threshold can be expressed in the remarkably simple forms

\[
|\alpha_p|^2 = \frac{1}{(1 - x) G_1^2} \kappa_a
\]

\[
|\alpha_s|^2 = \frac{1}{(1 - x^2) G_1} \left( \sqrt{r_p} - 1 \right)
\]

\[
|\alpha_{as}|^2 = x \frac{\kappa_s}{\kappa_{as}} |\alpha_s|^2
\]

where \( r_p \equiv |\alpha_p^{in}|^2/|\alpha_p^{th}|^2 \) is the incident unitless pump power normalized to threshold, which is given by

\[
|\alpha_p^{th}|^2 = \frac{1}{(1 - x) G_1 T_{p,0}} \frac{\kappa_p^2 \kappa_s T_{rt}^2}{\kappa_{as}}
\]

and the critical parameter \( x \) is defined as

\[
x = \frac{G_2}{G_1} \frac{\kappa_a}{\kappa_{as}} = \frac{G_2}{G_1} \frac{\ln(R_{s,rt})}{\ln(R_{as,rt})}
\]

where the \( R_{rt} \)'s are round-trip power reflectivities. We have used the fact that \( \kappa = -\ln(R_{rt})/2\tau_{rt} \).

There are three important aspects of Eqs. (H.2)-(G.12) that deserve discussion. First, the parameter \( x \) can be interpreted as the relative effective gain for the two cavity-enhanced Raman processes present in this system. Experimentally, one can therefore adjust the parameter \( x \) by changing the relative reflectivities of the cavity
mirrors at the two wavelengths. Next, it is emphasized that when \( x \ll 1 \), corresponding to no anti-Stokes cavity enhancement, the equations directly match the theoretical [27, 154] and experimental [27] behavior of cw Raman lasers from previous work.

The third important aspect can be observed most directly from Eq. (G.11). Specifically, as \( x \) approaches unity, the Stokes laser threshold increases dramatically. This is a direct manifestation of a Raman gain suppression phenomenon that was predicted by Bloembergen and Shen long ago [155] and has since been observed experimentally by several groups in the pulsed laser regime [156, 157]. In this cavity-enhanced version of the phenomenon, complete Stokes gain suppression occurs (for perfect phase matching and mode overlap) when the cavity-enhanced gains for the two Raman processes are equal \( (x = 1) \).

Although enhancement of the anti-Stokes field can therefore suppress the Stokes gain, one finds that this gain suppression is also directly associated with an increased conversion efficiency to anti-Stokes photons. In order to obtain photon conversion efficiencies for this system, one can convert the intracavity powers given by Eqs. (G.9) and (G.10) into powers exiting the cavity (multiply by the output coupler mirror transmissivity). It is then straightforward to show that the peak conversion efficiency for both the Stokes and the anti-Stokes occurs for a pump rate of four times the Stokes laser threshold \( (r_p = 4) \). At this pump rate, the peak conversion efficiencies
are given by

\[
\eta_{s,\text{peak}} = \frac{|\alpha_{\text{out}}^0|^2}{4|\alpha_p^{\text{th}}|^2},
\]

\[
= \frac{1}{(1 + x)} \frac{T_{p,0}}{(1 - R_{p,rt}) (1 - R_{s,rt})},
\]

\[
\eta_{as,\text{peak}} = \frac{|\alpha_{as}^{\text{out}}|^2}{4|\alpha_p^{\text{th}}|^2},
\]

\[
= \frac{x}{(1 + x)} \frac{T_{p,0}}{(1 - R_{p,rt}) (1 - R_{as,rt})},
\]

\[\text{(G.13)}\]

\[\text{(G.14)}\]

where the power reflectivities have been assumed nearly unity \((- \ln R_{rt} \approx 1 - R_{rt})\).

The conversion efficiencies, with the peak values given by Eqs. (G.13) and (G.14), are plotted as functions of the pump rate in Fig. 65 for several different values of the critical parameter \(x\). For the plots, we have assumed that the input/output coupler dominates the other mirror losses in the cavity \([T_0 \approx (1 - R_{rt})]\). As the ratio \(x\) is increased from near zero to unity, the peak Stokes conversion falls from 100% to 50%, while the peak anti-Stokes conversion grows from zero to 50%. Therefore, when the cavity-enhanced gains for the two Raman processes are nearly equal \((x \approx 1)\), the equations predict a conversion efficiency that approaches the fundamental quantum limit for anti-Stokes generation via the Raman-resonant FWM process. That is, a pair of pump photons is exchanged for one Stokes photon and one anti-Stokes photon, as shown in Fig. 64(b).
Figure 65. Photon conversion efficiencies for (a) the Stokes and (b) the anti-Stokes fields as functions of the pump rate. Curves are given for several different values of the relative gain parameter $x$, and for idealized experimental conditions.

However, Eq. (G.11) emphasizes one of the obstacles in experimentally obtaining such high conversion. Specifically, although Fig. 64(b) illustrates that a value of $x$ approaching unity is desirable for high conversion to anti-Stokes, this limit also corresponds to a prohibitive increase in the Stokes laser threshold, making it more difficult to achieve the four-times-threshold condition that is required for optimal conversion. It is interesting that the Stokes gain suppression that was traditionally avoided in the production of anti-Stokes in the past can now be interpreted as actually providing the means for anti-Stokes conversion in this system.

One can view the gain suppression from several other angles as well. For instance, one can interpret this effect as a form of electromagnetically-induced transparency. That is, without the anti-Stokes enhancement, the input pump light is absorbed and converted to Stokes. But by simply making the cavity resonant for the anti-Stokes, the Stokes laser threshold increases, causing the medium to be transparent to the pump
wavelength. Furthermore, one can also view this process as quantum interference. The two Raman processes that exist in Fig. 64 represent two separate paths between the lower two states. During gain suppression, these pathways interfere destructively to eliminate the 1–2 coherence that would exist if only one pathway were present.

To elucidate the relationship between the rising threshold and the increasing conversion, Fig. 66(a) shows the Stokes laser threshold normalized to its value for $x = 0$ and Fig. 66(b) shows the anti-Stokes peak photon conversion efficiency, both as functions of the parameter $x$. The solid curves (labeled A) are from Eqs. (G.11) and (G.14) and are associated with the idealized case at hand for which spatial concerns and mirror losses are neglected. The dotted curves (labeled B and C) represent departures from these ideal conditions and are discussed later. From the solid curves one can see that, despite the prohibitive threshold increase as $x$ approaches unity, substantial conversion can still be achieved for slightly lower values of $x$, corresponding to very mild threshold increases. For instance, when $x = 0.5$, the threshold only doubles relative to that when $x = 0$, but the conversion efficiency reaches 33%.
Figure 66. Plots showing (a) the normalized laser threshold and (b) the photon conversion efficiency as functions of the relative gain parameter $x$. The solid curves represent idealized conditions, the dotted curves labeled B include imperfect spatial overlap and phase matching, and the dotted curves labeled C additionally include mirror losses.

Experimental Realization

In the analysis up to this point, several simplifying assumptions have been made in order to clarify the physical processes involved. In reality, imperfect phase matching, spatial overlap, and mirror reflectivities can limit anti-Stokes conversion in addition to the increased threshold. The mathematics are similar, but a bit more algebraically involved when these debilitating effects are included. For the sake of brevity, the modified versions of the above equations are not shown. However, the predicted
results for realistic experimental conditions are provided below.

For an initial experiment, a relatively small Raman shift (rotational transition in diatomic hydrogen gas [158]) can be used so that all three fields (pump, Stokes, and anti-Stokes) can be simultaneously cavity-enhanced within the bandwidth of a single mirror coating. The small shift and gaseous medium also allow near-perfect phase matching for the proposed cavity length of 30cm. Phase matching and imperfect mode overlap will reduce the maximum conversion by less than 15% for a traveling-wave cavity (much greater for a standing-wave cavity) containing 10 atmospheres of gas pressure. Therefore, a four-mirror bow-tie ring cavity is considered with three mirrors coated as highly reflective as possible. Current technology allows for mirrors with combined transmission and absorption losses below ~10 ppm.

The requirements placed on the final input/output coupler mirror collectively represent the most significant technical challenge of experimentally realizing this system. The reason for this is as follows: The pump laser will be phase/frequency stabilized to a cavity resonance and the Stokes field will build automatically on the cavity resonance nearest to its gain peak, as discussed in Appendix D. Now, in the absence of dispersion, the anti-Stokes field that is generated by the pump and Stokes fields will automatically fall exactly on a resonance of the HFC, as depicted in Fig. 67(a). However, the dispersion of the hydrogen gas will prevent the generated anti-Stokes frequency from automatically matching a cavity resonance, as depicted in Fig. 67(b). The dispersion is exaggerated in the figure. In reality, the generated anti-Stokes is
offset from the cavity resonance by $\sim 50 - 100 MHz$. The two-photon dispersion is too weak to compensate for this and the offset is too small to bump it to the next longitudinal mode. One thought is to adjust the cavity length and mirror curvature until the anti-Stokes generation occurs on a higher order (spatial) resonance of the cavity. Aside from tricks like this, one must require that the input/output coupler compensate for this dispersion.

Figure 67. Schematic illustrating how dispersion can inhibit the generation of anti-Stokes. When no dispersion exists (a), the anti-Stokes is automatically generated on a cavity resonance. However, due to dispersion in the Raman medium, the anti-Stokes frequency does not automatically match that of a cavity resonance.

Fortunately, such dispersion-compensated or “chirped” mirrors have recently been
realized and have received a good deal of attention due to their roll in the generation of femtosecond laser pulses [159]. In light of these recent advances in mirror technology, it is assumed that an input/output mirror can be manufactured to compensate for the dispersion of hydrogen gas, while still exhibiting a 99.95% reflectivity and 60 ppm loss. This mirror loss is a conservative estimate from our experience with mirrors coated separately for two disparate wavelengths [34], but is a bit lower than other chirped-mirror loss estimates [159]. Note that the gas pressure and the reflection angle can be used to fine tune the dispersion compensation and therefore to coerce the anti-Stokes field into resonance.

The dotted curves in Figs. 66(a) and 66(b) show the resulting normalized threshold and peak anti-Stokes photon conversion when spatial concerns (curve b) and, additionally, mirror losses (curve c) are included. Note that the imperfect phase matching prevents complete gain suppression from ever occurring, but that the peak conversion still occurs for $x = 1$. In the absence of anti-Stokes enhancement ($x \approx 0$), the laser threshold would be under 2.5mW. With the $\sim 9$-fold increase in the laser threshold that is due to anti-Stokes enhancement at $x = 1$ and mirror losses, anti-Stokes conversion can exceed 30% (occurring at four times threshold) with less than 100mW of injected pump light. The luxury of greater available pump power can afford conversion efficiencies that approach the fundamental limit of 50% by reducing the cavity length and increasing the input/output coupler transmissivity relative to its loss.
In closing it is noted that, due to the high-finesse cavity enhancement, the first anti-Stokes order will build in a single high-quality spatial mode and will exhibit a predicted linewidth in the kHz range. The experimental conditions and techniques that are required to achieve this system are realistic and have already been proven in related work, with the notable exception of the low-loss chirped mirror. Moreover, by using a frequency-doubled Nd:Yag pump source to drive the vibrational shift in deuterium gas, this technique is very attractive as a simultaneous generator of red (Stokes = 633nm), blue (anti-Stokes = 459nm), and green (pump = 532nm) for color projection systems. The experimental realization of this system will therefore not only offer a probe into the fundamental limits and gain suppression subtleties of Raman-resonant FWM, it will also provide a practical method of efficiently up-converting the frequency of visible and near-infrared cw laser light.
APPENDIX H

SIMPLIFIED PUMP EQUATION
This appendix is devoted to deriving the simplified pump equation given in Eq. (3.38) from Eq. (3.21). Derivation of the Stokes equation follows analogously.

Setting Eqs. (3.26) and the $S_{23}$ counterparts to zero (these coherences can be adiabatically eliminated due to the large detunings present) and plugging the steady-state Fourier components of the coherences into Eq. (3.21) yields

$$\dot{a}_p = - (\kappa_p + i \Delta_p) a_p - \frac{g_{p,13}}{(\gamma_{31} + i \Delta)} \left[ g_{p,13}^* a_p (S_{11} - S_{33}) + g_{s,23}^* a_s S_{12} + F_{13}^{(-\omega_p)} \right]$$

$$+ \frac{g_{p,13}^*}{(\gamma_{31} - i \Delta_{as})} \left[ g_{p,13} a_p (S_{11} - S_{33}) + \left( F_{13}^{(+\omega_p)} \right)^\dagger \right]$$

$$- \frac{g_{p,23}}{(\gamma_{32} + i \Delta_{as})} \left[ g_{p,23}^* a_p (S_{22} - S_{33}) + F_{23}^{(-\omega_p)} \right]$$

$$+ \frac{g_{p,23}^*}{(\gamma_{32} - i \Delta')} \left[ g_{p,23} a_p (S_{22} - S_{33}) + g_{s,13} a_s S_{12} + \left( F_{23}^{(+\omega_p)} \right)^\dagger \right]$$

$$+ \sqrt{2\kappa_p} a_p^{in}. \quad (H.1)$$

It is now possible to neglect all $F_{13}$ and $F_{23}$ components compared to the input pump noise due to the large detunings in the denominators. This can be shown rigorously by either retaining the noise terms to the end or by calculating the second order correlation functions of the noise terms in Eq. (H.1) at this point. Comparison of the correlations from each of the noise terms reveals that the input pump field operator noise dominates. Performing this, noting that the $\Delta'$s are much greater than any $\gamma$'s
and collecting terms yields

\[ \dot{a}_p = -\left\{ \kappa_p + |g_{p,13}|^2 \gamma_{31} \left[ \frac{1}{\Delta^2} - \frac{1}{(\Delta'_{as})^2} \right] (S_{11} - S_{33}) + |g_{p,23}|^2 \gamma_{32} \left[ \frac{1}{\Delta_{as}^2} - \frac{1}{(\Delta')^2} \right] (S_{22} - S_{33}) \right\} a_p \\
- i \left\{ \Delta_p - |g_{p,13}|^2 \left[ \frac{1}{\Delta} - \frac{1}{\Delta'_{as}} \right] (S_{11} - S_{33}) - |g_{p,23}|^2 \left[ \frac{1}{\Delta_{as}} - \frac{1}{\Delta'} \right] (S_{22} - S_{33}) \right\} a_p \\
- g_{p,13} g_{s,23}^* \left[ \frac{1}{(\gamma_{31} + i\Delta)} - \frac{1}{(\gamma_{32} - i\Delta')} \right] a_x S_{12} + \sqrt{2\kappa_p a_p^{in}} \right\} , \quad (H.2) \]

where the fact that \( g_{p,13} g_{s,23}^* = g_{s,13} g_{p,23}^* \) has been used based on Eq. (??). In this form, it becomes clear that the second and third terms in the first curly brackets will effectively increase the cavity decay rate in proportion to the population differences. These single-photon interactions therefore serve to broaden the cavity linewidth. However, since \( \Delta^2 \kappa_p \gg \gamma_{31} |g_{p,13}|^2 \), the cavity decay \( (\kappa_p) \) dominates any such broadening and the second and third terms in the first curly brackets can be ignored.

Along similar lines, the second and third terms in the second curly brackets effectively alter the cavity detuning, \( \Delta_p \). These terms therefore "pull" the frequency of the cavity mode. Such mode pulling can actually be considerable compared to the cavity linewidth. However, in the present system the active electronic locking will nullify such pulling effects and lock the pump laser's frequency to the active cavity line center. The entire cavity detuning term is therefore neglected.

Finally, since \( \Delta \) and \( \Delta' \) are much greater than \( \gamma_{31} \) and \( \gamma_{32} \) one can make the approximation \( (\gamma_{31} + i\Delta)^{-1} \approx (i\Delta)^{-1} \). This actually isn't necessary, but it simplifies
future calculations quite a bit and the results come out the same even if the real parts are retained. Applying all the above simplifications to Eq. (H.2) yields Eq. (3.38).
APPENDIX I

SIMPLIFIED $S_{12}$ EQUATION
This appendix is devoted to deriving the simplified $S_{12}$ equation given in Eq. (3.40) from Eq. (3.23). The simplifications used are very similar to those used in Appendix H.

Setting Eqs. (3.26) and the $S_{23}$ counterparts to zero (these coherences can be adiabatically eliminated due to the large detunings present) and plugging the steady-state Fourier components of the coherences into Eq. (3.23) yields

$$\dot{S}_{12} = -(\gamma_{s1} + i\Delta_{12})S_{12}$$

$$- \frac{a_{p}^{*}g_{p,23}}{(\gamma_{s1} + i\Delta_{ss})} \left[ g_{p,23}^{\ast} a_{p}S_{12} - F_{13}^{(-\omega_{ae})} \right]$$

$$- \frac{a_{p}^{*}g_{p,13}}{(\gamma_{s2} - i\Delta_{ss})} \left[ g_{p,13} a_{p}S_{12} + F_{23}^{(+\omega_{ae})} \right]$$

$$- \frac{a_{p}g_{p,23}}{(\gamma_{s1} + i\Delta')} \left[ g_{s,13} a_{s}^{\dagger}(S_{11} - S_{33}) + g_{p,23}^{\ast} a_{p}S_{12} + F_{13}^{(-\omega_{e})} \right]$$

$$- \frac{a_{p}g_{p,13}}{(\gamma_{s2} - i\Delta)} \left[ g_{s,23} a_{s}^{\dagger}(S_{22} - S_{33}) + g_{p,13} a_{p}S_{12} - (F_{23}^{(+\omega_{e})})^{\dagger} \right]$$

$$- \frac{a_{s}^{\dagger}g_{s,23}}{(\gamma_{s1} + i\Delta')} \left[ g_{p,23} a_{p}(S_{22} - S_{33}) + g_{s,23}^{\ast} a_{s}S_{12} - F_{13}^{(-\omega_{e})} \right]$$

$$- \frac{a_{s}^{\dagger}g_{s,13}}{(\gamma_{s2} - i\Delta') \left[ g_{p,23} a_{p}(S_{22} - S_{33}) + g_{s,13} a_{s}S_{12} + (F_{23}^{(+\omega_{e})})^{\dagger} \right]$$

$$- \frac{a_{s}g_{s,23}}{(\gamma_{s1} + i\Delta_{ss})} \left[ g_{s,23} a_{s}^{\dagger}S_{12} + F_{13}^{(+\omega_{ae})} \right]$$

$$- \frac{a_{s}g_{s,13}}{(\gamma_{s2} - i\Delta_{ss})} \left[ g_{s,13} a_{s}^{\dagger}S_{12} - (F_{23}^{(-\omega_{ae})})^{\dagger} \right]$$

$$+ F_{12},$$

(1.1)

As in Appendix H, one can neglect all $F_{13}$ and $F_{23}$ components compared to the $F_{12}$ due to the large detunings in the denominators. Performing this, and collecting terms
yields

\[
\dot{S}_{12} = -\left\{ \gamma_{21} + \left[ \frac{1}{\Delta'^2} + \frac{1}{\Delta^2} \right] \gamma_{31} \Omega_{p,23}^2 \\
+ \left[ \frac{1}{(\Delta'^2)} + \frac{1}{\Delta^2} \right] \gamma_{32} \Omega_{p,13}^2 \\
+ \left[ \frac{1}{(\Delta'^2)} + \frac{1}{\Delta^2} \right] \gamma_{31} \Omega_{s,23}^2 \\
+ \left[ \frac{1}{\Delta'^2} + \frac{1}{\Delta^2} \right] \gamma_{32} \Omega_{s,13}^2 \right\} S_{12} \\
- i \left\{ \Delta_{12} - \left[ \frac{1}{\Delta'_{as}} + \frac{1}{\Delta'} \right] \Omega_{p,23}^2 \\
+ \left[ \frac{1}{\Delta'_{as}} + \frac{1}{\Delta'} \right] \Omega_{p,13}^2 \\
- \left[ \frac{1}{\Delta'_{as}} + \frac{1}{\Delta'} \right] \Omega_{s,23}^2 \\
+ \left[ \frac{1}{\Delta'_{as}} + \frac{1}{\Delta'} \right] \Omega_{s,13}^2 \right\} S_{12} \\
- g_{p,13}^* g_{s,23} \Omega_{p,23}^2 \left\{ \left[ \frac{1}{(\gamma_{31} + i\Delta)} + \frac{1}{(\gamma_{31} + i\Delta')} \right] (S_{11} - S_{33}) \\
+ \left[ \frac{1}{(\gamma_{32} - i\Delta)} + \frac{1}{(\gamma_{32} - i\Delta')} \right] (S_{22} - S_{33}) \right\} \\
+ F_{12}, \quad (I.2)
\]

where the fact that \( g_{p,13}^* g_{s,23}^* = g_{s,13} g_{p,23}^* \) has been used based on Eq. (??) and the Rabi frequency has been identified as \( |\Omega_{p,13}|^2 = |g_{p,13}|^2 a_p^\dagger a_p \). In this form, it becomes clear that the second through the fifth terms in the first curly brackets will increase the atomic dephasing rate of the 1–2 transition, \( \gamma_{21} \), in proportion to the Rabi frequencies squared. These interactions therefore serve to broaden the 1–2 transition linewidth. However, since \( \Delta^2 \gamma_{21} \gg \gamma_{31} |\Omega_{p,13}|^2 \), the line broadening terms will not significantly alter \( \gamma_{21} \) and can be ignored.

Along similar lines, the second through the fifth terms in the second curly brackets
effectively alter the two-photon Raman detuning, $\Delta_{12}$, by shifting the atomic resonance. These so-called AC stark shifts are also proportional to the Rabi frequency (i.e. the stronger the fields, the larger the shift). The AC Stark shifts can be in the 10's of kHz range, but are still much smaller than the two-photon Raman linewidth and will therefore be neglected.

Finally, since $\Delta$ and $\Delta'$ are much greater than $\gamma_1$ and $\gamma_2$ one can make the approximation $(\gamma_1 + i\Delta)^{-1} \approx (i\Delta)^{-1}$. This actually isn't necessary, but it simplifies future calculations quite a bit and the results come out the same even if the real parts are retained. Applying all the above simplifications to Eq. (1.2) yields Eq. (3.40)
APPENDIX J

COHERENT STATE POWER SPECTRAL DENSITY
This appendix is devoted to deriving the power spectral density of a coherent state. The procedure is identical to that used to obtain Eq. (3.107), but with a different representation for $\delta I_n(t)$. For the sake of notational clarity, the superscript "out" is omitted in the following calculations. However, the calculation is equally valid for the internal field operator as given.

Specifically, with $I_n(t) = a_n a_n^\dagger$, and $a_n = \alpha_n + \delta a_n$, one obtains

$$I_n(t) = (\alpha_n + \delta a_n) (\alpha_n^* + \delta a_n^\dagger)$$

$$\approx |\alpha_n|^2 + \alpha_n \delta a_n^\dagger + \alpha_n^* \delta a_n, \quad (J.1)$$

so that

$$\delta I_n(t) = \alpha_n \delta a_n^\dagger + \alpha_n^* \delta a_n. \quad (J.2)$$

The key here is that this time, no change to amplitude/phase variables is performed. As will be shown, this will allow the commutation relations for the field fluctuation operators to be used. With this expression for the intensity fluctuation operator, the
power spectral density becomes

\[
S_n^{\text{coh}}(\omega) = \int_{-\infty}^{\infty} \langle \delta I_n^{\text{out}}(t + \tau) \delta I_n^{\text{out}}(t) \rangle e^{-i\omega \tau} d\tau \\
= \int_{-\infty}^{\infty} \left[ \alpha_n \delta a_n^\dagger(t + \tau) + \alpha_n^* \delta a_n(t + \tau) \right] \\
\times \left[ \alpha_n \delta a_n^\dagger(t) + \alpha_n^* \delta a_n(t) \right] e^{-i\omega \tau} d\tau \\
= \int_{-\infty}^{\infty} \left\{ \alpha_n^2 \langle \delta a_n^\dagger(t + \tau) \delta a_n^\dagger(t) \rangle \\
+ \alpha_n \alpha_n^* \langle \delta a_n^\dagger(t + \tau) \delta a_n(t) \rangle \\
+ \alpha_n^* \alpha_n \langle \delta a_n(t + \tau) \delta a_n^\dagger(t) \rangle \\
+ (\alpha_n^*)^2 \langle \delta a_n(t + \tau) \delta a_n(t) \rangle \right\} e^{-i\omega \tau} d\tau. \tag{3.3}
\]

Now, since the system is in a coherent state for this calculation, \( \langle \delta a_n^\dagger = 0 \), and \( \langle \delta a_n \rangle = 0 \). This simplifies Eq. (3.3) to

\[
S_n^{\text{coh}}(\omega) = |\alpha_n|^2 \int_{-\infty}^{\infty} \langle \delta a_n(t + \tau) \delta a_n^\dagger(t) \rangle e^{-i\omega \tau} d\tau. \tag{3.4}
\]

The commutation relations for the field operators, \([a_n(t), a_n^\dagger(t')] = \delta(t - t')\), lead directly to those for the field fluctuation operators, which are therefore given by

\[
[\delta a_n(t), \delta a_n^\dagger(t')] = \delta(t - t'), \tag{3.5}
\]

and similarly for the input operators. Using these commutation relations, the power spectral density for a coherent state is then simply

\[
S_n^{\text{coh}}(\omega) = |\alpha_n|^2. \tag{3.6}
\]

For the output fluctuations, the superscript "out" should be included on \( \alpha_n \). One
can also interpret this result as proof that the variance is equal to the mean for a coherent state (Poissonian statistical distribution).
APPENDIX K

DIFFUSED WAIST
This very brief appendix is devoted to deriving the thermal waist. That is, this is the waist of the level 2 population, modified to account for diffusion during the vibrational decay time. The creation of molecular excitations due to the Raman process can be described with a Gaussian radial distribution with a standard deviation given by

$$\sigma_{\text{int}} = \frac{\omega_{0,\text{int}}}{2},$$

(K.1)

After a time $\tau_{\text{vib}}$, the standard deviation of this distribution is assumed to increase due to random walk of the molecules. The new standard deviation is then given by

$$\sigma_{\text{th}}^2 = \sigma_{\text{int}}^2 + 2D\tau_{\text{vib}},$$

(K.2)

where $D$ is the molecular diffusion constant. The thermal waist, which accounts for this diffusion is then given by

$$\omega_{0,\text{th}}^2 = \omega_{0,\text{int}}^2 + 8D\tau_{\text{vib}},$$

(K.3)

Eq. (4.4) follows directly from this result.
APPENDIX L

TEMPERATURE DISTRIBUTION
This appendix is devoted to deriving the steady state temperature distribution inside the Raman laser cavity. With the knowledge of the heat deposition profile, the diffusion equation is used to determine how the temperature varies as a function of radial distance from the optical axis.

In steady state, the temperature distribution can be determined by solving the following Poisson equation [160]

$$\nabla^2 T(r) = -\frac{Q(r)}{K_{th}}, \quad (L.1)$$

where $\nabla^2$ is the Laplacian operator, $T(r)$ is the temperature profile that results from the radially dependent heat deposition profile $Q(r)$ and where $K_{th}$ is the thermal conductivity of the gas. In cylindrical coordinates, the Laplacian operator is given by

$$\nabla^2 = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2}. \quad (L.2)$$

With this, Eq. (L.1) can be written

$$\frac{d}{dr} \left( r \frac{dT(r)}{dr} \right) = -\frac{rQ(r)}{K_{th}}, \quad (L.3)$$

where the fact has been used that the heat deposition profile is assumed to be independent of both $\theta$ and $z$. Integrating both sides of this equation from zero to $r$ with $Q(r)$ given by Eq. (4.5) gives

$$\frac{dT(r)}{dr} = -\frac{1}{r} \int_0^r \frac{r'Q_0}{K_{th}} \exp \left( -\frac{2(r')^2}{\omega_{0,th}^2} \right) dr'$$

$$= \frac{Q_0 \omega_{0,th}^2}{4rK_{th}} \left[ \exp \left( -\frac{2r^2}{\omega_{0,th}^2} \right) - 1 \right]. \quad (L.4)$$
This equation can be integrated again, this time with indefinite limits to give

\[ T(r) = \frac{Q_0 \omega_{0,th}^2}{8K_{th}} \left[ \text{Ei} \left( -\frac{2r^2}{\omega_{0,th}^2} \right) - 2 \ln r \right] + C, \quad (L.5) \]

where \( \text{Ei}(x) \) is the exponential integral function and \( C \) is a constant of integration. The value of this constant can be determined by requiring \( T(r = r_c) = T_0 \). That is, one must assume that there exists a surface at the radius \( r_c \) is held at the constant temperature \( T_0 \). With this, \( C \) can be evaluated and one can obtain the following expression for the temperature within the cavity relative to the constant temperature \( T_0 \)

\[ \Delta T(r) = T(r) - T_0 = \frac{Q_0 \omega_{0,th}^2}{8K_{th}} \left[ \ln \frac{r^2}{r_c^2} + \text{Ei} \left( -\frac{2r^2}{\omega_{0,th}^2} \right) - \text{Ei} \left( -\frac{2r_c^2}{\omega_{0,th}^2} \right) \right]. \quad (L.6) \]

Assuming ideal gas conditions, the resulting refractive index profile is given by

\[ \Delta n(r) = -(n_0 - 1) \frac{\Delta T(r)}{T_0}, \quad (L.7) \]

where \( n_0 \) is the refractive index of the gas in the absence of optical power. The refractive index profile resulting from the heat deposition is therefore not Gaussian.
APPENDIX M

SERVO CIRCUIT
Figure 68. Servo circuit for fast feedback to the diode laser injection current.
Figure 68 shows the circuit diagram for the fast servo path that feeds back to the diode laser injection current. The first stage is just a fast buffer. The second stage is an integrator with a zero. The third stage is a phase lead compensator [104, page 396]. Resistor R15 controls a dc voltage offset to compensate for any offset present on the error signal from the mixer. One can put resistors on either side of R15 to decrease the dc offset range. In later versions, resistor R4 was eliminated. Capacitors C1 and C2 are removable so that the two corner frequencies can be adjusted. It is also sometimes possible to squeeze out a bit more phase by dropping R7 a little, so this resistor is also removable. Resistor R13 is used to trim the overall gain. A fixed resistor can be placed in series with R13 to to obtain a different gain range, but problems are encountered when trying to make R13 large.
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