



Quantum fluctuations in stimulated Raman scattering
by Rand Curtis Swanson

A thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in
Physics

Montana State University

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Abstract:

Results from a study of quantum noise and its macroscopic manifestations in stimulated Raman scattering are presented. Shot to shot fluctuations in quantum noise initiated Stokes spectra are considered. The Stokes spectra were recorded experimentally using a Fabry-Perot interferometer. For comparison, numerically generated Stokes spectra were produced using the semi-classical model of stimulated Raman scattering with a numerically generated "vacuum" input field. The amount of structure on each spectrum was quantified by calculating the spectrum's standard deviation from its mean frequency. It was found that ensembles with the noisy spectra removed had narrower distributions of mean frequencies than ensembles that included noisy spectra.

The effects of these shot to shot spectral fluctuations on soliton formation in stimulated Raman scattering are presented next. Experimentally, the solitons were formed by using a Pockels cell to put a TC phase shift on the input Stokes pulse to a Raman amplifier. Theoretical solitons were produced by numerically placing a π phase shift in a Stokes field that was amplified using the semi-classical model of stimulated Raman scattering. Experimentally, noisy input spectra were discarded to insure the TC phase shift was the dominant phase structure. Good agreement was found with the numerical model when the noisy spectra were removed from the ensemble. These results indicate that soliton decay is directly related to quantum noise induced spectral fluctuations.

Results from studies on amplifier added noise are presented. The amount of noise added to a signal during amplification was studied by placing an amplifier in each leg of an interferometer. The noise added by the amplifier was deduced from the degradation of the output fringe pattern from the interferometer. This was done experimentally using Raman amplifiers and numerically using ideal, single mode amplifiers and Raman amplifiers. The results indicate that the amount of noise added by the Raman amplifiers corresponds to 1 photon per mode input to a noiseless amplifier. This is the lowest amount of noise allowed by quantum mechanics.

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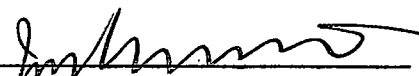
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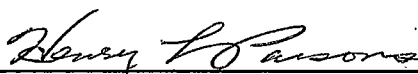
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ABSTRACT

Results from a study of quantum noise and its macroscopic manifestations in stimulated Raman scattering are presented. Shot to shot fluctuations in quantum noise initiated Stokes spectra are considered. The Stokes spectra were recorded experimentally using a Fabry-Perot interferometer. For comparison, numerically generated Stokes spectra were produced using the semi-classical model of stimulated Raman scattering with a numerically generated "vacuum" input field. The amount of structure on each spectrum was quantified by calculating the spectrum's standard deviation from its mean frequency. It was found that ensembles with the noisy spectra removed had narrower distributions of mean frequencies than ensembles that included noisy spectra.

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CHAPTER 1

INTRODUCTION

Spontaneous emission is an every day phenomenon that occurs, for example, whenever a fluorescent light or neon sign is turned on. In addition to such obvious occurrences of spontaneous emission in the physical world, it also plays more subtle roles such as placing an absolute limit on the fidelity of amplifiers. Since spontaneous emission is so ubiquitous in nature and because of its role in useful devices such as amplifiers, obtaining an understanding of its characteristics is important from a practical perspective. From a fundamental point of view, spontaneous emission is also an interesting topic to pursue because it provides an accessible testing ground for quantum optics.

In this thesis, I report on three experimental and theoretical studies in Raman scattering. The common denominator of all three studies is that the macroscopic results are critically dependent on the characteristics of microscopic spontaneous emission in Raman scattering. Consequently, the experiments I discuss provide a "microscope" to study spontaneous emission. Additionally, the results from these experiments are modeled with the quantum theory of Raman scattering, thus testing the validity of Raman scattering theory.¹⁻³

This thesis is organized into five chapters. In the first Chapter I present a brief

introduction to Raman scattering, and then give a short description of the three experiments and explain what we hoped to learn from them and why. The second Chapter is devoted to the mathematical theory of Raman scattering.¹⁻⁴ The essentials of the theory are presented in Chapter 2 with further details provided in appendices. The third Chapter discusses quantum noise induced spectral fluctuations in Raman scattering.⁵⁻⁷ Results from this Chapter are then used to gain an understanding of soliton decay in Raman scattering, the topic of Chapter 4.^{8,9} In the fifth and final Chapter, the limitations that spontaneous emission places on the Raman amplifier are tested.^{10,11}

Raman Scattering

Raman scattering is an inelastic photon scattering process where an input beam of light is red shifted as it scatters off a Raman active medium. This process is well known and has been utilized in a broad range of applications. In this thesis I study the quantum mechanical treatment of Raman scattering and explore the predictions of the theory. Thus I am primarily concerned with the fundamental understanding of the Raman scattering process itself rather than its applications.

The two photon Raman scattering process is shown in terms of an energy level diagram in Fig.1. The Raman active molecule consists of "three" levels, the ground state labeled $|1\rangle$, a level of opposite parity from the ground state labeled $|2\rangle$ (actually this can represent many high lying levels), and a level with the same parity as the ground state

labeled $|3\rangle$. In our experiment level $|3\rangle$ was the first vibrational level of H_2 . The molecule is pumped with a large laser field at angular frequency ω_p referred to as the "pump" that, for the experiments I describe, is far from any energy level of the molecule as indicated in Fig.1. The molecule is driven into level $|3\rangle$ by either of the two pathways shown in Fig.1. It can absorb a pump photon and then emit a lower energy photon at angular frequency ω_s referred to as a "Stokes" photon, or it may first emit the Stokes photon and then absorb the pump field energy. Both pathways contribute mathematically.

It should be noted that the energy levels do not have arbitrarily narrow widths. When, for example, collisions between molecules in the Raman medium take place, their energy levels are perturbed. This causes the energy levels to broaden, and consequently the linewidth of the Stokes field also broadens.

The Raman scattering process can occur spontaneously, or it can occur via stimulated emission and thus act as an amplifier of an input Stokes field. The origin of the spontaneous Stokes emission has two distinct interpretations that depend on the Stokes field operator ordering.^{7,12}

If the Stokes field operators are normally ordered to calculate the intensity, the Stokes field is interpreted as arising from the beating of the pump field with the quantum polarization fluctuations of the Raman medium. On the other hand, if the Stokes field operators are anti-normally ordered, the spontaneous Stokes field is interpreted as arising due to the stimulation from the vacuum field quantum noise. Identical results are obtained from either ordering, and both orderings rely on quantum fluctuations to initiate

the Stokes field. Consequently, convenience becomes the criterion used to choose the ordering scheme. Both orderings are used in this thesis.

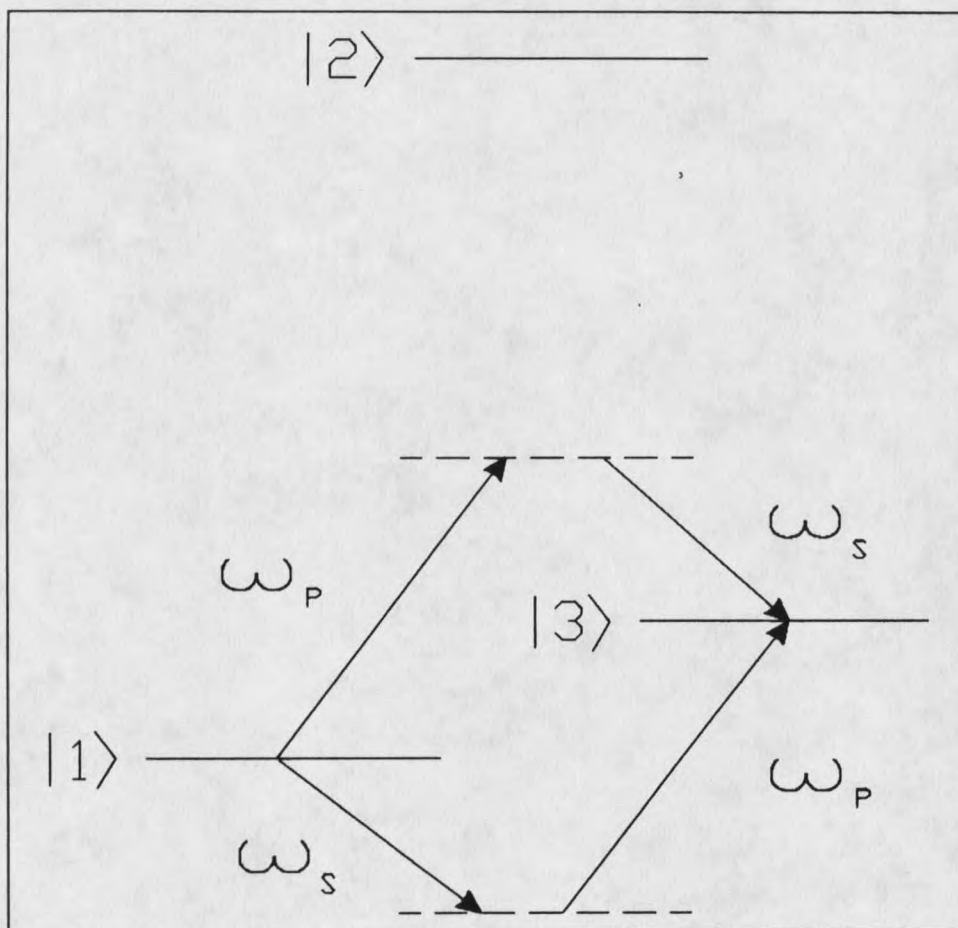


Figure 1 The energy level diagram of the Raman scattering process.

The Raman medium for our experiments was hydrogen gas, typically at 10 atm. pressure. The Raman medium was pumped by a pulsed, "single mode" laser, meaning that its linewidth was determined only by the Fourier transform of its temporal duration. Or, in other words, there were no phase shifts on the pump laser pulse. The Stokes field

was initiated by spontaneous Raman scattering. In some experiments, the spontaneously generated Stokes field was input to a second Raman cell to observe Raman amplification.

Spectral Fluctuations in Raman Scattering

Spectral fluctuations in Raman scattering are discussed in Chapter 3 of this thesis. Under the proper circumstances, the spontaneously initiated Stokes field can show dramatic shot to shot spectral fluctuations even though the experimental operating conditions are identical. Individual spectra may exhibit multiple spectral peaks that span a frequency range broad compared to the theoretically predicted gain narrowed linewidth, or on some shots single peaks may be much narrower than the gain narrowed linewidth. Since Raman scattering is often used for frequency conversion, it is important to understand these spectral fluctuations.

One can trace spectral fluctuations back to spontaneous Stokes emission. As mentioned earlier, spontaneous emission can be interpreted as a beating between the polarization fluctuations in the medium and the pump. Since the Raman medium undergoes random phase shifts, the spontaneous Stokes emission spectra contain a range of frequency components. This spontaneous Stokes emission is amplified as it propagates through the Raman medium, leading to a macroscopic output Stokes pulse, which has the frequency components of the originating spontaneous emission. (This picture is somewhat complicated by gain narrowing of the Stokes spectra that occurs during amplification.) Thus fluctuations in the output spectra are manifestations of the initiating quantum noise.

Chapter three will include experimental results of measured spectral fluctuations as well as results from theoretical modeling.⁷ Good agreement was found between theory and experiment. In our studies we found subtle differences between noisy Stokes spectra and single peak, near transform limited spectra. Near transform limited spectra tend to be located near line center, whereas the noisy spectra have a broad range of mean values. This turns out to be extremely important for explaining soliton decay in Raman scattering, the subject of Chapter 4.

Soliton Decay in Raman Scattering

In Raman scattering experiments, pump energy is diminished as it is converted into Stokes energy and vibrational energy in the Raman medium. However, if a π phase shift is inserted into the middle of the Stokes pulse, the process can be reversed with Stokes energy converted back to the pump. This reverse in gain leads to a short, high intensity peak in the pump temporal profile that, at large gain, has been shown to be a soliton solution to the Raman equations.¹³

These soliton pulses were first reported in Raman scattering in 1983.¹⁴ Subsequent efforts to produce solitons with electro-optical π phase shifts on the Stokes pulse had limited success.¹⁵ Although the solitons have been produced reliably, the amount of pump depletion varied from shot to shot, leading to solitons with amplitudes that vary from shot to shot.⁸ The origin of these soliton amplitude fluctuations was a mystery for some time.

Druhl *et. al.*¹⁶ predicted that solitons would decay if the Stokes pulse was off

resonance. Thus, shot to shot variations in the Stokes spectra could lead to the observed soliton decay.⁷⁻⁹ If the spontaneous spectral fluctuations are indeed the cause of soliton decay, the soliton amplitude distribution should be intimately related to the statistics associated with the spectral fluctuations. This is shown to be the case in Chapter 4.

Since soliton amplitude decay is due to shot to shot spectral fluctuations, one might wonder if this problem could be avoided by seeding the Raman amplifier with a cw source that is on resonance for the Raman transition. For this to work, however, the input signal must be large enough to dominate the noise introduced by the Raman amplifier. The issue of how large an input signal must be to dominate the noise introduced by a Raman amplifier is taken up in Chapter 5.

The Raman Amplifier

All light amplifiers necessarily add quantum noise.¹⁷⁻²⁰ This noise, in fact, is required by quantum mechanics. An understanding of how much noise an amplifier adds is important from a practical point of view in applications like trans-oceanic optical fiber communication lines where the signals have to be periodically amplified. It is also important to understand amplifier noise when considering fundamental questions such as the famous gedanken experiment with Schrodinger's cat, a radioactive atom, and an amplifier that triggers a lethal device.¹⁸

One can get a physical picture of the origin of quantum noise in an amplifier by considering an amplifier that consists of a medium of two level atoms with all the atoms

in their excited state. A light signal that has the frequency corresponding to the atoms' transition will amplify as it passes through the medium due to stimulated emission. However, we know that if an atom is placed in its excited state and left alone, it will decay to its ground state via spontaneous emission. The spontaneous emission, which can also amplify as it passes through the medium, is the noise.

Thus, from this simple model one expects that the output from this idealized amplifier will consist of a superposition of amplified signal and noise. The question that comes to mind is: How large does the signal have to be to dominate this noise?

To answer this question for Raman amplifiers we constructed an interferometer with Raman amplifiers in each leg. With no amplification, an input signal will form a set of fringes that depends on the geometry of the interferometer. This set of fringes is dependent on the correlation between the fields in each leg of the interferometer. When the amplifiers are turned on, noise is introduced to the field. This noise field is uncorrelated between the two amplifiers and leads to a degradation of the output fringe pattern. The relative amount of noise to signal can be inferred from the fringe degradation.

Experimental as well as theoretical results from this experiment are presented in Chapter 5. The exciting bottom line of the experiment is: The Raman amplifier emits the lowest noise allowed by quantum mechanics, which corresponds to 1 photon of noise per mode at the input of a noiseless amplifier.¹¹ Consequently, the noise can be dominated with signals having only a few photons per mode.

CHAPTER 2

THE RAMAN EQUATIONS

In Raman scattering an intense beam of light (referred to as the "pump") interacts with a Raman active medium, resulting in the excitation of the medium coincident with the emission of red-shifted light referred to as "Stokes" radiation. The Stokes radiation can be initiated spontaneously and can be amplified via stimulated emission.

The first mathematical models of Raman scattering were developed in the late twenties and early thirties shortly after the discovery of Raman scattering in 1928.^{21,22} The theory of Raman scattering has, as one would expect, evolved considerably in the more than sixty years since its discovery.

This Chapter will be devoted to a discussion of the mathematical modeling of Raman scattering. In particular, I will consider the semi-classical c-number model⁴ (i.e. there are no quantum mechanical operators used) as well as the fully quantum mechanical operator model^{1-3,23} (where quantum mechanical operators are used extensively). The semi-classical model is used in Chapters 3 and 4 where I consider spectral fluctuations and soliton decay. The fully quantum mechanical model is employed in Chapter 5, where Raman amplification is considered.

The Semi-Classical Model of Raman Scattering

The semi-classical model of Raman scattering mathematically couples the Raman active medium, the pump field, and the Stokes field. In this theory, the Raman active medium is treated quantum mechanically while the pump and Stokes fields are treated classically via Maxwell's equations, thus the label "semi-classical." The derivation of the semi-classical model is rather long and will not be presented here but can be found in the literature.⁴

If one considers a pencil shaped region of Raman active medium irradiated by a laser pulse, the slowly varying, differential equations governing Raman scattering are given by:

$$\frac{\partial E_s(z, \tau)}{\partial z} = -i\kappa_2 E_p(z, \tau) Q^*(z, \tau) \quad (2.1)$$

$$\frac{\partial E_p(z, \tau)}{\partial z} = -i(\omega_p/\omega_s)\kappa_2 E_s(z, \tau) Q(z, \tau) \quad (2.2)$$

$$\frac{\partial Q^*(z, \tau)}{\partial \tau} = -\Gamma Q^*(z, \tau) + i\kappa_1 E_p^*(z, \tau) E_s(z, \tau) \quad (2.3)$$

where $E_s(z, \tau)$ is the Stokes field at position z along the pencil shaped Raman medium at

retarded time τ , $E_p(z, \tau)$ is the pump field, and $Q(z, \tau)$ is the coherent superposition between the initial and final state of the Raman transition in the Raman active medium referred to as the "polarization." The coupling constants κ_1 and κ_2 depend primarily on the strength of the atomic transitions of the Raman active medium, ω_s and ω_p are the Stokes and pump frequencies respectively, and Γ is the dephasing rate of the Raman medium that indicates how fast the polarization of the Raman medium decays due to collisions between the molecules.

Due to the complex coupling between Eqs.(2.1-2.3), the solutions to these equations for a given set of input fields are usually found via numerical integration and good agreement with experiment has been found.²⁴

However, these equations have one serious shortfall in that they do not account for spontaneous Raman scattering. One can see in Eqs.(2.1-2.3) that if the initial Stokes field is set to zero and there is no initial polarization, then the output Stokes field remains at zero. These equations thus predict that when a Raman active medium in its ground state is pumped by a laser with no input field at the Stokes frequency, no Stokes radiation will be generated. Nature strongly disagrees with this result.

To account for spontaneous emission a "vacuum" field can be inserted artificially as the input Stokes field to the Raman active medium.⁷ One can justify the inclusion of this field in the following manner. If the Stokes field is treated quantum mechanically, it can be expanded mathematically into a set of orthonormal modes.^{25,26} The energy associated with each mode is proportional to the average number of quanta (photons) in that mode plus one half. The extra one half quanta corresponds to the zero point energy of the field

and is not detectable by photodetectors. Although not detectable, this extra one half of quanta can be thought to be the "stimulating" field for spontaneous scattering and is referred to as the "vacuum" field. Consequently, if one includes the "vacuum" field in the calculations using the semi-classical model, spontaneous Raman scattering is included in the theoretical predictions. As long as the unobservable "vacuum" field is either subtracted off the amplified field or is insignificant in comparison to the "real" field, the results should model Stokes amplified spontaneous emission.

As is well known, however, an infinite number of modes are required to model the full spectrum of the electromagnetic field. Thus, it is not obvious how to include the "vacuum" field as the initiating field for the semi-classical model.

Fortunately, the difficulty with the infinite number of modes necessary to model the electromagnetic field is easily avoided in many applications due to a finite frequency bandwidth and the finite temporal durations of the fields of interest. The argument proceeds as follows: In Raman scattering only a narrow bandwidth of frequencies are amplified. Therefore "vacuum field" frequency components that are far from the resonant (central) bandwidth experience no amplification. Since these components are not detectable (so they should be subtracted off at the end of the calculation anyway) and since they are not amplified, they can be dropped from the calculation without affecting the result. Thus only a narrow bandwidth of "vacuum field" is included to account for spontaneous emission. The spacing of the modes within the linewidth of interest need only be close enough that the system cannot resolve one mode from the next over the duration of the pulse.

For the purposes of modeling this vacuum field, all that is left to do is explain how the magnitude of the "vacuum field" was calculated.

Spontaneous Raman scattering (before amplification) is emitted into a Lorentzian lineshape with a halfwidth of Γ .^{2,4,27} With this information, we assumed that the effective energy in the "vacuum field" was:

$$\mathcal{E}_{vac} = (\hbar\omega_s/2) \sum_n \frac{\Gamma^2}{\Gamma^2 + (\omega_n - \omega_s)^2} \quad . \quad 2.4$$

Thus the "vacuum" energy is weighted by a Lorentzian lineshape. Converting this to an integral and using the density of states in one dimension allows the "vacuum" energy to be calculated for an arbitrary quantization length. Furthermore, dividing this quantity by the characteristic time associated with the quantization length yields the effective power under the Raman linewidth due to the vacuum field:⁷

$$E_{vac}c/L = P_{eff} = \hbar\omega_s\Gamma/2 \quad 2.5$$

where L is the quantization length. This result is consistent with the phenomenological photon rate equation.²

This "vacuum" field was numerically generated by summing together 136 modes (frequencies) with random phases that spanned roughly twice the Raman linewidth. The mode spacing was chosen to be small enough that individual modes could not be resolved within the duration of the pump pulse.

This artificially generated "vacuum" field was the input Stokes field that was

amplified by Eqs.(2.1-2.3) to simulate experiment. Since the output field was approximately 14 orders of magnitude larger than the initiating "vacuum" field, it could safely be regarded as negligible and was not subtracted from the output.

Using numerically generated "vacuum" fields allowed us to generate ensembles of macroscopic fields that could be compared with experiment. Since this method of modeling Raman scattering produces single shot results, one can very quickly observe shot to shot variations. This theoretical method of generating Stokes radiation was employed in the analysis of spectral fluctuations (Chapter 3) and soliton decay (Chapter 4).

The Quantum Model of Stimulated Raman Scattering

Like the semi-classical model of Raman scattering, the fully quantum mechanical model has been developed elsewhere and will not be repeated here.^{1-3,23} This theory predicts that the Stokes field will be initiated by spontaneous emission, and consequently the problem of Raman scattering can be treated more rigorously than with the semi-classical model. Additionally, since the results from the fully quantum model are obtained by taking expectation values of operators, ensembles of numerically generated shots do not have to be produced to compare with experiment. The fully quantum model, however, does not take pump depletion into account. Consequently, this theory does not model Raman scattering well once the Stokes field energy gets large enough that pump depletion becomes important.

For the most part, the derivation of the fully quantum model of Raman scattering is straightforward but time consuming. The operator equations can be derived from the Hamiltonian of the system and Maxwell's equations, with the coupled equations closely resembling Eqs.(2.1,2.3). There are, however, some interesting mathematical gymnastics performed in the derivation of the Raman medium polarization that I will discuss in Appendix A. The differential equations can be solved with Laplace transforms to find the operator expression for the Stokes field:²

$$\begin{aligned} \hat{E}_s^{(-)}(z, \tau) = & \hat{E}_s^{(-)}(0, \tau) + \int_0^z dz' A(z, z', \tau) \hat{Q}^\dagger(z', 0) \\ & + \int_0^\tau d\tau' B(z, \tau, \tau') \hat{E}_s^{(-)}(0, \tau') + \int_0^\tau d\tau' \int_0^z dz' C(z, z', \tau, \tau') \hat{F}^\dagger(z', \tau') \end{aligned} \quad 2.6$$

where $\hat{E}_s^{(-)}(z, \tau)$ is the slowly varying Stokes electric field operator at position z and

retarded time τ , $\hat{Q}^\dagger(z, 0)$ is the initial polarization operator for the Raman medium, and

$\hat{F}^\dagger(z', \tau')$ is a quantum Langevin operator needed to maintain operator consistency when

damping of the polarization is included phenomenologically (see Appendix A). The kernels (A, B , and C), which account for the gain, are not important for the present discussion and are given in Appendix B.

The various terms on the right hand side of Eq.(2.6) are interpreted as follows: The first term is the input Stokes field to the Raman medium. The third term is the amplified Stokes field. And the second and fourth terms do not involve the input Stokes field, so they represent the amplified spontaneous emission, or the noise added by the Raman amplifier.

To test this theory, the Stokes intensity must be calculated since that is what is measured experimentally. This may be done either by normal ordering or anti-normal ordering.⁷ The Stokes operator $\hat{E}_s^{(-)}(z, \tau)$ can be expanded in terms of creation operators that act on a set of orthonormal modes that span the field.^{25,26} If the intensity is calculated by normal ordering the Stokes field operators ($\langle \hat{E}_s^{(-)}(z, \tau) \hat{E}_s^{(+)}(z, \tau) \rangle$), the destruction operators are always to the right of the creation operators. Therefore, modes with no photons in them do not contribute, as we would like. In anti-normal ordering the creation operators are to the right and all modes contribute. This additional contribution must be subtracted off to compare with experiment. To avoid this complication, the calculations we present here that use the operator formalism use normal ordering.

When the Stokes field operators are normally ordered, the following, fairly long expression is found:⁷

$$\begin{aligned}
\langle \hat{E}_s^{(-)}(z, \tau) \hat{E}_s^{(+)}(z, \tau) \rangle &= \langle \hat{E}_s^{(-)}(0, \tau) \hat{E}_s^{(+)}(0, \tau) \rangle \\
&+ \int_0^z dz' \int_0^z dz'' \langle \hat{Q}^\dagger(z', 0) \hat{Q}(z'', 0) \rangle A(z, z', \tau) A^*(z, z'', 0) \\
&+ \int_0^\tau d\tau' \int_0^\tau d\tau'' \langle \hat{E}_s^{(-)}(0, \tau') \hat{E}_s^{(+)}(0, \tau'') \rangle B(z, \tau, \tau') B^*(z, \tau, \tau'') \\
&+ \int_0^\tau d\tau' \langle \hat{E}_s^{(-)}(0, \tau') \hat{E}_s^{(+)}(0, \tau) \rangle B(z, \tau, \tau') + \int_0^\tau d\tau'' \langle \hat{E}_s^{(-)}(0, \tau) \hat{E}_s^{(+)}(0, \tau'') \rangle B^*(z, \tau, \tau'') \\
&+ \int_0^z dz' \int_0^z dz'' \int_0^\tau d\tau' \int_0^\tau d\tau'' \langle \hat{F}^\dagger(z', \tau') \hat{F}(z'', \tau'') \rangle C(z, z', \tau, \tau') C^*(z, z'', \tau, \tau'')
\end{aligned} \tag{2.7}$$

where I have neglected the plethora of cross terms that contain terms such as

$$\langle \hat{Q}^\dagger(z', 0) \hat{E}_s^{(+)}(0, \tau'') \rangle \quad \text{since the polarization of the Raman medium, the input Stokes field,}$$

and the collisional Langevin operator have no correlation with each other and do not contribute.

When the input Stokes field is the vacuum field, this normally ordered expression, Eq.(2.7), simplifies considerably because all but the second and last terms on the right hand side vanish. The remaining terms account for the amplified spontaneous emission, which depends on the polarization operator, and the Langevin operator that accounts for collisional effects in the polarization (see Appendix A). Thus normally ordering the Stokes field operators leads to the interpretation that spontaneous Raman scattering is dependent on the polarization fluctuations in the Raman medium.

Equation (2.6) predicts that the output field from a Raman cell will be a superposition

of the field initiated by spontaneous emission and the field due to stimulated emission. Despite the fact that this theory was presented ten years ago, there have not been any experiments performed that tested to see if this theory predicts the correct superposition of the spontaneously generated field and the stimulated field until this research.^{10,11} The results of the experiment and the comparison with the theory are presented in Chapter 5.

CHAPTER 3

SPECTRAL FLUCTUATIONS IN RAMAN SCATTERING

Spectral fluctuations in Raman scattering were first observed in 1988⁵ in conjunction with studies of soliton decay in Raman scattering. During these studies, it was found that the Stokes spectra varied dramatically from shot to shot with some shots having spectra much narrower than the gain narrowed linewidth. Since the Stokes field arose from spontaneous emission, it was evident that the spectral fluctuations represented a macroscopic manifestation of the quantum fluctuations that initiated the field. As such, these quantum fluctuations represent the fundamental limit associated with the noise found on any light amplifier.

The spectral fluctuations also proved to be of interest in the study of soliton decay in Raman scattering. As will be discussed in the following Chapter, the shot to shot spectral fluctuations are directly related to observed soliton decay.⁸ Additionally, the quantum noise that leads to the spectral fluctuations is also responsible for spontaneous solitons in Raman scattering that have been predicted^{28,29} and observed.³⁰

In this Chapter I will first discuss some background, then explain why spectral fluctuations are observed. After this, the experiment used to observe spectral fluctuations is presented followed by the theoretical modeling.

Spectral Fluctuations

Spontaneous Raman scattering can be viewed as arising from the beating between the pump laser field and the medium's polarization. I will assume that the pump field is a single mode, pulsed field, as used in our experiments. The Raman medium consists of a gas of molecules (H_2 for our experiment) that undergo collisions with a characteristic time $1/\Gamma$. When molecules in the Raman medium experience collisions their polarization undergoes phase shifts. Consequently, the Stokes field that is produced by the beating of the polarization with the pump field has corresponding phase shifts.

There are several ways of considering the consequence of these phase shifts in the spectra of the Stokes field. From a mathematical point of view, one can note that the Stokes field becomes uncorrelated temporally with its value a few $1/\Gamma$ ago. Therefore its characteristic time is set by the collision time, and, performing a Fourier transform, one can see its power spectrum will be of the order Γ in width.

Another way is to note that a phase shift occurs as the result of the beating between more than one frequency. Thus, every time the field has a phase shift, its spectrum is necessarily broader than what it would have been if it had none because of the extra frequencies needed to produce the phase shift.

This picture is complicated somewhat due to gain narrowing. Gain narrowing results because those frequency components near line center experience more gain than those in the wings of the spectral profile. As a result of this, the spectral linewidth narrows. Conversely, the characteristic time between phase shifts in the Stokes pulse is lengthened.

The prediction for the gain narrowed linewidth in the steady-state, high gain regime is given by:^{2,27}

$$P_s(\omega) \propto \exp\left[-\frac{gz}{\Gamma^2} \omega^2\right] \quad 3.1$$

Here gz is the gain and ω is the Stokes frequency as measured from resonance. This result is valid only before pump depletion becomes important. However, it is expected that pump depletion does not have a significant effect on gain narrowing.⁵

Now, consider an experiment where the single mode pump laser is somewhat longer in duration than the characteristic time of the Stokes pulses (the average time between phase shifts). The phase shifts are initiated by random collisions. Due to the random nature of the initiating procedure it is possible that the Stokes pulse might have many phase shifts or none. Consequently, the output Stokes spectrum may have considerable structure or very little. From shot to shot, of course, the spectra will vary as long as the variations in the number of phase shifts is large compared to the average number of phase shifts in a pulse.

For example, if there were 10^6 phase shifts per Stokes pulse on average with fluctuations of the order of 10^3 , one would expect that the spectra would not show much variation from shot to shot since all the shots have around a million phase shifts. On the other hand, if there were on average 2 phase shifts per pulse, it would not be unusual to see shots with as many as 4 phase shifts or as few as none. There should be significant spectral variations seen in this situation.

Spectral Fluctuations Experiment

In this section I describe the experiment⁵⁻⁷ performed to observe spectral fluctuations in Raman scattering. Additionally, I will present some of the experimental results and introduce a characterization scheme to differentiate "noisy" spectra from near transform limited spectra. This, in turn, leads to further understanding of the Raman scattering process and provides results necessary to explain soliton decay.

The experimental apparatus used to study spectral fluctuations is diagramed in Fig. 2. A single mode, frequency doubled Nd:YAG laser at 532 nm was used to pump a Raman generator. The input pump pulse had a near gaussian temporal envelope with approximately 1 mJ of energy at the input of the Raman generator. The full width at half maximum of the pump pulse was 17.7 ns. To insure that data were taken only during periods of pump laser stability, the pump was monitored with a Fabry-Perot interferometer. The Raman generator consisted of a cell of hydrogen gas at 10 atm placed in a multipass cell.²⁴ The experiment was performed at room temperature. The pump laser pulse made 15 passes through the 1.5 m cell with a confocal parameter of 38 cm. The Stokes pulse was initiated by spontaneous emission and grew through pump depletion. Competing factors such as second Stokes and anti-Stokes were not important.²⁴

The output Stokes pulse was expanded and passed through a second Fabry-Perot interferometer. This interferometer had a free spectral range of 1 GHz with a finesse of approximately 70 and was temperature controlled to approximately a tenth of a degree centigrade for stability.

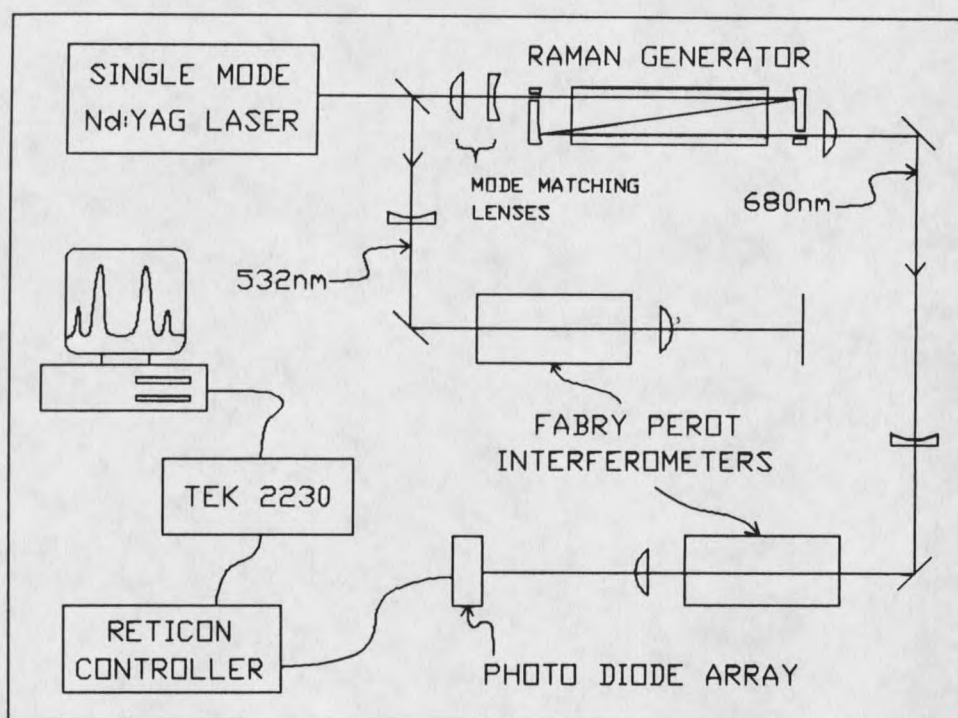


Figure 2 Experimental apparatus used to measure spectral fluctuations in Raman scattering.

To get maximum resolution, the Stokes beam was made slightly divergent such that we could image two rings onto a linear photodiode array, which measured a cross section of the rings. The linear photodiode array was interfaced to a computer for data analysis.

In Figs.3 and 4 are two typical single-shot spectra with the analytical gain narrowed linewidth (Eq.(3.1)) shown as a dashed curve for comparison. The spectrum in Fig.3 is clearly narrower than the gain narrowed linewidth, and in fact is near the Fourier transform limit of the Stokes pulse. Figure 4 is notably different with two distinctive peaks.

To distinguish between the clean spectra like the one shown in Fig.3 and the noisy

spectra such as that shown in Fig.4, we calculated the mean frequencies (f_{mean}) of the spectra and their standard deviations (σ) from their means with the following formulas:

$$f_{mean} = \frac{\int f I(f) df}{\int I(f) df} , \quad (3.2)$$

$$\sigma^2 = \frac{\int (f - f_{mean})^2 I(f) df}{\int I(f) df} , \quad (3.3)$$

where $I(f)$ is the measured intensity component at the frequency f .

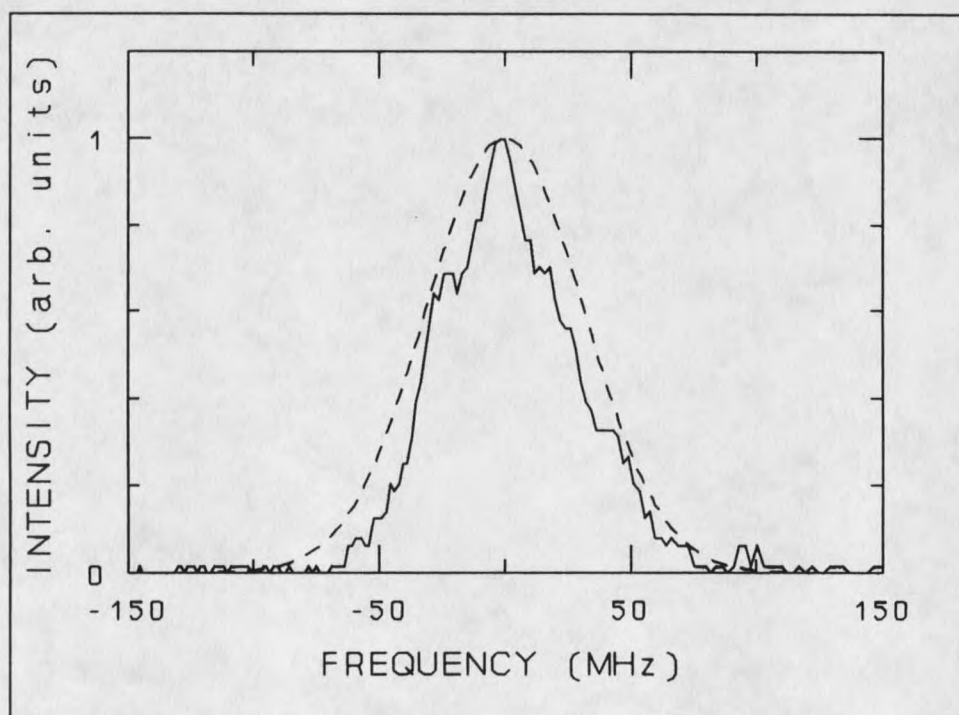


Figure 3 An experimentally measured, near transform limited Stokes spectrum.

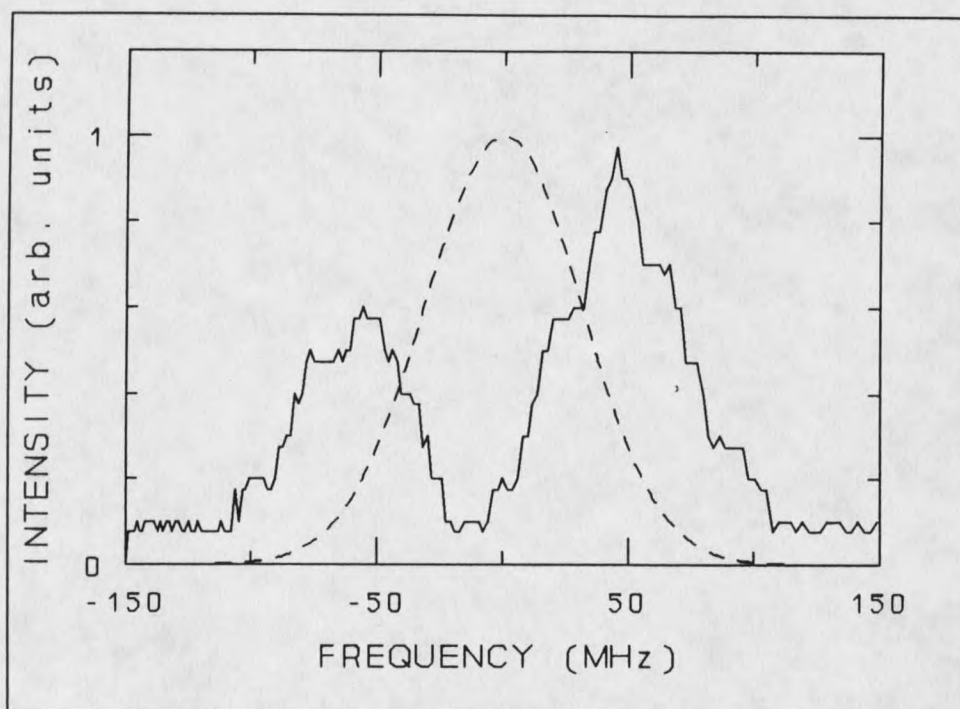


Figure 4 An experimentally measured noisy Stokes spectrum. The gain narrowed linewidth is shown for comparison as a dashed line.

Noisy spectra as in Fig.4 necessarily have larger frequency components far from their mean frequencies than clean spectra such as in Fig.3. Consequently, the clean spectra had much smaller standard deviations (σ 's) than the noisy spectra, yielding a quantitative measure of how "noisy" a single-shot spectrum was. For example, the spectrum in Fig.3 had a mean frequency of 0.52 MHz with a standard deviation of 26.5 MHz, while on the other hand the noisy spectrum in Fig.4 had a mean frequency of 2.3 MHz and a standard deviation of 73.9 MHz. Thus the standard deviation of a spectrum was an excellent indicator of how noisy an individual shot was.

Typically, data was taken several hundred shots at a time during which time the apparatus had to remain stable. Besides monitoring the pump with a Fabry-Perot

interferometer, we found another more sensitive method to discover small drifts in frequency. The mean frequencies of the first 100 shots recorded were used to generate a histogram. This histogram was constructed by dividing the frequency axis into 5 MHz bins and plotting the number of shots with mean frequencies that fell into each bin on the vertical axis. The same was done with the next 100 shots and so on. These histograms were then compared with one another. Any drift in frequency that occurred during data collection caused the histogram of the first 100 shots to be centered about a different frequency than a histogram of a group of 100 shots that occurred after the drift. Histograms compiled with shots that were taken during a drift were smeared out. Only those runs that did not show significant drifts in frequency were used.

While it is clear from Fig.3 that some shots can have a spectrum narrower than the gain narrowed linewidth, when an ensemble average of spectra is formed, the result is close to the gain narrowed linewidth. A large ensemble of experimental shots was obtained by combining several runs together. The data from the Fabry Perot interferometer yields the relative frequencies, but not the absolute frequencies. Therefore, resonance was chosen as the center of a gaussian fitted to the ensemble average of the spectra from each run. The experimental runs had between 300 and 600 individual shots. The data from three runs were then combined straightforwardly into one large ensemble. An ensemble of 1381 spectra was generated in this way.

Figure 5 shows the experimental ensemble average of 1381 spectra (solid curve) and the analytical gain narrowed linewidth (short dashed curve). The large dashed curve indicates the results from the numerical approach, which will be described later in this

Chapter. Note the excellent agreements in the width, although the wings of the experimental and numerical results are somewhat broader than the analytical result. The steady state approximation, which is made in deriving the analytical result for the gain narrowed linewidth, is valid for the condition $\Gamma\tau > gL$.² In our experiment, $1/\Gamma = 0.65$ ns, and the pump pulse had a full width at half maximum of about 17.7 ns. Thus as the pulse is developing with low gain, the steady state approximation is valid. However, at the output of the Stokes generator $gL = 30.8$, so in the latter stages of development the steady state approximation is not well justified. Therefore it is not surprising to find some deviation between the analytical steady state gain narrowed linewidth and the ensemble average of the Stokes spectra we measure experimentally.

Besides allowing a sensitive test of the shot to shot stability during the experiment, the mean frequencies of the single shot spectra provide a means of discovering the character of the shot to shot fluctuations. In Fig.6 the distribution of mean frequencies from the experimental ensemble is plotted. This distribution was constructed in the same manner as described for the 100 shot histograms above. The solid curve is a χ^2 gaussian fit to the mean frequency distribution, and the dashed curve is the gain narrowed linewidth plotted in relative units for comparison. Note that the distribution of mean frequencies is clearly narrower than the gain narrowed linewidth. This is easily understood. Since each spectrum has a width of the same order as the gain narrowed linewidth, the distribution of the means must be narrower than the gain narrowed linewidth.

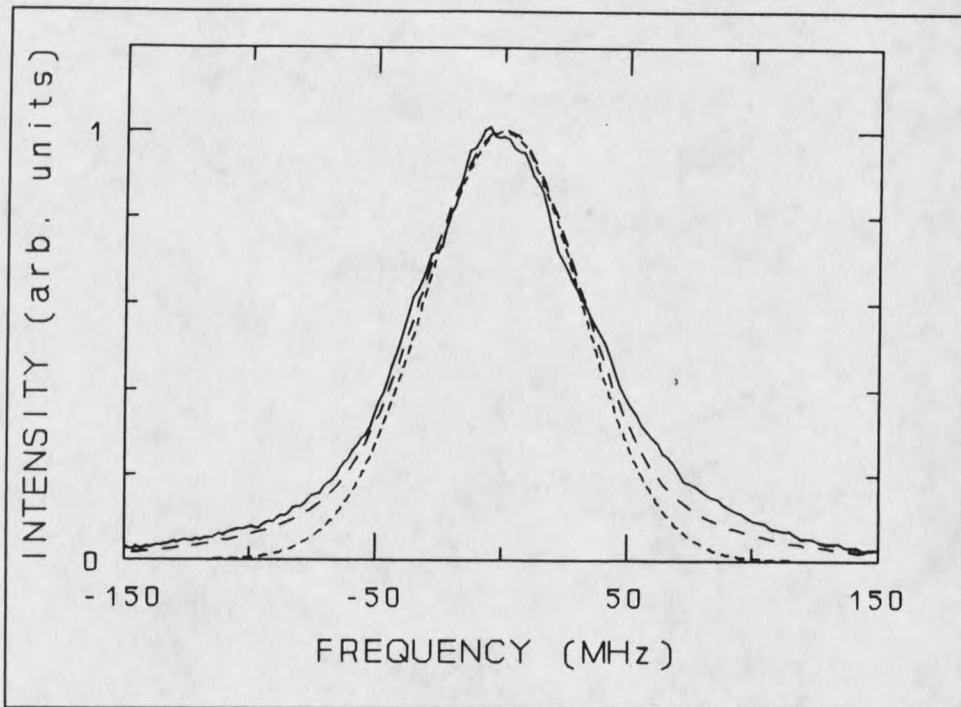


Figure 5 The Raman gain narrowed linewidth. The solid curve is the experimental result, the long dashed curve is the numerical result, and the short dashed curve is the analytical result.

To look for a difference between the characteristics of clean, near transform limited shots and the ensemble as a whole, we considered the distribution of mean frequencies of only those shots with a standard deviation less than 35 MHz and compared this with the distribution of the mean frequencies of the whole ensemble. These shots correspond to approximately 43% of the total ensemble. The comparison is shown in Fig.7. The + ticks indicate the distribution of the entire ensemble and * ticks indicate the distribution of the "clean" shots with $\sigma < 35$ MHz. Both distributions have been fitted with a gaussian. Surprisingly, this plot indicates that near transform limited shots are much more likely to be near resonance than are noisy shots.

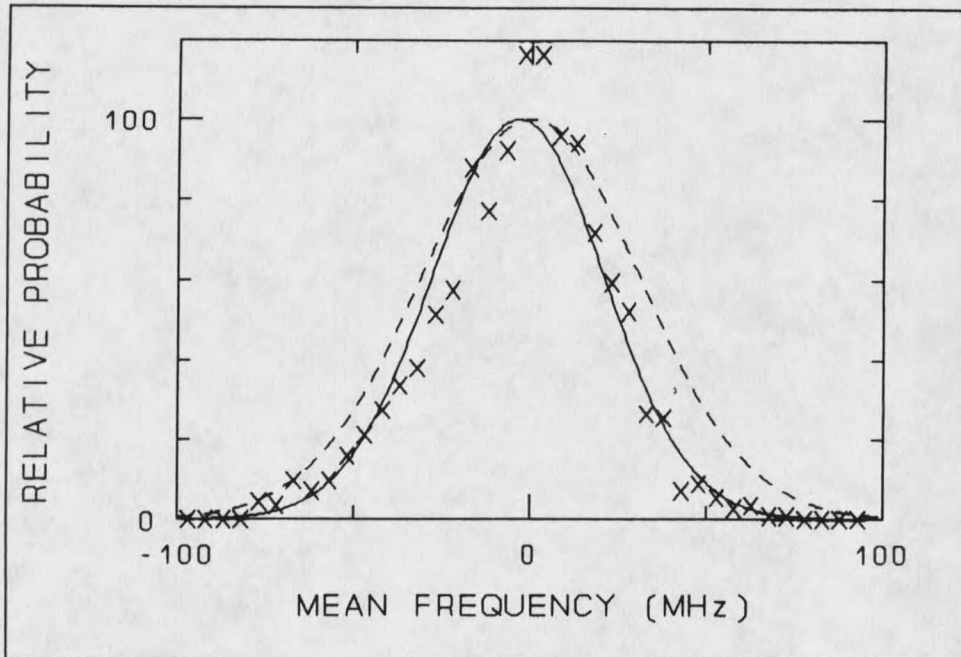


Figure 6 Measured distribution (X's) of the mean frequencies of Stokes spectra. The solid curve is a gaussian fit. The gain narrowed linewidth (dashed curve) is shown for comparison.

Numerical Modeling of Spectral Fluctuations

To determine if this narrowing of the distribution of mean frequencies was consistent with theory, we numerically modeled the semi-classical equations described in Chapter 2. Using the semi-classical approach with a numerically generated "vacuum" field corresponds to the anti-normally ordered approach. As discussed in Chapter 2, this approach has the advantage of producing single shot results that can be compared with experimental results rather than just the ensemble average. This approach also allows the inclusion of pump depletion.

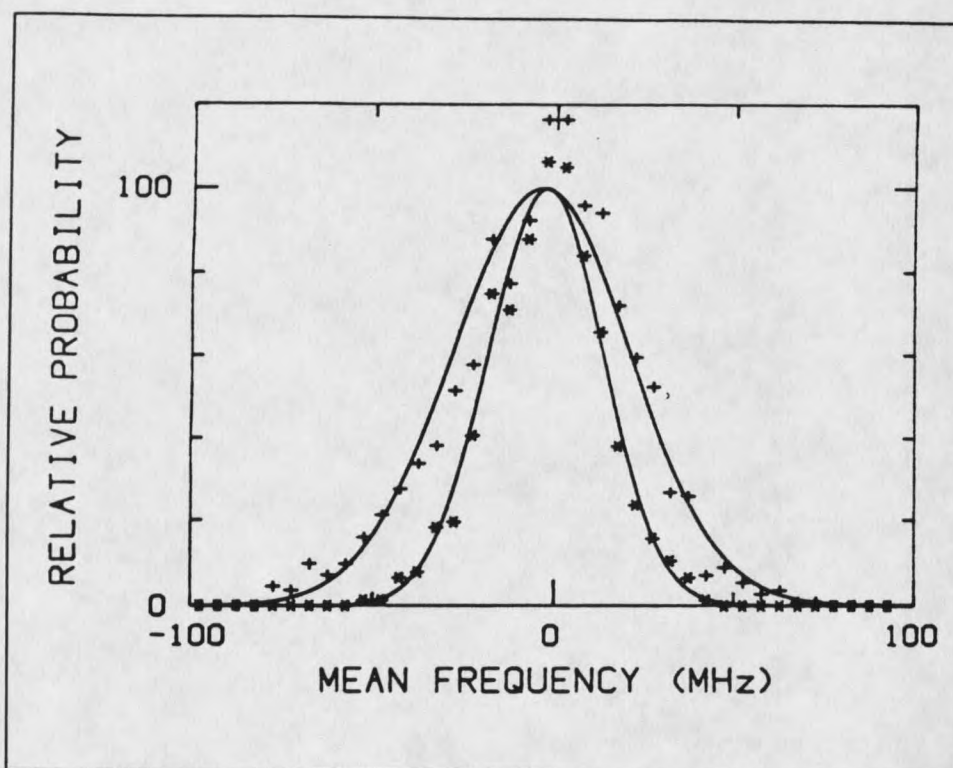


Figure 7 The + ticks indicate the distribution of all mean frequencies. The * ticks indicate the distribution of "clean" spectra means. Both distributions have been fitted with gaussians.

To compare the numerical approach with experiment, each numerically generated Stokes output pulse was Fourier transformed to obtain its spectrum. The mean frequency and standard deviation from its mean were then calculated as was done for the experimental data. In Figs.8 and 9 are two typical numerical Stokes spectra (solid curves) with the gain narrowed linewidth shown for reference (dashed curve).

These two shots were chosen to show that the numerically generated shots show structure similar to the experimentally observed spectra. As seen in the experimental spectrum in Fig.3, Fig.8 is narrower than the gain narrowed linewidth and is near the Fourier transform limit of the Stokes pulse. Similar to the experimental shot shown in

Fig.4, the numerically generated shot shown in Fig.9 is considerably broader with two distinctive peaks.

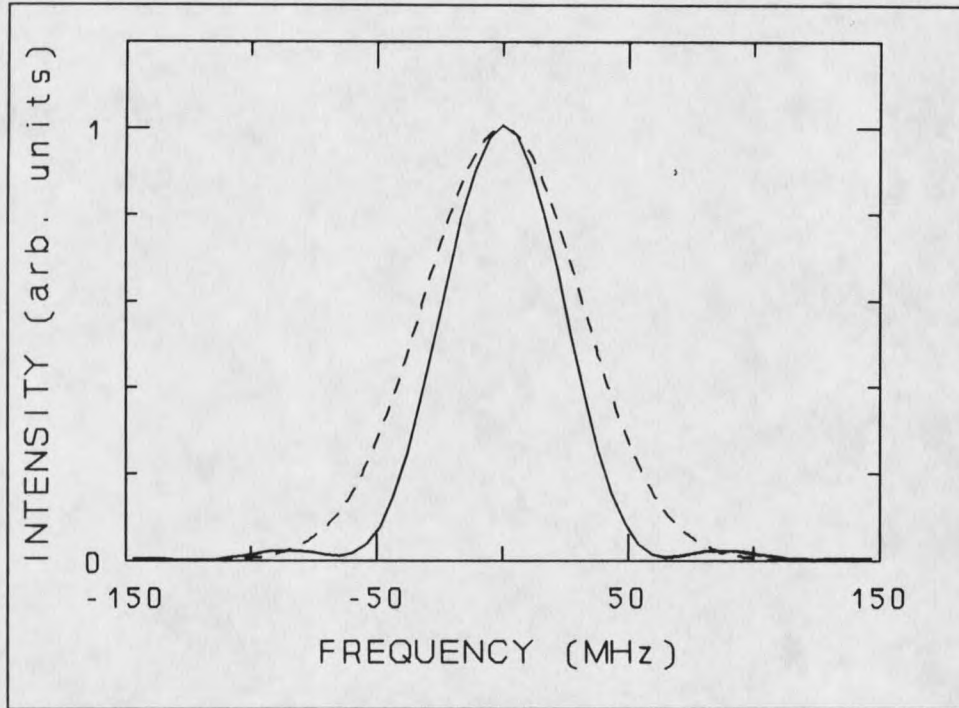


Figure 8 A numerically generated, near transform limited Stokes pulse.

As shown in Fig.7, the distribution of means for the experimental shots became narrower as we eliminated those shots from the ensemble which had a standard deviation greater than some maximum allowed standard deviation (referred to as the cutoff). The smaller the cutoff, the narrower the distribution of means became. In order to make a quantitative comparison between theory and experiment, we fit gaussians to the distribution of means of both the numerical and experimental ensembles for various cutoffs. The half width at half maximum (HWHM) of the gaussian fit was then used as a measure of the width of

the distribution of means. In Fig.10 we have plotted the HWHM versus cutoff for both the experimental ensemble (X's) and the numerical ensemble (solid curve). The dashed lines indicate the uncertainty in the gaussian fits of our numerical data. Similarly, the error bars indicate the uncertainty in the fits of the experimental ensembles.

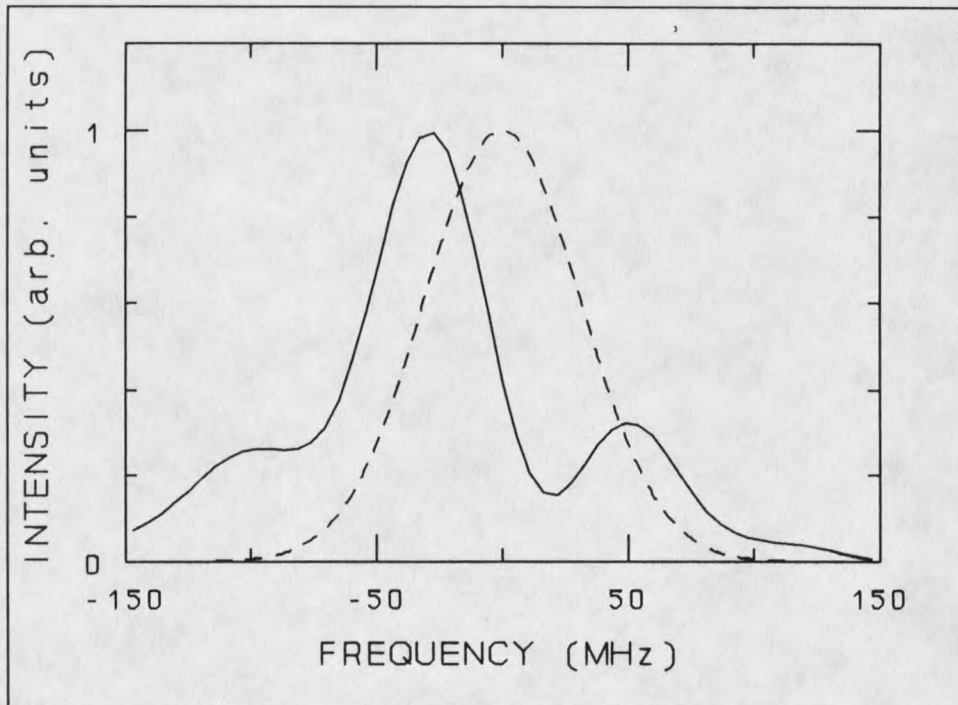


Figure 9 Numerically generated Stokes spectrum. Note that this spectrum is quite noisy.

This plot shows that as we make the cutoff value smaller, the distributions narrow more rapidly. Of course no spectrum can have a width less than the Fourier transform limit of the Stokes envelope (approximately 26.5 MHz), so for cutoff values less than this no spectra were left in the ensemble and the curve in Fig.10 ended.

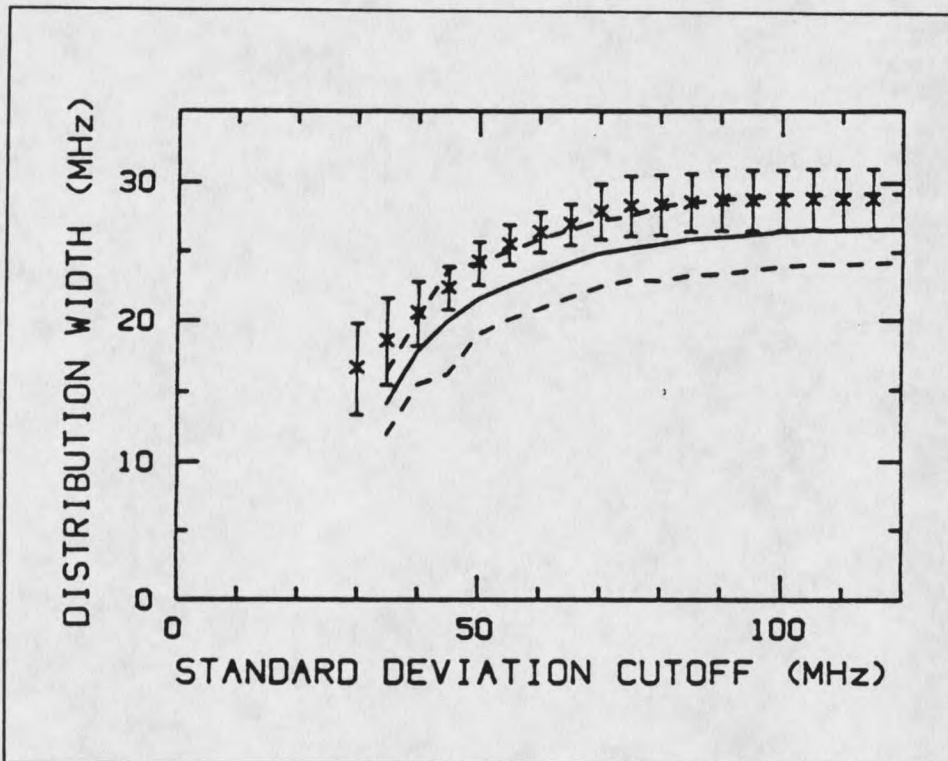


Figure 10 The experimental and numerical widths of the distributions versus cutoff for standard deviation.

There are a variety of possible reasons for the slight difference between the numerical and experimental results. All experimental spectra analyzed had noise subtracted off to prevent the noise from affecting the calculated mean frequencies and standard deviations. Small frequency components that could not be distinguished from noise were also subtracted off. The loss of these frequency components had little effect on the mean frequency of the pulse because on average the absence of these components is distributed equally above and below the "true" mean frequency. However, these missing components tended to make the experimental standard deviation too small because missing components from above *and* below the mean frequency would have

added to the standard deviation. Thus experimental shots were included in distributions of low cutoffs that would not have been if we could have resolved the small frequency components. The overall effect is that the experimental data are shifted slightly to the left in Fig.10.

Although the analysis ensured there was no major drift in pump frequency, a slow drift of the order of 10 MHz would have been unresolvable. Small shot to shot jitter in the pump frequency would also have been unresolvable. Such effects would cause the experimental distributions to widen, and therefore be shifted up in Fig.10. Despite these uncertainties, Fig.10 still shows quite good agreement between experiment and theory, both strongly indicating that near transform limited shots tend to have a narrower distribution than the ensemble as a whole.

Results and Interpretation

These results indicate that the ensemble average of single shot spectra is very nearly the gain narrowed linewidth. Because individual spectra can have widths and structure that differ considerably from the ensemble average, we interpret each shot as a realization of the quantum mechanical ensemble.

The distribution of mean frequencies is seen to be narrower for clean, near transform limited shots than for noisy shots. As one becomes more particular as to how clean the spectrum must be (that is, requires all shots to have smaller standard deviation from their mean frequencies), the distribution narrows more rapidly.

This can be understood qualitatively with the following argument. A developing Stokes pulse with its dominant frequency components near line center is unlikely to develop secondary peaks in its spectrum because all other frequency components are in spectral regions with less gain. On the other hand, a developing Stokes pulse with its dominant frequency components well off line center is likely to develop secondary peaks because small frequency components near resonance may experience enough gain to become significant. Thus one might well expect the noisy shots with large standard deviations from their mean values to have a broader distribution than clean shots. The more stringent the requirement as to how "clean" a shot has to be, that is, how small the largest allowed standard deviation is allowed to be, the narrower the distribution.

The mean frequency is typically more or less centrally located in each shot, while the gain narrowed linewidth is the ensemble average of all frequency components in the shots. Thus one might expect the gain narrowed linewidth to be the convolution of the distribution of mean frequencies with the typical width of the spectra. Using the numerical results to avoid the uncertainties associated with the experiment, we checked this by adding in quadrature the half width at half maximum of the typical Fourier transform limit of the Stokes pulse (approximately 26.5 MHz) with the HWHM of the distribution of the mean frequencies (26.7 MHz). This gave a result of 37.6 MHz compared to the measured HWHM of the ensemble average of the numerically generated shots of 38.4 MHz, which shows quite good agreement.

The results of these studies on spectral fluctuations are critical for an understanding of the decay of solitons in Raman scattering, the topic of the next Chapter of this thesis.

CHAPTER 4

SOLITON DECAY IN RAMAN SCATTERING

In this Chapter I will discuss the decay of "solitons" in stimulated Raman scattering. First I will discuss what a "soliton" in stimulated Raman scattering is and how one can be observed. Additionally the relationship between "soliton" decay and spectral fluctuations in Raman scattering will be considered. After that the experiment⁸ that was performed to observe the statistics associated with "soliton" decay is presented. And finally I will discuss the theoretical modeling of "soliton" decay and show that the observed decay is directly related to the quantum fluctuations that initiate the Stokes field in Raman scattering.

"Solitons" in Stimulated Raman Scattering

In stimulated Raman scattering pump energy is transferred to the Stokes field via stimulated emission. This process, of course, cannot go on indefinitely due to the finite energy in the pump laser pulse. Once the pump energy is diminished the pump is said to be depleted. The conversion of pump energy to Stokes energy occurs most efficiently in the region where the pump intensity is the highest. In Fig.11 is a temporal profile of the pump intensity before depletion (the higher curve) and after depletion (the lower

curve). The region in the center of the depleted pump pulse where the pump intensity has been diminished is called the depletion region. Note that the depletion region is temporally in the location where the input pump had the highest intensity.

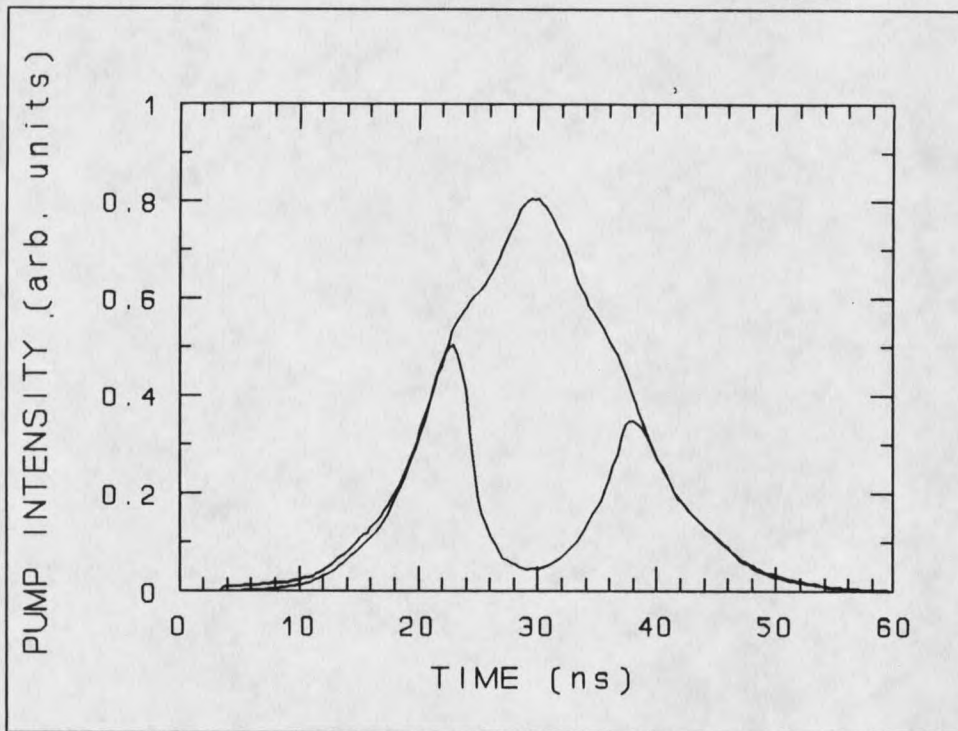


Figure 11 The temporal profile of the pump pulse before (upper curve) and after (lower curve) depletion.

One can think of a soliton as a particular solution to a set of mathematical equations (the Raman equations for example) that has a solitary wave shape.^{13,31,32} When a phase flip occurs in the Stokes field, a sharp peak in intensity can occur in the center of the depletion region of the pump. This sharp peak within the depletion region has been shown to evolve into a pulse that is very close to the exact soliton solution.³³ During the

evolution of this pulse into the near soliton solution, it narrows in duration until it is short compared to the Raman coherence time $1/\Gamma$.

Unfortunately, once the pulse has narrowed this much it is not resolvable with our detection equipment. Consequently, we can only observe the sharp peak in its nascent stages before it becomes a soliton. Thus I have put the word "soliton" in quotes above. From here on, however, I will drop the quotes with the understanding that the phenomenon of interest is not yet, strictly speaking, a soliton.

For the benefit of the skeptical reader, an experimentally measured soliton pulse is shown in Fig.12. Note that the intensity peak is clearly visible in the center of the depletion region. The input, undepleted pump pulse is included for reference.

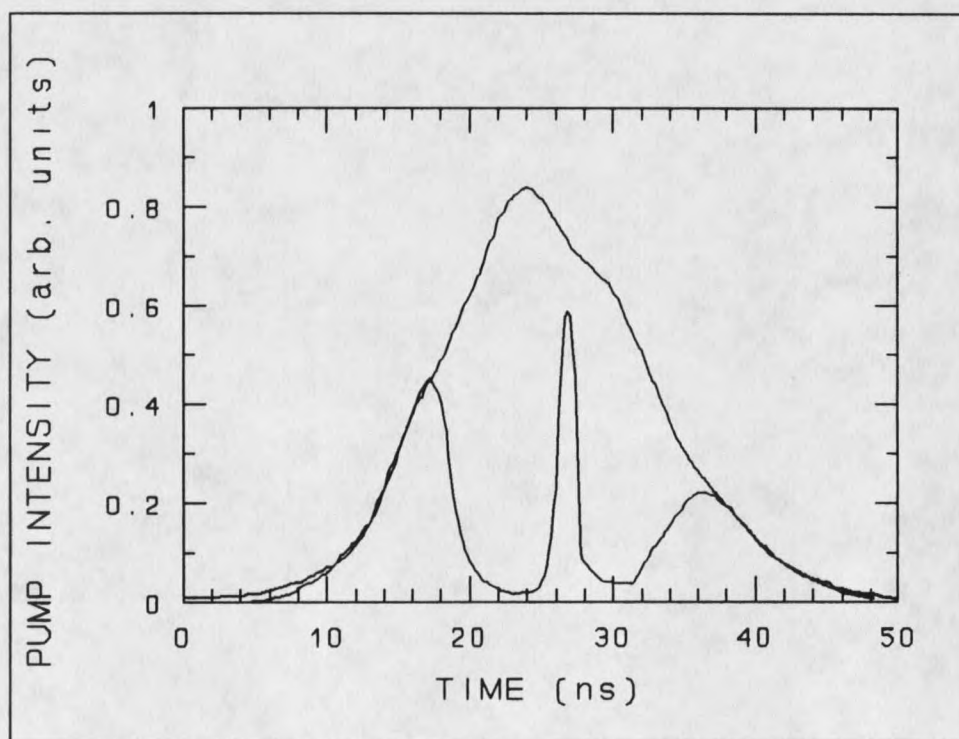


Figure 12 Experimentally recorded soliton in Raman scattering. This was one of the largest solitons recorded.

Mathematically, one finds that the repletion of the pump into the soliton is predicted when a π phase shift is inserted into the Stokes pulse.³⁴ This phase shift causes the Stokes field to experience loss and the pump field to achieve gain. Thus the Raman scattering process is reversed for a brief period. The exact mechanism of this process is rather complex since Raman scattering is described by three coupled, partial differential equations, so it is usually treated numerically.

To obtain a heuristic understanding of how a phase shift can reverse the process of gain and loss in a system we can consider the simple case of a driven harmonic oscillator. When a harmonic oscillator is driven at a frequency much below resonance, there is no phase shift between the oscillator and the driving force. This may be seen very easily by swinging a pendulum very slowly in your hand. If the oscillator is driven at a frequency much higher than the resonant frequency, the driving force leads the oscillator by a half cycle, or π . This is also easily observed with a hand held pendulum.

In the interesting case when the oscillator is driven at resonance, the driver (your hand with the pendulum for example) leads the oscillator by a quarter cycle, or $\pi/2$. Thus the driver is always slightly ahead of the oscillator mass, pulling it along and thereby transferring energy to it.

Consider what would happen if suddenly the driver shifted back by a half cycle, or π . Now the oscillator would be "in front" of the driver by $\pi/2$ and would be pulling the driver along, thereby transferring the energy in the oscillator back to the driver. This role reversal, of course, would not last very long because the oscillator would soon run out of energy, at which point one would expect the driver to regain its $\pi/2$ lead on the

oscillator and once again transfer energy to the oscillator in the manner we are familiar with.

The Raman system is in some ways analogous to the driven harmonic oscillator. In Raman scattering in H_2 the Stokes and pump fields beat at the resonant frequency of the molecule, thereby driving it into an excited vibrational state. The π phase shift in the Stokes field causes the molecules and Stokes field to give their energy back to the pump, thereby causing a brief depletion of the pump. This is the Raman soliton we observe experimentally.

Decay of Solitons in Raman Scattering

Theoretically, solitons were predicted to be possible in Raman scattering as early as 1975,¹³ however they were not expected to be experimentally realized due to the difficulty in producing the exact form of the fields and polarization necessary for propagation. Subsequent theoretical modeling³⁴ indicated that an instantaneous π phase shift in the input Stokes beam could initiate the formation of a soliton in a Raman amplifier. Since then solitons have been observed experimentally by placing a π phase shift electro-optically on the input beam to a Raman amplifier. However, the solitons amplitudes were found to vary from shot to shot.⁸ This was a mystery in the study of solitons in Raman scattering for some time.

Druhl *et al.*¹⁶ predicted that frequency detuning of the input Stokes beam from Raman resonance would lead to soliton decay. After this prediction MacPherson *et al.*⁵ found

that the output signal from a Raman generator can have frequency variations of the order of the gain narrowed linewidth. Since this output was used as the input signal for the soliton experiments, one might expect that the quantum initiated spectral fluctuations would lead to the observed soliton decay. As will be shown below, this suspicion is correct.

Experimental Observation of Soliton Decay

The experimental configuration for studying the statistics of soliton generation and decay is shown in Fig.13. A single-mode, frequency doubled Nd:YAG laser at 532 nm was used to pump both a Raman generator and a Raman amplifier. Multipass cells were used to reduce higher order processes such as second Stokes generation and anti-Stokes generation.²⁴ The Raman scattering was done in 10 atm. of hydrogen on the vibrational $Q_{01}(1)$ transition at room temperature.

An electro-optic crystal was used to place a π phase shift on the Stokes beam input to the Raman amplifier. Temporal profiles of the input and output pulses of the Stokes and pump beams were monitored with fast photodiodes and 500 MHz oscilloscopes. Overall our detection system had a 0.81 ns resolution full width at half maximum. The 532 nm pump laser had a temporal intensity profile that was near gaussian with a full width at half maximum of 17.7 ns. The Stokes beam input to the Raman amplifier was monitored with a Fabry-Perot interferometer in a diverging beam mode capable of single shot frequency resolution of 15 MHz. This interferometer had a free spectral range of 1

GHz with a finesse of approximately 70. The interferometer allowed us to monitor the Stokes frequency spectrum and reject shots with multiple peaks in frequency. This was done to insure that the π phase shift which was placed on the Stokes beam was the dominant phase information of the pulse.

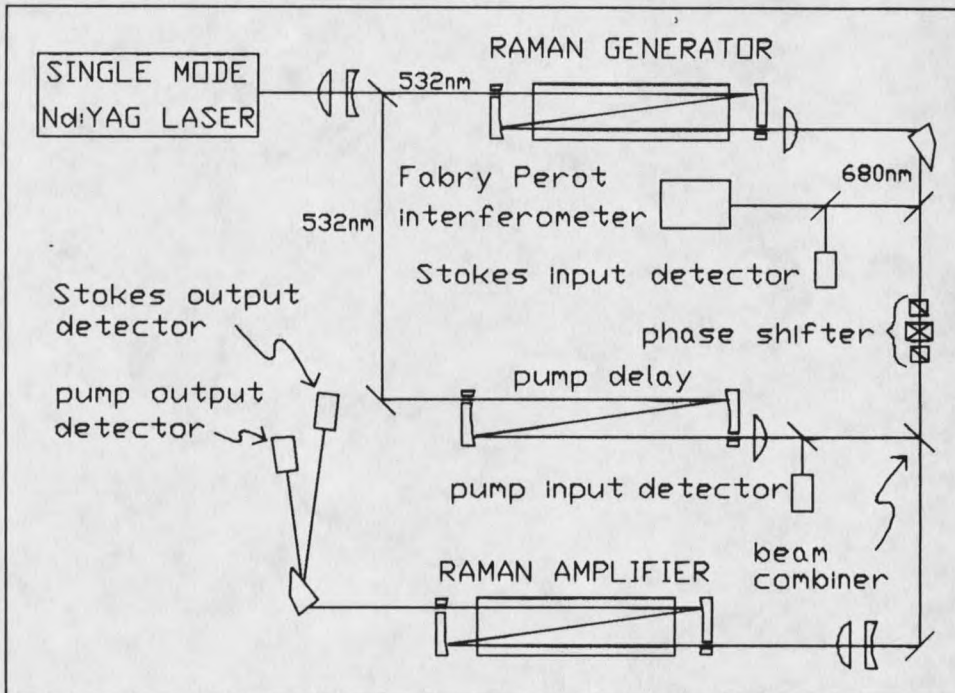


Figure 13 Experimental apparatus used to induce and record solitons in Raman scattering.

We found that every time a π phase shift was placed on the input Stokes beam, a soliton pulse in the region of pump depletion appeared. However, the amplitude of the soliton varied shot to shot indicating that the solitons were decaying. The soliton pulse shown in Fig.12 was one of the largest solitons seen for this level of pump depletion.

To study soliton decay we measured the fractional amplitude of 238 soliton pulses

which had a level of pump depletion similar to that shown in Fig.12.⁸ The fractional amplitude was taken to be the height of the soliton as a fraction of the maximum possible. For example, the soliton in Fig.12 has a fractional amplitude of 0.84. These shots were grouped into bins of size 0.05 and plotted as a histogram shown in Fig.14. Note that the majority of the solitons were large. Since the resolution of our detection system was only slightly narrower than the solitons, a full amplitude soliton would appear to be only about 84 percent of its true height. Since the peak of the distribution is near 0.84 this indicates that most of the input Stokes pulses were near resonance and experienced very little decay. If detuning of the injected Stokes beam caused the observed soliton decay, this fractional amplitude distribution should be directly related to the quantum statistics of the frequency output of the Raman generator. We explore this possibility in the next section.

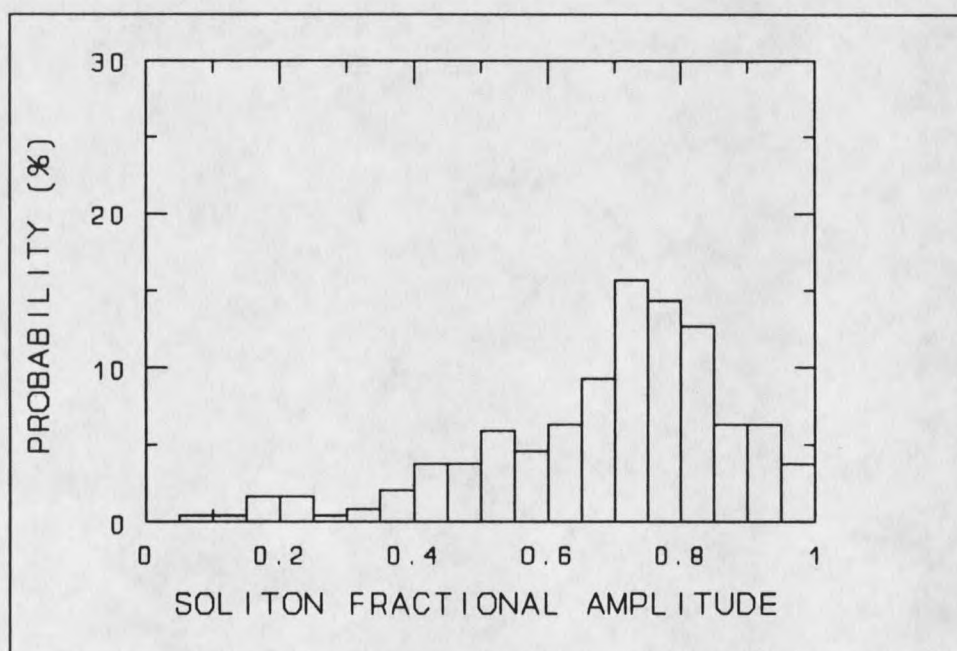


Figure 14 Histogram of experimentally measured solitons. Note that most of the solitons were large.

Theoretical Studies of Soliton Decay

In this section I will describe the method used to generate the theoretical distribution of Raman solitons when the input signal came from a Raman generator.

As described in Chapter 3 of this thesis, an ensemble of Stokes pulses can be generated by amplifying the field of a numerically generated "vacuum" field. Due to the random nature of the microscopic initiating field, the output field exhibits macroscopic variations from shot to shot. For example, in Fig.15 the spectrum of a Stokes pulse that happened to be near resonance is shown.

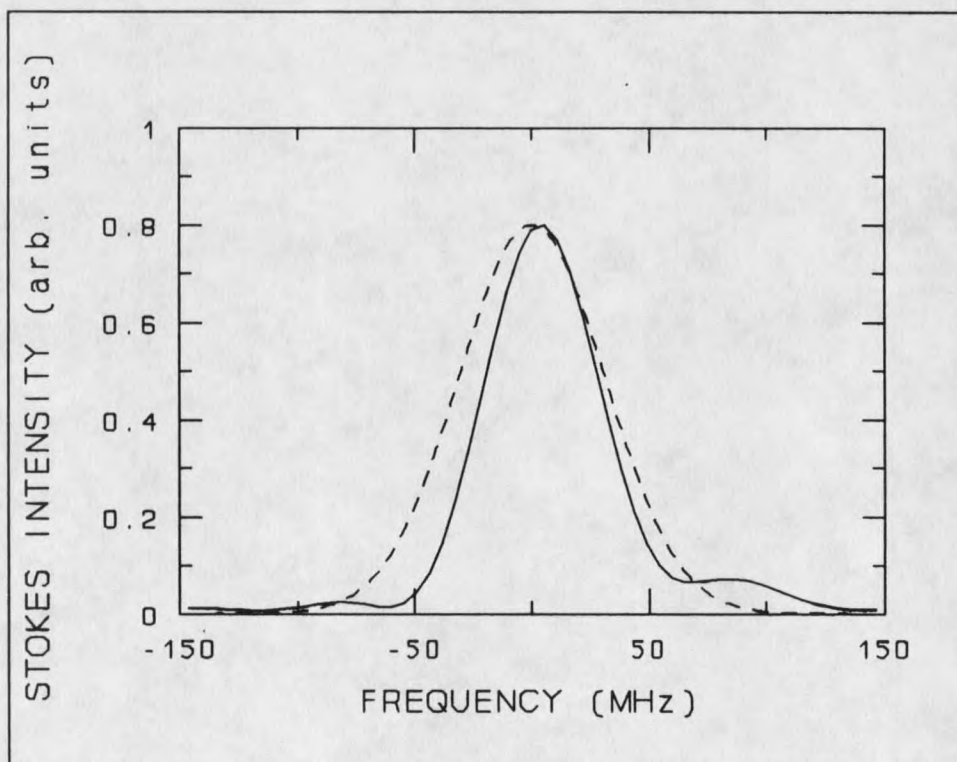


Figure 15 Near resonant, single shot Stokes spectrum. The analytical gain narrowed linewidth is shown as a dashed curve for comparison.

In Fig.16 an off resonance spectrum is presented with the gain narrowed linewidth again shown as a dashed curve. Note the distinct differences between these two spectra even though the only difference in the generation of these two spectra was the initiating "vacuum" field that was many orders of magnitude smaller than the output pulse. The spectra of the output Stokes pulses from the generator were found with a fast Fourier transform. Theoretically, this gave us the same information about the Stokes pulses as the Fabry-Perot interferometer did experimentally. The mean frequencies of an ensemble of numerically generated spectra and their standard deviations were calculated and recorded for every shot.

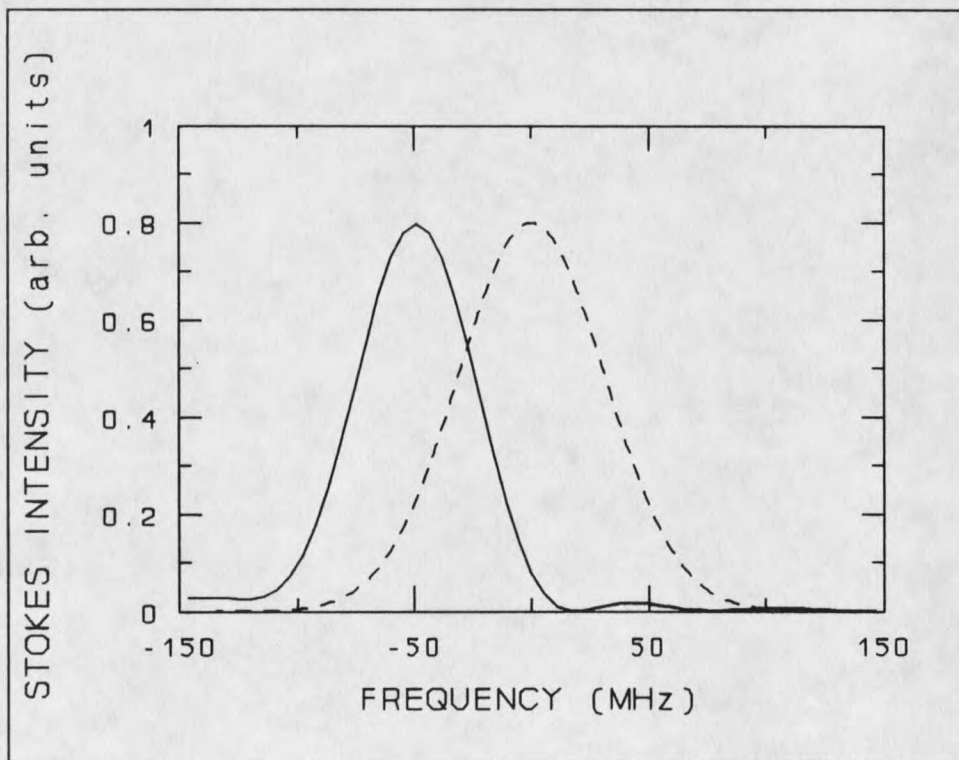


Figure 16 Single shot Stokes spectrum that is off resonance. The gain narrowed linewidth is shown as a dashed line for comparison.

To simulate the π phase shift introduced experimentally, the following phase shift function $B(\tau)$ was multiplied with each numerically generated Stokes field:

$$B(\tau)=1, \quad \tau < \tau_0$$

$$B(\tau) = \cos\left(\frac{\pi(\tau - \tau_0)}{\Delta\tau}\right), \quad \tau - \tau_0 \leq \Delta\tau \quad 4.1$$

$$B(\tau) = -1, \quad \tau > \tau_0 + \Delta\tau$$

The value for $\Delta\tau$ used was 2 ns. This additional phase shift inserted onto the Stokes pulse is, of course, in addition to the phase structure that occurred spontaneously during quantum initiation.

The Stokes pulse with the added phase shift was then input to a Raman amplifier along with fresh pump for numerical amplification, thus simulating the experiment shown in Fig.13.

Numerical amplification of this pulse to a level of depletion similar to that observed experimentally yielded pump intensity profiles with solitons in the depletion region. For example, the soliton resulting from the near resonant input signal shown in Fig.15 yielded the full amplitude soliton shown in Fig.17. The soliton that was obtained when the off resonant signal shown in Fig.16 was used as the input is shown in Fig.18. Note that the off resonant pulse produced a soliton that has decayed, in this case to a fractional amplitude of 0.54, reinforcing Druhl's¹⁶ claim that detuning of the input signal leads to

soliton decay.

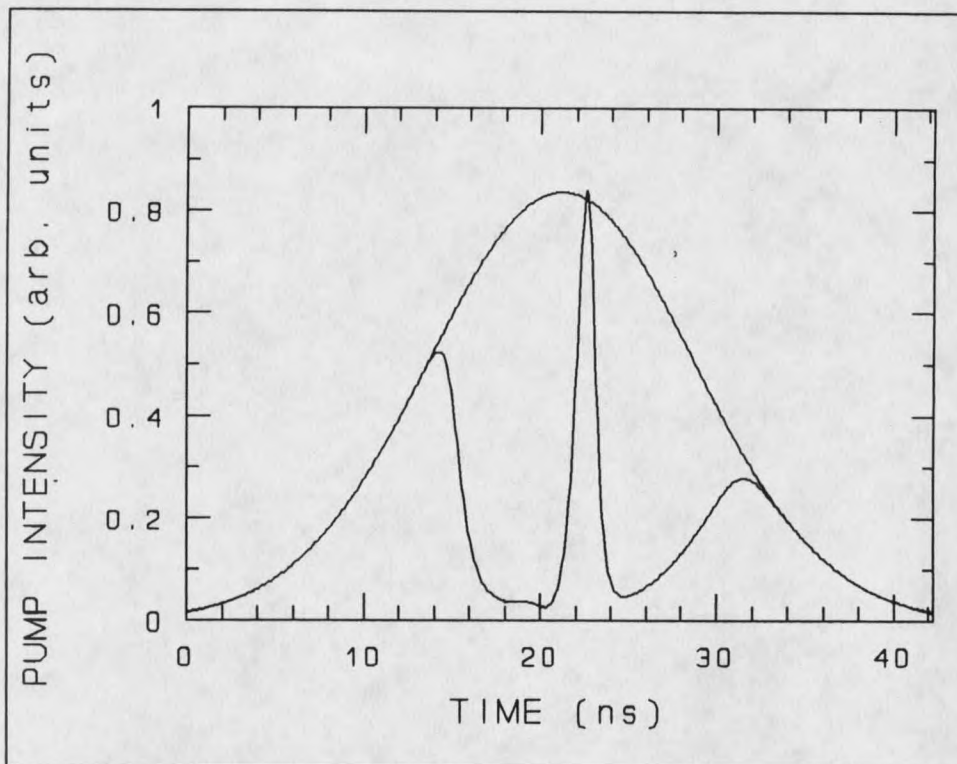


Figure 17 The soliton that resulted from the near resonance Stokes pulse shown in Fig.15.

To confirm that the experimentally observed soliton decay is related to the shot to shot fluctuations in the input Stokes signal, we numerically generated a soliton distribution analogous to the one measured experimentally. However, several experimental details had to be accounted for, such as the resolution of our detection system and variations in pump depletion due to shot to shot variations in the pump intensity. The temporal resolution of our experimental detection system was simulated by convoluting the numerically generated output pulses with a Gaussian, full width at half maximum 0.81 ns.

Numerically it was found that the soliton pulses narrow and, if they are off resonance, decay with additional pump depletion. Thus variations in the level of pump depletion leads to a spreading of the soliton amplitude distribution both because of the variations in decay and because the solitons in the more heavily depleted shots were not as well resolved. Therefore, an ensemble of solitons from a range of depletion levels was generated.

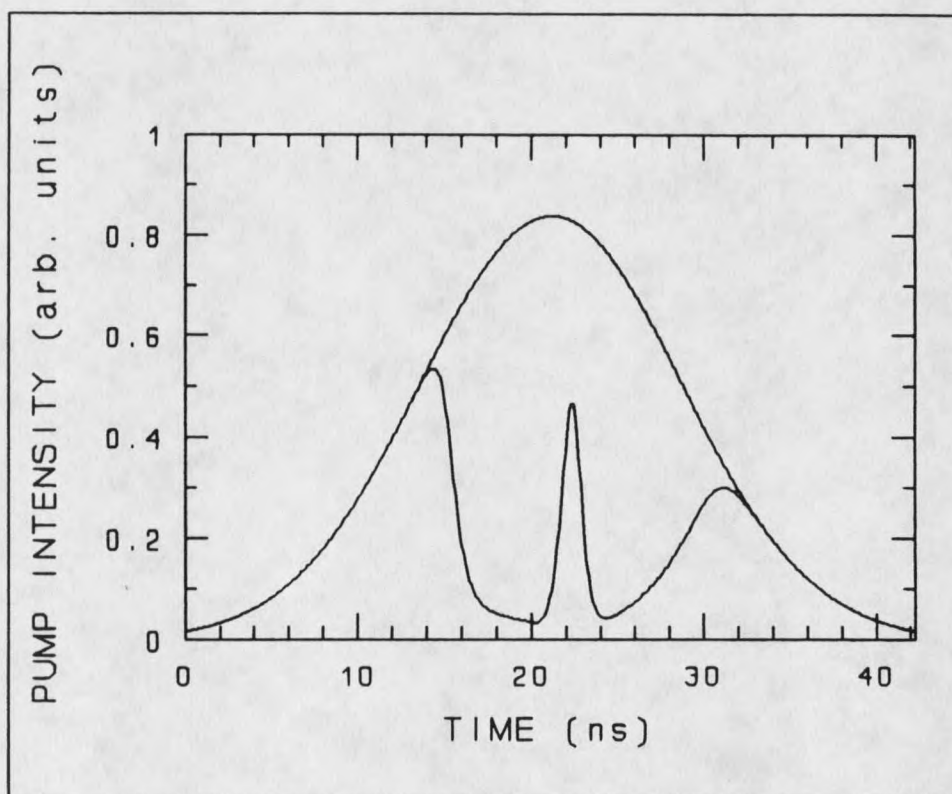


Figure 18 Soliton resulting from the off resonance Stokes spectrum shown in Fig.16.

A histogram of the predicted soliton amplitude distribution including these effects is presented in Fig.19. This distribution is similar to the experimentally measured histogram

in Fig.14 with most of the solitons having a large fractional amplitude. However, note that the theoretical distribution predicts more small amplitude solitons than were observed experimentally.

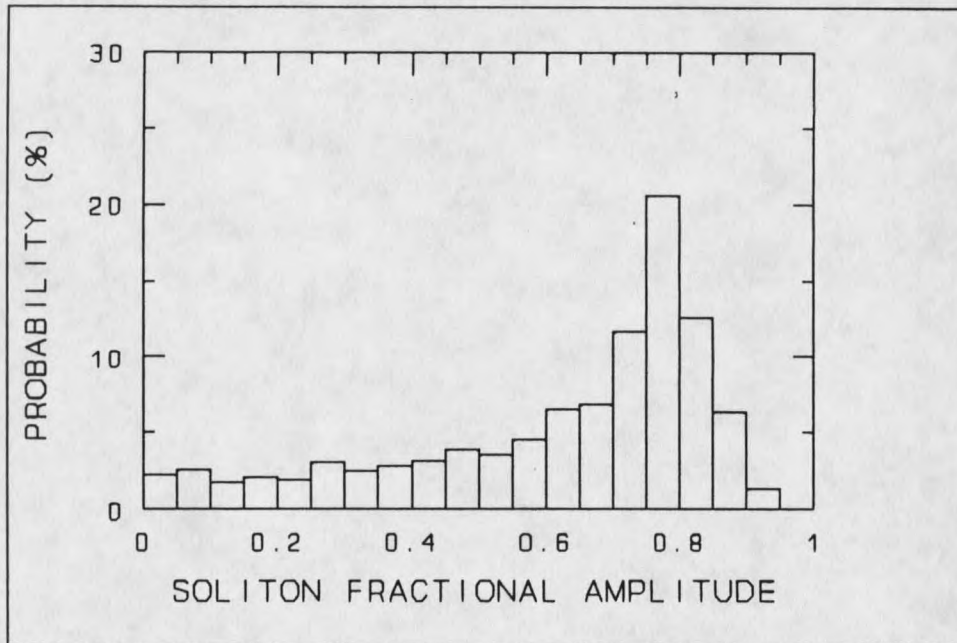


Figure 19 Histogram of all theoretically generated soliton amplitudes.

When the experiment was performed, those shots with noisy spectra were discarded from the ensemble. This was done to insure that the electro-optically induced π phase shift was the dominant phase information on the input signal.

As discussed in the last Chapter, noisy spectra can be identified by their large standard deviations from their mean values. (Noisy spectra have large frequency components far from their mean values, thus generating the large standard deviations.) Thus, by removing those shots that had large standard deviations (greater than 50 MHz)

from the ensemble, we effectively removed the noisy spectra shots as was done experimentally. When this was done, the distribution shown in Fig.20 was generated. Note that this distribution is quite similar to the experimental distribution in Fig.14, supporting the conjectured relationship between observed soliton decay and shot to shot fluctuations in the Stokes input signal initiated by quantum fluctuations.

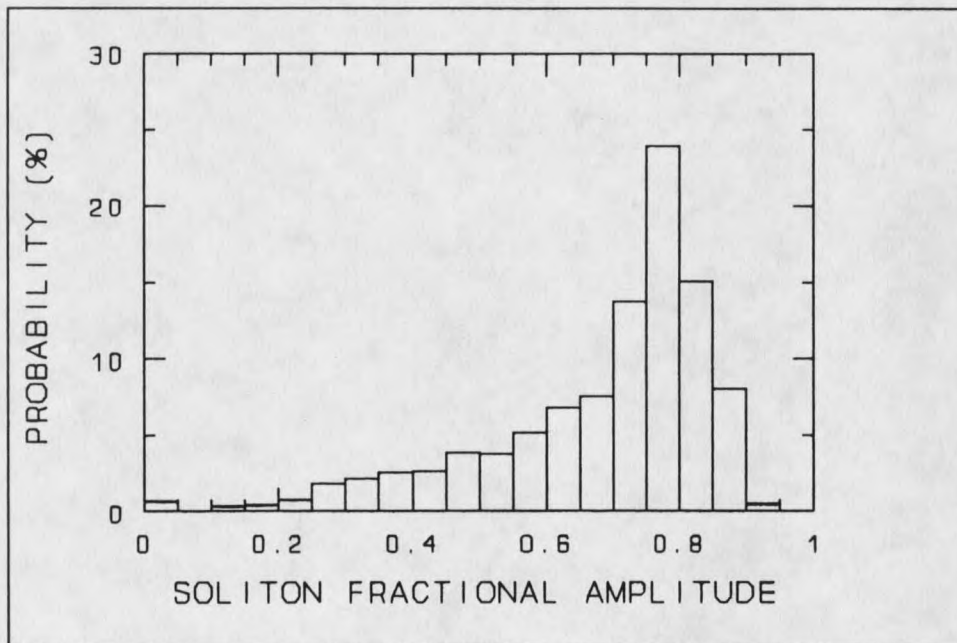


Figure 20 Histogram of soliton amplitudes when Stokes pulses with noisy spectra were excluded.

One might observe that the experimental distribution in Fig.14 appears somewhat wider than the theoretical distribution. We postulate that this may be due to shot to shot jitter in the temporal location of the electro-optic π phase shift. Phase shifts that occurred, for example, toward the rear of the pulse initiated solitons in regions where the pump intensity was lower. Consequently, those solitons experienced less gain. This is

analogous to experiencing less depletion, which leads to less narrowing of the pulse. These broader pulses were more resolvable to our detection system and therefore the distribution was broadened.

In conclusion, the distribution of observed soliton amplitudes in Raman scattering is directly related to the spectral fluctuations observed in the generation of the Stokes pulse. This research suggests that large amplitude solitons could be reproducibly generated with a stable, on resonance input signal such as tunable cw dye laser or Ti:Sapphire laser. One might be concerned that the power levels of these cw input sources would be so small that they would be dominated by the noise generated in the Raman amplifier. However, it appears that this is not a concern since the noise in a Raman amplifier can be dominated with only a few photons per mode. The studies that lead to this conclusion are the topic of the next Chapter of this thesis.

CHAPTER 5

THE QUANTUM MECHANICAL AMPLIFIER

The role of amplifiers in physics has been studied and discussed considerably. This is appropriate since the microscopic phenomena frequently studied emit signals that must be amplified for observation. One must then be concerned about whether the output of the amplifier yields an accurate representation of the microscopic phenomena or if it has been significantly altered during amplification.

Theoretical work has shown that bosonic linear amplifiers necessarily add quantum noise to the output signal.¹⁷⁻²⁰ (A linear amplifier is a device whose output modes are linearly related to its input modes.) For the output field to satisfy the commutation relations, noise must be added during the amplification, or put another way, noise is fundamentally required by the Heisenberg uncertainty relation. For large gain, the amount of noise that a linear amplifier will add to a signal will be at least what one would expect from the amplification of one quanta per mode at the input of a noiseless amplifier.

Past studies have shown that some real amplifiers can approach the minimum amount of noise required by quantum mechanics. For example, amplifiers based on dc SQUIDS^{35,36} have noise characteristics that approach the quantum limit. Other experiments using laser gain tubes have shown that the measured noise power^{37,38} is consistent with the quantum limit. The frequency noise in a four-frequency laser gyro has

also been found to be consistent with a laser gain tube operating near the quantum limit.³⁹ Recent results indicate that fiber amplifiers are approaching the quantum limit.⁴⁰ In a highly sensitive projection system using both a laser gain tube and a phase-conjugating mirror, the quantum limit was approached with only five photons required per resolution element.⁴¹

In Raman scattering a pump photon is scattered off a molecule, leaving it in an excited state and emitting a red shifted photon, referred to as a Stokes photon. This process can occur spontaneously, or it can occur via stimulation from an input Stokes field and thus act as an amplifier of the Stokes field. Experiments in Raman scattering have tended to fit into two distinct categories: Either they rely on spontaneous Raman scattering for initiation, where there is no input signal to amplify,⁴² or they have such a large input signal that spontaneous Raman scattering (noise) is insignificant.²⁴ The regime where both spontaneous emission and the input signal have a significant effect at the output of the amplifier occurs when the input signal has only a few photons per mode. Probing this region is interesting not only because it allows one to rigorously test the full quantum theory of Raman scattering,¹⁻³ but also because it explicitly demonstrates how good the Raman amplifier is.

In the regime of Raman scattering where both quantum noise and the input field significantly affect the output, recent work studying the spatial modes of a Raman amplifier⁴³ has shown that approximately 30 photons per mode in the input signal are required for the input signal's spatial mode to dominate the output spatial signal for 50% of the shots. Although this experiment indicated that there is not an excessive amount

of noise introduced in a Raman amplifier, a rigorous test of the theory with these results appears to be difficult.

In this Chapter I present recent results from an experiment using Raman amplifiers in each leg of a modified Mach-Zender interferometer.¹¹ In this experiment small signals were input to the interferometer which were then amplified within the interferometer. The quantum noise added to the signal during amplification manifested itself by degrading the ensemble average fringe pattern from the interferometer, which was quantified by measuring the fringe visibility.

Theoretical calculations of the fringe visibility are given in appendices. For simplicity, the analysis in Appendix C uses ideal, single mode amplifiers. This shows how the noise introduced by an amplifier degrades the visibility of the output fringe pattern with a model that can be analyzed analytically. Additionally, I show what conditions the amplifier must satisfy to minimize the number of noise photons. In Appendix D, the analysis is extended to Raman amplifiers, with similarities between the ideal, single mode amplifier and the Raman amplifier noted.

Interferometric Measurement of Quantum Noise

To study the noise added by an amplifier, consider the experiment diagramed schematically in Fig.21. A generator provides a signal for a Mach-Zender interferometer with amplifiers in each leg. The amplified fields from each leg of the interferometer are combined at the exit beam splitter with a slight angle between the combined fields to

produce a fringe pattern on the screen.

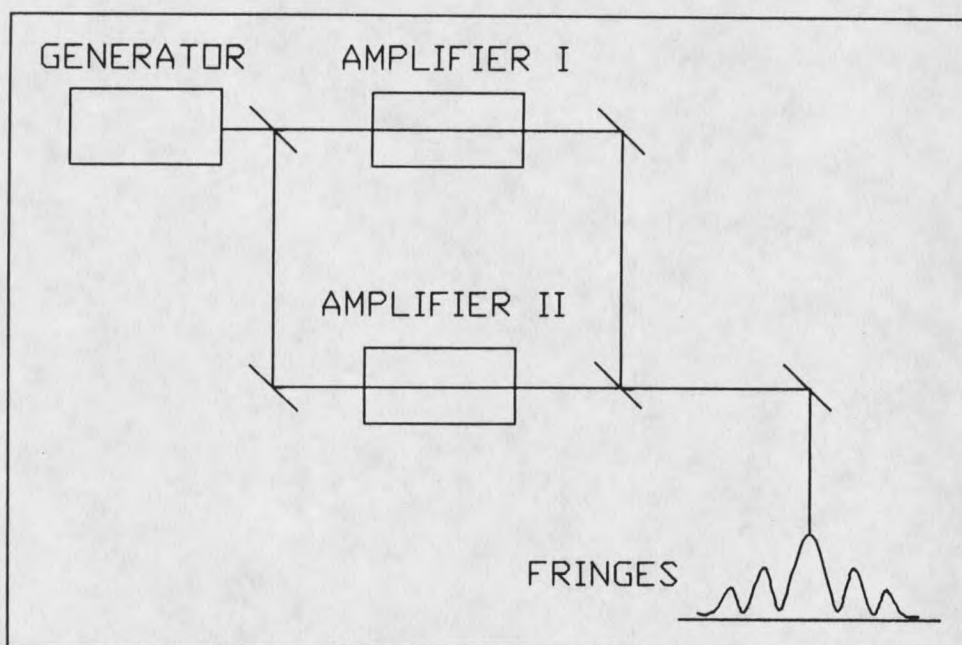


Figure 21 A Mach Zehnder interferometer with an amplifier in each leg. The noise added by the amplifiers degrades the output fringes.

The output field from an amplifier can be thought to consist of a superposition of the field due to stimulated emission from the input signal along with the field due to amplified spontaneous emission.⁴⁴ As one would expect, the amplified spontaneous emission (noise)⁴⁵ from two separate amplifiers is uncorrelated in phase⁴⁶ and therefore the relative phase difference between the combined fields is random from shot to shot, leading to random positions of the peaks and troughs in the output fringe pattern. Since the peak and trough positions are random from shot to shot, the ensemble average of such fringe patterns yields a smooth profile.

On the other hand, amplification of the input signal by stimulated emission

preserves the phase of the input field. Therefore, the amplified fields resulting from stimulated emission of the input signal have a well defined phase difference between the two legs that is dependent on the interferometer. This leads to fringe patterns that are stationary from shot to shot. Thus the ensemble average of the fringe patterns due to the field from stimulated emission retains well defined fringes.

In general the ensemble average of the output fringe patterns will consist of a smooth profile with a fringe pattern on top.⁴⁷ The fringe pattern is attributed to the amplified signal while the smooth profile is attributed to the noise. The larger the field arising from the input signal relative to the amplifier added noise, the larger the peaks will be compared to the smooth profile. To obtain a quantitative measure of the degradation of the fringe pattern due to the amplifier added noise the visibility V is calculated:

$$V = \frac{\langle I \rangle_{\max} - \langle I \rangle_{\min}}{\langle I \rangle_{\max} + \langle I \rangle_{\min}} \quad (5.1)$$

where $\langle I \rangle_{\max}$ is the intensity of the peak of the ensemble fringe pattern and $\langle I \rangle_{\min}$ is the intensity of the trough of the ensemble fringe pattern. A smooth intensity profile is one where $\langle I \rangle_{\max} = \langle I \rangle_{\min}$, and, as can be seen from Eq.(5.1) has a visibility of 0. On the

other extreme, a "perfect" ensemble fringe pattern is one in which $\langle \hat{f} \rangle_{\min} = 0$ and $\langle \hat{f} \rangle_{\max}$

is non-zero. Such a fringe pattern has a visibility of 1. For this to happen the phase information of the input signal has to be preserved on every shot, which means that the amplified signal must dominate the quantum noise added during amplification. As we will see, this limit is approached asymptotically when the input signals become large.

In the following section we consider the results one expects when ideal, single mode, linear amplifiers are used in Fig.21.

Ideal, Single Mode, Linear Amplifier

In this section an idealized amplifier whose output electric field can be expanded in some set of modes such that only one mode is significantly populated is considered. Rather than concern ourselves with the specifics of the modes chosen, we simply assume the operator characteristics of this field are described by Bosonic creation and annihilation operators.

With these ideal, single mode, linear amplifiers, the visibility of the interferometer diagramed in Fig.21 is calculated to demonstrate the role of quantum noise in this experiment. Using a formalism similar to that presented by Caves,¹⁷ the single mode field is described with creation and annihilation operators that have the free propagation time dependence ($\exp(\pm i\omega t)$) folded out. In particular we choose the model where the

amplifier output mode is linearly related to the input mode by:

$$\hat{b}_\alpha = M_\alpha \hat{a}_\alpha + \hat{N}_\alpha \quad (5.2)$$

where $\hat{b}_\alpha$ is the annihilation operator describing the output field of the amplifier, M_α

is related to the gain of the amplifier, $\hat{a}_\alpha$ is the input field annihilation operator, and $\hat{N}_\alpha$

is the operator governing the growth of noise in the amplifier, which depends on the amplifier internal modes. (This is not the most general form for an ideal, single mode amplifier, but one that readily demonstrates the analysis of the interferometric method used to measure noise.) The subscript α designates the amplifiers in Fig.21 and can take the values *I*, *II*, or *G* for amplifier I, amplifier II, or the generator respectively. Note that these subscripts do not imply complete independence, for example, $[\hat{b}_I, \hat{b}_{II}^\dagger] \neq \delta_{I,II}$. We

will assume that the noise field has random phase, and therefore:

$$\langle \hat{N}_\alpha \rangle = 0 \quad (5.3a)$$

Additionally, we assume the internal modes of one amplifier are independent from the internal modes of another. This assumption along with Eq.(5.3a) leads to:

$$\langle \hat{N}_\alpha \hat{N}_\beta^\dagger \rangle = \langle \hat{N}_\alpha^\dagger \hat{N}_\beta \rangle = 0 \quad \alpha \neq \beta \quad (5.3b)$$

and similarly for all other combinations of the noise operator and its Hermitian adjoint for $\alpha \neq \beta$.

Since photons are bosons, the input and output field operators of the amplifiers must satisfy the Bose-Einstein commutation relations:

$$[\hat{a}_\alpha, \hat{a}_\alpha^\dagger] = 1 \quad (5.4a)$$

$$[\hat{b}_\alpha, \hat{b}_\alpha^\dagger] = 1 \quad (5.4b)$$

One can see that if there was not a noise term in Eq.(5.2), the output field mode would not satisfy the commutation relation Eq.(5.4b). Combining Eqs.(5.2) and (5.4), one finds:

$$[\hat{N}_\alpha, \hat{N}_\alpha^\dagger] = (1 - |M_\alpha|^2) \quad (5.5)$$

where we have used the independence of the internal modes of the amplifier from the external, input field mode.

These are the essentials of the ideal, single mode amplifier model. Equation (5.5) places a considerable restriction on the noise operator, but does not distinguish between high noise and low noise amplifiers. The condition the noise operator $\hat{N}^\dagger$ must satisfy for a low noise amplifier is discussed in Appendix C.

Using this model for the ideal, single mode amplifier, the output visibility of the interferometer shown in Fig.21 as a function of the input field can be calculated straightforwardly. This is also done in Appendix C. For the specific example of a balanced interferometer where the amplifiers are identical as are the input and output beam splitters, the following form for the visibility is found:

$$V = \frac{\langle \hat{b}_G^\dagger \hat{b}_G \rangle}{\langle \hat{b}_G^\dagger \hat{b}_G \rangle + \frac{1 - |M|^{-2} + \langle \hat{N} \hat{N}^\dagger \rangle}{2t^2 r^2}} \quad (5.6)$$

where t (r) is the transmission (reflection) coefficient of the beam splitters in Fig.21 and

$\langle \hat{b}_G^\dagger \hat{b}_G \rangle$ is the expectation value for the number of photons input to the interferometer.

If the term $\langle \hat{N} \hat{N}^\dagger \rangle$ is large, which as shown in Appendix C indicates a noisy amplifier,

then the visibility is diminished. Not surprisingly, the condition for maximum visibility from the interferometer is the same as required for minimizing the number of noise photons as discussed in Appendix C:

$$\langle \hat{N} \hat{N}^\dagger \rangle = 0 \quad (5.7)$$

If one chooses a single mode, amplifier with the minimum noise requirement of

Eq.(5.17), uses beam splitters with 50/50 transmission/reflection ratios, and goes to high gain (i.e. $|M|^2 \gg 1$), the visibility expression reduces to the simple form:

$$V = \frac{\langle \hat{b}_G^\dagger \hat{b}_G \rangle}{\langle \hat{b}_G^\dagger \hat{b}_G \rangle + 2} \quad (5.8)$$

Note that with two photons input ($\langle \hat{b}_G^\dagger \hat{b}_G \rangle = 2$) to the interferometer (and therefore an average of one photon to each amplifier), the visibility is 0.5. The visibility as a function of the average number of photons input to each amplifier is shown in Fig.22. This plot shows that even with an input signal of only 8 photons (or 4 photons on average per amplifier), the output visibility is 0.8, indicating that the noise can be dominated with quite small inputs.

In principle, the visibility found when using Raman amplifiers in Fig.21 can be calculated in the same manner as using the ideal, single mode amplifiers. Unfortunately, the mathematics is considerably more complex. Additionally, we were interested in the slightly different experimental configuration shown in Fig.23. The details of the visibility calculations using Raman amplifiers in both the ideal Mach-Zender (Fig.21) and the experimental (Fig.23) interferometers are given in Appendix D. Results from this theoretical model are given later in this Chapter when we present the experimental results.

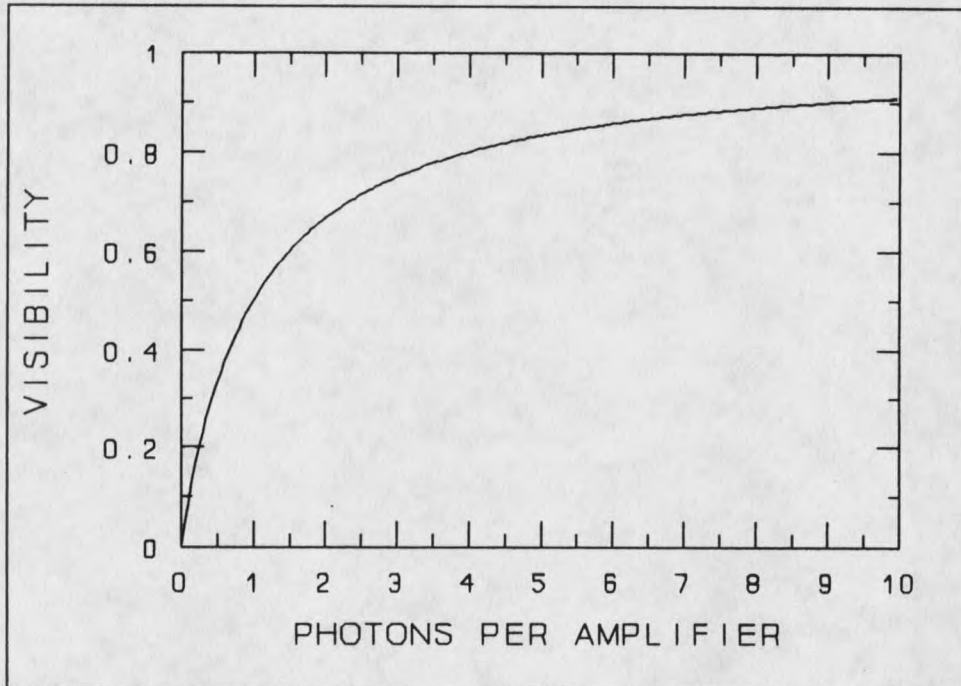


Figure 22 The visibility as a function of the average number of photons in each leg of the interferometer shown in Fig.1.

Interferometer Experiment and Results

The experimental apparatus used to measure the noise added by a Raman amplifier is diagramed in Fig.23. One should note that the apparatus diagramed in Fig.23 is essentially the same as the Mach-Zender interferometer shown in Fig.21. A generator provides a signal to an interferometer with amplifiers in each leg. The output fringes are imaged onto a linear diode array for data collection. The only difference from Fig.21 is

that in Fig.23 the interferometer has a cyclic geometry for improved stability over the standard Mach-Zender interferometer. A detailed description of the apparatus and experimental details is given in Appendix D.

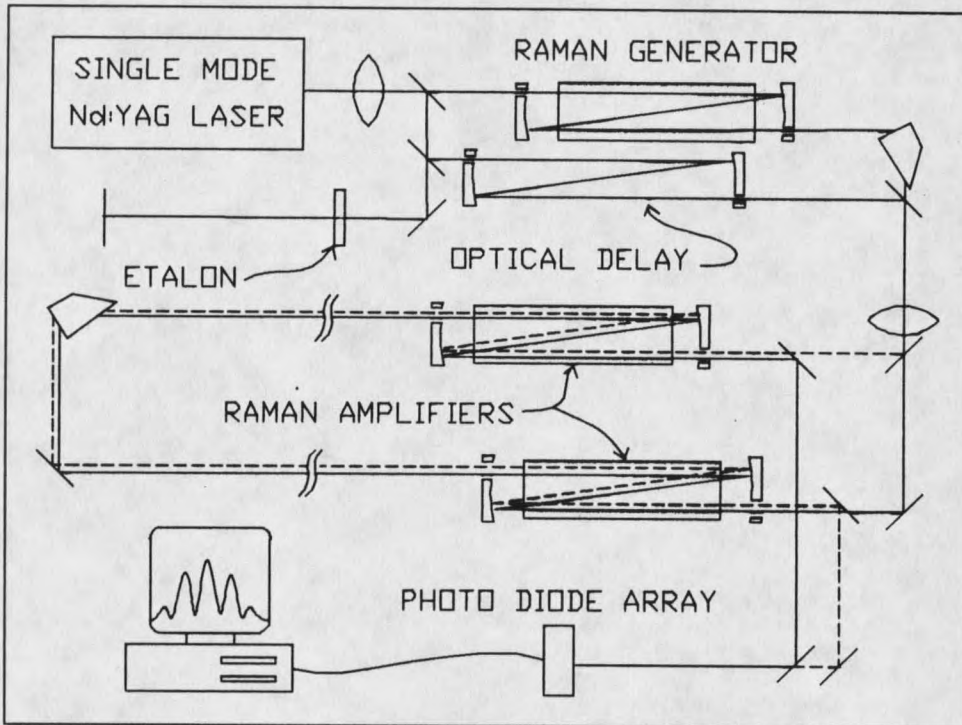


Figure 23 Experimental apparatus used to measure noise in a Raman amplifier. The cyclic geometry of the interferometer greatly improved the stability.

To compare theory with experiment, ensemble averages of the output fringe patterns from the interferometer were formed. From these, the visibility was measured. The visibility was expected to increase with an increased input Stokes signal which was a function of the pump energy input to the Raman generator. Therefore, numerous runs were made with different input pump energies to the Raman generator. This allowed us

to obtain the visibility as a function of the pump energy input to the Raman generator.

To clarify the significance of our results we used the quantum mechanical theory of stimulated Raman scattering¹⁻³ to calculate the number of photons emitted from the Raman generator for various input pump energies.

Then, including the effects of the transmission of the optics between the Raman generator and the interferometer (68% transmission) and the two beam splitters a beam encounters before reaching an amplifier,⁴⁸ the average number of photons reaching an amplifier was calculated. Finally, we estimated the number of Stokes temporal modes that had a significant population of photons to be the ratio of the pump linewidth to the Stokes gain narrowed linewidth. Due to gain narrowing,³¹ this ratio becomes smaller at large gains in the generator. For example, the ratio ranges from 22.3 for spontaneous Raman scattering at extremely low gain to around 4 at large gains. In this manner, the input signal was transformed to units of number of input photons per mode per amplifier.

In Fig.24 the ensemble average of 284 fringe patterns is presented when no input Stokes signal was put into the interferometer. As one would expect, there is no well defined fringe pattern. However, as the input signal to the interferometer is increased, the fringe pattern develops. For example, in Fig.25 an ensemble of 832 fringe patterns with an average of .13 photons per mode per amplifier shows well defined fringes on top of a smooth background. As noted above, the fringe pattern can be thought to arise from the stimulated signal, and the smooth background is the result of amplified noise in the amplifiers.

As the input signal is increased further, the amplified signal becomes more

dominant, as shown in Fig.26 with an input of .59 photons per mode per amplifier, and Fig.27 with an input of over one hundred photons per mode per amplifier where the noise is quite well dominated.

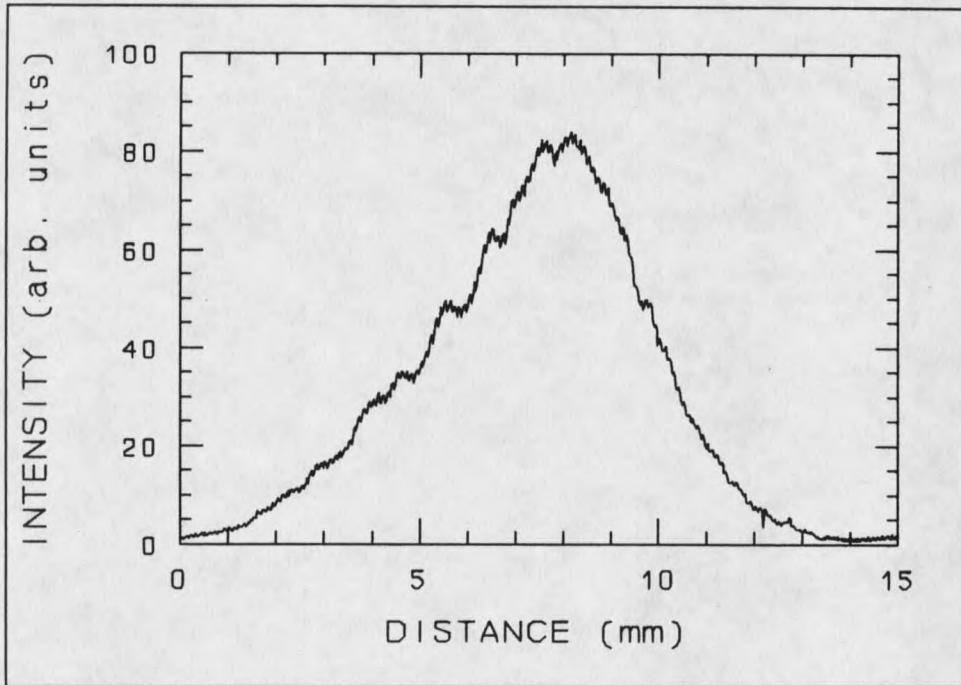


Figure 24 Ensemble average of 284 fringe patterns when no input signals were input to the interferometer. The measured visibility is .06.

The measured visibility for Fig.24, which consists entirely of noise, is near zero ($V=.06$). The visibility of the fringe pattern in Fig.27 where the noise is quite well dominated approaches 1 ($V=.95$). The effect of the gaussian envelope has been folded out of the visibility measurements. These plots very clearly demonstrate how the output visibility is quantitatively related to the relative amount of noise in the output pulses of the amplifiers. In Fig.28 we present the visibility as calculated from the theoretical model

of the interferometer shown in Fig.23 along with our experimental measurements.

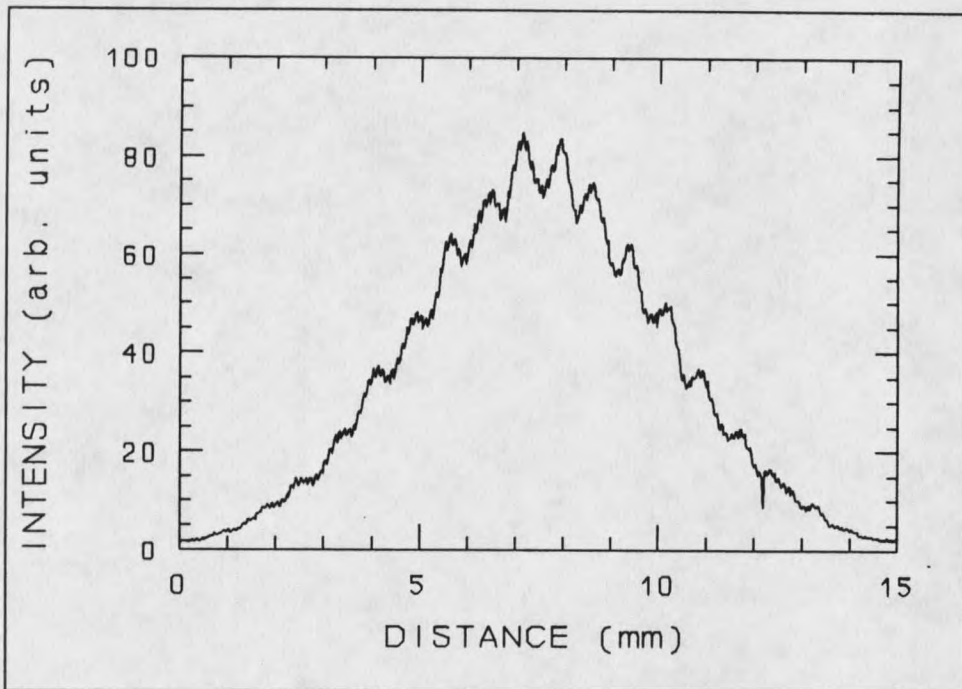


Figure 25 Small fringes are visible on this ensemble of 832 shots with an average of .13 photons per mode input into each amplifier. The measured visibility was .09.

Figure 28 shows the experimentally measured visibility (X's) as a function of the average number of photons per mode input to the amplifiers. The solid curve in Fig.28 represents the theoretical results from the Raman theory given in Appendix D. For comparison, the results for the same interferometer using ideal, single-mode amplifiers in place of the Raman amplifiers are shown as a dashed line. Note the similarity between the results when using ideal, single mode amplifiers and the results when using Raman amplifiers. As one would expect, the visibility goes up as the number of photons per mode input to the amplifiers increases.

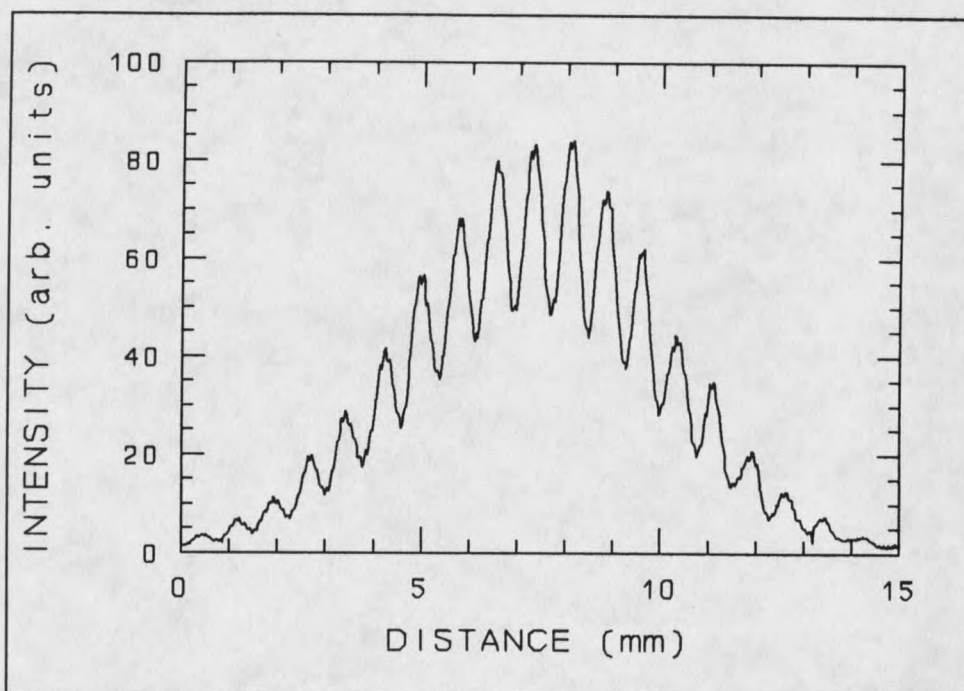


Figure 26 With an average of .59 photons per mode input into each amplifier, the ensemble average fringe pattern has a visibility of .30. There were 644 shots included in this ensemble.

One may note that the experimentally measured visibility in Fig.28 appears to rise more steeply than the theory. This may be due to slight variations in the pump temporal width that occurred during the data collection. To see how this affects the theoretical results the visibility is plotted as a function of the input pump energy to the Raman generator in Fig.29 for two different temporal widths of the pump. The X's are the experimentally measured results, the solid line is the theoretical result with the measured temporal width of the pump, and the dashed line is the theoretical result with the temporal width of the pump reduced by ten percent, roughly the uncertainty in our measurement. When the experiment was performed the temporal width of the pump was only monitored occasionally. Thus temporal narrowing of the pump during the later part of the day when

the higher visibility data points were taken could have lead to the steeper than expected curve in the experimental results.

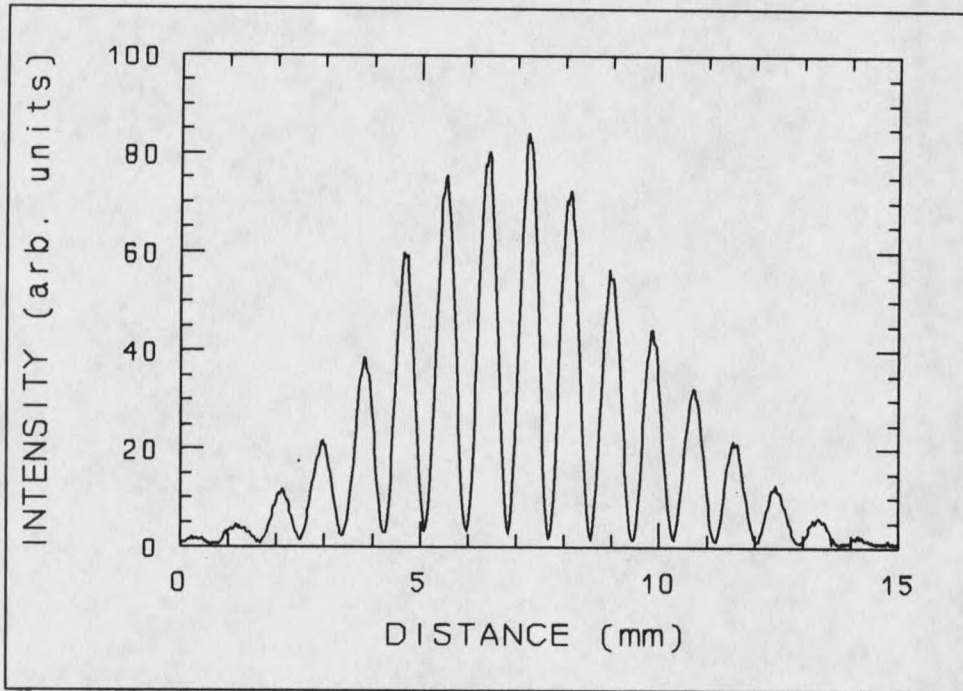


Figure 27 Over one hundred photons per mode were input into each amplifier on average to obtain these deep fringes. The measured visibility for this ensemble of 188 shots was .95.

Additionally, the experimental data appears to asymptotically approach a maximum visibility somewhat lower than what theory predicts, which could be caused by an imbalance in the interferometer. To test the interferometer we periodically measured the visibility with no pump supplied to the amplifiers, thus making the amplifiers passive. These measurements yielded an average visibility of .96, very near the asymptotic value the experimental data appears to approach.

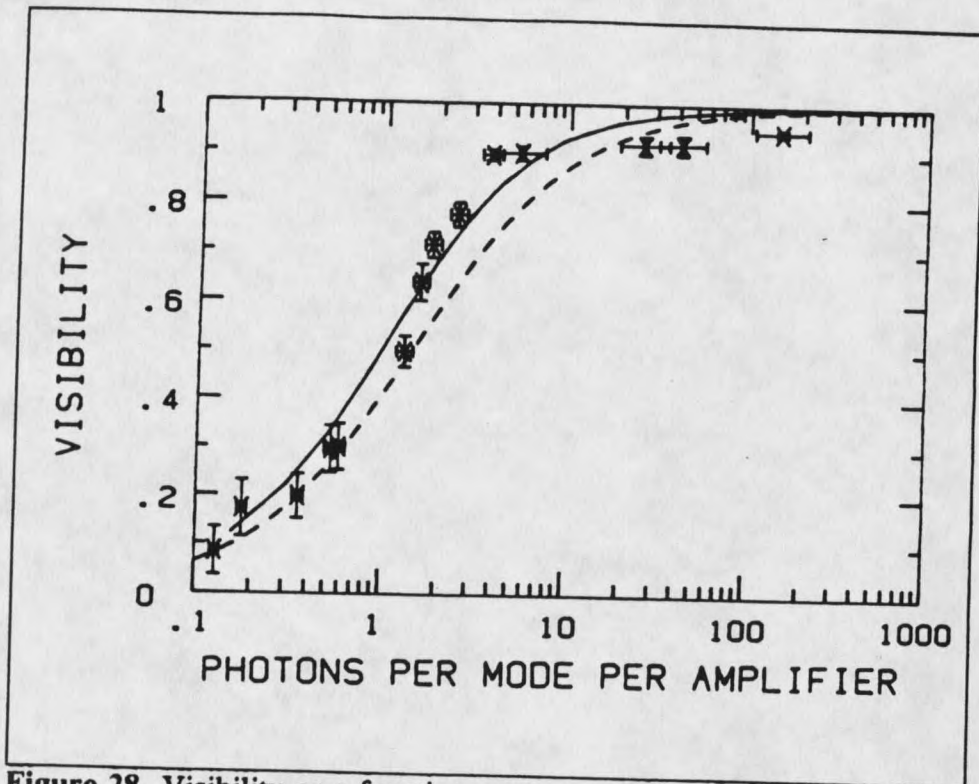


Figure 28 Visibility as a function average number of photons per mode input per amplifier. The X's are experimental results, the solid curve is from Raman theory, and the dashed curve is from ideal, single mode amplifier theory.

In conclusion, the above data indicate that the noise added in Raman amplification is in agreement with what is predicted theoretically by the theory of Raymer and Mostowski.¹⁻³ Since the noise emitted by the amplifiers is fundamentally required by the Heisenberg uncertainty relation, the Raman amplifier is an example of quantum mechanically limited linear amplifier. This noise can be dominated by input signals with only a few photons per mode, indicating that extremely small signals can be amplified with little distortion. And finally, the interferometric method described above is a practical method to measure the noise characteristics of light amplifiers, providing easily

interpretable results.

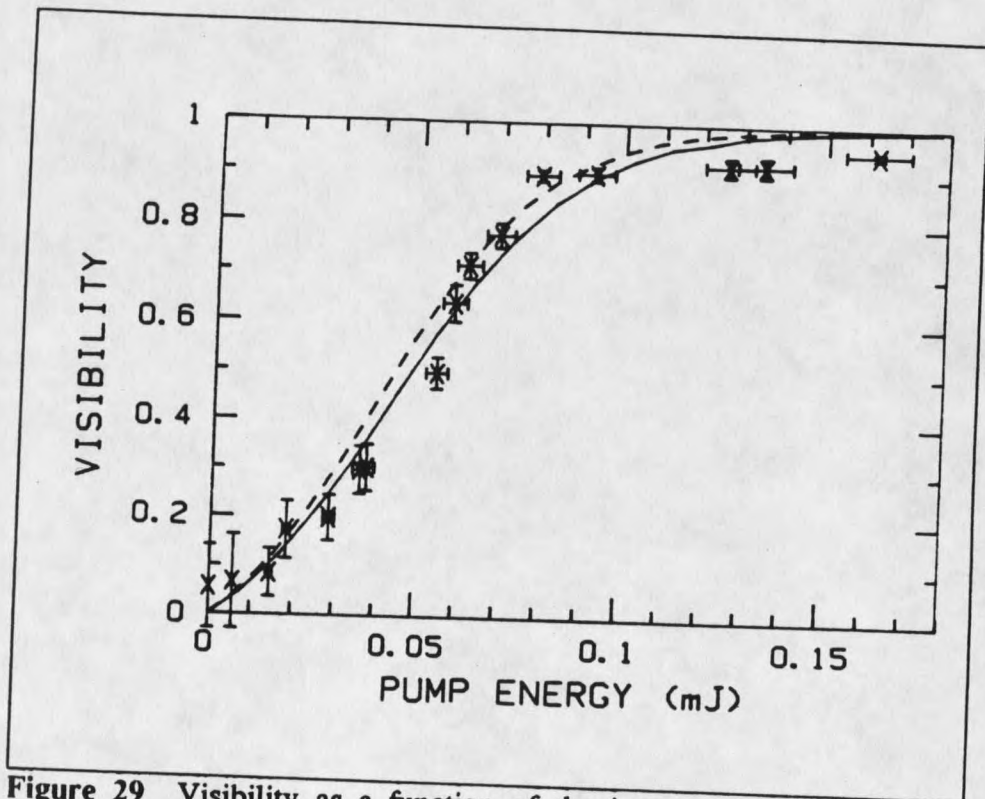


Figure 29 Visibility as a function of the input pump energy to the generator. The X's indicated experimental results. The two curves are from Raman theory with different duration pump pulses.

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APPENDICES

APPENDIX A

COLLISIONAL DEPHASING OF THE RAMAN MEDIUM

In this Appendix the damping of the Raman medium polarization is considered. The phenomenological damping of the medium requires an additional Langevin operator to be included to maintain operator consistency.^{2,3}

Using the Heisenberg picture, the following operator for a molecule in the Raman medium polarization can be obtained:

$$\frac{d\hat{Q}^\dagger(t)}{dt} = -i\kappa_1 E_p^*(z,t) \hat{E}_s^{(-)}(z,t) (\hat{\sigma}_{33}(t) - \hat{\sigma}_{11}(t)) \quad (\text{A.1})$$

where the slowly varying polarization operator $\hat{Q}^\dagger(t) = \hat{\sigma}_{31}(t) \exp[-i(\omega_p - \omega_s) + i(k_p - k_s)]$,

t is the time, z is the position of the Raman active molecule along the pumped pencil shaped region, and $\hat{\sigma}_{kj}(0) = |k\rangle\langle j|$. The terms not defined here are given in the main

text.

To include collisional dephasing into the theory, Eq.(A.1) was phenomenologically altered to be:

$$\frac{d\hat{Q}^\dagger(t)}{dt} = -\Gamma \hat{Q}^\dagger(t) - i\kappa_1 E_p^*(z,t) \hat{E}_s^{(-)}(z,t) [\hat{\sigma}_{33}(t) - \hat{\sigma}_{11}(t)] + \hat{F}^\dagger(t) \quad (\text{A.2})$$

where the first term on the right hand side ($-\Gamma\hat{Q}^\dagger(t)$) accounts for collisional damping, and the last term on the right hand side ($\hat{F}^\dagger(t)$) is a quantum statistical Langevin operator necessary to maintain operator consistency. One can think of the Langevin operator as the operator that accounts for the random collisions in the medium. The Langevin operator is needed to maintain equalities such as $\langle\hat{Q}^\dagger(t)\hat{Q}(t)\rangle=\hat{\sigma}_{11}(t)$.

The quantum Langevin operator has the following properties that can be found from quantum reservoir theory:^{3,49}

$$\langle\hat{F}(t)\rangle=0 \quad , \quad (A.3)$$

$$\langle\hat{F}^\dagger(t)\hat{F}(t')\rangle=2\Gamma\langle\hat{\sigma}_{11}(t)\rangle\delta(t-t') \quad , \quad (A.4)$$

$$\langle\hat{F}(t)\hat{F}^\dagger(t')\rangle=2\Gamma\langle\hat{\sigma}_{33}(t)\rangle\delta(t-t') \quad . \quad (A.5)$$

To demonstrate the need for the Langevin operator, consider the simplest situation. Let the Raman active molecule be in its ground state at $t=0$. Then, without turning the pump laser on, observe the predicted evolution of the molecule. Equation (A.2) is easily integrated to obtain:

$$\hat{Q}^\dagger(t) = \hat{Q}^\dagger(0)e^{-\Gamma t} + \int_0^t dt' e^{-\Gamma(t-t')} \hat{F}^\dagger(t') \quad (\text{A.6})$$

The normally ordered polarization operator is calculated to be:

$$\langle \hat{Q}^\dagger(t) \hat{Q}(t) \rangle = \langle \hat{Q}^\dagger(0) \hat{Q}(0) \rangle e^{-2\Gamma t} + \int_0^t dt' \int_0^t dt'' e^{-\Gamma(2t-t'-t'')} \langle \hat{F}^\dagger(t') \hat{F}(t'') \rangle \quad (\text{A.7})$$

where cross terms between the polarization operator and the Langevin operator do not contribute. Now, noting that $\langle \hat{Q}^\dagger(t) \hat{Q}(t) \rangle = \hat{\sigma}_{11}(t)$ one can see that without the Langevin operator, the inclusion of the damping term alone would predict spontaneous disappearance of matter. This is, of course, not observed experimentally.

In this example, $\hat{\sigma}_{11}(t)$ is time independent, so Eq.(A.7) can easily be solved to yield:

$$\langle \hat{Q}^\dagger(t) \hat{Q}(t) \rangle = \langle \hat{\sigma}_{11}(t) \rangle \quad (\text{A.8})$$

which is the desired result.

To treat the spacial evolution of the Stokes field, a slice averaged atomic operator was constructed. So, for example the slice averaged polarization operator is:²

$$\hat{Q}(z,t) = \frac{1}{n} \sum_{\alpha} \hat{Q}^{\alpha}(t) \quad (\text{A.9})$$

where α signifies the Raman active molecules in a slice of material at position z that is thin compared the Stokes wavelength. Similar slice averaged operators were constructed for the other atomic operators as well.

The correlation functions for the slice averaged polarization and Langevin operators are:

$$\langle \hat{Q}^{\dagger}(z,0) \hat{Q}(z',0) \rangle = (AN)^{-1} \langle \hat{\sigma}_{11}(z,0) \rangle \delta(z-z') \quad (\text{A.10})$$

$$\langle \hat{Q}(z,0) \hat{Q}^{\dagger}(z',0) \rangle = (AN)^{-1} \langle \hat{\sigma}_{33}(z,0) \rangle \delta(z-z') \quad (\text{A.11})$$

$$\langle \hat{F}^{\dagger}(z,t) \hat{F}(z',t') \rangle = \frac{2\Gamma}{AN} \langle \hat{\sigma}_{11}(z,t) \rangle \delta(z-z') \delta(t-t') \quad (\text{A.12})$$

$$\langle \hat{F}(z,t) \hat{F}^{\dagger}(z',t') \rangle = \frac{2\Gamma}{AN} \langle \hat{\sigma}_{33}(z,t) \rangle \delta(z-z') \delta(t-t') \quad (\text{A.13})$$

For our experiments, the Raman medium remained primarily in its ground state. Another way of thinking of this is that very few molecules in any slice of the Raman active

medium participated in the Raman scattering process. Consequently the approximations

$\langle \hat{\sigma}_{11}(z,t) \rangle \cong 1$ and $\langle \hat{\sigma}_{33}(z,t) \rangle \cong 0$ are made. Therefore only the normally ordered

polarization (Eq.A.10) and Langevin (Eq.A.12) operators contribute. As is discussed in

Appendix D, this is the condition that minimizes the quantum noise emitted from a

Raman amplifier.

APPENDIX B
STOKES FIELD OPERATOR

The Stokes field operator is presented in detail in this Appendix. Since this operator is used in Appendix D where there is more than one Raman amplifier, subscripts are included to distinguish between various amplifiers.

The full expression for the Stokes field operator is:^{2,3}

$$\begin{aligned} \hat{E}_\alpha^{(-)}(z, \tau) = & \hat{E}_\alpha^{(-)}(0, \tau) + \int_0^z dz' \hat{Q}_\alpha^\dagger(z', 0) A_\alpha(z, z', \tau) \\ & + \int_0^\tau d\tau' \hat{E}_\alpha^{(-)}(0, \tau') B_\alpha(z, \tau, \tau') \\ & + \int_0^\tau \int_0^z dz' d\tau' \hat{F}_\alpha^\dagger(z', \tau') C_\alpha(z, z', \tau, \tau') \end{aligned} \quad (\text{B.1})$$

where $\hat{E}_\alpha^{(-)}(z, \tau)$ is the slowly varying, negative frequency Stokes field operator at position z in the Raman cell, $\hat{E}_\alpha^{(-)}(0, \tau)$ is the input slowly varying, negative frequency Stokes field operator to the Raman cell, $\hat{Q}_\alpha^\dagger(z', 0)$ is the initial atomic polarization operator at position z' (see Appendix A), and $\hat{F}_\alpha^\dagger(z', \tau')$ is a quantum Langevin operator needed to maintain operator consistency when collisional dephasing is included (see Appendix A). The kernels in Eq.(B.1) are^{2,3}:

$$A_\alpha(z, z', \tau) = -i\kappa_2 E_{p\alpha}(\tau) e^{-\Gamma\tau} I_0[[4\kappa_1\kappa_2(z-z')p_\alpha(\tau)]^{1/2}] \quad (\text{B.2})$$

$$B_\alpha(z, \tau, \tau') = (\kappa_1\kappa_2 z)^{1/2} E_{p\alpha}(\tau) e^{-\Gamma(\tau-\tau')} E_{p\alpha}^*(\tau') \frac{I_1[[4\kappa_1\kappa_2 z(p_\alpha(\tau) - p_\alpha(\tau'))]^{1/2}]}{[p_\alpha(\tau) - p_\alpha(\tau')]^{1/2}} \quad (\text{B.3})$$

$$C_{\alpha}(z, z', \tau, \tau') = -i\kappa_2 E_{p\alpha}(\tau) e^{-\Gamma(\tau-\tau')} I_0[[4\kappa_1 \kappa_2 (z - z')(p_{\alpha}(\tau) - p_{\alpha}(\tau'))]^{1/2}] \quad (B.4)$$

Here κ_1 and κ_2 are constants that depend on the strengths of the transitions⁵⁰, Γ is the Raman medium dephasing rate, z is the coordinate labeling the position along the pumped, pencil shaped region in the Raman medium, τ is the retarded time ($\tau=t-z/c$), and I_0 and I_1 are modified Bessel functions. $E_{p\alpha}(\tau)$ is the classically treated pump field

(assumed to be large) and $p_{\alpha}(\tau)$ is defined by: $p_{\alpha}(\tau) \equiv \int_0^{\tau} |E_{p\alpha}(\tau'')|^2 d\tau''$.

In our analysis we normally order the field operators to calculate the expectation value of the intensity pattern. With this ordering, the only non-zero noise operator correlation functions are (see Appendix A):

$$\langle \hat{Q}_{\alpha}^{\dagger}(z', 0) \hat{Q}_{\beta}(z'', 0) \rangle = [AN]^{-1} \delta(z' - z'') \delta_{\alpha\beta} \quad (B.5)$$

$$\langle \hat{F}_{\alpha}^{\dagger}(z', \tau') \hat{F}_{\beta}(z'', \tau'') \rangle = \frac{2\Gamma}{AN} \delta(z' - z'') \delta(\tau' - \tau'') \delta_{\alpha\beta} \quad (B.6)$$

Here A is the cross sectional area of the pencil shaped region pumped in the Raman cell and N is the density of molecules in the Raman medium. Equation (B.5) states that the initial polarization fluctuations at one location in the Raman medium are uncorrelated with the polarization fluctuations at another location. Similarly, Eq.(B.6) states that the quantum statistical collisional effects in the medium are uncorrelated position to position

and time to time. The Kroneker delta functions simply indicate that these fluctuations are also uncorrelated between different Raman amplifiers, as one would expect.

The anti-normally ordered correlation functions of the operators governing the growth of the noise are (see Appendix A):

$$\langle \hat{Q}_\alpha(z,0) \hat{Q}_\beta^\dagger(z',0) \rangle = 0 \quad (\text{B.7})$$

$$\langle \hat{F}_\alpha(z',\tau') \hat{F}_\beta^\dagger(z'',\tau'') \rangle = 0 \quad (\text{B.8})$$

These results assume that the Raman medium remains primarily in its ground state throughout the interaction with the radiation fields.

This notation is somewhat different from that used in Appendix D. The second and fourth terms on the right hand side of Eq.(B.1) do not depend on the input Stokes field, so they

represent the noise operator $\hat{N}_\alpha^\dagger(z,\tau)$ in Eq.(D.1) of Appendix D. One can see that

Eqs.(B.7,B.8) lead to the result Eq.(D.2) that the expectation value of the anti-normally ordered noise operator given in Eq.(D.1) is 0.

APPENDIX C

VISIBILITY CALCULATIONS USING IDEAL AMPLIFIERS

In Chapter 5 a simple model for a single mode amplifier was introduced in Eq.(5.2). In the first part of this Appendix we deduce the condition required for the amplifier to emit the least noise consistent with quantum mechanics. Once this is done, the model is applied to the Mach-Zender interferometer shown in Fig.21. Predictions for the output visibility as a function of the input signal are quite easily obtained.

The annihilation operator for the ideal, single mode amplifier's output mode $\hat{b}_\alpha$ is linearly related to the input annihilation operator $\hat{a}_\alpha$:

$$\hat{b}_\alpha = M_\alpha \hat{a}_\alpha + \hat{N}_\alpha \quad (\text{C.1})$$

where M_α is related to the gain of the amplifier, and $\hat{N}_\alpha$ is a noise operator that depends only on the internal modes of the amplifier.

As discussed in Chapter 5, the field operators must satisfy the Bose-Einstein commutation relation. One can quickly see that without the noise operator, Eq.(C.1) could not satisfy the commutation relation except for in the uninteresting case of no gain ($|M_\alpha| = 1$). To maintain the commutation relations the noise operator must satisfy:

$$[\hat{N}_\alpha, \hat{N}_\alpha^\dagger] = (1 - |M_\alpha|^2) . \quad (C.2)$$

Equation (C.2) provides one condition on the noise operator but does not distinguish between low noise and high noise amplifiers. To see what condition leads to, for example, a low noise amplifier, we calculate the expectation value of the number of photons emitted from one of these amplifiers:

$$\langle \hat{b}_\alpha^\dagger \hat{b}_\alpha \rangle = |M_\alpha|^2 \langle \hat{a}_\alpha^\dagger \hat{a}_\alpha \rangle + \langle \hat{N}_\alpha^\dagger \hat{N}_\alpha \rangle . \quad (C.3)$$

The first term on the right hand side of Eq.(C.3) is interpreted as the amplified signal; the second is the number of amplified spontaneous emission photons, or noise.⁴⁵ Rearranging Eq.(C.2) and inserting it into Eq.(C.3), one obtains:

$$\langle \hat{b}_\alpha^\dagger \hat{b}_\alpha \rangle = |M_\alpha|^2 \langle \hat{a}_\alpha^\dagger \hat{a}_\alpha \rangle + \langle \hat{N}_\alpha^\dagger \hat{N}_\alpha \rangle + |M_\alpha|^2 - 1 . \quad (C.4)$$

The last three terms in Eq.(C.4) correspond to the number of noise photons emitted by the amplifier. To achieve low noise, we wish to find the condition necessary for this amplifier to emit the minimum number of noise photons allowed by quantum mechanics.

Noting that $\langle \hat{N}_\alpha^\dagger \hat{N}_\alpha \rangle \geq 0$, we see that the condition for the minimum number of noise

photons is:

$$\langle \hat{N}_\alpha^\dagger \hat{N}_\alpha \rangle = 0 . \quad (C.5)$$

When this condition is satisfied and the amplifier is operated in the high gain regime ($|M_\alpha|^2 \gg 1$), one can see from Eq.(C.4) that the average number of output noise photons is equal to the average number of output signal photons when the input signal has an average of 1 input photon. This is why the noise is said to be equivalent to at least 1 photon per mode at the input to a noiseless amplifier.

No specific operating mechanism has been indicated here to satisfy the minimum noise condition of Eq.(C.5), so from this analysis it is unclear exactly what operating conditions in the amplifier are required to achieve the minimum number of noise photons. However, in Appendix D, which discusses Raman scattering we will observe that the analogous condition arises and can be traced to specific operating conditions.

Now, with this model of an ideal, single mode amplifier, we calculate the fringe visibility of the interferometer diagramed in Fig.21. To do this, we start with the annihilation operator of the output field of the generator, then follow the transformation of the operator through the interferometer to the output screen.

The output field from the generator is described by the annihilation operator $\hat{b}_G$.

This field is split by the input beam splitter to the interferometer. We will assume the reflection and transmission coefficients are constant over the bandwidth of the mode. Then the input field operator to the upper (lower) leg of the interferometer is written

$t_i \hat{b}_G$ ($r_i \hat{b}_G$) where t_i (r_i) is the transmission (reflection) coefficient of the input beam

splitter.⁵¹ (Since we will employ normal ordering of the field operators to calculate the intensity, the vacuum field contribution from the other port of the beam splitter does not contribute and can be ignored.)

Using Eq.(C.1), the output field operator from the upper amplifier is:

$$\hat{b}_I = M_I(t_i \hat{b}_G) + \hat{N}_I \quad (\text{C.6})$$

and the output field from the lower amplifier is found by replacing the subscript I with II and inserting r_i for t_i . The final output field operator ($\hat{b}_{Tot}(x)$) from the interferometer can be written:

$$\hat{b}_{Tot}(x) = r_o \hat{b}_I + t_o \hat{b}_{II} e^{i\phi(x)} \quad (\text{C.7})$$

where x is a transverse coordinate of the field and t_o and r_o are the transmission and reflection coefficients of the output beam splitter respectively. The phase factor $\phi(x)$ results because a small angle was placed between the combined fields at the output beam splitter.

The intensity pattern is calculated using the quantum mechanical expectation value for the normally ordered product:

$$\begin{aligned} \langle \hat{I}(x) \rangle &= K \langle \hat{b}_{Tot}^\dagger(x) \hat{b}_{Tot}(x) \rangle \\ &= K [|r_o|^2 \langle \hat{b}_I^\dagger \hat{b}_I \rangle + |t_o|^2 \langle \hat{b}_{II}^\dagger \hat{b}_{II} \rangle + r_o^* t_o \langle \hat{b}_I^\dagger \hat{b}_{II} \rangle e^{i\phi(x)} + r_o^* t_o \langle \hat{b}_{II}^\dagger \hat{b}_I \rangle e^{-i\phi(x)}] \end{aligned} \quad (\text{C.8})$$

where K keeps the units correct and may include temporal information about the pulse but is unimportant for the discussion at hand. We assume good spatial overlap of the fields from each leg of the interferometer. In Eq.(C.8) one sees that replacing the complex transmission and reflection coefficients with their absolute values shifts the positions of the maximum and minimum intensity profiles along the x coordinate. Since the location of the maximum and minimum intensity profiles is unimportant in the visibility calculation, we make this simplification without affecting the results of our calculations. The expectation value for the maximum intensity is found by choosing the position where the phase is such that constructive interference results. Similarly, the minimum intensity is found by choosing the position where destructive interference occurs.

$$\langle \hat{I} \rangle_{Max} = K [r_o^2 \langle \hat{b}_I^\dagger \hat{b}_I \rangle + t_o^2 \langle \hat{b}_{II}^\dagger \hat{b}_{II} \rangle + r_o t_o \langle \hat{b}_I^\dagger \hat{b}_{II} \rangle + r_o t_o \langle \hat{b}_{II}^\dagger \hat{b}_I \rangle] \quad (C.9a)$$

$$\langle \hat{I} \rangle_{Min} = K [r_o^2 \langle \hat{b}_I^\dagger \hat{b}_I \rangle + t_o^2 \langle \hat{b}_{II}^\dagger \hat{b}_{II} \rangle - r_o t_o \langle \hat{b}_I^\dagger \hat{b}_{II} \rangle - r_o t_o \langle \hat{b}_{II}^\dagger \hat{b}_I \rangle] \quad (C.9b)$$

The visibility is then calculated to be:

$$V = \frac{r_o t_o (\langle \hat{b}_I^\dagger \hat{b}_{II} \rangle + \langle \hat{b}_{II}^\dagger \hat{b}_I \rangle)}{r_o^2 \langle \hat{b}_I^\dagger \hat{b}_I \rangle + t_o^2 \langle \hat{b}_{II}^\dagger \hat{b}_{II} \rangle} \quad (C.10)$$

The visibility is large when the cross correlation between the two amplified fields is large,

and is small when the cross correlation is small relative to the correlation of the output fields of the amplifiers with themselves. This is what one expects since only the input signal will sample both legs of the interferometer and can contribute to the cross correlation between the two amplified fields.

To see the effects of the amplifier noise, we substitute Eq.(C.6) into Eq.(C.10). Using the fact that the noise operator is independent of the input field mode and has random phase, the following form for the visibility is found:

$$V = \frac{r_o t_o r_i t_i (M_I^* M_{II} + M_{II}^* M_I) \langle \hat{b}_G^\dagger \hat{b}_G \rangle}{(r_o^2 t_i^2 |M_I|^2 + r_i^2 t_o^2 |M_{II}|^2) \langle \hat{b}_G^\dagger \hat{b}_G \rangle + r_o^2 \langle \hat{N}_I^\dagger \hat{N}_I \rangle + t_o^2 \langle \hat{N}_{II}^\dagger \hat{N}_{II} \rangle} \quad (C.11)$$

Note that noise terms do not appear in the numerator since terms such as $\langle \hat{N}_I^\dagger \hat{N}_{II} \rangle$ are zero. The noise terms are present only in the denominator where, as one would expect, they diminish the output visibility. The term $\langle \hat{b}_G^\dagger \hat{b}_G \rangle$ is simply the expectation value of the number of input photons to the interferometer. In Eq.(C.11) one can see that if zero photons are input into the interferometer the visibility vanishes, again the result one would expect. The visibility increases as one increases the number of photons input to the interferometer because there is more signal to compete with the noise.

APPENDIX D

VISIBILITY CALCULATIONS USING RAMAN AMPLIFIERS

In this Appendix we replace the ideal, single mode, linear amplifiers in Fig.21 with Raman amplifiers and repeat the analysis done in Appendix C. This analysis will show that the procedure used in Appendix C works on a real system and, in fact, is essentially the same. To treat this problem we employ the fully quantum mechanical model of Raman scattering.¹⁻³ This plane wave model assumes a pencil shaped region of Raman active medium is pumped with a large, classical field. Additionally, the pump laser is assumed to remain undepleted as it passes through the Raman medium. The slowly varying, negative frequency Stokes field operator is written:

$$\hat{E}_{\alpha}^{(-)}(z, \tau) = \hat{E}_{\alpha}^{(-)}(0, \tau) + \int_0^{\tau} d\tau' \hat{E}_{\alpha}^{(-)}(0, \tau') B_{\alpha}(z, \tau, \tau') + \hat{N}_{\alpha}^{\dagger}(z, \tau) \quad (\text{D.1})$$

where $\hat{E}_{\alpha}^{(-)}(z, \tau)$ is the field operator at position z in the Raman cell and at retarded time τ , $\hat{E}_{\alpha}^{(-)}(0, \tau)$ is the input Stokes field operator to the Raman cell, and $\hat{N}_{\alpha}^{\dagger}(z, \tau)$ is the noise operator that is independent of the input field operator. The exact expressions for the noise operator $\hat{N}_{\alpha}^{\dagger}(z, \tau)$ and the kernel $B_{\alpha}(z, \tau, \tau')$ are not important for the immediate discussion, however they are given in Appendix B and discussed more completely in Chapter 2. As in the example with ideal, single mode amplifiers, the subscript α can take the values *I*, *II*, and *G* to designate amplifier I in the interferometer, amplifier II, and the generator respectively.

Before proceeding we pause to note the similarities of Eq.(D.1) for the Raman system and Eq.(C.1) for the ideal, single mode amplifier discussed above. The output field operator $\hat{E}_\alpha^{(-)}(z,\tau)$ is analogous to $\hat{b}_\alpha^\dagger$. Similarly, $\hat{E}_\alpha^{(-)}(0,\tau)$, the Stokes field at $z=0$, is analogous to $\hat{a}_\alpha^\dagger$ as the input field operator. There is also a clear analogy between the noise operators for the ideal, single mode amplifier in Eq.(C.1), $\hat{N}_\alpha^\dagger$, and the noise operator in the Stokes field in Eq.(D.1), $\hat{N}_\alpha^\dagger(z,\tau)$. Also, like the ideal, single mode amplifier, the Raman Stokes field equation satisfies bosonic commutation relations.²⁵ So, like the ideal, single mode amplifier, the quantum noise added by the Raman amplifier is required by the commutation relations and the Raman amplifier can be thought of as a quantum limited amplifier. With these similarities it should not be surprising to find that the analysis using Raman amplifiers closely follows the procedures in Appendix C.

It is interesting to note that the anti-normally ordered noise operator correlation function is:

$$\langle \hat{N}_\alpha(z,\tau) \hat{N}_\beta^\dagger(z',\tau') \rangle = 0 \quad (D.2)$$

for all values of the variables (see Appendix A). For the ideal, single mode amplifier we found that this was the condition that led to the minimum number of noise photons; thus one would also expect that the Raman amplifier would emit the minimum number of noise photons.

With the Raman system, however, we know the operating conditions of the amplifier and can gain an understanding of why the Raman amplifier should emit the minimum number of noise photons. The result of Eq.(D.2) is dependent on the condition that the Raman system remains primarily in its ground state (see Appendix A). A molecule that participates in Raman scattering is initially in its ground state and, after the two photon Raman scattering process, it is left in an excited state. Thus, the condition that the Raman system remains primarily in its ground state means that very few molecules of the active medium are in the final state after the emission of the Stokes photon.

One can get a physical understanding of why the noise is low in an amplifier that has nearly all of its molecules in the state that is "ready" to emit photons rather than in the final state the molecules are in after emission of a photon by considering a two level system that has population in both states. In a two level system the upper state is "ready" to emit a photon and the ground state is the final state after emission of a photon. With more molecules in the upper state than in the lower state, an input signal photon at the transition frequency is more likely to amplify via stimulated emission than to be absorbed, a necessary condition for amplification. However, any signal photons that are absorbed by molecules in the lower state are potentially lost from the signal. Therefore, one would expect that the fewer molecules in the lower state (i.e. molecules that are available to absorb the photons of interest), the less noise the amplifier will emit. One then expects that the lowest noise characteristics will be achieved when the system is completely inverted so no molecules are in the final state of the amplifier transition.⁵²

Since the Raman amplifier we consider is assumed to have insignificantly few molecules in the final state of the amplifier transition, one expects that the minimum number of noise photons are emitted.

Now, with this model of stimulated Raman scattering, we proceed to calculate the expected fringe visibility for the apparatus shown in Fig.21 following a routine very similar to the one used in Appendix C. Note that it is assumed that the input field to the Raman generator is the vacuum field, which does not contribute when the field operators are normally ordered. Similarly, the vacuum field input at the beam splitter does not contribute in normal ordering, so it will be ignored.

The output field of the Raman generator is written $\hat{E}_G^{(-)}(L_G, \tau)$ where L_G is the length of the Raman generator. Assuming that the beam splitter coefficients are constant over the Raman linewidth, the input field to the upper (lower) amplifier in Fig.21 is

$t_i \hat{E}_G^{(-)}(L_G, \tau)$ ($r_i \hat{E}_G^{(-)}(L_G, \tau)$) where t_i (r_i) is the transmission (reflection) coefficient of the input beam splitter. Using Eq.(D.1), the output field from the upper amplifier is:

$$\hat{E}_I^{(-)}(L, \tau) = t_i \hat{E}_G^{(-)}(L_G, \tau) + \int_0^\tau d\tau' t_i \hat{E}_G^{(-)}(L_G, \tau') B_I(L, \tau, \tau') + \hat{N}_I^\dagger(z, \tau) \quad (D.3)$$

where L is the length of the Raman amplifiers (assumed to be identical in length). The output field from the lower amplifier in Fig.21 is the same as Eq.(D.3) with I 's changed to II 's and t_i 's changed to r_i 's.

The output fields from the two amplifiers combine at the exit beam splitter to give:

$$\hat{E}_{Tor}^{(-)}(x, \tau) = r_o \hat{E}_I^{(-)}(L, \tau) + t_o \hat{E}_{II}^{(-)}(L, \tau) e^{i\phi(x)} \quad (D.4)$$

Here $t_o(r_o)$ is the transmission (reflection) coefficient of the exit beam splitter of the interferometer. As in the analysis above for the single mode amplifier, the output field is now a function of the transverse coordinate x because the beams have been combined with a slight angle between them.

The intensity pattern in units of photons per second emitted from the pumped pencil shaped region of Raman medium can now be calculated using the normally ordered product:

$$\begin{aligned} \langle \hat{I}(x, \tau) \rangle &= \frac{Ac}{2\pi\hbar\omega_s} \langle \hat{E}_{Tor}^{(-)}(x, \tau) \hat{E}_{Tor}^{(+)}(x, \tau) \rangle \\ &= \frac{Ac}{2\pi\hbar\omega_s} [|r_o|^2 \langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle + |t_o|^2 \langle \hat{E}_{II}^{(-)}(L, \tau) \hat{E}_{II}^{(+)}(L, \tau) \rangle \\ &\quad + r_o^* t_o \langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_{II}^{(+)}(L, \tau) \rangle e^{-i\phi(x)} + r_o t_o^* \langle \hat{E}_{II}^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle e^{i\phi(x)}] \end{aligned} \quad (D.5)$$

where c is the speed of light and $\hbar\omega_s$ is the energy of a Stokes photon. Since replacing the complex transmission and reflection coefficients with their absolute values leads only to an unimportant shift of the fringe pattern along the x coordinate, we make this simplification. The quantum mechanical expectation values for the maximum and minimum intensities can now be found.

$$\begin{aligned} \langle \hat{I}(\tau) \rangle_{Max} &= \frac{Ac}{2\pi\hbar\omega_s} [r_o^2 \langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle + t_o^2 \langle \hat{E}_{II}^{(-)}(L, \tau) \hat{E}_{II}^{(+)}(L, \tau) \rangle \\ &\quad + r_o t_o \langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_{II}^{(+)}(L, \tau) \rangle + t_o r_o \langle \hat{E}_{II}^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle] \end{aligned} \quad (D.6a)$$

$$\begin{aligned} \langle \hat{f}(\tau) \rangle_{Min} = & \frac{Ac}{2\pi\hbar\omega_s} [r_o^2 \langle \hat{E}_I^{(-)}(L,\tau) \hat{E}_I^{(+)}(L,\tau) \rangle + t_o^2 \langle \hat{E}_H^{(-)}(L,\tau) \hat{E}_H^{(+)}(L,\tau) \rangle \\ & - r_o t_o \langle \hat{E}_I^{(-)}(L,\tau) \hat{E}_H^{(+)}(L,\tau) \rangle - t_o r_o \langle \hat{E}_H^{(-)}(L,\tau) \hat{E}_I^{(+)}(L,\tau) \rangle] \end{aligned} \quad (D.6b)$$

Note the similarity between Eqs.(C.9) and Eqs.(D.6) in the ideal, linear amplifier section.

For our experiment, the fringe pattern from the interferometer was imaged onto a linear diode array. The diodes in the array integrated the intensity over the duration of the pulse. Therefore, to obtain theoretical predictions appropriate for the experiment, Eqs.(D.6) were integrated over the pulse. Consequently, the expression for the visibility V_{Pulse} was found to be:

$$V_{Pulse} = \frac{\int_{Pulse} \langle \hat{f}(\tau) \rangle_{Max} d\tau - \int_{Pulse} \langle \hat{f}(\tau) \rangle_{Min} d\tau}{\int_{Pulse} \langle \hat{f}(\tau) \rangle_{Max} d\tau + \int_{Pulse} \langle \hat{f}(\tau) \rangle_{Min} d\tau} \quad (D.7)$$

Note that Eq.(D.7) for the visibility is similar to Eq.(5.1) except that the effects of the pulse duration are now included.

Thus using the "real" Raman amplifiers in the experimental setup diagramed in Fig.21 is very similar to using ideal, single-mode amplifiers. To find quantitative values for the visibility, numerous integrals had to be evaluated. Additionally, we were particularly interested in the slightly different experimental setup shown in Fig.23. The only changes that were needed to calculate the expected visibility from the interferometer shown in Fig.23 were the inclusion of more beam splitters, which was straightforward and is discussed below.

Since the interferometer shown in Fig.23 is nearly cyclic, both beam paths experience most of the optics in the interferometer, which makes the interferometer fairly insensitive to vibrations and thermal variations. Additional beam splitters, which we refer to as "pick-offs," allowed easy beam manipulation, but also led to a loss of signal input to the amplifiers that needed to be accounted for. Another loss of signal that needed to be included in the formalism came from the optics necessary for beam alignment between the generator and the interferometer. The loss from these optics was treated as an attenuation beam splitter between the generator and the interferometer. Including the losses from the beam splitters into the formalism for calculating the total output field from the interferometer gives:

$$\hat{E}_{Tot}^{(-)}(x, \tau) = t_o r_{p1} \hat{E}_I^{(-)}(L, \tau) + r_o r_{p2} \hat{E}_{II}^{(-)}(L, \tau) e^{i\phi(x)} \quad (D.8)$$

where r_{p1} (r_{p2}) is the reflection coefficient of the pickoff optic in front of the upper (lower) amplifier in Fig.23 . Similarly, the input field to the upper (lower) amplifier is found to be $t_A r_{p1} \hat{E}_G^{(-)}(L_G, \tau)$ ($t_A t_{p2} \hat{E}_G^{(-)}(L_G, \tau)$) where t_{p1} (t_{p2}) is the transmission coefficient of the pickoff optic in front of the upper (lower) amplifier and t_A is the transmission coefficient that accounts for the loss between the generator and the interferometer. All the beam splitter coefficients are assumed to be constant across the Raman linewidth. Thus the inclusion of the extra optics affects the output fields only by scale factors.

Using normal ordering, the visibility was derived as indicated above to get:

$$V_{Pulse} = \frac{r_{p2}^2 r_{p1}^2 r_o^2 t_o^2 \left(\int_{Pulse} \langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_H^{(+)}(L, \tau) \rangle d\tau + \int_{Pulse} \langle \hat{E}_H^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle d\tau \right)}{r_{p2}^2 r_o^2 \int_{Pulse} \langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle d\tau + r_{p1}^2 t_o^2 \int_{Pulse} \langle \hat{E}_H^{(-)}(L, \tau) \hat{E}_H^{(+)}(L, \tau) \rangle d\tau} \quad (D.9)$$

Inserting the full expressions for the amplified and generator fields as given in Eq.(B.1) of Appendix B into Eq.(D.9) leads to a fairly long expression for the visibility. This expression can be written in terms of the following integrals:

$$\mathfrak{S}_1(z, E_{p\alpha}, \tau) = \int_0^z dz' [NA]^{-1} |A_\alpha(z, z', \tau)|^2 \quad (D.10)$$

$$\mathfrak{S}_2(z, E_{p\alpha}, \tau) = \int_0^z \int_0^\tau dz' d\tau' \frac{2\Gamma}{NA} |C_\alpha(z, z', \tau, \tau')|^2 \quad (D.11)$$

$$\mathfrak{S}_3(L_G, L, \tau, E_{pG}, E_{p\alpha}) = \int_0^\tau d\tau' \int_0^{L_G} dz' [NA]^{-1} A_G(L_G, z', \tau') A_G^*(L_G, z', \tau) B_\alpha(L, \tau, \tau') \quad (D.12)$$

$$\begin{aligned} & \mathfrak{S}_4(L_G, L, \tau, E_{pG}, E_{p\alpha}) \\ &= \int_0^\tau d\tau' \int_0^{L_G} dz' \int_0^\tau d\tau'' \frac{2\Gamma}{[NA]} C_G(L_G, z', \tau, \tau'') C_G^*(L_G, z', \tau, \tau') B_\alpha(L, \tau, \tau') \end{aligned} \quad (D.13)$$

$$\begin{aligned} & \mathfrak{S}_5(L_G, L, \tau, E_{pG}, E_{p\alpha}, E_{p\beta}) \\ &= \int_0^\tau d\tau' \int_0^\tau d\tau'' \int_0^{L_G} dz' [NA]^{-1} A_G(L_G, z', \tau') A_G^*(L_G, z', \tau'') B_\alpha(L, \tau, \tau') B_\beta(L, \tau, \tau'') \end{aligned} \quad (D.14)$$

$$\begin{aligned}
& \mathfrak{S}_6(L_G, L, \tau, E_{pG}, E_{p\alpha}, E_{p\beta}) \\
& = \int_0^\tau d\tau' \int_0^\tau d\tau'' \int_0^{L_0} dz' \int_0^\tau d\tau^o \frac{2\Gamma}{NA} C_G(L_G, z', \tau', \tau^o) C_G^*(L_G, z', \tau'', \tau^o) \\
& \quad \Theta(\tau'' - \tau^o) B_\alpha(L, \tau, \tau') B_\beta^*(L, \tau, \tau'')
\end{aligned} \tag{D.15}$$

where $\Theta(\tau'' - \tau^o)$ indicates a step function. Note that in these integrals the dependence on the pump field amplitude has been included. (See Eqs.(B.2-B.4) for the dependence on the pump field amplitude.) The correlation function between the two output fields of the amplifiers that appears in the numerator of Eq.(D.9) can be written:

$$\begin{aligned}
\langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle &= t_i r_i t_{pI} t_{p2} [\mathfrak{S}_1(L_G, E_{pG}, \tau) + \mathfrak{S}_2(L_G, E_{pG}, \tau) \\
& \quad + \mathfrak{S}_3(L_G, L, \tau, E_{pG}, E_{pI}) + \mathfrak{S}_4(L_G, L, \tau, E_{pG}, E_{pI}) \\
& \quad + \mathfrak{S}_5(L_G, L, \tau, E_{pG}, E_{pII}) + \mathfrak{S}_6(L_G, L, \tau, E_{pG}, E_{pII}) \\
& \quad + \mathfrak{S}_5(L_G, L, \tau, E_{pG}, E_{pI}, E_{pII}) + \mathfrak{S}_6(L_G, L, \tau, E_{pG}, E_{pI}, E_{pII})]
\end{aligned} \tag{D.16}$$

Similarly, the autocorrelation function of the amplified field from the upper amplifier in Fig.23 is:

$$\begin{aligned}
\langle \hat{E}_I^{(-)}(L, \tau) \hat{E}_I^{(+)}(L, \tau) \rangle &= t_i^2 t_{pI}^2 [\mathfrak{S}_1(L_G, E_{pG}, \tau) + \mathfrak{S}_2(L_G, E_{pG}, \tau) \\
& \quad + 2\mathfrak{S}_3(L_G, L, \tau, E_{pG}, E_{pI}) + 2\mathfrak{S}_4(L_G, L, \tau, E_{pG}, E_{pI}) \\
& \quad + \mathfrak{S}_5(L_G, L, \tau, E_{pG}, E_{pI}, E_{pI}) + \mathfrak{S}_6(L_G, L, \tau, E_{pG}, E_{pI}, E_{pI}) \\
& \quad + \mathfrak{S}_1(L, E_{pI}, \tau) + \mathfrak{S}_2(L, E_{pI}, \tau)]
\end{aligned} \tag{D.17}$$

To obtain the autocorrelation function of the amplified field from the lower amplifier, simply replace the subscripts *I* with *II* and put r_i in place of t_i . One can see that the expression in Eq.(D.16) is nearly identical to Eq.(D.17), except for the last two terms in

Eq.(D.16). These terms represent the amplified spontaneous emission (noise) in the amplifier.

Thus, the calculation of the visibility is reduced to the evaluation of some rather long integrals. The integrations over the spatial coordinate can be done analytically,⁵³ but numerical techniques are required for the temporal integrations.

To obtain quantitative predictions using the above formulas, the beam splitter coefficients given in Appendix E along with the parameters listed below are needed. The effective cross sectional area pumped by the pump laser was calculated to be 4.25×10^{-3} cm² and the effective length of the Raman generator (amplifiers) was found to be 1665cm (1860cm).²⁴ To model the high gain in the amplifiers in the interferometer, the pump energy input to the amplifiers was taken to be approximately .37 mJ. The plane wave gain coefficient⁵⁴ ($\alpha = 2.5 \times 10^{-9}$ cm/W) was used to calculate⁵⁰ the coupling constants. There were no free parameters employed in these calculations.

APPENDIX E
EXPERIMENTAL DETAILS OF INTERFEROMETER EXPERIMENT

The detailed description of the experiment diagramed in Fig.23 is presented here in Appendix E. As noted in Chapter 5, the apparatus consists essentially of a generator and an interferometer with amplifiers in each leg.

A pulsed, near single-mode, frequency doubled, Nd:YAG laser (532 nm) with a gaussian temporal profile, 21.6 ns full width at half maximum, was used to pump the Raman amplifiers. The Raman scattering was done in 10 atmospheres of H_2 on the vibrational $Q_{01}(1)$ transition at room temperature. For this system the dephasing time for the Raman medium was $1/\Gamma = 7$ ns,⁵⁴ considerably shorter than the pump duration which meant that collisional effects were important.

An etalon was used to monitor the pump laser stability to insure that no mode hops occurred during data collection. Additionally, an energy meter that was interfaced to a computer (not shown in Fig.23), provided energy measurements of every pump laser shot. The output Stokes signal that was input to the interferometer was varied by placing pump attenuators in front of the Raman generator (not shown in Fig.23).

After the pulse exited the generator the residual pump was removed from the beam with a modified Pellin-Broca prism, and then additional pump energy needed for the amplification in the interferometer was added to the beam. No temporal delay was introduced between the Stokes pulse and the new pump pulse. The optics necessary for beam manipulation between the generator and the interferometer transmitted approximately 68 percent of the Stokes signal.

To achieve equal gain in the two amplifiers, the input beam splitter to the interferometer had a 50/50 transmission/reflection ratio at the pump wavelength to within a percent. The pump intensity input to the amplifiers was always large enough to achieve high gain (the signal was amplified by approximately 10 orders of magnitude for small input signals), but not so large as to lead to pump depletion.

Both the input and exit beam splitters of the interferometer had a 78/22 transmission/reflection ratio at the Stokes wavelength (680 nm). For the total output field of the interferometer to obtain equal contributions of amplified signal from each leg, the Stokes field that was transmitted (reflected) as it entered the interferometer was reflected (transmitted) at the exit beam splitter.

In the near cyclic design for the interferometer shown in Fig.23, most of the optics in the interferometer were common to both beam paths. Consequently, slight shifts in the optics positions affected both beam paths identically, and therefore did not affect the relative phase between the two beam. The cyclic design greatly increased the stability of the interferometer, which was critical since the beams traversed approximately 70 m in each leg of the interferometer. One beam path is shown as a dashed line in Fig.23 to differentiate it from the other. Note that each beam was amplified in a separate Raman amplifier placed in a multipass cell. The multipass cells limit the amplification to a single spatial mode. After the beam was amplified it encountered a modified Pellin-Broca prism before entering a second amplifier. This removed the residual pump energy from the pulse so no additional amplification occurred in the second amplifier. To insure that the amplification of the two pulses was independent, the beam path between the two

amplifiers was made long enough that the two pulses never overlapped inside an amplifier. The residual pump beams removed from the beam by the modified Pellin-Broca prisms were monitored with fast photo diodes (not shown in Fig.23) to make certain that no pump depletion occurred during amplification.

For additional beam manipulation at the expense of only a slight loss of stability, identical "pickoff" beam splitters were used. These optics had transmission/reflection coefficients of 50/50 at the Stokes wavelength and mark the ends of the cyclic region of the interferometer in Fig.23.

The output fringe patterns were imaged onto a linear photo-diode array with 25 μm resolution that was interfaced to a computer, allowing the fringe pattern from every shot to be recorded. Ensembles of these fringe patterns could then be formed and the resulting visibility measured.

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