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

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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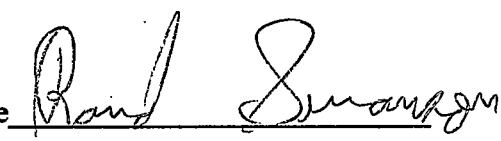
Signature 

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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.

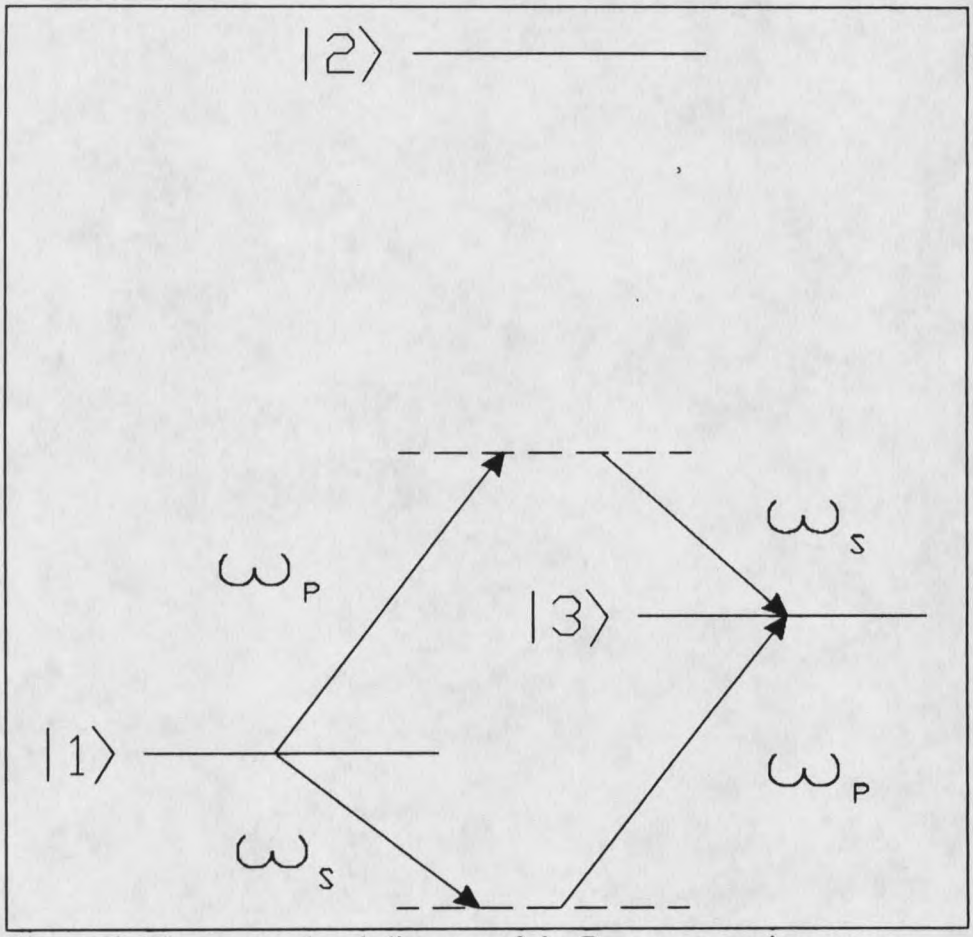

Figure 1 The energy level diagram of the Raman scattering process.

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 repletion 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 introduce 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 Schodinger'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 model[4] (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:

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

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

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

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:

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

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]

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

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]

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

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 phenominologically (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 $\langle \hat{Q}^{\dagger}(z',0)\hat{E}_s^{(+)}(0,\tau'')\rangle$ 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 1988[5] 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]

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

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 experiment[5-7] 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.

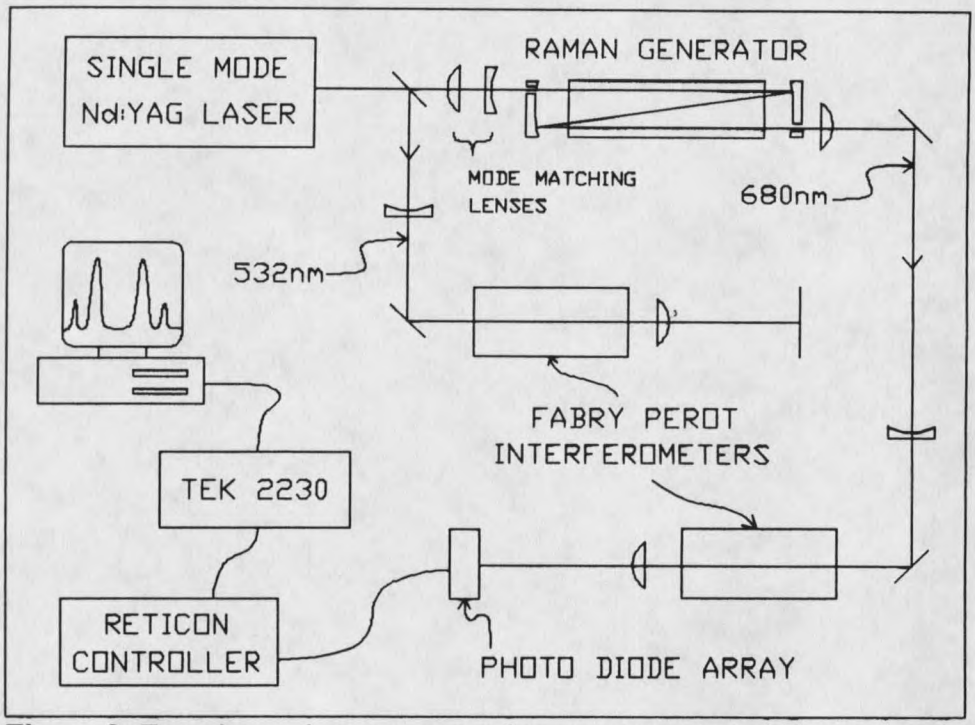

Figure 2 Experimental apparatus used to measure spectral fluctuations in Raman scattering.

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:

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

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

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

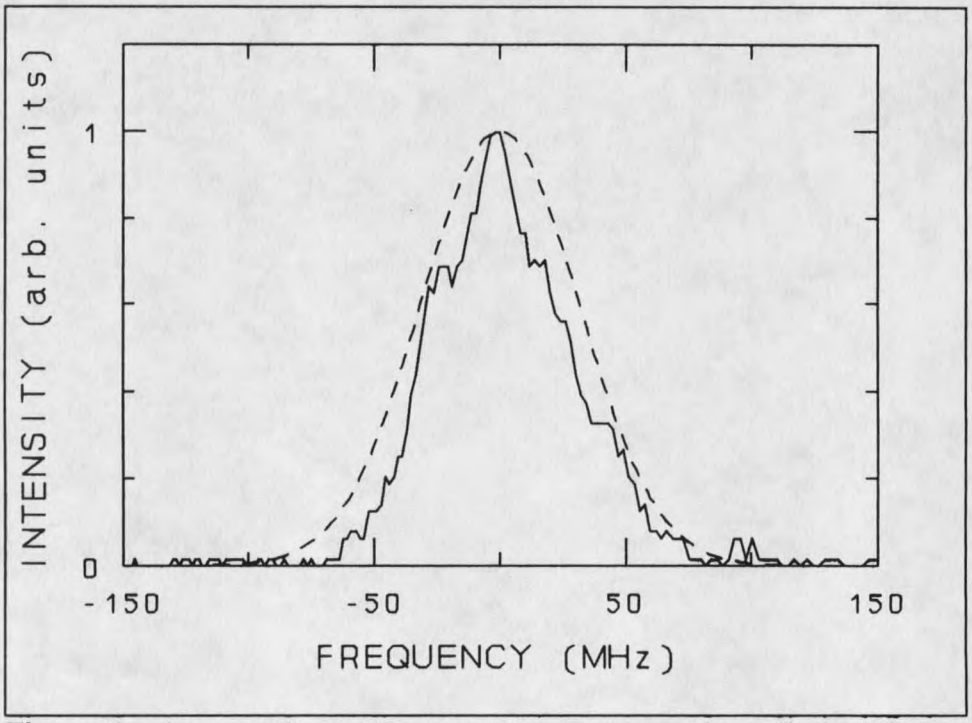

Figure 3 An experimentally measured, near transform limited Stokes spectrum.