Gain-guiding effects in stimulated Raman scattering
by Philip Ross Battle

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

Abstract:
Measurements of the output of a Raman amplifier when there is no input Stokes seed are presented. It is shown that a fully quantum mechanical scaled planewave theory describes the growth from spontaneous emission well in the high gain limit. However, in the low gain regime there is a significant increase in the amount of Stokes output which is not predicted by the plane wave theory. To account for the deviation from planewave growth observed at low gain, a fully quantum mechanical theory of amplification in a focused gain geometry is presented. The theory is based on a non-orthogonal mode expansion of the amplified field and incorporates the effects of both gain-guiding and diffraction. In addition to accounting for the observed Stokes output at low gain the non-orthogonal mode theory predicts that the Stokes field cart experience enhanced gain as well as have excess noise. Results from an experiment are presented which verify the existence of enhanced gain in a multipass Raman amplifier. Though the experiment does not give a direct measure of the excess noise factor the results indicate that there may be more than the usual quantum limit of 1 photon of noise effectively seeding the Raman amplifier.

GAIN-GUIDING EFFECTS IN STIMULATED RAMAN SCATTERING

by

Philip Ross Battle

A thesis submitted in partial fulfillment
of the requirements for the degree

of

Doctor of Philosophy

in

Physics

MONTANA STATE UNIVERSITY
Bozeman, Montana

April 1993

APPROVAL

of a thesis submitted by

Philip Ross Battle

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

4-2-93
Date

John L Cashman
Chairperson, Graduate Committee

Approved for the Major Department

4-2-93
Date

Head, Major Department

Approved for the College of Graduate Studies

4/6/93
Date

Graduate Dean

STATEMENT OF PERMISSION TO USE

In presenting this thesis in partial fulfillment of the requirements for a doctoral degree at Montana State University, I agree that the Library shall make it available to borrowers under rules of the Library. I further agree that copying of this thesis is allowable only for scholarly purposes, consistent with "fair use" as prescribed in the U.S. Copyright Law. Requests for extensive copying or reproduction of this thesis should be referred to University Microfilms International, 300 North Zeeb Road, Ann Arbor, Michigan 48106, to whom I have granted "the exclusive right to reproduce and distribute my dissertation for sale in and from microfilm or electronic format, along with the right to reproduce and distribute my abstract in any format in whole or in part."

Signature ____________________

Date 4/6/93

ACKNOWLEDGEMENTS

I would like to thank my advisor, John Carlsten, for allowing me the opportunity to work in his lab. His constant encouragement, inexhaustible patience, and boundless enthusiasm has made my tenure here a wonderful experience. Also I would like to thank Rand Swanson for introducing me to many of the interesting and fun ideas which this thesis is based on.

Thanks are also due to the students in John's lab. I am indebted to Jim Wessel for the many hours of insightful and sometimes whimsical conversation and the seemingly endless hours spent working with me on experiments. In addition I would like to acknowledge the many useful discussions with Tim Heumier and Meg Hall.

I am also grateful for the expert advise given by Erik Anderson and Norm Williams.

Further, I wish to thank Dr. Robiscoe and Dr. Cone for their critical reading of this thesis. Their comments and suggested revisions have been incorporated into this thesis.

Perhaps the people I owe the most are my friends outside the physics department. In particular I wish to thank Marilou Turrentine, David Wilson, and Tim Fry for the countless hours of fun we have had together.

Finally I owe my family more than I'll ever be able to repay. Throughout my academic career they have been a constant source of support and encouragement.

TABLE OF CONTENTS

Page

1. INTRODUCTION TO RAMAN SCATTERING 1

   Background and motivation 4

2. SPONTANEOUS EMISSION IN RAMAN SCATTERING 10

   Plane wave theory of stimulated Raman scattering 11
   Extension to a multipass cell 15
   Experimental setup and results 17

3. GAIN-GUIDING EFFECTS IN A FOCUSED GAIN AMPLIFIER 27

   Quantum theory of focused gain amplification 29

4. GAIN-GUIDING EFFECTS IN RAMAN AMPLIFICATION 40

   Experimental results and analysis 41

5. SUMMARY 51

   Future 52

REFERENCES CITED 55

APPENDICES 60

A. Details of generator experiment 61
B. Calculation of the modes in a gain-guided amplifier 65
   Some properties of the non-orthogonal modes 70
   Stucture of the non-orthogonal modes 72
C. Solution to the mode correlation function 75
D. Extension to a multipass cell 80
E. Details of gain-guiding experiment 85

LIST OF FIGURES

Figure Page

1. Energy level diagram 2
2. Components of Raman scattering experiments 3
3. Energy level diagram for molecular hydrogen 6
4. Diagram of focused gain geometry 16
5. Experimental arrangement for spontaneous emission experiment 18
6. Stokes energy as function of pump energy 19
7. Stokes energy in the linear regime 20
8. Stokes energy as a function of gz 22
9. Comparison of single mode theory to focused gain theory 23
10. Number of non-gaining spatial modes 24
11. Corrected plane wave theory 25
12. Coordinates of a focused gain geometry 31
13. Gain of dominant non-orthogonal mode 38
14. Experiment apparatus used to study gain guiding effects 43
15. Stokes Energy as a function of the gain length parameter(long cell) . . 45
16. Stokes Energy as a function of the gain length parameter(short cell) . . 46

LIST OF FIGURES - Continued

Figure Page

17. Transverse spatial profile of free space mode and non-orthogonal mode 48

18. Excess noise factor for short and long cell experiment 49

19. Spatial profile of both non-orthogonal mode and free space mode 73

20. Coordinates for extension of results to a multipass cell 81

ABSTRACT

Measurements of the output of a Raman amplifier when there is no input Stokes seed are presented. It is shown that a fully quantum mechanical scaled planewave theory describes the growth from spontaneous emission well in the high gain limit. However, in the low gain regime there is a significant increase in the amount of Stokes output which is not predicted by the plane wave theory. To account for the deviation from planewave growth observed at low gain, a fully quantum mechanical theory of amplification in a focused gain geometry is presented. The theory is based on a non-orthogonal mode expansion of the amplified field and incorporates the effects of both gain-guiding and diffraction. In addition to accounting for the observed Stokes output at low gain the non-orthogonal mode theory predicts that the Stokes field can experience enhanced gain as well as have excess noise. Results from an experiment are presented which verify the existence of enhanced gain in a multipass Raman amplifier. Though the experiment does not give a direct measure of the excess noise factor the results indicate that there may be more than the usual quantum limit of 1 photon of noise effectively seeding the Raman amplifier.

CHAPTER 1

INTRODUCTION

The term Raman scattering is used to describe an inelastic photon scattering process. When a photon scatters off a Raman active medium it can either deposit or extract energy from the medium depending on the initial state of the medium. Though Raman scattering is typically done using the vibrational and/or rotational states of molecules in a gas or liquid it can also be done using electronic states of an atom or can occur through an interaction of the incident photon with lattice or phonon modes of a solid.

When a photon is red shifted in the Raman process it is commonly referred to as a Stokes photon. In Fig.1 an energy level diagram of the Stokes process is shown. The Raman active molecule starts in the ground state $|1\rangle$. An incident pump photon with energy $\hbar\omega_p$ scatters off the molecule leaving it in level $|3\rangle$. In order to conserve energy the scattered Stokes photon energy is given by $\hbar\omega_s = \hbar\omega_p - \hbar\omega_{31}$. The Raman process can also lead to blue shifted photons referred to as anti-Stokes photons.

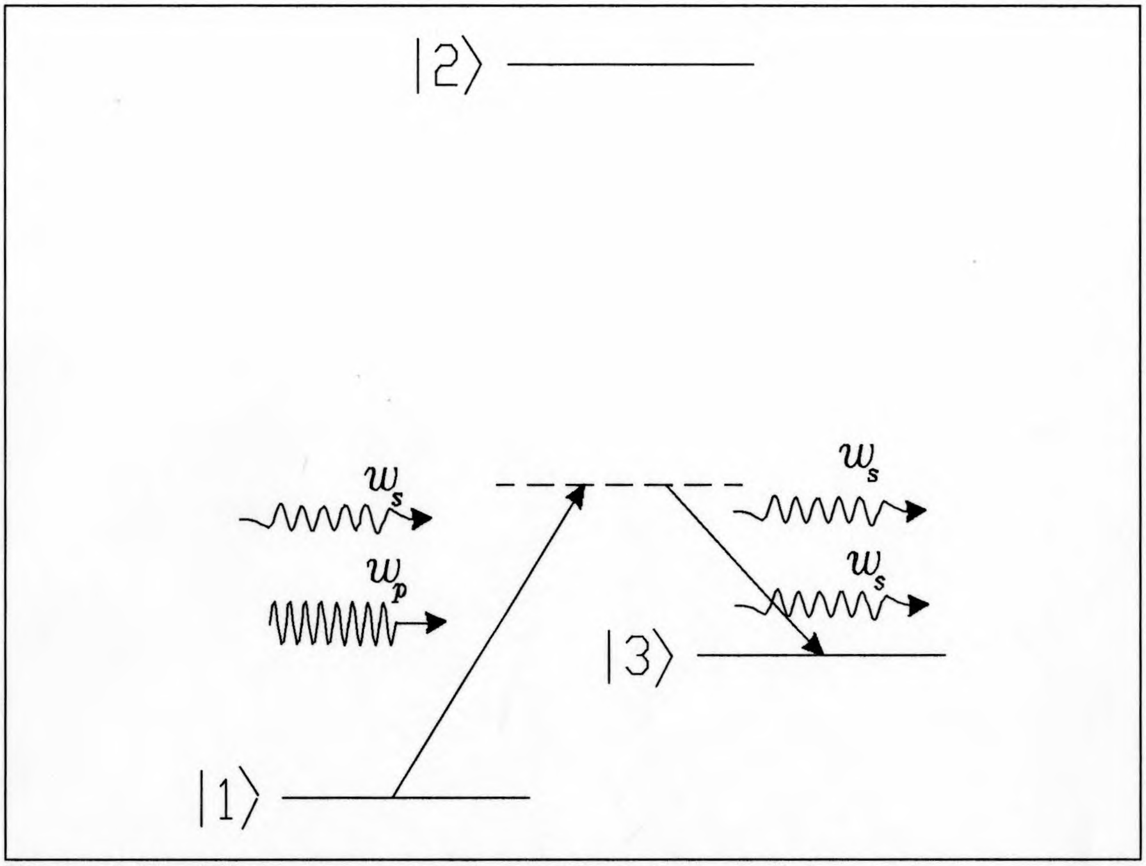

Figure 1 Simplified energy level diagram for Raman scattering process

There are two scattering mechanisms which contribute to the production of anti-Stokes photons. The non-resonant contribution to anti-Stokes scattering occurs when the pump photon scatters off a molecule which starts in the excited state, $|3\rangle$. The scattered photon is blue shifted in energy and the molecule is left in the ground state, $|1\rangle$. In resonant or coherent anti-Stokes scattering (CARS) the blue shifted photon results from a four-wave mixing between the Stokes and pump fields. In this case the molecule starts in the ground state and ends there as well. For both paths energy conservation dictates

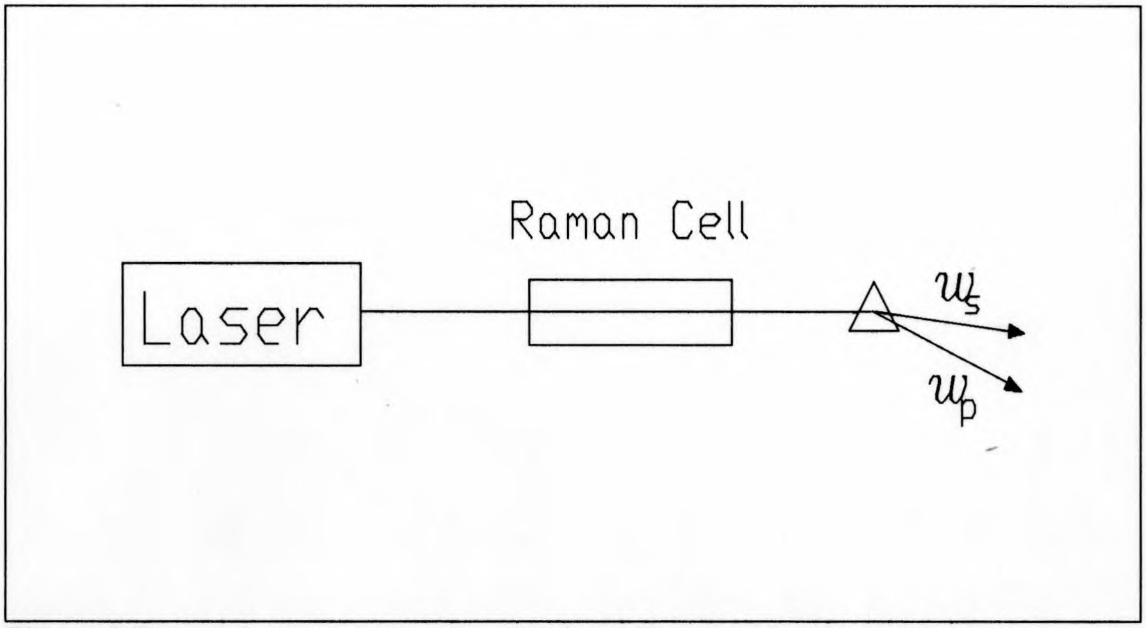

Figure 2 Essential components of a stimulated Raman scattering experiment.

that the resulting anti-Stokes photon has an energy given by $\hbar\omega_{AS} = \hbar\omega_p + \hbar\omega_{31}$.

Stokes scattering can occur spontaneously or, as alluded to in Fig.1, can be stimulated by other Stokes photons which are incident on the molecule at the same time the pump photon interacts with the molecule. The latter process is referred to as stimulated Raman scattering. Even in the absence of an initial Stokes field, stimulated Raman scattering can still occur. In Fig.2 the essential details of a Raman scattering experiment are shown. An intense laser is used to pump a Raman active medium. Since no Stokes photons are incident with the pump field only spontaneous Raman scattering occurs. However some of the initial spontaneous Stokes photons travel in the direction of the pump and are subsequently amplified through the stimulated Raman process discussed above. Exploring both experimentally and theoretically the various aspects of quantum initiated stimulated Raman scattering is the central theme of this thesis.

Background and Motivation

Stimulated Raman scattering(SRS), although actively studied since 1962,[1] continues to be an important and exciting research field as well as a practical and useful tool. Along with the well known applications in spectroscopy[2-5] such as generation of efficient tunable lasers in the infrared, SRS has recently been used to generate very narrow linewidth pulses.[6] In the non-linear regime it has been shown that the equations describing SRS admit soliton solutions.[7] These solitons have been observed to occur spontaneously and recently much effort has gone into studying and understanding the role of quantum fluctuations in the generation and propagation of the SRS solitons.[8-11]

SRS has also been used to gain insight into the microscopic quantum world, through amplification of the vacuum field the microscopic quantum fluctuations are manifest in frequency, time, spatial and energy fluctuations of the macroscopic output Stokes field.[12-15] In addition it has been demonstrated both theoretically and experimentally that a Raman amplifier can be operated at the phase insensitive quantum noise limit of 1 photon per mode.[16,17] In this thesis I present and discuss experimental data on SRS generation in a multipass cell (MPC).

There are several advantages to doing SRS, as well as other stimulated emission processes, in a MPC. In a MPC, the amplified field and the pump field are focused on each pass. In a focused geometry where the confocal parameter of the pump field is small compared to the medium length the threshold for amplification is minimized.[6,18,19]

The MPC can also be used to increase the gain. The gain enhancement is due to the multiple passes which effectively increase the length of the medium. This gain enhancement, which can easily be as much as an order of magnitude with the use of a MPC,[24]is extremely useful in cases where the gain is very small, such as SRS of CO_2 lasers in the IR.

As well as threshold reduction and gain enhancement, the MPC provides an excellent arena for studying first Stokes generation and amplification since focusing and wavelength selective mirrors each reduce the gain of higher order emission processes. For example, it has been shown that for single focus SRS, the ratio of anti-Stokes to Stokes is reduced to the 10^{-3} level.[18,20] For a MPC configured with mirrors that reflect strongly only at the Stokes and pump wavelengths the anti-Stokes to Stokes ratio is further reduced, and other four-wave mixing processes such as second or higher order Stokes are inhibited. In addition, the MPC can be configured, as in our experiment, so that the transit time of the pump through the MPC is greater than the temporal width of the pump. In this configuration the growth of backward Stokes is also greatly reduced. Thus the amplification of the Stokes pulse in a MPC can be studied without the usual complications of higher order mixing processes which accompany a high gain single pass system.

In both the experimental and theoretical work discussed in this thesis the Raman medium used was molecular hydrogen. Molecular hydrogen, H_2, is a homonuclear diatomic molecule. In Fig.3 a detailed (though not to scale) diagram of the energy levels

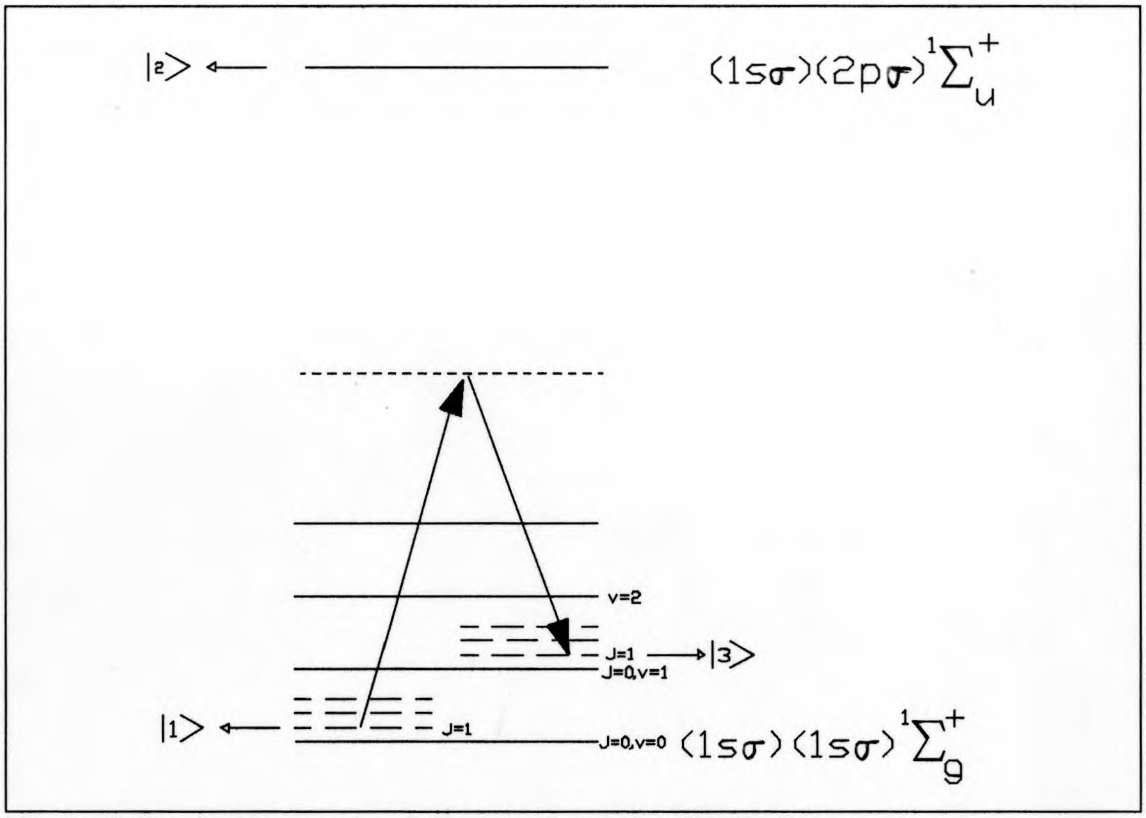

Figure 3 Detailed energy level diagram for molecular Hydrogen.

for H_2 is shown. The ground electronic state is denoted $(1s\sigma)(1s\sigma)\ ^1\Sigma_g^+$, where Σ indicates that the state has zero projected angular momentum, the indices + and g indicate the symmetry properties of the electronic wavefunction and the superscript 1 indicates that the electrons are in a singlet spin configuration.[21] The coarsely-spaced energy levels above the ground state are due to the vibrational degree of freedom in the molecule and are denoted by $v=0,1,2..$. The first few vibrational levels are separated in energy by approximately 0.5 eV. In addition, for each vibrational level there is a finer energy level structure corresponding to the rotational degrees of freedom which are denoted by J=0,1,2... and are separated in energy by approximately 0.03eV. The first electronic excited state denoted $(1s\sigma)(2p\sigma)\,{}^1\Sigma_u^+$, is separated from the ground state by approximately 90,000cm^{-1} or 11.2eV. Though not shown, there is a similar coarse and fine scale energy level structure associated with this state.

Raman scattering transitions amongst these levels do not take place arbitrarily but are constrained by the selection rules $\Delta v=0, \Delta J=0,\pm 2$ or $\Delta v=\pm 1, \Delta J=0,\pm 2$. Because in our work the pump photons were linearly polarized, we only had to consider pure vibrational Raman scattering, $\Delta v=\pm 1, \Delta J=0$. The nomenclature used to label pure vibrational transitions is $Q_{v,v'}(J)$ where v is the initial vibrational level, v' is the final vibrational level and J is the rotational state of the molecule.

At room temperature, which is the temperature at which our work was done, the molecule is primarily in the ground electronic state with v=0. However, due to the combined nuclear and rotational degeneracies, a majority of the molecules(66%) populate the J=1 rotational state rather then the lowest, J=0, rotational state. The following correspondences can be made to the simplified energy level diagram given in Fig.1; the ground state, $|1\rangle$, corresponds to the electronic ground state $(1s\sigma)^2\,{}^1\Sigma_g^+$

with v=0 and J=1, level $|2\rangle$ is identified as the first electronic excited state, and level $|3\rangle$ again corresponds to the ground electronic state but now has v=1 and J=1. This is summarized by saying that Raman scattering was done using the $Q_{01}(1)$ branch of H_2. In this case a pump photon at 532nm(green) undergoing Raman scattering will be Stokes shifted to become a photon at 683nm(red).

In the following chapter a brief description of the fully quantum mechanical plane wave theory of stimulated Raman scattering is presented. The theory accounts for both quantum initiation and propagation of the Stokes field. In addition we present a method for extending the jurisdiction of this theory to Raman scattering in a multipass cell. Finally we show the results of several experiments which point out some limitations of the plane wave theory in describing the growth from spontaneous emission in a multipass cell.

In Chapter 3 we develop a general three dimensional theory of steady state amplification in a focused gain geometry. The theory is based on a nonorthogonal mode expansion of the amplified field. The implications of including both the effects of diffraction and focused gain are explored. We show that the gain-guided nature of a focused-gain amplifier can have a large and profound effect on its output. In particular our analysis shows that the gain of the field can become larger than the usual plane wave gain and that the amount of noise effectively seeding the amplified field can be larger than the usual quantum limit of one photon per mode.

In Chapter 4 we summarize the results of two experiments designed to look for the excess noise in output of the gain-guided Raman amplifier. The experiments were performed using essentially the same apparatus as in the first set of experiments described in Chapter 2. The only notable change was the replacement of our old Nd:YAG laser with a new injection seeded YAG laser that might rightly be characterized as the "cadillac" of YAG's. The results of the experiments indicate that gain-guiding can indeed affect the noise properties of SRS.

Finally Chapter 5 concludes with a brief summary of the results in each of these chapters. In addition, directions that future work could follow are discussed.

CHAPTER 2

GROWTH FROM SPONTANEOUS EMISSION IN STIMULATED RAMAN SCATTERING

Recently, in one of the many theoretical and experimental investigations of amplification in a multipass cell (MPC),[20,22-24] Mac Pherson *et al.* demonstrated that, for stimulated Raman scattering (SRS) in the visible, amplification of an input Stokes pulse is well described by a semi-classical transient plane wave theory, even in the non-linear regime where pump depletion is significant. The focusing and multiple passes that occur in the MPC were accounted for by simply scaling the plane wave gain coefficient. In this chapter we show that the growth from spontaneous emission can also be described by a fully quantum mechanical transient plane wave theory, but only for relatively high gain. In the high gain regime we find that if the gain coefficient is scaled in the same manner as previous work[4;20,24] the growth of the Stokes field is well described.

In the low gain regime we find there is much more Stokes energy observed than predicted by the simple scaled plane wave theory. In order to obtain an insight into the origin of the excess Stokes energy, we considered the full three dimensional propagation/amplification problem. The solution to this was presented by Perry *et al.*,[22] for the case of steady state growth. By including the effects of spontaneous emission phenomenologically we are able to calculate the Stokes intensity from the MPC. In the low gain regime, we find that the effects of higher order spatial modes become significant, indicating that a simple transient plane wave model is no longer adequate to describe the Stokes growth.

Plane Wave Theory of Stimulated Raman Scattering

Raman scattering is driven by a coherent interaction between the medium, a pump field and a Stokes field. The beating of the pump and Stokes field produce a coherence between states $|1\rangle$ and $|3\rangle$ in the Raman active medium which, for our experiment, correspond to the ground state and first vibrational state in H_2. The induced molecular coherence then couples back to the pump field to produce the non-linear polarization at the Stokes frequency, $P_{nl}^{\omega_s} \sim (E_p^* E_s) E_p = |E_p|^2 E_s$. Stimulated Raman scattering (SRS) occurs because the rate of Stokes growth, which is proportional to the nonlinear polarization, is directly proportional to the amount of Stokes present. This implies that the Stokes growth will have exponential character, a hallmark of stimulated emission processes.

On first inspection of the non-linear polarization given above, one might conclude that if there is no injected Stokes field in the medium then, because the polarization is "zero", no Raman scattering occurs. Experimentally, however, this is not the case; when a Raman active medium is pumped in the absence of an initial Stokes field, spontaneous Raman scattering is observed. This apparent paradox is resolved when a fully quantum mechanical description of Raman scattering is considered.

In the fully quantum mechanical approach both the molecular response and the Stokes field are quantized. Then, just as the harmonic oscillator in its ground state has zero point fluctuations, the Stokes field, even with no photons in it, also has quantum zero point fluctuations. It is these quantum fluctuations in the Stokes field, often referred to as vacuum fluctuations, which are the origin of spontaneous Stokes scattering. A compelling feature of the fully quantum mechanical approach is that it ties together two seemingly distinct scattering mechanisms; using this model spontaneous emission can be thought of as simply, stimulated emission of the vacuum field.

A fully quantum mechanical description of plane wave amplification process for SRS has been presented by Raymer and Mostowski.[25] In deriving the Maxwell-Bloch equations that account for both growth and propagation of the Stokes field, they consider a collection of molecules in a pencil shaped region of length l_m and cross sectional area, A, uniformly pumped by a plane monochromatic field. The assumptions made are that the medium is left primarily in the ground state, and the pump field remains undepleted. In the absence of pump depletion, the output Stokes field $\hat{E}_s(z,\tau)$ is related linearly to the input Stokes field $\hat{E}_s(0,\tau)$, where $\tau = t - z/c$ is the retarded time, and z labels the position of some point along the axis of the amplifier. $\hat{E}_s^{(-)}(z,\tau)$ is the slowly varying negative frequency amplitude of the Stokes electric field

$$\hat{E}_s(z,\tau)=\hat{E}_s^{(+)}(z,\tau)e^{-i\omega_s\tau} + \hat{E}_s^{(-)}(z,\tau)e^{i\omega_s\tau} \quad (2.1)$$

In terms of the retarded time parameter, the differential equations describing the growth of the Stokes field and its coupling to the medium are given by

$$\frac{\partial}{\partial z}\hat{E}_s^{(-)}(z,\tau)=-ik_2\hat{Q}^\dagger(z,\tau)E_p(z,\tau) \quad (2.2a)$$

and

$$\frac{\partial}{\partial \tau}\hat{Q}^\dagger(z,\tau)=-\Gamma\hat{Q}^\dagger(z,\tau)+ik_1E_p^*(z,\tau)\hat{E}_s^{(-)}(z,\tau)+\hat{F}^\dagger(z,\tau) \quad (2.2b)$$

where $\hat{Q}^\dagger(z,\tau)$ represents the polarization fluctuations in the medium, $\hat{F}^\dagger(z,\tau)$ is a quantum Langevin operator added to account for collisional dephasing, and Γ is the dephasing rate in the Raman medium. Also, in Eq.(2.2) k_1 and k_2 are constants related to the field-molecule coupling strength[25] and can be expressed in terms of the plane wave gain coefficient, α, by $k_1k_2=\frac{\Gamma c\alpha}{16\pi}$

Though somewhat disguised, the first equation, 2.2a, is simply the steady state slowly varying form of Maxwell's wave equation. The right hand side of this equation is proportional to the non-linear polarization which is produced from the coupling between the induced molecular coherence, $\hat{Q}^{\dagger}(z,\tau)$ and the pump field E_p. The second equation, 2.2b, represents growth of the molecular coherence which results from the beating of the pump and Stokes fields.

The solution to Eq.(2.2) in terms of the Stokes field $\hat{E}_s^{(-)}(z,\tau)$ can be found in ref.[25]. The average Stokes power is related to the expectation value of the normally ordered product of the Stokes field and can be written as[26]

$$P_s(\tau)=\frac{Ac}{2\pi}\langle\hat{E}_s^{(-)}(z,\tau)\hat{E}_s^{(+)}(z,\tau)\rangle \tag{2.3}$$

$$=gz|\gamma(\tau)|^2\Gamma\hbar\omega_s\left(\frac{1}{2}+gz\Gamma\int_0^{\tau}d\tau'\,|\gamma(\tau')|^2e^{-2\Gamma(\tau-\tau')}\frac{I_1^2(\sqrt{q(\tau,\tau')})}{q(\tau,\tau')}\right).$$

where the pump field has the form $E_p(\tau)=E_o\gamma(\tau)$, $g = \alpha I_p$ and I_1 is a modified Bessel function of first order. In the steady state, $\Gamma\tau_p$>>1, high gain limit the Stokes intensity increases exponentially with increasing pump power. In this limit Eq.(2.3) becomes

$$P_s\approx\frac{\hbar\omega_s\Gamma}{2(\pi gz)^{1/2}}e^{gz} \tag{2.4}$$

which resembles the usual semi-classical high gain result and justifies calling α the plane wave gain coefficient.

Extension to a Multipass Cell

A theoretical investigation of amplification in a focused gain geometry, such as a multipass cell, has shown that when the gain per pass is small the spatial modes which describe the growth act independently.[22] Additionally when the amplified field's wavelength is comparable to the pump field's wavelength only one mode has dominant growth.[22] For SRS with a visible laser, the above conditions can easily be met and thus single mode growth can be expected. Previous theoretical and experimental studies of Raman amplification in a MPC have shown that Stokes amplification can be described by a plane wave theory.[20,23] In this approach, however, the usual gain length coefficient, gz, is scaled to account for the focusing and multiple passes.

In the next section we compare the output from a Raman generator in a MPC to the fully quantum mechanical plane wave theory presented above. As in previous work, to account for focusing, we replace the non-uniformly pumped geometry by a uniformly pumped region with an effective area given by[20]

$$A_{eff}=\frac{l_m(\lambda_s+\lambda_p)}{4\tan^{-1}(l_m/b)} \tag{2.5}$$

where, as shown in Fig.4, l_m is the medium length and b is the confocal parameter of