



Gain enhancement in a XeCl pumped Raman amplifier
by Jeffrey Rifkin

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in
Physics

Montana State University

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Abstract:

Gain enhancement in an excimer-pumped Raman amplifier consisting of molecular hydrogen is experimentally measured. Gain enhancement is measured both as a function of optical delay and input pump intensity, and in the two limits when the laser mode spacing is large, and comparable to the linewidth associated with the Raman medium. Experimental results are compared with the predictions of a transient multimode theory. This theory is based on the coupled equations of Raman scattering which describe the transient growth of the Stokes field and coherence of the Stokes transition. A sum over longitudinal modes with fixed and totally random phases is assumed for both the pump laser and Stokes fields.

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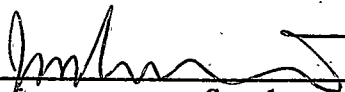
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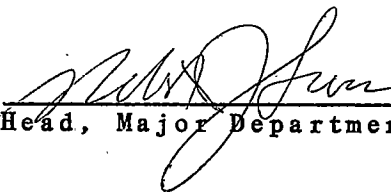
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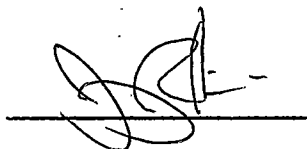

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ABSTRACT

Gain enhancement in an excimer-pumped Raman amplifier consisting of molecular hydrogen is experimentally measured. Gain enhancement is measured both as a function of optical delay and input pump intensity, and in the two limits when the laser modespacing is large, and comparable to the linewidth associated with the Raman medium.

Experimental results are compared with the predictions of a transient multimode theory. This theory is based on the coupled equations of Raman scattering which describe the transient growth of the Stokes field and coherence of the Stokes transition. A sum over longitudinal modes with fixed and totally random phases is assumed for both the pump laser and Stokes fields.

The theory compares well with experiment in the regime where the Raman linewidth is large compared to the modespacing of the laser. The theory also makes the interesting prediction that in this limit, the broadband gain will be larger than the gain for a narrowband laser.

CHAPTER ONE

INTRODUCTION

Raman scattering was first observed in 1928 by C.V. Raman¹. A Raman medium is defined as a medium capable of coupling optical frequencies which differ by a vibrational, rotational, or electronic frequency. Raman scattered light is referred to as either Stokes or anti-Stokes radiation² depending upon whether the coupling downshifts or upshifts the frequency of the scattered light.

The laser or pump used for Raman scattering can be categorized in one of two ways, either narrowband (monochromatic) or broadband. Numerous models exist to describe the broadband pump. The three most common are the phase diffusion, chaotic, and multimode models. These models all describe the pump as a broadband wave with either fluctuations in amplitude and/or fluctuations in phase. The phase diffusion model³ assumes constant amplitude with randomly fluctuating phases, e.g. $E(t) = E \cos(\omega t - k z + \phi(t))$, where E is the amplitude and ϕ is the phase. The chaotic model⁴ assumes both randomly fluctuating amplitudes and phases, e.g. $E(t) = E(t) \cos(\omega t - k z + \phi(t))$, while the multimode model⁴ uses a distribution of widely separated modes, fixed in both amplitude and phase so that $E(t) = \sum E_n \cos(\omega_n t - k_n z + \phi_n)$.

When the pump is of low enough intensity, only spontaneous Raman scattering (spontaneous emission) or SE will take place. As pump intensity is increased, stimulated Raman scattering or SRS will occur. SRS can be divided into two categories: Raman generation and Raman amplification.

The Raman generator has been studied by numerous authors⁴⁻¹⁴. The result from these studies relevant to this work is the prediction that in certain cases, the generated Stokes radiation, or field, may be correlated with the pump during its growth from SE in the generator. By correlation, one means that fluctuations in the Stokes field correspond to fluctuations in the pump field. This is predicted to be the case for the 1) phase diffusion model and 2) both the chaotic and multimode models in the limit when the laser bandwidth, Γ_L , is larger than the Raman linewidth, Γ_R .

Raman amplification, although studied by an equally large number of authors¹⁵⁻³⁴ has been mostly theoretical, rather than experimental in nature. Numerous applications of Raman amplification however have been exploited. Cleanup of spatially poor quality pump lasers through Raman conversion¹⁵⁻¹⁹, and access to high energy beams through the multiplexing of several pump lasers with a single Stokes beam in a Raman amplification medium¹⁸⁻²² are two of the most common applications.

Early work on Raman amplification^{3,23-29} using models derived from the phase diffusing and multimode models showed

that the amplified Stokes spectrum was always narrower than the pump spectrum below a critical pump intensity. Above this critical intensity, the Stokes spectrum approached or even equaled the pump spectrum in width. It was also discovered that the gain for broadband amplification was nonlinear and always less than the narrowband gain below the critical pump intensity. These two effects were attributed to dispersion in the Raman amplification medium and hence related to the spectral width of the pump. As pump intensity was increased above the critical intensity, broadband gain became independent of both pump bandwidth and dispersion, and consequently approached the narrowband value. Raymer et al.³, using the phase diffusion model, predicted theoretically that the pump and Stokes fields may become correlated in phase, which leads to monochromatic gain. Lombardi et al.³⁰, later showed experimentally that the Stokes does indeed acquire the same phase as the pump.

Vokhnik et al.³¹ and Stappaerts et al.³² were the first authors to investigate the effects of correlation between the pump and injected Stokes seed on gain enhancement. These authors showed that when the two fields were perfectly correlated, the gain enhancement is a maximum and approaches the narrowband value. Both Agarwal³³ and Georges³⁴ using the chaotic model made the interesting prediction that when the pump linewidth is narrower than the Raman linewidth, and when the two beams are well correlated, gains which are

larger than the monochromatic gain are to be expected.

It appears that no single model for Raman scattering will describe every experimental situation and each researcher must decide which model will work for any given laser and each particular experiment. In Dr. Carlsten's lab a XeCl laser (described in chapter three) is used to do Raman scattering. This thesis results from a first attempt at describing the radiation from a XeCl laser and it's interaction with the Raman medium. A multimode-fixed-random-phase model is developed, and using this model, the effects of correlation, linewidth, and input pump intensity on the broadband gain are studied. Experiments are performed and experimental results are compared with the theoretical predictions of this model.

CHAPTER TWO

DERIVATION OF THE RAMAN SCATTERING EQUATIONS

Introduction

A Raman medium is one capable of coupling optical frequencies which differ by a vibrational, rotational, or electronic frequency. The body of the work presented in this thesis will be concerned with vibrational Raman scattering; thus a brief description is appropriate.

Raman scattering is a two photon processes. Physical insight into vibrational Raman scattering is obtainable with a simple model. Consider the three level molecule in Fig.1. When a pump photon of frequency ω_p (also referred to as ω_L) is absorbed by the molecule, a Stokes photon of frequency ω_s is emitted and the molecule is left vibrating at the difference frequency ω_{13} , where $\hbar\omega_{13}$ is the energy difference between the molecular levels one and three. This difference frequency is referred to as the Raman shift.

A theoretical description of Raman scattering is considerably more complicated since both Schrodinger's equation and Maxwell's equations must be used. A feel for the theoretical approach may be obtained by referring to the block diagram in Fig.2. The interaction between the pump and Stokes fields in the medium is described by

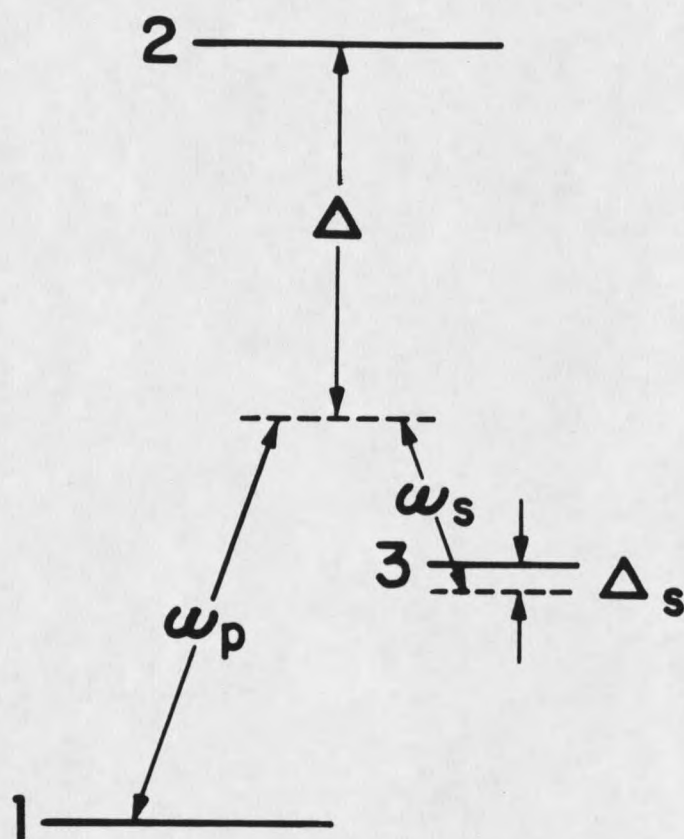


Figure 1. Energy level diagram for stimulated Raman scattering. A three level molecule interacts with a pump photon of frequency ω_p and a Stokes photon of frequency ω_s . The molecule is left vibrating at the difference frequency ω_{13} .

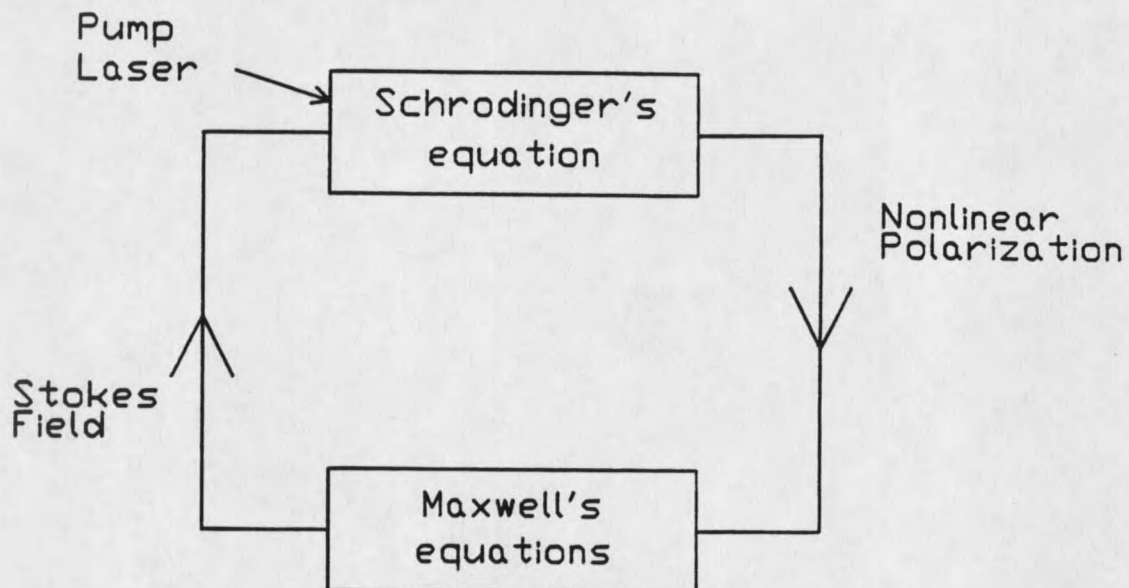


Figure 2. Self consistency diagram for stimulated Raman scattering. The pump and Stokes fields interact via Schrodinger's equation and produce a non-linear polarization in the medium. The non-linear polarization then drives the Stokes field via Maxwell's equation.

Schrodinger's equation. A nonlinear polarization in the medium results from this interaction and becomes the driving term in Maxwell's wave equation. The nonlinear polarization completely describes the macroscopic response of the medium to the incident fields, while Maxwell's wave equation completely describes the response of the fields to the medium. The remainder of this section will be devoted to the details of this process.

Response of the Medium

One begins by writing out the wave function for a system of three level molecules (described in chapter three) interacting with a laser field of frequency $\hbar\omega_L$ and a Stokes field of frequency $\hbar\omega_S$. An energy level diagram for the system is shown in Fig.1. Following Raymer et al.³ we let

$$\psi = a_1 \exp^{-i\Phi_1} U_1 + a_2 \exp^{-i(\Phi_1 + \Phi_L)} U_2 + a_3 \exp^{-i(\Phi_1 + \Phi_L - \Phi_S)} U_3. \quad (1)$$

The a_i are the amplitudes of level i , Φ_i is equal to $\omega_i t - k_i z$ and the U_i are the stationary states having energies $\hbar\omega_i$. Since we are free to choose the zero of energy it is convenient to choose it to be the ground state with the phase, $\Phi_1 = 0$.

The Hamiltonian for the system is

$$H = H_0 + H'. \quad (2)$$

H_0 is the Hamiltonian for the unperturbed system having eigenvalues $E_i = \hbar\omega_i$, while H' , the perturbing Hamiltonian, describes the interaction of light with the system and is

given by the dipole interaction³⁵

$$H' = \mu \cdot E = -ex \frac{1}{2} (E \exp i\Phi + \text{c.c.}). \quad (3)$$

We let the system evolve by putting ψ into Schrodinger's equation, $i\hbar\dot{\psi} = H\psi$. This yields

$$\begin{aligned} i\hbar(a_1\dot{U}_1 + [a_2 - ia_2\omega_L]\exp^{-i\Phi_L}\dot{U}_2 \\ + [a_3 - ia_3(\omega_L - \omega_S)]\exp^{-i(\Phi_L - \Phi_S)}\dot{U}_3) \\ = Va_1U_1 + (\hbar\omega_2 + V)a_2\exp^{-i\omega_L t}U_2 \\ + (\hbar\omega_3 + V)a_3\exp^{-i(\omega_L - \omega_S)t}U_3. \end{aligned} \quad (4)$$

Perturbing the system with H' creates the probability of finding populations in excited states. The amplitude for populating excited states is determined by evaluating

$$i\hbar\langle\dot{\psi}|\dot{\psi}\rangle = \langle\dot{\psi}|H|\dot{\psi}\rangle. \quad (5)$$

Collecting terms proportional to U_1 we obtain

$$i\hbar\dot{a}_1 = V_{11}a_1 + V_{12}a_2\exp^{-i\Phi_L} + V_{13}a_3\exp^{-i(\Phi_L - \Phi_S)}. \quad (6)$$

The dipole interaction only couples adjacent states,

therefore $V_{11}=V_{13}=0$, and Eqn.6 simplifies to,

$$ia_1 = -\Omega_{12}a_2 \quad (7)$$

where the Rotating Wave Approximation (RWA)³⁵ has been used

and $\Omega_{ij} = eX_{ij}E/2\hbar$ is the Rabi frequency³⁶. Similarly, we find

$$ia_2 = \Delta_L a_2 - \Omega_{21}^* a_1 - \Omega_{23}^* a_3 \quad (8)$$

and

$$ia_3 = a_3 \Delta_S - \Omega_{32} a_2. \quad (9)$$

$\Delta_L = \omega_2 - \omega_L$ describes how far the laser is tuned from the resonant 1-2 transition and is assumed to be very large for the purposes of this derivation (see appendix A), while

$\Delta_S = \omega_3 - (\omega_L - \omega_S)$ is the detuning for the Raman transition and assumed to be negligibly small.

Determination of the population $|a_i|^2$ of level i requires the simultaneous solution of the system of equations 7, 8, and 9. It is convenient to solve Eq. 8 first. Integrating both sides of this expression yields

$$a_2(t) = \int_0^t G(t') \exp^{i\Delta_L(t'-t)} dt', \quad (10)$$

where $G(t') = i(\Omega_{12}^* a_1 + \Omega_{32}^* a_3)$.

Integrating by parts gives

$$a_2(t) = \frac{G(t) - G(0)\exp^{-i\Delta_L t}}{i\Delta_L} - \frac{G(t) - G(0)\exp^{-i\Delta_L t}}{(i\Delta_L)^2} - \frac{G(t) - G(0)\exp^{-i\Delta_L t}}{(i\Delta_L)^3} \quad (11)$$

The second and third terms in this expression will be small compared to the first term for large Δ_L and can be ignored. At $t=0$, $a_1(0)=1$ and $a_2(0)=a_3(0)=0$ so that $G(0) = -i\Omega_{12}^* a_1$.

Thus

$$a_2(t) = \frac{(\Omega_{12}^* a_1 + \Omega_{32}^* a_3)}{\Delta_L} - \frac{\Omega_{12}^* a_1 \exp^{-i\Delta_L t}}{\Delta_L} \quad (12)$$

For large Δ_L , the second term will be oscillating very fast compared to the first term and can be ignored so that

$$a_2(t) \approx \frac{(\Omega_{12}^* a_1 + \Omega_{32}^* a_3)}{\Delta_L} \quad (13)$$

This is tantamount to $\dot{a}_2 \approx 0$. These approximations conveniently reduce the three level problem to a two level

problem; the system can be considered to be oscillating between levels 1 and 3 only.

Rewriting equations 7 and 9 with the expression for a_2 given by Eq.13 we obtain

$$a_1 = \frac{i(|\Omega_{12}|^2 a_1 + \Omega_{12} \Omega_{32}^* a_3)}{\Delta_L} \quad (14)$$

and

$$a_3 = \frac{i(\Omega_{32} \Omega_{12}^* a_1 - |\Omega_{32}|^2 a_3)}{\Delta_L} \quad (15)$$

Again, rather than solving these two simultaneous equations it is expedient to introduce the density matrix formulation³⁵ by defining

$$Q^* = 2a_1^* a_3 \quad (16)$$

where Q^* is related to the off diagonal density matrix elements and has the physical interpretation of a coherence between the states a_1 and a_2 . Additionally, with this formulation it is possible to include the effects of collisions between molecules in the medium. The effect of collisions is to introduce dephasing between the ground and excited states which in turn destroys the coherence between these states. The time evolution of the coherence is given by

$$\frac{\partial Q^*}{\partial t} = 2(a_1^* \dot{a}_3 + \dot{a}_1^* a_3) - \Gamma Q^* \approx \frac{i2\Omega_{32}\Omega_{12}^*}{\Delta_L} - \Gamma Q^* \quad (17)$$

or

$$\frac{\partial Q^*}{\partial t} = -\Gamma Q^* + ik_1 E_S E_L^* \quad \text{with } k_1 = \frac{d_{12}d_{23}}{2\Delta_L \hbar^2} \quad (18)$$

where terms proportional to a_3 have been assumed small and neglected, collisional dephasing is accounted for by the inclusion of the damping term Γ , and the d_{ij} are the matrix elements $e\langle U_i | X | U_j \rangle$. Eq.18 tells us that the pump and Stokes fields create coherence in the medium and collisions between molecules destroy it. Eq.18, then describes the microscopic response of the medium to the applied fields.

The macroscopic response of the medium is manifest in the polarization which is given by

$$P_{ij} = N\langle \psi_i | d \cdot X | \psi_j \rangle + \text{c.c.} \quad (19)$$

where $d \cdot X$ is the dipole moment per unit volume and N is the number of dipoles contained within the volume. The component of the polarization which drives the Stokes field is given by

$$P_{23} = N\langle \psi_2 | d \cdot X | \psi_3 \rangle + \text{c.c.} \quad (20)$$

With the wave function given by Eq.1 along with Eq.13 and Eq.16 the polarization can be written in terms of the coherence Q^* , yielding

$$P_{23} = Nd_{23}(\Omega_{21}Q^* \exp^{i\Phi S} + \Omega_{21}^*Q^* \exp^{-i\Phi S}). \quad (21)$$

So, using the pump and Stokes fields in Schrodinger's equation we have created a polarization in the medium, and thus completely described the macroscopic response of the medium to the applied fields.

Response of the Field

The polarization drives the Stokes field via Maxwell's

wave equation whose derivation follows. We begin with Maxwell's equations³⁷

$$\text{I. } \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad \text{II. } \nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t} \quad \text{III. } \nabla \cdot \mathbf{D} = \rho$$

and Ohm's law

$$\text{IV. } \mathbf{J} = \sigma \mathbf{E}$$

along with the relations

$$\text{V. } \mathbf{B} = \mu \mathbf{H} \quad \text{VI. } \mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$$

and the vector identity

$$\text{VII. } \nabla \times \nabla \times \mathbf{A} = (\nabla \cdot \mathbf{A}) \nabla - \nabla^2 \mathbf{A}.$$

Substituting V. into I. and taking the curl of both sides results in

$$\nabla \times \nabla \times \mathbf{E} = -\mu \frac{\partial (\nabla \times \mathbf{H})}{\partial t}.$$

Applying the identity VII. and substituting IV. and VI.

$$(\nabla \cdot \mathbf{E}) \nabla - \nabla^2 \mathbf{E} = -\mu \sigma \frac{\partial \mathbf{E}}{\partial t} - \mu \epsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} - \mu \frac{\partial^2 \mathbf{P}}{\partial t^2}. \quad (22)$$

The polarization is now separated into linear and nonlinear parts, that is, $\mathbf{P} = \mathbf{P}_L + \mathbf{P}_{NL}$. The linear polarization is given by³⁷ $\mathbf{P}_L = \epsilon_0 \chi_L \mathbf{E}$, so that Eq.22 can be rewritten

$$\nabla^2 \mathbf{E} - (\nabla \cdot \mathbf{E}) \nabla = \mu \sigma \frac{\partial \mathbf{E}}{\partial t} + \mu \epsilon \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2} \quad (23)$$

where $\epsilon = (1 + \chi_L) \epsilon_0$ is the permittivity³⁷ of the medium having a linear susceptibility χ_L . For wave propagation in gaseous media, there are no free charges which means that both the conductivity σ , and the divergence of $\mathbf{E}$ can be taken to be

zero.

Then, writing μ as $1/v^2$, Eq.23 simplifies

$$\nabla^2 E_S - \frac{1}{v^2} \frac{\partial^2 E_S}{\partial t^2} = \frac{4\pi \partial^2 P_{23}}{c^2 \partial t^2} \quad (24)$$

where the Stokes field E_S has been substituted for E , the polarization from Eq.21 has been substituted for P_{NL} , and a transformation to Gaussian units³⁸ has been made.

Equations 18 and 24 are sufficient to describe stimulated Raman scattering, though Eq.24 is more commonly written as a first order differential equation and in a slightly different form. This transformation will be derived next.

Considering propagation in the z direction only, the Stokes field can be written

$$E_S = \frac{1}{2} (E_S \exp^{i\Phi_S} + \text{c.c.}) \quad (25)$$

where $\Phi_S = \omega_S t - k_S z$ and E_S is complex with a slowly varying phase. Substituting this field into the left side of Eq.24 yields

$$-ik_S \exp^{i\Phi_S} \bar{D} E_S + \text{c.c.} \quad (26)$$

where the Slowly Varying Envelope Approximation³⁶ (SVEA) has been evoked, and D is the differential operator defined by

$$\bar{D} = \frac{\partial}{\partial z} + \frac{1}{v} \frac{\partial}{\partial t} \quad (27)$$

Writing out the complex exponential as a real and imaginary term, and letting $\alpha = D E$ and $\alpha^* = D E^*$, the expression given by

26 can be rewritten

$$-ik_S[(a-a^*)\cos\Phi_S + i(a+a^*)\sin\Phi_S] . \quad (28)$$

Similarly, the right side of Eq.24 can be written

$$- \frac{2\pi N d_{23} \omega_S^2}{c^2 \Delta_L} [(\beta+\beta^*)\cos\Phi_S + i(\beta-\beta^*)\sin\Phi_S] \quad (29)$$

where $\beta = \Omega_{12} Q^*$, $\beta^* = \Omega_{12}^* Q$, and SVEA was again invoked. But now note, for any complex number $\gamma = x + iy$,

$$x = \text{Re } \gamma = \frac{\gamma + \gamma^*}{2} \quad \text{and} \quad y = \text{Im } \gamma = \frac{\gamma - \gamma^*}{i2} \quad (30)$$

so that

$$\begin{aligned} \text{Im } \bar{D} E \cos\Phi_S + \text{Re } \bar{D} E \sin\Phi_S = & \frac{-N 2 \pi \omega_S v_S d_{23}}{c^2 \Delta_L} (\text{Re } \Omega_{12} Q^* \cos\Phi_S \\ & - \text{Im } \Omega_{12} Q^* \sin\Phi_S). \end{aligned} \quad (31)$$

Using the results of Eq.30, Eq.31 can be written as

$$\bar{D} E = -ik_2 Q^* E_L \quad \text{with } k_2 = \frac{N \pi \Omega \omega_S v_S d_{12} d_{23}}{\hbar c^2 \Delta_L}. \quad (32)$$

Eq.32 is the complete wave equation for the Stokes field. It tells us that spatial and temporal changes in the propagation of the Stokes field are driven both by the coherence and the pump laser. It should be noted that an equivalent expression to Eq.32 can also be written for the pump field, but this is of no interest in the present work, which is concerned only with the evolution of the Stokes field.

In summary then, the two equations, 18 and 32, give a complete description of the problem. They describe, as

promised, the response of the medium to the applied fields and the response of the fields to the medium.

CHAPTER THREE

ACCOUTERMENTS

Equipment and Technique

Spatial profiles and interference patterns, discussed in the next section of this chapter, were digitized using an EG&G Reticon "G" series self-scanning linear photodiode array. The array uses 1024 photodiodes on 25 μ m centers. The arrays are typically encased by a glass window. Since glass strongly attenuates the UV and adds the additional complication of interference from its two surfaces, the arrays used in these experiments were specially ordered without windows.

The array collects light continuously and is cleared by a scan. Each scan is triggered by a read pulse and takes 4ms to complete. In practice, the array is triggered by an external source with a repetition rate of approximately .5Hz. To reduce the background arising from stray room light, the array is read twice. At the onset of the read pulse the array scans once which clears it and after a .2ms wait, scans again. The second scan is read either with a storage oscilloscope or with digitizing electronics which send the information directly to a computer for storage. When using the photodiode array to measure spatial profiles

of the excimer laser, a 4.2ms delay is introduced between the trigger and the laser to insure that the second scan begins after the arrival of the laser pulse.

Usually, the spatial profiles of interest are inordinately small in diameter and in utterly inaccessible locations e.g. inside of a Raman cell. In these not so special cases, an imaging technique developed by Dr. Carlsten was used. An $f=10\text{cm}$ focal length lens is inserted in the beam, 12cm from the location of interest and the photodiode array is located at the image distance, 60cm from the lens. The photodiode array "sees" a magnified image of the beam 72cm from the point of interest.

An experimental measurement of the magnification was made using the apparatus shown in Fig.3. A 1mm aperture was placed in the attenuated laser beam which had been clipped "upstream" with a 4.5mm aperture to obtain a diffraction limited beam, as discussed in the next section. The photodiode array was initially placed immediately in front of the aperture and a measurement was made to confirm that the diameter of the aperture was 1mm. Next, the $f=10\text{cm}$ lens was placed in the system and the aperture was imaged onto the array where another measurement was made. A digitized plot of the measured results are shown in figures 4a for the actual aperture and 4b for the image of the aperture. One calculates the magnification by simply taking the ratio of the image to object distance, which for this case is 5. The

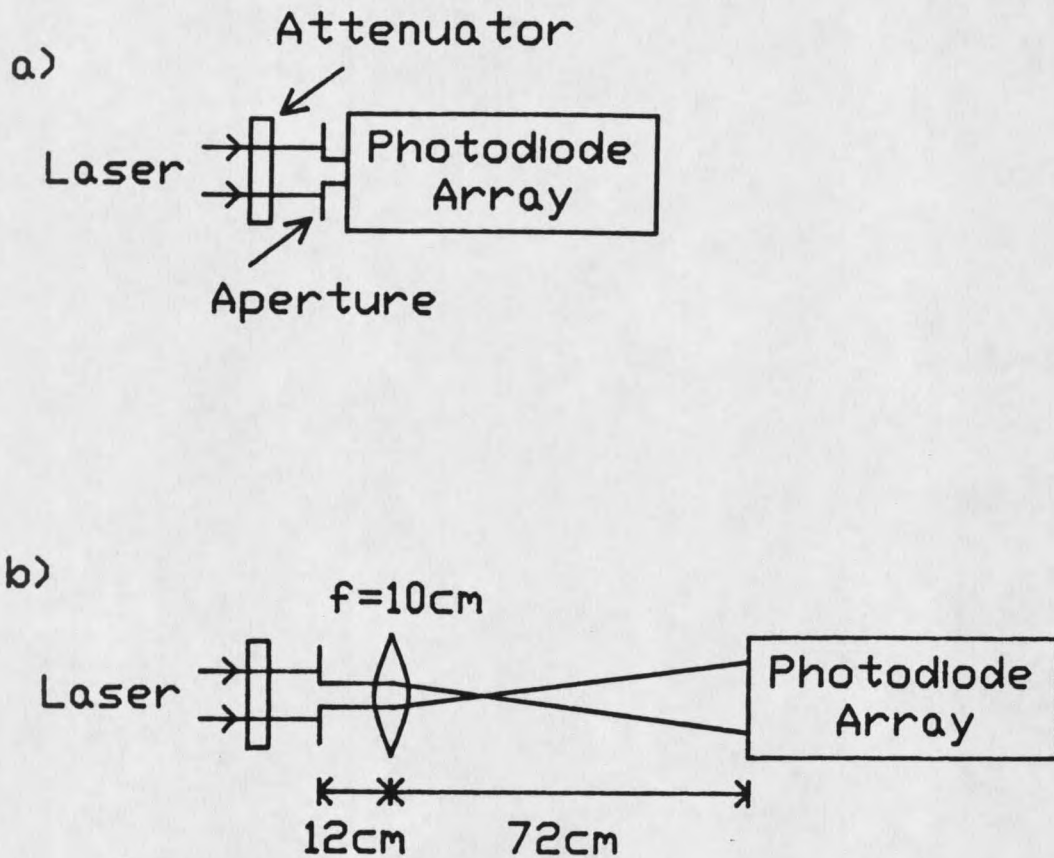


Figure 3. Schematic of experiment to measure the magnification of the $f=10\text{cm}$ lens. The laser was attenuated with an appropriate dielectric filter and then clipped with a 1mm aperture. The aperture was first measured (a) with a photodiode array, and then (b) the image of the aperture was projected onto the array.

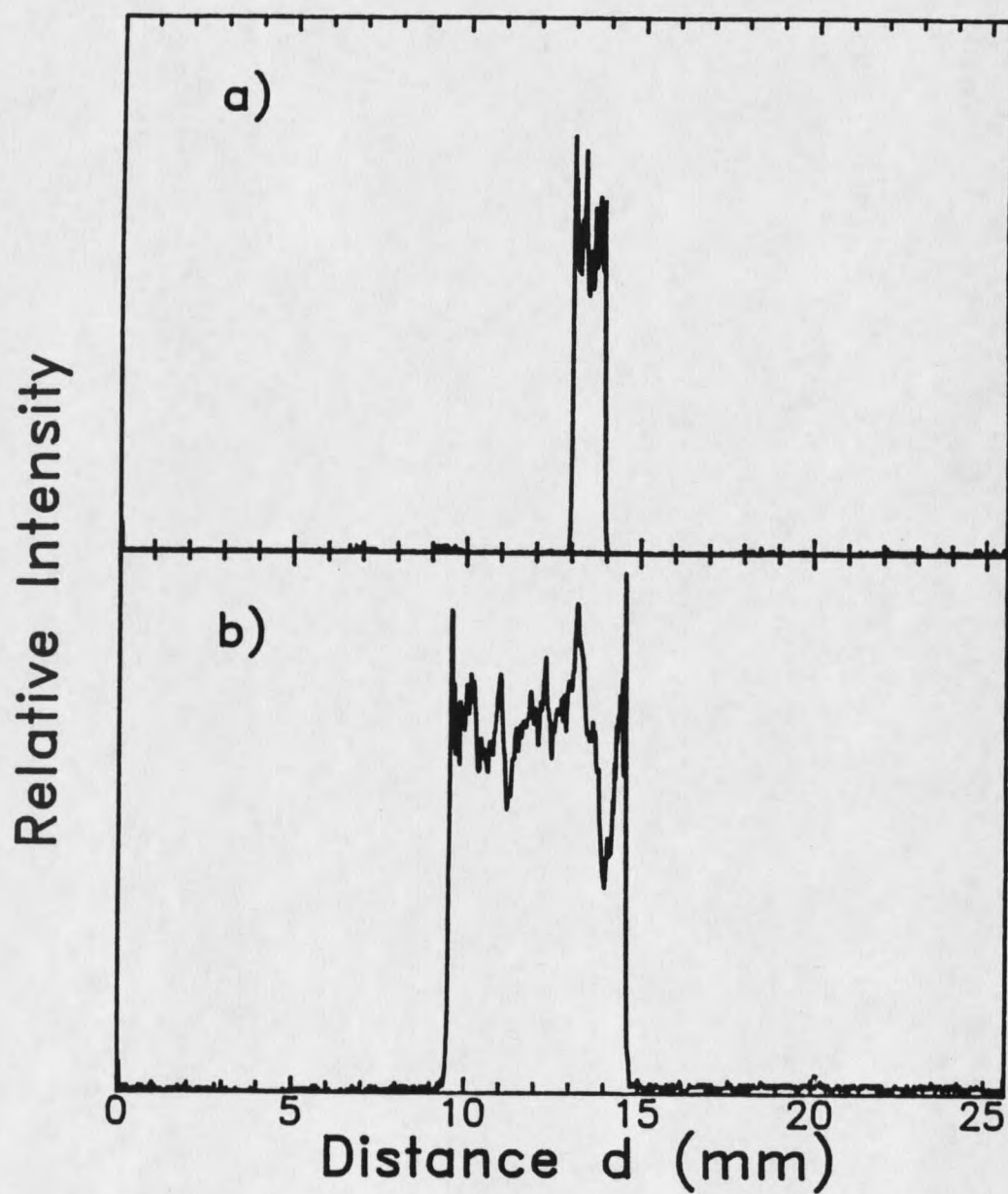


Figure 4. Digitized plot of (a) the aperture measured with the photodiode array and (b) the image of the aperture measured with the array.

experimentally measured value was 5.13.

Energy measurements were made with a Laser Precision Rj-7200 energy ratiometer, RjP-700 energy probe, and RjI serial to parallel interface. An HP-85 computer was interfaced to the forementioned electronics which were used in combination to collect the data.

In the next section, several plots of laser lines are shown. These lines were resolved with a Bausch and Lomb spectrograph which had a second order dispersion of 7.5 Å/mm and second order spectral range of 1850-3700 angstroms. A photodiode array was placed at the output of the spectrograph and monitored with a Tektronix 2230 100MHz digital storage oscilloscope. The spectrograph, though inadequate as a device to accurately measure the width of each laser line, was immensely useful as a monitor of changes in linewidth during the course of an experiment.

The Laser

The laser used in this thesis work was an injection locked Lambda Physik EMG-150ET, which is a pulsed, gas discharge laser. A schematic of the laser is shown in Fig.5a. The laser uses an oscillator-amplifier configuration. The oscillator light, after passing through a number of intra-cavity Brewsters prisms becomes significantly narrowed and polarized to better than 90% in the horizontal. The light, after leaving the oscillator,

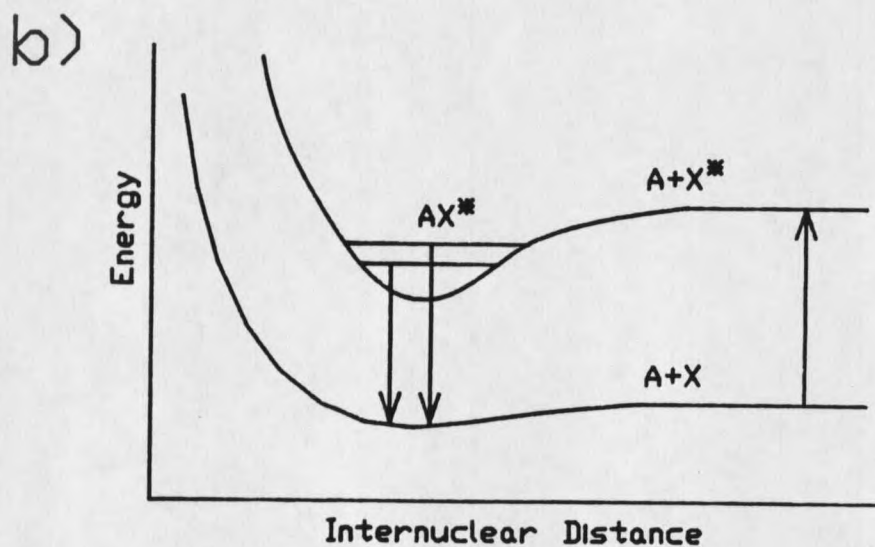
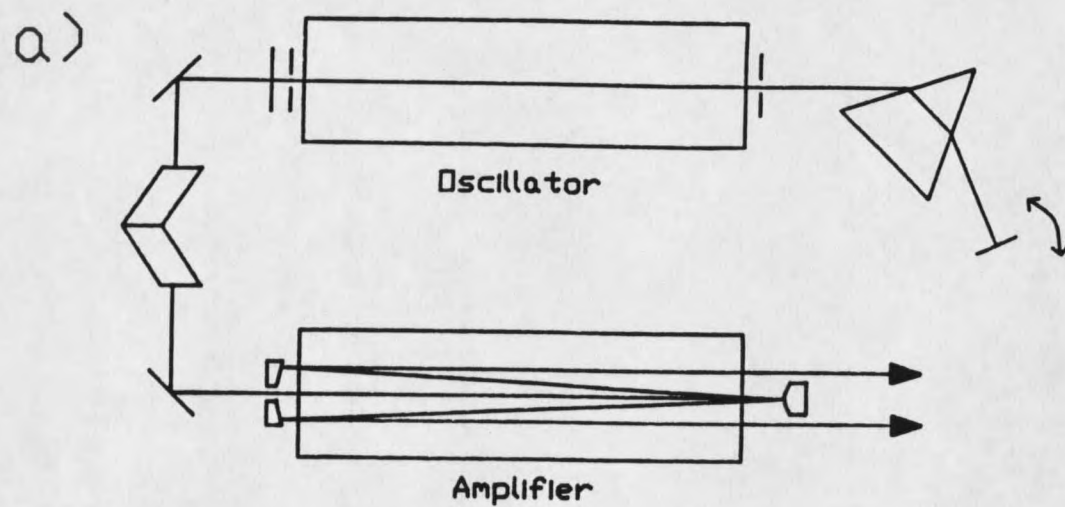


Figure 5. (a) schematic for the XeCl laser and (b) energy level diagram for the XeCl bond.

passes through two Fresnel rhombs which rotate the polarization from the horizontal into the vertical and then the light is injected into the amplifier, a three pass unstable resonator. The amplifier light becomes locked into the same vertical state of polarization as the injected oscillator light with an efficiency of better than 80%.

The laser's active medium, a gas, consists of the halogen gas, chlorine and the noble gas, xenon. Since xenon is inert, the ground state for the laser is repulsive and the molecules bond only in the excited state forming excimers³⁹ (molecules which exist only in the excited state). As is shown in Fig.5b, lasing only takes place on the transition between bound and unbound states with the ground state always empty. Thus, the excimer laser is considered to be very efficient.

According to the Lambda Physik catalog⁴⁰, the laser spectrum consists of four lines, two strong lines at 307.9nm and 308.15nm, and two weak lines at 307.6nm and 308.56nm, along with a weak subsidiary line at 308.2nm. Shown in Fig.6 is a plot of the digitized output from the spectrograph described in the previous section. As is shown in Fig.6a, we typically see only the 307.9nm, 308.1nm and 308.2nm lines. By tuning in the horizontal the rear mirror of the oscillator cavity, we are able to select to a certain degree which frequency light returns to the Brewster prisms at the appropriate angle for maximum transmission. Thus we

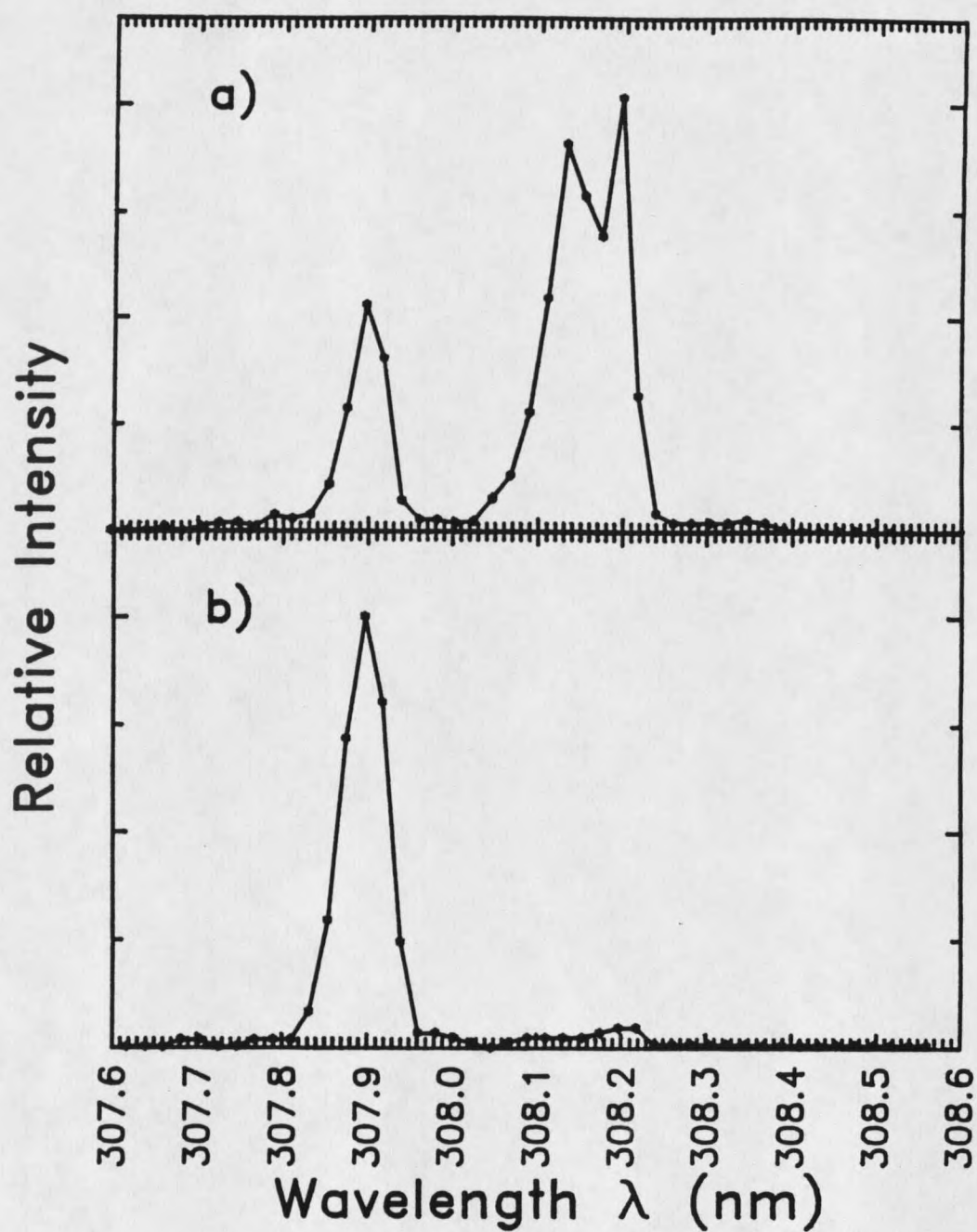


Figure 6. Digitized plot from the spectrograph showing (a) the three lines of the XeCl laser and (b) the result of tuning the laser on the 307.9nm line.

are able to tune between the three lines. We chose to tune the laser on the 307.9 line and as is shown in Fig.6b, in doing so, suppress the other two.

The temporal profile of the laser was measured with a Hamamatsu R1193U-02 phototube and a Tectronix 7834 500MHz storage oscilloscope. A digitized plot of the temporal profile is shown in Fig.7. By approximating the pulse as rectangular with a height equal to the peak power and an area equal to the total energy, the pulse width was calculated and equal to 15.3ns.

A measurement was also made of the laser linewidth. To make this measurement, a Mach-Zehnder interferometer shown schematically in Fig.8 was constructed. The path length of one beam was varied relative to the other using a three mirror corner-cube mounted on a linear motor drive which was originally constructed for the gain enhancement experiment discussed in chapter 6.

The output of the interferometer, was a series of interference profiles or interferograms. Each interferogram was for a different path length difference and was recorded with a linear photodiode array. An example of an interferogram is shown by the solid line in Fig.9.

The interference fringes are described by the irradiance function⁴¹ which is defined as

$$I(x) = I_1 + I_2 + 2(I_1 I_2)^{1/2} \text{Re} \gamma(x). \quad (33)$$

$\gamma(x)$ is the normalized autocorrelation function and is

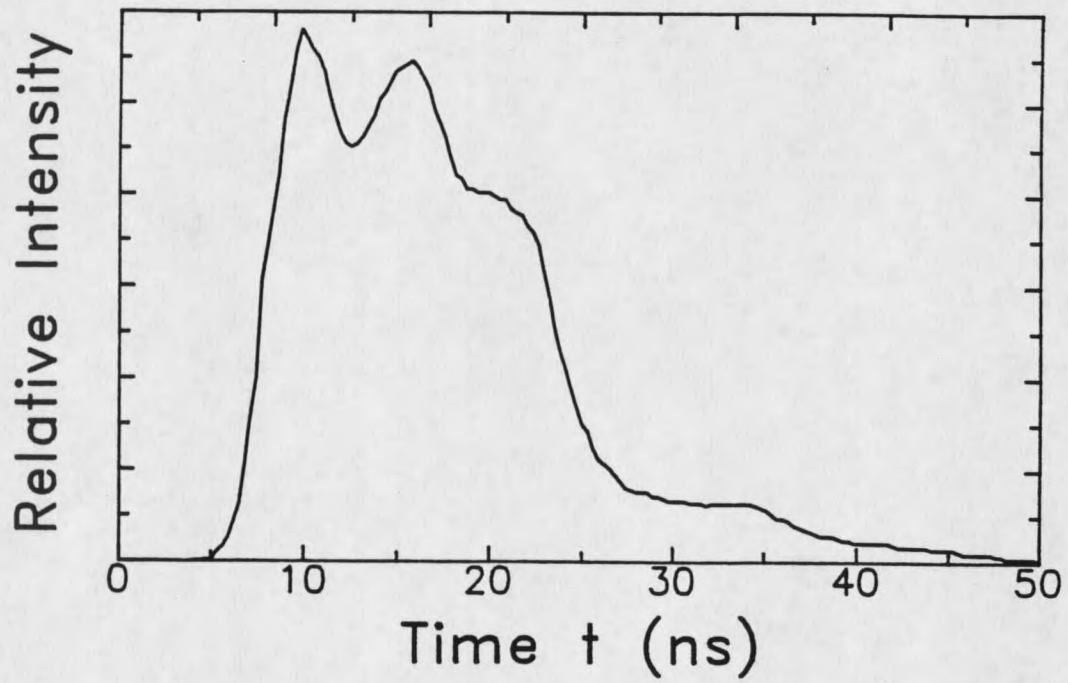


Figure 7. Temporal profile of a typical laser pulse.

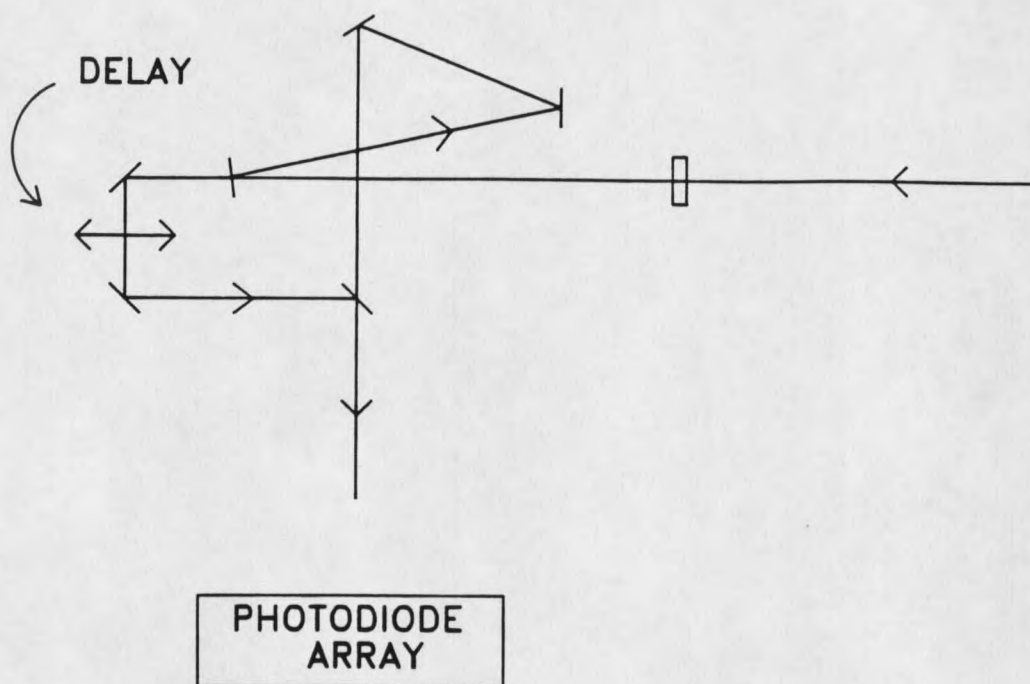


Figure 8. Schematic of the Mach-Zehnder interferometer used to measure the linewidths of the input Stokes and pump.

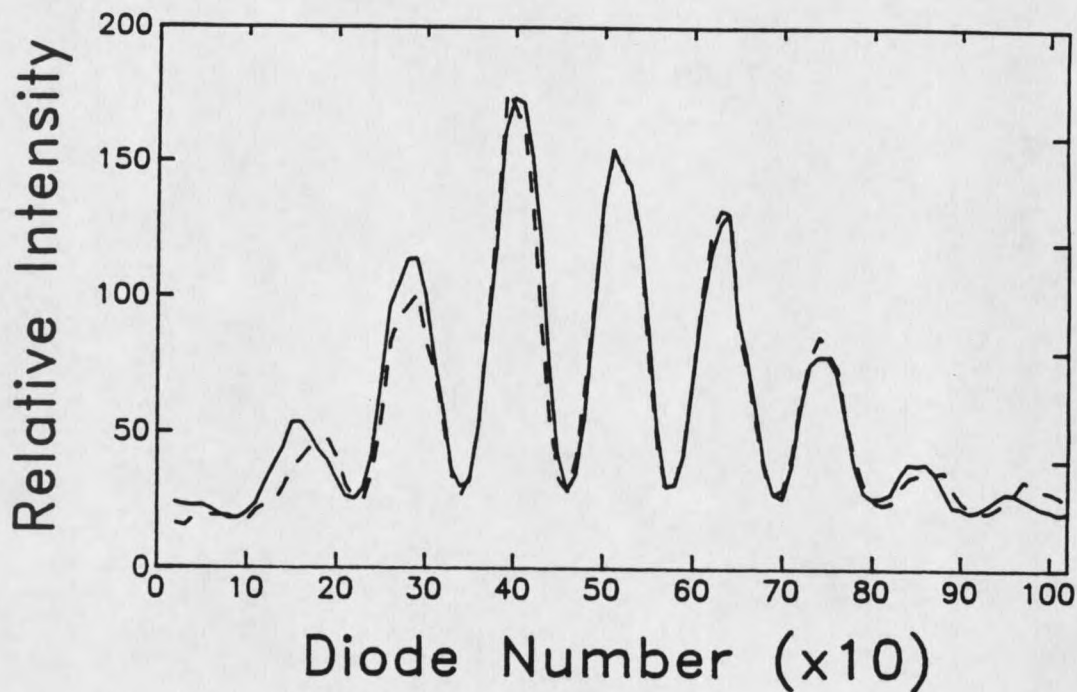


Figure 9. Interference profile generated with the Mach-Zehnder interferometer. The solid line is the actual data and the dashed line is a least squares fit using eqn.33. For this data $|\gamma|$ was .91.

given by

$$\gamma_{12}(x) = \frac{\langle E_1(x) E_2(x+\Delta x) \rangle}{(I_1 I_2)^{1/2}}$$

so that

$$\text{Re} \gamma_{12}(x) = |\gamma_{12}| \cos(\alpha x - \phi) \quad (34)$$

where α is the periodicity of the irradiance function $I(x)$ and ϕ is the phase.

A least squares fit was made to each interference profile using equations 32 and 33 with $|\gamma(x)|$, α , and ϕ as the fitting parameters. A typical fit is shown as the dashed line in Fig.9. A plot was then made of the magnitude of γ as a function of path length difference and is given by the X's in Fig.10b. The auto correlation function and the power spectrum form a Fourier transform pair, thus fitting $|\gamma|$ with an exponential as shown by the solid line in Fig.11b, results in a Lorentzian spectral distribution. The exponential fit had a half width at $1/e^2$ of 6.9mm which corresponds to $.47\text{cm}^{-1}$ HWHM for the Lorentzian frequency distribution. The coherence length for the laser can also be obtained from Fig.10b and is given by the HWHM at $1/e$ which is 3.5mm or 23.5ps.

The length of the oscillator cavity of the laser was measured to be 1.43m which corresponds to a longitudinal mode spacing δ , of $.0035\text{cm}^{-1}$. Thus there are 134 longitudinal laser modes out to the point at which the Lorentzian frequency distribution has fallen in intensity by

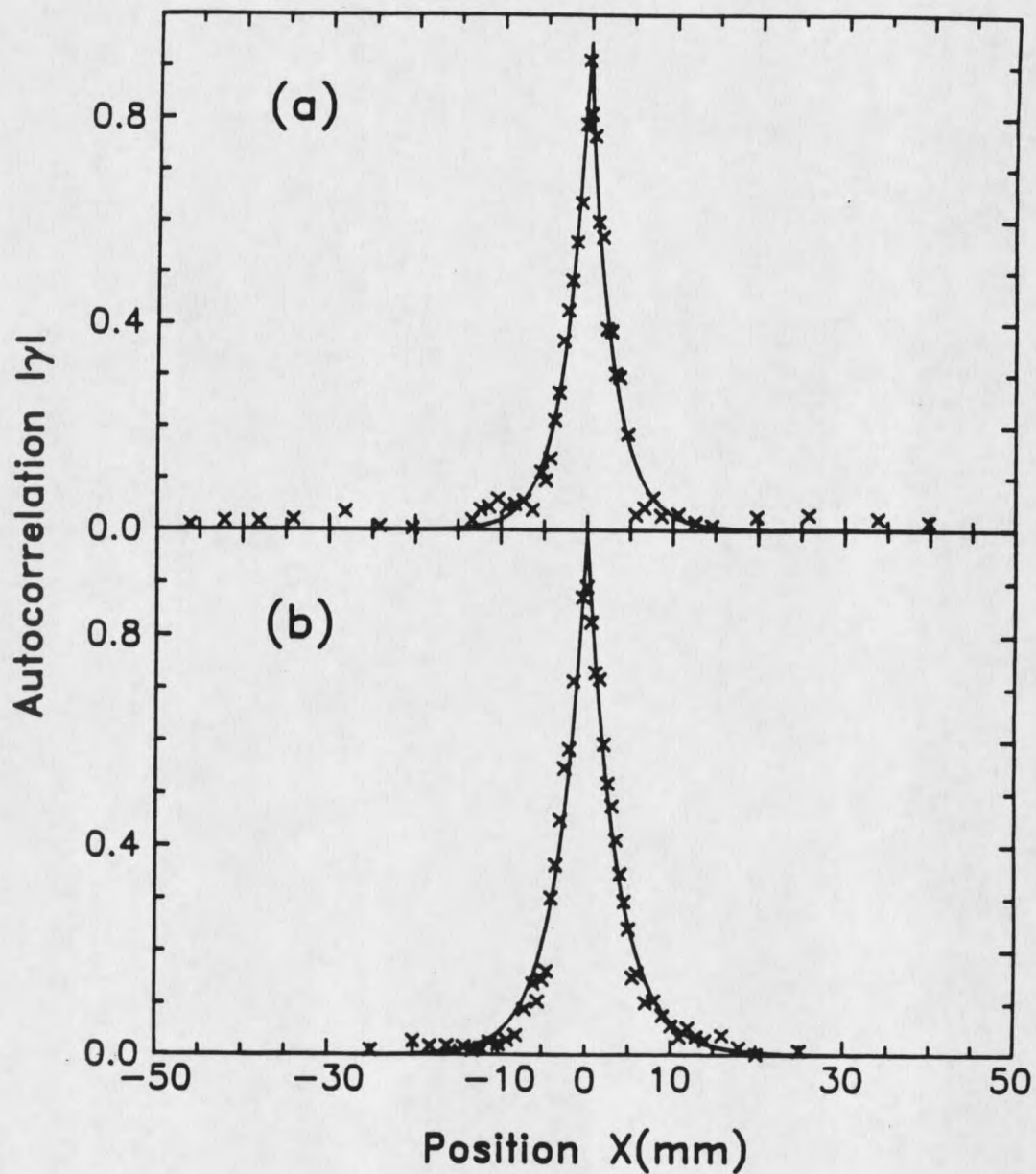


Figure 10. Magnitude of the normalized autocorrelation function γ , for (a) the Stokes beam and (b) the laser beam. The X's represent specific values of $|\gamma|$ for different interferometer path length differences and were generated as parameters from fitting a series of interference profiles similar to Fig.9. The solid line is an exponential fit to $|\gamma|$.

50%.

In all of the experimental work performed for this thesis, the laser was clipped with a 4.5mm aperture resulting in a flat tophat spatial distribution. The laser operating in this configuration is near diffraction limited, where a diffraction limited beam is defined as one which is a plane wave at focus. A simple check for diffraction limited operation can be made by making use of the fact that the image focal plane for a lens is the Fourier transform of the object focal plane⁴².

To determine diffraction limited operation⁴³, the apparatus in Fig.11 was used. A lens, L1, of focal length 198.6 cm was inserted 3cm from the aperture. The profile at focus was imaged with a second lens, L2, onto a linear photodiode array. The resulting pattern is shown by the solid line in Fig.12. The Fourier transform of a flat tophat spatial distribution is given by the Airy pattern⁴²

$$I(r) = I_0 \frac{[2J_1(\xi r)]^2}{(\xi r)^2}$$

where $\xi = 2\pi r / \lambda f$ and I_0 is the peak intensity at focus. The dashed line in Fig.12 is the calculated profile. The two compare well which shows that the beam is near diffraction limited.

The Medium

The medium used for scattering in this thesis work was

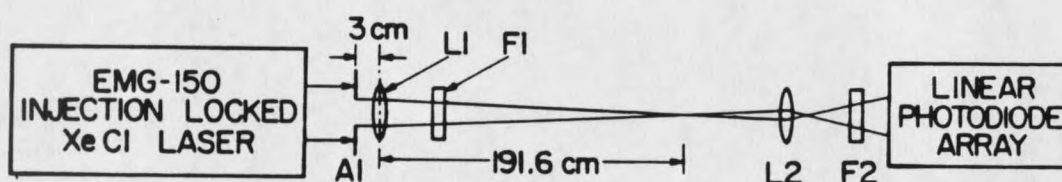


Figure 11. Experimental apparatus used to measure diffraction limited operation of the XeCl laser. The incident beam was clipped by aperture A1 and focused with a 191.6cm focal length lens L1. A second lens L2 was used for imaging the profile at focus onto a photodiode array. Filters F1 and F2 were used to attenuate the beam.

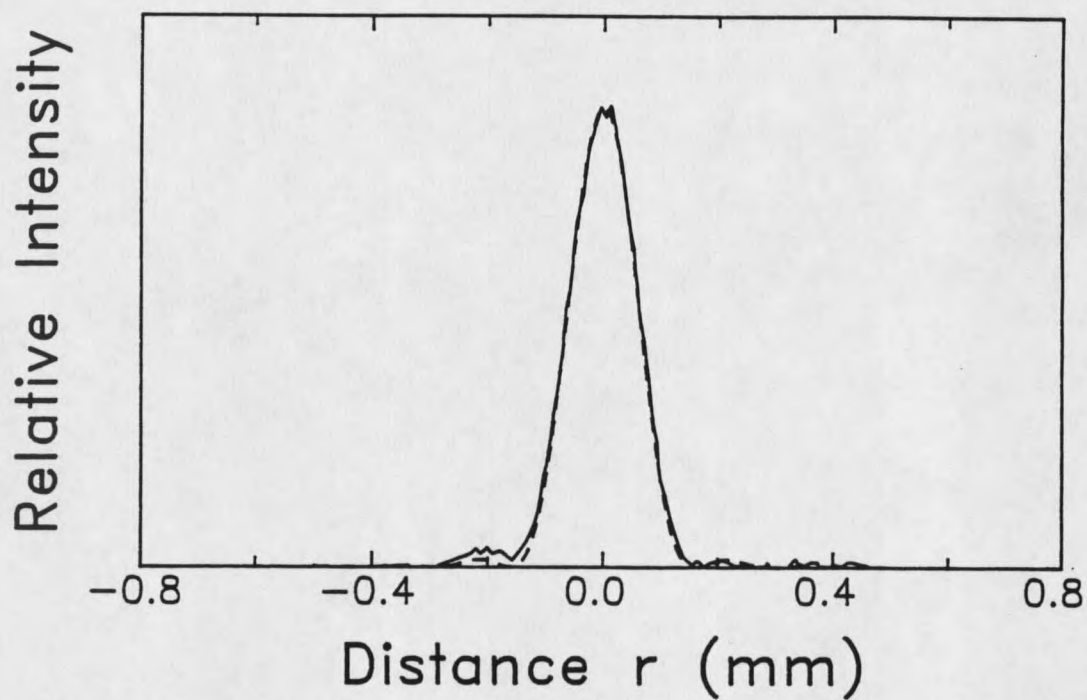


Figure 12. The solid line is the experimentally measured laser beam at focus and the dashed line is the calculated theoretical profile at focus for a diffraction limited beam.

molecular Hydrogen, H_2 . Molecular hydrogen is a diatomic homonuclear molecule⁴⁴ and has energy levels divisible into three groups or modes of motion⁴⁵. The primary energy levels are electronic states, with each electronic state divisible into approximately equal groups of energy levels corresponding to vibrational states. Each vibrational level in turn exhibits a fine structure attributable to rotational states⁴⁶. An energy level diagram for the H_2 is shown in Fig.13.

The quantum numbers associated with these levels are n , v , J , and m which are identified respectively as electronic and vibrational quantum numbers; and rotational angular momentum and it's projection along an internuclear axis¹³. The selection rules for two photon Raman scattering are $\Delta v=0$, $\Delta J=\pm 2$, or $\Delta v=\pm 1$ and $\Delta J=0, \pm 2$. The branches of the $\Delta J=-2, 0, 2$ are labeled O, Q, and S, respectively⁴⁷. The selection rules for v come from perturbation theory for dipole radiation while the selection rules for J arise from the fact that the H_2 absorbs two photons which each have a spin of ± 1 .

At room temperature the separation between vibrational levels is large compared to kT so that all of the molecules exist in the vibrational ground state. The rotational states however are separated by energies which are comparable to kT and the molecules have a distribution of rotational energies with approximately 66% of the population

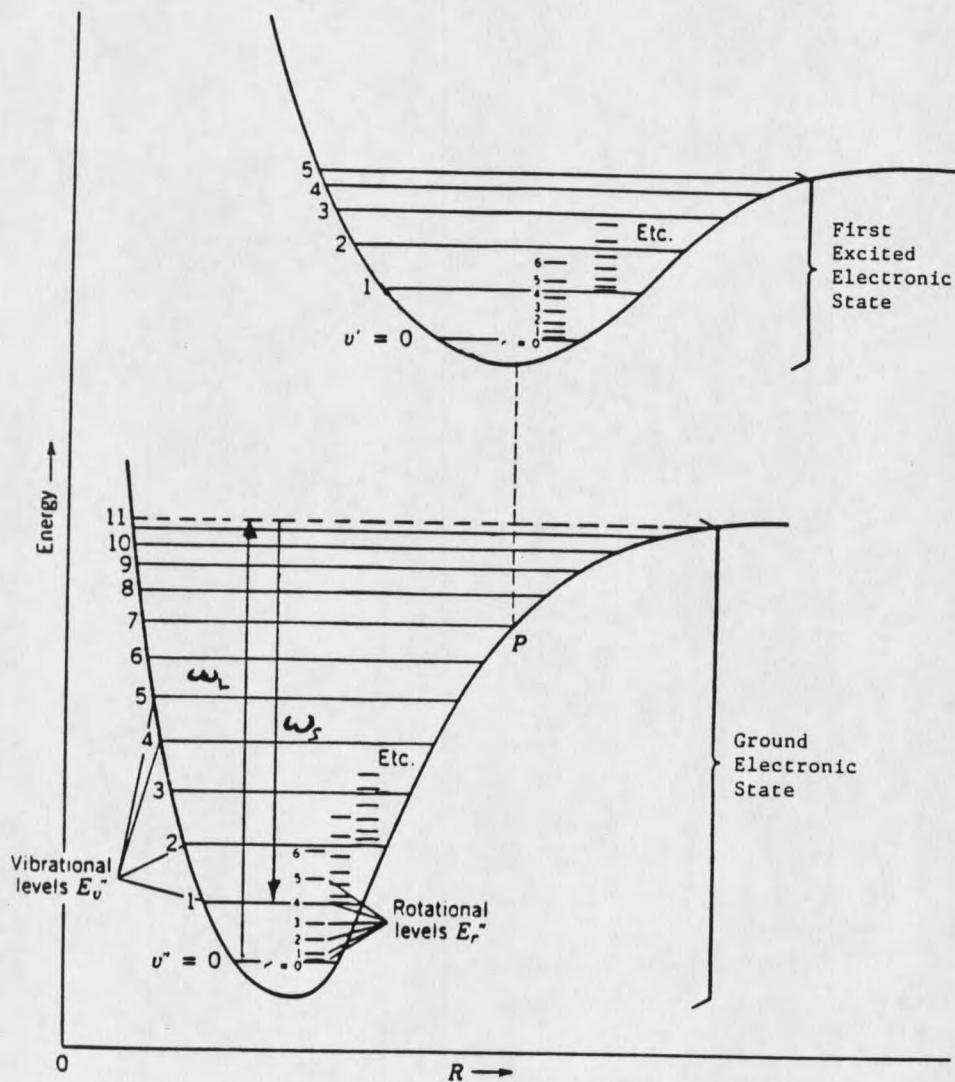


Figure 13. Energy level diagram for H_2 (adapted from Eisberg and Resnick, p.430).

in the $J=1$ level⁴⁷.

The scattering for this thesis work is from the $Q_{01}^{(1)}$ vibrational transition. With this notation the initial rotational level is given in the superscripted parenthesis, while the initial and final vibrational levels are given by the subscripts.

CHAPTER FOUR

THEORY

Physics

In this section the equations appropriate for a multimode, fixed-random-phase, broadband laser are described. This model is a straightforward extension of the coupled equations of transient stimulated Raman scattering derived in chapter 2. These equations, written in the travelling frame (see appendix B) are

$$\begin{aligned} d/dz E_S &= -ik_2 Q^* E_L \\ d/d\tau Q^* &= -\Gamma Q^* + ik_1 E_L^* E_S. \end{aligned} \quad (35)$$

These expressions, as previously mentioned, describe the transient growth of the Stokes field and coherence of the Raman transition for a medium of three level molecules interacting with two fields, a pump, E_L , and a Stokes, E_S , which differ in frequency by the Raman shift of the medium. The retarded time τ measures time in a reference frame traveling with the laser pulse and is given by $\tau = t - z/c$, where z is a measure of position in the Raman medium. Q^* is related to the off diagonal density matrix elements and can be interpreted as the coherence of the transition between the initial and final levels. Γ is the Raman linewidth

(HWHM) arising from collisional dephasing in the medium, and k_1 and k_2 are constants given by equations 18 and 32.

For simplicity, it is assumed that dispersion and pump depletion can be ignored. Additionally, we include a delay τ_D which corresponds physically to an optical delay between the Stokes and pump fields. The solution of equations 35 is given by³

$$E_S(z, \tau + \tau_D) = E_S(0, \tau + \tau_D) + (k_1 k_2 z)^{1/2} \int_0^\tau \frac{\exp[-\Gamma(\tau - \tau')]}{[p(\tau) - p(\tau')]^{1/2}} \times I_1(\{4k_1 k_2 z [p(\tau)p(\tau')]\}^{1/2}) E_L(\tau) E_L^*(\tau') E_S(0, \tau' + \tau_D) d\tau'. \quad (36)$$

The constant $(k_1 k_2 z)^{1/2}$ is related to the exponential gain G , for a narrow band laser with $G = 2E_L E_L^* k_1 k_2 z / \Gamma$ and will be discussed in the next section. $p(\tau)$ is the energy in the laser pulse up to the time τ , and I_1 is the modified Bessel function of order one⁴⁹.

To extend the theory using a multimode, fixed-random-phase model, the input pump and input Stokes fields are taken as a sum over longitudinal modes with fixed but random phases. The fields are given by

$$E_L(\tau) = \sum_n E_{Ln} \exp^{i(n\delta\tau + \phi_{Ln})}$$

and

$$E_S(0, \tau) = \sum_m E_{Sm}(0) \exp^{i(m\delta\tau + \phi_{Sm}(0))} \quad (37)$$

where the input field amplitudes E_{Ln} and $E_{Sm}(0)$ are fixed and real. It is assumed that the mode spacing, δ , is identical for both the pump and the Stokes fields, as are the distributions of the initial random phases, ϕ_{Ln} and

$\phi_{Sm}(0)$. The appropriateness of these assumptions requires consideration but for the purposes of this work, their validity is justified by the success of the model.

Computer Model

The computer programs are printed in their entirety in appendix C. The first program `Lfields` calculates the laser field and input Stokes field, integrated laser energy $p(\tau)$, and the difference $p(\tau) - p(\tau')$.

The code is very straightforward. First, the relevant parameters are loaded e.g. the halfwidth of the Lorentzian frequency distribution, pulse length, pulse resolution in time, and number of modes. Next, each mode is assigned a random phase, weighted with a Lorentzian distribution in intensity, and normalized.

Though it is not obvious from the code, the following is actually accomplished. Consider two fields, one broadband and one narrowband, and each normalized to the same energy. The amplitude of each broadband mode can be written as some function of the narrowband amplitude, so that

$$bb_A^2 = nb_A^2 (B_i^2) \quad (38)$$

where nb_A is the amplitude of a narrowband mode and equal to a constant and B_i gives the Lorentzian distribution with

$$B_i = \frac{C}{\left(1 + \frac{(k_0 - k_i)^2}{\Gamma_L^2}\right)^{1/2}} \quad (39)$$

where C is a normalization constant and Γ_L is the bandwidth HWHM of the laser. Each laser mode has an amplitude and a phase so that $E_i(t) = C A_i \exp^{i\Phi_i}$, where $\Phi_i = \omega_i t - k_i z + \phi_i$. The total field (i.e. the superposition of all the modes) then, can be written

$$E(t) = \sum_i E_i(t) = C \sum_i A_i \exp^{i\Phi_i}. \quad (40)$$

The multimode laser field is thus written in terms of the monochromatic field.

Finally, `Lfields` calculates the integrated laser energy $p(t)$ at each instant of time (with the input time resolution) over the pulse length along with the difference $p(t) - p(\tau)$. A relatively short pulse length of 4.685ns was chosen to minimize the calculation time.

The program `Mode` calculates gain enhancement as a function of input pump energy. It will calculate a delay profile for a fixed input pump energy as shown by the dashed line in Fig.14, or a table of correlated and uncorelated gains for varying pump energy as shown by the solid line in Fig.15. Though it is essentially nothing more than a straightforward numerical integration of Eq.36 using the trapezoidal integration method⁴⁹, it involves many nested loops over many different array elements. In an effort to optimize the algorithm, much of the code became necessarily obscure.

A simplified flow chart for the program showing critical decisions and paths is shown in Fig.16. `Mode` begins by

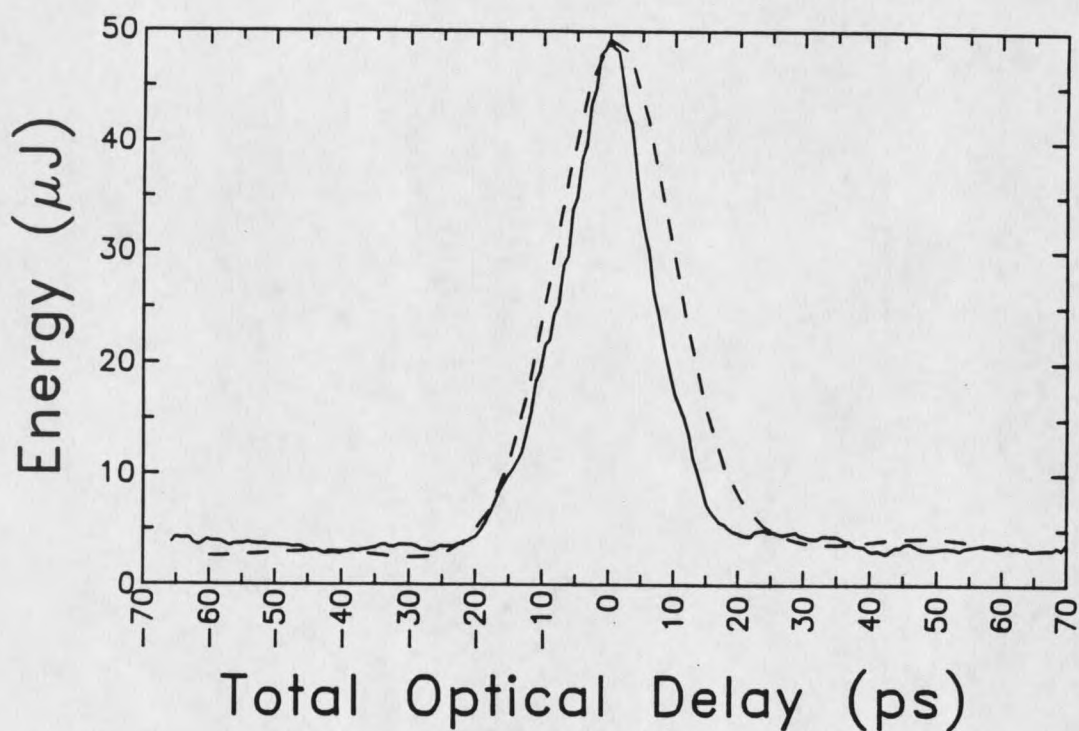


Figure 14. Comparison of the experimental (solid line) and theoretical (dashed line) values of the gain enhancement as a function of optical delay. The energy of the injected Stokes and input pump was $.43\mu\text{J}$ and $.42\text{mJ}$, respectively.

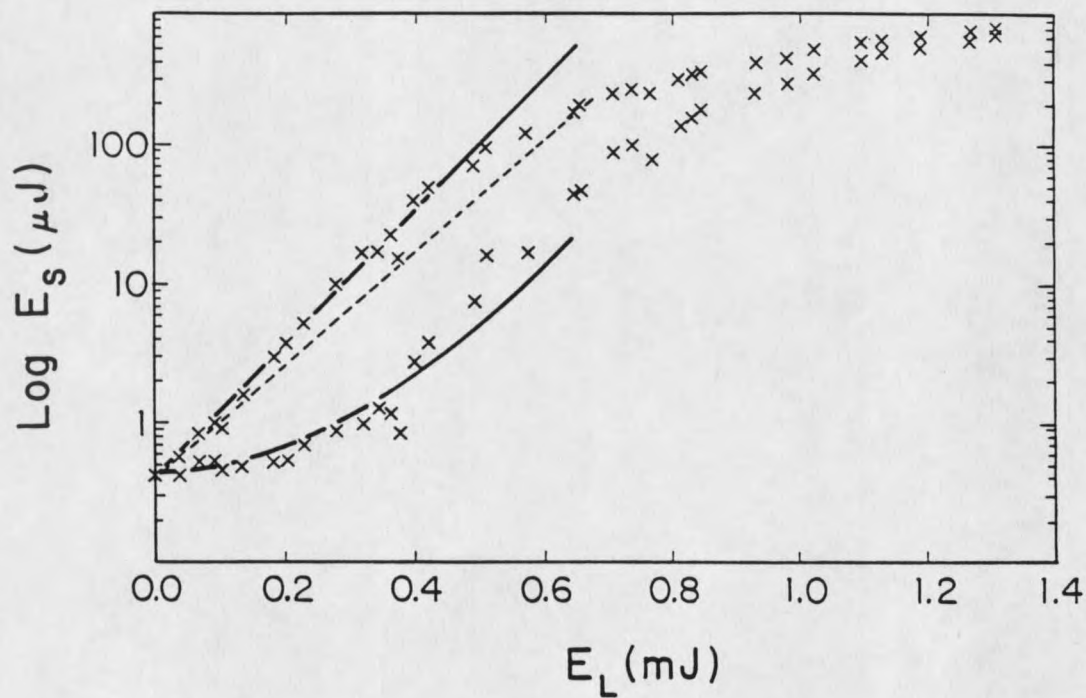


Figure 15. Correlated and uncorrelated gain enhancement as a function of input pump energy. Superimposed on the data (the X's) is the theoretical result (solid line). Also shown is the monochromatic gain (dashed line).

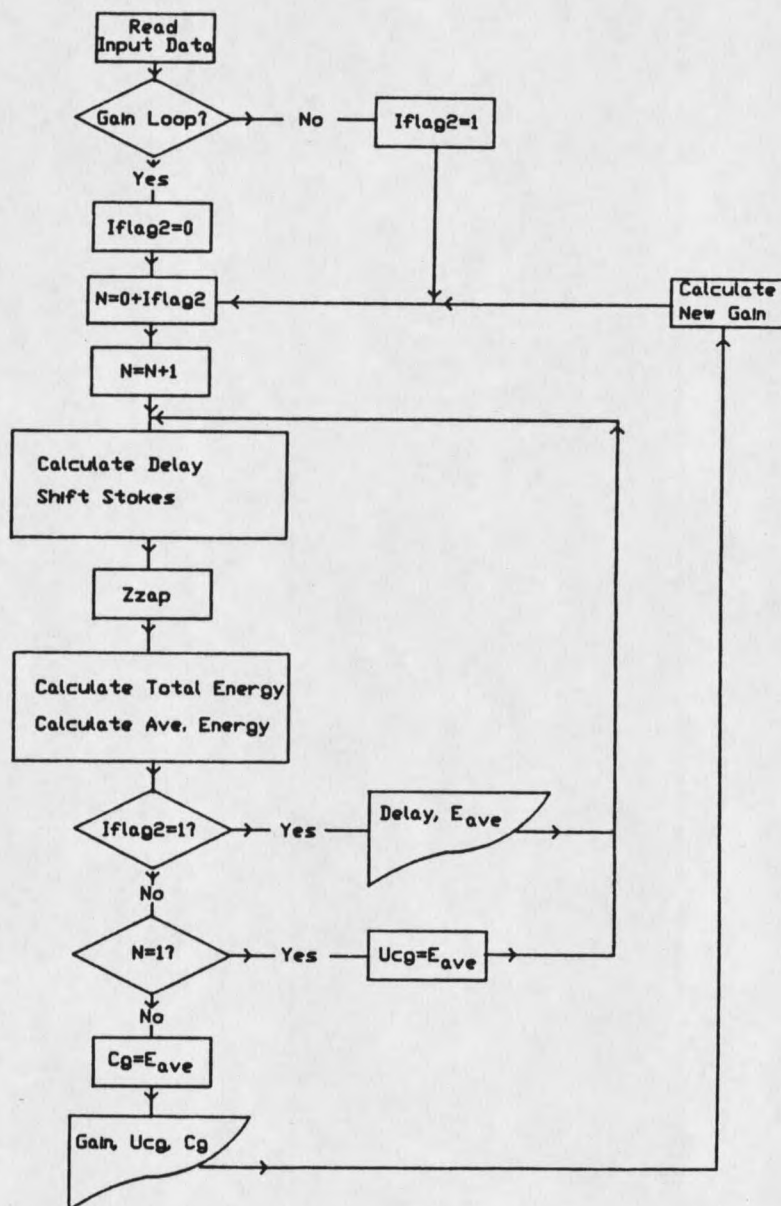


Figure 16. Flow chart for the program Mode used to calculate the theoretical predictions for gain enhancement as a function of optical delay.

reading the previously calculated values of the laser field $E_L(\tau)$, input Stokes field $E_S(\tau)$, integrated laser energy $p(\tau)$, and the difference $p(\tau)-p(\tau')$.

Mode also reads the results of another program in which the Bessel function $I_1()$ is calculated using a series approximation given in reference 48. Values are calculated over a range appropriate for solving the integral in **MODE** and the results are stored in an array called **bdata**. This program also calculates the slope between the elements of **bdata** and stores these values in an array called **bdif**. With the function call **Fbessel**, **MODE** uses these results, the method of linear interpolation, and the slopes stored in **bdif** to calculate Bessel function values which lie between the array elements of **bdata**.

Next **Mode** sets the flag **Iflag2** which is later used to determine whether a delay profile or gain enhancement loop is in use. **Mode** then proceeds into the gain loop. In the gain enhancement experiment, gain was increased by increasing the laser intensity. The computer program accomplishes this by varying the constant **C1**. To see how this is done we rewrite Eq.36 using Eq.40

$$E_S(z, \tau + \tau_D) = E_S(0, \tau + \tau_D) + (k_1 k_2 z |^{nb} E|^2)^{1/2} E_L(\tau) \int_0^\tau \frac{\exp^{-\Gamma(\tau - \tau')}}{[p(\tau) - p(\tau')]^{1/2}} x \quad (41)$$

$$I_1(\{4k_1 k_2 z |^{nb} E|^2 [p(\tau) p(\tau')]\}^{1/2}) E_L^*(\tau') E_S(0, \tau' + \tau_D) d\tau.$$

where we have used the fact that $^{nb} A^2 = |^{nb} E|^2$. The constant

C_1 is related to the gain for a narrowband laser by

$$c_1 = \left(\frac{G\Gamma_R}{2} \right)^{1/2} = (k_1 k_2 z |^{nb} E|^2)^{1/2}.$$

By varying c_1 , we are able to vary the gain.

The optical delay is implemented by shifting the Stokes field with respect to the pump field. This means that time is measured relative to the beginning of the pump pulse. Note that in Eq.41, the Stokes field at time $\tau + \tau_D$ depends on the laser field during the time interval 0 to τ .

Physically, this means that during the time interval $0 < t < \tau_D$, when the laser pulse precedes the Stokes pulse, the medium "sees" no Stokes field and we have no Raman scattering, i.e. $E_S = 0$. Conversely, for $\tau - \tau_D < t < \tau$, when the laser pulse follows the Stokes pulse, again, no Stokes field is present, there can be no Raman scattering and $E_S = 0$. The shift takes place in the do loop to 2700 where IDELAY corresponds to τ_D .

Zzap is the subroutine which contains the algorithm for calculating the integral, while Bbambi is a subroutine which calculates the integrand. The variable Iflag1 in the subroutine Bbambi is used to halt the iteration when the exponential $\exp^{-\Gamma x}$ becomes inconsequentially small.

CHAPTER FIVE

EXPERIMENTS

Gain Enhancement

The experimental apparatus used to collect the gain enhancement data is shown in Fig.17. The laser is described in chapter three. Two Raman cells were used and both were filled with H_2 initially at 100 atmospheres and later for a second experiment at 10 atmospheres. One was 50cm in length and was used as a Raman generator and the other was 100cm in length and was used as a Raman amplifier. As in previous Raman generator studies⁴³, the laser beam was clipped with a 4.5mm aperture to obtain a diffraction limited beam (see chapter three). A beam splitter was used to obtain two beams. One was brought to focus in the center of the Raman generator cell with an $f=1m$ lens. The Stokes emission from the generator cell was collimated with another $f=1m$ lens, passed through an optical delay (a three mirror corner-cube mounted on a linear motor drive), attenuated, and finally combined with the other (pump) beam at a beam combiner. The other beam, the pump, was also used to monitor the laser linewidth, power, and energy. The pump intensity was varied with dielectric attenuators before it was combined with the Stokes beam. After the beam combiner, the two beams were

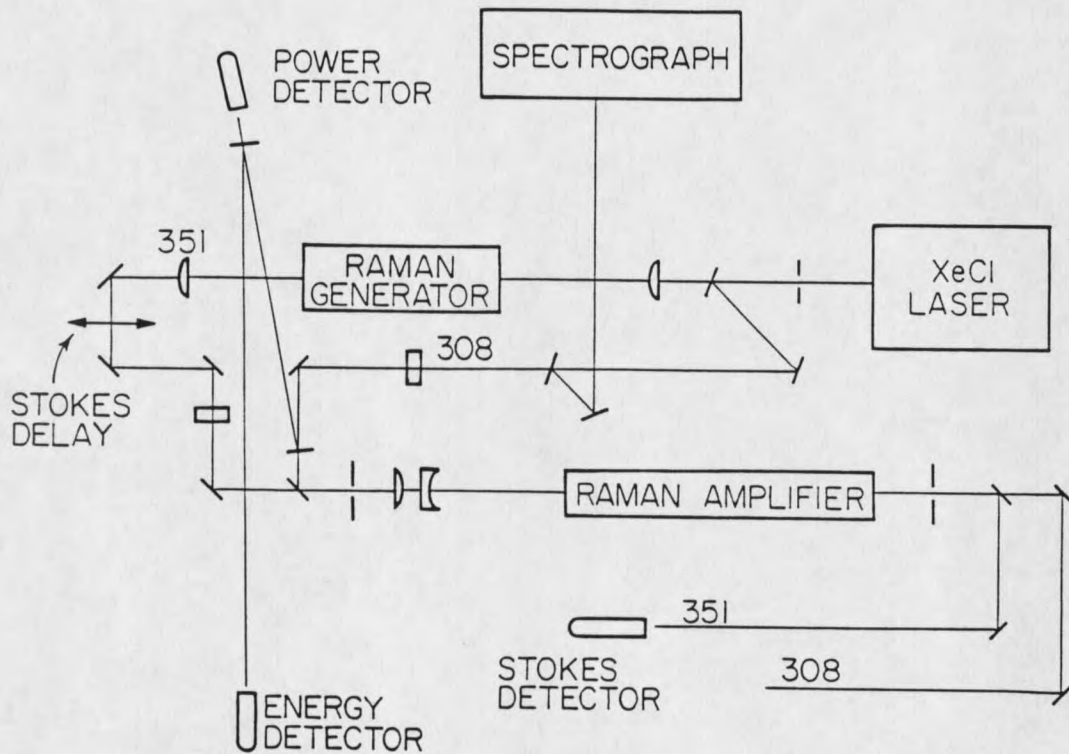


Figure 17. Schematic of the gain enhancement experiment.

made collinear by centering each beam in a 1.5mm aperture on each side of the Raman amplifier cell. To check that the beams were centered in the apertures, we examined the image of each beam in the respective aperture. A beam contractor was inserted to reduce the diameters of the two beams. An $f=500\text{mm}$ focal length lens was used to reduce the beam diameter and an $f=-250\text{mm}$ focal length lens was used to recollimate it, which is to say the optics were adjusted to give as uniform a profile as possible through the amplifier cell. The two beams were then injected into the amplifier.

A measurement of the Stokes linewidth was also made with the same Mach-Zehnder interferometer described in chapter 3. The optics were exchanged for optics coated at the appropriate wavelength for the Stokes but in all other respects the apparatus was identical as were the method of data collection and analysis.

Measurement of Beam Profiles

The spatial profiles of both the Stokes and pump were measured at the amplifier cell entrance, center and exit. The cell was first removed and then an $f=10\text{cm}$ focal length lens was used to image the desired location onto a linear photodiode array, using the imaging technique described in chapter 3.

Measurement of Zero of Optical Delay

An experiment was performed to measure exactly, the point at which the two legs of the gain enhancement apparatus were identical in length. The results of this experiment are discussed in chapter 6. A schematic of the experiment is shown in Fig.18. As can be seen from this figure there were no changes made from the gain enhancement experiment discussed above. As it happens, there is sufficient pump reflection from the surfaces of optics coated at the Stokes wavelength that the experimental apparatus could be used, unaltered, as another Mach-Zehnder interferometer. The pump traverses both legs of the apparatus and is combined at the beam combiner where the recombined beams were examined with a linear photodiode array 90 degrees from the previous direction of propagation. A measurement of the polarization through the corner-cube revealed that the polarization rotates by approximately 10%. A Glan-Thompson polarizer was inserted between the recombined beams and the photodiode array and was used in conjunction with attenuators allowing the beams to be fairly well matched in intensity. An attenuator was also inserted between the recombined beams and the photodiode array to block the Stokes radiation.

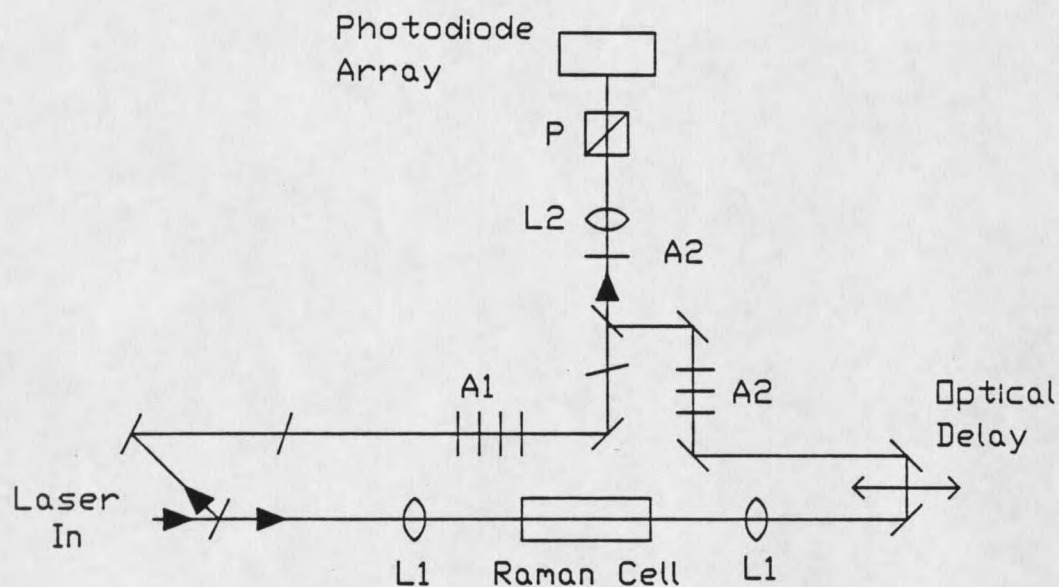


Figure 18. The gain enhancement experimental apparatus was used as a Mach-Zehnder interferometer to measure the zero of optical delay. The filters A2 were used to remove all light at 351nm. After the beams were recombined, they were projected with lens L2 onto a photodiode array. A polarizer P2 was used to vary the intensity of the two beams relative to one another.

CHAPTER SIX

EXPERIMENTAL RESULTS

General Results

Both the Stokes and pump beam profiles were measured with a linear photodiode array at the amplifier cell entrance, center, and exit. The measured spatial profiles are shown in figures 19 and 20. The waists were found to vary by $\pm 20\%$ through the cell. The profiles at cell center, figures 19b and 20b were fit to a gaussian profile, giving beam waists (half width at $1/e^2$) of .28mm for the Stokes and .33mm for the pump.

A pump energy of .42mJ and a pulselength of 15.3ns corresponds to an average power of 27.5kW. But the average power is also equal to the peak power density integrated over the cross-sectional area of the beam, that is

$$P_{ave} = I_0 \int_0^{\infty} \exp^{-2(r/w_0)^2} 2\pi r dr = \frac{1}{2} I_0 (\pi w_0^2).$$

This expression can be solved for the peak power density I_0 , which at cell center for a beam waist of .33mm, corresponds to 8.0MW/cm².

The linewidth of both the Stokes and pump were measured as described in chapter 3. In Fig.10a, the autocorrelation

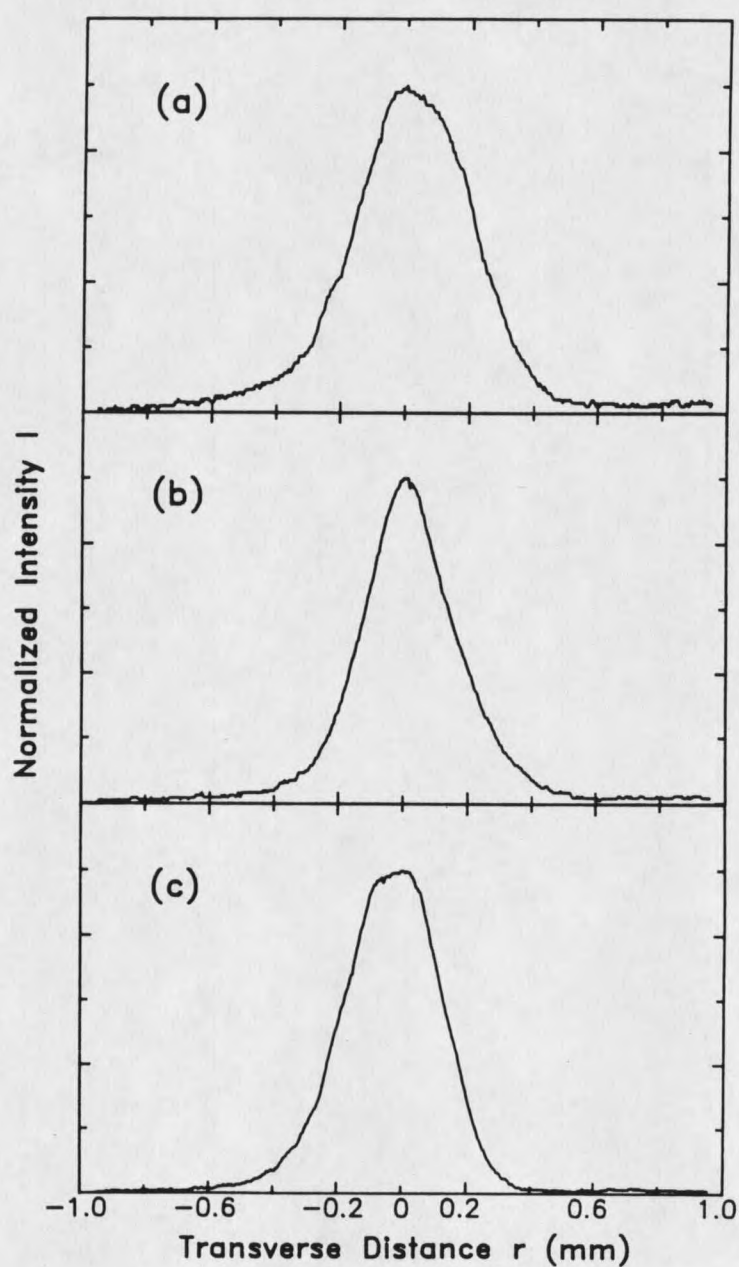


Figure 19. Spatial profile of the input Stokes beam at the amplifier (a) cell entrance, (b) cell center, and (c) cell exit.

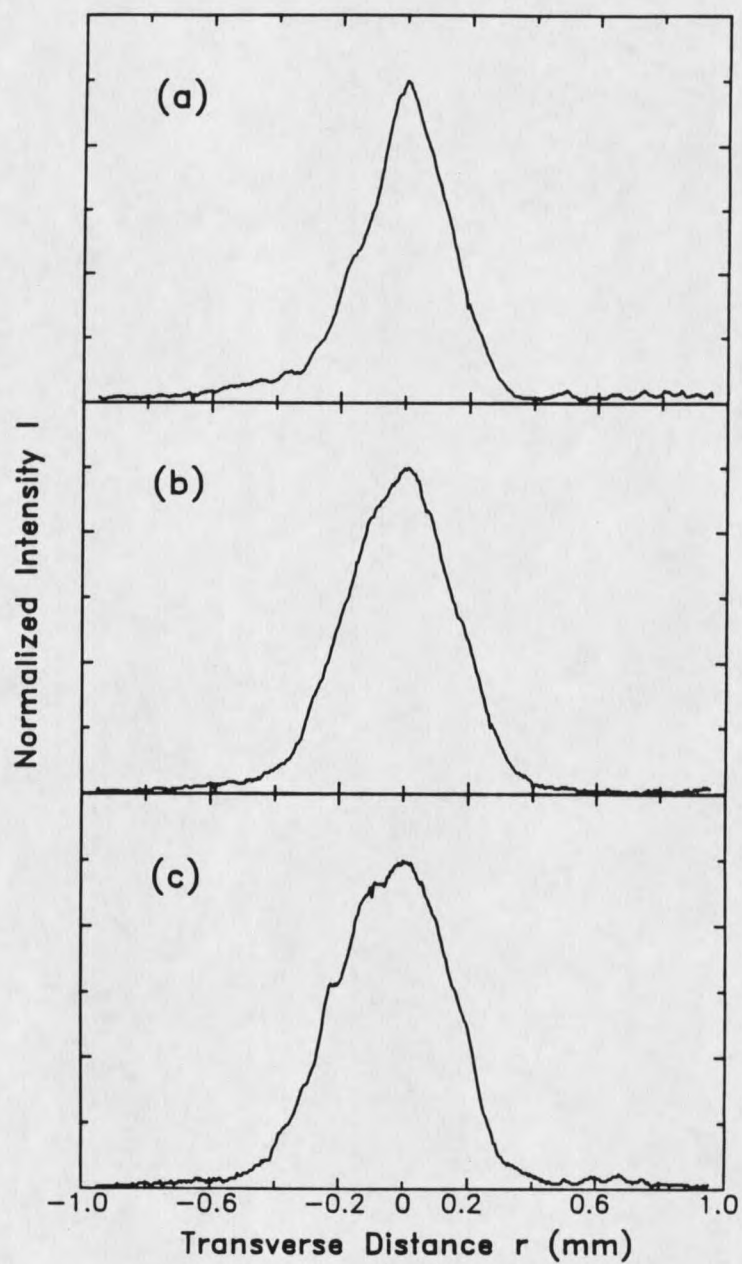


Figure 20. Spatial profile of the input pump beam at the amplifier (a) cell entrance, (b) cell center, and (c) cell exit.

function for the Stokes and it's exponential fit are shown. The Stokes distribution had a HWHM of $.58\text{cm}^{-1}$ while that of the pump was $.47\text{cm}^{-1}$. The larger linewidth for the Stokes beam is not surprising since the Raman linewidth at 100 atmospheres of hydrogen⁵⁰ is 0.08cm^{-1} .

Gain Enhancement at 100 Atmospheres

Fig.14 is a plot of the amplified Stokes energy (solid line) as a function of optical delay. As the optical delay is varied, the pump and Stokes beams become correlated and we begin to see gain enhancement. The gain enhancement continues to increase, experiences a maximum when the pump and injected Stokes beams become highly correlated, and then falls as the two beams once again become uncorrelated. For the data in Fig.8 the incident pump energy was 0.42mJ and the injected Stokes energy was 0.43μJ. The correlated gain enhancement is approximately 13 times the uncorrelated value, and the profile has a temporal width (FWHM) of 14.3ps corresponding to 4.3mm of optical delay in path length.

A measurement of the correlated and uncorrelated gain for various pump intensities was made, thus documenting the dependence of the gain enhancement on pump energy. The data was obtained from a number of plots similar to Fig.14 but at different pump energies. The results, (the X's) shown in Fig.15, are plotted on a log linear scale so that the usual exponential growth will be a linear function of pump energy.

The correlated Stokes gain increases linearly from the value of the injected Stokes and eventually saturates due to pump depletion. The uncorrelated Stokes gain initially grows much slower than the correlated gain, but eventually develops a slope comparable to that of the correlated gain before saturating.

Zero of Optical Delay

Note that in Fig.14 on page 41, the point of maximum gain is the point of zero delay. This result is artificial. An unsuccessful attempt was made to determine the zero delay point interferometrically with the experiment apparatus described in chapter 5. A number of interference profiles similar to those obtained in the linewidth experiment and shown in Fig.10 were obtained. These, too, were fit with Eq.33 to obtain the normalized autocorrelation function. A plot of the normalized autocorrelation function for this experiment is shown in Fig.21.

Though it was impossible to obtain a fit to this data directly it seems reasonable to assign the location of the visibility peak at 22.6mm with .5mm error bars. The optical path at the Stokes frequency is different from that for the pump. Corrections for this difference in optical path shift the peak by .702mm. The new location for the peak is at 21.9mm $\pm$.5mm.

With this identical configuration of quartz, a run was made

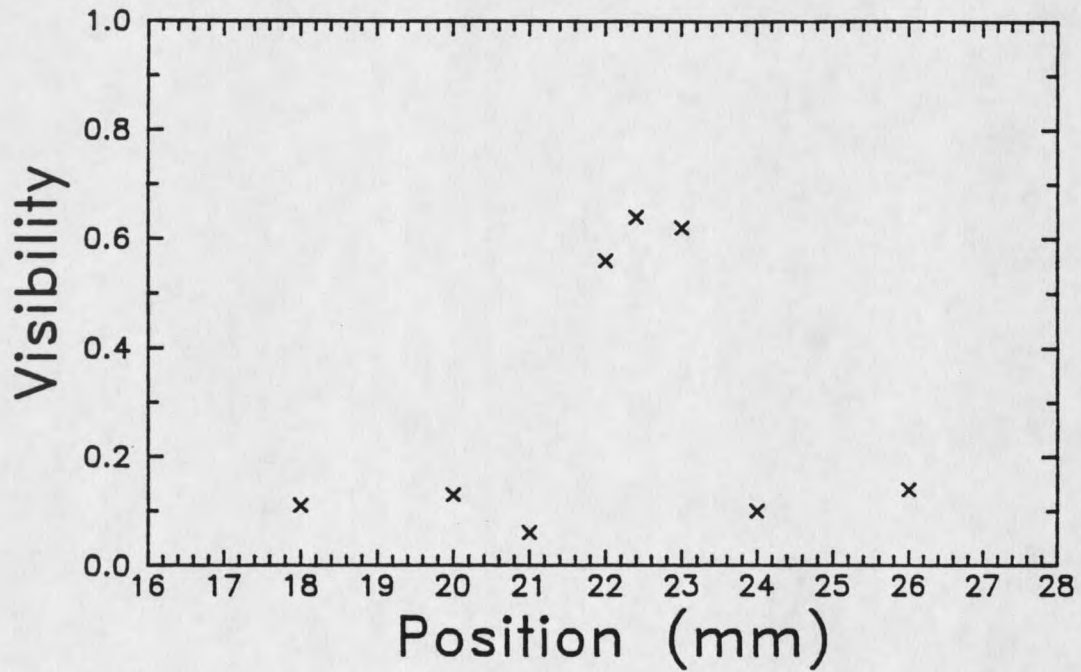


Figure 21. Experimental data for the autocorrelation function showing approximately the point of zero delay: the point at which both paths through the gain enhancement apparatus are optically equivalent.

to obtain a gain enhancement profile which is shown in Fig.22. The enhancement peak is located at $19.8\text{mm} \pm .1\text{mm}$, occurring $2.1\text{mm} \pm 1.5\text{mm}$ sooner than the interferometrically determined zero of optical delay. This corresponds to $7\text{ps} \pm 5\text{ps}$ in time. Theoretical calculations, along with arguments from physical considerations, both discussed in the next chapter, indicate that the zero of optical delay is expected to precede the gain enhancement peak. Even assuming the largest error, this measurement indicates that the converse is true. On theoretical grounds then, we are compelled to reject this experimental measurement. Perhaps the choice of error bars was too optimistic, but in any case, another more careful experiment is needed either to confirm or refute this result.

Gain Enhancement at 10 Atmospheres

The gain enhancement experiment discussed in the previous section was performed again in an identical fashion except that the pressure in the Raman generator and amplifier was reduced to 10 atmospheres. The motivation for this experiment was to study gain enhancement in the limit when the Raman linewidth becomes comparable to the laser mode spacing. Since the Raman linewidth in H_2 at room temperature depends linearly on pressure in the range from 5 atmospheres to greater than 100 atmospheres⁴⁷, reducing the pressure by a factor of ten, in effect, reduces the

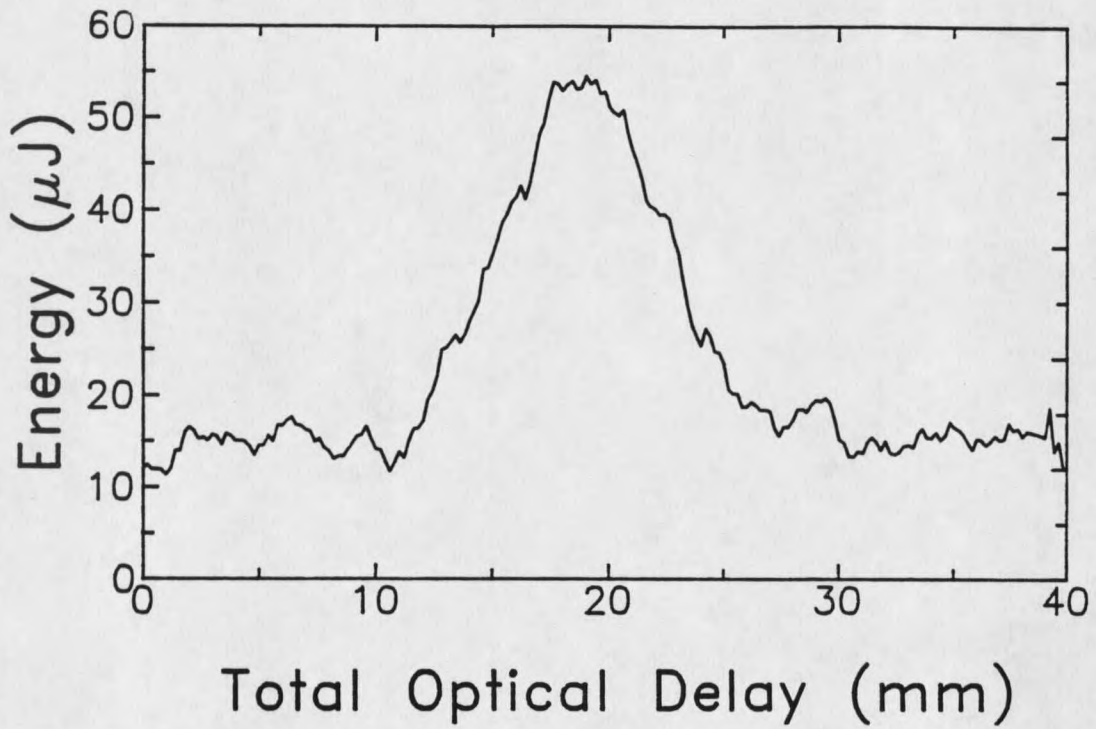


Figure 22. Gain enhancement profile showing the location of the enhancement peak.

linewidth by a factor of ten.

The results of this measurement are given by the X's in Fig.23. Both the correlated and uncorrelated gain again grow from the value of the energy of the injected Stokes beam, but in a manner quite different than the 100 atmosphere data. For this case, both the correlated and uncorrelated gain seem to be restrained from growth until some minimum pump energy is reached ($\approx 3\text{mJ}$ for the correlated gain). After this, both grow linearly with quite comparable slopes until saturation, at which point they both converge. In the region where the correlated and uncorrelated gains are both linear, the correlated gain is approximately one and a half orders of magnitude greater than the corresponding uncorrelated gain.

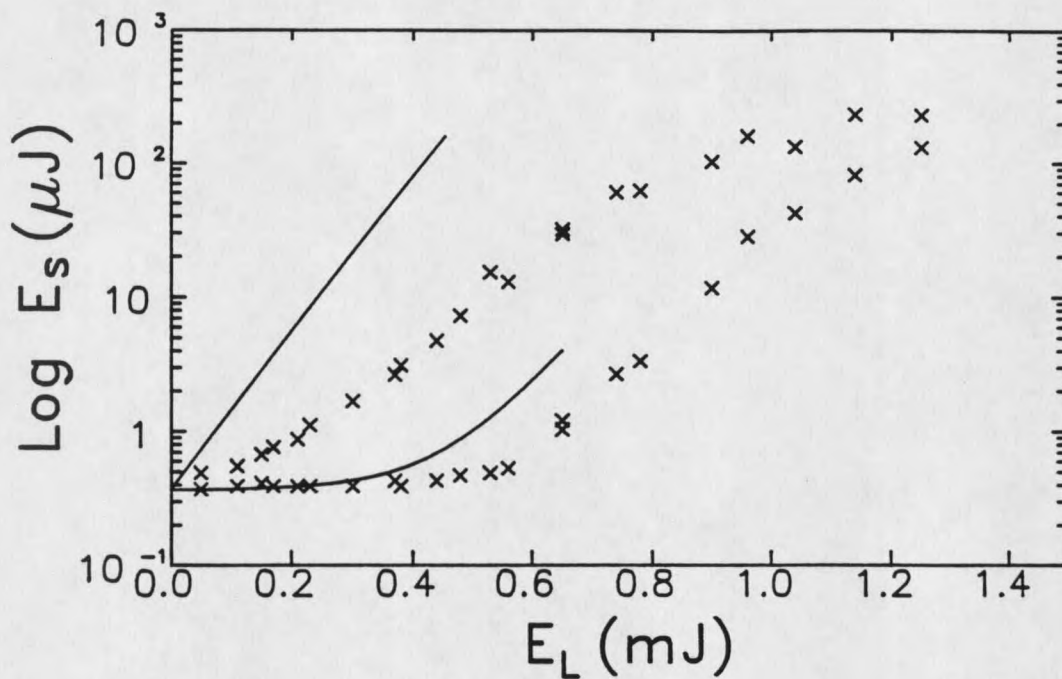


Figure 23. Correlated and uncorrelated gain enhancement as a function of input pump energy at 10 atmospheres. The experimental data are given by the X's while the solid line shows the theoretical prediction.

CHAPTER SEVEN

COMPARISON WITH THEORY

Introduction

In this section a comparison is made between the experimental results of chapter 6 and the predictions of the multimode, fixed-random-phase theory given by equations 36 and 37 in chapter 4. For simplicity, the input Stokes field was taken to have the same Lorentzian lineshape, mode spacing, and phase distribution as the pump. The appropriateness of these assumptions merit consideration, but were not, for lack of time, explored in this work.

The mode distribution was artificially chopped at 640 modes to make computation tractable. For a laser modespacing of $.0035\text{cm}^{-1}$, the magnitude of the intensity at the cutoff point is about $.14I_{\text{max}}$. This mode distribution along with the Raman linewidth at 100 atmospheres for comparison is shown on two different scales in figures 24 and 25. The individual modes are shown in Fig.24 and the overall envelope is shown in Fig.25. As one can see graphically from these figures, our calculations at 100 atmospheres are for a Raman linewidth large compared to the mode spacing of the pump and Stokes fields, but small compared to their linewidths.

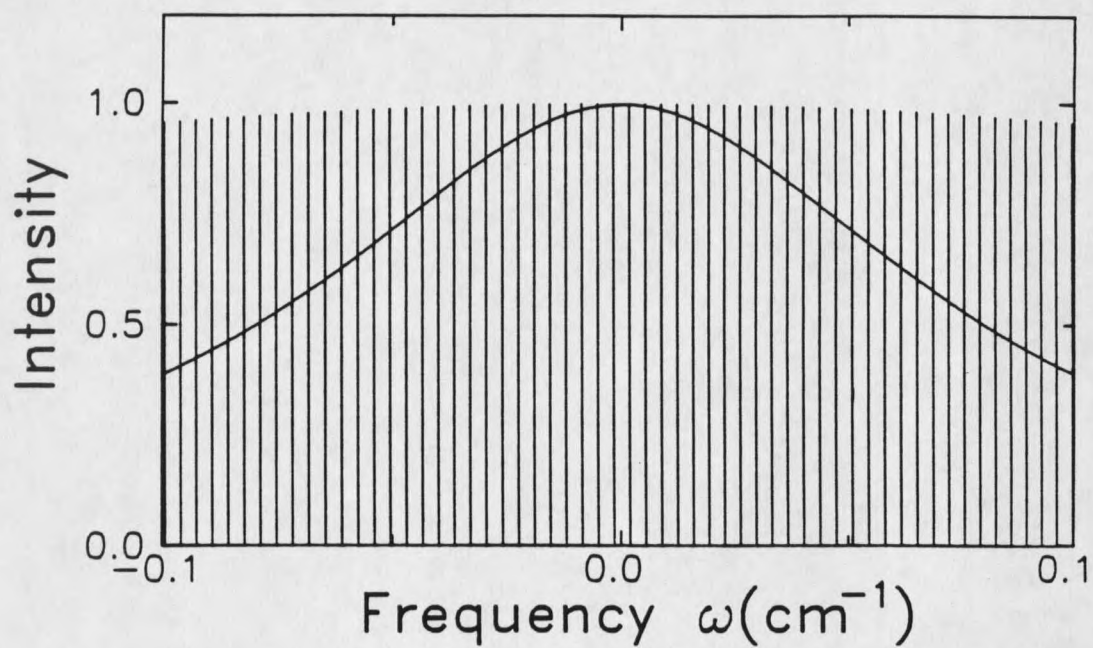


Figure 24. Distance between laser modes (vertical lines) separated by $.0035 \text{ cm}^{-1}$ is small compared to the Raman linewidth of $.08 \text{ cm}^{-1}$ (solid line).

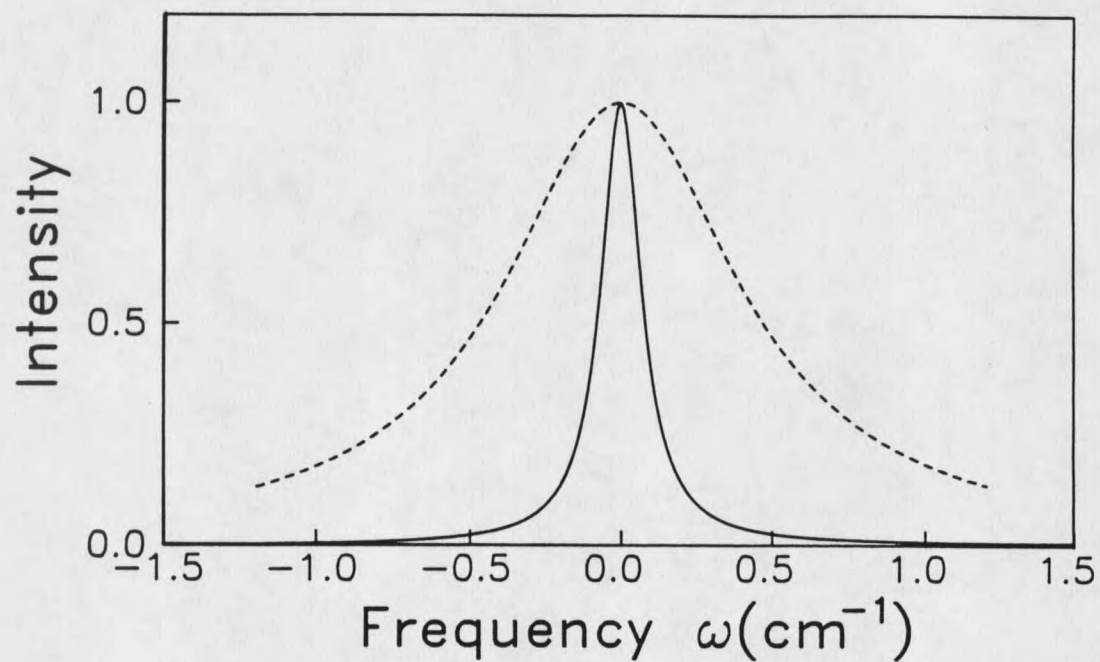


Figure 25. Lorentzian frequency distribution of the laser given by the dashed line was chopped at 1.12cm^{-1} and is large compared to the Raman linewidth of $.08\text{cm}^{-1}$ plotted as the solid line.

A random number generator was used to give each mode a random phase. For a 4.685ns pulse, the superposition of these modes results in the spiky envelope shown in Fig.26.

Using these values for the linewidth and number of modes, and using gain and optical delay as parameters, Eq.36 was numerically integrated to give the Stokes field at the amplifier cell exit.

100 Atmospheres

The dashed line in Fig.14 on page 41 is the predicted Stokes energy plotted as a function of optical delay. The profile has a temporal width of 21ps corresponding to 6.3mm of optical path delay, and a correlated to uncorrelated gain ratio of approximately 13, comparing well with the experimental result. The gain coefficient⁵¹ for this calculation was 6.8×10^{-9} cm/W giving a theoretical power density equal to 71% of the experimentally measured power density. This is reasonable since the on axis power density for the pump varies by approximately a factor of two across the 1m amplifier cell.

An interesting feature of the theoretical prediction is a 2ps asymmetry in the gain enhancement profile, which indicates a larger gain enhancement when the Stokes field precedes the pump field. This effect is related to the transiency of the Raman transition. When the Stokes pulse precedes the pump pulse, that is, when a spike in the Stokes

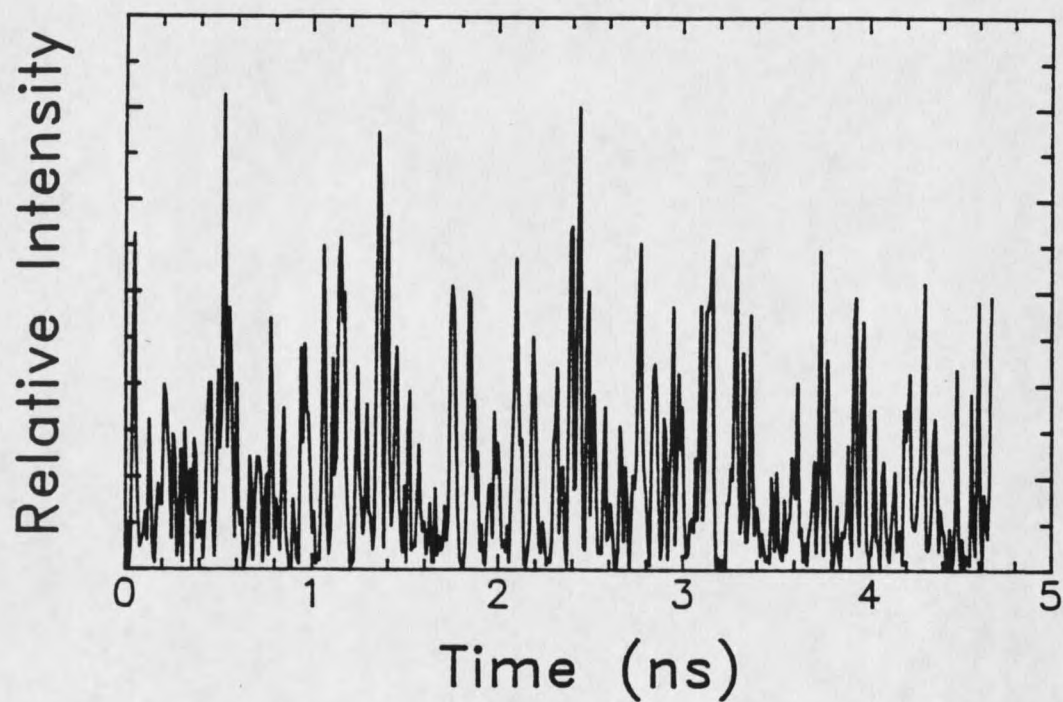


Figure 26. Plot of the theoretically calculated intensity envelope for the laser. The laser field was given a 4.65ps pulse length and results from the superposition of 640 longitudinal modes with fixed but random phases.

field precedes the corresponding spike in the pump field (see Fig.26) by some time τ_D which is short compared to the coherence time of both the medium and the pump, then a strong Q^* with the proper phase will be present in the medium as the pump spike arrives. The strong Q^* and pump fields are then maximally effective in driving Stokes amplification. In these calculations the coherence time for the medium was 60ps while that of the pump was approximately 14ps so that a 2ps asymmetry is not unreasonable.

The solid line in Fig.15 on page 42 is the theoretical prediction for the gain as a function of input pump energy for the correlated and uncorrelated cases. The theory fits the data well in the region before pump depletion. Note that in the region in which both the correlated and uncorrelated gains are linear, the theory predicts that the correlated and uncorrelated gains grow at slightly different rates.

Another interesting prediction of this theory is a correlated gain which is larger than the monochromatic gain. The monochromatic gain is plotted as the dashed line in Fig.15 for comparison. Since the phases of the modes are random, this enhancement is not the result of modelocking, as one might naively expect. Instead, it comes from amplitude modulation in the pump field due to beating of the various random-phase modes, and results in peaks in intensity which are larger than the intensity of a

monochromatic field of the same average intensity. Since the gain depends exponentially on the pump intensity, the correlated gain for a multimode pump is larger than the monochromatic gain. Gains which are higher than monochromatic have also been predicted for the chaotic model^{33,34}. The multimode, fixed-random-phase model is expected to approach the chaotic model in the limit when the mode spacing of the pump is small compared to the Raman linewidth. The results presented here are consistent with that prediction.

10 Atmospheres

At 10 atmospheres, the value for the Raman linewidth is smaller than the value at 100 atmospheres by a factor of ten. In this regime, the laser mode spacing is no longer small compared to the Raman linewidth and the correlated gain is expected to fall.

The solid line in Fig.23 on page 60 is the theoretical prediction for gain enhancement at 10 atmospheres. As one can readily see, the theory fits the data poorly. The experimental data seems to experience a nonlinear exponential growth below some critical input pump value for both the correlated and uncorrelated cases, while the theory predicts that only the uncorrelated gain should grow this way. The correlated gain is predicted to grow linearly from the value of the input Stokes energy in a manner identical

to the 100 atmosphere case. Though the theory correctly predicts the slope for the uncorrelated gain in the region where it grows linearly, it fails to do so for the correlated gain. Also, whereas the experimental data indicates that the correlated gain should be approximately one and a half orders of magnitude greater than the uncorrelated gain, the theory indicates that this value should be closer to three orders of magnitude, twice the experimental result.

In trying to discover the discrepancy between the theoretical and experimental results, several factors have been ruled out as possible explanations. The first is dispersion. Because of dispersion, different frequency components of the pump and Stokes pulses should accumulate relative phase shifts as they propagate through the dispersive medium of the experiment. Since the index at the pump frequencies will be different than the index at the Stokes frequencies, one would expect the correlated gain to fall because the two pulses spread out differently, which is to say that the two pulses will no longer be well correlated. In fact though, $dn/d\lambda$ is approximately constant over $\Delta\lambda$, the bandwidth of the pump or Stokes pulse and consequently the change in phase across $\Delta\lambda$ is negligible. The effect of dispersion will only be to shift the envelope of one pulse relative to the other and this can be compensated for with the optical delay τ_D .

Another possibility is a change in the spatial overlap of the input pump and Stokes pulses in the amplifier cell. We found the gain enhancement to be a very sensitive function of the spatial overlap or alignment of the two beams in the amplifier cell so that vibration of the optics during an experimental run would displace the two beams relative to one another causing the gain to fall. The slope of the correlated gain however depends not on the alignment of the two beams, but instead on the constant k_1 given by Eq.18 and the input pump intensity. The input pump intensity does vary from shot to shot experimentally, but time averages to a constant value during an experimental run. So, it would not be surprising to see the experimental value for the correlated gain falling below the theoretical prediction but it should none-the-less grow with the same slope.

One possibility which merits serious investigation is the laser linewidth which was also found to vary during the course of an experiment. The spectrograph used to monitor the laser linewidth reveals only gross characteristics i.e. whether one line or many lines were present in the laser spectrum. It was insensitive to the width of the actual laser line. Since several months had elapsed between the 100 atmosphere and 10 atmosphere experiments, very possibly the operating characteristics of the laser had changed. It may be that the second experiment was run with a different

laser linewidth than the first. Should this have been the case, then the discrepancy between the experimental results and theoretical predictions would come as no surprise. Before anything more can be learned, a more sensitive monitor of the laser linewidth is needed. Future experiments are planned using a Fabry-Perot to accurately measure and monitor the laser linewidth.

CHAPTER EIGHT

CONCLUSIONS

Conclusions for this thesis have already been given in previous chapters; only a brief summary will be given here. A multimode fixed-random-phase model works well to describe gain enhancement with a XeCl laser when pumping H_2 at 100 atmospheres. The model successfully describes gain enhancement as a function of optical delay (correlation between the pump and Stokes fields) and gain enhancement as a function of input pump intensity. The model also predicts that the gain enhancement will be larger than the monochromatic gain. Larger than monochromatic gains have also been predicted when a chaotic model for broadband pumping has been used^{33,34}. It is expected that the multimode fixed-random-phase model becomes approximately equivalent to the chaotic model in the limit when the laser modespacing becomes much smaller than the Raman linewidth. At 100 atmospheres of H_2 the Raman linewidth is small compared to the laser linewidth but large compared to the laser modespacing thus the results presented in this work support that prediction.

The model is much less successful at predicting gain enhancement at 10 atmospheres of H_2 when the Raman linewidth

becomes comparable to the modespacing of the laser. It may be that the theoretical prediction differs from the experimental results because the value used for the laser linewidth in the calculations was incorrect. The bandwidth of the laser was found to vary dramatically with time so that the linewidth of the laser measured during the 100 atmosphere experiment- and used for all calculations in this thesis- may have been different than the linewidth of the laser during the 10 atmosphere experiment. Future experiments are planned which will include a Fabry-Perot interferometer to monitor the linewidth in real time.

The multimode fixed-random-phase theory predicts an asymmetry in the profile of gain enhancement as a function of optical delay. The experiment to measure this asymmetry yielded a curious and likely erroneous result: an asymmetry was present in the experimental data, but unfortunately went in the wrong direction. A more careful or perhaps more clever measurement is still needed.

Whether, ultimately, the multimode fixed-random-phase model will prove to be the correct model for the XeCl laser is yet unknown. Many uncertainties about the model still exist. The fact that so many of the predictions given by the model are either correct or close gives one cause for optimism.

REFERENCES CITED

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1. C.V. Raman, K.S. Krishna, "A new type of secondary radiation," Nature **121**, 501(1928).
2. G. Herzberg, Spectra Of Diatomic Molecules. New York: Van Nostrand Reinhold, 1950.
3. M.G. Raymer, J. Mostowski, J.L. Carlsten, "Theory of simulated Raman scattering with broad-band lasers," Phys. Rev. A **19**, 6, 1979.
4. L. Westling, "Stimulated Raman scattering with a multimode pulsed dye laser," PhD. dissertation, Institute of Optics, Rochester, New York, 1986
5. N. Bloembergen, Y.R. Shen, "Multimode effects in stimulated Raman emission," Phys. Rev. Let. **13**, 720(1964).
6. Y.E. D'yakov, "Excitation of stimulated light scattering by broad spectrum pumping," ZHETF Pis. Red. **11**, 362(1970).
7. S.A. Akhmanov, K.N. Drabovich, A.P. Sukhornkov, A.S. Chirkin, "Stimulated Raman scattering in a field of ultrashort light pulses," Sov. Phys. JETP **32**, 266(1971).
8. T. Von Forester, R.J. Glauber, "Quantum theory of light propagation in amplifying media," Phys. Rev. A **3**, 1484(1971).
9. S.A. Akhmanov, Y.E. D'yakov, L.I. Pavlov, "Statistical phenomena in Raman scattering stimulated by a broadband pump," Sov. Phys. JETP **39**, 249(1974).
10. J. Cooper, A. Szoke, "Effects of bandwidth on intense-field Raman scattering," Phys. Rev. A **23**, 378(1981).
11. M.G. Raymer, J. Mostowski, "Stimulated Raman scattering: unified treatment of spontaneous initiation and spatial propagation," Phys. Rev. A **24**, 1980(1981).
12. M.G. Raymer, M. Trippenbach, K. Rzazewski, "Stimulated Raman scattering of colored chaotic light," JOSA B **1**, 671(1984).

13. M.G. Raymer, L.A. Westling, "Quantum theory of Stokes generation with a multimode laser," JOSA B 2, 1417(1985).
14. M. Trippenbach, C. L. Van, "Intensity cross-correlation functions in stimulated Raman scattering of colored chaotic light," JOSA B 3, 879(1986).
15. G.C. Valley, "Transfer of pump spatial variations to Stokes phase in Raman amplifiers," IEEE J.Q.E. 9, 1370(1982).
16. J. Reintjes, R.H. Lehmberg, R.S.F. Chang, M.D. Duignan, G. Calame, "Beam cleanup with stimulated Raman scattering in the intensity-averaging regime," JOSA B 3, 1408(1986).
17. H. Komine, W.H. Long, Jr., E.A. Stappaerts, S.J. Brosnan, "Beam cleanup and low-distortion amplification in efficient high-gain hydrogen Raman amplifiers," JOSA B 3, 1428(1986).
18. D. Korff, E. Mazur, C. Dkuzy, A. Flusberg, "Raman conversion using crossed broadband pump beams and bisecting stokes," JOSA B 3, 1333(1986).
19. S.F. Fulghum, D. Klimek, A. Flusberg, D.W. Trainor, C. Duzy, H. Hyman, J.D. Daughery, D. Korff, "Stokes phase preservation during Raman amplification," JOSA B 3, 1448(1986).
20. S.J. Pfeifer, "Multiline Raman beam combination," JOSA B 3, 1363(1986).
21. J.P. Partanen, M.J. Shaw, "High-power forward Raman amplifiers employing low-pressure gases in light guides. I. Theory and applications," JOSA B 3, 1374(1986).
22. J.M. Eggleston, "Solutions to Raman- scattering equations in the presence of multiple equivalent incoherent multiaxial-mode laser beams," JOSA B 3, 1390(1986).
23. S.A. Akhmanov, Y. E. D'yakov, A.S. Chirkin, "Accumulating nonlinear optical effects in a pump field with a broad frequency spectrum," ZhETF Pis. Red. 13, 724(1971).

24. A.Z. Grasyuk, I.G. Zubarev, N.V. Suyazov, "Influence of spectral line width of exciting radiation on the gain in stimulated scattering," ZhETF Pis. Red. **16**, 237(1972).
25. F.A. Korolev, O.M. Vokhnik, V.I. Odintsov, "Signal gain in stimulated scattering with broadband pumping," Sov. Tech. Phys. Lett. **2**, (1976).
26. G.P. Dzhotyan, Y.E. D'yakov, I.G. Zubarev, A.B. Mironov, S.I. Mikhailov, "Amplification during stimulated Raman scattering in a nonmonochromatic pump field," Sov. Phys. JETP **46**, 3(1977).
27. W.R. Trutna, Jr., Y.K. Park, R.L. Byer, "The dependence of Raman gain on pump laser bandwidth," IEEE J.Q.E. **15**, 648(1979).
28. J. Eggleston, R.L. Byer, "Steady-state stimulated Raman scattering by a multimode laser," IEEE J.Q.E. **16**, 850(1980).
29. G.P. Djotyan, L.L. Minasyan, "On the theory of the simulated Raman scattering of nonmonochromatic optical radiation," Op. Comm. **58**, 278(1986).
30. G.G. Lombardi, H. Injeyan, "Phase correlation in a Raman amplifier" JOSA B **3**, 1461(1986).
31. O.M. Vokhnik, I.V. Nikitin, V.I. Odintsov, "Delay of broadband-pumped stimulated Raman gain," Opt. Spectr. (USSR) **45**, 1(1978).
32. E.A. Stappaerts, W.H. Long, Jr., H. Komine, "Gain enhancement in Raman amplifiers with broadband pumping," Op. Lett. **5**, 4(1980).
33. G.S. Agarwal, "Stimulated Raman scattering in chaotic fields," Op. Comm. **35**, 267(1980).
34. A.T. Georges, "Theory of stimulated Raman scattering in a chaotic incoherent pump field," Op. Comm. **41**, 61(1982).
35. A. Yariv, Quantum Electronics. New York: John Wiley and Sons, 1975.
36. Y.R. Shen, The Principles Of Nonlinear Optics. New York: John Wiley and Sons, 1984.
37. P. Lorrain, D. Corson, Electromagnetic Fields And Waves. San Francisco: W.H. Freeman and Company, 1970.

38. J.D. Jackson, Classical Electrodynamics. New York: John Wiley and Sons, 1975.
39. O. Svelto, Principles Of Lasers. New York: Plenum Press, 1976.
40. Lambda Physik, Supplement-EMG 150.
41. E. Hecht, A. Zajac, Optics. California: Addison-Wesley, 1974.
42. J.W. Goodman, Introduction To Fourier Optics. San Francisco: McGraw-Hill, 1968.
43. J.L. Carlsten, J. Rifkin, D.C. MacPherson, "Spatial mode structure of stimulated Stokes emission from a Raman generator," J. Opt. Soc. Am. B 3, 1986.
44. S. Gasiorowicz, Quantum Physics. New York: John Wiley and Sons, 1974
45. G. Herzberg, Spectra Of Diatomic Molecules. New York: Van Nostrand Reinhold, 1950.
46. R. Eisberg, R. Resnick, Quantum Physics Of Atoms, Molecules, Solids, Nuclei, And Particles. New York: John Wiley and Sons, 1985.
47. R.W. Minck, E.E. Hagenlocker, W.G. Rado, "Consideration and evaluation of factors influencing the stimulated optical scattering in gases," Ford Motor Company, 1966.
48. M. Abramowitz, I.A. Stegun, Handbook of Mathematical Functions. National Bureau of Standards, 1972.
49. S. Koonin, Computational Physics. Menlo Park: The Benjamin Cummings Publishing Company, 1986.
50. O.W. Young, "High-resolution cw stimulated Raman spectroscopy in molecular hydrogen," Opt. Lett. 2, 91(1978).
51. W.K. Bischel, M.J. Dyer, "Wavelength dependence of the absolute Raman gain coefficient for the Q(1) transition in H₂," J. Opt. Soc. Am. B 3, 677(1986).

APPENDICES

APPENDIX A

RELATIVE SIZE OF Δ_L

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RELATIVE SIZE OF Δ_L

The energy of the first electronically excited state of hydrogen is approximately 10.2eV^{10} . This corresponds to approximately $82,200\text{cm}^{-1}$. The laser, at a wavelength of 308nm , and Stokes at 351nm , corresponds to $32,450\text{cm}^{-1}$ and $28,500\text{cm}^{-1}$ respectively. Δ_L , which is the difference between the first electronically excited state of hydrogen and the laser energy is $49,759\text{cm}^{-1}$.

APPENDIX B

TRANSFORMATION TO THE TRAVELING REFERENCE FRAME

APPENDIX B

TRANSFORMATION TO THE TRAVELING REFERENCE FRAME

Fig.27 shows both the traveling and laboratory reference frames. Both z and Z measure distances in the medium so that $z(t,z)=Z$. τ on the other hand measures time at rest with respect to a point on the pulse; therefore $\tau(t,Z)=t-Z/v_s$, where v_s is the velocity of the pulse in the medium. Defining the differential operator D as in chapter 2, we simply apply the chain rule

$$\frac{\partial}{\partial z} \frac{\partial z}{\partial Z} \bar{D} + \frac{\partial}{\partial \tau} \frac{\partial \tau}{\partial Z} \bar{D} = \frac{\partial}{\partial z} \frac{\partial z}{\partial Z} + \frac{1}{v_s} \frac{\partial}{\partial z} \frac{\partial z}{\partial t} + \frac{\partial}{\partial \tau} \frac{\partial \tau}{\partial Z} + \frac{1}{v_s} \frac{\partial}{\partial \tau} \frac{\partial \tau}{\partial t} = \frac{\partial}{\partial z}$$

since

$$\frac{\partial z}{\partial Z} = 1, \quad \frac{\partial z}{\partial t} = 0, \quad \frac{\partial \tau}{\partial Z} = -\frac{1}{v_s} \quad \text{and} \quad \frac{\partial \tau}{\partial t} = 1.$$

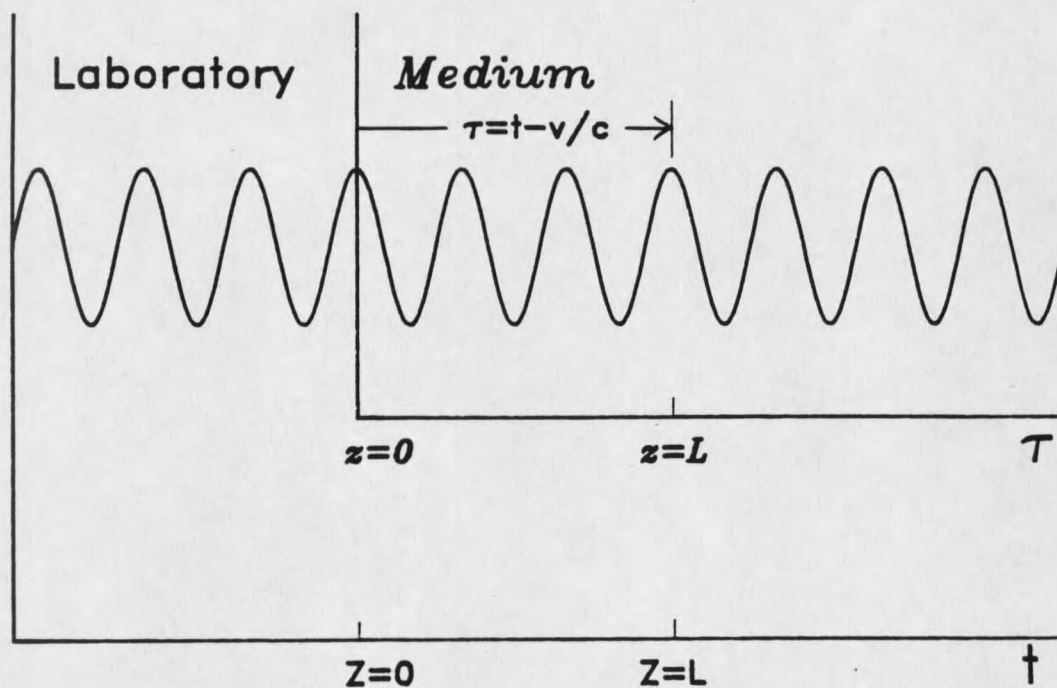


Figure 27. The traveling frame is at rest with respect to the wave. Points on the wave are located relative to an arbitrary origin, here chosen at the boundary of the medium with $z=Z=0$.

APPENDIX C

COMPUTER PROGRAMS

Figure 28. Computer program Lfields.

```

C
C *****
C      LFIELDS.FOR                      7-08-87
C *****
C      INITIALIZATION
C
C      REAL P(0:5000), P2(5000), RN(640), ELR, ELI, PHIL(640),
*      PHIS(640),
*      AMPL(640), AMPS(640), REESO(0:5000)
C
C      COMPLEX EL(0:5000), ES0(0:5000), ES(0:5000),
*      LASER, STOKES
C
C      INTEGER*4 ID
C
C      OPEN (1, FILE='FIELDS.DAT', STATUS='OLD')
C      OPEN (2, FILE='EL.DAT', STATUS='OLD')
C      OPEN (3, FILE='ES0.DAT', STATUS='OLD')
C      OPEN (4, FILE='REESO.DAT', STATUS='OLD')
C      OPEN (5, FILE='P.DAT', STATUS='OLD')
C
C      READ(1, *) DELTA, HWIDTH, TMAX, TSTEP, IRI, MODES, IRANDOM
C
C      C=3.E-2
C      PI=3.1415926
C      C2=2.*PI*C*DELTA
C
C *****
C      LOAD RANDOM NUMBER ARRAY AND CALCULATE INITIAL PHASES
C
C      IF (IRANDOM.NE.1) GOTO 50
C          M=32753
C      DO 40 I=1, MODES
C          JRI=IRAND(IRI, M)
C          IRI=JRI
C          RN(I)=IRI*2.*PI/M
C          PHIL(I)=RN(I)
C          PHIS(I)=RN(I)
40      CONTINUE
C      GOTO 70
50      DO 60 K=1, MODES
C          PHIL(K)=0
C          PHIS(K)=0
60      CONTINUE
C
C *****
C      CALCULATE THE LASER AND STOKES FIELDS AND THE LASER ENERGY

```

Figure 28. Continued.

```

C *****
C CALCULATE WEIGHTED AMPLITUDES USING A LORENTZIAN LINESHAPE
C
70    J0=MODES/2
      ASUM=0
      DO 200,J=1,MODES
          AMPL(J)=SQRT(1/(HWIDTH**2+(DELTA*(J-J0))**2))
c      OPEN (9,FILE='AMPL.DAT',STATUS='OLD')
c      WRITE (9,*) J,AMPL(J)
          ASUM=ASUM+AMPL(J)**2
200    CONTINUE
          SCALE=1/SQRT(ASUM)
      DO 300,J=0,MODES
          AMPL(J)=SCALE*AMPL(J)
          AMPS(J)=AMPL(J)
300    CONTINUE
      P1=0.
      PSUM=0.

C
C CALCULATE THE FIRST LATTICE POINT OF P,THE FIRST ELEMENT C
OF LASER THE LASER ENERGY, AND SIMULTANEOUSLY CALCULATE
C THE INPUT STOKES FIELD.
      WRITE(*,*) 'CALCULATING THE LASER ENERGY'
      T=0.
      I=0
      CALL FIELD(LASER,STOKES,AMPL,AMPS,C2,MODES,T,
        PHIL,PHIS)
      EL(I)=LASER
      P1=EL(I)*CONJG(EL(I))
      ES0(I)=STOKES
      P(I)=0.

C
      WRITE(2,*) EL(I)
      WRITE(3,*) ES0(I)
      WRITE(5,*) P(I)

C
      REESO(I)=ES0(I)*CONJG(ES0(I))
      WRITE(4,*) T,REESO(I)
c      S0=REESO(I)
C
C CALCULATE THE REMAINING ELEMENTS OF P AND LASER
      I=1
      DO 1200,T=TSTEP,TMAX-TSTEP,TSTEP
C
      CALL FIELD (LASER,STOKES,AMPL,AMPS,C2,MODES,T,
* PHIL,PHIS)
          EL(I)=LASER

```

Figure 28.. Continued.

```

      P2(I)=EL(I)*CONJG(EL(I))
      P(I)=PSUM+(P1+P2(I))*TSTEP/2.
      PSUM=P(I)
      P1=P2(I)
      ES0(I)=STOKES
      WRITE(2,*) EL(I)
      WRITE(3,*) ES0(I)
      WRITE(5,*) P(I)
C
      REES0(I)=ES0(I)*CONJG(ES0(I))
      TIME=T*.001
      WRITE (4,*) TIME, REES0(I)
C
      S0=S0+REES0(I)
C
      I=I+1
1200  CONTINUE
C
      END
C
      *****
      SUBROUTINE FIELD(LASER,STOKES,AMPL,AMPS,C2,MODES,T,
* PHIL,PHIS)
C
      COMPLEX LASER,STOKES
      REAL PHIL(640),PHIS(640),AMPL(640),AMPS(640)
      ELR=0.
      ELI=0.
      ESR=0.
      ESI=0.
      DO 200,M=1,MODES
      C3=C2*M*T
      UL=C3+PHIL(M)
      ELR=ELR+AMPL(M)*COS(UL)
      ELI=ELI+AMPL(M)*SIN(UL)
      US=C3+PHIS(M)
      ESR=ESR+AMPS(M)*COS(US)
      ESI=ESI+AMPS(M)*SIN(US)
200  CONTINUE
      LASER=CMPLX(ELR,ELI)
      STOKES=CMPLX(ESR,ESI)
      RETURN
      END
C
      *****
C
      INTEGER FUNCTION IRAND(IRI,M)
C
      INTEGER*4 ID

```

Figure 28. Continued.

```
IA=201  
IC=3679  
ID=IA*IRI+IC  
IRAND=MOD(ID,M)  
END
```

Figure 29. Computer program Mode.

```

C *****
C      MODE.FOR                      2-23-87
C *****
C      INITIALIZATION
C
C      REAL FBESSEL,BESSEL,BDATA(850),BDIF(850),ELR,ELI,
*      P(0:2000),REES(0:2000),ENERGY(500)
C
C      COMPLEX L(0:2000);ES0(0:2000),ES(0:2000),
*      ES1(0:2000),
*      BAMBI,ZAP,C3
C
C      INTEGER*4 ID
C      *****
C      OPEN (1,FILE='MODE.DAT',STATUS='OLD')
C      READ(1,*) DELTA,GAMMA,TMAX,STEP,DMIN,DMAX,DSTEP,
*      GMIN,GMAX,GSTEP
C      *****
C      C=3.E-2
C      PI=3.1415926
C      NOW CONVERT GAMMA FROM WAVENUMBERS TO TIME
C      GAMMA=2.*PI*C*GAMMA
C      C2=2.*PI*C*DELTA
C      N1=(TMAX-STEP)/STEP
C
C      **** READ THE INPUT FIELDS INTO ARRAYS
C      WRITE (*,*) 'TRANSFERRING FIELDS NOW...'
C      OPEN (1,FILE='ES0.DAT',STATUS='OLD')
C      OPEN (2,FILE='EL.DAT',STATUS='OLD')
C      DO 200 K=0,N1
C          READ(1,*) ES0(K)
C          READ(2,*) EL(K)
200  CONTINUE
C      *****
C      OPEN (1,FILE='P.DAT',STATUS='OLD')
C      DO 250 L=0,N1
C          READ (1,*) P(L)
250  CONTINUE
C      ***** READ BESSEL FUNCTION INTO AN ARRAY
C      WRITE(*,*) 'READIN THE OLD BESSEL FUNCTION....'
C      OPEN (1,FILE='BDATA.DAT',STATUS='OLD')
C      OPEN (2,FILE='BDIF.DAT',STATUS='OLD')
C      DO 300 I=1,800
C          READ(1,*) BDATA(I)
C          READ(2,*) BDIF(I)
300  CONTINUE
C *****

```

Figure 29. Continued.

```

C                      CALCULATE THE STOKES FIELD
C *****
  WRITE(*,*) 'CALCULATING THE STOKES FIELD....'
  OPEN (2,FILE='REES.DAT',STATUS='OLD')
  OPEN (3,FILE='GAIN1.DAT',STATUS='OLD')
  OPEN (4,FILE='GAIN2.DAT',STATUS='OLD')
C  UCG IS THE UNCORRELATED GAIN. CG IS THE CORRELATED GAIN.
  ES(0)=ES0(0)
C  CHECK TO SEE IF GAIN LOOP IS BEING USED
      IF (GMIN.EQ.GMAX) THEN
          IFLAG2=1
      ELSE
          IFLAG2=0
      END IF
  DO 8000 GL=GMIN,GMAX,GSTEP
      N=0+IFLAG2
400      C1=SQRT(GL*GAMMA/2.)
      DO 7000 D=DMIN,DMAX,DSTEP
          N=N+1
          IDELAY=NINT(D/STEP)
C
          REES(0)=ES(0)*CONJG(ES(0))
C          WRITE (2,*) 0, REES(0)
          ENERGY(N)=REES(0)
          DO 2700, L=0,N1
              K=L+IDELAY
              IF ((K.LT.0).OR.(K.GT.N1)) THEN
                  ES1(L)=CMPLX(0.,0.)
              ELSE
                  ES1(L)=ES0(K)
              END IF
2700      CONTINUE
C
          J=1
          DO 3000, T0=STEP,TMAX-STEP, STEP
              C3=C1*EL(J)
              CALL ZZAP(ZAP,STEP,T0,GAMMA,EL,J,C1,P,BDATA,
* BDIF,ES1,ARG)
              ES(J)=ES1(J)+C3*ZAP
C
              REES(J)=ES(J)*CONJG(ES(J))
              REES0=ES0(J)*CONJG(ES0(J))
              REES(J)=AMIN1(10000.*REES0,REES(J))
              TIME=.001*T0
C              WRITE (2,*) TIME, REES(J)

              ENERGY(N)=ENERGY(N)+REES(J)

```

Figure 29. Continued.

```

          J=J+1
3000    CONTINUE
        WRITE (*,*)
        WRITE (*,*) 'DELAY IS ',D,'ps          GAIN IS ',GL
        ENERGY=ENERGY(N)/TMAX*STEP
        WRITE (*,*) 'AVG OUTPUT ENERGY= ',ENERGY
        WRITE (*,*)
        IF (IFLAG2.EQ.1) THEN
C THE VARIABLE DELAY IS USED FOR PLOTTING ONLY ( => (X,Y) )
          DELAY=D*.001
          WRITE (3,*) DELAY,ENERGY
          GOTO 7000
        ELSE
          IF (N.NE.1) GOTO 4000
          UCG=ENERGY
        END IF
4000    CG=ENERGY
7000    CONTINUE
        WRITE(4,*) GL,UCG,CG
        WRITE(1,*) GL,ARG
8000    CONTINUE
C
      END
C *****
C                               SUBROUTINES
C *****

      SUBROUTINE ZZAP(ZAP,STEP,TO,GAMMA,EL,J,C1,P,BDATA,
* BDIF,ES1,ARG)
C      ZZAP IS THE INTEGRAL HAVING THE INTEGRAND BAMBI
C
      COMPLEX EL(0:2000),SUM,SUM0,SUMTO,BAMBI,ZAP,
* ES1(0:2000)
      REAL BDATA(850),BDIF(850),P(0:2000),ARG
C
      SUM=CMPLX(0.,0.)
C
C      CALCULATE FIRST AND LAST ELEMENTS OF INTEGRAND
      I=0
      T=0
      CALL BBAMBI(TO,T,GAMMA,EL,I,J,C1,P,BDATA,BDIF,
* ES1,IFLAG1,BAMBI,ARG)
      SUM0= BAMBI
      T=TO
      SUMTO= C1*CONJG(EL(J))*ES1(J)
      IF(J.EQ.1) GOTO 3210
C

```

Figure 29. Continued.

```

C      LOOP TO CALCULATE ES(Z,TAU)
      I=J-1
      DO 3200,T=T0-STEP,STEP,-STEP
C
          CALL BBAMBI(T0,T,GAMMA,EL,I,J,C1,P,BDATA,BDIF,
*              ES1,IFLAG1,BAMBI,ARG)
C
          IF (IFLAG1.EQ.1) GOTO 3205
          SUM=SUM+BAMBI
C
          I=I-1
3200    CONTINUE
C
3205      SUM= 2.*SUM
3210      ZAP= STEP/2.*(SUM0+SUM+SUMT0)
C
      RETURN
      END
C
C      *****
      SUBROUTINE BBAMBI(T0,T,GAMMA,EL,I,J,C1,P,BDATA,
*          BDIF,ES1,IFLAG1,BAMBI,ARG)
C      BAMBI IS THE INTEGRAND
C
      COMPLEX BAMBI
      COMPLEX EL(0:2000),ES1(0:2000)
      REAL BDATA(850),BDIF(850),P(0:2000),
*      FBESSEL,ARG
C
      X=T0-T
      PDIF=SQRT(P(J)-P(I))
      ARG=2*C1*PDIF
      BESSEL=FBESSEL(ARG,BDATA,BDIF)
      GX=GAMMA*X
      IF(GX.GT.80) GOTO 4900
      EBP=EXP(-GX+ARG)*BESSEL/PDIF
      IF (EBP.LE.1.E-4) THEN
          IFLAG1=1
      ELSE
          IFLAG1=0
      END IF
      BAMBI=EBP*CONJG(EL(I))*ES1(I)
      GOTO 5000
4900    BAMBI=0.
C
5000    RETURN
      END

```

Figure 29. Continued.

```

C
C *****
C                               FUNCTIONS
C *****
C     CALCULATE THE BESSEL FUNCTION
C     FBESSEL CALCULATES THE BESSEL FUNCTION OF AN ARGUMENT
C     BY INTERPOLATING BETWEEN PREVIOUSLY CALCULATED
C     RESULTS (PDATA).
C     THE METHOD OF LINEAR INTERPOLATON IS USED:
C
C      $Y=Y1+(Y2-Y1)/(X2-X1)*(X-X1)$ 
C
C     BDATA ARE THE BESSEL FUNCTION ARRAY ELEMENTS WHICH
C     HAVE ALREADY BEEN CALCULATED AND READ INTO PROGRAM.
C     BDIF IS AN ARRAY CONTAINING THE SLOPES BETWEEN ANY
C     TWO POINTS OF THE BDATA ARRAY.  ARG IS THE ARGUMENT
C     OF THE BESSEL FUNTION.  BARG PUTS ARG WITHIN THE
C
C     RANGE OF THE BDATA ARRAY.  IARG  AND JARG ARE THE
C     NUMBERS OF THE BDATA ARRAY ELEMENTS SURROUNDING BARG.
C
C     REAL FUNCTION FBESSEL(ARG,BDATA,BDIF)
C
C     REAL BDATA(850),BDIF(850)
C
C     CALCULATE THE ARGUMENT OF THE BESSEL FUNCTION
C
C     BARG=ARG/.05
C     IBARG=BARG
C     JBARG=IBARG+1
C     BESSEL1= BDATA(IBARG)+BDIF(JBARG)*(ARG-IBARG*.05)
C     FBESSEL=BESSEL1
C
C     END

```

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