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UCRL-51446

**STIMULATED BRILLOUIN AND
BACKWARD RAMAN LIGHT SCATTERING**

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Ph.D. Thesis

MS. date: October 1, 1973

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Abstract

Brillouin scattering is scattering of light by acoustic waves. Raman scattering is light scattering by a molecular rotation or vibration. In stimulated scattering the acoustic or molecular vibration is strengthened by intense laser light. In Brillouin and Raman scattering, monochromatic light interacts with one vibration frequency. This frequency is determined by the following conditions:

$$\omega_L = \omega_S + \omega_V$$

$$\vec{k}_L = \vec{k}_S + \vec{k}_V$$

The subscripts L, V, and S denote laser, vibration, and scattered; ω is angular frequency, and $\vec{k}$ is wave vector. If these conditions are multiplied by $\hbar$, Planck's constant, and $\hbar\omega$ and $\hbar\vec{k}$ are identified as quantum energy and momentum, then the conditions correspond to conservation of energy and momentum for the quanta. The conditions also ensure that the incident light and sound interact in phase over large distances compared to wavelengths and over long periods of time compared to periods of oscillation.

One way to produce stimulated scattering is by means of an intense nanosecond pulse from a ruby laser

incident on a scattering cell. The scattered light intensity is then measured.

The standard way of analyzing the scattering is using the following partial differential equations.

$$\frac{\partial E_L}{\partial z} + \frac{1}{c} \frac{\partial E_L}{\partial t} = -\frac{g E_S S}{2}$$

$$\frac{\partial E_S}{\partial z} - \frac{1}{c} \frac{\partial E_S}{\partial t} = -\frac{g E_L S^*}{2}$$

$$\frac{\partial S}{\partial t} + \frac{S}{2T} = -\frac{E_L E_S^*}{2T} + A \sum_m e^{i\phi_m(t-t_m)}$$

Stochastic terms have been added to these equations to account for noise. Here E_L is laser amplitude, E_S scattered light amplitude, and S sound amplitude. These equations apply to both Brillouin and Raman scattering. Analytical solutions in various approximations have been obtained by others. In this thesis, the equations are solved exactly using numerical methods and a digital computer. A distributed noise source is added to the vibration equation to stimulate thermal acoustic waves or zero point vibrations of molecules.

In Brillouin scattering a transient phase occurs while the sound increases to a large enough intensity to scatter most of the laser light. The increase of

the light intensity during this transient phase is computed. Since the laser beam is not uniform but is assumed to have a Gaussian radial intensity distribution, both beam center and average scattered light intensity are computed. The average scattering shows a more gradual increase than at beam center. The time required for the calculated transient to develop is close to experimental values. When the scattered light intensity becomes approximately equal to the laser intensity, the transient scattering phase ends. The scattering is then steady state, and time derivatives are small. The numerical solutions are within 5% of the values obtained from steady state theory, near the peak of the laser pulse where intensities are changing most slowly. Statistical fluctuations in the transient scattering due to the random noise sources are computed. Statistical fluctuations in the laser threshold intensity, the intensity corresponding to 1% of the laser light scattered, are

2%. Different sizes of noise source give very nearly equal steady state scattering.

In Raman scattering, backward scattered pulses appear owing to self-focusing of the laser light. The pulses are amplified to many times the unfocused laser intensity in propagating through the cell toward the laser. Self-focusing is simulated by increasing the laser intensity near the end of the cell. A parameter study involving variation of self-focused intensity and length of self-focused filament is done. The laser intensity is increased from 30 to 100 times to simulate self-focusing, and the self-focusing is assumed to occur at distances from 5 to 1 cm from the end of the cell. The intensity of scattered light fluctuates statistically owing to random origin of scattered light, and the magnitude of fluctuations is comparable with experimental values. The pulse temporal widths are wider than those given in the literature. The widths also fluctuate.

I. Introduction

1. LASERS

The laser, a new source of light, was discovered in 1960. The term laser is an acronym that stands for light amplification by stimulated emission of radiation. The principle of stimulated emission was discovered in 1918 by Einstein.¹ This principle implies that if an atom is excited, it can be triggered by a weak electromagnetic field of the right frequency. The direction and polarization of emitted light are the same as the

direction and polarization of the stimulating field. Masers, or microwave amplifiers by stimulated emission of radiation, were discovered in 1953.² In either device a large number of atoms or molecules are made ready to radiate in phase, i.e., coherently. This situation is made possible by a population inversion. For systems of molecules that are allowed to interact with surroundings, an equilibrium state is reached where most of the molecules have low energies and only a few have high energies. In

a population inversion, most of the molecules have high energies and are ready to give up the energy as radiation. When a weak field is applied, the atoms are stimulated to radiate at the same time. The nature of the light emitted is different from ordinary light, since the atoms radiate in phase. The amplitudes of the radiation from each atom add; in an incandescent lamp, on the other hand, it is the intensities that add. Consequently, higher intensities occur than in incandescent lights. By enclosing the excited atoms in a resonant cavity, the light can be made to go in a single direction with very slight divergence. Resonant cavities are used in microwave generation and filtering. There they are of the order of a wavelength in size. The cavity Q , which is the frequency divided by the band width of the frequencies passed, is large compared to that of low frequency radio circuits; there is a lot of energy stored relative to that absorbed. Q is a measure of the width of the band of frequencies at which the cavity is resonant, and is proportional to the energy stored divided by the energy dissipated per cycle. The Q of optical laser cavities can be larger, as many more wavelengths are stored. Dissipation occurs at the ends. To minimize the loss, highly reflecting mirrors are used. Most of the loss then is the light allowed to pass through one end and subsequently used in applications. Even though the Q of an optical cavity may be large, the band of frequencies passed is large because of the high frequency of the light.

Laser light is narrow in bandwidth. Many fewer energy levels in the atoms

give off energy than in incandescent light sources. The frequency of the light emitted is proportional to the difference in energy between the levels. This light is highly monochromatic, like a single line in the spectrum of the atom. The spectra of elements are used to identify the elements. The width of the lines in the spectra of gases are often very narrow. Bands of frequencies occur in liquids and solids owing to the interaction of molecules with each other.

There are two kinds of lasers. One delivers a steady beam that does not vary in intensity with time. These are continuous or cw lasers. The other kind gives a pulse or sequence of pulses. These are pulse lasers. One method of generating pulses of very high intensity is called Q spolling. The radiation is made to build up in a cavity with high Q . When it has reached a high intensity, the Q is switched or spoiled to a low value by allowing light to leave the cavity. The energy stored in the cavity leaves in a pulse.

One way of switching the laser is to have a bleachable dye in the cavity. When the light intensity is low, the dye is opaque. When the dye bleaches, light is allowed to reach both mirrors. The population inversion in solid state lasers is created using flash tubes. For continuous lasers electric discharge in a gas is used. The helium neon laser is a continuous laser. A ruby laser is a pulsed laser.

Numerous uses for lasers have been found. The continuous type is used as a straight line in surveying. Pulse lasers are used to drill tiny holes and in welding.

2. HOLOGRAPHY

One of the exciting uses of lasers is in holography. Holograms are a new invention made possible by the monochromatic plane wave light of some lasers.

To give a plane wave, the mirrors of a laser must be flat. This is not commonly found, as flat mirrors are more difficult to align. Some lasers have confocal mirrors. Here the light from one mirror is focused on the other mirror. The divergent light can be made into a plane wave using external lenses. If the light reflected from an object can be exactly recreated, the object can be observed in depth. This is the principle of the hologram.

In conventional movies, the light from an object is focused on a film sensitive to variations in intensity. The more intense the light, the darker the film. When light is directed through a positive of the film, the light and dark intensity patterns are projected on a screen. No depth is observed. In holograms light reflected from an object is made to interfere with light from the illuminating source, a laser. The interference pattern is recorded on photographic film. To view the hologram, laser light is directed through the film. The light reflected from an object is then exactly recreated. Consequently the depth of the object can still be observed. In an ordinary photograph the intensity of the light from each point on an object is recorded. In a hologram the intensity and the phase of the light relative to other points are recorded. The phase of a wave is a measure of when it started.

A wave consists of alternating maxima and minima. The phase tells where a maximum occurs. The position of all subsequent maxima and minima is then determined if the wavelength of the wave is known. Two light beams incident on a surface interfere. The intensity is not the sum of the two intensities, since maxima and minima do not coincide. If the phase difference is complete, one wave can cancel out the other. At a different place on the film the waves will add. A weird interference pattern results. Shining light through the pattern translates the pattern into a plane wave with the proper phase and intensity at each point.

3. LIGHT

Light is a familiar physical phenomenon. Objects are distinguished by the color of the light they reflect. Sunlight has a wide spectrum of colors, as does any light that appears white. Objects absorb some of the colors, reflect others, and transmit (are transparent to) still others. Geometrical optics is the study of phenomena that are related to lenses.

Although common experience gives no clue, it was discovered in the last century that light and radio waves are very closely related. They both are electromagnetic waves. Electric and magnetic fields in a part of space, if they vary in a certain way in space and time, can move from place to place. This motion takes place at the speed of light, even if the fields are radio waves. The wavelength of the electromagnetic radiation distinguishes light waves from radio waves. The wavelengths of light

waves are much shorter than the wavelength of radio waves. The wavelength of AM radio waves are from 600 to 200 m. Visible light has wavelengths from 3×10^{-5} to 7×10^{-5} cm. It is this difference in wavelength that causes the different properties of light and radio waves. Light travels in straight lines, while radio waves can go around obstacles. Radio waves can be generated, modulated, amplified, and demodulated in electric circuits of macroscopic size. Light waves, because of their short wavelength, cannot. Radiation interacts strongly with objects that are smaller than the wavelength.

Radio waves and light waves are only two of many different kinds of electromagnetic radiation. Electromagnetic radiation is named according to the uses or perceptions that it gives. It is customarily classified in a spectrum, where it is arranged according to decreasing wavelength or increasing frequency. The frequency and wavelength are related by $f = c/\lambda$; c is speed of light, f is frequency; and λ is wavelength. The electromagnetic spectrum is like the periodic table of the elements. Electric power has the lowest frequencies. There is some radiation from power lines, but since the wavelength is 3000 miles, it is inappreciable, as most power lines are much shorter than this. Next comes ordinary or AM radio, short wave radio, FM (frequency modulation) radio, and TV. The most familiar use of microwaves is radar. Infrared rays are light rays that are invisible, but can be felt as heat. Hot objects that are not incandescent radiate in the infrared. Very hot objects such as filaments in lamps

give off visible light. While still in an atom, the electrons are set into oscillatory motion of the order of the wavelength of light in amplitude; consequently, they can radiate. Actually the sizes of atoms are generally of the order of tens of angstrom units. Visible light has wavelengths from 3000 to 8000 Å. Beyond visible light is ultraviolet light. Ultraviolet light is invisible. It is called ultraviolet because it is of shorter wavelength than violet light, which has the shortest wavelength of visible light.

4. SOUND

Sound consists of compressions and rarefactions of a material. Since the elastic properties of different substances are different, the sound velocity varies widely. In air the speed of sound is about 300 m/sec. The wavelength of audible sound is a few millimeters to about 100 meters. The wavelength of hypersonic sound is 3000 Å, which is the same as that of visible light. The speed of sound can depend very strongly on the frequency. For high hypersonic frequencies, the frequency becomes independent of the wavelength. The minimum wavelength here is the distance between the molecules. This is the highest frequency that can exist in a simple crystal. A crystal is a solid in which the atoms are arranged in a regular array. Higher frequency sound can exist in which the molecules are set into vibration. There is no translational motion, but individual molecules are deformed and pulsate. They can also rotate. There are other vibrations in materials that are not commonly called

sound, such as spin waves. Molecules have a spin and a magnetic moment. They are like tiny permanent magnets. These magnets tend to line up in an external magnetic field, just as a compass needle does in the earth's magnetic field. They can oscillate about the field direction. There is some interaction between adjacent molecules. This interaction, or force exerted on one molecule by its neighbor, can set up a wave. Analogously to ever-present waves on the ocean, there are always sound waves in a material. They are disordered and are called noise or heat. Temperature measures the direction in which heat flows when two objects are brought into contact. The energy to be found in a mode in equilibrium at a sufficiently high temperature is proportional to the temperature. A mode is a sound wave that can exist in an object, a degree of freedom in which an object can vibrate. The number of independent sound waves that can have energy in an object is the number of modes. For reflection boundary conditions, the number is proportional to the volume of the object. Consequently, the total energy is proportional to the temperature and the size of the material. Heat is a type of energy. Two objects in contact in equilibrium then have the same average energy per mode.

5. LIGHT SCATTERING

An outgrowth of the development of lasers has been a renewed interest in light scattering. Scattered light is the name given to light that has interacted with a material other than the source.

Reflected light is the most familiar example of scattered light. Reflected light can have no wavelengths other than those incident on it. Some of the wavelengths incident on it may be absorbed, so that the scattered light can be a different color than the incident light. This occurs when sunlight containing all colors is incident on ordinary objects. Such objects absorb some light and reflect the color that they appear to be to observers. When objects are in motion relative to an observer, the frequencies can be shifted. This is a less well known phenomenon called the Doppler shift. The Doppler effect occurs for sound as well as light. The whistle of a train is heard at a higher pitch when it is approaching than when it is receding. The frequency shift is due to the Doppler effect. The light from distant stars is shifted to red because of their motion away from the earth. Red is the longest wavelength of visible light, and it has the lowest frequency. When light is reflected from a moving object, it is also scattered light. Since it has different frequencies than if the object were at rest, this kind of scattering is called inelastic light scattering. It is analogous to tennis balls thrown at a moving wall. When they bounce off the wall, they have the speed with which they hit it and twice the speed of the wall added on. The energy is different from when they were thrown as a result of collision with the wall. Collisions of this kind are called inelastic. If a tomato were thrown against the wall instead, this would also be an inelastic collision, as the tomato would be annihilated and would not have its original energy. The energy of electromagnetic

waves is related to the frequency, so shifts in wavelength are inelastic scattering. Light reflected from a mirror at rest is called elastically scattered light. The law that determines the frequency shift is conservation of momentum. Let the ball strike the wall with velocity V , and suppose the wall is moving toward the thrower with velocity v . The momentum of the ball before the collision is MV , where M is its mass. Relative to the wall, the momentum is $M(V + v)$. After the collision the ball has the same momentum, $M(V + v)$, relative to the wall. It has velocity $V + v$ relative to the wall, but now it is moving in the same direction as the wall. Its speed relative to the thrower is $V + 2v$. For a long time light was thought to consist of particles. As a result of the development of electromagnetic theory, we now know that light is an electromagnetic wave. It was found with the development of the theory of the structure of matter known as quantum mechanics that light behaves like particles when it interacts with matter. The energy of the light particles is proportional to the frequency, and the momentum is proportional to the wave vector. The wave vector is 2π divided by the wavelength, and points in the direction in which the wave moves. Conservation of momentum then says that the incident and scattered wave vectors should be different. The speed of the particles is unchanged though; it is always c , the speed of light. To calculate the change, the frequency rather than the velocity must be considered. As seen from the moving wall the frequency of the light is different. The time between wave crests is shorter,

because the wall has moved to meet the next crest. The wall moves vT , where T is the period, the reciprocal of the frequency, between crests. So the wavelength of the light as observed from the wall is reduced by VT .

$$\lambda_w = \lambda - VT.$$

The momentum is proportional to the reciprocal of the wavelength, and is changed by the factor $1/(1 - VT/\lambda)$. Since the wavelength divided by the period, or the wavelength times the frequency, is the speed of light, the momentum is increased by $1/(1 - v/c)$. In leaving the wall it has this same momentum relative to the wall. For exactly the same reason, it will be increased again when the observer sees it. The total change is $1/(1 - 2V/c)^2$, or approximately $1 + 2V/c$. The frequency is increased by $1 + 2v/c$ because of the Doppler shift. The same formula applies to sound if c is interpreted as the speed of sound. This is because sound is also a wave disturbance.

6. BRILLOUIN SCATTERING

A less familiar example of inelastic light scattering than the Doppler shift is light scattering from vibrations in a material. Light is scattered from waves in an aquarium, but because of the slow motion of the waves it is not noticeably shifted in frequency. The Doppler shift is again the reason that the light would be shifted at all. The scattered light is of high intensity, comparable to the incident light, since it can be distinguished by the naked eye. There are other vibrations in the water, of smaller

amplitude than the surface waves, but of higher frequencies. These are the waves that are associated with the temperature of the water. These thermally excited acoustic waves should cause scattering in the same way that the surface waves do. The surface waves cause scattering by changing the index of refraction of the space that was previously occupied by air. The index of refraction is momentarily that of water, but was previously that of air. If waves in the material also change the index of refraction, then light will be scattered for the same reason. The index of refraction is the ratio of the speed of light in a material to the speed of light in a vacuum. The index of refraction depends on the density of the substance. The changes in density associated with the heat causes the index of refraction to change. Light scattering should occur. Brillouin predicted light scattering of this variety in 1922.³

The maxima and minima of the sound waves move back and forth at the speed of sound. One expects from the Doppler shift that a change in frequency of $(2V/c) \times 100\%$ should occur. Here V is the sound speed, and c is the speed of light in the material. The light scattering from sound waves is ordinarily of very low intensity. It is called Brillouin scattering. The changes in the index of refraction are very small, and changes in the color of light are not noticeable. The change in frequency also is very small, as the ratio of the speed of sound to the speed of light is about 10^{-6} . The shift is a small fraction of an angstrom unit. The narrow line width of modern continuous lasers makes this shift

observable. The line width of a helium neon laser is much less than the expected shift. To observe the scattering, precise optical instruments must be used to separate the scattered and laser light. One such device is the Fabry Perot etalon, in which light is reflected back and forth between parallel mirrors. This is a high Q optical cavity. If the wavelength of the light does not divide the length of the cavity exactly, it cannot have minimum intensity on both mirrors and is damped out. Light of proper wavelength builds up to high intensities.

The interaction of a large continuous wave with a weak signal is familiar in microwave amplifiers called parametric amplifiers. There, a nonlinear element is used to amplify the signal at the expense of the continuous carrier. A third frequency is present, called the idler. The difference between the signal and carrier is the idler frequency. The interaction is coherent, as it takes place in an object of the same size as the wavelength. Incoherent interaction occurs when objects large compared to the wavelength reflect or absorb radiation, which can be adequately described as rays. Coherence can also occur in objects large relative to the wavelength. Light scattering can be described in these same terms. Generally speaking, a system generates new frequencies only if it is nonlinear. Here the scattered light is at a new frequency. The simplest nonlinearity is $y = x^2$. If the input x is made up of two frequencies, ω_V and ω_L , the sound and laser frequencies, the output will have an up-shifted and a down-shifted frequency.

$$\begin{aligned}
(\cos \omega_L t + \cos \omega_V t)^2 &= \cos^2 \omega_L t \\
&+ \cos^2 \omega_V t + \cos (\omega_L + \omega_V) t \\
&+ \cos (\omega_L - \omega_V) t
\end{aligned}$$

In the case of light scattering the interaction between the sound and light takes place over many wavelengths. Light reflected from a single maximum of the sound wave is of low intensity. Light reflected from many wavelengths in phase can be of large intensity. Consequently there is also a condition on the wavelength of the sound. Clearly a half integral number of light wavelengths must equal a sound wavelength. In terms of wave vectors we now have the relation $2\vec{k}_L = \vec{k}_V$. In general the scattered and incident light will not be collinear. There is scattering in other than forward and backward directions. The relation between the incident light and sound wave vectors is changed.

7. RAMAN SCATTERING

Brillouin scattering is only one of many kinds of inelastic light scattering. Light can be scattered in materials by other vibrations than acoustic sound. Brillouin scattering occurs for the part of the sound spectrum where frequency and wave vector laws are satisfied. Different light frequencies and directions scatter from different sound frequencies, up to a maximum frequency of scattering in the backward direction. Backward direction means toward the source of light. If the speed of sound were constant, there would be no further possible scattering from sound. Higher frequency vibra-

tions, where the molecules pulsate, do not have the same speed of sound. When molecules change states of vibration or rotation, they emit electromagnetic radiation because changes move in these molecular motions. The radiation is of lower frequency than visible light, in which electrons within the molecules change energy states. This radiation is in the infrared.

Molecules are characterized by their IR spectra, both of the absorption and emission varieties. An absorption spectrum is obtained by comparing incident and transmitted light. A source with a continuous spectrum of frequencies is used. Light is absorbed at specific frequencies rather than uniformly. The frequencies corresponding to this absorption give the absorption spectrum. If the substance is heated or an electric discharge passed through it, light is emitted in the infrared at specific frequencies. These frequencies give the emission spectrum. Infrared spectra characterize also groups of atoms. Groups of atoms occurring in different substances give similar spectra. The spectra corresponding to molecules and groups of molecules have been widely tabulated in handbooks.

As well as emitting and absorbing light, the vibrations that cause infrared spectra can scatter light. This phenomenon was discovered in the 1930's, and is called Raman scattering. The vibration or rotation occurs in separate molecules in contrast to Brillouin scattering, in which there is collective vibration of the medium. In Brillouin scattering aggregates of many molecules move back and forth, so that the substance

can be described as a continuum. None of the wavelengths are short enough to cause a continuum description to break down, and none of the frequencies cause the individual molecules to change energy states. The light frequencies could lead to absorption at the light frequency, but the optical lines of the scattering medium do not correspond to the emission frequency of the laser. Consequently, the molecules can be treated as particles; and, since the wavelengths are long, the particles can be treated as a continuum.

For Raman scattering this continuum description no longer applies. A continuum is a substance that does not appear to be made up of particles, like water ordinarily. The light interacts with molecules separately. The molecules do not interact with each other strongly, but rotate and vibrate individually. In liquids there is motion of the molecules, but the frequencies are small compared to the IR scattering frequencies. An infrared frequency is of the order of 10^{13} cps. With the sound speed of 300 m/sec, the wavelength would be 0.3 Å, which is of the order of the size of the molecules. This is too short a wavelength to interact with light. In travelling a wavelength of light, a molecule goes through 10,000 or more cycles of oscillation. The broadening of the line due to motion of the molecules is expected to be of the order of 0.01% of the infrared frequency. This broadening is called Doppler broadening. Molecules in motion experience frequencies different from the frequencies of the incident light due to the Doppler shift. In infrared spectra, light is

absorbed and emitted over a band of frequencies due to Doppler shift. The width of the line, or band, is of the order of $2v/c$, or 0.0002% of the center frequency. The wave vector condition for scattering can no longer be derived from the Doppler shift. One does expect to have the scattered and laser light interacting over long distances with a vibration wave. Also the argument based on conservation of momentum for the quanta indicates the wave vectors match. In fact the same law as for Brillouin scattering holds.

$$K_L = K_S + K_V \quad (1)$$

Here L, S, and V stand for laser, scattered, and vibration. The wave vector of a vibration wave is almost independent of the frequency. This follows from the fact that the vibration is local to one molecule and does not depend on interaction between molecules. The molecules do interact in vibrations, and Brillouin and Raman scattering are often present at the same time. Since the frequencies are so widely different, 10^{13} and 10^{19} cps, often they can be discussed separately. The only effect of the motion on Raman scattering is to broaden the Raman line. The wavelength in Raman scattering is established by a progressive difference in phase between the vibration of molecules with distance. The wave vector equation can consequently be ignored, as it is usually satisfied by many frequencies. From the description of the scattering as collision between quanta, one also expects to have a conservation of energy law. This gives a

law for the frequencies in Raman scattering. This law also follows from the description of scattering as generation of frequencies by a nonlinearity. It is the same as for Brillouin scattering.

$$\omega_L = \omega_S + \omega_V \quad (2)$$

Again, L, S, and V stand for laser, scattered, and vibration. The vibration frequency is now determined by the molecules themselves. It is a frequency characteristic of molecules rotation or vibration. For Raman scattering the frequency shift is determined independently of the laser frequency. It corresponds to a line in the emission and absorption spectra of the substance.

Scattering also occurs from spin waves and from plasmons, which are sound waves in a plasma. A plasma is a gas of electrons. The frequency and wave vector equations again hold, but the relation between scattering frequency and wave vector depends on the substance and type of vibration.

8. SCATTERED FREQUENCY

In three dimensions the equation relating the wave vectors is $\vec{k}_S + \vec{k}_V = \vec{k}_L$. Here, the bars indicate that the wave vectors may be in different directions. This situation corresponds to throwing tennis balls at a wall at an angle. The velocity in the direction of the motion of the wall will be reversed, but the velocity parallel to the wall will be unchanged. The motion of the wall is analogous to the sound wave. Wave vector $\vec{k}_V$ will be in the plane determined by $\vec{k}_S$ and $\vec{k}_L$. In this plane one has the

sum of the parallel components of $\vec{k}_S$ and $\vec{k}_L$ equal to $\vec{k}_V$. Let θ be the angle between $\vec{k}_S$ and $\vec{k}_V$. Since the frequency shift is small, $\vec{k}_V$ and $\vec{k}_L$ are approximately equal in magnitude. Both quantities are proportional to the frequency. The angle between $\vec{k}_L$ and $\vec{k}_V$ will also be θ . Consequently $2\vec{k}_L \cos \theta$ must equal $\vec{k}_V$. With $\vec{k}_L$ written as ω_L/c and $\vec{k}_V$ as ω_V/V , where V and c are velocity of sound and of light, one has

$$\omega_V = \frac{2V\omega_L \cos \theta}{c} \quad (3)$$

This formula gives the relation between the shift in the frequency of the scattered light and the direction in which the light is observed. The maximum frequency shift is found in the backward direction. In the forward direction there is no frequency shift. In the wave description the addition law for wave vectors corresponds to interaction in phase of the waves over long distances. With waves having wave vectors $\vec{k}_L$ and $\vec{k}_V$ present, there is also a wave with wave vector $\vec{k}_L + \vec{k}_V$ present as long as there is nonlinearity. This occurs for the same reason that different frequencies give sum and difference frequencies.

$$\begin{aligned} (\cos k_L z + \cos k_V z)^2 &= \cos^2 k_L z \\ &+ \cos^2 k_V z + \cos(k_L + k_V)z \\ &+ \cos(k_L - k_V)z \end{aligned}$$

If there is light of wave vectors $\vec{k}_L$ and $\vec{k}_S$ present, the difference is a wave of wave vector $\vec{k}_V$. Sound waves of other wave vectors will interact with $\vec{k}_V$, but the effect at different places will be

opposite owing to the lack of phase matching. The cumulative effect over the whole volume will be small.

In quantum theory the momentum of a particle is equal to the wave vector times Planck's constant, $P = \hbar k$. The addition rule for wave vectors has the form of a momentum conservation law. In the interaction with matter, light appears to be made up of particles. These particles are called photons. Snell's law in geometrical optics states that the ratio of the sine of the angle of incidence to the sine of the angle of refraction equals the ratio of the indices of refraction. The speed of light is inversely proportional to the index of refraction. Since the frequency is unchanged when the light passes from one material to another, the wave vector is proportional to the indices of refraction. Snell's law states that the parallel momenta in the two substances are equal. When light interacts with objects whose size is of the order of a wavelength the light acts like a wave. This is the case for small holes, where diffraction of the wave is observed. Diffraction is the occurrence of patterns of light different from the image of the hole through which it shines, if the image is constructed by drawing straight lines from the source through the hole. Somewhat surprisingly, light again acts like a particle when it interacts with an object of smaller size than its wavelength (e. g., with an atom). It is absorbed in amounts of energy that are integral multiples of Planck's constant times the frequency. The momentum is identified with Planck's constant times the wave vector. The two basic laws for Brillouin scattering are:

$$\omega_L = \omega_S + \omega_V,$$

and

$$\vec{k}_L = \vec{k}_S + \vec{k}_V.$$

The symbols can be displayed as

These laws correspond to the conservation of wave energy and momentum. Equation (3), which was derived previously from the Doppler shift,³ can also be derived from the laws for Brillouin scattering.

The frequency shift for Brillouin scattering gives a value for the speed of sound in a material. The width of the Brillouin shifted line is of interest as well. The width depends on the damping of the sound waves in the medium. The laser light interacts with one space mode of sound. The shift in the frequency produces a negligible change in the wave vector, since the frequency shift is so small compared to the light frequency. Since the broadening is of smaller amount than the shift, it also does not change the wave vector. The sound wave in a mode lasts for a short time. It is only if a sound wave lasts for a long time that its frequency is known precisely. If the wave is observed for time T and the cycles are counted, the number will be in error by about 1 cycle. The error then is 1 in ωT , so the expected error is $1/\omega T$ times ω . The spread in frequencies is equal to $1/T$, the time for a wave to

die out, where T is called the phonon lifetime. Using the Fabry Perot etalon or light filter, the width of the Brillouin line has been measured and phonon lifetimes calculated. The phonon lifetimes are tools in other scientific investigations. If a chemical reaction is taking place, the phonon lifetime gives information on how fast the reaction is proceeding. If there are large fluctuations in density, like those near the critical point of a substance, the phonon lifetime is also important.

The spectra of Brillouin scattering and Raman scattering for a low intensity steady laser are used to determine phonon lifetimes. When the spectrum is narrow, as in Rayleigh scattering, self-beating of monochromatic laser light is used. When the spectrum is wide, as for Brillouin scattering, the spectrum is measured using Fabry Perot spectrometer. The spectrum is approximately Lorentz and the phonon lifetime T is related to the width of the line by $\Delta\omega = \frac{1}{T}$. These spectroscopic methods of determining line width apply to liquids, gases, and solids. They are also used to determine the processes responsible for the line width. One source of line broadening is viscosity of liquids. The accuracy of the determination of line width is about 10%. When the density is so low that the collision time between molecules is of the order of the line width, other treatments must be used, as a phenomenological phonon lifetime no longer provides an acceptable description of damping of the sound. Low intensity scattering is observed in directions other than backward, although backward scattering is generally the most intense because

the interaction length is largest in this direction. Stimulated Brillouin scattering is observed only in the backward direction. Both backward and forward Raman scattering are observed.

Light scattering is used as a tool to investigate other phenomena in materials, such as fluctuations. Properties of density fluctuations, like dependence on temperature, are of interest particularly near the critical point of a substance where they become very large. Specific heats determine the damping time of the fluctuations. The velocity of sound in materials can be determined from the frequency shift for Brillouin scattering by setting $\theta = 0$ in Eq. (3). The resulting formula is:

$$\omega_B = \frac{2V}{c} \omega_L$$

9. STIMULATED BRILLOUIN SCATTERING

There is a branch of light scattering that is concerned with very high intensities of light. The intensities are only obtainable with pulses from a pulse laser. This type of scattering is called stimulated light scattering. It is different from ordinary light scattering in that the vibration is strengthened by the light. This corresponds to a tennis ball being thrown so hard at a moving wall that it speeds the wall up. Since the ball on the rebound is travelling slower than when approaching the wall, the ball has lost energy. This energy has been given to the wall. If the wall is moving toward the thrower, the ball is speeded up and the wall slowed down. Here the ball gains some energy and the wall loses some. The situation is more complicated when

the wall is moving obliquely or the ball is thrown at an angle. In light scattering, the change in frequency of the scattered light is given by the two laws expressed in Eqs. (1) and (2). These relations are also obeyed in stimulated light scattering. If the scattered light is of higher frequency than the laser light, the photons are more energetic. The difference in energy is made up by the sound wave, which loses energy. This kind of scattering is called anti-Stokes scattering. If ω_v is smaller than ω_L , laser energy is contributed to the sound wave. Thus, every scattered photon has lower energy than an incident photon. The difference in energy is a phonon energy. When the incident light is not strong, the increase in the sound is dissipated in the medium before it grows to any appreciable level. With high intensity laser pulses, the sound can be increased to large levels before it distributes itself among other vibration modes in the solid as heat. It appears as a large and monochromatic wave, and subsequently increases the temperature of the medium slightly. In this way high intensity laser light brings about an opacity of the medium at the laser frequency. The sound increases to such high levels that almost all of the laser light is scattered. The energy removed from the laser beam to give the high sound level is quite small, as the ratio of the energies of the photons to phonons is $2v/c$, or about 10^{-6} . A steady state occurs when the sound in the medium reaches a level where most of the light is scattered. The reason for the steady state is that the laser light is weakened in the scattering process. The higher the

sound amplitude, the nearer to the front of the scattering cell is most of the beam scattered. Further along the scattering cell the laser beam is weaker and less effective in causing the sound amplitude to build up. The sound wave has its minimum amplitude at the end of the scattering cell farthest from the laser. It builds up to a maximum at the front of the cell. When a steady state exists, the amount of energy left in the cell is only 10^{-6} of the incident energy. An incident photon enters the cell, is changed into a scattered photon, and leaves the cell. The number of photons leaving is equal to the number incident. With 100% conversion, the sound intensity is increased by 10^{-6} of the laser energy. The reason is that 10^{-6} is the relative difference in energy of incident and scattered photons. The amount of energy left in the medium in the steady state is small. Laser pulses of power LMW, lasting 20 ns leave 20×10^{-3} joule of energy in the scattering cell. This is not enough energy to raise the temperature of a cubic centimeter of water even 10^{-3} degrees.

The specific subject of this thesis is the description of scattering of this sort, stimulated light scattering. Stimulated Brillouin scattering was discovered in 1964;⁴ it is an area for which few applications have been found. Its uses are scientific, such as the measurement of the phonon lifetime in substances. A description of the stimulated scattering process, more detailed than the spectrum of the scattered light, can be given. For Brillouin scattering, a transient time passes while the sound intensity builds up to a high enough level

to scatter most of the light. The time required for this transient can be calculated. The intensity of the scattered light as a function of time can also be computed. This is not a problem of fundamental theoretical importance, but involves the use of well known tools. In particular, Maxwell's equations and hydrodynamics give a description. Descriptions using quantum ideas have also appeared.⁵ When light intensities are sufficiently high, a classical description is accurate.

10. STIMULATED RAMAN SCATTERING

In addition to stimulated Brillouin scattering, there is stimulated Raman scattering, which was discovered in 1962.⁶ It is similar to ordinary Raman scattering, but the intensity of the vibration is again increased by the light. The damping time of the vibration is much shorter than for Brillouin scattering. The damping time determines how long the transient scattering lasts. Having a shorter lifetime, transient Raman scattering is of much briefer duration than transient Brillouin scattering. This difference leads to qualitative differences between Raman and Brillouin scattering. Again laser pulses are made necessary by the high intensities required to give stimulated scattering. A Raman pulse can build up in a short time compared to that required by light to traverse the cell. In backward Raman scattering, Raman pulses arise at the end of the cell farthest from the laser and then travel toward the laser. They sweep out all the energy in the cell and grow to large intensities relative to the laser intensity,

Brillouin scattering never exceeds the laser intensity. Aside from being a source of short pulses, applications for this phenomenon have not been found. Backward Raman scattering is treated here. There is also forward Raman scattering, since the wave vector conservation is not required. There is no forward Brillouin scattering. Stimulated Brillouin scattering is always Stokes, or down-shifted, as energy must be added to the sound.

Filamentary effects during self-focusing and amplification of noise give a stochastic character to the scattering, even when laser emission is known.

Striking patterns of colors can be produced when higher order Raman emission occurs. Since the setup is cylindrically symmetric, different colors occur in rings of different radii.

Raman emission in the forward direction has been investigated experimentally and theoretically.⁶ Picosecond pulses are used, and mode structure of the laser adds another random parameter.

Here the anti-Stokes emission does not satisfy the wave vector and frequency conditions and is absent. However, induced anti-Stokes absorption is to be expected, if a primary source of shifted light is present. The spectrum of the absorption has been measured. Time dependence of transmitted pulses is also of interest, both for Brillouin and Raman scattering. The absorption spectra are significant as they are characteristic of the substance or substances in the scattering cell. The fraction of the light incident that is absorbed has been found to be proportional to laser power, in

accord with a weak exponential gain of the form:

$$e^{g I_L z}$$

Stimulated Brillouin and Raman emission often occur simultaneously. Fortunately the time scale for Raman emission is much shorter than for Brillouin emission, so that the Raman pulse occurs prior to and with a briefer duration than the Brillouin scattering. Therefore the two processes can be considered separately, as neither scattered intensity is large when the other is.

Phonon echoes occur when the time scale of scattering is long compared to the time required for sound to traverse the scattering cell. Then boundary conditions on the sound are important. In the case that we consider, these boundary conditions are not important. The duration of the laser pulse is 50 nsec, during which the sound propagates only 0.05 mm. When the distance between the laser and scattering cell is small compared to the distance light travels during the experiment, multiple Brillouin scattering is observed. In this phenomenon scattered light reenters the laser and is amplified, and subsequently produces additional shifts when it illuminates the scattering cell. With short pulses this can be eliminated by long path lengths between the laser and scattering cell.

11. APPLICATIONS

Stimulated light scattering is a topic of interest to scientists as a tool to study

properties and phenomena of materials. One of the scientific applications of inelastic light scattering is the study of the relaxation of fluctuations in density in materials.⁷ This is of interest when a chemical reaction takes place, as the rates of change of substances can be measured. It is also of interest when a material nears its critical point where fluctuations become very large. The phenomenon of critical opalescence occurs when transparent material turns white at its critical point. For the measurement of rates of relaxation, low intensity light is enough because only the spectra of the scattered light need to be measured. Infrared spectra of molecules or molecular groups is called Raman scattering. Low intensity or ordinary Raman scattering has long been used as a tool to identify substances in mixtures in Raman spectroscopy. Lasers are used here as a source of nearly monochromatic light.

An offshoot of the field of inelastic light scattering is stimulated light scattering or nonlinear optics. It has been found that the high intensities in pulse lasers gives very large scattering efficiencies. Nearly all the incident light is scattered, hence the name stimulated. Early work was directed at measuring the spectra of the scattered light. With fast detection systems it is possible to observe the time development of the scattering. This approach gives a detailed description of the scattering process.

II. Light Scattering

1. ORDINARY LIGHT SCATTERING

Light incident on matter is scattered in various directions, and often the scattered light is at a different frequency than the incident light. This change in frequency spectrum is usually caused by the absorption of some frequencies in the incident light and the reflection of others. This behavior characterizes a linear system. No new frequencies are generated, but some can be emphasized and others attenuated. A less familiar phenomenon is the shift in frequency of monochromatic light in matter. This change is caused by nonlinear properties of matter, and is called nonlinear optics. The shift results from the interaction of light with a vibration. The vibration is said to scatter light inelastically if the frequency of the scattered light is different from the incident light. The term inelastic also refers to collision where energy is not conserved. The same term is used because the energy of light is associated with the frequency.

One vibration that scatters light is an acoustic wave. This kind of scattering is called Brillouin scattering. The frequency shift for Brillouin scattering is always small compared to the frequency of the light. The reason is that light and sound wavelengths have to be comparable, and the speed of sound is very much smaller than the speed of light. The frequency of the vibration equals the frequency shift of the light.

Raman scattering is light scattering by a molecular vibration or rotation.

The vibrations cause the infrared spectra of materials. The wavelengths corresponding to different molecules and groups of molecules have been tabulated at length in handbooks. In Raman scattering the shifts in the light frequency are in the infrared. Often the light itself is visible. Raman scattering was investigated in the 1930's. In these investigations highly filtered light sources were used as monochromatic light. The measurement of the linewidth in Brillouin scattering was made possible by the monochromaticity of laser light. The linewidth of the scattered component is important as it is determined by the lifetime of the acoustic wave.

2. STIMULATED LIGHT SCATTERING

At high incident light intensities stimulated light scattering occurs. The incident light strengthens the material vibration in both Brillouin and Raman scattering. The strengthened vibration in turn gives rise to more scattered light. This stimulated scattering process builds up as an instability. Its growth is limited only by the power in the scattering beam. Eventually the vibration makes the material opaque to the laser beam and the beam is absorbed. The laser beam intensity is absorbed near the surface of the scatterer. The vibration wave does not grow further, and the scattering becomes steady. The vibration wave amplitude increases exponentially from a low level at the end of the scattering cell farthest from the laser.

When light is enclosed in a reflecting container it exhibits quantum properties as does sound. The reason is that radiation exists only in a set of modes characteristic of the container. These modes are determined by the boundary conditions. They are spatial modes. The energy in the container is a sum of the energies in each of the modes. Associated with each mode is a frequency. The energy in a single mode has two forms: kinetic and potential, or electric and magnetic. The modes are completely analogous to harmonic oscillators, and energy exchanges can occur only in discrete quanta. For light the quanta are called photons and for sound, phonons.

In stimulated scattering the vibration is strengthened by the light. In terms of quanta, the scattering process can be explained as follows. A laser photon is destroyed while a scattered photon and phonon are created. Here a laser photon is a quantum of energy with the laser frequency. A scattered photon has the Brillouin or Raman shifted frequency, which differs from the laser frequency by the frequency of the phonon. The conservation laws of energy and momentum apply to the scattering process. Associated with a quantum is the energy $\hbar\omega$ and momentum $\hbar\mathbf{k}$. These are basic quantum laws that allow the association of particles and waves. Conservation of energy for the quanta is given by

$$\hbar\omega_L = \hbar\omega_S + \hbar\omega_V, \quad (4)$$

where the subscripts L, S, and V stand for laser, scattered, and vibration. The left side of the equation is the energy of a laser photon prior to scattering. After

the interaction, the laser photon is replaced by a phonon and a scattered photon. The right side of the equation is the energy of the scattered photon and phonon. A momentum conservation equation holds as well. The direction of the momentum vector is the direction of the propagation of the wave. The momentum conservation equation is obtained by equating the momentum of an incident photon and its decay products, a scattered photon and a sound phonon:

$$\hbar\mathbf{k}_L = \hbar\mathbf{k}_S + \hbar\mathbf{k}_V. \quad (5)$$

Again L, S, and V stand for laser, scattered, and vibration, where vibration can be either acoustic or molecular for Brillouin or Raman, respectively. The symbol $\hbar$ is Planck's constant over 2π , and ω and $\mathbf{k}$ are angular frequency and wave vector.

3. BACKWARD SCATTERING

This thesis is confined to the problem of backward scattering or scattering toward the laser. There is no stimulated Brillouin scattering in the forward direction. The reason is that the energy and momentum conservation laws Eqs. (4) and (5) cannot be satisfied in this case. For Raman scattering the frequency of the molecular vibration is quite independent of the wave vector. Consequently there can be scattering in the forward direction, as only energy conservation law (Eq. 4) need be satisfied. Forward Raman scattering is another topic and is not treated here. Equations for the frequencies and wave vectors are derived from the energy and momentum laws by dividing by $\hbar$.

$$\omega_L = \omega_S + \omega_V \quad (6)$$

$$\vec{k}_L = \vec{k}_S + \vec{k}_V \quad (7)$$

Evidently the sound frequency is the difference between laser and scattered frequencies.

For backward scattering the wave vector of the scattered light points in the opposite direction to the incident light. Using vectors, this is written $\vec{k}_S = -\vec{k}_L$. The scattered and laser wave vectors are very nearly equal in magnitude, since the frequency shift is minute compared to the light frequencies. Even for Raman scattering the shift is only an infrared frequency.

For Brillouin scattering the frequency and wave vector are related by the speed of sound. The relation is

$$\omega_V = v k_V. \quad (8)$$

That v is the speed of sound is established as follows. The group velocity of a wave is given by the derivative of the frequency with respect to wave vector. The group velocity gives the speed at which information travels. Consequently the constant V must be the speed of sound. The magnitude of wave vector is denoted by k without a bar. A similar relation holds for the light frequencies and wave vectors:

$$\omega_L = c k_L \text{ and } \omega_S = c k_S. \quad (9)$$

The symbol c represents the speed of light. Substituting $k_S = -k_L$ into the wave vector conservation law gives $k_V = 2k_L$. Writing the wave vectors in terms of frequencies gives

$$\frac{\omega_V}{V} = \frac{2\omega_L}{c}.$$

Finally, using the frequency conservation condition Eq. (6),

$$\omega_S = 1 - \frac{2V}{c} \omega_L.$$

For Raman scattering the conservation of wave vector (Eq. 5) is satisfied approximately independently of frequency. A very small change in the frequency gives a large wave vector. If the molecules did not interact at all, there would be no relation between frequency and wave vector of the sound wave. Since this is never true in practice, there is some very slight dependence of frequency on wave vector. The frequency is determined by the IR spectral lines of the molecules, that is, individual molecular energy levels. These levels are generally rotational or vibrational. For Raman scattering the frequency is treated as determined by molecular structure.

4. CLASSICAL DESCRIPTION

A classical description applies to light scattering when the light is intense and there is no reflection. There are many quanta in the wave and no measuring apparatus used is delicate enough to distinguish energies of the size of a quantum. The radiation appears to be continuously distributed in energy, as is the case for ordinary light. No quantum effects are apparent until the light interacts strongly with individual atoms. In Brillouin scattering the light interacts only with large aggregates of atoms, changing only their motion. An analogue is the reflecting light vanes suspended in glass containers and painted black on one

side. Sunlight turns the vane as a result of pressure exerted by light. A quantum description is not required. Quantum descriptions are most successful when boundary conditions are simple, so that well defined modes exist, or for the case of atoms and molecules when there are no boundaries. Here the boundary conditions do not produce modes, and the system is not infinite. In Raman scattering the light does interact with individual molecules and atoms, and a quantum description applies. It is sometimes called semi-classical, as the light is treated classically and the atoms by quantum theory. Assuming the matter has a vibration coordinate that influences the index of refraction allows a complete classical treatment. For low intensity scattering a quantum description should be used. Here quantum ideas are used to evaluate the noise source, which alone is responsible for low intensity scattering.

Classical equations for sound and light amplitudes are derived using Maxwell's equations and either the Navier Stokes equations or a molecular vibration equation restricted to one space variable. One space variable is used in the equations as all growth is concentrated in the longitudinal direction. Radial derivatives are small compared with longitudinal derivatives, except when self-focusing is present. Self-focusing is treated approximately here by increasing the laser intensity near the end of the scattering cell. Coupling between sound and light waves is caused by the dependence of the index of refraction on both light and sound intensities. This appears as polarization depending on light and sound variables. To describe a coherent

interaction, the amplitudes are written as a slowly varying quantity multiplying a plane wave. The light and sound vary slowly over a wavelength and in a period of wave oscillation. A plane wave is the solution to wave equations in one space variable. The source of the acoustic vibrations is a random term added to the vibration equation. The equations describe both Brillouin and Raman scattering.

In the scattering experiments the sound in a scattering cell becomes intense enough after many phonon lifetimes to scatter most of the light. Initially the sound is of very low intensity. It doesn't build up instantaneously, as it requires considerable energy. Initially the cell is almost transparent to the laser light. The interaction is weak. Subsequently, the interaction becomes stronger, and the sound builds up faster. After many phonon lifetimes, it reaches a steady state where little light penetrates into the medium, and where most of the light is scattered near the front end of the cell. To obtain formulas for the buildup of intensities, Kroll⁸ solves the equations for transient Brillouin scattering. He uses constant laser intensity, because prior to steady scattering the laser beam is only slightly affected by the sound. He keeps a space derivative of the sound amplitude, which is neglected by Hagenlocker⁹ and by Carman¹⁰ for Raman scattering. This term is small compared to a time derivative as it is multiplied by the velocity of sound. Space derivatives multiplied by the velocity of light are kept. This term could be important in the steady state. Grob¹¹ keeps it and describes the growth of sound from zero at the front of the cell.

If the mesh boundary is assumed to be inside the scattering cell, the sound will be large at the left boundary. If the mesh boundary is taken to be in the surface of the scattering cell, the sound will be zero at both ends. In the numerical work presented here, the boundary conditions of both zero and peak sound at the front of the cell were used. The difference was not remarkable, as growth of light and sound in the first mesh interval, although larger than in other mesh intervals, is relatively small. Maier and Renner¹² give a solution with varying laser intensity for transient scattering.

A numerical solution for Brillouin scattering is computed here. A new model of the scattering process is used. The differential equations are the same as those used by other investigators, but a distributed noise source is used. This noise source is a random term added to the vibration equation. Since the equations cannot be solved analytically, numerical methods are used to obtain approximate solutions. The aim is to calculate the consequences of the model, and to obtain new understanding of Brillouin scattering. Also additions are made to the mathematical analogue so that it is more representative of the physical experiments. Stimulated scattering is computed as a function of radius and the scattered power obtained by integrating over beam cross section.

The difference between average transient scattering and beam center transient scattering, as given by Maier and Renner,¹² is a much less abrupt threshold. The light approaches the steady-state scattering condition more gradually. Steady-state scattering

appears to occur near the peak of the pulse, where the laser intensity is changing least rapidly in time. Different noise strengths are tried, as there is no unanimous agreement on the magnitude. The energy in a mode is known, but there is disagreement over the number of modes. The scattering is found to be quite independent of noise constant. Statistical fluctuations are calculated.

The Raman and Brillouin scattering described here are a part of a large literature concerning inelastic light scattering. A plane laser wave is assumed. Some experiments have been done with circularly polarized light, but often the end of the laser is a Brewster angle window in which the reflection is high only for one direction of polarization and the light is consequently linearly polarized. Multimode effects in which the laser emission occurs over a wide range of frequencies have been observed, particularly for the generation of short (picosecond) laser pulses. Here the laser is single mode, as is evidenced in spectroscopic measurements of the emission where the linewidth is given by the duration of the pulse. Pulse durations of 15 ns are typical of ruby laser emission.

Associated with Raman scattering is generation of light in higher orders. The scattered light becomes intense and acts in place of the laser beam. This is observed primarily in forward scattering. It is observed also to a small extent in backward scattering.

Both Brillouin and Raman scattering in solids must be described by susceptibility tensors. In the simpler case of

liquids and gases, where birefringence is not observed, the polarization of the scattered light and of the laser beam are

the same. A nonlinear polarization proportionality constant then suffices to describe the scattering.

III. Description of Experiments

Raman scattering is different from Brillouin scattering in part because the phonon lifetime is much shorter. Pulses computed for Brillouin scattering are of the order of nanoseconds, while Raman pulses are picoseconds in length. The model with distributed noise source for Raman scattering is also new. The same type of noise is used as for Brillouin scattering. Backward scattered pulses are computed to obtain durations and peak intensities of the pulses. Large statistical variations in both variables are found. Peak intensities are comparable to experimental values, but time durations are longer.

Self-focusing is the source of the Raman pulse in liquids. In the experiments of Maier¹³ a ruby laser pulse is directed into a cell containing carbon disulphide. The light focuses itself by changing the index of refraction in such a way that the light concentrates near the axis of the cell. This phenomenon is called self-focusing.¹⁴ Light is observed in filaments with diameters of a few microns at the end of the scattering cell farthest from the laser.¹⁵ Since there is no stimulated Raman scattering without filaments, it is concluded that the filaments are the source of Raman scattering. What is observed is very short pulses emitted toward the laser.

The light scattering experiments can be schematized as shown in Fig. 1.

Fig. 1. Schematic of light scattering experiments, where L is laser, PD is photodiode, and SC is scattering cell.

The photodiodes measure laser and scattered light reflected by a beam splitting plate. Other apparatus such as light filters to separate Raman and laser light, lenses to focus the light, etc., are also used. Rates of growth of pulses in space are determined by placing glass plates inside the scattering cell. The gain is determined in this way. The pulse from the photodiodes is displayed using oscilloscopes. Since the pulse length is much shorter than the response time of oscilloscopes, it must be measured by other methods. An autocorrelation method is used in which the pulse is split and sent over different light paths. It is then recombined in a substance that indicates how the pulses overlap. Sometimes two-photon fluorescence is used, where light is emitted at twice the laser frequency when the pulses recombine. When the light paths are the same, the peak intensity is large. When the pulses do not arrive at the same time at a given

place, a smaller peak intensity is obtained. The pulses are absorbed when they overlap, as the threshold for two-photon fluorescence is exceeded. The total energy is measured on photographic plates. By varying the light paths a known distance, various delays are obtained.

For Brillouin scattering similar apparatus is used. Brillouin and Raman scattering can occur in the same scattering experiment. Usually either one or the other occurs, because the gain of Raman dominates the gain of Brillouin.

The pulses obtained in stimulated Brillouin scattering are of the same order of magnitude duration as the laser pulses. The laser pulses are 15 nsec in width. Consequently, Brillouin pulses can be reproduced on oscilloscopes, and no autocorrelation is needed to measure width. Again, the gain is obtained by immersing plates in the scattering cell to determine rate of growth in space. Phonon lifetime is obtained from spontaneous scattering line widths. A Lorentz shape is assumed for the line. This corresponds to a simple phenome-

nological damping time in the vibration wave equation. The wave equation becomes

$$\frac{d^2 y}{dt^2} + \frac{1}{T} \frac{dy}{dt} + \omega_0^2 y$$

equal driving terms. Substituting $i\omega$ for derivatives gives a spectrum proportional to

$$\left(\omega_0^2 - \omega^2 + \frac{i\omega}{T} \right)^{-1}.$$

The real part of this expression involving the square root of -1 , written i , is taken. Replacing $\omega_0^2 - \omega^2$ by $2\omega\Delta\omega$ gives a spectrum proportional to

$$\left[(\Delta\omega)^2 + \left(\frac{1}{2T} \right)^2 \right]^{-1}.$$

Half power occurs when $\omega = 1/2T$, so the full width at half power is $1/T$. Here, ω is angular frequency and T is phonon lifetime. The spectrum for stimulated Brillouin scattering is much narrower, as is expected for a system with considerable gain. The phonon lifetime is determined from low intensity ordinary scattering.

IV. Differential Equations

1. MODEL

The model applies to an object resembling a scattering cell. The model is assumed to be one dimensional for Raman scattering and two dimensional (length and radius) for Brillouin scattering. The side boundaries of the cell are assumed to have no influence. All reflections at the ends also are assumed

to vanish. This corresponds to anti-reflection coatings on the ends. The laser is not a part of the model, as its effect is limited to giving a pulse of known shape. Nor is any of the measuring apparatus included in the model. The model is limited to a somewhat simplified scattering cell of 30 cm length and with physical constants corresponding to ethyl ether for Brillouin scattering and CS_2 for

Raman scattering. For the two-dimensional calculation, a pulse with Gaussian radial dependence of the intensity is assumed. The Gaussian dependence is described by

$$e^{-\left(\frac{r}{r_0}\right)^2},$$

where r is the radius, r_0 is a constant, and e is the base of the natural logarithms. Noise in the medium is modeled by means of a random source term in the partial differential equations. The model geometry for differential equations is given in Fig. 2.

2. MAXWELL'S EQUATIONS

Partial differential equations describing stimulated Brillouin scattering are derived from Maxwell's equations and Newton's laws applied to fluids. Ampere's law and Faraday's law of induction are:

$$\nabla \times \vec{B} = \frac{1}{c} \frac{\partial \vec{D}}{\partial t} \quad (10)$$

and

$$\nabla \times \vec{E} = -\frac{1}{c} \frac{\partial \vec{B}}{\partial t} \quad (11)$$

For transparent materials there is no conduction current. If there were conduction current, there would be joule heating. Some of the light would change to heat, and the material would not be transparent. Associated with Maxwell's equations are constitutive relations by which properties of macroscopic materials are accounted for. An electric field polarizes atoms. The polarization changes the field inside a material. If

Fig. 2. Model geometry for differential equations.

there is gain it increases the field. A given charge causes a smaller electric field in the absence of gain. Coulomb's law, which gives electric field proportional to charge, is modified by multiplying by the ratio of fields in the material and in space.

In Maxwell's equations an effective electric field D is used, and materials are otherwise ignored. The true electric field is obtained by dividing by the polarization constant, which is called permittivity, and is conventionally written ϵ . The displacement D is proportional to the electric field by the permittivity.

$$D = \epsilon E$$

The permittivity depends on the density of material. Dense material has different polarization than tenuous material. The permittivity is expanded about the steady density; higher order terms are neglected, as changes in the density are very small.

$$\vec{D} = \left[\epsilon_0 + \left(\frac{\partial \epsilon}{\partial \rho} \right)_0 \rho_1 \right] \vec{E}$$

Here ϵ_0 is permittivity in equilibrium, $\frac{\partial \epsilon}{\partial \rho}$ is the rate of change of ϵ with density in equilibrium, ρ_1 is the departure of the density from its equilibrium value, and

E is the electric field. When there is absorption in the medium, the permittivity also varies because of temperature changes. Scattering from temperature fluctuations is called Rayleigh scattering. Scattering from acoustic waves produced by absorptive heating is called stimulated thermal Brillouin scattering. To describe stimulated thermal Brillouin scattering a diffusion equation for heat flow must be used as well as Maxwell's equations, and the Navier-Stokes equation. When transparent materials are used there is very little heating, so that ϵ depends only on density.

3. VECTOR IDENTITIES

The del symbol in Maxwell's equations indicates either curl or divergence. If a cross follows del, it is curl, and if a dot follows del, it is divergence. Curl is an abbreviation for

$$\left(\frac{\partial A_z}{\partial y} - \frac{\partial A_y}{\partial z}, \frac{\partial A_x}{\partial z} - \frac{\partial A_z}{\partial x}, \frac{\partial A_y}{\partial x} - \frac{\partial A_x}{\partial y} \right)$$

considered as a vector where $\vec{A}$ is the vector whose curl is taken. The divergence of $\vec{A}$ is a scalar, that is, a number, equal to

$$\frac{\partial A_x}{\partial x} + \frac{\partial A_y}{\partial y} + \frac{\partial A_z}{\partial z}.$$

The subscripts on A indicate the Cartesian components of the vector in the x , y , and z directions. The z axis is assumed to be the axis of the scattering cell. The curl of Eq. (11) is taken. The curl commutes with the time derivative resulting in:

$$\nabla \times \nabla \times \vec{E} = \frac{1}{c} \frac{\partial (\nabla \times \vec{B})}{\partial t}. \quad (12)$$

Using the value of $\nabla \times \vec{B}$ given in Eq. (10), we get:

$$\nabla \times \nabla \times \vec{E} = -\frac{1}{c} \frac{\partial^2 \vec{D}}{\partial t^2}. \quad (13)$$

The curl of the curl can be simplified to $\nabla(\nabla \cdot) - \nabla^2$. This is a vector identity. It can be derived with shorthand notation for the curl as $\epsilon_{\alpha\beta\gamma} A_{\gamma,\beta}$. Here γ is a component of A , and β indicates a partial derivative with respect to coordinate β . The symbol $\epsilon_{\alpha\beta\gamma}$ is a number that equals 1 when α, β , and γ are in cyclic order, (i.e., xyz, yzx, xzy) and -1 when $\alpha\beta\gamma$ are in acyclic order (i.e., yxz, zyx, xzy). If two indices are repeated, $\epsilon_{\alpha\beta\gamma}$ equals zero. The curl of the curl is $\epsilon_{\alpha\beta\gamma} (\text{curl } A)_{\gamma,\beta}$. Two quantities after the comma indicate two partial derivatives.

Another identity is used for the product of two ϵ symbols. It is $\epsilon_{\alpha\beta\gamma} \epsilon_{\gamma\alpha\beta} = \delta_{\alpha\alpha} \delta_{\beta\beta} - \delta_{\alpha\beta} \delta_{\beta\alpha}$. When two letters are repeated, they are summed on. The delta symbols are ones when the subscripts are equal; otherwise, they are zeros. This formula is verified by substitution. For instance, if $\alpha = a$ and $\beta = b$, there is only one letter of xyz left for γ . If $\alpha\beta\gamma$ is cyclic, then $\gamma\alpha\beta$ is cyclic also; and both must be acyclic if either is. Consequently, the product is 1 unless α and β are also equal. In the latter case, it is zero. The right hand side assumes the same values. The curl of the curl becomes $(\delta_{\alpha\alpha} \delta_{\beta\beta} - \delta_{\alpha\beta} \delta_{\beta\alpha}) A_{b,\alpha\beta}$. We sum on the repeated indices to get $A_{b,\alpha\beta} - A_{\alpha,aa}$. This expression is $\nabla(\nabla \cdot A) - \nabla^2 A$ in the α direction. Equation (13) becomes

$$\nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} = -\frac{1}{c^2} \frac{\partial^2 \mathbf{D}}{\partial t^2}. \quad (14)$$

It is assumed that $\mathbf{E}$ is a transverse plane wave, that is, that the direction $\mathbf{n}$ of $\mathbf{E}$ is perpendicular to the direction of propagation. This holds true in an infinite medium with no free charge sources for $\mathbf{D}$. By Coulomb's law, another of Maxwell's four equations is

$$\nabla \cdot \bar{\mathbf{D}} = 4\pi \times \text{free charge density}.$$

If $\nabla \cdot \bar{\mathbf{D}} = 0$, then $\nabla \cdot \bar{\mathbf{E}} = 0$, since $\mathbf{E}$ is proportional to $\mathbf{D}$, and $\mathbf{E}$ must be perpendicular to the direction in which it varies. The wave equation for electric field becomes

$$\nabla^2 \bar{\mathbf{E}} = -\frac{1}{c^2} \frac{\partial^2 \bar{\mathbf{D}}}{\partial t^2} \quad (15)$$

4. FLUID EQUATIONS

Newton's second law for fluids is

$$\rho \frac{d\bar{\mathbf{v}}}{dt} = -\bar{\nabla} p \quad (16)$$

Here ρ is density of material, $\bar{\mathbf{v}}$ is velocity of a fluid element, and p is pressure. The equation equates the mass of a fluid element times its acceleration to a force, the gradient of the pressure. This force is actually the difference of the pressures pushing on two sides of the fluid element. The density and velocity also satisfy a continuity equation since if the velocity of the fluid at all points near a point is directed toward the point, the density must increase at that point. This principle gives the following equation:

$$\frac{\partial \rho}{\partial t} + \bar{\nabla} \cdot \rho \bar{\mathbf{v}} = 0 \quad (17)$$

The divergence is the total rate of flow into a cubic volume element. Each derivative gives the difference in the rate of flow across opposite sides, divided by the distance between the sides. The sum of the three derivatives must be balanced by the rate of change in density. A similar argument applies to Coulomb's law, where the total flux of the electric field, $\nabla \cdot \mathbf{E}$, is proportional to the charge contained in the volume. Since the density is nearly constant, while $\bar{\mathbf{v}}$ is small, $\nabla \cdot (\rho \bar{\mathbf{v}})$ is approximately $\rho_0 \nabla \cdot \bar{\mathbf{v}}$. Taking another derivative of the continuity equation gives

$$\frac{\partial^2 \rho}{\partial t^2} + \rho_0 \nabla \cdot \frac{\partial \bar{\mathbf{v}}}{\partial t} = 0.$$

Newton's law is used to substitute for $\rho_0 \frac{\partial \bar{\mathbf{v}}}{\partial t}$, and gives

$$\frac{\partial^2 \rho}{\partial t^2} = \nabla^2 p \quad (18)$$

The symbol ∇^2 is an abbreviation for divergence of gradient.

The pressure in the acoustic waves is due to two sources. One is the change in density, the other is electrostriction. The density increase associated with an increase in pressure is common experience, while electrostriction is less well known. Electrostriction is similar to piezoelectricity, where a crystal contracts because of an applied electric field. The change in polarization exerts forces on the material. In electrostriction the same phenomenon occurs in a much wider variety of substances. Applying an electric field

results in energy storage in the medium. To minimize this energy, the medium tends to shrink. The force exerted is proportional to electric field squared, and consequently is smaller than the piezoelectric force proportional to electric field. To evaluate electrostrictive force, we equate stored electromagnetic and acoustic energy in a virtual displacement:

$$P_{EM} \delta \rho = - \frac{E^2}{8\pi} \frac{\partial \epsilon}{\partial \rho} \delta \rho. \quad (19)$$

The acoustic pressure is proportional to change in density by the square of the velocity of sound, written v . Combining the two pressures, we get:

$$p = v^2 \rho_1 = - \frac{E^2}{8\pi} \frac{\partial \epsilon}{\partial \rho} \quad (20)$$

Damping of sound is added phenomenologically. If T is the phonon lifetime, then density satisfies the following equation.

$$\frac{\partial^2 \rho}{\partial t^2} + \frac{1}{T} \frac{\partial \rho}{\partial t} = \nabla^2 \left(v^2 \rho_1 - \frac{\partial \epsilon}{\partial \rho} \frac{E^2}{8\pi} \right) \quad (21)$$

Variation in one space direction is assumed, since in the absence of self-focusing radial changes are small compared to longitudinal growth.

$$\frac{\partial^2 E}{\partial z^2} - \frac{\epsilon}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{c^2} \frac{\partial \epsilon}{\partial \rho} \frac{\partial^2}{\partial t^2} (\rho_1 E) \quad (22)$$

$$\frac{\partial^2 \rho}{\partial t^2} + \frac{1}{T} \frac{\partial \rho}{\partial t} - v^2 \frac{\partial^2 \rho}{\partial z^2} = - \frac{1}{8\pi} \frac{\partial \epsilon}{\partial \rho} \frac{\partial^2}{\partial z^2} (E^2) \quad (23)$$

5. SLOWLY VARYING AMPLITUDE EQUATIONS

Fields are written in the form of slowly varying complex amplitudes that multiply plane wave exponentials:

$$E = E_L e^{i(\omega_L t - k_V z)} + E_S e^{i(\omega_S t + k_S z)} + C. C. \quad (24)$$

$$\rho_1 = \rho_2 e^{i(\omega_V t - k_V z)} + C. C. \quad (25)$$

The exponentials with constant amplitudes satisfy the wave equation without coupling terms. The coupling causes changes which are small in an optical period or over a wavelength distance. To determine the change, the amplitudes are allowed to vary slowly relative to periodic variation of fields in space and time. So that the amplitudes will indeed be slowly varying, the laser and scattered electric fields are written separately. The conservation conditions discussed in the introduction are assumed to relate $\omega_L, \omega_S, \omega_V$, and k_L, k_S, k_V . With this assumption, a set of equations for amplitudes that is devoid of exponentials is derived. Fields in the above form are substituted in the differential equations.

$$\begin{aligned} & -2ik_L \frac{\partial E_L}{\partial z} - k_L^2 E_L - \frac{\epsilon}{c^2} \\ & \times \left(2i\omega_L \frac{\partial E_L}{\partial t} - \omega_L^2 E_L \right) \\ & = - \frac{\omega_L^2}{c^2} \frac{\partial \epsilon}{\partial \rho} \rho_2 E_S \end{aligned} \quad (26)$$

$$2ik_S \frac{\partial E_S}{\partial z} - k_S E_S = -\frac{\epsilon}{c}$$

$$\times \left(2i\omega_S \frac{\partial E_S}{\partial t} - \omega_S^2 E_S \right) \\ = -\frac{\omega_S^2}{c} \frac{\partial \epsilon}{\partial \rho} \rho_2^* E_L \quad (27)$$

$$-\omega_V^2 \rho_2 + 2i\omega_V \frac{\partial \rho_2}{\partial t} + \frac{i\omega_V}{T} \rho_2 \\ - V^2 k_V^2 \rho_2 = + \frac{k_V^2}{8\pi} \frac{\partial \epsilon}{\partial \rho} 2 E_L E_S^* \quad (28)$$

All derivatives of amplitudes are assumed to be negligibly small compared with a frequency. The space derivative of the density is neglected as it is multiplied by the sound velocity $v \ll c$. Only terms which have the same space and time exponentials are equated, so that all exponentials can be cancelled from the slowly varying equations. Using the proportionalities between frequency and wave vectors,

$$\omega_{L,S}^2 = \frac{ck}{\sqrt{\epsilon}} L, S \omega_V = vk_V,$$

one has:

$$\frac{\partial E_L}{\partial z} + \frac{\sqrt{\epsilon}}{c} \frac{\partial E_L}{\partial t} = \frac{\omega_L^2}{2ik_L c^2} \frac{\partial \epsilon}{\partial \rho} \rho_2 E_S \quad (29)$$

$$\frac{\partial E_S}{\partial z} - \frac{\sqrt{\epsilon}}{c} \frac{\partial E_S}{\partial t} = -\frac{\omega_S^2}{2ik_S c^2} \frac{\partial \epsilon}{\partial \rho} \rho_2^* E_L \quad (30)$$

$$\frac{\partial \rho_2}{\partial t} + \frac{\rho_2}{2T} = + \frac{k_V^2}{2i\omega_V} \frac{\partial \epsilon}{\partial \rho} \frac{E_L E_S^*}{4\pi} \quad (31)$$

A new variable S is defined

$$\rho_2 = 2T \frac{k_V^2}{2i\omega_V} \frac{\partial \epsilon}{\partial \rho} \frac{S}{4\pi} \times 10^{13} \quad (32)$$

E_L and E_S squared are proportional to the light intensity measured in MW/cm². The units for E_L and E_S are chosen as $\sqrt{\text{MW/cm}}$. To convert from cgs, one multiplies by 10^{-13} , the number of megawattsec in an erg. The gain $g/2$

$$g/2 = \frac{\omega_L^2}{2c^2 k_L} \frac{\partial \epsilon}{\partial \rho} \frac{2T k_V^2}{2\omega_V} \frac{\partial \epsilon}{\partial \rho} \frac{S}{4\pi} \times 10^{13}.$$

In the following equations, c denotes speed of light in the scatterer. The equations for Brillouin scattering are:

$$\frac{\partial E_L}{\partial z} + \frac{1}{c} \frac{\partial E_L}{\partial t} = -\frac{g}{2} E_S S \quad (33)$$

$$\frac{\partial E_S}{\partial z} - \frac{1}{c} \frac{\partial E_S}{\partial t} = -\frac{g}{2} E_L S^* \quad (34)$$

$$\frac{\partial S}{\partial t} + \frac{S}{2T} = \frac{E_L E_S^*}{2T} \quad (35)$$

6. RAMAN SCATTERING

The molecules in Raman scattering are treated as a two level system. There are many energy levels, but most molecules are found in the ground state, as the temperature of the substance is low. Transitions to the first excited level are the only one considered. Quantum theory is used, since the vibrations are internal to molecules. In quantum theory a wave function is associated with energy levels. These wave functions can be written in bra and ket notation. This notation names functions by a single letter and determines if they are of bra variety or ket

variety. Any function can have either of the two forms. A bra form is written $\langle a|$, and a ket form is written $|a\rangle$. Inner products of a bra and ket are written $\langle a|b\rangle$. A general wave function for this system is a mixture of two states; so, it is written as $|\psi\rangle = a|a\rangle + b|b\rangle$. Here, a and b are complex amplitudes. The states are orthogonal, or the bracket $\langle a|b\rangle$ is zero. They are eigenstates of a Hamiltonian H satisfying

$$H|a\rangle = E_a |a\rangle \quad (36)$$

$$H|b\rangle = E_b |b\rangle \quad (37)$$

Writing Eq. (36) for bras gives $\langle a|H = E_a \langle a|$. The Hamiltonian, H has the property that $\langle a|H|b\rangle = \langle b|H|a\rangle^*$. Taking brackets gives $\langle a|H|b\rangle = E_b \langle a|b\rangle = \langle b|H|a\rangle^* = \langle b|a\rangle^* E_a = \langle a|b\rangle E_b$. Since E_a and E_b are different, $\langle a|b\rangle = 0$. The star signifies complex conjugation. The interpretation is that $|\langle a|\psi\rangle|^2$ is the probability that the system is in state a , and $|\langle b|\psi\rangle|^2$ is the probability that it is in state b . Using the orthogonality of $|a\rangle$ and $|b\rangle$, the probability of state $|a\rangle$ is $|a|^2$ and of state $|b\rangle$ is $|b|^2$. The coefficients a and b depend on time as described by the Schrödinger equation, a basic postulate of quantum theory.

$$H|\psi\rangle = i\hbar \frac{\partial |\psi\rangle}{\partial t} \quad (38)$$

The Hamiltonian is the sum of a large molecular part H_0 and an interaction term. The interaction term is

$$\frac{\partial \epsilon}{\partial Q} \frac{QE^2}{8\pi} \quad (39)$$

The symbol Q is now interpreted in the same way as H in terms of its effect on state functions. The interaction term is an interaction energy owing to change in the dielectric constant. The symbol Q is a vibration coordinate describing the dependence of dielectric constant on molecular vibrations, and E is the electric field. From the Schrödinger equation and orthogonality one gets:

$$i\hbar \dot{a} = E_a a + \frac{\partial \epsilon}{\partial Q} \langle b|Q|a\rangle \frac{E^2}{8\pi} b \quad (40)$$

$$i\hbar \dot{b} = E_b b + \frac{\partial \epsilon}{\partial Q} \langle a|Q|b\rangle \frac{E^2}{8\pi} a \quad (41)$$

Here the dot indicates time derivative. Using $a^* \dot{b} = \dot{a}^* b + a^* \dot{b}$, and taking the conjugate of Eq. (40)

$$i\hbar a^* \dot{b} = (E_b - E_a) a^* \dot{b} + \frac{\partial \epsilon}{\partial Q} \langle a|Q|b\rangle \frac{E^2}{8\pi} (|a|^2 - |b|^2) \quad (42)$$

The expectation of Q is $\langle \psi|Q|\psi\rangle$. The numbers $\langle a|Q|a\rangle$ and $\langle b|Q|b\rangle$ are zero by symmetry, as in an eigenstate the vibration coordinate is as likely to be positive as negative; so $\langle \psi|Q|\psi\rangle = \langle a|Q|b\rangle a^* b + \langle b|Q|a\rangle ab^* = 0$. When a ket is changed to a bra, the complex conjugate of coefficients is taken. With $\langle Q\rangle = \langle \psi|Q|\psi\rangle$, Eq. (42) gives:

$$i\hbar \dot{\langle Q\rangle} = (E_b - E_a) (a^* \dot{b} \langle a|Q|b\rangle - ab^* \langle b|Q|a\rangle) + \frac{\partial \epsilon}{\partial Q} (\langle b|Q|a\rangle)^2 |a|^2 - |b|^2 \quad (43)$$

Also Eq. 42 we get:

$$i\hbar(a^* \dot{b} - \dot{a} |Q| b) - \frac{\partial}{\partial Q} (b |Q| a) = (E_b - E_a) \langle Q \rangle + \frac{\partial \epsilon}{\partial Q} \frac{E^2}{8\pi} (|b|^2 - |a|^2) \langle b |Q| a \rangle^2. \quad (44)$$

The term $(a |Q| b) a^* b - (b |Q| a) ab^*$ and its derivative are eliminated by differentiating Eq. (43) and substituting from Eq. (44). The result is the following:

$$i\hbar \dot{\langle Q \rangle} = (E_b - E_a) \frac{1}{\hbar} (E_b - E_a) \langle Q \rangle + \frac{1}{\hbar} \frac{\partial \epsilon}{\partial Q} \frac{E^2}{8\pi} (|b|^2 - |a|^2) \langle b |Q| a \rangle^2. \quad (45)$$

Letting $E_b - E_a = \hbar \omega_{ab}$, and adding a phenomenological damping time T , one has

$$\langle \dot{Q} \rangle + \frac{\langle \dot{Q} \rangle}{T} + \omega_{ab}^2 \langle Q \rangle = - \frac{\omega_{ab}}{4\pi} \times |a |Q| b|^2 \frac{\partial \epsilon}{\partial Q} E^2 (|b|^2 - |a|^2) \quad (46)$$

A slowly varying approximation is made as before, with $\langle Q \rangle$

$$\langle Q \rangle = Q e^{i(\omega_{ab} t - k_V z)} + C. C.,$$

and k_V is the sum of k_S and k_L . Since the molecules are probably in the ground state, $|b|^2 - |a|^2 \approx 1$.

$$-\omega_{ab}^2 Q + 2i\omega_{ab} \frac{\partial Q}{\partial t} + \frac{i\omega_{ab}}{T} Q + \omega_{ab}^2 Q = -\omega_{ab} \frac{|a |Q| b|^2}{4\pi} \frac{\partial \epsilon}{\partial Q} 2 E_L E_S^* \quad (47)$$

Equation (47) is of the same form as the slowly varying equation corresponding to the sound amplitude S for Brillouin scattering. The light amplitudes are treated classically as before. The final system for Raman scattering is:

$$\frac{\partial E_L}{\partial z} + \frac{1}{c} \frac{\partial E_L}{\partial t} = - \frac{g}{2} E_S Q \quad (48)$$

$$\frac{\partial E_S}{\partial z} - \frac{1}{c} \frac{\partial E_S}{\partial t} = - \frac{g}{2} E_L Q^* \quad (49)$$

$$\frac{\partial Q}{\partial t} + \frac{Q}{2T} = - \frac{E_L E_S^*}{2T} \quad (50)$$

7. BOUNDARY CONDITIONS

The boundary conditions for Eqs. (48), (49), and (50) are values for E_L at the left boundary corresponding to an incident laser pulse, value for E_S at the right boundary corresponding to no incident scattered light, and values for S at both boundaries. The values for S on the boundaries are calculated as for interior points of the medium. The glass boundaries of the scattering cell are assumed to have no effect on the sound. Actually reflection of sound occurs at the boundary, but the duration of the laser pulse, on the order of 50 nsec, is so short that the sound propagates by only tenths of a millimeter. Since light amplification in the distance the reflection could travel is negligible; the reflected wave is neglected. This same approximation is made in the steady-state theory, and good agreement with experimental values is obtained.

V. Noise Source

1. DAMPING AND SPECTRUM

The spectrum of light scattered from a low intensity laser beam in Brillouin scattering is approximately Lorentz.⁷ This corresponds to the simplest phenomenological damping of sound with a phonon lifetime T .

$$\frac{\partial S}{\partial t} + \frac{S}{2T} = \text{driving terms} \quad (51)$$

Assuming that S varies with $e^{i\omega t}$, one gets

$$S = \frac{\text{constant}}{i\omega + \frac{1}{2T}} \quad (52)$$

The spectrum is $|S|^2$, where bars indicate absolute value.

$$|S|^2 = \frac{\text{constant}}{\omega^2 + \left(\frac{1}{2T}\right)^2} \quad (53)$$

This spectrum is called Lorentz. Its width between half power points is $\Delta\omega_{1/2} = 1/T$, where ω denotes angular frequency. The experimental spectra fit a curve of this form.

2. SHOT NOISE SOURCE

To reproduce this spectrum, a noise term is added to the vibration equation. A pulse added to the right side of Eq. (51) gives a Lorentz spectrum. The reason is that a pulse has equal power in all frequencies. A single pulse would not give a steady sound level; therefore, a sum of pulses is added. So that sound will not accumulate with any particular

phase, the pulses are given random phases. The sound equation becomes:

$$\frac{\partial S}{\partial t} + \frac{S}{2T} = \frac{E_L E_S^*}{2T} + A \sum_0^\infty e^{i\phi_m} \times \delta(t - m\Delta t) \quad (54)$$

Here $\{\phi_m\}$ are independent random variables uniformly distributed on $[0, 2\pi]$. The sound pulses are centered at equally spaced intervals: Δt . To evaluate S for ordinary scattering, the small term $E_L E_S^*$ is neglected. Thus,

$$S = A \sum e^{i\phi_m} \frac{e^{-\frac{t-m\Delta t}{2T}}}{2T} u(t - m\Delta t). \quad (55)$$

The autocorrelation of S is:

$$\begin{aligned} \langle S(t) S^*(t + \tau) \rangle &= A^2 \sum \sum e^{-\frac{t-m\Delta t}{2T} - \frac{t+\tau-m\Delta t}{2T}} \\ &\times u(t - m\Delta t) u(t + \tau - m\Delta t) \\ &= A^2 e^{-\frac{|\tau|}{2T}} \sum e^{-\frac{t-m\Delta t}{T}} u(t - m\Delta t) \\ &= A^2 e^{-\frac{|\tau|}{2T}} \frac{1}{1 - e^{-\frac{\Delta t}{T}}} \\ &\approx A^2 \frac{T}{\Delta t} e^{-\frac{|\tau|}{2T}} \quad (56) \end{aligned}$$

where brackets denote ensemble average. This correlation, a bilateral exponential,

corresponds to a Lorentz spectrum, since a Fourier transform gives the Lorentz denominator. The spectrum of the scattered light is the same, since the laser light is nearly monochromatic.

3. QUANTUM RATE EQUATION

To evaluate A, a classical rate equation is derived in the following operation and is compared with a quantum rate equation derived at length in.⁵ Since only the form of the quantum equation is needed here, physical constants such as $\hbar$ and $\frac{\partial \epsilon}{\partial \rho}$ will be omitted. As shown for Raman scattering the interaction part of the Hamiltonian is proportional to $E^2 S$. This can be shown by replacing $\frac{\partial \epsilon Q}{\partial Q}$ by $\frac{\partial \epsilon S}{\partial S}$ in Eq. (39).

A more complete quantum discussion is given here; the light and sound fields are written in terms of quantum amplitudes. Previously the light was not quantized.

$$E_L = b_L e^{i(\omega_L t - k_L z)} + b_L^+ e^{-i(\omega_L t - k_L z)} \quad (57)$$

$$E_S = b_S e^{i(\omega_S t + k_S z)} + b_S^+ e^{-i(\omega_S t + k_S z)} \quad (58)$$

$$S = a e^{i(\omega_V t - k_V z)} + a^+ e^{-i(\omega_V t - k_V z)} \quad (59)$$

The quantum amplitudes have the following properties:

$$b|n\rangle = \sqrt{n} |n-1\rangle \quad (60)$$

$$b^+|n\rangle = \sqrt{n+1} |n+1\rangle \quad (61)$$

The ket $|n\rangle$ is an eigenstate corresponding to n quanta, which can be either photons or phonons for light or sound respectively, in a mode. By time dependent perturbation theory Fermi's rule may be derived from the Schrödinger equation. Fermi's rule states that the rate of change of probability that a final state is occupied is proportional to the square of the matrix element of the Hamiltonian between initial and final states. The probability of the initial state is approximately 1. The time dependent amplitudes vary approximately as $e^{i\omega t}$, where ω is the energy of the state divided by $\hbar$. Without interaction, the Schrödinger equation gives

$$\frac{\partial a}{\partial t} = \frac{iE_a}{\hbar} a.$$

With interaction between initial and final states, the Schrödinger equation for slowly varying amplitudes multiplying $e^{i\omega t}$ gives:

$$\frac{\hbar}{i} \frac{\partial a}{\partial t} = \langle f | H_1 | i \rangle e^{-i\omega_f t}, \quad (62)$$

with

$$|\psi\rangle = e^{i\omega_i t} + a e^{i\omega_f t} |f\rangle.$$

Here, f and i stand for final and initial, respectively. The interaction part of the Hamiltonian, H_1 , has no diagonal matrix elements, so $\langle f | H_1 | f \rangle = \langle i | H_1 | i \rangle = 0$. The reason is that H_1 depends on a single coordinate, and in i or f the coordinate is equally likely to be positive or negative. This equation is integrated to give:

$$a = \frac{\hbar}{i} \langle f | H_1 | i \rangle \frac{e^{i(\omega_i - \omega_f)t} - 1}{i(\omega_i - \omega_f)}. \quad (63)$$

Taking the absolute square gives

$$|a|^2 = \hbar^2 |f| |H_1| |y|^2 \frac{\sin^2 \frac{\omega_{fi} t}{2}}{\left(\frac{\omega_{fi}}{2}\right)^2}$$

Here, $\omega_f - \omega_i = \omega_{fi}$. The sine oscillates rapidly. To obtain an average effect,

$$\frac{\sin^2 \frac{\omega_{fi} t}{2}}{\left(\frac{\omega_{fi}}{2}\right)^2}$$

is integrated over all ω_{fi} . This integral is proportional to t ; that is,

$$\int \frac{\sin^2 \frac{\omega_{fi} t}{2}}{\left(\frac{\omega_{fi}}{2}\right)^2} d\omega_{fi} = t \int \frac{\sin^2 \frac{\omega_{fi} t}{2}}{\left(\frac{\omega_{fi} t}{2}\right)^2} d(\omega_{fi} t).$$

When the sound and light fields are substituted in the interaction Hamiltonian, one has $\hbar \alpha (b_L + b_L^\dagger + b_S^\dagger + b_S) (a + a^\dagger)^2$ corresponding to $E^2 S$. Only energy conserving terms are kept; that is, terms corresponding to creation of scattered photon and phonon and destruction of laser photon, or destruction of scattered photon and phonon and creation of laser photon.

$$\hbar \alpha b_L b_S^\dagger a^\dagger + b_L^\dagger b_S a \quad (64)$$

Using the properties of the creation and destruction operators given above, we get:

$$\begin{aligned} \langle n_L, n_S, n_a | b_L^\dagger b_S a^\dagger | n_L - 1, \\ n_S + 1, n_a + 1 \rangle \\ = \sqrt{n_L (n_S + 1) (n_a + 1)} \quad (65) \end{aligned}$$

$$\begin{aligned} \langle n_L, n_S, n_a | b_L b_S a^\dagger | n_L + 1, \\ \times n_S - 1, n_a - 1 \rangle \\ = \sqrt{(n_L + 1) n_S n_a} \quad (66) \end{aligned}$$

By Fermi's rule,

$$\begin{aligned} \frac{dn_S}{dt} &= \sigma [n_L n_S (n_a + 1) - (n_L + 1) n_S n_a] \\ &= \sigma n_L (n_S + n_a + 1), \quad (67) \end{aligned}$$

where σ is a cross section evaluated explicitly in Ref. 5. The term $n_S n_a$ is negligible compared to $n_L n_a$ because in ordinary scattering the scattered light is much weaker than the laser beam.

4. CLASSICAL RATE EQUATION

A classical rate equation is derived by applying the partial differential equations. A situation in which the space derivative would have no effect for short times is near the center of the medium. Neither sound source nor laser beam depends on position. Before boundary conditions affect the center of the medium, growth of the scattering should be independent of position. The classical and quantum equations are compared for this situation. For weak scattering the laser beam is nearly constant. The equations reduce to:

$$\frac{\partial E_S}{\partial t} = -\frac{c g}{2} E_L S^* \quad (68)$$

$$\begin{aligned} \frac{\partial S}{\partial t} + \frac{S}{2T} &= \frac{E_L E_S^*}{2T} + A \sum e^{i\phi_m} \\ &\times \delta(t - m\Delta t) \quad (69) \end{aligned}$$

Multiplying Eq. (68) by E_S^* and adding the complex conjugate equation gives:

$$\frac{\partial \langle |E_S|^2 \rangle}{\partial t} = \frac{gc}{2} E_L \langle S^* E_S^* + C.C. \rangle \quad (70)$$

Integrating Eq. (68), multiplying by S , and taking ensemble averages, we get:

$$\langle E_S S \rangle = \frac{gc}{2} \int \langle S S^* \rangle dt \quad (71)$$

Using the correlation of S given in Eq. (56), we find:

$$\langle E_S S \rangle = \frac{gc E_L}{2} 2T \frac{A^2 T}{\Delta t}$$

Equation (72) is substituted into Eq. (70) resulting in:

$$\frac{\partial \langle |E_S|^2 \rangle}{\partial t} = gc |E_L|^2 gc \frac{A^2 T}{\Delta t} \quad (73)$$

In deriving Eq. (56), the term $E_L E_S^*$ in Eq. (69) was omitted. This term is assumed to be constant, so that

$$S = E_L E_S^* \quad (74)$$

Substituting this value for S into Eq. (70) gives

$$\frac{\partial \langle |E_S|^2 \rangle}{\partial t} = gc |E_L|^2 |E_S|^2 \quad (75)$$

The total rate of change of $|E_S|^2$ is obtained by adding the right sides of Eqs. (73) and (75);

$$\frac{\partial \langle |E_S|^2 \rangle}{\partial t} = gc |E_L|^2 \left(\langle |E_S|^2 \rangle + gc \frac{A^2 T}{\Delta t} \right) \quad (76)$$

Equation (76) is the classical rate equation.

5. THERMAL NOISE

The slowly varying amplitudes include more than one mode. The quantum rate equations refer to just one mode. The number of modes for the slowly varying amplitudes is estimated by multiplying the one-mode noise source by the free space mode density. For a standing wave, the wave vector times the length must equal some multiple of 2π . The minimum spacing in wave vector between modes is consequently 2π length. In three-dimensional space the minimum spacing between modes is $(2\pi/L)^3$, corresponding to a cube. The number of modes in a piece of a spherical shell of volume $\Delta\Omega k^2 \Delta k$ is $\Delta\Omega k^2 \Delta k \left(\frac{L}{2\pi}\right)^3$. Here $\Delta\Omega$ is solid angle, k is wave vector, L is length, and Δk is difference in length of wave vector. For arbitrarily shaped objects, the following is true:

$$\text{mode density} = \Delta\Omega \frac{k^2 \Delta k}{(2\pi)^3} \quad (77)$$

The bandwidth Δk is assumed to correspond to the bandwidth of the scattered light determined from ordinary scattering spectra. The value of $\Delta\Omega$ is taken as the solid angle of the laser beam $\Delta\Omega_L$. The wave vector bandwidth is $1/T_c$.

Number of modes per unit volume

$$= \Delta\Omega_L \frac{k^2}{(2\pi)^3} \frac{1}{T_c} \quad (78)$$

Each mode is given thermal energy $K T_e$, where T_e is temperature and K is

Boltzmann's constant. This is the energy that an acoustic mode has in thermal equilibrium with a heat reservoir at temperature T_e . The energy is in the form of phonons. Each phonon is converted to a photon in the scattering process. The photon energy corresponding to the phonon source will be larger by the ratio of photon to phonon energies. For backward scattering, $k_V - 2k_L = 2\omega_L/c = \omega_V/V$. Here, ω_L and ω_V are laser and vibration angular frequency, and V and c are sound and light speeds. The ratio of energies equals the ratio of frequencies, or $c/2V$. The scattered photon energy density is

$$E_p = \left(\frac{c}{2V}\right)(KT_e) \left(\Delta\Omega_L \frac{k^2}{(2\pi)^3} \frac{1}{cT}\right) \quad (79)$$

The light intensity is obtained by multiplying the energy density by c .

scattered light intensity

$$= \frac{c}{2V} kT_e \Delta\Omega_L \frac{k^2}{(2\pi)^3} \frac{1}{T} \quad (80)$$

This scattered intensity corresponds to the noise term in Eq. (7). Equating the two quantities gives

$$KT_e \frac{c}{2V} \frac{k_L^2}{(2\pi)^3} \Delta\Omega_L \frac{1}{T} = gc \frac{A^2 T^2}{\Delta t} \quad (81)$$

This determines the value used for A in the noise source. Since the intensity is in MW/cm^2 , the above quantity is multiplied by 10^{-13} , the number of MWsec/erg .

$$gc \frac{A^2 T^2}{\Delta t} = KT_e \frac{ck_L^2}{(2\pi)^3} \Delta\Omega_L \frac{1}{2T} \frac{c}{2V} \times 10^{-13}$$

For Raman scattering, $KT_e = \hbar\omega_R$ and $c/2V = 1$.

The noise constant for ethyl ether is calculated using the following numbers:

$$g = 0.02 \text{ MW/cm}$$

$$T = 0.55 \text{ nsec}$$

$$KT_e = 1.38 \times 10^{-16} \times 300 \text{ ergs}$$

$$\Delta\Omega_L = \pi \times 10^{-6}$$

$$V = 10^5 \text{ cm/sec}$$

$$\lambda_L = 7 \times 10^{-5} / 1.35 \text{ cm}$$

A value of $(0.6 \Delta t)^{\frac{1}{2}}$ is obtained.

The Raman noise constant for CS_2 is calculated using:

$$g = 10^{-2}$$

$$T = 10^{-11}$$

$$\hbar\omega_R = 6.6 \times 10^{-27} \times 656 \times 3 \times 10^{10} \text{ erg}$$

A value of $(4\Delta t)^{\frac{1}{2}}$ is obtained.

VI. Difference Methods

1. INTEGRAL EQUATIONS

The partial differential equations to be approximated by difference equations are:

$$\frac{\partial E_L}{\partial z} + \frac{1}{c} \frac{\partial E_L}{\partial t} = -\frac{g}{2} E_S S \quad (82)$$

$$-\frac{\partial E_S}{\partial z} - \frac{1}{c} \frac{\partial E_S}{\partial t} = -\frac{g}{2} E_L S^* \quad (83)$$

$$\frac{\partial S}{\partial t} + \frac{S}{2T} = \frac{E_L E_S^*}{2T}$$

$$+ A \sum e^{i\phi_m} \delta(t - m\Delta t) \quad (84)$$

The normal to a surface $E_L(z, t)$ is

$$\frac{\partial E_L}{\partial z}, \frac{\partial E_L}{\partial t} - 1.$$

By Eq. (82), curves satisfying the equation

$$dz = c dt = dE_L / (-gE_S S/2) \quad (85)$$

are perpendicular to the normal. These curves lie in the surface $E_L(z, t)$ and are called characteristic curves. The characteristics for Eq. (83) satisfy the following equation:

$$dz = -c dt = dE_S / (-gE_L S^*/2). \quad (86)$$

Integrating Eqs. (85) and (86) gives:

$$\int dE_L = \int -\frac{gE_S S}{2} c dt \quad (87)$$

$$\int dE_S = \int \frac{gE_L S^*}{2} c dt \quad (88)$$

Position is related to time in the integrands by $dz = c dt$ for Eq. (87) and by $dz = -c dt$ for Eq. (88). The integrands are evaluated at points in the z, t plane corresponding to a characteristic curve:

$$E_L(z, t) = \int_0^t \left(-\frac{gE_S S}{2} \right) \times (z - c(t - t'), t') c dt'; \quad (89)$$

$$E_S(z, t) = \int_0^t \left(\frac{gE_L S^*}{2} \right)$$

$$\times (z + c(t - t'), t') c dt'. \quad (90)$$

Equation (84) is integrated in time:

$$S(z, t) = \int_0^t \frac{E_L E_S^*}{2T} e^{-\frac{t-t'}{2T}} dt' + A \sum e^{i\phi_m} \frac{t-m\Delta t}{2T} u(t - m\Delta t) \quad (91)$$

2. DIFFERENCE EQUATIONS

In principle, the integral equations can be solved by successive approximations. In this method a first approximation to E_L, E_S, S is assumed and substituted into the integrals. The values obtained by evaluating the integrals are again substituted into the integrals. This procedure is repeated until successive approximations differ by less than some desired amount. This procedure is not readily adaptable to digital computers, as values of the variables for all position and times would have to be stored. Instead difference equations that approximate the integral equations are used to advance the solution from one time to the next. A computational mesh where $\Delta z = c\Delta t$ is used. The subscript j and superscript n denote position $j\Delta z$ and time $n\Delta t$ in the mesh. Difference equations are obtained by applying the integral equations between mesh points.

The integrals are evaluated by expanding the integrands l in Taylor series.

$$\int_{n\Delta t}^{(n+1)\Delta t} l dt' = l^n \Delta t + \left(\frac{dl}{dt} \right)^n \frac{(\Delta t)^2}{2} + \frac{d^2 l}{dt^2} \frac{(\Delta t)^2}{6} \quad (92)$$

The first derivative is approximated by a difference quotient.

$$\int l dt' = l^n \Delta t + \left(\frac{l^{n+1} - l^n}{\Delta t} - \frac{d^2 l}{dt^2} \frac{\Delta t}{2} \right) \frac{\Delta t^2}{2} + \frac{d^2 l}{dt^2} \frac{(\Delta t)^2}{6} \quad (93)$$

The difference quotient is obtained from Taylor series. Derivatives without time superscript are evaluated at an unspecified time between $n\Delta t$ and $(n+1)\Delta t$.

$$l^{n+1} = l^n + \left(\frac{dl}{dt} \right)^n \Delta t + \frac{d^2 l}{dt^2} \frac{(\Delta t)^2}{2} \quad (94)$$

The terms in Eq. (93) that multiply $(\Delta t)^3$ are written $O(\Delta^3)$

$$\int l dt' = \frac{l^n + l^{n+1}}{2} \Delta t + O(\Delta^3) \quad (95)$$

Equation (95) is applied to the integrals in Eqs. (89), (90), and (91). The terms of order Δ^3 are omitted. Equation (89) is integrated between mesh points 2 and 1, where $\delta z = c\delta t$. Equation (90) is integrated between points 3 and 1, where $\delta z = -c\delta t$. Equation (91) is integrated from point 4 to 1, where $\delta z = 0$.

$$E_{Lj}^{n+1} - E_{Lj-1}^n = -\frac{g\Delta z}{4}$$

$$+ \left[(E_S S)_j^{n+1} + (E_S S)_{j-1}^n \right] \quad (96)$$

$$E_{Sj}^{n+1} - E_{Sj+1}^n = \frac{g\Delta z}{4} + \left[(E_L S^*)_j^{n+1} + (E_L S^*)_{j+1}^n \right] \quad (97)$$

$$S_j^{n+1} = S_j^n e^{-\frac{\Delta t}{2T}} + (E_L E_S^*)_{Sj}^{n+1} \frac{\Delta t}{4T} + (E_L E_S^*)_{Sj}^n \frac{\Delta t}{4T} e^{-\frac{\Delta t}{2T}} + A e^{i\phi_m} \quad (98)$$

Equation (95) indicates difference in z or t between mesh points. The difference $c\Delta t$ is replaced by Δz in Eqs. (96) and (97). Equation (91) is equivalent to

$$S(z, t) = \int_{t-\Delta t}^t \frac{E_L E_S^*}{2T} e^{-\frac{t-t'}{2T}} S(z, t - \Delta t) + A e^{i\phi_m} \quad (99)$$

This equation is approximated above in Eq. (98).

3. SOLVING THE DIFFERENCE EQUATIONS

Equations (96), (97), and (98) do not give explicitly values for E_L^{n+1} , E_S^{n+1} , and S^{n+1} in terms of E_L^n , E_S^n , and S^n . Algebraic solution of the equations is difficult. Instead they are solved by using a simpler equation for S^{n+1} to obtain approximate values of S . Values of E_L and E_S are computed using the approximate value of S^{n+1} . The more accurate Eq. (98) is used later to improve the initial estimate. The rough value for S^{n+1} is obtained by omitting

$O(\Delta^2)$ rather than $O(\Delta^3)$ terms. In place of Eq. (95), we use

$$\text{Idt}' = \Gamma^n \Delta t + \frac{\partial \Gamma}{\partial t} \frac{\Delta t^2}{2}, \quad (100)$$

which in Eq. (91) gives

$$S_j^{n+1} = S_j^n e^{\frac{\Delta t}{2T}} + (E_L E_S^*)_j^n \frac{\Delta t}{2T} + A e^{i\phi_m}. \quad (101)$$

The reason for calculating approximate values for S is that, in Eqs. (96) and (97), S is multiplied by Δ . Terms of $O(\Delta^2)$ in S contribute only in $O(\Delta^3)$ to E_L and E_S . The values of E_{Lj}^{n+1} and E_{Sj}^{n+1} are obtained using Cramer's rule and the following equations:

$$E_{Lj}^{n+1} + \frac{g\Delta z}{4} (E_S S)_j^{n+1} = E_{Lj-1}^n - \frac{g\Delta z}{4} (E_S S)_{j-1}^n \quad (102)$$

and

$$-\frac{g\Delta z}{4} (E_L S^*)_j^{n+1} + E_{Sj}^{n+1} = E_{Sj-1}^n + \frac{g\Delta z}{4} (E_L S^*)_{j-1}^n \quad (103)$$

The resulting values are:

$$E_{Lj}^{n+1} = \frac{E_{Lj-1}^n - \frac{g\Delta z}{4} (E_S S)_{j-1}^n - \frac{g\Delta z}{4} S_j^{n+1} \left[E_{Sj+1}^n + \frac{g\Delta z}{4} (E_L S^*)_{j+1}^n \right]}{1 + \left| \frac{g\Delta z}{4} S_j^{n+1} \right|^2} \quad (104)$$

$$E_{Sj}^{n+1} = \frac{E_{Sj-1}^n + \frac{g\Delta z}{4} (E_L S^*)_{j-1}^n + \frac{g\Delta z}{4} S_j^{n+1} \left[E_{Lj-1}^n - \frac{g\Delta z}{4} (E_S S)_{j-1}^n \right]}{1 + \left| \frac{g\Delta z}{4} S_j^{n+1} \right|^2} \quad (105)$$

A final value for S_j^{n+1} is obtained using the values of E_{Lj}^{n+1} and E_{Sj}^{n+1} and the more accurate Eq. (98).

At the left boundary E_L is specified. A rough value of S is computed using Eq. (101), E_S is computed as above, and S is calculated again. At the right boundary E_S is specified, and E_L and S are calculated.

4. COMPUTATIONAL METHODS

To save running time in the computations, the real and imaginary parts of the equations were written separately. The corresponding real and imaginary quantities were stored separately. The routines for handling complex numbers proved to be slower than this direct approach. So that only one set of fields E_L , E_S , and S need be stored; the right hand sides in Eqs. (102) and (103) are computed and stored for all j .

$$X_j = \text{Re} \left[E_{Lj-1}^n - (E_S S)_{j-1}^n \right]$$

$$Y_j = \text{Im} \left[E_{Lj-1}^n - (E_S S)_{j-1}^n \right]$$

$$P_j = \text{Re} \left[E_{Sj+1}^n + (E_L S^*)_{j+1}^n \right]$$

$$Q_j = \text{Im} \left[E_{Sj+1}^n + (E_L S^*)_{j+1}^n \right]$$

A new variable equaling the previous S multiplied by $g\Delta z/4$ is used above. Real and imaginary quantities are denoted Re and Im respectively. Next, part of the precise value for S_j^{n+1} is computed.

$$\text{Re}S_j^{n+1} = \text{Re} \left[S_j^n e^{-\frac{\Delta t}{2T}} + \frac{\Delta t}{4T} (E_L E_S^*)^n_j \right] \quad (106)$$

$$\text{Im}S_j^{n+1} = \text{Im} \left[S_j^n e^{-\frac{\Delta t}{2T}} + \frac{\Delta t}{4T} (E_L E_S^*)^n_j \right]$$

The approximate value for S is calculated and used to obtain values for E_L and E_S . The new values for E_L and E_S are used to complete the calculation of S_j^{n+1} in Eq. (106) using Eq. (98). The calculations following Eq. (106) are done for each value of j , then the cycle beginning with Eq. (106) is repeated.

VII. Brillouin Scattering

In this section and the following one the results or applications of the theory that has been developed thus far are presented. The difference equations are applied to various specific situations that approximate conditions under which experiments have been performed. For Brillouin scattering the specific experiments that are modeled were performed by Maier in 1966.¹⁵ In these experiments the intensity distribution in time of backward scattered light is measured. Difference methods are used to calculate the intensity of the scattered light. The results of the numerical calculations are similar to the experimental results and approximate theory. Two scattered pulses are shown in Figs. 3 and 4.

1. ERROR

There is inherent error in a finite difference calculation owing to the replacement of derivatives by difference ratios. Generally, the error is larger

the coarser the net. To examine the error, calculations were done with meshes of different sizes. The other parameters in the calculation (length of scattering cell, noise constant, incident laser pulse, and material in the scattering cell) were the same. To eliminate random differences, the source of noise was a constant scattered intensity on the boundary of the cell farthest from the laser. Use of the more realistic distributed source would cause additional statistical differences owing to different sets of random numbers. A constant scattered intensity of $9 \times 10^{-11} \text{ MW/cm}^2$ was used. This number is equal to gcA^2T^2/t , which is the intensity of light due to scattering from thermal sound in the medium. Meshes of 100, 200, 300, and 400 points were used. The length of scattering cell was 50 cm, so that mesh intervals varied between 0.5 cm and 0.125 cm. The scatterer was ethyl ether. A pulse with intensity normally distributed in time and with width

between half intensity points of 15 nsec was used as laser pulse. The peak intensity was 800 MW/cm^2 . This is the peak intensity of pulses used by Maier in his experiments.¹⁵ The calculated scattered intensity at different times after incidence of the laser pulse is displayed in Table 1. The laser pulse is assumed to be incident 25 nsec ahead of peak intensity. The effect of the different mesh widths is apparent.

No intensity could be calculated for 25 nsec with the 100-point mesh. There was an instability that disrupted the computation, causing numbers too large for the computer to store to appear. With a mesh this coarse, the changes in intensities between mesh points are so large that a constant rate of change between mesh points is a bad approximation. The difference equations are valid for small linear changes in the variables between mesh points. At 9 ns the error apparently reduces in proportion to the time step. The error is halved from an estimated 0.136 in going from 100 to 200 points, and is again halved in going from 200 to 400 points. The error for the 100-point mesh is estimated by doubling the difference between the 100- and 200-point calculations. Using this estimate for the error corresponding to the

100-point calculation, the error for the 400-point mesh is estimated at 2% for 9 ns. At 16 ns the error is less than 0.05% beyond the 200-point mesh, which is negligible for these computations. At 25 ns the error appears to be of the order of 1%, since differences between successive calculations are of this size. The scattering here is approximately steady state, as the laser pulse is flattest, i.e., changing least rapidly in time, at 25 nsec after the pulse is incident.

2. DISTRIBUTED NOISE

A more realistic noise source is a distributed noise source. Here thermal density fluctuations are assumed to occur throughout the cell rather than just at the boundary. A random pulse is added to the density at each mesh point at each time step. The above calculations were performed for this noise source and a 200-point mesh. The length of the scattering cell was taken as 50 cm.; ethyl ether in the scattering cell was assumed; and a laser intensity normally distributed in time with a 15-ns half-width was used. All of these choices were made to model Maier's experimental results.¹⁵ The peak laser intensity was 800 MW/cm^2 . In the experiments the intensity of the laser beam varies over the beam cross section. The laser intensity is distributed normally in radius. In this calculation and in the transient theory described later, the laser intensity is assumed constant with radius. The scattered pulse is shown in Fig. 4. A calculation with varying laser intensity is described later in "steady state

Table 1. Calculated scattered intensity for different times and mesh sizes.

Time (nsec)	Mesh size (points)			400 MW/cm ²
	100	200	300	
9	2.174	2.106	2.084	2.073×10^{-7}
16	2.650	2.637	2.637	2.536×10^{-2}
25		7.493	7.560	7.640×10^{-2}

computation" and in "transient computation". The peak scattered percentage was 94% for 800 MW/cm² laser intensity. Transient scattering occurs before the pulse maximum, when the sound wave is small compared to its maximum. The scattering reaches 1% of the laser intensity for the first time at 13 ns from the time of incidence of the laser pulse. This is called transient scattering.

These results, the peak scattering percentage and the time for the transient scattering to reach 1% of the laser intensity, were the same for the distributed noise as for the previous boundary noise used in the discussion of the error. The calculations were insensitive to the kind and size of noise source. A distributed noise source attenuated by a factor of 30 was tried. Transient scattering occurred 1.5 ns later.

3. MAIER'S THEORY

The noise source used by Maier¹⁵ is different from the one used in this computational treatment in two respects. He uses a boundary noise source of constant amplitude, as opposed to the distributed noise source with randomly phased amplitudes used here. He also includes fewer modes in the scattered intensity because of thermal fluctuations. Here all modes within the bandwidth of low intensity or ordinary scattered light are given thermal energy KT and are assumed to scatter light. Maier uses a more restricted bandwidth in the noise source. The bandwidth is assumed limited by spectral narrowing. Owing to high gain, the spectrum of the scattered light is narrower than the spectrum of

low intensity scattered light in unstimulated or ordinary scattering.

The theory of spectral narrowing is developed in the following paragraph. Assume the sound to vary sinusoidally at frequency ω . The sound equation is

$$\frac{\partial S}{\partial t} + \frac{S}{2T} = \frac{E_L E_S^*}{2T} \quad (107)$$

With S and E_S amplitudes multiplying the exponential $e^{-i\omega t}$ and constant laser amplitude, Eq. (107) becomes:

$$S = \frac{E_L E_S^*}{-i\omega + \frac{1}{2T}} \quad (108)$$

The equation for the scattered light amplitude E_S is

$$\frac{\partial E_S}{\partial z} - \frac{1}{c} \frac{\partial E_S}{\partial t} = -\frac{g E_L S^*}{2} \quad (109)$$

For high-gain steady scattering, the time derivative can be neglected. For monochromatic scattered light, Eq. (109) together with Eq. (108) gives:

$$\frac{\partial E_S}{\partial z} = \frac{g}{2} |E_L|^2 \frac{E_S}{-i\omega + \frac{1}{2T}} \quad (110)$$

A solution to Eq. (109) is:

$$E_S = \exp \left(\frac{g |E_L|^2 l}{4T(1/2T - i\omega)} \right) \quad (111)$$

where l is length of the scattering cell. Other solutions are proportional to this one. The spectrum of the scattered intensity is obtained by taking the real part of the exponential corresponding to

multiplying E_S by E_S^* , its complex conjugate. The intensity spectrum is

$$\exp \left\{ \frac{g|E_L|^2 \ell}{8T^2 [\omega^2 + (1/2T)^2]} \right\}.$$

For large gain, $g|E_L|^2 \ell / 8T^2$, the frequency half width between half intensity points is much narrower than $1/T$, which is the frequency width for ordinary scattered light. The factor $g|E_L|^2 \ell / 8T^2$ can be as large as 200×10^{18} . When ω is 0, the exponent is 200; when ω is $1/2T$, the exponent is 100. The value of e^{100} is much less than half that of e^{200} . The half width is obtained by solving

$$\exp \left\{ \frac{g|E_L|^2 \ell}{8T^2 [\omega^2 + (1/2T)^2]} \right\} = \frac{1}{2} e^{\frac{g|E_L|^2 \ell}{2}} \quad (112)$$

the solution is

$$\frac{g|E_L|^2 \ell}{8T^2 [\omega^2 + (1/2T)^2]} = \frac{g|E_L|^2 \ell}{2} - \ln 2 \quad (113)$$

$$\omega^2 = \frac{g|E_L|^2 \ell}{8T^2 \left(\frac{g|E_L|^2 \ell}{2} - \ln 2 \right)} - \left(\frac{1}{2T} \right)^2 \quad (114)$$

$$\omega^2 \approx \left(\frac{1}{2T} \right)^2 \frac{\ln 2}{\frac{g|E_L|^2 \ell}{2}} \quad (115)$$

Equation (115) is obtained by expanding the denominator in Eq. (114). Maier¹⁵ attenuates his noise source because of the restricted bandwidth indicated by Eq. (115). The factor by which the bandwidth is restricted in Eq. (115) is

25. In the calculations presented here, the bandwidth is not attenuated as the calculation is started from ordinary scattering. The noise has its largest effect at the beginning of the computation, where the gain is small. To determine the effect of using an attenuated noise source, a noise constant attenuated by a factor of 30 was tried. The effect was small. Transient scattering reached 1% of the laser intensity 1.5 ns later than with the larger noise source. The scattered intensity was insensitive to this change in the noise except for initial scattering, which is unobserved in the experiments. Near the pulse maximum, Maier's steady-state theory applies. Here the laser intensity, which is normally distributed in time, is changing least rapidly in time. The rate of growth or decay of the sound wave is determined almost completely by small changes in the beam intensity, as transient scattering has already occurred. Maier's steady-state theory is described in the following paragraph.

Equations for the light intensities are obtained by letting $S = E_L E_S^*$ in Eqs. (33) and (34). This substitution corresponds to neglecting the time derivative in Eq. (106). Here $I_L = E_L E_L^*$ and $I_S = E_S E_S^*$, the laser and scattered light intensities. Time derivatives in Eqs. (33) and (34) are neglected as well. Multiplying (34) by E_S^* gives:

$$E_S^* \frac{\partial E_S}{\partial z} = - \frac{g|E_L|^2 |E_S|^2}{2} \quad (116)$$

Adding the complex conjugate equation, we get:

$$\frac{\partial I_S}{\partial z} = - g I_L I_S \quad (117)$$

Similarly, one obtains from Eq. (34)

$$\frac{\partial I_L}{\partial z} = -g I_L I_S \quad (118)$$

Letting $\partial I = I_L(0) - I_S(0)$, the difference between laser and scattered intensity at the end of the scattering cell nearest the laser, Eqs. (116) and (118) give:

$$I_L(z) - I_S(z) = \Delta I \quad (119)$$

Using Eq. (119) in Eq. (118), we find:

$$\frac{\partial I_S}{\partial z} = -g I_S (I_S + \Delta I) \quad (120)$$

A solution of Eq. (120) is:

$$I_S = \frac{\Delta I}{2} \left(\coth \frac{g \Delta I z}{2} - 1 \right) - \frac{\Delta I}{e^{g \Delta I z} - 1} \quad (121)$$

Equation (121) can be verified by substitution into (120). For the case of high gain, $g \Delta I \ell \gg 1$, Eq. (121) gives:

$$I_S(\ell) = \Delta I e^{-g \Delta I \ell} \quad (122)$$

Equation (122) is obtained by neglecting the 1 in the denominator of Eq. (121), which is valid for high gain.

Using Eq. (121), a ΔI of 27 MW/cm² is computed corresponding to $I_S(\ell) = 8 \times 10^{-11}$ MW/cm². This $I_S(\ell) = g c A^2 T^2 / \Delta t$, which is the thermally scattered light intensity corresponding to one photon scattered by each phonon in the modes in the laser bandwidth. Each mode is given energy $K T$. For laser intensity of 800 MW/cm², the peak scattering percentage corresponding to 27 MW/cm² is 97%. As stated in the

section "Distributed Noise Source," difference method calculations give a value of 94%. One reason for the discrepancy is that the scattering is not precisely steady state. Another is the lack of precision of the difference method computation. This high scattering percentage is not expected in experiments with laser intensity varying with radius. The attenuated noise source of Maier was tried; peak scattering of 96% was calculated. Again the scattering is insensitive to the size of noise source, as was found previously for transient scattering.

3. STEADY-STATE COMPUTATION

The calculations described to this point; the boundary noise scattering and error, the distributed noise calculation of transient and steady scattering, and different sizes of noise source; have all been based on a model with one space dimension—length. The actual laser intensities are normally distributed in radius. Consequently a two-dimensional model, with length and radius, is more realistic.

To simulate a cylindrically symmetric beam with intensity normally distributed in radius, scattered intensity is calculated for different laser intensities corresponding to different radii. The intensities are then used to calculate the energy by multiplying 2π times the radius times the difference between successive radii, and then adding. In Fig. 1, the scattered light power, i.e., intensity times cross section, is shown for a laser beam with intensity normally distributed in time, with half-width of 15 nsec. The

Intensity is normally distributed in radius as well, with peak intensity of 800 MW/cm^2 and peak power of 1 MW.

Fig. 3. Brillouin scattering by a cylindrically symmetric beam; average scattering calculated from scattering at various radii showing gradual approach to steady scattering.

Fig. 4. Brillouin scattering at beam center; the approach to steady scattering at beam center is abrupt.

The scatterer is 50 cm of ethyl ether. The scattering is computed at 12 radii. The constant boundary noise and a 200-point mesh are used. We emphasize the qualitative distinction between Figs. 3 and 4. Figure 2 corresponds to scattering at beam center, and shows an abrupt approach to steady state scattering. The transient is nearly vertical. In Fig. 3, the scattered power increases more gradually. The transient is no longer readily apparent. The reason is that at the larger radii the transient occurs later, and the cumulative transient occurs over a longer period of time.

The peak percentage of light power scattered that we computed is close to Maier's measured and theoretical values.¹⁵ He applies the steady-state theory described above to a nonuniform laser pulse. The average scattering is much less than at beam center. Maier's experimental and theoretical values are 80 and 83%, respectively. Our computed value is 87%.

4. TRANSIENT COMPUTATION

Transient scattering occurs prior to the time when steady-state theory applies. For transient scattering the sound wave limits the scattering to a small value. When the sound wave becomes large enough to scatter most of the laser light, steady-state theory becomes applicable. Subsequent changes in the sound intensity are caused by changes in the laser intensity and are slow. During transient scattering the laser intensity is constant. The laser beam is not affected by the scattered light, as the scattered light is

weak relative to the laser beam. Maier and Renner give a solution based on constant laser intensity, that is, laser intensity unaffected by the scattering.¹² A laser intensity corresponding to scattered light of 1% of the laser beam is predicted. Maier and Renner use a constant (nonfluctuating) noise source. The scattering is computed here at beam center in accord with Maier and Renner's treatment. The threshold for 1% scattering is compared with values from a theoretical curve¹² in Table 2.

Table 2. Comparison of computed and theoretical transient threshold intensities.

Phonon lifetime (nsec)	Theory, (MW/cm ²)	Computed (MW/cm ²)
0.26	90	106
0.55	107	127

The phonon lifetime 0.55 nsec corresponds to ethyl ether, while 0.26 nsec does not correspond to a specific substance. The scattering cell was 50 cm long. A laser pulse, the intensity of which was normally distributed in time with a 15-nsec half width, was used. Other

physical constants, index of refraction and sound speed, were the same for the two computations. The agreement between theoretical and computed intensities is satisfactory in view of the approximate nature of the Maier-Renner theory.

The previous computation was done at beam center to facilitate comparison with theory. A more realistic computation of average scattering (i.e., scattering corresponding to a laser pulse with intensity varying with radius) was also done. The result was that the average intensity corresponding to 1% of the laser power scattered is 145 MW/cm² for ethyl ether. The difference from the value of 127 MW/cm² for beam center scattering is notable. The average intensity was calculated as scattered power divided by cross section.

To determine the effect of statistical fluctuations and distributed noise, the distributed random noise source was used. Scattering was calculated at beam center. The distributed noise source scatters 1% of the laser intensity at laser intensity 143 MW/cm² $\pm$ 3. The sample average and standard deviation are computed from 10 runs.

VIII. Raman Scattering

The experiments of Maier, Kaiser, and Giordmaine¹⁵ on backward Raman scattering are treated here. Forward Raman scattering has also been observed and treated theoretically.¹⁰ In contrast to Brillouin scattering, the wave vector conservation condition in Raman scattering is not important. The Raman

vibration has frequency to a large extent independent of wave vector. Consequently slight changes in frequency can occur to bring about matching of backward or forward scattered wave vectors and the incident laser wave vector. Forward stimulated Brillouin scattering is not observed.

As for Brillouin scattering, the down shifted or Stokes frequency is the one treated. The differential and difference equations are of the same form as for Brillouin scattering.

Maier, Kaiser, and Goirdmaine's work concerns short pulses arising in a cell containing CS_2 . An incident laser pulse undergoes self-focusing at the end of the scattering cell farthest from the laser. The laser light becomes concentrated in filaments of micron diameter at the end of the cell. Raman scattered light arises in these filaments, then propagates toward the laser as a pulse. When the pulse becomes as intense as the laser beam, it depletes the laser beam in the cell, leaving the cell momentarily without illumination. Maier, Kaiser, and Goirdmaine obtained pulse intensities from 20 to 30 times the laser intensity and pulse widths of 30 psec.

1. MAIER, KAISER, and GIORDMAINE'S TREATMENT

In their discussion, Maier, Kaiser, and Giordmaine do not treat the self-focusing in detail. They solve rate equations obtained by setting $S = E_L E_S^*$ in Eqs. (33) and (34), multiplying by complex conjugate amplitudes, and adding complex conjugate equations. This procedure is similar to that used to derive Eqs. (116) and (117). The time derivatives in the equations for the light intensities are kept. With $I_L = E_L E_L^*$ and $I_S = E_S E_S^*$, they obtain:

$$\begin{aligned} \frac{\partial I_L}{\partial z} + \frac{1}{c} \frac{\partial I_L}{\partial t} &= -g I_L I_S \\ &= \frac{\partial I_S}{\partial z} - \frac{1}{c} \frac{\partial I_S}{\partial t} \end{aligned} \quad (123)$$

These equations are solved subject to short pulse boundary conditions.

Here, we adopt a model for self-focusing consisting of intensifying the laser intensity near the end of the cell. The pulses from a random source as in Brillouin scattering are computed.

2. MODEL FOR SELF-FOCUSING

In the attempt to obtain pulses of the same 30-psec width reported by Maier, Kaiser, and Giordmaine, the laser beam was intensified by various factors and at various distances from the end of the cell. The intensification was accomplished by multiplying the laser intensity by a constant factor at each mesh point in a length of 1 cm. Table 3 shows the overall intensification and position tried before arriving at one that gives the appropriate peak intensity. A constant 70 MW/cm^2 laser intensity is used as in Maier's discussion. The justification is that the laser intensity changes little in the time required by a pulse to arise and propagate through the cell. As in the work of Maier, Kaiser, and

Table 3. Self-focusing parameter study.

Intensification factor	Self-focus position (cm)	Peak (MW/cm^2)	Width (psec)
100	25-26	5500	
100	28-29	4700	
100	29-30	0	
100	$28\frac{1}{2}$ - $29\frac{1}{2}$	2200	93
50	28-29	1480	120
		2200	114
40	28-29	2800	85
30	28-29	2350	106
100	$28\frac{1}{2}$ - $29\frac{1}{2}$	1650	107

Fig. 5. Random Raman pulse 1.

Fig. 6. Random Raman pulse 2.

Fig. 7. Random Raman pulse 3; half width (duration between half maximum intensities) is 125 psec.

Giordmaine, a one-dimensional geometry is used.

The computations were done for a 30-cm cell of CS_2 using a 300-point mesh. The width reported by Maier was 30 psec. It was hoped that some combination of intensification factor and position would give a pulse width of this order and peak intensity in the range 1400 to 2100 MW/cm^2 corresponding to 20 to 30 times the laser intensity. All pulse widths calculated were longer than 30 psec. The

pulse widths and peak intensities were subject to statistical fluctuations owing to the random nature of the noise source. Accordingly, for the self-focus position of 28-29 cm from the front end of the cell and intensification factor of 30, average widths and peak intensities were calculated. The sample averages and standard deviations for 10 runs were 106 ± 22 ps and $2350 \pm 450 \text{ MW}/\text{cm}^2$ peak intensity. The intensity fluctuation here is about 40%, corresponding to the

Fig. 8. Random Raman pulse 4 (from the sample used to calculate average peak intensities and pulse duration).

Fig. 9. Random Raman pulse 5; duration of 150 psec is the longest in the sample.

observed peak intensity fluctuations of Maier, Kaiser, and Giordmaine. Some of the pulses used in the calculation of the averages are shown in Figs. 5-12A. Different sets of random numbers are used in each figure. Because of self-focusing, the solid angle used in determining the number of modes for the noise source is uncertain. Accordingly a larger noise constant is used in the last pulse calculated, Fig. 13. The pulse

width was not dramatically different from the other cases. Figure 13 shows a typical pulse width near the self-focusing end of the cell. Its width of 100 psec is not dramatically changed in propagating through and amplifying in the rest of the cell. The calculations using difference methods and a distributed noise source, with phenomenological self-focusing, give longer pulse widths than observed by Maier, Kaiser, and Giordmaine.

Fig. 10. Random Raman pulse 6.

Fig. 11. Random Raman pulse 7; distribution of scattered intensity in time showing peak pulse intensity and width.

Fig. 12. Random Raman pulse 8; noise constant increased by a factor of 100; change in pulse width is not remarkable.

Fig. 13. Raman pulse leaving self-focused region: typical pulse leaving a filament of laser light.

Fig. 14. Raman amplified normal pulse; a 47-psec pulse assumed to arise from self-focusing is amplified.

3. AMPLIFIED BOUNDARY PULSE

Better agreement with Maier and Kaiser's pulse width is obtained by amplifying a normal pulse, that is, a pulse with intensity normally distributed in time. The peak intensity is assumed to be 0.007 MW/cm^2 (0.01% of the laser

intensity), and width between half maximum intensity points is assumed to be 47 psec. The peak pulse intensity and width correspond to values used by Maier, Kaiser, and Giordmaine in solving their rate equations. No self-focusing is assumed for this calculation, and a 300-point mesh was used. The

Fig. 15. Autocorrelation of normal pulse corresponding to experimental determination of pulse duration.

emitted pulse is shown in Fig. 14. Its width is 41 psec, in better agreement with the 30-psec width reported for the experiments.

An autocorrelation of the intensity for this pulse is computed by multiplying the

Raman intensity by a delayed intensity, $I(t)I(t + \tau)$, and integrating in t . The normalized correlation gives a measure of the pulse width. Figure 15 shows the normalized correlation. The width is 36 psec.

Acknowledgments

The author wishes to acknowledge the assistance of Dr. Joseph Fleck in the origination of this project and in the final stages of its completion. The help generously given by Dr. John Killeen in preparing the final draft is also gratefully

acknowledged. Thanks are given to Mr. Robert Anderson and Mr. Don Reeves for running some computer programs. The author is indebted to the staff of Lawrence Livermore Laboratory for preparing the final manuscript.

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