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Diffractive Scattering of Picosecond Light Pulses in Absorbing Liquids[*]

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A mode-locked neodymium-glass laser with a fast shutter was used to study light scattering from an absorbing liquid. Two equal high-intensity beams from the laser intersected at a small angle in the liquid, and the resulting stimulated diffractive scattering was observed. Scattering was found on two time scales: a fast effect, developing within the 10-psec duration of a mode-locked laser pulse, and a slow effect requiring many nanoseconds to develop. An analysis is presented which shows the slow effect to be a combination of stimulated thermal Rayleigh scattering and stimulated thermal Brillouin scattering. The fast effect is explained as a result of saturation of the absorbing liquid in an interference pattern.

I. INTRODUCTION

Molecular scattering of light, in the form of Rayleigh, Rayleigh-wing, and Brillouin scattering, is now known in both spontaneous and stimulated versions. The stimulated scattering was first demonstrated for Brillouin scattering by Chiao, Townes, and Stoicheff.[1] Characteristic of their experiment, and almost all of those that followed, is the use of two light beams passing through the same region of the material being studied. One beam is a high-intensity laser beam, and the second, at some angle to the first, is of very low intensity.

In the sample, then, is an interference pattern of electromagnetic fields caused by the two beams. The fields are coupled to some property of the sample (density, polarizability, etc.). Hence, the condition of the sample is altered—in stimulated Brillouin scattering (SBS), for example, a sound wave is induced. The changed properties of the sample, acting back on the light through the dielec-

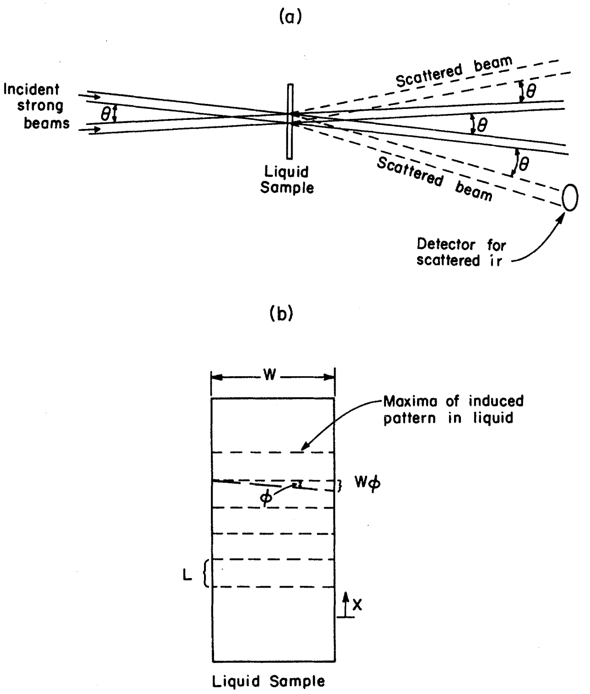


FIG. 1. (a) Geometry of light beams and sample; (b) detail of sample, showing symbols used in the text.

tric susceptibility, then scatter the light.

If the two original beams are properly arranged in direction and frequency, the induced scattering will be such as to divert energy from the strong beam into the same direction, phase, and frequency as the weak beam. Hence, the weak beam will grow, so the interference between the two beams will increase, increasing the effect on the sample. This feedback system will result in exponential growth of the weak beam.

The requirement for the proper geometry and frequency of the two beams is equivalent to a momentum-conservation condition. This requirement has the advantage of allowing the experimental separation of effects with different conditions, but has disadvantages in making the effects more difficult to find and to analyze.

In the experiments reported here,[2,3] a different geometry is used. As shown in Fig. 1, two equal, coherent, strong laser beams are incident on the sample, which is a thin volume of liquid. The interference pattern of the fields of these beams can strongly influence the liquid with no requirement for feedback or momentum conservation. The altered liquid can then scatter light (either the strong beams already present, or an additional weak beam used as a probe) in the manner of a diffraction grating.

The analysis to be given here will consider only the effect of the light on the simplest thermodynamic properties of the liquid (S, T, ρ, P) and the dependence of the index of refraction on these properties. Effects such as nonlinear or anisotropic polarizability will be ignored.

Conventionally, the state variables ρ and T are chosen to study the behavior of the index of refraction, so

$$n = n_0 + \left(\frac{\partial n}{\partial \rho}\right)_T \delta\rho + \left(\frac{\partial n}{\partial T}\right)_\rho \delta T \, . \qquad (1)$$

The behavior of the liquid is more easily studied in terms of the variables P and S. Thus, there are two separable cases: If only variations in pressure are present and S is constant (adiabatic scattering), then

$$n - n_0 = \delta n = \left[\left(\frac{\partial n}{\partial \rho}\right)_T \left(\frac{\partial \rho}{\partial P}\right)_S + \left(\frac{\partial n}{\partial T}\right)_P \left(\frac{\partial T}{\partial P}\right)_S\right] \delta P \, ; \qquad (2)$$

or, if only entropy fluctuations are present and P is constant (isobaric scattering), then

$$\delta n = \left[\left(\frac{\partial n}{\partial \rho}\right)_T \left(\frac{\partial \rho}{\partial S}\right)_P + \left(\frac{\partial n}{\partial T}\right)_\rho \left(\frac{\partial T}{\partial S}\right)_P\right] \delta S \, . \qquad (3)$$

If neither the pressure nor the entropy is constant, then Eqs. (2) and (3) must be added together.

The fluctuations δP and δS are most easily studied in terms of their spatial Fourier components. Equation (2) then represents scattering of light by ordinary adiabatic pressure waves—sound waves. Such waves propagate through the liquid, and the scattered light is Doppler shifted. Light scattered in this manner forms the "Brillouin doublet," symmetrically located in frequency about the frequency of the incident light.

The fluctuations represented by Eq. (3) are nonpropagating, as they have no pressure component. They are local density fluctuations, associated with temperature changes resulting in no change in pressure. They move and change only with the relatively slow time scale of thermal conductivity in the liquid. Hence, light scattered from these fluctuations is essentially unshifted in frequency and forms the central Rayleigh component of the scattering.

In many of the stimulated scattering experiments that have been reported, transparent samples are used, and the principal influence of the light on the sample is electrostriction. For the short laser pulses that are usually used, this is an adiabatic pressure effect, and it can be analyzed with Eq. (2). The Brillouin scattering is enhanced, but there is no effect on the Rayleigh scattering.

If the sample material shows optical absorption at the frequency of the laser light, then the main influence of the light will be to heat the sample. This addition of energy is described by Eq. (3),

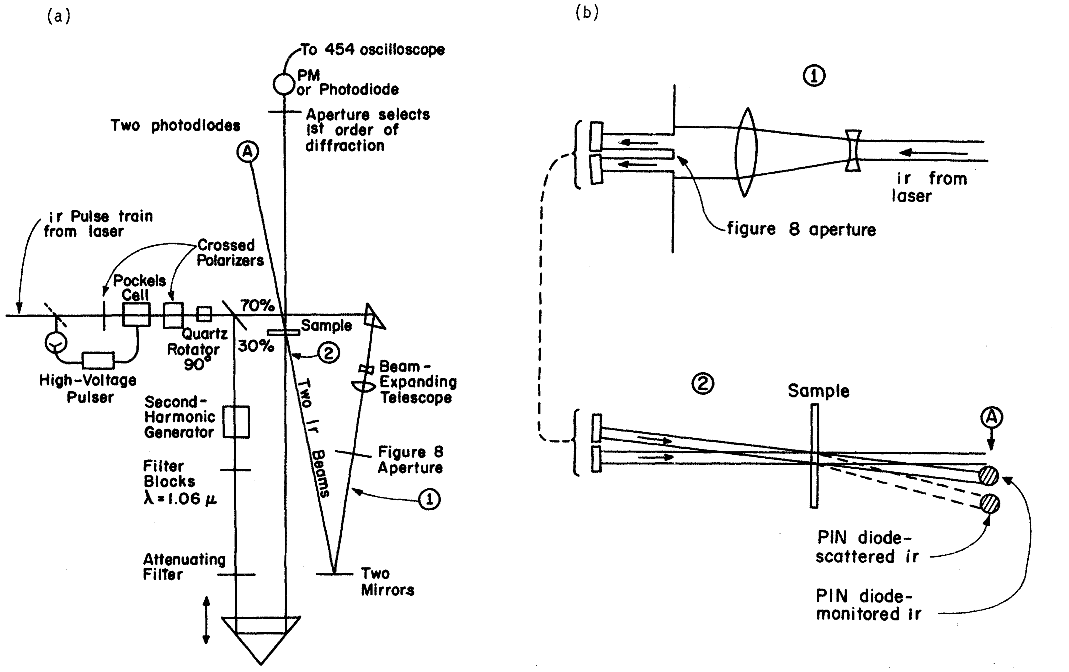


FIG. 2. Apparatus of the probe experiment. (a) Complete view; (b) side views of the system which provided two converging beams.

and the result will be an increase of the Rayleigh scattering. The effect is known as stimulated thermal Rayleigh scattering (STRS).[4,5] The heating of the sample is not isobaric, so terms from Eq. (2) will also be involved, and stimulated thermal Brillouin scattering (STBS) may also be found.

In Eqs. (2) and (3), the first term is much larger than the second. Hence, experimental study of the pure temperature dependence of the index of refraction is difficult.

The availability of the very short (10^{-11} sec or less) pulses from mode-locked lasers suggests an experimental approach to studying the temperature term. Mode-locked laser pulses may be used for a stimulated scattering experiment. In the duration of one of these pulses, the density of a liquid cannot change appreciably. Hence, a study of the temperature effect is possible by simply doing the experiment before the density has a chance to change.

An experiment of this kind was reported by Mack.[6,7] However, he used a long (many nanoseconds) train of pulses from a mode-locked laser. In these pulse trains, the amplitude of the early pulses is small, and scattering is not seen until the pulses are large. Hence, a slow density effect accumulating throughout the early part of the pulse train cannot be ruled out.

To study fast scattering more carefully, scattering experiments were performed with the two-strong-beam geometry described above.[2,3] A mode-locked Nd-glass laser was used for the strong beams. To eliminate ambiguity about the time scale, the laser beam was passed through a fast shutter before being used for the experiment. The shutter was opened during the middle of the pulse train from the laser. Hence, the first pulse reaching the sample was large, and it was possible to observe whether or not scattering occurred with a single short pulse.

II. EXPERIMENTS AND RESULTS

The apparatus used for most of the experiments is shown in Fig. 2. The Nd-glass laser is Q switched and mode locked with EK9860 dye. The output is a train of pulses at $\lambda = 1.06\ \mu$, about 10 psec long, spaced 4 nsec apart. The entire pulse train lasts about 150 nsec. The beam diameter is about 3 mm, and the power density in the short pulses is about 4.5 GW/cm^2.

The fast shutter consists of a Pockel's cell and two polarizers, with associated electronics. The shutter has a rise time of about 3 nsec, an extinction ratio better than 500 : 1, and a transmission when open of about 80%.

The second-harmonic generator was a potassium dihydrogen phosphate (KDP) crystal with a conversion efficiency of about $\frac{1}{2}$%. The second-harmonic intensity was roughly proportional to the intensity of the fundamental in the region of operation. Intensity of the second harmonic was further

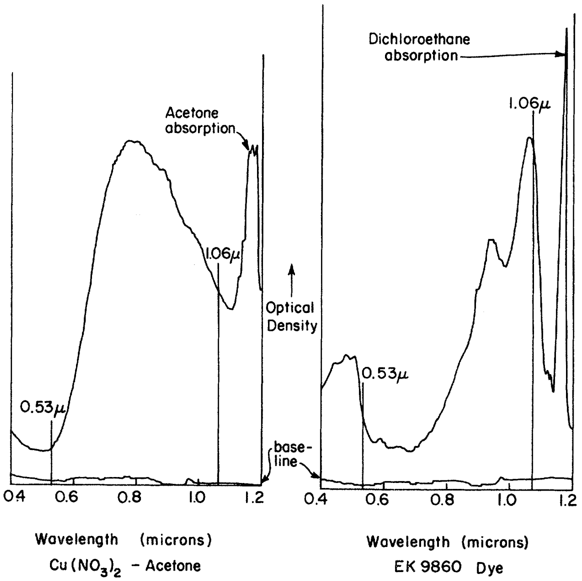


FIG. 3. Absorption spectra of $Cu(NO_3)_2$ in acetone and EK9860 dye in dichloroethane.

attenuated by filters, so the intensity at the sample was less than 500 kW/cm^2.

The aperture and mirror arrangement shown in Fig. 2(b) produced two converging beams at the sample. The angle between the beams was 5 mrad, and the intensity in each beam was roughly 0.5 GW/cm^2.

By changing the optical path length, the pulses in the second-harmonic beam could be delayed or advanced relative to the intense ir pulse pairs at the sample. The location of the zero-delay position was determined accurately with the use of an optical Kerr shutter[8] at the sample position.

Samples were held in a glass windowed cell 0.07 cm thick. Two different liquids were studied: EK9860, an organic dye in dichloroethane solvent, and $Cu(NO_3)_2$ dissolved in acetone. The absorption spectra of both samples are shown in Fig. 3. For both liquids, the transmission of the sample cell for low-intensity light at $\lambda = 1.06\ \mu$ was 10%.

The ir light at 1.06 μ was detected with PIN photodiodes. They had a response time of less than 0.3 nsec and were linear up to output currents of at least 0.5 A. To detect the (green) second-harmonic pulses at 0.53μ, a type-931A photomultiplier was used. To obtain sufficient time resolution with this tube, the output was taken from dynode No. 7, and the voltage from the photocathode to this dynode was set to 2200 V. Connected this way, the tube was able to completely resolve the separate pulses (4 nsec apart). Outputs from the detectors were observed with Tektronix Nos. 519 and 454 oscilloscopes.

III. EXPERIMENTS WITH ir ALONE

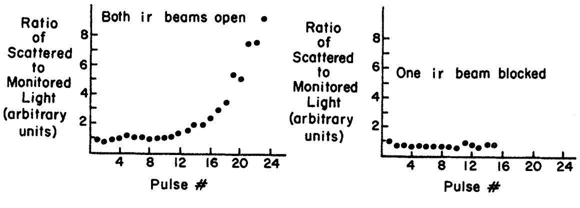
The scattered ir intensity in the first-order diffraction (see Fig. 1) from the sample was measured and was normalized by dividing by the intensity of one of the transmitted beams. Figure 4 shows this scattering ratio for typical pulse trains for the $Cu(NO_3)_2$-acetone and EK9860 samples.

For the $Cu(NO_3)_2$-acetone sample, the noise level was established by blocking one of the strong ir beams to the sample. For the pulse trains shown for the EK9860 sample, the apparatus of Fig. 5 was used. This is basically the same experiment as the one shown in Fig. 2, but without the second-harmonic beam. In addition, the pulses in the two strong beams may be advanced or delayed relative to each other. As shown in Fig. 4(b), scattering is observed when the delay between the two beams is zero; this is compared with the noise level found when the delay between the two beams is 10 psec.

As shown in Fig. 4, the scattering ratio for both liquids rises after about ten pulses have passed through the sample, ultimately growing to about 5–10 times the noise level by the end of the pulse train.

For the $Cu(NO_3)_2$-acetone sample, the scattering ratio for the first few pulses was the same as the noise level to within the experimental error of about 50%. For the EK9860 dye, however, immediate scattering was observed with a scattering ratio for the first few pulses of 2–4 times the noise level. [This initial scattering from EK9860 was also observed with the apparatus of Fig. 2 to provide unambiguous comparison with the $Cu(NO_3)_2$

(a) $Cu(NO_3)_2$ - acetone



(b) EK 9860 - dichloroethane

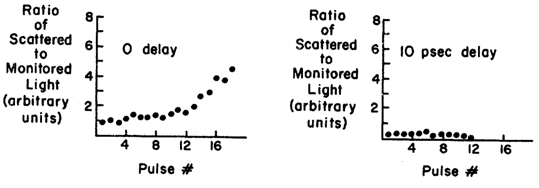


FIG. 4. Ratio of scattered to monitored ir light. (a) $Cu(NO_3)_2$-acetone sample, apparatus of probe experiment but with the green beam blocked; (b) EK9860 sample, apparatus of the delay experiment.

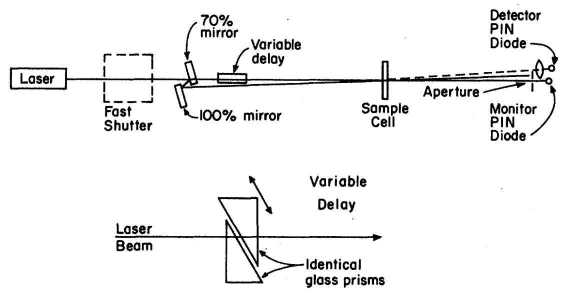


FIG. 5. Apparatus of delay experiment.

acetone. Results were the same as those shown in Fig. 4(b).]

The apparatus of Fig. 5 was also used without the fast shutter to observe the scattering of ir light in EK9860 as a function of the delay between the strong ir beams. Results are shown in Fig. 6. Each point is the average of several pulse trains, looking only at the scattering during the strongest part of each pulse train.

IV. EXPERIMENTS WITH SECOND HARMONIC

The low-intensity train of green pulses was used as a probe to study the grating produced by the intense ir pulse pairs. The apparatus of Fig. 2 was used for both samples.

Typical scattering ratios are shown in Fig. 7 for both liquids. The noise level was established by blocking one of the strong ir beams. For both liquids, no immediate scattering above noise level was found. The scattering started to grow larger after about ten pulses had passed through the sample, eventually becoming 5–10 times the noise level.

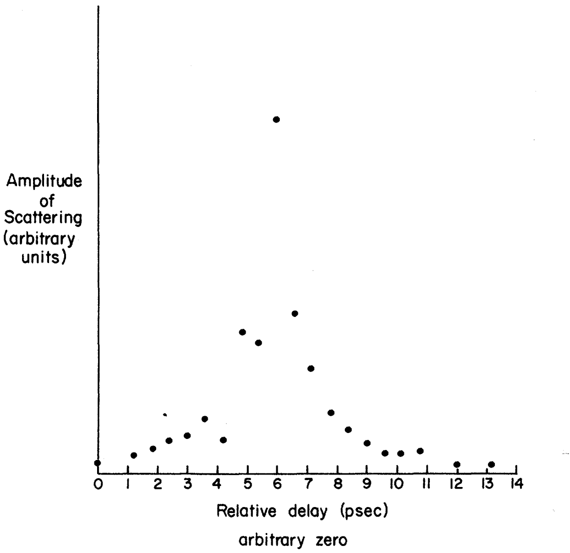
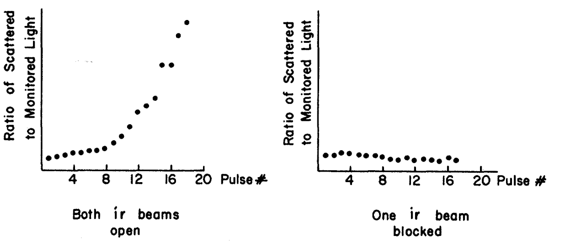


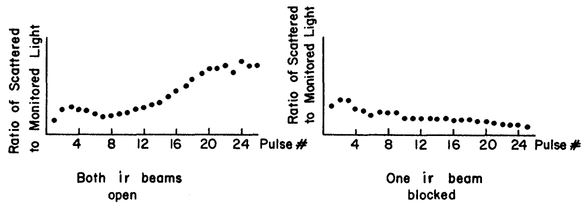
FIG. 6. ir scattering as a function of the delay between the two strong beams. Average of several pulse trains per point, with no fast shutter.



Both ir beams open

One ir beam blocked

EK 9860 sample



Both ir beams open

One ir beam blocked

$Cu(NO_3)_2$ – Acetone sample

FIG. 7. Ratio of scattered green light to monitored ir light. Apparatus of probe experiment.

With the fast shutter removed and using the EK9860 sample, a sheet of Polaroid film (no lens system) was exposed to a single-pulse train from the laser at the location of the photomultiplier. Three orders of diffraction may be seen, as shown in Fig. 8. When the fast shutter was in place for the same experiment, only the first order could be seen on the film.

Additional Observations

Scattering was studied without the fast shutter for the EK9860 dye. For both the fundamental and the second harmonic, the scattering was large (10–20 times background noise) for all pulses in the pulse train. However, it should be noted that the pulse train develops gradually, and some significant fraction (~ 10%) of the total energy in the pulse train has already passed through the sample by the time the pulses are large enough to be clear-

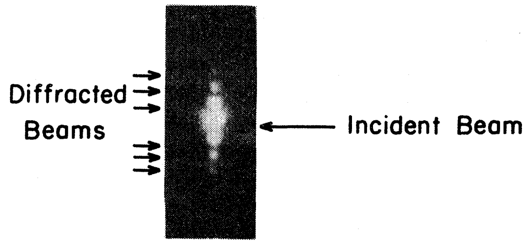


FIG. 8. Polaroid film exposed to scattered green light. Apparatus of probe experiment without the shutter.

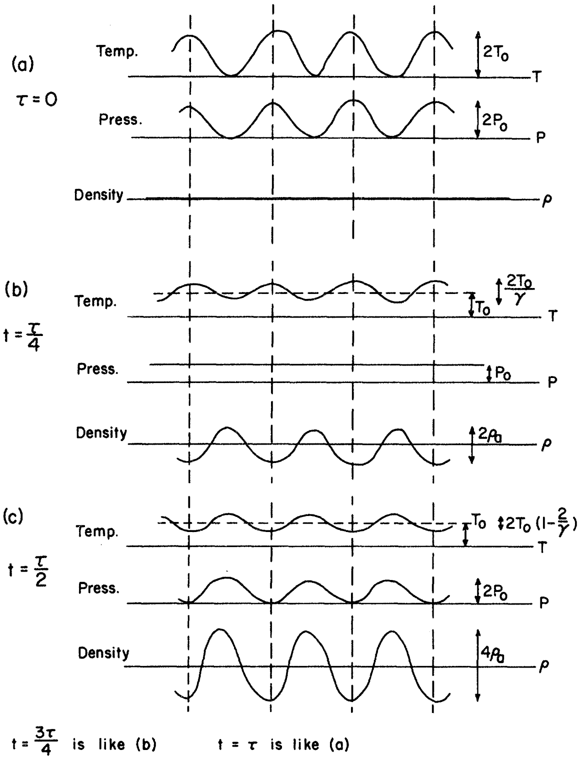


FIG. 9. Behavior of liquid heated by one ir pulse pair at time $\tau = 0$.

ly discernable.

Attempts were made to observe scattering above noise level with the liquids (dichloroethane and acetone) without any absorbers. These attempts were unsuccessful.

V. ANALYSIS OF RESULTS

The general problem of stimulated scattering in absorbing liquids has been reviewed in several recent papers,[9–11] for experiments with one strong beam and one weak beam. The following is a brief consideration of scattering induced in an absorbing liquid by two strong beams. Slow effects related to density changes will be discussed first, and the fast effect observed with EK9860 dye will be interpreted afterwards.

Calculation of Behavior of Liquid

The arrangement of the light and the sample is shown in Fig. 1. The strong input beams consist of trains of short coherent pulses with wavelength λ. The angle between the beams is θ, and hence the intensity of the light in the sample varies as

$$I(x) = 2I_0[1 + \cos(2\pi x/L)] ,$$

where I_0 is the intensity in each beam, $L = \lambda/\sin\theta$ is the wavelength of the pattern induced in the liquid, and x is transverse to the average direction of the beams, as shown. It is assumed that the pulses in the two beams are arriving simultaneously.

If the liquid absorbs a fraction $f = 1 - e^{-\alpha W}$, where α is the attenuation coefficient and W is the sample thickness, of a pulse of duration t_p, then the average heat absorbed per unit volume in the sample is

$$Q(x) = I(x)t_p f/W = Q_0[1 + \cos(2\pi x/L)] , \quad Q_0 = 2I_0 t_p f/W . \tag{4}$$

This energy absorbed from the light will appear as a change of temperature of the liquid. The resulting temperature distribution is

$$T(x) = T + T_0[1 + \cos(2\pi x/L)] , \tag{5}$$

$$T_0 = Q_0\left(\frac{\partial T}{\partial Q}\right)_V = \frac{Q_0}{C_V} , \tag{6}$$

where T is the temperature before the radiation reaches the sample and C_V is the specific heat per unit volume at constant volume.

The pressure distribution caused by this heating has the same form,

$$P(x) = P + P_0[1 + \cos(2\pi x/L)] \tag{7}$$

with

$$P_0 = T_0\left(\frac{\partial P}{\partial T}\right)_V = T_0\frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P \Big/ \frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_T = T_0\frac{\beta}{K_T} , \tag{8}$$

where β is the isobaric thermal expansion coefficient and K_T is the isothermal compressibility.

A liquid that has a pressure-temperature distribution of this form will react as shown schematically in Fig. 9. The easiest way to understand this behavior is to consider the induced distribution of $T(X)$ and $P(X)$ as the sum of two waves: an isobaric temperature-density wave as shown in Fig. 10(a) and an adiabatic pressure (sound) wave as shown in Fig. 10(b).

The pressure wave, with amplitude P_0 from Eq. (8), will have associated with it a temperature

$$T_a = P_0\left(\frac{\partial T}{\partial P}\right)_S . \tag{9}$$

By applying one of the thermodynamic Maxwell relations, this can be expressed as

$$T_a = P_0\left(\frac{\partial V}{\partial S}\right)_P = P_0\left(V\frac{\partial T}{\partial S}\right)_P\left(\frac{1}{V}\frac{\partial V}{\partial T}\right)_P = P_0\frac{T\beta}{C_P} , \tag{10}$$

where C_P is the specific heat per unit volume at constant pressure.

Combining (8) and (10) gives

$$T_a = T_0\frac{T\beta^2}{C_P K_T} = T_0\frac{\gamma - 1}{\gamma} , \tag{11}$$

where γ is the usual ratio C_P/C_V and use has been

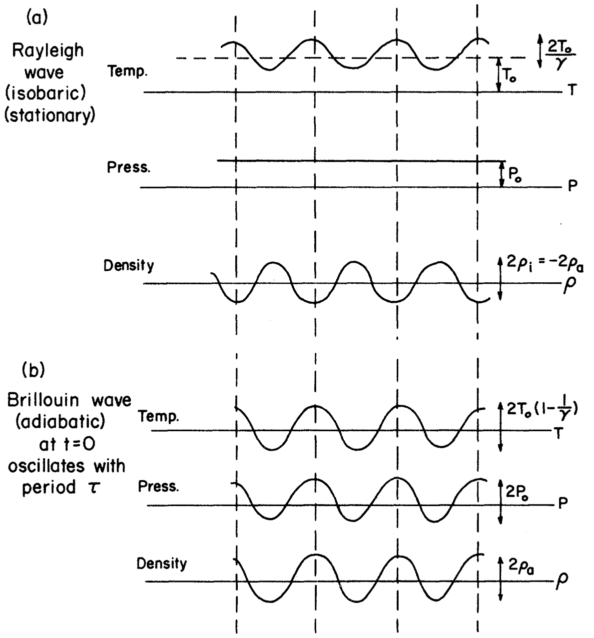


FIG. 10. Two component waves equivalent to those in Fig. 9.

made of the relation $\gamma - 1 = T\beta^2/C_V K_T$.

The remaining fraction of the temperature wave is associated with the isobaric wave and has an amplitude

$$T_i = T_0 - T_a = T_0/\gamma \ . \tag{12}$$

The density waves associated with these two components may also be determined. For the adiabatic wave,

$$\rho_a = P_0\left(\frac{\partial\rho}{\partial P}\right)_S = P_0\left(-\frac{\partial V}{\partial P}\right)_S \frac{1}{V^2} \ ,$$
$$\frac{\rho_a}{\rho} = P_0\left(-\frac{1}{V}\frac{\partial V}{\partial P}\right)_S = P_0 K_S = P_0\frac{K_T}{\gamma} = T_0\frac{\beta}{\gamma} \ , \tag{13}$$

and for the isobaric wave

$$\frac{\rho_i}{\rho} = T_i\left(-\frac{1}{V}\frac{\partial V}{\partial T}\right)_P = -T_i\beta = -T_0\frac{\beta}{\gamma} \ . \tag{14}$$

Therefore the two density waves have equal and opposite amplitudes, which is not surprising since uniform density for their sum was an initial condition of the problem.

These two waves behave quite differently. The isobaric "wave" just stands where it is and shows only slow (milliseconds) thermal diffusive decay. The pressure wave is an acoustic standing wave, oscillating with period $\tau = L/c_v$, where c_v is the speed of sound in the liquid. The preceding analysis depends on the inequality $t_p \ll \tau$, which is easily satisfied in these experiments.

Table I shows numerical values of the various quantities for the liquids in these experiments, as excited by a single ir pulse pair.

In the actual experiments, the liquid is acted upon by a train of pulses. The model of Figs. 9 and 10 allows easy visualization of the action of successive pulses, with results essentially the same as those obtained by Pohl.[10]

Each successive pulse pair imparts a new contribution to the already present waves. For the isobaric wave, the new contributions may simply be added up, as evolution of this wave during the course of the entire pulse train is negligible. However, evolution of the adiabatic wave must be considered, and the addition must take proper account of the phases of the new and exciting waves.

Let $\rho_{i,l}$ and $\rho_{a,l}$ be the amplitudes of the isobaric and adiabatic density wave produced by the lth pulse pair ($\rho_{i,l} = -\rho_{a,l}$), arriving at the liquid at time t_l. Then at time t, the total isobaric wave will be

$$\rho_R = \sum_{t_p < t} \rho_{i,l} \ , \tag{15}$$

the adiabatic wave will have an amplitude

$$\rho_B = \left| \sum_{t_p < t} \rho_{a,l}\, e^{i2\pi t_l/\tau} \right| \ , \tag{16}$$

and the total density wave will be

$$\rho_T = \rho_R + \rho_B \cos(2\pi t/\tau) \ . \tag{17}$$

Each pulse pair, considered by itself, creates equal density waves of the two types. If the pulse pairs are spaced any integer times τ apart, the summed adiabatic and isobaric waves remain equal in amplitude. In all other cases, ρ_R becomes larger than ρ_B.

As Pohl has pointed out,[10] the case when the two waves are equal is of special importance. In this situation, the total density wave becomes null once in each cycle of the adiabatic wave, as indicated by Eq. (17). When the liquid is in this

TABLE I. Thermodynamic parameters for the experiment.

	Acetone	Dichloroethane	Units
C_V	1.30	1.10	J/°K cm^3
β	1.390×10^{-3}	1.135×10^{-3}	°K^{-1}
κ_T	1.27×10^{-10}	0.797×10^{-10}	cm/dyn
ρ	0.7911	1.2521	g/cm^3
γ	1.34	1.43	...
c_V	1.2×10^5	1.1×10^5(calc)	cm/sec
I_0	0.4×10^9 W/cm^2		
L	0.02 cm		
τ	170	180	nsec
T_0	0.08	0.05	°K
P_0	8.8×10^5	7.5×10^5	dyn/cm^2
ρ_a/ρ	7.5×10^{-5}	4.0×10^{-5}	...

constant-density configuration, only a temperature wave is present, and one may try to observe pure temperature scattering; that is, scattering represented by the second terms of Eqs. (2) and (3). This configuration of the liquid may be achieved by careful synchronization or, probably more easily, by using only one exciting pulse pair.

Actually, even for properly timed pulses, two effects tend to introduce disparity between the amplitudes of the two waves. First, the damping times of the two waves are different; the isobaric wave decaying through simple thermal diffusion and the adiabatic wave through dynamic acoustic damping. Second, the wave pattern is only created in a small area, and the standing acoustic wave actually consists of two equal wave pulses traveling in opposite directions that happen to overlap. After they are created, they will travel away from the region where they were produced.

Calculations of the Effect on Light

With the configuration of the liquid known at any time, the scattering of the light may be determined. Several influences are important: The density variations cause variations in the refractive index and in the absorption coefficient, and the temperature variations also cause changes in the refractive index. Needed, then, is the scattering of light from a volume with a sinusoidally varying index of refraction and attenuation.

The analysis is simplified by the thinness of the sample and the small angles of incidence and scattering. If one is concerned only with radiation within an angle ϕ of the normal on a sample of thickness W in which the wavelength of the pattern is L [see Fig. 1(b)], and $\phi W \ll L$, then the sample may be considered simply to be a diffraction grating.[12,13] In the experiments reported here $L = 0.02$ cm, $\phi \leq 0.005$ rad, and $W = 0.07$ cm, so $\phi W \leq 0.05L$ and a diffraction grating calculation is adequate.

The optical properties of the liquid are the index of refraction

$$n(x) = n_0 + N\cos(2\pi x/L) \tag{18}$$

and the absorption coefficient

$$\alpha(x) = \alpha_0 + A\cos(2\pi x/L)\ , \tag{19}$$

where n_0 and α_0 are the values before the waves are induced. The amplitudes of these variations are related to quantities considered earlier by

$$A/\alpha_0 = \rho_T/\rho \tag{20}$$

and $N = \delta n$ from Eqs. (1) and (2) or (3), respectively.

Far from the sample, the amplitude of the light field is found by integrating the amplitude contribution across the sample in the usual manner. The result is that at an angle ω from the direction of the incident beam, the light intensity is

$$I(\omega) = I_0 e^{-\alpha_0 W}\left|\sum_{m=0}^{\infty}(iU)^{|n|+2m}\frac{1}{(|n|+m)!\,m!}\times\left(1-\frac{iUV}{|n|+m+1}+i\frac{V}{U}(|n|+m)\right)\right|^2 \tag{21}$$

$$= I_0 e^{-\alpha_0 W}\left|J_{|n|}(2U)+iV[J_{|n|-1}(2U)-J_{|n|+1}(2U)]\right|^2, \tag{22}$$

where

$$U = \pi W N/\lambda \quad \text{and} \quad V = \tfrac{1}{4}WA\ , \tag{23}$$

for an angle ω which satisfies the expected relation $L\sin\omega = n\lambda$, and $I(\omega) = 0$ for other angles. (The inclusion of the variable transmission is a modification of an otherwise well-known calculation.[13,14]) This calculation assumes that $e^{-V} \approx 1 - V$.

For the slow effect, V may be found using Eq. (20). To find N and U, it is necessary to estimate values of the derivatives in Eq. (1).

The quantity most accessible to measurement, and hence found in the literature, is $(dn/dT)_P$. This is related to the quantities of interest here by

$$\left(\frac{dn}{dT}\right)_P = \left(\frac{\partial n}{\partial\rho}\right)_T\left(\frac{\partial\rho}{\partial T}\right)_P + \left(\frac{\partial n}{\partial T}\right)_\rho\ . \tag{24}$$

To get some idea of the relative contribution of the two terms, it is useful to refer to the Lorentz–Lorenz relation. This indicates that, *if* the molecular polarizability remains constant,

$$\frac{\partial n}{\partial\rho} = \frac{(n^2+2)(n^2-1)}{6\rho n}\ . \tag{25}$$

Using this expression, and the measured value of dn/dT for acetone, and putting the numbers into Eq. (24) gives

$$-5.3\times10^{-4} = (0.504)(-1.10\times10^{-3}) + \left(\frac{\partial n}{\partial T}\right)_\rho\ ,$$
$$-5.3\times10^{-4} = -5.5\times10^{-4} + \left(\frac{\partial n}{\partial T}\right)_\rho\ . \tag{26}$$

Mack[6] in fact used this as a means of calculating $(\partial n/\partial T)_\rho$, but this does not seem valid in view of the restrictions on Eq. (25). In fact, calculating $(\partial n/\partial T)_\rho$ this way is equivalent to assuming that the molecular polarizability depends much more strongly on T than it does on P or ρ; an assumption that is not at all obvious. What is really needed is measured values of one of the partial derivatives, and they are lacking.

However, the numbers in (26) do indicate that the direct effect of T on n is quite small compared with the indirect effect through the density, as is well known. So when density changes are present, as they are in the slow effect, it will suffice to consider only $\partial n/\partial\rho$ and ignore $(\partial n/\partial T)_\rho$. Hence, we have

TABLE II. Optical parameters for the experiment.

	Acetone	Dichloroethane
n_D	1.359	1.444
$\frac{\partial n}{\partial \rho}_{\text{Lorentz-Lorenz}}$	0.504	0.410
$\frac{\partial n}{\partial \rho} = -\frac{1}{\beta}\frac{dn}{dT}_{\text{measured}}$	0.480	...
$N_{\tau/2} = 2\frac{\partial n}{\partial \rho}\rho_{a\,1\ \text{pulse pair}}$	6×10^{-5}	4.2×10^{-5}
$U = \frac{1}{2} WkN_{\tau/2}$	0.12	0.09

$$N = \frac{\partial n}{\partial \rho}\, \rho_T \; . \tag{27}$$

Numerical values for these experiments are shown in Table II.

Using (20) and (27) for the present experiments, after about ten pulses have passed through the sample, ρ_T will be about 3×10^{-4} g/cm^3 for acetone, so

$$U = 0.3 \text{ and } V = 3 \times 10^{-5}.$$

Clearly in this case, terms containing V in expressions (21) or (22) are unimportant. Considering the first term ($m = 0$) in the first sum of expression (21), for $n = 1$, a rough estimate is that it will represent the entire sum to within 4%, and for higher values of n (higher orders) the approximation will be even better. So for U small compared with 1, the phase grating produces a diffraction pattern, in which the intensity in the nth order is proportional to $(U^{|n|}/|n|!)^2$.

If U is not small compared with 1, the intensity of the nth order is proportional to $[J_{|n|}(2U)]^2$.

The experimental results for the slow scattering are in accord with this analysis. The two density waves ρ_R and ρ_B grow with additional pulses passing through the sample. Their sum ρ_T, which is responsible for the scattering ($U \propto \rho_T$), has a free-oscillation period of 170 nsec. Thus, in the fast-shutter experiments (Figs. 4 and 7), the 40-nsec time required for scattering to grow large is quite reasonable.

In experiments without the fast shutter, ρ_T is growing slowly with new input pulses by the time the pulses in the train are large. And the oscillations of ρ_T are fairly small, as ρ_R is larger than ρ_B; therefore the scattering seems fairly constant.

From the preceding analysis, the two density waves ρ_R and ρ_B may be shown to be responsible for Rayleigh and Brillouin scattering, respectively. For example, consider the scattering of a weak, continuous probe beam from a liquid in which ρ_R and ρ_B are equal. Then the scattering in the first order, proportional to $(\rho_T)^2$, will oscillate as $\cos^2(\pi f t)$, where $f = 1/\tau$ is the frequency of the acoustic wave in the liquid. Thus the first-order scattered light may be considered to be composed of three frequency components: one unshifted, or Rayleigh component, arising from the stationary wave ρ_R; and components shifted by $\pm f$, each half the intensity of the unshifted component, representing Brillouin scattering from the wave ρ_B. In the higher orders, appropriately higher multiples of the frequency shift will be found.

Fast Scattering

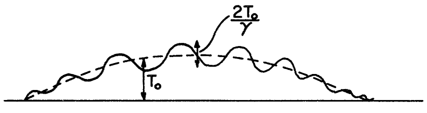
Observations of scattering from EK9860 with the fast shutter [Fig. 4(b)] show significant scattering of the first ir pulse pair to pass through the sample. Density fluctuations of the kind described above cannot develop in the 10-psec duration of the pulses; therefore, another mechanism must cause the scattering.

The absorption peak at 1.06μ of the EK9860 dye is known to be saturable,[15] and this saturation seems the most likely cause of the scattering.[16] The saturation of absorption of this dye is well known, and has made this dye popular for Q-switching and mode-locking neodymium lasers. An intensity of 56 MW/cm^2 reduces the absorption of a thin or dilute sample by a factor of 2, and the dye recovers in 6–9 psec. Presumably, the intense ir pulses will bleach a diffraction pattern in the dye, which will then scatter the latter part of the pulses. The green light will be unaffected, as the dye is practically transparent to that wavelength anyway.

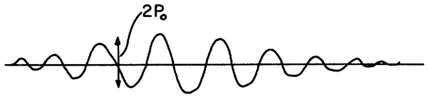
The low-intensity transmission of the sample was 10%, but the over-all transmission for the high-intensity pulse pairs was roughly 50%. As a rough approximation, the net transmission of the sample will be considered to be 10% at the minima of the diffraction pattern and 60% at the maxima. In terms of the model used for the slow effect, this means A [Eq. (19)] is $0.25/W$ and V is 0.06. Studying Eq. (22) for $n = 1$, the second term will be the important one when $U = 0$ at the beginning. And it will be roughly the same size as the first term when $U = 0.1$ as the slow scattering develops. This is in accord with the observations [Fig. 4(b)], which found slow and fast effects of roughly the same size.

Other possible causes of fast scattering have been described in several recent articles[9–11] which review the problem of scattering in absorbing liquids. These other effects do not seem to fit the results reported here, for the reasons enumerated below.

Fast scattering caused by changes in the index of refraction as a simple temperature dependence [second term of Eq. (1)] is ruled out by the fact that the green probe light was unaffected. Also,



"Realistic" Rayleigh Temperature Wave



"Realistic" Brillouin Pressure Wave

FIG. 11. More realistic representation of the waves of Fig. 10.

a true temperature effect would be cumulative over several pulses until the density effect became dominant, and this was not observed.

Actually, in the duration of a single pulse, a new temperature is not established. Mack proposed[6] that the energy input to the solvent, as entropy rather than temperature, changed the index of refraction. Again, this is ruled out in the present results by the fact that the probe light was not scattered.

Four-photon interactions[17,18] (light-by-light scattering) have a geometry similar to these experiments. But they do not depend on the presence of an absorber in the liquid.

Four-photon interactions from the EK9860 molecules are possible and similar to some proposed stimulated-scattering effects.[9] But all such light-by-light scattering effects would exhibit a sensitive relation between power levels and the angle between the beams, even for a two-strong-beam experiment. The fast effect reported here was essentially independent of angle over a factor of 2, and simply increased regularly (faster than linearly) with input power.

Scattering of picosecond pulses in nonsaturable absorbers and in transparent liquids has been reported by Mack.[6,7] No equivalent of the fast shutter was employed, so the time development is not completely certain, though two different time scales of scattering were clearly involved.[7] His results are difficult to compare with the negative results reported here, because the experiment was of the one-strong-beam–one-weak-beam type in a much thicker sample, and the pulses were of higher intensity than those used for this experiment,[19] though the pulses were also shorter and the energy density was about the same. So the experimental situation regarding picosecond scattering in nonsaturable liquids at extremely high power levels is not yet completely settled.

Approximations

In the preceding analysis, the behavior of the liquid has been severely simplified from reality. Without more precise measurements available, it does not seem productive to carry this analysis much further. However, a listing of some of the factors that have been ignored may be useful.

First, this analysis has assumed that the "waves" created in the liquid are quite extensive. In actuality, they are rather short "wave" trains, covering only ten or so cycles, as indicated in Fig. 11 which is to be compared with the idealized case represented in Figs. 9 and 10.

The average pressure and temperature in the excited region are higher than that in the surrounding liquid. Hence, superposed on the wave behavior will be broad *T* and *P* peaks, which will diffuse and radiate outward from the region of excitation. Since they contain spatial frequencies much lower than those of the waves, they will have small effect on the scattering.

Another complication is the effect of the weak scattered radiation on the liquid. This diffracted light will interfere with the high-power beams, and produce small additional effects in the liquid. This is described in the original theories of STBS, STRS, and STS, where it is in fact the only effect. Particularly in the case of samples that are "thin" enough to be treated as diffraction gratings, this effect will be very small.

The induced transmission grating in saturable absorbers is not a simple sine wave, and it is not small in the sense required for the calculation of the integral [Eq. (21) or (22)]. This results in coupling to more terms in an expression like (22) for a given order of diffraction. This would be important if accurate comparisons of the intensity in various orders were to be made.

Radiation passing through the liquid will exert influences, such as electrostriction, additional to the heat-absorption effect considered here. These could be taken into account by adding additional terms to Eqs. (5) and (7), and would not produce any qualitative changes in the theory.

Also, the radiation energy is not absorbed uniformly throughout the thickness of the sample, as was assumed to get Eq. (4). Instead, the absorbed light intensity is greatest where the light enters the sample and decreases as the light passes through. Compared with the idealized distribution used here, this actually produces a slightly lessened phase retardation, as there is some "washing out" of the pressure distribution as the front and rear of the cell come into equilibrium. However, the only effect on the form of the phase retardation will be some deviation from the simple sine-wave shape.

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Plasma Density Fluctuations in the Presence of Optical Mixing of Light Beams*

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The density-fluctuation spectrum and wave-vector distribution are calculated for a plasma subjected to the influence of two intense laser beams. The influences of the plasma ions, a possible external uniform magnetic field, the spectral intensity distribution of the mixed laser beams, and their finite diameter are taken into account. The spectrum is shown to consist of two contributions, of which one is recognized as the thermal-fluctuation spectrum while the other is induced by a force generated by the mixed laser beams. Both contributions simply add as a result of the incoherent nature of the thermal fluctuations. It is shown that the theoretical results are in agreement with recent experimental observations. Special attention is also directed to the case in which two beams, generated by the same laser, are mixed in a plasma, and it is shown that enhancement of the ion feature in the spectrum of more than two orders of magnitude against the thermal case can be produced by employing common *Q*-switched ruby lasers.

I. INTRODUCTION

Methods for generation of electrostatic waves of controlled amplitude, spectrum, and wave-vector distribution are of great interest in plasma physics since they are essential for the study of many nonlinear plasma phenomena. One of the most promising methods, which does not rely on perturbing mechanical probes and which can be used in high-density high-temperature plasmas, makes use of the mixing effect of two laser beams passing through a plasma. First proposed for a Lorentz plasma,[1] the mixing effect has meanwhile been observed in an experiment,[2,3] while the theory has been extended to include the effect of the ions and a uniform external magnetic field.[4] So far, however, theoretical calculations have treated the mixed light waves as plane and monochromatic, and a comparison between observation and theory has not been possible, since the nonlinear character of the light mixing effect prohibits a straightforward extension of the theoretical results to the case of polychromatic light beams of finite diameter. For light scattering experiments, the knowledge of spectral and wave-vector distributions of the density fluctuations are important for the design of the optical detection system. In the present study we therefore propose to develop the theory for physically realistic situations, which would make possible accurate predictions of and comparison with experimental results. In general, the effects of plasma ions, a possible uniform external magnetic field, and the initial plasma conditions are included.

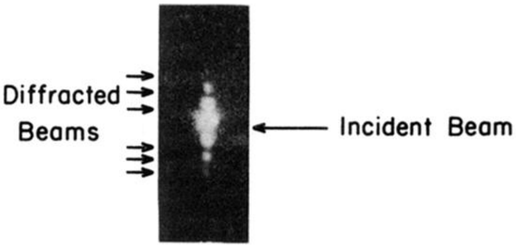

FIG. 8. Polaroid film exposed to scattered green light. Apparatus of probe experiment without the shutter.