ments[2] indicate that both $C_P$ and $\alpha$ diverge as $\ln|T-T_\lambda|$, so that their ratio is well behaved. This method of heat (or cool) pulses in fact offers a method of verifying that both $C_P$ and $\alpha$ diverge in the same manner. Such a study would only be interesting very close to $T_\lambda$, which was beyond the scope of the present experiment.

This observation of "cool" pulses may be rather unique in that although it should occur in all materials that have $\alpha<0$, the actual observation is favored by low specific heat (hence low temperatures) and large $|\alpha|$ [cf. Eq. (1)]. Many solids have negative $\alpha$ at low temperatures, but their ratios $\alpha/C_P$ are typically many orders of magnitude less than for He. (This is also true of water near its freezing point.) Thus only under very large shock heating might "cool" pulses be observed in materials other than He II.

I wish to thank Mr. John P. Anderson for skillful technical assistance. Conversations with Professor K. Dransfeld and Professor I. Rudnick have been helpful in understanding the phenomenon reported here.

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[2]M. J. Buckingham and W. M. Fairbank, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1961), Vol. 3, p. 80.

[3]This was suggested by I. Rudnick, private communication.

[4]K. R. Atkins, *Liquid Helium* (Cambridge Univ. Cambridge, England, 1959).

[5]This differs from the observation by J. R. Pellam [Phys. Rev. 75, 1183 (1949)] of a pressure (first sound) pulse generated by a heat pulse which is much larger in He II than in He I. Dr. Pellam does not mention any *heat* arriving at the velocity of first sound.

# Diffractive Scattering of Picosecond Light Pulses in Absorbing Liquids*

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The time development of diffractive scattering of picosecond light pulses from optically induced gratings in absorbing liquids is studied experimentally. Scattering begins quickly (in picoseconds) in saturable absorbers, but the scattering (stimulated thermal scattering) in a nonsaturable absorber requires many nanoseconds to develop. This resolves a discrepancy between theory and a previous experiment on stimulated thermal scattering.

When optical energy is absorbed in a liquid, changes occur in the index of refraction of the liquid. Usually, the principal coupling is through the density changes resulting from heating.

Mack[1] reported scattering of laser pulses which were incident on an absorbing liquid in such a manner as to produce a grating pattern of absorption. The incident light consisted of a train of picosecond pulses, spaced several nanoseconds apart, from a mode-locked ruby laser. Scattering was caused by a periodic change induced in the refractive index of the liquid.

Mack interpreted the results as indicating that the scattering of each pulse proceeded independently of other pulses in the train. Hence index changes had to occur in a few picoseconds, which is fast enough to rule out the usual coupling through density change. Recent theories of stimulated thermal scattering (STS)[2,3] do not account for such rapid changes in the refractivity of the liquid.

The present experiment was planned to eliminate any possible amibguity concerning the time required for scattering to begin. The essential improvement compared with Mack's arrangement is the use of a fast electro-optic shutter between the laser and the sample liquid. The shutter has an extinction ratio better than 500:1, and a rise time of 3 nsec. The shutter does not open until the pulses from the laser are near maximum amplitude. Hence the sample is not exposed to more or less continuous radiation during the onset of lasing and mode-locking.

By observing the diffractively scattered light with fast detectors, two possible cases can be distinguished: Scattering begins with the first pulse that hits the sample, indicating a picosecond-response effect; or scattering begins only after several pulses have hit the sample, indicating a much slower coupling to the mechanism

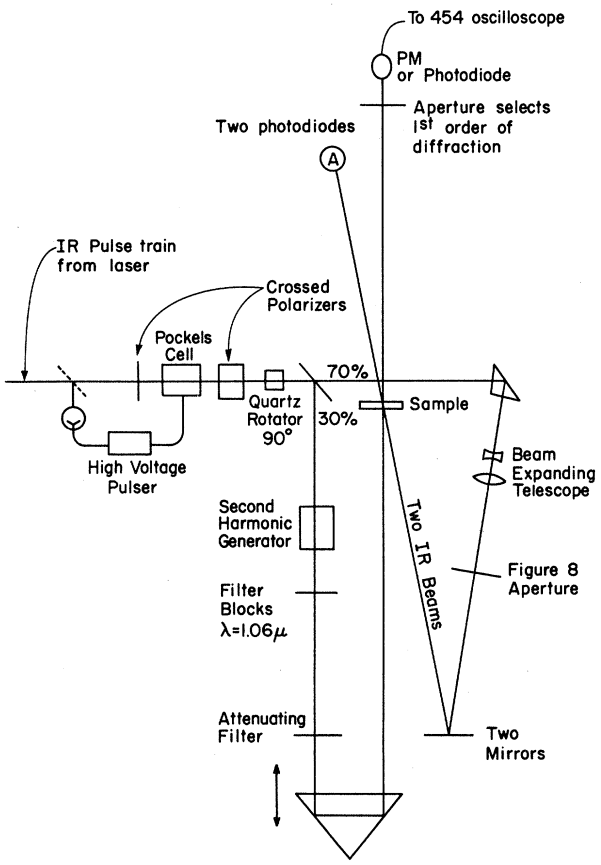


FIG. 1. Arrangement of the apparatus. At the point marked *A* are two photodiodes, one above the other. The upper one is connected to a 519 oscilloscope and monitors the IR pulse train by looking at the lower of the two transmitted beams. The lower diode is positioned in the first scattered IR beam below the two transmitted beams, and can be connected to the 454 oscilloscope to study the IR scattering.

of diffraction.

Observations are that scattering generally does not occur with the first pulse, but rather begins about 50 nsec after the pulse train first hits the sample in agreement with theories of STS.[2,3] Scattering of the first pulse occurred only when the sample was a saturable absorber (EK 9860 dye).

The apparatus employed is depicted in Fig. 1. The Nd-glass laser is mode-locked with EK 9860 dye, and several different techniques of pulse-width measurement yield values of 5-8 psec. Spacing between pulses is about 4 nsec.

After passing through a figure-eight shaped aperture, the light at $\lambda = 1.06$ $\mu$m was reflected by two plane mirrors arranged in the reflective equivalent of a Fresnel biprism. The resulting two beams were coincident on the sample. The power density in each beam was about $1\text{-}2\times10^{9}$ W/cm$^2$; the angle between them was about $5\times10^{-3}$ rad, resulting in a grating pattern of about 5 lines/mm.

The apparatus also allowed probing the grating with a single beam of pulses at $\lambda = 0.53$ $\mu$m. These green pulses could be advanced or delayed up to 70 psec relative to the infrared (IR) pulses that created the grating.[4] Location of zero delay was determined by substituting $CS_2$ for the sample and using one of the IR beams to produce an optical Kerr effect.[5]

A glass cell provided a 0.7 mm long path through the liquid samples. The first sample was EK 9860 dye (an organic dye in dichloroethane). Low-intensity absorption coefficients for this dye were $\alpha = 10$ cm$^{-1}$ at $\lambda = 1.06$ $\mu$m, and $\alpha = 2$ cm$^{-1}$ at $\lambda = 0.53$ $\mu$m. The absorption peak at 1.06 $\mu$m is known to be saturable—an input intensity of 56 MW/cm$^2$ reduces $\alpha$ to half its low-intensity value,[6] and the dye recovers its low-intensity absorption about 8 psec after the high-intensity light is turned off.[6,7]

The second sample was cupric nitrate in acetone. Absorption coefficients were $\alpha = 10$ cm$^{-1}$ at $\lambda = 1.06$ $\mu$m and $\alpha = 1$ cm$^{-1}$ at $\lambda = 0.53$ $\mu$m. No saturation of absorption has been observed for this material, which was chosen for its similarity to samples studied in Ref. 1.

Two *p-i-n* photodiodes were aligned to observe one of the transmitted IR beams and the adjacent diffracted IR signal. The diffracted green signal was detected with a third diode, or with a 931A photomultiplier, overvoltaged and with modified output connections to increase speed of response.

In observations on EK 9860 dye with the fast shutter omitted, diffraction was similar to previous results.[1] Three orders of diffraction of the green light were observed on a photographic plate 1 m behind the sample. Detected with the photodiode, the green diffracted signal followed in amplitude the monitored IR radiation[8]; the same was true for the diffracted IR. Typically, diffracted signals were 5-10 times greater than background noise observed when one of the IR beams to the sample was blocked.

With the fast shutter in use, and with the same sample, diffracted green light was detected with the photomultiplier. The signal rose above noise level only after about 12 pulses had passed through the sample. With the diffracted IR however, a signal 2-3 times noise level was present from the very first pulse; after about 12 pulses the diffracted signal began rising again, finally

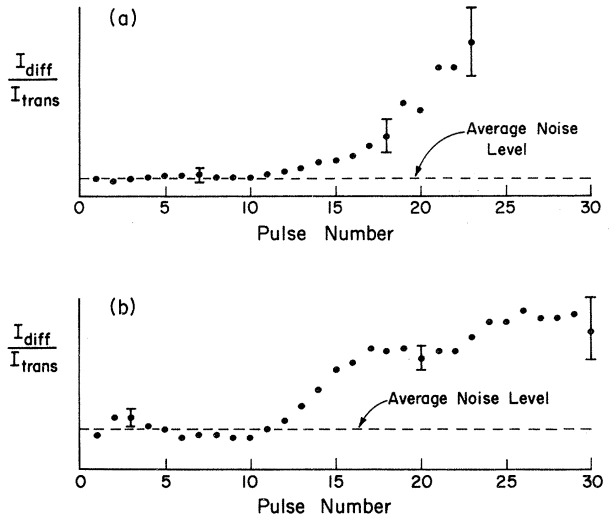


FIG. 2. Typical ratios of diffracted light to transmitted IR light for the cupric nitrate sample (arbitrary units). Each graph is taken from a single-pulse train recorded simultaneously with two oscilloscopes. (a) Shows diffracted IR light; (b) shows diffracted green. The average noise level is the ratio observed when one of the two IR beams aimed at the sample is blocked.

reaching 5-10 times noise level.

When the sample was cupric nitrate in acetone, with the fast shutter in use, immediate scattering was never seen. Instead, both IR and green pulses showed diffraction that did not rise above noise until after about 12 pulses had passed through the sample (see Fig. 2).

In these experiments, no dependence of green-light scattering with delay could be discerned. However, as delay observations depended on reproducibility of the laser between successive shots, a variation as large as 50% might have gone unnoticed.

For samples of pure dichloroethane or acetone, no diffraction was observed.

A likely explanation of the delayed scattering is the formation of both oscillating (Brillouin) and and stationary (Rayleigh) density waves in the liquid. Recent papers[2,3] on STS have presented the theory of a very similar experiment in which a strong and weak beam intersect at a liquid sample, instead of two equal beams as in the present experiment.

For the liquids and geometry employed here, the delay of 50 nsec before the detection of scattering corresponds to about $\frac{1}{4}$ of an acoustic cycle, which seems quite reasonable. (In the notation of Ref. 2, $\omega_B t_s/\pi = 0.05$.) The behavior of the density wave is complicated by the continuing input of energy, and its exact evolution depends on details of the envelope function of the laser pulse train.[2] Possible applications of this type of acoustic wave generation have been discussed by Auth.[9]

When the entire laser pulse train is allowed to interact with the sample, the density wave apparently reaches a large value by the time the mode-locked pulses have fully developed. (This requires that a significant amount of the laser's output energy is not mode-locked at the onset of lasing, in agreement with present understanding.)

On the other hand, density changes cannot explain the observed scattering of the first IR pulse from the EK 9860 dye. This prompt scattering is presumably caused by the saturation of the dye.[6] The incoming IR radiation bleaches the dye in a grating pattern, and the resulting amplitude grating then diffracts both IR beams. The green light is not directly affected, since the sample is nearly transparent to green light anyway. This type of saturated-dye diffraction grating has previously been observed[11] only with much longer (nanosecond) pulses.

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