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EXPERIMENTAL OBSERVATION OF STIMULATED THERMAL BRILLOUIN SCATTERING

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Stimulated thermal Brillouin scattering—a phenomenon which exists in absorbing media—has been experimentally investigated. The gain of a light amplifier was measured as a function of frequency, absorption coefficient, and time during the laser pulse. The striking difference between absorbing and nonabsorbing liquids was found to be in excellent agreement with theoretical predictions.

It has been shown in several recent publications that the application of light amplifiers offers a series of advantages for the investigation of stimulated scattering phenomena; e.g., quantitative data of the gain factors for Raman,[1] Rayleigh wing,[2] and Brillouin[3,4] scattering were obtained in oscillator-amplifier systems. Very recently Herman and Gray[5] have theoretically analyzed stimulated processes in absorbing liquids. They predicted thermal scattering phenomena in the neighborhood of the Rayleigh and Brillouin lines. While the first process has been investigated experimentally,[6] the second stimulated phenomenon has not yet—to our knowledge—been experimentally observed. We wish to report on the experimental confirmation of this new scattering process. Using an oscillator-amplifier system, quantitative data were obtained and excellent agreement with the theoretical predictions was achieved.

The change in density ρ under the influence of an electric field E can be written in the form[5,7,8]

$$\frac{\partial^2}{\partial t^2}\rho - v^2\nabla^2\rho - \frac{\eta}{\rho_0}\frac{\partial}{\partial t}\nabla^2\rho = \frac{1}{8\pi}\nabla^2[\gamma^e E^2 + 2\pi\nu_B\gamma^a\int E^2 dt]. \quad (1)$$

The left-hand side of Eq. (1) represents the wave equation for ρ, while the driving terms on the right-hand side consist of an electrostrictive component with a coupling coefficient γ^e,[8] and a component due to absorptive heating with a coupling coefficient γ^a. ν_B is the Brillouin shift of the amplifier, $\nu_B = 2\nu_L n v/c$, and $\nu = \nu_L - \nu_S$, where ν_L and ν_S are the laser and Brillouin-Stokes frequencies, respectively. For $\gamma^a = 0$ we obtain the gain of the well-known stimulated Brillouin scattering with a Lorentzian frequency profile[4,8]:

$$g^e(\nu) = g_{max}^e/[1 + 4(\nu-\nu_B)^2/\delta\nu^2], \quad (2)$$

where

$$g_{max}^e = (2\pi\nu_L^2\gamma^{e2})/(c^3 n\nu\rho\delta\nu) \quad (3)$$

and

$$\gamma^e \simeq \tfrac{1}{3}(n^2-1)(n^2+2); \quad (4)$$

when absorptive heating is included an additional gain factor $g^a(\nu)$ is obtained[5]:

$$g^a(\nu) = g_{max}^a \times 2\frac{2(\nu-\nu_B)/\delta\nu}{1+4(\nu-\nu_B)^2/\delta\nu^2}, \quad (5)$$

where

$$g_{max}^a = (2\pi\nu_L^2\gamma^e\gamma^a)/(2c^3 n\nu\rho\delta\nu) \quad (6)$$

and

$$\gamma^a = \alpha(vc^2\beta)/(2\pi\nu_L c_p). \quad (7)$$

The parameters in Eqs. (1) to (7) are defined as follows: v is the velocity of sound, η the bulk viscosity, ρ_0 the equilibrium density, β the cubic thermal expansion coefficient, c_p the specific heat, α the absorption coefficient at the laser frequency ν_L, $n=\sqrt{\epsilon}$ the refractive index, and $\delta\nu$ the full half-width of the Brillouin line of the material investigated; g^e and g^a are steady-state gain factors (see below).

The experimental system (described previously[4]) is depicted in Fig. 1(a). It consists, briefly, of a single-mode giant-pulse ruby laser with an output pulse of power P_L (pulse maximum ~1.5 MW), a generator for back-reflected Brillouin power P_i, and an amplifier for Brillouin light producing a signal P_a. The use of a light amplifier is essential for the investigation of stimulated thermal Brillouin scattering since it allows the elimination of other competing stimulated processes. The parameters of the amplifier and the intensity level of the laser were chosen in such a way that—in spite of its higher gain constant—stimulated thermal Rayleigh scattering did not reach the threshold value for oscillation. For each laser pulse the three signals P_L, P_i, and P_a were recorded simultaneously with a photoelectric detection system having an overall time constant of 0.3 nsec. By varying the liquid medium in the generator the Brillouin frequency of signal P_i was shifted by 1000 Mc/sec; in this way the gain profile of the medium in the amplifier cell could be investigated. In the experiments reported here a constant mixture of 66% CS_2 and 34% CCl_4 (by volume) was chosen in the amplifier cell in order to make the necessary frequency shifts possible for P_i (by changing the ratio of the two liquid components in the generator). In Fig. 1(b) the experimental gain factor g_{exp} is presented as a function of the frequency difference $\nu_B-\nu$ (difference between the frequency of the incoming light $\nu_S=\nu_L-\nu$ and the frequency of the center of the Brillouin line of the amplifier medium $\nu_L-\nu_B$). The gain factor was calculated from the pulses P_L, P_i, and P_a according to[4]

$$P_a/(P_i e^{-\alpha l}) = 1 + g_{exp} P_L l/(\pi w^2). \quad (8)$$

α is the absorption coefficient of the amplifier medium. The length of the amplifier cell $l=0.2$ cm, the spot size[9] of the laser beam at the position of the amplifier cell $w=4\times10^{-2}$ cm, and the incoming signal P_i were adjusted for small signal theory to apply. Figure 1(b) contains two sets of points. The solid squares were obtained

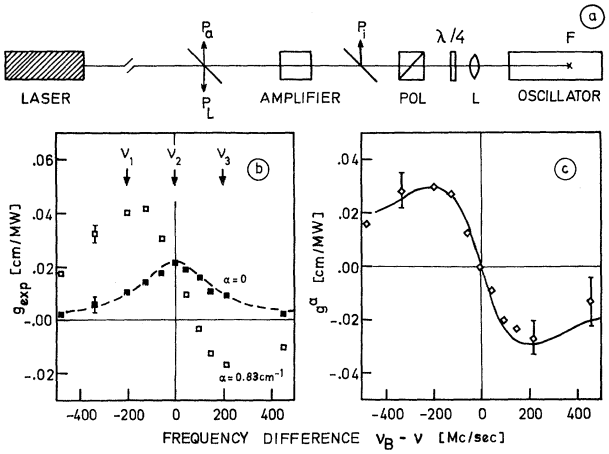

FIG. 1. (a) Schematic of the experimental system. (b) Experimental gain factor versus frequency for a pure liquid ($\alpha=0$, solid squares, Lorentz curve drawn through points) and an absorbing liquid ($\alpha=0.83$ cm^{-1}, open squares). (c) Difference of gain factors of the two liquids presented in (b); curve calculated according to Eqs. (4)-(7). The positive gain maximum is red shifted.

for a transparent liquid ($\alpha<10^{-2}$ cm^{-1}) in the amplifier while the open squares were found when the liquid was slightly colored (by dissolving a small amount of iodine) with an absorption coefficient of $\alpha=0.83$ cm^{-1}. There is a striking change between these two measurements. It is clearly seen from Fig. 1(b) that for the absorbing liquid the gain maximum is increased and shifted to a smaller frequency by approximately $\frac{1}{2}\delta\nu$ and—most interesting—that a negative gain factor is observed for higher frequencies. A negative value of g_{exp} indicates that $P_a/P_i e^{-\alpha l}<1$, i.e., the incident signal P_i is attenuated in passing through the "amplifier" cell. In this case Brillouin-Stokes photons plus energy from the thermal fluctuations are converted back into laser photons.

Through the solid squares (transparent liquid) a Lorentz curve is drawn which gives a width at half-maximum gain of $\delta\nu=360$ Mc/sec indicating a phonon lifetime of our liquid mixture of $\tau=0.4$ nsec. (The frequency width of the laser pulse and of the incident signal of $\simeq20$ Mc/sec each have been taken into account.)

In Fig. 1(c) the difference of the gain factors of the two liquids, with and without absorption, $g^a=g_{exp}^{with}-g_{exp}^{without}$ is plotted as a function of frequency. The curve drawn through our experimental points is calculated from Eqs. (4)-(7). The good agreement between theory and

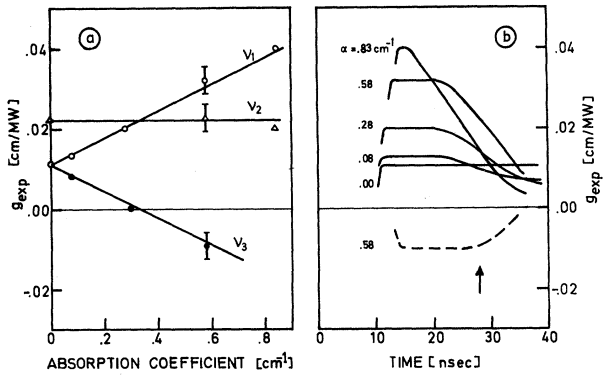

FIG. 2. (a) Gain factor versus absorption coefficient for three frequencies: $\nu_B-\nu_1=-200$ Mc/sec (open circles); $\nu_B-\nu_2=0$ (triangles); $\nu_B-\nu_3=+200$ Mc/sec (solid circles). (b) Experimental gain factors versus time during the laser pulse for liquids with different absorption coefficients. Data are presented for the Stokes shifted frequency difference $\nu_B-\nu_1=-200$ Mc/sec (solid lines) and for the anti-Stokes shifted frequency difference $\nu_B-\nu_3=+200$ Mc/sec (dashed line). The arrow indicates the maximum of the giant pulse.

experiment—including the predicted red shift—is noteworthy. There are no fitting parameters in Fig. 1(c). The physical parameters entering the quantitative determination of the factor g^a were taken from standard handbooks. The following numbers were used: $n=1.56$, $\nu=1.17\times10^5$ cm sec^{-1}, $\beta=1.20\times10^{-3}$ (°C)$^{-1}$, $c_p=0.91$ J g^{-1} (°C)$^{-1}$, $\nu_L=4.3\times10^{14}$ sec^{-1}, $\rho=1.31$ g cm^{-3}, $\delta\nu=3.6\times10^8$ sec^{-1}.

We have extended our investigations to liquids with different absorption coefficients α. In Fig. 2(a) experimentally observed values of $g_{exp}=g^e+g^a$ are plotted as a function of α for three different frequencies [which are indicated by arrows in Fig. 1(b)]. As expected from Eqs. (5) and (6), for $\nu=\nu_B$, we obtain $g=g^e$ which is constant, independent of α, while for $\nu\neq\nu_B$ a clear linear dependence of g on the absorption coefficient is observed. The lines drawn through our experimental data are again calculated from Eqs. (2)-(7). There is good quantitative agreement for the absorption coefficients used in our investigations.

The time dependence of the gain factor g_{exp} during the course of the laser pulse is depicted in Fig. 2(b) for various liquids with absorption coefficients $\alpha=0$, 0.08, 0.28, 0.58, and 0.83 cm^{-1}. It is seen from Fig. 2(b) that for nonabsorbing liquids a stationary value of g_{exp} is rapidly reached (transient time approximately $2\tau\simeq1$ nsec) which was previously shown to correspond to the steady-state gain factor of the medium.[4] For absorbing liquids a stationary value g_{exp} is reached very rapidly again [as expected from Eq. (1)]. The comparison with the results for $\alpha=0$ strongly suggests that we have obtained true steady-state values; these data were presented in Figs. 1(b) and 2(a) The decrease of g_{exp} for later times seems to be caused by an overall rise in temperature of the amplifying medium. From Fig. 2(b) together with the quantitative agreement with a steady-state theory, we wish to conclude that we have indeed observed steady-state gain factors in our experiments.

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