PHONON LIFETIMES MEASURED IN AMPLIFIERS FOR BRILLOUIN RADIATION

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The transient and steady-state behavior of stimulated Brillouin scattering in amplifiers was investigated theoretically and experimentally. There is good agreement between calculations and measurements with high time resolution. Direct methods for measuring phonon lifetimes are presented.

Recently considerable theoretical[1-3] and experimental interest has been focused on the steady-state[4-6] and transient[7] problem of stimulated Brillouin scattering. Most investigations were concerned with an analysis of the oscillator where a steady-state regime in liquids is now well established.[5,6] In this paper we wish to report on a number of experiments of an oscillator-amplifier system.[8] By proper choice of the input signal into the amplifier we were able to observe steady-state and transient phenomena; the steady-state gain factor $g(0)^2$ for stimulated Brillouin scattering was obtained using a single laser pulse. Phonon lifetimes can be determined from $g(0)$ or by introduction of a definite frequency difference between oscillator and amplifier.

The experimental setup is depicted in Fig. 1(a). The light of a ruby giant pulse laser (operating in a TEM_{00} mode) traverses first the amplifier liquid cell (length between 0.1 and 1 cm) and then generates intense Brillouin radiation (conversion efficiency $\leq 90\%$) in an oscillator cell. The backward traveling Brillouin light is strongly attenuated by the use of a polarizer and a $\frac{1}{4}\lambda$ plate. For each laser pulse three signals, the laser power P_L, the incoming Brillouin power P_i, and the amplified Brillouin power P_a, were measured with the same phototube (using appropriate time delays). The over-all time constant of the photodetection system is approximately 0.3 nsec. We have carefully investigated the change in frequency occurring during the emission of a giant pulse laser.[9,10] We found a frequency drift of $\simeq$ Mc/sec per nsec (or approximately 350 Mc/sec per pulse).[10] This value, while small compared with the laser frequency of 4.3×10^{14} cps, is quite important in our investigations since the phonon linewidths can be as small as 63 Mc/sec (in CS_2).

Our experimental system has a series of advantages which allow a direct comparison between theory and experimental results. (1) The amplification is measured as a function of time making transient and steady-state phenomena readily observable. (2) During each pulse the amplification is determined as a function of instantaneous laser intensity which allows an immediate determination of the gain coefficient g (see below). (3) The incoming Brillouin signal P_i can be tailor made by proper adjustment of the oscillator. We have analyzed the amplification of slowly and abruptly rising incoming signals P_i. The small value of P_i ($P_i/P_L \simeq 10^{-2}$) avoids saturation of the amplifier and allows a comparison of our data with a small-signal theory (outlined below). (4) Well-defined frequency differences between oscillator and amplifier are readily introduced, which provide a most direct determination of phonon lifetimes. (5) Since our system allowed the observation of small amplification values (<2) with good accuracy, small light intensities and short lengths of the amplifier cells could

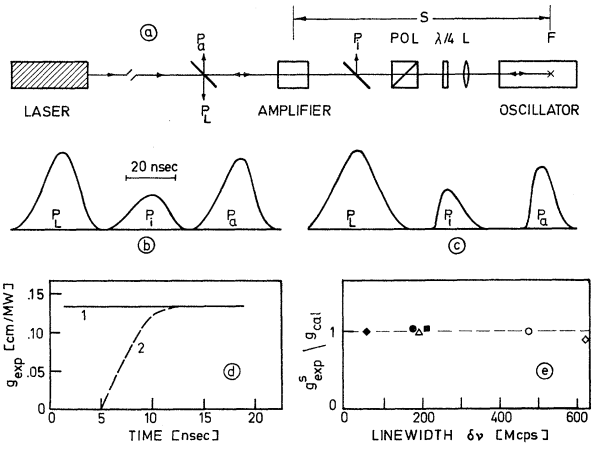

FIG. 1. (a) Schematic of the experimental system. (b) Oscilloscope traces depicting the laser pulse P_L, the incoming signal P_i, and the amplified signal P_a ($\Delta\nu=0$). (c) same as (b) with abruptly rising signal P_i ($\Delta\nu=0$). (d) Experimental gain factor of CS_2 versus time ($\Delta\nu=0$): (1) slowly rising P_i and (2) abruptly rising P_i. (e) Ratio $g_{exp}{}^S/g_{cal}$ versus phonon linewidth $\delta\nu$ for six different liquids: solid diamond, CS_2; solid circle, acetone; triangle, methanol; solid square, n-hexane; open circle, toluene; and open diamond, CCl_4.

be used. In this way, the occurrence of other competing nonlinear processes such as stimulated Raman scattering, self-focusing, and self-trapping was avoided.

Starting with nonlinear wave equations for the electric and acoustic fields[3,7] one derives the following equation for the Brillouin field G:

$$\frac{\partial G(z,t)}{\partial z}\frac{1}{2\tau'}+\frac{\partial^2 G(z,t)}{\partial t\,\partial z}=\frac{g(0)I_L}{4\tau}G(z,t), \tag{1}$$

where

$$\tfrac{1}{2}\tau' = \tfrac{1}{2}\tau + i2\pi\Delta\nu$$

$$\text{and } g(0) = 2\pi\nu_L{}^2\gamma^2/c^3 n\rho v\,\delta\nu; \tag{2}$$

$g(0)$ is the steady-state gain factor.[2] τ and v are the lifetime and velocity of the acoustic phonons, respectively. $\delta\nu = \frac{1}{2}\pi\tau$ is the width of the classical Brillouin line and $\Delta\nu$ is the frequency difference between the incoming signal and the peak of the classical Brillouin line in the amplifier. I_L and ν_L are the intensity and frequency of the laser light; γ is the elasto-optic coupling coefficient,[2] n the index of refraction, c the velocity of light, and ρ the density. In deriving Eq. (1) the assumption (fulfilled for the materials investigated) is made that the phonons are heavily damped, i.e., $\tau \ll l/v$ (l is the length of the amplifier) and that the time l/c is small compared with the signal risetime. For the case of small Brillouin signals, small amplification, and for an incoming step function, the gain in an amplifier has the form[11]

$$P_a/P_i = 1 + g(\Delta\nu)P_L\, l/\pi w^2[1 + e^{-t/2\tau} \times (2\Delta\nu/\delta\nu \sin 2\pi\Delta\nu t - \cos 2\pi\Delta\nu t)], \tag{3}$$

where the gain factor $g(\Delta\nu) = g(0)/[1 + (2\Delta\nu/\delta\nu)^2]$. w is the spot size[12] of the laser and Brillouin beam.

Our experimental observations are briefly summarized as follows: (A) First, the frequency difference $\Delta\nu$ is kept to a minimum, then (B) a well-defined frequency difference of $\Delta\nu$ <150 Mc/sec is introduced.

(A) The distance S between the point of generation of the back reflected Brillouin signal and the short amplifier cell was held small. The experimental system was designed in such a way that S was approximately 5 cm, which results in $\Delta\nu \simeq 5$ Mc/sec.[10] In Fig. 1 (b) a typical oscilloscope trace containing the three relevant signals P_L, P_i, and P_a is shown; the investigated material is CS_2. By proper choice of the lens L it is possible to initiate stimulated Brillouin emission in the oscillator at the very beginning of the laser pulse resulting in a slowly rising signal P_i. An analysis of the signals P_L, P_i, and P_a clearly indicates that steady-state amplification is obtained for the total time of the pulse. The gain factor for small amplification g_{exp} determined from $P_a/P_i = 1 + g_{\text{exp}}P_L l/\pi w^2$ is plotted as a function of time in Fig. 1(d). The time (and intensity!) independent value of g_{exp} indicates that we are working under quasi-steady-state conditions.

Quite different is the situation when the incoming Brillouin signal P_i consists of an abruptly rising pulse [Fig. 1(c)]. Such pulses are readily generated in the oscillator by increasing the focal length of lens L and initiating the stimulated Brillouin emission at a time when the laser pulse has reached a large fraction of its peak power. An analysis of the signals shows a distinct transient response of the system. In Fig. 1(d), the experimental gain factor g_{exp} is seen to approach the steady-state value $g_{\text{exp}}{}^S$ with a time constant which corresponds according to Eq. (3) to approximately twice the phonon lifetime of the material investigated. In Fig. 1(e) the ratio $g_{\text{exp}}{}^S/g_{\text{cal}}$ is plotted as a function of the phonon linewidth $\delta\nu = 1/2\pi\tau$ for six liquids, CS_2, acetone, n-hexane, methanol, toluene, and CCl_4. The excellent agreement between experimental and calculated gain factors for values of $\delta\nu$ between 60 and 630 Mc/sec is noteworthy.

For materials where the phonon lifetime is unknown, the experimentally determined steady-state gain factors allow a direct calculation of τ [Eq. (2)]. It should be emphasized that values of τ can be obtained with good accuracy from a single laser pulse. The shortcoming of this technique rests in the fact that we need absolute laser intensities for the determination of τ (calibrated phototubes). This difficulty is avoided in the next section where a different technique for the determination of τ will be discussed.

(B) A well-defined frequency difference $\Delta\nu$ was established in two ways: first, by increasing the distance S and taking advantage of the inherent frequency drift of the laser itself, and second, by slightly changing the composition of the medium in the oscillator; i.e., the Brillouin shift $\nu_B = 2\nu_L vn/c$ is varied by changing the values of v and n. Similarly to Sec. (A),

our time-resolved observations have to be subdivided depending upon the pulse shape of P_i. For a slowly increasing signal P_i again a steady-state amplification is found. But—as expected—the observed gain factor $g_{\mathrm{exp}}{}^{S}$ is reduced [see Fig. 2(a)] by the Lorentz factor $[1+(2\Delta\nu/\delta\nu)^2]^{-1}$ [compared with the gain factor $g(0)$ with $\Delta\nu \simeq 0$]. When an abruptly rising incoming pulse P_i was used, an oscillating transient signal P_a was observed. A typical time-dependent gain factor is presented in Fig. 2(a). Such an oscillatory behavior is predicted from the small-signal theory outlined above. In fact the agreement between the experimentally observed oscillation of g_{exp} and a more extended calculation is excellent when the true pulse shape of the signals is taken into account.[11]

The most direct method of determining the phonon lifetime consisted in using a slowly rising pulse P_i and a well-defined $\Delta\nu$ value by changing the material in the oscillator (CS_2:CCl_4 mixtures[13]). In Fig. 2(b), curve 1, the normalized experimental steady-state gain factors for CS_2 are plotted as a function of $\Delta\nu$. The curve drawn through the experimental points provides us—after the laser and incident signal linewidths of approximately 20 Mc/sec each have been taken into account—with a value of the phonon lifetime of $\tau = 2.7$ nsec which compares favorably with the value of $\tau = 2.5$ nsec obtained from classical Brillouin measurements.[14] The accuracy of our experimental technique is shown quite vividly in curve 2 of Fig. 2(b), where a mixture of 97.5% CS_2 and 2.5% (by volume) CCl_4 was investigated. The decrease of

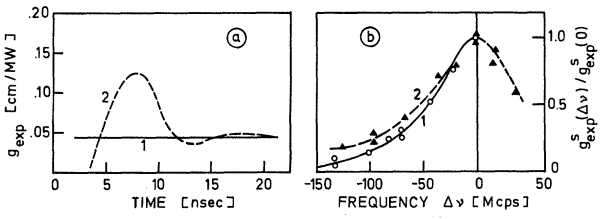

FIG. 2. (a) Experimental gain factor of CS_2 versus time for $\Delta\nu = 75$ Mc/sec: (1) slowly rising Pi and (2) abruptly rising P_i. (b) Normalized experimental gain factor of CS_2 (circles) and a mixture of 97.5% CS_2 and 2.5% (by volume) $CC|_4$ (triangles) ($T = 20°C$).

phonon lifetime in the mixture to $\tau = 1.8$ nsec is easily deduced from our data.

Our investigations clearly indicate that (especially for substances with long phonon lifetimes) a high time resolution is important for the investigation of stimulated Brillouin scattering. With time integrating devices—mostly used in previous experiments—existing transient phenomena are not recognized and accurate determinations of phonon lifetimes are not generally possible.

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[1]R. Y. Chiao, E. Garmire, and C. H. Townes, in Quantum Electronics and Coherent Light, Proceedings of the International School of Physics "Enrico Fermi," Course XXXI, edited by P. A. Miles (Academic Press, Inc., New York, 1964); Y. R. Shen and N. Bloembergen, Phys. Rev. 137, A1786 (1965); K. Grob, Z. Physik 201, 59 (1967).

[2]C. L. Tang, J. Appl. Phys. 37, 2945 (1966).

[3]N. M. Kroll, J. Appl. Phys. 36, 34 (1965).

[4]R. Y. Chiao, C. H. Townes, and B. P. Stoicheff, Phys. Rev. Letters 12, 592 (1964); R. G. Brewer, Phys. Rev. 140, A800 (1965); A. S. Pine, Phys. Rev. 149, 113 (1966).

[5]J. Walder and C. L. Tang, Phys. Rev. 155, 318 (1967); M. Maier, W. Rother, and W. Kaiser, Appl. Phys. Letters 10, 80 (1967).

[6]M. Maier, Phys. Rev. 166, 113 (1968).

[7]E. E. Hagenlocker, R. W. Minck, and W. G. Rado, Phys. Rev. 154, 226 (1967).

[8]P. Lallemand, P. Simova, and G. Bret, Phys. Rev. Letters 17, 1239 (1966); G. Bret and M. Denariez, Compt. Rend. 264, 1815 (1967); M. Denariez and G. Bret, to be published.

[9]V. V. Korobkin, A. M. Leontovich, M. N. Poprava, and M. Y. Shchelev, Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt. 3, 301 (1966) [translation: JETP Letters 3, 194 (1966)]; D. J. Bradley, G. Magyar, and M. G. Richardson, Nature 212, 63 (1966); A. Orszag and R. Saron, Compt. Rend. 264B, 1580 (1967).

[10]D. Pohl, to be published.

[11]A detailed solution will be published elsewhere.

[12]G. D. Boyd and J. P. Gordon, Bell System Tech. J. 40, 489 (1961); G. D. Boyd and A. Kogelnik, ibid. 41, 1347 (1962).

[13]F. Barocchi, M. Mancini, and R. Vallauri, Nuovo Cimento 49B, 233 (1967).

[14]P. A. Fleury and R. Y. Chiao, J. Acoust. Soc. Am. 39, 751 (1966).