Growth of Optical Plane Waves in Stimulated Brillouin Scattering

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Several classical plane-wave theories have been proposed recently to explain the observations of stimulated Brillouin scattering in solids and liquids. While previous experiments have been successful in demonstrating the effect, the use of focused excitation has prevented a critical test of theory. In the present work, the collimated beam of a giant pulse laser source excites a liquid, *n*-hexane, external to a cavity, and allows the Brillouin scattering intensity to be measured as a function of optical path length. The type of transient solution considered by Kroll comes closest to describing the observed gain curve, which includes a large spatial amplification and no apparent threshold condition. The results lead to values for the Pockel's photoelastic constant and the hypersonic acoustic lifetime. With collimated excitation, a striking "acoustic burst" whose intensity parallels the path-length dependence of the Brillouin scattering is noticed. This suggests that the acoustic wave generated in the Brillouin process initiates a shock wave which could lead to material fracture or cavitation. With focused excitation, an additional mechanism involving plasma formation appears to operate.

INTRODUCTION

THE prediction by Chiao, Garmire, and Townes[1] that stimulated Brillouin scattering should occur in transparent media exposed to an intense optical maser has now been verified by experiments on solids[2] and liquids.[3,4] The theory has also been investigated by Kroll[5] who has emphasized that transient-type solutions to the coupled electromagnetic and acoustic wave equations should be important when the excitation period is short, as is the case for a giant pulse. These solutions are characterized by an exponential growth in space and time with no threshold condition. More recently, Shen and Bloembergen[6] have proposed that steady-state solutions, exhibiting only spatial gain, are operative during stimulated Brillouin and Raman scattering.

The purpose of this paper is to determine which of these theories describes the observed growth of the scattered light wave when the exciting source is a giant pulse laser. The earlier stimulated Brillouin measurements[2–4] which utilize focused laser excitation, to enhance the effect, are obviously inadequate for testing theories which treat the scattering of plane waves. Unfortunately, a theoretical analysis for focused excitation which has not been attempted would entail at least the additional difficulties of a complex field distribution[7] and possibly the interference of several incident and scattered optical modes.[8] In the experiments to be described, the collimated beam of a laser source excites a liquid of variable length, thereby allowing a study of the Brillouin scattering buildup with distance and a comparison with theory.

THEORETICAL OUTLINE

The scattering of light by acoustic waves was first predicted by Brillouin.[9] From a classical viewpoint, the incident light wave, of frequency ω and phase vector k, is modulated by the acoustic wave (ω_1,k_1) which provides a periodic variation in the density and in the polarizability of the medium. Thus, the scattered light will be displaced from the exciting light by the acoustic frequency. A simple consideration of energy and phase-vector conservation

$$\pm\omega_1+\omega_2=\omega, \tag{1}$$

$$\pm k_1+k_2=k, \tag{2}$$

appropriate to a traveling acoustic wave, leads immediately to the well-known Brillouin expression for the frequency shift of the scattered light wave (ω_2,k_2)

$$\omega-\omega_2=\pm\omega_1\cong\pm 2(v_1/c)n\omega\sin(\tfrac{1}{2}\theta). \tag{3}$$

Hence, Brillouin scattering resembles a Raman process with the acoustic vibrations replacing molecular vibrations. In (3), v_1 and (c/n) are the acoustic and optical phase velocities in the medium, n is the index of refraction which will be essentially the same for incident and scattered light rays in isotropic media, and θ is their angular separation. Equation (3) will also be recognized as a Doppler shift.

Equations (1)–(3) also imply that acoustic waves can either be produced or annihilated in the scattering process. When acoustic waves are generated, an enhancement in light scattering can occur. With a weak exciting source, the enhancement is negligible, but with

[1] R. Y. Chiao, E. Garmire, and C. H. Townes, *Proceedings of the Enrico Fermi International School of Physics, Course XXXI*, edited by P. A. Miles (Academic Press Inc., New York, 1964), p. 326.
[2] R. Y. Chiao, C. H. Townes, and B. P. Stoicheff, Phys. Rev. Letters **12**, 592 (1964).
[3] R. G. Brewer and K. E. Rieckhoff, Phys. Rev. Letters **13**, 334A (1964).
[4] E. Garmire and C. H. Townes, Appl. Phys. Letters **5**, 84 (1964).
[5] N. M. Kroll, J. Appl. Phys. **36**, 34 (1965); Bull. Am. Phys. Soc. **9**, 222 (1964).
[6] Y. R. Shen and N. Bloembergen, Phys. Rev. **137**, A1787 (1965). *Note added in proof.* Another theoretical discussion has been given just recently by D. L. Bobroff, J. Appl. Phys. **36**, 1760 (1965).
[7] A. Boivin and E. Wolf, Phys. Rev. **138**, B1561 (1965); Bull. Am. Phys. Soc. **9**, 728 (1964).
[8] N. Bloembergen and Y. R. Shen, Phys. Rev. Letters **13**, 720 (1964).
[9] L. Brillouin, Ann. Phys. (Paris) **17**, 88 (1922); Compt. Rend. **158**, 1331 (1914).

a giant pulse laser, the effect is profound. The process then leads to a marked regenerative behavior of amplification or oscillation. The case of collinear backscattering is favored because of the possibility of a large spatial overlap between the two optical waves and the acoustic wave.

A brief sketch of Kroll's transient-type solution will be presented now as it is this case which is in closest agreement with the experimental observations to be presented in the next sections. In contrast, the steady-state solutions are found to be less appropriate and will not be considered at this point. The development which follows is introduced only for the purpose of illustrating its application, and additional details are to be found in the original derivation of Kroll.

The coupling of electromagnetic and elastic waves can be described by an electrostrictive force whose Lagrangian density is

$$L_{\text{int}}=-(1/8\pi)p_{ijkl}D_iD_j(\partial u_k/\partial x_l), \tag{4}$$

where $\delta\nu_{ij}=p_{ijkl}(\partial u_k/\partial x_l)$ is the variation in the reciprocal dielectric tensor ν_{ij} resulting from the strain $(\partial u_k/\partial x_l)$, u_k is the elastic displacement, D_i and D_j are electric displacement vectors corresponding to incident and scattered light waves, and p_{ijkl} is the Pockel's photoelastic constant. Equation (4) implies that any two of the three waves can be used to generate the third. The complete Lagrangian density results in the classical equations of motion for coupled acoustic and optical waves:

$$\rho\ddot{u}_i+\eta_{ij}\dot{u}-\lambda_{ijkl}\frac{\partial^2 u_l}{\partial x_j\partial x_k}=\frac{1}{8\pi}p_{klji}\frac{\partial}{\partial x_j}D_kD_l, \tag{5}$$

$$\frac{1}{c^2}d^2D_j/dt^2-\nu_{ij}\left(\delta_{ik}\nabla^2-\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_k}\right)\times D_j=p_{kjlm}\left(\delta_{ik}\nabla^2-\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_k}\right)D_j\frac{\partial u_l}{\partial x_m}. \tag{6}$$

For the acoustic equation (5), ρ is the mass density, η_{ij} is an acoustic damping coefficient, and λ_{ijkl} is an elastic constant. We shall neglect optical damping.

Kroll shows that (5) and (6) can be reduced by a linearization procedure to a pair of first-order differential equations with constant coefficients

$$(\partial F/\partial\tau)+\beta F+\gamma(\partial F/\partial\xi)=G, \tag{7}$$

$$(v_1/v_2)(\partial G/\partial\tau)+(\partial G/\partial\xi)=F. \tag{8}$$

In this form, the acoustic and optically scattered amplitudes are F and G, respectively. These waves are characterized by a polarization vector $\hat{e}_i$, propagation direction $\hat{q}_i$, frequency ω_i, energy flow v_i, and phase vector k_i with the subscripts 1 and 2 denoting an elastic and a scattered light wave, respectively. We will be interested in the case of a collinearly backscattered light wave with $\gamma=\hat{q}_2\cdot\hat{q}_1=-1$. The reduced quantities time τ, position ξ, and acoustic damping parameter β are given by

$$\tau=v_1t/X,\quad \xi=x/X,\quad \beta=\alpha X, \tag{9}$$

$$X=[32\pi\rho v_1v_2\omega_2\omega_1/P^2k_2^2c^2]^{1/2}, \tag{10}$$

$$P=D_{0k}\hat{e}_{2l}k_1\hat{e}_{1i}p_{klji}.$$

Furthermore, we have

$$F=-(8\pi\rho\omega_1v_1k_2^2c^2/\omega_2v_2)^{1/2}F_1 \tag{11}$$

and

$$u=F_1\hat{e}_1\exp i(\omega_1t-k_1\cdot r). \tag{12}$$

Equations (7) and (8) can be transformed into two integral equations. For the optical wave,

$$G(\xi,\tau)=G_0+\int_{\bar{\xi}}^{\xi}F[\xi_1,\ \tau-(v_1/v_2)(\xi-\xi_1)]d\xi_1 \tag{13}$$

with

$$\begin{aligned}&\bar{\xi}=0 \quad \text{for} \quad \xi<(v_2/v_1)\tau,\\ &\bar{\xi}=\xi-(v_2/v_1)\tau \quad \text{for} \quad \xi>(v_2/v_1)\tau.\end{aligned} \tag{14}$$

The limits satisfy the boundary conditions $G(\xi 0)=G_0=G(0,\tau)$, and it is assumed that the exciting light is turned on suddenly at $\tau=0$. In practice, $\xi<(v_2/v_1)\tau$ and hence the lower limit $\bar{\xi}=0$ is maintained while the term $(v_1/v_2)(\xi-\xi_1)$ can be dropped in the argument of the integrand.

The acoustic amplitude is given by

$$F(\xi,\tau)=F_0\exp(-\beta\tau)+\int_0^{\bar{\tau}}\exp(-\beta\tau_1)G(\xi+\tau_1,\ \tau-\tau_1)d\tau_1 \tag{15}$$

with

$$\bar{\tau}=\tau \quad \text{for} \quad \xi+\tau<L, \tag{16}$$

$$\bar{\tau}=L-\xi \quad \text{for} \quad \xi+\tau>L, \tag{17}$$

and the boundary conditions $F(\xi,0)=F_0$ and $F(L,\tau)=F_0\times\exp(-\beta\tau)$ have been imposed. In the experiments described here, measurements are made of essentially $G^2(L,\tau)$ as a function of L where L is the over-all reduced length of the medium. The question then arises as to whether both upper limits (16) and (17) are required in the evaluation of (15). The limit $\bar{\tau}=\tau$ will hold only for $\xi=0$ to $\xi=L-\tau$, but, if $\tau\ll L$, as is frequently the case, this limit will be a good approximation even for $\xi=L$, and (17) can be neglected.[10]

Substitution of (15) into (13) with the above limits

[10] To demonstrate that this approximation is valid, we first evaluate $G(L-\tau,\tau)$ using (13), (15), and the first upper limit of (15), $\bar{\tau}=\tau$. The contribution of the second upper limit $\bar{\tau}=L-\xi$ is estimated by introducing $G(L-\tau,\tau)$ in the integrand and assuming it to be a constant for the purpose of integrating (15) and (13) again. This iteration results in an approximate value for $G(L,\tau)$ which differs by less than 10% from $G(L-\tau,\tau)$ using values of the parameters appropriate to the present experiments. Therefore, (17) can be neglected.

of integration then yields

$$G(\xi,\tau)=G_0+F_0\xi\exp(-\beta\tau)+\int_0^{\xi}d\xi_1\int_0^{\tau}d\tau_1 G(\xi_1+\tau_1,\tau-\tau_1)\exp(-\beta\tau_1). \quad (18)$$

Equation (18) expresses that the optical amplitude at a point ξ in space and at time τ is an appropriate summation of optical waves generated between $\xi=0$ and ξ over the interval τ. Scattering events between ξ and $\xi=L$ will make no contribution to the amplitude at the point ξ according to (18).

Equation (18) can be re-expressed as

$$G(\xi,\tau)=G_0U(\xi,\tau)+F_0(\partial/\partial\tau)U(\xi,\tau), \quad (19)$$

where

$$U(\xi,\tau)=1+\int_0^{\xi}d\xi_1\int_0^{\tau}d\tau_1 U(\xi_1+\tau_1,\tau-\tau_1)\times\exp(-\beta\tau_1). \quad (20)$$

The first term of (19) corresponds to a buildup of $G(\xi,\tau)$ initiated by optical noise G_0 which is indeed very small under normal conditions. The second term, which is the one of interest, arises from a buildup initiated by a significant thermal elastic noise of amplitude F_0.

The evaluation of (20) is greatly facilitated by its iterated solution

$$U(\xi,\tau)=1+\xi\int_0^{\tau}d\tau_1\exp(-\beta\tau_1)\frac{I_1(Z)}{(Z/2)} \quad (21)$$

with I_1 being the modified Bessel function of order one of the first kind and with

$$Z=2\tau_1^{1/2}(\xi+\tau_1)^{1/2}.$$

It should be remarked that the tractable form of (21) only follows when the approximations in the limits of integration of (13) and (15) are made.

Substituting (21) into (19), averaging $G^2(\xi,\tau)$ in time for a thermal elastic noise source having random amplitude and phase at random times, and transforming to optical and acoustic power fluxes results in

$$P_2=\frac{\omega_2}{\omega_1}P_{1t}\left\{\left[\frac{\partial U(\xi,\tau)}{\partial\tau}\right]^2+2\beta\int_0^{\tau}d\tau_1\left[\frac{\partial U(\xi,\tau_1)}{\partial\tau}\right]^2\right\}. \quad (22)$$

This is the final expression which is to be evaluated numerically and compared with the experimental results. The power density of the thermal elastic noise source is given by

$$P_{1t}=(K\theta)(\Delta\Omega_1/\lambda_1^2)\Delta\nu_1, \quad (23)$$

where K is the Boltzmann constant, θ is the absolute temperature; the solid angle of the acoustic wave is $\Delta\Omega_1=(\lambda_1/\lambda_2)\Delta\Omega_2$, where $\Delta\Omega_2$ is the solid angle of the scattered light, and the bandwidth $\Delta\nu_1$ is determined in the present case by the inverse acoustic lifetime $\Delta\nu_1=(\alpha v_1/2\pi)$.

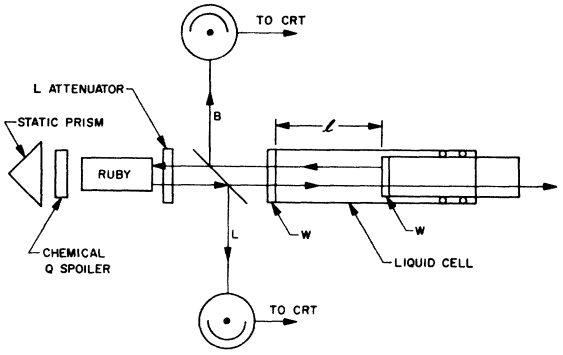

FIG. 1. Experimental arrangement for monitoring exciting light (L) and Brillouin scattered light (B). The liquid cell length l can be varied by movement of a plunger. Cell windows marked W are tilted to avoid reflections along optic axis.

The first term in (22) takes the form

$$[\partial U(\xi,\tau)/\partial\tau]^2=\xi^2\exp(-2\beta\tau)[I_1^2(Z)/(Z/2)^2]. \quad (24)$$

The second term of (22) must be integrated numerically, and in the present case will be comparable to the first term at large ξ; at small ξ, the second term will be larger than the first by at least one order of magnitude.

It will be noticed that a large growth in space and time can occur in the scattered light wave by expressing $I_1(Z)$ in its asymptotic form for large ξ, $I_1(Z)\sim\exp[2(\tau\xi)^{1/2}]/[4\pi(\tau\xi)^{1/2}]^{1/2}$, where it is assumed that $\xi\gg\tau$ as in the case of giant pulse excitation. This condition results in the approximate but convenient analytical expression, valid at large ξ,

$$P_2\cong\frac{\omega_2}{\omega_1}P_{1t}\xi^{1/2}\frac{\exp[4(\tau\xi)^{1/2}-2\beta\tau]}{4\pi\tau^{3/2}}, \quad (25)$$

where the second term in (22) has been neglected.

Of course, it is to be realized that (22) applies both to steady-state and transient-type solutions.

EXPERIMENTAL CONSIDERATIONS

Liquids are the most suitable media for the present study since the optical path length can be varied easily and made arbitrarily large. A particular liquid may be used if it exhibits a measurable intensity variation over convenient distances, 1 m or less, and preferably with a laser power of a few MW/cm^2. Thus the Pockel's coefficient should be large and, more important, the lifetime of the acoustic wave should be long. Another consideration is that the medium must not undergo stimulated Raman scattering which would compete with the stimulated Brillouin scattering, complicating the analysis unnecessarily. These requirements are satisfied by the liquid *n*-hexane which was of spectroscopic grade and was rendered dust-free by chromatographic filtering, an important precaution when long path lengths are involved. It may be of interest that we have also observed stimulated Brillouin scattering with

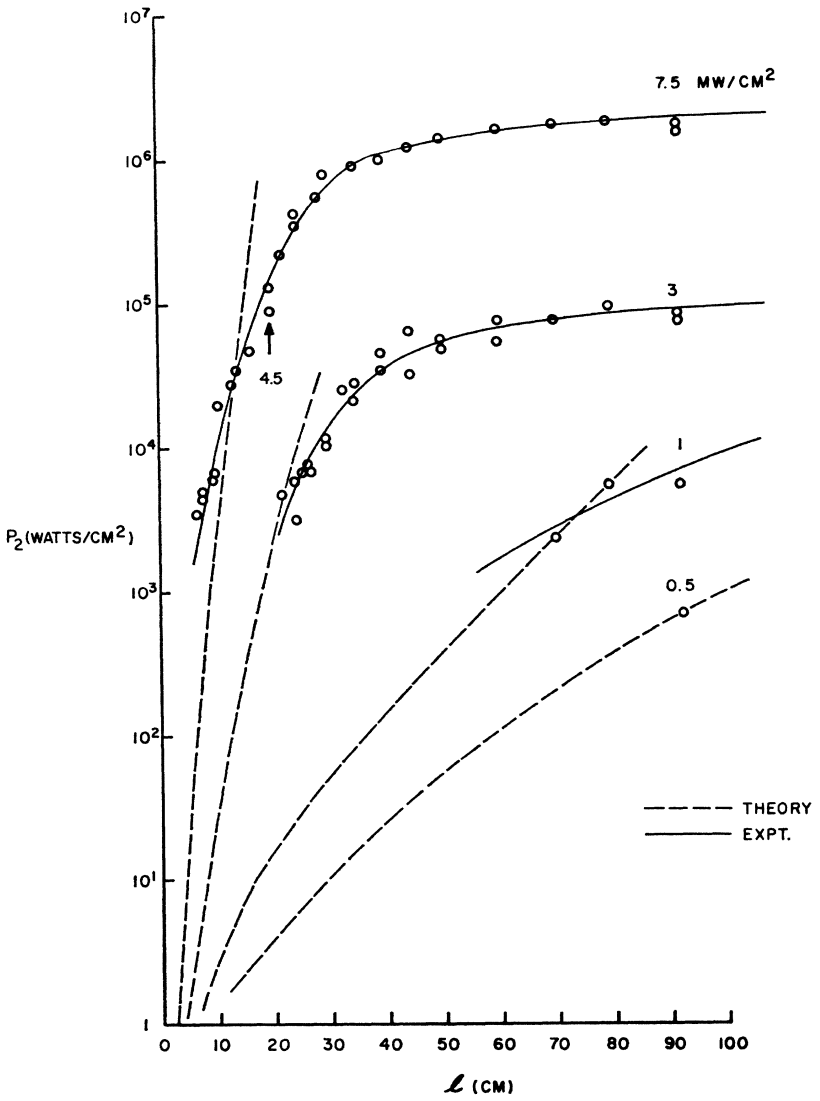

FIG. 2. Intensity of Brillouin scattered light versus optical path length. The theoretical curves (dashed lines) represent Eq. 22 of the text. The power density of the exciting beam is indicated on each graph.

benzene, but not with water, using a collimated beam for excitation.

A schematic of the experimental arrangement is shown in Fig. 1 and includes a liquid cell whose length l can be varied from 0 to 92 cm by means of a plunger. The cell is in close proximity to the laser head in order to utilize fully the available radiation density of the laser.

The exciting source is a giant pulse ruby laser which has the following characteristics: a beam divergence of $\sim 0.3°$ angular diameter, linear polarization, a single longitudinal mode output at ~ 6940 Å, 30-nsec duration, and a peak power of ~ 3 MW/cm^2. The chemical Q spoiler[11] contains vanadium phthalocyanine in a nitrobenzene solvent and has an optical density of 0.15 at 6940 Å. Corning filters are used to attenuate the laser output to the desired level.

The backscattered light is found to converge in a cone of $\sim 0.3°$ angular diameter and exhibits a negative frequency shift (Stokes scattering) from the incident light of 0.14 cm^{-1} as determined by interferometry.

The intensity of incident and backscattered light are diodes equipped with optical filters and with a Tektronix 555 dual beam oscilloscope containing type-L preamplifiers.

We draw attention to the fact that the ruby laser and the Brillouin scattering medium are coupled to each other. Backscattered Brillouin light, on retracing the path of the incident laser beam, enters the laser cavity whereupon it is amplified[4,12] and reflected back into the medium for a subsequent Stokes scattering.[13] Because of this feedback, the exciting light intensity will in general be dependent on the intensity of the scattered light. Thus, in the uppermost curve of Fig. 2, the exciting

[11] P. P. Sorokin, J. J. Luzzi, J. R. Lankard, and G. D. Pettit, IBM J. Res. Develop. 8, 182 (1964).

[12] R. G. Brewer, Appl. Phys. Letters 5, 127 (1964).

[13] For further details on these iterative scatterings see R. G. Brewer and D. C. Shapero, Conference on the Physics of Quantum Electronics, 1965 (to be published).

light power is 7.5 MW/cm² when the Brillouin conversion is 25% and levels off to 4.5 MW/cm² when the conversion is 3% or less. Obviously, when the feedback is small, the excitation is essentially constant as is true in the other experimental curves of Fig. 2.

GROWTH OF THE SCATTERED LIGHT WAVE

The intensity of backscattered Brillouin light versus optical path is given in Fig. 2 for n-hexane at 23°C. The solid curves are experimental, the dashed curves are theoretical, and the number on each curve designates the power level of excitation in MW/cm².

We first notice that a pronounced saturation is evident in the experimental points whenever the path length and the exciting power are sufficiently large. Since 3% or more of the exciting light is converted into Brillouin light under these conditions, this result suggests that saturation is caused simply by the depletion of exciting light with distance.

Below the saturation region, the experimental intensities drop off smoothly, covering $2\frac{1}{2}$ orders of magnitude in the 7.5-MW/cm² curve, as the path length is decreased. It is to be noted that there is no critical distance which can be associated with an instability or threshold condition. Therefore, within the range of these experimental points, there is no evidence for an unstable backward-wave amplifier, as described by Kroll[14] or by Chiao, Garmire, and Townes.[1] However, a possible threshold at shorter l's cannot be excluded.

Next consider the backward-wave amplifier-type solution as proposed by Bloembergen and Shen. For this steady-state case, the optical wave grows exponentially in space as e^{gl} where g is the gain constant. The experimental points do not follow such a dependence, however, as can be seen in the semilog plots of Fig. 2 which exhibit significant curvature well beyond the region of saturation. Even if a linear curve were forced on the data, its extrapolation to $l=0$ would account for an intensity variation of only four orders of magnitude or less. This feature is particularly noteworthy in the 1-MW/cm² curve. For spontaneously scattered light to build up to the saturation level, the amplification must exceed roughly eight orders of magnitude. Thus, this description does not readily account for the present observations.

To determine the applicability of Kroll's transient solution, we evaluate (22) numerically with the aid of an IBM-7094 computer assuming values for the Pockel's coefficient p and the acoustic damping constant α. By varying these parameters over the range $p=0.36$ to 1.00 and $\alpha=100$ to 10 000 cm^{-1}, a family of P_2-versus-l curves can be generated. The curves which agree most closely with the experimental points, as shown in Fig. 2, are confined to the values

$$p=0.5\pm0.1\,,\qquad \alpha=900\pm400\ \text{cm}^{-1}\,, \tag{26}$$

where the following constants[15] for n-hexane have been used: $\rho_{20°}=0.6595$ g/cm³; $v_1=1.06\times10^5$ cm/sec at 4 Gc/sec; $n=1.37$ at 6560 Å and $X=0.1/P^{1/2}$ cm with laser power P in MW/cm². There should, of course, be only one value of α and p necessary for describing all of the experimental curves. The distribution in α and p as indicated by (26), while not excessively broad, does reflect a certain degree of error in the measurements, in the theory, or both. Some of this uncertainty undoubtedly arises from an inability to measure accurately the power of the exciting light which enters in a sensitive way in the result, as do α and p.

The theoretical curves certainly account for the rapid buildup, which can occur at high exciting powers (7.5-MW/cm² curve) within the first few cm of the liquid and for the absence of a threshold condition. Also, the variation of the gain curve with exciting power is predicted but the rather large curvature (semilog plot) of the experimental graphs is not. No attempt has been made here to include optical loss, and clearly a large curvature will result as the saturation condition is approached, i.e., when a few precent or more of the exciting light is converted to Brillouin light. It is significant, however, that the experimental curves exhibit curvature well below the saturation region. Other optical-loss mechanisms, such as thermal currents or residual dust, might be contributing in this region. In addition, the possibility of a multi-mode laser source could contribute to curvature.[8] This is a point which should receive further investigation.

The magnitude of the Pockel's coefficient, 0.5 as indicated in (26), tends to support Kroll's theory since this value is in reasonable agreement with an estimate based on the Clausius-Mossotti relation, $p\sim\rho(\partial\epsilon/\partial\rho)\sim\frac{1}{3}(n^2+2)(n^2-1)$ or $p=1$. It should be noted, however, that an error of 2 in the laser power would modify p by 40% according to (10), and this is perhaps a more reasonable uncertainty in p.

The damping constant $\alpha=900$ cm^{-1} corresponds to an acoustic lifetime of 10^{-8} sec. This is consistent with the technique of time-resolved Brillouin orders,[16] which yields a value of less than 2×10^{-8} sec. An extrapolation of the ultrasonic damping constant,[17] assuming α is proportional to the square of the frequency, gives 10^{-9} sec. Obviously, an independent determination of α would be helpful as a check on the present results.

Thus, of the existing theories the transient solution of Kroll appears to be the most suitable for explaining at least the course features of the experimental gain curve of n-hexane. Presumably, other liquids and solids would behave similarly at room temperature since the

[14] See p. 38 and also footnote 8 of Ref. 5.

[15] ρ and n are taken from *Landolt-Bornstein Zahlenwerte und Funktionen* (Springer-Verlag, Berlin 1960, 1962), Vol. II, Part 2, p. 196 and Part 8, pp. 5–575. v_1 is obtained from the measured Brillouin frequency shift using Eq. (3) of the text.

[16] R. G. Brewer, Appl. Phys. Letters **6**, 165 (1965) and Ref. 13.

[17] K. F. Herzfeld and T. A. Litovitz, *Absorption and Dispersion of Ultrasonic Waves* (Academic Press Inc., New York, 1959), p. 357.

nature of the buildup will be determined primarily by the condition $\tau \ll 4\xi/\beta^2$ [see Eq. (25)], which will usually hold for giant pulse excitation. Whether the inclusion of loss in the optical component and a consideration of multimode behavior will improve the agreement with experiment remains to be seen. Also, the extension of the experimental curves to shorter path lengths would be desirable if the separation of Brillouin light from stray exciting light can be achieved.

Finally, the present technique of collimated excitation would be useful in testing current theories of the stimulated Raman scattering.[18]

ACOUSTIC BURSTS AND MATERIAL FRACTURE

In this section we consider whether laser excitation leading to fracture in solids[2,5] and cavitation in liquids[3] results from the acoustic wave produced in the Brillouin process or from other mechanisms.

Prevously we reported[3] that focused giant pulse excitation in liquids produces rather spectacular effects in addition to stimulated Brillouin emission. These include acoustic pulses which reflect off the walls of the liquid cell, audible bursts, cavitating bubbles which emit white light, and also the generation of second harmonic acoustic waves at $\sim$10 Gc/sec.[16] It was shown that the acoustic energy imparted to the windows of the liquid cell was at least 900 ergs whereas the maximum acoustic energy generated in the Brillouin process could have been only 50 ergs, assuming 100% conversion. This evidence and the fact that stimulated Brillouin scattering is observed at lower powers without the above effects indicates that more than one process is involved. Furthermore, the observation of dielectric breakdown suggests a mechanism involving the growth of a plasma.

On the other hand, we find that material fracture can take place without stimulated Brillouin emission when the crystals CaF_2, MgO, and NaCl are subjected to focused laser radiation. Clearly the mechanism operating here cannot be ignored when intense Brillouin scattering does occur. This damage might also result from dielectric breakdown and a rapidly expanding plasma.[19]

In the present study with a collimated beam of exciting light, occasionally a very striking acoustic burst, having a characteristic audible snap, is heard whenever the stimulated Brillouin process saturates. For example, with an exciting power of 3 MW/cm^2, the *n*-hexane column must be longer than 35 cm for this effect to occur. Since the electric field of the exciting light is only $\sim 5\times 10^4$ V/cm, dielectric breakdown is not expected, and no visible evidence was found as in the case of a focused beam. Indeed, if breakdown occurred, there should be little dependence on path length. The correlation of these bursts with the path-length dependence of Brillouin backscattering, as given in Fig. 2, indicates that they originate in the Brillouin acoustic wave of frequency 4 Gc/sec. The observation of audible frequency components suggests an intense shock wave with the eventual conversion of hypersonic to lower frequency waves. While cavitation might take place under these conditions, more refined techniques will be needed for its detection.

Another effect which occurs in the collimated excitation of benzene, hexane, carbon tetrachloride, and presumably in other liquids is the liberation of a uniform stream of stable bubbles along the entire optical path (up to 92 cm). These bubbles are not obviously associated with the above acoustic bursts and are not formed from gaseous decomposition products of the liquid, a possibility which might be expected if dielectric breakdown had occurred. For example, in CCl_4 the evolved gas does not contain chlorine. Rather, these bubbles are composed of air, and their formation can be enhanced or prevented by controlling the amount dissolved. In addition, there is a power and path-length dependence which is not necessarily associated with stimulated Brillouin or Raman scattering. This is indicated in CCl_4 which, within our limits of detection, shows neither stimulated Brillouin nor Raman scattering, but does exhibit bubbles, when the collimated excitation is a few MW/cm^2 and the liquid path length is 45 cm or more.

If bubble formation results from a weak stimulated Brillouin process, it is peculiar that the bubbles are not concentrated near the entrance window where the acoustic amplitude would be a maximum. Furthermore, an acoustic wave of high frequency would probably be damped in a few wavelengths and could not be expected to exert an influence over several cm. Also, local heating effects should be negligible for a 0.25 J collimated source. This evidence might suggest that another process is occurring such as a high-frequency Kerr effect involving molecular orientation or an electrostrictive effect of the type discussed recently in connection with the self-trapping of optical beams.[20] Additional studies will be needed to clarify this point.

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[18] E. Garmire, F. Pandarese, and C. H. Townes, Phys. Rev. Letters **11**, 160 (1963); R. W. Terhune, Solid State Design **4**, 38 (1963); Ref. 6; C. L. Tang and T. F. Deutsch, Phys. Rev. **138**, A1 (1965). Measurements of this kind were performed recently by P. Lallemand and N. Bloembergen, Appl. Phys. Letters **6**, 210 and 212 (1965).

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[20] R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Letters **13**, 479 (1964).