# STIMULATED BRILLOUIN SCATTERING IN LIQUIDS

Richard G. Brewer and Klaus E. Rieckhoff
IBM Research Laboratory, San Jose, California
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We have observed that when a liquid is in the path of a focused beam of a giant-pulse ruby laser there results stimulated Brillouin scattering[1,2] and, in addition, an intense acoustic wave, dielectric breakdown, and cavitation. This acoustic or shock wave is too intense to be confused with the coherent wave of frequency ~6 $\times 10^9$ cps produced in the stimulated Brillouin scattering process itself and very likely originates in the resulting plasma, causing cavitation in liquids and fracture in solids. In addition, we observe that stimulated Brillouin scattering in liquids consists of several orders of Stokes and anti-Stokes light, which behavior contrasts with the single Stokes emission seen in quartz and sapphire by Chiao, Townes, and Stoicheff,[2] and whose origin is not explained by existing theories.

In this work, back-scattered Brillouin light is identified interferometrically, and traveling acoustic waves are detected ballistically and also by an optical method to be described. All measurements were performed using giant ruby-laser pulses, which when unfocused lie in the power range 10 to 20 MW/cm$^2$. Fabry-Perot interferograms (etalon separation equal to 1 cm) of the laser source alone reveal one sharp ring system with each line exhibiting a full width at half-maximum of ~0.01 cm$^{-1}$. A 5-cm focal-length lens was used to concentrate the laser light in a sturdy rectangular glass cell of 10 cm length containing the liquid.

Fabry-Perot interferograms of the back-scattered light of the liquid were obtained using a 45° glass beam splitter between the laser and the 5-cm lens. Back-scattered light from the beam splitter was directed onto a ground glass placed in front of a Fabry-Perot interferometer whose transmitted light came into focus, by a 50-cm focal-length lens, on a I-N plate. The laser rings were intensified by placing a mirror to one side of the beam splitter and facing the interferometer. Figure 1 is an interferogram of stimulated Brillouin scattering of water. The etalon spacing is 0.30 cm, corresponding to an interorder separation of 1.7 cm$^{-1}$. If the water is removed from its cell, only a single ring system appears as a result of the laser light. The most intense ring in each Fabry-Perot order is the laser emission which separates three orders of Stokes (inner rings) and two orders of anti-Stokes (outer rings). The full width at half-maximum of the Stokes lines is ~0.1 cm$^{-1}$ and the anti-Stokes lines ~0.03 cm$^{-1}$ when the laser width is ~0.01 cm$^{-1}$. These values are to be compared with the widths for stimulated vibrational Raman scattering which are of similar magnitude.[3] The existence of a threshold, the narrow linewidths, the high directionality of this light, and the fact that the intensity of this light is comparable to the laser light itself, shows that the observed Brillouin scattering is a stimulated emission process.

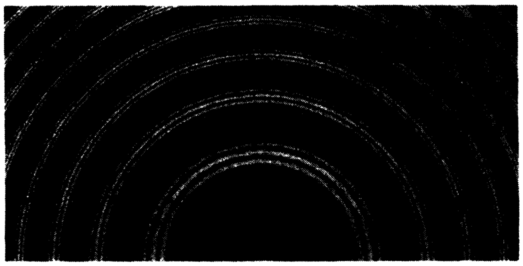
FIG. 1. Fabry-Perot interferogram of ruby laser and back-scattered Brillouin light of liquid water. Each Fabry-Perot order consists of three Stokes, two anti-Stokes, and a laser ring, which is the most intense. The etalon separation is 0.30 cm.

The experimental and calculated frequency shifts for water and benzene are given in Table I. The uncertainty in the experimental values is about 5%. The calculated fundamental acoustic frequency $\nu_S$ is given by the Brillouin expression

$$\nu_S = 2\nu_0 (nv/c) \sin(\theta/2), \tag{1}$$

$n$ being the index of refraction of the liquid, $v$ the velocity of sound in the liquid, and $\theta$ the angle between the incident laser beam and the scattered light. The agreement between the observed and calculated frequency shifts is seen to be quite satisfactory in spite of the fact that the dispersion of velocity with frequency has been ignored. Earlier measurements by Gross and

Table I. Measured and calculated frequencies of Brillouin-scattered light.

| Liquid | Brillouin order | | Observed shift[a] | Calculated shift[a, b] |
|---|---|---|---|---|
| | Stokes | Anti-Stokes | ($cm^{-1}$) | ($cm^{-1}$) |
| Water | 3 | | −0.59 | −0.57 |
| | 2 | | −0.40 | −0.38 |
| | 1 | | −0.21 | −0.19 |
| | | 1 | +0.20 | +0.19 |
| | | 2 | +0.42 | +0.38 |
| Benzene | 1 | | −0.22 | −0.19 |
| | | 1 | +0.19 | +0.19 |

[a] Relative to the ruby-laser frequency. The temperature is ~23°C.

[b] The index of refraction and the velocity of sound are taken from Landolt-Börnstein, Zahlenwerte und Funktionen (Springer-Verlag, Berlin, 1962), Vol. II, Pt. 8, p. 562, and Vol. IV, Pt. 1, p. 818, respectively.

others[4] are also in reasonable agreement with these results. The result that water produces more orders of Stokes and anti-Stokes light than does benzene is unexpected, since their efficiency is reversed in the ordinary Brillouin scattering.[4] Additional studies of other molecules will be required to determine if this result is a general one.

The generation of stimulated Brillouin scattering appears to be analogous to stimulated vibrational Raman scattering,[5] although certain differences might be expected. In particular, for maximum gain the laser ($\vec{k}_0$) will generate back-scattered Brillouin-Stokes radiation ($\vec{k}_{-1}$) and a forward-scattered acoustic wave ($\vec{k}_S$), of frequency ~$6\times10^9$ cps for liquids, in accordance with energy conservation and the momentum matching condition $\vec{k}_0 = \vec{k}_{-1} + \vec{k}_S$. The present observation of a back-scattered anti-Stokes emission, on the other hand, obeying $\vec{k}_0 + \vec{k}_S = \vec{k}_1$, requires the presence of an intense coherent acoustic wave traveling in the backward direction and raises the question of how such a wave can be produced. A tentative explanation is that the intense forward-scattered acoustic wave is reflected off a considerably more energetic thermal wave, as in the four phonon interactions postulated for some solids.[6] This view is consistent also with the absence of stimulated anti-Stokes emission in quartz and sapphire and the contention that a three-phonon mechanism, which cannot produce a back-scattered acoustic wave of the correct energy (~$6\times10^9$ cps), dominates hypersonic absorption in these substances.[7] Additional experiments are being conducted now to test whether the above mechanism applies.

Evidence for laser-induced plasma formation and cavitation in a liquid is given in Fig. 2, as indicated by a stream of small bubbles which emit intense white light in the vicinity of the focal region. Laser-induced plasma formation in gases[8] and solids[9] has been reported previously with up to 90 % of the exciting laser radiation being converted into thermal energy. We believe that once the plasma is initiated further absorption of laser light results in a shock wave or an intense acoustic wave.

Observation of these acoustic waves was achieved here by monitoring the light of a second source, a cw helium-neon laser operating at 6328 Å, which traverses the liquid at an angle of about 10° with respect to the ruby-laser beam. The passage of the cw light through the medium is interrupted periodically by the acoustic wave originating with the giant pulse (30 nsec duration) and persisting long after the giant pulse has terminated by repeated reflections off the walls of the cell containing the liquid. The acoustic wave causes the liquid to behave as an intermittent lens (or possible as an intermittent diffraction grating as in the Debye-Sears[10] effect)

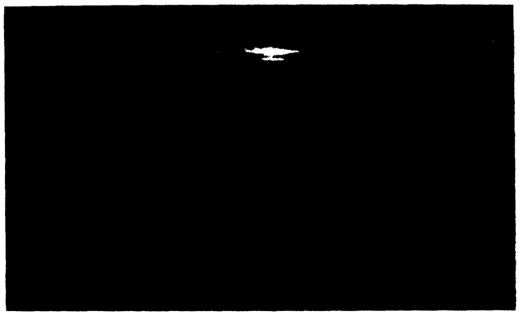

FIG. 2. Dielectric breakdown and cavitation in water as observed at right angles to the incident laser beam and through a filter which rejects scattered laser light.

which produces a train of optical pulses possessing a repetition interval of about 20 μsec (approximately liquid-cell dimensions/velocity of sound) and a pulse width of 2 μsec. A question arises as to the upper limit of the frequency of these pulses since the high frequency components (~$10^9$ cps) will be attenuated over short distances (~$2\times10^4$ dB/cm in benzene),[11] as will the acoustic Brillouin waves. DeMaria[12] has also observed acoustic waves in liquids by an independent method.

A lower limit to the energy stored in this acoustic wave is 900 ergs, this value being derived solely from the kinetic energy delivered to a pendulum whose lower end is in loose contact with the window of a cell containing the liquid. Thus, the energy stored in this acoustic wave is considerably higher than the maximum value which can be achieved in stimulated Brillouin scattering, namely ($0.2\ \text{cm}^{-1}/14\,400\ \text{cm}^{-1} = 10^{-5}$ or ~50 ergs). This result strongly suggests that plasma formation rather than stimulated Brillouin scattering is the dominant process leading to fracture in solids[2] and cavitation in liquids.

Liquids excited by a giant pulse should be attractive as a general source of intense hypersonic waves at frequencies near $6\times10^9$ cps. While liquids under these conditions cavitate, they are not subject to fracture as are the solids which have been studied thus far, and hence possess the advantage of reproducibility as well as higher homogeneity.

It would appear that several possibilities exist for future studies in this area. For example, stimulated Brillouin scattering and the accompanying hypersonic waves should be observable in supercooled liquids at low temperatures with the advantage of enhanced acoustic waves due to reduced attenuation and at frequencies beyond $10^{10}$ cps. Also, hypersonic waves generated at harmonic frequencies of the fundamental might be detectable and would allow an upward extension in acoustic frequency.

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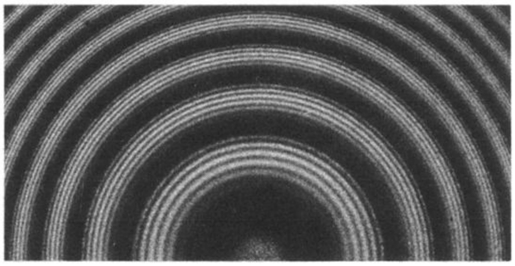

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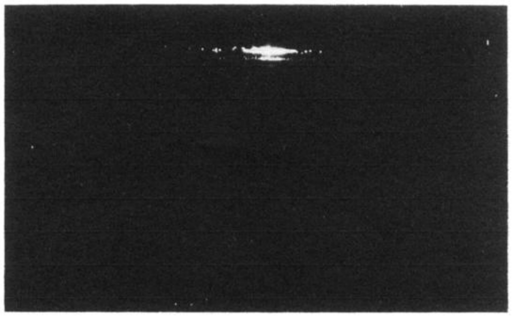

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