UNPUBLISHED PRELIMINARY DATA

STIMULATED BRILLOUIN SCATTERING IN LIQUIDS *

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Stimulated Brillouin scattering of intense laser light, with a build-up of coherent hypersonic waves, has been observed in a number of liquids in an arrangement which allows multiple Brillouin scattering processes and rather precise measurement of the velocity of hypersonic waves. Stimulated Brillouin scattering, which has already been reported in solids[1], can be considered parametric generation of an acoustic wave and a scattered light wave from an initial light wave. The non-linear coupling of the three waves is typically electrostrictive. Much of the theory involved has already been discussed[2][3] In solids, a single scattering of the incident light was observed, with a shift to lower frequencies equal to the frequency of the acoustic wave. In the present experiment with liquids, however, as many as eight orders of successively scattered light waves appear. Each order is generated backward from the incident wave and finds its way back to the laser cavity where it is amplified. This component again enters the liquid giving rise to its own Brillouin scattering, which appears as the next order.

A giant pulse ruby laser provided the incident light in an experimental arrangement shown schematically in Figure 1. A glass flat with parallel sides was introduced into the beam as an additional optical resonator in

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order to separate longitudinal modes and produced a single mode with a frequency spread less than .04 cm^{-1}. Liquids were placed in a cell at or near the focal point of a lens and the frequencies of light generated near 6943 Å were studied with a Fabry-Perot interferometer. The interferometer was placed at sites A, B, or C. Figure 2 shows a typical interferometer pattern. Stimulated Brillouin scattering was also observed with the ruby laser under normal (not Q-switched) operation, but the giant pulse system was used for measurements of frequency shifts because of its high spectral purity.

Photographs of the Fabry-Perot rings observed at sites A and C were similar, but with the total intensity at C perhaps a factor of five less than at A. Both the stray laser light and the Brillouin components were too weak to be detected through the Fabry-Perot placed at B. This evidence, along with the multiple orders of Brillouin shifts, show that the Brillouin components are being amplified in the ruby, and follow the path indicated by the dashed line in Figure 1. The width of the dashes represents the relative intensity of the Brillouin component as it is amplified. The intensity at B is too weak to be detected, while after amplification the signal at A may be large. Once the Brillouin component is amplified, it may be as strong as the original laser frequency and in fact acts somewhat like light from another mode of the laser, re-entering the liquid and causing another Brillouin scattering. This process can occur a number of times, since the frequency shift for Brillouin scattering in liquids is around 0.2 cm^{-1}. Thus a number of

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orders are well within the ruby linewidth and may be amplified. This is not the case for the solids previously reported,[1] where the shifts were around 1 cm^{-1}. It is to be emphasized that the reiterative effect in this particular arrangement is different from that normally causing the higher orders of stimulated Raman scattering. As long as the liquid is outside a cavity (and the backward scattered light wave is weak) there can be no "anti-Stokes" wave, since the required backward-going acoustic wave is not present. With sufficiently intense effects, one might expect multiple-order stimulated Brillouin scattering by higher order processes within the liquid and without further amplification of the first-order wave. This has not been observed. For, if the first order stimulated Brillouin scattering occurs at 180° to the incident light, the second order would occur at 0°, and one would expect alternating orders of Brillouin components at either A or C, which do not occur.

One might also wonder if the Brillouin components amplified in the ruby are from spontaneous Brillouin back-scattering, rather than stimulated Brillouin scattering. If such were the case, Brillouin shifts to higher frequencies would occur with the same intensity as shifts to lower frequencies, which is not observed. Furthermore, the large variation in relative intensity of the Brillouin components and the original laser light with a rather small variation of input laser power shows conclusively that stimulated emission dominates.

There is a threshold power density below which the Brillouin components

were not observed at all. This was not necessarily identical with the threshold for initiation of amplification of the Brillouin components, since the sensitivity of detection was not high. In the case of carbon disulfide, stimulated Brillouin scattering was observed when the focal point of a 1 7 cm focal length lens was 8 cm beyond the cell. This gave a threshold of 30 M watts/cm, the lowest observed in any of the liquids studied. The threshold in benzene was 1200 M watts/cm^2, and most of the liquids had comparable thresholds. Surprisingly, nitrobenzene had a much higher threshold than any other liquid studied. It is nearly as strong a spontaneous Brillouin scatterer as carbon disulfide and has almost 100 times less acoustic absorption in the ultrasonic region where it has been measured. The high threshold indicates that nitrobenzene may be unusually lossy to kilomegacycle acoustic waves. It is interesting to note that many weak Brillouin scatterers had thresholds comparable with some of the strongest scatterers, presumably because of their lower acoustic losses. Water is a notable example. Stimulated Brillouin scattering should prove valuable in the study of such weak scatterers where it is difficult to obtain sufficient spontaneous signal.

In carbon disulfide, nitrobenzene, toluene, benzene and acetone, stimulated Raman scattering occurred along with the stimulated Brillouin scattering. The Raman threshold in these cases is appreciably lower than that for Brillouin scattering. For water, carbon tetrachloride and methanol, stimulated Raman scattering did not occur at the experimental power levels.

The presence of simultaneous Raman and Brillouin scattering did not appear to markedly affect the Brillouin scattering.

From the measured frequency shift $\Delta\nu$, the hypersonic acoustic velocity v of the liquids can be calculated from the equation $\Delta\nu = 2\,\nu_0\,\frac{v}{c}\,n$, where ν_0 is the frequency of the incident light, and n is the refractive index. Table 1 lists the frequency shifts measured at about 22°C, the calculated sound velocities, and results of measurements of spontaneous Brillouin shifts given in Herzfeld and Litovitz.[4] The values are seen to agree within stated experimental errors.

Stimulated Brillouin scattering is a useful method of measuring hypersonic velocities to a high degree of accuracy. The directionality of the amplified Brillouin components, the many orders, the line sharpening of stimulated Brillouin scattering, and the sharp frequency of the single mode laser, all contribute to an inherent accuracy which should allow measurement of velocities about two orders of magnitude better than the rough measurements made here. The most accurate experimental means for determining the frequency shift would probably be observation of the microwave beats from a photo cathode mixing the laser and the Brillouin components. This method would be especially convenient since all the components are of comparable intensity and in a single directional beam.

As in the case of stimulated Brillouin scattering in solids, intense hypersonic acoustic waves are generated in the liquids. Contrary to the

case of crystals, neither the liquid nor the cell in which it is contained are damaged by the stimulated Brillouin effect. This simplifies observations, and detailed studies can be made of the acoustic properties of the liquids. Probably Brillouin components due to solids with sufficiently small sound velocities can also be amplified by this method. It may thus allow convenient excitation of ultrasonics and stimulated Brillouin effects in crystals without their being fractured.

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References

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TABLE 1

Brillouin shifts, calculated acoustic velocities, and previously measured velocities.

Liquid	Brillouin Shift cm^{-1}	Calculated sound velocity m/sec	Previous * results m/sec
CCl_4	.141	1007 ± 7	1040 ± 27
Methanol	.139	1100 ± 11	
Acetone	.153	1174 ± 7	1190 ± 40
CS_2	$.192_5$	1242 ± 6	1265 ± 22
H_2O	$.188_5$	1471 ± 8	1509 ± 25
Aniline	$.257_5$	1699 ± 8	

* acoustic velocities given in Reference 4, which are calculated from measurements of spontaneous Brillouin scattering.

FIGURE 1

Experimental arrangement, showing path of stimulated Brillouin scattering. A Fabry-Perot interferometer was placed at sites A, B, and C.

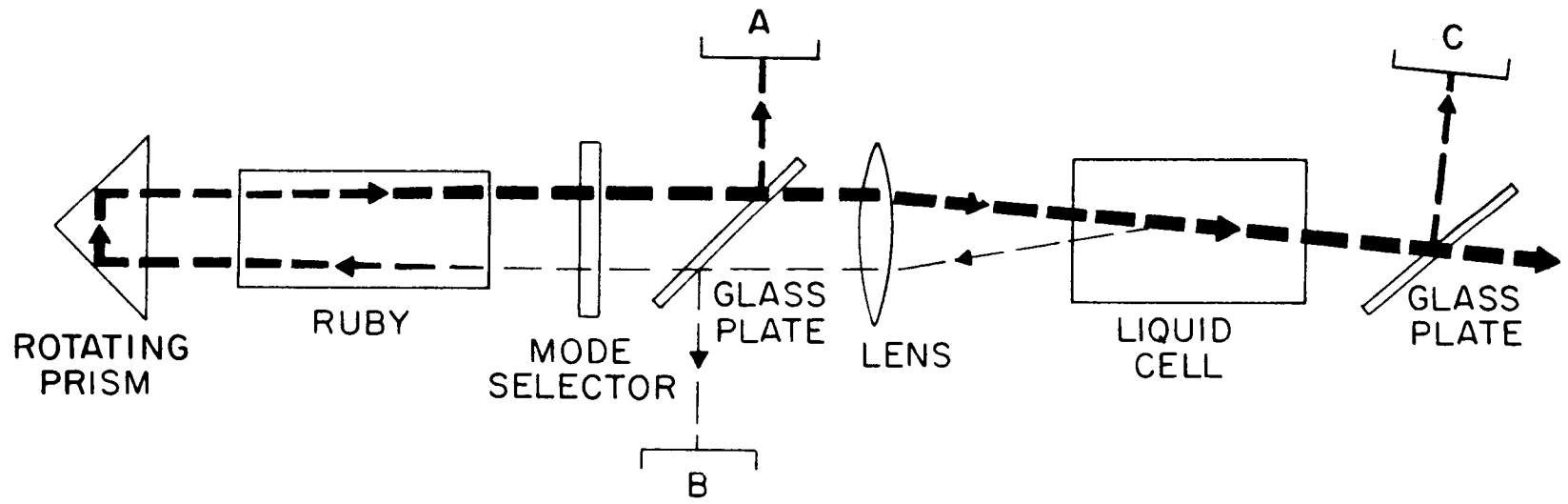
A
C
ROTATING
PRISM
RUBY
MODE
SELECTOR
GLASS
PLATE
LENS
LIQUID
CELL
GLASS
PLATE
B

FIGURE 2

Fabry-Perot Interferograms from site A with water in the liquid cell.

Left: Single frequency of laser with intensity below threshold for stimulated Brillouin scattering.

Right: Above threshold, with three Brillouin components from water in addition to the original laser frequency. Interorder spacing is .701 cm^{-1}.