point phonon splitting determined from the long-wavelength approximation to that determined from the short-wavelength approximation and obtained a value of four. Thus, both approximations yield the same order of magnitude for the ground-state splitting when used in conjunction with the Debye spectrum.

III. CONCLUSIONS

We have demonstrated that, even when admixtures arising from spin-orbit coupling are included, the orbit-lattice interaction yields a ground-state splitting for $CaF_2:Gd^{3+}$ which is orders of magnitude smaller than the measured value, gives the incorrect order of the levels, and increases instead of decreases with temperature. Consequently, in concurrence with our initial findings[1] on this subject, it is concluded that the orbit-lattice interaction, without configuration mixing, may be eliminated as a possible mechanism which is responsible for the splitting of the ground level of S-state ions.

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Transverse Stimulated Emission in Liquids*

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Stimulated Brillouin scattering transverse to the laser beam, external to the laser cavity and free from frequency-dependent feedback, has been studied using a specially designed laser. The laser beam is focused by a cylindrical lens so that the direction of maximum gain is transverse to the beam. The hypersonic velocities of several liquids have been measured at high power levels without frequency-pulling effects. Transverse filaments have been observed, and an approximate measurement of the power contained in a single filament has been obtained. A stimulated anti-Stokes scattering in benzene has also been observed, which has a 200-MHz frequency shift. This scattering is tentatively identified as stimulated thermal Rayleigh scattering.

I. INTRODUCTION

WE have studied stimulated Brillouin scattering transverse to the laser beam and external to the laser cavity. With this arrangment no *frequency-dependent* feedback occurred in either the longitudinal or transverse direction. This experiment has been made possible by the design and construction of a mode-selected 350-MW ruby laser whose spectral width is approximately 300 MHz. With this device we have been able to observe stimulated Raman and stimulated Brillouin scattering transverse to the laser beam in almost any liquid without any external resonator. Thus, it has been possible to measure the hypersonic velocities of the liquids at high power levels without the frequency pulling usually involved. Because of the good separation of the incident and scattered light, we have been able to observe light shifted by less than 0.01 cm^{-1} from the laser frequency. It has also been possible to study the spatial distribution of light intensity in the scattering region.

Several studies of stimulated scattering in liquids have been carried out in which the direction of the stimulated scattering was not parallel to the laser beam.[1–4] Dennis and Tannenwald[1] used a transverse resonator external to the laser cavity to study stimulated Raman scattering emitted at 90° to the laser beam. Dennis[2] observed stimulated Brillouin scattering in a transverse resonator, but the observed frequency shift was somewhat ambiguous because of the multimode nature of the laser source. Takuma and Jennings[3] studied stimulated Brillouin scattering in an off-axis external resonator at small angles to the laser beam. In the latter experiment two frequency shifts were observed, corresponding to the two angles from the forward laser beam direction. Pine[4] has studied stimulated Brillouin scattering from many different liquids using an external transverse resonator. His experiments were performed with a laser with sufficiently good mode selection to allow unambiguous determination of the Brillouin shift and threshold phenomena. Pine pointed out the difficulty with frequency-pulling effects caused by the external resonator. Frequency pulling by the laser resonator has also been noted[5,6] in longitudinal

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[1] J. Dennis and P. Tannenwald, Appl. Phys. Letters **5**, 58 (1964).
[2] J. Dennis, Lincoln Laboratory, Solid State Research Report No. ESD-TDR-65-31, 1964 (unpublished).
[3] H. Takuma and D. Jennings, Appl. Phys. Letters **5**, 239 (1964).
[4] A. Pine, Phys. Rev. **149**, 113 (1966).
[5] R. G. Brewer, Appl. Phys. Letters **9**, 51 (1966).
[6] T. A. Wiggins, R. V. Wick, D. H. Rank, and A. H. Guenther, Appl. Opt. **4**, 1203 (1965).

experiments, and it complicates accurate measurements of the hypersonic velocity in a stimulated scattering experiment.

In addition, the lack of stimulated anti-Stokes Brillouin scattering in the transverse geometry has been reported[4] even though acoustic waves and pump source propagate in antiparallel directions. Stimulated anti-Stokes scattering has been reported by Burlefinger and Puell[7] in the longitudinal direction from a liquid cell placed in the laser cavity. Their paper did not, however, indicate how an unambiguous determination was made as to which of the three lines observed was the laser. It is, therefore, possible that they observed the laser plus two successive Stokes-Brillouin shifts. Wick *et al.*[8] have clearly demonstrated stimulated anti-Stokes scattering of multiple orders in the longitudinal direction, external to the laser cavity although the exact mechanism is somewhat unclear.

II. EXPERIMENTAL

The equipment used in the transverse stimulated scattering experiment is shown in Fig. 1. The laser consists of a sapphire etalon output mirror mode selector,[9] a ruby rod 6 in. long, $\frac{1}{2}$ in. diam, with ends cut at the Brewster angle, a cryptocyanine Q switch, a reflection etalon mode selector,[10] and a Brewster-angle roof prism. The dye cell and reflection mode selector are combined, such that the two surfaces of the reflection mode selector are the correctly aligned

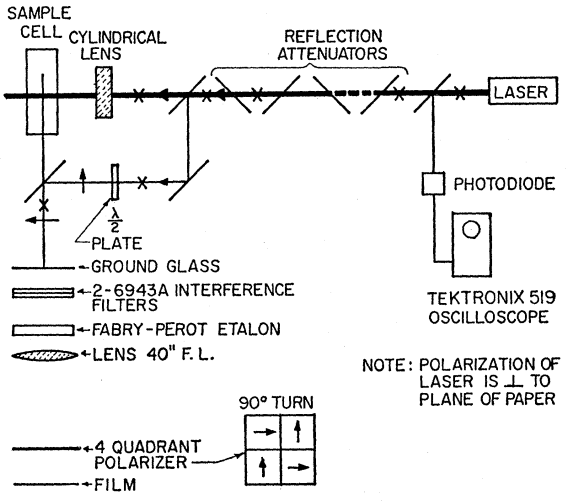


FIG. 1. Experimental setup used to study transverse stimulated scattering.

ends of the dye cell. This reduces the number of optical surfaces subject to degradation. The cryptocyanine-methanol solution flows continuously through the dye cell, eliminating the degradation of the dye in the Q switch, which otherwise occurs from pulse to pulse. The laser is usually operated at a peak power of 300 MW with a pulsewidth of 8 nsec. Changes in laser intensity are accomplished by a series of reflection attenuators so that the operating characteristics of the laser do not have to be altered. This provides greater pulse-to-pulse reproducibility of beam intensity profile and spectral width. Under these conditions it is possible to maintain a spectral width of less than 300 MHz and to ensure operation in a single longitudinal mode.

A cylindrical lens of 10–30-cm focal length is used to focus the beam into a cell containing the liquid under study. The ends of the sample cell are not parallel, which prevents them from operating as an inadvertent resonator. In addition, neither end is perpendicular to the focal line. With this equipment it is possible to observe stimulated Brillouin scattering and stimulated Raman scattering transverse to the laser beam along the line focal region in almost any liquid. The laser was separated 2.5 m from the liquid cell. This provided a 14-nsec delay between the laser pulse and any back-scattered radiation at the monitor. It was thus possible to observe and eliminate any iteration of pulses between liquid cell and the laser. Since only stimulated Brillouin scattering was studied, filters were used to remove the stimulated Raman scattering. The reflection attenuators allow the beam intensity to be adjusted from well below to well above threshold for the transverse stimulated scattering processes. Observations of both stimulated Raman scattering and stimulated Brillouin scattering indicated that the polarization of the transverse scattering was the same as the laser to 1 part in 1000. It is a significant advantage of the transverse geometry that scattered laser light can be reduced to a negligible level by proper baffling of the equipment.

Since the stimulated transverse scattering is plane polarized and the scattered laser light can be completely eliminated, it is possible to display separately both the laser and the stimulated Brillouin scattering on the same interferogram. As shown in Fig. 1, a portion of the laser beam is picked off and its polarization rotated 90° by a half-wave plate. This is combined with the stimulated Brillouin scattering on the beam splitter just ahead of the Fabry-Perot interferometer. In the film plane of the camera is a four-quadrant polaroid analyzer which provides the separation of the laser and the scattering.

An interferogram of water at 20.7°C taken with this apparatus is shown in Fig. 2(a). The laser is displayed in the upper right and lower left quadrants while the stimulated Brillouin scattering is displayed in the remaining two quadrants. The most intense line in the scattered spectrum corresponds to the Stokes shift for Brillouin scattering at 90° from the laser beam. The

[7] E. Burlefinger and H. Puell, Phys. Letters **15**, 313 (1965).
[8] R. V. Wick, D. H. Rank, and T. A. Wiggins, Phys. Rev. Letters **17**, 466 (1966).
[9] J. M. Burch, in *Proceedings of the Third International Quantum Electronics Conference*, edited by P. Grivet and N. Bloembergen (Columbia University Press, New York, 1964).
[10] S. A. Collins and G. R. White, in *Proceedings of the Third International Quantum Electronics Conference*, edited by P. Grivet and N. Bloembergen (Columbia University Press, New York, 1964).

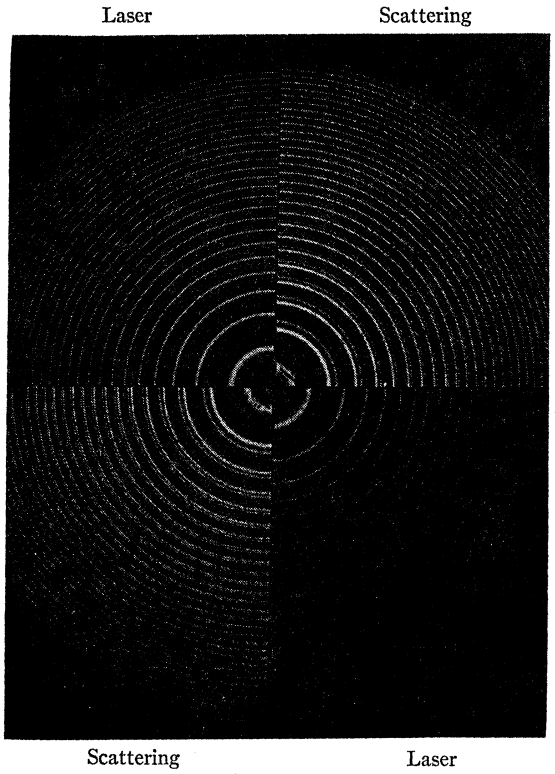


FIG. 2. Transverse stimulated Brillouin spectrum of water at 20.7°C. (See text for line identification.)

second line in the scattered spectrum is stimulated Brillouin scattering, corresponding to 180° scattering driven by the 90° line. It also propagates transverse to the laser beam. The 180° line in Fig. 2 has overlapped one order of the Fabry-Perot interferometer. Interferometers with interorder spacings of 0.2680, 0.5560, and 0.8950 cm^{-1} are used in order to evaluate overlap of orders. Thus, it is possible to measure the hypersonic velocity at two frequencies simultaneously. Table I compares the values of the hypersonic velocities obtained from the transverse stimulated Brillouin scattering with those obtained from spontaneous Brillouin scattering. It is easily seen that, as expected, the values agree to within less than 5%. The hypersonic velocity of water at two different temperatures has been included in Table I. This shows that the high power density does not greatly alter the apparent temperature characteristics of the medium during the laser pulse. A temperature rise of 15–20°C in the scattering region would have been easily observable.

TABLE I. Comparison of stimulated and spontaneous hypersonic velocities.

Liquid	T(°C)	Hypersonic velocity (this work) (m/sec)	Hypersonic velocity spontaneous measurement (m/sec)
H_2O	20.7	1485±25	1485,[a] 1485[b]
H_2O	58.5	1545±20	1550[b]
CH_3OH	20.7	1130±25	1118[a]
Acetic acid	20.7	1035±50	1169[a]
CCl_4	20.7	1020±37	1020[a]
$SiCl_4$	20.7	780±20	•••
Chloroform	20.7	1050±25	•••
C_6H_6	20.7	1500±28[c] 1420±28[d]	1501[a] •••

[a] R. Y. Chiao and P. A. Fleury, in *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Co., New York, 1966). (No error limits given.)
[b] Ultrasonic velocity measurement from M. Greenspan and C. Tschiegg, J. Res. Natl. Bur. Std. **59**, 249 (1957).
[c] Below threshold for appearance of anti-Stokes line.
[d] Above threshold for appearance of anti-Stokes line.

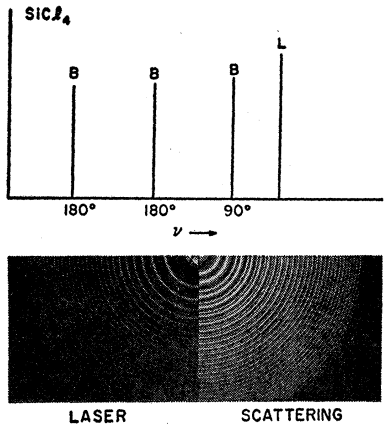


FIG. 3. Transverse stimulated Brillouin spectrum of $SiCl_4$ showing multiple 180° shifts produced just above threshold.

It should be noted that in these experiments stimulated anti-Stokes Brillouin scattering is never observed. At high power levels it is possible to produce several iterative 180° shifts, as is shown in Fig. 3. Since in this case both light waves and acoustic waves are traveling antiparallel in the active medium, stimulated anti-

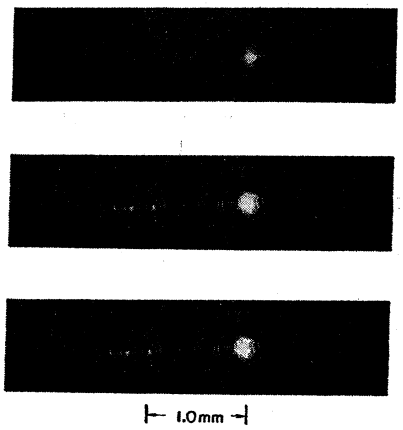


FIG. 4. Three photographs of the end of the line focal region showing large numbers of filaments.

Stokes Brillouin scattering should be observed. The lack of this anti-Stokes scattering was also observed by Pine[4] in his work on stimulated Brillouin scattering in a transverse resonator. He explains that the coherence length $\pi c/2n(\omega_{90°}-\omega_{180°})$ is smaller than a multiple pass length in his cavity. The insufficient coherence length would then seriously reduce the total gain available to the anti-Stokes Brillouin scattering. The coherence length between the Stokes and anti-Stokes Brillouin scattering is 1–2 cm for common materials, and is about the same length as the line focal region in the present experiments. In this case, the finite coherence length would only slightly reduce the gain available to the anti-Stokes Brillouin scattering. Thus, it is necessary to suggest another mechanism to explain the lack of stimulated anti-Stokes Brillouin scattering.

To better understand the processes involved, photographs were taken of one end of the line focal region. Essentially the same equipment was used as shown in Fig. 1 with the interferometer and its camera replaced with a camera to produce photographs of the end of the focal region magnified by a factor of 40. Three of these photographs taken just above threshold for the appearance of stimulated Brillouin scattering in C_6H_6 are shown in Fig. 4. Numerous filaments of 10-μ diameter or less are easily seen. Since the filaments in benzene scatter light from the side[11] a top view of the focal region was photographed. This is shown in Fig. 5. It is easily observed that the laser beam breaks up into filaments and, in addition, transverse filaments are formed intersecting the laser filaments. These filaments, which are transverse to the laser beam, did not start out as a large-diameter laser beam and self-focus. Reducing the power of the laser beam reduces the number of filaments observed, but under no condition is stimulated Brillouin scattering observed that is not contained in filaments. Very similar results are observed for such diverse materials as carbon tetrachloride and water.

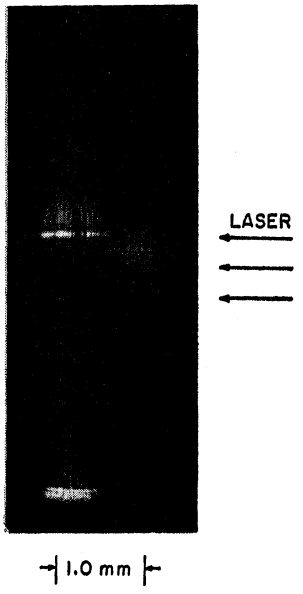


FIG. 5. Top view of a portion of the line focal region showing transverse and longitudinal filaments. Laser beam is incident from right.

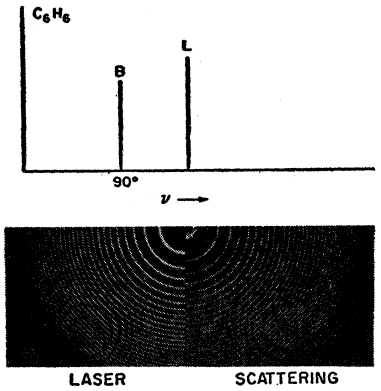


FIG. 6. Transverse stimulated Brillouin spectrum for benzene below threshold for the appearance of the anti-Stokes line.

The transverse filamentation suggests an explanation for the lack of stimulated anti-Stokes Brillouin scattering. If the light waves and acoustic waves traveling antiparallel in the focal region are not contained in the same volume of space the anti-Stokes process could not be stimulated. Since the light waves are contained in discrete filaments, it is quite probable that different filaments are propagating in different directions. Thus, the spatial overlap required for the stimulation of the anti-Stokes Brillouin scattering is not achieved.

Measurements have been attempted of the power contained in individual filaments. Just above threshold it is sometimes possible to observe only several filaments rather than the several hundred usually observed. To estimate the power in a filament, we have made the following assumptions: (1) All filaments are equal with respect to power contained. (2) Average time duration of the filaments is given by the spectral width of the Brillouin line. (3) The filaments contain only stimulated Brillouin scattering. This last statement is probably justified, since near threshold the total integrated stimulated Brillouin scattering greatly exceeds the total stimulated Raman scattering. The total energy output was observed with an ITT FW 128 biplanar photodiode with an S-1 photosurface. These estimates for benzene, which are admittedly crude, indicate that the power contained in a filament is approximately 10 kW. If we use the relation[12]

$$P_c \geq (1.22\lambda)^2 c/64n_2,$$

we can estimate the magnitude of the nonlinear refrac-

[11] R. G. Brewer and C. H. Townes, Phys. Rev. Letters **18**, 196 (1967).

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

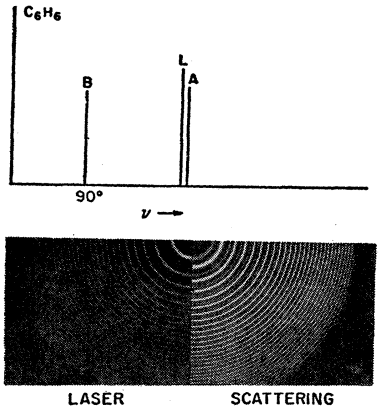


FIG. 7. Transverse stimulated Brillouin spectrum for benzene above threshold for the anti-Stokes line.

tive index n_2 to be 21×10^{-12} esu. This value is above most previous estimates. However, recently Hellwarth[13] has calculated the nonlinear refractive index of a wide variety of liquids. His theory yields an even larger value for the nonlinear index for benzene than our estimate, although there are still serious uncertainties in the calculation for densities as high as those of liquids.

While observing stimulated Brillouin scattering in benzene, it was noticed that a line appeared in the scattered spectrum at an almost identical frequency with the laser. Detailed measurements of many interferograms showed that this line was shifted upward from the laser in frequency about 200 MHz. This line was observed to have a definite threshold which was above that of the 90° stimulated Brillouin scattering. The angular divergence of this anti-Stokes line was approximately 3° which was about one-third that of the stimulated Brillouin scattering. It was also noted under these conditions that the 90° stimulated Brillouin shift was approximately 200 MHz low. Figures 6 and 7 show the transverse stimulated spectrum of benzene below and above threshold for observation of the anti-Stokes line.

Since the experimental work discussed here was completed, two papers have appeared discussing the origins of such anti-Stokes lines with small frequency shifts. Herman and Gray[14] have predicted the existence of a stimulated thermal Rayleigh scattering in liquids with an anti-Stokes frequency shift of approximately one-half the laser linewidth. This has been observed by Rank *et al.*[15] in carbon tetrachloride and carbon disulfide to which iodine has been added to enhance the laser absorption. This is very similar to the frequency shifts we have observed. In addition the theory[14] also predicts the stimulated scattering to the Stokes side of the Stokes-Brillouin line with a shift of similar magnitude. The present experiment would not have been able to separate clearly this new stimulated process from stimulated Brillouin scattering. However, a composite of both lines would be observed. By reference to Table I it can be seen that the 90° stimulated Brillouin line shift, measured at a power level above threshold for the appearance of the anti-Stokes line, is clearly less by about 200 MHz than that measured below threshold for this process. Thus, it is possible that both these effects have been noticed in these experiments. A detailed comparison with the theory would require a detailed knowledge of the various complex nonlinear multiphoton absorption processes that are taking place.

III. CONCLUSIONS

Stimulated scattering in liquids is easily obtained without the necessity of external resonators. In the absence of frequency-dependent feedback, the stimulated Brillouin frequency provides a reliable measurement of the hypersonic velocity. Near-field photographs have shown the existence of transverse filamentation which has provided a convenient explanation for the lack of stimulated anti-Stokes Brillouin scattering. It has also reemphasized that the details of the stimulated processes in liquids are really controlled almost entirely by the self-focusing properties of liquid. We have also indicated the possibility of self-focusing thresholds much below those usually considered. In addition observations have been made in benzene indicating the possible presence of stimulated thermal Rayleigh scattering.

ACKNOWLEDGMENT

We would like to thank J. F. Holzrichter for his assistance in reducing the Fabry-Perot interferograms to usable data and for his help in preparing this manuscript.

[13] R. W. Hellwarth, Phys. Rev. **152**, 156 (1966); **163**, 205 (1967).

[14] R. M. Herman and M. A. Gray, Phys. Rev. Letters **19**, 824 (1967).

[15] D. H. Rank, C. W. Cho, N. D. Foltz, and T. A. Wiggins, Phys. Rev. Letters **19**, 828 (1967).

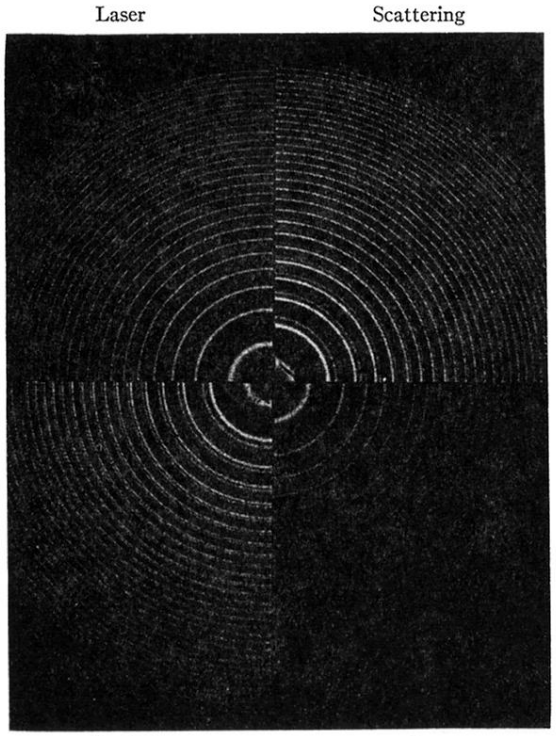


FIG. 2. Transverse stimulated Brillouin spectrum of water at 20.7°C. (See text for line identification.)

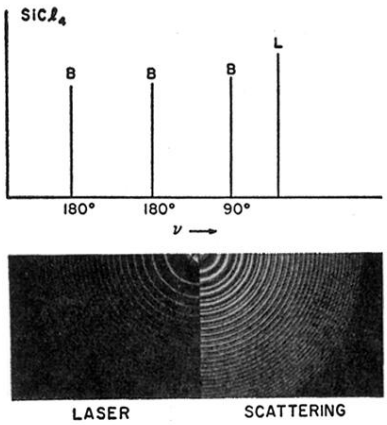


FIG. 3. Transverse stimulated Brillouin spectrum of $SiCl_4$ showing multiple 180° shifts produced just above threshold.

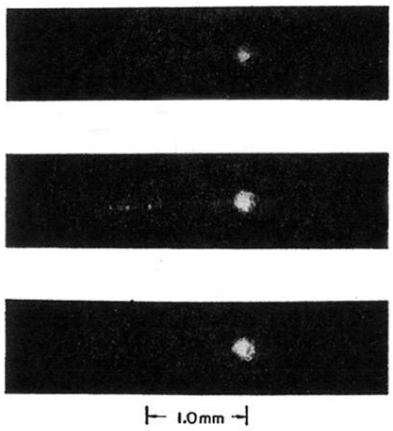


FIG. 4. Three photographs of the end of the line focal region showing large numbers of filaments.

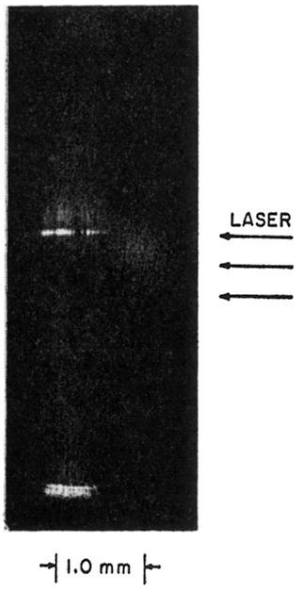


FIG. 5. Top view of a portion of the line focal region showing transverse and longitudinal filaments. Laser beam is incident from right.

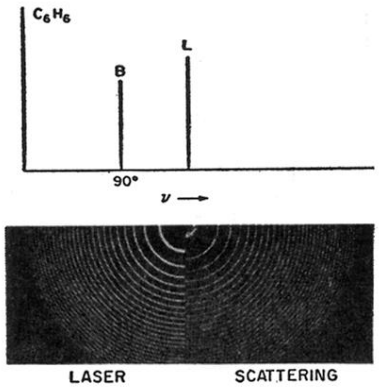
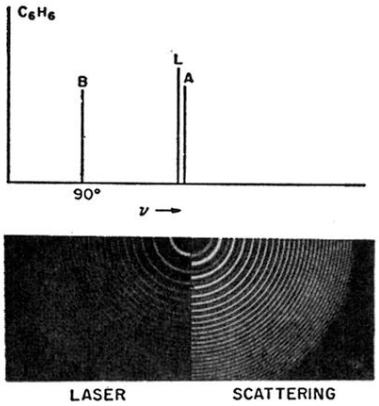


FIG. 6. Transverse stimulated Brillouin spectrum for benzene below threshold for the appearance of the anti-Stokes line.



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