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(December, 1967)

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NORMAL AND STIMULATED RAMAN SPECTROSCOPY

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University of Toronto
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RAMAN LINEWIDTHS FOR STIMULATED THRESHOLD
AND GAIN CALCULATIONS*

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Linewidths in the normal Raman spectra of several liquids of interest in stimulated Raman scattering have been measured including carbon disulfide, benzene, toluene, oxygen and nitrogen. The spectra were excited with $6328 \text{ \AA}$ radiation from a He-Ne laser and examined with a pressure-scanned Fabry-Perot interferometer.

*This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the U.S. Office of Naval Research, and the Department of Defense. Also supported by the National Research Council, Canada, and the University of Toronto.

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While it has long been known that Raman bands of totally symmetric vibrations exhibit extremely sharp lines even in liquids and solids, very few studies of the normal Raman effect have been concerned with accurate linewidth measurements. However, with the discovery of stimulated Raman scattering¹ there has been an increasing need for such measurements since both the threshold for stimulated scattering and the gain are dependent on the linewidths appearing in the normal Raman spectra. In the last few years, several linewidth measurements for liquids² and one for a solid³ (calcite) have been reported, all making use of grating instruments. In the present paper we describe a method for normal Raman linewidth measurements which is capable of even higher accuracy. Radiation at $6328 \text{ \AA}$ from a He-Ne laser is used for excitation of the spectra and a high resolution Fabry-Perot interferometer for measurement. With this apparatus linewidths of interest in stimulated Raman scattering have been measured for several liquids including carbon disulfide, benzene, toluene, oxygen and nitrogen.

The experimental arrangement is shown in Fig. 1. It is based on the arrangement used by Chiao and Stoicheff⁴ for

Brillouin spectroscopy and is similar to systems used by Murray and Javan,⁵ and Ducuing et al⁶ for Raman scattering with excitation by argon-ion lasers. In the present arrangement, the He-Ne laser has a power output of 150 mW and a linewidth of 0.025 cm^{-1} (full width at half intensity). The $6328 \text{ \AA}$ radiation is isolated with a $10 \text{ \AA}$ band-pass filter and reflected into the sample cell. For the organic liquids the cell is a quartz capillary 2.5 mm in diameter and 50 cm in length which acts as a light guide for the incident as well as for the scattered radiation. For liquid N_2 and O_2 the cell is a simple dewar providing a scattering length of 7 cm between two flat windows. Raman radiation scattered in the backward direction is selected with a $100 \text{ \AA}$ band-pass filter and analysed with a pressure-scanned Fabry-Perot interferometer. For CS_2 , O_2 and N_2 the interferometer spacing was 2.950 mm; for N_2 a larger spacing of 9.943 mm was also used; for benzene and toluene the spacing was reduced to 0.869 mm in order to increase the spectral free range to 576 cm^{-1} . Pressure scanning with CO_2 from vacuum to 2 atm provided a chart of 4 interferometer orders when the 2.95 mm spacer was used. The measured instrumental width (made up of contributions from the laser width, Fabry-Perot response function for a reflectivity of 0.995 and the size of the detector aperture) is 0.050 cm^{-1} at $6328 \text{ \AA}$ and at almost all of the Raman wavelengths. At the wavelength of the N_2 Raman line ($7423 \text{ \AA}$) the mirror reflectivity decreased

to 0.923 resulting in a calculated instrumental width of 0.063 cm^{-1} with the 2.95 mm spacer and 0.038 cm^{-1} with the 9.943 mm spacer.

In Fig. 2 is shown the 656 cm^{-1} Raman line of CS_2 in two adjacent orders. The line is relatively narrow and symmetric. It is superimposed on a broad background made up of overlapping of the wings of the line from different orders; in addition, several nearby Raman lines (at 645.9 cm^{-1} due to $\text{CS}^{32}\text{S}^{34}$ and at 647.5, 652.7 cm^{-1} due to "hot" bands) contribute to the low intensity peak observed between the adjacent orders of the 656 cm^{-1} line. To determine the Raman linewidth the observed spectrum was compared with a computed spectrum obtained by assuming a Lorentzian line shape and convoluting this with the instrumental profile. Various Lorentzian linewidths were used in the calculations until good agreement between the observed and computed spectra was achieved taking into account the overlap of the different orders.

Similar computations were carried out for the most intense Raman lines in the several liquids studied here to obtain the values of full-width at half intensity, listed in Table I. The estimated errors of the linewidths are approximately 5% of the measured widths although slightly larger for C_6H_6 and O_2 . The 992 cm^{-1} line of C_6H_6 is the broadest of those observed and is slightly asymmetric due to overlapping by the much weaker lines at 983.6 cm^{-1} of $\text{C}^{13}\text{C}_5^{12}\text{H}_6$ and at

998.0 cm^{-1} of a "hot" band. The O_2 Raman line exhibited the lowest intensity of the liquids studied, since the laser radiation falls in the region of the strongest absorption band of liquid O_2 .

The narrowest Raman line observed is that of liquid N_2 , equal to $0.067 \pm 0.004 \text{ cm}^{-1}$. Its intensity profile is shown in Fig. 3 where it is compared with that of the computed line. It may be noted that the same linewidth was obtained for both forward and backward scattering within the quoted experimental error. This value of the linewidth together with the relatively high intensity of the N_2 Raman line indicates perhaps one of the lowest thresholds for stimulated Raman emission and one of the highest gains.

The linewidth of 0.50 cm^{-1} obtained for liquid CS_2 is especially noteworthy in relation to the gain in stimulated Raman scattering. Although the recent literature² gives linewidths as narrow as 0.9, 1.0 and 1.4 cm^{-1} the value usually used in gain calculations is 3 cm^{-1} . The present measurement shows that the linewidth is approximately 1/6 of this value and hence the gain is a factor of 6 larger than that quoted in the recent literature.⁷

In conclusion we should like to point out that not only does this method allow the accurate measurement of relatively narrow Raman lines in liquids (with possible extension to gases and solids) but it also provides a means of studying

line shapes.

We are very grateful to G.I.A. Stegeman for the computer program used in the analysis.

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TABLE I

LINEWIDTH MEASUREMENTS OF TOTALLY
SYMMETRIC VIBRATIONAL RAMAN LINES

<u>Liquids</u>	<u>Raman Shift</u> <u>(cm⁻¹)</u>	<u>Full Width at</u> <u>Half Intensity</u> <u>(cm⁻¹)</u>
Nitrogen	2331	0.067 ± 0.004
Oxygen	1555	0.117 ± 0.008
Carbon disulphide	656	0.50 ± 0.02
Toluene	1002	1.94 ± 0.07
Benzene	992	2.15 ± 0.15

FIGURE CAPTIONS

- Fig. 1 Experimental apparatus for Raman linewidth measurements.
- Fig. 2 Interferogram of the 656 cm^{-1} Raman line of liquid CS_2 . Two adjacent orders are shown recorded with an interferometer spacer of 0.295 cm. The smooth line is the computed spectrum.
- Fig. 3 Interferogram of the Raman line of liquid N_2 recorded with an interferometer spacer of 0.295 cm. The smooth line is the computed intensity profile.

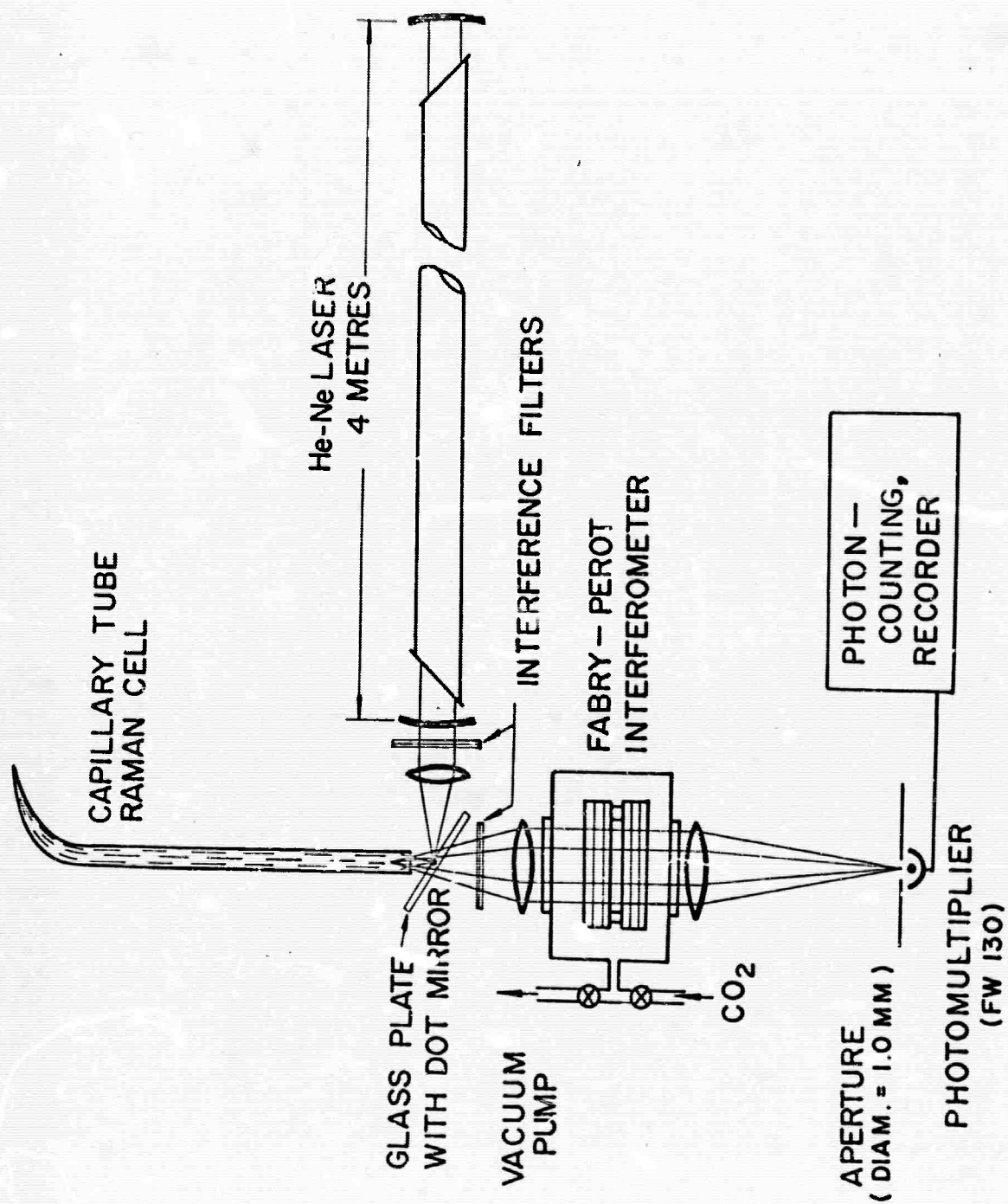
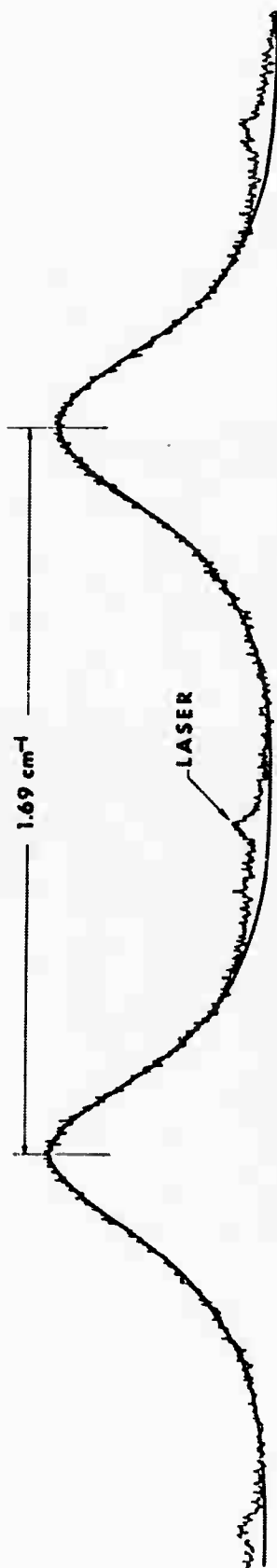


FIG. 1 CLEMENTE & STUCHIKOFF

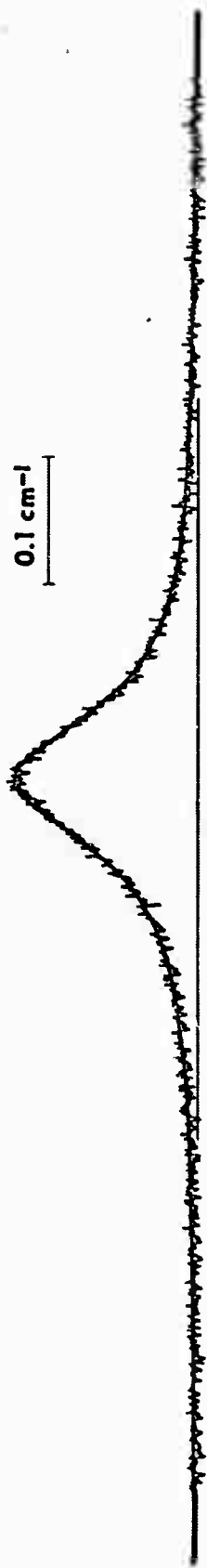
CS_2



1.692 cm^{-1} CS_2

N_2

0.1 cm^{-1}



J. R. CLEMENTS & STONCHER

ANGULAR DISTRIBUTION OF "SURFACE" RADIATION IN
STIMULATED RAMAN SCATTERING*

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ABSTRACT

Anti-Stokes emission in sharply defined cones has been observed from fine filaments in a mixture of acetone with 5 to 10% CS₂. The cone angle is in agreement with the momentum-matching condition that the longitudinal but not the transverse components sum to zero.

*This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the U.S. Office of Naval Research, and the Department of Defense. Also supported by the National Research Council, Canada, and the University of Toronto.

ANGULAR DISTRIBUTION OF "SURFACE" RADIATION IN STIMULATED RAMAN SCATTERING

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The emission of anti-Stokes radiation during the stimulated Raman process is known to have specific directional distributions which are strongly dependent on the experimental conditions. The two commonly observed distributions have been labelled Class I and II by Garmire.¹ Class I distribution is given by the momentum-matching condition for plane waves, $\vec{k}_0 + \vec{k}_{n-1} = \vec{k}_{-1} + \vec{k}_n$ where nth-order anti-Stokes radiation with wave vector $\vec{k}_n$ is generated from laser radiation ($\vec{k}_0$) and Stokes radiation of the first and n-1 orders ($\vec{k}_{-1}$ and $\vec{k}_{n-1}$). Such a distribution of anti-Stokes radiation has been observed in calcite,² in diamond,³ and in several liquids¹ when feedback of Stokes radiation at the phase-matching angle is present. Class II distribution occurs at larger angles than Class I. It has been observed in many liquids, but is not well understood.

A distribution of anti-Stokes emission which is different from Class I has been proposed by Szöke⁴ and labelled "surface" radiation. The first-order anti-Stokes emission is characterized by the relation

$$\vec{k}_1 \cos \theta = 2 \vec{k}_0 - \vec{k}_{-1} \quad (1)$$

where θ is the angle between the first-order anti-Stokes

emission and the forward-directed laser and first-order Stokes radiation. Such an angular distribution may occur when first-order Stokes radiation is strongly directional and parallel to the incident laser wave. Maker and Terhune⁵ have also suggested the occurrence of such a distribution in the limit that the longitudinal but not the transverse components of the phase velocities sum to zero. More recently Shimoda⁶ has presented a comprehensive account of possible angular distributions and suggested that "surface" radiation may occur in the presence of first-order Stokes radiation in filaments which are very long in comparison with their diameter.

We report here the observation of "surface" radiation in mixtures of acetone and carbon disulfide under experimental conditions which satisfy the boundary conditions of the theory. We have observed first-order anti-Stokes emission in sharply-defined cones in the forward direction at the angle θ given by Eq. (1). At the same time, first-order Stokes radiation has been observed in very fine filaments produced by self-trapping of the radiation.

The radiation used in these experiments was produced by a giant-pulse ruby laser. The laser pulses had a duration of ~ 40 nsec, a power output of ~ 5 MW and a spectrum composed of one strong axial mode with one or two weaker modes. The liquids used were pure acetone and various concentrations of carbon disulfide in acetone, in cell lengths of 10 and 20 cm. Both parallel and focused laser beams were used to excite the

stimulated Raman emission. Also, the effect on these experiments of possible stimulated Brillouin scattering was checked by separating the laser and sample cell by 6 m. The results were similar under these various conditions of excitation.

In pure acetone, usually only Class I distribution of anti-Stokes radiation is produced. An example of the far-field "ring" pattern is shown in Fig. 1a. This photograph was obtained on Polaroid film placed at right angles to the incident laser beam and at the focal plane of a lens ($f = 17$ cm). When a small amount (5 to 10 per cent by volume) of carbon disulfide is added to acetone, the angular distribution is completely changed. It usually consists of a sharp ring whose diameter is close to that of surface radiation represented by Eq. (1), together with a broad ring of smaller diameter (due to Class II radiation) as shown in Fig. 1b. When the excitation is close to threshold only the sharp ring is observed, Fig. 1c: at relatively high excitation the two rings merge and are not as distinct as in Fig. 1b. The half-angle θ of the sharp rings can be accurately determined and appears to vary slightly, from 6.5 to 6.8×10^{-2} rad, in different photographs taken with the 10% CS_2 -acetone mixture. (The half-angle of the broad ring is $\sim 5.7 \times 10^{-2}$ rad.) According to Eq. (1) the angle θ is calculated to be 7.3×10^{-2} rad, based on the available data of the refractive index in this mixture.⁷ (For pure acetone Eq. (1) gives $\theta = 6.4 \times 10^{-2}$ rad.)

Photographs (Fig. 2) of the exit end of the cell (that

is, near-field patterns) revealed the presence of filaments of laser, Stokes and anti-Stokes radiation in the CS_2 -acetone mixtures and their absence in pure acetone. Near threshold, the laser and Stokes filaments are of comparable intensity and occur with diameters of 20 to 100 μ . The corresponding anti-Stokes filaments appear to have smaller diameters, 8 to 16 μ ; they are of higher intensity when they arise from the small diameter Stokes filaments; also the narrowness of the resulting rings indicates that the emission region is 1 to 2 mm long. The total energy emitted from a small Stokes filament is estimated to be $\sim 10^{-6}$ joule, while the intensity ratio of radiation from a laser or Stokes filament to that from an anti-Stokes filament is approximately $1:10^{-3}$.

The broad anti-Stokes rings are also emitted from filaments, but with Stokes radiation ~ 100 times as intense as that of the filaments giving rise to the sharp anti-Stokes rings. They appear to be produced in a long emission region approximately 5 to 10 mm.

At the laser power levels used in the present experiments only stimulated radiation at $\Delta\nu = 2920 \text{ cm}^{-1}$ was observed in pure acetone and in the mixtures with CS_2 : no stimulated radiation at $\Delta\nu = 656 \text{ cm}^{-1}$ due to CS_2 was detectable. The role of the CS_2 is to produce filaments, and since its concentration in acetone is small, the threshold for filament formation in the mixture is estimated to be much higher than that in pure CS_2 (~ 10 to 20 kW).

From the present observations we conclude that the sharp anti-Stokes rings arise from "surface" radiation. Not only is their diameter in good agreement with the condition that only the longitudinal components of the phase velocities sum to zero, but they are also observed only in the presence of optical filaments in the liquid mixture. The 10 per cent difference between the observed and calculated cone angles may be explained in terms of the intensity dependent refractive index (and the finite length of the filaments) which tends to decrease the angle of emission, as observed. Following Shimoda,⁶ we write for the emission angle θ of the first anti-Stokes radiation, $\frac{1}{2}(\frac{\theta}{n})^2 = \Delta k/k_1 - g\delta - r^2/(4\ell^2)$ where $\Delta k = k_1 + k_{-1} - 2k_0$, r and ℓ are the radius and length of filament, δ is the fractional change of the wave vector due to the intensity dependent refractive index and g is a correction factor (~ 1) due to the waveguide effect. The sharpest anti-Stokes rings are only slightly affected by the finite lengths of the filaments since $r^2/(4\ell^2) \sim 10^{-6}$, whereas $\Delta k/k_1 = 1.3 \times 10^{-3}$. An estimate of the correction $g\delta$ ($\sim \delta$) for the intensity dependent index can be obtained from the approximate value of the intensity ratio ($\sim 10^{-3}$) of anti-Stokes to Stokes intensity in the filaments. For plane waves this ratio is calculated⁸ to be approximately $(\text{Stokes gain})^2/(4\Delta k)^2$. With the available data for the Raman gain,⁹ the Stokes and laser intensity in a filament is determined to be $\sim 15 \text{ GW/cm}^2$. The value of 10^{-11} esu for the intensity-dependent refractive index of CS_2 ¹⁰ (and therefore 10^{-12} esu for this mixture) then leads to $\delta \sim 10^{-4}$ which accounts

for the difference between the observed emission angle and that calculated from Eq. (1). Slight differences in the laser intensity of filaments would thus also explain the small observed variation in emission angle from pulse to pulse.

Similar experiments were carried out with cyclohexane, another liquid known to emit Class I radiation.¹ In mixtures with CS₂, again a sharp ring was observed, the measured emission angle being 6.8×10^{-2} rad for a 5% CS₂ mixture. Comparison with the angle for surface radiation, Eq. (1), was not possible since refractive index data for the mixture are not available, but for pure cyclohexane Eq. (1) gives 6.8×10^{-2} rad.

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FIGURE CAPTIONS

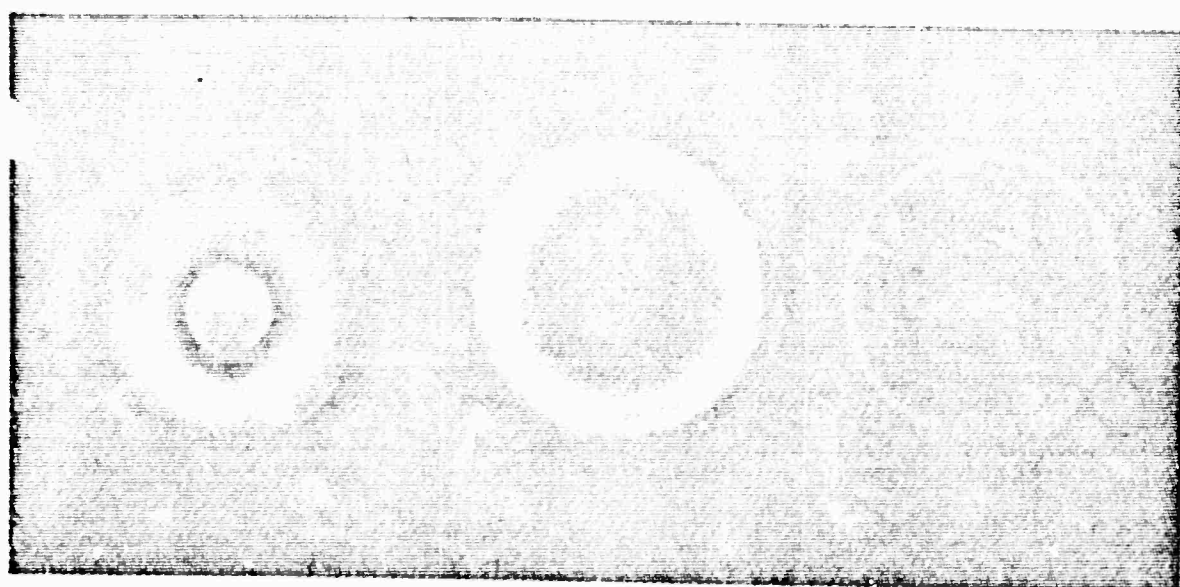
Fig. 1. Far-field "ring" patterns of anti-Stokes radiation in (a) pure acetone, (b) 10% CS₂-acetone mixture above threshold; (c) 10% CS₂-acetone mixture near threshold. There is no apparent difference in the spectral width (at $\Delta\nu = 2920 \text{ cm}^{-1}$) of the anti-Stokes stimulated radiation in (b) and (c).

Fig. 2. Stimulated Raman emission from a 10% CS₂-acetone mixture, (a)(b)(c) photographed simultaneously.

(a) Near-field photograph of anti-Stokes radiation showing filaments and associated "ring" patterns. The differences in ring diameters are due to the fact that the generating filaments occur at different distances from the exit window.

(b) Near-field photograph of Stokes radiation (approximately the same magnification as (a)). A comparison of (a) and (b) shows that the anti-Stokes rings are generated from radiation in the filaments of smallest diameter.

(c) Far-field photograph of anti-Stokes radiation.

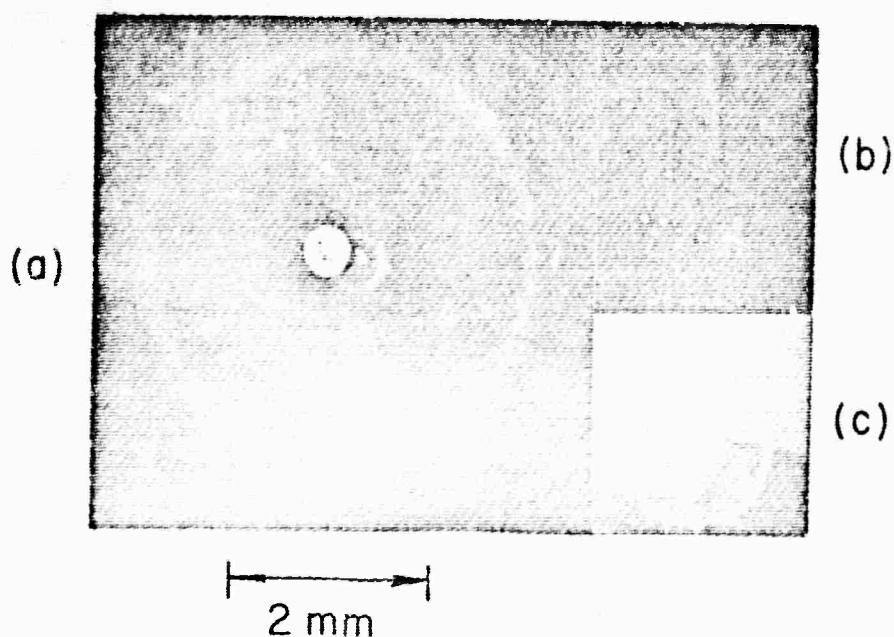


(a)

(b)

(c)

Fig. 1.



(a)

(b)

(c)

2 mm

Fig. 2.

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