

INVESTIGATION OF STIMULATED RAMAN SCATTERING AND SELF FOCUSING
OF LIGHT IN CARBON DISULFIDE

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Stimulated Raman scattering (SRS) and self focusing of light in liquid carbon disulfide were studied at various temperatures. The intensities of a number of Stokes and anti-Stokes SRS components were measured at various temperatures of the medium. The cross section of the exciting line beams and the first and second Stokes components in an aperture of 3.5° in the direction of the exciting light and in the opposite direction were also investigated. The evidence indicates that self focusing of light changes when the medium is cooled. The intensities of these beams were compared by simultaneous recording, yielding information on the effect of temperature on the asymmetry of the indicatrix of SRS.

STIMULATED Raman scattering (SRS) of light and self focusing of a light beam in a medium are among the most interesting phenomena of nonlinear optics currently studied in numerous publications. Self focusing and the attendant splitting of the light beam into fine light-conducting filaments change the excitation conditions of the stimulated Raman scattering. Therefore the connection between these two phenomena is studied in a number of papers.^[1, 2]

In the present paper we consider SRS and self focusing in liquid carbon disulfide at various temperatures. A change in temperature changes the optical constants of the material, the Kerr constant, and the probability of stimulated Raman scattering. Consequently by varying the temperature we can change the mode of self focusing of light in the material and the conditions of excitation of SRS, and we can study the interaction of these two effects.

EXPERIMENTAL METHOD

Stimulated Raman scattering was excited by a giant pulse from a ruby laser. Figure 1 shows the optical diagram of the experiment. The flat dielectric mirrors of the resonator had reflection coefficients of 100 and 8%. A ruby 12 mm in diameter and 120 mm long was used. An alcohol solution of cryptocyanine was used as the Q-switch. The giant pulse power was 10 MW. The ruby was maintained at constant temperature to ensure stability of the exciting radiation. Nitrogen vapor served to cool the ruby. The temperature in the reflector was monitored by a thermocouple. The ruby was pumped by two IFP-2000 lamps. The variation of ruby emission energy did not exceed 5%. The exciting light was focused by a lens with $F = 250$ mm inside the capsule with the investigated material. A portion of the exciting light was reflected by a plane parallel plate towards a photodiode serving to monitor the stability of the giant pulse. Lenses (6 and 8) were placed confocally. As a result, the light scattered in the forward and backward directions formed practically parallel beams after passing through the lenses. The beams were then reflected by flat plates onto prisms that rotated them through 90° and arranged them at different heights so

that both beams could be observed at the same time.

A gas laser was used to check the symmetry of the experimental arrangement. A plane-parallel plate was placed normal to the axis of the system in place of the cell with the scattering material. The light of the gas laser was first directed along the axis of the system and then was reversed. A comparison showed that the intensities of the light beams reflected from the plate in the forward and reverse directions were the same within the limits of experimental error.

The arrangement allowed us to record beams with apertures up to 3.5° . The cylindrical lens focused the scattered light onto the slit plane of a three-prism Huet spectrograph (the dispersion of the spectrograph in the red region was $47 \text{ \AA}/\text{mm}$). To avoid errors caused by a shift of the scattered light spot, spectra were recorded on photographic plates without the spectrograph slit in a number of experiments. Image density marks together with the slit were photographed with the aid of a ribbon lamp. We used the INFRA-760 plates in our experiments. The experiments showed that the slope of the density curve for these plates is the same when a ribbon lamp is used to illuminate the marks as when a giant pulse ruby laser light is used for this purpose.

To ensure photometric reliability, the intensity of some SRS components was attenuated with neutral filters. Filter transmission was measured under experimental conditions with a spectrograph at the required wavelength. The intensities of SRS components were

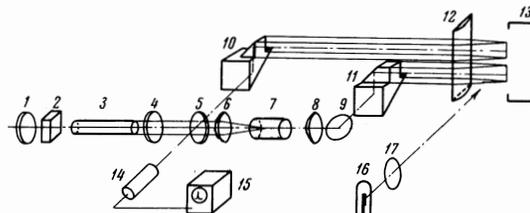


FIG. 1. Optical diagram of the experimental setup. 1, 4 – dielectric resonator mirrors, 2 – cell with cryptocyanine, 3 – ruby, 5, 9 – plane parallel plates, 6, 8 – lenses with $F = 250$ mm, 7 – cell with carbon disulfide, 10, 11 – prisms, 12 – cylindrical lens, 13 – spectrograph, 14 – photodiode, 15 – oscilloscope, 16 – ribbon lamp, 17 – lens.

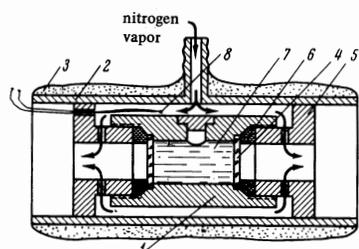


FIG. 2. Cell for the study of SRS at low temperatures. 1 – body of cell, 2 – metallic cylinder, 3 – asbestos thermal, 4 – quartz windows, 5 – metal bushings, 6 – teflon washers, 7 – investigated liquid, 8 – thermocouple.

obtained by averaging the data of 15–20 measurements. The rms error of the measurements was 15%.

The SRS spectra were investigated in the cell shown in Fig. 2. The scattering material was held in a brass cylinder with quartz windows and was placed in a thermal insulation jacket. Cold nitrogen vapor was passed through the cell in the directions indicated by arrows to obtain low temperature. The temperature in the cell was measured with a copper-constantan thermocouple. The cell windows did not fog up when temperature was lowered to -100°C and the liquid carbon disulfide remained transparent. The latter effect was verified experimentally. The filled cell was exposed to a free-running laser light (SRS was not excited in this case). The intensity of the light remained constant after the passage through the carbon disulfide at room and low temperatures. Analogous results were obtained when a helium–neon laser was used as the light source.

RESULT OF MEASUREMENTS

1. Thermal Dependence of SRS Intensity.

Stimulated Raman scattering in carbon disulfide at room temperature was observed at 656 cm^{-1} (Stokes and anti-Stokes components) and 1312 cm^{-1} (doubled frequency of the first Stokes component). The above lines were recorded both in the direction of the exciting light (“forward” direction) and in the reverse direction (“backward” direction). The observation of the anti-Stokes component in the “forward” direction apparently indicates the presence of “class II” SRS in our experiment. The scattering which occurs in fine filaments according to Garmire^[3] and Shimoda,^[4] propagates along the axis of the exciting light and in the cones, without meeting Townes^[5] phase matching conditions. The “backward” scattering of SRS light was not discussed in these papers. We note that under the experimental conditions (the aperture did not exceed 3.5°) the anti-Stokes radiation governed by the phase matching condition could not fall into the spectrograph.

The results of measurements of the relative intensity of SRS components in the “forward” direction at various temperatures are given in the table (St is the Stokes component and aSt is the anti-Stokes component).

The table shows that decreasing temperature causes a considerable rise in intensity of all the SRS components. Particularly strong is the rise of intensity of the second Stokes component, which varies by more than two orders of magnitude within the temperature interval from $+20^{\circ}$ to -90°C . A mutual saturation of intensity occurs in the first Stokes and the first anti-Stokes components at -30 and -70°C respectively; these tem-

Table. Relative intensity of SRS lines at various temperatures

$t, ^{\circ}\text{C}$	$[I(t, ^{\circ}\text{C})/I(20^{\circ}\text{C})] \cdot 100$		
	1 St	1 aSt	2 St
+20	100	100	100
-30	700	300	4000
-70	500	500	38500
-90	440	300	36000

peratures also give rise to the third Stokes and the second anti-Stokes components in the SRS spectrum. Figure 3 shows the general view of the intensity distribution in the SRS spectrum at various temperatures. Owing to the large difference in the component intensities, their relative magnitudes are displayed on a logarithmic scale in the graph. According to Fig. 3, a drop in the temperature of the medium increases the intensity of the SRS spectrum, which redistributes itself in favor of the higher-order components. Some intensity saturation is observed at -80 – -90°C .

The rise in intensity with decreasing temperature is probably due to two factors: increasing intensity of the ordinary Raman scattering, and a change in the self-focusing of light in carbon disulfide. As we know, the intensity of stimulated Raman scattering is associated with that of the ordinary Raman scattering. An investigation of the thermal dependence of ordinary Raman scattering in carbon disulfide showed that a drop in temperature from $+20$ to -90°C increases the intensity of the 656 cm^{-1} line (the first Stokes component) approximately by a factor of three.^[6, 7] If the dependence of the intensity of the first Stokes component of the SRS on that of the ordinary Raman scattering is assumed to be exponential,^[8] then the intensity of the Stokes SRS component should rise about ten times within the investigated temperature interval.

On the other hand, a number of authors^[9, 10] reported observing the self focusing phenomenon in liquids as a result of giant pulses of light. So-called “hot” filaments containing very high energy density are formed in the liquid. This phenomenon may lead to a decrease in the SRS threshold. It was shown in^[11, 12] that the self focusing effect depends on the Kerr and electrostriction constants. In carbon disulfide the self focus-

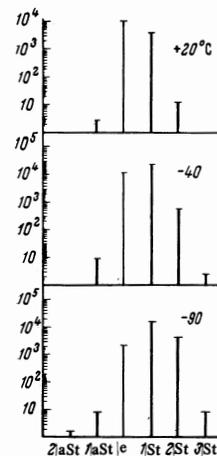


FIG. 3. Relative intensities (logarithmic scale) of lines in the SRS spectrum of carbon disulfide at three temperatures (e – exciting line).

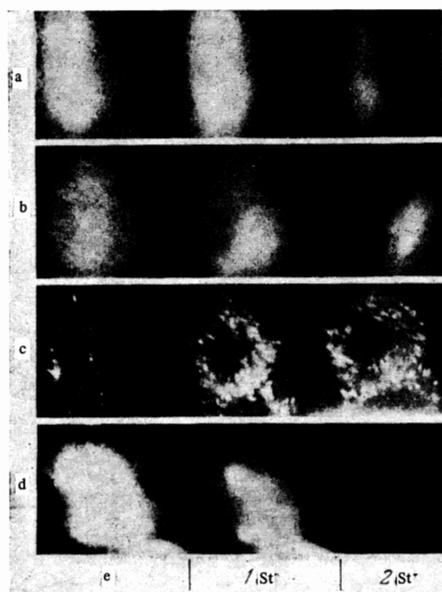


FIG. 4. Cross section of the beam of exciting line and SRS Stokes components. a – “forward” direction at +20°C, b – “backward” direction at +20°C, c – “backward” direction at -70°C, d – “backward” direction at -90°C.

ing action is mainly dependent on the Kerr constant^[11], which is sufficiently high in this material (at room temperature it equals $32 \times 10^{-8} \text{ cm}^{-1} \text{ cgs esu}^{-2}$). The Kerr constant increases approximately by 80% when the temperature drops 100°C.^[13] It is possible that such a rise in the Kerr constant can cause a substantial change in the self focusing of light in the material and consequently can affect the SRS intensity.

2. Self Focusing of Light in Carbon Disulfide at Various Temperatures

Our equipment allowed us to record the intensity distribution in the cross section of the beam propagating in the “backward” direction. In this case the spectrum was photographed without the slit and the cylindrical lens. A redistribution of the intensity in the cross section of scattered light beams of various frequencies was observed in the plane of the photographic plate. Figure 4 shows the most typical photographs obtained at +20, -70, and -90°C.

In the “forward” direction, the scattering of all frequencies was observed in the form of slightly blurred spots whose diameter was only insignificantly larger than that of the spot produced by the exciting laser emission (the photographs always showed two spots slightly displaced from one another, owing to the reflection from two surfaces of the pivoting plates). Diffusion rings or a number of points were observed at room temperature in the “backward” direction. When the temperature fell to -60–-70°C the rings broke up into individual bright points distributed along their perimeters or over the entire area within the rings. When the material was cooled to -90°C the scattered light coalesced into two or three individual points in most of the photographs. Figure 4 shows that in the cross section of the “backward” scattered beam the intensity

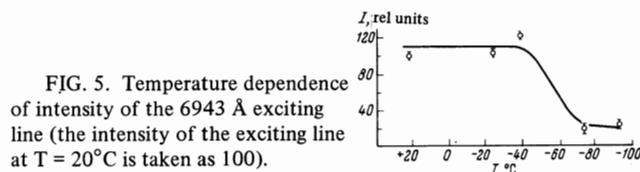


FIG. 5. Temperature dependence of intensity of the 6943 Å exciting line (the intensity of the exciting line at $T = 20^\circ\text{C}$ is taken as 100).

distribution is identical for the exciting¹⁾ component and for the first and second Stokes components. The origin of the rings that preferentially attract the bright points is not quite clear. The appearance of bright points in place of the diffusion scattering seems to be due to the change of the self focusing of light. A drop in temperature gives rise to many light-conducting filaments which coalesce at -90°C into individual light-conducting channels.

The change occurring in the self focusing mode during the transition from room to lower temperatures seems to cause a significant change in the coefficient of transformation of the exciting light into SRS. This change is manifested by a rise in the total SRS intensity (Fig. 4) and an attenuation of the exciting line intensity. Figure 5 shows the temperature dependence of the intensity of the exciting line. When the material is cooled below -40°C the intensity of the exciting line drops sharply (approximately by a factor of six within the temperature interval from -40 to -80°C), reaching some degree of saturation at -80–-90°C.

An additional confirmation of the fact that a change in the self focusing mode affects the coefficient of transformation of light into SRS is furnished by the data we obtained on the intensity of SRS as a function of the intensity of the exciting line I_0 and the coefficient K of the ordinary Raman scattering. This dependence can be theoretically regarded as exponential.^[8] If we plot the experimental points on a graph where the ordinate axis represents the logarithm of SRS intensity and the abscissa represents the product I_0K , the points will break up into two groups (Fig. 6). The first group of points corresponds to high temperatures and a lower coefficient of transformation, and the second group to low temperatures and a higher coefficient of transformation of the radiation into stimulated Raman scattering.

3. Asymmetry of the Indicatrix of SRS.

Our equipment permitted a simultaneous measurement of SRS intensity in the “forward” (I_f) and “backward” (I_b) directions. The ratio I_f/I_b is an important characteristic of the Raman scattering of light. According to the general theory of the phenomenon, $I_f/I_b = 1$ for ordinary Raman scattering. The experiments revealed some deviations from this relationship.^[14, 15] As far as SRS is concerned, the available data^[16, 17] indicate an asymmetry of the “forward” and “backward” scattering; however, these data were obtained by a method that is not beyond criticism, and are somewhat contradictory. According to our measurements within the investigated temperature interval, and also at room temperature,^[16, 17] the intensity of light scattered “forward” in the case of Stokes and anti-Stokes components

¹⁾Intensity of unshifted-frequency radiation that passed through the cell.

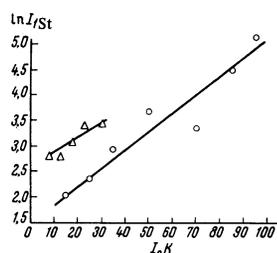


FIG. 6. The logarithm of intensity of the exciting line by the intensity of ordinary Raman scattering. Δ — points corresponding to low temperatures, \circ — points corresponding to high temperatures.

is considerably larger than the intensity of light scattered "backward." This ratio for the first Stokes component is 10:1 and does not change markedly over the entire temperature interval, while the component intensity itself undergoes large changes. At room temperature this ratio is 1:4 for the first Stokes component and 1:100 for the second. When the temperature is lowered these components manifest an ever increasing preference for the "forward" scattering of light.

The asymmetry of the SRS indicatrix could be due to the different polarization of light scattered by the material "forward" and "backward." The large number of reflecting surfaces can in this case attenuate the measured intensities to a different degree. In order to investigate the role played by this factor we repeated the measurements with the polaroid situated in the slit plane and oriented in two mutually perpendicular directions. The results of these two series of measurements were in agreement within the limits of experimental error. Consequently the observed asymmetry of the indicatrix does not depend on the method of measurements but doubtlessly characterizes the SRS phenomenon itself. The asymmetry of the SRS has not yet found a satisfactory theoretical explanation. The thermal dependence of the ratio of the "forward" and "backward" intensities for the first anti-Stokes and the second Stokes components suggests that the magnitude of the observed asymmetry of the indicatrix may possibly depend on the mode of self focusing of light in matter.

CONCLUSIONS

1. The intensity of stimulated Raman scattering of light in carbon disulfide responds to decreasing temperature by increasing and redistributing in favor of

higher-order components. Some saturation of SRS intensity takes place at the temperature of $-80 - -90^\circ\text{C}$. This dependence of SRS intensity on temperature is apparently due to the following two reasons: the increasing intensity of the ordinary Raman scattering with decreasing temperature, and the change in the coefficient of transformation of the exciting light into SRS accompanying the cooling of the material.

2. The increasing coefficient of transformation of light into SRS is due to a change in the mode of self focusing in carbon disulfide. The following stages are observed in the course of self focusing of light: splitting of the light beam into fine light-conducting filaments, and consolidation of these filaments into broader channels.

3. A considerable asymmetry of the SRS indicatrix is observed within the temperature interval from $+20$ to -90°C . The intensity of light scattered "forward" is much larger than that scattered "backward." When the material is cooled down, the first anti-Stokes and second Stokes components exhibit an ever increasing prevalence of forward scattered light.

¹D. L. Close, C. R. Guiliano, R. W. Hellwarth, F. J. McClung, and W. G. Wagner, *IEEE J. of Quantum Electronics*, QE-2, 1111 (1966).

²R. G. Brewer and J. R. Lifshitz, *Phys. Lett.* **23**, 79 (1966).

³E. Garmire, *Phys. Lett.* **17**, 251 (1965).

⁴K. Shimoda, *Japan. J. Appl. Phys.* **5**, 33 (1966).

⁵C. H. Townes, *Phys. Rev. Lett.* **11**, 160 (1963).

⁶A. I. Sokolovskaya, *Trudy FIAN* **27**, 63 (1964).

⁷A. I. Sokolovskaya and Z. Kecki, *Bull. de L'Acad. Polonaise des sciences, serie des sci. chem., geol. et geogr.* **6**, 133 (1958).

⁸V. A. Zubov, M. M. Sushchinskiĭ, and I. K. Shuvalov, *Usp. Fiz. Nauk* **89**, 49 (1966) [*Sov. Phys.—Uspekhi* **9**, 346 (1966)].

⁹C. C. Wang and G. W. Rarette, *Appl. Phys. Lett.* **8**, 256 (1966).

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