STIMULATED RAMAN SCATTERING AND SELF-FOCUSING OF LIGHT IN LIQUID NITROGEN

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The spectral and spatial characteristics of the first Stokes component of stimulated Raman scattering of light (SRS) are measured in liquid nitrogen. A single-mode ruby laser is used for excitation. SRS is observed in the direction of the exciting radiation and in the opposite direction in nitrogen layers of different thicknesses and at different pumping energies. The SRS characteristics in these directions differ considerably for a given nitrogen thickness and pumping energy. In particular, the scattered power per unit solid angle in the direction opposite to the exciting radiation exceeds that in the direction of the exciting radiation by about an order of magnitude. Self-focusing of light in liquid nitrogen is observed.

SOME ot the first experimental investigators of the characteristics of stimulated Raman scattering of light (SRS)[1] observed that the scattered intensity is asymmetric. In benzene the first Stokes component of SRS was considerably more intense in the direction of the exciting radiation than in the opposite direction. This result was later confirmed in other investigations,[2-4] although the quantitative data disagreed widely.

The observed "anomaly" was not accounted for by the existing theories.[1,5] The discovery that light is self-focused in liquids and the subsequent experimental studies showed that many "anomalies" of SRS result from this phenomenon. The low threshold and large conversion coefficient of light in SRS were explained in the case of liquids with a large Kerr coefficient.[6-8] In carbon disulfide ultrashort Stokes pulses of SRS were observed[9] in the direction opposite to the propagation direction of the exciting radiation, and the effect was considered to be associated with the self-focusing of light. However, despite the considerable augmentation of experimental information and the development of theoretical ideas,[3,9-11] no final explanation has been given for the different forms of SRS in the direction of excitation and in the opposite direction.

All the aforementioned investigations were concerned with either the SRS energy or power. The spectral width, divergence, and spatial intensity distribution of SRS remained unknown. A sufficiently complete picture could therefore not be obtained of SRS formation in different substances nor could the cause of the observed asymmetry be ascertained. It was therefore of great immediate importance to study the indicated SRS parameters.

In the present work we measured, in the direction of the exciting light ("forward") and in the opposite direction ("backward"), the total energy, brightness, intensity distribution in the beam cross sections, divergence (spread), and spectral width of the first Stokes component of SRS in liquid nitrogen. We observed SRS in nitrogen layers that were 8, 20, 40, and 100 mm thick at different pumping rates.

Liquid nitrogen possesses several advantages as the object of investigation and has been widely used. It is very important that there is a low SRS threshold in liquid nitrogen and a high SMBS (stimulated Mandel'shtam-Brillouin scattering) threshold, thus permitting "decoupling" of the laser and nitrogen-containing vessel.

SRS energy was measured by means of thermocouples that were calibrated with a power meter. The intensity distribution within the beam cross sections of the first Stokes component of SRS forward and backward, as well as within the cross section of the exciting light beam, were observed photographically. The divergence (spread) of the beams was determined at the half-maximum intensity of the spot obtained at the focus of a lens and was calculated using the formula $\theta = d/F$, where d is the beam diameter on a photographic plate and F is the focal length of the lens (F = 120 mm). Line widths were measured with Fabry-Perot interferometers of 30-mm thickness and cameras with F = 840 and 1300 mm. To isolate the required wavelength we used interference filters and glass filters. Blackening of the photographic plates was converted to intensities with the aid of a density scale. A stepped optical wedge was illuminated with ruby laser light or with SRS light.

A ruby laser with a passive shutter was used to excite SRS. The temperature of the ruby was kept constant by blowing liquid nitrogen vapor through the illuminator. The maximum laser energy was 0.25 J; the giant pulse duration was 20 nsec. A single mode was observed in the ruby oscillation spectrum. The stability of laser oscillation with respect to energy, time, and frequency was monitored with a calorimeter, I2-7 oscillograph, and 30-mm Fabry-Perot interferometer, respectively. It was verified experimentally that feedback between the nitrogen vessel and laser did not occur. The energy of the unshifted backward light component was about 10^{-4} of the energy passing through the nitrogen. The Dewar holding the nitrogen had plane-parallel windows placed at an angle $\approx 10°–12°$. Light reflected from the windows was deflected from the optical axis of the system.[1)] [11]

[1)] Our experimental observations and calculations based on the gain for nitrogen (given in [12]) show that generation did not occur at the end faces of our experimental vessel (in the 8-mm vessel because the amplifying layer was very thin, and in the vessels from 20 to 100 mm because the Dewar windows were considerably in need of adjustment).

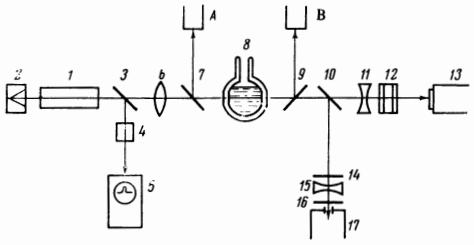

FIG. 1. Scheme of apparatus. 1—ruby laser; 2—calorimeter; 3, 7, 9, and 10—rotatable plates; 4—photodiode; 5—oscillograph; 6—lens (F = 230 mm); 8—Dewar containing nitrogen; 11 and 15—scattering lenses; 12—Fabry-Perot interferometer; 13—camera; 14—matte plate; 16—image density scale; 17—spectrograph.

The experimental arrangement is represented in Fig. 1. The thermocouples, Fabry-Perot interferometers and cameras, and photographic plates were located at A or B, depending on the measurements to be obtained. The exciting light was focused inside the Dewar by means of a long-focus lens (F = 230 mm). The diameter of the light spot at the lens focus was 0.8 mm. The maximum power density was 2.5×10^9 W/cm^2.

For all the utilized thicknesses of liquid nitrogen the intensity distribution within the cross section of the first Stokes component in the forward direction has the form of a diffuse spot. Figure 2 shows the beam cross sections of the first Stokes and anti-Stokes components in the forward direction for a 100-mm layer of nitrogen. The diffuse spot of first Stokes line scattering surrounds a ring of anti-Stokes absorption; this is observed especially well in the cases of the 40- and 100-mm vessels. The ring of anti-Stokes radiation represents absorption; the angle of the anti-Stokes cone is $\approx 180'$. As the nitrogen thickness is increased from 20 to 100 mm there is almost no intensity rise at the maximum of the first Stokes beam; the integral forward SRS intensity increases as a result of the wing intensities.

Figure 3 shows how the energy of the first Stokes line propagated forward depends on pump energy in the 20- and 100-mm nitrogen layers. In the case of the 20-mm Dewar we worked with the region where SRS intensity saturation is observed for the center of the beam; in the case of the 100-mm Dewar saturation occurs over the entire beam diameter.

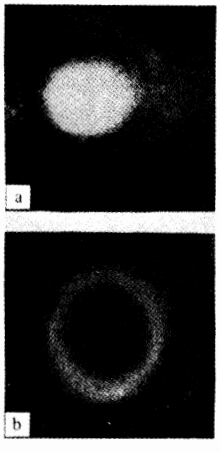

FIG. 2. Cross section of the forward SRS beam: a—first Stokes component; b—first anti-Stokes component.

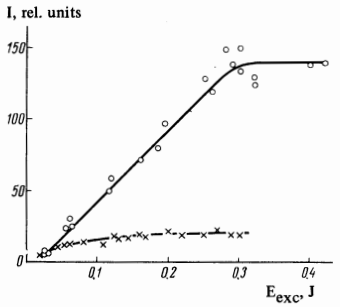

FIG. 3. Energy of the first Stokes line in the forward direction versus energy of the exciting radiation. O—20-mm nitrogen thickness; X—100-mm nitrogen thickness (the ordinate scale is reduced by the factor 10 for this curve).

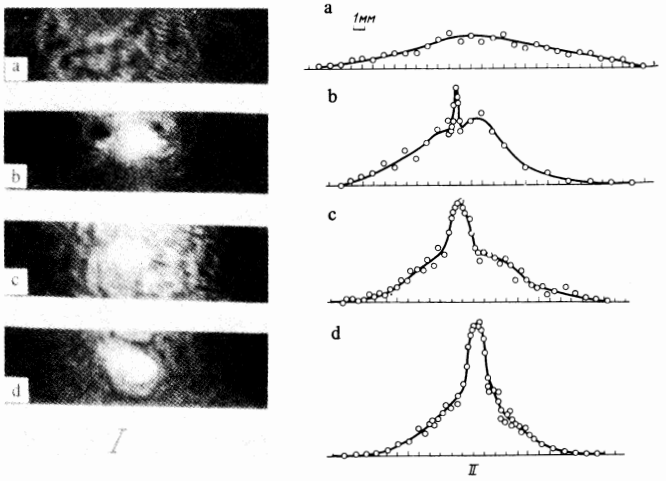

FIG. 4. Intensity distribution within the beam cross section of the first Stokes line in the "backward" direction for different thicknesses of the nitrogen layer: a—8 mm, b—20 mm, c—50 mm, d—100 mm (I—photographs, II—photometric measurements).

Figure 4 illustrates how the first Stokes line in the backward direction depends on the nitrogen thickness. In the case of the 8-mm vessel the same kind of diffuse spot is observed in both the backward and forward directions. As the vessel length is increased, at the center of the diffuse spot radiation appears having greater brightness and considerably smaller spread. With 0.2-J pumping 8 mm of nitrogen is close to the threshold, while 20 mm is the threshold for the appearance of intense scattering at the center of the first Stokes line beam. In the 100-mm Dewar practically the entire energy of the first Stokes line going forward is concentrated in a spot that is many times brighter than the diffuse part of the scattering. In the 40- and 100-mm cases a fairly intense ring is seen around a bright point. Anti-Stokes backward scattering is practically absent.

Figure 5 shows photographs and photometry results for the beam cross section, forward and backward, of the first Stokes line in the 100-mm vessel, registered on plates placed at an identical distance from the vessel; these SRS beam cross sections are markedly different. We also measured the relative energies of the first Stokes line for the two directions in vessels of the different lengths:

Dewar length, mm:	8	20	40	100
I_f/I_b:	1.6±0.6	4.3±1	3.0±0.5	5±0.6

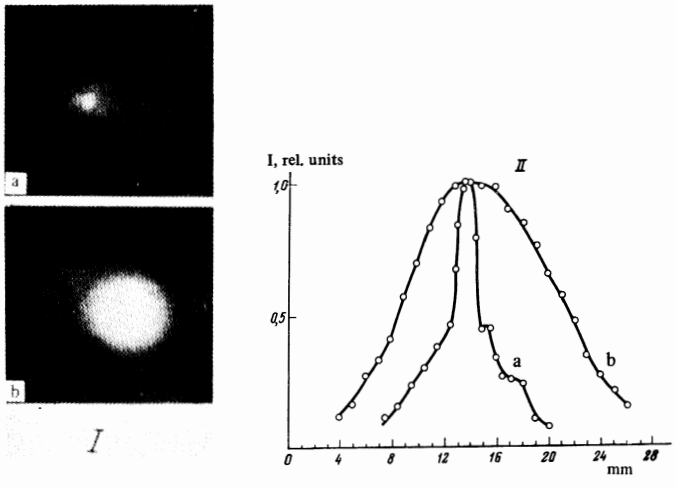

FIG. 5. Intensity distribution within the beam cross section of the Stokes line for a 100-mm nitrogen layer: a—in the backward direction, b—in the forward direction (I—photographs, II—photometric measurements).

In the 8-mm vessel the forward/backward SRS energy ratio is close to unity.[2)] In greater lengths the scattered energy is peaked forward. We also measured the divergences and spectral widths of the ruby radiation and of the first Stokes line of SRS. We obtained 46′ ± 2′ for the ruby light spread, 180′ ± 6′ for the first Stokes line forward, 170′ ± 8′ and 30′ ± 2′ for the backward direction in the 8-mm and 100-mm vessels, respectively.

With 8-mm nitrogen thickness and 10^9 W/cm^2 pump power the forward and backward beams of the first Stokes line of SRS are closest with respect to both spatial structure and total energy. Their spreads are also close at 180′, which considerably exceeds the spread of the ruby radiation.

Above 20-mm nitrogen thickness the SRS energy, spread, and intensity distribution within the SRS beam cross section differ in the forward and backward directions.

Finally, above 40-mm nitrogen thickness the pattern becomes stable and further increase of the thickness at constant pump energy produces no changes in the energy and spatial characteristics of the first Stokes line. It is noteworthy that the SRS power per unit solid angle is one order greater in the backward than in the forward direction.

The spectral width of the investigated SRS lines does not change essentially with the thickness of the nitrogen layer. We obtained for SRS the line widths forward and backward (in cm^{-1}):

	Forward	Backward
First Stokes line	0.053 ± 0.005	0.043 ± 0.004
Ruby radiation	0.028 ± 0.003	—
Ruby radiation passing through nitrogen	0.037 ± 0.006	—

These data show that the observed width of the SRS line exceeds the width of the exciting line and is smaller than the width of an ordinary Raman scattering line, which, with exclusion of the excitation and instrumental functions, is 0.067 ± 0.004 cm^{-1}, according to [12]. The observed spectral widths of the first Stokes line are close in the forward and backward directions. It should be noted, however, that for the backward direction several photographs in the 100-mm vessel show splitting of the first Stokes line into components of shifted frequencies.

The asymmetry of SRS was formerly linked, as a rule, to the self-focusing of the exciting radiation in the medium and to SRS radiation from light-conducting fibers. These were usually materials with a high Kerr constant (such as carbon disulfide). Heretofore self-focusing has not been observed in liquid nitrogen, and many authors have assumed that it does not occur because the electrostriction constant of nitrogen is small and its Kerr constant is an order smaller than that of carbon disulfide. However, a calculation of the critical power for self-focusing[13,14] shows that its value is ≈ 550 kW, which means that the self-focusing threshold can be reached with the customary experimental pump powers.

To elucidate the role of this factor we performed the following experimental work. A lens with F = 100 mm produced in the photographic plane a 10-fold enlarged image of a plane in nitrogen located 8 mm from the end face of the vessel. In this way we photographed the SRS field near the front and back faces of the 100-mm Dewar and the ruby field near the front face with and without nitrogen.[3)]

The photographs are shown in Figs. 6 and 7. A comparison of Fig. 6a and b shows a single bright point of 30-μm diameter in nitrogen instead of a broad diffuse spot of exciting radiation. The spread of the ruby radiation (100′ ± 10′) passing through the nitrogen considerably exceeds that of ruby radiation in air. The interference pattern includes one component with the width 0.037 ± 0.006 cm^{-1}. The foregoing results show that in liquid nitrogen self-focusing of an unshifted component occurs.

A comparison of Fig. 7a and b shows that the intensity distribution of the first Stokes line near the front face is diffuse, but that near the back face it consists of a series of points of about 30-μm diameter. The SRS field distribution near the faces of the vessel does not duplicate the distribution of the exciting radiation in nitrogen.

It appears possible that the observed formation of SRS in nitrogen layers of different thicknesses will permit two variant explanations, because the SRS energy, on the basis of the conversion coefficient,[17] is of the order of the critical self-focusing energy.

Variant 1. This explanation is based on the assumption of SRS self-focusing in liquid nitrogen. This is supported by the existence of a critical length and by the size of the points in the SRS field that are observed at a vessel face, although the combined spread backward is less than the divergence that could come from each point separately. Different nonlinear mechanisms can

[2)] The data given here for the 8-mm vessel correspond to a quite high pumping level, although they do not lie in the region of SRS saturation. The problem of SRS symmetry is of special interest at near-threshold excitation, [11] but we have not attempted to arrive at a solution in the present work.

[3)] A similar technique was used in [15], where it was confirmed that self-focusing points exist in liquid toluene. [16]

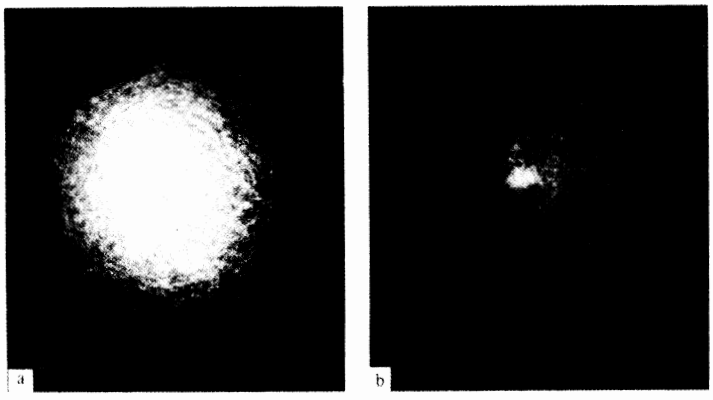

FIG. 6. Ruby field near the front face of the vessel: a—empty Dewar, b—nitrogen-filled Dewar.

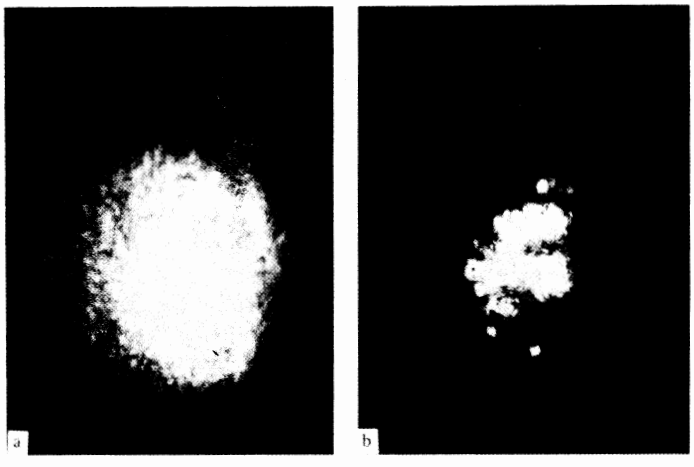

FIG. 7. SRS field: a—near the front face of the vessel, b—near the back face of the vessel.

possibly produce self-focusing in SRS. For example, the refractive index could be varied by the Kerr effect or by increased molecular polarizability under excitation.[17-19]

Variant 2. If the self-focusing threshold for SRS is not reached the observed intensity distribution can be explained as follows. SRS would arise in inhomogeneities of the ruby radiation field (in the regions of highest energy), being excited mainly in the back part of the vessel, while in the remaining part the excitation is less effective because of pumping depletion associated with both the production of SRS and the self-focusing of the light. As the radiation passes through the medium in the forward direction, the SRS at first emphasizes the inhomogeneities of the exciting radiation in virtue of the exponential pump dependence of SRS, after which saturation sets in and the inhomogeneities are gradually smoothed out. For SRS going forward the front of the vessel operates as a nonlinear amplifier in the saturation regime.

Since SRS is excited most effectively near the back face of the nitrogen vessel, radiation in the backward direction does not become saturated over the entire cross section but only wherever the ruby radiation is most intense; these inhomogeneities are thus emphasized, in virtue of the exponential dependence of SRS on pumping. A critical length exists for the production of radiation in the backward direction; thereafter an increase in the thickness of the nitrogen layer leads to no change of the SRS parameters.

In the presence of intense stimulated Mandelstam-Brillouin scattering and other nonlinear effects the development of SRS in liquids becomes even more complex.[20]

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