Fine structure of stimulated Raman scattering line in substances with small kerr constants

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Spectral splitting of the Stokes and anti-Stokes components of stimulated Raman scattering was observed in liquid nitrogen and in single-crystal calcite. No splitting was observed for the unshifted component. The total width of the spectral distribution and the spacing between the fine-structure components were measured as functions of the excitation conditions. Near the stimulated Raman scattering threshold, the first Stokes component was a signle one with a width much smaller than the line width of the exciting radiation. An asymmetry of the spectral composition of the first stimulated Raman scattering Stokes component was observed in the propagation direction of the exciting radiation and in the opposite direction. Self-focusing of the stimulated Raman scattering of light was observed in the absence of self-focusing of the exciting radiation.

It has been observed many times that the spectral distribution of laser-emission intensity becomes more complicated after passing through a medium with large polarizability anisotropy[1-7]. Thus, when ruby-laser light passes through carbon disulfide, additional components are produced near the laser line, or else a continuous spectrum extending from several dozen to several hundred reciprocal centimeters. Most workers have attributed the spectral broadening of the laser emission to phase modulation of the light, due to the change in the nonlinear refractive index in the self-focusing filaments in media with large Kerr constants. Quantitative comparisons of the spectral broadening of a laser line in carbon disulfide were carried out by Wong and Shen[8] on the basis of the theory of moving foci[9], and they found good agreement between experiment and this theory. In addition to a spectral broadening extending over hundreds of reciprocal centimeters, Mash et al.[10] observed a fine structure of the Rayleigh-line wing in media with large polarizability anisotropy, with components ~ 0.2 cm^{-1} away from the unshifted frequency. The appearance of these components was attributed by them to stimulated temperature scattering of the Rayleigh-line wing.

While the features of the spectral distribution of the unshifted component in anisotropic liquids have been investigated in sufficient detail, the number of experimental studies of the spectral distribution of first- and higher-order Stokes and anti-Stokes components of stimulated Raman scattering (SRS) is quite limited. All that are available are studies of the broadening of the first Stokes component of nitrobenzene and carbon disulfide[11,12], and indications of the presence of a fine structure in the first Stokes components of SRS in nitrobenzene, carbon disulfide[10,13], and single-crystal calcite[14]. There are practically no systematic investigations, however. This applies in particular to substances with small Kerr constants, since it has been assumed for a long time that the SRS in such substances is not complicated by the effect of self-focusing of the light, and all the observed irregularities agree with the theory[15].

We have shown in an earlier paper that when liquid nitrogen and calcite are excited by pulses of 20-nsec duration, self-focusing of the SRS components is observed, with a threshold lower than the self-focusing threshold of the exciting radiation[16]. The present paper is devoted to the spectral composition of SRS components in these substances.

The SRS was excited by a giant ruby-laser pulse with a passive shutter. When a Fabry-Perot etalon with a 150-mm base was used, one frequency was observed in the spectrum of the exciting radiation. The line width was 0.019 cm^{-1} and the pulse duration 20 nsec. The exciting radiation was focused into the medium by a lens with f = 230 mm, and the maximum power density at the focus of the lens was 160 MW/cm^2. The dispersing system was a three-prism spectrograph crossed with a Fabry-Perot etalon. The thickness of the etalon ranged in the experiments from 7 to 150 mm, and the apparatus function was 0.002 and 0.04 cm^{-1} for Fabry-Perot etalons of thickness 75 and 7 mm, respectively. The error in the measurements of the spectral distribution is estimated at 5–10%. The experimental setup is shown in Fig. 1. To prevent feedback from affecting the results, an optical delay was placed between the laser and the cell containing the investigated substance. The cells with nitrogen and the single-crystal calcite, with respective thicknesses 50 to 10 mm, had end faces inclined at an angle ~ 10 – 12° to eliminate SRS generation. The optical axis of the crystal was oriented along the system axis. We observed the frequencies 1085 cm^{-1} of calcite and 2329.7 cm^{-1} of liquid nitrogen in the SRS spectrum, and also the anti-Stokes and the second Stokes components.

A typical spectrogram of SRS in calcite at two temperatures and maximum excitation energy is shown in Fig. 2. At room temperature, using a Fabry-Perot etalon 7 mm thick, a unique distribution of the intensity with a distinct structure was observed in all three SRS components. It is important that no structure was observed on the laser line after passing through the medium. A structure was always produced at a given excitation energy and crystal temperature, although the number and relative intensities of the components varied somewhat. The results of the measurement of the total width of the distribution, of the number of the components, and of the magnitude of the splitting are given in Table I.

The spectral width of each structure component in calcite was approximately equal to the exciting-radiation line width (0.019 cm^{-1}), and the distance between components was larger than the distance between the longitudinal resonator modes (0.01 cm^{-1}). The number of components and the total width of the distribution were maximal for the second SRS Stokes component. When the temperature was lowered to −196°C, the structure was weakly resolved upon increase of the Fabry-

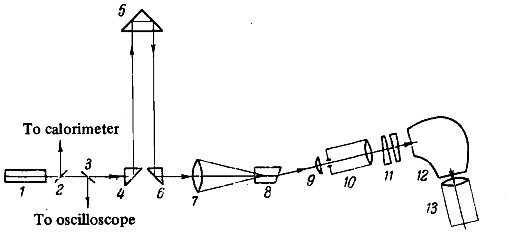

FIG. 1. Optical diagram of setup: 1—ruby laser, 2, 3—rotating plates, 4, 5, 6—optical-delay prisms, 7—lens; 8—sample; 9—lens; 10—collimator, 11—Fabry-Perot etalon, 12—prisms; 13—camera.

Perot etalon thickness to 30 mm, and the total width of the distribution decreased to approximately one-third of the value at room temperature. We note that the width of the distribution of the SRS components lies within the limits of the SRS line width obtained at the given temperatures by Park[17] (1.1 cm^{-1} at t = 20°C and 0.5 cm^{-1} at t = −196°C).

In the present study, we investigated the SRS in calcite at an exciting-radiation power density corresponding to the SRS saturation regime. The existence of components in the spectral distribution of the first SRS Stokes line in the saturation regime at room temperature and the equality of the width of one component to the width of the exciting radiation were noted also by Chirikov and co-workers[14]. It should be noted that the results, which were obtained using a different experimental setup and different excitation-source parameters, agree in the main with our results.

Investigations of the intensity distribution in the SRS field near the exit face of the crystal have shown that distinct self-focusing points are observed at the first and second SRS Stokes components at both room and nitrogen temperatures. No self-focusing points were observed in the field of the unshifted component.

The fine structure of the first Stokes component in liquid nitrogen was investigated with a Fabry-Perot etalon having a 75-mm base. We investigated the dependence of the number of components of the first Stokes line and the total spectral width of the distribution on the power density of the exciting radiation. Table II lists the measurement results. We see that the number of components increases from one at an exciting-radiation power density 10 MW/cm^2 to five at 65 MW/cm^2. The dependence of the number of components on the excitation power density in liquid nitrogen agrees with the analogous dependence observed in[14] in single-crystal calcite. The width of one component was 0.006 cm^{-1}, much less than the width of the exciting-radiation line (0.019 cm^{-1}). This is the first time that such a result was obtained for SRS of light. The feasibility of obtaining SRS with a line width smaller than the exciting-line width was indicated in[18].

We have compared the spectral distribution of the first SRS Stokes component of nitrogen with the distribution of intensity in the SRS field near the end faces of the cell, using different exciting-radiation power densities1). The comparison has shown that the appearance of one component in the structure of the first Stokes line corresponds to one self-focusing point in the SRS field and to a minimal coefficient of conversion of the excitation energy into SRS (near the threshold). A larger number of components corresponds to a larger number of self-focusing points in the SRS field in the discontinuity region (3 – 4 components) and points of saturation of the conversion coefficient (5 – 9 components).

TABLE I

t, °C		1 St	2 St	1 aSt
+20	Total width of distribution, cm^{-1}	0.14	0.23	0.18
	Number of components	3÷4	8—9	6
	Splitting, cm^{-1}	0.05	0.03	0.03 0.02
−196	Total width of distribution, cm^{-1}	0.067	0.104	0.070

TABLE II

Exciting-radiation power density, MW/cm^2	10	13	31	38	55	65
Number of components	1	3	3	4	5	5—9
Total spectral width, cm^{-1}	0.006	0.010	0.016	0.019	0.022	~0.050

Simultaneous observation of SRS with "forward" and "backward" excitation directions has shown that the first Stokes line has different spectral components in the different directions. A typical interference pattern is shown in Fig. 3.

The number of components and the magnitude of the "backward" splitting changed somewhat from experiment to experiment, but, as a rule, the asymmetry of the splitting in these two directions remained the same.

It is difficult at present to interpret unambiguously the observed fine structure of the SRS lines. The splitting is probably not connected with the modulation of the SRS by the laser modes, as was observed in a number of studies of substances with large Kerr constants. The laser employed by us generated only one longitudinal mode and no spectral splitting was observed at the unshifted component. Taking into account the value of the splitting of the SRS components, the absence of a structure in the unshifted component, and also the reproducibility of the results under different experimental

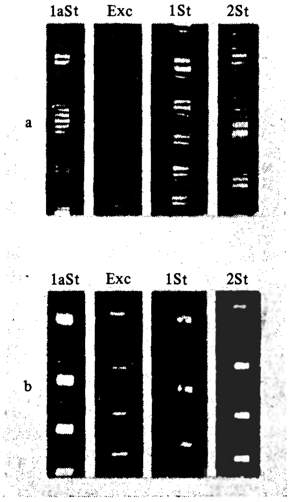

FIG. 2

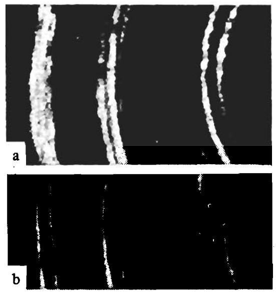

FIG. 3

FIG. 2. Interference pattern of SRS of light in single-crystal calcite: a–t = 20°C, b–t = 196°C. 1St, 2St–first and second Stokes components; 1aSt–first anti-Stokes component.

FIG. 3. Interference pattern of first Stokes component of SRS of light in liquid nitrogen: a—"forward," b—"backward."

conditions, it is also difficult to assume that the spectral distribution of the SRS is connected with mode selection caused by some element of the experimental setup.

One might assume that the large power density due to the self-focusing SRS gives rise to stimulated Mandel'shtam-Brillouin scattering (SMBS), for which the SRS line serves as the exciting line. However, an estimate of the splitting due to the SMBS line in liquid nitrogen yields a value 0.016 cm^{-1}, which is much larger than the experimentally expected splitting.

It should be noted that Brewer[13] observed in nitrobenzene and in carbon disulfide (substances with large Kerr constants) a fine structure both in the unshifted component and in the first SRS Stokes line. The splitting in the unshifted component was somewhat larger than in the SRS. This splitting was attributed to stimulated temperature scattering of the Rayleigh-line wing[10]. The mechanism whereby the structure is produced in the SRS line is not clear, but the cause of the splitting was assumed by the authors of[10] to be self-focusing of the light.

The absence of the structure in the unshifted component in our experiments appears to be due to the fact that self-focusing at the exciting line in nitrogen and calcite has a much lower threshold than SRS self-focusing[19]. It is important that the self-focusing in SRS is also shifted in time relative to the self-focusing of the laser pulse. The latter was observed experimentally by Korobkin, Lugovoĭ, and co-workers[20]. They have shown that the self-focusing points in SRS are delayed in time relative to the self-focusing at the exciting line in carbon disulfide and nitrobenzene by approximately 10 nsec.

The SRS line structure observed in our case is apparently due to self-focusing in SRS with amplification of the light at definite frequencies in the Raman-scattering line wing. We note that this process may be caused not by thermal reorientation of the molecules, as in the case of Rayleigh scattering[10], but by relaxation of the intramolecular oscillations[21]. There is no doubt that this point of view is at present still unsufficiently confirmed and that more detailed experimental research is necessary.

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[1)] Detailed investigations of self-focusing of light in liquid nitrogen were carried out by us earlier [16, 19].