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EFFECT OF TEMPERATURE ON STIMULATED RAMAN SCATTERING LIGHT IN SUBSTANCES WITH VARIOUS KERR CONSTANTS

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The temperature dependence of the spectral energy distribution of stimulated Raman scattering (SRS) and the coefficient of conversion of light into SRS are measured in a calcite single crystal and liquid carbon disulfide, i.e., in substances with significantly different Kerr constants. With decrease of temperature, the SRS intensity increases and the energy is redistributed in favor of higher-order components; the conversion coefficient increases. To a great extent the changes of the SRS parameters are determined by the decrease of the line width and growth of the cross section for ordinary Raman scattering of light with decreasing temperature. The formation of thin light-conducting filaments results in a change in distribution of the light intensity over the beam cross section and in additional growth of the intensity and coefficient of conversion into SRS.

IN our earlier paper[1] we investigated the connection between the stimulated Raman scattering of light (SRS) and self-focusing in carbon disulfide at different temperatures. On the basis of the experimental data obtained, it was suggested that an appreciable enhancement of the SRS intensity, observed in carbon disulfide when the latter is cooled, is due to two factors: The growth of the intensity of the ordinary Raman scattering of light, and the change in the light self-focusing regime, connected with the growth of the Kerr constant with decreasing temperature of the medium.

In the present investigation we measured the distribution of the energy among the components, the light-conversion coefficient, the distribution of the intensity in the cross sections of beams of different components, and the ratio of the SRS energy in the direction of propagation of the excited light, and in the opposite direction in calcite at different temperatures. Calcite is one of the few objects in which the SRS phenomenon prevails over other nonlinear effects. A comparison with the previously published data for CS_2, in which extraneous nonlinear effects are quite appreciable, makes it possible to obtain a qualitative idea of the influence of these effects on SRS.

1. DISTRIBUTION OF THE ENERGY IN THE SPECTRUM AND THE CONVERSION COEFFICIENTS IN SRS

The SRS was excited with the aid of a giant pulse from a ruby laser, of power 2×10^7 W. The experimental setup and the measurement procedure were described in[1]. In the present investigation we took additional precaution to stabilize the operation of the setup. To monitor the stability of the giant pulse, a fraction of the exciting light was diverted to a photodiode, and the signal was then applied to an S1-4 oscilloscope.

The measurements were made both photographically and photoelectrically. In the photographic measurements, the beam was focused on a thermocouple, the signal from which was fed to a mirror galvanometer M17/3. Light filters transmitting radiation of the required wavelength were placed in front of the receiver. The receiver and the galvanometer were calibrated with the aid of a power meter (IMO-1). In the investigation of the intensity distribution in the cross section of the scattered-light beams, the different SRS components were separated with the aid of a spectrograph with the slit removed. In many cases, the cross sections of the beams were photographed also through appropriate filters directly on photographic film.

The Q-switch was an alcohol solution of cryptocyanine. The exciting radiation was a single pulse of 15 nsec duration. Measurements with the aid of a Fabry-Perot interferometer with a gap of 15 cm between plates have shown that the main part of the radiation is concentrated in one structureless line. In addition, there are two other modes. We were unable to effect simultaneously single-mode and single-spike regimes. However, the additional modes in our setup had an intensity not exceeding 10% of the intensity of the total radiation. The mode composition of the radiation remained constant during the course of the experiments.

The investigated calcite single crystal measured $10 \times 10 \times 10$ mm. Two faces perpendicular to the optical axis of the crystal were polished. The crystal axis was oriented along the optical axis of the system. The scattering substances were placed in special cells, the construction of which made it possible to observe SRS at different temperatures. The cell is described in[1]. The radiation was focused in the central cross section of the scattering substance by a lens of focal length 250 mm. The beam cross section in the region of the focus was less than 1 mm. In the investigation of carbon bisulfide in[1], the cell was 20 mm long. Under the geometrical conditions realized in our setup, the variation of the beam cross section over the length of the cell or the crystal could be neglected, so that the region of the nonlinear interaction of the light with the medium coincided with the length of the scattering medium.

The intensity of the SRS components was obtained as a result of averaging 15–30 measurements. The rms measurement error was 15%. The investigations were carried out at a constant pump energy. The intensity of the exciting radiation was determined with the aid of a calorimeter that measured all the ruby-frequency radiation entering the cell.

In the investigation of the SRS phenomena, the main difficulty was the elimination of the influence of other nonlinear effects, viz., SMBS, stimulated Rayleigh-wing scattering, and self-focusing. These phenomena were also aggravated by the possible feedback between the investigated medium and the laser, due to reflection of the light by the windows of the cell or by the end faces of the crystal. Complete separation of these effects was difficult. In[2], the influence of the SMBS was excluded by using subnanosecond light pulses of duration shorter than the lifetime of the acoustic phonons. In our investigations we confined ourselves to systematic measurements of the radiation energy at the ruby frequency, going both in the direction of the exciting radiation ("forward") and in the opposite direction ("backwards"). For calcite, the energy of the backward radiation did not exceed 5% of the forward radiation, and this ratio remained unchanged under variation of the temperature, so that it could be assumed that the SRS in calcite was not complicated by either nonlinear effects or feedback. In the case of CS_2, the ruby-frequency radiation energy in the backward direction was about 40% of the forward radiation energy, so that a considerable connection with SMBS and other nonlinear phenomena could be assumed here. However, the ratio $I_{\text{forward}}/I_{\text{backward}}$ did not change with changing temperature, although the energy of the different SRS components increased appreciably with decreasing temperature, and the intensity of the exciting radiation passing through the cell decreased. It was shown in[1] that these effects are due mainly to changes in the self-focusing regime. Distorting action of other nonlinear effects cannot be excluded, but their role is apparently not very large.

Measurements of the SRS energy in calcite have shown that with decreasing temperature there are observed a general growth of the energy of the stimulated Raman scattering of the light, a redistribution of the energy in favor of the higher-order components, and an increase in the coefficient of conversion of the exciting light into SRS. The general character of variation of the energies of the individual components in calcite is the same as in CS_2[1]. The table lists the distribution of the energy in the SRS spectrum of calcite at different temperatures.

The maximum energy is carried by the first Stokes component. With decreasing temperature of the medium, saturation takes place of the first Stokes component, whereas changes of the energy of the second Stokes component and the two anti-Stokes components are largest in this temperature region.

Absolute values of the energy of the SRS components in calcite, in units of 10^{-6} J. Pump energy 0.3 J

Component.	t, °C				
	150	20	−20	−110	−196
1 St*	57000	68000	126000	197000	177000
2 St	3	8	17	16	590
1 aSt**	10	8	8	12	310
2 aSt	—	0.01	0.01	0.03	0,85

*Stokes components.

**Anti-Stokes components.

With increasing order of the component, its temperature threshold becomes more and more pronounced and the dependence of the intensity and the temperature become stronger. When the medium is cooled, the coefficient of conversion of the light into SRS also increases. The conversion coefficient was defined as the ratio of the total energy of all the observed SRS lines to the energy of the exciting radiation, and the energy of the SRS lines was measured both in the excitation direction and in the opposite direction. The results of measurements of the conversion coefficient for calcite at different temperatures are given below:

Conversion	150	20	−20	−110	−196
coefficient, %:	23	33	56	76	76

For comparison, we present data for carbon bisulfide

Conversion	20	−40	−75	−90
coefficient, %:	8	59	54	46

2. TEMPERATURE DEPENDENCE OF THE SRS INTENSITY

The authors of a number of recent publications ascribe the increase of the SRS intensity with decreasing temperature of the medium exclusively to the influence of self-focusing of the light on the SRS[3,4]. As we have already noted in[1], an appreciable contribution to the temperature dependence of SRS is made, apparently, also by the growth of the cross section of ordinary Raman scattering with decreasing temperature. Our investigations of spontaneous Raman scattering (SpRS) of carbon disulfide have shown that in the temperature interval from +20 to −100°C the integral intensity of the 656 cm^{-1} line increases by a factor of 3, whereas its width decreases from 0.9 to 0.7 cm^{-1}.

According to the data of Tulub and Bobovich[5], the integral intensity of the 1086 cm^{-1} line decreases by 30% when a calcite crystal is heated from 20 to 277°C. The width of this line, measured by Park[6] at room temperature, amounted to 1.1 cm^{-1}, and at 40°K—about 0.5 cm^{-1}. When the crystal was heated to 670°C, the width changed by 15 cm^{-1}.

An important factor in the excitation of SRS is the intensity of the corresponding SpRS at the maximum, i.e., the integral intensity divided by the width. The temperature dependence of the intensity of the 656 cm^{-1} of carbon disulfide and 1086 cm^{-1} of calcite at the maximum, obtained from the foregoing references, is shown in Fig. 1 dashed. The solid lines are the relative intensities of the first Stokes components of SRS of carbon disulfide and calcite.

A comparison of the plots shown in Fig. 1 indicates that a definite correspondence exists between them: the SpRS of carbon bisulfide, as well as the SRS, change with temperature much more strongly than SpRS and SRS of calcite. The second Stokes components therefore are the result of a successive excitation, or else of parametric interaction of components of lower order, and the temperature dependence of the intensity of the second Stokes components is determined to a considerable degree by the temperature dependence of the first components. Figure 2 shows the relative intensities of the second Stokes components of carbon disulfide and calcite at different temperatures. In the temperature interval from +20 to −100°C, the intensity of the second Stokes component of carbon disulfide increases by 2.5 orders of magnitude, and that of calcite increase by 1.5 orders.

Thus, when a medium with low Kerr constant (calcite) is cooled, the SRS energy increases and becomes redistributed in favor of the components of higher order. The coefficient of conversion of the exciting radiation into SRS increases. It should be noted that analogous changes in the SRS intensity were observed by Tulub et al.[7] and by Faĭzullov and co-workers[8] for radiation with different energy applied to the scattering medium. The authors obtained a monotonic increase of the SRS component intensity and of the conversion coefficient with increasing energy of the exciting radiation.

The temperature dependence of the SRS intensity of liquid carbon bisulfide has certain singularities, which apparently are connected with the self-focusing of light. In order to establish the connection between the temperature dependence of the SRS intensity and self-focusing, we have observed simultaneously with the intensity measurement, also the distribution of the intensity in the cross section of the scattered-light beam in the observation direction and in the opposite direction.

3. ANGULAR DISTRIBUTION OF THE INTENSITY OF THE SRS COMPONENTS

Investigations of the angular distribution of the intensity of the SRS components at room temperature in calcite and in carbon bisulfide were carried out a number of times[9-11]. According to these investigations, the intensity distribution of the Stokes and anti-Stokes components over the cross section of the beams in calcite single crystals, at an optical-axis orientation along the direction of propagation of the exciting light, satisfied the phase-synchronism conditions[12]. Recently, Ataev and Lugovoĭ[11] observed and explained theoretically the additional radiation of the anti-Stokes components when the optical axis of the crystal is oriented at certain angles to the exciting radiation. In carbon disulfide, an angular distribution of the intensity of class II was observed, and its occurrence was attributed to the self-focusing of the light. The number of investigations of SRS in the direction opposite to the propagation of the exciting light is quite small.

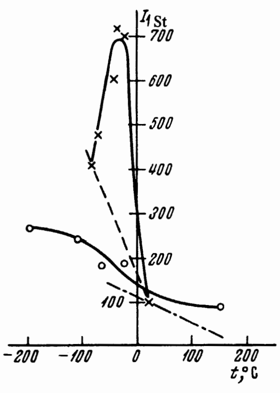

FIG. 1

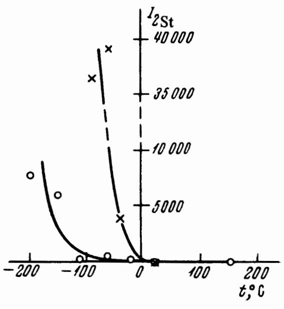

FIG. 2

FIG. 1. Temperature dependence of the intensity of the first Stokes component of SRS of carbon disulfide (×) and calcite (○), and of the corresponding SpRS of carbon bisulfide (dashed) and calcite (dash-dot).

FIG. 2. Temperature dependence of the intensity of the second Stokes component of carbon disulfide (×) and calcite (○).

Among the recent papers, notice should be taken of the article by Shen and Shaham[13], who measured the SRS and SMBS power in the "forward" and "backward" directions as functions of the exciting-radiation power in toluene at 20 and 80°C. It is shown that the ratio of the "forward" and "backward" radiation powers depends on the power of the exciting radiation. The radiation power in either direction is higher at 20°C than at 80°C.

In our experiments, the optical axis of the calcite crystal was oriented along the direction of propagation of the exciting light. The observed distribution of intensity of the second Stokes and anti-Stokes components in the cross sections of the beams obeyed the conditions of phase synchronism. In addition, at the center of the ring of the radiation of the second Stokes component we observed scattering along the axis, apparently connected with the consecutive excitation of the second Stokes component. The cross section of the backward-scattered light beams had the form of diffuse spots.

Measurement of the ratio of the intensities in the direction of propagation of the exciting light and in the opposite direction has shown that for the first Stokes components the ratio $I_{\text{forward}}/I_{\text{backward}} = 5$, while for the second Stokes component this ratio is equal to 4. When the temperature decreases, the intensity of the SRS increases over the entire section of the beam, but the angular distribution of the intensity and the ratio of the "forward" and "backward" intensities remained unchanged. In carbon disulfide in the propagation direction of the exciting radiation, we observed scattering of "class II"[1]. In the investigated angle interval (3.5°) we observed no noticeable changes in the intensity distribution over the SRS beam cross sections at different temperatures, whereas the distribution of the intensities over the "backward" radiation beam cross sections changed appreciably. Thus, in the temperature interval from −40 to −70°C, the cross section of the unshifted component and of the Stokes components of the "backward" scattering had the form of distinct points. Apparently, thin light-conducting filaments were produced in the carbon disulfide in this temperature region. This temperature region corresponded to maxima in the intensities of the first Stokes and anti-Stokes components, i.e., the formation of thin light-conducting filaments in the medium led to an increase of the SRS intensity. With further decrease of the temperature, the structure of the beam changed and the filaments vanished.

Inasmuch as the self-focusing of the light is connected with the Kerr constant, which generally speaking depends on the temperature, it can be assumed that the change of the self-focusing regime in liquid carbon disulfide is connected precisely with the change of the Kerr constant. It would be of interest to compare the temperature dependence of the Kerr constant and the energy characteristics of the SRS. However, at the present time there are only static measurements of the

Kerr constant in carbon disulfide at different temperatures. According to these data, when the temperature changes from +20 to − 60° C, the Kerr constant increases by 80%. It is not excluded that when giant light pulses are applied, a role is played both by the high-frequency Kerr constant, which is connected with the electron polarizability, and by the Kerr constant connected with the orientation of the molecules in the liquid. It is possible that the thin light-conducting filaments in the carbon disulfide are produced in the temperature region where both processes are sufficiently intense. In the case of deeper cooling of the medium, the viscosity begins to increase and the orientation processes become hindered. Indeed, other measurements of the relaxation time of the carbon disulfide molecule, as determined from the widths of the depolarized lines, have shown that in the temperature interval from +20 to − 100° C the relaxation time of the molecule increases by a factor 5—6. This can lead to a change of the Kerr constant as a result of the decreased role of the orientation processes, and to a vanishing of the thin light-conducting filaments at low temperatures.

CONCLUSIONS

Investigations of the SRS of carbon disulfide in calcite, substances with essentially different Kerr constants, have shown that in either case an increase of the SRS components and a redistribution of the energy in favor of the components of higher order is observed with decreasing temperature. The coefficient of conversion of light into SRS also increases.

A comparison of the temperature dependence of the SRS intensity with the temperature dependence of the width and intensity of the corresponding SpRS oscillation has shown that the change of the SRS intensity is due mainly to a narrowing of the line and increase of the SpRS probability with decreasing temperature.

Self-focusing of light and carbon bisulfide leads to a change in the distribution of light over the beam cross section and to additional growth of the intensity of the components and of the coefficient of conversion into SRS. When thin light-conducting filaments appear in the carbon disulfide, the intensity of the first Stokes component and the coefficient of conversion into SRS are maximal.

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