Stimulated Raman scattering and self-focusing of light in substances with different Raman cross sections

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Self-focusing of the first Stokes component of SRS and of the exciting light is investigated in substances in which the Kerr constant and the Raman cross section are appreciably different. SRS was induced by a giant pulse from a single-mode ruby laser. Quantities that characterize the evolution of the SRS and the self-focusing of the light in the substances are compared (e.g., the Kerr constant, gain, efficiency of conversion of the laser light into SRS, power density at the minimum-diameter self-focusing points, etc.) The causes of the similarities and differences in the development of self-focusing in substances with appreciably different Kerr constants are discussed.

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The results of theoretical and experimental investigations of the propagation of laser radiation in substances with large Kerr constants have shown that the change produced in the refractive index of light by the orientational Kerr effect leads under the influence of a strong electric field to the appearance of regions in which light with high-power density is concentrated. This phenomenon has been called self-focusing of light. The role of the stimulated Raman scattering (SRS) in this phenomenon has remained unclear. It was customary to assume that in substances with large Kerr constants an increase of the laser-radiation power density, due to its self-focusing, leads to a "jump" in the SRS energy (E_R) by several orders of magnitude ($\sim 10^5$), whereas the laser-radiation energy (E_L) changes only by several percent. It was assumed that SRS is produced and propagates in the self-focusing filaments of the laser radiation.[1–3]

In 1970 we have observed self-focusing of SRS of light in liquid nitrogen, a substance with a small Kerr constant.[4,5] In our subsequent studies we have shown[6–8] that in substances having a large probability of Raman scattering (RS) of light and different values of the Kerr constant, the SRS undergoes self-focusing. The SRS produces in this case focal regions that are autonomous with respect to the exciting radiation. These results were subsequently confirmed by other workers.[9–11]

We present in this paper the results of an investigation of the self-focusing of the first Stokes component of SRS and of laser light for a large group of substances with different optical characteristics. These substances were chosen such that the Kerr constant and the effective SRS cross section were varied in a wide range. A comparison was made of the quantities characterizing the evolution of the SRS and of the self-focusing of light in different substances (Kerr constants, gains, relative SRS thresholds, etc.). The following substances were investigated: CS_2, $C_6H_5NO_2$, C_6H_6, $CHCl_3$, N_2 $CaCO_3$, C_6H_{12}, CCl_4, CH_3COCH_3.

1. EXPERIMENTAL SETUP AND MEASUREMENT PROCEDURE

The optical system of the experimental setup is shown in Fig. 1a. The SRS was excited by a giant pulse of ruby laser *l*, which emitted a single longitudinal mode. The laser emitted a single pulse of 20 nsec duration and 30 MW maximum power. The distribution of the intensity of the exciting radiation in the far field was nearly Gaussian. The laser radiation was focused into the cell with the medium by lens L_1 of focal length 250 mm; in a number of experiments, the SRS was excited by an unfocused beam. In this case, to decrease the beam diameter, the telescopic system AF was used. Feedback was prevented by placing an optical delay in the propagation path of the exciting radiation between the laser and the cell with the scattering medium, and by using cells with skewed windows. The energy of the exciting radiation was varied by means of calibrated neutral light filters F_4 and was measured with the aid of the IMO-2 instrument or with photodiodes Edgerton SGD_{100} (PM), while the mode content of the radiation was moni-

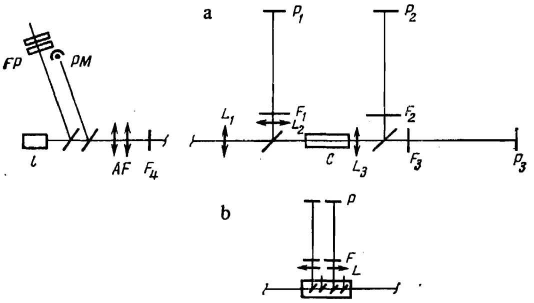

FIG. 1. a) Optical system of the experimental setup: l—ruby laser, FP—Fabry-Perot etalon, PM—photodiode, AF—telescopic system, F_4—neutral light filters, L_1, L_2, L_3—lenses, F_1, F_2, F_3—selective light filters, P_1, P_2, P_3—photographic plates, C—cells with investigated medium. b) Cell for the investigation of the structure of the beam in planes situated at various distances from the entrance window. L—lenses, F—light filters, P—photographic plates.

tored with the aid of a Fabry–Perot (FP) etalon. The structures of the SRS and laser beams were photographed in a plane near the exit window of the cell C inside the medium, with tenfold magnification, on photographic plates of the type Kodak 1N or Infrarot-Platte (P_1, P_2, P_3). The radiation was projected on the photographic plates by lenses L_2 and L_3. The resolving power of the system was 10 μ. The spectral separation of the laser radiation and of the SRS components was with the aid of selective light filters F_1, F_2, F_3 or with a spectrograph without a slit.

Figure 1b shows the cell used to investigate the beam structure in planes situated at different distances from the entrance window of the cell. The radiation was extracted from the cell with the aid of glass plane-parallel plates oriented at an angle to the cell axis, and was then projected by lenses L on the photographic plates P.

The distributions, over the beam cross section, of the intensity of the first Stokes component of the SRS and of the exciting radiation inside the cell with the scattering medium was investigated at different excitation energies, from the SR threshold to the region of the SRS saturation. The energy of the first Stokes component of the SRS at saturation and the energy of the exciting radiation incident on the scattering medium were measured with the aid of Edgerton SGD_{100} photodiodes, the signals from which were fed to sensitive galvanometers.

2. EXPERIMENTAL RESULTS

The quantities characterizing the evolution of the SRS and of the self-focusing of light in the investigated media are listed in Table I: ν—frequency of the molecular vibration excited in the case of SRS of light; B_0—Kerr constant[14,16]; E_R/E_L—ratio of SRS energy at saturation to the exciting-radiation energy; g—gain calculated from the effective RS cross section (the effective cross sections were obtained from the results of a number of studies[16–20]; P_R—SRS threshold (in relative units); E_R—change of SRS in joules, corresponding to the region of the "jump" on the $E_R(E_L)$ curves; P_c/P_l—ratio of the threshold powers of the SRS for the exciting radiation with circular and linear polarization[21,22]; change of polarization ellipse—change of polarization ellipse of the laser radiation when the excitation energy is changed from threshold to SRS saturation.[22] In addition, the table lists the characteristic distribution of the intensity in the cross section of the exciting-radiation and first Stokes SRS component beams near the SRS threshold and above the threshold (close to the SRS saturation) in the plane of the exit end face of the cell inside the medium; P/S—SRS power density in the self-focusing regions having the minimum diameter.[7]

In accordance with the results given in the table, the investigated substances can be divided into three groups:

Group A: (CS_2, $C_6H_5NO_2$, C_6H_6, $CHCl_3$) substances with large Kerr constants and large ratio E_R/E_L.

Group B: (N_2, $CaCO_3$) substances with small Kerr constants and large ratio E_R/E_L.

Group C: (C_6H_{12}, CCl_4, CH_3COCH_3) substances with small Kerr constants and small ratio E_R/E_L.

TABLE I.

	Group A				Group B		Group C		
	CS_2	$C_6H_3NO_2$	C_6H_6	$CHCl_3$	N_2	$CaCO_3$**	C_6H_{12}	CCl_4	CH_3COCH_3
ν, cm^{-1}	656	1345	992	667	2330	1086	2852	459	2921
B_0, 10^9 cgs esu	418	290	40	18	6	—	4.1	5.1	14
E_R/E_L	$8 \cdot 10^{-2}$	$3 \cdot 10^{-1}$	$5 \cdot 10^{-2}$	10^{-1}	$5 \cdot 10^{-1}$	$3 \cdot 10^{-1}$	10^{-2}	10^{-2}	10^{-3}
g, 10^{-2} cm/MW	1.9*	3.4	0.44	0.03	2.1*	2.3	0.17	0.13	0.14
P_R	1	1.25	3.6	20	1.6	0.3	≥50	≥50	≥50
ΔE_R, J	$>10^4$	10^5	$>10^2$	—	10^4	10^7	—	—	—
P_c/P_l	1.9	1.7	1.6	—	1	—	1	1	1
Change of polarization ellipse	Appreciable change				No change		Very small change		
Intensity distribution									
Near threshold ER***	No change (just as below threshold)								
Near threshold SRS	One or two intense spots 100–300 μ.								
Above threshold ER	Absorption regions 100μ Self-focusing regions 10μ						No change (just as below threshold)		
Above threshold SRS	Self-focusing regions 10μ			25μ	10μ		100–300μ		
P/S, 10^{-3} MW/cm^2	100	100	—	—	10	6	—	—	—

*Experimentally confirmed values.[12,13]
**Data for $CaCO_3$ at −196 °C.
***ER—exciting radiation.

At an excitation energy greatly exceeding the threshold, the contrast between the points and the background in the cross section of the SRS beam decreases as a result of the saturation.

The distribution of the light intensity in the SRS beam cross section, observed at various points inside the cell, confirm the results listed in the table for the exit plane of the cell. The evolution of the SRS beam was straight as it passed through the cell. Large regions in the SRS beam section at the start of the cell are converted into bright and small points at the exit from the medium in substances of type A and B. The diameters of these points are determined by the maximum resolution of the system and equal 10 μ. No points with this diameter were observed in substances of type C.

As seen from the table, substances of type A have certain common properties with substances of type B, namely a large ratio E_R/E_L, a large gain, a low SRS threshold, and the presence of a jump on the $E_R(E_L)$ curves. Above the SRS threshold, in substances of types A and B, self-focusing points are observed in the cross section of the SRS beam, with diameter ~10 μ, as well as absorption regions in the section of the laser-light beam, the intensity in which decreases practically to zero. It is important that distinct self-focusing points with dimensions 10 μ are observed at the centers of certain absorption regions.

However, certain properties of substances of type A and type B are different, for example, there is a strong difference between the values of the Kerr constants, the power density at the points of self-focusing with minimum diameter is lower in substances of type B, the ratio of the SRS thresholds for the circularly and linearly polarized exciting radiation is not the same, and the change of the polarization ellipse of the laser light in the region of the SRS energy jump is different.

It should be noted that the properties that are different for substances of type A and B are common to substances of types B and C (with the exception, of course, of the power density at the minimal-diameter self-focusing points).

There exist, however, differences between substances of type B and C: the gain is less in substances of type C, for which no jump regions on the $E_R(E_L)$ curves are observed,[23] and in substances of type C the diameter of the SRS spots is the same above the threshold as near the threshold (~200 μ).

3. DISCUSSION OF RESULTS

When comparing the results of investigations of the SRS and self-focusing of light in the substances given in the table, the following questions arise:

1. Why is the self-focusing picture in substances of types A and B similar, despite the appreciable difference between the Kerr constants? Yet no self-focusing points of 10 μ diameter arise in substances of type C, where the Kerr constant is practically the same as in substances of type B.

2. What causes the differences between certain properties (the power density in the self-focusing regions P/S, the ratio P_c/P_l, the change in the polarization ellipse) of substances of classes A and B when there is no difference in the character of the evolution of the self-focusing?

3. What causes the appearance of absorption regions with self-focusing points sometimes located at the center in the section of the exciting-radiation beam passing through the substance, which are observed simultaneously with the self-focusing points in the section of the beam of the first Stokes component of the SRS?

The following explanations can be suggested.

In our preceding investigation,[4,5] it was shown that the self-focusing of SRS, especially in media with small Kerr constants, is caused by processes connected with the SRS effect itself. The possibility of self-focusing of light when the polarizability of the molecules is changed by excitation was indicated in[24,25]. The nonlinear increment to the refractive index, due to the excitation of the SRS, was estimated theoretically in[26,27]. The results of these estimates show that the change of the refractive index upon excitation of the SRS can be of the same order as in the optical Kerr effect, and in substances with small Kerr constants it is much higher. In substances of types A and B, despite the appreciable difference between the Kerr constants, the coefficient of conversion of light into SRS upon saturation and the gain are very large. In these media, self-focusing points of 10 μ diameter are observed, as well as regions of absorption in the section of the exciting-radiation beam. At the same time, in substances of type C, where the coefficient of conversion into SRS and the gain are small, no points of 10 μ diameter are observed.

If the SRS is the cause of the self-focusing in substances of type B, then the ratio P_c/P_l in these substances can differ from the value of this ratio in substances of type A, where an appreciable contribution to the change of the nonlinear refractive index is introduced by the optical Kerr effects. For the same reason, the results can differ also for substances of types A and B when measurements are made of the changes in the polarization ellipse of the laser radiation and in the SRS power density at the self-focusing point. It should be noted that in the case of substances of type A (carbon disulfide or nitrobenzene), the energy and power density at the points of the self-focusing of the SRS light and of the exciting radiation are close in order of magnitude to those calculated theoretically with allowance for the Kerr effect[28] and to the experimental estimates of the energy at the points of the self-focusing of the exciting radiation.[24,29] In substances of type B, the power density at the points of the self-focusing of the SRS of light is much larger than that expected in the case when the principal mechanism that leads to the nonlinearity of the refractive index is the quadratic Kerr effect.

The absorption regions (sometimes with a bright point at the center) in the section of the laser-light beam can be attributed to competition between processes due to the increase of the intensity as a result of self-focusing of the laser radiation and the decrease as a result of conversion into SRS.

The jump of the SRS intensity is not connected with the Kerr self-focusing of the pump, since in substances of class A and B the jump is observed at close values of E_L. Nor is the difference between the parameters of the SRS propagating "forward" and "backward" a direct consequence of self-focusing.[4] When the diameter of the SRS self-focusing regions decreases the divergence decreases in "backward" SRS, and the brightness of the light flux increases, in contrast to "forward" SRS. At a definite pump energy and a definite layer thickness, in the presence of self-focusing regions, the distribution of the intensity in the far field and the divergence of the SRS "backward" and the divergence of the exciting light are close.[4] That is to say, in the case of "backward" SRS, in spite of the occurring optical inhomogeneities in the medium, the wavefront of the SRS duplicates the wavefront of the pump. It is interesting that the results obtained[29] for SMBS are analogous to our results for SRS. This seems to indicate that the physical natures of the observed phenomena are close.

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