Self-focusing of the higher Stokes and anti-Stokes SRS components in liquid carbon disulfide

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New data are reported on the development of self-focusing, the variation in spectral composition, and the spatial distribution of Stokes and anti-Stokes SRS components in liquid carbon disulfide as functions of the energy of the exciting radiation. It is shown that, at low pump energies, there is self-focusing of the first Stokes components, and emission cones are observed for the higher Stokes and anti-Stokes components. At energies above a certain definite value, SRS becomes time-dependent and spectral broadening of the order of a few hundred cm^{-1} is observed at self-focusing points for all the SRS components. The results are explained in terms of the theory of propagation of a light beam in a nonlinear medium in the form of a multifocus structure.

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Self-focusing[1-3] and the attendant variation in the spectral and spatial characteristics of radiation in liquid carbon disulfide have been investigated both experimentally and theoretically. Considerable spectral broadening (up to a few hundred cm^{-1}) has been observed by a number of workers for the laser radiation transmitted by a material and for the first Stokes component of SRS.[4] In carbon disulfide, SRS is known to be accompanied by the appearance of ultrashort pulses[5] which, like the considerable spectral broadening, were initially associated with the appearance of light-guiding "filaments" in the medium[6] and, subsequently, with the motion of focal regions of the multifocus structure of the light beam.[3] However, despite the large number of published papers, many aspects of the propagation of a laser beam through a nonlinear medium and, especially, the relation between self-focusing and stimulated scattering (SRS, SMBS) have so far remained somewhat controversial. In particular, there are practically no data on the effect of self-focusing in carbon disulfide on the development of SRS, including the higher-order Stokes and anti-Stokes components.

In this paper, we report new data on self-focusing, spectral composition, and spatial distribution of Stokes and anti-Stokes components of SRS in carbon disulfide as functions of the energy of the exciting radiation. The spatial and spectral distributions of SRS components were observed simultaneously on planes inside the medium at different distances from the exit window of the cell. The spectral and angular distributions of SRS components outside the medium were also observed under the same experimental conditions.

The source of the exciting radiation was a ruby laser with a single-mode amplifier, and also a ruby laser generating two longitudinal modes of roughly equal intensity and a separation of $0.01\ \mathrm{cm}^{-1}$. The giant-pulse energy was varied between 0.008 and 0.5 J with the aid of a set of calibrated neutral light filters. An optical delay system was used to reduce feedback between the cell containing the medium and the laser. To prevent feedback within the cell itself, the windows of the cell were slanted or the cell was placed at an angle to the beam of exciting radiation. The exciting radiation was focused inside the cell containing the medium by a lens of long focal length ($f = 420$ mm). The maximum power density at the focus of the lens was $200\ \mathrm{MW/cm}^2$. Self-focusing and spectral broadening of the SRS components were observed with the aid of the STÉ-1 spectrograph. The intensity distribution on different planes inside the cell containing the medium was projected onto the slit of the spectrograph with a fivefold magnification. The resolving power of the system was 10 μ, the depth of definition was $\lesssim 1$ mm, and the linear dispersion of the spectrograph between 6000 and 9000 Å was 12.8 Å/mm.

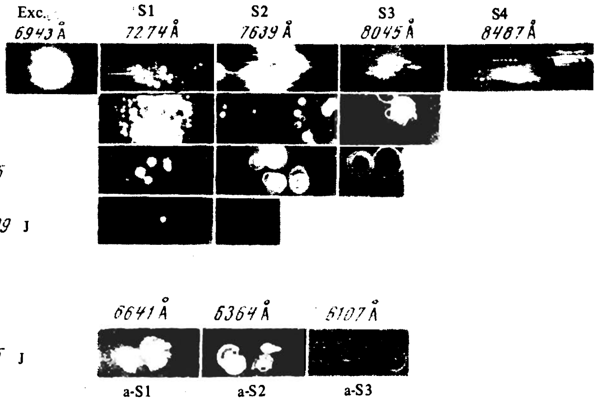

FIG. 1. Intensity distributions of the exciting radiation, four Stokes components, and three anti-Stokes components of SRS in the case of carbon disulfide at different excitation energies.

The intensity distribution on a plane near the end of the cell is shown in Fig. 1 for different excitation energies. The figure shows the exciting radiation transmitted by the medium, the first, second, third, and fourth Stokes components and three anti-Stokes components of SRS. Self-focusing points were not observed in the cross section of the beam of exciting radiation at the exit window of the cell. This may be connected with the large value of the SRS transformation coefficient for the exciting radiation. Near the SRS threshold (0.009 J), there is one self-focusing point in the field of the first Stokes component, which corresponds to rings in the case of the second and third Stokes components.[1)] Figure 1 shows only the ring corresponding to the second Stokes component because the intensity of the ring associated with the third Stokes component is too low to be seen on the photographic plate. As the excitation energy is increased, the cross sections of the Stokes and anti-Stokes beams contain a large number of different rings, including a system of concentric rings. When beams of the SRS components and of the unshifted component near the entrance to the cell were examined in the backward direction, there was clear evidence of self-focusing points but there were no rings.

The number of self-focusing points in the case of the first Stokes component, and the diameter and number of rings corresponding to higher order components observed in the forward direction, were found to depend on the energy of the exciting radiation. As the pump energy was increased, the number of self-focusing points across the beam cross section in the case of the first Stokes component was found to increase, and their size to decrease. There was a corresponding increase in the number, and a reduction in the diameter of rings, corresponding to the higher Stokes and anti-Stokes components. At a certain particular excitation energy, the rings corresponding to higher order components were found to disappear and were replaced by points. Further increase in the pump energy ensures that each point corresponding to the SRS components spreads over the spectrum to a band of a few tens or hundreds of cm^{-1}.

Figure 2 shows graphically the width of the spectral distribution at the self-focusing points corresponding to different components as a function of pump energy. The spectral broadening appears when the excitation energy reaches the threshold value (0.08—0.09 J when the thickness of the scattering layer is 50 mm) for practically all the observed components. In the case of the first Stokes component, the broadening extends preferentially into the anti-Stokes region, whereas for the higher-order components, this occurs into the Stokes region. The results obtained with a laser generating one and two longitudinal modes show no qualitative difference.

The contour of the first Stokes line consist of a strong central part and low-intensity wings. High resolution observations of the spectral distribution of the first Stokes component of SRS shows that the strong part of the profile has a complicated structure consisting of individual components of width roughly equal to 0.1 cm^{-1} (width of the instrumental function of the spectrometer).2) The separation between the components is 0.21 cm^{-1}, which corresponds to the separation between the SMBS components in liquid carbon disulfide. The resultant width of the strong part of the profile is 1.6 cm^{-1}, i.e., somewhat greater than the linewidth of the completely symmetric oscillation at 656 cm^{-1} of carbon disulfide in the ordinary Raman effect. The weaker wings provide a continuous background on the interference pattern.

To estimate the angle of the cones which produce rings inside the medium (Fig. 1), the intensity distribution at different depths within the absorbing cell was projected onto the plane of the slit of the STÉ-1. The energy of the exciting radiation was held constant and near the threshold. Several rings (between one and four) with only slightly different diameters were obtained in the plane of the photographic plate for a given SRS component. The planes inside the cell, which were projected onto the plane of the slit of the STÉ-1, were 1.5 mm apart. The ring diameters were measured on the photographic plate and a graph was plotted of this diameter as a function of the position of the plane of observation. Finally, a calculation was made of the angle of the cone of radiation corresponding to the given component. The angle obtained in this way for the second Stokes component was 0.032 rad (corrected for the refractive index).

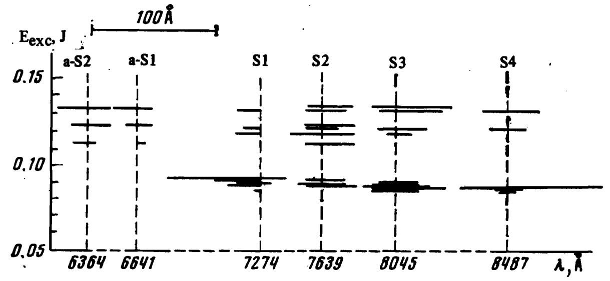

FIG. 2. Spectral width at self-focusing points corresponding to the SRS components in carbon disulfide for different excitation energies.

The overall angular distribution of the SRS components can also be observed in the focal plane of a collecting lens. This method was used to investigate the angular distribution of SRS components in all the previous papers.[11,12] The cone angle corresponding to the second Stokes component, which we obtained from the resultant angular distribution, was 0.035 rad (in air), which agreed to within experimental error with the angle corresponding to class 2 emission.[12] Comparison of the results shows that the radiation cones observed inside the medium have roughly equal angles at the apex, which agree with the cone angle in the resultant angular distribution of the given component to within experimental error.

The appearance of a large number of rings of different diameter in sections through cones of equal angle at excitation energies of between 0.01 and 0.05 J for the higher Stokes and anti-Stokes components of SRS suggests that the cones belong to one type of radiation (class 2) and originate at definite points of space that do not lie in a single plane inside the cell containing the medium.

At excitation energies near the threshold, the centers of the rings belonging to the higher Stokes and anti-Stokes components show a correspondence with the self-focusing points of the first Stokes component. This suggests that the cones originate in regions of self-focusing. The appearance of the systems of concentric rings shows that the regions of self-focusing that produce the radiation cones may lie on a single axis. This picture is consistent with the theory describing the propagation of a light beam in a nonlinear medium in the form of a multifocus structure.[3] The apices of the radiation cones corresponding to the higher-order components may be associated with the foci of the structure which appears in the first Stokes component when SRS is quasistationary. As the pump energy increases, the ring diameters corresponding to the SRS components are found to decrease until, in a particular cross section through the cell, there are independent focal regions for each component. This reduction in diameter may be connected with the approach of the voci in the multifocal structure of the first Stokes component of SRS to the exit plane of the cell. When the excitation energy approaches a definite threshold value, there is considerable spectral broadening of the components (see Figs. 1 and 2), which probably indicates the appearance of ultrashort SRS pulses[5] and of an independent multifocal structure in them. The pulse length estimated from the observed broadening is 10^{-12}—10^{-13} sec. The characteristic asymmetry of the broadening and, in some cases, the quasiperiodic structure of the observed spectral broadening (see Fig. 1) can be explained by the different rates of motion of the focal regions and the interference of radiation originating in different focal points.[3]

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[1] The self-focusing of the first Stokes component of SRS has frequently been observed in carbon disulfide and in a number of other media. [7-10]

[2] The dispersive system was a Fabry-Perot etalon, 1 mm thick, used in conjunction with the ISP-51 spectrograph.

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