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Investigation of the Angular Distribution of the First Stokes Component for Stimulated Raman Scattering

A. N. Arbatskaya, K. A. Prokhorov, and M. M. Suschinskiĭ

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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The angular distribution of the first Stokes component of stimulated Raman scattering is investigated in a number of substances for various excitation conditions. It is shown that the stimulated Raman scattering angular distribution approximately "repeats" the angular distribution of the exciting radiation. Some possible causes of the repetition effect are discussed.

INTRODUCTION

INVESTIGATIONS of the angular distribution of SRS are of great interest for the explanation of the nature of this phenomenon. In such investigations, greatest attention is paid to radiation rings at anti-Stokes and higher Stokes frequencies, to which many investigations have been devoted. The angular distribution of SRS radiation propagating near the axis of the exciting light beam has not yet been sufficiently well investigated. Garmire's paper[1] contained individual remarks concerning this distribution. More detailed investigations[2,3] have shown that the angular distribution of the first Stokes component of SRS is characterized by a sharply pronounced forward directivity. When ruby radiation is focused inside a cell with the investigated substance, practically the entire indicated radiation is concentrated near the geometrical cone of the exciting-light rays emerging from the cell.

In the present paper, which is a continuation of[2,3], our task was a more systematic investigation of the angular distribution of the first Stokes SRS component with allowance for a number of factors capable of influencing this distribution.

EXPERIMENTAL PROCEDURE

The light source for the excitation of the SRS was a ruby laser. The SRS was excited in most experiments with unfocused beams. To investigate the influence of different operating regimes of the laser on the angular distribution of the first Stokes component of the SRS, the investigations were carried out with two different setups. In the first we used a laser in which the pulse Q-switching was produced with a rotating total-internal-reflection prism. In this case the laser radiation contained several axial and angular modes. The radiation power directly ahead of the cell with the investigated liquid ranged from 10 MW for benzene to 32 MW for acetone. In the second setup, the Q-switching was with a cryptocyanine solution in ethyl alcohol. The emission spectra of such a laser contained one axial mode and a large set of angular modes. Such a radiation has made it possible to illuminate uniformly a diaphragm with diameter up to 10 mm, which was placed directly ahead of the input window of the cell with the investigated liquid. A number of measurements were performed with a single-mode laser. The diagram of the setup for this case is seen in Fig. 1. The selection of the transverse modes was by means of diaphragm 6 of 1–1.2 mm diameter, depending on the quality of the ruby. The selection of the longitudinal modes was by two plane-parallel glass plates that served as a compound output mirror 7. TEM_{00} modes were obtained in this case by adjusting the resonator.

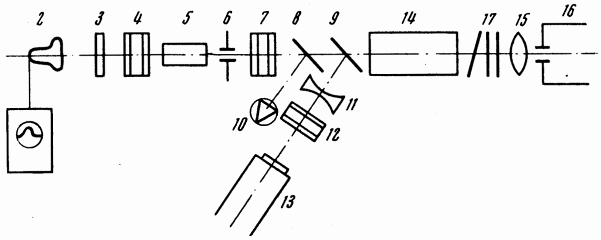

FIG. 1. Experimental setup with laser working in the single-mode regime: 1–12-7 oscilloscope, 2–FÉK-09 coaxial photocell, 3–dielectric mirror (100% reflection at λ = 6943Å), 4–cell with bleachable filter, 5–ruby rod, 6–diaphragm, 7–pair of plane-parallel glass plates, 8, 9–rotary plates, 10–calorimeter, 11–scattering lens, 12–Fabry-Perot etalon, 13–chamber, 14–cell with substance, 15–lens with f = 120 mm, 16–spectrograph, 17–set of filters for attenuating the SRS.

The laser parameters in the single-mode regime were as follows: maximum energy 0.032 J, pulse duration at half height 30 nsec, diameter of beam leaving the resonator ~ 1 mm. The SRS radiation was gathered with the aid of lens 15 in the plane of the slit of spectrograph 16 (the slit was removed during the operation). The investigated liquid was in cells ranging in length from 10 to 40 cm. The cells had exit windows that were inclined 5–10° to the axis. This excluded the possibility of SRS generation as a result of light reflection from the cell windows. To prevent the radiation emerging from the cell from being reflected back from the lens or from other elements of the optical scheme, neutral and selecting optical filters were placed immediately behind the cell at an inclination to the axis. It was verified experimentally that there was no feedback between the cell and the laser.

The angular divergences of the exciting radiation and of the first Stokes component of the SRS were measured by photometry of the obtained photographs. The density on the photographic plates was converted into intensity with the aid of a density scale produced by illuminating a stepped attenuator either by light from a ruby laser or by the SRS light.

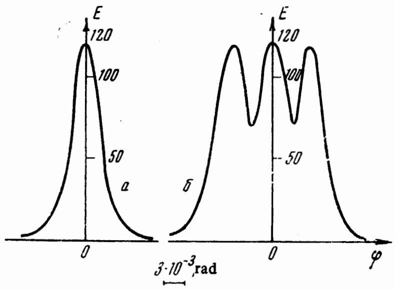

FIG. 2. Angular distribution of the radiation of the first SRS Stokes component in acetone following excitation by a multimode laser: a—perpendicular to the arrangement of the small spots of the structure, b—along the arrangement of the small spots. The energy units are arbitrary.

MEASUREMENT RESULTS

We have studied the influence exerted on the angular distribution of the SRS by such factors as self-focusing, the mode composition of the exciting radiation, and the conditions of SRS excitation.

The influence of self-focusing on the SRS phenomenon is by now universally known. To trace the influence of self-focusing on the angular distribution of the SRS, we have carried out parallel investigations with several substances in which there is self-focusing (carbon disulfide, benzene), and substances in which under our conditions there was apparently no excitation of the self-focusing (acetone, calcite crystals). In addition, we investigated a mixture of 92% acetone and 8% carbon disulfide. Self-focusing does take place in this mixture, as is very clearly manifest in the structure of the anti-Stokes scattering rings, which are much more diffuse and broad in comparison with pure acetone[4,5]. By comparing the angular distribution of the first Stokes component of SRS in pure acetone (3020 cm^{-1} line) with the same line in the mixture of acetone with carbon disulfide, it was possible to trace the influence of self-focusing on the investigated angular distribution.

A qualitative comparison of the photographs of SRS in substances with and without self-focusing has shown that there is no strong difference between them. The widths of the angular distribution for the exciting radiation past the cell and for the first SRS stokes component in the substances investigated by us turned out to be close. To be sure, when a laser with several axial modes was used, the pattern of the angular distribution of the first SRS stokes component became more complicated. Namely, the scattering spot was split in many cases into several smaller spots, usually disposed along the spectrum. The total width of this complex distribution exceeds by 10—15 times the width of the ruby-radiation distribution. In the direction perpendicular to the arrangement of the small spots (along the diameter of the individual spots), the widths of the angular distribution of the SRS and of the ruby radiation differ less noticeably. As an illustration, Fig. 2 shows the distributions obtained for acetone.

A quantitative comparison of the angular distribution of the ruby radiation passing through the cell and of the first SRS Stokes component, as indicated above, was carried out by photometry of the photographs. The photometry was carried out in the same direction on the photographic plate, for example, perpendicular to the line joining the small spots of the structure. The obtained data are listed in Table I. They confirm the qualitative conclusions drawn above, that self-focusing does not influence greatly the width of the angular distribution of the SRS Stokes component.

Table I. Widths of angular distribution (at half height) of the laser radiation past the cell and of the radiation of the first SRS Stokes component.

Substance	Δφ, 10^{-3} rad	
	Exciting radiation	First Stokes component
Acetone	2,4	5.6
Acetone with carbon disulfide	3,2	6.7
Benzene	3,2	6,7

*Cell length 30 cm, photometry perpendicular to the spectrum.

FIG. 3

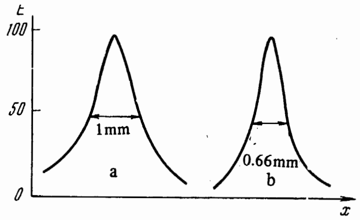

FIG. 4

FIG. 3. SRS spectrum in benzene excited by a laser with one longitudinal and many transverse modes (ER—exciting radiation, 1St—first Stokes component). The photography was with a spectrograph without an input slit. Cell length 10 cm. The densities on the photographic plates were equalized with the aid of optical filters.

FIG. 4. Distribution of radiation intensity for benzene excited with a single-mode laser. a—first Stokes component of SRS, b—ruby radiation passing through the cell (dimensions of the spots on the photographic plate are indicated).

We have already indicated above that the presence of several axial modes in the radiation causes a complication of the angular distribution of the first SRS Stokes component. To ascertain the influence of the transverse modes in the excited radiation, we have compared the photographs obtained in the multimode and in single-mode regimes of the ruby laser operation. The ruby excitation conditions were first chosen such that the presence of a large number of transverse modes and their mutual overlap would cause the radiation from the ruby to have the most uniform possible distribution of intensity over the cross section. In such a laser regime, possible distribution of intensity over the cross section. In such a laser regime, SRS was excited in benzene in cells 100 and 400 mm long. In the former case, approximately 1% of the exciting radiation was converted into the first Stokes component, i.e., the excitation was carried out near the SRS threshold. In the latter case, the intensity of the first Stokes component was close to the intensity of the ruby radiation emerging from the cell.

Table II. Widths of angular distribution (at half height) of laser radiation past the cell and of the radiation of the first Stokes component of SRS in benzene for single-mode laser operation

Photograph	$\Delta\varphi$, 10^{-3} rad	
	Exciting radiation	First Stokes component
1	4.6	6.8
2	3.6	5.6
3	1.7	2.6
4	2	3.3
5	3.5	2.1
Average	3	4

Under these conditions, the SRS is excited under saturation conditions[3]. A photograph of one of the obtained SRS spectra in benzene is shown in Fig. 3. SRS was excited with the same cells under conditions of single-mode laser operation. The data obtained for one of the measurement runs are listed in Table II. Figure 4 shows the results of photometry of the angular distribution in benzene at a cell length 10 cm.

These experiments have shown that variation of the cell length and of the SRS excitation conditions do not exert a noticeable influence on the angular distribution.

DISCUSSION OF RESULTS

A systematic study of the angular distribution of the first SRS Stokes component, carried out in[2] and in the present paper, shows that this distribution is characterized by a sharply pronounced forward directivity. This directivity is so strong that the angular distribution of the first SRS Stokes component "repeats", as it were, the distribution of the exciting radiation. Of course, the angular distribution of the SRS "repeats" the corresponding distribution of the exciting radiation only with a certain approximation. This is quite natural, if it is recognized that there are always processes that lead to a broadening of the indicated distribution.

To explain the predominant directivity of the first SRS Stokes component along the axis of the exciting radiation, it is customary to cite the difference between the optical path l along the laser beam and off the beam. Since the intensity of the SRS lines depends exponentially on l, this factor must, of course, be taken into account. But a simple geometrical analysis shows that the experimental data cannot be explained in this manner.

The simplest to analyze is the case when a parallel beam of exciting radiation passes through the cell with the investigated substance. The beam entering the cell contains only photons with the laser frequency ω. These photons first excite the usual Raman scattering at the Stokes frequency ω', and their angular distribution in the angle interval of interest to us (near the axis) can be regarded as almost uniform.

We shall henceforth follow the usual amplification scheme. Each Stokes photon induces photons in its own direction. But then, under identical geometrical conditions, as is the case near the axis, the amplification should be the same in all directions. The section through the exciting-radiation beam along the cell axis has in this case the form of a rectangle. The optical path lengths at small angles θ to the axis are obviously larger than along the cell axis. Accordingly, the SRS intensity at small angles θ to the axis, for example along the diagonal of the beam section, should not be smaller than along the axis.

The experimental data strongly contradict this conclusion. By way of example, Fig. 3 shows dashed the circle in which the uniform distribution of the SRS should be distributed in accordance with the foregoing calculation, and the experimentally obtained distribution.

The SRS angular distribution depends, generally speaking, on the mode composition of the exciting radiation, and also on side effects, for example self-focusing. However, the experiments described above have shown that the repetition effect becomes distinctly pronounced if the laser operates in the single-mode regime (Fig. 4). Since the repetition effect is observed under conditions when there is no self-focusing, this effect can likewise not be attributed to self-focusing.

It should be noted that the disparity we observed between experiment and the SRS theory is not limited to the angular distribution of the SRS radiation. An analogous disparity is also obtained with respect to the spectral width of the SRS radiation, which turned out to be much narrower for the first Stokes component than expected from the theory of the phenomenon[6,7]. In accordance with suggestions made in[3], both anomalies, called "repetition effects," are closely related and can be explained on a common basis.

A number of "anomalies" of SRS can be explained by taking into account the coherent processes in the scattering of light. In the usual analysis of scattering processes, the phases of the incident and scattered photons play no role and can be disregarded. Such an approach is insufficient in the investigation of a simultaneous interaction of two photons with a scattering molecule, wherein two photons with frequencies ω_a and ω_b and wave vectors $\mathbf{k}_a$ and $\mathbf{k}_b$ are incident on the molecules and two new photons with frequencies ω_a' and ω_b' and wave vectors $\mathbf{k}_a'$ and $\mathbf{k}_b'$ are scattered. The role of such "four-photon" processes in the SRS phenomenon is universally known. Similar processes explain the occurrence of radiation rings of anti-Stokes and higher Stokes components of SRS. A more detailed analysis shows that the SRS intensity in coherent processes does not vanish only if the coherence conditions are satisfied, namely

$$\Delta\mathbf{k} = \mathbf{k}_a - \mathbf{k}_a' - \mathbf{k}_b + \mathbf{k}_b' = 0 \quad \Delta\omega = \omega_a - \omega_a' - \omega_b + \omega_b' = 0.$$

The first formula is the well-known condition of phase synchronism, while the second expresses the law of energy conservation in each Raman-scattering process.

Let us consider now the particular case when both incident photons are photons of exciting radiation. A coherent process of the type indicated above is superimposed on the usual SRS process that leads to the formation of the first Stokes component. For such a coherent process, the condition $\Delta\omega = 0$ leads to the conclusion that if $\omega_a - \omega_b = \delta\omega$ we also have $\omega_a' - \omega_b' = \delta\omega$. But this does not mean that the SRS line width coincides in the case of coherent scattering with the width of the exciting line, i.e., that the "repetition" effect with respect to the line width takes place. From the condition of phase synchronism we obtain analogously that if $\mathbf{k}_a - \mathbf{k}_b = \delta\mathbf{k}$ then $\mathbf{k}'_a - \mathbf{k}'_b = \delta\mathbf{k}$. This means that the angular divergence of the scattered radiation does not go beyond the limits of the divergence of the exciting radiation, i.e., the "repetition effect" with respect to the angles takes place. All this radiation is directed only "forward," thus explaining the asymmetry of the SRS distribution.

The considered coherent (four-photon) RS is added to the usual incoherent (two-photon) RS. In the case of SRS, this leads to a preferred development of processes satisfying the coherence conditions. If the coherence conditions cannot be satisfied (for example, in an off-axis resonator), then the processes of amplification of the incoherent SRS develop. Saturation also leads to an increased role of the incoherent SRS and should become manifest in a broadening of the spectral line and of the angular distribution of the SRS.

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