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A STUDY OF THE SPATIAL DISTRIBUTION OF ANTI-STOKES RADIATION OF STIMULATED RAMAN SCATTERING

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The spatial distribution of the first and second anti-Stokes components of stimulated Raman scattering generated by a self-focused laser beam was investigated. The experimental results were explained in terms of a four-photon model.

It is a well known experimental property of stimulated Raman scattering (SRS) that several anti-Stokes components are observed travelling along co-axial cones [1]. There is extensive literature on the theory of SRS. However, the generation process of the anti--Stokes radiation of SRS (AS SRS) in self-focusing liquids is still not clear [2, 3].

To explain the generation process of AS SRS caused by a self-focused laser beam, Lugovoi and Prokhorov [5] recently considered the interference of a plane laser wave and a focused Stokes first component. They found that the value of the cone angle should vary within a wide range depending on experimental conditions. On the other hand, Sushchinskii [6] attempted to explain SRS-generation as a result of the coherence of two scattering processes in which laser, Stokes and anti-Stokes photons are involved. None of these theories has been experimentally confirmed [1, 6–8].

We present here the results of investigations of SRS associated with a self-focused laser beam in nitrobenzene [4]. The cones of the first and second components of the anti--Stokes radiation and their structure have been determined using beams differing in power and diameters.

A single transverse mode Q-switch ruby laser with a plane resonator was used. The pulse duration was 20–30 nsec, the beam diameters were 1, 1.2, 1.5 mm and the maximum power was between 0.9 and 2.5 MW. The transverse mode selection and the required beam diameters were obtained using a diaphragm inside the resonator.

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The first anti-Stokes cone was studied using the experimental system shown in Fig. 1a. This system provides the possibility for the simultaneous registration of two cross-section in planes perpendicular to the cone axis. The beam splitter (BS) divides the scattered light which is reflected back by mirrors M1 and M2 toward the photographic plate P. Knowing the path difference between the split beams we could determine the numeric values of the

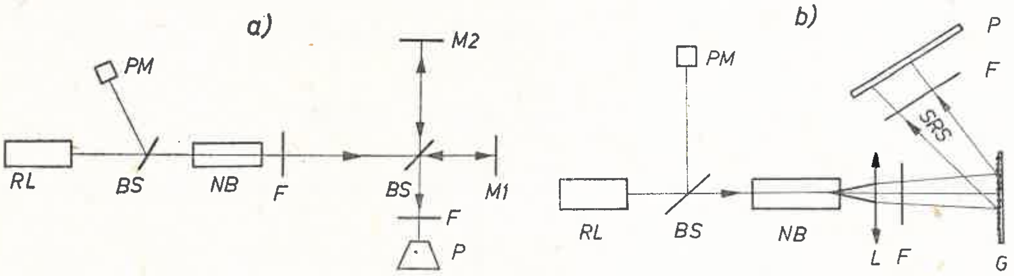


Fig. 1. Schematic arrangement of experimental systems: a) for the investigation of the first anti-Stokes component (AS_1); b) for measuring of the ratio $\varnothing_{AS2}/\varnothing_{AS1}$. RL — ruby laser, L — lens, G — grating, P — camera, F — filter, PM — power meter, BS — beam splitter, M1, M2 — mirrors, NB — cell with nitrobenzene

cone angle. The setup for investigations of the second anti-Stokes cones is shown in Fig. 1b. The grating (G) separates the first and second components of AS SRS. The cross-sections of the cones registered on the plate (P) have an elliptical shape. The length of the vertical axis of such an ellipse is a function of the cone angle (Fig. 2). We measured the emission angle of the first AS-component (AS_1) and varied the self-focusing length through a range of 11–20 cm. We found this angle to be 2.16 ± 0.01 degrees in all cases within the liquid. Simultaneously, the energy distribution at different distances from the cone top was obtained (Fig. 3). This distribution is asymmetric and has a "tail" in the outer part of the cone.

The second AS SRS component (AS_2) was studied by determining the ratio of the diameters of cone cross-sections obtained for the first and second component, $\varnothing_{AS2}/\varnothing_{AS1}$, for different distances from the nitrobenzene sample. The results are shown in Fig. 4. The ratio of $\varnothing_{AS2}/\varnothing_{AS1}$ was constant in all measurements, being 1.95 ± 0.02.

Our results can only be explained satisfactorily through a four-photon model modified by Sushchinskii [6, 8]. The L–P theory is in disagreement with these results because it implies that the angles of emission of both AS-components should vary by some percentage.

The four-photon model assumes that the AS SRS cones will be created if both scattering processes (Stokes and anti-Stokes) are coherent. Phase-matching is the condition of this coherence and for the AS_1 it can be satisfied in two ways. These two cases may be described using wave-vectors (Fig. 5):

$$\text{case A} \quad \vec{k}_L + \vec{k}'_L = \vec{k}_{AS1} + \vec{k}_{S1} \qquad \vec{k}_L \nparallel \vec{k}'_L \nparallel \vec{k}_{AS1} \nparallel \vec{k}_{S1},$$

$$\text{case B} \quad \vec{k}_L + \vec{k}_{S1} = \vec{k}_{AS1} + \vec{k}_{S2} \qquad \vec{k}_L \nparallel \vec{k}_{S1} \nparallel \vec{k}_{S2} \nparallel \vec{k}_{AS1}.$$

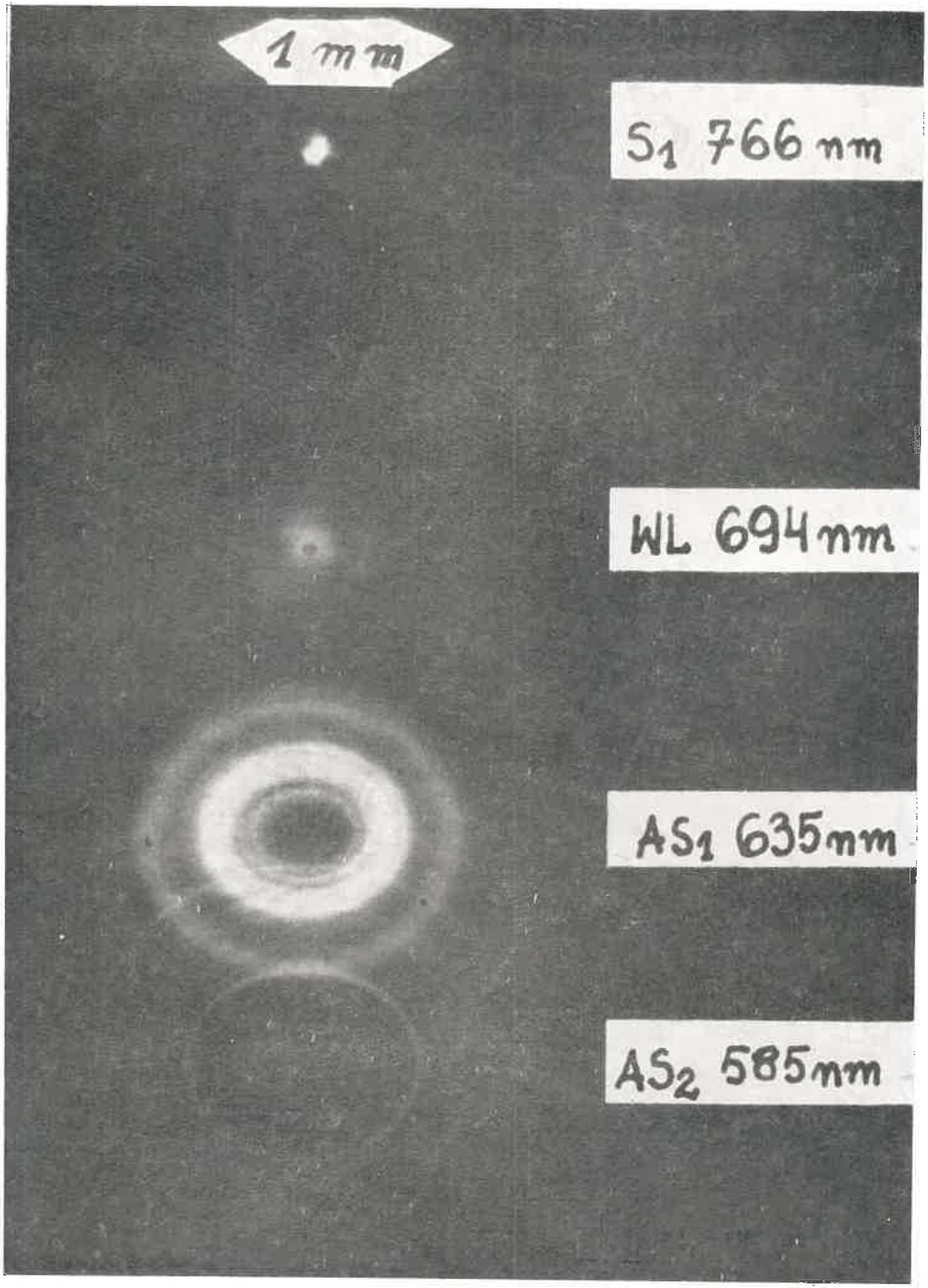


Fig. 2. Cross-section of cones obtained using the experimental setup shown in Fig. 1b. S, WL, AS — the images of Stokes, laser and anti-Stokes radiations

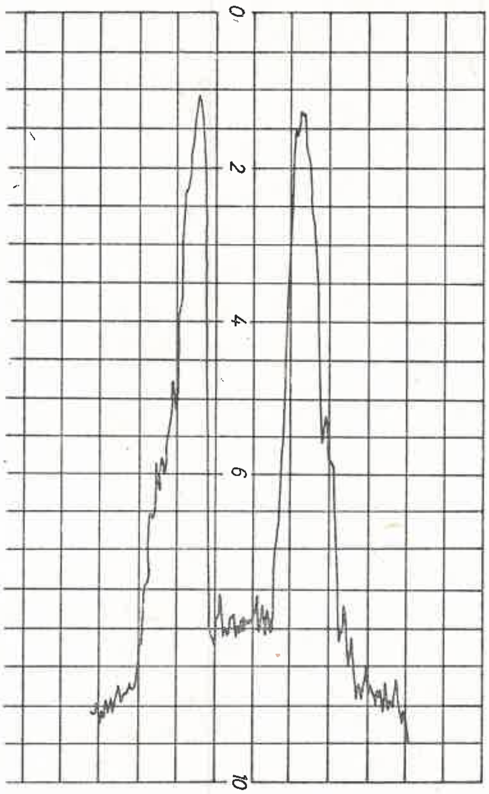


Fig. 3. Distribution of energy density in the cone of the first AS SRS component

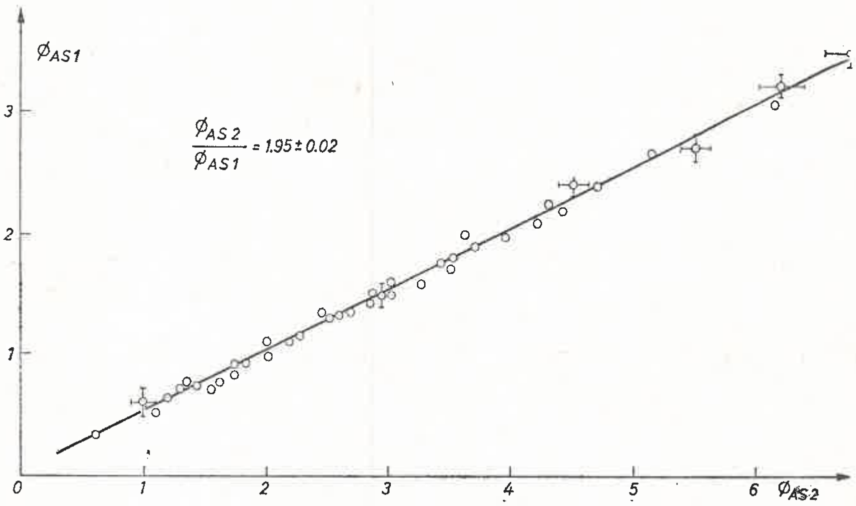
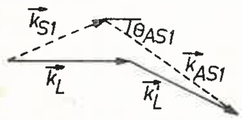
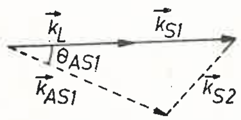


Fig. 4. The diameter of the cone of AS_2 plotted as a function of the diameter of the cone of AS_1 for differen experimental conditions

The experimental results presented are in agreement only with the four-photon B-model. In model A the AS_1 angle values would be correct provided the angle between $\vec{k}_L$, $\vec{k}'_L$ is close to 1 degree. Then, however, the energy distribution in the cone would be highly diffused toward the cone axis and unlike thats hown in Fig. 3. To explain the results for the second AS-component we must assume that (AS_2) is created in a four-photon



A



B

Fig. 5. Schematic diagram describing phase-matching conditions for the two four-photon models A and B

scattering where the S_1 and AS_1 photons produce S_2 and AS_2 photons. The wave-vector $\vec{k}_{S1}$ is parallel to the laser beam axis. Within the liquid the angle between $\vec{k}_{S1}$ and $\vec{k}_{AS1}$ is 2.16 degrees. In this case the ratio of $\varnothing_{AS2}/\varnothing_{AS1}$ is 1.83. The discrepancy between the experimental (1.95) and theoretical (1.83) values may be caused by nonlinear changes of the refractive index.

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