PHOTON DISTRIBUTION IN THE COMPONENTS OF STIMULATED RAMAN SCATTERING IN LIQUID NITROGEN

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The spectrum of stimulated Raman scattering (SRS) usually contains several Stokes and anti-Stokes components (see [1]). If there is no absorption for any of the components, then the total number of photons should be conserved in SRS. This communication deals with the question of the distribution of the photons among the components at different laser-emission intensity.

The active medium employed was liquid nitrogen. The frequency of the Raman scattering in nitrogen is $\vec{\nu}' = 2326.5$ cm^{-1} [1]. The primary emitter was a ruby laser ($\vec{\nu}_0 = 14400$ cm^{-1}). Six Stokes components could thus be produced.* We list below the wavelengths (in microns) of some of these components. The +, -, and 0 signs denote respectively the Stokes and anti-Stokes components and the laser emission.

-6	-5	-4	-3	-2	-1	0	+1....	+8
22.0	3.61	1.982	1.347	1.026	0.828	0.694	0.598....	0.303

The absorption in nitrogen is small for all of these components. High transformation coefficients were obtained for SRS in liquid nitrogen in [2].

The experimental setup is shown in Fig. 1. The radiation passes through two confocal lenses L_1 and L_2, between which a cell N_2 with liquid nitrogen was placed. The cell was 6 cm long. The laser pulse applied to the nitrogen was 11 ± 1 nsec in duration and had a divergence 7 ± 1' and a maximum energy ~2.8 J. Filter F_1 made it possible to vary the input energy from the maximum value to zero. Filter F_2 separated the radiation of one component only. Calorimeters C_1 and C_2 measured the input and output energies.

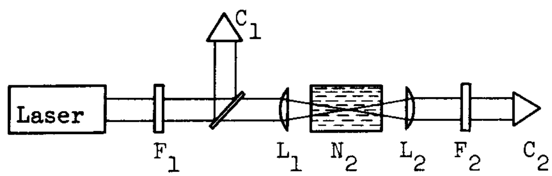

Fig. 1. Diagram of experimental setup

The energy of the forward radiation was measured for all the components from +2 to -4 (with L_1 having a focal length 3 cm). The energy distribution among the components is highly sensitive to the purity of the nitrogen. The presence of solid H_2O and CO_2 particles causes scattering of the light in the liquid nitrogen. The results shown in Fig. 2 were obtained for a scattering coefficient ~0.02 cm^{-1}.

The anti-Stokes components were registered also with an ISP-30 spectrograph. It was observed that the number of anti-Stokes components depends strongly on the focal length of the lens L_1 and ranges from five (when $f_1 = 9$ cm) to eight (when $f_1 = 3$ cm). Their intensity decreases with decreasing wavelength of the component (Fig. 3).

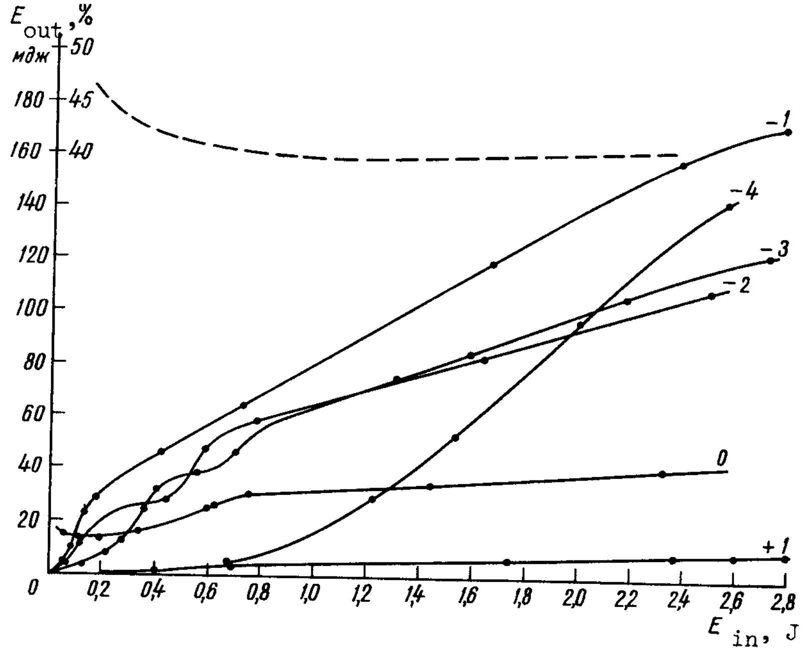

Fig. 2. Energy and relative number of photons in SRS lines vs. laser-emission energy. Solid - radiation energy in the different components, millijoules; dashed - relative number of photons, per cent.

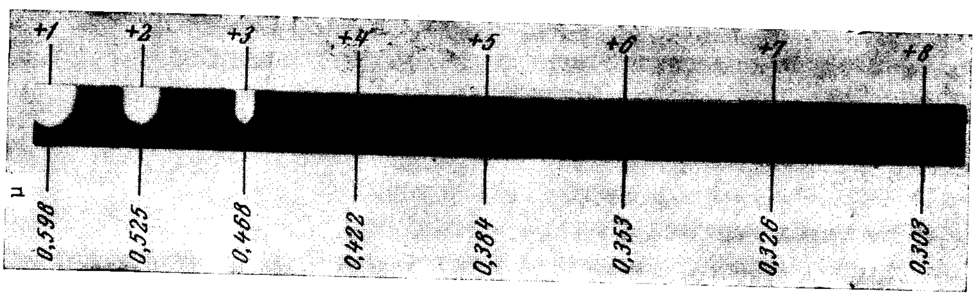

Fig. 3. SRS spectrum in nitrogen

It can be concluded from Fig. 2 that the radiation of all the Stokes components appears simultaneously. Visual observations have shown that the anti-Stokes components appear simultaneously with the Stokes components when the laser emission energy is ~20 mJ. The form of the curve for the zero component indicates that practically the entire laser emission is converted when this threshold is exceeded.

As seen from Fig. 2, an effective conversion into the -4 component is possible and the number of photons in it reaches ~16% at maximum input energy. However, an appreciable conversion into the -4 component is attained much later than into the -3 component. This is apparently due to the decrease in the Raman-scattering probability with decreasing radiation frequency. On this basis, we can hardly expect a noticeable photon fraction in components -5 and -6 at the presently attainable laser intensity. The radiation energy in the +2 component, which is not shown in Fig. 2, is approximately 2.5 times as small as in the +1 component.

The curves of Fig. 2 give, apart from constants, the photon distribution among the components as a function of the number of photons in the laser pulse. Using these data we can readily calculate what fraction of the primary photons emerges from the nitrogen forward. This is shown by the dashed curve of Fig. 2. As seen from this curve, in a wide range of laser power, approximately half the photons emerge forward. The photons in the anti-Stokes components were disregarded in the calculations, but it is clear that no significant changes will occur when they are taken accurately into account.

No anti-Stokes components were observed in the backward radiation. At an input energy larger than 2 J, the backward radiation is approximately double the forward radiation; in the -4 component, the forward radiation is several times larger than the backward one.

The foregoing results make it obvious that the theory of stepwise SRS is not valid, at least at large excitation intensities. There exists a strictly defined threshold, beyond which practically all the laser emission is transformed into the Stokes region. An insignificant fraction of the photons goes into the anti-Stokes region. When the intensity of the laser emission is increased, the photons are redistributed among the Stokes components. The higher the laser intensity, the farther it is possible to go into the Stokes region. If the intensity is high enough, we can expect transfer of the photons to the extreme Stokes components. This uncovers the possibility of obtaining powerful laser emission in the far infrared.

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[1] B. P. Stoicheff, "Enrico Fermi" Internat. School of Physics, Course XXXI, 19-31 August 1963.

[2] N. G. Basov, A. Z. Grasyuk, and V. A. Katulin, Dokl. Akad. Nauk SSSR 161, 1306 (1965) [Sov. Phys.-Dokl. 10, 343 (1965)].

* Raman scattering of light of frequency ν can occur if $\vec{\nu} > \vec{\nu}'$. Therefore the number of possible Stokes components is equal to the integer part of $\vec{\nu}_0/\vec{\nu}'$.