STIMULATED SCATTERING OF LIGHT OF THE RAYLEIGH LINE WING IN AN EXTERNAL RESONATOR

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The phenomenon of stimulated scattering of light of the Rayleigh line wing (SRW), observed in [1], was investigated experimentally [1 - 5] and theoretically [1,4, 6 - 8] a number of times.

We report here the first of a study of generation of SRW light in an external transverse resonator (see Fig. 1), in which a cylindrical lens L_1 focuses the light of a ruby-laser pulse polarized in a plane perpendicular to the scattering plane, of power ~100 mW and duration ~12 nsec.

The generation of the SRW was investigated in eight liquids, and was observed from the first time in n-xylol, benzene, and toluene. If the resonator mirror M_1 is covered, the SRW spectrum disappears.

Depending on the character of the generated SRW spectrum, the investigated liquids can be arbitrarily subdivided into two groups.

In the liquids of the first group [benzaldehyde (spread of generated SRW spectrum 3 cm^{-1}), nitrobenzene (1.5 cm^{-1}), quinoline (2.5 cm^{-1}), o-xylol (2.5 cm^{-1}), and n-xylol (4.5 cm^{-1}) (see Fig. 2)], only Stokes SRW is observed.

The second group consists ofcarbon disulfide, benzene, and toluene. The SRW generation

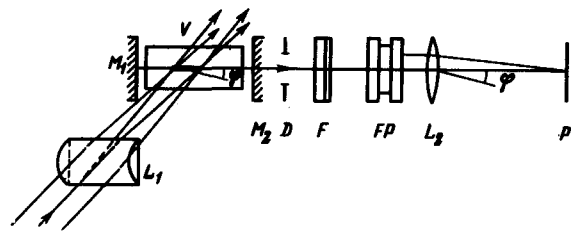

Fig. 1. Diagram of setup. L_1 - cylindrical lens (f = 5 - 7 cm), V - vessel with investigated liquid. M_1 and M_2 - mirrors with reflectances 100 and 50%, respectively (distance between mirrors 10 cm), FP - Fabry-Perot interferometer, ϕ - angle between resonator axis and registration direction.

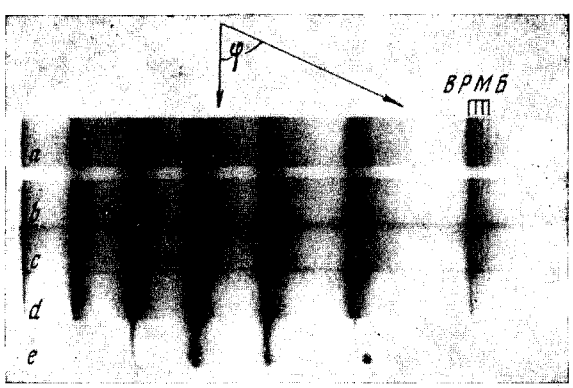

Fig. 2. SRW spectrum in n-xylol. Interferometer dispersion region $\Delta\nu$ = 5 cm^{-1}: a - exciting light not attenuated; b, c, d, e - exciting light attenuated by 1.5, 2.3, 3, and 5 times, respectively

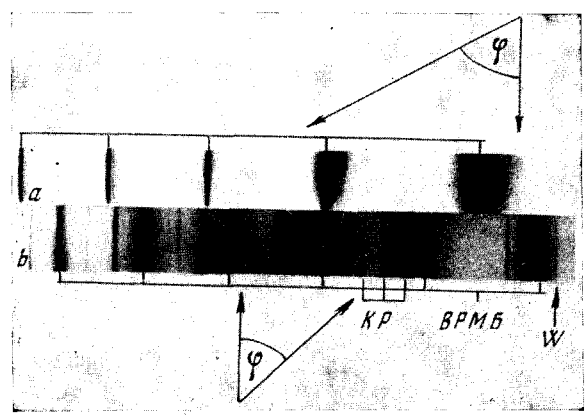

Fig. 3. SRW spectrum: a - in toluene ($\Delta\nu$ = 5 cm^{-1}), b - in CS_2 ($\Delta\nu$ = 5 cm^{-1}). W - maximum in SRW ($\phi \sim 2^\circ$). SR - stimulated Raman scattering lines. SMBS - stimulated Mandel'shtam-Brillouin scattering components.

spectrum is more complicated in this case than in the liquids of the first group (Fig. 3). At an angle $\phi \sim 0.5 - 1^\circ$ relative to the resonator axis (Fig. 1), the SRW spectrum in these liquids consist of Stokes and anti-Stokes parts. At angles $\phi > 1^\circ$, only the Stokes wing is observed in the scattered light.

The most complicated is the SRW spectrum in carbon disulfide (Fig. 3). In the cone of angles $\phi \sim 0.7^\circ$, a Stokes (~ 2.5 cm^{-1}) and anti-Stokes (~ 1.5 cm^{-1}) part of the wing is observed in the SRW spectrum of CS_2. In the angle region $\phi \sim 1 - 2^\circ$, only the Stokes wing, extending about 7 cm^{-1} and having no visible maximum in the intensity distribution, is observed. Only several interference patterns show, at $2 - 3^\circ$, an SRW line whose shift $\Delta\nu_{max} = 2.5$ cm^{-1} corresponds to the frequency of the maximum gain $g_1(\Omega)$ [6].

A possible explanation of the results is as follows. In carbon disulfide, benzene, and toluene in an external resonator, intense SMBS takes place and propagates along the resonator axis. The intensity of the SMBS light is strong enough to cause four-photon interaction in SRW at small scattering angles [3, 6, 7]. According to (14) of [6], the gain $g_2(\Omega)$ in four-photon interaction is maximal at a scattering angle $\theta = \theta_{opt} = A^{1/2}|E_0|$, is the same for the Stokes and anti-Stokes parts of the wing, and has a maximum (unlike $g_1(\Omega)$) when $\theta >> \theta_{opt}$, at a frequency $\Omega = 0$.

At relatively large angles θ (scattering angle larger than θ_{opt}), no four-photon interaction is realized and ordinary Stokes SRW sets in either as a result of the intensity of the intensity of the SMBS or as a result of the intensity of the exciting radiation (scattering angle $\theta = 90^\circ$), which has the character of either a continuous spectrum or a line at frequency $\Omega = 1/\tau$ (Fig. 3).

In liquids belonging to the first group, the hypersound absorption coefficient is larger, and consequently the SMBS intensity is smaller. In this case, the SRW is due to the intensity of the exciting light with scattering angle $\theta = 90^\circ$, and consequently has only a Stokes component.

It is still unclear why, with rare exceptions, a continuous spectrum and not a line were produced in the SRW spectrum in these and in the earlier experiments [1, 2].

One of the possible explanations was advanced by us [2] and was subsequently developed in detail in [8]. According to it, the criterion $[(a_1 - a_2)/kT]|E_0|^2 << 1$ is not satisfied under ordinary experimental conditions. For carbon disulfide, the violation of this criterion occurs in fields $|E_0| > 10^7$ V/cm. In such fields, saturation of the molecule orientation sets in and the maximum of the SRW spectrum may vanish. However, in the experiments reported here, $|E_0| \sim 10^6$ V/cm or less, and therefore this explanation does not hold in our case.

An examination of the processes occurring in stimulated scattering has led us to the conclusion that ultrashort pulses can occur in the SRW, and the usually observed continuous broad spectrum of SRW light (in place of the expected line) can be attributed to the formation of such ultrashort pulses of scattered light. The formation of picosecond pulses was already observed in stimulated Raman scattering [9].

The mechanism of formation of such ultrashort pulses in stimulated scattering is analogous to the already described mechanism [10] of narrowing of a laser pulse propagating in a

medium with inverted population.

The action of such a mechanism of shortening the scattered light pulse is clear from an analysis of backward scattering ($\theta = 180°$). In the case of large gain, the leading front of the scattered-light pulse produced, for example, in the focal region and propagating in a direction opposite to that of the exciting light will draw from the latter an appreciable fraction of the energy, and with this the slope of the leading front will increase while the trailing edge will practically not be amplified. As a result, the scattered-light pulse can have a duration t_s much shorter than the total duration t_0 of the exciting light. Within a time t_0 we can expect several such short pulses of scattered light, and their limiting duration is set by the anisotropy relaxation time, $(t_s)_{lim} \sim \tau$, and consequently it can reach a value $\sim 10^{-12}$ sec. This estimate of the limiting duration of the pulse in SRW agrees with the experimentally observed broadening of the spectrum of the Stokes SRW ($\Delta\Omega \sim 1/\tau$). The considered mechanism of formation of ultrashort pulses is most effective at scattering angles $\theta = 180°$, but can occur at any $\theta \neq 0$.

The existence of ultrashort SRW pulses with duration shorter than the lifetime of the acoustic phonons makes it possible to explain the competition observed in these and earlier [2] experiments and leading to a total or partial suppression of the SMBS components upon occurrence of SRW. Apparently, the SMBS phenomenon does not have time to become established while the ultrashort SRW pulses occur.

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[1] D. I. Mash, V. V. Morozov, V. S. Starunov, and I. L. Fabelinskii, ZhETF Pis. Red. 2, 41 (1965) [JETP Lett 2, 25 (1965)].
[2] G. I. Zaitsev, Yu. I. Kyzylasov, et al., ibid. 6, 505 (1967) [6, 35 (1967)
[3] G. I. Zaitsev, Yu.I. Kyzylasov, et al., ibid. 6, 695 (1967) [6, 180 (1967)
[4] V. S. Starunov, Trudy FIAN 39, 151 (1967).
[5] N. D. Foltz, C. W. Cho, D.H. Rank, and T. A. Wiggins, Phys. Rev. 165, 396 (1968).
[6] V. S. Starunov, Dokl. Akad. Nauk SSSR 179, 65 (1968) [Sov. Phys.-Dokl. 13, 217 (1968)].
[7] R. Chiao, P. Kelly, and E. Garmire, Phys. Rev. Lett. 17, 1158 (1966)
[8] R. M. Herman, Phys. Rev. 164, 200 (1967).
[9] M. Maier and W. Kaiser, Phys. Rev. Lett. 17, 1275 (1966).
[10] N. G. Basov, R. V. Ambartsumyan, V. S. Zuev, P. G. Kryukov, and V. S. Letokhov, Zh. Eksp. Teor. Fiz.50, 23 (1966) [Sov. Phys.-JETP 23, 16 (1966)].

SHAPE OF COEXISTENCE CURVE OF THE LIQUID AND GASEOUS PHASES OF ARGON

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It is known that there is no meeting of minds concerning the shape of the coexistence curve of pure liquids near the critical point. In the first approximation, this curve can be expressed by the formula

$$|\rho - \rho_c|/\rho_c = A[(T_c - T)/T_c]^{\beta},$$

where ρ is the density of the liquid or saturated vapor at temperature T, and A and β are universal contacts.