STIMULATED SCATTERING OF LIGHT OF THE RAYLEIGH-LINE WING

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A phenomenon, called stimulated scattering of light, is produced in the field of the optical wave of the giant pulse of a ruby laser, as a result of nonlinear interaction between the light and the medium. Stimulated Raman scattering of light was previously observed [1], as well as stimulated Mandel'shtam-Brillouin scattering [2]. In this note we report the observation of a new phenomenon - stimulated scattering of light of the Rayleigh-line wing.

The spectrum of thermal scattering of light in liquids consisting of anisotropic molecules contains a more or less broad section of continuous spectrum, the maximum intensity of which occurs at the frequency of the exciting line and decreases on both sides of this frequency, reaching sometimes 100 - 150 cm^{-1} [3].

In the thermal light scattering, the wing of the Rayleigh line is due to modulation of the scattered light as a result of the fluctuations of the anisotropic-molecule orientations, which attenuate in time. The weak field of the exciting optical wave (ordinary source of light) turns out to exert such a negligible influence on the anisotropy of the medium, that it can be disregarded. The situation is different when the scattering is excited by a focused giant laser pulse. Then the intensity of the electric field of the light wave is so large that it produces, together with the field of the thermal scattering, an appreciable force [4] 1): $f \cong (\alpha_1 - \alpha_2)E^2/\tau kT$ (α_1, $\alpha_2 = \alpha_3$ - principal polarizabilities of the molecule, τ - anisotropy relaxation time, k - Boltzmann's constant, T - absolute temperature, and E - sum of the fields of the exciting and scattered light). The low-frequency component of this force causes anisotropy of the medium as a result of the orientation of the anisotropic molecules.

For a classical solution of the problem of stimulated scattering of the light from the Rayleigh-line wing it is necessary to solve simultaneously the nonlinear Maxwell's equations and the nonlinear equations for the anisotropy tensor of the medium [4]. Assuming here that the nonlinear terms are small compared with the linear ones, and using the method of the small parameter with abbreviated equations [3,5], we can derive the following expression for the threshold of the Stokes part of the stimulated wing of the Rayleigh line (the anti-Stokes component does not arise in the approximation considered here):

$$\frac{E^2}{8\pi} \geq \frac{45 n k_\omega kT}{16\pi |k_1| \frac{\partial\epsilon}{\partial s} \frac{I(\Omega)}{I(0)} \Omega\tau(\alpha_1 - \alpha_2)} \qquad (1)$$

Here n - refractive index of the medium, k_ω - summary coefficient characterizing the optical losses, k_1 - wave number of the Stokes component of the wing, s - anisotropy tensor, Ω - frequency measured from the maximum of the wing, ω - dielectric constant, $I(\Omega)/I(0)$ - ratio of the intensity at the frequency Ω to the intensity at the maximum of the Rayleigh-line wing, due to the fluctuations (thermal scattering). The quantity $\Omega\tau(I(\Omega)/I(0))$ has a maximum. Indeed, if we assume, making a slight simplification [3], that

$$\frac{I(\Omega)}{I(0)} = \frac{1}{1 + \Omega^2\tau^2} \qquad (2)$$

then $\Omega\tau(I(\Omega)/I(0))$ will have a maximum at a frequency $\Omega = 1/\tau$. Consequently, at this frequency the expression for the threshold (1) will have a minimum, and this in turn signifies that the stimulated scattering of light of the Rayleigh-line wing will have a maximum at a frequency Ω, and that this maximum does not coincide with the maximum of the intensity of the light of the wing in thermal scattering.

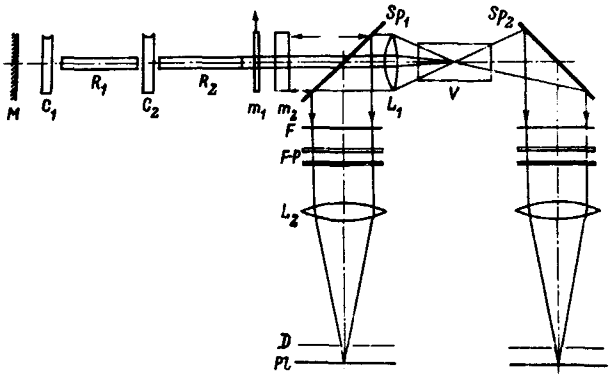

Fig. 1. Diagram of setup. M - mirror ($R \approx 100\%$), R_1, R_2 - rubies of length 12 cm and diameter 1.2 cm, m_1, m_2 - plane-parallel plates. Sp_1, sp_2 - beam splitting glass plates, L_2 - objective (s = = 120 cm).

We chose for the investigation the following liquids: carbon disulfide, benzene, toluol, nitrobenzene, acetic acid, salol, and triacetin. The exciting light came from a ruby laser (Fig. 1) with Q modulated by two cuvettes C_1 and C_2 one cm thick, filled with a solution of cryptocyanin in methyl alcohol with concentration 1.8×10^{-6} M. In the mode employed for observation of the stimulated scattering of light of the Rayleigh-line wing, the pulse power was approximately 100 MW. The light from the pulse was focused by a lens L_1 (f = 3 cm) inside a vessel V filled with the scattering liquid. The light scattered at angles 180° and 0° relative to the direction of the exciting light was guided through a filter F to a Fabry-Perot interferometer F-P and photographed on a photographic plate Pℓ. The exciting light was alternately attenuated by introducing between m_2 and Sp various types of flat glass plates. The pictures were obtained on the Fabry-Perot etalon in the dispersion regions 50, 16, 8, and 1 cm^{-1}. The stimulated scattering of the light of the Rayleigh-line wing was observed in carbon disulfide and nitrobenzene at room temperature and in salol at 170°C. In benzene, toluol, acetic acid, and triacetin the phenomenon was not observed under the same conditions.

The stimulated Mandel'shtam-Brillouin scattering was observed in all these liquids except triacetin.

Figure 2 shows the stimulated scattering of light of the Rayleigh-line wing in nitrobenzene and salol. The stimulated wing in nitrobenzene extends over approximately 1 cm^{-1}, and a maximum is noted in its intensity distribution; it is difficult, however, to establish the position of this maximum, since it lies close to the Stokes component of the stimulated Mandel'shtam-Brillouin scattering, ~ 0.23 cm^{-1}. In the case of salol one can clearly see a band (stimulated wing) with maximum near 0.16 cm^{-1}.

In carbon disulfide, at the used intensity of the exciting light, the stimulated wing of the Rayleigh line extends to 15 cm^{-1}. When the intensity of the exciting light was reduced by 10 - 15%, the stimulated wing disappeared. These changes are shown in Fig. 3.

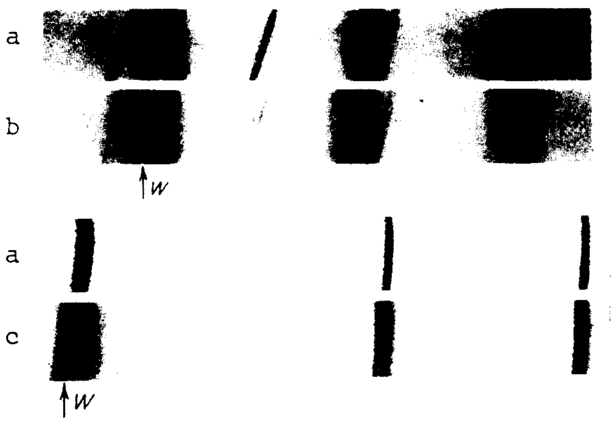

Fig. 2. Spectra of stimulated scattering of light of the Rayleigh-line wing (interferometer dispersion region 5 cm^{-1}): a - Emission spectrum of ruby laser, b - nitrobenzene at 20°C, c - salol at 170°C.

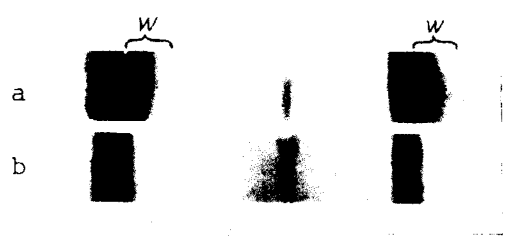

Fig. 3. Spectrum of stimulated scattering of light of the Rayleigh-line wing in carbon disulfide at 20°C: a - At an exciting light-pulse power ~ 100 MW (interferometer dispersion region 50 cm^{-1}), b - the same with power reduced 10 - 15%.

The stimulated wing of the Rayleigh line has a pronounced threshold in all three liquids.

The existence of a threshold, the fact that the scattering intensity is comparable with the intensity of the exciting light, the presence of a maximum in the wing, and the absence of an anti-Stokes wing - all prove that stimulated scattering of the light from the Rayleigh-line wing was observed in the three cases described above.

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[1] E. J. Woodbury and W. K. No, Proc. IRE 50, 23 and 67 (1962).

[2] Chiao, Townes, and Stoicheff, Phys. Rev. Lett. 12, 592 (1964); R. G. Brewer and K. E. Rieckhoff, Phys. Rev. Lett. 13, 334 (1964).

[3] I. L. Fabelinskii, Molecular Scattering of Light, Trudy FIAN (Physics Institute, Academy of Sciences) 9, 181 (1957).

[4] J. I. Frenkel, Kinetic Theory of Liquids, Dover, 1954.

[5] S. A. Akhmanov and R. V. Khokhlov, Problems of Nonlinear Optics (1962 - 1963), AN SSSR, Information Institute, 1964.

1) In deriving the expression for f it was assumed that $(\alpha_1 - \alpha_2)E^2 < kT$ (this is satisfied in fields up to $E \sim 10^7$ V/cm).