MEASUREMENT OF THE AMPLIFICATION COEFFICIENT OF STIMULATED LIGHT SCATTERING OF THE RAYLEIGH LINE WING

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Stimulated scattering of light of the Rayleigh line wing (SRW) is investigated experimentally at various intensities and polarizations of the exciting radiation. The SRW amplification is measured at intensities $I \leq 10^2$ MW/cm^2 and the shape of the amplification band is studied. At intensities $\sim 10^4$ MW/cm^2 an SRW line is observed in the back-scattered light spectrum whose position is identical with that of the amplification coefficient. Under the same conditions, a diffuse wing with a spread of 1 cm^{-1} and a smeared-out maximum in a region ~ 0.7 cm^{-1} is observed in the forward-scattered light spectrum. A discussion is given of the experimental results.

STIMULATED scattering of the Rayleigh line wing (SRW)[1] has been studied frequently, but there have been no quantitative data to date that would allow us to decide as to the applicability of the formulas of the linearized theory of this phenomenon.[1,2] Moreover, at high intensities of the exciting radiation, the experimentally observed picture of the SRW spectrum differs qualitatively from that which one should have expected from existing theory, and it is frequently not clear why and under what conditions the linearized theory ceases to describe the observed phenomenon.

The goal of the present research—quantitative measurements of the amplification coefficient of the SRW $g(\Omega)$ as a function of the frequency $\Omega = \omega_0 - \omega_1$, where ω_0 and ω_1 are the frequencies of the exciting and scattered light, respectively, the measurements were made for o-xylol in excitation by light of comparatively low intensity ($I \leq 10^2$ MW/cm^2), polarized both linearly and circularly. In addition to this, for high intensities ($I \sim 10^4$ MW/cm^2) the SRW spectrum and the stimulated Mandel'shtam-Brillouin scattering (SMBS) were studied.

It follows from the linearized theory[1,2] that $g(\Omega)$ for the SRW has a maximum at the frequency $\Omega = 1/\tau$, where τ is the relaxation time of the anisotropy. In correspondence with this, for large amplification coefficients and not too small scattering angles ($\vartheta > 1-2°$), at a frequency $\omega_1 = \omega_0 - 1/\tau$, a sharp SRW line should be expected. However, both in the first research,[1] and in a number of later works,[3-6] a Stokes diffuse wing was frequently observed along with the SRW line, sometimes with a weak smeared-out maximum in the region about the frequency $\Omega = 1/\tau$. Such a broadening of the SRW spectrum can be due to the following causes: saturation in the orientation of anisotropic molecules,[2,3,6,7] multimode structure of the laser radiation,[8] four-photon interaction in forward scattering,[2,9,10] phase modulation,[11] and formation of picosecond pulses in the SRW.[4,12] For low intensities of single-mode laser radiation and scattering at large angles, the effect of the indicated factors on the SRW should be insignificant, and under these conditions, a quantitative comparison of the experimental results with the linearized theory should be possible. The data thus far obtained[13] turn out to be insufficient to make a decision as to the agreement between the results of experiment and the theory relative to the shape of the amplification band of the SRW, and this gap is filled here.

The measurement of the amplification coefficient was carried out by us for conditions under which the increment $J = gI_0L$ (L is the region of nonlinear interaction) for the SRW was of the order of unity. The experimental arrangement is shown in Fig. 1. A ruby single mode laser (TEM_{00}) was used with a power of 1–5 MW and pulse length ~30 picosec. The laser radiation of frequency ω_0 passed through the vessel W with the liquid under study and was focused on the sample, MB is a generator of SMBS radiation of frequency $\omega_1 = \omega_0 - \Omega_{MB}$. This SMBS radiation, attenuated to the intensity $I_{MB} < 0.1I_0$, was propagated opposite to the laser radiation and, encountering it in the vessel W (the length of the vessel L = 10 cm), was amplified because of the nonlinear interaction. The photoelectric detectors Ph_1, Ph_2, and Ph_3 measured the power P' of the SMBS radiation entering the vessel W, and the power P'' of the SMBS radiation after passage through the vessel W. All three pulses were fed to a calibrated oscillograph with corresponding delays. Measurement of the ratio P''/P', of the quantity P_0

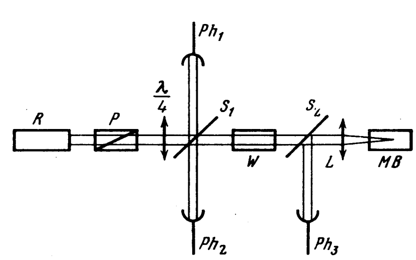

FIG. 1. Arrangement of apparatus for the measurement of the amplification coefficient of the SRW. R—Ruby laser, P—Glan prism, S_1, S_2—separating plates, Ph_1, Ph_2, Ph_3—radiation detectors, W—vessel with test liquid, L—focusing lens, MB—SMBS generator.

and of the cross section of the radiation bundles allows us to determine the amplification g at the frequency $\omega = \omega_0 - \Omega_{MB}$. By changing the material of the SMBS generator, measurements could be made of the frequency Ω_{MB} and, consequently, the dependence of g on the frequency $\Omega = \Omega_{MB}$ could be investigated.

The experimental conditions were so chosen that the SMBS intensity in the vessel was much smaller than the other recorded signals. The substance o-xylol was chosen as the object of investigation. The molecules of this liquid have large anisotropies in the polarizability and, consequently, large amplification coefficients. Moreover, the expected position of the maximum in the amplification coefficient (at a distance $\sim 0.7\ cm^{-1}$ from the frequency of the exciting radiation) lies in a region easily accessible to investigation.

As the generator of the SMBS test signal we used liquid carbon tetrachloride, plastic, TF-5 glass, fused quartz, and sapphire crystal. Consequently, the shape of the SRW amplification band was studied in the region of frequencies $\Delta\nu = 0.14-2\ cm^{-1}$, where $\Delta\nu$ is measured from the frequency of the exciting radiation.

The results of the measurement of the dependence of the relative amplification coefficient $g(\Omega)/g_{max}$ on the frequency for circular $g_0(\Omega)/g^0_{max}$ and linear $g''(\Omega)/g''_{max}$ polarizations of the exciting radiation are shown in Fig. 2. Comparison of the obtained results with the theory (solid curve) shows that there is excellent agreement between them. The measured location of the maximum and that computed from data on the quantity τ from the investigation of the thermal Rayleigh line wing[14] agree with one another ($\Delta\nu_{max} \approx 0.7\ cm^{-1}$). The accuracy of measurement of the relative value of g was no worse than 50%. Because of the difficulty of accounting for the spatial distribution of the radiation, the accuracy of the measurement of the absolute value of the amplification coefficient was worse (by a factor of ~ 2). The amplification coefficient, averaged over all the measurements, for linear polarization of the exciting light, after reduction of the measurements to the frequency $\Omega = \Omega_{max}$ was found to be equal to $g''_{max} = 5 \times 10^{-3}$ cm/MW, which agrees well with the quantity 5.7×10^{-3} cm/MW calculated from the formula with account of correction for the internal field:[7]

$$g'' = 4\pi\left(\frac{\varepsilon+2}{3}\right)^4 \frac{N(\alpha_\parallel - \alpha_\perp)^2 |k_1| \Omega\tau}{45kT(1+\Omega^2\tau^2)} |\varepsilon_L|^2,$$

where ϵ_L is the amplitude of the laser radiation, N the number of molecules per unit volume, $\Omega = \omega_0 - \omega_1$ the difference in frequencies of laser and scattered light, $|\mathbf{k}_1|$ the modulus of the wave vector of the scattered wave, $\alpha_\parallel$ and $\alpha_\perp$ the principal components of the polarizability tensor of the molecules, and τ the anisotropy relaxation time. Thus, our measurements show that the linearized theory for small intensities of the exciting radiation quantitatively describes the SRW phenomenon.

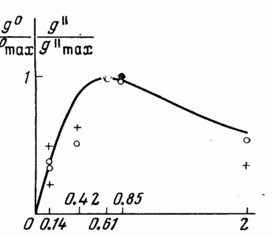

FIG. 2. Dependence of the relative amplification coefficient for circular (O–$g^0(\Omega)/g^0_{max}$) and linear (+–$g^{\parallel}(\Omega)/g^{\parallel}_{max}$) polarizations of the exciting radiation. The continuous curve is the theoretical curve. To the given measurements one should add the quantity $g^0/g^0_{max} = g^{\parallel}/g^{\parallel}_{max} = 0.96$ for $\Delta\nu = 0.54\ cm^{-1}$.

We also investigated the spectrum of light scattered forward $\vartheta \approx 0$ and backward ($\vartheta \approx 180°$) for excitation of the scattering by circularly polarized radiation. This radiation was focused into a vessel with o-xylol by a lens with a focal length ~ 2 cm. The spectrum of the scattered radiation was analyzed by means of a Fabry-Perot interferometer with a dispersion range of $2.5\ cm^{-1}$.

Figure 3 shows the interferograms of scattered light for observations forward (a) and backward (b), when the SRW increment was $J = gI_0L \sim 4-5$ and for observation forward (c) and backward (d) when the radiation intensity was large ($J \sim 8-10$). In the case when $J \sim 4$, only the lines of SMBS for which the amplification coefficient is much greater than for SRW were observed in the scattered light spectrum. For values of the increment $J \sim 8$, lines of SMBS are observed in the spectrum of back scattered light (Fig. 3d) and the sharp line of SRW, displaced relative to the exciting line by an amount $\Delta\nu = 0.67\ cm^{-1}$, which corresponds to an anisotropy relaxation time $\tau = 7.9 \times 10^{-12}$ sec. The position of this line is in excellent agreement with the position of the maximum of the amplification coefficient (see Fig. 2). For the same value of the increment, a diffuse wing is observed in the spectrum of forward scattered light (see Fig. 3c), with extension up to $\sim 1\ cm^{-1}$ with a broad maximum in the range $0.5-0.8\ cm^{-1}$ and several (~ 5) lines of SMBS, and the four lines of SMBS, located at a distance of $\sim 0.72\ cm^{-1}$ from the exciting line have greater intensity than the third line. Inasmuch as the laser in our experiments was decoupled from the scattering volume, the principal reason for the appearance of a large number of Stokes components of the SMBS must be the repeated scattering of the region of nonlinear interaction.[15] Here the appearance of odd lines of SMBS is connected with the reflection from the apertures of the vessel with the scattering liquid. Moreover, the features of the forward scattering that have been described can also be explained as the effect of four-photon[2,10,15] interaction in SRW at small angles. For such four-photon process, in contrast with the two-photon scattering, the maximum of the amplification coefficient is achieved at the frequency of the scatter-

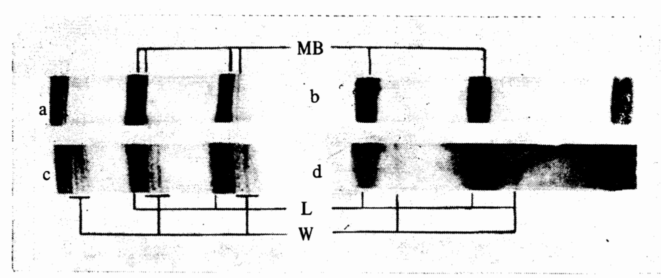

FIG. 3. Interferograms of the scattered radiation: forward (a, c) and backward (b, d). a, b–increment of amplification ~4; d, c–increment of amplification ~8; L–line of exciting radiation, MB–SMBS components, W–lines and spectrum of SRW. The dispersion region of the interferometer is 2.5 cm^{-1}.

ing radiation. The total effect of two- and four-photon processes can lead to the emergence of a broad SRW spectrum, to an increase in the amplification of the SMBS components and, as a consequence, to the appearance of a large number of these components.

For the SMBS lines, which coincide with the maximum of the amplification band for SRW, the amplification coefficient will be greater than for the neighboring lines, as has been observed experimentally.

We also observed that, for intensities I_0 close to the threshold of recording the SRW, the SMBS components broaden in the Stokes direction, as is also indicated in[6]. This broadening may be due to the same processes which produce the broadening of the lines of the exciting radiation. In addition to this, for high intensities of the fields of the light waves, when the molecules are oriented preferentially in a plane perpendicular to the direction of propagation of the light (circularly polarized), during the light pulse, the parameters of the medium which determine the velocity and absorption of hypersound will change. This can lead to a continuous change in the SMBS spectrum over the duration of the light pulse.

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