Fine structure of stimulated light scattering of the Rayleigh line wing

D. V. Vlasov and I. L. Fabelinskiĭ

P. N. Lebedev Physical Institute, USSR Academy of Sciences

(Submitted October 8, 1975)
Zh. Eksp. Teor. Fiz. 70, 1350–1362 (April 1976)

The temperature kinetics of a new phenomenon, the splitting of the stimulated scattering line of the Rayleigh line wing (RLW), is observed and investigated by a specially developed experimental technique. The measurements were performed in four organic liquids with optically anisotropic molecules, viz., salol, α-chloronaphthalene, nitrobenzene, and quinoline. A number of general properties of the spectral distributions of the stimulated depolarized scattering increments and of the corresponding thermal scattering cross sections are established on the basis of calculations. A method is shown for the experimental determination of the signs of the two mechano-optical coefficients, which are the parameters of the phenomenological relaxation theory. The results of the experiments are compared with calculations based on the formulas of the phenomonological theory.

PACS numbers: 78.30.Ly

The spectrum of the light scattered by fluctuations of the anisotropy in a liquid constitutes a broad band with a rather complicated intensity distribution. This band, or wing of the Rayleigh line, extends 100–150 cm^{-1} in both directions from the frequency of the exciting light. Even before the advent of laser sources, it had been experimentally established that in acetic acid and triacetate,[1] and also in a number of other liquids,[2] a narrow portion of the wing is observed, the halfwidth of which does not exceed 0.3 cm^{-1}, and it can be seen on an interferogram as a distinct line if the polarization of the scattered light is such that the linearly polarized Mandel'shtam–Brillouin components and the central component are completely extinguished.

The application of a spectrally narrow, intense gas-laser line enabled us to study the structure of the Rayleigh line wing and to discover a new phenomenon[3] in the J_{yz} and J_{zx} spectra1) of depolarized scattered light. In this phenomenon, the narrow portion of the wing is a doublet consisting of broad components which are considerably superimposed. This phenomenon was not described by the then-existing theories of the distribution of the depolarized scattering of light. Within a year, this phenomenon has been confirmed experimentally[4] and maxima have been observed for the J_{yz} component of the depolarized scattering spectrum in the frequency range of the Mandel'shtam–Brillouin components, due, according to modern representations, to the interaction of acoustic longitudinal waves with the modes of oriented motion of the medium.

The purpose of the present research was to investigate experimentally the stimulated light scattering, brought about by the J_{yz} and J_{yx} components of the spectra of thermal depolarized scattering. The threshold of stimulated scattering on the wing of the Rayleigh line (RLW) is considerably higher than the threshold of the scalar stimulated Mandel'shtam–Brillouin scattering; therefore, a special method of selection was developed[5–7] for the depolarized types of stimulated scattering (SS), which enables us to suppress scalar types of scattering, and also the phenomena of self-focusing and phase modulation, and thus, to observe and study the stimulated scattering of the Rayleigh line wing in two components of polarization of the scattered light: J_{yz} and J_{yx}.[5–7] Numerical calculations are given for the spectral distributions of the increments of the stimulated scattering and the thermal scattering cross section, and where possible, the results of calculations are compared with the experimental results.

1. CALCULATION OF THE COEFFICIENTS OF AMPLIFICATION AND THE CROSS SECTIONS OF THERMAL SCATTERING

The phenomenological relaxation theory of thermal light scattering yields expressions for the spectral intensity densities of the scattered light $J_{\alpha\beta}$ or the corresponding differential cross sections of thermal scattering $dR^2_{\alpha\beta}/d\Omega\,d\theta$ in a medium with an arbitrary number of relaxing scalar and symmetric tensor parameters.[8] The corresponding amplification coefficients for stimulated scattering are found with the help of the universal relation[9]

$$g_{\alpha\beta}(\Omega,\theta)=\frac{(2\pi)^3J_L\Omega}{kT\omega_Lk_L{}^2}\frac{d^2R_{\alpha\beta}}{d\Omega\,d\theta}, \tag{1}$$

where $\Omega=\omega_L-\omega_s$ is the difference in frequencies of the incident and scattered light, J_L (W/cm^2) is the intensity of the pump light, $g(\Omega,\theta)$ is the local increase in amplification in cm^{-1}, ω_L and k_L are the frequency and wave number of the pump light wave, k is the Boltzmann constant, and T is the absolute temperature.

On the basis of experimentally known parameters, we limit ourselves to a single relaxation time for the bulk viscosity (one scalar relaxation parameter) and to two relaxation times for the shear viscosity (two tensor relaxation parameters). In such a medium, the spectral density of the light intensity of components scattered at 90° takes the form[8]

$$J_{zz}\sim\frac{1}{i\Omega}\left\{\frac{1}{\Delta}\left[{}^1/_3Xq^2({}^1/_3X-2Y)+Y^2q^2\right]+\frac{m^2}{\lambda_c}+\frac{n_1{}^2}{\lambda_1}+\frac{n_2{}^2}{\lambda_2}-\text{c.c.}\right\}, \tag{2}$$

$$J_{yz}=J_{zx}\sim\frac{1}{i\Omega}\left\{\frac{X^2q^2}{4A}+\frac{1}{2}\left(\frac{n_1{}^2}{\lambda_1}+\frac{n_2{}^2}{\lambda_2}\right)-\text{c.c.}\right\}, \tag{3}$$

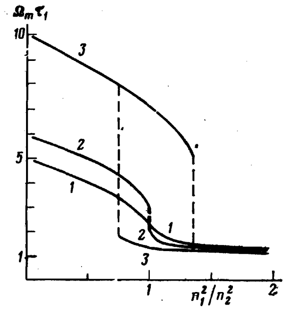

FIG. 1. Dependence of the relative displacement of the maximum $\Omega_m\tau_1$ (or of the maxima $\Omega_{m1}\tau_1$ and $\Omega_{m2}\tau_2$) of the increment g_0 (5) on the parameter n_1^2/n_2^2 or $\tau_1/\tau_2=5$ (curve 1, $\tau_1/\tau_2=6$ (curve 2), $\tau_1/\tau_2=10$ (curve 3).

$$J_{yx}\sim\frac{1}{i\Omega}\left\{\frac{q_2^2X^2}{2\Delta}+\frac{1}{2}\left(\frac{n_1^2}{\lambda_1}+\frac{n_2^2}{\lambda_2}\right)-\text{c.c.}\right\},\tag{4}$$

where $\mathbf{q}=\{k_L,\,-k_L,\,0\}$; $|\mathbf{q}|=2nk_{L0}$; n is the index of refraction of the medium; k_{L0} is the wave number of the pump in vacuum; X, Y are the complex optical moduli of the medium; n_1, n_2, m are the actual optical moduli, which determine the dependence of the permittivity on the relaxation parameters. In addition, we introduce the notation $\lambda_e=1+i\Omega\tau_c$ and $\lambda_c=1+i\Omega\tau_i$, where τ_c is the bulk viscosity relaxation time and $\tau_i(i=1,\,2)$ are the relaxation times of the tensor parameters, $\Delta=-\rho_0\Omega^2+Kq^2=(4/3)\mu q^2$ is the resonant denominator of the Mandel'shtam–Brillouin doublet, $A=\mu q^2-\rho_0\Omega^2$ is the resonant denominator of the shear doublet, K and μ the bulk and shear complex elastic moduli of the medium, and ρ_0 is the mean density of the medium.

The term X^2/Δ in Eq. (4) leads to the appearance of maxima in the intensity distribution at frequencies close to the resonances of the denominator Δ, i.e., to frequencies of the shift of the Mandel'shtam–Brillouin components. The relative shift in the maxima of the Mandel'shtam–Brillouin, Y^2/Δ in the J_{zz} component (2) or X^2/Δ in the J_{yx} component, is due to the strong dispersion of the mechanical-optical coefficient $X(\Omega)$, and in particular, for liquids, $X=0$ at $\Omega=0$.

Far away from the resonances of the denominators of A and Δ, the spectral distribution of the increments of the depolarized scattering are identical for g_{yx} and g_{yz} and with account of the two anisotropy relaxation times, then are written in the form of a sum of two Lorentzians:

$$g_0\approx g_{yx}\approx g_{yz}\approx g_{zx}\approx\frac{\Omega\tau_1n_1^2}{1+\Omega^2\tau_1^2}+\frac{\Omega\tau_2n_2^2}{1+\Omega^2\tau_2^2}.\tag{5}$$

In the general case, the spectral distribution (5) can have one or two maxima, depending on the ratio of the parameters τ_1/τ_2 and n_1^2/n_2^2.

The regions of existence of one and two maxima of the distribution (5) were calculated with a computer (we call these regions 1 and 2, respectively). Inside region 2, the amplification coefficient (5) has two maxima Ω_{m1} and Ω_{m2}, located at frequencies approximately equal to $1/\tau_1$ and $1/\tau_2$, respectively, and in region 1 it has one maximum. In this range of values of the parameters, and also in the region of transition through the boundary from 1 to 2, there is no simple relation between the location of the maxima of the amplification coefficient and the relaxation times. Figure 1 shows the characteristic dependence of the location of the maximum amplification (5) on the value of n_1^2/n_2^2 for τ_1/τ_2 equal to 5, 6, 10. It is obvious that the experimental measurement of the locations of the maxima (or maximum) of the amplification (5) enables us to assess only the absolute values of the quantities n_1 and n_2 and not the signs of these optical moduli. Account of the terms previously-descarded in Eqs. (2), (3), (4) also has a significant effect on the location and value of the maxima (or maximum) of the distribution of the increment of stimulated scattering.

In the range of frequencies where A and Δ have resonances, the spectral distributions (2), (3), (4) can be significantly different from a superposition of two Lorentzian contours. Since only a single relaxation time of the anisotropy has been measured for most liquids at the present time, we tentatively analyze the spectral distributions of the increments in the approximation $\tau_2=0$. The corresponding densities of the amplification coefficients are of the form

$$g_{yz}\sim\frac{n_1^2\tau_1\Omega}{\Lambda_1}\left[1+\frac{\Omega_T^2\tau_1^2(2\Omega^2-\Omega_T^2)}{2\Lambda_1(\Omega^2+\tau^2(\Omega^2-\Omega_T^2)^2}\right]\tag{6}$$

$$g_{yx}\sim\frac{n_1^2\tau_1\Omega}{\Lambda_1}\left[1+\frac{\Omega^2\Omega_T^2}{2\Lambda_1}\frac{(2\tau_1\Delta'-\Delta''(\Omega^2\tau_1^2-1))}{|\Delta|^2}\right],\tag{7}$$

where $\Lambda_1=1+\Omega^2\tau_1^2$; the frequency of the shear wave is $\Omega_T^2=\mu_\infty g^2/\rho$; Δ' and Δ'' are real quantities, determined by the equation

$$\Delta=\Delta'+i\Omega\Delta''=(\Omega^2-\Omega_{\text{MB}}^2)+i\Omega\Delta'';$$

$\Omega_{\text{MB}}=(g^2/\rho)\operatorname{Re}(K+(4/3)\mu)$ is the frequency of longitudinal sound; $\Delta''=(q^2/\rho\Omega)\operatorname{Im}(K+(4/3)\mu)$ is its attenuation.

The spectral densities of the amplification coefficients (6) and (7) were calculated with a computer for salol, nitrobenzene, quinoline and a number of other liquids. The spectral distribution of the amplification coefficient g_{yx} for the range of temperatures where $\Omega_{\text{MB}}\tau\gtrsim1$ has, in addition to the maximum corresponding to the Lorentzian ($\Omega=1/\tau$), a more or less intense second maximum near the frequency Ω_{MB}. At a certain temperature, both maxima have approximately the same value and consequently one can expect the appearance of a doublet in the stimulated-scattering spectrum. For $\Omega_{\text{MB}}\tau_1>1$, the second maximum becomes small in amplitude and vanishes. The value and shift of the second maximum of the increment g_{yx} depend essentially on the relation of the parameters τ, η', $\Omega_{\text{MB}}\Omega_t$ (η' is the bulk viscosity).

The location of the maxima in the spectral distribution of g_{yz} can be found by solving the equation $dg_{yz}/d\Omega=0$. A computer calculation just as in the direct solution of this equation, shows that in the temperature range $\Omega_t\lesssim1/\tau$ the amplification coefficient (6) has a single maximum (Fig. 2) corresponding to $\Omega=1/\tau$. Thus, in this range of temperatures, there are no components of the corresponding fine structure of the Ray-

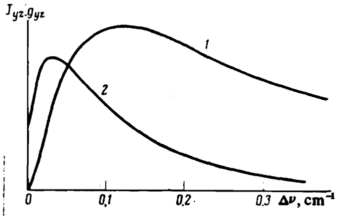

FIG. 2. Spectral distribution of the increment g_{yz} (1) and the thermal scattering cross section (2) for J_{yz} polarization. Quinoline at 20 °C.

leigh line wing in stimulated radiation. In the temperature range $\Omega_t \lesssim 1/\tau$, the effect of the fine structure manifests itself in an insignificant increase in the amplification coefficient g_{yz} relative to g_{yx}. The asymmetries of the increments should lead in the experiment to a relative change in the thresholds of the lines of the RLW in these components of the scattered light. Calculation of the amplitudes of the maxima of the increments in this temperature range showed that, for example in salol, in the temperature region $T \sim 10$ °C, $g_{yz} \sim 2g_{yx}$.

A more detailed analysis of the spectral distributions of the amplification coefficients requires account of two anisotropy relaxation times τ_1 and τ_2 (for definiteness, let $\tau_1 > \tau_2$). In this case,

$$g_{yx} \sim \left\{ \frac{\Omega^3 q^2}{\rho_0 |\Delta|^2} \left[\frac{N_1^2 n_1^2 \tau_1^2}{\Lambda_1^2} M_1 + \frac{2N_1 N_2 n_1 n_2 \tau_1 \tau_2}{\Lambda_1 \Lambda_2} M_{1,2} + \frac{N_2^2 n_2^2 M_2 \tau_2^2}{\Lambda_2} \right] + g_0 \right\} \quad (8)$$

$$g_{yz} \sim \left\{ \frac{\Omega^3 q^2}{\rho_0 |A|^2} \left[\frac{N_1^2 n_1^2 \tau_1^2}{\Lambda_1^2} L_1 + \frac{2N_1 N_2 n_1 n_2 \tau_1 \tau_2}{\Lambda_1 \Lambda_2} L_{1,2} + \frac{N_2^2 n_2^2 \tau_2^2}{\Lambda_2^2} \right] + L_2 \right\};$$

$$\begin{gathered} M_i = 2\tau_i \Delta' + \Delta''(\Omega^2 \tau_i^2 - 1),\ M_{1,2} = (\tau_1 + \tau_2)\Delta' + \Delta''(\Omega^2 \tau_1 \tau_2 - 1), \\ L_i = 2\tau_i A' + A''(\Omega^2 \tau_i^2 - 1),\ L_{1,2} = (\tau_1 + \tau_2) A' + A''(\Omega^2 \tau_1 \tau_2 - 1); \\ A = A' + i\Omega A'',\ \ A' = \mathrm{Re} A,\ A'' = \Omega^{-1} \mathrm{Im}\, A. \end{gathered} \quad (9)$$

It is not possible to find the maxima of Eqs. (8), (9) in analytic form; therefore, all the calculations were carried out with a computer. The difficulty of such calculations lies in the fact that there is no complete set of parameters entering into the formulas as relaxation theory for the liquids which have been studied experimentally, or for those similar to them, and for the calculations one needs not only the parameters but also their temperature dependences. Therefore, we have made a number of simplifications in the calculations. In particular, it has been assumed that the bulk viscosity relaxation time changes with temperature in the same way as the large anisotropy relaxation time. The temperature dependences of the parameters τ_1, τ_2, η_0, n_1^2/n_2^2, μ_∞, n, ρ have been investigated in greatest detail in salol[10] and also in quinoline.[11][2] The bulk viscosity and the compressibility modulus were obtained from acoustic measurements.[12–14] If a standard set of experimental parameters is used, the sign of the ratio n_1/n_2 remains undetermined; therefore, both possibilities were taken into account in the calculations, i.e., $n_1/n_2 > 0$ and $n_1/n_2 < 0$.

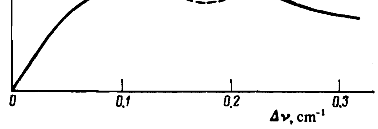

FIG. 3. Spectral distribution of g_{yx} with account of one (dashed line) and two anisotropy relaxation times (continuous line). Quinoline at 20 °C.

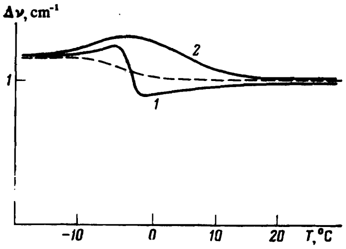

FIG. 4. Calculated temperature dependence of the displacement of the maximum of the fine structure in the J_{yx} component in salol in the range of temperatures $\Omega_{MB}\tau_2 \sim 1$. Curve 1 corresponds to the case $n_1 n_2 < 0$; 2—$n_1 n_2 > 0$. In these calculations, the temperature dependence of the bulk elasticity modulus K was not taken into account. The dashed curve is the displacement of the maximum of the Mandel'shtam–Brillouin component in J_{zz} polarization.

The computed coefficients of amplification of stimulated scattering in the temperature range $\Omega_{MB}\tau_2 \sim 1$, with account of the two relaxation times for quinoline, are shown in Fig. 3. Figure 4[3] shows the results of calculation of the thermal scattering cross section in the spectrum of J_{yx} in salol in the temperature range $\Omega_{MB}\tau_2 \sim 1$ for the cases $n_1 n_2 > 0$ and $n_1 n_2 < 0$. As is seen from Fig. 4, these cases give qualitatively different spectra, namely, for $n_1 n_2 < 0$ the maximum is shifted relative to Ω_{MB} toward the exciting line, and for $n_1 n_2 > 0$, to the contrary, the shift of the maximum of the J_{yx} polarization spectrum is greater than the shift in the Mandel'shtam–Brillouin component of the J_{zz} spectrum. The variation of the parameters of relaxation theory within reasonable limits leads to a change in the absolute value of the shift and the amplitude of the maximum, but does not change the picture of the phenomenon qualitatively.

2. EXPERIMENTAL APPARATUS AND METHOD OF EXPERIMENT

As was pointed out in the Introduction, the basic experimental difficulty in the investigation of the RLW line and its fine structure is that the threshold of this form of stimulated scattering is high in comparison with the thresholds of other nonlinear effects, in particular, Mandel'shtam–Brillouin scattering. Therefore, for the study of weak depolarized types of scattering, the method of polarization-angular selection of the scattered light was used in this research.[5–7] The essence of this method is that the nonlinear interaction region, the dimensions of which are determined by the conditions of focusing of the exciting radiation and the dimensions of the sample investigated, is strongly elongated in the direction parallel to the electric field vector of the pump (we denote this direction by **y**; see Fig. 5). The intensity of the pump in the region of nonlin-

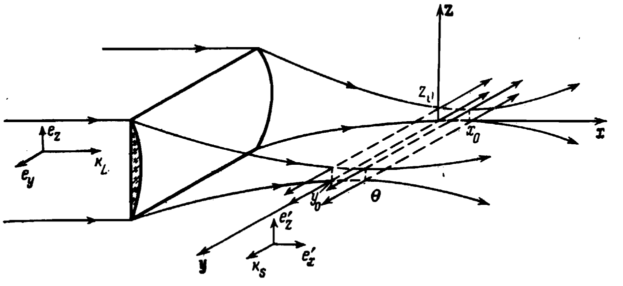

FIG. 5. Diagram of the region of nonlinear interaction for excitation of the RLW at an angle of 90°. The continuous lines with arrows indicate the direction of propagation of the pump light. The region of nonlinear interaction is tentatively assumed to be a parallelepiped (dashed lines); e_z, e_y are unit vectors of polarization of the incident, and e'_z, e'_x of the scattered light; $\mathbf{k}_L$ and $\mathbf{k}_s$ are the wave vectors of the incident and scattered light.

ear interaction is approximately equal to $J_L = W_L/y_0z_0$, where y_0 and z_0 are the dimensions of the region of nonlinear interaction (Fig. 5), and W_L is the radiation power of the pump.

Upon increase in the dimension y_0, the quantity J_L decreases as $1/y_0$. On the other hand, in the y direction (scattering angle 90°), the total gain $G = J_L g y_0 = gW_L/z_0$ does not depend on the quantity y_0. Thus, upon increase in y_0, the amplification for all scattering angles falls off approximately as $1/y_0$,[7] while it remains constant for a scattering angle of 90°. Consequently, by choosing a sufficiently large value of y_0 and orienting the electric vector of the exciting radiation strictly along the **y** axis, we can suppress completely the scalar modes of stimulated scattering and excite stimulated scattering due to low intensity lines of the depolarized light scattering.

The scheme of the experimental apparatus is shown in Fig. 6. In the experiments, a Q-switched ruby laser and an amplifier were used as the pump source. We give the basic characteristics of the pump radiation: power—up to 200 MW, pulse length $\tau_L \sim 20$ nanosec, spectral width $\Delta\nu_L \sim 0.02$ cm^{-1}, angular spread $\sim 10^{-2}$ rad. The pump radiation was focused into the vessel with the scattering liquid V by means of two crossed cylindrical lenses L$_1$ and L$_2$. To broaden the beam in the **y** direction, we used the negative cylindrical lens L$_1$ with focal length $f_1 = -25$ cm. By means of the short-focus positive lens L$_2$ ($f_2 = +5$ cm) the pump radiation was focused along the vertical **z** axis. In this case, the dimensions of the region of nonlinear interaction amounted to ~ 5 cm along the **y** axis. To increase the amplification in the **y** direction, we used a confocal spherical resonator, consisting of two dielectric mirrors with a radius of curvature of 15 cm and reflection coefficient ~ 100%. The axis of the resonator coincided with the **y** direction. Inside the resonator was a circular diaphragm D (Fig. 6) of diameter ~ 3 mm, which limited the angular spectrum of scattered radiation and improved the selection of depolarized modes of stimulated scattering.

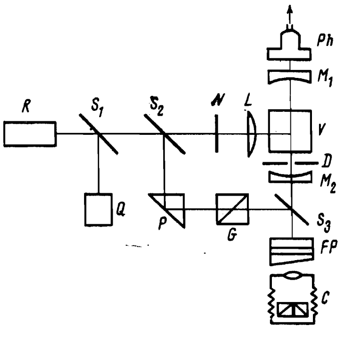

FIG. 6. Experimental apparatus for observation of the RLW at an angle of 90°. R—ruby laser, S$_1$, S$_2$, S$_3$ are glass plates, Q—a calorimeter, M$_1$, M$_2$—mirrors with dielectric coating, L—a cylindrical lens, N—a quartz plate of half wavelength, used for balancing the optical system, D—3-mm diaphragm, P—glass rectangular prism, G—Glan prism, Ph—coaxial photomultiplier connected to a high-speed oscilloscope I-2-7, V—thermostated container with the liquid under study, FP—Fabry-Perot interferometer, C—photocamera with polarizers, which enables the observer to see the scattered light in two mutually perpendicular polarizations.

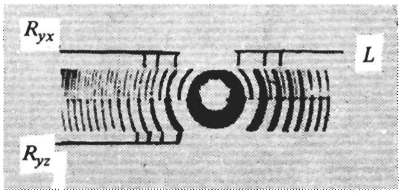

FIG. 7. Spectrum of the components of the RLW line in nitrobenzene (30 °C), L is the spectrum of the line of laser light; R_{yz} and R_{yx} are the components of the RLW line with vertical and horizontal polarizations, respectively.

The spectrum of the scattered radiation was recorded by a Fabry-Perot interferometer at two polarizations, J_{yx} and J_{yz}. By means of the beam-splitting plates S$_2$ and S$_3$ and the prism P, part of the pump radiation was directed into the interferometer FP through a frosted plate and analyzer G in such fashion that the polarization of the pump radiation could be changed (by turning the analyzer G) for convenience of measurement of the line shifts in the spectra corresponding to different polarizations of the scattered light.

Vessels of stainless steel in the form of cubes with four windows of diameter 60 mm were used in the experiments. Above and below the vessel, there were special cavities, connected with the thermostat, to maintain the desired temperature. To remove temperature gradients, the vessel was placed in a special box with heat-insulating walls. The liquids studied were first purified by means of filters and vacuum distillation and then either poured directly or distilled into a dust-free metal cell. This made it possible to supercool the salol 35–40 °C below its crystallization temperature (42°).

3. EXPERIMENTAL RESULTS

The temperature dependence of the spectra of depolarized stimulated light scattering was measured experimentally in quinoline salol, nitrobenzene, and α-chloronaphthalene. The characteristic spectrum of the lines of stimulated scattering, far from the resonances of the denominators of A and Δ, are shown in Fig. 7.

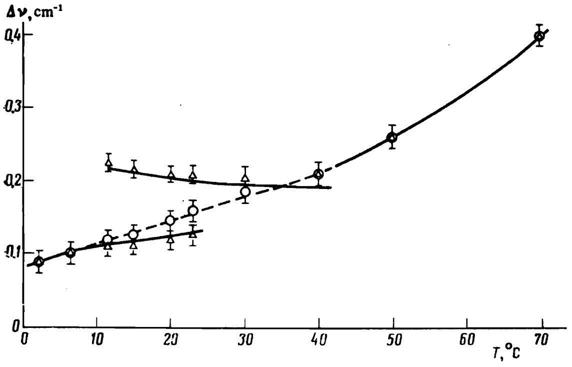

FIG. 8. Dependence of the location of the components of the RLW wing ($\Delta\nu$ in cm^{-1}) on the temperature in quinoline, o—displacement of the RLW line in the J_{yz} component of the scattered light, Δ—in the J_{yx} component, the continuous line is the calculation according to Eq. (7).

In both polarizations of the scattered light, the intensity and line shift of the RLW are approximately the same. The character of the temperature dependence of the line shift of the RLW in the J_{yx} and J_{yz} polarizations of the scattered light is shown in Figs. 8, 9, 10 for the liquids studied. The greatest splitting of the lines of the RLW in the temperature range $\Omega_{MB} \sim 1/\tau$ is observed in the spectra of light scattered in quinoline. The corresponding spectrum for a temperature of 15 °C is shown in Fig. 11. The observed splitting of the lines of the RLW in α-chloronaphthalene is much less than in quinoline and corresponds approximately to the halfwidth of the lines in the RLW spectrum. Actually, in the region of temperatures where $\Omega_{MB} \sim 1/\tau$, the line of the RLW in α-chloronaphthalene in the J_{yx} polarization was not observed.

Another region in which, according to calculations, an asymmetry should appear in the amplification coefficients g_{yx} and g_{yz}, as a consequence of the fine structure is that corresponding to $\Omega_t \sim 1/\tau$. In salol, this region of temperatures corresponds to the supercooled state in the liquid. The spectra of the lines of the RLW for salol are shown in Fig. 13 for measurements at 10 °C. The threshold of the RLW line in the J_{yx} polarization at this temperature is 1.3–1.5 times greater than in the J_{yz} polarization. In the range 2–10 °C, this difference of the thresholds in the experiment agrees with the calculated dependence. The characteristic spectra of broadening of the RLW line in α-chloronaphthalene are shown in Fig. 14.

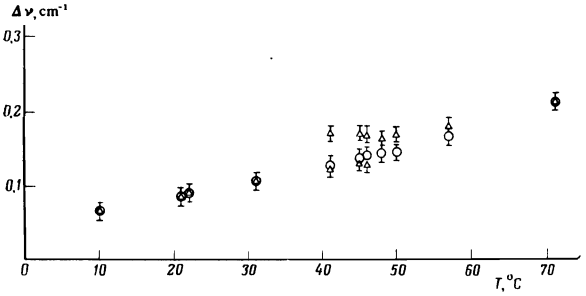

FIG. 9. Temperature dependence of the displacement of the RLW lines in α-chloronaphthalene; o—in the J_{yz} component, Δ—in the J_{yz} component.

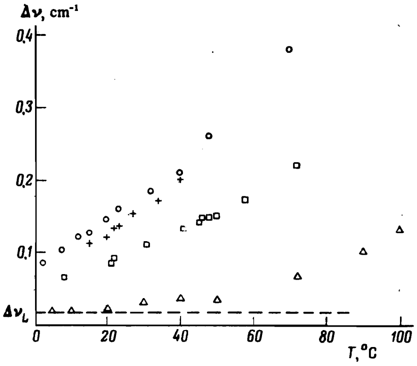

FIG. 10. Temperature dependence of the displacement of the RLW line in four liquids for the R_{yz} component; Δ—in salol, +—in nitrobenzene, o—in quinoline, □—in α-chloronaphthalene. In salol and nitrobenzene the displacement of the RLW lines in both components of the scattered light are identical to all temperatures within the limits of experimental error. The halfwidth of the laser line $\Delta\nu_L$ is shown by the dashed line.

The error in the measurement of the quantity τ in the shift of the RLW line consists of several independent parts. First, the accuracy of the measurements is determined by the width of the RLW line, which has a tendency to increase as the shift from the exciting line increases. This broadening can be due to an increase in the spectral amplification band $\Delta\nu_g$ in the RLW, which is equal to $1/\tau$, i.e., the shift of the RLW line. Moreover, for a given shift, the width of the RLW line depends essentially on the intensity of the pump.

Another source of error is connected with the asymmetries of the contour of the amplification coefficient curve (see Figs. 2, 3), because of which the error in the determination of the actual location of the maximum of the amplification coefficient can also be increased. Finally, as follows from calculations, the maximum of the amplification coefficient (with account of the two relaxation processes) is not necessarily located exactly at the frequency $1/\tau$.

The error in the measurement of the value of the RLW line splitting can also be due to a number of reasons. First, the structure of the characteristic modes of the resonator has an effect on the splitting of the RLW line. In our experiments, the interval of frequencies between the neighboring modes of the spherical

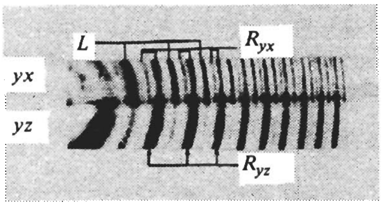

FIG. 11. Interferograms of the spectrum of light scattered in quinoline in J_{yx} and J_{yz} polarizations. The dispersion range of the Febry-Perot interferometer is $\Delta\nu = 0.33$ cm^{-1}; L is the line of the ruby laser, R_{yx} and R_{yz} the RLW lines in the J_{yx} and J_{yz} polarizations, respectively; at $T = 15$ °C, the displacement of the line R_{yz} corresponds to $\Omega = 1/\tau$, the line R_{yx} splits into two lines.

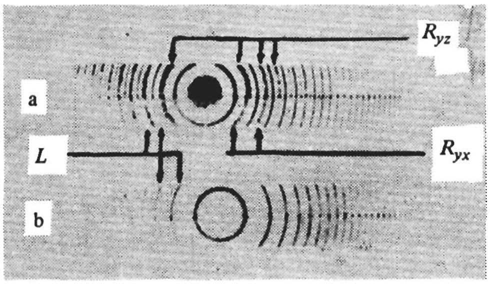

Fig. 12. Asymmetry of the RLW line in α-chloronaphthalene (50 °C). The dispersion range of the interferometer is $\Delta\nu = 0.33$ cm^{-1}; a) R_{yz} is the RLW line corresponding to the vertical direction of the electric vector in the scattered wave, R_{yx} is the vector of the electric field of the scattered wave lies in the plane of scattering (the horizontal plane), L is the spectrum of the laser radiation. In this experiment, the exciting light incident on the chamber was polarized at an angle of 45° to the horizontal plane. The presence of the exciting light in both polarizations simplifies the measurement of the displacements of the components of the RLW line. b) The spectrum of the laser radiation.

confocal resonator amounted to $\delta\nu = 1/2L = 0.025$ cm^{-1}, and had no effect on the value of the RLW line splitting. Thus, when the length of the resonator L changed by a factor of 1.5, the value of the splitting of the RLW wing (in quinoline) remained unchanged. Second, the scalar contribution of the amplification coefficient can distort the picture of splitting of the RLW line. This arises as a consequence of the finite aperture of the pump beam and the beam of the scattered light. At scattering angles 90° ± δ, where $\delta \sim d/L$ is the halfwidth of the aperture and d is the radius of the diaphragm, we have $d/L \sim 10^{-2}$ and, consequently, the value of the scalar contribution of g_{zz} to g_{yx} is $\approx \delta^2 \approx 10^{-4}$.

The possible effect of the finite aperture was verified experimentally by the method of increasing and decreasing the diaphragm radius by several times (Fig. 6). Here the splitting of the RLW line (Fig. 11) or the absence of such (Fig. 7) did not depend on the dimension d and, consequently, the finite aperature of the beams had no effect on the value of the splitting of the RLW line in our experiments. If this were not the case, and if a small part of the increment of the Mandel'shtam–Brillouin stimulated scattering from the g_{zz} polarization of the scattered light were added to the amplification coefficient in the g_{yz} polarization, then the greatest line splitting of the RLW would have arisen in nitrobenzene, and not in quinoline, since it is in nitrobenzene that the threshold of stimulated Mandel'shtam–Brillouin scattering is the lowest of the four liquids studied and is approximately one half that of quinoline.

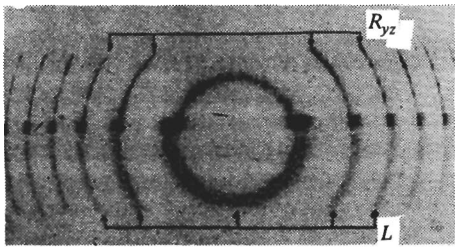

FIG. 13. Asymmetry of the amplification of the RLW line in salol at $t = 10$ °C. The RLW line corresponding to vertical direction of the electric vector in the scattered wave has a large increment and a correspondingly high intensity in the spectrum.

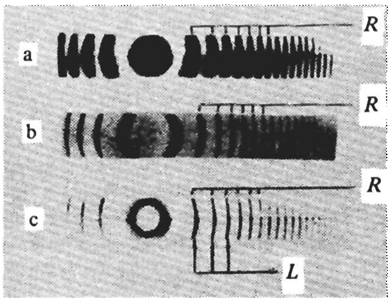

FIG. 14. Broadening of the RLW lines in α-chloronaphthalene with increasing intensity ($c \to b \to a$). L is the line of the radiation of the ruby laser. a) $J_L \sim 1.5 \times 10^9$ W/cm^2, b) $J_L \approx 1.2 \times 10^9$ W/cm^2, c) $J_L \sim 0.5 \times 10^9$ W/cm^2.

4. DISCUSSION OF THE RESULTS

The characteristic features of the spectral distributions obtained in the research admit of a simple qualitative explanation. First, in the range of temperatures in which the location of the maximum amplification of the wing ($\tau_2 = 0$) is near the frequency Ω_{MB}, i.e., $\Omega_{MB} \approx 1/\tau$, we get from Eqs. (6), (7), taking it into account that $\Omega_T \ll \Omega_{MB}$ and $\Omega \sim \Omega_{MB}$,

$$g_{yz} \approx n_1^2 \tau \Omega / \Lambda_1 \tag{9a}$$

$$g_{yx} = g_0 [1 + \Omega_T^2 (\Omega_{MB}^2 \tau_1^2 - 1) / \Lambda_1 \Delta'']. \tag{10}$$

We can thus register the singularity in g_{yx} in the form of an addition to the purely Lorentzian gain contour $g_0 \approx g_{yz}$. At $\Omega_{MB} > 1/\tau$, the addition to the maximum gain is positive and at $\Omega_{MB} < 1/\tau$, it is negative.

An additional maximum of the amplification coefficient and of the line in the spectrum at a frequency close to Ω_{MB} should be observed at $\Omega_{MB} > 1/\tau$. The value of the maximum is determined from the relation (10):

$$\frac{\Omega_T^2}{\Delta''(\Omega_{MB})} = \frac{\eta(\Omega)}{\eta'(\Omega_{MB}) + {}^4/_3 \eta(\Omega_{MB})} = \frac{\alpha'(\Omega_{MB})}{\alpha_{tot}(\Omega_{MB})} \tag{11}$$

and, consequently, the fine structure in the J_{yx} component of the scattered light is determined by the ratio of the shear viscosity to the total viscosity, or the ratio of the partial sound absorption due only to the shear viscosity to the total absorption.

The disappearance of the maximum at $\Omega_{MB}\tau_1 < 1$ can be explained also by considering the exchange of energy (within the framework of molecular theory) between the acoustic and orientation modes.[15] Thus, the greatest part of the increase in stimulated scattering is made by the fine structure in the temperature range in which $\Omega_{MB}\tau_1 \gtrsim 1$. Using these simple considerations on the dependence of the fine structure in the J_{yx} component of the scattered light, with account of the single relaxation time, we can analyze qualitatively the temperature dependence of the spectral increment (8) with account taken of two relaxation times, taking it into consideration that it consists of three terms, each of which tion is similar to (7).

In the range of temperatures $\Omega_{MB}\tau_1 \sim 1$, the fundamental contribution to the fine structure is made by the first term, which is identical with (7) at $\tau_2 \to 0$. Its value is proportional in this case to the ratio of the part of the shear viscosity that relaxes with the time τ_1 to the total viscosity $(\eta' + (4/3)\eta)$ or the ratio of the corresponding sound absorption coefficients. Two other terms in (8) are small at the frequency Ω_{MB}, since $\tau_2 \ll \tau_1$ and are negative. It is essential that account of the second relaxing process can significantly reduce the amplitude of the maximum of the fine structure, since in this case the basic role is played not by the total shear viscosity, which can be measured in independent experiments, but only by that part which relaxes with the time τ_1. In the temperature range

$$\Omega_{MB}\tau_2 < 1 \lesssim \Omega_{MB}\sqrt{\tau_1\tau_2} < \Omega_{MB}\tau_1$$

the principal contribution is made by the term

$$N_1N_2n_1n_2\tau_1\tau_2M_{1,2}/\Lambda_1\Lambda_2.$$

The first term is small, because the quantity $(\Omega_{MB}\tau_1)^4 \sim \Lambda_1^2$ appears in the denominator, and the third term is smaller by a factor of approximately τ_2/τ_1 then the second, and is negative at $\Omega \sim \Omega_{MB}$. The sign of the second term, except for the factor $M_{1,2}$ considered above, is determined by the sign of the product n_1n_2. Here, if $n_1n_2 < 0$, then the displacement of the maximum of the fine structure relative to the positive MB component will be of the other sign, i.e., the maximum will be located closer to the frequency of the exciting light than the MB component, as follows from the calculations.

Finally, in the temperature range $1 \lesssim \Omega_{MB}\tau_2$, the first two terms are small and the basic role is played by the term $N_2^2n_2^2\tau_1^2M_2/\Lambda_2^2$, and the picture of the splitting of the RLW line in this case is about the same as for the first term.

We note that we can find only n_1^2 and n_2^2 from the distribution of the intensity in the wing of the Rayleigh line. The relative displacement of the maximum of the fine structure in the temperature range

$$\Omega_{MB}\tau_2 < 1 < \Omega_{MB}\sqrt{\tau_1\tau_2}$$

offers a unique possibility of determining the sign of the product n_1n_2 or, since the sign of n_1 is known $(n_1 > 0)$, the sign of n_2, directly from the relative displacement of the two lines in the spectrum. As long as the sign of this coefficient is chosen either arbitrarily[10] in these calculations, or both variants are considered,[14] then, as the calculations carried out by Zubkov, Rozhdestvenskiĭ and Ramanov have shown,[11] the spectrum in the J_{yz} component can be written both with $n_2 > 0$ and with $n_2 < 0$. The mechanism of the second relaxation process is actively discussed in the literature at the present time; therefore, the determination of the sign of n_2 can significantly ease the search and construction of corresponding molecular model.

It is shown that the sign of n_2 is more conveniently determined in experiments on the thermal scattering of light, while in the measurement of the temperature dependence of τ, it is more effective to use the spectra of stimulated light scattering. In the investigation of the spectral lines of the scattered light with a complicated intensity distribution, experiments on the thermal and stimulated light scattering can complement each other, and improve the accuracy of the result.

[1)] The first subscript of J denotes the direction of the electric vector of the light wave of the exciting light, and the second that of the scattered light. The exciting light is propagated along the x axis.

[2)] The complete set of parameters describing the relaxation of the two tensor parameters has been measured only at a single temperature.

[3)] The temperature dependence of the shift of the component J_{zz} has been normalized to unity.

---

[1] I. L. Fabelinskiĭ, Izv. Akad. Nauk SSSR, ser. fiz. **17**, 538 (1953).

[2] I. L. Fabelinskiĭ, Molecular Scattering of Light (English translation, Plenum Press, 1968).

[3] V. S. Starunov, E. V. Tiganov and I. L. Fabelinskiĭ, Pis. Red. Zh. Eksp. Teor. Fiz. **5**, 317 (1967) [JETP Lett. **17**, 260 (1967)].

[4] G. I. A. Stigman and B. P. Stoicheff, Phys. Rev. Lett. **21**, 202 (1968).

[5] D. V. Vlasov and I. L. Fabelinskiĭ, Pis. Zh. Eksp. Teor. Fiz. **17**, 476 (1973) [JETP Lett. **17**, 342 (1973)].

[6] D. V. Vlasov, Candidate's dissertation, Phys. Inst. Acad. Sci., 1973.

[7] D. V. Vlasov, Kvant. Elektr. (Moscow) **2**, 1439 (1975) [Sov. J. Quantum Electron. **2**, 772 (1975)].

[8] S. M. Rytov, Zh. Eksp. Teor. Fiz. **58**, 2154 (1970) [Sov. Phys. JETP **31**, 1163 (1970)].

[9] B. YA. Zel'dovich and I. I. Sobel'man, Usp. Fiz. Nauk **101**, 3 (1970) [Sov. Phys. Usp. **13**, 307 (1970)].

[10] V. S. Starunov and I. L. Fabelinskiĭ, Zh. Eksp. Teor. Fiz. **66**, 1740 (1974) [Sov. Phys. JETP **39**, 854 (1974)].

[11] L. A. Zubov, N. B. Rozhdestvenskiĭ and V. P. Romanov, Zh. Eksp. Teor. Fiz. **65**, 1782 (1973) [Sov. Phys. JETP **38**, 891 (1974)].

[12] G. I. A. Stigman, Light scattering from liquids. Ph.D. thesis.

[13] E. V. Tiganov, Trudy (Works) Fiz. Inst. Akad. Nauk **58**, 42 (1972).

[14] Chang-Hyum Chung and Sidney Yip, Phys. Rev. **4**, 928 (1971).

[15] N. D. Gershan and I. Oppenheim, Physica **71**, 134 (1974).

Translated by R. T. Beyer