

DETERMINATION OF THE VELOCITY AND ABSORPTION OF HYPERSOUND  
IN VISCOUS LIQUIDS ON THE BASIS OF SCATTERED LIGHT SPECTRA

L. M. SABIROV, V. S. STARUNOV, and I. L. FABELINSKIĬ

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

Submitted August 3, 1970

Zh. Eksp. Teor. Fiz. 60, 146-159 (January, 1971)

The fine structure of the Rayleigh line wing<sup>[6]</sup> in salol and benzophenone is studied experimentally under conditions when the viscosity of the substances is varied by several orders of magnitude. Two branches of transverse hypersonic oscillations are observed in the liquid. One is a low temperature ("normal") branch in which the "transverse" components in the scattered light move apart with lowering of temperature; the other is a high temperature ("anomalous") branch in which the components move together with lowering of the temperature. The propagation velocities of the transverse hypersonic oscillations are measured. The velocities and absorption coefficients are measured for longitudinal hypersonic waves in salol, benzophenone and triacetin. A series of experimental problems are discussed.

It is well known that much unique information on hypersonic propagation can be obtained from the study of the scattered light spectra.<sup>[1-5]</sup> The discovery of the fine structure of the Rayleigh line wing<sup>[6-8]</sup> and the subsequent experimental and theoretical study of the new phenomenon<sup>[9-16]</sup> have made it possible to understand this phenomenon and to investigate the propagation not only of longitudinal but also transverse hypersonic waves in liquids<sup>[17]</sup> over a wide range of variation of the viscosity.

The phenomenon of the fine structure of the Rayleigh line wing (WFS) consists of the following. A doublet is observed in the spectrum of depolarized scattered light under certain conditions. This phenomenon has already been observed in several dozen liquids.<sup>[14-16]</sup> In this paper, we give the results of the experimental study of the kinetics of the fine structure of the Rayleigh line wing in salol and benzophenone, when their viscosity was changed over several orders of magnitude.

The velocity and the absorption coefficient of longitudinal hypersonic have been measured in these media and in triacetin of varying viscosity. The completed investigations have shown that relaxation theory<sup>[18]</sup> cannot explain sound propagation in viscous media.<sup>[19,20]</sup> The nonlocal theory of sound propagation of Isakovich and Chaban<sup>[21]</sup> was specially developed by them in order to describe the observed regularities, and the formulas of this theory actually describe the phenomenon. Therefore, we shall compare our results of the measurement of the velocity and the absorption coefficient of longitudinal hypersonic with the theory of Isakovich and Chaban.

1. BASIC THEORETICAL REPRESENTATIONS AND RELATIONS

A. Theory of the Fine Structure of the Rayleigh Line Wing

We have shown<sup>[11]</sup> that all the features of the complex picture of the temperature dependence of the distance between the "transverse" components can be explained if we assume that the subsidence of the fluctuations of the anisotropy is characterized by two relaxation times. Attempts have been made by other authors

to explain this phenomenon by the introduction of two relaxation times for the anisotropy.<sup>[12-13]</sup> The general form of light scattering from fluctuations of the anisotropy with account of two relaxation times was studied by Rytov.<sup>[22,23]</sup> We shall give the basic results of the Rytov theory here. This is needed for comparison with our results (Sec. 3).

The intensity distribution for any scattering angle  $\vartheta$  for the scattered radiation  $I_{z\Gamma}$ , which is polarized in the plane of scattering (the exciting light is polarized along the z axis) and for the scattered radiation  $I_{yZ}$ , which is polarized perpendicular to the scattering plane, is determined by the expression

$$I_{z\Gamma} = I_{yZ} = -\frac{kT}{2\pi i\omega} \left\{ \left[ \frac{i\omega}{(1+i\omega\tau_1)(1+i\omega\tau_2)} \times \frac{q_z^2}{4\rho} \frac{(2\epsilon_0 M \eta_0 + i\omega X_\infty \tau_1 \tau_2)^2}{\eta_0 q^2 / \rho_0 + i\omega \omega_T \tau_1 \tau_2 + i\omega(1+i\omega\tau_1)(1+i\omega\tau_2)} - \text{c.c.} \right] - i\omega \left( \frac{n_1^2 \tau_1}{1+\omega^2 \tau_1^2} + \frac{n_2^2 \tau_2}{1+\omega^2 \tau_2^2} \right) \right\} \quad (1)$$

Here we have introduced the following notation:  $\omega$  is the variable frequency, measured from the frequency of the exciting light,  $\tau_1$  and  $\tau_2$  are the relaxation times,  $q_z^2 = k^2 \sin^2 \vartheta$ ,  $q^2 = 4k^2 \sin^2 (\vartheta/2)$ ,  $k$  is the wave number of the exciting radiation,  $\epsilon_0$  the dielectric constant,  $M$  the Maxwell constant,  $\omega_T = (\mu_\infty / \rho)^{1/2} q$ ,  $n_1$  and  $n_2$  constants,  $\mu_\infty$  and  $X_\infty$  the limiting values of the shear modulus  $\mu$  and the photoelastic coefficient  $X$  as  $\omega \rightarrow \infty$ . The following expressions hold for  $\mu$  and  $X$ :

$$\mu = \frac{i\omega(\eta_0 + i\omega\mu_\infty\tau_1\tau_2)}{(1+i\omega\tau_1)(1+i\omega\tau_2)}, \quad X = \frac{i\omega(2\epsilon_0 M \eta_0 + i\omega X_\infty \tau_1 \tau_2)}{(1+i\omega\tau_1)(1+i\omega\tau_2)} \quad (2)$$

The static value of the viscosity  $\eta_0$  satisfies the condition (for  $\tau_1 \gg \tau_2$ )

$$\mu_\infty \tau_2 \leq \eta_0 \leq \mu_\infty \tau_1 \quad (3)$$

For  $\tau_1 = \tau_2$  and for the angle  $\vartheta = 90^\circ$ , the expression (1) for  $I_{z\Gamma} + I_{yZ}$  coincides with the corresponding formulas of the Leontovich theory<sup>[17]</sup> for the spectral distribution of the light intensity scattered by the fluctuations of the anisotropy with account of only one relaxa-

tion time.<sup>1)</sup> It follows from Eq. (1) that discrete components are also possible in the scattered light, in addition to the Rayleigh line wing with a maximum at the frequency of the exciting radiation (a component of the form  $\text{const}(1 + \omega^2\tau_1^2)^{-1}$ ). This is possible if the cubic polynomial in the square brackets in the denominator of the first component in (1) has a pair of complex conjugate roots

$$i\omega_m = \omega_r(-\zeta \pm i\Delta). \quad (4)$$

In this case, one should observe two Lorentz lines, which are shifted relative to the frequency of the exciting light by a quantity  $\omega_m = \pm\omega_T\Delta$  and which have the halfwidth  $2\delta = 2\zeta\omega_T$ .

For the frequently encountered case  $\epsilon = \tau_2/\tau_1 \ll 1$  the solution of the cubic polynomial leads to the following expressions for  $\Delta$  and  $\zeta$ :

$$\Delta = \frac{\sqrt[3]{3}}{2}(u-v), \quad \zeta = \frac{u+v}{2} + \frac{1}{3}\gamma^{\frac{2}{3}}, \quad (5)$$

where  $u$  and  $v$  are determined by the formulas<sup>2)</sup>

$$\left. \begin{aligned} u \\ v \end{aligned} \right\} = \pm \frac{1}{3} \left\{ \sqrt[3]{3} \left[ \frac{1}{3}(6\beta+1)\gamma^2 + (27\beta^2-1)\gamma \right. \right. \\ \left. \left. + 1 - \frac{1}{4}\gamma^3\epsilon^2 \pm \gamma^{\frac{2}{3}}(\gamma+27\beta) \right]^{\frac{1}{3}} \right\}^{\frac{1}{2}}. \quad (6)$$

Here we have introduced the notation

$$\gamma = \frac{1}{\omega_r^2\tau_2^2}, \quad \beta = \frac{1}{2} \left( \frac{\eta_0}{\mu_\infty\tau_1} - \frac{1+\epsilon}{3} \right). \quad (7)$$

It follows from (3) that for  $\epsilon \ll 1$ , the following condition should be satisfied:

$$-1/6 \leq \beta \leq 1/3. \quad (8)$$

From (5) and (6), for  $\gamma \ll 1$  (low temperatures), we get  $\Delta \approx 1$  and  $\omega_m \approx \omega_T$ , i.e., the result which also follows from the theory of Leontovich. In the case  $\gamma \gg 1$  (high temperatures), we get for  $\Delta$

$$\Delta = \left( \frac{6\beta+1}{3} - \frac{1}{4}\gamma\epsilon^2 \right)^{\frac{1}{2}}. \quad (9)$$

if  $\gamma \gg 1$  as before. In order that the doublet exist for  $\gamma \gg 1$ , we need, in addition to (8), the satisfaction of the condition  $2\Delta^2 > 0$  or, in other words,

$$\eta_0\tau_1 > \rho/4q^2. \quad (10)$$

For wavelength of the exciting light  $\lambda = 6328 \text{ \AA}$ ,  $\vartheta = 90^\circ$ ,  $\rho \sim 1$  and  $n \sim 1.5$ , this condition has the form  $\eta_0\tau_1 > 5 \times 10^{-12} \text{ g/cm}$ . In this case, if the left hand side of the inequality (10) is much greater than the right hand side, we get  $\omega_m \approx (\eta_0/\tau_1\rho_0)^{1/2}q$  for the shift of the WFS components for  $\gamma \gg 1$  and consequently, if we set  $\tau_1 \sim \eta_0/\mu_1$ , then  $\omega_m$  is dependent on the "partial" shear modulus associated with long relaxation time, as was shown by us earlier.<sup>[11]</sup>

According to the Rytov theory, the dependence of  $\Delta$  on  $\gamma$  for different values of  $\beta$  should have the form

<sup>1)</sup>For  $\tau_1 = \tau_2 = \tau$  we should have [23]  $\mu_\infty\tau = \eta_0$ ,  $X_\infty\tau = 2\epsilon_0M\eta_0$ ,  $n_1^2 = n_2^2 = X_\infty^2/4\mu_\infty$ .

<sup>2)</sup>In his approximations, Rytov [23] set  $\epsilon = 0$  and, consequently,  $\gamma^2\epsilon^2 = 0$ . However, in the real case (for example, salol), in spite of the fact that  $\epsilon \ll 1$ , the value of  $\gamma$  for high temperatures is so great that one cannot neglect the term  $\frac{1}{4}\gamma^3\epsilon^2$ .

shown in Fig. 3 of [23]. It follows from this drawing that for  $\beta < -1/9$  there are two "branches" of the dependence of  $\Delta$  on  $\gamma$ . For arbitrary  $\beta$  and  $\gamma < 3$ , the value of  $\Delta$  increases with decrease in  $\gamma$  (the low temperature branch). For  $\beta \leq -1/9$  there is also a range of values of  $\gamma$  ( $\gamma > 3$ ) where  $\Delta$  increases with increase in  $\gamma$  (the high temperature branch). For the values  $\beta < -1/9$ , the doublet will not be observed in the intermediate range of values of  $\gamma$ .

## B. Nonlocal Theory of the Propagation of Longitudinal Hypersound

Basing their work on the results of the experimental investigation of hypersonic propagation in strongly viscous liquids<sup>[19,20]</sup> and starting from the assumption of the microinhomogeneous structure of such media, Isakovich and Chaban constructed a nonlocal theory of sound propagation.<sup>[21]</sup> All the known experimental results of the study of the acoustic properties of liquids are well described by the formulas of this theory.

Theory gives the following expression for the velocity of hypersound  $V$  and the absorption coefficient  $\alpha$ :

$$\frac{1}{V} - i\frac{\alpha}{\omega} = \frac{1}{V_\infty} \left[ 1 - i\frac{V_\infty^2 - V_0^2}{V_0^2} F(\omega\tau) \right]^{\frac{1}{2}} \quad (11)$$

Here  $\tau = a^2/2D$  is the time of diffusion equalization,  $a$  the radius of the microinhomogeneities,  $D$  the diffusion coefficient,  $F(\omega\tau)$  a function of the form

$$F(\omega\tau) = \frac{1}{2}(1+x^{-1})(x - \text{tg } x)(1 + \text{tg } x)^{-1}, \\ x = (1-i)(\omega\tau)^{\frac{1}{2}}. \quad (12)$$

The graphs of the dependence of the real and imaginary parts of the function  $F(\omega\tau)$  on  $\omega\tau$  are given in [19-21]. The equalization time  $\tau$  in the simplified variant of the theory is equal to

$$\tau = \frac{5}{3} \frac{V_\infty^2}{V_0^2} \frac{(\eta_0 + \frac{1}{3}\eta')}{\rho(V_\infty^2 - V_0^2)} = \frac{5}{3} R \frac{V_\infty^2\eta_0}{V_0^2(V_\infty^2 - V_0^2)}, \quad (13)$$

where  $\eta'$  is the coefficient of bulk viscosity,  $R = (\alpha/\alpha\eta)\omega\tau \ll 1$  the ratio of the sound absorption coefficient for  $\omega\tau \ll 1$  and the absorption coefficient determined by  $\eta_0$ . By knowing  $V_\infty$  and  $V_0$  for each value of the temperature, one can compute  $\tau$  from (13) if the value of  $R$  is known.

In the comparison of the theory with the results of experiment, we also needed the following formulas, which follow from (11):

$$\text{Re } F(\omega\tau) = \frac{2\alpha}{\omega} \frac{V_0^2}{V} \frac{V_0^2}{V_\infty^2 - V_0^2}, \\ \text{Im } F(\omega\tau) = \left( 1 - \frac{V_0^2}{V^2} + \frac{V_0^2\alpha^2}{\omega^2} \right) \frac{V_0^2}{V_\infty^2 - V_0^2}. \quad (14)$$

For  $\omega\tau \gg 1$ , it follows from the asymptotic value of the function  $F(\omega\tau)$ , and (11) and (13), that

$$\tau = \left( \frac{3}{8} \frac{V_\infty^2 - V_0^2}{V_0^2} \frac{V_\infty}{V - V} \right)^2 \frac{1}{\omega}, \quad (15)$$

$$V = V_\infty \left( 1 - \frac{3}{8} \frac{V_\infty^2 - V_0^2}{V_0^2} \frac{1}{\sqrt{\omega\tau}} \right), \quad (16)$$

$$\frac{\alpha}{\omega} = \frac{3}{8} \frac{V_\infty^2 - V_0^2}{V_0^2 V_\infty} \frac{1}{\sqrt{\omega\tau}}. \quad (17)$$

All the relations given above follow from the simplified variant of the nonlocal theory, in which the effect

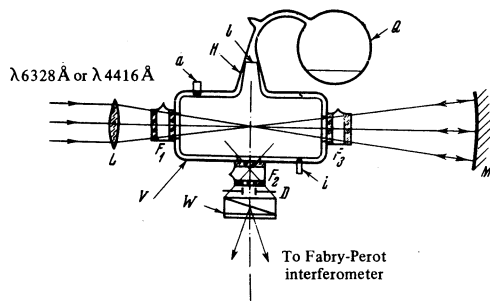


FIG. 1. Schematic diagram of the vessel for a scattering medium in an external optical resonator: V—cylindrical part of the vessel with double walls for transmission through thermostatic material, H—horn-shaped appendage for producing a dark background, F<sub>1</sub>, F<sub>2</sub> and F<sub>3</sub>—windows and vacuum pockets to prevent steaming of the glass, L—objective ( $f=28$  cm, aperture  $1^\circ$ ), M—spherical mirror, Q—vessel for distilling the liquid, i and a—entrance and exit of the thermostatic material, l—level of the liquid, D—diaphragm. The entire vessel was roughened, except for the window, W—Wollaston prism.

of the shear deformations on the propagation of longitudinal hypersound is not taken into account. In the general theory of Isakovich and Chaban,<sup>[21]</sup> shear elastic deformations are considered; however, the comparison of our experimental results with this theory has not been accomplished, because of the absence of data on the temperature dependence of the limiting values of the shear moduli  $\mu_\infty$  ( $\omega \rightarrow \infty$ ) and  $\mu_0$  ( $\omega \rightarrow 0$ ).

## 2. METHODS OF MEASUREMENT

In principle, the scheme of our apparatus did not differ in its general form from that described earlier.<sup>[1, 6-8, 10, 11]</sup>

The scattered light was excited by light from gas lasers, either from the mixture Ne-He,  $\lambda = 6328 \text{ \AA}$  (40 mW), or from the mixture He-Cd<sup>[14]</sup>,  $\lambda = 4416 \text{ \AA}$  (20 mW)<sup>[24]</sup> operating in a single mode regime. A special vessel (Fig. 1) permitted change of temperature of the studied liquid over a wide range and, in the case of necessity, permitted us to carry out repeated purification of the observed liquid.<sup>[1]</sup> The full aperture of the apparatus was limited by the dimensions of the diaphragm D or of the Wollaston prism W, and did not exceed 0.03 radian.

By way of special apparatus, we used a Fabry-Perot interferometer. The distance between the mirrors was varied, depending on the character of the problem, from 0.3 to 10 mm. The reflection coefficient of the mirrors was 96%. The focal length of the objective ranged from 600 to 1200 mm.

The method of determination of the velocity of hypersound follows from the well-known formula of Mandel'shtam-Brillouin:<sup>[1]</sup>

$$V = c\Lambda\nu = c\Delta\nu / 2nv \sin(\vartheta/2), \quad (18)$$

where  $c$  is the speed of light,  $\Delta\nu$  the displacement of the components. The scattering angle  $\vartheta$  was established to within  $\pm 0.5$  deg. The amplitude absorption coefficient  $\alpha$  was determined from the width  $\delta\nu$  of the Mandel'shtam-Brillouin components (MBC)<sup>[1]</sup>

$$\alpha = \pi c \delta\nu / V. \quad (19)$$

For the conditions of our experiment, the effect of finite aperture on the width and position of the MBC and WFS was negligibly small.<sup>[1, 25]</sup> The relative error of measurement of the longitudinal and transverse hypersound was chiefly determined by the roughness of the measurement of  $\Delta\nu$ . For longitudinal hypersound, it amounted to 0.5%, for transverse (low temperature branch)—3–5%, and for the components of the WFS (high temperature branch)—10%. The error in the determination of the MBC was 20–30%. Present-day photoelectric methods of recording<sup>[14, 26]</sup> have many advantages over the photographic method and we did not use it in this research for reasons that did not depend on us.

The anisotropy relaxation time was determined from the intensity distribution in the  $I_{yx}$ -polarization component. The work was carried out with a Fabry-Perot interferometer and in light not decomposed by polarization; the photos were taken with a spectrograph (26  $\text{\AA}/\text{mm}$ ). In both cases, the determination of  $\tau_1$  and  $\tau_2$  was made by the well-known method of reduction of the experimental data,<sup>[1, 17]</sup> according to which the dependence of the value of the reciprocal intensity  $I^{-1}$  as a function of  $\omega^2$  was constructed. Then the relaxation time will be expressed in the following way:<sup>[1]</sup>

$$\tau = \left[ \frac{1}{I_0} \frac{d(I^{-1})}{d(\omega^2)} \right]^{-1/2}.$$

The use of the photoelectric method of recording undoubtedly makes it possible to increase the accuracy of the measurements, by at least an order of magnitude.

## 3. RESULTS OF THE INVESTIGATION OF THE PROPAGATION OF TRANSVERSE HYPERSOUND IN LIQUIDS

### A. Dependence of the Distance between the Components of the Fine Structure of the Wing on the Wavelength of the Exciting Radiation

The phenomenon of the WFS is due to the modulation of the scattered radiation from the oriented microinhomogeneities brought about by shear deformations (transverse hypersound). Such an explanation, advanced by us in our first research,<sup>[6]</sup> was confirmed by a study of the angular dependence of the distance between WFS components<sup>[6-9]</sup> and by polarization studies of this phenomenon.<sup>[6-10]</sup>

A new confirmation of the proposed interpretation has been obtained in the present research. We measured the distance between the components of the WFS in quinoline for two wavelengths of the scattered light  $\lambda = 6328 \text{ \AA}$  and  $\lambda = 4416 \text{ \AA}$ . These distances were shown to be equal to 0.07 and 0.1  $\text{cm}^{-1}$ , respectively. Their ratio amounts to 0.70. If the WFS arises as a result of the modulation of the scattered light by transverse waves, then, just as in the case of longitudinal waves (see Eq. (18)), the distance should be proportional to the frequency or inversely proportional to the wavelengths of the exciting light. (In the latter case, the difference in the refractive indices is neglected.) Such a ratio is equal to 0.69. For a control, we measured the ratio of the MBC distances for the same wavelengths; this amounted to 0.68. Thus we obtained yet another proof of the validity of the given interpretation.

**B. Temperature Dependence of the Distance between the Transverse Components**

The dependence of the distance between the transverse components  $2\Delta\nu_m$  on the temperature in salol and benzophenone was determined under conditions when the viscosity of the latter was varied from  $10^{-2}$  poise up to the viscosity of the vitreous state. The transverse doublet was photographed in the  $I_{ZX}$  polarization. Figure 2 shows the dependence of  $2\Delta\nu_m$  on the temperature for salol and benzophenone.

In the temperature range 120–46°C the WFS doublet was observed in salol. The distance between the components of the doublet decreased with decrease in temperature. In the given temperature range, the viscosity changed from  $10^{-2}$  to  $8.5 \times 10^{-2}$  poise. In the temperature range –2– +45°C, the transverse component could not be recorded. Beginning with –2.5°C ( $\eta_0 \sim 4.5$  poise) the transverse components again appeared and the distance between them increased with decrease in temperature and increase in viscosity (for  $t = -48^\circ\text{C}$ , the viscosity  $\eta_0 \sim 10^9$  poise).

In the range of low temperatures, from –2.5 to –62°C, we observed a triplet—two transverse components and a central line along with the transverse doublet (WFS), which is observed at high temperatures. For low temperatures, the character of the intensity distribution in the scattered light in the  $I_{ZX}$  polarization is similar to the distribution of light scattered by glass in this same polarization component.

Thus, two “branches” are observed in the temperature dependence of the distance between the transverse components. In the low temperature branch the depen-

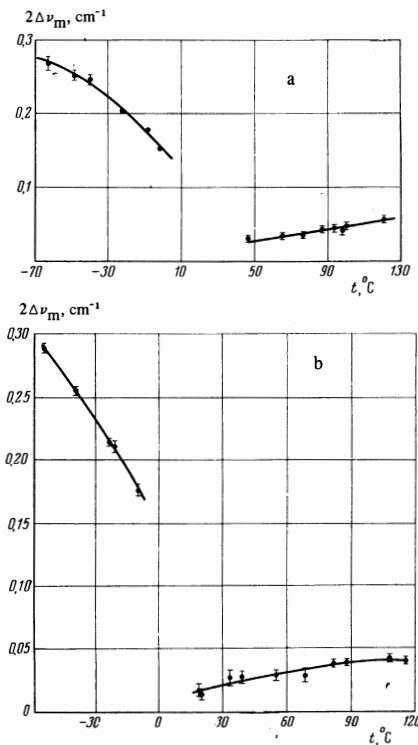


FIG. 2. Temperature dependence of the distance  $2\Delta\nu_m$  between the components of the transverse doublet; a—in salol, b—in benzophenone.

dence of  $\Delta\nu_m$  on the temperature is in agreement with the Leontovich theory,<sup>[17]</sup> which takes into account a single relaxation time. In the high temperature branch, the dependence  $\Delta\nu_m(t)$  is not included in the framework of this representation. The same picture of the temperature dependence of  $2\Delta\nu_m$  was also observed in benzophenone (Fig. 2b).

**C. Measurement of the Parameters Necessary for Comparison of Experimental Results with Theory**

In Eqs. (5) and (6), which follow from the Rytov theory, and which describe the observed phenomena, we have the parameters  $\epsilon$ ,  $\gamma$ , and  $\beta$ . Consequently, for comparison with the theory of the observed dependence of the distance between the transverse components on the temperature, it is necessary to know the temperature dependence of  $\eta_0$ ,  $\rho$ ,  $n$ ,  $\omega_T$  (or  $\mu_\infty$ ),  $\tau_1$  and  $\tau_2$ .

The temperature dependence of  $\rho$ ,  $n$  and  $\eta_0$  for salol are known from the literature.<sup>3)</sup>

The limiting value of  $\mu_\infty$  or  $\omega_T$  ( $\omega \rightarrow \infty$ ) was determined by extrapolation of the measurements of  $\omega_T$  at low temperatures to the region of high temperatures. We note also that for high temperatures (the region of most interest to us), when  $\gamma \gg 1$ , the distance between the components depends little on  $\mu_\infty$ . Actually, in this case, as follows from (9), it should be

$$\omega_m \approx \left( \frac{q^2 \eta_0}{\rho \tau_1} - \frac{1}{4\tau_1^2} \right)^{1/2} \quad (20)$$

For the determination of the anisotropy relaxation times we can use the intensity distribution in the Rayleigh line wing ( $I_{YX}$  polarization).

It is known from the previous investigations<sup>[1,25,28]</sup> that the intensity distribution in the Rayleigh line wing in liquids has a complicated character, which is represented schematically in Fig. 3. In salol, the parts of the wing dD and bD were studied previously by one of us.<sup>[1,28]</sup> In the present research, we studied the temperature dependence of the distribution of the intensity in the parts AB ( $0-0.15 \text{ cm}^{-1}$ ) and Cd ( $0.8-5 \text{ cm}^{-1}$ ) with the help of Fabry-Perot interferometers. The relaxation time  $\tau_1$  was determined from the portion of the wing AB by the method described in Sec. 2, and the relaxation time  $\tau_2$  was determined from the linear portion bd. The part of the wing Bb forms the transitional region from the part of the wing AB that is close to the unperturbed line to the further region of the wing bE. The relaxation time  $\tau_2$ , which is determined from the part of the wing bd, was larger than the relaxation time determined from

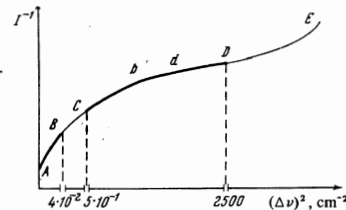


FIG. 3. Dependence of the reciprocal of the intensity on the square of the frequency in the Rayleigh line wing in salol.

<sup>3)</sup> The viscosity of salol in the temperature range 40–100°C was measured by us on a Keppler viscometer, since we could not find reliable data for these temperature.

Table I. Parameters necessary for the calculation of the temperature dependence of the distance between the transverse components in salol

$t, ^\circ\text{C}$	$\rho, \text{g/cm}^3$	$n$	$\eta,$ centipoise	$\tau_1 \cdot 10^{10}, \text{sec}$	$\tau_2 \cdot 10^{10}, \text{sec}$	$\mu_{\infty} \cdot 10^{-10},$ dyne/cm <sup>2</sup>	$\gamma \cdot 10^{-2}$
100	0.986	1.558	3	1.2	1.52	1.30	7.26
90	1.020	1.562	4	1.4	1.62	1.35	6.09
78	1.056	1.568	5	1.7	1.8	1.40	4.74
65	1.10	1.573	6.5	2.1	2.08	1.45	3.55
50	1.15	1.580	8	3.0	2.5	1.55	2.46
40	1.181	1.584	9.1	4.0	2.9	1.56	1.83
30	1.216	1.589	14	6.1	4.5	1.60	0.77
20	1.25	1.593	39	17	12.4	1.64	0.10
10	1.28	1.597	71	31	23	1.68	$3 \cdot 10^{-2}$
0	1.312	1.601	316	140	100	1.72	$1.54 \cdot 10^{-2}$

Note: It is assumed that the limiting value of the velocity of transverse sound  $V_T(\omega \rightarrow \infty) = 1120 \text{ m/sec}$  does not depend on the temperature. For  $t < 40^\circ\text{C}$ , it is assumed that  $\tau_1$  and  $\tau_2$  vary in proportion with the viscosity. The viscosity for  $t < 40^\circ\text{C}$  is determined by the extrapolation formula  $\ln \eta = -11.45 + 1.01 \times 10^3/(T-193)$ , where  $T$  is the absolute temperature.

the part dD.<sup>4)</sup> [28] We attribute such a departure to the fact that the effect of inertia is already seen in the part dD,<sup>5)</sup> [1,25] and, if we take this into account, then the results of [28] will possibly come close to the results obtained here.

Another possibility is that the process of subsidence of the fluctuations of the anisotropy is characterized not by two, but by a larger number of relaxation times. Leaving the discussion of this question to the obtaining of more detailed results of the investigation of the entire Rayleigh line wing (AE), we note that  $\tau_2$ , as follows from (20), has no significant effect on the distance between the WFS components at high temperatures ( $\gamma \gg 1$ ). This effect will also be small for  $\gamma \ll 1$ . But, of course, the temperature region in which the high temperature branch of the dependence of  $2\Delta\nu_m$  on  $t$  goes over into the low-temperature branch does depend on the quantity  $\tau_2$ .

We measured the relaxation times  $\tau_1$  and  $\tau_2$  at temperatures of 43, 78 and  $120^\circ\text{C}$ . In this range of temperatures, both  $\tau_1$  and  $\tau_2$  vary approximately linearly with change in temperature. The time  $\tau_1$  decreases somewhat more rapidly than the viscosity, and  $\tau_2$  more slowly, although the difference is not large. Thus, all the necessary parameters are known to us in the temperature range  $40\text{--}120^\circ$ . For lower temperatures, measurement of  $\tau_1$  and  $\tau_2$  encounters great difficulties. In the region of temperatures  $43\text{--}78^\circ\text{C}$ , the times  $\tau_1$  and  $\tau_2$  change in approximate proportion with the viscosity. Therefore, for  $t < 40^\circ\text{C}$ , the values of  $\tau_1$  and  $\tau_2$  were determined from the assumption that, beginning with  $t = 40^\circ$ , the values increase with decrease in temperature, also in proportion with the viscosity. This assumption agrees rather well with reality.<sup>[1,25]</sup>

The needed parameters that have been determined in the manner outlined above are given in Table 1.

<sup>4)</sup>In this research, a spectrogram of the Rayleigh line wing was obtained in salol at room temperature with the use of a spectrograph (see Sec. 2) and the relaxation time  $\tau \sim 1.5 \times 10^{-13} \text{ sec}$  was found from the portion dD. This time agrees satisfactorily with the results of the research [28].

<sup>5)</sup>Inertia in the specified region of frequencies decreases the rate of decrease of the intensity (the portion dD); afterwards, at higher frequencies, the intensity will fall off very rapidly the portion dE). [1,25]

## D. Discussion of the Measurement Results and Comparison with Theory

For salol, all the parameters necessary for calculation are known (see Table 1). Since  $\epsilon < 1$ , one uses formulas (5) and (6) for the determination of  $\Delta$  and, consequently of  $\omega_m = \omega_T \Delta$ . Figure 4 (see also Table 2) gives the experimental results of the measurement of the dependence of  $2\Delta\nu_m$  on temperature in salol (circles) and the results of calculations from the formulas (5) and

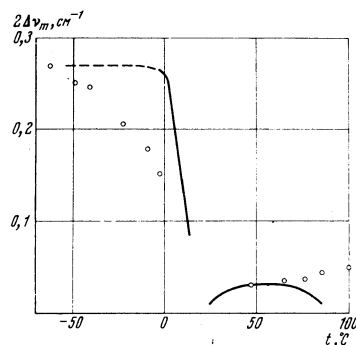


FIG. 4. Temperature dependence of  $2\Delta\nu_m$  in salol. The circles indicate experimental data. The continuous curve is theoretical (see Sec. 1). The dashed curve corresponds to the assumption of the independence of  $\omega_T$  of the temperature.

Table II. Results of measurement and calculation of the parameters of the "transverse" components in salol

$t, ^\circ\text{C}$	$2\Delta\nu_m \cdot 10^3, \text{cm}^{-1}$		$V_T, \text{m/sec}$		$2\delta\nu_m \cdot 10^3, \text{cm}^{-1}$	
	measurement	calculation	measurement	calculation	measurement	calculation
100	4.9	0	211	---	---	---
78	3.6	2.04	154	---	4	---
65	3.5	2.8	149	---	3.3	---
50	3.2**	3.1	136**	---	2.7	---
40	---	2.7	---	---	2.1	---
30	---	2.2	---	---	2.2	---
20	---	0	---	---	---	---
10	---	13.2	---	---	23	---
0	15.1***	26	630***	---	1.2	---

\*A more detailed table of measured values of  $2\Delta\nu_m$  can be found in [11] (see also Fig. 2a). For  $t = -62.5^\circ\text{C}$  in salol,  $2\Delta\nu_m = 0.269 \text{ cm}^{-1}$ .

\*\*Measurement carried out at  $t = 46.5^\circ\text{C}$ .

\*\*\*Measurement carried out at  $t = -2.5^\circ\text{C}$ .

(6) of the theory and the parameters of Table 1 (continuous curve). It is seen from Fig. 4 that for  $t < 80^\circ\text{C}$  we have qualitative agreement of the results of calculation and experiment. The temperature change of  $2\Delta\nu_m$  in the low temperature branch is slower in comparison with the calculation, which indicates that for both transverse and longitudinal sound, relaxation theory is inapplicable for large values of  $\eta$ . In this connection, the problem arises of the construction of a theory of the spectrum of scattered light on the basis of the scheme of a non-local theory.<sup>[21]</sup>

In the high temperature branch for  $t > 80^\circ\text{C}$  the reasons for the divergence between theory and experiment are not yet clear. Possibly, if we take more than two relaxation times of the anisotropy into account, the agreement between the computed and measured values of  $2\Delta\nu_m$  is improved. Some divergence can be attributed to the low accuracy of the determination of  $\tau_1$  and  $\tau_2$  (20–30%). Inasmuch as the result of the calculation depends critically on the accuracy of measurement of  $\tau_1$ , one could argue that if we were to obtain a value of  $\tau_1$ , one half the value given in Table 1, then calculation would have yielded  $\omega_m = 0$ . There is also the danger that at high temperatures, when the WFS components have a large width and small displacement, the subjective systematic error of measurement  $2\Delta\nu_m$  on the comparator gives exaggerated values. Additional more precise and objective measurements will provide the possibility of explaining this question.

Apparently the main laws governing the kinetics of transverse sound and the WFS phenomenon in liquids are qualitatively described, in to some degree also quantitatively, by the theory that takes two relaxation times into account.

4. VELOCITY AND ABSORPTION OF LONGITUDINAL HYPERSOUND IN A WIDE INTERVAL OF VARIATION OF THE VISCOSITY OF LIQUIDS

The velocity of hypersound was determined from the positions of the Mandel'shtam-Brillouin components and thermal-scattering components (formula (18)) in viscous liquids only for glycerine and triacetin<sup>[29]</sup>, a mercury lamp being used as the exciting light source.

The velocity of hypersound in 1,2 propylene glycol was determined from the spectrum of stimulated Mandel'shtam-Brillouin scattering for change in viscosity up to the vitreous state.<sup>[20,30]</sup> Investigations of the propagation of hypersound in viscous media were practically exhaustive in these researches, while significant and diverse experimental material have also been obtained for low-viscosity liquids.<sup>[1,3,7,8,25]</sup>

The measurement of the absorption coefficient from the width of the Mandel'shtam-Brillouin components (formula (19)) in viscous liquids therefore has a special value.

Table 3 and Fig. 5 show the results of our measurements of the velocity and absorption of hypersound and the dispersion of the sound velocity in salol, benzophenone and triacetin. The velocity of hypersound was measured in the change of viscosity from a small fraction of a poise to the viscosity of the vitreous state. The coefficient of absorption was measured over a somewhat smaller range of variation of the viscosity but, as is seen from Fig. 5 and Table 3, the entire region of max-

Table III. Results of the measurement of velocity and absorption coefficient of longitudinal hypersound in various materials

$t, ^\circ\text{C}$	$f \cdot 10^9, \text{Hz}$	$V, \text{m/sec}$	$V_0, \text{m/sec}$	$V_\infty, \text{m/sec}$	$\frac{V - V_0}{V_0} \cdot 10^4$	$\tau \cdot 10^{12}, \text{sec}$	$\alpha \cdot 10^{-1}, \text{cm}^{-1}$
Salol							
100	4.40	1265	1265	1620	0	1.0	
85	4.60	1312	1312	1640	0	1.4	0.78
76	4.76	1360	1360	1730	0	1.6	
65	4.80	1373	1373	1780	0	2.0	1.45
55	5.00	1421	1390	1840	2.17	2.5	
43	5.35	1510	1420	1900	6.35	4.0	
34	5.50	1570	1440	1940	9.00	6.3	1.44
22	6.10	1680	1470	2000	14.3	$1.6 \cdot 10^1$	2.02
9	6.45	1820	1500	2060	21.2	$8.0 \cdot 10^1$	1.4
-9	7.55	2120	1550	2150	36.8	$2.0 \cdot 10^2$	0.4
-13.5	7.65	2130	1560	2170		$4 \cdot 10^2$	
-34	8.3	2300	1610	2300	43.7	$2.5 \cdot 10^2$	
-40.5	8.65	2400	1630	2400	47.7	$1.2 \cdot 10^3$	
-48.5	8.85	2456	1640	2456	50.0	$1.6 \cdot 10^{11}$	
Benzophenone							
137	4.27	1360	1360	1510	0	--	0.83
115	4.65	1400	1400	1630	0	--	0.54
108	4.75	1410	1410	1670	0	1.58	
97	4.85	1420	1420	1730	0	1.62	
88	4.95	1450	1450	1780	0	1.78	
82	5.10	1455	1455	1810	0	2	0.52
67	5.40	1532	1532	1930	0	3.16	0.61
55	5.5	1530	1530	1960	0	3.5	1.35
42	5.70	1620	1560	2030	3.7	4.5	2.45
35	5.90	1650	1570	2070	4.85	5.6	2.90
19	6.45	1810	1610	2160	11	7.95	2.85
3	7.25	2080	1650	2230	23.8	20	1.45
-3	7.65	2160	1660	2280	31.0	40	1.53
-11	8.1	2258	1680	2320	34.4	124	
-22.5	8.7	2410	1700	2410	42	$1.22 \cdot 10^4$	0.11
-40	9.0	2480	1740		43.5		
-55	9.3	2570	1770		45		
Triacetin							
70	3.90	1260	1260	1430	0	1	1.05
50	4.20	1332	1332	1590	0	1.2	1.42
36	4.40	1400	1400	1700	0	2.3	1.28
22	4.80	1520	1520	1820	0	3.16	2.61
20	4.85	1530	1530	1830	0	3.54	2.23
12	5.15	1620	1550	1900	3.95	6.3	1.28
2	5.7	1785	1610	1980	10.8	15.8	1.00
-8	6.1	1915	1660	2060	15.3	50	0.39
-34	7.23	2260	1810	2230	25.0	$5 \cdot 10^3$	
-43	7.5	2320	1860	2320		$5 \cdot 10^4$	
-51	7.8	2402	1900	2402	26.5	$9.7 \cdot 10^6$	
-55	8.2	2520	1980	2520	27.3	$8.7 \cdot 10^9$	

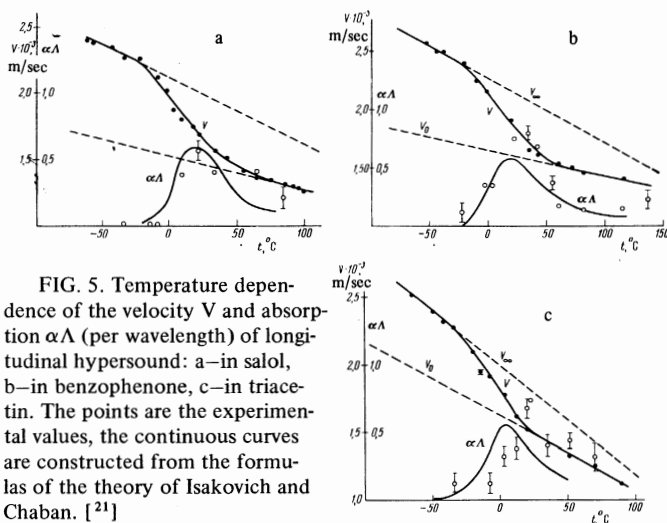


FIG. 5. Temperature dependence of the velocity V and absorption  $\alpha A$  (per wavelength) of longitudinal hypersound: a—in salol, b—in benzophenone, c—in triacetin. The points are the experimental values, the continuous curves are constructed from the formulas of the theory of Isakovich and Chaban.<sup>[21]</sup>

imum change of the absorption coefficient is sufficiently well represented.

In correspondence with the earlier researches,<sup>[19,20]</sup> the temperature coefficients for  $V_\infty$  and  $V_0$  are different (see the table), while in all cases,  $dV_\infty/dt > dV_0/dt$ . By virtue of this circumstance, the dispersion of the veloc-

ity should be greater than in <sup>[20]</sup> at low sound frequencies, in the same range of variation of the viscosity.

It was pointed out in the introduction that one should compare the results of the measurements at high viscosities with the conclusions of nonlocal theory, <sup>[21]</sup> inasmuch as relaxation theory does not describe the phenomenon. It is clear from Eq. (11) of Sec. 1 that in order to carry out the comparison of experimental data with theoretical calculations, we need to know the relaxation time  $\tau$ , in addition to the frequency (in our experiments, the frequency depends somewhat on the temperature, but it is always known).

The absorption coefficient at low sound frequency enters into Eq. (13). This coefficient was not measured for salol and benzophenone, <sup>6)</sup> so that the value of  $R$  is known. In order to determine  $R$ , we used the following method. The values of the real and imaginary parts of the function  $F(\omega\tau)$  were found (Eqs. (11) and (14)). The value of  $\omega\tau$  was determined for each temperature from the values thus found and the graph of the function  $F(\omega\tau)$ . <sup>[21]</sup> The frequency  $\omega$  was determined from the position of the MBC and known  $\tau$  were thus established. Further,  $\tau$  has been computed from Eq. (13) under the assumption that  $R = 1$ , and the value of  $R$  was determined from the ratio of the previously found value of  $\tau$  to the value of  $\tau$  for  $R = 1$ . For all the cases studied,  $R$  did not depend on temperature and was equal to 4.6 and 1 for salol and triacetin, respectively. For benzophenone at high temperatures, the value of  $R$  found by such a method was found to be equal to 4.3, but in the low temperature region, the viscosity for benzophenone is unknown and therefore  $\tau$  in this region ( $\omega\tau \gg 1$ ) was determined from the approximate formula (15).

Thus, for all materials and the interval of temperatures studied, computed values were obtained for the velocity and sound absorption and were compared with experimental results (see Fig. 5). This comparison shows that there is excellent agreement between theory and experiment over the entire range of variation of the viscosity.

There is also agreement between the measured and computed values of the absorption coefficient, but it is considerably poorer. This is partially explained by the fact that we could not take the effect of the shear modulus into consideration in the calculations, inasmuch as the temperature dependence of the limiting shear modulus  $\mu_0$  is not known. At the same time, such consideration significantly improves the agreement of the experimental and theoretical values of  $\alpha$ . <sup>[20,21]</sup> We also note that these first determinations of the absorption coefficient were made less satisfactory by appreciable error. Therefore, if the accuracy is to be considerably improved, which is quite realistic, and if the effect of the shear modulus is to be taken into account in the calculation, then one can expect better agreement of theory with experiment. But even after such improvements, it

is possible that the location of the maximum calculated and found experimentally ( $\alpha\Lambda$  as a function of  $t$ ) will not agree with each other. Some results obtained in <sup>[20]</sup> also suggest this. Therefore, the problems of the development and perfection of the theory of Isakovich and Chaban, <sup>[21]</sup> and also the problem of the direct testing of the assumptions on which the theory is constructed, all remain urgent.

In conclusion, the authors express their gratitude to V. P. Zaĭtsev and S. V. Krivokhizha for assistance in the research.

<sup>1</sup>I. L. Fabelinskiĭ, *Molekulyanoe rasseyanie sveta* (Molecular Scattering of Light) (Nauka, 1964) [Plenum Press, 1968].

<sup>2</sup>P. F. Flubacher, A. L. Leodbetter, I. A. Harrison, and B. P. Stoicheff, *J. Phys. Chem. Solids* **12**, 53 (1960).

<sup>3</sup>D. I. Mash, V. V. Morozov, V. S. Starunov, E. V. Tiganov and I. L. Fabelinskiĭ, *ZhETF Pis. Rev.* **2**, 246 (1965) [*JETP Lett.* **2**, 157 (1965)].

<sup>4</sup>T. S. Velichkina, O. A. Shustin, and I. A. Yakovlev, *ZhETF Pis. Red.* **2**, 189 (1965) [*JETP Lett.* **2**, 119 (1965)].

<sup>5</sup>S. M. Shapiro, R. W. Gammon, and H. Z. Cummins, *Appl. Phys. Lett.* **9**, 157 (1966).

<sup>6</sup>V. S. Starunov, E. Tiganov, and I. L. Fabelinskiĭ, *ZhETF Pis. Red.* **5**, 317 (1967) [*JETP Lett.* **5**, 260 (1967)].

<sup>7</sup>I. L. Fabelinskiĭ and V. S. Starunov, *Appl. Optics* **6**, 1793 (1967).

<sup>8</sup>E. V. Tiganov, *Propagation of Longitudinal and Transverse Hypersound in Liquids* (Candidate's dissertation), *Phys. Inst., Acad. Sci., Moscow*, 1967.

<sup>9</sup>G. I. Stigeman and B. Stoicheff, *Phys. Rev. Lett.* **21**, 202 (1968).

<sup>10</sup>L. M. Sabirov, V. S. Starunov, and I. L. Fabelinskiĭ, *ZhETF Pis. Red.* **8**, 399 (1968) [*JETP Lett.* **8**, 246 (1968)].

<sup>11</sup>I. L. Fabelinskiĭ, L. M. Sabirov, and V. S. Starunov, *Phys. Lett.* **29A**, 414 (1969).

<sup>12</sup>V. Volterra, *Phys. Rev.* **180**, 156 (1969).

<sup>13</sup>E. F. Gross, V. P. Romanov, A. V. Solov'ev, and E. O. Chernysheva, *Fiz. Tverd. Tela* **11**, 3686 (1969) [*Soviet Phys.-Solid State* **11**, 3101 (1970)].

<sup>14</sup>G. I. Stigeman, *Light Scattering from Thermal Shear Waves in Liquids*, Ph.D. Thesis, U. Toronto, 1969.

<sup>15</sup>N. B. Rozhdestvenskaya and L. A. Zubkov, *Optika i spektroskopiya* (Optics and Spectroscopy) **28**, 599 (1970).

<sup>16</sup>L. M. Sabirov, *Light Scattering Spectra and the Propagation of Transverse and Longitudinal Hypersound in Viscous Liquids and Glasses* (Candidate's thesis), *Phys. Inst., Acad. Sci. Moscow*, 1970.

<sup>17</sup>M. Leontovich, *J. Phys.* **4**, 499 (1941).

<sup>18</sup>L. I. Mandel'shtam and M. A. Leontovich, *Zh. Eksp. Teor. Fiz.* **7**, 439 (1937).

<sup>19</sup>S. V. Krivokhizha and I. L. Fabelinskiĭ, *Zh. Eksp. Teor. Fiz.* **50**, 3 (1966) [*Soviet Phys.-JETP* **23**, 1 (1966)].

<sup>20</sup>S. V. Krivokhizha, *Investigation of the Propagation of Ultrasound and Hypersound in Viscous Liquids, Glasses, and Crystalline Quartz Over a Wide Range of Temperatures* (Candidate's dissertation), *Phys. Inst. Acad. Sci., Moscow*, 1969.

<sup>6)</sup>Salol and benzophenone are comparatively easily supercooled and can remain many years in the supercooled state, but they are crystallized by a sufficiently strong mechanical action (shaking) and especially by radiation by ultrasound. For this reason, the application of an optical method of investigation of the acoustic properties of such a class of materials is particularly appropriate.

- <sup>21</sup>M. A. Isakovich and I. A. Chaban, Zh. Eksp. Teor. Fiz. 50, 1343 (1966) [Soviet Phys.-JETP 23, 893 (1966)].
- <sup>22</sup>S. M. Rytov, Zh. Eksp. Teor. Fiz. 58, 2154 (1970) [Soviet Phys.-JETP 31, 1163 (1970)].
- <sup>23</sup>S. M. Rytov, Zh. Eksp. Teor. Fiz. 59, 2130 (1970) [Soviet Phys.-JETP 32, No. 12 (1971)].
- <sup>24</sup>V. P. Zaitsev and L. M. Sabirov, PTÉ (in press).
- <sup>25</sup>V. S. Starunov, Trudy (Works) phys. Inst. Acad. Sci. 39, 151 (1967).
- <sup>26</sup>D. A. Jackson and E. R. Pike, J. Phys. E1, 394 (1968).
- <sup>27</sup>I. L. Fabelinskiĭ, Izv. Akad. Nauk SSSR, ser. fiz. 9, 186 (1945).
- <sup>28</sup>I. L. Fabelinskiĭ, Izv. Akad. Nauk SSSR, ser. fiz. 11, 382 (1947).
- <sup>29</sup>M. S. Pesin, I. L. Fabelinskiĭ, Dokl. Akad. Nauk SSSR 129, 299 (1959); 135, 114 (1960) [Soviet Phys.-Doklady 4, 1264 (1960)].
- <sup>30</sup>I. Sh. Zlatin, S. V. Krivokhizha, and I. L. Fabelinskiĭ, Zh. Eksp. Teor. Fiz. 56, 1186 (1969) [Soviet Phys.-JETP 29, 683 (1969)].

Translated by R. T. Beyer

20