SHEAR DOUBLET IN RAYLEIGH SCATTERING OF LIGHT IN LIQUIDS WITH TWO RELAXATION TIMES

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The narrow doublet observed in depolarized components of light scattered by certain liquids[1-3] is considered, and it is assumed that the singularities in the temperature dependence of the interval between the maxima of this doublet, $2\Delta\nu_{tr}$, are due to the presence of two relaxation times in the liquid.[6] Application of the usual relaxation theory of Rayleigh scattering[9] to this case makes it possible to ascertain the conditions for the existence of a shear doublet (Secs. 2 and 3) and to demonstrate that in the case when the relaxation times differ strongly (Sec. 4) it is possible to obtain satisfactory quantitative description of both the temperature (Sec. 5) and the angular (Sec. 6) dependences of $2\Delta\nu_{tr}$.

1. FORMULATION OF PROBLEM

THE presence of a narrow doublet in the spectrum of the depolarized components $J_x^z = J_z^y$ [1)] in the scattering of light in liquids was first established by Starunov, Tiganov, and Fabelenskiĭ.[1-3] On the basis of polarization investigations and of the study of the angular dependence of the displacement $\Delta\nu_{tr}$ of the doublet maxima observed in nitrobenzene, quinoline, and aniline, the authors have reached the conclusion that this doublet is due to a shear wave satisfying the Bragg condition and to anisotropy fluctuations associated with this wave. Subsequent experiments by these authors, and also the results of Stegeman and Stoicheff[4] pertaining to three more liquids in addition to those mentioned above, have shown that the temperature dependence of $\Delta\nu_{tr}$ is the opposite of that expected from the Leontovich relaxation theory.[5]

Further experiments by Fabelinskiĭ et al.[6] with salol in a wide temperature interval (from +120 to −48°C) revealed the presence in this liquid of two branches of the temperature curve of $\Delta\nu_{tr}$—one decreasing and one rising with increasing temperature (Fig. 1). In the interval from +46 to −2.5°C, no fine structure of the wing (henceforth called the shear doublet) is observed between the two branches.[2)] An explanation of this phenomenon was proposed already in [6], where it was shown qualitatively that the presence of two relaxation times makes it possible to describe satisfactorily the appearance of both branches of the temperature curve of $\Delta\nu_{tr}$.

At approximately the same time, Volterra[7] developed a relaxation theory of scattering in the case of two relaxation times and offered, independently of [6], an explanation (likewise qualitative) of the increasing temperature branch of $\Delta\nu_{tr}$ in quinoline. In a paper by

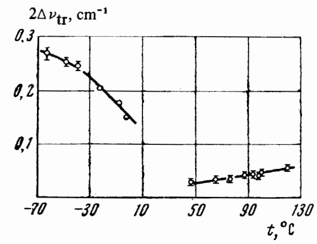

FIG. 1

V. P. Romanov and V. A. Solov'ev delivered at the 9th Conference on the Physics of Liquids (Kiev, 1969), independently of [6,7], an explanation was also proposed for the shear doublet. This explanation was subsequently reported in [8] where, in addition, the results were confirmed for quinoline and data were given for benzylalcohol.

We present in this article a quantitative analysis of the conditions for the existence of a shear doublet in liquids having two anisotropy relaxation times. The analysis is based on the results of the general relaxation theory in [9] for Rayleigh scattering.

2. SPECTRAL INTENSITY OF DEPOLARIZED LIGHT

The indicated theory gives for the spectral intensities of the depolarized components of scattered light the formula

$$J_h^z(\omega, \mathbf{q}) = J_z^y(\omega, \mathbf{q}) = -\frac{k_B T}{2\pi i\omega}\left[\frac{q_2^2 X^2}{4(\mu q^2 - \rho\omega^2)} + \frac{1}{2}\sum_k \frac{n_k^2}{1 + i\omega\tau_k} - \text{c.c.}\right]. \quad (1)$$

Here ω is the frequency reckoned from the frequency of the primary wave, $\mathbf{q}$ is the scattering vector ($q_2^2 = k^2 \times \sin^2\theta$, $q^2 = 4k^2\sin^2(\theta/2)$, θ is the scattering angle, and k is the wave number of the primary wave), μ is the complex shear modulus, X is the complex mechano-optical coefficient, ρ and T are the density and temperature of the medium, and c.c. denotes the complex conjugate. For liquids in the case of two anisotropy relaxation times τ_1 and τ_2, the values of μ and X are given by [9]

$$\mu = \frac{i\omega}{2}\left(\frac{N_1^2\tau_1}{1 + i\omega\tau_1} + \frac{N_2^2\tau_2}{1 + i\omega\tau_2}\right), \quad X = i\omega\left(\frac{n_1 N_1\tau_1}{1 + i\omega\tau_1} + \frac{n_2 N_2\tau_2}{1 + i\omega\tau_2}\right), \quad (2)$$

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1) The superior indices denote the polarization of the primary wave propagating along the x axis, and the inferior ones the observed polarization of the wave scattered in the direction of the y axis. In formula (1) below, the inferior index h denotes horizontal polarization.

2) I. L. Fabelinskiĭ was kind enough to inform me that similar results were also obtained for benzophenone.

where $n_{1,2}$ and $N_{1,2}$ are real constants, As $\omega \to 0$, we have, accurate to the first order in $i\omega$

$$\mu = \frac{i\omega}{2}(N_1^2\tau_1 + N_2^2\tau_2) = i\omega\eta_0, \quad X = i\omega(n_1N_1\tau_1 + n_2N_2\tau_2) = i\omega\cdot 2\varepsilon_0 M\eta_0,$$

where η_0 is the static viscosity, ϵ_0 the optical dielectric constant of the liquid, and M its Maxwell's constant. As $\omega \to \infty$ we have

$$\mu = {}^1/_2(N_1^2 + N_2^2) = \mu_\infty, \quad X = n_1N_1 + n_2N_2 = X_\infty.$$

Using the last four relations to express $N_{1,2}^2$ and $n_{1,2}N_{1,2}$ in terms of η_0, M, μ_∞, and X_∞, we can easily reduce (2) to the form

$$\mu = \frac{i\omega(\eta_0 + i\omega\mu_\infty\tau_1\tau_2)}{(1+i\omega\tau_1)(1+i\omega\tau_2)}, \quad X = \frac{i\omega(2\varepsilon_0 M\eta_0 + i\omega X_\infty\tau_1\tau_2)}{(1+i\omega\tau_1)(1+i\omega\tau_2)}, \quad (3)$$

and from the positiveness of $N_{1,2}^2$, assuming $\tau_1 \geq \tau_2$, it follows that

$$\mu_\infty\tau_2 \leqslant \eta_0 \leqslant \mu_\infty\tau_1. \quad (4)$$

Substituting (3) in (1), we obtain

$$J_h^z = J_z^y = -\frac{k_BT}{2\pi i\omega}\left\{\frac{q_2^2}{4\rho}\cdot\right.$$
$$\times\left[\frac{i\omega(2\varepsilon_0M\eta_0 + i\omega X_\infty\tau_1\tau_2)^2}{(1+i\omega\tau_1)(1+i\omega\tau_2)[\omega_\eta + i\omega\omega_T^2\tau_1\tau_2 + i\omega(1+i\omega\tau_1)(1+i\omega\tau_2)]} - \text{c.c.}\right]$$
$$\left. - i\omega\left(\frac{n_1^2\tau_1}{1+\omega^2\tau_1^2} + \frac{n_2^2\tau_2}{1+\omega^2\tau_2^2}\right)\right\}, \quad (5)$$

where

$$\omega_\eta = \frac{\eta_0 q^2}{\rho}, \quad \omega_T^2 = \frac{\mu_\infty q^2}{\rho}. \quad (6)$$

In the case of one relaxation time, i.e., when $\tau_1 = \tau_2 = \tau$, and, as can be readily seen,

$$\mu_\infty\tau = \eta_0, \quad X_\infty\tau = 2\varepsilon_0M\eta_0, \quad n^2 = \frac{X_\infty^2}{4\mu_\infty},$$

formula (5) yields

$$J_h^z = J_z^y = \frac{k_BTX_\infty^2\tau}{4\pi\mu_\infty}\left\{\frac{q_2^2\omega_T^2\tau}{2q^2}\cdot\right.$$
$$\left.\times\left[\frac{1}{(1+i\omega\tau)[(\omega^2-\omega_T^2)\tau - i\omega]} - \text{c.c.}\right] + \frac{1}{1+\omega^2\tau^2}\right\}. \quad (7)$$

In turn, in observations at a right angle ($\theta = 90°$, $q_2^2 = q^2/2$), Eq. (7) goes over into Leontovich's well-known formula.[5, 10]

3. CONDITIONS FOR THE EXISTENCE OF THE SHEAR DOUBLET

A thorough knowledge of the extent to which the doublet becomes pronounced against the relaxation background requires, of course, an analysis of the frequency dependence of the complete expressions (5) and (7). For definite values of the parameters, the corresponding curves can be calculated with a computer, but to ascertain the conditions under which the doublet is in general possible it suffices to investigate the roots of the cubic polynomial in $i\omega$ contained in the denominator of (5) and of the quadratic polynomial in the denominator (7). Factors of the type $1 + i\omega\tau$ in the denominators can always be separated by expanding in partial fractions, and contribute only to the relaxation background, i.e., they produce in the intensity terms of the form $(1 + \omega^2\tau^2)^{-1}$, which have a maximum at $\omega = 0$. Obviously, the doublet is possible when the indicated polynomials have a pair of complex conjugate roots

$$i\omega = \omega_T(-\zeta \pm i\Delta), \quad (8)$$

and constitute in this case two Lorentz lines with half-width ζ, shifted by intervals $\pm\Delta$ from $\omega = 0$ (in units of ω_T).

In the case of one relaxation time, the quadratic trinomial in (7) has roots in the form (8) when $2\omega_T\tau > 1$, with

$$\Delta = \sqrt{1 - \frac{\gamma}{4}}, \quad \zeta = \frac{\sqrt{\gamma}}{2} \quad \left(\gamma = \frac{1}{\omega_T^2\tau^2}\right).$$

Although the doublet exists when $\gamma < 4$, it is clear that, even if we disregard the relaxation background, one can count on observing the doublet only in the region $\gamma < 1$ ($\omega_T\tau > 1$), where the half-width ζ is much smaller than the shift Δ. With increasing temperature, the relaxation time τ decreases (γ increases), and, consequently, in the case of one relaxation time the doublet is possible only in the region of not too high temperatures.

One root of the polynomial cubic in $i\omega$ in (5) is always real (it contributes only to the relaxation background), and the condition for the existence of two other roots of the type (8), as well as the expressions for the roots themselves, can be obtained in accordance with the Cardan formulas.

We introduce the variable $z = i\omega/\omega_T$ and the parameters

$$\varepsilon = \frac{\tau_2}{\tau_1}, \quad \beta = \frac{1}{2}\left(\frac{\eta_0}{\mu_\infty\tau_1} - \frac{1+\varepsilon}{3}\right), \quad \gamma = \frac{1}{\omega_T^2\tau_2^2}. \quad (9)$$

According to (4)

$$\varepsilon \leqslant \eta_0/\mu_\infty\tau_1 \leqslant 1$$

and accordingly, by virtue of (9)

$$-{}^1/_6(1 - 2\varepsilon) \leqslant \beta \leqslant {}^1/_3(1 - \varepsilon/2). \quad (10)$$

When ϵ changes from zero ($\tau_2 \ll \tau_1$) to unity ($\tau_2 = \tau_1$), the interval of permissible values of β narrows down from the segment $(-1/6, 1/3)$ at $\epsilon = 0$ to a fixed value $\beta = 1/6$ at $\epsilon = 1$.

By equating the cubic polynomial in (5) to zero, we obtain in the notation of (9) the equation

$$z^3 + \sqrt{\gamma}(1+\varepsilon)z^2 + (1+\varepsilon\gamma)z + \sqrt{\gamma}\left(2\beta + \frac{1+\varepsilon}{3}\right) = 0. \quad (11)$$

Cardan's formulas give for its complex-conjugate roots in the form (8)

$$\Delta = \frac{\sqrt{3}}{2}(u - v), \quad \zeta = \frac{u+v}{2} + \sqrt{\gamma}\frac{1+\varepsilon}{3}, \quad (12)$$

where

$$\left.\begin{matrix}u\\v\end{matrix}\right\} = \pm(\sqrt{q^2+p^3} \mp q)^{1/3}, \quad q = \sqrt{\gamma}\left[\beta + \frac{\gamma}{27}(1+\varepsilon)(1-2\varepsilon)\left(1 - \frac{\varepsilon}{2}\right)\right]$$

$$q^2 + p^3 = \beta^2\gamma + \frac{2}{27}\beta\gamma^2(1+\varepsilon)(1-2\varepsilon)\left(1 - \frac{\varepsilon}{2}\right) \quad (13)$$
$$+ \frac{1}{27}\left[1 - \gamma(1 - \varepsilon + \varepsilon^2) + \frac{1}{3}\gamma^2(1 - \varepsilon + \varepsilon^2)^2 - \frac{1}{4}\gamma^3\varepsilon^2(1-\varepsilon)^2\right].$$

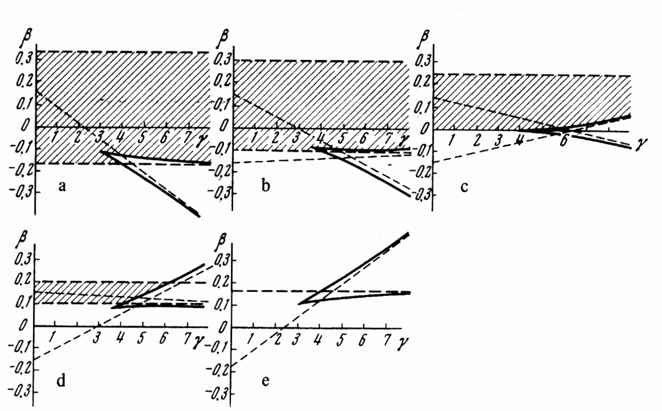

FIG. 2. The (β, γ) plane in the case $\epsilon = 0$ (a), $\epsilon = 0.2$ (b), $\epsilon = 0.5$ (c), $\epsilon = 0.8$ (d), and $\epsilon = 1$ (e).

The condition for the existence of such roots (the condition that u and v be real) is

$$q^2 + p^3 > 0. \tag{14}$$

Formulas (12) and (13) determine Δ and ζ as functions of ϵ, β and γ. Figures 2a–e show, shaded, the region of the plane (β, γ) where the doublet exists, for five values of ϵ. The condition (10) determines the permissible band of β; in addition, a curvilinear wedge penetrates into this band, and the condition (14) is violated within this wedge. The wedge crosses the upper boundary of the shaded band at the point $\gamma = 4/\epsilon^2$, and the lower boundary at $\gamma = 4$. At equal relaxation times, the permissible region reduces to a segment from $\gamma = 0$ to $\gamma = 4$ on the straight line $\beta = 1/6$ (Fig. 2e).

4. CASE OF STRONGLY DIFFERING RELAXATION TIMES

Let $\tau_1 \gg \tau_2$, i.e., let the parameter ϵ be small enough to be negligible compared with unity (Fig. 2a). The permissible values of β lie in the interval $(-1/6, 1/3)$, and the top of the forbidden wedge is at the point $\beta = -1/9$, $\gamma = 3$. At $\epsilon = 0$, expressions (13) become much simpler, namely

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

Plots of Δ and ζ as functions of γ are shown for a number of values of β in Figs. 3 and 4, respectively. So long as $\beta > -1/9$ the shift Δ does not vanish for any value of γ. The straight line $\beta = -1/9$ already passes through the top of the forbidden wedge (Fig. 2a), and accordingly, for this value of β the shift Δ vanishes when $\gamma = 3$. The straight lines $\beta = \text{const} < 1/9$ intersect the wedge, and correspondingly there appears in Fig. 3 an interval of γ in which there is no doublet (all three roots of Eq. (11) are real). The ends of this interval are the points

$$\gamma_{1,2} = \frac{3}{2(6\beta+1)}\left[1-27\beta^2 \pm 27\left(\beta+\frac{1}{9}\right)\sqrt{\left(\beta+\frac{1}{9}\right)\left(\beta-\frac{1}{3}\right)}\right].$$

At these points $\Delta = 0$ ($u = v = -q^{1/3}$), and the half width ζ of the doublet lines is

$$\zeta_{1,2} = \frac{1}{3}\left\{\gamma_{1,2}^{1/2} - \left[\gamma_{1,2}^{1/2}(\gamma_{1,2}+27\beta)\right]^{1/3}\right\}.$$

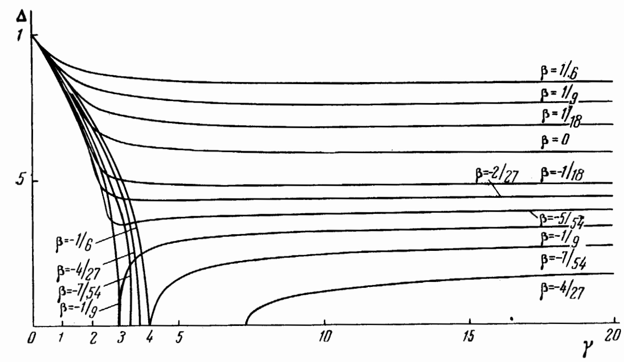

FIG. 3

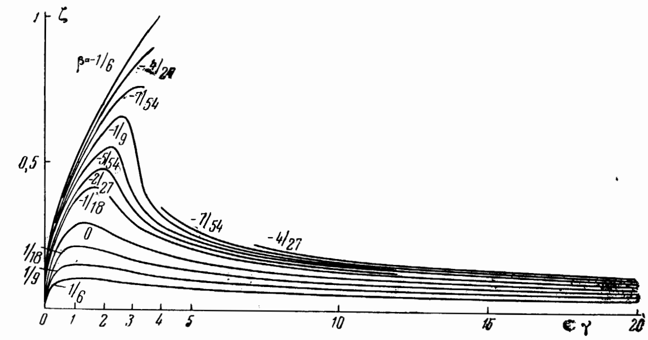

FIG. 4

It is seen from Fig. 3, furthermore, that when $\beta \lesssim -2/27$ the $\Delta(\gamma)$ curves acquire sections on which Δ increases with increasing γ. Starting with $\beta = -1/9$, this section forms a separate branch, which continuously drops and moves farther away to the right along the γ axis with further decrease of β. The limiting value $\beta = -1/6$ returns us to the case of one relaxation time, when there remains only one $\Delta(\gamma)$ branch that decreases from unity to zero as γ increases from zero to four.

We note that when $\epsilon \neq 0.2$ (Fig. 2b), the rising branch of $\Delta(\gamma)$ practically vanishes. It can thus exist only at a sufficiently small ratio of the relaxation times $\epsilon = \tau_2/\tau_1$.

5. TEMPERATURE DEPENDENCE OF Δ IN THE CASE WHEN $\epsilon = 0$

It is easy to note that there is a complete qualitative correspondence between those curves of Fig. 3 which have two branches (decreasing and increasing with increasing γ) and the experimental $\Delta\nu_{tr}(T)$ plot (see Fig. 1). By choosing a certain monotonic temperature dependence of the parameters β and γ (which does not violate the condition $\epsilon \ll 1$, under which the curves of Fig. 3 have been plotted), it is also possible to obtain fair quantitative agreement.[3] By way of an example, Fig. 5 shows the $\Delta(\beta, \gamma)$ curve taken from Fig. 3, corresponding to $\beta = -4/27$ and transformed into a function of the temperature T from a function of γ by means of the exponential dependence of γ on T, namely

[3]Of course, it would be better justified and simpler to compare the theoretical curves with direct experimental data on the temperature dependence of the parameters of the liquid (μ_∞, η_0, ρ, τ_1, τ_2), but the necessary information either does not cover the entire temperature interval of Fig. 1, or is generally nonexistent.

$$\lg \gamma = -2.3079 + 0.010034T. \qquad (15)$$

The same Fig. 5 shows the experimental points for salol (Fig. 1), the ordinate scale being chosen such that the experimental point $\Delta\nu_{tr} = 0.075\ \text{cm}^{-1}$ ($T = 270.5\,°K$) falls on the curve recalculated in this manner. However, one must not attach too great a value to the resultant fairly good agreement between theory and experiment. First, the theoretical curves do not take into account the relaxation background, which can decrease the values of Δ by 15–20%, and whose influence on the experimental $\Delta\nu_{tr}$ cannot be excluded.[4)] Second, at low temperatures (high viscosities) the applicability of the relaxation theory is in general subject to question.

Let us nevertheless ascertain what can be said concerning the temperature dependence of the parameters of salol, by starting from the relation (15).

The temperatures 200 and 400 °K correspond in accordance with (15) to values $\gamma \simeq 0.47$ and 50. Accordingly, the product $\Delta = 0$ ($u = v = -q^{1/2}$), decreases by one order of magnitude on going from the left edge of the plot in Fig. 5 to the right one. At very low temperatures, when $\Delta \to 1$, the doublet lines are separated by an interval $2\,\omega_T$. Judging from the experimental curve of Fig. 1, this limiting value of Δ corresponds to $2\Delta\nu_{tr} \simeq 0.3\ \text{cm}^{-1}$, i.e., $\omega_t \simeq 2.83\times 10^{10}$ rad/sec. Consequently, at 200 °K we have

$$\tau_2 = 1/\omega_T\sqrt{\gamma} \cong 5.15\cdot 10^{-11}\,\text{sec}.$$

The lack of data on the temperature dependence of μ_∞ prevents us from estimating ω_T and τ_2 separately for other temperatures.

If we assume arbitrarily that ω_T retains the value given above in the entire temperature interval of interest to us, then we obtain at 273 and 400 °K, respectively, $\tau_2 = 2.15\times 10^{-11}$ and 5×10^{-12} sec.

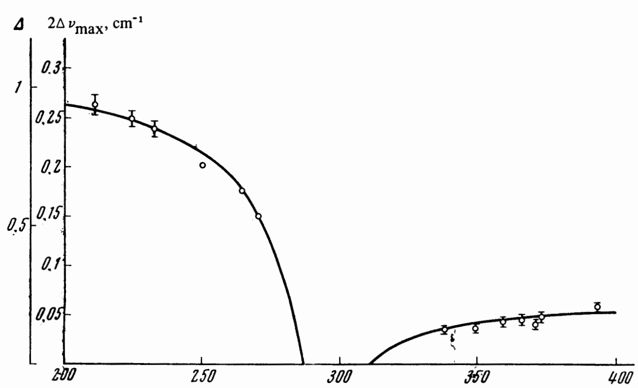

FIG. 5. Dependence of Δ on T at $\beta = -4/27$.

The temperature 200 °K corresponds to the following (extrapolated) values of the density and of the refractive index: $\rho = 1.59$ and $n = 1.637$. For the wavelength $\lambda = 6328$ Å we obtain

$$q^2 = 8\pi^2 n^2/\lambda^2 = 5.3\cdot 10^{10}\,\text{cm}^{-2},$$

which yields at $\omega_T = 2.83\times 10^{10}$

$$= \frac{\rho\omega_T^2}{q^2} = 2.4\cdot 10^{10}\,\text{dyn-cm}^{-2},\quad V_\infty = \sqrt{\frac{\mu_\infty}{\rho}} = \frac{\omega_T}{q} = 1230\,\text{m-sec}^{-1}.$$

One could use the ratio

$$\mu_\infty\tau_1/\eta_0 = 27,$$

which follows from (9) when $\epsilon \ll 1$ and $\beta = -4/27$. However, there is hardly any advantage to carrying out further quantitative estimates in view of the patent lack of actual data on the temperature dependence of various parameters of salol.

6. ANGULAR DEPENDENCE OF THE INTERVAL BETWEEN THE DOUBLET LINES

The scattering angle θ enters the half-interval Δ between the doublet lines only via the square of the modulus of the scattering vector $q^2 = 4k^2\sin^2(\theta/2)$, i.e., via

$$\omega_T^2 = \frac{\mu_\infty q^2}{\rho} = 2\omega_{90}^2\sin^2\frac{\theta}{2},$$

where $\omega_{90} = \sqrt{2k^2\mu_\infty/\rho}$ is the value of ω_T at $\theta = 90°$. The parameters ϵ and β do not depend on ω_T, and for γ we have

$$\gamma = \frac{1}{\omega_T^2\tau_2^2} = \frac{\gamma_{90}}{2\sin^2(\theta/2)},\quad \gamma_{90} = \frac{1}{\omega_{90}^2\tau_2^2}.$$

Variation of θ to 180° corresponds to a change of γ from ∞ to $\gamma_{90}/2$.

According to (8), the dimensional half-interval between the doublet lines is $\Delta\omega_{tr} = \omega_T\Delta$, where Δ is determined by formulas (12) and (13). In order to use the already calculated curves of $\Delta = \Delta(\beta, \gamma)$ to plot $\Delta\omega_{tr}$ against θ (we confine ourselves, as before, to the case $\epsilon \ll 1$), it is advisable to retain the parameter γ as the argument:

$$\Delta\omega_{tr} = \omega_T\Delta(\beta,\gamma) = \omega_{90}\sqrt{\gamma_{90}/\gamma}\,\Delta(\beta,\gamma).$$

It would be natural to refer $\Delta\omega_{tr}$ to the value of this half-interval at $\theta = 90°$:

$$\Delta\omega_{90} = \omega_{90}\Delta(\beta,\gamma_{90}), \qquad (16)$$

i.e., to plot the following curves as functions of θ (or of $\sin(\theta/2)$):

$$\frac{\Delta\omega_{tr}}{\Delta\omega_{90}} = \sqrt{\frac{\gamma_{90}}{\gamma}}\frac{\Delta(\beta,\gamma)}{\Delta(\beta,\gamma_{90})},\quad \sin\frac{\theta}{2} = \sqrt{\frac{\gamma_{90}}{2\gamma}}. \qquad (17)$$

By specifying the value of β, i.e., by choosing any one of the curves of Fig. 3, and then fixing the value of γ_{90} (and by the same token, in accord with (16), also the ratio $\Delta\omega_{90}/\omega_{90}$), it is easy to plot $\Delta\omega_{tr}/\Delta\omega_{90}$ against $\sin(\theta/2)$ by using the chosen curve of Fig. 3 and formulas (17).

Figure 6 shows three such plots corresponding to $\beta = -4/27$. Two of them ($\gamma_{90} = 20$ and $\gamma_{90} = 12$) pertain to the case when the width of the doublet is determined, for observation at an angle $\theta = 90°$, by the rising branch

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4) The third root z_1 of (11) is always real (and negative). Therefore the expansion of the cubic polynomial (11) contains the factor

$$z - z_1 = \frac{i\omega}{\omega_T} - z_1 = -z_1(1 + i\omega\tau'),$$

where $\tau' = -1/\omega_T z_1$ plays the role of a certain additional relaxation time, which, generally speaking, differs from τ_1 and τ_2 and depends on the parameters ϵ, β and γ. Thus, the spectrum of the relaxation background does not reduce in the general case to a superposition of two wings of the form $(1+\omega^2\tau_1^2)^{-1}$ and $(1+\omega^2\tau_2^2)^{-1}$, and its form requires a more detailed analysis.

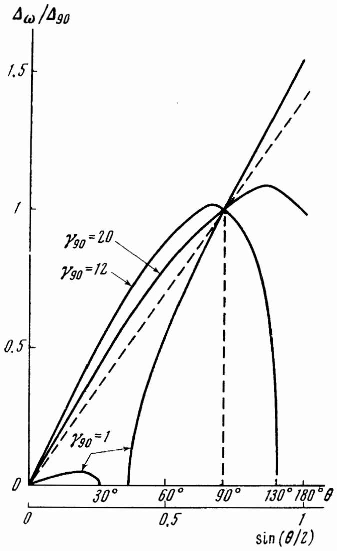

FIG. 6. Dependence of $\Delta_\omega/\Delta_{90}$ on θ at $\beta = -4/27$ for three values of γ_{90}.

of the $\Delta(\gamma)$ curve, while the third ($\gamma_{90} = 1$) pertains to the case when this width is determined by the decreasing branch.

The larger γ_{90}, the closer $\Delta(\beta, \gamma)$ is to its asymptotic value $\Delta(\beta, \infty)$ at all values of the scattering angle $0 < \theta < 180°$ ($\infty > \gamma > \gamma_{90}/2$). For example, if $\gamma_{90} = 30$, so that γ changes from 15 to ∞, then we find for $\beta = -4/27$ (Fig. 3) that $\Delta(\beta, \gamma)$ changes only from 0.15 to the asymptotic value 0.193. In the first-order approximation we can assume, at sufficiently large γ_{90}, that $\Delta(\beta, \gamma) = \Delta(\beta, \gamma_{90})$ in the entire interval of γ from $\gamma_{90}/2$ to ∞. According to (17), this means that for all values of θ we have

$$\frac{\Delta\omega_{tr}}{\omega_{90}} = \sqrt{\frac{\gamma_{90}}{\gamma}} = \sqrt{2}\sin\frac{\theta}{2},$$

i.e., the dependence of $\Delta\omega_{tr}$ on $\sin(\theta/2)$ becomes linear.

Thus, direct proportionality of $\Delta\omega_{tr}$ to $\sin(\theta/2)$ is not at all obligatory. If the width of the doublet is determined by the rising branch of the $\Delta(\gamma)$ curve, then the plots of $\Delta\omega_{tr}$ against $\sin(\theta/2)$ can have a maximum (see the curves for $\gamma_{90} = 20$ and 12 on Fig. 6), as was obtained by Stegeman by direct measurements for a number of liquids.[11]

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243