SOVIET PHYSICS JETP VOLUME 22, NUMBER 6 JUNE, 1966

INTENSITY AND WIDTH OF THE FINE STRUCTURE COMPONENTS OF SCATTERED LIGHT IN LIQUIDS AND THE ATTENUATION OF HYPERSOUND

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Submitted to JETP editor July 20, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) **49**, 1764-1773 (December, 1965)

The fine structure of the scattered light excited by $\lambda = 6328$ Å radiation from a He-Ne gas laser is investigated in five liquids. The hypersonic velocity is determined at a frequency $f \sim 4 \times 10^9$ cps on the basis of the position of the Mandel'shtam-Brillouin components. The hypersonic absorption coefficient is determined on the basis of the width of these lines. It is shown that for some of the liquids the experimental results can be described quantitatively by relaxation theory formulas involving only a single relaxation time. The relaxation times for the bulk viscosity coefficient are determined. The relative intensities of the fine structure components are measured and compared with the theoretical values.

INTRODUCTION

THE study of the spectral component of Rayleigh light scattering in liquids and solids allows us to obtain extensive information on the kinetics of the adiabatic and isobaric density fluctuations and anisotropy fluctuations.[1-3] The present research is devoted to the study of the spectrum of the adiabatic density fluctuations in a liquid in the excitation of light scattering of the narrow and intense spectral line $\lambda = 6328$ Å of stimulated emission of an He-Ne gas laser.

The spectrum of adiabatic density fluctuations consists of the discrete Stokes and anti-Stokes Mandel'shtam-Brillouin components (MBC), the separation of the maxima of which is determined by the ratio of the hypersonic velocity to the light velocity, while the width of the components is determined by the damping of the corresponding hypersonic waves. From a study of the fine structure upon excitation of scattered light by lines of the mercury spectrum, it has been possible to find the dispersion of the sound velocity in some liquids,[1, 2, 4-6] although the accuracy of these measurements was low. The attenuation of the hypersound in previous measurements could not generally be found because of too great a width of the lines of the excited light.[1, 2]

The first application of the radiation $\lambda = 6328$ Å of an He-Ne gas laser to the study of the fine structure made it possible to increase the accuracy of determination of the hypersonic velocity by an order of magnitude and, most important, made it possible to find the attenuation of the hypersound (frequency $\sim 5 \times 10^9$ cps) over the range of MBC in liquids.[7] The subsequent investigations have widened the range of studied objects.[8, 9] The application of the narrow excited line further made it possible to find, with accuracy higher than before, the ratio of the intensities of the central component of the fine structure to the intensity of the MBC and thus verify the Landau-Placzek relation, which has been done in this research for several liquids.

FUNDAMENTAL THEORETICAL RELATIONS

Light scattering by adiabatic density fluctuations must be regarded as the diffraction of light by thermal elastic Debye waves in a condensed medium. The theory of Mandel'shtam[10] and Brillouin[11] which is based on this idea, leads to the conclusion that the light scattered by the adiabatic density fluctuations will be modulated by the frequency f of the thermal Debye wave, where

$$f = \Delta\nu = 2n\nu\frac{v}{c}\sin\frac{\vartheta}{2}. \qquad (1)$$

Here $\Delta\nu$ is the frequency difference between the maxima of the displaced fine structure lines, brought about by the isobaric density fluctuations and by the displaced MBC; n, ν, v, c, ϑ are respectively the index of refraction of the medium, the frequency of the excited light, the velocity of sound, the velocity of light, and the scattering angle. The spectroscopic determination of $\Delta\nu$ makes it possible to find from Eq. (1) the frequency of the elastic thermal wave $f \equiv \Delta\nu$ and its phase velocity of propagation v.

An elastic wave exhibits considerable attenuation in propagation in a liquid, especially at high frequencies. This leads to a broadening of the MBC by an amount

$$\delta\nu_{\mathrm{MB}} = \alpha v / \pi c \quad [\mathrm{cm}^{-1}], \tag{2}$$

where α is the acoustic amplitude absorption coefficient. The hydrodynamic theory yields:[12]

$$\alpha = \alpha_\eta + \alpha_{\eta'} = (\Omega^2 / 2v_0^3\rho)\{{}^4/_3\eta + \eta'\} \tag{3}$$

(v_0 is the sound velocity at low frequency, ρ the density, η and η' the shear and bulk viscosity coefficients, respectively, and $\Omega = 2\pi f$).

Relaxation theory leads to the well known result[13]

$$\alpha_{\eta'} = \frac{\Omega^2\tau(v_\infty^2/v_0^2 - 1)}{2v_0(1 + \Omega^2\tau^2)} \tag{4}$$

(τ is the relaxation time of the bulk viscosity coefficient, v_∞ is the sound velocity at infinite frequency). Comparing $\alpha_{\eta'}$ from (3) and (4) at low frequencies ($\Omega\tau \ll 1$), where they ought to be identical, one can obtain an equation for $\alpha_{\eta'}$ which is different from (4), and which is valid at any frequency:

$$\alpha_{\eta'} = \frac{\Omega^2}{2\rho v_0^3}\frac{\eta_0'}{1 + \Omega^2\tau^2}. \tag{4a}$$

From (4), (4a), and the expression

$$\frac{v_\Omega^2 - v_0^2}{v_\infty^2 - v_0^2} = \frac{\Omega^2\tau^2}{1 + \Omega^2\tau^2} \tag{5}$$

(v_Ω is the hypersonic velocity at the given frequency), we get an expression for the relaxation time of the bulk viscosity coefficient:

$$\tau = \frac{a \pm (a^2 - 4\Omega^2)^{1/2}}{2\Omega^2}, \quad a = \frac{\Omega^2\eta_0'}{\rho(v_\Omega^2 - v_0^2)}. \tag{6}$$

Equations (4) and (6) make it possible to find the relaxation time τ from two independent measurements—the absorption and velocity dispersion of the sound—and at the same time verify quantitatively the applicability of the relaxation theory with a single relaxation time.

The clear picture of the structure that arises in the excitation by light from the stimulated emission of a gas laser makes it possible to find not only the half-width of the shifted components, but also to measure the integral intensities of all three components of the fine structure and the distribution of the intensity in them, comparing the results obtained with the results of calculation:

$$\frac{J_c}{2J_{\mathrm{MB}}} = K\frac{\sigma^2 T}{c_p\rho\beta_s}, \quad K = \left(\frac{1}{\sigma}\frac{\partial\varepsilon}{\partial T}\right)_p^2 \Big/ \left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_s^2, \tag{7}$$

where c_p is the specific heat at constant pressure, σ is the volume expansion coefficient, and β_s the adiabatic compressibility.

If we neglect the difference between $(\rho\partial\varepsilon/\partial\rho)_s$ and $|(\sigma^{-1}\partial\varepsilon/\partial T)_p|$ and do not take into account sound velocity dispersion, then Eq. (7) reduces to the well-known Landau-Placzek formula:[14]

$$J_c / 2J_{\mathrm{MB}} = \gamma - 1; \tag{8}$$

Here $\gamma = c_p/c_v$ (c_v is the specific heat at constant volume).

EXPERIMENTAL PART

The study of the fine structure of the Rayleigh line has been carried out on the apparatus shown schematically in Fig. 1. The vessel V with the scattering liquid is placed inside a laser working with the mixture He-Ne ($\lambda = 6328$ Å). The exciting light is polarized in a plane perpendicular to the scattering plane. The scattered light was observed in a direction perpendicular to the direction of propagation of the exciting light. The spectrum of the scattered light was analyzed with a Fabry-Perot interferometer (FP) with multilayered dielectric mirrors and a thickness of the separator ring of 8 mm (dispersion region 0.625 cm^{-1}). The camera lens L_3 had a focal length F = 415 mm. The aperture of the scattered light was limited by the diaphragms D_1 and D_2 and had a value of ~0.03. The interferometer was placed in an isothermal box B which maintained a constant temperature within 0.03° C during the exposure. The half-width of the exciting line, together with the half-width of the apparatus function amounted to ~0.02 cm^{-1}. The scattered light was observed at an angle $\vartheta = 90° \pm 0.5°$. The liquids used were cleaned and rendered dust-free by multiple

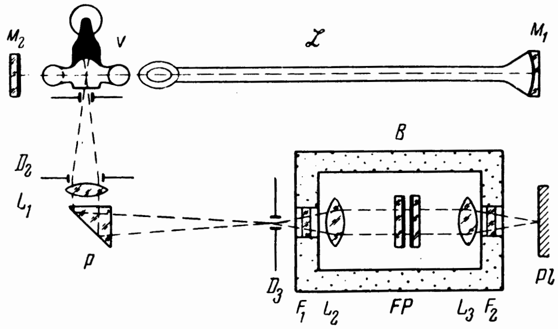

FIG. 1. The diagram of the apparatus for study of the fine structure of the Rayleigh line in liquids: $\mathscr{L}$ = laser tube, M_1 and M_2 are dielectric mirrors, V = vessel with the liquid to be studied, FP – Fabry-Perot interferometer, L_1, L_2, L_3 – objectives, D_1, D_2, D_3 diaphragms, P – reflecting prism, F_1 and F_2 – light filters, Pl – film, B – isothermal chamber.

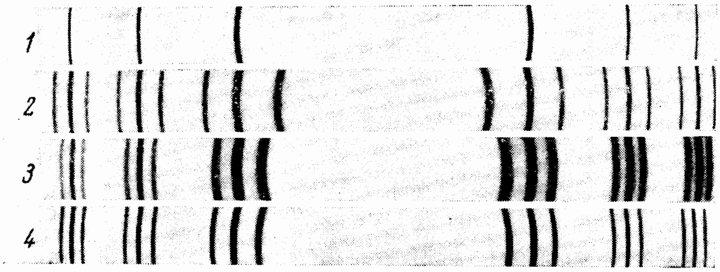

FIG. 2. Photographs of the fine structure of the Rayleigh scattering lines in liquids; 1 – exciting line λ = 6328Å, 2 – benzene, 3 – carbon tetrachloride, 4 – chloroform.

vacuum distillation in a sealed-off vessel by Martin's method. The vessel V (Fig. 1) for the liquid studied was made of quartz. The windows located in the path of the excited light beam were set at the Brewster angle.

Figure 2 shows photos of the fine structure in several liquids. Figure 3 gives the intensity distribution in the components of the fine structure of the Rayleigh line in benzene, obtained according to the rules of photographic photometry.

To determine the actual contours of the components of the fine structure, the procedure customarily used in such cases was applied.[2] The treatment of the photographs of the fine structure for the determination of the hypersonic velocity was carried out by the off-center method[15] on an IZ A-2 comparator. The error in the measurement of the hypersonic velocity amounted to ±0.5%. The value of this error is made up of the random errors in the determination of the location of the components of the fine structure and of the setting of the scattering angle. The values of the hypersonic velocities were referred to 20 ° C by means of the temperature coefficient for ultrasound.

The measurements have shown that contours of the central component and the MB components are close to those of a Gaussian distribution. The actual half-width of the MBC was determined as the difference between the visible half-width of the MBC and that of the central component. The broadening of the central component in the scattered light amounts to $\sim 10^{-4}$ cm^{-1}, which lies beyond the limits of accuracy of our method. Therefore the central component is suitable for calculation of the width of the exciting line together with the apparatus half-width. The accuracy of measurement of the width of the MBC amounted to $\pm 2 \times 10^{-3}$ cm^{-1}. Measurements of the ratio of the integral intensities were carried out with an accuracy of ±10–15%.

DISPERSION OF THE SOUND VELOCITY AND HYPERSONIC ABSORPTION IN LIQUIDS

The new measurements of the velocity of hypersound by the optical method give an accuracy that is an order of magnitude greater than that of previous measurements. In the case of benzene, carbon tetrachloride, carbon disulfide and toluene, the previous measurements of the sound velocity agree with the new data within the limits of experimental error. In the case of chloroform and methylene chloride, the accuracy of the previous measurements[6] was evidently overestimated and therefore the new data deviate from the previous measurements. The values of the hypersonic velocities obtained in the present work agree with the the data of other authors.[8, 9] For toluene, dispersion of the sound velocity was not observed

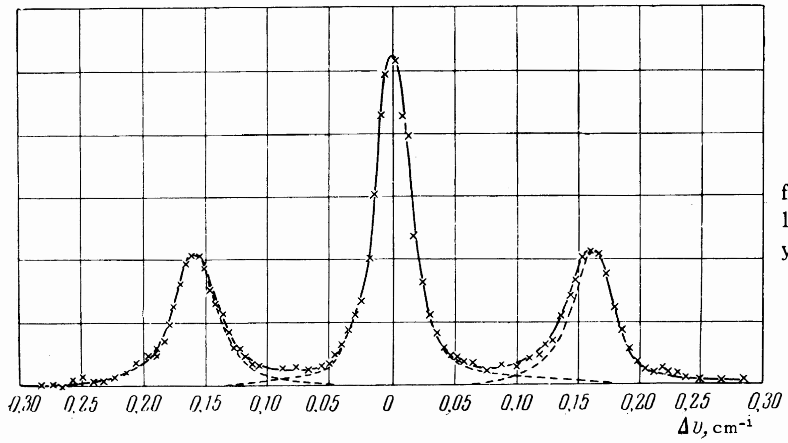

FIG. 3. Intensity distribution in components of the fine structure of the scattering lines in benzene: solid line – experimental curve, broken line – result of analysis.

Table I. Results of measurement of the hypersonic velocity in liquids.

Substance	$f \times 10^{-9}$, cps	v, m/sec	$\frac{\Delta v}{v_\Omega} \cdot 10^2$	ρ, σ/cm	$\eta \times 10^3$, poise	η'_0, poise	$\tau \times 10^{10}$, sec	$\alpha \times 10^{-2}$, cm from (4)	v_∞, m/sec.
C_6H_6	4,94±0.02	1471±8	10	0,878	6.5	0.921	2,5±0.1	5.5±0.4	1474±8
CCl_4	3,31±0.01 (3.54) *	1015±6 (1022) *	10	1.595	9.7	0,339	0,93±0.1 (0.82) *	14,4±2.3 (18.1) *	1036±13 (1043) *
$CHCl_3$	3.4±0.01	1055±5	5	1.498	5.8	0.306	1.68±0.2	4.52±1.0	1096±14
$C_6H_5CH_3$	4.42±0.02	1326±7	0	0.867	5.9	0.079	—	—	—
CH_2Cl_2	3.53±0.02	1113±6	2	1.336	4.3	1,071	17±5	0.91±0.1	1113±8

*These values for CCl_4 are obtained if one takes into account the effect of absorption of the hypersound at the location of the MBC maximum. In this case,[1] $\Delta\nu_{max} = 1 - (\frac{1}{2})\, \delta\nu^2_{MB}/\Delta\nu^2)^{1/2} \Delta\nu$, where $\Delta\nu$ is determined from (1). This correction was negligible in the other liquids studied.

within the range of accuracy of the measurement[1)] (Table I).

The relaxation time of the bulk coefficient of viscosity τ was determined by means of ultrasonic measurement of η'_0 and also of the values of v_Ω and v_0 from Eq. (6). This quantity is listed in Table I. The limiting value of the velocity v_∞, found from Eq. (5), is given in the last column of the table.

The absorption coefficient $\alpha_{\eta'}$, can also be calculated from the data already obtained and from Eq. (4). On the other hand, the absorption α is directly computed from the width of the MBC according to Eq. (2). The results of such calculations are shown in Table II. The absorption coefficient, which is due to the shear viscosity coefficient, is shown in the fourth column of the table (the calculations are carried out from Eq. (3) for $\eta'_0 = 0$). The absorption coefficient due to the bulk viscosity coefficient $\alpha_{\eta'} = \alpha - \alpha_\eta$. In this case, the relaxation term of the bulk viscosity coefficient was found from the absorption data and Eq. (4a) [see the fifth column of Table II].

In those cases in which the values of τ have been obtained from the dispersion of the sound velocity and the absorption of the hypersound from the formulas of relaxation theory, one can say that the theory gives an excellent description of the phenomenon. Such a conclusion can be made relative to benzene, carbon tetrachloride, methylene chloride (see below), and the previously studied carbon disulfide. For chloroform, the τ computed from the dispersion of the sound velocity and from the hypersonic absorption differ widely.

In the already mentioned work of Chiao and Stoicheff,[9] it is emphasized that for benzene, carbon tetrachloride, chloroform and toluene, $\tau \geq 4 \times 10^{-10}$, which contradicts our measurements of the width of the MBC and the dispersion of the sound velocity.

Graphs of the dependence of α/f^2 on log f were constructed for all the cases studied (Fig. 4). Here, the formula of relaxation theory with a single relaxation was used for $\alpha_{\eta'}$. For the region of ultrasonic frequencies, the data of ultrasonic measurements were used, while for the hypersonic region, we employed the values from our own optical measurements (Tables I and II).

The curves constructed for benzene, carbon tetrachloride, and methylene chloride are very well described by the relaxation theory formula with a single τ if one assumes that α/f^2 is correctly measured for ultrasonic and hypersonic frequencies. Especially noteworthy is the curve for methylene chloride, where the different ultrasonic and our own hypersonic measurements lie on a single relaxation curve. However, it is necessary to note that direct measurements of the absorption of hypersound in methylene chloride have not yet been made, since this quantity is at the limit of sensitivity of our method. The absorption is computed from the dispersion of the sound velocity.

The relaxation curves for chloroform are plotted in Fig. 5. The three different curves correspond to the case in which the curve is con-

Table II. Results of measurement of the width of the Mandel'shtam-Brillouin components in liquids

Substance	$\delta\nu \times 10^3$, cm^{-1}	$\chi \times 10^{-3}$, cm^{-1}	$\alpha_\eta \cdot 10^{-3}$, cm^{-1}	$\alpha_\eta \times 10^{-2}$, cm^{-1}	$\tau \times 10^{10}$	$\alpha/f^2 \times 10^{17}$ cm^{-1}
C_6H_6	7±2	4.5±1.3	2,0	2.5	3.03±1.0	18.4±0.5
CCl_4	17±3	16 ±3	2,0	14	0.88±0.1	146 ±27
$CHCl_3$	11±2	10 ±2	1.15	9	0,96±0.1	87 ±17
$C_6H_5CH_3$	10±2	7 ±1,5	1.48	5.6	0.46±0.1	36 ±8
CH_2Cl_2	~2	—	—	—	—	—
CS_2	~3	—	—	—	—	—

[1)] According to the data of Chiao and Stoicheff[9] a dispersion of about 4% was obtained in toluene at a frequency $f = 6.4 \times 10^9$ cps.

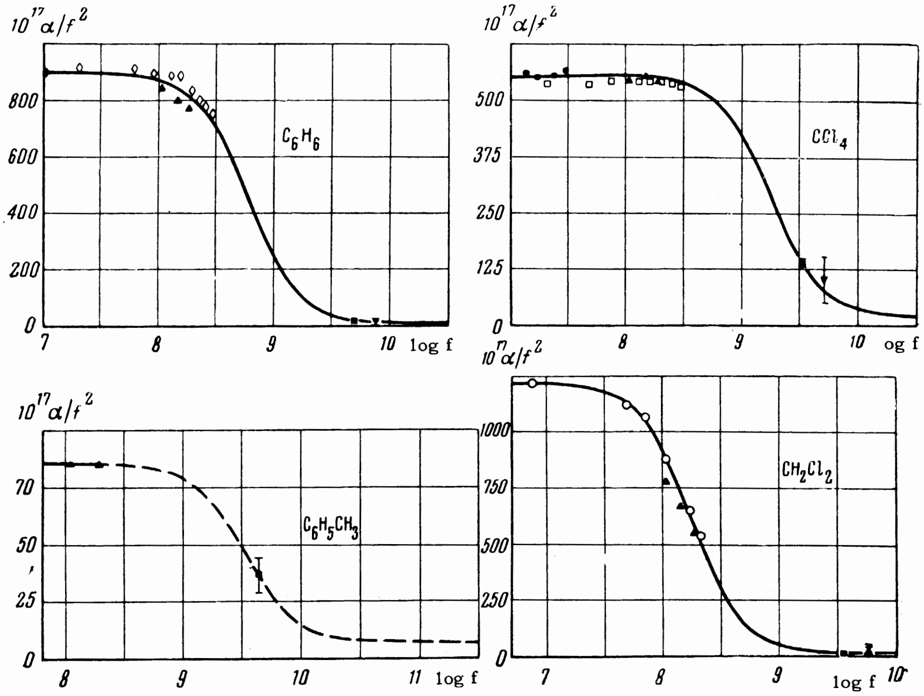

FIG. 4. Dependence of α/f^2 on log f for benzene, carbon tetrachloride, toluene, methylene chloride ■ – present research, ▼ – [2], ◆ – [21] ▲ – [16], ◇ – [18], □ – [19], ● – [17], × – [6], o – [20]. In the case of toluene, the point ■ was obtained from the width of the MBC.

structed (1) from ultrasonic measurements,[19] (2) from ultrasonic data and hypersonic data for the absorption, computed from the dispersion of the sound velocity, and (3) from ultrasonic data[19] and hypersonic data, obtained from the MBC curve, that is, for hypersonic absorption. It is possible that the ultrasonic and hypersonic data in the case of chloroform cannot be described by the formulas of relaxation theory, which take into account only a single relaxation time.

THE RATIOS OF THE INTENSITY OF THE FINE STRUCTURE COMPONENTS

The accurate separation of the fine structure components in the scattered-light spectrum for excitation by light with $\lambda = 6328$ Å of the stimulated emission of a He-Ne gas laser makes it possible to bring about an improvement in the accuracy of earlier measurements. In the study of the fine structure components in benzene, 15 different photographs were taken and it was made clear that the intensity distribution over all three components was close to Gaussian. The intensity between the n-th order Stokes component and the (n−1)st order anti-Stokes components was considered by us to be equal to the intensity of the continuous spectrum produced by the wings of the Rayleigh line.2) The intensity at the same point, brought about by the departure from unity of the reflection coefficient of the mirror of the interferometer, was a negligibly small quantity in our experiment.

The method used for processing the fine-structure spectra can bring about appreciable errors in the "tails" of the intensity distribution

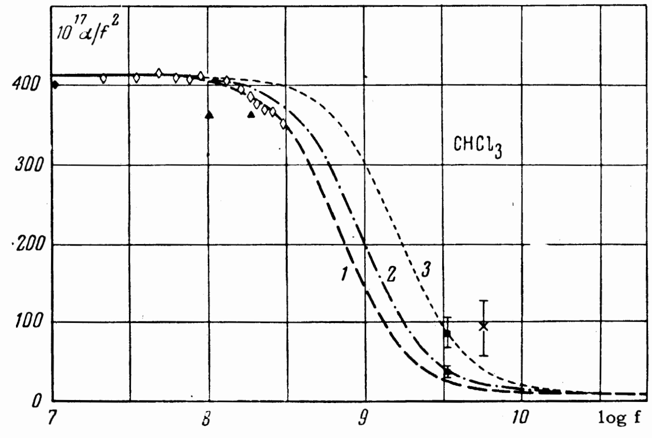

FIG. 5. Dependence of α/f^2 on log f for chloroform (for the meaning of the points, see Fig. 4).

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2)In the present research, the scattered light was produced by an optical laser polarized in a plane perpendicular to the scattering plane. The photograph of the scattered light spectrum showed, besides the polarized discrete components, also continuous, depolarized light (the wing of the Rayleigh line). The scattered light was not photographed in two mutually perpendicular polarizations; therefore, interpretation by the method described in[2] was not carried out. The dispersion region of the interferometer was 0.625 cm^{-1}, while the half-width of the same narrow part of the wing was ~ 2 cm^{-1}. Therefore, we have assumed that in our spectra the intensity distribution in the continuous spectrum was uniform.

Table III. Ratio of intensities in fine structure components ($I = J_c/2J_{MB}$)

Substance	I, experiment		I calc. from (8)	Calc. from (7)*		Calc. from (7)**			
	Present research	Previous measurements		*K*	*I*	"low" $R_{\pi/2}$		"high" $R_{\pi/2}$	
						K	*I*	*K*	*I*
C_6H_6	0.72 ± 0.1	0.97 [22] 0.98 ± 0.25 [2]	0,46	1.10	0,60	1,11	0.60	0.62	0.34
CCl_4	0.67 ± 0.15	0.84 [22] 1.10 ± 0.3 [2]	0.46	—	—	1,04	0.56	0.65	0,35
$CHCl_3$	0.58 ± 0.1	—	0.51	1.08	0.58	1,41	0,75	0.79	0,42
$C_6H_5CH_3$	0.45 ± 0.06	0.40 [23]	0.36	1.24	0.43	1.74	0,60	0.93	0.32
CH_2Cl_2	0.72 ± 0.1	—	0.54	—	—	—	—	—	—

*Dynamical values of $(\rho\partial\epsilon/\partial\rho)_s$ are used.

**Values of $(\rho\partial\epsilon/\partial\rho)_s$ are computed from the light scattering coefficient.

of all three components. Therefore, it was deemed advantageous to determine the integral intensity of the components not by the direct measurement of the areas enclosed by the contours of the components, but by measurement of the maximal intensities of the components and their visible half-widths. In this case, as is well known,

$$\frac{J_c}{2J_{MB}} = \left(\frac{J_c}{2J_{MB}}\right)_{max} \frac{\delta\nu_c}{\delta\nu_{MB}}. \tag{9}$$

The results of the experimental data for $J_c/2J_{MB}$ are given in Table III. In this table are shown the previous measurements, made with a mercury arc as a source of the exciting light. It is necessary to note that although the previous data, within the limits of a rather large experimental error, practically overlaps the recent results, the latter are systematically lower than the former.

As liquids to be studied, we also chose those in which there is observed a dispersion in the sound velocity (an exception is toluene). These very liquids, as is clear from Eq. (7) are of greatest interest, inasmuch as they must reveal the greatest divergence from the formulas of the theory, which do not take into account the sound dispersion and the $K = 1$ assumed in Eq. (7).[3)]

To compare the experimental data with that calculated by Eq. (8) is of no difficulty. Such a comparison (see Table III) shows that the agreement with experiment in a majority of the materials investigated here is unsatisfactory. A comparison of the measured values of $J_c/2J_{MB}$ with the result of calculation from Eq. (7) encounters purely technical difficulties, which consist of the fact that the quantities entering into Eq. (7) are either completely unknown or are known with great uncertainty. This applies primarily to such quantities as $(\rho\partial\varepsilon/\partial\rho)_s$ and $(\sigma^{-1}\partial\varepsilon/\partial T)_p$ (see Appendix).

In principle, the best possibility of finding $(\rho\partial\varepsilon/\partial\rho)_s$ is the computation of this quantity from the experimentally determined scattering coefficient $R_{\pi/2}$ and the general formula for light scattering from which it follows that

$$\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_s^2 = \rho v^2\left[\frac{2\lambda^4 R_{\pi/2}}{\pi^2 kT f(\Delta)} - \left(\frac{1}{\sigma}\frac{\partial\varepsilon}{\partial T}\right)_p^2 \frac{\sigma^2 T}{\rho c_p}\right], \tag{10}$$

where $f(\Delta) = (1 + \Delta)/(1 - (7/6)\Delta)$, Δ is the coefficient of depolarization of the scattered light. But so far the accuracy of the measurement of $R_{\pi/2}$ is not large. Furthermore, there exist two groups of measurements of the scattering coefficient—the so-called "high" and "low" values.[1, 2] There is excellent agreement between the experimental results inside each group, but between these groups, the measurements diverge—for benzene, for example, by 40%. Therefore, we determined $(\rho\partial\varepsilon/\partial\rho)_s$ separately, from the "low" and "high" values of $R_{\pi/2}$. Moreover, we calculated the value of K. This same quantity was determined from the dynamical measurements of $(\rho\partial\varepsilon/\partial\rho)_s$.[5]

According to thermodynamic calculations, K should be equal to or greater than unity.[2] This is also obtained from the dynamic values of $(\rho\partial\varepsilon/\partial\rho)_s$, and the "low" values of $R_{\pi/2}$ and Eq. (10). From the "high" values of $R_{\pi/2}$ and Eq. (10), one systematically gets $K < 1$, which we are inclined to consider as evidence that the "high" values are incorrect.

Undoubtedly, the application of gas lasers to the

[3)] In the work of Sen,[24] it was shown that if one computes K, using the formula of Clausius-Mosotti for the connection between ϵ and ρ, then the calculation yields $K = 1$. In the calculation it is assumed that the constant C in the Clausius-Mosotti formula does not depend on the temperature or, in other words, it is assumed that $(\partial\epsilon/\partial T)_\rho = 0$. In this approximation, as shown previously,[2] the value of K is unity for any connection of ϵ and ρ. But just such a definition is inadmissible in a number of cases[1,2] (see also Table III).

Table IV

Substance	ρ, g/cm^3	n	$\sigma \cdot 10^5$, град$^{-1}$	$c_p \times 10^{-7}$ erg/deg	$\frac{dn}{dT} \cdot 10^5$	$R_{\pi/2} \cdot 10^6$		$\left(\rho \frac{\partial \varepsilon}{\partial \rho}\right)_s$	$\Delta \cdot 100$
						"low"	"high"		
C_6H_6	0,879	1.499	121	1.70	64	32	48	1.56	43
CCl_4	1.595	1.462	122	0.84	56	10.4	14	—	6
$CHCl_3$	1,489	1,445	126	0,966	61	12,8	18.6	1.36	20
$C_6H_5CH_3$	0,865	1,494	106	1,68	61	35,8	55	1,6	48

Note: For $(\rho \partial \epsilon/\partial \rho)_s$, the dynamical values are given.

investigation of the intensity of the components of the fine structure makes it possible to measure the value of J_c/J_{MB} with still greater accuracy than at present; it will then be advantageous to determine the value of $(\rho \partial \varepsilon/\partial \rho)_s$ from these measurements and Eq. (7). It is possible that even now such a determination would be of interest.

In conclusion, the authors thank V. P. Zaĭtsev, who made work possible with mirrors of multiple dielectric coatings.

APPENDIX

For the calculation of the ratios of the intensities from Eq. (7), which are given in Table III, we used numerical values given in Table IV.

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Translated by R. T. Beyer
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