

Temperature dependence of the velocity and absorption of hypersound in the critical region of binary mixtures

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The velocity and absorption of hypersound are measured in critical mixtures by the light scattering technique over a broad temperature range. It is found that as the critical temperature is approached the hypersonic velocity increases and remains practically constant near the critical temperature. The absorption of the hypersound increases near the critical temperature. The results are compared with ultrasonic data obtained in the same mixtures. It is shown that current theories cannot describe the velocity and absorption of hypersound in critical mixtures at frequencies above 4 GHz.

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1. INTRODUCTION

Molecular acoustics makes it possible to obtain extensive information on critical phenomena and the physical characteristics of an investigated substance.¹ The purpose of our research was to study the behavior of the velocity and the absorption coefficient of hypersound, the velocity of ultrasound in binary mixtures of aniline-cyclohexane and nitrobenzene-n-hexane over a wide range of temperatures and at the closest possible approach to the critical temperature T_{cr} .

The studies of the hypersound at frequencies exceeding 4.5 GHz were carried out by measurement of the location and width of the Mandel'shtam-Brillouin components (MBC). Such measurements are greatly complicated by the extraordinarily intense central component, instrumental wings which are superposed on the MBC and distort them. It is possible to avoid most of the difficulty by increasing the contrast of the spectrum, using a multipass Fabry-Perot interferometer.² A two-pass interferometer was used in the present work, constructed at Samarkand University.³ The temperature dependence of the ultrasonic velocity at a frequency of 2.8 MHz was measured in the same critical-mixtures sample by a method previously described.⁴

The results obtained are discussed in the final section on the basis of theories existing at the present time.

2. EXPERIMENT

The schematic diagram of the experimental setup is the same as in Refs. 1 and 3 but with a two-pass interferometer.

The scattering was excited by radiation of a single-frequency He-Ne laser LG-159 ($\lambda = 6328 \text{ \AA}$), with a power $\sim 2 \text{ mW}$. The spectrum of the light scattered at an angle $90 \pm 0.2^\circ$ relative to the direction of propagation of the excited beam was investigated. The polarization of the exciting and scattered light was perpendicular to the scattering plane. The spectrum of the scattered light was scanned by variation of the pressure in the interferometer chamber.⁵ The nonlinear distortion of the spectrum did not exceed 1% over the range of three orders of the interferogram.

The diaphragm in front of the photomultiplier was so chosen that its magnitude did not have a significant effect on the total value of the instrumental contour. To record the light signal we used an FÉU-79 photomultiplier cooled to a temperature of -30°C and operating in the photon-counting regime. Upon cooling, the noise of the photomultiplier decreased to 5–10 counts/sec, and the sensitivity was not lowered appreciably. The signal from the photomultiplier was fed to a linear intensimeter PI-4 and was recorded with a potentiometer KSP-4.

The investigation of stratified mixtures was carried out both at a temperature above the critical temperature of stratification T_{cr} , when the medium is homogeneous and at a temperature below T_{cr} , when the mixture is stratified. The measurements were carried out for each of the two components separately. The constructed thermostat made it possible to maintain the temperature of the vessel with the liquid under study constant over a wide range of temperatures, and to maintain the temperature of the sample constant to within 0.05°C . The temperature was controlled by a Beckman thermometer to within $\pm 0.02^\circ\text{C}$. The investigated samples were prepared from optically purified materials of ChDa (analytically pure) grade by the method of Martin.¹ The measurement of the velocity of propagation of the ultrasound was performed by the method previously described in Ref. 4, with an error not exceeding 0.25%. The contrast $C = 6 \times 10^4$ and the finesse $F = 35 - 30$, by which our apparatus was characterized, made it possible to measure the value of the shift with accuracy to within $\pm 0.5\%$ up to $\Delta T = \pm 1.5^\circ\text{C}$ and $\mp 1\%$ upon closer approach to T_{cr} , while the value of the absorption, i.e., the width of the MBC, was measured to within 5–10%, significantly decreasing the effect of the central component of the MBC.

The velocity of propagation of the hypersound is equal to

$$V = \Delta v_0 c / 2n \nu \sin(\theta/2), \quad (1)$$

where c , n and θ are the velocity of light, index of refraction, and scattering angle, respectively.

In this case, the actual value Δv_0 was determined from

the scattering spectra with account taken of their finite width:^{1, 6, 7}

$$\Delta\nu_0 = \Delta\nu(1 - \delta\nu^2/2\Delta\nu^2)^{-1/2} \quad (2)$$

Here $\Delta\nu$ is the visible shift of the MBC, $\delta\nu$ is the width of the MBC. This allowance for the effect of the widths of the MBC on their shift gives a correction $\sim 1\%$ to the value of the hypersonic velocity for mixtures of nitrobenzene-n-heptane.

The absorption coefficient of the hypersound α was calculated from the relation¹:

$$\alpha = \pi c \delta\nu_0/V, \quad (3)$$

where $\delta\nu_0$ is the true value of the width of the MBC, obtained by elimination of the instrumental width of the contour $\delta\nu_{inst}$ from the observed width.

The value of the actual width of the components was obtained from a graph of the dependence of the ratios $\delta\nu_0/\delta\nu_{inst}$ and $\delta\nu/\delta\nu_{inst}$ calculated for our two-pass interferometer similarly to what was done in Ref. 8 for three-pass and five-pass Fabry-Perot interferometers.

3. EXPERIMENTAL RESULTS

a) *Aniline-cyclohexane*. The dependences of the shifts of the MBC on the degree of approach to the critical temperature $T_{cr} = 32.2^\circ\text{C}$ are given in Fig. 1 for an aniline-cyclohexane mixture of critical concentration ($k = 0.44$ molar fraction of aniline). At $T < T_{cr}$, the shifts of the MBC in each component of the mixture differ considerably from one another, and as the critical temperature is approached the difference decreases, and near the critical temperature T_{cr} the shifts approach the shift for a homogeneous mixture. If the critical temperature is approached from above, $T > T_{cr}$, then in the temperature range $\Delta T = 1.65 - \sim 7^\circ\text{C}$, an increase in the MBC shift is observed. In the temperature range $\Delta T \sim 7 - 1.5^\circ\text{C}$, the MBC shift practically does not change and remains essentially constant within the limits of accuracy of the experiment. Upon further approach to the critical temperature ($\Delta T \sim 1.5 - 0.3^\circ\text{C}$) the MBC shift evidently increases somewhat, although the accuracy of our experiment did not permit us to make an unambiguous inference.

Since the value of the propagation velocity of hypersound is linearly related with the MBC shift, the char-

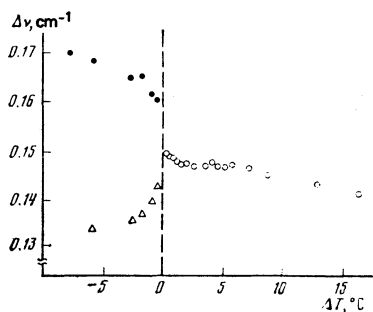


FIG. 1. Dependence of the value of the MBC shift as one approaches the critical temperature T_{cr} in the aniline-cyclohexane mixture: ●—aniline component, Δ—cyclohexane component (stratified mixture); ○—homogeneous mixture.

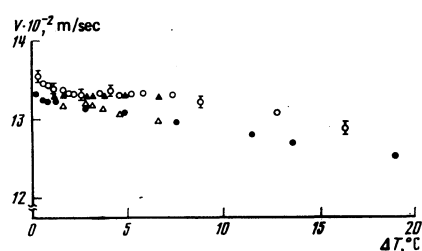


FIG. 2. Dependence of the sound propagation velocity in the aniline-cyclohexane mixture as T_{cr} is approached: ○—hypersound 4.7 GHz and ●—2.8 MHz—our measurements; ▲—15 MHz and Δ—2 MHz—data of Ref. 9.

acter of the change in the velocity, calculated from the MBC shift as a function of temperature, corresponds fully to the character of the shift of the MBC (Fig. 2).

Figure 2 shows, in addition to the dependence of the propagation velocity of the hypersound on the temperature, the dependence of the ultrasound of frequency 2.8 MHz, measured in the same mixtures in this research, as well as the data of Ref. 9, obtained at frequencies of 15 and 2 MHz. As is seen in Table I, the dispersion of the sound velocity reaches a value of $\sim 1.5\%$, which is beyond the limit of error of both experiments. As the temperature increases, the dispersion of the velocity falls off gradually.

The agreement of our hypersonic results with the data of Ref. 9 for a frequency of 15 MHz shows that in this range of frequencies, sound velocity dispersion is absent, within the limits of error of both experiments, in the temperature range $\Delta T = 2 - 6^\circ\text{C}$. The data of our measurements at a frequency of 2.8 MHz and the measurements of Ref. 9 at the frequency 2 MHz are in complete agreement.

Figure 3 gives the dependence of the MBC width on the temperature as measured by us. Just as in the case of the MBC shifts, the widths were measured for each component of the mixture at $T < T_{cr}$ in the stratified mixture. As T_{cr} is approached, the MBC width in each component of the mixture increases and reaches a maximum value near T_{cr} .

If T_{cr} is approached from above (homogeneous phase), then it is seen from Fig. 3 that, far from T_{cr} , the width

TABLE I. Aniline-cyclohexane ($k_{cr} = 0.44$ molar fraction aniline; $T_{cr} = 32.2 \pm 0.05^\circ\text{C}$).

$\Delta T, ^\circ\text{C}$	$\Delta V/V, \%$		$\alpha \cdot 10^{-3}, \text{cm}^{-1}$		η, cP
	experiment	calculated by formula (5)	experiment	calculated by formula (6)	
0.3	1.5	0.15	27	0.004	1.895
0.5	1.8	0.15	23	0.003	1.87
0.8	1.5	0.15	21	0.005	1.855
1.1	—	—	18.6	—	1.835
1.2	1.3	0.3	18.6	0.006	1.83
1.6	—	—	16.3	—	1.80
1.9	1.2	0.4	14.4	0.009	1.785
2.5	—	—	—	—	1.745
2.6	—	—	13.9	—	1.74
3.6	—	—	14.1	—	1.695
4.1	—	—	14.1	—	1.675
4.6	1.8	0.6	13.6	0.013	1.66
5.2	—	—	13.1	—	1.635
5.8	—	—	14.4	—	1.58
7.3	—	—	14.8	—	1.565
7.6	—	—	—	—	1.55
8.9	—	—	14.2	—	1.51
12.9	3	1.1	13.5	0.025	1.41
16.4	—	—	13.8	—	1.335

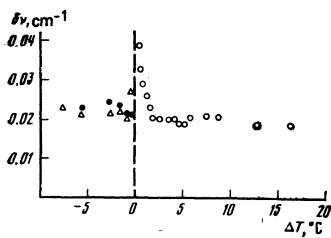


FIG. 3. Dependence of the MBC width in the aniline-cyclohexane mixture as T_{cr} is approached. Notation is the same as in Fig. 1.

of the MBC increases slowly as T_{cr} is approached, and a rapid increase in the MBC width is observed in the temperature range $\Delta T = 1.5 - 0.3^\circ\text{C}$. Such a change in the MBC width corresponds to a change in the absorption coefficient of hypersound from $13.76 \times 10^3 \text{ cm}^{-1}$ to $27 \times 10^3 \text{ cm}^{-1}$ (Table I). According to our measurements, the character of the change in the hypersonic absorption upon approach to the critical temperature agrees qualitatively with the ultrasonic data.¹⁰

b) *Nitrobenzene-n-hexane*. Figure 4 shows the results of measurement of the MBC shift in a nitrobenzene-n-hexane mixture of critical concentration ($k = 0.4$ molar fraction of nitrobenzene) ($T_{cr} = 19.95 \pm 0.05^\circ\text{C}$) as a function of the approach to T_{cr} . Figure 5 gives the values of the hypersonic propagation velocity, obtained from the MBC shifts and from the ultrasonic propagation velocity measured in the same mixtures.

The measurements of the MBC width in this same mixture and the values of the absorption coefficient calculated from formula (1) are given in Fig. 6 and Table II.

The behavior of the propagation velocity and the hypersonic absorption coefficient in the nitrobenzene-n-hexane mixture is analogous to the change in these quantities in the aniline-cyclohexane mixture. It must be noted that the sound velocity dispersion in the nitrobenzene-n-hexane mixture is significantly greater than the sound velocity dispersion in the aniline-cyclohexane mixture, and reaches a value of 6%.

In the present research, measurements of the MBC shifts and their widths have also been made in aniline-cyclohexane and nitrobenzene-n-hexane mixtures with different concentrations at temperatures of 36.5 and 25.5°C , respectively.

4. DISCUSSION OF RESULTS

We now discuss the results that we have obtained from the viewpoint of current theories which describe the

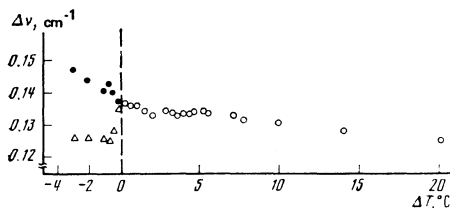


FIG. 4. Dependence of the value of the behavior of the MBC as T_{cr} is approached in the nitrobenzene-n-hexane mixture: ●—nitrobenzene component, Δ —n-hexane component (stratified mixture); ○—homogeneous mixture.

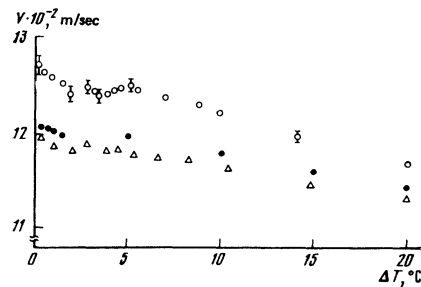


FIG. 5. Dependence of the sound propagation velocity in the nitrobenzene-n-hexane mixture as T_{cr} is approached; ○—hypersound 4.7 GHz and Δ —2.8 Mhz—our measurements; ●—15 MHz—data of Refs. 9 and 10.

sound propagation in critical mixtures.

In the region of applicability of the formulas of hydrodynamic theory ($q\xi \leq 1$, where q is the wave vector, ξ is the correlation radius) the sound absorption coefficient is described by the expression

$$\alpha = \frac{2\pi^2 f^2}{\rho V_0^3} \left\{ \frac{4}{3} \eta + \eta' \right\}, \quad (4)$$

where η is the shear viscosity, η' the bulk viscosity. For a critical mixture consisting of two liquids, this region extends to $\Delta T \sim 0.1^\circ\text{C}$, which in our experiment includes the entire range of temperatures in which the experiments were carried out.

According to the expression (4), in a medium the sound absorption coefficient described by hydrodynamic theory is the sum of the absorption coefficient η determined by the shear viscosity and the absorption coefficient η' determined by the bulk viscosity. Knowing the value of the shear viscosity η , measured with a viscosimeter, we can calculate the absorption coefficient connected with it and, consequently, calculate the absorption coefficient determined by the bulk viscosity. Figures 7 and 8 give the temperature dependences of the quantities α_η/f^2 and $\alpha_{\eta'}/f^2$, calculated from formula (4) for the aniline-cyclohexane and nitrobenzene-n-hexane mixtures. As is seen in Fig. 8, the absorption coefficient determined by the shear viscosity is smaller by a factor of severalfold than the absorption coefficient determined by the bulk viscosity, while the absorption coefficient $\alpha_{\eta'}/f^2$ increases rather strongly as T_{cr} is approached closer than 1.5°C . Such an increase in $\alpha_{\eta'}/f^2$ as T_{cr} is approached indicates that the bulk viscosity increases as T_{cr} is approached, since the remaining

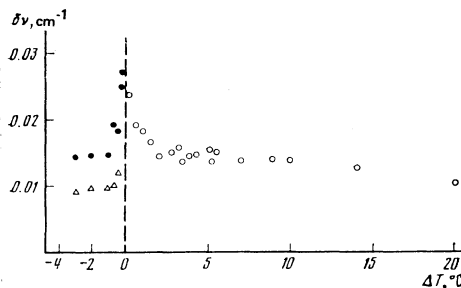


FIG. 6. Dependence of the MBC width as T_{cr} is approached in the nitrobenzene-n-hexane mixture. Notation is the same as in Fig. 4.

TABLE II. Nitrobenzene-n-hexane ($k_{cr} = 0.4$ molar fraction nitrobenzene, $T_{cr} = 19.95 \pm 0.05$ °C).

$\Delta T, ^\circ C$	$\Delta V/V, \%$		$\alpha \cdot 10^{-3}, cm^{-1}$		η, cP
	experiment	calculated by formula (5)	experiment	calculated by formula (6)	
0.2	7	—	47	—	0.715
0.6	—	—	44	—	0.702
1	6.3	0.3	13.7	0.09	0.695
1.5	5	0.3	12.6	0.1	0.685
2	5	0.35	11.3	0.1	0.672
2.9	—	—	11.5	—	0.657
3.3	5	0.4	11.8	0.15	0.652
3.5	—	—	10.8	—	0.65
4	5	—	11.1	—	0.64
4.3	—	—	11.3	—	0.637
4.6	5	0.6	11	0.15	0.632
5.2	—	—	11.7	—	0.625
5.5	5.6	0.6	11.4	0.16	0.62
7.1	—	—	10.8	—	0.598
8.9	—	—	10.9	—	0.575
10	5	—	10.5	—	0.562
14.1	—	—	9.9	—	0.528
20.1	—	—	8.8	—	0.49

quantities entering into (4) have a weak temperature dependence.

Plots of the ultrasonic and hypersonic propagation velocities are presented in Figs. 2 and 9. These were obtained by us and other authors as T_{cr} is approached in aniline-cyclohexane and nitrobenzene-n-hexane mixtures.

It is seen from Fig. 9 that our results and the measurements of the hypersonic velocity in the nitrobenzene-n-hexane mixture as T_{cr} is approached are in excellent agreement with the data of Ref. 11 over the entire temperature interval, and differ somewhat from the values of the hypersonic velocity obtained in Ref. 12. This is possibly connected with the fact that a rather powerful argon laser (250 mW) was used in Ref. 12, which evidently led to local heating of the sample and to some inaccuracy in the measurement of the mixture temperature.

A significant decrease in the hypersonic velocity was observed in Ref. 11 in the temperature range $\Delta T \sim 0.5$ °C; it fell to a value of 1210 m/sec, while in our experiment a practically constant hypersonic velocity was observed (Figs. 2 and 9) or even a rather weak tendency to increase as T_{cr} is approached, although it is not possible to consider the latter as certain because of the rather large error ($\sim 1.0\%$) in the determination of the hypersonic propagation speed in this temperature range.

In our work, we made an attempt to describe the observed sound velocity dispersion (Figs. 2 and 5, Tables I and II) and the increase in the absorption coefficient (Tables I and II) as the critical temperature is ap-

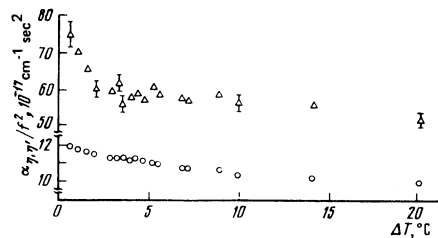


FIG. 7. Temperature dependence of the quantity $\alpha_{\nu}/f^2(\Delta)$ and $\alpha_{\nu}/f_2(\circ)$ in the aniline-cyclohexane mixture.

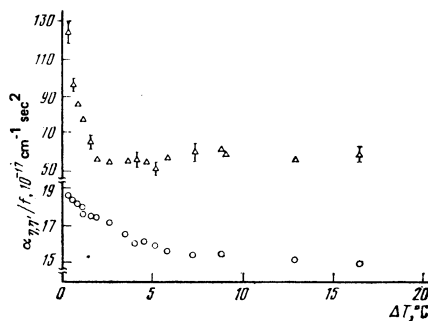


FIG. 8. Temperature dependence of the quantities $\alpha_{\nu}/f^2(\Delta)$ and $\alpha_{\nu}/f_2(\circ)$ in the nitrobenzene-n-hexane mixture.

proached, from the viewpoint of existing theories of coupled modes, developed by Kawasaki¹³ and Chaban^{14,15}. Both theories give expressions characterizing the propagation velocity and the sound absorption coefficient in critical mixtures as T_{cr} is approached, while the formulas of both theories are practically identical. We use the formulas of the Chaban theory¹⁵ and consider the results that we obtain from the viewpoint of this theory.¹⁾

The sound velocity dispersion and the absorption coefficient are described by the following expressions¹⁵:

$$\frac{V-V_0}{V_0} = -\frac{MV_0}{\tau_1} [|T-T_{cr}| + d |k-k_{cr}|^3]^{-0.2} \left[F_2(\Omega\tau) - \frac{b \arctg b}{12} \right], \quad (5)$$

$$\frac{\alpha}{2\pi f^2} = M [|T-T_{cr}| + d |k-k_{cr}|^3]^{-2} F_1(\Omega\tau) + L, \quad (6)$$

where

$$M = \frac{1.44 k_B T \rho V_0 \tau_1}{4\pi^2 \xi_1^3} \left(-\frac{\partial T_{cr}}{\partial P} + \frac{T\kappa}{\rho C_p} \right)^2. \quad (7)$$

b is a constant of the order of several units, k_B is the Boltzmann constant, κ is the coefficient of thermal expansion; C_p is the heat capacity at constant pressure, $\tau_1 = \xi_1^2/D_1$; ξ_1 and D_1 are the correlation radius and the coefficient of mutual diffusion, respectively, at the temperature $\Delta T = 1$ °C, k is the concentration of the mixture and differs from the critical k_{cr} , $F_1(\Omega\tau)$ and $F_2(\Omega\tau)$ are functions determined on a highspeed computer up to the value $\Omega\tau = 10^6$. In calculations with formulas (5) - (7), the quantities given in Table III were used.

The sound velocity dispersion obtained from the experimental values near T_{cr} reached values of 1.5 and 6% respectively in the aniline-cyclohexane and nitrobenzene-n-hexane mixtures. The sound velocity dispersion ob-

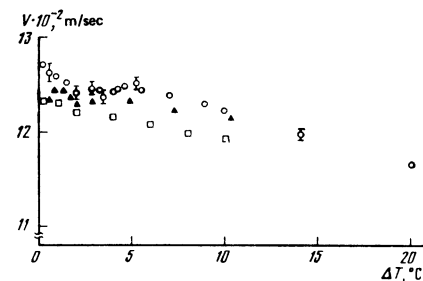


FIG. 9. Temperature dependence of the propagation velocity of hypersonic in the nitrobenzene-n-hexane mixture; \blacktriangle —data of Ref. 11, \square —data of Ref. 12, \circ —our measurements.

TABLE III. Quantities used in the calculation by formulas (5)–(7).

	Aniline-cyclohexane	Nitrobenzene-n-hexane
ξ_1 , cm	$6.997 \cdot 10^{-7}$	$4.411 \cdot 10^{-7}$
D_1 , cm ² ·sec ⁻¹	$2.136 \cdot 10^{-7}$	$5.85 \cdot 10^{-7}$
Ω_1 , Hz	$2.79 \cdot 10^{10}$	$2.57 \cdot 10^{10}$
$\partial T_{cr}/\partial P$, deg·cm ² ·dyn ⁻¹	$0.68 \cdot 10^{-8}$	$-8 \cdot 10^{-9}$
$T \times (\rho C_p)$, deg·cm ² ·dyn ⁻¹	$1.23 \cdot 10^{-8}$	$1.63 \cdot 10^{-8}$
$F_1(\Omega\tau)$	$1.52 \cdot 10^{-7} - 1.43 \cdot 10^{-4}$	$1.065 \cdot 10^{-7} - 4.45 \cdot 10^{-5}$
$F_2(\Omega\tau)$	0.0148–0.096	0.013–0.069
M , sec ² ·deg ² ·cm ⁻¹	$4 \cdot 10^{-14}$	$4.9 \cdot 10^{-13}$
τ_1 , sec	$2.3 \cdot 10^{-6}$	$3.3 \cdot 10^{-6}$
$\Omega\tau$	$4.4 \cdot 10^5 - 313.6$	$8.5 \cdot 10^5 - 780$

* ξ_1, D_1 —data obtained by L. L. Chaikov in our mixtures on the apparatus of the Physics Institute, Acad. Sci., USSR.

tained from computations with formulas (5) – (7) in this same range of temperatures were significantly smaller than those observed experimentally and amounted to only 0.15 and 0.3% (see Tables I and II). The hypersonic absorption coefficient, calculated from the given formulas is much smaller than the measured value, because of the smallness of the quantity $F_1(\Omega\tau)$ (see Tables I and II).²⁾

It follows from the above that the cited theories do not describe the frequency dependence of the velocity of propagation and the absorption coefficient of sound in critical mixtures and a departure from the experimental values is observed in the hypersonic range of frequencies.¹⁶⁻¹⁹

We assume that for a complete description of the experimental results, over the entire range of acoustical frequencies in critical mixtures, it is necessary to take into account the relaxation mechanisms studied by Mandel'shtam and Leontovich.²⁰ These, as a consequence of the concentration fluctuations in the critical region, can strongly increase the bulk viscosity and thus affect the absorption and dispersion of the sound velocity.

In conclusion, the authors thank I. L. Fabelinskiĭ for attention to the work, useful advice, and a discussion of the results.

¹⁾The theory of Ref. 15 takes into account the dependence of T_{cr} on the pressure $\partial T_{cr}/\partial P$ and is therefore different from

the theory of Ref. 13.

²⁾The quantity L , which characterizes the contribution to the sound absorption from all the mechanisms except the mechanism described by the theories of Refs. 13 and 15, was not calculated theoretically and therefore could not be taken into account.

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