



UDC: 531.567:535

MANDELSTAM-BRILLUEN SCATTERING IN A STRETCHING SOLUTION

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Annotatsiya: Suyuqliklarda gipertovush tezligi va yutilishini o'rganishga mo'ljallangan ikki o'tishli Fabri-Pero interferometri asosida ishlovchi tajriba qurilmasining ishlash prinsipi batafsil yoritilgan, aniline-siklogeksan eritmasida kritik nuqta atrofida Mandelshtamm-Brillyuen komponentalarining siljishi va kengligi o'rganilgan, bir jinsli faza tomonidan t_k ga yaqinlashganda Mandelshtamm-Brillyuen komponentalarining siljishining sekin o'sishi, bir fazali tomonidan t_k ga yaqinlashganda esa Mandelshtamm-Brillyuen komponentalari kengligining sekin o'sishi kuzatiladi, olingan natijalar asosida kritik nuqta atrofida gipertovush chastotasida sezish mumkin bo'lgan relaksatsion jarayonlar mavjudligi taxmin qilingan.

Kalit so'zlar: Gipertovush, tovush tezlig va yutilishi, binary aralashmalar, dispersiya, kritik nuqta, yorug'likning sochilishi, Fabri-Pero interferometri, Mandelshtamm-Brillyuen komponentalari, qatlamlanuvchi eritmalar, relaksatsion jarayonlar, konsentrasiya fluktuasiyasi.

Аннотация: Подробно описано принцип работы экспериментальной установки по изучению скорости и поглощения гиперзвука в жидкостях на базе



двухпроходного интерферометра Фабри-Перо, измерено смещения и ширины компонент Мандельштама-Бриллюэна в растворе анилин-циклогексан в окрестности критической точки, при приближении к t_k со стороны однородной фазы обнаружено медленное увеличение смещения компонент Мандельштама-Бриллюэна, а при приближении к Δt со стороны однофазного состояния наблюдается медленный рост значений ширины компонент Мандельштама-Бриллюэна, по полученным результатам предположено существование релаксационных процессов осязаемых на гиперзвуковых частотах в окрестности критической точки.

Ключевые слова: Гиперзвук, скорость и поглощение звука, бинарные смеси, dispersiya, критическая точка, рассеяние света, интерферометр Фабри-Перо, компоненты Мандельштама-Бриллюэна, расслаивающиеся растворы, релаксационные процессы, флуктуация концентрации.

Abstract. The principle of operation of an experimental setup for studying the velocity and absorption of hypersound in liquids based on a two-pass Fabry-Perot interferometer is described in detail, the displacements and widths of the Mandelstam-Brillouin components in aniline-cyclohexane solution in the vicinity of the critical point are measured, and a slow increase is observed when approaching t_k from the side of the homogeneous phase the displacement of the Mandelstam-Brillouin components, and when approaching Δt from the side of the single-phase state, a slow increase in the widths of the Mandelstam-Brillouin components is observed, according to the results obtained, it was assumed that relaxation processes exist at hypersonic frequencies in the vicinity of the critical point.

Keywords: Hypersound, speed and absorption of sound, binary mixtures, dispersiya, critical point, light scattering, Fabry-Perot interferometer, Mandelstam-Brillouin components, stratified solutions, relaxation processes, concentration fluctuation.

Introduction. Ultrasonic measurements of absorption and dispersion of the speed of sound near the critical point of separation of liquid binary mixtures are a fruitful method for studying the kinetics of second-order phase transitions. The available experimental and theoretical materials mainly contain the results of anomalous absorption of ultrasonic waves. At the same time, there are no data in the literature on the absorption of the hypersonic range in general. Studies of Mandelstam-Brillouin scattering [MB], it seemed, should have provided a lot of information about physical processes near the critical point, just as the corresponding studies in pure liquids [1] provided valuable data on relaxation processes affecting the propagation of sound.

However, such studies in solutions are still little used when discussing various theoretical models of the Critical State because of certain experimental difficulties and large errors arising in the study of the scattering spectrum near the critical point. In most works, a low-contrast single-pass Fabry-Perot interferometer was used. Therefore, the light of a very intense central component arising from scattering by concentration fluctuations partially falls into the frequency range where the Mandelstam-Brillouin components are observed. This leads to a noticeable but poorly

controlled decrease in the measured values of their displacements, and, consequently, the corresponding, experimentally determined, values of the speed of sound.

The inaccuracy of the results obtained as a result of this using a low-contrast Fabry-Perot interferometer can explain the appearance of a report on the observation of a negative dispersion of the speed of sound near the critical point of separation of solutions [2]. Later measurements with a two-pass interferometer did not confirm this result [3]. When measuring the displacement of the Mandelstam-Brillouin components using a two-pass high-contrast Fabry-Perot interferometer, the central component due to light scattering by concentration fluctuations does not affect the position of the Mandelstam-Brillouin components. However, in work [3] performed with such an interferometer, only the speed of hypersound was measured and the width of the Mandelstam-Brillouin components and, consequently, the absorption coefficient of hypersound were not measured, which significantly reduces information on the nature of the processes affecting its propagation. This obviously also limits the possibility of using the experimental results to discuss physical processes near the critical point.

Experiment.

The present work assumes an attempt to fill, as far as possible, this gap. For this, similarly to [3,4,11-15], we used a high-contrast two-pass Fabry-Perot interferometer developed by us. The schematic diagram of the experimental setup is shown in Fig. 1.

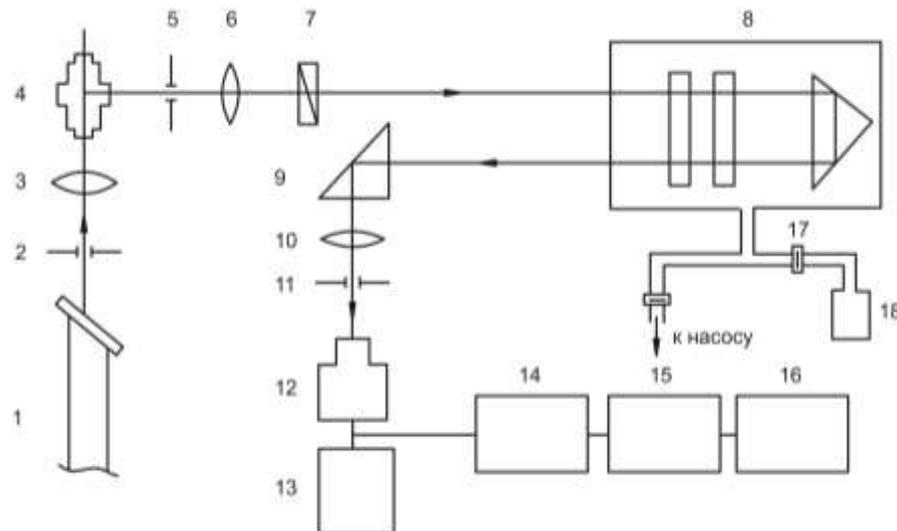


Fig. 1.

Schematic diagram of the experimental setup for recording the fine structure spectra of the Rayleigh line:

1- He-Ne laser; 2 - diaphragm; 3 - lens (120 mm); 4 - a vessel with a test liquid; 5 - diaphragm; 6 - collimator lens (210 mm); 7 - polarizer (Frank-Ritter prism); 8 - pressure chamber with a two-pass Fabry-Perot interferometer; 9 - rotary prism; 10 - camera lens (270 mm); 11 - aperture (0.25 mm); 12 - Photomultiplier tube (PMT)-79; 13 - PMT power supply unit; 14 - emitter follower; 15 - linear intensity meter; 16 - KSP-4 recorder; 17 - supersonic needle leakage valve; 18 - nitrogen cylinder.

It turned out that with our accuracy of setting the angles (± 0.20), the error in determining the displacement of the Mandelstam-Brillouin components for a



scattering angle of 90° did not exceed 1%. To reduce the influence of random errors, the spectra were recorded at least four times (four orders of the spectrogram), and the results of processing the spectra were averaged. An He-Ne laser (wavelength 632.8 nm, radiation power about 15 mW) was used as a source of exciting light (1). The laser beam was focused by a long-focus lens (3) into a cell (4). The scattering angle was set using a pentaprism (accuracy $\pm 0.2^\circ$). In the path of the scattered light, there was a Frank-Ritter prism (7), which made it possible to select the scattered light of the required polarization. The polarizer alignment accuracy ($\pm 0.5^\circ$) was quite satisfactory for the experiment. The scattering volume was at the focus of the objective (6), which formed a parallel beam of rays passing through the Frank-Ritter prism and then incident on the Fabry-Perot interferometer (8).

To ensure the linearity of the gas leak during scanning, we used a needle supersonic leak (17). To increase the linearity of scanning, a ballast volume was used. The gas (nitrogen) pressure at the inlet of the leak was 6-8 atm. Such a gas supply system allowed us to achieve that the nonlinearity of scanning at three orders of the interferogram was no more than 0.5%. After a two-pass interferometer, the scattered light, passing through a rotating prism (9), was collected in the focal plane of a camera lens (10) with a focal length of 270 mm. Aperture (11) is installed in the focal plane of the camera lens. The radius of the diaphragm was selected empirically, proceeding from the condition of the minimum broadening of the instrumental function. For example, in the case when an interferometer with a dispersion region of 0.417 cm was used, the diaphragm had a diameter of 0.25 mm.

The half-width of the instrumental function becomes minimum when the exit diaphragm is placed in the center of the interference pattern. The diaphragm was adjusted using two micrometric screws moving it in mutually perpendicular directions in the focal plane of the objective (10). A cooled photomultiplier PMT-79 operating in the photon counting mode was used as a photodetector in our setup.

The PMT cooling circuit is assembled on the basis of a semiconductor microcooler operating on the principle of the Peltier effect. Cooling of the PMT to -25°C was achieved within 1 hour. Upon cooling to this temperature, the number of dark pulses decreased from 100-150 pulses / s to 10-15 pulses / s, with the same photocathode sensitivity.

Pulses from the photomultiplier anode were fed to the input of the emitter follower (14). The emitter follower has a sufficiently high input impedance and low input capacitance, which, along with a low output impedance, is necessary for pulse transmission. Then the signal entered the input of the discriminator of the PI-4-1 linear analog intensimeter (15), which passed the pulses with the amplitude specified by the discriminator. At the output of the intensimeter, a voltage constant in sign appeared, the value of which was proportional to the number of pulses per second, which was then fed to the KSP-4 potentiometer (16), the recorder of which recorded the signal on a chart tape. The spectrum was scanned by changing the pressure in the body of the Fabry-Perot interferometer using a leak system 17, 18. The linearity of the leak [1%] by three orders of interference was ensured by using a needle leak 17. The Fabry-Perot interferometer used had a contrast ratio of $6 \cdot 10^4$ and a sharpness of 40.

The cuvette with the solution under study was placed in a specially made thermostat, the temperature in which was maintained with an accuracy of 0.05 ° C.

The discussion of the results.

The high value of contrast and sharpness in the interferometer used made it possible to measure the displacement Δv and the width δv of the Mandelstam-Brillouin components with high accuracy in a stratified aniline-cyclohexane solution near the critical point of separation and quite close to the critical point of separation in temperature.

The calculation of the hypersound speed was carried out according to the well-known expression

$$g = \frac{\Delta v_0 C}{2vn \sin \theta/2} \quad (1)$$

where Δv_0 , is determined from the displacement of the Matzdelaitama-Brillouin components Δv taking into account their finite width from the expression

$$[5] \Delta v = \Delta v \left(1 - \frac{\delta v^2}{2\Delta v} \right)^{-\frac{1}{2}} \quad (2)$$

In (1) and (2) n is the refractive index, C is the speed of light, v is the frequency of the exciting light, Δv is the displacement of the Mandelstam-Brillouin components obtained from the experiment.

The data on the width of the Mandelstam-Brillouin components made it possible to determine the hypersound absorption coefficient α from the relation

$$\alpha = \frac{\delta v_{ncm} \pi C}{V} 3$$

Where δv_{ncm} is the true width of the Mandelstam-Brillouin components

The measurement accuracy of Δv and δv is $\pm 0.5\%$ and $\pm 5\%$, respectively.

The results of measuring the displacement and width of the Mandelstam-Brillouin components in the critical solution of aniline-cyclohexane are shown in Fig. 2. The measurements were carried out over a wide temperature range. In the homogeneous phase, the spectrum of light scattered from the volume was recorded in the region coinciding with the delamination region as the temperature dropped below t_k .

In fig. 2 shows the results both for a homogeneous phase above the critical temperature $\Delta t > 0$ (Δ , \circ is, respectively, the displacement and width of the Mandelstam-Brillouin components), and for temperatures below $\Delta t < 0$ in both phases (\bullet , \blacktriangledown - corresponds to a phase saturated aniline; \blacksquare , \blacktriangle - phase saturated with cyclohexane) at a distance of 10 mm from the delamination boundary.

Using expressions (1) and (3), the values of the rate \mathfrak{g} and the absorption coefficient α were determined for an aniline-cyclohexane solution with a concentration of 0.44 ppm. aniline having a critical delamination temperature $t_k = 2.2 \pm 0.05$ ° C.

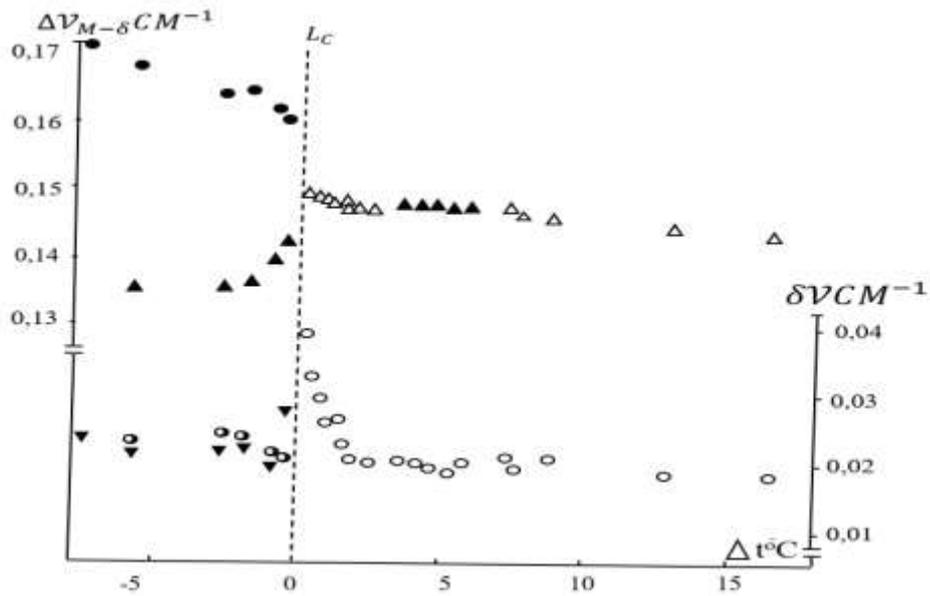


Fig 2.

In fig. 3 and 4 show typical spectra of scattered light near the critical temperature at and, respectively, obtained on a two-pass Fabry-Perot interferometer.

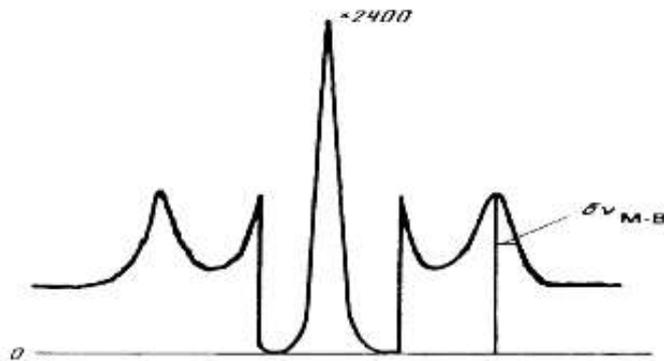


Fig 3.

It can be seen from Fig. 2 that, when approaching t_k from the side of the heterophase state, the difference in the values of the displacement Δv of the Mandelstam-Brillouin components for different phases decreases and tends to zero.

When approaching t_k from the side of the homogeneous phase to a slow increase in the displacement of the Mandelstam-Brillouin components is observed. From $\Delta t \sim 7^\circ \text{C}$ to $\Delta t \sim 2^\circ \text{C}$, within the experimental error, the shift of the Mandelstam-Brillouin components remains constant. At $\Delta t \sim 2^\circ \text{C}$, when approaching t_k , an increase in the mixing of the Mandelstam-Brillouin components is observed.

This figure also shows the temperature dependence of the width of the Mandelstam-Brillouin components. It can be seen from this dependence that for the width of the Mandelstam-Brillouin components, when approaching t_k from the side of the heterophase state ($t < t_k$) to $\Delta t = -0.4^\circ \text{C}$, no features are observed within the experimental errors.[11]

$t^\circ \text{C}$	$\Delta v \text{ sm}^{-1}$	$\delta v \text{ sm}^{-1}$	$\vartheta \text{ m/s}$	$\alpha 10^3 \text{ sm}^{-1}$
32,5	0,1497	0,0388	1361	27,04
32,8	0,1490	0,0329	1345	23,04

33,0	0,1487	0,0296	1342	20,78
33,3	0,1476	0,0263	1332	18,59
33,4	0,1482	0,0265	1338	18,65
33,8	0,1477	0,0232	1334	16,33
34,1	0,1476	0,0204	1333	14,44
34,8	0,1470	0,0197	1328	13,94
35,8	0,1474	0,0201	1332	14,09
36,3	0,1480	0,0200	1337	14,10
36,8	0,1471	0,0192	1329	13,63
37,4	0,1473	0,0156	1332	13,13
38,0	0,1475	0,0203	1334	14,36
39,5	0,1469	0,0208	1329	14,77
41,1	0,1448	0,0197	1310	14,19
45,1	0,1445	0,0188	1309	13,54
48,6	0,1419	0,0188	1287	13,76

It is also seen that when approaching Δt from the side of the single-phase state ($t > t_k$) to $\Delta t \sim 2^\circ \text{C}$, a slow increase in the widths of the Mandelstam-Brillouin components is observed, and at $\Delta t < 2^\circ \text{C}$, the width of the Mandelstam-Brillouin components increases very bistro. The acoustic properties of solutions with a critical separation point have been studied theoretically and experimentally for a long time and intensively. There are many theoretical works done to describe the experimental results obtained by measuring the velocity and absorption of ultrasound [6, 7, 8, 11-15] taking into account the interaction of sound waves with slowly absorbing concentration fluctuations. As is known, these theories describe quite satisfactorily the propagation of ultrasound near t_k . at frequencies not exceeding several tens of megahertz At high frequencies, such agreement of these theories with the experimental results could not be obtained [6, 7, 8 and 9, 12].

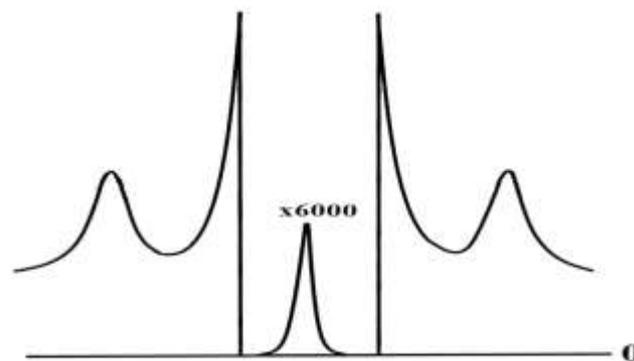


Fig 4.

Conclusion. Perhaps it should be assumed that these theories do not take into account the physical processes that significantly affect the nature of the propagation of high-frequency sound [109 Hz and more]. Indeed, the results of the calculated absorption values from the Mandelstam-Brillouin scattered light spectra show an anomalous strong absorption of hypersound at a temperature $\Delta t_k < 2^\circ \text{C}$ and its growth as the critical separation point is approached. The preliminary estimate of the dispersion of the speed of sound in the aniline-cyclohexane system near the critical temperature is 1.5%, which is beyond the experimental error. The results obtained allowed us to assume significant relaxation processes at hypersonic frequencies in the vicinity of the critical point, which differ from the mechanism noted in [6, 7, 8, 11].

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