TEMPERATURE DEPENDENCE OF THE SPEED OF PROPAGATION OF GEPER SOUNDS IN AQUEOUS SOLUTIONS

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ABSTRACT

A study was made of the nature of hypersound propagation in an aqueous solution of 4-methylpyridine with a concentration of 0.06 m.f. by method of Mandelshtam-Brillouin light scattering spectroscopy. It is shown that a positive sound velocity dispersion is observed in the studied solution in the frequency range from 5.4 MHz to 4.8 GHz, which is due to the process of bulk viscosity relaxation. In a narrow frequency range from 4.8 to 6.1 GHz, a negative dispersion of the hypersonic velocity has been experimentally revealed. The negative dispersion is associated not with the relaxation process, but with the spatially inhomogeneous structure of the solution on scales comparable to the hypersonic wavelength

Keywords: *Hypersound, Mandelshtam-Brillouin, Ultra-and hyperacoustic parameters, 4-methylpyridine, ultrasonic speed, Fabry-Perot, Cylindrical glass, calculation, Mandelstam-Leontovich relaxation theory, Navier-Stokes.*

INTRODUCTION

The propagation of sound velocity is one of the main issues in Condensed Matter Physics, and has been studied repeatedly, both theoretically and experimentally, a problem studied with great interest. 0.06-0.08 mol share (m.q), at normal atmospheric pressure and temperature $t_0 \approx 70\ ^0C$, aqueous solutions of pyridine methyl spert have minimal thermodynamic stability (called"single point. State diagram in the coordinates "temperature-concentration". Single-point systems have been little studied experimentally and are therefore an interesting and important object for high-frequency acoustic studies of flocculation processes and structural changes located close to a point. In our laboratory, we worked on the study of the Ultra-and hyperacoustic parameters of one of the objects belonging to this solution class (an aqueous solution of 3-methylpyridine). In the temperature range from 20 to 80 ^0C, the temperature dependence of the ultra-and hyperplussive speed made it clear that there is a positive dispersion of the speed of Sound near a separate point. Analyzing the experimental results obtained for this solution at a frequency of about 2.6 GHz, the basis is to assume that negative velocity dispersion can also exist near a single point (i.e., a decrease in velocity with an increase in sound frequency) in the narrow hyperplane frequency range. To verify this assumption, we have a share of

0.06 mol (m.q),we studied the temperature dependence of the speed of sound in a concentrated 4-methylpyridine (4MP) aqueous solution. this corresponds to a single point concentration. An experimental study of the fine structure spectra of the relay light scattering line at a wide range of temperatures and concentrations in aqueous solutions of 4-methylpyridine was carried out by US. The results of the works [1-4] made it possible to reveal some interesting and important features of the processes of structure formation in the studied systems, which were later confirmed in the work of other authors (see, for example, [5]) The conclusions of some works [1-4] are involved in the discussion of the results obtained in this process of work

DISCUSSION AND RESULTS

Compared to our previous studies, the range of sound frequencies in this study was significantly expanded, from 5 MHz to 6.1 GHz.The ultrasonic speed was measured by an acousto-optical method based on the phenomenon of diffraction of laser radiation with ultrasonic waves.

The hyperplane velocity was calculated from the frequency shift of the fine structure components of the relay light scattering line (Mandelshtam-Brillouin component). Studies were conducted at 20 and 80 ^{0}C light scattering angles corresponding to sound frequencies of ~4.8 and ~6.1 GHz. The excited radiation was a He-Ne laser (radiation wavelength 632.8 nm, power 15 MW)

Scattered light spectra were recorded using an experimental device with a two-pass Fabry-Perot interferometer. The value of the interference wave was 5×10^4, the accuracy of the edges was ~40. Mandelstam-Brillouin (KMB) component studies $\Delta\nu$ was placed in a cuvette thermostat with a sample of no less than an e-tube, whose circuit allowed the temperature to be stabilized with an accuracy of less than ±0,05 °C in the tube. Error measuring frequency shift did not exceed 0.5%

The optically pure components of the solution were obtained by triple dispersion. Solution samples Mandelstam-Brilluene from light scattering spectra the difficulties of the hyperplane experiment in measuring velocity require special attention. When using a cylindrical resanotor, the effect of cylindrical lenses appears. As a result, the collimator lens of the installation receives light scattered not only at a certain angle, but also within larger and smaller angles than indicated. In our experiments, an ampoule with a test solution was placed in an octahedral cylinder glued together from optical prisms to eliminate the effects of cylindrical lenses

Cylindrical glass is sealed into cubes under pressure below atmospheric pressure. An immersion fluid was poured into the space between the ampoule and the cylinder walls, the refractive index of which was close to that of the cylinder. When we studied this case, we determined the angle of propagation. Error setting the angle of spread 0.2 ^{0} does not exceed.

The temperature dependencies of the Mandelstam-Brillouin (KMB) components of the beat of a clock are 20^{0} and 80^{0}. studied for scattering angles. the values of the hyperplane velocity defined from the Momenta are shown in Figure 1. The same picture shows the results of measuring the speed of ultrasound. As can be seen from the picture, the temperature movement of the speed is basically the same for ultrasound and hyperplastic frequencies. As the temperature of the solution increases, the speed of sound monotonically decreases, but the dependence of V(t) is not linear, but dv/dt is characterized by an increase in the temperature coefficient of speed (at absolute value). The frequency dependence of the sound speed turns out to be much more complicated. Figure 1 shows that an increase in sound frequency from 5.4 MHz to 4.8 GHz is accompanied by an increase in sound speed by about 1.05 times, and this value remains virtually unchanged throughout the time studied (temperature range). An increase in sound frequency to 4.8 GHz - 6.1 GHz leads to a decrease in sound speed by about 1.03 times, and this value practically does not change even with the temperature of the solution.

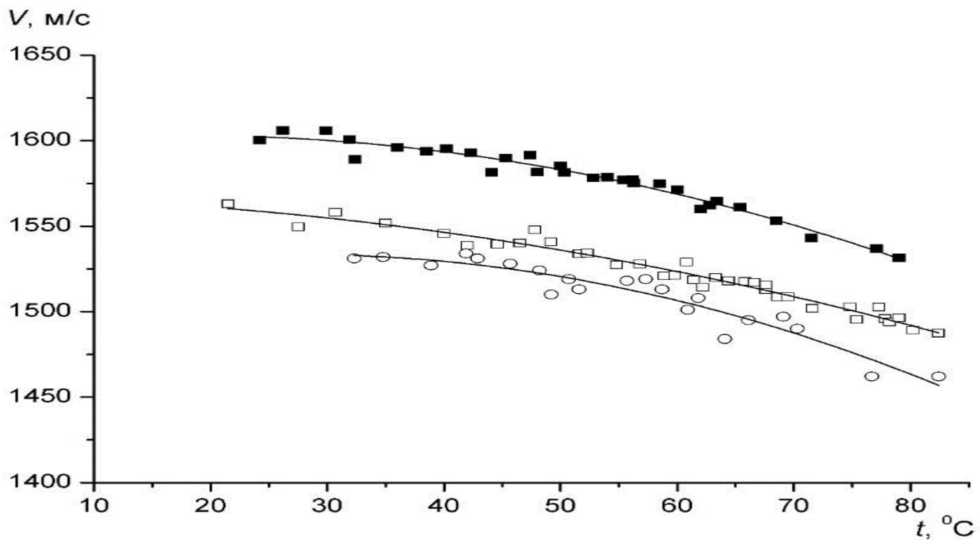

Figure 1. 4 MP (0.06 mol share m.q) temperature dependence of the speed of sound in aqueous solution 6.1 GHz,– ~4.8 GHz,– 5.4 MHz.

Table. Positive sound speed temperature dependence graph in the frequency range from 5.4 MHz to 4.8 GHz

t				80 ^{0}C		
f	5.4 MHz	3.4 GHz	4.8 GHz	5.4 MHz	3.4 GHz	4.8 GHz
V, м/с	1540	1590	1609	1534	1583	1602

About calculation		
V_0, м/с	1540	1534
V_∞, м/с	1609	1602
□×10^11^ c	9.3	7.7

1.Thus, the results of our study of sound speed at frequencies from 5.4 MHz to 6.1 GHz show that: 1) there is a positive dispersion of sound speed in the frequency range from 5.4 MHz to 4.8 GHz; 2) a negative speed dispersion occurs in the narrow frequency range from 4.8 to 6.1 GHz; 3) the value of the dispersion (both positive and negative) does not depend in 4-methylpyridine (0.06 m.q) in aqueous solution, in the frequency range from 5.4 MHz to 4.8 GHz in the temperature range from 20 ^{0}C to 80 ^{0}C, there is a positive dispersion of sound speed and a negative dispersion of hyperplasia.

The analysis of the frequency dependence of the sound velocity was carried out based on the following experimental results: 1) in the frequency range from 5.4 MHz to 4.8 GHz, the speed behavior corresponds to a positive dispersion due to the relaxation process of volumetric viscosity; 2) the hypersound speed with a frequency of 6.1 GHz is lower as a result of the existence of a negative dispersion

The value of the positive dispersion of the speed of sound in the frequency range from 5.4 MHz to 4.8 GHz was calculated using the formula:

$$\frac{\Delta V}{V} = \frac{V_1 - V_2}{V} \quad (1)$$

where V_1 and V_2 are the speeds of hypersound (f=4.8 GHz) and ultrasound (f=5.4 MHz), respectively, and V=(V_1+V_2)/2is the average speed in this frequency range [1].

The value of the negative variance was calculated using the same formula. The sound velocity values at frequencies 6.1 and 4.8 GHz, respectively, were used as V_1 and V_2. The calculated temperature dependences of the velocity dispersion are shown in

As can be seen from the value of the positive dispersion is approximately 4% and slightly depends on the temperature of the solution. The value of the negative dispersion is approximately 3% and also practically does not depend on the temperature of the solution. The obtained dispersion values significantly exceed the error of measuring the speed of sound by the displacement of the Mandelstam-Brillouin components.

The parameters of the relaxation process of volumetric viscosity, leading to a positive dispersion of the speed of sound, were calculated in accordance with the formulas of the Mandelstam-Leontovich relaxation theory [6]. The calculation is based on the formula

$$V^2 = \frac{\Omega^2\tau^2}{1+\Omega^2\tau^2}\left(V_\infty^2 - V_0^2\right) + V_0^2 \quad (2)$$

Here *W*, *t*, V_0 and V_∞ are the sound frequency, acoustic relaxation time, and speed limits, respectively

Using the values of the velocities V for the frequencies 5.4 MHz, 2.6 GHz (obtained earlier) and 4.8 GHz, we compiled 3 equations in accordance with the formula (2) and the values of the values *t*, V_0 and V_∞ were obtained by sequential exclusion of unknowns (an example of the calculation results is given in the table). Following these calculations, the following conclusions can be drawn:

1. in an aqueous solution of 4MP (0.06 m.q) there is a relaxation process, the acoustic relaxation time of which varies from t=9.3×10^{-11} s to t=7.7×10^{-11} s with a temperature change from 30 ^{0}C to 50 ^{0}C;

2. The values of V_0 and V_∞ depend on temperature, and the relaxation time decreases with increasing temperature;

3. The values of ultrasound and hypersound speeds at 5.4 MHz and 4.8 GHz are close to the limiting speeds of V_0 and V_0. (the calculated relaxation curves are shown in Fig.3.3)

Table. Parameters of the relaxation process of volumetric viscosity, which causes a positive dispersion of the speed of sound in the frequency range from 5.4 MHz to 4.8 GHz.

t	30 ^{0}C			50 ^{0}C		
f	5.4 MHz	2.6 GHz	4.8 GHz	5.4 MHz	2.6 GHz	4.8 GHz
V, м/с	1533	1586	1600	1520	1566	1583
Calculation results						
V_0, м/с	1533			1520		
V_∞, м/с	1608			1594		
t×10^{11}, с	9.3			7.7		

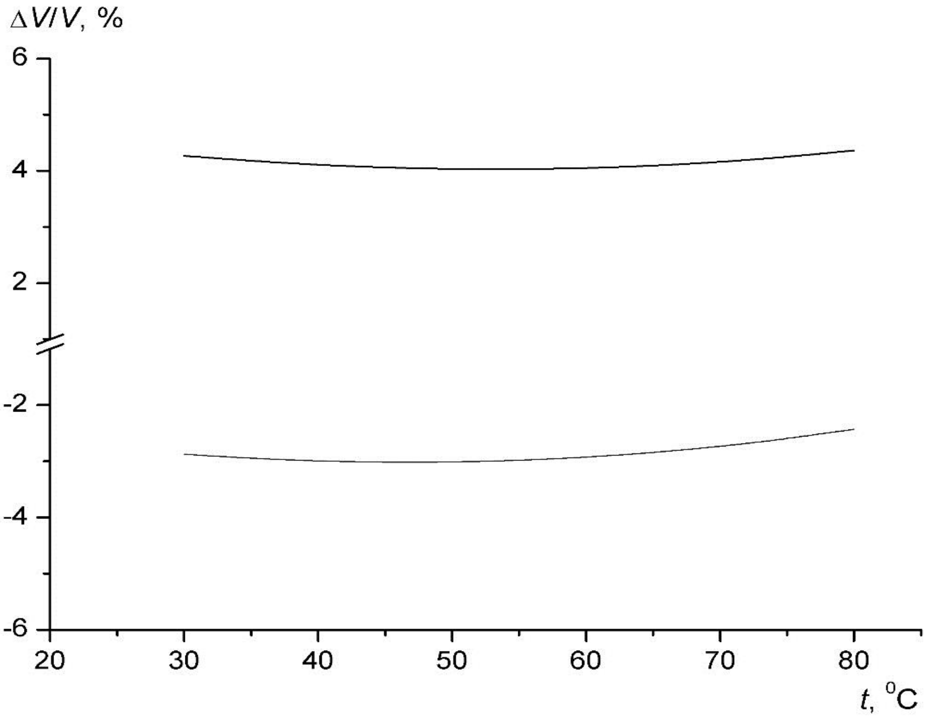

Fig.2. Temperature dependences of the sound velocity dispersion in the frequency ranges from 5.4 MHz to 4.8 GHz (upper curve) and in the frequency range from 4.8 to 6.1 GHz (lower curve)

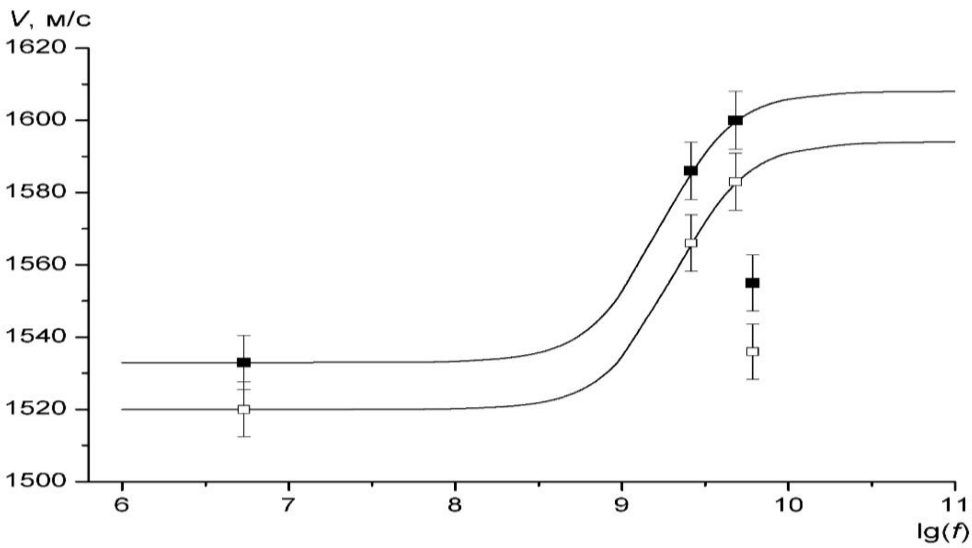

Fig.3. Relaxation curves describing the frequency dependence of the speed of sound due to relaxation of the volumetric viscosity in a 4MP-water solution:

30 ^{0}C-50 ^{0}C.

Consider the possibility of negative variance. There are no physical reasons for observing a negative dispersion of the speed of sound in ordinary liquids and solutions. Another thing is in associated liquid media. Vladimirsky [7] and Ginzburg [8] pointed out that, in principle, a negative dispersion of the speed of sound in liquids can be observed. In [7], the dispersion of the speed of sound is considered, which is not associated with relaxation, but is due to intermolecular interaction. In [8], the Navier-Stokes equation, presented in the second approximation, is investigated, and it is assumed that the absorption is very small, that is, the effect of viscosities on dispersion is not essentially considered, but other causes of dispersion associated with the features of the spatial structure of the medium under consideration, and, consequently, with the spatial dispersion of the speed of sound, are identified. According to [8], the amount of variance can be estimated by the expression

$$\frac{\Delta V}{V} = \frac{2\pi^2 f^*}{V^2 \Lambda^2} \quad (3)$$

where L is the wavelength of the sound. The sign of sound dispersion in (3.) will be determined by the sign f^* and the latter depends on the nature of intermolecular interactions. For example, in a molecular aggregate with attractive forces acting between the particles, i.e. $f^*<0$, the dispersion according to (3.) will be negative [8]. If we assume that что $f^*\sim V^2 r^2$, where r is the distance at which the molecular interaction is significant (correlation radius or diameter of the associated aggregate), then (3) can be rewritten as

$$\frac{\Delta V}{V} \approx 2\pi^2 \left(\frac{r}{\Lambda}\right)^2 \quad (4)$$

It is known [17-19] that aqueous solutions of methylpyridines have a rather complex phase diagram in the three-dimensional space "temperature-concentration-pressure". In the temperature range below and above the singular point, these solutions are associated due to the formation of

hydrogen bonds of various intermolecular character. Thus, the conditions under which the existence of a negative dispersion is allowed in an aqueous solution of 4MP are

The diameter of the associated aggregate can be estimated using the formula (4) using experimentally observed negative variance. Our estimates of the size of the associated aggregate turned out to be on the order of 100 Å, which is in satisfactory agreement with the sizes of aggregates observed in other similar solutions by other methods. For example, in [9], based on measurements of the concentration dependences of the spectral intensity of the Rayleigh line and calculations of the correlation functions of the electric field of scattered light, the cluster sizes in aqueous solutions of tertiary butyl alcohol and 2-butoxyethanol at a temperature of 40 ^{0}C and in the range of nonelectrolyte concentrations from 0.02 to 0.2 mole fractions are estimated at about 50 Å

Negative dispersion during the transition from ultrasound to hypersonic was previously observed by Zaitsev in aqueous electrolyte solutions [10]. In [10], the negative variance was estimated at 1.46%, and the size of the associates according to the formula (4) was about 90 Å. The existence of a negative dispersion was associated with a strong association of aqueous electrolyte solutions.

CONCLUSION

In an aqueous solution of 4-methylpyridine (0.06 ppm) in the temperature range from 20 0C to 80 0C, there is a positive dispersion of the sound velocity in the frequency range from 5.4 MHz to 4.8 GHz and a negative dispersion of the hypersound velocity in the range from 4.8 to 6.1 GHz. Moreover, the positive variance, according to the expression (1), is characterized by the fact that the higher the frequency of sound, the higher the speed of sound corresponding to this frequency. Negative dispersion, according to expression (2), is characterized by the fact that with increasing frequency, the speed of hypersound decreases. These two processes, which are based on completely different dispersion mechanisms, lead to the experimentally observed complex dependence of velocity on frequency (Fig.13). Although the effect of decreasing velocity with frequency is quite noticeable, it should be borne in mind that the formula (3) is derived without taking into account the behavior of viscosity and thermal conductivity and, accordingly, does not take into account dispersion of the relaxation type.

The mechanism discussed in [8] is not the only possible one. There is a generally accepted idea of the structure of aqueous solutions of nonelectrolytes as a continuous grid of hydrogen bonds [10]. An aqueous solution of 4MP is capable of forming clathrate-like structures and thereby contribute to strengthening the structure of the solution, i.e. strengthening the network of hydrogen bonds. This is also evidenced by studies of acoustic absorption in aqueous solutions of nonelectrolytes of small and medium concentrations [11], which led the authors [11] to the conclusion that hydrated molecules of solute form a clathrate-like structure. The presence of nonelectrolyte molecules in the clathrate structure can be considered as point defects that can form dislocation lines in the continuum of the hydrogen bond grid with a lifetime corresponding to the lifetime of hydrogen bonds. Sound propagation in the presence of dislocations can be accompanied by a dispersion of the resonant type of sound velocity. The interaction of sound with these inhomogeneities occurs when the frequency of sound is close to the resonant frequency of dislocation motion [11]. In this case, both positive and negative dispersion of the speed of sound can be observed.

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List of used Internet resources

1. www.ufn.ru – Сайт журнала «*Успехи физических наук*»
2. www.quantum-electron.ru – Сайт журнала «*Квантовая электроника*»
3. www.maik.ru – Портал издательства научной литературы МАИК, содержание журналов «*Журнал экспериментальной и теоретической физики*», «*Акустический журнал*», «*Оптика и спектроскопия*», «*Physics of wave phenomena*»