LIGHT SCATTERING AND PROPAGATION OF ELASTIC WAVES IN SATURATED WATER SOLUTION OF POTASSIUM CHLORIDE RESEARCH

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The analysis of potassium chloride solubility in water showed, that in saturated solution near 22 °C there occurs second order phase transition, which is accompanied with strongly developed fluctuations concentration. Therefore, in this conditions there should be observed anomalous characteristics of light scattering and propagation of elastic waves. With the method of low-frequency light scattering there was experimentally researched the temperature dependence of speed and absorption coefficient of longitudinal hypersound with frequency about 5 megahertz, and also the correlation of integral intensities in Rayleigh triplet's components. The temperature of solution was changing in the interval from 18,6 to 27,4 °C. Dependence curves of absorption coefficient and Landau-Plazchek relation on temperature resembled λ – curves within second order phase transitions. Speed of hypersound and ultrasound at 5 megahertz frequency increased monotonously with temperature coefficient of 2,2 m/(sec·degree). Expected dispersion of sound speed – positive or negative – wasn't revealed in work.

Keywords: light scattering, hypersound, fluctuations, phase transition

Phase transition has been attracting attention of researchers for long time because of their effects of many anomalies. Especially interesting are considered to be second order phase transitions (SOPT) and due to isomorphism they are convenient to be researched at binary liquid mixes because of low critical temperature. Most often there were researched mixes of organic liquids [4]. The aim of this work is experimental research of light scattering and ultra- and hypersound spreading at SOPT point's neighborhood in saturated water solution of potassium chloride.

Initial opinion that SOPT appears in it was formed on the base of *KCl* solubility's analysis. Curve of solubility has break in the neighborhood of $t = 22–27\,°C$, derivative of concentration dc/dt changes unevenly. In work [5] it was showed that dc/dt was connected firstly to fluctuations concentration's kinetics $\overline{(\Delta c)^2}$. They also define the intensity of central component of Rayleigh triplet in the scattered light's spectrum, and the behavior of acoustic waves' spreading depends on them [4].

Molecular light spreading in every clear liquid is owing to optical heterogeneity, which is aroused by isobaric and adiabatic density fluctuations, and concentration fluctuations in solution. Because of different type of their dynamics there is observed a Rayleigh triplet in the scattered light's spectrum – central line and two moved Mandelstam – Brillouin components (MBC). MBC occurs as a result of scattering at adiabatic fluctuations of density, central component occurs at isobaric fluctuations of density and at fluctuation of concentration. Appeared adiabatic fluctuations of density resolve as elastic waves with and give MBC in spectrum. They are moved in frequency to quantity:

$$\Delta\nu = \pm\frac{2n\upsilon}{\lambda c}\sin\left(\frac{\theta}{2}\right). \qquad (1)$$

Here n – indicator of liquid refraction; λ – wave length of exciting light; c – speed of light; θ – scattering angle.

The width of MBC Γ is defined with absorption coefficient α of elastic waves:

$$\Gamma = \frac{\alpha\upsilon}{\pi c}. \qquad (2)$$

Ration of integral intensities of central component to two MBS (Landau-Plazchek ratio (LP):

$$\frac{I_c}{2J_{MB}} = \frac{I_p + I_c}{I_s}, \qquad (3)$$

where I_p, I_s and I_c – integral components of light scattered at isobaric, adiabatic fluctuations of density and fluctuations of concentration.

Thereby, from the MBS width move one can define the speed and coefficient of elastic waves absorption, which get to hypersound diapason at 5 GHz frequency. Out of LP ratio one can draw a conclusion about the fluctuations of concentration dynamics $\overline{(\Delta c)^2}$, because $I_c \sim \overline{(\Delta c)^2}$.

In this work we have researched the low-frequency spectrum of scattered light in saturated water solution of potassium chloride in the range of temperatures from 18,6 to 27,4 °C, where, apparently, phase transition of higher order exists and where deeply changing fluctuations of concentration should reflect in the spectrum.

Experimental facility included single-frequency laser, producing light at wave length of $\lambda = 632{,}8$ nm, scanned interferometer of Fabry-Perot and cooled photomultiplier, working as photon counter [2, 3].

To define the speed of hypersound with formula (1) one have to know the indicator of refraction of scattering medium at corresponding temperature and wave length. It was measured at refractometer with the use of the same laser accurate to five decimal digits. Total error of hypersound's speed definition was 0,5 %, the coefficient of hypersound absorption and Landau-Plazchek ratio were found out 4 % accurate.

Temperature dependence of LP ratio is shown at Fig. 1. It's obvious that to 22,2 °C it increase, then in the narrow range from 22,2 to 23,5 °C it dramatically decreases and to 27,4 °C stays practically invariable. Thereby, LP ratio is anomalous just in that narrow temperature range, where it has jump of KCl solubility's derivative by temperature. It confirms the suggestion that both quantities in solution $J_c/2J_{MB}$ and dc/dt are connected with one reason – nonmonotonic change of fluctuation of concentration.

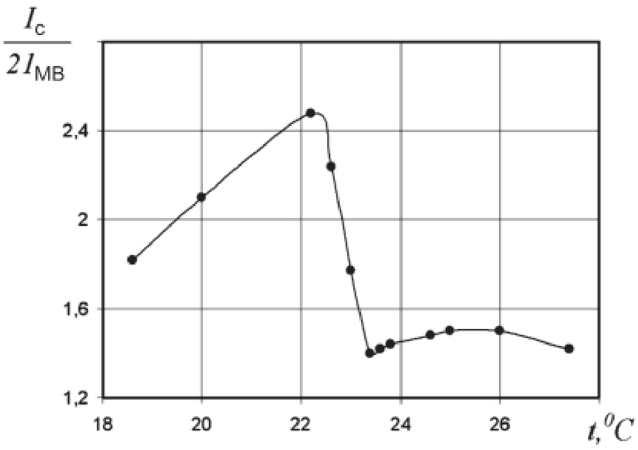

Fig. 1. Landau – Plazchek ratio at different temperatures

Change of medium size of fluctuations always leads to anomalous behavior of medium's acoustic characteristics [4]. At Fig. 2 there showed the results of hypersound absorption's coefficient research. As LP ratio, the coefficient of absorption reaches its maximum at 22,2 °C temperature and decreases dramatically both ways from this point. The type of dependence of α and $J_c/2J_{MB}$ on t resembles λ – the curve of absorption at phase transition of higher order, for example, helium I to helium II.

But at temperature dependence of υ_{GZ} hypersound waves' speed, defined out of MBC move, no features were revealed (Table 1). With solution's heating the speed of hypersound increases monotonously, what is usually observed at water solutions of electrolytes [1]. Within temperature increase from 18,6 to 27,4 °C the speed of hypersound changes linearly from 1670 to 1692 m/sec, that means that temperature coefficient of speed $\beta = d\upsilon/dt$ is 2,2 m/(sec·degree), that is less that in pure water, where $\beta = 2{,}9$ m/(sec·degree).

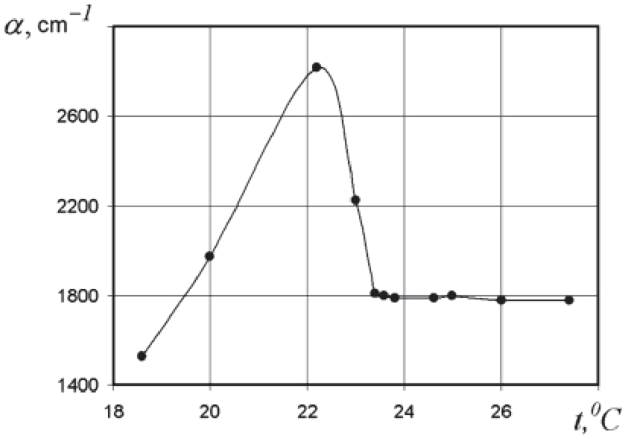

Fig. 2. Temperature dependence of hypersonic absorption coefficient

Table 1

Temperature dependence of υ_{GZ} speed with 5 GHz frequency

t, °C	18,6	20,0	22,2	23,0	23,4	23,6	23,8	24,6	25,0	26,0	27,4
υ_{GZ}, m/sec	1670	1675	1678	1680	1681	1682	1682	1684	1684	1687	1692

For comparison we have researched the temperature dependence of υ_{UZ} hypersound speed with 5 MHz frequency. It was changing with echo-impulse method accurate to 0,15 %. Quantities υ_{UZ} and υ_{GZ} within the error limits agree, that means that both positive and negative dispersion of sound speed, in spite of difference in their frequencies to thousand times (Table 2).

Table 2

υ_{UZ} hypersound speed with 5 MHz at different temperatures

t, °C	20,6	21,6	22,4	23,2	24,0	24,6	27,4	28,8	29,6
υ_{UZ}, m/sec	1675	1678	1679	1681	1683	1687	1691	1693	1695

Usually in phase transition of higher order range, the diagram of υ_{GZ} dependence on t has a braek, there is observed comparatively big positive dispersion ($\upsilon_{GZ} > \upsilon_{UZ} >$) [4]. In our solution positive dispersion, apparently, is veiled by negative one ($\upsilon_{GZ} > \upsilon_{UZ} >$), which was revealed in the water solution of electrolytes [1]. Possibly, far from phase transition of higher order point the phenomenon of negative dispersion will reveal at higher temperatures, as it in [1].

How one can imagine molecular mechanism of the experimental process? In the solution far from critical range, as it's known [5], there exists the short-range order of particles' disposition, which is characterized with average coordinating number z. If z changes in narrow range of temperatures, then quantity of connections between molecules of dissolvent and particles of dissolved substance also changes. It's analogous to phase transition of higher order: regions of solution with one coordinating number pass to regions with other z. In potassium chloride solution the temperature rise and KCl concentration at 22 °C leads to z rise, and in the range of 22–23,5 °C, z dramatically decreases. Tend to chaotic condition at particles' spreading means the rise of fluctuations. Because of medium's heterogeneity there appear features of light scattering and elastic waves' absorption.

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