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Issue Article

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DISPERSION OF THE SPEED OF SOUND IN WATER-ORGANIC SOLUTIONS

Abstract: *Concentration and temperature studies of spontaneous Mandelstam-Brillouin scattering of light in aqueous solutions of g-picoline showed the manifestation of negative dispersion of hypersound velocity at concentrations C J 0.2 mole fractions of g-picoline. The temperature and concentration dependence of the negative dispersion is associated with the process of solution structurization, which reaches its optimum value at a concentration of C = 0.06 mole fractions of g-picoline.*

Key words: *dispersion, relaxation of bulk viscosity, hypersound, relaxation time, water-organic solution.*

Language: *English*

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Introduction

The work is devoted to the description of an experiment on measuring the temperature dependences of the hypersound velocity at different frequencies in water-organic solutions with singular points. Interest in the study of such solutions is due to the following. As is known (see, for example, monograph [1]), there is a class of water-organic solutions in which an anomalously high scattering peak unrelated to molecular scattering is observed in a certain fairly narrow range of concentrations. The physical nature of this peak is currently being actively discussed (see, for example, [2] and references cited in this work). It is believed that the main reason for the appearance of this peak is the appearance of a supramolecular structure in a certain range of temperatures and concentrations. It is necessary to mention the previous works [3 - 6], in which it has been shown that stable liquid molecular complexes with sizes of 100 - 200 nm spontaneously arise in

aqueous solutions of polar organics, the density of which differs from the density of the surrounding liquid. It has been shown in [7 - 12] that anomalous light scattering is associated with the formation of a nanodroplet phase in water-organic mixtures.

In this work, a mixture of "water - g-picoline" is studied. Interest in this particular mixture is due to the fact that in this solution in a certain concentration range there is an abnormally high light scattering peak at a content of g-picoline » 0.06 mole fractions (ppm), see Fig. III.14 in the monograph [1] and comments on this figure. At the same time, in accordance with the literature data (see, for example, [11]), g-picoline has infinite solubility in water, i.e. the mixture "g-picoline - water" does not separate at any concentration in the entire temperature range at which the liquid mixture exists. The refractive index of g-picoline at a wavelength of l = 589.3 nm is 1.5, so it should be expected that in the concentration range for which an anomalous light scattering peak is recorded, a nanodroplet phase enriched in g-picoline can be observed, and the phase contrast, in this case, will be high enough to study these nanodroplets with a phase microscope, see [7 - 10]; we are currently experimenting with. In addition, in [12 - 15] it was shown that the Landau-Placek relation $\left(\frac{I_\omega}{I_{\omega-\delta\omega}+I_{\omega+\delta\omega}}\right)$, where I_w - unbiased component intensity, $I_{w\pm dw}$ - the intensity of the displaced components increases with decreasing solution concentration over a wide temperature range. In [16], it was suggested that the increase in $\left(\frac{I_\omega}{I_{\omega-\delta\omega}+I_{\omega+\delta\omega}}\right)$ is due to an increase in the isothermal compressibility b_T with an increase in the water content. This, in turn, may be due to the appearance of stable supramolecular structures (nuclei of the nanodroplet phase) with a decrease in the concentration of g-picoline; this phase, in accordance with the model presented in [7 - 10], causes anomalous scattering in an aqueous solution of g-picoline at its concentration of 0.06 ppm. in solution; henceforth, we will call this concentration the critical concentration. This paper presents the results of a study of the frequency dispersion of hypersound velocity in aqueous solutions of g-picoline in a wide range of temperatures and concentrations; special attention is paid to the concentration region near the critical one.

Experiment.

The experimental setup is described in detail in [17]. The spectra of polarized light scattering were studied by using a two-pass Fabry-Perot interferometer. The light scattering angle was ~90° and 135°. The error in setting the angle did not exceed 0.2°. The dispersion region of the interferometer was б0.625см$^{-1}$ and б0.417см$^{-1}$с. The interference contrast of the pattern reached 4Ч10^5, the sharpness of the interference maxima was 35. The radiation source was a He-Ne laser with a wavelength of 632.8 nm and a power of 115 mW. The shift of the Mandelstam - Brillouin components Dw was measured with an error not exceeding 0.5%. The hypersound velocity v was calculated using the formula:

$$v = \frac{\Delta\omega \cdot c}{\omega_0 2n \sin\frac{\theta}{2}}, \qquad (1)$$

where c– is the speed of light, w_0 – is the frequency of the exciting light, n – is the refractive index, q – is the scattering angle.

Experimental results and discussion.

On Fig. 1 (a) – (f) panels show the temperature dependences of the hypersound velocity v for frequencies of 6.2 GHz and 4.5 GHz for pure g-picoline (panel (a)) and for concentrations of g-picoline C = 0.4 ppm .(panel (b)); C = 0.2 ppm (panel (c)); C = 0.1 ppm (panel (d)); C = 0.06 ppm (panel (e)) and C = 0.05 ppm. (panel (f)). Figure 2 shows the dependence of the area difference DS under the curves for frequencies of 6.2 GHz and 4.5 GHz on the concentration C of the solution.

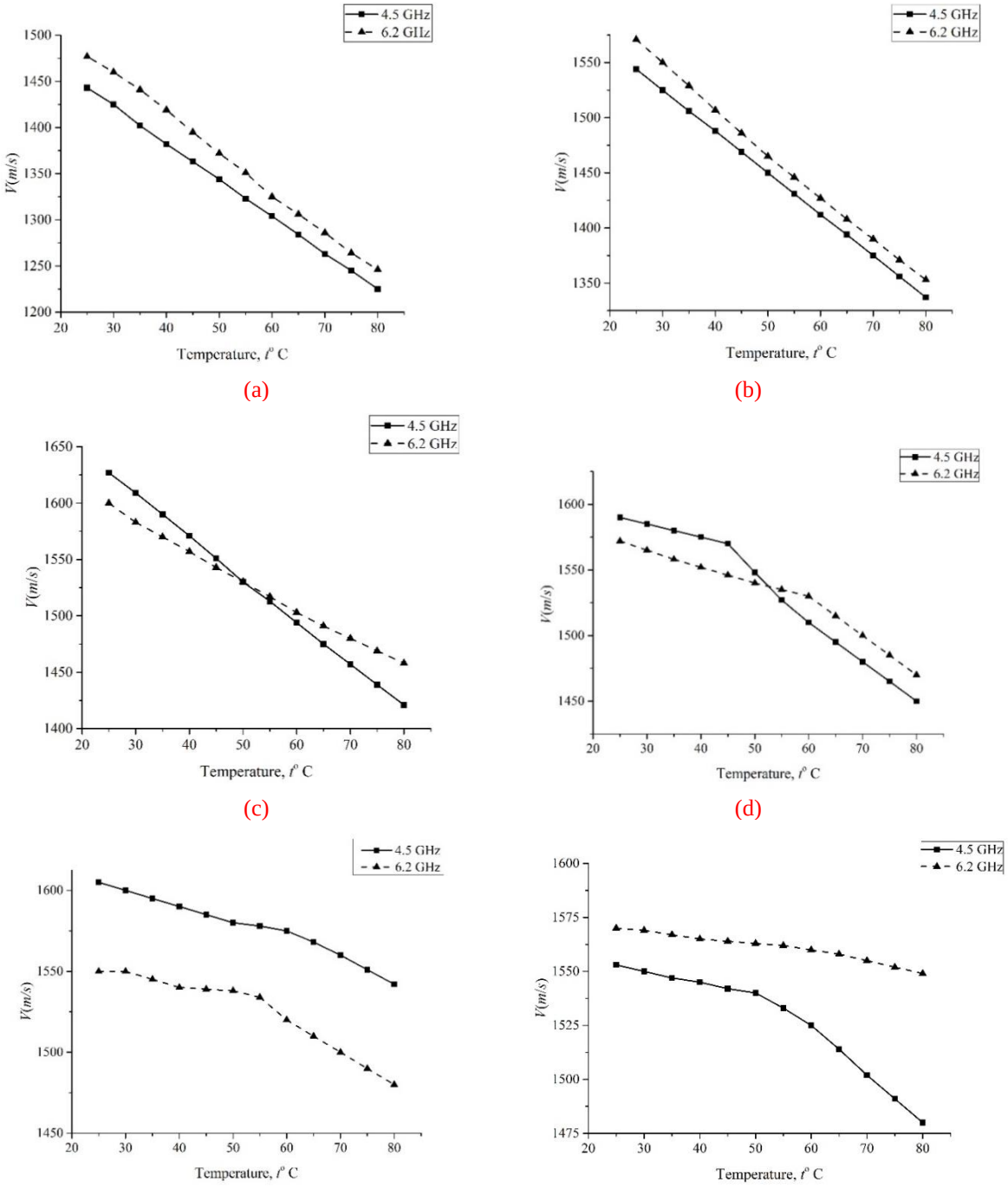

Fig. 1. Dependences of the velocity *V* of hypersound at frequencies 4.5 and 6.2. GHz on temperature for pure g-picoline (a), mixture "g picoline - water" at a concentration of g-picoline 0.4 mole fractions (b), mixture "g-picoline - water" at a concentration of g- picoline 0.2 mole fractions (c), mixtures "g-picoline - water" at a concentration of g-picoline 0.1 mole fractions (d), mixtures "g- picoline - water" at a concentration of g-picoline 0.06 mole fractions (e), mixtures "g-picoline - water" at a concentration of g- picoline 0.05 mole fraction (f).

As can be seen from the figure (panels (a) and (b)), the nature of the temperature dependence at the indicated frequencies for pure g-picoline and 0.4 ppm is approximately the same for both frequencies. Differences appear when the concentration of g-picoline is C = 0.2 ppm. and 0.1 ppm (panels (c) and (d)). It can be seen that at $t > 50°$ C, there is a positive and at $t < 50°$ C, a negative hypersonic speed dispersion in the frequency interval between 6.2 and 4.5 GHz.

Figure 2 shows the dependence of the area difference DS = $S_{6.2}$ –$S_{4.5}$, where $S_{6.2}$ and $S_{4.5}$ are the areas under the graphs in Fig. 1 for frequencies of 6.2 and 4.5 GHz, respectively, depending on the concentration of g-picoline. Since DS is the temperature-averaged difference in hypersonic speeds at the specified frequencies, this value can be conditionally considered as a quantitative characteristic of the hypersound velocity dispersion.

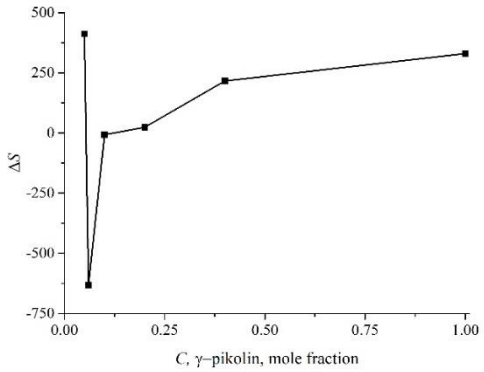

Fig. 2. Concentration dependence of *DS*

In this case, the concentration dependence of DS anticorrelates with the concentration dependence of the scattering coefficient (see [1]): a sharp negative minimum of DS at *C* = 0.06 ppm. coincides with the position of the scattering peak at the same concentration. Thus, it has been found that the appearance of a nanodroplet phase (peak of anomalous scattering) leads to a change in the sign of the dependence *dV/dw*, where *V* - is the hypersound velocity.

Conclusions

In aqueous solutions of g-picoline, the situation occurs when the bulk viscosity relaxes at a relatively low frequency compared to other aqueous solutions. This led to the fact that at frequencies *f*>4GHz, hypersonic speed dispersion appeared, which was not associated with the processes of relaxation of bulk viscosity. The temperature, concentration, and frequency dependences of the hypersound velocity consistently reflect the following: a) Positive sound velocity dispersion for pure g- picoline and for a solution with a concentration of C=0.4 ppm. g-p. b) Positive dispersion for C=0.2 and 0.1 ppm. g-p. for temperature t>50^{0}C, negative dispersion for temperatures t<50^{0}C, c) For a concentration of 0.06 ppm g-p. only negative variance is observed. Such a transformation of the sign of the dispersion can be associated with the process of solution structurization, which begins at a concentration of 0.2 ppm. g-p. and reaches its maximum value at C=0.06 ppm. g-p. In parallel with structurization, the thermodynamic state of the solution approaches (ECP) or (LCP) [16].

With a decrease in the concentration of g-picoline, the solution is closer to the periphery of the closed separation curve, and therefore the influence of the Upper critical point and the Lower critical point becomes weaker. With a decrease in the concentration of g-picoline in water, the process of structurization intensifies and its influence increases to a maximum associated with the optimal number of g-picoline molecules.

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