

Negative Dispersion of Hypersonic Velocity in an Aqueous Solution of Gamma-Picoline

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Aqueous solutions of some non-electrolytes (alcohols, acetone, etc.) at a certain temperature and concentration of non-electrolyte possess a minimum of thermodynamic stability according to the TC-diagram (a so-called singular critical point or double critical point). To explain the experimental result for these solutions, several approaches have been developed, such as the closeness to a “liquid-liquid” phase transition, a structural phase transition in a scale of near-crystalline order, a concept of existence of an unattainable critical point, as well as strengthening the local structure due to formation of over-molecular structures (clathrates). Nevertheless, there is no united opinion regarding the whole complex of effects observed in such systems.

In this work, we carried out measurements of the hypersonic velocity in aqueous solutions of gamma-picoline to study the behavior of the dispersion of high-frequency sound at different concentrations of gamma-picoline in a wide temperature interval. The velocity of sound was determined from spectra of polarized light scattering, which were registered with the help of a high-resolution two-pass Fabry-Perot interferometer. Three angles of scattering allowed us to study the propagation of sound for frequencies 2.6, 4.8 and 6.2 GHz. An analysis of experimental data showed that the character of hypersonic sound propagation is very complicated with respect to the variation of the solution's temperature and the concentration of gamma-picoline. For high concentrations of gamma-picoline, we observed an increase of hypersonic velocity with rising frequency of sound. However, for smaller concentrations (below 0.4 mole fraction), we observed both positive dispersion (the velocity increases with increasing frequency) and negative dispersion (the velocity decreases with increasing frequency). For example, for a solution with a concentration 0.2 m.f. of gamma-picoline, we observed a positive dispersion of sound velocity in the temperature interval from 283 K to 333 K, and a negative dispersion in the temperature interval from 333 K to 353 K. The experimental results are discussed in terms of a local strengthening of water's H-bond continuum due to the formation of a clathrate structure.