Analysis of the spectrum of light scattering and ultra- and hypersonic propagation in β-picoline-water solution

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The temperature dependence of the intensity of the central component in the spectrum of Mandel'shtam-Brillouin scattering in a β-picoline-water solution is investigated. An intensity maximum was observed in the temperature range 60–70 °C. A maximum for the absorption of hypersound and an absence of the temperature dependence of ultra- and hypersound, velocities, $\partial V/\partial T \approx 0$, were observed in the same temperature interval. At lower temperatures $\partial V/\partial T = -0.8$ and at higher temperatures $\partial V/\partial T = -3.6$.

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1. Analysis of linearly polarized, spectrally undecomposed light in some unstratified solutions indicated the existence of binary solutions in which the intensity of the named scattering greatly increases upon approaching a certain concentration and temperature (see Ref. 1 and the references given therein). Such solutions, for example, are acetone-water and β-picoline-water. These solutions, considered binary solutions with a closed stratification region that has been contracted to a point, are called solutions with a "special point." [2]

Until now, the temperature dependence of hypersonic and ultrasonic velocity and the corresponding absorption coefficients, as well as the intensity distributions of linearly polarized light with respect to velocities sensitive to phase transformations have not been analyzed in such solutions.

Our goal is to study the temperature dependences of hypersonic and ultrasonic velocity, the absorption coefficient of hypersound and the kinetics of the spectrum of light scattering in β-picoline-water solution at β-picoline concentration of 28.4%. According to the data,[1] a sharp increase of the scattering intensity of linearly polarized light has been observed at such concentration.

2. In our experiment we investigated the propagation of hypersound according to the spectra of Mandel'shtam-Brillouin scattering.[3,4] The propagation velocity of hypersound was determined with an accuracy of ± 1% from the shift of the Mandel'shtam-Brillouin components. The absorption coefficient of hypersound was determined from the true width of the Mandel'shtam-Brillouin components after eliminating the instrumental width of the profile from the observed width.[5] The absorption coefficient of hypersound was determined with an accuracy of ± 5%.

Figure 1 shows the curves for the temperature dependence of the central component I'_c relative to the intensity of the central component at 40 °C, I'_{c40}. Both central components were normalized in all the cases by the total intensity of the Mandel'shtam-Brillouin components, $I'_c = I_c/2I_{M-B}$.

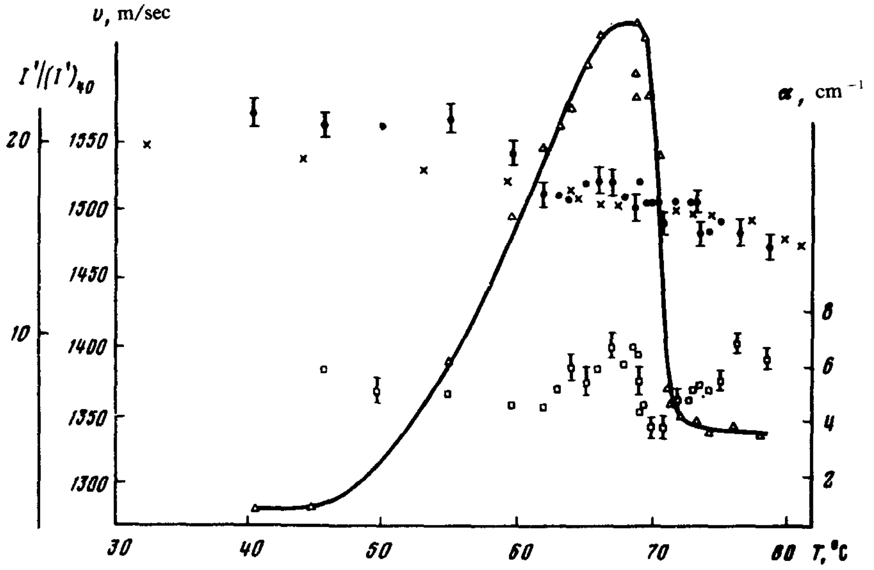

FIG. 1. Temperature dependence of the intensity of the central component in the spectrum of Mandel'shtam-Brillouin scattering by a normalized intensity of the Mandel'shtam-Brillouin components $I'_c = T_c/2I_{M-B}$ in a β-picoline-water solution. □, Temperature dependence of the absorption coefficient of ~4.6-Ghz hypersound in the same solution; ●, temperature dependence of the propagation velocity of ~4.6-GHz hypersound in a β-picoline-water solution; ×, temperature dependence of the propagation velocity of 2.8-MHz ultrasound in the same solution.

We can see in Fig. 1 that I_c exceeds I'_{c40} by more than a factor of 20. The maximum lies in the temperature range of 50 to 70 °C. The hypersonic absorption maximum is also located in this temperature range (Fig. 1).

The results of measurement of the propagation velocity of hypersound in the 40 to 78 °C temperature range are shown in Fig. 1.

It can be seen in the plot of the propagation velocities that the ultrasonic and hypersonic velocities can be broken down into three distinct regions. In the 40 to 55 °C temperature range we can see a monotonic decrease of the ultrasonic and hypersonic velocities with the temperature, where the value $\partial V/\partial T = -0.8$. In the second temperature interval of 62 to 72 °C the ultrasonic and hypersonic velocities are almost independent of the temperature $\partial V/\partial T \approx 0$ within the accuracy limits of the experiment. In this temperature interval we can see the maximum of the hypersonic absorption, and, as mentioned above, the intensity maximum of the central component.

The propagation velocity of ultrasound was measured with an accuracy of $\pm 0.2\%$ by using a method described in Ref. 6. In the third temperature interval of 72 to 78 °C, $\partial V/\partial T = -3.6$, which is several factors higher than the temperature dependence of the hypersonic velocity in the temperature range of 40 to $+60$ °C.

A comparison of the ultrasonic and hypersonic velocities shows that there is a ~2.2% maximum positive dispersion of the sound velocity in the 40 to 60 °C tempera-

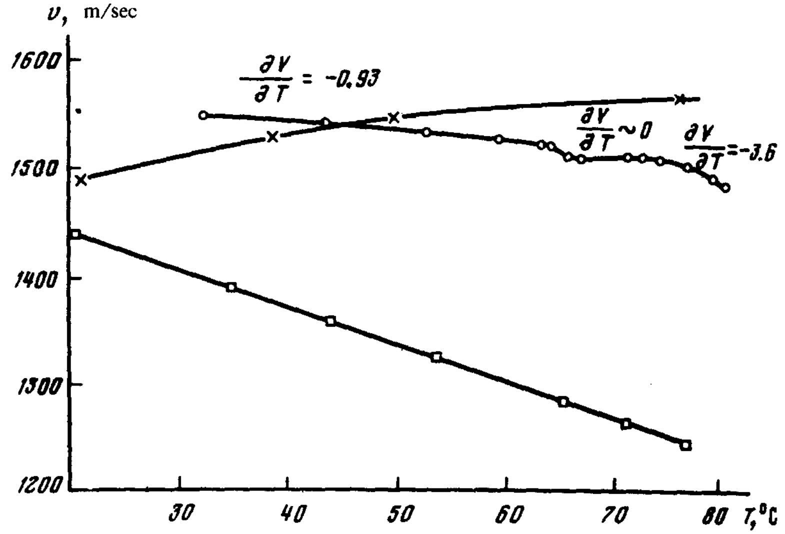

FIG. 2. ○, temperature dependence of the propagation velocity of 2.8-MHz ultrasound in a β-picoline-water solution; □, temperature dependence of the propagation velocity of ultrasound in a pure β picoline; ×, temperature dependence of the propagation velocity of ultrasound in pure water (our measurements and table values[8]).

ture range, and there is no sound velocity dispersion with further increase of temperature within the error limits of the experiment.1)

Figure 2 shows the measurements of the temperature dependence of the ultrasonic velocity in pure β-picoline and pure water, in addition to the temperature dependence of the propagation velocity of ultrasound in β-picoline-water solution. Here, we consider it important to point out one more peculiarity of β-picoline-water solution, i.e., the propagation velocity of ultrasound in a solution is higher than in any one of the pure components in the temperature range of 32 to 42 °C; this means that the solution's compressibility is smaller than that of water.

3. Although there is no theory that can describe the behavior of the propagation velocity of ultrasound and hypersound in such solutions, an attempt was made to develop one for absorption[7]; however, the necessary parameters for comparison of this theory with the experiment are not available.

The results of investigation of the propagation of sound in the investigated β-picoline-water solution are of special interest. Initially, we expected no anomalies in the behavior of the sound velocity at some distance from the "special point" at lower and higher temperatures than it, since the concentration fluctuations are weak at these temperatures. However, an experimental study in the ultrasonic and hypersonic frequency ranges showed that the sound velocity has a considerably smaller temperature dependence at lower temperatures than the special point than at higher temperatures than the special point. Our result seems to indicate that there is a certain structural rearrangement of the material occurring as a result of passage through the region of the special point.

A comparison of the measurements performed by us with the results of earlier experiments involving stratified solutions shows that on the basis of the behavior of the intensity I'_c, α, and V the solution must be close to the critical stratification point, although the point itself has not been formed in our case. However, the behavior of the propagation velocity of sound in β-picoline-water solution, which is still unclear, requires an explanation.

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[1)] In the investigation performed by us the propagation velocity of ultrasound in the temperature range above 70 °C coincides with the velocity of hypersound within the error limits of the measurements, but the ultrasonic data systematically lie above the hypersonic data in all the series of the experiment. The reason for this requires an explanation.

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