Behavior of the electrostriction coefficient in a β-picoline–water solution with a singular point

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Stimulated Brillouin scattering has been studied in a β-picoline–water solution with a singular point. The temperature dependence of the electrostriction coefficient has been determined. This coefficient changes abruptly near the singular point.

Certain aqueous solutions of nonelectrolytes, at certain concentrations and certain temperatures, exhibit a state of an unstable thermodynamic equilibrium: The second derivative of the molar thermodynamic potential is close to zero. For a β-picoline–water solution, this state (the so-called singular point) corresponds to a β-picoline mole fraction of 0.06 and a temperature of 69 °C (Ref. 1). It is believed that at this concentration and this temperature the solution is at its closest to the critical point for stratification. The susceptibility of the system should accordingly differ from that of ordinary solutions. A study of stimulated Brillouin scattering in a β-picoline–water solution reveals a nonmonotonic increase in the intensity of the stimulated scattering.[2]

In this letter we are reporting a determination of the temperature dependence of the electrostriction coefficient $Y=\rho(\partial\epsilon/\partial\rho)$ (ρ and ϵ are respectively the density and dielectric constant of the medium) from measurements of the excitation threshold and reflection coefficients for stimulated Brillouin scattering in a β-picoline–water solution with a singular point.

The stimulated Brillouin scattering was excited by the second harmonic from a neodymium laser in single-mode operation with a pulse length of 20 ns. The laser light was focused at the center of the cell holding the solution, whose temperature was regulated within 0.1 °C. We studied the temperature dependence of the excitation threshold E_{th} and the dependence of the reflection coefficient R on the intensity of the laser light. These measurements were carried out over the temperature range 20–86 °C.

Figure 1 shows the temperature dependence of the reflection coefficient $R(\cdot)$ under saturation conditions and the temperature dependence of the normalized threshold for stimulated Brillouin scattering, $k=E_{th}/E_{th}^{20}$, where E_{th}^{20} is the value of the threshold found at a solution temperature of 20 °C. We see that the reflection coefficient increases nonmonotonically under saturation conditions, while the threshold for stimulated Brillouin scattering decreases with increasing solution temperature.

Near the excitation threshold, the intensity of the stimulated Brillouin scattering, I_{MB}, is given by[3]

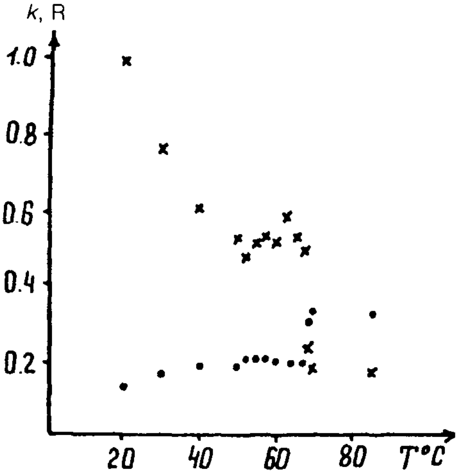

FIG. 1. The reflection coefficient under saturation conditions, $R(\cdot)$, and the threshold energy for stimulated Brillouin scattering, $k(x)$, versus the solution temperature.

$$I_{MB}=I(0)\exp(GIz). \quad (1)$$

Here $I(0)$ is the intensity of spontaneous Brillouin scattering, I is the intensity of the laser light in the caustic, and z is the nonlinear-interaction length, which we take to be the length of the caustic of the laser light focused in the cell. In accordance with the constant experimental geometry and in accordance with expression (1), the experimental temperature dependence of E_{th} and R implies a temperature dependence of the gain G. The value of G, whose temperature dependence is shown in Fig. 1, was found from expression (1) for various solution temperatures on the basis of the conditions for the focusing of a Gaussian beam by a lens with a focal length of 10 cm and for an intensity $I(0)=10^{-11}$. The values shown here, as in Fig. 1, are normalized to the value of G at 20 °C. We see that the change in G with increasing temperature is rather complex.

Near the Stokes resonance the value of the constant G is given by[3]

$$G=Y^2\omega^3/\Omega\Gamma c^4\rho. \quad (2)$$

Here Ω is the frequency shift and Γ the half-width of the Brillouin maximum, c is the velocity of light, and ω is the frequency of the laser light. This relation was used along with values of Γ, Ω, and ρ from Ref. 4 to calculate $G(t)$. The behavior found as a result is shown by the solid line in Fig. 2. Comparison of the experimental and theoretical results shows that at temperatures between 20 and 67.5 °C the experimental results agree with the theory, *i.e.*, are determined by the values of Ω, Γ, and ρ. Near and above the singular point of this solution, on the other hand, there is a discrepancy of a factor $\sim$2.5.

According to expression (2), the only parameter which determines the experimental dependence of G_{expt} near the singular point is the electrostriction coefficient, relative values of which were calculated from the experimental data and expression (2).

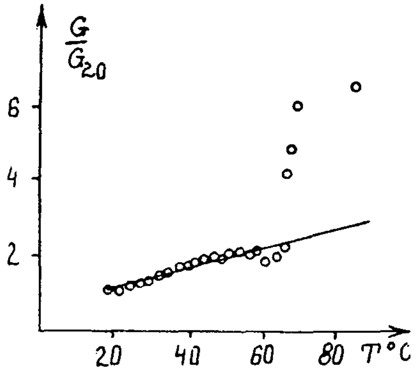

FIG. 2. Temperature dependence of the gain for stimulated Brillouin scattering in a β-picoline–water solution. Circles—Experimental data; solid lines—theoretical results.

Determining numerical values of Y in the hypersonic range is a rather complex problem, involving an extrapolation of data found for ultrasound.[5] The calculation method used in the present study does not yield Y very accurately, because approximate values are used for such parameters as z and $I(0)$. Determining accurate values of Y for these media will apparently require the use of both the method described in the present study and that of Ref. 5.

We see in Fig. 3 that near the singular point there is an abrupt change in Y, by a factor of 1.4–1.6, from the values found in the temperature range 20–67.5 °C.

The observed behavior of Y near and above the singular point is apparently attributable to the same processes which account for the narrow peak in the intensity of light scattering[1] and for the minimum of the diffusion coefficient.[6] The nature of these processes is currently being studied.

This study of the temperature dependence of the threshold and reflection coefficient for stimulated Brillouin scattering in a β-picoline–water solution thus indicates that there is an abrupt increase in the electrostriction near the singular point of the solution. Since the cubic nonlinear susceptibility, which describes the stimulated Brillouin scattering within a constant factor, is proportional to the square of the electros-

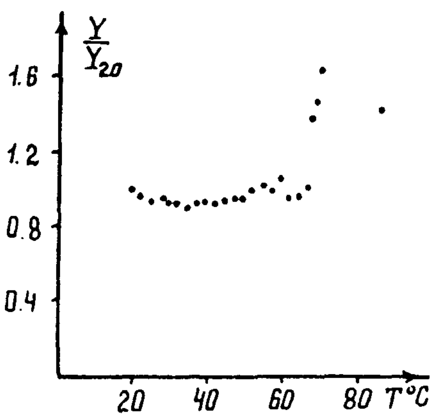

FIG. 3. Temperature dependence of the electrostriction coefficient in a β-picoline–water solution.

triction coefficient, there are substantial changes in the stimulated Brillouin scattering in the temperature interval in which the solution is at the boundary of thermodynamic instability. These substantial changes are manifested as a pronounced decrease in the excitation threshold.

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