Field dependence of depolarized critical opalescence

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The depolarization coefficient Δ of scattered light along the critical isotherm of n-pentane has been investigated experimentally for two visible wavelengths $\lambda_1 = 435.8$ nm and $\lambda_2 = 632.8$ nm, and a qualitative difference has been detected in the height behavior $\Delta(H)$ of an anisotropic medium for different λ. The field dependence of depolarized critical opalescence, which is determined by the tensor fluctuations of the dielectric constant of the medium, was investigated on the basis of the obtained data.

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It was shown previously[1] that one possible cause of the depolarization of scattered Rayleigh radiation near the critical point is the tensor fluctuations of the dielectric constant of the medium. According to Ref. 1, the intensity of depolarized scattering of light by these fluctuations varies as $I \sim \lambda^{-4}\beta_T^{1/2} \sim \lambda^{-4}R_c$ (here λ is the light wavelength, and β_T and R_c are the compressibility and the correlation radius of the substance, respectively). Until now, these assumptions of the theory of depolarized scattering of light have not been confirmed experimentally.

We must also note that at present only the temperature dependence of the intensity of depolarized light scattering along the critical isochore or phase boundary is normally investigated. A study of the field characteristics of depolarized light scattering in an optically anisotropic system near the critical point is just as important and urgent.

The purpose of this paper is to investigate the field (height) dependence of the intensity of depolarized scattering caused by the tensor fluctuations of the dielectric constant of the medium.[1]

The intensity of polarized and depolarized light scattering at a 90° angle was measured using the experimental apparatus described in detail elsewhere.[2] The exciting radiation I_0^X polarized along the X axis was directed vertically upward into the chamber with the substance being studied, i.e., it was directed parallel to the Z axis. Using this arrangement, we analyzed the light scattering of the two polarizations I_X^X and I_Z^X along the Y axis, as a function of the height of the test sample.

We used n-pentane at the critical temperature $T_c = 469.98$ K as the subject of our investigation. The critical temperature was determined with an accuracy[2] of 0.01°. Before measuring the light-scattering intensity, the system was temperature stabilized for a day with an accuracy of 0.005°. The intensity of the polarized I_X^X and depolarized I_Z^X components was measured for two visible wavelengths $\lambda_1 = 435.8$ nm (the source was a DRSh-250 mercury lamp) and $\lambda_2 = 632.8$ nm (LG-78 He-Ne laser).

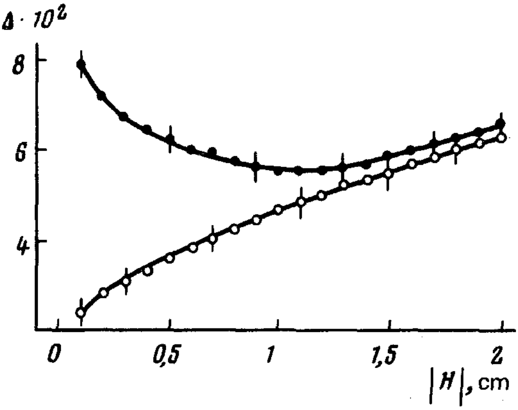

FIG. 1. Height dependence of the depolarization coefficient along the critical isotherm of *n*-pentane for two visible wavelengths: •—λ_1 = 435.8 nm, and ○—λ_2 = 632.8 nm.

We calculated the depolarization coefficients $\Delta = I_Z^X / I_X^X$ on the basis of the experimental data for I_X^X and I_Z^X. The obtained results are shown in Fig. 1. As seen from this figure, the height dependence $\Delta(H)$ for the two visible wavelengths is significantly different. For $\lambda_1 = 435.8$ nm the Δ decreases with increasing H; for $\lambda_2 = 632.8$ nm the depolarization coefficient increases as one goes upward from the $H = 0$ level at which the critical density of the substance is realized. In the region $H \geqslant 1$ cm Δ is almost independent of λ within the limits of experimental errors. This is the first time that such behavior of the depolarization coefficient has been observed along the critical isotherm in the range of the field variable $h = \rho_c g H P_c^{-1}$ from 0 to $h \approx 10^{-5}$ The obtained result can be explained qualitatively by the following two factors. Experimental measurements of critical opalescence in an anisotropic medium near the critical point have shown[2] that the intensity of scattered light in the region of the field variable $h \geqslant 10^{-6}$ obeys the Rayleigh-Einstein law ($I_X^X \sim \lambda^{-4}\beta_T$) at all temperatures, including the critical temperature. In this region of heights both secondary scattering $I_{2Z}^X \sim (\lambda^{-4}\beta_T)^2$ and, according to Ref. 1, depolarized single scattering of light $I_{1Z}^X \sim (\lambda^{-4} R_c)$ associated with tensor fluctuations of the dielectric constant of the medium contribute to the depolarized light scattering I_Z^X. Thus the component measured experimentally is $I_Z^X = I_{1Z}^X + I_{2Z}^X$, and the height dependence of the depolarization coefficient $\Delta(H)$ is determined by the relative contribution to I_Z^X of these two dissimilar types of scattered light,

$$\Delta = (I_{1Z}^X + I_{2Z}^X)(I_X^X)^{-1} = A R_c^{-1} + B \lambda^{-4} \beta_T \,. \tag{1}$$

Here A and B are constant quantities for a given substance. In the region of heights $h \geqslant 10^{-6}$ for those wavelengths λ for which $I_{2Z}^X \gg I_{1Z}^X$ ($\lambda_1 = 435.8$ nm) the depolarization coefficient must decrease as $\Delta \sim \lambda^{-4}\beta_T \sim \lambda^{-4} H^{1/\delta - 1}$ with increasing field variable h.[3] As the wavelength of excitation radiation increases the relative fraction of the rescattered light in the depolarized component I_Z^X decreases. When $I_{1Z}^X \gg I_{2Z}^X$ ($\lambda_2 = 632.8$ nm) the depolarization coefficient $\Delta \approx I_{1Z}^X / I_X^X$ increases as $\Delta \sim R_c^{-1} \sim H^{(\delta-1)/2\delta}$. The performed analysis was confirmed qualitatively by the

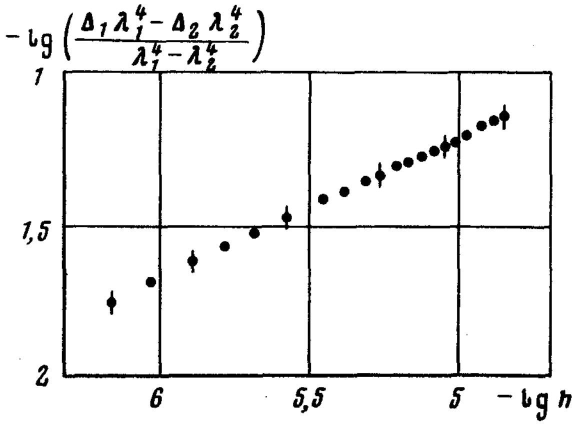

FIG. 2. Field dependence of the depolarized critical opalescence.

obtained experimental results (Fig. 1).

On the basis of these experimental data of the behavior of the depolarization coefficient for two wavelengths of visible light, it is possible to examine the field dependence of the depolarized critical opalescence.[1] According to Eq. (1),

$$\frac{I_{1Z}^X}{I_X^X} \approx \frac{\Delta_1(\lambda_1)\lambda_1^4 - \Delta_2(\lambda_2)\lambda_2^4}{\lambda_1^4 - \lambda_2^4}. \quad (2)$$

This function in the region of the field variable $h = 10^{-6}$-10^{-5} is shown in Fig. 2, in which it can be seen that the field dependence of the ratio I_{1Z}^X/I_X^X along the critical isotherm obeys the relation $(I_{1Z}^X)(I_X^X)^{-1} \sim h^{\xi}$, where $\xi = 0.4 \pm 0.6$. This result, which confirms the prediction of the theory[1] within the experimental error limits

$$(I_{1Z}^X \sim R_c \sim h^{-(\delta-1/2\delta)}; \quad \delta = 5[4]),$$

is in agreement with the experimental studies of the field dependence of the correlation radius along the critical isotherm of n-pentane[5] ($R_c \sim h^{-\xi}$, $\xi = 0.405 \pm 0.025$).

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