DEPOLARIZED LIGHT SCATTERING IN INHOMOGENEOUS SYSTEMS UNDER GRAVITY NEAR THE CRITICAL POINT

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The results of experimental studies of the intensities of the polarized and depolarized components of light scattered in inhomogeneous n-pentane and cyclopentane under gravity near their critical temperatures are presented. On the basis of these data, it has been shown that the depolarized light scattering in such inhomogeneous systems consists of the depolarized secondary and depolarized primary light scattering, the latter being caused by the tensor character of fluctuations of the dielectric permittivity of the substance in the vicinity of the critical point.

Experimental and theoretical researches of the secondary depolarized light scattering in a substance, which is in its critical state, have been started relatively long ago (see, e.g., [1,2]), but their basic results correspond, as a rule, to spatially homogeneous systems.

When analyzing the results obtained during the studies of the secondary depolarized light scattering in such spatially homogeneous systems near the critical points [1, 2], the following conclusions were drawn: in the Rayleigh region ($k^2R_c^2 \ll 1$), the intensity of the secondary light scattering I_2 is proportional to the square of the intensity of the primary light scattering I_1 ($I_2 \propto I_1^2 \propto \beta_T^2\lambda^{-8}$). Hence, the depolarization factor $\Delta = I_2I_1^{-1} \propto I_1 \propto \beta_T\lambda^{-4}$ is proportional to the intensity of a single light scattering. This means that if the critical point is approached, the value of the depolarization factor Δ grows. Moreover, it follows from this relation that the shorter the wavelength of the scattered light, the larger the depolarization factor.

The problem becomes considerably complicated when analyzing the light scattering near the critical points in spatially inhomogeneous systems, which are under the action of the gravitational field. Such researches of weakly inhomogeneous systems, for which the relative variation of the chemical potential $\Delta\mu = (\mu - \mu_c)/\mu_c$ is supposed to be equal to the relative variation of the hydrostatic pressure $h = \rho_c gz/P_c$, i.e. $|\Delta\mu| = |h|$, have been started earlier in works [3, 4] and others. Here, μ_c, ρ_c, and P_c are the critical values of the chemical potential, density, and pressure, respectively. The drawn conclusions concerning the features in the behavior of the secondary light scattering in those weakly inhomogeneous systems are close to the conclusions made in [1,2].

However, it should be emphasized that the condition $|\Delta\mu| = |h|$ is not fulfilled in the majority of real systems. All experimental researches of the gravitational effect, without exceptions, carried out at the faculty of molecular physics at the Taras Shevchenko Kyiv National University (see, e.g., [5–7]) testify to that, in inhomogeneous systems with large critical temperatures, $T_c \geq 300$ K, the value of $|\Delta\mu|$ is about $10 \div 10^2$ times $|h|$. The relation $|\Delta\mu| \approx |h|$ is valid only for objects with small critical temperatures $T_c \approx 5 \div 20$ K, e.g., H_2 and O_2.

Therefore, nowadays, vital is the experimental research of the behavior of the depolarized light scattering in real, essentially inhomogeneous systems near the critical point with the purpose of verifying the applicability of the theory of secondary light scattering in homogeneous systems to spatially inhomogeneous ones.

Such experimental researches have been started earlier in [5, 6]. In these works, the depolarized light scattering in an inhomogeneous system under gravity was shown for the first time to consist of the depolarized

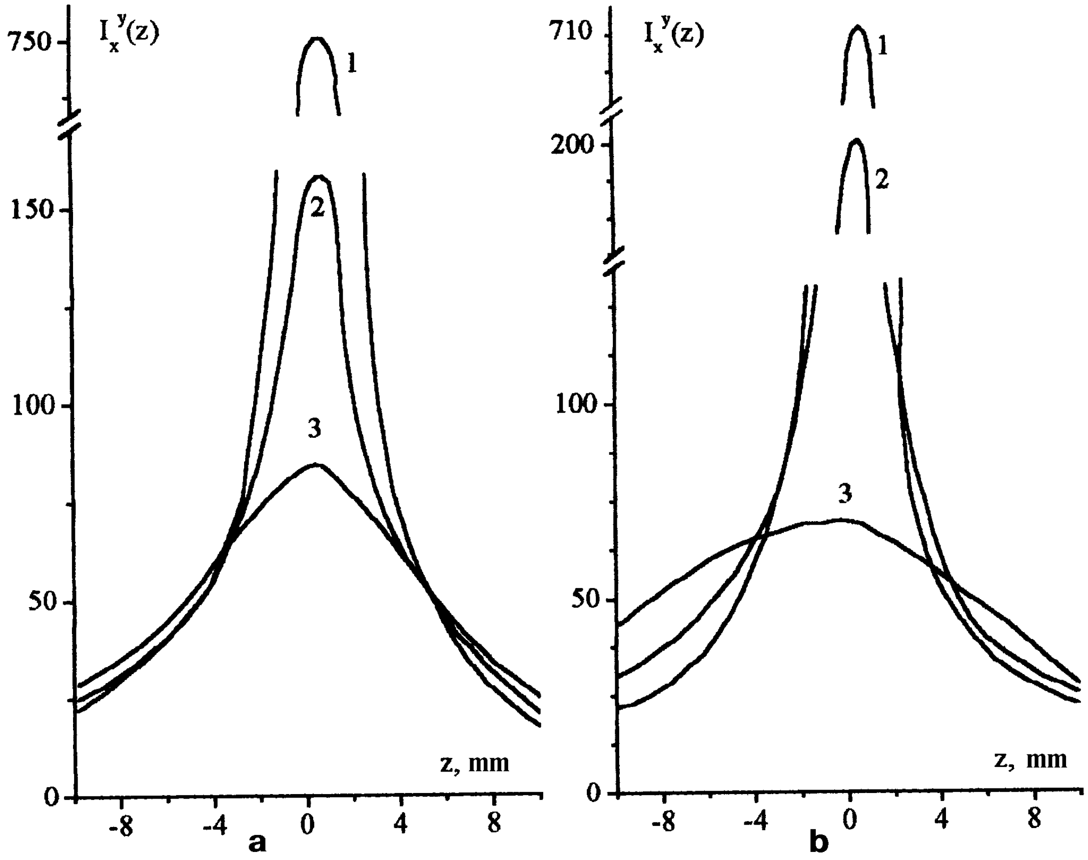

Fig. 1. Height dependences of the intensity of the depolarized light scattering in an inhomogeneous substance under gravity near the critical point at $\Delta T > 0$ in (*a*) n-pentane ($\Delta T = 0.03$ (*1*), 0.42 (*2*), and 0.85 K (*3*)) and (*b*) cyclopentane ($\Delta T = 0.02$ (*1*), 0.37 (*2*), and 0.85 K (*3*))

secondary light scattering [1–4] and the depolarized primary critical opalescence caused by the tensor character of fluctuations of the dielectric permittivity of the system [7].

Following those studies, in this work, we carried out the measurements of both the field and temperature dependences of the depolarized light scattering and the depolarization factor in essentially inhomogeneous systems ($|\Delta\mu| \gg |h|$) near the liquid–vapor critical temperature and tested the validity of some fundamental statements of the theories of the secondary light scattering [1–4] for such inhomogeneous systems.

The measurements of the intensity of the polarized and depolarized light scattering at an angle of 90° in essentially inhomogeneous pure substances, namely, n-pentane and cyclopentane, were carried out using the experimental installation described in [6, 7]. The experimental procedure was as follows. The exciting radiation of one of two polarizations, I_0^x or I_0^y, polarized with the help of a polaroid was sent vertically upwards through the researched system along the z axis. In so doing, the intensity of light possessing either I_x^x or I_x^y polarization (the superscript indicates the polarization direction of the exciting wave and the subscript does the polarization of the scattered light) was analyzed along the z-direction with the help of another polaroid.

The measurements of the height and temperature dependences of the polarized and depolarized components of the scattered light were carried out for five light wavelengths in the visible range, $\lambda = 633$, 579, 546.1, 436, and 405 nm. The experimental results obtained for the depolarized light scattering in an inhomogeneous environment for $\lambda = 546.1$ nm are presented in Fig. 1. From this figure, one can see that, similarly to the case of the polarized light scattering in an inhomogeneous environment I_x^x [6], the intensity of the depolarized light scattering I_x^y also varies strongly along the height of the optical cell. As one would expect, the sharpest dependence of the intensities of the depolarized

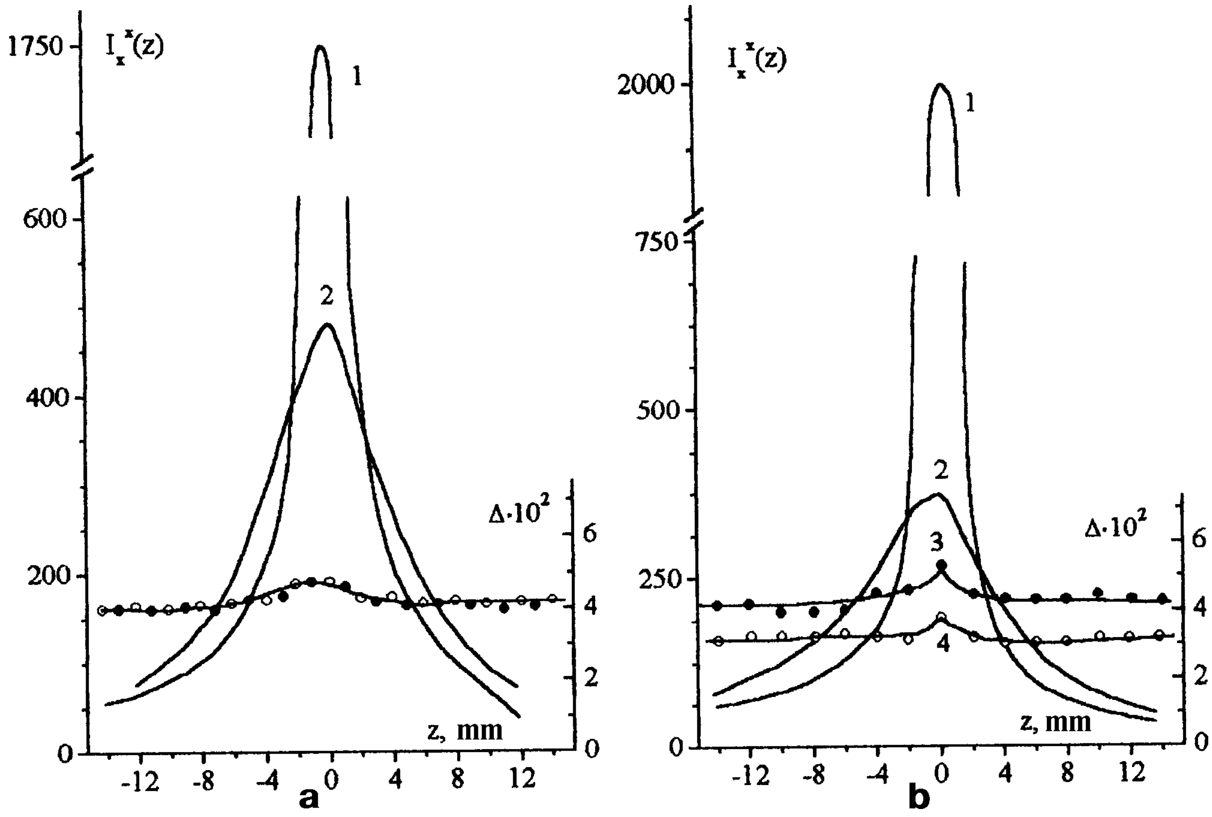

Fig. 2. Height dependences of the intensity of the polarized light scattering I_x^x (curves) and of the depolarization factor $\Delta = I_x^y/I_x^x$ (circles) in an inhomogeneous substance at various temperatures ΔT in (*a*) n-pentane ($\Delta T = 0.03$ (*1* and hollow circles) and 0.58 K (*2* and solid circles)) and (*b*) cyclopentane ($\Delta T = 0.02$ (*1* and solid circles) and 0.54 K (*2* and hollow circles))

components of the scattered light I_x^y on the height is observed near the critical temperatures. For example, at the dimensionless temperature $t = (T - T_c)/T_c \approx 10^{-4}$, the intensity of the depolarized scattering at the height $z = 0$, where the critical density of the substance takes place, is almost 50 times as much as the intensity of light scattering at height $z \geq 1$ cm. While moving away from the critical temperature, the dependence I_x^y on the height becomes more and more smooth.

Experimental data concerning the height and temperature dependences of the intensities of the polarized, I_x^x, and depolarized, I_x^y, light scattering were used for the calculations of the depolarization factor $\Delta = I_x^y/I_x^x$ (Figs. 2 and 3). Then, the obtained results were used for checking the applicability of some above-indicated consequences of the theory of the secondary light scattering in homogeneous systems [1–3] to essentially inhomogeneous systems.

If one assumes that the depolarization of the scattered light near the critical point is connected mainly with the secondary scattering [2] ($I_x^y \propto (I_x^x)^2$), then the dependence of the depolarization factor Δ on the height must repeat the height profile of the polarized component I_x^x.

However, as is seen from Fig. 2, the depolarization factor Δ remains practically constant within the limits of experimental errors (±10%) in the whole investigated intervals of temperatures $t \geq 10^{-4}$ and height $z \geq 0.2$ cm. For example, at $t \approx 10^{-5}$, $\Delta \approx 4 \times 10^{-2}$ for n-pentane and $(3 \div 4) \times 10^{-2}$ for cyclopentane. At the same time, the intensity of the polarized component I_x^x changes more than by a factor of 10 in the same interval of height under the appropriate conditions.

Let us now analyze the temperature dependence of the depolarization factor $\Delta = I_x^y/I_x^x$ along the line of critical isochore $z = 0$ (see Fig. 3). As is seen from the figure, the depolarization factor both in n-pentane and in cyclopentane changes only by 20—25% in the temperature range of $\Delta T = T - T_c$ from 0.8 to 0.04 K. At the same time, the experimentally measured intensity of

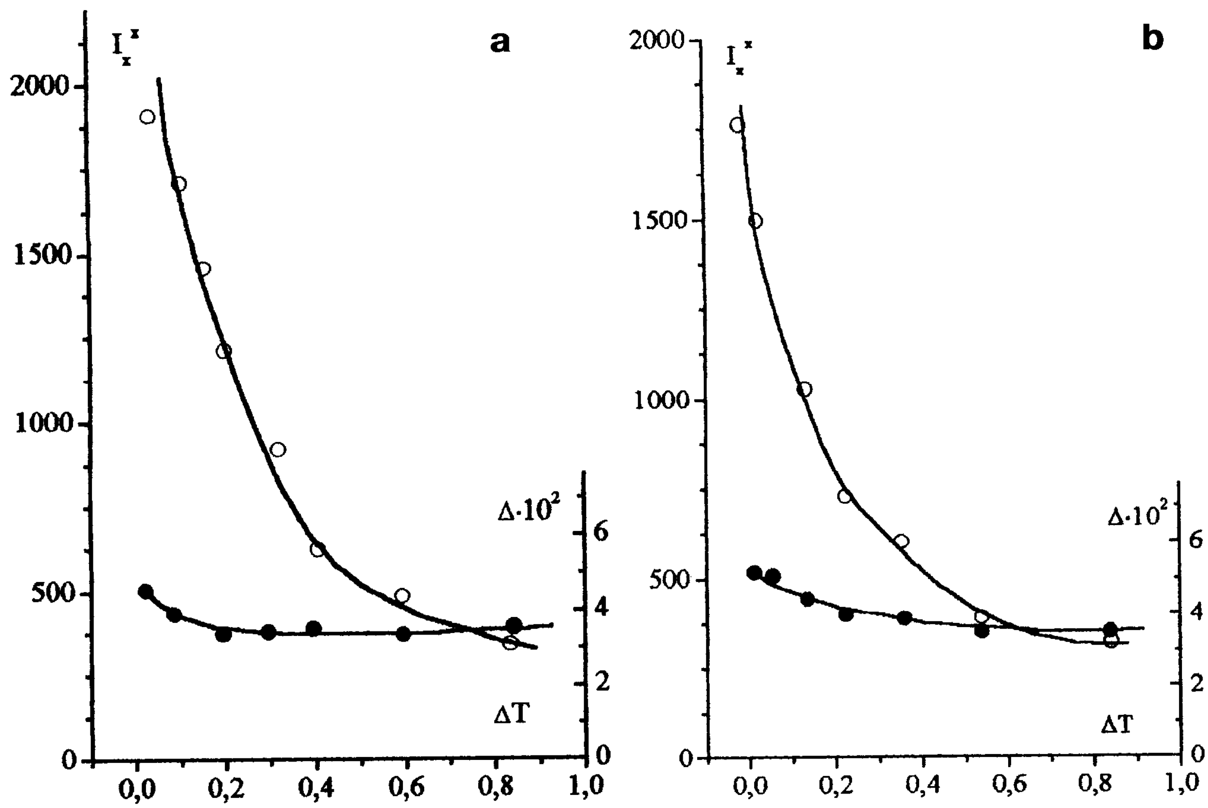

Fig. 3. Temperature dependences of the intensity of the polarized light scattering I_x^x (hollow circles) in an inhomogeneous substance along the critical isochore ($z = 0$) and the depolarization factor $\Delta = I_x^y/I_x^x$ (solid circles) in (a) n-pentane and (b) cyclopentane

the polarized light I_x^x increases by an order of magnitude in this temperature interval.

It follows from here that the obtained experimental data for I_x^y and $\Delta(z,t)$ cannot be explained only in the framework of the known theories of the secondary scattering of light in homogeneous systems.

For a more detailed analysis of the behavior of the depolarization factor Δ in the vicinity of the critical isochore in the height range $z < 0.2$ cm, the measurements of the polarized, I_x^x, and depolarized, I_z^x, components of the scattered light intensity were carried out at the critical temperature of n-pentane for five wavelengths of exciting radiation $\lambda = 632.7$, 579.0, 546.1, 435.8, and 404.7 nm.

The obtained experimental results are shown in Fig. 4,a. On the basis of these data, the depolarization factors $\Delta(z) = i_z^x/i_x^x$ in a unit volume along the critical isotherm were calculated (Fig. 4,b). Here, $i_x^x = I_x^x/V_1$, $i_z^x = I_z^x/V_2$, and V_1 and V_2 are the volumes which emitted the polarized and depolarized radiation, respectively. As is seen, the height dependences of the depolarization factor $\Delta(z)$ have qualitatively different characters for different λ. For $\lambda = 632$ nm, Δ grows with z; for $\lambda = 546.1$ nm, Δ is almost independent of the height; for $\lambda = 404.7$ nm, the depolarization factor decreases as z increases. Such a behavior of the depolarization factor near the critical point in an optically inhomogeneous environment has been revealed experimentally for the first time and, as earlier, cannot be explained by assuming the presence of only the secondary light scattering.

For the explanation of the results obtained, we engaged [5] a qualitatively different mechanism of the possible depolarization of scattered radiation near the critical point, connected with the tensor character of fluctuations of the dielectric permittivity of the medium [7]. In accordance with [7], the intensity of the depolarized light scattering by these fluctuations changes according to the law $I_{1z}^x \propto \lambda^{-4}\beta_T^{1/2} \propto \lambda^{-4}R_c$.

Then, using the consequences of this theory [7] and the theory of the secondary light scattering [1–4], we can present the experimentally measured intensity of the depolarized light scattering in an inhomogeneous

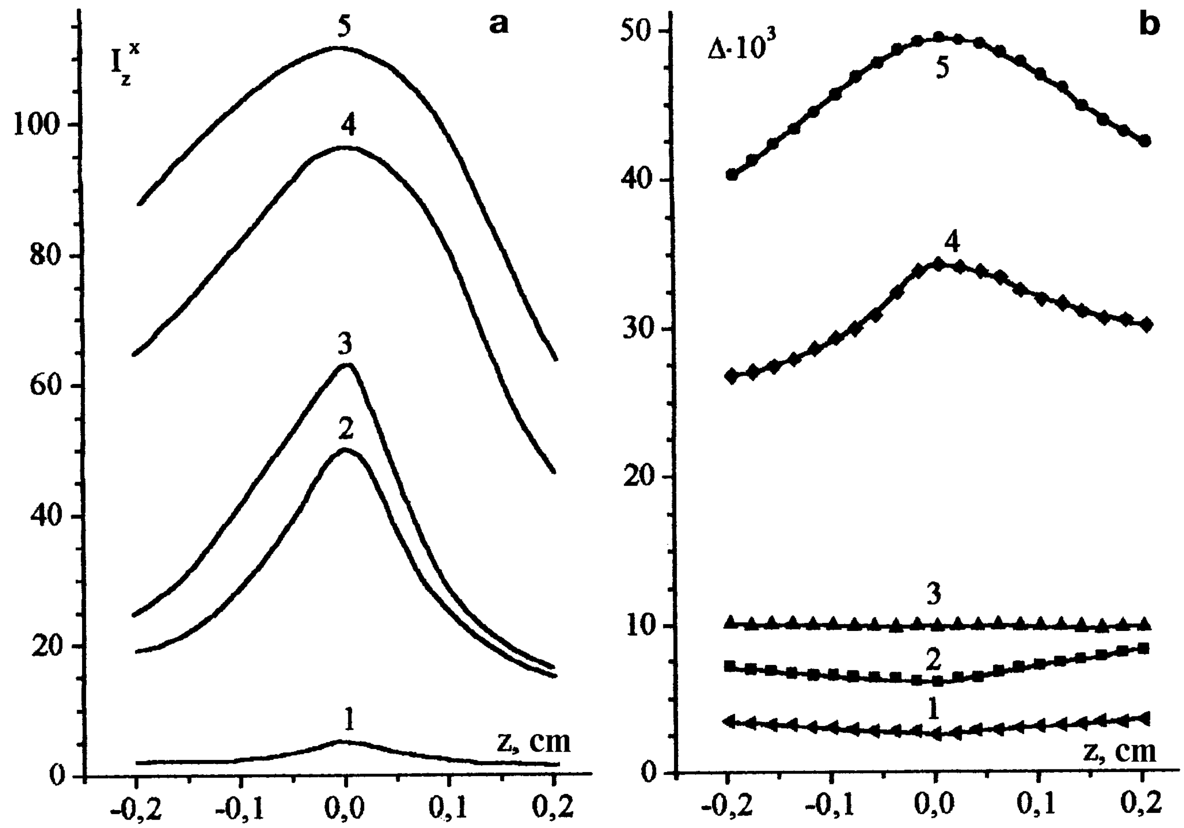

Fig. 4. Height dependences of the intensity of the depolarized light scattering I_z^x (*a*) and of the depolarization factor $\Delta = i_z^x / i_x^x$ (*b*) at the critical temperature of inhomogeneous n-pentane for various wavelengths of visible light $\lambda = 632.8$ (*1*), 579 (*2*), 546.1 (*3*), 435.8 (*4*), and 404.7 nm (*5*)

medium I_z^x as $I_z^x = I_{1z}^x + I_{2z}^x$, where $I_{1z}^x \propto \beta_T^{1/2} \propto R_c$ and $I_{2z}^x \propto \beta_T^2$. Hence, the behavior of the depolarization factor is defined by a relative contribution of those two kinds of the scattered light, being different by their nature, to the depolarized component I_z^x ($\Delta = (I_{1z}^x + I_{2z}^x)/I_x^x$). On the basis of this ratio, we can analyze the height dependences of the depolarization factor of the scattered light obtained at the critical temperature of n-pentane and displayed in Fig. 4,*b* for various wavelengths of the exciting radiation.

For those wavelengths, for which $I_{2z}^x \gg I_{1z}^x$ ($\lambda = 404.7$ nm), the depolarization factor has to change according to the law $\Delta = I_{2z}^x / I_x^x \propto \beta_T(z) \propto z^{(1-\delta)/\delta}$ while the field variable z increases. Provided the wavelength of the exciting radiation becomes longer, the fraction of the secondary scattering in the depolarized component diminishes. In this case, where $I_{1z}^x \gg I_{2z}^x$, the depolarization factor $\Delta = I_{1z}^x / I_x^x \propto R_c^{-1} \propto z^{(\delta-1)/2\delta}$. Therefore, as the field variable increases, Δ decreases for small wavelengths of the exciting radiation ($\lambda = 404.7$ nm) and increases for relatively large ones ($\lambda = 637.5$ nm). For the intermediate wavelength $\lambda = 546$ nm, one might expect that $I_{2z}^x \approx I_{1z}^x$. In the last case, the depolarization factor $\Delta(\lambda = 546 \text{ nm}) \propto O_1 z^{(1-\delta)/\delta} + O_2 z^{(\delta-1)/2\delta}$ and will weakly depend on the height near the critical temperature. As we see, these conclusions are completely proved by the experimental data concerning the dependences of the depolarized light scattering and the depolarization factor on the height obtained in n-pentane at its critical temperature for five wavelengths of the exciting radiation (Fig. 4,*b*). As for the temperature dependence of the depolarization factor along the direction of the critical isochore $\Delta(t, \lambda = 546 \text{ nm}) \propto O_1' t^{-\gamma} + O_2' t^{\nu}$, it must be also a weak function of temperature for the intermediate wavelength $\lambda = 546$ nm, which is completely confirmed by the experimental data presented in Fig. 3.

Thus, on the basis of the fulfilled researches, it is possible to draw conclusion that the depolarized light scattering in inhomogeneous systems near their critical points is composed of the secondary light scattering [1–4] and of the primary depolarized critical opalescence, the latter being caused by the tensor character of fluctuations of the dielectric permittivity of the system [7].

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ДЕПОЛЯРИЗОВАНЕ РОЗСІЯННЯ СВІТЛА В НЕОДНОРІДНИХ СИСТЕМАХ В ГРАВІТАЦІЙНОМУ ПОЛІ ПОБЛИЗУ КРИТИЧНОЇ ТОЧКИ

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Резюме

Наведено результати експериментальних досліджень поляризованої та деполяризованої компонент інтенсивності розсіяного світла в неоднорідному н-пентані та циклопентані у гравітаційному полі поблизу критичної температури. На основі цих даних показано, що деполяризоване розсіяння світла в таких неоднорідних системах складається з деполяризованого вторинного, а також деполяризованого первинного розсіяння світла, зумовленого тензорним характером флуктуацій діелектричної проникності середовища поблизу критичної точки.