
THE NEUTRON REFRACTION INDEX OF CONFINED LIQUIDS IN THE CRITICAL REGION

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The problem of both density profile and neutron refraction index is studied for a non-uniform liquid in a reduced geometry. A special attention is paid to the influence of gravitation on the properties of liquid systems. New theoretical results are obtained for the shifts of the critical temperature on the phase coexistence curve and the positions of inflection points on the critical isotherm in confined liquids.

1. Introduction

The finite-size effects of the systems of different nature (liquids, magnetics, liquid crystals, biological objects, etc.) near the critical points and the points of phase transitions cause the promoted interest in the theoretical and experimental studies (see, for example, [1–4]). The main reasons which can explain the desire of researchers to study the systems with reduced geometry in details are related to at least two factors: (a) finite-size objects which are frequent in the nature have got a wider practical application; (b) reliable and adequate theoretical approaches that are similar to the finite-size scaling hypothesis and renormalization-group computations for the systems with reduced geometry have been already developed.

Neutron methods that use the transmission of neutrons and their coherent and incoherent scattering give additional possibilities (relative to, e.g., the light scattering method) for the research of rather fine aspects of the critical behavior of individual liquids and mixtures in both large and small volumes (see, in particular, [5,6]). As is known, because of the small length of a de Broglie wave, the anomalous fluctuations of the order parameter are manifested in neutron experiments with the liquids in large volumes in states with thermodynamical parameters (temperature, density, concentration, etc.) that are far from the critical values. Although the role of fluctuations is not completely defining in small volumes of liquids, the features related to the fluctuation effects must be observed in the neutron experiments. So, from the features of the critical behavior of equilibrium and nonequilibrium properties

of the confined liquids (isothermal compressibility, diffusion coefficients, temperature conductivity, etc.) [1–3, 7–10], we have to expect noticeable temperature shifts of the maxima of coherent scattering and a change of the width of the diffusive peak of incoherent neutron scattering.

In the given paper, such important factor as the spatial heterogeneity of liquid near the critical point will be taken into account. Indeed, it is clear from the physical analysis that liquid in the critical region must be effected the Earth's gravitational field. The heterogeneity of liquid properties over the height that is caused by this factor is described in accordance with the "gravitation effect" theory (see review [11]). The reason for the gravitation effect is the matter distribution over the height of a chamber due to a growth of isothermal compressibility with approaching the critical point.

Thus, the study of features of the critical behavior of the density (in particular, the study of shifts of the critical temperature and other parameters) and the neutron refraction index of liquids on the basis of the scaling hypothesis for the finite-size systems with regard for the gravitation effect is the purpose of this work.

2. Neutrons Refraction Index in the Critical Region

We will consider the liquid volume geometry as: (1) a slit-like pore (plane-parallel layer) that has finite linear sizes $0 \leq x \leq L_1$, $0 \leq y \leq L_2$, $0 \leq z \leq L_3$; (2) a cylindrical pore of small radius R with the generatrix length L_3 ($0 \leq z \leq L_3$). The chambers of such geometries are often used in experiments with the near-critical liquid systems [12–14]. Hereafter, we will consider that $L_1 \ll L_2, L_3$, which means that the only one linear size along the x -axis not much exceeds the correlation length ξ of density fluctuations of liquid. This statement is also relevant to the cylinder radius in the second-type geometry ($R < L_3$). In the other words, the explored volume of liquid is reduced only in one direction in the case of a slit-like pore and in two directions in the case of a cylindrical pore.

We assume that the z -axis coincides with the direction of the Earth's gravitational field. Also we consider that the filling of the volume corresponds to the situation, where the critical density of a one-component liquid will be realized right in the middle of the chamber, i.e. at the level of $z = L_3/2$.

The problem of the distribution and scattering of neutrons in such confined heterogeneous liquid near the critical point with the application of the Fermi pseudopotential [15]

$$U(\vec{r}) = 2\pi\hbar^2 b_{\text{coh}}\rho(\vec{r})m^{-1}, \quad (1)$$

where \hbar — Planck constant, b_{coh} — coherent length (amplitude) of the neutron scattering, $\rho(\vec{r})$ — the local value of the numerical density of nuclei, and m — neutron mass, is reduced to solving the Schrödinger equation for the wave function of a neutron which adopts the form of an electro-dynamical wave equation [5]:

$$\psi(\vec{r}) + k_0^2 n^2(\vec{r})\psi(\vec{r}) = 4\pi b_{\text{coh}}\rho'(\vec{r})\psi(\vec{r}). \quad (2)$$

In (2), $\rho'(\vec{r})$ is the nucleus density fluctuation, $k_0 = 2\pi/\lambda$ is the wave vector, λ is the de Broglie wavelength that is within the range $4 \cdot 10^{-10} - 10^{-7}$ m for cold neutrons, and n is the analog of the optical refraction index that is determined by the relation

$$n(\vec{r}) = (1 - b_{\text{coh}}\rho_0(\vec{r})\lambda^2/\pi)^{1/2}. \quad (3)$$

The second term under the radical sign in (3) is usually small enough. Indeed, for the average numerical density of nuclei, we have $\rho_0 = \rho_c(1 + \Delta\rho)N_A/M$, where ρ_c is the critical volume density, N_A is the Avogadro constant, M is the molar mass, and $\Delta\rho = (\rho_0 - \rho_c)/\rho_c$ is a dimensionless deviation of the density from the critical value which is the order parameter for the liquid system (in the critical region, $\Delta\rho < 1$). Then, using the proper values of parameters for a specific substance (for example, for nitrogen N_2 , the coherent scattering length is $b_{\text{coh}}=1.87 \times 10^{-14}$ m, molar mass $M=28 \times 10^{-3}$ kg/mol, and critical density $\rho_c=0.304 \times 10^3$ kg/m³) under condition that the de Broglie wavelength $\lambda=9 \times 10^{-10}$ m (cold neutrons), one can get $s = b_{\text{coh}}\rho_c\lambda^2 N_A (\pi M)^{-1} \approx 3.2 \times 10^{-5}$ which characterizes the second term in (3). This estimation enables us to write the neutron refraction index near the critical point in the form

$$n = n_c - s\Delta\rho/2, \quad n_c = 1 - b_{\text{coh}}\rho_c\lambda^2 N_A (2\pi M)^{-1}, \quad (4)$$

where n_c is the critical value of the neutron refraction index [5]. The values of n_c for some substances are combined in Table 1. It is seen from this table that,

for the substances with the positive coherent scattering length b_{coh} (N_2 , CO_2 , C_6H_6 , and many others), the critical value of the neutron refraction index is slightly less than unity. Whereas for the substances with the negative b_{coh} (for example, H_2 , Li , H_2O), it slightly exceeds unity.

3. Height Density Profile in Confined Liquids

Hereafter, for the given geometry, we will take into account the action of gravitation by introducing a dimensionless height $\varsigma = \rho_c g(z - L_3/2)/P_c$ (g — acceleration due to the Earth's gravity, P_c — critical pressure). This height ς is counted from the level where the critical state of the unconfined liquid will be realized at the bulk critical temperature. The height ς will be assumed positive in the gas phase at $T > T_c$, i.e. for $z > L_3/2$, and will be assumed negative in the liquid phase at $z < L_3/2$.

The scaling theory of the gravitation effect for unconfined liquids (see, e.g., [16,17]) assumes that the height density profile is determined by such an expression:

$$\Delta\rho = h^{1/\delta} f_\rho(\tau/h^{1/\beta\delta}), \quad (5)$$

where $\tau = (T - T_c)/T_c$ is a dimensionless deviation of the temperature from the critical value, and $f_\rho(u)$ is a scale function with the asymptotics $f_\rho(u \rightarrow 0) = \text{const}$, $f_\rho(u \rightarrow \infty) \sim u^{-\gamma}$. Formula (4) along with the presented asymptotics gives such height density profile: a) $\Delta\rho \sim h^{1/\delta}$ at $u=0$, i.e. on the critical isotherm ($\tau=0$), b) $\Delta\rho \sim h/\tau^\gamma$ in the vicinity of the critical isochor ($h \ll \tau^{\beta\delta}$). The well-known relation for the critical indices $\beta + \gamma = \beta\delta$ is taken into account (the values of the critical indices for the liquid and other systems that are isomorphous to the 3-dimensional Ising model are as follows: $\beta \approx 0.325$, $\gamma \approx 1.24$, and $\delta \approx 4.8$).

According to the ideas which represent the basis of the static scaling hypothesis for the finite-size systems [1–3] in the case where the liquid has reduced geometry that is characterized by a linear size L , the equilibrium

Table 1. Critical values of the refraction index for cold neutrons ($\lambda = 9 \times 10^{-10}$ m)

Substance	Coherent dispersion length b_{coh} , m	Critical neutron refraction index $n_c - 1$
Nitrogen N_2	1.87×10^{-14}	-1.55×10^{-5}
Carbon dioxide CO_2	1.83×10^{-14}	-1.49×10^{-5}
Benzene C_6H_6	1.74×10^{-14}	-0.52×10^{-5}
Lithium Li	-0.19×10^{-14}	0.22×10^{-5}
Water H_2O	-0.17×10^{-14}	0.23×10^{-5}
Hydrogen H_2	-0.75×10^{-14}	0.89×10^{-5}

value of the order parameter of the liquid can be described by such an expression (see, e.g., [18]):

$$\Delta\rho = L^{-\beta/\nu} F(v, \omega), \quad (6)$$

where the scaling arguments $v = a\tau L^{1/\nu}$ and $\omega = bhL^{\beta\delta/\nu}$ determine the ratio of the linear size L of the system in the direction of spatial limitation to the correlation length ξ . In this case, the first argument v characterizes the temperature dependence of the correlation length $\xi \sim \tau^{-\nu}$ at $h \ll \tau^{\beta\delta}$, and the second argument ω defines the field dependence of the correlation length $\xi \sim h^{-\nu/\beta\delta}$ at $\tau \ll h^{1/\beta\delta}$. The constants a and b are nonuniversal, i.e. they are not identical for different liquids.

It is worth noting that the choice of the field variable h for the determination of the gravitation effect influence on the density profile of liquid near the critical point is not unique. In most theoretical and experimental papers (see references in review [11]) that are devoted to the research of the gravitation effect in liquids, the field variable h is selected as a height ς , which was introduced earlier. On other hand, in the fluctuation theory of phase transitions [19], the field h , which is conjugated to the order parameter $\Delta\rho$ in individual liquids, is introduced as a linear combination of deviations $\Delta\mu = (\mu - \mu_c)/\mu_c$ of the chemical potential and temperature τ from their critical values. That is, the field variable is selected in the form $h = a_1\Delta\mu + a_2\tau$. The third alternative for the field variable h was offered in [11] from the viewpoint of experimental results on the gravitation effect in liquids. In [11], it was considered to be reasonable to include, additionally to the height ς , some internal field in h which would be responsible for the interaction of density fluctuations.

According to the scaling hypothesis for the finite-size liquid systems [1–3], expression (5) can be rewritten by means of the equivalent expressions as

$$\Delta\rho = \tau^\beta F_1(v, \omega), \quad (7)$$

$$\Delta\rho = h^{1/\delta} F_2(v, \omega), \quad (8)$$

where the scale functions in Eqs. (6)–(8) are coupled between itself by such relations: $F_1 = (a/v)^\beta F$, $F_2 = (b/\omega)^{1/\delta} F$.

4. Height Profile Features of the Neutron Refraction Index

It is obvious that, according to Eq. (4), expressions (6)–(8) entirely sets the dependence of neutron refraction

index n on the temperature variable τ and the field variable h , as well as on the linear sizes of the finite-size liquid near the critical region.

In a bulk liquid, the neutron refraction index $n = n_c - s\Delta\rho/2$ (together with the order parameter $\Delta\rho$ of individual liquid) has a) the maximum of its temperature dependence on the coexistence curve of phases (CC) at the critical temperature ($T = T_c$) and b) the inflection point of its height dependence at the level with the critical density ($\rho = \rho_c$).

We will find the positions of the maximum and the inflection point in the temperature dependence and in the height dependence of the neutron refraction index and the order parameter for a spatially inhomogeneous liquid near the critical region in a small volume of the given geometry.

In order to determine the temperature which corresponds to the maximum points of the temperature dependences $n(\tau)$ and $\Delta\rho(\tau)$ on CC in the zero external field ($h=0, \omega=0$), we consider the following expression for the derivative that follows from Eqs. (4) and (7):

$$\begin{aligned} \partial n / \partial \tau = \\ = -(s/2) \left[\beta \tau^{\beta-1} F_1(v, 0) + \tau^\beta a L^{1/\nu} \partial F_1(v, 0) / \partial v \right]. \end{aligned} \quad (9)$$

For the temperature shift τ_m of the maximum of CC, this yields

$$\tau_m = -a_\tau L^{-1/\nu}, \quad (10)$$

where $a_\tau = \beta F_1 / a F_1'$. It becomes obvious from Eq. (10) that the maximum points of the temperature dependences of the neutron refraction index and order parameter of the liquid on CC will be realized at the bulk critical temperature $T = T_c$ ($\tau_m=0$) only in an unrestricted liquid, when $L \rightarrow \infty$. In the liquid with restricted geometry where the linear size of the system becomes less than the correlation length ($L \leq \xi$), this point must be observed at a smaller temperature $T_m < T_c$, because $\tau_m < 0$. The difference of temperatures $T_c - T_m$ grows with reduction of the linear size L : $(T_c - T_m) \sim L^{-1.6}$. Fig. 1 illustrates the derived temperature dependence of the neutron refraction index (in relative units $n^* = n2\rho_c/|s|$ at $s < 0$) on CC of bulk liquid (dotted line) and confined liquid (solid line).

Equation (10) for the position of a maximum as a function on the neutron refraction index $n(\tau)$ on CC is equivalent to the expression for a shift of the critical temperature $[T_c(\infty) - T_m(L)] \sim L^{-1/\nu}$ (that is known from the scaling hypothesis for finite-size systems [1–3]) to the region of smaller temperatures upon

the transition from the bulk systems to the systems with reduced geometry. Indeed, the temperature T_m , which corresponds to the maximum of CC in Fig. 1, is nothing but the critical temperature $T_c(L)$ for a confined liquid.

Therefore, it is interesting to compare the obtained results with the corresponding experimental data and results of computer simulation (see review [20]). In the reviewed works, the shifts of the critical temperature for liquids which are confined in cylinder and slit-like pores were determined by different experimental methods that are based on the observation of the temperature of disappearance of a) the jump on isochors and isotherms of adsorption, b) the hysteresis “adsorption-desorption”, and c) the coefficient of surface tension on the boundary of domains (see Table 2). The third column in Table 2 contains the data of theoretical computations of the critical temperature shift according to the expression

$$(\Delta T_c/T_c)_{\text{theor}} = (\mu_1 \xi_0/R)^{1/\nu} \quad (11)$$

which was obtained for cylindrical pores in [21]. In relation (11), $\mu_1=2.4048$ is the first zero of the Bessel function J_0 , and ξ_0 is the amplitude of the correlation length. From the data in Table 2, one can see that there is a good agreement between the theoretical and experimental results in a number of cases. However, there is a considerable disagreement of the data that were obtained experimentally by different methods for the same liquid in pores with approximately identical size. This indicates, on the one hand, the significant complication in conducting such experimental researches and, on the other hand, confirms the conclusion that there is no single critical temperature for the different physical properties of a definite confined liquid (see, e.g., [3,18]).

It is worth to mention that, in the computer experiment [20], there was a crossover in the dependence of the shift $\Delta T_c/T_c \sim L^{-1/\nu}$ of the critical temperature of water on the characteristic linear size L of the system. The linear size L is the radius R of a cylinder or the thickness H of a slit-like pore. It appears that, at $H < H_x$ for water in a finite volume, there is the dependence $\Delta T_c \sim L^{-1}$, which means that the critical index $\nu = 1$ (2-D critical behavior). At $H > H_x$, we observe the dependence $\Delta T_c \sim L^{-1.6}$ that corresponds to $\nu = 0.625$ (3D critical behavior). The value $H_x \approx 2.4$ nm is the crossover thickness of a slit-like pore which is approximately equivalent to 8 diameters of a water molecule. It should be noted that the calculations by expression (11) (see Table 2) were carried out for $\nu = 0.625$.

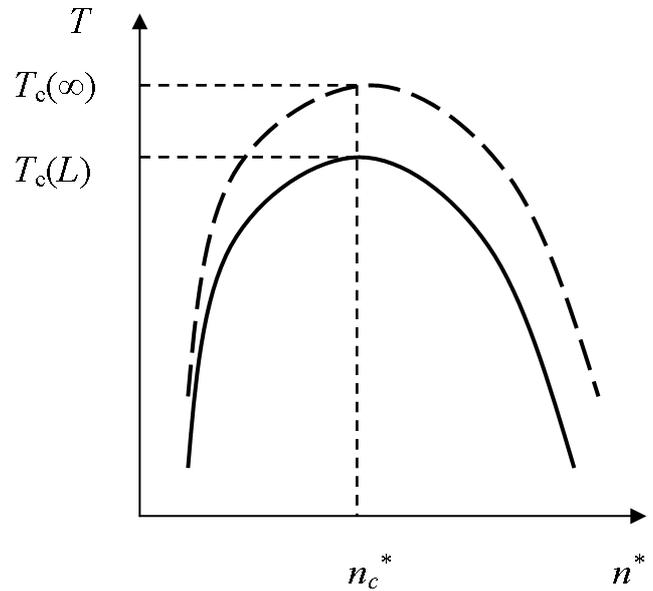


Fig. 1. Temperature dependences of the neutron refraction index n^* (in relative units $n^* = n2\rho_c/|s|$ at $s < 0$) on the coexistence curve of phases of the bulk liquid (dotted line) and confined liquid (solid line)

Like that a shift of the critical temperature was determined previously from the location of the maximum on CC, we now find the position of a inflection point h_{infl} on the field dependence of the neutron refraction index $n(h)$ and the field dependence of the order parameter $\Delta\rho(h)$ on the critical isotherm ($\tau=x=0$) in a confined liquid. For this purpose, we consider Eqs. (4) and (8) in order to calculate the derivative $\partial n/\partial h = -(s/2)\partial\Delta\rho/\partial h$:

$$\begin{aligned} \partial n/\partial h = & -(s/2) \left[\delta^{-1} h^{1/\delta-1} F_2(0, \omega) + \right. \\ & \left. + h^{1/\delta} b L^{\beta\delta/\nu} \partial F_2(0, \omega) / \partial \omega \right]. \end{aligned} \quad (12)$$

Table 2. Comparison of theoretical and experimental data for the shift of the critical temperature of liquids in cylindrical pores

Pore radius R , nm	$(\Delta T_c/T_c)_{\text{exp}}$	$(\Delta T_c/T_c)_{\text{theor}}$	Method of study
5.0	0.044	0.044	hysteresis [27]
11.0	0.033	0.013	hysteresis [29–32]
3.9	0.110	0.065	hysteresis [29–31]
3.9	0.047	0.065	adsorption curve [28,33]
12.1	0.029	0.011	adsorption curve [28,33]
15.7	0.0015	0.0071	adsorption curve [28,33]

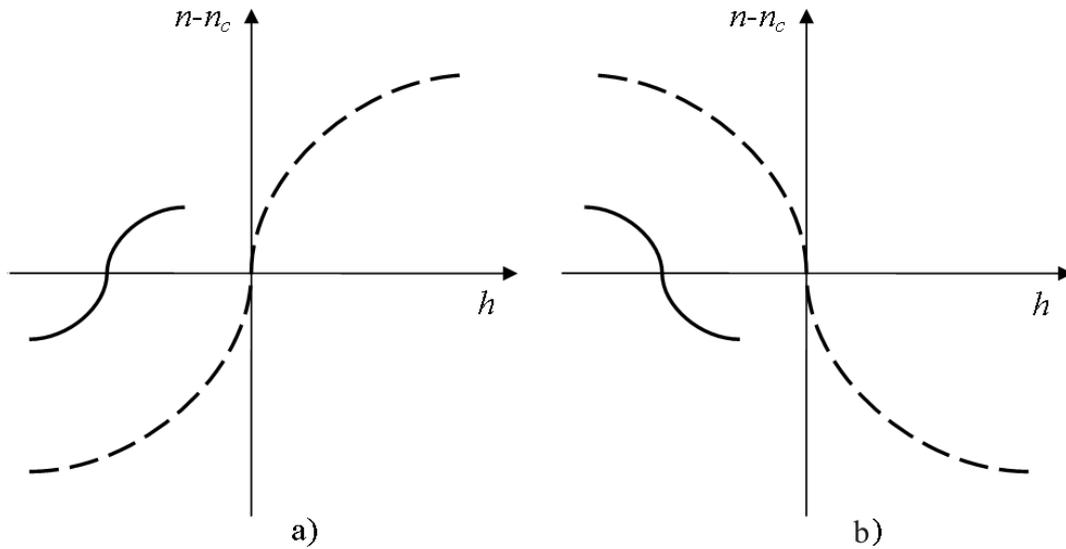


Fig. 2. Shift of the inflection point in the field dependence of the neutron refraction index on the critical isotherm. *a* — corresponds to the liquid system with positive coherent scattering length; *b* — corresponds to the case with $b_{\text{coh}} < 0$. The dotted lines describe the critical isotherms of the neutron refraction index in the bulk phase, while solid lines represent the dependence $n(h)$ in the confined liquid

Then the field variable h_{infl} at the inflection point is determined by the expression

$$h_{\text{infl}} = -b_h L^{-\beta\delta/\nu}, \tag{13}$$

where $b_h = F_2/\delta b F'_2$.

This situation is similar to the previous one, namely: as $L \rightarrow \infty$, i.e. for a unconfined (bulk) liquid, the inflection points in the dependence $n(h)$ and in the height density profile $\Delta\rho(h)$ are observed at the level $h = 0$, where the field variable conjugated to the order parameter is equal to zero. The nonzero and negative value of the field variable h_{infl} corresponds to the inflection point in the case of the reduced geometry of the liquid system. According to Eq. (13), the shift of the field variable for the liquid in the gravitational field means that the interface (meniscus) moves downward with respect to the level $h=0$. In this case, the modulus of this shift grows after a reduction of the system linear size L according to expression $h_{\text{infl}} \sim L^{-2.5}$. Obviously, the shift of the inflection point position h_{infl} in a finite-size liquid system has the same nature, as well as the above-mentioned shift of the critical temperature in confined liquids and the shift of the critical magnetic field $(H_c(\infty) + H_c(L)) \sim L^{-\beta\delta/\nu}$ in confined magnetics.

Fig. 2 illustrates a shift of the inflection point of the field dependence of the neutron refraction index on the critical isotherm. Thus, Fig. 2,*a* represents the case of

a liquid system with positive coherent scattering length, and Fig. 2,*b* shows the case with $b_{\text{coh}} < 0$.

In conclusion, we make a few important remarks.

1. In the wide vicinity of the critical point in respect to the temperature and field variables, the scaling corrections (see, e.g., reviews [22,23]) have to be accounted in addition to the basic asymptotic terms in Eqs. (5)–(8) for both the order parameter of a liquid and the neutron refraction index. For example, the density of coexisting phases on CC of individual liquids is described as

$$\rho_0 = \rho_c \pm B_0\tau^\beta \pm B_1\tau^\Delta + B_2\tau^{1-\alpha} + B_3\tau + \dots, \tag{14}$$

where B_i are system-dependent constants, $\Delta \approx 0.5$ is the critical index of the nonasymptotic correction [24]. The term with $\tau^{1-\alpha}$ gives, as is known, the singularity of the temperature derivative $d\rho_d/d\tau \sim \tau^{-\alpha}$ which demonstrates the near-critical behavior similar to that of isochoric heat capacity C_v , where $\rho_d = (\rho_{\text{liq}} + \rho_{\text{vap}})/2$ is the “diameter” of CC, and $\alpha \approx 0.11$ is the critical index.

2. Almost all results for the neutron refraction index, which were presented previously, are correct for the optical refraction index n_{opt} and for the dielectric permeability of a confined liquid. Indeed, the application of the Lorenz–Lorentz equation allows one to write the dielectric permeability as

$$\varepsilon(\vec{r}, \tau) = \varepsilon_c [1 + 3\varepsilon_c^{-1} r_c \rho_c \Delta\rho(\vec{r}, \tau)], \tag{15}$$

where $\varepsilon_c = 1 + 3r_c\rho_c$ is the critical value of the dielectric permeability, and r_c is the specific molecular refraction of the liquid system in the critical point. The practical similarity of Eqs. (15) and (4) for the neutron refraction index gives the background for the above-mentioned statement. At the same time, there are the definite distinctions in the critical behavior of the neutron refraction index and other physical values such as the dielectric permeability $\varepsilon(\vec{r}, \tau)$ and the optical refraction index $n_{\text{opt}} = \sqrt{\varepsilon}$ (at the magnetic permeability $\mu=1$). This distinction is determined by the condition that the specific molecular refraction r_c has a positive value in contrast to the coherent scattering length b_{coh} , which can be also negative.

3. As is known, the Lorenz–Lorentz equation does not take into account the contribution of fluctuation effects near the critical point. The same statement is correct in respect to Eq. (1) for the Fermi pseudopotential $U(r)$ and Eqs. (2), (3) for the neutron refraction index. The consideration of the binary and higher correlations of the order parameter (see reviews [17,25]) proves that the Lorenz–Lorentz equation acquires the additional contributions to the real part of the refraction index $\text{Re}(n)$ and to the imaginary part $\text{Im}(n)$ of 10^{-4} in the order of magnitude. This result can be spread on the neutron refraction index due to the absolute similarity of the methods of the account of correlation contributions in neutron optics and in liquids near the critical point. The imaginary part of the refraction index is determined by such expressions: a) in the region of weak spatial dispersion ($\xi q \ll 1$), $\text{Im}(n) \sim \int G_2(r) d\vec{r} \sim \beta_T$, where G_2 is the pair correlation function of density fluctuations, and β_T is the isothermal compressibility of liquid, b) in the region of strong spatial dispersion ($\xi q \gg 1$), $\text{Im}(n) \rightarrow \text{const}$, which means that the imaginary part of the refraction index does not become singular. From the physical reason, such a behavior of $\text{Im}(n)$ means that light and neutron beams will be weakened through the scattering in the liquid system while approaching its critical point. This statement is completely predictable and can be supported by the well-known optical theorem (see, e.g., [25,26]): $\sigma = 4\pi\text{Im}(n)/\lambda$, where σ is the extinction coefficient for light or the scattering cross-section for neutrons.

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НЕЙТРОННИЙ ПОКАЗНИК ЗАЛОМЛЕННЯ ОБМЕЖЕНИХ РІДИН В КРИТИЧНІЙ ОБЛАСТІ

К.О. Чалый

Р е з ю м е

Досліджено проблему профілю густини та нейтронного показника заломлення для неоднорідної рідини в обмеженій геометрії. Спеціальну увагу приділено впливу гравітації на властивості рідинних систем. Нові теоретичні результати отримано для зсуву критичної температури на кривій співіснування фаз та положення точок перегину на критичній ізотермі в обмежених рідинах.