
**LIQUID DISTRIBUTION IN A CONFINED SYSTEM
SUBJECTED TO A NONUNIFORM POTENTIAL****A.N. VASILEV,^{1,2} P.I. GORDIICHUK¹**¹**Taras Shevchenko National University of Kyiv***(2, Academician Glushkov Prosp., Kyiv 03022, Ukraine; e-mail: vasiliev@univ.kiev.ua)*²**National Technical University of Ukraine “KPI”***(16/2, Yangel’ Str., Kyiv 03056, Ukraine)*PACS 05.70.Fh, 05.70.Jk
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We calculate the density distribution profile of a spatially confined liquid system with cylindrical geometry under the action of a wall potential nonuniform along the cylinder axis. The problem is solved in the general case where the system is subjected to the action of an external field with radial symmetry. We also analyze a particular case of the wall potential of a special kind and perform numerical calculations.

1. Introduction

A significant breakthrough in the investigation of liquid systems, including the spatially confined ones, is related to the rapid development of computer technologies in the recent decades (see, e.g., [1–4]). The reason is simple and quite clear: a wealth of theoretical methods used for the investigation of liquid systems (though enriched with a number of rather efficient approaches and techniques) still can hardly be called sufficient. That is why a large number of practically important problems are solved with the use of computer calculations.

Among the most efficient computer methods successfully used for solving the problems of modeling and prediction of properties of liquid spatially confined systems, we can separate two fundamental directions or approaches. They are the direct computer modeling and the free-energy density-functional theory [5–11]. In the framework of the first approach, one carries out the imitation modeling of the behavior of a many-particle system. A characteristic feature of the given approach lies in the fact that a liquid is considered as an assembly of a large number of individual particles, whereas the statistical characteristics are calculated by averaging with

the use of the Monte-Carlo (MC) or molecular dynamics methods. In this field, the considerable success has been achieved; moreover, the majority of calculations allow one to consider the microstructure of a liquid substance in small pores, i.e. to perform studies on the level of atomic distribution functions ([12]). It is worth noting that, in spite of a certain specificity of the results obtained by the MC methods, they are extremely important, being directly applied in industry and allowing one to verify theoretically predicted effects. Another important field of application of the studies of liquids in small pores by the MC methods is the prediction and the research of various biophysical objects and systems. In this case, they allow one not only to obtain new interesting results, but sometimes to fill up certain gaps related to the impossibility of direct experimental measurements.

Another important direction of researches effectively developed during several recent decades is called the free-energy density-functional theory (DFT) and has several modifications depending on the object under study and the level of applied computer techniques. This method acquired a good reputation in the study of various systems and phenomena starting from quantum many-electron systems and finishing by the processes of sorption and the phase behavior of essentially heterogeneous liquids (see, e.g., [13–33]). As concerns the application of the DFT to the study of liquids (including those spatially confined in the presence of an external field), the basic assumption consists in the fact that a liquid represents a continuous medium described by the density distribution function. Respectively, the free energy of the system can be presented as a functional of the free energy density, which depends, in turn, on

the distribution density. That is why one of the problems successfully solved in the framework of the DFT approaches is the calculation of distribution profiles of liquids in nanopores. In spite of a considerable progress, a number of basic questions and problems still remain unsolved or little investigated. In particular, the numerical modeling (including the DFT-based one) gives rather good results for spatially confined microsystems, where dimensions of a system are only several-fold higher than those of substance's particles. An increase of this ratio (i.e. the transition to mesosystems) significantly complicates the numerical analysis at least due to the fact that the number of particles in a sample, over which one performs the statistical averaging, must essentially grow. Moreover, the high interest in these cases is attracted, first of all, by the smoothed density distribution function, where effects related to the finiteness of particles' sizes are smoothed over. Such functions can be restored on the basis of experimental data. For example, an important information on statistical and thermodynamic characteristics of liquid spatially confined systems can be obtained from experiments on neutron refraction [34–42]. In particular, it is known that the liquid density distribution in a cylindrical pore is essentially influenced by the wall potential [43–45], whereas the pattern of the distribution itself substantially depends on the temperature in the system. Of special interest is the behavior of the system in the neighborhood of a critical state. In spite of the complexity of this problem, it is rather theoretical in the sense that the use of the MC or molecular dynamics methods for its solving would allow one to obtain particular results, whereas it is desirable to have a general idea about the behavior of the system.

It is worth noting that, in a number of theoretical works, a similar problem of calculation of the density profile in a cylindrical system under the action of an external field (in particular, of the wall potential) was solved by solely analytical methods (see, e.g., [46–51]). In this case, it was considered that the wall potential is uniform along the cylinder axis. Naturally, it is not always true in practice. That is why the density distribution profile of a liquid is calculated in the given paper under the assumption of the nonuniformity of a wall potential. We use the approach proposed earlier in [46–50] and later developed in [51]. First, the problem is solved for a general external field characterized with radial symmetry and dependent on the coordinate along the cylinder axis. After that, we separately analyze the case where the wall potential represents a superposition of the basic potential uniform along the cylinder axis and a nonuniform addition. It is worth noting that the

methodology used in the work, though being rather simple, allowed one to obtain reliable results which were verified for a number of systems, in particular, on the basis of experiments on neutron scattering. In the case of systems uniform along the pore axis, the results obtained earlier with the help of this method are in good agreement with data of modeling by the MC methods (making allowance for the fact that all particles are considered to be point, that is why the calculated distribution function represents an analog of the smoothed density distribution function of finite-size particles). All these reasons give grounds to consider that the original results obtained for the critical behavior of a liquid in a pore subjected to a nonuniform wall potential are reliable as well.

2. System under Study

Let us consider a cylindrical pore of radius R filled with a one-component liquid. The liquid is subjected to the action of an external field $h(r, z)$, where r denotes the distance from the cylinder axis (i.e. $0 \leq r \leq R$), and z is the coordinate along the cylinder axis. Without loss of generality, we can consider that the field is periodic along the cylinder axis with period L , which means that $h(r, z + L) = h(r, z)$, $0 \leq z \leq L$.

Due to the action of the external field $h(r, z)$, the density distribution in the pore becomes nonuniform. Let us present the dependence of the liquid density $\rho(r, z)$ in the pore on the spatial coordinates in the form $\rho(r, z) = \rho_0 + \delta\rho(r, z)$, where ρ_0 denotes the liquid density in the absence of the field, and $\delta\rho(r, z)$ is a deviation of the density from the uniform distribution ρ_0 . The density ρ_0 corresponds to a minimum of the free energy functional in the absence of the field and, in particular, the wall potential. Here, one should also take into account that, in the framework of the proposed approach, the liquid particles are considered to be point, i.e. the liquid is considered as a continuum. Under such assumptions, the equilibrium distribution function of the liquid in the pore must evidently be constant, and the distribution is uniform.

In this case, the addition to the free energy (per a cell of the cylinder of length L) caused by the action of the field equals

$$\delta\Phi = \frac{1}{2} \int \left(a(\delta\rho)^2 + b(\nabla\delta\rho)^2 + 2h(r, z)\delta\rho \right) dV, \quad (1)$$

where a and b are the phenomenological parameters of the model, while the integration here and below is performed over the volume of a cylinder cell determined in

accordance with the period of the external-field potential (see Fig. 1). The problem is reduced to the search for a minimum of the functional under the additional condition

$$\int \delta\rho dV = 0 \quad (2)$$

which means the constancy of the total mass of a liquid in the system (see, e.g., ([46–50])). As a result, we obtain the equation

$$a\delta\rho - b\Delta\delta\rho = -(h(r, z) + \lambda), \quad (3)$$

where the unknown multiplier λ is sought from condition (2). In addition, it is necessary to satisfy the boundary conditions

$$\left. \frac{\partial\delta\rho}{\partial r} \right|_{r=R} = 0 \quad (4)$$

and the periodicity conditions

$$\delta\rho(r, z + L) = \delta\rho(r, z). \quad (5)$$

It is clear that the solution depends essentially on the form of the field $h(r, z)$. As was noted above, we will find firstly the general solution and after that analyze a particular case.

3. General Solution

In order to find the solution, the density deviation will be presented as

$$\delta\rho(r, z) = \xi(r, z) - \lambda/a. \quad (6)$$

After that, the parameter $\xi(r, z)$ can be obtained from the equation

$$a\xi - b\Delta\xi = -h(r, z) \quad (7)$$

with the boundary condition

$$\left. \frac{\partial\xi}{\partial r} \right|_{r=R} = 0 \quad (8)$$

and the periodicity condition

$$\xi(r, z + L) = \xi(r, z). \quad (9)$$

The parameter λ is determined by the relation

$$\lambda = \frac{2a}{R^2L} \int_0^L dz \int_0^R \xi(r, z) r dr. \quad (10)$$

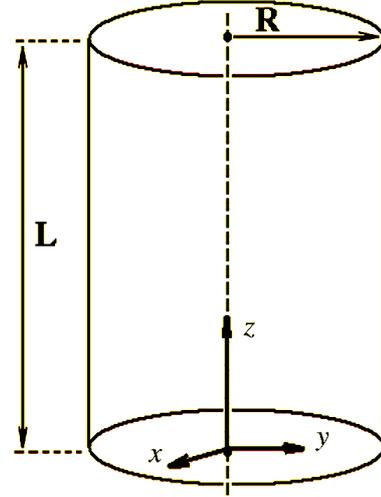


Fig. 1. Geometry of the investigated system: R is the cylinder radius, L is the field period along the cylinder axis

Thus, in order to obtain the general solution, it is necessary to find the function $\xi(r, z)$.

Taking the periodicity condition (9) into account, the required function $\xi(r, z)$ can be presented as the series

$$\xi(r, z) = \sum_{m=0}^{\infty} \xi_m^{(1)}(r) \cos\left(\frac{2\pi mz}{L}\right) + \xi_m^{(2)}(r) \sin\left(\frac{2\pi mz}{L}\right), \quad (11)$$

where the expansion coefficients $\xi_m^{(1,2)}(r)$ satisfy the equation

$$\left(a + \frac{4\pi^2 m^2}{L^2} b\right) \xi_m^{(k)} - b\Delta_r \xi_m^{(k)} = -h_m^{(k)}(r). \quad (12)$$

Here, $k = 1, 2$, $\Delta_r = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \right)$, while the expansion coefficients

$$h_m^{(k)}(r) = \frac{2 - \delta_{0m}}{L} \int_0^L h(r, z) \cos\left(\frac{2\pi mz}{L} - \frac{\pi(k-1)z}{2}\right) dz, \quad (13)$$

where δ_{ij} is the Kronecker symbol. In addition, the following boundary conditions must be satisfied:

$$\left. \frac{\partial\xi_m^{(k)}}{\partial r} \right|_{r=R} = 0. \quad (14)$$

The parameter λ is determined on the basis of the expansion coefficient $\xi_0^{(1)}(r)$ and, therefore, depends on the field $h(r, z)$ as

$$\lambda = -\frac{2}{R^2L} \int_0^L dz \int_0^R h(r, z) r dr. \quad (15)$$

The solution for the expansion coefficients $\xi_m^{(1,2)}(r)$ can be sought in the form of the series ($k = 1, 2$)

$$\xi_m^{(k)}(r) = \sum_{n=0}^{\infty} \xi_{m,n}^{(k)} J_0\left(\frac{\mu_n r}{R}\right), \quad (16)$$

where $J_0(u)$ are the zero-index Bessel functions, the eigenvalues μ_n are the zeros of the first-index Bessel function, i.e. $J_1(\mu_n) = 0$ (here, we consider $\mu_0 = 0$). It is easy to show that the expansion coefficients $\xi_{m,n}^{(k)}$ are determined by the relation

$$\xi_{m,n}^{(k)} = -\frac{h_{m,n}^{(k)}}{b\left(a/b + (2\pi m/L)^2 + (\mu_n/R)^2\right)}, \quad (17)$$

where the expansion parameters of the field $h_{m,n}^{(k)}$ ($k = 1, 2$) have the following form:

$$h_{m,n}^{(k)} = \frac{2}{R^2 J_0^2(\mu_n)} \int_0^R h_m^{(k)}(r) J_0\left(\frac{\mu_n r}{R}\right) r dr. \quad (18)$$

Relations (6), (11), (13), and (15)–(18) determine the solution of the problem. In particular, the density $\delta\rho(r, z)$ of the liquid distribution in the pore (in view of the facts that $h_{0,0}^{(1)} = -\lambda$ and $\xi_{0,0}^{(1)} = -h_{0,0}^{(1)}/a$) is given by the expression

$$\begin{aligned} \delta\rho(r, z) = & \\ = \sum_{k=0}^1 \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \xi_{m,n}^{(k+1)} J_0\left(\frac{\mu_n r}{R}\right) \cos\left(\frac{2\pi m z}{L} - \frac{\pi k}{2}\right), \quad (19) \end{aligned}$$

where the parameters $\xi_{m,n}^{(k)}$ are calculated according to the above-presented relation (17).

Let us analyze the obtained result for a particular case of the wall potential of a special kind.

4. Wall Potential with a Characteristic Periodicity Scale

In the general case, the wall potential can be presented as a superposition of space harmonics. Let us consider the situation where one harmonic is dominant among such a set (except for the harmonic corresponding to the field uniform along the cylinder axis). The characteristic nonuniformity scale corresponding to this harmonic will be denoted by L . For the sake of simplicity, we assume that the potential $h(r, z)$ is described by the dependence

$$h(r, z) = U(r) \left(1 + \varepsilon \cos\left(\frac{2\pi z}{L}\right)\right), \quad (20)$$

where ε denotes the nonuniformity degree of the potential, while the function $U(r) = U_0$ at $R - d \leq r \leq R$ and $U(r) = 0$ at $r < R - d$, which corresponds to the potential in the form of a “step” of thickness d .

For the wall potential of such a form, some of the above-presented relations become essentially simpler. In particular, the parameter λ takes the form

$$\lambda = -\frac{U_0 d}{R} \left(2 - \frac{d}{R}\right), \quad (21)$$

the solution for the parameter $\xi(r, z)$ is given by the relation

$$\xi(r, z) = \xi_1(r) + \xi_2(r) \cos\left(\frac{2\pi z}{L}\right). \quad (22)$$

Moreover,

$$\xi_1(r) = -\frac{1}{b} \sum_{n=0}^{\infty} \frac{h_n}{a/b + (\mu_n/R)^2} J_0\left(\frac{\mu_n r}{R}\right), \quad (23)$$

$$\xi_2(r) = -\frac{\varepsilon}{b} \sum_{n=0}^{\infty} \frac{h_n}{a/b + (2\pi/L)^2 + (\mu_n/R)^2} J_0\left(\frac{\mu_n r}{R}\right), \quad (24)$$

and, for $n > 0$,

$$\begin{aligned} h_n = & \frac{2U_0}{R^2 J_0^2(\mu_n)} \int_{R-d}^R J_0\left(\frac{\mu_n r}{R}\right) r dr = \\ = & -\frac{2U_0(1-d/R)J_1\left(\left(1-\frac{d}{R}\right)\mu_n\right)}{\mu_n J_0^2(\mu_n)}. \quad (25) \end{aligned}$$

Figure 2 shows the parameter $\Delta n = b\delta\rho/U_0$ as a function of the space coordinates for the values $\varepsilon = 0.1$, $d/R = 0.05$, $L/R = 10$, and $a/b = 1$.

The parameter $a/b \equiv \kappa^2$ has the dimension of inverse squared distance (in this case, the correlation radius $R_C = |\kappa|^{-1}$ [52]). According to the generally accepted assumption, this parameter depends on the temperature. Moreover, after the current ideas of the scale invariance theory (scaling), this dependence is power (see, e.g., [52–54]). Particularly, if we denote the dimensionless temperature by $\tau = (T - T_C)/T_C$ (T is the temperature, and T_C is its critical value), then we can put down $\kappa^2 = \kappa_0^2 \tau^{2\nu}$, where κ_0 is the inverse amplitude of the correlation radius and ν is the critical index (for a wide class of liquids, $\nu \approx 0.63$) [52]. Thus, with varying temperature, the correlation radius R_C of the system

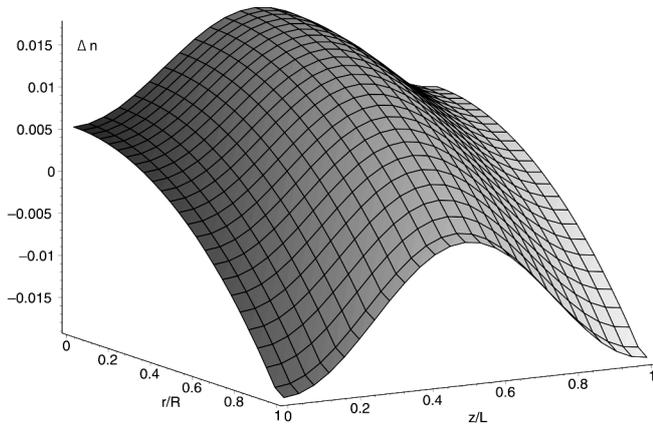


Fig. 2. Parameter $\Delta n = b\delta\rho/U_0$ as a function of space coordinates

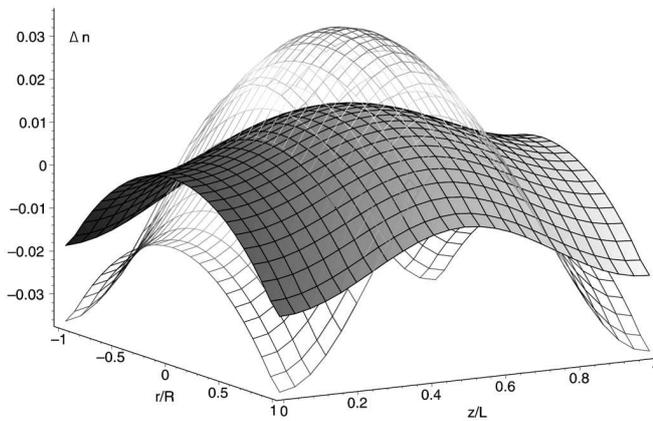


Fig. 3. Parameter $\Delta n = b\delta\rho/U_0$ as a function of space coordinates at the temperatures $\tau = 1$ (solid surface) and $\tau = 0.1$ (transparent surface)

changes within wide limits. On the other hand, the correlation radius represents a characteristic scale, to which other characteristic space parameters of the system are compared — in the given case, they are the pore radius and the size of the spatial nonuniformity mode of the wall potential.

That is why it is not surprising that a change of the temperature essentially affects the pattern of the liquid distribution in the pore via a change of the correlation radius. For comparison, Fig. 3 presents the distribution surfaces of Δn for the dimensionless temperatures $\tau = 1$ and $\tau = 0.1$.

As the system approaches the critical state (the parameter κ^2 tends to zero), the effect of the wall potential on the nonuniformity of the density distribution increases. This situation can be examined in more details with the help of the contour graphs in Figs. 4–6. They present the illustrations for the profiles of the density

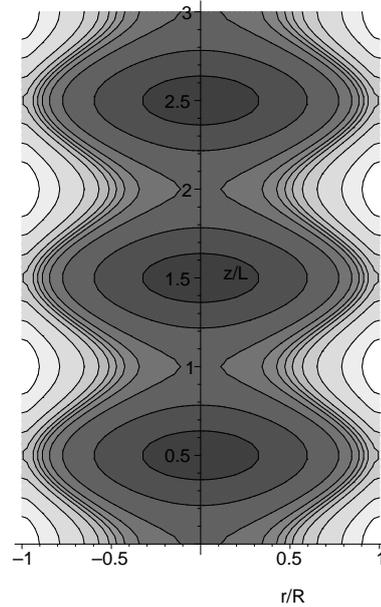


Fig. 4. Contour graph for the dependence of the liquid density profile (parameter $\Delta n = b\delta\rho/U_0$) in a pore on space coordinates at the temperature $\tau = 1$. Contour lines are presented for $\Delta n = 0, \pm 0.001, \pm 0.0025, \pm 0.005, \pm 0.01, \pm 0.015, \pm 0.02$

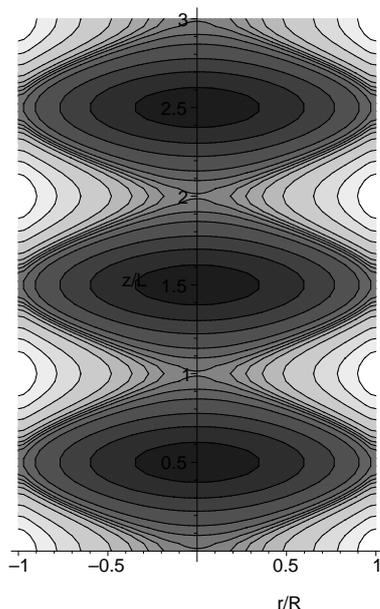
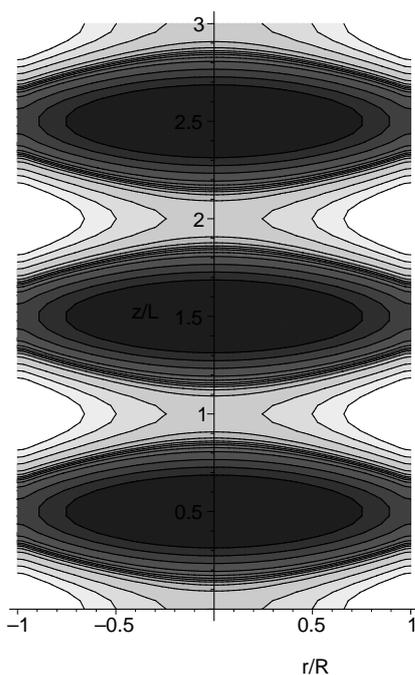
distributions at $\tau = 1$ (Fig. 4), $\tau = 0.5$ (see Fig. 5), and $\tau = 0.1$ (see Fig. 6).

For convenience, the graphs present three adjoining cells of the system under study. In such a form, it is easier to imagine the pattern of a liquid distribution in the whole system with regard for its periodicity.

The effect of the temperature variation is quite expected: in a state close to the critical one, a liquid becomes more “structured,” and the distribution extrema are sharper as compared with the case where the system is beyond the limits of the critical state ($\tau \sim 1$). In the given case, the effect of the temperature variation is estimated quantitatively.

5. Conclusions

The critical behavior of liquid systems in small volumes represents a perspective rather complicated problem (see, e.g., [55–59]). That is why, in spite of a considerable many years’ interest of researchers, there still remain a number of unsolved problems. Among them, there is the problem of the filling of low-dimension systems with fluids. For this purpose, the liquid-filler is usually converted to the critical or overcritical state. On the other hand, it is known that a liquid in the near-critical state is characterized by anomalous susceptibility. Particularly, as was shown above, there can arise a problem

Fig. 5. The same as in Fig. 4 at $\tau = 0.5$ Fig. 6. The same as in Fig. 4 at $\tau = 0.1$

related to the nonuniformity of the liquid distribution (along a pore) caused by an even insignificant nonuniformity of the wall potential. That is why the obtained results can be also useful for the prediction of the properties of porous and low-dimension liquid-filled systems and, in particular, for the determination of the bound-

ary values of nonuniformities of the wall potentials in the corresponding pores.

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РОЗПОДІЛ РІДИНИ В ОБМЕЖЕНІЙ СИСТЕМІ ПІД ДІЄЮ НЕОДНОРІДНОГО ПОТЕНЦІАЛУ

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Резюме

В статті розраховано профіль розподілу густини просторово обмеженої рідкої системи з геометрією циліндра за умови дії пристінкового потенціалу, неоднорідного вздовж напрямку циліндра. Задачу розв'язано в загальному випадку, коли система знаходиться під дією зовнішнього поля з радіальною симетрією, проаналізовано частинний випадок пристінкового потенціалу спеціального типу та проведено числові розрахунки.