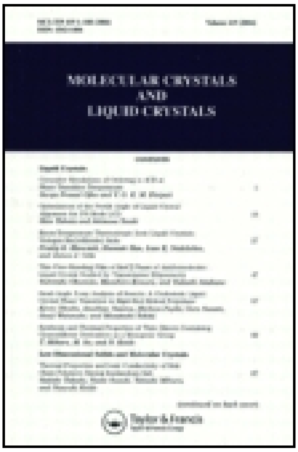

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Statistical-Mechanical Theory of Systems Composed of Interacting Ellipsoidal Molecules at a Solid Surface

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Statistical-Mechanical Theory of Systems Composed of Interacting Ellipsoidal Molecules at a Solid Surface

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A statistical-mechanical theory which takes into account translational-translational, orientational-orientational and mixed correlations based upon the method of conditional distribution functions is applied to calculations of the order parameters and the rotational viscosity coefficient (RVC) of a system composed of interacting ellipsoidal molecules near an interacting wall. The calculations have been carried out for cubic close packing with the nearest-neighbor Gay-Berne intermolecular potential and with a (9-3) like orientation-dependent molecule-wall interaction. The theory shows that the number of surface layers which must be taken into account depends more on the character of intercell correlations in the system than on the direct wall-nematic interaction. The equilibrium order parameters and rotational viscosity coefficient at the solid-nematic fluid interface have been calculated.

Keywords: Conditional distribution functions, nonlinear integral equations.

I. INTRODUCTION

A solid crystalline surface would be able to induce the additional ordering or disordering in the liquid crystals (LCs) which close to the one and thereby to effect on the macroscopic characteristics, such as a viscosity and elasticity.

Recently a kind of Bethe theory, which takes account of intercell pair correlations for describing a molecular systems composed of interacting ellipsoidal molecules near an interacting wall, has been proposed.[1,2] In the framework of that theory, which is based on the concept of average force potentials,[3,4] the influence of the interacting wall on the order parameter and surface free energy has been calculated. However, in,[1] the influence of the wall was considered as a small perturbations on the bulk values of the average force potentials, which is not likely to be valid in the case of a strong wall-molecule interaction.

In the present paper, I make an attempt to combine the previous[1,2] advantages of integral equation theory and of the cell model approach to construct a new statistical-mechanical theory for calculating equilibrium properties of NLCs and the rotational viscosity coefficient (RVC) near an interacting wall, which will be applicable to the case of a strong wall-molecule interaction.

This paper was presented at the XVth International Liquid Crystal Conference, Budapest, 3–8 July 1994.

The plan of this paper is as follows. A description of the model is given in section 2, the method of solving the resulting system of nonlinear integral equations is presented in section 3, the intermolecular potential is described in section 4, results of the numerical calculations are given in section 5, and finally I summarize my main results and conclusions in section 6.

II. MODEL

I consider a one-component fluid consisting of molecules with coordinates $i \equiv (\mathbf{q}_i, \mathbf{e}_i)$ describing molecular positions $\mathbf{q}_i$ and orientations $\mathbf{e}_i(|\mathbf{e}_i| = 1)$ of prolate ellipsoids of revolution, of major semiaxis $\sigma_\parallel$ and minor semiaxis $\sigma_\perp$, respectively, in contact with a solid planar wall. The x-axis of the coordinate frame is chosen to be normal to the wall, and the nematic molecules occupy the half-space $x \geq 0$. The space-fixed z-axis is chosen so that the nematic director lies in the yz plane.

One uses a statistical scheme in which the total volume V of the system of N molecules is divided into n molecular layers parallel to the solid wall. In layer L there are N_L cells, so that $N = \Sigma N_L$, and the volume of each cell is $v = V/N$. The potential energy associated with the N particles is a sum of potentials $U = \sum_{i \neq j} \Phi(ij) + \Phi(i)$, where $\Phi(ij)$ is the intermolecular pair potential, and $\Phi(i)$ is the molecule-wall potential. By integration of the Gibbs canonical distribution, a set of functions $F_L(i)$. $F_{LL}(ij)$, $F_{LK}(ij)$ etc., is introduced in order to define the probability densities for the molecules to be found about the positions $m = W_m$ ($W_m = v_m \otimes \alpha_m$, where α_m is the volume associated with orientations, $m = i, j$).[1, 3] Using mean force potentials (MFPs), I can present the above mentioned functions in the form

$$F_L(i) = Q_L^{-1} \exp\{-\beta \varphi_L(i)\}, \tag{1}$$

$$F_{LL}(ij) = Q_{LL}^{-2} \exp\{-\beta[\Phi(ij) + \varphi_{LL}(ij)]\}, \tag{2}$$

$$F_{LK}(ij) = \{Q_L * Q_K\}^{-1} \exp\{-\beta[\Phi(ij) + \varphi_{LK}(ij)]\}, \tag{3}$$

where L, K are the layer numbers, respectively, and

$$Q_L = \int_i d(i) \exp\{-\beta \varphi_L(i)\}, \quad \int_i d(i) \equiv \int_{w_i} d\mathbf{q}_i d\mathbf{e}_i.$$

Here $\beta \equiv (kT)^{-1}$ is the inverse temperature, and $\varphi_L(i)$, $\varphi_{LK}(ij)$ and $\varphi_{LL}(ij)$ are the single and binary mean force potentials belonging to the layers L and K, respectively. These are sums of the form

$$\varphi_L(i) = \sum_{j \neq i} \varphi_{i,j}(i), \quad i \in L, \tag{4}$$

$$\varphi_{LL}(ij) = \sum_{l \neq ij} \varphi_{ij,l}(ij), \quad i \in L, \quad j \in L, \tag{5}$$

$$\varphi_{LK}(ij) = \sum_{l \neq ij} \varphi_{ij,l}(ij), \quad j \in K, \quad i \in L, \tag{6}$$

Here $\varphi_{i,j}(i)$ and $\varphi_{ij,l}(ij)$ are the mean force potentials which are defined in Equations (4–5) of.[1] The quantities $\nabla_i \varphi_{i,j}(i)$ and $\nabla_i \varphi_{ij,l}(ij)$ are the mean forces acting on a molecule in the i^{th} cell due to the molecule in the l^{th} cell, averaged over the states of the latter. The subscripts before the comma correspond to the MFP dependence on the positions of the molecules; those after the comma correspond to the average states. Using now the relations between the single and binary functions which follow from their definition, one has

$$\int_i d(i) F_K(i) = 1, \quad \int_i d(j) F_{LK}(ij) = F_K(i), \quad j \in L. \tag{7}$$

The two-particle function is similarly related to a three-particle function by an integral relation, etc. Below I take into account only the first two functions of the infinite hierarchy; this corresponds to considering only pair correlations between cells. In order to make this a closed system of equations, I separate the mean force potentials into irreducible parts.[2] In the approximation considered here, which corresponds to neglecting three-cell and higher-order correlations, I have

$$\varphi_{ij,l}(ij) = \varphi_{i,l}(i) + \varphi_{j,l}(j), \tag{8}$$

and the expressions for the binary functions take the forms

$$F_{LK}(ij) = F_L(i) * F_K(j) * \exp\{\beta[\varphi_{i,j}(i) + \varphi_{j,i}(j)]\} * V(ij), \quad \text{for} \quad i \in L, \quad j \in K. \tag{9}$$

$$F_{LL}(ij) = F_L(i) F_L(j) * \exp\{\beta[\varphi_{i,j}(i) + \varphi_{j,i}(j)]\} * V(ij), \quad \text{for} \quad i \in L, \quad j \in L. \tag{10}$$

where $V(ij) = \exp\{-\beta\Phi(ij)\}$. The exponential factors in Equations (9–10) reflect the correlations between cells and distinguishes the approach used here from the mean field approximation. Substitution of Equations (9 and 10) into Equation (7) leads to closed system of nonlinear integral equations (NIEs) for the MFPs,[1,2]

$$\exp\{-\beta\varphi_{i,j}(i)\} = \int_j d(j) \exp\{\beta\varphi_{j,i}(j)\} * V(ij) * F_M(j), \quad M = L, K. \tag{11}$$

Since $\varphi_{i,j}(i)$, $\varphi_{j,i}(j)$ and $F_M(j)$ are related through Equations (1), (4–6), (8), Equation (11) provides a system of nonlinear integral equations for the mean force potentials. Knowing the solution of these equations, one can compute the microscopic characteristics of the LC system (expressed in terms of the one-particle and two-particle functions) and also the macroscopic characteristics, which can be expressed in terms of the free energy of the system. The Helmholtz free energy per molecule is given by,[2,3]

$$f = \sum_L f_L, \quad f_L = -\beta^{-1} \ln \int_i d(i) \exp\{-\beta\varphi_L(i)\}. \tag{12}$$

It should be noted that in the case of a homogeneous system, the MFPs $\varphi_{i,j}(i)$, $\varphi_{j,i}(j)$ do not depend on the positions of the cells i and j. This means that one reduces the system of NIE (11) to a single equation. Note that the same system of equations appear in the Bethe approximation for a lattice model of surfactant mixtures in[5] (see Eq. (12)).

Theoretically, of course, the system of Equation (11) is infinite, but in practice only a finite number of adjacent layers can be taken into account. It will be shown that the MFPs decay to their bulk values. The decay length, or the number of layers in which the MFPs are different from the ones in the bulk, depends strongly on the pairwise interaction and correlations between the cells. In this case, the system of Equation (11) must be truncated after a finite number of perturbed layers. Accordingly, one arrives at the finite system of NIEs

$$\left\{\begin{array}{ll} \psi_{1,k}(i)=\int_j d(j)\,\tilde{V}(ij)\,\psi_{1,k}^{-1}(j)\,F_1(j), & k=0,1 \\ \psi_{1,2}(i)=\int_j d(j)\,V(ij)\,\psi_{2,1}^{-1}(j)\,F_1(j), & \\ \vdots & \vdots \\ \psi_{n,m}(i)=\int_j d(j)\,V(ij)\,\psi_{m,n}^{-1}(j)\,F_n(j), & m=n-1, \\ \psi_{n,n}(i)=\int_j d(j)\,V(ij)\,\psi_{n,n}^{-1}(j)\,F_n(j), & \end{array}\right. \tag{13}$$

where $\psi \equiv \exp\{-\beta\varphi\}$, and

$$\tilde{V}=\left\{\begin{array}{ll} V_w(i), & \text{for} \quad k=0 \\ V(ij), & \text{for} \quad k=1 \end{array}\right.$$

and $V_w(i)$ is the kernal of the integral equation determined by the wall-molecule interaction potential which I shall identify shortly, and

$$F_L(i)=Q_L^{-1} * \prod_{j\neq i}\psi_{i,j}(i), \quad i\in L, \tag{14}$$

$$F_n(i)=Q_n^{-1} * \left\{\prod_{j\neq i}\psi_{i,j}(i)\right\} * \psi_{i,j}^{b}(i) \quad i\in n, \tag{15}$$

where $\psi_{i,j}^{b}(i)$ is the bulk MFP. It should be noted that in the present paper I have considered only a simple cubic structure where every cell has six neighbors.

III. METHOD OF SOLVING THE SYSTEM OF NONLINEAR INTEGRAL EQUATIONS

The method of solution of the five-dimensional problem (13) is very complicated and there are no good general methods for solving systems of more than one nonlinear equation. Except in linear problems, obtaining solutions invariably proceeds by numerical iteration by the method

$$\psi^{(k+1)}(P_i)=\{\psi^{(k)}(P_i)\,\hat{L} * \psi^{(k)}(P_i)\}^{1/2}, \tag{16}$$

where $\|\hat{L}\| \leq 1$ is the n-dimensional nonlinear integral operator defined in Equation (13) in the space $W_i = v_i \otimes \alpha_i$, and P_i is n-dimensional vector in that space. For smoothly varying functions, good algorithms will always converge, provided that the initial guess is good enough. Success strongly depends on having a good initial guess for the solution $\psi(i)$, and from that point of view it is convenient to chose $\psi^{(0)}(i) = \psi^b(i)$. The procedure of construction of the bulk MFPs $\psi^b(i)$ for the nematic phase was described in.[1,2] The algorithm (16) was implemented as follows: the initial approximation was set to be $\psi^{(0)}_{1,k}(P_i) = \psi^{(0)}_{1,2}(P_i) = \cdots = \psi^{(0)}_{n,n}(P_i) = \psi^b(P_i)$. Then the integrals of the right-hand side of (16) were calculated with the help of the Sobol's method,[6] using the Haar functions for calculating the multi-dimensional integrals as

$$\int_0^1 dx_1 \cdots \int_0^1 dx_n f(x_1 \cdots x_n) \approx \frac{1}{M} \sum_{\alpha=0}^{M-1} f(P_\alpha). \tag{17}$$

Here P'_α are the points uniformly (*mod* 1) distributed in the n-dimensional unit cube. These points belong to a LP_τ sequence[7] and was generated by means of LP_τ-generator,[6] and M is the number of points. The precision of the calculations is about $R = O(M^{-1} \ln^n M)$, where n is the dimensionality of space, whereas for Monte-Carlo integration the analogous estimate is $\sim O(1/\sqrt{M})$.

The procedure of calculations was repeated for all neighbors of the cells in the first n layers. The coordinates P_i in Equation (16) were chosen such that $\psi^{(1)}(P_i)$ could be calculated at the points forming the same stationary LP_τ sequence as used in the evaluation of the integrals with the initial approximation $\psi^{(0)}(P_i)$. Then $F^{(1)}_L(P_i)$ was calculated by a simple multiplication of $\psi^{(1)}(P_i)$. The procedure was then iterated until a given accuracy was achieved. The calculations were executed using $M = 200$, which corresponds to 200^2 points of a Monte Carlo method for the same estimates of the integrals. This has been achieved owing to fact that the LP_τ-sequences possess a more uniformly distributed set of M points in the n-dimensional unit cube than the independent random sequences.

IV. MODEL OF MOLECULAR INTERACTIONS

The kernel of the integral Equation (13) is determined by the molecular interaction potential. This was chosen to be the Gay-Berne[8] intermolecular potential

$$\Phi(\mathbf{q}_{ij}, \mathbf{e}_i, \mathbf{e}_j) = 4\varepsilon_0 \varepsilon(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij}) \left\{ \left[\frac{\sigma_0}{\mathbf{q}_{ij} - \sigma(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij}) + \sigma_0} \right]^{12} - \left[\frac{\sigma_0}{\mathbf{q}_{ij} - \sigma(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij}) + \sigma_0} \right]^{6} \right\}, \tag{18}$$

where $\mathbf{e}_i$, $\mathbf{e}_j$ are, respectively, unit vectors along the molecular symmetry axes, $\mathbf{q}_{ij}$ the vector between the molecular centers, and $\mathbf{e}_{ij} = \mathbf{q}_{ij}/|\mathbf{q}_{ij}|$. The potential energy parameter $\varepsilon(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij})$ and size parameter $\sigma(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij})$ are angle-dependent functions defined in Equations (3), (4) and (8)–(10), respectively, of.[8] The former depends on the molecular elongation or length-to-breadth ratio γ (denoted $\sigma_{\|}/\sigma_{\perp}$ in,[8] while $\varepsilon(\mathbf{e}_i, \mathbf{e}_j, \mathbf{e}_{ij})$ depends on both γ and another parameter which can be used to adjust the ratio between the end-to-end and side-by-side well depth, denoted $\varepsilon_l/\varepsilon_s$. In our calculations the parameters μ and v of[8] have been fixed at the values 1 and 2, respectively. The molecule-wall interaction is given by[9]

$$\Phi(\mathbf{e}_i, x_i) = \frac{2\pi}{3}\frac{\varepsilon_w}{\sigma_w^2}\left[\frac{2}{15}\left(\frac{\sigma_0}{\sigma_w x_i}\right)^9 - \left(\frac{\sigma_0}{\sigma_w x_i}\right)^3\right]. \tag{19}$$

The parameters in this potential are also orientation dependent, and are given by

$$\varepsilon_w = \varepsilon_{ow}(1 - \chi^2 e_{i,z}^2)^{-1/2}, \tag{20}$$

$$\sigma_w = \left(1 - \frac{\chi e_{i,x}^2}{1 - \chi^2 e_{i,z}^2}\right)^{-1/2}, \tag{21}$$

where $\chi^2 = (1 - \gamma^2)/(1 + \gamma^2)$, $\varepsilon_{ow} = \varepsilon\varepsilon_o$, and ε is a strength parameter, x_i denote the distance from the wall to the molecule i, and $\mathbf{e}_i = (e_{i,x}, e_{i,y}, e_{i,z})$.

V. RESULTS OF THE NUMERICAL CALCULATIONS

Within the framework of the theory, which is based upon the random walk theory[10] together with some ideas of Zubarev's nonequilibrium statistical operator[11] one can write the expression for rotational viscosity coefficient γ_1 as a function of temperature, molecule moment of inertia and set of the NLC order parameters[2]

$$\gamma_1 \Big/ \left\{\frac{\sqrt{\varepsilon_0 I}}{\sigma_0^3}\right\} = \frac{3\sqrt{2\pi}}{\pi^3} * \frac{\beta^{-1/2}}{v} * \frac{2f(\eta_2(L))}{1 - \frac{5}{2}\eta_2(L) + \frac{27}{8}\eta_4(L) - \frac{65}{16}\eta_6(L)} \tag{22}$$

where

$$\eta_k(L) = \langle P_k^{(L)}(\cos\theta_i)\rangle = \int_i d(i)\, P_k(\cos\theta_i)\, F_L(i), \tag{23}$$

and $P_k(\cos\theta_i)$ is the Legendre polynomial of order k, and $F_L(i)$ is the single function corresponding to the L^{th} layer, satisfying the system of NIE (13), and

$$f(\eta_2(L)) = \frac{(3.181 + 0.757\,\eta_2(L))\,\eta_2^2(L)}{2.881 + \eta_2(L) + 12.36\eta_2^2(L) + 4.69\eta_2^3(L) - 0.743\eta_2^4(L)}.$$

A final representation (22) imply a knowledge a set of the NLC parameters $\eta_k(L)$. Our system is characterized by reduced parameters: the reduced volume $v^* = v/\sigma_0^3$, the reduced temperature $\beta^{-1} \equiv k\,T/\varepsilon_o$, and anisotropy parameter γ. One can readily see from Figure 1 that the number of perturbed layers for the case of a strong interaction between the solid wall and nematic phase ($\varepsilon_{ow} = 5 * \varepsilon_o$) (Fig. 1, curves 1, 2, 3) and in the case of weak interaction ($\varepsilon_{ow} = 0.5 * \varepsilon_o$) (Fig. 1, curve 4) are the same and equal approximately four. The direct interaction has a strong influence on the behavior of the molecules only in the layer nearest to the wall. It is seen that the OP $\eta_2(L)$ varies rapidly with growth of the value L (typically over two or three cells) to the value of the bulk order parameter. In the previous calculation of OP $\eta_2(L)$ by linearization of the MFP's[1] the number of distinct layers which must be taken into account equals approximately three, whereas in the present theory four distinct layers are needed, and the degree of ordering near the solid surface is the greatest. The value of the OP $\eta_2(1)$ decreases with increasing temperature, as shown in Figure 2. The most rigorous test of the theory would be a comparison to the molecular dynamics OP $\eta_2(L)$ for the same pair potential. Such computer experiment data have been reported [9]. Generally, agreement between the calculated values and the computer experimental data are reasonable Figure 1. (open squares indicate points calculated by MD at the same thermodynamical conditions).

The dependence of the scaled RVC $\gamma_1/\{\sqrt{\varepsilon_0 I/\sigma_0^3}\}$ for reduced volume $v/\sigma_0^3 = 4.0$ on the layer L is plotted in Figure 3 for the case of a strong interaction ($\varepsilon_{0w} = 5.0 * \varepsilon_0$) and for different values of the reduced temperature $\beta^{-1} \equiv kT/\varepsilon_0 = 0.75$ (curve 1), 0.7885 (curve 2), 0.827 (curve 3). The values of the RVC decreases with the growth of the value L to the value in the bulk. One can ascribe such behavior of the rotational viscosity coefficient γ_1 to a tendency of the molecules to be more ordered near the surface than in the bulk. A similar behavior of the shear viscosity in the molecular liquids near a solid surfaces has been discovered in.[12]

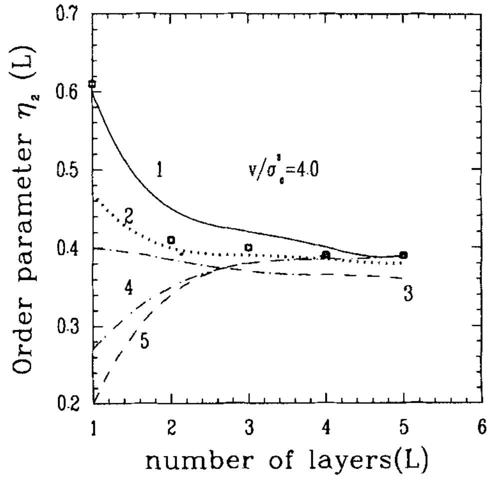

FIGURE 1 Dependence of the order parameter $\eta_2(L)$ on the number of cells L from the wall, for $\gamma = 3.0$, reduced volume $v/\sigma_0^3 = 4.0$, in the case of a strong interaction $\varepsilon_{ow} = 5.0 * \varepsilon_0$, for different values of the reduced temperature $\beta^{-1} \equiv kT/\varepsilon_0 = 0.5$ (curve 1), 0.6 (curve 2), 0.7 (curve 3), and in the case of a weak interaction $\varepsilon_{ow} = 0.5 * \varepsilon_0$ for $\beta^{-1} = 0.5$ in the framework of the present theory (curve 4) and by linearization of the nonlinear MEPs[1] (curve 5). Open squares indicate points calculated by means of MD[9] at the same thermodynamic conditions.

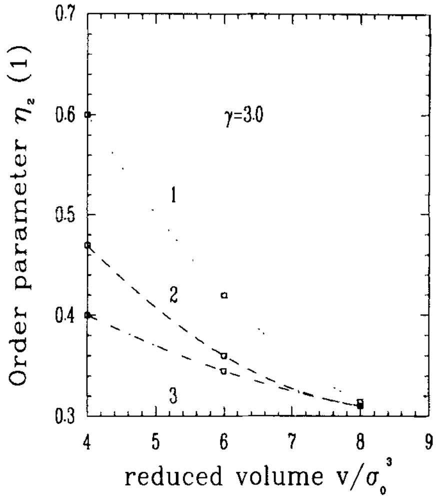

FIGURE 2 Volume dependence of the order parameter $\eta_2(1)$ for the first layer in the case of the strong interaction $\varepsilon_{0w} = 5.0 * \varepsilon_0$ and $\gamma = 3.0$, for different values of the reduced temperatures: $\beta^{-1} = 0.5$ (curve 1), 0.6 (curve 2) and 0.7 (curve 3).

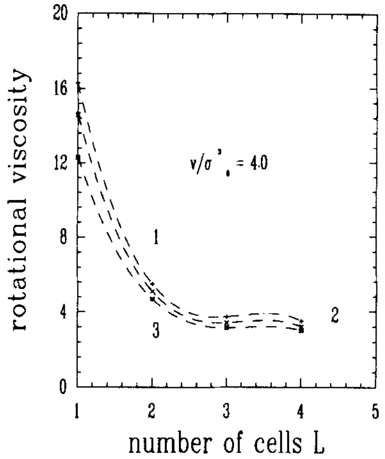

FIGURE 3 Dependence of the scaled rotational viscosity coefficient $\gamma_1/\{\sqrt{\varepsilon_0 I/\sigma_0^3}\}$ on the number of cells L from the wall, for $\gamma = 3.0$, reduced volume $v/\sigma_0^3 = 4.0$, in the case of a strong interaction $\varepsilon_{0w} = 5.0 * \varepsilon_0$, for different values of the reduced temperature $\beta^{-1} \equiv kT/\varepsilon_0 = 0.75$ (curve 1), 0.7885 (curve 2), 0.827 (curve 3).

VI. CONCLUSION

In this paper has been developed in a statistical-mechanical theory which takes into account the translational-translational, orientational-orientational and mixed correlations for describing the equilibrium order parameters and the rotational viscosity coefficient γ_1. The calculations have been carried out for a cubic close packed system composed of molecules interacting by means of the Gay-Berne potential and with a (9-3) like orientation dependent molecule-wall interaction. The calculations show that the number of distinct layers which must be taken into account depend mainly on the character of cell correlations. It should be mentioned that a similar problem for the same nonuniform system has been solved in the framework of linearization of the nonlinear functional[1] $\varphi_{i,j}(i) = \varphi^b_{i,j}(i) + h_{i,j}(i)$, where $|h_{i,j}(i)| \ll |\varphi^b_{i,j}(i)|$. In that case, one has an additional, to the bulk MFPs, system of linear integral equations but, in the case of the strong interaction ($\varepsilon_{ow} = 5.0 * \varepsilon_0$), it is questionable whether can one use a such decomposition. Here I have extended the conditional distribution function approach to the case of a strong wall-molecule interaction.

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