Statistical description of Coulomb-like systems

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(Received 31 July 2011; revised manuscript received 4 November 2011; published 8 December 2011)

The solution of the problem of the partition function calculation for a Coulomb-like system is proposed. The quantum-field-theory approach is used to give a statistical description of a system of interacting particles with due regard to arbitrary spatially inhomogeneous configurations. Formation of structures in a Coulomb-like system is analyzed and applied to the case of of dusty crystals and two-dimensional colloidal crystals. In the one-dimensional case, an exact solution for the spatial distribution of charged particles is obtained. In the two-dimensional case, the exact partition function for homogeneous distribution of particles is presented. We have analytically derived the necessary condition for the crystal formation in a system of interacting particles in the three-dimensional case.

DOI: 10.1103/PhysRevE.84.061115 PACS number(s): 05.70.Jk, 51.30.+i, 82.60.Lf

I. INTRODUCTION

Particle systems with Coulomb interaction (Coulomb-like systems), such as plasmas, colloidal particles, electrolyte solutions, electron gas in solids, etc., are widely presented both in nature and under laboratory conditions. Many soft-matter systems, e.g., surfactant solutions, colloids in various solvents, and dust particles in plasmas, exhibit self-assembling into various structures. The interest in this system is generated by its applications to the studies of a variety of peculiar phenomena in various fields of science [1–3]. One of the most important problems here is the statistical description of Coulomb-like systems with high concentrations of interacting particles [4]. The formation of different crystal structures, transitions between crystalline phases of different symmetries, and melting-like phenomena [5,6] are observed when the concentration increases [7]. Moreover, dusty plasmas as well as colloidal suspensions may serve as perfect media for the experimental investigation of classical fluids and solids [7–16], since both direct measurements of the interaction between colloidal particles [17,18] and theoretical treatment [19] reveal the Coulomb-like nature of the interaction in colloidal suspensions [2]. The theoretical description of such systems is one of the key problems of statistical physics.

It is rather difficult to solve this problem since traditional methods of statistical mechanics cannot be applied to inhomogeneous systems with Coulomb-like interactions. Here specific methods should be applied, which can take into account the inhomogeneity of particle distributions. In particular, these methods should employ an appropriate procedure to find the dominant contribution to the partition function and to avoid free-energy divergences when the volume grows infinite. Only a few model systems of interacting particles are known for which the partition function can be evaluated exactly in the thermodynamical limit [20–23]. As for the description of equilibrium states, only a few results have been obtained within the framework of exact equilibrium statistical mechanics. Now the known results can be obtained much more easily in terms of the method of collective variable and integral transformations [24]. Moreover, this method makes it possible to obtain the free energy of a classical plasma system in a regular manner up to an arbitrary order. A universal sequence of ordered structures was obtained from the mesoscopic description of the self-assembly using the functional and statistical field theory [25]. In this paper an alternative approach is proposed, resulting in a density-functional theory with rather simple structure. The theory makes it possible to include the contributions of long-range correlations between fluctuations in the grand potential. The effect of fluctuations on the stability of a periodic structure was investigated in Ref. [14] (see also [15]). The field-theory method was applied to show that a periodic structure is stabilized due to the low-amplitude order parameter. The nature of the transition and properties of various ordered phases were studied for the case of an isotropic system, either dusty plasma or a colloidal system.

One of the ways to describe the spatially inhomogeneous distribution in a system of interacting particles is to use the new unconventional method proposed in Refs. [26,27], which employs the Hubbard-Stratonovich representation of the partition function [28]. Now this method is extended and applied to a system with Coulomb-like interaction to find the solution for the particle distribution. It is important that this solution has no divergences for the thermodynamical limits.

The purpose of the present contribution is to apply the quantum-field-theory approach to the statistical description of a Coulomb-like system and to calculate the thermodynamic characteristics for both homogeneous and inhomogeneous distributions of interacting particles. The condensed spatially periodic structures are studied as well. We consider electrically neutral systems and use the pure Coulomb interaction potential or an effective screened potential, if the appropriate model can be introduced. We use the saddle-point approximation with regard to the conservation of the number of particles that yields a nonlinear equation for the new field variable. In the three-dimensional case, this equation reduces to the sine-Gordon equation whose solution determines the state associated with the dominant contribution in the partition function. This method makes it possible to describe the conditions of Wigner crystal formation in a system of dust particles in a plasma. There may exist various possibilities for different parameters corresponding to the interaction potential. Nevertheless, the results for simple and basic cases are most important for understanding the behavior of dusty plasmas in complex situations. The necessary condition for the crystal formation in a system of dust particles is derived analytically for the three-dimensional case. For the one- and

two- dimensional cases, exact solutions for various spatial distributions of charged particles are found.

II. STATISTICAL DESCRIPTION OF A COULOMB-LIKE SYSTEM

We begin with a brief account of an inhomogeneous system of interacting particles [26,27]. The method makes it possible to describe an equilibrium system of interacting particles taking into account the formation of a thermodynamically stable spatial particle distribution and to consider the collective behavior of the structures thus formed. The statistical investigation of particle interaction in condensed matter is based on the function of the canonical ensemble of particle sets $\{n\}$. We can write the partition function in the well-known form; i.e.,

$$Z_N = \sum_{\{n\}} \exp[-\beta H(n)], \tag{1}$$

where $\sum_{\{n\}}$ implies summation over all probable distributions of occupation number $\{n_s\}$, $\beta = \frac{1}{kT}$ is the inverse temperature, and $H(n)$ is the configuration Hamiltonian of the system. A system of interacting particles with regard to the type of statistics with quantum correlations can be treated in the classical manner with the model Hamiltonian [29] given by

$$H(n) = \sum_s \varepsilon_s n_s - \frac{1}{2}\sum_{s,s'} W_{ss'} n_s n_{s'}, \tag{2}$$

with ε_s being the additive part of the energy in the state s that in most cases is equal to the kinetic energy, $W_{ss'}$ being the interaction energies for the particles in the states s and s'. In this model, the macroscopic states of the system are described by a set of occupation numbers n_s. Index s labels an individual particle state that can correspond to a fixed site of the Ising lattice [21]. It is clear that calculating of the partition function makes a rather complicated problem even in the case of the Ising model. The partition function for the canonical ensemble of a system of interacting particles is given by Ref. [27]

$$\begin{aligned} Z_N &= \sum_{\{n\}} \exp[-\beta H(n)] \\ &= \sum_{\{n\}} \exp\left\{ -\beta\left[\sum_s \varepsilon_s n_s - \frac{1}{2}\sum_{s,s'} W_{ss'} n_s n_{s'} \right]\right\}. \end{aligned} \tag{3}$$

In order to perform a formal summation in Eq. (2), additional field variables can be introduced in terms of the theory of Gaussian integrals [28–30]:

$$\begin{aligned} &\exp\left\{ \frac{1}{2\theta}\nu^2 \sum_{s,s'} \omega_{ss'} n_s n_{s'} \right\} \\ &= \int_{-\infty}^{\infty} D\varphi \exp\left\{ \nu \sum_s n_s \varphi_s - \frac{1}{2\beta}\sum_{s,s'} \omega^{-1}_{ss'} \varphi_s \varphi_{s'} \right\}, \end{aligned} \tag{4}$$

where $D\varphi = \prod_s d\varphi_s / \sqrt{\det 2\pi\beta\omega_{ss'}}$ depends on the character of the interaction energy and $\omega^{-1}_{ss'}$ is the inverse interaction matrix. The latter satisfies the condition $\omega^{-1}_{ss''}\omega_{s''s'} = \delta_{ss'}$. The partition function of a system may be rewritten as

$$\begin{aligned} Z = \int_{-\infty}^{\infty} D\varphi \sum_{\{n_s\}} \exp\Bigg\{ &\sum_s (i\varphi_s - \beta\varepsilon_s) n_s \\ &- \frac{1}{2\beta}\sum_{s,s'} \left(W^{-1}_{ss'} \varphi_s \varphi_{s'} \right) \Bigg\}. \end{aligned} \tag{5}$$

In the above analysis we did not restrict the number of particles. Now let us fix the total number of particles in the system $N = \sum_s n_s$. To do this we use the well-known Cauchy formula [21]:

$$\frac{1}{2\pi i}\oint d\xi\, \xi^{\sum_s n_s - N - 1} = 1. \tag{6}$$

Then the partition function can be rewritten in the form

$$\begin{aligned} Z_N = \frac{1}{2\pi}\oint d\xi \int_{-\infty}^{\infty} D\varphi \exp\Bigg\{ &-\frac{1}{2\beta}\sum_{s,s'} \left(W^{-1}_{ss'} \varphi_s \varphi_{s'} \right) \\ &- (N+1)\ln\xi \Bigg\} \prod_s \sum_{\{n_s\}} [\xi \exp(i\varphi_s - \beta\varepsilon_s)]^{n_s}, \end{aligned} \tag{7}$$

where summation extends over the occupation numbers n_s. Finally it reduces to

$$Z_N = \frac{1}{2\pi}\oint d\xi \int_{-\infty}^{\infty} D\varphi \exp[-\beta F(\varphi,\xi)], \tag{8}$$

where the effective free energy can be present as

$$\begin{aligned} \beta F(\varphi,\xi) = &\frac{1}{2\beta}\sum_{s,s'} \left(W^{-1}_{ss'} \varphi_s \varphi_{s'} \right) \\ &+ \delta \sum_s \ln(1 - \delta\xi e^{-\beta\varepsilon_s}\cos\varphi_s) + (N+1)\ln\xi. \end{aligned} \tag{9}$$

$\delta = +1$ for Bose statistics and $\delta = -1$ for the Fermi statistics; $\xi \equiv e^{\beta\mu}$ is the absolute chemical activity of the chemical potential μ. This presentation of the partition function makes it possible to use the efficient methods developed in the quantum-field theory without imposing any additional restrictions on the integration over the field variables. The functional $\beta F(\varphi,\xi)$ depends on the distribution of field variables φ and the absolute chemical activity ξ. The field variable φ contains the same information as the original partition function with summation over the occupation numbers, i.e., all the information about probable states of the system. The partition function represented in terms of the functional integral over auxiliary fields corresponds to the construction of an equilibrium sequence of probable states of the system with regard to their weights. This representation provides a possibility to employ the well-known method of quantum-field theory and to make no use of the perturbation theory. The extension to the complex plane makes it possible to apply the saddle-point method.

Now we can employ the saddle-point method to find the asymptotic value of the partition function Z_N for $N \to \infty$; the dominant contribution is given by the states which satisfy the extremum condition for the functional. The particle

distribution is determined by the saddle-point solution of the equations; i.e.,

$$\frac{\delta\beta F}{\delta\xi} = \frac{\delta\beta F}{\delta\varphi} = 0, \tag{10}$$

which is valid in both cases of spatially homogeneous or inhomogeneous distributions. The solutions associated with finite effective free energy $F(\varphi,\xi)$ as the volume of the system tends to infinity are thermodynamically stable. The spatially inhomogeneous solution of these equations describes the distribution of interacting particles. Such inhomogeneous behavior is determined by the interaction intensity. In other words, the accumulation of particles in a finite spatial domain is related to the spatial distribution of the fields and the activity. In the case of a Coulomb-like system that is neutral as a whole, the interaction energy can be present as $\omega_{ss'} = \omega(|r_s - r_{s'}|) = (Q^2/|r_s - r_{s'}|)\exp(-\kappa|r_s - r_{s'}|)$. The inverse matrix $\omega^{-1}_{ss'}$ to such interaction in the continuum limiting case should be treated in the operator sense [29]; i.e.,

$$\omega^{-1}_{rr'} = \delta_{rr'}\widehat{L_{r'}} = -\frac{1}{4\pi Q^2}(\triangle - \kappa^2), \tag{11}$$

where Q is the particle charge, $\triangle$ is the Laplace operator, and κ is the screening length. With the accuracy up to the surface term, the effective free energy in the continuum case is given by

$$\beta F(\varphi,\xi) = \int dV \left\{ \frac{1}{8\pi Q^2\beta}[(\nabla\varphi)^2 + \kappa^2\varphi^2] + \delta\sum_p \ln(1 - \delta\xi e^{-\beta\varepsilon_p}\cos\varphi)\right\} + (N+1)\ln\xi. \tag{12}$$

Integration over the momentum and coordinates should be performed over the cell volume $(2\pi\hbar)^3$ in the phase space of individual states. To demonstrate the advantages of the approach we first derive the well-known result in the case of ideal gas. For ideal gas $\varphi = 0$ and the partition function may be written as

$$Z_N = \frac{1}{2\pi i}\oint d\xi \exp\left\{ -\delta\sum_p \ln(1 - \delta e^{-\beta\varepsilon_p}) - (N+1)\ln\xi\right\}, \tag{13}$$

which can be transformed to

$$Z_N = \frac{1}{2\pi i}\oint \frac{d\xi}{\xi^{N+1}}\prod(1 - \delta e^{-\beta\varepsilon_p})^{\delta}, \tag{14}$$

since the partition function of the grand canonical ensemble $Z = \prod(1 - \delta e^{-\beta\varepsilon_p})^{\delta}$ fully reproduces the well-known result for the partition function of an ideal quantum gas. As has been shown in Ref. [22] for the classical statistic we have $\xi \leqslant 1$ and $\delta\sum_p \ln(1 - \delta\xi e^{-\beta\sum_p\varepsilon_p}\cos\varphi) \approx -\xi e^{-\beta\varepsilon_p}\cos\varphi + \cdots$. The effective free energy for the Boltzmann statistic can be rewritten in the form [27]

$$\beta F(\varphi,\xi) = \int dV\left\{\frac{1}{2r_e}[(\nabla\varphi)^2 + \kappa^2\varphi^2] - \xi A\cos\varphi\right\} + (N+1)\ln\xi, \tag{15}$$

where we have introduced a new variable $r_e = 4\pi Q^2\beta$. Here $A \equiv \lambda^{-3} = (2\pi m/\beta h^2)^{3/2}$, where λ represents the quantum thermal wavelength. The effective free energy of noninteracting particles vanishes, $\varphi = 0$, and thus in this case of Boltzmann statistics the free energy can be written in the following simple form:

$$\beta F(\varphi,\xi) = -\int d\mathbf{r}\xi\left(\frac{2\pi m}{\beta\hbar^2}\right)^{3/2} + (N+1)\ln\xi. \tag{16}$$

The normalization condition reduces to the equation

$$V\xi\left(\frac{2\pi m}{\beta\hbar^2}\right)^{3/2} = N + 1, \tag{17}$$

which yields the absolute chemical activity $\xi = \frac{N}{V}(2\pi m/\beta\hbar^2)^{-3/2}$. Substituting this quantity into the expression for the effective free energy yields the effective free energy for fixed number of particles and energy, and the partition function of noninteracting particles is then given by $Z_N = \exp(-\beta F_B)$ where

$$\beta F_B = \frac{3N}{2} - \ln\frac{N!}{V}\left(\frac{\beta\hbar^2}{2\pi m}\right)^{3/2}, \tag{18}$$

which reproduces the well-known free energy for the ideal Boltzmann gas.

In the case of interacting particles, we propose a method that makes it possible to determine the states with the dominant contributions to the partition function. Namely, we use the saddle-point approximation which provides an efficient and powerful technique in the quantum-field theory. This approach has been successfully applied to many problems. In particular, such approach holds in the case of a high-temperature many-body system. We start from the one-dimensional case.

III. ONE-DIMENSIONAL COULOMB-LIKE SYSTEM

Let us consider a one-dimensional system with linear particle density. Charges distributed along a macromolecule can be treated as an example of such a system. We consider a cylindrical molecule of length L and radius $r \ll L$. Let the Coulomb charges lie on the cylinder axis. In this case the problem can be solved exactly [27]. The free energy of a system of charged particles in the one-dimensional case can be presented as

$$\beta F = \frac{V}{L}\int_0^L dz\left\{\frac{1}{r_e}\left(\frac{d\varphi}{dz}\right)^2 - \xi A\cos\varphi\right\} + (N+1)\ln\xi. \tag{19}$$

Then the saddle-point equation reduces to the sine-Gordon equation

$$\frac{1}{r_e}\left(\frac{d^2\varphi}{dz^2}\right) + \xi A\sin\varphi = 0. \tag{20}$$

The first integral of this equation is given by

$$\frac{1}{r_e}\left(\frac{d\varphi}{dz}\right)^2 + \xi A\cos\varphi = C. \tag{21}$$

It corresponds to the exact solution with the finite period; i.e.,

$$l = \frac{1}{\sqrt{2r_e}}\int \frac{d\varphi}{\sqrt{C-\xi A\cos\varphi}} = \frac{4K(p)}{\sqrt{2r_e(C+\xi A)}}, \tag{22}$$

where $K(p)$ is the full elliptic integral of the first kind with unknown argument $p = \sqrt{2\xi A/(C+\xi A)}$. This solution depends on the integration constant C [31]. Substituting the solution into the free energy yields

$$\beta F = 2\xi AV\left\{\frac{2E(p)}{p^2K(p)} - \frac{1}{p^2} + 1\right\} - \xi AV + (N+1)\ln\xi. \tag{23}$$

Here $E(p)$ is the full elliptic integral of the second kind with the same argument. The free energy extremum is achieved for $p = 1$ or $C = \xi A$ which corresponds to the soliton solution given by

$$\varphi = 4\arctan\exp(z\sqrt{r_e\xi A}). \tag{24}$$

This solution determines the state associated with the dominant contribution in the partition function. Thus, the free energy takes the form

$$\beta F = 8\frac{V}{L}\left(\frac{\xi A}{r_e}\right)^{1/2} - \xi AV + (N+1)\ln\xi. \tag{25}$$

Minimization of the free energy by chemical activity leads to the equation

$$4\frac{V}{L}\left(\frac{\xi A}{r_e}\right)^{1/2} - \xi AV + (N+1) = 0. \tag{26}$$

Its solution exists only for $(N+1)(r_e/L) \geqslant 16$ or $\pi Q^2/kT > 4$. This leads to the chemical activity $\xi = (N+1)/V$. Substituting this value in the free energy provides the following result for the one-dimensional Coulomb-like system:

$$\beta F = \beta F_B + 8\left[(N+1)\frac{L}{r_e}\right]^{1/2}. \tag{27}$$

In this case the free energy of interacting particles increases with the increase of number of particles and the size of the system. The possible periodic structure can be motivated by spatial boundary conditions. The period of the structure $l = L[L/(N+1)r_e]^{1/2}$ increases with the decrease of the number of particles. Such a system is homogeneous on the macroscopic scale, but the particle distribution can be spatially periodic.

IV. TWO-DIMENSIONAL CASE: HEXAGONAL STRUCTURE

As was shown in Ref. [19], the Debye-Huckel theory for the interfacial geometry predicts the Coulomb-like system for charged colloids or charged polymers at monolayers, solid substrates, and interfaces. The phase diagram of two-dimensional electron liquids was described in Refs. [32–34].

In the case of a two-dimensional system we can obtain an exact solution for the homogeneous distribution of particles. Let us consider the Coulomb-like potential

$$\omega_{ij} = -\frac{Q^2}{\langle r\rangle}\ln\kappa r_{ij}, \tag{28}$$

where $r_{ij} = |r_i - r_j|$ is the distance between particles and $\langle r\rangle$ is the average distance between particles. For charged colloidal particles we have $Q^2 = l_b\rho^2$, where $l_b = e^2/4\pi\varepsilon kT$ is the Bjerrum length, which defines the length at which two unit charges interact with thermal energy, and ρ is the charge density. This model potential has the same form as for the interaction between two homogeneously charged lines in three-dimensional space. Thus, the motion of real charges on the two-dimensional plane is similar to the motion of parallel lines oriented perpendicularly to the plane of the two-dimensional system. In the continuous limit the partition function in the case under consideration can be written in the standard form

$$Z_N = \int \exp[-\beta H(r,p)]d^N\mathbf{r}d^N\mathbf{p}, \tag{29}$$

where H is the Hamiltonian of the system

$$H(r,p) = \sum_i \frac{p_i^2}{2m} + \frac{1}{2}\sum_{i,j}\omega_{ij}. \tag{30}$$

In the two-dimensional case the equation of state can be derived from the relation

$$P = kT\frac{\partial\ln Z_n}{\partial S}, \tag{31}$$

where S is the square of a circle of radius R. Having introduced the dimensionless variable $r_i' = r_i/S^{1/2}$ we can rewrite the partition function in the form

$$Z_N = S^N\int \exp[-\beta H(r,p)]d^N\mathbf{r}'d^N\mathbf{p}'. \tag{32}$$

The derivative of the partition function in this case can be presented as

$$\frac{\partial Z_N}{\partial S} = NS^{-1}Z_N - \frac{S^N}{kT}\int \exp[-\beta H(r,p)]\frac{\partial H(r,p)}{\partial S}d^N\mathbf{r}'d^N\mathbf{p}' \tag{33}$$

or, in another representation,

$$\frac{\partial Z_N}{\partial S} = \frac{NZ_N}{S} - \frac{S^N}{kT}\int \exp[-\beta H(r,p)] \times \frac{1}{2}\sum_{i,j}\frac{r_{ij}}{2S}\frac{\partial\omega_{ij}}{\partial r_{ij}}d^N\mathbf{r}'d^N\mathbf{p}'. \tag{34}$$

This result can be presented in the dimensional form within the context of the explicit form of the potential

$$\frac{\partial Z_N}{\partial S} = \frac{NZ_N}{S} - \frac{N(N-1)}{4SkT}\int \exp[-\beta H(r,p)]d^N\mathbf{r}'d^N \equiv \frac{NZ_N}{S} - \frac{N(N-1)Z_N}{4SkT}. \tag{35}$$

Substituting this result into the equation of state generates the exact solution for the two-dimensional Coulomb-like system; i.e.,

$$PS = NkT\left\{1 + \frac{(N-1)Q^2}{4kT\langle r\rangle}\right\}. \tag{36}$$

If we take into account that $\langle r\rangle \sim n^{-1/2}$, where n is the concentration of particles, it is possible to show that, for a large concentration, the system of interacting particles will be unstable and the inhomogeneous particle distribution can appear. Such inhomogeneous distributions are caused by the long-range nature of the Coulomb interaction. In the case of intensive interaction, the Coulomb-like system is unstable as a whole, so the minimum value of the free energy is achieved in the case of inhomogeneous distribution of particles.

Now we employ the proposed approach in order to find the states associated with the Wigner crystal. In the two-dimensional case, we can write the effective free energy in the following simple form:

$$\beta F = h\int dxdy\left\{\frac{1}{r_e}[(\nabla\varphi)^2 + \kappa^2\varphi^2] - \xi A\cos\varphi\right\} + (N+1)\ln\xi, \tag{37}$$

where h is the thickness of the two-dimensional layer. In the general case, the equation for the saddle-point states $\delta\beta F/\delta\varphi = 0$ is given by

$$\frac{1}{r_e}\{\triangle_2\varphi - \kappa^2\varphi\} + \xi A\sin\varphi = 0, \tag{38}$$

where $\triangle_2$ is the two-dimensional Laplace operator. The chemical activity can be obtained from the normalization condition $\delta\beta F/\delta\xi = 0$:

$$h\int dxdy\xi A\cos\varphi = N+1. \tag{39}$$

It should be noted that this condition enables us to introduce the concentration of particles $\rho(\mathbf{r}) = \xi A\cos\varphi(\mathbf{r})$. The first integral of the equation for the field variable can be obtained from the first equation by multiplying this equation by $\nabla\varphi$ with regard to the relation $\triangle = \nabla^2$. Thus, we obtain the first integral in the form

$$\frac{1}{r_e}[(\nabla\varphi)^2 + \kappa^2\varphi^2] + \xi A\cos\varphi = E, \tag{40}$$

where E is an unknown integration constant that should be found from the condition of existence of the solution. Although this equation cannot be solved in the general case, it provides a tool to study many interacting Coulomb-like systems under various external conditions. In terms of the density function $\rho(\mathbf{r})$ the first integral also can be written as

$$\frac{1}{r_e}\left\{\frac{1}{(\xi A)^2 - \rho^2}(\nabla\rho)^2 - \kappa^2\arccos^2\left(\frac{\rho}{\xi A}\right)\right\} + \rho = E, \tag{41}$$

and the effective free energy in terms of the first integral has the form

$$\beta F = h\int dxdy\left\{E + 2\frac{\kappa^2}{r_e}\arccos^2\left(\frac{\rho}{\xi A}\right) - 2\rho\right\} + (N+1)\ln\xi. \tag{42}$$

Let us consider the case of the periodic distribution of particles in the system. Here it is necessary to assume that $\rho \sim \rho_0\{1 + \cos(\mathbf{k}\varrho)\}$ where ϱ is the surface variable and $\mathbf{k}$ is inverse lattice vector of the periodic distribution. The first integral should be constant for each point of the space including the spatial point where $\rho = 0$. In this case the first integral can be determined as

$$-\frac{\pi^2\kappa^2}{4r_e} = E. \tag{43}$$

Substituting this relation into the free energy, we can present it in the form

$$\beta F = V\left\{-\frac{\pi^2\kappa^2}{4r_e} + \frac{2\kappa^2}{r_e}\arccos^2\left(\frac{\rho_0}{\xi A}\right)\right\} + (N+1)\ln\xi, \tag{44}$$

where the value of the integral over the coordinate space is estimated in terms of averaged concentration and $V = hS$ is the volume of the system. From the condition of minimum effective free energy, we conclude that the solution exists if $\xi A = \rho_0$ and the effective free energy takes the form

$$\beta F \simeq \beta F_B + (N+1)\left\{\frac{\pi^2\kappa^2}{4nr_e} - 1\right\}, \tag{45}$$

which can be smaller than the free energy of gas if $8\pi^2\kappa^2/nr_e < 1$.

V. THREE-DIMENSIONAL CASE: WIGNER CRYSTAL

The three-dimensional structure of induced electrorheological solids was described in Refs. [35,36]. In this section we describe the system of dust particles in weakly ionized plasma but in the stationary case. In this case we again use a method that makes it possible to determine the states with the dominant contributions to the partition function. Namely, we use the saddle-point approximation. In the general case, the equation for the saddle-point states $\delta\beta F/\delta\varphi = 0$ is given by

$$\frac{1}{r_e}\{\triangle\varphi - \kappa^2\varphi\} + \xi A\sin\varphi = 0, \tag{46}$$

where $r_e = 2\pi Q^2\beta$. The chemical activity can be obtained from the normalization condition $\delta\beta F/\delta\xi = 0$:

$$\int dV\xi A\cos\varphi = N+1. \tag{47}$$

It should be noted that this condition enables us to introduce the concentration of particles as $\rho(\mathbf{r}) = \xi A\cos\varphi(\mathbf{r})$. This leads to the first integral given by

$$\frac{1}{r_e}\{(\nabla\varphi)^2 + \kappa^2\varphi^2\} + \xi A\cos\varphi = E, \tag{48}$$

where E is an unknown integration constant. Similarly to the two-dimensional case, this equation cannot be solved explicitly in the general case.

Let us start from the case of a homogeneous distribution of interacting particles. In this case we have to derive the

condition for the existence of the solution $\varphi = \varphi_0 =$ constant from the equation for the field variable

$$\frac{\kappa^2}{r_e}\varphi_0 + \xi A \sin\varphi_0 = 0 \tag{49}$$

and to find the chemical activity from the normalization condition

$$\xi A V \cos\varphi_0 = N + 1. \tag{50}$$

Within the context of the first integral and the equation for the chemical activity, the free energy can be written as

$$\beta F = F_B + N\left\{\frac{\kappa^2}{r_e n} - \ln\cos\varphi_0\right\}, \tag{51}$$

where we have introduced the concentration of particles $n = (N+1)/v$. Obviously, the second term in the free energy is always positive since $\cos\varphi_0 \leqslant 1$ and thus the free energy of a homogeneous system of interacting particle is greater than the free energy of the Boltzmann gas.

In the general case, particle distributions in Coulomb-like systems are inhomogeneous, the long-range nature of the Coulomb interaction being the reason. Similarly to the two-dimensional Coulomb-like system in the case of intensive interaction the system becomes unstable as a whole, so the minimum value of the free energy is achieved in the case of inhomogeneous distribution of particles.

Now we employ the proposed approach to show how to find the states associated with the Wigner crystal. Taking into account that the density function $\rho(\mathbf{r}) = \xi A\cos\varphi$ is only positive and assuming that the state of interest does exist, we take the periodic distribution function in the form

$$\begin{aligned}\rho(\mathbf{r}) &= \xi A\cos\varphi \\ &= \xi A\{1 + \cos(k_x x) + \cos(k_y y) + \cos(k_z z)\},\end{aligned} \tag{52}$$

which corresponds to the cubic lattice with the wave vector $\mathbf{k} = (k_x, k_y, k_z)$. If we assume that one charged particle is present at every lattice site and that the lattice is isotropic $k_x = k_y = k_z = 2\pi n^{1/3}$ [where $n = (N+1)/V$ is the particle density], then the normalization condition yields $\xi = n/A$ or $\xi = (N+1)/AV$. From the first integral for the field variable, one can conclude that $E^2 = \pi^2\lambda^2/4r_e$. We substitute this relation into the free energy written in terms of the first integral; i.e.,

$$\beta F = \int dV\left\{E + 2\frac{\kappa^2}{r_e}\varphi^2 - 2\xi A\cos\varphi\right\} + (N+1)\ln\xi. \tag{53}$$

Here φ is the the value for average concentration of particles. Now the effective free energy of the system can be written in the simple form given by

$$\beta F = \beta F_B + (N+1)\left\{\frac{\pi^2\kappa^2}{4nr_e} - 1\right\}. \tag{54}$$

Introducing the coupling parameter $\Gamma_e \equiv r_e n^{1/3}$ (which is the ratio of the Coulomb to kinetic energy) provides a relation for the critical value of the coupling parameter; i.e.,

$$\Gamma_e \geqslant 4\pi^2\kappa^2 n^{2/3} \equiv (2\pi\kappa L)^2, \tag{55}$$

where L is the lattice period. With this condition being satisfied, we can expect a crystal structure to be formed. Namely, such structures are observed in dusty plasmas [1]. In terms of the structure lattice parameter used in Ref. [1] $l \equiv \kappa L$ (the interparticle distance normalized by the effective screening length), the relation obtained is given by

$$\Gamma_e \geqslant (2\pi l)^2. \tag{56}$$

This relation gives the value of the same order as the result of the numerical simulation [1]. We cannot solve the problem of crystal structure formation in dusty plasmas exactly; nevertheless we can analytically predict the conditions for such formation. The problem is that we do not know the three-dimensional solution of the sine-Gordon equation that determines the field variable. Moreover, this approach provides a description of spatially periodic distributions. The partition function has no singularities for any values of the Coulomb-like field. It is shown that the minimum of the free energy sometimes does not correspond to a homogeneous particle distribution, but could indicate the formation of a crystal-like structure.

VI. CONCLUSION

We have developed a formalism to describe the spatially inhomogeneous distribution in a system of interacting particles. It employs the new unconventional method proposed earlier in Refs. [26,27], which employs the Hubbard-Stratonovich representation of the partition function. This method is now extended and applied to a system with Coulomb-like interaction to find the solution for the particle distribution. It is important that this solution has no divergences for the thermodynamical limits. We use the saddle-point approximation with regard to the conservation of the number of particles which yields a nonlinear equation for the new field variable. In the three-dimensional case, this equation reduces to the sine-Gordon equation whose solution determines the state associated with the dominant contribution in the partition function. This method makes it possible to describe the conditions of Wigner crystal formation in a system of dust particles in a plasma. There may exist various possibilities for different parameters corresponding to the interaction potential. However, the results for simple and basic cases are very important for understanding the behavior of a dusty plasma in complex situations. We have analytically derived the necessary condition for the crystal formation in a system of dust particles in the three-dimensional case. In the one- and two-dimensional cases, we have found exact solutions for various spatial distributions of charged particles. The proposed method is designed for studies of self-assembly systems where nonuniform distributions are found on the different length scales. We present the partition function and the equation of state in the most general form. In the three-dimensional case we obtained a condition (quadratic dependence of coupling parameter Γ on the experimental parameter l) which is in good agreement with the experimental data reported in Ref. [1]. In the one-dimensional case we obtained a structure of periodic distribution of charge particles along a cylindrical sample. In the two-dimensional case an exact result is obtained for the partition function for homogeneous distribution in a purely Coulomb system.

The periodic distribution of charged particles in the two-dimensional case is shown to be related of the minimum free energy, which can be calculated in this case for both homogeneous and inhomogeneous particle distributions. To conclude, we note that our method, proposed earlier in Refs. [26,27], can be used to describe systems with various inhomogeneous distributions of interacting particles regarding the thermodynamic function as the free energy and the equation of state. It is shown that the method proposed here makes it possible to obtain the earlier result much more easily.

ACKNOWLEDGMENTS

This work is partially supported by a joint grant of the National Academy of Science of Ukraine and the Russian Foundation of Fundamental Researches.

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