Statistical description of model systems of interacting particles and phase transitions accompanied by cluster formation

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We develop an approach to the statistical description of a system of interacting particles in order to describe spatially inhomogeneous structures. A criterion is proposed for selecting system states whose contributions in the partition function are dominant. A nonperturbative calculation of the partition function is demonstrated. The known results for various systems (hard sphere model, gravitating gas, etc.) are reproduced. Spatially inhomogeneous system states are considered. The conditions for the phase transition accompanied with cluster formation are found for model systems. Cluster size distribution and cluster interaction residual energy are estimated. The formation of new spatial structures in a cluster system is considered. [S1063-651X(98)07206-7]

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I. INTRODUCTION

The formation of spatially inhomogeneous particle and field distributions is a topical problem in condensed matter physics. It concerns the study of physical grounds of the optimum states of the system and is of value for applications in practice [1–3]. Earlier investigations of the formation conditions and behavior of the inhomogeneous states have mainly employed the statistical theory of nonequilibrium processes. However, spatially inhomogeneous particle and field distributions can also be formed in equilibrium systems. The conditions for the formation of such structures and their physical manifestation are determined first of all by the type of interaction. So, we have to formulate an adequate mathematical method that would describe formation and behavior of spatially inhomogeneous equilibrium particle and field distributions.

The purpose of this paper is to develop an approach [4] to the statistical description of a system of interacting particles with regard for spatially inhomogeneous particle distribution. In order to describe such structures, it is necessary to work out a method that would enable us to select the states with thermodynamically stable particle distributions in the partition function. The representation of the partition function in terms of a functional integral over auxiliary fields makes it possible to employ the methods of quantum field theory [5–11]. An attempt to apply the functional integral in the description of multiparticle systems was discussed for the first time in Ref. [12]. The advantages and difficulties of this approach were described in [5]. The extension to the complex plane provides a possibility to apply the saddle-point method making no use of the perturbation theory. It allows one to select the system states associated with both homogeneous and inhomogeneous particle distributions.

A few model systems of interacting particles are known, for which the partition function can be evaluated exactly, at least in the thermodynamical limit. In this paper we demonstrate the efficiency of the proposed approach by a nonperturbative calculation of the partition function for the known model systems with interaction (hard sphere model, Coulomb gas, gravitating gas, etc.). This approach allows one to describe any system of interacting particles with regard for spatially inhomogeneous particle distributions. A typical physical situation that involves bound states in a particle system occurs when the interaction consists of long-range attraction and short-range repulsion. Another realistic situation is associated with the contrary case when the repulsion range is longer than the attraction range. Such physical systems are, e.g., electrons on the liquid helium surface [13], polar atoms and molecules on a metal or dielectric surface [14,15], and ions implanted in silicon [1,16]. As long as such interaction is present, the system cannot be homogeneous and hence it involves spatially inhomogeneous particle distributions—finite-size clusters.

The proposed approach allows one to describe such particle distributions, to calculate cluster size, to estimate the number of particles within a cluster, and to determine the temperature of the phase transition to the state under consideration. The number of particles in a cluster and the size of the latter depend on the interaction type and intensity as well as on external parameters. The residual interaction (uncompensated after the cluster formation) produces interaction between clusters that, in turn, can cause formation of new spatial structures in the cluster system. The problem of how to find the cluster distribution and to estimate the influence of the external factors also can be solved in terms of this approach.

Thus, a unified approach makes it possible to describe equilibrium systems of interacting particles allowing for the formation of thermodynamically stable spatially inhomogeneous particle distributions and to consider the collective behavior of the structures formed. The topicality of this problem is confirmed by the recent attempts made by other authors [1,5,6].

II. PARTITION FUNCTION FOR MODEL SYSTEMS WITH INTERACTION. THE STATE SELECTION CRITERION

A wide range of systems of interacting particles occur, for which the statistics must be involved in consideration while

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dynamical quantum correlations can be disregarded. This means that the interaction can be treated classically. The Hamiltonian of such a system can be written as [17–22]

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

where ε_s is the additive part of the particle energy in the state s (in most cases it is the kinetic energy), $W_{ss'}$ and $U_{ss'}$ are the absolute values of the attraction and repulsion energies of particles in the states s and s', respectively. The macroscopic state of the system is determined by the occupation numbers n_s. The subscript s corresponds to the variables that describe individual particle states. It also can enumerate the lattice sites [18,20]; the specifics of the lattice does not influence the result. Though the approach is adequate for considering the discrete case as well, in this study we are interested only in the continuum approximation and assume that the medium is isotropic.

The partition function of the grand canonical ensemble is given by

$$Z=\sum_{\{n\}} \exp(-\beta H)=\sum_{\{n\}} \exp\left\{-\beta\left[\sum_s \varepsilon_s n_s-\frac{1}{2}\sum_{s,s'} W_{ss'}n_s n_{s'}+\frac{1}{2}\sum_{s,s'} U_{ss'}n_s n_{s'}\right]\right\}, \tag{2}$$

where $\Sigma_{\{n\}}$ implies summation over all possible distributions $\{n_s\}$, $\beta\equiv 1/kT$, and T is the absolute temperature. The summation in Eq. (2) can be formally carried out by introducing auxiliary field variables and making use of the known properties of the Gauss integrals [6,8–11], i.e.,

$$\exp\left\{\frac{\nu^2}{2\theta}\sum_{s,s'} \omega_{ss'}n_s n_{s'}\right\}=\int_{-\infty}^{\infty} D\varphi \exp\left\{\nu\sum_s n_s\varphi_s-\frac{\theta}{2}\sum_{s,s'} \omega_{ss'}^{-1}\varphi_s\varphi_{s'}\right\}, \tag{3}$$

with

$$D\varphi=\frac{\Pi_s d\varphi_s}{\sqrt{\det(2\pi\beta\omega_{ss'})}}.$$

Here $\omega_{ss'}^{-1}$ is the inverse matrix that satisfies the equation $\omega_{ss''}^{-1}\omega_{s''s'}=\delta_{ss'}$ and $\nu^2=\pm 1$ depending on the sign of the interaction or the potential energy.

Within the context of Eq. (3), the partition function can be written as

$$Z=\int D\varphi\int D\psi\sum_{\{n_s\}} \exp\left\{\sum_s (\varphi_s+i\psi_s-\beta\varepsilon_s)n_s-\frac{1}{2\beta}\sum_{s,s'} (W_{ss'}^{-1}\varphi_s\varphi_{s'}+U_{ss'}^{-1}\psi_s\psi_{s'})\right\}. \tag{4}$$

In the above analysis, we did not restrict the number of particles. Now let us fix the number of particles in the system, $N(n)=\Sigma_s n_s$. This means that we consider the canonical ensemble. To do this, we use the well-known Cauchy formula, i.e.,

$$\frac{1}{2\pi i}\oint \xi^{\Sigma_s n_s-N-1}d\xi=1.$$

Then, making use of the contour integral [21,22], we write the partition function of the system of N particles in terms of the grand partition function. Thus we have

$$Z_N=\frac{1}{2\pi i}\oint d\xi\int D\varphi\int D\psi \exp\left\{-\frac{1}{2\beta}\sum_{s,s'} (W_{ss'}^{-1}\varphi_s\varphi_{s'}+U_{ss'}^{-1}\psi_s\psi_{s'})-(N+1)\ln\xi\right\}\prod_s \sum_{\{n_s\}} [\xi\exp(\varphi_s+i\psi_s-\beta\varepsilon_s)]^{n_s}. \tag{5}$$

After summing over the occupation numbers n_s, the partition function reduces to

$$Z_N=\frac{1}{2\pi i}\oint d\xi\int D\varphi\int D\psi e^{-S(\varphi,\psi,\xi)}, \tag{6}$$

where

$$S(\varphi,\psi,\xi)=\frac{1}{2\beta}\sum_{s,s'} (W_{ss'}^{-1}\varphi_s\varphi_{s'}+U_{ss'}^{-1}\psi_s\psi_{s'})+\delta\sum_s \ln(1-\delta\xi e^{-\beta\varepsilon_s+\varphi_s}\cos\psi_s)+(N+1)\ln\xi. \tag{7}$$

In the last expression, the sign $\delta=\pm 1$ depends on the statistics (plus and minus for the Bose and Fermi systems, respectively).

The partition function representation 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 for their weights. This representation enables us to employ the well known methods of quantum field theory and to avoid using the perturbation theory. The extension to the complex plane makes it possible to apply the saddle-point method [7].

It should be emphasized that such a description is useful [5,6] because it advances the study of thermodynamical characteristics of model systems and their dependence on the medium. If interaction of some type does not occur, the general representation (6) and (7) allows one to reproduce the partition function for pure gravitational attraction given in Ref. [6], and the partition function of the sine-Gordon model for pure Coulomb repulsion given in Ref. [5]. This approach also makes it possible to consider the states with spatially inhomogeneous particle distributions. To do this, one has to vary the functional $S(\varphi,\psi,\xi)$ for the fields φ and ψ and the analog of the chemical potential ξ, and then to apply the saddle-point method in order to obtain the asymptotic value for the partition function Z_N as $N\to\infty$. The solutions, in which the action S is finite as the volume of the system tends to infinity, can be interpreted as thermodynamically stable particle distributions. Whether the distribution is homogeneous or inhomogeneous depends on the solutions that satisfy the extremum condition for the functional S, i.e., $\delta S/\delta\varphi=\delta S/\delta\psi=\delta S/\delta\xi=0$. The equations for the saddle-point states are given by

$$\frac{1}{\beta}\sum_{s'} W_{ss'}^{-1}\varphi_{s'}-\frac{\xi_s e^{\varphi_s}\cos\psi_s}{1-\delta\xi_s e^{\varphi_s}\cos\psi_s}=0,$$

$$\frac{1}{\beta}\sum_{s'} U_{ss'}^{-1}\psi_{s'}+\frac{\xi_s e^{\varphi_s}\sin\psi_s}{1-\delta\xi_s e^{\varphi_s}\cos\psi_s}=0,$$

$$\sum_{s}\frac{\xi_s e^{\varphi_s}\cos\psi_s}{1-\delta\xi_s e^{\varphi_s}\cos\psi_s}=N+1, \tag{8}$$

where $\xi_s=\xi e^{\beta\varepsilon_s}=e^{\beta(\varepsilon_s-\mu)}$ and μ is the chemical potential.

This set of equations provides a solution of the above many-particle problem in the sense that it selects the system states whose contributions in the partition function are dominant. Inasmuch as the inverse of the interaction matrix is not defined uniquely in the general case, it is impossible to find the general solution of the set (8). And even if the last problem can be solved, there still remain technical difficulties associated with solving a set of nonlinear equations and interpreting the solutions thereof. In the sections that follow we consider the solutions for some model systems.

It should be noted that the normalization condition [the third equation in the set (8)] enables us to regard the expression

$$f_s=\frac{\xi_s e^{\varphi_s}\cos\psi_s}{1-\delta\xi_s e^{\varphi_s}\cos\psi_s}$$

as the particle distribution function determined by the auxiliary fields. It is obvious that, for given statistics, the distribution function depends on the interaction nature and intensity. Moreover, the proposed representation can be used to extend the treatment of the Bose condensation to the coordinate space. The cluster formation corresponds to particle localization within a limited space. In our treatment, the effect is reflected in the behavior of the auxiliary fields and chemical potential. Probably, the proposed approach will improve the understanding of the fractional statistics of particles too [8,23,24].

In what follows, we apply the approach to describe model systems with various interactions, for which the partition function can be estimated in the thermodynamical limit. With this purpose in view, we make use of the continuum approximation, which makes it possible to obtain analytic expressions even for inhomogeneous particle distributions. In the continuum approximation, the subscript s runs through a continuum of values in the system volume V. When integrating over momenta and coordinates, we bear in mind that the unit cell volume in the space of individual states is equal to $\omega=(2\pi\hbar)^3$.

In the continuum case, the inverse matrix $\omega_{ss'}^{-1}$ for the interaction $\omega_{ss'}=\omega(|\vec{r}_s-\vec{r}_{s'}|)$ is given by [8,10,13]

$$\omega_{rr'}^{-1}=\delta_{rr'}\hat{L}_{r'}, \tag{9}$$

where $\hat{L}_{r'}$ is the operator for which the interaction potential is the Green function. For the screened Coulomb or Newtonian potential, the inverse operator may be written as [8–11]

$$\hat{L}_{r'}=-\frac{1}{4\pi q^2}(\Delta_{r'}-\lambda^2), \tag{10}$$

where q^2 is the interaction constant and λ^{-1} is the screening length. The number of realistic interactions, for which the inverse operator can be found, is limited. The difficulties in obtaining the inverse operator can be avoided by introducing a collective variable [25,26] that corresponds to the relationship between the introduced fields on the saddle-point trajectory. It is easier to find the required parameters of inhomogeneous structures in this approach, however, it is difficult to trace the formation details for such particle distributions. Without loss of generality within the above approximations, in what follows we describe the model systems of interacting particles taking into account their spatially inhomogeneous distributions. First of all we demonstrate the advantage of the approach for the well-known models, and then describe real systems with interaction and find the conditions for the cluster formation and cluster parameters.

III. MODEL SYSTEMS WITH INTERACTION

To demonstrate the advantages of the approach, we first derive the well-known results.

A. Ideal Bose and Fermi gases

For the ideal gases, $\varphi=\psi=0$ and the partition function (6) may be written as

$$Z_N=\frac{1}{2\pi i}\oint d\xi\,\exp\left\{-\delta\sum_s \ln(1-\delta\xi e^{-\beta\varepsilon_s}) -(N+1)\ln\xi\right\}=\frac{1}{2\pi i}\oint\frac{d\xi}{\xi^{N+1}}\prod_s(1-\delta\xi e^{-\beta\varepsilon_s})^{\delta}. \tag{11}$$

Since the partition function of the grand canonical ensemble [21,22] $Z=\Sigma_N\xi^N Z_N$, we thus obtain the known result

$$Z=\prod_s(1-\delta\xi e^{-\beta\varepsilon_s})^{\delta}. \tag{12}$$

B. Ideal Boltzmann gas

For the Boltzmann statistics in the continuum case, Eq. (6) reduces to

$$Z_N=\frac{1}{2\pi i}\oint d\xi\,\exp\left\{\frac{1}{\omega}\int dV\int d^3p[\xi e^{-\beta\varepsilon} -(N+1)\ln\xi]\right\}. \tag{13}$$

Since $\varepsilon=p^2/2m$, where m is the particle mass, then $\int d^3pe^{-(p^2/2m)\beta}=(2\pi m/\beta)^{3/2}$ and hence

$$Z_N=\frac{1}{2\pi i}\oint d\xi\,\exp\left\{\xi V\left(\frac{2\pi m}{\beta\hbar^2}\right)^{3/2}-(N+1)\ln\xi\right\} =\frac{1}{2\pi i}\oint d\xi e^{-S(\xi)}. \tag{14}$$

We find the saddle-point value $\tilde{\xi}$ from the equation $\delta S/\delta\xi=0$ and thus obtain $\tilde{\xi}=(2\pi m/\beta\hbar^2)^{-3/2}(N+1)/V$. Now we substitute this value in Eq. (14) and apply the Stirling's formula $N-N\ln N\simeq\ln N!$. This yields the partition function of the Boltzmann gas to be

$$Z_N^0=\frac{V^{N+1}}{(N+1)!}\left(\frac{2\pi mkT}{\hbar^2}\right)^{3/2(N+1)}. \tag{15}$$

These well known results for the ideal systems show the consistency of the proposed approach with the traditional methods. Later on we demonstrate new advantages of this statistical description.

C. Hard sphere model

We generalize the hard sphere model by assuming that the potential barrier U_0 is finite. This value is determined by the mechanism of particle collisions. The interaction energy can be written as $U_{ss'}=U_0\delta_{ss'}$. In this case, the inverse operator is described by the expression $U_{ss'}^{-1}=U_0^{-1}\delta_{ss'}$. In the continuum case, we can approximately invert the potential. When doing this, we have to remove the self-interaction terms that arise as we pass from the discrete sum $\Sigma_{s,s'}U_{ss'}n_sn_{s'}$ to continuum. Thus, Eqs. (6) and (7) reduce to

$$Z_N=\frac{1}{2\pi i}\oint d\xi\int D\psi e^{-S(\psi,\xi)} \tag{16}$$

and

$$S(\psi,\xi)=\int dV\left[\frac{1}{2\beta U_0V}\psi^2-\xi A\cos\psi\right]+(N+1)\ln\xi, \tag{17}$$

where $A=(2\pi m/\beta\hbar^2)^{3/2}$. The saddle-point equations that satisfy the extremum condition for the action S are given by

$$\frac{1}{\beta U_0V}\psi+\xi A\sin\psi=0,$$

$$\int dV\,\xi A\cos\psi=N+1. \tag{18}$$

From the behavior of the interaction potential ($U=U_0$ for $r<r_0$, and $U=0$ for $r>r_0$ where r_0 is the particle radius) we can conclude that $\psi=0$ everywhere except for the volume $V_0=2v_0(N+1)$ (v_0 is the particle volume), and $\psi=\tilde{\psi}$ in the volume V_0 in which particle interaction occurs. The quantity $\tilde{\psi}$ can be found from the equation

$$\tilde{\psi}+\xi A\beta U_0V\sin\tilde{\psi}=0 \tag{19}$$

with the normalization condition

$$\xi A(V-V_0)+\xi AV_0\cos\tilde{\psi}=N+1. \tag{20}$$

If the interaction energy is finite, then the solution of Eq. (19) may be written in the form $\tilde{\psi}\simeq\pi+\alpha$, $\alpha\ll\pi$, and we find that $\alpha=\pi/\xi A\beta U_0V$. As $\beta U_0\to\infty$, we have $\tilde{\psi}=\pi$. This corresponds to the pure hard sphere model. Having applied the successive approximations to calculate the chemical potential, we rewrite Eq. (20) in the form

$$\xi A(V-V_0)-\xi AV_0\simeq N+1$$

then $\xi\simeq(N+1)/[AV(1-2V_0/V)]$, and thus $\tilde{\psi}\simeq\pi(N+1)\beta U_0/[(N+1)\beta U_0-(1-2V_0/V)]$.

The solutions obtained allow one to estimate the action. We have

$$S=\frac{\pi^2}{\beta U_0}\frac{V_0}{V}\left[\frac{(N+1)\beta U_0}{(N+1)\beta U_0-(1-2V_0/V)}\right]^2 -(N+1)+(N+1)\ln\frac{N+1}{AV(1-2V_0/V)} \tag{21}$$

and thus the partition function is given by

$$Z_N=\frac{V^{N+1}}{(N+1)!}\left(\frac{2\pi mkT}{\hbar^2}\right)^{3/2(N+1)}\left(1-\frac{2V_0}{V}\right)^{N+1} \times\exp\left\{-\frac{\pi^2}{\beta U_0}\frac{V_0}{V}\left[\frac{(N+1)\beta U_0}{(N+1)\beta U_0-(1-2V_0/V)}\right]^2\right\}. \tag{22}$$

The last expression may be rewritten in a compact form

$$Z_N = Z_N^0 \left(1 - \frac{2V_0}{V}\right)^{N+1} \left\{1 - \frac{\pi^2}{\beta U_0} \frac{V_0}{V} \times \left[\frac{(N+1)\beta U_0}{(N+1)\beta U_0 - (1 - 2V_0/V)}\right]^2\right\}, \tag{23}$$

where Z_N^0 is the partition function of the ideal Boltzmann gas (15). For $U_0 \to \infty$, we find the partition function of the pure hard sphere model to be of the form

$$Z_N = Z_N^0 \left(1 - \frac{4v_0(N+1)}{V}\right)^{N+1}. \tag{24}$$

This result can be immediately obtained from the solution of Eq. (18) under the assumption $U_0 \to \infty$. It should be emphasized that solution (24) exactly reproduces the partition function of the hard sphere model. In this approach, it is derived without calculating virial coefficients.

D. Model with Coulomb repulsion. Wigner crystal

Let us consider another model with repulsion. Suppose the interaction is the screened Coulomb potential

$$U_{ss'} = \frac{q^2}{|\vec{r}_s - \vec{r}_{s'}|} e^{-\lambda |\vec{r}_s - \vec{r}_{s'}|},$$

where λ^{-1} is the screening radius. This model describes likely charged particles whose Coulomb repulsion is screened by the uniformly charged background of opposite sign. In this case, the inverse operator is given by Eq. (10). Thus the action S reduces to

$$S = \int dV \left\{\frac{1}{8\pi q^2 \beta} [(\nabla\psi)^2 + \lambda^2 \psi^2] - \xi A \cos\psi\right\} + (N+1) \ln \xi, \tag{25}$$

where $A = (2\pi m/\beta\hbar^2)^{3/2}$ has the same meaning as before. It should be emphasized that this field representation is completely similar to the lattice sine-Gordon model described in [22]. In this book, the sine-Gordon model is proved to reduce to a system of particles with Coulomb repulsion. The partition function is transformed to an expression in terms of occupation numbers. Actually, the authors have solved the inverse problem. However, the representation of the partition function in terms of occupation numbers does not allow one to find the states associated with inhomogeneous particle distributions since it does not provide any criterion for the selection of such states.

Now we employ the proposed approach in order to show how to find the states corresponding, say, to the Wigner crystal. Suppose that a state of interest occurs and can be described as given by

$$\psi = b(\cos k_x x + \cos k_y y + \cos k_z z). \tag{26}$$

For a cubic sample with linear dimensions L, substituting the trial function (26) in the action (25) yields

$$S = \frac{V(k_x^2 + k_y^2 + k_z^2 + 3\lambda^2)}{16\pi r_e} b^2 - \xi A V J_0^3(b) + (N+1) \ln \xi, \tag{27}$$

where $r_e \equiv 4\pi q^2 \beta$, $J_0(b)$ is the zero-order Bessel function of the argument b. If we assume that one charged particle is present at every lattice site and that the lattice is isotropic, we obtain $k_x = k_y = k_z = 2\pi n^{1/3}$, where $n = (N+1)/V$ is the particle density. Then, minimizing Eq. (27) with respect to ξ and b, we find that

$$S = \frac{3\pi}{4}\left[1 + \left(\frac{\lambda}{2\pi n^{1/3}}\right)^2\right] \frac{\tilde{b}^2}{\Gamma_e} (N+1) - (N+1) + (N+1) \ln \frac{N+1}{AVJ_0^3(\tilde{b})}, \tag{28}$$

where $\tilde{b}$ is governed by the equation

$$\frac{\pi}{2}\left[1 + \left(\frac{\lambda}{2\pi n^{1/3}}\right)^2\right] \frac{\tilde{b}}{\Gamma_e} + \frac{J_1(\tilde{b})}{J_0(\tilde{b})} = 0$$

with $\xi = (N+1)/AVJ_0^3(\tilde{b})$. Here $\Gamma_e \equiv r_e n^{1/3}$ is the coupling parameter equal to the Coulomb to kinetic energy ratio. Within the context of the general expressions (6) and (7), we obtain the partition function of the form

$$Z_N = \tilde{Z}_N^0 \left\{1 - \frac{3\pi}{4}\left[1 + \left(\frac{\lambda}{2\pi n^{1/3}}\right)^2\right] \frac{\tilde{b}^2}{\Gamma_e}\right\}^{N+1}, \tag{29}$$

where $\tilde{Z}_N^0$ is the partition function of the ideal Boltzmann gas with renormalized volume $\tilde{V} = VJ_0^3(\tilde{b})$.

It seems to be worthwhile to consider a one-dimensional analog of the system considered above since in this case the problem can be solved exactly. Physically this corresponds to one-dimensional molecular systems with free charges.

Let us consider a cylindrical body of length L and radius $r \ll L$. Let the Coulomb-repulsing charges lie on the cylinder axis. In this case we have

$$S = \frac{V}{L} \int_0^L dz \left\{\frac{1}{2r_e}\left(\frac{d\psi}{dz}\right)^2 - \xi A \cos\psi\right\} + (N+1) \ln \xi. \tag{30}$$

Then the Euler-Lagrange equation reduces to the sine-Gordon one, i.e.,

$$\frac{1}{r_e} \frac{d^2\psi}{dz^2} - \xi A \sin\psi = 0. \tag{31}$$

The first integral

$$\frac{1}{2r_e}\left(\frac{d\psi}{dz}\right)^2 + \xi A \cos\psi = C \tag{32}$$

corresponds to the solution with the period

$$l = \frac{1}{\sqrt{2r_e}} \int \frac{d\psi}{\sqrt{C - \xi A \cos\psi}} = \frac{4K(p)}{\sqrt{2r_e(C + \xi A)}}, \tag{33}$$

where $K(p)$ is the full elliptic integral of the first kind with the argument $p=\sqrt{2\xi A/(C+\xi A)}$. Substituting the solution (32) in the action (30) yields

$$S=2\xi AV\left\{\frac{2}{p^2}\frac{E(p)}{K(p)}-\frac{1}{p^2}+1\right\}-\xi AV+(N+1)\ln\,\xi. \tag{34}$$

Here $E(p)$ is the full elliptic integral of the second kind with the same argument p. The action (34) is extremum for $p=1$; this corresponds to the soliton solution given by

$$\psi=4\arctan\exp(z\sqrt{r_e\xi A}) \tag{35}$$

with the action

$$S=8\left(\frac{\xi A}{r_e}\right)^{1/2}\frac{V}{L}-\xi AV+(N+1)\ln\,\xi. \tag{36}$$

Thus we come to an expression for the partition function, i.e.,

$$Z_N=Z_N^0\left[1-\frac{8}{\sqrt{nr_eL^2}}\right]^{N+1}. \tag{37}$$

The multisoliton solution can be obtained in a manner similar to the analysis in Ref. [30].

The above model systems are homogeneous on the macroscopic scale. Particle distributions, however, can be spatially periodic. Moreover, this approach allows one to describe spatially inhomogeneous particle distributions, i.e., finite-size macroscopic clusters. Now let us consider model systems with interaction of the type that admits existence of such thermodynamically stable formations.

IV. CLUSTER FORMATION IN CONDENSED MEDIA

A. Gravitating gas model

Let us consider a system of particles whose interaction consists of gravitational attraction and hard sphere repulsion. For the Newtonian attraction, the inverse operator is known to be

$$W_{rr'}^{-1}=-\frac{1}{4\pi Gm^2}\Delta_{r'}\delta_{rr'},$$

where G is the gravitational constant, m is the particle mass, and Δ_r is the d'Alembert operator. Using the results of the hard sphere model yields an expression for the action, i.e.,

$$S=\int_{V_0}dV\left\{\frac{1}{4r_m}(\nabla\varphi)^2-\xi Ae^{\varphi}\right\}+\xi AV_0+(N+1)\ln\,\xi, \tag{38}$$

where $r_m\equiv 2\pi Gm^2\beta$, and the integration is carried out over the whole space except for the volume occupied by particles. An expression, analogous to Eq. (38), was obtained in Ref. [6]. However, the authors did not fix the number of particles and disregarded particle repulsion. The result of [6] can be supplemented with the solutions that allow for inhomogeneous particle distributions.

We introduce a dimensionless quantity $r=R/r_m$ and denote $\gamma^2\equiv\xi Ar_m^3$. Then the action (in spherical coordinates) can be written in terms of a new variable $\sigma=\exp(\varphi/2)$, i.e.,

$$S=4\pi\int_{r_0}^{\infty}\left\{\left(\frac{1}{\sigma}\frac{d\sigma}{dr}\right)^2-\gamma^2\sigma^2\right\}r^2dr+\xi AV_0+(N+1)\ln\,\xi. \tag{39}$$

If the cluster surface contribution in the action (39) is negligible, then the term $(2/r)d\sigma/dr$ can be omitted [27,28]. Then the saddle-point equation reduces to

$$\frac{d^2\sigma}{dr^2}-\frac{1}{\sigma}\left(\frac{d\sigma}{dr}\right)^2+\gamma^2\sigma^3=0. \tag{40}$$

The first integral of this equation is given by

$$\left(\frac{1}{\sigma}\frac{d\sigma}{dr}\right)^2+\gamma^2\sigma^2=\Delta^2, \tag{41}$$

where Δ^2 is an unknown integration constant. It should be noted that the first integral of Eq. (40) is similar to that of the nonlinear Schrödinger equation. The solution of Eq. (40) with the first integral (41) is given by

$$\sigma=\frac{\Delta}{\gamma}\frac{1}{\cosh\Delta r}. \tag{42}$$

Thus, introducing the ansatz $\varphi=\ln\sigma^2$ enabled us to find the solution of the nonlinear equation

$$\frac{1}{2}\frac{d^2\varphi}{dr^2}+\gamma^2e^{\varphi}=0,$$

which satisfies the extremum condition of the functional $S=\int r^2dr\{\frac{1}{4}(\nabla\varphi)^2-\gamma^2e^{\varphi}\}$. When considering the one-dimensional solution, we assumed that the variation range of φ or σ is smaller than the dimension of the soliton formation described by the solution (42). In our interpretation, any soliton solution corresponds to a spatially inhomogeneous particle distribution—a finite-size cluster. It depends on the interaction parameters, chemical potential, and temperature, which solution is realized. In the model under consideration, the soliton solution is associated with the case when, by virtue of gravitational attraction, particles are concentrated in a volume limited by their sizes. This corresponds to the solution (42) with the asymptotics $\sigma^2=1$, $\varphi=0$ for $r=d$, where d is the cluster size, $\sigma\to 0$, $\varphi\to-\infty$ as $r\to\infty$. Physically, this solution describes the presence of particles in the inhomogeneous formation of the size d and the absence of particles at infinity, since in this case the spatial distribution function is $f(r)=\xi Ae^{\varphi}$. Within the context of Eq. (42), the action (39) can be rewritten in the form

$$S=4\pi\int_{r_0}^{d}(\Delta^2-2\gamma^2\sigma^2)r^2dr+\gamma^2\frac{V_0}{r_m^3}+(N+1)\ln\frac{\gamma^2}{Ar_m^3}. \tag{43}$$

Now we perform integration,

$$2\gamma^2\int_{r_0}^{d}\sigma^2 r^2 dr = 2\Delta^2\int_{r_0}^{d}\frac{r^2 dr}{\cosh^2\Delta r} = \frac{2}{\Delta}\Bigg\{\Delta^2 d^2 \tanh\,\Delta d - \Delta^2 r_0^2 \tanh\,\Delta r_0 - 2\sum_{k=1}^{\infty}\frac{2^{2k}(2^{2k}-1)}{(2k+1)(2k)!}B_{2k}(\Delta\,d)^{2k+1} + 2k,$$

$$+2\sum_{k=1}^{\infty}\frac{2^{2k}(2^{2k}-1)}{(2k+1)2k!}B_{2k}(\Delta r_0\}^{2k+1},$$

where B_{2k} are the Bernoulli numbers, and expand the result in power series of $\Delta d \ll 1$. Then the action (43) reduces to

$$S = -\frac{\Delta^2}{r_m^3}(V - V_0) + \gamma^2\frac{V_0}{r_m^3} + (N+1)\ln\frac{\gamma^2}{A r_m^3}. \tag{44}$$

Inasmuch as $\sigma^2 = 1$ for $r = d$, we have $\gamma^2 \simeq \Delta^2(1 - \Delta^2 d^2)$. Substituting the last expression in (44), we find the action to be given by

$$S = \Delta^2\frac{V_0}{r_m^3} - \Delta^2\frac{V - V_0}{r_m^3} + (N+1)\ln\frac{\Delta^2}{A r_m^3} + (N+1)\ln(1 - \Delta^2 d^2) - \frac{V_0}{r_m^3}\Delta^4 d^2. \tag{45}$$

In the next step, we find the extremum of Eq. (45) with respect to Δ by means of the iteration method. Thus we obtain $\tilde{\Delta}^2 = r_m^3(N+1)/(V - 2V_0)$. We substitute the last expression in Eq. (45) to obtain the action in the form

$$S = -(N+1) + (N+1)$$
$$\times\ln\frac{N+1}{AV[1 - 2V_0/V + \tilde{\Delta}^2 d^2(1 - 2V_0/V)]}. \tag{46}$$

Now we have to find d. In what follows we shall see that its value can be found by minimizing the action. Here, however, we estimate it in terms of physical reasoning. Let us consider the condition for particle confinement within a cluster. For $r = 2r_0$, $\varphi = \varphi_0$ and hence $d = 2r_0 + \varphi_0/2\tilde{\Delta}$. If particles are not confined within the cluster, $d \simeq 2r_0$ and thus

$$\tilde{\Delta}^2 d^2\left(1 - \frac{2V_0}{V}\right) = \frac{(N+1)r_m^3 4r_0}{V} = \frac{V_0}{V}\frac{6Gm^2}{r_0}\beta.$$

Therefore, within the context of Eq. (46), the partition function of the system is given by

$$Z_N = Z_N^0\left[1 - \frac{2V_0}{V} + \frac{V_0}{V}\frac{6Gm^2}{r_0 kT}\right]^{N+1}. \tag{47}$$

It should be noted that the gravitating gas partition function thus obtained exactly reproduces the known expression [21,22] that was derived with calculating the virial coefficients. The proposed approach is more general because it allows one to estimate the partition function in the presence of clusters of arbitrary size $R = d r_m$, i.e.,

$$Z_N = Z_N^0\left[1 - \frac{2V_0}{V} + \frac{R^2 r_m(N+1)}{V}\right]^{N+1}, \tag{48}$$

where Z_N^0 is the partition function of the ideal Boltzmann gas.

B. Models with attraction and repulsion

Now we consider a system of particles whose interaction consists of attraction and repulsion. This problem cannot be solved in the general case. Let us reveal the main features of spatially inhomogeneous particle distribution formation in the cases when the inverse interaction operator is known. We consider the screened Coulomb repulsion and attraction. Using the known form of the inverse operators (10) yields

$$S = \int dV\Bigg\{\frac{1}{2r_m}[(\nabla\varphi)^2 + \chi^2\varphi^2] + \frac{1}{2r_e}[(\nabla\psi)^2 + \lambda^2\psi^2]$$
$$-\xi A e^{\varphi}\cos\,\psi\Bigg\} + (N+1)\ln\,\xi, \tag{49}$$

where $A \equiv (2\pi m/\beta\hbar^2)^{3/2}$ as before; χ^{-1} and λ^{-1} are the attraction and repulsion screening radii, respectively; $r_m \equiv 4\pi Q^2\beta$ and $r_e \equiv 4\pi q^2\beta$; Q^2 and q^2 are interaction constants.

The saddle-point equations are given by

$$\frac{1}{r_m}(\Delta\varphi - \chi^2\varphi) + \xi A e^{\varphi}\cos\,\psi = 0,$$

$$\frac{1}{r_e}(\Delta\psi - \lambda^2\psi) - \xi A e^{\varphi}\sin\,\psi = 0, \tag{50}$$

$$\int dV\xi A e^{\varphi}\cos\,\psi = N+1.$$

This set of nonlinear equations determines spatially inhomogeneous field distributions that correspond to the formation of finite-size clusters. In some cases, these equations can be solved analytically and thus the parameters of such formations can be found.

Let us consider the case when the effective change of the parameters of the system occurs for the distances $\lambda^{-1} < r$ and $\chi = 0$. Physically, this corresponds to the long-range attraction and short-range repulsion. Suppose that $\psi \ll 1$. We expand the second equation of the set (50) in power series of the "slow" field component ψ to obtain the relation

$$\xi A e^{\varphi}\,\frac{\psi^2}{2}=3\,\frac{\lambda^2}{r_e}+3\xi A e^{\varphi}.$$

Having substituted the latter in Eq. (49), we can write the effective action in the form

$$S_{\rm eff}=2\int d\tilde{V}\left\{\frac{1}{4}\,(\nabla\varphi)^2+\xi A r_m^3 e^{\varphi}+\frac{3}{2}\,\frac{\lambda^2}{r_e}\,r_m^3\right\}+(N+1)\ln\,\xi, \tag{51}$$

where the dimensionless length $\tilde{r}=R/r_m$ is introduced and the integration extends over the dimensionless volume $\tilde{V}$. The physical situation described by the effective action (51) corresponds to the long-range gravitational attraction and effective repulsion for distances smaller than the interaction radius λ^{-1}.

Let us introduce the notation $\gamma^2\equiv\xi A r_m^3$ and $\alpha^2\equiv3\lambda^2 r_m^3/2r_e$. Then we have the action for the function $\sigma=\exp(\varphi/2)$ in the form

$$S_{\rm eff}=2\int d\tilde{V}\left\{\left(\frac{1}{\sigma}\,\frac{d\sigma}{dr}\right)^2+\gamma^2\sigma^2+\alpha^2\right\}+(N+1)\ln\,\frac{\gamma^2}{A r_m^3}. \tag{52}$$

This functional crucially differs from Eq. (39): the sign of the second term is opposite and, moreover, it contains an additional term given rise to by the interaction renormalization in the presence of effective repulsion. The extremum condition of the action (52) is realized for the solution of the equation

$$\frac{d^2\sigma}{dr^2}-\frac{1}{\sigma}\left(\frac{d\sigma}{dr}\right)^2-\gamma^2\sigma^3=0 \tag{53}$$

with the first integral

$$\left(\frac{1}{\sigma}\,\nabla\sigma\right)^2-\gamma^2\sigma^2=\Delta^2. \tag{54}$$

The solution of the latter equation is given by

$$\tilde{\sigma}=\frac{\Delta}{\gamma}\,\frac{1}{\sinh\,\Delta(r-r')},$$

where r' is the soliton center coordinate.

As follows from the form of the distribution function $f(r)=A\xi e^{\varphi}$, this solution describes a spatially inhomogeneous particle distribution. We regard it as a finite-size cluster, with the cluster size to be found. If a multisoliton solution is realized, in which the soliton centers are dispersed but numbers of particles in the solitons are equal, then $S_{\rm eff}=nS^0_{\rm eff}$, where

$$S^0_{\rm eff}=\left[8\pi\int r^2dr\{\Delta^2+\alpha^2+2\gamma^2\tilde{\sigma}^2\}+k\,\ln\,\frac{\gamma^2}{A r_m^3}\right]. \tag{55}$$

Here n is the number of clusters and $k=(N+1)/n$ is the number of particles within a cluster. In a manner similar to the analysis of new phase bubbles formation [27,28], we write the effective action per cluster, i.e.,

$$S^0_{\rm eff}=8\pi\left\{\frac{1}{3}\,\tilde{R}^3(\Delta^2+\alpha^2)+2\gamma^2\tilde{R}^2S_1\right\}+k\,\ln\,\frac{\gamma^2}{A r_m^3}, \tag{56}$$

where $\tilde{R}$ is the dimensionless cluster size and

$$S_1=\int_{2r_0}^{R}\tilde{\sigma}^2dr=\int_{\sigma_0}^{1}\frac{\sigma d\sigma}{\sqrt{\Delta^2+\gamma^2\sigma^2}}.$$

In the last expression, the cluster center is assumed to lie at the spherical coordinate system origin. Actually, S_1 describes the cluster surface energy. The above formulas are valid when the transition layer thickness is considerably smaller than the cluster size [29].

For physical reasons, the asymptotic behavior of the solution is the following: $\sigma^2=1$ for $r=R$, then $\Delta\sim R^{-1}$ and $\gamma e\sim\Delta$; for $r=2r_0$, we have $\sigma_0\simeq(4\gamma^2r_0^2)^{-1}$. Thus we obtain $S_1\simeq-(2r_0\gamma^2)^{-1}$ and the effective action, in terms of cluster size, is given by

$$S^0_{\rm eff}\simeq8\pi\left\{\frac{\tilde{R}^3}{3}\left(\alpha^2+\frac{1}{\tilde{R}^2}\right)-\frac{\tilde{R}^2}{r_0}\right\}-k\,\ln(A r_m^3\tilde{R}^2). \tag{57}$$

Minimizing the action over $\tilde{R}$ yields the value of $\tilde{R}$. It is evident that the solution with finite size of spatially inhomogeneous particle distribution can be realized only for $\alpha^2Rr_0>3$. The phase transition occurs for $R=2r_0$, this corresponds to the condition $2\alpha^2r_0^2=3$. Thus we obtain the value of the transition temperature to be given by $\theta_c\simeq\pi b^2Q^2/r_0$, where $b=(Q/q)\lambda r_0\gg1$.

If in this case $\alpha R>1$, then the effective action reduces to

$$S^0_{\rm eff}\simeq\frac{8\pi}{3}\,\alpha^2\tilde{R}^3-k\,\ln(A r_m^3\tilde{R}^2). \tag{58}$$

Having minimized the action with respect to $\tilde{R}$, we find the cluster size to be $\tilde{R}_0^3=k/4\pi\alpha^2$, and the action to be given by

$$S^0_{\rm eff}=\frac{2}{3}\,k\left\{1-\frac{6}{\alpha^2\tilde{R}_0r_0}-\ln\,\frac{k}{4\pi\alpha^2}\,(A r_m^3)^{3/2}\right\}. \tag{59}$$

The minimum of this functional is realized for the optimum value of the number of particles within a cluster that is determined by the equation

$$k_c=\frac{4\pi\alpha^2}{(A r_m^3)^{3/2}}\exp\left(-\frac{24\pi}{\alpha^{4/3}k_c^{1/3}r_0}\right). \tag{60}$$

The critical size of a cluster is $\tilde{R}_c^3=k_c/4\pi\alpha^2$. Expanding the action (59) in power series in the vicinity of the critical value of the number of particles within a cluster, we obtain

$$S^0_{\rm eff}=\frac{2}{3}\,k_c\left[1-\frac{1}{2}\left(\frac{k}{k_c}\right)^2\right].$$

The probability of finding a cluster of k particles is $P\sim\exp(-S^0_{\rm eff})$.

At last, the free energy of the gas of noninteracting clusters is $F=-\theta\ln Z$. In our case, with regard for the zero modes [30], this reduces to

$$F=n\theta\left\{S_{\rm eff}^0-\frac{1}{2}\ln\frac{6}{\pi}S_{\rm eff}^0\right\}. \tag{61}$$

C. Model with long-range repulsion and short-range attraction

Now let us consider the contrary case when the repulsion range is longer than the attraction range, so that $\lambda=0$ and $\chi\neq 0$. We assume that $\varphi\ll 1$ and retain only the first term in the action expansion in power series of the field φ. Then we find from the first equation of the set (50) that $\tilde{\varphi}=(\xi A r_m/\chi^2)\cos\psi$ and substitute this value in the action (49). Thus we have

$$S_{\rm eff}=\int d\tilde{V}\left\{\frac{1}{2}(\nabla\psi)^2-\gamma^2\cos\psi-\gamma^2\alpha^2\cos^2\psi\right\}+(N+1)\ln\frac{\gamma^2}{Ar_e^3}, \tag{62}$$

where we have introduced the dimensionless length $\tilde{r}=R/r_e$, and denoted $\gamma^2\equiv\xi A r_e^3$ and $\alpha^2\equiv r_m/2\lambda^2 r_e^3$.

Since $\gamma\alpha\ll 1$, we obtain an equation for ψ in the spherically symmetric case, i.e.,

$$\frac{d^2\psi}{dr^2}+\frac{2}{r}\frac{d\psi}{dr}-\gamma\sin\psi=0. \tag{63}$$

In the general case, the solution of this equation describes a soliton that may be regarded as a spatially inhomogeneous formation. Similarly to Eq. (40), we can neglect the term $(2/r)d\psi/dr$ when the dimension of this formation is larger than the transition layer thickness [27,28]. In this case the first integral exists, i.e.,

$$\frac{1}{2}\left(\frac{d\psi}{dr}\right)^2+\gamma^2\cos\psi=C. \tag{64}$$

For $C=\gamma^2$, the solution of Eq. (63) is given by $\psi=\arctan\exp[-\gamma(r-r')]$. Its asymptotics is $\psi=\pi$ as $r\to 0$, and $\psi=0$ as $r\to\infty$. Physically this solution describes the formation of a pore in the continuum distribution of particles, i.e., absence of particles within a limited volume of the size $d\sim\gamma^{-1}$ which encloses the soliton center r'. In the case of a multisoliton solution, which corresponds to the formation of a finite number of pores in the system, we can write

$$S_{\rm eff}=4\pi n\int_0^d r^2dr\left[\left\{\frac{4\gamma^2(1+\gamma^2\alpha^2)}{\cosh^2(yr)}-\frac{4\gamma^4\alpha^2}{\cosh^4(yr)}\right\}-\gamma^2(1+\gamma^2\alpha^2)\right]-\gamma^2(1+\gamma^2\alpha^2)(V-V_d)+(N+1)\ln\frac{\gamma^2}{Ar_e^3}. \tag{65}$$

Here we could use the results of the previous subsection and write the effective action in terms of the pore size. Our purpose, however, is to show the possibility of describing spatially inhomogeneous formations by means of statistical methods only. In order to obtain the final result we make use of the integrals

$$4\pi\int_0^d\frac{r^2dr}{\cosh^2(yr)}=\tilde{V}_d\quad\text{and}\quad 4\pi\int_0^d\frac{r^2dr}{\cosh^4(\gamma r)}=\frac{5}{3}\tilde{V}_d,$$

where $\tilde{V}_d=4/3\pi d^3$. Thus we obtain the final expression for the action, i.e.,

$$S_{\rm eff}=4\gamma^2\left(1-\frac{2}{3}\gamma^2\alpha^2\right)\tilde{V}_d-\gamma^2(1+\gamma^2\alpha^2)\tilde{V}+(N+1)\ln\frac{\gamma^2}{Ar_e^3}. \tag{66}$$

The iteration procedure for γ^2 yields a simplified expression for the action: $S_{\rm eff}^0=-\gamma^2\tilde{V}+(N+1)\ln(\gamma^2/Ar_e^3)$. This corresponds to the approximation that all the pores occupy a volume that is small as compared to the system volume. Thus we obtain $\tilde{\gamma}^2=(N+1)/\tilde{V}$. Substituting this result in the action (66), we obtain the final expression for the partition function of the system with regard for the spatially inhomogeneous particle distribution, i.e.,

$$Z_N=Z_N^0\left\{1+\gamma^2\alpha^2-4\left(1-\frac{2}{3}\gamma^2\alpha^2\right)\frac{\tilde{V}_d}{\tilde{V}}\right\}^{N+1}. \tag{67}$$

If we set $\gamma\alpha\ll 1$ and $V_d=V_0$ (the particle volume), then we obtain the same result as for the hard sphere model (24). Evidently, the phase transition occurs for $\gamma\alpha\to 1$, which corresponds to the temperature $\theta_c\simeq 2\pi\rho Q^2/\lambda^2$. The last expression can be derived from the condition that the stability of homogeneous distribution in the system of likely charged particles is violated.

V. DISCUSSION

The results obtained in this paper illustrate the possibility to describe a system of interacting particles with regard for their spatially inhomogeneous distribution by means of the statistical theory. Representation of the partition function in terms of the functional integral over the auxiliary fields corresponds to the construction of an equilibrium sequence of probable states with regard for their weights. With the partition function being treated in this way, we can employ the methods of the quantum field theory. The extension to the complex plane provides a possibility to apply the saddle-point method and thus to select the system states whose contributions in the partition function are dominant. The solutions associated with the finite values of the "action" functional may be regarded as thermodynamically stable particle distributions. Whether the distribution is homogeneous or inhomogeneous, depends on the solutions that satisfy the extremum condition for the functional. Thus the spatially inhomogeneous distribution of the auxiliary fields can be unambiguously related to the spatially inhomogeneous particle distribution. It is also possible to find the parameters of such formations and the temperature of the phase transition accompanied by the formation of finite-size clusters of the new phase. Actually, this approach extends the average field approximation to involve into consideration spatially inhomogeneous field distributions.

In the proposed approach, there is no need to introduce two auxiliary fields that correspond to attraction and repulsion, respectively. We may introduce one complex field $\varphi + i\psi$ associated with interaction of any type, and carry out the procedure in the complex plane. We only have to know the inverse operator of the interaction. Dividing the interaction into several parts provides a better understanding of the mechanisms of spatially inhomogeneous particle distribution formation. Actually, this method describes the first kind phase transitions to the states that contain implantations of the new phase [31,32].

The proposed approach provides an advance in the study of the behavior of the clusters formed. In the case of multisoliton solutions, the residual interaction (uncompensated in the course of cluster formation) produces new spatial structures. The soliton interaction energy is described by an expression of the form $\omega_{rr'} = A \exp[-k(r-r')]$ [29,32]. Obviously, this system of clusters may be regarded as a gas of interacting particles and traditional methods of statistical physics, e.g., the simplest Ising model, may be employed to estimate the temperature of the phase transition to the spatially ordered state.

Thus, the proposed approach provides a unified statistical description for systems of interacting particles with regard for spatially inhomogeneous particle distributions. It also allows one to consider the collective behavior of such formations.

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