Accurate first-principle equation of state for the One-Component Plasma

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Abstract

Accurate "first-principle" expressions for the excess free energy $F_{ex}$ and internal energy $U_{ex}$ of the classical one-component plasma (OCP) are obtained. We use the Hubbard-Schofield transformation that maps the OCP Hamiltonian onto the Ising-like Hamiltonian, with coefficients expressed in terms of equilibrium correlation functions of a reference system. We use the ideal gas as a reference system for which all the correlation functions are known. Explicit calculations are performed with the high-order terms in the Ising-like Hamiltonian omitted. For small values of the plasma parameter $\Gamma$ the Debye-Huckel result for $F_{ex}$ and $U_{ex}$ is recovered. For $\Gamma \gg 1$, these depend linearly on $\Gamma$ in accordance with the Monte Carlo findings for the OCP. The MC data for the internal energy are reproduced fairly well by the obtained analytical expression.

I. INTRODUCTION

The one component plasma (OCP) model is one of the basic models in the condensed matter physics [1,2]. Besides of its direct astrophysical applications [2], to model the ionized
matter in white dwarfs [3], outer layers of neutron stars and interiors of heavy planets [4,5], OCP is widely used as a reference model for a variety of systems, ranging from alkali metals [6–8] to colloidal solutions [9–11]. The application of the OCP is not restricted by classical systems; it is also used when quantum effects are important [12].

The OCP model is formulated as a system of point particles, interacting via the Coulombic potential, which move in a uniform neutralizing background [12]. All thermodynamic properties of the OCP depend only on the dimensionless plasma parameter \( \Gamma = l_B/a_c \), where \( l_B = e^2/k_B T \) is the Bjerrum length (\( e \) is the charge of the particles, \( k_B \) is the Boltzmann’s constant, \( T \) is the temperature) and \( a_c = (3/4\pi \rho)^{1/3} \) is the ion-sphere radius with \( \rho = N/\Omega \) being the concentration of particles (\( N \) is the number of particles, \( \Omega \) is the volume of the system).

For small values of \( \Gamma \), which correspond to a hot and/or dilute system, the Debye-Huckel theory accurately describes the thermodynamic properties of the OCP. For the excess free energy and internal energy this gives in the \( \Gamma \to 0 \) limit:

\[
\frac{F_{ex}}{k_B T N} = -\frac{1}{\sqrt{3}} \Gamma^{3/2}, \quad \frac{U_{ex}}{k_B T N} = -\frac{\sqrt{3}}{2} \Gamma^{3/2}
\]  

(1)

Abe expansion [13] for the classical OCP provides next terms for that limit (in the present study we consider the classical case):

\[
\frac{F_{ex}}{k_B T N} = -\frac{1}{\sqrt{3}} \Gamma^{3/2} - \frac{c}{3} \Gamma^3 - \frac{1}{8} \Gamma^3 (3 \log \Gamma - 1) + \cdots 
\]

(2)

\[
\frac{U_{ex}}{k_B T N} = -\frac{\sqrt{3}}{2} \Gamma^{3/2} - \left( c + \frac{3}{8} \right) \Gamma^3 - \frac{3}{8} \Gamma^3 (3 \log \Gamma - 1) + \cdots 
\]

(3)

where \( c = \frac{9}{8} \log 3 + \frac{3}{2} \gamma - 1 = 1.101762 \ldots \), and \( \gamma \) is the Euler’s constant. The analytical Abe expansion seems to be fairly accurate for \( \Gamma \) up to 0.1 [14]. The next few terms for the \( \Gamma \to 0 \) expansion calculated in Ref. [13] allow to use the small-\( \Gamma \) expansion up to \( \Gamma \leq 0.4 \).

For larger values of \( \Gamma \) the OCP was studied numerically, by means of integral equations, such as Percus-Yevick, hypernetted-chain equations [16], and (most successively) by modified hypernetted-chain equation [17]. Extensive numerical studies have been performed.
by Monte Carlo (MC) \cite{15,16,20,21} and Molecular Dynamics \cite{22} technique. To fit available “experimental” data for the excess thermodynamic functions simple analytical fits were proposed \cite{23}:

\[
\frac{U_{ex}}{k_B T N} = A\Gamma + B\Gamma^s + C
\]

\[
\frac{F_{ex}}{k_B T N} = A\Gamma + \frac{B}{s}\Gamma^s + (3 + C) \log \Gamma - D
\]

with \(A = -0.8992, B = 0.596, s = 0.3253, C = -0.268\) and \(D = A + B/s + 1.1516\). Eqs.(4) and (5) are fairly accurate for the interval \(1 \leq \Gamma \leq 220\) \cite{23}, but unfortunately they do not give a correctly behavior at \(\Gamma \rightarrow 0\). Pade approximants for the \(U_{ex}\) suggested in \cite{24,25} remedy the failure of (4) at small \(\Gamma\); one thus obtains very precise description for the whole interval \(0 \leq \Gamma \leq 200\) \cite{25}.

To describe the limit of very large \(\Gamma\) a perturbation theory was proposed \cite{26}; it agrees well with the MC data for large \(\Gamma\), but also lack the proper small-\(\Gamma\) behavior. The correct Debye-Huckel behavior at \(\Gamma \rightarrow 0\) together with a reasonable \(\sim 10\%\) accuracy for \(0 \leq \Gamma \leq 100\) has been obtained in a simple semiphenomenological “Debye-Huckel plus hole” (DHH) theory \cite{27}; it fails, however, for \(\Gamma > 125\) \cite{27}. A modified DHH theory proposed recently \cite{28} using only one fitting parameter accurately reproduces the MC results for the large values of \(\Gamma\) \((1 < \Gamma < 200)\) and demonstrates a correct behavior at \(\Gamma \rightarrow 0\).

Thus, up to now, no “first-principle” theory of OCP exists which describes accurately the thermodynamic properties in the whole range of \(\Gamma\) from the Debye-Huckel limit \(\Gamma \rightarrow 0\) up to \(\Gamma \gg 1\) limit, where the Wigner crystallization \cite{2} occurs. In the present study we report a simple “first-principle” equation of state for the OCP which has the correct Debye-Huckel behavior for small \(\Gamma\) and demonstrates a linear dependence on \(\Gamma\) for \(\Gamma \gg 1\). It reproduces within \(1 - 2\%\) accuracy the experimental data for the most of range of \(\Gamma\) \((0 \leq \Gamma \leq 250)\) and has a typical deviation of the order of \(2 - 5\%\) (with a maximal one \(\approx 8\%\)) for \(0.01 < \Gamma < 1\). The rest of the paper is organized as follows: in the next Sec.II we consider the Hubbard-Schofield transformation that maps the OCP Hamiltonian onto the Ising-like Hamiltonian and calculate the coefficients of the effective Hamiltonian. In this section we also present the
field-theoretical formulation for the statistical sum of the OCP which directly follows from
the transformation used. Within the Gaussian approximation for the effective Hamiltonian
we derive the equation of state for the OCP. In Sec.III we discuss the equation of state
obtained and compare the analytical results for the internal excess free energy with the
available Monte Carlo data for the OCP. In the last Sec.IV we summarize our findings.

II. EFFECTIVE HAMILTONIAN AND EQUATION OF STATE FOR THE OCP

We start from the OCP Hamiltonian which may be written as follows ($\beta^{-1} = k_BT$):

$$H = \frac{1}{2} \beta^{-1} \sum_k \nu_k (\rho_k \rho_k - \rho) + H_{id} \quad (6)$$

where the first term in the right-hand side of Eq.(6) refers to the Coulombic interactions,
written in terms of the collective variables,

$$\rho_k = \frac{1}{\sqrt{\Omega}} \sum_{j=1}^{N} e^{-ikr_j} \quad (7)$$

where $r_j$ denotes coordinate of $j$-th particle, $\nu_k = 4\pi l_B^2/k^2$ and $H_{id}$ is the ideal-gas part of the
Hamiltonian. Summation in Eq.(6) is to be performed over the wave-vectors $k = \{k_x, k_y, k_z\}$
with $k_i = \frac{2\pi}{L_i} l_i$ ($i = x, y, z$), where $l_i$ are integers, $L^3 = \Omega$, and the prime over the sum denotes
that the term with $k = 0$ is excluded [29].

A. Hubbard-Schofield transformation

The configurational integral may be then written in terms of the configurational integral
of the reference (ideal gas) system $Q_R$ [30,31] as

$$Q = \left\langle \exp \left\{ -\frac{1}{2} \sum_k \nu_k (\rho_k \rho_k - \rho) \right\} \right\rangle_R \quad (8)$$

where $\langle (\cdots) \rangle_R = Q_R^{-1} \int d\mathbf{r}^N (\cdots)$ denotes the averaging over the reference system. In accord-
dance with the Hubbard-Schofield scheme [30] we use the identity,
\[ \exp\left(-\frac{1}{2}a^2x^2\right) = \left(2\pi a^2\right)^{-1/2} \int_{-\infty}^{+\infty} \exp\left(-\frac{1}{2}y^2/a^2 + ixy\right)dy \]

and arrive after some algebra at:

\[ Q = Q_R \int \prod_k c_k d\varphi_k \exp \left\{ -\frac{1}{2} \sum_k' \nu_k^{-1} \varphi_k \varphi_{-k} \right\} \left\{ \exp \left\{ i \sum_k' \rho_k \varphi_{-k} \right\} \right\}_R \tag{9} \]

where \( c_k = (2\pi \nu_k)^{-1/2} e^{\frac{1}{4} \nu_k \rho} \), and where the integration is to be performed under the restriction, \( \varphi_{-k} = \varphi_k^* \) (\( \varphi_k^* \) is the complex conjugate of \( \varphi_k \)). \[ \text{[32]} \]

Applying the cumulant theorem \[ \text{[33]} \] to the factor \( \langle \exp \left\{ i \sum_k' \rho_k \varphi_{-k} \right\} \rangle_R \) one obtains:

\[ Q = Q_R \int \prod_k c_k d\varphi_k e^{-\mathcal{H}}, \quad \text{with} \]

\[ \mathcal{H} = \sum_{n=2}^{\infty} \Omega^{1-\frac{n}{2}} \sum_{k_1,\ldots,k_n} \left. u_n \left( k_1, \ldots, k_n \right) \varphi_{k_1} \cdots \varphi_{k_n} \right| \]

\[ u_2 \left( k_1, k_2 \right) = \frac{1}{2} \delta_{k_1+k_2,0} \left\{ \frac{k_1^2}{4\pi l_B} + \langle \rho_{k_1} \rho_{-k_1} \rangle_c_R \right\} \]

\[ u_n \left( k_1, \ldots, k_n \right) = (-1)^n \frac{\Omega^{n-1}}{n!} \langle \rho_{k_1} \cdots \rho_{k_n} \rangle_{c_R} \quad n > 2 \tag{10} \]

here \( \langle \ldots \rangle_{c_R} \) denotes cumulative average \[ \text{[33]} \] for the reference ideal-gas system. As it follows from Eqs.(10), \( Q \) is written in a similar way as the partition function for the magnetic system having the Ising-like Hamiltonian \[ \text{[34]} \], where \( \varphi_{\vec{k}} \) are the Fourier components of the “spin-field”, \( \varphi(\vec{r}) \). Note that the coefficients of the effective Hamiltonian (10) are expressed in terms of the correlation functions of the ideal-gas system, which are perfectly known (e.g. \[ \text{[35]} \]):

\[ \Omega^{n-1} \langle \rho_{k_1} \cdots \rho_{k_n} \rangle_{c_R} = \rho \delta_{k_1+\cdots+k_n,0} \tag{11} \]

This yields the effective Hamiltonian:

\[ \mathcal{H} = \frac{1}{2} \sum_k' \left[ \rho + (4\pi l_B)^{-1} k^2 \right] \varphi_k \varphi_{-k} - \sum_{n=3}^{\infty} \frac{\Omega^{n-1}}{n!} \rho \sum_{k_1,\ldots,k_n} \varphi_{k_1} \cdots \varphi_{k_n} \delta_{k_1+\cdots+k_n,0} \]

\[ = \mathcal{H}_G + \mathcal{H}_1 \tag{12} \]

where we write explicitly the Gaussian part \( \mathcal{H}_G \):
\[ \mathcal{H}_G = \frac{1}{2} \sum_k' \left[ \rho + (4\pi l_B)^{-1} k^2 \right] \varphi_k \varphi_{-k} \]  

Since all the coefficients of the effective Hamiltonian are known, one can develop the usual perturbation expansion, with \( \mathcal{H}_G \) being a reference part of the Hamiltonian and with \( \mathcal{H}_1 \) being perturbation (e.g. [34,36,37]).

A simple structure of the effective Hamiltonian \( \mathcal{H} \), Eq.(12) suggests a simple, closed-form field-theoretical formulation for the partition sum of the OCP, akin the Sine-Gordon representation of the Coulombic gas [38], or the restricted primitive model of electrolytes [36,39].

**B. Field-theoretical model for the OCP**

Using the space-dependent field \( \varphi(r) \)

\[ \varphi(r) = \frac{1}{\sqrt{\Omega}} \sum_k \varphi_k e^{-ikr} \]

It is easy to show that under this transformation the terms in Eq.(12) containing products \( \varphi_k \cdots \varphi_k \) give rise to the terms \( \int dr \varphi^*(r) \), the terms \( k^2 \varphi_k \varphi_{-k} \) give rise to the term \( \int dr (\nabla \varphi)^2 \), so that one can write

\[ \mathcal{H} = \rho \int dr \left[ \frac{1}{2} (4\pi l_B \rho)^{-1} (\nabla \varphi)^2 - \sum_{n=2}^{\infty} \frac{i^n}{n!} \varphi^n \right] \]  

(14)

and recognize the expansion of \( e^{i\varphi} \).

Integration over the Fourier-components \( \varphi_k \) in Eq.(13) converts into "field"-integration over the field \( \varphi(r) \). It may be shown that the Jacobian of this transformation does not depend on \( \varphi(r) \) and appears as a normalization constant.

Noticing that

\[ \prod_k' (2\pi \nu_k)^{-\frac{1}{2}} = \left[ \int \prod_k' d\varphi_k \exp \left\{ -\frac{1}{2} \sum_k' \frac{k^2}{4\pi l_B} \varphi_k \varphi_{-k} \right\} \right]^{-1} \]

(with the restriction \( \varphi^*_k = \varphi_{-k} \)) and that

\[ \prod_k' e^{\frac{1}{2} \nu_k \rho} = \exp \left\{ \rho \Omega \frac{U(0)}{2k_B T} \right\} \]
where $U(0)$ is the so-called "self-energy" [38,40], one arrives after some algebra at the field-theoretical expression of the statistical sum $Z_{OCP}$ of the OCP:

$$e^{-\beta F} \equiv Z_{OCP} = \frac{\int \mathcal{D}\varphi \exp \left\{ -\mathcal{H}(\varphi) \right\}}{\int \mathcal{D}\varphi \exp \left\{ -\int \text{d}r \left[ \frac{\kappa_D^2}{2} (\nabla \varphi)^2 - \beta \tilde{\mu} \right] \right\}}$$  \hspace{1cm} (15)

where $\mathcal{D}\varphi$ denotes the "field"-integration,

$$\mathcal{H} = \int \text{d}r \left[ \frac{\kappa_D^2}{2} (\nabla \varphi)^2 + i\varphi - e^{i\varphi} \right],$$  \hspace{1cm} (16)

$k_D^2 = 4\pi l_B\rho = 4\pi e^2 \beta \rho$ is the inverse Debye screening length and

$$\tilde{\mu} = \mu_{id} + k_BT - U(0)/2.$$  \hspace{1cm} (17)

Here $\beta \mu_{id} = \log(\Lambda^3/\Omega)$ is the ideal-gas chemical potential ($\Lambda = \hbar/(2\pi mk_BT)^{1/2}$ is the thermal wavelength). Deriving Eq.(15) we write the statistical sum of the ideal gas as

$$Z_{id} = \exp \left\{ -N \log(\Lambda^3/\Omega) \right\} = \exp \left\{ -\beta \mu_{id} \rho \int \text{d}r \right\}$$  

and rescale the length $\rho \text{d}r \rightarrow \text{d}r$.

As it follows from Eq.(15) the potential function of the effective field-theoretical Hamiltonian for the OCP reads $V(\varphi) = i\varphi - e^{i\varphi}$. This may be compared with the potential function $V_{SG}(\varphi) = \cos \varphi$ of the Sine-Gordon model for the Coulombic gas [38]. Note, that all imaginary terms in Eq.(15) vanish after the field integration due to the symmetry properties of the Hamiltonian under the transformation $\varphi \rightarrow -\varphi$.

Consider now the "saddle-point" approximation to the numerator in Eq.(15). The equation for the "extremal" field which minimizes the effective Hamiltonian reads:

$$\nabla^2 \varphi = \kappa_D^2 \left( i - ie^{i\varphi} \right).$$  \hspace{1cm} (18)

Under transformation $\varphi = ie^2\phi/k_BT$ Eq.(18) converts into

$$\nabla^2 \phi = -4\pi \rho \left[ e^{-\epsilon\phi/k_BT} - 1 \right],$$  \hspace{1cm} (19)

which is the usual mean-field Poisson-Boltzmann equation for the OCP. This is not surprising since the "saddle-point" approximation is essentially the mean-field one.
C. Equation of state for the OCP

Now we concentrate on the Gaussian part of the effective Hamiltonian and show that even neglecting the non-Gaussian contribution to the effective Hamiltonian, one can obtain fairly accurate equation of state for the OCP, provided that a correct value of the ultraviolet cutoff in the $k$-space is used. The Gaussian approximation to $H$ corresponds actually to the Random Phase, or Debye-Hückel approximation, (e.g. [41]). Using $H_G$, Eq.(13) and performing (Gaussian) integration (e.g. [38]) over $\varphi_k$ in (10), one easily finds for the excess free energy of the OCP:

$$-\beta F_{ex} = \log\left(\frac{Q}{Q_R}\right) = \frac{1}{2} \sum_k [\rho \nu_k - \log (1 + \rho \nu_k)]$$  (20)

We argue that the summation in Eq.(21) should be carried out over a finite number of the wave-vectors $k$. In this we follow the Debye theory of the specific heat of solids (e.g. [42]). Namely, we assume that the total number of degrees of freedom in the system, $3N$, should be equal to the total number of physically different modes with the wave-vectors $k$ within the spherical shell of radius $k_0$ in the $k$-space. The number of modes is twice the number of the wave-vectors, since for each $k$ one has a sine and cosine mode (the amplitude of the $k$-th mode is a complex number) [43]. Thus we obtain:

$$2 \frac{\Omega}{8\pi^3} 4\pi \int_0^{k_0} k^2 dk = 3N$$  (21)

where the factor $\Omega/8\pi^3$ appears when the integration in $k$-space is used instead of summation. From Eq.(21) follows that $k_0 = (9\rho\pi^2)^{1/3}$. A similar Debye-like scheme to find the cutoff $k_0$ was first proposed for plasma in [14], where somewhat different value of the cutoff wave-vector was reported. Using the $k_0$ obtained we write:

$$-\frac{\beta F_{ex}}{N} = \frac{1}{2} \frac{\Omega}{8\pi^3} N \int_0^{k_0} k^2 dk \left[ \log \left(1 + \frac{\kappa_D^2}{k^2}\right) - \frac{\kappa_D^2}{k^2}\right]$$

$$= \frac{9}{4} \int_0^1 x^2 dx \left[ \log \left(1 + \frac{b\Gamma}{x^2}\right) - \frac{b\Gamma}{x^2}\right]$$  (22)

where $b = \frac{2}{3} \left(\frac{2}{\pi^2}\right)^{1/3}$. The last integral is easily calculated to obtain for the free energy
\[
\frac{F_{\text{ex}}}{k_B T N} = \frac{3}{4} \left[ \log (1 + b\Gamma) - b\Gamma \right] - \frac{3}{2} (b\Gamma)^{\frac{3}{2}} \arctan \left( \frac{1}{\sqrt{b\Gamma}} \right) 
\]  
(23)

and for the internal energy:

\[
\frac{U_{\text{ex}}}{k_B T N} = -\frac{9}{4} (b\Gamma)^{\frac{3}{2}} \arctan \left( \frac{1}{\sqrt{b\Gamma}} \right) 
\]  
(24)

of the OCP. To obtain Eq.(24) we use the relation \( U_{\text{ex}} = \Gamma \partial F_{\text{ex}} / \partial \Gamma \). Again we note that the same functional dependence for the excess internal energy (but with different coefficients) has been obtained in [44].

### III. RESULTS AND DISCUSSION

As it follows from Eqs.(23) and (24), for \( \Gamma \to 0 \) the Debye-Huckel behavior is recovered. On the other hand in the opposite limit \( \Gamma \gg 1 \) Eqs.(23) and (24) demonstrate the linear behavior on \( \Gamma \) with the leading term \(-A\Gamma\) in accordance with fits (4) and (5) to the MC data. The constant \( A \) reads

\[
A = \frac{9}{4} b = \frac{3}{2} \left( \frac{2}{\pi^2} \right)^{1/3} = 0.881 \ldots 
\]  
(25)

which is fairly close to the constant \( A = 0.899 \ldots \) of the fits (4), (5).

In Fig.1 the excess internal energy given by Eq.(24) is compared to the Monte Carlo data, taken from Ref. [14,18] for \( 0.1 \leq \Gamma \leq 1 \) and Ref. [3,20] for \( \Gamma > 1 \). Fig.2 shows the relative error of the analytical expression (24). As it follows from Fig.1 and Fig.2 the equation of state is fairly accurate in the most of range of the plasma parameter. The maximal deviation of the analytical expression from the numerical data occurs at the intermediate values of the plasma parameter, \( 0.01 < \Gamma < 1 \).

To analyze the reason of the enhanced deviation of the theoretical results from the numerical data at \( 0.1 < \Gamma < 0.5 \), one can address the small-\( \Gamma \) expansion of \( U_{\text{ex}} \) [28]. It was observed [28] that in spite of the correct Debye-Huckel limit, this does not reproduce correctly the next-order terms of the Abe expansion (3). This occurs due to limitations of the Gaussian approximation for the effective Hamiltonian. Since all the coefficients of
the effective Hamiltonian are known, one can go beyond the Gaussian approximation and
develop a usual perturbation scheme, based on the Gaussian Hamiltonian.

FIGURES

FIG. 1. Shows the dependence of the excess internal energy of the OCP $U_{ex}/Nk_B T$ on the
plasma parameter $\Gamma = l_B/a_c$ ($l_B = e^2/k_B T$, $a_c^{-3} = 4\pi \rho/3$). Points give the Monte Carlo data
(Ref. [14,18] for $0.1 \leq \Gamma \leq 1$ and Ref. [6,20] for $\Gamma > 1$); lines represent the theoretical result,
Eq. (24)

FIG. 2. Shows the relative error (%) of the analytical expression (24) for the excess internal
energy of the OCP $U_{ex}/Nk_B T$ as a function of the plasma parameter $\Gamma$. The Monte Carlo data
of Ref. [14,18] for $0.1 \leq \Gamma \leq 1$ and of Ref. [6,20] for $\Gamma > 1$ are used. For $\Gamma < 0.1$ Eq.(24) is
compared with the Abe expansion Eq.(3)
In particular the equation of state in a form of the virial expansion may be recovered \cite{37}. Unfortunately this expansion does not provide the closed analytical expression for the excess thermodynamic functions, which may be used with acceptable accuracy for all the range of $\Gamma$.

Thus, dealing with a problem where the relevant range of plasma parameter is not known in advance one should preferably use the simple closed-form equations suggested in the present study.

IV. CONCLUSION

A ”first-principles” equation of state for the one-component plasma is derived that has a correct Debye-Huckel behavior at the limit of small plasma parameter $\Gamma$ and demonstrates a linear dependence on $\Gamma$ at $\Gamma \gg 1$. The obtained coefficient 0.881 at the linear leading term is close to the corresponding coefficient 0.899 found in the Monte Carlo simulations. The simple analytical expression for the excess internal energy reproduces the MC data within $1 - 2\%$ accuracy for the most of range of $\Gamma$ ($0 \leq \Gamma \leq 250$) and has a typical deviation of the order of $2 - 5\%$ (with a maximal one $\approx 8\%$) for $0.01 < \Gamma < 1$.

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