Electrostatic Attraction of Coupled Wigner Crystals: Finite Temperature Effects

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Abstract

In this paper, we present a unified physical picture for the electrostatic attraction between two coupled planar Wigner crystals at finite (but below their melting) temperature. At very low temperatures, we find a new regime where the attraction, arising from the long-wavelength excitation of the plasmon mode, scales with the interplanar distance $d$ as $d^{-2}$. At higher temperatures, our calculation agrees with known results. Furthermore, we analyze the temperature dependence of the short-ranged attraction arising from “structural” correlations and argue that thermal fluctuations drastically reduce the strength of this attraction.

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I. INTRODUCTION

Electrostatic interactions play an important role in a system of charged macroions in an aqueous solution of neutralizing counterions [1]. The macroions may be charged membranes, stiff polyelectrolytes such as DNA, or charged colloidal particles. Recently, there has been a great interest in understanding the attraction arising from correlations between highly-charged macroions as evidenced in experiments [2] and in simulations [3]. This attraction cannot be explained by the standard Poisson-Boltzmann (PB) treatment, even for an idealized system of two highly charged planar surfaces, with counterions distributed between them, since PB, being a mean-field theory, neglects correlations. Indeed, it has been proven recently that PB theory predicts only repulsions between two likely-charged objects [4]. Recall that the PB solution [4] for a single charged surface with charge density \( en \) – where \( e \) is the elementary charge and \( n \) the areal density – immersed in a solution of neutralizing counterions of valence \( Z \), predicts a length scale \( \lambda_{GC} = 1/(2\pi l_B Z n) \) (where \( l_B = e^2/\varepsilon k_B T \approx 7 \AA \) is the Bjerrum length below which electrostatics dominates the thermal energy in an aqueous solution of dielectric constant \( \varepsilon = 80 (H_2O) \), \( k_B \) is the Boltzmann constant, and \( T \) is the temperature.) Physically, this Gouy-Chapman length \( \lambda_{GC} \) defines a sheath near the charged surface within which most of the counterions are confined [5]. For a moderately charged surface of \( n \sim 1/100 \AA^{-2} \), \( \lambda_{GC} \) is of the order of few angstroms, and for highly charged surfaces and multivalent counterions \( Z > 1 \), we have \( \lambda_{GC} < l_B \), signaling the breakdown of PB theory. In this limit, fluctuations and correlations about the mean-field potential become so large that the solution to the PB equation is no longer valid [6].

To account for the attraction arising from correlations, two distinct approaches have been proposed. The first approach, based on charge fluctuations, treats the “condensed” counterion fluctuations in the Gaussian approximation. This theory predicts a long-ranged attraction which vanishes as \( T \to 0 \) [7]. In the other approach based on “structural” correlations first proposed by Rouzina and Bloomfield [8], the attraction comes from the ground state configuration of the “condensed” counterions. Indeed, at low temperature, the
“condensed” counterions crystallize on the charged surface to form a 2D Wigner crystal. When brought together, the counterions of two Wigner crystals correlate themselves to minimize the electrostatic energy. These staggered Wigner crystals attract each other via a short-ranged force that is strongest at $T = 0$. Although the physical origin of the attraction is clear in each approach, the relationship between them remains somewhat obscure and their results in the $T \to 0$ limit are somewhat contradictory. Therefore, it is desirable to formulate a unified approach which captures the physics of both mechanisms.

To this end, we attempt in this paper to develop a detailed physical picture of the electrostatic interaction at finite temperature between two planar Wigner crystals in the strong Coulomb coupling limit. Since correlation effects are essentially two-dimensional, we consider a model system composed of two uniformly charged planes a distance $d$ apart, each having a charge density $\rho$. Confined on the surfaces are negative point-like mobile charges of magnitude $e$. In order to understand correlation effects that are not captured by PB theory, we assume that the charges form a system of interacting Wigner crystals (see Fig. 1). In particular, we compute the electrostatic attraction between the two layers by explicitly taking into account both correlated fluctuations and “structural” correlations. (By “structural” correlations, we mean the residual ground state spatial correlations which remain at finite temperature.) In the former case, we obtain a long-ranged force ($\sim 1/d^3$), which agrees with the result based on Debye-Hückel (Gaussian) approximation [7]. For the latter, a simple expression for the short-ranged force is derived, which shows that thermal fluctuations reduce its range, and which in the $T \to 0$ limit agrees with the known exponentially decaying result [8,9]. Furthermore, we argue that at zero temperature, there must also be a long-ranged force derived from the quantum fluctuations of the plasmons [9], in addition to this zero-temperature exponentially decaying force. At very low temperatures in the quantum regime, we obtain a new length scale $\lambda_L$ (to be defined below), where the attraction scales like $d^{-2}$ when $d < \lambda_L$.

It should be pointed out that we have assumed a uniform charge distribution on the surface of the charged plates in our model for electrostatic attraction, mediated by the
“condensed” counterions. This assumption of a uniform neutralizing background may not be a good approximation to real experimental settings, since charges on macroion surfaces are discrete. For monovalent counterions, they tend to bind to the charges on the surface and form dipolar molecules. Therefore, the ground state for this system may not be a Wigner crystal, which relies on mutual repulsion among charges for its stability, and short-ranged effects are likely to be important. However, for polyvalent counterions, a Wigner crystal is likely to form since each counterion does not bind to a particular charge on the surface, and a uniform background may be more appropriate. The detailed structure of the ground state as determined by short-ranged effects and valences will be the subject for another study.

Another point worth mentioning concerns the ordering of 2D solids which exhibit quasi-long-range-order (QLRO) [10]. It is well-known that a true long-range order is impossible for 2D systems with continuous symmetries. For a 2D solid, which may be described by continuum elasticity theory with nonzero long-wavelength elastic constants, the Fourier components of the density function $n(r) = \sum G n_G(r) e^{iG \cdot r}$ average (thermally) out to zero for a nonzero reciprocal lattice vector $G$, i.e. $\langle n_G(r) \rangle = \langle e^{iG \cdot u(r)} \rangle = 0$, where $u(r)$ are the displacements of the particles from their equilibrium positions, while the correlation function decays algebraically to zero: $\langle n_G(r) n^*_G(0) \rangle \sim r^{-\eta_G(T)}$ with $\eta_G(T) = \frac{k_B T G^2 (3\mu + \lambda)}{4\pi \mu (2\mu + \lambda)}$, where $\mu$ and $\lambda$ are Lamé elastic constants. This slow power-law decay of the correlation function is very different from the exponential decay one would expect in a liquid. Hence the term QLRO. For a single 2D Wigner crystal, QLRO implies that the thermal average of the electrostatic potential at a distance $d$ above the plane is zero at any non-zero temperature, in contrast to a perfectly ordered lattice ($T = 0$) where the electrostatic potential decays exponentially with $d$. This may lead to the conclusion that at finite temperatures the short-ranged force between two coupled Wigner crystals should likewise be zero. As we show below, this is not the case because the susceptibility, which measures the linear response of a 2D lattice to an external potential, nevertheless diverges at the reciprocal lattice vectors as in 3D solids [11].

This paper is organized as follows. In Sec. [11] we derive an effective Hamiltonian which
describes two interacting planar Wigner crystals starting from the zero temperature ground state. The total pressure is then decomposed into a long-ranged and a short-ranged component, which are evaluated in Sec. IIA and IIB respectively, and a detailed discussion of our results is presented in Sec. IIC. In Sec. II, we present an argument for a long-ranged attractive force arising from the zero-point fluctuations at zero temperature. In addition, we use the Bose-Einstein distribution to calculate the attractive long-ranged pressure in the quantum regimes.

II. EFFECTIVE HAMILTONIAN AND PRESSURE

We start with the Hamiltonian for two interacting Wigner crystals: \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{int} \). Here, \( \mathcal{H}_0 \) is the elastic Hamiltonian for two isolated Wigner crystals \(^2\)

\[
\beta \mathcal{H}_0 = \frac{1}{2} \sum_i \int \frac{d^2q}{(2\pi)^2} \Pi_{\alpha\beta}(q) u^{(i)}_{\alpha}(q) u^{(i)}_{\beta}(-q),
\]

where \( \beta^{-1} = k_B T \), \( u^{(i)}_{\alpha}(q) \) is the Fourier transform of the in-plane displacement field of the charges in the \( i \)th layer \( (i = A \text{ or } B) \), \( \Pi_{\alpha\beta}(q) = \left[ \frac{2\pi l_B n^2}{q} P^L_{\alpha\beta} + \mu P^T_{\alpha\beta} \right] q^2 \) is the dynamical matrix, \( \mu \approx 0.245 n^{3/2} l_B \) is the shear modulus \(^3\) in units of \( k_B T \), \( P^L_{\alpha\beta} = q_\alpha q_\beta / q^2 \) and \( P^T_{\alpha\beta} = \delta_{\alpha\beta} - q_\alpha q_\beta / q^2 \) are longitudinal and transverse projection operator, respectively. Here, Greek indices indicate Cartesian components. \( \mathcal{H}_{int} \) is the electrostatic interaction between the two layers:

\[
\beta \mathcal{H}_{int} = l_B \int d^2x d^2x' \frac{\left[\rho_A(x) - n\right] \left[\rho_B(x') - n\right]}{\sqrt{(x - x')^2 + d^2}},
\]

where \( \rho_i(x) \) is the number density of charges in the \( i \)th layer. In order to capture the long-wavelength coupling as well as discrete lattice effects which are essential for our discussions on the short-ranged force, we employ a method, similar to that in Ref. 14, which allows us to derive an effective Hamiltonian that is valid in the elastic regime where the density fluctuations are slowly varying in space, i.e. \( \nabla \cdot \mathbf{u}^{(i)}(x) \ll 1 \), but \( |\mathbf{u}^A(x) - \mathbf{u}^B(x)| \) need not be small compared to the lattice constant \( a \).
Let us introduce a slowly varying field for each layer:

\[ \phi_\alpha^{(i)}(x) = x_\alpha - u_\alpha^{(i)}[\bar{\phi}^{(i)}(x)], \]  

where the displacement field \( u^{(i)}(x) \) is defined in such a way that it has no Fourier components outside of the Brillouin Zone (BZ). Then, the density \( \rho_i(x) \) can be written as:

\[ \rho_i(x) = \sum_l \delta^2(R_l - \bar{\phi}^{(i)}(x)) \det[\partial_\alpha \phi_\beta^{(i)}(x)], \]  

where \( R_l \) are the equilibrium positions of the charges, i.e. the underlying lattice sites. Using the Fourier representation of the delta function and solving \( \phi_\alpha^{(i)}(x) \) iteratively in terms of the displacement field, we obtain a decomposition of the density for the \( i \)th layer into a slowly and a rapidly spatially varying pieces:

\[ \rho_i(x) - n \cong -n \nabla \cdot u^{(i)}(x) + \sum_{G \neq 0} n e^{i G \cdot [x + u^{(i)}(x)]}, \]  

where \( G \) is a reciprocal lattice vector. Note that we have neglected terms that are products of the slowly and the rapidly varying terms. Physically, the first term represents density fluctuations for wavelengths greater than the lattice constant, and the second term represents the underlying lattice, modified by thermal fluctuations. Using the density decomposition (5), \( \mathcal{H}_{\text{int}} \) may be written as

\[ \beta \mathcal{H}_{\text{int}} = \int \frac{d^2 q}{(2\pi)^2} \frac{2\pi l_B}{q} e^{-qd} \int d^2 x \int d^2 x' e^{i q \cdot (x - x')} \left( n \nabla \cdot u^A(x) - \sum_{G \neq 0} n e^{i G \cdot [x + u^A(x)]} \right) \times \left( n \nabla \cdot u^B(x') - \sum_{G' \neq 0} n e^{i G' \cdot [x' + u^B(x')]}, \right) \]

where \( c \) is the relative displacement vector between two lattices of the different plane and we have used the fact that \( \frac{1}{\sqrt{x^2 + d^2}} = \int \frac{d^2 q}{(2\pi)^2} e^{i q \cdot x \frac{2\pi}{q}} e^{-qd} \). Again neglecting the products of slowly and rapidly varying terms, which give vanishingly small contributions when integrating over all space, \( \mathcal{H}_{\text{int}} \) separates into two pieces: a long-wavelength term

\[ \beta \mathcal{H}_{\text{int}}^L = \int \frac{d^2 q}{(2\pi)^2} \frac{2\pi l_B n^2}{q} e^{-qd} q_\alpha q_\beta u_\alpha^A(q) u_\beta^B(-q), \]

and a short-wavelength term
\[
\beta H^S_{\text{int}} = + \sum_{G \neq 0} \sum_{G' \neq 0} \int \frac{d^2 q}{(2\pi)^2} \frac{2\pi l_B n^2}{q} e^{-qd} \times \int d^2 x \int d^2 x' e^{i q (x-x')} e^{i G \cdot [x + u^A(x)]} e^{i G' \cdot [x' + u^B(x')]}.
\] (8)

In order to obtain a tractable analytical treatment, we approximate this expression by splitting the sum over \(G'\) into two parts. The dominant part, with \(G' = -G\) is
\[
\beta H^S_{\text{int}} = - \sum_{G \neq 0} \int \frac{d^2 q}{(2\pi)^2} \frac{2\pi l_B n^2}{q} e^{-qd} \int d^2 x \int d^2 x' e^{i (q+G) \cdot (x-x')} e^{i G \cdot [u^A(x) - u^B(x')]},
\] (9)

where we have used \(e^{iG \cdot c} = -1\). The second part (those terms with \(G' \neq -G\)) contains extra phase factors which tend to average to zero in the elastic limit. As a first approximation, we neglect such terms. Finally, Eq. (9) can be systematically expanded using a gradient expansion:
\[
\beta H^S_{\text{int}} = - \sum_{G \neq 0} \Delta_G(d) \int d^2 x \cos \left\{ G \cdot \left[ u^A(x) - u^B(x) \right] \right\} + O(\partial_x u^i(\partial_y u^j)),
\] (10)

where \(\Delta_G(d) = \frac{4\pi l_B n^2}{G} e^{-Gd}\). Putting Equations (1), (7), and (10) together, we obtain an effective Hamiltonian for the coupled planar Wigner crystals:
\[
\beta \mathcal{H}_e = \beta H_0 + \frac{d^2 q}{(2\pi)^2} \frac{2\pi l_B n^2}{q} e^{-qd} q_\alpha q_\beta u^A_\alpha(q) u^B_\beta(-q) - \sum_{G \neq 0} \Delta_G(d) \int d^2 x \cos \left\{ G \cdot \left[ u^A(x) - u^B(x) \right] \right\}.
\] (11)

The second term in Eq. (11) comes from the long-wavelength couplings while the third term reflects the periodicity of the underlying lattice structure. This particular structure in the effective Hamiltonian, as will be demonstrated below, leads to a total force which is comprised of two pieces – an exponentially decaying (short-ranged) force and a long-ranged power-law force:
\[
\Pi(d) = -\frac{1}{A_0} \left\langle \frac{\partial H_{\text{int}}}{\partial d} \right\rangle_{\mathcal{H}_e} = -\frac{1}{A_0} \left\langle \frac{\partial H^S_{\text{int}}}{\partial d} \right\rangle_{\mathcal{H}_e} - \frac{1}{A_0} \left\langle \frac{\partial H^L_{\text{int}}}{\partial d} \right\rangle_{\mathcal{H}_e} = \Pi_{SR}(d) + \Pi_{LR}(d),
\] (12)

where \(A_0\) is the area of the plane. It is important to emphasize that both forces are present simultaneously, although each force dominates at a different spatial scale – the long-ranged force dominates at large separations while the short-ranged force at small separations.
To calculate various expectation values in Eq. (12), it is convenient to transform the displacement fields into in-phase and out-of-phase displacement fields by 

\[ u^+(x) = u^A(x) + u^B(x) \] and

\[ u^-(x) = u^A(x) - u^B(x), \] respectively, so that the effective Hamiltonian (11) separates into two independent parts: \( \mathcal{H}_e = \mathcal{H}_+ + \mathcal{H}_- \) with

\[ \beta \mathcal{H}_+ = \frac{1}{2} \int \frac{d^2q}{(2\pi)^2} \Pi_{\alpha\beta}(q) \ u^+_{\alpha}(q) \ u^+_{\beta}(-q), \] (13)

and

\[ \beta \mathcal{H}_- = \frac{1}{2} \int \frac{d^2q}{(2\pi)^2} \Pi_{\alpha\beta}(q) \ u^-_{\alpha}(q) \ u^-_{\beta}(-q) - \sum_{G \neq 0} \Delta_G(d) \int d^2x \ \cos[G \cdot u^-(x)], \] (14)

where \( \Pi_{\alpha\beta}^\pm(q) = \frac{1}{2} \left[ \frac{2\pi l_B n^2}{q} (1 \pm e^{-qd}) p_{\alpha\beta}^L + \mu p_{\alpha\beta}^T \right] q^2. \) Furthermore, at low temperature, where \( |u^-(x)| \) is small compared to the lattice constant \( a \), the cosine term in Eq. (14) can be expanded up to second order in \( |u^-(x)| \) to obtain the “mass” terms. Within a harmonic approximation \( \mathcal{H}_- \), up to an additive constant, may be written as

\[ \beta \mathcal{H}_- \simeq \frac{1}{2} \int \frac{d^2q}{(2\pi)^2} \Pi_{\alpha\beta}(q) \ u^-_{\alpha}(q) \ u^-_{\beta}(-q) + \frac{1}{2} \int \frac{d^2q}{(2\pi)^2} \left[ m_L^2 p_{\alpha\beta}^L + m_T^2 p_{\alpha\beta}^T \right] u^-_{\alpha}(q) \ u^-_{\beta}(-q). \] (15)

Here, \( m_{L,T} = \frac{4\pi l_B n^2}{\sum_{G \neq 0} G e^{-Gd}} = \frac{4\pi l_B n^2 \Delta_0(d)}{2}. \) Note that the mass terms vanish exponentially with \( d \) as also found in Ref. [14]. The fact that the transverse \( m_T \) and longitudinal “mass” \( m_L \) are degenerate is related to the underlying triangular structure of the lattices [15]. These “masses” are associated with the finite energy required to uniformly shear the two Wigner crystals, and thus give rise to a gap in the dispersion relations of the out-of-phase modes. In the next two subsections, we derive expressions for the long-ranged and the short-ranged pressure as given in Eq. (12) within the harmonic approximation.

**A. Long-ranged Pressure**

The long-ranged power-law force comes from the correlated long-wavelength density fluctuations (the plasmon modes). The shear modes do not contribute to this interaction since
\[ \partial_\alpha P_{\alpha\beta}^{(i)} u_\beta^{(i)}(x) = 0. \]

Using Eqs. (7) and (12), we obtain an expression for the long-ranged force:

\[ \beta \Pi_{LR}(d) = \frac{2\pi l_B}{A_0} \int \frac{d^2 q}{(2\pi)^2} e^{-qd} \langle \delta \rho_A(q) \delta \rho_B(-q) \rangle, \]

where \( \delta \rho_i(x) = -n \nabla \cdot u^{(i)}(x) \) is the long-wavelength density fluctuation to the lowest order. Making use of the equipartition theorem to evaluate

\[ \langle \delta \rho_A(q) \delta \rho_B(-q) \rangle \propto \int D u^\pm(q) \delta \rho_A(q) \delta \rho_B(-q) e^{-\beta[H_+ + H_-]} \]

and substituting this into Eq. (16), we have the result

\[ \Pi_{LR}(d) = -\frac{k_B T}{d^3} \alpha(\Delta_0 d), \]

where

\[ \alpha(x) = \frac{\zeta(3)}{8\pi} + \frac{x}{\pi} \left[ \text{Ci}(2\sqrt{x}) \cos(2\sqrt{x}) + \text{Si}(2\sqrt{x}) \sin(2\sqrt{x}) \right], \]

\( \zeta \) is the Riemann zeta function, and \( \text{Ci}(x) \) and \( \text{Si}(x) \) are the cosine and sine integral functions \[16\], respectively. In the large distance limit, the second term in Eq. (19) is exponentially suppressed and can be neglected, yielding \( \alpha = \frac{\zeta(3)}{8\pi} \). Therefore, for large \( d \) we have

\[ \Pi_{LR}(d) = -\frac{\zeta(3)}{8\pi} \frac{k_B T}{d^3}. \]

This is the well-known result from the Debye-Hückel approximation \[7\]. Note also that the amplitude \( \frac{\zeta(3)}{8\pi} \approx 0.048 \) is universal for this interaction, induced by the long wavelength fluctuations \[17\].

**B. Short-ranged Pressure**

The short-ranged force which decays exponentially owes its existence to the “structural” correlations. It survives even at non-zero temperature, in contrast to the conclusion drawn
from a single 2D Wigner crystal, as discussed in the Introduction. However, we expect on physical grounds the short-ranged force to be weakened by thermal fluctuations. To compute its temperature dependence explicitly, we start with the expression for this force derived from Eqs. (9) and (12):

$$\beta \Pi_{SR}(d) = -2\pi l_B n^2 \sum_{G \neq 0} e^{-\frac{G^2}{8\pi l_B^2 n^2}} f_G(d),$$

(21)

where $$f_G(d) = \int \frac{d^2 q}{(2\pi)^2} S(q - G) e^{-qd}$$, $$S(q - G) = \int d^2 r e^{i(q - G) \cdot r} e^{-\frac{q^2}{8\pi l_B^2 n^2} B^+(r) - B^-(r)}$$, and $$B^\pm(r) = \langle |u^\pm(r) - u^\pm(0)|^2 \rangle$$. Note that Eq. (21) is exact, provided all the averages are evaluated exactly. For a system of coupled perfect Wigner crystals at zero temperature, $$f_G(d) = e^{-Gd}$$. At finite temperature, but below the melting temperature $$T_m$$, we note that $$B^\pm(r)$$ varies very slowly in space, so that $$f_G(d)$$ can be approximated by its zero temperature value: $$f_G(d) \approx e^{-Gd}$$. Hence, we obtain

$$\beta \Pi_{SR}(d) \approx -2\pi l_B n^2 \sum_{G \neq 0} e^{-Gd} \langle e^{iG \cdot [u^A(0) - u^B(0)]} \rangle_{\mathcal{H}_e}. $$

(22)

The thermal average of the displacement fields in Eq. (22) resembles a “Debye-Waller” factor and indicate the degree to which the short-ranged force is depressed by thermal fluctuations from its zero temperature maximum value. Because of the cosine term present in Eq. (11), this “Debye-Waller” factor is in general not zero, unlike the case of a single 2D Wigner crystal. However, if the system has melted into a Coulomb fluid, this cosine term, which comes from the lattice structure, would have to be modified.

The required expectation value in Eq. (22) only involves $$\mathcal{H}_e$$. Within the harmonic approximation, the mean-square out-of-phase displacement field can be evaluated

$$\langle |u^{-}(x)|^2 \rangle \approx \frac{\lambda_D}{2\pi n d} \ln \left[ \frac{d}{4\Delta_0(d) a^2} \right] + \frac{1}{2\mu} \ln \left[ \frac{\mu}{8\pi l_B n^2 \Delta_0(d) a^2} \right] \approx G_0 d \left[ \frac{\lambda_D}{2\pi} \frac{1}{n d} + \frac{1}{\mu} \right],$$

(23)

where $$\lambda_D = 1/(2\pi l_B n)$$, $$a$$ is the lattice constant, $$\mu \approx 0.245 n^{3/2} l_B$$ is the shear modulus of an isolated Wigner crystal in units of $$k_B T$$, and in the last line, we have approximated $$\Delta_0(d)$$ by the first nonzero reciprocal lattice vector contribution: $$\Delta_0(d) \approx G_0 e^{-G_0 d}$$. Note also that the logarithmic dependence on the “mass” ($$= 4\pi n^2 l_B \Delta_0(d)$$) is a characteristic of 2D solids.
Inserting Eq. (23) into Eq. (22), we obtain an expression for the short-ranged pressure at finite temperatures

\[ \beta \Pi_{SR}(d) \simeq -2\pi l_B n^2 e^{-(1+\xi/2)G_0d}. \]  

(24)

Here, the parameter \( \xi \) defined by

\[ \xi = \frac{G_0^2}{2\pi} \left( \frac{\lambda_D}{nd} + \frac{1}{\mu} \right), \]

(25)

characterizes the relative strengths of thermal fluctuations and the electrostatic energy of a Wigner crystal, i.e. \( \xi \sim \frac{k_B T a}{e^2} \). Thus, the sole effect of thermal fluctuations on the short-ranged force is to reduce its range: \( G_0 \rightarrow G_0 \left( 1 + \frac{\xi}{2} \right) \).

C. Discussion of Results

In summary, we have shown that the total pressure can be decomposed into a long-ranged \( \Pi_{LR} \) and a short-ranged pressure \( \Pi_{SR} \). Each force is computed at low temperatures, where the harmonic approximation is expected to be valid. The result for the total force is

\[ \beta \Pi(d) \simeq -2\pi l_B n^2 e^{-(1+\xi/2)G_0d} - \frac{\alpha(\Delta_0d)}{d^3}, \]

(26)

where \( \xi = \frac{G_0^2}{2\pi} \left[ \frac{\lambda_D}{nd} + \frac{1}{\mu} \right] \) and \( \alpha(\Delta_0d) = \frac{\xi(3)}{8\pi} \) for large \( d \). In Fig. 2, we have plotted \( \Pi_{SR} \) and \( \Pi_{LR} \) for two values of the coupling constant, \( \Gamma \equiv \frac{l_B}{a} = 150 \) and 50. Not surprisingly, they show that \( \Pi_{SR} \) dominates for small \( d \), and \( \Pi_{LR} \) for large \( d \). However, it is interesting to note that even for high values of \( \Gamma \), \( \Pi_{LR} \) dominates as soon as \( d \sim a \).

According to Eq. (24), the magnitude of \( \Pi_{SR} \) tends to decrease exponentially with temperature, as illustrated in Fig. 3. This strong decrease with increasing temperature is consistent with the Brownian dynamics simulations of Grønbech-Jensen et al. [3]. The shortening of its range may be attributed to the generic nature of strong fluctuations in 2D systems, and can also be understood by the following scaling argument. Referring back to \( \mathcal{H}_- \) in Eq. (14), one can show that the anomalous dimension of the operator \( \cos[\mathbf{G}_0 \cdot \mathbf{u}^{-}(\mathbf{x})] \) is \( [\text{Length}]^{-\xi} \) and correspondingly the dimension of \( \Delta_{\mathbf{G}_0}(d) \) is \( [\text{Length}]^{\xi-2} \). Since \( \Delta_{\mathbf{G}_0}(d) \)
is the only relevant length scale in $\mathcal{H}_-$, we must have $\langle e^{i G_0 \cdot u^- (0)} \rangle \sim \Delta_{G_0}^{xx}$. Therefore, the short-ranged pressure scales like

$$
\Pi_{SR}(d) \sim -\Delta_{G_0}(d) \times \langle e^{i G_0 \cdot u^- (0)} \rangle \sim -\Delta_{G_0} \times \Delta_{G_0}^{xx} \sim -e^{-G_0 d (\xi^2/2^2)}.
$$

(27)

In the low temperature limit ($\xi \ll 1$), we see that the range of $\Pi_{SR}$ is $G_0 \left(1 + \xi^2/2\right)$ as in Eq. (24). This scaling argument also suggests that at higher temperatures thermal fluctuations may have interesting nonperturbative effects. At zero temperature $\xi = 0$, so $\Pi_{SR}$ in Eq. (24) reproduces the known result of exponentially decaying attractive force [8,9].

The long-ranged pressure for large $d$ in Eq. (20) agrees exactly, including the prefactor, with the Debye-Hückel approximation. This is hardly surprising since the existence of long-wavelength plasmons (average density fluctuations) is independent of local structure, and they are present for solids and fluids alike. Thus, the asymptotic long-ranged power-law force must manifest itself even after QLRO is lost via a 2D melting transition driven by dislocations [19]. Therefore, our formulation captures the essential physics of the attraction not only arising from the ground state “structural” correlations, but also from the high temperature charge-fluctuations.

III. QUANTUM CONTRIBUTIONS TO THE LONG-RANGED ATTRACTION

According to the classical calculations above, correlation effects give rise to a “structural” short-ranged and a long-ranged attractive force. Recall that the long-ranged force vanishes as $T \to 0$, and that the short-ranged force is strongest at zero temperature but vanishes exponentially with distance. This observation suggests that for sufficiently large separations correlated attractions at finite temperatures are stronger than those arising from the perfectly correlated zero temperature ground state. However, we have pointed out in Ref. [9] that zero-point fluctuations of the plasmons lead to an attractive long-ranged interaction, which exhibits an unusual fractional-power-law decay ($\sim d^{-7/2}$), in contrast to the zero-temperature van der Waals interaction ($\sim d^{-4}$). Hence, in the $T \to 0$ limit, this
"zero-point attraction" dominates the short-ranged "structural" force at large separations. Furthermore, we expect that quantum fluctuations persist at finite temperature, and in this section, we compute their temperature dependence.

Within the harmonic approximation to the effective Hamiltonian, the dispersion relations for the plasmons can be readily obtained [9]:

\[
\omega_1^2(q) = \frac{8\pi e^2 n}{m\epsilon} \Delta_0(d) + \frac{2\pi e^2 n}{m\epsilon} q \left(1 - e^{-qd}\right); \\
\omega_2^2(q) = \frac{2\pi e^2 n}{m\epsilon} q \left(1 + e^{-qd}\right),
\]

where \(m\) is the mass of the charges and \(\Delta_0(d) \sim e^{-Gd}\) is proportional to the energy gap (the "mass" term) for the out-of-phase mode. The plasmon modes are related to the correlated charge-density fluctuations in the two layers. At any finite temperature, the free energy of the low-lying plasmon excitations is given by

\[
\mathcal{F}(d)/A_0 = \frac{\hbar}{2} \sum_{i=1,2} \int \frac{d^2q}{(2\pi)^2} \omega_i(q) + k_B T \sum_{i=1,2} \int \frac{d^2q}{(2\pi)^2} \ln \left[1 - e^{-\beta \omega_i(q)}\right],
\]

where \(A_0\) is the area of the plane. Since the energy gap \(\Delta_0\) is exponentially damped for large distances, its contribution to the free energy may be neglected in the large distance limit, where the long-ranged force is expected to be dominant.

The first term in Eq. (30) arising from the zero-point fluctuations has been computed in Ref. [9] and gives the \(d^{-7/2}\) power-law mentioned above. An additional contribution to the pressure at finite temperature arises from the second term in Eq. (30),

\[
\beta \Pi_{LR}(d) = -\frac{\hbar \Lambda}{4\pi d^{7/2}} \int_0^\infty dx \frac{1}{x^{5/2}} \left\{ \frac{1}{\exp[\eta \sqrt{x(1 - e^{-x})}] - 1} \frac{e^{-x}}{\sqrt{1 - e^{-x}}} \right. \\
\left. - \frac{1}{\exp[\eta \sqrt{x(1 + e^{-x})}] - 1} \frac{e^{-x}}{\sqrt{1 + e^{-x}}} \right\},
\]

where \(\Lambda = \sqrt{2\pi e^2 n / m\epsilon}\) and \(\eta = \beta \hbar \Lambda / \sqrt{d}\). We can evaluate this expression in two limits:

In the low-temperature limit \(\eta \gg 1\), Eq. (31) can be systematically expanded in powers of \(\eta^{-1}\). The lowest order term is given by \(\Pi_{LR}(d) = -\alpha k_B T / \lambda_L d^2\), where \(\lambda_L \equiv a_B L_B^2\), \(a_B \equiv e\hbar^2 / (m\epsilon^2)\) is the effective Bohr radius, \(\alpha \equiv \frac{1}{4\pi} \int_0^\infty dx \frac{x^2}{e^x - 1} = \zeta(3)/(2\pi)\), and \(\zeta\) is the Riemann
zeta function. We observe that the low temperature condition $\eta > 1$ is equivalent to the short distance limit $d < \lambda_L$.

In the high temperature limit $\eta \ll 1$ or the large distance limit $d > \lambda_L$, we expand the exponential in the denominator of Eq. (31) to obtain $\Pi_{LR}(d) = -\alpha \frac{k_B T}{d^3}$, where $\alpha = \frac{\zeta(3)}{(8\pi)}$. This result agrees with the classical calculation in Sec. II A as it should. Therefore, we have the following regimes for correlated attraction from plasmon fluctuations at finite temperature

$$\Pi_{LR}(d) \sim \begin{cases} 
-k_B T/d^3, & \text{for } \lambda_L < d, \\
-k_B T/(\lambda_L d^2), & \text{for } \lambda_L > d.
\end{cases}$$

(32)

We note that $\lambda_L$, in contrast to $\lambda_D$, increases with decreasing temperature, indicating, as one might expect, that quantum fluctuations are important at low temperatures. Furthermore, since $\Pi_{LR}(d) \to 0$ as $T \to 0$, the attractive interaction as $T \to 0$ is governed by zero-point fluctuations as emphasized above. In the strong Coulomb coupling limit $l_B/\lambda_D \sim 100$, we get $\lambda_L \sim 3\,\text{Å}$ for $\epsilon \sim 100$ and $a_B \sim 1/20\,\text{Å}$. Finally, it should be emphasized that quantum contributions to the long-ranged attraction are unlikely to be relevant for macroions. Our motivation here stems from the desire to understand the charge-fluctuation-induced attraction between coupled layers in a complete picture. However, our results may be relevant for electrons in bilayer semiconductor systems. Indeed, there are recent theoretical efforts devoted to this subject [20].

IV. DISCUSSION AND CONCLUSION

In this paper, we have studied analytically the electrostatic attraction between two planar Wigner crystals in the strong Coulomb coupling limit. We show that the total attractive pressure can be separated into a long-ranged and short-ranged component. The long-ranged pressure arises from correlated fluctuations and the short-ranged pressure from the ground state “structural” correlations. We also compute the very low temperature behavior of the fluctuation-induced attraction, where long-wavelength plasmon excitation must be described
by Bose-Einstein statistics. The results are summarized in Fig. 4, showing different regimes for the charge-fluctuation-induced long-ranged attraction, including the high temperature results in Ref. [7] and the characteristic decay length $l_{SR}$ for the short-ranged force. For small $d$, the short-ranged force is always dominant, but the decay length shrinks with increasing temperature. The crossover from the short-ranged to long-ranged dominant regimes occurs about $d \sim a$. Thus, for large $d \gg a$ only the long-ranged force is operative, which crosses over from $d^{-7/2}$ at zero temperature to the finite temperature distance dependence of $d^{-2}$ if $d < \lambda_L$ and $d^{-3}$ if $d > \lambda_L$. This provides a unified description to the electrostatic attraction between two coupled Wigner crystals.

In addition, our formulation may offer further insights into the nature of the counterion-mediated attraction at short distances. As discussed in Sec. II B, the reason that the short-ranged force in Eq. (22) does not vanish is because of the cosine term in $\mathcal{H}_-$, which represents the underlying lattice structures, and our results indicate that the strength of the short-ranged force decreases exponentially with temperature. However, at higher temperatures the expression for $\Pi_{SR}$ in Eq. (24) is no longer valid, since the harmonic approximation breaks down. Indeed, the scaling argument leading to Eq. (27) suggests that if the full cosine term is retained, $\Pi_{SR}$ may exhibit nonperturbative behaviors as $\xi \to 2^-$. 

To discuss qualitatively what happens at higher temperatures, we assume that $\Delta G(d)$ is sufficiently small and the system of interacting Wigner crystals is below its melting temperature $T_m$. Then, the charges between the two layers may unlock via a Kosterlitz-Thouless (KT) type of transition, determined by the relevancy of the cosine term in $\mathcal{H}_-$, at $\xi = 2$ [21]. (An order of magnitude estimate for the coupling constant is $\Gamma \sim 13$.) In the locked phase, $\xi \ll 2$, the periodic symmetry in $\mathcal{H}_-$ is spontaneously broken, and the resulting state is well captured by the harmonic approximation. On the other hand, when $\xi > 2$ the fluctuations are so large that the ground state becomes nondegenerate (gapless), i.e. the layers are decoupled. To compute $\Pi_{SR}$ in the unlocked phase, $\mathcal{H}_{int}^S$ given in Eq. (14) can be treated as a perturbation in evaluating the “Debye-Waller” factor in Eq. (22). To the lowest order, we obtain
\[ \Pi_{SR}(d) \simeq -\frac{k_B T}{\lambda_D^2 a} \left( \frac{\xi - 1}{\xi - 2} \right) e^{-2G_0d}. \] (33)

We first note that this expression diverges as \( \xi \to 2^+ \), indicating the breakdown of the perturbation theory as the temperature is lowered. Furthermore, in contrast to Eq. (24), the range of \( \Pi_{SR} \) remains constant and the amplitude acquires a temperature dependence of \( \sim 1/T \) (for large \( \xi \gg 2 \)), reminiscent of a high temperature expansion.

However, the above picture may be modified if the charges have melted into a Coulomb fluid via a dislocation-mediated melting transition [19] before \( \xi \to 2^- \). If this is the case, further analysis is necessary to obtain a more complete picture of the high temperature phase. Although the spatial correlations in a system of coupled 2D Coulomb fluids are expected to be somewhat different from 2D Wigner crystals, the solid phase results above suggest a qualitative lower limit of \( \Gamma \sim 13 \) at which \( \Pi_{SR} \) crosses over from low temperature to high temperature behavior. It may be of interest to note that in Ref. [1], an estimate for the upper limit of \( \Gamma \) at which the Poisson-Boltzmann equation breaks down is of the order of \( \Gamma \sim 3 \). To describe the melting of coupled 2D Wigner crystals, excitations of dislocations must be introduced into the effective Hamiltonian Eq. (11) similar to what is done in Ref. [22]. These considerations may help to establish an analytical theory of the attraction arising from counterion correlations, not captured by the Poisson-Boltzmann theory. The present formulation is a first step in that direction.

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FIG. 1. A schematic picture of two staggered Wigner crystals formed by the “condensed” counterions at very low temperatures.
FIG. 2. Plots of $\Pi_{SR}$ and $\Pi_{LR}$ versus $d$ for $\Gamma = 150$ (a) and 50 (b). Observe that the crossover ($\Pi_{LR} \approx \Pi_{SR}$) occurs at about $d \sim a$. $\Pi_0 \equiv k_B T (l_B/a^4) \times 10^{-3}$. 
FIG. 3. A plot of $\Pi_{SR}$ as a function of $\Gamma^{-1}$ at $d = 1.2a$, according to Eq. (24), which shows that $\Pi_{SR}$ exponentially decreases with increasing temperature.
FIG. 4. A schematic phase diagram summarizing different charge-fluctuation-induced attraction regimes. The characteristic decay length $l_{SR}$ of the short-ranged force is also shown.