Electrostatic attraction of coupled Wigner crystals: Finite temperature effects

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In this paper we present a unified physical picture for the electrostatic attraction between two coupled planar Wigner crystals at finite temperature. This model may facilitate our conceptual understanding of counterion-mediated attractions between (highly) similarly charged planes. By adopting an elastic theory, we show that the total attractive force between them can be (approximately) decomposed into a short-ranged and a long-ranged component. They are evaluated below the melting temperature of the Wigner crystals. In particular, we analyze the temperature dependence of the short-ranged attraction, arising from ground-state configuration, and we argue that thermal fluctuations may drastically reduce its strength. Also, the long-range force agrees exactly with that based on the charge-fluctuation approach. Furthermore, we take quantum contributions to the long-ranged (fluctuation-induced) attraction into account and show how the fractional power law, which scales as $d^{-7/2}$ for large interplanar distance d at zero temperature, crosses over to the classical regime d^{-3} via an intermediate regime of d^{-2}.

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I. INTRODUCTION

Electrostatic interactions play an important role in systems of charged macroions in aqueous solutions 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 attractive force arising from correlations between highly charged macroions as evidenced in experiments [2] and in simulations [3]. This attraction is not contained in the standard Poisson-Boltzmann (PB) treatment, even for an idealized system of two highly charged planar surfaces. Indeed, it has been proven recently that PB theory predicts only repulsions between two likely charged objects [4]. Recall that the PB solution [1] 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 $\lambda_G = 1/(2\pi l_B Z n)$ [where $l_B \equiv e^2/\epsilon k_B T \approx 7$ Å is the Bjerrum length below which electrostatics dominates the thermal energy in an aqueous solution of dielectric constant $\epsilon = 80$ (H_2O), k_B is the Boltzmann constant, and T is the temperature]. Physically, this Gouy-Chapman length λ_G defines a sheath near the charged surface within which most of the counterions are confined [5]. For a moderately charged surface of $n \sim 1/100$ Å^{-2}, λ_G is of the order of a few angstroms, and for highly charged surfaces and multivalent counterions $Z > 1$, we have $\lambda_G < l_B$, 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 no longer provides a reasonable approximation [6].

To account for the attraction arising from correlations, two distinct approaches have been proposed [7,8]. The first approach, based on charge fluctuation, treats the ''condensed'' counterion (in-plane) fluctuations in the Gaussian approximation. This theory predicts a long-ranged attraction which scales with the interplaner distance as d^{-3} and vanishes as $T \to 0$ [7]. Note that this long-ranged force belongs to the general class of fluctuation-induced forces [9]. In the other approach based on ''structural'' correlations proposed by Rouzina and Bloomfield [8], the attraction comes from the ground-state configuration of the condensed counterions. Indeed, at sufficiently low temperature, the condensed counterions crystallize on the charged surface to form a two-dimensional (2D) Wigner crystal. When brought together, the counterions of two Wigner crystals correlate themselves to minimize the electrostatic energy (see Fig. 1). The pressure between them can easily be calculated at zero temperature [8,10]

$$\Pi_{SR}(d) = -\frac{\partial}{\partial d}\left\{ \frac{e^2 n}{\epsilon} \sum_l \frac{1}{\sqrt{|\mathbf{R}_l + \mathbf{c}|^2 + d^2}} - \frac{(en)^2}{\epsilon} \int \frac{d^2\mathbf{r}}{\sqrt{\mathbf{r}^2 + d^2}} \right\} \simeq -\frac{2\pi (en)^2}{\epsilon} e^{-G_0 d} \quad (1)$$

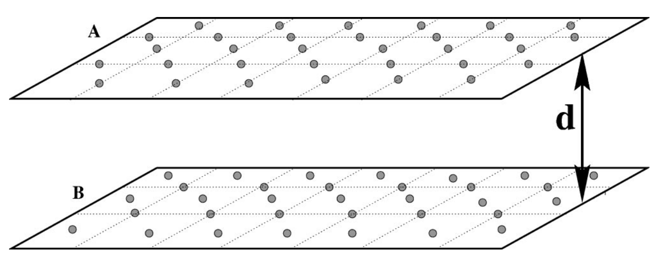

FIG. 1. A schematic picture of two staggered Wigner crystals formed by the ''condensed'' counterions at very low temperatures.

for large d, where $\mathbf{R}_l$ are the lattice sites, $\mathbf{c}$ is the relative displacement vector between two lattices of the different plane, $G_0 \equiv 4\pi/(\sqrt{3}a)$ is the magnitude of the first reciprocal-lattice vector, and $a \equiv \sqrt{2/(\sqrt{3}n)}$ is the lattice constant. Hence, these staggered Wigner crystals attract each other via a short-range force that decays exponentially with the lattice constant as the characteristic length scale. Clearly, this short-ranged force is strongest at zero temperature and thermal fluctuations diminish this attraction.

Although the physical origin of the attraction is clear in each approach, the relationship between them remains somewhat obscure, and this has generated a debate in the literature [11]. Therefore, it is desirable to formulate a unified approach that captures the physics of both mechanisms and addresses some important issues, for example, the temperature dependence of the short-ranged force, computed only at zero temperature in Eq. (1). It is the goal of this paper to formulate such an approach. Since at low temperature the counterion distribution is essentially two dimensional, we consider a model system composed of two uniformly charged planes a distance d apart, each having a charge density en. Confined on the surfaces are negative pointlike 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 and develop a detailed physical picture of the electrostatic interaction between them at finite temperatures but below their melting temperature [12].

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 that remain at finite temperature.) By adopting an elasticity theory, the total force of the system can be decomposed (approximately) into a short-ranged component arising from structural correlations and a long-ranged component from correlated fluctuations. They are calculated in Sec. II within the harmonic approximation using Boltzmann statistics (classical), which is valid below the melting temperature of the Wigner crystals. We show that the short-ranged force persists at finite temperature, and we obtain a simple expression—see Eq. (25)—which reduces to the zero-temperature result in Eq. (1) [8,10]. The interesting effect of thermal fluctuations is to reduce the *range* of this force and thus the effect is not negligible even below the melting temperature of the Wigner crystals. For the long-ranged force, this ''elastic'' calculation—see Eq. (21)—finds exactly the same result, even including the prefactor, as the Debye-Hückel (Gaussian) approximation [7]. This is to be expected since the long-wavelength density fluctuations, which give rise to the long-ranged force, are independent of the local Wigner crystal-like ordering. Thus, an important insight gained here is that what is previously thought of as disparate mechanisms for the attractions—the short-ranged attraction (ground state) for low temperature and the long-ranged attraction (charge fluctuation) for high temperature—are both captured within a single framework.

In addition, at zero temperature there must also be a long-ranged attraction derived from the *quantum* fluctuations of the plasmons [10]. This is the low-temperature counterpart of the long-ranged force arising from charge fluctuation at finite temperature. While this low-temperature result only bears a conceptual interest for macroions, it may have real relevance in the larger context of fluctuation-induced interactions in general, and in semiconductor bilayers in particular [13]. Interestingly, although similar to the Casimir effect, arising from zero-point fluctuations at $T=0$, the fluctuation-induced force associated with two-coupled Wigner crystals is fundamentally different. The Casimir effect pertains to two metal slabs separated by a gap of distance d, outside of which there is no electric field; this force scales d^{-4} at $T=0$ [14]. For the case of coupled Wigner crystals, zero-point fluctuations of the plasmons lead to a characteristically different force, which decays with a novel power law: $d^{-7/2}$ [10]. Hence, this long-ranged attractive force dominates the ground-state short-ranged attraction in Eq. (1) for large d. Furthermore, it is of fundamental interest to consider finite-temperature effects as well. This is done in Sec. III, where we first recall the phonon spectrum of the coupled Wigner crystals, identify the plasmon modes, which characterize the density fluctuations of the system, and compute the attractive force arising from fluctuations using explicitly the Bose-Einstein distribution, which appropriately captures quantum effects at very low temperatures and thermal effects at higher temperatures for phonons in general and for plasmons in this particular case. Our result in the classical regime, which scales d^{-3}, agrees exactly including the prefactor with that based on 2D Debye-Hückel theory and ''elasticity'' theory in Sec. II. Thus, we have provided an interesting but different perspective on the same problem and explicitly show how the $d^{-7/2}$ force law at zero temperature crosses over to the d^{-3} law at high temperature via an intermediate d^{-2} regime.

Another point worth mentioning concerns the ordering of 2D solids which exhibit quasi-long-range-order (QLRO) [15]. 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 $n(\mathbf{r}) = \Sigma_{\mathbf{G}} n_{\mathbf{G}}(\mathbf{r}) e^{i\mathbf{G}\cdot\mathbf{r}}$ average (thermally) out to zero for a nonzero reciprocal-lattice vector $\mathbf{G}$, i.e., $\langle n_{\mathbf{G}}(\mathbf{r})\rangle = \langle e^{i\mathbf{G}\cdot\mathbf{u}(\mathbf{r})}\rangle = 0$, where $\mathbf{u}(\mathbf{r})$ are the displacements of the particles from their equilibrium positions, while the correlation function decays algebraically to zero: $\langle n_{\mathbf{G}}(\mathbf{r}) n^*_{\mathbf{G}}(\mathbf{0})\rangle \sim r^{-\eta_{\mathbf{G}}(T)}$ with $\eta_{\mathbf{G}}(T) = k_B T G^2 (3\mu + \lambda)/4\pi\mu(2\mu+\lambda)$, where μ and λ 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 nonzero 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 three-dimensional (3D) solids [16].

However, in real biological systems, the ordering of the counterions may be far from a Wigner crystal. Nevertheless, it is important to understand counterion-mediated attractions between two highly charged surfaces in this Wigner-crystal limit, since it does provide useful *qualitative* insights into the nature of this problem, and moreover, the present formulation may serve as a starting point for a more sophisticated theory which includes the melting of coupled Wigner crystals.

This paper is organized as follows. In Sec. II we derive an effective Hamiltonian that 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 Secs. II A and II B, respectively, and a detailed discussion of our results is presented in Sec. II C. In Sec. III 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 [17]

$$\beta\mathcal{H}_0=\frac{1}{2}\sum_i \int \frac{d^2\mathbf{q}}{(2\pi)^2}\Pi_{\alpha\beta}(\mathbf{q})u_\alpha^{(i)}(\mathbf{q})u_\beta^{(i)}(-\mathbf{q}), \tag{2}$$

where $\beta^{-1}=k_BT$, $\mathbf{u}^{(i)}(\mathbf{q})$ is the Fourier transform of the in-plane displacement field of the charges in the ith layer ($i=A$ or B), $\Pi_{\alpha\beta}(\mathbf{q})=[(2\pi l_B n^2/q)P^L_{\alpha\beta}+\mu P^T_{\alpha\beta}]q^2$ is the dynamical matrix, $\mu\approx 0.245n^{3/2}l_B$ is the shear modulus [18] in units of k_BT, and $P^L_{\alpha\beta}=q_\alpha q_\beta/q^2$ 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,

$$\beta\mathcal{H}_{int}=l_B\int d^2\mathbf{x}d^2\mathbf{x}'\frac{[\rho_A(\mathbf{x})-n][\rho_B(\mathbf{x}')-n]}{\sqrt{(\mathbf{x}-\mathbf{x}')^2+d^2}}, \tag{3}$$

where $\rho_i(\mathbf{x})$ is the number density of charges in the ith 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. [19], 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., $\nabla\cdot\mathbf{u}^{(i)}(\mathbf{x})\ll 1$, but $|\mathbf{u}^A(\mathbf{x})-\mathbf{u}^B(\mathbf{x})|$ need not be small compared to the lattice constant a.

Let us introduce a slowly varying field for each layer,

$$\phi_\alpha^{(i)}(\mathbf{x})=x_\alpha-u_\alpha^{(i)}[\vec{\phi}^{(i)}(\mathbf{x})], \tag{4}$$

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

$$\rho_i(\mathbf{x})=\sum_l \delta^2[\mathbf{R}_l-\vec{\phi}^{(i)}(\mathbf{x})]\det[\partial_\alpha\phi_\beta^{(i)}(\mathbf{x})], \tag{5}$$

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

$$\rho_i(\mathbf{x})-n\cong -n\nabla\cdot\mathbf{u}^{(i)}(\mathbf{x})+\sum_{\mathbf{G}\neq 0} n e^{i\mathbf{G}\cdot[\mathbf{x}+\mathbf{u}^{(i)}(\mathbf{x})]}, \tag{6}$$

where $\mathbf{G}$ 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 (6), $\mathcal{H}_{int}$ may be written as

$$\begin{aligned}\beta\mathcal{H}_{int}=&\int\frac{d^2\mathbf{q}}{(2\pi)^2}\frac{2\pi l_B}{q}e^{-qd}\int d^2\mathbf{x}\int d^2\mathbf{x}'e^{i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}\\&\times\left(n\nabla\cdot\mathbf{u}^A(\mathbf{x})-\sum_{\mathbf{G}\neq 0} n e^{i\mathbf{G}\cdot[\mathbf{x}+\mathbf{u}^A(\mathbf{x})]}\right)\\&\times\left(n\nabla\cdot\mathbf{u}^B(\mathbf{x}')-\sum_{\mathbf{G}'\neq 0} n e^{i\mathbf{G}'\cdot[\mathbf{x}'+\mathbf{c}+\mathbf{u}^B(\mathbf{x}')]}\right),\end{aligned} \tag{7}$$

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

$$\beta\mathcal{H}^L_{int}=\int\frac{d^2\mathbf{q}}{(2\pi)^2}\frac{2\pi l_B n^2}{q}e^{-qd}q_\alpha q_\beta u_\alpha^A(\mathbf{q})u_\beta^B(-\mathbf{q}) \tag{8}$$

and a short-wavelength term

$$\begin{aligned}\beta\mathcal{H}^S_{int}=&+\sum_{\mathbf{G}\neq 0}\sum_{\mathbf{G}'\neq 0}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\frac{2\pi l_B n^2}{q}e^{-qd}\\&\times\int d^2\mathbf{x}\int d^2\mathbf{x}'e^{i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}e^{i\mathbf{G}\cdot[\mathbf{x}+\mathbf{u}^A(\mathbf{x})]}\\&\times e^{i\mathbf{G}'\cdot[\mathbf{x}'+\mathbf{c}+\mathbf{u}^B(\mathbf{x}')]}.\end{aligned} \tag{9}$$

In order to obtain a tractable analytical treatment, we approximate this expression by splitting the sum over $\mathbf{G}'$ into two parts. The dominant part, with $\mathbf{G}'=-\mathbf{G}$ is

$$\beta\mathcal{H}^{S}_{int}=-\sum_{\mathbf{G}\neq 0}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\frac{2\pi l_B n^2}{q}e^{-qd}\times\int d^2\mathbf{x}\int d^2\mathbf{x}'\,e^{i(\mathbf{q}+\mathbf{G})\cdot(\mathbf{x}-\mathbf{x}')}e^{i\mathbf{G}\cdot[\mathbf{u}^A(\mathbf{x})-\mathbf{u}^B(\mathbf{x}')]},\tag{10}$$

where we have used $e^{i\mathbf{G}\cdot\mathbf{c}}=-1$. The second part (those terms with $\mathbf{G}'\neq-\mathbf{G}$) contains extra phase factors that tend to average to zero in the elastic limit. As a first approximation, we neglect such terms. Finally, Eq. (10) can be systematically expanded using a gradient expansion,

$$\beta\mathcal{H}^{S}_{int}=-\sum_{\mathbf{G}\neq 0}\Delta_{\mathbf{G}}(d)\int d^2\mathbf{x}\cos\{\mathbf{G}\cdot[\mathbf{u}^A(\mathbf{x})-\mathbf{u}^B(\mathbf{x})]\}+O(\partial_\alpha u^{(i)}_\beta\partial_\gamma u^{(j)}_\tau),\tag{11}$$

where $\Delta_{\mathbf{G}}(d)=(4\pi l_B n^2/G)e^{-Gd}$. Setting Eqs. (2), (8), and (11) together, we obtain an effective Hamiltonian for the coupled planar Wigner crystals,

$$\beta\mathcal{H}_e=\beta\mathcal{H}_0+\int\frac{d^2\mathbf{q}}{(2\pi)^2}\frac{2\pi l_B n^2}{q}e^{-qd}q_\alpha q_\beta u^A_\alpha(\mathbf{q})u^B_\beta(-\mathbf{q})-\sum_{\mathbf{G}\neq 0}\Delta_{\mathbf{G}}(d)\int d^2\mathbf{x}\cos\{\mathbf{G}\cdot[\mathbf{u}^A(\mathbf{x})-\mathbf{u}^B(\mathbf{x})]\}.\tag{12}$$

The second term in Eq. (12) 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,

$$\begin{aligned}\Pi(d)&=-\frac{1}{A_0}\left\langle\frac{\partial\mathcal{H}_{int}}{\partial d}\right\rangle_{\mathcal{H}_e}\\&=-\frac{1}{A_0}\left\langle\frac{\partial\mathcal{H}^S_{int}}{\partial d}\right\rangle_{\mathcal{H}_e}-\frac{1}{A_0}\left\langle\frac{\partial\mathcal{H}^L_{int}}{\partial d}\right\rangle_{\mathcal{H}_e}\\&=\Pi_{SR}(d)+\Pi_{LR}(d),\end{aligned}\tag{13}$$

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 dominates at small separations.

To calculate various expectation values in Eq. (13), it is convenient to transform the displacement fields into in-phase and out-of-phase displacement fields by $\mathbf{u}^+(\mathbf{x})=\mathbf{u}^A(\mathbf{x})+\mathbf{u}^B(\mathbf{x})$ and $\mathbf{u}^-(\mathbf{x})=\mathbf{u}^A(\mathbf{x})-\mathbf{u}^B(\mathbf{x})$, respectively, so that the effective Hamiltonian (12) separates into two independent parts: $\mathcal{H}_e=\mathcal{H}_++\mathcal{H}_-$ with

$$\beta\mathcal{H}_+=\frac{1}{2}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\Pi^+_{\alpha\beta}(\mathbf{q})u^+_\alpha(\mathbf{q})u^+_\beta(-\mathbf{q}),\tag{14}$$

and

$$\beta\mathcal{H}_-=\frac{1}{2}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\Pi^-_{\alpha\beta}(\mathbf{q})u^-_\alpha(\mathbf{q})u^-_\beta(-\mathbf{q})-\sum_{\mathbf{G}\neq 0}\Delta_{\mathbf{G}}(d)\int d^2\mathbf{x}\cos[\mathbf{G}\cdot\mathbf{u}^-(\mathbf{x})],\tag{15}$$

where $\Pi^{\pm}_{\alpha\beta}(\mathbf{q})=\frac{1}{2}[(2\pi l_B n^2/q)(1\pm e^{-qd})P^L_{\alpha\beta}+\mu P^T_{\alpha\beta}]q^2$. Furthermore, at low temperature, where $|\mathbf{u}^-(\mathbf{x})|$ is small compared to the lattice constant a, the cosine term in Eq. (15) can be expanded up to second order in $|\mathbf{u}^-(\mathbf{x})|$ to obtain the ''mass'' terms. Within a harmonic approximation $\mathcal{H}_-$, up to an additive constant may be written as

$$\beta\mathcal{H}_-\simeq\frac{1}{2}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\Pi^-_{\alpha\beta}(\mathbf{q})u^-_\alpha(\mathbf{q})u^-_\beta(-\mathbf{q})+\frac{1}{2}\int\frac{d^2\mathbf{q}}{(2\pi)^2}[m_L^2P^L_{\alpha\beta}+m_T^2P^T_{\alpha\beta}]u^-_\alpha(\mathbf{q})u^-_\beta(-\mathbf{q}),\tag{16}$$

where $m^2_{L,T}=4\pi l_B n^2\Sigma_{\mathbf{G}\neq 0}Ge^{-Gd}=4\pi l_B n^2\Delta_0(d)$. This approximation is valid below the melting temperature of the Wigner crystals. Note that the mass terms vanish exponentially with d as also found in Ref. [20]. The fact that the transverse m_T and longitudinal ''mass'' m_L are degenerate is related to the underlying triangular structure of the lattices [20]. 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 Secs. II A and II B we derive expressions for the long-ranged and the short-ranged pressure as given in Eq. (13) 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 $\partial_\alpha P^T_{\alpha\beta}u^{(i)}_\beta(\mathbf{x})=0$. Using Eqs. (8) and (13), we obtain an expression for the long-ranged force

$$\beta\Pi_{LR}(d)=\frac{2\pi l_B}{A_0}\int\frac{d^2\mathbf{q}}{(2\pi)^2}e^{-qd}\langle\delta\rho_A(\mathbf{q})\delta\rho_B(-\mathbf{q})\rangle,\tag{17}$$

where $\delta\rho_i(\mathbf{x}) = -n\boldsymbol{\nabla}\cdot\mathbf{u}^{(i)}(\mathbf{x})$ is the long-wavelength density fluctuation to the lowest order. Making use of the equipartition theorem, the correlation function $\langle \delta\rho_A(\mathbf{q})\delta\rho_B(-\mathbf{q})\rangle$ can be evaluated

$$\begin{aligned}\langle \delta\rho_A(\mathbf{q})\delta\rho_B(-\mathbf{q})\rangle &\equiv \mathcal{N}^{-1}\int D\mathbf{u}^{\pm}(\mathbf{q})\,\delta\rho_A(\mathbf{q})\,\delta\rho_B(-\mathbf{q})e^{-\beta\mathcal{H}_e}\\ &= -A_0\frac{q^2}{4\pi l_B}\left[\frac{1}{q(1-e^{-qd})+4\Delta_0}-\frac{1}{q(1+e^{-qd})}\right],\end{aligned} \tag{18}$$

where $\mathcal{N}\equiv\int D\mathbf{u}^{\pm}(\mathbf{q})e^{-\beta\mathcal{H}_e}$ is the normalization factor and $\Delta_0\equiv\Sigma_{\mathbf{G}\neq 0}Ge^{-Gd}$. Substituting this result into Eq. (17), we find

$$\Pi_{LR}(d) = -\frac{k_BT}{d^3}\alpha(\Delta_0 d), \tag{19}$$

where

$$\alpha(x)\cong\frac{\zeta(3)}{8\pi}+\frac{x}{\pi}[\mathrm{Ci}(2\sqrt{x})\cos(2\sqrt{x})+\mathrm{Si}(2\sqrt{x})\sin(2\sqrt{x})], \tag{20}$$

ζ is the Riemann zeta function, and $\mathrm{Ci}(x)$ and $\mathrm{Si}(x)$ are the cosine and sine integral functions [21], respectively. In the large distance limit, the second term in Eq. (20) is exponentially suppressed and can be neglected, yielding $\alpha = \zeta(3)/8\pi$. Therefore, for large d we obtain the long-ranged attraction arising from correlated fluctuations

$$\Pi_{LR}(d) = -\frac{\zeta(3)}{8\pi}\frac{k_BT}{d^3}, \tag{21}$$

which is identical to the well-known result from the Debye-Hückel approximation [7]. Note that this long-ranged force belongs to the general class of fluctuation-induced forces, in which the Casimir effect is a prototype and the amplitude $\zeta(3)/8\pi\cong 0.048$ is universal for this interaction, induced by the long-wavelength fluctuations [9]. Although the scaling of this charge-fluctuation-induced force coincides with that of the finite temperature van der Waals interaction, they are very different at low temperature. This is explored in Sec. III.

B. Short-ranged pressure

The short-ranged force that decays exponentially owes its existence to the ‘‘structural’’ correlations. It survives even at nonzero 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. (10) and (13),

$$\beta\Pi_{SR}(d) = -2\pi l_B n^2\sum_{\mathbf{G}\neq 0} e^{-(G^2/2)\langle|\mathbf{u}^-(0)|^2\rangle}f_{\mathbf{G}}(d), \tag{22}$$

where $f_{\mathbf{G}}(d)=\int d^2\mathbf{q}/(2\pi)^2\mathcal{S}(\mathbf{q}-\mathbf{G})e^{-qd}$, $\mathcal{S}(\mathbf{q}-\mathbf{G}) = \int d^2\mathbf{r}e^{i(\mathbf{q}-\mathbf{G})\cdot\mathbf{r}}e^{-(G^2/8)[B^+(\mathbf{r})-B^-(\mathbf{r})]}$, and $B^{\pm}(\mathbf{r}) = \langle[\mathbf{u}^{\pm}(\mathbf{r})-\mathbf{u}^{\pm}(\mathbf{0})]^2\rangle$. Note that Eq. (22) is exact, provided all the averages are evaluated exactly. For a system of coupled perfect Wigner crystals at zero temperature, $f_{\mathbf{G}}(d)=e^{-Gd}$. At finite temperature, but below the melting temperature T_m, we note that $B^{\pm}(r)$ varies very slowly in space, so that $f_{\mathbf{G}}(d)$ can be approximated by its zero-temperature value: $f_{\mathbf{G}}(d)\simeq e^{-Gd}$. Hence, we obtain

$$\beta\Pi_{SR}(d)\cong -2\pi l_B n^2\sum_{\mathbf{G}\neq 0}e^{-Gd}\langle e^{i\mathbf{G}\cdot[\mathbf{u}^A(\mathbf{0})-\mathbf{u}^B(\mathbf{0})]}\rangle_{\mathcal{H}_e}. \tag{23}$$

The thermal average of the displacement fields in Eq. (23) resembles a ‘‘Debye-Waller’’ factor and indicates 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. (12), 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. (23) only involves $\mathcal{H}_-$. Within the harmonic approximation, the mean-square out-of-phase displacement field can be evaluated

$$\begin{aligned}\langle|\mathbf{u}^-(\mathbf{x})|^2\rangle &= \frac{\lambda_D}{2\pi nd}\ln\left[\frac{d}{4\Delta_0(d)a^2}\right]\\ &\quad+\frac{1}{2\pi\mu}\ln\left[\frac{\mu}{8\pi l_B n^2\Delta_0(d)a^2}\right]\simeq\frac{G_0 d}{2\pi}\left[\frac{\lambda_D}{nd}+\frac{1}{\mu}\right],\end{aligned} \tag{24}$$

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_BT, 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. (24) into Eq. (23), we obtain an expression for the short-ranged pressure at finite temperatures

$$\beta\Pi_{SR}(d)\simeq -2\pi l_B n^2 e^{-(1+\xi/2)G_0 d}. \tag{25}$$

Here, the parameter ξ defined by

$$\xi=\frac{G_0^2}{2\pi}\left(\frac{\lambda_D}{nd}+\frac{1}{\mu}\right) \tag{26}$$

characterizes the relative strengths of thermal fluctuations and the electrostatic energy of a Wigner crystal, i.e., $\xi \sim k_BTa/e^2$. Thus, the sole effect of thermal fluctuations on the short-ranged force is to reduce its *range*: $G_0 \to G_0[1+(\xi/2)]$.

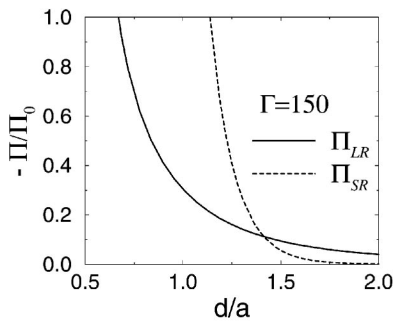

FIG. 2. Plot of Π_{SR} and Π_{LR} versus d for $\Gamma=150$. Observe that the crossover ($\Pi_{LR}\approx\Pi_{SR}$) occurs at about $d\sim a$. $\Pi_0 \equiv k_BT(l_B/a^4)\times 10^{-3}$.

C. Discussion of results

In summary, we have shown that the total pressure between two coupled Wigner crystals can be decomposed into a long-ranged Π_{LR} and a short-ranged pressure Π_{SR}. Each force is computed below their melting temperature, where the harmonic approximation is expected to be valid. The result for the total force is

$$\beta\Pi(d)\simeq -2\pi l_B n^2 e^{-(1+\xi/2)G_0 d}-\frac{\alpha(\Delta_0 d)}{d^3}, \qquad (27)$$

where $\xi=(G_0^2/2\pi)[(\lambda_D/nd)+(1/\mu)]$ and $\alpha(\Delta_0 d)=\zeta(3)/8\pi$ for large d. In Figs. 2 and 3, we have plotted Π_{SR} and Π_{LR} for two values of the coupling constant, $\Gamma\equiv l_B/a=150$ and 50. Note that $\Gamma\equiv e^2\sqrt{\pi n}/(\epsilon k_BT)$ is the ratio of the average Coulomb energy among charges and their thermal energy. Not surprisingly, they show that Π_{SR} dominates for small d, and Π_{LR} for large d. However, it is interesting to observe that even for high values of Γ, Π_{LR} dominates as soon as $d\sim a$. We note that the crossover condition here is not sensitive to the approximation leading to Eq. (25), since all higher-order terms neglected are strongly exponentially suppressed for distances larger than the lattice constant.

According to Eq. (25), the magnitude of Π_{SR} tends to decrease exponentially with temperature. 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. (15), 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 $\langle e^{i\mathbf{G}_0\cdot\mathbf{u}^-(\mathbf{0})}\rangle\sim\Delta_{\mathbf{G}_0}^{\xi/(2-\xi)}$ [22]. Therefore, the short-ranged pressure scales like

$$\begin{aligned}\Pi_{SR}(d)&\sim-\Delta_{\mathbf{G}_0}(d)\times\langle e^{i\mathbf{G}_0\cdot\mathbf{u}^-(\mathbf{0})}\rangle\\ &\sim-\Delta_{\mathbf{G}_0}\times\Delta_{\mathbf{G}_0}^{\xi/(2-\xi)}\\ &\sim -e^{-G_0 d[2/(2-\xi)]}.\end{aligned} \qquad (28)$$

In the low-temperature limit ($\xi\ll1$), we see that the range of Π_{SR} is $G_0(1+\xi/2)$ as obtained in Eq. (25). This scaling argument also suggests that at higher temperatures thermal fluctuations may have interesting nonperturbative effects. At zero temperature $\xi=0$, so Π_{SR} in Eq. (25) reproduces the known result of an exponentially decaying attractive force as obtained in Eq. (1). It should be mentioned that in real biological systems, counterions are likely to be a correlated fluid with short-ranged order. However, as long as $\Gamma\gg1$ and the lateral characteristic correlation length is much larger than the spacing between the layers, it is possible to have structural correlations and our calculation should capture the short-ranged attraction at least qualitatively.

The long-ranged pressure for large d in Eq. (21) 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 [23]. 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 fluctuation.

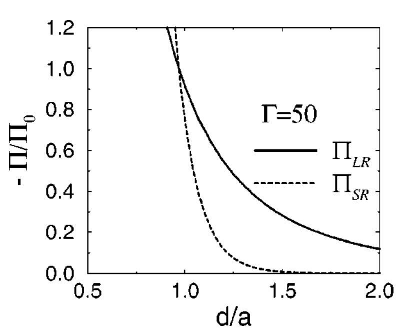

FIG. 3. Same as Fig. 2 for $\Gamma=50$.

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 $T\to0$, 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. [10] (and recall below) 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 $T\to0$ 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 also compute their temperature dependence.

To this end, it is more convenient to employ a method with which fluctuation-induced forces are usually calculated [9]. The advantage of the elastic approach in Sec. II is that the short-ranged force is captured more transparently. However, for the long-ranged force, an equivalent formulation in terms of the plasmon excitations seems more natural in the low-temperature regime where quantum effects are important. Below, we recall the phonon spectrum of the coupled Wigner crystals, identify the plasmon modes, which characterize the density fluctuations of the system, and compute the attractive force arising from fluctuations using explicitly the Bose-Einstein distribution, which appropriately captures quantum effects at very low temperature and thermal effects at higher temperature, for phonons in general and plasmons in this particular case.

Within the harmonic approximation to the effective Hamiltonian, the dynamical matrix can be diagonalized to yield four modes. Two of them are the shear modes of the system that do not contribute to the long-ranged force [10]. The dominate modes that lead to the long-ranged attraction are the two plasmon modes of two-coupled 2D Wigner crytals, which have the following dispersion relations:

$$\omega_1^2(q)=\frac{8\pi e^2 n}{m\epsilon}\Delta_0(d)+\frac{2\pi e^2 n}{m\epsilon}q(1-e^{-qd}), \tag{29}$$

$$\omega_2^2(q)=\frac{2\pi e^2 n}{m\epsilon}q(1+e^{-qd}), \tag{30}$$

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 the Bose-Einstein distribution

$$\mathcal{F}(d)/A_0=\frac{\hbar}{2}\sum_{i=1,2}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\omega_i(\mathbf{q})+k_BT\sum_{i=1,2}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\ln[1-e^{-\beta\hbar\omega_i(\mathbf{q})}], \tag{31}$$

where A_0 is the area of the plane. Since the energy gap Δ_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. (31) arising from the zero-point fluctuations leads to an attractive pressure [10]

$$\Pi_{LR}^0(d)=-\frac{1}{A_0}\frac{\partial\mathcal{F}_0(d)}{\partial d}=-\sqrt{\frac{\hbar^2e^2n}{m\epsilon}}\frac{\alpha_1}{d^{7/2}}, \tag{32}$$

where α_1 is a positive numerical constant of order unity, explicitly given by

$$\alpha_1=\frac{1}{4\sqrt{2\pi}}\int_0^\infty dx x^{5/2}e^{-x}\left\{\frac{1}{\sqrt{1-e^{-x}}}-\frac{1}{\sqrt{1+e^{-x}}}\right\}. \tag{33}$$

Thus, zero-point fluctuations induce a long-range attraction which decays with a power law $\sim d^{-7/2}$. This should be contrasted with the usual Casimir-like force $\sim d^{-4}$, which arises from, for example, the acoustic-phonon zero-point fluctuations. We note that this power law stems from the two-dimensional nature of charged systems: 2D plasmons do not have a finite gap, as they do in 3D. For an order of magnitude estimate, assuming $m\sim10^{-25}$ kg, $n\sim1/50$ Å^{-2}, $d\sim10$ Å, and $\epsilon\sim80$, we find $\Pi_{LR}^0\sim10^{-25}$ J/Å^3. This is close to the magnitude of the short-ranged force in Eq. (1) at zero temperature: $\Pi_{SR}(d)\sim10^{-24}$ J/Å^3, and thus may be just as important under suitable conditions.

An additional contribution to the pressure at finite temperature arises from the second term in Eq. (31),

$$\beta\Pi_{LR}(d)=-\frac{\hbar\Lambda}{4\pi d^{7/2}}\int_0^\infty dx x^{5/2}\times\left\{\frac{1}{\exp[\eta\sqrt{x(1-e^{-x})}]-1}\frac{e^{-x}}{\sqrt{1-e^{-x}}}-\frac{1}{\exp[\eta\sqrt{x(1+e^{-x})}]-1}\frac{e^{-x}}{\sqrt{1+e^{-x}}}\right\}, \tag{34}$$

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

In the limit $\eta\gg1$, Eq. (34) can be systematically expanded in powers of η^{-1}. The lowest-order term is given by $\Pi_{LR}(d)=-\alpha_2(k_BT/\lambda_Ld^2)$, where $\lambda_L\equiv a_B(l_B/2\lambda_D)$, $a_B\equiv\epsilon\hbar^2/(me^2)$ is the effective Bohr radius, $\alpha_2\equiv(1/4\pi)\int_0^\infty dx[x^2/(e^x-1)]=\zeta(3)/(2\pi)$, and ζ is the Riemann zeta function. We observe that the condition $\eta\gg1$ is equivalent to the short distance limit $d\ll\lambda_L$.

In the limit $\eta\ll1$ or the large distance limit $d\gg\lambda_L$, we expand the exponential in the denominator of Eq. (34) to obtain $\Pi_{LR}(d)=-\alpha(k_BT/d^3)$, where $\alpha=\zeta(3)/(8\pi)$. 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 arising finite temperature contributions

$$\Pi_{LR}(d)\simeq\begin{cases}-k_BT/d^3 & \text{for}\quad \lambda_L\ll d\\ -k_BT/(\lambda_Ld^2) & \text{for}\quad \lambda_L\gg d.\end{cases} \tag{35}$$

We note that λ_L, in contrast to λ_D, increases with decreasing temperature, indicating, as one might expect, that quantum fluctuations are important at low temperatures. Furthermore, since $\Pi_{LR}(d)\to0$ as $T\to0$, the attractive interaction as $T\to0$ is governed by zero-point fluctuations as emphasized above. In the strong Coulomb coupling limit $l_B/\lambda_D\sim100$, we get $\lambda_L\sim3$ Å for $\epsilon\sim100$ and $a_B\sim1/20$ Å. It should be emphasized that the results in this section are independent of the nature of the ground state. Thus, any system where low-temperature modes of plasmon are important may, in principle, exhibit the behavior predicted in this section. This means that quantum contributions to the long-

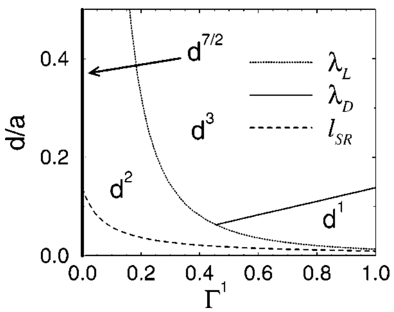

FIG. 4. A schematic phase diagram summarizing different charge-fluctuation-induced attraction regimes. The characteristic decay length l_{SR} of the short-ranged force is also shown.

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 have real impact in a greater field of fluctuation-induced forces in general and for electrons in bilayer semiconductor systems in particular. Indeed, there exist recent theoretical efforts devoted to this subject [13].

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 short-distance result, which scales as d^{-1} for $d<\lambda_D$ 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. (23) does not vanish is because of the cosine term in $\mathcal{H}_-$ given in Eq. (15), 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 Π_{SR} in Eq. (25) is no longer valid, since the harmonic approximation breaks down. Indeed, the scaling argument leading to Eq. (28) suggests that if the full cosine term is retained, Π_{SR} may exhibit nonperturbative behaviors as $\xi\to 2^-$.

To discuss qualitatively what happens at higher temperatures, we assume that $\Delta_{\mathbf{G}}(d)$ 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 $\xi=2$ [24]. 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 Π_{SR} in the unlocked phase, $\mathcal{H}^S_{int}$ given in Eq. (11) can be treated as a perturbation in evaluating the Debye-Waller factor in Eq. (23). To the lowest order, we obtain

$$\Pi_{SR}(d)\simeq -\frac{k_BT}{\lambda_D^2 a}\left(\frac{\xi-1}{\xi-2}\right)e^{-2G_0 d}, \tag{36}$$

where $\lambda_D=1/(2\pi n l_B)$. 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. (25), the range of Π_{SR} remains constant and the amplitude acquires a temperature dependence of $\sim T^{-1}$ (for large $\xi\gg 2$), reminiscent of a high-temperature expansion [25].

However, the above picture may be modified if the charges have melted into a Coulomb fluid via a dislocation-mediated melting transition [23] 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 Π_{SR} crosses over from low-temperature behavior in Eq. (25) to high-temperature behavior in Eq. (36). It may be of interest to note that in Ref. [6], an estimate for the *upper* limit of Γ at which the Poisson-Boltzmann equation breaks down is of the order of $\Gamma\sim 3$. For divalent counterions ''condensed'' onto a highly charged (opposite) plate of surface charge density $\sigma\sim e/10$ Å^{-2}, $\Gamma\sim 20$ at room temperature and the counterions are best described as a 2D correlated Coulomb fluid. However, as long as the characteristic lateral correlation length is much larger than the spacing between the two layers, our elastic approach should capture the qualitative behavior of the short-ranged attraction. A better theory should include the melting of coupled 2D Wigner crystals by introducing excitations of dislocations into the effective Hamiltonian Eq. (12) similar to what is done in Ref. [26]. 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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[12] 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.

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