Ground state of classical bilayer Wigner crystals

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Abstract – We study the ground-state structure of electronic-like bilayers, where different phases compete upon changing the inter-layer separation or particle density. New series representations with exceptional convergence properties are derived for the exact Coulombic energies under scrutiny. The complete phase transition scenario —including critical phenomena— can subsequently be worked out in detail, thereby unifying a rather scattered or contradictory body of literature, hitherto plagued by the inaccuracies inherent to long-range interaction potentials.

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The prediction by Wigner that strongly correlated charge carriers in a uniform compensating background could crystallize [1], was first realized experimentally with electrons at the surface of liquid helium [2], which form a two-dimensional structure. Since then, the study of low-dimensional electronic systems has shown no abating and in particular, the bilayer geometry singles out. It appears significantly richer than its monolayer counterpart and has been investigated in different settings: GaAs quantum wells [3] or other semiconductors [4], quantum dots [5], graphene [6], boron nitride [7], laser-cooled trapped ion plasmas [8], dusty plasmas [9] and colloids [10]. In light of these applications, it is essential to understand the ground-state features of Coulombic bilayers, starting with the classical limit. This problem has received significant attention in the last 20 years, as such [13–17] or supplemented with finite temperature analysis [18–20]. In addition, ground-state ordering impinges on strong-coupling expansions describing counterintuitive yet ubiquitous electrostatic phenomena, such as like-charge attraction [11,21–23] or charge reversal [12,24]. This body of work has revealed the main features of ground-state structure, but there exist surprising discrepancies in the literature, especially on the respective domains of existence of the different phases possible. The reason lies in the long-range nature of Coulombic interactions, a common bane for such analysis. Our goal here is to resolve existing controversies, precisely locate all phases and discuss the critical behavior associated. All results reported are exact; they are obtained from new series representations of lattice sums for Coulomb law.

We consider an ensemble of identical classical point charges \( q \), interacting through a \( 1/r \) pair potential, and confined between two symmetric parallel charged walls. These boundaries both bear a uniform surface charge of density \( -\sigma \), so that global electroneutrality holds. At finite temperature \( T \), the charges do populate the interior of the slab. For \( T = 0 \), the charges evenly condense on the opposing walls, thereby forming a bilayer ground state [25] the structure of which depends on a single dimensionless parameter \( \eta = d\sqrt{\sigma} \), where \( d \) is the interplate distance (see fig. 1). It is known that five structures can be realized upon increasing \( \eta \); they will be referred to following standard terminology, common to the classical [15] and quantum contexts [28]. To begin with, the limits of small and large \( \eta \) are both straightforward. For \( \eta \to 0 \), a genuine two-dimensional one-component plasma is produced [21], where the strong mutual repulsion between charges leads to a hexagonal Wigner crystal [29], the so-called structure I. Conversely, for \( \eta \to \infty \), the two layers decouple and a hexagonal crystal forms on each plate (structure V). These two crystals adopt a stag-

1Note though that in the soft matter realm, the charge carriers need to be multivalent in order to reach, at room temperature and in an aqueous dispersion, the strong couplings required for Wigner crystallization [11,12].

2Clearly, the ground-state structure does not depend on the precise value of the confining plates surface charge, as long as they are symmetric objects, thereby producing a vanishing electric field in the interstitial slab [27].
The inter-layer energy contribution $E_{\text{inter}}$ is amenable to a similar treatment [31], and the last step of the procedure provides us with a simple expression where the limit $R \to \infty$ can be readily taken:

$$
\sqrt{\pi} E_{\text{intra}} \frac{q^2}{2a} \sum_{j,k \neq (0,0)} \frac{1}{\sqrt{j^2 + k^2}} = \frac{q^2}{2a} \int_0^\infty \frac{dr}{\sqrt{r}} \left[ \sum_{j,k} e^{-tr^2} \frac{1}{\sqrt{r}} \right] - 1 - \frac{\pi}{4a}
$$

with the restriction $j^2 + k^2 \Delta^2 \leq (R/a)^2$. It is expedient and common procedure [20,30] to use the gamma identity $(\pi/2)^{1/2} = \int_0^\infty t^{-1/2} \exp(-zt) dt$ valid for $z > 0$, which
consists in introducing the function
\[
  z_\nu(x,y) = \int_0^{1/\nu} \frac{dt}{t^2} e^{-xt} e^{-y/t}, \quad \text{for } y > 0. \tag{4}
\]

We finally end up with the series representation for the total energy \( E(\Delta, \eta) = E_{\text{intra}} + E_{\text{inter}} \):
\[
  \frac{E(\Delta, \eta)}{q^2\sqrt{\sigma}} = 2 \sum_{j=1}^{\infty} \left[ \frac{z_{3/2}(0, j^2/\Delta) + z_{3/2}(0, j^2 \Delta)}{j^2 + \Delta} \right] + 4 \sum_{j,k=1}^{\infty} \frac{(-1)^j z_{3/2}((\pi j)^2, j^2 \Delta)}{j^2 + \Delta} + 2 \sum_{j,k=1}^{\infty} \frac{(-1)^j (-1)^k z_{3/2}((\pi j)^2, j^2 \Delta)}{j^2 + \Delta} + 2 \sum_{j,k=1}^{\infty} \frac{z_{3/2}(0, \eta^2 + (j - 1/2)^2/\Delta + (k - 1/2)^2 \Delta)}{j^2 + \Delta} - 2\sqrt{\frac{\pi}{2}} z_{1/2}(0, \eta^2).
\tag{5}
\]

The function \( z_\nu \) generalizes to two-layer problems the so-called Misra function [32] used extensively in single-layer lattice summation [33,34]. Our use of (5) will be three-pronged: it allows us to show analytically that \( \eta = 0 \), to calculate explicitly the singular behavior near critical points and is moreover particularly suited for numerical evaluations. From an operational point of view, the series (5) is indeed endowed with remarkable properties: it is free of singular terms, and importantly, converges extremely quickly. The error made upon truncating the series in the energy expression (5) at order \( j = k = M \) behaves like \( \exp(-cM^2)/M \), where \( c \) is a constant of order unity. We first document the convergence property on the single-layer case of structure I, for which the exact energy is \( E(\sqrt{\Delta}, 0)/q^2\sqrt{2\sigma} = -1.96051578931989006 \ldots \), which is directly the Madelung constant of the 2D hexagonal Wigner crystal. Cutting the series (5) at \( M \), we obtain the exact value with a precision of 2,5,10,17 digits with notably small cutoffs \( M = 1, 2, 3, 4 \), respectively. This makes numerical calculations extremely fast and efficient on any workstation. A similar accuracy is met for all other structures and parameter values reported here, and all numerical results quoted below have been obtained with the cutoff \( M = 5 \).

We now turn our attention to the threshold \( \eta_0 \) which defines the stability window of structure I. For a given distance \( \eta \), we proceed by calculating the Taylor expansion of (5) in the small parameter \( \epsilon = \sqrt{3} - \Delta \), which yields
\[
  \frac{E(\sqrt{3} - \epsilon, \eta)}{q^2\sqrt{2\sigma}} = \frac{E(\sqrt{3}, \eta)}{q^2\sqrt{2\sigma}} + f_1(\eta) \epsilon + f_2(\eta) \epsilon^2 + O(\epsilon^3),
\]
where
\[
  f_1(\eta) = \frac{1}{2^{1/2}\sqrt{\pi}} \left\{ 4 \sum_{j=1}^{\infty} j^2 \left[ z_{5/2}(0, j^2/\sqrt{3}) - \frac{1}{3} z_{5/2}(0, j^2/\sqrt{3}) \right] + 8 \sum_{j,k=1}^{\infty} \left( k^2 - \frac{j^3}{3} \right) z_{5/2}(0, j^2/\sqrt{3} + k^2/\sqrt{3}) + 2 \sum_{j=1}^{\infty} (-1)^j j^2 \left[ z_{5/2}(\pi j)^2, j^2/\sqrt{3} - \frac{1}{3} z_{5/2}(\pi j)^2, j^2/\sqrt{3} \right] + 4 \sum_{j,k=1}^{\infty} (-1)^k \left[ k^2 - \frac{j^2}{3} \right] z_{5/2}(\pi j)^2, j^2/\sqrt{3} + k^2/\sqrt{3}) + 4 \sum_{j,k=1}^{\infty} \left[ (k - 1/2)^2 - \frac{1}{3} \left( j - 1/2 \right)^2 \right] \right\} \times z_{5/2}(0, \eta^2 + (j - 1/2)^2/\sqrt{3} + (k - 1/2)^2/\sqrt{3}) \right\},
\tag{7}
\]
and the function \( f_2 \) is also explicitly known [31]. To investigate the stability of structure I, it is sufficient to study the sign of \( f_1 \), which is worked out from a Taylor expansion for small \( \eta \). The first two derivatives of this function \( f_1 \) vanish at \( \eta = 0 \) and we have \( f_1(\eta) = -0.5830359875 \ldots \eta^2 + \mathcal{O}(\eta^4) \), hence an energy decrease upon increasing \( \epsilon \) as compared to the \( \epsilon = 0 \) case (structure I). This implies that \( \eta = 0 \): at finite but small distances \( \eta \), the optimal phase is not structure I. To obtain the optimal value of \( \epsilon \) selected and that we denote \( \epsilon^* \), we further Taylor expand \( f_2(\eta) \) which yields \( f_2(\eta) = 0.0408440789 \ldots + \mathcal{O}(\eta^3) \). As a consequence,
\[
  \sqrt{3} - \Delta^* \equiv \epsilon^* = -\frac{f_1(\eta)}{2f_2(\eta)} = 7.14064 \ldots \eta^2 + \mathcal{O}(\eta^4),
\tag{8}
\]
which entails that the energy change scales like \( \eta^4 \). For the thresholds \( \eta_1 \) reported in refs. [15,19], a relative accuracy of \( 10^{-9} \) was therefore required to answer the finite or vanishing \( \eta_1 \) question. The accuracy of our findings is illustrated in fig. 3.

The above analysis shows that the evolution from structure I to structure II is not a phase transition in the common sense. The situation differs between structures II and III. To inspect the corresponding transition, we note that \( E(\Delta, \eta) \) enjoys the symmetry \( \Delta \rightarrow -\Delta^{-1} \), as is clear from fig. 2 where a global rotation of \( \pi/2 \) does not affect the energy but interchanges lattice vectors \( a_1 \) and \( a_2 \). The value \( \Delta = 1 \) characterizing structure III is therefore a self-dual point, and it will now be convenient to parameterize the aspect ratio as \( \Delta = \exp(\epsilon) \). All expressions will then be even in \( \epsilon \). The expansion of \( E(\epsilon^*, \eta) \) in small \( \epsilon \)-deviations yields
\[
  \frac{E(\epsilon^*, \eta)}{q^2\sqrt{2\sigma}} = \frac{E(1, \eta)}{q^2\sqrt{2\sigma}} + g_2(\eta) \epsilon^2 + g_4(\eta) \epsilon^4 + \mathcal{O}(\epsilon^6),
\tag{9}
\]
where \( g_2 \) and \( g_4 \) are explicitly known [31], in a series form that is very reminiscent of eq. (7). The bilayer energy
Proceeding along similar lines as for the I → II Taylor expansion was accurate: 0. The critical point field arguments usually underlying such approaches, our it should be emphasized that at variance with mean-

appears in a standard Ginzburg-Landau form [35], but it should be emphasized that at variance with mean-
field arguments usually underlying such approaches, our expression is exact. The critical point ηII sought for is the root of g2(η) = 0, which gives ηII = 0.2627602682 … It appears here that the thresholds reported in earlier works were accurate: 0.27 [13], 0.262 [15], 0.27 [17] and 0.28 [19]. Proceeding along similar lines as for the I → II crossover, we Taylor expand g2(η) and g4(η) to leading order around ηII. The former behaves like (η − ηII) while the latter is constant, a prototypical scenario for a continuous phase transition with critical index β = 1/2 [35]. Specifically, we get

$$\Delta^* - 1 \simeq \epsilon^* = \left( -\frac{g_2(\eta)}{2g_4(\eta)} \right)^{1/2} \simeq 1.48031\sqrt{\etaII - \eta}. \quad (10)$$

This expression applies for η < ηII, in the stability domain of our structure II, and is in excellent agreement with our numerical calculations from eq. (5), see fig. 4.

The task remaining is to find the series representations for structures IV and V. We first address structure IV. Implementation of the procedure that led to the series (5) becomes possible once the distance between a reference ion and an arbitrary ion located on the same layer at r(j, k) = ja1 + ka2 is expressed as

$$|r(j, k)|^2 = a^2 \left( j^2 + k^2 + 2jk \cos \varphi \right) = a^2 \left[ (j + k)^2 \cos^2(\varphi/2) + (j - k)^2 \sin^2(\varphi/2) \right]. \quad (11)$$

The latter “diagonalized” form in terms of indices, provides the starting point to write the intra-layer Coulomb energy (summing 1/√(|r(j, k)|^2), and suggests to introduce new indices n and m: if j + k is even, we define n = (j + k)/2 and m = (j − k)/2. If j + k is odd, we introduce indices n = (j + k + 1)/2 and m = (j − k + 1)/2.

Like wise for the inter-layer interactions, taking due account of the shift (a1 + a2)/2 between opposite layers. Building on the gamma identity and Poisson summation formula, the series form for the energy $E_{IV}$ ensues [31]. This energy depends on the angle $\varphi$ and of course on the distance $\eta$. For our purposes, rather than the lengthy explicit form, it is sufficient to report the Landau-like expansion of $E_{IV}$ in the vicinity of $\varphi = \pi/2$. A convenient expansion parameter is $\epsilon$ such that $\exp(\epsilon) = \tan(\varphi/2)$, and the invariance $\varphi \to \pi - \varphi$ makes $E_{IV}$ an even function of $\epsilon$. In the small $\epsilon$ region of interest associated to the vicinity of $\pi/2$ for $\varphi$, we obtain an expansion up to order $\epsilon^2$ of the same form as (9). This teaches us that structure III is unstable for $\eta > \eta_{III} = 0.6214809246 …$, to be compared to the thresholds 0.61 [14], 0.622 [15], 0.62 [17], 0.59 [19] while structure IV was not considered in [13]. We furthermore again obtain a second order phase transition with critical index 1/2 and explicit order parameter close to the transition point

$$\epsilon^* \simeq \frac{\pi}{2} - \varphi^* \simeq 1.24494\sqrt{\eta - \eta_{III}}, \quad (12)$$

in excellent agreement with our numerical data.

The transition IV → V is discontinuous, which made its characterization more elusive in previous publications. Our method, though, is easily adapted to the geometry of structure V. The series representation for $E_{V}(\eta)$ should be compared to $E_{IV}(\varphi^*, \eta)$ evaluated for the optimal distortion angle $\varphi^*(\eta)$. Requiring that $E_{V}(\eta) = E_{IV}(\varphi^*, \eta)$, we obtain the last $\eta$-threshold that was remaining to be specified: $\eta_{IV} = 0.73242 …$. Previous investigations gave 0.75 [14], 0.732 [15], 0.87 [17] and 0.70 [19]. For $\eta > \eta_{IV}$, structure V is energetically favorable. As a by-product of our analysis, we report the large distance behavior of the inter-plate pressure $P = -2\sigma \partial E_{V}/\partial d$:

$$P = -2\pi(\sigma q)^2 \exp \left( -\frac{4\pi}{\sqrt{2}\eta^{1/4}} \right), \quad (13)$$

in agreement with [15] but at variance with [14,22].
To summarize, we have derived series representations for the different Coulomb lattice sums pertaining to the ground state of classical bilayer systems. The derivation, worked out explicitly for the five different structures that were known to compete at vanishing temperature, results from a series of transformation rooted in the general theory of Jacobi $\theta$ functions [31]. The resulting series provide the thresholds delimiting the domains of validity of the different phases, that were prone to some fluctuations in previous works. Figure 5 provides an overview of our main findings. We could in particular show that the simple hexagonal structure I can only exist in the limiting case of a vanishing inter-plate distance, and is preempted by a buckled phase for all $\eta \neq 0$. This is the scenario first reported in ref. [16], which differs from several other studies that assigned a finite stability window to phase I. Whereas the evolution I $\rightarrow$ II is not a phase transition, we could show that the continuous transitions II $\rightarrow$ III and III $\rightarrow$ IV have critical index $\beta = 1/2$. In addition, our series representation is endowed with exceptional convergence properties, providing typically more than 10 digits of accuracy when retaining only the first 4 or 5 terms involved. Relinquishing the symmetry between the two plates to address the cases where they bear different surface charges is an interesting venue for future work. This brings the difficulty that local electroneutrality no longer holds at the single-plate level in the ground state [26], except presumably at large separations. Our approach can also be extended to bilayers and multilayers with repulsive Yukawa or inverse-power-law interactions, that deserve attention.

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