Lekner summations and Ewald summations for quasi-two dimensional systems.

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Abstract

Using the specific model of a bilayer of classical charged particles (bilayer Wigner crystal), we compare the predictions for energies and pair distribution functions obtained by Monte Carlo simulations using three different methods available to treat the long range Coulomb interactions in systems periodic in two directions but bound in the third one. The three methods compared are: the Ewald method for quasi-two dimensional systems [D.E. Parry, Surf. Sci. 49, 433 (1975); ibid., 54, 195 (1976)], the Hautman-Klein method [J. Hautman and M.L. Klein, Mol. Phys. 75, 379 (1992)] and the Lekner summations method [J. Lekner, Physica A176, 485 (1991)]. All of the three method studied in this paper may be applied to any quasi-two dimensional systems, including those having not the specific symmetry of slab systems. For the particular system used in this work, the Ewald method for quasi-two dimensional systems is exact and may be implemented with efficiency; results obtained with the other two methods are systematically compared to results found with the Ewald method. General recommendations to implement with accuracy, but not always with efficiency, the Lekner summations technique in Monte Carlo algorithms are given.

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I. Introduction.

In Chemical physics, Condensed matter physics and Molecular physics, numerical simulations are basic theoretical tools to study generic model systems and to predict qualitatively and/or quantitatively the properties of some materials. Molecular simulations allow to study the thermodynamical and the transport properties of some more or less detailed models of real substances. In Molecular simulations, there are mainly two procedures used: the Molecular Dynamics method and Monte Carlo algorithms, both being generally considered as complementary [1, 2].

The number of particles or the number of degree of freedom of a system generally considered in these studies is ranging from few hundreds to few millions. Compared to macroscopic materials, this number is small and far from the thermodynamical limit. To avoid irrelevant surface or finite-size effects, periodic boundary conditions are generally used.

When the interaction is short ranged, truncations of potentials or minimum image conventions are used to compute with accuracy the interaction energy or forces between molecules or particles.

On the other hand, when the interaction is long-ranged, as it is the case for Coulombic and Dipolar interactions, truncation of the potential may lead to severe bias [3]. For bulk-like systems with Coulombic or Dipolar interactions, Ewald methods are employed to take into account the long-ranged contribution of the potential to energy or forces when periodic boundary conditions are applied to the system [4-6].

In the context of molecular simulations, the interesting feature of Ewald summations method for bulk-like systems is that the cost in computing time for the calculation of the energy of a configuration may scales as $N^{3/2}$, where $N$ is the number of particles. For large systems, this computational time is still too expensive and some numerical procedures based on the Fast Fourier Transform algorithm like the particle-particle/particle-mesh (PPPM) method [7], or procedures based on multipole expansion like the fast multipole methods [8] have been implemented. For bulk-like systems, all these methods have been used and conveniently intercompared in the last two decades.

Some very interesting systems may not be considered as bulk-like systems, for instance when one or two dimensions of the system are small compared to the other dimensions: the quasi-one dimensional or quasi-two dimensional systems. Examples of such systems are fluid-fluid or fluid-solid interfaces, monolayers, biological membranes, free standing films, cylindrical pores, carbon nanotubes, etc. As for bulk-like systems, to avoid some irrelevant finite size effects, other than the finite extension of these systems, partially periodic boundary conditions are employed. Partially periodic boundary conditions consist of taking periodic boundary conditions for only one or two dimensions, while the remaining dimensions are considered with their finite extension. For these systems, straightforward applications of the Ewald summations lead to a computational effort that scales as $N^2$ and not as $N^{3/2}$, because the analytical formulation of the electrostatic energy of this kind of systems is more complicated than for bulk-like systems [9-15]. To compute
coulombic or dipolar energies in these quasi-two (or quasi-one) dimensional systems, several others methods have been proposed [9,16-23]. One has to note that some of these methods may be applied only to systems having a slab-like geometry, while the methods compared in this paper may be applied to any quasi-two dimensional systems.

The purposes of the present work is to exhibit a comparison between three different methods proposed to study quasi-two dimensional systems. The method compared in this work are: (a) the Ewald method for quasi-two dimensional systems (Ewald quasi-2D) [10,11,13-15], (b) the Hautman-Klein method [16] and (c) the Lekner summations technique [19]. All the three methods are applied to a very simple system: a bilayer Wigner crystal, described in the section II of this paper. As it is explained further in the paper, we have chosen to apply these methods to this particular system because for this system the Ewald quasi-2D method may be implemented with the same efficiency as the Ewald method for bulk-like systems. To make consistent intercomparisons between methods, one needs to have some reliable and solid reference points, the Ewald quasi-2D method is the best candidate to produce such reference points. All the results obtained by applying the Hautman-Klein method and the Lekner summations technique to this system are systematically compared to the results obtained by applying the Ewald quasi-2D method.

This work follows a previous study on the convergence of Lekner summations done by the author in ref.[24] and another study done in ref.[25] where we have compared the methods of references [16,18,20] on a system having a slab geometry. One of the aims of the present work is also to present explicitly a procedure to implement with accuracy (but not always with efficiency) the Lekner summations technique in Monte-Carlo simulations using the Metropolis algorithms.

Comparisons of Ewald summation techniques for confined and quasi-bidimensional systems have been done previously, such as in the interesting work by A.H. Widmann and D.B. Adolf [26]. In these studies, authors generally compare numerical accuracy and efficiency of methods on only few configurations of a simple system; for instance in the study of ref.[26], Widmann and Adolf compared Hautman-Klein, Nijboer-de Wette and Parry methods on five configurations of a system with 100 particles. In the present work, the Ewald quasi-2D, Hautmann-Klein and Lekner methods are implemented in Monte Carlo simulations; the influence of the accuracy on thermodynamical averages and on correlation functions is outlined. The purpose of the present paper is to concretely illustrate, on a very simple system, how some methods, and especially the Lekner method, may introduce biais on physical quantities in a Monte Carlo simulation, since relations between thermodynamical averages and accuracy of methods are needed to validate outputs of numerical simulations.

In section II, we describe the model on which comparisons between the different methods are made. In section III, we present the three methods that are studied and compared in this paper. Section IV is devoted to exhibit the results obtained with Monte Carlo simulations using the Metropolis algorithm. A general discussion and some recommendations to implement correctly and with accuracy the Lekner summations are given in section V.
For completeness, an appendix that gives the computation of the interaction between two charged surfaces and ions has been added at the end of the paper.

II. Description of the Model and simulations parameters.

A bilayer of charged particles is the reference system chosen in this work. The three methods used to compute the electrostatic energy of this system are presented in the next section.

This model is frequently used to give crude representations of very different systems. For instance such bilayer Wigner crystals are used to give representation of strongly coupled electronic bilayers of charged particles in two dimensional semiconductor heterostructures or in dusty plasmas. Some recent experiments on Laser-cooled $^9\text{Be}^+$ ions have provided direct observations of structural phase transition in these systems [27].

This very simple model is also used to give very crude representation of neutral lipid bilayers [28] or as basic theoretical model to facilitate the conceptual understanding of counterion-mediated attractions between similarly charged planes [29]. The monolayer version of this model is also useful to describe classical electrons trapped on the surface of liquid helium [30].

Our reference system is a bilayer made of $N = 2N_0$ point ions interacting by a Coulomb potential $1/r$. The ions are evenly distributed in two layers $L_1$ and $L_2$ separated by a distance $h$. A snapshot of an instantaneous configuration of a system of 512 point ions is represented on Figure 1. Each layer is a square of side $L$ and partial periodic boundary conditions, with a spatial periodicity $L$, are applied in both directions parallel to the layer (directions $x$ and $y$) while no periodic boundary conditions are taken in the third direction (direction $z$). The purpose of this work is not to study the phase diagram of this system (which may be very rich, see for instance ref.[31]), thus in all computations that we have done, the shape and area of the layers are constants; nevertheless most of the computations done in this paper can be compared to a more extensive thermodynamical study of this system done in ref.[32]. The charge of points ions is $q$ and to guaranteed charge neutrality, both layers $L_1$ and $L_2$ carry a uniform surface charge density $\sigma$ given by

$$Nq + 2\sigma L^2 = 0$$

In all computations done, we have taken $q = 14$. The characteristic length $a$, the ion-disk radius, is defined by $\pi\rho_0a^2 = 1$ where the ion density in each layer is $\rho_0 = N_0/L^2$. This ion-disk picture is a simplified version of the hexagonal Wigner-Seitz cell.

We have made computations using the three different methods described in the next section in six different situations resumed in TABLE I. For Runs a, c, d, e, e' and f, the coupling constant is $\Gamma = q^2/kTa = 196$ ($a = 1$), while for Runs b, it is $\Gamma = 98$ ($a = 2$).
For all computations, a Monte Carlo cycle (MC-cycle) is made of a random trial move of each particle in both layers, the trial moves are accepted or rejected according to the Metropolis algorithm applied to the NVT ensemble [33, 1, 2]; in a MC-cycle there are \( N \) trial moves. The amplitude of the trial displacement has been chosen such as the acceptance ratios ranged between 30\% and 60\%. No exchange of ions between layers \( L_1 \) and \( L_2 \) is allowed. From an initial configuration, \( t_{eq} \) MC-cycles are used for equilibration and after \( t_{av} \) MC-cycles are used to accumulate the thermodynamical averages. The differences between Runs e and e’ are in the values of \( t_{eq} \) and \( t_{av} \); Runs e’ were applied only to computations using the Ewald quasi-2D and Hautman-Klein methods.

In Runs a, c, d, e, e’ and f, the thermodynamical stable state is a square or hexagonal solid; for these computations, the average electrostatic energies per particle are closed to the corresponding Mandelung energies. The Mandelung energy, \( \beta u_0 \), for a square bidimensional lattice may be found in a computation done by Totsuji [5]. Taking the lattice length as \( L/\sqrt{N_0} \), the Mandelung energies computed in the work of Totsuji are given in TABLE I for all Runs done in the present work. Since the shape and the area of the bilayer are kept constant, this would impose preferentially a square lattice for the solid-like phase, even if the thermodynamical stable phase has another symmetry. To study rigourously the solid phase diagram of this bilayer system, one should allow at least the shape of the box to fluctuate. This study was done in ref.[32]. In the present work, the shape, a square of side \( L \), and the area \( L^2 \) of layers are constant and the average energy per particle found in Monte Carlo computations, except for Runs b, is closed to \( u_0 \). We also give in TABLE I, the preferential structures found in the computations done in ref.[32] for the bilayer Wigner crystal.

The order parameters \( \Psi_m \) used in ref.[32] to differentiate the square and hexagonal structures are not computed in the present study. In the spirit of the current work, these parameters are irrelevant and since the fixed square shape of layers would induced preferentially a square lattice for the solid-like phases, the values for \( \Psi_4 \) and \( \Psi_6 \) that would have been found might be misleading to further studies of the solid phase diagram of this bilayer system.

The energy of the bilayer system can be written as

\[
U = E_{intra} + E_{inter}
\]

where \( E_{intra} \) is the intralayer contribution and \( E_{inter} \) is the interlayer contribution, including the contribution of the surface-surface energy (see appendix). In the next section, we give the analytical expressions for \( E_{intra} \) and \( E_{inter} \) for each of the methods studied in this work.

The intralayer \( g_{11} \) and interlayer \( g_{12} \) pair distribution functions, corresponding to particles in the same layer and to particles belonging to different layers but positions projected onto the same plane respectively, are evaluated. These distribution functions
are computed as

\[
\begin{align*}
    g_{11}(s) &= \frac{L^2}{2N_0^2} < \sum_{i \in L_1} \sum_{j \in L_1, j \neq i} \delta(s - s_{ij}) > \\
    g_{12}(s) &= \frac{L^2}{N_0^2} < \sum_{i \in L_1} \sum_{j \in \bar{L}_2, j \neq i} \delta(s - s_{ij}) > 
\end{align*}
\]

(3)

where particles positions are noted by \( r_i = s_i + z_i e_z \) and \( < ... > \) denotes statistical averages.

In the next section, we describe the different methods used to compute the electrostatic energy, while in section IV we present the results obtained by applying these methods to the bilayer system.

III. Numerical methods to compute the electrostatic energy.

An efficient technique for computing the long ranged part of the intermolecular interactions for systems with periodic boundary conditions is provided by Ewald sums [1, 2, 4]. In this procedure, the interaction energy between molecules is separated into short ranged and long ranged contributions by using a damping function \( f(r; \alpha) \), where \( \alpha \) is a convergence parameter conveniently chosen. For the coulombic interaction, this separation is done by setting

\[
\frac{1}{r} = \frac{1 - f(r; \alpha)}{r} + \frac{f(r; \alpha)}{r}
\]

(4)

The short range contributions to the energy are given by the first term of the right handed side (r.h.s) of Eq.(1) and the long ranged contributions are given by the second term. To handle the long ranged contributions, Fourier transform of the second term is taken; the interaction energy can be set in the form

\[
E = E_r^{(s)} + E_k^{(l)}
\]

(5)

where \( E_r^{(s)} \) is the short ranged contribution and \( E_k^{(l)} \) the long ranged contribution, which for technical convenience, is expressed as

\[
E_k^{(l)} = E_{k=0}^{(l)} + E_{k \neq 0}^{(l)}
\]

(6)

The damping function \( f(r; \alpha) \) may be chosen such as both contributions of Eq.(5) are rapidly convergent. Such choices allow to derive efficient algorithm to compute the interaction energy.

The choice
\[ f(r; \alpha) = \text{erf}(\alpha r) = \frac{2}{\sqrt{\pi}} \int_0^{\alpha r} dt \, \exp(-t^2) \]

is frequently used. Other derivations using an integral representation of the Gamma function give the same damping function [13, 15]. We found for the Coulomb interaction energy of \( N \) particles in a simulation box of side \( L \) with periodic boundary conditions in the three directions,

\[
\begin{align*}
E^{(s)}_r &= \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} \sum_{n} q_i q_j \exp \left(\text{erfc}(\alpha |r_{ij} + L_n|) \right) \\
E^{(l)}_{k=0} &= \frac{2\pi}{3V} \left( \sum_{i=1}^{N} q_i r_i \right)^2 - \frac{\alpha}{\sqrt{\pi}} \sum_{i=1}^{N} q_i^2 \\
E^{(l)}_{k \neq 0} &= \frac{2\pi}{V} \sum_{k \neq 0} \frac{\exp(-k^2/4\alpha^2)}{k^2} \left| \sum_{j=1}^{N} q_j \exp(i k \cdot r_j) \right|^2
\end{align*}
\]

with

\[ L_n = n_x L_x \hat{e}_x + n_y L_y \hat{e}_y + n_z L_z \hat{e}_z \]

where \( n_x, n_y \) and \( n_z \) are relatives integers.

The primed sum in \( E^{(s)}_r \) indicates that for \( L_n = 0, i = j \) must be omitted. In Eq.(7), \( r_i = (x_i, y_i, z_i) \) are positions of particles, \( r_{ij} = r_i - r_j \) and \( k \) the lattice vectors in the tridimensional Fourier space.

In Eqs.(7), the long ranged contributions \( E^{(l)}_{k \neq 0} \) are expressed with a summation over the particles, \( N \) contributions to the sum, and not as a summation over the pair of particles, \( N(N-1)/2 \) contributions; this factorization is one of the technical features that allows to implement efficiently the Ewald-3D method. By reference to the crystalographic origin of the Ewald summations, \( \tilde{\rho}(k) = \sum_j q_j \exp(i k \cdot r_j) \) is sometimes called the structure factor (of the simulation box).

The same procedure, with the same damping function, has been applied to systems in
two dimensions \([5, 6, 34]\). In particular for Coulomb interaction, it gives

\[
\begin{align*}
E_s^{(s)} &= \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} \sum_{\nu} q_i q_j \frac{\text{erfc}(\alpha|s_{ij} + L\nu|)}{|s_{ij} + L\nu|} \\
E_s^{(l)}_{\kappa=0} &= -\frac{\sqrt{\pi}}{\alpha A} \left( \sum_{i=1}^{N} q_i \right)^2 - \frac{\alpha}{\sqrt{\pi}} \sum_{i=1}^{N} q_i^2 \\
E_s^{(l)}_{\kappa \neq 0} &= -\frac{\pi}{2A} \sum_{\kappa \neq 0} \sum_{i=1}^{N} \sum_{j=1}^{N} q_i q_j \frac{\text{erfc}(\kappa/2\alpha)}{\kappa} \exp(i\kappa \cdot s_{ij})
\end{align*}
\]

(8)

with

\[
L\nu = \nu_x L_x \hat{e}_x + \nu_y L_y \hat{e}_y
\]

where \(\nu_x\) and \(\nu_y\) are relative integers. \(\kappa\) are the lattice vectors in the bidimensional Fourier space, \(s_i = (x_i, y_i)\) the position of the particles in the two dimensional space and \(s_{ij} = s_i - s_j\). The convergence parameter \(\alpha\) is usually chosen such as the summations that give \(E_s^{(s)}\) can be restricted to \(\nu = 0\). This choice is often taken as \(\alpha L \simeq 5 - 8\).

For systems with a quasi-two dimensional geometry, as the bilayer Wigner crystal described in section II, the Ewald method for three dimensional systems, given by Eqs.(7), may be applied by taking a highly asymmetric box \([35, 20, 21(b)]\). For systems with a slab geometry, this procedure is certainly the most efficient \([21(b), 24]\). A careful examination of the summations may show that large errors, arising by a naive implementation of the tridimensional Ewald method can be corrected analytically by adding some corrections terms \([20, 21(b)]\). Nevertheless, these procedures may not easily be applied to systems that do not have the specific symmetry of slab-like systems, as it is the case for fluid-fluid, fluid-solid interfaces, monolayers, etc. For these reasons, several methods have been developed to take into account the finite extension of systems without any periodicity in the direction of the finite extension.

The method for real two dimensional systems, given by Eqs.(8), may of course not be applied to quasi-two dimensional systems, except as a limiting case when the length of the finite extension in the third direction tends to zero \((h \rightarrow 0)\).

### A. The Ewald method for quasi-two dimensional systems.

Following analytical computations done in refs.[10-15], one may derived an Ewald method for three dimensional systems with periodicity in only two directions.

One of the most simple derivation of the Ewald quasi-2D method is the analytical derivation done by Parry \([10]\) in 1975; this derivation of the Ewald quasi-2D method is also
very useful to link this method to the Ewald method for tridimensional systems. In the derivation done by Parry, the Ewald-3D method is taken as a starting point. By letting the periodicity of the periodic boundary condition in the direction where the system has a finite extension tending to infinity, an analytical formulation of the Ewald quasi-2D method is found. Other analytical derivations, that do not use the Ewald-3D method as a starting point, may also be found in refs.[11,13,15,35]. In particular, an elegant method may be found in a recent work by Grzybowski, Gwóźdź, and Bródka [15]; their derivation is also closely related to the rapidly convergent representations for Green’s functions for Laplace’s equations presented in a work by Linton [37].

We define our notations as following,

\[
\begin{align*}
    r_{ij} &= s_{ij} + z_{ij}\hat{e}_z \\
    L_\nu &= \nu_x L_x \hat{e}_x + \nu_y L_y \hat{e}_y \\
    L_n &= L_\nu + n_z L_z \hat{e}_z \\
    k &= \kappa + k_z \hat{e}_z
\end{align*}
\]

where \( k_z = 2\pi/L_z \).

One finds by taking \( L_z \to \infty \)

\[
E^{2D} = E_R + E_{\kappa=0} + E_{\kappa\neq0} = E_{pp}^{(cv)}
\]

with the contributions given by [10]

\[
E_R = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} \sum_{\nu} q_i q_j \frac{\text{erfc}(\alpha|\mathbf{r}_{ij} + L_\nu||)}{|\mathbf{r}_{ij} + L_\nu|}
\]

\[
E_{\kappa=0} = -\frac{\pi}{A} \sum_{i=1}^{N} \sum_{j=1}^{N} q_i q_j \left( \frac{\exp(-\alpha^2 z_{ij}^2)}{\alpha\sqrt{\pi}} + |z_{ij}|\text{erf}(\alpha|z_{ij}|) \right) - \frac{\alpha}{\sqrt{\pi}} \sum_{i=1}^{N} q_i^2
\]

\[
E_{\kappa\neq0} = \frac{\pi}{2A} \sum_{\kappa\neq0} \sum_{i=1}^{N} \sum_{j=1}^{N} q_i q_j F(\kappa, \alpha; z_{ij}) \frac{\exp(i\kappa . s_{ij})}{\kappa}
\]

where

\[
F(\kappa, \alpha; z_{ij}) = \exp(\kappa z_{ij})\text{erfc}(\frac{\kappa}{2\alpha} + \alpha z_{ij}) + \exp(-\kappa z_{ij})\text{erfc}(\frac{\kappa}{2\alpha} - \alpha z_{ij})
\]

and the contribution \( \kappa = 0 \) arising from \( k_z \neq 0 \).

When applied to the bilayer Wigner crystal described in section II, one may use the factorization as one particle summation in the long ranged contributions given by Eq.(10.c).
Thus, one may implement with the same efficiency the Ewald quasi-2D method for this system as it is done in the Ewald-3D method. This situation is very particular to the system considered in the present work. For other quasi-2D systems, because the complicated form of the function $F(\kappa, \alpha; z_{ij})$, one may not in general find a factorization in one particle summation as in the Ewald-3D method. For the bilayer Wigner crystal, $z_{ij}$ may take only two values: if both particles are in the same layer, then $z_{ij} = 0$ or else $z_{ij} = h$.

The intralayer contributions to the long ranged part of the electrostatic energy ($z_{ij} = 0$), for $\kappa \neq 0$ are given by

$$E_{\kappa \neq 0}^{\text{intra}} = \frac{\pi}{A} \sum_{\kappa \neq 0} \frac{\text{erfc}(\kappa/2\alpha)}{\kappa} \left( |\sum_{i \in L_1} q_i \exp(i\kappa.s_i)|^2 + |\sum_{i \in L_2} q_i \exp(i\kappa.s_i)|^2 \right)$$  \hspace{1cm} (12)$$

and for $\kappa = 0$

$$E_{\kappa = 0}^{\text{intra}} = -\frac{\sqrt{\pi}}{\alpha A} \left[ (\sum_{i \in L_1} q_i)^2 + (\sum_{i \in L_2} q_i)^2 \right] - \frac{\alpha}{\sqrt{\pi}} \left( \sum_{i \in L_1} q_i^2 + \sum_{i \in L_2} q_i^2 \right)$$  \hspace{1cm} (13)$$

Since the electroneutrality of the system is achieved with the surface charge density of layers $L_1$ and $L_2$, contributions with $(\sum_{i \in L_2} q_i)^2 \neq 0$ have to be included in the electrostatic energy (see appendix for more details).

The interlayer contributions to the long ranged part of the electrostatic energy for $\kappa \neq 0$ are given by

$$E_{\kappa \neq 0}^{\text{inter}} = \frac{\pi}{A} \sum_{\kappa \neq 0} \frac{F(\kappa, \alpha; h)}{\kappa} \Re\left[ (\sum_{i \in L_1} q_i \exp(i\kappa.s_i))(\sum_{j \in L_2} q_j \exp(-i\kappa.s_j)) \right]$$  \hspace{1cm} (14)$$

where $F(\kappa, \alpha; h)$ is given by Eq.(8) and $\Re[z]$ is the real part of the complex number $z$.

For $\kappa = 0$, the interlayer contribution given by Eq.(10.b) is

$$E_{\kappa = 0}^{\text{inter}} = -\frac{2\pi}{A} \left( \frac{\exp(-\alpha^2 h^2)}{\alpha \sqrt{\pi}} + |h|\text{erf}(\alpha |h|) \right) \left( \sum_{i \in L_1} q_i \right) \left( \sum_{j \in L_2} q_j \right).$$  \hspace{1cm} (15)$$

The electrostatic energy of a configuration of the bilayer Wigner crystal with the Ewald quasi-2D method is given by

$$E = E^{2D} + E_W = (E_{\kappa \neq 0}^{\text{intra}} + E_{\kappa = 0}^{\text{intra}} + E_{\kappa \neq 0}^{\text{inter}}) + (E_{\kappa \neq 0}^{\text{inter}} + E_{\kappa = 0}^{\text{inter}} + E_{\kappa \neq 0}^{\text{inter}} + E_W)$$

with Eqs. (10.a), (12-15) and (A.14).

This expression of the electrostatic energy is an exact application of the Ewald method to the bilayer system; all the computations with Monte Carlo algorithm done with this method are considered in section IV as reference points.

Generally, one may not implement with efficiency the Ewald quasi-2D method, thus there are very few uses of this method. Of course, the procedure described in this subsection
has been used in the study of the solid-phase diagram of the bilayer crystal [32] and also in other numerical studies of electronic bilayer systems [38]. Most applications of the Ewald quasi-2D method in computer simulations use precalculated tables of potential energy, forces and second derivatives on a three-dimensional grid and calculations are performed by interpolation of the tables [39, 35, 20]. In ref.[40], Arnold and Holm have shown that the MMM2D method has a computation effort that scales as $N^{5/3}(\log N)^2$. It is interesting to note that for the bilayer Wigner system, it is possible to implement the Ewald quasi-2D method with the same efficiency as the Ewald 3D method, the computation effort scaling as $N^{3/2}$. Thus, for this system, the Ewald quasi-2D method is slightly more efficient than the MMM2D method.

B. The Hautman-Klein method.

The computation of particle-particle interaction in the Hautman-Klein method starts with the identity [16]

\[ \frac{1}{r} = \frac{1}{r} - \sum_{n=0}^{M} a_n \frac{z^{2n}}{s^{2n+1}} + \sum_{n=0}^{M} a_n \frac{z^{2n}}{s^{2n+1}} \]  

with

\[ a_n = \frac{(-1)^n(2n)!}{2^{2n}(n!)^2} \]

By using damping functions $h_n(s; \alpha)$ on terms $1/s^{2n+1}$, one separates the electrostatic energy of the system in a short ranged part $E_s$ and long ranged part $E_l$,

\[ E^{HK} = E_s + E_l = E^{(ev)}_{pp} \]

where

\[ E_s = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} q_i q_j \sum_{\{\nu\}} \left( \frac{1}{r_{ij,\nu}} - \sum_{n=0}^{M} a_n z_{ij}^{2n} \frac{h_n(s_{ij,\nu}; \alpha)}{s_{ij,\nu}^{2n+1}} \right) \]  

and

\[ E_l = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} q_i q_j \sum_{n=0}^{M} a_n z_{ij}^{2n} \left( \sum_{\{\nu\}} \frac{h_n(s_{ij,\nu}; \alpha)}{s_{ij,\nu}^{2n+1}} \right) \]  

As in the Ewald method, the long ranged contributions are evaluated by using a Fourier transform. In the original derivation by J. Hautman and M.L. Klein the damping functions $h_n(s; \alpha)$ are chosen such as

\[ \begin{align*}
    h_0(s; \alpha) &= \text{erf}(\alpha s) \\
    \frac{h_n(s; \alpha)}{s^{2n+1}} &= \frac{1}{a_n(2n)!} \nabla^{2n} \left( \frac{h_0(s; \alpha)}{s} \right) .
\end{align*} \]
The derivation done by J. Hautman and M.L. Klein with the damping functions given in Eqs.(19) allows to recover easily the analytical derivation of the Ewald method for exact two dimensional systems given in Eqs.(8) by letting \(z_{ij} \to 0\). This is obtained obviously from equations (17-19). Using the damping functions of Eqs. (19), we have

\[
E_s = \frac{1}{2} \sum_{i=1}^{N} \sum_{j \neq i} q_i q_j \left( \frac{1}{r_{ij}} - \frac{\text{erf}(\alpha s_{ij})}{s_{ij}} - \sum_{n=1}^{M} \frac{1}{(2n)!} z_{ij}^{2n} \nabla^{2n} \left( \frac{\text{erf}(\alpha s_{ij})}{s_{ij}} \right) \right)
\]  

and

\[
E_l = \frac{\pi}{A} \sum_{i=1}^{N} \sum_{j=1}^{N} q_i q_j \sum_{n=0}^{M} \frac{1}{(2n)!} z_{ij}^{2n} \sum_{\kappa \neq 0} \kappa^{2n-1} \text{erfc}(\kappa/2 \alpha) e^{i\kappa \cdot s_{ij}}
\]

According Eqs.(16) for \(n = 1 \text{ to } 3\) we have

\[
\begin{align*}
h_1(s; \alpha) & = \text{erf}(\alpha s) - \frac{2\alpha s}{\sqrt{\pi}} e^{-\alpha^2 s^2} (1 + 2\alpha^2 s^2) \\
h_2(s; \alpha) & = \text{erf}(\alpha s) - \frac{2\alpha s}{\sqrt{\pi}} e^{-\alpha^2 s^2} \left(1 + \frac{2}{3} \alpha^2 s^2 - \frac{4}{9} \alpha^4 s^4 + \frac{8}{9} \alpha^6 s^6 \right) \\
h_3(s; \alpha) & = \text{erf}(\alpha s) - \frac{2\alpha s}{\sqrt{\pi}} e^{-\alpha^2 s^2} \left(1 + \frac{2}{3} \alpha^2 s^2 + \frac{4}{15} \alpha^4 s^4 + \frac{8}{25} \alpha^6 s^6 \right) \\
& \quad - \frac{112}{225} \alpha^8 s^8 + \frac{32}{225} \alpha^{10} s^{10})
\end{align*}
\]

The attractive feature of the method is that, by writing the \(z_{ij}^{2n}\) explicitly as polynomials in \(z_i\) and \(z_j\), the long ranged contribution given by Eq.(21) can be cast into a sum of terms each of which involving product of two functions having the general form

\[
F_p(\kappa) = \sum_{i=1}^{N} q_i z_i^p \exp(i\kappa \cdot s_i)
\]

A careful examination of Eqs.(20-21) in one hand and Eqs.(10) in the other hand, shows that Hautman-Klein method may not be considered exactly as Taylor expansion of the Ewald quasi-2D method. In particular, the complicated contribution \(E_{\kappa=0}^\kappa\) in Eq.(10.b) in the Ewald quasi-2D method is roughly approximated in the Hautman-Klein method by a constant. As a consequence, the contributions to intralayer and interlayer energies do not have exactly the same constant terms.

For the bilayer Wigner crystal, the long ranged intralayer contributions \(E_{\kappa \neq 0, HK}^{\text{intra}}\) are
exactly given by Eq.(12) and with Eq.(21), one may expressed $E_{\kappa=0,HK}^{\text{intra}}$ as Eq.(13). Long ranged interlayer contributions are given by

$$E_{\kappa \neq 0,HK}^{\text{inter}} = \frac{\pi}{A} \sum_{\kappa \neq 0} \frac{H(\kappa, \alpha; h)}{\kappa} \Re\left[ \left( \sum_{i \in L_1} q_i \exp(i\kappa.s_i) \right) \left( \sum_{j \in L_2} q_j \exp(-i\kappa.s_j) \right) \right]$$

(24)

with the same notations as in Eq.(14) and

$$H(\kappa, \alpha; h) = \sum_{n=0}^{M} \frac{1}{(2n)!} (\kappa h)^{2n} \text{erfc}(\kappa/2\alpha)$$

(25)

The contribution to interlayer energy for $\kappa = 0$, according to Eqs.(21) and (13) has to be written as

$$E_{\kappa=0,HK}^{\text{inter}} = -\frac{2\sqrt{\pi}}{\alpha A} \left( \sum_{i \in L_1} q_i \right) \left( \sum_{j \in L_2} q_j \right)$$

(26)

instead of the Taylor expansion of Eq.(15).

The electrostatic energy of a configuration of the bilayer Wigner crystal with the Hautman-Klein method is given by

$$E = E_{HK} + E_W = (E_{\kappa=0,HK}^{\text{intra}} + E_{\kappa=0,HK}^{\text{intra}} + E_{\kappa \neq 0,HK}^{\text{intra}})$$

$$+ (E_{\kappa \neq 0,HK}^{\text{inter}} + E_{\kappa=0,HK}^{\text{inter}} + E_{\kappa \neq 0,HK}^{\text{inter}} + E_W)$$

with Eqs.(20), (12), (13), (24), (26) and (A.14).

Because an expansion of $1/r$ for small $z$ is taken as a starting point in Hautman-Klein method, this method is often considered as inaccurate for rather thick systems. Despite its obvious inaccuracy, for slab-like systems, as the system studied in ref.[25], the results found with the Hautman-Klein method are in very good agreement with results obtained with a purely electrostatic model and results obtained with the methods of refs.[18,20]. The Hautman-Klein method may also be implemented with almost the same efficiency as Ewald methods for bulk-like systems, but without taking any periodic images in the direction of the finite extension of the system. Consequently, the Hautman-Klein method is specifically well adapted to systems as fluid-fluid or fluid-solid interfaces, that do not have a slab-like geometry. The Hautman-Klein method may also be implemented to take into account electrostatic images at the interface between two media with different dielectric constants. This method has been used recently in a study of a mixed lipid/non-ionic surfactant membrane [41].

C. The Lekner method.

The third procedure used in the present work is a method proposed by J. Lekner [19]. This method may also be more or less easily related to the original Ewald method.
When applied to quasi-one dimensional systems, Lekner summations method is an eigenfunction expansion of the Green function of Laplace equation with periodicity in only one direction (see for instance Eq.(3.10) in ref.[37]). Following the derivations done in refs.[13,15,37], an Ewald method for quasi-one dimensional systems may be derived [42], the Lekner summations for the quasi-one dimensional systems may be recovered by letting the convergence parameter $\alpha$, defined in Eq.(4), tends to infinity [37, 43].

For quasi-two dimensional systems, the relation between Lekner summations and Ewald summations is slightly more complicated. With the derivation that permits in quasi-one dimensional systems to relate the Lekner summations to an eigenfunction expansion of Green functions, one is obtaining for quasi-two dimensional systems the so-called Nijboer-de Wette representation [12, 13, 44], the same method as the one used recently in the method called MMM2D (see ref.[21(a)] and [45]); Lekner summations are related to these methods by an asymmetric use of the Poisson-Jacobi formula. As for quasi-one dimensional geometries, when the convergence parameter $\alpha$ tends to infinity, one may relate easily the Nijboer-de Wette representation (or the 'far formula' of the MMM2D method [21]) to the Ewald method [37].

Following the original derivation done by J. Lekner [19] and computations done by N. Gronbech-Jensen and co-workers [46-49] and by S. Marshall [50], the electrostatic energy of a quasi-two dimensional system can be computed as

$$E^{LC} = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1, j \neq i}^{N} E_{ij} + \sum_{i=1}^{N} U_{\text{Self}}^i$$

(27)

The interaction energy $E_{ij}$ between pairs is approximated in a numerical computation by one of the two formulas

$$V_{ij}(n_c, n_K) = 4 \frac{q_i q_j}{L_y} \sum_{m=1}^{n_c} \cos(2\pi \frac{y_{ij}}{L_y} m) \sum_{k=-n_K}^{n_K} K_0 \left[ 2\pi m \left( \frac{L_y}{L_y} \right)^2 \left( \frac{x_{ij}}{L_x} + k \right)^2 + \left( \frac{z_{ij}}{L_y} \right)^2 \right]^\frac{1}{2}$$

$$- \frac{q_i q_j}{L_y} \ln \left[ \cosh \left( 2\pi \frac{z_{ij}}{L_y} \right) - \cos \left( 2\pi \frac{x_{ij}}{L_x} \right) \right] - \frac{q_i q_j}{L_y} \ln 2$$

(28)

or

$$U_{ij}(n_c, n_K) = 4 \frac{q_i q_j}{L_x} \sum_{n=1}^{n_c} \cos(2\pi \frac{x_{ij}}{L_x} n) \sum_{k=-n_K}^{n_K} K_0 \left[ 2\pi n \left( \frac{L_x}{L_x} \right)^2 \left( \frac{y_{ij}}{L_y} + k \right)^2 + \left( \frac{z_{ij}}{L_x} \right)^2 \right]^\frac{1}{2}$$

$$- \frac{q_i q_j}{L_x} \ln \left[ \cosh \left( 2\pi \frac{z_{ij}}{L_y} \right) - \cos \left( 2\pi \frac{y_{ij}}{L_y} \right) \right] - \frac{q_i q_j}{L_x} \ln 2$$

(29)

The integers $n_c$ and $n_K$ are the truncation parameters that would govern the accuracy of the finite summations [24, 37]. The main technical problem that one would encounter in trying to implement straightforwardly this procedure is in the convergence rate of one of the two summations for some special configurations of the pair of particles $i, j$ relatively
to the geometry of the simulation box. For these particular configurations, that are very frequent when few billions of configurations are sampled, one of the two summations given by Eqs.(28) and (29) is very slowly convergent and a lot of contributions must be included to compute the correct value of the energy. The origin of this slow convergence rate is in the behavior of the modified Bessel function $K_0$ as its argument tends to zero (see ref.[24]). To implement correctly the Lekner summations in a Monte Carlo algorithm or in a Molecular dynamics simulation, one must use a procedure to overcome this poor convergence rate or else some bias would plague the simulation. To achieve this aim, several methods have been proposed. A procedure frequently used consist in applying a cyclic symmetry that is found in the original derivation of the method [19]. In the following, we restrict ourself to this procedure and the implementation of the Lekner summations according to this procedure is called the Lekner-cyclic method. At the end of this subsection we will review briefly other procedures proposed to overcome the poor convergence rate of one of the summations $U_{ij}(n_c, n_K)$ or $V_{ij}(n_c, n_K)$.

The cyclic symmetry consists in recognizing that as $n_c \to \infty$ and $n_K \to \infty$, we have

$$
\lim_{n_c \to \infty, n_K \to \infty} V_{ij}(n_c, n_K) = \lim_{n_c \to \infty, n_K \to \infty} U_{ij}(n_c, n_K) = E_{ij} \quad (30)
$$

For the particular configurations where one of the two summations is slowly convergent, to compute $E_{ij}$ one may use $V_{ij}(n_c, n_K)$ or $U_{ij}(n_c, n_K)$ (refs.[19,47,50]). Since for given $n_c$ and $n_K$, $V_{ij}(n_c, n_K)$ and $U_{ij}(n_c, n_K)$ may have very different numerical values, the application of the cyclic symmetry may introduce complicated bias [24]. In particular, if $n_c$ and $n_K$ are badly chosen, the value for $E_{ij}$ found in computing energy of particle $i$ in the electrostatic potential of the particle $j$ may be different of the value found for $E_{ji}$ when the energy is computed by considering the particle $j$. An inequality as $E_{ij} \neq E_{ji}$ happens here because the configuration of the vector $s_{ij}$, relatively to the simulation box, is different from the configuration of the vector $s_{ji}$, especially when the minimum image convention is used.

The study done in ref.[24] is helpful to define a criterion on $s_{ij}$ to choose to compute $V_{ij}(n_c, n_K)$ rather than $U_{ij}(n_c, n_K)$ or the converse.

For the bilayer Wigner crystal, the interaction energy $E_{ij}$ between pair of particles is computed as

$$
E_{ij} = \begin{cases} 
V_{ij}(n_c, n_K) & \text{if } |x_{ij}| > |y_{ij}| \\
U_{ij}(n_c, n_K) & \text{if } |x_{ij}| < |y_{ij}| 
\end{cases} \quad (31)
$$

The criterion used here is quite simple because shapes of the layers are squares of side $L$.

General formulas as Eqs.(28) and (29) for Lekner summations, when the base of unit cell is not a square ($L_x \neq L_y$) or do not have a rectangular shape, have been proposed in refs.[48,51-53]; for these geometries a careful examination of the criterion given by Eq.(31) should be done, especially when the base of the unit cell is not rectangular.

To compute, with the cyclic symmetry, the energy of a configuration made of $N$ particles, the number of different contributions to evaluate is of the order $n_c(2n_K+1)N^2$. As it was
shown in our previous work [24], when the criterion given by Eq.(31) is used, for some configurations of the vector \( s_{ij} \) the value computed by using the most rapidly convergent of the two formulas (28) and (29) becomes independent of the direction of the vector \( s_{ij} \) for rather high values of \( n_c \) (e.g. \( n_c \simeq 40 \) for a relative accuracy of \( 10^{-5} - 10^{-6} \)) [24, 37]. On the other hand, for most of the configurations of the vectors \( s_{ij} \), the convergence of summations (28) and (29) are more rapid, thus a value as large as \( n_c \simeq 40 \) is not needed in all computations done with the Lekner summations. Then, to improve the efficiency of the method, \( n_c \) is not chosen independently of the argument of the Bessel function. Taking into account the asymptotic behavior of the Bessel function \( K_0(x) \)

\[
K_0(x) \simeq \sqrt{\frac{\pi}{2x}} \exp(-x) \quad \text{as} \quad x \to \infty
\]

truncation to \( n_K = 3 \) or 4 is enough to achieve a good accuracy \( (K_0(19.) \simeq 1.6 \times 10^{-9}) \).

The value of \( n_c \) used is chosen such as

\[
1 \leq n_c(s_{ij}) \leq n_c^{\text{max}}
\]

where \( n_c^{\text{max}} \) is a prefixed value and \( n_c(s_{ij}) \) the value of \( n_c \) chosen in such a way that the output of the summation, Eq.(28) or Eq.(29), would be independent of the vector \( s_{ij} \).

In the computations presented in the next section, \( n_c(s_{ij}) \) is chosen as the first integer such as

\[
2\pi n_c(s_{ij})\left[\left(\frac{y_{ij}}{L} + k\right)^2 + \left(\frac{z_{ij}}{L}\right)^2\right]^{\frac{1}{2}} > 19. \quad (33)
\]

If the integer found by using Eq.(33) is greater than \( n_c^{\text{max}} \), then one takes \( n_c(s_{ij}) = n_c^{\text{max}} \).

For each value of the term between the brackets in Eq.(33), a value of \( n_c(s_{ij}) \) is taken from a precomputed table that takes into account the prescription imposed by \( n_c^{\text{max}} \).

This evaluation of \( n_c(s_{ij}) \) is done for all pairs of particles. For the bilayer Wigner crystal this procedure is used for both the intralayer and interlayer contributions.

This procedure has been applied to all computations done with the Lekner-cyclic method presented in section IV, in this section \( n_c \) denotes the value of \( n_c^{\text{max}} \) used in Eqs.(32) and (33).

For the bilayer Wigner crystal, the separation of the energy into intralayer and interlayer contributions is done as in the Ewald quasi-2D and Hautman-Klein methods as

\[
E^{LC} = E^{LC}_{\text{intra}} + E^{LC}_{\text{inter}} = E^{(cv)}_{pp}
\]

with

\[
E^{LC}_{\text{intra}} = \frac{1}{2} \sum_{i \in L_1, j \in L_1, i \neq j} E_{ij} + \frac{1}{2} \sum_{i \in L_2, j \in L_2, i \neq j} E_{ij} + \sum_{i \in L_1} U^\text{Self}_i + \sum_{i \in L_2} U^\text{Self}_i
\]

where

\[
U^\text{Self}_i = \frac{1}{2} \lim_{r_i \to r_j} \left( E_{ij} - \frac{q_i^2}{r_{ij}} \right) = \frac{q_i^2}{L} \left[ 4 \sum_{m=1}^{\infty} \sum_{k=1}^{\infty} K_0(2\pi mk) + \gamma - \ln(4\pi) \right]
\]
and

\[ E_{\text{inter}}^{LC} = \sum_{i \in L_1} \sum_{j \in L_2} E_{ij} \]  

(37)

For the bilayer Wigner crystal, we have

\[ V_{ij}^{\text{intra}}(n_c, n_K) = 4 \frac{q_i q_j}{L} \sum_{m=1}^{n_c} \cos\left(\frac{2 \pi m}{L} y_{ij}\right) \sum_{k=-n_K}^{n_K} K_0\left(2 \pi m \frac{x_{ij}}{L} + k\right) \]

\[ -\frac{q_i q_j}{L} \ln \left[1 - \cos\left(2 \pi \frac{x_{ij}}{L}\right)\right] - \frac{q_i q_j}{L} \ln 2 \]  

(38)

for intralayer contributions, and

\[ V_{ij}^{\text{inter}}(n_c, n_K) = 4 \frac{q_i q_j}{L} \sum_{m=1}^{n_c} \cos\left(\frac{2 \pi m}{L} y_{ij}\right) \sum_{k=-n_K}^{n_K} K_0\left[2 \pi m \left(\frac{x_{ij}}{L} + k\right)^2 + \left(\frac{h}{L}\right)^2\right]^{\frac{1}{2}} \]

\[ -\frac{q_i q_j}{L} \ln \left[\cosh\left(2 \pi \frac{h}{L}\right) - \cos\left(2 \pi \frac{x_{ij}}{L}\right)\right] - \frac{q_i q_j}{L} \ln 2 \]  

(39)

for interlayer contributions and similar relations for \(U_{ij}^{\text{intra}}\) and \(U_{ij}^{\text{inter}}\) (interchanging \(x_{ij}\) with \(y_{ij}\)).

The electrostatic energy of a configuration of the bilayer Wigner crystal with the Lekner-cyclic method is given by

\[ E = E^{LC} + E_W = E_{\text{intra}}^{LC} + (E_{\text{inter}}^{LC} + E_W) \]  

(40)

with equations (35) to (39) and (A.14).

The Lekner summations method has been used in few studies using molecular simulation algorithms.

The cyclic symmetry has been used recently in a Molecular Dynamics study of thin water-acetonitrile films [54]: in this work an appendix that describes the method used to implement the Lekner-cyclic method may be found. In recent Monte Carlo simulations of adsorption of colloidal particles on a charged surface [55], the Lekner-cyclic method is also used [56], but technical details on the implementation are not given in ref.[55].

To implement correctly the Lekner summations method, other procedures have been proposed. In the present work, none of these procedures, reviewed briefly below, have been neither implemented, nor compared to the Lekner-cyclic or Ewald quasi-2D methods.

In ref.[57], J.F. Harper proposed to use an integral representation of summations when arguments of Bessel functions are small. This method might be well adapted for quasi-one dimensional systems [42] where, instead of having two different analytical relations, Eqs.(28) and (29), one has only one summation. In these situations one cannot use the cyclic symmetry.

Another procedure is to derive an alternative expression which converges fast as the argument of the Bessel functions tends to zero. Using the Hurwitz zeta function, R. Sperb...
had proposed such alternative expressions [58]. The Sperb method has been used in a Molecular Dynamics study of strongly charged polyelectrolyte brushes [57]; in this work one would find an appendix that describes the procedures used by authors. In ref.[43], A. Grzybowski and A. Bródka also used a procedure close to the Sperb method for quasi-one dimensional systems. The Lekner-Sperb method was also used by A.G. Moreira and R.R. Netz in their studies of counterions distributions near charged plates [60, 61]. In ref.[61], Moreira and Netz give a detailed and very clear description of their implementation of Lekner summations. In their implementation they use Sperb transformation with the following procedure: if \( \rho = \sqrt{(y+k)^2 + z^2} \geq 1/3 \), they use relations similar to Eqs.(28) and (29) of the present work with truncation parameters \((n_c, n_K) \simeq (11, 2)\), if \( \rho < 1/3 \) Sperb formula is used. As it will be shown in section IV, when Lekner-cyclic method is used, truncation parameters as \((n_c, n_K) \simeq (10, 3)\) do not give accurate results. Nevertheless, one should not conclude that the truncation parameters used by Moreira and Netz are too small to give accurate results; in the core of their method they use Sperb formula which main purpose is to reduce these truncation parameters. Implementation done in ref.[61] and in the present work are not exactly identicals.

Another procedure has been developed by J.N. Newman based on two-dimensional Chebyshev expansions and economized polynomials [62].

There are few others studies that used Lekner summations on very different, interesting and complicated systems [55, 63-67], but details on implementation are not given there. Although the cyclic symmetry is certainly the procedure the mostly used for quasi-two dimensional systems, details on the implementation are needed. Firstly, there are at least three different methods to implement the Lekner summations method. Secondly, as it will be shown in section IV, the results of a computation may be depending on parameters and criterions used to correct the slow convergence rate of one of the two summations given by Eqs.(28) and (29). Moreover, the criterions used to implement the Lekner cyclic method in Eqs.(31), (32) and (33) are depending on the geometrical parameters of the simulation box or on parameters of the studied system.

### IV. Results.

In this section, we present the results obtained with the three methods of the previous section.

The three computer codes used in this work are very similar. The main differences between the three programs are in the subroutines that compute the electrostatic energy, all others parts of the programs are the same. In particular, the files in which configurations are written may be used in any of the three programs, the random numbers generator used is the same and it is possible, with our codes, to start computations for each of the three methods from exactly the same initial condition and with exactly the same sequence of random numbers for the trial moves of the particles and for the acceptance tests. This point was necessary not only to compare the compatibility of the methods,
but also to test the efficiency and the accuracy of the methods. This section is made of two subsections. In subsection IV.A, using the same initial configuration and the same random numbers sequence, we compare the sampling of the phase space of the bilayer Wigner crystal done with the three methods. In subsection IV.B, the results of computations for the Runs defined in TABLE I are given. All the computations were performed on a Silicon Graphics Origin 2000 (R10000/195 processors).

A. Accuracy of the sampling of the phase space.

In the Monte Carlo Metropolis algorithm [1, 2, 33], the phase space of the system studied is sampled by an importance-weighted random walk. From an initial configuration of the system, its phase space is sampled by a construction of configurations in such a way that configurations are generated with a probability proportional to the statistical weight of the configurations. A convenient way is to generate a trial configuration from an old configuration of the system and to accept or reject the trial configuration according to an acceptance probability defined with the help of the detailed balance condition. According to this procedure, if the trial configuration is accepted, it becomes the new configuration of the system, or else, if the trial configuration is rejected, the old configuration is taken as the new configuration. The procedure is implemented by building another trial configuration from the new configuration.

When the trial configuration is generated only from an old configuration and when the acceptance probability may only be defined with the knowledge of the old and trial configurations, the chain of configurations generated by the Metropolis algorithm has all the properties of a Markov chain. All ensembles of the statistical mechanics can be sampled with the Metropolis algorithm [1].

To apply the Monte Carlo algorithm to the canonical ensemble, the simplest procedure is to generate trial configurations from old configurations by a random displacement of one particle of the system. The displacement of the particle (the new configuration of the system) is then accepted according to a probability given by

\[
\text{acc}(o \rightarrow n) = \min(1, \exp(-\beta(E_n - E_o)))
\]

where \(E_n\) is the total energy of the trial (new) configuration and \(E_o\) the total energy of the old configuration. For these procedures, the accuracy of the computation of the energy is closely related to the accuracy of the sampling of the phase space of the system. For the bilayer Wigner crystal, the energy difference used in the acceptance probability of Eq.(41) is computed by using one of the three methods of section III and in the following we set

\[
\beta \Delta E^{(cv)}_{pp} = -\beta(E_n - E_o)
\]

Since for the Lekner-cyclic method, configurations where the point ions are on a regular two dimensional lattices are very penalizing for the convergence of summations, these kinds of configurations were not taken as initial configurations. To generate initial configurations on which an examination of the influence of the truncation parameters \(n_c\)
and \( n_K \) could be done, we have chosen to build initial configurations as follows. \( N_0 \) point ions are arranged on a square lattice in each layers; this configuration is taken as the initial configuration for the code using the Ewald quasi-2D method. From this initial configuration, 100 MC-cycles were performed with the Ewald quasi-2D method. After these 100 MC-cycles (200\( N_0 \) trial moves), the point ions are no longer arranged in a perfect regular lattice, thus this configuration may be taken as an initial configuration for all the three methods. This procedure presents also the advantage of allowing to start computations from a well defined configuration that we may reproduce easily, if the same sequence of random numbers is taken.

If codes are written such as the random numbers are generated following the same sequence and used for the same purpose in each of the three codes, then, if the three methods give exactly the same energy, the trajectory in the phase space obtained by the Markov chain of configurations would be exactly the same in the three computations. The trajectory followed by the system with the Ewald quasi-2D method is considered as the exact trajectory; the trajectories obtained with the Hautman-Klein and Lekner-cyclic methods are compared to this trajectory. In TABLE II, we give some elements to appreciate the accuracy of the sampling of the phase space by the Hautman-Klein and Lekner-cyclic methods relatively to the sampling obtained with the Ewald quasi-2D method for initial configuration that corresponds to Runs a (cf. TABLE I). The energy difference \( \beta \Delta E_{pp}^{(cv)} \) for the first trial move of the first MC-cycle from the same initial condition with the same sequence of random numbers is given for different truncation parameters for the Hautman-Klein and Lekner-cyclic methods. This first move is particularly interesting, whatever it is accepted or rejected, the move is identical in the three methods and the position of all the other particles are also the same in the three methods. The values of \( \beta \Delta E_{pp}^{(cv)} \) show that the truncation parameter \( n_c \) in the Lekner-cyclic method governs the accuracy of the computations, while \( n_K \) seems to have less incidence on accuracy. In agreement with our previous work [24], to reach a relative accuracy of \( 10^{-4} - 10^{-5} \), one needs to choose \( n_c \approx 40 \), while \( n_K = 2 \) or 3 seems to be sufficient. A careful examination of other energy differences shows that, for few particular moves, one needs to take \( n_K = 3 \) to avoid a lost of accuracy; nevertheless these events are rare and in almost all cases, \( n_K = 2 \) might be sufficient. On the other hand, \( n_c < 25 \) always leads to important differences in almost all energy differences.

Because energy differences in the three methods are not exactly the same, the trajectory in the phase space followed by the system are not exactly the same. In TABLE II, we give also the number of trial moves accepted after the first MC-cycle \((t=1)\) and after the first hundred MC-cycles \((t=100)\) from the same initial condition. In a deterministic point of view, the trajectory in the phase space, using the Metropolis algorithm, has to be independing of the method used to compute the energy. Of course, in almost all molecular simulations, such deterministic point of view is unreachable, at least because of round-off errors done by the machines on floating points [68]. In our study, as soon as a random trial move in the computation using the Ewald quasi-2D method is accepted (or rejected), while it is rejected (or accepted) in computation using Hautman-Klein of
Lekner-cyclic methods, the trajectories are no longer the same. Since in all situations examined, one may not exactly follow the same trajectory with the Hautman-Klein and Lekner-cyclic methods as with the Ewald quasi-2D method, we should find a criterion to estimate if these methods are enough accurate to give reliable statistical averages.

To answer this question, one has to come back to the principles of the Monte Carlo sampling. To obtain reliable thermodynamical averages by making a Monte Carlo sampling, one has to generate configurations with a probability that would be proportional, with a rather good accuracy, to the statistical weight of the configurations. Thus, it is not necessary that the trajectories followed by the system will be exactly the same, even if the initial configurations and the sequence of random numbers are the same. The important point is to generate configurations with a probability proportional to the statistical weight of the configurations; if this is not achieved, the thermodynamical averages are biased in a more or less complicated way. To estimate the bias from the ‘exact’ results given by the Ewald quasi-2D method, we have chosen to follow the instantaneous energy per particle. In Figure 2 (a-d), we have plotted for the first 500 MC-cycles the instantaneous energy per particle obtained with the Ewald quasi-2D on one hand, and with others methods on another hand, for the same initial configuration and with the same sequence of random numbers. The instantaneous energy of the initial configuration is given at $t = 0$.

In Figure 2.a, the energies using Hautman-Klein method ($M = 3$) and Ewald quasi-2d are shown. An examination of the first 50 MC-cycles shows that both trajectories are exactly the same for these first moves. After, it happens that a trial move is accepted in one of the two methods, while it is rejected in the other. Trajectories separate at this point. Nevertheless, fluctuations of this quantity are still in good agreement in the two methods. This is an indication, but not a demonstration, that both methods would give same statistical averages and thus it seems that there are no bias, relatively to the Ewald quasi-2D method, in using Hautman-Klein method for the geometrical parameters corresponding to Runs a.

The situation in Figure 2.b, where the same quantities are plotted for Ewald quasi-2D method and Lekner-cyclic method with $(n_c, n_K) = (10, 3)$, is different. Trajectories separate in the first trial moves of the first MC-cycle (see also TABLE II). For the first 50 MC-cycles, fluctuations of the instantaneous energy seem to be similar, but after 100 MC-cycles the trajectories clearly separate, much more than two magnitudes of the fluctuation. In this case, as it will be shown in the next subsection, the statistical averages are different.

Since for $n_c = 10$ and $n_K = 3$ as truncation parameters in the Lekner-cyclic method the accuracy is not correct, we made the same numerical analysis for higher value of these parameters. This is shown on Figure 2.c and 2.d. In Figure 2.c the truncation parameters are $n_c = 25$ and $n_K = 3$; the trajectories do not separate for the few first trial moves but only after the few first MC-cycles, despite the fact that the relative accuracy on the energy difference is only $10^{-2}$ compared to the energy difference found for the Ewald quasi-2D method. For the 500 MC-cycles shown in this Figure, with $n_c = 25$ and $n_K = 3$, trajectories stay in good agreement. If one increases the value of $n_c$, as it is done
in Figure 2.d where $n_c = 50$, one would find a better agreement between Ewald quasi-2D and Lekner-cyclic methods. For $n_c = 50$ and $n_K = 4$, the separation occurs only after 60 MC-cycles and after the trajectories stay very close. For both cases, $(n_c, n_K) = (25, 3)$ and $(n_c, n_K) = (50, 4)$ one may hope to obtain the same statistical averages than in the Ewald quasi-2D method. Of course a dramatic separation, as for $(n_c, n_K) = (10, 3)$, may still happen for both $(n_c, n_K) = (25, 3)$ and $(n_c, n_K) = (50, 3)$ after more MC-cycles, this had not happened for the first 10000 MC-cycles examined.

B. Energy and pair distribution functions.

To estimate statistical averages, we have mainly considered two different cases. A case where the separation between the two layers is small ($h = 1.0$) for which the coupling between the layers is important: this corresponds to Runs a and b. We have also considered another case where the separation between layers is large enough ($h = 4.0$) so that both layers are almost independent [32], Runs c to f.

Since for $h = 1.0$, the coupling between the layers is important, this has an incidence on the interlayer pair distribution functions $g_{12}$. This separation between the layers allows us to estimate the agreement between methods for small $z$. On the other hand, because for $h = 4.0$ the coupling between the two layers is very small, one would find for almost all $s$, $g_{12}(s) \simeq 1$. One may estimate on these computations with $h = 4.0$ the influence of the periodic boundary conditions by varying $N$.

In TABLE III, the results for Runs a and b are given, $U/N$ is the average energy per particle where $U$ is computed in the three methods as

$$U = <E>$$

$E_{\text{inter}}/N$ and $E_{\text{intra}}/N$ are respectively the interlayer and intralayer average energies per particle computed according section III; and $\sigma_U$ is an estimation of the statistical fluctuation of the total energy computed as

$$\sigma_U^2 = <E^2> - <E>^2$$

On Figures 3 to 6, we give the intralayer $g_{11}(s)$ and interlayer $g_{12}(s)$ pair distribution functions obtained with the Ewald quasi-2D and other methods for Runs a and b.

The value of $\sigma_U$ given in TABLE III can be used to give an estimation of the accuracy needed on the average energy. The fluctuation $\sigma_U$ is the average of the fluctuations observed on Figures 2(a-d). The results given on TABLE III show that the Ewald quasi-2D and Hautman-Klein methods in one hand and the Lekner-cyclic method with $(n_c, n_K) = (25, 3)$ on the other hand are in a very good agreement for an estimation of the energy for Runs a and b. Using the results of TABLE III, one may check that the difference between the average energy obtained using Ewald quasi-2D method and Hautman-Klein method or Lekner-cyclic method with $(n_c, n_K) = (25, 3)$ is lesser than
2 \sigma_U$. For Runs a, one has $|U_{Ewald} - U_{HK}| \simeq 0.2 \sigma_U$ for Hautman-Klein method and $|U_{Ewald} - U_{Lekner}| \simeq 0.4 \sigma_U$ for Lekner-cyclic method with $(n_c, n_K) = (25, 3)$, while for Runs b, one has $|U_{Ewald} - U_{HK}| \simeq 0.4 \sigma_U$ for Hautman-Klein method and $|U_{Ewald} - U_{Lekner}| \simeq 1.7 \sigma_U$ for Lekner-cyclic method with $(n_c, n_K) = (25, 3)$. The lost of accuracy in the Lekner-cyclic method with $(n_c, n_K) = (25, 3)$ for Runs b may be well understood by observing that as $L$ increases the argument of the Bessel functions in Eqs.(38) and (39) decreases, thus one has to increase $n_c$ to achieve a better accuracy. As shown on Figures 3 and 5 the pair distribution functions are also in good agreement, but with a small lost of accuracy for Runs b in both Hautman-Klein method and Lekner-cyclic method with $(n_c, n_K) = (25, 3)$ compared to the Ewald quasi-2D method. With $n_c$ raised to 40, one obtains a very good agreement between the Ewald quasi-2D and Lekner-cyclic method for Runs a and b, as it is shown on Figures 6.

With the same criterions, it appears that the results for the Lekner cyclic method with $(n_c, n_K) = (10, 3)$ are not in a good agreement with the Ewald quasi-2D method neither for Runs a, nor for Runs b. For Runs a, one has $|U_{Ewald} - U_{Lekner}| \simeq 4.2 \sigma_U$ (see also Fig.2(b)) and for Runs b $|U_{Ewald} - U_{Lekner}| \simeq 5.6 \sigma_U$. Then, as it was mentioned in the previous subsection, the truncation parameters $(n_c, n_K) = (10, 3)$ are badly chosen to give reliable statistical averages in this implementation of the Lekner-cyclic method. The bias induced by a bad choice of the convergence parameters in Lekner summations is exhibited on Figs.4(a-b). The pair distribution functions, computed with the Lekner-cyclic method $(n_c, n_K) = (10, 3)$ for Runs a, differ dramatically from the pair distribution functions computed with the Ewald quasi-2D method. As it is shown on Figs.5 (a-b), when the value of the truncation parameter $n_c$ is raised to 25, pair distribution functions $g_{11}$ and $g_{12}$ computed with the Lekner-cyclic method are exactly the same as those computed with the Ewald quasi-2D method. For Runs b, the observations done on Figs.4(c-d) and Figs.5(c-d) are not as clear as it was for Runs a; nevertheless one may not conclude that for the thermodynamical point corresponding to Runs b, truncation parameters $(n_c, n_K) = (10, 3)$ are enough to avoid bias, since the average energy is very badly estimated. This point could be misleading when testing the implementation of the Lekner method in a computer code where no reference points, as those provided by the Ewald quasi-2D method, can be found. As mentioned in TABLE I, for Runs b the preferential structure of the layers is a disordered fluid-like structure, thus there is less configurations of pairs of particles with $x_{ij} \simeq 0$ or $y_{ij} \simeq 0$ compared to a crystal-like ordered structure. As a consequence, summations given by Eqs.(38) and (39) converge rapidly for most of pairs of particles and the bias induced by the poor convergence rate of summations for few pairs of particles is hidden. This situation becomes very dangerous when a fluid-solid equilibrium is studied with these parameters. We must outlined also that even if the average energy is badly evaluated with truncation parameter $n_c = 10$, the value found with this parameter is not very different from the correct value given by the Ewald quasi-2D method, because of the oscillatory behavior of summations given by Eqs.(38) and (39) (see reference [24]). This point could also be misleading when tests on implementation of the Lekner method are done.

For $h = 4.0$ (Runs c to f), the statistical averages are given on TABLE IV with same
notations. All these computations are done for the same thermodynamical point but with different values of the number of particles ranging from \( N = 128 \) to 968. According to the results of TABLE IV, the Ewald quasi-2D and Hautman-Klein methods are in a good agreement despite the large separation of layers \((h = 4.0)\) and except for Runs c with \( N = 128 \). For Runs d, e’ and f, one has \(| U_{\text{Ewald}} - U_{\text{HK}} | \leq 0.7 \sigma U\). On the other hand, for Runs c, where \( N = 128 \) we have \(| U_{\text{Ewald}} - U_{\text{HK}} | \simeq 3.5 \sigma U\) and the value found for \( E_{\text{inter}} \) is not correct with the Hautman-Klein method. This is illustrated on Figs.7 (a-b) where it is shown that the pair distribution functions computed with Ewald quasi-2D and Hautman-Klein methods are not in agreement. By using the Hautman-Klein method with the geometrical parameters of Runs c, one finds a coupling between both layers, while with the Ewald quasi-2D method no coupling appears. This lost of accuracy in the Hautman-Klein method for Runs c is introduced by the expansion of \( 1/r \) for small \( z \), one has \( h/L \simeq 0.3 \) for \( N = 128 \), while for \( N \geq 338 \), we have \( h/L < 0.2 \), but with the same density of point ions. Figures 7 and TABLE IV show that to increase the accuracy of the Hautman-Klein method one may increase \( N \).

The finite-size effect is also outlined on Figures 7 for the Ewald quasi-2D method. The pair distribution function \( g_{11} \) is sensitive to the finite size of the simulation box, this is illustrated in Fig.7(a) up to Fig.7(g) where \( N \) is ranging from 128 to 968.

Comparison between Ewald quasi-2D and the Lekner cyclic methods are done in TABLE IV and in Figures 8 and 9, for Runs c to f. As for Runs a, the Lekner-cyclic method with \((n_c, n_K) = (25, 3)\) is in very good agreement with the Ewald quasi-2D method. For Runs c to f, one has \(| U_{\text{Ewald}} - U_{\text{Lekner}} | < 1.5 \sigma U\) for \( N \geq 338 \) and \(| U_{\text{Ewald}} - U_{\text{Lekner}} | \simeq 1.8 \sigma U\) for \( N = 128 \). For \((n_c, n_K) = (10, 3)\), TABLE IV shows that as \( N \) is increased, the Lekner-cyclic method lost its accuracy. For Runs c with \((n_c, n_K) = (10, 3)\), one has \(| U_{\text{Ewald}} - U_{\text{Lekner}} | \simeq 1.5 \sigma U\) and for Runs f one finds \(| U_{\text{Ewald}} - U_{\text{Lekner}} | \simeq 12 \sigma U\). This point is well illustrated on the representation of \( g_{11} \) given on Figures 8. As \( N \) is increased from 128 to 968, the structure functions computed with the Lekner-cyclic method become poorly evaluated. With \((n_c, n_K) = (10, 3)\), for \( N = 128 \), \( g_{11} \) and \( g_{12} \) are in very good agreement with the functions computed with the Ewald quasi-2D method for \( N = 128 \) (Figs.8 (a-b)), but as \( N \) is increased, this accuracy is lost. This lost of accuracy is similar to the one observed in Runs b for the Lekner-cyclic method with \((n_c, n_K) = (25, 3)\) since as \( N \) is increased, \( L \) is increased and thus the argument of the Bessel functions decreased. This point may also introduce bias when an implementation of the Lekner method is done. Before making extensive computations, one frequently checks the accuracy of the computation on systems having very few particles. For systems with long ranged interactions, a good estimation of some kind of Mandelung constants is sometimes taken as a test of the method [19, 69]. Thus, with this kind of tests, one may conclude using systems with very few particles that a small value of \( n_c \) gives accurate results, but this accuracy would be lost as the number of particle is increased. Tests on systems with a small numbers of particles (i.e. \( N \leq 150 \)) are not sufficient to check the accuracy of the implementation of the Lekner-cyclic method. As it is shown on Figure 9, if one raises \( n_c \) to 25, the Lekner-cyclic and the Ewald quasi-2D methods are in very good agreement for Runs c to f, both for the average energy and pair distribution functions.
On TABLES III and IV, we give also a crude estimation of the average CPU-time in seconds per MC-cycle. These estimations are depending on machines and codes. Improvement of the efficiency may certainly be achieved; nevertheless these CPU-times may be intercompared to give an estimation of the relative efficiency of the methods. The average CPU-time per MC-cycle found for the Ewald quasi-2D method in TABLE IV may not be considered as a representative efficiency of the method, since for the particular system considered in this work the Ewald quasi-2D is as efficient as an Ewald-3D method (see subsection III.A). TABLE IV shows that the Lekner-cyclic method may become particularly expensive as $N$ is increased (see also ref.[61]). With Eq.(33), one may estimate the value of $n_{c}^{\text{max}}$ to achieve enough accuracy with the Lekner-cyclic method as $N$ is increased, one roughly has

$$n_{c}^{\text{max}} \sim \frac{19L}{2\pi h} \sim N^{\frac{1}{2}}$$

(45)

Thus, the computing time with the implementation proposed in subsection III.C scales like

$$\delta t \sim N^{\alpha} \quad \text{with} \quad 2 \leq \alpha \leq \frac{5}{2}$$

(46)

V. Discussion

The study done in this paper gives a comparison between Lekner, Ewald quasi-2D and Hautmann-Klein methods when implemented in a Monte Carlo simulation. In particular the influence of the methods on thermodynamical and structural quantities is outlined on a very simple model on which the Ewald quasi-2D method may be efficiently implemented. Thus, the Ewald quasi-2D method and the bilayer Wigner system may serve as reference tools to gain on physical quantities of general interest, concretely (i.e. when implemented in Monte Carlo Metropolis or in Molecular Dynamics simulations) the accuracy and efficiency of all other methods proposed to handle quasi-bidimensional systems. One of the main results of this paper is to demonstrate that the Ewald quasi-2D method and the Lekner summation technique are in complete agreement. In refs.[51,67], authors have compared the Lekner method with the Hautman-Klein method; the outputs of the computations presented in this paper agree with the results of these authors.

According the computations done here, we may not conclude that the Lekner summations are an efficient alternative to the Ewald quasi-2D method, at least for the implementation that uses the cyclic symmetry (Lekner-cyclic method).

The procedure to implement the Lekner-cyclic method, described in the subsection III.C of the present paper, is successful to use correctly the Lekner summations in a Monte-Carlo Metropolis algorithm. This procedure was based on a previous work where a study of the convergence of the Lekner summations is done [24]. The truncation parameter $n_{c}$ defined in ref.[24] and in Eqs.(28) and (29) governs the accuracy of the sampling of the phase space of a system when Lekner summations are used in Monte-Carlo Metropolis
algorithms. Some severe bias may be induced when the method is straightforwardly implemented. These bias are generated by the diverging behavior of the Bessel functions as its argument tends to zero. We may not recommend to use a precise value of the truncation parameter \( n_c \) to always implement the Lekner method with accuracy, because the value of \( n_c \) that must be used is depending on the geometrical parameters of systems studied. Thus, for each system a convergence study is to make before performing intensive computations.

As explained in subsection IV.B, some tests may be misleading to check the accuracy of the implementation. One of the most difficult behavior to handle is the fact that the needed value of \( n_c \) to achieve good accuracy is depending on the number of particles, because the spatial periodicity appears explicitly in the argument of the Bessel function (see also Figures 8). The finite size effects in Lekner summations are much more difficult to handle, than in Ewald methods; especially because of the asymmetry of Eqs.(28) and (29).

In Ewald methods, it is required that energies are not depending on the convergence parameter \( \alpha \). The same requirement is to achieve with the truncation parameter \( n_c \) in Lekner methods.

The proposed procedure in this present work to implement the Lekner method in molecular simulations is accurate, but unfortunately this procedure is not very efficient. Others procedures have been proposed to correctly implement the Lekner summations [55, 56, 60]. These procedures have not been considered in the present work. Some of these procedures may improved the efficiency of the method.

The bilayer Wigner crystal used as a reference model in this work is very convenient, since with the Ewald quasi-2D method exact results may be obtained. Thus, this model and the results of TABLE III and TABLE IV may be used as references to test the accuracy of other methods for quasi-two dimensional systems.

Lekner summations have also been used to compute Mandelung constants [70]. In these computations, the Lekner summations are useful and may be considered as an alternative to Ewald methods, mainly because the spatial periodicities that are used in these computations are much smaller than the spatial periodicities used in molecular simulations.

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Appendix: Electroneutrality and computation of the interaction between two charged surfaces and ions.

For the system described in Section II of the present work and more generally for a system made of \(N\) charged particles of charge \(q\) and two parallel surfaces \((L_1 \cup L_2)\) separated by a distance \(h\) with a surface charge density \(\sigma(r)\), electroneutrality is given by

\[
Nq + \int_{L_1 \cup L_2} dr \sigma(r) = 0 \tag{A.1}
\]

Using conventional notations for the periodic boundary conditions, the electrostatic energy of this system is given by

\[
E = \frac{q^2}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} \left( \sum_{n} \frac{1}{|r_{ij} + n|} \right) - q \sum_{i=1}^{N} \int_{L_1 \cup L_2} dr \sum_{n} \frac{\sigma(r)}{|r_i - r + n|} + \frac{1}{2} \int_{L_1 \cup L_2} dr dr' \sum_{n} \frac{\sigma(r)\sigma(r')}{|r' - r + n|} \tag{A.2}
\]

\(= E_{pp} + E_{pS} + E_{SS}\)

The three contributions to the electrostatic energy are: (a) \(E_{pp}\) the particle-particle interaction energy evaluated in this work by using one of the three methods (i.e. Ewald quasi-2D, Hautman-Klein or Lekner-cyclic methods) where self energies are included in \(E_{pp}\), (b) \(E_{pS}\) is the particle-surface energy and (c) \(E_{SS}\) the surface-surface energy, including self energy of both surfaces.

In the following, we consider only uniform surface charge density and using Eq.(A.1), one has

\[
\sigma = -\frac{Nq}{2L^2} \tag{A.3}
\]

With these notations, the particle-surface energy is given by

\[
E_{pS} = -\frac{Nq^2}{2L^2} \sum_{i=1}^{N} \int_{L_1 \cup L_2} dr \sum_{n} \frac{1}{|r_i - r + n|} \tag{A.4}
\]

Following the derivation done by Grzybowski and co-workers in ref.[15] and using the periodic boundary conditions, we have

\[
E_{pS}(\epsilon) = -\frac{Nq^2}{2L^2} \sqrt{\pi} \sum_{i=1}^{N} \left( f_\epsilon(|z_i - \frac{h}{2}|) + f_\epsilon(|z_i + \frac{h}{2}|) \right) \tag{A.5}
\]

where

\[
f_\epsilon(|Z|) = \int_{\epsilon}^{\infty} \frac{dt}{\sqrt{2\pi}} e^{-Z^2t} = 2\sqrt{\pi} \left[ e^{-\frac{Z^2}{\epsilon}} \text{erf}(\sqrt{\epsilon}|Z|) \right] - 2\sqrt{\pi} |Z| \tag{A.6}
\]
The prescription $\epsilon$ has been introduced to outline the diverging behavior of the integral in the second member of Eq.(A.6). As $\epsilon \to 0$, we have

$$f_\epsilon(|Z|) = \frac{2}{\sqrt{\epsilon}} - 2\sqrt{\pi} |Z| + 2\sqrt{\epsilon} |Z|^2 + o(\epsilon^{3/2})$$  \hspace{1cm} (A.7)

As extensively commented in the literature, because of the electroneutrality of the system, the diverging behavior as $\epsilon \to 0$, is cancelled by adding all contributions to the electrostatic energy (including self energy contributions). For systems such as all particles are confined between charged surfaces (i.e. $-h/2 \leq z_i \leq h/2$), as the system studied in this work and the system studied in ref.[25], we have

$$f_\epsilon(|z_i - h/2|) + f_\epsilon(|z_i + h/2|) = 4\sqrt{\epsilon} - \sqrt{\pi h} + 2\sqrt{\epsilon}(|z_i - h/2|^2 + |z_i + h/2|^2) + o(\epsilon^{3/2})$$  \hspace{1cm} (A.8)

Following the same steps, the surface-surface contribution is

$$E_{SS}(\epsilon) = \frac{N^2 q^2}{2L^2} \sqrt{\pi} \sqrt{\epsilon} + \frac{N^2 q^2}{4L^2} \sqrt{\pi} f_\epsilon(h)$$  \hspace{1cm} (A.9)

When contributions $E_{SS}(\epsilon)$ and $E_{pS}(\epsilon)$ are added, the diverging behavior is not yet cancelled by using electroneutrality since one has to include the contribution of the particle-particle interaction with the same prescription.

With the notations of subsection III.A, one has

$$E_{pp}(\epsilon) = E_R + E_{\kappa \neq 0} + E_{\kappa = 0}$$  \hspace{1cm} \hspace{1cm} (A.10)

Computing the integral in the right handed side of Eq.(A.10), one finds

$$E_{pp}(\epsilon) = E_R + E_{\kappa \neq 0} + E_{\kappa = 0}$$

$$+ 2q^2 \pi L^2 \sum_{i=1}^{N} \sum_{j=1, j \neq i}^{N} \left( \frac{e^{-z_{ij}^2}}{\sqrt{\epsilon \pi}} + |z_{ij}| \text{erf}(\sqrt{\epsilon} |z_{ij}|) \right) + E_{pp}^{Self}(\epsilon)$$  \hspace{1cm} (A.11)

The particle-particle contribution is then separated into the converging contribution given in Subsection III.A and a diverging contribution as

$$E_{pp}(\epsilon) = E_{pp}^{(cv)} + E_{pp}^{(dv)}(\epsilon)$$  \hspace{1cm} (A.12)

with

$$E_{pp}^{(dv)}(\epsilon) = \frac{N^2 q^2}{2L^2} \sqrt{\pi} \sqrt{\epsilon} + \frac{q^2}{L^2} \left( \sum_{i=1}^{N} \sum_{j=1, j \neq i}^{N} z_{ij}^2 \right) \sqrt{\epsilon} + o(\epsilon^{3/2})$$  \hspace{1cm} (A.13)

Thus, adding the contributions given by Eqs.(A.5), (A.9), (A.11) and (A.13), and letting $\epsilon \to 0$, Eq.(A.2) becomes

$$E = E_{pp}^{(cv)} + \frac{\pi}{2} \frac{N^2 q^2}{L^2} h = E_{pp}^{(cv)} + 2\pi \sigma^2 L^2 h = E_{pp}^{(cv)} + E_W$$  \hspace{1cm} (A.14)
The contributions of the charged surfaces to the energy when the particles are confined between surfaces is simply a term that is not depending on the position of the particles. During the sampling of the phase space of the system described in section II, especially when the energy difference for a trial move of a particle is computed, the term added in Eq.(A.14), $E_W$ is constant as long as $h$, $L$ and $N$ are constants. This constant is included in the results given in TABLE III and IV, and Figures 2(a-d). $E_W$ is considered as a contribution to interlayer energies.

On the other hand, $E_{pp}^{(cv)}$ is evaluated using the Ewald quasi-2D, Hautman-Klein or Lekner-cyclic methods.
References

[1] D. Frenkel and B. Smit, *Understanding molecular simulation. From Algorithms to Applications.* (Academic Press, 1996)

[2] M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids*, Clarendon Press (1987)

[3] J.C. Shelley and G.N. Patey, Mol. Phys. **88**, 385 (1996)

[4] (a) S.W. De Leeuw, J.W. Perram and E.R. Smith, Proc. R. Soc. Lond. **A373**, 27 (1980); *ibid.*, 57 (1980). Ewald sums have also been developed recently for Yukawa potentials, see: (b) G. Salin and J.-M. Caillol, J. Chem. Phys. **113**, 10459 (2000).

[5] H. Totsuji, Phys. Rev. **A17**, 399 (1978)

[6] G.T. Gao, X.C. Zeng and W.Wang, J. Chem. Phys. **106**, 3311 (1997)

[7] M. Deserno and C. Holm, J. Chem. Phys. **109**, 7678 (1998); 7694 (1998)

[8] L. Greengard and V. Rokhlin, J. Comp. Phys. **73**, 325 (1987); K.E. Schmidt and M.A. Lee, J. Stat. Phys. **63**, 1223 (1991)

[9] Y.-J. Rhee, J.W. Halley, J. Hautman and A. Rahman, Phys. Rev. **B40**, 36 (1989)

[10] D.E. Parry, Surf. Sci. **49**, 433 (1975); *ibid.*, **54**, 195 (1976)

[11] S.W. De Leeuw and J.W. Perram, Mol. Phys. **37**, 1313 (1979)

[12] B.R.A. Nijboer, Physica **A 125**, 275 (1984); B.R.A. Nijboer and F.W. de Wette, Physica **23**, 309 (1957)

[13] E.R. Smith, Mol. Phys. **65**, 1089 (1988)

[14] D.M. Heyes, Phys. Rev. **B49**, 755 (1994)

[15] A. Grzybowski, E. Gwóźdź, and A. Bródka Phys. Rev. **B61**, 6706 (2001)

[16] J. Hautman and M.L. Klein, Mol. Phys. **75**, 379 (1992)

[17] D. Wolf, P. Keblinski, S.R. Phillpot and J. Eggebrecht, J. Chem. Phys. **110**, 8254 (1999)

[18] A. Delville, R.J.-M. Pellenq and J.M. Caillol, J. Chem. Phys. **106**, 7275 (1997); R.J.-M. Pellenq, J.M. Caillol and A. Delville, J. Phys. Chem. **B101**, 8584 (1997); J.M. Caillol, J. Chem. Phys. **96**, 1455 (1992); J.M. Caillol and D. Levesque, J. Chem. Phys. **96**, 1477 (1992); J.M. Caillol, J. Chem. Phys. **99**, 8953 (1993)

[19] J. Lekner, Physica **A157**, 826 (1989); J. Lekner, Physica **A176**, 485 (1991)
[20] (a) I.-C. Yeh and M.L. Berkowitz, J. Chem. Phys. 110, 7935 (1999); (b) I.-C. Yeh and M.L. Berkowitz, J. Chem. Phys. 111, 3155 (1999)

[21] (a) A. Arnold and C. Holm, Chem. Phys. Lett. 354, 324 (2002); (b) A. Arnold, J. de Joannis and C. Holm, J. Chem. Phys. 117, 2496 (2002); 2503 (2002)

[22] M. Kawata and M. Mikami, Chem. Phys. Lett. 340, 157 (2001); M. Kawata, M. Mikami and U. Nagashima, J. Chem. Phys. 115, 4457 (2001); M. Mazars, J. Chem. Phys. 117, 3524 (2002); M. Kawata, M. Mikami and U. Nagashima, J. Chem. Phys. 117, 3526 (2002); A. Grzybowski and A. Bródka, Chem. Phys. Lett. 361, 329 (2002)

[23] M. Kawata, M. Mikami and U. Nagashima, J. Chem. Phys. 116, 3430 (2002)

[24] M. Mazars, J. Chem. Phys. 115, 2955 (2001)

[25] M. Mazars, J.-M. Caillol, J.-J. Weis and D. Levesque, Condensed Matter Physics, 4, 697 (2001) (http://www.icmp.lviv.ua/journal/)

[26] A.H. Widmann and D.B. Adolf, Comp. Phys. Comm. 107, 167 (1997); in this paper the Ewald quasi-2D method is called HBC for D.M. Heyes, M. Barder and J.H.R. Clarke, J. Chem. Soc. Faraday Trans. II 73, 1485 (1977).

[27] T.B. Mitchell, J.J. Bollinger, D.H.E. Dubin, X.-P. Huang, W.M. Itano and B.M. Baughman, Science 283, 1290 (1998)

[28] S. Leikin, J. Chem. Phys. 95, 5224 (1991)

[29] A.W.C. Lau, D. Levine and P. Pincus, Phys. Rev. Lett. 84, 4116 (2000); A.W.C. Lau, P. Pincus, D. Levine and H.A. Fertig, Phys. Rev. E63, 051604 (2001); A.Yu. Grosberg, T.T. Nguyen and B.I. Shklovskii, Rev. Mod. Phys. 74, 329 (2002)

[30] R.S. Crandall and R. Williams, Phys. Lett. A34, 404 (1971); C.G. Grimes and G. Adams, Phys. Rev. Lett.42, 795 (1979)

[31] G. Goldoni and F.M. Peeters, Phys. Rev. B53, 4591 (1996)

[32] J.-J. Weis, D. Levesque and S. Jorge, Phys. Rev. B63, 045308 (2001)

[33] N. Metropolis, A.W. Rosenbluth, M.N. Rosenbluth, A.H. Teller, et E. Teller, J. Chem. Phys. 21, 1087 (1953)

[34] J.-J. Weis, D. Levesque and J.-M. Caillol, J. Chem. Phys 109, 7486 (1998)

[35] E. Spoehr, J. Chem. Phys. 107, 6342 (1997)

[36] B. Cichocki and B.U. Felderhof, Mol. Phys. 67, 1373 (1989)

[37] C.M. Linton, Proc. R. Soc. Lond. A455, 1767 (1999)
[38] R.E. Johnson and S. Raganathan, Phys. Rev. E63, 056703 (2001)
[39] S.W. de Leeuw and J.W. Perram, Fast Ion transport in solids, Electrode and Electrolytes, edited by P. Vashista, J.N. Mundy and G.K. Shenoy (North-Holland, 1982)
[40] A. Arnold and C. Holm, Comp. Phys. Comm. 148, 327 (2002)
[41] K.-P. Schneider and S. Eisenblätter, Chem. Phys. Lett. 303, 325 (1999)
[42] M. Porto, J. Phys. A: Math. Gen. 33, 6211 (2000)
[43] A. Grzybowski and A. Bródka, Mol. Phys. 100, 635 (2002)
[44] B.R.A. Nijboer and F.W. de Wette, Physica 24, 1105 (1958); 1422 (1958)
[45] R. Sperb, Mol. Simul. 22, 199 (1999); R. Sperb and R. Strebel, Mol. Simul. 27, 61 (2001)
[46] N. Grønbech-Jensen, Int. J. Mod. Phys. C7, 873 (1996)
[47] N. Grønbech-Jensen, Int. J. Mod. Phys. C8, 1287 (1997)
[48] N. Grønbech-Jensen, G. Hummer and K.M. Beardmore, Mol. Phys. 92, 941 (1997)
[49] N. Grønbech-Jensen, Comp. Phys. Com. 119, 115 (1999)
[50] S.L. Marshall, J. Phys.: Condens. Matter 12, 4575 (2000)
[51] A.T. Clark, T.J. Madden and P.B. Warren, Mol. Phys. 87, 1063 (1996)
[52] S.Y. Liem and J.H.R. Clarke, Mol. Phys. 92, 19 (1997)
[53] A. Grzybowski and A. Bródka, Mol. Phys. 100, 1017 (2002)
[54] R.D. Mountain, J. Phys. Chem. B 105, 6556 (2001)
[55] T. Terao and T. Nakayama, Phys. Rev. E63, 041401 (2001); 65, 021405 (2002)
[56] T. Terao and T. Nakayama, private communication.
[57] J.F. Harper, Physica A196, 255 (1993)
[58] R. Sperb, Mol. Simul. 13, 189 (1994); 20, 179 (1998)
[59] F.S. Gsajka and C. Siedel, Macromolecules, 33, 2728 (2000)
[60] A.G. Moreira and R.R. Netz, Europhys. Lett. 52, 705 (2000); Phys. Rev. Lett. 87, 078301 (2001); Europhys. Lett. 57, 911 (2002)
[61] A.G. Moreira and R.R. Netz, Eur. Phys. J. E8, 33 (2002)
[62] J.N. Newman, *Approximation for free surface Green function*. In: *Wave asymptotics* (ed. P.A. Martin and G.R. Wicklam), pp.107-135, Cambridge University Press (1992).

[63] N. Grønbech-Jensen, R.J. Mashl, R.F. Bruinsma and W.M. Gelbart, Phys. Rev. Lett. 78, 2477 (1997); R.J. Mashl and N. Grønbech-Jensen, J. Chem. Phys. 109, 4617 (1998); R.J. Mashl, N. Grønbech-Jensen, M.R. Fitzsimmons, M. Lütt and D.Q. Li, J. Chem. Phys. 110, 2219 (1999)

[64] B.P. Stojković, Z.G. Yu, A.R. Bishop, A.H. Castro Neto and N. Grønbech-Jensen, Phys. Rev. Lett. 82, 4679 (1999); B.P. Stojković, Z.G. Yu, A.L. Chernyshev, A.R. Bishop, A.H. Castro Neto and N. Grønbech-Jensen, Phys. Rev. B62, 4353 (2000)

[65] N. Grønbech-Jensen, K.M. Beardmore and P. Pincus, Physica A261, 74 (1998)

[66] A.H. Juffer, G.M. Shepherd and H.J. Vogel, J. Chem. Phys. 114, 1892 (2001)

[67] T. Terao, Phys. Rev. E66, 046707 (2002)

[68] A very interesting exception may be found in: *Molecular Dynamics and Time Reversibility* by D. Levesque and L. Verlet, J.Stat. Phys. 72, 519 (1993)

[69] D.J. Tildesley in *Computer Simulation in Chemical Physics*, p.23 edited by M.P. Allen and D.J. Tildesley (Kluwer Academic Publishers) (1993)

[70] J. Lekner, Physica B252, 149 (1998)
List of Tables

**TABLE I:** Geometrical and simulation parameters of the different Monte Carlo samplings. $L$ is the spatial periodicity, $h$ the distance between the layers, $N$ the number of point ions in the simulation, $\sigma$ the uniform surface charge density used to achieve electroneutrality, $t_{eq}$ the number of Monte Carlo cycles done in each Runs for equilibration, $t_{av}$ the number of Monte Carlo cycles to accumulate thermodynamical average, $\beta u_0$ the Mandelung energy computed for a square bidimensional lattice. The preferential structure is the structure found in the Monte carlo study of the solid-phase diagram of the classical bilayer crystal done in ref.[31].

**TABLE II:** Estimation of the accuracy of the sampling of the phase space by the three different methods. $\beta \Delta E_{pp}^{(cv)}$ is the energy difference computed with the three methods of the first trial move of the first MC-cycle from the same initial conditions and with the same random numbers. ACC($t = 1$) is the number of accepted trial moves after the first MC-cycle and ACC($t = 100$) the number of accepted trial moves after the first hundred MC-cycles (see also Figures 2).

**TABLE III:** Averages energies computed with the Monte Carlo algorithm for Runs a and b ($h = 1.0$). $\beta U/N$ is the average total energy per particle, $\beta E_{inter}/N$ and $\beta E_{intra}/N$ are respectively the average interlayer and intralayer energy per particle, $\beta \sigma_U/N$ an estimation of the statistical fluctuation $\sigma_U$ of the total energy and $\delta t$ is a crude estimation of the average CPU-time in seconds per MC-cycle ($\beta = 1/kT, T$ temperature).

**TABLE IV:** Same as TABLE III, but for Runs c, d, e, e’ and f ($h = 4.0$).
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Figure 1: Representation of the bilayer Wigner cristal. Periodic boundary conditions with a spatial periodicity $L$ are not represented. This representation is a snapshot of an instantaneous configuration of a system of 512 point ions with the geometrical parameters of the Runs e and e' given in TABLE I.

Figure 2: Instantaneous energy per particle for the first 500 Monte-Carlo cycles computed with different methods but with exactly the same initial configuration and with the same sequence of random numbers. The parameters of these computations are: $h = 1.0$, $N = 512$, $L = 28.36$ (Run a) (see also TABLE II). (a) Comparison of the sampling done with the Ewald quasi-2D and Hautman-Klein method. (b) Comparison between the Ewald quasi-2D and Lekner-cyclic ($n_c = 10$, $n_K = 3$) methods.

Figure 2: (c) Comparison between the Ewald quasi-2D and Lekner-cyclic ($n_c = 25$, $n_K = 3$) methods. (d) Comparison between the Ewald quasi-2D and Lekner-cyclic ($n_c = 50$, $n_K = 3$) methods.

Figure 3: Representation of the intralayer $g_{11}(s)$ and interlayer $g_{12}(s)$ pair distributions functions obtained with the Ewald quasi-2D and Hautman Klein methods for Runs a and b ($h = 1.0$). (a) $g_{11}(s)$ from Runs a; (b) $g_{12}(s)$ from Runs a; (c) $g_{11}(s)$ from Runs b; (d) $g_{12}(s)$ from Runs b.

Figure 4: Same as the Figure 3, but for the Ewald quasi-2D and Lekner-cyclic ($n_c = 10$, $n_K = 3$) methods for Runs a and b ($h = 1.0$). (a) $g_{11}(s)$ from Runs a; (b) $g_{12}(s)$ from Runs a; (c) $g_{11}(s)$ from Runs b; (d) $g_{12}(s)$ from Runs b.

Figure 5: Same as the Figure 3, but for the Ewald quasi-2D and Lekner-cyclic ($n_c = 25$, $n_K = 3$) methods for Runs a and b ($h = 1.0$). (a) $g_{11}(s)$ from Runs a; (b) $g_{12}(s)$ from Runs a; (c) $g_{11}(s)$ from Runs b; (d) $g_{12}(s)$ from Runs b.

Figure 6: Same as the Figure 3, but for the Ewald quasi-2D and Lekner-cyclic ($n_c = 40$, $n_K = 2$) methods for Runs a and b ($h = 1.0$). (a) $g_{11}(s)$ from Runs a; (b) $g_{12}(s)$ from Runs a; (c) $g_{11}(s)$ from Runs b; (d) $g_{12}(s)$ from Runs b.
Figure 7: Same as Figure 3, but for the Ewald quasi-2D and Hautman Klein methods for Runs c to f ($h = 4.0$). (a) $g_{11}(s)$ from Runs c; (b) $g_{12}(s)$ from Runs c ($N = 128$); (c) $g_{11}(s)$ from Runs d; (d) $g_{12}(s)$ from Runs d ($N = 338$); (e) $g_{11}(s)$ from Runs e and e'; (f) $g_{12}(s)$ from Runs e and e' ($N = 512$); (g) $g_{11}(s)$ from Runs f; (h) $g_{12}(s)$ from Runs f ($N = 968$).

Figure 8: Same as Figure 3, but for the Ewald quasi-2D and Lekner-cyclic ($n_c = 10, n_K = 3$) methods for Runs c to f' ($h = 4.0$). (a) $g_{11}(s)$ from Runs c; (b) $g_{12}(s)$ from Runs c ($N = 128$); (c) $g_{11}(s)$ from Runs d; (d) $g_{12}(s)$ from Runs d ($N = 338$); (e) $g_{11}(s)$ from Runs e and e'; (f) $g_{12}(s)$ from Runs e and e' ($N = 512$); (g) $g_{11}(s)$ from Runs f; (h) $g_{12}(s)$ from Runs f ($N = 968$).

Figure 9: Same as Figure 3, but for the Ewald quasi-2D and Lekner-cyclic ($n_c = 25, n_K = 3$) methods for Runs c to f' ($h = 4.0$). (a) $g_{11}(s)$ from Runs c; (b) $g_{12}(s)$ from Runs c ($N = 128$); (c) $g_{11}(s)$ from Runs d; (d) $g_{12}(s)$ from Runs d ($N = 338$); (e) $g_{11}(s)$ from Runs e and e'; (f) $g_{12}(s)$ from Runs e and e' ($N = 512$); (g) $g_{11}(s)$ from Runs f; (h) $g_{12}(s)$ from Runs f ($N = 968$).
Method Parameters of the method $\beta$ $\Delta E_{PP}^{(cv)}$ $\text{ACC}$ $\text{ACC}$

| Method               | Parameters | $\beta$ $\Delta E_{PP}^{(cv)}$ | $(t = 1)$ | $(t = 100)$ |
|----------------------|------------|-------------------------------|-----------|-------------|
| Ewald quasi-2D       | $\alpha = 0.3$, $\kappa \times \kappa = 8 \times 8$ | $-0.55046(0)$ | 243       | 23139       |
| Hautman-Klein        | $\alpha = 0.3$, $\kappa \times \kappa = 8 \times 8$ |                          |           |             |
|                      | $M = 1$    | $-0.5506(7)$                  | 243       | 23171       |
|                      | $M = 2$    | $-0.5506(1)$                  | 243       | 23171       |
|                      | $M = 3$    | $-0.5506(2)$                  | 243       | 23171       |
| Lekner-cyclic        | $n_c = 5$, $n_K = 1$ | $-1.37408(7)$                | 348       | 24623       |
|                      | $n_c = 5$, $n_K = 2$ | $-1.37404(2)$                | 348       | 24623       |
|                      | $n_c = 5$, $n_K = 3$ | $-1.37404(1)$                | 348       | 24623       |
|                      | $n_c = 5$, $n_K = 4$ | $-1.37404(1)$                | 348       | 24623       |
|                      | $n_c = 10$, $n_K = 1$ | $-0.77481(0)$                | 263       | 24775       |
|                      | $n_c = 10$, $n_K = 2$ | $-0.77476(4)$                | 263       | 24775       |
|                      | $n_c = 10$, $n_K = 3$ | $-0.77476(4)$                | 263       | 24775       |
|                      | $n_c = 10$, $n_K = 4$ | $-0.77476(4)$                | 263       | 24775       |
|                      | $n_c = 15$, $n_K = 1$ | $-0.51255(9)$                | 244       | 23601       |
|                      | $n_c = 15$, $n_K = 2$ | $-0.51251(3)$                | 244       | 23597       |
|                      | $n_c = 15$, $n_K = 3$ | $-0.51251(3)$                | 244       | 23597       |
|                      | $n_c = 15$, $n_K = 4$ | $-0.51251(3)$                | 244       | 23597       |
|                      | $n_c = 25$, $n_K = 1$ | $-0.5440(5)$                 | 243       | 23132       |
|                      | $n_c = 25$, $n_K = 2$ | $-0.54493(9)$                | 243       | 23132       |
|                      | $n_c = 25$, $n_K = 3$ | $-0.54493(9)$                | 243       | 23132       |
|                      | $n_c = 40$, $n_K = 1$ | $-0.5509(7)$                 | 243       | 23172       |
|                      | $n_c = 40$, $n_K = 2$ | $-0.55089(1)$                | 243       | 23147       |
|                      | $n_c = 40$, $n_K = 3$ | $-0.55089(1)$                | 243       | 23147       |
|                      | $n_c = 45$, $n_K = 1$ | $-0.5507(5)$                 | 243       | 23172       |
|                      | $n_c = 45$, $n_K = 2$ | $-0.55071(1)$                | 243       | 23172       |
|                      | $n_c = 45$, $n_K = 3$ | $-0.55071(2)$                | 243       | 23172       |

**TABLE II.** MAZARS, Lekner and Ewald sums.
| Name | $L$ | $h$ | $N$ | $\sigma$ | $t_{eq}$ | $t_{moy}$ | $\beta u_0$ | Preferential structure         |
|------|-----|-----|-----|--------|--------|--------|----------|-----------------------|
| a    | 28.36 | 1.0 | 512 | -4.456 | 50000  | 100000 | -215.64794 | staggered rhombic       |
| b    | 56.72 | 1.0 | 512 | -1.114 | 50000  | 100000 | -106.68091 | fluid-like, disordered  |
| c    | 14.18 | 4.0 | 128 | -4.456 | 400000 | 100000 | -215.64794 | staggered hexagonal     |
| d    | 23.04 | 4.0 | 338 | -4.456 | 100000 | 200000 | -215.64794 | staggered hexagonal     |
| e    | 28.36 | 4.0 | 512 | -4.456 | 50000  | 100000 | -215.64794 | staggered hexagonal     |
| e'   | 28.36 | 4.0 | 512 | -4.456 | 200000 | 100000 | -215.64794 | staggered hexagonal     |
| f    | 38.99 | 4.0 | 968 | -4.456 | 50000  | 100000 | -215.64794 | staggered hexagonal     |

**TABLE I.** MAZARS, Lekner and Ewald sums.
| Method                  | Runs | L  | $\beta U/N$ | $\beta E_{\text{inter}}/N$ | $\beta E_{\text{intra}}/N$ | $\beta \sigma_U/N$ | $\delta t(s)$ |
|-------------------------|------|----|-------------|-----------------------------|-----------------------------|---------------------|--------------|
| Ewald quasi-2d          | a    | 28.36 | -219.94 | -5.06(9) | -214.87(9) | 0.046 | 1.0 |
|                         | b    | 56.72 | -119.22 | -12.85(7) | -106.36(1) | 0.049 | 1.0 |
| Hautman-Klein           | a    | 28.36 | -219.95 | -5.06(7) | -214.88(3) | 0.045 | 1.3 |
|                         | b    | 56.72 | -119.20 | -12.76(3) | -106.44(1) | 0.046 | 1.3 |
| Lekner-cyclic           | a    | 28.36 | -219.75 | -4.97(9) | -214.79(1) | 0.045 | 3.9 |
| ($n_c = 10$, $n_K = 3$) | b    | 56.72 | -119.51 | -14.54(3) | -104.96(5) | 0.052 | 3.9 |
| Lekner-cyclic           | a    | 28.36 | -219.96 | -5.07(8) | -214.88(8) | 0.046 | 4.2 |
| ($n_c = 25$, $n_K = 3$) | b    | 56.72 | -119.30 | -12.73(5) | -106.56(6) | 0.047 | 4.2 |
| Lekner-cyclic           | a    | 28.36 | -219.95 | -5.06(6) | -214.88(4) | 0.045 | 3.8 |
| ($n_c = 40$, $n_K = 2$) | b    | 56.72 | -119.21 | -12.91(8) | -106.29(2) | 0.047 | 3.8 |

**TABLE III.** MAZARS, Lekner and Ewald sums.
| Method                  | Runs  | N  | $\beta U/N$ | $\beta E_{\text{inter}}/N$ | $\beta E_{\text{intra}}/N$ | $\beta \sigma U/N$ | $\delta t$(s) |
|------------------------|-------|----|-------------|-----------------|-----------------|-----------------|-------------|
| Ewald quasi-2d         | c     | 128| -215.57     | -0.03(3)        | -215.54(0)      | 0.101           | 0.1          |
|                        | d     | 338| -215.61     | -0.00(4)        | -215.60(5)      | 0.057           | 0.5          |
|                        | e'    | 512| -215.67     | -0.00(5)        | -215.66(2)      | 0.046           | 1.1          |
|                        | f     | 968| -215.72     | -0.00(6)        | -215.70(9)      | 0.034           | 3.1          |
| Hautman-Klein          | c     | 128| -215.91     | -0.22(6)        | -215.68(1)      | 0.099           | 0.1          |
|                        | d     | 338| -215.65     | -0.00(6)        | -215.64(2)      | 0.057           | 0.6          |
|                        | e'    | 512| -215.68     | -0.00(7)        | -215.67(2)      | 0.046           | 1.2          |
|                        | f     | 968| -215.70     | -0.01(0)        | -215.68(8)      | 0.037           | 4.7          |
| Lekner-cyclic          | c     | 128| -215.42     | -0.00(2)        | -215.41(9)      | 0.100           | 0.3          |
| ($n_c = 10, n_K = 3$)  | d     | 338| -215.39     | -0.00(4)        | -215.38(2)      | 0.061           | 1.7          |
|                        | e     | 512| -215.86     | -0.00(4)        | -215.85(6)      | 0.048           | 3.7          |
|                        | f     | 968| -216.27     | -0.01(2)        | -216.25(6)      | 0.045           | 13.5         |
| Lekner-cyclic          | c     | 128| -215.39     | -0.00(2)        | -215.38(9)      | 0.101           | 0.3          |
| ($n_c = 25, n_K = 3$)  | d     | 338| -215.61     | -0.00(3)        | -215.60(4)      | 0.057           | 1.8          |
|                        | e     | 512| -215.67     | -0.00(4)        | -215.66(9)      | 0.046           | 4.0          |
|                        | f     | 968| -215.77     | -0.00(5)        | -215.76(8)      | 0.034           | 14.6         |

**TABLE IV**, MAZARS, Lekner and Ewald sums.
Figure 1, MAZARS, Lekner and Ewald sums.
Figure 2(a), (b), MAZARS, Lekner and Ewald sums.
Figure 2(c),(d), MAZARS, Lekner and Ewald sums.
Figure 3 (a-d), MAZARS, Lekner and Ewald sums.
Figure 4 (a-d), MAZARS, Lekner and Ewald sums.
Figure 5 (a-d), MAZARS, Lekner and Ewald sums.
Figure 6 (a-d), MAZARS, Lekner and Ewald sums.
Figure 7 (a-h), MAZARS, Lekner and Ewald sums.
Figure 8 (a-h): MAZARS, Lekner and Ewald sums.
Figure 9 (a-h), MAZARS, Lekner and Ewald sums.

Ewald quasi-2D

Lekner-cyclic

\(n_c=25, n_K=3\)