RESEARCH ARTICLE

Ewald method for polytropic potentials in arbitrary dimensionality

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The Ewald summation technique is generalized to power-law $1/|r|^k$ potentials in three-, two- and one-dimensional geometries with explicit formulae for all the components of the sums. The cases of short-range, long-range and 'marginal' interactions are treated separately. The jellium model, as a particular case of a charge-neutral system, is discussed and the explicit forms of the Ewald sums for such a system are presented. A generalized form of the Ewald sums for a non-cubic (non-square) simulation cell for three- (two-) dimensional geometry is obtained and its possible field of application is discussed. A procedure for the optimization of the involved parameters in actual simulations is developed and an example of its application is presented.

Keywords: Ewald summation; power-law potentials

1. Introduction

The behaviour of many-body systems is often governed by the long-range Coulomb potential between charged particles. Numerical simulations of such systems are usually performed by considering a finite number of particles in a cell with periodic boundary conditions. The correct estimation of the potential energy in such systems requires of a summation over all images created by the periodic boundary conditions. For long-range interaction potentials such direct summation either converges slowly or it is conditionally convergent, making its evaluation computationally cumbersome. Instead, the performance of the calculation can be greatly improved by using Ewald summation methods [1]. In these methods, the slowly convergent tail of the sum in the potential energy is represented by a rapidly convergent sum in momentum space.

For a good performance in simulations of large $N$-particle systems, a number of modified summation methods has been developed. Historically, the first efforts to enhance the Ewald method consisted in looking for appropriate truncation schemes, but all of them were strongly dependent on the system properties, in particular on the system size. Tabulations of precalculated terms in both real space and momentum space sums [4], as well as polynomial approximations of the involved functions [5–7], were also proposed to look for a balance between calculation time and truncation errors. Nevertheless, these approximate methods suffer from error accumulation in simulations of large systems, and do not allow for reducing the overall $O(N^2)$ complexity of the original Ewald summation. The work of Perram et al. [8] was the first to give a way to optimize the splitting of the interparticle potential between the long-range and short-range parts to yield a total complexity of $O(N^{3/2})$. A special modification of the Ewald method called Wolf summation [9,10], based on a damping of the Fourier-transformed part of the sum, was posteriorly developed in order to render the original Ewald summation more efficient for non-periodic systems and large model sizes. The Ewald technique was also applied to develop the method of evaluation of electrostatic potential near the surfaces of ionic crystals [11,12].

Another way for improving the Ewald method is to perform fast Fourier transform (FFT) of a reciprocal
space sum on a mesh. The oldest algorithm of this kind is the so-called Particle–Particle Particle–Mesh (P3M) method, invented and improved to the complexity $O(N)$ in calculation of forces by Hockney and co-authors in the 1970s [13,14]. The P3M technique is based on a distribution of the charge density on a grid using a certain smooth assignment function and then the discrete Poisson equation is solved using FFT. This algorithm appeared to be less complex to yield $O(N \ln N)$ with an appropriate choice of the free parameters. The P3M algorithm was recently improved by Ballenegger et al. [15] for calculation of energies, bringing, as claimed, the maximal precision in the energy by an optimization of the ‘influence’ function (a substitution of the potential in the Fourier-transformed Poisson’s equation). For a comprehensive introduction to Ewald- and mesh-based techniques we recommend the reader refer to the cited work of Ballenegger and co-authors, where special attention is paid to the estimation of both sum truncation-imposed and grid-imposed errors. The extension of this method, called Particle Mesh Ewald (PME), makes use of the analytical form of the sum in the reciprocal space and evaluates potentials via FFT instead of interpolating them as P3M does. Although PME is slightly more complex than the P3M algorithm, it still is $O(N \ln N)$ and allows one to reduce significantly the memory expenses. Later the Particle Mesh Ewald method was reformulated by Essmann et al. [17], making use of cardinal B-splines to interpolate structure factors. This approach, called Smooth Particle–Mesh Ewald (SPME) substantially improved the accuracy of PME with a comparable computational cost, as it still scales as $O(N \ln N)$. SPME is also claimed to be applicable to potentials of the polytropic form $1/|r|^k$. In general, the conventional FFT-based approaches suffer from the severe fallback of requiring equidistant particle positions. The invention of the variant of Fourier transform for nonequispaced nodes (NFFT) opened a path to overcome this shortcoming, while keeping the introduced errors below the specified target levels. The nonequispaced fast Fourier transform is currently considered as a promising means to improve the Ewald summation performance, with open code implementations available [18]. The early variants of the NFFT algorithms are reviewed in the work of Ware [19]; a general approach to the fast summation methods based on NFFT can be found in the article of Steidl [20].

The most recent family of algorithms based on the Ewald approach are the tree-based algorithms, with the fast multipole method (FMM) being the most known and widely used among them. The algorithm, developed primarily by Greengard and Rokhlin [21], is based on the idea of keeping the direct summation of potentials or forces for the nearby atoms and approximating the interactions of the distant atoms by their multipole expansions. FMM offers the asymptotically fastest performance among the Ewald-related algorithms, being linear in $N$ in most cases and not worse than $O(N \ln N)$ with explicitly controlled accuracy. The FMM technique is naturally applicable to inhomogeneous and non-periodic systems, being also easy to parallelize since it is an entirely real-space summation. Since then the algorithm has been significantly improved in efficiency, mostly by introducing new diagonal forms of translation operators [22]. However, FMM has an intrinsic shortcoming, when applied to molecular dynamics calculations, as the energy conservation it brings is poor; the method per se is also rather cumbersome in implementation. Another group of methods, based on the multigrid methods of solving elliptic (in this particular case – Poisson’s) equations [23], was developed a decade ago [24]. These methods allow one to preserve the scaling $O(N)$ and parallelization advantages of tree-based methods, as well as the applicability in simulations without PBC, being on the other hand satisfactorily energy-conserving and additionally accelerated on all length scales. An efficient realization of the multigrid method and its analysis may be found in the work of Sagui et al. [25]. An advanced mesh Ewald technique, claimed to reduce significantly the computational costs of charge spreading in multigrid-based methods, was recently proposed by Shan et al. [26].

A detailed comparison of the optimized $O(N^{3/2})$ pure Ewald technique, FFT-based summations, and multipole-based methods was made by Petersen [27] for systems with approximately uniform charge distributions, taking into account a possible parallel implementation. According to Petersen, the method of choice with a number of particles below $10^4$ is the conventional Ewald summation, PME is preferable in the range $N \sim 10^4$–$10^5$, and the fast multipole method should overperform them with $N > 10^5$. A more recent and ample review of FMM, P3M and pure Ewald methods by Pollock and Glosli [28], based partially on their own calculations, implies that P3M is faster than the Ewald summation already for 500 particles, although it is stressed that the other factors such as the ease of the coding, the system geometry, as well as the code optimization can change the choice. We would also suggest a thorough survey of different Ewald summation techniques given in the work of Toukmaji and Board [29].
An approach, alternative to using cubic periodic boundary conditions in a calculation of long-range interactions, called Isotropic Periodic Sum (IPS), was recently proposed by Wu and Brooks [30]. The main goal of their approach is to deal with long-range interactions, avoiding artificial correlations and anisotropy bias induced by a PBC-based summation in a cubic box. In this technique, only the interactions of a particle $A$ with the others within a certain radius $r_c$ are taken into account (as in a plain cut-off scheme), and this spherical simulation zone is repeated in an infinite number of shifts by vectors $r_{sh}$, such that $|r_{sh}| = 2N|r_c|$. Therefore, the particle $A$ interacts not only with $B$ (within the sphere radius), but also with all the images of $B$, occupying homogeneously the shells of radii $2N|r_c|$, centred in $B$. The subsequent integration and summation over the shells allows one to obtain explicit expressions of forces and energies for a number of interactions of most physical interest, like electrostatic, Lennard-Jones and exponential potentials. The method is known to yield a performance close to the one shown by the Ewald summation, but without imposing unwanted symmetry effects.

Since its proposal, the Ewald method has been applied to a large number of physical problems, although mostly to systems with the Coulomb $1/|r|$ interaction potential. In a recent work by Johnson and Ranganathan [31], a generalized approach to Ewald summation is stated to obtain potential energy and forces for systems with a power-law, Yukawa potential and electronic bilayer systems. The Ewald method for two-dimensional systems with electrostatic interactions was developed by Parry [11], but his technique appeared to be computationally inefficient. Spohr [32] studied a slab geometry by treating the simulation cell as a fully three-dimensional one with the conventional Ewald summation. Later on, a significant advance was made by Yeh and Berkowitz [33], as these authors managed to obtain the explicit correction term for the rigorous three-dimensional Ewald summation, which brings the results for a slab system to a satisfactory agreement with the 2D summation. The 2D Ewald technique was also applied by Yang et al. [34] to calculate the energy of Coulomb particles in a slab system with a uniformly charged surface. One of the first two-dimensional variants of the Ewald summation was presented in [35], applied to the quasi-2D Stockmayer model with the potential $1/|r|^3$. Recent applications to dipolar bosons in a 2D geometry have been made by Mora et al. [36] and Lu and Wu [37]. On the other hand, the explicit forms of the Ewald sums for Yukawa interactions have also been reported: in 3D geometry, with partial periodic boundary conditions [38,39], and in 2D geometry [40]. The general approach to the Ewald summation in quasi-two-dimensional systems with power-law potentials and results for several values of power factor are given in [41]. The Ewald method can also be useful even applied to fast decaying power-law potentials. For instance, the Ewald formalism was developed in [42] for the dispersion interaction $1/|r|^6$ and later for the Lennard-Jones potential by Ou-Yang et al. [43]. Also, Shirts et al. in their recent work [44] argue the need for taking into account the effects of cut-offs in molecular dispersion interactions due to a Lennard-Jones potential, especially in non-isotropic and inhomogeneous media. The authors developed two formalisms for the estimation of these cut-off errors in binding free energy of macromolecular systems, which can in principle be extended to the other observables. However, it is claimed that the adequate implementation of the Ewald summation for this kind of system may render their corrections unnecessary by mostly eliminating the cut-off-dependent behaviour.

In the present work, we report explicit expressions of the Ewald sums for the general case of particles interacting via a $1/|r|^k$ polytropic potential and in 3D, 2D, and 1D geometries. The closed derivation of these sums is given, with special attention being paid to conditionally convergent potentials. One of the difficulties of the derivation is that different terms have to be considered in the cases of short-range, long-range or ‘marginal’ potentials. In the case of a short-range interaction, the original slowly convergent sum is represented as a linear combination of two rapidly convergent ones. For a long-range interaction, the condition of charge neutrality in the simulation cell is shown to be necessary to make the energy absolutely convergent within the considered scheme. The introduction of a uniform neutralizing charged background (jellium), as a particular case of a charge-neutral system, is also discussed. The explicit forms of the Ewald sums are reported for a jellium system and for an arbitrary polytropic potential. We explicitly calculate the expressions for physically relevant interactions as Coulomb, dipole–dipole, and Lennard-Jones potentials. Finally, we have extended the Ewald sums to the case of a noncubic simulation cell, which could be useful in simulations of hexagonal closed packed (hcp) and two-dimensional triangular solids. In addition, the general derivation path given in this work may be used to obtain the forms of Ewald sums for other interaction potentials.
The computational efficiency is another important issue of the practical implementation of the method. In fact, one needs to choose correctly a free parameter, appearing in the integral representation of the sums, and to decide which number of terms should be kept in spatial and momentum sums in order to reach the required accuracy. The choice of these three parameters affects the difference between the calculated result and the exact one as well as the calculation complexity. Therefore, a certain optimization of the parameters is always required. In the present work, this optimization process is formalized and it is shown that in following the described procedure the overall computation time is significantly reduced. The accuracy of the result is shown to be kept under control, with the only cost of a preliminary benchmark calculation.

The rest of the article is organized as follows. In Section 2, we formulate the problem, develop the general Ewald approach and report explicit expressions for the Ewald sums for a polytropic potential in a three-dimensional cubic simulation cell. Sections 3 and 4 contain derivations of the Ewald sums in two-dimensional and one-dimensional geometries, respectively. In Section 5, the case of a simulation cell with different side lengths is considered for three- and one-dimensional geometries, respectively. In Section 6, the case of a simulation cell with one-dimensional geometries, respectively. In Section 7, the case of a simulation cell with different side lengths is considered for three- and two-dimensional geometries. The final general expressions and their particularization to the most physically relevant cases are presented in Section 6. The practical algorithm for the parameter optimization and an actual application of the Ewald method is discussed in Section 7. Summary and conclusions are drawn in Section 8.

2. Ewald sum for an arbitrary polytropic potential

$1/|r|^k$ in 3D geometry

2.1. Basic assumptions and initial sums

We consider a system of $N$ particles inside a cubic simulation cell of size $L$ with periodic boundary conditions. Thus, each particle with coordinates $r$ in the initial cell has an infinite number of images $r + nL$ in the adjacent cells. The total potential energy is estimated by

$$U = \frac{1}{2} \sum_{n \in \mathbb{Z}^3} \left[ \sum_{i=1}^N \sum_{j=1}^N \phi(r_{ij} + nL) \right], \quad (1)$$

where $\phi(r)$ is the interparticle potential, $r_{ij} \equiv r_i - r_j$, and the prime in the first sum means that the summation over an integer vector $n$ must be done omitting the term $n = 0$ when $i = j$.

2.2. Analytic development

In many physical situations, the interaction potential between two particles $i$ and $j$ has the power-law form $q_i q_j / |r|^{k}$ with positive $k$ and $q_i$, $q_j$ being the generalized charges of the particles. This sort of interaction is generally referred to as polytropic potential.

First, let us consider the case of short-range potentials, $k \leq 3$. As we will see later, the potentials corresponding to $k > 3$ give a similar result. For $k \leq 3$, the right-hand part of Equation (1) diverges and it can be made convergent only if the restriction of charge neutrality is required, i.e. when $\sum_{i=1}^N q_i = 0$. It has also been shown [45] that for a pure electrostatic interaction the total energy (1) can be conditionally convergent even in a neutral simulation cell because of a higher multipole contribution. The energy and forces are therefore dependent on the order of summation, which can also be implicitly set by a choice of a convergence factor. The ambiguity usually appears in the form of a constant or a position-dependent term, vanishing in the limit $L \rightarrow \infty$. Hence, the preference in one or another factor should be dictated either by physical properties of a particular system or by arguments regarding rates of convergence to the thermodynamic limit. For a general discussion on the convergence issues appearing in periodic boundary conditions, see [46]. The main idea of the Ewald summation technique in the approach proposed by de Leeuw et al. [2] is to multiply each component of the sum by the dimensionless factor $\exp(-sr^2)$, with $s > 0$ being a dimensionless regularizing parameter, making the sum absolutely convergent. Then, the limit $s \rightarrow 0$ is taken, so that the singularity in the initial sum (1) can be explicitly separated into a term depending only on $s$, that finally can be cancelled due to the charge neutrality condition. We take a similar multiplier $c(n, r, s) = \exp(-s|n + r|^2)$ yielding the same rate of convergence (since $0 \leq r \leq 1$ in units of $L$). As the sum, multiplied by $c$, is invariant to an arbitrary substitution $r \rightarrow n + r$, the chosen convergence factor allows one to preserve the periodicity of the potential in order to avoid any possible artefacts in the final results.

For the sake of clearness of the derivation, it is convenient to use reduced length units, that is to use the size of the box $L$ as unity of length and substitute $r_{ij}$ by $r_{ij}/L$. From now on, and for simplicity, we use the notation $r$ for $r_{ij}$ and, in case of possible ambiguity, we will stick to the standard notation $r_{ij}$. Also, we rewrite the potential energy by splitting the total sum (1) into two terms: $I_{01}$ (the sum of the interactions between a particle with all the other particles in the box), and $I_{00}$ [the sum of the interaction of a particle with its own images, comprised of the components $i = j$]
in Equation (1)]. Explicitly,

\[ U = \frac{1}{L^2} (I_{01} + I_{00}), \]  

(2)

with

\[ I_{01} = \sum_{n \in \mathbb{Z}^3} \sum_{1 \leq i < j \leq N} \frac{q_i q_j \exp(-s|n|^2)}{|r_{ij} + n|^k}, \]  

(3)

\[ I_{00} = \frac{1}{2} \sum_{n \in \mathbb{Z}^3} \exp(-|n|^2) \sum_{j=1}^N q_j^2, \]  

(4)

where the shorthand notation \( n = |n| \) is used.

First, let us focus on the \( I_{01} \) term, which we rewrite as

\[ I_{01} = \sum_{1 \leq i < j \leq N} q_i q_j \psi(r, s), \]  

(5)

where we have defined the ‘screened’ interaction potential \( \psi(r, s) = \sum_n \exp(-s|n|^2)/|r + n|^k \), extended from a single cell to the whole coordinate space. Since the total potential energy consists of a sum of pair interaction components, we may consider a single pair without any loss of generality.

Let us apply the equation

\[ x^{-2s} = \frac{1}{\Gamma(s)} \int_0^\infty t^{s-1} \exp(-tx^2) \, dt, \]  

(6)

representing the definition of the gamma-function, to the polytropic potential \( |r + n|^k \). Then the function \( \psi \) may be represented in an integral form,

\[ \psi(r, s) = \frac{1}{\Gamma(k/2)} \int_0^{\infty} t^{k-1} \sum_n \exp(-t|r + n|^2) \times \exp(-s|n|^2) \, dt. \]  

(7)

We expect that the integral (7) contains a singularity that will be located in the vicinity of zero. Therefore, we split this integral into two domains \([0, a^2]\) and \([a^2, \infty)\), the corresponding integrals being denoted as \( \psi_{\text{fin}} \) and \( \psi_{\text{inf}} \), where \( \alpha \) is some arbitrary positive constant,

\[ \psi(r, s) = \psi_{\text{fin}}(r, s) + \psi_{\text{inf}}(r, s). \]  

(8)

In the following, we analyse the two terms of the previous sum (8).

(1) The explicit analytical form of the term \( \psi_{\text{inf}}(r, s) \) can be found

\[ \psi_{\text{inf}}(r, s) = \frac{1}{\Gamma(k/2)} \sum_n \int_0^{\infty} t^{k-1} \exp(-t|n|^2 - s|n|^2) \, dt \]

\[ = \sum_n \frac{\exp(-s|n|^2) \Gamma(k/2, \alpha^2|n|^2)}{\Gamma(k/2)}, \]  

(9)

where \( \Gamma(a, z) \) is the incomplete gamma function. From the large distance asymptotic expansion of this function, one obtains that the above lattice sum is absolutely and uniformly convergent if \( s \geq 0 \) and \( \alpha > 0 \). Therefore, one may simply take the limit of vanishing screening \( s \to 0 \),

\[ \psi_{\text{inf}}(r, s) \xrightarrow{s \to 0} \frac{1}{\Gamma(k/2)} \sum_n \frac{\Gamma(k/2, \alpha^2|n|^2)}{|n|^k}. \]  

(10)

(2) The calculation of \( \psi_{\text{fin}}(r, s) \) is done by making a separate analysis of the \( n = 0 \) case,

\[ \psi_{\text{fin}}(r, s) = \psi_{\text{fin}}^{m \neq 0}(r, s) + \psi_{\text{fin}}^{m = 0}(r, s). \]  

(11)

Explicitly,

\[ \psi_{\text{fin}}^{m \neq 0}(r, s) = \frac{\pi^{3/2}}{\Gamma(k/2)} \sum_{m \neq 0} \int_0^{\infty} t^{k-1} \exp[-\pi^2 m^2] \frac{1}{(t + s)^{3/2}} \, dt, \]  

(12)

\[ \psi_{\text{fin}}^{m = 0}(r, s) = \frac{\pi^{3/2}}{\Gamma(k/2)} \int_0^{\infty} t^{k-1} \exp[-\pi^2 m^2] \frac{1}{(t + s)^{3/2}} \, dt, \]  

(13)

where we have used the Jacobi transformation [47,48]

\[ \sum_n \exp(-s|n + r|^2) = (\frac{\pi}{s})^{3/2} \sum_m \exp[-\pi^2 m^2/s + 2\pi imr] \text{ for } m, r \in \mathbb{Z}^3, \]  

(14)

applied to

\[ \exp[-s|n + r|^2 - t|n|^2] = \exp[-(s + t)|n + r|^2]. \]  

(15)

We evaluate the integral \( \psi_{\text{fin}}^{m \neq 0}(r, s) \) by the following analysis. Consider separately the following factor of the integrated expression from (12)

\[ M = \frac{\exp[-\pi^2 m^2]}{(t + s)^{3/2}}. \]  

(16)

It is clearly continuous and bounded on \((0, +\infty)\) as a function of \((t + s)\), also notice that \( t^{k-1} \) is absolutely integrable on \((0, \alpha^2)\) for \( k > 0 \). In accordance with the standard convergence test for improper integrals, the integral \( \psi_{\text{fin}}^{m \neq 0}(r, s) \) converges absolutely and uniformly
with \( s \) being considered as a parameter. Then, the limit \( s \to 0 \) may be carried out and the integral becomes

\[ \psi_{\text{fin}}^{m=0}(r, s) = \frac{\pi^{3/2}}{\Gamma(k/2)} \sum_{m \neq 0} \exp(2\pi imn) \times \int_0^{\infty} \frac{r^{k-5/2}}{(1 + r)^{3/2}} \exp \left(-\frac{\pi^2 m^2}{t} \right) \, dt, \quad (17) \]

\[ = \sum_{m \neq 0} \frac{\pi^3 \cos(2\pi m) \Gamma(k/2)}{\Gamma(k/2)} \omega^k E_{(k-1)/2} \left( \frac{\pi^2 m^2}{\omega^2} \right). \quad (18) \]

The function \( E_n(z) \) is the exponential integral function, and we have cancelled the imaginary part of the sum \((17)\) by grouping the pairs with \( n \) and \( -n \).

Now, we analyse the second term of \( \psi_{\text{fin}}(r, s) \),

\[ \psi_{\text{fin}}^{m=0}(r, s) = \frac{\pi^{3/2}}{\Gamma(k/2)} \int_0^{\infty} \frac{r^{k-1}}{(r + s)^{3/2}} \, dr. \quad (19) \]

In terms of a new variable \( v = s/(t + s) \),

\[ \psi_{\text{fin}}^{m=0}(r, s) = \frac{\pi^{3/2}}{\Gamma(k/2)} \int_{s/(t + s)}^1 \frac{(1 - v)^{k-1}}{v^{k-3/2}} s^{(k-3)/2} \, dv. \quad (20) \]

The integration of \( \psi_{\text{fin}}^{m=0}(r, s) \) for a \( 1/|r|^k \) interaction has to be carefully analysed as a function of \( k \): \( 1 \leq k < 3 \), long-range potential; \( k = 3 \), marginal case; and \( k > 3 \), short-range potential.

(a) Suppose \( 1 \leq k < 3 \). The resulting integral,

\[ \psi_{\text{fin}}^{m=0}(r, s) = \psi_{\text{fin}}^{m=0}(r, s) = \frac{\pi^{3/2}}{\Gamma(k/2)} \int_{s/(t + s)}^1 \frac{(1 - v)^{k-1}}{v^{k-3/2}} s^{(k-3)/2} \, dv \]

may be given explicitly in terms of incomplete beta- and incomplete gamma-functions. Expanding the resulting function for small \( s \),

\[ \psi_{\text{fin}}^{m=0}(r, s) = s^{(k-3)/2} \frac{2\pi \Gamma}{2} \left[ \frac{3 - k}{2} \right] + \frac{2\pi \Gamma}{2} \left( \frac{3 - k}{2} \right) + O(s) \]

may be given explicitly in terms of incomplete beta- and incomplete gamma-functions. Expanding the resulting function for small \( s \),

\[ S(s) = s^{(k-3)/2} \frac{2\pi \Gamma}{2} \left[ \frac{3 - k}{2} \right] \quad (22) \]

(b) We remind that the choice of a convergence factor (that explicitly affects the summation order) may in principle lead to additional contributions in the total energy if the convergence of the sum is conditional (like for a charge-neutral cell of Coulomb particles with non-zero total dipole moment). In the original derivation of de Leeuw et al. [2], the factor \( \exp(-s\alpha^2) \) results in an additional dipole-like component in \( \psi_{\text{fin}}^{m=0} \), which breaks the periodicity of the potential and therefore complicates its use in simulations with periodic boundary conditions. Moreover, this procedure [2] yields a nonvanishing dipole term exclusively for \( k = 1 \) in 3D geometry, with the rest of the sums remaining unchanged. From our point of view, this discontinuity points to a nonphysical character of the dipole term appearing in the case of the Coulomb potential. Nevertheless, in a number of studies [45,46] it is considered as a first-order correction when the convergence to the thermodynamic limit is analysed. The mere fact that the results for the two different convergence multipliers coincide when \( k > 1 \) is a consequence of the absolute convergence of the higher multipole contributions in this case.

(c) Suppose \( k = 3 \). In this marginal case, the expression \((21)\) may be integrated directly to yield the following logarithmic dependence

\[ \psi_{\text{fin}}^{m=0}(r, s) = \frac{\pi^{3/2}}{\Gamma(2)} \left( \frac{2\pi \alpha}{(\alpha^2 + s)^{1/2} + \ln(s + 2\alpha^2 + 2\alpha(s + \alpha^2)^{1/2}) - \ln s} \right), \]

which close to \( s = 0 \) expands as

\[ \psi_{\text{fin}}^{m=0}(r, s) = -2\pi \ln s - 4\pi + 4\pi \ln(2\alpha) + O(s \ln s) \]

with the diverging term

\[ S(s) = -2\pi \ln s. \quad (26) \]

(d) Consider the remaining option \( k > 3 \). In this case, \( (1 - \frac{1}{k})^{-1} \) is bounded from above and \( (k - 1)/2 > 1 \). It means that the integral converges absolutely and the only finite contribution to the integral comes from the first (constant) term of the integral expansion for small \( s \),

\[ \psi_{\text{fin}}^{m=0}(r, s) = \frac{2\pi \Gamma}{(k - 3) \Gamma[ k/2 ]} \left( \frac{3 - k}{2} \right). \quad (27) \]

The second term of the total potential energy, \( I_{00} \)

(2) can be derived in a similar form to the first one. The procedure to find the form of \( \psi(r, s) \) is repeated here.
with \( r_{ij} = 0 \), hence the results are obtained straightforwardly via (10), (18), (25) and (27),

\[
I_{00} = \sum_{i=1}^{N} q_i^2 \left[ \frac{1}{\Gamma(k/2)} \sum_{n} \frac{\Gamma(k/2, \alpha^2 n^2)}{n^k} \right.
+ \sum_{m \neq k} \frac{\pi^{3/2} \alpha^{k-3} E_{(k-1)/2}(\pi^2 m^2/\alpha^2)}{\Gamma(k/2)}
\left. - \frac{\alpha^k}{\Gamma(\xi + 1)} + \psi_{\text{fin}}^{m=0}(r, s) \right],
\]

(28)

with the term \( \psi_{\text{fin}}^{m=0}(r, s) \) depending on the potential parameter \( k \) via (22), (25) or (27).

Putting all together, the potential energy can be written in a more compact form as

\[
U = \frac{1}{L^3} (I_{01} + I_{00})
\]

\[
= \frac{1}{L^3} \sum_{i<j} q_i q_j \psi(r_{ij}/L) + \frac{1}{2L^3} \sum_{i=1}^{N} q_i^2 \xi
+ \frac{1}{L^3} \sum_{i<j} q_i q_j S(s) + \frac{1}{2L^3} \sum_{i} q_i^2 S(s),
\]

(29)

with the generalized potential,

\[
\psi(r) = \sum_{n} R(n, r) + \sum_{m \neq 0} K(m, r) + C_1.
\]

(30)

A constant shift in the definition of \( \psi \) is introduced to satisfy the property \( \int_{\text{cell}} \psi dr = 0 \), convenient for a proper treatment of the background contributions (see Appendix). The functions entering in Equation (30) are defined as

\[
R(n, r) = \frac{\Gamma(k/2, \alpha^2 |r + n|^2)}{\Gamma(k/2) |r + n|^k}
\]

(31)

\[
K(m, r) = \kappa(m) \cos(2\pi mr),
\]

(32)

with

\[
\kappa(m) = \frac{\pi^{3/2} \alpha^{k-3}}{\Gamma(k/2)} E_{(k-1)/2}(\pi^2 m^2/\alpha^2).
\]

(33)

The explicit form of the function \( S(s) \) depends on the \( k \) value,

\[
S(s) = \begin{cases} 
2\pi^{3/2} m^{3/2} \xi & \text{if } k \leq 3 \text{ (singular term)}, \\
-2\pi \ln s & \text{if } k = 3 \text{ (singular term)}, \\
0 & \text{if } k > 3,
\end{cases}
\]

(34)

and the term \( \xi \) depends only on the choice of \( \alpha \),

\[
\xi = \sum_{n \neq 0} \rho(n) + \sum_{m \neq 0} \kappa(m) + C_1 + C_2,
\]

(35)

with

\[
\rho(n) = \frac{\Gamma(k/2, \alpha^2 n^2)}{\Gamma(k/2)n^k},
\]

(36)

and \( \kappa(n) \) defined in Equation (33). The constants \( C_1 \) and \( C_2 \) are explicitly,

\[
C_1 = \begin{cases} 
\frac{2\pi^{3/2} \alpha^{k-3}}{(k - 3) \Gamma(k/2)} & \text{if } k \neq 3, \\
-4\pi + 4\pi \ln(2\alpha) & \text{if } k = 3,
\end{cases}
\]

(37)

\[
C_2 = \frac{\alpha^k}{\Gamma(\xi + 1)}.
\]

(38)

**2.3. Removing singularities for \( k \leq 3 \)**

The diverging part \( U_s \) (containing a singularity) of the total potential energy is equal to

\[
U_s = \frac{1}{L^3} \sum_{i<j} q_i q_j S(s) + \frac{1}{2L^3} \sum_{i} q_i^2 S(s) = \frac{1}{2L} \left( \sum_{i} q_i \right)^2 S(s)
\]

(39)

and vanishes, if the charge neutrality condition \( \sum q_i = 0 \) is taken.

Consider now a charge-neutral system with a neutralizing background consisting of a large number of identical uniformly distributed particles of the opposite charge (the ‘jellium’ model). We denote the numbers of negatively charged particles \( q_- \) and positively charged (background) particles \( q_+ \) as \( N_- \) and \( N_+ \), respectively. By imposing charge neutrality, \( q_+ = -[N_-/N_+]q_- \), with \( N \) the total number of particles, \( N = N_- + N_+ \).

The potential energy for the jellium model can be written as

\[
U = \frac{1}{L^3} \sum_{i<j} q_i q_j \psi(r_{ij}/L) + \frac{N_- q_-^2 + N_+ q_+^2}{2L^3} \xi.
\]

(40)

The second term in Equation (41) has a component proportional to \( N_+ q_+^2 \). Note that the negative charges \( q_- \) and their number \( N_- \) is defined by the problem and therefore fixed. Hence, in the limit \( N_+ \to \infty \), this term cancels \( N_+ q_+^2 = (N_- q_-^2)/N_+ \to 0 \), and therefore this background contribution may be eliminated to yield

\[
\frac{N_- q_-^2 + N_+ q_+^2}{2L^3} \xi = \frac{N_- q_-^2}{2L^3} \xi.
\]

(41)

Concerning the first term of Equation (41), let us split it into three pieces,

\[
\frac{1}{L^3} \sum_{i<j \leq N} q_i q_j \psi(r_{ij}) = \frac{1}{L} (S_- + 2S_+ + S_{++}),
\]

(42)
where the first sum corresponds to the interaction between the negative charges

\[ S_{--} = \sum_{1 \leq i < j \leq N_{--}} q_i q_j \psi(r), \quad (43) \]

the second sum is the interaction of the negatively charged particles with the positive charges of the background

\[ S_{-+} = \sum_{i=1}^{N_{--}} \sum_{j=1+N_{--}} q_i q_j \psi(r), \quad (44) \]

and the third one is the interaction between the background charges

\[ S_{++} = \sum_{1+N_{--} \leq i < j \leq N_{--}+N_{--}} q_i q_j \psi(r). \quad (45) \]

The last two terms \( S_{-+} \) and \( S_{++} \) are easily shown to be zero in the limit \( N_{--} \to \infty \) as a consequence of the zero value of the integral of \( \psi \) over the simulation cell (see Appendix).

With the above considerations we can finally write the expression for the potential energy within the jellium model as

\[ U^{\text{jel}} = \frac{q^2}{L^k} \sum_{i<j} \psi(r_{ij}/L) + \frac{N q^2}{2L^k} \xi. \quad (46) \]

In the more general case of different charges in a charge-neutral simulation cell (with a long-range potential) or a system with an arbitrary short-range potential the potential energy is given by

\[ U^{\text{gen}} = \frac{1}{L^k} \sum_{i<j} q_i q_j \psi(r_{ij}/L) + \frac{\sum_{m=1}^{N} q^2_m}{2L^k} \xi. \quad (47) \]

A certain analytical conversion of the sum in the reciprocal space is also possible in order to sum it up faster. Expanding the sum that defines \( K(m, r) \) (32), one can simplify it in the following way,

\[ \sum_{i<j} q_i q_j \sum_{m \neq 0} K(m, r) \]

\[ \quad = \sum_{m \neq 0} \kappa(m) \sum_{i<j} q_i q_j \cos(2\pi m r) \]

\[ \quad = \frac{1}{2} \sum_{m \neq 0} \kappa(m) \sum_{i<j} q_i q_j \left[ \cos(2\pi m r_i) \cos(2\pi m r_j) + \sin(2\pi m r_i) \sin(2\pi m r_j) \right] - \frac{1}{2} \sum_{m \neq 0} q^2_i \sum_{m \neq 0} \kappa(m) \]

\[ \quad = \frac{1}{2} \sum_{m \neq 0} \kappa(m) \left[ \sum_{j} q_j \exp(2\pi i m r_j) \right]^2 - \frac{1}{2} \sum_{i} q^2_i \sum_{m \neq 0} \kappa(m). \quad (48) \]

In this form, the sum over all pairs of particles in the reciprocal space is represented as a single sum over particles and thus it scales as \( O(N) \) instead of \( O(N^3) \). Notice that the number of prefactors \( \kappa(m) \) and exponents in the sum depends on a chosen cut-off, which in general also might depend on \( N \), making the overall complexity of the \( k \)-space grow. Naïve schemes with \( \alpha \) and the cut-off not depending on \( N \) do not take into account the interplay between the \( r \)-space and \( k \)-space sum complexities, thus leaving at least \( O(N^3) \) in one of them. Nevertheless, as we show later, optimization with \( \alpha \) and cut-off depending on \( N \) gives a best total complexity of \( O(N^{3/2}) \). An alternative method to sum up the momentum space part is to use fast Fourier transform-based techniques (like PME), which is fast as \( O(N \ln N) \).

The last term in Equation (49) cancels the \( \kappa(m) \) component of \( \tilde{\xi} \). Introduce the notation

\[ \tilde{\psi}(r) = \sum_{n} R(n, r) + C_1, \quad (49) \]

\[ \tilde{\xi} = \sum_{n \neq 0} \rho(n) + C_1 + C_2, \quad (50) \]

\[ \tilde{S}_{\text{equal}}(m) = q \sum_{j} \exp(2\pi i m r_j/L), \quad (51) \]

\[ \tilde{S}_q(m) = \sum_{j} q_j \exp(2\pi i m r_j/L), \quad (52) \]

where \( \tilde{S}_{\text{equal}} \) is used when the system of equally charged particles \( q_- \) is considered. Within this notation the potential energy may be rewritten in the following forms, which are more efficient for numerical implementation,

\[ U^{\text{jel}} = \frac{q^2}{L^k} \sum_{i<j} \tilde{\psi}(r_{ij}/L) + \frac{1}{2L^k} \sum_{m \neq 0} \kappa(m) |\tilde{S}_{\text{equal}}(m)|^2 \]

\[ + \frac{N q^2}{2L^k} \tilde{\xi}, \quad (53) \]

\[ U^{\text{gen}} = \frac{1}{L^k} \sum_{i<j} q_i q_j \tilde{\psi}(r_{ij}/L) + \frac{1}{2L^k} \sum_{m \neq 0} \kappa(m) |\tilde{S}_q(m)|^2 \]

\[ + \frac{\sum_{m=1}^{N} q^2_m}{2L^k} \tilde{\xi}, \quad (54) \]

with \( r, r_{ij} \) in the original length units.

### 2.4. Short-range potentials and the marginal case

In the case of a short-range interaction \( (k > 3) \), the potential energy does not diverge, which is clear from
the form of the singular term $S(s)$ [see Equation (34)]. Hence, there is no need to add a neutralizing background and, even more, the background must be necessarily excluded since it leads to a divergence in the energy. This is easily seen by considering the potential energy of the background separately

$$U_{bg} = C \int_0^{\text{cell}} \frac{dr}{|r|^3},$$

(55)

which contains a singularity in zero. The expression for the potential energy is simply equal to Equation (48),

$$U = \frac{1}{L^d} \sum_{i,j} q_i q_j \psi(r_{ij}/L) + \sum_{i=1}^N q_i^2/2L^d \xi.$$  

(56)

When $k = 3$ (marginal case), both ultraviolet (coming from short-range contributions) and infrared (coming from long-range contributions) divergences arise in zero for the background as well as in the vicinity of infinity (the logarithmic divergence in the energy of negative charges). The only coherent model here is a plain 'quasi-neutral' gas consisting of a mixture of a finite number of charges per box with the constraint $\sum q_i = 0$, i.e. with the positive background excluded.

3. Ewald method for two-dimensional systems

3.1. General notes for lower dimensions

The Ewald sums can be extended to two-dimensional (2D) systems interacting through polytropic potentials. The difference with the 3D case comes from a different form of the Jacobi imaginary transformation for the Jacobi $\theta$-functions [its 3D form is given in Equation (14)].

The ‘third’ Jacobi $\theta$-function $\theta_3(z, \tau)$ is defined as

$$\theta_3(z|\tau) = \sum_{m=-\infty}^{+\infty} \exp(i\pi m^2) \exp(2\pi i m z),$$

(57)

and satisfies the Jacobi imaginary transformation,

$$\theta_3(z|\tau) = (-i\tau)^{-1/2} \exp(i\tau z^2/\pi) \theta_3(z|\tau'),$$

(58)

with $\tau = 1/\tau$. Under the change of variables, $z = \pi r$ and $\tau = i\tau/s$, the $\theta$-function becomes a Gaussian, which is the relevant function for performing the Ewald sums,

$$\sum_{n=-\infty}^{+\infty} \exp[-s(r + n)^2]$$

$$= (\pi/s)^{1/2} \sum_{m=-\infty}^{+\infty} \exp(-\pi^2 m^2/s) \exp(2\pi i m r).$$

(59)

This expression will be used later, in the derivation of the Ewald sum in one-dimensional systems. Equation (60) may be easily generalized to the 2D geometry,

$$\sum_n \exp(-s|r + n|^2)$$

$$= (\pi/s) \sum_m \exp(-\pi^2 m^2/s) \exp(2\pi i m r).$$

(60)

Comparing this result for 2D with its 1D (60) and 3D (14) counterparts one finds that the dimensionality $D$ affects only the constant multiplier as $(\pi/s)^{D/2}$.

3.2. Derivation

The analytical derivation of the Ewald sum in 2D proceeds similarly to the one already presented for 3D. Equations from (2) to (11) are also valid here because their derivation is done without explicit reference to the dimensionality of the problem. In particular, the integral $\psi_{\text{int}}(r, s)$ converges absolutely and to the same value

$$\psi_{\text{int}}(r, s) \rightarrow \frac{1}{\Gamma(k/2)} \sum_n \frac{\Gamma(k/2, \alpha^2|r + n|^2)}{|r + n|^k}. $$

(61)

We make the same decomposition of the integral $\psi_{\text{int}}(r, s)$ as in 3D,

$$\psi_{\text{int}}(r, s) = \psi_{\text{int}}^{m\neq 0}(r, s) + \psi_{\text{int}}^{m=0}(r, s),$$

(62)

with

$$\psi_{\text{int}}^{m\neq 0}(r, s) = \frac{\pi}{\Gamma(k/2)} \sum_{m \neq 0} \int_0^{\infty} \frac{\tau^{-1}}{t + s} \exp\left[-\pi^2 m^2 \frac{t + s}{t + s} + 2\pi i m r \right] dt,$$

(63)

$$\psi_{\text{int}}^{m=0}(r, s) = \frac{\pi}{\Gamma(k/2)} \int_0^{\infty} \frac{\hat{\tau}^{-1}}{t + s} \ dt,$$

(64)

where the two-dimensional variant of the Jacobi transformation (61) is used. The difference between the pair of equations (64) and (65) and their three-dimensional analogues (12) and (13) relies in a substitution of the 3D factor $(\pi/(t + s))^{3/2}$ by the 2D one $\pi/(t + s)$.

First, we consider the term $\psi_{\text{int}}^{m\neq 0}(r, s)$. Following the same analysis as for its 3D counterpart, it can be shown that this parametric integral also
converges absolutely. It yields

$$\psi_{\text{fin}}^{m=0}(r,s) = \frac{\pi}{\Gamma(k/2)} \sum_{m=0}^{\infty} \exp(2\pi im) \int_0^\infty \frac{\exp(-\pi m^2 t)}{t} dt$$

$$= \sum_{m=0}^{\infty} \frac{\pi \cos(2\pi m)}{\Gamma(k/2)} \frac{e^{-\pi m^2\alpha^2}}{k/2}.$$

(65)

The modification of the integral $\psi_{\text{fin}}^{m=0}$ is less straightforward, since it requires specific integrations and expansions in the series for small $s$. Namely, we have to evaluate the integral

$$\psi_{\text{fin}}^{m=0} = \frac{\pi}{\Gamma(k/2)} \sum_{s=0}^{\infty} \frac{(1-\psi)^{s-1}}{v^{k/2}} \frac{\sin(s\pi/\alpha^2)}{\Gamma(k/2)}.$$  

(66)

which is the 2D equivalent of Equation (20).

In the following, we consider separately the cases of long-range potential ($1 \leq k < 2$), marginal interaction ($k = 2$) and short-range potential ($k > 2$).

1. $1 \leq k < 2$. As in 3D, the integral can be found analytically via the incomplete beta- and incomplete gamma-function with known series expansions for small $s$. Omitting these unnecessary intermediate expressions, we give the final expansion for $\psi_{\text{fin}}^{m=0}$,

$$\psi_{\text{fin}}^{m=0} = \frac{\pi^2}{\sin(\pi k/2)\Gamma(k/2)} + \frac{2\pi\alpha^{k-2}}{(k-2)\Gamma(k/2)} + O(s^{k/2}).$$

(67)

The first term of the expansion,

$$S(s) = \frac{\pi^2}{\sin(\pi k/2)\Gamma(k/2)},$$

clearly diverges when $s \to 0$. Similarly to the 3D case, this term is cancelled in a charge-neutral cell and hence,

$$\psi_{\text{fin}}^{m=0} = \frac{2\pi\alpha^{k-2}}{(k-2)\Gamma(k/2)}. $$

(69)

2. $k = 2$. The integration of Equation (67) is performed to yield in the limit $s \to 0$ a marginal logarithmic dependence,

$$\psi_{\text{fin}}^{m=0} = -\pi \ln s + 2\pi \ln \alpha + O(s \ln s).$$

(70)

As for the 3D geometry, the jellium model is inapplicable in this particular case since the energy of the continuous background diverges. Nonetheless the diverging component

$$S(s) = -\pi \ln s$$

(71)

can be removed if we consider a charge-neutral system with a finite number of charges. In this case,

$$\psi_{\text{fin}}^{m=0} = 2\pi \ln \alpha.$$  

(72)

3. $k > 2$. The integral (67) can be evaluated by taking $s = 0$, since its convergence is absolute,

$$\psi_{\text{fin}}^{m=0} \to \frac{2\pi\alpha^{k-2}}{(k-2)\Gamma(k/2)}.$$  

(73)

The second potential energy component, $I_{00}(2)$, is calculated as in the 3D case. The result for 2D is

$$I_{00}(s) = \sum_{j=1}^{N} q_j^2 (\psi_{\text{fin}}^{m=0}(0,s) + \psi_{\text{inf}}(0,s) - \psi_{\text{inf}}^{m=0}(0,s)).$$

$$= \sum_{j=1}^{N} q_j^2 \left[ \frac{\pi}{\Gamma(k/2)} \frac{e^{-\pi m^2\alpha^2}}{k/2} \right] \times E_{k/2} \left( \frac{\pi^2 m^2}{\alpha^2} - \frac{\alpha^k}{\Gamma(k/2)} \right).$$

(74)

### 3.3. Final expressions

With respect to the 3D case, the changes in the 2D Ewald sum appear in those terms in which the Jacobi transformation is used, that is in $\kappa(n)$ and $C_1$,

$$\kappa(m) = \pi \left( \frac{e^{-\pi m^2\alpha^2}}{k/2} \right)^{1/2}.$$  

(75)

$$C_1 = \psi_{\text{fin}}^{m=0}. $$

(76)

The other terms, namely $R(r,n)$, $\rho(n)$ and $C_2$, are not affected by dimensionality and may be taken directly from the previous section.

Within the jellium model for a long-range potential ($k < 2$), the Ewald sum is given by

$$U^{\text{jel}} = \frac{q_j^2}{L^k} \sum_{i<j} \psi(r_{ij}/L) + \frac{Nq_j^2}{2L^k} \xi.$$  

(77)

A more general form, applicable to any system with a short-range potential ($k > 2$), a charge-neutral system with long-range interaction ($k < 2$), or a marginal ($k = 2$) potential is expressed as

$$U^{\text{gen}} = \frac{1}{L^k} \sum_{i<j} q_j \psi(r_{ij}/L) + \frac{\xi}{2L^k} \sum_{j=1}^{N} q_j^2.$$  

(78)

In the same way as for the 3D systems we can modify the sum in the reciprocal space, and with the same notations (50)–(53) ($\rho$, $R$ and the constants $C_1$, $C_2$ are the new ones, corresponding to 2D case).
the potential energy may be given by
\[ U^{\text{jd}} = \frac{q^2}{L} \sum_{i<j} \tilde{\psi}(r_{ij}/L) + \frac{1}{2L^2} \sum_{m \neq 0} \kappa(m) j_{\text{equal}}^2 + \frac{Nq^2}{2L^2} \tilde{\xi}, \]
(79)

\[ U^{\text{gen}} = \frac{1}{L^2} \sum_{i<j} q_i q_j \tilde{\psi}(r_{ij}/L) + \frac{1}{2L^2} \sum_{m \neq 0} \kappa(m) j_{\text{equal}}^2 + \sum \frac{q_i^2}{2L^2} \tilde{\xi}, \]
(80)

4. Ewald method for one-dimensional systems

As has been commented on before for the 2D case, the differences due to dimensionality are caused by the form of the Jacobi imaginary transformation. In the derivation for 1D, one needs the following ones

\[ x^{-2z} = \frac{1}{\Gamma(s)} \int_0^\infty t^{s-1} \exp(-tx^2) \, dt, \]
(81)

\[ \sum_{n=-\infty}^{+\infty} \exp(-n^2) = (\pi/s)^{1/2} \sum_{m=-\infty}^{+\infty} \exp(-\pi^2 m^2/s), \]
(82)

\[ \sum_{n=-\infty}^{+\infty} \exp(-n(n^2)) = (\pi/s)^{1/2} \sum_{m=-\infty}^{+\infty} \exp(-\pi^2 m^2/s) \exp(2\pi i m r). \]
(83)

Similarly to that discussed in the previous section, the only terms to be changed are those where the Jacobi transformation is used, namely \( \psi_{\text{fin}}^m = \psi_{\text{fin}}^0 \) (in \( I_0 \) in a radial-dependent form, in \( I_{00} \) for \( r = 0 \)). The difference arises from a different power exponent (1/2) in (83) and (84), that is in (18) \( k \) has to be substituted by \( k + 2 \) (and \( \pi^{3/2} \) by \( \pi^{1/2} \), respectively), yielding

\[ \psi_{\text{fin}}^m = \sum_{n \neq 0} \frac{\pi^{1/2} \exp(2\pi i m r)}{\Gamma(k/2)} d^{k-1} E_{(k+1)/2} \left( \frac{\pi^2 m^2}{\alpha^2} \right). \]
(84)

As far as the term \( \psi_{\text{fin}}^m = 0 \) is concerned, we should perform a simple integration and do a series expansion for small \( s \),

\[ \psi_{\text{fin}}^m = \frac{\pi^{1/2}}{\Gamma(k/2)} \int_0^1 \frac{(1 - v)(k+1/2)}{v^{(k+1)/2}} \, dv. \]
(85)

The estimation of this integral depends on the \( k \) value. In the following, we detail this analysis.

(1) \( k = 1 \), the marginal case,

\[ \psi_{\text{fin}}^m = \frac{\pi^{1/2}}{\Gamma(k/2)} (-\ln s - 2 + 2 \ln(2\alpha)) + O(s). \]
(86)

As before, we keep only the constant term, considering the diverging term absent due to the charge neutrality condition. Therefore, with \( \Gamma(1/2) = \pi^{1/2} \) one has

\[ \psi_{\text{fin}}^m = -2 + 2 \ln(2\alpha). \]
(87)

(2) \( k > 1 \), the short-range potential,

\[ \psi_{\text{fin}}^m = \frac{\pi^{1/2}}{\Gamma(k/2)} \frac{2\alpha^{k-1}}{k-1} + O(s) + O(s^{(k-1)/2} \ln s). \]
(88)

In the limit \( s \to 0 \), it yields

\[ \psi_{\text{fin}}^m = \frac{2\pi^{1/2} \alpha^{k-1}}{(k-1) \Gamma(k/2)}. \]
(89)

resembling the 3D result (27), with the change \( k \to k + 2 \) (except in the \( \Gamma \) term) and \( \pi^{3/2} \to \pi^{1/2} \).

The final result for the one-dimensional Ewald summation reads

\[ \psi(r) = \sum_n \rho(n,r) + \sum_{m \neq 0} K(m,n) + C_1, \]
(90)

\[ \xi = \sum_{n \neq 0} \rho(n) + \sum_{m \neq 0} K(m) + C_1 + C_2, \]
(91)

where \( C_1 = \psi_{\text{fin}}^m = 0 \) is taken from the expressions (88) (if \( k = 1 \)) or (90) (if \( k > 1 \)).

For \( k = 1 \), the only consistent system is the charge-neutral one with a finite number of particles. In this case and for a short-range potential \( (k > 1) \) one the potential energy is given by

\[ U^{\text{gen}} = \frac{1}{L} \sum_{i<j} q_i q_j \psi(r_{ij}/L) + \sum_{i=1}^N q_i^2 L^2 \tilde{\xi}. \]
(92)

Although the Ewald method is applicable to one-dimensional problems, there is a direct way to calculate the sums for polytropic potentials

\[ U = \frac{1}{L^k} \sum_{n=-\infty}^{+\infty} \frac{1}{|r + n|^k}. \]
(93)

For \( k > 1 \), this sum can be represented as a linear combination of the Hurwitz zeta functions,

\[ \frac{1}{L^k} \sum_{n=-\infty}^{+\infty} \frac{1}{|r + n|^k} = \frac{1}{L^k} (H_{k}(r) + H_k(1 - r)). \]
(94)

In particular, for \( k = 2 \) the sum converts into a familiar expression used in the Calogero–Sutherland model [49,50],

\[ \frac{1}{L^2} \sum_{n=-\infty}^{+\infty} \frac{1}{|r + n|^2} = \frac{\pi^2}{L^2} \sin^2(\pi r). \]
(95)
Notice that the sum (95) may be expressed in terms of trigonometric functions only for even values of $k$ via $(k-2)$ times differentiation of Equation (96). Anyway, the possibility to find exact expressions for infinite sums in 1D suggests that the use of the Ewald method might not be needed, but we keep it as a possibly useful mathematical relation and for completeness.

5. Ewald method in a rectangular box of arbitrary side lengths

5.1. 3D case

A special and interesting situation arises if we consider a simulation cell in a more general way, as a rectangular box with different side lengths ($L_x$, $L_y$, $L_z$ in the corresponding dimensions). The need to deal with a box of unequal size lengths may occur in the simulation of a solid with a noncubic lattice (the simplest examples include a hexagonal packed crystal in 3D geometry), since the lattice vectors $\mathbf{n}$ in the sum over images on (1) are no longer orthogonal. Focusing our analysis to a 3D geometry, the potential energy is now given by

$$ U = \frac{1}{2} \sum_{\mathbf{n} \in \mathbb{Z}^3} \left[ \sum_{i=1}^{N} \sum_{j=1}^{N} \phi(r_{ij} + L_0 \mathbf{n}_i) \right], $$

(96)

with $\mathbf{n} = (n_x L_x, n_y L_y, n_z L_z)/L_0$, $n_x, y, z$ being integer vectors along the corresponding axis $x, y, z$. We have introduced the geometric average $L_0 = (L_x L_y L_z)^{1/3}$ and we will use reduced $L_0$ units for $r_{ij}$, and hence $r_{ij}$ will be adimensional. Repeating the standard procedure, we multiply the potential energy by a Gaussian term $\exp(-s|\mathbf{n}_i + r|^2)$ and, at the end, we take the limit $s \to 0$, separating the converging part, if present. We group separately the interaction with images of other particles $I_{01}$ and the interaction of a particle with its own images $I_{00}$.

$$ U = \frac{1}{L_0^2} (I_{01} + I_{00}), $$

(97)

where

$$ I_{01} = \sum_{\mathbf{n} \in \mathbb{Z}^3} \left[ \sum_{1 \leq |l| < |n| \leq N} q_l q_j \exp(-s|\mathbf{n}_i + r|^2) \right], $$

(98)

$$ I_{00} = \frac{1}{2} \sum_{\mathbf{n} \in \mathbb{Z}^3 \setminus 0} \exp(-s|\mathbf{n}_i|^2) \sum_{i=1}^{N} q_i^2. $$

(99)

Comparing the relations (98)–(100) to the cubic case (2)–(4), one notices that these relations remain unchanged if $\mathbf{n}$ is formally substituted by $\mathbf{n}_i$ and the constant coefficient $1/L_0^3$ is replaced by $1/L_0^5$. Therefore, all the results found without the Jacobi transformation (14) remain the same with $\mathbf{n}_i$ instead of $\mathbf{n}$. In particular, Equation (10) transforms into the following

$$ \psi_{\text{inf}} = \frac{1}{\Gamma(k/2)} \sum_{\mathbf{n}} \frac{\Gamma(k/2, \alpha^2 |\mathbf{r} + \mathbf{n}_i|^2)}{|\mathbf{r} + \mathbf{n}_i|^k}. $$

(100)

The Jacobi transformation (14) in a noncubic box has the following form

$$ \sum_{\mathbf{n}_i} \exp(-s|\mathbf{n}_i + r|^2) $$

$$ = \prod_{i=x,y,z} \sum_{n_i} \exp[-s(n_i L_i/L_0 + r_i)^2] $$

$$ = \left[ \prod_{i=x,y,z} \left( \frac{\pi}{s(n_i L_i/L_0)^2} \right)^{1/2} \right] \prod_{i=x,y,z} \exp\left(-\frac{\pi^2 m_i^2}{s(L_i/L_0)^2}\right) $$

$$ \times \exp(2\pi im_i r_i/L_i) $$

$$ = (\pi/s)^{3/2} \sum_{m_i} \exp(-\pi|m_i|^2/s) \exp(2\pi im_i r_i), $$

(101)

with $\mathbf{m}_i = m_x L_x/L_0, m_y L_y/L_0, m_z L_z/L_0$ the normalized displacement vector in momentum space. The last equation is obtained from the original expression (14) by a formal substitution of the vector $\mathbf{m}$ by $\mathbf{m}_k$.

In order to calculate $\psi_{\text{fin}}$ we first modify Equation (15),

$$ \exp(-s|\mathbf{n}_i + r|^2 - t|\mathbf{n}_i + r|^2) = \exp[-(s + t)|\mathbf{n}_i + r|^2], $$

(102)

then insert it into the relation (102), and finally separate the summand $\mathbf{n} = 0$.

$$ \psi_{\text{fin}} = \frac{\pi^{3/2}}{\Gamma(k/2)} \sum_{m_i \neq 0} \int_0^{\alpha^2} \frac{t^{k-1}}{(t + s)^{3/2}} \exp\left[-\frac{\pi^2 m_i^2}{t + s} + 2\pi im_i r_i\right] dt $$

$$ + \frac{\pi^{3/2}}{\Gamma(k/2)} \int_0^{\alpha^2} \frac{t^{k-1}}{(t + s)^{3/2}} dt $$

$$ = \psi_{\text{fin}}^{m \neq 0} + \psi_{\text{fin}}^{m = 0}. $$

(103)

The subsequent derivation follows exactly the derivation for a cubic box, with the change of $\mathbf{n}$ by $\mathbf{n}_i$ and $\mathbf{m}$ by $\mathbf{m}_k$ for sums in the real and momentum spaces, respectively. The final result for a 3D system in a noncubic box can be summarized as follows

$$ \psi(r) = \sum_{\mathbf{n}_i} \frac{\Gamma(k/2, \alpha^2 |\mathbf{n}_i + r|^2)}{\Gamma(k/2)|\mathbf{n}_i + r|^k} $$

$$ + \sum_{m_i \neq 0} \frac{\pi^{3/2} \alpha^{-k-2} \cos(2\pi m_i r_i)}{\Gamma(k/2)} E_{(k-1)/2}\left(\frac{\pi^2 |m_i|^2}{\alpha^2}\right) + C_1 $$

(104)
\[ \xi = \sum_{n_r \neq 0} \frac{\Gamma(k/2, \alpha^2 |n_r|^2)}{\Gamma(k/2) |n_r|^k} + \sum_{m_i \neq 0} \frac{\pi^{3/2} \alpha^{k-3}}{\Gamma(k/2)} E_{(k-1)/2} \left( \frac{\pi^2 |m_i|^2}{\alpha^2} \right) + C_1 + C_2 \]  

(105)

\[ U = \frac{q^2}{L_0^2} \sum_{i<j} \psi(r_{ij}/L_0) + \frac{N q^2}{2 L_0^4} \xi \]  

(106)

with the constants \( C_1 \) and \( C_2 \) defined in (37) and (38). As done in the cubic box, the potential energy may also be given with the momentum space sum (linear in \( N \)). Applying the definitions, similar to Equations (50)–(53),

\[ \tilde{\psi}(r) = \sum_{n_r} R(n_r, r) + C_1, \]  

(107)

\[ \tilde{\xi} = \sum_{n_r \neq 0} \rho(n_r) + C_1 + C_2, \]  

(108)

\[ \tilde{S}_{eq}(m_k) = q_r \sum_j \exp(2\pi i m_j r_j/L), \]  

(109)

\[ \tilde{S}_q(m_k) = \sum_j q_j \exp(2\pi i m_j r_j/L), \]  

(110)

the potential energy for a one-component jellium model converts into

\[ U^1 = \frac{q^2}{L_0^2} \sum_{i<j} \tilde{\psi}(r_{ij}/L_0) + \frac{1}{2 L_0^k} \sum_{m_i \neq 0} \kappa(m_k) |\tilde{S}_{eq}(m_k)|^2 \]  

(111)

with a natural extension to the general case

\[ U^{\text{gen}} = \frac{1}{L_0^2} \sum_{i<j} q_i q_j \tilde{\psi}(r_{ij}/L_0) + \frac{1}{2 L_0^k} \sum_{m_i \neq 0} \kappa(m_k) |\tilde{S}_q(m_k)|^2 \]  

(112)

\[ + \sum_{m_i \neq 0} \frac{q_i^2}{2 L_0^2} \tilde{\xi}. \]

Note that the formulae are derived for an orthogonal basis set. Still many triclinic lattices can be sampled in a similar form. In such cases the crystal is constructed not by translating the smallest-volume unit cell along non-orthogonal vectors but rather by translating a pseudo-unit cell of size \( L_x/2 \times L_y/2 \times L_z \) containing more atoms along orthogonal directions. For example a hcp crystal can be summed in this way. Nonetheless, as the pseudo-elementary cell technique may be inconvenient in application to triclinic lattices, we would suggest the reader to rely on a reciprocal lattice technique (see [51]).

5.2. 2D case

The generalization of the formulae found in a square 2D geometry to a rectangular simulation box comes about in a similar manner. It is sufficient to take the resulting expressions for the two-dimensional problem (62) and (66), and to perform the necessary substitutions \( n_r \rightarrow n_r \) and \( n_i \rightarrow m_k \),

\[ \psi(r) = \sum_{n_r} \frac{\Gamma(k/2, \alpha^2 |n_r + r|^2)}{\Gamma(k/2) |n_r + r|^k} + \sum_{m_i \neq 0} \frac{\pi \alpha^{k-2} \cos (\pi m_i r)}{\Gamma(k/2)} E_{(k-1)/2} \left( \frac{\pi^2 |m_i|^2}{\alpha^2} \right) + \psi_{\text{fin}}^{m=0}, \]  

(113)

\[ \xi = \sum_{n_r \neq 0} \frac{\Gamma(k/2, \alpha^2 |n_r|^2)}{\Gamma(k/2) |n_r|^k} + \sum_{m_i \neq 0} \frac{\pi \alpha^{k-2} |m_i|^2}{\Gamma(k/2)} E_{(k-1)/2} \left( \frac{\pi^2 |m_i|^2}{\alpha^2} \right) + \psi_{\text{fin}}^{m=0}, \]  

(114)

where \( \psi_{\text{fin}}^{m=0} \) is given by the expressions (70), (73) or (74).

For a long-range interaction within the jellium model, the potential energy becomes

\[ U^{\text{gen}} = \frac{q^2}{L_0^2} \sum_{i<j} \psi(r_{ij}/L_0) + \frac{N q^2}{2 L_0^4} \xi \]  

(115)

with the notation

\[ L_0 = (L_x L_y)^{1/2}, \]  

(116)

\[ n_r = n_x L_x/L_0 + n_y L_y/L_0, \]  

(117)

\[ m_k = m_x L_0/L_x + m_y L_0/L_y. \]  

(118)

For a multicomponent gas (quasi-neutral in the case of a long-range potential), the potential energy is

\[ U^{\text{gen}} = \frac{1}{L_0^2} \sum_{i<j} q_i q_j \psi(r_{ij}/L_0) + \sum_{m_i \neq 0} \frac{q_i^2}{2 L_0^2} \tilde{\xi}. \]  

(119)
Finally, the usual modification to calculate the momentum space sum linearly in $N$ is given by

$$U^{\text{jel}} = \frac{q^2}{L_0^2} \sum_{i<j} \tilde{\psi}(r_{ij}/L_0) + \frac{1}{2L_0^2} \sum_{m \neq 0} \kappa(m_k) |\tilde{S}_\text{equal}(m_k)|^2$$

$$+ \frac{Nq^2}{2L_0^2} \tilde{\xi},$$

(120)

$$U^{\text{gen}} = \frac{1}{L_0^2} \sum_{i<j} q_i q_j \tilde{\psi}(r_{ij}/L_0) + \frac{1}{2L_0^2} \sum_{m \neq 0} \kappa(m_k) |\tilde{S}_\text{q}(m_k)|^2$$

$$+ \sum q_i^2 \tilde{\xi},$$

(121)

with $\tilde{\psi}, \tilde{\xi}, \tilde{S}_\text{equal}, \tilde{S}_\text{q}$ defined by (108)–(111) in their corresponding two-dimensional variants.

Some of non-orthogonal lattices can be sampled using the concept of pseudo-unit cell. For example a triangular lattice is constructed by translation of a single atom along two vectors with a 60° angle between them. The same filling can be obtained by translation of a rectangular pseudoelementary cell with two atoms which can be readily calculated with the presented formulae.

6. Equation summary

In the previous sections, we have derived general expressions of the Ewald sums for polytropic $1/|r|^k$ potentials in three- two- and one-dimensional systems. For integer values of $k$, the polytropic potential reduces to a power-law interaction, which comprises realizations of high physical relevance. Integer power-law potentials include

- $k = 1$ – Coulomb $1/|r|$ interaction;
- $k = 2$ – Calogero–Sutherland $1/|r|^2$ interaction;
- $k = 3$ – isotropic $1/|r|^3$ component of dipole–dipole interaction (one-dimensional systems; two-dimensional system of dipole oriented perpendicularly to the plane);
- $k = 4, 5, 6$ – interaction between different Rydberg atoms;
- $k = 6, 12$ – van der Waals interaction.

The expressions for the potential energy for both the jellium model and the general case of a charge-neutral simulation cell are the following

$$U^{\text{gen}} = \frac{1}{L_0^2} \sum_{i<j} q_i q_j \tilde{\psi}(r_{ij}/L_0) + \frac{1}{2L_0^2} \sum_{m \neq 0} \kappa(m_k) |\tilde{S}_\text{q}(m_k)|^2$$

$$+ \sum q_i^2 \tilde{\xi},$$

(122)

$$U^{\text{jel}} = \frac{q^2}{L_0^2} \sum_{i<j} \tilde{\psi}(r_{ij}/L_0) + \frac{Nq^2}{2L_0^2} \tilde{\xi},$$

(123)

$$\psi(r) = \sum_{n} R(n, r) + \sum_{m \neq 0} K(m_k, r) + C_1,$$

$$\xi = \sum_{n \neq 0} \rho(n_r) + \sum_{m \neq 0} \kappa(m_k) + C_1 + C_2,$$

$$R(n, r) = \rho(n + r),$$

$$K(n, r) = \kappa(m) \cos(2\pi mr),$$

$$C_1^{3D} = \begin{cases} 2\pi^{3/2} \alpha^k \sqrt{\pi} \frac{3}{2} & \text{if } k \neq 3, \\ \frac{4\pi}{(k - 3)!} \frac{1}{\sqrt{2\pi k}} & \text{if } k = 3, \end{cases}$$

$$C_1^{2D} = \begin{cases} 2\pi \alpha^{k-2} \frac{3}{2} & \text{if } k \neq 2, \\ \frac{2\pi \alpha}{(k - 2)!} \frac{1}{\sqrt{2\pi k}} & \text{if } k = 2, \end{cases}$$

$$C_2 = -\frac{\alpha^k}{(2\pi)^k} \frac{3}{2},$$

$$L_0 = \begin{cases} (L_x, L_y, L_z)^{1/3} & \text{in 3D}, \\ (L_x, L_y)^{1/2} & \text{in 2D}, \end{cases}$$

$$n_r = (n \cdot L)/L_0, \quad \text{with } L = (L_x, L_y, L_z),$$

$$m_k = (m \cdot L')L_0, \quad \text{with } L' = (1/L_x, 1/L_y, 1/L_z).$$

(124)–(133)

Alternatively, by performing a momentum space sum the above set of equations becomes

$$U^{\text{gen}} = \frac{1}{L_0^2} \sum_{i<j} q_i q_j \tilde{\psi}(r_{ij}/L_0) + \frac{1}{2L_0^2} \sum_{m \neq 0} \kappa(m_k) |\tilde{S}_\text{q}(m_k)|^2$$

$$+ \sum q_i^2 \tilde{\xi},$$

(134)

$$U^{\text{jel}} = \frac{q^2}{L_0^2} \sum_{i<j} \tilde{\psi}(r_{ij}/L_0) + \frac{Nq^2}{2L_0^2} \tilde{\xi},$$

(135)

$$\tilde{\psi}(r) = \sum_{n} R(n, r) + C_1,$$

$$\tilde{\xi} = \sum_{n \neq 0} \rho(n_r) + C_1 + C_2,$$

$$\tilde{S}_\text{equal} = \sum_{j} q_j \exp(2\pi im_r L),$$

$$\tilde{S}_\text{q} = \sum_{j} q_j \exp(2\pi im_r L).$$

(136)–(139)
Table 1. Coefficients $\rho(n)$ and $\kappa(n)$ taken from Equations (36) and (18) for 3D geometry. LR and SR stand for long range and short range, respectively.

|        | $\rho(n)$                                                                 | $\kappa(m)$                                                                 |
|--------|---------------------------------------------------------------------------|---------------------------------------------------------------------------|
| LR $\frac{1}{r}$ | $\text{erfc}(\alpha|n|) \frac{|n|}{r}$                                  | $\frac{1}{\pi m^2} \exp\left(-\frac{\pi^2 m^2}{\alpha^2}\right)$          |
| LR $\frac{1}{|n|^2}$ | $\exp(-\alpha^2 n^2) \frac{n^2}{|n|^2}$                              | $\frac{\pi}{|m| \text{erfc} \frac{\pi m}{\alpha}}$                        |
| SR $\frac{1}{r}$ | $\frac{\alpha^2 n^2 + 1}{n^4} \exp(-\alpha^2 n^2)$                    | $\frac{2\pi^{1/2}}{\pi^2} \alpha^2 \exp\left(-\frac{\pi^2 m^2}{\alpha^2}\right) - \pi^2 |m| \text{erfc} \frac{\pi |m|}{\alpha}$ |
| SR $\frac{1}{|n|^2}$ | $\text{erfc}(\alpha |n|) + \frac{4 \exp(-\alpha^2 n^2)}{|n|^2} \left( \frac{3|n|}{2} + (\alpha |n|)^3 \right)$ | $\frac{4 \pi \alpha^2}{3} \left( e^{-\frac{\pi^2 m^2}{\alpha^2}} - \frac{\pi^2 m^2}{\alpha^2} E_i \left( \frac{\pi^2 m^2}{\alpha^2} \right) \right)$ |
| SR $\frac{1}{r^2}$ | $\frac{\alpha^4}{n^4} + \frac{2 \alpha^2}{n^2} + \frac{1}{n^4} \exp(-\alpha^2 n^2)$ | $\frac{8 \pi^{1/2} \alpha^2}{9} \left( \exp\left(-\frac{\pi^2 m^2}{\alpha^2}\right) - \frac{2 \pi^2 m^2}{\alpha^2} + \frac{2 \pi^{1/2} \alpha^2}{\alpha^3} \text{erfc} \frac{\pi |m|}{\alpha} \right)$ |
| SR $\frac{1}{|n|^4}$ | $\sum_{i=0}^{\infty} \frac{10}{|i|} \text{erfc}(-\alpha^2 n^2) \frac{n^2}{|n|^4}$ | $\frac{2 \pi^2 m^2}{\alpha^2} \left( e^{-\frac{\pi^2 m^2}{\alpha^2}} \left( 1 - \frac{2 \pi^2 m^2}{\alpha^2} \right) + \frac{2 \pi^{1/2} \alpha^2}{\alpha^3} \text{erfc} \frac{\pi |m|}{\alpha} \right)$ |

Table 2. The coefficients $\rho(n)$ and $\kappa(m)$ taken from Equations (36) and (76) for 2D geometry. LR and SR stand for long range and short range, respectively.

|        | $\rho(n)$                                                                 | $\kappa(m)$                                                                 |
|--------|---------------------------------------------------------------------------|---------------------------------------------------------------------------|
| LR $\frac{1}{|r|}$ | $\text{erfc}(\alpha|n|) \frac{|n|}{|r|}$                                   | $\frac{1}{\pi m^2} \text{erfc} \frac{\pi m}{\alpha}$                      |
| SR $\frac{1}{|r|}$ | $\frac{2 \alpha^2}{|n|^2} \exp(-\alpha^2 n^2) + \frac{\text{erfc}(\alpha|n|)}{|n|^2}$ | $\frac{4 \pi \alpha^2}{3} \left( e^{-\frac{\pi^2 m^2}{\alpha^2}} - \frac{\pi^2 m^2}{\alpha^2} E_i \left( \frac{\pi^2 m^2}{\alpha^2} \right) \right)$ |
| SR $\frac{1}{|n|^2}$ | $\alpha^2 n^2 + 1 \frac{n^2}{|n|^2} \exp(-\alpha^2 n^2)$                       | $\frac{8 \pi^{1/2} \alpha^2}{9} \left( \exp\left(-\frac{\pi^2 m^2}{\alpha^2}\right) - \frac{2 \pi^2 m^2}{\alpha^2} + \frac{2 \pi^{1/2} \alpha^2}{\alpha^3} \text{erfc} \frac{\pi |m|}{\alpha} \right)$ |
| SR $\frac{1}{|n|^4}$ | $\frac{\alpha^4}{n^4} + \frac{2 \alpha^2}{n^2} + \frac{1}{n^4} \exp(-\alpha^2 n^2)$ | $\frac{2 \pi^2 m^2}{\alpha^2} \left( e^{-\frac{\pi^2 m^2}{\alpha^2}} \left( 1 - \frac{2 \pi^2 m^2}{\alpha^2} \right) + \frac{2 \pi^{1/2} \alpha^2}{\alpha^3} \text{erfc} \frac{\pi |m|}{\alpha} \right)$ |
| SR $\frac{1}{|r|}^2$ | $\sum_{i=0}^{\infty} \frac{10}{|i|} \text{erfc}(-\alpha^2 n^2) \frac{n^2}{|n|^4}$ | $\frac{2 \pi^2 m^2}{\alpha^2} \left( e^{-\frac{\pi^2 m^2}{\alpha^2}} \left( 1 - \frac{2 \pi^2 m^2}{\alpha^2} \right) + \frac{2 \pi^{1/2} \alpha^2}{\alpha^3} \text{erfc} \frac{\pi |m|}{\alpha} \right)$ |

In accordance with considerations discussed in preceding sections, the simulation cell has to fulfill the charge neutrality condition ($\sum_{i=1}^{N} q_i = 0$) for long-range potentials. Also, notice that in the particular case of a cubic simulation cell, $n_c = n, m_c = m$.

Explicit expressions of the coefficients $\rho(n)$ and $\kappa(m)$ for the most relevant interactions are summarized for 3D and 2D systems in Tables 1 and 2, respectively.

The expressions of the Ewald summation components are consistent in appropriate limits with the earlier published results by Mazars [41,52] (inverse power-law potentials in 2D and 3D geometries), by Mora et al. [36] (2D dipolar bosons), by Karasawa and Goddard [42] and Ou-Yang et al. [43] (Lennard-Jones potential).

7. Practical application and optimizations in the Ewald technique

7.1. Optimization scheme with $N^2$ dependence

The basic idea of the Ewald method is to calculate slowly decaying sums in a rapid manner by means of the Fourier transform of the slowly converging part.
Although conceptually it provides an exact result, the number of terms which has to be summed in order to reach the needed convergence is a priori unknown. Once we choose the interaction potential, this fixes the exact form of the sums to calculate, and the practical remaining question is the proper choice of the free parameter $\alpha$ and the numbers of terms to be calculated in the sums, originated from coordinate and momentum spaces. The total computer time $T$ is obtained from the time needed to evaluate different sums

$$T = (t_r N_r + t_k N_k) N^2 / 2,$$

(140)

with the constants $t_r$ and $t_k$ depending on the complexity of the coefficients in the sums and the factor $N^2 / 2$ approximating the number of pairs. Here $N_r$ and $N_k$ are numbers of terms which are summed for each pair $r_{ij}$, in particular the case $N_r = 1$ corresponds to the so-called minimum image convention. One can notice that $t_k$ is usually much less then $t_r$, since in the Jacobi-transformed sum we only calculate cosine functions, which is generally far less time-consuming than the complicated functions appearing in $R$. It is clear that the parameter $\alpha$ affects only the resulting error in the energy. In fact, the value of $\alpha$ being very small or very large eliminates errors in one of the sums, but amplifies them in the other, so there is an ‘optimal’ point for $\alpha$, yielding a minimum error in the total energy.

In the following, we discuss a way for error ($\delta E$) minimization assuming the calculation time $T$ fixed. From our point of view, a useful approach for practical implementation is represented by the following scheme

- We determine a time law $T = (t_r N_r + t_k N_k) N^2 / 2$ in a preliminary calculation and fix the values of $t_r$ and $t_k$.
- We take a set of configurations, corresponding to the equilibrated state using an initial Ewald summation. Then, we calculate the exact energies $E_{\text{ex}}$ (as a converged result of the Ewald summation) and the energies $E(\alpha, N_r, N_k)$ biased by a choice of $N_r$ and $N_k$. For each pair $(N_r, N_k)$, we find an optimal value of $\alpha = \alpha_{\text{opt}}(N_r, N_k)$.
- We choose the goal accuracy $\delta E_{\text{acc}}$ (normally, well below the statistical error). We plot the error as a function of the computer time spent and choose the less time consumption case among the points that lie below $\delta E_{\text{acc}}$, therefore obtaining all the parameters required: $\alpha$, $N_r$, and $N_k$. From now on, these parameters are used in actual simulations.

![Figure 1. Dependence of the calculation time $T$ on the number of terms $N_r$ in the coordinate space for fixed numbers of terms in the momentum space $N_k = 5, 9, 25, 45$.](image)

### 7.2. Example of optimization

Let us illustrate the scheme proposed in the preceding subsection taking as an example the problem of a two-dimensional zero-temperature Bose gas of particles, interacting through the pairwise repulsive $C_3 / |r|^3$ potential. The model corresponds to the dipole–dipole interaction with all dipole moments aligned perpendicularly to the plane of motion. The simulation is performed with $N = 108$ particles in a quadratic box ($L_x = L_y = L = 1$). The dimensionless Hamiltonian in the present example is taken to be $\hat{H} = - \sum_{i=1}^{N} \nabla_i^2 / 2 + \sum_{i<j} 1 / |r_{ij}|^3$ with the unit of energy being $\hbar^2 / m L^2$. To describe the ground-state properties of the system we use the variational Monte Carlo (VMC) method and a Jastrow wave function with a two-body correlation factor which is the solution of the two-body scattering problem [53].

The optimization is done by averaging over $N_{\text{conf}} = 50$ uncorrelated VMC configurations, sampled according to the chosen probability distribution. We define the error $\delta E(\alpha)$ as a sum over $N_{\text{conf}}$ configurations of the difference of the Ewald energy $E(\alpha; \xi_{\text{conf}}, \alpha, N_r, N_k)$, calculated for a given set of parameters ($\alpha, N_r, N_k$) and the converged energy $E_{\text{ex}}(\xi_{\text{conf}}) = \lim_{N_k \to \infty} \lim_{N_r \to \infty} E(\xi_{\text{conf}}, \alpha, N_r, N_k)$. The dependence of the computer time $T$, needed for the evaluation of Ewald sums, on the parameter set is shown in Figures 1 and 2. In Figure 1, we show the dependence of $T$ on the number of terms $N_r$ in real space for different fixed numbers of terms $N_k$ in the momentum space. The computation time is proportional to the number of terms and the resulting dependence is linear in $N_r$. A fixed number of terms $N_k$ requires a certain amount of calculations which results in a constant shift.
Similarly, keeping \( N_r \) fixed and varying \( N_k \) produces a linear dependence in \( N_k \) with a constant shift which depends on \( N_r \), as shown in Figure 2.

As one sees in Figures 1 and 2, the time dependence is linear both on \( N_k \) and \( N_r \), although the point corresponding to \((0,0)\) in \((N_r, N_k)\) does not necessarily give \( T = 0 \), since the reported time also contains some initializing calculations. The total error in the potential, as defined above, is given by

\[
\delta E(\alpha) = \left( \sum_{i=1}^{N_{\text{conf}}} \frac{(E(\alpha, i_{\text{conf}}) - E_{\text{ex}}(i_{\text{conf}}))^2}{N_{\text{conf}}} \right)^{1/2}. \tag{141}
\]

According to our previous considerations, in the case of very small or very large values of \( \alpha \) the error coming from one of the two sums, that is in the real or momentum space, grows and dominates over the error coming from the other sum; for a certain ‘optimal’ range of \( \alpha \) these two errors are of the same order. Notice that for each particular configuration, and each pair \((N_r, N_k)\), it is possible to find \( \alpha_{\text{opt}}(i_{\text{conf}}) \), such that \( E(\alpha_{\text{opt}}(i_{\text{conf}}), i_{\text{conf}}) - E_{\text{ex}}(i_{\text{conf}}) = 0 \). Instead, our task is to obtain a ‘universal’ parameter \( \alpha_0 \), minimizing the total error (142). The mean over the configuration set of the biased energies \( \overline{E}(\alpha_0, i_{\text{conf}}) \) is used as an estimation for the mean of the exact energies \( \overline{E}_{\text{ex}} \), introducing an inevitable systematic error. As it appears in typical calculations, this error is at least one order of magnitude smaller than the statistical error (142) given by the minimization of \( \delta E(\alpha) \). In our benchmark calculations we also checked the dependence of the total energy on the value \( \alpha \) for different pairs \((N_r, N_k)\), which revealed characteristic plateaus for certain ranges of \( \alpha \) (of the order of 1). It means that the Ewald summation indeed converges fast to a universal result. Nevertheless, the optimum value of the parameter \( \alpha \), minimizing the cumulative error, depends on the cut-off numbers in the sums in the range from \( \alpha = 1.45 \) \((N_r = 21, N_k = 5)\) to \( \alpha = 5.25 \) \((N_r = 1, N_k = 45)\).

A second step is the study of the dependence of the error and time on different pairs \((N_r, N_k)\). The calculation time can be split as the sum of times for summing up in coordinate and momentum spaces, \( T = (N_r t_r + N_k t_k) N_r^2 / 2 \) as in Equation (141), with \( N_r, N_k \) being the numbers of terms in each sum for a single pair. Every one of these sums converges when \( N_r, N_k \to \infty \) to a certain value, depending on \( \alpha \), while the sum of the limiting values is a constant. We can take into account the errors, corresponding to each of the sums separately. For \( \alpha \to 0 \) the error for the real space term is zero and the other one tends to infinity (and vice versa as \( \alpha \to \infty \)). The minimum total error should therefore correspond to the value of \( \alpha \), satisfying the relation \( d(\delta E_r + \delta E_k) / d\alpha = 0 \).

Focusing on the 2D system of our example, we note that the long-range expansions of the terms in (36) and (76) are similar, in a sense that the leading terms in both expressions are Gaussians,

\[
\frac{\Gamma(k/2, \alpha^2 n^2)}{\Gamma(k/2) m^k} = \exp(-\alpha^2 n^2) \left[ \frac{C_r}{n^2} + O \left( \frac{1}{m^3} \right) \right]. \tag{142}
\]

\[
\frac{\pi^{3/2} q^{k-3}}{\Gamma(k/2)} \frac{E(k-1) / \alpha^2}{\pi^2 m^2} \left[ \frac{C_k}{m^2} + O \left( \frac{1}{m^3} \right) \right]. \tag{143}
\]

The power-law terms in \( n, m \) and the constants \( C_r, C_k \) may be neglected since the leading behaviour is driven by the Gaussian. The cut-off errors due to finite numbers of elements in the sums can be evaluated by ignoring the discrete structure of the images and approximating the sums by uniform integrals,

\[
\delta E = \frac{N_r^2}{2} \int_{R}^\infty \exp(-\alpha^2 r^2) 2\pi r \, dr + \frac{N_k^2}{2} \int_{K}^\infty \exp(-\frac{\pi^2 K^2}{\alpha^2}) 2\pi k \, dk
\]

\[
= \frac{\pi N_r^2}{2} \exp(-\frac{\alpha^2 N_r/\pi}{\alpha^2}) + \frac{\alpha^2}{\pi^2} \exp(-\frac{\pi N_k / \alpha^2}{\alpha^2}) \tag{144}
\]

with \( R \approx (N_r/\pi)^{1/2} \) and \( K \approx (N_k/\pi)^{1/2} \) the approximate cut-off lengths in real and momentum spaces, respectively. The optimal value for \( \alpha \) can be obtained by solving the equation \( d\delta E_r / d\alpha = -d\delta E_k / d\alpha \). The first-order approximation of this equation is found by
taking logarithms of both sides and omitting constants and terms, depending on $\alpha$ logarithmically, that is

$$A_k/\alpha^2 - A_r/\alpha^2 = 0$$  \hspace{1cm} (145)

with $A_k = \pi N_k$ and $A_r = N_r/\pi$, which yields

$$\alpha = (A_k/A_r)^{1/4} = (\pi^2 N_k/N_r)^{1/4}. \hspace{1cm} (146)$$

Then, at lowest order one finds (145),

$$\delta E \sim \frac{N^2}{2} \exp(-\alpha^2 N_r/\pi) = \frac{N^2}{2} \exp(-(N_k N_r)^{1/2}). \hspace{1cm} (147)$$

Since the calculation time is linear with the numbers of elements $N_r$ and $N_k$, we may conclude that with $N_k$ fixed and comparatively large $N_r$, $\ln(\delta E) \sim (N_r)^{1/2} \sim T^{1/2}$ and vice versa, with $N_r$ fixed and large $N_k$, $\ln(\delta E) \sim N_k^{1/2} \sim T^{1/2}$. This power law may be easily checked in our calculations, as is shown in Figure 3. Note that for the obtained value of $\alpha$ the errors of the real- and momentum-space cut-offs are of the same order of magnitude, that is $\delta E_r \approx \delta E_k$, which may serve as a rough criterion to optimize the parameter $\alpha$.

### 7.3. Optimization scheme with $N^{3/2}$ dependence

A more advanced procedure for optimization of the parameters, proposed by Perram et al. [8], yields an asymptotic scaling $N^{3/2}$, with $N$ the number of particles. It is based on the form of Ewald summation with the rearranged momentum space sum, linear in $N$ (135). Note that here the linear $N$ dependence is obtained after a summation over particle coordinates, while in Sections 7.1 and 7.2 the momentum space sum was evaluated over the pairs of the particles and had $N^2$ dependence. Suppose the values of the calculation time $t_r$, $t_k$ to perform unit computations in both sums are known and the target error level $\exp(-p)$ is fixed. Then, the total execution time in the real and momentum spaces is

$$T = T_r + T_k = N^2 \pi R^2 t_r + N \pi R^2 t_k \hspace{1cm} (148)$$

with $p = \alpha^2 R^2 = \pi^2 K^2/\alpha^2$. Expressing $K$ as $K = p/(\pi R)$ we can see that the minimum of the total time $T$ corresponds to

$$R_{\text{opt}} = \left(\frac{p}{\pi}\right)^{1/2} \left(\frac{t_k}{t_r}\right)^{1/4} N^{-1/4}, \hspace{1cm} (149)$$

$$K_{\text{opt}} = \left(\frac{p}{\pi}\right)^{1/2} \left(\frac{t_k}{t_r}\right)^{-1/4} N^{1/4}, \hspace{1cm} (150)$$

$$\alpha_{\text{opt}} = \pi^{1/2} \left(\frac{t_k}{t_r}\right)^{-1/4} N^{1/4}. \hspace{1cm} (151)$$

The computation time is equally divided between the real and momentum space parts (this was also stated in our simple optimization scheme), with a scaling of the whole summation given by

$$T = 2N^2 \pi R^2 t_r = 2p(t_r t_k)^{1/2} N^{3/2}. \hspace{1cm} (152)$$

Notice that the values of the free parameters change very slowly when the simulation cell is enlarged, and in particular $\alpha$ is not affected by the choice of the precision. Similar formulae for the optimized parameters in three-dimensional systems, with a discussion of different techniques to improve performance of the Ewald summation, are given by Fincham [54]. A more precise and detailed analytic study of the cut-off errors with verifications of the analytic results in actual calculations can be found in the work of Kolafa and Perram [55]. An optimized method for treating the truncation error in Ewald sums with generic potentials was proposed by Natoli and Ceperley [56]. While the needed CPU time scales as $O(N \ln N)^{3/2}$, it was shown that in the example of the Coulomb potential the method resulted in greatly improved accuracy compared to that of standard Ewald technique for a comparable computational effort. This method is based on an expansion of the real space function in an arbitrary radial basis with a parametric set of numbers in place of the $k$-dependent prefactors of $\exp(2\pi i n r)$. The subsequent minimization of $\chi^2$ with respect to the whole set of parameters yields a final optimal solution, that is the real space expansion coefficients and the $k$-space factors. This technique was also applied to derive the optimized summation
formulae for the two-dimensional Coulomb system [57].

In general, the unit computation time in momentum space is 2–4 times faster than the one in real space. Taking the following reasonable assumptions \( p = 4\pi, t_1/t_2 = 3, \) we find \( R_{\text{opt}} \approx 2.6/N^{1/4} \) (the box size is taken to be 1). We want \( R \) to be below 0.5, since in this case the summation in the real space reduces to the accumulation of the single component \( n = 0 \) (‘minimum image convention’). This condition \( R_{\text{opt}} = 0.5 \), with our previous assumptions, corresponds to

\[
N_{\text{opt}} = 770, \quad K_{\text{opt}} = 8.0, \quad \alpha_{\text{opt}} = 7.1.
\]  

(153)

In smaller systems, the other components of the real sum, starting from \( |n| = 1 \), should be considered.

It is worth pointing out that if the interaction is very strong at short distances (as for the Lennard-Jones potential), then in principle the real-space cut-off \( R \) can be chosen below the ‘hard core radius’ with a large enough value of \( \alpha \). This leads to the possibility of dropping completely the real-space part of the total sum and treating the \( k \)-space only. This can be advantageous in different aspects, especially with the current progress in the development of efficient FFT-based methods. Nonetheless, we are not aware of any present application of a similar technique.

8. Conclusions

In the present work, we have applied the Ewald summation method to \( 1/|r|^s \) polytropic potentials in three-, two- and one-dimensional geometries in a simulation box with periodic boundary conditions. We have found the explicit functional forms for all the components of the sums in both real and momentum spaces, with special attention being paid to the cases of long-range interactions, that is conditionally convergent or divergent potentials (corresponding to \( k < D \), with \( D \) standing for the dimensionality), ‘marginal’ interactions (\( k = D \)), and short-range interactions (with \( k > D \)). For the latter case of short-range interaction potentials, where in principal a straightforward summation of the initial sum (1) is possible, the Ewald method is shown to be useful, as it yields the faster (Gaussian) convergence rate. A condition of charge neutrality of the simulation cell is stated to be necessary for conditionally convergent and divergent potentials; a homogeneous positive charge background (‘jellium’ model) is introduced as the most relevant and frequently used kind of neutralization. The conditionality of the convergence for a charge-neutral system, governed by the Coulomb interaction, is discussed with a justification of the use of a specific periodicity-preserving convergence factor. The derivation technique, presented in our work, is consistent with the arguments of de Leeuw et al. [2].

The results are first presented for the case of a 3D system in a cubic simulation box in order to explain the general mathematical procedure, which for the specific case of the Coulomb potential recovers well-known results [58]. Later on, the same mathematical technique is applied to 2D and 1D geometries. For the one-dimensional case the initial sum for the potential energy is explicitly evaluated (95), nonetheless the Ewald summation is developed for this case too and may be used as a mathematical equality. The special representations of the reciprocal space sums, linear in the number of particles \( N \) and hence more efficient in actual modelling, are presented for 3D and 2D systems. The explicit expressions for the terms of the Ewald sums are given in a tabular form for physically relevant potentials with small integer power indexes \( k \), as dipole–dipole interaction potential, Lennard-Jones potential and others in both three- and two-dimensional geometries (see Tables 1 and 2).

When the simulation box cannot be chosen as cubic, for example in a modelling of a three-dimensional hcp crystal structure, the Ewald method can also be applicable after certain modifications. Formally, it consists of the choice of an appropriate rectangular simulation box and a substitution of the vector \( r \) by \( r_0 = (n_x L_x + n_y L_y + n_z L_z)/L_0 \) and \( m_k = (m_x/L_x + m_y/L_y + m_z/L_z)L_0 \) in the real and momentum space sums, respectively [see (101) and (102)].

The optimization of the involved parameters, that is cut-off numbers in both sums and the integration parameter \( \alpha \), is a necessary operation in order to improve the convergence rates and avoid excessive calculations. The main idea of the optimization, proposed in the present work, is to perform a benchmark calculation, minimizing the variance of the result.

A particular example of the application of the technique is presented for a calculation of the potential energy of a two-dimensional gas of dipoles, aligned perpendicular to the plane of motion. This practical optimization technique is thought to be efficient for stationary and nearly uniform systems that appear, for instance, in Monte Carlo simulations. In spite of being very simple, it allows one to find rather quickly adequate parameter ranges. The analytical estimations of the parameters are given as well and are proven to be consistent with the results, obtained in our method. A more sophisticated method to optimize the calculation parameters, taking advantage of the \( \text{O}(N) \) representation of the Fourier transform sum, is also presented with explicit estimations of the parameters.
for a typical system simulated by Quantum Monte Carlo methods.

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Appendix

We prove that the sums $S_{-+}$ and $S_{+-}$ (45)-(46) vanish on average, allowing one to calculate the potential energy over the negatively charged particles’ positions only.

- First, let us show that the integral of $\psi$ over the cell is zero. Since the distances are in units of $L$, ...
consider the cubic cell $\mathbf{\Omega}=(x, y, z)\in[-1/2, 1/2]^3$, that yields
\[
\int_\mathbf{\Omega} \psi(r) \, dr = J_1 + J_2 + C_1,
\] (154)
where
\[
J_1 = \int_\mathbf{\Omega} \, dr \sum_n R(n, r),
\] (155)
\[
J_2 = \int_\mathbf{\Omega} \, dr \sum_{m \neq 0} K(m, r).
\] (156)

It can be easily seen that the second integral $J_2$ is zero,
\[
J_2 = \sum_{m \neq 0} \kappa(m, r) \int_\mathbf{\Omega} \cos(2\pi m r) \, dr \\
= \sum_{m \neq 0} \kappa(m) \frac{\sin(2\pi m_1 + m_2 + m_z))}{(2\pi)^3 m_1 m_2 m_z} = 0.
\] (157)

• As far as the integral $J_1$ is concerned, we can notice that the regions $\mathbf{\Omega}(n)=\mathbf{r}+\mathbf{n}$, where $\mathbf{r} \in \mathbf{\Omega}$, $\mathbf{n} \in \mathbb{Z}^3$ are the same cubic unit cells, displaced by an integer vector, thus covering all the coordinate space with only zero-measure intersections. It means that the summation of the integrals in (156) over the cell $\mathbf{\Omega}$ can be substituted by the integration over the whole coordinate space,
\[
J_1 = \sum_n \int_\mathbf{\Omega}(n) \, R(n, r) \, dr = \sum_n \int_\mathbf{\Omega}(n) \frac{\Gamma(k/2, \alpha^2 |r + \mathbf{n}|^2)}{\Gamma(k/2)} \, dr \\
= a^{k-3} \int_{\mathbf{R}^3} \frac{\Gamma(k/2, \rho^2)}{\Gamma(k/2)\rho^k} \, d\rho = -2\pi^{3/2}a^{k-3} \frac{G[k/2]}{(k-3)G[k/2]} = -C_1,
\] (158)
and thus the whole integral (155) is equal to zero.

• Consider two species of the particles: negative charges $q_i$ on positions $\mathbf{r}_i$ and a positively charged and uniformly distributed background with a total charge $q_+N_+=-q_-N_-$, ensuring the neutrality of the cell. Let us demonstrate that $S_{++}$ is equal to zero, when the number of background charges tends to infinity. In this case the sum (45) for $S_{++}$ may be rewritten as an integral over the background charges' positions
\[
S_{++} = \sum_i q_i \int_\mathbf{\Omega} \psi(\mathbf{r}_i - \mathbf{r}) \, d\mathbf{r}_p = \sum_i q_i \int_\mathbf{\Omega} \psi(\mathbf{r}) \sigma \, d\mathbf{r},
\] (159)
where we did the change of variables $\mathbf{r} = \mathbf{r}_p - \mathbf{r}_i$. The regions $\mathbf{\Omega}$ and $\mathbf{\Omega}_i$ refer to the original simulation cell and the same cell, moved by the vector $\mathbf{r}_i$, and $\sigma$ stands for the background charge density $\sigma = -q_+N_i/V(\mathbf{\Omega})$. It is clear that every vector $\mathbf{r}=(x, y, z) \in \mathbf{\Omega}_i$ can be displaced into the cell $\mathbf{\Omega}$ by the corresponding shift $\mathbf{\tilde{r}} = (\mathbf{x}, \mathbf{\tilde{y}}, \mathbf{\tilde{z}}) = (x-aL, y-bL, z-cL) \in \mathbf{\Omega}$ with integers $a, b, c$. The Jacobian $J$ of the change of variables $\mathbf{r} \to \mathbf{\tilde{r}}$ is obviously 1. On the other hand, due to the periodicity of $\psi$, $\psi(\mathbf{r}) = \psi(\mathbf{\tilde{r}})$, and $\tilde{r}$ runs over the whole region $\mathbf{\Omega}$ due to the conservation of the volume with $J=1$. Finally, Equation (160) can be written as
\[
S_{++} = \sum_i q_i \int_\mathbf{\Omega} \psi(\mathbf{\tilde{r}}) \sigma \, d\mathbf{\tilde{r}} = 0.
\] (161)

• In a similar manner, the interaction between the charges of the background $S_{++}$ in the limit $N_+ \to \infty$ is given by the double integral
\[
S_{++} = \frac{1}{2} \int_\mathbf{\Omega} \int_\mathbf{\Omega} \psi(\mathbf{r}_1 - \mathbf{r}_2) \sigma^2 = 0.
\] (162)

Since $\int_\mathbf{\Omega} \psi(\mathbf{r}_1 - \mathbf{r}_2) \, d\mathbf{r}_2 = 0$, following the same arguments as for the case of $S_{--}$. 