Convergence peculiarities of lattice summation upon multiple charge spreading generalizing the Bertaut approach

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Abstract. Within investigating the multiple charge spreading generalizing the Bertaut approach, a set of confined spreading functions with a polynomial behaviour, but defined so as to enhance the rate of convergence of Coulomb series even upon a single spreading, is proposed. It is shown that multiple spreading is ultimately effective especially in the case when the spreading functions of neighbouring point charges overlap. In the cases of a simple exponential and a Gaussian spreading functions the effect of multiplicity of spreading on the rate of convergence is discussed along with an additional optimization of the spreading parameter in dependence on the cut-off parameters of lattice summation. All the effects are demonstrated on a simple model NaCl structure.

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1. Introduction

It is known that the Ewald approach [1] is the most widespread implementation of the Poisson summation formula in crystals [2, 3, 4, 5]. Bearing in mind that such a treatment proposes that the overall summation is divided into sums over direct and reciprocal space, the enhancement of the rate of convergence is traditional in this problem and assumes the investigation of ranges of summation in either of those sums. Indeed, Epstein [6] discussing this problem for the first time has proposed that both the sums must be separated by a dimensionless parameter unity, in agreement with conventional mathematical approaches. According to Ewald [1], the corresponding parameter of splitting can still be chosen as a unique, but variable, so as to provide the most rapid rate of convergence of both the sums. This proposal remained appreciable for a long time [7]. However, in the last years one more standpoint arises, keeping in mind that the splitting parameter can also depend on the cut-off parameters of both the summations in question [8, 9]. In particular, such a treatment can reduce the computational efforts associated with the dimension of a supercell employed in molecular dynamics [10, 11].

It is important that the Ewald approach can be regarded as an effect of charge spreading with a Gaussian spreading function [1, 3, 11]. In this connection, the Bertaut treatment [12] is obviously the extension of the Ewald one to an arbitrary spreading
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It implies that the problems of adjustable parameters of splitting still remain actual in this generalized treatment as well \[8, 13\]. Of course, these problems become immaterial if the spreading functions applied to different point charges in a point-charge lattice do not overlap \[12, 14\]. In this case the sum over direct space is absent and so the error arising upon truncating the summation over reciprocal space is the only subject of interest \[15, 16, 17, 18, 19\].

It is worth noting that the charge spreading with a certain spreading function applied to all couples of interacting charges in the lattice was originally discussed \[12, 14\]. On the other hand, it turns out that spreading the charges generating the potential field is sufficient for enhancing the rate of convergence of the lattice series \[13, 20, 21\]. Nevertheless, the application of spreading to all the charges in the expression for the Coulomb energy appears to be somewhat more efficient \[14\]. The reason of this efficiency can be understood from the fact that, by symmetry, the latter effect may be treated as a double spreading of charges generating the potential field. As a result, the idea of a multiple charge spreading arises as a next step towards achieving the faster convergence \[22\].

In the present paper the problem of enhancing the rate of convergence is discussed in detail. We consider different principal classes of spreading functions, with concentrating our attention on the effects of multiple spreading. As particular cases of spreading functions extended to infinity, here we discuss a Gaussian spreading function and a simple exponential spreading. Furthermore, we examine different types of confined spreading functions in a sequence of enhancing their convergence efficiency, providing that the possibility of their overlap is accessible. To our mind, the latter effect extends the original ideas of Bertaut \[12, 14\].

2. Basic relations describing the multiple charge spreading

For convenience, here we compile some results describing the multiple charge spreading and obtained earlier \[22\]. Every perfect crystal can be specified by a unit-cell charge distribution \(\rho(r)\) subject to the condition of neutrality of a unit cell:

\[
\int_V \rho(r) \, dr = 0, \tag{1}
\]

where the integral is over the volume occupied by \(\rho(r)\). The corresponding structure factor as a function of a reciprocal lattice vector \(h\) is of the form

\[
F(h) = \frac{1}{v} \int_V \rho(r) \exp(-2\pi i hr) \, dr, \tag{2}
\]

where \(v\) is the unit cell volume. Then the relation \(F(h = 0) = 0\) readily follows from \[11\]. In the particular case of a point-charge lattice the charge distribution is converted into

\[
\rho(r) = \sum_j q_j \delta(r - b_j), \tag{3}
\]

where \(j\) runs over point charges \(q_j\) belonging to a unit cell and located at positions \(b_j\), \(\delta(r)\) is the Dirac delta function.

The effect of charge spreading will be described by a spherically symmetric spreading function \(\sigma(|r|)\) normalized by the condition

\[
\int \sigma(|r|) \, dr = 4\pi \int_0^\infty \sigma(r) r^2 \, dr = 1. \tag{4}
\]
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Here $r = |r|$. The application of $\sigma(|r|)$ to the charge distribution in a unit cell results in an additional multiplier

$$S(h) = \int \sigma(|r|) \exp(-2\pi i h r) \, dr$$

modifying structure factor \(F(h)\) as follows:

$$F(h) \rightarrow F(h)S^n(h)$$

if the spreading of interest is performed \(n\) times in a consecutive manner.

The corresponding potential effect of a multiple spreading in direct space is described by the function

$$\Omega^{(n)}(\tilde{R}) = \int \sigma(r_1) \cdots \sigma(r_n) \, dr_1 \cdots dr_n,$$

As a result, it is found that the electrostatic potential field arising within such a procedure of spreading can be written as

$$U(n)(r) = \frac{1}{\pi} \sum' h F(h)S^n(h) \frac{\exp(2\pi i h r)}{|h|^2}$$

$$+ \sum' \int_V dR \rho(R_1) W^{(n)}(|\tilde{R}_i|) \left\{ q_j \Omega^{(n)}(0) \right\}_{r=b_j},$$

where the prime on the summation sign over reciprocal lattice vectors \(h\) implies missing the term at \(h = 0\), the parameter \(i\) runs over vectors \(R_i\) determining the Bravais lattice and specifying different unit cells, we introduce the notation \(\tilde{R}_i = R_i + r_1 - r\), the prime on the summation sign over \(i\) means that a possible singularity associated with the denominator in the summand must be omitted as well,

$$\frac{W^{(n)}(R)}{R} = \frac{1}{R} - \Omega^{(n)}(R),$$

the last term on the right-hand side of \(8\) is the correction associated with a point charge \(q_j\) if it happens at \(r\). Taking potential \(8\) into account, we write down the Coulomb energy per unit cell in the form

$$\mathcal{E}_n = \frac{\nu}{2\pi} \sum' h F(h)S^n(h) \frac{\exp(2\pi i h r)}{|h|^2}$$

$$+ \frac{1}{2} \sum' \int_V dR_1 \rho(R_1) \rho(R_2)$$

$$\times \frac{W^{(n)}(|\tilde{R}_{12}|)}{|\tilde{R}_{12}|} - \frac{\Omega^{(n)}(0)}{2} \sum_j q^2_j,$$

where the summation over \(j\) is over all point charges \(q_j\) in the unit cell and \(\tilde{R}_{12} = R_i + r_1 - r_2\).

The recurrence relations associated with \(\Omega^{(n)}(R)\) and \(\Omega^{(n)}(0)\), respectively, take the form

$$\Omega^{(n)}(R) = \frac{2\pi}{R} \int_0^\infty \sigma(r) r \, dr \int_{|R-r|}^{R+r} \Omega^{(n-1)}(y) \, dy,$$

$$\Omega^{(n)}(0) = 4\pi \int_0^\infty \sigma(r) \Omega^{(n-1)}(r) r^2 \, dr.$$
The case of \( n = 1 \) is then straightforward \[^{22}\] and is described by
\[
W^{(1)}(R) = 4\pi \int_{R}^{\infty} \sigma(r)(r-R)\,dr,
\]
\[
\Omega^{(1)}(0) = 4\pi \int_{0}^{\infty} \sigma(r)\,dr,
\]
providing that \( \Omega^{(0)}(R) = 1/R \). In the case of \( n = 2 \) one can obtain
\[
W^{(2)}(R) = 4\pi^2 \left[ \int_{0}^{\infty} dr_1 \int_{0}^{\infty} dr_2 A(r_1, r_2) - \int_{0}^{R} dr_1 \int_{0}^{R-r_1} dr_2 \right] \times A(r_1, r_2) - 2 \int_{0}^{\infty} dr_1 \int_{R+r_1}^{\infty} dr_2 B(r_1, r_2),
\]
\[
\Omega^{(2)}(0) = 32\pi^2 \int_{0}^{\infty} \sigma(r_1) r_1 \, dr_1 \int_{0}^{r_1} \sigma(r_2)(r_2)^2 \, dr_2,
\]
where the following definitions
\[
A(r_1, r_2) = \sigma(r_1)\sigma(r_2) r_1 r_2 (R - r_1 - r_2)^2,
\]
\[
B(r_1, r_2) = \sigma(r_1)\sigma(r_2) r_1 r_2 (R + r_1 - r_2)^2
\]
are taken into account and notation \[^{9}\] is also utilized.

Employed to the simple exponential spreading specified by
\[
\sigma(r) = \frac{\alpha^3}{8\pi} \exp(-\alpha r),
\]
the latter results give rise to
\[
S(h) = \left[ 1 + \left( \frac{2\pi|h|}{\alpha} \right)^2 \right]^{-2},
\]
\[
\tilde{W}^{(1)}(z) = \left( 1 + \frac{z^2}{2} \right) \exp(-z),
\]
\[
\tilde{W}^{(2)}(z) = \left( 1 + \frac{11z^2}{16} + \frac{3z^4}{16} \right) \exp(-z),
\]
\[
\tilde{W}^{(3)}(z) = \left( 1 + \frac{193z^2}{256} + \frac{65z^4}{256} + \frac{37z^6}{768} + \frac{z^8}{192} + \frac{z^{10}}{3840} \right) \exp(-z),
\]
\[
\tilde{\Omega}^{(1)}(0) = \frac{\alpha}{2}, \quad \tilde{\Omega}^{(2)}(0) = \frac{5\alpha}{16}, \quad \tilde{\Omega}^{(3)}(0) = \frac{63\alpha}{256}.
\]

Here we go over to a new dimensionless variable \( z = \alpha R \), so that the notations introduced above are modified as follows:
\[
W^{(n)}(R) = \tilde{W}^{(n)}(z).
\]

The other important case is associated with a Gaussian spreading function
\[
\sigma_{\mu}(r) = \left( \frac{n\mu^2}{\pi} \right)^{3/2} \exp\left(-n\mu^2 r^2\right).
\]
The results appropriate to this case are of the form
\[
S_{\mu}^{(n)}(h) = \exp\left(-\frac{\pi^2|h|^2}{\mu^2}\right),
\]
\[
\tilde{W}^{(n)}(z) = \text{erfc}(z) = \frac{2}{\sqrt{\pi}} \int_{z}^{\infty} \exp(-u^2) \, du,
\]
\[
\tilde{\Omega}^{(n)}(0) = \frac{2\mu}{\sqrt{\pi}}.
\]
The fact that the final results are independent of \( n \) is the peculiar feature of the Ewald approach, as stressed earlier.
3. Spatially confined spreading

Let us now consider spherically symmetric spreading functions bounded by a radius $R_0$:

$$\sigma_k(r) = \begin{cases} g_{k(s)}(r) & \text{at } r \leq R_0, \\ 0 & \text{at } r > R_0, \end{cases}$$  \hspace{1cm} (30)

where we restrict ourselves to polynomials $g_{k(s)}$ of order $k$ which in turn result in

$$S_{k(s)}(h) \propto |h|^{-s}. \hspace{1cm} \text{The meaning of the parameter } s \text{ is defined therefrom.}$$

Then it is evident that relation (9) contributing to (8) still takes form (25), but at $z = R/R_0$. Furthermore, it will appear that $\tilde{W}_k^{(n)}(z) = 0$ as $z \geq n$. This fact is natural for the Coulomb potential generated by a spherically symmetric charge distribution and implies that the sum over $i$ in (3) is actually finite and includes only unit cells nearest to a reference point.

There are different polynomials discussed in the literature [12, 13, 14, 16, 18, 20, 21, 23, 24]. Interested in principal particular cases of spreading (30), we begin with a uniform spreading normalized by condition (4), as proposed by Bertaut [12]:

$$g_{0(2)}(r) = \frac{3}{4\pi R_0^3}. \hspace{1cm} (31)$$

According to [5], relations (30) and (31) yield

$$S_{0(2)}(h) = \frac{3}{Y^2} \left( \sin Y - \cos Y \right), \hspace{1cm} (32)$$

where $Y = 2\pi|h|/R_0$ and so $s = 2$ herein. If $n = 1$, then the other quantities of interest, which are specified by the subscript $k(s)$ and by the superscript $(n)$, are as follows:

$$\tilde{W}_{0(2)}^{(1)}(z) = \begin{cases} \frac{(1-z)^2(2+z)}{2} & \text{at } 0 \leq z \leq 1, \\ 0 & \text{at } z > 1, \end{cases} \hspace{1cm} (33)$$

$$\Omega_{0(2)}^{(1)}(0) = \frac{3}{2R_0}. \hspace{1cm} (34)$$

The fact that $S_0(h)$ contains the factor $Y^{-2}$ enhances the rate of convergence of a series over $h$ in (3) even at $n = 1$. This effect becomes stronger as $n$ increases. In the original approach of Bertaut [12] the case of $n = 2$ is considered. Making use of (15) and (16), one can show that

$$\tilde{W}_{0(2)}^{(2)}(z) = \begin{cases} \frac{(2-z)^4(10 + 8z + z^2)}{160} & \text{at } 0 \leq z \leq 2, \\ 0 & \text{at } z > 2, \end{cases} \hspace{1cm} (35)$$

$$\Omega_{0(2)}^{(2)}(0) = \frac{6}{5R_0}. \hspace{1cm} (36)$$

in this case. The event of $n = 3$ appears to be more complicated, though it is still described by formulae (11) and (12) based on relations (9) and (35). The results can be cast in the form

$$\Omega_{0(2)}^{(3)}(0) = \frac{1269}{1280R_0}, \hspace{1cm} (37)$$

$$\tilde{W}_{0(2)}^{(3)}(z) = \begin{cases} M_1(z) + M_2(z) & \text{at } 0 \leq z \leq 1, \\ M_1(z) & \text{at } 1 \leq z \leq 3, \\ 0 & \text{at } z > 3, \end{cases} \hspace{1cm} (38)$$
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where

\[ M_1(z) = \frac{(3-z)^6(72 + 81z + 18z^2 + z^3)}{53760}, \]
\[ M_2(z) = \frac{(1-z)^6(424 + 87z - 6z^2 - z^3)}{17920}. \]

It is important that, apart from enlarging the value of \( n \), another way of enhancing the rate of convergence of the series over reciprocal vectors can be arrived at by increasing the order \( k \) of \( g_{k(s)}(s) \) so as to enhance the degree \( s \) of \( Y \) in the denominator of \( S_{k(s)}(h) \) \[24\]. In particular, one can show that

\[ g_{1(3)}(r) = \frac{3(1-x)}{\pi R_0^3}, \]

where \( x = r/R_0 \), leads to

\[ S_{1(3)}(h) = \frac{12}{Y^3} \left( \frac{2(1-\cos Y)}{Y} - \sin Y \right). \]

In this case

\[ \tilde{W}_{1(3)}^{(1)}(z) = \begin{cases} (1-z)^3(1+z) & \text{at } 0 \leq z \leq 1, \\ 0 & \text{at } z > 1, \end{cases} \]
\[ \Omega_{1(3)}(0) = \frac{2}{R_0}. \]

Extending the consideration to the case of \( n = 2 \) here, we derive

\[ \Omega_{1(3)}(0) = \frac{52}{35R_0}, \]
\[ \tilde{W}_{1(3)}^{(2)}(z) = \begin{cases} M_3(z) + M_4(z) & \text{at } 0 \leq z \leq 1, \\ M_3(z) & \text{at } 1 \leq z \leq 2, \\ 0 & \text{at } z > 2, \end{cases} \]
\[ \text{where} \]
\[ M_3(z) = \frac{(2-z)^6(2+4z+z^2)}{140}, \]
\[ M_4(z) = \frac{(1-z)^7(3+z)}{35}. \]

Interested in polynomials of the lowest degree, the next several polynomials for the spreading function within this set are shown in Appendix A. Note that the couples of relations \[31\] and \[32\], \[33\] and \[42\] and finally \[A.1\] and \[A.2\] from Appendix A naturally agree with results proposed in \[13\] \[14\] \[19\] \[21\] \[23\]. One can see that the set of spreading functions is reduced to terms of the form \((1-x)^k\) \[13\] \[19\] \[20\] only at \( k = 1 \) and \( k = 2 \). This fact accounts for the known statement \[16\] that the simple form \((1-x)^k\) is not yet efficient at \( k > 2 \).

Note that various polynomials are possible for a given \( s \) if they are not restricted to the lowest degree. Of different polynomials associated with \( s = 3 \) and so competing with \(41\), here we consider the only one that is expected to be very effective \[24\] and is of the form

\[ g_{2(3)}(r) = \frac{5x(1-x)}{\pi R_0^3}. \]
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Its Fourier transform is also known:

\[
S_{2(3)}(\mathbf{h}) = \frac{20}{\sqrt{\pi}} \left( \frac{6 \sin Y}{Y^2} - \frac{2 + 4 \cos Y}{Y} - \sin Y \right).
\]  
\(50\)

All the other quantities appropriate to the case can be obtained in the manner considered above. The case of \(n = 1\) is described by

\[
\tilde{W}^{(1)}_{2(3)}(z) = \begin{cases} 
(1 - z)^3 \left( 3 + 4z + 3z^2 \right) / 3 & \text{at } 0 \leq z \leq 1, \\
0 & \text{at } z > 1,
\end{cases}
\]  
\(51\)

\[
\Omega^{(1)}_{2(3)}(0) = \frac{5}{3R_0}.
\]  
\(52\)

In the case of \(n = 2\), we reach

\[
\Omega^{(2)}_{2(3)}(0) = \frac{85}{63R_0},
\]  
\(53\)

\[
\tilde{W}^{(2)}_{1(3)}(z) = \begin{cases} 
M_5(z) - M_6(z) & \text{at } 0 \leq z \leq 1, \\
M_5(z) & \text{at } 1 \leq z \leq 2, \\
0 & \text{at } z > 2,
\end{cases}
\]  
\(54\)

where

\[
M_5(z) = \frac{(2 - z)^6 (28 + 24z + 42z^2 + 16z^3 + 3z^4)}{1512},
\]  
\(55\)

\[
M_6(z) = \frac{5(1 - z)^7 (7 + 4z + z^2)}{189}.
\]  
\(56\)

4. Optimization of spreading parameters

It is important that every spreading function is characterized by a certain parameter. The problem of its optimization is traditional. In particular, the value of \(\mu = 2\sqrt{\pi/d}\) describing a Gaussian spreading function was used as a unique for the NaCl structure, where \(d\) is the lattice spacing \[1, 7\]. The situation associated with a simple exponential spreading appears to be quite similar \[13, 25, 26\].

First of all, here we develop the treatment appropriate to this case. As mentioned earlier \[5\], it is based on the Coulomb characteristic of a Bravais lattice, the parameter put forward by Harris and Monkhorst \[27\]. In order to discuss this approach, we consider a Bravais lattice composed of unit point charges and immersed in a neutralizing uniform background. It is easy to show that the interaction of a background with the bulk potential field vanishes and the same is right for the background contribution to the sum over reciprocal lattice in expression \(10\) \[5, 22, 27\]. As a result, the effect of lattice summation in \(10\) is associated with the contribution of point charges alone. However, there is a remainder constituted of two simple finite terms there. One of those terms is determined by the last optional term in \(10\). The other one is the contribution of a background to the integral over real space and so is described by the negative of the quantity

\[
G^{(n)} = \frac{8\pi^2 n}{3v} \int_0^\infty r^4 \sigma(r) dr,
\]  
\(57\)

keeping in mind that a background is of negative charge \[22\].

In the case at hand equation \(2\) leads to \(F(h) = 1\). It implies that both the series over reciprocal and direct lattices in the original energy expressions are divergent and
the spreading length, denoted in general as \( \lambda \), actually forms cut-off parameters for both of those series in formula (10). Therefore it is expedient to cast the expression for the Coulomb characteristic \( C \) of a Bravais lattice in the following schematic form:

\[
\frac{A}{\lambda} + B\lambda^2 = G^{(n)}_\lambda + \Omega^{(n)}_\lambda(0) + C,
\]

where the terms on the left-hand side stand for the contributions of series over reciprocal and direct lattices, respectively, with singling out their principal dependence on \( \lambda \). Note that \( C \) is the value of the modulated lattice potential at the site of a unit point charge and so \( C/2 = \mathcal{E} \), where \( \mathcal{E} \) is the total energy per point charge in question [5]. It is significant that except for \( C \), all the terms in relation (58) are positive. Moreover, according to definitions (4), (12) and (57), one can readily show that

\[
G^{(n)}_\lambda \propto \lambda^2, \quad \Omega^{(n)}_\lambda(0) \propto \lambda^{-1}.
\]

Therefore \( G^{(n)}_\lambda \) and \( \Omega^{(n)}_\lambda(0) \) may be treated as counterparts to the direct-lattice series and to the reciprocal-lattice one in (58), respectively. Bearing in mind that \( C \) is a constant, we draw a conclusion that an optimum relation between the terms on the left-hand side of formula (58) is associated with a minimum of the right-hand side that is determined by the condition

\[
\frac{d}{d\lambda} \left[ G^{(n)}_\lambda + \Omega^{(n)}_\lambda(0) \right] = 0.
\]

The Gaussian spreading (26) substituted into formula (57) at a given \( n \) results in

\[
G_\mu = \frac{\pi}{\mu v^2}.
\]

Note that this functional form is still independent of \( n \). In this case \( \lambda = 1/\mu \). Inserting relations (29) and (61) into (60), we derive the result discussed earlier [4, 5]:

\[
\mu^{opt} = \sqrt{\pi} v^{-1/3},
\]

providing that the differentiation immediately with respect to \( \mu \) in (60) is also admissible.

In the cases of simple exponential spreading discussed above, the substitution of (19) into (57) yields

\[
G_\alpha^{(n)} = \frac{8\pi n}{v\alpha^2}.
\]

Now we substitute issue (63) along with one of the relations described by (24) into (60). The peculiar feature of relations (59) implies that both the differentiation with respect to \( \alpha \) and the differentiation with respect to \( \lambda = 1/\alpha \) in (60) also lead to the same result:

\[
\alpha^{opt} = \begin{cases} 
2 \left( \frac{4\pi}{v} \right)^{1/3} & \text{at } n = 1, \\
8 \left( \frac{\pi}{5v} \right)^{1/3} & \text{at } n = 2, \\
16 \left( \frac{\pi}{21v} \right)^{1/3} & \text{at } n = 3.
\end{cases}
\]

Note that the value of \( \alpha^{opt} \) increases when \( n \) changes from 1 to 3. It means that the initial spreading function becomes more compact and thus the enhancing effect of spreading at \( n = 2 \) and further at \( n = 3 \) turns out to be somewhat restricted.
Another case of optimization arises if the values of the spreading parameter $\alpha$, for definiteness, are supposed to depend on the cut-off parameter of summation $m$. If an integer value of $m$ is common to both summations over reciprocal and direct space, then the result for the energy can be written as $E^{[m]}(n)$. Apart from this quantity, it is also advantageous to consider the value of $E^{[m+m_a]}(n)$, where $m_a$ is a certain integer as well. The value of $\alpha^{[m]}(n)$ associated with a given $m$ can be determined by the condition

$$E^{[m+m_a]}(n) - E^{[m]}(n) = 0.$$  (65)

In order to understand this relation, one should point out that at least two different situations arise here. Indeed, if $m_a = 1$, then formula (65) is apparently reduced to the condition of stability with respect to small variations of $m$. The opposite limiting case arises when $m_a \gg 1$. This case corresponds to the fact that (65) evaluates all the rest removed within the cut-off procedure. The requirement of zero value of this remainder is then natural. It should be emphasized that both of these motifs are in accord with the statements discussed in the literature [9, 11]. In other words, our latter treatment, albeit original, is developed in a quite traditional manner, without recourse to any additional adjustable function simulating the contribution of the lattice sum over direct space [8].

It is worth noting that according to (65), the value of the spreading parameter $\alpha^{[m]}(n)$ is connected with the value of the cut-off parameter $m$. It is evident that this relation could be formally inverted, i.e., $m$ might be treated as a function of $\alpha^{[m]}(n)$ as well. However, contrary to $\alpha^{(n)}$ that is continuous initially, $m$ is a discrete parameter and so a discrete set of values $\alpha^{[m]}(n)$ appropriate to different $m$ actually arises as a solution of (65).

One more possibility to optimize the calculation of lattice series is associated with the fact that the lattice sum in direct space converges faster than that in reciprocal space. Therefore it may be advantageous to choose the cut-off parameter $m_a$ truncating the direct lattice sum larger than the cut-off parameter $m$ applied to the corresponding reciprocal lattice sum. In this case the correlation between the cut-off parameter $m_a$ and the spreading parameter $\alpha^{(n)}$ may be of interest, providing that the third parameter $m$ is fixed [8, 9, 11]. The foregoing treatment based on (65) can be easily extended to the present case if we adopt

$$E^{[m+m_a, m_R+m_a]}(n) - E^{[m, m_R]}(n) = 0,$$  (66)

where we conjecture that the increment $m_a$ in both $m$ and $m_R$ is the same, for simplicity. Note that the two limiting cases mentioned above as associated with $m_a$ still take place. As far as the choice of $m_R$ is concerned, it turns out that this value is to be as large as possible. Actually its value is restricted by machine accuracy. On the other hand, its effect on $\alpha^{(n)}$ is strong, as will be shown later on.

In the events of confined spreading functions restricted by $R_0$, the Bertaut version dealing with non-overlapping charge distributions is the most popular. As is well known, the sum over direct space is then zero and so it does not contribute to the result that is determined by the sum over reciprocal space alone [12, 14]. Keeping in mind that the value $R_0$ must still be as large as possible, one may conclude that $R_0$ is to be half the nearest interatomic distance [12, 13, 16, 18, 20, 21, 28, 29, 30]. This convention is sustained in modern papers devoted to this subject as well [24].
As a result, the assessment of the accuracy of computation is reduced to the classical consideration of the cut-off effect upon summation over reciprocal space \[15, 19\].

However, it is expedient to note that the aforementioned restriction on \( R_0 \) may be convenient, but is not principal. Indeed, some further growth of \( R_0 \) results in the appearance of a certain direct sum that is finite and can be calculated rigorously. On the other hand, the larger \( R_0 \) the faster convergence of the series over reciprocal space. This is the reason to make use of the largest value of \( R_0 \) consistent with machine accuracy of evaluation of the direct sum \[2, 18, 24\]. The latter is the problem typical of practical summation.

\section*{5. Effect of infinite normalized spreading functions}

Here we apply the foregoing results to the classical NaCl structure composed of point charges, which remains attractive as a model system \[21, 30, 31\]. As known, a face-centred cubic Bravais lattice describes this structure, with two sites per unit cell. If \( d \) is the edge of an elementary cube, then \( v = d^3/4 \) and the basis vectors for point charges \( \pm q \) may be chosen as:

\begin{equation}
+ q : \mathbf{b}_1 = (0, 0, 0), \quad - q : \mathbf{b}_2 = (d/2, 0, 0).
\end{equation}

In terms of the elementary translations

\begin{equation}
\mathbf{a}_1 = \frac{d(1, 1, 0)}{2}, \quad \mathbf{a}_2 = \frac{d(1, 0, 1)}{2}, \quad \mathbf{a}_3 = \frac{d(0, 1, 1)}{2},
\end{equation}

an arbitrary translation vector is of the form

\begin{equation}
\mathbf{R}_i = m_1 \mathbf{a}_1 + m_2 \mathbf{a}_2 + m_3 \mathbf{a}_3,
\end{equation}

where \( m_j \) are integers. The elementary reciprocal lattice translations appropriate to the vectors in \[68\] are defined by the scalar product \( (\mathbf{a}_i, \mathbf{h}) = \delta_{ij} \), where \( \delta_{ij} \) is the Kronecker delta, and are as follows:

\begin{equation}
\mathbf{h}_1 = \frac{(1, 1, -1)}{d}, \quad \mathbf{h}_2 = \frac{(1, -1, 1)}{d}, \quad \mathbf{h}_3 = \frac{(-1, 1, 1)}{d}.
\end{equation}

They compose a general reciprocal lattice vector of the form

\begin{equation}
\mathbf{h} = m_1 \mathbf{h}_1 + m_2 \mathbf{h}_2 + m_3 \mathbf{h}_3.
\end{equation}

Note that definitions \[68\] and \[70\] are conventional \[32\].

On taking formulae \[2, 4\] and \[67\] into account, expression \[10\] for the energy of interest is transformed into

\begin{equation}
\mathcal{E}^{(n)} = \frac{q^2}{\pi v} \sum_k \frac{\left| 1 - \cos[2\pi (\mathbf{h} \cdot \mathbf{b}_2)] \right| S^2(\mathbf{h})}{|\mathbf{h}|^2} + q^2 \sum_i \left( \frac{W^{(n)}(R_i)}{|R_i|} - \frac{W^{(n)}(|R_i + \mathbf{b}_2|)}{|R_i + \mathbf{b}_2|} \right)
\end{equation}

\begin{equation}
- q^2 \Omega^{(n)}(0).
\end{equation}

According to \[68 - 71\], the parameters necessary for the further summation take the form

\begin{equation}
|R_i| = \frac{d}{2} \left[ (m_1 + m_2)^2 + (m_2 + m_3)^2 + (m_3 + m_1)^2 \right]^{1/2},
\end{equation}

\begin{equation}
|R_i + \mathbf{b}_2| = \frac{d}{2} \left[ (m_1 + m_2 + 1)^2 + (m_2 + m_3)^2 + (m_3 + m_1 + 1)^2 \right]^{1/2},
\end{equation}

\begin{equation}
|h| = \frac{1}{d} \left[ (m_1 + m_2 - m_3)^2 + (m_2 + m_3 - m_1)^2 + (m_3 + m_1 - m_2)^2 \right]^{1/2},
\end{equation}

\begin{equation}
1 - \cos[2\pi (\mathbf{h} \cdot \mathbf{b}_2)] = \begin{cases} 0 & \text{at } m_1 + m_2 - m_3 \text{ even}, \\ 2 & \text{at } m_1 + m_2 - m_3 \text{ odd}. \end{cases}
\end{equation}
Table 1. The Coulomb energy $E$, in units of $q^2/d$, per unit cell of the NaCl point-charge lattice. The calculation is based on a Gaussian charge spreading with $\mu = 2\sqrt{\pi}/d$ proposed originally by Ewald ($E_{Ew}$) and with $\mu = \sqrt{\pi}v^{-1/3}$ in agreement with our proposal ($E_{our}$). The results, with significant figures only, are shown in dependence on $m$ restricting actual ranges of summation over direct and reciprocal space, in accord with condition (77).

| $m$ | $E_{Ew}$ | $E_{our}$ |
|-----|----------|----------|
| 1   | -3.5     | -3.5     |
| 2   | -3.49513 | -3.4951292 |
| 3   | -3.49512918927 | -3.49512918926636* |

* This value agrees with the result of Sakamoto [33].

Note that the latter relation is very specific [21]. Employing relations (73)–(76) in formula (72), we propose that the parameters of summation are restricted by a common condition

$$|m_j| \leq m,$$

where an integer $m$ is varying. The rate of convergence of series in equation (72) will be then studied in dependence on $m$.

Here we start from a Gaussian spreading function that is a classical example [2, 5, 13]. Bearing in mind that the effect of multiple charge spreading is reduced only to some definite scaling of Gaussian parameters in relations (74)–(76), we consider the case of a unique optimal value of a Gaussian parameter described by (62). The value of the Madelung energy is shown in table 1 in dependence on $m$. The effect of the original value of $\mu = 2\sqrt{\pi}/d$ proposed by Ewald [1, 7] is demonstrated in the same table for comparison. Table 1 shows that the rate of convergence is actually fantastic in both these cases, though our choice appears to be somewhat more advanced. Such results are in general expected [19]. This is the reason that we will not discuss further possibilities of optimization addressed to a Gaussian spreading function [9].

At this stage it is worth noting that the accuracy of results specified by Gaussian spreading function drops drastically if we restrict ourselves to spherical domains of summation over both the reciprocal and direct lattices. This fact is known [8]. Unfortunately, such a loss in accuracy caused by spherical modes of summation in the series at hand turns out to be typical of all the other cases discussed below. It means that modes of summation supporting the crystal symmetry are more advantageous for perfect crystals. On the other hand, very popular schemes of spherical summation seem to be essential in applications describing disordered systems [9, 11].

In the cases of a simple exponential spreading formulae (20)–(24) and (64) are utilized in (72) first of all. The results of summation are listed in table 2. As anticipated, the tendency towards enhancing the rate of convergence takes place upon increasing the order of multiplicity $n$. This effect is quite general and has been pointed out by Bertaut [14] in connection with the particular cases of $n = 1$ and $n = 2$ at spatially confined spreading functions, as will be discussed in the sequel.

However, table 2 also shows that, contrary to a Gaussian spreading function, the application of a simple exponential spreading with fixed optimal values of $\alpha(n)$ results in much more moderate rate of convergence of lattice series in question. Therefore further efforts towards optimizing lattice calculations seem to be necessary here. The next step of optimization discussed is associated with a possible variation of $\alpha(n)$ in
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Table 2. The Coulomb energy $\xi$, in units of $q^2/d$, per unit cell of the NaCl point-charge lattice. The calculation is based on a simple exponential charge spreading with optimal values of $\alpha(n)$ described by relation (65) for $n = 1$, $n = 2$ and $n = 3$. The results are shown in dependence on $m$ in condition (24) again.

| $m$ | $\xi_{(1)}$ | $\xi_{(2)}$ | $\xi_{(3)}$ |
|-----|-------------|-------------|-------------|
| 5   | $-3.5$      | $-3.4951$   | $-3.495129$ |
| 10  | $-3.5$      | $-3.495129$ | $-3.495129189$ |
| 15  | $-3.495$   | $-3.4951292$ | $-3.49512918927$ |
| 20  | $-3.495$   | $-3.49512919$ | $-3.4951291892664$ |
| 25  | $-3.495$   | $-3.495129189$ | $-3.4951291892664$ |
| 30  | $-3.4951$  | $-3.4951291893$ | $-3.4951291892664$ |
| 35  | $-3.4951$  | $-3.4951291893$ | $-3.49512918926636^a$ |

$^a$ This value agrees with the result of Sakamoto (33).

dependence on $m$. With the help of condition (65), this effect can be readily taken into account. Here two particular cases of $m_a = 1$ and $m_a = 10$ are studied. The results of calculation are compiled in table 2. We see that the tendency towards reducing the value of $\alpha(n)$ upon growing $m$ is common to all the events of $n$ under consideration. It implies that the effective charge distributions become more diffuse as the range of summation increases. On the other hand, the multiple spreading leads to larger values of $\alpha(n)$. This output agrees with the results of (64). It is important that the enhancement of the rate of convergence arises in both the cases of $m_a$, but the effect at $m_a = 10$ is somewhat stronger than that at $m_a = 1$.

The latter trend can be fruitful when we go over to the next step of optimization based on relation (62). The event of $m = 1$ and $m_a = 22$ is studied, providing that

Table 3. The Coulomb energy $\xi$, in units of $q^2/d$, per unit cell of the NaCl point-charge lattice in the case similar to that in table 2 but with $\alpha(n)$ of which variation with $m$ is specified by (65) at $m_a = 1$ and $m_a = 10$, respectively.

| $m$ | $\alpha_{(1)}$ | $\xi_{(1)}$ | $\alpha_{(2)}$ | $\xi_{(2)}$ | $\alpha_{(3)}$ | $\xi_{(3)}$ |
|-----|----------------|-------------|----------------|-------------|----------------|-------------|
| 1   | 6.8265         | $-3.5$      | 11.2028        | $-3.5$      | 14.4677        | $-3.495$   |
| 3   | 5.0228         | $-3.5$      | 8.7002         | $-3.49513$  | 11.5983        | $-3.4951292$ |
| 5   | 4.0395         | $-3.495$   | 7.2189         | $-3.4951292$ | 9.8319        | $-3.49512919$ |
| 7   | 3.4073         | $-3.495$   | 6.2024         | $-3.49512919$ | 8.5572        | $-3.49512918927$ |
| 9   | 2.9639         | $-3.49513$ | 5.4620         | $-3.49512919$ | 7.6008        | $-3.495129189266$ |
| 11  | 2.6336         | $-3.49513$ | 4.8970         | $-3.4951291893$ | 6.8568        | $-3.4951291892664$ |
| 13  | 2.3769         | $-3.49513$ | 4.4502         | $-3.49512918927$ | 6.2618        | $-3.49512918926636^a$ |

| $m_a = 10$ |
|-------------|-------------|-------------|-------------|-------------|-------------|
| 1   | 6.5898      | $-3.495$   | 11.1262      | $-3.4951292$ | 14.4308      | $-3.4951291893$ |
| 2   | 5.5308      | $-3.495$   | 9.6293       | $-3.4951292$ | 12.6762      | $-3.49512918927$ |
| 3   | 4.8102      | $-3.495$   | 8.6223       | $-3.49512919$ | 11.5650      | $-3.49512918927$ |
| 4   | 4.2661      | $-3.4951$  | 7.7974       | $-3.49512919$ | 10.5979      | $-3.49512918927$ |
| 5   | 3.8453      | $-3.4951$  | 7.1272       | $-3.495129189$ | 9.7854       | $-3.4951291892664$ |
| 6   | 3.5093      | $-3.49513$ | 6.5721       | $-3.4951291893$ | 9.0943      | $-3.4951291892664$ |
| 7   | 3.2346      | $-3.49513$ | 6.1058       | $-3.4951291893$ | 8.5019      | $-3.4951291892664$ |
| 8   | 3.0052      | $-3.49513$ | 5.7084       | $-3.4951291893$ | 7.9888      | $-3.49512918926636^a$ |

$^a$ This value agrees with the result of Sakamoto (33).
Table 4. The Coulomb energy $\mathcal{E}$, in units of $q^2/d$, per unit cell of the NaCl point-charge lattice in the case of a simple exponential charge spreading like that in table 3 but with $m = 1$ and $m_0 = 22$ restricting lattice summations over reciprocal and direct space, respectively, in accord with (77). Then the values of $\alpha(n)$ at $n = 1$, 2 and 3 are determined by (66) at $m_a = 1$ and $m_a = 10$.

| $m_a$ | $n$ | $\alpha(n)$ | $\mathcal{E}(n)$ |
|-------|-----|-------------|----------------|
| 1     | 1   | 1.0346      | -3.4951         |
|       | 2   | 1.9312      | -3.4951292      |
|       | 3   | 2.6808      | -3.495129189    |
| 10    | 1   | 1.0681      | -3.49512919     |
|       | 2   | 1.9562      | -3.49512918927  |
|       | 3   | 2.6986      | -3.49512918926364 |

$^a$ This value agrees with the result of Sakamoto [33].

due to the values $m_a = 1$ and $m_a = 10$ are utilized in (66). The corresponding results are listed in table 4. Table 4 shows that the values of $\alpha(n)$ corresponding to each value of $n$ appear to be close, but the effect of $m_a = 10$ is more pronounced again.

6. Trends in the application of confined spreading functions

As far as confined spreading functions are concerned, here we restrict ourselves to the situation opposite to the classical one described by charge distributions non-overlapping after spreading [12, 14]. In other words, we propose that the parameter $R_0$ in (30) is greater than the lattice parameter $d$. It is important that the sum over direct space is always finite in such a case and so it can be counted with a high precision. In practice our choice of $R_0$ is actually restricted by machine accuracy. As a result, optimum values of $R_0$ appear to be different and depend on each particular case under consideration. For example, if $k = 0$, then we adopt $R_0 = 7d$ upon considering all three cases for $n$ from one to three. Note that the value of $m$ in (77) restricts only the reciprocal lattice series now. The computation based on formulae (31)–(40) substituted into (72) gives rise to the results presented in table 5. Table 5 shows that

Table 5. The specific Coulomb energy $\mathcal{E}$, in units of $q^2/d$, for the NaCl point-charge lattice is obtained in dependence on the restricting parameter $m$ at a fixed value of $R_0 = 7d$ common to all the cases of the confined polynomial spreading function $g_0(2)(r)$ defined by formula (31). The cases of $\mathcal{E}(n)$ at $n = 1$, $n = 2$ and $n = 3$, which are specified by $g_0(2)$ in formula (31) are considered.

| $m$ | $\mathcal{E}(1)$ | $\mathcal{E}(2)$ | $\mathcal{E}(3)$ |
|-----|-----------------|-----------------|-----------------|
| 0   | -3.5            | -3.49513        | -3.495129189    |
| 1   | -3.495          | -3.4951292      | -3.49512918927  |
| 2   | -3.495          | -3.4951292      | -3.4951291892664 |
| 3   | -3.495          | -3.4951292      | -3.4951291892664 |
| 4   | -3.495          | -3.49512919     | -3.4951291892664 |
| 5   | -3.495          | -3.49512919     | -3.4951291892664 |
| 10  | -3.495129      | -3.49512919     | -3.495129189266364 |
| 15  | -3.495129189   | -3.495129189    | -3.495129189266364 |

$^a$ This value agrees with the result of Sakamoto [33] (see also references [34, 35]).
Table 6. The specific Coulomb energy $\varepsilon$, in units of $q^2/d$, for the NaCl point-charge lattice obtained with making use of either $g_{1(3)}(r)$ defined by (41) at $R_0 = 6.5d$ or $g_{2(3)}(r)$ defined by (49) at $R_0 = 7d$, in dependence on the cut-off parameter $m$. The cases of $n = 1$ and $n = 2$ for both these spreading functions are considered together for comparison.

| $m$ | $\varepsilon_{(1)}$ | $\varepsilon_{(2)}$ | $\varepsilon_{(1)}$ | $\varepsilon_{(2)}$ |
|-----|----------------------|----------------------|----------------------|----------------------|
| 0   | -3.495               | -3.4951292           | -3.495               | -3.4951292           |
| 1   | -3.49513             | -3.4951291890        | -3.49513             | -3.4951291890        |
| 2   | -3.495129            | -3.49512918927       | -3.49513             | -3.49512918927       |
| 3   | -3.495129            | -3.49512918927       | -3.49513             | -3.49512918927       |
| 4   | -3.495129            | -3.49512918927       | -3.49513             | -3.49512918927       |
| 5   | -3.495129            | -3.49512918927       | -3.49513             | -3.49512918927       |
| 10  | -3.495129            | -3.4951291892664     | -3.495129            | -3.4951291892664     |
| 15  | -3.4951292           | -3.4951291892664     | -3.4951292           | -3.4951291892664     |
| 20  | -3.49512919         | -3.49512918926636    | -3.49512919         | -3.49512918926636    |

$^a$This value agrees with the result of Sakamoto [33]
Table 7. The specific Coulomb energy $E$, in units of $q^2/d$, for the NaCl point-charge lattice is obtained in dependence on the restricting parameter $m$ at a fixed value of $R_0 = 15d$ common to all the cases of the confined polynomials specified by (A.1)–(A.12) in Appendix A at $n = 1$.

| $m$ | $E_{(1)}$ (case of $g_{2(4)}(r)$) | $E_{(1)}$ (case of $g_{4(5)}(r)$) | $E_{(1)}$ (case of $g_{5(6)}(r)$) |
|-----|----------------------------------|----------------------------------|----------------------------------|
| 0   | $-3.49513$                       | $-3.49512919$                   | $-3.49512919$                   |
| 1   | $-3.495129$                      | $-3.49512919$                   | $-3.49512919$                   |
| 2   | $-3.4951292$                     | $-3.4951291893$                 | $-3.4951291893$                 |
| 3   | $-3.4951292$                     | $-3.4951291893$                 | $-3.49512918927$                |
| 4   | $-3.49512919$                    | $-3.49512918927$                | $-3.49512918927$                |
| 5   | $-3.49512919$                    | $-3.49512918927$                | $-3.49512918927$                |
| 10  | $-3.49512919$                    | $-3.4951291892664$              | $-3.4951291892664$              |
| 15  | $-3.495129189$                   | $-3.49512918926636$             | $-3.49512918926636$             |
| 20  | $-3.495129189$                   | $-3.49512918926636$             | $-3.49512918926636$             |
| 25  | $-3.495129189$                   | $-3.49512918926636$             | $-3.49512918926636$             |
| 30  | $-3.495129189$                   | $-3.49512918926636$             | $-3.495129189266364$            |
| 35  | $-3.4951291893$                  | $-3.49512918926636$             | $-3.4951291892663644^a$         |

$^a$ This value agrees with the result of Sakamoto [33] (see also references [34, 35]).

k grows. Of course, this effect turns out to be less prominent in comparison with the cases of $n = 2$ and $n = 3$ mentioned above. Nevertheless, at $k = 5$ the limiting precision adopted in our calculations is eventually attained as well.

Note that if $n$ is odd, then the contribution of the reciprocal lattice sum tends to the exact value in an oscillatory manner due to trigonometric functions describing $S_k(s)(h)$ appropriate to all the events in this section [13, 21].

Some final comment is necessary on the question why all the foregoing tables contain limiting energy values with a rather large number of significant figures. Indeed, one may think that the lattice parameters of real structures are usually known up to four, at best six, figures only [37]. Nevertheless, even in this case the numerical calculation should be a bit more accurate. However, the reason of our ultimate accuracy is quite different. As shown, the analytic accuracy of a series is associated with the number of unit cells taken into account and restricted by a cut-off parameter introduced upon series computation. On the other hand, restricted by machine accuracy, the overall accuracy of computation depends on the total number of implemented operations and so is connected anyhow with the total number of point charges taken into account. The interplay between these tendencies implies that the effect of machine accuracy may be predominant if the number of charges per unit cell increases. In other words, an ultimate accuracy achieved for model structure with a small unit cell may be regarded as a guaranty of a sufficient accuracy while each of the approaches developed above is applied to modern compounds with large unit cells.

7. Conclusion

In summary, it has been confirmed that the effect of a multiple charge spreading results in increasing the rate of convergence of the lattice series. In other words, we deal with one more way to make the convergence faster, providing that this effect becomes stronger and stronger with every further repetition of the procedure of spreading. It is important that this is a property common to both the classes of infinite and confined
normalized spreading functions.

The optimization of the shape of spreading is more traditional in the problem of enhancing the rate of convergence. Nevertheless, we have proposed some novel approaches to this task. In particular, for infinite normalized functions of spreading two different situations are considered. In the case of a fixed spreading parameter, the universal approach of optimization is based on the investigation of the concomitant terms contributing to the Coulomb energy, which are independent of the lattice sums. The problem of optimizing the spreading parameter in dependence on the cut-off parameters of summation has been solved with the help of some conditions imposed on the remainder of the Coulomb energy, providing that this remainder is also treated in a truncated form. In the case of confined spreading functions the optimization in question is reduced to separating any main set of polynomials ensuring the progressive enhancement of the rate of convergence of the sum over reciprocal space.

It is found that the effect of optimization becomes much more efficient if the multiple spreading is applied as well. The only case independent of the multiple spreading is described by a Gaussian spreading function due to its invariance with respect to spreading in a multiple manner [22]. Moreover, it turns out that the convergence with a Gaussian spreading function is the most prominent even at a fixed optimized spreading parameter. This fact gives evidence that a Gaussian spreading seems to be the most suitable one in the treatment based on the charge spreading as a whole.

On the other hand, the investigation of confined functions of spreading shows that it is fruitful to choose larger values of the parameter restricting those functions. Of course, a certain finite sum over sites of the direct lattice then appears. However, there is no problem with its calculation. On the other hand, the contribution of the sum over reciprocal lattice becomes smaller, up to the case where the latter sum describes only a small correction to the result of direct summation. Note that this trend is eventually common to all the examples discussed above. By the way, it implies one more approach to the problem of direct summation of Coulomb series in crystals.

Appendix A. Particular forms of a polynomial spreading function

Here we continue the fundamental series of polynomials beginning with (31) and (41), which produce $S(h)$ with the enhancing power of $Y$ in the denominator, providing that the polynomial degree be a minimum. Note that the case of $n = 1$ alone will be considered here. So, the next example of such a polynomial, with including the concomitant relations, is of the form

$$g_{2(4)}(r) = \frac{15(1 - x)^2}{2\pi R_0^3}, \quad (A.1)$$
$$S_{2(4)}(h) = \frac{60}{Y^4} \left(2 + \cos Y - \frac{3\sin Y}{Y}\right), \quad (A.2)$$
$$W_{2(4)}^{(1)}(z) = \begin{cases} 
\frac{(1 - z)^4(2 + 3z)}{2} & \text{at } 0 \leq z \leq 1, \\
0 & \text{at } z > 1, 
\end{cases} \quad (A.3)$$
$$\Omega_{2(4)}^{(1)}(0) = \frac{5}{2R_0}, \quad (A.4)$$

where the notations of section 3 are utilized.
The next polynomial of spreading within the set under consideration is not simply defined by \((1 - x)^3\), but it is described as

\[
g_{4(5)}(r) = \frac{105(1 - x)^3(1 + 3x)}{16\pi R_0^3},
\]

with the concomitant quantities

\[
S_{4(5)}(h) = \frac{630}{Y^6}\left(\sin Y + \frac{8 + 7\cos Y}{Y} - \frac{15\sin Y}{Y^2}\right),
\]

\[
W_{4(5)}^{(1)}(z) = \begin{cases} 
(1 - z)^6(8 + 19z + 15z^2) & \text{at } 0 \leq z \leq 1, \\
0 & \text{at } z > 1,
\end{cases}
\]

\[
\Omega_{4(5)}^{(1)}(0) = \frac{21}{8R_0}.
\]

The relations specifying one more pattern of this set are of the form

\[
g_{5(6)}(r) = \frac{21(1 - x)^4(1 + 4x)}{2\pi R_0^3},
\]

\[
S_{5(6)}(h) = \frac{5040}{Y^6}\left[4 - \cos Y + \frac{9\sin Y}{Y} - \frac{24(1 - \cos Y)}{Y^2}\right],
\]

\[
W_{5(6)}^{(1)}(z) = \begin{cases} 
(1 - z)^6(1 + 3z + 3z^2) & \text{at } 0 \leq z \leq 1, \\
0 & \text{at } z > 1,
\end{cases}
\]

\[
\Omega_{5(6)}^{(1)}(0) = \frac{3}{R_0}.
\]

Interested in the cases of \(s \leq 6\), we will not propose the complete set of values characteristic of the next example. However, it is worth pointing out that the structure of the corresponding spreading function becomes of a more complicated form again and is as follows:

\[
g_{7(7)}(r) = \frac{45(1 - x)^5(1 + 5x + 8x^2)}{4\pi R_0^3},
\]

The corresponding concomitant quantities take the form

\[
S_{7(7)}(h) = \frac{75600}{Y^7}\left[-\sin Y + \frac{24 - 15\cos Y}{Y} + \frac{87\sin Y}{Y^2} - \frac{192(1 - \cos Y)}{Y^3}\right],
\]

\[
\Omega_{7(7)}^{(1)}(0) = \frac{25}{8R_0}
\]

and are sufficient to describe point-charge lattices at least while different spreading functions of form \((A.13)\) do not overlap.

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