Renormalization method for infinite lattice sums revisited: lattice sums with Bloch phase factor

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Abstract

Infinite lattice summation scheme based on the idea of renormalization is generalized to enable evaluation of infinite lattice sums with Bloch phase factors which can occur when treating long-range interactions in infinite periodic systems. The scheme is fast, with easy to control accuracy and is not limited to any choice of special points in the Brillouin zone. Illustrative calculation for a first few contributions for a simple cubic lattice is presented.

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I. INTRODUCTION

In electronic structure calculations of infinite or large finite systems the Laplace expansion of Coulomb potential \((R > a)\)

\[
\frac{1}{|R-a|} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} I_{lm}(R)R_{lm}^*(a)
\]

(1)

is an indispensable frequently used factorization tool when treating distant interactions. In (1), \(R\) and \(I\) are the scaled regular and irregular solid harmonics, respectively.

\[
R_{lm}(a) = \frac{1}{(l+m)!} P_{lm}(\cos \theta_a) e^{im\phi_a},
\]

(2)

\[
I_{lm}(R) = (l-m)! \frac{1}{R^{l+1}} P_{lm}(\cos \theta_R) e^{im\phi_R}
\]

(3)

\(P\) are the Legendre polynomials and \(R = (R, \theta_R, \phi_R), a = (a, \theta_a, \phi_a)\) in spherical coordinates. Apart from scaling factors \(R\) and \(I\) correspond to multipole moments and their Taylor counterparts.

In a number of calculations (e.g., post-Hartree-Fock density fitting calculations in extended systems) Bloch sums of (1) occur and the quality of long-range incorporation depends on how effectively one can cope with infinite lattice sums of the type

\[
\sigma_{lm}(q) = \sum_{R > R_0}^{\infty} e^{iq.R} I_{lm}(R).
\]

(4)

In (4), \(R_0\) indicates the short-range region of the infinite periodic lattice and \(q\) is a wave vector.

For one-dimensional periodicity (1D) infinite summations of the type (4) do not present a problem. For \(q = 0\) they lead to Riemann zeta functions, for general \(q \neq 0\) analytical formulas using Bernoulli numbers were derived. Beyond 1D, for high \(l\) one can profit from the fast decay of \(I(R)\) which restricts the range of summation to a reasonable size. Anyhow, for the lowest orders of \(I\) brute force summations (if convergent at all) hardly return a sufficiently accurate answer in a reasonable time.

For the \(q = 0\) case, several efficient techniques were already published. The idea which dates back to Nijboer and De Wette is based on splitting the infinite sum into two parts using Ewald-like partitioning. Treating each part separately one gets a rapidly convergent direct space term and a fast converging term in reciprocal space. The idea was recast in a computationally more suitable form by Challacombe et al.
An alternative way of treatment is based on a renormalization scheme. Looking at the multipole expansion at two different structural scales Bermman and Greengard\textsuperscript{8} succeeded in finding a relation between multipole expansion coefficients from which a recurrence formula for infinite lattice sums follows. The scheme was reinvestigated by Kudin and Scuseria\textsuperscript{9} who formulated the idea of renormalization in terms of rescaling and translation operators. Their recurrence relation for infinite lattice sums is more natural to work with since each iterative step can be interpreted as a contribution from certain part of the infinite lattice.

The simple elegant idea behind the renormalization method, the fast convergence and implementational ease make the scheme challenging for trying to extend the technique to infinite sums of the type (4) for nonzero \( q \). When attempting to do so one has to find a way how – in addition to profiting from the scaling properties of \( R, I \) – to cope with the Bloch phase factor scaling. To the author’s knowledge the only attempt to generalize the scheme of Kudin and Scuseria\textsuperscript{9} beyond \( q = 0 \) was published by Grundei and Burow (Appendix A of Ref. 4) where the authors simply got rid of the phase scaling problem for the price of limiting their scheme to the restriction to \( q \) being a fractional part of a reciprocal lattice vector, \( q = K/n \). Selecting \( q \) in this special form for a suitable choice of the initial cluster always returns an integer multiple of \( 2\pi \) phase and the phase scaling problem does not occur. We show that an easy way how to solve the problem for completely general \( q \) without any restriction to special type of wave vector exists for only modest additional costs.

In Sec. II we review the ideas of renormalization method in easy to follow geometric way first. Using this picture we derive the recurrence formula for fast evaluation of lattice sum (4) for general nonzero wave vector which does not suffer from any restrictions imposed on \( q \). In Sec. III numerical issues are discussed and an illustrative calculation for a simple cubic lattice is presented.

II. METHOD

The basic idea behind the technique is the same as for the \( q = 0 \) case\textsuperscript{9} however, our way of derivation is free of involved operator manipulations, it offers a simple geometric insight and enables an easy generalization to the nonzero wave vector case.

To understand the essence of the method let us describe the way how the infinite lattice is generated first. We will consider a system with translational periodicity in three dimensions
(3D). By a straightforward simplification, the scheme can be applied to periodicity in one or two dimensions as well.

The idea is illustrated in Fig. 1. A cluster consisting of the unit cell centered at the origin and its first \( n_{WS} \) neighbour shells (\( n_{WS} \) is the analogy of the well-separatedness criterion\(^{2,10}\)) we will refer to as the central cluster (CC). We will distinguish the CC (with a general integer \( n_{WS} > 0 \)) from a special case of CC with \( n_{WS} = 1 \) (always of size \( n_0 = 3 \times 3 \times 3 \) cells) which we will call the nearest neighbours cluster (NNC).

Consider now a layer of cells next to CC consisting of all nearest neighbour replicas of CC (its edge being 3-times the edge of CC). We denote the number of cells in this layer by \( N_0 \) (Layer I in Fig. 1). Let us label each cell by its lattice translation vector. When we stretch the lattice translation vector \( R_0 \) associated with a cell from Layer I by a factor of 3 we get a new lattice translation vector \( R_0' = 3R_0 \) which is now a center of \( 3 \times 3 \times 3 \) supercell (a periodic replica of NNC) in the next layer (Layer II in Fig. 1). When we repeat the process with all \( R_0 \) from Layer I we end up with a completely filled Layer II. Evidently, there are \( N_0 \) supercells of \( n_0 \) cells each in Layer II and the complete set of lattice translation vectors of all cells from Layer II consists of \( N_1 = N_0 \times n_0 \) vectors \( R_1 = R_0' + a = 3R_0 + a \), where \( a \) runs over all \( n_0 \) lattice translation vectors of the NNC.

This process can now be repeated recursively until sufficiently large cluster is generated. From the way of construction it follows that the relation between sizes of two successive layers will always be \( N_{n+1} = N_n \times n_0 \) so that the size of a layer exhibits geometric growth. Consequently, large enough cluster can be generated within a small number of recursive steps in this way.

Let us return to the evaluation of infinite sums (4) now. Decomposing the infinite sum into contributions from all layers we have

\[
\sigma_{lm}(q) = \sigma_{lm}^{(0)}(q) + \sigma_{lm}^{(1)}(q) + \ldots + \sigma_{lm}^{(n)}(q) + \ldots
\]

where

\[
\sigma_{lm}^{(n)}(q) = \sum_{R_n} e^{i\mathbf{q} \cdot \mathbf{R_n}} \mathcal{I}_{lm}(\mathbf{R_n})
\]

and the summation in (6) runs over all the \( N_n \) lattice translation vectors \( \mathbf{R_n} \) of the \( n \)-th layer. Let us suppose we already know \( \sigma_{lm}^{(n)}(q) \). Considering the way how layers were constructed
the contribution from the next layer will be

\[ \sigma_{lm}^{(n+1)}(q) = \sum_{R_{n+1}} e^{i q \cdot R_{n+1}} T_{lm}(R_{n+1}) \]

\[ = \sum_{R_n} e^{i q \cdot (3R_n + a)} T_{lm}(3R_n) \sum_a e^{i q \cdot a} \mathcal{R}^*_j(-a) \]

\[ = \sum_{jk} \xi_{l+j} \sum_{R_n} e^{i 3q \cdot R_n} T_{l+j,m+k}(R_n) M_{jk}^*(q). \]  

(7)

Use was made of the addition theorem for irregular solid harmonics\textsuperscript{11} which in normalization \textsuperscript{2–3} reads

\[ T_{lm}(R - a) = \sum_{j=0}^{\infty} \sum_{k=-j}^j T_{l+j,m+k}(R) \mathcal{R}^*_j(a). \]  

(8)

The factor \( \xi = 1/3^{l+1} \) is a result of scaling property of \( T \). Comparison with (3) yields the recurrence formula

\[ \sigma_{lm}^{(n+1)}(q) = \sum_{j=0}^{\infty} \sum_{k=-j}^j \sigma_{l+j,m+k}^{(n)}(3q) M_{jk}^*(q), \]  

(9)

or, in more compact form using the symbolics of Ref. 9,

\[ \sigma^{(n+1)}(q) = \mathcal{U}_L[\sigma^{(n)}(3q)] \otimes M^*(q). \]  

(10)

In (9) \( M(q) \) was introduced

\[ M_{jk}(q) = \sum_a e^{i q \cdot a} \mathcal{R}_{jk}(a) \]  

(11)

where the summation runs over all \( n_0 \) lattice translation vectors of the NNC. The recurrence relation (10) is the key formula of our scheme. As expected, for \( q = 0 \) it simplifies to a formula equivalent to Eq. (24) of Ref. 9, the formal difference is that we evaluate the layer contributions while in Ref. 9 the partial sums are treated directly.

III. CALCULATIONAL DETAILS AND DISCUSSION

Similar to the \( q = 0 \) scheme\textsuperscript{9} once we know \( \sigma^{(0)} \) and \( M \) all the contributions to full \( \sigma \) can be evaluated recursively using (10). Notice, however, that for knowing e.g. \( \sigma^{(1)}(q) \) we need to know \( \sigma^{(0)}(3q) \), etc. Consequently, to get \( \sigma^{(n)}(q) \) we need to start the recurrence
from $\sigma^{(0)}(3^n\mathbf{q})$. At the same time, $M(3^n\mathbf{q})$ will also be required, $m = 0, 1, \ldots, (n-1)$. This is the additional expense we have to pay in our scheme. Notice that there is no danger of numerical overflow for any $3^n\mathbf{q}$ since – owing to periodicity of $\sigma(\mathbf{q})$ in reciprocal space – each wave vector can be always kept within the first Brillouin zone (BZ) by a suitable reciprocal lattice translation.

Compared with the $\mathbf{q} = \mathbf{0}$ case general nonzero $\mathbf{q}$ calculation converges considerably faster. Typically, 8-9 iterations are sufficient for 16 digit accuracy for a general $\mathbf{q}$-vector from inside BZ for $n_{WS} = 1$ and $l \geq 3$. For special points at BZ edge number of iterations varies between 7 for $\mathbf{q} = (1/2, 1/2, 1/2)$ and 11 for $\mathbf{q} = (1/2, 0, 0)$ (in reciprocal lattice vector units) while for $\mathbf{q} = \mathbf{0}$ (and for any other reciprocal lattice vector) 16 iterations are needed for 16 digit convergence. For small $\mathbf{q}$ in symmetry positions number of iterations can be similar to the $\mathbf{q} = \mathbf{0}$ case.

Other numerical issues behave in a way similar to that in Ref.9. The infinite summation over angular momentum in (9) has to be truncated to some finite value $l_{max}$ in practice. We found $l_{max} = 40$ sufficient for all $\sigma_{lm}$ to be saturated. It is natural to require to have also large enough $l_{max}$ for which the results are already not sensitive to the choice of $n_{WS}$, i.e., for which the computationally least demanding choice $n_{WS} = 1$ is sufficient. To be able to compare the effect of the choice of $n_{WS}$ we unified $R_0$ in (4) for calculations with different $n_{WS}$ first. We set $R_0 = \mathbf{0}$ and the missing finite part beyond $\mathbf{R} = \mathbf{0}$ was added to each $\sigma_{lm}$ to make the comparison for different $n_{WS}$ possible. In all cases, the differences between $n_{WS} = 1$ and $n_{WS} = 2$ were below 15th decimal place for $l_{max} = 40$.

Working in double precision arithmetics, the symmetry-expected zero imaginary (or, real) parts of infinite sums we found all to be within $10^{-15}$ accuracy of corresponding $|\sigma|$. The zero-to-be sums were also within 15 digit accuracy compared with nonzero terms of the same $l$ (or, close $l$ if $\sigma_l = 0$ all). Of course, for large $l$ (where $|\sigma_l| \gg 1$) we can get spurious numbers orders of magnitude above $10^{-15}$ instead of true zeros. If wishing to get rid of these artifacts Legendre polynomials have to be evaluated in quadruple precision arithmetics.

In Table I lattice sums starting from $l = 3$ (the lowest order free of possible conditional convergence for 3D system) up to $l = 5$ are presented for $\mathbf{q} = (0.1, 0.1, 0.1)$ for a simple cubic lattice with unit lattice constant. For all numbers in Table I Legendre polynomials were evaluated in quadruple precision arithmetics.

Compared to $\mathbf{q} = \mathbf{0}$ case our algorithm is slowed down by the fact that for each $\sigma^{(n)}(\mathbf{q})$ sets
| $l$ | $m$ | $Re\{\sigma_{lm}(q)\}$ | $Im\{\sigma_{lm}(q)\}$ |
|-----|-----|-------------------------|-------------------------|
| 3   | 0   | 0.00000000000000000    | 7.461180731804426       |
| 3   | 1   | -3.730590365902213     | 3.730590365902213       |
| 3   | 2   | 4.575050777090631      | 0.00000000000000000    |
| 3   | 3   | -18.652951829511067    | -18.652951829511067    |
| 4   | 0   | 60.616977645071896     | 0.00000000000000000    |
| 4   | 1   | 0.907115047217165      | 0.907115047217165      |
| 4   | 2   | 0.00000000000000000    | 3.628460188868663      |
| 4   | 3   | 6.349805330520163      | -6.349805330520163     |
| 4   | 4   | 303.084888225359527    | 0.00000000000000000    |
| 5   | 0   | 0.00000000000000000    | 120.923466268988799    |
| 5   | 1   | 51.788023546202568     | -51.788023546202568    |
| 5   | 2   | 0.00000000000000001    | 0.00000000000000000    |
| 5   | 3   | 86.482861899369858     | 86.482861899369858     |
| 5   | 4   | 0.00000000000000000    | 51.533789562654259     |
| 5   | 5   | 984.480915344945550    | -984.480915344945550   |

of $\sigma(0)(3^m q)$ and $M(3^{n-1} q)$ have to be evaluated for each $q$. The special $q$-points scheme does not suffer from these extra expenses either, however, in their scheme the choice of central cluster is governed by the choice of $n$ in $q = K/n$, which for some $q$ makes the evaluations in the central cluster quite demanding.

In our scheme, each $q$-point of our need we get for the same price without any additional complication. Moreover, since the sums are sufficient to be pre-calculated at the initial stage once per calculation (and can be used for the same lattice with the same choice of $q$-points repeatedly) extreme speed of the summation scheme is not an issue that should bother us. The possibility to do the summation with any $q$ without any restriction in a reasonable time with a controllable accuracy is what is usually needed.

In spite of the need to start the recurrence from a new $\sigma(0)(3^m q)$ and have all $M(3^{m-1} q)$ available in each $m$-th recurrence the algorithm can be arranged so that the computational
costs scale linearly with the number of recursive steps. At the same time, number of terms included in summation grows geometrically.

IV. CONCLUSIONS

We generalized the renormalization idea based lattice summation method of Kudin and Scuseria\textsuperscript{9} to enable evaluation of infinite lattice sums with Bloch factor. The scheme is general and is not limited to any special form of the wave vector. As a by-product, we offer a simple novel way of looking at the renormalization scheme.

For a general point from inside the Brillouin zone the number of recurrence steps is typically lower than for the $q = 0$ case. Compared to the $q = 0$ scheme\textsuperscript{9} the method has only modest extra computational expenses.

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Fig. 1. Two-dimensional illustration of lattice generation for $n_{WS} = 2$: the central cluster, Layer I and a part of Layer II. $R'_0$ is the 3× stretched $R_0$ lattice translation vector; the $R_0$-cell and the corresponding supercell around $R'_0$ are set off by bold framing.