Brillouin-zone integration by Fourier quadrature: Special points for superlattice and supercell calculations

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(Received 11 October 1988)

A simple extension of the Monkhorst-Pack Fourier-quadrature (or special-points) method for Brillouin-zone integration makes it possible to determine equivalent (identical by symmetry) points for two different unit cells simply by identifying lattice points common to both cells. This is useful for superlattice or supercell geometries in order to minimize systematic errors.

I. INTRODUCTION

Calculations on superlattices of different orientations, frozen phonons, impurities in supercells, and other superstructures often involve comparisons of results from two or more different unit cells (geometries). All require the evaluation of Brillouin zone integrals, which are invariably approximated using so-called "special points"[1–3] methods. In order to minimize systematic numerical errors, it is advantageous to use the same (equivalent) set of "special points" in each geometry. Thus it becomes necessary to find the corresponding "special points" for one unit cell given a set for another and to determine if the two sets are indeed equivalent. Presented here is an alternate derivation and a simple extension of the method described by Monkhorst and Pack,[3] which simplify this process. The method will also reproduce the Chadi and Cohen points[2] for appropriate choices of parameters.

II. THEORY

We want to approximate the Brillouin zone (BZ) integral of a reciprocal-space function $F(\mathbf{k})$ with a discrete sum:

$$\Omega^{-1}\int_{\mathrm{BZ}} d^3k\, F(\mathbf{k})=\sum_n w_n F(\mathbf{k}_n), \tag{1}$$

where w_n are weight factors. $F(\mathbf{k})$ is periodic and can be expanded in a Fourier series

$$F(\mathbf{k})=\sum_{\mathbf{R}} F_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}}, \tag{2}$$

where $\mathbf{R}$ is a real space lattice vector. Our goal is to choose a small set $\{\mathbf{k}_n, w_n\}$ such that as many small-$\mathbf{R}$ terms as possible in the Fourier expansion of $F(\mathbf{k})$ are integrated exactly. The first term in the Fourier series that fails to integrate exactly is denoted $\mathbf{R}_{\max}$. Notice that this is exactly analogous to, e.g., Gaussian quadrature integration, where the order of the polynomial corresponds to $\mathbf{R}_{\max}$. The appropriate name for this process is therefore *Fourier quadrature* rather than the term "integration by special points."

In the following derivation it is convenient to allow integrations and sums to run unrestricted over all space, not just over the first Brillouin zone. Since $F(\mathbf{k})$ is periodic this will simply multiply the result by the number of Wigner-Seitz cells in the reciprocal lattice. Later translational invariance is used to restrict sums to the first Brillouin zone only.

One-dimensional Fourier quadrature is carried out using equally weighted, equidistant points. In three dimensions, we will try the linear generalization

$$\mathbf{k}_n \rightarrow \mathbf{k}_{\{n_j\}}=\mathbf{f}_0+\mathbf{f}_1 n_1+\mathbf{f}_2 n_2+\mathbf{f}_3 n_3 \tag{3}$$

and

$$w_n \rightarrow w_{\{n_j\}}=w, \tag{4}$$

where $\{\mathbf{f}_j\}$ are vectors in reciprocal space, the last three of which must be linearly independent. Monkhorst and Pack[3] chose them to be fractions of (and parallel to) the primitive translation vectors of the reciprocal lattice; however, as will become clear, this is an unnecessary and undesirable restriction. Combining Eqs. (1), (2), (3), and (4) and extending integrals and sums to all space we obtain

$$\Omega^{-1}\int d^3k\, F(\mathbf{k})=\sum_{\mathbf{R}} c_{\mathbf{R}} F_{\mathbf{R}} \tag{5}$$

with

$$c_{\mathbf{R}}=w e^{i\mathbf{f}_0\cdot\mathbf{R}} \prod_{j=1}^{3} \sum_{n_j=-\infty}^{+\infty} e^{i\mathbf{f}_j\cdot\mathbf{R} n_j}. \tag{6}$$

The correct solution for the integral is obtained if the coefficients $c_{\mathbf{R}}$ are all zero with the exception of c_0, which should be one. This is satisfied provided at least *one* of the inner products $\mathbf{f}_j\cdot\mathbf{R}$ is *not* an integer multiple of 2π, causing one or more of the sums over n_j in Eq. (6) to be zero. Thus, for a given set of vectors $\{\mathbf{f}_j\}$, the first term of the sum in Eq. (2) which fails to integrate correctly is the smallest $\mathbf{R}$ which satisfies

$$\mathbf{f}_j\cdot\mathbf{R}=2\pi\times(\text{integer}), \quad j=1,2,3. \tag{7}$$

This defines the vector $\mathbf{R}_{\max}$.

Equation (7) shows that the integral could be evaluated exactly by making $\mathbf{f}_i$ incommensurate with the reciprocal lattice primitive translation vectors. However, since the

quadrature grid would not be the same from one reciprocal unit cell to the next, this would necessitate summing over all space. To avoid this and reduce the sum to the first Brillouin zone, the grid must have the translational period of the reciprocal lattice. That is

$$\mathbf{k}_{\{n_j\}}+\mathbf{G}=\mathbf{k}_{\{n_j'\}} \tag{8}$$

where $\{n_j\}$ and $\{n_j'\}$ are integers for any reciprocal lattice vector $\mathbf{G}$. Equation (8) is satisfied provided integers $\{m_{ij}\}$ exist such that

$$\sum_{j=1}^{3} m_{ij}\mathbf{f}_j=\mathbf{b}_i, \quad i=1,2,3, \tag{9}$$

where $\{\mathbf{b}_i\}$ are the primitive translation vectors of the reciprocal lattice. Defining a set of vectors $\{\mathbf{g}_i\}$ by the conditions

$$\mathbf{f}_i\cdot\mathbf{g}_j=2\pi\delta_{ij}, \quad i,j=1,2,3. \tag{10}$$

Equation (9) can be written

$$\mathbf{g}_i=\sum_{j=1}^{3}\mathbf{a}_j m_{ji}, \tag{11}$$

where $\{\mathbf{a}_i\}$ are the primitive translation vectors in real space. Since Eq. (11) requires that each $\mathbf{g}_i$ must be a real space lattice vector, Eqs. (7) and (10) show that they coincide with $\mathbf{R}_{\max}$ and define the first terms in the Fourier series, Eq. (2), which fail to integrate exactly.[4]

Since Eq. (11) forces the quadrature grid to have the translational symmetry of the crystal, all the sums can be restricted to the first Brillouin zone. The weight factor, w, determined from Eq. (6) by the requirement $c_0=1$, is equal to the inverse of the number of quadrature points. So far the shift $\mathbf{f}_0$ is arbitrary.

The point-group symmetry of the crystal is normally used to reduce the quadrature grid to the irreducible part of the Brillouin zone by removing points that are related by symmetry. However, if the grid generated by Eq. (3) is without the full point-group symmetry, the point group may also be used to *symmetrize* the grid, creating additional grid points. The resulting grid is still described by Eq. (3), but with a different set of vectors $\{\mathbf{f}_j\}$. The symmetrization may modify the predictions of Eq. (7), changing $\mathbf{R}_{\max}$. (See below for an explanation of this apparent contradiction.) Since two quadrature grids (for different unit cells) are considered equivalent if they have the same $\mathbf{R}_{\max}$, we will avoid this complication by requiring that the grids described by Eq. (3) have the full point-group symmetry. That is,

$$O^{\alpha}\cdot\mathbf{k}_{\{n_j\}}=\mathbf{k}_{\{n_j'\}}, \tag{12}$$

where $\{n_j\}$ and $\{n_j'\}$ are integers for any point-group rotation O^{α}. Equation (12) places additional constraints on $\{\mathbf{f}_i\}$. Consider separately $\{n_i=0\}$ and $\{n_i\neq 0\}$ to obtain the requirements

$$\mathbf{g}_i\cdot O^{\alpha}\cdot\mathbf{f}_0-\mathbf{g}_i\cdot\mathbf{f}_0=2\pi\times(\text{integer}), \quad i=1,2,3 \tag{13}$$

and

$$\mathbf{g}_i\cdot O^{\alpha}\cdot\mathbf{f}_j=2\pi\times(\text{integer}), \quad i,j=1,2,3, \tag{14}$$

for all values of α.

The conditions imposed by Eqs. (13) and (14) simply mean that for an unsymmetrized grid, every vector in a star (as generated by $O^{\alpha}\cdot\mathbf{R}$) must be examined using Eq. (7). If any of the vectors fails to satisfy Eq. (7), all the terms in the star will integrate correctly, despite the prediction of Eq. (7). On the other hand, for a symmetric grid, it is sufficient to examine one member of each star.

III. SUMMARY

The process of selecting and generating a Fourier quadrature grid for Brillouin-zone integration can be summarized as follows.

(i) Select three linearly independent generating vectors $\{\mathbf{g}_i\}$ equal to three lattice vectors for which we tolerate integration error. The shortest of these defines $\mathbf{R}_{\max}$.[4]

(ii) Generate the vectors $\{\mathbf{f}_i\}$, $i=1,2,3$, from $\{\mathbf{g}_i\}$ using Eq. (10).

(iii) Check that the vectors satisfy Eq. (14). If they do not, go back to step (i) and select a new set.

(iv) Select a shift $\mathbf{f}_0$ satisfying Eq. (13). Since the shift has no effect on the quality of the grid, as given by Eq. (7), it should be chosen to optimize step (vi).

(v) Using Eq. (3), generate a grid of all points falling inside the first Brillouin zone and assign to each point a weight equal to the inverse of the number of points.

(vi) Reduce the grid to the irreducible part of the zone by examining each pair of points to see if they are related by an element in the point-group symmetry (with inversion added if missing). If they are, remove one member of the pair from the grid, and add its weight to the weight of the other.

The last two steps are best carried out using a computer.

IV. EXAMPLES

We are now in a position to discuss equivalent quadrature grids for different unit cells. The examples discussed below are superlattice cells based on the zinc-blende structure. The roman numerals refer to the steps outlined in the previous section. We first generate grids for the simple cubic unit cell formed by the conventional cube with edges of length a. Its real space primitive translation vectors are

$$\mathbf{a}_1=a\hat{\mathbf{e}}_x, \quad \mathbf{a}_2=a\hat{\mathbf{e}}_y, \quad \mathbf{a}_3=a\hat{\mathbf{e}}_z. \tag{15}$$

Choose (i)

$$\mathbf{g}_1=na\hat{\mathbf{e}}_x, \quad \mathbf{g}_2=na\hat{\mathbf{e}}_y, \quad \mathbf{g}_3=na\hat{\mathbf{e}}_z, \tag{16}$$

which gives (ii)

$$\mathbf{f}_1=\frac{2\pi}{na}\hat{\mathbf{e}}_x, \quad \mathbf{f}_2=\frac{2\pi}{na}\hat{\mathbf{e}}_y, \quad \mathbf{f}_3=\frac{2\pi}{na}\hat{\mathbf{e}}_z, \tag{17}$$

where n is an integer. These vectors satisfy Eq. (9) for translational symmetry, and they integrate exactly all terms in a Fourier series up to $|\mathbf{R}_{\max}|=na$. They also (iii)

satisfy Eq. (14) for the rotational symmetry of the full cubic group, $m3m$. Select (iv) a shift

$$\mathbf{f}_0=\tfrac{1}{2}(\mathbf{f}_1+\mathbf{f}_2+\mathbf{f}_3), \tag{18}$$

satisfying Eq. (13).

Since the point group of any supercell or superlattice based on the zinc-blende structure must be a subgroup of $m3m$, we need only consider the translational requirement, Eq. (9) or (11), in the following.

A. Face-centered-cubic structure

First let us identify the grids for the face-centered-cubic (fcc) cell. Since every lattice vector of the simple cubic cell above is also a lattice vector of the fcc lattice, all values of n in Eqs. (16) and (17) are allowed. If, for instance, we want our Brillouin-zone integration to be exact for all terms up to $|\mathbf{R}_{\max}|=2a$, choose $n=2$. This (v) gives a grid of 8 points in the simple cubic Brillouin zone and 32 points in the 4 times larger fcc zone. Reducing (vi) the 32 points with the 48 point-group operations gives 2 distinct points in the irreducible zone. These are identical to the 2 "special points" found by Chadi and Cohen.[2] Similarly, choosing $n=4$ and 8 gives the 10 and 60 Chadi-Cohen points, respectively. The choice $n=3$ was not considered by Chadi and Cohen and leads to 6 irreducible points. The only other shift allowed by Eq. (13) is $\mathbf{f}_0=\mathbf{0}$. This gives the same quality grids in terms of $\mathbf{R}_{\max}$, but leads to more quadrature points in the irreducible zone (6, 19, and 85 for $n=2$, 4, and 8, respectively) and is therefore inferior to the shift given by Eq. (18).

B. Superlattices in the [001] direction

Superlattices in the [001] direction have primitive translation vectors[5]

$$\mathbf{a}_1=\frac{a}{2}(\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y),\quad \mathbf{a}_2=\frac{a}{2}(-\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y),\quad \mathbf{a}_3=ma\hat{\mathbf{e}}_z, \tag{19}$$

where $2m$ is the superlattice repeat period [e.g., appropriate for $(GaAs)_m(AlAs)_m$ superlattices]. Equation (11) requires that each $\mathbf{g}_i$ be equal to a lattice vector, so the process of finding an equivalent quadrature grid is simply a matter of identifying common lattice vectors for the simple cubic structure, Eq. (15), and the superlattice structure, Eq. (19). We see that n in Eq. (16) must be a multiple of m. [For a $(GaAs)_4(AlAs)_4$ superlattice, the allowed values are $n=4$, 8, 12, etc.] The number of integration points in the irreducible part of the zone depends on the point-group symmetry of the superlattice. For superlattices with point-group symmetry $\bar{4}m2$ and repeat periods $m=1$–4, we find the points listed in Table I for $n=2$, 3, and 4. These are equivalent to the 2, 6, and 10 fcc points, respectively. An empty weight column means that no equivalent set exists for this combination of m and n. Table I exhausts the possible superlattice periods for which quadrature sets equivalent to the 2, 6, and 10 fcc points can be found. Ren and Dow[6] have applied the original Chadi-Cohen method to [001] and [111] superlattices and the set of 6 points for $m=4$ in Table I is identical to their 12 point set after the latter has been reduced using the $\bar{4}$ symmetry operation (present in [001] $(GaAs)_m(AlAs)_m$ superlattices). Note that although their quadrature sets give the same quality of sampling as

TABLE I. Reduced Fourier quadrature points in units of $2\pi/a$ and weights w for [001] $(AC)_m/(BC)_m$ superlattices.

				w			
n	k_x	k_y	k_z	$m=1$	$m=2$	$m=3$	$m=4$
2	−1/4	1/4	1/4	1/2	1/2		
	−1/4	3/4	1/4	1/2	1/2		
3	−1/2	1/2	1/6	2/27		1/9	
	−1/6	1/6	1/6	4/27		2/9	
	−1/6	1/2	1/6	8/27		4/9	
	−1/6	5/6	1/6	4/27		2/9	
	−1/2	1/2	1/2	1/27			
	−1/6	1/6	1/2	2/27			
	−1/6	1/2	1/2	4/27			
	−1/6	5/6	1/2	2/27			
4	−3/8	3/8	1/8	1/16	1/8		1/8
	−3/8	5/8	1/8	1/16	1/8		1/8
	−1/8	1/8	1/8	1/16	1/8		1/8
	−1/8	3/8	1/8	1/8	1/4		1/4
	−1/8	5/8	1/8	1/8	1/4		1/4
	−1/8	7/8	1/8	1/16	1/8		1/8
	−3/8	3/8	3/8	1/16			
	−3/8	5/8	3/8	1/16			
	−1/8	1/8	3/8	1/16			
	−1/8	3/8	3/8	1/8			
	−1/8	5/8	3/8	1/8			
	−1/8	7/8	3/8	1/16			

TABLE II. Reduced Fourier quadrature points in units of $2\pi/a$ and weights w for [110] $(AC)_m/(BC)_m$ superlattices. The [110] $m=1$ superlattice is identical to the [001] $m=1$ superlattice given in Table I.

				w		
n	k_x	k_y	k_z	$m=2$	$m=3$	$m=4$
2	−1/4	1/4	1/4	1/2		
	−1/4	3/4	1/4	1/4		
	1/4	1/4	1/4	1/4		
3	−1/2	1/2	1/6		1/9	
	−1/2	1/2	1/2		1/18	
	−1/6	1/6	1/6		2/9	
	−1/6	1/6	1/2		1/9	
	−1/6	1/2	1/6		2/9	
	−1/6	1/2	1/2		1/9	
	1/6	1/6	1/6		1/9	
	1/6	1/6	1/2		1/18	
4	−3/8	3/8	1/8	1/16		1/8
	−3/8	3/8	3/8	1/16		1/8
	−3/8	5/8	1/8	1/16		1/16
	−3/8	5/8	3/8	1/16		1/16
	−1/8	1/8	1/8	1/16		1/8
	−1/8	1/8	3/8	1/16		1/8
	−1/8	3/8	1/8	1/8		1/8
	−1/8	3/8	3/8	1/8		1/8
	−1/8	5/8	1/8	1/16		
	−1/8	5/8	3/8	1/16		
	1/8	1/8	1/8	1/16		1/16
	1/8	1/8	3/8	1/16		1/16
	1/8	3/8	1/8	1/16		
	1/8	3/8	3/8	1/16		

TABLE III. Reduced Fourier quadrature points in units of $2\pi/a$ and weights w for [111] $(AC)_m/(BC)_m$ superlattices.

				w	
n	k_x	k_y	k_z	$m=1$	$m=2$
2	−3/4	1/4	1/4	3/16	
	−1/4	1/4	1/4	3/8	
	−1/4	3/4	1/4	3/8	
	1/4	1/4	1/4	1/16	
4	−5/8	1/8	1/8	3/64	
	−5/8	1/8	3/8	3/32	3/16
	−5/8	1/8	5/8	3/32	3/32
	−5/8	3/8	3/8	3/64	3/64
	−3/8	1/8	1/8	3/64	3/32
	−3/8	1/8	3/8	3/32	3/16
	−3/8	1/8	5/8	3/32	3/32
	−3/8	3/8	3/8	3/64	3/32
	−3/8	3/8	5/8	3/32	
	−1/8	1/8	1/8	3/64	3/32
	−1/8	1/8	3/8	3/32	3/32
	−1/8	1/8	5/8	3/32	
	−1/8	3/8	3/8	3/64	
	1/8	1/8	1/8	1/64	1/64
	1/8	3/8	1/8	3/64	

the 10 Chadi-Cohen fcc points for any superlattice where m is even (as is stated in their paper), they are *equivalent* points only for $m=2$ and 4. This means that if, e.g., zinc-blende GaAs is calculated using this quadrature set in a supercell geometry with $m>4$, the result would be different from GaAs calculated with the 10 Chadi-Cohen points in the usual fcc cell.

C. Superlattices in the [110] direction

Superlattices in the [110] direction can be taken to have primitive translation vectors

$$\mathbf{a}_1=m\frac{a}{2}(\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y),\quad \mathbf{a}_2=\frac{a}{2}(-\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y),\quad \mathbf{a}_3=a\hat{\mathbf{e}}_z, \tag{20}$$

where again $2m$ is the superlattice repeat period. Comparing Eqs. (16) and (20) we find that again n must be an integer multiple of m. $m=1$ superlattices are identical in the [110] and [001] directions and Table II lists all remaining quadrature sets equivalent to the 2, 6, and 10 fcc sets for the point group $mm2$.

D. Superlattices in the [111] direction

For superlattices in the [111] direction there is no single set of primitive translation vectors that describe all repeat periods. We treat repeat periods $m=1$ and 2, which are the only ones where quadrature sets equivalent to the 2 and 10 fcc points can be found. No [111] superlattice has a quadrature set equivalent to the 6 fcc points. For $m=1$ the primitive translation vectors are

$$\mathbf{a}_1=\frac{a}{2}(\hat{\mathbf{e}}_x-\hat{\mathbf{e}}_z),\quad \mathbf{a}_2=\frac{a}{2}(\hat{\mathbf{e}}_y-\hat{\mathbf{e}}_z),\quad \mathbf{a}_3=\frac{a}{2}(\hat{\mathbf{e}}_x+\hat{\mathbf{e}}_y+2\hat{\mathbf{e}}_z), \tag{21}$$

and for $m=2$ they are

$$\mathbf{a}_1=\frac{a}{2}(\hat{\mathbf{e}}_x-\hat{\mathbf{e}}_z),\quad \mathbf{a}_2=\frac{a}{2}(\hat{\mathbf{e}}_y-\hat{\mathbf{e}}_z),\quad \mathbf{a}_3=\frac{a}{2}(3\hat{\mathbf{e}}_x+3\hat{\mathbf{e}}_y+2\hat{\mathbf{e}}_z). \tag{22}$$

A little algebra shows that the $\{\mathbf{g}_i\}$ of Eq. (16) fall on lattice points provided $n=2$ and 4 for $m=1$ or $n=4$ for $m=2$. (In general n must be an integer multiple of $3m-m$ mod 3.) Table III lists the resulting quadrature sets for the point-group symmetry $3m$. The set of 10 points for $m=2$ is identical to the one found by Ren and Dow[6] for [111] superlattices.

ACKNOWLEDGMENT

The author thanks D. M. Wood and Alex Zunger for helpful comments and for a critical reading of the manuscript.

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