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Expansion into lattice harmonics in cubic symmetries

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ABSTRACT

On the example of a few sets of sampling directions in the Brillouin zone, this work shows how important the choice of the cubic harmonics is on the quality of approximation of some quantities by a series of such harmonics. These studies led to the following questions: (1) In the case that for a given l there are several independent harmonics, can one use in the expansion only one harmonic with a given l?; (2) How should harmonics be ordered: according to l or, after writing them in terms of $(x^4 + y^4 + z^4)^n$ $(x^2y^2z^2)^m$, according to their degree $q = n + m$? To enable practical applications of such harmonics, they are constructed in terms of the associated Legendre polynomials up to $l = 26$. It is shown that electron momentum densities, reconstructed from experimental data for $ErGa_3$ and $InGa_3$, are described much better by harmonics ordered with q.

1. Introduction

In solids, having a periodic lattice, some quantities as electronic densities, Fermi surfaces, effective masses, and associated quantities (e.g., Compton scattering spectra) are invariant under transformations of the point group of the crystal. This property is expressed by their expansion into a series of lattice harmonics, $H_{l,\nu}(\theta,\varphi)$, of a given symmetry

$$f(\mathbf{p}) = \sum_{1,\nu} f_{1,\nu}(p) H_{1,\nu}(\Theta, \varphi) \tag{1}$$

where the index ν distinguishes harmonics of the same order l. In turn (Θ, φ) are the azimuthal and polar angles, respectively, of the direction $\boldsymbol{p}$ with respect to the lattice coordinate system, and $f_{l,\nu}(p)$ are the radial coefficients of the function $f(\boldsymbol{p})$.

Because of the orthogonality of harmonics, coefficients $f_{l,\nu}(p)$ are expressed by the integrals as follows:

$$f_{l,\nu}(p) = \int_\Omega f(p, \Theta, \varphi) H_{l,\nu}(\Theta, \varphi) sin(\Theta) d\Theta d\varphi \tag{2}$$

However, when values of $f(\mathbf{p})$ are known only along some limited number (N) of sampling directions (Θ_i, φ_i), either one cannot perform the exact integration in Eq. (2) or it is difficult, particularly in the case of cubic harmonics (CHs). In such a case, by assuming that series (1) converges and truncating it at a certain term, one can consider a set of algebraic linear equations with N unknown functions $f_{l,\nu}^a(p)$ (superscript "a" emphasizes that each truncation leads to an approximate value). Two

ways of determining $f_{l,\nu}^a(p)$ are described in detail in Ref. [1] – they are the Houston method [2] (commonly used) and the method based on the orthogonality relation (2). The smallest truncation error in calculating $f_{l,\nu}^a(p)$ provides the application of special directions (SDs) – such sampling directions, which take into account the replacement of integral (2) by a finite sum, require the use of the Gaussian quadratures. SDs were proposed for the first time by Bansil [3].

The expansion into lattice harmonics, which simplifies many theoretical calculations [4–10], allows also for providing analytical solutions to some inverse problems. They occur in, for example, medical tomography [11,12], determinations of the Fermi surface from de Haas van Alphen effect [13–16], or reconstructions of electron densities from the Compton scattering, and angular correlations of annihilation radiations (ACARs) spectra [17–23], when applying transform methods [11] (analytical inversion of the Radon transform [24]). The exception is the direct Fourier transform method, in which one can follow two different approaches: either an expansion into a harmonics series or some interpolation [25]. Series expansions method, known as algebraic reconstruction techniques [26–30], needs to use neither Eq. (1) nor SDs.

Lattice harmonics are linear combinations of the spherical harmonics $Y_{l,m}$, which are solutions of the angular momentum operator. Therefore, it is common practice that they are ordered with respect to l [13]. However, due to constructing SDs [31], Fehlner and Vosko [32] proposed CHs, denoted later as $K_{l,q}$, which are not ordered by l but by q. Do such harmonics describe the reality? On the example of "experimental" electron densities for $ErGa_3$ and $InGa_3$, it is demonstrated that for these materials such harmonics seem to be the best.

Table 1
The coefficients $c_{l,m}$ for harmonics $F_{l,\nu}$ and $K_{l,q}$ given in terms of the associated Legendre polynomials as in Eq. (3).

l,ν	$m = 0$	$m = 4$	$m = 8$	$m = 12$	$m = 16$
16,1	0.68136	0.27587	0.29049	0.32757	0.51765
16,2	0	0.63705	−0.32999	−0.64798	0.25573
18,1	0.45792	−0.38646	−0.40209	−0.43747	−0.53657
18,2	0	0.14873	−0.63775	0.72334	−0.21895

l,q	$m = 0$	$m = 4$	$m = 8$	$m = 12$	$m = 16$
16,3	0.30179	−0.44896	0.42452	0.726042	0
16,4	0.61088	0.52950	0.11428	0.00668	0.57737
18,3	0.17300	−0.28370	0.43858	−0.83501	0
18,4	0.42398	−0.30163	−0.61323	−0.13177	−0.57952

To study expansion (1), one should apply sampling directions. In this work, a few sets were chosen among 6 and 10 directions for the following reasons: (1) for 6 SDs, using $K_{l,q}$, one could determine a better set than the set given by Fehlner et al. [31]; (2) $K_{l,q}$ describes very particular sampling directions, measured by Hiraoka and co-workers [33], incomparably better than commonly used harmonics [13], which lead to nonsensical results, even for the isotropic component; (3) for $n = 6$ and 10 (also for $n = 15$ and 21), harmonics $K_{l,q}$ allow to estimate almost perfect SDs.

2. Construction of special directions and cubic harmonics

For structures with the unique R-fold axes (tetragonal, trigonal, or hcp lattices), lattice harmonics have very simple form in terms of associated Legendre polynomials: either $P_l(cos\Theta)$ or $P_l^{|m|}(cos\Theta)cos(m\varphi)$. Cubic structures (with three fourfold axes) have quite different lattice harmonics that are a linear combination of such polynomials.

Mueller and Priestley gave explicit forms of 27 CHs up to $l = 30$ (Table 1 in Ref. [13]), which were verified in our previous study [34]. The characteristic of these CHs, denoted later as $F_{l,\nu}$, is that the harmonics with $\nu = 1$ contain both polynomials $P_l(cos\Theta)$ and $P_l^{|m|}(cos\Theta)cos(m\varphi)$

$$F_{l,1} = c_{l,1}\sqrt{\frac{(2l+1)}{4\pi}}P_l(cos\,\theta)$$
$$+ \sum_m c_{l,m}\sqrt{\frac{(2l+1)(l-|m|)!}{2\pi(l+|m|)!}}P_l^{|m|}(cos\,\theta)cos(m\varphi) \quad (3)$$

By contrast, harmonics with $\nu > 1$ do not contain $P_l(cos\Theta)$. Moreover, if for a given l there are three CHs, $F_{l,2}$ does not also contain $P_l^{|4|}(cos\Theta)cos(4\varphi)$. In Eq. (3) $m = 0, 4, 8, \ldots, \leq l$ and $c_{l,m}$ denotes normalization constants. The most efficient way of calculating the Legendre polynomials is to use their recurrence relations.

When for a given l there is only one harmonic, which in the case of CHs takes place for $l = 0, 4, 6, 8, 10,$ and 14, CHs are determined unambiguously. Otherwise, one could define them in different ways by taking their suitable linear combinations – of course, to have always the same number of independent harmonics. However, there are still harmonics ordered by l.

To construct N-SDs the ideal situation is when lines of zeros of a few first harmonics, at least two first, which are omitted in expansion (1), intersect in N points. For CHs [13] this occurs only in the case of 1- and 3-SDs. This is a reason that Fehlner and co-workers [32] and next Prasad and Bansil [6,35] expressed CHs in terms of the functions $Q = x^4 + y^4 + z^4$ and $S = x^2y^2z^2$ (in the Cartesian coordinates), searching

the simultaneous N zeros of appropriate polynomials in the Q–S space (two polynomials for $N \leq 10$ and three for higher N). For such harmonics, roots of $K_{l,q}$ with the same degree q intersect in $M = 0.5q(q+1)$ points. An index q in $K_{l,q}$ denotes that $K_{l,q}$ is a polynomial of degree $q = n + m$ of a product $Q^n S^m$.

Fehlner and Vosko (Table 2 in Ref. [32]) wrote expressions for ten harmonics ($K_{l,q}$). However, because there is some error in the 10th harmonic $K_{18,3}$, they did it only for nine harmonics (i.e., up to $K_{16,3}$). Therefore, in this paper they are calculated, finding the relationships between $K_{l,q}$ and $F_{l,\nu}$. When for a given l there are two harmonics, they are the following:

$$K_{l,q} = c_1 F_{l,1} - c_2 F_{l,2} \text{ and } K_{l,q+1} = c_2 F_{l,1} + c_1 F_{l,2}$$

where $c_2 = \sqrt{1 - c_1^2}$ and $c_1 = 0.335132$ ($l = 12$); $c_1 = 0.442923$ ($l = 16$); $c_1 = 0.377803$ ($l = 18$); $c_1 = 0.508312$ ($l = 20$); $c_1 = 0.512705$ ($l = 22$); $c_1 = 0.590961$ ($l = 26$).

For $l = 24$, there are three harmonics: $K_{24,4} = 0.20301 F_{24,1} - 0.38417 F_{24,2} + 0.90067 F_{24,3}$; $K_{24,5} = 0.5155 F_{24,1} - 0.7401 F_{24,2} - 0.43187 F_{24,3}$ and $K_{24,6} = 0.8325 F_{24,1} + 0.55196 F_{24,2} + 0.04779 F_{24,3}$.

Based on Table 1 in Ref. [13] and the aforementioned relationships, one can obtain $K_{l,q}$ in terms of the associated Legendre polynomials. For harmonics $K_{l,q}$, it is characteristic that all of them contain both $P_l(cos\Theta)$ and $P_l^{|m|}(cos\Theta)cos(m\varphi)$, as given in Eq. (3). However, when for a given l there is more than one harmonic, $K_{l,q}$ with the lowest q does not contain $P_l^{|m|}(cos\Theta)cos(m\varphi)$ with the highest m – some examples are presented in Table 1. Table 2 illustrates their order of occurrence in expansion (1).

In the next subsections, how to use some sets of SDs when either harmonics $F_{l,\nu}$ [13] or $K_{l,q}$ [32] are applied is demonstrated. Weights of all components $f_{l,\nu}^a(p)$ were determined both by the Houston method and
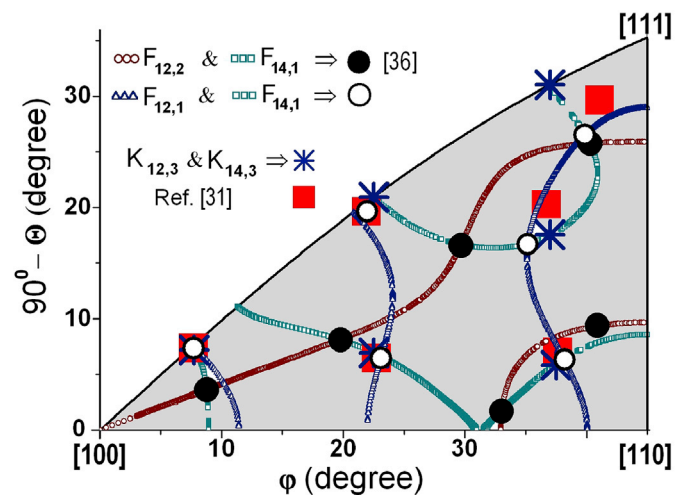

Fig. 1. Loci of zeros of harmonics $F_{12,1}$, $F_{12,2}$, and $F_{14,1}$ and four sets of 6-SDs, drawn in the irreducible part of the Brillouin zone with the high symmetry directions in the corners. Three sets are designated by the common roots (or are close to them) of two CHs: $F_{12,2}$ & $F_{14,1}$; $F_{12,1}$ & $F_{14,1}$; and $K_{12,3}$ & $K_{14,3}$. The set marked by the full squares [31] based on the common roots of two polynomials is the linear combination of harmonics K (see Table 4 in Ref. [32]).

Table 2
The order of occurrence in expansion (1) of CHs: $F_{l,\nu}$ [13] and $K_{l,d}$ [32].

no.→	**6**	7	8	9	**10**	11	12	13	14	**15**	16
$l,\nu \rightarrow$	**12,1**	12,2	14,1	16,1	**16,2**	18,1	18,2	20,1	20,2	**22,1**	22,2
$l,q \rightarrow$	**12,2**	12,3	14,3	16,3	**18,3**	16,4	18,4	20,4	22,4	**24,4**	20,5

no.→	17	18	19	20	**21**	22	23	24	25	26	27
$l,\nu \rightarrow$	24,1	24,2	24,3	26,1	**26,2**	28,1	28,2	28,3	30,1	30,2	30,3
$l,q \rightarrow$	22,5	24,5	26,5	28,5	**30,5**	24,6	26,6	28,6	30,6	28,7	30,7