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A general algorithm for generating isotropy subgroups in superspace

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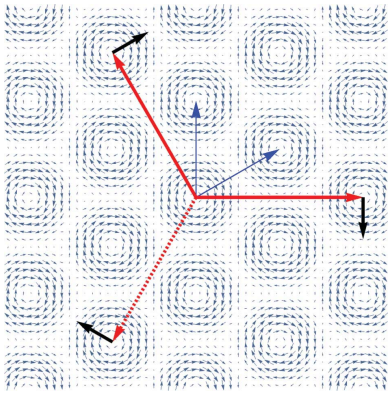
This paper presents a general algorithm for generating the isotropy subgroups of superspace extensions of crystallographic space groups involving arbitrary superpositions of multi-**k** order parameters from incommensurate and commensurate **k** vectors. Several examples are presented in detail in order to illuminate each step of the algorithm. The practical outcome is that one can now start with any commensurate parent crystal structure and generate a structure model for any conceivable incommensurate modulation of that parent, fully parameterized in terms of order parameters of irreducible representations at the relevant wavevectors. The resulting modulated structures have $(3 + d)$-dimensional superspace-group symmetry. Because incommensurate structures are now commonly encountered in the context of many scientifically and technologically important functional materials, the opportunity to apply the powerful methods of group representation theory to this broader class of structural distortions is very timely.

1. Introduction

The study of structural phase transitions in crystalline solids has greatly benefited from group-theoretical methods (Bradley & Cracknell, 1972; Birman, 1978; Authier, 2003; Kovalev, 1993; Toledano & Toledano, 1987; Howard & Stokes, 2005). Most commonly, the symmetry of some parent phase is lowered by the onset of a distortion, so that the symmetries of the parent and distorted phases have a group–subgroup relationship. The distortion, which might involve not only atomic displacements, but also lattice strain, site occupation or magnetic spin, can be classified as belonging to one or more irreducible representations (IRs) of the space-group symmetry of the parent phase.

This IR classification gives us predictive power. Given one or more IRs of a space group, we can use group-theoretical methods to calculate the possible subgroup symmetries that can arise from distortions belonging to those IRs. Such subgroups are called isotropy subgroups. For a given set of IRs, the complete list of possible isotropy subgroups is finite and can be calculated. Knowing the isotropy subgroup of a distorted structure facilitates the complete parameterization of the physical distortion in terms of symmetry-constrained order parameters of the relevant IRs.

The technical definition of an isotropy subgroup is as follows: given an n-dimensional matrix representation D of a space group G_0 and some n-dimensional vector $\boldsymbol{\eta}$ called the order parameter direction (OPD), the isotropy subgroup with respect to D and $\boldsymbol{\eta}$ is the set of all operators g in G_0 which satisfy



$$D(g)\boldsymbol{\eta} = \boldsymbol{\eta}. \quad (1)$$

For the case of superposed IRs, the representation would contain each IR and would not of itself be reducible. For a given representation, we consider all possible OPDs. Even though the possible number of OPDs is infinite, the resulting number of distinct isotropy subgroups is finite. Of course, once we find an isotropy subgroup, we cast the OPD into its most general form for which equation (1) is satisfied simultaneously for every operator g in the subgroup. Generating a list of isotropy subgroups simply means finding all possible solutions to equation (1) for all possible vectors $\boldsymbol{\eta}$. This paper describes the algorithm for accomplishing this seemingly simple task.

Almost 30 years ago, Stokes & Hatch (1988) implemented on computer an algorithm for generating isotropy subgroups and their corresponding OPDs and published a complete list of isotropy subgroups for all IRs associated with special **k** vectors (**k** points of symmetry) for each of the 230 crystallographic space groups. Since then, the computer programs *ISOTROPY*, *ISODISTORT* (Campbell *et al.*, 2006) and *ISOSUBGROUP* (Stokes *et al.*, 2016), available in the *ISOTROPY* software suite (http://stokes.byu.edu/iso/isotropy.php), have greatly extended the scope of the original table by providing real-time calculation of isotropy subgroups for IRs associated with non-special **k** vectors, and even arbitrary superpositions of special and non-special **k** vectors, including both magnetic and non-magnetic cases.

New crystallographic and group-theoretical infrastructure for generating the isotropy subgroups has greatly facilitated our understanding of symmetry breaking in crystals *via* (i) the fast and convenient generation of distorted structure models, (ii) the enumeration, classification and parameterization of families of structures arising from phase transitions (Howard & Stokes, 2004, 2005; Carpenter & Howard, 2009; Mshumi *et al.*, 2014), (iii) parametric studies of order-parameter evolution (Müller *et al.*, 2010, 2014), (iv) the symmetry-adapted exploration of energy landscapes and transition mechanisms (Hatt *et al.*, 2010; Wojdeł & Íñiguez, 2010; Stroppa *et al.*, 2011; Yamauchi *et al.*, 2011), and (v) the symmetry-enhanced solution and compact description of complex and/or subtle structural distortions (Peel *et al.*, 2012; Senn *et al.*, 2012; Campbell *et al.*, 2015; Khalyavin *et al.*, 2015; Lewis *et al.*, 2016).

A decade ago, the systematic symmetry-mode parameterization of crystal distortions at incommensurate **k** vectors (Stokes *et al.*, 2007) was not possible due to a lack of critical infrastructure. At that time, there were no exhaustive tables of $(3+d)$-dimensional superspace groups (SSGs) for $d = 2$ or $d = 3$ (Stokes *et al.*, 2011), no efficient algorithms for establishing the equivalence of SSGs from their operators (van Smaalen *et al.*, 2013), no IRs of superspace extensions of space groups (SSESGs) (Stokes *et al.*, 2013), and no method of generating isotropy subgroups of SSESGs or comparing them to known $(3+d)$-dimensional SSGs. We present the solutions to these last two problems. The projection of IR basis functions at incommensurate **k** vectors will be treated in a later work.

Through the recent implementation of all of these new innovations, the *ISOTROPY* software suite now allows one to generate isotropy subgroups that arise from arbitrary combinations of commensurate and incommensurate **k** vectors, while simultaneously parameterizing a variety of physical order parameters (*i.e.* lattice strains, atomic displacements, magnetic moments, occupancy variations and rigid-body rotations) that can coexist and cooperate in such a distortion. Though we will not discuss those tools in any detail, interested readers can use them to gain immediate practical access to the novel theoretical developments presented here.

2. Modulation vectors

A modulated distortion is characterized by the **k** vectors of its physical modulation waves. Given a space group G_0, the little group G_0^k of modulation vector **k** is the subgroup of G_0 containing all operators for which the point part R leaves **k** invariant [obeys the relation $\mathbf{k}\cdot R^{-1} = \mathbf{k}$ (mod **K**), where **K** is a vector of the reciprocal lattice]. The star of **k** is the set of reciprocal-space vectors contained in the orbit of **k** with respect to G_0^k.

To make these concepts more relevant to SSGs, where **k** and $-\mathbf{k}$ both correspond to the same internal superspace dimension, Stokes *et al.* (2013) defined the little group of $\pm\mathbf{k}$ (denoted by $G_0^{k\bar{k}}$) as containing all operators for which $\mathbf{k}\cdot R^{-1} = \pm\mathbf{k}$ (mod **K**), and defined the star of $\pm\mathbf{k}$ as the set of reciprocal-space vectors contained in the orbit of **k** with respect to $G_0^{k\bar{k}}$. If no operator in G_0 takes **k** into $-\mathbf{k}$, or if $\mathbf{k} = -\mathbf{k}$ (mod **K**) [such as $\mathbf{k} = (0, 0, 1/2)$ for a space group in a primitive setting], then $G_0^{k\bar{k}} = G_0^k$; otherwise, $G_0^{k\bar{k}}$ is twice the size of G_0^k and the star of $\pm\mathbf{k}$ is half the size of the star of **k**. For the sake of brevity, from this point on, we will simply use the word 'star' when referring to the set of **k** vectors in the star of $\pm\mathbf{k}$.

Consider a set of modulated distortions that break the symmetry of a parent space group, resulting in some subgroup G_S of the parent. For a given structure, the set of **k** vectors that actually characterize the modulated distortions are called the 'active' **k** vectors. In general, this set can be comprised of subsets of one or more stars in the parent.

The number of rationally independent **k** vectors determines the dimension of the superspace in which the symmetry of the distorted structure is conventionally described. By 'rationally independent', we mean that the irrational parts of the **k** vectors are linearly independent with respect to rational coefficients. In the context of **k** vectors, we will usually shorten 'rationally independent' to simply 'independent'. The active **k** vectors are not necessarily rationally independent.

For a given **k** vector, we will write the star in the parent as $\{\mathbf{k}^{(\mathrm{F})}\}$, where the superscript '(F)' denotes the 'full' star; and we will define $d^{(\mathrm{F})}$ as the number of **k** vectors in $\{\mathbf{k}^{(\mathrm{F})}\}$. Similarly, we will write the set of active **k** vectors as $\{\mathbf{k}^{(\mathrm{A})}\}$, where the superscript '(A)' denotes 'active'; and we will define $d^{(\mathrm{A})}$ as the number of **k** vectors in $\{\mathbf{k}^{(\mathrm{A})}\}$. Lastly, we will write the set of **k** vectors from $\{\mathbf{k}^{(\mathrm{A})}\}$ that are independent as $\{\mathbf{k}^{(\mathrm{I})}\}$, where the superscript '(I)' denotes 'independent'; and we will define

$d^{(\mathrm{I})}$ as the number of **k** vectors in $\{\mathbf{k}^{(\mathrm{I})}\}$. By definition, $\{\mathbf{k}^{(\mathrm{I})}\} \subseteq \{\mathbf{k}^{(\mathrm{A})}\} \subseteq \{\mathbf{k}^{(\mathrm{F})}\}$ so that $d^{(\mathrm{I})} \leq d^{(\mathrm{A})} \leq d^{(\mathrm{F})}$.

If **k** is active, then all other **k** vectors related to **k** by point operations of the subgroup are also active. Thus, $\{\mathbf{k}^{(\mathrm{A})}\}$ contains the full stars of each of its **k** vectors in the subgroup. We could also say that $\{\mathbf{k}^{(\mathrm{A})}\}$ is formed as a union of active stars in the subgroup, which are mutually disjoint by definition. The selection of a given subgroup G_S restricts the possible subsets $\{\mathbf{k}^{(\mathrm{A})}\}$ of $\{\mathbf{k}^{(\mathrm{F})}\}$ in the parent to unions of stars in the subgroup.

3. Superspace dimensions

The symmetries of incommensurate structures are usually described in $(3+d)$-dimensional $[(3+d)\mathrm{D}]$ superspace, where d is the number of incommensurate modulations, each represented by a **k** vector (Janner & Janssen, 1977; de Wolff, 1974; Janssen *et al.*, 2004; van Smaalen, 2007). The 3-dimensional part of the superspace is called the *external* space and the d-dimensional part is called the *internal* space. An operator in $(3+d)$D superspace can be written as an augmented matrix:

$$g = \{R|\mathbf{v},\boldsymbol{\delta}\} = \begin{pmatrix} R & 0 & \mathbf{v} \\ M & \varepsilon & \boldsymbol{\delta} \\ 0 & 0 & 1 \end{pmatrix}, \tag{2}$$

where R is a three-dimensional point operator in external space, ε is a d-dimensional point operator in internal space, **v** is a three-dimensional translation in external space and $\boldsymbol{\delta}$ is a d-dimensional translation in internal space which can be interpreted in terms of phase shifts along each of the d incommensurate modulations of the structure. R, ε, M are integer matrices such that M can be calculated from R and ε using

$$M_{jm} = \sum_{i=1}^{3} k_{ji} R_{im} - \sum_{i=1}^{d} \varepsilon_{ji} k_{im}, \tag{3}$$

where k_{ji} is the ith component of the jth **k** vector.

By convention, the operators of a given SSG are expressed in $(3+d^{(\mathrm{I})})$D superspace in terms of a set $\{\mathbf{k}^{(\mathrm{I})}\}$ of $d^{(\mathrm{I})}$ **k** vectors whose irrational components are linearly independent with respect to rational coefficients. The $d^{(\mathrm{A})}$ active **k** vectors $\{\mathbf{k}^{(\mathrm{A})}\}$ can be generated by performing the operation $\mathbf{k}_i^{(\mathrm{I})} \cdot R_j^{-1}$ for every point operator R_j and every independent vector $\mathbf{k}_i^{(\mathrm{I})}$ of the SSG. Each of the resulting vectors in $\{\mathbf{k}^{(\mathrm{A})}\}$ can be written as linear combinations of the vectors in $\{\mathbf{k}^{(\mathrm{I})}\}$ with integer coefficients:

$$\mathbf{k}_j^{(\mathrm{A})} = \sum_{l=1}^{d^{(\mathrm{I})}} T_{jl} \mathbf{k}_l^{(\mathrm{I})} \pmod{\mathbf{K}}, \tag{4}$$

where the T_{jl} are integers.

SSG operators can also be presented in $(3+d^{(\mathrm{A})})$D superspace, as well as in the conventional $(3+d^{(\mathrm{I})})$D superspace. However, since the $d^{(\mathrm{A})}$ **k** vectors defining the $(3+d^{(\mathrm{A})})$D superspace are not generally independent, the $\varepsilon^{(\mathrm{A})}$, $M^{(\mathrm{A})}$ and $\boldsymbol{\delta}^{(\mathrm{A})}$ parts of the operators $g^{(\mathrm{A})}$ in this superspace are not well defined unless we require that each row and column of $\varepsilon^{(\mathrm{A})}$ contain only one nonzero entry (± 1) which reflects the fact that, in this space, the ε component of an operator merely permutes the active **k** vectors. We then obtain $M^{(\mathrm{A})}$ using equation (3), and obtain the $d^{(\mathrm{A})}$ components of $\boldsymbol{\delta}^{(\mathrm{A})}$ from

$$\delta_j^{(\mathrm{A})} = \sum_{l=1}^{d^{(\mathrm{I})}} T_{jl} \delta_l^{(\mathrm{I})} \pmod 1, \tag{5}$$

using the same integers T_{jl} as in equation (4). Note that when expressed in $(3+d^{(\mathrm{A})})$D superspace, the $d^{(\mathrm{A})}$ phase shifts in the operators are *not* independent, because they must obey equation (5).

This relationship between operators in the $(3+d^{(\mathrm{A})})$D and $(3+d^{(\mathrm{I})})$D superspaces is important since our algorithm generates isotropy subgroups in $(3+d^{(\mathrm{A})})$D superspace. We then want to identify these as entries in a table of SSGs in $(3+d^{(\mathrm{I})})$D superspace.

3.1. Example 1

As an example, consider SSG 143.2.80.4 $P3(\alpha,\beta,0)0(-\alpha-\beta,\alpha,0)0$ [see ISO(3+d)D table at http://iso.byu.edu/iso/isotropy.php]. There are two independent **k** vectors ($d^{(\mathrm{I})}=2$): $\mathbf{k}_1^{(\mathrm{I})} = (\alpha,\beta,0)$ and $\mathbf{k}_2^{(\mathrm{I})} = (-\alpha-\beta,\alpha,0)$. Using the **k**-vector labels of Cracknell *et al.* (1979), we describe the **k** vectors as lying in the '*B*' plane of the first Brillouin zone of space group No. 143 *P*3. There are actually three **k** vectors in $\{\mathbf{k}^{(\mathrm{A})}\}$ for this **k** point: $\mathbf{k}_1^{(\mathrm{A})} = \mathbf{k}_1^{(\mathrm{I})}$, $\mathbf{k}_2^{(\mathrm{A})} = \mathbf{k}_2^{(\mathrm{I})}$ and $\mathbf{k}_3^{(\mathrm{A})} = \mathbf{k}_2^{(\mathrm{A})} R^{-1}(3[001]) = -\mathbf{k}_1^{(\mathrm{I})} - \mathbf{k}_2^{(\mathrm{I})} = (\beta,-\alpha-\beta,0)$, where 3[001] denotes a threefold rotation about the [001] axis. The matrix T in equations (4) and (5) is given by

$$T = \begin{pmatrix} 1 & 0 \\ \bar{1} & 1 \end{pmatrix}. \tag{6}$$

In $(3+2)$D space, the augmented matrix for the operator 3[001] is given by

$$g^{(\mathrm{I})}(3[001]) = \left(\begin{array}{ccc|cc|c} 0 & \bar{1} & 0 & 0 & 0 & 0 \\ 1 & \bar{1} & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ \hline 0 & 0 & 0 & \bar{1} & \bar{1} & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ \hline 0 & 0 & 0 & 0 & 0 & 1 \end{array}\right). \tag{7}$$

In $(3+3)$D space, the matrix for this same operator is given by

$$g^{(\mathrm{A})}(3[001]) = \left(\begin{array}{ccc|ccc|c} 0 & \bar{1} & 0 & 0 & 0 & 0 & 0 \\ 1 & \bar{1} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ \hline 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ \hline 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{array}\right), \qquad (8)$$

where the ε part of the matrix has an extra row and column for the extra $\mathbf{k}$ vector, and now contains only one nonzero element in each row and column, as explained above. From equation (5), the phase shift for $\mathbf{k}_3^{(\mathrm{A})}$ should be $\delta_3^{(\mathrm{A})} = -\delta_1^{(\mathrm{I})} - \delta_2^{(\mathrm{I})}$, which in this case is zero since both $\delta_1^{(\mathrm{I})}$ and $\delta_2^{(\mathrm{I})}$ are zero.

Though the conventional setting for SSGs is in $(3 + d^{(\mathrm{I})})$D superspace, operators have more intuitive meaning in $(3 + d^{(\mathrm{A})})$D superspace, since there really are $d^{(\mathrm{A})}$ fundamental modulations. In the above example, the $\mathbf{k}_3^{(\mathrm{A})}$ modulation is not merely a harmonic of $\mathbf{k}_1^{(\mathrm{A})}$ and $\mathbf{k}_2^{(\mathrm{A})}$. Point symmetries relate all three of the modulations, putting them on equal footing. Because no parent symmetry group was identified in this example, it would make no sense to discuss $\{\mathbf{k}^{(\mathrm{F})}\}$ here, though one could define a parent.

4. Algorithm for generating isotropy subgroups

The SSG symmetry of a modulated structure is not a subgroup of the crystallographic space group G_0 of the unmodulated parent structure, but it is a subgroup of a superspace extension G of G_0. In a SSESG, one adds to each operator in G_0 a continuously infinite set of translations in internal space. See Stokes *et al.* (2013) for a discussion of SSESGs. Given a space group G_0 and its superspace extension G, the central problem of this work is to generate all of the inequivalent isotropy subgroups of G with respect to an IR associated with a $\mathbf{k}$ vector containing at least one irrational component. For now, we treat the case of a single IR, but in §5 we extend the algorithm to multiple superposed IRs.

4.1. Selection of independent k vectors

Step 1. To generate the isotropy subgroups with $d^{(\mathrm{I})}$ independent $\mathbf{k}$ vectors, we try every inequivalent set of $d^{(\mathrm{I})}$ independent vectors from $\{\mathbf{k}^{(\mathrm{F})}\}$. Note that two sets of $\mathbf{k}$ vectors are equivalent if some external point operator R of G takes one set into the other, to within a permutation of the vectors in the set.

4.2. Point group H_A of all possible active k vectors

Step 2. For a given set $\{\mathbf{k}^{(\mathrm{I})}\}$ of $d^{(\mathrm{I})}$ $\mathbf{k}$ vectors, we find the external point operators of G which take each of the vectors in $\{\mathbf{k}^{(\mathrm{I})}\}$ into linear combinations of the vectors in $\{\mathbf{k}^{(\mathrm{I})}\}$. This may generate extra $\mathbf{k}$ vectors not in $\{\mathbf{k}^{(\mathrm{I})}\}$, which together with those in $\{\mathbf{k}^{(\mathrm{I})}\}$ form a possible set of active $\mathbf{k}$ vectors. These point operators form a point group H_A, where the subscript 'A' denotes 'active'. H_A is the point group that leaves the set of active $\mathbf{k}$ vectors invariant, to within a permutation of the vectors.

4.3. Subgroups H_S of H_A

Step 3. For a given point group H_A, we find all of the point groups H_S which are subgroups of H_A, and consider them one at a time, beginning with the largest, which is H_A itself, and ending with the smallest, which contains only the identity point operator.

4.4. Active k vectors

Step 4. For a given point group H_S, we let each point operator in H_S act on $\{\mathbf{k}^{(\mathrm{I})}\}$, forming a set $\{\mathbf{k}^{(\mathrm{A})}\}$ of active $\mathbf{k}$ vectors. The extra active $\mathbf{k}$ vectors not in $\{\mathbf{k}^{(\mathrm{I})}\}$ are then written as a linear combination of those in $\{\mathbf{k}^{(\mathrm{I})}\}$, as in equation (4). This generates the coefficients T_{jl} that are needed in the next step. Note that sometimes two sets of inequivalent independent $\mathbf{k}$ vectors can generate equivalent sets of active $\mathbf{k}$ vectors. When this occurs, we detect this equivalence and keep only inequivalent sets of $\{\mathbf{k}^{(\mathrm{A})}\}$ for further consideration. An example of this will be shown below.

4.5. Subgroup G_S of G

Step 5. Given H_S and the associated active $\mathbf{k}$ vectors $\{\mathbf{k}^{(\mathrm{A})}\}$, we next attempt to form from H_S a subgroup G_S of G by adding to each operator of H_S a three-dimensional fractional translation and a set of $d^{(\mathrm{A})}$ phase shifts, one for each $\mathbf{k}$ vector in $\{\mathbf{k}^{(\mathrm{A})}\}$. The three-dimensional lattice of G_S is the same as that of G, so the three-dimensional fractional is simply taken from the corresponding operator in G. We have only the freedom to choose phase shifts for each operator but are restricted by the following two requirements: (i) the operators in G_S must form a group with a valid multiplication table and (ii) the phase shifts must obey the relation in equation (5) using the coefficients T_{jl} determined in the previous step. [See Stokes *et al.* (2011) for more details about finding the allowed phase shifts.] We consider all possible choices of phase shifts consistent with these restrictions. For each choice of phase shifts that obey these restrictions, we obtain a subgroup G_S that leaves $\{\mathbf{k}^{(\mathrm{A})}\}$ invariant.

4.6. Order parameter direction

Step 6. We now must test G_S to determine if it is an isotropy subgroup for the selected IR. We require that a nonzero vector $\boldsymbol{\eta}$ exists which satisfies

$$D(g_S)\boldsymbol{\eta} = \boldsymbol{\eta}, \qquad (9)$$

simultaneously for every operator g_S in G_S. $D(g_S)$ is the n-dimensional IR matrix for the operator g_S, and $\boldsymbol{\eta}$ is a vector in the n-dimensional carrier space of that IR. The vector $\boldsymbol{\eta}$ is called the OPD.

In the ISO-IR tables of Stokes *et al.* (2013), the IR matrices were chosen to contain rows and columns of smaller matrix blocks, one for each of the $d^{(\mathrm{F})}$ vectors of $\{\mathbf{k}^{(\mathrm{F})}\}$, where each row and column contain exactly one nonzero block matrix of

dimension $b = n/d^{(\mathrm{F})}$. Because of this simple structure, the OPD can be written as

$$\boldsymbol{\eta} = (\boldsymbol{\eta}_1, \boldsymbol{\eta}_2, \ldots, \boldsymbol{\eta}_{d^{(\mathrm{F})}}), \tag{10}$$

where $\boldsymbol{\eta}_i$ is a b-dimensional vector corresponding to the ith vector of $\{\mathbf{k}^{(\mathrm{F})}\}$ and where only the vectors $\boldsymbol{\eta}_i$ corresponding to active $\mathbf{k}$ vectors, *i.e.* those of $\{\mathbf{k}^{(\mathrm{A})}\}$, are allowed to be nonzero. Thus, only the rows and columns in the IR matrix corresponding to the active $\mathbf{k}$ vectors enter into equation (9). Note that because the phase shifts for the non-active $\mathbf{k}$ vectors do not enter into equation (9), there was no need to determine them in previous steps.

4.7. Equivalent OPDs

Step 7. Each nonzero solution for $\boldsymbol{\eta}$ arising from equation (9) is an isotropy subgroup, though multiple equivalent isotropy subgroups are often generated *via* this procedure. We eliminate equivalent isotropy subgroups and apply the chain rule (Birman, 1978; Jarić, 1981, 1982) all in one step by eliminating isotropy subgroups with equivalent OPDs. During the search for isotropy subgroups, we compare the OPD $\boldsymbol{\eta}$ with the OPDs $\boldsymbol{\eta}'$ of all previously found isotropy subgroups for the same set $\{\mathbf{k}^{(\mathrm{A})}\}$, regardless of which point group H_S generated it. Specifically, $\boldsymbol{\eta}$ and $\boldsymbol{\eta}'$ are equivalent if there exists an operator g in G for which $\boldsymbol{\eta} = D(g)\boldsymbol{\eta}'$. See Appendix *A* for more details.

4.8. Find G_S in ISO(3+d)D table

Step 8. For each inequivalent isotropy subgroup G_S, we further identify it with one of the entries from the ISO(3+d)D table of SSGs at http://iso.byu.edu/iso/isotropy.php. Because the setting of G_S is in $(3 + d^{(\mathrm{A})})$D superspace, whereas tabulated SSGs are all set in $(3 + d^{(\mathrm{I})})$D superspace, we must convert each tabulated candidate to a setting in $(3 + d^{(\mathrm{A})})$D superspace (see §3) before comparing it to G_S, using a method similar to that described in van Smaalen *et al.* (2013).

4.9. Example 2

4.9.1. Two independent k vectors (example 2*a*).

As an example, we consider IR Σ_1 (SM1) of space-group No. 221 $Pm\overline{3}m$ extended to superspace. This IR belongs to a $\mathbf{k}$ vector on the Σ (SM) line in the first Brillouin zone. The star $\{\mathbf{k}^{(\mathrm{F})}\}$ contains six vectors: $(\alpha, \pm\alpha, 0)$, $(\alpha, 0, \pm\alpha)$, $(0, \alpha, \pm\alpha)$. Let us specifically search for isotropy subgroups associated with $d^{(\mathrm{I})} = 2$ independent modulations. (Step 1) The inequivalent sets of $d^{(\mathrm{I})} = 2$ independent $\mathbf{k}$ vectors are found to be

$$\begin{aligned} &(1)\,\{(\alpha, \alpha, 0), (\alpha, \overline{\alpha}, 0)\} \\ &(2)\,\{(\alpha, \alpha, 0), (\alpha, 0, \alpha)\}. \end{aligned} \tag{11}$$

(Step 2) In set (1) of equation (11), the point operators in G which take $\{\mathbf{k}^{(\mathrm{I})}\}$ into linear combinations of $\{\mathbf{k}^{(\mathrm{I})}\}$ form a point group $H_{\mathrm{A}} = 4/mmm$ with the fourfold axis along the cubic [001] direction. None of the point operators in H_{A} produce any extra $\mathbf{k}$ vectors not in $\{\mathbf{k}^{(\mathrm{I})}\}$ so that in this case $\{\mathbf{k}^{(\mathrm{A})}\} = \{\mathbf{k}^{(\mathrm{I})}\}$. In order to focus attention on the most interesting scenarios, we will pass over set (1) and move on to set (2).

The point group H_{A} for set (2) of equation (11) is $\overline{3}m$ with the threefold axis along the cubic $[\overline{1}11]$ direction. In this case, these point operators do produce an extra $\mathbf{k}$ vector $(0, \alpha, \overline{\alpha}) = (\alpha, \alpha, 0) - (\alpha, 0, \alpha)$ not in $\{\mathbf{k}^{(\mathrm{I})}\}$. This is a possible active $\mathbf{k}$ vector, depending on the point group H_S, as will be seen. (Step 3) Detailed calculations show that only six subgroups H_S of H_{A} result in inequivalent isotropy subgroups for IR SM1: $\overline{3}m$, 32, $2/m$, 2, $\overline{1}$, 1, where the twofold axes in the monoclinic point groups are along the cubic [100] axis.

(Step 4) For $H_S = \overline{3}m$, the action of the point operators produces the active $\mathbf{k}$ vectors $\{\mathbf{k}^{(\mathrm{A})}\} = \{(\alpha, \alpha, 0), (\alpha, 0, \alpha), (0, \alpha, \overline{\alpha})\}$. The first two are independent and the third can be expressed as $\mathbf{k}_3 = \mathbf{k}_1 - \mathbf{k}_2 = (0, \alpha, \overline{\alpha}) = (\alpha, \alpha, 0) - (\alpha, 0, \alpha)$. Therefore, the $d^{(\mathrm{A})}$ phase shifts in each operator in G_S must satisfy $\delta_3^{(\mathrm{A})} = \delta_1^{(\mathrm{A})} - \delta_2^{(\mathrm{A})}$. (Step 5) When we solve for the possible phase shifts in the operators of G_S under this restriction, we find two distinct solutions, (Step 6) only one of which satisfies equation (9) and therefore is an isotropy subgroup for IR SM1: all phase shifts in every operator equal to zero. The resulting OPD is $\boldsymbol{\eta} = (a, 0; 0, 0; a, 0; 0, 0; 0, 0; a, 0)$, where we have used semicolons to separate the parts of the OPD belonging to each of the six arms of $\{\mathbf{k}^{(\mathrm{F})}\}$. The three active $\mathbf{k}$ vectors under consideration are the first, third and sixth arms of that star and give rise to the nonzero parts of the OPD. (Step 8) We identify this isotropy subgroup as 166.2.77.3 $R\overline{3}m(\alpha, \alpha, 0)00(-2\alpha, \alpha, 0)00$ in the ISO(3+d)D table of SSGs (http://iso.byu.edu/iso/isotropy.php). The basis vectors of the isotropy subgroup in terms of those of the parent can be shown to be $(\overline{1}, 0, \overline{1}, 0, 0)$, $(0, \overline{1}, 1, 0, 0)$, $(\overline{1}, 1, 1, 0, 0)$, $(0, 0, 0, \overline{1}, 0)$, $(0, 0, 0, 0, 1)$ with an origin shift $(0, 0, 0, 0, 0)$.

We can obtain the $\mathbf{k}$ vectors in the setting of the subgroup in terms of the $\mathbf{k}$ vectors in the setting of the parent using information in these basis vectors. We form two transformation matrices from the basis vectors. From the first three components of the first three basis vectors, we obtain a transformation in external space,

$$A^{(\mathrm{ext})} = \begin{pmatrix} \overline{1} & 0 & \overline{1} \\ 0 & \overline{1} & 1 \\ \overline{1} & 1 & 1 \end{pmatrix}, \tag{12}$$

where we put one basis vector in each column. Similarly, from the last two components of the last two basis vectors, we obtain a transformation in internal space,

$$A^{(\mathrm{int})} = \begin{pmatrix} \overline{1} & 0 \\ 0 & 1 \end{pmatrix}. \tag{13}$$

Using these transformations, we can obtain a relationship between the $\mathbf{k}$ vectors in the parent and subgroup settings:

$$k_{ml}^{(\mathrm{s})} = \sum_{i,j} A_{mi}^{(\mathrm{int})} k_{ij}^{(\mathrm{p})} A_{jl}^{(\mathrm{ext})}, \tag{14}$$

where $k_{ij}^{(\mathrm{p})}$ is the jth component of the ith $\mathbf{k}$ vector in the setting of the parent, and $k_{ml}^{(\mathrm{s})}$ is the lth component of the mth $\mathbf{k}$ vector in the setting of the subgroup. In the present case,

$\mathbf{k}_1^{(p)} = (\alpha, \alpha, 0)$ and $\mathbf{k}_2^{(p)} = (\alpha, 0, \alpha)$ (the two independent $\mathbf{k}$ vectors), so, using equation (14), we obtain $\mathbf{k}_1^{(s)} = (\alpha, \alpha, 0)$ and $\mathbf{k}_2^{(s)} = (-2\alpha, \alpha, 0)$, which are identical to the $\mathbf{k}$ vectors in the SSG symbol for the subgroup.

For $H_S = 32$, we obtain the same three active $\mathbf{k}$ vectors as above, resulting in $\boldsymbol{\eta} = (a, b; 0, 0; b, a; 0, 0; 0, 0; a, \overline{b})$. We identify this isotropy subgroup as 155.2.77.3 $R32(\alpha, \alpha, 0)00(-2\alpha, \alpha, 0)00$ with the same basis vectors as above but with a nonzero origin shift $(0, 0, 0, 3/4, 1/2)$.

For $H_S = 2/m$, none of the point operators take $(\alpha, 0, \alpha)$ or $(\alpha, \alpha, 0)$ into $(0, \alpha, \overline{\alpha})$, so now we only have two active $\mathbf{k}$ vectors: $\{\mathbf{k}^{(A)}\} = \{\mathbf{k}^{(I)}\}$. There are no restrictions on phase shifts in the operators in G_S since T is a unit matrix. We find a solution where all phase shifts are zero, resulting in $\boldsymbol{\eta} = (a, 0; 0, 0; a, 0; 0, 0; 0, 0; 0, 0)$. We identify this isotropy subgroup as 12.2.17.13 $B2/m(\alpha, \beta, \gamma)00(-\alpha, -\beta, \gamma)$. The basis vectors of the subgroup are $(0, 1, 1, 0, 0)$, $(1, 0, 0, 0, 0)$, $(0, 1, \overline{1}, 0, 0)$, $(0, 0, 0, \overline{1}, 0)$, $(0, 0, 0, 0, 1)$ with zero origin shift. Forming the transformation matrices and applying equation (14) to all three parent $\mathbf{k}$ vectors, we obtain $\mathbf{k}_1^{(s)} = (\overline{\alpha}, \overline{\alpha}, \overline{\alpha})$ and $\mathbf{k}_2^{(s)} = (\alpha, \alpha, \overline{\alpha})$. Comparing these vectors to those in the SSG symbol, we see that the value of α^p in the parent and the values of $\alpha^s, \beta^s, \gamma^s$ in the subgroup are related by $\alpha^s \simeq \beta^s \simeq \gamma^s \simeq -\alpha^p$. These relations are approximate because the subgroup symmetry is sufficiently low to allow the $\mathbf{k}^{(s)}$ vectors to distort slightly into a more general direction.

The results for the three remaining possibilities for H_S are similar to those of $H_S = 2/m$ except that the OPDs are in more general directions. For $H_S = 2$, we obtain 5.2.17.9 $B2(\alpha, \beta, \gamma)0(-\alpha, -\beta, \gamma)0$ with $\boldsymbol{\eta} = (a, b; 0, 0; a, \overline{b}; 0, 0; 0, 0; 0, 0)$; for $H_S = \overline{1}$, we obtain 2.2.1.1 $P\overline{1}(\alpha_1, \beta_1, \gamma_1)0(\alpha_2, \beta_2, \gamma_2)0$ with $\boldsymbol{\eta} = (a, 0; 0, 0; b, 0; 0, 0; 0, 0; 0, 0)$; for $H_S = 1$, we obtain 1.2.1.1 $P1(\alpha_1, \beta_1, \gamma_1)0(\alpha_2, \beta_2, \gamma_2)0$ with $\boldsymbol{\eta} = (a, b; 0, 0; c, d; 0, 0; 0, 0; 0, 0)$. The results of this example can be readily reproduced using the internet-based *ISOSUBGROUP* software.

4.9.2. Three independent k vectors (example 2*b*). It would be instructive at this point to briefly examine the case with three independent $\mathbf{k}$ vectors. The inequivalent sets of $d^{(I)} = 3$ independent $\mathbf{k}$ vectors are found to be

$$\begin{aligned} &(1)\ \{(\alpha, \alpha, 0), (\alpha, \overline{\alpha}, 0), (\alpha, 0, \alpha),\\ &(2)\ \{(\alpha, \alpha, 0), (\alpha, 0, \alpha), (0, \alpha, \alpha)\}. \end{aligned} \tag{15}$$

With either of these sets $\{\mathbf{k}^{(I)}\}$, every external point operator of G takes $\{\mathbf{k}^{(I)}\}$ into linear combinations of $\{\mathbf{k}^{(I)}\}$, so that $H_A = m\overline{3}m$. The point operators produce all six of the $\mathbf{k}$ vectors in $\{\mathbf{k}^{(F)}\}$. In the case where $H_S = m\overline{3}m$, we have $\{\mathbf{k}^{(A)}\} = \{\mathbf{k}^{(F)}\}$ for either of the inequivalent sets $\{\mathbf{k}^{(I)}\}$ in equation (15). We thus consider $H_S = m\overline{3}m$ for only one of the sets in equation (15) to avoid generating equivalent isotropy subgroups. Furthermore, any other subgroup H_S of H_A which generates the same set $\{\mathbf{k}^{(A)}\}$ for both sets $\{\mathbf{k}^{(I)}\}$ in equation (15) should only be considered with one of those sets.

For the present case of $H_S = m\overline{3}m$, we obtain $\boldsymbol{\eta} = (a, 0; a, 0; a, 0; a, 0; a, 0; a, 0)$ and the $(3+3)$D SSG is 221.3.210.7 $Pm\overline{3}m(0, \beta, \beta)000(\beta, 0, \beta)000(\beta, \beta, 0)000$.

Various other subgroups of H_A result in seven additional nonequivalent isotropy subgroups (eight total) for set (1) and seven for set (2), none of which share the same set of active $\mathbf{k}$ vectors.

5. Superposed IRs

When two or more IRs are superposed, the generation of isotropy subgroups becomes more complicated. This could either involve the superposition of multiple incommensurate IRs or the superposition of incommensurate and commensurate IRs. We only consider cases that have at least one incommensurate IR, *i.e.* its $\mathbf{k}$ vectors have at least one irrational component.

5.1. Incommensurate IRs

First of all, consider the relatively simpler case of two or more superposed incommensurate IRs. We follow the same algorithm as in §4 but with the following minor modifications.

In Step 1, we select one or more independent $\mathbf{k}$ vectors for each IR. (Note that if two IRs are associated with the same star, we require that they share the same independent $\mathbf{k}$ vectors.)

In Step 2, we form the point group H_A with point operators that simultaneously take each set of independent $\mathbf{k}$ vectors into linear combinations of themselves.

In Step 6, we require that for each IR, there exists a nonzero OPD which satisfies equation (9) simultaneously for every operator in G_S.

5.2. Incommensurate and commensurate IRs

Now consider the more complicated case when one or more incommensurate IRs are superposed with one or more commensurate IRs. Commensurate IRs are associated with $\mathbf{k}$ vectors having all rational components.

We first generate a list of isotropy subgroups G_C of G that arise from the superposed commensurate IRs alone (see Appendix *B*; the subscript 'C' denotes commensurate $\mathbf{k}$ vectors). For our purposes here, we include in the list all equivalent isotropy subgroups, including each of the domains of each isotropy subgroup. We then follow the same algorithm as in §4 but with the following modifications.

In Step 1, we select one or more independent $\mathbf{k}$ vectors for each incommensurate IR.

We split Step 3 into two steps. In Step 3*a*, we consider the commensurate isotropy subgroups G_C, one at a time. Let H_C be the point group of subgroup G_C.

In Step 3*b*, we identify subgroups H_S of H_A that are also subgroups of H_C (*i.e.* $H_S \subset H_C \cap H_A$) and consider these one at a time. We consider only subgroups for which the commensurate part of the resulting OPD is equal to that of the selected G_C. See the example below for clarification.

In Step 5, we note that the three-dimensional lattice of G_S is the same as that of G_C; so we form G_S by attaching to each point operator in H_S the three-dimensional fractional for that operator in G_C.

In Step 6, we require that for each incommensurate IR, there exists a nonzero OPD which satisfies equation (9) simultaneously for every operator in G_S.

5.3. Example 3

As an example, we extend example 2*a* by superposing a commensurate IR Γ_4^- (GM4−) associated with $\mathbf{k} = 0$ which itself yields four commensurate isotropy subgroups: G_C = *R*3*m*, *Amm*2, *Cm* and *P*1. The inequivalent sets of active incommensurate **k** vectors for $d^{(1)} = 2$ are the same as in equation (11), where we will again consider in detail only set (2). As before, the point group H_A is $\overline{3}m$ with the threefold axis along the cubic $[\overline{1}11]$ direction.

For $G_C = R3m$ (threefold axis along the cubic $[1\overline{1}\overline{1}]$ direction), the commensurate part of the OPD is $(a, \overline{a}, \overline{a})$. One of the subgroups of H_A $(\overline{3}m)$ is equal to H_C $(3m)$. Choosing $H_S = 3m$, we obtain the isotropy subgroup 160.2.77.3 $R3m(\alpha, \alpha, 0)00(-2\alpha, \alpha, 0)00$ with OPD $(a, \overline{a}, \overline{a}|b, 0; 0, 0; b, 0; 0, 0; 0, 0; b, 0)$. Here we use the vertical bar | to separate the parts of the combined OPD belonging to IRs GM4− and SM1.

One other subgroup of $H_C \cap H_A$ generates an additional inequivalent isotropy subgroup in which the commensurate part of the OPD is equal to $(a, \overline{a}, \overline{a})$. For $H_S = 3$, we obtain 146.2.73.1 $R3(\alpha, \beta, 0)0(-\alpha-\beta, \alpha, 0)0$ with OPD $(a, \overline{a}, \overline{a}|b, c; 0, 0; c, b; 0, 0; 0, 0; b, \overline{c})$.

For $G_C = Amm2$, the commensurate part of the OPD is $(0, a, \overline{a})$, The largest point group which is a subgroup of both $H_C = mm2$ and $H_A = \overline{3}m$ is $H_S = 2$ with the twofold axis along the cubic [100] direction. Using $H_S = 2$, we obtain the isotropy subgroup 5.2.17.9 $B2(\alpha, \beta, \gamma)0(\overline{\alpha}, \overline{\beta}, \gamma)0$ with OPD $(0, a, \overline{a}|b, c; 0, 0; b, \overline{c}; 0, 0; 0, 0; 0, 0)$.

Note that $H_S = 2$ is also a subgroup of 3*m*, but we did not include it when considering $G_C = R3m$ since the commensurate part of the OPD for that case is not $(a, \overline{a}, \overline{a})$ but $(0, a, \overline{a})$. When considering subgroups of $H_C \cap H_A$, we consider only those for which the commensurate part of the OPD is equal to that of G_C.

There are two additional isotropy subgroups, one for $G_C = Cm$ and one for $G_C = P1$, which we will not analyze in detail.

6. Conclusion

Since the 1960s, a wealth of new crystallographic and group-theoretical infrastructure had been developed for exploring symmetry-lowering distortions of crystalline materials. Whereas these capabilities were previously restricted primarily to distortions involving only commensurate order parameters, we now demonstrate their extension to the general case of incommensurate order parameters. Here, we present a general algorithm to generate isotropy subgroups involving arbitrary superpositions of multi-**k** order parameters from incommensurate and commensurate **k** vectors, resulting in modulated structures possessing $(3 + d)$D superspace-group symmetry. Several examples are described in detail.

We believe the algorithm to be sufficiently general to apply to any incommensurate crystal modulation, regardless of the dimension of the resulting superspace-group symmetry. The scope of potential applications includes theoretical, computational and experimental investigations of any material classes where incommensurate modulations are possible. Incommensurate structure models generated using this algorithm can be completely parameterized in terms of order parameters of irreducible representations of the parent symmetry group, while also being fully constrained by the relevant $(3 + d)$D superspace group. We note that the *ISODISTORT* and *ISOSUBGROUP* programs of the internet-based *ISOTROPY* software suite have now been extended to make use of this algorithm for cases with up to $d = 3$ superspace dimensions.

APPENDIX A
Equivalent isotropy subgroups

Two isotropy subgroups G_{1S} and G_{2S} are equivalent if there exists an operator g in the parent G such that

$$g(G_{1S})g^{-1} = G_{2S}. \tag{16}$$

Since they are isotropy subgroups, there are corresponding OPDs $\boldsymbol{\eta}_1$ and $\boldsymbol{\eta}_2$ such that

$$D(g_1)\boldsymbol{\eta}_1 = \boldsymbol{\eta}_1 \quad \text{and} \quad D(g_2)\boldsymbol{\eta}_2 = \boldsymbol{\eta}_2 \tag{17}$$

for any $g_1 \in G_{1S}$ and any $g_2 \in G_{2S}$. Combining equations (16) and (17), we obtain $\boldsymbol{\eta}_2 = D(g_1)\boldsymbol{\eta}_1$. For our purposes, this provides a more useful criterion for equivalence: two OPDs $\boldsymbol{\eta}_1$ and $\boldsymbol{\eta}_2$ generate equivalent isotropy subgroups if there exists an operator g in G such that

$$\boldsymbol{\eta}_2 = D(g)\boldsymbol{\eta}_1. \tag{18}$$

We say that these two OPDs are equivalent.

We can also use equation (18) to generate all equivalent OPDs for an isotropy subgroup. Whenever we find a new isotropy subgroup not equivalent to any previously found isotropy subgroup, we save not only its OPD but all equivalent OPDs as well. Then, whenever we generate an isotropy subgroup, we can compare its OPD with those on the saved list and quickly find out whether or not it is a new inequivalent subgroup.

The chain rule (Birman, 1978; Jarić, 1981, 1982) states that if two isotropy subgroups have a group–subgroup relation and if they both have the same subduction frequency, we do not keep the smaller of the two. This rule can be restated as follows. If two isotropy subgroups have the same OPD, we keep the one with the larger index with respect to the parent. So, if we generate isotropy subgroups with a decreasing number of point operators, we simply discard any new isotropy subgroup with an OPD equivalent to that of a previously found isotropy subgroup.

For commensurate IRs, generating a complete list of equivalent OPDs is straightforward. We first expand the parent group G in terms of cosets of the subgroup G_S:

$$G = \sum_i g_i G_S, \tag{19}$$

where $\{g_i\}$ are the coset representatives. We put each coset representative, one at a time, into equation (18).

Because the resulting list may contain some OPDs which are equal to one another, we eliminate the duplicates. Though the symbolic forms of two OPDs may appear to be different, we say that they are equal if their variables can take on nonzero values that make the OPDs numerically equal.

The general form of an OPD is given by

$$\eta_j = \sum_{i=1}^{n_s} a_i \eta_{ij}^{(\mathrm{A})}, \tag{20}$$

where a_i are arbitrary parameters and n_s is called the subduction frequency (number of independent parameters in the OPD). To make equality immediately apparent, we bring each OPD into a 'standard' form by bringing the matrix $\eta^{(\mathrm{A})}$ into row echelon form using linear row operations. In this form, the first nonzero element of each row occurs in a column where it is the only nonzero element. We also normalize each row so that the first nonzero element is equal to 1.

As an example, consider the OPD

$$\boldsymbol{\eta} = (a_1 + a_2, a_1, a_2) = a_1(1, 1, 0) + a_2(1, 0, 1). \tag{21}$$

Bringing the general-form matrix

$$\eta^{(\mathrm{A})} = \begin{pmatrix} 1 & 1 & 0 \\ 1 & 0 & 1 \end{pmatrix} \tag{22}$$

into row echelon form, we obtain

$$\eta^{(\mathrm{A})} = \begin{pmatrix} 1 & 0 & 1 \\ 0 & 1 & \overline{1} \end{pmatrix} \tag{23}$$

and the standard form,

$$\boldsymbol{\eta}' = a_1(1, 0, 1) + a_2(0, 1, \overline{1}) = (a_1', a_2', a_1' - a_2'). \tag{24}$$

Note that for $a_1' = a_1 + a_2$ and $a_2' = a_1$, these two OPDs $\boldsymbol{\eta}$ and $\boldsymbol{\eta}'$ are exactly equal.

For incommensurate IRs, the generation of all equivalent OPDs becomes impossible in practice. The parent symmetry group G is a space group extended to superspace and contains one or more phase shifts with undetermined and continuously variable values between 0 and 1. The isotropy subgroup G_S is an SSG with discrete phase shifts that are determined. Thus, there are an infinite number of cosets in equation (7) and accordingly an infinite number of distinct equivalent OPDs.

IR matrices of operators g in G can have a particularly simple form (Stokes *et al.*, 2013):

$$D(g) = Q(\boldsymbol{\delta})P(R), \tag{25}$$

where R is the three-dimensional point operator in external space and $\boldsymbol{\delta}$ is the $d^{(\mathrm{A})}$-dimensional vector containing the phase shift for each incommensurate modulation. Note that $D(g)$ does not depend on the three-dimensional translations in external space since the IR maps all lattice translations onto the unit matrix.

Let H be the point group of G and let H_S be the point group of G_S. We expand H in terms of cosets of H_S:

$$H = \sum_i R_i H_S, \tag{26}$$

where the coset representatives R_i are now point operators. Similar to equation (18), we can form a list of equivalent OPDs with respect to point groups using

$$\boldsymbol{\eta}_2 = P(R_i)\boldsymbol{\eta}_1 \tag{27}$$

for each coset representative R_i.

The remaining infinite number of equivalent OPDs could in theory by generated by

$$\boldsymbol{\eta}_3 = Q(\boldsymbol{\delta})\boldsymbol{\eta}_2 \tag{28}$$

using all possible values of $\boldsymbol{\delta}$. (In G, each component of $\boldsymbol{\delta}$ can take on any value between 0 and 1.) So, instead of using equation (28) to generate a list of equivalent OPDs, we use it to *compare* two OPDs. If some choice of $\boldsymbol{\delta}$ can satisfy equation (28), then the two OPDs are equivalent.

Whenever we find a new isotropy subgroup not equivalent to any previously found isotropy subgroup, we save not only its OPD but also all equivalent OPDs with respect to point groups. Then, whenever we generate a new isotropy subgroup, we compare its OPD with those on the saved list using equation (28). If a solution for $\boldsymbol{\delta}$ is found, then the two OPDs are equivalent and we do not keep the new result.

Solving equation (28) for $\boldsymbol{\delta}$ is not straightforward since both $\boldsymbol{\eta}_2$ and $\boldsymbol{\eta}_3$ contain parameters with unknown values, and since $Q(\boldsymbol{\delta})$ is not linear in $\boldsymbol{\delta}$. However, $Q(\boldsymbol{\delta})$ *is* linear in $\cos 2\pi\delta_i$ and $\sin 2\pi\delta_i$. If we introduce an arbitrary wave amplitude A_i, then $\lambda_i^{\mathrm{c}} = A_i \cos 2\pi\delta_i$ and $\lambda_i^{\mathrm{s}} = A_i \sin 2\pi\delta_i$ can be treated like two independent variables. The amplitudes A_i will be normalized to 1 only after a solution has been found. Lastly, we choose random values for the parameters in $\boldsymbol{\eta}_2$. Now equation (28) is a set of equations linear in the variables λ_i^{c} and λ_i^{s}, and the parameters in $\boldsymbol{\eta}_1$, and is easily solved. We assume that if there is a solution for one set of random values for the parameters in $\boldsymbol{\eta}_2$, then there will be a solution for any set of random values. Just to be certain that we did not accidentally choose some special values, we repeat for a second set of random values.

As an example, let us consider a simple case with one active **k** vector and two-dimensional IR matrices. We choose the two OPDs we wish to compare to be $(a, 0)$ and (b, b). The form for $Q(\boldsymbol{\delta})$ is simply

$$Q(\delta) = \begin{pmatrix} \cos 2\pi\delta & \sin 2\pi\delta \\ -\sin 2\pi\delta & \cos 2\pi\delta \end{pmatrix}. \tag{29}$$

Assigning a random number $b = 0.37$, equation (28) becomes

$$\begin{pmatrix} \lambda^{\mathrm{c}} & \lambda^{\mathrm{s}} \\ -\lambda^{\mathrm{s}} & \lambda^{\mathrm{c}} \end{pmatrix} \begin{pmatrix} 0.37 \\ 0.37 \end{pmatrix} = \begin{pmatrix} a \\ 0 \end{pmatrix}. \tag{30}$$

Note that there are two equations and three variables, so we are free to assign any value to one of the variables. We choose $\lambda^{\mathrm{s}} = 1$. With this choice, we easily obtain $\lambda^{\mathrm{c}} = 1$ and $a = 0.74$,

from which we calculate $A = 1.414$ and $\delta = 0.125$. The existence of a solution indicates that these two OPDs are indeed equivalent.

APPENDIX *B*
Isotropy subgroups of commensurate IRs

An algorithm for generating isotropy subgroups was proposed by Hatch (1984) and then implemented on computer (Stokes & Hatch, 1984, 1988; Hatch & Stokes, 1984, 1986). Although this algorithm has been in use for many years as part of the *ISOTROPY* software suite, the details of its implementation have never been published. We add these details here, in some sense making this paper complete, so that commensurate and incommensurate cases, and even their superpositions, can be treated within a common framework.

B1. Lattice of the subgroup

The algorithm for generating isotropy subgroups of some parent space group G_0 for commensurate IRs is very similar to that for incommensurate IRs described in the body of this paper. However, there are some important differences. For one, the translation group T_S containing all of the translations $\mathbf{t}_s$ of the subgroup G_S may be a proper subgroup of the translation group T_0 containing all of the translations of the parent group G_0. In other words, the lattice of the subgroup may be smaller than the lattice of the parent which requires a different strategy for identifying the set of active $\mathbf{k}$ vectors $\{\mathbf{k}^{(A)}\}$ associated with a given isotropy subgroup G_S.

Consider the requirement that there exists a nonzero OPD $\boldsymbol{\eta}$ which satisfies

$$D(g_S)\boldsymbol{\eta} = \boldsymbol{\eta} \tag{31}$$

for every operator g_S in the subgroup G_S, including the translation operators $\mathbf{t}_S$ in T_S. Our standard form of the IR matrix $D(\mathbf{t}_S)$ (Stokes *et al.*, 2013) for a translation operator $\mathbf{t}_S$ is block diagonal with $d^{(F)}$ blocks, such that the ith block of dimension b is given by

$$U_i(\mathbf{t}_S) = \begin{pmatrix} \mathbf{1}_{b/2}\cos(2\pi\mathbf{k}_i\cdot\mathbf{t}_S) & \mathbf{1}_{b/2}\sin(2\pi\mathbf{k}_i\cdot\mathbf{t}_S) \\ -\mathbf{1}_{b/2}\sin(2\pi\mathbf{k}_i\cdot\mathbf{t}_S) & \mathbf{1}_{b/2}\cos(2\pi\mathbf{k}_i\cdot\mathbf{t}_S) \end{pmatrix}, \tag{32}$$

where $\mathbf{k}_i$ is the ith vector of $\{\mathbf{k}^{(F)}\}$ and $\mathbf{1}_{b/2}$ is a $b/2$-dimensional unit matrix. Recall that the OPD can be written as

$$\boldsymbol{\eta} = (\boldsymbol{\eta}_1, \boldsymbol{\eta}_2, \ldots, \boldsymbol{\eta}_{d^{(F)}}), \tag{33}$$

where $\boldsymbol{\eta}_i$ is a b-dimensional vector corresponding to the ith vector of $\{\mathbf{k}^{(F)}\}$ and therefore to the block $U_i(\mathbf{t}_S)$, so that equation (31) can be written separately for each block,

$$U_i(\mathbf{t}_S)\boldsymbol{\eta}_i = \boldsymbol{\eta}_i. \tag{34}$$

Since equation (34) must be satisfied simultaneously for every lattice translation $\mathbf{t}_S$ in T_S, it is clear that there is no nonzero solution for $\boldsymbol{\eta}_i$ unless

$$\mathbf{k}_i\cdot\mathbf{t}_S = \text{integer} \tag{35}$$

for every lattice translation $\mathbf{t}_S$ in T_S. Thus, the requirement that there exists a nonzero OPD $\boldsymbol{\eta}$ which satisfies equation (31) for every lattice translation $g_S = \mathbf{t}_S$ in T_S cannot be met unless equation (35) is satisfied for at least one of the $\mathbf{k}$ vectors in $\{\mathbf{k}^{(F)}\}$. The $\mathbf{k}$ vectors in $\{\mathbf{k}^{(F)}\}$ for which equation (35) is satisfied, if any, are called the active $\mathbf{k}$ vectors and form the set $\{\mathbf{k}^{(A)}\}$. Note that any OPD $\boldsymbol{\eta}$ that satisfies equation (31) for each of the three basis translations of T_S will also satisfy it for every translation $\mathbf{t}_S$ of T_S.

Now we are ready to describe the steps in the algorithm for generating isotropy subgroups.

B2. Active k vectors

Step 1. Find every possible set $\{\mathbf{k}^{(A)}\}$ of nonequivalent active $\mathbf{k}$ vectors. To accomplish this, we identify every nonequivalent subset of $\{\mathbf{k}^{(F)}\}$, and for each subset we find the subgroup T_S comprised of the lattice vectors $\mathbf{t}_S$ which satisfy equation (35) for every $\mathbf{k}$ vector in the subset. We then determine if any of the other $\mathbf{k}$ vectors in $\{\mathbf{k}^{(F)}\}$ also satisfy equation (35) for every lattice vector in T_S, and, if so, we discard the subset under consideration and move on to the next one. The subsets that survive are 'complete' sets $\{\mathbf{k}^{(A)}\}$ of active $\mathbf{k}$ vectors.

B3. New fractional translations in the subgroup

Step 2. For a given set $\{\mathbf{k}^{(A)}\}$ and associated translation group T_S, we decompose the translation group T_0 of the lattice translations in the parent G_0 into cosets with respect to T_S:

$$T_0 = \sum_i \mathbf{t}_i T_S, \tag{36}$$

where $\mathbf{t}_i$ are the coset representatives (reps), which (except for $\mathbf{t}_1 = 0$) become new fractional translations in the subgroup.

B4. Point group H_A of the subgroup lattice

Step 3. Given a set $\{\mathbf{k}^{(A)}\}$ and associated translation group T_S, we find the point operators of G_0 which leave the lattice invariant, *i.e.* which take each basis translation of T_S into a linear combination of the three basis translations. These point operators form a point group H_A.

B5. Subgroups H_S of H_A

Step 4. For a given point group H_A, we find all of the point groups H_S which are subgroups of H_A, and consider them one at a time, beginning with the largest, which is H_A itself, and ending with the smallest, which contains only the identity point operator.

B6. Subgroups G_S of G_0

Step 5. From a given point group H_S, we next attempt to form subgroups G_S of G_0. For each generator h in H_S, we take the corresponding operator g_h in G_0 and add to its translational part one of the coset reps $\mathbf{t}_i$ in equation (36). These become the generators of G_S. We obtain the other operators in G_S using group multiplication. If the resulting set of operators obeys a valid multiplication table, it is saved for further consideration as an isotropy subgroup. All possible ways of combining the coset reps $\mathbf{t}_i$ with the generators of H_S must be explored.

B7. Order parameter direction

Step 6. We must now test G_S to determine if it is an isotropy subgroup for the selected IR, by requiring that there exists a nonzero OPD $\boldsymbol{\eta}$ which satisfies equation (31) simultaneously for every operator g_S in G_S, not just for the lattice translations.

B8. Equivalent OPDs

Step 7. For each isotropy subgroup G_S, we require that it not be equivalent to any previously found isotropy subgroup, which can be tested by verifying that their OPDs are not equivalent (see Appendix *A*).

B9. Find G_S in standard tables

Step 8. For a given inequivalent isotropy subgroup G_S, we identify it as one of the 230 crystallographic space groups and determine the transformation (lattice basis vectors and origin) that takes the operators of the subgroup from the parent setting to the standard space-group setting of the subgroup (Hatch & Stokes, 1985).

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