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Order parameters for phase transitions to structures with one-dimensional incommensurate modulations

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Phase transitions that result in incommensurate structural modulations are widely observed in crystalline solids and are relevant to a broad range of physical phenomena in magnetic, electronic, optical and structural materials. While the (3+1)-dimensional superspace-group symmetries associated with one-dimensional modulations have been tabulated, the order parameters that produce these modulations have not been explored in detail. Here, using group-theoretical methods, we present a unique and exhaustive enumeration of the isotropy subgroups (and their corresponding order-parameter directions) belonging to irreducible representations of the (3+1)-dimensional superspace extensions of the 230 crystallographic space groups at all incommensurate k points. The vast majority of experimentally observed incommensurately modulated structures have order parameters belonging to one of these subgroups.

1. Introduction

Over the years, the study of structural phase transitions in crystalline solids has greatly benefited from group-theoretical methods (Bradley & Cracknell, 1972; Birman, 1978; Toledano & Toledano, 1987; Kovalev, 1993; Janssen *et al.*, 2004; Howard & Stokes, 2005). In the largest class of these transitions, we observe the onset of some distortion which lowers the symmetry of a parent phase. In this paper, we use the word 'distortion' in a general sense to represent any physical order parameter that lowers the symmetry of the parent phase. Distortions can involve not only atomic displacements but also site occupation, electron density and magnetic spin, for example. When the symmetries of the parent and distorted phases have a group–subgroup relationship, the distortion can be classified as belonging to one or more irreducible representations (IRs) of the space-group symmetry of the parent phase.

This 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. The symmetries of these distorted structures are called isotropy subgroups. For a given set of IRs, the complete list of possible isotropy subgroups is finite.

Order parameters are vectors in representation space and determine the distortions that arise in a phase transition. The direction of the order parameter determines the symmetry of the distortion and thus determines the isotropy subgroup symmetry. For multidimensional IRs, there is a many-to-one correspondence between directions of order parameters and isotropy subgroup symmetries. For each isotropy subgroup symmetry, there is a continuous range of directions of the order parameter that produce distortions with that symmetry.

About 20 years ago, Stokes & Hatch (1988) implemented an algorithm for generating isotropy subgroups and their corresponding order-parameter directions 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. Subsequently, the *ISOTROPY* computer program was created (available over the internet, see Stokes & Hatch, 1998). *ISOTROPY* can find the isotropy subgroups for IRs associated with non-special **k** vectors having rational components (*i.e.* general commensurate **k** vectors) and for coupled IRs that appear simultaneously in a transition. All of these isotropy subgroups describe transitions to structures that are commensurate with the parent phase.

Perez-Mato *et al.* (1984*a*,*b*) and Janssen & Janner (1984) demonstrated how to apply group-theoretical methods to phase transitions which produce modulations that are incommensurate with the periodicity of the parent phase. Incommensurate modulations belong to IRs associated with **k** vectors having irrational components. We have extended the method of Perez-Mato *et al.* and have developed a general algorithm for generating the isotropy subgroups, and their corresponding order-parameter directions, for such IRs. The method described here is limited to structures with one-dimensional incommensurate modulations but may be easily extended to the higher-dimensional cases.

We have implemented this algorithm on computer and have tabulated 7799 isotropy subgroups and their corresponding order-parameter directions for the 5508 IRs of the (3+1)-dimensional superspace extensions of the 230 crystallographic space groups associated with irrational **k** vectors. This table is

available on the internet (Stokes *et al.*, 2007) and will hereafter be referred to by the name ISO(3+1)D. The scope of this table is limited to one-dimensional incommensurate modulations arising from single uncoupled IRs.

While the present discussion aims to describe and illustrate the group-theoretical methods used to generate the ISO(3+1)D tables, the tables themselves are still useful to those who are not interested in the underlying methods, as illustrated by the examples in §4.

2. Background

We first review some principles of IRs, order parameters, isotropy subgroups and superspace groups essential for understanding our algorithm.

2.1. IRs of three-dimensional space groups

IRs of a three-dimensional crystallographic space group G are induced from IRs of G_k, the little group of **k**. This is what is meant by the IR being associated with a **k** vector. G_k is a subgroup of G that consists of all symmetry operators of G which contain point operators that take **k** into itself (modulo a reciprocal-lattice vector **K**). Following the treatment of Bradley & Cracknell (1972), we factorize G into left cosets with respect to G_k so that

$$G = \sum_i g_i G_k, \tag{1}$$

where g_i are coset representatives (reps). We choose g_1 to be the identity operator so that the first coset is simply G_k itself.

Let $D(g)$ denote the N-dimensional matrix onto which an IR Γ maps symmetry operators g of the parent space group G and let $D_k(g)$ denote the N_k-dimensional matrix onto which an IR Γ_k maps symmetry operators of G_k, the little group of **k**. Then the matrices $D(g)$ are induced from the matrices $D_k(g)$:

$$D(g)_{ij} = M_{ij} D_k(g_i g g_j^{-1}), \tag{2}$$

where $M_{ij} = 1$ if $g_i g g_j^{-1}$ is in G_k and zero otherwise. The subscripts i, j refer to blocks of N_k-dimensional matrices within $D(g)$. Each allowed IR of G_k induces an IR of G.

Since we are considering physical distortions, we need real-valued matrices. Generally, the matrices obtained from equation (2) are complex. In the case of type-1 IRs, the matrices $D(g)$ are brought to a real form with a similarity transformation, $D_R(g) = SD(g)S^{-1}$. In the case of type-2 and type-3 IRs, a physically irreducible representation is formed from a direct sum of $D(g)$ and its complex conjugate which is then brought to real form with a similarity transformation.

2.2. Isotropy subgroups and order parameters

An IR induces distortions which can be decomposed into sets $\{\phi_i\}$ which transform like basis functions of that IR, *i.e.* in representation space, a space-group operator g acting on one element in the set results in a linear combination of elements in the set given by

$$g\phi_i = \sum_{j=1}^{N} \phi_j D(g)_{ji}. \tag{3}$$

When we say that the distortion arising in a phase transition 'belongs' to an IR, we mean that the distortion is some linear combination of distortions ϕ_i that transform like basis functions of that IR:

$$\phi = \sum_{i=1}^{N} \eta_i \phi_i. \tag{4}$$

The coefficients η_i are components of an N-dimensional vector $\boldsymbol{\eta}$ called the order-parameter direction.

When a space-group operator g acts on the total distortion ϕ, we obtain

$$g\phi = \sum_{i=1}^{N} \phi_i \sum_{j=1}^{N} D(g)_{ij} \eta_j. \tag{5}$$

Comparing equations (4) and (5), we see that the operator g will leave the distortion invariant ($g\phi = \phi$) if

$$\eta_i = \sum_{j=1}^{N} D(g)_{ij} \eta_j. \tag{6}$$

The symmetry group of the distortion will contain all operators g which satisfy this equation. This collection of operators g is the isotropy subgroup. If we diagonalize $D(g)$, we can see from equation (6) that an isotropy subgroup can only contain operators which map onto matrices $D(g)$ with at least one eigenvalue equal to 1.

Isotropy subgroups can be generated without reference to order parameters by testing subgroups G' of G with the following two rules (Birman, 1978; Jaric, 1981, 1982).

(I) Subduction rule. The subduction frequency must be non-zero:

$$i(G') = \frac{1}{|G'|} \sum_{g \in G'} \chi(g) \neq 0, \tag{7}$$

where $\chi(g)$ is the character of $D(g)$. In practice, the sum is taken over operators $g \in G'$ that are mapped onto distinct matrices $D(g)$, and $|G'|$ is the number of those distinct matrices.

(II) Chain rule. We require that $i(G') > i(G'')$ for all isotropy subgroups G'' which are supergroups of G'.

We first find isotropy subgroups using the subduction and chain rules, and then obtain the corresponding order-parameter directions by solving sets of simultaneous equations for the components of $\boldsymbol{\eta}$. We obtain N equations like equation (6) for each generator of the isotropy subgroup. The solution to these equations yields relationships between the components of $\boldsymbol{\eta}$ and thereby restrict the order-parameter direction to a specific invariant subspace of representation space. Any physical distortion ϕ from equation (4) with a direction of $\boldsymbol{\eta}$ thus restricted will possess the symmetry of the corresponding isotropy subgroup.

2.3. Superspace groups

In a phase transition to an incommensurately modulated structure, three-dimensional translational symmetry is lost. A translation **n** of the lattice in the parent phase causes a phase shift $\mathbf{k}\cdot\mathbf{n}$ in the modulation. If the phase shift is irrational, the structure has no translational symmetry in the direction of **n**, so that its symmetry cannot be described by a crystallographic space group. In such a case, the symmetry of the structure is best described (in the case of one-dimensional modulations) by a (3+1)-dimensional superspace group (Janssen *et al.*, 2004), where translations along the axis in the fourth dimension (called the t axis) correspond to phase shifts of the modulation. Because a translation $-\mathbf{k}\cdot\mathbf{n}$ along the t axis corresponds to a phase shift which undoes the phase shift caused by the translation **n**, the combined (3+1)-dimensional translation $(\mathbf{n}, -\mathbf{k}\cdot\mathbf{n})$ in superspace is a symmetry operator of the modulated structure. Thus, in superspace, modulated structures still have translational symmetry.

Following Janssen *et al.* (2004), we denote an operator in superspace by $\tilde{g} = \{(R, \varepsilon)|(\mathbf{v}, \Delta)\}$, where the three-dimensional part $\{R|\mathbf{v}\}$ is a point operation R followed by a translation **v**, and the part in the fourth dimension $\{\varepsilon|\Delta\}$ is a point operation ε (which operates on the t coordinate) followed by a translation Δ along the t axis. [Here, g refers to operators in three-dimensional space, and $\tilde{g}$ refers to operators in (3+1)-dimensional space.] It is convenient to write the full (3+1)-dimensional operator as an affine transformation matrix $A(\tilde{g})$ which operates on a position vector **x**,

$$A(\tilde{g})\mathbf{x} = \begin{pmatrix} R_{11} & R_{12} & R_{13} & 0 & v_1 \\ R_{21} & R_{22} & R_{23} & 0 & v_3 \\ R_{31} & R_{32} & R_{33} & 0 & v_3 \\ 0 & 0 & 0 & \varepsilon & \Delta \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x \\ y \\ z \\ t \\ 1 \end{pmatrix}. \tag{8}$$

We define basis vectors of the superspace lattice,

$$\begin{aligned} a_1 &= (\mathbf{a}, -\mathbf{k}\cdot\mathbf{a}), \\ a_2 &= (\mathbf{b}, -\mathbf{k}\cdot\mathbf{b}), \\ a_3 &= (\mathbf{c}, -\mathbf{k}\cdot\mathbf{c}), \\ a_4 &= (0, 1), \end{aligned} \tag{9}$$

and a position in superspace as $x_1a_1 + x_2a_2 + x_3a_3 + x_4a_4$. By changing from x, y, z, t to x_1, x_2, x_3, x_4 coordinates, equation (8) becomes

$$A(\tilde{g})\mathbf{x} = \begin{pmatrix} R_{11} & R_{12} & R_{13} & 0 & v_1 \\ R_{21} & R_{22} & R_{23} & 0 & v_2 \\ R_{31} & R_{32} & R_{33} & 0 & v_3 \\ M_1 & M_2 & M_3 & \varepsilon & \delta \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \\ 1 \end{pmatrix}, \tag{10}$$

where

$$M_i = -\varepsilon k_i + \sum_j k_j R_{ji} \tag{11}$$

and

$$\delta = \Delta + \sum_i k_i v_i. \tag{12}$$

The requirement that point operations on lattice vectors must result in a lattice vector demands that the values of M_i in equation (11) be integers. Thus, the superspace group may only contain point operators that take **k** into either (i) **k** itself (modulo a reciprocal-lattice vector **K**) in which case $\varepsilon = 1$, or (ii) $-\mathbf{k}$ (modulo **K**) in which case $\varepsilon = -1$.

Two superspace groups $\tilde{G}_1$ and $\tilde{G}_2$ are equivalent if there exists a one-to-one correspondence $\tilde{g}_{2i} \leftrightarrow \tilde{g}_{1i}$ between operators in $\tilde{G}_1$ and $\tilde{G}_2$ such that

$$A_2(\tilde{g}_{2i}) = S^{-1}A_1(\tilde{g}_{1i})S, \tag{13}$$

where A_1 and A_2 are affine transformation matrices of the form in equation (10) and S is an affine similarity transformation matrix,

$$S = \begin{pmatrix} S_{R,11} & S_{R,12} & S_{R,13} & 0 & S_{v1} \\ S_{R,21} & S_{R,22} & S_{R,23} & 0 & S_{v2} \\ S_{R,31} & S_{R,32} & S_{R,33} & 0 & S_{v3} \\ S_{M1} & S_{M2} & S_{M3} & S_\varepsilon & S_\delta \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \tag{14}$$

such that S_R, S_M and S_ε contain only integer elements. The basis vectors a_{2i} of the lattice in G_2 are given in terms of the basis vectors a_{1i} of the lattice in G_1 by

$$a_{2i} = \sum_{j=1}^{4} a_{1j}S_{ji}, \tag{15}$$

and the position of the origin τ of G_2 with respect to G_1 is given by

$$\tau = \sum_{j=1}^{4} a_{1j}S_{j5}. \tag{16}$$

3. Incommensurate modulations

We next consider the ways we deal with IRs and isotropy subgroups when the **k** vector is irrational and the isotropy subgroup must have superspace-group symmetry. In order for superspace groups to be subgroups of the parent group, we must first extend the parent group to include translations along the t axis in superspace. Since the modulation amplitude is zero in the parent phase, such translations along the t axis are symmetry operators of the parent phase.

Therefore, we define

$$\tilde{G} = G \otimes E_1(\Delta), \tag{17}$$

where $E_1(\Delta)$ is the one-dimensional Euclidean group of all translations Δ along the t axis. [As in the previous section, G refers to a group of operators in three-dimensional space, and $\tilde{G}$ refers to a group of operators in (3+1)-dimensional superspace.]

3.1. IRs and isotropy subgroups

For an incommensurate modulation in the direction of **k**, the resulting superspace symmetry can only contain point operators that take **k** into $\pm\mathbf{k}$. In order to generate the

isotropy subgroups of $\tilde{G}$ that lead to incommensurate modulations, we find it useful to first identify a suitable subset of $\tilde{G}$ that contains only these operators. We will first consider the analogous subset of the three-dimensional space group G, and then extend the results to $\tilde{G}$.

In equation (1), the first coset is G_k which contains all of the operators in G that take $\mathbf{k}$ into $\mathbf{k}$ itself. Suppose that one of the coset reps g_i contains a point operator that takes $\mathbf{k}$ into $-\mathbf{k}$. We denote this coset rep by $g_{\bar{k}}$. (When no such coset rep exists, the following discussion is greatly simplified.) The coset $g_{\bar{k}}G_k$ then contains all of the operators in G which take $\mathbf{k}$ into $-\mathbf{k}$. We construct a group $G_{k\bar{k}}$ which contains only the operators in these two cosets:

$$G_{k\bar{k}} = G_k + g_{\bar{k}}G. \tag{18}$$

IR matrices $D_{k\bar{k}}(g)$ for operators g in $G_{k\bar{k}}$ are easily obtained from equation (2) by truncating the matrices $D(g)$ to include only the indices i, j for the cosets G_k and $g_{\bar{k}}G_k$. The resulting matrices $D_{k\bar{k}}(g)$ are $2N_k$-dimensional.

If g is in G_k, the matrix $D_{k\bar{k}}(g)$ has the form

$$D_{k\bar{k}}(g) = \begin{pmatrix} D_{11} & \mathbf{0} \\ \mathbf{0} & D_{22} \end{pmatrix}, \tag{19}$$

where D_{11} and D_{22} are N_k-dimensional matrices and $\mathbf{0}$ is the N_k-dimensional zero matrix. In particular, if g is a lattice translation,

$$D_{k\bar{k}}(\{E|\mathbf{n}\}) = \begin{pmatrix} \mathbf{1}\exp(i2\pi\mathbf{k}\cdot\mathbf{n}) & \mathbf{0} \\ \mathbf{0} & \mathbf{1}\exp(-i2\pi\mathbf{k}\cdot\mathbf{n}) \end{pmatrix}, \tag{20}$$

where $\mathbf{1}$ is the N_k-dimensional identity matrix. If g is in $g_{\bar{k}}G_k$, the matrix $D_{k\bar{k}}(g)$ has the form

$$D_{k\bar{k}}(\{R|\mathbf{v}\}) = \begin{pmatrix} \mathbf{0} & D_{12} \\ D_{21} & \mathbf{0} \end{pmatrix}. \tag{21}$$

We now extend the above treatment to include translations along the t axis so that equation (18) becomes

$$\tilde{G}_{k\bar{k}} = \tilde{G}_k + \tilde{g}_{\bar{k}}\tilde{G}_k. \tag{22}$$

The IR matrices $\tilde{D}_{k\bar{k}}(\tilde{g})$ for $\tilde{g}$ in $\tilde{G}_{k\bar{k}}$ are simply obtained from

$$\tilde{D}_{k\bar{k}}[\{(R, \varepsilon)|(\mathbf{v}, \Delta)\}] = \tilde{D}_{k\bar{k}}[\{(E, 1)|(0, \Delta)\}]D_{k\bar{k}}(\{R|\mathbf{v}\}), \tag{23}$$

where, from Perez-Mato *et al.* (1984*a*,*b*),

$$\tilde{D}_{k\bar{k}}[\{(E, 1)|(0, \Delta)\}] = \begin{pmatrix} \mathbf{1}\exp(i2\pi\Delta) & \mathbf{0} \\ \mathbf{0} & \mathbf{1}\exp(-i2\pi\Delta) \end{pmatrix}. \tag{24}$$

From these equations, we obtain

$$\tilde{D}_{k\bar{k}}[\{(E, 1)|(\mathbf{n}, -\mathbf{k}\cdot\mathbf{n})\}] = \begin{pmatrix} \mathbf{1} & \mathbf{0} \\ \mathbf{0} & \mathbf{1} \end{pmatrix}, \tag{25}$$

as expected.

Phase transitions to structures with modulation vector $\mathbf{k}$ will have the symmetries of isotropy subgroups of $\tilde{G}_{k\bar{k}}$. Our task is to find the isotropy subgroups of $\tilde{G}_{k\bar{k}}$. Since $\tilde{G}_{k\bar{k}}$ contains an infinite number of operators, it is not practical to find the isotropy subgroups by applying the subduction and chain rules to every subgroup of $\tilde{G}_{k\bar{k}}$. We must somehow limit the number of subgroups to try.

3.2. Translations

From equation (25), we see that every IR maps the lattice translations $(\mathbf{n}, -\mathbf{k}\cdot\mathbf{n})$ onto identity matrices. Therefore, these same lattice translations appear in every isotropy subgroup. The collection of all such lattice translations forms a translation group which we denote by $\tilde{T}$.

We now determine which translations Δ along the t axis can appear in isotropy subgroups. An operator $\tilde{g}$ can be a member of an isotropy subgroup only if at least one of the eigenvalues of $\tilde{D}_{k\bar{k}}(\tilde{g})$ is equal to 1. Let us denote these operators as being 'eligible'. Consider an operator $\tilde{g} \in \tilde{G}_k$. Combining equations (19) and (24), we find that matrix $\tilde{D}_{k\bar{k}}(\tilde{g})$ has the form

$$\tilde{D}_{k\bar{k}}[\{(R, 1)|(\mathbf{v}, \Delta)\}] = \begin{pmatrix} D_{11}\exp(i2\pi\Delta) & \mathbf{0} \\ \mathbf{0} & D_{22}\exp(-i2\pi\Delta) \end{pmatrix}, \tag{26}$$

where the matrices D_{11} and D_{22} are the diagonal elements of $D_{k\bar{k}}(\{R|\mathbf{v}\})$. The eligible operators are therefore those for which $\exp(-i2\pi\Delta)$ is equal to one of the eigenvalues of D_{11} or $\exp(i2\pi\Delta)$ is equal to one of the eigenvalues of D_{22}. This limits us to a finite number of possible values for Δ.

Next consider an operator $\tilde{g} \in \tilde{g}_{\bar{k}}\tilde{G}_k$. From equations (21) and (24), we find that the matrix $\tilde{D}_{k\bar{k}}(\tilde{g})$ has the form

$$\tilde{D}_{k\bar{k}}[\{(R, -1)|(\mathbf{v}, \Delta)\}] = \begin{pmatrix} \mathbf{0} & D_{12}\exp(i2\pi\Delta) \\ D_{21}\exp(-i2\pi\Delta) & \mathbf{0} \end{pmatrix}, \tag{27}$$

which can be written in the form

$$S\begin{pmatrix} \mathbf{0} & D_{12} \\ D_{21} & \mathbf{0} \end{pmatrix}S^{-1}, \tag{28}$$

where

$$S = \begin{pmatrix} \mathbf{1}\exp(i\pi\Delta) & \mathbf{0} \\ \mathbf{0} & \mathbf{1}\exp(-i\pi\Delta) \end{pmatrix}. \tag{29}$$

Because a similarity transformation does not change the eigenvalues of a matrix, if $D_{k\bar{k}}(\{R|\mathbf{v}\})$ contains at least one eigenvalue equal to 1, then so will $\tilde{D}_{k\bar{k}}(\{(R, -1)|(\mathbf{v}, \Delta)\})$ for any value of Δ. Let $\tilde{T}_\Delta$ be the translation group containing operators $\{(E, 1)|(0, \Delta)\}$ for all possible values of Δ. Factorizing $\tilde{g}_{\bar{k}}\tilde{G}_k$ into left cosets with respect to $\tilde{T}_\Delta$, we obtain

$$\tilde{g}_{\bar{k}}\tilde{G}_k = \sum_i \tilde{g}_{\bar{k},i}\tilde{T}_\Delta, \tag{30}$$

where $\tilde{g}_{\bar{k},i}$ are coset reps. If one operator in a coset is eligible, then all operators in that coset are eligible.

3.3. The group $\tilde{H}$

We next need to restrict the search for isotropy subgroups to a manageably finite number of possibilities. We first form a set containing every eligible operator in $\tilde{G}_k$, and at least one operator from every coset in equation (30) having eligible operators, and then use group multiplication to complete a

group that we call $\tilde{H}$. Because the product of two eligible operators is not necessarily eligible, $\tilde{H}$ may contain operators which are not eligible. Note that the construction of $\tilde{H}$ is not unique. We can choose any operator from the eligible cosets in equation (30). However, we strategically choose them so as to minimize the total number of operators in $\tilde{H}$. We also find that it is always possible to choose operators $\tilde{g}$ so that none of the matrices $\tilde{D}_{k\bar{k}}(\tilde{g})$ depend on $\mathbf{k}$. Other possible choices for $\tilde{H}$ result in equivalent sets of isotropy subgroups which are related by translations of the origin along the t axis. In the ISO(3+1)D tables, we list the generators $\tilde{g}$ of $\tilde{H}$ along with the matrices $\tilde{D}_{k\bar{k}}(\tilde{g})$ for each of the 5508 IRs associated with incommensurate $\mathbf{k}$ vectors. The group of distinct matrices $\tilde{D}_{k\bar{k}}(\tilde{g})$ onto which the IR maps the operators $\tilde{g}$ of $\tilde{H}$ is called the image of $\tilde{H}$. Among these IRs, we find that only 15 inequivalent images of $\tilde{H}$ arise.

Now consider some isotropy subgroup $\tilde{G}'$ of $\tilde{G}$. The operators in $\tilde{G}'$ may be divided into two sets: (i) those contained in $\tilde{G}_k$ and (ii) those contained in $\tilde{g}_{\bar{k}}\tilde{G}_k$ (if any). By definition, every operator in $\tilde{G}'$ must be eligible. Thus, every operator in set (i) must also be contained in $\tilde{H}$, since $\tilde{H}$ contains every eligible operator in $\tilde{G}_k$. The operators in set (ii), however, may not be contained in $\tilde{H}$. Here we use the fact that a translation of the origin of $\tilde{G}'$ along the t axis changes the value of Δ for operators in $\tilde{g}_{\bar{k}}\tilde{G}_k$ but not for those in $\tilde{G}_k$. We can therefore move the origin of $\tilde{G}'$ so that at least one of the operators in set (ii) is in $\tilde{H}$. If we do this, then, by group multiplication, all of the operators in set (ii) will be in $\tilde{H}$. In summary, there exists a translation of the origin along the t axis such that every operator in $\tilde{G}'$ is in $\tilde{H}$. This means that every isotropy subgroup of $\tilde{G}_{k\bar{k}}$ is equivalent, *via* an origin shift along the t axis, to a subgroup of $\tilde{H}$.

As a final step, we factorize $\tilde{H}$ into left cosets with respect to the translation group $\tilde{T}$:

$$\tilde{H} = \sum_i \tilde{h}_i \tilde{T}. \tag{31}$$

The number of cosets is finite. Since every isotropy subgroup will contain every element in $\tilde{T}$, isotropy subgroups will contain only whole cosets from equation (31). Finding the non-equivalent isotropy subgroups of $\tilde{G}_{k\bar{k}}$ is now reduced to finding the non-equivalent sets of cosets from equation (31) which form groups satisfying the subduction and chain rules. In Appendix *A*, we give a detailed example of generating $\tilde{H}$ and its isotropy subgroups.

3.4. Violation of the chain rule

This is best illustrated by a simple example. Suppose there are two cosets in equation (31) so that

$$\tilde{H} = \tilde{h}_1\tilde{T} + \tilde{h}_2\tilde{T}, \tag{32}$$

where h_1 is the identity operator and

$$\tilde{D}_{k\bar{k}}(\tilde{h}_1) = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \tag{33}$$

$$\tilde{D}_{k\bar{k}}(\tilde{h}_2) = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{34}$$

and every operator in $\tilde{T}$ is mapped onto the identity matrix. It would appear that there are two isotropy subgroups, $\tilde{G}'_1 = \tilde{h}_1\tilde{T} + \tilde{h}_2\tilde{T} = \tilde{H}$ and $\tilde{G}'_2 = \tilde{h}_1\tilde{T} = \tilde{T}$ with subduction frequencies $i(\tilde{G}'_1) = 1$ and $i(\tilde{G}'_2) = 2$. Both the subduction and chain rules are satisfied. The order parameter for $\tilde{G}'_1$ is $\boldsymbol{\eta} = (a, 0)$, and the order parameter for $\tilde{G}'_2$ is $\boldsymbol{\eta} = (b, c)$, where a, b, c are arbitrary constants.

Suppose that for $\tilde{g} = \{(E, 1)|(0, \Delta)\}$ we have

$$\tilde{D}_{k\bar{k}}(\tilde{g}) = \begin{pmatrix} \cos 2\pi\Delta & \sin 2\pi\Delta \\ -\sin 2\pi\Delta & \cos 2\pi\Delta \end{pmatrix}. \tag{35}$$

Then $\tilde{G}''_1 = \tilde{g}\tilde{H}\tilde{g}^{-1}$ is an isotropy subgroup equivalent to $\tilde{G}'_1$ with order parameter $\boldsymbol{\eta} = (a\cos 2\pi\Delta, -a\sin 2\pi\Delta)$. Since Δ can take any value, this order parameter can point in any direction in two-dimensional representation space. Thus, for any order parameter (b, c), there exists some value of Δ for which the order parameter of $\tilde{G}''_1$ is in the same direction. Because there does not exist any direction of the order parameter for which the operators in $\tilde{G}'_2$ are the only operators that satisfy equation (6), $\tilde{G}'_2$ is not really an isotropy subgroup of $\tilde{G}_{k\bar{k}}$, even though it satisfies the chain rule.

In the example above, $\tilde{h}_1$ belongs to $\tilde{G}_k$ and $\tilde{h}_2$ belongs to $\tilde{g}_{\bar{k}}\tilde{G}_k$. The off-diagonal form of the matrix in equation (21) is diagonal in equation (32) because the similarity transformation that brings $\tilde{D}_{k\bar{k}}(\tilde{h}_2)$ to real form also diagonalizes it. The assignment of operators to $\tilde{G}_k$ and $\tilde{g}_{\bar{k}}\tilde{G}_k$ can be checked in the following way. The eigenvalues of $\tilde{D}_{k\bar{k}}(\tilde{g})\tilde{D}_{k\bar{k}}(\tilde{h}_1)$ depend on Δ and thus $\tilde{h}_1$ belongs to $\tilde{G}_k$. The eigenvalues of $\tilde{D}_{k\bar{k}}(\tilde{g})\tilde{D}_{k\bar{k}}(\tilde{h}_2)$ do not depend on Δ and thus $\tilde{h}_2$ belongs to $\tilde{g}_{\bar{k}}\tilde{G}_k$.

We add an extension to the chain rule: if $\tilde{G}'$ contains only operators in $\tilde{G}_k$ and if there exists an isotropy subgroup $\tilde{G}''$ which contains all of the operators in $\tilde{G}'$ plus some operators in $\tilde{g}_{\bar{k}}\tilde{G}_k$, we require that $i(\tilde{G}') > i(\tilde{G}'') + 1$. In the above example, the extended chain rule is not satisfied and $\tilde{G}'_2$ is not an isotropy subgroup.

3.5. Identifying superspace groups

Once we obtain the operators g in an isotropy subgroup, we need to identify its (3+1)-dimensional superspace-group symmetry as being equivalent to one of the 775 such groups that have been tabulated (Janssen *et al.*, 2004). Let $\tilde{G}_1$ be the group of operators $\{\tilde{g}_{1i}\}$ in our isotropy subgroup (in the setting of the parent space group), and let $\tilde{G}_2$ be the group of operators $\{\tilde{g}_{2i}\}$ (in the standard setting of one of the supergroups). [ISO(3+1)D contains what we consider to be the 'standard setting' of each superspace group.] Our task is to try to find the matrix S in equation (13) for some one-to-one mapping of operators in $\tilde{G}_1$ onto operators in $\tilde{G}_2$. If successful, we have identified the superspace-group symmetry of the isotropy subgroup.

Table 1
Isotropy subgroups of the (3+1)-dimensional superspace extension of space group No. 220 $I\bar{4}3d$.

For each IR, we give the order-parameter direction $\boldsymbol{\eta}$, the superspace-group symmetry of the subgroup, the basis vectors of the lattice of the subgroup in terms of the basis vectors of the lattice of the parent group and the origin of the subgroup with respect to the origin of the parent group.

k	IR	$\boldsymbol{\eta}$	Subgroup	Basis vectors	Origin
$\Delta = (0, \beta, 0)$	Δ_1	$(a, 0)$	122.1 $I\bar{4}2d(00\gamma)$	(1, 0, 0, 0), (0, 0, −1, 0), (0, 1, 0, 0), (0, 0, 0, 1)	(1/4, 3/8, 0, 0)
	Δ_2	$(a, 0)$	122.1 $I\bar{4}2d(00\gamma)$	(1, 0, 0, 0), (0, 0, −1, 0), (0, 1, 0, −2), (0, 0, 0, 1)	(1/4, 3/8, 0, 1/4)
	$\Delta_3\Delta_4$	$(a, 0, b, 0)$	43.2 $Fdd2(00\gamma)s0s$	(−1, 0, 1, 0), (1, 0, 1, 0), (0, 1, 0, 2), (0, 0, 0, 1)	(−1/4, 1/4, 1/2, 0)
		(a, a, b, b)	24.2 $I2_12_12_1(00\gamma)00s$	(1, 0, 0, 0), (0, 0, −1, 0), (0, 1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/4, 0)
		(a, b, c, d)	5.3 $B2(00\gamma)s$	(1, 0, −1, 0), (−1, 0, 0, 0), (0, 1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/2, 0)
$\Lambda = (\alpha, \alpha, \alpha)$	$\Lambda_1\Lambda_1$	(a, b)	161.1 $R3c(00\gamma)$	(1, 0, −1, 0), (−1, 1, 0, 0), (1/2, 1/2, 1/2, 0), (0, 0, 0, 1)	(0, 0, 0, 0)
	$\Lambda_2\Lambda_2$	(a, b)	161.1 $R3c(00\gamma)$	(1, 0, −1, 0), (−1, 1, 0, 0), (1/2, 1/2, 1/2, 3), (0, 0, 0, 1)	(0, 0, 0, 0)
	$\Lambda_3\Lambda_3$	$(a, b, 0, 0)$	146.2 $R3(00\gamma)t$	(−1, 1, 0, 0), (0, −1, 1, 0), (1/2, 1/2, 1/2, 0), (0, 0, 0, 1)	(0, 0, 0, 0)
		$(a, b, a, \bar{b})$	9.1 $Bb(\alpha\beta0)$	(0, 0, −1, 0), (1/2, 1/2, −1/2, 0), (1, −1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/4, 0)
		$(a, b, \bar{a}, b)$	9.1 $Bb(\alpha\beta0)$	(0, 0, −1, 0), (1/2, 1/2, −1/2, 1), (1, −1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/4, 0)
		(a, b, c, d)	1.1 $P1(\alpha\beta\gamma)$	(−1/2, 1/2, 1/2, 0), (1/2, −1/2, 1/2, 0), (1/2, 1/2, −1/2, 0), (0, 0, 0, 1)	(0, 0, 0, 0)
$\Sigma = (\alpha, \alpha, 0)$	Σ_1	$(a, 0)$	43.3 $F2dd(00\gamma)$	(0, 0, −1, 0), (−1, 1, 0, 0), (1, 1, 0, 0), (0, 0, 0, 1)	(0, 1/4, 0, 0)
	Σ_2	$(a, 0)$	43.3 $F2dd(00\gamma)$	(0, 0, −1, 0), (−1, 1, 0, 0), (1, 1, 0, 2), (0, 0, 0, 1)	(0, 1/4, 0, 0)
$D = (1/2, 1/2, \gamma)$	D_1D_1	$(a, b, 0, 0)$	9.1 $Bb(\alpha\beta0)$	(0, 0, −1, 0), (1/2, 1/2, −1/2, 0), (1, −1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/4, 0)
		$(a, b, a, \bar{b})$	5.4 $B2(0\frac{1}{2}\gamma)$	(1, −1, 0, 0), (−1, 2, 0, 0), (0, 0, 1, 0), (0, 0, 0, 1)	(0, 1/4, 0, 0)
		(a, b, b, a)	9.1 $Bb(\alpha\beta0)$	(0, 0, 1, 1), (−1/2, 1/2, 1/2, 0), (−1, −1, 0, −1), (0, 0, 0, 1)	(0, 0, 0, 0)
		(a, b, c, d)	1.1 $P1(\alpha\beta\gamma)$	(−1/2, 1/2, 1/2, 0), (1/2, −1/2, 1/2, 0), (1/2, 1/2, −1/2, 0), (0, 0, 0, 1)	(0, 0, 0, 0)
$G = (\alpha, \bar{\alpha}, 1)$	G_1G_2	$(a, 0, b, 0)$	9.1 $Bb(\alpha\beta0)$	(0, 0, 1, 2), (−1/2, 1/2, 1/2, 0), (−1, −1, 0, −2), (0, 0, 0, 1)	(0, 0, 0, 0)
		(a, a, b, b)	5.1 $B2(\alpha\beta0)$	(1, −1, 0, 1), (−1, 2, 0, −1), (0, 0, 1, 1), (0, 0, 0, 1)	(0, 1/4, 1/4, 0)
		(a, b, c, d)	1.1 $P1(\alpha\beta\gamma)$	(−1/2, 1/2, 1/2, 0), (1/2, −1/2, 1/2, 0), (1/2, 1/2, −1/2, 0), (0, 0, 0, 1)	(0, 0, 0, 0)
$C = (\alpha, \alpha, \gamma)$	C_1C_1	(a, b)	9.1 $Bb(\alpha\beta0)$	(0, 0, −1, 0), (1/2, 1/2, −1/2, 0), (1, −1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/4, 0)
	C_2C_2	(a, b)	9.1 $Bb(\alpha\beta0)$	(0, 0, −1, 0), (1/2, 1/2, −1/2, 1), (1, −1, 0, 0), (0, 0, 0, 1)	(−1/4, 1/4, 1/4, 0)
$A = (\alpha, \beta, 0)$	A_1	$(a, 0)$	5.1 $B2(\alpha\beta0)$	(1, −1, 0, 0), (−1, 2, 0, 0), (0, 0, 1, 0), (0, 0, 0, 1)	(0, 1/4, 0, 0)
$GP = (\alpha, \beta, \gamma)$	GP_1GP_1	(a, b)	1.1 $P1(\alpha\beta\gamma)$	(−1/2, 1/2, 1/2, 0), (1/2, −1/2, 1/2, 0), (1/2, 1/2, −1/2, 0), (0, 0, 0, 1)	(0, 0, 0, 0)

Our method for finding S is similar to the algorithm used by Hatch & Stokes (1985) for identifying the three-dimensional space-group symmetry of isotropy subgroups for commensurate **k** vectors. Writing equation (13) as $SA_2(\tilde{\mathbf{g}}_{2i}) = A_1(\tilde{\mathbf{g}}_{1i})S$ and setting $S_\varepsilon = 1$ (to preserve the right-handedness of the coordinate system), we obtain

$$\varepsilon_{2i} = \varepsilon_{1i}, \tag{36}$$

$$S_R R_{2i} = R_{1i} S_R, \tag{37}$$

$$S_M R_{2i} + M_{2i} = M_{1i} S_R + \varepsilon_{1i} S_M, \tag{38}$$

$$S_R v_{2i} + S_v = R_{1i} S_v + v_{1i}, \tag{39}$$

$$S_M v_{2i} + \delta_{2i} + S_\delta = M_{1i} S_v + \varepsilon_{1i} S_\delta + \delta_{1i}. \tag{40}$$

Using these equations, we construct an algorithm for finding the transformation S between two equivalent superspace groups. If such a transformation cannot be found, then the two groups are not equivalent.

Step 1. Choose n generating operators $\{\tilde{\mathbf{g}}_{2i}\}$ of $\tilde{G}_2$. Find a mapping of these operators onto n operators $\{\tilde{\mathbf{g}}_{1i}\}$ of $\tilde{G}_1$ and test the one-to-one correspondence $\tilde{\mathbf{g}}_{2i} \leftrightarrow \tilde{\mathbf{g}}_{1i}$. From equation (36), we see that we must choose the elements in $\tilde{G}_1$ such that $\varepsilon_{1i} = \varepsilon_{2i}$. From equation (37), we see that we must also choose the elements in G_1 such that $\det R_{1i} = \det R_{2i}$ and $\mathrm{tr}\, R_{1i} = \mathrm{tr}\, R_{2i}$. This means that each pair, R_{1i} and R_{2i}, must be the same physical type of point operator, *i.e.* twofold rotation, reflection *etc*. At this point, we specify the translational parts v_{1i}, δ_{1i} of $\tilde{\mathbf{g}}_{1i}$ only to within an integer.

Step 2. Use equation (37) to determine possible integer values for the matrix S_R. We use the same algorithm in this step as in Hatch & Stokes (1985), and require that $\det S_R = 1$ to preserve the right-handedness of the coordinate system.

Step 3. Use equation (38) to determine possible integer values for the row matrix S_M.

Step 4. Use equations (39) and (40) to solve for the column matrix S_v and the element S_δ. Since we have only specified v_{1i} and δ_{1i} to within an integer, we must solve these equations modulo 1. We do this by computing the Smith normal form of those equations (Grosse-Kunstleve, 1999). This allows us to determine whether or not there are any solutions and, if there are, to find the solutions. Using the Smith normal form is an improvement over the algorithm in Hatch & Stokes (1985).

3.6. IR matrices

We obtained the matrices $D_k(g)$ from tables in Cracknell *et al.* (1979) (CDML), which are extensions of the tables of Miller & Love (1967). In some cases, the CDML tables contain multiple IRs which belong to the same **k**-vector type. Because the results in our ISO(3+1)D tables are valid for any choice of parameters α, β, γ, even values outside the first Brillouin zone, we list only one **k** vector of each type. As a result, some of the IRs in CDML do not appear in our tables. For example, for space group No. 220 $I\bar{4}3d$, **k** vectors along the F and Λ lines are of the same type. If the Λ line is extended beyond the boundary of the first Brillouin zone, it becomes translationally equivalent to the F line. Also, the **k** vectors in the J, B and C planes are of the same type. Thus, we do not include the F, J or B IRs in our tables.

Furthermore, for many space groups, CDML omits some **k** vectors that are present for the holosymmetric space group of the same Bravais-lattice type. We include IRs for these **k** vectors in our tables. For example, for the cubic I Bravais

lattice, the A IRs included for the holosymmetric space group No. 229 $Im\bar{3}m$ are missing in the CDML table of IRs for space group No. 220 $I4\bar{3}d$, since for $I4\bar{3}d$ the A IR is just a special case of the IR at the general point (GP). We include the A IR in our tables. As we see in Table 1, in this case, the IR at A generates a different isotropy subgroup from that of the IR at the general point.

3.7. Reliability of the tables

We carefully implemented on computer each part of the algorithm and compared the results with particular examples that could be hand-calculated. In fact, parts of the algorithm were developed specifically to address problems discovered in the testing procedure (for example, the violation of the chain rule).

Our previous tables of isotropy subgroups (Stokes & Hatch, 1988) are virtually error free, and we expect the ISO(3+1)D tables to be likewise accurate. The previous tables were printed in a book. The ISO(3+1)D tables reside on the internet where we have the possibility to correct errors, if necessary.

4. Examples: using the tables

4.1. Isotropy subgroups of $I\bar{4}3d$

In Table 1, we extract from ISO(3+1)D the isotropy subgroups of the (3+1)-dimensional superspace extension of space group No. 220 $I\bar{4}3d$. Complex IRs of type 2 or 3 which have been brought to real form are denoted by symbols showing two IRs. For example, $\Delta_3\Delta_4$ is the direct sum of Δ_3 and Δ_4 which are complex conjugates of each other. $\Lambda_1\Lambda_1$ is the direct sum of Λ_1 with itself which is equivalent to its own complex conjugate.

The first isotropy subgroup in the table is 122.1 $I\bar{4}2d(00\gamma)$, which arises from the IR Δ_1 associated with the **k** vector $\Delta = (0, \beta, 0)$. The incommensurate modulation is along the y axis $(0, \beta, 0)$ in the setting of the parent group and along the z axis $(0, 0, \gamma)$ in the setting of the superspace group. We see that the third basis vector is $(0, 1, 0, 0)$, which means that the z axis in the superspace-group setting is the y axis in the parent space-group setting. From the ISO(3+1)D table of IRs, we find that the generators of $\tilde{H}$ are mapped onto the matrices in equations (33) and (34). We therefore obtain the same result as in the example in §3.4: there is one isotropy subgroup, its order-parameter direction is $(a, 0)$, and the operators in the isotropy subgroup include all of the operators in $\tilde{H}$. From the basis vectors and origin, we obtain the affine transformation matrix

$$S = \begin{pmatrix} 1 & 0 & 0 & 0 & 1/4 \\ 0 & 0 & 1 & 0 & 3/8 \\ 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \qquad (41)$$

which takes operators in the parent space-group setting into operators in the superspace-group setting *via* equation (13).

Table 1 lists four isotropy subgroups for the IR $\Lambda_3\Lambda_3$. From ISO(3+1)D, we find there are two generators of $\tilde{H}$, each mapped onto a four-dimensional matrix. By group multiplication, we obtain 36 operators in $\tilde{H}$. The image of $\tilde{H}$ is identical to image *D*36*b* listed in Stokes & Hatch (1988). This image appears for 16 different IRs associated with rational **k** vectors (*e.g.* IR H_2H_3 of space group 147 $P\bar{3}$). In each of those cases, there are also four isotropy subgroups with the same order-parameter directions as in Table 1.

4.2. K_2SeO_4

At 130 K, K_2SeO_4 undergoes a transition from a commensurate phase with crystallographic space group 62 *Pmcn* to an incommensurate phase with **k** vector $(0, 0, 1/3 - \delta)$ and superspace-group symmetry $P^{Pnam}_{\bar{1}ss} = Pmcn(00\gamma)ss0$ (Perez-Mato *et al.*, 1984*a*,*b*; Cummins, 1990), which is seen to be equivalent to 62.2 $Pmcn(00\gamma)s00$ in the ISO(3+1)D table of superspace groups. Perez-Mato *et al.* found that this transition is driven by a soft-phonon mode belonging to IR Σ_2. This transition appears in ISO(3+1)D under the three-dimensional Σ_2 IR of space group 62 *Pnma* with order-parameter direction $(a, 0, 0)$.

At 93 K, the **k** vector locks in at $(0, 0, 1/3)$ so that the Σ_2 IR produces space-group symmetry 33 $P2_1cn$, a result that we also obtain using the *ISOTROPY* program.

4.3. TTF–TCNQ

At 54 K, TTF–TCNQ undergoes a transition from a commensurate phase, space group 14 $P2_1/c$, to an incommensurate charge-density-wave phase (known as the M_{III} phase), with **k** vector $(1/2, \beta, 0)$, $\beta \approx 0.295$, and superspace-group symmetry $C^{P2_1/c}_{1\bar{1}} = 14.3\,P2_1/b(\frac{1}{2}0\gamma)$ (Bak & Janssen, 1978; Wang *et al.*, 2003). We find transitions with this symmetry in ISO(3+1)D under IRs W_1 and W_2 of space group 14 $P2_1/c$. The IRs W_1 and W_2 produce modulations which are physically different but which have the same superspace-group symmetry. With TTF at the origin in $P2_1/c$, W_1 produces a structure where the modulated displacements of the TCNQ molecules are all in phase in a given (100) plane but the TTF molecules are not. W_2 produces a structure where the modulated displacements of the TTF molecules are all in phase in a given (100) plane but the TCNQ molecules are not. It is not clear from experimental data in the literature which of these two IRs is active in the M_{III} phase of TTF–TCNQ.

At 49 K, there is another transition to a triclinic incommensurate M_{II} phase with superspace-group symmetry $P^{P\bar{1}}_{\bar{1}} = 2.1\,P\bar{1}(\alpha\beta\gamma)$, $1/4 < \alpha < 1/2$, $\beta \approx 0.295$, $\gamma \approx 0$. This symmetry is found in ISO(3+1)D under the IR for the general point (GP1) of space group 14 $P2_1/c$.

4.4. $NaNO_2$

At 436 K, $NaNO_2$ undergoes a transition from a commensurate phase, space group 71 *Immm*, to an incommensurate phase with **k** vector $\Lambda = (0, 0, \gamma)$, $\gamma \approx 0.108$, and superspace-group symmetry $P^{I2mm}_{ss1} = 44.2\,Imm2(00\gamma)s0s$ (Ziegler, 1931; Kucharczyk & Paciorek, 1985; McConnell, 1991). From

ISO(3+1)D, we see that there are four Λ IRs, all of which result in isotropy subgroups with superspace Bravais class $mmmI(00\gamma)$. In order to obtain $Imm2(00\gamma)s0s$, we must couple two different IRs. From ISO(3+1)D, we find the generators of $Imm2(00\gamma)s0s$ to be $(\bar{x}_1, x_2, x_3, x_4 + 1/2)$ and $(x_1, \bar{x}_2, x_3, x_4)$. These are operators in $Immm(00\gamma)s00$, the isotropy subgroup for IR Λ_4, so that $Imm2(00\gamma)s0s$ is a subgroup of $Immm(00\gamma)s00$. [$Immm(00\gamma)s00$ is also the isotropy subgroup for IR Λ_3 but with the a_1 and a_2 axes interchanged so that its symmetry relative to the parent is actually $Immm(00\gamma)0s0$.] In order to reduce the symmetry from $Immm(00\gamma)s00$ to $Imm2(00\gamma)s0s$, we need a ferroelectric distortion along the a_3 axis. From Table 4 in Stokes & Hatch (1988), we find that the IR of *mmm* which is associated with the z component of a polar vector is Γ_2^-. Therefore, the transition from *Immm* to $Imm2(00\gamma)s0s$ is accomplished by coupling Λ_4 and Γ_2^-.

APPENDIX A
Example of generating $\tilde{H}$

As an example of generating the group $\tilde{H}$, consider the IR A_1 of space group 17 $P222_1$. The A point is at $\mathbf{k} = (\alpha, 0, 1/2)$. The little group of $\mathbf{k}$ consists of all space-group elements containing the point operators x, y, z and $x, \bar{y}, \bar{z}$. We choose the coset representative $g_{\bar{k}} = (\bar{x}, y, \bar{z} + 1/2)$. Every operator in G takes $\mathbf{k}$ either into $\mathbf{k}$ or into $-\mathbf{k}$, so that, in this case, $G_{k\bar{k}} = G$.

The IR matrices $D(g)$ are two-dimensional:

$$\begin{aligned}
D(x, y, z) &= \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \\
D(x, \bar{y}, \bar{z}) &= \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \\
D(\bar{x}, y, \bar{z} + 1/2) &= \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \\
D(\bar{x}, \bar{y}, z + 1/2) &= \begin{pmatrix} 0 & i \\ i & 0 \end{pmatrix} \\
D(x + 1, y, z) &= \begin{pmatrix} \exp(i2\pi\alpha) & 0 \\ 0 & \exp(-i2\pi\alpha) \end{pmatrix}, \\
D(x, y + 1, z) &= \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \\
D(x, y, z + 1) &= \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}.
\end{aligned} \tag{42}$$

We also have from equation (24)

$$\tilde{D}(x, y, z, t + \Delta) = \begin{pmatrix} \exp(i2\pi\Delta) & 0 \\ 0 & \exp(-i2\pi\Delta) \end{pmatrix}. \tag{43}$$

The eigenvalues of $D(x, y, z)$ are 1 and of $D(x, \bar{y}, \bar{z})$ are ± 1, so the eligible operators in $\tilde{G}_k$ are (x, y, z, t), $(x, \bar{y}, \bar{z}, t)$ and $(x, \bar{y}, \bar{z}, t + 1/2)$. The eigenvalues of $D(\bar{x}, y, \bar{z} + 1/2)$ are ± 1 and of $D(\bar{x}, \bar{y}, z + 1/2)$ are $\pm i$. Therefore, all of the operators $(\bar{x}, y, \bar{z} + 1/2, \bar{t} + \Delta)$ are eligible, and none of the operators $(\bar{x}, \bar{y}, z + 1/2, \bar{t} + \Delta)$ are eligible.

We generate $\tilde{H}$ from the eligible operators, (x, y, z, t), $(x, \bar{y}, \bar{z}, t)$, $(x, \bar{y}, \bar{z}, t + 1/2)$ and $(\bar{x}, y, \bar{z} + 1/2, \bar{t})$. We arbitrarily choose $\Delta = 0$ for the last generator. Through group multiplication, we obtain an additional four operators in $\tilde{H}$: $(x, y, z, t + 1/2)$, $(\bar{x}, y, \bar{z} + 1/2, \bar{t} + 1/2)$, $(\bar{x}, \bar{y}, z + 1/2, \bar{t})$ and $(\bar{x}, \bar{y}, z + 1/2, \bar{t} + 1/2)$.

We can generate $\tilde{H}$ with two operators: $(x, \bar{y}, \bar{z}, t + 1/2)$ and $(\bar{x}, y, \bar{z} + 1/2, \bar{t})$. These two operators are listed in our ISO(3+1)D table of IRs in terms of the superspace lattice: $(x_1, \bar{x}_2, \bar{x}_3, \bar{x}_3 + x_4 + 1/2)$ and $(\bar{x}_1, x_2, \bar{x}_3 + 1/2, \bar{x}_4)$.

The IR A_1 is of type 3 because IR A_1^* is equivalent to IR A_2. We obtain real matrices from the physically irreducible representation A_1A_2 which maps operators g onto matrices $D^{A_1A_2}(g) = D^{A_1}(g) \oplus D^{A_2}(g)$. Using equation (23), the generators of $\tilde{H}$ are thus mapped onto

$$\begin{aligned}
\tilde{D}^{A_1A_2}(x, \bar{y}, \bar{z}, t + 1/2) &= \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \\
\tilde{D}^{A_1A_2}(\bar{x}, y, \bar{z} + 1/2, \bar{t}) &= \begin{pmatrix} 0 & -i & 0 & 0 \\ i & 0 & 0 & 0 \\ 0 & 0 & 0 & i \\ 0 & 0 & -i & 0 \end{pmatrix}.
\end{aligned} \tag{44}$$

We next bring these matrices to real form with a transformation

$$S = (1/2)^{1/2} \begin{pmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 \\ i & 0 & -i & 0 \\ 0 & i & 0 & -i \end{pmatrix}, \tag{45}$$

resulting in

$$\begin{aligned}
S\tilde{D}^{A_1A_2}(x, \bar{y}, \bar{z}, t + 1/2)S^{-1} &= \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \\
S\tilde{D}^{A_1A_2}(\bar{x}, y, \bar{z} + 1/2, \bar{t})S^{-1} &= \begin{pmatrix} 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ -1 & 0 & 0 & 0 \end{pmatrix}.
\end{aligned} \tag{46}$$

Note that the choice of S is not unique. We simply require *any* transformation S which brings the IR matrices to real form. Our computer algorithm found the matrix S shown above.

At this point, we determine that the eight matrices in the image of $\tilde{H}$ are reducible to block-diagonal form and contain two copies of the image B8a which we found in our previous work (Stokes & Hatch, 1988). We find the transformation

$$S_2 = 1/2 \begin{pmatrix} -1 & 1 & -1 & -1 \\ 1 & 1 & 1 & -1 \\ 1 & 1 & -1 & 1 \\ -1 & 1 & 1 & 1 \end{pmatrix}, \tag{47}$$

which brings the matrices to block-diagonal form with exact copies of the image B8a as diagonal elements:

$$S_2 S\tilde{D}^{A_1A_2}(x, \bar{y}, \bar{z}, t+1/2)S^{-1}S_2^{-1} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix},$$

$$S_2 S\tilde{D}^{A_1A_2}(\bar{x}, y, \bar{z}+1/2, \bar{t})S^{-1}S_2^{-1} = \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (48)$$

Again, the choice of S_2 is not unique. Our computer algorithm found the matrix S_2 shown above. We label this image B8aB8a since it contains two copies of B8a. Among the 5508 IRs in our ISO(3+1)D tables, we find only 15 inequivalent images of $\tilde{H}$. Each of these images can be brought to a form that contains exact copies of images found in our previous work (Stokes & Hatch, 1988). The transformation S_2 is important since it brings uniformity to our tables. IRs with the same images give rise to isotropy subgroups with the same order-parameter directions.

Using the two generators above, we can easily generate the eight matrices onto which the IR maps the operators in $\tilde{H}$ and use equation (6) to determine the isotropy subgroups of $\tilde{H}$:

$$\begin{aligned}
&[1] \quad (x, y, z, t), (x, \bar{y}, \bar{z}, t) : \ \boldsymbol{\eta} = (a, \bar{a}, b, \bar{b}), \\
&[2] \quad (x, y, z, t), (x, \bar{y}, \bar{z}, t+1/2) : \ \boldsymbol{\eta} = (a, a, b, b), \\
&[3] \quad (x, y, z, t), (\bar{x}, \bar{y}, \bar{z}+1/2, \bar{t}) : \ \boldsymbol{\eta} = (0, a, 0, b), \\
&[4] \quad (x, y, z, t), (\bar{x}, \bar{y}, \bar{z}+1/2, \bar{t}+1/2) : \ \boldsymbol{\eta} = (a, 0, b, 0), \\
&[5] \quad (x, y, z, t) : \ \boldsymbol{\eta} = (a, b, c, d).
\end{aligned} \quad (49)$$

Subgroups [1] and [2] are equivalent, and subgroups [3] and [4] are equivalent (*via* a similarity transformation). Therefore, there are three non-equivalent isotropy subgroups for the IR A_1A_2.

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