Superspace Groups without the Embedding: The Link between Superspace and Fourier-Space Crystallography

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The symmetry classification of three-dimensional periodic or aperiodic crystals is given a coordinate independent formulation which establishes the precise connection between Fourier-space and superspace crystallography. Superspace groups emerge without having to embed an aperiodic crystal in a higher-dimensional space.

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Superspace crystallography classifies the symmetry of aperiodic crystals by extending them to periodic crystals in a higher-dimensional space. Their symmetries can then be expressed in terms of the conventional point groups and space groups of higher-dimensional periodic crystals. The key steps are embedding the aperiodic crystal in more than three dimensions and applying the higher-dimensional symmetry classification of periodic crystals [1].

Fourier-space crystallography treats aperiodic crystals the same way it treats ordinary three-dimensional periodic crystals, using a broader concept of three-dimensional point-group symmetry that applies equally well to both types. This generalization of real-space point-group symmetry has simple consequences in three-dimensional Fourier space. The key steps are redefining point-group symmetry, and expressing space-group symmetry in terms of sets of functions (called *gauge functions*) linear on the set of wave vectors at which the density has nonzero Fourier coefficients [2].

From the Fourier-space perspective the superspace approach is underdetermined, since the embedding is not unique. In this Letter we eliminate this arbitrariness by developing the superspace formalism in terms of the transformations induced on the space of its gauge functions by the three-dimensional point group of a crystal. To do this we introduce coordinate-independent definitions of Bravais class, arithmetic crystal class, and space-group type from which follow both the informal geometric definitions of the Fourier-space approach and the coordinate-based algebraic definitions of the superspace approach (as well as the conventional approach to three-dimensional periodic crystals [3]). This union of the Fourier-space and superspace approaches gives the complete symmetry classification of aperiodic crystals without making any use of higher-dimensional point groups.

We first review the implications of the redefinition of the point group G of a crystal [4] with equilibrium density $\rho(\mathbf{r})$ as the subgroup of $O(3)$ that leaves invariant the positionally averaged nth order autocorrelation functions of $\rho(\mathbf{r})$,

$$\lim_{V\to\infty} \frac{1}{V} \int_V d\mathbf{r}\, \rho(\mathbf{r}_1 - \mathbf{r}) \cdots \rho(\mathbf{r}_n - \mathbf{r}), \qquad (1)$$

for all $n \geq 2$. For periodic crystals this is equivalent to the conventional definition, but in aperiodic crystals there are in general no rotations that leave the density invariant to within a three-dimensional translation, even though there can be subgroups of $O(3)$ that leave all density autocorrelation functions invariant. This new definition is appropriate because two densities ρ and ρ' that agree in all positionally averaged autocorrelation functions are *indistinguishable*: Any finite subregion is as likely to have been taken from one density as from the other [5]. This expansion of the concept of point-group symmetry is essential for understanding the symmetry of aperiodic crystals and for the compact coordinate-independent construction of superspace groups that follows. It takes on a particularly simple form when expressed in terms of the density Fourier coefficients $\rho(\mathbf{k})$.

The set L of all integral linear combinations of those wave vectors at which $\rho(\mathbf{k}) \neq 0$ is called the (reciprocal) *lattice* or *Fourier module*. A set S of vectors *generates* L if every vector in L is an integral linear combination of vectors in S. The *rank* D of L is the smallest number of wave vectors necessary to generate it. (Periodic three-dimensional crystals have rank 3.) A set of wave vectors $\mathbf{b}^{(1)}, \ldots, \mathbf{b}^{(D)}$ that generates L is a *basis* if the $\mathbf{b}^{(i)}$ themselves are all in L. A fundamental theorem of module theory [6] establishes that L can always be given a basis. The *point group* G_L of a lattice L is defined in the conventional way as the subgroup of $O(3)$ that permutes its wave vectors. The point group G of a crystal is necessarily a subgroup of the point group G_L of its lattice of wave vectors.

If we express the density $\rho(\mathbf{r})$ in terms of its Fourier coefficients $\rho(\mathbf{k})$, then it easily follows that two densities are indistinguishable if and only if their density Fourier coefficients are related by

$$\rho'(\mathbf{k}) = e^{2\pi i \chi(\mathbf{k})} \rho(\mathbf{k}), \qquad (2)$$

where the *gauge function* χ is a real-valued linear function on L (which cannot, in general, be extended to a function linear on all of three-dimensional Fourier space, when $D > 3$). Density Fourier coefficients at

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wave vectors that differ by a point-group operation g are related by a particular gauge function Φ_g:

$$\rho(g\mathbf{k}) = e^{2\pi i\Phi_g(\mathbf{k})}\rho(\mathbf{k}). \quad (3)$$

The gauge functions Φ_g associated in this way with point-group operations are called *phase functions.* Phase functions Φ'_g and Φ_g that characterize indistinguishable densities ρ' and ρ are associated with the same symmetry type. It follows from (2) and (3) that two such phase functions are related by

$$\Phi'_g(\mathbf{k}) \equiv \Phi_g(\mathbf{k}) + \chi(g\mathbf{k} - \mathbf{k}), \quad (4)$$

where "$\equiv$" signifies equality to within an additive integer. Sets of phase functions related by (4) are said to be *gauge equivalent*.

A relation between phase functions associated with different point-group elements, the *group compatibility condition*, follows directly from (3) and the identity $\rho([gh]\mathbf{k}) = \rho(g[h\mathbf{k}])$:

$$\Phi_{gh}(\mathbf{k}) \equiv \Phi_g(h\mathbf{k}) + \Phi_h(\mathbf{k}). \quad (5)$$

This completes our review of point-group symmetry.

The concepts of Bravais class, arithmetic crystal class, and space-group type can now be defined directly in terms of the rank-D lattice L of three-dimensional wave vectors, its point group G_L, the point group G of the periodic or aperiodic crystal, and the associated phase functions.

Two lattices of wave vectors L and L' are in the same *Bravais class* if their point groups G_L and $G_{L'}$ are conjugate subgroups of $O(3)$,

$$G_{L'} = r\,G_L\,r^{-1}, \quad (6)$$

for some proper [7] three-dimensional rotation r, and there is an invertible linear map f from L to L' that commutes with their point groups; i.e., for all $\mathbf{k}$ in L,

$$f(g\mathbf{k}) = g'f(\mathbf{k}), \quad g' = rgr^{-1}. \quad (7)$$

The existence of f ensures that the lattices are isomorphic as Abelian groups and therefore, in particular, that they have the same rank. That isomorphism is required by (7) to preserve the action of the point groups on their lattices. (It is this proviso that distinguishes, for example, the three rank-3 cubic Bravais classes.) Since a point-group operation induces a linear map on L, we can define a product of linear maps,

$$f \circ g\ (\mathbf{k}) = f(g\mathbf{k}), \quad g' \circ f\ (\mathbf{k}) = g'[f(\mathbf{k})], \quad (8)$$

in terms of which condition (7) requires that

$$rgr^{-1} = f \circ g \circ f^{-1}. \quad (9)$$

Two crystals are in the same *arithmetic crystal class* if their lattices L and L' are in the same Bravais class, and it is possible to choose the proper rotation r and the lattice isomorphism f in (9) so that the point groups G and G' of the crystals are also conjugate under the conjugacy between the point groups G_L and $G_{L'}$ of their lattices [8]:

$$G' = rGr^{-1}. \quad (10)$$

Two crystals have the same *space-group type* if they are in the same arithmetic crystal class, and to within a transformation of the form (4) the mapping f preserves the values of the phase functions Φ_g and $\Phi'_{g'}$ of the two crystals:

$$\Phi'_{rgr^{-1}}(f\mathbf{k}) \equiv \Phi_g(\mathbf{k}) + \chi(g\mathbf{k} - \mathbf{k}), \quad (11)$$

where χ is linear on L.

To recover from this framework the conventional Fourier-space description of the space-group types, note that in calculating or displaying the space-group types for a given Bravais class and point group G, it suffices to use a single lattice L from the class. The lattice isomorphism f and rotation r in (11) then become lattice and point-group automorphisms. The case in which f and r are both the identity establishes that each gauge-equivalence class of phase functions on L is associated with the same space-group type. The existence of nontrivial f and/or r satisfying (9) permits the possible further grouping of different gauge-equivalence classes of phase functions on L into the same space-group type. This is called the *scale equivalence* of the gauge-equivalence classes, a nomenclature reflecting the fact that just such a mapping, with f a uniform rescaling and r the identity e, played a role in the proper identification of the rank-6 icosahedral space groups [9].

If the point-group automorphism r is actually *in* G, and if f is just the linear transformation induced on L by r, then it is an elementary exercise to deduce from the group compatibility condition (5) that $\Phi'_{rgr^{-1}} \circ f$ and Φ'_g are gauge equivalent. Consequently, the scale equivalence of distinct gauge-equivalence classes is only an issue when the lattice transformation f is not induced by a member of the point group G. In aperiodic crystals such lattice transformations often involve rescalings (sometimes combined with rotations), or more general linear transformations of L that cannot be realized by any operation in $O(3)$. They also arise in the periodic case, a familiar example being permutations of the three orthogonal twofold axes that characterize the point group of an orthorhombic crystal [10]. In this case the distinct gauge-equivalence classes belonging to a single space-group type are just its *settings*.

Within the framework described above we can also give a coordinate independent definition of superspace and recover the conventional superspace formalism. *Superspace* is the set V^* of *all* real-valued linear functions Ψ on the lattice L of three-dimensional wave vectors [11]. Note that V^* is a D-dimensional vector space over the real

numbers, for if $\mathbf{b}^{(i)}$, $i = 1 \ldots D$, are a basis for L, then the set of D linear functions $\Psi^{(i)}$ on L defined by

$$\Psi^{(i)}(\mathbf{b}^{(j)}) = \delta_{ij} \tag{12}$$

are a basis for V^*: The $\Psi^{(i)}$ are complete since any Ψ can be expanded as

$$\Psi = \sum_{i=1}^{D} \Psi(\mathbf{b}^{(i)})\Psi^{(i)}, \tag{13}$$

and the $\Psi^{(i)}$ are linearly independent over the reals, since if

$$\sum_i x^i \Psi^{(i)} = 0 \tag{14}$$

then applied to $\mathbf{k} = \mathbf{b}^{(j)}$ (14) gives $x^j = 0$.

The *superlattice* L^* is the subset (a Z module) of V^* consisting of the integral-valued linear functions on L. Since phase functions Φ_g need only be specified to within an additive function from L^*, any vector Ψ_g in the set $L^* + \Phi_g$ can serve equally well as the phase function Φ_g.

Note that we have constructed the superspace V^* and superlattice L^* without going through the conventional procedure of choosing a basis for L, adding $D - 3$ additional components to each basis vector $\mathbf{b}^{(i)}$ to get a set of D wave vectors $\mathbf{B}^{(i)}$ in D dimensions, constructing a dual set of D-dimensional vectors $\mathbf{A}^{(i)}$ satisfying

$$\mathbf{A}^{(i)} \cdot \mathbf{B}^{(j)} = \delta_{ij}\,, \tag{15}$$

and defining superspace to be the set of real linear combinations of the $\mathbf{A}^{(i)}$. Our construction of superspace is just the usual coordinate-independent way of associating a dual space with a vector space, generalized to Z modules. It makes no use of any basis [12] for L or any additional components. By not extending L to a lattice of wave vectors in D dimensions, our definition of V^* makes explicit the fact that all relevant features of superspace are independent of any such extension.

The only relevant point-group symmetries in superspace are those induced by operations from the three-dimensional group of symmetries G_L of L: Any g in G_L takes a linear function Ψ on L into $\Psi \circ g$, defined by

$$\Psi \circ g\ (\mathbf{k}) = \Psi(g\mathbf{k})\,. \tag{16}$$

Superspace groups are the familiar generalization of such point-group transformations of V^*, constructed out of the set of pairs $\{g, \Psi_g\}$ with g in the point group G and Ψ_g in $L^* + \Phi_g$. The action of such a pair on a linear function Ψ in V^* is defined by

$$\Psi \circ \{g, \Psi_g\} = \Psi \circ g + \Psi_g\,. \tag{17}$$

It follows from repeated application of (17) that the group combination law must be the semidirect product

$$\{g, \Psi_g\} \circ \{h, \Psi_h\} = \{gh, \Psi_g \circ h + \Psi_h\}. \tag{18}$$

That the function paired with gh in (18) is indeed in $L^* + \Phi_{gh}$ follows from the group compatibility condition (5).

The superspace groups are conventional space groups in the D-dimensional space V^* of real-valued linear functions on the lattice of wave vectors L. The associated point group is just the three-dimensional point group G of the crystal, through its action on V^* given by (16). The phase function Φ_g is, to within an additive vector of the superlattice L^*, the superspace translation that accompanies the point-group operation g in the pair $\{g, \Psi_g\}$.

We recover the algebraic description of space groups (when $D = 3$) or of superspace groups (when $D > 3$) by picking a particular basis of D three-dimensional wave vectors $\mathbf{b}^{(i)}$ for the lattice of wave vectors L. The transformation of these under the point group G defines a D-dimensional representation of G by matrices $\mathcal{D}$ of integers:

$$g\mathbf{b}^{(i)} = \sum_j \mathbf{b}^{(j)} \mathcal{D}^{ji}(g)\,. \tag{19}$$

The transformation (16) of the associated basis (12) for the space V^* of linear functions on L becomes

$$\Psi^{(i)} \circ g = \sum \mathcal{D}^{ij}(g)\Psi^{(j)}. \tag{20}$$

There is a similar expression for the transformation (17) on V^* induced by the pairs $\{g, \Psi_g\}$. Each phase function Ψ_g associated with g has the expansion

$$\Psi_g = \sum a_g^i \Psi^{(i)}, \quad a_g^i = \Psi_g(\mathbf{b}^{(i)})\,. \tag{21}$$

In terms of the D-dimensional matrices $\mathcal{D}(g)$ and the D-dimensional vectors a_g, the superspace product (18) acquires the coordinate-dependent form

$$\{\mathcal{D}(g),\, a_g\}\{\mathcal{D}(h),\, a_h\} = \{\mathcal{D}(g)\mathcal{D}(h),\, a_g\mathcal{D}(h) + a_h\}. \tag{22}$$

The definition of space-group type given above reduces to the conventional definition [3] when translated into the language of these matrices.

This completes the unification of the superspace and Fourier-space symmetry classifications. There is, however, an important nomenclatural difference that should be noted. As originally developed for the treatment of modulated crystals and as subsequently applied to intergrowth compounds, the superspace concepts of Bravais class and space group make finer discriminations by further restricting the linear maps f in (7) and (11). Various kinds of restrictions specify various *equivalence relations.* In the

case of modulated crystals these restrictions require the lattice automorphism f in (7) to take an appropriate rank-3 sublattice of L, the *lattice of main reflections*, into itself. In the case of intergrowth compounds f might be required to take two or more such subspaces into themselves. In the case of icosahedral quasicrystals no such restrictions are appropriate or possible. In the case of crystals with rank-6 tetrahedral lattices [13] such restrictions are possible, but whether they are appropriate or not depends on whether one is using the space-group type to describe tetrahedrally modulated cubic crystals or tetrahedrally distorted icosahedral quasicrystals. For these reasons we believe it is better to retain the nomenclature used in the periodic case, characterizing any such restrictions as specifying different *settings* of the Bravais class or space-group type.

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[1] For a review, see T. Janssen, A. Janner, A. Looijenga-Vos, and P. M. de Wolff, in *International Tables for Crystallography*, edited by A. J. C. Wilson (Kluwer Academic, Dordrecht, 1992), Vol. C, p. 797.

[2] For a review, see D. A. Rabson, N. D. Mermin, D. S. Rokhsar, and D. C. Wright, Rev. Mod. Phys. **63**, 699 (1991); N. D. Mermin, Rev. Mod. Phys. **64**, 3 (1992).

[3] For a summary, see H. Wondratschek, in *International Tables for Crystallography*, edited by T. Hahn (Kluwer Academic, Dordrecht, 1995), Vol. A, p. 711.

[4] We use the term "crystal" to mean periodic *or* aperiodic crystal, refining the term further only when we wish to distinguish between periodic and aperiodic crystals, or to emphasize that we have both types in mind.

[5] Two *tilings* have indistinguishable densities if they belong to the same *local isomorphism class*. Indistinguishable densities that differ by more than a three-dimensional translation are said to differ by a *phason.*

[6] The theorem establishes that every Z module has a basis. See, for example, B. Hartley and T. O. Hawkes, in *Rings, Modules and Linear Algebra* (Chapman and Hall, London, 1970). A Z module is defined as a vector space except that the scalars are restricted to be integers. The lattice L generated by a set of three-dimensional wave vectors is precisely such a structure.

[7] The exclusion of conjugacy via improper rotations is irrelevant in three dimensions, since proper and improper rotations differ by the inversion, which drops out of (6). If the physical space of interest has an even number of dimensions, however, requiring conjugacy by a proper rotation preserves the distinction between enantiomorphic pairs of Bravais classes. This possibility arises, for example, in the aperiodic two-dimensional case. See N. D. Mermin, D. S. Rokhsar, and D. C. Wright, Phys. Rev. Lett. **58**, 2099 (1987).

[8] An important but peripheral technical point that arises in the periodic case under the discussion of *Bravais flocks*: Two crystals are also assigned the same arithmetic crystal class even if one of the lattices (say, L') has a higher symmetry than the other, provided the two crystals continue to be related by (6)–(10), where $G_{L'}$ is a *subgroup* of the full point group of L'. This is because the extra symmetry of L' is, as a matter of physics, accidental and irrelevant. Thus a crystal with a tetragonal point group G whose c/a ratio drops from 1.01 to 0.99 as a function of temperature happens to have a lattice in a cubic Bravais class at a single temperature. But even at that unique temperature the crystal is placed in a tetragonal arithmetic crystal class, because the point group of the crystal is not cubic. An arithmetic crystal class is identified by its point group G and the lowest symmetry Bravais class associated with it in this extended sense.

[9] D. S. Rokhsar, D. C. Wright, and N. D. Mermin, Phys. Rev. B **37**, 8145 (1988).

[10] A less familiar example is the tetrahedral space-group type $P\frac{2_1}{a}\overline{3}$ ($Pa3$) which has two distinct gauge-equivalence classes related by the lattice transformation f induced by a fourfold rotation.

[11] The $*$ used here and below is the conventional sign for a dual space; it is not the $*$ used to distinguish reciprocal space from real space.

[12] We introduced a basis to prove that V^* was a D-dimensional vector space, but that basis played no role in the construction of V^*.

[13] J. Dräger, R. Lifshitz, and N. D. Mermin, in *Proceedings of the 5th International Conference on Quasicrystals*, edited by C. Janot and R. Mosseri (World Scientific, Singapore, 1995), p. 72.