Copernican Crystallography

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Abstract. Redundancies are pointed out in the widely used extension of the crystallographic concept of Bravais class to quasiperiodic materials. Such pitfalls can be avoided by abandoning the obsolete paradigm that bases ordinary crystallography on microscopic periodicity. The broadening of ordinary crystallography to include quasiperiodic materials is accomplished by defining the point group in terms of indistinguishable (as opposed to identical) densities.

A periodic material (crystal) is characterized by a lattice of vectors [1] specifying the translations that leave its density unchanged. A century and a half ago Frankenheim classified such lattices by their symmetry, counting 15 types. A few years later Bravais pointed out that two of the Frankenheim classes contained identical lattices, and today the edifice of crystallography rests on a foundation of 14 Bravais classes [2].

Within the past two decades the discovery of many quasiperiodic materials (displacively modulated crystals, substitutionally modulated crystals, incommensurate intergrowth compounds, quasicrystals) has stimulated efforts to extend the crystallographic classification scheme to include these novel structures. Roughly speaking quasiperiodic materials have the property — reminiscent of but weaker than periodicity — that subregions of arbitrary size can be found reproduced elsewhere in the material at distances of the order of that size. This notion is made precise by the Fourier space definition of quasiperiodicity, which gives a simple and natural expression of the close connection between periodic and quasiperiodic materials. Densities of either type of material are superpositions of plane waves whose wave vectors can be expressed as a lattice of integral
linear combinations of $3+d$ primitive wave vectors that span a three dimensional space and are linearly independent over the integers. A material is periodic if $d = 0$ and quasiperiodic if $d > 0$. To emphasize that the vectors in such lattices are wave vectors rather than translations, one may refer to them as reciprocal lattices.

Although originally viewed as containing lattices of translations, the Bravais classes of periodic materials can equally well be regarded as classes of reciprocal lattices. Since quasiperiodic materials have no 3-dimensional translational symmetry but continue to be described by a lattice of wave vectors, it is only in Fourier space that the concept of Bravais class can directly be applied to them. The first attempt at such a classification, using the less direct superspace formalism described below, was made over a decade ago by Janner, Janssen, and de Wolff (JJdW) [3,4] for the simplest quasiperiodic materials with crystallographic point group symmetries. Such materials can be described by a reciprocal lattice with $d = 1$ for 6 of the 7 crystals systems; for the 7th (cubic) system the minimum $d$ is 3. The JJdW catalog lists 24 (3+1) Bravais classes, and 14 (3+3) cubic Bravais classes.

The narrow purpose of this Letter is to point out that 8 pairs of the (3+1) JJdW Bravais classes are identical — *i.e.* each of the two classes contains exactly the same lattices of 3-dimensional wave vectors — as are 3 pairs and one trio of the (3+3) cubic Bravais classes. The specific pairs and the trio are listed in Tables 1 and 2. Those engaged in crystallographic studies of incommensurately modulated materials can readily confirm these identifications by simply working out the general form of the lattices of ordinary 3-dimensional wave vectors belonging to each JJdW Bravais class. A direct derivation in 3-dimensional Fourier space of these 16 (3+1) and 9 (3+3) cubic Bravais classes is given elsewhere [5].

My broader purpose is to comment on the reasons behind this redundancy of description [6]. These and other anomalies in the existing generalization of crystallography to quasiperiodic materials are unlikely to disappear until crystallographers abandon the venerable but outdated enshrinement of periodicity as the *sine qua non* of their taxon-
omy. By the time quasiperiodic materials were discovered the view that crystallography is limited to the classification of periodic materials was so entrenched that the extension to quasiperiodic materials was achieved only by expressing them as 3–dimensional sections of materials periodic in more than three dimensions, to which the higher dimensional crystallography of periodic materials could then be applied. By liberating crystallography from its historic reliance on periodicity, one can avoid climbing up into superspace in search of periodicity for the hazardous purpose of coming back down with a bag full of categories, simply by taking a step sideways from 3–dimensional position space into 3–dimensional Fourier space. In Fourier space, as already noted, the distinction between periodic and quasiperiodic materials is elementary, and the fundamental concept of a Bravais class of lattices can be trivially extended from the periodic to the quasiperiodic case [7] without ever leaving 3 dimensions.

The virtues of Fourier space, even as the venue for the traditional crystallography of periodic materials, were celebrated by A. Bienenstock and P. P. Ewald [8] three decades ago. They pointed out that the 230 crystallographic categories of Schönflies, Fedorov, and Barlow could be derived simply and efficiently in Fourier space as classes of phase relations between density Fourier coefficients at wave vectors related by point–group operations. Quasiperiodic materials not then having attracted serious attention, Bienenstock and Ewald presented their method only as a more powerful approach to the ordinary crystallography of periodic materials. Their Fourier space classification scheme can, however, be directly derived without any appeal to periodicity in 3 or any other number of dimensions as the natural way to classify the broader class of quasiperiodic materials [9–11]. Since Fourier space offers an unorthodox but more effective route to the ordinary crystallography of periodic materials, since Fourier space provides the simplest definition of quasiperiodicity, and since the Fourier space route to crystallography applies equally well to both periodic and quasiperiodic materials, the single advantage of ascending to superspace in search of a classification scheme based on periodicity, is that it relieves one of
having to take a radical new look at the foundations of ordinary crystallography [12].

The key to a reformulation of crystallography that does not rely on periodicity, and perhaps the most important inducement for working in Fourier space [13], emerges from the concept of indistinguishable — as opposed to identical — densities. Two densities are indistinguishable if their positionally averaged \( n \)-point autocorrelation functions are the same for all \( n \) — \textit{i.e.} if any substructure on any scale that occurs in one occurs in the other with the same frequency. Two periodic densities are so strongly constrained by the condition of periodicity that they can be indistinguishable only if they differ by at most a translation, but two quasiperiodic densities can be indistinguishable even when they are not so simply related.

The concept of indistinguishable densities resolves a puzzle about quasiperiodic materials (such as 5-fold Penrose tilings). Many of them clearly appear to possess certain real-space point-group symmetries even though no origin can be found about which a point group operation takes the density into itself at arbitrary distances. (Although this is not often emphasized, they therefore lack strict rotational as well as translational symmetry.) How is this to be understood? The puzzle is resolved by redefining the point group of a quasiperiodic material to be the set of operations from \( O(3) \) that take the density into one that is not \textit{identical to}, but merely \textit{indistinguishable from} the original. With the relaxation of identity to indistinguishability, such point group operations become strictly valid, and in fact apply about any origin whatever.

Should a material happen to be periodic, indistinguishability reduces to identity to within a translation; one is then led to extend the point group to a space group which includes the translations that combine with point group operations to leave the density identical to what it was. Should the material be quasiperiodic, however, translations cease to be relevant. The role of the space group in classifying such materials is played by the point group, supplemented by an elementary consequence of the use of indistinguishability in the definition of the point group. Indistinguishability assumes a very simple form in
Fourier space: two densities are indistinguishable if the products of their Fourier coefficients over any set of wave vectors summing to zero always agree. This leads straightforwardly to a classification scheme, valid for periodic or quasiperiodic materials, that is based on point group symmetry and the phase relations between density Fourier coefficients at symmetry related wave vectors [9–11]. For periodic materials the resulting categories can also be regarded as subgroups of the real–space Euclidean group (the crystallographic space groups) because of the simple form indistinguishability assumes in the presence of periodicity. For quasiperiodic materials, however, and for a powerful and broader approach to periodic materials as well, one is better off building the classification scheme out of the point group of operations that take the density into something indistinguishable, and the phase relations between Fourier coefficients related by operations from that point group [14].

Tables 1 and 2 provide a simple example of the importance of staying in 3-dimensions. The redundancies in the JJdW catalog correspond to the different (3+0) crystallographic sublattices that can be found within any 3–dimensional lattice from the given (3+d) Bravais class [15]. The particular way in which the lattices in two such JJdW classes are embedded in superspace obscures the fact that both classes contain identical sets of ordinary 3–dimensional wave-vectors. Information about the different (3+0) sublattices of the (3+d) lattices in a given Bravais class is quite useful in applying the scheme to the diffraction patterns of those quasiperiodic materials where such (3+0) sublattices can be associated with a crystallographic lattice of main reflections and the remaining wave vectors, with weaker satellite peaks. But to count as distinct Bravais classes these different ways in which one and the same class of (3 + d) lattices can be manifest in particular materials is to abandon the view of a Bravais class as a class of lattices of 3–dimensional wave vectors, that serve as a template for a diffraction pattern independent of the particular intensities of the Bragg peaks that are actually observed [16]. By building such distinctions into the concept of Bravais class one unnecessarily creates different crystallographies for
displacively modulated crystals (where the distinctions are relevant) and quasicrystals or
incommensurate intergrowth compounds (where they are not); one obscures relations such
as that between the 3 Bravais classes of icosahedral quasicrystals and the 3 (not 4) (3+3)
cubic Bravais classes with tetrahedral symmetry, noted in the caption of Table 2; and
when one comes to compute the classes of phase relations (which give the space groups
in the periodic case and are currently displayed in the form of superspace groups in the
quasiperiodic case), by failing to recognize that some Bravais classes contain identical
3–dimensional lattices of wave vectors one ends up calculating and tabulating the same
information more than once.

If the Sun were the only thing of interest in the heavens, it would be foolish not
to regard it as moving around the Earth. Because this view became firmly entrenched,
generations of astronomers had to learn about epicycles to account for the motions of the
planets. While it was wrenching to shift to a heliocentric perspective, the resulting simpli-
fication in the more broadly applied scheme more than made up for the pain of abandoning
the Ptolemaic view. When all materials of interest were periodic, a crystallography based
on periodicity grew and thrived. Epicycles appeared, in the form of superspace groups,
when the scheme was extended to quasiperiodic materials without abandoning its concep-
tual reliance on periodicity. While unwilling to burn for it at the stake, I would like to
suggest that others could spare themselves significant pain by abandoning Ptolemaic crys-
tallography and learning how to classify both periodic and quasiperiodic materials, not by
ascending to superspace but by resting the foundations of crystallography on the concept
of a point group of operations that change the density into something indistinguishable.

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lography might not be in need of a paradigm shift. Veit Elser, Chris Henley, and Michael Widom commented helpfully, with varying degrees of approbation or disapprobation, on earlier versions of the manuscript. This research is supported by the National Science Foundation, Grant No. DMR 8920979.

References

[1] A set of vectors is a lattice if $v - w$ is in the set whenever $v$ and $w$ are.

[2] The contributions of Frankenheim (1842) and Bravais (1848) are described in A. E. H. Tutton, *Crystallography and Practical Crystal Measurement*, MacMillan, London, 1922.

[3] P. M. de Wolff, T. Janssen, A. Janner, Acta Cryst. A37, 625 (1981).

[4] A. Janner, T. Janssen, and P. M. de Wolff, Acta Cryst A39, 658, 667, 671 (1983).

[5] N. D. Mermin and R. Lifshitz, submitted to Acta Crystallographica A.

[6] I stress that the redundancy is there, independent of my diagnosis of the underlying problem. I comment further on the redundancy in the context of that diagnosis, at the end of this Letter.

[7] Ref. [5] gives a geometrical definition for when two $(3 + d)$ lattices are in the same Bravais class, but it is not relevant to the problem with the JJdW Bravais classes noted here, that Bravais classes counted as distinct in their scheme sometimes contain *identical* lattices of 3-dimensional wave-vectors.

[8] A. Bienenstock, and P. P. Ewald, Acta Cryst.15, 1253 (1962).

[9] D. S. Rokhsar, D. C. Wright, and N. D. Mermin, Acta Cryst. A44, 197 (1988).

[10] D. A. Rabson, N. D. Mermin, D. S. Rokhsar, and D. C. Wright, Revs. Mod. Phys. 63, 699 (1991).

[11] N. D. Mermin, to appear in Revs. Mod. Phys. 64, January, 1992.

[12] Superspace can, of course, be used advantageously for other purposes, such as building models that suggest where the atoms might be in real 3-space. See, for example, V.
[13] X-ray diffractionists should need no such inducement. Since they see the microscopic material directly in Fourier space, what could be more natural than to formulate the crystallographic categories there as well?

[14] These phase relations play no role in the particular application made here, which addresses only the concept of Bravais class. I mention them (a) because a simplification of the concept of Bravais class that left one unable to formulate finer details of the classification scheme would be worthless, and (b) because they provide, through their connection to the real space concept of indistinguishable densities, one of the most important conceptual reasons for working in Fourier space.

[15] This is particularly clear for the pairs of JJdW Bravais classes associated with the classes I have numbered 20, 21, and 22 in Table 2.

[16] All but a finite number of the peaks will, of course, not be observed at all. The lattice itself should be viewed as a mathematical abstraction from the diffraction pattern: the set of all integral linear combinations of wave vectors determined by the observed Bragg peaks.

**Table Captions**

**Table 1.** The 16 (3+1) Bravais classes for the non-cubic crystal systems. They contain lattices with the point group symmetry of the crystal system, made up of vectors that are integral linear combinations of 4 integrally independent vectors. As the nomenclature in the first column is intended to indicate, 14 of the 16 Bravais classes contain lattices that are simply given by all the sums of all pairs of vectors, one from a lattice in a conventional crystallographic Bravais class, and the other, an arbitrary integral multiple of a single additional vector. The second and third columns list the names and numbers of the corresponding JJdW Bravais class (or classes). When two JJdW classes appear on the same line, they describe identical lattices of wave-vectors, but viewed from the perspective of one
or the other of two different 3+0 crystallographic sublattices. Redundant JJdW classes are listed within braces ({}). The correspondence between the Bravais class designations in the first and second columns should be obvious in the 14 simple cases. The JJdW labels characterize the lattices in terms of a crystallographic sublattice (specified by the capital letter) to each vector of which is added arbitrary integral linear combinations of the vector listed in parentheses and all its images under the point group of the crystal system. In the first column the designations \( I^* \) and \( F^* \) refer to what are conventionally called the orthorhombic \( F \) and \( I \) Bravais classes, as is appropriate for a classification scheme based in Fourier space.

**Table 2.** The 9 (3+3) cubic Bravais classes. The first 6 have full cubic symmetry; the last 3, only tetrahedral symmetry. Lattices in all 6 of the full cubic Bravais classes contain sums of all pairs of vectors taken from two incommensurate (3+0) cubic lattices, each of these crystallographic lattices belonging to any of the 3 crystallographic cubic Bravais classes, as the nomenclature in the first column indicates.) The JJdW symbols and numbers are given in the second column and (when a Bravais class occurs under more than one name in their catalog) in the third and fourth. The convention behind their nomenclature is as described in the caption of Table 1. The JJdW symbols in the second column for the lattices with full cubic symmetry clearly reveal their relation to the designations in the first column. Redundant JJdW symbols in the third and fourth columns are given in braces ({})) to emphasize that they do not describe additional classes of lattices. As in Table 1, \( F^* \) is synonymous with but preferable to \( I \) (and similarly for \( I^* \) and \( F \)) since the Bravais classes contain lattices of vectors in Fourier space. If one ignores superspace and considers wave-vectors in ordinary 3-dimensional Fourier space, it should be evident that the Bravais classes with full cubic symmetry described by the redundant JJdW symbols in the third column contain lattices of wave-vectors identical to those in the Bravais classes described by the symbols in the second. Establishing this for the symbol in the fourth column requires a small amount of analysis. The remaining three cubic (3+3)
Bravais classes contain lattices with only tetrahedral symmetry. Quasicrystallographers might note that the three icosahedral $F^*$, $P$, and $I^*$ lattices are nothing but the (3+3) cubic lattices $T_0$, $T_1$, and $T_2$ (respectively), with the ratio of the two incommensurate length scales set at a special value that raises the point group symmetry from tetrahedral to icosahedral.
## TABLE 1

|        |     |
|--------|-----|
| **Triclinic** |     |
| $P + 1$ | 1   |
| $P(\alpha\beta\gamma)$ | 1   |
| **Monoclinic** |     |
| $P + 1_{ab}$ | 2   |
| $P2/m(\alpha\beta\delta)$ | 2   |
| $B + 1_{ab}$ | 3   |
| $B2/m(\alpha\beta\delta)$ | 4   |
| $\{ P2/m(\alpha\beta\delta^3/2) \}$ | 6   |
| $P + 1_c$ | 4   |
| $P2/m(00\gamma)$ | 5   |
| $C + 1_c$ | 5   |
| $B2/m(00\gamma)$ | 6   |
| $\{ P2/m(1/20\gamma) \}$ | 8   |
| $M$ | 6   |
| $B2/m(0\gamma)$ | 7   |
| **Orthorhombic** |     |
| $P + 1$ | 7   |
| $Pmmm(00\gamma)$ | 9   |
| $I^* + 1$ | 8   |
| $Fmmm(00\gamma)$ | 12  |
| $\{ Pmmm(1/2\gamma) \}$ | 11  |
| $F^* + 1$ | 9   |
| $Immm(00\gamma)$ | 12  |
| $\{ Cmmm(01\gamma) \}$ | 14  |
| $C + 1$ | 10  |
| $Cmmm(00\gamma)$ | 13  |
| $A + 1$ | 11  |
| $Ammm(00\gamma)$ | 15  |
| $\{ Pmmm(0\gamma) \}$ | 10  |
| $O$ | 12  |
| $Ammm(1/2\gamma)$ | 16  |
| $\{ Fmmm(01\gamma) \}$ | 18  |
| **Tetragonal** |     |
| $P + 1$ | 13  |
| $P4/mmm(00\gamma)$ | 19  |
| $I + 1$ | 14  |
| $I4/mmm(00\gamma)$ | 21  |
| $\{ P4/mmm(1/2\gamma) \}$ | 20  |
| **Trigonal** |     |
| $R + 1$ | 15  |
| $R3m(00\gamma)$ | 22  |
| $\{ P31m(1/3\gamma) \}$ | 23  |
| **Hexagonal** |     |
| $P + 1$ | 16  |
| $P6/mmm(00\gamma)$ | 24  |
|       | Cubic                                                                 |
|-------|----------------------------------------------------------------------|
|       |                                                                     |
|       | **Cubic**                                                            |
| $P + P$ | $17 \quad Pm3m(\alpha 00) \quad 208$                              |
| $I^* + I^*$ | $18 \quad Fm3m(\alpha \alpha \alpha) \quad 217$                      |
| $F^* + F^*$ | $19 \quad Im3m(0\beta \beta) \quad 213$                                |
| $P + I^* = I^* + P$ | $20 \quad Pm3m(\alpha \alpha \alpha) \quad 215$                                |
| $P + F^* = F^* + P$ | $21 \quad Pm3m(0\beta \beta) \quad 212$                                |
| $I^* + F^* = F^* + I^*$ | $22 \quad Im3m(\alpha \alpha \alpha) \quad 216$                                |

**Tetrahedral**

|       |                                                                     |
|-------|----------------------------------------------------------------------|
| $T_0$ | $23 \quad Pm3(\frac{1}{2}\beta \beta + \frac{1}{2}) \quad 206$ |
| $T_1 = T_0 + I^*$ | $24 \quad Fm3(1\beta \beta + 1) \quad 207$                                |
| $T_2 = T_0 + I^* + I^*$ | $25 \quad Fm3(\alpha 10) \quad 205$                                |

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