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Group Cohomology and Quasicrystals II: The Three Crystallographic Invariants in Two and Three Dimensions

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Abstract

In Fourier-space crystallography, space groups are classified by their phase functions, Φ, and we can determine Φ (up to a gauge) by its invariants. Usually, the invariants imply necessary extinctions in diffraction, but among the 157 periodic non-symmorphic space groups, two are distinguished not by extinctions—they have none—but by a second type of invariant. We give a non-periodic example of a third type of invariant and assert that all invariants can be expressed as sums of just these three types.

1. Introduction

Absent some accidental vanishing of a structure factor, the space group of a crystal determines the pattern of extinctions in X-ray diffraction. Does an absence of extinctions imply symmorphicity? Among the periodic space groups, usually it does, but there are two non-symmorphic space groups with no extinctions at all, $I2_12_12_1$ and $I2_13$. In terms of the Fourier-space approach,[1–4] given a reciprocal lattice L and a point group G, the space group is determined[5] by the phase-function values $\Phi_g(k)$, where $k \in L$ and $g \in G$; this function generalizes the non-primitive translations in the non-symmorphic space groups of periodic crystals. The non-vanishing of $\Phi_g(k)$ for some k fixed by g necessarily leads to an extinction in diffraction. Functions Φ related by a gauge transformation (the generalization of translating the origin) are identified, but when $gk = k$, the phase $\Phi_g(k)$ is independent of gauge. This is an invariant (value) of type I. Mermin[4] noted that in the two exceptional periodic space groups lacking a type-I invariant, one could still construct a gauge-invariant value of the form $\Phi_g(k_h) + \Phi_h(k_g)$, where g, h belong to a particular subgroup of G and k_g, $k_h \in L$, and with König[6–8] attached this invariant to a necessary electronic degeneracy ("band sticking"). This is an example of what we call an invariant of type II. We have discovered a third type of invariant first appearing in a modulated tetragonal structure, but this is the end of the story: we will assert that under quite general conditions, any invariant can be expressed as a linear combination of invariants of the three types.

Much previous work in Fourier-space crystallography has limited its attention to reciprocal lattices of minimal rank with two-dimensional "standard" sublattices equivalent to the cyclotomic integers.[2,9,3] Progress has also been made on "modulated" (or non-minimal-rank) structures but only on a case-by-case basis, fixing both the point group and the rank.[10–11] The scheme we outline here is more general, covering all finite point groups and all finitely-generated reciprocal lattices in two and three dimensions.

The enumeration of invariants takes advantage of the homological machinery developed in references 12–15, the first of which is tutorial. In the new language, the additive group of invariants (identifying those trivially related by group compatibility) is called $H_1(G, L)$, first homology of G with coefficients in L; a representative invariant in this group is a linear combination $z = \sum_{g \in G} k_g[g]$, with $k_g \in L$, satisfying the "cycle condition" $\partial z = \sum_g (g k_g - k_g) = 0$. The cycle condition is equivalent to gauge invariance. Translating back to the original language, a phase function Φ couples with invariant z to give an invariant value through the pairing $\langle \Phi, z \rangle = \sum_g \Phi_g(k_g)$. A type-I invariant (leading to an extinction) is thus a linear "combination" of the single element, $k[g]$, where $gk = k$.

2. The three types of irreducible invariant

This section will assume the background and notations of references 12–15. A type-I invariant is a cycle of the form $k[g]$ if this is not a boundary. A type-II invariant of the König-Mermin form looks like $k_g[h] - k_h[g]$, where $k_a = q - aq \in L$, $a = g, h$, for some reciprocal-space vector q not necessarily in the lattice and where g and h commute. The existence of a non-trivial invariant of this form implies that there exists a space group for which every energy level at q is at least doubly degenerate. In proposition 8.1 of reference 14, we show that a more general form of type-II invariant implies the same degeneracy; the general form is

$$z_{\mathrm{II}} = \sigma \cap c \quad , \tag{1}$$

where $\sigma \in Z^1(H, L)$ is a 1-cocycle taking values in L, $c \in Z_2(H, \mathbb{Z})$ is a 2-cycle with coefficients in the integers, H is a subgroup of the point group G, and $\cap$ is the "cap" operator.[16–17] A type-I invariant, leading to an extinction at a lattice vector k, also implies a Bragg plane of vanishing strength perpendicular to and passing through $k/2$, so extinctions may themselves lead to band sticking.[18] König and Mermin[6] discuss some type-II invariants that are sums of type-I invariants. To distinguish these type-II invariants from those that do not arise from extinctions, we define an *irreducible* invariant of type n as one that cannot be written as a sum of invariants all of types less than n.

In the example of the next section, we shall find an irreducible invariant that can be written in the form

$$z_{\mathrm{III}} = \tfrac{2}{N} \sigma \cap c \quad , \tag{2}$$

where again $\sigma \in Z^1(H, L)$ and $c \in Z_2(H, \mathbb{Z})$ and where $N \geq 4$, the order of a rotational subgroup of G, is a power of 2. Multiplying such an invariant by $N/2$, yields, if not a boundary, an invariant of type II.

The proof that all irreducible invariants are of these three types begins with a cyclic point group H (generated by an n-fold rotation r or rotoinversion $\bar{r}$). According to §III.1 of reference 16, $H_1(H, L)$ has representatives $\{k[g] \,|\, gk = k\}$ where $g = r$ or $\bar{r}$; lattice vectors are taken modulo $(1 + g + g^2 + \ldots + g^n)L$. Any such invariants are of type I. If a point group G contains a normal, cyclic subgroup H of small index, we take advantage of the restriction-inflation sequence,[16,14] which tells us that $H_1(G, L)$ (which is what we want to calculate) is generated by the image of $H_1(H, L)$ when it's mapped back to $H_1(G, L)$ and by representatives in $H_1(G, L)$ of the much smaller $H_1(Q, L_H)$, where $Q = G/H$ and $L_H = L/\{hk - k \,|\, h \in H\}$ is the largest quotient of L fixed by H. Since most (quasi)crystallographic groups admit normal, cyclic subgroups of low index, this considerably simplifies the calculation. There are a few point groups (*e.g.*, octahedral, icosahedral) that do not fit this description; for these, we use the fact that $H_1(G, L)$ is generated by $H_1(H_i, L)$ where H_i runs through the subgroups of G of prime-power order. We find in all cases that the only irreducible invariants are of types I, II, and III.

3. An example of the type-III irreducible invariant

Irreducible type-III invariants occur for point groups $D_{2^j} = 2^j22$, $j \geq 2$, but only with certain lattices containing points related by a mirror but *not* by an axial rotation; the simplest example occurs in a tetragonal crystal ($j=2$) with a rank-5 lattice. Think of Fourier three-space as generated by the complex plane $\mathbb{C}$ and one additional vector, $\hat{z}$, so that the four-fold rotation r corresponds to multiplication by i. Pick $b \in \mathbb{C}$. Construct the lattice $L = \langle b, ib, b^*, ib^*, b_3 \rangle$, where $b_3 = \hat{z} + \frac{1+i}{2}(b - b^*)$. (The notation $\langle \ldots \rangle$ means "generated by.") Straightforward calculation shows that L is invariant under $G = D_4 = 422 = \langle r, d \rangle$, where d is the two-fold rotation about the real axis; a little more work verifies that L is incommensurate if and only if $\tan(\arg[b])$ is irrational. Calculation of the 1-cycles proceeds as in references 12–13, and we find that $H_1(G, L)$ is generated by

$$z = b_3[r] - b[d] \quad . \tag{3}$$

Four times z is a boundary, and every boundary turns out to have a coefficient in front of $b_3[r]$ equal to some multiple of 4; it follows that $2z$ is not a boundary, so there exists a space group, Φ_0, satisfying $\langle \Phi_0, z \rangle = 1/4$. We can list generators for all the cycles of types I and II, and they all have even coefficients of $b_3[r]$, so z is not an invariant of type I or type II. However, we can find an explicit $c \in Z_2(G, \mathbb{Z})$ such that $z = \frac{1}{2}\sigma \cap c$, so z is an invariant of type III. Since $2z$ is an invariant of type II, reducible to invariants of type I, the space group Φ_0 exhibits both extinctions and band sticking. More notably, the space group $2\Phi_0$ admits only the type-III invariant, since $\langle 2\Phi_0, 2z\rangle = 0$.

Finally we note that physical structures with two incommensurate modulations (although not matching those described here) have been reported in SiO_2-tridymite [19] as well as in a nearly-tetragonal high-temperature superconductor. [20] It is likely that these ideas will find application.

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