EXTINCTIONS IN SCATTERING FROM MAGNETICALLY ORDERED QUASIPERIODIC CRYSTALS

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1 Main discussion

I shall discuss here the relation between the symmetry of a magnetic crystal and extinctions in its neutron diffraction diagram. In doing so I shall provide an outline of a symmetry classification scheme for magnetically ordered crystals: A scheme which is inspired by the spin-group classification of Litvin and Opechowski [1] in which the magnetic crystal is described by a spin density field $\mathbf{S}(\mathbf{r})$, left invariant by a set of rigid motions in physical space combined with primed or unprimed [1] proper rotations in spin space. Two aspects of my presentation should be emphasized from the start:

(i) I have extended Litvin and Opechowski's original scheme to deal with quasiperiodic as well as periodic crystals. This includes any spin density field whose Fourier expansion requires only a discrete set L of wave vectors

$$\mathbf{S}(\mathbf{r}) = \sum_{\mathbf{k}\in L} \mathbf{S}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{r}} \ . \tag{1}$$

As in the case of ordinary space groups there are two approaches for such an extension. The first, as suggested by Janner and Janssen [2], is to view the quasiperiodic magnetic crystal as a 3-dimensional slice of a structure periodic in a higher-dimensional "superspace", where the classification of Litvin and Opechowski can be directly applied using *high-dimensional* spin groups. I follow the Fourier-space approach of Rokhsar, Wright, and Mermin [3], which leads one to a redefinition of the concept of a spin point group and allows for a *3-dimensional* classification scheme for any magnetic crystal — periodic or quasiperiodic.

(ii) Rather than taking the usual geometric route I shall motivate the Fourier-space classification scheme by asking a question in physics: Are there any restrictions which are imposed on the Fourier expansion (1) of a spin density field $\mathbf{S}(\mathbf{r})$ describing a *physically stable* magnetic crystal? [2] Any such restrictions will manifest themselves experimentally in a diffraction diagram produced by elastic scattering of unpolarized neutrons, since the scattering intensity at wave vector $\mathbf{k}$ is proportional to

$$|\mathbf{S}(\mathbf{k})|^2 - |\hat{\mathbf{k}} \cdot \mathbf{S}(\mathbf{k})|^2, \tag{2}$$

as shown, for example, by Izyumov *et al.* [5, equation (17.10)].

[1] A primed rotation is one which is followed by time inversion. Time inversion takes $\mathbf{S}$ into $-\mathbf{S}$.
[2] Mermin [4] has answered a similar question regarding a density function $\rho(\mathbf{r})$ describing a physically stable non-magnetic crystal. Here I extend his line of argument to the slightly more involved case of a spin density field $\mathbf{S}(\mathbf{r})$.

Since $\mathbf{S}(\mathbf{r})$ is real one has

$$\mathbf{S}(\mathbf{k})^* = \mathbf{S}(-\mathbf{k}), \tag{3}$$

so that if $\mathbf{k}$ is in L then so is $-\mathbf{k}$. The question is whether any further restrictions on the structure of L or that of the Fourier coefficients $\mathbf{S}(\mathbf{k})$ follow from the requirement of physical stability.

A stable spin density field is one which makes stationary a free energy of the general form

$$F = \sum_{n=2}^{\infty} \sum_{\mathbf{k}_1 \ldots \mathbf{k}_n \in L} \sum_{i_1 \ldots i_n \in \{x,y,z\}} A^{i_1 \ldots i_n}(\mathbf{k}_1, \ldots, \mathbf{k}_n) S_{i_1}(\mathbf{k}_1) \cdots S_{i_n}(\mathbf{k}_n). \tag{4}$$

In the absence of any external fields the interactions giving rise to the magnetic crystal are translationally and rotationally invariant. This imposes two constraints on the free energy coefficients A:

$$A^{i_1 \ldots i_n}(\mathbf{k}_1, \ldots, \mathbf{k}_n) = 0 \quad \text{unless} \quad \mathbf{k}_1 + \cdots + \mathbf{k}_n = 0, \tag{5}$$

$$\sum_{j_1 \ldots j_n} \gamma^{i_1}_{j_1} \ldots \gamma^{i_n}_{j_n} A^{j_1 \ldots j_n}(g\mathbf{k}_1, \ldots, g\mathbf{k}_n) = A^{i_1 \ldots i_n}(\mathbf{k}_1, \ldots, \mathbf{k}_n) \tag{6}$$

for any rotation g in physical space and any proper rotation γ in spin space possibly combined with time inversion. The only exception to condition (6) is that in systems with "spin-orbit" coupling the spin-space rotation γ must be the axial-vector representation of the physical space rotation g. A generic set of coefficients A can vary freely with temperature and pressure subject to these two constraints and no others. Any condition on the Fourier expansion of $\mathbf{S}$ must follow from these constraints alone.

An immediate consequence of condition (5) is that if two spin density fields $\mathbf{S}$ and $\mathbf{S}'$ are related by

$$\mathbf{S}'(\mathbf{k}) = e^{2\pi i \chi(\mathbf{k})} \mathbf{S}(\mathbf{k}), \tag{7}$$

where χ is linear to within an additive integer on the set L of wave vectors at which $\mathbf{S}(\mathbf{k}) \neq 0$, then the two spin density fields have identical free energies.[3] If the magnetic crystal happens to be periodic then one easily shows that $2\pi\chi(\mathbf{k})$ is of the form $\mathbf{k} \cdot \mathbf{d}$, so that $\mathbf{S}$ and $\mathbf{S}'$ differ only by a translation. Since the degeneracy of their free energies is generic the two spin density fields must contain the same spatial distribution of bounded substructures of arbitrary size. Otherwise, one could construct a generic free energy that would discriminate between the two. The two fields are appropriately called *indistinguishable*, and the function χ relating them is called a *gauge function*.

We adopt the notion of indistinguishability as the underlying criterion of symmetry, defining the *point group* G of the magnetic crystal as the set of operations g from $O(3)$ that leave it indistinguishable to within a transformation γ in spin space. Accordingly,

[3] We [6] have also shown the converse: that any two fields, whose Fourier coefficients have identical products $S_{i_1}(\mathbf{k}_1) \cdots S_{i_n}(\mathbf{k}_n)$ over *all* sets of wave vectors summing to zero and for *all* choices of spin components, must be related by (7).

for every pair (g,γ) there exists a gauge function, $\Phi_g^\gamma(\mathbf{k})$, called a *phase function*, which satisfies

$$\mathbf{S}(g\mathbf{k}) = e^{2\pi i\Phi_g^\gamma(\mathbf{k})}\gamma\mathbf{S}(\mathbf{k}). \tag{8}$$

Since $\mathbf{S}([gh]\mathbf{k}) = \mathbf{S}(g[h\mathbf{k}])$, one easily establishes that the transformations γ in (8) form a group Γ and that the pairs (g,γ) satisfying (8) form a subgroup of $G\times\Gamma$ called the *spin point group* $G_{\mathbf{S}}$. The corresponding phase functions must satisfy the *group compatibility condition*

$$\forall(g,\gamma),(h,\eta)\in G_{\mathbf{S}}: \quad \Phi_{gh}^{\gamma\eta}(\mathbf{k})\equiv\Phi_g^\gamma(h\mathbf{k})+\Phi_h^\eta(\mathbf{k}), \tag{9}$$

where "$\equiv$" denotes equality to within an additive integer.

A further consequence of the condition (5) of translational invariance is that if $\mathbf{k}$ is not in L then there is no linear instability against a non-zero $\mathbf{S}(\mathbf{k})$ unless there are already $\mathbf{k}_1\ldots\mathbf{k}_n$ in L with $\mathbf{k}+\mathbf{k}_1+\cdots+\mathbf{k}_n=0$. In that case there may be non-zero terms in a generic free energy of the form

$$A^{i_1\ldots i_n,i}(\mathbf{k}_1,\ldots,\mathbf{k}_n,\mathbf{k})S_{i_1}(\mathbf{k}_1)\cdots S_{i_n}(\mathbf{k}_n)S_i(\mathbf{k}), \qquad i=x,y,z, \tag{10}$$

which are linear in the components of $\mathbf{S}(\mathbf{k})$. The appearance of a non-zero $\mathbf{S}(\mathbf{k})$ with an appropriate phase can lower the free energy and would be expected unless the additional condition (6) of rotational invariance provides some generic reason for the sum of all such terms to vanish. In the absence of such a reason we conclude, using (3), that L is a *lattice* — it is closed under the addition and subtraction of its wave vectors.

The only combination of terms linear in a given component of $\mathbf{S}(\mathbf{k})$ that one can expect to generically vanish are terms related by elements (g,γ) of the spin point group with g leaving $\mathbf{k}$ invariant. These elements form a subgroup of $G_{\mathbf{S}}$ which we call the *little spin group of* $\mathbf{k}$, $G_{\mathbf{S}}^{\mathbf{k}}$. The rotational invariance (6) of the free energy coefficients A then enables one to combine all terms (10) linear in a given component of $\mathbf{S}(\mathbf{k})$ into a smaller number of terms of the form

$$A^{i_1\ldots i_n,i}(\mathbf{k}_1,\ldots,\mathbf{k}_n,\mathbf{k})\sum_{(g,\gamma)\in G_{\mathbf{S}}^{\mathbf{k}}}[\gamma^{-1}\mathbf{S}]_{i_1}(g\mathbf{k}_1)\cdots[\gamma^{-1}\mathbf{S}]_{i_n}(g\mathbf{k}_n)[\gamma^{-1}\mathbf{S}]_i(\mathbf{k}). \tag{11}$$

Using the point group condition (8) one can rewrite the sum in (11) as

$$S_{i_1}(\mathbf{k}_1)\cdots S_{i_n}(\mathbf{k}_n)\sum_{(g,\gamma)\in G_{\mathbf{S}}^{\mathbf{k}}}e^{2\pi i\left(\Phi_g^\gamma(\mathbf{k}_1)+\cdots+\Phi_g^\gamma(\mathbf{k}_n)\right)}[\gamma^{-1}\mathbf{S}]_i(\mathbf{k}). \tag{12}$$

If indeed $\mathbf{k}=-\mathbf{k}_1-\cdots-\mathbf{k}_n$ is in L then the linearity of the phase function Φ_g^γ further reduces the sum on the right side of (12) to

$$\sum_{(g,\gamma)\in G_{\mathbf{S}}^{\mathbf{k}}}[e^{-2\pi i\Phi_g^\gamma(\mathbf{k})}\gamma^{-1}\mathbf{S}]_i(\mathbf{k}) = N_{\mathbf{k}}[\mathcal{P}\mathbf{S}]_i(\mathbf{k}), \tag{13}$$

where the normalization factor $N_{\mathbf{k}}=|G_{\mathbf{S}}^{\mathbf{k}}|$ is just the order of the little spin group of $\mathbf{k}$. Thus, by summing all generically related terms we find that it is not the components of $\mathbf{S}(\mathbf{k})$ which couple linearly in terms of the form (10) but rather the components of

$\mathcal{P}\mathbf{S}(\mathbf{k})$, where $\mathcal{P}$ is an operator in spin space defined by (13). All that is left to complete the characterization of the Fourier expansion of a physically stable magnetic crystal is to examine the nature of this operator.

One can verify that $\mathcal{P}$ is a projection operator by noting that

$$\mathcal{P}^2\mathbf{S}(\mathbf{k}) = \frac{1}{{N_\mathbf{k}}^2} \sum_{(h,\eta)\in G_\mathbf{S}^\mathbf{k}} \sum_{(g,\gamma)\in G_\mathbf{S}^\mathbf{k}} e^{-2\pi i \Phi_{gh}^{\gamma\eta}(\mathbf{k})} \eta^{-1}\gamma^{-1}\mathbf{S}(\mathbf{k}) = \mathcal{P}\mathbf{S}(\mathbf{k}), \tag{14}$$

where the first equality holds due to the group compatibility condition (9) and the second uses the fact that multiplying all the elements of the group $G_\mathbf{S}^\mathbf{k}$ by a fixed one of them, (h,η), merely rearranges them. Multiplying by η and again using the group compatibility condition, we also have

$$\forall (h,\eta) \in G_\mathbf{S}^\mathbf{k}: \quad \eta[\mathcal{P}\mathbf{S}](\mathbf{k}) = e^{-2\pi i \Phi_h^\eta(\mathbf{k})}[\mathcal{P}\mathbf{S}](\mathbf{k}). \tag{15}$$

Thus, $\mathcal{P}$ projects $\mathbf{S}(\mathbf{k})$ into a subspace whose spins are simultaneous eigenvectors of all spin rotations η appearing in the little spin group $G_\mathbf{S}^\mathbf{k}$. The corresponding eigenvalues are predetermined by the symmetry of the spin density field as described by its set of phase functions. Only the component of $\mathbf{S}(\mathbf{k})$ along this subspace can couple linearly to terms of the form (10) in a generic free energy. One can also show directly from the point group condition (8) that $\mathbf{S}(\mathbf{k})$, if not equal to zero, must completely lie within this subspace.

We have thus arrived at a general characterization of the Fourier expansion (1) of a spin density field describing a physically stable magnetic crystal:

(i) Every Fourier coefficient $\mathbf{S}(\mathbf{k})$ is required to be a simultaneous eigenvector of all spin transformations in the little spin group of $\mathbf{k}$, with the eigenvalues given by the corresponding phase functions. This leads directly to the idea of *extinctions* where:

(ii) The set L of wave vectors appearing in the Fourier expansion must be a lattice (*i.e.* closed under addition and subtraction) except that if, for a given $\mathbf{k}$, a non-trivial eigenvector does not exist which satisfies the first requirement then $\mathbf{S}(\mathbf{k})$ may (and must) be absent from the Fourier expansion.

If, for example, $G_\mathbf{S}^\mathbf{k}$ contains (e,γ) and its powers where γ is an n-fold rotation about the z-axis in spin space, then repeated applications of (9) to $\Phi_e^{\gamma^n}(\mathbf{k})$ give $n\Phi_e^\gamma(\mathbf{k}) \equiv 0$, so $\Phi_e^\gamma(\mathbf{k}) \equiv j/n$. One can then verify that $\mathbf{S}(\mathbf{k})$ vanishes unless $j = 0 \bmod n$ in which case $\mathbf{S}(\mathbf{k}) = (0,0,S)$, or unless $j = \pm 1 \bmod n$ in which case $\mathbf{S}(\mathbf{k}) = (S, \pm iS, 0)$.

Those fluent in the language of group representations will have immediately recognized that due to the group compatibility condition (9) the set of eigenvalues in (15) for all the elements of the group $G_\mathbf{S}^\mathbf{k}$ form a 1-dimensional representation of that group. The operator $\mathcal{P}$ is simply the projection operator onto the subspace transforming under this 1-dimensional representation. Using this language one can easily determine whether $\mathbf{S}(\mathbf{k})$ must vanish by checking whether the 1-dimensional representation in question is contained within the decomposition of the 3-dimensional axial-vector representation when the latter is restricted to the subgroup $G_\mathbf{S}^\mathbf{k}$.

2 Some technical details

One thus needs to determine the distinct symmetry classes of quasiperiodic spin density fields in order to establish the constraints imposed by stability on their Fourier expan-

sions and through (2) on the outcome of neutron diffraction experiments. This involves classifying the types of lattices, determining the distinct spin point groups compatible with those lattices, and finding all the inequivalent sets of phase functions satisfying the associated group compatibility conditions (9), as briefly outlined below.

2.1 Lattices and Bravais classes

The lattices of wave vectors which appear in the Fourier expansions of magnetic crystals are the same lattices that appear in the expansions of non-magnetic crystals. They are characterized by a *rank* — the smallest number of vectors required to generate the whole lattice — and a lattice point group G_L — the subgroup of $O(3)$ that leaves the lattice invariant. The point group G of the crystal is necessarily a subgroup of G_L. The classification of lattices into Bravais classes is identical to the non-magnetic case. For more details see, for example, Lifshitz [7].

2.2 The spin point group

The spin point group $G_{\mathbf{S}}$ is a subgroup of $G\times\Gamma$ with the property that every element of G and every element of Γ appears in at least one pair of $G_{\mathbf{S}}$. Such a subgroup is easily shown (Ref. [6]) to have the following structure: The set of point-group operations G_ϵ that are paired in $G_{\mathbf{S}}$ with the identity ϵ of Γ forms a normal subgroup of G; the set of spin rotations Γ_e paired with the identity e of G forms a normal subgroup of Γ. The pairs appearing in $G_{\mathbf{S}}$ associate all the elements of each coset of G_ϵ with all the elements of a single corresponding coset of Γ_e. This correspondence between cosets is an isomorphism between the two quotient groups G/G_ϵ and Γ/Γ_e.

The group compatibility condition (9) further implies that the subgroup Γ_e is abelian. If it contains more than just the identity or the identity and time inversion then it must be either *axial*, containing only primed or unprimed rotations about a single axis, or *orthorhombic*, containing primed or unprimed 2-fold rotations about three mutually orthogonal axes. In both cases it may also contain the time inversion.

Because Γ_e is also a normal subgroup of Γ, for every δ in Γ, $\gamma\in\Gamma_e$ implies $\delta\gamma\delta^{-1}\in\Gamma_e$. If Γ_e is axial then Γ may only include additional (primed or unprimed) rotations about the same axis, 2-fold rotations about orthogonal axes, and time inversion. If Γ_e is orthorhombic then additional operations in Γ may only permute the three 2-fold axes and are therefore restricted to operations in the proper cubic point group and time inversion.

Elements of the form (e,γ) describe spin rotations that leave the spin density field indistinguishable without requiring any rotation in physical space. In the special case of periodic crystals, these are spin rotations that when combined with a translation leave the magnetic crystal invariant. The phase functions $\Phi_e^\gamma(\mathbf{k})$ therefore contain the information which generalizes to the quasiperiodic case the so-called "spin translation groups" of Litvin and Opechowski.

2.3 The spin space group

The *spin space-group* classification is an organization of sets of phase functions satisfying the group compatibility condition (9) into equivalence classes according to two criteria:

(i) Two sets of phase functions Φ and Φ' that describe indistinguishable fields $\mathbf{S}$ and $\mathbf{S}'$, related by a gauge function χ, should clearly be associated with the same spin space group. Two such sets are related by a *gauge transformation*

$$\Phi'^{\gamma}_{g}(\mathbf{k}) \equiv \Phi^{\gamma}_{g}(\mathbf{k}) + \chi(g\mathbf{k} - \mathbf{k}), \tag{16}$$

and belong to the same *gauge-equivalence class*. Note that the constraints on the form of the Fourier coefficients $\mathbf{S}(\mathbf{k})$ involve only values of $\Phi^{\gamma}_{g}(\mathbf{k})$ for g's that leave $\mathbf{k}$ invariant and are therefore, as one may expect, gauge-invariant.

(ii) Two sets of phase functions Φ and Φ' may also be counted as *scale-equivalent* if there is a symmetry s of the lattice L, for which $G \rightarrow sGs^{-1}$ is an automorphism of G, and there is an automorphism σ of Γ, which together take one set into the other

$$\Phi'^{\gamma}_{g}(\mathbf{k}) = \Phi^{\sigma\gamma\sigma^{-1}}_{sgs^{-1}}(s\mathbf{k}). \tag{17}$$

The classes of phase functions under gauge and scale equivalence for a given spin point group and lattice correspond to the spin space group types in the periodic case, and constitute the extension of the classification scheme to the quasiperiodic case.

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