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Magnetic short-range order diffuse scattering in quasicrystals

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An analytical expression is derived for the short-range order (SRO) magnetic neutron diffuse scattering intensity in quasicrystals, and it is applied to a fictitious model of spin-orientation disorder in the Penrose pattern. The SRO diffuse scattering intensity depends on the overlapped volume of the occupation domains which are separated from each other by distances less than the correlation length and the SRO correlation functions. Analytical results for four different spin arrangements in the Penrose pattern are compared with numerical ones. The corresponding analytical and numerical results for all the cases are quite similar, suggesting the validity of the analytical expression.

1. Introduction

X-ray and neutron scattering experiments provide important information on the ordered and disordered states of solids. In particular, the latter are powerful for determining magnetic structures of crystals and quasicrystals. The scattered intensity is classified into Bragg reflection and diffuse scattering intensities. The former has information on the average ordered structure, while the latter gives information on the correlation between atoms in the disordered state. An expression for X-ray Bragg reflection intensities of quasicrystals has been derived on the basis of an n-dimensional (n = 5, 6) description of the structure of quasicrystals (Yamamoto, 1996) and its extension to nuclear neutron diffraction is straightforward. An n-dimensional description is also available to calculate short-range order (SRO) diffuse X-ray or neutron scattering intensity. In a previous paper (Yamamoto, 2010) the author has given an analytical formula for the X-ray SRO diffuse scattering intensity and applied it to the diffuse scattering of phason flips of the Penrose pattern (PP). The formula has been validated by comparing the analytical and numerical results. Its extension to neutron *nuclear* scattering is straightforward, since it is obtained from the formula for X-ray scattering by replacing the atomic scattering factors with the neutron scattering lengths. For magnetic neutron scattering, however, different expressions have to be used.

Quasicrystals are ordered solids without periodicity and show non-crystallographic point symmetries. Many quasicrystals contain rare-earth or transition-metal atoms with magnetic moments (Tsai, 1999; Tsai *et al.*, 1994; Luo *et al.*, 1993; Niikura *et al.*, 1994). Therefore, whether or not the magnetic properties of quasicrystals are different from those of periodic crystals is an interesting subject in solid-state physics. Quasicrystals usually have some structural disorder. Their average structures can be determined by the n-dimensional description, in which a quasicrystal structure is given by a three-dimensional intersection of an n-dimensional (n = 5 or 6) periodic structure (Yamamoto, 1996).

So far, magnetic structures with long-range order have not been observed in quasicrystals (Sato *et al.*, 1998). Therefore, we can observe neither magnetic Bragg reflections nor magnetic diffuse scattering caused by random phason or phonon disorder, since such diffuse scattering intensities are proportional to the Bragg reflection intensity (Jarić & Nelson, 1988; Lei *et al.*, 1999; Ishii, 2000). However, magnetic SRO diffuse scattering has been observed in an icosahedral Zn–Mg–Ho (i-Zn–Mg–Ho) quasicrystal and it shows icosahedral symmetry (Sato *et al.*, 2000). This gives information on the correlation between magnetic moments of different atoms so it is important for understanding magnetic properties in quasicrystals. No theory for analyzing such a SRO diffuse scattering has, however, yet been proposed.

Sato *et al.* (2000) have performed single-crystal neutron scattering experiments and observed a complicated magnetic diffuse scattering intensity distribution from Ho electron spins in i-Zn–Mg–Ho quasicrystals below the spin-glass transition temperature. The intensity distribution has been analyzed based on a six-dimensional model and the model could reproduce the observed patterns well. The analysis assumes a magnetic modulation with a single wavevector with an ∼11 Å period, which is parallel to a twofold axis of an icosahedral lattice. In addition, the correlation length of this magnetic modulated structure is determined to be ∼10 Å. Such a modulation breaks icosahedral symmetry. Therefore, at least 15 magnetic domains with different orientations should exist to recover the average icosahedral symmetry, provided that the magnetic domain has the point symmetry $2m'm'$. Noting that the correlation length is nearly equal to the wavelength of the modulation wave, it is a highly dampened wave.

The existence of such a wavy magnetic modulated structures is, however, not a necessary condition for icosahedral diffuse scattering. It is conceivable that a magnetic cluster

leads to a diffuse scattering intensity distribution with icosahedral symmetry. In this case, if an atom has a magnetic moment directed along some direction, many other atoms related to it by symmetry operations should exist, since the site symmetry compatible with the magnetic moment is low.

In this paper, an analytical formula for magnetic SRO diffuse scattering intensity is derived and the derived formula is applied to the magnetic moment arrangement in the PP, in which the magnetic moments are located at the phason flip sites in the PP (Yamamoto, 2010). This simple model can simulate the magnetic disorder caused by a phason flip, although such a magnetic spin arrangement is not realistic. When magnetic atoms form a cluster and within the cluster the correlation is complete, we can simplify the intensity formula and easily include medium-range order correlations in the calculation. Since most quasicrystals are considered to consist of some kind of cluster, this treatment may be efficient for the analysis of magnetic diffuse scattering in many quasicrystals.

In the next section, a general formula for SRO diffuse scattering intensity is derived. In §3 a simplified formula is given for the case where the magnetic structure consists of similar magnetic (spin) clusters. Its simplest case without intercluster correlation is discussed in §4. As an example of this case, it is finally applied to four different spin arrangements at the phason flip sites of the PP and the analytical and the corresponding numerical results are compared.

2. Scattering intensity

The scattering cross sections of non-spin-flip (↑↑) and spin-flip (↑↓) scattering are related to the pair-correlation functions, $\langle M^{\alpha}(\mathbf{x}^{\rm e})M^{\beta}(\mathbf{x}^{\rm e\prime})\rangle$, of the magnetic moment densities at the points $\mathbf{x}^{\rm e}$ and $\mathbf{x}^{\rm e\prime}$, where $\alpha, \beta = x, y, z$ and the superscript e indicates vectors in external space. They are (Sato *et al.*, 2000)

$$\left(\frac{{\rm d}\sigma}{{\rm d}\Omega}\right)_{\uparrow\uparrow} = \xi \sum_{\alpha\beta} (e_s^{\alpha} e_s^{\beta}) \left\langle M^{\alpha}(\mathbf{q}) M^{\beta}(-\mathbf{q}) \right\rangle \qquad (1)$$

and

$$\left(\frac{{\rm d}\sigma}{{\rm d}\Omega}\right)_{\uparrow\downarrow} = \xi \sum_{\alpha\beta} [\delta_{\alpha,\beta} - (e_q^{\alpha} e_q^{\beta} + e_s^{\alpha} e_s^{\beta})] \times \left\langle M^{\alpha}(\mathbf{q}) M^{\beta}(-\mathbf{q}) \right\rangle, \qquad (2)$$

with $\xi = (\gamma r_0)^2 (g/2)^2$ and

$$\left\langle M^{\alpha}(\mathbf{q}) M^{\beta}(-\mathbf{q}) \right\rangle = \int_V {\rm d}\mathbf{x}^{\rm e} \int_V {\rm d}\mathbf{x}^{\rm e\prime} \exp[2\pi i \mathbf{q} \cdot (\mathbf{x}^{\rm e} - \mathbf{x}^{\rm e\prime})] \times \langle M^{\alpha}(\mathbf{x}^{\rm e}) M^{\beta}(\mathbf{x}^{\rm e\prime}) \rangle, \qquad (3)$$

where $\langle \cdots \rangle$ denotes the statistical average, V is the volume of a crystal, and γ, r_0 and g are the gyromagnetic ratio of a neutron, the classical electron radius and the Landé g factor, respectively. The normalized scattering vector, $\mathbf{q}/|\mathbf{q}|$, is denoted by $\mathbf{e}_q$, while $\mathbf{e}_s$ represents the up-spin direction of a neutron. The total cross section for the unpolarized neutron is given by the sum of (1) and (2),

$$\frac{{\rm d}\sigma}{{\rm d}\Omega} = \xi \sum_{\alpha\beta} [\delta_{\alpha,\beta} - (e_q^{\alpha} e_q^{\beta})] I^{\alpha\beta}(\mathbf{q}), \qquad (4)$$

where $I^{\alpha\beta}(\mathbf{q}) \equiv \langle M^{\alpha}(\mathbf{q}) M^{\beta}(-\mathbf{q}) \rangle$. This formula includes the magnetic Bragg reflection intensity in general. Thus, if we know the Fourier transformation of the magnetic spin-density correlation functions, we can calculate the magnetic scattering intensity. The correlation function $I^{\alpha\beta}(\mathbf{q})$ can be split into two parts: $I_0^{\alpha\beta}(\mathbf{q}) \equiv \langle M^{\alpha}(\mathbf{q}) \rangle \langle M^{\beta}(-\mathbf{q}) \rangle$ and $I_1^{\alpha\beta}(\mathbf{q}) \equiv \langle \Delta M^{\alpha}(\mathbf{q}) \Delta M^{\beta}(-\mathbf{q}) \rangle$, where $\Delta M^{\alpha}(\mathbf{q}) = M^{\alpha}(\mathbf{q}) - \langle M^{\alpha}(\mathbf{q}) \rangle$. The first and second parts contribute to the Bragg and diffuse scattering intensities, respectively.

The calculation of the Fourier integral for the magnetic moment density [see (3)] is, however, not straightforward, since a quasicrystal structure is neither periodic nor random. According to the calculation based on the higher-dimensional description of quasicrystals (Yamamoto, 1996), the SRO diffuse scattering intensity can be expressed by the overlapped occupation domains (ODs) for the pair of atoms (see Appendix *A*)

$$I_1^{\alpha\beta}(\mathbf{q}) = \kappa \sum_{ijl} f f^* \langle \Delta\sigma_{i0}^{\alpha} \Delta\sigma_{jl}^{\beta} \rangle v_{ijl} \exp(2\pi i \mathbf{q} \cdot \Delta\mathbf{x}_{ijl}^{\rm e}), \qquad (5)$$

where $\kappa = V/\Omega_n$, V is the volume of the crystal and Ω_n is the unit-cell volume of the n-dimensional lattice; $f \equiv f(\mathbf{q})$ is the magnetic form factor; σ_{il}^{α} is the α component of the unit spin vector for the magnetic atom which is located at the site generated by the ith OD in the lth unit cell in n-dimensional space, and $\Delta\sigma_{il}^{\alpha}$ is the deviation from its average value: $\sigma_{il}^{\alpha} - \langle \sigma_i^{\alpha} \rangle$. $v_{ijl} \equiv v_{ij}(\Delta\mathbf{x}_{ijl}^{\rm i})$ is the overlapped volume (area) of ODs located at $\mathbf{x}_i$ in the unit cell at the origin and $\mathbf{x}_j$ in the unit cell at lattice vector $\mathbf{x}_l$: $\Delta\mathbf{x}_{ijl} \equiv \mathbf{x}_{i0} - \mathbf{x}_{jl}$. The latter symbol, $v_{ij}(\Delta\mathbf{x}_{ijl}^{\rm i})$, is used only when the vector $\Delta\mathbf{x}_{ijl}^{\rm i}$ has to be specified explicitly. (Superscripts e and i denote the external and internal space components of an n-dimensional vector.) v_{ijl} determines the frequency of pairs of magnetic atoms with a distance of $\Delta\mathbf{x}_{ijl}^{\rm e} \equiv \mathbf{x}_{i0}^{\rm e} - \mathbf{x}_{jl}^{\rm e}$ in external space. A higher-dimensional model (n-dimensional model) of a magnetic structure of quasicrystals is determined by the locations and shapes of ODs for magnetic atoms (see Yamamoto, 2010). Therefore, if we assume that the relevant correlation function is $\langle \Delta\sigma_{i0}^{\alpha} \Delta\sigma_{jl}^{\beta} \rangle$, the diffuse scattering intensity can be calculated based on a higher-dimensional model.

To analyze magnetic SRO diffuse scattering in quasicrystals, we need to give the correlation functions $\langle \Delta\sigma_{i0}^{\alpha} \Delta\sigma_{jl}^{\beta} \rangle$ in (5). In general, spin orientations of atoms are not collinear so the number of possible orientations may be larger than two. We denote a different spin orientation by μ, which runs from 1 to m. Like the case of atomic short-range order (Yamamoto, 2010), we introduce a statistical variable z_{il}^{μ}, where z_{il}^{μ} takes values 1 or 0 when the spin of the ith site at $\mathbf{x}_{il}$ is or is not in the μth orientation. Let the elements of the transformation matrix of the local Cartesian coordinates to the common coordinates be $(S_i^{\mu})^{\alpha\beta}$. The unit spin orientation vector is then represented by $(S^{\mu})^{\alpha z} z_{il}^{\mu}$. The correlation function $\langle \Delta\sigma_{i0}^{\mu\alpha} \Delta\sigma_{jl}^{\nu\beta} \rangle$ can be written as $(S^{\mu})^{\alpha z} (S^{\nu})^{\beta z} \langle \Delta z_{i0}^{\mu} \Delta z_{jl}^{\nu} \rangle$, where $\langle \Delta z_{i0}^{\mu} \Delta z_{jl}^{\nu} \rangle \equiv \langle z_{i0}^{\mu} z_{jl}^{\nu} \rangle - \langle z_{i0}^{\mu} \rangle \langle z_{jl}^{\nu} \rangle$. We denote $\langle \Delta z_{i0}^{\mu} \Delta z_{jl}^{\nu} \rangle$ as $g_{ijl} \equiv g_{ij}^{\mu\nu}(\Delta\mathbf{x}_{ijl}^{\rm e})$.

Using this correlation function and symmetry operator R in the point group, (5) can be rewritten as

$$I_1^{\alpha\beta}(\mathbf{q}) = \kappa \sum_{ijl} v_{ijl} a_{ijl} \sum_{\mu\nu R} Rf^{\mu\alpha} Rf^{*\nu\beta} \times g_{ijl}^{\mu\nu} \exp(2\pi i \mathbf{q} \cdot R\Delta\mathbf{x}_{ijl}^{\rm e}), \quad (6)$$

where $Rf^{\mu\alpha} \equiv f(S_i^{\mu}(R))^{\alpha z}$, i and j run over the independent OD pairs for the magnetic atoms in the unit cell which are located at $\mathbf{x}_{i0}$ and $\mathbf{x}_{jl}$ in n-dimensional space, and R runs over all the symmetry operators in the point group. $a_{ijl} \equiv a_{ij}(\Delta\mathbf{x}_{ijl})$ is the multiplicity of a vector $\Delta\mathbf{x}_{ijl}$, $(S_i^{\mu}(R))^{\alpha\beta} \equiv (S_i^{\mu} R)^{\alpha\beta}$ (Yamamoto, 2010). (Note that the matrix representation of the rotation operator R for the spin is different from that for the atom position by the determinant of the latter.) Therefore, from (4), the scattering cross section is given by

$$\frac{{\rm d}\rho}{{\rm d}\Omega} = \xi\kappa \sum_{ijl} v_{ijl} a_{ijl} \sum_{\mu\nu R} \left[(R\mathbf{f}^{\mu} \cdot R\mathbf{f}^{*\nu}) - (\mathbf{e}_q \cdot R\mathbf{f}^{\mu})(\mathbf{e}_q \cdot R\mathbf{f}^{*\nu}) \right] \times g_{ijl}^{\mu\nu} \exp(2\pi i \mathbf{q} \cdot R\Delta\mathbf{x}_{ijl}^{\rm e}), \quad (7)$$

where $R\mathbf{f}^{\mu}$ is the magnetic structure factor of the magnetic atom with a μ orientation, which is rotated by R.

The correlation functions fulfill $g_{ijl}^{\mu\mu} = -\sum_{\mu\neq\nu} g_{ijl}^{\mu\nu}$ (Hayakawa & Cohen, 1975). This leads to the final form of the SRO diffuse scattering cross section,

$$\frac{{\rm d}\rho}{{\rm d}\Omega} = -\xi\kappa \sum_{ijl} v_{ijl} a_{ijl} \times \sum_{\mu>\nu R} \left[|\Delta R\mathbf{f}^{\mu\nu}|^2 - |\mathbf{e}_q \cdot \Delta R\mathbf{f}^{\mu\nu}|^2 \right] \times g_{ijl}^{\mu\nu} \exp(2\pi i \mathbf{q} \cdot R\Delta\mathbf{x}_{ijl}^{\rm e}), \quad (8)$$

where $\Delta R\mathbf{f}^{\mu\nu} = R\mathbf{f}^{\mu} - R\mathbf{f}^{\nu}$ (see Appendix *B*).

3. Diffuse scattering from magnetic atom clusters

We consider the case where the correlation between the spins of atoms in a cluster is complete and atoms are not shared between different clusters. In such a case, $I_1^{\alpha\beta}(\mathbf{q})$ can be expressed by the magnetic structure factor of the magnetic atom cluster as shown below. In (6) the summation with respect to i, j, l and R can be replaced by the summation between all atomic pairs in a cluster and summation over the cluster centers. Therefore, (6) can be calculated by introducing the vector from the cluster center, $\Delta\mathbf{x}_{ikl}^{\rm e} = \Delta\mathbf{x}_{in}^{\rm e} + \Delta\mathbf{x}_{jn'}^{\rm e} + \Delta\mathbf{x}_{nn'l}^{\rm e}$, where the vectors $\Delta\mathbf{x}_{in}^{\rm e}$ and $\Delta\mathbf{x}_{jn'}^{\rm e}$ are the magnetic atom positions measured from the n and n' cluster centers $\mathbf{x}_n$ and $\mathbf{x}_{n'}$ in the unit cell, while $\Delta\mathbf{x}_{nn'l} \equiv \mathbf{x}_n - \mathbf{x}_{n'} - \mathbf{x}_l$. We denote the kind of cluster and their orientations by a single letter μ. Then the correlation function of each atomic pair $g_{ijl}^{\mu\nu}$ is given by the correlation function of the cluster center $g_{nn'l}^{\mu\nu}$, provided that atoms located at $\mathbf{x}_{i0}$ and $\mathbf{x}_{jl}$ belong to the n and n'th clusters.

In terms of the correlation functions for the cluster centers, (7) is rewritten as (see Appendix *C*)

$$\frac{{\rm d}\rho}{{\rm d}\Omega} = \xi\kappa \sum_{nn'l} v_{nn'l} a_{nn'l} \times \sum_{\mu\nu R} \left[(R\mathbf{F}^{\mu} \cdot R\mathbf{F}^{*\nu}) - (\mathbf{e}_q \cdot R\mathbf{F}^{\mu})(\mathbf{e}_q \cdot R\mathbf{F}^{*\nu}) \right] \times g_{nn'l}^{\mu\nu} \exp(2\pi i \mathbf{q} \cdot R\Delta\mathbf{x}_{nn'l}^{\rm e}). \quad (9)$$

where n, n' and l run over the independent OD pairs for the magnetic cluster centers located at $\mathbf{x}_n$ and $\mathbf{x}_{n'}$ in the unit cell and the lattice vector $\mathbf{x}_l$. $a_{nn'l} \equiv a_{nn'}(\Delta\mathbf{x}_{nn'l}^{\rm i})$ is the multiplicity of the nth and n'th cluster pair, $v_{nn'l} \equiv v_{nn'}(\Delta\mathbf{x}_{nn'l}^{\rm i})$ is the overlapped area of the cluster center ODs located at $\mathbf{x}_{n0}$ and $\mathbf{x}_{n'l}$, and $R\mathbf{F}^{\mu}$ is the structure factor for the cluster with μth orientation rotated by R, the α component of which is given by

$$(R\mathbf{F}^{\mu})^{\alpha} = \sum_i f(S^{\mu}(R))^{\alpha z} \exp(2\pi i \mathbf{q} \cdot R\Delta\mathbf{x}_{in}^{\rm e}), \quad (10)$$

where i runs over all the atoms in a cluster.

In the derivation of this expression, we used the fact that the overlapped area v_{ijl} is the same as $v_{nn'l}$ since each magnetic atom is assumed to be not shared by two clusters.

Similar to $g_{ijl}^{\mu\nu}$, the correlation functions $g_{nn'l}^{\mu\nu}$ fulfill $g_{nn'l}^{\mu\mu} = -\sum_{\mu\neq\nu} g_{nn'l}^{\mu\nu}$. Therefore, (9) is rewritten as

$$\frac{{\rm d}\rho}{{\rm d}\Omega} = -\xi\kappa \sum_{nn'l} v_{nn'l} a_{nn'l} \times \sum_{\mu>\nu R} \left[|\Delta R\mathbf{F}^{\mu\nu}|^2 - |\mathbf{e}_q \cdot \Delta R\mathbf{F}^{\mu\nu}|^2 \right] \times g_{nn'l}^{\mu\nu} \exp(2\pi i \mathbf{q} \cdot R\Delta\mathbf{x}_{nn'}^{\rm e}), \quad (11)$$

where $\Delta R\mathbf{F}^{\mu\nu} = R\mathbf{F}^{\mu} - R\mathbf{F}^{\nu}$. This shows that when the correlation between constituent atoms in all the clusters is complete, the diffuse scattering intensity is given by the correlation function of the cluster centers and the magnetic structure factors of the clusters. This expression is simplified when the inter-cluster correlations are negligibly small, as shown in the next section.

4. Magnetic clusters without inter-cluster correlations

In the case where the correlations between clusters are negligibly small, the diffuse scattering intensity calculation is reduced to the calculation of the structure factor of the magnetic atom cluster. In such a case, only the correlation functions with $\Delta\mathbf{x}_{nn'l}^{\rm e} = \mathbf{0}$, that is $n = n'$ and $\mathbf{x}_l = \mathbf{0}$, are non-zero so that the magnetic scattering intensity is given by

$$\frac{{\rm d}\rho}{{\rm d}\Omega} = \xi\kappa \sum_n v_{nn}(\mathbf{0}) \times \left[\langle |\Delta\mathbf{F}^{\mu}|^2 \rangle - \langle |\mathbf{e}_q \cdot \Delta\mathbf{F}^{\mu}|^2 \rangle \right], \quad (12)$$

where $\Delta\mathbf{F}^{\mu} \equiv \mathbf{F}^{\mu} - \langle \mathbf{F} \rangle$ (see Appendix *D*). Note that $\langle X^{\mu} \rangle \equiv \sum_{\mu} X^{\mu} \langle z_n^{\mu} \rangle$ denotes the average over all the orientations of X^{μ}. It is remarkable that (12) is independent of the location of the cluster center, so that it is determined by the structure factor of magnetic clusters and the number of cluster centers. [Note that $\kappa v_{nn}(\mathbf{0})$ gives the number of nth cluster centers.] Since quasicrystals can be considered to consist of some kind of cluster (usually with high symmetry), if magnetic atoms are

located at some shell of the cluster, this simple formula may be applicable to the magnetic diffuse scattering in such cases.

5. Magnetic diffuse scattering due to phason flips of the Penrose patterns

The magnetic scattering intensity of a given spin arrangement in external space can be directly calculated from the magnetic structure factor of a given magnetic moment distribution by calculating the Fourier transformation of its magnetic moment and by taking an ensemble average [see (3) and (4)]. In numerical calculations of the diffuse scattering intensity, however, we need to use the deviation of the spin magnetic moment from its average value in order to remove the contributions of Bragg reflections, as mentioned in Yamamoto (2010). In this section, numerical results for several spin arrangements are compared with their analytical results to confirm the validity of the derived expression. For simplicity the $|\mathbf{q}|$ dependence of the magnetic form factor $f^{\alpha}(\mathbf{q})$ is neglected.

We employ a simple magnetic atom arrangement, where the magnetic atom is located at the vertices with three outgoing edges in the PP and their phason flip sites are generated by the ODs 5 and 6 in Fig. 1 (since the phason flip sites are generated only by ODs 5 and 6 and sites generated by ODs 3 and 5 are included in the PP). For convenience, an OD which is related to OD i by inversion is denoted as OD i'. As is well known, the PP is given by four pentagonal ODs. They are composed of ODs 1 and 3, and 2 and 4 shown in Fig. 1, and another two given by ODs 1′–4′ (see Yamamoto, 2010). The symmetry of the PP is specified by a four-dimensional space group. We represent it by a layer-group symbol $p10/mmm(10^{7}1mm)$ (Kopský & Litvin, 2006) instead of using a five-dimensional space-group symbol for real decagonal quasicrystals (Janssen *et al.*, 2002).

The phason flip sites are located in two kinds of hexagons in the framework (which are generated by ODs 1, 2, 1′ and 2′; see Fig. 2). Blue points are the vertices of the PP generated by ODs 3, 4, 3′ and 4′. Their phason flip sites in the same hexagon are generated by ODs 5′ 6′, 5 and 6, respectively. Their spin direction depends on the magnetic layer group used. In order to introduce disorder in the structure, we assume that one of these two sites in the hexagon is randomly occupied, while the spin orientation is fixed for each site. Depending on the spin orientation used for each site, a different diffuse scattering intensity is obtained as shown below. Since we assume that the sites generated by ODs 1, 2, 1′ and 2′ are fully occupied by the same magnetic atoms and its spin magnetic moment is completely ordered, any spin arrangement for such sites contributes only to the Bragg intensity so that these spins are ignored in the following.

We consider six models with different spin arrangements, which can be classified by magnetic layer groups. To specify the spin direction of atoms which are generated by ODs 3–6, we use a six-coordinate representation of a five-dimensional vector for convenience, although one of the six basis vectors shown below is redundant. Five-dimensional vectors are represented by coordinates with respect to the basis vectors $\mathbf{e}_j$ ($j \leq 6$) with

$$\mathbf{e}_j = \frac{2a}{5^{1/2}}[c_j\mathbf{a}_1 + s_j\mathbf{a}_2 + c_{2j}\mathbf{a}_4 + s_{2j}\mathbf{a}_5] \ (j \leq 5) \qquad (13)$$

$$\mathbf{e}_6 = \mathbf{a}_3, \qquad (14)$$

where $c_j = \cos(2\pi j/5)$ and $s_j = \sin(2\pi j/5)$, and $\mathbf{a}_1$, $\mathbf{a}_2$ and $\mathbf{a}_3$ are the unit vectors of the external space, while $\mathbf{a}_4$ and $\mathbf{a}_5$ are those of the internal space. We write a five-dimensional vector $\mathbf{s}$ in terms of coordinates with respect to the basis vectors $\mathbf{e}_j$:

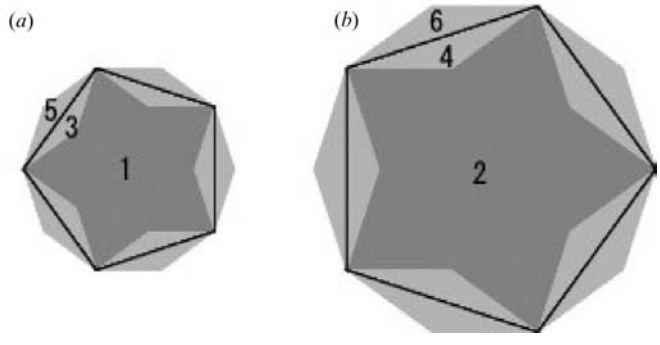

Figure 1
Independent occupation domains for the Penrose pattern and its phason flip sites, located at (*a*) (1, 1, 1, 1)/5 and (*b*) (2, 2, 2, 2)/5 in the unit cell of a four-dimensional decagonal lattice (Yamamoto, 1996, 2010). The occupation domains 3, 4, 5 and 6 and their inverted ODs located at (*a*) −(1, 1, 1, 1)/5 and (*b*) −(2, 2, 2, 2)/5 generate the positions flipped by the random phason into each other.

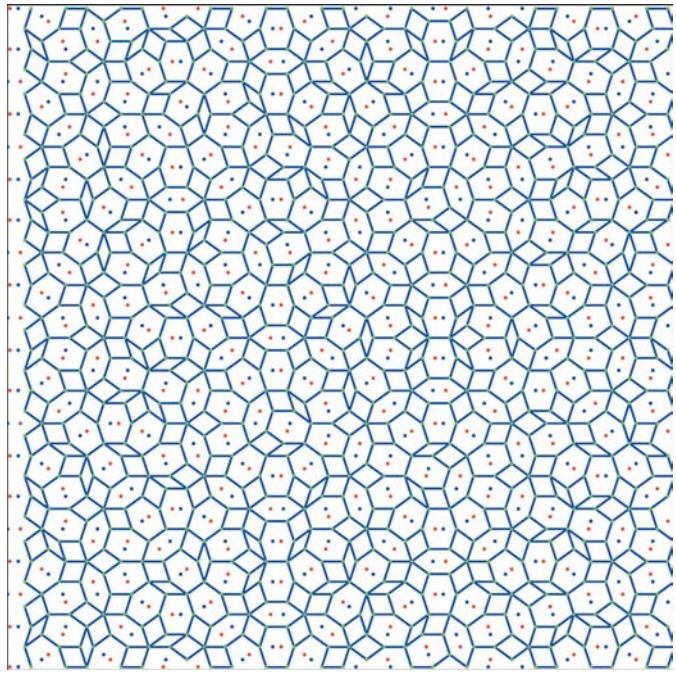

Figure 2
Phason flip sites in the Penrose pattern. The atom positions flipped by the random phason into each other are shown in hexagons.

$(s_1, s_2, s_3, s_4, s_5, s_6) = \sum_{j=1}^{6} s_j \mathbf{e}_j$ (Yamamoto, 1996). Note that $(1, 1, 1, 1, 1, 0)$ equals the zero vector, because $\sum_{j=1}^{5} \mathbf{e}_j = \mathbf{0}$.

The spin arrangements of the six models are shown in Table 1. In the first two models all the spins are in the plane normal to the tenfold axis. (The tenfold axis is parallel to $\mathbf{e}_6$.) Their (average-structure) symmetries are however different. In model (I) there is the inversion $\{I|0\}$, while in model (II) the antisymmetric inversion $\{I|0\}'$ exists in contrast. Note that the latter flips the spin direction, while the former does not change the spin direction. In model (I) the spin of an atom denoted by a blue point in Fig. 2, which is generated by the ODs 3 and 3′ (or 4 and 4′), is directed to the center of the hexagon. Thus in models (I) and (II) the spin direction of the phason flip sites (denoted by red points) is parallel and antiparallel to that of the original site. The spin arrangement in the two hexagons for model (II) is shown in Fig. 3. As mentioned above, the difference between a real magnetic moment and its average contributes to the diffuse scattering intensity, so that the difference of the magnetic moments for these sites is, in contrast, antiparallel and parallel. Owing to the symmetry of the PP, the spins on the site generated by equivalent ODs are rotated by $2j\pi/5$ when the ODs are obtained from independent ODs by $4j\pi/5$.

Models (III) and (IV) have the spin directions parallel and antiparallel to the tenfold axis, respectively, for all the sites generated by ODs 3, 4, 5 and 6. In the former the existence of $\{I|0\}$ is assumed, while in the latter, $\{I|0\}'$ instead. Thus, the spins in the hexagon are parallel and antiparallel in models (III) and (IV), and again their difference from the average spins are antiparallel and parallel.

The layer group allowing such a spin configuration is different in different models. In all the models, the (positional) site symmetry of ODs 3 (5) and 4 (6) is $2mm(mm1)$. Note that the normal mirror m and antisymmetric mirror m' allow the spins which are normal and parallel to the mirror plane, respectively. Thus, in models (I) and (II) the site symmetry of the spin arrangement in ODs 3–6 should be $m'2m'(m'm1)$, while in models (III) and (IV), it is $m'2'm(m'm'1)$. As mentioned above, in models (I) and (III) the inversion symmetry is assumed, while in models (II) and (IV) antisymmetric inversion is required. Therefore, models (I) and (II) have the layer groups $p10'/m'm'm(10^71mm)$ and $p10/m'm'm'(10^71mm)$, while those of models (III) and (IV) are $p10/mm'm'(10^71mm)$ and $p10/mm'm(10^71mm)$. (The time-reversal operation inverts the spin direction and it is defined in the external space, so that the prime denoting the time-reversal operation combined with another operation is used only in the external space parts.)

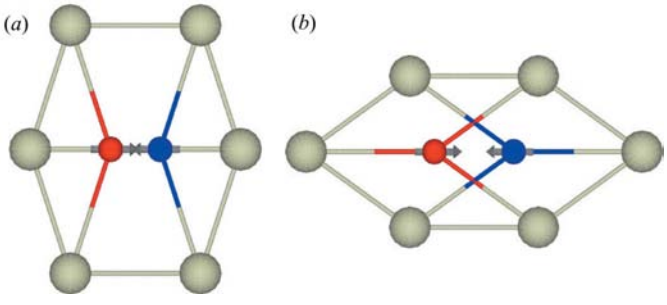

Figure 3
Spin arrangements in the phason flip sites in two hexagons (*a*) and (*b*) of the Penrose pattern. The arrows show the spin orientation in model (II) (drawn by *VESTA*; Momma & Izumi, 2008) when the sites are occupied. In model (IV) the arrows are replaced by the up and down arrows normal to the plane of the hexagon.

Table 1
Spin orientations for models (I) to (VI).

The models are defined by the unit vectors $\mathbf{s}_j^e/|\mathbf{s}_j^e|$ ($j = 1, 2, 3$). The vectors $\mathbf{s}_j^e$ are the external space components of vectors expressed by a six-coordinate representation of a five-dimensional vector with respect to the basis $\mathbf{e}_j$ ($j \leq 6$), for example $\mathbf{s}_1 = (0, 0, 0, 0, 1, 0)$, $\mathbf{s}_2 = (0, 1, 0, 0, 1, 0)$, $\mathbf{s}_3 = (0, 0, 0, 0, 0, 1)$. Note that the sixth coordinate represents the component parallel to the tenfold axis, so that spins in models (I) and (II) are on the plane normal to the tenfold axis and those in models (III)–(VI) are parallel or antiparallel to the tenfold axis.

	Model					
OD	(I)	(II)	(III)	(IV)	(V)	(VI)
3	$\mathbf{s}_1$	$\mathbf{s}_1$	$\mathbf{s}_3$	$\mathbf{s}_3$	$\mathbf{s}_3$	$\mathbf{s}_3$
5′	$\mathbf{s}_1$	$-\mathbf{s}_1$	$\mathbf{s}_3$	$-\mathbf{s}_3$	$\mathbf{s}_3$	$-\mathbf{s}_3$
4	$\mathbf{s}_2$	$\mathbf{s}_2$	$\mathbf{s}_3$	$\mathbf{s}_3$	$\mathbf{s}_3$	$\mathbf{s}_3$
6′	$\mathbf{s}_2$	$-\mathbf{s}_2$	$\mathbf{s}_3$	$-\mathbf{s}_3$	$\mathbf{s}_3$	$-\mathbf{s}_3$
3′	$\mathbf{s}_1$	$-\mathbf{s}_1$	$\mathbf{s}_3$	$-\mathbf{s}_3$	$-\mathbf{s}_3$	$\mathbf{s}_3$
5	$\mathbf{s}_1$	$\mathbf{s}_1$	$\mathbf{s}_3$	$\mathbf{s}_3$	$-\mathbf{s}_3$	$-\mathbf{s}_3$
4′	$\mathbf{s}_2$	$-\mathbf{s}_2$	$\mathbf{s}_3$	$-\mathbf{s}_3$	$-\mathbf{s}_3$	$\mathbf{s}_3$
6	$\mathbf{s}_2$	$\mathbf{s}_2$	$\mathbf{s}_3$	$\mathbf{s}_3$	$-\mathbf{s}_3$	$-\mathbf{s}_3$

If we regard the atoms in the hexagons in Fig. 2 as a cluster, we have two kinds of clusters which are accommodated in the two hexagons. Then the diffuse scattering intensity can be calculated by (12), since all the models have no correlation between these clusters. As mentioned in Yamamoto (2010), v_{44} is τ^2 times larger than v_{33}. Noting that

$$\Delta\mathbf{F}^{\mu} = \frac{\mathbf{s}_{\mu}}{|\mathbf{s}_{\mu}|}[1 \mp \exp(2\pi\mathbf{q}\cdot\Delta\mathbf{x}_{\mu})] \quad (\mu = 1, 2), \qquad (15)$$

with $\Delta\mathbf{x}_1 = (1, 1, 0, 1)$ and $\Delta\mathbf{x}_2 = (-1, 1, -1, 0)$, give the fluctuation in magnetic moment in models (I)/(II) and (III)/(VI) for the upper/lower sign, their diffuse scattering intensities shown in Fig. 4 are obtained for models (I) and (II). On the other hand, in models (III) and (IV) $\mathbf{s}_1/|\mathbf{s}_1|$ and $\mathbf{s}_2/|\mathbf{s}_2|$ are both replaced by $\mathbf{s}_3/|\mathbf{s}_3|$ in Table 1. This leads to Fig. 5. It should be noted that Fig. 4 is quite different from Fig. 5 since the magnetic structure factor depends strongly on the spin direction because of the existence of the second term in the square brackets in (12). In models (III) and (IV) the second term vanishes since the scattering vector $\mathbf{q}$ is normal to $\mathbf{s}_3$. Thus, the intensity of Fig. 5 only comes from the first term, which is independent of the spin direction. From (12) and (15), the diffuse scattering intensity can be expressed as the sum of a constant term (the so-called Laue monotonic diffuse scattering) and the deviations from it. The second term in (15) gives the same amplitude for models (I) and (II) for the same $\mathbf{q}$, while its sign is negative and positive, respectively. Such inverted intensity modulations are clearly seen in Figs. 4(*a*) and (*b*) or Figs. 5(*a*) and (*b*).

The numerical calculations of the diffuse scattering intensity are given in Fig. 6. The numerical calculations of one ensemble showed strong speckle noise, so that the figure is obtained after averaging intensities of 1000 ensembles. Although Fig. 6 still includes weak speckle noise, it is evident that the numerical calculations for these models give diffuse scattering intensities quite similar to the corresponding analytical ones, suggesting the validity of the analytical formula.

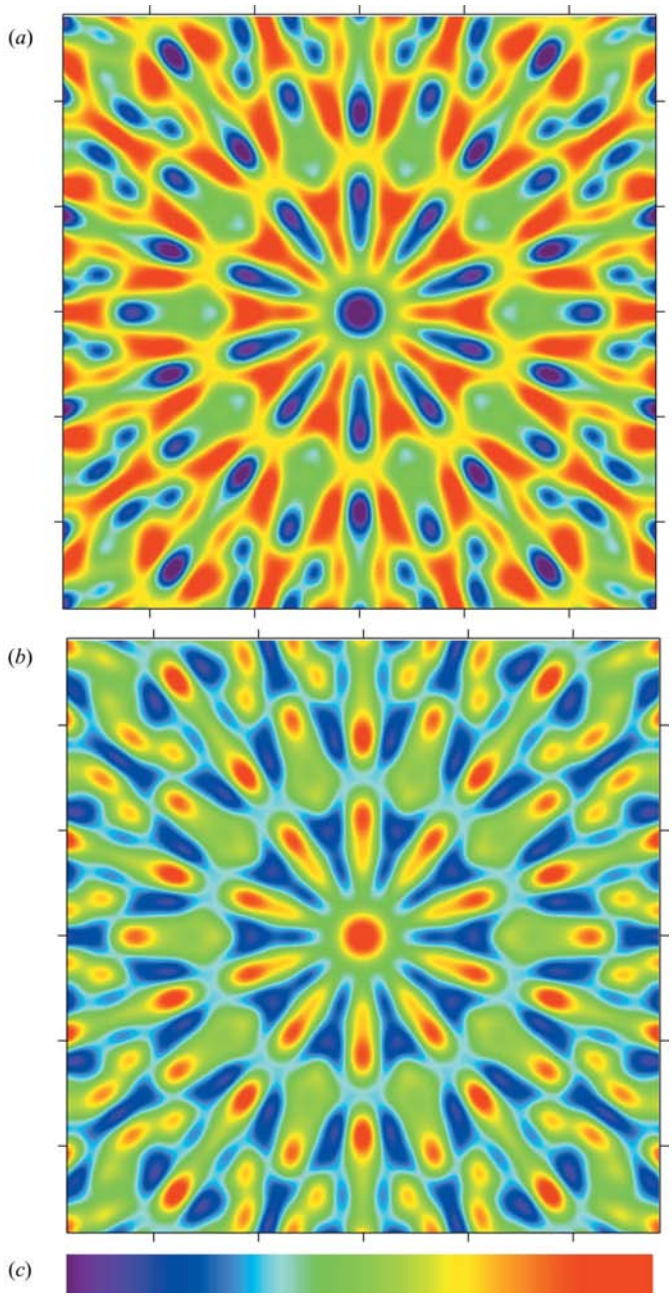

Figure 4
Diffuse scattering intensity of (*a*) model (I) and (*b*) model (II). The intensity is expressed by the rainbow colors shown in (*c*). The interval of the scale is $5a^*$ [$a^* = 2/(5^{1/2}a_r)$, where a_r is the edge length of the Penrose pattern shown in Fig. 2].

In Table 1, two other models, (V) and (VI), are given, in which the spins of ODs 3, 4, 5′ and 6′ are the same as in (III) and (IV), respectively, while those of ODs 3′, 4′, 5 and 6 are flipped. The magnetic structure factors of the clusters in models (V) and (VI), which are formed by the sites generated by ODs 3′ and 5 or 4′ and 6, change signs compared with those of the corresponding clusters in models (III) and (IV). Therefore, their Bragg reflection intensities are different. However, model (V) gives the same diffuse scattering intensity as that of model (III) and also model (VI) leads to the same pattern as in model (IV). This is because if the sign of the cluster structure factor is changed, the diffuse scattering intensity does not change whenever the inter-cluster correla-

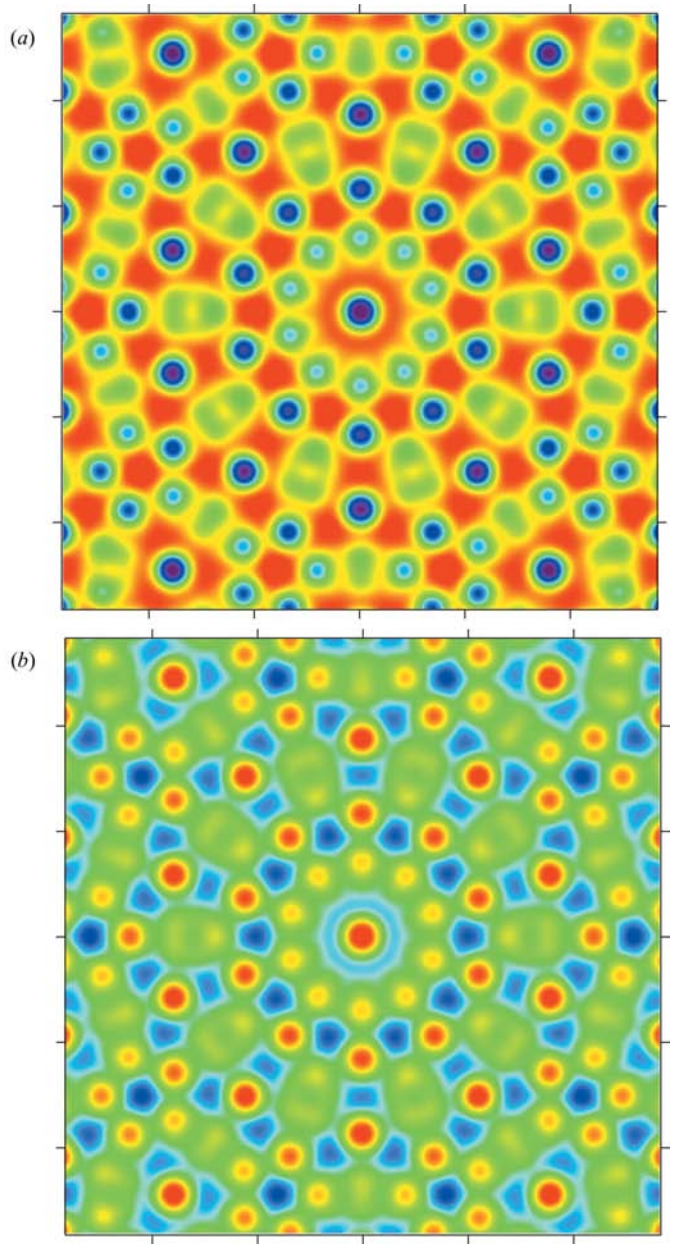

Figure 5
Diffuse scattering intensity of (*a*) model (III) and (*b*) model (IV). The scale interval is $5a^*$ as in Fig. 4.

tion is zero, as is clear from (12). Similarly, we can consider a model with different Bragg intensities but with the same diffuse scattering intensity for the case where the spin is on the plane normal to the tenfold axis.

6. Discussion

We consider in more detail the special case where all magnetic atoms are included in clusters and the correlation between the clusters is negligibly small. We compare (12) with the magnetic Bragg reflection intensity formula, which is given by (see Appendix *E*)

$$I(\mathbf{q}) = \xi\kappa\Big[\big|\mathbf{F}(\mathbf{q}')\big|^2 - \big|\mathbf{e}_q \cdot \mathbf{F}(\mathbf{q}')\big|^2\Big], \tag{16}$$

with the magnetic structure factor

$$\mathbf{F}(\mathbf{q}') = \sum_n a_n \sum_{\mu\{R|\tau\}} \langle z_n^\mu \rangle \mathbf{F}^\mu(\mathbf{q}) D_n(\mathbf{q}^{\rm i}) \times \exp[2\pi i \mathbf{q}' \cdot (R\mathbf{x}_n + \tau)], \tag{17}$$

where $\mathbf{q}'$ is a reciprocal-lattice vector of an n-dimensional lattice, the external and internal space components of which are $\mathbf{q}$ and $\mathbf{q}^{\rm i}$, a_n is the multiplicity of the nth cluster center, $\{R|\tau\}$ runs over the symmetry operations of an n-dimensional space group, while $D_n(\mathbf{q}^{\rm i})$ is the Fourier integral of the OD for the cluster center which is located at $\mathbf{x}_n$ in an n-dimensional unit cell (Yamamoto, 1996).

In both cases the intensity can be calculated by the structure factor of the magnetic atom clusters. An important difference is that (16) gives an integrated intensity in contrast to the scattering cross section in (12). Therefore, the peak height obtained from experiments does not correspond to (16) and it

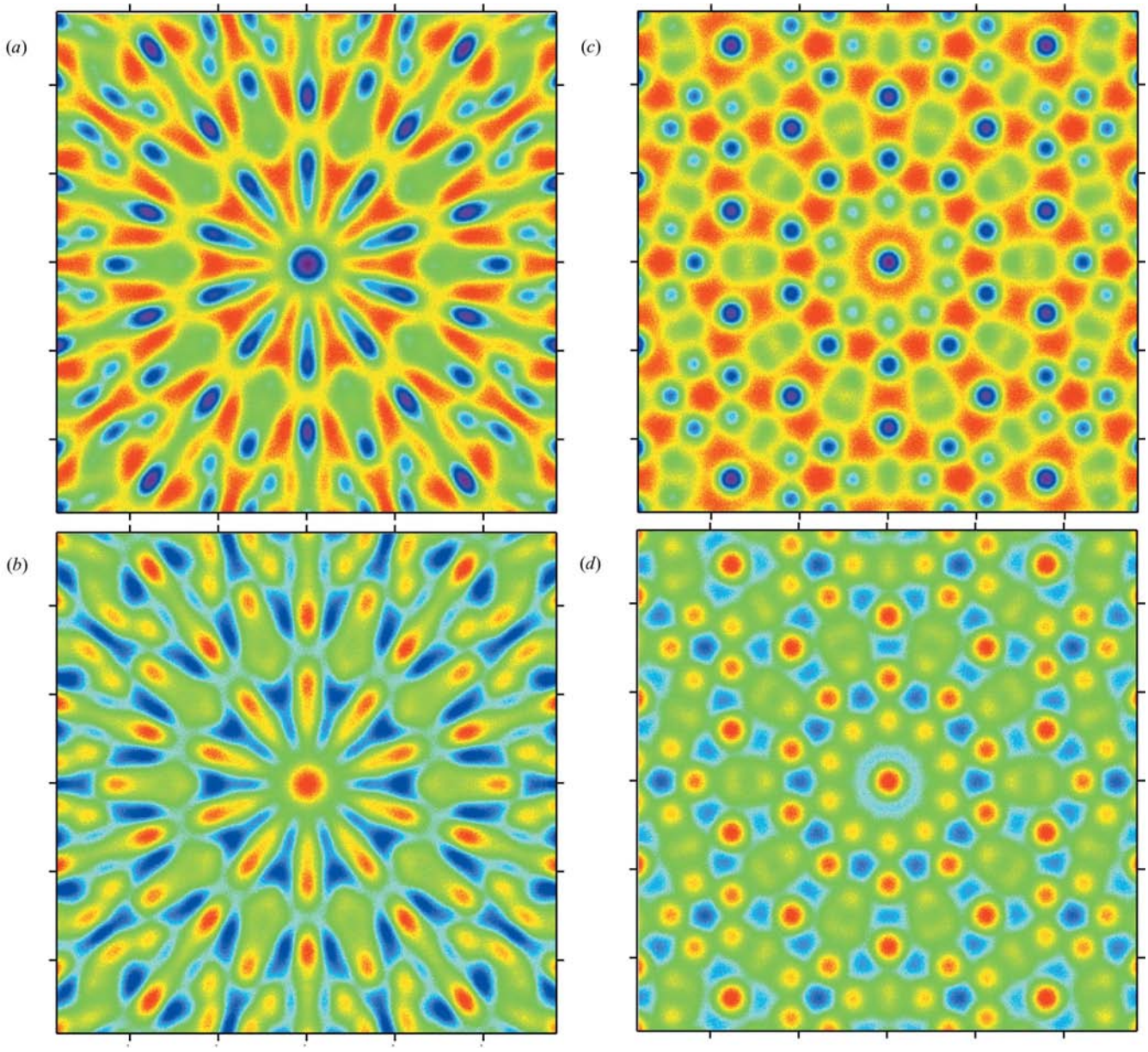

Figure 6
Diffuse scattering intensities calculated by numerical calculations for models (I)–(IV) in (*a*)–(*d*). The scale interval is $5a^*$ as in Fig. 4.

largely depends on the resolution of the equipment used even if the same sample is measured. The scattering vector $\mathbf{q}$ in (12) is not always a reciprocal-lattice vector, although the expression can give the diffuse scattering intensity at the positions of Bragg reflections. In an experiment, the measured intensity is always an integrated intensity within a small solid angle, which is determined by the slit width or pixel size of the detector. Note that the same scale factor applies to the integrated intensity in the solid angle which covers a peak in the Bragg reflection position. Therefore, if we know the scale factor $\xi\kappa$ from the Bragg reflection intensity, we can calculate the diffuse scattering intensity with the same scale.

The intensity formula for magnetic neutron diffuse scattering is similar in form to the corresponding formula for diffuse X-ray scattering. The main difference exists in the structure factors. In neutron magnetic scattering, an electron spin in a material scatters a neutron so that a scattering factor is given by a vector, while an X-ray is scattered owing to the charge of an electron and leads to a scalar structure factor. Owing to the vector properties, the scattering cross section of the neutron scattering depends on the direction of the scattering vector $\mathbf{q}$. In special cases, both X-ray and neutron scattering have a similar intensity distribution. In fact, it can be shown that the intensity distribution in Fig. 5 agrees with that of the X-ray diffuse scattering intensity shown in Figs. 4(*a*) and 5(*a*) in Yamamoto (2010). In this case $|\Delta\mathbf{F}^{\mu}|$ agrees with $|\Delta f^{\mu}|$ used in Yamamoto (2010) since no $|\mathbf{q}|$ dependences are assumed for the atomic structure factor and magnetic form factor.

The derived formulae, equations (7) or (9), are applicable to any case with inter-magnetic atom or inter-magnetic cluster correlations. However, when long-range correlations between them exist, we need to take into account the difference in atom or cluster environments up to far neighbors in the modeling of quasicrystals, since each small OD has to generate atoms or clusters with a similar environment within such a long distance. If we consider an environment up to a long distance, the number of different environments will increase compared with that of the nearest neighbor. This increases the number of independent ODs to be considered. Except for such a practical difficulty, the formulae are applicable to any cases in principle.

In icosahedral Zn–Mg–Ho (i-Zn–Mg–Ho) quasicrystals, a magnetic cluster model without inter-cluster interaction seems to be a good approximation. Then a simpler expression, such as (12), can be applied. The analysis of this case is, however, beyond the scope of this paper. Detailed analysis of the diffuse scattering in i-Zn–Mg–Ho (Sato *et al.*, 2000) will be described in a separate paper.

7. Summary

An analytical intensity formula for the SRO magnetic neutron diffuse scattering has been derived and it has been applied to the diffuse scattering of randomly distributed electron spins on the phason flip sites of the PP. The SRO magnetic diffuse scattering intensity in quasicrystals can be expressed by the overlapped volume of ODs in the higher-dimensional expression of quasicrystals and pair-correlation functions. To validate the derived formula, analytical results in the four examples with different spin orientations in the PP are compared with the results of numerical calculations. Agreement between analytical and numerical results confirms the validity of the formula. The SRO magnetic diffuse scattering intensity in quasicrystals strongly depends on the spin direction. This is demonstrated in the same examples.

APPENDIX *A*
Correlation function of magnetic moments

The Fourier integral in equation (3) can be calculated by using a higher-dimensional (n-dimensional) description of quasicrystals. Each atom position in the three-dimensional external space is given as an intersection of an OD, so that the magnetic moment density in the external space is given by

$$M^{\alpha}(\mathbf{x}^{\rm e}) = \sum_{l}\sum_{i} M_i^{\alpha}(\mathbf{x}^{\rm e} - \mathbf{x}_{il}^{\rm e}) D_i(-\mathbf{x}_{il}^{\rm i}), \tag{18}$$

where $M_i^{\alpha}(\mathbf{x}^{\rm e})$ is the magnetic moment density of the ith atom located at the origin, while $\mathbf{x}_{il}^{\rm e}$ and $\mathbf{x}_{il}^{\rm i}$ are the external and internal space components of the positional vector of the center of the ith OD in the lth unit cell of an n-dimensional lattice, $\mathbf{x}_i + \mathbf{x}_l$. The function $D_i(\mathbf{x}^{\rm i})$ is the occupation probability and takes a value of one when $\mathbf{x}^{\rm i}$ is within the ith OD located at the origin, otherwise it is zero. The sum over l runs over all the lattice points in the n-dimensional lattice and i runs over all the ODs in the unit cell. Owing to the existence of $D_i(-\mathbf{x}_{il}^{\rm i})$ in the above expression, only ODs near the three-dimensional external hyperplane give the atom position (see Fig. 1 in Yamamoto, 2010) Using this expression, (3) is rewritten as

$$\begin{aligned}
\langle M^{\alpha}(\mathbf{q}) M^{\beta}(-\mathbf{q})\rangle &\equiv I^{\alpha\beta}(\mathbf{q}) \\
&= \int_V {\rm d}\mathbf{x}^{\rm e} \int_V {\rm d}\mathbf{x}'^{\rm e} \sum_{lm}\sum_{ij} \exp[2\pi i \mathbf{q}\cdot(\mathbf{x}^{\rm e} - \mathbf{x}^{\rm e\prime})] \\
&\quad \times \langle M_i^{\alpha}(\mathbf{x}^{\rm e} - \mathbf{x}_{il}^{\rm e}) M_i^{\alpha}(\mathbf{x}'^{\rm e} - \mathbf{x}_{jm}^{\rm e})\rangle \\
&\quad \times D_i(-\mathbf{x}_{il}^{\rm i}) D_j(-\mathbf{x}_{jm}^{\rm i}) \\
&= \sum_{lm}\sum_{ij} ff \langle \sigma_{il}^{\alpha}\sigma_{jm}^{\beta}\rangle D_i(-\mathbf{x}_{il}^{\rm i}) D_j(-\mathbf{x}_{jm}^{\rm i}) \\
&\quad \times \exp(2\pi i \mathbf{q}\cdot\Delta\mathbf{x}_{iljm}^{\rm e})
\end{aligned} \tag{19}$$

where $f = f(\mathbf{q})$ is the magnetic form factor of atoms which occupy the ith OD in the lth unit cell of the n-dimensional lattice, while σ_{il}^{α} is the α component of the unit spin vector for that atom. This expression includes the Bragg reflection intensity in addition to the diffuse scattering intensity. The former and the latter come from the first and second terms of $\langle\sigma_{il}^{\alpha}\sigma_{jm}^{\beta}\rangle = \langle\sigma_i^{\alpha}\rangle\langle\sigma_j^{\beta}\rangle + \langle\Delta\sigma_{il}^{\alpha}\Delta\sigma_{jm}^{\beta}\rangle$, where $\Delta\sigma_{il}^{\alpha} = \sigma_{il}^{\alpha} - \langle\sigma_i^{\alpha}\rangle$. (We assume that $\langle\sigma_i^{\alpha}\rangle$ is independent of l). Therefore, $I_1^{\alpha\beta}(\mathbf{q}) \equiv \langle\Delta M^{\alpha}(\mathbf{q}) M^{\beta}(-\mathbf{q})\rangle$ is given by

$$\sum_{lm}\sum_{ij} ff\langle\Delta\sigma_{il}^{\alpha}\Delta\sigma_{jm}^{\beta}\rangle D_i(-\mathbf{x}_{jm}^{\rm i})D_j(-\mathbf{x}_{il}^{\rm i})\exp(2\pi i\mathbf{q}\cdot\Delta\mathbf{x}_{iljm}^{\rm e}) = \kappa\sum_{ijl} f_i f_j\langle\Delta\sigma_{i0}^{\alpha}\Delta\sigma_{jl}^{\beta}\rangle v_{ijl}\exp(2\pi i\mathbf{q}\cdot\Delta\mathbf{x}_{ijl}^{\rm e}). \quad (20)$$

where $v_{ijl}\equiv v_{ij}(\Delta\mathbf{x}_{ijl}^{\rm i})$ is the overlapped area (volume) of the ith and jth ODs located at $\mathbf{x}_i$ and $\mathbf{x}_{jl}$, respectively. Thus, we obtain equation (5).

APPENDIX *B*
Another expression for diffuse scattering intensity

In equation (7) we consider the parts

$$\sum_{\mu\nu}(R\mathbf{f}^{\mu}\cdot R\mathbf{f}^{*\nu})g_{ijl}^{\mu\nu} \quad (21)$$

and

$$\sum_{\mu\nu}(\mathbf{e}_q\cdot R\mathbf{f}^{\mu})(\mathbf{e}_q\cdot R\mathbf{f}^{*\nu})g_{ijl}^{\mu\nu} \quad (22)$$

separately. Since $g_{ijl}^{\mu\mu} = -\sum_{\nu\neq\mu} g_{ijl}^{\mu\nu}$ (Hayakawa & Cohen, 1975), equation (21) is rewritten as

$$\sum_{\mu\langle\rangle\nu} R\mathbf{f}^{\mu}\cdot(R\mathbf{f}^{*\nu}-R\mathbf{f}^{*\mu})g_{ijl}^{\mu\nu} = -\sum_{\mu>\nu}(\Delta R\mathbf{f}^{\mu\nu}\cdot\Delta R\mathbf{f}^{*\mu\nu})g_{ijl}^{\mu\nu}, \quad (23)$$

where $\sum_{\mu\langle\rangle\nu}$ is the summation with respect to μ and ν excluding $\mu=\nu$ and $\Delta R\mathbf{f}^{\mu\nu}\equiv R\mathbf{f}^{\mu}-R\mathbf{f}^{\nu}$. From similar considerations, equation (22) is rewritten as

$$-\sum_{\mu>\nu}(\mathbf{e}_q\cdot\Delta R\mathbf{f}^{\mu\nu})(\mathbf{e}_q\cdot\Delta R\mathbf{f}^{*\mu\nu})g_{ijl}^{\mu\nu}. \quad (24)$$

Using (23) and (24), (7) is rewritten as (8).

APPENDIX *C*
Diffuse scattering from magnetic clusters

By replacing $\langle\Delta\sigma_{i0}^{\mu\alpha}\Delta\sigma_{jl}^{\nu\beta}\rangle$ with $(S^{\mu})^{\alpha z}(S^{\nu})^{\beta z}\langle\Delta z_{i0}^{\mu}\Delta z_{jl}^{\nu}\rangle$, equation (5) is rewritten as

$$I_1^{\alpha\beta}(\mathbf{q}) = \kappa\sum_{ijl} v_{ijl}\sum_R ff^*(S^{\mu})^{\alpha z}(R)(S^{\nu})^{\beta z}(R) \times\langle\Delta z_{i0}\Delta z_{jl}\rangle\exp(2\pi i\mathbf{q}\cdot\Delta\mathbf{x}_{ijl}^{\rm e}), \quad (25)$$

where $(S^{\mu})^{\alpha\beta}(R)\equiv(S^{\mu}R')^{\alpha\beta}$. The matrix element of the rotation operator for the spin is different from that of the positional vector, since the former is an axial vector while the latter is a polar vector. To avoid the confusion, R' is used here for the rotation matrix for the spin.

As stated in the main text, the correlation functions of each atomic pair within a cluster, $\langle\Delta z_{i0}^{\mu}\Delta z_{jl}^{\nu}\rangle$, are equal to the correlation function of the cluster, $\langle\Delta z_{n0}^{\mu}\Delta z_{n'l}^{\nu}\rangle$, and the magnetic structure factor of a cluster is given by equation (10). This leads to equation (9).

APPENDIX *D*
Diffuse scattering for clusters without inter-cluster correlations

In the case where the correlations between clusters are zero, we have $\langle z_{n0}^{\mu}z_{n'l}^{\nu}\rangle = \langle z_n^{\mu}\rangle\delta_{nn'}\delta_{0l}\delta_{\mu\nu}$, where $\langle z_n^{\mu}\rangle$ is the probability with which the cluster at the nth site takes the μth orientation. Then the non-zero correlation function is given by $g_{nn}^{\mu\nu}(\mathbf{0}) = \langle\Delta z_n^{\mu}\Delta z_n^{\nu}\rangle = \langle z_n^{\mu}\rangle(\delta_{\mu\nu}-\langle z_n^{\nu}\rangle)$. In equation (11) we consider the parts $-\sum_{\mu>\nu}|\Delta\mathbf{F}^{\mu\nu}|^2 g_{nn}^{\mu\nu}(\mathbf{0})$ and $-\sum_{\mu>\nu}|\mathbf{e}_q\cdot\Delta\mathbf{F}^{\mu\nu}|^2 g_{nn}^{\mu\nu}(\mathbf{0})$ separately. The first part is given by

$$\begin{aligned}\sum_{\mu>\nu}|\mathbf{F}^{\mu}-\mathbf{F}^{\nu}|^2\langle z_n^{\mu}\rangle\langle z_n^{\nu}\rangle &= \frac{1}{2}\sum_{\mu,\nu}|\mathbf{F}^{\mu}-\mathbf{F}^{\nu}|^2\langle z_n^{\mu}\rangle\langle z_n^{\nu}\rangle\\ &= \frac{1}{2}\sum_{\mu,\nu}\left[|\mathbf{F}^{\mu}|^2-2Re(\mathbf{F}^{\mu}\mathbf{F}^{\nu*})+|\mathbf{F}^{\nu}|^2\right]\\ &\quad\times\langle z_n^{\mu}\rangle\langle z_n^{\nu}\rangle\\ &= \sum_{\mu}\left[|\mathbf{F}^{\mu}|^2-|\langle\mathbf{F}\rangle|^2\right]\langle z_n^{\mu}\rangle, \end{aligned} \quad (26)$$

where $\langle\mathbf{F}\rangle=\sum_{\mu}\mathbf{F}^{\mu}\langle z_n^{\mu}\rangle$. (Note that $\langle\mathbf{F}\rangle$ does not depend on μ, so that $\sum_{\mu}\langle\mathbf{F}\rangle\langle z_n^{\mu}\rangle=\langle\mathbf{F}\rangle$ since $\sum_{\mu}\langle z_n^{\mu}\rangle=1$.) This is equal to the average of the squared deviation of the structure factor of the μth orientation from the average structure factor,

$$\sum_{\mu}|\mathbf{F}^{\mu}-\langle\mathbf{F}\rangle|^2\langle z_n^{\mu}\rangle\equiv\langle|\Delta\mathbf{F}^{\mu}|^2\rangle, \quad (27)$$

where $\Delta\mathbf{F}^{\mu}=\mathbf{F}^{\mu}-\langle\mathbf{F}\rangle$, since this gives

$$\sum_{\mu}\left[|\mathbf{F}^{\mu}|^2-2Re\mathbf{F}^{\mu}\langle\mathbf{F}^*\rangle+|\langle\mathbf{F}\rangle|^2\right]\langle z_n^{\mu}\rangle. \quad (28)$$

Noting that $Re\sum_{\mu}\mathbf{F}^{\mu}\langle z_n^{\mu}\rangle\langle\mathbf{F}^*\rangle=|\langle\mathbf{F}\rangle|^2$, this leads to the last expression of (26).

The second part is obtained from (26) by replacing $\mathbf{F}^{\mu}$ and $\mathbf{F}^{\nu}$ with $\mathbf{e}_q\cdot\mathbf{F}^{\mu}$ and $\mathbf{e}_q\cdot\mathbf{F}^{\nu}$, so that it is given by

$$\langle|\mathbf{e}_q\cdot\mathbf{F}^{\mu}-\langle\mathbf{e}_q\cdot\mathbf{F}\rangle|^2\rangle=\langle|\mathbf{e}_q\cdot\Delta\mathbf{F}^{\mu}|^2\rangle. \quad (29)$$

Expressions (26) and (29) for the first and second parts in (11) lead to (12).

APPENDIX *E*
Bragg reflection intensity

The Bragg intensity is obtained from (4) by replacing $I^{\alpha\beta}(\mathbf{q})=\langle M^{\alpha}(\mathbf{q})M^{\beta}(-\mathbf{q})\rangle$ with $I_0^{\alpha\beta}(\mathbf{q})=\langle M^{\alpha}(\mathbf{q})\rangle\langle M^{\beta}(-\mathbf{q})\rangle$. From (18) the term contributing to the Bragg reflection intensity, $\langle M^{\alpha}(\mathbf{q})\rangle$, is given by

$$\langle M^{\alpha}(\mathbf{q})\rangle=\sum_{li} f\langle z_l^{\alpha}\rangle D_i(-\mathbf{x}_{il}^{\rm i})\exp(2\pi i\mathbf{q}\cdot\mathbf{x}_{il}^{\rm e}). \quad (30)$$

Using $D_i(\mathbf{x}_{il}^{\rm i})=\int d\mathbf{q}^{\rm i}\,D_i(\mathbf{q}^{\rm i})\exp(-2\pi i\mathbf{q}^{\rm i}\cdot\mathbf{x}_{il}^{\rm i})$ [or $D_i(\mathbf{q}^{\rm i})=(2\pi)^{-d/2}\int d\mathbf{x}^{\rm i}\,D_i(\mathbf{x}^{\rm i})\exp(2\pi i\mathbf{q}^{\rm i}\cdot\mathbf{x}_{il}^{\rm i})\quad(d=n-3)$], this is rewritten as

$$\langle M^{\alpha}(\mathbf{q})\rangle=\int d\mathbf{q}^{\rm i}\,L(\mathbf{q}')F^{\alpha}(\mathbf{q}'), \quad (31)$$

where $L(\mathbf{q}') = \sum_l \exp(2\pi i \mathbf{q}' \cdot \mathbf{x}_l)$ is the periodic delta function (the Laue function) in n-dimensional space and $F^\alpha(\mathbf{q}')$ is the α component of the magnetic structure factor. This is given by

$$F^\alpha(\mathbf{q}') = \sum_i f\langle z_i^\alpha \rangle D_i(\mathbf{q}^{\rm i}) \exp(2\pi i \mathbf{q}' \cdot \mathbf{x}_i), \tag{32}$$

with the n-dimensional vectors $\mathbf{q}' = \mathbf{q} + \mathbf{q}^{\rm i}$ and $\mathbf{x}_i = \mathbf{x}_i^{\rm e} + \mathbf{x}_i^{\rm i}$. In this expression i runs over all the ODs of the magnetic atoms in the n-dimensional unit cell. Using (31), (32) and (4) the integrated intensity of the Bragg reflection at the reciprocal-lattice point $\mathbf{q}$ is given by

$$I(\mathbf{q}) = \xi\kappa\Big[\big|\mathbf{F}(\mathbf{q}')\big|^2 - \big|\mathbf{e}_q \cdot \mathbf{F}(\mathbf{q}')\big|^2\Big]. \tag{33}$$

Equations (32) and (33) mean that the Bragg reflection intensity can be obtained by projection of the structure factor in n-dimensional space onto the external space. The summation over i in (32) can be replaced by the summation over the cluster centers and a summation for the ODs of the constituent atoms (cluster ODs). As stated in the text, the ODs for constituent atoms of a cluster (cluster ODs) are the same as that of the cluster center but they are shifted by $\Delta\mathbf{x}_{in}^{\rm e}$ from the OD of the cluster center in parallel to the external space. Then $\mathbf{x}_i$ is given by $\Delta\mathbf{x}_{in}^{\rm e} + \mathbf{x}_n$, where $\Delta\mathbf{x}_{in}^{\rm e}$ is a vector from the cluster center and $\mathbf{x}_n$ is the positional vector of the nth cluster center. Let the α component of the structure factor for the cluster ODs of the nth cluster be $F^\alpha(\mathbf{q})$. Then (32) is rewritten as

$$F^\alpha(\mathbf{q}') = \sum_n F^\alpha(\mathbf{q}) D_n(\mathbf{q}^{\rm i}) \exp(2\pi i \mathbf{q}' \cdot \mathbf{x}_n) \tag{34}$$

with the structure factor of the cluster

$$F^\alpha(\mathbf{q}) = \sum_i f\langle z_i^\alpha \rangle \exp(2\pi i \mathbf{q} \cdot \Delta\mathbf{x}_{in}^{\rm e}), \tag{35}$$

where i runs over the atoms forming the nth cluster and $D_n(\mathbf{q}^i)$ is the Fourier integral of the OD for the nth cluster center. Instead of considering all the clusters, the use of independent ones and symmetry operations and (34) and (35) lead to (16) and (17).

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