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Phases of Intense Peaks in IPIC Quasicrystals Models. Formal Patterson Analysis

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Abstract. We show that, within the context of Interpenetrating Pseudo-Icosahedral Clusters (IPIC) Models for i-quasicrystals, formal expressions of phases of intense peaks can be provided as quotients of formal series of several variables. We comment the roles played by the deformations of pseudo-icosahedral clusters, their chemical disorders on shells, the site multiplicities - diffractive graduated skeleton of sites - and their mutual interactions through characteristic quadratic, cubic, ... forms invariant under the action of the icosahedral group. The Patterson function is carried out formally with the IPIC intensity expressions as series and is associated to a geometrical iteration process associated to the atomic surfaces in the internal space.

1. Introduction

It is well-known that the Patterson function is the convolution of the electron density $\rho(r)$ with itself inverted in the origin $\rho(-r)$; and, for crystals, can be defined as the integral $P(u) = \int_{u.c.} \rho(x)\rho(-u+x)dx$ over the unit cell. It represents all the vectors $r_j - r_i$ between atoms (N^2 for N atoms in the unit cell) present in the electron density. It is always centrosymmetric, $P(u) = P(-u)$, since, for every interatomic vector $r_j - r_i$, there exists in the reverse direction the interatomic vector $r_i - r_j$. The symmetry of the Patterson function may therefore be different from that of the crystal. The Patterson function admits symmetry invariants the analysis of which allows the characterization of the structure. An easier way to view the Patterson function is to express it as the Fourier transform of the intensity distribution in reciprocal space. For this, phases are set to zero, and peaks are punctualized.

For icosahedral quasicrystals in the 3D Euclidean space E_n in $\mathbb{R}^6$, the situation is different. The set of interatomic vectors $\mathcal{IV} = \{r_j - r_i \mid i \neq j\}$ becomes infinite, discrete, invariant under inversion, without any translation vector leaving it invariant. No periodicity occurs, and the convolution $\rho(x) * \rho(-x)$ would be an integral over $\mathbb{R}^3$ and, therefore, has to be handled differently. It is by now one of the basic tools

to understand the 6D crystallography of a quasicrystal, the atomic surfaces [1, 2], whereas the 6D Bravais lattice class can be guessed by analyzing the peak positions in reciprocal space, and their extinctions according to the type $Pm\bar{3}5$, $Fm\bar{3}5$, or $Im\bar{3}5$, or else [3].

The present paper aims at showing that under IPIC conditions, that is when i-phases are formed by infinite collections of parallel (-orientational order-) pseudo-icosahedral clusters disposed quasiperiodically which can in some sense be averaged, IPIC models of i-phases [4] give formal expressions of the phases of intense peaks and of the intensity distribution in reciprocal space, and that the set $\mathcal{TV}$, i.e. the Patterson function, is in fact structured with respect to the icosahedral symmetry.

2. Phases in intense peaks from IPIC models

We use the same definitions as in [4] for an IPIC state of matter, where $CC = \{s_1, s_2, s_3, \dots\}$ is the set of cluster centers in E_n , and which represents, under a slightly axiomatic form, an i-phase. Centered at each s_i , which can be an occupied site or not, we find one cluster which is a deformation $\delta(X(n))$ of an average perfectly icosahedral (that is G -invariant, where $G = m\bar{3}5$ is the icosahedral group fixed in $O(3, \mathbb{R})$) colored cluster $X(n)$, n denoting the number of layers. We assume here, for simplicity's sake, that only one orientation of clusters exists, i.e. that the orientational order is unique ($J = 1$ in Verger-Gaugry [4]). Coloration is G -invariant, and uniform on each shell of $X(n)$. A shell is constituted by the vertices of an icosahedral polyhedron. Since the radii of the shells in a cluster increase, and that icosahedral clusters in "metallic" i-phases are compact, some hierarchical sequences of icosahedral polyhedra necessarily appears such as those proposed from E_8 by Sadoc and Mosseri [5].

The average cluster, corresponding to the infinite set of deformed clusters, may be very large, eventually infinite, and exhibit diffraction properties typical to the succession of averaged layers it contains. An IPIC model in some sense assumes that this averaging is possible in direct space, cluster by cluster, while an atomic surface is associated to the averaging of the perp components of equivalent atomic positions, atom by atom, in the internal space $E_\perp$. The layers of the average cluster are constituted by icosahedral polyhedra, for which the vertices are diffracting, with non-realistic scattering factors, including temperature-dependant terms, since they are obtained as averaged actual scattering factors from the actual sites. When the icosahedral symmetry extends over very long distances in clusters, compared to a typical distance separating two adjacent cluster centers, interpenetration occurs with a lot of atomic sites belonging simultaneously to several clusters, that is they are counted several times since viewed from several cluster centers at the same time. If the average cluster $X(n)$ is assumed infinite, $n = \infty$, some sites will be counted infinitely many times, while others will have a finite and small multiplicity. Those sites whose multiplicity is very large, since included in icosahedral shells, constitute a kind of diffractive skeleton for the icosahedral symmetry, and are responsible in the G -invariance of the diffraction patterns in reciprocal space. This set of sites

is graduated by the number of clusters to which a site belongs. The diffractive "weight" of the other sites becomes not significant in the diffraction process and those sites of low multiplicity can almost be eliminated from the modelling of the IPIC phase, since they almost do not contribute to the diffracting intensities, as far as the icosahedral symmetry is concerned. Their proportion is assumed small. These atoms are not the so-called "glue atoms" inbetween clusters. When modelling an icosahedral phase on the computer with a finite number of icosahedral clusters, it is easy to compute the sites with low and elevated diffractive weights. However, in infinite models, this computation gives under-estimated values, since the number of shells in pseudo-icosahedral clusters cannot be defined precisely, nor their types at large distances.

To allow the analysis of the various contributions: defects of coloration in icosahedral shells, site multiplicities, deformations, due to interpenetration, and their mutual interactions, we have developped a certain scattering function $H(q, 1, 1, 1)$ as a series [4]. The intensity $I(q)$ at a scattering vector q is given by:

$$(1) \quad I(q) = |H(q, 1, 1, 1)|^2$$

where, for $q \in E_n^* \equiv E_n$ the dual space of E_n and $0 \leq \gamma_i \leq 1$ ($i = 1, 2, 3$):

$$(2) \quad H(q, \gamma_1, \gamma_2, \gamma_3) = \sum_{s_k \in CC} \sum_{l=1}^n \sum_{x_j \in L_{lk}} \frac{K_{lk} + \gamma_1 z_j}{\Lambda + \gamma_2 \lambda_j} \exp[2i\pi q \cdot (x_j + \gamma_3 [\delta_k(x_j) - x_j])]$$

Λ is the average site multiplicity, δ_k is the deformation of the k th-cluster, z_j the atomic scattering factor excess for the j th atom lying in (resp. K_{lk} the average scattering factor relatively to) the l th icosahedral layer L_{lk} of the k th cluster, constituted by n layers, λ_j the site multiplicity excess for the atom j ($\equiv x_j$), x_j the current atomic site.

$H(q, 1, 1, 1)$ is equal to the infinite convergent Taylor series:

$$(3) \quad H(q, 1, 1, 1) = H(q, 0, 0, 0) + \sum_{i=1}^3 \frac{\partial H}{\partial \gamma_i} \Big|_{\gamma_i=0} + \sum_{i,j} \frac{\partial^2 H}{\partial \gamma_i \partial \gamma_j} \Big|_{\gamma_i=\gamma_j=0} + \dots$$

All the derivatives can easily be computed from Eq.(2). Convergence is ensured by the convergence of $\sum_{s_k \in CC} \exp[2i\pi q \cdot s_k]$, where we consider q as a certain g_n , and by the finite summation of $\int_V \exp[-2i\pi g_{\perp} \cdot y] dy$, over a G -invariant volume V in the internal space $E_{\perp}$, which may be not connected, nor smooth [4]. Since $I(q)$ is G -invariant and expressed by Eq.(1), we slightly modify the series given by Eq.(3) to obtain a still converging series constituted by G -invariant terms. In Verger-Gaugry [4], this geometrical renormalizing process was made up till the first order term. It can be seen as follows: we want $H(q, 1, 1, 1)$ G -invariant, that is $H(q, 1, 1, 1) = H(q, 1, 1, 1)^G$. We write:

$$H(q, 1, 1, 1) = H(q, 0, 0, 0)^G + (H(q, 0, 0, 0) - H(q, 0, 0, 0)^G) +$$

$$(4) \quad \sum_{i=1}^3 \frac{\partial H}{\partial \gamma_i} |_{\gamma_i=0} + \sum_{i,j} \frac{\partial^2 H}{\partial \gamma_i \partial \gamma_j} |_{\gamma_i=\gamma_j=0} + \dots$$

Now, $H(q, 0, 0, 0) - H(q, 0, 0, 0)^G$ is a first-order term, where $H(q, 0, 0, 0)^G$ is the closest G -invariant expression to $H(q, 0, 0, 0)$, and we group it with $\sum_{i=1}^3 \frac{\partial H}{\partial \gamma_i} |_{\gamma_i=0}$. Since $[(H(q, 0, 0, 0) - H(q, 0, 0, 0)^G) + \sum_{i=1}^3 \frac{\partial H}{\partial \gamma_i} |_{\gamma_i=0}] - [(H(q, 0, 0, 0) - H(q, 0, 0, 0)^G) + \sum_{i=1}^3 \frac{\partial H}{\partial \gamma_i} |_{\gamma_i=0}]^G$ is a second-order term, we group it with $\sum_{i,j} \frac{\partial^2 H}{\partial \gamma_i \partial \gamma_j} |_{\gamma_i=\gamma_j=0}$, and so on; thus, we obtain the development containing only G -invariant terms:

$$(5) \quad H(q, 1, 1, 1) = H(q, 0, 0, 0)^G + [(H(q, 0, 0, 0) - H(q, 0, 0, 0)^G) + \sum_{i=1}^3 \frac{\partial H}{\partial \gamma_i} |_{\gamma_i=0}]^G + \dots$$

It is clear that this process can be continued up till infinity, and amounts to renormalizing sequentially the symbolic powers of order $p \geq 0$ of H evaluated at 1 according to the main features of clusters, with respect to G . The closest G -invariant expressions can easily be found by averaging each time the suitable products and sums of variables in the expressions [4]. This approach is fundamental to obtain G -invariant phases of peaks in reciprocal space ($H(q, 1, 1, 1)$ is G -invariant, not only its module), which is expected experimentally. If we denote by $K_{av(l)}$ the average value of the K_{ik} s, k varying, for the l th layers, K the average atomic scattering factor of the i -phase, $\delta_{av(l)}$ (resp. $\lambda_{av(l)}$) the average deformation (resp. site multiplicity excess) restricted to the l th layers of all the clusters, we obtain the phases $\Phi(q)$ by separating real and imaginary terms in Eq.(5) and the contribution of the cluster centers CC from that of the average cluster (Eq.(13) and (14) in Verger-Gaugry [4]). We set $q = g_n$, and $\frac{1}{2}L_{l1}$ for all the sites of L_{l1} in a half-space passing through the center of the average cluster centered at s_1 . Let

$$A(g_n) = \sum_{i=1}^n [K_{av(l)} (1 - \frac{\lambda_{av(l)}}{\Lambda}) \sum_{\frac{1}{2}L_{l1}} \cos(2\pi g_n \cdot (x_j - s_1))]$$

$$B(g_n) = \sum_{i=1}^n [2\pi g_n \cdot (\delta_{av(l)}(x_j) - x_j) K_{av(l)} \sum_{\frac{1}{2}L_{l1}} \cos(2\pi g_n \cdot (x_j - s_1))]$$

$C(g_\perp) = \int_V \cos(2\pi g_\perp \cdot y) dy$ and $D(g_\perp) = -\int_V \sin(2\pi g_\perp \cdot y) dy$, for $g_\perp$ the corresponding vector to g_n . We obtain:

$$(6) \quad \Phi(g_n) = \text{Arctan} \left(\frac{A(g_n)D(g_\perp) + B(g_n)C(g_\perp)}{A(g_n)C(g_\perp) - B(g_n)D(g_\perp)} \right)$$

Summation is expressed here at the first order, and from Eq.(5) the exact expression of $\tan(\Phi(q))$ is a quotient of two formal G -invariant series, each of them viewed as a sum of G -invariant quadratic (for second-order terms), cubic (for third-order terms),

... forms. A lot of information on the QC structure is in the phases and is necessary for reconstructing it [6].

3. Intensities and Patterson analysis

The intensity $I(g_n)$ can be expressed as $|\frac{1}{\Lambda}(A(g_n) + iB(g_n))(C(g_\perp) + iD(g_\perp))|^2$ and the "trace" of the atomic surfaces appears through the Patterson function:

$$(7) \quad P(u) = \frac{1}{\Lambda^2} \sum_{g_n} (A(g_n)^2 + B(g_n)^2)(C(g_\perp)^2 + D(g_\perp)^2) \cos(2\pi g_n \cdot u)$$

where the term $A(g_n)^2 + B(g_n)^2$ outlines the influence of deformations (in A), of multiplicities (in B), and the rôle played by the succession of icosahedral polyhedra in layers (summations over $\frac{1}{2}L_{II}$ and l in A and B) on the determination of the atomic surfaces. The maxima of $P(u)$ are referred to a first-order truncation of Eq.(5). The atomic surfaces are therefore a limit, when taking into account all the terms of Eq.(5).

4. Conclusions

The analysis of the formal expressions of phases and Patterson function, under IPIC conditions, is fruitful to understand the rôles played by interpenetration of pseudo-icosahedral clusters.

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