Diffuse scattering in quasicrystals

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Defects in ideal quasicrystalline lattices associated with Goldstone phason modes are analyzed. The diffuse scattering which they cause is calculated. The relationship with a recent experiment by Denoyer *et al.* is discussed.

A diffuse scattering with a complicated k-space distribution was recently observed[1] in the quasicrystal Al_6CuLi_3. The compound Al_6CuLi_3 is probably a thermodynamic-equilibrium quasicrystal, since a structure with icosahedral symmetry is found at an arbitrarily small cooling rate (for the quasicrystals which have been discovered previously, this rate is very high). In this connection there is the question of a disorder in regular quasicrystalline lattices. In this letter we analyze defects which might lead to a scattering like that observed by Denoyer *et al.*[1] Since the arrangement of atoms in the compound Al_6CuLi_3 has not been precisely established, the arguments below are of basically a qualitative nature.

Several models have been proposed to describe the structure of quasicrystals.[2–4] In terms of their essential features these models are equivalent to each other. According to them, the positions of atoms are found by projecting entire points of a six-dimensional lattice which lie within a tube. The feature of this construction which is of importance to the present letter is the sharp edge of the tube, which leads to a special class of defects, as we will see below.

Let us assume that the interaction between the atoms making up the structure makes the configuration described above favorable from the energy standpoint. We consider the presence of three additional phason degrees of freedom for a structure of this sort. These degrees of freedom correspond to the possibility of a parallel displacement of the tube, which does not result in a change in the energy. In the course of such a displacement, local restructurings occur in the structure, since certain points disappear, intersecting the boundary of the tube, while others, on the contrary, appear. This behavior means that the local configurations containing points found by projecting the points near the edge of the tube form two-level systems, which undergo transitions from one state to the other upon a small phason shift. Each such two-level system can be in two states: "regular" and "irregular" with respective energies E_r and E_i. The regular (irregular) state corresponds to a point inside (outside) the tube and close to its boundary. We adopt an assumption which is consistent with the Goldstone nature of the phasons: the difference $\Delta E = E_i - E_r$ approaches zero as the distance (x) from the corresponding point to the tube boundary vanishes:

$$\Delta E(x) \to 0, \quad \text{as} \quad x \to 0. \tag{1}$$

The condition for thermodynamic stability holds: $\Delta E > 0$. Using condition (1), we can describe the defects in the following way. We assume that each two-level system is in one of the two states with a probability which depends on ΔE [or on x; see (1)], as shown in Fig. 1. Just how this distribution originates is not very important for our purposes; the origin may be either thermodynamic or associated with the growth kinetics. If the blurring of the tube, x_0, is slight, the concentration of two-level centers in the irregular position is on the order of x_0.

We will describe the defects in more detail for the so-called standard model, in which the tube is found with the help of a six-dimensional cube,[5] and its cross section is a triacontahedron. In this case the two-level systems lie along 15 families of parallel planes which are oriented perpendicular to the twofold axes of the diffraction pattern. The two-level systems which belong to a common plane are distributed along it with a finite x_0-independent density. The distance between adjacent parallel planes is on the order of x_0^{-1} (see Subsections 5 and 7 in Ref. 5). Since for any two-level system there are other two-level systems close to it, the various two-level systems cannot be regarded as statistically independent. It is reasonable to suggest that at a small value of x_0 i.e., at a large distance between planes, the correlation among the two-level systems which are close to a single plane is strong (the correlation radius is large), while the two-level systems corresponding to different planes are correlated only weakly.

For the system found, consisting of a sparse distribution of defects which themselves consist of highly correlated two-level systems, we calculate the diffuse scattering under the assumption that the various defects scatter incoherently (this assumption is

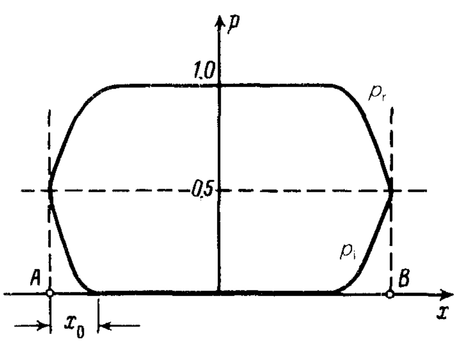

FIG. 1. For the regular and irregular states of two-level systems, the respective probabilities p_r and p_i are shown as functions of the position in the tube.

valid at small values of x_0). We consider a single defect, assuming at first that the correlation radius of the two-level systems along its plane is infinite. The positions of the atoms in the regular and irregular states are found by projecting the points at opposite parallel faces of the tube (in the case of projection onto R^{3*} these points fall on opposite rhombic faces of the triacontahedron). In order to calculate the incoherent part of the scattering, we must use the negative values of the contributions of the regular and irregular positions to the Fourier transform. Let us assume that the unit vector **n** is directed along a twofold axis, while unit vectors **a** and **b** form the sides of a face of the triacontahedron: projections of a six-dimensional cube parallel to the plane of the defect. Here is the result for the Fourier transform of a defect, $F(\mathbf{k}) = F(k_{\parallel}, \mathbf{k}_{\perp})$ $(\mathbf{k} = k_{\parallel}\mathbf{n} + \mathbf{k}_{\perp}, k_{\parallel} = (\mathbf{k},\mathbf{n}), (\mathbf{k}_{\perp},\mathbf{n}) = 0)$.

$$F(k_{\parallel}, \mathbf{k}_{\perp}) = 2i\sin(\lambda k_{\parallel}) \sum_{m} \delta(\mathbf{k}_{\perp} - \mathbf{k}_{m}) \frac{\sin(\mathbf{k}_m^*, \mathbf{a})\sin(k_m^*, \mathbf{b})}{(\mathbf{k}_m^*, \mathbf{a})(\mathbf{k}_m^*, \mathbf{b})}, \tag{2}$$

where $\lambda = \cos(\theta/2) - \sin(\theta/2), \cos\theta = 1/\sqrt{5}$ (the summation in (2) is carried out over the integer four-dimensional vectors $\mathbf{m} = (m_1, m_2, m_3, m_4)$]. The two-dimensional vectors $\mathbf{k}_m$ and $\mathbf{k}_m^*$ are defined by

$$(\mathbf{k}_m, \mathbf{x}_i) + (\mathbf{k}_m^*, \mathbf{y}_i) = 2\pi m_i, \tag{3}$$

where $\mathbf{x}_1 = \mathbf{a}$, and $\mathbf{x}_2 = \mathbf{b}$, $\mathbf{x}_3 = \varphi(\mathbf{b} - \mathbf{a})$, $\mathbf{x}_4 = \varphi^{-1}(\mathbf{a} + \mathbf{b})$, $\mathbf{y}_1 = \mathbf{a}$, $\mathbf{y}_2 = -\mathbf{b}$, $\mathbf{y}_3 = \varphi^{-1}(a + b)$, $\mathbf{y}_4 = \varphi(\mathbf{b} - \mathbf{a})$ and $\varphi = (\sqrt{5} + 1)/2$. The vectors **a** and **b** have been chosen in such a way that we have $(\mathbf{a},\mathbf{b}) = 1/\sqrt{5}$. The function $F(\mathbf{k})$ is nonzero on lines running parallel to the vector **n**. To find the intensity of the diffuse scattering, we need to smear the δ-function singularity in the function $|F(\mathbf{k})|^2$ in the direction perpendicular to **n**, by an amount on the order of the reciprocal of the correlation radius in the plane of the defect. We also need to allow for the circumstance that diffuse lines of the same sort are found for other twofold axes.

Here are the most important features of the pattern of diffuse scattering calculated in this way: 1) The system of diffuse lines has an icosahedral symmetry and a fairly complex distribution in k space. 2) Each diffuse line passes through peaks in the diffraction pattern. 3) The scattering intensity oscillates along a diffuse line. 4) Peaks having small values of the R^{3*} projections of the wave vector fall in those regions of diffuse lines where the scattering intensity is low. This comment applies to all of the sufficiently bright peaks (see Ref. 6 regarding the relationship between this property and the Goldstone nature of phasons).

Let us examine the experimental data available. Figure 2a shows the peaks reported in Ref. 1, which lie in a twofold symmetry plane; Fig. 2b shows the R^{3*} projections of these peaks. We first note that the intensities of the peaks fall off on the average with increasing length of the R^{3*} projection, in agreement with the tube model. We also see, however, that the intensity of the peaks depends on more than the R^{3*} projection. This circumstance is probably due to the complex structure of the unit cell (the material is a three-component material), which renders the standard model inapplicable from the quantitative standpoint.

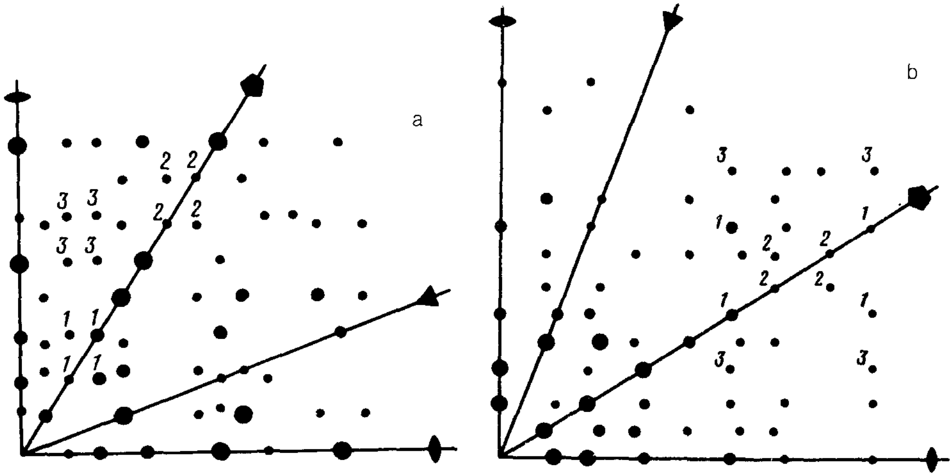

FIG. 2. a: The peaks found in Ref. 1, which lie in a twofold plane of the diffraction pattern, are shown. The peaks are classified somewhat arbitrarily into three groups on the basis of their intensity (high, intermediate, and low), as indicated by the sizes of the circles. Three groups of peaks which fall in the diffuse-scattering region are labeled 1, 2, and 3. b: The peaks shown in Fig. 2a when projected onto the orthogonal complement of R^{3*} (the notation is the same).

The diffuse-scattering regions which were observed in Ref. 1 contain the peaks shown in Fig. 2a. The projections of these peaks onto the orthogonal complement of R^{3*}, which are shown in Fig. 2b, are comparatively long. This result agrees with properties 2) and 4).

We see that the diffuse scattering in quasicrystals depends strongly on the configuration of the surface of the tube. Precise measurements of the scattering intensity distribution in k space might be useful for determining the atomic structure of these substances.

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