

Density of quasicrystals

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The density of atoms in an ordered icosahedral quasicrystal is related to the diffraction wave vector Q_{100000} : $n = a(\Lambda_1 + \Lambda_2\tau)Q_{100000}^3$, where Λ_1 and Λ_2 are integers, and τ is the golden section. In particular, for the stable quasicrystal $Al_{5.1}CuLi_3$ the numbers (Λ_1, Λ_2) are (2, 1) for Li and (4, 2) for Al (Cu).

The arrangement of atoms in ordered icosahedral quasicrystals is known to be described by the intersection of a periodic family of “atomic surfaces” in the $6D$ space R^6 with the $3D$ physical space R^3 (Refs. 1–3). The condition that a quasicrystal be completely ordered imposes some severe restrictions on possible discontinuities of atomic surfaces, however. Specifically, any point at the edge of a discontinuity can be associated with another point on the edge in such a way that the line segment which connects them is parallel to the physical space and does not exceed a few atomic spacings.¹ Our purpose in the present letter is to show that this condition leads to restrictions on the permissible densities of atoms in quasicrystals.

We factorize the atomic surface in terms of translations of a $6D$ periodic lattice. The surface is converted into a $3D$ manifold M^3 (which may have an edge), which is nested in a $6D$ torus T^6 . To calculate the density of atoms in real space, we recall that in the model of Ref. 4 this density is equal to the ratio of the volume of the cross section of the “tube” to the volume of the unit cell in R^6 . The reason is the complete incommensurability of the physical subspace, which leads to a dense filling of the cross section of the tube. A direct generalization of the expression for the density of atoms in this case takes the form

$$n = V_{\perp} / V_T, \tag{1}$$

where V_{\perp} is the $3D$ volume of the projection of the atomic surface onto the space which is orthogonal to R^3 , i.e., $R^{3\perp}$, and V_T is the volume of the $6D$ unit cell (of the torus T^6). The volume V_{\perp} can be written

$$V_{\perp} = \int_{M^3} d\omega^{\perp}, \tag{2}$$

where $d\omega^{\perp}$ is a 3-form of the volume in the space $R^{3\perp}$, defined on T^6 . Adding to M^3 some $3D$ films which repair discontinuities and which are made up of segments parallel to R^3 does not change the value of integral (2). The reason is that $d\omega^{\perp}$ vanishes on such films. We will accordingly assume that the discontinuities are repaired, and we will assume that M^3 is a manifold without an edge.

To evaluate integral (2), we note that $d\omega^{\perp}$ is a closed form on T^6 , and the integration over M^3 can be replaced by an integration over a combination of basis 3-

cycles on T^6 which is homologous with M^3 :

$$M^3 \propto \sum_{1 \leq i < j < k \leq 6} \lambda_{ijk} c^{ijk}, \quad (3)$$

where λ_{ijk} are integers, and c^{ijk} are basis 3-cycles. The conditions for icosahedral symmetry of the manifold M^3 , however, lead to the result that only two of the coefficients in (3) are independent:

$$M^3 \propto \Lambda_1 (c^{123} - c^{126} + c^{134} + c^{145} + c^{156} + c^{235} + c^{245} + c^{246} + c^{346} + c^{356}) \\ + \Lambda_2 (c^{124} - c^{125} + c^{135} - c^{136} + c^{146} + c^{234} + c^{236} + c^{256} + c^{345} + c^{456}), \quad (4)$$

The basis vectors in (4) are numbered in such a way that when projected onto R^{31} vectors 2, 3, 4, 5, and 6 make acute angles with the projection of vector 1 and are in clockwise order (when we look along the projection of vector 1).

The integrals of $d\omega^1$ over the basis 3-cycles are equal to the corresponding oriented volumes of parallelepipeds constructed in R^{31} on the projections of the basis vectors. Using (4), we find that an evaluation of the integrals leads to the following expression for the atomic density:

$$n = \frac{20\tau}{(2 + 2\tau^2)^{3/2}} (\Lambda_1 + \Lambda_2 \tau), \quad \text{where } \tau = (\sqrt{5} - 1)/2. \quad (5)$$

Here the volume of the $6D$ unit cell has been assigned a unit value. The value of n should be expressed in terms of quantities which can be found experimentally, e.g., the wave vector Q_{100000} , which can be measured in a diffraction experiment:

$$n = \frac{(\Lambda_1 + \Lambda_2 \tau) 20\tau}{(1 + \tau^2)^{3/2}} \left(\frac{Q_{100000}}{2\pi} \right)^3. \quad (6)$$

It is useful to compare (6) with the expression for the density of atoms in a cubic crystal:

$$n_c = N (Q_{100}/2\pi)^3, \quad (7)$$

where N is the number of atoms of the given species in the unit cell. We thus see that there is a pair of integers (Λ_1, Λ_2) corresponding to each species of atoms in the quasicrystal, just as a number N corresponds to each species of atoms in a crystal.

Before we apply (6) to real substances we should note that the vector Q_{100000} in quasicrystals, which have a simple cubic $6D$ lattice, is defined within a scaling transformation⁵:

$$S: \begin{cases} Q_{100000} \rightarrow \tau^3 Q_{100000} \\ \Lambda_1 \rightarrow 55 \Lambda_1 + 34 \Lambda_2 \\ \Lambda_2 \rightarrow 34 \Lambda_1 + 21 \Lambda_2 \end{cases}, \quad (8)$$

Accordingly, from all of the values of Q_{100000} generated by transformation S we are to select those for which Λ_1 and Λ_2 are sufficiently small. In this manner, we select the least intricate of all the atomic surfaces generated by transformation (8).

In conclusion let us examine the real, stable quasicrystal⁶ with the composition $Al_{5.1}CuLi_3$ from the standpoint of (6). In describing the positions of the Al and Cu atoms in the substance we should apparently use the same atomic surface. The reason is the pronounced mixing of the Al and Cu atomic positions which has been observed in the so-called R phase of Al_5CuLi_3 , which has a local order similar to that of a quasicrystal.⁷ A diffraction experiment yields two possible values for the wave vector Q_{100000} : 0.623 \AA^{-1} and 2.639 \AA^{-1} , which are related by transformation (8). The quantities $\Lambda_1 + \Lambda_2\tau$ found from (6) through the use of the value $Q_{100000} = 0.623 \text{ \AA}^{-1}$ are 2.610 for Li and 5.290 for Al (Cu). We are thus very close to

$$\begin{array}{l} \Lambda_1 = 2 \\ \Lambda_2 = 1 \end{array} \quad \text{for Li and} \quad \begin{array}{l} \Lambda_1 = 4 \\ \Lambda_2 = 2 \end{array} \quad \text{for Al (Cu)}. \quad (9)$$

That the numbers Λ_1 and Λ_2 have turned out to be small seems to mean that $Al_{5.1}CuLi_3$ is an ordered quasicrystal and obeys constraints (6).

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