Phase diagram of inelastic instability of icosahedral quasicrystals

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The expansion of the elastic energy of icosahedral quasicrystals in the "phonon" displacements is considered. It is shown that an appropriate Ginzburg–Landau functional describes a first-order phase transition in which the symmetry is lowered to a pentagonal or triangular symmetry. A suitable phase diagram is constructed.

Since the discovery of quasicrystals[1] this new type of solids has stimulated increasing interest.[2] Many quasicrystals obtained experimentally have a definite disorder relative to an ideal Penrose lattice, which manifests itself in the displacement and broadening of the diffraction peaks.[3] This disorder is customarily regarded as a certain displacement field which corresponds to the "phason" degrees of freedom and which arises during the growth process.[4–6]

However, because of the presence, in the decay of the elastic energy of an icosahedral quasicrystal, of a term which relates the phason components of strain to the phonon components,[7,8] such a disorder may accompany ordinary displacements.[9] Using the method of atomic density functional, Jaric and Mohanthy[10] have calculated the elastic constants of an icosahedral quasicrystal and detected a mechanical instability due to spontaneous strain which corresponds to one of the four irreducible representations of the icosahedral group Y. From the number of these representations it can be inferred that fifteen components of the strain tensor of the quasicrystal are transformed. The symmetry of the phason spontaneous strain was analyzed by Ishii.[11]

In the present letter we will examine the Ginzburg–Landau expansion as an order parameter for a phonon-type spontaneous deformation.

The Landau theory of such phase transitions, sometimes called ferroelastic transitions, has been developed extensively.[12,13] The general procedure involves the determination, based on the symmetry considerations, of the nonzero components of the spontaneous strain tensor of the low-symmetry phase and the determination, in the next expression, of the coefficients of the free-energy expansion in the magnitude of the strain as a one-component order parameter in terms of the elastic constants of different powers.

In an icosahedral medium the strain tensor transforms in accordance with the direct sum of two irreducible representations of the group Y: a single and a $5D$ irreducible representation. Since a single representation does not correspond to a lowering of the symmetry, the structure of the low-symmetry phase will be determined by those maximum subgroups of Y, upon the reduction to which from a $5D$ irreducible representation a single representation of the subgroup splits off. It is easy to see that D_5 and D_3 are such subgroups, which were detected previously[14] in the analysis of the expansion of the free energy in the amplitudes of the density waves. The strain refers to the even representations, so we can replace D_m by C_m which has no reflections.

The symmetry of the shear tensor in C_m gives rise to a uniaxial strain

$$\epsilon_{ij} = \epsilon(3n_in_j - \delta_{ij}), \tag{1}$$

where **n** is a unit vector directed along the axis of rotation. In Voigt's notation we have

$$\begin{aligned} \eta_1 &= 3n_1^2 - 1; & \eta_2 &= 3n_2^2 - 1; & \eta_3 &= 3n_3^2 - 1; \\ \eta_4 &= 3n_2n_3; & \eta_5 &= 3n_1n_3; & \eta_6 &= 3n_1n_2 \ . \end{aligned} \tag{2}$$

We restrict the expansion to the fourth order, as is usually the case in problems of this sort.[15] The number of independent second-, third-, and fourth-order elastic constants for an icosahedral medium is equal to the number of times a single irreducible representation is included in the second, third, and fourth symmetric degrees of representation corresponding to the strain tensor (2, 4, and 6, respectively).

In the basis, which determines the direction of the 5-fold axis of symmetry as

$$\begin{aligned} \mathbf{e}_{m+1} &= (\sin(\frac{2\pi m}{5})\sin\theta; \quad \cos(\frac{2\pi m}{5})\sin\theta; \ \cos\theta) \\ m &= 1, 2, 3, 4, 5, \qquad \mathbf{e}_1 = (0, 0, 1), \end{aligned} \tag{3}$$

where $\theta =$ arctan of (2), there arise 26 third-order nonvanishing elastic constants. Of these constants, twelve are distinct. For the fourth order, 64 elastic constants are respectively nonvanishing and 28 are distinct.

Substituting into the Ginzburg–Landau expression for the elastic energy the order parameter such as (2), we obtain an expansion in powers of the vector components **n**.

Here the νth order in the strains corresponds to the power 2ν in $\{n_i\}$. The number of effective elastic constants which determine the expansion of the energy in powers of the uniaxial strain will then be equal to the number of times a single irreducible representation is included in the corresponding symmetric power of the canonical $3D$ irreducible representation, according to which the coordinates of the vector $\mathbf{n}$ are transformed. For an icosahedral symmetry, these multiplicities are 1, 2, and 2 for the fourth, sixth, and eighth powers, respectively. Consequently the number of effective "uniaxial" elastic constants of the second, third, and fourth orders are 1, 2, and 2.

In basis (3) the expression for the order parameter corresponding to the symmetry C_5 with a 5-fold axis along the z axis can be written

$$\eta = \frac{1}{\sqrt{6}}(-\eta_1 - \eta_2 + 2\eta_3),$$

and the free-energy expansion can be written

$$\Delta F = \frac{C_{11} - C_{12}}{2}\eta^2 + \frac{1}{6\sqrt{6}}(4C_{114} + C_{456})\eta^3 + (-3C_{1444} + \frac{3}{8}C_{4444})\frac{\eta^4}{36}. \quad (4)$$

Similarly, for symmetry C_3 we find

$$\eta = \sqrt{\frac{5}{23-3\tau}}\left(-\frac{\eta_1}{\tau} - \frac{\eta_2}{\tau+2} + \frac{2\eta_3}{\sqrt{5}} + \eta_4\sqrt{\frac{3\tau+2}{5}} + \eta_5\sqrt{\frac{\tau+1}{\tau+2}} + \frac{\eta_6}{\sqrt{\tau+2}}\right),$$

where $\tau = (\sqrt{5}+1)/2$ is the "golden section."

$$\Delta F = \frac{15}{23-3\tau}(C_{11} - C_{12})\eta^2 + \frac{5\sqrt{5}}{(23-3\tau)^{3/2}}(-4C_{114} + C_{456})\eta^3 + \frac{25}{(23-3\tau)^2}(C_{1444} + \frac{3}{8}C_{4444})\eta^4. \quad (5)$$

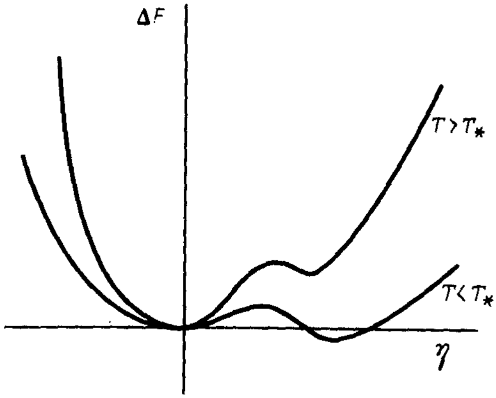

FIG. 1. Free energy (6) vs the order parameter for various temperatures. $T_* = T_c + (2/9)\theta$ corresponds to a first-order phase transition.

We recall that the Landau–Ginzburg expression for a one-component order parameter

$$\Delta F = \frac{\alpha}{2}(T - T_c)\eta^2 + \frac{\beta}{3}\eta^3 + \frac{\gamma}{4}\eta^4 ; \qquad \alpha, \gamma > 0 \tag{6}$$

at $\beta \neq 0$ describes a first-order phase transition at a temperature $T_* = T_c + 2/9\theta$, where $\theta = \beta^2/\alpha\gamma$ (Fig. 1). Transforming expansions (4) and (5) to (6) and introducing the notation

$$\alpha_0 = \frac{\partial}{\partial T}\Big|_{T=T_c}(C_{11} - C_{12}); \quad \theta_0 = \frac{C^2_{456}}{\alpha_0 C_{4444}}; \quad \zeta_3 = \frac{C_{114}}{C_{456}}; \quad \zeta_4 = \frac{C_{1444}}{C_{4444}}$$

we find two ways to lower the symmetry

$$Y \to C_s \qquad \theta_s = \frac{(1 + 4\zeta_3)^2}{1 - 8\zeta_4}\theta_0$$

$$Y \to C_3 \qquad \theta_3 = \frac{3(1 - 4\zeta_3)^2}{3 + 8\zeta_4}\theta_0$$

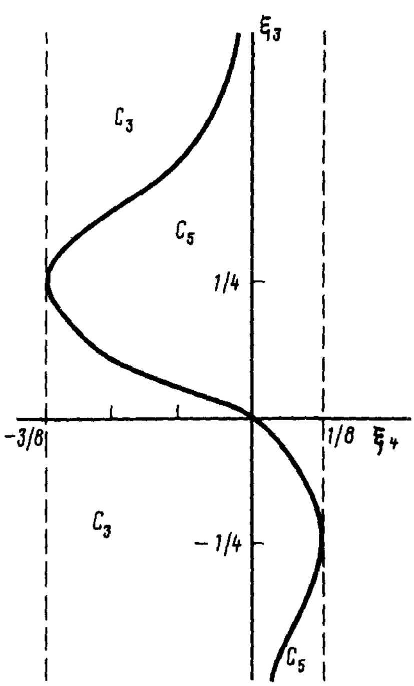

FIG. 2. Phase diagram showing the symmetry of the low-symmetry phase as a function of the relationship between the effective elastic constants.

In the coordinates ζ_3, ζ_4 we can construct a phase diagram in which the regions corresponding to the condition $\theta_5 > \theta_3$, where the symmetry is lowered to C_5, are separated from the regions where $\theta_3 > \theta_5$ and the low-symmetry phase is described by the subgroup C_3.

Let us assume that $C_{4444} > 0$. The condition $\gamma > 0$ for the two expansions, (4) and (5), singles out in the plane the band $-3/8 < \zeta_4 < 1/8$, in which the symmetry can be lowered to each subgroup, depending on the relationship between θ_3 and θ_5 (Fig. 2). At $\zeta_4 \geqslant 1/8$ there can be a transition only to C_3 and at $\zeta_4 \leqslant -3/8$ a transition only to C_5 can occur. An interesting point is that there is no loss of stability due to a second-order phase transition at $\beta = 0$ (i.e., $\zeta_3 = 1/4$ for the symmetry C_3 and $\zeta_3 = -1/4$ for C_5 in the region in which the various low-symmetry phases compete with each other.

If $C_{4444} < 0$, then a transition to C_5 occurs to the right of the line $\zeta_4 = 1/8$, while a transition to C_3 occurs to the left of the line $\zeta_4 = -3/8$, and a competition region does not arise.

We can therefore conclude that the spontaneous deformation leads to a lowering of the symmetry from the icosahedral to the pentagonal or triangular symmetry, depending on the relationship between the effective elastic constants of the third and fourth orders. In the region in which there is a competition between the various low-symmetry phases the transition is a first-order transition.

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