Quasicrystals with Dodecahedral Equilibrium Faceting

Two recent Letters[1,2] addressed the issue of equilibrium faceting in icosahedral quasicrystals. Ho *et al.*[1] considered lattice models with purely attractive short-range two-body forces and argued that the equilibrium shape could not be a dodecahedron. However, dodecahedral faceting has been observed in some quasicrystals.[3] If Ho *et al.*'s result could have been generalized, it would have meant that such faceting could only occur in nonequilibrium growth processes. In this Comment we show that a more general model admits a wider range of equilibrium quasicrystal shapes, including the dodecahedron.

The equilibrium shape of a material can be obtained if we first calculate the surface free-energy density $\gamma(\hat{\mathbf{n}})$ for creation of a plane surface normal to angular direction $\hat{\mathbf{n}}$, and then use the Wulff construction.[4] At $T=0$, $\gamma(\hat{\mathbf{n}})$ for icosahedral models with two-body interactions has the form

$$\gamma(\hat{\mathbf{n}})=\sum_\mu g^{(\mu)}\sum_\alpha |\hat{\mathbf{n}}\cdot\hat{\mathbf{A}}_\alpha^{(\mu)}|. \qquad (1)$$

The unit vectors $\hat{\mathbf{A}}_\alpha^{(\mu)}$ depend on the details of the model; in general, the vectors can be divided into finite sets, labeled by μ, each with icosahedral point-group symmetry. In Refs. 1 and 2, the coefficients $g^{(\mu)}$ are all positive, representing purely attractive interactions within the material. If a great circle normal to each vector $\hat{\mathbf{A}}_\alpha^{(\mu)}$ is constructed in angular space, then $\gamma(\hat{\mathbf{n}})$ has a cusp in the direction $\hat{\mathbf{n}}$ where two or more such circles intersect.

Surface energies of the form of Eq. (1) lead to Wulff shapes which are completely faceted, with no rounded corners or edges.[5] Each facet is normal to one of the cusp directions. Moreover, when all the coefficients $g^{(\mu)}$ are positive, every cusp direction must produce a facet normal to it. In this case, a necessary, but not sufficient, condition for a given faceted shape to be a possible equilibrium shape is that there exist a set of great circles whose intersections are in one-to-one correspondence with the facets, and such that each intersection lies along the outward normal direction to a facet. A dodecahedron does not satisfy this criterion because any set of great circles with intersections along the outward normal direction to each of the twelve facets necessarily has many other intersections as well.

Our principal point is that new equilbrium shapes, including the dodecahedron, are possible if one considers repulsive, as well as attractive, interactions. Repulsion could arise because of either inherently repulsive two-body forces or steric crowding of atoms or clusters within a material. Generalizing the model of Ref. 2 to include repulsive bonds leads to a free energy of the form of Eq. (1) with coefficients $g^{(\mu)}$ of mixed sign. With mixed signs, the Wulff facets normal to some cusps in $\gamma(\hat{\mathbf{n}})$ are reduced in size or completely suppressed. Figure 1 illustrates this, showing the equilibrium shapes obtained from two sets of vectors: $g^{(1)}$ is positive and held fixed; $g^{(2)}$ is positive in Fig. 1(a), zero in Fig. 1(b), and progressively more negative in Figs. 1(c)–1(e). As the repulsive term increases, all but twelve facets shrink and eventually disappear, leaving a dodecahedron [Fig. 1(e)] as the final shape. At each stage the net energy required to dissociate all bonds within the bulk remains attractive, and $\gamma(\hat{\mathbf{n}})$ remains positive for all $\hat{\mathbf{n}}$.

We believe that our model, while not physically realistic, indicates that the dodecahedral faceting observed in some quenched quasicrystals is not precluded from being the equilibrium shape as well. Further details of the model and possible equilibrium shapes will be published at a later date.

This work has been supported in part by the National Science Foundation Materials Research Laboratories Program, under Grant No. DMR-8519059.

Kevin Ingersent and Paul J. Steinhardt
Department of Physics
University of Pennsylvania
Philadelphia, Pennsylvania 19104

Received 11 February 1988
PACS numbers: 61.50.Em, 61.50.Jr

[1]T. L. Ho, J. A. Jaszczak, Y. H. Li, and W. F. Saam, Phys. Rev. Lett. **59**, 1116 (1987).

[2]A. Garg and D. Levine, Phys. Rev. Lett. **59**, 1683 (1987).

[3]A.-P. Tsai, A. Inoue, and T. Masumoto, Jpn. J. Appl. Phys. Pt. 2 **26**, L1505 (1987); W. Ohashi and F. Spaepen, Nature (London) **330**, 555 (1987).

[4]G. Wulff, Z. Kristallogr. **34**, 449 (1901).

[5]C. Herring, Phys. Rev. **82**, 87 (1951).

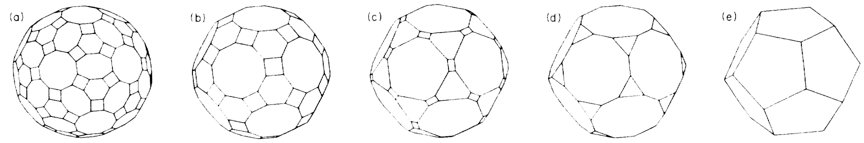

FIG. 1. A sequence of equilibrium shapes for icosahedral quasicrystals, showing the effect of the incorporation of a combination of attractive and repulsive bond forces. See text for details.

