Comment on "Icosahedral Quasiperiodic Ground States?"

Narasimhan and Jarić have recently suggested that a particular optimized icosahedral quasicrystal structure may actually minimize the enthalpy for a monatomic solid with a certain square-well potential.[1] Their model uses a hard-sphere diameter σ with an attractive square well of width w; this potential has been used elsewhere as a simple model for a liquid phase.[2] For values of the ratio w/σ between $[2(1+1/\sqrt{5})]^{1/2}-1\approx 0.701$ and $\sqrt{3}-1\approx 0.732$, Narasimhan and Jarić found that their icosahedral quasicrystal model had an energy below those of the several simple periodic structures they tried: fcc, bcc, and hcp. We have found a periodic crystal (albeit one with a thirteen-atom basis) that does considerably better than the authors' icosahedral quasicrystal in this range; furthermore, because it also has a higher packing fraction, it has a lower enthalpy than the quasicrystal at any pressure (again in the given range for w/σ). This may not be the most energetically favored of all possible structures, but it appears to rule out the possibility of a ground-state one-component quasicrystal of the class considered by Narasimhan and Jarić, at least with this simple potential.

For hard spheres with a square-well attraction, the total potential energy of a particular structure is determined solely by the effective coordination number, by which we mean the average number of neighbors in the distance range from σ to $\sigma+w$. The effective coordination numbers for some simple periodic structures are 16 (diamond), 18 (fcc), and 26 (bcc), giving energies of -8ϵ, -9ϵ, and -13ϵ, respectively, where ϵ is the depth of the attractive part of the potential. The energy of -13.5ϵ quoted in the Letter[1] is only slightly more favorable than the bcc value. The cubic and close-packed structures gain a coordination shell at $w=0.732$; see Fig. 1 of Ref. 1. The proposed icosahedral quasicrystal occupies only a small height of the phase diagram because it has a much lower packing fraction than its bcc competitor.

We have found that the $\{W/Mo/(Mn,Cr)\}Al_{12}$ structure first determined by Adam and Rich[3] has a lower enthalpy in the entire range shown as occupied by the quasicrystal by Narasimhan and Jarić. (We replace the transition metals of the structure with a thirteenth Al.) This structure comprises regular icosahedra placed at the points of a bcc lattice; it can also be described as a packing of rhombohedra and rhombic dodecahedra and is related to a simple rational approximant of the three-dimensional Penrose tiling.[4] For $0.701\leq w<0.973$, the twelve icosahedral vertices in a cell each have effective thirtyfold coordination. The remaining point (the icosahedron center) is 36-fold coordinated; this gives a potential energy per atom of -15.23ϵ, a significant improvement over the quasiperiodic structure.

Our monatomic periodic structure is not intended as an actual structural prediction but rather as a counterexample to the proposal advanced by Narasimhan and Jarić in the Letter. While we cannot rule out the possibility that the ground state of some monatomic system is quasiperiodic, we see no reason that a more general member of the class of structures they investigated will provide the ground state of a simple square-well potential. This suggests that quasiperiodic ground states may be limited to two-or-more-component compounds.

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[1]Subha Narasimhan and Marko V. Jarić, Phys. Rev. Lett. **62**, 454 (1989).

[2]See, e.g., J. P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic, Boston, 1986), 2nd ed., p. 5.

[3]J. Adam and J. B. Rich, Acta Crystallogr. **7**, 813 (1954); see also D. P. Shoemaker and C. B. Shoemaker, in *Introduction to Quasicrystals,* edited by M. V. Jarić (Academic, Boston, 1988), p. 15.

[4]V. Elser and C. L. Henley, Phys. Rev. Lett. **55**, 2883 (1985); C. L. Henley and V. Elser, Philos. Mag. B **53**, L59 (1986).