Understanding Quasicrystals

Abstract

We have examined the possibility that quasicrystals can be better understood as condensed orientation waves than as condensed density waves. We have studied simulation data of hard tetrahedra that result in a quasicrystal approximate using a Fourier transform of the spherical harmonics associated with the orientation of the tetrahedra, rather than a Fourier transform of the locations of the centers or corners of the tetrahedra. We find that some orientation peaks have significantly higher brightness when the quasicrystal forms, demonstrating that among quasicrystals, at least the hard tetrahedra approximate is better understood as a condensed orientation wave than a condensed density wave.

Background

Quasicrystals (quasiperiodic crystals) have been observed in a variety of metallic systems\(^1\) and recently also in simulations of hard tetrahedra.\(^2\) In quasicrystals atoms are arranged in such a way that the Fourier transform of the positions of their centers, or indeed of any characteristic of the structure has sharp peaks at a dense number of points. These peaks cannot be indexed by a simple three-dimensional lattice but are indexed by a higher dimensional lattice, with the height of the peak related to the index of the lattice.

The Landau theory of phase transitions suggests that quasicrystal systems should be characterized by a single order parameter, but such a parameter is not yet known. Proposed descriptions of quasicrystals assume the order parameter is a density wave. However, orientation waves are also consistent with crystalline order, and the central hypothesis of the project is that orientation waves serve as order parameters for quasicrystals.

Prior work on simulations of hard tetrahedra have shown they form a dodecagonal quasicrystal or its approximate at moderate pressures.

Methods

Tetrahelices (rotated tetrahedra in a helical arrangement) are common in quasicrystals, and are composed of tetrahedra with spatially varying orientations; it is natural, then, to suppose that the important order parameter in quasicrystals containing them is an orientation wave associated with tetrahedra. The simplest way to describe the orientation of a tetrahedron is to use a three rank traceless symmetric tensor, an object that has the eigenvalue of the generator of rotations (the angular momentum) \(l \geq 3\) and which is odd under spatial inversion. It makes sense to try to describe these phases by order parameters that vary in space, are odd under inversion, and have angular momentum around the helix axis of \(m = 0, 1, 2, 3\). Each tetrahedron can thus be described by the position of its center and a single \(l = 3\) spherical harmonic that describes its orientation. We then do a spatial Fourier transform of these spherical harmonics in order to examine order parameters associated with orientation waves.

We received data of simulations of a hard tetrahedra fluid packing into a quasicrystalline approximate structure at pressure (above). We have redone the scattering for the density and the \(l = 3\) spherical harmonic orientation waves of the tetrahedra for all values of \(m\) and both high and low pressure (below). The scattering is indexed by the reciprocal lattice of the approximant. Only data in the basal plane – the plane of the poster in the diagram above – is shown.

Results & Conclusions

The “Low Pressure” diagrams correspond to a simulation pressure, just above the pressure at which the approximant forms in equilibrium. The “High Pressure” diagrams are from a pressure 5 times larger, with the crystal nearly fully formed. The brightness of a peak indicates its intensity, here shown by the area of the peak.

The High Pressure diagrams have many intense scattering spots. Most of the orientation waves with \(m \neq 0\) can be understood as resulting from plausible tetrahelices, and the density and \(m = 0\) peaks all correspond to \(m = 0\) orientation waves.

Only a few intense peaks are seen in the Low Pressure diagrams. The most intense peaks are the \(m = 0\) and \(m = 1\) peaks at \((n, n, m) = ( \pm 3, \pm 3, 0)\), \(( \pm 1, \pm 1, 0)\), and \(( \pm 4, \pm 4, 0)\). Moreover, the locations of all intense peaks in the basal plane at high pressure (which are quite scattered and hard to rationalize otherwise) can be written the sum of a small number of these wavevectors.

This shows that these waves are the order parameter for the approximant, and in turn demonstrates that at least the hard tetrahedra approximant is better understood as a condensed orientation wave than a condensed density wave. We speculate that other quasicrystals can also be rationalized in this way. Preliminary Landau Theories based on such order parameters, and specifically on the waves observed in this quasicrystal approximate are in progress and seem to naturally result in quasicrystals.

Acknowledgments

I would like to thank Professor Rolfe Petschek for his continued guidance and support, as well as Professors Amir Haji-Akbari, Michael Engel, and Sharon C. Glotzer (Department of Chemical Engineering, University of Michigan) for providing the data on the quasicrystal approximate.

References

1. See e.g. Quasicrystals : the state of the art / editors, D.P. DiVincenzo, J.P. Steurer (Imprint Singapore) / River Edge, NJ : World Scientific, c1999
2. Haji-Akbari, Amir; Engel, Michael; Keys, Aaron S.; Zheng, Xiaoyu; Petschek, Rolfe G.; Palffy-Muhoray, Peter; Glotzer, Sharon C.; “Disordered, quasicrystalline and crystalline phases of densely packed tetrahedra” Nature 462: 773-777 http://dx.doi.org/10.1038/nature08641

Density

Intensity of orientation and density waves in basal plane (a) at lowest pressures for which quasicrystal is stable, indexed by the reciprocal lattice of the approximant

Density

Intensity of orientation and density waves in basal plane at a pressure 5 times the lowest pressure for which quasicrystal is stable, indexed by the reciprocal lattice of the approximant