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Open Theoretical Problems in the Physics of Aperiodic Systems*

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Abstract. Quasicrystals have intrigued and stimulated research in a large number of disciplines. Mathematicians, physicists, chemists, metallurgists and materials scientists have found in them a fertile ground for new insights and discoveries. In the quarter century that has ensued since the publication of the experimental observation of a quasiperiodic Al-Mn alloy,¹ many different kinds of quasiperiodic alloys have been manufactured and studied. The physical properties of quasicrystals are no less interesting than the unusual structural properties that led to their discovery in 1984. In this review, some of the properties that characterize quasicrystals are presented, several types of theories that have been put forward briefly discussed, and some new behaviors that might be investigated by experiment described.

Keywords: complex metallic alloys

WHAT CHARACTERIZES A QUASICRYSTAL?

Diffraction patterns are widely used to distinguish different types of structures in condensed matter physics. The 1984 paper of Shechtman *et al.*¹ entitled "Metallic phase with long ranged orientational order and no translational symmetry" studied the alloy using electron and X-ray diffraction. The two main characteristics of these patterns seemed to contradict each other: a) there were well-defined spots, indicating a long range order as in crystals in the way atoms were placed, and b) the bright spots in one of the planes were arranged in a 5-fold symmetric way. Elementary geometrical arguments rule out, as is well-known, the possibility of such 5-fold axes in a periodic crystalline array. As better and better quasicrystals have appeared on the scene, the diffraction patterns have evolved to eliminate much of the initial diffuse scattering, showing that these materials have excellent long range structural order, rivaling those of the best crystals.

Since those early days, many new experimental probes have become available, including high resolution transmission electron microscope (HRTEM), scanning tunneling microscopy and spectroscopy (STEM) or low energy electron diffraction (LEED), to name just a few that are commonly used to study surface layers of quasicrystals. Such studies lead in a natural way to the notion of atomic clusters. These are used to explain, for example, the repeating motifs that are seen in the STM or the

LEED patterns. The question of how such clusters can be put together to form an ordered quasicrystal then arises, and many authors have addressed this nontrivial problem (see for example the compilations in Refs. 2 and 3).

While the concept of clusters arises when viewing quasicrystalline surfaces at close range, long range quasiperiodic order as seen in diffraction studies is perhaps more naturally described in terms of quasiperiodic tilings. These are the analogs of the "Bravais lattices" for periodic systems, and give a good description of the orientational order and positional long range order seen in quasicrystal diffraction patterns. Tilings are constructed from a small number of tiles arranged so as to avoid overlapping or holes. Quasiperiodic tilings usually possess a hierarchically organized structure, where tilings can be "inflated" ("deflated") so as to obtain a similar tiling on a larger (smaller) length scale. It was realized early on that quasiperiodic tilings can be obtained by a projection down from a higher dimensional periodic array. The important early papers can be found in Ref. 4, and the basic idea is illustrated in Figure 1 below, which indicates schematically how one can obtain quasiperiodic tilings such as the Fibonacci chain, the Penrose tiling, or the icosahedral tiling. Points of the D dimensional hyper cubic lattice that lie within an infinite strip are projected onto the irrationally oriented $d < D$ dimensional physical space in which the tiling is defined. The points in the strip are chosen such that their

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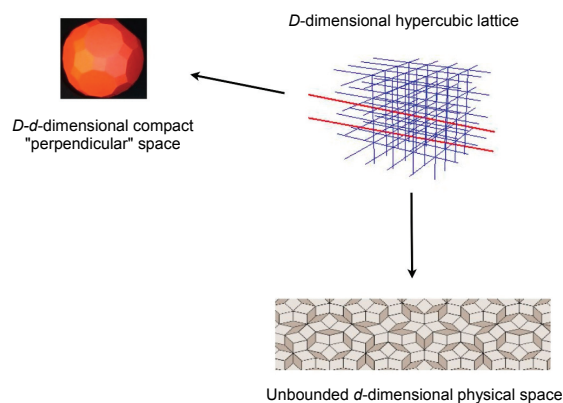


Figure 1. Schematic view of the cut-and-projection method: all the vertices of a D -dimensional lattice (center) in the region delineated by irrational planes are chosen for projection. The two possible projections give (bottom) vertices of a tiling in the physical space, or (left) a densely filled region of the complementary "perpendicular" space. Polyhedron reproduced with permission from the collection of T. Uchida.

projections onto the perpendicular subspace (the remaining $D-d$ perpendicular directions) fall inside a polyhedral volume, called the selection window. Thus, every vertex of the tiling possesses a counterpart in the compact perpendicular space window. This property provides one with a convenient way of representing certain spatial properties of quasicrystals, as we will see in our magnetic model later.

The method of projection manifestly implies long range order, admittedly of a complex type. It also leads to repetition of local atomic clusters (obtained by decorating the vertices of the D dimensional lattice). The method can be generalized to obtain many types of structures, including various kinds of disordered ones. In addition, by increasing the dimension D , one can go from the crystal ($D = d$) to more and more strongly aperiodic structures. This provides interesting models, for example, when one wishes to study the localization of electrons, or, as we consider here, magnons in different quasiperiodic structures.

ELECTRONIC, MAGNETIC AND VIBRATIONAL PROPERTIES

Leaving the subject of determination of the structures of specific quasicrystals, we now raise the question: can one make some general statements about the consequences of quasiperiodic structural order on physical properties of these materials?

A common feature of most quasicrystalline alloys is the dip, or pseudogap, in the electronic density of states close to the Fermi level.⁵ This pseudogap presumably leads, following the argument of Hume-Rothery,⁶

to the formation of the quasicrystalline phase for certain values of the electron concentration. The pseudogap is found in approximants as well. In other words, it depends primarily on the short range structural details, with modifications due to the long range order. Short range structural order also determines the magnitude of the electrical conductivity to a large extent. It plays an important role in magnetic properties of magnetic quasicrystalline alloys such as the Curie-Weiss temperature, or the spin glass transition temperature.

To observe properties arising due to long range quasiperiodic order, one has to go to low temperatures and of course make measurements on systems which are perfectly quasiperiodically ordered on a sufficiently long length scale. Some phenomenological models have been proposed for the transport in quasicrystals, such as a prediction based on hierarchical structure models for the temperature dependence of the electrical conductivity of icosahedral alloys.^{7,8} Vibrational properties, similarly, should reflect the hierarchical structure present in quasicrystals. At low energies, phonon modes may for example show anomalous dispersion laws. This in turn could lead to a nontrivial temperature dependence of the lattice contribution to the thermal conductivity. In the case of aerogels which are disordered fractals, such effects have been shown in neutron scattering and by thermal conductivity experiments. Recently studies have been carried out to investigate the thermal conductivity in a number of quasicrystals.⁹

Computational approaches such as *ab-initio* and molecular dynamics calculations have been successful in fitting some experimentally observed electronic¹⁰ and vibrational¹¹ properties. Analytical results are on the other hand scarce, even in the simplest of models, due to the lack of appropriate theoretical tools. It is however important to study simplified models for their ability to isolate some simple generic behaviors. Thus, for example, tight-binding models are useful for systems such as graphene, to mention one much studied current problem, or high-temperature superconductors. Such models allow to predict behaviors independent (to a first approximation) of details of chemistry and could be applied to a family of related compounds.

In this context, a promising direction of research, in terms of isolating the physics of quasiperiodicity, is the attempt to generate a simple single-element quasicrystal by deposition on an appropriate substrate. This is described in several contributions in the CMA-Zagreb '08.¹² As will be discussed in more detail in the last section, surface probes on such quasicrystalline films should provide direct evidence of some of the most interesting properties of quasiperiodic materials. One interesting challenge would be to excite and observed critical states, somewhat along the lines of the "quantum corral" experiment on a copper substrate¹³ in which it was possible

to image a electron state localized on Fe atoms that were dragged into a ring-shape by an STM tip. The electron problem is unfortunately complicated by electron-electron interactions, but magnon modes on a quasiperiodic surface should exhibit this type of critical states, as we discuss next.

The following section describes a problem that was studied recently for two dimensional quasicrystals: the antiferromagnetic ground state, and magnon excitations. We will see that many properties observed in the phonon and electron problems are carried over to the magnetic case, showing that quasiperiodic antiferromagnets could provide a new testing ground for unusual properties of quasicrystals.

MODEL OF A QUASIPERIODIC QUANTUM ANTIFERROMAGNET

We illustrate the properties of a Heisenberg antiferromagnet in a 2 dimensional quasiperiodic structures such as the Penrose rhombus tiling or the Ammann-Beenker (octagonal) tiling. The Hamiltonian is taken to be of the simplest possible form:

$$H = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j \quad (1)$$

where the spins are situated on vertices of the tiling, and the bonds act along edges connected nearest neighbors. The bonds are all assumed of the same magnitude, $J > 0$, and the most interesting situation corresponds to the case of $S = 1/2$ spins. The questions we address are a) what is the quantum ground state of this system? and b) what are the equivalent of magnons (the elementary excitations in a periodic antiferromagnet) in a quasiperiodic material?

The answers were found by approximate methods (both numerical, using a spin wave expansion and analytical, using renormalization group) and by Quantum Monte Carlo (QMC) calculations (see Ref. 14 for details and references). The spin wave method, analogous to the one used for periodic systems, does not require wave properties, as one can work in a real space basis without need for any Fourier transforms. In agreement with the QMC results, we find the result shown in Figure 2 for the ground state magnetic configuration of the spins in one of the approximants of the Penrose tiling. In the figures, each site has been colored according to the absolute value of its local magnetization $|\langle S_{iz} \rangle|$. The upper figure shows the sites in real space (the tile edges have not been drawn), showing that the ground state magnetization is quite complex. One can discern a self-similar structure, even within the small region that is shown. In the lower figures (Figures 2a and 2b), the same ground state is represented using the perpendicular space coordinates of the sites of the tiling (Figures 2a and 2b indi-

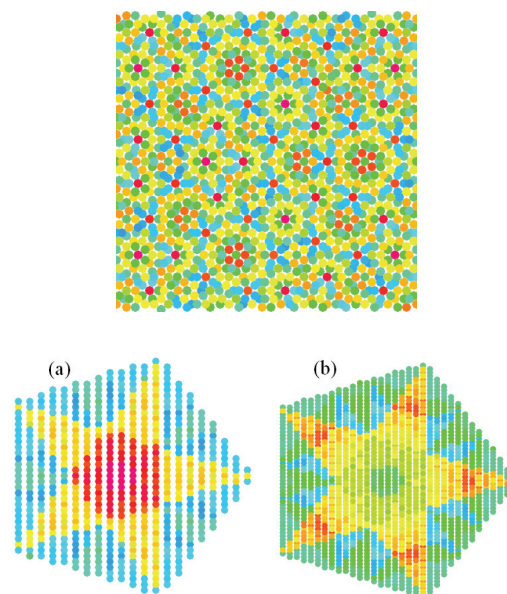


Figure 2. Color representation of the distribution of absolute values of the local magnetizations in the ground state of a Penrose antiferromagnet. (Upper figure) as seen in real space. (Lower figures (a) and (b)) after projection onto two regions of the perpendicular space (colors range from red for the smallest values, to blue for the highest values)

cate two out of the four domains that constitute the full set of vertices). These figures show an alternative view of the ground state structure, which is perhaps simpler, with colors grouped together in domains. This representation is a compact way to show the ground state magnetic structure, and also provides an elegant way to demonstrate self-similarity (not discussed here).

The ground state shown above is the result of quantum fluctuations – magnons, or spin excitations – of the quasiperiodic Heisenberg model. The magnon spectrum and the real space-dependence of the magnons have been determined in spin wave theory. The density of magnon states is very reminiscent of that of scalar phonons in quasiperiodic tilings,¹⁵ with a linear low energy "dispersion relation" for low energy modes, as in other types of solids. The values of the effective spin wave velocity depend of course on the specific structure. One can compare the values of the velocity with that found in the square lattice, which is a reference system having the same average connectivity ($z = 4$) as the Penrose tiling or the octagonal tiling. One finds that the velocities decrease in the order square lattice \rightarrow octagonal tiling \rightarrow Penrose tiling, reflecting the rise in complexity as one goes from one structure to the next. When the quasiperiodic tiling is disordered by permuting tiles (adding frozen phason disorder), the sound velocity increases, whereas experience based on periodic systems would indicate the contrary.¹⁶ This is the equivalent of a phenomenon already observed in studies of electron

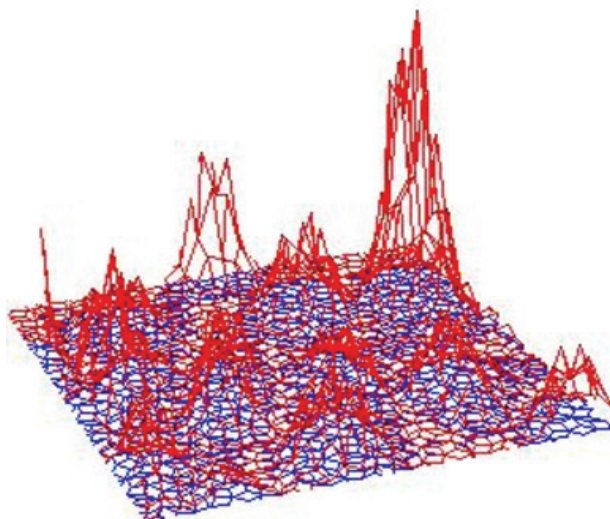


Figure 3. (Color online) A representation of a magnon wavefunction in a Penrose tiling, corresponding to a low energy state $E = 0.82$, showing the square of the wavefunction for each of the sites in a finite region of the tiling. Although the system size is small, it illustrates the recurrent peak structure and power law decay.

motion in quasiperiodic environment, as was shown for example by Passaro *et al.*,¹⁷ who showed that the diffusion exponent for spreading of a wave packet increased in a disordered two dimensional tiling compared to the nonrandom tiling.

Other interesting properties of the magnetic model are related to the spatial behavior of the spin excitations. The magnons have an extended character at low energies, with possibly critical states, as shown in Figure 3. The wave function for the energy eigen state shown has a central peak, and lesser peaks at sites situated at large distances. This type of wave function leads to spin-spin correlations that are very large for different subsets of sites, depending on the energy.¹⁸ Such effects would be observable for quasiperiodic magnetic layer of atoms, a difficult but in principle reachable experimental goal! The results obtained for our spin model lead us to speculate that nonlocal effects and quantum entanglement features should be particularly interesting in these systems.

DISCUSSION AND CONCLUSIONS

Where quasicrystals are concerned, theory has lagged behind experiment in terms of explaining the observed electrical, thermal and magnetic properties. This is due to the fact that the available theoretical tools developed for periodic crystals are either not valid or else not easily

applicable in these aperiodic materials. Simulations of quasicrystals are computationally intensive, and hence results are obtained for rather small system sizes, depending on the available memory and time constraints. On the other hand, simple models such as those outlined in this paper predict that quasicrystals may possess highly unusual and distinctive physical properties that are not shared by crystalline or amorphous materials. The experimental realization of such simplified models is difficult but not impossible, in view of the recent achievements in the deposition of atoms on quasiperiodic surfaces to form two dimensional structures. Further theoretical and experimental progress is clearly needed in order to understand the properties of this class of materials.

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SAŽETAK

Otvorena teorijska pitanja u fizici aperiodičnih sistema

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Kvazikristali su podstakli istraživanje u velikom broju disciplina. Matematičari, fizičari, kemičari, metalurzi i ostali našli su u njima plodno tlo za nove uvide i otkrića. Unutar četvrt stoljeća proteklog od objavljivanja eksperimentalnih rezultata za kvaziperiodičnu Al-Mn leguru proizvedene su i proučene mnoge različite vrste kvaziperiodičnih slitina. Fizikalna svojstva kvazikristala nisu ništa manje interesantna od njihovih neobičnih strukturnih svojstava koja su dovela do njihovog otkrića 1984 godine. U ovom radu dan je pregled svojstava koja karakteriziraju kvazikristale, ukratko razmatrano je nekoliko tipova do sada postavljenih i ponuđenih teorija, te opisana neka nova svojstva koja bi valjalo detaljnije eksperimentalno istražiti.