

Magnetic states induced by electron-electron interactions in a plane quasiperiodic tiling

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We consider the Hubbard model for electrons in a two-dimensional quasiperiodic tiling using the Hartree-Fock approximation. Numerical solutions are obtained for the first three square approximants of the perfect octagonal tiling. At half-filling, the magnetic state is antiferromagnetic. We calculate the distributions of local magnetizations and their dependence on the local environments as the on site repulsion parameter U is varied. The inflation symmetry of quasicrystals results in a corresponding inflation symmetry of the magnetic configurations when passing from one tiling to the next in the progression towards the infinite quasicrystal. [S0163-1829(97)00413-X]

Experimental studies of magnetism in quasicrystal-forming alloys have shown that, for a given composition, the magnetic structure of quasicrystalline, amorphous, and crystalline phases were different. The earliest studies, carried out in Al-Mn-Si alloys,¹ found that the quasicrystalline samples had a small proportion of magnetic atoms and appeared to undergo a spin-glass transition. The crystalline phase of similar composition was, on the contrary, nonmagnetic, and this difference stimulated interest in magnetic properties of quasicrystalline media. The proportion of moment-carrying atoms was difficult to estimate, in the absence of any information as to the values of the moments and as to the magnetic entities (possibly clusters of atoms). Efforts were made nevertheless to extract such information from data obtained in NMR, Mössbauer, and susceptibility measurements.² The early quasicrystals were relatively disordered, and could not be improved by annealing, so that the role of disorder was difficult to disentangle from that of quasiperiodicity. Since then, it has become possible to study quasicrystals of far better structural quality, which are thermodynamically stable, in contrast to their predecessors. Recently, results were reported for a “good” quasicrystal, Al-Pd-Mn.³ Moments were observed to exist at high temperature, with a marked increase in value at the transition to the molten state. Again, the values and the distribution of magnetic moments remain to be determined.

Motivated by this experimental situation, as well as the intrinsic interest of a quasiperiodic Hubbard model, we have chosen to investigate the influence of quasiperiodic order in a *single* orbital case. We consider the octagonal tiling, the “fruit fly” of calculations on quasiperiodic systems. It is a two-dimensional (2D) analog of the 3D Penrose tiling often encountered in models of real three-dimensional quasicrystals and used to describe certain two-dimensional quasicrystals exhibiting eightfold symmetry in their diffraction spectrum. The calculations were carried out for the first three square approximants⁴ of the octagonal tiling, the perfect quasicrystal being obtained in the limit of infinite size. These square pieces are then periodically continued in the plane. The tight-binding Hamiltonian matrix is given by

$$H = \sum_{i,j,s} t_{ij} a_{is}^\dagger a_{js} + U \sum_i n_{i\uparrow} n_{i\downarrow},$$

where a_{is} is the fermion destruction operator on site i with spin projection s . The energy scale is chosen so that t_{ij} is one if the sites are one tile length apart, and zero otherwise, and U is the on-site repulsion strength, multiplying the up- and down-spin densities. In the Hartree-Fock approximation this second term is approximated by $U \sum_\sigma \langle n_{i\sigma} \rangle n_{i-\sigma}$, and the resulting Hamiltonian matrix can then be diagonalized numerically. The iterative method used consists of diagonalizing the Hartree-Fock Hamiltonian matrix for an arbitrary initial distribution of electrons, finding the lowest energy state, computing the Hartree-Fock interaction for the new configuration and continuing until a self-consistent solution is attained. As can be seen from our introduction of a quantization axis, rotational symmetry has been explicitly broken, and a collinear structure is thus assumed in this calculation. However, one can do a more general calculation without this choice of parametrization — indeed for fillings away from the special values considered here, it is probably essential to allow for noncollinear structures (see last section).

The model presented here is a precursor to models containing both s and d orbitals, which would permit actual local moments as opposed to the itinerant state of local magnetizations shown further below. The case of an alloy material would then be described by taking a certain number of sites to possess d and s orbitals. One could then consider the situations where the d orbitals are randomly placed (the melt), or placed in well-defined sites, to investigate the corresponding moments. Similar questions have been considered previously from an *ab initio* standpoint in the melt,⁵ for crystalline Hume-Rothery alloys,⁶ and for small clusters.⁷ Those studies however include the full complexity of the local atomic structure and therefore are limited to rather small approximant phases. On the contrary, the relative simplicity of the Hubbard model allows us to study bigger approximants and therefore to concentrate on the effects of long-range quasicrystalline order on magnetism without being sidetracked by the effects of the local atomic and electronic structure.

In Ref. 8 the octagonal tiling was studied in the paramagnetic phase. Within the random-phase approximation (RPA), a magnetic transition was found to occur for a critical value U_c where U_c depends on the Fermi energy. The values of the $U_c(E_F)$ fluctuate strongly when E_F is varied, reflecting the

huge fluctuations of the density of states of the noninteracting model. More interesting was the observation which are the sites “most susceptible” to going magnetic. The answer depends on the position of the Fermi level in the band, which is symmetric for the bipartite case that we have studied here. For E_F near the band edges, the sites of high coordination number have the strongest divergence of susceptibility at the transition. (There exist six possible coordination numbers ranging from 3 to 8.) For $E_F \approx 0$, i.e., near the band center, on the other hand, it is the *low* coordination sites that are the most strongly susceptible at the U -driven magnetic transition. Therefore it appears natural to expect that the new magnetic state that we shall discuss in this paper will have big “moments” (hereafter we will use this term to denote the value of the local magnetization density) associated with the high, or low coordination numbers, depending on the value of the filling. This is indeed found, as Figs. 1 and 5 illustrate below.

We now take up the case of half-filling: one electron per site, for tilings of size 41, 239, and 1393 sites. (The actual sizes used in our calculations are four times as large, as we have diagonalized 2×2 samples of each approximant, to avoid distortions due to frustrating boundary conditions.) Since each of the square approximants has in addition a mirror plane, the number of nonequivalent sites is in fact smaller than the numbers cited above. Concerning numerical difficulties of convergence to the correct ground state: if one iterates using an arbitrary set of initial values of the on-site moments, one finds several metastable solutions when U is large. To overcome this problem, we have used the fact that the sign of the moments is alternating on first-neighbor sites, in making up initial conditions that give rapid convergence to the ground state. For arbitrary values of the filling, where the ground state is unknown this method cannot be used.

At half-filling the nonmagnetic state obtained for $U=0$ gives way for finite values of U to an “antiferromagnetic” state where moments on adjacent sites (one hop distance apart) are antiparallel. This antiferromagnetic ordering in this Hartree-Fock approach is to be expected⁹ because the tiling is bipartite — the sites and their links can be obtained by projecting a four-dimensional cubic lattice down into the plane. That the occupancy of each site is exactly one (and equal on all sites, for half-filling only) is also easy to see. This is another consequence of the bipartite character of the tiling, which ensures that not only the total density of states but each *local* on-site density of states is perfectly symmetric about the band center (see plots in Ref. 8). For this case of one particle per site, one can formulate the usual argument for minimizing the total energy in the large- U limit, to obtain an effective antiferromagnetic Heisenberg model with an energy of exchange proportional to $1/U$.

The on-site moments are distributed in a range of values. Figures 1(a) and 1(b) illustrate the distribution of moments, for $U=0.5$, for the two bigger tilings. The radius of the circle at each vertex of the tilings is proportional to the size of the moment developed on that vertex, and the signs of the magnetization are indicated by the color of the spots. Figure 1(c) shows the vertices of the smallest approximant, from which the two successive tilings can be obtained by one inflation/two inflations, respectively. Note that the rings of moments of Fig. 1(b) are in one-to-one correspondence with

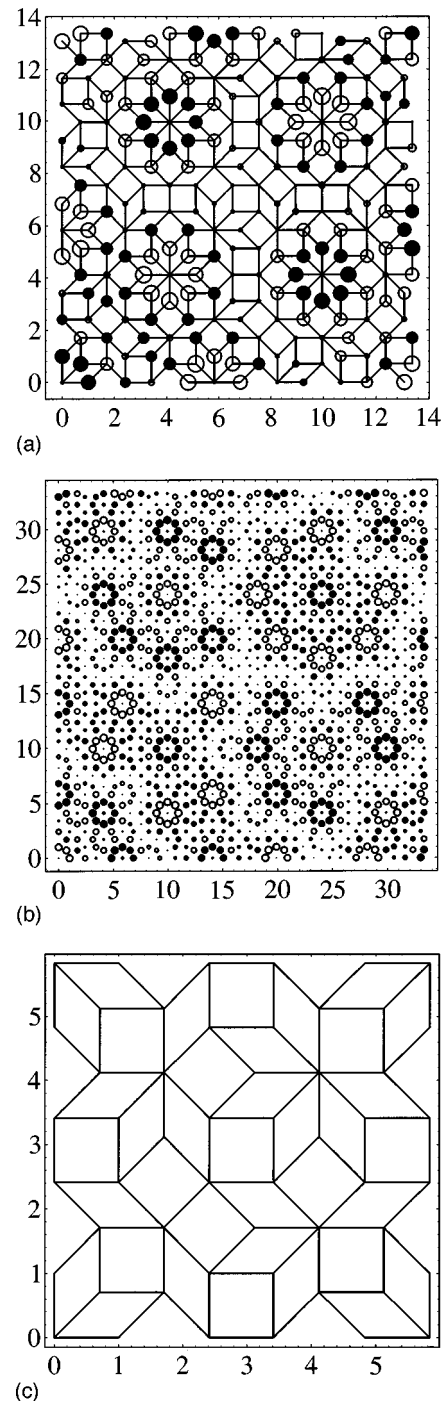


FIG. 1. (a) 239 site approximant tiling showing moments at half-filling. Circle radii correspond to moment size, filled/empty circles correspond to positive/negative moments. (b) 1393 site tiling (links between sites are not shown) for half-filling. Note the similarity with (a). (c) The first tiling of the series, from which the other two can be deduced. Note the position of the rings of moments of (b) coinciding with the vertices of this tiling.

the vertices of Fig. 1(c). This is related to the fact that after two inflations *all* vertices become the centers of eightfold rings. Thus, the geometrical construction relating successive approximants has its consequence for the magnetic structure, which is also self-similar.¹⁰ We note that the biggest circles, or largest moments, correspond to sites of coordination 3 and

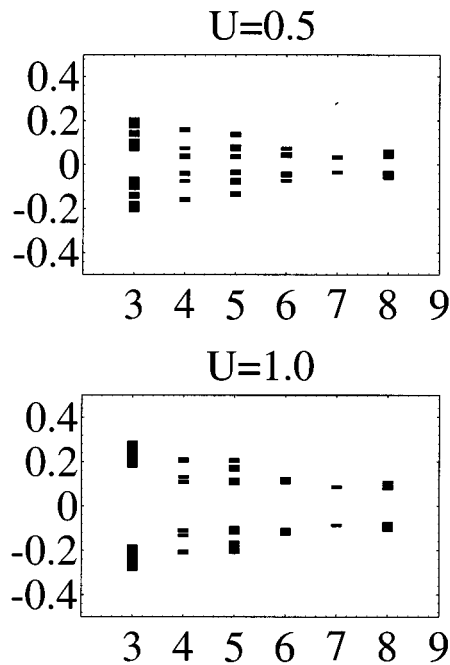


FIG. 2. Values of the on-site moments (positive and negative) plotted against the site coordination number for two different U .

4, while the high coordination sites have small values. Figure 2 shows the distribution of moments grouped according to the coordination number z . The typical (or average) moment is small for the large z sites, while the average moment is largest for the $z=3$ sites. Increasing the value of U can be seen to shift the values further towards the maximum possible values of ± 1 , while at the same time reducing the width of the distribution of the values. Figure 2 also shows an asymmetry between positive and negative moments, a feature more easily seen in the detailed distribution of moments (below). This is due to the finite size of the tilings, and one expects this asymmetry to be reduced as tilings are increased in size, to reach the quasicrystalline limit.

In the limit of large U , the moments on all the sites tend to the value of 1, which is the *exact* occupancy of each site for *any* value of U . Figure 3 shows the approach to this asymptotic value for two different sites chosen at random. The variation of each moment as a function of U appears to be site dependent, and we have not found a compact way to characterize these variations. Turning now to the details of the distribution of moments, we have shown as an example,

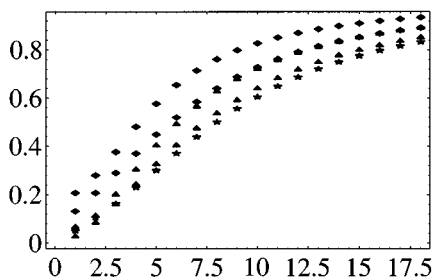


FIG. 3. Plot of moment versus U for a selection of $z=3$ sites (arbitrary).

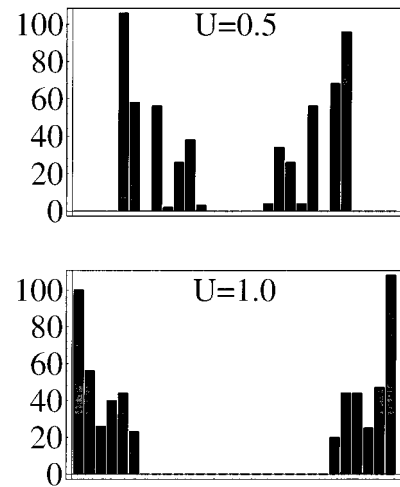


FIG. 4. Histogram of moment values at given values of U for the sites of coordination number $z=3$.

the histogram of moments for sites of $z=3$ in Fig. 4. The distributions show a structure of peaks within peaks. A distribution of values is of course to be expected, this being a reflection of the fact that the local density of states is not identical for two sites of the same coordination number, because of the differences in the background. The peak structure arises because in a quasicrystal one can always find a set of sites that have the same environment out to an arbitrarily large distance. One sees that the distribution of moments shrinks in width as U gets larger, and the center of gravity shifts to larger values of the moment. It is tempting to compare the histograms with the plots in Ref. 5 for the case of Mn in a disordered background of Al. In that work, the authors study the moments developed on Mn atoms in a variety of environments in liquid Al-Mn. There, a “bias” towards higher values of the moment was remarked. This *may* be related to the fact that there are more sites of small coordination (for our octagonal tiling, as an example, the density of $z=3$ sites is about 40%, while that of $z=8$ sites is about 3%).

For small fillings, the Hartree-Fock ground state is ferromagnetic. The occupancy ($n_{\uparrow} + n_{\downarrow}$) of sites is no longer uni-

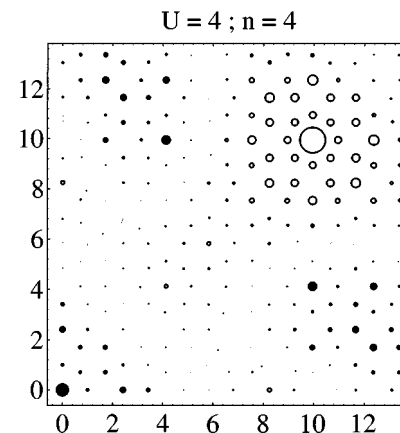


FIG. 5. Magnetizations on the 239 site tiling showing the ferromagnetic state for small filling (four electrons).

form as for half-filling but depends on the coordination number as well as on U . Figure 5 shows the second approximant (containing 239 sites), with circles of diameter corresponding to the value of the moment at that site. A domain wall separating the positive (filled circles) from negative (open circles), appears, created by our imposition of a zero magnetization initial condition. The reflection symmetry along the diagonal of our tiling imposes the orientation of the domain wall.

Finally, we end with some remarks on what could be expected for other values of the filling. We have found that convergence is very poor and trapping in metastable states occurs. One might expect the magnetic state to consist of a mix of negative and positive moments, or more likely, the moments may not be collinear. In this case, taking a fully spin rotationally invariant RPA Hamiltonian could not only allow for a reasonable description of such ground states, but in addition provide a means to overcome the problem of trapping in metastable states, since now there will be soft modes in the transverse direction that will allow the spins to move out of a local minimum of energy. The numerical cal-

culations are bigger, as all spin orientations must now be allowed for, and a new set of calculations is presently underway to examine the magnetic state for arbitrary fillings. One particular case that will be interesting to examine is that of filling the band up to the pseudogap (at $E \approx \pm 2$). This may be closest to the experimental case, where a variety of measurements as well as numerical calculations on “realistic” models have shown that E_F lies in a local minimum of the density of states. The pseudogap at the Fermi energy observed (measured/calculated E_F being a third to a fifth of the expected free-electron values¹¹) in quasicrystalline and nearby crystalline alloys is thought¹² to be due to a Hume-Rothery-Jones mechanism.¹³ The corresponding band-filling for our octagonal tiling model is about 30% (electrons or holes). A study of the magnetic state for this case will be a further step in the understanding of the magnetism of quasicrystalline magnetism.

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