Density of states and magnetic susceptibilities on the octagonal tiling

A. Jagannathan

Laboratoire de Physique des Solides [9], Université Paris-Sud, 91405 Orsay, France

Abstract

We study electronic properties as a function of the six types of local environments found in the octagonal tiling. The density of states has six characteristic forms, although the detailed structure differs from site to site since no two sites are equivalent in a quasiperiodic tiling. We present the site-dependent magnetic susceptibility of electrons on this tiling, which also has six characteristic dependences. We show the existence of a non-local spin susceptibility, which decays with the square of the distance between sites and is the quasiperiodic version of Ruderman-Kittel oscillations. These results are obtained for a tight-binding Hamiltonian with pure hopping. Finally, we investigate the formation of local magnetic moments when electron-electron interactions are included.

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This study of a tight-binding model on the two-dimensional quasiperiodic tiling with eight-fold symmetry, the octagonal tiling, is aimed at understanding electronic properties in real space. There is not much known about quasiperiodic Hamiltonians in two and higher dimensions. To date, studies on quasiperiodic tight-binding models have focused on the form of the density of states, on the response of energy levels to changes of boundary condition (which gives a measure of the degree of localization of the wavefunctions), and on the conductance of such tilings (reviewed in [1]). These studies are important for understanding experiments on transport phenomena, and for resolving whether quasicrystals are intrinsically metals or insulators. However the properties studied are site-averaged properties, and are characteristic of the tiling as a whole. Quasicrystals are interesting for their local properties as well, since they allow local
atomic environments which are different from those found in periodic ordered structures. One indication of site-dependent effects is given by nuclear magnetic resonance and Mössbauer experiments \[\text{\cite{2}}\] which find that in magnetic quasicrystals, the value of the local magnetic moment on a site appears to vary with the type of site. There are no theoretical calculations to date that predict the distribution of magnetic moments of ions on quasiperiodic structures. This motivates our present study of local, site-dependent electronic properties in quasicrystals.

We begin with calculating the local density of states, within a non-interacting tight-binding model, for each of the six local environments on the octagonal tiling. Next we present the onsite magnetic susceptibility of conduction electrons on this tiling. This is proportional to the spin polarization density at the site in question when an external magnetic field is applied. Another interesting quantity calculated is the nonlocal susceptibility, which tells how two magnetic moments interact with each other as a function of their positions within the quasicrystal. This changes sign as a function of the distance, as in a crystalline medium, the quasicrystalline version of Ruderman-Kittel oscillations. Finally, we present results on the magnetic instability induced by electron-electron interactions. Upon adding an onsite repulsion $U$ local moments appear on certain sites depending on the type of site and the position of the Fermi level.

This study hopes as well to add to the scant intuition existing about physical properties of quasiperiodic systems, which lack models, even phenomenological, of their own. It is not the intent here to produce a realistic band structure for a quasicrystal – for which certainly far more complex calculations using realistic parameters for atomic orbitals in binary and ternary alloys have been carried out \[\text{\cite{3}}\]. Instead, we will consider possibly the simplest of models, whose properties we hope will not be overly model-specific. The octagonal tiling (which is infrequently encountered in real materials) is appealing in that it is the two-dimensional cousin of the 3d Penrose tilings used to model a large class of quasicrystals, and is much closer mathematically to these than is the 2d Penrose tiling. Thus we hope that much of the discussion that follows will be carried over to three dimensions.

The approximants of the octagonal tiling are generated by the projection method described in \[\text{\cite{4}}\], which yields square pieces of the infinite tiling, and allows for their periodic repetition in the x-y plane. These approximants are obtained by taking successive rational approximants of $\lambda = \sqrt{2}$, setting it to equal $O_k/O_{k+1}$ where $1, 2, 5, 12...O_k...$ is the so-called Octonacci sequence \[\text{\cite{4}}\]. The number of sites in the square approximants depend on these Octonacci numbers as well, and is $41, 239, 1393...$ etc for $k=1,2,3...$

The tightbinding model chosen in this work takes a single hopping matrix element $t=1$ between sites connected by a bond in Fig.1. The six types of site have been labeled in the figure. This Hamiltonian
possesses a symmetry $t \rightarrow -t$ (since the structure shown is bipartite) and the energy band is thus symmetric about zero. This is no longer the case when hopping is allowed across the short diagonals of the rhombus, for instance, or as in the study of vibrational spectra on these lattices, when diagonal terms appear the Hamiltonian.

We obtain first the local density of states (LDOS), using a continued fraction expansion and recursion method [5] for the Green's functions. The continued fraction is summed to infinite order, in an approximation used with good results in crystals and amorphous solids (reviewed in [6]) and seen to yield satisfactory results in our present case. The form of the LDOS depends on the site coordination number for its overall shape. Superposed on this is a "fine structure" which varies from site to site, and shows the intrinsically multifractal scaling property that is associated with the spectra of quasiperiodic Hamiltonians, whether for the 1d Fibonacci chain [7] or the vibrational spectrum on the octagonal tiling [8]. The present work is not concerned so much with the fractal properties of the spectrum [8] and of the associated spiky behavior of the susceptibilities, but rather with extracting systematic variations obtained, statistically speaking, for sites of a given local environment.

The continued fraction representation of the Green’s function $G_{ii}(z)$ corresponding to a given site $i$ of the approximant can be written

$$G_{ii}(z) = \frac{1}{z - \frac{b_1}{z - \frac{b_2}{z - \frac{b_3}{z - \ldots}}}}$$

(1)

where the parameters $b_n$ are the offdiagonal matrix elements in the tridiagonalized form of the operator $(z - H)^{-1}$ (the diagonal elements are zero due to the symmetric form of the energy spectrum of the Hamiltonian). To calculate the $b_n$ corresponding to a given site, we have shifted that site to the center of the square tiling, and by repeatedly applying the Hamiltonian to the original basis vector consisting of that site, generated successive basis vectors in the tridiagonal basis, along with the sequence of parameters $b_n$. This procedure yields a finite number of the parameters in the continued fraction in Eq.1, as the procedure is stopped upon reaching the boundary. The first twenty $b_n$ have been calculated for the 1393 site tiling in this manner. One may truncate the continued fraction and evaluate the density of states. A better approach is possible in view of the fact that there are no gaps in the spectrum [10]. In this situation it is easy to show that if one replaces the exact $b_n$ by a fixed value $b_\infty$ the relation $b_\infty = \Delta^2/16$ holds (where $\Delta = 2E_{max}$ is the band width). The band width can be inferred from $b_\infty$ and vice versa, but of course neither is known apriori [11]. In practice, one can estimate $b_\infty$ by calculating the local density of states $\rho_i(E) = -\lim_{\delta \to 0} \text{Im}G_{ii}(E + i\delta)/\pi$ by summing the infinite continued fraction and requiring
that $\rho$ be normalized to unity. This allows the determination of a minimum value for $b_\infty$. Setting an upper limit on it is a more delicate affair and requires inspecting the calculated $\rho_i$ for spurious oscillations induced by a too-high value of $b_\infty$.

The six local densities of states obtained after averaging over all sites of a given coordination are shown in Fig.2. The two curves correspond to the LDOS obtained for the 239 site approximant, with eight nontrivial $b_n$ in the continued fraction, and for 1393 sites with 20 nontrivial $b_n$ and, clearly, more fine structure. This is to be expected because replacing the exact $b_n$ by $b_\infty$ corresponds to replacing the true quasicrystalline environment by some averaged effective medium. Retaining 20 exact coefficients in the continued fraction yields the exact quasicrystal up to a greater distance, hence to a finer energy resolution. The tendency is for A and B sites to have their greatest weight at the band edges, while the E and F sites are most active in states at the band center. Not shown are the individual LDOS for each particular site. These differ from the averaged curves in that sharp spikes appear superimposed on the overall form, in ”random” positions along the energy axis. We note also that that the density of states thus found is in good accord with that determined by numerical diagonalization.

The point to be stressed about these LDOS and their smoothed forms (with increasingly wriggly shapes as one calculates further $b_n$ exactly in the continued fraction) is not that there are only six different kinds of sites in the quasicrystal (which naturally is quite false), but that local environments are helpful in understanding certain properties in a statistical sense. Thus, one may expect to find that A sites have in general small amplitudes on mid-band wavefunctions. In fact ”localized” or ”confined” states of zero energy can be constructed readily as for the Penrose tiling [12], and lead to the enormous degeneracy found at $E=0$. These are the wavefunctions that are particularly concentrated on the F sites surrounding each of the A sites, leading to the peak that one finds in the F site LDOS at $E=0$. Compared to the density of states calculated by diagonalizing the Hamiltonian, the smoothed version represented by our calculation is preferable on physical grounds for two reasons: firstly, the presence of disorder in a real quasicrystal will probably act to smoothen the jagged edges in the density of states of a perfect quasicrystal. Finite temperature will also make it impossible to see the selfsimilar structure at very small energy scales. Thus it makes sense to look at smoothed out functions. Second, the boundary conditions imposed when diagonalizing the Hamiltonian always frustrate the tiling, and thus distort the energy spectrum, leading for example to spurious peaks in the onsite susceptibility (see below), if one is not careful.
Now we turn to the calculations of electron spin susceptibility in this model, using the relation

\[ \chi_{ij}(E_F) = \frac{1}{\pi} \int dE \text{Im}[G_{ij}(E)G_{ji}(E)] \text{sgn}(E_F - E) \]  

(2)

where \( N \) is the number of sites, and \( i \) and \( j \) site indices. This is the standard formula for spin susceptibility written in real space coordinates. \( \chi_{ij} \) gives the spin polarization at site \( i \) induced by a small magnetic field at site \( j \). For the onsite susceptibility \( \chi_{ii}(E_F) \), we have used, first, Eq[1] of the Green’s function, which yields a smoothed function similarly to the LDOS in Fig.2. Second, we have calculated it directly by finding the eigenvectors of the Hamiltonian, using mixed boundary conditions, for 41, 239 and 1393 sites, and using a discrete version of Eq[2]. The two methods yield the same envelope curves for the local susceptibilities and are shown in Figs.3 for each site type (averaged over sites of a given type). The susceptibility is roughly flat for A and B sites, while the F sites are ”more susceptible” for \( E_F \) close to zero.

Next we examine the nonlocal susceptibilities \( \chi_{ij}(E_F) \) as a function of the distance of site \( j \) from a given site \( i \). These oscillate, although not with a defined period as do the RKKY oscillations in a crystal. However the number of zero crossings of the susceptibility increases with the Fermi energy, reaching a maximum at \( E_F = 0 \). Figs.4a) and b) show the oscillations for two different values of \( E_F \), over a range of distance that corresponds to half the lattice size of the 1393 site tiling. Also indicated is the \( 1/r_{ij}^2 \) envelope of these ”quasi-RKKY-oscillations” – which are an amusing demonstration of metallic character in the quasicrystal.

The effect of an onsite Coulomb repulsion \( U \) is now considered. We have treated this in the random phase approximation (RPA), and calculated numerically the site susceptibilities given by the equation

\[ \chi_{RPA} = \chi_0(1 - U\chi_0)^{-1} \]

(3)

\( \chi_{RPA} \) being the RPA susceptibility matrix, and \( \chi_0 \) the susceptibility matrix of Eq[2]. This equation represents the real space version of the Stoner equation [13] leading to itinerant ferromagnetism in metals. A magnetic instability is signalled when an eigenvalue of this matrix diverges. This occurs when \( U = \lambda_{max}^{-1} = U_c \) where we have determined \( U_c(E_F) \) numerically. Although it does not vary smoothly, \( U_c \) is about 0.5 at the band center, increasing to values of the order of 1 as the band edge is approached. The sites that are ”most susceptible” to going magnetic are found by studying the eigenvectors of \( \chi_0 \) corresponding to \( \lambda_{max} \). These are, speaking statistically, almost exclusively the F sites when \( E_F \) is close to zero (about 25 % of these sites), and include all of the A and B sites when \( E_F \) is near the band edges.
A more detailed look at the distribution of susceptibilities will be given elsewhere. For completeness, we may mention that Deng and Carlsson [14] have calculated magnetic moments of Mn atoms in a cluster of AlMnSi, finding two types of site carry no moment, while a third type of site does. This last case is distinguished by the presence of a nearest neighbor Mn. This shows perhaps the importance of including such pairwise interactions if one wishes to study the distribution of moments (which is outside the scope of the present paper).

Thus, numerical diagonalization and the continued fraction technique yield similar results, showing the applicability of the latter method to this quasiperiodic system, despite the singular features in the energy spectrum. The smoothing features of the latter method are in fact useful in helping identify overall features which could be obscured by the irregular spikes of the exact solution (which depends on tiling size and boundary conditions). There is seen to be a progression in the local density of states and the susceptibility of individual sites, and A and B sites behave quite differently from E and F sites, while the remaining two have in-between character. Coulomb interactions enhance the magnetic susceptibility of a given class of sites, depending on Fermi energy, and could lead to the observed NMR observation of nonhomogeneous values of the local moment. Finally, the mechanism that is responsible for RKKY or Friedel oscillations in crystalline metals gives rise, in the quasicrystal, to changes of sign of the susceptibility as a function of distance, the frequency of which is determined by $E_F$. For nearest neighbor sites, the "quasi-RKKY" interaction is initially positive, but goes negative upon raising the Fermi level, staying negative all the way until the upper edge of the band. For sites that are several bond lengths apart, on the other hand, $\chi_{ij}(E_F)$ is a chaotically behaved function of the Fermi energy.

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References

[*] Laboratoire associé au CNRS.

[1] Takeo Fujiwara and Hirokazu Tsumetsugu Quasicrystals: The State of the Art, Eds.D. P. Divincenzo and P. J. Steinhart World Scientific, 1991
A rather simple calculation gives one a first estimate of the band width: the infinite system of coupled equations contained in the tight-binding eigenvalue equation $H|\Psi_0\rangle = E_{max}|\Psi_0\rangle$ ($\Psi_0$ everywhere positive) is truncated to a small set of approximate equations written for the six local environments, which can be solved for the energy $E_{max}$ analytically. The value (necessarily approximate) obtained agrees with the result of numerical diagonalization $\approx 4.2$. 

[12] Mahito Kohmoto and Bill Sutherland Phys. Rev. B 34, 3849 (1986)

[13] R.M. White Quantum Theory of Magnetism, Springer Series in Solid State Sciences, Ed. P. Fulde vol.32

[14] A. E. Carlsson and Rob Phillips Quasicrystals: The State of the Art, Eds.D. P. Divincenzo and P. J. Steinhardt World Scientific, 1991
Figure Captions

1. 239-site tiling with local environments labeled.

2. Local density of states. Two curves are shown for each site type, corresponding to taking 8 and 20 non-equal $b_n$ in Eq.1 and taking $b_\infty = 4.48$.

3. $\chi_{ii}$ versus $E_F$. The continuous curves show the result using continued fractions, while the points are the result of numerical diagonalization, for the 239 site tiling.

4. Quasi-RKKY oscillations along a path of 19 sites (running index $j$), taking $i$ to be an A site on the 1393 site tiling. a) shows the slower oscillations at the lower end of the energy spectrum and b) the rapid oscillations near the band center. Dashed lines indicate $1/r^2$ decay. The continuous lines are simply a guide for the eye.