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Screening of a magnetic impurity in a quasicrystal

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Abstract. The problem of screening of local moments in a metallic environment is revisited, in the context of quasiperiodic solids. The motivation for this study comes from experimental studies showing that low temperature thermodynamic properties are strongly anomalous in the quasicrystalline phase, but less or not anomalous in the related approximant phase. A model for a magnetic impurity in a quasiperiodic environment is introduced and solved numerically. Arguments are given for the existence of a singular distribution of Kondo temperatures in the quasicrystal, thereby providing a possible explanation for the experimental observations.

1. Introduction

The discovery of a heavy fermion quasicrystal, YbAuAl, with anomalous thermodynamic properties at low temperature [1, 2] raised fundamental questions regarding the electronic states in metallic quasicrystals, and of the role played by electron-electron interactions in these materials. Already at the single particle level, numerous studies in a variety of models [3] have shown that quasiperiodicity has strong consequences for the nature of electronic states. Their spatial characteristics are very different from those of the usual Bloch states of a periodic system, bearing, instead, closer resemblances to states at the critical point of the metal-insulator transition in disordered metals. The local and global density of states have strong fluctuations, with pseudogaps of varying amplitudes. These fluctuations in energy and in position can be expected to have experimentally detectable consequences. One consequence concerns transport properties, and indeed experiments show quasicrystals to be very poor electrical conductors. Theoretical calculations of the mean square displacement and the distribution of band velocities in tight binding models [4] show that these systems indeed cannot be described by a standard Boltzmann type theory.

A good way to probe electronic properties in real space consists of introducing impurities, and studying the effects of the perturbation. For noninteracting electrons this leads to indirect exchange interactions between magnetic impurities, and can result in a complex magnetic order [5]. These result from the nontrivial, spatially inhomogeneous response functions for an electron in the quasicrystal. This type of spatial fluctuation leads, for the many body interacting system, to novel behaviors as well. Thus, as a mean-field treatment of the Hubbard model showed [6], inhomogeneous ferromagnetic or antiferromagnetic states can appear for arbitrarily small values of the Hubbard parameter U.

Here we consider the screening of a local moment due to hybridization between a single localized state and the band of conduction electrons in a quasicrystal. In a periodic metallic solid, antiferromagnetic coupling between the conduction electron gas and a localized spin-1/2 leads to the Kondo effect, namely the screening of the moment at low temperatures. In the quasicrystal,

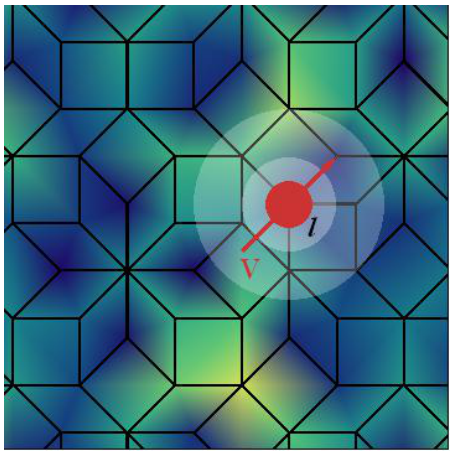

Figure 1.
Schematic drawing of an impurity atom (red) bearing a local moment, located on a given site of a quasiperiodic tiling. The background colors depict the values of the local conduction electron density of states.

one can expect that the screening will be strongly dependent on spatial position of the impurity and on the Fermi energy of the system. This is effectively the case, as was found in [7] for a specific single impurity model. Fig.1 gives a schematic view of the model. An impurity in the quasicrystal background sees a fixed certain local environment and a corresponding local density of states (LDOS). To model the quasicrystalline background, we consider the tight-binding approximation in a quasiperiodic tiling in d dimensions (d=2 or 3), simplified by assuming a single-orbital per site and hopping terms only. The magnetic impurity is described in terms of the infinite U Anderson model, where a single f-orbital is located at a specific site of the quasicrystal. A hybridization term allows for the hopping of electrons between the conduction band and the f-level.

Sec.2 presents the model and Sec.3 the main results for the Kondo temperatures. In Sec.4 the mechanism underlying this form of the distribution is discussed. In Sec.5 temperature-dependence of the average magnetic susceptibility and relation to experimental data in the YbAuAl compounds are discussed.

2. The model

We define the tight binding Hamiltonian for the conduction electrons on a tiling in d dimensions, with a single orbital on each of the vertices. The particles hop along the edges joining the vertices with a hopping amplitude t, assumed to be the same for all edges. The onsite energies are taken to be uniform and are set henceforth equal to zero. The tight-binding Hamiltonian is then

$$\mathcal{H}_c \;=\; -t \sum_{\langle ij\rangle,\sigma} \left(c^\dagger_{i\sigma} c_{j\sigma} + c^\dagger_{j\sigma} c_{i\sigma} \right). \qquad (1)$$

where $c^\dagger_{i\sigma}$ ($c_{i\sigma}$) is the creation (annihilation) operator of a c-electron at site i with spin σ. The tilings considered for a detailed analysis were square periodic approximants of the Ammann-Beenker (octagonal) tiling in two dimensions (2D) and of the rhombohedral tiling in three dimensions (3D). The most detailed studies were carried out for the 2D system, where it was possible to consider a series of finite size approximants ranging from N = 41 to 8119, where N

is the number of sites in the approximant. The 3D system was studied next and served to check the robustness of our results under change of dimensionality.

The Hamiltonian for the conduction electrons is diagonalized assuming twisted boundary conditions, ie, $\psi(x+L,y+L) = e^{i(k_xL+k_yL)}\psi(x,y)$. The densities of states (DOS) are then averaged over a large number of values of $\vec{k}$. This results in a local smoothing of the DOS, and better stability of the numerical calculations. The results thus obtained for the Kondo temperatures are thus valid in a statistical sense. This procedure is also physically pertinent in that it mimics the experimental systems, where finite size effects due to grain boundaries or defects tend to smear out the fine structure in the electronic structure of the quasicrystal.

The impurity is described by the $U \to \infty$ Anderson impurity model or AIM (a discussion of models for local moments in solids can be found in, for example, [8])

$$\mathcal{H} \;=\; \mathcal{H}_c + E_f \sum_\sigma n_{f\sigma} + V \sum_\sigma \left(f^\dagger_{\ell\sigma} c_{\ell\sigma} + c^\dagger_{\ell\sigma} f_{\ell\sigma} \right). \tag{2}$$

This model describes a band of non-interacting electrons (c-band) which hybridize with a localized f-orbital located at site ℓ. The operator $f^\dagger_{\ell\sigma}$ ($f_{\ell\sigma}$) creates (destroys) an electron with spin σ at the impurity site ℓ. E_f is the f-level energy, measured with respect to the chemical potential μ, and the hybridization V couples the impurity site to the conduction band. The $U \to \infty$ limit imposes the constraint $n_{f\sigma} = f^\dagger_{\ell\sigma} f_{\ell\sigma} \leq 1$

In terms of the parameters of the AIM, the so-called Kondo coupling is defined by $J = 2V^2/|E_f|$. The Kondo regime corresponds to the limit in which the impurity f-orbital is singly occupied and only spin fluctuations are relevant. The Kondo temperature then gives the temperature below which the impurity moment couples strongly to the conduction electrons, and the moment is effectively quenched.

In order to obtain quantitative results, we consider a large-N (N here represents the quantum number of the spin) limit of Eq. 2 where a variational method can be applied [9]. The two variational parameters are Z_ℓ (quasiparticle weights) and $\tilde{\varepsilon}_{f\ell}$ (renormalized f-energy levels). These are site-dependent in the case of a quasicrystal. The f-level occupation is then given by $n_{f\ell} = 1 - Z_\ell$.

3. Solutions in the Kondo limit

The variational calculation involves placing the impurity site in an effective bath depending on the local environment of the site, ℓ. The hybridization functions $\Delta_{f\ell}$, which gives the broadening of the f-level, are related to the local (onsite) conduction electron Green's functions $G_{c\ell}$ by $\Delta_{f\ell}(\omega) = V^2 G_{c,\ell}(\omega)$. The integral equations involving $\Delta_{f\ell}$ and variational parameters are solved numerically, for each of the sites of the tiling. The Kondo limit is achieved when the impurity site is singly occupied, $n_{f\ell} \approx 1$. The Kondo temperature is, in this limit, given by the width of the resonance at the Fermi level, $T^\ell_{\rm K} \equiv Z_\ell {\rm Im}\left[\Delta_{f\ell}(0)\right]$. The distribution of Kondo temperatures $P(T_{\rm K})$ can then be calculated. The calculations are repeated for different values of the chemical potential μ and of the parameters V and E_f.

The results show the result for the distribution $P(T_{\rm K})$ of Kondo temperatures T_K for different locations of the impurity (sample size N=1393, $\mu = -1.8t$, $V = 1.0t$, $E_f - \mu = -3t$). One sees that for small T_K, the distribution follows a power law $P(T_{\rm K}) \propto T_{\rm K}^{\alpha-1}$ where the exponent $\alpha < 1$, ie, there is a singular tail for small T_K. It is seen that the singular behavior is obtained for all the values of μ tested, providing that the Kondo coupling J is small enough. The exponent α is non-universal, and depends on the chemical potential and the value of J. In the next section, we will discuss the possible mechanisms underlying this power law.

The wide spread of values of $T_{\rm K}$ in the Kondo regime is reminiscent of the distributions found in disordered metals in the non-Fermi liquid regime [11]. In contrast to disordered metals,

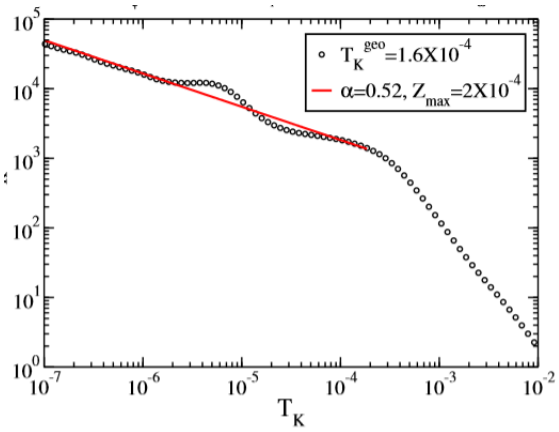

Figure 2.
Distribution function $P(T_K)$ for chemical potential $\mu = 1.8t$ and system size N=1393. The best fit values of the exponent α and of the geometric mean of the Kondo temperatures (defined as in the text, with Z_{max} the upper cutoff in the integral) are given.

however, in the quasicrystal the distribution is *intrinsic*, and does not depend on parameters such as the strength of the disorder. The typical temperature scale in both these situations is measured not by the arithmetic mean, but the geometric mean, $\langle T_K \rangle$, which is defined by

$$\langle T_K \rangle = \exp\left(\int dT_K \log T_K P(T_K) \right) \tag{3}$$

In the computation the integral runs over the interval of T_K in which P satisfies the power law.

4. Analysis of results

The observed distribution of screening temperatures T_K results from a combination of factors. Clearly, the spread of values of T_K is the result of the inhomogeneous local density of states, $\rho_{c\ell}(E) = (-1/\pi) G_{c\ell}(E)$, in the quasicrystal. The form of the LDOS (see Fig.3) can be qualitatively understood as a signature of multifractal "critical" states. Multifractal properties of electronic states are well-understood in one dimension, where the characteristic exponents can be analytically found in some cases [10]. However, in higher dimensions, it has not been possible to obtain a theoretical description of wavefunctions of the standard tight-binding models, except in some special cases.

To probe the mechanism underlying the distributions $P(T_K)$, let us consider how T_K is related to the electronic properties of the tiling. Although there are no exact solutions of the mean field equations, one can make some simplifying assumptions in the Kondo limit, when E_f lies well below the Fermi level, and $n_{f\ell}$ tends to 1. In this limit, the mean field equations yield an expression of the form

$$\begin{aligned} T_{K\ell} &= D \exp\left[-\frac{\pi}{2} \left(\frac{Re[\Delta_{f\ell}(\mu)] + |E_f - \mu|}{Im[\Delta_{f\ell}(\mu)]} \right) \right] \\ &= D \exp\left[-\frac{1}{2J\rho_{c\ell}(\mu)} + \left(\frac{\mu - Re[\Delta_{c\ell}(\mu)]}{Im[\Delta_{c\ell}(\mu)]} \right) \right] \end{aligned} \tag{4}$$

where the hybridization function was eliminated in favor of the real and imaginary parts of the cavity function $\Delta_{c\ell}$, defined in terms of the electron Green's function by $G_{c\ell}(\omega) = (\omega - \Delta_{c\ell})^{-1}$.

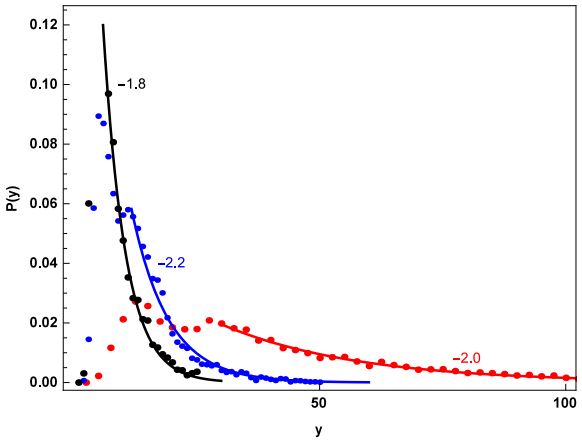

Figure 3.
Distribution functions $W(y)$ of the inverse LDOS y, with fits to an exponential tail for three values of the chemical potential: $\mu = -1.8t$(black), $-2t$(red) and $2.2t$(blue). System size N=1393.

In the Kondo limit, assuming that $J \ll 1$, one can write

$$T_{K\ell}/D = \mathrm{e}^{-\frac{1}{2J}(y_\ell + \mathcal{O}(J))} \tag{5}$$

where the term $\mathcal{O}(J)$ in the exponent can be neglected in the small J limit. The variable $y_\ell = 1/\rho_{c\ell}$ is the inverse LDOS on site ℓ. In this expression for the Kondo temperature, the first term in the exponent dominates for small J, leading to vanishingly small T_K on sites with smallest LDOS. To calculate the distribution for small T_K, we need the distribution function for large values of y_ℓ. These typically correspond to sites where large fluctuations of the real and imaginary parts of the electron self-energy occur for small energy and boundary condition changes. In consequence, as numerical calculations confirm for the three choices of μ shown in Fig. 3, the y_ℓ behave like Poisson-distributed pseudo-random quantities. The distribution of the variable y (computed for finite broadening of levels and averaged over an energy interval of width 2δ where δ is the mean level spacing) is well-approximated by an exponential form $W(y) = e^{-cy}$. The constant c depends on the nature of the wavefunctions, and thus varies strongly with the position of μ, ranging from 0.05 to 0.2, in the figure. One can now deduce from this form the power law for Kondo temperatures, $P(T_\mathrm{K}) \sim T_\mathrm{K}^{\alpha-1}$ where $\alpha = 2Jc$.

The exponents α predicted by this argument are non-universal. The constants c vary significantly when the Fermi level is varied. Their values depend slightly on finite size effects and the energy-broadening introduced in the calculation. The smallest value of c occurs for $\mu = -\pm 2t$ i.e. at the main pseudo-gaps of the pure hopping model, where the distribution of y is widest. The exponents for fixed chemical potential are, secondly, proportional to the coupling J. The numerical results for T_K indeed show these trends [7] for J small.

The asymptotic form of the distribution is valid in the limit of large size, where distributions of the LDOS and the $\Delta_{f\ell}$ are sufficiently wide. For small approximants, the LDOS distribution will not display an exponential tail, and the minimal value of T_K, T_{min}, will not be particularly small. In numerical calculations, the size of the approximant is thus an important limiting factor, and the singular power law dependence is clearly seen only for sufficiently large systems.

5. Discussion and conclusions

The singular (power law) form of $P(T_{\rm K})$ indicates that, for a finite fraction of sites, quenching of the impurity local moments does not occur, down to very low temperatures. This leads to a non-Fermi-liquid (NFL) behavior as we now argue. Recall that $\chi(T)$ of the Fermi liquid is finite as $T \to 0$, while a free moment shows Curie behavior, ie, has a diverging magnetic susceptibility $\chi(T)$. The zero temperature susceptibility $\chi(T = 0)$ in a system with a quenched moment has a finite value, of the order of $T_{\rm K}^{-1}$. These behaviors can be compactly written in terms of an interpolative form of the susceptibility for low T, $\chi(T) \sim (T + T_{\rm K})^{-1}$. In a simple lattice, $T_{\rm K}$ is single valued, whereas in disordered metals – or as we have seen, in a quasicrystal – $T_{\rm K}$ can have different values depending on the local environment.

Let us now consider a quasicrystal with a finite but low concentration of impurities, and let us suppose that one can neglect interactions between them. The experimentally measured susceptibility is the magnetic susceptibility averaged over all sites of the sample, defined by $\overline{\chi}(T) = \int dT_{\rm K} P(T_{\rm K})\chi(T)$. Substituting the form of the distribution for low $T_{\rm K}$, a power law extending down to $T_{\rm K} = 0$ for the infinite quasicrystal, one finds a power law $\overline{\chi}(T) \propto T^{\alpha-1}$, ie, the susceptibility is singular for $\alpha < 1$. Our scenario thus explains the power law of the susceptibility found experimentally as reported in [1, 2]. In this model, the spin contribution to other thermodynamic quantities such as the specific heat should also possess power law singularities. Experiments show however that the singularity of the specific heat is weaker. This could be an indication that impurity-impurity correlations, which were neglected in this simple model, are important.

Small approximants of the quasicrystal should, in contrast, show a Fermi liquid low temperature behavior. For, as the periodic length (and number of local environments) gets smaller, the Kondo temperatures increase, and their lower bound, T_{min}, will rise. For T lower than the value of T_{min}, the susceptibility will thus level off to a constant. This is in accord with experiments which show a nonsingular behavior of the susceptibility in the approximant crystal [1].

To conclude, we have presented an argument for NFL behavior in quasicrystals based on the existence of critical states at the Fermi energy and numerical results that support the scenario. The results are robust under a change of dimension: similar results were found in [7] for a three dimensional tiling. Our model considered properties in the Kondo regime where spin fluctuations are dominant, while theoretical models to study valence fluctuation effects have been investigated elsewhere [12, 13]. More theoretical studies are clearly necessary in order to understand the real-world experimental systems.

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