Absence of Spin Diffusion in Most Random Lattices

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We examine the metallic phase of the positionally disordered Anderson-Hubbard model as a prototype of a strongly correlated, disordered metal. We find that the low-temperature thermodynamics is dominated by spin excitations in rare regions, leading to non-Fermi-liquid behavior as $T \to 0$, characterized by a diverging susceptibility, a specific heat rising faster than T, and a vanishing spin diffusion coefficient. We argue that the results should, in principle, apply throughout the metallic phase of random alloys which undergo a transition, as a function of concentration, to an insulating phase with local moments.

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Recent experiments on weakly metallic samples of uncompensated doped semiconductors (Si:P, Ge:Sb, Si:B) using magnetic [1] and thermodynamic [2] probes have provided strong evidence that highly disordered correlated metals contain localized moments [3–5]. These dominate the magnetic susceptibility and specific heat at low temperatures (< 1 K). Experiments on compensated materials (Si:P;B) show even stronger effects [6].

The idea of local moments existing in disordered systems on the metallic side of the metal-insulator (MI) transition is not new [4]. A recent study [5] of the positionally disordered Hubbard model in three dimensions found local moment instabilities on $\sim 10\%$ of the sites at densities $\sim 25\%$ above the critical density n_c of the MI transition. The issue is whether such moments persist down to low temperatures, so that they are an integral part of the low-energy description, or are quenched (due to coupling to other electrons) at a high-energy scale resulting in (finite) renormalization of Fermi-liquid parameters, but no dramatic consequences.

In this Letter we examine this question in some detail, focusing on the consequences of *quenched randomness*. Our principal result is that at *any* composition in such dirty metallic systems, rare statistical density fluctuations give rise to local moments which are so weakly coupled to the itinerant electron fluid that they cannot be effectively quenched via either the Kondo mechanism or RKKY interactions. These residual moments give rise, in the $T \to 0$ limit, to a divergent magnetic susceptibility $\chi(T)$ and specific-heat coefficient $\gamma(T) \equiv C_v(T)/T$, and a vanishing spin diffusion coefficient. These effects will occur at any composition provided the system undergoes a MI transition as a function of composition to an insulating phase with local moments. Far away from the transition the effects will be very small and only appear at exponentially low temperature, but as the transition is approached the effects should become much larger. This implies that a *minimal* phenomenological description of such a metallic disordered system at low temperatures is not in terms of a single-component Fermi fluid, but must consist of *two* entities—including both the local moments and the itinerant electron fluid. This is in contrast to uniform systems (such as heavy fermions [7]) which form local moments at intermediate temperatures but (barring other instabilities such as superconductivity) exhibit Fermi-liquid behavior as $T \to 0$, characterized by a *finite* χ and γ, albeit with very large [7] renormalization $\sim 10^3$. These differences [5], due to the quenched disorder, are analogous to Griffiths singularities [8] in the disordered phase of classical random magnets, which can dominate the dynamical properties [9], and for quantum systems, the static properties as well [10]. Alternative explanations [11–13] of the experimental observations have been based on renormalization-group treatments of interacting electrons in a disordered medium, but these have focused on long-wavelength rather than localized effects.

For concreteness, we first consider a specific model —the Anderson-Hubbard model with sites positioned randomly in a d-dimensional continuum. Such a model is a rather good caricature of doped semiconductor [14] systems; further, as we shall argue later, most of our arguments can be generalized to systems with other kinds of quenched random disorder. The Hamiltonian is

$$H = \sum_{i\sigma} (\epsilon_i - \mu) n_{i\sigma} + \sum_{ij\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} , \qquad (1)$$

where the sites i are distributed *randomly* in d dimensions with density n and some spread in the on-site energies ϵ_i. The hopping integral $t_{ij} \sim t_0 \exp(-r_{ij}/a)$, where $t_0 \sim U$, the on-site Coulomb repulsion, μ is the chemical potential, and σ the spin index. This model is believed to have a transition from an insulating state to a metallic state for $d > 2$ as the site density n is increased. In $d = 3$, the critical density n_c is expected to be near the Mott criterion [15,16] value ($n_c^{1/3} a \approx \frac{1}{4}$) especially close to half filling, which we primarily consider.

Well above n_c, most of the sites will be coupled to an infinite cluster by hopping elements $t_{ij} \sim U$. Nevertheless, there exist large density fluctuations because of the random distribution of sites. Thus, for example, the density

of isolated sites which have their nearest neighbor at a distance greater than r is given by $n_I(r) \sim n \exp(-c_d \times n r^d)$, with c_d a dimensionality-dependent constant. For sufficiently large r, such strongly isolated sites are occupied by single electrons and because of their weak coupling to the surroundings form local moments [3,5], analogous to d or f levels in the Anderson model [3], whereas the other, well-coupled sites are like conduction electron levels. The exchange coupling J_i of a moment at strongly isolated site i (with a nearest neighbor k at distance r_i) with the rest of the system is of order $J_i \sim t_{ik}^2/u \sim \exp(-2r_i/a)$, which falls off relatively rapidly with r_i. u is an energy involving U and $\epsilon_i - \mu$, which does not depend on r_i for such isolated sites. Such a local moment (Fig. 1), in the absence of J_i, would give a divergent Curie susceptibility $\chi_C(T) \sim \mu^2/T$. However, the coupling to other sites can quench the local moment.

Since quenched moments do not contribute significantly to the susceptibility, the low-energy magnetic behavior is controlled by the density of moments $\rho_u(\epsilon)$ which are *not* quenched at an energy scale above ϵ. Since these moments will only have a Curie susceptibility at temperature T if $\epsilon < T$, the resulting susceptibility is approximately $\chi(T) \sim (\mu^2/T)\rho_u(T)$. The entropy from the unquenched sites gives rise to a contribution to the specific heat $C_v(T) \equiv T\gamma(T) \approx T\, d\rho_u(T)/dT$. One mechanism for quenching local moments is through coupling to the sites which has formed a "Fermi fluid," i.e., via the Kondo effect [17]. This quenching is effective below the Kondo temperature given by [18] $T_K^i \approx E_0 (J_i \rho_F)^{1/2} \times e^{-1/J_i \rho_F}$, where E_0 is of the order of the bandwidth and ρ_F is the density of states at the Fermi level. Note that because of the strong (exponential) dependence of J_i, which in turn depends exponentially on r_i, the Kondo temperature for relatively isolated sites is *extremely* low, as can be explicitly seen by substituting for J_i in T_K^i. If the Kondo effect were the only mechanism for quenching moments, then $\rho_u(T)$ would be simply $n_I(r_T)$ where the size of the isolation hole r_T around sites not quenched to temperature T is given by $T_K(r_T) = T$. This would give

$$\chi(T) \sim (n\mu^2/T) \exp\{-f_d \ln^d[\ln(T_0/T)]\}, \qquad (2)$$

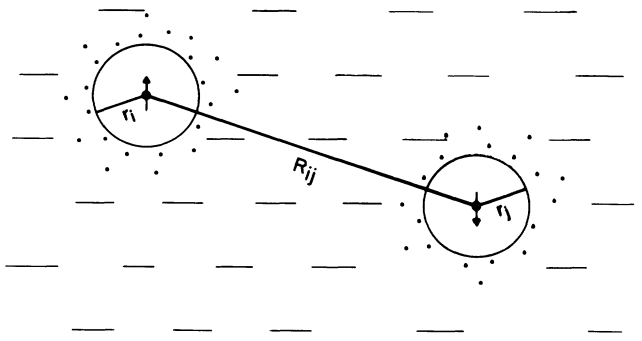

FIG. 1. Two isolated sites i and j with local moments separated by a distance R_{ij} in the midst of a Fermi liquid.

where $T_0 \sim t$, and $f_d = c_d n (a/2)^d$. This $\chi(T)$ diverges at low T in all dimensions, as does $\gamma(T)$ [19].

This arises because, even though the density $n_I(r)$ has a rapid falloff with r, the few sites in the tail of the distribution are too weakly coupled to be quenched effectively via the Kondo effect. Consequently, in the absence of any other quenching mechanism, they would give a divergent susceptibility, and dominate the thermodynamics at low enough temperatures, and the system would not be a Fermi liquid in the conventional sense as $T \to 0$.

In real systems, we must also consider quenching of the local moments due to interactions with other local moments. In the metallic phases of interest, this occurs via the conduction electron mediated RKKY interaction. Between two spins with separation R_{ij} coupled to the conduction electrons via exchanges J_i and J_j (see Fig. 1), the effective RKKY exchange is $K_{ij} \sim J_i J_j \rho_F g_d(k_F \mathbf{R}_i, k_F \mathbf{R}_j)/|R_{ij}|^d$, with g_d an oscillatory function of its arguments, typically varying on a scale of unity, and exhibiting randomness in a dirty metal arising from the behavior of the conduction electron wave functions near the Fermi surface [20]. The RKKY interactions between local moments at large separation ($R_{ij} \gg k_F^{-1}$) are thus randomly ferromagnetic or antiferromagnetic. For spin-$\frac{1}{2}$ local moments, however, the latter have a much stronger effect due to the factor of 3 difference in exchange energy between a singlet and triplet. Since we are interested in a *lower* bound on the susceptibility, we first assume that the RKKY interactions are purely antiferromagnetic with $g_d = 1$, and then return to the more realistic case.

Bhatt and Lee [21] have shown that *exponentially* decaying antiferromagnetic interactions J_{ij} between randomly placed spin-$\frac{1}{2}$ impurities (e.g., insulating Si:P) give rise to a "random singlet" phase with the spins paired into singlets (or occasionally grouped in larger clusters) with a distribution of separations. In the simplest approximation, the most strongly coupled pair of spins are paired, then the strongest coupled pair of the *remaining* spins are paired and the process continued until all the spins are exhausted. The susceptibility is due to spins which are eventually paired with an energy smaller than ϵ, $\rho_u(\epsilon)$, as discussed above. If all the exchanges J_i were equal, this would lead to $\rho_u(\epsilon) \sim \epsilon/J^2\rho_F$, yielding a *finite* susceptibility as in uniform systems.

With the broad spectrum of J's which occurs in our case, the behavior is somewhat more subtle. Most of the spins with largest J, i.e., J_0, will pair with those with comparable J. Some, however, will find a rarer spin with a smaller J, J_1, with which they can bind more strongly than the unpaired J_0 spins. Because of the rarity of the J_1 spins, this will only happen occasionally. (Indeed, the resulting pairing strengths K_{01} when it does occur will usually be comparable to the typical coupling strength K_{11} between J_1 spins.) Thus the J_1 spins will again pair primarily with spins of their own ilk. A good approximation to the number of unpaired spins at temperature T is

thus given by the density $n_I(r_T)$ of those with isolation holes of size r_T such that the RKKY interaction $K_T = J(r_T)^2 \rho_F R_T^{-3}$ between them and similarly isolated spins is of order T. The typical separation between such spins is $R_T \sim [n_I(r_T)]^{-1/d}$. By substituting for $\rho_u(T) \sim n_I(r_T)$ we then obtain a low-temperature susceptibility:

$$\chi(T) \sim [J^2(r_T)\rho_F]^{-1} \sim \exp[C_d \ln^{1/d}(T_0/T)] . \quad (3)$$

This diverges as $T \to 0$ albeit more slowly than any power of T, as does $\gamma(T)$ (up to extra $\ln T$ factors).

Several factors might affect the susceptibility. First, excitation of an already paired singlet of intervening spins can give rise to an effective indirect interaction $\tilde{K}$ between spins, larger than the direct interaction. In the insulating case with exponential decay of K_{ij}, this is an important effect [21], especially in low dimensions. In our case, on the other hand, because of the power-law dependence of K_{ij} on R_{ij}, this can only be an important effect if the unpaired spins of interest are located near the opposite ends of the preformed singlet. This can be shown to occur only rarely and thus it does not significantly affect the results.

A potentially more important effect is due to the ferromagnetic nature of some of the interactions (and effective interactions). If the system remains in a random singlet phase, then these should only enhance the susceptibility via the formation at temperature T of clusters with larger magnetic moments. It is possible, however, that the competition between the ferromagnetic and antiferromagnetic interactions drives the system towards a spin-glass phase with broken spin rotational symmetry at $T=0$. Interactions decaying as $1/R^d$ are believed to be marginal for classical Heisenberg spin glasses, i.e., a positive temperature spin-glass transition ($T_c > 0$) might occur in their presence if it did not do so in their absence [22]. If $T_c > 0$, $\chi(T)$ should exhibit a cusp and then saturate to a finite value at $T=0$. If, however, $T_c = 0$, $\chi(T)$ is likely to be more strongly divergent than Eq. (3), probably a Curie form due to excitations of size of order the T-dependent spin-glass coherence volume [22].

A random singlet phase appears most likely in three dimensions for a spin-$\frac{1}{2}$ system due to the dominance of antiferromagnetic interactions. If so, the susceptibility of Eq. (3) is probably a lower bound, although it may asymptotically be the correct form. But, in any case, the zero-temperature metallic phase will *not* be a "normal" dirty metal with Pauli susceptibility, linear specific heat, and no broken symmetries; it will have a divergent susceptibility, a broken spin symmetry, or both.

In a random singlet phase with no spin-nonconserving interaction, the spin transport will be anomalous. Spins will tend to get "trapped" by the localized moments at low temperature so the spin diffusion coefficient $D_s(T) \to 0$ as $T \to 0$. If the spin is carried between the local moments by the same Fermi liquid quasiparticles which carry the charge then the Wiedmann-Franz law gives

$$D_s(T) = [\chi_0/\chi(T)]\sigma/K_0 , \quad (4)$$

where χ_0 and K_0 are the susceptibility and compressibility of a noninteracting Fermi gas and σ is the conductivity of the actual system [11,12]. In a metallic phase with a finite conductivity at $T=0$, this yields $D_s(T) \sim \chi^{-1}(T)$. Note that at any *fixed* temperature the susceptibility and other properties will be essentially homogeneous on sufficiently long length scales ($\gg [\rho_u(T)]^{-1/d}$), thus Eq. (4) should obtain. Further understanding of the transport processes is needed to validate or invalidate this result, in particular the effects of the RKKY coupling between the local moments.

We now consider the generality of our results. In one-dimensional systems, the results do not apply directly for a variety of reasons, including the absence of Fermi-liquid behavior even in a spinless system. Nevertheless, other arguments again suggest a divergent susceptibility [23]. In higher dimensions, however, our results should, at least in principle, be quite general. All that is needed is the possibility of forming an isolated spin in a region of radius r which is sufficiently isolated from the rest of the system that its exchange with the conduction electrons, $J \sim e^{-br}$. This may be on single sites as we have considered, or on clusters of sites. If this *can* occur, in a random alloy, then it will *always do so* with probability no smaller than e^{-Cr^d} since any possible event in a region of size r will occur with at least this probability. Thus we expect that in the presence of electron-electron interactions in *any random alloy* which has a metal-insulator transition to an insulating phase with local moments as a function of concentration, isolated moments, and the associated destruction of the dirty Fermi liquid will occur for *any concentration.* This should obtain provided the Fermi level in the alloy lies *below* the electron affinity level of the pure insulating component and between the odd (lower) and even (higher) electron states of *any* cluster embedded in the insulating matrix. Exceptions would be systems in which it is impossible to isolate an *odd* number of electrons, or those which are metallic at all compositions. The best systems for observing the effects discussed here are clearly those near to a metal-insulator transition; deep in the metallic phase they will occur only at inaccessibly low T.

So far, we have assumed only Heisenberg spin interactions. In the presence of spin-orbit interactions, crystal fields, etc., the spin diffusion would of course no longer exist even in a conventional dirty Fermi liquid. Nevertheless, the thermodynamic properties of the random singlet phase should still persist at least for weak symmetry-breaking perturbations. With predominantly antiferromagnetic RKKY exchange, clusters of even numbers of spins will typically have nondegenerate ground states with a gap. Odd-numbered clusters will, because of time-reversal invariance, have degenerate Kramers doublet ground states. This strongly suggests that the basic

mechanism which causes the random singlet phase (i.e., weakly coupled spins interacting directly and via excitation of intervening pairs with a gap) will persist. In the most general case the pairs will not have any good quantum number except time reversal but the weakly coupled pairs will nevertheless still give rise to divergent susceptibilities and specific-heat coefficients of the same form as the isotropic case studied here.

Our results cast doubts over perturbative field theoretic approaches to the MI transition from the metallic side [11,12] which leave out the effects of rare regions. Such difficulties may be related to the direct breakdown of the spin transport in perturbation theory investigated recently [13]. This field-theoretic treatment to two-loop order finds a transition as disorder is increased from a metallic phase with both charge and spin diffusion to a metallic phase with charge diffusion but no spin diffusion, *before* the onset of charge localization (MI transition) at still higher disorder. Our results suggest that quite generally the zero-temperature spin diffusion coefficient at long wavelengths is zero in the metallic phase—presumably their transition is destroyed by nonperturbative effects.

For a random site model with an energy scale ~ 1–10 eV and $n \sim 1.5 n_c$, we estimate that these effects should be observable at millikelvin temperatures. However, the estimates are exponentially sensitive to input parameters, and therefore difficult to make reliable. It would nevertheless be of interest to look at the magnetic and thermodynamic behavior of other atomically disordered metal-insulator composites (where the metal atom has unpaired spins and little tendency to form atomic pairs with zero spin) near the MI transition, to see how universal these features are. Other candidate systems include disordered heavy fermions (with low or zero superconducting temperatures) where the Fermi liquid in the crystalline material is already fragile, and low-dimensional systems (e.g., δ-doped semiconductors) where some of these ideas can be tested more quantitatively.

In conclusion, we have shown that for the positionally disordered Anderson-Hubbard model, the low-temperature thermodynamics (in the absence of instabilities like superconductivity) is *not* that of a Fermi liquid, even deep in the metallic phase, but rather a random singlet metal with a divergent susceptibility and specific-heat coefficient, and concomitant absence of spin diffusion in the long-wavelength limit. This behavior, due to rare fluctuations, should be ubiquitous and may be observable in other quenched disordered systems [24].

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