Non-Fermi Liquid Behavior and Griffiths Phase in f-Electron Compounds

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We study the interplay among disorder, RKKY, and Kondo interactions in f-electron alloys. We argue that the non-Fermi liquid behavior observed in these systems is due to the existence of a Griffiths phase close to a quantum critical point. The existence of this phase provides a unified picture of a large class of materials. We also propose new experiments that can test these ideas. [S0031-9007(98)07424-9]

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The observation of non-Fermi-liquid (NFL) exponents in the thermodynamic and transport properties of f-electron alloys has stimulated considerable interest in the study of these materials [1]. The alloys in which NFL behavior is observed fall into two categories: (1) Kondo hole systems, in which the f-electron atoms (R) are replaced by nonmagnetic metallic atoms (M) according to the formula $R_{1-x}M_x$, and (2) disordered ligand systems, in which the metallic atoms are substituted for a different metallic atom according to the formula $R(M1)_{1-y}(M2)_y$. Notice that due to alloying these compounds have a high probability of being disordered. That disorder is indeed a very important factor in bringing about the NFL behavior in these compounds has been shown in recent experiments [2]. This is in addition to the fact that most of these systems are close to a phase transition. Then, we claim that the NFL properties of these compounds are a consequence of the competition between the intrasite Kondo and the intersite Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions taking place in the midst of a disordered environment. If disorder were not present, there are two possibilities: the compound will have long-range magnetic order when the RKKY interaction is sufficiently large compared with the Kondo interaction or the compound will be paramagnetic due to the quenching of the magnetic moments of the rare earth atoms. However, the experimental observations show that the NFL behavior generally appears between these two phases [1]. Several proposals have been put forward as the possible explanations for the NFL behavior. A possible scenario is based on single impurity models with particular symmetries such as the multichannel Kondo effect of magnetic [3] and electric origin [4,5]. Another possible scenario attributes the NFL behavior to proximity to a quantum critical point [6,7]. Recently, a route to NFL behavior that emphasizes a disorder-driven mechanism, known as "Kondo disorder," has been suggested [8,9]. All of these proposals have had partial success in explaining some of the experiments [1]. In particular, conformal invariance scaling gives a good description of the dynamic susceptibility $\chi''(\omega, T)$ and the electrical resistivity in $UCu_{5-x}Pd_x$ [10], a Kondo disorder model can explain the temperature dependence of the electrical resistivity [9], and the phase diagram of spin glass models is qualitatively similar to the one observed in these alloys [7]. Nevertheless, there is no final word as to the origin of the NFL behavior in these alloys. Here, we propose a framework that incorporates what we believe are the essential aspects of the problem: disorder and the competition between RKKY and Kondo effect. In this framework the presence of disorder leads to the coexistence of a metallic (paramagnetic) phase with a *granular* magnetic phase. We show that this coexistence phase is equivalent to the *Griffiths phase* of dilute magnetic systems [11]. In our scenario, we have two electronic fluids: one of them is quenched by the Kondo interaction, behaving as a Fermi liquid, and the other is dominated by the RKKY interaction, leading to ordered regions of two-level systems. We have, therefore, an inhomogeneous environment which is brought about by disorder. This scenario is reminiscent of the one found in compensated doped semiconductors (Si:P, Ge:Sb) [12–14]. In these systems, disorder leads to local density fluctuations which result in the formation of magnetic moments. The Griffiths phase is characterized by the formation of rare strongly coupled magnetic clusters which have large susceptibilities. In this phase the thermodynamic functions show essential singularities with strong effects at low temperatures. At these low temperatures, clusters of interacting magnetic moments can be thought of as "giant" spins which can tunnel over classically forbidden regions. In the Griffiths phase magnetic clusters with N spins have a relaxation time which is given by [15] (we use units such that $\hbar = k_B = 1$)

$$\tau_R = \omega_0^{-1} e^{N\zeta}, \tag{1}$$

where ω_0 is an attempt frequency and ζ is a characteristic parameter that we discuss below. Because of cluster formation in the paramagnetic phase of these systems, we have the following predictions for the thermodynamic

functions:

$$\begin{aligned}\gamma \equiv C_V/T &\propto [\chi(T)]_{\rm av} \propto T^{-1+\lambda},\\ [\chi_{\rm nl}(T)]_{\rm av} &\propto T^{-3+\lambda},\\ [\chi_L(\omega)'']_{\rm av} &\propto \omega^{-1+\lambda}\tanh(\omega/T),\\ T_1^{-1}(\omega) &\propto \omega^{-2+\lambda}T\tanh(\omega/T),\\ \delta\chi(T)/\chi(T) &\propto T^{-\lambda/2},\end{aligned} \tag{2}$$

where $[\cdots]_{\rm av}$ means average over disorder. C_V is the specific heat, $\chi(T)$ is the static susceptibility, $\chi_{\rm nl}(T)$ is the nonlinear static susceptibility, $\chi_L(\omega)$ is the local frequency dependent susceptibility, $1/T_1$ is the NMR relaxation rate, and $\delta\chi(T)$ is the mean square deviation of the susceptibility due to the distribution of susceptibilities in the system [16]. Here $\lambda = -\zeta^{-1}\ln(c)$, where c denotes the density of the spins. The Griffiths phase is characterized by $\lambda < 1$ so that the susceptibilities diverge at zero temperature. We propose that Griffiths singularities dominate the physics of the system at low temperatures leading to NFL behavior. Let us note here that power-law behaviors for γ and χ have also been obtained by other researchers using different approaches [10,13,14]. Notice that the logarithmic behavior observed in some NFL compounds [1] can as well be fitted by small power laws ($\lambda \approx 1$). Also, it follows from (2) that NFL systems should have *positively* divergent nonlinear susceptibilities ($\lambda < 3$). Indeed, $U_{0.9}Th_{0.1}Be_{13}$ shows a tendency to a positively divergent susceptibility [17], in contrast to the usual negative divergence of the paramagnetic susceptibility of UBe_{13}. Systems like $UCu_{5-y}Pd_y$ show even stronger divergent behavior and can be considered "deep" inside of the Griffiths phase. Recent neutron scattering experiments show that the imaginary part of the susceptibility, the specific heat, and the static susceptibility can be exactly fitted by the result (2) with $\lambda = 2/3$ [18]. To compare this result with NMR and μSR, we calculated the variation of the Knight shift, $\delta K/K \propto \delta\chi/\chi$, for the same material [8]. Our result is shown in Fig. 1 for $\lambda = 2/3$.

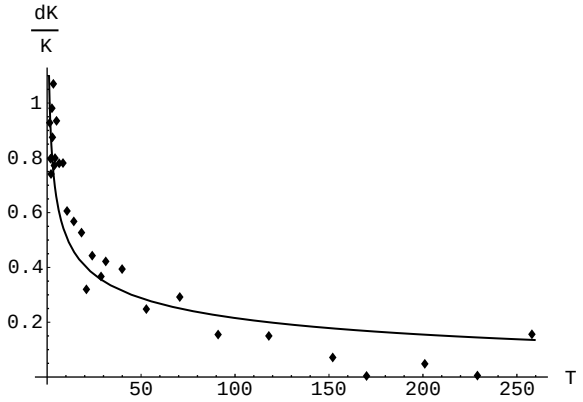

FIG. 1. Mean square deviation of the Knight shift as a function of temperature given in (2). Diamonds: Experiments for UCu_4Pd from Ref. [7].

The agreement between theory and experiment is very good. Our predictions are robust in the sense that *all* the results in (2) have to be self-consistent. Moreover, we have a definite prediction for the NMR relaxation rate $1/T_1(\omega)$, which should be largely frequency dependent. We also predict that under pressure the exponent λ should change inside of the Griffiths phase [19]. It would be interesting to plot the logarithmic derivative of the susceptibility (and/or specific heat) as a function of temperature and pressure, $\lambda(T,P) = 1 + \partial\ln\chi(T,P)/\partial\ln T$, for various NFL systems to verify our predictions. The main characteristics of f-electron systems studied here are interplay between RKKY and Kondo effects, magnetic anisotropy, and disorder due to alloying. It is well known that f-electron systems are characterized by their strong magnetic properties. This magnetism arises from the crystal field interactions and the strong spin-orbit coupling; in some of these materials, the large f-electronic clouds lead to an anisotropy comparable in magnitude to the exchange energy. In addition to this ion anisotropy, one expects a Dzyaloshinsky-Moriya (DM) exchange interaction [20] generated by the spin-orbit coupling. This type of interaction is forbidden only in highly symmetric situations which are not commonly realized in alloys with very complex unit cells [21]. That this is the case, it can be seen from the coexistence of weak parasitic ferromagnetism within the antiferromagnetic phase in some materials such as R-CrO_3 systems [22]. We believe that this DM interaction accounts for the recent neutron scattering data in the NFL compound $CeCu_{6-y}Au_y$ [23]. Consequently the RKKY and Kondo interactions will be strongly anisotropic in the f-electron systems. The simplest Hamiltonian that describes the situation above is the anisotropic Kondo model

$$H = H_e + \sum_n \left[J_n^z S_n^z s_n^z + \frac{J_n^\perp}{2}\left(S_n^+ s_n^- + S_n^- s_n^+\right)\right], \tag{3}$$

where the sum is over all the R atoms with spin $\mathbf{S}_n$. H_e is the conduction electron Hamiltonian that can be obtained from band structure calculations, and $s_n^a = \sum_{\sigma,\sigma'} c_{n,\sigma}^\dagger \tau_{\sigma,\sigma'}^a c_{n,\sigma'}$ with $a = x, y, z$ is the conduction electron spin (τ^a are Pauli matrices). The main difference between Kondo hole and disordered ligand systems is the alloying process. In the Kondo hole the magnetic atom is replaced which leads to a reduction of the number of magnetic moments. Moreover, the substitute atom has a different size from the original R atom which will lead to local changes in the lattice structure. In disordered ligand systems the lattice spacing is also affected by the substitution of the ligand atoms. As is well known, hybridization matrix elements between the conduction band and the f-electron system are exponentially sensitive to changes such as the type of lattice and characteristics of the substitute atom, and this will in turn affect the local values of the exchange constants in (3). Therefore, alloying leads to a situation where local variations result in parts of the sample having larger exchange interactions than others. The magnetism in these systems

can be understood by looking at the effective interaction between the R atoms due to the conduction electrons. The magnetic Hamiltonian in second order perturbation theory relative to the free electron problem is

$$H_M \approx \sum_{n,m,a,b} S_n^a \, \Gamma_{n,m}^{a,b} \, S_m^b \,, \tag{4}$$

where $\Gamma_{n,m}^{a,b}$ is the RKKY interaction mediated by the conduction electrons. In the limit of large anisotropy, $J^z \gg J^\perp$, we have $\Gamma^{z,z} \sim \mathcal{O}[(J^z)^2/E_F]$ where E_F is the Fermi energy. In a clean d-dimensional system this interaction decays like $1/r^d$. In the presence of disorder, however, it decays like $e^{-r/\ell}$ where ℓ is the spin-orbit diffusion length [24]. Since we are dealing with disordered systems the effective interaction $\Gamma_{n,m}^{a,b}$ is actually short ranged. The important point here is that the ordering temperature of the magnetic system, T_c, is of order of the exchange interaction, that is, $T_c(J^z) \propto \Gamma \propto (J^z)^2/E_F$. The critical ordering temperature is not the only energy scale in the problem since the Kondo effect can take place and quench the magnetic moment.

Consider, for instance, a particular position in the system, say, $n = M$, where, due to the alloying, J_M^z is much larger than average. For simplicity we disregard all the other sites and look at the physics at this particular site. The Hamiltonian of interest can be written as $H_M = H_0 + H_I$, where

$$\begin{aligned} H_0 &= H_e + J_M^z S_n^z (n_{M,\uparrow} - n_{M,\downarrow}) \,, \\ H_I &= \frac{J_M^\perp}{2} (S_M^+ s_M^- + S_M^- s_M^+) \,, \end{aligned} \tag{5}$$

and in the limit of large anisotropy we treat H_I as a perturbation of H_0. Observe that H_0 can be easily diagonalized in the S^z basis. If the R atom is in a state such that S^z is $\Uparrow$ ($\Downarrow$) the energy of the system is minimized by making a bound state with an electron with spin $\downarrow$ ($\uparrow$), respectively. The bound state energy is just the Kondo temperature

$$T_K(M) = W e^{-1/N(0)J_M^z}, \tag{6}$$

where $N(0)$ is the renormalized density of states at the Fermi energy, and W is the conduction electron bandwidth. This situation is similar to the approach to NFL behavior which takes into account a disordered distribution of Kondo temperatures [8,9]. The ground state of H_0 is doubly degenerate corresponding to the spin configurations $|\Uparrow,\downarrow\rangle$ and $|\Downarrow,\uparrow\rangle$. Application of first order perturbation theory in H_I shows that the singlet state $|\Uparrow,\downarrow\rangle - |\Downarrow,\uparrow\rangle$ is lower in energy than the triplet state $|\Uparrow,\downarrow\rangle + |\Downarrow,\uparrow\rangle$ by an amount $J_M^\perp$. In this case H_I acts as a *transverse field* and lifts the degeneracy of the bound state. In fact, using the bosonized version [25] of Hamiltonian (5) and mapping the problem into the dissipative two-level system [26], we can show that tracing out the electrons of the problem results in a S_M^x operator for the R atoms spin degrees of freedom [27]. The competition between the RKKY interaction and the Kondo effect can be understood in terms of the two relevant energy scales, that is, $T_K(M, J^z)$ and $T_c(J_z)$: (i) If $T_K(M) \gg T_c$ as we lower the temperature of the system below $T_K(M)$ the local magnetic moment is quenched and order is inhibited; and (ii) if $T_K(M) \ll T_c$ and the temperature is lowered below T_c there is local magnetic order and the Kondo effect is suppressed [28]. If we now take into account the magnetic moments that are left to interact via RKKY, as in (4), together with the sites which are quenched by the Kondo effect, as in (5), we see that the magnetism of the original problem in the limit where $J_n^z \gg J_n^\perp$ is described by

$$H_{\text{eff}} \approx \sum_{\langle i,j \rangle} \Gamma_{i,j} S_i^z S_j^z + \sum_i t_i S_i^x \,, \tag{7}$$

where the brackets $\langle i, j \rangle$ imply nearest neighbor coupling. Notice that $\Gamma_{i,j} \sim \mathcal{O}((J^z)^2/E_F)$ and $t_i \sim \mathcal{O}(J^\perp)$ are random (but intercorrelated) variables dependent on the alloying and lattice structure. Thus we have mapped our problem into the random Ising model in a random transverse magnetic field [15]. The phase diagram of this model follows: at small doping the RKKY interaction dominates and the system can order magnetically (the ordered phase can be antiferromagnetic, ferromagnetic, or spin glass [7] depending if the mean value of $\Gamma_{i,j}$ in (7) is positive, negative, or null, respectively). With increasing doping the quantum fluctuations grow due to the Kondo effect and the bulk critical temperature decreases until it vanishes for some critical value of doping. At this quantum critical point the system percolates. For large values of doping, inside of the paramagnetic phase, only finite clusters of magnetic atoms can be found. Among these clusters there are some rare ones which are large and strongly coupled. Within these clusters the spins behave coherently as a giant spin or a magnetic grain. We can describe the cluster in terms of a "classical" degree of freedom which can be parametrized by Euler angles (θ, ϕ). The classical energy, $E(\theta, \phi)$, will have at least two minima due to the original degeneracy of the magnetic ground state. In the simplest of cases, $E(\theta, \phi)$ can be written in terms of a classical spin with X-easy axis and XY-easy plane,

$$E(\theta, \phi) = N(-\epsilon_\perp + \epsilon_{||} \sin^2\phi) \sin^2\theta \,, \tag{8}$$

where $\epsilon_\perp > \epsilon_{||} > 0$ are the anisotropy energies perpendicular and parallel to the easy axis, respectively. These energies depend on the microscopic coupling constants in (7). Observe that the energy has two minima at $(\pi/2, 0)$ and $(\pi/2, \pi)$ with an energy barrier between them. When the temperature is higher than the barrier height the cluster is thermally activated and behaves classically. At lower temperatures the cluster can undergo quantum tunneling between the two minima. Using standard instantons methods [29], we can calculate the parameters that appear in (1):

$$\begin{aligned} \omega_0 &= 2\sqrt{\epsilon_{||}\epsilon_\perp} \,, \\ \zeta &\approx \ln\left(\frac{1 + \sqrt{\epsilon_{||}/\epsilon_\perp}}{1 - \sqrt{\epsilon_{||}/\epsilon_\perp}} \right) . \end{aligned} \tag{9}$$

Notice that in the absence of anisotropy ($\epsilon_{\|} = \epsilon_{\perp}$) tunneling cannot occur ($\zeta \to \infty$ and $\tau_R \to \infty$), as expected. Then, the low energy physics for a cluster Ω with N spins reduces to the Hamiltonian

$$H_{\Omega(N)} = \Delta_{\Omega(N)} \tau^x, \tag{10}$$

where $\Delta_{\Omega(N)} = 1/\tau_R$ is the tunneling energy and is given in (1), and it can be related to the anisotropy energies by (9). Using (10) and averaging over cluster with different sizes, we arrive at the predictions given in (2) [16]. In conclusion, we propose that the NFL behavior observed in f-electron systems can be attributed to the existence of Griffiths singularities close to the quantum critical point. These singularities have their origin on the interplay between the RKKY and Kondo interactions in the presence of magnetic anisotropy and disorder. These conclusions have similarities with those discussed recently by Sachdev [30]. We were also able to map the disordered Kondo lattice problem into the random Ising model in a random transverse magnetic field where disorder is correlated. In the paramagnetic phase of this Ising model the physics of clusters can be understood in terms of the quantum tunneling of intrinsic magnetic grains which are described by a classical spin model. At low temperatures the spin degrees of freedom of a magnetic grain can tunnel over classically forbidden regions and at finite temperatures they can be thermally activated. At very low temperatures the problem reduces to a two-level system problem which, when appropriately averaged over disorder, leads to the Griffiths singularities and to the predictions in (2). This Griffiths phase will depend strongly on the type of lattice structure and value of the local microscopic exchange constants. This would explain why systems like $CeCu_{6-y}Au_y$ have to be fine-tuned for NFL behavior to be observed, while others, like $Th_{1-x}U_xPd_3$, have large regions of NFL behavior. It is indeed possible, as in 1D systems [15], that Griffiths behavior can extend over large regions of doping [31].

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Note added.—After this paper was completed, experimental indications of possible Griffiths phase behavior have been reported [32].

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