Possible origin of the resistivity maximum in heavy-fermion systems

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The temperature T_M of the resistivity maximum in $CeCu_2Si_2$ and other heavy-fermion compounds has been assumed by some authors to be directly related to the value of the Kondo temperature T_K in the same compound. In the present paper, I present evidence that, as is well known for spin-glasses, the Ruderman-Kittel-Kasuya-Yosida interaction between magnetic ions in heavy-fermion systems plays an important or even dominant role in determining the value of T_M. It is argued that across the quasiternary series $La_{1-x}Ce_xCu_2Si_2$ the increase of T_K with x is small, only doubling in value across the series.

The temperature dependence of the electrical resistivity of numerous heavy-fermion systems—including $CeCu_2Si_2$, $CeCu_6$, UBe_{13}, U_2Zn_{17}, and $NpBe_{13}$—displays a maximum at a temperature T_M which generally lies below 40 K.[1] On a linear plot of resistivity versus temperature, the drop in the resistivity for $T < T_M$ appears quite precipitous. This has prompted some experimentalists to identify the resistivity maximum as a crossover point between incoherent single-ion Kondo scattering at high temperatures $T > T_M$ and progressively coherent Kondo-lattice behavior at lower temperatures $T < T_M$; T_M has been taken as proportional to (or at least a monotonic function of) the Kondo temperature T_K when analyzing shifts of T_M with compound composition[2-4] or under high pressure.[3,5,6] This interpretation of T_M was given theoretical support by Lavagna *et al.*,[7] who derive $T_M \propto T_K$ for constant conduction-electron number; this model does consider coherence effects in a periodic Anderson lattice, but neglects the influence of the normal Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction between magnetic ions which contains interaction components not involved in the many-body resonance.[8] In this paper, I will discuss evidence that at the present state of knowledge the value of T_M in heavy-fermion compounds should be considered to be a function of both T_K *and* the mean RKKY-interaction strength Δ_c, in which case T_K and T_M would not be proportional.

In heavy-fermion systems the magnetic ions are present in high concentrations and possess sizeable local magnetic moments over an appreciable temperature range.[1] In $CeCu_2Si_2$ the magnetic susceptibility approximately follows a Curie-Weiss law above 30 K with an effective moment per Ce ion of $2.5\mu_B$, the full value expected for trivalent Ce local moments. One way to arrive at a better understanding of such a complex concentrated magnetic system is to follow the evolution of the magnetic behavior from the dilute to the concentrated limit. Steglich *et al.*[4] have pointed out that possession of giant specific-heat γ values is not limited to concentrated heavy-fermion compounds, but is also shared by very dilute Kondo alloys. Cu—81 ppm Fe ($T_K \simeq 28$ K) and Cu—51 ppm Cr ($T_K \simeq 2.1$ K) have values of γ per mole of impurity of ~ 1 J/K^2 mole Fe and 16 J/K^2 mole Cr, respectively.[9] These values thus roughly follow the expected relation $\gamma \propto T_K^{-1}$. To achieve even larger values of γ we need only to choose systems with much lower values of the Kondo temperature. An excellent candidate would be *Ag*Mn with $T_K \simeq 10^{-16}$ K,[10] which would imply $\gamma \simeq 10^{17}$ J/K^{2}mole Mn, a truly monumental γ value! However, to confirm this value experimentally one would have to not only cool the sample down to temperatures well below $T_K \simeq 10^{-16}$ K, *but also* reduce the impurity concentration to a level $< 10^{-14}$ ppm Mn in Ag so that the RKKY interactions between the impurities could be neglected, i.e., $\Delta_c \ll T_K$. Should the RKKY interactions become so high that $\Delta_c \gg T_K$, the Kondo resonance would be quenched and γ would revert to normal values. Because for a system with a given value of T_K, the RKKY interaction is much (100 times) larger for $3d$ compared with $4f$ impurities, $4f$-impurity systems will be more likely to show heavy-fermion behavior. For example, dilute *Au*Fe and *La*Ce both have $T_K \simeq 0.2$ K,[8] but Au-1 at.% Fe experiences spin-glass freezing at 10 K, whereas 100% Ce (β phase) orders antiferromagnetically only at 13 K.

From the above it is clear that (i) dilute local-moment Kondo systems can also show giant values of γ, and (ii) rare-earth ions can more easily maintain their γ values in magnetically concentrated compounds like the heavy fermions because of weaker RKKY-interaction effects. I will now address the question of the change in the values of Δ_c and T_K when the concentration of the Kondo ion in a system, for example, $La_{1-x}Ce_xCu_2Si_2$, is increased from the dilute to the concentrated limit.

Electrical resistivity is an especially sensitive tool for studying not only the gradual buildup with temperature of the single-impurity Kondo resonance but also even subtle perturbations of this resonance due to the effects of impurity-impurity interactions, applied magnetic field, or nonmagnetic impurities. The evolution of the Kondo resistivity anomaly as the magnetic impurity concentration and thus the RKKY-interaction strength Δ_c increases is clearly shown by the calculated resistivity curves of Larsen[8,11] in Fig. 1. As the value of the ratio Δ_c / T_K increases from zero, for example, by increasing the magnetic impurity concentration c ($\Delta_c \propto c$) with T_K remaining essentially constant, the low-temperature plateau of the

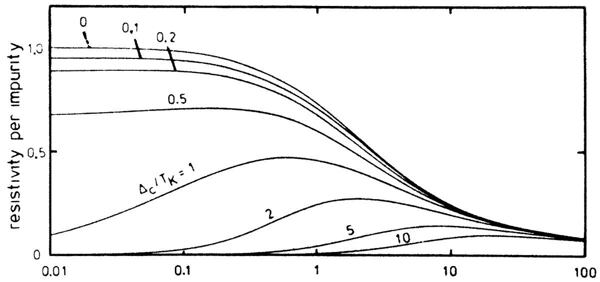

FIG. 1. Functional dependence of the electrical resistivity per impurity (linear scale) on the relative temperature T/T_K (logarithmic scale) and on the relative interaction strength Δ_c/T_K according to Larsen (Ref. 11). The uppermost curve is for $\Delta_c=0$, the single-impurity limit.

resistivity anomaly *per impurity* is seen to be depressed. A well-defined resistivity maximum at a temperature T_M develops for $\Delta_c/T_K \geq 0.5$, which moves to higher temperatures as Δ_c/T_K increases further (note that T_M can be both smaller or greater than T_K, depending on the value of Δ_c). If the RKKY interactions are sufficiently strong, magnetic ordering or spin-glass freezing may occur at a temperature T_0 which is generally less than T_M, as indicated in Fig. 2. These basic trends were given ample experimental verification in the 1970s by careful studies on numerous Kondo and spin-glass systems with both transition-metal and rare-earth impurities.[12] The relative value of Δ_c/T_K determines whether magnetic ordering

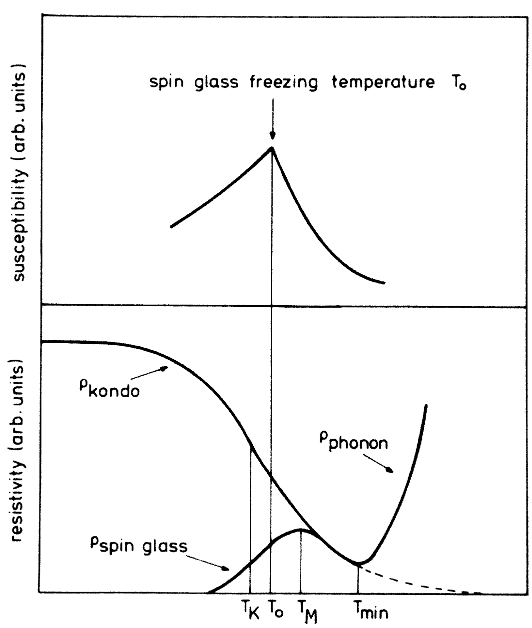

FIG. 2. Schematic diagram of the resistivity of isolated impurities ρ_{Kondo} and interacting impurities $\rho_{\text{spin-glass}}$ as a function of $\log T$. Interactions between impurities give rise to a resistivity maximum at T_M and a susceptibility peak at a lower temperature T_0.

quenches the Kondo effect, or vice versa. It is important to realize that the presence of a resistivity maximum does *not* imply that magnetic ordering or freezing phenomena actually occur, but rather marks the approximate temperature below which RKKY interactions between magnetic ions begin to convert single-ion Kondo behavior into correlated-ion behavior. From the above discussion it is clear that T_M is neither a direct measure of Δ_c, as was often assumed in the 1960s and early 1970s, nor of T_K, but is, unfortunately, a complicated function of both [for $\Delta_c \gg T_K$ Larsen derives the relation $T_M \simeq \frac{1}{2}\Delta_c \ln(\Delta_c/T_K)$]. This fact is perhaps most clearly brought out by high-pressure experiments on the system *Au*Fe where T_M decreases under pressure[8] in spite of the fact that both the spin-glass freezing temperature T_0,[13,14] where $T_0 \propto \Delta_c$, and the Kondo temperature T_K increase. Further high-pressure work[8,13,14] has confirmed the basic correctness of the derived functional dependence of T_M on Δ_c and T_K.

Perhaps the most representative measurements of all for the above phenomena were carried out by Winzer[15] on the $La_{1-x}Ce_xB_6$ series, where the entire evolution of the magnetic properties from the single-impurity Kondo anomaly in dilute $La_{1-x}Ce_xB_6$ to spin-glass freezing for intermediate x to long-range magnetic ordering for CeB_6 is displayed. In Fig. 3 the magnetic contribution to the resistivity of this series is plotted versus $\ln T$ for values of x from 0.02 to 0.7. The gradual monotonic increase of T_M with Ce concentration seen in the inset of Fig. 3 has been observed on numerous other systems[12,16] and arises from the increase in the strength of the RKKY interactions. Whereas T_M increases by more than a factor of 6 across this series, the Kondo temperature T_K is only expected to change by a factor of 2, as will be discussed below.

In Fig. 4 are shown the results of Aliev *et al.*[2] on the quasiternary compound series $La_{1-x}Ce_xCu_2Si_2$. The development of the resistivity maximum at lower Ce concentrations and its almost linear increase with x all the

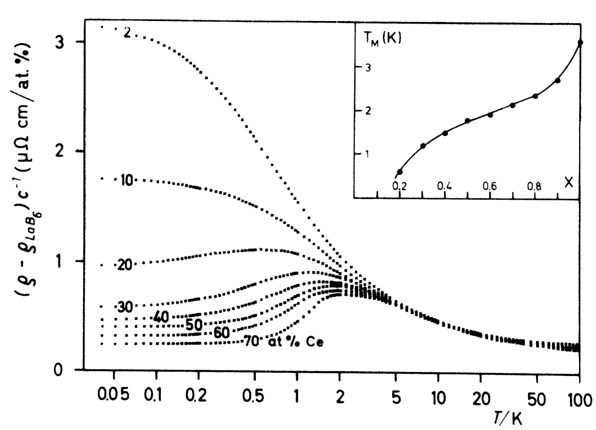

FIG. 3. Logarithmic temperature dependence of magnetic contribution to resistivity (measured resistivity minus phonon resistivity of LaB_6) per atomic percent of Ce across the quasiternary series $La_{1-x}Ce_xB_6$ from Winzer (Ref. 15). Inset shows dependence of temperature of resistivity maximum T_M on Ce concentration x.

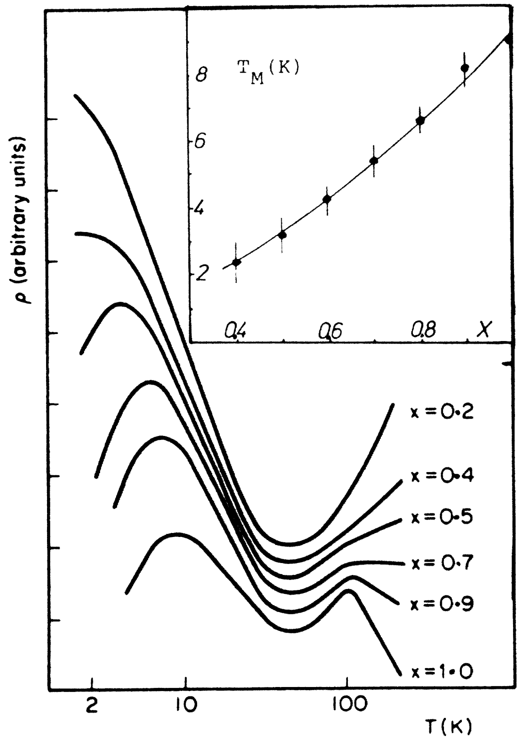

FIG. 4. Resistance in arbitrary units (linear scale) versus T (logarithmic scale) across quasiternary series $La_{1-x}Ce_xCu_2Si_2$ from Aliev *et al.* (Ref. 2). Inset shows dependence of temperature of resistivity maximum T_M on Ce concentration x.

way across the series to $x=1$ ($CeCu_2Si_2$) closely parallels the behavior of the $3d$- and $4f$-impurity systems discussed above. This is strong evidence that RKKY interactions play an important role in the origin of the resistivity maximum in stoichiometric $CeCu_2Si_2$, the first heavy-fermion superconductor.[17] A similar discussion applies to the $La_{1-x}Ce_xAl_3$ series where the resistivity maximum, which is clearly visible for $x \geq 0.7$, moves into a crystal-field resistivity peak for $x \to 1$.[18,19] Aliev *et al.*[2] and Brandt and Moshchalkov[3] attempt to account for the resistivity data in Fig. 4 by a rapid increase in T_K with x. Substituting Ce for La in $La_{1-x}Ce_xCu_2Si_2$ does decrease the volume of the unit cell V_{uc} which would be expected to lead to an increase in T_K.[8] However, the measured decrease in V_{uc} ($\sim 2\%$) is far too small to account for the large T_K increase (100 times) proposed by the above authors. Direct high-pressure studies on dilute *La*Ce ($T_K \simeq 0.2$ K) and *Y*Ce ($T_K \simeq 40$ K) give the volume dependences $\gamma_m \equiv -\partial \ln T_K / \partial \ln V \simeq +50$ and $+30$, respectively. Using the value $\gamma_m = +40$ for $CeCu_2Si_2$ ($T_K \simeq 10$ K) would imply that the small 2% decrease in V_{uc} from very dilute $La_{1-x}Ce_xCu_2Si_2$ to $CeCu_2Si_2$ should result in only a doubling of the value of T_K [$\delta T_K / T_K \simeq (\delta V/V)\gamma_m \simeq 2\%(40) = 80\%$]. This twofold T_K increase is far less than the hundredfold increase suggested by Aliev *et al.*[2] and Brandt and Moshchalkov[3] who attempted to use universal resistivity curves or paramagnetic Curie temperatures to estimate $T_K(x)$. Unfortunately, the sizeable RKKY interactions in $La_{1-x}Ce_xCu_2Si_2$ (and $La_{1-x}Ce_xAl_3$) make such a simple analysis unreliable.

One significant difference between $CeCu_2Si_2$ and $CeAl_3$ and the other concentrated systems such as CeB_6 discussed above is the lack of magnetic ordering. It is, however, important to note that for $x \geq 0.7$ the quasiternary series $La_{1-x}Ce_xCu_2Si_2$ displays low-field susceptibility peaks suggesting spin-glass freezing.[2,3] One possible reason for the disappearance of magnetic ordering or freezing phenomena as $x \to 1$ might be related to the increasing coherence of the Kondo-lattice behavior as $x \to 1$. Acting together in a coherent fashion the Kondo ions may be much more effective in quenching the magnetic order. Substituting La for Ce breaks up this coherence and allows magnetic ordering or freezing. It is interesting to note that the Kondo temperatures of $CeCu_2Si_2$ ($T_K \simeq 10$ K) and $CeAl_3$ ($T_K \simeq 5$ K) (Ref. 4) lie above that of CeB_6 ($T_K \simeq 1$ K) (Ref. 15); it would thus be expected that CeB_6 would have a greater tendency to order magnetically.

In the present discussion I have tried to emphasize the care which must be taken in separating out those effects which might be characteristic for heavy-fermion compounds from those which are well known from previous investigations on Kondo and spin-glass systems. Heavy-fermion systems contain a high concentration of magnetic ions and the effects of the RKKY interactions between these ions must be carefully considered. It seems very likely that these interactions play an important role in the formation of the low-temperature resistivity maximum in $CeCu_2Si_2$, $CeAl_3$, and Ce-based heavy fermions as well as in the uranium systems. It is conceivable that UPt_3 also has a resistivity maximum near 40 K, which, however, is buried under the phonon scattering. It would be of considerable benefit to a detailed analysis if experimental results were plotted versus $\ln T$ and the phonon resistivity were at least approximately subtracted.

It would seem reasonable to divide up the magnetic contributions to the resistivity of heavy-fermion systems into three regions: (1) a high-temperature region where single-impurity Kondo scattering dominates, (2) an intermediate region where RKKY interactions lead to correlations between impurities, thus reducing the very large Kondo scattering and leading to a resistivity maximum, and (3) a low-temperature region where long-range coherence effects couple the Kondo scattering of one magnetic ion with many others, leading to a nonmagnetic Fermi-liquid ground state. Coherence effects have recently been studied by Lavagna[20] in the two-impurity Kondo problem. Applying high pressures to a heavy-fermion system will shift T_K to higher temperatures and extend the temperature range where strong coherency effects are important, at the cost of region (1) and, particularly, region (2). Of course, since $\gamma \propto T_K^{-1}$, a heavy-fermion system will rapidly "lose weight" under pressure. Concerning the resistivity of $CeCu_2Si_2$, it cannot be excluded that coherency effects play some role in determining the value of T_M. The magnetic state of this system in the temperature range near 10 K is obviously in a very delicate state of balance. Two or more characteristic energies are certainly important, as evidenced by high-pressure studies[6,21,22] which show that the resistivity cannot be scaled by a single characteristic energy, as is possible for single-impurity Kondo systems.[8,23]

To put the main message of this paper conservatively: at the present state of understanding it is certainly unwarranted to attempt to quantitatively analyze the resistivity data on heavy-fermion systems under the assumption that the temperature of the resistivity maximum T_M is a function of T_K alone. Resistivity data at very low temperatures, on the other hand, should allow a more straightforward analysis.

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