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Resistivity and magnetoresistance of $(U_{1-x}M_x)Be_{13}$ (M = La,Th,Y,Sc,Hf)

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Electrical resistivity and longitudinal magnetoresistance of $(U_{1-x}M_x)Be_{13}$, with M = La, Th, Y, Sc, and Hf, were investigated in the temperature range 0.4–300 K and fields to 14 T. Alloying-caused changes of the resistivity can be correlated with variations of U-Be separation, or localization or delocalization of U $5f$ states. The large, negative magnetoresistance of UBe_{13} is severely depressed when only a minor, ~5%, amount of impurities are introduced for U. The changes of magnetoresistance, for $x \leq 0.05$, cannot be related to either lattice-pressure effects or the electronic configuration of M. It is suggested that the large, negative magnetoresistance, similarly to the enhanced specific heat, results from intersite $5f$ correlations existing in UBe_{13} at low temperatures.

I. INTRODUCTION

U- and Ce-based compounds and alloys are under close theoretical and experimental scrutiny for their unusual physical properties with heavy-fermion behavior as the most notable. Although most of the physical properties of Ce-based heavy fermions can be qualitatively understood in the framework of a single ion Kondo model,[1,2] the origin of the heavy-electron mass in U-containing-systems is far less-well understood and much more controversial.[3] An essential ingredient of microscopic $4f$ system models is that $4f$ electrons are well localized and only weakly hybridized with ligand atom orbitals. This, in turn, leads to pronounced crystal field effects at low temperatures which are inseparable from and intertwined with a formation of the heavy-fermion state.[4] Larger spatial extend of $5f$ orbitals in U systems, on the other hand, results in much larger hybridization and, in general, much larger crystal-field-level separation, such that the crystal-field effects are not as important at low temperatures. Consequently, temperature and magnetic-field dependencies of various thermodynamic properties of both classes of systems can be qualitatively different. UBe_{13} is one of a few exceptions. A remarkable similarity has been pointed out, by several authors, between the temperature dependence of zero-field properties, such as specific heat and electrical resistivity, of UBe_{13} (Ref. 5) and of a Ce compound, $CeCu_2Si_2$, considered as a prime example of a Kondo lattice. Moreover, it has been shown that UBe_{13} can be easily alloyed with a large number of other MBe_{13} compounds, where M is a trivalent or tetravalent metal, and both the crystal structure and the nonmagnetic ground state are preserved. The degree of hybridization of $5f$ and conduction electrons of the $U_{1-x}M_xBe_{13}$ alloy can be controlled by a proper selection of MBe_{13}. In this respect, UBe_{13} could stand as an important system bridging the $4f$ and $5f$ heavy-fermion systems. A systematic study of hybridization effects on electrical resistivity for several pseudobinary $U_{1-x}M_xBe_{13}$ alloy systems is presented in this paper.

Despite the above-mentioned zero-field similarities of UBe_{13} and $CeCu_2Si_2$, their magnetic field responses are markedly different with the most pronounced differences observed in the magnetoresistivity. The field-induced change of the electrical resistivity of $CeCu_2Si_2$ at sufficiently low temperatures is positive[6] and is interpreted as due to coherence effects.[7] At higher temperatures, $T > 4$ K, the magnetoresistance becomes negative as expected from the single ion Kondo model.[1] Contrary to that behavior, the resistivity of UBe_{13} decreases as a function of magnetic field, down to the lowest attainable temperatures.[8,9,10] Moreover, in order to describe the magnetoresistivity of UBe_{13} at temperatures lower than 20 K by a single-ion Kondo model, the Kondo temperature (T_K) has to be considered a strongly varying function of temperature, an obvious deficiency of the theory. Our recent specific heat study of a number of pseudobinary systems $(U_{1-x}M_x)Be_{13}$ (Ref. 11) revealed drastic changes of the Sommerfeld coefficient, γ, with x, for $x < 0.2$ and nearly perfect scaling with U concentration for $x \geq 0.2$ and M = Y or Sc. The strong dependence of γ on x for $x < 0.2$ has been explained by U-U correlation effects existing in unalloyed UBe_{13}. Whether these correlation effects are responsible for the large, negative mangetoresistance of UBe_{13} is a subject of our investigation.

Finally, a more general issue of classifying which properties of heavy-fermion systems are sensitive to correlation effects and which may be explained by a single ion picture is considered.

EXPERIMENTAL

The material preparation has been discussed in detail in Ref. 11. The electrical resistivity samples have been cut from large, about 1 g, buttons. Because of the affinity of UBe_{13} to develop internal cracks upon cooling, most of the samples used in the measurements were cut in the form of short 3–4 mm bars to avoid the cracks, with a cross sectional area of about 1/4 mm^2. Owing to such a geometry our systematic errors were ±10%. Although all samples were carefully examined under a microscope we cannot completely exclude the possibility that some of them possessed internal cracks. Thus, consistency in resistivity values amongst a series of related (e.g., by composition) samples was used as a further check for lack of internal cracks. The electrical resistivity measurements were performed with a standard, dc four-probe method. Magnetic fields were always applied in the sample current direction. The temperature in magnetic field was measured with either a 220 Ω, 1/4 W Speer carbon resistor (below 1 K) or a Lake Shore capacitance sensor (above 1 K). Small magnetoeffects of the Speer resistor have been taken into account following a prescription given in Ref. 12.

ZERO-FIELD RESISTIVITY

The electrical resistivities of $(U_{1-x}Y_x)Be_{13}$ for $x=0, 0.03, 0.2, 0.8$, and 0.9 are presented in Fig. 1. The resistivity of UBe_{13} has several interesting features. First, it exhibits a Kondo-like temperature dependence, i.e., increases with a decrease of temperature, in the 300 K to 2.2 K temperature range. At 2.2 K it displays a maximum which is suppressed with only a minor amount of impurities, for example, the $x=0.03$ sample does not show the maximum down to 0.4 K. A similar behavior was observed, when other elements were partially substituted for U (Ref. 13), and therefore this has led to the widely accepted speculation that a change from noncoherent to coherent electron scattering on U ions takes place at about 2.2 K. Another not fully understood aspect of the UBe_{13} resistivity is a broad shoulder at about 25 K. This shoulder is also extremely sensitive to Y alloying, see Fig. 1. Neither the $x=0.03$ nor higher Y concentration samples exhibit this anomaly. Thorium alloying, on the other hand, transforms (see Fig. 2) the 25 K shoulder into a rather pronounced maximum and shifts it to higher temperatures. The resistivities of several $U_{0.1}M_{0.9}Be_{13}$ materials are displayed in Fig. 2. The samples with $M=$Y, Sc, and Hf, thus with elements leading to the contraction of the UBe_{13} lattice, have very similar resistivity vs temperature curves with no sign of the 25 K anomaly. The Th and La samples, on the other hand, have very noticeable shoulders (maxima after subtraction of the corresponding $ThBe_{13}$ or $LaBe_{13}$ resistivities) at temperatures >50 K. The most widely accepted interpretation of the 25 K structure in the resistivity of pure UBe_{13} is that it arises from a combination of a crystal-field splitting and a Kondo effect.[14]

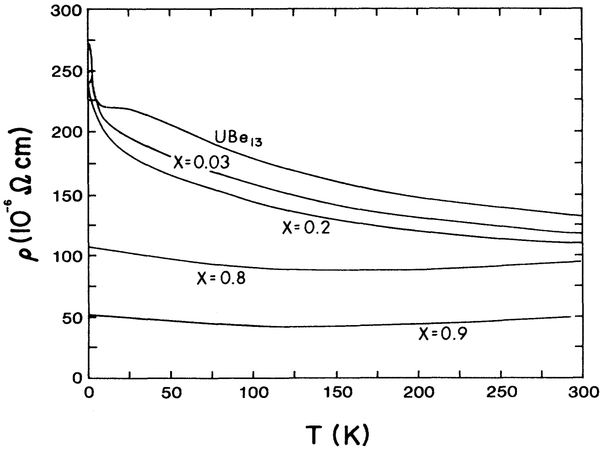

FIG. 1. Zero-field resistivity of $U_{1-x}Y_xBe_{13}$; $x=0$, 0.03, 0.2, 0.8, and 0.9. Note that only $x=0$ sample shows a low-temperature peak in the resistivity.

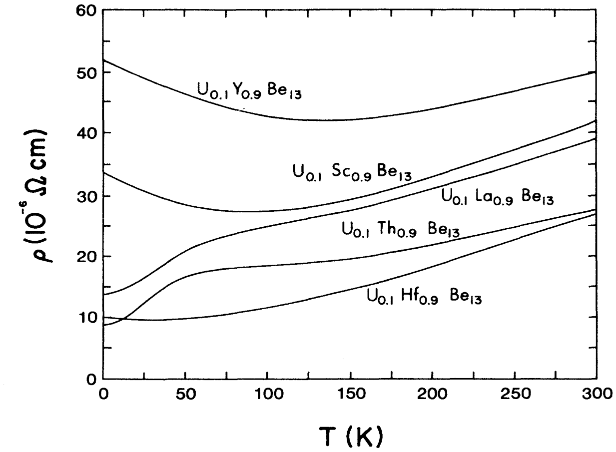

FIG. 2. Zero-field resistivity of $U_{0.1}M_{0.9}Be_{13}$; $M=$Y, Sc, Hf, La, and Th.

Our results are consistent with this interpretation. Note that the position of the high-temperature anomaly (or lack of the anomaly) in $U_{1-x}M_xBe_{13}$ alloys can be related to U-Be separation that is directly linked to a localization (Th,La) or delocalization (Y,Hf,Sc) process of U-5*f* states through the change of hybridization with Be *sp* states. This interpretation is further supported by studies of UBe_{13} at nonzero applied hydrostatic pressure.[15] Application of an external pressure is detrimental to this high-temperature anomaly in the UBe_{13} resistivity. There is, however, a quantitative difference between the chemical pressure (alloying) and the external pressure effects. The shoulder disappears for a pressure somewhere between 4.4 and 9.9 kbar, that corresponds to at least 1.4×10^{-3} relative decrease of the lattice constant.[16] The positive chemical pressure (alloying with Y) is much more effective in removing the anomaly, since replacement of 3% of U by Y results in only 5×10^{-5} relative reduction of the lattice constant. It is also interesting to note, that when the resistivity of YBe_{13} is subtracted from that of $U_{0.1}Y_{0.9}Be_{13}$, the resultant curve has a shape characteristic of 3*d* Kondo impurities such as *Cu*Fe, *Au*Fe (Ref. 17) and can be well fit to the Hamann[18] expression derived for transition-metal Kondo systems. On the other hand, the analogous procedure in the case of $U_{0.1}La_{0.9}Be_{13}$ and $U_{0.1}Th_{0.9}Be_{13}$ leads to curves very similar to those of some Ce compounds, e.g., $CeAl_2$ (Ref. 19), with a ground-state doublet and the crystal-field splitting of the order of 100 K. This again points up the contrast between delocalization (Y) and localization (Th,La) effects. Finally, the lowest temperature resistivities and

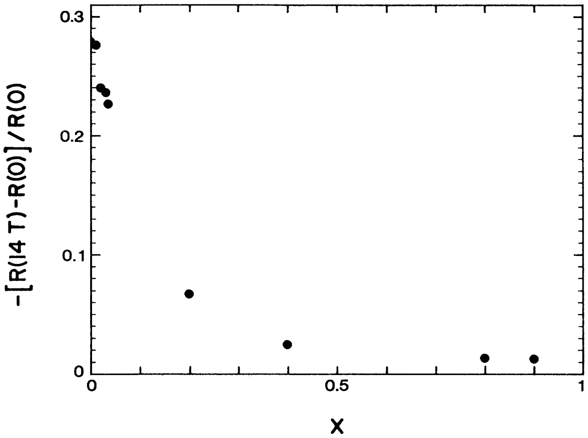

FIG. 3. Relative depression of resistivity of $U_{1-x}Y_xBe_{13}$ by 14 T field at $T=4.2$ K.

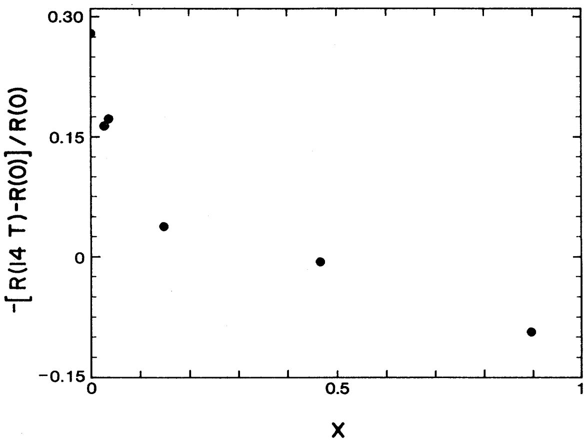

FIG. 4. Relative depression of resistivity of $U_{1-x}Th_xBe_{13}$ by 14 T field at 4.2 K.

temperature position of the resistivity minima (T_{min}) of $U_{0.1}M_{0.9}Be_{13}$, where M is an element smaller (e.g., Y, Sc, Hf) than U, also correlate with U-Be distance (see Fig. 2). Increase of the hybridization (smaller U-Be distance) leads to a quick suppression of the residual resistivities and a decrease of T_{min}. (Due to the presence of important crystal-field effects for the case of more localization (e.g., La, Th), such a clear-cut correlation cannot be separated out.)

MAGNETORESISTANCE

In the present studies we concentrated on two pseudobinary systems, $(U_{1-x}Y_x)Be_{13}$ and $(U_{1-x}Th_x)Be_{13}$. The 14-T depression of ρ at 4.2 K is shown in Figs. 3 and 4 for Y and Th alloying, respectively. Both Th and Y substitutions lead to dramatic effects in the magnetoresistance. An especially steep reduction of the negative magnetoresistance is observed at the 1–4 % level of impurities with the initial depression of magnetoresistance for Th alloying larger than for Y alloying. The 4.2 K magnetoresistance of $U_{1-x}Y_xBe_{13}$ exhibits a tendency towards saturation for $x>0.2$, while in the case of $U_{1-x}Th_xBe_{13}$ the magnetoresistance becomes positive for $x>0.4$.

Let us now consider this positive magnetoresistance in the case of $U_{1-x}Th_xBe_{13}$. Recall that the single-ion Kondo model describes the magnetic part, ρ_m, of the total resistivity ρ. Therefore, a comment on the difference between total ρ and ρ_m is in order. The total resistivity of a pseudobinary compound $AB_{1-x}C_x$, obeying a Matthiessen's rule, can be written as a sum of ρ_{imp}, ρ_{phonon}, ρ_{Nordheim}, and ρ_m, where the first three subscripts stand for the resistivity due to chemical impurities and lattice imperfections, resistivity due to electron-phonon scattering, and resistivity due to a random distribution of B and C ions, respectively.[21] Thus, extraction of ρ_m from ρ is, in general, a formidable and not very reliable procedure. Even in a system such as $U_{1-x}Th_xBe_{13}$, where the resistivity seems to be completely dominated by its magnetic contribution, the nonmagnetic part cannot be completely neglected. This can be seen from Table I, where ρ, $\tilde{\rho}$ ($\tilde{\rho}$ denotes the difference between ρ of $U_{1-x}M_xBe_{13}$ and ρ of MBe_{13} in the same field and at the same temperature) and their changes in 14 T at 4.2 K for several compositions are assembled. (The resistivity of MBe_{13} is used to approximate $\rho_{\mathrm{imp}}+\rho_{\mathrm{phonon}}$ of $U_{1-x}M_xBe_{13}$. We are unable to account for the disorder

TABLE I. Resistivity for some $(U_{1-x}Th_x)Be_{13}$ and $(U_{1-x}Y_x)Be_{13}$ alloys at 4.2 K and in $H=0$ or 14 T.

	$\rho(H=0)$ ($\mu\Omega$ cm)	ρ(14 T) ($\mu\Omega$ cm)	$\tilde{\rho}(0)$ ($\mu\Omega$ cm)	$\tilde{\rho}$(14 T) ($\mu\Omega$ cm)	$\frac{\rho(14\ \mathrm{T})}{\rho(0)}$	$\frac{\tilde{\rho}(14\ \mathrm{T})}{\tilde{\rho}(0)}$
UBe_{13}[a]	230	166	226.5	160.4	0.72	0.71
$U_{0.1}Th_{0.9}Be_{13}$	9	9.85	5.5	4.25	1.09	0.77
UBe_{13}[b]	230	166	227	158	0.72	0.70
$U_{0.97}Y_{0.3}Be_{13}$	235	180	232	172	0.77	0.74
$U_{0.8}Y_{0.2}Be_{13}$	220	205	217	197	0.93	0.91
$U_{0.6}Y_{0.4}Be_{13}$	166	162	163	154	0.975	0.94
$U_{0.2}Y_{0.8}Be_{13}$	106	104.5	103	96.5	0.986	0.94
$U_{0.1}Y_{0.9}Be_{13}$	51.6	50.9	48.6	42.9	0.986	0.88

[a] $\tilde{\rho}$ calculated using $ThBe_{13}$ results.

[b] $\tilde{\rho}$ calculated using YBe_{13} results.

term; however, the very unsymmetrical shape of the residual resistivities of $(U_{1-x}Y_x)Be_{13}$ and $(U_{1-x}Th_x)Be_{13}$ with respect to $x=0.5$ argues for a small magnitude of this term in comparison with ρ_m.) As can be seen from Table I, $\tilde{\rho}(14\ \mathrm{T})/\tilde{\rho}(0)$ for $U_{0.1}Th_{0.9}Be_{13}$ at 4.2 K is close to 0.77, which is almost as small as $\rho(14\ \mathrm{T})/\rho(0)$ or $\tilde{\rho}(14\ \mathrm{T})/\tilde{\rho}(0)$ for undoped (coherent) UBe_{13}. We believe that this result is a consequence of overestimating the nonmagnetic part of the $U_{0.1}Th_{0.9}Be_{13}$ resistivity. On the other hand, the corresponding ratios, $\tilde{\rho}(14\ \mathrm{T})/\tilde{\rho}(0)$ and $\rho(14\ \mathrm{T})/\rho(0)$ for $(U_{1-x}Y_x)Be_{13}$ are reasonably close to one another and a somewhat quantitative analysis is possible.

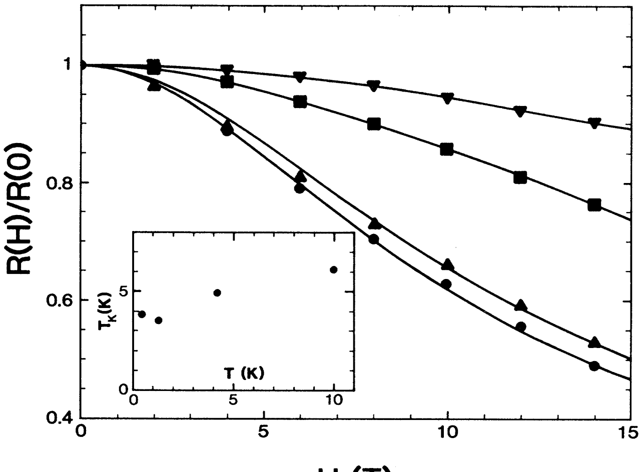

FIG. 6. Resistance of $U_{0.97}Y_{0.03}Be_{13}$ vs field for $T=0.45$ K (●), 1.3 K (▲), 4.2 K (■), and 10 K (▼). Inset shows T_K vs T.

Let us therefore focus on $U_{1-x}Y_xBe_{13}$. In Figs. 5, 6, and 7 we plot $R(H)/R(0)$ isotherms for UBe_{13}, $U_{0.97}Y_{0.03}Be_{13}$, and $U_{0.8}Y_{0.2}Be_{13}$. All isotherms were fit to a formula derived for a $S=1/2$ Coqblin-Schrieffer model[20] with only two parameters: the g factor and T^*, where $T^*=T+T_K$. T_K is proportional to the single energy scale (Kondo temperature) and is independent on a magnetic field and temperature. However, in order to describe our data qualitatively, we purposely lifted the last requirement (temperature independence) and assigned a different T_K for each isotherm. Following Refs. 22 and 23 we assumed the following relation between a magnetic field value (in Tesla) and temperature (in K): 1 $T \leftrightarrow g\mu_B \times (1\ \mathrm{T})=0.49$ K ($g=8/11$ for U^{3+}). The magnetoresistance of undoped UBe_{13} was measured at 1.3, 4.2, and 10 K. The best fits for the UBe_{13} data were obtained with the following T_K's: 1.6, 4, and 6.1 K for these temperatures, respectively (inset of Fig. 5). These results are in an agreement with others previously reported.[8,9,10] In particular, the T_K parameter is strongly temperature dependent and approaches a very small value for $T \rightarrow 0$ K. Similarly obtained T_K for $U_{0.97}Y_{0.03}Be_{13}$ have much weaker temperature dependence, and approaches $T_K \approx 4$ K for $T=0$ K (Fig. 6). The magnetoresistance for $x=0.2$, thus a concentration for which the magnetoresistance ceases to be strongly concentration dependent, is displayed in Fig. 7. T_K, derived from the magnetoresistance curves at 0.4, 4.2, and 10 K is within the 16–18 K temperature range. (If T_K is calculated not from total ρ, but from $\tilde{\rho}$, defined previously, all T_K values for $U_{0.8}Y_{0.2}Be_{13}$ are about 3 K lower.) Therefore, it can be concluded that, within the accuracy of our analysis, T_K obtained for $U_{0.8}Y_{0.2}Be_{13}$ is temperature independent and is of the order of 16 K. T_K calculated within the same model from the low-temperature susceptibility, $T_K=\mu_B^2/4\chi_0$ ($\chi_0 \simeq 13$ memu/mol), is about 7.2 K and is independent on Y concentration. Assuming the relevancy of the Kondo model, there are at least two major possible sources for the disagreement between the characteristic temperatures derived from the magnetic susceptibility and magnetoresistance of $U_{0.8}Y_{0.2}Be_{13}$. First, the g factor used is only a crude approximation and it can be strongly renormalized for the ground-state doublet with respect to the free ion value, and second, the Van Vleck

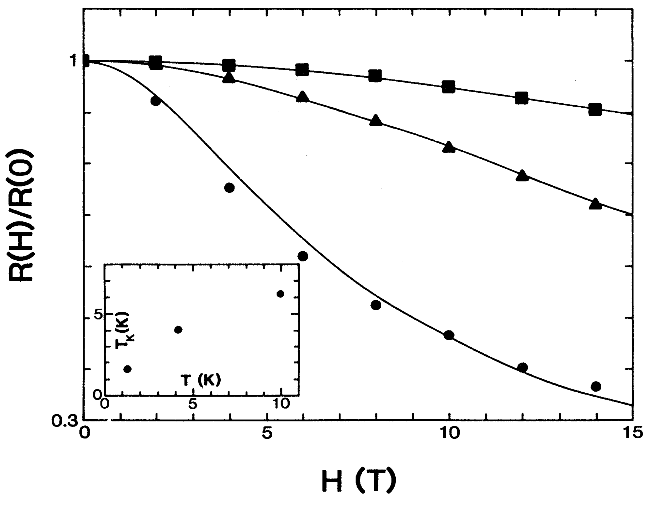

FIG. 5. Resistance of UBe_{13} vs field for $T=1.3$ K (●), 4.2 K (▲), and 10 K (■). Inset shows T_K vs T (see text).

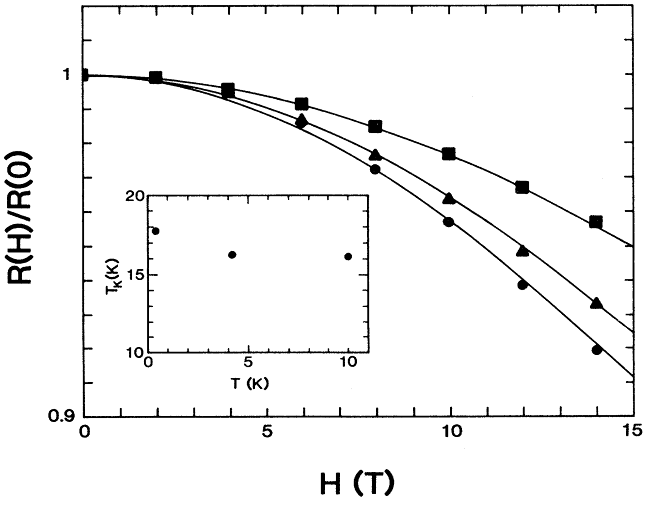

FIG. 7. Resistance of $U_{0.8}Y_{0.2}Be_{13}$ vs field for $T=0.4$ K (●), 4.2 K (▲), and 10 K (■). Inset shows T_k vs T.

mechanism can significantly enhance the magnetic susceptibility. The ratio of the low-temperature susceptibility and $\gamma(\sim 400\ \mathrm{mJ/K^2\,mol})$ is larger than for typical Ce Kondo impurity systems with the ground-state doublet [e.g., it is about 1.5 times larger than for (Ce,La)Pb_3 system]. This strongly indicates that the susceptibility of UBe_{13} can be significantly effected by the Van Vleck mechanism.

In order to explain the unusual temperature dependence of the magnetoresistance of pure UBe_{13}, a coherent Kondo lattice formation idea has been invoked.[8] This idea gained some support from the fact that only 3% of Th impurities introduced for U dramatically alter the magnetoresistivity, in particular decrease its temperature dependence.[10] Our results are certainly in agreement with this line of reasoning. Note that the relative magnetoresistance vs x plots for $U_{1-x}Y_xBe_{13}$ and $U_{1-x}Th_xBe_{13}$ (Figs. 3 and 4) bear a remarkable resemblance to the Sommerfeld coefficient γ vs x dependences for these systems.[11] It has been suggested[11] that about 60% of the large γ value for UBe_{13} results from intersite correlation effects in the U sublattice. A similar explanation for the magnetoresistance changes seems to be the most natural. Note that these changes, which are similar for Y and Th doping, are not related to either lattice pressure effects or the electronic configuration of the doping element since Th is four valent and larger than U in UBe_{13} while Y is trivalent and slightly smaller than U in UBe_{13}. The T_K parameter, derived from the magnetoresistance, becomes temperature independent for sufficiently diluted UBe_{13} ($U_{0.8}Y_{0.2}Be_{13}$) as expected for the Kondo impurity system. Whether a Kondo lattice (UBe_{13}), on the other hand, can be described in terms of a Kondo system with a temperature dependent T_K is an interesting question (posed already in Refs. 8 and 24) and should be addressed theoretically.

Our work allows us to explain the apparent lack of any correspondence between high field magnetization and magnetoresistance of UBe_{13}, a problem first pointed out in Ref. 8. Our extensive alloying study of several $(U_{1-x}M_x)Be_{13}$ systems[11] revealed that the magnetic susceptibility as measured at $T=2$ K and in 0.5 T field, scales with U concentration and does not depend on x or the metal element M. Magnetization of UBe_{13} is then a single-ion property, contrary to the magnetoresistance or the specific heat. It is more appropriate then to compare UBe_{13} magnetization with magnetoresistance of $U_{1-x}Y_xBe_{13}$ alloys with $x \geq 0.2$ (where the relative magnetoresistance for these alloys is only weakly concentration dependent, see Fig. 3). According to the Kondo model, an 8% decrease in the resistivity (1.5 K, 14 T for $U_{0.8}Y_{0.2}Be_{13}$) corresponds to only about a 4% deviation from linearity for the magnetization M vs H. Our magnetization measurement for pure UBe_{13} showed about 6.6% deviation from linearity at $T=1.4$ K and $H=20$ T (Ref. 25), certainly a consistent result.

In summary, our results indicate that magnetoresistance is another property, after specific heat and low-temperature resistivity, sensitive to the U-U correlations existing in undoped UBe_{13}. A very large negative magnetoresistance at low temperatures is a symptom of the correlation effects in UBe_{13} in contrast to the behavior of Ce compounds, such as $CeCu_2Si_2$ and $CeAl_3$, where positive magnetoresistance is taken as a sign of the correlations in Ce lattices. Although we can not perform an accurate and detailed analysis of the magnetoresistance of strongly diluted UBe_{13} samples, we observe a clear tendency in magnetoresistance behavior upon dilution. When the correlation effects in UBe_{13} are suppressed, both the magnetoresistance and magnetization can be self-consistently described by the Kondo model. This is an important observation since this discrepancy in the sensitivity of these two properties to magnetic field in UBe_{13} was one of the most important motives for the formulation of other heavy-fermion theories, like a quadrupolar Kondo model.[3] The zero-field resistivity results are consistent with a localized character of $5f$ electrons in UBe_{13}. Yet small variations in U-Be separation lead to dramatic effects in the resistivity that can be explained by either further localization or delocalization of U $5f$ electrons. This makes UBe_{13} and its alloys a convenient system to study the formation of the heavy-fermion ground state and especially the role of crystal fields.

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