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INVITED PAPER

MAGNETISM OF ACTINIDE COMPOUNDS

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Actinide compounds exhibit most of the known magnetic phenomena. Their properties are easily modified in intermetallic compounds. We present some new results on uranium ferromagnets and heavy fermion antiferromagnets and explain their implications. We show magnetic ordering in a high T_c superconductor.

The actinide, 5f-electron elements lie between the 4f and 3d series in the degree of their localization. The 3d elements never do form local moment magnets while the 4f elements do so at the beginning of the series. All of these transition metal series begin by showing bonding or itineracy of their respective d or f electrons. As the shells fill, the pertinent wave functions become more compact, and this leads toward electron localization and magnetism in the more atomic-like wavefunctions. As the elements pass through this crossover in behavior, there is a great sensitivity of the electronic structure to perturbations, and there are many possible ground states. This means that more interesting physics takes place here [1]. The simplest systematic way to explore all of this is to study the metallic compounds.

For the actinides, Hill showed that the behavior of compounds of the lighter elements was largely determined by the spacing between the actinide atoms, independent of the intervening atoms [2]. Thus closely spaced f atoms have overlapping or strongly hybridized wavefunctions, and these itinerant f-electrons can become superconducting. Widely spaced f-electron atoms tends to have more localized wave functions and may show magnetism. Hill's criterion survived reasonably well until heavy fermion superconductors showed up with their spacing well into the magnetic region [3]. The heavy fermion ground states of superconductivity, antiferromagnetism, and Fermi-liquid

behavior show up in compounds that have their f electrons on the very edge of being fully localized.

The heavier actinide elements and compounds would form local moment magnets, but the elements are often too difficult to handle or even to produce [4]. Fortunately the most interesting behavior occurs in the lighter elements that are more readily available. Here we find such things as itinerant magnets, large magnetocrystalline anisotropies, and the so-called heavy fermion magnets. We will briefly discuss some recent results on these topics and then show a recent, rather trivial example of local moment behavior as a contrast to these subjects.

There are some families of compounds of the rare earths in which the cerium or neodymium members order at temperatures close to that of the gadolinium member such as CeBi, CeSb, and CeRh₃B₂ [5]. However, for conventional spin-driven magnetic interactions it is instead expected that ordering temperatures for a family should decrease on both sides of the gadolinium maximum as $s(s+1)$ or faster, see fig. 1. Furthermore, the same exceptions show up for some uranium and plutonium magnets, but because there are fewer measured actinide compounds, the overall pattern is less obvious than for the 4f elements. For both 4f and 5f elements, within the transition region from itinerant to localized behavior, the rather diffuse f-electron ionic charge can engage in an orbitally-driven magnetic interaction [6]. Thus f-electrons can interact with each other through a hybridization with non-f-electron atoms. This very anisotropic interaction is in parallel with the

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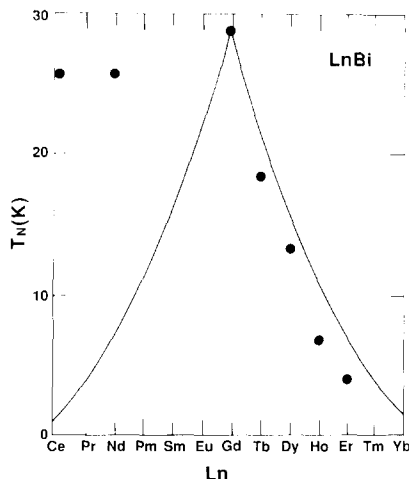


Fig. 1. Ordering temperatures for lanthanum bismuth compounds. Curve is proportional to $s(s+1)$ and normalized to gadolinium.

RKKY interaction and can yield the observed higher ordering temperature, as well as the even more common reduced, ordered moments.

Along with higher ordering temperatures, another parameter that can be of technological value is magnetocrystalline anisotropy, which is one of the major factors in producing large coercive fields in ferromagnets. Orbitaly driven magnetism is extremely anisotropic because it is a first order effect rather than a secondary effect as is the RKKY interaction. It is well known that neodymium is useful in practical ferromagnets. We thought we would see what could be done with uranium. We began with UCu_2Si_2 , which has the ThCr_2Si_2 -type structure. We measured our polycrystalline, arc-melted, and annealed samples with a vibrating sample magnetometer. They had a Curie temperature of 100 K and showed a saturation magnetization of 7.7 emu/g and a coercive field of 14 kOe. Then we made replacements of the copper with iron and manganese. The highest T_c of 145 K was achieved for $\text{UCu}_{1.49}\text{Mn}_{0.51}\text{Si}_2$, and in fig. 2 we show a hysteresis loop at 4 K for this compound. The saturation magnetization and coercive fields for these two series of compounds are shown in figs. 3 and 4. No attempt was made to improve the coercive field by modification of the microstructure of these materials. We note simply that as in the study of permanent magnets,

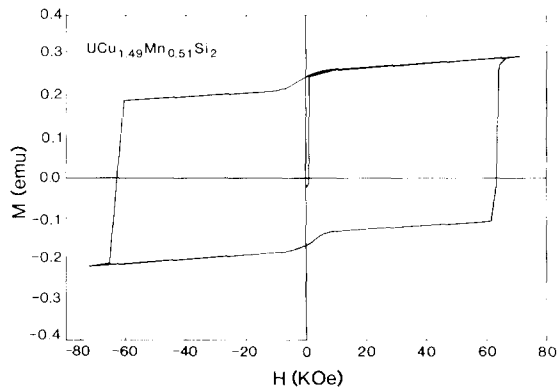


Fig. 2. Hysteresis loop for $\text{UCu}_{1.49}\text{Mn}_{0.51}\text{Si}_2$ taken in a vibrating sample magnetometer at 4 K.

uranium can be as useful as the light rare earths for producing coercive fields of practical magnitudes.

The heavy fermion compounds represent a new class of materials with several possible ground states [3,7,8]. The name originates from the low-temperature, electronic heat capacity that indicates an effective mass for the conduction electrons that is several hundred times that of a free electron. At higher temperatures these compounds behave as a rather ordinary collection of decoupled spins, a Kondo impurity system, that could be expected to order antiferromagnetically. How-

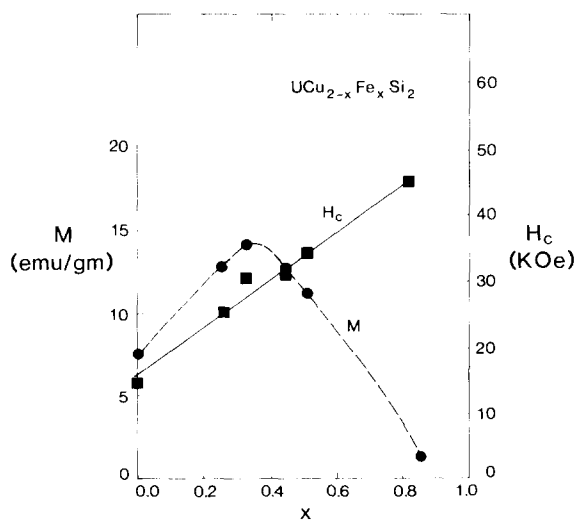


Fig. 3. Saturation magnetization and coercive fields for $\text{UCu}_{2-x}\text{Fe}_x\text{Si}_2$ at 4 K.

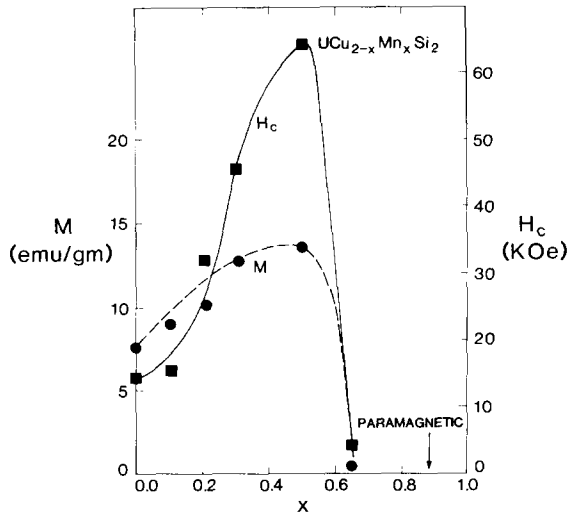


Fig. 4. Saturation magnetization and coercive fields for $UCu_{2-x}Mn_xSi_2$ at 4 K.

ever, at temperatures near 20 K the electrons begin to form a coherent electron state that resembles a Fermi liquid. The spin system becomes a Pauli paramagnet. Some of these compounds are superconducting, and one reason they are currently believed to be non-electron-phonon superconductors is that their properties are so dominated by magnetic interactions. Other heavy fermion compounds, such as U_2Zn_{17} , order antiferromagnetically. Their electronic heat capacity shows that a sufficiently small part of the Fermi surface is involved in the magnetic ordering that the electronic heat capacity of the remaining Fermi surface continues to increase at lower temperatures. In spite of an extremely simple magnetic structure [9], these are clearly unusual antiferromagnetic compounds. Indeed they are magnets that sit between typical itinerant-electron and local-moment magnets [3]. Their magnetic ordering is driven by the temperature dependence of the RKKY interaction, rather than the usual increase of the single-ion susceptibility [8,10]. These antiferromagnets have their Kondo coupling and RKKY interactions sufficiently well balanced that a smaller energy scale can emerge to dominate the low-temperature properties [10].

This fence-sitting of the properties of U_2Zn_{17} has been demonstrated by the unusual sensitivity of the Néel temperature to depression by impuri-

ties. This sensitivity has been seen with copper on the zinc sites [11] and cerium on uranium sites [12]. We have now substituted thorium on the uranium sites and deduced the Néel temperature from the peak in a plot of heat capacity divided by temperature versus temperature. This peak correlates well with the T_N determined from susceptibility and resistivity measurements. Fig. 5 shows the depression which occurs at a rate of about -0.5 K/at% Th. For all of these impurity substitutions, the rate of depression is about -4.5 K/% of total atoms replaced in the compound. As these impurity atoms have very different electronic properties and substitute for either uranium or zinc, it seems that the effect of impurities is simple disorder. This compound is then unusually sensitive to impurities for an antiferromagnet because it is sitting so very delicately between itinerant and local electron behavior.

In contrast to the rich variation of behavior as f electrons begin to localize, fig. 6 shows magnetic ordering of gadolinium spins at $T_N = 2.24$ K for a crystallographically ordered compound of gadolinium [13]. From the data, which includes an extrapolation to $T = 0$ in the inset, we can see that the full magnetic entropy of $R \ln 8$ is available below $T = 6$ K. This is a fine example of what we call local-moment behavior. We show it here at a symposium on magnetism in intermetallic compounds because the same compound shows an

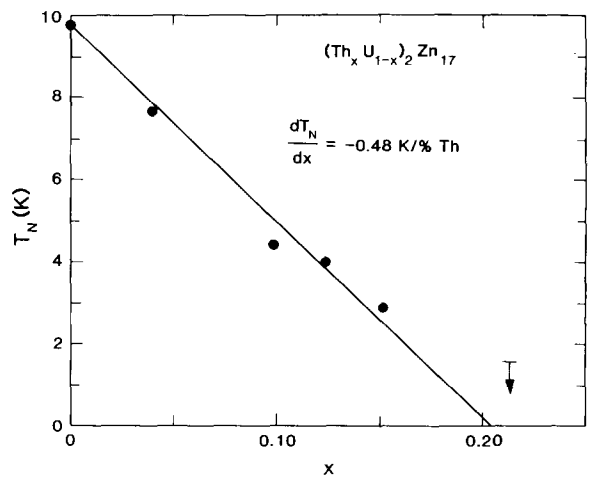


Fig. 5. Depression of Néel temperature of U_2Zn_{17} by thorium impurities.

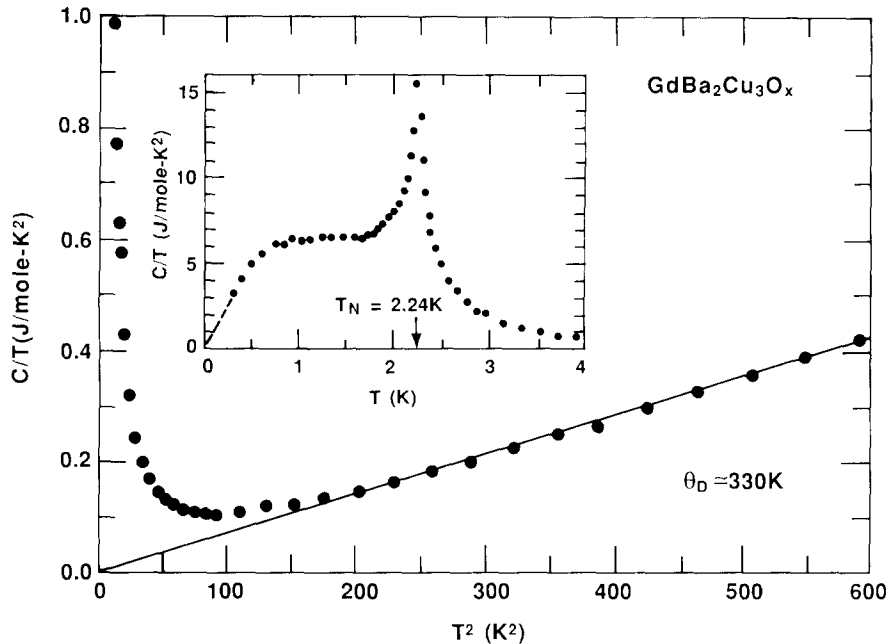


Fig. 6. Heat capacity divided by temperature plotted as a function of T and T_2 for $\text{GdBa}_2\text{Cu}_3\text{O}_x$. This compound shows an onset of superconductivity at (93 ± 1) K by dc susceptibility measurements.

onset of superconductivity at $T = 93$ K. Although we may not learn much physics from this magnetic ordering, clearly there is a place for the new high T_c superconductors at our symposium.

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