UMn_2Al_{20}: Small Itinerant Moment or Induced Local Moment Ferromagnetism?

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Abstract. The magnetic susceptibility, specific heat, resistivity and inelastic neutron scattering spectrum were measured on the cubic heavy Fermion compound UMn_2Al_{20}. A ferromagnetic transition is observed in the magnetic susceptibility at T_c=20 K, but no anomaly is seen in the specific heat or resistivity at T_c. This behaviour is similar to that of the single-triplet induced moment ferromagnet Pr_3Tl. Since the 5f electrons of uranium have the possibility of being in the $5f^2$ electron configuration with a nonmagnetic J=0 ground state and a triplet excited state, such induced moment behaviour is a possibility in UMn_2Al_{20}. Since the linear coefficient of specific heat at T = 0 is large (0.3 J/mol-K^2) a second possibility is moderately small (~ 0.9 μ_B) moment itinerant ferromagnetism in a heavy Fermion system. In the energy transfer range 5 < ΔE < 60 meV the inelastic neutron scattering spectra appear to be dominated by phonon scattering, with no obvious magnetic scattering. Hence, if either scenario is correct, there must be magnetic excitations at lower (1-5 meV) energies.

1. Introduction

Due to the large spatial extent of the 5*f* orbitals in actinide compounds, the *f* electrons typically exhibit band-like behavior. For example, the magnetic order in such compounds as UGe_2 [1] is clearly itinerant ferromagnetism. Localized 5*f* behavior and clearly defined crystal field excitations are rare in the metallic U compounds, but there are exceptions, such as UPd_3[2]. A "dual nature" of 5*f* electrons, where some of the 5f electrons are localized while others are itinerant, has also been suggested [3,4]. Therefore, whether the 5*f* electrons are localized or itinerant remains as an important issue for uranium intermetallic compounds, and more detailed experiments on good crystals are needed.

UMn_2Al_{20} is a member of the family of lanthanide and actinide compounds RT_2M_{20} (R=Ce, Yb, U; T=transition metal; M=Zn, Al). These materials crystallize with the $CeCr_2Al_{20}$ cubic structure (F d -3 m) and display several interesting features, such as heavy Fermion or intermediate valence behavior [5,6,7]. In this family, the fraction of lanthanide or actinide atoms in the crystal is relatively small (~ 4

%), allowing for studies close to the impurity limit, but in an ordered system. Furthermore, the *f*-atoms are separated by a U-U distance greater than 6 Å and are surrounded by a nearly spherical cage of Zn atoms in cubic site symmetry, which makes for very small crystal field splitting[5].

In this paper, we present low temperature magnetic susceptibility, specific heat, resistivity and inelastic neutron scattering measurements for UMn_2Al_{20}. The magnetic susceptibility shows a ferromagnetic phase transition at 20K but no associated anomaly is observed in the specific heat or resistivity. The inelastic neutron scattering spectrum shows no obvious magnetic excitations in the energy range 5 to 60 meV. We propose that the magnetic behaviour is either due to induced ferromagnetism arising from a low energy (1-5 meV) singlet-triplet crystal field excitation of localized 5f electrons, or is due to heavy Fermion ferromagnetism of itinerant 5*f* electrons.

2. Experimental details

Single crystals were grown in Al flux with an elemental starting ratio U:Mn:Al=1:1:50. The crucible was sealed under vacuum in a quartz tube. The sample was heated to 1050^0C quickly in order to avoid the reaction between Al and the quartz tube, held at 1050^0C for 4 h, then cooled at a rate 5^0C/h to 700^0C. At this point the excess Al flux was removed by using a centrifuge. The magnetization was measured in a commercial superconducting quantum interference device (SQUID) magnetometer. The specific heat measurements were performed in a commercial physical properties measurement system (PPMS). The electrical resistivity was also measured in the PPMS using the four wire method. The inelastic neutron scattering experiment was performed on a 35 gram powder sample using the high resolution chopper spectrometer (Pharos) at the Lujan Center, LANSCE, at Los Alamos National Laboratory.

2. Results

The magnetic susceptibility $\chi(T)$ of UMn_2Al_{20} is shown in figure 1. The data shows a dramatic enhancement at low temperature (see inset (a)), indicating a ferromagnetic transition at $T_c \approx 20K$. A Curie-Weiss fit of the form $\chi(T)= C/(T+\theta)$ at high temperature (T > 295 K) gives an effective moment $\mu_{eff} = 5.03\ \mu_B$ and a large Weiss temperature as $\theta = 238.6K$. Since the data for $1/\chi$ are not actually linear, and since this value of Curie constant is much larger than the value for either the $5f^2$ ($\mu_{eff} = 3.58\ \mu_B$) or the $5f^3$ ($\mu_{eff} = 3.62\ \mu_B$) configuration, we do not view this fit as meaningful. The data can also be fit to the formula $\chi(T) = \chi_0 + C/(T-\theta)$ where $\chi_0 = 0.0023$ emu/mole, C=1.0101 emu-K/mole (μ_{eff} =2.84), and $\theta = 23.1$ K. While the value of θ for this fit is close to the magnetic phase transition temperature, the Curie constant seems too small and the value of $\chi_0 = 0.0023$ too large for the fit to be meaningful. While we are thus uncertain as to the interpretation of the susceptibility at high temperature, ferromagnetic fluctuations appear to play an important role.

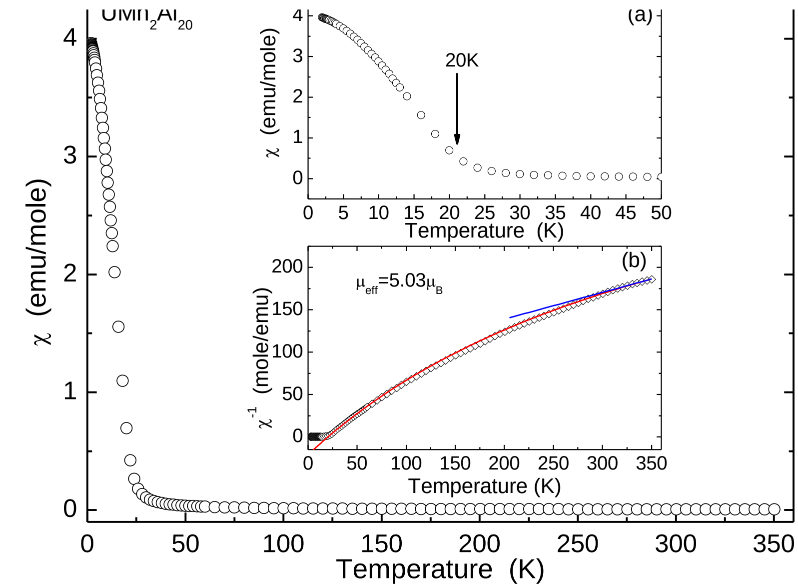

Figure 1: Magnetic susceptibility χ(T) for UMn_2Al_{20} at H = 0.1 Tesla. Inset (a) shows zero field cooling (open circles) and field cooling curves (red line). Inset (b) gives the inverse magnetic susceptibility $\chi^{-1}(T)$. The blue line is the high temperature Curie-Weiss fit. The red line in inset is a fit to the formula $\chi(T)=C/(T-\theta)+\chi_0$ with C=1.0101 emu-K/mole, θ=23.1 K and χ_0 = 0.0023 emu/mole.

The isothermal magnetization results at 2 K and 5 K of UMn_2Al_{20} are displayed in figure 2. The full hysteresis loop at 2 K is shown in the inset. Both the coercive field and remnant magnetization are very small (H_c~ 9 Oe, M_R ~ 0.03 μ_B), indicating that UMn_2Al_{20} is a soft ferromagnet. Linear fits to the magnetization data give the saturation magnetization as $M_{sat}(2K)$ = 0.90 μ_B and $M_{sat}(5K)$ = 0.81 μ_B.

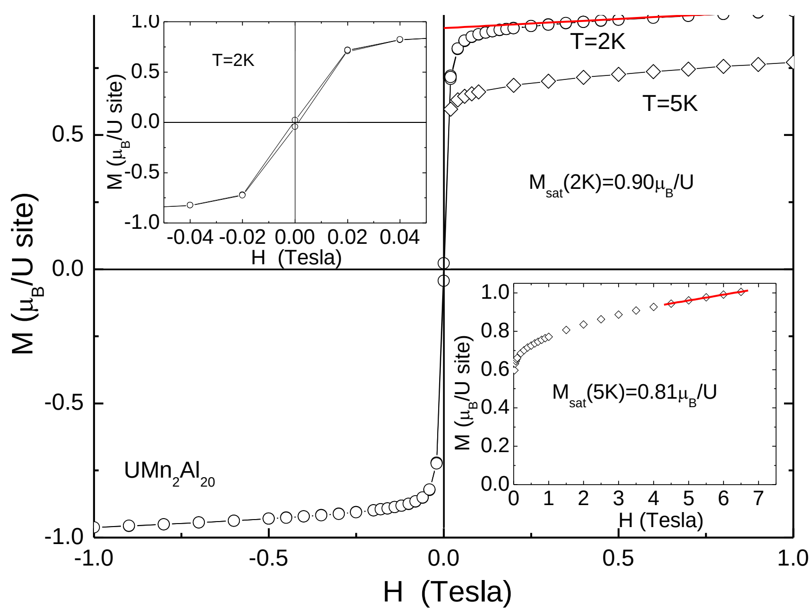

Figure 2 Magnetization M(H) of UMn_2Al_{20} at 2K (open circles) and 5K (open diamonds).

The red line is linear fitting to data. Inset is the hysteresis loop at 2K.

Specific heat measurements on UMn_2Al_{20} and the nonmagnetic counterpart ThV_2Al_{20} are shown in figure 3. There is no obvious anomaly in the total (as measured) specific heat of UMn_2Al_{20} near T_c = 20K. The magnetic contribution to the specific heat C_{mag} is obtained by subtracting the lattice contribution to the specific heat of the nonmagnetic counterpart ThV_2Al_{20}. The resulting C_{mag} and C_{mag}/T are shown in the insets of figure 3 (a) and (b), respectively. The quantity C_{mag} shows a peak at around 16 K which corresponds to a small anomaly in the slope of C_{mag}/T at the same temperature. The entropy associated with the magnetic specific heat is shown in figure 3 (b), giving a value for the magnetic entropy of Rln2 at 46 K. The entropy curve shows a curvature change at T ~ T_c.

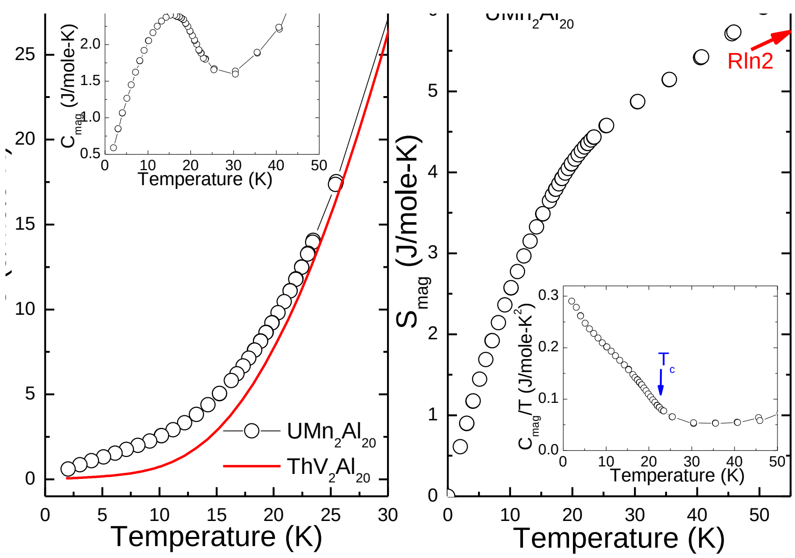

Figure 3 (a) Specific heat of UMn_2Al_{20} and ThV_2Al_{20}. (b) Entropy associated with the magnetic specific heat. The inset of (a) shows the magnetic contribution to the specific heat. The inset of (b) is the linear coefficient C_{mag}/T

The electrical resistivity of UMn_2Al_{20} is shown in figure 4. The data decreases with decreasing temperature down to 10 K, below which it is constant. There is no anomaly at 22 K associated with the ferromagnetic transition. To enhance the possibility of a tiny anomaly in the resistivity, we display the temperature differential curve $d\rho(T)/dT$ in the inset (a); again, there is no obvious anomaly.

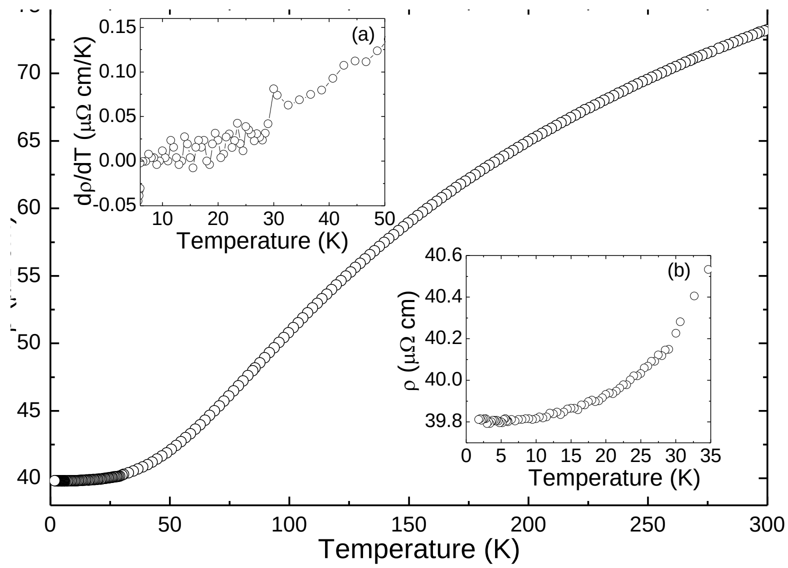

Figure 4 Electrical resistivity ρ(T) of UMn_2Al_{20}. Inset (a) is the temperature differential curve.

3. Discussion

The magnetization measurements show clearly that there is a ferromagnetic transition in UMn_2Al_{20} at $T_c \approx 20$ K. However, there is no anomaly in the electrical resistivity associated with this transition, nor is there any obvious anomaly in the total (as measured) specific heat C(T). An anomaly in C(T) only becomes apparent after subtraction of the specific heat of the counterpart compound, ThV_2Al_{20}. The resulting peak in C_{mag}(T) is quite broad, although the entropy $S_{mag}(T_c)$ is a large fraction of R ln 2. The linear coefficient of specific heat C_{mag}/T appears to be very large (~ 0.3 J/mol-K^2) at the lowest temperatures, suggesting that the ferromagnetic order occurs within a heavy Fermion state. It is plausible that the moderately small moment (0.90 μ_B), the weak anomaly in C(T) at T_c, and the absence of an anomaly in ρ(T) at T_c may reflect heavy Fermion itinerant ferromagnetism.

A small anomaly in the specific heat was observed at the spin glass transition temperature in $Hg_{0.65}Mn_{0.35}Te$ [8]. However, from the rapid and large enhancement of the magnetic susceptibility of UMn_2Al_{20}, where the value of the susceptibility at 2 K is more than 20 times larger than the value above 20 K, spin glass behavior is very unlikely.

Small anomalies in C(T) and ρ(T) were also observed in Pr_3Tl[9] (Pr_3In[10]), where *induced* ferromagnetic (antiferromagnetic) order occurs at 12 K [11, 12]. For these compounds, the Pr^{3+} $4f^2$ ground multiplet is split by the crystal field such that the Γ_1 singlet is the ground state and the Γ_4 triplet is the lowest excited state. The Γ_1 ground state couples with Γ_4 triplet states through the intersite magnetic exchange interaction to induce a magnetic moment on the ground state [13,14]. In mean field theories of the induced magnetic order, the ordering occurs within the singlet without loss of degeneracy, so that a very weak anomaly in the specific heat and resistivity is expected, reflecting the lack of a significant magnetic entropy change at the magnetic transition temperature. This has been taken as the explanation of the small anomalies in C(T) and ρ(T) in Pr_3Tl and Pr_3In.

In UMn_2Al_{20}, the uranium 5f electrons have the possibility of being in a $5f^2$ local moment configuration with a nonmagnetic J=0 ground state and a triplet excited state, which is the same as $4f^2$ configuration as in the rare earth Pr^{3+}. Coupled with the absence of a specific heat C(T) and electrical resistivity ρ(T) anomaly at the transition temperature, this raises the possibility that this compound has a similar induced local moment behaviour.

It has been proposed that the phase transition in induced moment systems is actually brought about by a softening of the crystal field excitation at the Q vector which corresponds to the magnetically ordered phase (Q = 0 for ferromagnetism; $Q = Q_N$ for antiferromagnetism). At a temperature much higher than the ordering temperature, well-defined non-dispersive crystal field excitations are expected but in the ordered state the singlet-triplet excitation would be dispersive [13,14,15]. These effects should be readily observable in neutron scattering spectra.

In order to check for induced moment behaviour, we performed inelastic neutron scattering on a polycrystalline sample of UMn_2Al_{20}. The spectra are shown in figure 5. There are no obvious magnetic excitations; all the peaks appear to be phonon contributions. To explore this further, we utilize the observation that in the neutron scattering spectra measured[6] for the nonmagnetic compound $ThCo_2Zn_{20}$, the phonon contribution measured at high Q is roughly 3 times larger than at low Q. If we scale the high Q spectra of UMn_2Al_{20} by 1/3 we find that the low Q spectra are nearly identical with the scaled curve (blue lines in Fig. 5) for energy transfer $\Delta E > 10$ meV, suggesting that there are no magnetic excitations in this energy range. However, the scaled data appears to be smaller than the low-Q data at smaller energy transfer, so that there may be a magnetic contribution in the 1-5 meV range. As mentioned above, in this crystal structure, the uranium atom is surrounded by 16 Zn atoms to form a nearly spherical cage; and the resulting crystal field splittings in the corresponding rare earth compounds are small, occurring at these small (1-5 meV) energies[5]. Therefore, the possibility remains that there is a crystal field excitation below 5 meV in UMn2Al20. The inelastic neutron scattering spectra shown in Fig. 5b gives a hint that there may be such scattering around 4 meV. Further neutron scattering experiment on a higher resolution instrument are needed to confirm this.

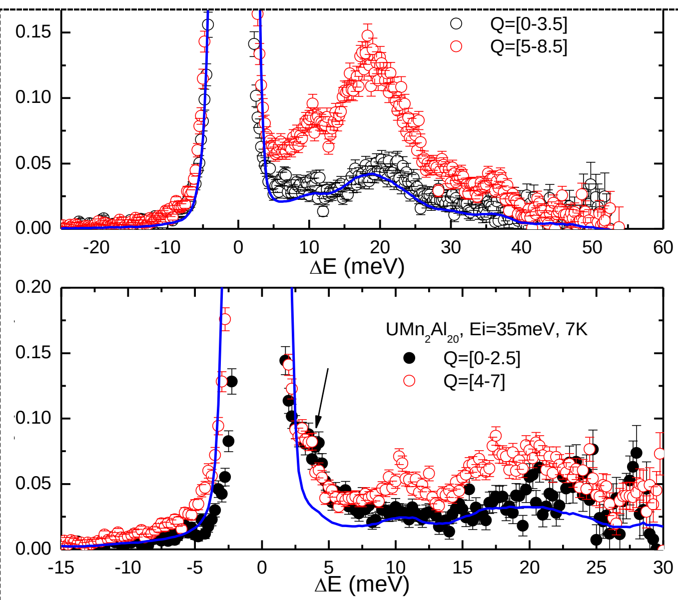

Figure 5. The inelastic neutron spectra of UMn_2Al_{20} measured on Pharos with two different incident energies. The black open circles are the low Q spectra, the red open circles are the high Q spectra. The blue line is the high Q data divided by 3.

In summary, a ferromagnetic transition was observed in magnetization measurements on UMn_2Al_{20} but no strong anomaly was observed in the specific heat and electrical resistivity measurements. There appear to be two possibilities to explain this behaviour: moderately small moment itinerant ferromagnetism occurring in a heavy Fermion state, and singlet-triplet induced local moment behaviour. The inelastic neutron scattering spectra show no magnetic excitations between 5 meV to 60 meV which would correspond to either the spin fluctuation scattering expected for heavy Fermion compounds or the crystal field excitations expected for a singlet-triplet induced moment system. Since the crystal field excitation in this compound is expected to be smaller than 5 meV, further neutron scattering experiment at higher resolution are in order.

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