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RAPID COMMUNICATIONS

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Specific heat, magnetic susceptibility, and resistivity of an $U_{0.85}La_{0.15}Al_2$ alloy

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The compound UAl_2 exhibits ferromagnetic spin fluctuations at low temperatures. When 15 at.% La is substituted for U in UAl_2, we find the low-temperature magnetic susceptibility increases by more than a factor of 2. We have also measured specific heat on this sample at low temperatures. We find that the electronic density of states has increased by 60% and the spin-fluctuation contribution has increased by a factor of 2. We conclude from these measurements that the Stoner enhancement factor has increased by ~30% and the spin-fluctuation temperature has increased by ~50% as compared with UAl_2. In addition, we have measured the electrical resistivity of $U_{0.85}La_{0.15}Al_2$. We find the residual resistivity to be approximately an order of magnitude larger than that of UAl_2. We attribute this effect to the breaking of U–U bonds by the La atoms.

INTRODUCTION

There has been considerable interest recently in materials systems that are nearly magnetic. Among these materials are those that are thought to be spin-fluctuation systems, e.g., UAl_2,[1] $TiBe_2$,[2] Pd,[3] Sc,[4] $LuCo_2$,[5] and $CeSn_3$.[6] Of these, only the first two exhibit the $T^3 \ln T$ term in the low-temperature specific heat (LTSH) predicted by theory[7] for ferromagnetic spin fluctuations. In the rest of these materials it is still a matter of discussion whether a spin-fluctuation description is appropriate.

UAl_2 is a compound with close (3.36 Å) U-U distance, which is thought to form an f band.[8] Increasing the U-U separation, either by dilution or by lattice expansion, should tend to localize the f electrons and thus narrow the f band causing an increase in the electronic density of states at the Fermi level, $N(0)$. We were therefore interested in how increasing the U-U distance in UAl_2 by alloying would affect its properties. With some approximations we calculate a spin-fluctuation temperature T_{sf} from the $T^3 \ln T$ term in the LTSH and from magnetic-susceptibility measurements.

In this investigation we have measured magnetic susceptibility, specific heat, and resistivity at low temperatures on an $U_{0.85}La_{0.15}Al_2$ alloy to study these effects. We have also measured magnetic susceptibility and resistivity of samples of UAl_2, $U_{0.93}La_{0.07}Al_2$, $U_{0.9}Y_{0.1}Al_2$, and $U_{0.93}Sc_{0.07}Al_2$ for comparison.

EXPERIMENTAL PROCEDURE

The samples for this investigation were prepared by arc melting the pure elements together on a water-cooled copper hearth in a zirconium-gettered argon atmosphere. The $U_{0.85}La_{0.15}Al_2$ sample was then annealed at 1450 ℃ in argon for 9 h. A Debye-Scherrer x-ray analysis of this sample showed it to be single-phase C-15 structure with lattice parameter $a_0 = 7.8211$ Å vs $a_0 = 7.766$ Å for pure UAl_2. The $U_{0.93}La_{0.07}Al_2$ sample was annealed for 8 d at 1100 ℃ in argon.

The resistivity measurements were made on a spark-cut bar of each of these samples with leads attached using silver epoxy. The measurements were taken by a four-lead ac technique at 220 Hz. Temperature, accurate to ±0.1 K, was measured with Ge and Pt resistance thermometers.

The magnetic-susceptibility measurements were made in a vibrating sample magnetometer described elsewhere.[9] The temperature was measured with a carbon glass resistance thermometer located near the sample on the sample rod.

The LTSH was measured by the time-constant method in an apparatus described elsewhere.[10-13]

RESULTS

The magnetic-susceptibility measurements (Fig. 1) show that at low temperature the susceptibility of $U_{0.85}La_{0.15}Al_2$ is significantly larger than that of UAl_2. The value of the susceptibility extrapolated to 0 K is 14.6×10^{-6} emu/g G for UAl_2 as opposed to 32.8 $\times 10^{-6}$ for $U_{0.85}La_{0.15}Al_2$. The sample showed no ferromagnetic remnant to the lowest temperatures reached (1.4 K).

Figure 2 shows a plot of the specific-heat data for $U_{0.85}La_{0.15}Al_2$ and that of UAl_2 (Ref. 14) for comparison. It can be seen that the low-temperature upturn in C/T is much larger in $U_{0.85}La_{0.15}Al_2$ than it is in UAl_2 and this upturn starts at higher temperature. In a system with spin fluctuations the specific heat should be of the form[7]

$$C = \gamma_0(1 + \lambda_{e\text{-ph}} + \lambda_{\text{sf}})T + \left\{\beta - \delta\left[\ln\left(\frac{4ST_{\text{sf}}}{\pi(S-1)}\right) - B_0\right]\right\} + \delta T^3 \ln T + \alpha T^5 \ , \qquad (1)$$

where $\gamma_0 = \pi^2 k_B^2 N(0)/3$, $\lambda_{e\text{-ph}}$ and λ_{sf} are the electron-phonon and spin-fluctuation interaction parameters, respectively, $\beta = 12\pi^4 k_B / 5\Theta_D^3$ where Θ_D is the Debye temperature, δ is the coefficient of the spin-fluctuation term, S is the Stoner enhancement factor, B_0 is a numerical constant, and α is the coefficient of the next-order term of the lattice specific heat. The solid line in Fig. 2 is a least-squares fit of this functional form to the data for $U_{0.85}La_{0.15}Al_2$. The coefficients from this fit are tabulated in Table I. Also shown in Fig. 2 are some specific-heat data points taken in a magnetic field of 10 T. The 5% decrease in the LTSH at 2 K in 10 T is comparable to the decrease for UAl_2 in this field,[14] and consistent with the predictions of Hertel, Appel, and Fay.[15]

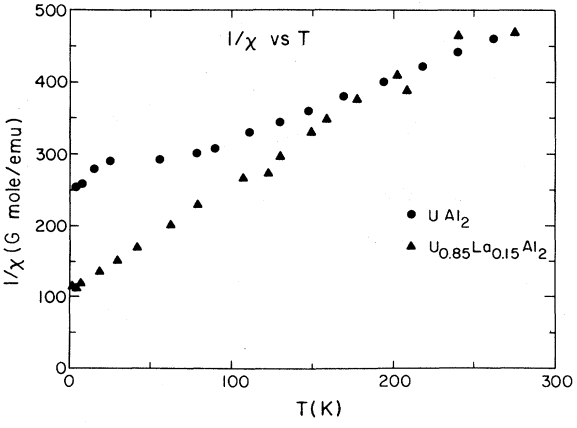

FIG. 1. Inverse susceptibility vs temperature for UAl_2 and $U_{0.85}La_{0.15}Al_2$.

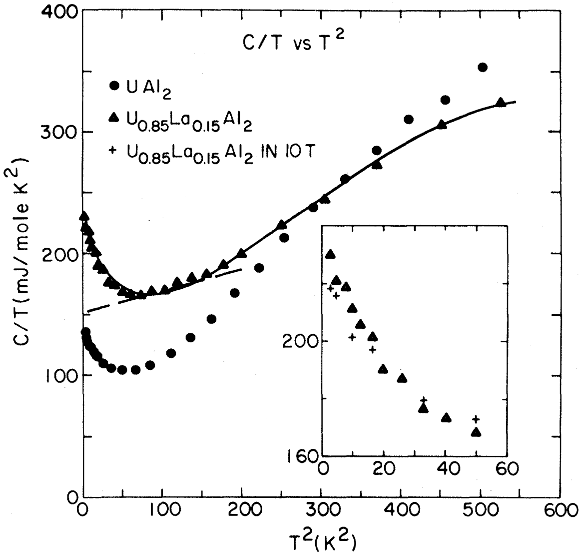

FIG. 2. Specific heat divided by temperature plotted vs temperature squared for UAl_2 from Stewart *et al.* (Ref. 14) and $U_{0.85}La_{0.15}Al_2$. The inset shows that low-temperature data for $U_{0.85}La_{0.15}Al_2$ in 0 and 10 T magnetic field. The solid line is a least-squares fit to the zero-field data for $U_{0.85}La_{0.15}Al_2$. The dashed line is explained in the text.

The resistivity data for $U_{0.85}La_{0.15}Al_2$ and arc melted UAl_2 are shown in Fig. 3. The $U_{0.85}La_{0.15}Al_2$ sample resistivity has the same general shape as that of UAl_2, but it has an unusually large residual resistivity, ρ_0, for this La concentration. We believe this is due to interference by the La atoms with the U–U bonding and thus with f-band formation. The data in Table II, which compares ρ_0 and χ of various samples, indicate that this increase in ρ_0 is not correlated with the increase in magnetism. The results for $U_{0.9}Y_{0.1}Al_2$ are in qualitative agreement with the work of Buschow and van Daal.[16] We also note that at

TABLE I. The results of a least-squares fit of the zero-field specific-heat data for $U_{0.85}La_{0.15}Al_2$ to the form $C = \gamma_0(1 + \lambda_{e\text{-ph}} + \lambda_{\text{sf}})T + BT^3 + \delta T^3 \ln T + \alpha T^5$, in mJ/mol K.

Parameter	Value
$\gamma_0(1 + \lambda_{e\text{-ph}} + \lambda_{\text{sf}})$	238.3
V	−5.21
δ	2.04
α	−0.001 93

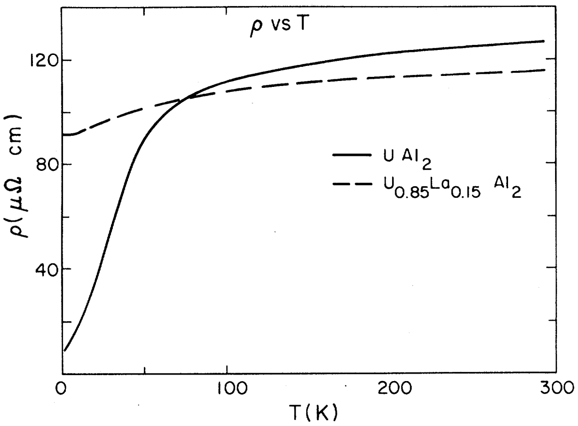

FIG. 3. Resistivity of UAl_2 and $U_{0.85}La_{0.15}Al_2$ vs temperature. Both samples were polycrystalline material.

TABLE II. The residual resistivity ρ_0 and magnetic susceptibility χ at 1.4 K are shown for some UAl_2-based alloys.

Sample	ρ_0 ($\mu\Omega$ cm)	χ (10^{-6} emu/g G)
$U_{0.93}La_{0.07}Al_2$	65	25.7
$U_{0.9}Y_{0.1}Al_2$	79	18.9
$U_{0.93}Sc_{0.07}Al_2$	68	14.4
UAl_2	7.6	14.6

high temperatures the resistivity is nearly saturated and thus has very little temperature dependence.

DISCUSSION

It is of interest to investigate how the increases in magnetic susceptibility and LTSH correspond to changes in T_{sf} and the Stoner enhancement factor S. To find a value for T_{sf} and S we follow the argument of Stewart *et al.*[14] We need to find S using the relation

$$S=\frac{\chi^m}{\mu_B^2 N(0)} ,$$

where χ^m is a measured magnetic susceptibility at 0 K and μ_B is the Bohr magneton. For a normal metal the LTSH is $C=\gamma_0(1+\lambda_{e\text{-ph}})T+\beta T^3$; therefore, in a plot of C/T vs T^2 this is a straight line with slope β and intercept $\gamma_0(1+\lambda_{e\text{-ph}})$. To find γ_0 and thus $N(0)$, we estimate Θ_D from the known value of Θ_D for $LaAl_2$(374 K),[17] using the relation

$$\Theta_D^{U_{0.85}La_{0.15}Al_2} \simeq \left(\frac{M^{LaAl_2}}{M^{U_{0.85}La_{0.15}Al_2}}\right)^{1/2} \Theta_D^{LaAl_2} .$$

We obtain $\Theta_D^{U_{0.85}La_{0.15}Al_2} \simeq 312$ K, or $\beta \simeq 0.192$ mJ/mol K^4. A dashed line with the slope β found above and tangent to the specific-heat data is shown in Fig. 2. This gives an intercept of $\gamma_0(1+\lambda_{e\text{-ph}}) = 154$. If we use an estimate of 0.3 for $\lambda_{e\text{-ph}}$, this gives $N(0)=16.8$ states/eV atom. Using our measured value of $\chi=32.8\times10^{-6}$ emu/g G we obtain $S=5.4$. This is considerably larger than the value for UAl_2 of $S=4.1$. Now we can calculate a value for T_{sf} from the relation[7]

$$T_{sf}=\frac{3\pi^2}{4}\left(\frac{\gamma_0 S}{5\delta}\right)^{1/2}\left(1-\frac{1}{S}\right)^2 ,$$

where δ is the coefficient of the $T^3 \ln T$ term in Eq. (1). Using the value of δ from Table I, we get $T_{sf}=39$ K which is 48% higher than the value for UAl_2 calculated in the same way.

We note that an increase in both the magnetic susceptibility and spin-fluctuation temperature seems to be inconsistent with theoretical predictions,[18] but even using a different, previously used method[2] for calculating T_{sf} gives an increase in T_{sf}.

We are engaged in a continuing effort to study the very interesting effects caused by ternary additions to the nearly magnetic UAl_2 system. We hope that our future investigations will lead to a better understanding of the magnetic properties and their relation to the chemical bonding involved in these alloys.

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