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MAGNETIC BEHAVIOUR IN THE $U_{1-x}Th_xCu_2Si_2$ SYSTEM

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SUMMARY

The compound UCu_2Si_2 has been found to be ferromagnetic at low temperatures with a Curie temperature of 101 K. The magnetization follows a Curie-Weiss law in the paramagnetic region with an effective magneton number of 2.12 Bohr magnetons per uranium atom. Partial substitution of Th atoms for the U atoms results in an expansion of the lattice, a rapid drop in the Curie temperature, and a sizeable increase in the coercive field.

INTRODUCTION

A very large number of ternary compounds crystallize in the body-centered-tetragonal $ThCr_2Si_2$ structure (see Fig. 1) and have been the subject of numerous investigations. These compounds have the general formula RM_2B_2 in which R can be a rare earth element, an actinide, or Y, Ca, and Sr while M is Cr, Fe, Co, Ni, Cu, Ag, Au, Pd, or Rh and B is either Si or Ge. A wide range of interesting phenomena have been found to occur in these materials including superconductivity, ferromagnetism, antiferromagnetism, and valence fluctuations.

The discovery of bulk superconductivity and "heavy fermion" behaviour in $CeCu_2Si_2$ by Steglick et al. in 1979 (ref. 1) has focused recent interest in the low temperature porperties of the rare earth and actinide members of this crystal group. Grier et al. (ref. 2) investigated the magnetic ordering in the CeM_2Si_2 compounds where M= Ag, Au, Pd, or Rh and Novion et al. (ref. 3) examined the magnetic structure of $NpCu_2Si_2$ and $NpCo_2Si_2$. The present study was undertaken to investigate the low temperature magnetic properties of UCu_2Si_2 and to determine the effect of partial substitution of Th atoms for the U atoms.

SAMPLE PREPARATION AND CHARACTERIZATION

The polycrystalline compounds UCu_2Si_2 and $ThCu_2Si_2$ were first prepared by arc melting stochiometric amounts of crystal bar Th, ^{238}U, Cu(99.9999% purity), and Si(99.99999% purity) together on a water-cooled copper hearth in a purified argon atmosphere. The meltings were repeated four times with the button turned over between meltings to insure homogeneity. The intermediate compositions having the general formula $U_{1-x}Th_xCu_2Si_2$ with x= .017, .07, .15, .25, .50, and .75 were prepared by repeated arc melting of proper weights of the U and Th compounds to form the desired compositions. The weight loss for all prepara-

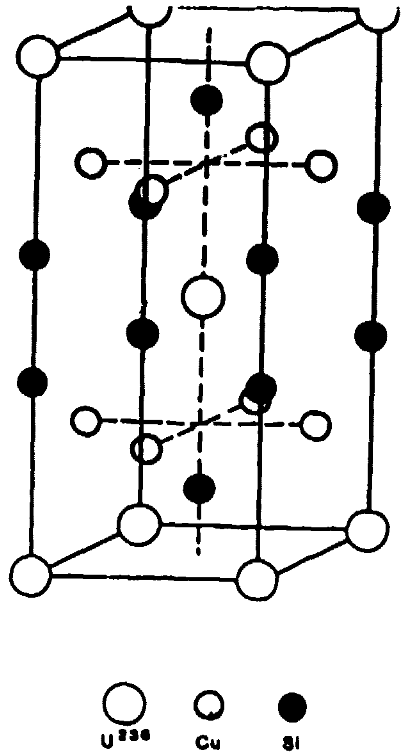

Fig. 1. Crystal structure of UCu_2Si_2. The space group is I4/mmm.

tions was less than 0.2 percent. The various samples were wrapped in Ta foil, sealed in quartz tubes under a He atmosphere and annealed at 800° C for 120 h.

Room temperature x-ray diffraction patterns of the finely powdered materials were taken with a General Electric XRD-6 diffractometer using Ni-filtered Cu-radiation. The various lattice parameters were determined and are listed in Table 1.

The magnetic properties were measured in a P.A.R. Model 150A vibrating sample magnetometer which is interfaced to a HP 86 computer. The power leads to the superconducting solenoid were modified so that the field direction could be reversed allowing M versus H measurements in all four quadrants.

RESULTS AND DISCUSSION

The $U_{1-x}Th_xCu_2Si_2$ samples in which x= 0.0, 0.017, 0.07, 0.15, and 1.00 gave x-ray diffraction patterns which were fully indexed suggesting these compositions are single phase. The samples with x= 0.25, 0.50, and 0.75 contain small amounts (estimated between 5 and 10 percent) of an unidentified second phase. Also the diffraction lines for these compositions were quite broad and diffuse so that the doublets in the back angle region were not resolved indicating considerable strain in the lattice results as the Th concentration is increased.

Measurements of the magnetic moment of UCu_2Si_2 sample were made at 3.97 K as a function of the applied field H. The complete hysteresis curve is shown

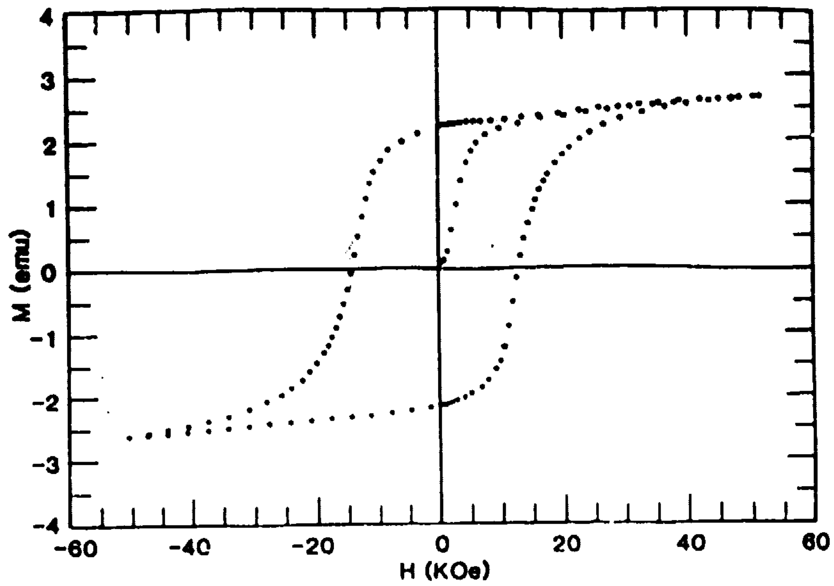

Fig. 2. Hysteresis loop for UCu_2Si_2 at 3.97 K.

in Fig. 2. The initial magnetization curve is seen to rise rapidly toward the saturation magnetization value of 4900 emu/U-mole. Since Th doesnot contribute to the magnetic moment, the magnetization will be defined as emu/U-mole. Isothermal curves of M versus H were measured over the temperature range of 50 to 200 K and the Curie temperature determined as the isothermal curve that intersects the origin. At temperatures above 100 K a plot of $1/\chi$ versus T is linear and yields a Curie-Weiss moment of 2.12 Bohr magnetons per U atom and an intercept with the T axis at 90.9 K.

Low-temperature magnetization measurements on the samples containing Th showed rather drastic effects on the magnetic properties as the Th content was increased. The replacement of only 1.7 percent of the U atoms by Th resulted in a sizeable decrease in the saturation magnetization, a lowering of the Curie temperature and an almost two fold increase in the coercivity. As the Th content is increased further, the magnetization curve shows no sign of saturation for applied fields up to 70 KOe and the coercive force increases sharply. The Curie temperature decreases almost linearly with increasing Th content. These data are listed in Table 1.

Clearly the UCu_2Si_2 is a ferromagnet with considerable anisotropy as evidenced by the large hysteresis. The introduction of Th atoms into the lattice acts as a lattice defect and causes microstresses which impedes both domain wall motion and domain rotation. This results in increased resistance to magnetization and in turn produces very high values of coercivity. It thus appears that small additions of Th tend to improve the hard ferromagnetic proper-

TABLE 1

Lattice parameters and magnetic data of the $U_{1-x}Th_xCu_2Si_2$ samples.

Th content at%	Lattice parameters a_o	c_o (Å)	T_c(K)	Saturation magnetization (emu/U-mole)	Residual induction (emu/U-mole)	Coercivity H_c(KOe)
0	3.978	9.933	101	4900	3890	14
1.7	3.982	9.931	98	2160	1750	26[b]
7	3.988	9.927	92	a	a	44[b]
15	3.995	9.920	85	a	a	54
25	4.016	9.914	78	a	a	61
50	4.048	9.900	58	a	a	--
75	4.075	9.885	32	a	a	--
100	4.104	9.864	--	--	--	--

[a] Magnetic saturation not achieved with maximum applied field of 70 KOe.
[b] Coercive force required to reduce the remanence from the 70 KOe applied field to zero.

ties of the UCu_2Si_2 but unfortunately it does so at the expense of a decrease in the saturation magnetization value and a lower Curie temperature.

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