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Magnetic properties of doped UCoSn

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Abstract

The magnetic properties of $UCo_{0.9}T_{0.1}Sn$ quasiternary solid solutions (T = Fe, Ni, Ru and Pd) have been studied. The magnetic moment increases with Fe and Ru substitutions and decreases in the case of Ni and Pd substitutions, the Curie temperature changes in similar way. Such a behaviour is unexpected considering the properties of ternary 1:1:1 uranium intermetallics.

Keywords: Uranium intermetallics; Magnetic properties

1. Introduction

The UCoSn compound belongs to uranium ternary equiatomic intermetallics UTX (X is a metal of the 3rd or 4th group of the periodic table and T is a late transition metal). It has a crystal structure of the ZrNiAl type (the ternary variant of the Fe_2P structure, the $P6m2$ space group). The compound is a ferromagnet with the highest ordering temperature ($T_C = 83$ K) of the whole UTX family [1,2]. The value of the uranium magnetic moment, $\mu_U = 1.2\ \mu_B$, is not the maximum (for UTX compounds with ZrNiAl structure this is 1.5 μ_B in antiferromagnetic UPdIn [3]), but is also rather large. The hybridization between 5f and 3d, 4d, 5d electrons is considered to be a main mechanism for the 5f-electron delocalization. This hybridization increases with decreasing number of d electrons in the 3d, 4d, 5d-band, which results in a nonmagnetic state of U in most of UTX with T = Fe and Ru, whereas compounds with T = Ni, Pd, Pt exhibit magnetic ordering with pronounced features of 5f-electron localization [4,5]. Consequently, one can expect a reduction of the magnetism in quasiternaries when Co is replaced by Fe and Ru and an enhancement in the case of Ni and Pd substitution. However, an unexpected increase of both T_C and μ_U was observed in $UCo_{1-x}Fe_xSn$ solid solutions with increasing Fe content [6].

In order to check whether the results of Ref. [6] reflect a systematic trend, we have studied the changes in magnetic properties of UCoSn upon replacement of 10% of Co by different late transition metals.

2. Experimental details

The $UCo_{0.9}T_{0.1}Sn$ (T = Fe, Ni, Ru, Pd, Cu and Ag) alloys were prepared by melting the components (uranium of 99.8%, other metals of 99.99%) in an arc furnace on a water-cooled copper bottom under a protective argon atmosphere. The ingots (of 3 g mass) were turned several times in order to avoid inhomogeneities, and afterwards they were wrapped in a Ta foil and annealed in vacuum at 750 °C in a sealed quartz tube for 1 week. The phase composition and lattice parameters were determined by a standard X-ray diffractometry.

Magnetization measurements were performed by an induction method in pulsed fields up to 40 T with a rise time of about 5 ms at 4.2 K on powder samples. The powders with randomly oriented particles were fixed by frozen alcohol to avoid an alignment of the particles in high field. This corresponds to an ideal polycrystal.

The Curie temperatures were determined from a.c. susceptibility measurements in magnetic field of 1 mT amplitude and 80 Hz frequency.

3. Results and discussion

Since of the chosen T metals only Ru forms a UTSn compound with ZrNiAl structure, a limited solubility of the other T metals was expected in the $UCo_{1-x}T_xSn$ system. We anticipated that alloys with $x = 0.1$ are of

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single phase, because the homogeneity range was found to be up to $x = 0.15$ in the case of T = Fe [6]. Indeed, the $UCo_{0.9}T_{0.1}Sn$ alloys with T = Fe, Ni, Ru and Pd were of single phase and their lattice parameters are listed in Table 1. However, in the case of Cu and Ag a considerable amount of extraneous phases was observed and these alloys were not further studied.

One can see from Table 1 that the Fe and Ni substitutions do not strongly influence lattice parameters. The deviations do not exceed the literature spread of these parameters in UCoSn, which reflects a homogeneity range of the compound. In the case of Ru, a slight lattice expansion within the basal plane is compensated by a shrinkage along the c axis, so that the unit-cell volume remains unchanged. With further increasing Ru content, the $c(x)$ dependence passes through a broad minimum in vicinity of $x = 0.6$, which is explained by a preference in the T sites occupation of Ru atoms upon the Co–Ru substitution [7]. Such a preference, where the T atoms of smaller size occupy mostly positions in the U-containing planes and where the larger T atoms come in the U-free planes, is found in several other quasiternary $U(T^1,T^2)X$ systems [8]. Pd expands the lattice in all directions, but due to its low content the expansion does not reach 1%. Thus, the difference in lattice parameters for all compounds studied is rather small and, as shown below, cannot be responsible for changes in magnetic properties.

Fig. 1 shows a field-down part of high-field magnetization curves for compounds studied at 4.2 K. Values of magnetic moment, obtained from these curves, are also listed in Table 1. $\mu(0)$ is the molecular magnetic moment extrapolated to zero field after application of 40 T. In the present compounds, due to high magnetic hardness, $\mu(0)$ is practically equal to the remaining magnetic moment. $\mu(40)$ is the molecular magnetic moment in 40 T, the highest available field. μ_s is the spontaneous magnetic moment. For an ideal polycrystal with uniaxial anisotropy, $\mu_s = 2\mu(0)$, but it is usually difficult to determine $\mu(0)$ properly due to an overlap of field intervals associated with domain-wall movement and with moment rotation. A proper determination of $\mu(0)$ becomes the more easy, the larger the anisotropy. As shown by a comparison of high-field magnetization curves of free and fixed powders in long-pulse fields for most of the ZrNiAl-type UTX compounds (including UCoSn), these intermetallics are very suitable subjects for such determination of actual values of spontaneous moment in polycrystals [9]. Therefore, we consider $2\mu(0)$ as representative for μ_s.

Table 1
Structural and magnetic properties of $UCo_{0.9}T_{0.1}Sn$ compounds

T	a (pm)	c (pm)	T_C (K)	$\mu(0)$ (μ_B)	$\mu(40)$ (μ_B)	μ_S (μ_B)	dμ/dB (10^{-3} μ_B/T)	B_C (T)
Co	716.0	400.0	83	0.60	0.86	1.21	5.9	1.0
Fe	716.0	400.1	93.5	0.62	0.88	1.25	6.1	1.1
Ru	718.1	396.5	83	0.66	0.95	1.31	6.4	1.0
Ni	715.3	399.7	79	0.54	0.75	1.08	4.8	1.3
Pd	720.7	403.7	75	0.57	0.86	1.14	6.5	2.8

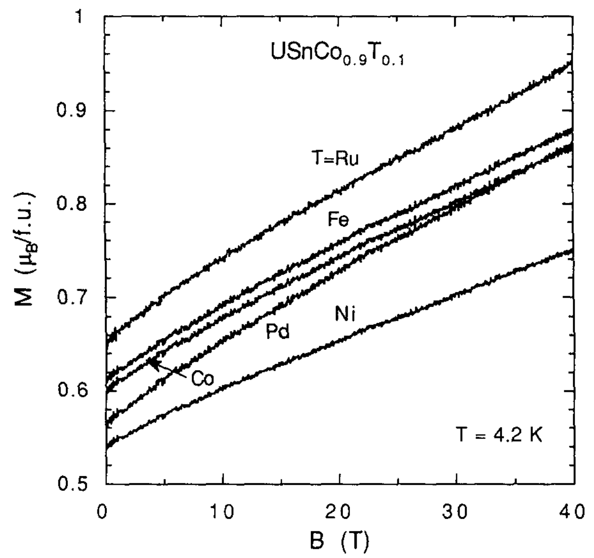

Fig. 1. High-field magnetization curves at 4.2 K (field-down branch).

One can see that a small but significant increase of μ_s exists in the case of T = Fe and Ru, whereas Ni and Pd reduce μ_s compared to that of the parent compound. A similar behaviour is found for T_C with the only difference that it does not change in the case of Ru. Such a development is rather unexpected on the basis of analysis of pure UTX ternaries. The similarity in behaviour of the compounds doped by Fe and Ru on one side, and by Ni and Pd on the other side, shows that the magnetic property variations are predominantly due to changes of the d-band filling. The size effect mentioned above is relatively unimportant.

Fig. 2 shows the virgin magnetization curves and hysteresis loops after application of a 7 T field. All the compounds exhibit low initial susceptibility, rapid rise of magnetization above a critical field and a rectangular hysteresis loop. These features of the magnetization process correspond to the narrow domain wall model of coercivity. This model may be applied to a ferromagnet in which the anisotropy energy exceeds the exchange energy. The ZrNiAl-type UTX compounds with their huge uniaxial anisotropy and relatively low μ_U and T_C evidently belong to this class of ferromagnets [5]. As for the parent compound [1], all the doped samples exhibit a very large magnetic anisotropy. Their magnetization curves do not tend to saturation even in 40 T (Fig. 1).

Due to the extremely high anisotropy field in the uranium magnets (of the order of hundreds of Tesla), it is very difficult to obtain quantitative data on the magnetic anisotropy energy even for single crystals and,

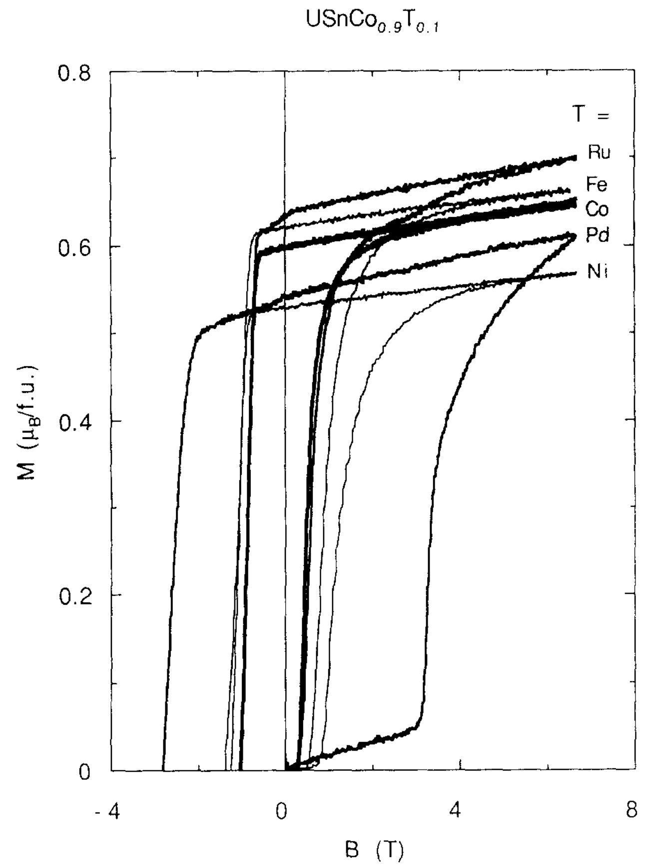

Fig. 2. Virgin magnetization curves and hysteresis loops at 4.2 K (with maximum applied field of 7 T).

especially, for polycrystals. In some cases, a qualitative behaviour of the magnetic anisotropy can be estimated indirectly from the high-field susceptibility and from the coercivity. However, in the compounds studied these results are not consistent. Table 1 presents dμ/dB at 40 T and the coercive field B_c at 4.2 K. An increase in dμ/dB in the Fe and Ru containing compounds, which could be considered as evidence for decreasing anisotropy, is not accompanied by a decrease in coercivity. In the case of Ni both quantities show an increase of the anisotropy. On the other hand, a considerable rise of B_c in the compound with Pd coexists with a moderate value of dμ/dB. Since both B_c and dμ/dB depend not exclusively on the anisotropy, this is not too surprising. An increase of the anisotropy seems, however, to be more realistic because of rather strong enhancement of coercivity.

Thus, we can conclude that the anisotropy at least does not increase with Fe and Ru doping and probably increases with Ni and Pd doping. Such a conclusion is in agreement with general trends in UTX compounds mentioned in the introduction. The increase of T_C with increasing hybridization was already observed and satisfactorily explained in the $URh_{1-x}Ru_xAl$ compounds with low Ru content [9]. However, magnetic moment decreases monotonically with x and leads to the disappearance of the magnetic ordering at $x = 0.5$. Therefore, the behaviour of magnetic moment observed in the present work is rather unusual and might be a subject of further study.

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