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Magnetic properties of $UCo_{0.9}T_{0.1}Al$ (T = Fe, Ni, Ru, Pd)

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Abstract

Magnetic properties of the $UCo_{0.9}T_{0.1}Al$ quasiternary solid solutions (T = Fe, Ni, Ru and Pd) have been studied. The base compound UCoAl is a spin-fluctuating system with a low-field metamagnetic transition. Whereas introduction of Fe and Ru leads to the appearance of strong ferromagnetism (T_C of 44 K and μ_U of 0.5 μ_B), in the case of Ni and Pd substitution a suppression of metamagnetism is observed. The results are compared with other UTX quasiternaries.

Keywords: Uranium intermetallics; Magnetic properties

1. Introduction

Magnetic as well as other electronic properties of the 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) were studied in great detail in the last decade ([1,2] and references therein). All the compounds display a huge magnetic anisotropy and relatively weak exchange interactions. The U magnetic moment varies from zero to 1.6 μ_B, depending on the degree of itinerancy of the 5f electrons of uranium. Other components do not contribute much to the magnetic moment, but affect the magnetic properties of the compounds by influencing the state of 5f electrons.

The hybridization between 5f and 3,4,5d electrons is considered to be a main mechanism for the 5f-electrons delocalization. This hybridization increases with decreasing number of d electrons in the 3,4,5 d-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 a relatively high U moment μ_U and exhibit pronounced features of 5f-electron localization [1–4]. Consequently, in the UTX compounds with T = Co and Rh one can expect a suppression of magnetism when Co (or Rh) is replaced by Fe (or Ru) and an enhancement in the case of Ni (or Pd) substitution. An increase in the Curie temperature T_C with increasing hybridization observed in the $URh_{1-x}Ru_xAl$ system at low Ru content can be satisfactorily explained [5], but the magnetic moment decreases monotonically in this system when going from ferromagnetic URhAl to paramagnetic URuAl. When studying the compound with the highest T_C within the whole UTX series (UCoSn), we found an unexpected increase of the magnetic moment upon Ru [6] and Fe [7] substitutions for Co as well as a moment decrease upon Ni and Pd substitutions [8]. The observed changes do not exceed 15% of μ_U, but show a certain trend. In order to check this trend, we have studied the influence of such substitutions on the magnetic properties of another representative of the UTX family, namely paramagnetic UCoAl having a low-field metamagnetic transition. Since the transition field in metamagnetic materials is usually very sensitive to substitutions, we expected more pronounced effects in UCoAl than in the strong ferromagnet UCoSn. For our study, the alloys with a 10% substitution of Co by Fe, Ni, Ru and Pd were chosen, as has been done in the case of UCoSn in Ref. [8].

2. Experimental details

All experimental procedures were the same as in Ref. [8], where the $UCo_{0.9}T_{0.1}Sn$ alloys were studied. The $UCo_{0.9}T_{0.1}Al$ (T = Fe, Ni, Ru, Pd) alloys were prepared by arc melting of the components. The ingots were annealed in vacuum at 750 °C for one week. The magnetization was measured in pulsed fields up to 35

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T at 4.2 K on powder samples. The powders consisting of randomly oriented particles were fixed by frozen alcohol to avoid an alignment of the particles in high field. This corresponds to an ideal polycrystalline material. The Curie temperatures were determined by ac susceptibility measurements.

3. Results and discussion

UCoAl has the hexagonal crystal structure of the ZrNiAl type (the ternary variant of the Fe_2P structure, the P$\bar{6}$m2 space group) [9,10]. All alloys studied were found to be single phase, including the sample with Pd. We note that Pd does not form a ternary compound UPdAl with ZrNiAl structure. The lattice parameters are listed in Table 1. For the parent compound, both the *a* and the *c* parameters are in good agreement with literature data [3,10,12–14]. As in the $UCo_{0.9}T_{0.1}Sn$ system, the Fe and Ni substitutions do not influence lattice parameters strongly because of the almost equal size of the Co, Fe and Ni atoms (the metallic radii are 125, 126 and 125 ppm, respectively). The considerably larger Ru and Pd atoms (134 and 137 pm) expand the lattice, but due to their low concentration this expansion is rather moderate. We would like to underline here the similarity in the behavior of Fe and Ni, from one side, and Ru and Pd, from another side. This is in contrast to corresponding magnetic properties discussed below, and shows that the difference in lattice parameters for all compounds studied cannot be taken as responsible for changes in magnetic properties.

Regarding the magnetic properties, we would like to recall in what direction we move upon substitution. UAlFe is a paramagnet with an almost temperature-independent magnetic susceptibility [1,15,16]. URuAl is also a paramagnet, but has features of a spin-fluctuating system (a broad maximum in temperature dependence of susceptibility around 50 K and a magnetization curve with an s-shaped anomaly around 25 T) [17]. UNiAl is a uniaxial antiferromagnet with $T_N = 19$ K, $\mu_U = 0.8$ μ_B and exhibits a sharp metamagnetic transition in 11 T [16]. UPdAl, as already mentioned, does not form.

High-field magnetization curves (field-down branches) at 4.2 K are shown in Fig. 1. One can see considerable changes in magnetization behavior upon substitution. As shown by single-crystal measurements, UCoAl has very large susceptibility even in the easy-magnetization direction (along the *z* axis) up to 35 T due to its paramagnetic ground state and its instability of the magnetic moment [17]. In the compounds with Fe and Ru, the high-field susceptibility decreases strongly compared to UCoAl (see Table 1). The Fe and Ru compounds behave similarly to each other as well as to many UXT ferromagnets with ZrNiAl structure. The ratio between the magnetic moments in 35 T and in zero field is equal to 1.45 (Fe) and 1.57 (Ru) in these compounds. This is a typical value observed for fixed powders with a random orientation of the particles in UTX ferromagnets having stable magnetic

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

T	*a* pm	*c* pm	T_c K	$\mu(0)$ μ_B	$\mu(35)$ μ_B	μ_s μ_B	$d\mu/dB$ 10^{-3} μ_B/T	B_c T
Co	668.5	397.0	16 [a]	< 0.01	0.47	0.3 [b]	5.7	–
Fe	669.8	396.4	44	0.27	0.39	0.54	2.9	0.28
Ni	670.7	397.2	–	0	0.35	0	7.7	–
Ru	672.9	397.3	43	0.24	0.38	0.48	2.9	0.63
Pd	674.1	399.2	9	0.04	0.41	–	8.9	< 0.05

[a] Curie temperature of the field-induced ferromagnetic phase determined as the point where the metamagnetism vanishes. It is also equal to T_c of the ferromagnetic contribution.

[b] Moment value after metamagnetic transition obtained on a single crystal [11].

T = Co corresponds to the UCoAl compound without substitution; *a* and *c* are the lattice parameters; T_c is the Curie temperature; $\mu(0)$ is the magnetic moment extrapolated to zero field after application of 35 T field; $\mu(35)$ is the magnetic moment in 35 T; μ_s is the spontaneous magnetic moment; $d\mu/dB$ is the differential susceptibility at 35 T; B_c is the coercive field after application of a 7 T field.

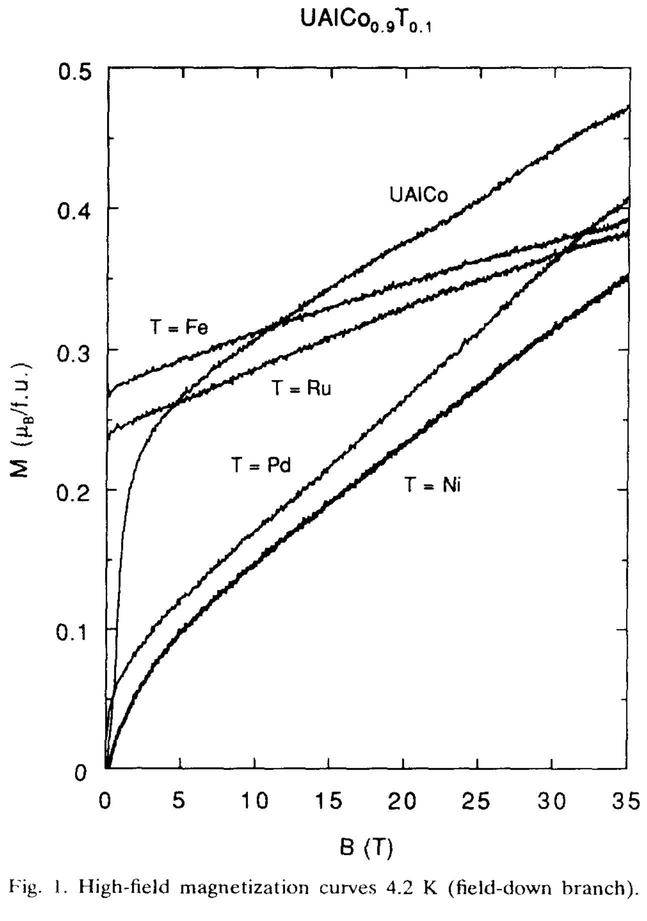

Fig. 1. High-field magnetization curves 4.2 K (field-down branch).

moments (for example, it is 1.4 in UCoSn and 1.6 in URuSn, [6]). For such compounds, this ratio is determined mainly by the magnetic anisotropy and shows that the anisotropy field may reach a typically huge value of the order of a hundred Tesla. On the other hand, the samples with Ni and Pd display completely different behavior. The low-field rise of the magnetic moment is almost suppressed whereas the high-field susceptibility increases considerably (Table 1).

For closer inspection of the low-field behavior, the virgin magnetization curves and hysteresis loops after application of a 7 T field are presented in Fig. 2. The parent compound exhibits an s-shaped magnetization curve with a broad metamagnetic transition in the vicinity of 0.8 T. The transition has a hysteresis with half-width of 0.03 T. In zero field, a small remanent moment of 0.01 μ_B reflects a ferromagnetic contribution. The results obtained are in agreement with single crystal data [11,14,17], only the transition in the random polycrystalline sample is not so well pronounced as along the *z* axis in the single crystalline sample.

The rather special magnetic properties of UCoAl were the subject of several studies and led to a disagreement in the conclusions regarding the ground state of this compound. First, an antiferromagnetic ordering (T_N = 16 K) with a metamagnetic transition, as in UNiAl, but with very low transition field, was proposed on the basis of magnetization, magnetostriction and ac susceptibility measurements of a single crystal [11]. Later it was shown that temperature dependence of the specific heat has no anomaly, indicative of magnetic ordering (as well as temperature dependence of resistivity) and a metamagnetic transition occurs from the paramagnetic ground state [17]. Subsequently a strong influence of deviations from the 1:1:1 stoichiometry on the magnetic behavior was observed, and a coexistence of ferromagnetic and metamagnetic states in some off-stoichiometric single-phase samples was found [14]. The Curie temperature of the ferromagnetic component and the temperature of disappearance of the metamagnetic behavior practically coincide (16 K). This can explain the anomaly in the ac susceptibility interpreted in Ref. [11] as due to magnetic ordering, because a very small ferromagnetic contribution (about 0.005 μ_B per formula unit) is found in the stoichiometric single crystal, which is enough to considerably change the temperature dependence of susceptibility compared to the pure metamagnet [18]. We should also mention, that the opinion exists about a ferromagnetic ground state of UCoAl [19].

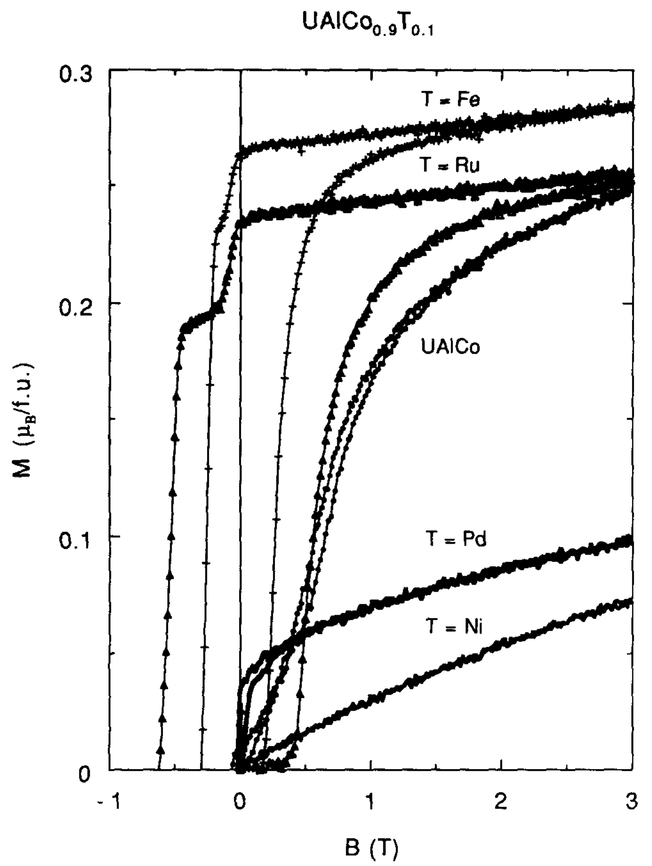

Fig. 2. Virgin magnetization curves and hysteresis loops at 4.2 K (with maximum applied field of 7 T).

The samples with Fe and Ru exhibit a low initial susceptibility, a rapid rise of magnetization above a critical field and an almost rectangle hysteresis loop. These features of the magnetization process are rather typical for the UTX ferromagnets, including UCo$_{0.9}$T$_{0.1}$Sn [8]. In Table 1, $\mu(0)$ is the molecular magnetic moment extrapolated to zero field after application of a 35 T field. As in other UTX ferromagnets, $\mu(0)$ is practically equal to the remanent magnetic moment. μ_s is the spontaneous magnetic moment, determined as $\mu_s = 2\mu(0)$. This relation is valid for an ideal polycrystalline material with uniaxial anisotropy. The ZrNiAl-type UTX compounds are suitable for such determinations of the true values of spontaneous moment in polycrystals, as was inferred from a comparison of the high-field magnetization curves of free and fixed powders in long-pulse fields [5]. In the presently studied compounds, μ_s is considerably lower than the maximum value found in UTX (1.6 μ_B), but there exist several compounds in this class with similar μ_s values (e.g. 0.6 μ_B in UCoGa [20]). The values of T_C of both Fe and Ru compounds are almost the same (Table 1). Ferromagnetism in U(Co,Fe)Al quasiternaries has already been observed [19]. The maximum T_C value (42 K) was found just for the UCo$_{0.9}$Fe$_{0.1}$Al composition, in good agreement with our result. The μ_s value reported for this compound (0.4 μ_B) is difficult to compare with our result because we do not know the method used for its determination [19].

The compound with Ni exhibits a simple paramagnetic magnetization curve (Fig. 2) without hysteresis, metamagnetic transition and spontaneous moment. The temperature dependence of ac susceptibility shows no anomaly indicative of magnetic ordering. Compounds of the $UCo_{1-x}Ni_xAl$ series with high Ni content ($0.6 \leqslant x \leqslant 1$) have already been studied in Ref. [21]. A rapid suppression of antiferromagnetism in UNiAl was observed. Compounds with 20 and more at.% Co are paramagnets, which was confirmed by the disappearance of an anomaly at T_N in the temperature dependence of the specific heat. Unexpectedly, Co influences the properties of UNiAl in a similar way to the influence of Ni on UCoAl. Pd substitution in UCoAl also leads to a strong reduction of the magnetization. The high-field magnetization curve of the Pd compound is very similar to that of the Ni compound. But the Pd sample exhibits a small ferromagnetic component with $T_C = 9$ K. The question is whether this is an intrinsic property of the compound or whether it is contributed by an impurity. The latter is more probable because of the rather low coercivity of this ferromagnet compared to the compounds with Fe and Ru (Table 1) and other UTX ferromagnets. Another feature that is not clear should be mentioned. This involves a step in low negative fields on the hysteresis loop of the Fe and Ru compounds. It might point to some degree of inhomogeneity of the samples.

Concluding, we found a similar development of the magnetic properties in $UCo_{0.9}T_{0.1}Al$ and $UCo_{0.9}T_{0.1}Sn$ compounds. In both cases, Fe and Ru substitutions lead to an enhancement of ferromagnetism, whereas the Ni and Pd substitutions suppress the magnetic moment. These results are in contradiction to general trends in magnetic properties known in uranium intermetallics. For their explanation, further investigations should be performed including specific heat measurements and studies of the site occupation of the T atoms because of the two nonequivalent d atom positions in the ZrNiAl structure.

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