Pressure influence on magnetism of UCo$_{1-x}$T$_x$Al solid solutions with T = 3d- and 5d- metals

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UCoAl belongs to a large UTX group of uranium intermetallics (T is a late transition metal of 3d, 4d or 5d series, X is a p-metal Al, Ga, In or Sn) with the hexagonal crystal structure of the ZrNiAl type. Compounds of this group exhibit very diverse magnetic properties from temperature-independent paramagnetism to ferromagnetism or antiferromagnetism [1,2]. UCoAl is a unique itinerant 5f electron metamagnet which exhibits a field-induced transition from the paramagnetic to the ferromagnetic state at a critical field $H_{cr} = 0.7$ T applied along the c-axis [2,3]. Due to so low H_{cr}, UCoAl is extremely sensitive to any influence, such as off-stoichiometry, dilution of the U sublattice, substitution within the Co or Al sublattices, external pressure, temperature and magnetic field. A magnetic moment at the transition is induced only on U [4], but the state of the 5f electrons depends very drastically on the interaction with ligands, especially with late d-metals.
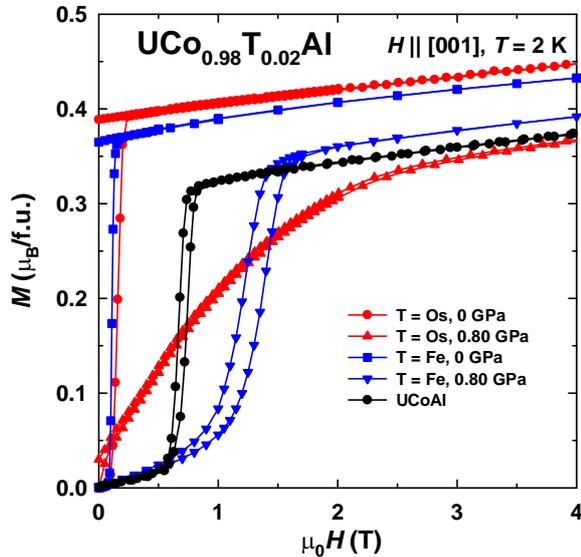
Fig. 1. Magnetization curves along the c axis of single crystals of UCo$_{0.98}$Fe$_{0.02}$Al and UCo$_{0.98}$Os$_{0.02}$Al compounds under ambient pressure and at 0.8 GPa at 2 K in comparison with ambient-pressure curve of UCoAl.

When Co is substituted by T = Fe, Ru, Os, Rh and Ir in the UCo$_{1-x}$T$_x$Al systems, H_{cr} rapidly decreases and already less than 1 % doping stabilizes spontaneous ferromagnetism [5-8]. Opposite, for T = Ni, Pd and Pt, H_{cr} increases and compounds become paramagnetic [5,9]. UCoAl and solid solutions on its base exhibit a huge uniaxial magnetic anisotropy.

In the present work we compare influence of hydrostatic pressure on magnetic properties of UCoAl-based compound with the same content of either 3d or 5d metals. Magnetization was measured in the temperature range 2-60 K using MPMS-7 cryomagnetic installation in fields up to 7 T applied along the c axis on single crystals. A miniature CuBe clamp pressure cell was used to apply hydrostatic pressure up to 0.83 GPa. The crystals were grown by the modified Czochralski method from 8 g stoichiometric mixtures of the pure elements (99.9% U, 99.99% Co and other d-metals, and 99.999% Al) in a tri-arc furnace with a water-cooled copper crucible.

Figure 1 shows magnetization curves of compounds with 2% Fe or Os in the Co sublattice. At ambient pressure both compounds are ferromagnetic with very similar magnetization curves with slightly higher magnetic moment in the case of Os (the Curie temperature T_C is 17 K for the Fe compound and 26 K for the Os one). However, under pressure they behave in very different ways. In the case of Fe substitution, metamagnetic transition characteristic for UCoAl restores at 0.2 GPa and moves to higher field with increasing pressure. The magnetization curve at the transition is slightly wider than in UCoAl. The broadened metamagnetic magnetization curve of UCo$_{0.98}$Fe$_{0.02}$Al for 0.8 GPa can be described well in the assumption of Gaussian distribution of the critical field values H_{cr} with respect to the average critical field H_{cr}^0. The distribution is characterized by the probability

$$P(H_{cr}) = \frac{1}{w\sqrt{\pi/2}} \exp[-2\frac{(H_{cr}-H_r^0)^2}{w^2}]$$

with $H_{cr}^0 = 1.34$ T and the distribution width $w = 0.35$ T.

The compound with Os demonstrates under pressure a mixture of the spontaneous ferromagnet and paramagnet. Ferromagnetic part almost disappears at 0.8 GPa (Fig. 1). Paramagnetic part does not demonstrate clear magnetization jump like in UCoAl or UCo$_{0.98}$Fe$_{0.02}$Al. However, the magnetization curve might be described by a very wide distribution of H_{cr} over crystal volume. Best fit of the magnetization curve of UCo$_{0.98}$Fe$_{0.02}$Al at 0.8 GPa gives the value of the average critical field $H_{cr}^0 = 0.8$ T and the distribution width $w = 1.7$ T.

The difference in the distribution width of the metamagnetic transition between the Fe and Os substituted UCoAl can be due to difference in the atomic radii. The metallic radius of Os (135 pm) is considerably larger than that of Co (125 pm). Therefore, random

substitution of Os for Co leads to significant local distortions of the lattice, which produce local variations of the 5*f-d* hybridization and distribution of parameters of the metamagnetic transition. On contrary, the metallic radius of Fe (126 pm) is almost the same as that of Co, and variation of the critical field of metamagnetic transition is moderate.

In Figure 2 we compare pressure behavior of the compounds in which 5% of Co is substituted by Ni and its 5*d* analogue Pt. In $UCo_{0.95}Ni_{0.05}Al$ the metamagnetic transition is shifted at ambient pressure to 2.5 T compared to 0.7 T in UCoAl. Under external pressure the transition moves to higher field. Magnetization gain over transition decreases. Extrapolation gives 3 GPa as a critical pressure where the transition disappears and the compound becomes usual paramagnet. Magnetization curve of $UCo_{0.95}Pt_{0.05}Al$ at ambient pressure might be described by very wide distribution of H_{cr} over crystal volume, similar to that of $UCo_{0.98}Os_{0.02}Al$. It includes even zero value which means that the compound has small (0.03 μ_B) but non-negligible spontaneous moment (which was not observed on random polycrystal [9]). This ferromagnetic component has T_C = 7 K. The crystal becomes forced ferromagnet at approx. 1.4 T, so the average H_{cr} is close to 0.7 T as in UCoAl. At external pressure 0.33 GPa the ferromagnetic component (with M_s = 0.15 μ_B and T_C = 5 K) is still observed. It vanished at 0.58 GPa. The magnetization curve at 0.83 GPa does not exhibit any trace of field-induced transition, the compound behaves as usual paramagnet.
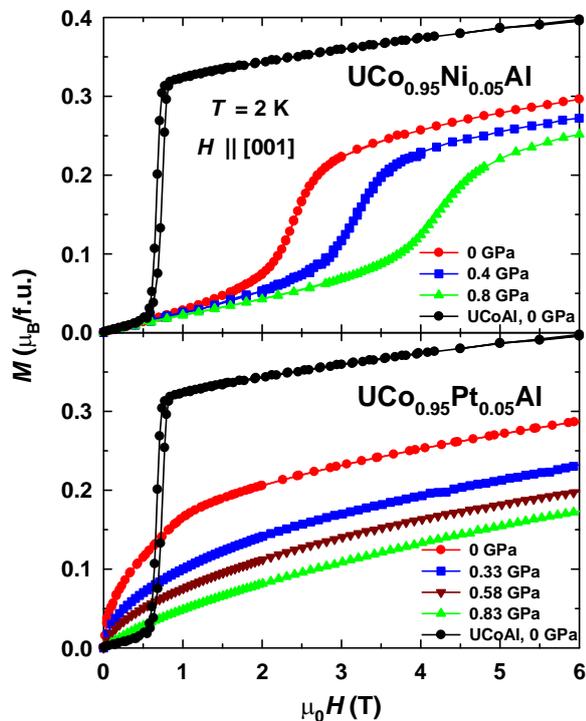
Fig. 2. Magnetization curves along the c axis of single crystals of $UCo_{0.95}Ni_{0.05}Al$ and $UCo_{0.95}Pt_{0.05}Al$ compounds under different pressure at 2 K in comparison with ambient-pressure curve of UCoAl.

We also compared pressure behavior of compounds with equal concentration of metals which enhance and suppress ferromagnetism, $UCo_{0.90}Fe_{0.05}Ni_{0.05}Al$ and $UCo_{0.90}Os_{0.05}Pt_{0.05}Al$. It was known that in the first case of 3d metals influence of Fe is stronger that of Ni, H_{cr} field decreases compared to that of UCoAl just to zero, $UCo_{0.90}Fe_{0.05}Ni_{0.05}Al$ at 2 K consists of equal amounts of ferromagnet (T_C = 16 K) and metamagnet. In the case of 5d metals, Os (analogue of Fe) also dominates over Pt (analogue of Ni). $UCo_{0.90}Os_{0.05}Pt_{0.05}Al$ is pure ferromagnet with M_s = 0.34 μ_B and T_C = 26 K).

Magnetic ordering of uranium moments in the UTX compounds is determined by the *f - d* and *f ó p* hybridization. A huge magnetic anisotropy of UTX is mainly due to the *f - d* hybridization within the basal plane of the lattice. It was empirically established that the interplane *f - d* hybridization favours ferromagnetic ordering, while the *f ó p* hybridization is responsible for antiferromagnetic interplane interactions. The difference in the magnetic behavior of UCoAl upon the Fe and Ni alloying is due to weakening of interplane *f - d* hybridization caused by filling the *d*-electron shell. When we consider the difference between the Os and Pt alloying, we have to take into account an additional effect of variation of interatomic distances. Since both Os and Pt have larger metallic radii than Co, the substitution leads to an appreciable increase in the lattice volume and especially in the *a* parameter, which plays the role of õnegativeö pressure, reduces the hybridization and increases ferromagnetic interactions.

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