ARTICLE IN PRESS

Journal of Magnetism and Magnetic Materials xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Research articles

Alloying and pressure effects on itinerant-electron metamagnetism of the UCoAl-based compounds

N.V. Mushnikov [a,*], A.V. Andreev [b], Z. Arnold [b], K. Shirasaki [c], T. Yamamura [c]

[a] Institute of Metal Physics UB RAS, S. Kovalevskaya 18, 620990 Ekaterinburg, Russia
[b] Institute of Physics, Academy of Sciences, Na Slovance 2, 182 21 Prague, Czech Republic
[c] Institute for Materials Research, Tohoku University, Katahira 2-1-1, 980-8577 Sendai, Japan

ARTICLE INFO

Article history:
Received 10 July 2017
Received in revised form 20 October 2017
Accepted 25 October 2017
Available online xxxx

Keywords:
Uranium intermetallics
UCoAl
Itinerant electron metamagnetism
Ferromagnetism
Pressure effects

ABSTRACT

With the $UCo_{1-x}Os_xAl$ single crystals, we studied interplay between alloying and pressure effects on magnetism of the itinerant electron metamagnet UCoAl. The Os alloying for $x \geq 0.005$ switches the UCoAl to ferromagnetism. For the $UCo_{0.995}Os_{0.005}Al$ single crystal, even lowest applied pressure 0.11 GPa is sufficient to suppress ferromagnetism. A sharp metamagnetic transition for various pressures is observed in magnetic fields along the *c* axis of the crystal. Concentration magnetic phase diagram of the $UCo_{1-x}Os_xAl$, which includes the quantum phase transition from ferromagnetic to metamagnetic state, and *P*–*T*–*H* phase diagram of the $UCo_{0.995}Os_{0.005}Al$ are determined using the data of magnetization measurements at various pressures and temperatures.

1. Introduction

The field of quantum criticality attracts widespread experimental and theoretical attention because of its importance in understanding the behavior of correlated electron systems [1–3]. In some itinerant-electron ferromagnets, the Curie temperature can be brought to zero by controlling external pressure or composition. In the critical region of the quantum phase transition, the itinerant-electron ferromagnets can exhibit exotic states such as unconventional superconductivity [4], skyrmion lattice ordering [5], nematic state [6]. Magnetic phase diagrams of various compounds near the quantum phase transition have several common features. The ferromagnetic transition temperature decreases with increasing pressure and the magnetic transition changes from second order to first order at a tricritical point (TCP). The ferromagnetic ground state disappears at a quantum transition point (QTP). At higher pressures, the Pauli-paramagnetic ground state is realized, and the first-order metamagnetic transition appears with application of magnetic field.

UCoAl is a well-known 5*f* itinerant electron metamagnet [7]. The ground state of the UCoAl is paramagnetic. In magnetic fields of approximately 0.7 T applied along the *c* axis, this compound undergoes the metamagnetic transition from paramagnetic to a ferromagnetic-like state with the U magnetic moment of 0.3 μ_B. Such first-order field-induced transition is considered to originate from a special shape of the density-of-states curve around the Fermi level [8]. A theory of the itinerant electron metamagnetism based on the spin-fluctuation model [9,10] is commonly used to describe the main features of magnetic behavior of UCoAl.

UCoAl belongs to a wide group of the compounds with general formula UTX (T is a late transition metal of 3*d*, 4*d* or 5*d* series, X is a *p*-metal Al, Ga, In or Sn) with the ZrNiAl-type structure (space group $P\bar{6}2m$). The structure is built up of U-T and T-X layers alternating along the *c*-axis [7]. In all the studied to date UTX compounds, only U atoms can carry magnetic moment. Magnetic properties of UTX compounds strongly depend on the overlap of the 5*f* wave functions of neighboring U atoms and the hybridization of the 5*f* states of U with the *s*, *p* and *d* valence states of T and X atoms [11]. The overlap leads to a huge magnetic anisotropy with the easy magnetization direction parallel to the hexagonal *c*-axis. The U-U interaction along the *c*-axis involves the *f-d* and *f-p* hybridization, which can provide either ferromagnetic (UCoGa, UCoSn, etc.) or antiferromagnetic (UNiAl, UNiGa, etc.) ordering of the U magnetic moments. A balance of ferromagnetic and antiferromagnetic interactions along the *c*-axis and strong in-plane 5*f*-3*d* hybridization prevents the formation of the spontaneous U magnetic moment in UCoAl.

The metamagnetic transition in UCoAl is sensitive to pressure [12]. Critical field of the transition H_{cr} increases with increasing

* Corresponding author.
E-mail address: mushnikov@imp.uran.ru (N.V. Mushnikov).

https://doi.org/10.1016/j.jmmm.2017.10.099
0304-8853/

hydrostatic pressure at a rate 2.6 T/GPa [13]. On the other hand, uniaxial pressure applied along the *c*-axis stabilizes ferromagnetic ground state [14,15]. Application of the uniaxial pressure makes the transition very broad, since the stresses are non-uniformly distributed over the sample. Therefore, the pressure-induced magnetic phase diagram of the UCoAl can be drawn only schematically [15,16].

Magnetic properties of the UCoAl are strongly affected by the alloying. Only 1–2% doping of the Co sublattice by Fe, Ru, Rh and Ir stabilizes ferromagnetic ground state [17–19]. For the ferromagnetic compositions that are close to the phase boundary between ferromagnetic and metamagnetic phases, application of hydrostatic pressure provides reentrant metamagnetism [20,21]. The interplay between alloying and pressure, however, is usually accompanied by broadening of the transition because of inhomogeneous distribution of the components even for single-crystalline samples.

Recently it was found that in $UCo_{1-x}Os_xAl$ solid solution small Os alloying is able to switch the UCoAl to the ferromagnetic state [22]. In the present work, we studied the alloying and pressure effects on magnetic properties of the $UCo_{1-x}Os_xAl$ single crystals, including the pressure-induced quantum phase transition from ferromagnetic to metamagnetic state. We found the metamagnetic transition to be very sharp for low Os contents. This makes it possible to correctly determine magnetic phase diagram of the itinerant 5*f* electron ferromagnet near the quantum phase transition.

2. Experimental

The $UCo_{1-x}Os_xAl$ single crystals were grown by a modified Czochralski method in a tri-arc furnace from the metals with 99.9% (U), 99.99% (Co and Os) and 99.999% (Al) purity on a rotating water cooled copper crucible under protective Ar atmosphere. Since the melting temperature of Os exceeds the boiling point of Al, we used a binary precursor $Co_{80}Os_{20}$ [22]. X-ray diffraction patterns measured on powdered sample prepared from the ingot were used to control the phase state of the alloys. The single crystal quality was checked by back-scattered X-ray Laue analysis.

Magnetization was measured in the temperature range 2–40 K in magnetic fields up to 7 T using MPMS-7 cryomagnetic installation (Quantum Design Inc). A miniature CuBe clamp pressure cell with a mixture of mineral oils as a pressure transmitting medium was used to apply hydrostatic pressure up to 0.82 GPa [23]. The pressure was determined at low temperatures using the known pressure dependence of the critical temperature of the superconducting state of the pure Pb (5N) sample. Magnetic fields were applied only along the *c*-axis of the sample, since in magnetic fields along the *c*-plane, the samples behave Pauli-paramagnetically and no metamagnetic transition is observed [24].

3. Results and discussion

Since UCoAl forms the ZrNiAl crystal structure and UOsAl crystallizes with the hexagonal $MgZn_2$ type Laves phase structure, the solid solutions in the $UCo_{1-x}Os_xAl$ system are formed in limited concentration ranges. For small Os contents, single-phase state with the ZrNiAl type structure is observed for $x \le 0.2$ [22]. Parent UCoAl is characterized by the lattice parameters $a = 0.668$ pm, $c = 0.3966$ pm. Within the solubility limit, the crystallographic *c* parameter remains virtually unchanged, while the *a* parameter increases with increasing *x* up to 0.674 pm for $x = 0.2$ [22]. The lattice deformation induced by the Os alloying is of the same type as that under uniaxial pressure, where the lattice expansion in the basal plane pushes the UCoAl metamagnet to the ferromagnetic state.

Fig. 1 shows magnetization curves of $UCo_{1-x}Os_xAl$ single crystals measured at $T = 2$ K in magnetic fields applied along the *c*-axis for different $x \le 0.1$. Parent compound UCoAl shows a very sharp metamagnetic transition at a critical field $\mu_0H_{cr} = 0.7$ T, in good agreement with previous studies [13,25]. The field of the metamagnetic transition decreases with increasing *x* (inset in Fig. 1) and ferromagnetic ground state is stabilized at $x \ge 0.005$. Besides, the Os alloying leads to a substantial increase in the magnetic moment in ferromagnetic state. Perhaps, due to decrease of the in-plane 5*f*-*d* hybridization, localization of the 5*f* electrons gradually increases.

$UCo_{0.98}Os_{0.02}$ at 2 K shows typical ferromagnetic behavior with a rectangular hysteresis loop that is characterized by the spontaneous magnetic moment of 0.4 μ_B/f.u and the coercivity 0.12 T. With increasing temperature, the transition to the paramagnetic state occurs at the Curie temperature $T_C = 26$ K which is the point of a second-order magnetic phase transition. Thus, 2% of Os substitution for Co shifts the magnetic state of UCoAl from metamagnetic to ferromagnetic region of the phase diagram across the quantum critical point.

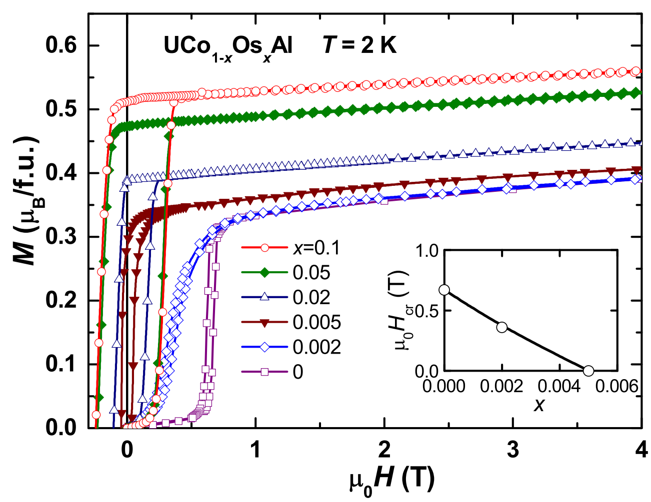

Fig. 1. Magnetization curves of $UCo_{1-x}Os_xAl$ for different *x* at $T = 2$ K. The inset shows concentration dependence of the critical field of metamagnetic transition.

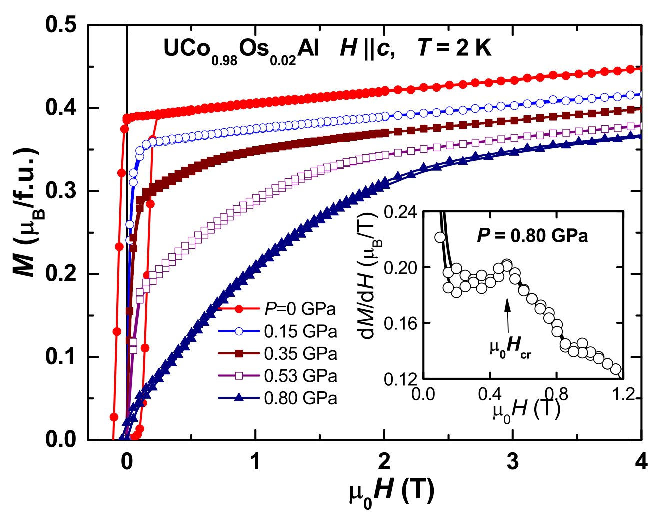

Fig. 2. Magnetization curves of $UCo_{0.98}Os_{0.02}Al$ for $T = 2$ K at various pressures. Inset: field derivative of the magnetization d*M*/d*H* as a function of external field for $P = 0.80$ GPa.