Evolution of magnetic structures in $U(Ni_{1-x}Pd_x)_2Si_2$ system

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Abstract

Influence of Pd-Ni substitution on the formation of magnetic phases in the tetragonal $U(Ni_{1-x}Pd_x)_2Si_2$ system and the corresponding magnetic phase diagrams are presented. The single crystals of two different substitutions $x = 0.15$ and 0.10 were grown and detailed studies by neutron diffraction were performed in horizontal magnetic fields up to $B = 6$ T. Both compounds order antiferromagnetically (AF) and exhibit three different AF structures below the Néel temperature. All three structures are formed by ferromagnetic basal planes stacked along c-axis with different $\mathbf{q} = (0, 0, q_z)$ propagation. For $x = 0.10$, the co-existence of two magnetic phases in the ground-state was observed.

Key words: magnetic phase diagram, antiferromagnetism, uranium intermetallic compounds

1. Introduction

Most of the ternary uranium silicides UT_2Si_2 with transition metal T crystallize in the centrosymmetric tetragonal structure of the $ThCr_2Si_2$-type with the space group *I4/mmm*, among them both UNi_2Si_2 and UPd_2Si_2, as well as the pseudoternary $U(Ni_{1-x}Pd_x)_2Si_2$ system ($0 \leq x \leq 1$). Owing to similarity of lattice parameters the Ni – Pd substitution is possible in the whole concentration range. The border compounds UNi_2Si_2 and UPd_2Si_2 attracted much attention by their huge magnetocrystalline anisotropy, complexity of magnetic phase diagrams and availability of high quality single crystals. Both border compounds order antiferromagnetically (AF) below the Néel temperatures $T_N = 125$ K and 133 K, respectively [1]. All magnetic structures are formed by ferromagnetic basal planes with U magnetic moments ($\mu_U \approx 2.2\mu_B$) parallel to the c-axis [1–4]. The individual magnetic phases depend on various stacking or modulation of these planes along the c-axis, described by various propagation vectors expressed generally as $\mathbf{q} = (0, 0, q_z)$.

In UNi_2Si_2 the ground-state phase AF3 is an uncompensated aniferromagnet (UAF) with the $++-$ stacking of the basal planes ($q_z = 2/3$). Between 40 and 110 K there is simple AFI-type structure AF2 with $+-+-$ stacking and $q_z = 1$. Finally, between 110 K and the T_N there is an incommensurate longitudinal spin-density wave (ILSDW) phase AF1 with $q_z \approx 0.74$. In UPd_2Si_2 the ground-state phase is just the AF2 structure, then at 108 K there is a transition to the ILSDW phase AF1 ($q_z \approx 0.73$) which is stable up to T_N.

In UNi_2Si_2 was found that the formation of the UAF phase is promoted in high magnetic fields parallel to the c-axis [4] or in elevated pressures [5]. This caused natural question about the concentration stability of the UAF phase in $U(Ni_{1-x}Pd_x)_2Si_2$. The aim of this work was to find the critical concentration of Pd, where the UAF ground state converts into the AF2. The pilot powder neutron diffraction study had shown that this concentration should be lower than $x = 0.25$ [6].

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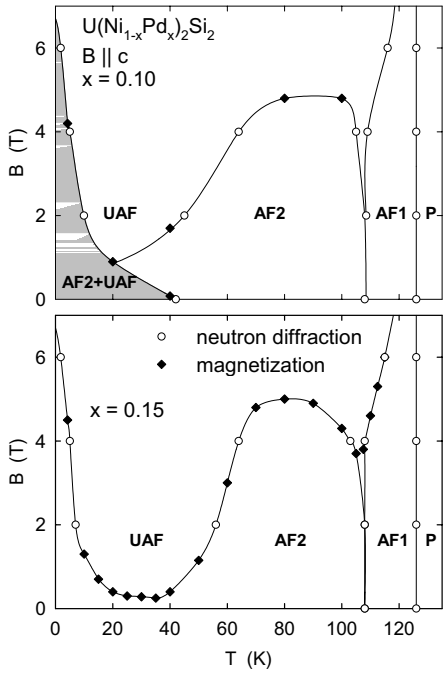

Fig. 1. Magnetic phase diagrams of $U(Ni_{1-x}Pd_x)_2Si_2$ for $x = 0.10$ and 0.15, respectively. The gray area on the $x = 0.10$ graph shows the region of the co-existence of the UAF and AF2 phases.

2. Experimental, Results and Discussion

Two single crystals for $x = 0.15$ and 0.10, respectively, were selected for the detailed microscopic study. For the description of the crystal growth and the results of the bulk studies please see [7]. The neutron diffraction experiment was performed in BENSC, HMI-Berlin using the E4 diffractometer with the incident wavelength $\lambda = 2.41$ Å. The samples were mounted within the $a - c$ scattering plane into the 6 T horizontal field cryomagnet HM1 with the field parallel to the crystallographic c-axis.

To exclude possible magnetic history effects the data were taken after zero field cooling from the paramagnetic range to the lowest available temperature ($\approx$ 1.6 K), then the magnetic field was applied and the measurements were performed while heating.

The neutron diffraction has confirmed the good quality of both crystals with low-temperature lattice parameters almost identical with those of UNi_2Si_2 ($a \approx$ 3.95 Å, $c \approx 9.52$ Å) within the experimental error of neutron diffraction experiment. (The detailed room-temperature X-ray analysis yields $a = 3.93125(9)$ and 3.96837(8) Å, $c = 9.46020(1)$ and 9.55750(3) Å for $x = 0.10$ and 0.15, respectively.) Only the three above described magnetic phases were found in the magnetically ordered state in fields up to 6 T and in all of them the uranium magnetic moment is parallel to the c-axis.

In the $x = 0.15$ crystal the ground state is the AF2-phase as in pure UPd_2Si_2 and the magnetic field of at least 0.25 T is necessary to produce the UAF structure. On the other hand, the co-existence of AF2 and UAF phases was found as a ground state of the $x = 0.10$ crystal. Here, with the increasing magnetic field the intensity of the AF2 reflections irreversibly drops down revealing the rapid diminishing of the AF2 volume fraction.

The temperature and field dependence of the q_z propagation in the ILSDW phase AF1 of both crystals is within the experimental error identical to that of pure UNi_2Si_2 [4], approaching the value $q_z = 2/3$ with increasing field and decreasing temperature.

All these data enabled us to construct magnetic phase diagrams for both samples (see Fig. 1) which at temperatures above $\approx$ 60 K strongly resemble that of pure UNi_2Si_2 [1,4]. This fact makes both crystals good candidates for the studies under high hydrostatic pressures, where the AF2 $\Rightarrow$ UAF change of the ground state is expected, similar to [5].

The influence of the Pd substitution on UNi_2Si_2 magnetism may be attributed to a 'negative chemical pressure' in conjunction with modification of the electronic structure (reduced $5f$ – ligand hybridization). In this scenario the application of the high pressure may re-establish the UAF ground state. Such experiments are now in progress.

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