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Evolution of magnetism in $U(Ni_{1-x}Pd_x)_2Si_2$ single crystals

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Abstract. Single crystals of $U(Ni_{1-x}Pd_x)_2Si_2$ with $x = 0.05$, 0.09 and 0.135 have been grown. Magnetization and electrical resistivity measurements were performed in a wide range of temperatures and magnetic fields in order to study stability of magnetic phases in the solid solutions between UNi_2Si_2 and UPd_2Si_2 with a special emphasis on the type of ground state. In UPd_2Si_2 the simple AFI-type antiferromagnetic structure of U moments is observed at low temperatures. UNi_2Si_2 adopts the uncompensated AF structure (UAF) with the $++-$ stacking of U moments along the c-axis and consequently this compound exhibits a spontaneous magnetization corresponding to 1/3 of the U moment. The substitution of Pd for Ni leads to a rapid decay of the spontaneous magnetization. The evolution of magnetization and electrical resistivity behavior with Pd doping is tentatively attributed to the coexistence of the AF-I and UAF phases in the ground state of $U(Ni_{0.91}Pd_{0.09})_2Si_2$ and $U(Ni_{0.865}Pd_{0.135})_2Si_2$. In this scenario, the volume fraction of the AF-I phase rapidly grows with Pd doping on account of the UAF. At lowest temperatures an irreversible transition to the UAF phase is observed when a sufficiently high magnetic field is applied along the c-axis.

PACS. 75.50 Ee Antiferromagnetic materials – 75.50 Gg Ferrimagnetics – 75.30 Gw Magnetic anisotropy – 75.30 Kz Magnetic phase transitions

1 Introduction

The UT_2Si_2 (T: transition metals) compounds crystallize in the body-centered $ThCr_2Si_2$-type of tetragonal structure characterized by the space group I4/mmm [1]. The crystal structure consists of the U basal planes, which are separated by the T-Si slabs. The UT_2Si_2 compounds with various transition metals exhibit variety of interesting physical properties such as the heavy-fermion state, superconductivity, $5f$-electron magnetism or coexistence of the two cooperative phenomena [2]. Closer inspection of experimental data collected for the UT_2Si_2 family reveals that the compounds with the transition metals from the same column of the Periodic table frequently exhibit common features. Both, the UPd_2Si_2 and UNi_2Si_2 compounds order magnetically at relatively high temperatures ($T_N = 133$ and 124 K, respectively) [3,4]. In a limited temperature range (down to $T_2 = 108$ and 103 K, respectively) both compounds exhibit an incommensurate magnetic structure of U magnetic moments, which is frequently quoted as the longitudinal spin density wave (LSDW). At T_2 the both compounds undergo the first-order magnetic phase transition to the body-centered antiferromagnetic AF-I structure characterized by the simple $+-+-+-$ coupling of the U magnetic moments along the c-axis. In UPd_2Si_2 this magnetic structure persists down to lowest temperatures and is characteristic for the ground state. In UNi_2Si_2 the AF-I phase is stable only in a limited temperature range and at $T_1 = 53$ K the AF-I phase transforms to the ground-state phase, which is characterized by a spontaneous magnetization.

Magnetism in UPd_2Si_2 and UNi_2Si_2, as well as in the other UT_2Si_2 counterparts is characterized by the strong uniaxial anisotropy with the easy-magnetization direction along the c-axis. On the microscopic level this anisotropy is associated with the ordered U magnetic moments being locked always along c in this $ThCr_2Si_2$-type structure. All the magnetic structures observed in the two compounds have in common basal-plane sheets of ferromagnetically coupled U moments whereas they differ in the propagation of the U moment along the c-axis; *i.e.* all the propagation vectors have the general form $\boldsymbol{q} = (0,0,q)$. The LSDW phase in UNi_2Si_2 is described by the temperature dependent value of $q \sim 0.74$ [4,5] whereas the value of 0.732 was determined for UPd_2Si_2 [3]. The AF-I phase is characterized by $q = 1$ whereas the ground-state phase in UNi_2Si_2, which exhibits the spontaneous moment equal

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to 1/3 of the U moment as a result of the $++-$ stacking, is quoted as the uncompensated antiferromagnetic phase (UAF) with $q = 2/3$ [4,5]. A detailed analysis of magnetization data revealed that besides the main AF-I phase the low-temperature state in UPd_2Si_2 contains about 1% of the UAF phase. The results of magnetization measurements at 1.3 K [6] indicate a metamagnetic transition into the UAF phase in a field of $\sim$ 15 T applied along the c-axis.

The magnetic phase transitions of UPd_2Si_2 and UNi_2Si_2 are accompanied by clear anomalies in the electrical resistivity [7,8], which makes the resistivity measurements a useful tool to study magnetism in these materials.

The stability of the AF-I and UAF phases with respect to the concentration of Ni and Pd is a subject of the present research. The pilot study performed on polycrystals of the $U(Ni_{1-x}Pd_x)_2Si_2$ solid solutions led to the conclusion that in compounds for $x \geq 0.25$ the ground-state phase is characterized by the entire AF-I phase [9]. In the present work, we have prepared several $U(Ni_{1-x}Pd_x)_2Si_2$ single crystals with various Pd content $x \leq 0.135$ and performed magnetization and electrical resistivity measurements. The main motivation was to investigate the evolution of the ground state for the Ni/Pd concentration region, for which the transformation between the AF-I and UAF phase is expected.

2 Experimental

The $U(Ni_{1-x}Pd_x)_2Si_2$ single crystals used for the present study were grown in a tetra-arc furnace by a modified Czochralski method from initial mixtures of components corresponding to $x = 0.05$, 0.10 and 0.15. Phase purity was confirmed by X-ray powder diffraction and by a microprobe analysis. The microprobe analysis confirmed the exact 1:2:2 stoichiometry for all the compounds studied (within a 1% error). As regards to the Pd-Ni sublattice, the crystals were found to correspond to average $x = 0.05$, 0.09 and 0.135, respectively. The composition varies over the crystals within $\Delta x = 0.01$ for $x = 0.05$ and 0.09 and $\Delta x = 0.015$ for $x = 0.135$. The lattice parameters determined as $a = 396.5$ pm, $c = 953.0$ pm for $x = 0.05$; $a = 397.6$ pm, $c = 956.7$ pm for $x = 0.09$; $a = 398.7$ pm, $c = 959.4$ pm for $x = 0.135$ are in satisfactory agreement with the expected linear increase of lattice parameters between UNi_2Si_2 and UPd_2Si_2 [2]. The crystal quality was checked and crystal orientation was facilitated by means of X-ray Laue method.

The magnetization curves were measured at various temperatures (4.2−130 K) using a vibrating sample magnetometer with a superconducting magnet in fields up to 12 T with continuous field sweeps at a rate $\mathrm{d}B/\mathrm{d}t = \pm\ 0.75$ T/min.

A SQUID magnetometer with magnetic fields up to 4 T was employed for measurements of the temperature dependence of the magnetization (magnetic susceptibility) in the temperature interval 5−300 K. The electrical resistivity for current along the c-axis was measured in the temperature range 3−300 K by a conventional four-probe AC method.

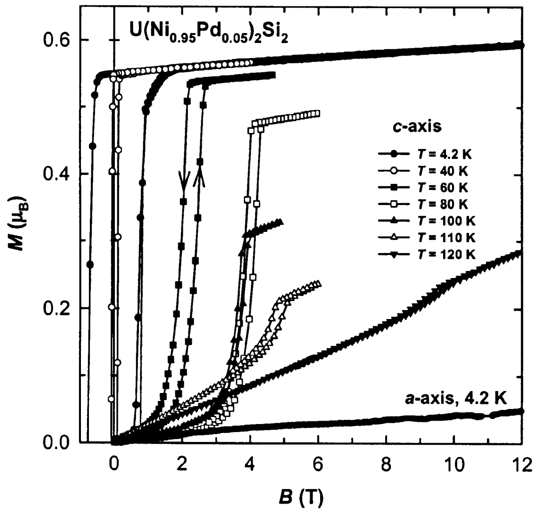

Fig. 1. The magnetization isotherms along the c-axis of the $U(Ni_{0.95}Pd_{0.05})_2Si_2$ crystal at 4.2 K and elevated temperatures. The magnetization curve along the a-axis at 4.2 K is also shown.

3 Results and discussion

3.1 $U(Ni_{0.95}Pd_{0.05})_2Si_2$

The magnetization curves measured on $U(Ni_{0.95}Pd_{0.05})_2Si_2$ in magnetic fields applied along the c-axis are shown in Figure 1 for several representative temperatures. At temperatures up to 40 K one observes a nearly rectangular hysteresis loop with an invariable remanent magnetic moment of 0.55 μ_B. The value of coercive force is gradually decreasing with increasing temperature (0.8 T at 4.2 K, 0.1 T at 40 K). This magnetization behavior is identical with that of UNi_2Si_2 [2,10], *i.e.* the UAF phase is characteristic for the ground state of both, the UNi_2Si_2 and $U(Ni_{0.95}Pd_{0.05})_2Si_2$.

The magnetization curves measured at 60, 80 and 100 K are compatible with the existence of the AF-I phase at these temperatures. Only these typical curves are shown for clear illustration although measurements at several other temperatures between 60 and 100 K were performed, as well. These $M(B)$ curves show no spontaneous magnetic moment but exhibit a sharp increase at a critical field B_c at which the AF-I structure transforms to the UAF phase. This metamagnetic transition exhibits a pronounced hysteresis. The value of B_c reaches maximum at temperatures around 80 K while the hysteresis becomes strongly reduced.

The magnetization curves measured at 110 and 120 K are considerably different from those observed at lower temperatures. This is because the zero-field state is no more characterized by the AF-I but it is the LSDW phase. For this temperature range, the low-field slope of the $M(B)$ curve is rather high and the metamagnetic transition becomes broadened.

In Figure 1 also the magnetization curve is shown as measured on $U(Ni_{0.95}Pd_{0.05})_2Si_2$ at 4.2 K in a magnetic field applied along the a-axis. This M *vs.* B dependence is

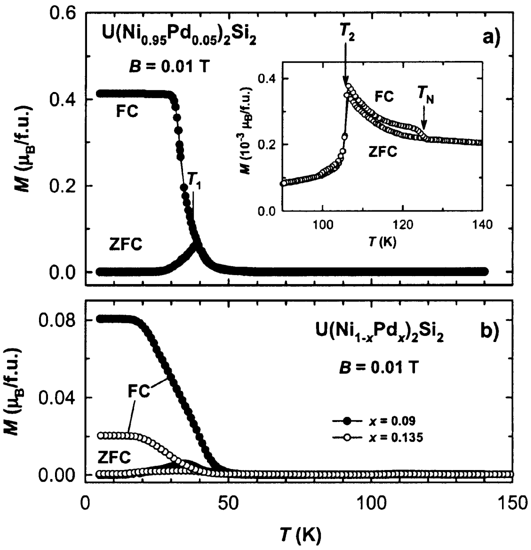

Fig. 2. a). Temperature dependence of magnetization along the c-axis of the $x = 0.05$ sample for ZFC and FC processes measured in a 0.01 T magnetic field. Expanded $M(T)$ curves around T_2 and T_N are in inset. b) Temperature dependence of magnetization along the c-axis of the sample with $x = 0.09$ and 0.135.

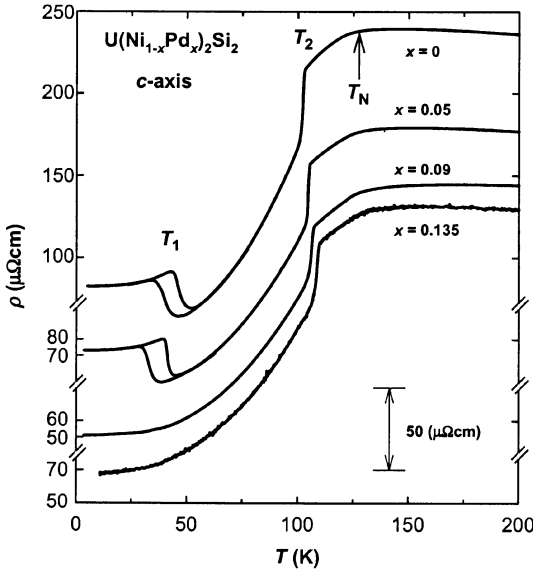

Fig. 3. Temperature dependence of electrical resistivity along the c-axis of the $U(Ni_{1-x}Pd_x)_2Si_2$ crystals with $x = 0.0 - 0.135$ for heating and cooling.

not far from being linear (except the initial part which is sensitive to a possible small misorientation of the crystal) with a very low susceptibility and is nearly identical with the basal-plane magnetization data reported for the parent compounds UNi_2Si_2 [2] and UPd_2Si_2 [6,10]. Moreover, we obtained almost the same a-axis magnetization data also for the other two crystals $U(Ni_{0.91}Pd_{0.09})_2Si_2$ and $U(Ni_{0.865}Pd_{0.135})_2Si_2$. This confirms that the huge uniaxial magnetocrystalline anisotropy of parent compounds persists also in the $U(Ni_{1-x}Pd_x)_2Si_2$ solid solutions. Since the essential features of magnetism are concentrated to the magnetization along the c-axis, the following magnetization data will be presented and discussed only for magnetic fields applied along this direction.

Figure 2a displays the variation of the magnetization of $U(Ni_{0.95}Pd_{0.05})_2Si_2$ in the field $B = 0.01$ T with increasing temperature. Note that the ZFC (FC) curve for all three compounds was measured on heating after cooling the sample in zero magnetic field (in the magnetic field of measurement). The ZFC data exhibit negligible magnetization ($M < 10^{-3}$ μ_B/f.u.) except for the temperature interval between 25 and 50 K, where it shows a peak reaching $M \simeq 0.05$ μ_B/f.u. at 38 K. The peak may be associated with the UAF $\rightarrow$ AF-I magnetic phase transition. Another sharp anomaly although much weaker ($M \simeq 4 \times 10^{-4}$ μ_B/f.u.) peaks at 105.5 K (see inset of Fig. 2a) and marks the AF-I $\rightarrow$ LSDW magnetic phase transitions. No anomaly, however, shows up in the ZFC curve at T_N.

The low temperature FC-magnetization data up to approximately 25 K show a high constant value of 0.41 μ_B/f.u. At temperatures increasing above 25 K the magnetization precipitously decreases and joins the ZFC curve at 38 K. The FC and ZFC curves for temperatures above 38 K are identical except for the temperature interval between T_2 and T_N, where the FC curve shows somewhat higher values, which is connected with a tiny step at T_N as seen in the inset of Figure 2a.

As can be seen in Figure 3, the three magnetic phase transitions revealed by the magnetization measurements on $U(Ni_{0.95}Pd_{0.05})_2Si_2$ are accompanied by corresponding anomalies on the temperature dependence of electrical resistivity. The transitions at T_1 (T_2) are associated with a rapid decrease (increase) of resistivity with increasing temperature. One can see that the $\rho(T)$ curves measured for $U(Ni_{0.95}Pd_{0.05})_2Si_2$ and UNi_2Si_2 are qualitatively very similar. The only differences are in the location of the two above mentioned $\rho(T)$ anomalies, which reflects a slight increase of T_2 and a stronger decrease of T_1 observed with the Pd doping in UNi_2Si_2.

The obtained magnetization and resistivity results provide a strong evidence for nearly identical magnetic phase diagrams for $U(Ni_{0.95}Pd_{0.05})_2Si_2$ and UNi_2Si_2, *i.e.* the two compounds condense in the same ground state characterized by the UAF magnetic structure although the temperature interval of the stability of the AF-I phase becomes somewhat expanded with Pd doping, especially on the low-temperature side.

3.2 $U(Ni_{0.91}Pd_{0.09})_2Si_2$

When the Pd content is increased, dramatic changes of magnetism can be recognized. The most striking change is seen in the temperature dependence of resistivity where one observes no anomaly below 50 K, which is accompanying the UAF $\Longleftrightarrow$ AF-I magnetic phase transition in $U(Ni_{0.95}Pd_{0.05})_2Si_2$ and UNi_2Si_2. On the other hand,

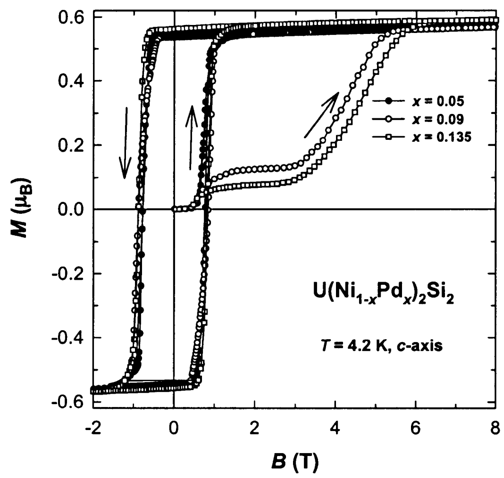

Fig. 4. Comparison of the virgin magnetization curves and hysteresis loops at 4.2 K for compounds with $x = 0.05$, 0.09 and 0.135.

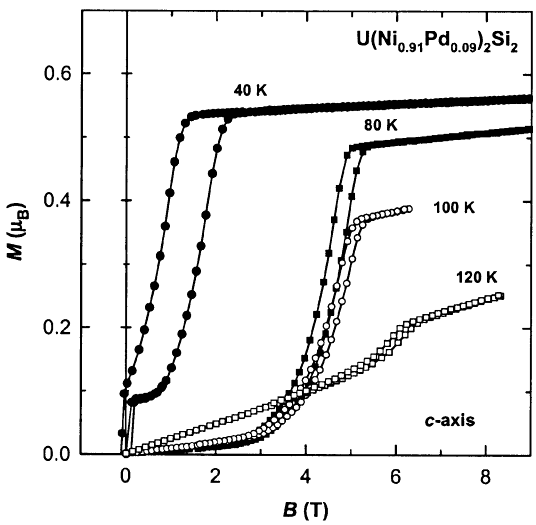

Fig. 5. The magnetization isotherms along the c-axis of the $U(Ni_{0.91}Pd_{0.09})_2Si_2$ crystal at selected temperatures.

the resistivity behavior of $U(Ni_{0.91}Pd_{0.09})_2Si_2$ is reminiscent of UPd_2Si_2 [6]. This would, however, suggest the entire AF-I structure in the ground state exists also for $U(Ni_{0.91}Pd_{0.09})_2Si_2$.

On the other hand, the temperature dependences of magnetization (both for the ZFC and FC curves) measured in 0.01 T (see Fig. 2b) exhibit anomalies below 50 K, which are qualitatively similar to those observed for $U(Ni_{0.95}Pd_{0.05})_2Si_2$ and UNi_2Si_2 [2]. The low-temperature FC magnetization value is, however, 5 times lower than that for $U(Ni_{0.95}Pd_{0.05})_2Si_2$. The $M(T)$ dependence in the vicinity of T_2 and T_N is very similar to that for $x = 0.05$ and is not shown in Figure 2. The positions of the two magnetization maxima in the ZFC curve reflect a slight increase (decrease) of T_2 (T_1) observed with increasing Pd content, which points to stabilization of the AF-I phase. Also the T_N value is somewhat higher with respect to that of the 5% -Pd compound. Similar conclusions can be drawn from the analysis of resistivity data.

Also the magnetization curves measured for $U(Ni_{0.91}Pd_{0.09})_2Si_2$ at temperatures up to 40 K are different from corresponding data obtained for the sample with lower Pd concentration. At 4.2 K, two successive magnetization steps around $0.5 (= B_{c1})$ and 4.5 T $(= B_{c2})$ are seen in the virgin curve (Fig. 4). The hysteresis loop, however, resembles strongly $U(Ni_{0.95}Pd_{0.05})_2Si_2$ and UNi_2Si_2 [2]. The remanent magnetic moment equals to 0.53 μ_B/f.u. Further $M(B)$ cycling repeats the hysteresis loop and the heating up to ~50 K is needed to restore the initial state. This magnetization behavior serves an outstanding example of a material, which exhibits the virgin curve being mostly outside the hysteresis loop. In our understanding, the steps on the virgin magnetization curve reflect field induced magnetic phase transitions. The type of hysteresis loop confirms that we deal with an irreversible transition from the AF-I to the UAF phase. In loose words we may say that $U(Ni_{0.91}Pd_{0.09})_2Si_2$ at 4.2 K represents a rare case of a permanent magnet (ferromagnet) induced by magnetic field from an antiferromagnet.

At 40 K a two-step metamagnetic transition is observed in the magnetization curve (Fig. 5). In this case the values of critical fields are $B_{c1} \simeq 0.1$ T and $B_{c2} \simeq 1.5$ T. The upper transition shows a large hysteresis of approximately 1 T. Except for the small spontaneous magnetization component ($\sim$ 0.1 μ_B/f.u.) the 40 K magnetization curves can be attributed to metamagnetic processes in an antiferromagnet. The spontaneous magnetization component vanishes above 40 K and the magnetization curves at higher temperatures resemble the behavior of the compounds with lower Pd content. The metamagnetic transition is, however, shifted to higher (lower) fields with increasing Pd concentration in the interval of $80 < T < 100$ K ($100 < T < 120$ K). This result suggests that the Pd substitution for Ni stabilizes the AF-I phase and destabilizes the LSDW phase with respect to the application of magnetic field.

3.3 $U(Ni_{0.865}Pd_{0.135})_2Si_2$

Electrical resistivity data obtained for this composition (Fig. 3) strongly resemble results observed for $U(Ni_{0.91}Pd_{0.09})_2Si_2$, only the T_2-anomaly is shifted to slightly higher temperatures. The temperature dependence of magnetization seen in Figure 2b also shows features analogous to these observed for the compounds with lower Pd content. The low temperature magnetization measured in 0.01 T is reduced in this case down to about 5% of the value observed for $U(Ni_{0.95}Pd_{0.05})_2Si_2$.

The $M(B)$ curves (Figs. 4, 6) in many aspects resemble the corresponding magnetization data obtained for the compound with $x = 0.09$. At 4.2 K, a two-step metamagnetic transition is observed with $B_{c1} \sim 0.5$ T. The upper magnetization step becomes broader and is shifted to slightly higher magnetic fields (Fig. 4). With increasing temperature, the evolution of magnetization curves

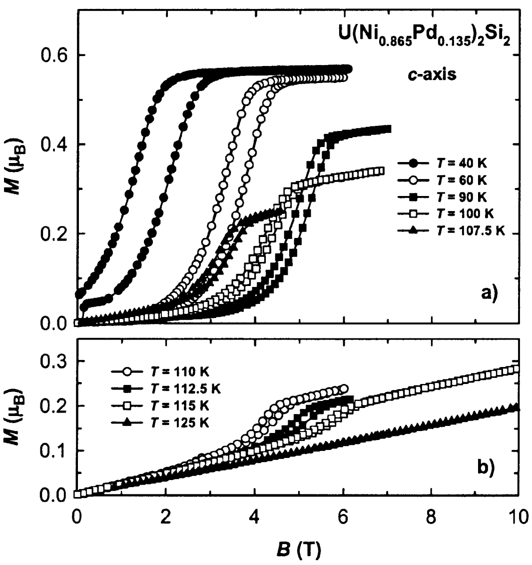

Fig. 6. The magnetization isotherms along the c-axis of the $U(Ni_{0.865}Pd_{0.135})_2Si_2$ crystal at selected temperatures in the AF-I range (a) and in the LSDW range (b).

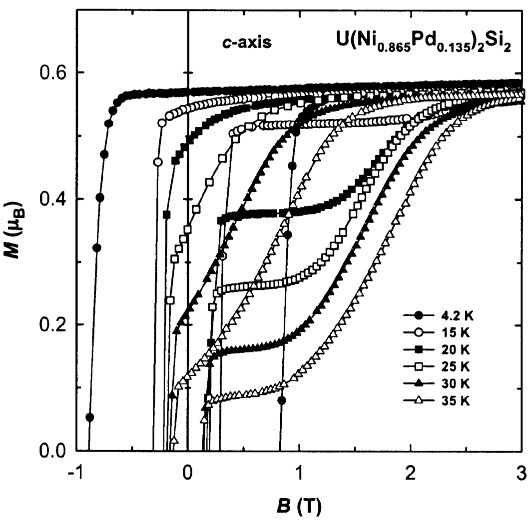

Fig. 7. Details of low temperature hysteresis loops of $U(Ni_{0.865}Pd_{0.135})_2Si_2$.

is very similar to that observed for $U(Ni_{0.91}Pd_{0.09})_2Si_2$ (Fig. 6). The close similarity of the 4.2 K magnetization curves measured for the two compounds shows that the conclusion about the irreversible AF-I → UAF transition drawn above for $U(Ni_{0.91}Pd_{0.09})_2Si_2$ is without doubt valid also for $U(Ni_{0.865}Pd_{0.135})_2Si_2$. With increasing temperature, the irreversibility becomes less pronounced; a part of volume transfers back to the AF-I state. Figure 7 shows details of hysteresis loops in $U(Ni_{0.865}Pd_{0.135})_2Si_2$ below 40 K. In a wide field interval, the AF-I and UAF phases coexist which results in the plateau in the $M(B)$ curve. The decreasing height of plateau shows how the AF-I volume fraction grows on account of UAF with heating.

The low-temperature magnetization values observed for $U(Ni_{0.91}Pd_{0.09})_2Si_2$ and $U(Ni_{0.865}Pd_{0.135})_2Si_2$ in 0.01 T represent only small fractions of those observed for $U(Ni_{0.95}Pd_{0.05})_2Si_2$. This result may be interpreted either in terms of emerging new "UAF" phases with very complicated stacking that would yield a spontaneous moment of approximately 0.08 μ_B/f.u. and 0.02 μ_B/f.u. This could be achieved only by a very complex stacking of U moments leading to a very large magnetic elementary cell, which would imply observation of a strong increase of the electrical resistivity at T_1 with decreasing temperature, which is apparently not the case. In this model also the state at the plateau in the $M(B)$ curves in Figure 7 would be considered as a homogeneous state with complex stacking of U moments instead of a mixture of AF-I and UAF phases.

Another scenario may be formulated in terms of the above discussed coexistence of the AF-I and UAF phases at temperatures below T_1 with fractional volumes depending on the composition of the Ni/Pd sublattice and on external parameters (magnetic field, temperature or pressure). The dramatic reduction of the low temperature FC (in 0.01 T) magnetization with Pd doping should then reflect the rapidly decaying fractional volume of the UAF phase to become less than 4% of the total volume in $U(Ni_{0.865}Pd_{0.135})_2Si_2$. This scenario could explain also the absence of resistivity anomaly at T_1 both for the $U(Ni_{0.91}Pd_{0.09})_2Si_2$ and $U(Ni_{0.865}Pd_{0.135})_2Si_2$ compounds. The majority volume with the AF-I characterized by the lower resistivity in these two compounds bypasses the more resistive UAF clusters, which are emerging below T_1. Nevertheless, the possible appearance of a new magnetic phase with different propagation vector cannot be entirely ruled out. Therefore, the microscopic experiments, especially the neutron diffraction, are strongly desired in order to confirm the ground state of the $x = 0.09$ and 0.135 samples.

We measured also the temperature dependence of susceptibility at temperatures between T_N and room temperature in a field of 4 T applied along the c-axis. In case of all our samples and also UNi_2Si_2 and UPd_2Si_2 the susceptibility follows well the Curie-Weiss law. The values of Θ_p vary from −4 K in UNi_2Si_2 [10] to 2 K for $U(Ni_{0.865}Pd_{0.135})_2Si_2$. A linear extrapolation of this trend to UPd_2Si_2 yields 36 K, which compares very well with 40 K [6]. The values of the effective moment μ_{eff} for $U(Ni_{1-x}Pd_x)_2Si_2$ appear in the narrow interval between 3.4 and 3.5 μ_B/U atom. (3.34 μ_B for UNi_2Si_2 and 3.5 μ_B for UPd_2Si_2). The ordered-moment value does not show any considerable dependence on the Pd concentration ($\mu_{ord} = 0.55 \pm 0.02$ μ_B). This reflects the stability of the U magnetic moment with respect to the varying chemical neighborhood of U atoms whereas the exchange interactions between the U moments is somewhat sensitive to the changes in the Ni/Pd sublattice.

4 Conclusions

In conclusion, we have prepared single crystals of $U(Ni_{1-x}Pd_x)_2Si_2$ compounds with $x = 0.05$, 0.09 and 0.135. Magnetization isotherms at different temperatures and temperature dependences of magnetic susceptibility and electrical resistivity have been measured. The magnetic properties of $x = 0.05$ sample is similar to that of pure UNi_2Si_2 and it seems to have entirely the UAF ground state. On the other hand, in the ground state of the samples with $x = 0.09$ and 0.135, very small magnetic moment has been observed in the ZFC sample at low temperatures. Moreover the increase of electrical resistivity accompanied by a magnetic order-order transition from AF-1 to UAF has not been detected for $x \geq 0.09$. Application of a sufficiently high magnetic field along the c-axis causes an irreversible transition to the UAF phase at lowest temperatures. The UAF phase persists at low temperatures after removing external magnetic field. These phenomena can be attributed to the ground state of $x = 0.09$ and 0.135 samples being characterized by the coexistence an AF-I and UAF with a small volume fraction of the UAF phase. In the mean time between submission of this paper and final acceptance we performed a neutron diffraction study of the two $U(Ni_{1-x}Pd_x)_2Si_2$ single crystals with $x = 0.09$ and 0.135 [11]. We have confirmed validity of the proposed scenario including the fact that the Néel temperature (T_N) of both these compounds is almost the same as in UNi_2Si_2, *i.e.* $\simeq 125$ K, because clear magnetic neutron diffraction reflections relevant to AF ordering with $\boldsymbol{q} = (0,0,0.76)$ have been observed below this temperature.

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