Modulated spin-density waves in uranium intermetallic compounds with ThCr$_2$Si$_2$ structure

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Abstract

The crystal and magnetic structures of U(Pd$_{1-x}$M$_x$)$_2$Ge$_2$ compounds with M = Fe, Co, Ru are studied by neutron powder diffraction and µSR. The effects of M doping and external applied pressure on the crystal and magnetic structures are compared. We find that even at small M-doping level the magnetic structure drastically changes, while the values of the lattice parameters and interatomic distances change only slightly. In contrast, a high external pressure modifies the crystal structure more significantly, while the magnetic structure remains the same. These observations allow us to conclude that the drastic changes of magnetic structure of U(Pd$_{1-x}$M$_x$)$_2$Ge$_2$ compounds with increased M doping could result from modifications of the RKKY indirect exchange interactions owing to a variation of conduction-electron number per U atom, rather than from lattice contraction.

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Neutron-diffraction studies of UPd$_2$Ge$_2$ [1] below the Néel temperature $T_N \sim 140$ K established that the magnetic structure is a square-modulated commensurate antiferromagnetic (AF) longitudinal spin-density wave (LSDW), propagating along the c-axis for $T < 75$ K, and that it is an incommensurate LSDW for $75 < T < 140$ K.

Magnetization measurements of U(Pd$_{1-x}$Fe$_x$)$_2$ Ge$_2$ [2] indicated two additional magnetic transitions near 87 and 50 K and the possible coexistence of three magnetic phases: ferromagnetic, AF, and spin glass. The most prominent changes are found at the Fe doping level $x = 0.02$. The end member of U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$–UFe$_2$Ge$_2$ is paramagnetic over the entire measured temperature region.
Neutron-diffraction studies of U(Pd$_{0.98}$Fe$_{0.02}$)$_2$Ge$_2$ are described elsewhere [3]. Increasing of doping level up to 2% of Fe in U(Pd$_2$Ge$_2$ changes the magnetic structure. In this article, we present the results of neutron diffraction and µSR studies of U(Pd$_{1-x}$M$_x$)$_2$Ge$_2$ with $x = 0.015, 0.03$ and M = Fe, Ru and Co.

Polycrystalline samples U(Pd$_{1-x}$M$_x$)$_2$Ge$_2$ were prepared by arc melting under an Ar atmosphere using stoichiometric quantities of the initial components.

Neutron-diffraction experiments were performed at the high-resolution fourier diffractometer and DN-12 diffractometer at the IBR-2 pulsed reactor in the Frank Laboratory for Neutron Physics (Dubna), G4.2 and G6.1 diffractometers at the Orphée reactor at the Léon Brillouin Laboratory (Saclay), and DMC diffractometer at the SINQ spallation source at the Paul Scherrer Institute (Villigen). The data were analyzed by the Rietveld method using MRIA and Fullprof programs [4,5]. Zero-field µSR experiments were performed with the GPD instrument at the Paul Scherrer Institute.

All samples had the same body-centered-tetragonal crystal structure of the ThCr$_2$Si$_2$-type with space group I4/mmm and atoms in the following positions: U in 2(a): (0 0 0) Pd (Fe) in 4(d) $(0 \frac{1}{2} \frac{1}{2})$, Ge in 4(e) $(0 \frac{1}{2} 0)$. Our previous study [6] showed that the magnetic structure of pure U(Pd$_2$Ge$_2$ and of the compound with 1% Fe are practically the same. At $T < 50$ K, we observed only the first- and third-order magnetic satellites in neutron-diffraction patterns. In this case, the magnetic structure can be described as a square-modulated LSDW with propagation vectors $k = (0 0 k_z)$ and $k_z = \frac{1}{3}$. The modulation is changed in the region of 65–90 K from square-shaped to more sinusoidal and the third harmonic decreases rapidly but $k_z$ is locked at $k_z = \frac{1}{3}$. Above 90 K, the $k_z$ value reduces down to 0.72 and all magnetic reflections disappear near $T_N \sim 140$ K.

The neutron-diffraction pattern of U(Pd$_{0.98}$Fe$_{0.02}$)$_2$Ge$_2$ at low temperature is shown in Fig. 1. From these data it follows that at low temperature the sample with 1.5% of Fe possesses a simple AF structure with wave vector $k = c^*$. This magnetic structure is similar to that observed in samples with $x = 0.02$ [3]. Thus, we can conclude that increasing the doping level from $x = 0.01$ to 0.015 leads to changes of the magnetic structure at $T < 65$ K from square-modulated LSDW to simple AF.

Our experiments with $x = 0.03$ and 0.08 have shown that additional doping with Fe does not lead to changes in the type of magnetic structure. Only a slight decrease of the Néel temperature with increasing $x$ was observed.

Zero-field µSR studies of U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ with $x = 0.01, 0.015, 0.02$ have also been performed. In the sample with simple AF structure ($x = 0.02$), we observed one-component muon-spin precession (see Fig. 2) with frequency $F \sim 35$ MHz in the region with simple AF structure ($T < 85$ K), the frequency decreasing to zero in the region of incommensurate sinusoidal modulation $85 < T < 140$ K [7]. In the sample with $x = 0.01$, the muon-spin precession in zero external field was also observed below $T_N$. The signal has one component at $100 < T < 140$ K, and two components with two different muon-precession frequencies below $T = 95$ K [7]. This result is in good agreement with the neutron-diffraction data [8]. In the case of a square-modulated commensurate magnetic structure, U-atoms occupy two different magnetically equivalent positions. So, two different dipole fields, $B_1$ and $B_2$ are seen by the muons.

In Fig. 3, the temperature dependence of the muon precession frequency for the sample with
transition from square-modulated to simple AF occurs at low temperature, while at intermediate temperatures 55 \( < T < 80 \) K a mixed square-modulated/sinusoidal modulated phase exists. This phase completely disappears with further increase of the iron doping level up to 2%.

Hence, the magnetic structure in the U(Pd\( _{1-x} \Fe_x \))\(_2\)Ge\(_2\) series undergoes significant changes with variation of the Fe-doping level between \( x = 0.01 \) and 0.015. There were several theoretical attempts to explain the magnetic phase diagram of A(M\( _{1-x} \M' \))\(_2\)Ge\(_2\) compounds. In Ref. [6], a phenomenological theory of the magnetic phase transitions in these compounds taking into account the heterogeneities of the structure produced by doping was developed. It explains the change in the magnetic structure by a decrease of the unit-cell volume owing to the substitution of Fe atoms for Pd ones.

We performed series of neutron powder diffraction measurement of U(Pd\( _{1-x} \M_x \))\(_2\)Ge\(_2\), with \( M = Ru, Co \), \( x = 0.01, 0.015 \). These elements were chosen because their atomic radii differ from that of Fe. We found that the magnetic structure of these compounds is the same as that of compounds with Fe substitution at similar \( x \) values. However, the changes in unit-cell volume at these concentrations is only \( \sim 0.05\% \), while for Fe a concentration of 0.1% leads to changes of unit-cell volume by \( \sim 0.2\% \) [9].

We performed neutron-diffraction experiments on U(Pd\( _{0.985} \Fe_{0.015} \))\(_2\)Ge\(_2\) and U(Pd\( _{0.98} \Fe_{0.02} \))\(_2\)Ge\(_2\) at external pressures up to 4.1 GPa. The comparison between the lattice parameters and interatomic distances [9] shows that high external pressure induces more pronounced changes in the crystal structure of U(Pd\( _{1-x} \Fe_x \))\(_2\)Ge\(_2\) compound than the “chemical” pressure (variation of Fe content). An increase of Fe content \( x \) from 0 to 0.03 at fixed temperature \( T = 10 \) K leads to relative decreases of the lattice parameters \( \Delta a/a_0 = -1.41 \times 10^{-3} \), \( \Delta c/c_0 = -5.19 \times 10^{-4} \) and of interatomic distance U–M (M = Pd/Fe) \( \Delta d_{U-M}/d_{(U-M)} = -8.49 \times 10^{-4} \). A pressure increase up to 4.1 GPa leads to much higher changes of the corresponding values for U(Pd\( _{0.98} \Fe_{0.02} \))\(_2\)Ge\(_2\):

\[
\begin{align*}
\Delta a/a_0 &= -1.48 \times 10^{-2}, \\
\Delta c/c_0 &= -3.24 \times 10^{-3}, \\
\Delta d_{U-M}/d_{(U-M)} &= -7.89 \times 10^{-3}
\end{align*}
\]

at ambient
temperature. The $c/a$ ratio remains nearly the same with an increase of $x$ at ambient pressure, $c/a \approx 2.44$ and increases slightly with increasing pressure up to 4.1 GPa from 2.44 to 2.46 (for the sample with $x = 0.02$).

Endstra et al. [10] suggested the “f–d hybridization model” to explain the magnetic-ordering temperatures of $MT_2X_2$ compounds ($M = U, Ce; T = 3d, 4d, or 5d$ transition metal; and $X = Ge, Si$). In this model, an ordering of 5f spins of $U$ appears as the result of competition between an indirect f–d–f magnetic interaction of the local U moments (RKKY-type interaction) and a Kondo-type spin-compensating mechanism. However, in our case the observed changes of magnetic structure of $U(Pd_{1-x}Fe_x)_2Ge_2$ caused by Fe substitution of Pd atoms at ambient pressure could not be explained in terms of the “f–d hybridization model” by the modification of hybridization matrix elements since their values $V_{df} \propto 1/d_{U-M}^6$ and the U–M distance vary very slightly for the studied Fe doping level $x < 0.03$.

A possible explanation of the drastic changes in the magnetic structure of $U(Pd_{1-x}Fe_x)_2Ge_2$ caused by Fe substitution is the modification of the RKKY-type indirect exchange interaction via variations of the conduction-electron number per U atom.

References