Modulated magnetic structure of U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ studied by µSR

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Abstract. The magnetic ordering of the ternary intermetallic compound U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ ($x = 0.01$, 0.02) was studied by μSR in the temperature range $T = 15$–300 K. The low temperature state changes from the modulated longitudinal spin density wave LSDW ($x = 0.01$) with a propagation vector $k = [0\,0\,3/4]$ ($T_N = 140$ K) to a simple antiferromagnetic structure AF ($x = 0.02$) with $k = [0\,0\,1]$ ($T_N = 140$ K). In the AF phase (sample with $x = 0.02$) the uranium magnetic moment shows a monotonic temperature dependence. In the LSDW phase with $k = [0\,0\,3/4]$ (sample with $x = 0.01$) the magnetic moments of uranium atoms located in two magnetically nonequivalent positions $(0\,0\,0)$ and $(0\,0\,1)$ have different temperature dependences. One moment assumes a maximum at $T_{m1} = 50$ K, while the other one does not display any peculiarities. The results are discussed within a model of the superposition of a square and sine modulation and are compared with neutron diffraction data. The observed peculiar temperature behaviour of the moments is connected with the temperature variation of the initial phase φ of the LSDW modulation by $\Delta\varphi = 0.2$ rad.

1. Introduction

Most of the intermetallic compounds UT$_2$X$_2$ (where T = 3d, 4d or 5d transition metal and X = Ge or Si) crystallize in a body centred tetragonal structure. High magnetic anisotropy along the tetragonal c axis and indirect RKKY interaction between uranium magnetic moments lead to a great variety of magnetic states in these systems [1]. Magnetic susceptibility and magnetization studies of U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ showed that even a small Fe doping completely changes the type of magnetic ordering [2]. From magnetic susceptibility data in the range of $x = 0.01$–0.02 two additional transitions $T_{m1} \sim 50$ K and $T_{m2} \sim 80$ K have been found below the Néel temperature $T_N = 140$ K. The intensities of the magnetic susceptibility peaks are strongly dependent on the iron doping level x. The results of macroscopic investigations at temperatures below T_N in the range of $x = 0.01$–0.02 were interpreted in terms of the coexistence of antiferromagnetic (AFM), spin-glass (SG) and ferromagnetic (FM) phases. According to neutron diffraction

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data [3] UPd_2Ge_2 has an antiferromagnetic structure with longitudinal spin density wave modulation (LSDW), which has a commensurate form of modulation with a propagation vector $k = [0\,0\,3/4]$ at low temperatures. The shape of the modulation changed from 'squared' to 'sinusoidal' at 100 K and, finally, has an incommensurate sine modulation with $k = [0\,0\,k_z]$ in the temperature range $100 < T < T_N$. Iron doping changes the magnetic structure [4], and the low temperature state of the composition with $x = 0.02$ is a collinear antiferromagnet with $k = [0\,0\,1]$, and in the range $70\,K < T < T_N$ the magnetic structure is an incommensurate LSDW similar to the $x = 0$ compound.

In general, for the modulated structures with a rational value of the propagation vector k, the magnetic moments on particular atoms cannot be determined from non-polarized neutron diffraction data. For example, for the sine-modulated magnetic structure of LSDW type in UPd_2Ge_2 the value of the magnetic moment is $m(z_i) = \mu_0 \cos(2\pi k z_i + \varphi)$, where z_i is a uranium atom coordinate along the c axis. From the diffraction data one can determine the modulation amplitude μ_0 and the propagation vector k, but the value of the initial phase of modulation φ does not affect the intensity of the Bragg magnetic peaks and, hence, is not accessible. If several propagation vectors k are present the calculation of the magnetic moments of the atoms becomes even more complicated. Obviously, the real values of the magnetic moments of atoms located inside the magnetic unit cell are dependent on the initial phase φ and, usually, some reasonable value of φ is assumed. μSR, being a local probe technique, can provide complementary information, since it allows one to determine the local magnetic field in the crystal lattice, which is directly connected with the real magnetic moments of the atoms. In addition, in a μSR experiment the atomic magnetic moments are measured with higher accuracy in comparison with neutron diffraction experiments, because μSR is very sensitive to small changes in the local magnetic configuration (of the order of 0.01 μ_B per atom.) Also, the neutron diffraction technique yields the product of the volume fraction of the magnetically ordered phase and the magnetic moment, while μSR can determine these two parameters separately, allowing us to detect the potential presence of additional phases of e.g. spin-glass-like type. We have employed the μSR technique for refining the $U(Pd_{1-x}Fe_x)_2Ge_2$ magnetic structure. Preliminary results were published elsewhere [5].

2. Experiment

Polycrystalline samples of $U(Pd_{0.99}Fe_{0.01})_2Ge_2$ and $U(Pd_{0.98}Fe_{0.02})_2Ge_2$ were prepared by arc-melting of the initial components (U, Pd, Ge, Fe) in an Ar atmosphere. The samples were annealed at $T = 800\,°C$ for 1000 h. According to the x-ray diffraction data they are crystallized in a tetragonal body centered lattice of $ThCr_2S_2$ type with $I4/mmm$ space group and unit cell parameters $a = b = 4.18$ Å, $c = 10.21$ Å. Zero field (ZF) μSR experiments have been performed with the GPD instrument at the Paul Scherrer Institute (Villigen, Switzerland) in the temperature range of 15–300 K. The samples had a disc shape with a diameter of 10 mm, a thickness of 5 mm and a weight of 5g. They were glued to a silver sample holder and mounted on the cold finger of a closed cycle refrigerator.

2.1. $U(Pd_{0.98}Fe_{0.02})_2Ge_2$ ($x = 0.02$)

According to neutron diffraction data, the low temperature state of this composition is collinear antiferromagnetic (AF) [4] with $k = [0\,0\,1]$. The uranium magnetic moments form ferromagnetic planes (*ab*) with m_U oriented parallel and antiparallel to the c axis in adjacent planes. The reported value of the magnetic moment amounts to $m_U = 2.3(1)$ μ_B [4] or

$m_U = 2.5(1)\ \mu_B$ [6].

Due to the Coulomb repulsion from the ions of UPd$_2$Ge$_2$, the positive muon is localized in symmetric interstitial sites. Possible muon sites in the compounds with crystal structures similar to UPd$_2$Ge$_2$ were determined from the angular and temperature dependence of the Knight shift in a transverse magnetic field [7], and from the muon spin depolarization, caused by the interaction with the dipolar nuclear moments [8]. Thus, in CeRu$_2$Si$_2$ with the same crystal structure as in UPd$_2$Ge$_2$ the most suitable muon site was $(1/2\ 1/2\ 0)$ [7]. In [8] Ce$_{0.95}$La$_{0.05}$Ru$_2$Si$_2$ was studied, and the $(0\ 1/2\ 1/8)$ muon site was selected among 11 possible positions as the most appropriate.

The muon spin polarization function $P(t)$ in the magnetically ordered state in zero external field can be expected to read:

$$P(t) = A \exp(-\lambda t) \cos(2\pi F t + \varphi) + A_0 \exp(-\lambda_0 t) \tag{1}$$

where $F = (\gamma_\mu/2\pi) B_\mu$ is the muon spin precession frequency in the local magnetic field B_μ acting on the muon, γ_μ is the gyromagnetic ratio $(\gamma_\mu/2\pi = 13.55\ \text{MHz kG}^{-1})$; λ is a muon spin relaxation rate; A and A_0 are the initial asymmetries (or amplitudes). The first term in equation (1) reflects the precessing component of the μ^+ polarization perpendicular to B_μ; the second term is the component along B_μ. In a polycrystalline sample $A = \frac{2}{3} A_{tot}$ of the total polarization amplitude A_{tot}. If the muons are localized in several sites possessing different magnetic fields, than the total muon spin polarization is the sum of oscillating components given by formula (1).

One-component precession with frequency $F = 35.07(4)$ MHz at 15 K (figure 1(a)) was observed in the ZF-μSR experiment in the U(Pd$_{0.98}$Fe$_{0.02}$)$_2$Ge$_2$ sample. The transition to the AF state occurs below $T_N = 140$ K. The peculiarity in the temperature dependence of the frequency $F(T)$ at $T_{m1} = 75$ K reflects the change of the local magnetic field B_μ at the transition from the incommensurate LSDW to the AF state. Figure 2 shows the experimental polarization functions $P(t)$ for the different magnetic states of the U(Pd$_{0.98}$Fe$_{0.02}$)$_2$Ge$_2$ sample.

The magnetic field B_μ acting on the muon, in an antiferromagnet, is a sum of the dipole and the contact magnetic fields:

$$B_\mu = B_{dip} + B_c \tag{2}$$

where B_{dip} is calculated as a sum of dipole fields produced by the local uranium magnetic moments $m_U(r)$:

$$B_{dip} = \sum_r \frac{1}{r^3} \left(\frac{3(r \cdot m_U(r))r}{r^2} - m_U(r) \right) \tag{3}$$

where r is a vector from the muon position to the uranium atom. B_c is a contact field produced by a nonzero conduction electron spin density n_s at the muon site by means of the RKKY interaction:

$$B_c \sim J_{sf}(0)/E_F n_s \sum_r F(2k_F r) m_U(r) \tag{4}$$

where $J_{sf}(0)$ is the s–f exchange integral and $F(x)$ is the Rudderman–Kittel function $F(x) = (1/x^4)(x \cos x - \sin x)$. Since both n_s and $J_{sf}(0)$ are unknown, one cannot estimate the contribution of the contact field to B_μ. We therefore neglect B_c. However, we note that the RKKY interaction with the localized U moments decreases with distance similar to the dipole contribution ($\sim 1/r^3$), so that the closest neighbours of the muon give the main contribution.

The values of the dipole fields B_{dip} calculated for possible muon sites in the UPd$_2$Ge$_2$ lattice are presented in table 1. A comparison of the dipole magnetic field in the AF state at different muon sites shows that the muon site $(0\ 1/2\ 1/8)$ proposed in [8] would yield a
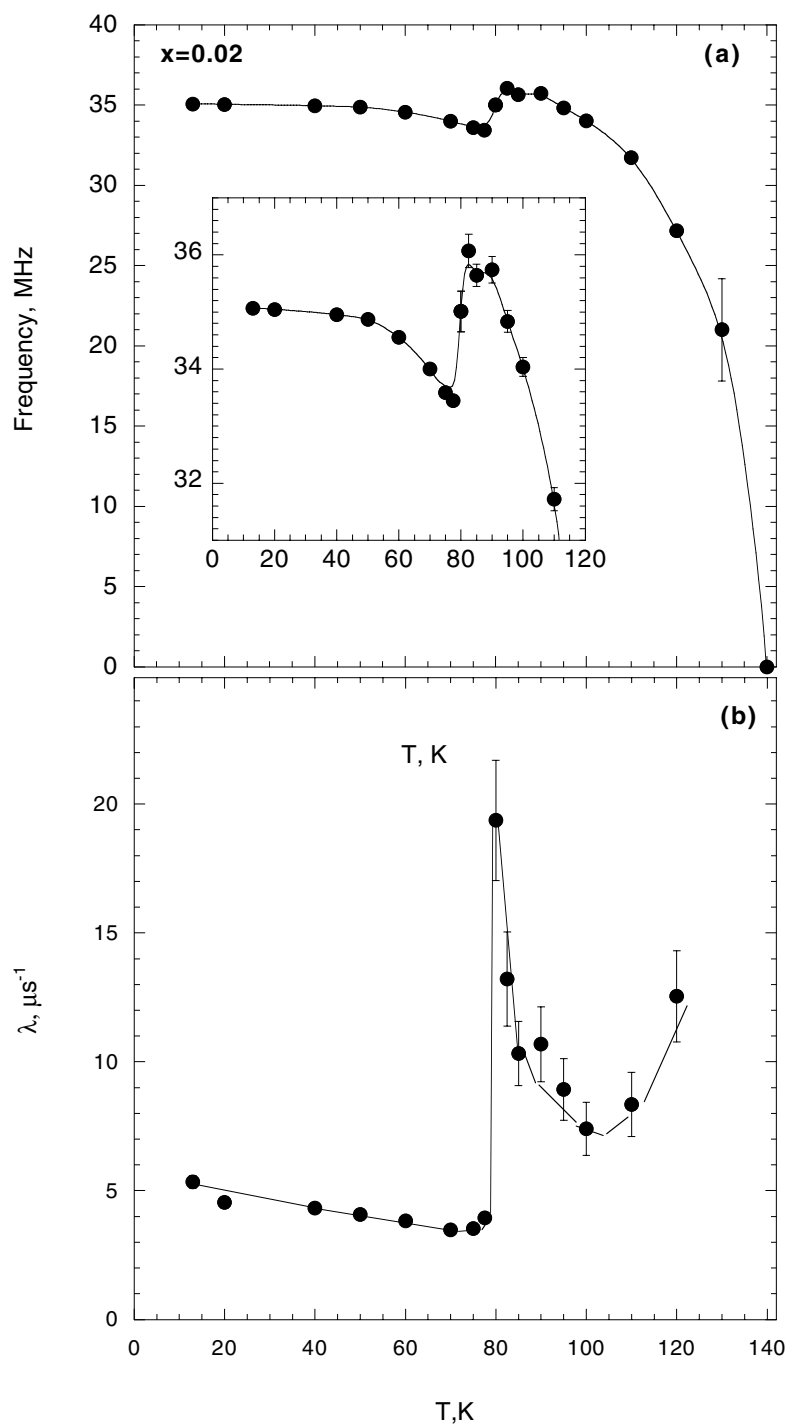
Figure 1. Temperature dependences of (a) the muon precession frequency F. The inset shows the frequency in an enlarged scale. (b) Muon spin relaxation rate λ in zero external magnetic field in $U(Pd_{0.98}Fe_{0.02})_2Ge_2$. Error bars are given for all points. They are not seen if less than the symbol size.
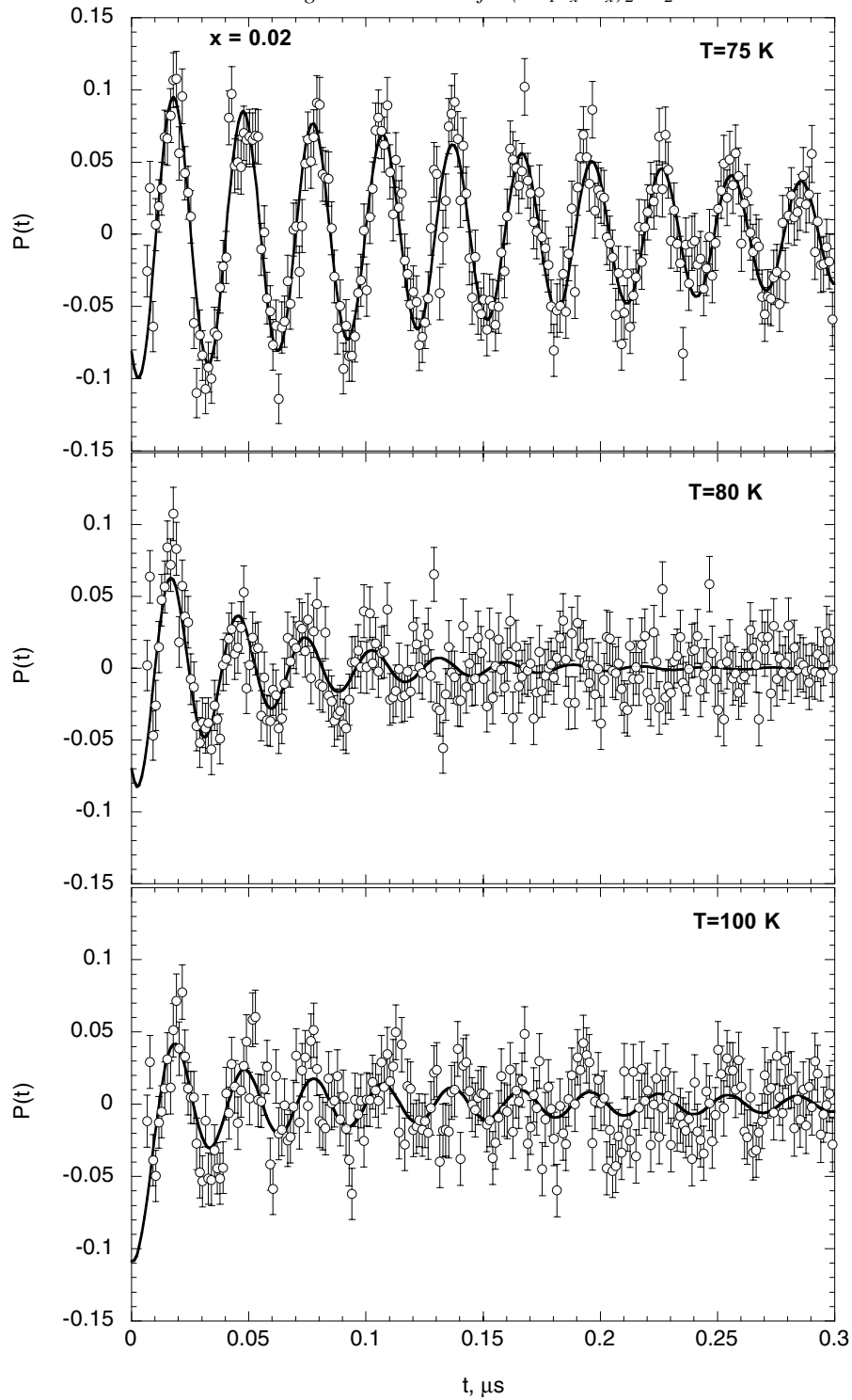
Figure 2. Time dependence of muon spin polarization $P(t)$ in zero external magnetic field in U(Pd$_{0.98}$Fe$_{0.02}$)$_2$Ge$_2$ at $T = 75$ K (in the AF phase), $T = 80$ K (at the transition from AF to LSDW) and $T = 100$ K (in the incommensurate LSDW phase). The non-oscillating part of the polarization is subtracted.

Table 1. The dipole field B_{dip} calculated for possible muon sites in the $U(Pd_{0.98}Fe_{0.02})_2Ge_2$ lattice ($I4/mmm$, unit cell parameters $a = b = 4.18$ Å, $c = 10.21$ Å) for the simple antiferromagnetic structure with $k = [0\,0\,1]$ for the uranium magnetic moment $\mu_U = 1\ \mu_B$ along the c-axis and the experimentally measured B_μ at $T = 15$ K. The column μ_U shows the uranium magnetic moment expected in the dipolar approximation from the experimentally measured B_μ.

Muon site	$B_{dip}/1\ \mu_B$ (G)	μ_U (μ_B)
0 1/2 1/8	1016	2.55
0 0 1/2	2113	1.22
1/2 1/2 0	2113	1.22
1/4 1/4 1/4	0	—
0 0 1/4	901	2.87
1/2 1/2 5/16	2067	1.25
1/2 1/4 0	2485	1.04
1/2 0 1/16	2212	1.17
1/8 1/2 3/32	1607	1.61
1/4 1/4 5/32	1099	2.35
1/8 1/8 7/32	884	2.93
Experimental B_μ	2588(3)	

uranium magnetic moment close to the experimental one implying a negligible contribution from the contact field. For the muon site $(1/2\,1/2\,0)$ found in [7] one would have to invoke the presence of a considerable negative contribution from the contact field, but since both B_c and B_{dip} are proportional to m_U this would not affect the functional form of the temperature dependence.

The peculiarity of the temperature dependence of the precession frequency F at $T_{m1} = 75$ K (figure 1(a)) is accompanied by a sharp increase of the muon spin relaxation rate (figure 1(b)) due to the increase in the magnetic field distribution at the muon site in the incommensurate modulated phase. In addition, the time dependence of the polarization function is changed: the initial phase $\phi(T)$ of the muon spin precession at $T > T_{m1}$ undergoes a jump $\Delta\phi \approx -\pi/4$, which is a direct indication of an incommensurate magnetic structure. In this case the muon spin polarization function $P(t)$ is described by a Bessel function $J_0(2\pi Ft)$ if the local field is simply represented by $B_\mu(z) = B_0 \cos(2\pi kz)$ [9]. $J_0(2\pi Ft)$ is proportional to $(\pi(Ft)^{1/2})^{-1} \cos(2\pi Ft - \pi/4)$ for large values of $2\pi Ft$. The polarization in the temperature range of the incommensurate LSDW is well described by the damped Bessel function $A \exp(-\lambda t) J_0(2\pi Ft)$. To make the relaxation rate in the AF and LSDW phases comparable we show (figure 1(b)) the values of λ obtained by fitting the experimental $P(t)$ to the formula (1) for both AF and LSDW phases. For the damped Bessel function (BF) the additional damping λ is less because of the intrinsic damping of BF: e.g. $\lambda = 2.5\ \mu s^{-1}$ instead of $\lambda = 7\ \mu s^{-1}$ at $T = 100$ K (figure 1(b)). In the whole investigated temperature range the signal amplitudes $(3/2)A = A_{tot}$ correspond to the initial polarization A_{tot}. Thus, our data do not need any assumptions about the presence of any additional magnetic phases with short range magnetic order, as was proposed in [2].

2.2. $U(Pd_{0.99}Fe_{0.01})_2Ge_2$ ($x = 0.01$)

The muon spin precession in zero external magnetic field was also observed below $T_N = 140$ K (figure 3). The signal possesses one oscillating component in the range $100K < T < 140$ K, and two oscillating components with two different muon precession frequencies F_1 and F_2 (figure 4) below $T = 95$ K. $F_1(T)$ has a monotonic temperature dependence while $F_2(T)$ has
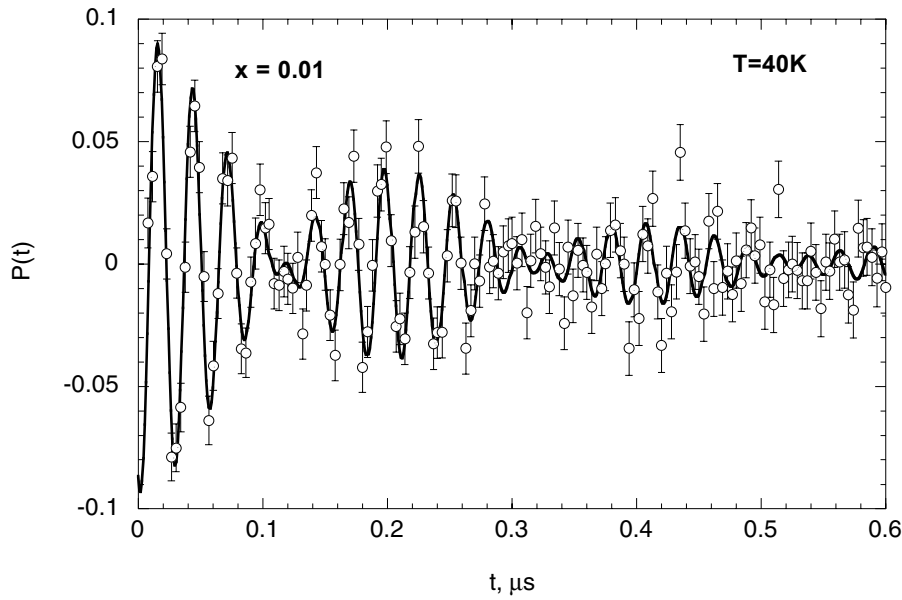
Figure 3. Time dependence of muon spin polarization $P(t)$ in zero external magnetic field in U(Pd$_{0.99}$Fe$_{0.01}$)$_2$Ge$_2$ at $T = 40$ K in the commensurate LSDW phase. The non-oscillating part of the polarization is subtracted.

a well pronounced maximum at $T_{m1} = 50$ K (the inset in figure 4). Both frequencies decrease as the temperature increases until $T_{m2} = 100$ K, where F_2 vanishes and F_1 shows a small anomaly. Above the temperature T_{m2} this sample is completely identical to the composition with $x = 0.02$: the polarization function is also well described by the damped Bessel function in the temperature range of the incommensurate phase, and the uranium magnetic moments have the same temperature dependence at $T > 80$ K.

The modulated magnetic structure of U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ is shown in figure 5. Four crystal cells along the c-axis are shown, containing three magnetic unit cells. Figure 5 also shows the variation of the z-component of the U magnetic moment $m_z(z/c)$ in the case of sine- and square-modulation forms. In general, for the magnetic structure with the propagation vector $k_z = 3/4$ there are four different absolute values of the magnetic moment of the uranium ions: m_1, m_2, m_3, m_4. In the case of a square modulation all four moments are equal. For the sine form of the modulation $m(z) = \mu_0 \cos(2\pi k z + \varphi)$ with an arbitrary value of the phase φ all four moments are different: $m_1 = \mu_0 \cos\varphi$, $m_2 = \mu_0 \cos(3\pi/4 + \varphi)$, $m_3 = \mu_0 \cos(3\pi/2 + \varphi)$, $m_4 = \mu_0 \cos(\pi/4 + \varphi)$. If one takes $\varphi = -\pi/8$ [3], then the structure is described by the two moments: $m_1 = \mu_0 \cos\pi/8$, $m_2 = \mu_0 \sin\pi/8$, $m_3 = m_2$, $m_4 = m_1$. Since at low temperatures the modulation is close to a square shape and is readily transformed to a sine shape upon increasing temperature, the suggestion that $\varphi = -\pi/8$ is most reasonable: this is the simplest magnetic configuration of the U moments in the magnetic unit cell which satisfies the neutron diffraction data. Thus, m_1 and m_3 are changed from $m_1 \approx m_3$ (square modulation) to $m_1 = m_3 \cos\pi/8/\sin\pi/8$ (sine modulation) as the temperature increases. For an arbitrary ratio between m_1 and m_3, the structure can be considered as a superposition of 'square' and 'sine'.

The muon site (0 1/2 1/8) and equivalent ones are presented in figure 5 in a unit cell quadruplicated along the c-axis. The sites $\mu 1$ and $\mu 3$ are magnetically equivalent as well
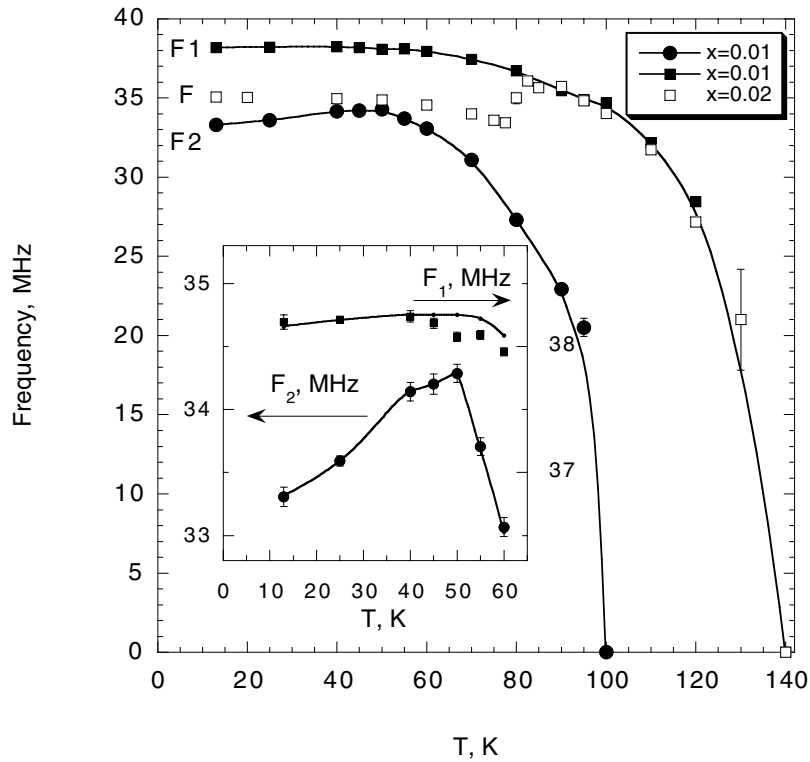
Figure 4. Temperature dependence of the muon precession frequencies F_1, F_2 in zero external magnetic field in $U(Pd_{0.99}Fe_{0.01})_2Ge_2$ (closed symbols). The lines are guides for the eyes. For comparison, the precession frequency F in $U(Pd_{0.98}Fe_{0.02})_2Ge_2$ is also shown (open symbols). The lines in the inset are the result of fitting to (3) with $m_\square = 2.45\ \mu_B$, $\mu_0 = 0.29\ \mu_B$.

as the sites $\mu2$ and $\mu4$. The value of the dipole magnetic field at the $\mu1$ site is equal but antiparallel in orientation to the dipole magnetic field at the $\mu3$ site. The same situation applies for the $\mu2$ and $\mu4$ sites. So, only two different dipole fields, B_1 and B_2, are seen by the muons. Since the crystal lattice is highly anisotropic ($c/a = 2.4$) the main contribution to the dipolar field B_1 and B_2 arises from the moment m_1 for B_1, and from m_3 for B_2 respectively. Dipole field calculations give the following numerical coefficients α for the contributions of the uranium moments m_1 and m_2 to the experimentally observed frequency: $F_1 = \alpha_1 m_1 + \alpha_2 m_2$. $\alpha_1 = 13.87$ and $\alpha_2 = 0.11$ MHz μ_B^{-1}. Thus, with an accuracy of 0.8% the frequency F_1 depends only on the moment m_1. Apparently, the same is valid for $F_2 = \alpha_1' m_3 + \alpha_2' m_4$. Due to slightly different configurations of the moments m_2 and m_4 around the μ_1 and μ_2 muon sites α_2' is slightly different from α_2 by $\alpha_2 - \alpha_2' = 0.001$ MHz μ_B^{-1}, while $\alpha_1' = \alpha_1$. The experimentally observed difference between the precession frequencies is $F_1 - F_2 \approx 5$ MHz (see figure 4), and also the pronounced change in the frequency $\Delta F_2 \approx 1$ MHz in the vicinity of the maximum at $T = 50$ K exceeds by far possible contributions from the moments m_2 and m_4. So, the experimental frequencies are a direct measure of the values of the moments m_1 and m_3 and figure 4 directly reflects the temperature dependence of the corresponding magnetic moments $m_1(T) = F_1(T)/\alpha_1$ and $m_3(T) = F_2(T)/\alpha_1$ with an accuracy of 0.8% arising from the small contribution from m_2 and m_4.

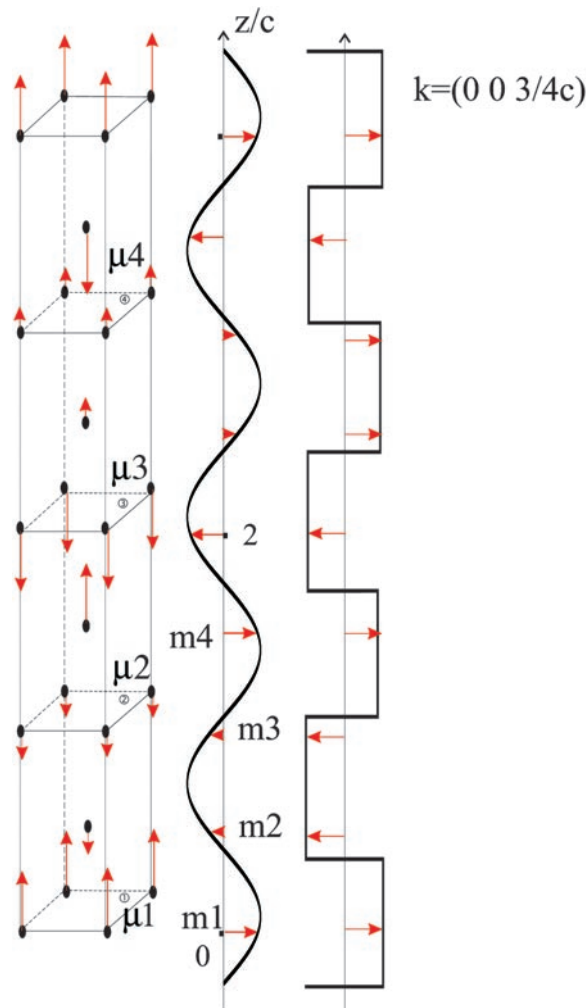For the following discussion an important point concerning the muon site is that the

Figure 5. Modulated magnetic structure of U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ (only uranium atoms are shown). Four crystal unit cells are shown containing three magnetic unit cells. The muon sites (0 1/2 1/8) are marked as $\mu1$, $\mu2$, $\mu3$, $\mu4$. The plots show the z-component of the uranium moments for the sine and square modulation shapes as a function of z.

frequencies F_1 and F_2 are mostly determined by the nearest uranium magnetic moments, so that $\alpha_1 \gg \alpha_2$. This will also be valid for the second possible muon site (1/2 1/2 0) [7] because it lies even closer to the U plane and both dipole and contact contributions from the U moments in the (0 0 1/2) and (0 0 1) (ab)-planes are small due to the ($1/r^3$) strength of both types of interaction (formulae (3), (4)). The dipolar contribution to α_2 for this site is $\alpha_2/\alpha_1 = 0.5\%$, similar to the (0 1/2 1/8) muon site. The absolute values of the uranium magnetic moment will be slightly different due to the different value of the α_1-coefficient (α_1 for this muon site can include both the dipole and the contact contributions), but the magnetic moment ratio (m_1/m_3) and all the relative changes in the moments as a function of temperature will be preserved.

3. Discussion

The real physical values are the magnetic moments of the uranium atoms. However, an expansion of the moments into two component ('square' + 'sine') modulations allows one to perform a direct comparison with the neutron diffraction data. The total moments are given by:

$$m_1 = m_\square + \mu_0 \cos \varphi$$
$$m_3 = m_\square - \mu_0 \sin \varphi \tag{5}$$

where $\varphi = -\pi/8$; m_1 and m_3 are the absolute values of the uranium magnetic moments at the $(0\,0\,0)$ and $(0\,0\,1)$ positions (see figure 5), which are directly measured in the μSR experiment; μ_0 and $m_\square$ are the amplitudes of square and sine modulation. After the transformation of (5) the amplitudes of modulation read:

$$\mu_0 = (m_1 - m_3)/(\cos \pi/8 - \sin \pi/8)$$
$$m_\square = (m_3 \cos \pi/8 - m_1 \sin \pi/8)/(\cos \pi/8 - \sin \pi/8). \tag{6}$$

In a neutron diffraction experiment, the measured values are the modulation amplitudes μ_k, corresponding to the harmonics with the propagation vectors k, $3k$, ... The actual values of magnetic moments of the atoms in the lattice are reconstructed from the amplitudes μ_k. Taking into account the Fourier expansion of the square modulation

$$m_\square(z) = m_\square 4/\pi (\cos 2\pi k z - \tfrac{1}{3} \cos 6\pi k z + \cdots) \tag{7}$$

we have the values of modulation amplitudes for two phases with the propagation vectors k and $3k$:

$$\mu_k = \mu_0 + 4/\pi m_\square$$
$$\mu_{3k} = 4/3\pi m_\square. \tag{8}$$

Figure 6 shows $\mu_k(T)$ and $\mu_{3k}(T)$ calculated from the precession frequencies $F_1(T)$ and $F_2(T)$ by using formulae (6), (8). In UPd$_2$Ge$_2$ which possesses a magnetic structure similar to $x = 0.01$ the amplitudes μ_k and μ_{3k} were directly measured by neutron diffraction [3]. The neutron and muon data are in qualitative agreement, but μ_k and μ_{3k} measured by neutron diffraction have no peculiarities around $T = 50$ K [3], while the real magnetic moment $m_3(T)$ shows a maximum. One explanation is that the neutrons do not see this maximum, because the change of the amplitude of μ_{3k} is about 0.05 μ_B, which is within the errorbars of 0.1 μ_B of the diffraction data. In terms of the 'square' + 'sine' model, the temperature of the maximum of $m_3(T)$ corresponds to the maximum contribution of the 'square' modulation.

The maximum of the magnetic moment $m_3(T)$ can be related to the temperature variation of the initial phase of the modulation $\varphi(T)$. Indeed, if φ is close to zero, then the variation of the phase obtained by changing the temperature would lead to a pronounced change of the moment $m_3(\varphi)$. On the other hand, $m_1(\varphi)$ would be almost unaffected, because the derivative of the cosine is close to zero at $\varphi \approx 0$. Eliminating the phase φ from equation (5), we derive m_1 as a function of m_3:

$$m_1(m_3) = m_\square + (\mu_0^2 - (m_3 - m_\square)^2)^{1/2}. \tag{9}$$

Figure 7 shows experimental values of m_1 as a function of m_3 and the result of fitting to the formula (9) with the assumption that $m_\square$ and μ_0 are constant values in the temperature range of $T \leqslant 60$ K. Equation (9) is in fact the equation of a circle with the centre at a point $(m_\square, m_\square)$ and μ_0 as radius. The value of the phase is $\varphi = -0.2$ rad $(\approx -\pi/16)$ at $T = 15$ K.
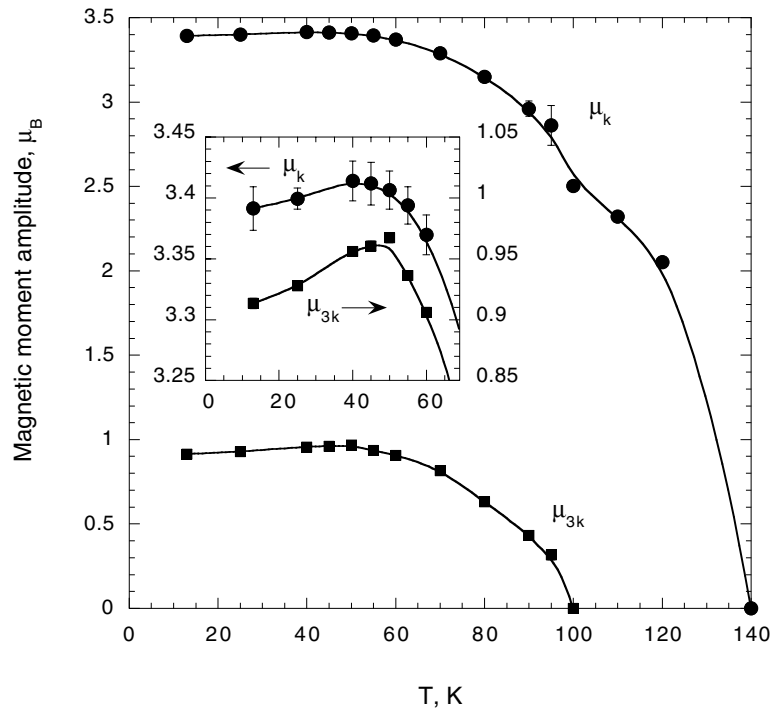
Figure 6. Temperature dependence of the magnetic moment modulation amplitudes μ_k and μ_{3k} for the propagation vectors $k = 3/4$ and $9/4$, calculated from the magnetic moments m_1 and m_3, determined from the muon spin precession frequencies in U(Pd$_{0.99}$Fe$_{0.01}$)$_2$Ge$_2$. The lines are guides for the eyes.

By increasing the temperature up to $T = 50$ K, the phase reaches zero value. With further temperature increase, $\varphi(T)$ recovers its low temperature value again. Due to the body centred lattice the modulation of the U moments at the position $(+1/2 + 1/2 + 1/2)$ should have the phase shift $-\varphi - \pi/4$. Otherwise the U moments m_2 and m_3 would have different values and the μSR signal would contain four frequencies instead of two. The temperature dependences of the $F_1(T)$ and $F_2(T)$ precession frequencies were calculated according to equation (5), with the $\varphi(T)$-values obtained by fitting equation (9) to the experimental $m_1(m_3)$. The result of the calculation (solid line) together with the experimental data (symbols) is presented in the inset of figure 4. The maximum of the temperature dependence of the $m_3(T)$ moment at $T_{m1} = 50$ K coincides with the maximum of the magnetic susceptibility $\chi(T)$ [2]. However, with respect to our data, the temperature T_{m1} cannot be considered as a phase transition temperature, because no change of the magnetic state is observed.

4. Conclusions

The intermetallic compound U(Pd$_{1-x}$Fe$_x$)$_2$Ge$_2$ ($x = 0.01$, 0.02) was studied by ZF μSR in the temperature range $T = 15$–300 K. The low temperature state is changed from simple antiferromagnetic AF ($x = 0.02$) with a propagation vector $k = [0\,0\,1]$ to a modulated longitudinal spin density wave LSDW ($x = 0.01$) with $k = [0\,0\,3/4]$. Both compositions have a Néel temperature $T_N = 140$ K and their magnetic state is incommensurate LSDW at temperatures above $T_m = 75$ K and 100 K for the samples with $x = 0.02$ and 0.01
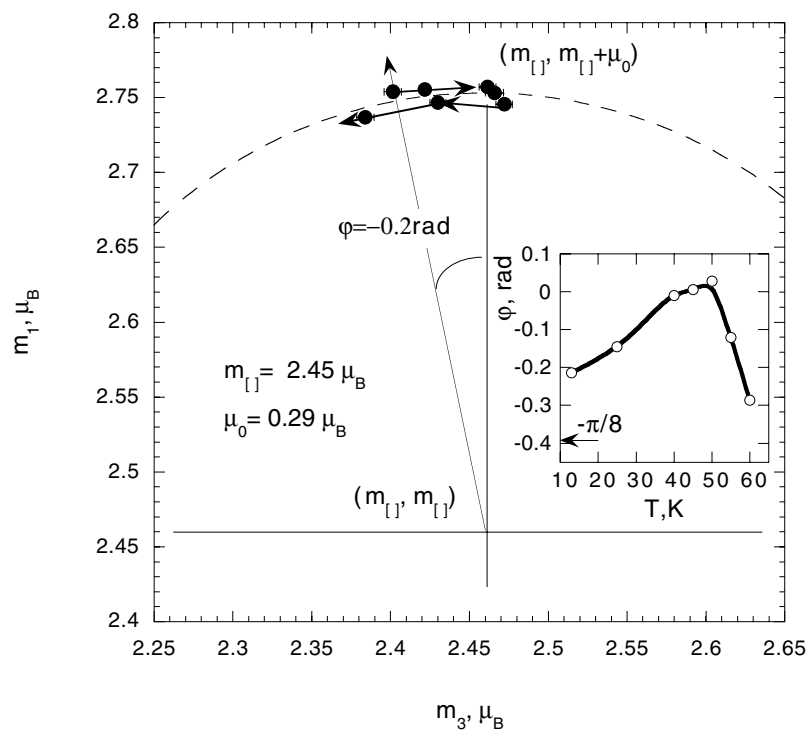
Figure 7. The magnetic moment m_1 as a function of m_3 with temperature as a parameter in $U(Pd_{0.99}Fe_{0.01})_2Ge_2$. The temperature range of $T < 60$ K is shown; the temperature decreasing is shown by arrows. The dashed line is a result of fitting to formula (6): the amplitudes of modulation are $m_\square = 2.45$ μ_B, $\mu_0 = 0.29$ μ_B. The co-ordinate system has a centre at $(m_\square, m_\square)$; a radius-vector indicates the value of phase. The inset shows the initial phase of modulation φ as a function of temperature.

respectively. In the AF phase (sample with $x = 0.02$) the uranium magnetic moment has monotonic dependence on temperature. In the LSDW phase with $k = [0\,0\,3/4]$ (sample with $x = 0.01$), the magnetic moments of uranium atoms in two magnetically nonequivalent positions $(0\,0\,0)$ and $(0\,0\,1)$ have different temperature dependences. One moment has a maximum at $T_{m1} = 50$ K, while the other one does not exhibit any peculiarities. The peculiar temperature behaviour of the moments is connected to the temperature variation of the initial phase φ of the LSDW modulation by $\Delta\varphi = 0.2$ rad.

Acknowledgments

References

[1] Sechovsky V and Havela L 1998 Magnetism of ternary intermetallic compounds of uranium *Handbook of Magnetic Materials* vol 11, ed K H J Buschow (Amsterdam: Elsevier–North-Holland) p 127
[2] Duh H M, Lyubutin I S, Jiang I M, Hwang G S and Lain K D 1996 *J. Magn. Magn. Mater.* **153** 86

Duh H M, Lyubutin I S, Wur C S, Lin K J, Hwang J S, Chang I J and Tien C 1995 *J. Phys.: Condens. Matter* **7** 2165

[3] André G, Bourée F, Olés A, Sikora W, Penc B, Szytula A and Tomkovicz Z 1996 *Solid State Commun.* **97** 923

[4] Balagurov A M, Raspopina E V, Sikolenko V V, Lyubutin I S, Stepin A S, Gribanov A V, André G, Bourée F and Duh H M 2000 *J. Magn. Magn. Mater.* **210** 225

[5] Raspopina E V, Balagurov A M, Sikolenko V V, Pomjakushin V Yu, Amato A, Schenck A and Gribanov A V 2000 *Physica* B **289–290** 282

[6] Sikolenko V V 1997 private communication

[7] Amato A, Feyerherm R, Gygax F N and Schenck A 1997 *Hyperfine Interact.* **104** 115

[8] Yamamoto Y, Marumoto K, Miyako Y, Nishiyama K and Nagamine K 1997 *Hyperfine Interact.* **104** 227

[9] Dalmas de Réotier P and Yaouanc A 1997 *J. Phys.: Condens. Matter* **9** 9113