Magnetic anomalies in UPd_2Ge_2 induced by iron doping

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From susceptibility and magnetization measurements in pure and Fe-doped UPd_2Ge_2 two magnetic transitions are found below the antiferromagnetic (AF) transition at $T_N=140$ K. The transition at $T_{m1}\approx 80$ K is very sharp and the transition at $T_{m2}\approx 50$ K is broad. The intensity of the peak susceptibility at T_{m1} and T_{m2} strongly depends on the iron doping level x, and the $x=0.02$ value is found to be a critical concentration x_c, at which many parameters change drastically. The T_{m1} peak intensity has a sharp maximum at $x=0.02$, while the T_{m2} peak almost vanishes at $x=0.02$. Both peaks disappear at $x=0.08$. At low iron doping only the AF phase is found in the range $T_{m1}<T<T_N$. At $T_{m2}<T<T_{m1}$ a spin-glass (SG) phase exists along with the AF phase, but the SG phase changes with temperature. Around the T_{m2} transition a ferromagnetic (FM) component is also present and a mixed AF-SG-FM phase is observed. At 5 K $<T<T_{m2}$ the SG component disappears, but in the AF phase a spin flip can be induced by an applied field, which implies a metamagnetic state. It seems the SG state appears at the boundary of the AF and longitudinal spin-density wave structures as a result of competition between coexisting phases. The UPd_2Ge_2 doped by 2% Fe is suggested as a candidate for the Stoner spin glass with the spin freezing temperature at 74 K.

I. INTRODUCTION

The intermetallic compound UPd_2Ge_2 has the $ThCr_2Si_2$-type crystal structure and exhibits interesting magnetic properties. At $T_N=140$ K it transforms from a paramagnetic to an antiferromagnetic (AF) state.[1–4] Neutron-diffraction studies[1] have found an incommensurate longitudinal spin-density wave (LSDW) structure in UPd_2Ge_2 at $T<140$ K. Recent magnetic investigations[2–4] revealed at least two additional magnetic transitions at 87 and around 50 K. From those studies, the coexistence of spin-glass (SG), ferromagnetic (FM), and AF states at low temperatures has been suggested.[4]

For the present studies, we have prepared a set of the compounds $U(Pd_{1-x}Fe_x)_2Ge_2$, where the Fe was substituted for a part of the Pd in steps of $\Delta x=0.1$. The end member of this series UFe_2Ge_2 also has the $ThCr_2Si_2$-type crystal structure and down to 4.2 K it is a simple Pauli paramagnet.[5,6] ^{57}Fe Mössbauer spectroscopy measurements[5] have proved that the Fe atoms possess no magnetic moments, and Fe occupies only one type of site, i.e., there is no site exchange of the Fe and Ge atoms. However, our preliminary measurements revealed drastic changes in the magnetic properties of UPd_2Ge_2 even at a relatively low level of Fe doping. In particular, the two magnetic anomalies present in UPd_2Ge_2 at 87 and 50 K, disappear completely when $x=0.1$.

In order to investigate these changes in the magnetic behavior on a finer scale, we have prepared a new set of $U(Pd_{1-x}Fe_x)_2Ge_2$ samples in the range $0.0\le x\le 0.1$ with the step $\Delta x=0.01$ ten times smaller. At low iron doping we found a very sharp peak in the susceptibility at around 85 K, whose intensity strongly depends on the doping level with a maximum at $x=0.02$. The maximum of susceptibility at around 50 K on the other hand, rapidly decreases in the $0.00<x<0.02$ region.

In this paper we present detailed measurements of susceptibility and magnetization in the $U(Pd_{1-x}Fe_x)_2Ge_2$ compounds for the range $0.0\le x\le 0.1$. The data for the range $0.1\le x\le 0.9$ will be submitted later.

II. EXPERIMENT

Polycrystalline $U(Pd_{1-x}Fe_x)_2Ge_2$ samples with $x=0.00$–0.20 were prepared by melting of stoichiometric amounts of the constituent materials in an argon arc furnace. The purities of the materials were U 99.95% and Pd, Fe, and Ge 99.9999%. To improve homogeneity, the resulting metallic buttons were remelted several times after turning them over. During melting the overall weight loss was less than 1%. Then all samples were sealed in a quartz tube and annealed at 800°C for 7 days.

X-ray measurement, performed on a Regaku D/Max III x-ray diffractometer, showed that the crystal structure of all $U(Pd_{1-x}Fe_x)_2Ge_2$ compounds with $x=0.01$–0.20 is of the $ThCr_2Si_2$ type with space group $I4/mmm$. The materials are pure single phases. The crystal lattice parameters a and c, the unit cell volume V, and calculated interatomic distances $d_{U\text{-}U}$ for all samples are given in Table I. We believe that upon substitution Fe atoms locate at the Pd atoms sites.

The magnetic susceptibility and magnetization measurements were performed in a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-5 type) in the temperature range 5–300 K and in an applied magnetic field H of up to 55 kG. In all measurements the samples were first cooled down to a specified temperature in zero field, then an external magnetic field was applied, and all data were taken at each step of the temperature and field variations.

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TABLE I. The crystal lattice parameters a and c and magnetic transition temperatures T_N, T_{m1}, and T_{m2} in the $U(Pd_{1-x}Fe_x)_2Ge_2$ system. V is the unit-cell volume and $d_{U\text{-}U}$ is the interatomic distance between the nearest two U atoms. Typical errors are ± 0.005 for a and ± 0.010 for c.

x	a (Å)	c (Å)	V (Å^3)	$d_{U\text{-}U}$ (Å)	T_N (K)	T_{m1} (K)	T_{m2} (K)
0.00	4.200	10.230	180.457	5.915	140	87	50
0.01	4.190	10.192	178.903	5.895	135	92	50
0.02	4.190	10.192	178.903	5.895	135	74	50
0.03	4.190	10.192	178.903	5.895	135	81	50
0.04	4.185	10.229	179.161	5.909	135	88	50
0.05	4.181	10.221	178.671	5.904	135	95	47
0.06	4.178	10.214	178.290	5.900	135	102	44
0.07	4.178	10.214	178.290	5.900	130	107	41
0.08	4.175	10.206	177.857	5.895	130	112	
0.09	4.174	10.206	177.858	5.895	130	115	
0.10	4.174	10.206	177.858	5.895	130	115	
0.20	4.156	10.212	176.399	5.891	130		

III. RESULTS AND ANALYSIS

The x-ray studies showed an unusual anomaly in the x dependence of the lattice parameters c and $d_{U\text{-}U}$ [Figs. 1(a) and 1(b)]. As the iron concentration increases, c and $d_{U\text{-}U}$ decrease at low x and have minimum values at around $x=0.02$. As x increases further, c and $d_{U\text{-}U}$ values return to the initial values, and then decrease gradually.

Figures 2(a) and 2(b) show the temperature dependence of the magnetic susceptibility of $U(Pd_{1-x}Fe_x)_2Ge_2$ measured in the applied field $H=2$ kG. Three peaks are observed in the $\chi(T)$ curves for $x=0.00$–0.09. The temperature of the peaks are listed in Table I, and the x dependence of these magnetic transition points is plotted in Fig. 3(a). The position of the high-temperature peak at T_N changes from 140 to 130 K with change in iron doping from $x=0.00$ to 0.2. The peaks at around 74–115 K (the T_{m1} transition point) are narrow and their typical width at half maximum is 8 K. The peaks at around 50 K (the T_{m2} transition point) are broad. Both T_{m1} and T_{m2} vary with the iron concentration [Fig. 3(a)], with T_{m1} having a minimum value of 74 K at $x=0.02$.

Figure 3(b) shows the x dependence of the peak intensity at the T_{m1} and T_{m2} transition points. The T_{m1} peak intensity has a sharp maximum at $x=0.02$, whereas the intensity of the T_{m2} peak decreases rapidly as x approaches 0.02. Both peaks are hardly visible at $x=0.09$, and the small remnant of the T_{m1} peak disappears at $x=0.2$ [Fig. 2(b)]. Figure 2(b) also shows that at $x \geq 0.07$ the magnetic susceptibility increases rapidly as temperature decreases in the region far below T_{m2}.

At low iron concentration the behavior of the peak susceptibility at the T_{m1} point is correlated with the behavior of the crystal lattice parameters c and $d_{U\text{-}U}$. The maximum of the peak intensity and the minimum in the T_{m1} value at $x=0.02$ correspond to the minimum of the c and $d_{U\text{-}U}$ parameters.

To study the nature of these peaks, we also measured their field dependence in the sample with $x=0.05$ (Fig. 4). At low field H the intensity increases with H for both the T_{m1} and T_{m2} peaks. However, a higher field suppresses the peaks, but in different ways: the T_{m2} peak disappears at $H \geq 20$ kG, whereas the T_{m1} peak intensity decreases with increasing field but still exists even at $H=50$ kG (Fig. 4).

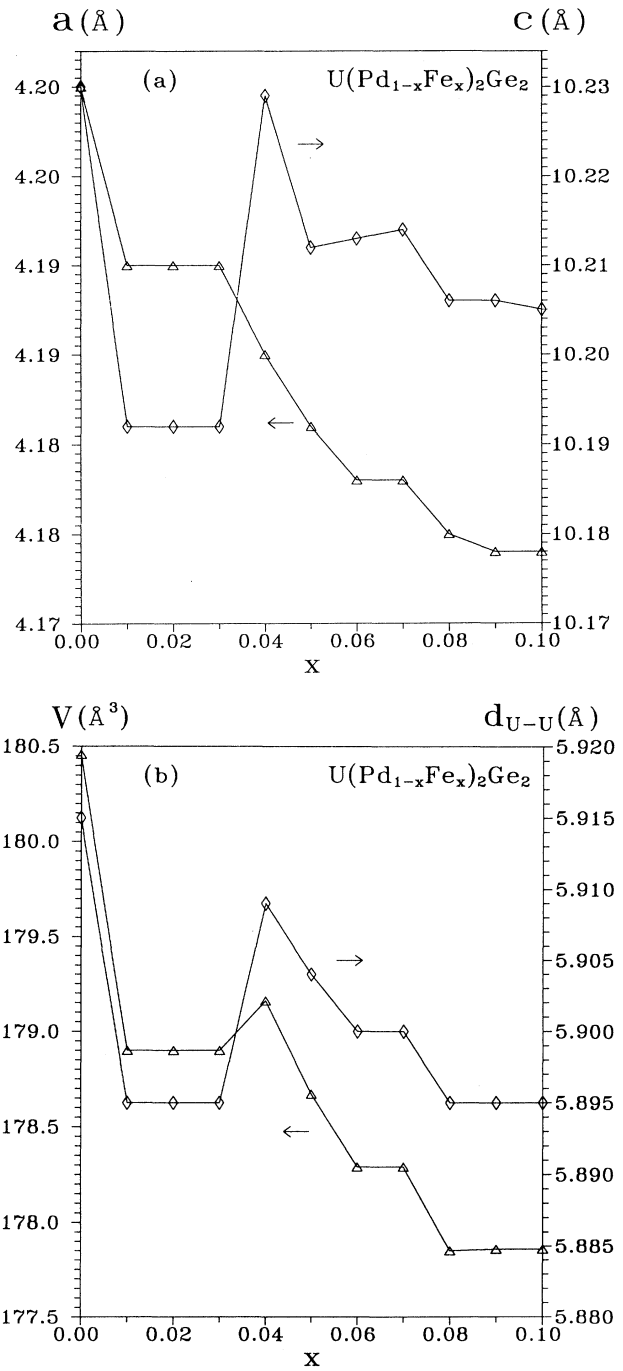


FIG. 1. The x dependence (a) of the crystal lattice parameters a and c, and (b) of the unit cell volume V and the calculated interatomic distance $d_{U\text{-}U}$ between the nearest U atoms.

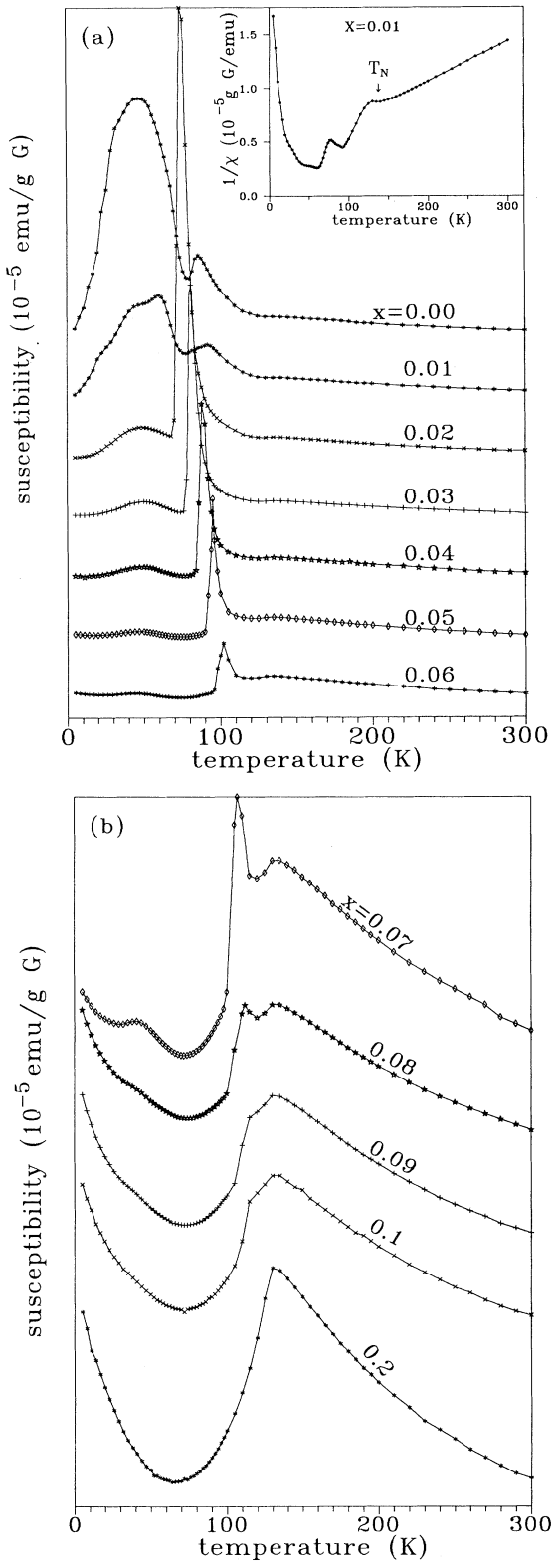


FIG. 2. The temperature dependence of the magnetic susceptibility in $U(Pd_{1-x}Fe_x)_2Ge_2$ measured in an applied field of 2 kG: (a) for $x=0.0$–0.06 and (b) for $x=0.07$–0.2. The inset in (a) shows the temperature dependence of the inverse magnetic susceptibility for $x=0.01$.

Figures 5–8 show the magnetization curves measured at several temperatures for $x=0.00$–0.09.

The low-temperature region. $T<T_{m2}$. We found that at 5 K the hysteresis loops for $x=0.00$ and 0.01 show ferromagnetic behavior, but the initial magnetization curve is not normal [Fig. 5(a)]. At low fields it is weakly dependent on the field and the curve is linear up to a certain field H_a ($H_a\approx 5$ and ≈ 10 kG for the $x=0.00$ and 0.01 samples, respectively). Then it drastically increases and saturates in higher fields. Such behavior is typical of metamagnetism and implies a spin-flip process induced by an applied field in an antiferromagnetic system.[7] In the susceptibility curves of Fig. 2(a), the FM component is not visible at low temperature. This shows that the AF

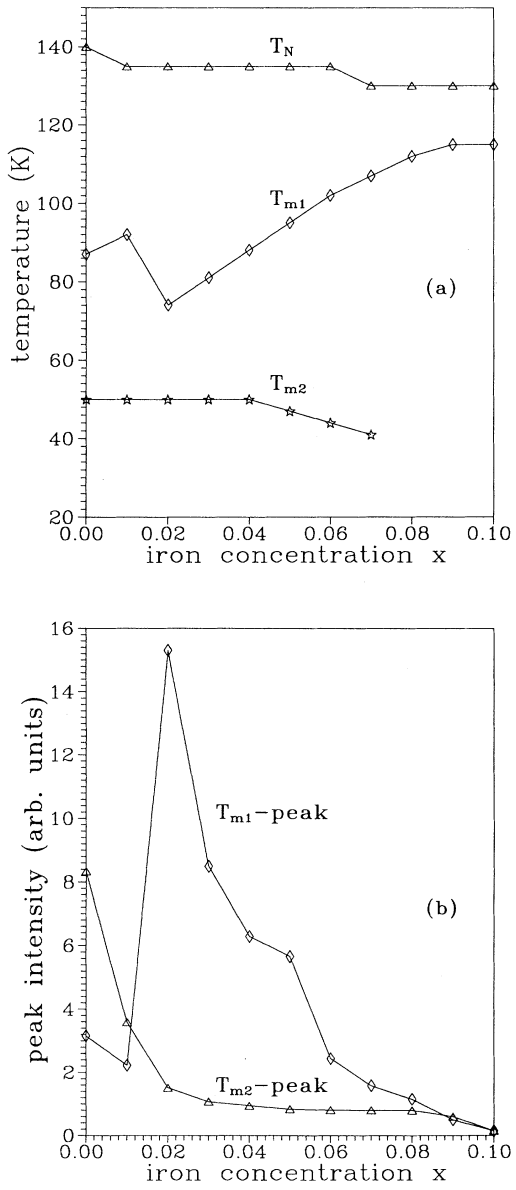


FIG. 3. The x dependence (a) of the magnetic transition temperatures T_N, T_{m1}, and T_{m2} and (b) of the susceptibility peak intensities at the T_{m1} and T_{m2} points in $U(Pd_{1-x}Fe_x)_2Ge_2$.

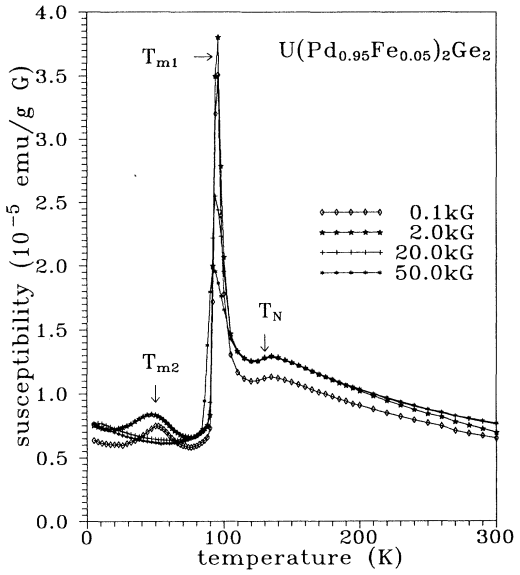


FIG. 4. The temperature dependence of the magnetic susceptibility of $U(Pd_{0.95}Fe_{0.05})_2Ge_2$ in different applied fields $H=0.1$, 2, 20, and 50 kG.

phase is dominant.

For $x \geq 0.02$, the hysteresis loop changes drastically [Fig. 5(b)] and the magnetization becomes small. At 5 K the initial curve of magnetization is linear, as it is in an AF material, but in the cyclic loop irreversibility appears, as if a very small FM component is still induced by the field. The FM component disappears rapidly with further iron doping. It is hardly visible even at $x=0.02$ and it is absent at $x=0.08$ [Fig. 5(b)]. The hysteresis area decreases as x increases. For $x=0.09$, the magnetization curve is composed of two linear and reversible parts, and the slopes are different in the $|H|<35$ kG and $|H|>35$ kG regions. This indicates that the iron-doped compounds become purely antiferromagnetic at low temperature. These effects can be related to changes in the magnetic anisotropy of the crystal UPd_2Ge_2. It seems that a small amount of iron doping increases the anisotropy greatly, and FM ordering, easily enhanced by an applied field in pure UPd_2Ge_2, cannot be induced by the field in the iron-doped compounds. The $x=0.02$ value is the critical concentration x_c at which many parameters change drastically.

The T_{m2} temperature region. Figure 6 shows the field dependence of the magnetization for the $x=0.01$ sample at $T_{m2}=50$ K. The curve exhibits some ferromagnetic-like behavior but it is different from the hysteresis loop at 5 K of Fig. 5(a). The magnetization is not saturated, the remanence is small, and the initial magnetization curve crosses that of the second increasing field after cycling to ± 50 kG, which is the signature of a spin glass.

For the $x=0.02$ sample, which has the critical parameters, we have measured the hysteresis loops at the transition temperatures $T_{m1}=74$ and $T_{m2}=50$ K and at $T=5$ K (Fig. 7). At all these temperatures the hysteresis curves are unusual. At $T_{m2}=50$ K the initial magnetization curve is linear and reversible up to the field $H \approx 10$ kG. With further field increase the magnetization increases suddenly, and irreversibility appears at higher fields. The initial magnetization curve (curve 1 in Fig. 7) is outside the hysteresis loop. The rise of magnetization at $H > H_a$ can be explained by spin flopping in an AF system, and H_a corresponds to an anisotropy field. Similar hysteresis curves have been observed by Senoussi[8,9] in $Ni_{79}Mn_{21}$ and $Au_{81}Fe_{19}$ alloys, where SG and FM order-

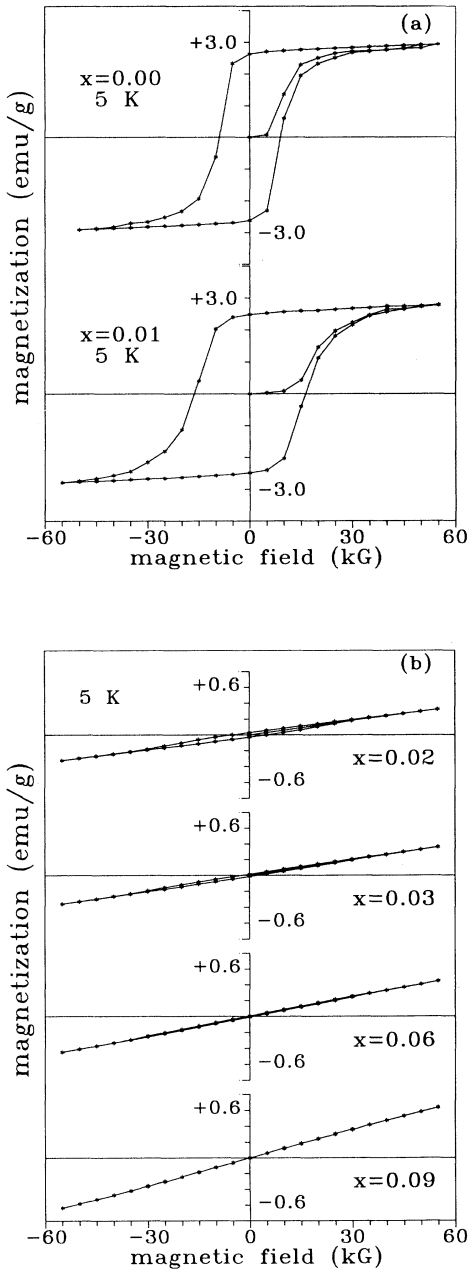


FIG. 5. The low-temperature magnetization curves of $U(Pd_{1-x}Fe_x)_2Ge_2$ (a) for $x=0.00$ and 0.01 and (b) for $x=0.02$–0.09.

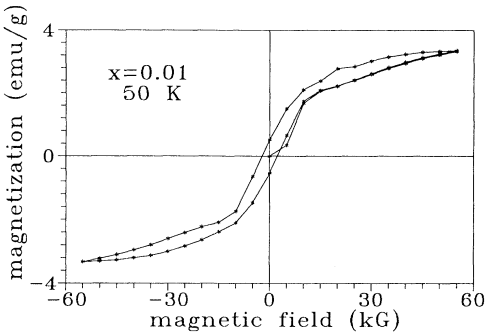


FIG. 6. The field dependence of the magnetization in $U(Pd_{0.99}Fe_{0.01})_2Ge_2$ at 50 K.

ing coexists. The specific features of the hysteresis curves, obtained under various zero-field-cooled (ZFC) and field-cooled (FC) conditions, were explained in terms of anisotropy effects.

Thus, in the T_{m2} temperature region, the AF, FM, and SG states coexist in both pure and iron-doped UPd_2Ge_2 compounds. With higher iron concentration the FM component becomes smaller and disappears at around $x=0.10$ (in a field of 2 kG). We note that this component can be strongly enhanced by an applied field in pure UPd_2Ge_2, and it is suppressed by a field of about 20 kG in the iron-doped material.

The T_{m1} temperature region. The magnetization curves at $T\approx T_{m1}=74$ K (Fig. 7) also exhibit SG-like behavior. The area of the hysteresis loop is largest in the $x=0.02$ sample, where the T_{m1} susceptibility peak intensity is highest [Fig 2(a)]. The magnetization curve at T_{m1} is irreversible but it has no remanence and no coercitivity. In addition, the initial magnetization curve (curve 1 in Fig. 7) is completely outside the hysteresis loop. It is interesting to note that the hysteresis loop at 74 K may be considered as similar to that at 50 K, if one takes $H_a\approx 0$.

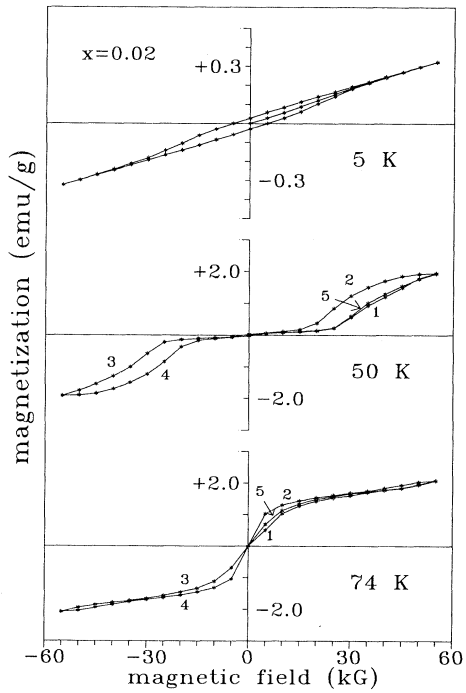
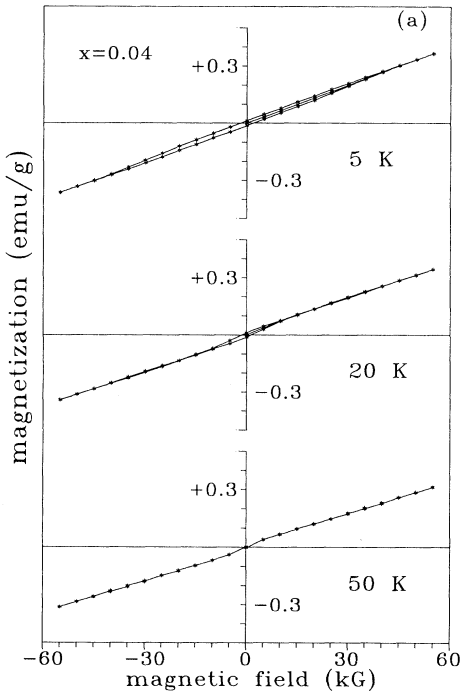


FIG. 7. The magnetization curves of $U(Pd_{0.98}Fe_{0.02})_2Ge_2$ at different temperatures.



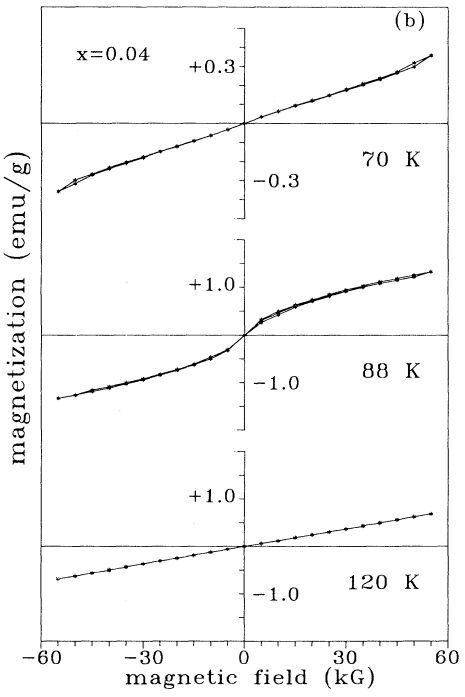


FIG. 8. The magnetization curves of $U(Pd_{0.96}Fe_{0.04})_2Ge_2$ temperatures (a) 5, 20, and 50 K and (b) 70, 88, and 120 K.

This means that anisotropy is very weak at the T_{m1} point. Thus, in the T_{m1} region AF order coexists with SG order; this is most pronounced in the sample with the critical iron concentration $x_c = 0.02$.

Thus, in the $T_{m2} < T < T_{m1}$ range, the pure and doped compounds exhibit the coexistence of AF and SG order with a field-induced FM component. At $T < T_{m2}$, the SG component vanishes as temperature decreases and AF order dominates. In the $T_{m1} < T < T_N$ range, the pure and doped compounds exhibit AF order without coexisting FM and SG phases.

Figures 8(a) and 8(b) show the magnetization curves for the $x = 0.04$ sample at several temperatures, including $T_{m1} = 88$ K and $T_{m2} = 50$ K. The shapes of the curves are similar to those in the $x = 0.02$ sample at the corresponding temperatures, but the hysteresis area is considerably decreased. At $T_{m1} = 88$ K, the hysteresis loop is also typical of a SG, but with much smaller area of the loop as compared with the $x = 0.02$ sample. At $T_{m2} = 50$ K, the curve is typical of the AF state, but at 70 K it still shows SG properties. The $x \geq 0.04$ samples mainly exhibit AF behavior at $T < 50$ K and at $T > T_{m1}$, and coexistence of the AF and SG states at $T_{m2} < T < T_{m1}$. For the higher iron concentrations, $x = 0.1$ and 0.2, the magnetization curves at $T < T_N$ are typical of antiferromagnets.

IV. DISCUSSION

Duh *et al.*[4] studied the nature of the T_{m1} peak in pure UPd_2Ge_2. The hysteresis curves, ZFC and FC magnetic moments, and magnetoresistance provided strong evidence for a spin-glass transition at this temperature. A similar sharp peak of the magnetization has recently been found at 90 K in UPd_2Si_2.[10,11] Neutron diffraction showed that this peak was accompanied by a maximum intensity of the magnetic Bragg peaks from a small component of the commensurate LSWD structure. This component appears at the boundary of the transition of the high-temperature incommensurate LSWD structure to the low-temperature simple AF structure at around 108 K. An anomaly in the magnetization induced by an applied field was also found in UNi_2Si_2 at 103 K by Rebelsky *et al.*[12] It follows from the neutron diffraction that the applied field induces a commensurate LSDW phase in this temperature region. The temperature of these anomalies is again around the transition temperature, $T \simeq 103$ K, from the high-temperature incommensurate LSDW phase to the simple AF phase. Thus this magnetic anomaly appears at the boundary of three phases, incommensurate LSDW, commensurate LSDW, and simple AF ("the triple point" in the magnetic phase diagram of Ref. 12).

We suggest that the peak of susceptibility in $U(Pd_{1-x}Fe_x)_2Ge_2$ at T_{m1} also relates to the appearance of a new magnetic phase. However, the present studies show that at $T < T_{m1}$ this material is not a simple AF, as in the case of UPd_2Si_2 and UNi_2Si_2, since it shows clear SG properties. Competition between different phases in the T_{m1} region probably introduces frustration into the magnetic order and the SG state appears. A small concentration of Fe impurity in UPd_2Ge_2 enhances the magnetic disorder, which becomes most pronounced at $x = 0.02$. It seems that development of the SG structure in the region $T_{m2} < T < T_{m1}$ is governed by the temperature dependence of the anisotropy energy. Thus in pure and doped UPd_2Ge_2 there are at least three temperature ranges, $T < T_{m2}$, $T_{m2} < T < T_{m1}$, and $T_{m1} < T < T_N$, where different magnetic phases appear. Our data do not support the results of Ref. 1, where only one magnetic phase was found in UPd_2Ge_2 at $T < 140$ K. Detailed neutron-diffraction studies of the temperature variations of the magnetic structure in UPd_2Ge_2 are necessary.

On the other hand, the peak of susceptibility at the T_{m1} point is very sharp, which is not usual for conventional spin glasses. Here we might note that at low iron concentration the $U(Pd_{1-x}Fe_x)_2Ge_2$ system possesses some features expected of a Stoner spin glass.[13,14] Stoner-type spin-glass ordering in a disordered itinerant-electron system with a very high Kondo temperature ($T_K \gg T_{SG}$) has been suggested for some binary alloys (e.g., Rh-Fe, Rh-Co), where both host and impurity are transition metals. UFe_2Ge_2 is known to be a Pauli paramagnet down to 4.2 K,[5] and our studies showed that the substitution of Fe for Pd in UPd_2Ge_2 introduces itinerant character into the high-temperature paramagnetism. In the Stoner SG approximation the local spin fluctuations at Fe impurity sites can be coupled via the magnetic response of the host-matrix electrons. When this interaction is random, a frozen-spin state characterized by an Edwards-Anderson spin-glass order parameter may occur.

V. CONCLUSION

In compounds like UPd_2Ge_2 and UPd_2Si_2, the nature of the magnetic phases is governed by the temperature dependence of the anisotropy energy. In regions of coexistence of different phases, a spin-glass state appears as a result of competition between the coexisting phases. Small iron doping decreases the c lattice parameter, increases the anisotropy energy, and brings an itinerant character into the electronic system. This may result in a Stoner spin-glass condensation.

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