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Spin-glass-like behaviour in UPd_2Ge_2

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Abstract. The magnetic susceptibility, hysteresis curves, ZFC and FC magnetic moments and magnetoresistance measurements indicate the appearance of a spin glass state in UPd_2Ge_2 below 87 K, and the existence of a ferromagnetic phase below 80 K. From these studies and taking into account the neutron diffraction data, a mixed spin glass–ferro–antiferromagnetic state is suggested in the low-temperature region.

1. Introduction

The uranium ternary intermetallic compound UPd_2Ge_2 has the $ThCr_2Si_2$ type crystal structure and shows interesting features in its magnetic behaviour [1–3]. The neutron diffraction investigation [1] has shown that below 140 K the compound becomes a collinear antiferromagnet, but the magnetic ordering has an oscillatory character. The magnetic properties can be explained by an incommensurate magnetic structure with a modulated amplitude of the magnetic moments. Tien *et al* [2] have found that besides the phase transition at 140 K, the magnetic state of UPd_2Ge_2 changes near 70 K and 50 K. They also suggested the coexistence of ferromagnetic and antiferromagnetic states in this material and the appearance of ferromagnetism at low temperature. Duh *et al* [3] suggested that the peak at 80 K in the $1/\chi = f(T)$ curves corresponds to the appearance of the ferromagnetic phase. In addition, they observed an anomaly at 20 K and classified the magnetic states below the Néel temperature into three different regions. It was also found that the magnetic structure of UPd_2Ge_2 is strongly modified by the applied field [3]. To study the abnormal magnetic properties of UPd_2Ge_2, in this report we present the measurements of magnetic susceptibility, hysteresis curves, ZFC and FC magnetic moments and magnetoresistance. Evidence for the spin glass behaviour is found at temperatures below 87 K.

2. Experimental details

The method of preparation of the UPd_2Ge_2 samples and x-ray analysis were described in detail in [3]. The susceptibility and magnetization measurements were performed in a

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SQUID magnetometer (Quantum Design MPMS-5 type) in the temperature range 4.2–300 K and in an applied magnetic field $H_{\rm ext}$ up to 55 kG. The longitudinal magnetoresistance measurements were carried out at 70 K in fields up to 50 kG generated by the Oxford superconducting magnet. A standard four-probe DC technique was used for the magnetoresistance measurements. The measurements were automated using an IBM compatible PC/AT through an IEEE-488 interface.

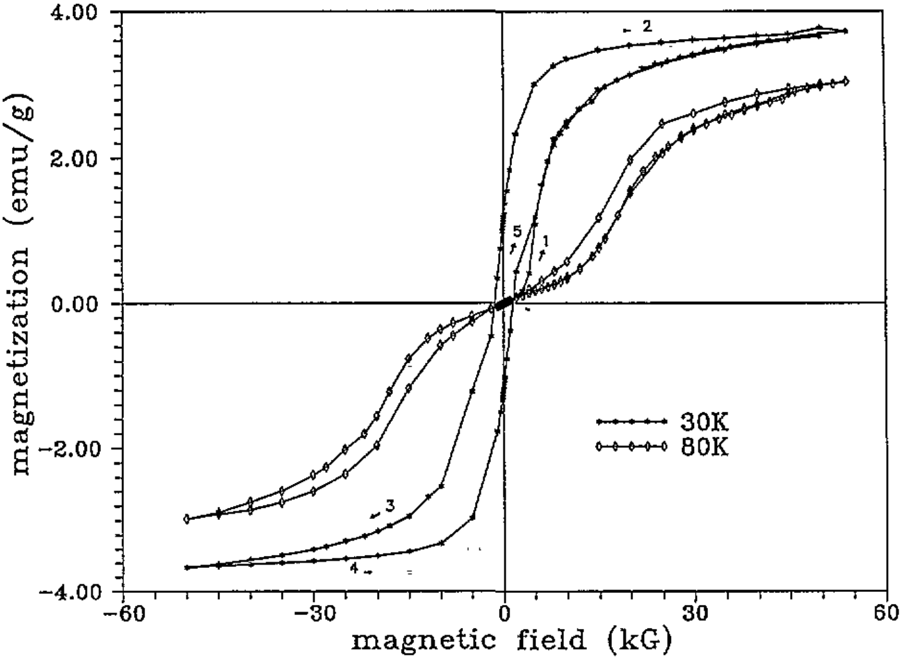

Figure 1. The magnetization curves of UPd_2Ge_2 at 30 K and 80 K. Curves 1, 2, 3, 4 and 5 correspond to different stages of the magnetization. Curve 1 corresponds to the initial stage of magnetization and 2, 3, 4 and 5 are the cyclic loop.

3. Results and discussions

Figure 1 shows the hysteresis loops in UPd_2Ge_2 at 30 K and 80 K. The curves are very similar to those observed in the concentrated spin glass (SG) system $Ni_{79}Mn_{21}$ [4]. The hysteresis curve for 30 K in figure 1 shows different paths for two 0–50 kG increasing field periods, and the later one crosses the former one at a certain field. The initial curve (curve 1 in figure 1) is outside the cyclic loop. This behaviour is different from that typical of ferromagnets. In the magnetization curve we also found a sharp change at ±1 kG field, which shows that almost the entire remanence is reversed in a narrow range of values of field. Similar behaviour has been observed in the SG system $Au_{81}Fe_{19}$ [4]. Theoretical analysis of various SG models [5, 6], based on mean field calculations suggested that a ferromagnetic short-range order is responsible for such sharp magnetization reversals. This particularly

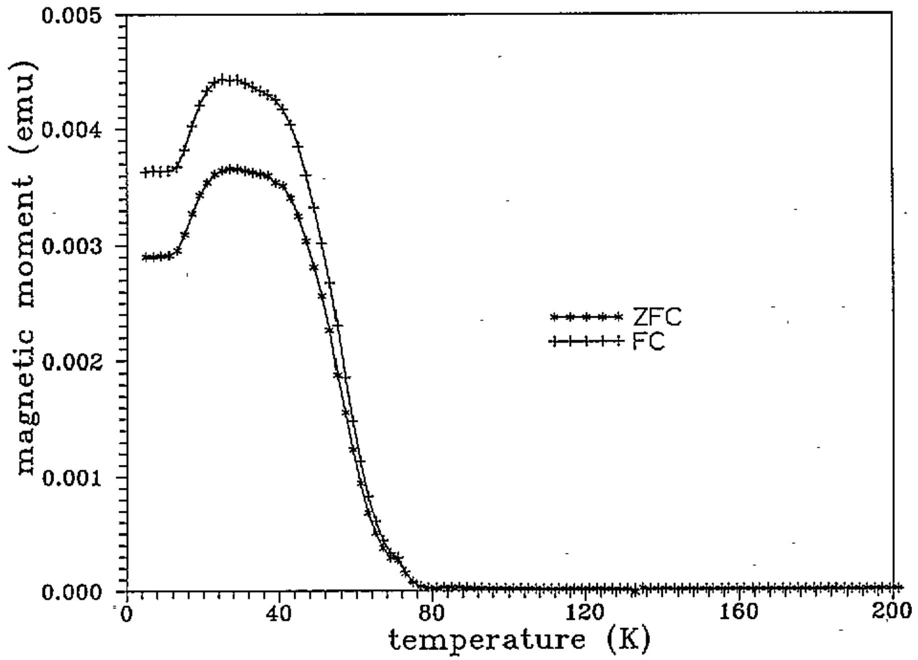

Figure 2. The temperature dependences of magnetic moments in UPd$_2$Ge$_2$ for ZFC and FC regimes in the field $H_{\text{ext}} = 50$ G.

complicated behaviour may occur in systems where SG and ferromagnetic ordering compete and the states with mixed ordering can be produced.

Figure 2 shows the temperature dependence of magnetic moments for ZFC and FC regimes in the field $H_{\text{ext}} = 50$ G. It is clearly seen in figure 2 that a ferromagnetic phase appears at $T = 80$ K. However, the magnetization curves for ZFC and FC regimes are split below 80 K and magnetic moments for FC are higher than for ZFC. This behaviour can also be explained by the SG effect and it was predicted theoretically in [6] for the Ising type of SG and for the Heisenberg type with large anisotropy. Since the moments in an SG will be frozen at a temperature below the freezing temperature T_{SG}, the field applied before cooling down will line up some magnetic moments. However, in classical concentrated SGs (with a ferromagnetic component), the FC and ZFC magnetizations are split at roughly the temperature at which the ZFC magnetization has a maximum, but the shape of the ZFC curves depends on the value of anisotropy and on the exchange interaction parameter J_0 [5, 6]. We suppose that in our case the FC and ZFC curves are different from the classical SGs because of the presence of the antiferromagnetic phase along with the SG and ferromagnetic states. Maybe the s–f exchange interaction in UPd$_2$Ge$_2$ (in contrast to the s–d interaction in classical SGs) introduces its own specificity.

SGs are magnetic systems in which the exchange interactions are randomly distributed in sign and magnetic moments are frozen over a macroscopic scale at temperatures below the freezing temperature T_{SG}. As an accompaniment to spin freezing one observes a cusp in the magnetic susceptibility at T_{SG} and hysteresis and remanent magnetization below the freezing temperature.

Figure 3 shows the temperature dependences of the susceptibility of UPd$_2$Ge$_2$ under different external magnetic fields H_{ext}. An abnormal phenomenon was observed below

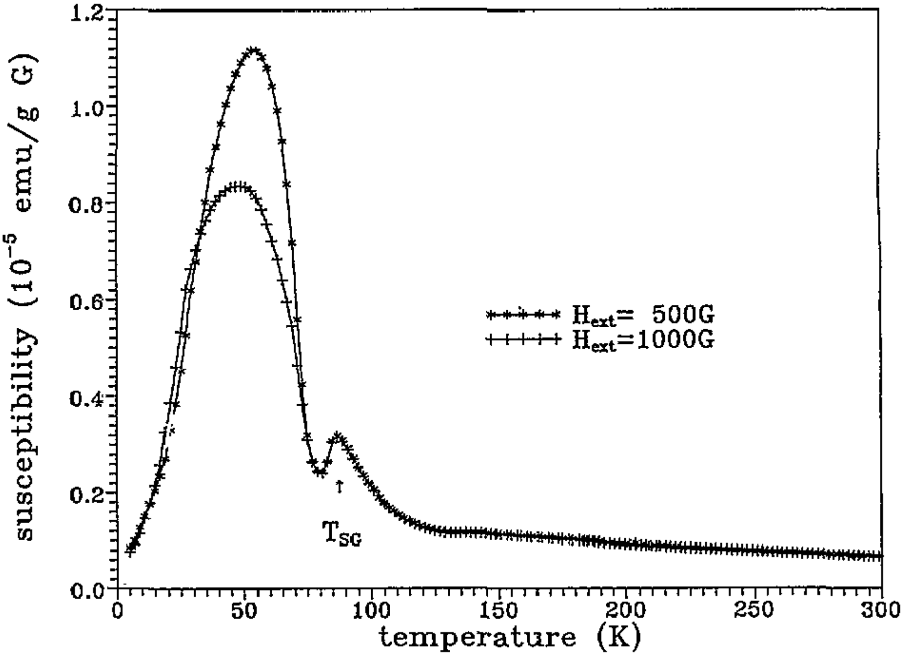

Figure 3. The temperature dependences of the magnetic susceptibility in UPd_2Ge_2 for $H_{ext} = 500$ G and 1000 G.

140 K. Near 87 K a small cusp appears in the $\chi = f(T)$ curves and maximum susceptibility is observed around 50 K. The position of the cusp at 87 K does not change with different applied field, but the maximum around 50 K shifts at different applied fields. According to the phenomenological theory of SGs [7], in zero or low fields a weak singularity of susceptibility (an upward cusp) appears near the transition point T_{SG}, and with decreasing temperature, the SG phase normally appears first and the ferromagnetic phase appears at lower temperature, i.e. $T_{SG} > T_C$. It was found in our previous studies [3] that a ferromagnetic phase appeared at 80 K and ferromagnetic and antiferromagnetic states coexisted in the 20–80 K region. Perhaps we can suggest that the cusp in the $\chi = f(T)$ curves of figure 3 appearing at 87 K in UPd_2Ge_2 is the SG transition point.

The SG character can also be observed in the behaviour of the magnetoresistance [8, 9]. Figure 4 shows the field dependence of magnetoresistance $\Delta\rho/\rho$ of UPd_2Ge_2 at 70 K. The $\Delta\rho/\rho$ is negative and it exhibits a strong hysteresis effect as has also been seen in the magnetization measurements. At $H_{ext} \sim 10$ kG, the $\Delta\rho/\rho$ values drop sharply. Nigam *et al* [8] shows that such behaviour of the magnetoresistance is the indication for a metamagnetic transition due to applied field. The result of figure 4 suggests that a field higher than 10 kG will turn the antiferromagnetic or SG state to the ferromagnetic state. This is consistent with the result of our previous study [3], in which it was found that a strong applied field induces the ferromagnetic phase in this material. From the field dependence of $\Delta\rho/\rho$ (see figure 4) we also found that magnetoresistance is negative for all fields, and at low fields, before the metamagnetic transition, $\Delta\rho/\rho$ is proportional to H^n where $n > 1$. The theory of magnetoresistance for the canonical SGs (such as AuFe, AuMn, CuMn, AgMn and AuCr) based on an Edwards–Anderson type model [9], when the local spins interact through the conduction electrons via the s–d exchange coupling, predicts $\Delta\rho < 0$ and $\Delta\rho/\rho \propto H^n$ with

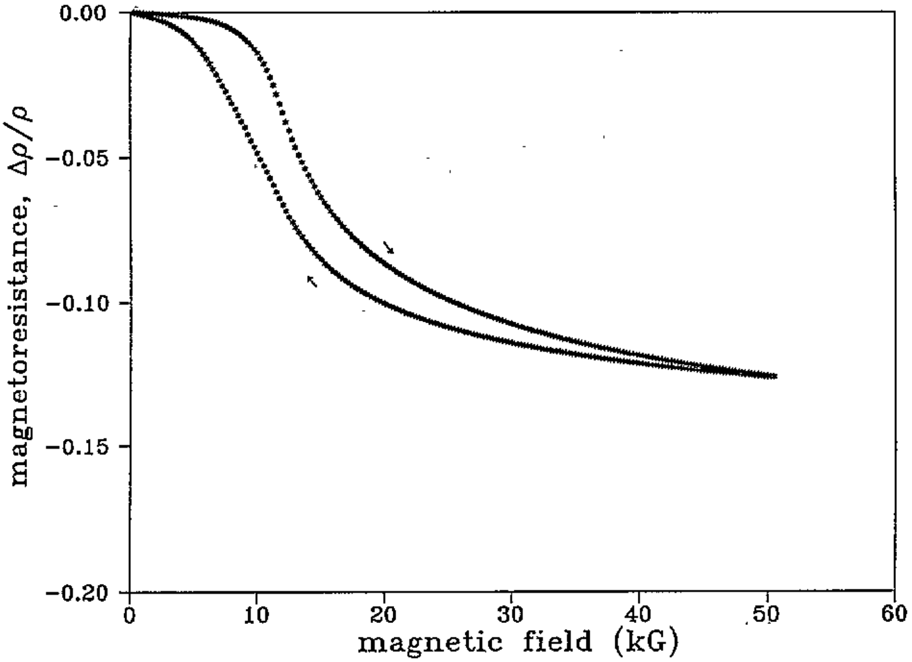

Figure 4. The field dependence of the magnetoresistance in UPd_2Ge_2 at 70 K.

$n > 1$. Thus, the above features in the behaviour of the magnetoresistance in UPd_2Ge_2 are due to the SG state. The field dependence of magnetoresistance in Upd_2Ge_2 observed in our measurements is similar to that for the SG systems AuFe [9] and UCu_2Ge_2 [8].

In conclusion, the field dependence of the magnetization and the temperature dependence of FC and ZFC magnetic moments, as well as the susceptibility and magnetoresistance behaviour, indicate the existence of an SG state in UPd_2Ge_2 below 87 K, and the appearance of a ferromagnetic phase below 80 K. Thus, the mixed SG–ferromagnetic state is present at $T < 80$ K. Taking into account the neutron diffraction data [1], which have found the antiferromagnetic state in UPd_2Ge_2 at 4.2 K and at 80 K, and the results of our previous studies [3], we suggest the presence of a mixed SG–ferro–antiferromagnetic state in this temperature region.

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