

# Effect of randomness on anomalous Hall coefficient in antiferromagnet $U_2PdGa_3$

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The Hall effect has been studied in antiferromagnet  $U_2PdGa_3$ . In the paramagnetic state, the Hall data can be interpreted in terms of skew scattering theory for heavy fermion systems. We observe a large contribution of anomalous Hall coefficient, indicating a dominating contribution of incoherent skew scattering by uranium 5f moments. An interesting behaviour of the  $R_H(T)$  dependence is found at low temperatures: the  $R_H(T)$  curve displays a plateau between  $T^* = 12$  K and  $T_N = 33$  K, followed by a shoulder upon further decreasing temperature. The appearance of the anomaly around  $T^*$  suggests the existence of a new scattering mechanism, in addition to well known skew and jump-side scatterings. We compare the Hall data of  $U_2PdGa_3$  with those observed for strongly correlated electron systems and with those of spin-glasses. Like the latter systems, non-zero spin chirality seems to play a considerable role in the anomalous Hall effect of  $U_2PdGa_3$ .

Key words:  $U_2PdGa_3$ ; Hall effect; strongly correlated electron systems; randomness; spin chirality

## 1. Introduction

$U_2PdGa_3$  is an orthorhombic, collinear antiferromagnet with the Néel temperature  $T_N \approx 33$  K [1, 2]. In this compound, there is a competition between the Kondo effect and randomness for long-range antiferromagnetism. The Kondo effect manifests itself in electrical resistivity, magnetoresistance properties at high temperatures and causes an enhancement in the electronic heat capacity at low temperatures ( $\gamma_0 = 72$  mJ/(mol·K<sup>2</sup>). Furthermore, like in many other compounds with randomness, the specific heat of  $U_2PdGa_3$  does not show a mean-field discontinuity at the Néel temperature but, instead, the dc magnetic susceptibility exhibits magnetic history phenomena. As a result, the antiferromagnetic state with the ordered moments of about  $0.3 \mu_B$  exists only with finite magnetic correlation lengths (below 150 Å) [2].

The measurement of the Hall effect is one of the tools of studying scattering mechanisms of conduction electrons on magnetic moments in metals. For ordinary

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magnetic materials, the Hall resistivity  $\rho_H$  measured in a field  $B$  can be approximately described by the equation:

$$\rho_H = R_0 B + 4\pi R_s M \quad (1)$$

where  $R_0$  and  $R_s$  are the ordinary and anomalous Hall coefficients and  $M$  is the magnetization [3, 4]. Usually, the sign and value of  $R_0$  determine the type and carrier concentration, respectively, while the  $R_s$  value and its temperature dependence provide information on scattering mechanisms of the charge carriers.

In the  $f$  electron Kondo lattice systems, the electronic and magnetic properties are sensitive to a change in the coupling strength between  $f$  and conduction electrons. At high temperatures, weak  $f$ - $spd$  hybridization enables  $f$  electrons to be localized ones. On the second hand, at low temperatures the strength of the  $f$ - $spd$  hybridization becomes stronger. Thus, the ground state of more itinerant  $f$  electrons involving heavy mass quasiparticles emerges. According to the theory of Hall effect in heavy-fermion systems [5, 6], the Hall effect is mainly determined by coherent skew scattering at  $T < T_{\text{coh}}$  and incoherent skew scattering for  $T > T_{\text{coh}}$ . The skew scattering contribution  $R_{sk}$  was predicted to be proportional to the product of magnetic susceptibility and magnetic resistivity, i.e.,  $R_{sk} = \gamma \tilde{\chi} \rho$ , where  $\gamma$  is a parameter related to the phase shift  $\delta$  by the dependence  $\gamma = -(5/7)g\mu_B k_B^{-1} \sin\delta \cos\delta$ ,  $\tilde{\chi} = \chi/C$  with  $C$  being the relevant Curie parameter deduced from the susceptibility data and  $\rho$  is the magnetic resistivity [5, 6].

In this contribution we address the question whether the carrier scattering mechanisms inferred from measuring the Hall resistivity comply with those from the magnetization and electrical resistivity measurements.

## 2. Experimental details

Polycrystalline samples of  $\text{U}_2\text{PdGa}_3$  were fabricated by arc-melting in a high-quality pure argon atmosphere. The samples were annealed at 650 °C for one week. Phase purity and composition were checked by EDX and X-ray powder diffraction. dc magnetization measurements were carried out by using a Quantum-Design SQUID magnetometer. Electrical resistivity was measured by an ac conventional four-probe method. The measurements of Hall effect were performed in the temperature range 2–300 K and in magnetic fields up to 7 T, using an ac conventional four-probe technique. Hall voltage was recorded with a low-frequency (37 Hz) excitation current of 10 mA on a 0.5 mm thick sample mounted on a horizontal rotator.

## 3. Results and discussion

Figure 1 shows the temperature dependence of the Hall coefficient measured at 7 T. We notice that  $R_H$  has a positive sign over the entire temperature range of investi-

gation. In the paramagnetic state,  $R_H$  increases with decreasing temperature, resembling the temperature dependence of the magnetization and resistivity reported previously [1]. Since theory of the Hall effect for heavy fermions [5, 6] predicts the following dependence of  $R_H(T)$ :

$$R_H = R_0 + \frac{\gamma\chi\rho}{C} \quad (2)$$

this form was used to fit the data. In the inset of Fig. 1 we plot the experimental  $R_H$  values against the product  $\rho\chi/C$  and the solid line is the result of the fitting for the data in the temperature range 60–300 K. The agreement between the experimental and theoretical data may support the dominance of the incoherent skew scattering, being responsible for the anomalous Hall effect in the paramagnetic state. The constant  $\gamma = 0.042$  K/T, derived from the fit is the same order of magnitude as those found in heavy-fermions like  $CeAl_3$ ,  $CeCu_6$  [6] and  $UCu_5Al$  [7]. From the fit  $R_0$  has also been estimated to be  $-2.28 \pm 0.05 \times 10^{-10} \text{ m}^3/\text{C}$ , which corresponds to a single-band concentration of electron-like carriers  $n = 6.5$  e/f.u.

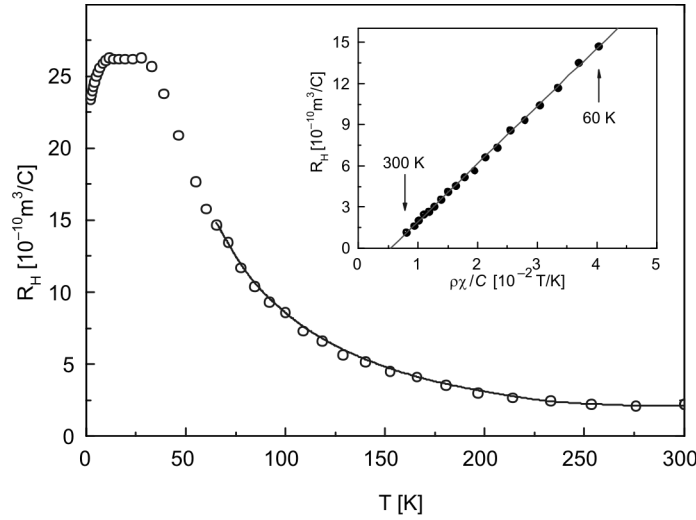


Fig. 1. Temperature dependence of the Hall coefficient of  $U_2PdGa_3$  measured at 7 T. The inset shows the dependence of  $R_H$  on  $\rho\chi/C$  with  $C = 0.58 \text{ cm}^3 \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ . The solid lines present the fits of the experimental data to Eq. (2)

In order to get access to information on scattering mechanisms at low temperatures, we compare the  $R_H$  data with the electrical resistivity  $\rho$  and volume magnetic susceptibility  $\chi_V$  in Fig. 2. As shown in Fig. 2a, below the magnetic phase transition  $T_N$  the Hall coefficient levels off until it approaches  $T^*$  of about 12 K. On further decreasing temperature,  $R_H$  rapidly decreases. In the magnetically ordered state of ordinary magnets, with decreasing temperature the strength of the asymmetric scattering de-

increases continuously due to a continuous decrease of the spin fluctuations, and obviously no distinct anomaly would be expected at temperatures far below magnetic ordering temperature. The fact that a shoulder appears in  $R_H(T)$  curve of  $U_2PdGa_3$  at temperatures near  $T^*$  ( $\sim T_N/2$ ), and the slope of the  $R_H(T)$  curve has different values in the temperature ranges 2–12 K and 16–30 K, is remarkable. One explanation is that in these temperature ranges different mechanisms of scatterings appear. In other words, the strength of the asymmetric scattering changes dramatically when the system goes from one to another temperature range.

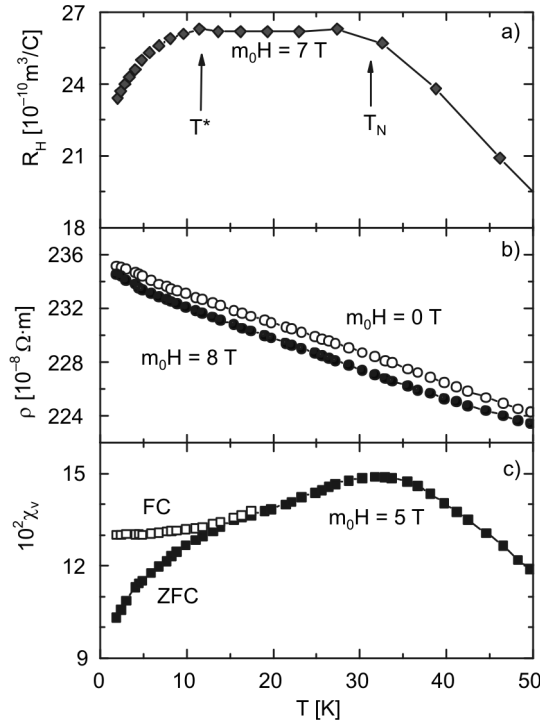


Fig. 2. Temperature dependence of a) the Hall coefficient, b) electrical resistivity at 0 and 8 T, and c) volume susceptibility at 5 T after zero-field cooling and field-cooling in  $U_2PdGa_3$

In contrast to the  $R_H$  behaviour, the resistivity measured at 0 and 8 T (Fig. 2b) does exhibit weak temperature dependence. This fact, together with the lack of a clear singularity at the magnetic ordering temperature, may point to a significant influence of the short-range interactions and/or to the Kondo effect, though the magnetoresistance,  $[\rho(8 \text{ T}, T) - \rho(0 \text{ T}, T)]/\rho(0 \text{ T}, T)$  displays a negative minimum at  $T_N$  [2]. Moreover, since the resistivity has no anomaly at  $T^*$ , it is difficult to ascribe the behaviour of  $R_H$  to the same scattering mechanism as for the resistivity solely. In fact, the magnetic susceptibility taken at 5 T after zero-field cooling and field cooling exhibits an irreversibility point around 15 K and a broad maximum at  $T_N \approx 33$  K (Fig. 2c). These features point to closely correlated behaviour between  $R_H(T)$  and  $\chi_v(T)$ . Previously,

the irreversibility effect observed in the susceptibility curves was interpreted in terms of short-range magnetic interactions dictated by disorder effects [2]. It is accepted that the atomic disorder is favourable for the system to develop magnetic clusters. In terms of the electronic transport properties, magnetic moments belonging to clusters may stand for new scattering centres, in addition to magnetic moments not belonging to any cluster yet. Therefore, the randomness not only breaks the coherent scattering but may also modify the magnitude of the anomalous Hall component which in natural manner depends on centres scattering electrons.

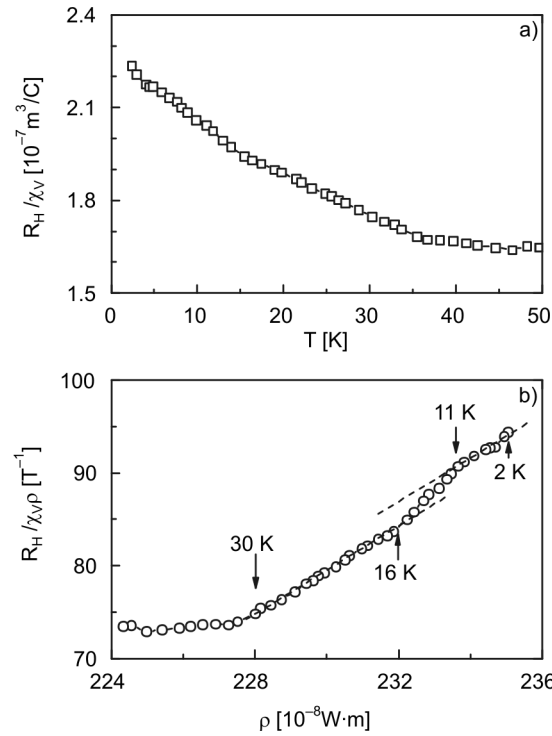


Fig. 3. Temperature dependence of the ratio  $R_H/\chi_V$  (a) and the ratio  $R_H/\chi_V \rho$  vs.  $\rho$  in  $U_2PdGa_3$

If the ordinary component of  $R_H$  is small compared to the anomalous one, the ratio  $R_H/\chi_V$  should approximately represent the anomalous contribution  $R_s$ . When inspecting Fig. 3a, where the plot of the ratio  $R_H/\chi_V$  vs.  $T$  is shown, we see that the ratio  $R_H/\chi_V$ , i.e.,  $\sim R_s$ , increases with decreasing temperatures. A comparison of  $R_H/\chi_V(T)$  with  $\rho(T)$  suggests a contribution of the electrical resistivity to the anomalous Hall coefficient. Usually, the anomalous Hall coefficient in a magnetically ordered state consists of two contributions: the skew scattering proportional to  $\rho$ , and the side-jump scattering proportional to  $\rho^2$  [4]. We have considered the relationship between  $R_H/\chi_V$  and  $\rho$  given by

$$\frac{R_H}{\chi_V} \sim R_s = a\rho + b\rho^2 \quad (3)$$

Parameters  $a$  and  $b$  are the coefficients of the skew and jump-side scattering contributions, respectively. According to Eq. (3), a plot  $R_s/\rho$  versus  $\rho$  should be a straight line, and the coefficients  $a$  and  $b$  can be deduced. In Figure 3b, the ratio  $R_H/\chi_V\rho$  is plotted in function of  $\rho$ . The data indeed approximately lie on a straight line. Interestingly, the slopes of the curve  $R_H/\chi_V\rho$  vs.  $\rho$  (i.e., parameter  $b$ ) in the temperature ranges 2–11 K and 16–30 K are the same, suggesting that there is no change in strength of the side-jump scattering across  $T^*$ , whilst for the skew scattering, the coefficient  $a$  (i.e., the intercept of the plot) is different above and below  $T^*$ . As we have considered above, the formation of magnetic clusters below  $T^*$  should bring in new scattering centres, yielding additional contribution to skew scattering.

Recently, the chirality mechanism [8] has been proposed as an important mechanism for the anomalous Hall effect in spin glass systems [9–12]. Tataru and Kawamura [8] have shown that the uniform chirality  $\chi_0$  contributes to anomalous Hall effect as follows:  $R_s = (a\rho + b\rho^2) + c\chi_0$ . Here,  $c$  is a constant relevant to the detailed band structure of the conduction electrons and a uniform chirality  $\chi_0$  is the sum of the local chirality defined as  $\chi_{ijk} = S_i(S_jS_k)$  for three spins  $S_i$ ,  $S_j$  and  $S_k$ . Clearly, the chirality contribution to  $R_s$  is remarkable when the net uniform chirality is finite, i.e.,  $\chi_0 \neq 0$ . In fact, the contribution from  $\chi_{ijk}$  to  $\chi_0$  decays rapidly as  $\exp(-3r/2l)/(k_F r)^3$ , where  $r$  is the distance between the spins,  $k_F$  is the Fermi wave number, and  $l$  is the mean free path. Thus, one expects a large contribution from  $\chi_{ijk}$  to  $\chi_0$  when the distance between spins  $r$  is shorter than  $l$ . Using the value of  $R_H$  and  $\rho$  at 2 K, one estimates the mean free path to be  $28.6 \times 10^{-10}$  m, being larger than the average distance between the spins of the order of  $3.5 \times 10^{-10}$  m. This means that a number of triangles of three spins may give their contribution to  $\chi_0$  in the studied compound.

#### 4. Conclusions

We have studied the Hall effect in the short-range antiferromagnet  $\text{U}_2\text{PdGa}_3$ . The experimental Hall coefficient was compared to the magnetic susceptibility and electrical resistivity. From the relationship between  $R_H$ ,  $\chi_V$  and  $\rho$ , we infer that the incoherent skew scattering dominates in the paramagnetic state. Anomalies found in the  $R_H(T)$  dependence at both the Néel temperature  $T_N \approx 33$  K and  $T^* = 12$  K provide a new evidence in support that  $\text{U}_2\text{PdGa}_3$  is not a classical long-range antiferromagnet. The development of magnetic clusters below  $T^*$ , and the resulting randomness have been suggested to cause an anomaly in the  $R_H(T)$  curve at  $T^*$ . We considered the observed behaviour to connect with the chiral mechanism of the anomalous Hall effect. We conclude that the low-temperature  $R_H$  data of  $\text{U}_2\text{PdGa}_3$  may be qualitatively under-

stood in the framework of contributions from the skew, jump-side scatterings and from the spin chirality.

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