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Coherence effects in the low-temperature Hall coefficient of the heavy-fermion system UPd_2Al_3

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The temperature dependence of the Hall resistivity of thin epitaxial UPd_2Al_3 films with low defect densities is investigated. It shows a minimum at 6 K for fields up to 12 T. This feature is attributed to the superposition of a coherence-derived low-temperature Hall coefficient and a skew-scattering contribution. Further implications on the Hall effect in the coherence regime of Kondo lattices are discussed.

Heavy-fermion systems differ from diluted Kondo systems due to the regular arrangement of the Kondo ions in a lattice. While the high-temperature properties of Kondo lattices find a conclusive interpretation by simply scaling single-impurity Kondo behavior to the effective concentration of one Kondo ion per unit cell, the low-temperature properties show characteristic new features due to the necessarily coherent ground state of the lattice system.[1] In particular, the onset of coherence of the scattering events on the Kondo-lattice sites below the coherence temperature T^* leads to a decreasing resistance with decreasing temperature. The dynamical and equilibrium properties of Kondo systems are determined through the temperature-dependent evolution of a many-body resonance of the local f states [Abrikosov-Suhl-resonance (ASR) or Kondo resonance] in the vicinity of the Fermi level E_F.[2] This resonance occurs due to an antiferromagnetic coupling of the band-electron spins on the impurity spin. Concerning the transport properties, the Hall coefficent represents a sensitive probe for the details of the scattering mechanism due to its dependence on higher-order transport integrals. Above T^* skew scattering was shown to give a reasonable explanation for the observed variation of the Hall coefficient with temperature. Based on a s-d (s-f) exchange model for the interaction of band electrons with magnetic impurity sites the scattering matrix in momentum representation contains an imaginary orbital-exchange part that changes its sign under time reversal [$i(\mathbf{k}\times\mathbf{k}')\cdot\mathbf{J}$, ($\mathbf{J}$ is the magnetic moment)]. This results in a left-right imbalance of the Hall currents.[3,4] While spin-flip scattering is explicitly excluded in the calculation of the Hall coefficient, the resonant scattering in the $l=3$ channel for f Kondo ions substantially increases the skew-scattering contribution. In the low-temperature regime skew-scattering contributions are reduced due to the growing dominance of small-angle scattering. Since existing calculations base on the single-impurity limit the skew-scattering effect in the coherent regime of heavy-fermion systems still lacks a theoretical understanding (for a recent review see Ref. 5). Nevertheless, in most of the heavy-fermion systems the single-impurity skew-scattering model is able to reproduce, at least qualitatively, the behavior of the Hall coefficient below T^*.[3] In the Fermi-liquid regime strong deviations from a simple skew-scattering mechanism become obvious. Among the heavy-fermion systems a wealth of different low-temperature characteristics in $R_H(T)$ can be found with especially interesting behavior in $CeCu_6$,[6,7] UPt_3,[8,9] and CeNi.[9] These systems show a minimum in $R_H(T)$ well below T^* whose origin is unclear.[5]

In this paper we stress the similarities to the temperature dependence of the Hall coefficient in UPd_2Al_3, that shows an analogous minimum at low temperatures. An explanation of the low-temperature behavior of R_H in UPd_2Al_3 is given based on a contribution due to coherence effects, $R_H^{\text{coh}}(T)$, which is superimposed by a skew-scattering part, $R_H^{\text{skew}}(T)$. The combination of both results in a net Hall coefficient

$$R_H(T)=R_H^0+R_H^{\text{coh}}(T)+R_H^{\text{skew}}(T) \quad . \tag{1}$$

The possible relevance of this ansatz for the Hall coefficient in $CeCu_6$ and other heavy-fermion systems is discussed.

UPd_2Al_3 is a heavy-fermion superconductor with $T_c \simeq 2$ K. Below 14 K the system orders antiferromagnetically.[10] The moments are oriented in the basal plane of the hexagonal lattice.[11] The measurements of the temperature dependence of the Hall coefficient were performed on thin epitaxial films that were deposited onto heated single-crystalline $LaAlO_3$ substrates in (111) orientation by means of a molecular-beam-epitaxy preparation technique. The films grow with the c axis perpendicular to the substrate.[12] X-ray diffraction reveals the single-phase nature of the films. For the transport measurements the films were wet-chemically patterned. Electrical contacts were prepared with copper wires attached to the films using silver paint. Resistivity measurements yield residual resistance ratios [$\rho(300\text{ K})/\rho_0$] of 2.6, 5.5, and 13.3, respectively, for the films denoted here as samples No. 1 to No. 3. Sample No. 1 did not become superconductive, whereas films No. 2 and No. 3 show a superconducting onset at 1.74 K and 1.97 K, respectively. For all the data presented here the magnetic field was oriented parallel to the c axes of the films.

In Fig. 1 the striking different behavior of $R_H(T)$ for the film with high defect density [Fig. 1(a), film No. 1] and the films with lower defect density [Fig. 1(b), films No. 2 and No. 3] becomes obvious. For all samples the slope of $R_H(T)$ is negative at elevated temperatures, passes a maximum (at 60 K for sample No. 1 and at 50 K to 60 K for samples No. 2 and No. 3, respectively) and, for the present, decreases with further decreasing temperature. The position of this maximum usually represents a good measure for the Kondo–lattice temperature T^*.[3] At 6 to 7 K minima in the Hall coefficient $R_H(T)$ of films No. 2 and No. 3 occur. Film

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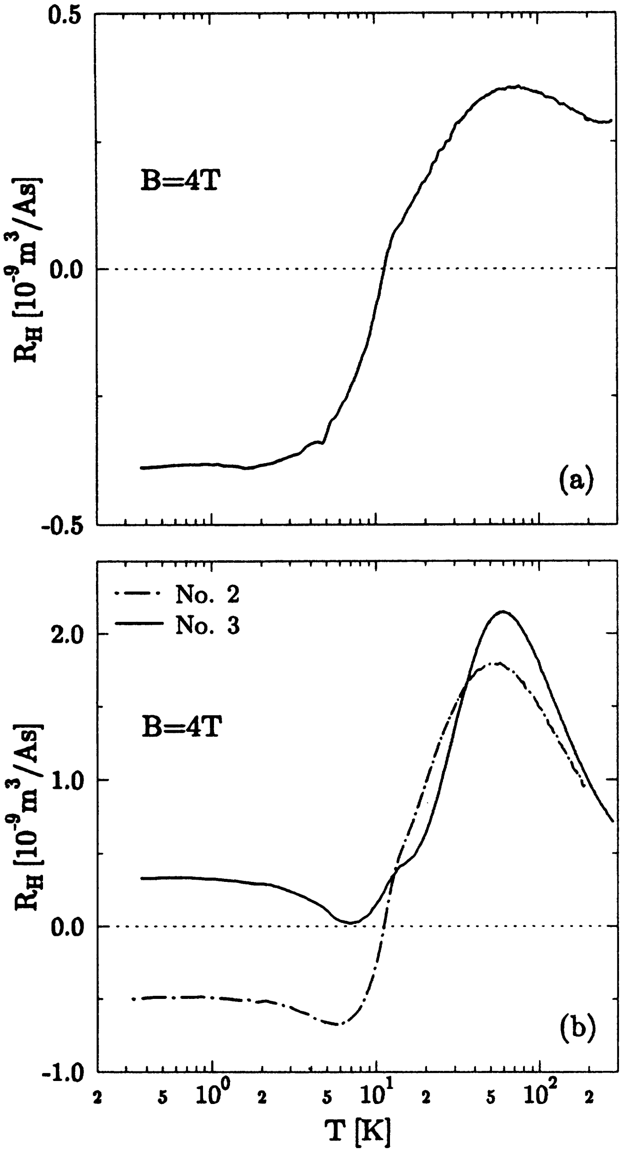

FIG. 1. Hall coefficient R_H as a function of temperature for the films No. 1 (a) and Nos. 2 and 3 (b). The magnetic field, 4 T, is oriented parallel to the films' c axes.

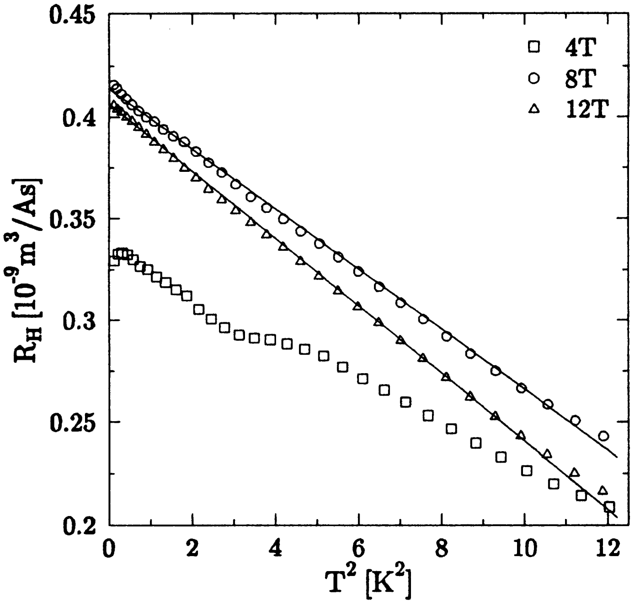

FIG. 2. Low-temperature Hall coefficient of film No. 3 in magnetic fields of 4 T, 8 T, and 12 T. The solid lines correspond to a fit of the 8 T and 12 T data according to $R_H^{\mathrm{coh}}(T)=R_H^0-a'T^2$ with $R_H^{0(8\ \mathrm{T})}=4.14\times10^{-10}$ m^3/A s, $a'^{(8\ \mathrm{T})}=1.48\times10^{-11}$ m^3/As K^2 and $R_H^{0(12\ \mathrm{T})}=4.07\times10^{-10}$ m^3/A s, $a'^{(12\ \mathrm{T})}=1.66\times10^{-11}$ m^3/As K^2.

No. 1, on the other hand, shows a nearly flat saturation behavior with evidence for a shallow minimum at 2 K. The maximum at 60 K is less pronounced as compared to samples No. 2 and No. 3. We consider the almost complete depression of the minimum at lower temperatures to be a clear sign for strongly disturbed coherence in this sample. The further discussion will therefore concentrate on samples No. 2 and No. 3.

In the temperature range above 60 K the Hall coefficient is dominated by skew scattering (see Fig. 1). The behavior can be described by the well-known dependence[3,4]

$$R_H^{\mathrm{skew}}(T)=\gamma\rho_m(T)\chi'(T) \qquad (2)$$

with the magnetic resistivity contribution $\rho_m(T)=\rho(T)-\rho_0-\rho_{\mathrm{Phonon}}(T)$, the reduced susceptibility $\chi'(T)=\chi(T)/C$ (C is the Curie constant), and the parameter γ which is constant in the temperature regime above T^* and becomes temperature dependent below. This will be discussed in more detail in the range $T<T_N$ later on. The susceptibility follows a Curie-Weiss law for the magnetic field oriented parallel to the c axis in the temperature range above 50 K changing to a nearly temperature-independent Van Vleck paramagnetism below.[13] The onset of antiferromagnetic order manifests itself in a slope anomaly at T_N. Based on a Curie-Weiss law for $\chi(T)$ the high-temperature data ($T>60$ K) of samples No. 2 and No. 3 were analyzed according to a least-squares fit of $R_H(T)$ vs $[\rho(T)-\rho_0]/(T-\Theta)$ with Θ as a free parameter (the phonon contribution has been neglected). This yields Curie-Weiss temperatures of $\Theta=-160$ K ($T_N=11.7$ K) and $\Theta=-95$ K ($T_N=10.8$ K) for samples No. 2 and No. 3, respectively.

In the temperature range below $T_N\simeq11$ K, which lies clearly in the coherent regime ($T\leqslant T^*\simeq60$ K), we limit our analysis to the data of film No. 3 since it shows the largest residual resistance ratio.

Below 3.5 K the resistivity shows the typical Fermi-liquid behavior $\rho(T)=\rho_0+aT^2$.[14] Therefore we denote $T_F\simeq3.5$ K as the Fermi-liquid temperature. In the same temperature range the Hall coefficient can be described almost perfectly by a dependence of the form

$$R_H(T)=R_H^0+R_H^{\mathrm{coh}}(T)=R_H^0-a'T^2 \quad (a'>0) \qquad (3)$$

in the magnetic field range above 8 T (see Fig. 2). (The anomaly at 2 K for a field of 4 T corresponds to an anomaly

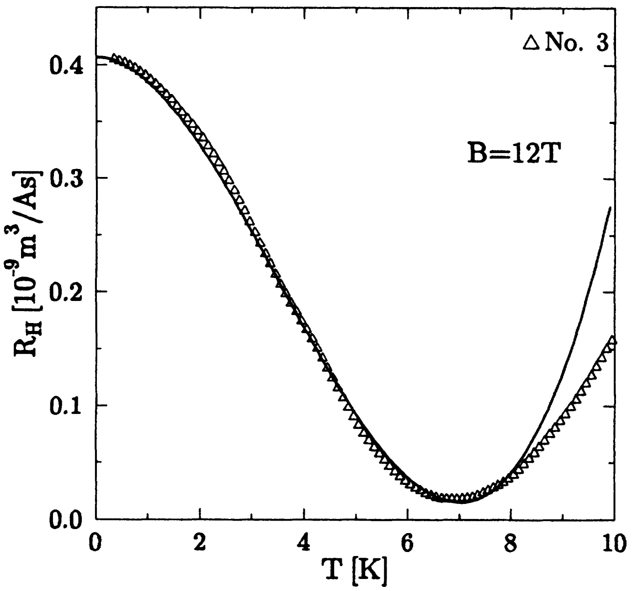

FIG. 3. Hall coefficient of sample No. 3 below 10 K in a magnetic field of 12 T. The solid line corresponds to a fit according to the assumption of independent Hall currents for the coherent and skew-scattering contribution. See text for details.

in the field dependence of the Hall resistivity at low temperatures that will be discussed in a forthcoming publication.) The same T^2 behavior is observed in sample No. 2 with almost the same coefficient a', whereas the residual Hall value R_H^0 shows the opposite sign as compared to sample No. 3. The reason for this sign reversal is still unclear.

Skew scattering can be excluded as a reason for this temperature dependence for at least two reasons. First, the dominance of small-angle scattering in the low-temperature regime leads to a freezing out of skew scattering in the lattice for $T\rightarrow 0$. Secondly, assuming that the proportionality $R_H(T) \propto \rho_m(T)$ with $\chi'(T)=\text{const}$ is still valid a T^2 behavior with positive coefficient should result. Side-jump effects (arising from parts in the interaction Hamiltonian that do not commute with the position operator) can be excluded due to a proportionality $R_H^{sj} \propto \rho_m^2(T)$ that would lead to a T^4 temperature dependence of $R_H(T)$.[15]

We assume the coherent ground state itself to be responsible for the temperature dependence given by Eq. (3). The Hall coefficient, among other transport properties, is given through the transport integrals (for a spherical Fermi surface):[16]

$$L_n^m=\int_{-\infty}^{\infty}\left(-\frac{\partial f}{\partial\epsilon}\right)\tau^m\epsilon^n d\epsilon\ , \quad R_H=-\frac{1}{n|e|}\frac{L_0^2}{(L_0^1)^2} \tag{4}$$

According to the lattice version of the noncrossing approximation (LNCA) for the periodic Anderson Hamiltonian $\tau(\epsilon)$ is basically determined through the Green's function of the local states whose imaginary part represents the Kondo resonance in the vicinity of E_F and can be calculated in the LNCA procedure.[17] The calculation leads to a quadratic temperature dependence of the Hall coefficient as it is observed in UPd$_2$Al$_3$ (see Fig. 2). This is due to an initially T^2-like reduction of the resonance height at low temperatures.[18]

The main physical cause for our derivation is the strong temperature dependence of the height of the ASR. It alone basically determines the low-temperature properties of the Hall coefficient and can account for the observed $R_H^{\text{coh}}(T)$ behavior. This is true not only for the system investigated here but in principle for all Kondo lattices in the low-temperature regime. As an example, measurements of the Hall coefficient in the Kondo-lattice system CeCu$_6$ show striking similarities to UPd$_2$Al$_3$ as presented here. In CeCu$_6$ a low-temperature minimum in $R_H(T)$ occurs at 0.3–0.5 K, which is roughly one-tenth of $T^*\simeq 4.6$ K.[6,7] Below 0.3 K $R_H(T)$ is negative and shows an almost linear temperature dependence with a negative slope. The different $R_H(T)$ characteristics in the Fermi-liquid regime of heavy-fermion systems can be accounted for principally in the following way: The position of the ASR strongly depends on crystal-field effects and spin-orbit coupling (see, for example, Ref. 19). The temperature-dependent height variation of the resonance itself can be modified by band-structure effects on the resonance, arising at low temperatures when coherent f-derived quasiparticle bands evolve.[20]

With increasing temperature (but below $T_N=10.7$ K for sample No. 3) the analysis of the resistivity data reveals an additional T^3 contribution which is most likely due to electron-magnon scattering. The deviations of $R_H(T)$ from the T^2 behavior are assumed to be caused by increasingly relevant skew scattering that is connected to the magnetic scattering part of the resistivity [see Eq. (2)]. The skew-scattering contribution for the single-impurity system below the Kondo temperature is determined by the interference between the $l=2$ (nonresonant) and $l=3$ (resonant) partial-wave states.[3,4] This results in the following γ factor [see Eq. (2)]

$$\gamma=-\frac{\pi g\mu_B\sin(\delta_2-2\delta_3)}{(2J+1)k_B\sin\delta_2} \tag{5}$$

with the partial-wave scattering phases δ_2 and δ_3. For a tentative extrapolation of this single-impurity result to the coherent regime we choose the following parameters in order to estimate the γ factor for sample No. 3: $J=1/2$ and $g=2$ according to the results of neutron scattering on single crystals which show a temperature-dependent variation of the magnetization following approximately a $S=1/2$ Brillouin function.[21] Furthermore, $\chi'=(T-\Theta)^{-1}|_{T=50\text{ K}}$ according to the above mentioned nearly temperature-independent paramagnetism below 50 K with $\Theta=-95$ K. In order to account for the crude approximations we introduce an additional factor α for the skew-scattering part in Eq. (3). This results in the following temperature dependence of the Hall coefficient below T_N:

$$R_H(T)=R_H^0-a'T^2+\alpha\cdot\gamma\chi'(50\text{ K})bT^3 \tag{6}$$

With b deduced from the T^3 contribution in the resistivity (in a magnetic field of 12 T) and $\alpha=1.28$, which is close to unity, we get the behavior depicted in Fig. 3 by the solid line. The fit is satisfactory below 8 K with clear deviations above. Since the susceptibility shows a cusp at T_N (Ref. 13) and the phase shift δ_3 is reduced with increasing temperature from $\delta_3(T=0)=\pi/2$ (for twofold degeneracy) the skew-scattering part will gain a different temperature dependence close to T_N.

The minimum in the Hall coefficients of $\mathrm{CeCu_6}$, $\mathrm{UPt_3}$, and CeNi could be explained in the same qualitative manner even if the skew-scattering contribution should be completely different due to the varying importance of magnetic correlations in these systems. In $\mathrm{UPd_2Al_3}$ the magnetism is close to the limit of well localized moments.[10] As presented here, a minimum in $R_H(T)$ is observed for the magnetic field oriented perpendicular to the easy plane of magnetization. In $\mathrm{UPt_3}$ the minimum is observed only in single crystals with the magnetic-field orientation parallel to the c axis, i.e., perpendicular to the orientation of the small spin-density-wave-derived moments in the antiferromagnetic regime.[8,9] For $\mathrm{CeCu_6}$ only local magnetic correlations exist below $T^*\simeq 4.6$ K.[22] CeNi, finally, is a mixed valence compound with compensated moments due to the enhanced hybridization broadening of the localized f states.[23] Since the minimum in the Hall coefficient is observed in different systems covering the whole range of possible magnetic correlations we conclude that magnetism alone cannot be the reason for the observed minimum. This underlines the necessary inclusion of the coherent part to $R_H(T)$ that was performed here.

In conclusion, Hall-effect measurements on thin epitaxial $\mathrm{UPd_2Al_3}$ films with various defect densities were performed. In films of improved purity (coherent Kondo lattices) the Hall coefficient varies in the Fermi-liquid temperature range below $T_F \simeq 3.5$ K like $R_H(T)=R_H^0+R_H^{\mathrm{coh}}(T)=R_H^0-a'T^2$ $(a'>0)$. With increasing temperature a minimum at roughly one-tenth of the Kondo-lattice temperature $T^*\simeq 60$ K is observed. These features can be explained, if the additivity of independent Hall currents arising from a coherent part (varying like $-a'T^2$, $a'>0$) and a skew-scattering part (varying like the magnetic part of the resistivity $\propto T^3$), respectively, is assumed. The observed minima in the Hall coefficients of $\mathrm{UPt_3}$, $\mathrm{CeCu_6}$, and CeNi are assumed to be due to the same superposition of coherent and skew-scattering contributions.

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