PHYSICAL REVIEW B VOLUME 32, NUMBER 10 15 NOVEMBER 1985

Hall effect in $Ce_{1-x}Y_xPd_3$ mixed-valence alloys

A. Fert, P. Pureur,* and A. Hamzic†
Laboratoire de Physique des Solides, Université de Paris-Sud, F-91405 Orsay, France

J. P. Kappler
Laboratoire de Magnetisme et de Structure Electronique des Solides, 3 rue de l'Université, F-67084 Strasbourg, France

(Received 3 September 1985)

Mixed-valence and Kondo lattice systems exhibit large anomalous Hall coefficients with a striking change of sign at low temperature in several systems ($CePd_3$, $CeCu_6$, . . . , etc.). We have studied the Hall effect of $Ce_{1-x}Y_xPd_3$, in which the substitution of small amounts of Y for Ce prevents the development of coherence at low temperature. We find that the Hall coefficient does not change its sign at low temperature and can be well understood in the one-impurity model of Ramakrishnan, Coleman, and Anderson. We infer that the change of sign observed in $CePd_3$ is an effect of coherence.

Mixed-valence and Kondo rare-earth systems exhibit anomalous Hall coefficients, as has been observed in Ce (Refs. 1 and 2) and Tm (Ref. 3) dilute alloys and in several intermetallic compounds.[4-9] A striking result is the change of sign of the Hall coefficient in $CePd_3$,[5] SmB_6,[6] $CeBe_{13}$,[5] $CeCu_6$,[9] and YbCuAl.[5] A plausible origin of these Hall-effect anomalies is the existence of skew scattering.[10] An early model[11] of the Hall effect induced by Kondo Ce impurities was based on the calculation of the skew scattering by a Coqblin-Schrieffer interaction[12] between conduction electrons and Ce impurities. This model is valid only in the high-temperature limit, i.e., $T >> T_K$. Recently, a more general model has been proposed by Ramakrishnan, Coleman, and Anderson.[13,14] The expression of the Hall coefficient found by these authors [see Eqs. (4) and (9) in Ref. 13] is written as

$$R_H = R_H^0 + g\mu_B|\alpha|\rho\sin(\phi+\delta_2)/\sin\delta_2 \ , \tag{1}$$

with $\phi = -2\delta_3$ in the low-temperature limit ($T << T_K$) and $\phi = -\pi$ in the high-temperature limit. δ_3 is the phase shift associated with the resonance scattering in the $l=3$ channel at low temperature, δ_2 is the phase shift due to additional potential scattering in the $l=2$ channel, ρ is the resistivity, and $|\alpha|$ is proportional to $\tilde{\chi}(1-\tilde{\chi}T)$, where $\tilde{\chi}$ is the reduced magnetic susceptibility, i.e., $\tilde{\chi} = 3\chi/g^2\mu_B^2 J(J+1)$. The change of ϕ from $-2\delta_3$ to $-\pi$ as the temperature increases is related to the renormalization of the f-level position. Ramakrishnan *et al.*[13,14] have proposed to ascribe the change of sign of the Hall effect at low temperature to the change of sign of $\sin(\phi+\delta_2)$ as ϕ shifts from $-\pi$ to $-2\delta_3$. However, the model of Ramakrishnan *et al.*[13,14] describes the skew scattering by *independent* Ce impurities and does not take into account the coherence effects occurring at low temperature in mixed-valence and Kondo lattices. Alternatively, it is tempting to ascribe the change of sign of the skew scattering term to the onset of coherence and to the resulting changes in the scattering processes. In $CePd_3$, for example, R_H is found to drop in the temperature range where the coherence appears (drop of the resistivity). In the same way, in $CeCu_6$, the change of sign of R_H seems to be related to coherence effects.[9] In order to establish if the decrease and the change of sign of R_H in $CePd_3$ is due to the onset of coherence, we have studied the Hall effect of $Ce_{1-x}Y_xPd_3$ alloys with $x = 0, 0.1, 0.3$. It is known that the substitution of a few atomic percent of Y for Ce in $CePd_3$ breaks the coherent state: with only 3 at.% of Y (i.e., $x = 0.03$) the resistivity drop at low temperature disappears almost completely;[15] for $x = 10$ and 30 the resistivity levels off at a very high value in the low-temperature limit,[16] which is the typical behavior for independent mixed-valence and Kondo impurities.[17] To probe the role of coherence in the change of sign of R_H, we compare the Hall effect of coherent $CePd_3$ and incoherent $Ce_{1-x}Y_xPd_3$.

Samples of $Ce_{1-x}Y_xPd_3$ with $x = 0, 0.1$, and 0.3 were prepared from 99.99% pure metals by arc melting under a pure argon atmosphere. The magnetic and transport properties of samples prepared in this way have been described previously.[16] The samples for Hall-effect studies were in the form of platelets ($1\times3\times10$ mm^3) and the measurements were performed by a standard ac technique up to 7 T between 1.2 K and room temperature. We present below our results on the initial Hall coefficient R_H.

In Fig. 1 we show R_H vs T for our three samples. Our results for $CePd_3$ confirm the change of sign at about 20 K already observed by Cattaneo, Häfner, and Wohlleben.[5] On the contrary, for $Ce_{0.9}Y_{0.1}Pd_3$ and $Ce_{0.7}Y_{0.3}Pd_3$, R_H increases monotonically when T is lowered from room temperature to 1.2 K. For these two $Ce_{1-x}Y_xPd_3$ alloys the variation of R_H with T fits roughly the variation of $\rho\tilde{\chi}(1-\tilde{\chi}T)$ calculated from independent measurements of ρ and $\tilde{\chi}$ (Refs. 16 and 19) and represented by dashed curves in Fig. 1. This rough agreement between the variation of R_H and $\rho\tilde{\chi}(1-\tilde{\chi}T)$ (Ref. 20) means the Hall effect of our two $Ce_{1-x}Y_xPd_3$ alloys is approximately explained by the one-impurity model of Ramakrishnan *et al.* with temperature-independent values of ϕ in Eq. (1) (from the neutron quasielastic width[21] and the susceptibility data,[16] the Kondo temperature T_K of our $Ce_{1-x}Y_xPd_3$ alloys is in the range 500–1000 K, which is consistent with an approximately constant value of ϕ in our experimental range 1–300 K).

Now, if the behavior of R_H for $x = 0.1$ and 0.3 is characteristic of incoherent Ce and explained by the one-impurity model of Ramakrishnan *et al.*, we infer that the behavior of R_H in $CePd_3$, with a drop below 100 K and a change of sign at about 20 K, has to be ascribed to the onset of coherence. It has been suggested to one of us (A.F.) by Coleman that the effect of coherence might be described by introducing a crystal-field splitting in the coherent state and the resulting

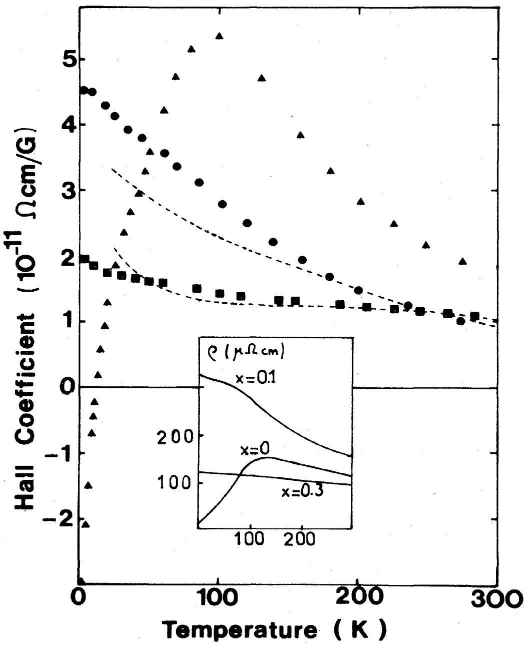

FIG. 1. Hall coefficient R_H vs temperature for $CePd_3$ (▲) (Ref. 18), $Ce_{0.9}Y_{0.1}Pd_3$ (●), and $Ce_{0.7}Y_{0.3}Pd_3$ (■). The dashed curves represent the variation (arbitrary scale) of $\rho\tilde{\chi}(1-\tilde{\chi}T)$ calculated from independent measurement of the resistivity ρ and reduced susceptibility $\tilde{\chi}$ (see Ref. 16). Inset: Resistivity vs temperature for the same alloys (see Ref. 16).

new phase shifts in Eq. (1). However, comparing the susceptibility and the Wilson ratio of $CePd_3$ and $Ce_{1-x}Y_xPd_3$ (Ref. 16) hardly supports the idea of different crystal-field splittings in the coherent and incoherent states. Rather we believe that a specific treatment of the scattering processes in the coherent state is needed to explain the Hall effect. In the presence of coherence the electrons are scattered only by the nonperiodic part of the potential, i.e., by fluctuations about the coherent state. A similar situation occurs in the Hall effect of ferromagnets such as Gd, Tb, Dy, . . . , : In the paramagnetic state the anomalous Hall effect is due to independent orbital exchange terms, $V^i_{\rm scatt} \sim \mathbf{l}\cdot\mathbf{J}_i$, whereas, in the ferromagnetic state, the scattering is due to fluctuations about the ferromagnetic state, $V^i_{\rm scatt} \sim \mathbf{l}\cdot(\mathbf{J}_i - \langle\mathbf{J}\rangle)$, and this even leads, in some cases, to a change of sign of the anomalous Hall effect between T_c and 0 K.[22] Of course, the problem of the scattering by fluctuations about the coherent state is likely much more complex. In addition, the change of the band structure and Fermi surface at the onset of coherence could also affect the scattering processes significantly.

In conclusion, our results on $Ce_{1-x}Y_xPd_3$ alloys show that the change of sign of the Hall effect in $CePd_3$ is a coherence effect and disappears when Y impurities prevent the development of coherence. For the incoherent $Ce_{1-x}Y_xPd_3$ alloys, the Hall coefficient fits the predictions of the independent impurity model of Ramakrishnan *et al.*[13,14] The relation between the change of sign of the Hall coefficient and the onset of coherence seems to have also been observed in other systems and poses an interesting problem.

*Permanent address: University of Rio Grande do Sul, Porto Alegre, Brasil.

†Permanent address: Institute of Physics of the University, 41001 Zagreb, Yugoslavia.

[1]A. Fert, and O. Jaoul, Solid State Commun. **11**, 759 (1972).

[2]R. Dreyer and K. Winzer, Solid State Commun. **46**, 7 (1983).

[3]P. Haen and F. Lapierre (unpublished).

[4]M. Christen, Ph.D. thesis, Genève, 1979 (unpublished).

[5]E. Cattaneo, U. Häfner, and D. Wohlleben, *Valence Instabilities*, edited by P. Wachter and H. Boppart (North-Holland, Amsterdam, 1982), p. 451; E. Cattaneo, J. Magn. Magn. Mater. **47-48**, 529 (1985).

[6]J. W. Allen, B. Batlogg, and P. Wachter, Phys. Rev. B **20**, 12 (1979).

[7]N. B. Brandt, V. V. Moshchalkov, N. E. Slachanks, E. M. Savitskii, and T. M. Shkatover, Solid State Commun. **53**, 645 (1985).

[8]J. Flouquet (private communication).

[9]T. Penney, J. Stankiewicz, S. von Molnar, Z. Fisk, J. L. Smith, and H. B. Ott (unpublished).

[10]For a review on skew scattering, see A. Fert and A. Hamzić, *The Hall Effect and Applications*, edited by C. L. Chien and C. Westgate (Plenum, New York, 1980).

[11]A. Fert, J. Phys. F **3**, 2126 (1973); D. Sorg, Ph.D. thesis, Geneve, 1974 (unpublished).

[12]B. Coqblin and J. R. Schrieffer, Phys. Rev. **185**, 847 (1969).

[13]T. V. Ramakrishnan, P. Coleman, and P. W. Anderson. J. Magn. Magn. Mater. **47-48**, 493 (1985).

[14]P. Coleman, P. W. Anderson, and T. V. Ramakrishnan, Phys. Rev. Lett. **55**, 414 (1985).

[15]H. Schneider and D. Wohlleben, Z. Phys. B **44**, 193 (1981).

[16]M. J. Besnus, J. P. Kappler, G. Krill, M. F. Ravet, N. Handaoui, and A. Meyer, *Valence Instabilities*, edited by P. Wachter and H. Boppart (North-Holland, Amsterdam, 1982), p. 163.

[17]N. E. Bichers, D. L. Cox, and J. W. Wilkins, Phys. Rev. Lett. **54**, 230 (1985).

[18]In fact, our $CePd_3$ sample is a $Ce_{26}Pd_{74}$ alloy for which the coherence effects are slightly stronger than in $Ce_{25}Pd_{75}$; see M. J. Besnus, J. P. Kappler, and A. Meyer, J. Phys. F **13**, 597 (1983).

[19]For $Ce_{0.9}Y_{0.1}Pd_3$; and $Ce_{0.7}Y_{0.3}Pd_3$, $1-\tilde{\chi}$ departs little from 1 in our temperature range, so that the variation of $\rho\tilde{\chi}(1-\tilde{\chi}T)$ is essentially that of $\rho\tilde{\chi}$.

[20]The ordinary Hall coefficient R^0_H is approximately known from data on YPd_3, see Ref. 5. It is relatively small and, in first approximation, can be neglected in Eq. (1).

[21]E. Holland-Moritz, D. Wohlleben, and M. Loewenhaupt, Phys. Rev. B **25**, 7482 (1982).

[22]B. Giovannini, Phys. Lett. **36A**, 381 (1971).