APPENDIX: DERIVATION OF EQS. (25) AND (26)

Consider

$$D_w \equiv (e^{-\beta H})_w \ . \tag{A1}$$

Then,

$$\frac{d}{d\beta} D_w = -H_w G D_w \ , \tag{A2}$$

where

$$H_w = -\frac{1}{4}\sum_{i \ne l} J_{il}\ \vec{\Omega}_i \cdot \vec{\Omega}_l \ . \tag{A3}$$

Try the form

$$D_w = e^{A(\beta)}, \quad A(\beta) = \sum_{n=1}^{\infty} a_n \beta^n \ . \tag{A4}$$

Then, substituting (A4) into (A2) and equating the coefficients of β^n in both sides, we obtain

$$a_1 = -H_w, \quad a_2 = \tfrac{1}{2}\ [(H^2)_w - H_w^2] \ ,$$

$$a_3 = -\frac{1}{3!}\ [(H^3)_w - H_w^3] + a_2 H_w \ , \tag{A5}$$

and Eq. (26) follows straightforwardly.

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Field Dependence of the Anomalous Hall Coefficient in Dilute Magnetic Alloys*

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(Received 12 June 1970)

The anomalous Hall coefficient R of dilute magnetic alloys exhibiting the Kondo effect is calculated in the second Born approximation using the s-d exchange model. The variation of R as a function of the magnetic field, for fixed temperature, is studied. Comparisons are made with other theories and recent experimental data.

I. INTRODUCTION

The effect of an external magnetic field on the Kondo[1] behavior of the electrical resistivity in dilute magnetic alloys has been studied theoretically from two different approaches: The S-matrix theory of More and Suhl[2] and a second-Born-approximation calculation by the authors.[3] The first approach is in principle correct at all temperatures and fields, but because of its complexity and the amount of computer calculation needed, it cannot easily be fitted to the experimental results. The second one gives explicit formulas which can readily be compared with experiment, but it is only a rough approximation, and the limits of its validity can only be fixed through the exact theory. Both papers contained calculations of the conduction-electron relaxation times for spin up (τ_+) and spin down (τ_-). Recently, More has used the S-matrix life-times to compute the Hall coefficient as a function of the magnetic field $R(H)$ for fixed temperatures.[4] He obtains curves of $R(H)$ increasing more or less rapidly with H, which he compared with the experimental data on impure gold due to Gaidukov.[5] However, the physical explanation of this behavior could not be obtained from this calculation. In the present paper we shall calculate $R(H)$ using the expressions for τ_+ and τ_- obtained in third-order perturbation theory.[3] We show that the field dependence of R is essentially explained in terms of the square of the impurity spin magnetization[6] for low fields $g\mu_B H/kT < 1$. We compare this result with More's and with the recent data.[7] We also obtain the very high-field $(g\mu_B H/kT > 10)$ behavior of $R(H)$. This last case is discussed more extensively elsewhere.[8] The behavior of R is compared with the similar behavior of the magnetoresistivity, and suggestions are made for further experiments.

II. CALCULATION OF THE HALL COEFFICIENT

In this section we calculate $R(H)$ for a very dilute alloy, in order to deal with the one-impurity problem only. The assumptions are the same as those we used in calculating the magnetoresistivity,[3] and we will omit the details of the calculation here. We suppose the electrons to be free in an applied magnetic field H; the perturbations are an ordinary spin-independent potential V and the s-d exchange potential $-2J\vec{s}\cdot\vec{S}$ between the conduction-electron spin density $\vec{s}$ at the impurity, and the impurity spin $\vec{S}$. We will restrict ourselves to the case $J \ll V \ll \epsilon_F$, the Fermi energy. Following More,[4] we define the Hall coefficient in a "two-band" model of spin-up and spin-down conduction electrons, which is valid for $\omega_c \tau \ll 1$, ω_c being the cyclotron frequency. (We shall return to this point in comparing our results to the experimental data.) The calculation of the corresponding relaxation times τ_+ and τ_-[9] is given in detail in Ref. 3. We have

$$R(H)/R(0) = 2(\tau_+^2 + \tau_-^2)/(\tau_+ + \tau_-)^2 \qquad (1)$$

In principle, energy averages, with a weight factor given by the derivative of the Fermi function, and averages over the impurity spin orientations are involved in Eq. (1). Suhl[10] has shown that the energy average of τ and τ^2 for a vanishingly small field leads to a 20% effect in the temperature dependence of R. As we are only interested in fixed temperatures here, we shall drop, as More[4] does, the energy average effect which would give a different normalization for R-vs-H curves at different temperatures. We therefore evaluate τ_+ and τ_- at ϵ_F. Assuming the impurity g value to be 2, one gets, after some algebra,

$$1/\tau_\pm = a \mp b \quad, \qquad (2)$$

$$a = (k_F m v_0 c/\pi\hbar^3)\{V^2 + J^2[S(S+1) - \langle S_z\rangle \tanh(\alpha/4)]$$
$$\times [1 + 4JG(H,T)]\} \quad,$$

$$b = (k_F m v_0 c/\pi\hbar^3)\, 2VJ\langle S_z\rangle\, [1 + 2JG(H,T)] \quad, \qquad (3)$$

where v_0 is the atomic volume of the host metal, k_F the Fermi wave number, c the impurity concentration, $\langle S_z\rangle$ the thermal average of the impurity spin operator S_z, $\alpha = g\mu_B H/kT$, and

$$G(H,T) = \frac{3z}{2\epsilon_F}\left[1 + \frac{1}{2}\ln\frac{kT}{4\epsilon_F} + \frac{1}{2}I\left(\frac{\mu_B H}{kT}\right)\right], \qquad (4)$$

$$I(x) = \frac{1}{4}\int_0^\infty \operatorname{sech}^2\frac{x'}{2}\ln|x'^2 - x^2|\,dx' \quad, \qquad (5)$$

where z is the number of electrons per atom. In Fig. 1 we plot $4I(x)$ against $x = \alpha/g$; the asymptotic behavior of $I(x)$ for small and large x is

$$I(x) \simeq -0.1256 + 0.2132x^2 \quad, \quad x \ll 1$$
$$I(x) \simeq \ln x - 0.8221x^{-2} \quad, \qquad x \gtrsim 10 \qquad (6)$$

Combining Eqs. (1) and (2) gives

$$R(H)/R(0) = 1 + b^2/a^2 \quad, \qquad (7)$$

so that

$$\frac{\Delta R}{R(0)} \equiv \frac{R(H) - R(0)}{R(0)} = \frac{b^2}{a^2} = 4\left(\frac{J}{V}\right)^2 \langle S_z\rangle^2 \frac{[1 + 2JG(H,T)]^2}{\{1 + (J/V)^2[S(S+1) - \langle S_z\rangle\tanh\frac{1}{4}\alpha][1 + 4JG(H,T)]\}^2} \qquad (8)$$

We shall be interested in the behavior of Eq. (8) as a function of H for fixed temperature. Then the small-field behavior $(g\mu_B H/kT \ll 1)$ is dominated by the prefactor $\langle S_z\rangle^2$, and the high-field behavior $(g\mu_B H/kT \gg 1)$ will be given by the $\ln(\mu_B H/4\epsilon_F)$ part of $G(H,T)$ once $\langle S_z\rangle$ has saturated.

III. COMPARISON WITH THEORY AND EXPERIMENT

A. Theory

Here we shall compare our results with More's computed curves for $R(H)$ in the full S-matrix theory.[4] He finds that R increases with H, more rapidly for lower temperatures, and has a zero slope for $H = 0$. This is in agreement with the behavior we find from our perturbative formulas. In curve (b) of Fig. 2 we have plotted Eq. (8) as a function of H for fixed temperature ($T = 1.2\,°K$) with the following choice of parameters: $\epsilon_F = 7.1$
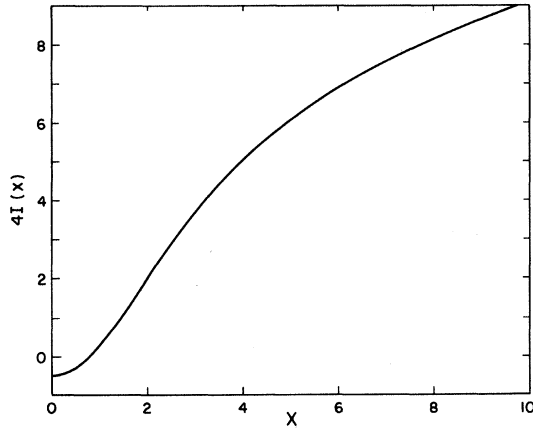
eV, $J = -0.12$ eV, $|J|/V = 0.1$, and $S = 2$. (This choice was obtained from a fit to the data on CuMn alloys; see Sec. III B below.) We have also assumed that $\langle S_z\rangle$ obeys a Brillouin law,

$$\frac{\langle S_z\rangle}{S} = B_S(y) = \frac{2S+1}{2S}\coth\left(\frac{2S+1}{2S}y\right) - \frac{1}{2S}\coth\left(\frac{y}{2S}\right),$$

$$y = Sg\mu_B H/kT \quad. \qquad (9)$$

Equations (8) and (9) then imply that $\Delta R/R(0)$ will have a zero slope at $H = 0$ and will increase more rapidly with H for lower T. The first Born approximation to the scattering amplitudes gives

$$\frac{\Delta R}{R(0)} = 4\left(\frac{J}{V}\right)^2 \langle S_z\rangle^2 \bigg/ \left\{1 + \left(\frac{J}{V}\right)^2\left[S(S+1) - \langle S_z\rangle\tanh\frac{\alpha}{4}\right]\right\}^2,$$

$$(10)$$

FIG. 1. The function $4I(x)$ vs x.

which has the same low-field behavior. The second Born approximation [Eq. (8)] changes the coefficient of $\langle S_z \rangle^2$, but the field dependence of these modifications (for $g\mu_B H/kT \lesssim 1$ and at fixed temperature) is quite small. Curve (a) of Fig. 2 corresponds to Eq. (10) with the same value of $|J|/V$, 0.1. Curves (a) and (b) exhibit the same qualitative behavior in the low-field regime $g\mu_B H/kT \lesssim$, so Eq. (10) gives a rough but physical explanation for the increase of $R(H)$ in More's[4] results: This corresponds to the rapid variation of $\langle S_z \rangle^2$ for small H.

Calling M the impurity magnetization in Bohr magnetons per atom, we have

$$M = g\langle S_z \rangle \quad , \tag{11}$$

and we may say that the dominance of the $\langle S_z \rangle^2$ term in Eqs. (8) and (10) for $g\mu_B H/kT < 1$ implies that

$$\Delta R/R \propto M^2 \quad , \tag{12}$$

at least in perturbative treatments. The same behavior was noted in the second Born result for the negative magnetoresistivity,[3] where in the low-field regime the dominant field and temperature dependence was given by

$$\frac{\rho(H) - \rho(0)}{\rho(0)} \equiv \frac{\Delta\rho}{\rho(0)} \propto M^2 \quad . \tag{13}$$

This dominance of the M^2 part of $\Delta\rho/\rho(0)$ and $\Delta R/R(0)$ in the low-field regime has been verified experimentally (see Ref. 3 and Sec. III B below) and in these perturbative calculations may be traced back to the relatively weak field and temperature dependence of the scattering amplitudes compared to the rapid variation of M^2 for small $g\mu_B H/kT$. This observation allows us to generalize a remark we made in connection with the magnetoresistivity: In zero field the behavior of the transport coefficients of Kondo-effect systems is given by the perturbation series (or more refined nonperturbative

theories) for the scattering amplitudes, which display $\ln T$ behavior. But as soon as a magnetic field is present, the deviation of the transport coefficients from their zero-field behavior is dominated by the impurity magnetization because it is much more rapidly varying for small $g\mu_B H/kT$ than the logarithmic terms which come from the scattering amplitudes. Furthermore, this suggests that to a good approximation we can ignore the field dependence of the scattering amplitudes in obtaining the low-field behavior of the transport coefficients, so that we may write

$$[\tau_{\pm}(\epsilon)]^{-1} \propto \text{Im}t(\epsilon) \pm M \text{Im}\tau(\epsilon) \quad , \tag{14}$$

where t and τ are the ordinary and spin-flip scattering amplitudes, respectively, *in zero field*. This automatically leads to

$$\frac{\Delta\rho}{\rho(0)} \propto \frac{\Delta R}{R(0)} \propto M^2 \quad , \tag{15}$$

to all orders of perturbation theory, for small enough M. It also allows us to clarify Tsuji's conjecture, based on a first Born calculation, that $\Delta R/R(0)$ and $\Delta\rho/\rho(0)$ are equal.[6] Different energy averages of the lifetimes given by Eq. (14) enter into the calculation of $R(H)$ and $\rho(H)$. These differences do not effect the first-Born-approximation
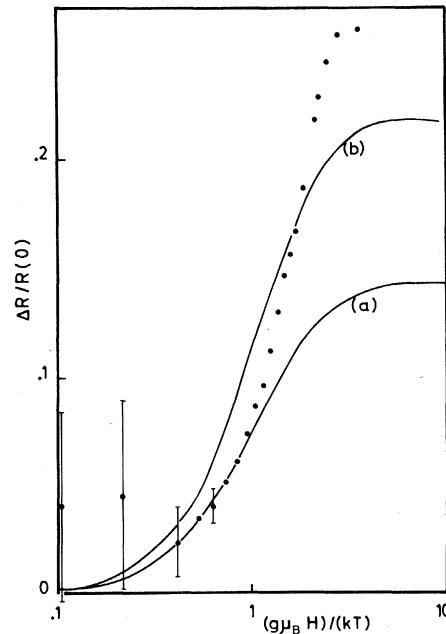

FIG. 2. $\Delta R/R(0)$ vs $g\mu_B H/kT$ for $g=2$ and $T=1.2\,^\circ\text{K}$. Curve (a) is the first Born fit [Eq. (10)] with the parameters: $\epsilon_F = 7.1$ eV, $|J|/V = 0.1$, and $S=2$; curve (b) is the second Born fit with the same parameters and $J=-0.12$ eV. The points are the experimental results of Ref. 7 for a CuMn alloy with 150 ± 10 ppm Mn. The error bars are largest for the lowest fields; the precision increase rapidly for higher fields.

results, but we expect the rapid variation of the higher-order scattering amplitudes near the Fermi surface, which makes the transport coefficients extremely sensitive to the energy averaging procedure, to lead to appreciable deviations from equality between $\Delta R/R$ and $\Delta\rho/\rho$.

Finally, we note that, like More, we do not find a discontinuous jump in $R(H)$ as was found experimentally by Gaidukov.[5] However, it seems that this behavior is due to too crude an analysis of the Hall voltage in these early experiments,[7] and it is most likely that the actual variation of $R(H)$ is a smooth one. We briefly note that in the high-field regime $(g\mu_B H/kT \gtrsim 10)$ the magnetization is saturated and the behavior of $R(H)$, as well as that of $\rho(H)$, is given by $\ln H$ terms in the scattering amplitudes. Further details of this high-field behavior will be given elsewhere.[8]

B. Experiment

Here we shall compare our calculation with recent experiments on CuMn alloys.[7] We have chosen the CuMn system for this comparison for the following reasons. CuMn has a low Kondo temperature $(T_K < 0.1\,°K)$ much below the temperature $T = 1.2\,°K$, where we make the comparison, so we expect perturbation theory to be valid; second, Mn is most likely the transition impurity which exhibits the smallest degree of anisotropy in its potential
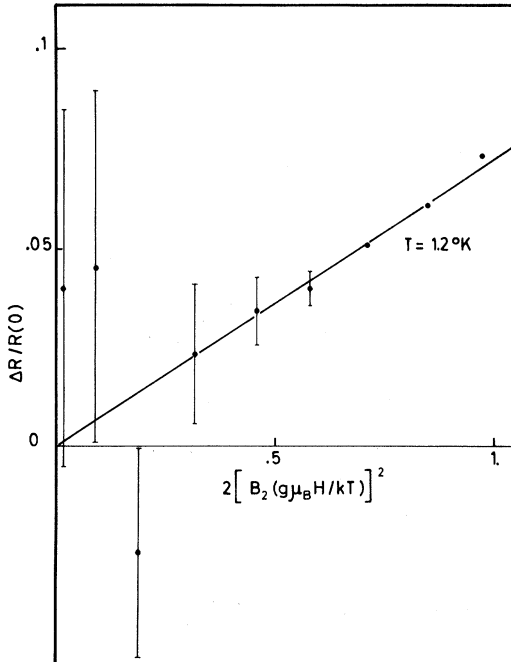

FIG. 3. Experimental points of Ref. 7 for $\Delta R/R(0)$ at $T = 1.2\,°K$ plotted against twice the square of the Brillouin function for spin 2. The straight line corresponds to the theoretical prediction of Eq. (15).

scattering, so that the spin effect we are studying in this paper is expected to be the dominant contribution to $R(H)$ in CuMn alloys. The above anisotropy, which we will call the nonspin effect, is assumed to be temperature independent, so that it is the only contribution to the field dependence of the Hall coefficient at temperatures where the spin effects become negligible (say, $20\,°K$ for fields less than 30 kG). We have made the assumption, as in our magnetoresistivity analysis,[3] that the spin effect and the nonspin effect are additive. Hence, we have subtracted the nonspin field dependence measured at $20.2\,°K$ [and identified with $R(H=0)$] from the raw experimental data at $T = 1.2\,°K$ to obtain the $R_{spin}(H)$ we compare with our Eqs. (8) and (10).

Both Eqs. (8) and (10) imply that for $S = 2$,

$$\frac{\Delta R}{R(0)} \propto \left[B_2\left(2\frac{g\mu_B H}{kT}\right)\right]^2 . \tag{16}$$

The coefficient of proportionality depends on $(J/V)^2$ and, in Eq. (8), J/ϵ_F and $\ln(kT/4\epsilon_F)$. In Fig. 3 we have plotted the experimental points[7] for a CuMn alloy of 150 ± 10 ppm Mn at $T = 1.2\,°K$, in fields between zero and 15 kG, vs $[B_2(2g\mu_B H/kT)]^2$. The straight line is the expected theoretical behavior. The agreement is reasonable, considering the nature of the approximation and the scatter of the experimental points. We have used the slope of this straight line in Fig. 3 to obtain a value of $|J|/V$. However, as we have already pointed out,[3] perturbative expressions such as Eqs. (8) and (10) can only give order-of-magnitude estimates of $|J|/V$ and cannot be expected to agree very well with each other except in order of magnitude. This conclusion has been borne out by the fits we have made here: Eq. (10) gives $|J|/V \simeq 0.1$. We have used this value together with $J = -0.12$ eV to draw our theoretical curves in Fig. 2. Considering the crudeness of the analysis, these rough estimates agree surprisingly well with the value of $|J|/V \simeq 0.16$ obtained from a similar analysis of the negative magnetoresistivity of CuMn alloys.[3]

The actual experimental points at $T = 1.2\,°K$ are also shown in Fig. 2. Although the high-field behavior has been discussed elsewhere,[8] let us note an important point here: The high-field behavior of the experimental data, for $g\mu_B H/kT > 1$, cannot be compared with our theory, for, at these fields and for that impurity concentration, $\omega_c \tau$ is no longer $\ll 1$. Hence, our simplified model ignoring Fermi-surface effects, which is reasonable at low fields, is not valid in this region.[11]

In conclusion, we would like to point out that the encouraging agreement between experiment and theory we have found in the low-field region suggests that systematic measurements of the Hall coefficient, the negative magnetoresistivity, and

the magnetization on the same sample will be most useful in verifying our suggestion that $R(H) - R(0)$ and $\rho(H) - \rho(0)$ are proportional to each other and to the square of the magnetization.[11]

ACKNOWLEDGMENTS

We are grateful to Dr. P. Monod and A. Fried-
rich for providing us with some of their results prior to publication. We have enjoyed fruitful discussions with Dr. H. Suhl, Dr. R. More, and Dr. C. Hurd on both the theoretical and experimental aspects of this work. We also thank Dr. D. Schiff for his assistance in the computations.

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Temperature-Dependent Scattering in Paramagnetic PdCo Alloys*

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(Received 15 September 1970)

Measurements are presented of the resistivities of some dilute PdCo alloys at temperatures above the magnetic-ordering temperature. The excess resistivity $\Delta\rho$ can be represented by the expression $\Delta\rho = A + B \ln T$, where B is positive and is proportional to the Co concentration. Magnetoresistance measurements on one of the alloys are also presented. It is shown that an implausibly large positive value for the exchange coupling between s electrons and local spins is required to account for the magnetoresistance and zero-field measurements if the temperature dependence is assumed to result from a Kondo scattering of the s electrons from local Co moments. We conclude that the mean-square moment on the impurity site may be temperature dependent, due either to partial spin compensation by the itinerant d electrons or localized spin fluctuations on the Co sites, and that this leads to a temperature-dependent scattering of the s electrons and the resistivity behavior which we observe.

INTRODUCTION

It is well known that the alloy systems PdFe and PdCo which exhibit the giant-moment phenomenon[1-4] also exhibit ferromagnetic ordering below a Curie temperature T_C, which increases rapidly with Fe or Co concentration.[5] Many of the properties of these systems show anomalous features, but perhaps none more striking than the electrical resistivity for which a sharp change in slope occurs in the neighborhood of T_C; below T_C the resistivity decreases rapidly as the temperature is further reduced.[6-9] At temperatures above the Curie temperature, in the PdFe system, the incremental resistivity $\Delta\rho(T) = \rho_{alloy}(T) - \rho_{Pd}(T)$ in this paramagnetic region is almost temperature independent below 10 K, but in the PdCo system[9] a substantially larger temperature dependence is observed.[10] In this paper we examine the temperature dependence of $\Delta\rho(T)$ above the Curie temperature in greater detail, in PdCo alloys containing 0.05-, 0.1-, and 0.2-at. % Co. Alloys of higher Co concentration were not included in this investigation as their Curie temperature falls in a temperature region where the "pure" Pd resistivity is increasing rapidly, so that uncertainties resulting from the breakdown of