Negative Magnetoresistivity in Dilute Alloys*

M.-T. BÉAL-MONOD† AND R. A. WEINER
Department of Physics, University of California, San Diego, La Jolla, California

(Received 25 October 1967)

The negative magnetoresistivity of dilute alloys containing magnetic transition impurities is calculated in the second Born approximation using an *s-d* exchange model. Physically the variation of the magnetoresistivity is the product of: (a) the field and temperature dependence of the conduction-electron scattering amplitudes, (b) the freezing out of the impurity's spin degree of freedom by the magnetic field. In zero field, the former contribution leads to the well-known Kondo logarithmic series in T, whereas the latter remains constant in temperature. But in the presence of a magnetic field and for $g\mu_B H/k_B T \lesssim 2$, the freezing out of the spins, mainly described by the square of the magnetization, varies, and much more rapidly than the perturbation expansion of the scattering amplitudes. This is verified experimentally in *Cu*Mn alloys (for $T \gg T_{\text{Kondo}}$), and allows us to phenomenologically extend our results to $T \approx T_{\text{Kondo}}$, for which we get the same good fit with experiments in *Cu*Fe alloys. For $g\mu_B H/k_B T \gtrsim 4$, the impurity spins are completely aligned with the field; the scattering amplitudes become the main source of variation in the magnetoresistivity. In this case, as in absence of field, an exact theory in needed.

I. INTRODUCTION

AFTER some comments on previous work on the same subject, we calculate the effect of a parallel[1] magnetic field on the low-temperature resistivity of dilute alloys such as *Cu*Mn and *Cu*Fe, showing a Kondo[2] resistivity anomaly. Our calculation is just a third-order perturbation expansion of the *s-d* exchange Hamiltonian, and is therefore, in principle, restricted to temperatures much greater than the Kondo temperature T_K. Work is in progress on a calculation using Suhl's scattering-matrix methods[3] which will be valid at all temperatures and magnetic fields.[4]

Although the *s-d* exchange model is known to be incorrect for transition impurities, it has been shown[5] that the Anderson model, which describes them correctly in terms of virtual *d* bound states, leads to the same anomalous Kondo behavior as the truly bound spins involved in the *s-d* exchange interaction. However, calculations of the resistivity in the Anderson model are extremely complicated even in zero field,[6] and consequently we have worked with the *s-d* exchange model to obtain explicit results.

We shall assume the impurity concentration to be small enough to neglect completely the effects of internal fields due to correlations between the impurities. Our last section will briefly comment on the internal-field case in the absence of an external field. The resistivity can then be calculated by considering the scattering of the electrons from one impurity located at the origin; then the total impurity resistivity (we will drop, in the following, the phonon part) will be obtained by multiplying the result by the number of impurities.

The unperturbed Hamiltonian is

$$\mathcal{H}=\sum_{\mathbf{k},\sigma}\epsilon_{\mathbf{k}}a_{\mathbf{k},\sigma}^{\dagger}a_{k,\sigma}-2\mu_B H\sum_{\mathbf{k},\sigma,\sigma'}a_{\mathbf{k},\sigma}^{\dagger}s_{\sigma,\sigma'}{}^{z}a_{\mathbf{k},\sigma'}-g\mu_B HS_z, \tag{1}$$

where $a_{\mathbf{k}\sigma}^{\dagger}$ ($a_{\mathbf{k}\sigma}$) is the creation (destruction) operator for an electron of momentum $\mathbf{k}$ and spin component σ in the z (field) direction, and $\epsilon_{\mathbf{k}}$ is its kinetic energy. The second and third terms represent the Zeeman energies of the electron and impurity, respectively; H is the magnetic field, μ_B the Bohr magneton, and g and 2 the g values of the impurity and the conduction electron. S_z is the component along the z axis of the impurity spin $\mathbf{S}$. The perturbing Hamiltonian is

$$\mathcal{H}'=(V/N)\sum_{k,k',\sigma}a_{k\sigma}^{\dagger}a_{k'\sigma}-2(J/N)\sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'}a_{\mathbf{k}'\sigma'}^{\dagger}\mathbf{s}_{\sigma'\sigma}\cdot\mathbf{S}a_{\mathbf{k}\sigma}, \tag{2}$$

where $\mathbf{s}$ and $\mathbf{S}$ are the spins of the conduction electron and the impurity, respectively, and N the number of atoms of the crystal.

For $g\mu_B H/k_B T<2$, the most rapidly varying part of the negative magnetoresistivity is caused by the freezing out of spin-flip scattering due to the aligning of the impurity spins by the magnetic field. That is, consider the following spin-flip scattering process: initial state of an electron of momentum $\mathbf{k}_i$, spin down, and impurity with spin component M_S, going into the final state of an electron of momentum $\mathbf{k}_f$, spin up, and impurity spin component M_S-1. In the simplest case, $g=2$, energy conservation gives $|\mathbf{k}_f|=|\mathbf{k}_i|$, so for electrons initially within $k_B T$ of the Fermi surface (the only ones which contribute to the resistivity) the final-state spin-up electron has a total energy (kinetic+Zeeman) less than ϵ_F by at least $2\mu_B H-k_B T$, and this scattering process is forbidden by the exclusion principle. Since the field tends to align the impurity spins

* Research sponsored by the Air Force Office of Scientific Research, Office of Aerospace Research, U.S. Air Force, under Grant No. AF-AFOSR-610-67.

† Permanent address: Physique des Solides, Faculté des Sciences, 91, Orsay, France.

[1] We ignore the usual positive magnetoresistivity due to the $\mathbf{v}\times\mathbf{H}$ term in the Boltzmann equation ($\mathbf{v}$ is the velocity of the conduction electrons).

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along the positive z direction, more of the spin-flip processes are of this forbidden electron-spin-down into electron-spin-up type than of the spin-up into spin-down type allowed by the exclusion principle, and the total contribution of the spin-flip scattering amplitudes to the resistivity is decreased. This is a much more important source of the variation of the resistivity with field (or temperature at fixed field) than the field and temperature dependence of the scattering amplitudes themselves, at least until the spins are completely aligned. In this regime the leading term in the magnetoresistivity is then given by the square of the magnetization M^2 to both second and third order in $\mathcal{H}'$.[7] An extension of our results for temperatures close to T_K can then be made (for $g\mu_B H/k_B T$ sufficiently small) by replacing the Brillouin function for M by the experimental magnetization. Since it appears experimentally[8,9] that M is not given by a Brillouin function of H/T, and, on the other hand, that theoretical calculations of M exhibit the same kind of logarithmic behavior in T as the resistivity,[10,11] this extension should include the most important part of the Kondo effect in the negative magnetoresistivity by correctly accounting for the freezing out of the spin-flip scattering.

Our results are compared with experimental data[12] for both $T\gg T_K$ (CuMn) and $T\approx T_K$ (CuFe). In both cases, fairly good fits can be made and estimates are obtained for the effective values of the exchange integral J. These estimates should be taken only as order-of-magnitude results, since the perturbation series is not rapidly converging in CuMn, and we use rough approximations in fitting CuFe. The case of high $g\mu_B H/k_B T$ is briefly discussed.

II. COMMENTS ON PREVIOUS CALCULATIONS OF THE MAGNETORESISTIVITY

A. Second-Order Calculations

We would like to make several remarks concerning previous calculations of the resistivity to second order in $\mathcal{H}'$.[7,13] As was shown in these papers, the magnetic field appears in the transport equations via the equilibrium distribution of the spin states of the impurity as well as in shift of the fermi distributions to $f(\epsilon_k \mp \mu_B H)$. This leads to relaxation times for spin up and down, $\tau_\pm$, which are energy-dependent through ϵ_k and $f(\epsilon_k)$:

$$\tau_\pm^{-1} \equiv \tau_\pm^{-1}(\epsilon_k, f(\epsilon_k)). \tag{3}$$

Therefore, because of the presence of $f(\epsilon_k)$, which is rapidly varying at the Fermi energy ϵ_F, it is incorrect to write directly, as has been done before,[5,10] ρ_H proportional to $\tau_{\epsilon_F}^{-1}$ with

$$\tau_{\epsilon_F}^{-1} = \tau^{-1}(\epsilon_F, f(\epsilon_F)) = \tau^{-1}(\epsilon_F, \tfrac{1}{2}). \tag{4}$$

Rather, one has to use $\tau_\pm^{-1}$ given by Eq. (3) to calculate the conductivities $\sigma_\pm$ from

$$\sigma_\pm = -\frac{e^2}{6\pi^2 m}\int k^3 \tau_\pm \frac{\partial f(\epsilon_\pm)}{\partial \epsilon_\pm}\, d\epsilon_\pm, \tag{5}$$

$$\epsilon_\pm = \epsilon_k \mp \mu_B H, \tag{6}$$

where m and e are the mass and charge of the electron, respectively. The energy integrals involved in Eq. (5) can be done exactly, with the total conductivity given by

$$\sigma = \sigma_+ + \sigma_- \tag{7}$$

and the resistivity by

$$\rho_H = \sigma^{-1}. \tag{8}$$

For the simple case of $g\mu_B H/k_B T \ll 1$, for example, while the calculations using Eq. (4) find[7,13]

$$\frac{\rho_H}{\rho_{H=0}} \simeq 1 - \left(\frac{g\mu_B H}{k_B T}\right)^2 \frac{J^2 S(S+1)}{9[V^2 + J^2 S(S+1)]^2} \times \{[\tfrac{3}{2} + 4S(S+1)]V^2 + \tfrac{3}{2}J^2 S(S+1)\}, \tag{9}$$

the correct second-order result is

$$\frac{\rho_H}{\rho_{H=0}} \simeq 1 - \left(\frac{g\mu_B H}{k_B T}\right)^2 \frac{J^2 S(S+1)}{9[V^2 + J^2 S(S+1)]^2} \times \{[1 + 4S(S+1)]V^2 + \tfrac{4}{3}J^2 S(S+1)\}. \tag{10}$$

The discrepancies resulting from such approximations are much more important in the third-order calculation.

B. Third-Order Calculations

A recent paper by Harrison and Klein[14] contains a third-order perturbation calculation of ρ_H for the case $|J| \ll V$. Although the transition probabilities are calculated correctly, their result for ρ_H is incorrect because of several numerical errors and a more fundamental error in the way they handle the expansion in J/V. They ignore the cross term in $(V - 2J\mathbf{s}\cdot\mathbf{S})^2$, which is in fact the largest contribution to the magnetoresistivity. The present paper will follow essentially the same procedure as HK but (we hope) with fewer errors and with some generalizations.

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[14] R. J. Harrison and M. W. Klein, Phys. Rev. **154**, 540 (1967); we will henceforth refer to this paper as HK.

III. CALCULATION OF THE MAGNETORESISTIVITY

The relaxation times $\tau_\pm$ to third order in $\mathcal{H}'$ are given by

$$\frac{1}{\tau_\pm}=\frac{kmv_0c}{\pi\hbar^3}\Big\{V^2\mp 2VJ\langle S_z\rangle+J^2\langle S_z^2\rangle + J^2\frac{S(S+1)-\langle S_z^2\rangle\mp\langle S_z\rangle}{1-f_\pm(1-e^{\mp\alpha})} + [\mp 4VJ^2\langle S_z\rangle+4J^3\langle S_z^2\rangle]g^\mp(\epsilon_\pm\pm g\mu_BH) + 2J^3[S(S+1)-\langle S_z^2\rangle\mp\langle S_z\rangle] \times\frac{g^\mp(\epsilon_\pm\pm g\mu_BH)+g^\pm(\epsilon_\pm)}{1-f_\pm(1-e^{I\alpha})}\Big\}, \quad (11)$$

where v_0 is the atomic volume of the host metal, k the wave number of the conduction electron, and c the impurity concentration. $\langle S_z\rangle$ and $\langle S_z^2\rangle$ are the equilibrium averages of the operators S_z and S_z^2 at the temperature T:

$$\langle S_z\rangle=\sum_{M_S=-S}^{+S}P_{M_S}M_S,$$

$$\langle S_z^2\rangle=\sum_{M_S=-S}^{+S}P_{M_S}M_S^2,$$

$$P_{M_S}=e^{M_S\alpha}/\sum_{M_S=-S}^{+S}e^{M_S\alpha},$$

$$\alpha=g\mu_BH/k_BT. \quad (12)$$

M_S is the expectation value of S_z.

We note the useful relation

$$\langle S_z^2\rangle=S(S+1)-\langle S_z\rangle\coth\tfrac{1}{2}\alpha. \quad (13)$$

The Fermi distribution $f_\pm$ is

$$f_\pm=[\exp((\epsilon_\pm-\epsilon_F)/k_BT)+1]^{-1}, \quad (14)$$

with $\epsilon_\pm$ given by Eq. (6). The functions $g^\pm(\epsilon)$ are given by

$$g^\pm(\epsilon)=N^{-1}\sum_q\frac{f_\pm(q)}{\epsilon_{q\pm}-\epsilon} =\frac{3z}{2\epsilon_F}\left\{1+\tfrac{1}{2}\ln\left(\frac{k_BT}{2\epsilon_F}\right)-\tfrac{1}{2}I^\pm\left(\frac{\epsilon}{k_BT}\right)\right\}, \quad (15)$$

$$I^\pm(x)=\int\frac{\partial f(\epsilon')}{\partial\epsilon'}\ln\left|\frac{\epsilon'}{2k_BT}-\tfrac{1}{2}x\right|d\epsilon', \quad (16)$$

where z is the number of conduction electrons per atom. These $g(\epsilon)$ integrals give the contributions of the intermediate states responsible for the anomalous behavior of the resistivity.

We have dropped, as Kondo[2] and HK do, the terms in the third-order part of Eq. (11) which do not involve integrals over the Fermi distribution:

$$h^\pm(\epsilon)=N^{-1}\sum_q(\epsilon_{q\pm}-\epsilon)^{-1}. \quad (17)$$

To make such terms converge we would have to take into account the energy dependence of V and J, but then the problem would be practically impossible to solve explicitly. Whereas these terms were temperature-independent in the $H=0$ case, here their contribution to the resistivity depends on the temperature through their product with $\langle S_z\rangle$ or $\langle S_z^2\rangle$. However, they do not contribute to the $\ln T$ terms and their omission causes only slight changes in the coefficients of $\langle S_z\rangle$ and $\langle S_z^2\rangle$.

The scheme we use for approximate calculations of the resistivity is to write, from Eq. (11),

$$1/\tau_\pm=(kmcv_0/\pi\hbar^3)[\text{second order}+\text{third order}]. \quad (18)$$

Then, on the assumption that the third-order term is much smaller than the second-order term, we expand $\tau_\pm$ as

$$\tau_\pm\simeq\frac{\pi\hbar^3}{kmcv_0}\frac{1}{\text{second order}}\left[1-\frac{\text{third order}}{\text{second order}}\right], \quad (19)$$

which enables us to do the integrals in Eq. (5) without too much difficulty in the limiting cases given below.

A. $|J|\ll V$, Arbitrary H and T

This is the case studied by HK. We expand Eq. (19) in powers of J/V to order $(J/V)^2J/\epsilon_F$. From the first term of Eq. (19) we obtain three terms in $(J/V)^2$: two from the two J^2 terms in Eq. (11) and another one from the square of the VJ term of Eq. (11). Similarly, among the $(J/V)^2J/\epsilon_F$ terms in Eq. (19) we obtain a cross term between the VJ and VJ^2 parts of Eq. (11). It is these last two terms which have been omitted in HK and which give the largest contribution to the magnetoresistivity (the $4\langle S_z\rangle^2$ term below).

To complete the calculation, we need the following integrals:

$$\int\frac{\partial f(\epsilon)}{\partial\epsilon}\frac{1}{1-f(\epsilon)(1-e^{\mp\alpha})}d\epsilon=-\tfrac{1}{2}\alpha\frac{e^{\pm\alpha/2}}{\sinh(\frac{1}{2}\alpha)},$$

$$\int\frac{\partial f(\epsilon)}{\partial\epsilon}I^\mp\left(\frac{\epsilon}{k_BT}\pm\alpha\right)d\epsilon=I_1(\alpha),$$

$$\int\frac{\partial f(\epsilon)}{\partial\epsilon}\frac{I^\mp((\epsilon/k_BT)\pm\alpha)}{1-f(\epsilon)(1-e^{\mp\alpha})}d\epsilon =\int\frac{\partial f(\epsilon)}{\partial\epsilon}\frac{I^\pm(\epsilon/k_BT)}{1-f(\epsilon)(1-e^{\mp\alpha})}d\epsilon =e^{\pm\alpha/2}\tfrac{1}{2}\alpha\frac{I_2(\alpha)}{\sinh(\frac{1}{2}\alpha)}. \quad (20)$$

The functions $I_1(\alpha)$ and $I_2(\alpha)$ are the same as those in HK. They are computed numerically and shown in

Fig. 1. We agree on their asymptotic expansion for $|\alpha|<2$:

$$I_1(\alpha)\simeq -0.432+0.091\alpha^2,$$
$$I_2(\alpha)\simeq -0.432+0.030\alpha^2, \quad (21)$$

but we find the following for $|\alpha|>10$:

$$I_1(\alpha)\simeq \ln|\alpha/2|-(3.27/\alpha)+\cdots,$$
$$I_2(\alpha)\simeq \ln|\alpha/2|-1+(3.27/\alpha^2)+\cdots. \quad (22)$$

In terms of I_1, I_2, α, and $\langle S_z\rangle$ the resistivity in the presence of the magnetic field is

$$\rho_H=\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0\left\{V^2+J^2[S(S+1)-A(\alpha)]+\frac{3zJ^3}{\epsilon_F}\left[(S(S+1)-A(\alpha))\times\left(1.568+\ln\frac{k_BT}{2\epsilon_F}\right)+B(\alpha)\right]\right\}, \quad (23)$$

where

$$A(\alpha)=4\langle S_z\rangle^2+\langle S_z\rangle(\coth\tfrac{1}{2}\alpha-\tfrac{1}{2}\alpha[1/\sinh^2(\tfrac{1}{2}\alpha)]),$$

and

$$B(\alpha)=[S(S+1)-4\langle S_z\rangle^2-\langle S_z\rangle\coth\tfrac{1}{2}\alpha]\times[I_1(\alpha)-I_1(0)]+\tfrac{1}{2}\alpha[\langle S_z\rangle/\sinh^2(\tfrac{1}{2}\alpha)]\times[I_2(\alpha)-I_2(0)], \quad (24)$$

and where we have used Eq. (13) to simplify the expressions for A and B. Equations (23) and (24) replace Eq. (2.27) of HK. Subtracting $\rho_{H=0}$, we obtain the negative magnetoresistivity $\Delta\rho$ as

$$\Delta\rho=\rho_H-\rho_{H=0}=-\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0J^2\left\{A(\alpha)\left[1+\frac{3Jz}{\epsilon_F}\left(1.568+\ln\frac{k_BT}{2\epsilon_F}\right)\right]-\frac{3Jz}{\epsilon_F}B(\alpha)\right\}. \quad (25)$$

For $\alpha\ll 1$ this becomes

$$\Delta\rho\simeq-\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0\frac{\alpha^2}{q}J^2S(S+1)\left\{1+4S(S+1)+\frac{3zJ}{\epsilon_F}\left[(1+4S(S+1))\ln\frac{k_BT}{2\epsilon_F}+6.27S(S+1)+1.11\right]\right\}, \quad (26)$$

while for $\alpha>10$

$$\rho_H\simeq\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0\left\{V^2+J^2[S(S+1)-4\langle S_z\rangle^2-\langle S_z\rangle]\times\left[1+\frac{3J_z}{\epsilon_F}\left(2+\ln\frac{g\mu_BH}{4\epsilon_F}\right)\right]\right\}. \quad (27)$$

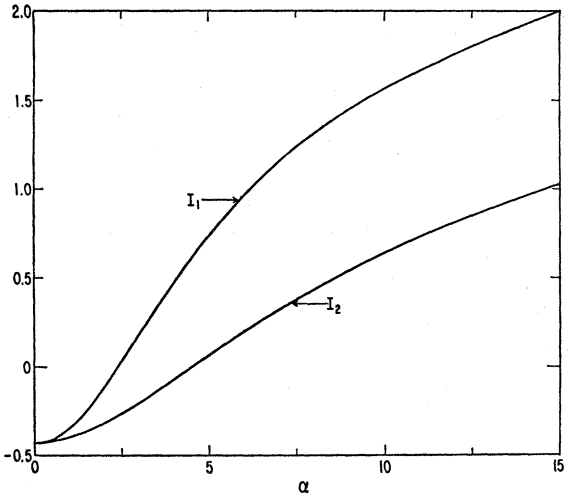

FIG. 1. The functions $I_1(\alpha)$ and $I_2(\alpha)$ versus α.

The $\ln H$ terms appearing here reflect Kondo-type anomalous behavior in the applied field[11] when all the spins are aligned by the magnetic field, so that $\langle S_z\rangle$ is constant. Presumably an exact theory[4] is needed to explain correctly such large-α behavior, just an exact theory is needed for the low-T behavior in absence of field.

B. $g\mu_BH/k_BT\ll 1$, Arbitrary V and J

In the limit of high temperatures or small magnetic fields, we can expand Eq. (19) in powers of α for arbitrary V and J. To order α^2, we need the following integrals:

$$\int\frac{\partial f(\epsilon)}{\partial\epsilon}I^{\mp}\left(\frac{\epsilon}{k_BT}\pm\alpha\right)d\epsilon=-0.432+0.091\alpha^2,$$
$$\int\frac{\partial f(\epsilon)}{\partial\epsilon}f(\epsilon)I^{\mp}\left(\frac{\epsilon}{k_BT}\pm\alpha\right)d\epsilon=-0.216\mp0.091\alpha,$$
$$\int\frac{\partial f(\epsilon)}{\partial\epsilon}f^2(\epsilon)I^{\mp}\left(\frac{\epsilon}{k_BT}\right)d\epsilon=-0.114, \quad (28)$$

and

$$\langle S_z\rangle\simeq S(S+1)\alpha/3. \quad (29)$$

The result for general V and J is very complicated, so we will not write the general formula. However, the interest of this small-α expansion is that we can, then, compare two simple extreme cases: $|J|\ll|V|$ and $|V|\ll|J|$. Since this ratio is not experimentally well determined, our calculation allows us to know which of these orders of magnitude for $|J|/|V|$ corresponds to the physical situations. By comparison with experiments for $\Delta\rho$ we can obtain an order of magnitude for $|J|$ in both limits. Then a comparison with the V's obtained from $\rho_{H\,\min}$ at higher temperatures ($g\mu_BH/k_BT\simeq 0$), using the above J's, should enable us to know which order of magnitude of $|J|/|V|$ is con-

sistent with the hypothesis from which we started. Since we use only a truncated perturbation series, these estimates are obviously subject to considerable error; consequently the values obtained for $|J|$ and $|V|$ must be definitely regarded as rough order-of-magnitude estimates. But as long as one of the two extreme cases seems to reflect the experimental situation, the order of magnitude of the ratio $|J|/|V|$ should be correctly estimated.

For $|J|\ll|V|$ the small-α limit for the magnetoresistivity is given by Eq. (26). For $|V|\ll|J|$ [15] we have

$$\Delta\rho=-\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0\frac{J^2S(S+1)\alpha^2}{27}\times\left[4+\frac{3Jz}{\epsilon_F}\left(4\ln\frac{k_BT}{2\epsilon_F}+3.48\right)\right]. \quad (30a)$$

As an example of an intermediate case for the $|V|/|J|$ ratio, the result for $|V|/|J|$ is given in Eq. (30b):

$$\Delta\rho=-\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0\frac{J^2S(S+1)\alpha^2}{9[1+S(S+1)]^2}\left\{1+\tfrac{19}{3}S(S+1)+\tfrac{136}{3}S^2(S+1)^2+\frac{3zJ}{\epsilon_F}\left(\ln\frac{k_BT}{2\epsilon_F}\right)\times[1+\tfrac{20}{3}S(S+1)+\tfrac{4}{3}S^2(S+1)^2]+\frac{6zJ}{\epsilon_F}[0.559+4.717S(S+1)+0.580S^2(S+1)^2]\right\}. \quad (30b)$$

We note also that for fixed J/ϵ_F the magnitude of the magnetoresistivity decreases with V and that the ratio of the $\ln T$ term to the first Born term appears to be smallest for $|V|\approx|J|$, for all values of $S\geq 1$.

IV. COMPARISON WITH EXPERIMENTS

We will restrict ourselves to comparison with those of the experimental data[12] which are obtained in a fairly large range of temperatures and fields, for concentrations low enough to consider the internal fields as unimportant compared with the external field, and where the negative magnetoresistivity is reasonably well separated from its positive part. Other magnetoresistivity measurements can be found in Ref. 12.

A. *Cu*Mn Alloys

We will draw graphs and speak in terms of $\rho_H-\rho_{H=0}=\Delta\rho$ and not in terms of the full ρ_H, because, experimentally, the actual measured quantities are $\rho_{H=0}$ and $\Delta\rho$, and not ρ_H directly.

The experimental data[12] on the negative magnetoresistivity in *Cu*Mn alloys have been obtained in the temperature range 1.4–9.3°K. From previous estimates of J and the behavior of the zero-field resistivity, T_K seems to be of the order of a few tenths of a degree Kelvin,[16] so these alloys appear to offer a significant test of our perturbation results. Using the procedures described in Sec. IV B, a rough estimate gives $|J|/|V|\simeq 0.16$, so we have used Eq. (25) to fit the data. We have tried two fits, using Brillouin functions for $\langle S_z\rangle$,[17] by keeping, first, only the J^2 part of Eq. (25), and second, with both the J^2 and J^3 parts of Eq. (25). In the first case we used $J\simeq-0.33$ eV, in the second case $J\simeq-0.24$ eV. Both fits are fairly good (within 10%) with a barely perceptible improvement in the second case. Figure 2 shows the variation of $\Delta\rho$ versus T in both cases. The fit of the second case implies $J<0$.

The most striking feature of these results is that both the first and second Born approximations for the electron lifetimes can give equally good fits to the negative magnetoresistivity. This indicates that most of the observed temperature and field variation of the magnetoresistivity is not due to the temperature and field variation of the scattering amplitudes, but rather is caused by the freezing out of spin-flip scattering as H/T is increased, as explained in the Introduction. Since the same factor, $A(\alpha)$ in Eq. (25) accounts for this freezing out in both the first and second Born approximations,[18] the fact that both will give a good fit to the experimental results indicates that the freezing out is by far the dominant contribution to the magnetoresistivity in the temperature and field range of the experiments. At this point, we note that the main contribution to $A(\alpha)$ is the $4\langle S_z\rangle^2$ factor which is proportional to the square of the magnetization M^2. Therefore, one conclusion is that for $g\mu_BH\lesssim 2k_BT$, $\Delta\rho$ is essentially proportional to M^2 and this is verified experimentally. This is true as long as the magnetization has not yet saturated; once it does saturate ($\alpha>4$), the freezing-out factors will cease to change and the variation of the negative magnetoresistivity will be given by that of the scattering amplitudes. We expect that this would be correct even if M is not given by a thermal-equilibrium spin distribution.

One further point must be made in connection with our estimate of J from the second Born fit to experiment. Even at these high temperatures (10 to 100 times T_K) and very small J/ϵ_F ($\simeq-0.04$), the J^3 term

[15] The $|V|\ll|J|$ case is extremely difficult for arbitrary α, due to the presence of the $[1-f_\pm(1-e^{\mp\alpha})^{-1}]$ terms which must be kept in the denominator in the expansion in powers of V/J. Fortunately it seems that this case has not yet occurred experimentally, since in both *Cu*Mn and *Cu*Fe alloys we find that $|J|\ll|V|$ is the case consistent with experiments.

[16] M. D. Daybell and W. A. Steyert, Rev. Mod. Phys. (to be published).

[17] In this temperature and field range, the Kondo effect does not cause significant shifts in the magnetization from a Brillouin function with $S=2$. This can be seen in the results of Refs. 8 and 12 which lead to a susceptibility $\chi=g^2\mu_B^2S(S+1)/3k_B(T+T')$ with $T'=0.2\pm0.1$°K.

[18] The $B(\alpha)$ term in Eq. (25), which includes both the effects of freezing out and changes in the scattering amplitudes due to the application of the field, is completely negligible for $\alpha<4$.

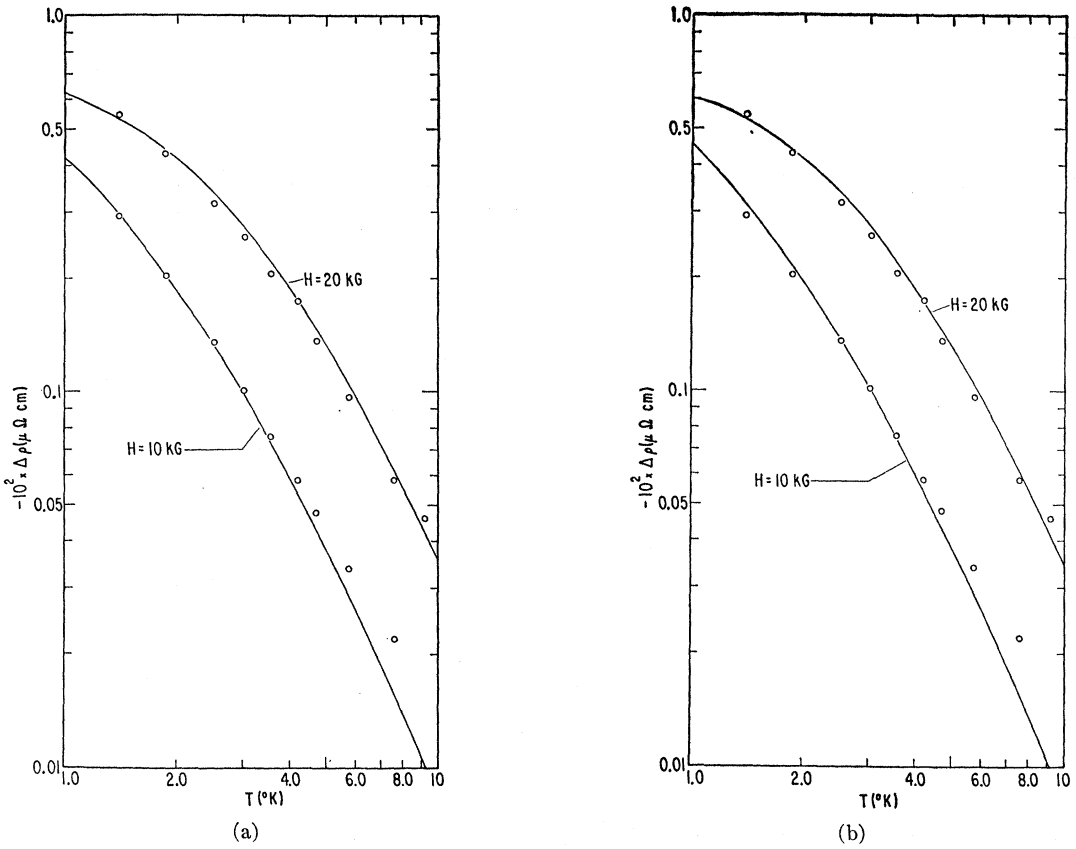

FIG. 2. Negative magnetoresistivity $-\Delta\rho$ as a function of temperature T in CuMn ($c=75$ ppm) for $H=10$ and 20 kg. The smooth curves are the theoretical curves; the open circles are the experimental points (Ref. 12). (a) Theoretical results given by the first Born approximation, $J=-0.33$ eV; (b) theoretical results given by the first and second Born approximations, $J=-0.24$ eV.

made as large a contribution to the magnetoresistivity as the J^2 term. That is, the perturbation series is not converging rapidly and the value of J we have obtained indicates nothing more than the order of magnitude of J. This is one of the dangers of doing perturbative expansions in a theory with $\ln T$ anomalies: The $\ln T$ is so slowly varying that even far above the critical temperature associated with the logarithm, its value is large enough to require summing all orders of perturbation theory, as was pointed out by Suhl and Wong.[19] The only reason we have obtained reasonably good fits to the experimental results is that the $\ln T$ terms contribute very little to the magnetoresistivity temperature and field variation in the region of the experimental temperatures and fields; in other words $\ln T$ varies less rapidly with respect to T than $M^2 \propto 1/T^2$ for small α.

B. *Cu*Fe Alloys

The magnetoresistivity measurements for CuFe alloys are at temperatures between 1.4 and 7°K.[12] Unfortunately, the estimates of T_K [9,20] are spread between 5 and 16°K, and our results cannot, in principle, be used in this temperature region. However, as we have seen in the CuMn alloys, the most important contribution to the negative magnetoresistivity for the experimental temperatures and fields is due to the freezing out of spin-flip scattering; the temperature and field dependence of the scattering amplitudes, for which exact results are needed for $T \approx T_K$, have little effect on a fit to the experiments. We have therefore made a rough extension of our perturbation results to $T \approx T_K$.

Using the procedures of Sec. III B, we find $|J|/|V| \simeq 0.2$. Rewriting Eq. (25) in terms of the magnetization M (in Bohr magnetons per atom),

$$M = g\langle S_z \rangle \tag{31}$$

(we shall take $g=2$ in the following), we can express the freezing-out factor $A(\alpha)$ in Eq. (25) as

$$A(\alpha) = M^2\left(1+(1/2M)\left\{\coth\tfrac{1}{2}\alpha - \left[\alpha/2\sinh^2(\tfrac{1}{2}\alpha)\right]\right\}\right). \tag{32}$$

For $\alpha \leq 1$ the second term in Eq. (32) is very well approximated by

$$\alpha/6M = \mu_B H/3k_B TM \simeq \mu_B{}^2 N/3k_B T\chi \equiv \mu_B{}^2/\mu_{\text{eff}}{}^2, \tag{33}$$

[19] H. Suhl and D. Y. Wong, Physics **3**, 17 (1967).
[20] C. M. Hurd, Phys. Rev. Letters **18**, 1127 (1967); M. A. Jensen, A. J. Heeger, L. B. Welsh, and G. Gladstone, Phys. Rev. Letters **18**, 997 (1967).

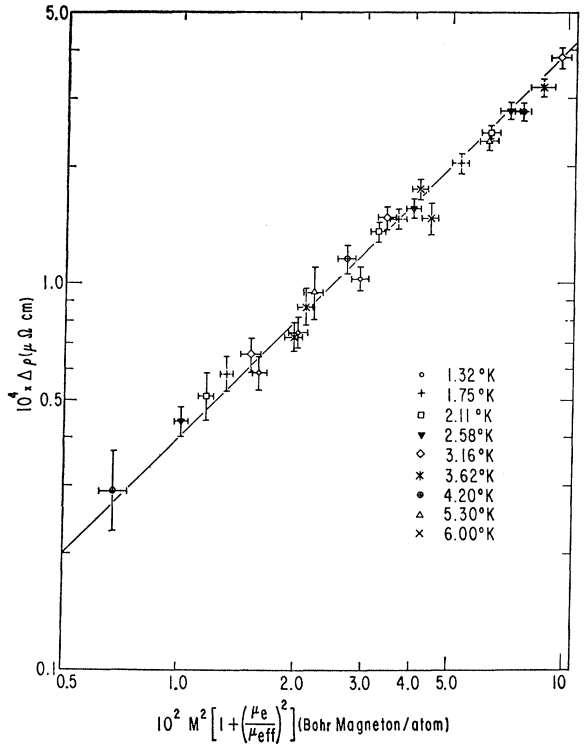

FIG. 3. Negative magnetoresistivity $-\Delta\rho$ as a function of $M^2(1+\mu_B^2/\mu_{\text{eff}}^2)$ in *Cu*Fe ($c=110$ ppm). The experimental points are from Ref. 12. The solid line corresponds to a slope of one (in a ln−ln plot) and is given by the J^2 part of Eq. (34) with $|J|=0.91$ eV.

where χ is the susceptibility and μ_{eff} an "effective" moment defined by (33). Then the expression for the magnetoresistivity is

$$\Delta\rho \simeq -\frac{3\pi}{2\epsilon_F}\frac{m}{e^2\hbar}cv_0J^2M^2\left(1+\frac{\mu_B^2}{\mu_{\text{eff}}^2}\right)\times\left[1+\frac{3J_z}{\epsilon_F}\left(1.568+\ln\frac{k_BT}{2\epsilon_F}\right)\right]. \quad (34)$$

We can reasonably suppose that a possible extension of this formula to $T\approx T_K$ which will take account of the freezing out of the spin-flip scattering is to replace M, which is given by a Brillouin function in Eq. (34), by the experimental magnetization (which deviates significantly from a Brillouin function[8,12]) and to obtain μ_{eff}^2 from the experimental susceptibility by Eq. (33). In this manner we hope to take account of the logarithmic anomalies in the magnetization[17] and through that, their most important effects on the magnetoresistivity.

Since the J^3 terms in Eq. (34) are clearly superfluous in the face of the guesses we have made to extend it to $T\approx T_K$, we have attempted to fit only the J^2 part to the experimental data. The result, with $|J|\approx 0.91$ eV, is given in Fig. 3. Once more, this value for J must be regarded as an order-of-magnitude estimate. Considering the crudeness of the way we have extended the perturbation-theory results, the fit is fairly good.

V. DISCUSSION

The estimates we obtain for $|J|$ in *Cu*Mn and *Cu*Fe indicate that $|J|_{CuFe} > |J|_{CuMn}$ and hence $(T_K)_{CuMn} < (T_K)_{CuFe}$, which seems to be the case experimentally. We do not wish to calculate the T_K corresponding to our values of $|J|$ as these are only order-of-magnitude values, and a change of 10% in J gives a huge change (approximately one order of magnitude) in T_K.

Our second Born result for the magnetoresistivity indicates that there are two ranges of the value of H/T in which specific mechanisms for the H and T behavior of the magnetoresistivity can be separated. The first is $\alpha \lesssim 2$, in which all the observed variation of $\Delta\rho$ is given by the magnetization. In the other range, $\alpha \gtrsim 4$, the magnetization is saturated and all the variation is due to the temperature and field dependence of the scattering amplitudes themselves. In the intermediate region $4 \gtrsim \alpha \gtrsim 2$, both effects are of the same order of magnitude.

All the present experimental data has been obtained in the first region and can be successfully fitted by the first Born approximation to the lifetimes, which adequately account for the freezing out of spin-flip scattering. Experimental data would be welcome at much higher fields[21] and/or lower temperatures to give some indication of the temperature and field variation of the scattering amplitudes. Since the perturbation series converges so slowly, an exact theory[4] is needed in the second region for both $T \gg T_K$ and $T \leq T_K$.

The total resistivity in the presence of a magnetic field is the sum of $\rho_{H=0}$ and of $\Delta\rho$. The negative contribution of $\Delta\rho$ may then introduce a maximum, below the well-known minimum (when the phonon part is added), in ρ_H, as is observed experimentally.[12] Since $|\Delta\rho|$ is proportional to M^2 (for $\alpha \lesssim 2$), $\partial|\Delta\rho|/\partial H$ decreases for increasing fields. Therefore, the difference between the values of the maximum and the minimum decreases for increasing fields.[12] It is reasonable to think that in the very-high-field region the maximum will disappear. This maximum is analogous to the one introduced in $\rho_{H=0}$ by internal fields (for much higher concentrations) in the absence of an external field. In this case, the internal fields due to correlations between the impurities increase with the impurity concentration and the induced maximum of $\rho_{H=0}$ disappears gradually as the concentration increases.[22] We expect that high external

[21] Previous experiments up to 100 kG on both *Cu*Fe and *Cu*Mn [see, Y. Muto, K. Noto, and F. T. Hedgcock, Can. J. Phys. **42**, 15 (1964)] are difficult to analyze quantitatively as the negative magnetoresistivity is not correctly separated from the normal positive magnetoresistivity.

[22] See, for example, a survey of such experiments in G. J. van Den Berg, *Low Temperature Physics* (Plenum Press, Inc., New York, 1965), p. 955.

magnetic fields would, in the same way, make the maximum of ρ_H disappear completely. The high-field experiments suggested above would be able to verify this conclusion.

VI. INTERNAL FIELDS

In the absence of external fields, but when the concentration is large enough so that the correlations between impurities can no longer be neglected, the impurities spins are then subjected to internal fields H_i due to these correlations. We agree with HK that, in principle, the resistivity is then given by the formula

$$\rho = aT^5 + \int_{-\infty}^{+\infty} p(g\mu_B H_i, T)\rho_{Hi}(T)\, d(g\mu_B H_i), \quad (35)$$

where aT^5 is the phonon contribution; $\rho_{Hi}(T)$ should be given by Eq. (23) with $H=H_i$, and $p(g\mu_B H_i, T)$ is a statistical factor[23,24] which describes the distribution of the internal fields H_i. We would like to mention first that the second-order contribution to (35), which HK find to be the main one, already has been studied by one of us[24] with the same approximation that HK has used:

$$p(g\mu_B H_i, T) \simeq p(g\mu_B H_i, 0) \simeq \text{Lorentzian distribution}, \quad (36)$$

and led to the same main conclusion: At very low temperature the resistivity ρ varies linearly with the temperature with a slope independent of the concentration:

$$\rho_{T\to 0} \propto cS[k_B T/\Delta(T=0)], \quad (37)$$

where Δ is the width of the distribution $p(g\mu_B H_i, 0)$:

$$\Delta \propto cS. \quad (38)$$

Therefore

$$\rho_{T/0} \propto \text{const.} \times T. \quad (39)$$

As the expression for $p(g\mu_B H_i, T)$ for $T\neq 0$ was not (and so far is still not) established, we restricted ourselves[23] to study the resistivity in the neighborhood of 0°K. However, our result (39) was sufficient to point out, for the first time, that one could expect a maximum in the resistivity between 0°K and $T_{\min}$, in agreement with experiment. We would like to point out that a detailed investigation of the low-temperature resistivity is complicated for the following reasons: (a) Although the calculation of (35) with the approximation (36) is reasonable for $T \ll T_{\max}$ one has to be careful in calculating the temperature of the maximum $T_{\max}$ by simply differentiating (35) with respect to T, with $p(g\mu_B H_i, T)$ approximated by $p(g\mu_B H_i, 0)$, as HK do. One will thus miss a term coming from the temperature dependence of $p(g\mu_B H_i, T)$ near $T_{\max}$, which may not be negligible. (b) Although we think the qualitative result (39) should hold, the way of calculating it by means of perturbation theory seems controversial. Between 0°K and $T_{\max}$, (35) involves values of $g\mu_B H_i$ such that $\alpha_i = g\mu_B H_i/k_B T > 4$. In this region, we noted in the previous sections that perturbation theory is almost meaningless. Therefore, we believe that to correctly describe the very-low-temperature behavior of $\rho_{Hi}(T)$ in (35) requires an exact theory of the scattering amplitudes. Then a comparison between the measured resistivity in the very dilute case in presence of an external field, and the measured resistivity in a more concentrated case with no external field, could give some information on the internal fields H_i.

Note added in proof. Since the present paper was submitted for publication, other experimental results on *Cu*Cr[25] and *Cu*Fe[26] alloys have become available. We should like to emphasize that rather than plotting the experimental $\Delta\rho/\rho_{H=0}$ against the experimental M^2 for small $g\mu_B H/kT$, as was proposed by Yosida[7] on the basis of a first Born calculation, which did not contain any of the Kondo logarithmic terms, the conclusions of this paper show that Eqs. (25) or (34) are more correct for alloys exhibiting the Kondo anomalies. In addition, the exact theory of More and Suhl[4] has now been published. It shows that perturbation theory is convergent not only for $T \gg T_{\text{Kondo}}$ and $g\mu_B H \ll kT$, but also for $g\mu_B H \gg kT_{\text{Kondo}}$, $T \ll T_{\text{Kondo}}$. This indicates that our high-field result, Eq. (27), should apply which implies that if M is saturated in temperature, $(\rho_H)_{\text{total}}$ should be practically temperature-independent.

ACKNOWLEDGMENTS

We would like to thank P. Monod for providing us with his data prior to publication and for many useful discussions. We are indebted to Professor H. Suhl and to R. M. More for many illuminating conversations. One of us (M.-T. B.-M.) is grateful to Professor W. Kohn for his hospitality during 1966–67.

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