# Magnetoresistance of Dilute Alloys

H. ROHRER
*IBM Zurich Research Laboratory, 8803 Rüschlikon, Switzerland*
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Measurements of the negative magnetoresistance in high fields of dilute Au–Mn and Au–Fe alloys reveal a qualitative difference between the anomalous scattering of the conduction electrons by manganese impurities and by iron impurities. The measurements were carried out on a series of alloys with impurity concentrations from 0.11 to 1.01 at.% Fe and 0.22 to 1.20 at.% Mn in pulsed magnetic fields up to 200 kOe and in the temperature range from 2.19 to 50°K. Whereas the effective exchange model of the *s-d* scattering is adequate in the case of Au–Mn, it fails to account for the anomalous features of the temperature and field dependence of the magnetoresistance in Au–Fe. Furthermore, the magnetoresistance of Au–Fe exhibits an unexpectedly strong concentration dependence which was not observed in Au–Mn and which could not be explained by any magnetic ordering of the Fe impurities. Moreover, the decrease of the zero-field resistivity due to short-range order has been calculated from the magnetoresistance and is found to be consistent with the location of the maximum of the zero-field resistivity.

## I. INTRODUCTION

EXTENSIVE theoretical and experimental studies on the problem of localized moments of paramagnetic impurities in dilute alloys have been concerned with the resistivity anomalies[1–3] and the response of the impurity magnetic moment to an external field.[1,4–6] The experimental results agree qualitatively with the predictions of the effective exchange model of the anomalous electron scattering. The application of the various theories based on the effective exchange model to the Kondo effect[7] of Au-, Ag-, Cu-, Mg-, and Zn-based alloys containing transition metal impurities yields $\eta|J|\ll 1$, where $\eta$ is the density of states per atom of the host metal and $J$ the effective exchange constant. Under this condition the Schrieffer-Wolff[8] transformation is valid and the application of the effective exchange model is consistent. However, the value of $\eta J$ varies only little[9] (less than a factor 2) for the different alloys in question—$\eta J\approx -0.06$ using Abrikosov's[10] approximation and about $-0.09$ using the theories of Hamann[2] and of Suhl and Wong[11]—so that $J$ is roughly proportional to $\eta^{-1}$. Such a result is not consistent with the form of $J$ given by Schrieffer and Wolff[8] and Kondo,[12] where $J$ is a sensitive function of the relative position of the Fermi energy of the host metal and the unperturbed $d$ levels of the impurity.

Results of the high-field magnetoresistance of dilute Au–Mn and Au–Fe are presented here, which show a qualitative difference in the anomalous scattering of the conduction electrons by Mn and Fe impurities. Whereas the magnetoresistance of Au–Mn is well described by the *s-d* exchange model and is consistent with the temperature dependence of the zero-field resistivity, this model is not adequate in the case of Au–Fe. This is unexpected since the Kondo effect of these two materials is very similar[13] and leads to the same results within the effective exchange model. In addition, the concentration dependence of the magnetoresistance is much more pronounced in Au–Fe than in Au–Mn, although the location of the resistivity maximum,[13] the excess specific heat,[14,15] and the anomalies of the low-temperature susceptibility[16] indicate impurity interactions of comparable strength in both alloys.

For the anomalous *s-d* scattering study, magnetoresistance measurements are useful in three respects. First, the magnetoresistance is characteristic of the bulk of the *s-d* resistivity and not only to some selected higher-order scattering processes as in the case of the temperature dependence of the zero-field resistivity. The saturation value of the magnetoresistance is closely related to the *s-d* resistivity $\rho_m$ and thus provides a direct and independent determination of the effective exchange constant $J$ which can then be compared to the results obtained from the Kondo effect. Second, the temperature and field dependence of the magnetoresistance $\Delta\rho_m/\rho$ is essentially determined by the response of the impurity moments to

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TABLE I. Properties of the Au–Mn and Au–Fe alloys.

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Au–Fe | | | | | | | | | |
| $c$ (at.% Fe) (chemical analysis) | 0.11 | 0.14 | $0.19_5$ | $0.24_2$ | 0.27 | 51 | 66 | 72 | 1.01 |
| $\rho$(4.2°K) ($\mu\Omega$ cm/at.%) | 7.46 | 7.30 | 7.34 | 7.72 | 7.66 | 7.80 | 7.78 | 7.45 | 7.38 |
| Au–Mn | | | | | | | | | |
| $c$ (at.% Mn) (nominal) | 0.15 | 0.22 | 0.33 | 0.40 | 0.57 | 0.83 | 1.20 | | |
| $\rho$(4.2°K) ($\mu\Omega$ cm/at.%) | 3.18 | 2.84 | 2.82 | 2.72 | 2.56 | 2.52 | 2.48 | | |

an external field. In particular $\Delta\rho_m/\rho$ is proportional to the magnetization squared if the moment is not field-dependent. Third, the magnetoresistance gives an experimental check of the theories proposed by Silverstein[17] and Harrison and Klein[18] to explain the appearance of a resistivity maximum on the basis of statistical internal fields.

Magnetoresistance measurements have been previously used to study the anomalous $s$-$d$ scattering. In the experimental studies of Gerritsen[19] and Los and Gerritsen,[20] the experimental data were not sufficiently complete to be able to draw definite conclusions. In other cases insufficient attention has been given to the interference of the potential and the spin scattering of the paramagnetic impurity[21] or to the violation of Mathiessen's rule in the presence of a magnetic field[22] (see Appendix). Therefore, the conclusions drawn from these experiments have to be accepted with reservation.

## II. EXPERIMENT

### A. Samples

The Au–Fe alloys have been prepared from 99.9999% pure Au and 99.9% pure Fe, starting with a master alloy of 1 at.% Fe and diluting it to the desired Fe concentration. After being kept for 30 min first above and then 20°C below the melting point, the alloy was quenched to room temperature by cold helium gas and then cold-worked. The concentrations were determined by chemical analysis, and agreed with the nominal concentrations within the experimental errors. Some properties of the alloys are given in Table I. The error in $\rho/c$ due to chemical analysis and geometry is estimated at 5%, so $\rho/c$ can be considered constant for the whole concentration range studied. The mean value of $\rho/c$ is 7.54 $\mu\Omega$ cm/(at.% Fe). This is slightly higher than in Gerritsen's work,[23] where for the same concentration range 7.24 $\mu\Omega$ cm/(at.% Fe) was found. This discrepancy may be due to the cold-working of our samples. The location and depth of the minimum and the maximum of the zero-field resistivity agree well with other experimental data.[13,23] The fact that $\rho/c$ is constant indicates that the iron is in solution.

The Au–Mn alloys were prepared by Méteau Précieux, Neuchâtel, Switzerland. Since Mn is easily soluble in Au, no further metallurgical analysis was carried out. The value of $\rho/c$ increases slightly with decreasing impurity concentration as also observed by MacDonald *et al.*[13] This is believed to be partly due to the presence of other impurities, and partly to the concentration dependence of the $s$-$d$ scattering.

### B. Apparatus

The magnetoresistance was measured in pulsed magnetic fields obtained by discharging a battery of capacitors through the field coil. The pulse duration was about 4 msec, so that skin effects were negligible. Longitudinal fields were used to keep the normal magnetoresistance low. The field inhomogeneity never exceeded 7%. In order to obtain accurate results over the whole field range, five different measurements were made for each temperature with peak field values of 20, 50, 100, 150, and 200 kOe. The different magnetic-field sweep rates (about a factor 10 in low fields) are useful because in this way the relaxation time effects can be shown to be unimportant. The resistivity was measured by the four-probe method. The temperature of the sample was varied by a heater wound around the sample which was then embedded in a Plexiglass tube. Liquid helium was allowed to flow between the field coil and the sample holder so that the heat was dissipated radially, and axial heat dissipation was small. The inhomogeneity of the sample temperature, indicated by the appearance of thermal voltages at the voltage leads (the sample copper-wire connections act as thermocouples) when heating the sample, amounted to about 10% or 7°K at 77°K. The temperatures were measured by Au+2.1 at.% Co thermocouples, which were calibrated by measuring the electrical resistivities first with the sample in liquid nitrogen and then again after heating it up from 4.2°K to a temperature of

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77°K as measured by the thermocouple placed in the middle of the sample. A comparison of these resistivities gave a systematic discrepancy of about 1.5°K. The measurement of the average temperature of the sample was therefore accurate to 2%. Below 4.2°K the temperatures were reached by pumping on the liquid helium.

### C. Measurements

Figures 1 and 2 show the negative magnetoresistance $(\Delta\rho_m/\rho_{tot})_{expt}$ of Au–Mn and Au–Fe, respectively, as a function of magnetic field and temperature for different impurity concentrations. In the high-concentration limit, $\Delta\rho_m$ is equal to the measured magnetoresistance $\Delta\rho$, whereas for low concentrations the normal, positive magnetoresistance is no longer negligible. $\Delta\rho_n$ was eliminated as follows. It is shown in the Appendix that $\Delta\rho_n$ and $\Delta\rho_m$ are, in general, not additive but that $\Delta\rho_m \sim (\Delta\rho - \Delta\rho_n)(1+0.8\Delta\rho_n/\rho)$. The correction factor $0.8\ \Delta\rho_n/\rho$ appears because in a magnetic field the relaxation times are different for spin-up and spin-down conduction electrons. This correction factor was neglected by Muto *et al.*,[22] who assumed that $\Delta\rho_m$ and $\Delta\rho_n$ are additive. However, for the very dilute alloys considered by these authors, the correction factor is essential. $\Delta\rho_n$ is obtained from the Kohler diagram[24] given by Lüthi[25] and Los and Gerritsen.[20] Kohler's rule[24] states that $\Delta\rho_n/\rho_0$ is a function of $H/\rho_0$ where $\rho_0$ is the zero-field resistivity. However, Kohler's rule in its usual form is only applicable if the relaxation time is field-independent, which is not the case for scattering by paramagnetic impurities. To account for the field dependence of the relaxation time, $\rho_0$ has to be replaced by $\rho_0+\Delta\rho_m$. The negative magnetoresistance $\Delta\rho_m$ may then be determined by an iteration procedure. As a first step $\rho_0$ is replaced by $\rho_0+\Delta\rho$ which, when put into a Kohler plot, yields a first approximation $\Delta\rho_n'$ for the normal magnetoresistance and $\Delta\rho_m'$ for the exchange part of the magnetoresistance. Next $\rho_0+\Delta\rho_m'$ is used for the Kohler plot and so on. This iteration converges rapidly and only for low concentrations and high temperatures was a second step necessary.

For Au+0.33 at.% Mn and Au+1.01 at.% Fe, we plotted the measured values of magnetoresistance in order to indicate the experimental accuracy.

## III. RESULTS AND DISCUSSION

### A. Negative Magnetoresistance

The magnetic-field-dependent part of the anomalous *s-d* scattering gives a negative contribution to the magnetoresistance. Whether the total magnetoresistance is negative or positive depends on the impurity concentration. The negative exchange contribution $\Delta\rho_m$ decreases linearly with decreasing impurity concentration, whereas $\Delta\rho_n$, the ordinary, positive part, increases with decreasing impurity concentration. Since it is the exchange scattering that is of interest here, $\Delta\rho_m$ should be the dominant part of the magnetoresistance. Unfortunately, this limits the impurity concentration of the alloys to values that are too high for impurity correlations to be completely excluded. In the Au–Mn system, the minimum useable concentration is of the order of 0.2 at.%, and for Au–Fe of the order of 0.1 at.%. In both cases, impurity correlation effects already show up in the Kondo effect. However, the concentration effects in the magnetoresistance can be eliminated if necessary.

Before writing down explicit expressions for the magnetoresistance based on recently derived theoretical expressions for the field-dependent relaxation times, the main features of the magnetoresistance will be described using a simple exchange scattering model, considering only the first Born approximation. Such a short discussion of the essential features of the negative magnetoresistance seems appropriate in view of the inadequate analysis of previous magnetoresistance work.[20–22] The scattering processes involving a spin flip of the conduction electron contribute $\frac{2}{3}$ to the zero-field resistivity and are frozen out in sufficiently high magnetic fields ($\mu_B H > 3kT$, where $\mu_B$ is the Bohr magneton and $H$ the applied field). The zero-field resistivity due to exchange scattering not involving a spin flip of the conduction electron is $\frac{1}{3}\rho_m = \frac{1}{3}AJ^2S(S+1)$, where $A$ is a constant of the host metal, and increases in an applied field to $AJ^2S^2$. The saturation value of the magnetoresistance due to these processes is therefore $\Delta\rho_{ms} = -\rho_m/(S+1)$. In addition to these two terms, there appears a third negative contribution to the magnetoresistance because of the interference of potential and exchange scattering not involving a conduction electron spin-flip. Although this term does not contribute to the zero-field resistivity (in this simple model) a substantial part of the negative magnetoresistance is generally due to this term. The size of the interference term depends on the ratio of the potential and the exchange scattering strengths, $|V/J|$. This term cannot only compensate the positive magnetoresistance due to the non-spin-flip exchange scattering processes, but for conduction electrons with spins antiparallel to the applied field, it also compensates the potential scattering. It is evident from the relaxation times given by van Peski-Tinbergen and Dekker[26] that in the high-field limit ($\mu_B H > 3kT$), this compensation is complete if $V^2 = J^2S^2$, thus leading to a saturation value of the magnetoresistance of $\Delta\rho_{ms}/\rho = -1$, where $\rho = \rho_m + \rho_{pot}$. The negative magnetoresistance is therefore in general a function of both $V$ and $J$, and to

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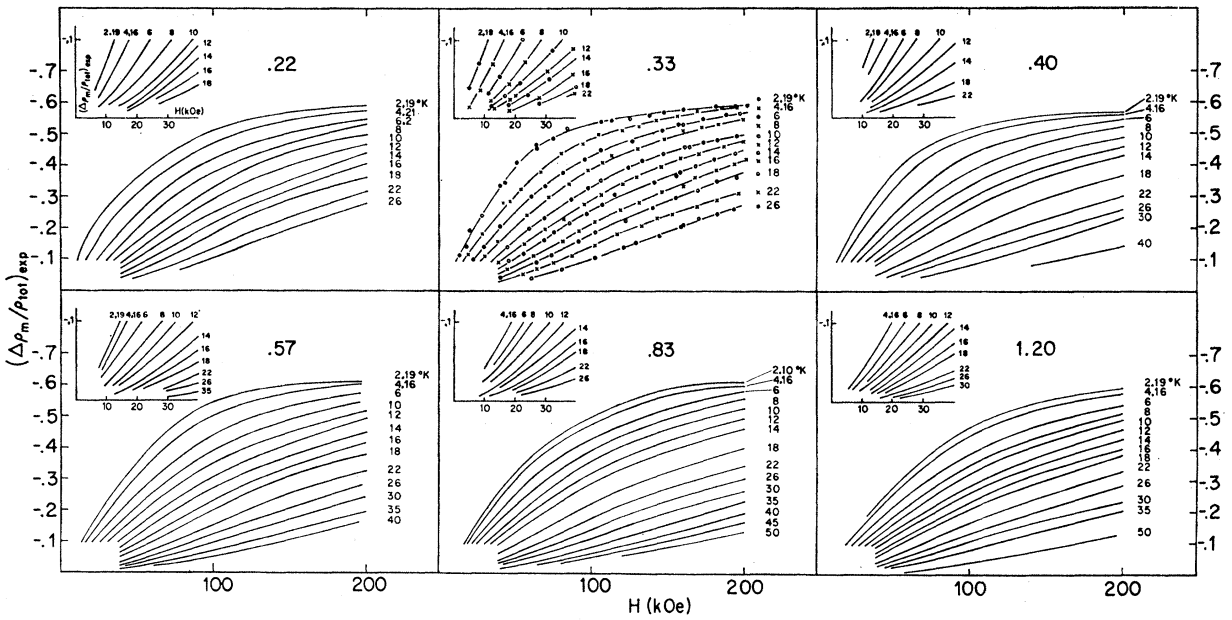


FIG. 1. Magnetoresistance $(\Delta\rho_m/\rho_{tot})_{expt}$ as a function of magnetic field and temperatures for Au–Mn. The large numbers on each plot indicate the manganese concentrations in at.%. The total measured zero-field resistance at temperature $T$ is $\rho_{tot}$.

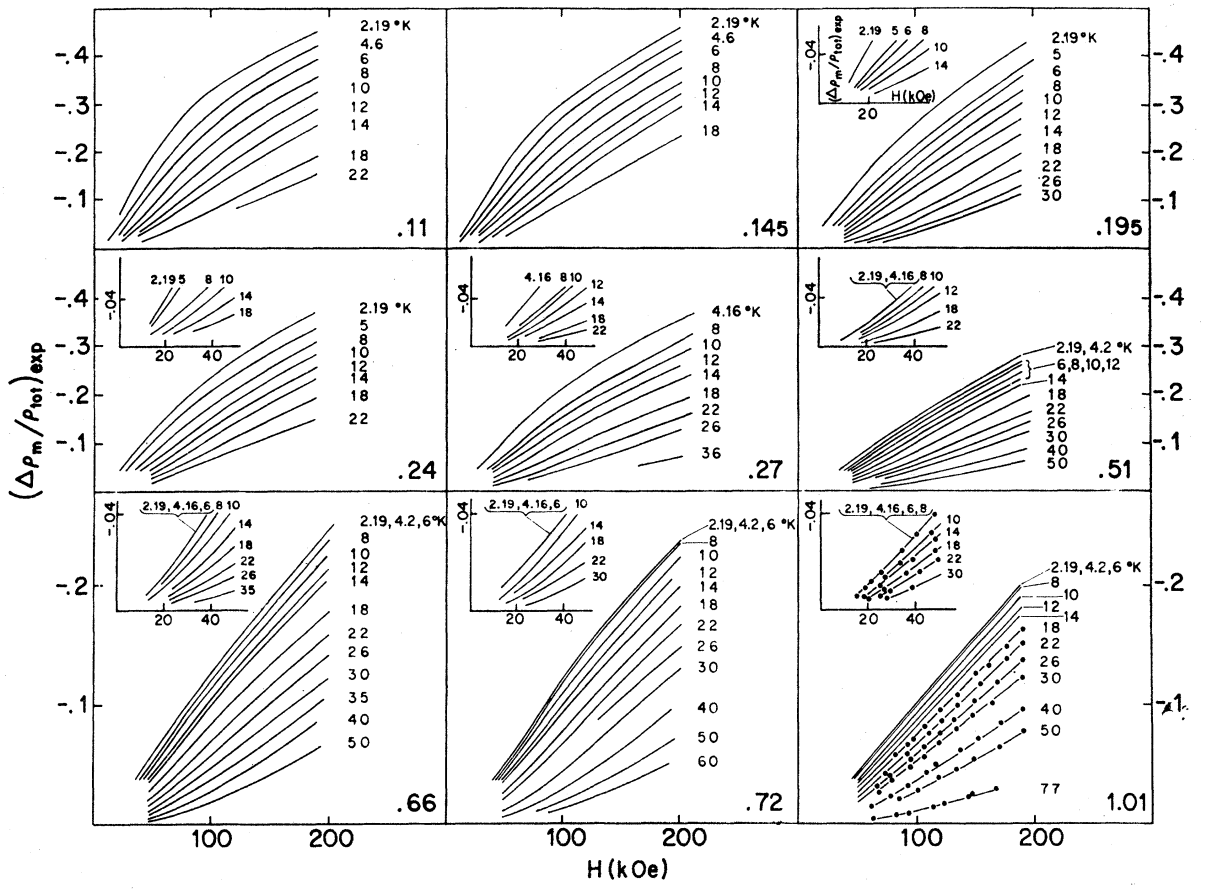


FIG. 2. Magnetoresistance $(\Delta\rho_m/\rho_{tot})_{expt}$ as a function of magnetic field and temperature for Au–Fe, The large numbers on each plot indicate the iron concentration in at.%.

determine $J$ from the magnetoresistance measurements it is necessary to know $V$. For the analysis of the magnetoresistance it is then useful to introduce a parameter $\beta$ for the ratio of the potential to exchange scattering, $\rho_{\text{pot}}/\rho_m$, and to consider only the ratio $\Delta\rho_m/\rho=(1+\beta)^{-1}(\Delta\rho_m/\rho_m)$. This has the advantage that the calculations are independent of the host-metal parameters contained in $A$ and that uncertainties in the sample dimensions and sample concentration are eliminated.

For a detailed calculation of the magnetoresistance we have used the relaxation times given by Abrikosov,[27] whose model of the $s$-$d$ exchange scattering should apply at temperatures well above the Kondo condensation temperature $T_c$, i.e., as long as the moment compensation is negligible. The Kondo effect of Au–Mn and Au–Fe indicates[9] that $T_c$ lies far below 1°K in both cases such that this condition is satisfied. Abrikosov's relaxation times can be written in the following form, when one assumes that the magnetization $M$ of the impurities is described by a Brillouin function $B_S(x)$:

$$\tau_{\pm}^{-1}\sim V^2\pm 2VJSB_S(x) + [J^2S(S+1)-J^2SB_S(x)\tanh(x/2S)] \times[1+\eta J\ln(E_F/Q)]^{-2}, \quad (1)$$

where $\tau_+$ and $\tau_-$ are the relaxation times for the conduction electrons with spins parallel and antiparallel to the applied field, respectively; $x=Sg\mu_BH/kT$, $g$ is the Landé splitting factor, and $Q=\mu_BH$ or $kT$, whichever is larger. Following van Peski-Tinbergen and Dekker,[26] the magnetoresistance can be written in terms of the conduction electron relaxation times

$$\Delta\rho_m/\rho=[\rho(H)-\rho(0)]/\rho(0)=2\tau_0/(\tau_++\tau_-)-1, \quad (2)$$

where $\tau_0$ is the zero-field relaxation time for the conduction electron of each spin direction. Substituting Eq. (1) into Eq. (2) gives

$$\Delta\rho_m/\rho=-(1+\beta)^{-1}\left\{1+\left(\frac{B_S(x)\tanh(x/2S)}{S+1}-1\right)\left(\frac{1+\eta J\ln(E_F/kT)}{1+\eta J\ln(E_F/Q)}\right)^2 + \frac{4\beta SB_S^2(x)}{\beta(S+1)[1+\eta J\ln(E_F/kT)]^{-2}+[S+1-B_S(x)\tanh(x/2S)][1+\eta J\ln(E_F/Q)]^{-2}}\right\}, \quad (3a)$$

where

$$\beta=\rho_{\text{pot}}/\rho_m=\frac{V^2}{J^2S(S+1)[1+\eta J\ln(E_F/kT)]^{-2}}$$

and

$$J=J(\beta)=\frac{(1+\beta)(T/\rho)(d\rho/dT)}{2\eta[1-0.5(1+\beta)(T/\rho)(d\rho/dT)\ln(E_F/kT)]}.$$

In deriving Eq. (3) it is assumed that no scattering centers other than the paramagnetic impurities are present. It should be mentioned that $\langle M^2\rangle$ of Eq. (13) in Abrikosov's paper has been correctly replaced by $\langle M^2\rangle-\langle M\rangle^2$.[28] The first two terms in Eq. (3) represent the magnetoresistance due to exchange scattering alone, the third is due to the interference of potential and exchange scattering. This third term is, in first Born approximation, proportional to the magnetization squared as previously shown by Yosida.[29] The expression for $\Delta\rho_m/\rho$ given by Eq. (3a) is proportional to $H^2$ in low fields ($\mu H\ll kT$) and is nearly field-independent in fields sufficient to align the impurity moments ($\mu_BH>3kT$). The weak field dependence in high fields is due to the higher-order spin-flip scattering processes involving no net spin flip of the impurity. These processes persist beyond fields sufficient to align the impurity moments and give rise to the terms depending logarithmically on $H$. Since these terms vary very slowly with $H$, $\Delta\rho_m/\rho$ can be considered saturated in fields for which $\mu_BH>3kT$. This quasisaturation value is denoted in the following by $\Delta\rho_{ms'}/\rho$.

To determine $\beta$ and thus $\rho_m$ and $J$ from the measured magnetoresistance, we consider merely the quasisaturation value $\Delta\rho_{ms}'/\rho$, such that $B_S(x)\approx 1$ and $\tanh(x/2S)\approx 1$, thus eliminating the spin, field, and temperature dependence of $\Delta\rho_m/\rho$ due to these functions. In the high-field limit, Eq. (3a) reduces to

$$\Delta\rho_{ms'}/\rho=-(1+\beta)^{-1}\left\{1-\frac{S}{(S+1)}\frac{[1+\eta J\ln(E_F/kT)]^2}{[1+\eta J\ln(E_F/\mu_BH)]^2} + \frac{4\beta S}{(S+1)[1+\eta J\ln(E_F/kT)]^{-2}+S[1+\eta J\ln(E_F/\mu_BH)]^{-2}}\right\}. \quad (3b)$$

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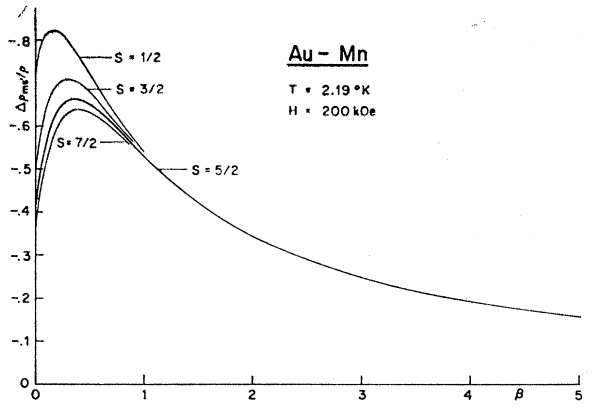


FIG. 3. Quasisaturation value $\Delta\rho_{ms'}/\rho$ of the magnetoresistance of Au–Mn as a function of $\beta$ for $H=200$ kOe, $T=2.19$°K, and $(T/\rho)(d\rho/dT)=-0.085$. For $\beta>1$, the curves for different spins converge to a single curve.

The temperature and field dependence of $\Delta\rho_{ms'}/\rho$ is negligible for $\mu_B H>3kT$. The spin dependence of $\Delta\rho_{ms'}/\rho$ in the case of Au–Mn alloys is shown in Fig. 3, where $\Delta\rho_{ms'}/\rho$ is plotted as a function of $\beta$ with $S$ as variable parameter. A maximum is shown by $\Delta\rho_{ms'}/\rho$ in the region where $V^2=J^2S^2$ (as for first-order processes alone). The maximum value is lower than in the case where only first-order scattering processes are considered when $(\Delta\rho_{ms}/\rho)_{\max}=-1$, because the scattering processes leading to the logarithmic terms in Eqs. (3a) and (3b) persist beyond experimentally accessible fields. It is interesting to note that for $\beta>1$, $\Delta\rho_{ms'}/\rho$ no longer depends on the value of $S$. This allows $\beta$ to be determined without knowing the actual value of $S$. This is quite useful since in many cases $S$ is not very well known. Once $\beta$ has then been determined from graphs like those shown in Fig. 3, $\Delta\rho_m/\rho$ can be calculated as a function of $H$ and $T$.

This way of determining $\beta$ involves some ambiguity as soon as $|\Delta\rho_{ms'}/\rho|_{\text{expt}}>|\Delta\rho_{ms'}/\rho|_{\beta=0}$. In this case, there exist two values of $\beta$ for each value of $\Delta\rho_{ms'}/\rho$ and some additional information is needed to decide which of the two values, $\beta_1$ or $\beta_2$ ($\beta_1<\beta_2$), is the appropriate one. In many cases (like Au–Mn, Au–Fe, Cu–Mn, and others) the lower value $\beta_1$ can be excluded, because it would lead to values of $\rho_{\text{pot}}$ that are much smaller than those due to nonmagnetic impurities with corresponding ionicity and ionic radii.

### B. Analysis of the Experimental Results

We now analyze the experimental results in terms of the equations derived in the previous section. A direct comparison of the measured magnetoresistance with the expressions given by Eqs. (3a) and (3b) is not feasible, because the contribution due to scattering centers other than the paramagnetic impurities and impurity correlation have not been taken into account in the expressions given in the previous section.

It is shown in the Appendix that the effect of the nonmagnetic scattering centers is to multiply $(\Delta\rho_m/\rho_{\text{tot}})_{\text{expt}}$ by a correction factor $(1+\Delta)$, where $\Delta$ is given by

$$\Delta=1.8\tau_0/\tau_a.$$

In this expression, $\tau_0$ is the relaxation time of the conduction electrons due to scattering by the paramagnetic impurities in zero field, and $\tau_a$ is that due to the other scattering centers. The phonon contribution to $\tau_a$ is obtained from the resistivity of pure gold, the contribution due to other impurities and crystal defects is not well known. However, this latter contribution was estimated from the residual resistivity of a pure Au sample prepared in the same way as the alloys.

The impurity correlation effects are not to be treated theoretically. In the case of Au–Mn, they are not important, and as will be shown below, in the case of Au–Fe, they will be taken into account empirically.

#### *1. Gold-Manganese Alloys*

The results for the gold-manganese alloys are shown in Fig. 1. The magnetoresistance of the samples containing 0.22, 0.33, and 0.40 at.% Mn does not exhibit any noticeable concentration effect down to 2.19°K, which is the lowest temperature considered. Only those samples for which the impurity correlations are not important will be considered in the discussion below. The quasisaturation of the magnetoresistance, $\Delta\rho_{ms'}/\rho$, yields for $S=\frac{5}{2}$, $\beta_2(2.19°\text{K})=0.64$, $\beta_2(4.16°\text{K})=0.69$, $\beta_2(6°\text{K})=0.74$, and $\beta_2(8°\text{K})=0.76$. If the value of $\beta_2$ for the three higher temperatures is normalized to 2.19°K, 0.63, 0.65, and 0.64, are found, respectively, giving an average value of 0.64. The corresponding values of $\beta_1$ can be ruled out because they imply a value for $\rho_{\text{pot}}$ that is too small. Using the value

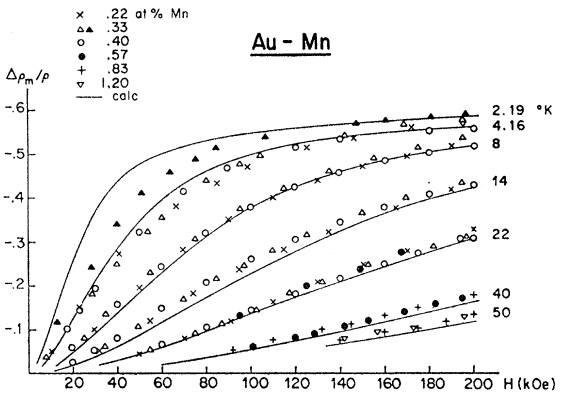


FIG. 4. Comparison of the experimentally determined magnetoresistance $(\Delta\rho_m/\rho_{\text{tot}})_{\text{expt}}$ of Au–Mn with the magnetoresistance calculated by Eq. (3a).

obtained for $\beta_1$ gives a value for the potential scattering for $Mn^{++}$ impurities in Au $\rho_{pot}(Mn^{++}) = 0.45\ \mu\Omega$ cm/at.%, which is slightly less than the potential scattering of $Cu^{+}$ in Au, $\rho_{pot}(Cu^{+}) = 0.46\ \mu\Omega$ cm. However, one expects the potential scattering due to $Mn^{++}$ impurities to be considerably larger than that due to $Cu^{+}$.

For the value of $\beta_2$ deduced above, one obtains for the exchange resistivity $\rho_m = 1.7\ \mu\Omega$ cm/at.%, for the effective exchange constant $J = -0.18$ eV and for the Kondo condensation temperature $T_c \approx 10^{-2}$ °K. Applying Abrikosov's approximation[10] to the temperature dependence of the zero-field resistivity[13] one finds

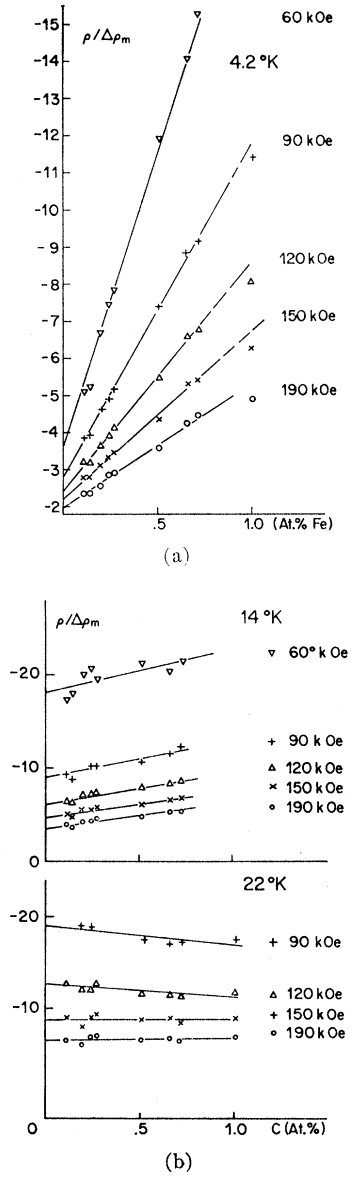


FIG. 5. Concentration dependence of the magnetoresistance of Au–Fe for the temperatures 4.16, 14, and 22°K.

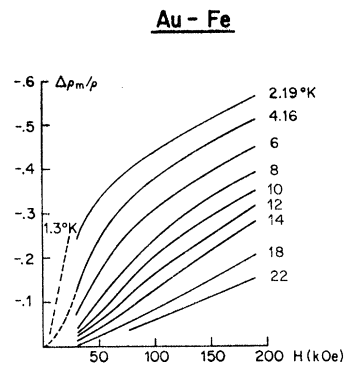


FIG. 6. Magnetoresistance of Au–Fe after elimination of concentration effects. The dotted lines have been determined from low-field magnetoresistance measurements of Gerritsen (see Ref. 23).

$\rho_{mA} = 1.1\ \mu\Omega$ cm/at.%, $J_A = -0.16$ eV, and $T_{cA} \sim 5 \times 10^{-3}$ °K,[9] where the index A refers to Abrikosov's approximation. As far as the effective exchange constant and the Kondo condensation temperature is concerned, the agreement is quite good. The difference between $\rho_m$ and $\rho_{mA}$ is, on the other hand, larger than can be explained by any uncertainties of the host-metal parameters. Better agreement is obtained by using Hamann's expression for the zero-field resistivity. The temperature dependence of the zero-field resistivity yields then $\rho_{mH} = 1.6\ \mu\Omega$ cm/at.%, which is very close to the value obtained from the magnetoresistance. Although the *s-d* resistivity $\rho_m = 1.7\ \mu\Omega$ cm/at.% has been derived from the magnetoresistance using Abrikosov's model, it is not inconsistent to compare $\rho_m$ with $\rho_{mH}$. Abrikosov's expression for the zero-field resistivity $\rho_{mA}$ can be considered as an expansion of $\rho_{mH}$ to first order in

$$\gamma/x^2 = \pi^2 S(S+1)/\ln(T/T_c)^2.$$

Neglecting the higher-order terms in $\gamma/x^2$ is expected to have only a slight influence on $\Delta\rho_m/\rho$ as long as $\rho_m$ is the dominant part of $\rho$, although $\rho_{mH}$ and $\rho_{mA}$ may differ considerably even above $T_c$.

With $\beta_2 = 0.64$, the field and temperature dependence of the magnetoresistance was computed using Eq. (3a). A comparison with the experimental results is shown in Fig. 4. At low temperatures, only the three purest samples were used, since for higher impurity concentrations, the influence of impurity correlation is already noticeable although it is small. At the highest temperatures considered, the normal, positive magnetoresistance and the correction due to the phonon resistance are comparable with the measured magnetoresistance in the case of the low-concentration samples. A reliable determination of $\Delta\rho_m$ is then no longer possible. Therefore only the results for the impure samples are shown which, at these temperatures, no longer exhibit any concentration effects. The agreement between theory and experi-

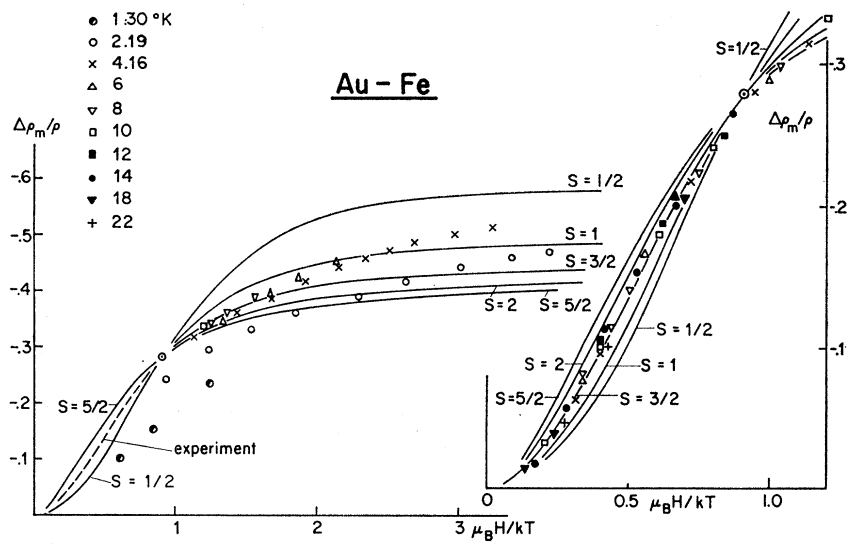


FIG. 7. Comparison of $(\Delta\rho_m/\rho)_{id}$ with Eq. (3a). The calculations have been fitted to the experiment at 190 kOe and 14°K, which corresponds to $\mu_B H/kT=0.91$, denoted on the graphs by ⊙. The detailed behavior of the experimental and theoretical magnetoresistance for $\mu_B H/kT<1.2$ is shown in the insertion.

ment is quite satisfactory. Some deviation from the theory is found for intermediate values of the fields at 4.16°K and more pronounced at 2.17°K. The reason for this discrepancy is not known.

### 2. Gold-Iron

In the case of Au–Fe alloys, an analysis of the experimental results is not so straightforward as for Au–Mn. When Figs. 1 and 2 are compared, two striking differences between the magnetoresistance of Au–Fe and Au–Mn are noticed.

(a) In high magnetic fields and at low temperatures, $\Delta\rho_m/\rho$ for Au–Fe does not saturate. In the case of the purer samples, there is a knee in the $\Delta\rho_m/\rho$ versus $H$ curve but $\Delta\rho_m/\rho$ keeps increasing for fields up to 200 kOe.

(b) $\Delta\rho_m/\rho$ exhibits a pronounced concentration effect both in the magnitude and in its magnetic field and temperature dependence.

This strong concentration dependence makes a comparison between the experimental results and the theory more difficult. The expression for $\Delta\rho_m/\rho$ given by Eq. (3a) does not contain any concentration dependence, while the concentration dependence introduced by the correction factor $(1+\Delta)$ (see the previous subsection) is an order of magnitude smaller than those observed in AuFe and also of opposite sign.

However, phenomenologically it has been found that the concentration dependence of the magnetoresistance for the entire concentration, temperature, and magnetic field range used in the present experiment can be described by

$$(\Delta\rho_m/\rho_{tot})_{expt}=(B+cD)^{-1}, \qquad (4)$$

where $B$ and $D$ are functions of the magnetic field and temperature. This concentration dependence of $\Delta\rho_m/\rho$ is shown in Fig. 5 for $T=4.16$, 14 and 22°K. The concentration-independent part of $\Delta\rho_m/\rho$, given by $1/B$, is obtained by extrapolation $(B+Dc)$ to zero concentration as shown in Fig. 5. This value of $1/B$ should not be considered as the magnetoresistance of an alloy with very low impurity concentration but should be considered rather as the magnetoresistance of an idealized material in which the impurities are not correlated, but with an impurity concentration high enough to keep $\Delta\rho_n$ and the corrections involving $\rho_0/\rho_a$ negligible.

Figures similar to Fig. 5 were drawn for each temperature, and $B$ was obtained by a least-mean-square fit, assuming the concentrations to be correct and weighing the results of the different samples equally. The results are shown in Fig. 6; $1/B$, the magnetoresistance of an idealized alloy, being denoted by $(\Delta\rho_m/\rho)_{id}$. Also plotted in Fig. 6 are the results obtained from the static low-field measurements of Gerritsen.[23]

The temperature and field dependence of $(\Delta\rho_m/\rho)_{id}$ deviates in two respects from the predictions of Eq. (3a). First, according to Eq. (3a), the magnetoresistance should be a function of $H/T$ only, except for the slowly varying logarithmic terms containing $H$ and $T$ separately. In a plot of $(\Delta\rho_m/\rho)_{id}$ versus $H/T$, the experimental points should therefore lie on a single curve. As shown in Fig. 7 this is only the case at 4.16°K and higher temperatures. Second, the expected saturation of $\Delta\rho_m/\rho$ in high fields $(\mu_B H/kT>2)$ is not found.

Outside the region of these two anomalies, i.e., for $T\geq 4.16$°K and $\mu_B H/kT<1.8$, the experimental data are best fitted by Eq. (3a) if $S=\frac{3}{2}$ and $\beta(4.16°K)=1.8$ (see Fig. 7). Such a value of $S$ is consistent with a magnetic moment $\mu_{eff}=3.6\mu_B$ found by Lutes and Schmit[16] from magnetization measurements. However,

the value of $\beta$ derived this way is not consistent with the temperature dependence of the zero-field resistivity. The value of $\beta$ yields $\rho_m = 2.8\ \mu\Omega$ cm/(at.% Fe), whereas the temperature dependence of the zero-field resistivity yields $\rho_{mA} = 1.25\ \mu\Omega$ cm/(at.% Fe) at 4.16°K.[9] Therefore, the model of Abrikosov of the anomalous *s-d* scattering is not adequate to describe the magnetoresistance of Au–Fe.

As mentioned above, there exists a temperature $T_0$, below which $(\Delta\rho_m/\rho)_{id}$ is no longer a function of $H/T$ only. This temperature $T_0$ has also been determined for the actual Au–Fe samples and is found to increase rapidly with the Fe concentration.

In Fig. 8, $T_0$ is plotted versus the sample concentration. Empirically, $T_0$ can be described by

$$T_0 = \alpha + \gamma c$$

in the concentration range 0.1 to 1 at.% Fe, where $\alpha = 3.7$°K and $\gamma = 52$°K/at.% Fe. It is interesting to note that $\alpha$ coincides with $T_0$ obtained from $(\Delta\rho_m/\rho)_{id}$, thus supporting the extrapolation procedure used to obtain $(\Delta\rho_m/\rho)_{id}$.

### 3. *Low-Temperature Resistivity Maximum and Internal Fields*

With the magnetoresistance measured up to very high fields we are in a position to check experimentally the theories of Silverstein[17] and Harrison and Klein[18] concerning the local maximum of the zero-field resistivities at low temperatures. According to their theories, the decrease of the resistivity, $\Delta\rho_T$, due to impurity correlation is given by

$$\Delta\rho_T = \int_{-\infty}^{+\infty} p(H)\,\Delta\rho_m(H)\,dH, \qquad (5)$$

where $p(H)$ is the probability that an impurity experiences a field $H$ due to other impurities. The interaction of the impurity is assumed to be of the Marshall-Klein-Brout (MKB)[14,30] type. With a $p(H)$ derived from specific-heat anomalies and $\Delta\rho_m(H)$ calculated from the experimentally determined temperature dependence of the zero-field resistivity, Harrison and Klein obtained the location of the resistivity maximum and the subsequent linear decrease of $\rho(T)$ of Au–Fe in very good agreement with experiment. If, however, the experimentally determined $\Delta\rho_m(H)$ is inserted in Eq. (5), the reduction of the resistivity, $|\Delta\rho_T|$, is about nine times larger. If the influence of the MKB fields on the magnetoresistance itself is also considered, $|\Delta\rho_T|$ is increased even further. Therefore Eq. (5) does not give results consistent with the experiments, contrary to the conclusion reached by Harrison and Klein. This discrepancy is due to the fact that the effect of short-range internal fields is qualitatively different from that of a uniform external field. It is evident from the form of the field-dependent relaxation times [Eq. (1)] that the interference term, which is the dominant term in the magnetoresistance of Au–Fe and Au–Mn, disappears for statistical short-range fields of the Marshall-Klein-Brout type. The efficiency of the spin-flop term to freeze out the *s-d* scattering depends on the amount the bands of spin-up and spin-down conduction electrons is split locally by the internal fields. Therefore the influence of statistical internal fields on the resistivity is substantially smaller than that of a uniform external field, and is similar to the case of long-range antiferromagnetism treated by Yosida.[29] Therefore it is incorrect to use the uniform field value $\Delta\rho_m(H)$ in Eq. (5).

For a rough estimate of the influence of MKB fields on the zero-field resistivity we make the following assumptions: (1) $\langle M_S\rangle = 0$ and $\langle M_S^2\rangle = \frac{1}{3}S(S+1)$, where $M_S$ is the magnetic spin quantum number of the impurity. (2) The conduction electron bands are fully split by the internal fields. (3) Collective excitations of the impurity clusters can be neglected. Then the relaxation times for spin-up and spin-down conduction electrons are in the first Born approximation given by

$$1/\tau_\pm \sim V^2 + J^2S(S+1) \pm \tfrac{2}{3}J^2S(S+1)\tanh(x/2S). \qquad (6)$$

Setting $V^2/J^2S(S+1) = \beta$, the effective magnetoresistance—the magnetoresistance to be inserted in Eq.

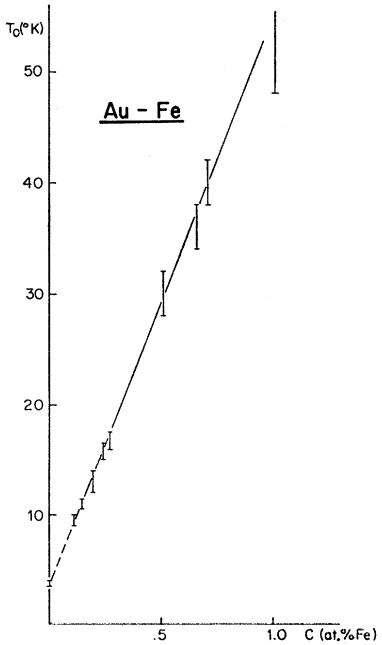


FIG. 8. The characteristic temperature $T_0$ as a function of the Fe concentration. The point at $c = 0$ represents $T_0$ for $(\Delta\rho_m/\rho)_{id}$.

[30] W. Marshall, Phys. Rev. **118**, 1520 (1960).

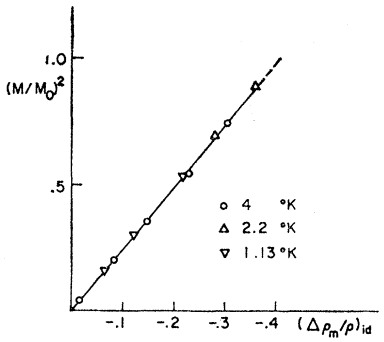


FIG. 9. $(\Delta\rho_m/\rho)_{id}$ versus $(M/M_0)^2$, the relative magnetization squared, for Au–Fe in the temperature range 1.13 to 4°K and in magnetic fields up to 65 kOe.

(5)—can then be written

$$(\Delta\rho_m)_{eff}/\rho = -\frac{(4/9)\tanh^2(x/2S)}{(1+\beta)^2}. \quad (7)$$

Using the value of $\beta$ derived earlier, in the case of Au–Fe, $(\Delta\rho_m)_{eff}/\rho \approx -0.06$ is found for the high-field limit of the effective magnetoresistance. This is about a factor 9 smaller than the measured high-field magnetoresistance. Therefore if $\Delta\rho_m(H)$ in Eq. (5) is replaced by $(\Delta\rho_m)_{eff}$, good agreement is again obtained with the experimentally determined temperature dependence of the zero-field resistivity.

### 4. Discussion

The magnetoresistance of Au–Mn is in good agreement with the prediction of the *s-d* exchange model. The magnetoresistance $(\Delta\rho_m/\rho)_{id}$ of Au-Fe, on the other hand, deviates in two ways from the expected behavior. First, below 4.16°K, $\Delta\rho_m/\rho$ is not a function of $H/T$ alone. Second, the expected saturation in high fields is not found. The field and temperature dependence of the magnetoresistance is closely connected to the magnetization $M$, and the anomalies of $\Delta\rho_m/\rho$ may be related to anomalies of the magnetization. The dominant part of the magnetoresistance of Au–Fe (and also of Au–Mn) is due to the interference term, which amounts to about 75%. In this case the temperature and field dependence of the magnetoresistance given by Eq. (3a) reduces to the simple expression given by Yosida[29]:

$$\Delta\rho_m/\rho = P(M/M_0)^2, \quad (8)$$

where $P$ is a constant containing the ratio of potential to spin scattering amplitude, and $M_0$ is the saturation magnetization. Equation (8) is expected to be valid only for field-independent impurity moments.

In Fig. 9, the magnetoresistance $(\Delta\rho_m/\rho)_{id}$ is compared with the magnetization found by Kitchens *et al.*[4] The magnetoresistance for 4 and 1.13°K has been obtained from the magnetoresistance at 4.16 and 1.30°K assuming an $H/T$ dependence. Although $(\Delta\rho_m/\rho)_{id}$ does not depend only on $H/T$ in this temperature region, the deviations are small for the temperature differences in question. Below 65 kOe (the field up to which the hyperfine field of the Fe impurities has been measured), the magnetoresistance is proportional to the square of the magnetization, showing that the low-temperature anomaly of the magnetoresistance is caused by the anomalous magnetization. In high fields, however, $(\Delta\rho_m/\rho)_{id}$ increases beyond the saturation value of $-0.4$ indicated by Fig. 9.

A possible explanation for the increase of the magnetoresistance beyond the expected saturation is the breaking up of a Nagaoka bound state,[31] which should further reduce that part of the *s-d* scattering that has not yet been frozen out by aligning the impurity moments. However, the Mössbauer experiment by Kitchens *et al.*[4] shows that above 30 kOe, the saturation hyperfine field is practically independent of the applied field. Therefore a possible bound state would already be completely broken up by a field of 30 kOe. This is consistent with the Kondo condensation temperature $T_c$ of about 0.1°K derived above. The field necessary to break up a bound state completely is about $5\,kT_c$,[32] which corresponds to a magnetic field of about 10 kOe. It is then evident that the high-field anomaly of the magnetoresistance—the deviation from the expected saturation behavior—is not related to the Nagaoka condensation. The low-temperature anomaly of the magnetoresistance—the deviation from the $H/T$ dependence—is, for low fields, compatible with the breaking up of a bound state associated with a condensation temperature of about 0.1°K. However, since it persists up to very high fields, this anomaly is probably not related to the Nagaoka condensation, either. The anomalies of the magnetoresistance of Au–Fe must therefore be related to some property of the Fe moment other than the compensation of the moment by the conduction electrons.

Another unexpected feature of the magnetoresistance of Au-Fe is the strong concentration dependence. The location of the maximum of the zero-field resistivity,[13] the excess low-temperature specific heat,[14,15] and the susceptibility anomalies[16] indicate impurity interactions to be of similar strength in both Au–Mn and Au–Fe. Yet the concentration dependence of the magnetoresistance is very pronounced in Au–Fe but is hardly noticeable in Au–Mn. This suggests that, for the magnetoresistance, the impurity correlations are of different origin than those responsible for the concentration dependence of zero-field resistivity. The

[31] Y. Nagaoka, Phys. Rev. **138**, A1112 (1965); Progr. Theoret. Phys. (Kyoto) **37**, 13 (1967).
[32] Sang Boo Nam and James Wing Fai Woo, Phys. Rev. Letters **19**, 649 (1967).

strong decrease of the low-field magnetoresistance with increasing Fe concentration indicates that the impurity interactions are predominantly antiferromagnetic. Ferromagnetic interactions would make it easier to align the impurity moments by the applied field and thus increase the low-field magnetoresistance. In order to account for a characteristic temperature $T_0=50°K$ and a reduction of the high-field magnetoresistance by a factor 2.5 in Au+1% Fe, the average impurity interactions have to be of the order of 50°K. These interactions are an order of magnitude stronger than expected from the low-temperature specific heat[14] and susceptibility anomalies.[16] Since it is especially the anomalies of the magnetoresistance which exhibit a strong concentration dependence, the apparently strong interaction of the Fe impurities may be related to the problems of the magnetoresistance anomaly.

## IV. CONCLUSIONS

1. The negative magnetoresistance of Au–Mn is well described by Abrikosov's model of the exchange resistivity, and is consistent with the observed Kondo effect. The Mn moment is well localized and not compensated by the conduction electrons; its response follows the Brillouin function for $S=\frac{5}{2}$ up to 200 kOe.

2. The magnetoresistance of Au–Fe exhibits some anomalous features: (a) Below a certain temperature $T_0$, the magnetoresistance is not a function of $H/T$ only; (b) no saturation is found in fields for which $\mu_B H \gg kT$. These anomalies are not compatible with the prediction of Abrikosov's *s-d* exchange model and cannot be accounted for by a possible Nagaoka-type compensation of the impurity moment. It appears therefore that the effective exchange model is not appropriate for Fe impurities in Au.

3. The very strong concentration dependence of the magnetoresistance of Au-Fe indicates impurity interactions which are an order of magnitude larger than expected from specific heat or susceptibility anomalies. It is suggested that these apparently strong impurity interactions are related to the magnetoresistance anomalies.

4. The model of Silverstein and of Harrison and Klein for the resistivity maximum and the subsequent resistivity decrease is in good agreement with experiment, if their magnetoresistance is replaced by an effective magnetoresistance which is essentially that derived by Yosida for long-range antiferromagnetism.

## ACKNOWLEDGMENTS

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## APPENDIX

Here we show that Mathiessen's rule is generally not obeyed in the presence of a magnetic field. That is, $\Delta\rho_m(H)$ is changed by the additional field-independent scattering centers such as phonons and nonmagnetic impurities. We assume that the effect of the additional scattering centers is to add $1/\tau_a$ to the inverse of the relaxation times of the conduction electrons for each spin direction. With the same method[26] as that used to derive Eq. (2) one obtains for the magnetoresistance

$$\Delta\rho_m(H, 1/\tau_a)=\frac{(2\tau_0-\tau_+-\tau_-)\tau_a+\tau_0(\tau_++\tau_-)-2\tau_+\tau_-}{2\tau_0\tau_a(\tau_++\tau_-)+4\tau_0\tau_+\tau_-}. \tag{A1}$$

If no additional scattering is present, then $1/\tau_a=0$ and $\Delta\rho_m(H, 1/\tau_a)$ reduces to $\Delta\rho_m$ of Eq. (2).

According to Eq. (1), the field-dependent relaxation times $\tau_+$ and $\tau_-$ are of the form

$$1/\tau_\pm=1/\tau_0-f\pm g,$$

where $g$ stands for the second and $f$ for the fourth term on the right-hand side of Eq. (1). Inserting these relaxation times into Eq. (A1), one obtains the following expression for the relative deviation from Mathiessen's rule:

$$\delta=\frac{\Delta\rho_m(H, 1/\tau_a)-\Delta\rho_m(H, 0)}{\Delta\rho_m(H, 0)}=-\frac{g^2/f}{[g^2/f+1/\tau_0-f][\tau_a(1/\tau_0-f)+1]}. \tag{A2}$$

We note that $\delta\leq 0$, since $f, g>0$ and $1/\tau_0\geq f+g$.

The presence of field-independent scattering generally decreases the negative magnetoresistance $\Delta\rho_m$. Mathiessen's rule is satisfied only in the absence of interference terms, and the deviation is most pronounced when the interference terms are dominant.

If the case of Au–Mn, one finds in the high-field limit ($\mu_B H>3kT$) at 2.19°K, $g_{sat}=4f_{sat}$ and $1/\tau_0=6f_{sat}$, where $g_{sat}$ and $f_{sat}$ denote the quasisaturation values of $g$ and $f$, respectively. Inserting these values into Eq. (A2) one obtains for the high-field limit of the correction $\delta$

$$\delta=-0.83\tau_0/\tau_a \quad \text{(high fields)} \tag{A3a}$$

provided $\tau_0/\tau_a\ll 1$. For small fields ($\mu_B H\ll kT$), $a\ll 1/\tau_0$ and

$$g^2/f\approx(g_{sat}^2/f_{sat})\frac{B_S(x)}{2\tanh(x/2S)}=\tfrac{1}{3}(S+1)(g_{sat}^2/f_{sat}).$$

In this equation $(1+\eta J\ln E_F/Q)^{-2}$ has been approxi-

mated by 2. One obtains then for $\delta$

$$\delta=-0.76\tau_0/\tau_a \quad \text{(low fields)}. \tag{A3b}$$

This shows that the correction $\delta$ depends only weakly on field and temperature. We have therefore used

$$\delta=-0.8\tau_0/\tau_a \tag{A3c}$$

for all values of field and temperature.

The magnetoresistance calculated by Eq. (3a), $(\Delta\rho_m/\rho)$ Eq. (3a), and the experimentally determined magnetoresistance $(\Delta\rho_m/\rho_{\text{tot}})_{\text{expt}}$, of Figs. 1 and 2 are then related by

$$(\Delta\rho_m/\rho)_{\text{Eq. (3a)}}=(\Delta\rho_m/\rho_{\text{tot}})_{\text{expt}}(1+1.8\tau_0/\tau_a) \tag{A4}$$

as long as $\tau_0/\tau_a\ll 1$.

A second, similar correction factor is due to $\Delta\rho_n$ and $\Delta\rho_m$ not being additive. That Mathiessen's rule does not hold in this case either can immediately be seen if $\Delta\rho_n$ is described by an additional scattering rate $1/\tau_n$, which is field-dependent but equal for the conduction electrons of either spin direction. The deviation $\delta_n$ from Mathiessen's rule is obtained by replacing $\tau_a$ by $\tau_n$. With the same values for $f$, $g$, and $\tau_0$ one then obtains

$$\delta_n\approx-0.7\Delta\rho_n/\rho \tag{A5}$$

provided $\Delta\rho_n/\rho\ll 1$. $\Delta\rho_m$ and $\Delta\rho$ are then related by

$$\Delta\rho_m\approx(\Delta\rho-\Delta\rho_n)(1+0.7\Delta\rho_n/\rho). \tag{A4'}$$

It is interesting to consider the case $1/\tau_a\gg 1/\tau_0$, which might occur in ternary alloys or in the case of very large positive magnetoresistance. One finds from Eq. (A1) that the interference terms no longer contribute to the negative magnetoresistance and one has $\Delta\rho_m=-f/2$.

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# Electronic Spin Polarization around a Magnetic Impurity Using Perturbation Theory*

H. U. Everts† and B. N. Ganguly‡
*Department of Physics, University of Toronto, Toronto, Canada*

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The electronic spin polarization $p(r)$ arising from the $s$-$d$ exchange interaction is computed using iterated solutions of Nagaoka's truncated equations and direct perturbation theory. Most of our detailed calculations are only to order $J^2$ (where $J$ is the exchange constant). However, arguments are given to suggest that certain qualitative features [such as the oscillatory behavior of $p(r)$] will remain even if we work to all orders in $J$. We critically discuss the work of Falk and Fullenbaum as well as Suhl, whose results agree with ours apart from the important difference that our Ruderman-Kittel-Kasuya-Yosida term is proportional to the average of an effective spin rather than the bare impurity spin. This effective spin also enters the static susceptibility $\chi$. In one of the Appendices, we briefly consider the effect of potential scattering on the spin polarization.

## I. INTRODUCTION

It seems natural to expect that one of the most important manifestations of the Kondo effect[1] will be in the conduction-electron spin polarization $p(r)$ around a magnetic impurity. Historically, the first such study was made by Nagaoka[2] using a self-consistent solution of the decoupled equations of motion for retarded double-time Green's functions. He found that in contrast to the Born approximation for the polarization (associated with the names Ruderman-Kittel-Kasuya-Yosida[3] and thus referred to as RKKY) his result had a much longer range and moreover the electron spins were favored to align antiparallel to the impurity atom spin. This result led Nagaoka to the conclusion that below a critical temperature $T_K$ there exists some sort of quasibound state between the electron spins and the impurity spin. The coherence length of this quasibound state was estimated to be $10^{-4}$ cm. It has since been realized that Nagaoka's original solution of his equations was incorrect and that his equations do not give rise to any bound state. As a consequence the result for the electron spin polarization given in Ref. 2 should be disregarded. In contrast, Suhl[4,5] made use of his alterna-

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† One of the authors (H.U.E.) gratefully acknowledges a travel grant from the Deutsche Forschungsgemeinschaft.
‡ Present address: Department of Physics and Astronomy, Louisiana State University, Baton Rouge, La.
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