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Negative Transverse Magnetoresistance in Dilute Polycrystalline Alloys of *Au*Fe

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The field dependence of the negative transverse magnetoresistance has been evaluated from the total effect measured in polycrystalline wires of the *Au*Fe system at 2 and 4.2 °K. The solute concentration ranged from 10–400 atomic ppm, and the applied field strengths ranged up to 26 kOe. In making this evaluation, consideration was given to the high-field–low-field transition, the anomalous size effects, the field dependence of the electron's relaxation time, and possible changes in the anisotropy of the solute scattering—considerations which will be relevant to similar measurements carried out on dilute alloys of analogous Kondo systems. For sufficiently concentrated samples, the negative component is shown to vary linearly with the square of the effective magnetization obtained from other work.

I. INTRODUCTION

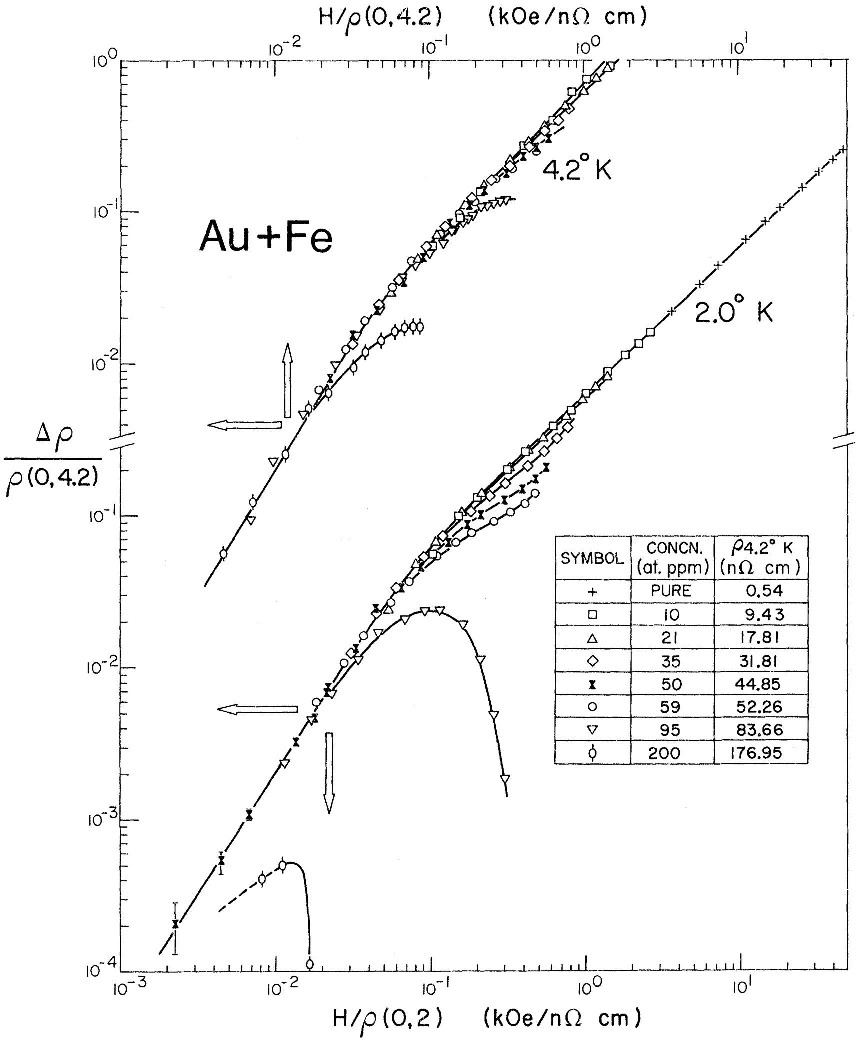
The field dependence of the negative component of the transverse magnetoresistance observed in polycrystalline wires of the *Au*Fe system has been evaluated at 4.2 and 2 °K. The experiments involved the determination of the total magnetoresistance at these temperatures, in fields ranging to 26 kOe, and for alloys having concentrations in the range 10–400 atomic ppm. The negative component was extracted from these data taking into consideration possible complications by high-field–low-field effects, anomalous size effects, changes in the electron's scattering anisotropy, and the field dependence of the electron's relaxation time. Although this paper is restricted to a particular alloy system, we believe that many of the remarks relating to these complications will be relevant to similar experiments on dilute polycrystalline alloys containing a magnetic solute (Kondo alloys).

The experiments follow the increasing tendency to study very dilute alloys in order to minimize solute-solute interaction effects. *Au*Fe was chosen since there now exist several studies of the magnetic[1] and galvanomagnetic[2–6] effects in its dilute alloys which provide useful data upon which to draw when discussing the negative component. To the best of our knowledge there are, however, no other published measurements of the magnetoresistance in very dilute alloys of this system.[7] This paper can therefore fill this gap, while also hopefully clarifying some of the empirical considerations required when evaluating the negative magnetoresistance in very dilute Kondo alloys.

II. EXPERIMENTAL DETAILS

Polycrystalline ingots of 0.375 in. diam were prepared in a high-vacuum induction furnace as described previously.[2,8] The starting materials were Au of 99.9999 at.% purity[9] [$\rho_{4.2\,^\circ\mathrm{K}}$ obtained for an annealed wire of 0.040 in. diam is given in Fig. 1; it corresponds to a residual resistance ratio (RRR) ($=\rho_{273\,^\circ\mathrm{K}}/\rho_{4.2\,^\circ\mathrm{K}}$) of about 3700] and Fe of approximately[10] 99.99 at.% purity. After casting, the alloy ingots were rotary swaged at room temperature to a final diameter of 0.080 in. Before measurement, the samples were degreased, washed, and chemically etched to remove about 0.001 in. from the diameter, then annealed at 550 °C for 24 h in an evacuated Pyrex container. This was air cooled to ambient temperature before opening. Sections from each sample were analyzed by the Analysis Section of the National Research Council of Canada (using either a spectrographic or the atomic-absorption method) with the results shown in Fig. 2. Also shown there is $\rho_{4.2\,^\circ\mathrm{K}}$ versus Fe concentration for our samples together with corresponding data either obtained in this laboratory or taken from the literature. (The gradient of the solid line corresponds to 7.98 $\mu\Omega$ cm/at.%.) Values of the concentration determined from $\rho_{4.2\,^\circ\mathrm{K}}$ as in Fig. 2 are probably more appropriate than those obtained by the above chemical methods since they directly reflect the amount of solute in solution and the amount "seen" by the conduction electrons. They also have the advantage of the supporting evidence from other sources shown in Fig. 2. Consequently, where there was significant difference between the results from the physical and chemical methods, we accepted the former. Values of $\rho_{4.2\,^\circ\mathrm{K}}$ and the corresponding concentrations are tabulated in Fig. 1.

The resistivity of specimens of length 2.25 in. was determined by the usual four-probe technique using[11] a primary current of approximately 5.8 A. The separation of the potential probes was determined directly from the knife-edge indentations, while the form factor was obtained from the sample's weight and known density (taken to be that of pure Au). A standard cryostat-electromagnet combination was used to give fixed temperatures of 2.0 or 4.2 °K with a field ($\vec{H}$) ranging up to 26



SYMBOL	CONCN. (at. ppm)	ρ4.2° K (nΩ cm)
+	PURE	0.54
□	10	9.43
△	21	17.81
◇	35	31.81
⌶	50	44.85
○	59	52.26
▽	95	83.66
ϕ	200	176.95

FIG. 1. Kohler plots showing the field dependence of the total transverse magnetoresistance $\Delta\rho = \rho(H, T) - \rho(0, T)$ observed at $T = 2.0$ and 4.2 °K.

kOe. The dc potentiometric arrangement and the measurement techniques were identical to those described previously.[11] Estimates of the experimental accuracy are given in Sec. III.

III. TOTAL MAGNETORESISTANCE

Figure 1 shows the field dependence observed for the total transverse magnetoresistance $\Delta\rho = \rho(H, T) - \rho(0, T)$ at $T = 2.0$ and 4.2 °K (ρ is the resistivity). The results are shown in Kohler plots having $\Delta\rho/\rho(0, 4.2)$ as the ordinate in each case and with $H/\rho(0, T)$ as the abscissa. The ordinate can be regarded as the change in resistance per unit concentration c, where the relationship between c and $\rho(0, 4.2)$ is given in Fig. 2. We estimate the precision of any value of $\Delta\rho/\rho(0,4.2)$ [i.e., neglecting the error in the form factor] to be better than ± 0.001; such error bars are indicated in Fig. 1 for the case of Au + 50 ppm Fe at 2.0 °K. The corresponding accuracy of any $\Delta\rho/\rho(0, 4.2)$ is estimated to be ± 0.002, and the resulting uncertainty in the evaluated negative component (± 0.004) is shown in typical cases in Figs. 5–7.

Throughout this paper, we shall concentrate upon the results at 2.0 °K, since the negative component is greatest there and possible interference from phonon effects will be least. (Previous work[3] suggests that, in fact, phonon effects can safely be neglected below 4 °K.) Note that the behavior of the total magnetoresistance in the high- and low-field regions, where $\omega\tau \gg 1$ and $\omega\tau \ll 1$, respectively (ω is here the cyclotron frequency corresponding to the applied field $\vec{H}$ and τ is an average electronic relaxation time for an orbit), can be fitted quite well by an H^n dependence, where $n = 0.95 \pm 0.01$ and 1.5 ± 0.04, respectively. Concerning the low-field dependence, the discrepancy between observation and the expected quadratic behavior (see Sec. IV) probably indicates only that the true low-field condition has not been reached in the experiment. [The lowest value of $H/\rho(0, 2)$

in Fig. 1 corresponds to $H=100$ Oe. Any reduction of H below this is self-defeating since the imprecision of the final datum becomes overwhelmingly large.] Turning to the high-field condition, since the arguments leading to the H^1 dependence are at best qualitative[12-14] the slight discrepancy between the expected exponent of unity (see Sec. IV) and that observed is perhaps not of great significance. What we would like to draw attention to, however, is a feature which is not obvious on the scale of Fig. 1: Close scrutiny of the high-field background on an expanded scale suggests that n is field dependent. There is evidence just within the experimental precision of a tendency for $\Delta\rho$ to saturate as H increases. [A similar more pronounced effect has also been seen[15] in pure polycrystalline Cu at comparable values of $H/\rho(0)$.] Clearly, there is a need for further investigation of this effect.

IV. EVALUATION OF NEGATIVE COMPONENT

The negative component is observed as a deviation from Kohler's rule. This rule, and its requirements for validity, have been discussed on numerous occasions[12,16-18] so that it is unnecessary to go into detail here. Briefly, it states that for a given temperature the fractional change in resistivity, $\Delta\rho/\rho(0)$, where $\rho(0)$ is here the resistivity in zero applied field, can be written

$$\Delta\rho/\rho(0)=f(H/\rho(0)) \quad . \qquad (1)$$

f is a function which in general depends on the orientation in the lattice of $\vec{H}$ and the primary current, as well as upon the type of metal,[12,19] but in a polycrystalline alloy this orientation dependence averages to zero.

Since the extraction of the negative component concerns deviations from Kohler's rule, the first step in the analysis is therefore to establish what form a behavior, which follows Kohler's rule, would take in equivalent circumstances. The second step is then to attribute the observed deviations from this behavior to the negative component, but taking account of other possible sources of deviation such as were referred to in Sec. I.

Figure 3 is intended to clarify this by showing schematically a comparison of Kohler plots of the isothermal field dependence of the total magnetoresistance observed in very dilute polycrystalline alloys typical of those under consideration. (The temperature is assumed low enough that the solute scattering is the dominant scattering mechanism throughout.) With a nonmagnetic solute, Kohler's rule is obeyed and the Kohler plot is a unique function as indicated in Fig. 3(a): It has distinct sections of gradients of approximately 1 and 2 which correspond, respectively, to the field dependence observed in the high- and low-field conditions. (The quadratic low-field dependence is expected from theory,[12,19] but the linear high-field dependence is an unexplained empirical fact[2,12-14] ob-

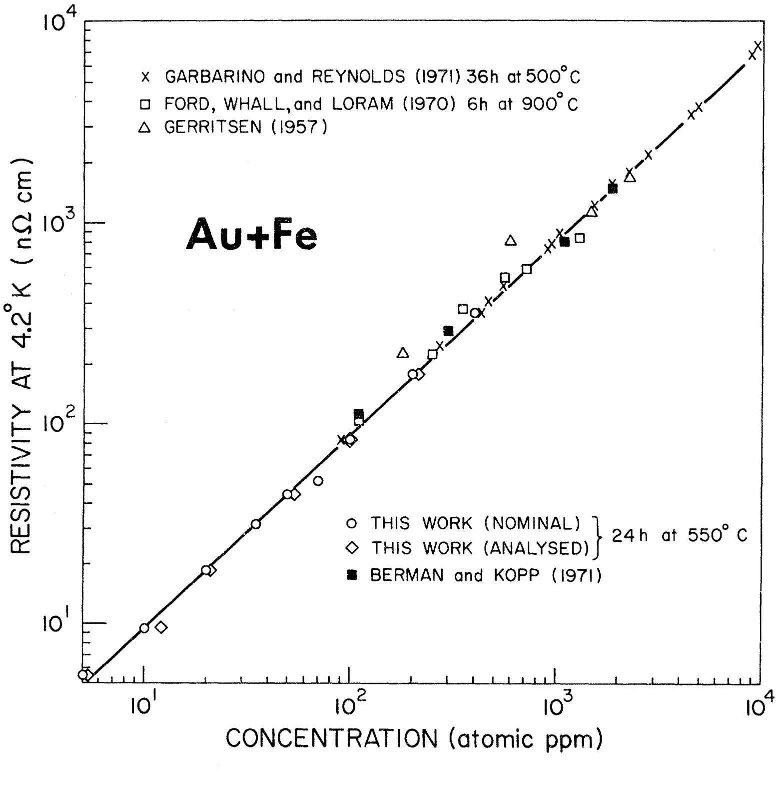


FIG. 2. Resistivity at 4.2 °K vs Fe concentration taken from the sources indicated. Solid line has a gradient of 7.98 $\mu\Omega$ cm/at.%. Reference citations are: Berman and Kopp (Ref. 6), Garbarino and Reynolds (Ref. 5), Ford *et al.* (Ref. 4), and Gerritsen (cited in Ref. 7).

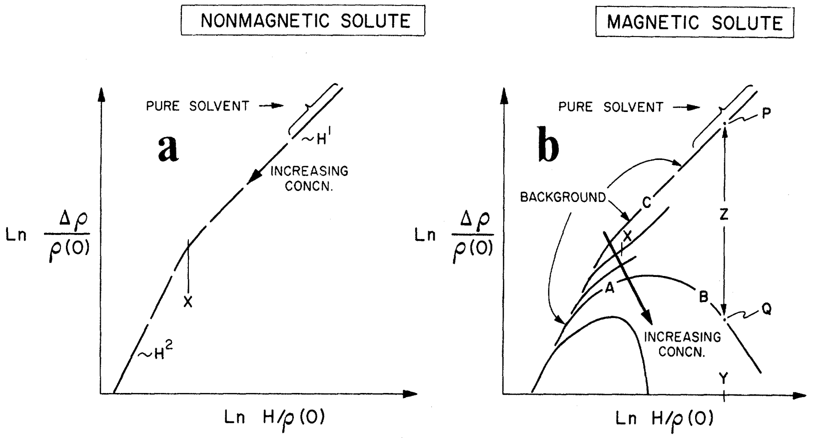


FIG. 3. Schematic view of the isothermal field dependence of the transverse magnetoresistance of very dilute polycrystalline alloys where the solvent metal has a multiply connected Fermi surface. The temperature is assumed low enough that the solute is always the dominant scattering mechanism. The solute may be either nonmagnetic (a) or magnetic (b).

served for a polycrystalline uncompensated metal having a multiply connected Fermi surface.) In measurements on very dilute alloys at low temperatures the experiment inevitably encompasses the transition X between the high- and low-field conditions, and it can be precipitated by varying either the amount of electron scattering or the applied field strength. Examples of Fig. 3(a) can be found in the literature for the case of a $1B$ metal where the dominant scattering source is phonons,[20] lattice defects,[20] or a neighboring polyvalent solute.[21,22]

Turning to the case with a magnetic solute [Fig. 3(b)], the negative component of the total magnetoresistance leads to deviations from Kohler's rule as shown. For sufficiently dilute alloys the total effect can remain positive in the range of fields available, and even undergo a transition to the high-field condition as at X. As the solute concentration is reduced the negative component diminishes continuously to the point where it is no longer detectable within the precision of the experiment. Curve C represents such a case. These data form part of what we shall call[2] the "background" behavior of $\Delta\rho/\rho(0)$ observed when the negative component is vanishingly small. This background is the normal positive magnetoresistance which in the low-field region is characteristic of the Coulombic scattering of the solute, while in the high-field region it is determined by the topology of the solvent-metal's Fermi surface.[2] It includes the effect of the high-field–low-field transition, and its establishment is the first step referred to above. An example of Fig. 3(b) for dilute *Ag*Mn appears in Alderson and Hurd,[2] while Fig. 1 shows the corresponding case for *Au*Fe alloys. Referring to the data at 2.0 °K in Fig. 1, we find that an alloy containing 10 ppm Fe shows no detectable negative magnetoresistance within our precision; these data join up smoothly with those for pure Au to give the background behavior used in the subsequent analysis. Furthermore, the more dilute samples (10–50 atomic ppm, for example) show evidence of the transition to the high-field condition [as at X in Fig. 3(b)] and this feature reappears in the data shown in Fig. 5.

Returning to Fig. 3(b), suppose we wish to proceed to the second step referred to above and attribute the deviation from Kohler's rule at some chosen abscissa (say Y) to the negative component (ρ_m). We have stated elsewhere[2,11] that ρ_m is given directly as the product $Z\rho(0)$. (Z, as shown, is the vertical separation between the background and the measured value of the total magnetoresistance.) The idea behind this approach was to account for the effect of the low-field–high-field transition which, if ignored in very dilute alloys, can lead to an erroneous field dependence of the negative component.[2] Although to this extent the idea was an improvement upon earlier methods, it is still incorrect in principle[23] and needs to be modified.

The error arises from our implicit assumption that τ is field independent. The above procedure assumes that in plots such as Fig. 1 or Fig. 3(b) a given value of $H/\rho(0)$ corresponds to a certain value of $\omega\tau$ which is fixed for alloys of different concentrations. This would be correct if τ were field independent, as in the case of a nonmagnetic solute, but (as Rohrer has already pointed out[24]) this is not so with a magnetic solute. In that case the application of H inevitably lengthens τ through the freezing out of the spin-flip scattering.[25,26]

Specifically in Fig. 3(b), suppose we obtain a certain value of $\omega\tau$ from the experimental value of $H/\rho(0)$ at Y. The τ so obtained corresponds to the appropriate $\rho(0)$ and is correct for the point P (since the background behavior involves no negative component). But this same value of $\omega\tau$ is not valid for the point Q, for in this case as the field was applied τ increased from the value given by $\rho(0)$. In fact, at Q some value $\omega\tau'$ applies, where $\tau' > \tau$ is always true. As Rohrer has indicated,[24] to get the form of Fig. 3(b) corrected for the field dependence of τ we should have as the abscissa

$\ln H/[\rho(0) \pm \rho_m]$. (Comment on the choice of signs is given below.) Since making the correct plot therefore requires prior knowledge of the quantity we are seeking, we are obviously forced to a less direct approach. This can be made through an iterative procedure as suggested by Rohrer.[24] We shall differentiate between two cases depending on whether the field dependence of the alloy' s magnetoresistance has a positive or negative gradient [portions A and B of Fig. 3(b), respectively]. Whether a positive or negative gradient is observed depends on the experimental conditions and on the solute's concentration (in Rohrer's case[24] the conditions were selected so that only a negative gradient was observed).

Figure 4(a) shows in detail how the iterative procedure is carried out in the second case above; i.e., when the field dependence of the magnetoresistance has a negative gradient. A is the background curve representing alloys so dilute that no negative component is measurable, while B is that for an alloy having an appreciable negative component. Taking a given abscissa, say X_B, the negative component read directly from the graph ($\rho_m' = \rho_m + \epsilon_3 + \epsilon_2 + \epsilon_1$) is always greater in magnitude than the true value (ρ_m). [This follows since $\bar{\tau}$ and the divergence of A and B in Fig. 4(a) both increase with H.] To correct for the discrepancy between ρ_m' and ρ_m, the first iteration (curve 1) is obtained by shifting each abscissa to a new value $H/[\rho(0) - \rho_m']$. The second iteration (curve 2) is obtained from curve 1 by shifting each abscissa to $H/[\rho(0) - \rho_m'']$, where $\rho_m'' = \rho_m + \epsilon_3 + \epsilon_2$. This procedure is continued until the change produced in the negative component (ϵ_n) is less than the precision of the data in curve B. In Fig. 4(a) this has been reached with ϵ_4, and hence curve 3 represents the required plot of $\Delta\rho/\rho(0)$ versus $H/[\rho(0) - \rho_m]$. The negative component at any abscissa can therefore be read directly off the corrected curve as $\rho_m = Z\rho(0)$.

Note that if the gradient of curve B in Fig. 4(a) had been positive [as, for example, in portion A of Fig. 3(b)], the same procedure would be followed except that the new values of the abscissa after each iteration are $H/[\rho(0) + \rho_m']$, $H/[\rho(0) + \rho_m'']$, etc. In other words, the choice of sign in the denominator must be made to give a *reduction* in the magnitude of ρ_m' at each step which, as we saw above, is required on physical grounds. In any case, this choice is also forced by the algebra since, as is easily shown, the iteration is only convergent if the magnitude of ρ_m' is reduced at each step.

The field dependence of the negative component from Fig. 1 is shown in Fig. 5.[27] The open and closed symbols represent, respectively, the corresponding values obtained before and after the above iterative correction. The significance of this correction obviously depends on the relative magnitudes of $\rho(0)$ and ρ_m, and upon the precision of the measured $\Delta\rho/\rho(0)$; Fig. 5 shows that in this particular case it is only significant for alloys containing less than about 60 atomic ppm—and then only at the higher field strengths.

In evaluating the negative component we have so far referred to the allowances made for the high-field–low-field transition and for the field dependence of the electron's relaxation time. The anomalous size effects and the possibility of changes in the anisotropy of the electron's scattering remain to be considered from the list of Sec. I. Turning first to the size effects (which were probably first observed by Berlincourt[28]), these occur[11,29] in "thick" polycrystalline wires (i.e., where the calculated electron mean free path is less than the wire's diameter) and are greater than expected from simple theory. A systematic study of size effects in alloys is in progress, but already we have referred to their manifestation in the transverse magnetoresistance.[11] To the best of our present knowledge, size effects always produce deviations from Kohler's rule in the direction shown in Fig. 4(b). Clearly, the effects become less important as either the solute's concentration or the wire's

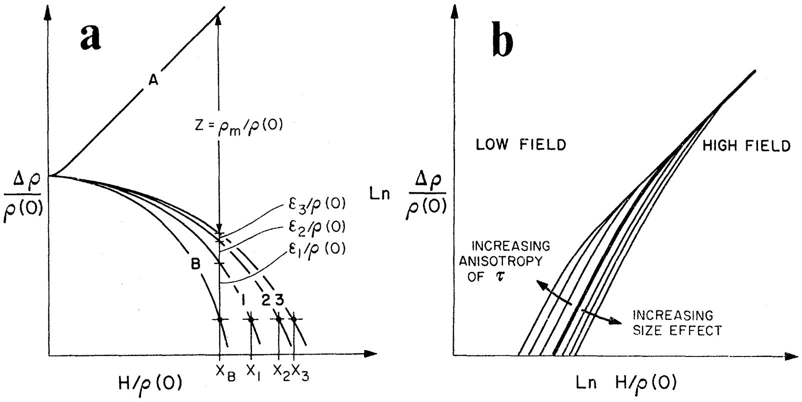


FIG. 4. Schematic view of the low-temperature isothermal field dependence of the transverse magnetoresistance of very dilute polycrystalline alloys. (a) Indicating the iterative procedure described in the text for extracting the negative component of the magnetoresistance. (b) Kohler plot showing the deviations from Kohler's rule observed for increasing size effects and for increasing anisotropy of either $\tau(\vec{k})$ or $\epsilon(\vec{k})$.

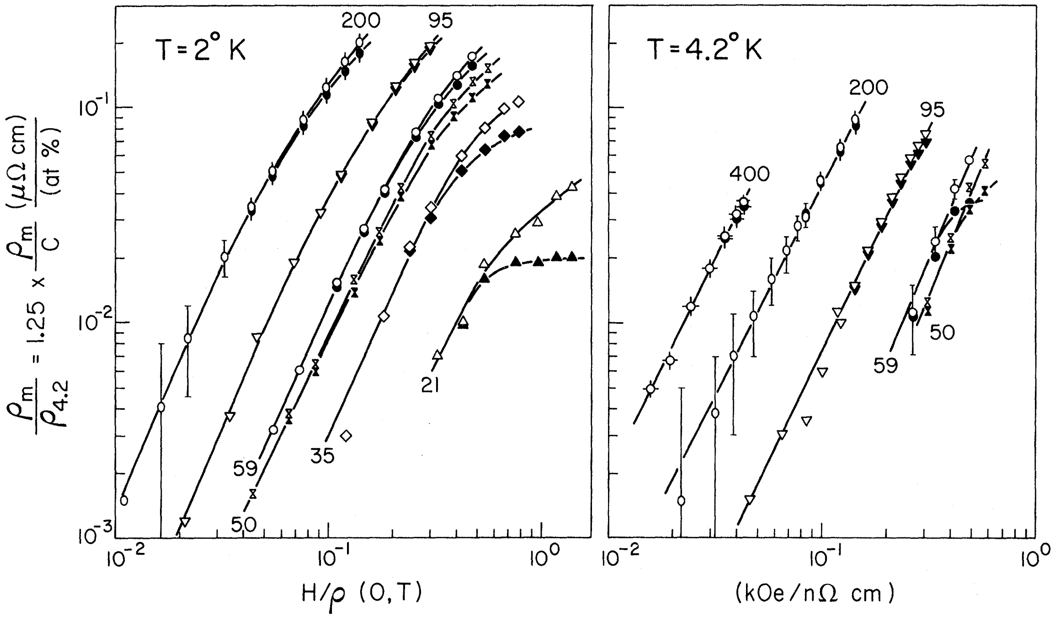


FIG. 5. Kohler plots showing the field dependence of the negative component ρ_m obtained from the data in Fig. 1. The abscissa is $H/\rho(0,T)$, where T is either 2 or 4.2 °K, while the ordinate is effectively the negative component per unit solute concentration c. (The latter is related to $\rho_{4.2\,°\mathrm{K}}$ as shown in Fig. 2.) At 4.2 °K the negative component in samples having less than 50 ppm was not detectable. The iterative procedure described in Sec. IV has been carried out to account for the field dependence of $\tau(\vec{k})$. The open and closed symbols represent, respectively, the behavior before and after correction. The numbers refer to the solute concentration in atomic ppm.

diameter is increased, and our experience suggests that if the wire samples are at least 0.080 in. in diameter size effects are unlikely to be significant at 4.2 °K for concentrations above a few ppm. It was determined empirically that the data of Fig. 1 represent samples free from size effects.

Turning to the remaining possible complication cited in Sec. I, it is a requirement for the validity of Kohler's rule[12,16-18] that during the experiment there shall be no variation in the anisotropy of either the electron's relaxation time $\tau(\vec{k})$ or of the solvent-metal's Fermi surface $\epsilon(\vec{k})$. In principle, both of these quantities could vary as the solute concentration is changed; the former through solute-solute interaction and the latter through changes in band structure upon alloying. Furthermore, a possible variation of $\tau(\vec{k})$ is not restricted to alloys: It could occur in a pure metal as the amount of the dominant scattering process is changed in some way. There are in the literature several examples of this effect in a Kohler plot of the form of Fig. 1 or 3(b): In Cu, for example, can be seen the changing anisotropy of phonon scattering with temperature,[20] of dislocation scattering with plastic deformation,[30] of defects with neutron irradiation[18] (in longitudinal magnetoresistance), and of solute scattering with the nature of the solute[11] (Zn compared with Mn, Fe, or Co). From all of this it appears that an increase in the anisotropy of $\tau(\vec{k})$ produces, for a polycrystalline sample at least, an increasing intercept on the Kohler plot as indicated in Fig. 4(b). The data of Fig. 1 showed no evidence of this, and so support the assumption that the effects of either solute-solute interaction or band-structure changes are not manifested as apparent changes in the anisotropy of $\tau(\vec{k})$ at such low solute concentrations.

V. DISCUSSION OF NEGATIVE COMPONENT

One result[26] of the freezing out of spin-flip scattering (Sec. IV) responsible for the negative magnetoresistance ρ_m is that below saturation, i.e., for $g\mu_B H/kT <\sim 2$ (in the usual terminology), ρ_m is essentially proportional to M^2, where M is the magnetization. Explicitly, the relationship is

$$\frac{\rho_m}{c} = \frac{3\pi}{2\epsilon_f}\,\frac{m}{e^2\hbar}\,\Omega J^2 M^2\left[1+\left(\frac{\mu_B}{\mu_{\mathrm{eff}}}\right)^2\right] . \qquad (2)$$

Here the common symbols have their usual meanings, while c is the atomic concentration of magnetic solute (i.e., c is atomic ppm/10^6), Ω is the atomic volume of the solvent, $\mu_{\mathrm{eff}} = g\mu_B[S(S+1)]^{1/2}$ is the effective moment per solute atom, and J is the s-d exchange constant. It is not difficult to make a qualitative test of Eq. (2) since $M \propto H$, providing the conditions are such that the Brillouin function is approximately linear. All that is required is to demonstrate that $\rho_m \sim H^2$. (Such is shown in Fig. 5—to which we return shortly.) But to apply Eq. (2) qualitatively requires a knowledge of M and μ_{eff}. For this we shall use the results of Loram *et al.*[1]

First, note that 2.0 °K, with a maximum field of 26 kOe, $g\mu_B H/kT$ in our experiment ranges up to 1.7. The conditions are, therefore, generally within the subsaturation requirement. Turning now to the qualitative features of Fig. 5, note that for the two most dilute samples there is a pronounced tendency toward saturation of the negative component at the highest field strengths. This is most noticeable at 2.0 °K and it correlates with the onset of the high-field condition [as at X in Fig. 3(b)] and was referred to in Sec. IV. [We are referring throughout to the closed symbols in Fig. 5 which are the data corrected for the field dependence of $\tau(\vec{k})$.] This saturation illustrates what might be expected on physical grounds: Since in the high-field condition the galvanomagnetic effects are not influenced by the electron scattering processes,[2] ρ_m becomes field independent there.

Apart from this high-field behavior, ρ_m determined at 4.2 °K follows an H^2 dependence to within experimental precision. This qualitatively confirms Eq. (2) and, of course, has been observed in other systems.[26,31] At 2.0 °K, however, there is a noticeable deviation from this quadratic behavior for all concentrations. This is partly due to the effects of magnetic saturation (i.e., to the increasing curvature of the Brillouin function as $\mu_B H/kT$ increases) and partly to the magnetization of the polarized cloud of spin-compensating electrons about the solute atom (the Nagaoka correlation[32]). When both are allowed for (as in Fig. 6 discussed below) a more linear variation of ρ_m with M^2 is observed.

Turning to the quantitative application of Eq. (2), Loram *et al.*[1] have measured the magnetic susceptibility of very dilute (29–100 ppm) *Au*Fe alloys in the range 1.5–4.2 °K. Their data indicate a μ_{eff}/μ_B of about 3.41 corresponding to $S=1.28$ when $g=2$. The field dependence of the magnetization of one of these alloys was determined at 2.02 and 4.2 °K. It was found that this could not be fitted by a simple Brillouin function with $S=1.28$ and $g=2$ (which gave values up to 12% too great), but that account had to be taken of the magnetization of the above polarized conduction-electron cloud. This can be expressed[1] in an effective g factor (g_{eff}) such that the magnetization per Fe atom due to the solute (expressed in Bohr magnetons) is

$$M = S g_{\text{eff}} B_S (g_{\text{eff}} S \mu_B H/kT) . \tag{3}$$

B_S is the Brillouin function for spin S, and g_{eff} is given by

$$g_{\text{eff}} = g\left(1 - \frac{0.5}{\log_{10}[T_{\text{eff}}/T_K]}\right) . \tag{4}$$

T_K is the Kondo temperature, and T_{eff} is approximated by

$$kT_{\text{eff}} = [(kT)^2 + (g\mu_B H)^2]^{1/2} . \tag{5}$$

Using $S=1.28$ and $T_K=0.1$ °K (the empirical values obtained from the susceptibility), Loram *et al.* find that the observed isothermal field dependence of M (obtained at 2.0 or 4.2 °K and in fields up to 59 kG) can satisfactorily be fitted by Eqs. (3)–(5). Accordingly, we can use this approach to calculate M for substitution in Eq. (2).

Figures 6 and 7 show the negative component per unit solute concentration (exactly as the ordinate of Fig. 5) obtained at 2.0 and 4.2 °K, respectively, plotted against $M^2[1+(\mu_B/\mu_{\text{eff}})^2]$. μ_{eff} is taken[1] as $3.41\mu_B$ and M is calculated from Eqs. (3)–(5) using $S=1.28$ and $T_K=0.1$ °K. According to Eq. (2), such a plot should be linear and concentration independent in the dilute range. Figures 6 and 7 show that this is the case for the more concentrated samples: The gradients at 2.0 °K corresponding to samples 200 and 95 ppm and those at 4.2 °K corresponding to 400, 200, and 95 ppm are equal within experimental precision. The more dilute samples (viz., 59–21 ppm at 2.0 °K and 59–50 ppm at 4.2 °K) show deviations from linearity because of the onset of the high-field conditions as H and hence $M^2[1+(\mu_B/\mu_{\text{eff}})^2]$ is increased.[33] (This can also be judged from the data shown in Fig. 1.) As we have pointed out above, the negative component becomes independent of H as this condition is approached because the scattering process becomes increasingly unimportant in the manifestation of the electron's dynamics. The theory relating to the freezing out of spin-flip processes does not account for this and is thus essentially a low-field theory.[33] Hence the observed deviations from linearity in Figs. 6 and 7, and hence the increasing tendency towards saturation observed for ρ_m as temperature and solute concentration are decreased.

The insert in Fig. 6 shows the effect of allowing for the magnetization of the polarized charge cloud about the solute atom. We referred above to the fact that Loram *et al.* found that M could not be fitted with a simple Brillouin function but required a g_{eff} as in Eq. (4). The same is observed in the magnetoresistance: Taking the 200-ppm alloy at 2.0 °K as an example, the insert shows $\Delta\rho/\rho_{4.2}$ plotted against M^2 using $g=2$ in the usual form $M=SgB_S$. This is compared with the modified function ($M=Sg_{\text{eff}}B_S$) which accounts for the polarized electron cloud. The improvement in linearity in the latter case is obvious, and here perhaps is indirect evidence of the influence of the Nagaoka state upon the magnetoresistance.

For what it is worth, the mean value of J, calculated by means of Eq. (2) from those data in Figs. 6 and 7 which show no appreciable effects

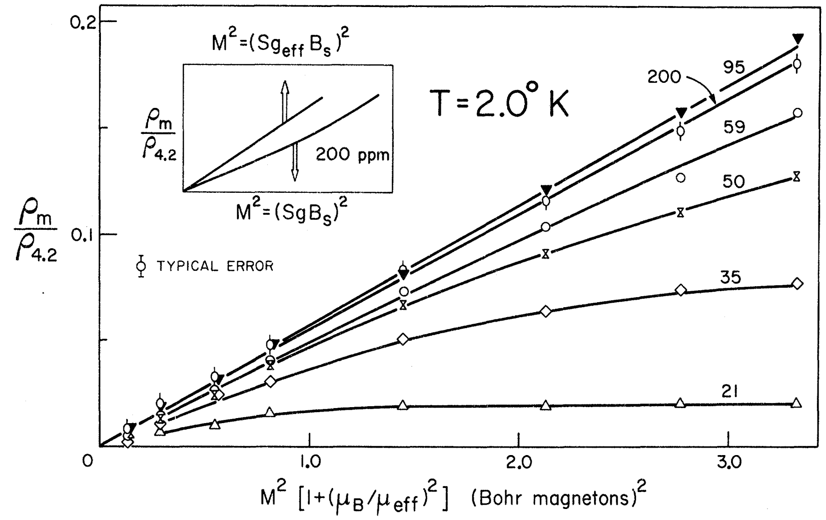


FIG. 6. Negative component per unit solute concentration obtained at 2.0 °K vs the square of the effective magnetization [Eq. (2)]. M is calculated from the data of Loram *et al.* (Ref. 1). Solute concentrations are indicated in atomic ppm. Insert shows the effect, for the example of the 200 ppm alloy at 2.0 °K, of replacing $g=2$ by g_{eff} of Eq. (4). This is introduced to account for the magnetization of the polarized electron cloud about the solute atom.

due to the onset of the high-field condition ($\omega\tau \gg 1$), is -0.81 eV. This is somewhat larger than the values (-0.31 and -0.28 eV) obtained[3,34] by other methods, and is also larger than the value (-0.26 eV) corresponding[35] to the assumed value of T_K ($=0.1$ °K). But quantitative comparisons can easily be pushed too far here: Quite apart from the experimental uncertainty in the J values obtained from Figs. 6 and 7 (probably about ± 0.06 eV), the derivation of Eq. (2) is approximate to the extent that the resulting J values should only be regarded as order-of-magnitude quantities.[26,31] However, we note that this overestimation of $|J|$ from Eq. (2) compared with other methods is carried through to other systems. Using Eq. (2), Monod[31] finds 0.91 and 0.40 eV for $-J$ in *Cu*Fe and *Cu*Mn, respectively, while magnetic susceptibility results lead[34] to corresponding values of 0.34 and 0.29 eV.

VI. CONCLUSIONS

In order to avoid solute-solute interaction effects in Kondo systems, there is an increasing tendency to study very dilute alloys. In the case of the galvanomagnetic effects at low temperatures, this can raise new problems since the experiment inevitably encompasses a dynamical intermediate-field condition; i.e., a condition which does not approximate closely to either the high- or low-field limits.[33] Physically, some of the electrons are "high field" and find themselves on cyclotron orbits for which $\omega\tau > 1$, while others, because of the peculiarities of the dominant electron scattering process, remain "low field" and are more suitably described by the usual diffusion equations. It is a region for which

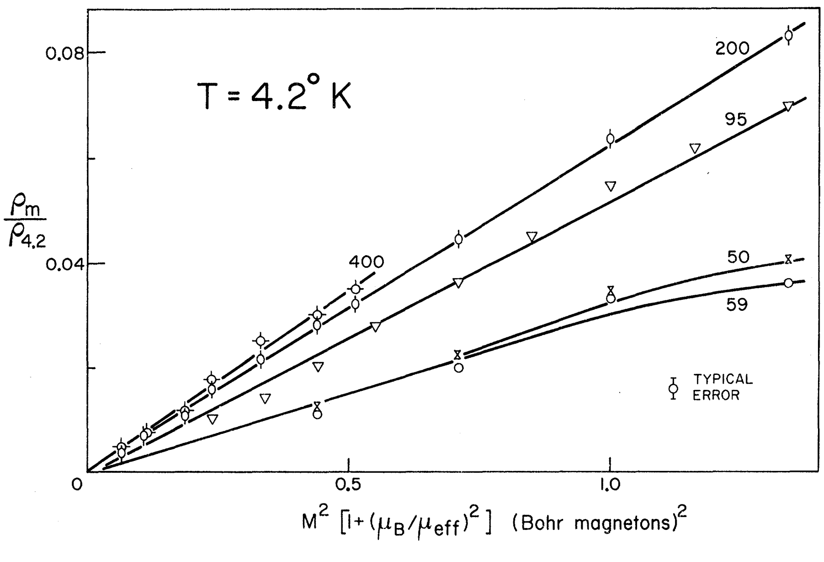


FIG. 7. Negative component per unit solute concentration obtained at 4.2 °K vs the square of the effective magnetization [Eq. (2)]. M is calculated from the data of Loram *et al.* (Ref. 1). Solute concentrations are indicated in atomic ppm.

no electron transport theory exists and therefore we doubt whether galvanomagnetic effects measured in it will be very fruitful. (In the cases of Figs. 6 and 7, for example, it is only the samples sufficiently concentrated to avoid intermediate-field effects which lead to useful results.) Nevertheless, it is important to recognize the condition in such experiments, otherwise it can lead to a misinterpretation of the data.

With the benefit of hindsight, we can say that we have probably already made this mistake in a recent analysis[11] of the transverse magnetoresistance in dilute *Cu*Mn alloys. Based upon results shown in Fig. 2 of that paper, we found an apparently anomalous dependence of the negative component upon solute concentration. But a comparison of that Fig. 2 and Figs. 6 and 7 of the present paper shows clearly the similarity between the various sets of data: The more dilute alloys in each case show evidence of the onset of high-field effects. For them the experimental condition is not the full low-field one required for the valid application of the theory of freezing out spin-flip processes, but the intermediate condition. Consequently, as we discussed in Sec. V, the deviation of ρ_m from a concentration-independent linear dependence upon H^2 is to be expected. Hence the anomalous dependence on solute concentration found for ρ_m in alloys with less than about 70 ppm (Fig. 3 of Ref. 11) is a consequence of the onset of the high-field condition in the experiment, and is not an intrinsic characteristic of the solute atom.

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[35] $T_K = T_F e^{|J\rho|}$, where T_F is the Fermi temperature and ρ is the unperturbed single-particle conduction-electron density of states. For Au the latter has a value of about 0.29 eV^{-1}.