PHYSICAL REVIEW B 1 NOVEMBER 1985

Anisotropic magnetoresistance of amorphous and concentrated polycrystalline iron alloys

A. P. Malozemoff

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598 (Received 8 July 1985)

The Campbell-Fert-Jaoul theory for anisotropic magnetoresistance is extended and a new formula derived for amorphous metalloid-containing alloys, as well as for weak ferromagnets. It accounts semiquantitatively for experimental data on amorphous Fe-B, Fe-Ni-B, and Fe-Ni-P-B, and also on concentrated polycrystalline Fe-Ni.

Anisotropic magnetoresistance¹ ($\Delta \rho / \rho$) – or more properly, spontaneous anisotropic magnetoresistance —measures the difference in resistance when the magnetization vector of a single-domain ferromagnet in zero field lies either parallel or perpendicular to the current. Arising from spinorbit coupling, 2 it is a subtle effect of considerable theoretical and applied interest.

The Campbell, Fert, and Jaoul (CFJ) two-current conduction model $3-6$ has been remarkably successful in accounting for $\Delta \rho / \rho$ in a permalloy and other *dilute* nickel-based crystalline alloys. However, the interpretation of data on more concentrated alloys^{1,4,7,8} as well as more recent data on
amorphous alloys^{9–23} has remained controversial. In this article, a new formula for $\Delta \rho / \rho$ is presented, extending the CFJ theory to such strongly disordered or weakly ferromagnetic materials. With this formula and fresh insights into the various contributions to resistivity, the first semiquantitative predictions for low-temperature $\Delta \rho / \rho$ are obtained for amorphous Fe alloys like Fe-Ni-B, Fe-Ni-PB, and Fe-B. The falloff in $\Delta \rho / \rho$ for crystalline $Ni_{1-x}Fe_x$ alloys $(0.2 < x_{\text{Fe}} < 0.4)$ is also explained in a new and consistent way.

In the two-current conduction model schematized in Fig. 1(a), conduction proceeds through two parallel channels with resistivities ρ_{\uparrow} and ρ_{\downarrow} corresponding to spin-up and spin-down conduction electrons, respectively. In the absence of spin-orbit coupling, each channel has series resistivity contributions ρ_{ss} arising from scattering between likespin s states and ρ_{sd} arising from scattering between like-

FIG. l. (a) Schematic two-current conduction model for ferromagnetic transition-metal alloys with s and d scattering. For strong ferromagnetism ρ_{sd} ₁ = 0. Usually ρ_{ss} ₁ = ρ_{ss} ₁ is assumed. (b) Schematic d density of states $N_d(E)$ for up- and down-spin bands of a strongly ferromagnetic Fe-Ni or Fe-Ni-8 alloy. For sufficient Fe, the Fermi level lies above the divide between Fe and Ni s bands.

spin s and d states; spin mixing can be ignored for highresistivity materials. ⁵

Spin-orbit coupling introduces scattering between up- and down-spin d states² with a small proportionality factor γ which is about 0.01 for the 3d transition metals.³⁻⁶ What is more, this scattering is anisotropic. In the limit of low temperature CFJ write³

$$
\rho_{\parallel \parallel} = \rho_{\perp \parallel} + \gamma \rho_{\perp \parallel} ,
$$

\n
$$
\rho_{\parallel \parallel} = \rho_{\perp \parallel} - \gamma \rho_{\perp \parallel} ,
$$
\n(1)

where ρ_{\parallel} and ρ_{\perp} refer to resistivities for current parallel and perpendicular to the field, respectively. Implicit in these formulas are two key assumptions appropriate to dilute nickel alloys: (1) ρ_1 is predominantly due to s-d scattering, i.e., $p_1 = p_{sd} + p_{sd}$ + p_{sd} ; this is why the sd label is dropped in the $\gamma \rho_{\perp \perp}$ terms above. (2) $\rho_{\perp \uparrow}$ is entirely due to s-s scattering because the strong ferromagnetism of nickel pushes spin-up d states below the Fermi surface; this is why here are no $\gamma \rho_{\perp}$ terms above. To first order in γ , CFJ use Eq. (1) to derive their classic formula for the anisotropic magnetoresistance

$$
\Delta \rho / \rho \equiv (\rho_{\parallel} - \rho_{\perp}) / \rho = \gamma [(\rho_{\perp} / \rho_{\uparrow}) - 1] \quad . \tag{2}
$$

Independent determinations of γ and ρ ₁/ ρ ₁ give excellent agreement with data on moderately dilute Ni alloys^{1,4,7} $(0.2 \ge x > 0.05)$ as illustrated for $Ni_{1-x}Fe_x$ in Fig. 2 by the dashed line. In the dilute range $\Delta \rho / \rho$ is independent of concentration for a crystal (except at the lowest concentrations, where spin mixing must be included') because both p_1 and p_1 increase linearly with x.

For the more concentrated regime and for amorphous alloys, the interpretation of $\Delta \rho / \rho$ has been less clear. Campbell,⁴ for example, suggested that if ferromagnetic weakness sets in, holes appear in the up-spin d band, causing ρ_1 to increase and $\Delta \rho / \rho$ in Eq. (2) to decrease. He explained the falloff in Fig. 2 for crystalline $Ni_{1-x}Fe_x$ (x > 0.2) in this qualitative way, even though Slater-Pauling analysis of moment data and the resistivity anomaly indicate significant magnetic weakness only for $x \ge 0.5$.²⁴ Furthermore, amorphous Ni-Fe-B and Ni-Fe-PB alloys show much reduced $\Delta \rho / \rho$ and the opposite concentration trend^{13, 19, 21} with $\Delta \rho / \rho$ *increasing* with increasing x_{Fe} , as shown in Fig. 3. Kaul and Rosenberg²¹ and Kaul²² have attempted to explain this trend, following Campbell by invoking ferromagnetic weakness with decreasing x_{Fe} even though magnetic data indicate iust the opposite, $25, 26$ namely, magnetic weakness at high x_{Fe} . As an alternative "explanation," many other authors have noticed that $\Delta \rho / \rho$ tends to track the magnetic moment

FIG. 2. Low-temperature anisotropic magnetoresistance of crystalline $Ni_{1-x}Fe_x$. Data from van Elst (Ref. 7) (+), Campbell (Ref. 4) (x) , and McGuire and Potter (Ref. 1) $(*)$, theory from Campbell et al. (Refs. 4 and 6) $(- - -)$ and this paper $(-)$.

in amorphous alloys, $12, 13, 16, 19, 21, 23$ although no justification has been given. Still others have invoked the split-band model^{$27-29$} and a scattering resonance description, but without quantitative success.

A new interpretation of $\Delta \rho / \rho$ springs from the key insight that the underlying assumptions of the crystalline nickel alloy derivation for Eq. (2) are no longer valid in amorphous alloys and weak ferromagnets. Firstly, metalloid impurities perturb the s-electron potential, causing ρ_{ss} to be strongly increased. This is apparent from many amorphous alloys with no *d* states at the Fermi surface: Systems with 15 or more percent metalloid like Pd-Si, Pd-Ge, and Ag-Cu-Ge

FIG. 3. Low-temperature anisotropic magnetoresistanceof amorphous $Ni_{0.8-x}Fe_xB_{0.2}$ (- - -) and $Ni_{0.8-x}Fe_xP_{0.14}B_{0.06}$ (--). Data from Kaul and Resenberg (Ref. 21), theory from this paper.

show resistivities in the range of 100 $\mu \Omega$ cm often increasing linearly with metalloid concentration, 30 while systems with simple metals only like Ag-Cu-Mg or Mg-Zn are usually in the range of 50 $\mu \Omega$ cm or less.³⁰ Since ρ_{ss} and ρ_{ss} act in parallel, and assuming they are equal $(\rho_{ss1} - \rho_{ss1} - 2\rho)$, we expect ρ_{ss} to be of order 200 $\mu \Omega$ cm in metalloid-containing systems.

values in the range of 100 $\mu \Omega$ cm in typical cases, which is A second insight is that for nondilute concentrations ρ_{sd} should be comparable in concentrated, amorphous, and crystalline alloys of similar composition, as long as the Fermi density of d states $N_d(E_F)$ is comparable.^{31,32} Indeed, taking ρ_{sd} as proportional to $N_d(E_F)$ times a squared matrix element, one can expect that matrix element to be similar for amorphous and crystalline systems containing Fe because of the localized nature of the d-scattering potential. $N_d(E_F)$ can be estimated empirically from electronic specific heats^{30,33} which are typically in the range of $3-7$ mJ/mole K² (taking care to correct for magnetic terms, especially where spin-glass behavior occurs). About 1 mJ/mole K^2 of this comes from the s electrons, as is evident from the electronic specific heats of non- d systems³⁰ like Pd-Si, Ag-Cu-Ge, Ag-Cu-Mg, and Mg-Zn, which range from 0.7 to 1.2 mJ/mole K². Thus, in what follows, $N_d(E_F)$ for various systems will be scaled to specific-heat data by simply subtracting off 1 mJ/mole K². As will be seen, this implies ρ_{sd} comparable to what CFJ would estimate for 15% Fe in Ni, for example.³ Clearly, ρ_{ss} equals if not exceeds ρ_{sd} in the resistor network of Fig. 1(a) for amorphous materials, in contrast to the crystalline case where $\rho_{sd} \gg \rho_{ss}$. In this case, the aproximations used in deriving Eq. (2) from Eq. (1) must be revised, and instead of $\rho \sim \rho_{ss}$ one has

$$
\rho = (\rho_{ss} + \rho_{sd\uparrow})(\rho_{ss} + \rho_{sd\downarrow})/(2\rho_{ss} + \rho_{sd\uparrow} + \rho_{sd\downarrow})
$$
 (3)

The third insight is that for ferromagnetically weak systems where $\rho_{sd} \neq 0$, Eq. (1) must be generalized to

$$
\rho_{\parallel \parallel} = \rho_{\perp \parallel} + \gamma (\rho_{sd\perp \parallel} - \rho_{sd\perp \parallel}),
$$

\n
$$
\rho_{\parallel \parallel} = \rho_{\perp \parallel} - \gamma (\rho_{sd\perp \parallel} - \rho_{sd\perp \parallel}),
$$
\n(4)

where

$$
\rho_{\perp\uparrow} = \rho_{ss\uparrow} + \rho_{sd\perp\uparrow}
$$

and so forth. Combining these equations to first order in γ but with no approximations on the relative sizes of ρ_{ss} and p_{sd} leads to the principal result of this paper:

$$
\frac{\Delta \rho}{\rho} = \frac{\gamma (\rho_{sd} - \rho_{sd\uparrow}) (\rho_{sd\downarrow} + \rho_{ss\downarrow} - \rho_{sd\uparrow} - \rho_{ss\uparrow})}{(\rho_{sd\downarrow} + \rho_{ss\downarrow}) (\rho_{sd\uparrow} + \rho_{ss\uparrow})} \quad . \tag{5}
$$

For strong ferromagnets and $\rho_{ss\uparrow} = \rho_{ss\downarrow}$, this simplifies to

$$
\Delta \rho / \rho = \gamma \rho_{sd}^2 / \rho_{ss} (\rho_{ss} + \rho_{sd}) \quad . \tag{6}
$$

 $\Delta p/p = \gamma p_{sd} \gamma / p_{ss} \sqrt{p_{ss} + p_{sd}}$
Obviously, if $\rho_{sd} \gg \rho_{ss}$, this reduces to the earlier CFJ esult of Eq. (2). But these formulas differ significantly from Eq. (2) for either weak ferromagnets or amorphous materials with $\rho_{sd} \sim \rho_{ss}$. As we shall now see, these new results permit semiquantitative agreement with experiment in many cases.

For crystalline $Ni_{1-x}Fe_x$, Eq. (2) is still valid, but in the range $0.2 < x_{\text{Fe}} < 0.4$, where $\Delta \rho / \rho$ drops off abruptly, ρ_{sd} and ρ_{ss} are no longer proportional as they were in the diind ρ_{ss} are no longer proportional as they were in the di-
ute limit. Indeed, although $\rho \sim \rho_{ss}$ continues to increase almost linearly³⁴⁻³⁶ ($\sim 20x_{\text{Fe}} \mu \Omega \text{ cm}$), electronic specific

heat (4 mJ/mole K² for $0.2 < x_{\text{Fe}} < 0.4$) indicates N_{d} ₁(E_F) is essentially constant, 37 in agreement with band calculations.³⁸ The well-established split-band picture^{27, 28} for these alloys, shown in Fig. 1(b), implies that above $x_{Fe} = 0.2$, the Fermi level moves up into the down-spin iron d band. The area of this band in Fig. 1(b) will go up as x_{Fe} while the energy width also increases as x_{Fe} (because of Fe-Fe interactions), explaining the constant state density provided the band profile is flat as a function of energy.

To determine the *d*-scattering matrix element, the $\Delta \rho / \rho$ To determine the *a*-scattering matrix element, the $\Delta \rho / \rho$
data are fitted at $x_{Fe} \sim 0.3$, using Eq. (2) and the above values of ρ_{ss} and $\gamma (=0.01)$. This gives $\rho_{sd} = 72.5 \mu \Omega$ cm independent of concentration, which in turn implies the theory prediction (solid lines) of Fig. 2. The falloff in $\Delta \rho / \rho$ can now be understood primarily as a consequence of the increase of $p = \rho_{ss}$. The further decrease beyond $x_{Fe} = 0.4$ can now be more plausibly attributed to magnetic weakness, though no quantitative prediction using Eq. (5) is attempted here.

The same kind of analysis, applied to amorphous $Ni_{0.8-x}Fe_xB_{0.2}$ and $Ni_{0.8-x}Fe_xP_{0.14}B_{0.06}$, reveals just the opposite behavior from the crystalline alloys: ρ_{ss} is essentially constant while ρ_{sd} varies as a function of x. The ρ_{sd} variation is revealed by the electronic specific heats, $30,33$ which tion is revealed by the electronic specific heats, ^{30, 33} which
track the $\Delta \rho/\rho$ trends remarkably well.²¹ ρ_{ss} can be determined from Eq. (3) using the values $\rho = 120$ and 140 $\mu \Omega$ cm for the B_{0.2} and P_{0.14}B_{0.06} series, resepctively;³⁰ within error bars these appear to be independent of x_{Fe} . Once again, to determine ρ_{sd} and the matrix element, Eq. (6) is fitted to the data at a single point $(Fe_{0.8}P_{0.14}B_{0.06})$, and the other ρ_{sd} values are scaled for both series from the measured specific heats as described earlier. The predicted $\Delta \rho / \rho$ from Eq. (6), shown as "theory" in Fig. 3, is in remarkable agreement with the data. What is more, the (squared) matrix element deduced from the fitting point is only 25% larger than that determined for crystalline $Ni_{1-x}Fe_{x}$. In effect, this means the amorphous $\Delta \rho / \rho$ can be predicted from crystalline data and vice versa.

Thus, the underlying cause of the trends in Fig. 3 is the density-of-states profile of the down-spin Fe band [see Fig. 1(b)]. That this is not directly related to magnetization (as many authors have attempted to prove) is emphasized by differing profiles for cystalline and amorphous analogs, reflected in the differing electronic specific heats. While the crystalline profile was flat in the range of interest, the amorphous one is peaked, which is most likely due to hybridization with antibonding metalloid sp states.³⁹

 $Fe_{1-x}B_x$ shows a low temperature $\Delta \rho / \rho$ which peaks at 0.9% for $x_B \sim 0.15$ and drops off to 0.35% for $x_B = 0.25^{16}$ Again electronic specific heat tracks this trend, 30 dropping from 9 to 5.8 MJ/mole K^2 at the two concentrations, while ρ increases only slightly from 116 to 125 $\mu \Omega$ cm.⁴⁰ With the same ρ_{sd} scaling as before and Eq. (3) to determine ρ_{ss} , $\Delta\rho/\rho$ comes out 0.88 and 0.31% for the two compositions, espectively, in excellent agreement with experiment. The alloff for $x_B < 0.15$ is presumably due to magnetic weakness.

Actually, there is evidence⁴¹ that weakness sets in already for $x_B \le 0.20$. At $x_B = 0.15$, for example, the actual atomaveraged moment μ_a of 1.85 μ_b , differs from the hypothetical strong moment μ_s of 2.1 μ_B , implying 0.125 holes per atom in the up-spin d band (see Ref. 41, Fig. 13, Z_m = 1.25). To calculate the effect on $\Delta \rho / \rho$ we must know the band shape; let us take as a model assumption $N_d(E) \propto (E_t - E)^n$, where E_t is the top of the band and n is some power, In this case, it is easy to show that for rigidband exchange splitting in a Stoner model, the ratio of up and down d-state densities at the Fermi level is $[(\mu_s - \mu_a)/(\mu_s + \mu_a)]^{n/n+1}$. Using this ratio to determine p_{sd} and p_{sd} from the specific heat as before, and using Eqs. (3) and (5), we find $\Delta\rho/\rho = 0.50\%$ for $n = 2$. Results are sensitive to the band-shape parameter n and increase to 0.71% as $n \rightarrow \infty$. While high *n* plausibly mimics a band tail at the top of the band, uncertainty about the actual band shape prevents a more conclusive comparison with experiment.

In summary, the new formulas Eqs. (5) and (6) and a more careful consideration of the different contributions to resistivity appear to account for the anisotropic magnetoresistance of a variety of amorphous as well as crystalline Fe-containing alloys in a consistent way.

The author thanks L. Berger for first pointing out the importance of metalloids on the ss scattering. He acknowledges valuable conversations with T. R. McGuire, R. J. Gambino, M. Continentino, K. Fukamichi, T. Egami, R. Hasegawa, H. Bernas, J. Hafner, T. Massalski, and I. A. Campbell.

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