Magnetoresistance of PdFe and PdNiFe alloys

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Magnetoresistance of Pd-Fe and Pd-Ni-Fe alloys[a)]

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The electrical resistivity of Pd-Fe alloys has been measured in magnetic fields parallel and perpendicular to the current, for a wide range of alloy compositions. The influence of atomic ordering on the magnetoresistance has been investigated. The ferromagnetic anisotropy of resistance $\Delta\rho/\rho_0$ reaches a maximum value of $+9\%$ in the ordered state between 30 and 35 at. % Fe, and $+6\%$ in the disordered state between 40 and 60 at. % Fe, at 4.2 K. At 295 K, $\Delta\rho/\rho_0$ never exceeds $+1\%$. Similar measurements have been done on $(Pd_{50}\,Ni_{50})_{100-x}\,Fe_x$ for $x \leqslant 30$ at. % Fe. The maximum $\Delta\rho/\rho_0$ value of $+7\%$ is obtained for 10 at. % Fe at 4.2 K.

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I. INTRODUCTION

In most metallic ferromagnets, the electrical resistivity $\rho_\parallel$, measured in the saturated state with the magnetization $\mathbf{M}_s$ parallel to the current, is larger than the resistivity $\rho_\perp$ obtained when $\mathbf{M}_s$ is perpendicular to the current. The so-called ferromagnetic anisotropy of resistivity $\Delta\rho/\rho_0$ is defined[1,2] by

$$\frac{\Delta\rho}{\rho_0} = \frac{\rho_\parallel - \rho_\perp}{\rho_0}, \tag{1}$$

where $\rho_0 = (1/3)\rho_\parallel + (2/3)\rho_\perp$ is approximately equal to the resistivity in the demagnetized state at zero external field. Usually, $\Delta\rho/\rho_0$ is found to take larger values for impurity scattering than for scattering by phonons, magnons or lattice defects. Smit has proposed a rather successful theory[3] of $\Delta\rho/\rho$: in the presence of spin-orbit interaction, the total scattering cross section presented by a transition-metal impurity depends on the angle between the electron spin and the momentum of the incoming conduction electron.

Campbell, Fert, and Jaoul[4] have shown experimentally that the value of $\Delta\rho/\rho_0$ in dilute nickel alloys is an increasing function of the ratio $\rho_\downarrow/\rho_\uparrow$. Here, $\rho_\uparrow$ and $\rho_\downarrow$ are the resistivities for spin-up and spin-down electrons, which can be derived[4,5] from the observed deviations from Matthiessen's rule. This dependence of $\Delta\rho/\rho_0$ on $\rho_\downarrow/\rho_\uparrow$ is correctly predicted by Smit's theory.

The large values $\Delta\rho/\rho_0 \simeq +20\%$ reached[3] in fcc $Ni_{85}Fe_{15}$ and $Ni_{75}Co_{25}$ at low temperatures [Fig. 1(b)] are of scientific as well as technological interest. Note also the value $\Delta\rho/\rho_0 \simeq +12\%$ for bcc $Fe_{94}V_6$ at low temperatures.[6] From existing data and theory, we can state[7] the following conditions to obtain large $\Delta\rho/\rho_0$.

(a) Scattering by impurities (i.e., by minority components of the alloy) is dominant.

(b) The alloy is among those following the left-side or right-side part of the Slater–Pauling curve for saturation magnetization data.

(c) The impurities are located to the left of nickel in the periodic table in nickel-rich alloys, or to the left of iron in iron-rich alloys.

(d) The concentration of these impurities is lower than $\simeq$20 at. %, so that the spin-down Fermi level is still located[7] in the d band of the matrix (i.e., of the majority component such as nickel), rather than in the impurity band (e.g., Fe↓ band). See Figs. 1(a) and 1(b).

Condition (b) minimizes $\rho_\uparrow$ in Ni-rich alloys, by insuring that the spin-up d band is full. On the other hand, conditions (c) and (d) have the effect of maximizing $\rho_\downarrow$ by creating a scattering resonance[7] at the Fermi level in the spin-down d band. Indeed, a calculation of s-d scattering probability, based on the Slater–Koster formalism for dilute alloys, predicts[7] the existence of such a scattering resonance in the case of a repulsive scattering potential. The existence and location of that resonance is directly confirmed by the data of Szentirmay and Kedves,[8] which show a large peak of $\rho_\downarrow$ at the composition $Ni_{85}Fe_{15}$. A somewhat similar peak of $\rho_\downarrow$ is predicted by coherent-potential calculations done by Akai,[9]

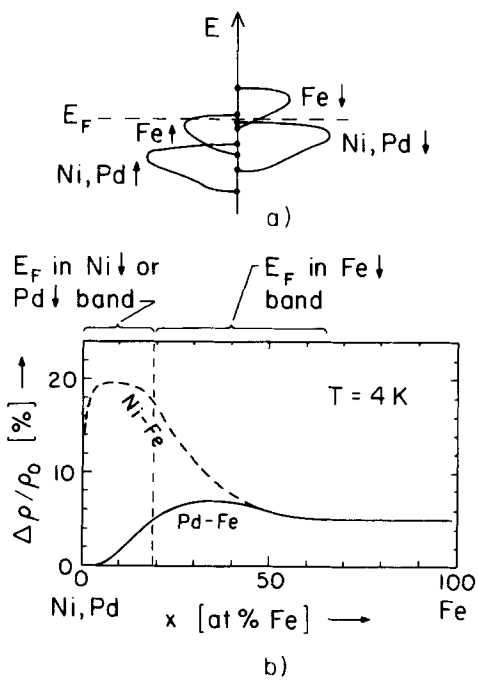

FIG. 1. (a) Band structure of Ni-Fe or Pd-Fe alloys, with distinct iron and nickel (or palladium) d bands, according to Ref. 7. (b) Magnetoresistance of $Ni_{1-x}Fe_x$ alloys. It is very large for $x < 20$ at. % Fe, when the Fermi level is in the Ni↓ band. The expected magnetoresistance of Pd-Fe is smaller in that range due to incomplete filling of the spin-up bands.

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but without clear explanation of its location and physical origin.

The failure of condition (c) explains partially why $\Delta\rho/\rho_0$ is usually smaller in pure nickel than in Ni-Fe [see left end of Fig. 1(b)].

The physical and magnetic properties of Pd-Fe alloys are similar to those of Ni-Fe alloys. One difference between these two series is that the saturation magnetization of Pd-Fe falls[10] below the linear Slater–Pauling curve, for iron concentrations below 20 at. %. This indicates that the spin-up d band is not full in that composition range, which should lead to an increase of $\rho_\uparrow$ and, consequently, to a decrease of $\rho_\downarrow/\rho_\uparrow$. In turn [see condition (b) above], this should cause $\Delta\rho/\rho_0$ to be smaller in Pd-Fe than in Ni-Fe, in the range below 20 at. % Fe.

In other words, the large $\rho_\downarrow/\rho_\uparrow$ and $\Delta\rho/\rho_0$ values observed in Ni-Fe below 20 at. % Fe, caused by the $\rho_\downarrow$ resonance discussed earlier, may disappear in Pd-Fe because of the large $\rho_\uparrow$. This predicted behavior is shown in Fig. 1(b).

Existing magnetoresistance data[11–13] in Pd-Fe cover only the range up to 35 at. % Fe, and only for $T = 1.5$ K. The purpose of the present investigation is to obtain $\Delta\rho/\rho_0$ data for the whole Pd-Fe series, at several temperatures. We have already published[14] a detailed study of the effect of atomic ordering on $\Delta\rho/\rho_0$ in $Pd_{70}Fe_{30}$, $Pd_{75}Fe_{25}$, and $Pd_{50}Fe_{50}$.

Although very little information is available about Pd-Ni-Fe alloys, we can expect their properties to be intermediate between those of Ni-Fe and of Pd-Fe.

II. EXPERIMENTAL DETAILS

Sample preparation and measurement techniques were already described in Ref. 14. However, among the final thermal treatments listed in Ref. 14, only the two treatments labeled (a) and (d) are used in the present work. Treatment (a) consists of an anneal for 2 h in a vacuum at 1100 °C, followed by quenching in water under atmospheric pressure. Treatment (d) is an anneal for 20 h in a vacuum at 550 °C, followed by rather slow furnace cooling; however, a temperature $\leqslant$400 °C was used in the case of the Pd-Fe sample with 20 at. % Fe because of its lower ordering temperature. Treatment (a) is designed to minimize the degree of atomic ordering, and treatment (d) to maximize it. Extensive studies[15–17] show that fcc Pd-Fe alloys between $\simeq$20 and $\simeq$37 at. % Fe tend to assume an fcc ordered state of the Cu_3 Au type, which we call the Pd_3Fe state. Between $\simeq$37 and $\simeq$55 at. % Fe, an ordered face-centered tetragonal state of the CuAu type is possible, which we call the PdFe state. However, between $\simeq$55 and 98 at. % Fe, a bcc phase coexists with the face-centered phases. Above 98 at. % Fe, the bcc phase exists alone.

We have obtained x-ray diffraction and electrical resistance data[14] showing that a $Pd_{70}Fe_{30}$ sample quenched in oil, although not yet exhibiting any superlattice Bragg reflections corresponding to the Pd_3Fe ordered state, already has a substantially lower value of the resistivity ρ_0 than a sample quenched in water. This suggests that physical properties such as the resistivity are sensitive not only to long-range order, but even to short-range order present in the oil-quenched sample. For that reason, we have used ρ_0 rather than x-ray data, as an indicator of the degree of order of our samples. For the same reason, we have quenched our samples in water rather than in oil.

These ρ_0 data, together with existing x-ray studies of ordering kinetics in Pd-Fe (Refs. 15,16) suggest that treatments (a) and (d) yield, respectively, complete disorder and substantial degree of order in most cases.

The external magnetic field $\leqslant$0.3 T used during measurements is large enough to saturate most samples, in a direction parallel or perpendicular to the current. Due to the tetragonal distortion of the PdFe fct ordered state, ordered samples between 37 and 98 at. % Fe have a large uniaxial magnetic anisotropy and are very difficult to saturate.[18] Thus, for compositions in the range 55–98 at. % Fe, which have mixed face-centered and bcc phases, we have measured $\Delta\rho/\rho_0$ and ρ_0 only in the disordered state, i.e., after treatment (a). However, for the (more interesting) purely face-centered compositions 40 and 50 at. % Fe, data in the ordered state at 4.2 K were obtained with the help of a superconducting magnet reaching 6.2 T. This field was large enough to saturate the samples.

The Pd-Ni-Fe alloys were prepared in the same fashion as the Pd-Fe. The liquidus and solidus curves are available in the literature.[19] Since the atomic ordering temperatures are not known, treatment (d) was carried out for 25 h at temperatures declining gradually from 550 to 330 °C.

III. EXPERIMENTAL RESULTS

Measured values of ρ_0 for Pd-Fe at 4.2, 77, and 295 K are given in Fig. 2. At 77 and 4.2 K, ρ_0 is always larger in the disordered state [treatment (a)] than in the ordered state [treatment (d)]. Paradoxically, at 295 K the opposite is true for compositions between 30 and 35 at. % Fe. As noted earlier,[14] this implies that Pd_3Fe ordering does more than reduce electron scattering by the alloy disorder; it also changes the band structure by introducing new band gaps. In Fig. 2,

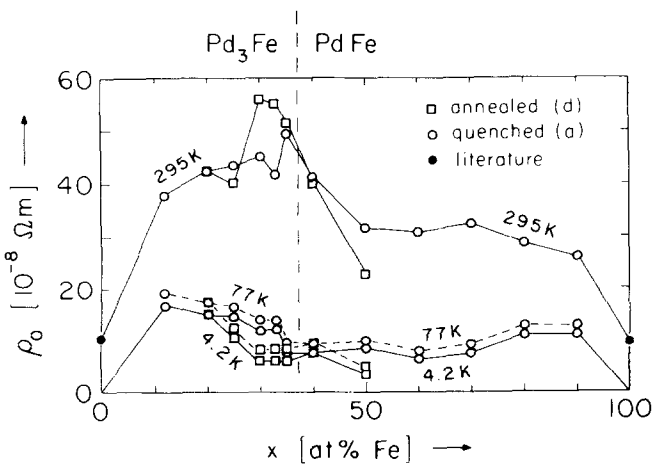

FIG. 2. Electrical resistivity of $Pd_{1-x}Fe_x$ alloys. Treatment (d) results in atomic order of Pd_3Fe or PdFe type. Treatment (a) results in atomic disorder.

TABLE I. Measured values of density and of saturation magnetization at 295 K.

$Pd_{100-x}Fe_x$		$(Pd_{50}Ni_{50})_{100-x}Fe_x$		$M_s(T)$	
x(at. %)	δ (10^3 kg/m^3)	x(at. %)	δ (10^3 kg/m^3)	Treat. (a)	Treat. (d)
70	8.957	10	10.473	0.65	0.66
80	8.582	20	10.147	0.88	0.90
90	8.359	30	9.976	1.15	1.16

we also show ρ_0 values for pure Pd and pure Fe, taken from tables of constants.

Our ρ_0 values in Pd-Fe are in reasonable agreement with those obtained by other authors.[20]

We have measured the density δ of some Pd-Fe samples by the buoyancy method. It agrees within $\pm 2\%$ with the value derived from fcc or fct lattice parameters in the composition range where these are available.[21] In Table I, we give our measured δ in the range 70–90 at. % Fe where no previous information existed.

The measured values of $\Delta\rho/\rho_0$ for Pd-Fe are shown in Fig. 3, at 4.2, 77, and 295 K. At 295 K, $\Delta\rho/\rho_0$ is smaller than $+1\%$ in all cases. It increases considerably as T decreases and impurity scattering becomes dominant [condition (a)]. In the 30, 33, and 35 at. % Fe samples at 4.2 and 77 K, the effect of ordering of the Pd_3Fe type is to increase $\Delta\rho/\rho_0$ considerably, as reported earlier.[14] Values $\Delta\rho/\rho_0 \simeq +9.5\%$ are reached. On the other hand, the effect of Pd_3Fe ordering on the 25 at. % Fe sample at the same temperatures is to decrease $\Delta\rho/\rho_0$ somewhat (Fig. 3). A similar reversal of the effect of ordering on $\Delta\rho/\rho_0$, with varying composition, was observed in Ni-Fe by Kondorskii and Ozhigov.[22] Data for samples with 40 and 50 at. % Fe show that ordering of the PdFe type causes a small decrease of $\Delta\rho/\rho_0$ at 4.2 K (Fig. 3).

In Fig. 4 we give our measured values of ρ_0 for $(Pd_{50}Ni_{50})_{100-x}Fe$ at 4.2, 77, and 295 K. On the same figure, we show ρ_0 values of $Pd_{50}Ni_{50}$ at about the same temperatures, from the literature.[23]

The data show that the nature of the thermal treatment has little effect on ρ_0. This is somewhat surprising, since an atomic ordering similar to Pd_3Fe is known[24] to exist below 503 °C in Ni-Fe for compositions between 20 and 40 at. % Fe. As in Ni-Fe, it might be very sluggish, so that our samples remain disordered.

Our $\Delta\rho/\rho_0$ data for $(Pd_{50}Ni_{50})_{100-x}Fe_x$ at 4.2, 77, and 295 K are shown on Fig. 5. The investigation is limited to the range $x \leqslant 30$ at. % Fe because this range is most likely to yield large $\Delta\rho/\rho_0$ values [see condition (d)]. Again, the type of thermal treatment has little effect on $\Delta\rho/\rho_0$. The maximum value observed is $\Delta\rho/\rho_0 \simeq +7\%$ in $(Pd_{50}Ni_{50})_{90}Fe_{10}$ at 4.2 K. We also show the $\Delta\rho/\rho_0$ value for $Pd_{50}Ni_{50}$ at 4.2 and 295 K, from the literature.[25,11]

The density and saturation magnetization of these ternary alloys have been measured at 295 K and are given in Table I.

IV. DATA INTERPRETATION

The $\Delta\rho/\rho_0$ data for Pd-Fe (Fig. 3) are more or less in agreement with the ideas described in the introduction. As expected, the large $\Delta\rho/\rho_0$ values present in Ni-Fe below 20 at. % Fe, caused by the $\rho_\downarrow$ resonance, are absent [Fig. 1(b)] in Pd-Fe because of the large $\rho_\uparrow$ associated with the violation of condition (b). Some vestige of the edge of the $\rho_\downarrow$ scattering resonance may remain visible around 30–40 at. % Fe, and explain the somewhat larger $\Delta\rho/\rho_0$ values observed there.

The low $\Delta\rho/\rho_0$ values observed below 20 at. % Fe had

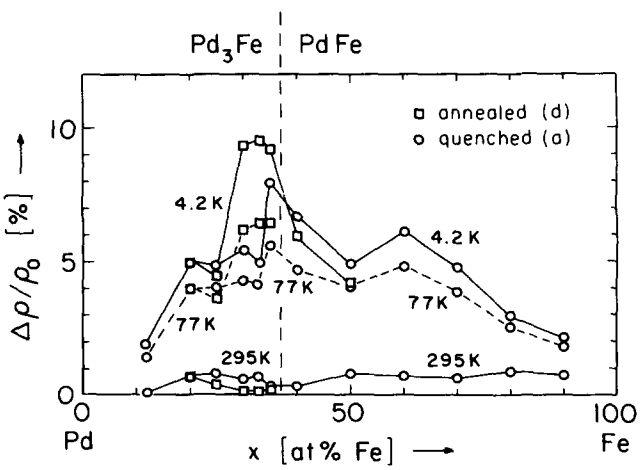

FIG. 3. Magnetoreistance of $Pd_{1-x}Fe_x$ alloys. Treatment (d) results in atomic order of Pd_3Fe or PdFe type. Treatment (a) results in atomic disorder.

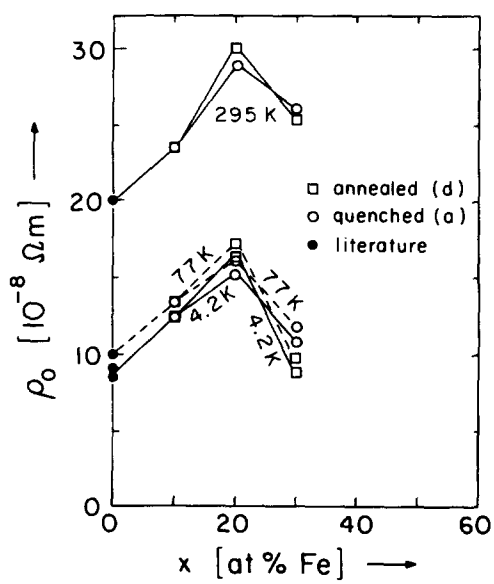

FIG. 4. Electrical resistivity of $(Pd_{50}Ni_{50})_{1-x}Fe_x$ alloys.

already been discovered by Senoussi, Campbell, and Fert.[11]

The effect of Pd_3Fe ordering on $\Delta\rho/\rho_0$ at 4.2 and 77 K, including its sign reversal with varying composition, can also be explaind in terms of condition (b). Ryzhenko, Sidorenko, Karpov, and Gel'd[26] have found that Pd_3Fe ordering increases the saturation magnetization M_s appreciably at $x \geqslant 30$ at. % Fe, but decreases it somewhat at 25 at. % Fe (Fig. 6). Incidentally, the effect of ordering on the hyperfine field at Fe nuclei, although of the opposite sign, also undergoes a sign reversal between $\simeq$30 and 25 at. % Fe (Fig. 6).[26] Now, a M_s increase would correspond to a more complete filling of the spin-up d band, bringing M_s in better agreement with the Slater–Pauling curve. In turn, by condition (b), a larger $\Delta\rho/\rho_0$ should result. This agrees with our low-temperature experimental findings for Pd_3Fe ordering at 30–35 at. % Fe. Similarly, the M_s decrease caused by Pd_3Fe ordering at 25 at. % Fe should cause a drop of $\Delta\rho/\rho_0$, as is actually observed (Fig. 3) at low temperatures. A different explanation was given by us in Ref. 14.

At 295 K, the effect of Pd_3Fe ordering on $\Delta\rho/\rho_0$ is opposite to the effect at 77 or 4.2 K (Fig. 3). This is explained by an additional mechanism: atomic ordering depresses impurity scattering as compared to phonon scattering. By condition (a), this tends to reduce $\Delta\rho/\rho_0$.

We now try to explain the small $\Delta\rho/\rho_0$ decrease caused by ordering of the PdFe type (Fig. 3). Neutron diffraction data and Mossbauer effect data[26] suggest that a ferrimagnetic spin arrangement exists in the ordered state at zero field. This is evidence of antiferromagnetic interactions between Fe and Pd atoms in the ordered state, and of the failure of strong ferromagnetism as represented by the Slater–Pauling curve. Then, by condition (b), $\Delta\rho/\rho_0$ is expected to be smaller in the ordered state, in agreement with our data.

As expected, the behavior of $\Delta\rho/\rho_0$ in $(Pd_{50}Ni_{50})_{100-x}Fe_x$ below 20 at. % Fe (Fig. 5) is intermediate between that of Pd-Fe and that of Ni-Fe. As noted earlier, our (PdNi)-Fe samples are probably always disordered. Hence, they should be compared to disordered Pd-Fe and Ni-Fe, displayed in Fig. 1(b).

The abrupt drop of $\Delta\rho/\rho_0$ between $(Pd_{50}Ni_{50})_{90}Fe_{10}$ and $Pd_{50}Ni_{50}$ (Fig. 5) is a good demonstration of the validity of our condition (c): an impurity (here Fe) located to the left of Ni or Pd in the periodic table is needed for large $\Delta\rho/\rho_0$.

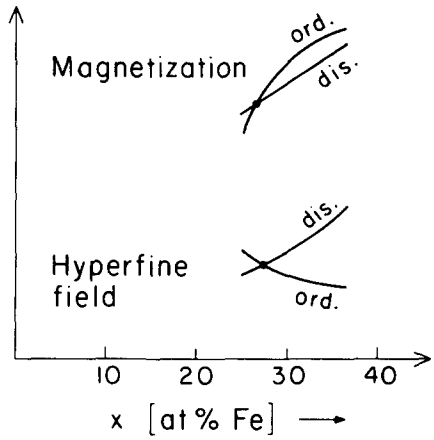

FIG. 6. Effect of Pd_3Fe atomic order on the saturation magnetization and on the Fe hyperfine field in $Pd_{1-x}Fe_x$ changes sign between 25 and 30 at. % Fe, according to Ref. 26.

V. CONCLUSIONS

Although the physical properties of Pd-Fe are very similar to those of Ni-Fe alloys, the large $\Delta\rho/\rho_0 \simeq 20\%$ values observed in Ni-Fe below 20 at. % Fe are absent in Pd-Fe because their magnetization drops below the Slater–Pauling curve. At 4.2 K, the variation of $\Delta\rho/\rho_0$ with atomic ordering of Pd_3Fe type also tracks the variation of the magnetization with ordering. Ordered alloys with 30–35 at. % Fe have a $\Delta\rho/\rho_0$ reaching $+9.5\%$ at 4.2 K. At 295 K, all $\Delta\rho/\rho_0$ values in Pd-Fe are below $+1\%$. In the $(Pd_{50}Ni_{50})_{100-x}Fe_x$ series, the maximum $\Delta\rho/\rho_0$ reached is $+7\%$, for 10 at. % Fe at 4.2 K.

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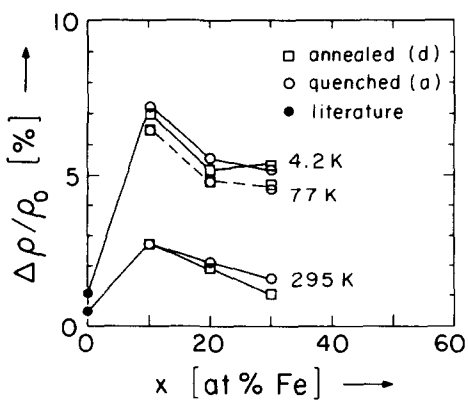

FIG. 5. Magneotoresistance of $(Pd_{50}Ni_{50})_{1-x}Fe_x$ alloys. Its dependence on x is intermediate between that of $Ni_{1-x}Fe_x$ and that of $Pd_{1-x}Fe_x$.

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