the magnetization on the same sample will be most useful in verifying our suggestion that $R(H) - R(0)$ and $\rho(H) - \rho(0)$ are proportional to each other and to the square of the magnetization.[11]

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PHYSICAL REVIEW B VOLUME 3, NUMBER 9 1 MAY 1971

Temperature-Dependent Scattering in Paramagnetic *Pd*Co Alloys*

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Measurements are presented of the resistivities of some dilute *Pd*Co alloys at temperatures above the magnetic-ordering temperature. The excess resistivity $\Delta\rho$ can be represented by the expression $\Delta\rho = A + B \ln T$, where B is positive and is proportional to the Co concentration. Magnetoresistance measurements on one of the alloys are also presented. It is shown that an implausibly large positive value for the exchange coupling between s electrons and local spins is required to account for the magnetoresistance and zero-field measurements if the temperature dependence is assumed to result from a Kondo scattering of the s electrons from local Co moments. We conclude that the mean-square moment on the impurity site may be temperature dependent, due either to partial spin compensation by the itinerant d electrons or localized spin fluctuations on the Co sites, and that this leads to a temperature-dependent scattering of the s electrons and the resistivity behavior which we observe.

INTRODUCTION

It is well known that the alloy systems *Pd*Fe and *Pd*Co which exhibit the giant-moment phenomenon[1-4] also exhibit ferromagnetic ordering below a Curie temperature T_C, which increases rapidly with Fe or Co concentration.[5] Many of the properties of these systems show anomalous features, but perhaps none more striking than the electrical resistivity for which a sharp change in slope occurs in the neighborhood of T_C; below T_C the resistivity decreases rapidly as the temperature is further reduced.[6-9] At temperatures above the Curie temperature, in the *Pd*Fe system, the incremental resistivity $\Delta\rho(T) = \rho_{\text{alloy}}(T) - \rho_{\text{Pd}}(T)$ in this paramagnetic region is almost temperature independent below 10 K, but in the *Pd*Co system[9] a substantially larger temperature dependence is observed.[10] In this paper we examine the temperature dependence of $\Delta\rho(T)$ above the Curie temperature in greater detail, in *Pd*Co alloys containing 0.05-, 0.1-, and 0.2-at.% Co. Alloys of higher Co concentration were not included in this investigation as their Curie temperature falls in a temperature region where the "pure" Pd resistivity is increasing rapidly, so that uncertainities resulting from the breakdown of

Matthiessen's rule[11,12] make the determination of $\Delta\rho(T)$ above T_C unreliable.

For the alloys here examined, the incremental resistivity $\Delta\rho(T)$ can be represented, at temperatures above the magnetic-ordering temperature, by the expression $\Delta\rho(T) = A + B \ln T$, where B is positive and proportional to the concentration of the Co impurities. This result conclusively demonstrates that the observed contribution to the incremental resistivity derives from conduction electrons scattering from essentially isolated impurities. The possible origins of such effects are discussed in the light of current theories.

EXPERIMENTAL DETAILS

The alloys were prepared by "diluting" the appropriate amounts of a Pd 5-at.% Co master alloy with pure Pd; this master alloy was itself prepared from 99.999% pure Pd (Johnson Matthey and Co., London) and 99.99% pure Co (Koch-Light Ltd., England) by a method previously described.[8,9] The resistivity samples were in the form of carefully etched and homogenized strips, approximately 0.01 cm thick, 0.2 cm wide, and 8 cm long; their resistance was measured using a four-probe technique in which the current through the sample was varied to balance a highly stable voltage. A Keithley 149 Millimicrovoltmeter was used to detect the balanced condition. Reproducible measurements to $\pm 10^{-9}$ V were achieved by measuring the specimen voltage and current using Hewlett-Packard digital voltmeters, leading to a resistance measurement accurate to approximately 1 part in 10^4. The area-to-length ratios of the specimens were measured to an accuracy of $\pm 0.5\%$ using a method described by Loram *et al.*,[13] thus enabling reliable estimates of the incremental resistivity to be made.

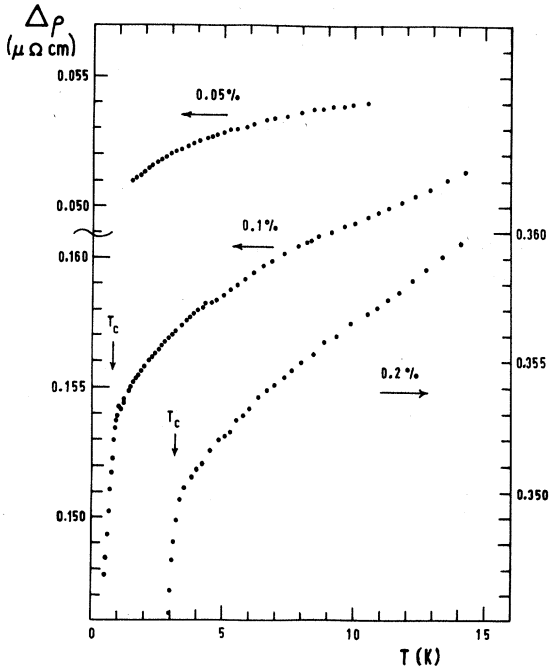

FIG. 1. Excess resistivity $\Delta\rho$ as a function of temperature for *Pd*Co alloys containing 0.5-, 0.10-, 0.20-at.% Co. Relevant resistivity scales for each alloy are indicated by the arrow.

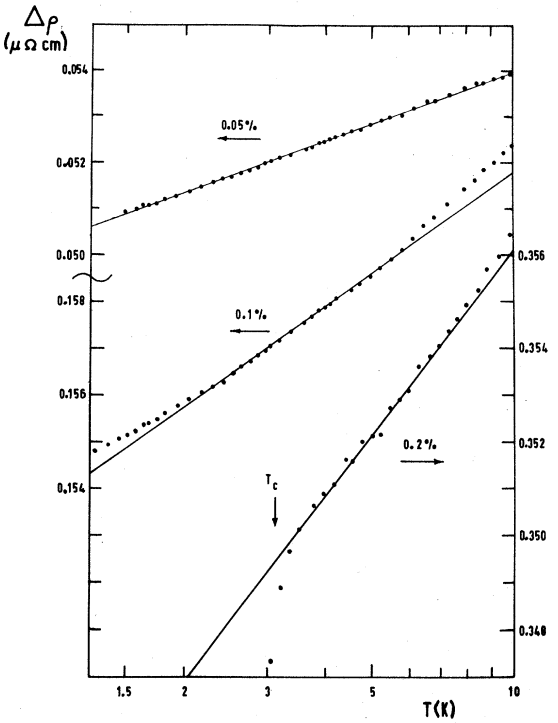

FIG. 2. Excess resistivity $\Delta\rho$ as a function of temperature (on a logarithmic scale) for the *Pd*Co alloys.

Temperature stabilization was achieved by using a carbon resistance thermometer in a feedback control circuit, which allowed temperatures below 4.2 K to be stabilized and measured (via the He^4 vapor pressure) to ± 1 mdeg and above 4.2 K to better than $\pm 1\%$ (using a gas thermometer).

Magnetoresistance measurements were made by mounting the sample in the longitudinal field of a superconducting solenoid, the latter being locked in its persistent mode during the course of the measurements. The applied magnetic field, measured to within ± 0.1 kOe using a Hall probe, varied by approximately 2 parts in 10^3 over the length of the specimen.

RESULTS AND DISCUSSION

From the measurements taken it was possible to estimate the incremental resistivity $\Delta\rho(T) = \rho_{\text{alloy}}(T) - \rho_{\text{Pd}}(T)$ due to the presence of the Co impurities as a function of temperature. Figure 1 shows $\Delta\rho(T)$ plotted against temperature T for the three alloys investigated; the Curie temperature T_C occurs at

TABLE I. Estimates of the coefficients of the "ln" term.

Alloy[a]	Estimate of B(in nΩ cm)
Pd 0.05-at.% Co	1.5±0.1
Pd 0.1-at.% Co	3.1±0.3
Pd 0.2-at.% Co	5.8±0.5

[a]Sample analyses were carried out by Daniel Griffiths and Co., London.

2.95 K for the 0.2-at.% Co alloy, at 0.79 K for the 0.1-at.% Co alloy,[9] while that for the 0.05-at.% Co alloy is expected to be below 0.4 K. The rapidly increasing slope of the $\Delta\rho(T) - T$ curve with decreasing temperature suggests a logarithmic variation; consequently, $\Delta\rho(T)$ was plotted against $\ln T$ (Fig. 2). This figure clearly demonstrates that the variation of $\Delta\rho(T)$ at temperatures above the magnetic-ordering temperature is well represented by

$$\Delta\rho(T) = A + B\ln T \ . \tag{1}$$

Figure 2 also enables the coefficient B to be estimated; these are listed in Table I. Within experimental error, B varies linearly with the Co contration. At temperatures above 8 K the experimental points deviate from a logarithmic temperature dependence by an amount which is proportional to the "pure" Pd resistivity. This can probably be attributed to the breakdown of Matthiessen's rule,[11,12] which is expected whenever several competing scattering mechanisms of comparable magnitude and different anisotropy are present (in this case impurity, paramagnon,[14] and phonon scattering).

We will now discuss the possible origins of a temperature-dependent impurity resistivity above the magnetic-ordering temperature T_C. In *Pd*Fe alloys of comparable concentration and *over the relevant temperature range*, critical fluctuations in the local magnetization yield a contribution to the resistance whose slope varies as $(T - T_C)^{-n}$, with n estimated to be >1.[15-17] While such a dependence is consistent with our own measurements above T_C on *Pd*FE alloys, it does not account for the much larger temperature variation observed in *Pd*Co alloys; in addition, one would expect any short-range order above the Curie temperature to yield a temperature-dependent contribution to $\Delta\rho(T)$ which varied nonlinearly with concentration, and to be of comparable magnitude in both the *Pd*Fe and *Pd*Co systems. This result, combined with the linear concentration dependence of the coefficient B in Eq. (1), suggests that the $\ln T$ dependence results from a temperature-dependent scattering of s-like conduction electrons from *noninteracting* Co impurities. (In such alloys the conductivity is dominated by the contribution from s-like conduction electrons in view of their relatively low effective mass.[18,19]) A very similar temperature dependence (though considerably larger in magnitude) has been observed in *Rh*Fe,[20,21] IrFe,[22] and *Pt*Co,[23] and in ternary *RuRh*Fe and *RhPd*Fe[24] alloys, and it is reasonable to suppose that the effects in these systems and in *Pd*Co have a similar origin.

Let us first examine the possibility that this "single-particle" $\ln T$ variation results from Kondo[25] scattering of s electrons from the local Co moments. The positive sign for B requires that the exchange coupling constant J (between the s electrons and the local moment) be positive. The Kondo effect, as such, has been taken to apply to localized-impurity moments in otherwise nonmagnetic hosts.[26,27] Its extension to the case of an exchange-enhanced host metal is not without objection, and obviously awaits theoretical justification (or refutation); we nevertheless include the discussion in this section for completeness. Abrikosov[28] has derived the following expression for the excess resistivity (in zero applied magnetic field), which should be valid at $T=0$ for positive J, using the s-d Hamiltonian $\mathcal{H} = V - J\vec{S}\cdot\vec{\sigma}$ [where V represents the screened-Coulomb spin-independent potential which arises from the departure of the lattice from perfect periodicity, $-J\vec{S}\cdot\vec{\sigma}$ represents the isotropic exchange terms between the impurity spin ($\vec{S}$) and conduction-electron spins ($\vec{\sigma}$)]:

$$\Delta\rho = \Delta\rho_{\text{potential}} + \Delta\rho_{\text{spin}} = AV^2 + AJ_{\text{eff}}^2(T)S(S+1) \ , \tag{2}$$

where

$$A = \frac{3\pi m^*}{8E_F e^2\hbar N} \ \mu\Omega\,\text{cm/eV}^2\text{/at.\%} \ . \tag{3}$$

with m^* being the conduction-electron effective mass, E_F the Fermi energy (in eV), and N the number of lattice sites per unit volume. In addition,

$$J_{\text{eff}}(T) = \frac{J}{1 + Jn_s(E_F)n(D/T)} \ , \tag{4}$$

where $n_s(E_F)$ is the s-electron density of states at the Fermi energy E_F, and D is an energy of the order of the bandwidth. The coefficient B is thus given by

$$B = \frac{d(\Delta\rho)}{d(\ln T)} = 2J_{\text{eff}}(T)n_s(E_F)\Delta\rho_{\text{spin}} \ . \tag{5}$$

In an applied magnetic field H, the resistance tends to Eq. (2) for $T \gg H$, and falls to a value $\Delta\rho_H(0)$ at $T=0$, where

$$\Delta\rho_H(0) = AV^2 - 3AJ_{\text{eff}}^2(H)S^2 \tag{6}$$

and

$$J_{\text{eff}}(H) = \frac{J}{1 + Jn_s(E_f)\ln(D/H)} \ . \tag{7}$$

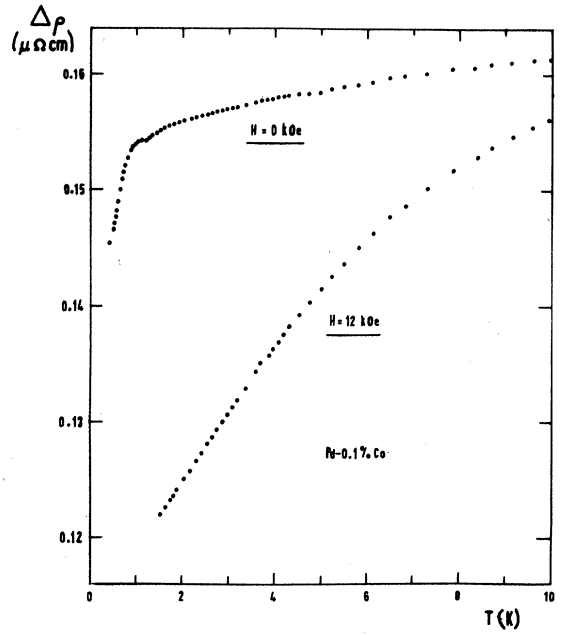

FIG. 3. Excess resistivity $\Delta\rho$ as a function of temperature, in zero applied field and in an applied magnetic field of 12 kOe for the Pd 0.1%-Co alloy.

In Fig. 3 the resistance of the Pd 0.1-at.% Co alloy is plotted against temperature in zero field and in a field of 12 kOe. The decrease in resistance $\Delta\rho'$ from $T \gg H$ to $T \ll H$ is given approximately by[28]

$$\Delta\rho' \sim A J_{\text{eff}}(T')(4S^2+S) , \tag{8}$$

where T' is approximately the temperature at which $\Delta\rho$ has decreased by $\frac{1}{2}\Delta\rho'$. Finally, we have

$$\frac{B}{\Delta\rho'} \sim \frac{2S+2}{4S+1} J_{\text{eff}}(T')n_s(E_f) . \tag{9}$$

From the measured values of $\Delta\rho'$ and B, and assuming $S=1$ (as obtained from neutron-diffraction data[29]), $J_{\text{eff}}(5\text{ K})n_s(E_F) = 0.092$. Assuming that the current is confined to a parabolic s band, with 0.36 s electrons per atom, and with effective mass $m^* = 2.3\,m$,[18,19] where m is the free-electron mass, $E_F = 1.33$ eV, and $A = 1.64\ \mu\Omega$cm/eV/at.%. With this value of A and using Eqs. (5), (8), and (9), one obtains $J_{\text{eff}}(5\text{ K}) = 0.23$ eV, $n_s(E_F) = 0.40$ per eV atom, and $J = 0.86$ eV. This value for $n_s(E_F)$ may be compared with the value of 0.20 per eV atom estimated by assuming a parabolic s band with the values of m^* and the number of electrons per atom above.[30]

It is evident that if this interpretation of the result is correct, then the Kondo temperature [defined for $J>0$ as the temperature at which $Jn_s(E_F)\ln(D/T) = 1$] is of the order of 1000 K for the *Pd*Co system, and that the spin resistivity at very high temperatures is more than an order of magnitude larger than its value in the liquid-helium temperature range. The value of J derived from the above analysis is considerably larger than that expected from normal ferromagnetic atomic exchange,[31] while the effective exchange coupling derived from covalent admixture,[32] although large under certain circumstances, is always antiferromagnetic.[33] Thus, although this explanation cannot be entirely discounted, it does seem unlikely that the observed temperature dependence can be attributed to Kondo scattering of s electrons from local Co moments with a *ferromagnetic* exchange coupling.

Fischer[34] has suggested that a positive logarithmic temperature dependence in the resistivity could result from a *negative* exchange constant J if the potential-scattering phase shift exceeds $\frac{1}{4}\pi$. For the system under consideration here, the effective exchange interaction is almost certainly an "extended" interaction of $l=2$ (d-like) symmetry,[35] which should, in addition, due to covalent admixture, be negative. However, since Pd and Co lie in consecutive columns of the Periodic Table, charge-screening considerations limit the $l=2$ potential phase shift to a value significantly smaller than $\frac{1}{4}\pi$. Such an explanation does not, therefore, seem plausible even for the contribution to the resistivity from the $l=2$ component of the conduction band (and is thus even more implausible for, say, the $l=0$ component, which presumably makes a significantly larger contribution to the conductivity). Similar considerations also make this explanation unacceptable in *Pt*Co, *Rh*Fe, etc.

In this latter respect a suggestion put forward by Knapp[36] in this discussion of the *Rh*Fe and *Ir*Fe systems seems more promising. This author suggests that there is an antiferromagnetic exchange coupling between the itinerant d electrons and the local spins, leading via the Kondo effect to a progressive compensation of the local moments as the temperature falls (the coexistence of spin compensation and giant moment being understandable in terms of the discussion in Ref. 33). The s electrons which dominate the conductivity are assumed to be weakly coupled to these local moments plus a compensating cloud. From the first Born approximation the zero-field spin resistivity is predicted to be proportional to $\langle M^2\rangle$, the mean-square magnetization, and hence to χT (where χ is the impurity susceptibility), a relation which has been experimentally verified for the *Rh*Fe,[36] *Ir*Fe,[36] and *Pt*Co[23] alloy systems. If this is a correct description of the temperature dependence of the *Pd*Co resistivity, then it may be estimated from the present results, assuming $S=1$, that $\Delta\rho_{\text{spin}}$ and hence $\langle M^2\rangle$ has decreased by approximately 30% as the temperature is reduced from 7 to 1.4 K. The "Kondo" temperature of the *Pd*Co system on this model must therefore be below 1 K.[37]

A further possibility that should be considered is

that the temperature-dependent resistivity may result from s-electron scattering from enhanced spin fluctuations *within the impurity cell*. In an extension of the theory of Lederer and Mills,[38] Kaiser and Doniach[39] have shown that this type of model can be used to explain the temperature dependence of the resistivity and susceptibility of *Rh*Fe, *Ir*Fe, and related systems. A central feature of the theory is that the characteristic spin fluctuation temperature T_s is inversely proportional to the exchange-enhancement factor on the impurity site $1/(1-U\chi_0)$, and goes to zero at the (static) Hartree-Fock magnetic instability $U\chi_0=1$ (where U is the intra-atomic exchange interaction relative to the host, and χ_0 is the host susceptibility). Significant temperature dependences in the liquid-helium temperature range are predicted only when the system is very close to this limit.

In contrast to the systems *Rh*Fe, *Pt*Co, *Ir*Fe, etc., *Pd*Co exhibits normal paramagnetic behavior, a Curie-Weiss susceptibility with a small Curie temperature, and a large *negative* magnetoresistance, and is, therefore, presumably well beyond the Hartree-Fock instability. From the similarity in the resistive behavior in *Pd*Co and these other systems, we conclude that spin fluctuation may not be confined to the region $U\chi_0 \sim 1$, but may extend well into the "magnetic" region. It is, in fact, reasonable to suppose that in a volume of atomic dimensions, fluctuations in the magnetization will be comparable to the mean magnetization even in a magnetic system.[40] The zero-frequency enhancement factor $1/(1-U\chi_0)$ diverges at the Hartree-Fock magnetic instability, but this factor does not fully characterize the magnetic response of the system since the dynamic (fluctuating) aspects are not included in it; Hamann[41] has recently demonstrated that, in fact, T_s goes to zero only in the limit $U\to\infty$.

SUMMARY

In conclusion, we have demonstrated that the temperature-dependent resistivity in *Pd*Co is unlikely to result from the Kondo scattering of s electrons from local Co moments via a ferromagnetic s-d exchange coupling J, as this leads to the prediction of an unreasonably large value for J. An explanation in terms of a negative s-d exchange coupling and a potential phase shift greater than $\frac{1}{4}\pi$ appears equally untenable since Pd and Co lie in consecutive columns of the Periodic Table. Operationally the approaches of Knapp and of Kaiser and Doniach yield very similar results, and so it is not possible, from resistivity measurements, to distinguish between the rival claims of a Kondo compensation by the itinerant d electrons, or of localized spin-fluctuation effects. However, from the observation of a well-defined paramagnetic behavior in the liquid-helium range, we may conclude that the relevant characteristic temperature T_K or T_s is very much less than 1 K.

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Longitudinal Dynamical Susceptibility of the Heisenberg Ferromagnet at Short Wavelengths and Low Temperatures*

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The longitudinal dynamical susceptibility of the Heisenberg ferromagnet is studied at short wavelengths and low temperatures. It is shown that identical results to order $1/S$ are obtained using (a) a spin decoupling technique, (b) a diagrammatic method using the Holstein-Primakoff transformation, and (c) a diagrammatic method using the Dyson-Maleev transformation. We thus conclude that there are no significant kinematic effects at low temperatures. Using the random-phase approximation, we find that the Dyson-Maleev interactions between magnons are too weak to support the existence of a zero-sound mode. Both these conclusions disagree with the recent results of other authors.

I. INTRODUCTION

The increasing resolution attainable in inelastic neutron-scattering experiments has stimulated interest in the various collective excitations in magnetic systems, since these excitations are potentially observable via such experiments. Thus, detailed investigations of two-spin-wave bound states, [1-4] of second magnons, [5-7] and more recently, of zero sound[8,9] have been carried out. With regard to zero sound, the work of Ranninger and Natoli (RN)[9] is especially provocative. By analyzing the longitudinal dynamical susceptibility RN have concluded that (a) there is a well-defined collective excitation for wave vectors near the zone boundary, and (b) kinematic interactions play an important role in the kinematics of this mode. The purpose of this paper is to investigate these points in greater detail, since such conclusions have rather fundamental implications for both theoretical and experimental programs in magnetism.

The motivation for reexamining these conclusions is that the theory of RN appears to embody two physically unsatisfactory aspects. First, they claim to have detected effects of the kinematic interaction on the zero-sound mode, but the effects they find are simply proportional to various powers of Bose occupation numbers. In other words, the kinematic interaction in their theory gives rise to effects of order $(kT/JS)^n$, where n is of order 3. On the other hand, such large effects at low temperature are not to be expected in view of Dyson's arguments, [10] which suggest that these effects are of order $\xi \equiv e^{-aT_C/T}$, where T_C is the Curie temperature and a is a constant of order unity. Indeed, up to now, no one has been able to construct a theory which is accurate enough to detect effects of order ξ at low temperatures. (In this connection, it is worth noting that treatments of the two-spin-wave bound states via a hard-core potential, [11,12] which rigorously exclude kinematic effects, have thus far only taken account of two-spin-wave states, and hence do not yield any conclusions about the kinematic effects of states involving more than two spin