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Magnetoresistance of a Dilute Ferromagnetic *Pd*Mn Alloy: Acoustic- and Optic-Mode Excitations*

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Magnetoresistance measurements on a Pd–2.91-at.% Mn alloy in the temperature range 1.4–20 K and in applied fields up to 60 kOe are presented. The data demonstrate the effects of *inter*impurity (near-neighbor) covalent admixture, the antiferromagnetic coupling from which appears to be in qualitative agreement with the calculations of Moriya. These data also allow an estimate of the near-neighbor Mn-Mn interaction energy to be made, and this is compared with quantitative estimates on the basis of the double-resonance coupling mechanism of Caroli and Blandin. By including a magnetic field in the collective-excitation model of Long and Turner, the magnetoresistance data can be fitted once allowance has been made for near-neighbor antiferromagnetic coupling. The acoustic-spin-wave stiffness D, which is obtained by fitting the data to this model, is found to be field dependent; it decreases rapidly with increasing field. The possibility of optical-mode collective excitations is briefly discussed.

I. INTRODUCTION

The static response of the conduction electrons of some hosts to moment bearing impurities, particularly impurities from the first transition series, has been the subject of considerable theoretical speculation. That group of hosts, especially, in which exchange-enhancement effects play an important role, has had much attention focussed on it. A phenomenological account of the static properties of alloys in this latter group, based essentially on a rigid-band model, has been given by Rhodes and Wohlfarth[1]; in contrast, Clogston and co-workers[2] used an approach in which the near-neighbor host "lattice sites" were perturbed by a magnetized virtual-bound impurity state. This latter approach, however, suffered from the obvious drawback of predicting an induced moment in the surrounding matrix which was not only parallel to the "on-site" impurity moment but also related to the magnitude of this moment by a constant factor.

Moriya[3,4] has subjected this problem to a detailed analysis based on both the Anderson[5] and the Wolff–Clogston[6,7] (tight-binding) models. Within the framework of the Anderson model, the physical origin of the various contributions to the induced conduction-band polarization are easily identified. The admixture term in this model [$\sum_k \sum_\sigma V_{kd}(c^*_{k\sigma} c_{d\sigma} + c^*_{d\sigma} c_{k\sigma})$ in Anderson's[5] notation] causes part of the spin-up (occupied) localized d state to be mixed into the empty conduction-band states of the same spin orientation, while occupied spin-down conduction-band states are mixed into the unoccupied spin-down localized d state. This is the covalent-admixture contribution, which is clearly parallel to the impurity moment (and in hosts where exchange-enhancement effects are unimportant, the integrated conduction-band polarization from covalent admixture must exactly compensate the reduction in local moment). The admixture term also causes an energy shift (in second order) of the conduction-band states; this energy shift is positive for the spin-up and negative for the spin-down band states. The consequent redistribution of conduction electrons leads to a contribution to the induced conduction-band polarization which is antiparallel to the local moment.

The sign and magnitude of the net conduction-band polarization is thus dependent on the relative magnitudes of the covalent-admixture and energy-shift contributions, which, in turn, are closely related to the slope of the density of states at the Fermi level, and to the arrangement of the spin-up and spin-down localized d states relative to the Fermi energy.[3,4] It is this net contribution which is subject to the effects of intraband exchange. Indeed, detailed application of Moriya's calculations to first-row transition elements in Pd not only predicts the correct sign of the induced polarization in the host's d band (i.e., the "giant-moment" phenomenon associated with Fe and Co impurities in Pd[2,8–10] along with the negative polarization induced by Cr impurities in the same host), but also gives realistic quantitative estimates for the magnitude of this polarization when allowance is made for the effects of exchange enhancement. At this point the polarization induced by Mn impurities in Pd warrants some comment. Calculations of the above type indicate that the induced polarization is small and negative; the magnitude of the induced polarization has been confirmed by impurity NMR measurements[11] (these indicate that the on-site moment is close to the total moment). However, it seems possible that when the effects of true s-d exchange are taken into account,[4,5] a small *positive* polarization will result, which will then be in quantitative agreement with paramagnetic resonance ($g = 2.09$)[12] measurements, and with the analysis of resistivity measurements on *Pd*Mn alloys[13,14] (the latter indicate that the ferromagnetic ordering temperature T_c per atomic percent Mn is much less than that for Fe or Co impurities in the same host).

In the low-concentration limit (< 1-at.% impurity) the nature of the ordered ground state of the alloy is determined essentially by the spatial variation of the conduction-band polarization. Typically, the Ruderman-Kittel-Kasuya-Yosida (RKKY) oscillations[15] induced in the host conduction band lead to a "disordered" antiferromagnetic ground state.[16] In hosts where intraband exchange effects are important, the RKKY oscillations are suppressed out to relatively large distances,[17,18] leading to a ground state of largely ferromagnetic character. However, in alloys containing several atomic percent impurity, this latter behavior can be modified by the effect of *direct* interimpurity coupling (operative virtually only when impurities are nearest neighbors). Near-neighbor-interaction effects have been studied by several authors; Alexander and Anderson[19] and Moriya[4] have investigated interaction effects due to admixture between two near-neighbor magnetic impurities. The relevant conclusions reached on the basis of this approach are that impurities with nearly half-filled d shells tend to couple antiferromagnetically; ferromagnetic coupling is increasingly favored as the number of d electrons increase. These conclusions appear to be in good qualitative agreement with the observed concentration dependence of the magnetic ordering temperature T_c for Fe, Co, and Mn impurities in Pd.[14] For concentrations of the order of a few atomic percent, dT_c/dc is an increasing function of concentration for Fe and Co impurities, but is a decreasing function of concentration for Mn in Pd.

Theoretical investigations[20,21] of the dynamic properties of the ordered phase of alloys formed from impurities in the latter half of the first transition series with Pd, on the basis of an s-d model in which the impurity spin ($\vec{S}$) is coupled to itinerant d electrons (of spin $\vec{\sigma}_d$) via an isotropic exchange coupling $-2J_{d\,\text{local}}\vec{S}\cdot\vec{\sigma}_d$, have indicated that at temperatures well below T_c, the elementary excitations of the coupled local-moment d-hole system can be approximately described by spin waves. The conductivity of this ordered phase has been calculated[22] on the basis of s electrons scattering from these elementary excitations via a coupling of the form $-2J\vec{S}\cdot\vec{\sigma}_s$, where the effective exchange constant J has contributions from both "direct" and "indirect" processes.[22] The random distribution of impurities implies a lack of translational symmetry in the s-d Hamiltonian for the alloy, which thus predicts a $T^{3/2}$ limiting low-temperature form for the incremental resistivity $\Delta\rho(T)$; the latter has been observed in dilute *Pd*Fe,[23] *Pd*Co,[24] and *Pd*Mn[14,25] alloys. This model has also been extended to take into account the effects of an applied magnetic field,[26] and appears to give a reasonably good account of the observed magnetoresistance in dilute ferromagnetic *Pd*Fe and *Pd*Co alloys.[27]

In this paper magnetoresistance measurements are presented on a Pd–2.91-at.% Mn alloy over the temperature range 1.4–20 K, and in applied magnetic fields up to 60 kOe. These data can be analyzed within the framework of the above model when allowance is made for the effects of near-neighbor interimpurity interactions.

II. EXPERIMENTAL DETAILS

The specimen used in this investigation was in the form of a carefully etched and annealed strip of approximate dimensions 0.01 cm thick, 0.3 cm wide, and 7 cm long. It was prepared in a previously described manner[24] from 99.999%-pure Pd (from Johnson Matthey and Co., London) and 99.95%-pure Mn (supplied by Koch-Light Ltd., England) and had been subjected to measurements down to 0.41 K in zero applied field.[14] During the course of the present measurements the sample was mounted in the longitudinal field of a superconducting magnet operating in a persistent mode. This magnetic field was measured by a Hall probe to within ± 0.1 kOe, and varied by about 2 parts in 10^3 over the length of the sample. While the magnet was immersed in a He^4 bath at atmospheric pressure, the sample was mounted in a copper enclosure which was inserted into the bore of the solenoid. Temperature stabilization of this enclosure was achieved by using a carbon resistor in a feedback control circuit; this allowed temperatures below 4.2 K to be stabilized and measured (via He^4 vapor pressure) to ± 1 mdeg and above 4.2 K to better than 1% (using a gas thermometer).

A four-probe technique was used to measure the sample resistance; in this the sample current was varied to balance a highly stable voltage. Balanced conditions reproducible to $\pm 10^{-9}$ V were achieved, leading to a resistance measurement accurate to

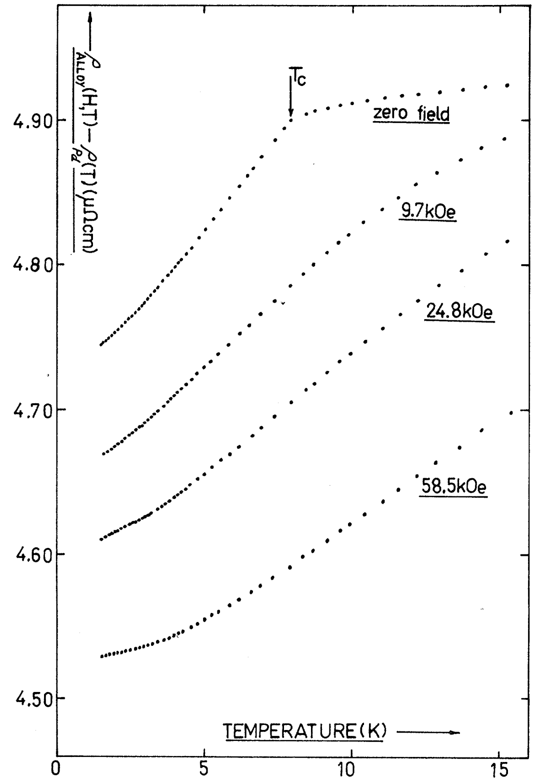

FIG. 1. Incremental resistivity $\rho_{\text{alloy}}(H, T) - \rho_{\text{Pd}}(T)$ ($\mu\Omega$ cm) plotted against temperature (K) in zero field and in applied fields of 9.7, 24.8, and 58.5 kOe.

1 in 10^4. Reliable estimates of the sample resistivity were made by measuring the area-to-length ratio of the specimen to an accuracy of ±0.5%.[28]

III. RESULTS AND DISCUSSION

In Fig. 1 the measured incremental resistivity $\Delta\rho(H, T) = \rho_{\text{alloy}}(H, T) - \rho_{\text{Pd}}(T)$ is plotted as a function of temperature up to 16 K in zero field and in applied fields of 9.7, 24.8, and 58.5 kOe.[29] The measurements in zero field and in 9.7 kOe were extended up to 25 K, and indicate that $\Delta\rho(H=0, T=25\text{ K}) - \Delta\rho(H=9.7\text{ kOe}, T=25\text{ K})$ is vanishingly small. This latter result demonstrates that the effects of "normal" magnetoresistance due to the cyclotron curvature of conduction-electron orbits is not an important consideration in the present investigation. This appears to be in quantitative agreement with the work of Campbell *et al.*[30] in which it is suggested that such effects are unimportant in alloys whose resistivity is greater than 1 $\mu\Omega$ cm. From Fig. 1 the magnetic ordering temperature T_c is estimated at 7.85 ± 0.1 K, while the coefficient of the $T^{3/2}$ limiting form for $\Delta\rho(H=0, T)$ valid up to about 3.5 K (see Fig. 2) is $(8.95 \pm 0.1) \times 10^{-3}$ $\mu\Omega\,\text{cm/K}^{3/2}$. These estimates compare favorably with previous zero-field measurements on this Pd–2.91-at.%-Mn sample[14]; previous data on this and less concentrated *Pd*Mn allows are summarized in Table I.

In zero applied field it is usual[31] to take the difference $\Delta\rho(T_c) - \Delta\rho(T=0)$ as a measure of the "spin-disorder" contribution to the resistivity, $\Delta\rho(T_c)$ representing the contribution from the totally disordered state and $\Delta\rho(T=0)$ that from the fully ordered state. In the random-phase approximation (RPA), $\Delta\rho(T_c) - \Delta\rho(T=0)$ is linearly proportional to the impurity concentration c.[31] An inspection of Table I reveals that this approximation is not a good description of the zero-field data on *Pd*Mn [for example, $\Delta\rho(T_c) - \Delta\rho(T=0)$ is larger for 2.4 than 2.91-at.% Mn]. This is confirmed by the data plotted in Fig. 1; in examining these data it is clear that $\Delta\rho(H, T_c)$ can no longer be taken as the contribution from the totally disordered state. However, the opening observations of this section indicate that neglecting the contribution to the incremental resistivity from the breakdown of Matthiessen's rule[32] (in this case due to the competing effects of spin disorder and phonon scattering), then $\Delta\rho(H=0, T_c) = \Delta\rho(H, T \gg T_c)$—the contribution from the totally disordered state does not depend on the applied field—in which case $\Delta\rho(H=0, T_c) - \Delta\rho(H, T=0)$ represents the appropriate measure of the spin-disorder contribution in an applied field H. The observation of a field dependence in this latter difference is thus clearly inconsistent with our assigning $\Delta\rho(H=0, T=0)$ as the contribution from the fully ordered state. This

TABLE I. Summary of existing zero-field data on dilute *Pd*Mn alloys.

Mn concentration (at. %)	T_c(K)	$\Delta\rho(T=0)$[a] ($\mu\Omega$ cm)	Coefficient of the[a] $T^{3/2}$ term ($\mu\Omega$ cm/K$^{3/2}$)	$\Delta\rho(T_c) - \Delta\rho(T=0)$[a] ($\mu\Omega$ cm)
1.05(14)	3.90 ± 0.02	1.363(5) ± 0.001	$(15.6 \pm 0.1) \times 10^{-3}$	0.101(5) ± 0.001
2.40(14)	7.35 ± 0.02	3.413(0) ± 0.001	$(10.7 \pm 0.1) \times 10^{-3}$	0.187(0) ± 0.001
2.91(14)	7.71 ± 0.02	4.788(1) ± 0.001	$(9.0 \pm 0.1) \times 10^{-3}$	0.177(9) ± 0.001
1.0(25)	3.4	1.647	15.1×10^{-3}	0.092
0.5(13)	1.7			
1.0(13)	3.7			
2.0(13)	5.25			

[a]The errors quoted in these columns exclude errors in the measured area-to-length ratio of the samples.

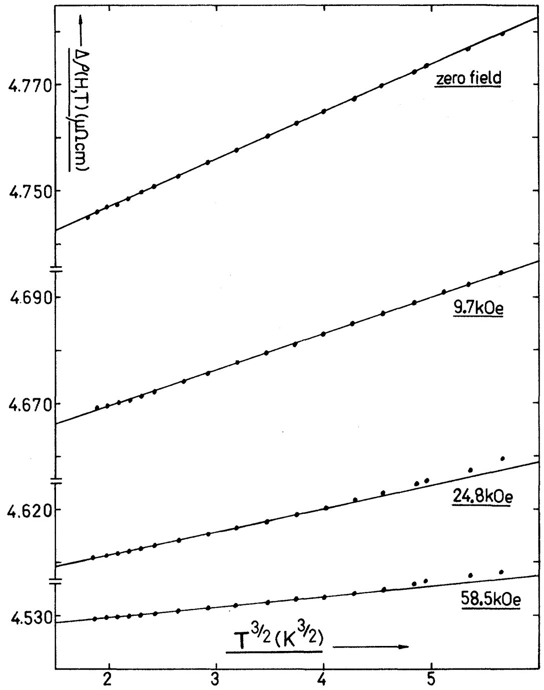

FIG. 2. Incremental resistivity $\Delta\rho(H,T)$ ($\mu\Omega$ cm) plotted against $(T^{3/2})(K^{3/2})$ in zero field, and in applied fields of 9.7, 24.8, and 58.5 kOe.

inconsistency is removed by conceding that in zero field the ground state of more concentrated alloys contains a significant number of antiferromagnetically coupled, near-neighbor Mn pairs, in qualitative agreement with calculations based on interimpurity covalent admixture.[4]

A. Near-Neighbor Interactions

In order to treat the effects of near-neighbor interactions on a quantitative basis it is necessary to obtain reasonably accurate estimates for $\Delta\rho(H, T=0)$. According to Long and Turner,[22] at temperature $T \ll T_c$,

$$\Delta\rho(H=0,\,T)=Ac+Bc^{-1/2}T^{3/2}\,, \tag{1}$$

where A and B are constants. In Fig. 2, $\Delta\rho(H, T)$ for $T \ll T_c$ are plotted against $T^{3/2}$; as expected $\Delta\rho(H=0, T)$ is well described by Eq. (1) up to about 3.5 K; unexpectedly $\Delta\rho(H, T)$ also follow a $T^{3/2}$ limiting form up to about 2.8 K ($\approx \frac{1}{3}T_c$), with B decreasing with increasing H—this is not the situation in dilute ferromagnetic *Pd*Fe and *Pd*Co alloys.[27] This result enables $\Delta\rho(H, T=0)$ to be accurately evaluated (see Table II). The difference $\Delta\rho(H=0, T_c) - \Delta\rho(H, T=0)$ is plotted against the applied field H in Fig. 3, in this way the "infinite-field" step height is estimated at 0.44 ± 0.04 $\mu\Omega$ cm (the estimate is admittedly crude, this is reflected in the $\pm 10\%$ error). One can write

$$\Delta\rho(H=0,\,T_c)-\Delta\rho(H,\,T=0)=E-Fe^{-\alpha H}\,, \tag{2}$$

which is essentially a molecular-field description of the antiferromagnetic Mn-Mn nearest-neighbor interaction. E is identified with the infinite-field step height, F is a parameter with the dimensions of $\mu\Omega$ cm, while

$$\alpha \approx g\mu_B S/E_{\mathrm{int}}\ . \tag{3}$$

Here E_{int} is the assumed unique nearest-neighbor interaction energy due to interimpurity covalent admixture, g is the impurity Landé factor, and μ_B

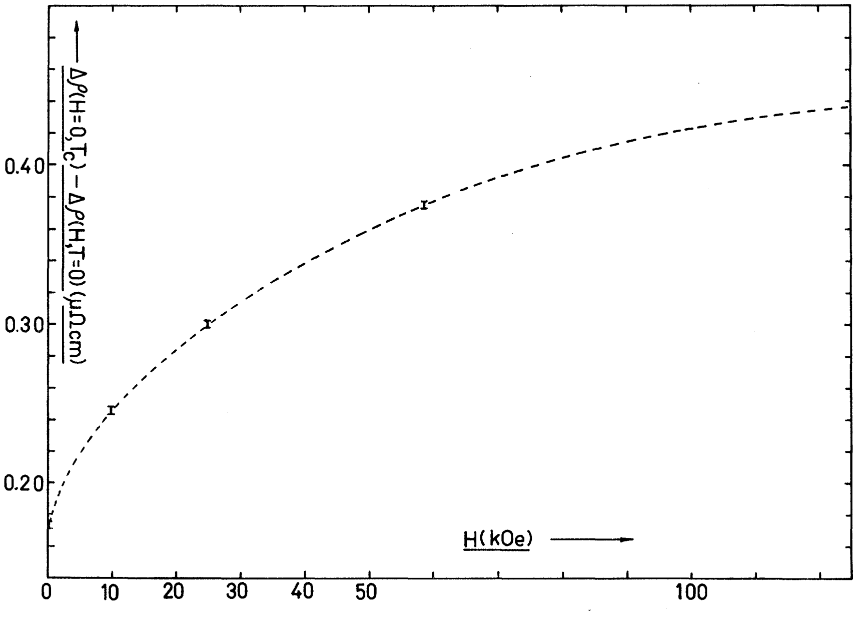

FIG. 3. "Step height" $\Delta\rho(H=0,\ T_c) - \Delta\rho(H, T=0)$ ($\mu\Omega$ cm) plotted against applied field (kOe).

TABLE II. Magnetoresistance data on Pd–2.91-at.% Mn.

Magnetic field (kOe)	Coefficient of the $T^{3/2}$ [a] term ($\mu\Omega$ cm/$K^{3/2}$)	$\Delta\rho(H, T=0)$ [a] ($\mu\Omega$ cm)	$\Delta\rho(H=0, T_c) - \Delta\rho(H, T=0)$ [a] ($\mu\Omega$ cm)
zero	$[8.9(5) \pm 0.1] \times 10^{-3}$	$4.728(5) \pm 0.001$	$0.171(5) \pm 0.001$
9.7 ± 0.1	$[7.0(0) \pm 0.1] \times 10^{-3}$	$4.655(0) \pm 0.001$	$0.245(0) \pm 0.001$
24.8 ± 0.1	$[4.5(5) \pm 0.1] \times 10^{-3}$	$4.602(0) \pm 0.001$	$0.298(0) \pm 0.001$
58.5 ± 0.1	$[2.0(6) \pm 0.1] \times 10^{-3}$	$4.525(4) \pm 0.001$	$0.374(6) \pm 0.001$

[a] Errors quoted in these columns exclude errors in the measured area-to-length ratio.

is the Bohr magneton. Figure 4, in which $\ln[E - \Delta\rho(H=0,\ T_c) + \Delta\rho(H,\ T=0)]$ is plotted against the applied field H, demonstrates that the assumption of a unique value for E_{int} is reasonable, while the slope of this plot gives

$$\alpha \approx (2.40 \pm 0.06) \times 10^{-5}.$$

Using $g = 2.09$[12] and $S = 2.5$ [33] yields

$$E_{\text{int}} = (1.21 \pm 0.05) \times 10^{-3}\ \text{eV}.$$

Although the effects of interimpurity covalent admixture, as discussed by Moriya,[4] appears to account qualitatively for the observed near-neighbor coupling, quantitative estimates on the basis of this model are difficult to obtain. However, the *double*-resonance coupling model of Caroli[34] and Blandin[35] may be applied directly to the present situation. Double-resonance coupling between two near-neighbor impurities separated by a distance R leads to an interaction energy

$$E_{\text{int}}(R) = \sum_l \frac{(2l+1)^2}{\pi} E_F \sin\eta_l^{\sigma}$$

$$\times \sin\eta_l^{-\sigma} \frac{\cos(2k_F R + \eta_l^{\sigma} + \eta_l^{-\sigma})}{(k_F R)^3}. \qquad (4)$$

l labels various partial waves and σ is a spin index, E_F is Fermi energy, and $\eta_l^{+\sigma}$, $\eta_l^{-\sigma}$ are the phase shifts for the appropriate (spin-up or -down) partial waves; k_F is the Fermi momentum. The phase shifts appearing in Eq. (4) may be estimated from

$$Z = \sum_l \frac{2l+1}{\pi} [\eta_l^{\sigma}(E_F) + \eta_l^{-\sigma}(E_F)]\ . \qquad (5)$$

Z is the charge difference between host and impurity—this is the Friedel sum rule[36]—while

$$S = \sum_l \frac{2l+1}{\pi} [\eta_l^{\sigma}(E_F) - \eta_l^{-\sigma}(E_F)]\ . \qquad (6)$$

Assuming d-wave ($l = 2$) contributions dominate Eqs. (4)–(6), and using $Z = -3$ and $S = \frac{5}{2}$,[37] Eqs. (5) and (6) yield $\eta_2^{+} = -\frac{1}{20}\pi$ and $\eta_2^{-} = -\frac{11}{20}\pi$. To estimate $E_{\text{int}}(R)$ from Eq. (4) it is necessary to evaluate k_F and E_F. Recent band-structure calculations for Pd [38] have indicated that the Fermi velocity v_F for $XW5$, the open d-hole surface in the fifth zone, averages approximately to 0.2 a.u. The bare

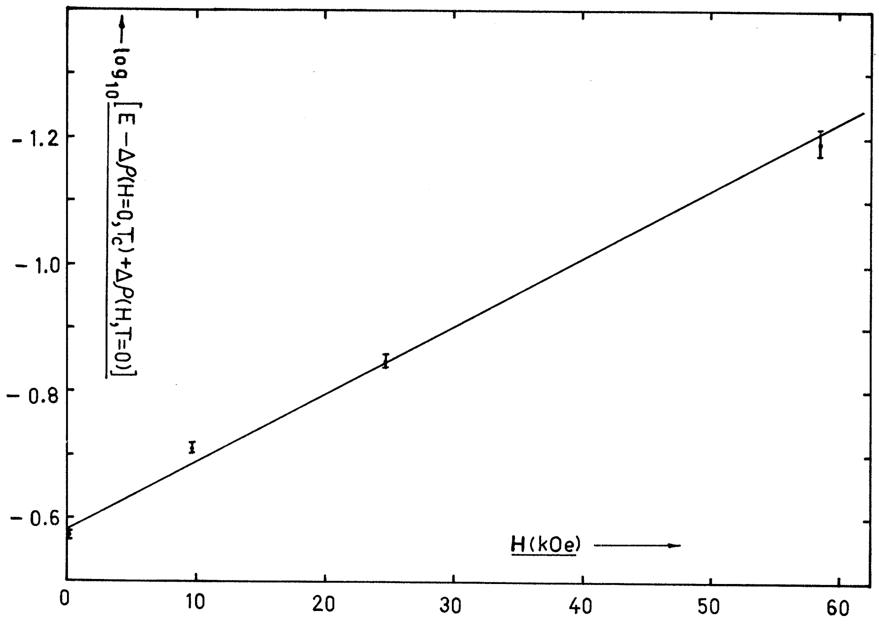

FIG. 4. ln(E − step height) plotted against applied field (kOe).

TABLE III. RPA estimates of the concentrations ordering ferromagnetically (c_1) and antiferromagnetically (c_2) in various fields. These estimates are based on an infinite field step height of 0.44 $\mu\Omega$ cm.

Magnetic field (kOe)	c_1 (at.%)	c_2 (at.%)
zero	0.955	1.955
9.7 ± 0.1	1.49	1.42
24.8 ± 0.1	1.87	1.04
58.5 ± 0.1	2.44	0.47

band mass for this surface[39] is 9.1 and the enhancement factor 1.66,[38] the effective mass m_d^* is thus estimated at $15.1m_0$, which gives $k_F = 5.7$ Å^{-1}, and the product $k_F R$ for nearest-neighbor impurities is 15.6. The estimated Fermi energy E_F is 7.6 eV[38] (relative to the potential zero between muffin-tin spheres); from Eq. (4) one obtains

$$E_{\text{int}}(\text{nearest neighbors}) = 1.3\times 10^{-3}\ \text{eV},$$

which appears to be in good agreement with that estimated from the experimental data.

B. Low-Temperature Magnetoresistance Data

The extension of Eq. (1) to include the effects of an applied field H yields for $\Delta\rho(H, T)$ for $T \ll T_c$ and in those alloys in which charge scattering is much stronger than "exchange" scattering[26]:

$$\Delta\rho(H, T) = Ac(V^2 - 3J^2S^2) + \frac{AcJ^2S\Gamma(\frac{3}{2})}{2\pi^2}\left(\frac{\Omega}{N}\right) \times\left(\frac{k_B T}{D}\right)^{3/2}\left(4\sum_{n=1}^{\infty}\frac{(e^{-t})^n}{n^{3/2}} - \sum_{n=1}^{\infty}\frac{(-e^{-t})^n}{n^{3/2}}\right). \tag{7}$$

Here

$$A = 3\pi m^*\Omega/2e^2hE_F N\ . \tag{8}$$

Ω is the atomic volume and N the number of atoms in that volume; m^* is the conduction-electron effective mass. For Pd, an effective-mass treatment of the s band with $m_s^* = 2.2m_0$ and 0.36 s electrons per atom[40,41] gives $A = 6.56$ $\mu\Omega$ cm/eV^2 at.%. In Eq. (7), V is the spin-independent screened Coulomb potential arising from the departure of the lattice field from perfect periodicity, and J is the effective exchange coupling discussed in Sec. I. $\Gamma(x)$ is the appropriate Γ function, k_B is Boltzmann's constant, and

$$t = g\mu_B H/k_B T\ .$$

D is the acoustic spin-wave stiffness constant. Owing to the complications of mixed ferromagnetic and antiferromagnetic coupling in *Pd*Mn, a direct analysis of the present magnetoresistance data using Eq. (7) is not possible. The following approximate scheme was thus adopted; if, in some applied field H, the concentration of impurities ordering ferromagnetically is c_1 and concentration c_2 order antiferromagnetically, then in the RPA[31] the following expression results:

$$\Delta\rho(H=0, T_c) - \Delta\rho(H, T=0) = AJ^2S[c_1(1+4S)+c_2]\ . \tag{9}$$

For the 2.91-at.% Mn alloy examined here, obviously $c_1 + c_2 = 2.91$. $|J|$ may be calculated from the infinite field step height (since $c_2 = 0$ for $H = \infty$); using the previously listed parameters,

$$|J| = (2.9 \pm 0.2)\times 10^{-2}\ \text{eV}.$$

Using this estimate, V can also be evaluated, since in infinite field

$$\Delta\rho(H=\infty, T=0) = A(c_1+c_2)(V^2 - 3J^2S^2), \tag{10}$$

from which

$$V = 0.49(5)\ \text{eV}.$$

Using the above value of $|J|$ in conjunction with Eq. (9) and the measured values of $\Delta\rho(H=0, T_c)$ and $\Delta\rho(H, T=0)$ leads to the estimates of c_1 and c_2 listed in Table III.

In fitting the present magnetoresistance data to Eq. (7), we make use of the concentration c_1 of impurities ordering ferromagnetically and assume that those impurities antiferromagnetically ordered at $T = 0$ are not significantly thermally randomized at temperatures of interest here (up to about 4 K)[42] and thus make no significant temperature-dependent contribution to $\Delta\rho(H, T)$ for $T < T_c$. Figures 5, 6, and 7 show the fit to the experimental data in applied fields of 9.7, 24.8, and 58.5 kOe, respectively, obtained using the above scheme, and with $g = 2.105$ as measured in the ferromagnetic phase using resonance methods.[12,43] The zero-field $T^{3/2}$ expansion [Eq. (1)] is found to be valid up to about 3.5 K, a similar range of validity is expected for the field-modified form [Eq. (7)]. An inspection of Figs. 5–7 confirms this. The values of D required to fit these data are listed in Table IV, along with that value of D derived from the observed zero-field $T^{3/2}$ coefficient and the appropriate effective concentration. The origin of the relatively small magnitudes for D estimated from resistivity measurements compared with those derived from analyses of other data[44] is discussed

TABLE IV. Estimates of the acoustic spin-wave stiffness from the magnetoresistance data.

Magnetic field (kOe)	D(K Å^2)
zero	4.89 ± 0.03
9.7 ± 0.1	4.82 ± 0.04
24.8 ± 0.1	4.33 ± 0.04
58.5 ± 0.1	2.99 ± 0.20

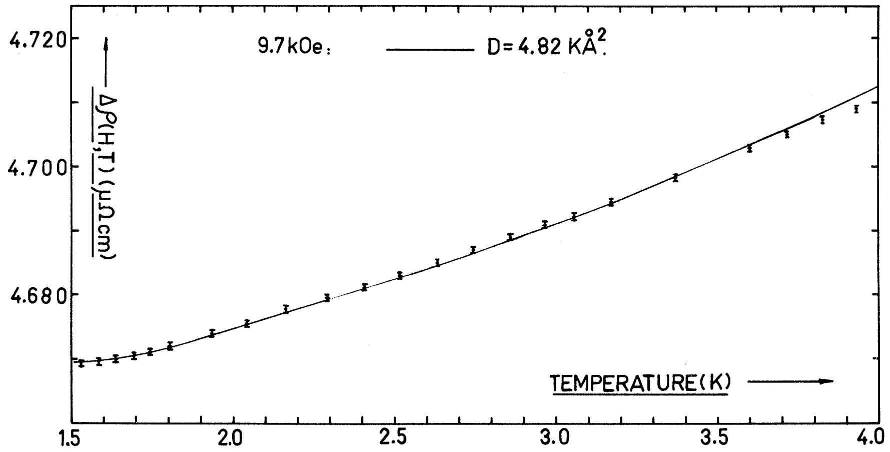

FIG. 5. Low-temperature (< 4 K) incremental resistivity data ($\mu\Omega$ cm) in a field of 9.7 kOe, plotted against temperature (K). The solid line is a calculated fit using $D = 4.82$ K Å^2.

elsewhere.[45]

From Table I it can be seen that for those alloys examined in zero field, the $T^{3/2}$ coefficients are within experimental error, proportional to $c^{-1/2}$, as predicted in Eq. (1).[22] However, it was pointed out in Ref. 14 that this agreement must be regarded as fortuitous since Eq. (1) is based on a straightforward molecular-field model in which both T_c and D are proportional to c; experimentally, T_c is clearly not proportional to c for concentrations of interest here. The present measurements emphasize this point since they demonstrate that not all impurities are free to participate in acoustic-mode collective excitations. From previous work on dilute ferromagnetic *Pd*Fe and *Pd*Co alloys,[27] the acoustic-spin-wave stiffness D was found to increase with both increasing field and concentration; however an examination of Table IV indicates that for *Pd*Mn, D is a decreasing function of H (and hence of c_1). While it could be argued with some justification that the absolute values of D listed in this table may be in error due to our use of an admittedly simplified RPA scheme for separating the ferromagnetic and antiferromagnetic contributions,[46] a general trend has nevertheless been established—this can be demonstrated as follows: The expression for $\Delta\rho(H, T)$ can simply be rewritten

$$\Delta\rho(H, T) = Ac(V^2 - 3J^2S^2) + \gamma T^{3/2} \times\left(4\sum_{n=1}^{\infty}\frac{(e^{-t})^n}{n^{3/2}} - \sum_{n=1}^{\infty}\frac{(-e^{-t})^n}{n^{3/2}}\right), \quad (11)$$

where γ can be obtained from comparison with Eq. (7). Changes in γ reflect changes in D, but obviously the values obtained for γ do not depend on a particular "separation" scheme. In Table V the various values for γ used to fit the present data are listed as a function of field; the corresponding γ's for Pd–0.78-at.% Fe[27] are listed for comparison; the opposing trends are apparent. Available calculations of the acoustic spin-wave stiffness D^{20-22} in dilute binary alloys predict that this quantity should be proportional to the impurity concentra-

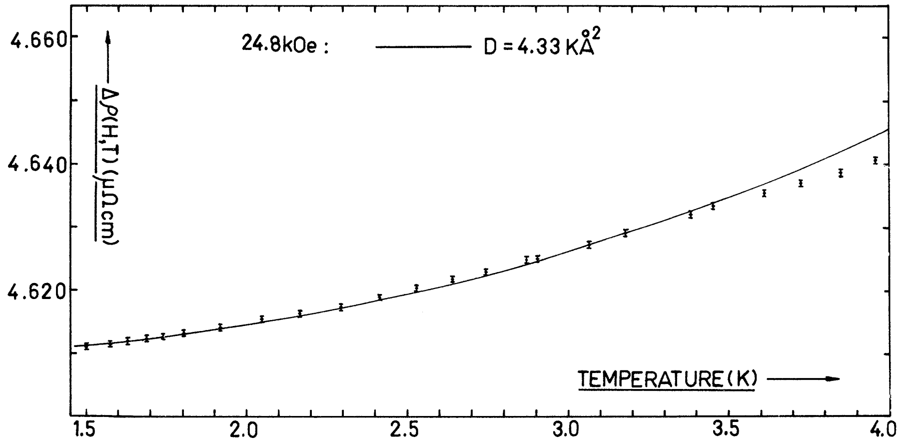

FIG. 6. Low-temperature incremental resistivity data ($\mu\Omega$ cm) in a field of 24.8 kOe, plotted against temperature (K). The solid line is a calculated fit using $D = 4.33$ K Å^2.

TABLE V. Estimates of the parameter γ.

Magnetic field (kOe)	$\gamma(Pd\mathrm{Mn})$ $(10^{-4}\ \mu\Omega\ \mathrm{cm/K^{3/2}})$	$\gamma(Pd\mathrm{Fe})$ $(10^{-4}\ \Omega\mu\Omega\ \mathrm{cm/K^{3/2}})$
zero	$9.0(5) \pm 0.1$	1.38 ± 0.01
9.7 ± 0.1	14.6 ± 0.2	1.37 ± 0.01
24.8 ± 0.1	21.4 ± 0.2	1.31 ± 0.01
58.5 ± 0.1	48.5 ± 5.5	1.14 ± 0.01

tion c. The present measurements suggest that in the *Pd*Mn system the concentration of impurities ordering ferromagnetically increases with increasing field, yet paradoxically D appears to decrease rapidly with increasing field.[47]

In the ordered phase, analyses of the spectrum of the collective excitations previously discussed[20,21] reveal two branches (due to the inequivalent contributions from conduction electrons and local moments to the total magnetization). According to Doniach and Wohlfarth,[20] the energetically low-lying acoustic mode is separated from the optical mode by an energy gap $E_G(\vec{q})$ which, at $\vec{q}=0$, has the value

$$E_G(\vec{q}=0)=\frac{J_{d\,\mathrm{local}}\,c\mu}{50\mu_B}\ , \qquad (12)$$

where μ is the moment per impurity site. Using $\mu \approx 2.5\ \mu_B$, $E_G(\vec{q}=0)$ then depends on the value estimated for $J_{d\,\mathrm{local}}$. The latter can be evaluated in two ways: (i) from the measured g shift and (ii) from the observed magnetic-ordering temperature.

(i) *g shift.* From the work of Peter *et al.*[12] it follows that the g shift (Δg) due to an isotropic exchange coupling of the form discussed in Sec. I is[48]

$$\Delta g = 2J_{d\,\mathrm{local}}\,\chi_z/g_e\mu_B\ , \qquad (13)$$

where χ_z is the host susceptibility (per atom) and g_e is the conduction-electron g factor. For $\Delta g = 0.1$[12] and $\chi_z = 7.25\times10^{-6}$ emu/g, and for simplicity putting $g_e=2$, then

$$J_{d\,\mathrm{local}} = 4.2\times10^{-3}\ \mathrm{eV} \qquad (14)$$

and from Eq. (12)

$$E_G(\vec{q}=0)=6\times10^{-4}\ \mathrm{eV}\ (\approx 7\ \mathrm{K}).$$

(ii) *Magnetic-ordering temperature.* Assuming that in alloys of this type, the onset of magnetic ordering is controlled by d-band properties, Long and Turner[22] arrived at the following expression for T_c:

$$k_B T_c = \frac{|J_{d\,\mathrm{local}}|^2 cS(S+1)N(E_F)}{30K_0^4(Z/2)^{2/3}}\ ,$$

where $N(E_F)$ is the total unenhanced density of d states at the Fermi level,[49] K_0^{-2} $[=1-IN(E_F)$, I being the d electron, intra-atomic Coulomb repulsion$]$ is the Stoner enhancement factor and Z is the number of d holes per atom. The product $N(E_F)K_0^{-2}$ can be estimated from the low-temperature susceptibility of the host, since

$$N(E_F)K_0^{-2}=4\chi_z/g_e^2\mu_B^2\ . \qquad (15)$$

Putting $g_e=2$ yields

$$N(E_F)K_0^{-2}=23.8\ \mathrm{(states/eV)/atom}\ .$$

With[50] $K_0^2=0.14$ and $T_c=7.85$ K, Eq. (14) yields

$$J_{d\,\mathrm{local}}=1.2\times10^{-2}\ \mathrm{eV};$$

hence

$$E_G(\vec{q}=0)=17.5\times10^{-4}\ \mathrm{eV}\ (\approx 19\ \mathrm{K}).$$

J is, of course, wave-vector-($\vec{q}$) dependent[12]; clearly, the relevant J for Eq. (12) is $J_{d\,\mathrm{local}}(\vec{q}=0)$. The $J_{d\,\mathrm{local}}$ derived from Eq. (13) is in fact $J_{d\,\mathrm{local}}(\vec{q}=0)$, since the g shift probes the static response of the system, whereas that derived from the magnetic-ordering temperature (as estimated from resistivity data) is a weighted "average" over the range $\vec{q}\simeq 0$ to $\vec{q}=2k_F$. The former should consequently be more relevant. However, using either estimate it would thus appear that on the basis of the Doniach–Wohlfarth model there is a possibility of optical-mode excitations being pro-

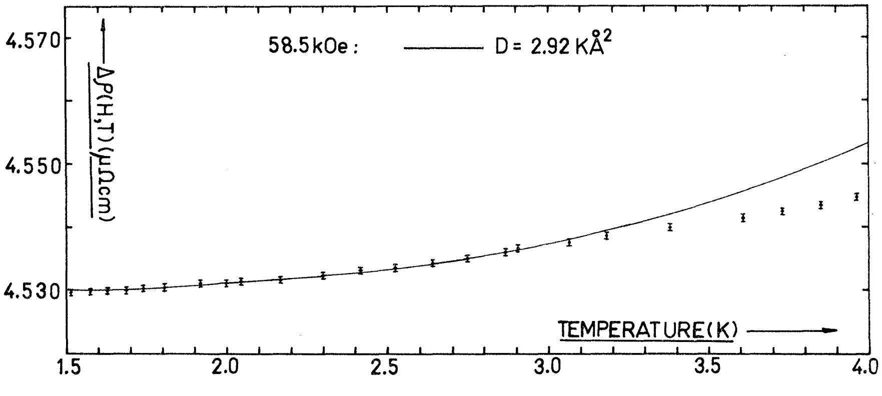

FIG. 7. Low-temperature incremental resistivity data ($\mu\Omega$ cm) in a field of 58.5 kOe, plotted against temperature (K). The solid line is calculated using $D=2.92$ K Å^2.

duced at temperature of interest here in *Pd*Mn. The smallness of $E_G(\vec{q}=0)$ in *Pd*Mn compared with that in the giant-moment systems *Pd*Fe and *Pd*Co[51] is readily understood in terms of the discussion in the introductory section. At $\vec{q}=0$, optical-mode excitations correspond physically to a reversal of an "on-site" impurity spin relative to the surrounding conduction-band polarization; this reversal occurs in the exchange field of this conduction band polarization, hence the smaller the polarization the smaller $E_G(\vec{q}=0)$ becomes.

Further, one not only expects $E_G(\vec{q}\neq 0) < E_G(\vec{q}=0)$—the optic-acoustic mode gap further narrows as $\vec{q}$ increases from zero[20]—but also the application of an applied magnetic field should assist optic-acoustic mode coupling. Optical-mode excitations would add to an already complicated system; it would thus appear that in many respects the *Pd*Mn system is more complicated than the *Pd*Fe or *Pd*Co system. Considerably more theoretical and experimental effort (particularly neutron scattering and magnetization measurements) is required to clarify the ground state of *Pd*Mn and elementary excitation from it.

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lated from Eq. (9) by putting $c_2 = 0$, thus obtaining magnetic field (kOe) of zero, 9.7, 24.8, and 58.5 for c_1 (at.%) of 1.13, 1.62, 1.94, and 2.48, respectively. These estimates are not appreciably different from those listed in Table III; the corresponding values for D are, for magnetic field (kOe) of zero, 9.7, 24.8, and 58.5, D(K Å^2) of 5.42, 5.05, 4.43, and 3.00, respectively.

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[47] D could certainly be field dependent (as opposed to effective-concentration dependent). However previous measurements on *Pd*Fe and *Pd*Co (Ref. 27) indicate that the scale of this variation is considerably smaller [$D(H) - D(0) \simeq 10^{-1} D(0)$ at 60 kOe for Pd − 0.78-at.% Fe, compared with approximately $-0.4\,D(0)$ here]. The sign is also reversed. If D is truly field dependent, an applied field dependence of the saturation moment should be observed (Refs. 27 and 44).

[48] We attribute the entire g shift to coupling with the d band since the dominant contribution to χ_g is attributed to this band. This procedure represents an upper limit on $J_{d\ \mathrm{local}}$ (as derived from Δg).

[49] In Ref. 14, $N(E_F)$ was incorrectly identified as the unenhanced density of d states from one spin orientation. $J_{d\ \mathrm{local}}$ was thus overestimated by a factor of 2.

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