

## Magnetoresistance in Dilute Pd-Based Alloys\*

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(Received 17 November 1970)

Measurements have been made of the magnetoresistance of dilute (approximately 0.1 at.%) alloys of Fe and Co in a Pd matrix over a temperature range from 1.5 to 10 K and for fields of up to 60 kOe. The magnetoresistances follow the predictions of Yosida and show that the giant moments in these alloys can be described by  $g$  factors of  $(2.9 \pm 0.1)$  for both alloys and by  $S$  values of  $(4.5 \pm 0.1)$  for Fe and  $(4.7 \pm 0.1)$  for Co. The enhancement of the  $g$  factors is interpreted in terms of the enhanced Pauli susceptibility of the host and the exchange interaction between the local spins and the  $d$ -band conduction electrons, and is found to correlate well with the ferromagnetic ordering temperatures observed in these alloys if only the on-site spin is considered.

### INTRODUCTION

The giant-moment phenomenon observed in dilute alloys of iron and cobalt in palladium has attracted considerable experimental and theoretical interest in recent years. It was originally revealed by analysis of the paramagnetic susceptibility<sup>1</sup> and the ferromagnetic saturation magnetization of such alloys<sup>2</sup> which indicated anomalously large magnetic moments associated with the impurity ions. The long-range ferromagnetic interaction between the impurities, inferred from the observation that ferromagnetic coupling was found in alloys with as little as 0.1 at.% of impurity,<sup>2</sup> suggested the existence of a long-range spin polarization in the host metal and direct evidence for such a polarization was supplied by neutron-scattering<sup>3</sup> and Mössbauer-effect studies.<sup>4</sup>

The physical picture provided by the neutron scattering is of an on-site impurity moment of  $3.5 \mu_B$  in PdFe, and  $2.1 \mu_B$  in PdCo with an induced polarization extending over some 200 Pd near neighbors of the impurity ion, each contributing an average of about  $0.05 \mu_B$  to the giant moment. The spatial extent of this induced polarization has been attributed to the nearly ferromagnetic itinerant nature of the  $d$  band of the host which suppresses the Ruderman-Kittel-Kasuya-Yosida (RKKY) oscillations and results in an exchange enhancement of the range of the polarization.

A theoretical treatment of the properties of these alloys has been given by Moriya,<sup>5</sup> concentrating firstly on the mechanism by which a local moment is formed upon the impurity site and in its immediate neighborhood, and then upon the enhancement of this resultant moment by the exchange interaction in the host metal.

This paper presents the results of magnetoresistance measurements in such dilute alloy systems in the temperature range 1.5–10 K and for

fields of up to 60 kOe. The measurements show that the giant moments can be described by  $g$  factors of  $(2.9 \pm 0.1)$  for both alloys and by  $S$  values of  $(4.5 \pm 0.1)$  for Fe and  $(4.7 \pm 0.1)$  for Co. The enhancement of the  $g$  factors is interpreted in terms of the enhanced Pauli susceptibility of the host and the exchange interaction between the local spins and the  $d$ -band conduction electrons, and is found to correlate well with the ferromagnetic ordering temperatures observed in these alloys<sup>6</sup> if only the on-site spin is considered.

### EXPERIMENTAL

The alloy specimens were selected from those studied by Williams and Loram<sup>6</sup> in zero field down to 0.5 K and analyzed concentrations of 0.1-at.% Fe and 0.098-at.% Co. They are in the form of foils 2 to 3 mm wide and 10 cm long, and are bent into a hairpin shape before mounting inside a superconducting solenoid in such a way as to keep 9.5 cm of the foil in a longitudinal field and only 0.5 cm in a transverse field. The field homogeneity over the sample length was better than  $4$  in  $10^3$ . Previous investigations<sup>7</sup> have shown that both longitudinal and transverse magnetoresistances of polycrystalline paramagnetic specimens of magnetic alloys have the same field and temperature dependence.

Because of the small changes expected in the resistivity, the foil resistances were measured by altering the current through the specimen to balance a highly stable voltage, the off-balance signal being detected by a Keithley 149 millimicro-voltmeter and the current and voltage being read simultaneously to a resolution of 1 in  $10^5$  on Hewlett-Packard digital voltmeters. The equipment was capable of reproducible measurements to 1 nV and an accuracy in resistance measurement of 2 in  $10^4$ . Temperatures below 4.2 K were stabilized and measured to within 1 mdeg and those

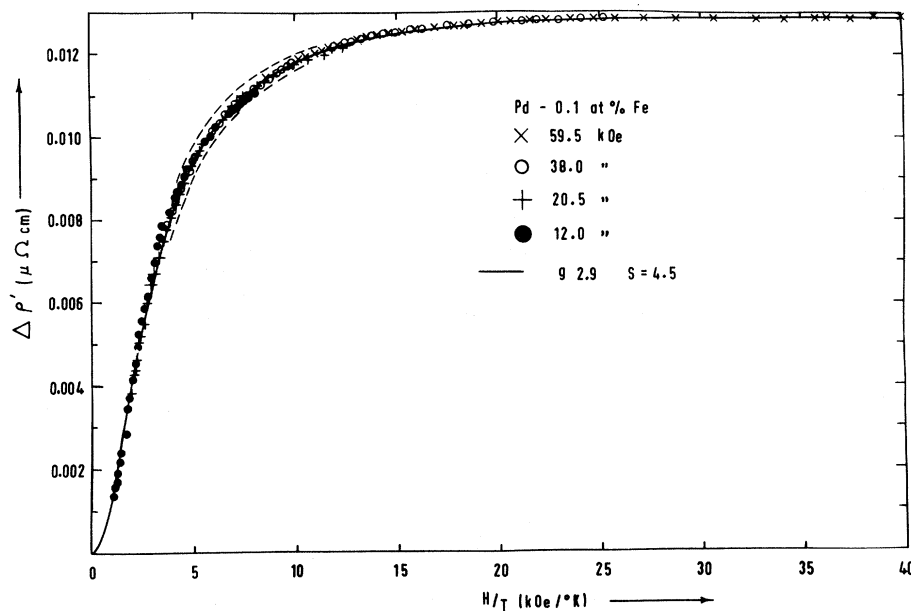


FIG. 1. Impurity magnetoresistivity of Pd 0.1-at. % Fe vs  $H/T$ . The solid line represents Eq. (2), evaluated for the parameters shown in Table I. The dashed curves at the knee of the curve are curves plotted for  $g = 3.1$  and  $g = 2.7$ .

above 4.2 K to better than 1% using vapor pressure and gas thermometer techniques, in combination with carbon resistance thermometers in the temperature control system. The area to length ratios of the samples were measured by a technique similar to that of Dugdale and Basinski.<sup>8</sup>

To facilitate analysis of the results, resistivities were measured in constant fields as a function of temperature between 1.5 and 10 K. The upper limit to the temperature range chosen is such as to eliminate resistivity changes due to the breakdown of Matthiessen's rule.<sup>6,8</sup>

#### ANALYSIS OF RESULTS

The resistivity due to the magnetic impurities at a particular temperature was calculated by subtracting the resistivity of a pure Pd specimen, of resistance ratio 100, from the measured resistivities. It is important to correct for the normal Kohler magnetoresistance due to potential scattering in the host metal and by the magnetic impurities themselves. This was done by assuming that the resistivity due to the impurity spins would be a universal function of  $H/T$  for a given impurity,

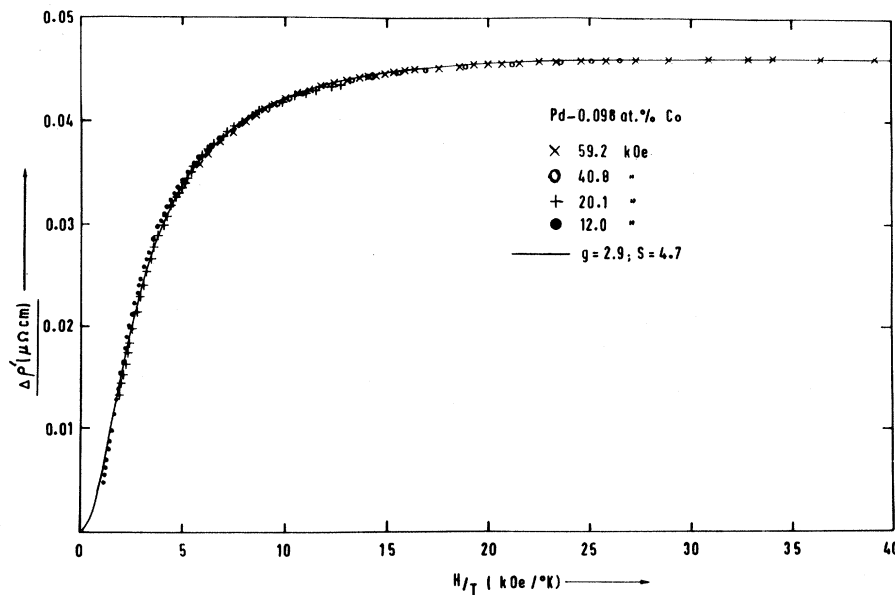


FIG. 2. Impurity magnetoresistivity of Pd 0.098-at. % Co vs  $H/T$ . The solid line represents Eq. (2) evaluated for the parameters shown in Table I.

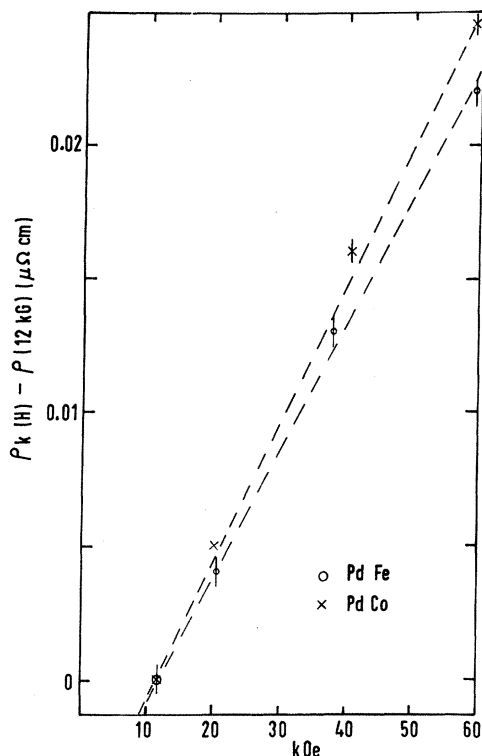


FIG. 3. Kohler magnetoresistivities for the two alloy systems as a function of field.

as shown below, and by calculating the Kohler magnetoresistivity required to ensure this. Figures 1 and 2 demonstrate that the resistivity data

for both alloy systems can be thus plotted as universal function of  $H/T$ , taking

$$\Delta\rho' = \rho_{\text{alloy}}(H, T) - \rho_{\text{Pd}}(T) - \rho_{\text{Kohler}}(H, T) - \rho(H=0, T=\infty),$$

the value of  $\rho(H=0, T=\infty)$  being deduced from the curve-fitting procedure described below.

The Kohler magnetoresistivities were then plotted for the two alloy systems as functions of the magnetic field, as shown in Fig. 3, and were found to agree quite well. The maximum Kohler magnetoresistivity  $[\rho_K(H) - \rho_K(12 \text{ kOe})]$  was of the order of  $0.025 \mu\Omega \text{ cm}$ , whereas the total resistivity of the alloy at the same field was of the order of  $0.3 \mu\Omega \text{ cm}$ . The Kohler magnetoresistivities were found to be temperature independent to within the experimental error of the measurements.

The apparent saturation of the spin-disorder magnetoresistivity appears to indicate that the local spins are all aligned and accordingly the departure of the resistivities from saturation might be expected to follow a Boltzmann temperature dependence of the form

$$\Delta\rho'(H, T) - \Delta\rho'(H, 0) = A e^{-g\mu_B H/kT}, \quad (1)$$

where  $g$  relates to the local spin. Figure 4 demonstrates such behavior for the *PdCo* specimen, and it is clear that the value of  $g$  required is 2.6, in excess of the free-electron value of 2. A similar increase of  $g$  above 2 is observed for *PdFe*.

The temperature dependence of the magnetoresistivity of dilute magnetic alloys is discussed by Yosida<sup>9</sup> and by Beal-Monod and Weiner.<sup>10</sup> The lat-

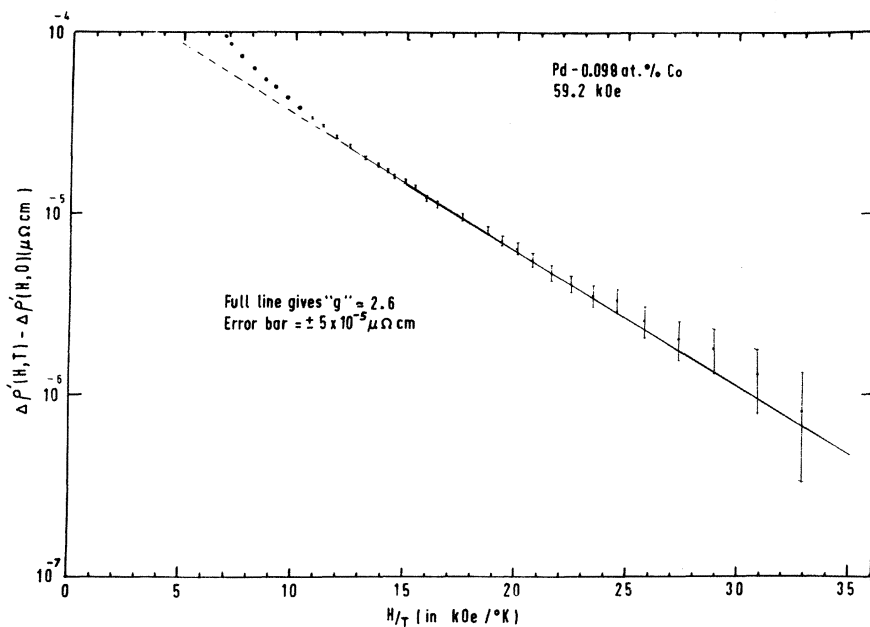


FIG. 4. Impurity magnetoresistivity of Pd 0.098-at. % Co vs  $H/T$ , demonstrating the exponential departure from spin saturation.

ter authors take their calculations to third order to include the effect of a spin-compensated state but the *PdFe* system shows no sign of an appreciable temperature dependence of the zero-field resistivity and such third-order terms may be presumed to be negligible. The *PdCo*, however,

does exhibit some temperature dependence<sup>11</sup> but the magnitude of the change is small enough to suggest that an attempt be made to fit not only the *PdFe* data to the second-order theories but also the *PdCo*. For the purpose of analysis, we use the expression derived by Yosida:

$$\rho(H, T) = Ac \left( V^2 + J^2 \{ \langle S_z^2 \rangle + [S(S+1) - \langle S_z^2 \rangle - \langle S_z \rangle] [1 + \tanh(\frac{1}{2}\alpha)] \} \right. \\ \left. \frac{4V^2 J^2 \langle S_z \rangle^2}{V^2 + J^2 \langle S_z^2 \rangle + J^2 [S(S+1) - \langle S_z^2 \rangle - \langle S_z \rangle] [1 + \tanh(\frac{1}{2}\alpha)]} \right), \quad (2)$$

where  $c$  is the impurity concentration in at. %,  $\alpha = g\mu_B H/kT$ ,  $\langle S_z \rangle$  and  $\langle S_z^2 \rangle$  are the expectation values at the temperature  $T$  of the operators  $S_z$  and  $S_z^2$ , respectively, and the constant  $A$  is given by

$$A = 3\pi m V / 2\hbar e^2 E_f$$

with  $V$  being the host atomic volume. The assumption of isotropic potential and exchange scattering has been made. It should be emphasized that this formulation by Yosida is valid for arbitrary  $V$  and  $J$ . By evaluating the expectation values in zero field, the general expression for the magnetoresistance is given by

$$\Delta\rho(H, T) = \rho(0, T) - \rho(H, T) = Ac \left( J^2 \langle S_z \rangle \tanh(\frac{1}{2}\alpha) + \frac{4V^2 J^2 \langle S \rangle^2}{V^2 + J^2 [S(S+1) - \langle S_z \rangle \tanh(\frac{1}{2}\alpha)]} \right), \quad (3)$$

which can be seen to yield a negative magnetoresistance.

Assuming  $|V| \gg |J|$ , the limiting behavior of the magnetoresistance for small  $\alpha$  is given by

$$\Delta\rho(H, T) = (Ac/18) J^2 S(S+1)(8S^2 + 8S + 3)\alpha^2, \quad (4)$$

assuming Brillouin behavior of the spins, and Fig. 5 shows a plot of  $[\Delta\rho'(H/T) - \Delta\rho'(0)]$  vs  $(H/T)^2$  for the lowest field investigated, the linearity for small values of  $(H/T)^2$  apparently agreeing with Eq. (4). Using the value of  $g$  deduced from the high-field low-temperature data of Fig. 4, values of  $g[S(S+1)]^{1/2}$  can be calculated, yielding values of 12 for *PdFe* and 14.5 for *PdCo*. It will be shown later that this agreement is fortuitous, and that if more accurate values of  $g$  and  $S$ , derived from a fit of the magnetoresistivities over the whole range of field and temperature, are combined with a lifting of the restriction that  $|V| \gg |J|$ , then the values of  $\alpha$  are not sufficiently small to exhibit the true linear dependence. This is shown in Fig. 5 where the dotted line indicates the behavior to be expected at higher temperatures, where, however, the experimental data become increasingly inaccurate due to the breakdown of Matthiessen's rule. Extrapolation of the observed data in Fig. 5 does yield an approximate value for the over-all

change of spin-disorder resistivity, the "step height"  $[\rho(0, T) - \rho(H, 0)]$ .

The magnetoresistivity for various combinations of  $g$ ,  $S$ , and step height can now be computed from Eq. (2) for all values of field and temperature, the value of  $A$  being  $5.78 \mu\Omega\text{cm/at. \%}$  if  $V$  and  $J$  are expressed in eV and a parabolic  $s$  band is assumed with 0.36 electrons per Pd atom and an effective mass  $m/m_0 = 2.2$ .<sup>12</sup> The solid lines in Fig. 1 represent the best fit to the experimental points and the resulting values of  $g$  and  $S$  are shown in Table I. The agreement between the calculated curves and the experimental points is shown at the knee of the curve, the most sensitive region to such curve fitting, and the data can be seen to fall between the curves for the quoted limits. Further confirmation of the assignment of  $g$  and  $S$  is shown in Fig. 6, where the low-temperature high-field data are plotted as a function of  $x = \mu_B H/kT$  with the calculated curve superimposed. Using these values of  $g$  and  $S$ , true saturation of the spin system in the sense of description by a single Boltzmann exponential of the type in Eq. (1) is to be expected for values of  $x$  greater than 1.34, a value not attained in our experiments.

From the resistivities, we are further able to deduce a value for the exchange constant  $J$  since it satisfies the equation<sup>6</sup>

$$|J_{s\text{-local}}|^2 = \frac{2(2S+1)\rho(0, T) - \rho(H, 0) \pm [8S(2S+1)\rho(0, T)\rho(H, 0) + \rho(H, 0)^2]^{1/2}}{2Ac S(2S+1)^2}. \quad (5)$$

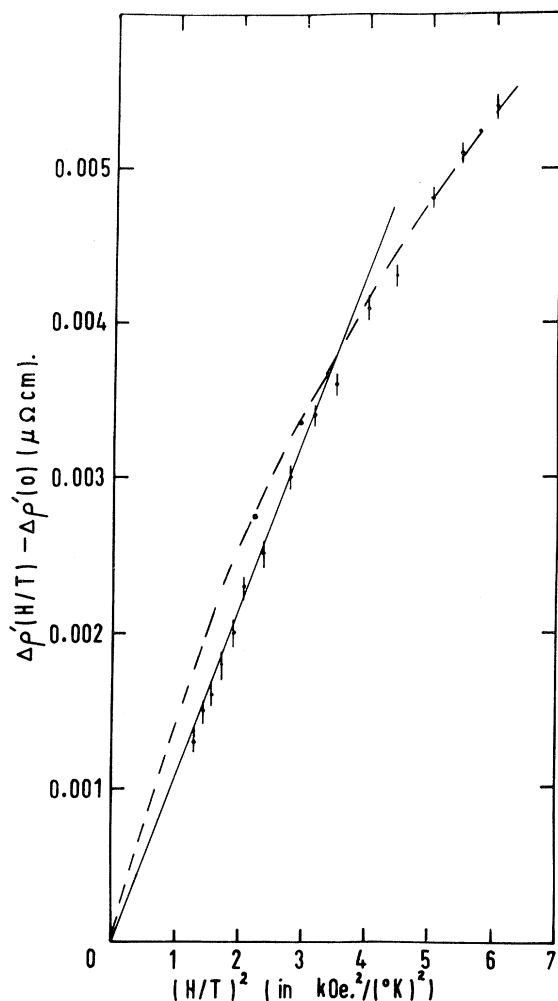


FIG. 5. Impurity magnetoresistivity of Pd 0.1-at.% Fe vs  $(H/T)^2$ .

A value for the potential scattering  $|V|$  due to the impurity spins can similarly be deduced, and the values of  $V$  and  $J$  are included in Table I. It should be noted that there is no evidence from the data for any interaction field other than an exchange field, in contrast to the behavior of the magnetization described by Manuel and McDougald.<sup>13</sup> It can be seen that the values of  $V$  are of comparable magnitude as would be expected from the agreement between the Kohler magnetoresistivities shown in Fig. 3.

TABLE I. Values of  $V$ ,  $J$ ,  $g$ , and  $S$  for the alloy systems studied.

	$V$ (eV)	$J$ (eV)	$g$	$S$	$gS$
<i>PdFe</i>	0.582	0.0163	$2.9 \pm 0.1$	$4.5 \pm 0.1$	$13.1 \pm 0.2$
<i>PdCo</i>	0.496	0.0307	$2.9 \pm 0.1$	$4.7 \pm 0.1$	$13.6 \pm 0.2$

It is of interest to compare the step heights deduced from the present work with those deduced from the zero-field resistivities of the same alloys<sup>6</sup> and Table II shows the comparison. It can be seen that the step height from magnetoresistivity in each case is the larger, indicating that in the zero-temperature limit not all the spins in the alloys are aligned. If we assume that the step height from the magnetoresistivity corresponds to complete alignment of the spins over a distance of the order of magnitude of a mean free path for spin-flip scattering, then it can be shown that the ratio  $\beta$  of step heights in zero field and in a magnetic field will be given by

$$\beta = 1 - [4Sa/(1+4S)](1-a),$$

where  $a$  is the fraction of spins aligned parallel to the  $z$  axis with the remaining spins aligned antiparallel. This expression can be derived from Eq. (2) by substituting  $\langle S_z^2 \rangle = S^2$  and  $\langle S_z \rangle = (1-2a)S$  over the distances considered and assuming, as has been shown to be the case, that  $|V| \gg |J|$ . Such analysis suggests values of  $a = 0.86$  in *PdFe* and  $0.76$  in *PdCo*. These figures may be explained in a semiquantitative way by considering the spin polarization around the impurity ion in Pd. The neutron diffraction experiments suggest a range

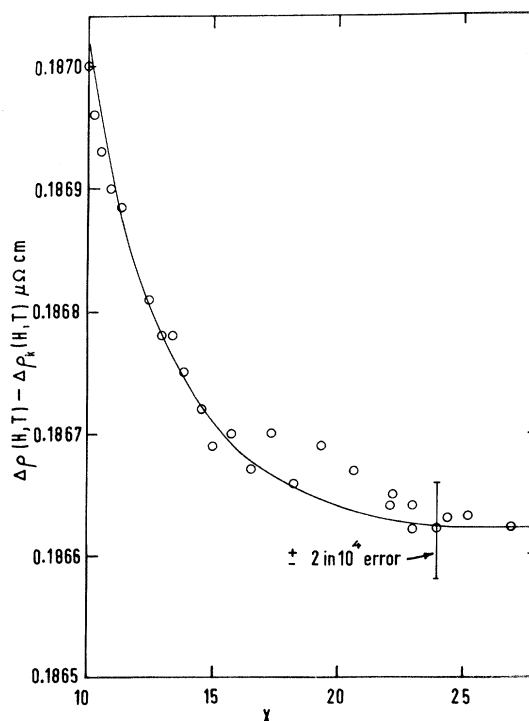


FIG. 6. High-field low-temperature magnetoresistivity of Pd 0.1 at.% vs  $x = \mu_B H / KT$ . The solid curve represents Eq. (2) evaluated for the parameters shown in Table I.

TABLE II. Comparison of step heights for alloy systems studied.

	Zero-field step height $\rho(T_c) - \rho(0)$ $\mu\Omega \text{ cm}$	Magnetoresistivity step height $\rho(0, T) - \rho(H, 0)$ $\mu\Omega \text{ cm}$
<i>PdFe</i>	0.0071	0.0129
<i>PdCo</i>	0.0139	0.046

of the giant moment spin cloud of approximately  $10 \text{ \AA}$ , roughly  $2\frac{1}{2}$  lattice spacings. Beyond this range the spin polarization is oscillatory, so we may assume that impurities sitting in the oscillatory part will order in a random antiferromagnetic way. If  $n$  is the number of atoms within the range of the giant moment and  $c$  is the fractional concentration of impurities, then the probability of finding an impurity outside the range of the giant moment will be  $(1-c)^n$ . Of these impurities, half will align themselves parallel to the spin giving rise to the giant moment, leading to the identification of the parameter  $a$  as  $\frac{1}{2}(1-c)^n$ . Values of  $n$  equal to 720 and 530 in *PdFe* and *PdCo*, respectively, result. In a fcc lattice such as Pd, there are 256 atoms within two lattice spacings and 864 within three. The fact that the observed values of  $n$  fall between these limits shows rough agreement with the neutron-scattering results, and suggests that the polarization range for Fe in Pd is slightly larger than that of Co in Pd.

It has been shown<sup>14</sup> that a  $g$  shift occurs in the process of  $s$ - $d$  exchange given by

$$g_{\text{eff}} = g_s + 2J\chi/g_e\mu_B, \quad (6)$$

where  $g_s$  is the  $g$  factor for the localized spin,  $\chi$  the susceptibility of the host, and  $g_e$  the  $g$  factor for the conduction electrons. In Pd-based alloys, the static spin susceptibility is given by

$$\chi = g_e^2 \frac{\mu_B^2}{4} \left( \frac{N(E_F)}{1 - IN(E_F)} \right) \text{ or } \frac{K_0^{-2} g_e^2 \mu_B^2 N(E_F)}{4}, \quad (7)$$

where  $I$  is the electron-electron interaction in the host  $d$  band and  $N(E_F)$  is the total  $d$ -band density of states. The exchange parameter relating to the largest possible  $g$  shift will accordingly be that

\*Work sponsored in part by the Air Forces Materials Laboratory (AFSC) through the European Office of Aerospace Research (OAR), United States Air Force, under Contract No. F61052-68-C-0011.

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coupling the local spin to the  $d$ -band  $J_{\text{local-d}}$ .

Turner and Long<sup>15</sup> have derived an expression for the Curie temperature of an enhanced system,

$$kT_c = \frac{|J_{\text{local-d}}|^2 CS(S+1)N(E_F)}{30K_0^4 (\frac{1}{2}Z)^{2/3}}, \quad (8)$$

where  $S$  is the on-site spin,  $Z$  the number of conduction electrons per atom, and  $C$  the concentration. Substituting Eqs. (7) and (8) into (6) we have

$$g_{\text{eff}} = g_s \pm \left( \frac{T_c K_0^2 (\frac{1}{2}Z)^{2/3} \chi N_A 5.18 \times 10^3}{CS(S+1)} \right)^{1/2}, \quad (9)$$

where  $N_A$  is the atomic number of the host. For the Pd 0.1-at. % Fe alloy we find  $T_c = 0.78 \text{ K}$ ,<sup>6</sup>  $S = 1.5$  from neutron diffraction,<sup>3</sup> and  $\chi = 7.5 \times 10^{-6} \text{ emu/gm}$ .<sup>16</sup> Assuming  $g_s = 2$  and taking the plus sign in Eq. (9), we have  $g_{\text{eff}} = 2.70$ . In the case of the Pd 0.098-at. % Co, we have  $T_c = 0.79 \text{ K}$ ,<sup>6</sup>  $S = 1$ ,<sup>3</sup> and  $g_{\text{eff}} = 2.93$ .

The enhancement of the spin deduced from the magnetoresistivities above the expected value of 1.5 for Fe and 1 for Co, and in particular the greater enhancement of the Co spin, cannot be simply explained. An attempt to repeat Moriya's calculation for the double-peaked density of states expected to be applicable to Pd proved to be too sharply dependent on the parameters selected to be meaningful.

#### CONCLUSION

We have used magnetoresistivity data on dilute alloys of Fe and Co in Pd to assign values to the potential scattering, the exchange scattering, and the  $g$  and  $S$  values of the impurity giant moments. It is found that the results are compatible with the zero-field measurements on the same alloys over a wider temperature range, if the "on-site" moment alone is involved in the exchange interaction between local spins and the  $d$ -band conduction electrons to give ordering in the alloys.

#### ACKNOWLEDGMENTS

We should like to thank Dr. R. E. Turner for effective and stimulating discussions, and the Science Research Council for financial support for one of us (G. A. S.).

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VOLUME 3, NUMBER 12

15 JUNE 1971

## Structure and Dynamics of a Submonolayer Film Adsorbed on Solid Surfaces\*

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(Received 4 March 1971)

The equilibrium configuration of a submonolayer film adsorbed on a solid surface is analyzed via a model classical Hamiltonian. In this model, the adsorbed atoms or molecules are represented by point particles interacting through connecting springs and the substrate is simulated by a two-dimensional periodic potential. The resulting structure of the film is found to be either commensurate or incommensurate with the periodic potential, depending on the force constant of the springs, the amplitude of the periodic potential, and the degree of mismatch between the length of the unstretched springs and the lattice parameter of the underlying periodic potential. Next, the lattice-vibration spectrum of the film is studied. It is found that when the film structure is incommensurate, the excitation spectrum starts from zero energy. In the commensurate structure the phonon spectrum always starts at a finite frequency. It is noted that this difference between commensurate and incommensurate behavior has an important influence on the thermodynamic properties of the film. Qualitative comparison with experimental data is made.

## I. INTRODUCTION

A submonolayer film adsorbed on a solid surface is a system with complexity rivaling that of a three-dimensional solid: The atoms or molecules in the film can exist in various phases (and undergo phase transitions); and they interact in a complicated manner with the substrate underneath. Recently, the system of He<sup>3</sup> or He<sup>4</sup> submonolayers adsorbed on solid surfaces has received considerable experimental and theoretical consideration.<sup>1</sup> Dash and co-workers have performed a series of experiments with He submonolayers adsorbed on a copper sponge, and found that at low temperatures the contribution of the adsorbed layer to the specific heat has a  $T^2$  dependence. This peculiar behavior seems to indicate that the film behaves like a two-dimensional solid. However, if we neglect the substrate completely, the concentration of He atoms and vapor pressure under those experimental conditions indicates that it is very unlikely for the adsorbed atoms to condense into a solid phase. Taking the substrate into account, condensation into complete registry seems possible, but then the specific heat would have an exponential behavior at low temperature. This apparent paradox remains unexplained at the present time. However, the latter alternative moti-

vates one to ask the following: When the atoms or molecules exist in a solid phase<sup>2</sup> on the surface, does their configuration bear any simple relation with the lattice underneath, and how will their configuration influence thermodynamic properties? In this paper, we attempt to answer these questions for a system described by a model classical Hamiltonian. The equilibrium configuration (the ground state) is obtained by minimizing the system potential energy, i.e., the kinetic part of the energy is neglected. This is equivalent to neglecting zero-point vibrations. Because of the extreme light mass of helium<sup>3</sup> atom, this model may not be directly applicable to a system of He molecules adsorbed on surface: It would be more appropriate for an adsorbate of other rare gas species such as Ne and Ar.

## II. HAMILTONIAN AND THE DIFFERENCE EQUATION

In actuality, for many physisorption systems, various central factors, such as the nature of the adsorbate-adsorbate and adsorbate-substrate interactions, are little known. It seems worthwhile, therefore, to investigate idealized models of the system to arrive at a qualitative understanding of the interplay of various factors in determining the equilibrium configuration and excitation spectrum.