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PHYSICAL REVIEW B

VOLUME 4, NUMBER 1

1 JULY 1971

## Temperature Dependence of the Electrical Resistivity of Dilute Ferromagnetic Alloys

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(Received 20 January 1971)

We present a theoretical discussion of the contribution to the temperature-dependent portion of the electrical resistivity from electron-magnon scattering in dilute ferromagnetic alloys. The transport relaxation rate may be written as a sum of two terms, one (the coherent part) which arises from wave-vector-conserving electron-magnon scattering, and a second (the incoherent part) arises from wave-vector-nonconserving scattering processes. The incoherent part is found to be proportional to  $T^{3/2}$ , and to the impurity concentration, at low concentrations. We also present new data on the temperature dependence of the electrical resistivity of dilute NiMn alloys. A term proportional to  $T^{3/2}$  and to the Mn concentration is observed. We suggest that this term arises from the incoherent scattering of conduction electrons by magnons. The general features of the incoherent contribution to the resistivity relaxation rate are discussed. We have also applied the theory to the ferromagnetic alloys PdFe and PdMn to extract the concentration dependence of the spin-wave stiffness constant  $D$ . The results suggest that the Mn moment is localized at the impurity cell and its near vicinity, in contrast to the well-known giant character of the moment associated with Fe and Co in Pd.

### I. INTRODUCTION

A study of deviations from Matthiessen's rule provides insight into a number of aspects of electric-current conduction in solids. Several mechanisms that lead to violations of Matthiessen's rule have been discussed recently.<sup>1</sup>

In particular, it has been demonstrated that in ferromagnetic metals and alloys, it is necessary to employ a two-current model in order to interpret data on the electrical resistivity  $\rho$  of these systems,<sup>2</sup> since deviations from Matthiessen's rule arise mainly from spin-flip processes that mix the currents associated with the up- and down-spin electrons. However, in many systems, these deviations appear to follow a  $T^{3/2}$  law<sup>3</sup> in the liquid-helium temperature range. This behavior cannot be explained by theories of electron-magnon scattering appropriate to the pure material. The temperature dependence of this contribution to  $\rho$  is similar to that observed in ferromagnetic Pd alloys.<sup>4</sup>

In this paper, we present the results of an experimental study of the temperature dependence of the electrical resistivity of dilute alloys of Mn in Ni. We find that at liquid-helium temperatures, the re-

sistivity contains a term proportional to  $T^{3/2}$ , and to the Mn concentration. Since this contribution is not present in the pure metal, one has a striking violation of Matthiessen's rule associated with the presence of the impurity.

We also present a theoretical model that appears to provide a satisfactory account of the data. We consider the contribution to  $\rho$  from electron-magnon scattering in the alloy. In the disordered system, wave vector is no longer conserved when the electron is scattered by magnons. On the basis of a simple model, we find that the transport relaxation rates can be written as a sum of two terms. One term comes from the coherent scattering of the electrons from the magnons. This term exhibits the same temperature dependence that obtains in the absence of impurities. The second, or incoherent contribution to the relaxation rate, results from the breakdown of wave-vector conservation. This term is proportional to  $T^{3/2}$ , and to the impurity concentration for small concentrations. We suggest that this contribution to the relaxation rate is responsible for the temperature dependence of  $\rho$  observed in the ferromagnetic alloys mentioned above.

In Sec. II, the theory will be discussed. In Sec. III, we present data on the temperature dependence of the electrical resistivity of dilute NiMn alloys, and in Sec. IV, we comment on the variation with concentration of the temperature-dependent portion of  $\rho$  in ferromagnetic Pd alloys.<sup>4</sup>

II. THEORETICAL DISCUSSION

Since it is not possible to perform realistic theoretical studies of the transport coefficients of transition-metal alloys at the present time, we shall resort to a simple model which we feel contains the essential ingredients. We consider a band of conduction electrons exchange coupled to a ferromagnetically aligned system of local moments. For simplicity, we presume the electron wave functions to be plane waves.<sup>5</sup> We use a notation similar to that of an earlier work,<sup>6</sup> and take the conduction-electron-local-moment coupling of the form

$$H_I = V_c \sum_i J_i \vec{S}_i \cdot \int d^3r f_i(\vec{r}) \vec{s}(\vec{r}). \quad (1)$$

In Eq. (1),  $V_c$  is the volume of the unit cell;  $\vec{s}(\vec{r})$  the conduction-electron spin density;  $J_i$ ,  $\vec{S}_i$ , and  $f_i(\vec{r})$  are the exchange constant, spin, and form factor of the local moment in the  $i$ th unit cell, respectively [ $f_i(\vec{r})$  is normalized so  $\int d^3r f_i(\vec{r}) = 1$ ].

We consider a binary alloy, with two magnetic species  $A$  and  $B$  present. Then  $J_i$ ,  $\vec{S}_i$ , and  $f_i(\vec{r})$  will vary from site to site. As a consequence, if an excitation of wave vector  $\vec{q}$  is impressed on the local-moment system (a spin wave), the disturbance seen by the conduction electron contains a mixture of Fourier components, and not just the single component  $\vec{q}$ . Thus, when the electron absorbs or emits a spin wave, the wave vector need not be conserved.

We begin by considering the spin-mixing relaxation rate  $\tau_{ii}^{-1}$ . This quantity can frequently be deduced from resistivity data by employing the two-current model of conduction.<sup>2</sup> From the form of the Boltzmann equation appropriate to the two-current model, one finds<sup>7</sup>

$$\frac{1}{\tau_{ii}} = \frac{2m}{\hbar_B T} \frac{1}{N} \sum_{\vec{k}\vec{k}'} [\hat{x} \cdot \vec{V}(k \uparrow)] [\hat{x} \cdot \vec{V}(k' \downarrow)] f_0(\vec{k} \uparrow) \times [1 - f_0(\vec{k}' \downarrow)] W(\vec{k} \uparrow \rightarrow \vec{k}' \downarrow). \quad (2)$$

In this expression,  $m$  and  $\vec{V}_k$  are the effective mass and group velocity of an electron of wave vector  $\vec{k}$ ,  $\xi$  is the number of conduction electrons per unit cell,  $f_0(k\sigma)$  is the equilibrium distribution function for electrons with spin  $\sigma$ , and  $W(\vec{k} \uparrow \rightarrow \vec{k}' \downarrow)$  is the transition rate for the process  $\vec{k} \uparrow \rightarrow \vec{k}' \downarrow$ .

The electron makes the transition  $\vec{k} \uparrow \rightarrow \vec{k}' \downarrow$  by absorbing a magnon. We calculate  $W$  for this process from Eq. (1). Since the details of the calculation are quite straightforward, and follow earlier work closely, we only sketch the calculation here.

We introduce the operator  $a_i$  that destroys a spin deviation on site  $i$ :

$$S_i^\dagger = (2S_i)^{1/2} a_i.$$

At low temperatures, the electron scatters only from magnons of very long wavelength. Even in the impure system, these waves have the plane-wave form. Thus,

$$a_i = N^{-1/2} \sum_{\vec{k}} a_{\vec{k}} e^{i\vec{k} \cdot \vec{R}_i}.$$

For the transition rate we find

$$W(\vec{k} \uparrow \rightarrow \vec{k}' \downarrow) = \frac{\pi}{N^3} \sum_{ij} \sum_{\vec{q}} I_i(\vec{k} - \vec{k}') I_j^*(\vec{k} - \vec{k}') \times e^{i(\vec{k} - \vec{k}' - \vec{q}) \cdot (\vec{R}_i - \vec{R}_j)} n_{\vec{q}} \delta(\epsilon_i(\vec{k}) - \epsilon_i(\vec{k}') + \hbar\omega_{\vec{q}}). \quad (3)$$

In Eq. (2),  $\epsilon_\sigma(\vec{k})$  is the energy of the electron ( $\vec{k}\sigma$ ),  $\omega_{\vec{q}}$  and  $n_{\vec{q}}$  are the frequency and occupation number of the magnons of wave vector  $\vec{q}$ , and if  $F_i(\vec{k})$  is the Fourier transform of  $f_i(\vec{r})$ , we define

$$I_i(\vec{k}) = J_i (S_i)^{1/2} F_i(\vec{k}).$$

We need to consider the sum

$$S = \frac{1}{N^2} \sum_{ij} I_i I_j^* e^{i\vec{q} \cdot (\vec{R}_i - \vec{R}_j)}.$$

To evaluate  $S$ , we replace  $I_i$  by  $\bar{I} + \delta I_i$ , where  $\bar{I} = C_A I_A + C_B I_B$  is the average value of  $I_i$ , and  $\delta I_i = I_i - \bar{I}$ . Then

$$S = \bar{I}^2 \delta_{\vec{q},0} + \frac{1}{N} \sum_{ij} \delta I_i \delta I_j^* e^{i\vec{q} \cdot (\vec{R}_i - \vec{R}_j)} + \frac{2}{N^2} \text{Re} \left( \bar{I} \sum_{ij} \delta I_i^* \delta I_j e^{i\vec{q} \cdot (\vec{R}_i - \vec{R}_j)} \right).$$

The third term vanishes upon summing over  $i$ . In the second term, the subset of terms with  $i = j$  all give positive definite contributions to  $S$ , while the remainder average to zero for a random alloy. We then find

$$S = [C_A I_A + C_B I_B]^2 \delta_{\vec{q},0} + (1/N) C_A C_B [I_A - I_B]^2, \quad (4)$$

where  $C_A$  and  $C_B$  are the concentration of species  $A$  and  $B$ .

From Eq. (4), one sees that  $W(k \uparrow \rightarrow k' \downarrow)$  consists of a sum of two terms. The first term describes "coherent" scattering of the electron by a uniform background matrix described by the average value  $\bar{I}$  of the coupling parameter. In this contribution to  $W$ , the wave vector is conserved. The second term describes the "incoherent" or wave-vector-non-conserving part of the relaxation rate that arises from the presence of a disorder.

It is straightforward to insert Eq. (4) into Eq. (3), and then to obtain the transport relaxation rate  $\tau_{ii}^{-1}$ . We have carried out such a calculation for a spherically symmetric parabolic electron band with an exchange splitting  $\Delta$  between the up- and down-

spin bands. We find for the coherent part

$$\left(\frac{1}{\tau_{\uparrow\downarrow}}\right)_{\text{coh}} = \frac{3\pi}{8} (C_A J_A S_A^{1/2} + C_B J_B S_B^{1/2})^2 \times \frac{\xi k_B T}{\hbar^2 \epsilon_F D k_F^2} f\left(\frac{\epsilon_m}{k_B T}\right), \quad (5a)$$

where

$$f(x) = \int_x^\infty \frac{d_y y e^y}{(e^y - 1)^2} = \begin{cases} -\ln x, & x < 1 \\ x e^{-x}, & x > 1 \end{cases}.$$

For the pure metal (where  $C_A$  or  $C_B = 1$ ) this result reduces to the form given previously by Fert and Campbell<sup>2</sup> although the present notation differs slightly. In Eq. (5a),  $\epsilon_F$  and  $k_F$  are the Fermi energy and wave vector,  $D$  is the spin-wave stiffness constant, and  $\epsilon_m = \hbar D k_F^2 \Delta^2 / 2\epsilon_F^2$ . When  $k_B T \ll \epsilon_m$ , there are no thermal magnons with wave vector large enough to scatter an electron from the up-spin to the down-spin Fermi surface, so  $\tau_{\uparrow\downarrow}^{-1}$  varies like  $e^{-\epsilon_m/k_B T}$  with  $T$  in this region. When  $k_B T \gg \epsilon_m$ , one has  $\tau_{\uparrow\downarrow}^{-1} \propto T \ln(k_B T/\epsilon_m)$ . This dependence on  $T$  obtains rather than the often quoted  $T^2$  law for the electron-magnon contribution to the transport relaxation rate because the  $(1 - \cos\theta)$  factor is absent from the expression for  $\tau_{\uparrow\downarrow}^{-1}$ .

To compute the contribution to  $\tau_{\uparrow\downarrow}^{-1}$  from incoherent scattering, we assume the form factors for the  $A$  and  $B$  spins are identical. Then

$$\left(\frac{1}{\tau_{\uparrow\downarrow}}\right)_{\text{incoh}} = \frac{27\pi^3}{32} \frac{\xi}{\hbar \epsilon_F} C_A C_B (J_A S_A^{1/2} - J_B S_B^{1/2})^2 \Gamma\left(\frac{3}{2}\right) \times \xi \left(\frac{3}{2}\right) \left(\frac{k_B T}{\hbar D k_F^2}\right)^{3/2} \int_0^2 d\eta \eta (1 - \frac{1}{2}\eta^2) [F(k_F \eta)]^2. \quad (5b)$$

Since even long-wavelength magnons give rise to large-angle scattering of the electron in the presence of disorder, the form factor appears in Eq. (5b).

We next comment on the results in Eqs. (5). While our simple model cannot be applied to a real alloy in a reliable manner for quantitative purposes, a number of features can be expected to be generally valid.

In the pure material, the results reduce to the values appropriate to the pure matrix. For small impurity concentration  $c$ ,  $(\tau_{\uparrow\downarrow}^{-1})_{\text{coh}}$  is well approximated by its value in the host, and  $(\tau_{\uparrow\downarrow}^{-1})_{\text{incoh}}$  is proportional to  $c$ .

As noted above, the contribution to  $\tau_{\uparrow\downarrow}^{-1}$  from coherent scattering "freezes out" when  $k_B T \ll \epsilon_m$ . However,  $(\tau_{\uparrow\downarrow}^{-1})_{\text{incoh}}$  exhibits the power-law dependence given in Eq. (5b) at all temperatures, even when  $k_B T \ll \epsilon_m$ . By means of a wave-vector-nonconserving collision, even a very-long-wavelength magnon can scatter an electron from the up-spin Fermi surface to a point near the down-spin Fermi surface. Thus, at sufficiently low temperatures, the incoherent scattering should dominate the co-

herent scattering in ferromagnetic alloys.

The electron-magnon interactions also contribute to the intraband scattering probabilities  $1/\tau_{\uparrow\uparrow}$  and  $1/\tau_{\downarrow\downarrow}$ . From the Boltzmann equation we have

$$\left(\frac{1}{\tau_{\sigma\sigma}}\right) = \frac{2m}{\xi k T N} \sum_{\mathbf{k}\mathbf{k}'} \hat{x} \cdot \vec{V}(\mathbf{k}\uparrow) \{ \hat{x} \cdot [\vec{V}(\mathbf{k}\uparrow) - \vec{V}(\mathbf{k}'\downarrow)] \} f_0(\mathbf{k}\uparrow) \times [1 - f_0(\mathbf{k}'\downarrow)] W_s(\mathbf{k}\uparrow - \mathbf{k}'\downarrow), \dots, \quad (6)$$

with the same definitions as for Eq. (2). From a very similar calculation to that for  $1/\tau_{\uparrow\downarrow}$ , we have the electron-magnon contributions through coherent and incoherent scattering:

$$\left(\frac{1}{\tau_{\sigma\sigma}}\right)_{\text{coh}} = \frac{3\pi\xi}{16} \frac{(C_A J_A S_A^{1/2} + C_B J_B S_B^{1/2})^2}{\hbar E_F} \left(\frac{k_B T}{\hbar D k_F^2}\right)^2 \xi(2) \quad (7)$$

and

$$\left(\frac{1}{\tau_{\sigma\sigma}}\right)_{\text{incoh}} = \frac{27\pi^3}{64} C_A C_B \frac{|J_A S_A^{1/2} - J_B S_B^{1/2}|^2}{\hbar \epsilon_F} \times \xi \left(\frac{k_B T}{\hbar D k_F^2}\right)^{3/2} \Gamma\left(\frac{3}{2}\right) \xi\left(\frac{3}{2}\right) \int_0^2 d\eta \eta^3 |F(k_F \eta)|^2. \quad (8)$$

When  $k_B T \ll \epsilon_m$ , we again find  $(\tau_{\sigma\sigma}^{-1})_{\text{coh}} \sim e^{-\epsilon_m/k_B T}$ , whereas  $(\tau_{\sigma\sigma}^{-1})_{\text{incoh}}$  remains proportional to  $T^{3/2}$ .

It should be mentioned that Kagan and co-workers<sup>8</sup> have also presented a theory of the electrical resistivity of ferromagnetic alloys, in which both incoherent and coherent scattering of the electrons from the spins is considered. They also find a  $T^{3/2}$  term in the low-temperature resistivity of the alloy. There are significant differences in detail between the present theory and the theory of Kagan *et al.* In Ref. 8, the two-current nature of the conduction is ignored, and the presence of the exchange splitting of the conduction band is not taken into account. As we have seen, these features of the problem must be taken into account to produce the correct temperature dependence of the electrical resistivity. Furthermore, we find that the exchange constant  $D$  appears in the final expressions for the relaxation rate, while Kagan *et al.* have apparently employed an approximate theory of the spin dynamics of the disordered system which inserts the Curie temperature  $k_B T_C$  in place of  $D$ . In their model,  $D$  and the Curie temperature are proportional. We find that it is the spin-wave exchange constant  $D$  of the alloy that appears in the formulas for the low-temperature resistivity, and not the Curie temperature. This is an important distinction, since it is not clear that in the disordered system,  $D$  and  $T_C$  are proportional to each other. This is particularly the case for transition metals and their alloys where a complete theory must take account of the itinerant character of the magnetic electrons. We shall see below that one may extract the concentration dependence of  $D$  from resistivity data, if the  $T^{3/2}$  term

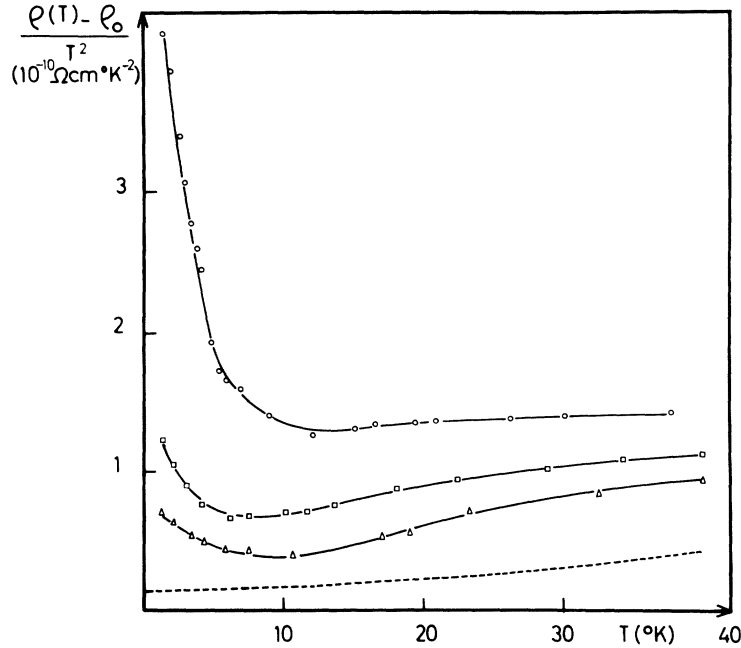


FIG. 1. A plot of  $[\rho(T) - \rho(0)]/T^2$  as a function of temperature for NiMn with 0.4 at. % Mn ( $\Delta$ ); 1 at. % Mn ( $\square$ ); and 2 at. % Mn ( $\circ$ ). The upturn of the curves for  $T < 10^\circ\text{K}$  is the  $T^{3/2}$  behavior discussed in the text. The dotted curve is  $[\rho(T - \rho(0))]/T^2$  for pure Ni. The term  $\rho_{\uparrow\downarrow}(T)$  for the three alloys corresponds approximately to the difference between the full and dotted curves.

and its concentration dependence can be observed. It is important to realize that one obtains information only about the long-wavelength magnons by this procedure, and  $D$  and  $T_C$  may not exhibit the same concentration dependence.

### III. EXPERIMENTS ON Ni ALLOYS

A two-current model has been shown experimentally to describe electrical conduction rather well in dilute Ni-based alloys, the resistivity being given in this model by

$$\rho = \frac{\rho_{\uparrow}\rho_{\downarrow} + \rho_{\uparrow\downarrow}(\rho_{\uparrow} + \rho_{\downarrow})}{\rho_{\uparrow} + \rho_{\downarrow} + 4\rho_{\uparrow\downarrow}}, \quad (9)$$

where  $\rho_{\uparrow}$ ,  $\rho_{\downarrow}$  are the resistivities for spin-up ( $\uparrow$ ) and spin-down ( $\downarrow$ ) conduction electrons and  $\rho_{\uparrow\downarrow}(T)$  is proportional to the scattering probability  $1/\tau_{\uparrow\downarrow}$ , between spin-up ( $\uparrow$ ) and spin-down ( $\downarrow$ ) bands.

It is assumed that a quasi-Matthiessen's rule holds for intraband scattering, so  $\rho_{\uparrow\downarrow}(T)$  is zero. Then, at low temperatures, when the residual resistivity is dominant  $\rho_{\uparrow}^0, \rho_{\downarrow}^0 \gg \rho_{\uparrow}^{\uparrow}, \rho_{\uparrow\downarrow}$ , we can expand Eq. (9) to give

$$\rho(T) - \rho_0 = \left(1 + \frac{(\alpha - \mu)^2}{(\alpha + 1)^2 \mu}\right) \frac{\rho_{\uparrow}^{\uparrow}(T)\rho_{\downarrow}^{\downarrow}(T)}{\rho_{\uparrow}^{\uparrow}(T) + \rho_{\downarrow}^{\downarrow}(T)} + \left(\frac{\alpha - 1}{\alpha + 1}\right) \rho_{\uparrow\downarrow}(T),$$

where  $\alpha = \rho_{\uparrow}^0/\rho_{\downarrow}^0$  and  $\mu = \rho_{\uparrow}^{\uparrow}(T)/\rho_{\downarrow}^{\downarrow}(T)$ .

The  $\alpha$  values are characteristic of different impurities, whereas the  $\mu$  and  $\rho_{\uparrow\downarrow}$  for coherent scattering are independent of the impurity; these values have been tabulated by Fert.<sup>7</sup> It can be seen that

very different low-temperature behavior can be expected for alloys where the  $\alpha$  values are very different. In particular, if  $\rho_{\uparrow\downarrow}(T)$  is the dominant thermal term at low temperatures, it will be visible only for alloys where  $\alpha$  is very different from 1.

For NiMn alloys ( $\alpha \approx 15$ ) with Mn concentration varying from 0.2 at. % to 2 at. %, it has been found that  $\rho(T) - \rho_0$  varies as  $T^{3/2}$  at low temperatures, and with the strength of the  $T^{3/2}$  term approximately proportional to the Mn concentration (see Fig. 1). Both the temperature and the concentration dependence appear to identify this behavior with the incoherent magnon scattering of the model of Sec. II. For Ni alloys with  $\alpha$  near 1 (V or Cr impurities) only very small  $T^{3/2}$  terms are seen, as would be expected, but for NiCo and NiFe with large  $\alpha$  values, a  $T^{3/2}$  is observed; but it is smaller than for Mn and not proportional to the impurity concentration. NiMn seems to be a special case, with a high effective value of  $J$  for Mn. It is known<sup>9</sup> from the magnetic properties of Ni alloys that NiMn is close to the critical condition for passing from a strong positive impurity moment (such as in NiFe) to a negative impurity moment (such as in NiCr). It appears that the strong effective  $J_{Mn}$  is a consequence of this instability.

### IV. TEMPERATURE DEPENDENCE OF THE RESISTIVITY IN FERROMAGNETIC Pd-BASED ALLOYS

For a ferromagnetically aligned set of local moments randomly distributed throughout a paramagnetic host, one expects the dominant contribution to  $\rho$  to come from the incoherent scattering, at small concentrations. From Sec. II, when  $C_B = 0$  and  $C_A$

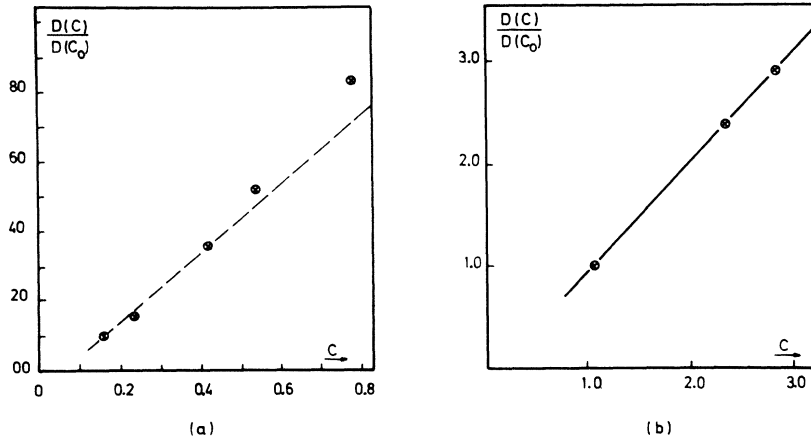


FIG. 2. The dependence of the spin-wave stiffness constant  $D$  on impurity concentration for (a)  $PdFe$  alloys, and (b)  $PdMn$  alloys.

$= C \ll 1$ , the coherent scattering contribution to  $\Delta\rho$  is proportional to  $C^2$ , whereas the incoherent contribution is proportional to  $C$ . The present theory gives for the temperature-dependent portion of  $\rho$  the relation

$$\rho(T) = cKT^{3/2}/D^{3/2}, \quad (10)$$

where  $K$  is a constant that may be obtained from Sec. II. As we have commented earlier, a contribution to  $\rho$  with the  $T^{3/2}$  temperature dependence has been observed in  $PdMn$ ,  $PdFe$ , and  $PdCo$ .<sup>4</sup>

Turner and Long<sup>10</sup> have also presented a theory of the contribution to  $\rho$  from electron-magnon scattering in these alloys. They find the dependence on  $T$  exhibited in Eq. (10). However, in their theory, they employ a specific approximation scheme to describe the dynamical properties of the local moments. They find  $D$  is proportional to the concentration  $c$ , so that  $\rho \sim T^{3/2}/c^{1/2}$  in their theory.

We feel it is important to realize that Eq. (10) holds for  $c \ll 1$ , independent of any specific description of the dynamics of the local-moment system. The constant  $D$  is the spin-wave exchange stiffness of the alloy; it may not be proportional to  $c$  even for  $c \ll 1$ , particularly in the giant-moment Pd alloys. Equation (10) may not be used to obtain reliable absolute values of  $D$  in the alloys because our oversimplified model gives an unreliable value for  $K$ . However, we feel the dependence of  $\rho(T)$  on  $c$ ,  $T$ , and  $D$  is correctly given by Eq. (10). As a consequence, since the  $T^{3/2}$  law is accurately obeyed for  $T \ll T_c$  in the Pd-based alloys, one can extract the concentration dependence of  $D$  from the data. We have done this for  $PdFe$  and  $PdMn$  and the results are presented in Fig. 2. We comment on the results for each system.

(i)  $PdFe$ . The most striking feature of the plot

is the strong deviation of  $D(c)$  from a linear dependence on  $c$ , even for  $c < 1\%$ . This is not surprising, in view of the well-known giant moment associated with Fe in Pd. Some authors<sup>11</sup> have analyzed magnetization and specific-heat data on more concentrated samples, and have presumed  $D(c)$  to vary linearly with  $c$  for  $c < 1\%$ . We see that quantitative conclusions based on such an extrapolation are subject to error.

(ii)  $PdMn$ . One sees that  $D(c)$  is accurately linear with  $c$  up to 3%, the highest concentration studied. The linear dependence of  $D$  on  $c$  gives strong support to other experimental results which indicate that  $PdMn$  local moment is well localized spatially in contrast to  $PdFe$  and  $PdCo$ , which produce spread-out giant moments.

Physically, this can be understood by considering only the  $d$  electrons. When a virtual bound state is formed just above or just below a band edge, it is spatially very extended (see, e. g., Kanamori<sup>12</sup>); for Pd the Fermi energy is very close to the top of the  $d$  band. For  $PdMn$  the full spin-up ( $\uparrow$ )  $d$  states are well below  $E_F$  and the empty spin-down ( $\downarrow$ ) states are well above, so any  $d$  disturbance is short range. For  $PdCo$  or  $PdFe$ , on the other hand, the spin-up ( $\uparrow$ ) states are still well below  $E_F$ , but the spin-down ( $\downarrow$ ) states are partially occupied (because of the neutrality condition), and so are around  $E_F$ . They are thus spatially extended, with an associated disturbance in the Pd  $d$  band which is ferromagnetic and quite long ranged, since the range is greatly increased by the strong exchange enhancement in the host matrix. For  $PdCr$ , this picture gives an antiferromagnetic coupling to any Pd neighbor polarization. We refer the reader to Ref. 13, where a quantitative discussion of these points is presented.

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<sup>1</sup>For recent discussions of the breakdown of Matthiessen's rule, see M. J. Rice, Phys. Rev. Letters **23**,

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## Theory of Macroscopic Excitations of Magnons\*

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(Received 4 February 1971)

The excitation of ferromagnetic magnons by a macroscopic time-varying magnetic field is analyzed quantum mechanically. The quantization of the spin excitations is made by the method of Holstein and Primakoff, generalized for the case of a general nonuniform static field. Both linear and nonlinear excitation mechanisms are considered, with the driving field being either perpendicular or parallel to the static field. Particular attention is given to the coherence properties of the magnon states generated.

### I. INTRODUCTION

It is well known that strongly magnetic systems have low-energy wavelike excitations called spin waves or magnons. The concept of spin waves was introduced by Bloch<sup>1</sup> in 1930 to explain the thermodynamic properties of ferromagnets at low temperatures. The interest in this field was renewed by the work of Suhl,<sup>2</sup> which explained the saturation effects observed in ferromagnetic resonance experiments as arising from the unstable growth of spin waves. Considerable progress in the subject was made after this work with the development of several methods for generating and detecting spin waves, measuring their properties, and studying their interactions with other elementary excitations.

Besides Suhl's nonlinear process, it was found by Morgenthaler<sup>3</sup> and Schlömann<sup>4</sup> that spin waves could be parametrically excited by an rf field applied parallel to the static field, instead of perpendicular. Schlömann<sup>5</sup> also first proposed a linear excitation mechanism, making use of an rf field perpendicular to a nonuniform static field. This excitation method proved to be very useful in low-

magnetic-loss materials, such as yttrium iron garnet, providing a new possibility for delaying and processing information contained in electromagnetic signals. It has been used in several experimental investigations of properties of spin waves, such as their coupling to elastic waves in spatially<sup>6,7</sup> and time-varying<sup>8</sup> magnetic fields, their amplification by parametric pumping,<sup>9</sup> their interaction with light beams,<sup>10</sup> and others.

The macroscopic excitation of spin waves in ferromagnets has been discussed both semiclassical and quantum mechanically. The semiclassical treatments are mostly based on the magnetization equation-of-motion method introduced by Herring and Kittel.<sup>11</sup> In most of the previous quantum treatments one uses the equations of motion for the magnon creation and annihilation operators of the Holstein-Primakoff<sup>12</sup> formalism. The solutions of these equations are used to find the time dependence of the expectation values of the magnetization and other related operators. These results, however, do not contain all the information that can be obtained from the knowledge of the time-evolution operator.