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s-d Exchange Integrals Obtained from Experiment in Cu:Mn, Ag:Mn, and Au:Mn

D. A. Smith and G. B. Smith*
Department of Physics, Monash University, Clayton, Victoria, Australia

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s-d exchange integrals are estimated (i) from the energies of virtual bound states appearing in the optical absorption and (ii) from susceptibility, specific-heat, and resistivity measurements above T_K, using the high-temperature results of the s-d exchange model. The resistivity gives values which are much too low; it is suggested that this arises from neglecting nonlogarithmic terms especially important in the theory of transport properties.

I. INTRODUCTION

There are two parts to this paper. First, we wish to point out that the validity of the s-d exchange model of dilute alloys is proscribed by optical absorption and photoemission experiments, and the s-d exchange integral can be estimated. The second part is a quantitative test of the available predictions of the s-d exchange model at high temperatures, where perturbation theory *à la* Abrikosov[1] might be expected to hold good. That the current theories are not quantitatively successful even at 100 times the Kondo temperature is due, we believe, to neglecting nonleading logarithmic terms. Nevertheless, values for the s-d exchange integral J can be obtained by force fitting the available experimental data, which we do for Mn impurities in the noble metals. J values from susceptibility and specific-heat measurements are similar and also compare favorably with $J = -0.45$ eV derived from optical data on Ag : Mn. But the resistivity yields J's which are much too low, and it is suggested that this is again due to the omitted nonleading logarithmic terms.

II. s-d EXCHANGE INTEGRALS FROM OPTICAL DATA

Recent optical absorption[2] and photoemission[3] experiments on Ag : Mn reveal the presence of two widely separated virtual bound states, one above and one below the Fermi level. This is strong evidence for large intra-atomic Coulomb interactions and hence a well-defined localized spin (presumably $S=\frac{5}{2}$) at each Mn impurity. This is precisely the situation where one ought to be able to do perturbation theory starting from the "atomic limit," or, more specifically, the canonical transformation first given by Schrieffer and Wolff[4] which leads to the s-d exchange model.

For Mn which is assumed to be an S-state ion ($3d^5$ or $3d^5 4s^2$) the passage from Anderson's model to the s-d exchange model is straightforward and is given by Schrieffer.[5] One finds an impurity Hamiltonian with potential and exchange scattering terms

$$H_{\text{imp}} = \frac{5}{N} \sum_{\vec{k}\vec{k}'\sigma\sigma'} P_2(\cos\theta_{\vec{k}\vec{k}'}) \{V\delta_{\sigma\sigma'} - J\vec{S}\circ\vec{s}_{\sigma\sigma'}\} c^{\dagger}_{\vec{k}\sigma} c_{\vec{k}'\sigma'} , \tag{2.1}$$

where the fivefold degeneracy of the impurity d orbitals is responsible for the $\vec{k}$-space form factor P_2. Here $\vec{s}_{\sigma\sigma'}$ are matrix elements of half the Pauli matrices and V and J are fixed by the energies of the two virtual bound states measured from the Fermi level. Let T_+ and $-T_-$ be the energy changes on adding an electron and a hole, respectively, at the Fermi level to the Mn impurity in its ground state; for stability, both must be positive. Then T_+ and T_- are the one-electron energy levels of the virtual bound states from the Fermi level. One finds that

$$V = -\tfrac{1}{2} v^2 (T_-^{-1} + T_+^{-1}), \qquad J = \tfrac{2}{5} v^2 (T_-^{-1} - T_+^{-1}), \tag{2.2}$$

where

$$v = (4\pi N)^{1/2} \int_0^\infty r^2 dr\, \phi(r)\, V(r)\, j_2(k_F r) \tag{2.3}$$

is the covalent admixture matrix element and its energy dependence is suppressed by putting $k = k' = k_F$. For Mn, J is necessarily negative leading to the Kondo-Suhl instability, but since J is really energy dependent and changes sign for energies below T_- or above T_+, the cutoff energy D in the usual theories is not the Fermi energy but more nearly $|T_+ T_-|^{1/2}$. Contrary to general expectation, exchange scattering dominates potential scattering here; V is small because the two resonances contribute with opposite signs and this is borne out by a very small residual thermopower.[6]

Now Meyers, Wallden, and Karllson[2] found impurity absorption in Ag : Mn below the d-band edge at 1.6 and 3.25 eV which they attribute to virtual bound states. Photoemission work by Norris and Wallden[3] shows extra structure at 3.25 eV only so that $T_+ = 1.6$ and $T_- = 3.25$ eV. Myers *et al.* also studied Cu : Mn and found no discernible structure from virtual bound states, presumably because they are hidden in the d band, so $|T_\pm|$ must be > 2 eV in Cu : Mn. Since $v \simeq 1.1$ eV for Mn in Ag, we estimate $J = -0.45$ eV and $V = -0.19$ eV for Ag : Mn. J values for Cu : Mn and Au : Mn are not expected to be much different, but the potential scattering [which by Eq. (2.1) would be more sensitive to the level positions] can in fact be ignored altogether. It enters the theoretical expressions only as the $l = 2$ phase shift $\eta_2 = -\tan^{-1}(\pi\rho V)$, where ρ is the one-spin density of states per atom at the Fermi level, and for Ag : Mn η_2 is only 5°. Finally, the cutoff parameter D is 2.3 eV or 2.6×10^4 °K.

III. ANALYSIS OF EXPERIMENTAL DATA AT HIGH TEMPERATURES

We shall consider only the magnetic susceptibility, specific heat, and electrical resistivity, which manifest the Kondo effect in a fairly simple way and for which reasonable data are available. The first question to ask is whether the data fit the current theoretical predictions. If they do, then a value for J may be obtained from each experimental quantity. Only dilute alloys of Mn in the noble metals will be considered so that J's may be compared with the optical estimate for Ag : Mn.

Previous attempts to correlate J values in this way suffer by neglecting one or more of the following considerations: (i) The impurity is often considered to have no orbital degeneracy, so that the form factor $P_2(\cos\theta_{\vec{k}\vec{k}'})$ is left out of the exchange Hamiltonian. When it is included, the coefficients of J in expressions for various alloy properties are altered in different ways. (ii) Even if $T > T_K$, where perturbation theory should be applicable, the leading $\ln T$ terms in each order of perturbation theory must be summed to all orders, as Abrikosov first did, to avoid numerical error. To see this, note that the ratio of successive leading terms is

$$x = J\rho\ln(T/D) = \frac{\ln(T/D)}{\ln(T_K/D)} ,$$

where T_K is the Kondo temperature. Thus if $T_K = 0.026$ °K and $D = 2.6\times10^4$ °K, then $x = \frac{2}{3}$ at 2.6 °K and $\frac{1}{3}$ at 260 °K. For this typical example the first temperature is already 100 times the Kondo temperature yet increasing it by a further factor of 100 improves convergence only by a factor of 2. (iii) Nonleading $\ln T$ terms in each order of perturbation theory, viz., those with less than the maximum power of $\ln(T/D)$, are more important for d-orbital impurities than for s-orbital ones because they arise partly from unlinked diagrams which are associated with extra factors of 5. Consequently, expressions for various properties derived by summing only the leading $\ln T$ terms to all orders ought to describe the variation of these properties with $\ln T$ satisfactorily, but will not be adequate to give the constant temperature-independent contributions.

In view of these comments the following procedure will be adopted. First, an experimental quantity X is chosen which is expected to be some simple function of $\ln T$ only. Plotting X against $\ln T$ ought, in principle, to be the correct way of testing the theoretical expressions for X, but in most experiments to date the temperature has not been varied over more than a decade and in this range X is usually linear in $\ln T$ to the accuracy of the data. Remember that we have in mind only the high-temperature limit, say, $T/T_K > 100$; these remarks obviously do not apply to measurements near or below T_K. In these circumstances the theoretical expression must be assumed to give the slope $dX/d(\ln T)$ correctly, and then T_K and hence J may be obtained. The theories are tested by the self-consistency of J values required to fit different experimental quantities.

The predictions of the s-d exchange model, which is specified by (2.1) with $V = 0$, are conveniently presented in terms of the Kondo temperature and parameters b and λ:

$$T_K = D\, e^{1/J\rho}, \qquad b = \pi^2 S(S+1), \quad \lambda = \ln(T/T_K). \tag{3.1}$$

For Mn, take $S = \frac{5}{2}$ so that $b = 86.38$. The density of states ρ may be extracted from host specific-heat data[7] since it is really required at the Fermi surface. $\rho = 0.147\ \text{eV}^{-1}\,\text{spin}^{-1}\,\text{atom}^{-1}$ for Cu, 0.137 for Ag, and 0.155 for Au.

A. Magnetic Susceptibility

The contribution per impurity to the susceptibility can be expressed entirely as a renormalization of the Curie constant, which in the approximation of leading $\ln T$ terms only depends only on $\ln T$.[8] Thus

$$\Delta\chi = [g^2\mu_B^2 S(S+1)/3kT]\ C(T), \qquad C(T) = 1 - 5\lambda^{-1}, \tag{3.2}$$

which ought to give the $\ln T$ slope correctly but ought not (and in fact will not!) reproduce the constant term.

Figure 1 shows data by Hurd[9] on Cu : Mn, Ag : Mn, and Au : Mn presented in terms of the Curie constant $C(T)$ plotted against $\ln T$. We attempt to fit the slope $dC/d\lambda$ to the theoretical expression $5\lambda^{-2}$. Because of the considerable scatter in these results, and because the slope will change very little over a decade if the Kondo temperature is small, the experimental points may be fitted to a straight line by the method of least squares. By equating its slope to the slope of the theoretical curve in the *middle* of the experimental temperature range, we obtain

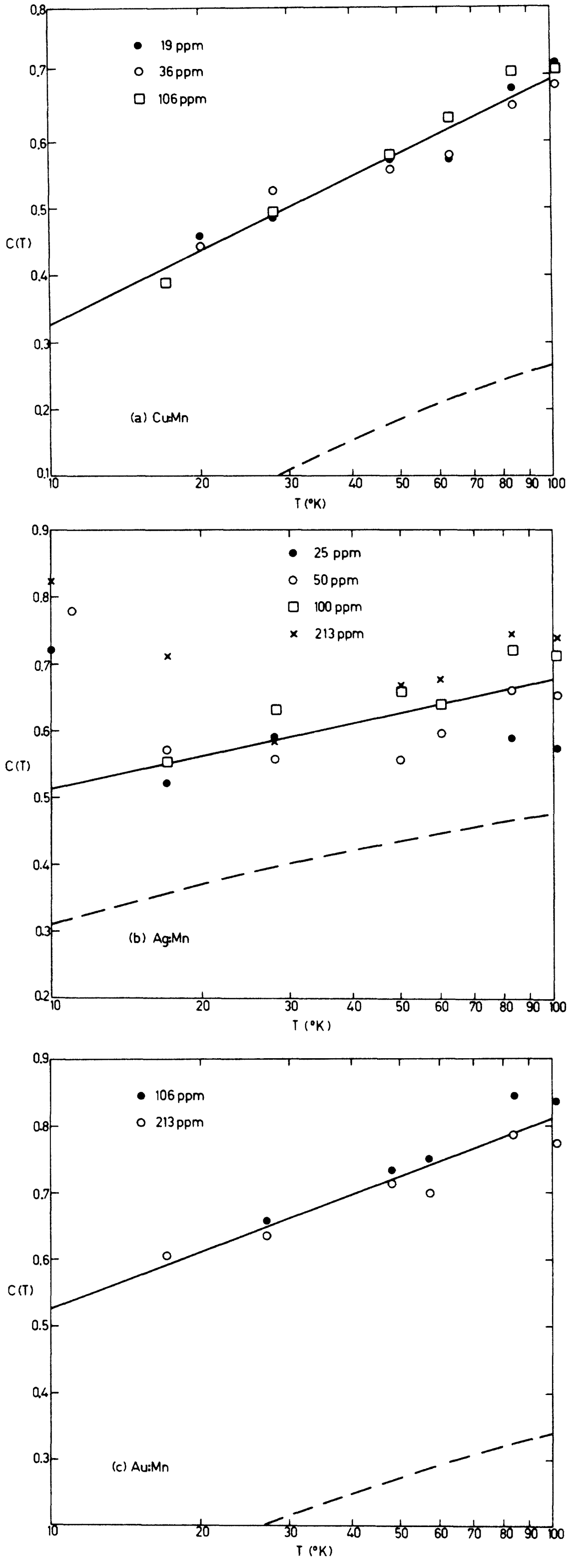

FIG. 1. Normalized Curie constant for Cu : Mn, Ag : Mn, and Au : Mn against ln$T$, from results of Hurd. Also shown is the best straight-line fit and the theoretical curve (dashed) with the same slope at 30 °K.

estimates for T_K and hence J. These are given in Table I.

Some discussion of errors is very necessary. First, the error involved in neglecting the temperature dependence of the slope can be assessed by plotting (3. 2) using the values for T_K obtained above, giving the dashed curves in Fig. 1. The curvatures are seen to be within experimental error in each case. In fact, the predicted curvature is very difficult to detect experimentally because T/T_K must be large (say, > 100) for (3. 2) to be useful and the curvature is a decreasing function of T/T_K. Second, the slope of the "line of best fit" can probably be varied by 20% in either direction for Cu : Mn and Au : Mn, and somewhat more for Ag : Mn. This produces errors of 50–100% in the Kondo temperature.[10] But an equally serious error arises from the somewhat arbitrary way in which (3. 2) is matched to the straight line. By varying the temperature at which the slopes were matched over a factor of 2 in each direction, the estimated T_K will vary proportionally without degrading the fitting of the slopes very much. To be conservative, the uncertainty in T_K is probably a factor of 10 in all, but even this means that J is accurate to about ± 15%.[10]

We should mention that Hurd's data above 100 °K have not been used because the density of states there is expected to change appreciably with temperature due to thermal expansion.[9] Some of his results from higher-concentration samples have been omitted because of obvious magnetic ordering, and the four points in the top left-hand corner of Fig. 1(b) were dropped from the least-squares program for the same reason. The Ag : Mn data are the least satisfactory of the three, and results from high-concentration samples were accepted in this case to get more points.

B. Specific Heat

According to a conjecture of Kondo's,[8,11] the impurity specific heat per impurity is

$$\Delta C = 5kb\lambda^{-4}, \qquad (3.3)$$

where the factor of 5 for d-orbital degeneracy has been supplied and k is Boltzmann's constant. This is supported by the calculations of Müller-Hartmann and Zittartz[12] based on Nagaoka's truncation scheme.

The only quantitative experiment showing ln$T$ behavior above T_K for dilute (noble-metal) : Mn alloys is on Cu : Mn by Tu and Heeger.[13] Their results are well represented by straight-line plots of ΔC against ln$T$, which are shown in Fig. 2 after normalizing to one impurity. The two lines for their two samples are separated outside experimental error (this may be checked from their publication), indicating that impurity interactions are important and lower concentrations necessary. But it appears

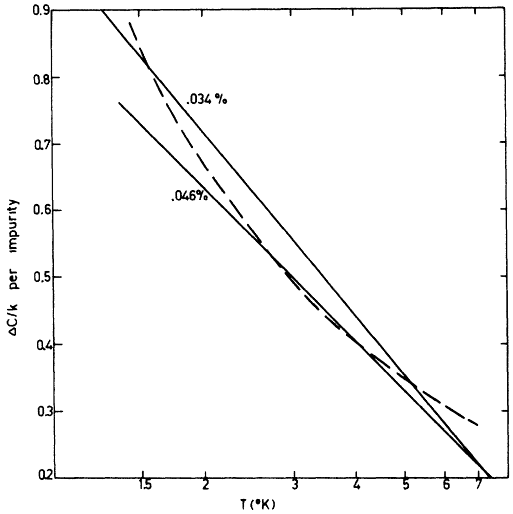

FIG. 2. Specific heat per impurity (in units of k) of two Cu : Mn samples, after Tu and Heeger, also the theoretical curves (3.3) (dashed) whose slope is the average of the experimental ones at 3 °K.

that the impurity specific heat is linear in $\ln T$, whereas the theoretical curve obtained by fitting the slope to $-20b\lambda^{-5}$ at 3 °K is definitely nonlinear. In spite of this, a reasonable value for J emerges (see Table I).

Assuming that the observed linearity is genuine, it is hard to believe that it could be a fortuitous combination of the theoretical curve (3.3) and interaction effects which would lower ΔC at lower temperatures. It seems more likely that (3.3) requires modification by nonleading $\ln T$ terms even at these high temperatures; it does after all diverge very rapidly at $T \to T_K$. So in view of this the value of J obtained here cannot be taken too seriously.

C. Residual Resistivity

The standard expression for the residual resistance per impurity[14] $\Delta\rho$ is given by

$$\Delta\rho/\rho_u = \tfrac{1}{4} b\, \lambda^{-2}\,,$$

where

$$\rho_u = 20\pi\hbar/k_F e^2 \tag{3.4}$$

is the d-wave unitarity limit for a monovalent metal, viz., 19.0 $\mu\Omega$cm/at.% for Cu and 21.4 for Ag and Au.

Most experimental results in the literature are unusable because the impurity concentration is too high and/or the host contribution is unknown and has not been subtracted. We use Gainon and Heeger' s result[15] on a single 110-ppm Cu : Mn sample, work by Malm and Woods[16] on Ag : Mn, and Loram, Ford, and Whall on Au : Mn.[17]

Now all these resistivity results plotted against $\ln T$ yield straight lines, shown in Fig. 3. T_K and J are obtained as before by fitting the slope at 3 °K to $-\frac{1}{2} b\, \lambda^{-3}$, and these estimates are then used to draw the theoretical curves (dashed) from (3.4). These display no curvature whatsoever, but that is because the Kondo temperatures obtained here are absurdly small, in fact quite unbelievable. So in spite of a perfect (because trivial) fit to the slope of each experimental curve, the $\ln T$ variation in electrical resistance cannot be described by the current high-temperature theoretical expressions. Nor does the nonperturbative result[18]

$$\Delta\rho/\rho_u = \tfrac{1}{2}[1 - \lambda/(\lambda^2 + b)^{1/2}] \tag{3.5}$$

represent much improvement here; applied to the Cu : Mn result it yields $T_K = 74\times10^{-6}$ °K instead of 2.2×10^{-6} °K. To fit the measured slope at 3 °K, λ^2 must be greater than b and then (3.5) is little different from (3.4). Experimental curves are often fitted to (3.5) by allowing b to be an adjustable parameter, but $b = \pi^2 S(S+1)$ and it is hard to conceive of the Mn spin being anything but $\frac{5}{2}$.

IV. DISCUSSION

Table I shows that values of T_K and J derived from the susceptibility and specific heat are in reasonable agreement considering the errors involved (a factor of 10 for T_K and ± 10–15% for J). Values for different hosts are not expected to differ much, and this allows a wider comparison to be made. Moreover, the Kondo temperatures are consistent with empirical T_K's obtained from *low*-temperature experiments.[19] But values derived from the resistivity are clearly much too low and in this sense the resistivity, which is a transport property, is anomalous.

It seems likely that this disagreement arises from the calculation of the resistivity from the s-d exchange model rather than from the model itself. Success with the susceptibility and specific heat, which are both equilibrium properties, was achieved by separating leading $\ln T$ terms from all others and this separation can be maintained at every stage of the perturbation calculation of the free energy. But

TABLE I. Values of the Kondo temperature (T_K) and s-d exchange integral (J) estimated from susceptibility, specific heat, and resistivity at the temperatures shown. Optical experiments on Ag : Mn indicate $J = -0.45$ eV and hence $T_K = 0.0023$ °K.

		$\Delta\chi$ (30 °K)	ΔC (3 °K)	$\Delta\rho$ (3 °K)
Cu : Mn	T_K (°K)	0.11	0.013	2.2×10^{-6}
	J (eV)	−0.55	−0.47	−0.29
Ag : Mn	T_K (°K)	0.0071		1.7×10^{-9}
	J (eV)	−0.48		−0.24
Au : Mn	T_K (°K)	0.052		1.2×10^{-11}
	J (eV)	−0.49		−0.17

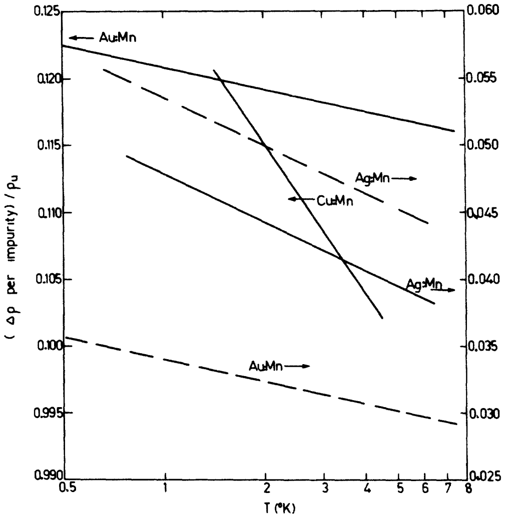

FIG. 3. Residual resistivity per impurity, expressed in terms of unitarity limits, for Cu : Mn (Gainon and Heeger), Ag : Mn (Malm and Woods), and Au : Mn (Loram, Ford, and Whall). Where absolute values were known, the dashed curves calculated from (3.4) are also displayed.

transport properties require the t matrix, which is a complex quantity; the leading $\ln T$ terms occur in the real part of the denominator of t but the imaginary part, which is often neglected, lies beyond logarithmic accuracy. Thus no such separation of leading $\ln T$ terms and others is possible. This may be why (3.4) fits neither the constant component nor the $\ln T$ slope of the resistivity. These remarks also apply to the nonperturbative treatments since they were checked with low orders of perturbation theory only to logarithmic accuracy. For example, Suhl's theory leads to Eq. (3.5) which also gives too small a J when fitted to the observed $\ln T$ slope. The linearized vertex equation proposed by Duke and Silverstein[20] is presumably worse since the imaginary part of the vertex function disagrees with perturbation theory in second order, which is not true of the nonlinear equations.

However, other explanations can also be countenanced. Gainon and Heeger[15] added heavy impurities to Cu : Mn which washed out the $\ln T$ slope in resistivity at lower temperatures and suggest that this reduces the memory of the Mn spin. Thus intrinsic spin fluctuations in the absence of heavy impurities might have a similar effect, but these are outside the framework of the s-d exchange model. An additional relaxation mechanism is required to reduce $\ln T$ slopes in transport properties but not equilibrium properties. Finally, one should not forget that impurity-impurity interactions tend to reduce the $\ln T$ slope[21] and the concentration dependence of the experimental values used here is not adequately established.

Can our analysis be applied to other Kondo systems? Optical experiments, by revealing either one or two virtual bound states, will in principle dictate the use of the spin fluctuation or localized spin models, respectively; e.g., recent work by Beaglehole[22] supports the spin fluctuation picture for Al : Mn. But if the localized spin picture is indicated, a difficulty arises for non-S-state ions; why is there no orbital moment? Crystal fields are much too weak to split the virtual bound states so one is forced to say that orbital fluctuation times are too short to permit a well-defined orbital moment. Less convincingly, one could imagine an orbital moment reduced by Kondo compensation with a very high orbital Kondo temperature because orbit-orbit plus orbit-orbit-spin-spin interactions can be shown to arise from the Schrieffer-Wolff transformation in this case.

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[*] Present address: School of Mathematical and Physical Sciences, University of Sussex, Brighton, United Kingdom.

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Temperature Dependence of the Electrical Resistivity of Dilute Ferromagnetic Alloys

D. L. Mills*
Theoretical Physics Division, AERE Harwell, Didcot, Berks, England

and

A. Fert and I. A. Campbell
Physique des Solides, Faculté des Sciences, 91-Orsay, France

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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 *Ni*Mn 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 *Pd*Fe and *Pd*Mn 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.[1]

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 ρ of these systems,[2] 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[3] 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 ρ is similar to that observed in ferromagnetic Pd alloys.[4]

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 resistivity 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 ρ 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 ρ observed in the ferromagnetic alloys mentioned above.