Resistivity from magnetic impurity pairs in metallic alloys

Fu-sui Liu,* W. A. Roshen, and J. Ruvalds
Department of Physics, University of Virginia, Charlottesville, Virginia 22901
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The spin-flip contributions to the electron scattering by a pair of magnetic impurities is found to exhibit a T^{-1} behavior in the temperature variation of the cross section. Our calculations provide an explanation for the sharp decrease in the low-temperature resistivities of several alloys in the intermediate concentration regime where the Kondo effect is inoperative and the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling between impurities is substantial. Accordingly, the size and magnitude of the impurity pair contribution to the resistivity is a senstive function of the impurity concentration and the electronic structure of the host metal. Our computations agree with the data of $CuMn_x$, $AgMn_x$, $AuMn_x$, and $AuCr_x$, and they predict the observed variation of the resistivity maximum with impurity concentration using a standard RKKY coupling between impurities.

I. INTRODUCTION

Dilute magnetic impurities yield a resistivity minimum in systems such as $CuMn_x$ which have been extensively studied experimentally.[1] The anomalous electron scattering responsible for the minimum was associated with the magnetic character of the impurity using susceptibility data, and the physical origin of the $\ln T$ resistivity divergence was first identified by Kondo[2] as a spin-flip mechanism caused by individual impurities. His result appears in the third-order perturbation theory and correlates extremely well with a wide range of experiments. In particular, his prediction for the resistivity ($\rho_K \propto \ln T$), and the minimum temperature $T_0 \propto x^{1/5}$ is very accurately confirmed in many cases. Also the disappearance of the Kondo effect at higher impurity concentrations, typically $x > 1\%$, is well established.[3] The Kondo calculations have stimulated a great deal of theoretical activity, culminating in recent exact solutions of the Kondo Hamiltonian over the entire temperature range of interest.[4,5]

The purpose of the present work is to demonstrate that coupled pairs of magnetic impurities may yield a divergent resistivity contribution of the form $\rho \propto x^2/T$ whose sign is determined by the coupling between impurities and the electronic structure of the conduction electrons. Unlike the Kondo scattering, the impurity pair contribution is enhanced at higher concentrations of the order $x \simeq 1-10\,\%$. This intermediate regime is notable for strong Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions among impurities and a lack of long-range magnetic order. Hence these metallic alloys provide an ideal testing ground for our mechanism, which is based on the sequential spin-flip scattering of an electron by two nearby impurities. By contrast, the spin-conserved scattering gives only a constant contribution by analogy to potential scattering.

The formalism and explicit calculation of the pair resistivity contribution is given in Sec. II, and the results are applied to various alloys in Sec. III. We find good agreement with the resistivity anomalies in $CuMn_x$, $AgMn_x$, $AuCr_x$, and other alloys and the conclusions of these results are summarized in Sec. IV.

II. FORMAL DEVELOPMENT

We consider the interaction of the electrons with the magnetic impurities via the standard *s-d* exchange represented by a coupling constant J according to the Hamiltonian

$$H = H_0 + H_1 \ , \tag{1}$$

where the unperturbed part is given by

$$H_0 = \sum_{k,\alpha} \varepsilon_k C^{\dagger}_{k\alpha} C_{k\alpha} - U \sum_{i \neq j} S^z_i S^z_j \ , \tag{2}$$

where ε_k is the electron energy and U represents the interaction between two magnetic impurities with spins S_i and S_j, respectively, situated at sites i and j. The electron creation and destruction operators are $C^{\dagger}_{k\alpha}$ and $C_{k\alpha}$, where α is the spin index and k is the momentum. The spin-conserving (z-component) part of the impurity-impurity coupling will not influence the spin states of the conduction electrons and thus is conveniently included in H_0. Potential scattering terms are not considered here, since the key features relate to spin-flip scattering of the conduction electrons. Hence the interaction Hamiltonian may be expressed as

$$H_1 = -J \sum_j \mathbf{S}_j \cdot \boldsymbol{\sigma} - \frac{U}{2} \sum_{\substack{i,j \\ (i \neq j)}} (S_i^+ S_j^- + S_i^- S_j^+) \ , \tag{2'}$$

where σ is the conduction electron spin and the raising (lowering) operators for the impurity spin are

$$S_j^{\pm} = S_j^x \pm i S_j^y \ . \tag{3}$$

In the event that U is determined by the RKKY indirect exchange of conduction electrons, it may be expressed directly in terms of the exchange coupling J. For a three-dimensional gas of free electrons the standard result is

$$U_{\mathrm{RKKY}}=\frac{9\pi J^2}{E_F}L(y)\ , \tag{4}$$

where

$$L(y)=\frac{y\cos y-\sin y}{y^4}\ , \tag{5}$$

with $y=2k_F\mid \mathbf{R}_i-\mathbf{R}_j\mid$, and $E_F=k_F^2/2m$ is the Fermi energy.

The scattering perturbation series for an electron described by the above Hamiltonian exhibits two fundamental difficulties. First of all the terms of order J^3 are divergent at low temperatures as noted by Kondo. Secondly, the impurity spin operators $\mathbf{S}_i$ do not obey simple boson or fermion commutation rules so that the traditional perturbation expansions cannot be applied directly. Thus it is convenient to use the "pseudofermion" technique invented by Abrikosov, which was designed to circumvent the above difficulties. Abrikosov introduces operators $a_{j\beta},a_{j\beta}^{\dagger}$ for a fermion field, and writes the spin operator $\mathbf{S}_j$ as

$$\mathbf{S}_j=\sum_{\beta,\beta'}a_{j\beta}^{\dagger}\mathbf{S}_{\beta\beta'}a_{j\beta'} \tag{6}$$

where $\mathbf{S}_{\beta\beta'}=\langle\beta\mid\mathbf{S}\mid\beta'\rangle$ are the standard matrix elements of the spin matrices. Each index β and β' assumes values corresponding to $2S+1$ Zeemann states. The operators $a_{j\beta},a_{j\beta}^{\dagger}$ obey the usual anticommutation relation for fermions,

$$\begin{aligned}&\{a_{i\beta}^{\dagger},a_{j\beta'}\}=\delta_{ij}\delta_{\beta\beta'}\ ,\\&\{a_{i\beta},a_{j\beta'}\}=0=\{a_{i\beta}^{\dagger},a_{j\beta'}^{\dagger}\}\ ,\end{aligned} \tag{7}$$

and thus they are suitable for standard perturbation expansions and diagrammatic analysis. The important difficulty with the pseudofermion method is the presence of spurious states with unphysical multiple occupation. However, these states are readily removed by assigning them an energy $\lambda\gg T$ and then normalizing the results to the probability of single occupation $(2S+1)\exp(-\lambda/T)$ with $\lambda\rightarrow\infty$.

Thus, each impurity state has an associated pseudofermion Green's function $\mathcal{G}$ (Ref. 6)

$$\mathcal{G}=\frac{1}{i\omega_m-\lambda},\quad \omega_m=(2m+1)\pi T \tag{8}$$

where the impurity energy is constant as a result of its localization. We follow the usual convention with solid lines representing the electron propagators and broken lines for the pseudofermion Green's function. Expressing the Hamiltonian in terms of these pseudofermion operators $a_{j\beta}^{\dagger}(a_{j\beta})$, we rewrite Eq. (2) as

$$\begin{aligned}H_1=&-J\sum_{\substack{\mathbf{k},\mathbf{k}',j,\\ \alpha,\alpha',\beta,\beta'}}e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_j}(\boldsymbol{\sigma}_{\alpha'\alpha}\cdot\mathbf{S}_{\beta',\beta})a_{j\beta'}^{\dagger}a_{j\beta}C_{\mathbf{k}'\alpha'}^{\dagger}C_{\mathbf{k}\alpha}\\&-\frac{U}{2}\sum_{\substack{i,j\\(i\neq j)}}(S_i^{+}S_j^{-}+S_i^{-}S_j^{+})\ ,\end{aligned} \tag{9}$$

where $\boldsymbol{\sigma}$ is the Pauli spin matrix for the conduction electrons, and the corresponding matrix for the impurity spin is $\frac{1}{2}\mathbf{S}$.

The lowest-order Born approximation for the electron scattering by a single impurity yields a self-energy Σ_{AG} shown in Fig. 1 and first calculated by Abrikosov and Gorkov (AG).[6] Their work yields a temperature-independent relaxation time τ_{AG} defined by

$$\frac{1}{\tau_{\mathrm{AG}}}=-\mathrm{Im}\sum_{\mathrm{AG}}=2\pi xN(0)J^2S(S+1)\ , \tag{10}$$

where $N(0)$ is the density of conduction states at the Fermi level. The quantities in Eq. (10) appear also in the higher-order terms and are subject to independent determination from measurements such as the depression of the superconducting transition temperature according to the Abrikosov-Gorkov theory.

In the intermediate concentration regime under consideration, the Kondo effect is suppressed by RKKY interactions and thus is not discussed here. A historical perspective on earlier work on the resistivity associated with dilute impurities may be found in Ref. 7, where the limiting criteria for the Kondo effect are reviewed. In addition, references to earlier work on coupled magnetic impurities may be found in Ref. 7. However, we remark that multiple scattering from a single impurity spin, which is responsible for the Kondo effect, can be successfully treated in the pseudofermion formalism. This was shown by Abrikosov in Ref. 6 where he obtains a logarithmic divergence in agreement with the results of Kondo[2] for order J^3. Higher-order contributions are also divergent and have only recently been calculated by exact methods.[4,5]

It is convenient to express our results in terms of the indirect exchange coupling (RKKY) between magnetic

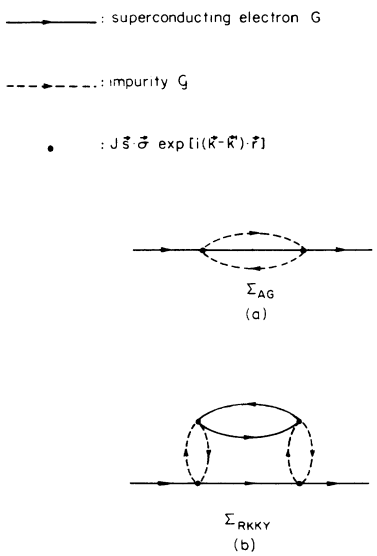

FIG. 1. Self-energy diagrams for an electron with a propagator G scattering from a magnetic impurity state represented by $\mathcal{G}$. The s-d exchange interaction J leads to the Born approximation scattering treated by Abrikosov-Gorkov and represented by Σ_{AG}. The next-order single-impurity Kondo diagrams are omitted for reasons expressed in the text.

impurities, primarily because this limits the need for adjustable parameters in the theory. Hence, we can express the final results in terms of the *s-d* exchange coupling J and the conventional parameters of the free-electron model. The form of the well-known RKKY interaction is given in Eqs. (4) and (5). It should be noted that the RKKY coupling is a sensitive function of the Fermi surface and becomes longer range at lowered dimensionalities. Furthermore, the limited mean free path of electrons in spin-glass alloys tends to significantly decrease the range of the RKKY interaction and further complicate the concentration variation of the coupling between impurities. Hence our analysis of data using Eqs. (4) and (5) for U are realistic only in a qualitative sense. However, our analysis may shed light on modifications of the RKKY coupling in representative compounds. Although the magnitude of the RKKY scattering contribution is proportional to U, the temperature variation of the electron cross section is a fundamental consequence of the spin dynamics and therefore will be present for alternate models of the impurity interactions which allow for elastic spin-flip scattering of the conduction electrons.

The physics of the spin-flip scattering of an electron by two nearby impurities can be visualized as in Fig. 1, where the corresponding electron self-energy is Σ_{RKKY}. This elastic scattering process yields a T^{-1} divergence in Σ_{RKKY}, which we now derive explicitly.

Following the usual rules for evaluating diagrammatic expansions of the scattering amplitude[8] we obtain the impurity pair-induced electron self-energy

$$\Sigma_{\mathrm{RKKY}}(\mathbf{p},\omega)=-J^4\sum_{j,l}T^2\sum_{\omega_1,\omega_2}A_s M^2(\omega-\omega_2)\times B(\mathbf{R}_j-\mathbf{R}_l,\omega_1,\omega)\ , \tag{11}$$

where A_s represents the spin summation result

$$A_s=\frac{1}{(2s+1)^2}\sum_{\substack{\beta,\beta',\alpha,\alpha',\\ \beta_1,\beta_1',\alpha_1,\alpha_1'}}(\mathbf{S}^j_{\beta'\beta}\cdot\boldsymbol{\sigma}_{\alpha'\alpha})(\mathbf{S}^j_{\beta'\beta}\cdot\boldsymbol{\sigma}_{\alpha_1'\alpha_1})(\mathbf{S}^l_{\beta_1'\beta_1}\cdot\boldsymbol{\sigma}_{\alpha,\alpha_1'})(\mathbf{S}^l_{\beta_1,\beta_1'}\cdot\boldsymbol{\sigma}_{\alpha\alpha'})\ , \tag{12}$$

which reduces to

$$A_s=\frac{[S(S+1)]^2}{24}\ . \tag{13}$$

The frequency summations lead to the factor

$$M(\omega-\omega_2)=\lim_{(\lambda/T)\to 0}e^{\lambda/T}T\times\sum_{\omega_1}\frac{1}{(i\omega_1-\lambda)}\frac{1}{i(\omega_1+\omega-\omega_2)-\lambda}\ , \tag{14}$$

this sum vanishes for $\omega\neq\omega_2$, and leads to a divergent temperature variation in the elastic case, i.e.

$$M_{\mathrm{imp}}(0)=\lim_{(\lambda/T)\to\infty}e^{\lambda/T}T\sum_{\omega_1}\left[\frac{1}{i\omega_1-\lambda}\right]^2=\lim_{(\lambda/T)\to\infty}e^{\lambda/T}\frac{\partial}{\partial\lambda}\left[T\sum_{\omega_1}\frac{1}{(i\omega_1-\lambda)}\right]\ , \tag{15}$$

where the sum over ω_1 is performed by contour integration; this introduces the Fermi function $n(\lambda)$ and yields

$$M_{\mathrm{imp}}=\lim_{(\lambda/T)\to\infty}e^{\lambda/T}\frac{\partial}{\partial\lambda}[n(\lambda)]=\lim_{(\lambda/T)\to\infty}e^{\lambda/T}\frac{-e^{\lambda}/T}{(e^{\lambda/T}+1)^2}\frac{1}{T}\ , \tag{16}$$

which gives the final result

$$M_{\mathrm{imp}}=-\frac{1}{T}\ . \tag{17}$$

This surprising temperature variation is a consequence of the spin-flip dynamics and was noticed previously in a preliminary report by Liu *et al.*[9] Finally, we calculate the frequency and momentum summations which involve the RKKY coupling. This factor is of the form

$$B(\mathbf{p},\boldsymbol{\Delta})=\frac{T}{(2\pi)^9}\sum_{\omega}d^3p_1d^3p_2d^3q e^{i(\mathbf{p}-\mathbf{p}_2+\mathbf{q})\cdot\boldsymbol{\Delta}}\times\left[\frac{1}{i\omega-\varepsilon_{p_2}}\frac{1}{i(2\omega-\omega')-\varepsilon_{\mathbf{p}_2-\mathbf{q}}}\times\frac{1}{i\omega''-\varepsilon_{p_1}}\right]\ , \tag{18}$$

where $\boldsymbol{\Delta}=\mathbf{R}_j-\mathbf{R}_l$, and we have set $\omega=\omega''$ in view of the fact that only the elastic scattering yields a finite contribution to the self-energy as shown above. Performing the frequency sum in Eq. (18) transforms $B(\mathbf{p},\boldsymbol{\Delta})$ into the product of two factors, the first being simply the RKKY coupling U defined in Eqs. (4) and (5), and the second factor involves a momentum average over the impurity phase factor: Thus, combining the results of Eqs. (8)–(13), we obtain the final result for the imaginary part of the self-energy

$$\mathrm{Im}\,\Sigma_{\mathrm{RKKY}}=\frac{2\pi N(0)J^2S^2(S+1)^2}{3T}\sum_{j,l}U(\Delta)\frac{\sin^2(k_F\Delta)}{(k_F\Delta)^2}\ , \tag{19}$$

where $N(0)$ is the density of states at the Fermi energy. This result is the electron lifetime which enters in calculations of the superconducting order parameter and electron spin susceptibility.[10] It is interesting to note that the sign of $\mathrm{Im}\Sigma_{\mathrm{RKKY}}$ is determined by the sign of the RKKY interaction.

Transport properties such as the resistivity require an additional weighting factor for large-angle scattering which is familiar in the Boltzmann formulation of the conductivity, but nevertheless requires a subtle

modification involving vertex corrections. The physical origins of the transport relaxation time τ, as well as the formal diagrammatic derivation are elucidated by Mahan (Ref. 8, Chap. 7). The net result for the transport relaxation time is the introduction of an additional factor $(1-\cos\theta)$ in the momentum summation of Eq. (18), where $\cos\theta=\mathbf{k}\cdot\mathbf{k}'$ defines the scattering angle for an electron with incoming momentum $\mathbf{k}$ being scattered to a state $\mathbf{k}'$. Performing the required angular integration and impurity average over nearest-neighbor pairs separated by a distance Δ, yields the transport relaxation time

$$\frac{1}{\tau_{\mathrm{RKKY}}}=-2\pi N(E_F)x^2z\left[\frac{J}{N}\right]^2\frac{U}{T}I(2k_F\Delta)\ , \tag{20}$$

where z is the number of nearest-neighbor sites, x is the impurity concentration, and

$$I(y)=-\frac{\cos(y)}{y^2}+\frac{2\sin(y)}{y^3}+\frac{2\cos(y)}{y^4}-\frac{2}{y^4}\ . \tag{21}$$

It is interesting to note that the RKKY impurity pair resistivity contribution may have the same sign as the RKKY coupling U only in certain region as the phase factor $2k_F\Delta$. The correspondence between the transport relaxation time and U in the RKKY model is shown in Fig. 2, which demonstrates small regions of the phase $2k_F\Delta$, where U and τ_{RKKY} differ in sign: In most regions the net result is a negative relaxation time contribution.

Electronic band-structure details could readily change the electron scattering rate because the momentum sums in Eq. (18) are quite sensitive to deviations from the free-electron model used here. In particular, nearly parallel sections of the Fermi surface in momentum space may change the sign as well as the magnitude of τ_{RKKY}, so that applications of Eq. (20) to metals with tightly-bound electron orbitals are expected to have qualitative reliability.

The primary significance of the impurity pair contribution is the T^{-1} divergence evident in Eq. (20). At the lowest temperatures, higher-order graphs need to be included to satisfy the unitarity limit on the scattering, and the treatment of this limit should consider the Kondo contributions from a single impurity as well.

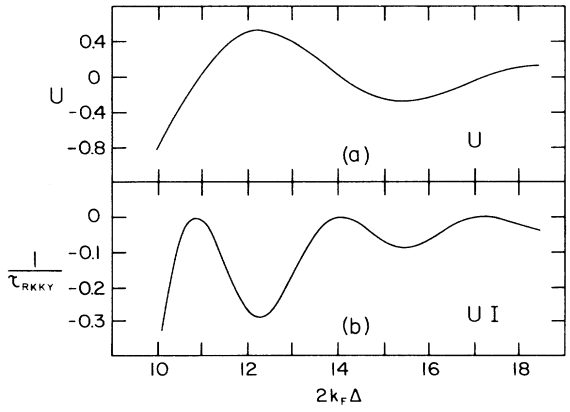

FIG. 2. Comparison of the RKKY coupling U with the electron scattering rate $(\tau_{\mathrm{RKKY}})^{-1}$ as a function of $2k_F\Delta$, where k_F is the Fermi momentum and Δ is the nearest-neighbor impurity separation. These quantities are given in Eqs. (4), (5), and (21)–(22); they are represented in arbitrary units.

A physical alternate derivation of the temperature variation of the electron scattering rate may be found by applying ordinary perturbation theory to the sequential spin-flip scattering of an electron: In each impurity site there is a divergent vertex of the form $(\varepsilon_k-\varepsilon_q)^{-1}$ arising from the elastic nature of the impurity scattering; then the intermediate electron state introduces a Fermi function $f_q(T)$ which leads to the final result

$$\frac{1}{\tau_{\mathrm{RKKY}}}\propto x^2J^4\sum_q\frac{f_q(T)}{(\varepsilon_k-\varepsilon_q)^2}\propto\frac{x^2J^4}{T}\ . \tag{22}$$

However, multiple scattering of an electron from a *single* impurity will yield a negligible result for the scattering when the limiting case of an RKKY "interaction" process of an impurity with itself is analyzed properly. Furthermore, spin-conserving processes will not yield transitions of the above form but rather will resemble ordinary potential scattering.

III. APPLICATIONS TO SPIN GLASSES

Resistivity data on a wide range of materials exhibits anomalous variations at low temperatures which may be attributed to the impurity pair mechanism of our theory. In a preliminary report[9] we had analyzed the resistance minimum observed in Fe_xTaSe_2 (Ref. 11) at $x=0.5\%$ and $x=5\%$. Our theory accounts for the temperature variation of the magnetic impurity scattering and gave the observed behavior of the temperature at the minimum, i.e., $T_m\propto x^{1/3}$, providing that the photon contribution to the resistivity was taken as the conventional T^5 form.[9] However, several complications in these types of layered alloys require further experiments to distinguish our mechanism from the Kondo effect. The primary uncertainty arises from the nonmagnetic scattering which includes a temperature variation from charge-density-wave (CDW) contributions in the pure $TaSe_2$. At higher Fe content, $x\sim5\%$, the CDW is presumably suppressed, but the temperature variation of the phonon term remains in doubt. Thus further data on these systems would be most useful in understanding the role of impurity interactions as well as the possibility of an unexpected persistence of Kondo scattering at such high impurity concentrations. Thus we were stimulated to search for other systems where the Kondo scattering is not competitive. Many compounds which have been investigated thoroughly in the dilute Kondo regime provide ideal tests for our theory at higher concentrations.

We focus attention on those systems where the Kondo scattering is unequivocally suppressed for magnetic impurity concentrations exceeding fractions of a percent.

First we consider the typical example of $AuFe_x$. Dilute concentrations of iron in gold yield a classic Kondo effect which has been extensively studied.[12] The Kondo temperature is quite low, i.e., $T_K\cong0.2$ K, and the interactions among Fe impurities are strong enough to suppress the

Kondo effect already at very low concentrations $x \geq 0.2\%$.[12] The disappearance of the Kondo minimum at low impurity concentration is a feature common to most Kondo systems including $CuMn_x$, $AgMn_x$, and many others. Resistivity data[13] on Fe_xAu in the spin-glass regime exhibits an anomalous drop at low temperatures which is shown in Fig. 3 and analyzed below.

The host resistivity of pure Au has been subtracted in the definition of $\Delta\rho$ and hence the low-temperature data illustrate primarily the spin-dependent contributions of the iron impurities. Our theory provides a good description of the lower-temperature data in the region 25 K $< T <$ 150 K as seen in Fig. 3. At higher temperatures $T >$ 150 K there is an additional temperature variation which is not considered by our mechanism, but may be in part due to the changes in the phonon spectrum of Au caused by the substantial amounts of impurities. Our fits to the $AuFe_{0.02}$ data are done with a ferromagnetic RKKY coupling $U = -0.01$ eV which is consistent with our independent analysis[14] of the susceptibility data on this alloy. Using values of $\varepsilon_F = 5.5$ eV and $J = 5$ eV from NMR data, we find the optimum fit with $2k_F\Delta \cong 15$ for the case of $z = 12$ nearest-neighbor sites. In the gold host $2k_F = 2.4$ Å^{-1}, so the corresponding impurity separation is $\Delta \simeq 6$ Å. For the $Fe_{0.08}Au$ case a good fit is obtained using the same J and a slightly smaller value of $2k_F\Delta = 14$. Also, to make a more realistic comparison we have added a small phonon scattering contribution $\Delta\rho_{\rm ph} = 0.004T$ to the fit of the 8% concentration sample,

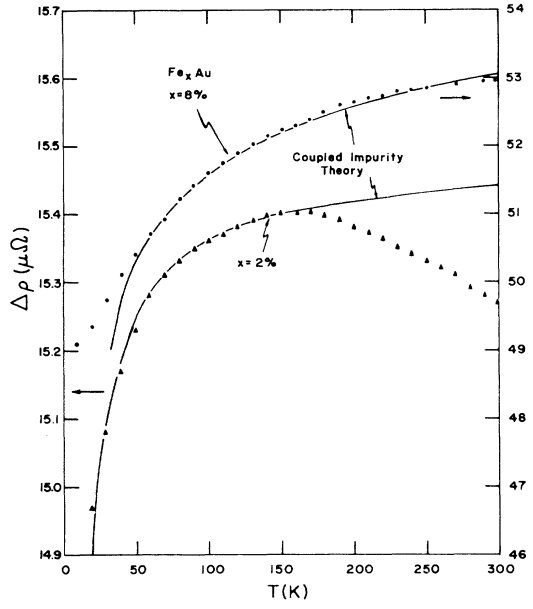

FIG. 3. Temperature dependence of the excess resistivity originating from Fe impurities in gold. The data of Ref. 13 display an anomalous drop in the range 25 K $< T <$ 150 K. The variations in $\Delta\rho$ at higher $T >$ 150 K may be in part associated with changes in the phonon spectrum of Au caused by alloying. Our theoretical curves provide a good description to the low-temperature region using parameters quoted in the text, and shown in the figure by solid curves.

and this term improves the fit to the high-T range as expected.

A systematic investigation of nondilute magnetic alloys is constructive in determining the universality, or lack thereof, of the impurity pair scattering, and it provides a useful guide to the trends in the coupling parameters as a function of concentration, host metal, and impurity characteristics. Since all of the parameters cannot be independently estimated we represent the resistivity in the form

$$\Delta\rho = a - bT - \frac{c}{T} , \tag{23}$$

where the modified phonon contribution $\Delta\rho_{\rm ph} \equiv -bT$ at higher temperatures seems to be a common feature of the alloys studies below. The data presented here are from Ref. 15, and in accord with their presentation, the resistivity of the pure noble metal host has been subtracted to excellent accuracy to give $\Delta\rho$ shown in the following figures. The impurity pair resistivity is simply related to the scattering time $\tau_{\rm RKKY}$ defined in Eq. (15) using the standard relation

$$\Delta\rho_{\rm pair} \equiv -\frac{c}{T} = \frac{m^*}{ne^2\tau_{\rm RKKY}} , \tag{24}$$

where m^* is the effective electron mass, e is the electron charge, and n denotes the electron density. Thus we proceed to analyze the resistivity data of $AgMn_x$, $CuMn_x$, $AuMn_x$, and $AuCr_x$. All of these are similar in that Kondo scattering has been well determined at dilute concentrations $x < 0.5\%$ and the suppression of the Kondo effect in the spin-glass cases considered here is unambiguously established.

The case of $AgMn_x$ is shown in Fig. 4 for three representative Mn concentrations. Our theory yields a good fit to the $AgMn_x$ data[15] in the temperature and concentration regime where it is expected to be valid: At

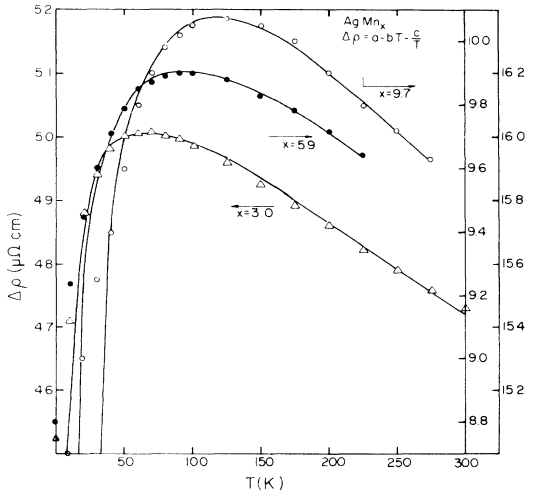

FIG. 4. The resistivity data of Ref. 14 for $AgMn_x$ are shown for three representative spin-glass concentrations as a function of temperature. Our theory yields the solid curves using the parameters listed in Table I and defined in Eq. (19).

high T ($\simeq$200–300 K) the gradual decline in $\Delta\rho$ is described by the term $-bT$ which is plausibly the modified phonon concentration; as the impurity atoms differ in valence and size from the host. The pair scattering term $-c/T$ yields a good description of the anomalous drop in the resistivity at temperatures below 100 K; the value of c which is proportional to the RKKY coupling was fixed to yield the correct position of the maximum in $\Delta\rho$, and hence the low-temperature variation of $\Delta\rho$ is determined. At the lowest temperatures the theory systematically falls below the data and it is interesting to note that the departure becomes more significant at higher concentrations. Such a trend may be attributed to higher-order scattering terms from a given pair of impurities or to larger impurity clusters. Another interesting possibility is the onset of short-range magnetic ordering which is characteristic of this spin glass as well as the analogous systems studied below. The parameters used in the fit are given in Table I.

A common feature in the $AgMn_x$ data, as well as the other compounds considered below is the anomalous resistivity maximum observed at a temperature T_M. This maximum provides an additional check on our theory because its concentration dependence can be predicted directly from Eq. (23) by examining the maximal criterion

$$\left.\frac{\partial(\Delta\rho)}{\partial T}\right|_{T=T_M} = 0 = -b + \frac{c}{T_M^2} ,$$

which yields

$$T_M = \left[\frac{c}{b}\right]^{1/2} \propto x\left[\frac{UI}{B}\right]^{1/2} . \tag{25}$$

Thus, providing that the concentration dependence of the phonon factor B and the RKKY coupling U is relatively smooth (or negligible) we expect the maximal temperature T_M to scale roughly linearly with magnetic impurity concentration. We shall see that this prediction in fact is reasonably supported by the data in our concluding analysis at the end of this section.

Other spin-glass alloys which exhibit the anomalous drop in resistivity include $CuMn_x$, $AuMn_x$, and $AuCr_x$: their resistivity data are examined in Figs. 5–7 using the parameters listed in Table I. With the exception of quantitative variations in their parameters, these resistivities are analogous to the case of $AgMn_x$ shown in Fig. 4 and discussed above in that they appear to have significant variations in the phonon spectrum upon alloying, and the impurity pair term of Eqs. (20)–(24) yields good agreement with the general structure of the data. The likely contributions of higher-order scattering become more important at higher concentrations, although they must be considered in all cases at sufficiently low temperatures.

Our analysis of electron scattering from impurity pairs neglects the thermodynamics distribution of spin-pair states. Hence the results should be valid for temperatures in excess of the RKKY coupling strength, and deviations from the data are expected as k_BT approaches U_{RKKY}. This situation is evident in all of the examples presented here in view of the estimated RKKY coupling given in Table II. For $AgMn_x$, $CuMn_x$, and $AuMn_x$ the fits start to deviate near $T \sim U_{RKKY} \sim 20$ K as expected, but surprisingly better fits are evident for $AuFe_x$ and $AuCn_x$ at the lowest concentrations. Including the thermal population of spin-pair singlet and triplet states would yield an additional smoother T variation as found by Brailsford and Overhauser, these exhibit the familiar exponential variation of the form $\exp(-U/T)$.[16]

Comparison of the parameters for the magnetic scattering for the spin-glass cases yields further insight into the

TABLE I. Parameters used in the fitting of Eq. (19) to the spin-glass resistivities of Ref. 15 shown in Figs. 4–7. The modified phonon scattering term is $\Delta\rho = bT$, a is the constant residual resistivity, and the strength of the magnetic impurity pair contribution is $\Delta\rho = -c/T$, where c is proportional to the RKKY coupling.

Material	x	a	b	c
$AgMn_x$	3.0	5.2	0.0015	6.0
	5.9	10.5	0.0032	26.0
	9.7	17.5	0.0049	65.0
$CuMn_x$	1.6	5.28	0.0018	4.0
	2.7	8.73	0.0027	10.0
	6.3	19.6	0.005	85.0
$AuMn_x$	1.5	3.98	0.0005	3.0
	2.8	6.88	0.001	4.5
	7.7	19.6	0.0035	55.0
$AuCr_x$	0.9	4.87	0.0029	3.5
	1.5	6.62	0.0038	7.0
	3.3	15.6	0.006	40.0
$AuFe_x$	2.0	15.5	0.0	12.0

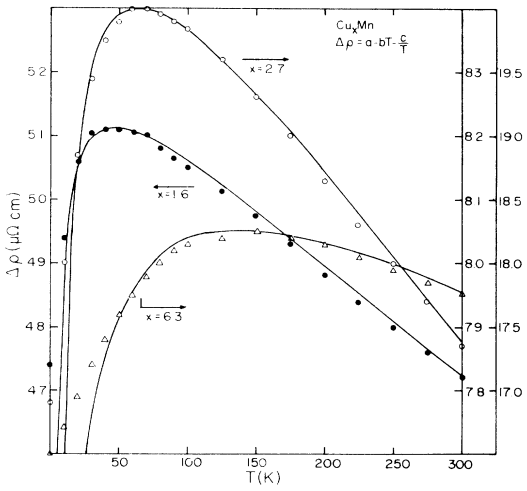

FIG. 5. The resistivity data of Ref. 15 for $CuMn_x$ at three concentrations is shown as a function of temperature. Our theory yields the solid curves using the parameters defined by Eq. (19) and listed in Table I.

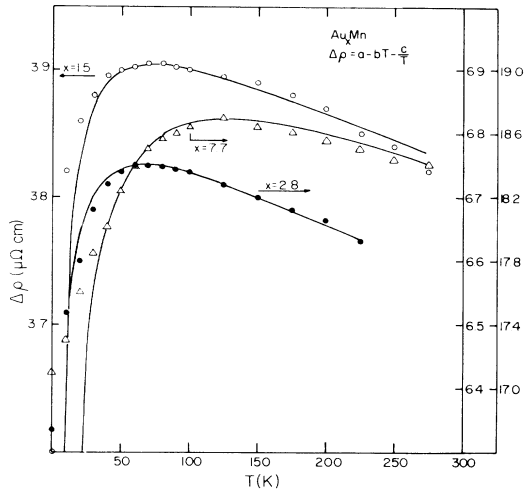

FIG. 6. The resistivity data of Ref. 15 for $AuMn_x$ at three concentrations is shown as a function of temperature. Our theory yields the solid curves using the parameters defined in Eq. (19) and listed in Table I.

RKKY mechanisms as shown in Fig. 8. At low concentrations a quadratic concentration variation of the parameter c would be expected from the lowest-order term [see Eqs. (20) and (22)] of the pair scattering, and such a behavior appears compatible with the data. At higher concentrations the pair scattering rises more slowly than x^2 as seen in Fig. 8 for the cases $AgMn_x$, $CuMn_x$, $AuMn_x$, and $AuCr_x$: Possible explanations for the slower dependence include higher-order RKKY processes or impurity cluster contributions beyond the nearest-neighbor approximation invoked here. Moreover, at the higher impurity contents, changes in the electronic band structure cannot be ruled out, and these would surely influence the range

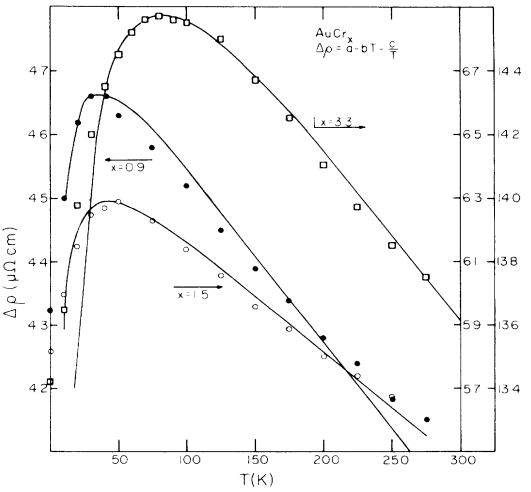

FIG. 7. The resistivity data from Ref. 15 for $AuCr_x$ at three concentrations are plotted as a function of temperature. Our theory yields the solid curves using the parameters defined in Eq. (19) and listed in Table I.

TABLE II. List of parameters for the alloys under consideration: the Fermi energy E_F is taken from the host metal values in Ref. 15; the exchange coupling J is from Ref. 3; the spin value for Fe in Au is from Ref. 18, whereas the other alloy estimates are from Ref. 3; the RKKY coupling parameter U_{RKKY} is estimated using Eq. (9) and the parameters listed below.

Material	E_F (eV)	J (eV)	S	U_{RKKY}/k_B (K)
$AgMn_{0.03}$	5.49	2.5	$\frac{5}{2}$	15.3
$CuMn_{0.016}$	7.00	2.5	2	19.7
$AuMn_{0.015}$	5.53	2.5	$\frac{5}{2}$	17.9
$AuFe_{0.02}$	5.53	5	$\frac{3}{2}$	44
$AuCr_{0.009}$	5.53	4.5	$\frac{3}{2}$	47(63)

and strength of the RKKY scattering.

Finally we test the theoretical prediction of the resistivity maximum in those cases where the modified phonon contribution can be expressed as $\Delta\rho_{ph} \simeq -bT$ at high temperatures. In combination with $\Delta\rho_{RKKY} = -c/T$, the total resistivity may exhibit a maximum at a temperature $T_M \propto x$ [as derived in Eqs. (24) and (25)]. This behavior is in good agreement with the data for $AuCr_x$ and $AgMn_x$ as shown in Fig. 9. Thus the qualitative features of the RKKY spin-flip scattering contribution to the resistivity appear to give a good account of the resistivity maximum over a wide concentration range.

IV. CONCLUSIONS

Our computations of electron scattering by a coupled pair of nearest-neighbor impurities yield a good account of the resistivities observed in many alloys with intermediate concentrations of magnetic impurities. Explicit comparison of our results to the data on $AgMn_x$, $CuMn_x$, $AuMn_x$, and $AuCr_x$ reveal interesting trends in the concentration variation of the magnetic scattering in the concentration regime where the Kondo effect is not operative.

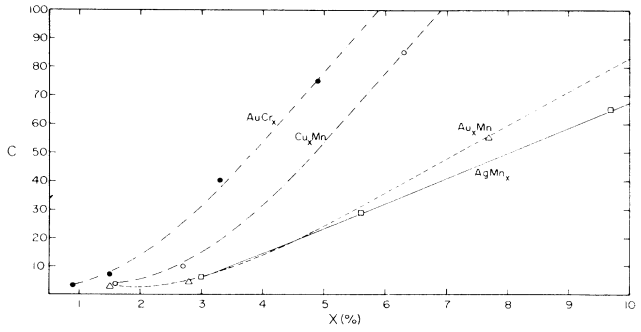

FIG. 8. The strength of the magnetic impurity pair scattering c is determined from a fit to the resistivity data using Eq. (19) and plotted here as a function of concentration for four spin-glass alloys. The lower concentrations are expected to yield a quadratic concentration behavior from the leading scattering term. The slower increases in c at higher concentrations may indicate higher order scattering contributions as discussed in the text.

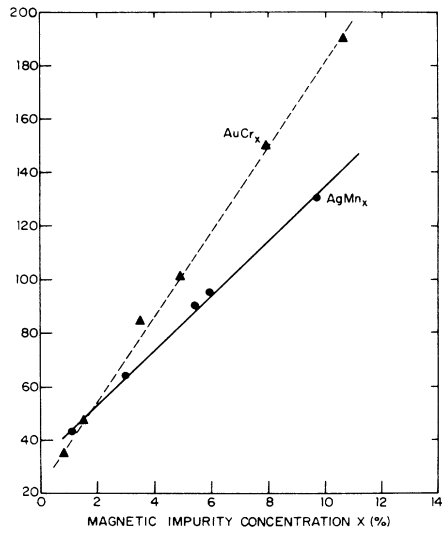

FIG. 9. The temperatures T_M characteristic of the resistivity maximum as a function of impurity concentration are shown at various alloy concentrations. The combined effects of a modified phonon contribution to the resistivity and the spin-flip RKKY scattering predict a maximum with T_M proportional to the concentration x.

Also, departures from the data at the lowest temperatures become more pronounced as the impurity concentration is increased, and they indicate directions for future analysis of higher-order scattering terms from the same pair of impurities as well the possibility of more extended impurity clusters. Such calculations may have a bearing on the short-range magnetic ordering which has been studied by neutron scattering and by magnetic susceptibility measurements.

Providing that modified phonon scattering or some other contribution yields a decline in the resistivity at high temperatures, the strong decrease in the low-temperature region created by the RKKY spin-flip mechanism may change the slope of the *total* resistivity and thereby yield a resistivity *maximum* at a characteristic temperature T_M. In $AgMn_x$ and $AuCr_x$, as well as similar alloys considered here, the prediction of a linear variation of the temperature T_M with impurity concentration is in accord with the data. Heavy-fermion compounds are noted for anomalous resistivity maxima as a function of temperature and recent research on these materials indicates a substantial contribution from the RKKY scattering.[17]

Rare-earth compounds provide other examples which are good candidates for the pair-induced resistivity mechanisms. Taking Th_xU as an example, it is interesting that the resistivity data[18] shows no minimum in the dilute (ppm) concentration range, but shows a pronounced minimum at $x=1\%$, 2%, etc. which scales according to the behavior expected from RKKY interactions.

At the lowest temperatures, the resistivity must of course saturate to comply with the unitarity limit on the scattering cross section. There are higher-order diagrams which represent scattering in the exchange interaction J arising form one pair of impurities. We find that these diagrams (which are not shown here) yield a leading correction of the type $\rho=\rho_{\text{pair}}(1+2U/15T+\ldots)$. Another series of corrections which become essential at low temperatures are the multiple scattering graphs commonly referred to as the "ladder" series. Summing these higher-order impurity cluster diagrams yields a resistivity of the form $\rho\cong\rho_{\text{pair}}T(T-T_N)^{-1}$, where the impurity magnetic ordering temperature is $T_N\propto xU$. This divergence at T_N coincides with the linked cluster calculations by Abe,[19] who used the mean-field approximation in the impurity averaging. Therefore an extension of the present analysis to higher-order clusters of impurities would be of interest with regard to the understanding of the short-range order observed in some spin glasses. Furthermore, as the tempeature approaches the strength of the RKKY coupling between impurities, the thermal distribution of spin pair states becomes important and must be explicitly included in the theory.

Concerning the regime of validity for our theory it may be helpful to emphasize the key role of the concentration of magnetic impurities. Recently several authors have distinguished regimes of interest for a *single* pair of impurities. By comparison, our examples are classic Kondo systems for concentrations measured in parts per million. However, the intermediate concentration regime of X ranging from 0.5% to 10% demonstrates a strong dominance of the impurity pair spin-flip scattering term, which has been previously neglected by others. Hence, these systems are characterized by coupling parameters U comparable to the Kondo temperature T_k, and the estimated parameters J, E_F, and U given in Table II support this notion. Hence, our analysis suggests that the pair spin-flip scattering emphasized here should be included in future calculations of concentrated Kondo systems.

Our mechanism does not give a divergence from the multiple scattering of an electron by a single impurity in the analogous fourth-order perturbation expansion $\rho\propto J^4$, because the "RKKY coupling" of an impurity spin to itself must be negligible. Nevertheless the single-impurity Kondo divergence in the scattering is quite sensitive to the RKKY coupling of neighboring spins as reviewed in Ref. 7, and analyzed recently by scaling methods.[19] Magnetoresistance measurements on spin-glass alloys may provide further clues to the magnetic scattering features discussed here. By the same token, the theory should be extended to take into account external fields as well as effective internal fields of other impurities generated by the exchange interaction.

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*Permanent address: Physics Department, Beijing University, Beijing, People's Republic of China.

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