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Kondo-Resistivity Suppression Due to Inelastic Scattering of Conduction Electrons by Magnetic Impurities in Alloys

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The interaction of localized magnetic moments of a magnetic impurity with the lattice causes relaxation of the states of that localized moment. The lifetime is represented as a broadening γ of the states of the localized moment. The introduction of this broadening changes the scattering of the conduction electrons by the localized moment from an elastic scattering to an inelastic one plus negligible elastic scattering. Thus, the electron of the average excitation energy above the Fermi level $\epsilon \approx T$ may take part in all inelastic allowed scattering processes only if $\epsilon \gtrsim \gamma$, i.e., $T \gtrsim \gamma$. For $T < \gamma$ this number of processes is reduced, and it vanishes for $T \to 0$. This effect was in fact found for the terms of the order J^2 and J^3 in the perturbation expansion, and the correct temperature dependence of the Kondo resistivity was obtained for $J < 0$. The resistivity for $J > 0$ is also discussed.

Recent measurements of the Kondo resistivity of Cu-Mn, [1] Au-Mn, [2] Ag-Mn, [3] and Au-Fe [4] alloys have shown deviation from the Kondo $1 - A\ln(\epsilon_F/T)$ behavior for low temperatures. It was found that at low temperatures the resistivity due to magnetic impurities first increases until it reaches a maximum (at T_M) and then decreases again and can be described by a $1 - A\ln(\epsilon_F/T)$ law for $T \gg T_M$. The Kondo temperature (T_K) for the Cu-Mn, Au-Mn, Ag-Mn systems is very low ($T \ll 0.1\ ^\circ\mathrm{K}$) and for Au-Fe it is [4] $T_K = 0.24\ ^\circ\mathrm{K}$. In all these cases [1-4] T_K is much smaller than T_M. Since $T_K \ll T_M$, the maximum in the resistivity cannot be associated with the formation of a quasibound state of conduction electrons around an impurity spin for $T < T_K$. [5] Thus a different mechanism, which suppresses the Kondo resistivity at $T < T_M$, must be looked for. Such a mechanism was suggested by Harrison *et al.* [6] They assume that the existence of an internal local magnetic field due to Ruderman-Kittel-Kasuya-Yosida (RKKY) impurity-impurity interaction suppresses the Kondo resistivity. [7] Silverstein [8] has considered the change of the mean magnetic moment of the impurity spin due to the change of the occupation of the Zeeman levels with the change in temperature in this internal local magnetic field. The calculated resistivity is in quite good agreement with experiment for T around T_M. [2-6] We, however, prefer to attribute to the RKKY impurity-impurity interaction merely a

broadening of the magnetic impurity spin states. It will be shown later that it is that broadening (γ) which suffices to explain the decrease in the resistivity below T_M. This broadening causes the scattering of the conduction electrons from the magnetic impurities to be mainly an inelastic one. The inelastic scattering governs the low-temperature behavior of the resistivity. The special characteristics due to the inelastic scattering vanish gradually for high temperatures ($T>\gamma$) and are negligible for $T\gg\gamma$. Suhl[9] has already shown that a Lorentzian broadening of the spin states will somewhat suppress the resistivity for $T> T_M$. We have obtained the changes in the resistivity due to the inelastic scattering for the whole temperature scale.

The Hamiltonian used in the calculation of the Kondo resistivity by many authors is the Kondo one,[10] where the impurity spin is assumed to have an infinite relaxation time, i.e., $\gamma=0$. For this system only elastic scattering of the conduction electrons from the impurity spins is possible. We consider an alloy where the magnetic impurity spin state relaxes and thus has a density of states of width γ. A possible mechanism for the broadening is the RKKY impurity-impurity interaction. Another possibility is the interaction of the magnetic spin with nonmagnetic impurities.[11] The magnetic impurity is assumed to scatter the conduction electrons by a $\vec{\sigma}\cdot\vec{S}$ interaction. We calculate the self-energy Σ of the conduction electrons Green's function in the first and second Born approximation under the following assumptions: (i) The impurity has spin $S=\frac{1}{2}$. The densities of states around each spin direction are assumed to be degenerate (i.e., the Zeeman levels are broadening but not split). This forms a band of spin states of width γ which is half-filled and hence has a Fermi level. This Fermi level need not be equal to the conduction electron Fermi level. (ii) γ is temperature independent.[2] (iii) The spin band is assumed to have a constant density of states over the width γ and to vanish elsewhere rather than to be Lorentzian. (iv) The spin Fermi level then lies in the middle of the spin band.[12]

Abrikosov[13] has shown that in the calculation of the resistivity, one should consider only $\mathrm{Im}\Sigma(\epsilon)$ of the conduction electrons possessing mean excitation energy $\epsilon\approx T$ above their Fermi level. Another dependence of $\mathrm{Im}\Sigma(\epsilon)$ on the temperature is due to the distribution functions of the spin and conduction electrons. Hence $\mathrm{Im}\Sigma(T,\epsilon)|_{\epsilon=T}$ is calculated. The resistivity is then

$$\rho_{\mathrm{alloy}}(T)-\rho_{\mathrm{alloy}}(0)=-(m/Ne^2)\,\mathrm{Im}\Sigma(T,\epsilon\to T)+\alpha T^5\ , \tag{1}$$

where m is the conduction-electron effective mass, N is the number of atoms, and e is the electron charge. The low-temperature phonon contribution is taken to be αT^5.[14] First $\mathrm{Im}\Sigma(0,\epsilon)|_{\epsilon=T}$ is calculated. In doing so no variation of the distribution functions with temperature is permitted. This expression can be evaluated analytically under the former assumptions (i)–(iv). We call this the $T=0$ approximation. The calculations give $\mathrm{Im}\Sigma(0,\epsilon)$, in terms of $E\equiv\epsilon/\gamma$, as

$$\mathrm{Im}\Sigma_{\mathrm{I}}(0,E)=-\frac{1}{\tau_0}\times\begin{cases}2E^2\ , & 0\le E\le\frac{1}{2}\\ 1-2(1-E)^2\ , & \frac{1}{2}\le E\le 1\\ 1\ , & 1\le E\end{cases} \tag{2}$$

for the first Born term, and

$$\mathrm{Im}\Sigma_{\mathrm{II}}(0,E)=-\frac{A}{\tau_0}\times\begin{cases}E^2[2\ln(2\epsilon_F/\gamma)+\frac{7}{3}]+\frac{1}{6}(\frac{1}{2}+2E)(1-2E)^2\ln(1-2E)+\frac{1}{6}(\frac{1}{2}-2E)(1+2E)^2\ln(1+2E)\ , & 0\le E\le\frac{1}{2}\\ (E-E^2)[2\ln(2\epsilon_F/\gamma)+\frac{7}{3}]-E(E-\frac{4}{3}E^2)\ln E+\frac{1}{3}(1-5E+4E^2)(1-E)\ln(1-E)\\ \quad+(2E-1)[\ln(\epsilon_F/\gamma)+\frac{3}{2}]+\frac{1}{4}(1-2E)^2\ln(2E-1)-\frac{1}{4}(1+2E)^2\ln(1+2E)+2E\ln 2\ , & \frac{1}{2}\le E\le 1\\ \ln[\epsilon_F/(\gamma E)]+\frac{3}{2}+(E-\frac{1}{2})^2\ln[1-1/(2E)]-(E-1)^2\ln(1-1/E)-(E+\frac{1}{2})^2\ln[1+1/(2E)]\ , & 1\le E\ll\epsilon_F/\gamma\end{cases} \tag{3}$$

for the second Born term. τ_0 and A are defined as

$$1/\tau_0\equiv\pi N_s(0)(J/N)^2S(S+1)N_i/2\ , \tag{4a}$$

$$A\equiv-4N_s(0)J/N\ , \tag{4b}$$

where $N_s(0)$ is the density of the conduction electrons at their Fermi level, J/N is the coupling constant, and N_i is the number of impurities.

In Fig. 1 $\mathrm{Im}\Sigma_{\mathrm{I}}(0,\epsilon)|_{\epsilon=T}$ and $\mathrm{Im}\Sigma_{\mathrm{II}}(0,\epsilon)|_{\epsilon=T}$ are shown for $\gamma=0.1\,^\circ$K and $\gamma=1\,^\circ$K, with $\epsilon_F=7$ eV (which applies to Cu alloys) and $A=0.045$. In Fig. 2

$$\Delta\rho(T)\propto\mathrm{Im}\Sigma_{\mathrm{I}}(0,\epsilon)|_{\epsilon=T}+(\mathrm{sign}\,J)\,\mathrm{Im}\Sigma_{\mathrm{II}}(0,\epsilon)|_{\epsilon=T}$$

is shown for both $J<0$ and $J>0$, where $\Delta\rho(T)$ is defined as $\Delta\rho(T)\equiv\rho_{\mathrm{alloy}}(T)-\rho_{\mathrm{alloy}}(0)-\alpha T^5$. The parameters of $\Delta\rho(T)$ in Fig. 2 were chosen to apply to the Ag-Mn system of Jha and Jerico[3] (alloy No. 2), with $\epsilon_F=5.5$ eV, $\gamma=7.4\,^\circ$K, and $A=0.05$. It is seen that the general correct be-

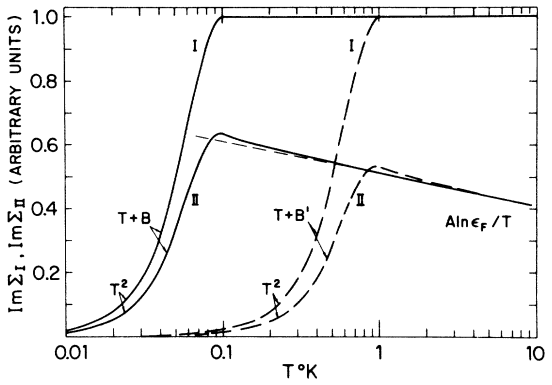

FIG. 1. $\mathrm{Im}\Sigma_I(0, \epsilon)|_{\epsilon=T}$ and $\mathrm{Im}\Sigma_{II}(0, \epsilon)|_{\epsilon=T}$ are shown for $\gamma = 0.1$ °K in bold line and for $\gamma = 1$ °K in dashed bold line. The dashed thin line is the continuation of the asymptotic $\ln(\epsilon_F/T)$ behavior to lower temperatures. B, B' are constants.

havior over the complete temperature scale is revealed by the present approximation. We have obtained the maximum at $T \approx \gamma$ a decrease in the resistivity from the maximum towards lower temperatures and a $1 - A\ln(\epsilon_F/T)$ behavior for $T \gg \gamma$. The decrease for $T < \gamma$ is interpreted as the exclusion of part of the inelastic scattering processes when $\epsilon \approx T < \gamma$. However, the quantitative fit is not complete, especially for $T < \gamma$ where the calculated resistivity is low. This limitation in the fit is mainly due to the $T = 0$ approximation. We have also calculated analytically $\mathrm{Im}\Sigma_I(T, \epsilon)|_{\epsilon=T}$ for $T \ll \gamma$ and $T \gg \gamma$ by taking into account the temperature dependence of the spin distribution functions. This has resulted in

$$\mathrm{Im}\Sigma_I(T, \epsilon)|_{\epsilon=T} = 5(T/\gamma)^2 , \qquad T \ll \gamma \qquad (5a)$$

$$\mathrm{Im}\Sigma_{II}(T, \epsilon)|_{\epsilon=T} = \tfrac{1}{2}[1 + \gamma/(6T)] , \quad T \gg \gamma . \qquad (5b)$$

The expressions obtained in Eqs. (5a) and (5b) give a temperature dependence of $\Delta\rho(T)$ which is similar to the asymptotic temperature dependence of $\Delta\rho(T)$ obtained from Eq. (2), except for the coefficients. In particular the low-temperature resistivity (for $T < T_\gamma$) is increased relative to that obtained from Eq. (2). It is expected that the second Born term will be changed similarly. The resistivity then obtained is shown in Fig. 2 by the solid lines. It is seen that the quantitative fit has improved considerably.

The following conclusions can be drawn from our results: (a) A simple model based on assumptions (i)–(iv) and the $T = 0$ approximation gives $\Delta\rho(T)$ similar to the experiment for the entire temperature scale. For $J < 0$ the decrease of $\Delta\rho(T)$ below T_M, the connection between T_M and γ, and the correct $1 - A\ln(\epsilon_F/T)$ law for $T \gg \gamma$ are obtained. (b) The *first* Born term gives the main contribution to the changes in $\Delta\rho(T)$ below T_M (as long as $\gamma \gg T_K$). It changes $\Delta\rho(T)$ also for $T > T_M$, and only for $T \gg T_M$ does it become a constant. (c) Since $\gamma = \gamma(c)$, where c is the impurity concentration, and $T_M \approx \gamma$, a relation $T_M = T_M(c)$ is obtained. In particular if $\gamma \propto c$, then $T_M \propto c$. Furthermore, if the relation $T_M = T_M(c)$ is once known, then c can be deduced from the value of T_M. (d) $\Delta\rho(T)$ for $J < 0$ will show a $1 - A\ln(\epsilon/T)$ behavior only for $T \gg \gamma$, and practically for $T \geq 10\gamma$, as is seen from Fig. 2. Therefore fitting a $1 - A\ln(\epsilon_F/T)$ expression to experimental results is plausible only for $T \geq 10T_M$. (e) For $T < \gamma$ $\Delta\rho(T)$ ($J < 0$) goes through various T dependence regions: For $T \to 0$ $\Delta\rho(T) \propto T^2$, for higher temperatures $\Delta\rho(T) \propto T + B$, then $\Delta\rho(T) \propto \ln T + B'$, until $\Delta\rho(T)$ flattens off and reaches the maximum. (f) $\Delta\rho(T)$ for $J > 0$ also has an interesting structure. It does not show a resistance maximum, but rather a shoulder at $T \approx \gamma$. For $T \gg \gamma$ it increases as $1 + A\ln(\epsilon_F/T)$.[15] (g) For $\Delta\rho(T)$ calculated from $\mathrm{Im}\Sigma(T, \epsilon)|_{\epsilon=T}$ it is interesting to see the dependence of it on the exact shape of the broadening. It is expected that for $T \ll \gamma$ or $T \gg \gamma$ the exact shape of the broadening is of no importance, but this is not so for $T \approx \gamma$. To show this we observe that in Fig. 2 the calculated resistivity at $T \approx T_M$ lies above the $1 - A\ln(\epsilon_F/T)$ line (for the $T = 0$ approximation). On the other hand, the resistivity for a Lorentzian broadening was shown by Suhl[9] to lie below the $1 - A\ln(\epsilon_F/T)$ line for $T \approx T_M$, in agreement with Gainon and Heeger's[11] experimental results. Thus if the $T = 0$ approximation does not cause the discrepancy, then it is the shape of the broadening used. (h) The divergence in the perturbation series given by Abrikosov[13] determines the Kondo temperature (T_K). This divergence is obtained when $\mathrm{Im}\Sigma_I(\epsilon) = \mathrm{Im}\Sigma_{II}(\epsilon)$.

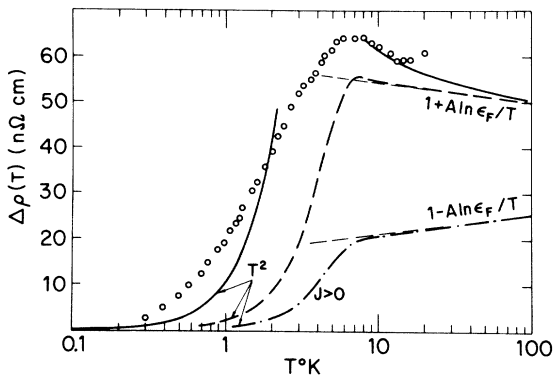

FIG. 2. $\Delta\rho(T) \propto \mathrm{Im}\Sigma_I(0, \epsilon)|_{\epsilon=T} + (\mathrm{sign}\,J)\,\mathrm{Im}\Sigma_{II}(0, \epsilon)|_{\epsilon=T}$ is shown for $J < 0$ by the dashed line and for $J > 0$ by the dash-dot line. The solid lines are $\Delta\rho(T) \propto \mathrm{Im}\Sigma_I(T, \epsilon)|_{\epsilon=T} + \mathrm{Im}\Sigma_{II}(T, \epsilon)|_{\epsilon=T}$ ($J < 0$) for $T \ll \gamma$ and $T \gg \gamma$. Open circle denotes the experimental values of $\Delta\rho(T)$ for Ag-Mn 0.332 at.% from Jha and Jericho (Ref. 3).

However this equality will never occur for a γ big enough, since the highest value of the second Born term is determined by $\ln(\epsilon_F/\gamma)$ [see Eq. (3) and compare it with Eq. (2)], i.e., $\mathrm{Im}\Sigma_{II}(\epsilon) < \mathrm{Im}\Sigma_I(\epsilon)$. For the weakly interacting system γ is always large enough for any practical impurity concentration. Hence $\mathrm{Im}\Sigma_{II}(\epsilon) < \mathrm{Im}\Sigma_I(\epsilon)$ for any ϵ. Then our results apply to $\Delta\rho(T)$ down to $T=0$. This is not always so for strongly interacting systems like Cu-Fe. There $\gamma < T_K$ is possible, and the whole perturbation series must be summed up.[13,16,17]

Measuring the resistivity of the following Kondo systems would be useful for comparison with the present theory: (a) a weakly interacting system like Cu-Mn, Ag-Mn, Au-Mn; (b) low impurity concentration, so that $T_{max} < 0.1\,T_{min}$, i.e., the experimental Kondo resistivity can be obtained up to $T \gg \gamma$; (c) very low temperature measurements down to $T < 0.1\,T_M$ in order that the residual $\rho_{alloy}(0)$ can be determined and subtracted from $\rho_{alloy}(T)$, and also that the behavior of $\Delta\rho(T)$ for $T \ll \gamma$ can be obtained.

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