

Consistent treatment of the effect of the impurity-impurity interaction on the Kondo resistivity

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We discuss the effect of the impurity-impurity interaction on the Kondo resistivity ρ_K without the explicit use of the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction between magnetic impurities. Both the RKKY and ρ_K originate from the s - d exchange interaction between the conduction electrons and the magnetic impurities. To be consistent we use *only the s - d interaction* to discuss the effect of the impurity-impurity interaction on the Kondo resistivity. For a system with a low Kondo temperature and for high temperatures, it is found that the $\ln T$ dependence of ρ_K is replaced by a $\ln(T^2 + c^2\bar{\Delta}^2)$ dependence (c is the impurity concentration and $\bar{\Delta}$ is independent of c and T). This is in accordance with results obtained when the RKKY interaction is explicitly included in the Hamiltonian.

I. INTRODUCTION

The Kondo resistivity ρ_K of dilute magnetic alloys was first discussed for the single-impurity case.¹ The results can be applied to dilute alloys provided the expected average Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction between neighboring magnetic impurities $J_{\text{imp,imp}}$ is weaker than the Kondo temperature T_K ,

$$J_{\text{imp,imp}} \approx AcJ_{sd}^2 S(S+1) \ll K_B T_K, \quad (1)$$

where J_{sd} is the coefficient of the s - d exchange interaction between the conduction electrons and the localized moments of the magnetic impurities with spin S , c is the concentration of impurities, ϵ_F is the Fermi energy of the conduction electrons, K_B is the Boltzmann factor, and A is a numerical coefficient. Experimental results for dilute Cu-Fe with $c < 560$ ppm seem to represent this case well. For these alloys² ρ_K/c is independent of c , i.e., ρ_K/c vs T can be presented by a universal curve for all $c < 560$ ppm. As T decreases below T_K the increase in ρ_K diminishes, and ρ_K levels off.

The high-concentration alloys in the present discussion are dilute alloys for which the inequality in Eq. (1) is inverted. Thus here high and low values of c are defined with respect to T_K . In all cases we consider dilute alloys, i.e., $c \ll 1$.

For a high concentration ρ_K exhibits a maximum³ at a temperature $T_m \gg T_K$, where T_m is of the order of $J_{\text{imp,imp}}/K_B$. ρ_K/c depends now on c and *decreases* at low temperatures. This temperature dependence of ρ_K is thus quite different from the one for low concentrations.

Let us consider ρ_K for large c . The decrease in ρ_K at low temperature is attributed to the RKKY interaction, while the RKKY interaction is assumed to exist independently of the Kondo effect.^{4,5} This is inconsistent with the fact that the RKKY inter-

action and the Kondo resistivity both originate from the s - d interaction and are therefore mutually dependent. For large c , however, this assumption seems to be valid. At first it was thought that the RKKY interaction resulted in the formation of internal magnetic fields which suppressed ρ_K for $T \rightarrow 0$.⁴ However, it was later shown that the maximum in ρ_K could also be due to correlations between impurities even if no internal field existed.⁵ Ford and Mydosh⁶ have recently shown experimentally that T_M indeed appears above the spin glass transition temperature. The decrease in ρ_K for low T is due to the quenching of inelastic scattering of conduction electrons by the system of interacting impurities.⁷

As c is lowered T_M decreases at first but then disappears when the expected value of T_m is less than T_K .^{2,8} The absence of a decrease in ρ_K as $T \rightarrow 0$ indicates that the RKKY interaction is quenched at low temperatures $T < T_K$, for systems with low c . This is probably due to the compensation of the impurity magnetic moment by the cloud of conduction electrons surrounding it.

We may conclude that at low temperature either the RKKY interaction exists and the Kondo resistivity is suppressed (large c), or the opposite happens—that ρ_K exists and the RKKY interaction is suppressed (low c).

To cover this wide spectrum of behavior of ρ_K/c in a consistent manner one has to calculate it from the s - d interaction only. The effect of the RKKY interaction (contained implicitly in the calculation) will show up at large c . Kozarzewski⁹ considered this problem using the coherent-potential approximation. His results, however, do not reveal a maximum in ρ_K for large c , nor do they give a universal curve for ρ_K/c vs T for low concentrations as observed in Cu-Fe dilute alloys.²

It is the purpose of this paper to present a calculation of ρ_K which incorporates the many-impurity effect on the Kondo resistivity using the s - d

interaction only. We make no explicit use of the RKKY interaction as done before.^{4,5} The discussion here is restricted for simplicity to systems with low T_K (i.e., relatively large c) and to high temperatures ($T > T_m \gg T_K$). We find that the many-impurity effect changes the $\ln T$ behavior of ρ_K to a $\ln(T^2 + c^2 \bar{\Delta}^2)$ behavior (here $\bar{\Delta}$ is independent of c and T). This is consistent with other theoretical work,⁵ where the RKKY interaction is explicitly used.

II. RESISTIVITY

Let us consider a system of noninteracting conduction electrons in an external small static electric field. N_i magnetic impurities are embedded in the N_0 atomic sites ($N_i \ll N_0$), and are randomly distributed in the system. The Hamiltonian of the system is

$$H = H_0 + H_{sd}. \quad (2a)$$

H_0 describes the conduction electrons in the presence of the electric field¹⁰:

$$H_0(t) = \sum_{\vec{p}, \nu} \epsilon_{\vec{p}} a_{\vec{p}, \nu}^{\dagger}(t) a_{\vec{p}, \nu}(t) - e \sum_{\vec{p}, \vec{p}'} U(\vec{p}' - \vec{p}) a_{\vec{p}', \nu}^{\dagger}(t) a_{\vec{p}, \nu}(t), \quad (2b)$$

where $a_{\vec{p}, \nu}^{\dagger}$ is the creation operator of a conduction electron with momentum \vec{p} and spin direction ν , e is the electronic charge, and $U(\vec{k})$ is the Fourier transform of the electric potential. H_{sd} is the s - d exchange interaction between the electrons and the magnetic impurities,

$$H_{sd}(t) = \left(-\frac{J}{N_0} \right) \sum_{\vec{p}', \vec{p}} F_n(\vec{p}' - \vec{p}) \vec{\sigma}_{\nu', \nu} \cdot \vec{S}_n(t) a_{\vec{p}', \nu'}^{\dagger}(t) a_{\vec{p}, \nu}(t) \quad (2c)$$

where J stands for J_{sd} , $\vec{\sigma}$ is the conduction-electron spin operator, and

$$F_n(\vec{p}' - \vec{p}) = e^{i(\vec{\sigma}' - \vec{\sigma}) \cdot \vec{R}_n}; \quad (3)$$

\vec{R}_n and \vec{S}_n are the position and the spin of the n th impurity atom. When we use the Heisenberg equation of motion ($\hbar = 1$)

$$i \frac{\partial}{\partial t} \hat{A}(t) = [\hat{A}(t), H(t)], \quad (4a)$$

as well as the relations

$$[a_{\vec{p}, \nu}^{\dagger}(t), a_{\vec{p}', \nu'}(t)]_{\pm} = \delta_{\vec{p}, \vec{p}'} \delta_{\nu, \nu'}, \quad [S_n^{\alpha}, S_n^{\beta}] = i \delta_{n, n'} \epsilon_{\alpha\beta\gamma} S_n^{\gamma}, \quad (4b)$$

then the rate of change of the electric current density, $\vec{J}(t)$, due to scattering of the electrons by the impurities is

$$\begin{aligned} \vec{I}(t) &= \frac{\partial}{\partial t} \vec{J}(t) \\ &= i \frac{J}{N_0} \\ &\quad \times \sum_{\substack{\vec{p}, \vec{q} \\ n, \alpha \\ \nu, \nu'}} \frac{\vec{p} - \vec{q}}{m_0} F_n(\vec{p} - \vec{q}) \sigma_{\nu', \nu}^{\alpha} g_{\nu', \nu}(n, \alpha | \vec{p}, \vec{q} | t), \end{aligned} \quad (5)$$

where m_0 is the electron mass, and the electron current density is,

$$\vec{J}(t) = \sum_{\vec{p}, \sigma} \frac{\vec{p}}{m_0} \langle a_{\vec{p}, \sigma}^{\dagger}(t) a_{\vec{p}, \sigma}(t) \rangle \equiv \sum_{\vec{p}, \sigma} \frac{\vec{p}}{m_0} g_{\sigma, \sigma}(\vec{p}, \vec{p} | t). \quad (6)$$

Let us define an M -electron, N -impurity, density-matrix element

$$\begin{aligned} &g_{\nu_1, \nu_2, \dots, \nu_{2M}}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{q}_1, \vec{q}_2; \dots; \vec{q}_{2M-1}, \vec{q}_{2M} | t) \\ &= \langle S_{n_1}^{\alpha_1}(t) \dots S_{n_N}^{\alpha_N}(t) a_{\vec{q}_1, \nu_1}^{\dagger}(t) a_{\vec{q}_2, \nu_2}(t) \dots a_{\vec{q}_{2M-1}, \nu_{2M-1}}^{\dagger}(t) a_{\vec{q}_{2M}, \nu_{2M}}(t) \rangle. \end{aligned} \quad (7)$$

The impurities in the system are randomly distributed. One must therefore average in (5) over all possible configurations of the impurities in the system.¹¹ To find the average in (5) we expand it in a power series in the electron-impurity interaction J by using the factorization technique.¹⁰⁻¹² We first separate (5) into two groups: group A,

for which $\vec{p} = \vec{q}$, and group B, for which $\vec{p} \neq \vec{q}$. The terms with $\vec{p} = \vec{q}$ vanish. For the terms in group B we write a higher-order equation of motion for the one-electron, one-impurity density matrix (with $\vec{p} \neq \vec{q}$). In general the equation of motion of the one-electron, N -impurity density matrix is (after taking a temporal Fourier transform and some re-

arrangements)

$$\begin{aligned}
& g_{\sigma\sigma}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{p}', \vec{p} | \omega) \\
&= \gamma(p', p, \omega) \left[-e \sum_{\vec{q}} [U(\vec{p} - \vec{q}) g_{\sigma\sigma}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{p}', \vec{q} | \omega) - U(\vec{q} - \vec{p}') g_{\sigma\sigma}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{q}, \vec{p} | \omega)] \right. \\
&\quad - \frac{J}{N_0} \sum_{\vec{q}, \nu, \beta} \left(\sum_{\vec{m}} [\sigma_{\sigma\nu}^\beta F_m(\vec{p} - \vec{q}) g_{\sigma\nu}(n_1, \alpha_1; \dots; n_N, \alpha_N; m, \beta | \vec{p}', \vec{q} | \omega) \right. \\
&\quad\quad\quad \left. - \sigma_{\nu\sigma}^\beta F_m(\vec{q} - \vec{p}') g_{\nu\sigma}(n_1, \alpha_1; \dots; n_N, \alpha_N; m, \beta | \vec{q}, \vec{p} | \omega)] \right. \\
&\quad\quad\quad \left. + i \sum_{\substack{j=1, N \\ \vec{q}', \nu', \gamma}} F_{n_j}(\vec{q}' - \vec{q}) \sigma_{\nu'\gamma}^\beta \epsilon_{\alpha_j \beta \gamma} \delta_{\alpha_j \nu'} g_{\nu'\sigma}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{q}', \vec{q}; \vec{p}', \vec{p} | \omega) \right] , \tag{8}
\end{aligned}$$

where m and β refer to the $(N+1)$ th impurity and

$$\gamma(p', p, \omega) \equiv [\epsilon_{p'} - \epsilon_p + \omega]^{-1} = P[\epsilon_{p'} - \epsilon_p + \omega]^{-1} - i\pi\delta(\epsilon_{p'} - \epsilon_p + \omega) . \tag{9}$$

At various orders we shall encounter products of the form $F_{n_1} \dots F_{n_N}$. The spatial average $\langle \dots \rangle$ of these products is,¹²

$$\begin{aligned}
& \langle F_{n_1}(\vec{q}_1) \dots F_{n_N}(\vec{q}_N) \rangle \\
&= \left(\frac{1}{N_0} \right)^N \sum_{n_1, \dots, n_N} \exp(i\vec{q}_1 \cdot \vec{R}_{n_1} + \dots + i\vec{q}_N \cdot \vec{R}_{n_N}) , \tag{10a}
\end{aligned}$$

where the sums in (10a) are over all atomic sites. In (10a) we do not consider disconnected averages that can be written in the form $\langle F_{n_1} \dots F_{n_j} \rangle \times \langle F_{n_{j+1}} \dots F_{n_N} \rangle$. For $N=1, 2$ Eq. (10a) becomes

$$\langle F_n(\vec{q}) \rangle = \delta_{\vec{q}, 0} , \quad \langle F_n(\vec{q}) F_{n'}(\vec{q}') \rangle = \frac{1}{N_0} \delta_{\vec{q}, -\vec{q}'} . \tag{10b}$$

We shall use next the following approximations: The external electric field is assumed to be small. Thus the distribution function of the electrons in equilibrium, $f(p)$, is a zero-order term, and we ignore second-order terms in the electric field. Density matrices of an order higher than a two-electron density matrix are ignored. We consider, however, many impurity correlations (to all orders), thus renormalizing the impurity spectral function. As to the electrons, we have only the lower-order Kondo terms, and we ignore correlations between electrons that may be important at very low temperatures. Two-electron, N -impurity density-matrix elements are approximated as products of a one-electron, N -impurity density

matrix and a density factor. This approximation is equivalent to the random phase approximation (RPA) for the electrons.¹³

To evaluate Eq. (5) we use Eq. (8) for the special case of one-electron, one-impurity density matrix and insert it into Eq. (5). The product of $F_{n_1} F_{n_2}$ then appears in Eq. (5). We divide all terms into two groups. For that selection we make a virtual averaging over the impurities and include in group A all terms for which $\langle F_{n_1} F_{n_2} \rangle \neq 0$, and in group B all the others. The importance of this partition is that we do not ignore the terms in group B which would otherwise vanish after decoupling the $F_{n_1} \dots F_{n_N}$ from the density-matrix elements and averaging over impurity sites. At this stage we have a one-electron, two-impurity density matrix and a two-electron, one-impurity density matrix. The latter is now decoupled to give a one-electron, one-impurity density matrix and a density factor $f(p)$. All terms now depend on one-electron density matrices. For the terms in group B we now use (8) and repeat the former process (starting with the equation of motion, etc.) to all orders in J . At each order we first separate the terms that can be added to group A from those that remain in group B. We then decouple the two-electron density matrix as before, and ignore terms which are of second order or higher in $f(p)$. This leaves us with one-electron, N -impurity density-matrix elements only. After carrying out this procedure to infinite order, we perform the averaging over the impurity sites for terms in group A, which is a trivial procedure, as mentioned before. Some of these terms vanish. Those left in the N th order are

$$\begin{aligned}
\langle\langle \tilde{\Gamma}^{(N)}(\omega) \rangle\rangle &= (-i) \left(-\frac{J}{N_0} \right)^N \\
&\times \sum_{\substack{\vec{p}, \vec{q}_1, \dots, \vec{q}_N \\ n_1, \dots, n_N, \alpha_1, \dots, \alpha_N \\ \mu_1, \dots, \mu_{N+1}}} \frac{\vec{p}}{m_0} \left[Q_{\mu_1, \dots, \mu_{N+1}}(\vec{p}, \vec{q}_1, \dots, \vec{q}_N | n_1, \alpha_1; \dots; n_N, \alpha_N | \omega) \right. \\
&\times \left(\langle\langle g_{\mu_1, \mu_{N+1}}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{p}, \vec{q}_N | \omega) \rangle\rangle - i \sum_{\substack{i=2, N \\ j=1, i-1 \\ \gamma}} f(q_{i-1}) \delta_{n_i, n_j} \epsilon_{\alpha_j \alpha_i \gamma} \right. \\
&\times \langle\langle g_{\mu_1, \mu_{N+1}}(n_1, \alpha_1; \dots; n_j, \gamma \dots; n_{i-1}, \alpha_{i-1}; n_{i+1}, \alpha_{i+1} \dots; n_N, \alpha_N | \vec{p}, \vec{q}_N | \omega) \rangle\rangle \\
&- Q_{\mu_1, \dots, \mu_{N+1}}^*(\vec{p}, \vec{q}_1, \dots, \vec{q}_N | n_1, \alpha_1; \dots; n_N, \alpha_N | \omega) \\
&\times \left(\langle\langle g_{\mu_{N+1}, \mu_1}(n_1, \alpha_1; \dots; n_N, \alpha_N | \vec{q}_N, \vec{p} | \omega) \rangle\rangle + i \sum_{\substack{i=2, N \\ j=1, i-1 \\ \gamma}} f(q_{i-1}) \delta_{n_i, n_j} \epsilon_{\alpha_j \alpha_i \gamma} \right. \\
&\times \langle\langle g_{\mu_{N+1}, \mu_1}(n_1, \alpha_1; \dots; n_j, \gamma \dots; n_{i-1}, \alpha_{i-1}; n_{i+1}, \alpha_{i+1} \dots; n_N, \alpha_N | \vec{q}_N, \vec{p} | \omega) \rangle\rangle \left. \right), \quad (11)
\end{aligned}$$

which depend on one-electron, N -impurity and one-electron, $(N-1)$ -impurity density matrices, and where

$$\begin{aligned}
Q_{\mu_1, \dots, \mu_{N+1}}(\vec{p}, \vec{q}_1, \dots, \vec{q}_N | n_1, \alpha_1; \dots; n_N, \alpha_N | \omega) \\
= \langle\langle F_{n_1}(\vec{p} - \vec{q}_1) F_{n_2}(\vec{q}_1 - \vec{q}_2) \dots F_{n_N}(\vec{q}_{N-1} - \vec{q}_N) \rangle\rangle \sigma_{\mu_1 \mu_2}^{\alpha_1} \dots \sigma_{\mu_N \mu_{N+1}}^{\alpha_N} \gamma(p, q_1, \omega) \dots \gamma(p, q_{N-1}, \omega). \quad (12)
\end{aligned}$$

The term $\langle\langle I^{(2N+1)}(\omega) \rangle\rangle$ gives contributions which are of the order $c^i J^{2N+1} \gamma^i$ in c , J , and γ , where $1 \leq i \leq N$. c is the impurity concentration, $c = N_i/N_0$. The highest order in c for a given N is c^N . Each order $2N+1$ in J introduces a higher-order term in c : $c^N J^{2N+1} \gamma^N$, which did not appear at a lower-

order term in J . On the other hand, it also gives lower powers of c , which appeared also at lower orders in J . We therefore neglect all these contributions except for $c^N J^{2N+1} \gamma^N$, which is the leading contribution to c^N . Thus the contributions to $\langle\langle I(\omega) \rangle\rangle$ are

$$\langle\langle \tilde{\Gamma}^{(2N+1)}(\omega) \rangle\rangle = -2i \left(\frac{J}{N_0} \right)^{2N+1} [i\pi S(S+1)N_i]^N [1 - (-1)^N] \sum_{\vec{p}, \vec{q}, \sigma} \{ \mathcal{H}(\epsilon_p) P[\gamma(p, q, \omega)] \}^N f(q) g_{\sigma, \sigma}(\vec{p}, \vec{p} | \omega) \frac{\vec{p}}{m_0}, \quad N=1, 2, \dots, \quad (13a)$$

$$\langle\langle \tilde{\Gamma}^{(2N)}(\omega) \rangle\rangle = \begin{cases} -2\pi \left(\frac{J}{N_0} \right)^2 N_i S(S+1) \sum_{\vec{p}, \sigma} \mathcal{H}(\epsilon_p) g_{\sigma, \sigma}(\vec{p}, \vec{p} | \omega) \frac{\vec{p}}{m_0}, & N=1, \\ 0, & N=2, 3, \dots, \end{cases} \quad (13b)$$

where $\mathcal{H}(\epsilon_p)$ is the density of the electron states of one-spin direction. $\omega \ll \epsilon_p$ because the important contribution comes from electrons with energy ϵ_p near the Fermi surface. In obtaining (13) we further approximate the one-electron, N -impurity density matrix as a product of an electron density matrix and an impurity density matrix. For the impurity density matrix we use a high-temperature approximation, since we are interested in temperatures above the spin glass transition temperature. We neglect correlations between different impurities. For each pair

$$\langle\langle S_n^\alpha S_{n'}^\beta \rangle\rangle = \delta_{\alpha\beta} \frac{S(S+1)}{3} \delta_{n, n'}, \quad (13c)$$

and the average over an odd number of spins vanishes.

Summing up all orders in (13) gives [together with Eq. (6)]

$$\tilde{\Gamma}(\omega) = -\frac{1}{\tau(\omega)} \vec{\mathcal{J}}(\omega), \quad (14)$$

where

$$\frac{1}{\tau(\omega)} = 2c\Delta \left(1 - \frac{2J}{N_0} \sum_{\mathbf{q}} \frac{P[\gamma(q_F, q, \omega)]f(q)}{1 + (P[\gamma(q_F, q, \omega)]c\Delta)^2} \right), \quad (15)$$

$$\Delta = \pi S(S+1) \frac{\mathcal{N}(0)}{N_0} J^2. \quad (16)$$

Assuming a constant density of state over a band of width $2D$ the static resistivity ρ_K is¹⁴

$$\rho_K = \frac{m_0}{n_e e^2} \frac{1}{\tau(0)}, \quad (17)$$

where from (15)

$$\begin{aligned} \frac{1}{\tau(0)} &= 2c\Delta \left(1 + \frac{2J\mathcal{N}(0)}{N_0} \int_{-D}^D \frac{f(\epsilon)\epsilon}{\epsilon^2 + (c\Delta)^2} d\epsilon \right) \\ &\approx 2c\Delta \left[1 + \frac{J\mathcal{N}(0)}{N_0} \ln \left(\frac{(2K_B T)^2 + (c\Delta)^2}{D^2} \right) \right], \end{aligned} \quad (18)$$

with $K_B T, \Delta \ll D$.

III. CONCLUSION

The resistivity calculated up to the Kondo term is modified to include the interaction between magnetic impurities which is implicitly included in the

s - d Hamiltonian, Eq. (2c). This results in the change of the $\ln T$ term in ρ_K to a $\ln[(2K_B T)^2 + c^2 \Delta^2]$ one [Eq. (18)]. This change will reduce the Kondo temperature, as was shown by Tsay and Klein.⁵ This temperature dependence is consistent with the deviation from an $\ln T$ behavior toward a concentration-dependent maximum observed in the large c alloys. Our calculation is valid at high temperatures. It is therefore not surprising that ρ_K obtains its maximum value only at $T=0$. The maximum value of ρ_K is obtained from Eq. (18),

$$(\rho_K)_{\max}/c \propto \left(1 + \frac{2J\mathcal{N}(0)}{N_0} \ln \frac{c\Delta}{D} \right), \quad (19)$$

which shows that $(\rho_K)_{\max}/c$ is expected to depend linearly on $\ln c$. This seems to be supported by the experimental results of Laborde and Radhakrishna³ for Cu-Mn alloys with $c = 25$ – 1000 ppm.

We have shown that it is possible to include the effect of the many-impurity interactions on the Kondo resistivity without considering the RKKY interaction as if it were independent of the Kondo effect. We have based our calculations only on the s - d interaction. It is believed that some approximations used here can be removed to extend the results to higher values of T_K and to low temperatures: $T < T_m, T_K$.

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