FOURTH-ORDER TERMS IN THE ELECTRICAL RESISTIVITY OF DILUTE MAGNETIC ALLOYS DUE TO A PAIR OF INTERACTING MAGNETIC IMPURITIES

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The effect of the exchange coupling of localized magnetic moments on the electrical resistivity of dilute magnetic alloys is investigated. The RKKY *s-d* exchange model is adopted and calculations of the electrical resistivity are performed up to fourth-order terms in the *s-d* exchange integral. It is found that as an effect of the magnetic interaction the Kondo results are strongly modified. In the case of the antiparallel ordering of the localized magnetic moments the spin-dependent part of the electrical resistivity is strongly reduced and the Kondo effect rather does not occur. For the ferromagnetic coupling it is verified that the electrical resistivity is generally greater than that of the noninteracting magnetic impurities. In the last case the Kondo minimum exists, but is shifted into the region of higher temperatures.

1. Introduction

The most interesting quantity in the Kondo problem is electrical resistivity [1, 2, 3, 4, 5]. The purpose of the present paper is to examine if the magnetic coupling between localized spins changes the second divergent term in the electrical resistivity of dilute magnetic alloys. The previous treatment of the Kondo effect by Beal-Monod, in which calculations were performed only up to the third-order terms and impurity spin was assumed to be 1/2, is generalized to arbitrary spin and fourth-order terms are taken into account. It is found that the exchange type coupling between impurities considerably modify the divergent terms. In our paper we perform the calculations on the basis of the RKKY model in the second Born approximation. We also include the exchange type coupling between a pair of localized moments. After lengthy but standard calculations we obtain general formulas for the electrical resistivity. The numerical results for spin 1/2 and 2 are presented in the last part of this paper. Our results may prove to be useful in the quest for a qualitative explanation of the temperature dependence of the electrical resistivity of

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dilute magnetic alloys when the concentration of the impurities increases and the interactions between magnetic atoms become more and more important. Preliminary results have been published recently [6].

2. *Hamiltonian of the system*

We consider the modified $s-d$ exchange model described by the Hamiltonian:

$$H = H + H_{\mathrm{ex}} + H_{s-d}, \tag{1}$$

where

$$H_b = \sum_{k,s} E_k a^+_{k,s} a_{k,s}$$

is the conduction band Hamiltonian for Bloch states of wave vector k, spin s and energy E_k $a^+_{k,s}$ and $a_{k,s}$ are the creation and annihilation operators of the electron in the state $|k, s\rangle$, respectively. The term H_{ex} represents the exchange type coupling of the pair of the magnetic atoms and is given as

$$H_{\mathrm{ex}} = -J_1 \cdot \vec{S}_1 \cdot \vec{S}_2, \tag{2}$$

where J_1 denotes the exchange parameter. $\vec{S}(S_1^x, S_1^y, S_1^z)$, $\vec{S}_2(S_2^x, S_2^y, S_2^z)$ are the spin operators of the magnetic atom. The last part of the Hamiltonian (1) describes the true $s-d$ exchange interaction and can be written as follows:

$$H_{s-d} = \frac{-2J}{N} \sum_{\substack{n,k,S \\ \vec{k}',S'}} [\exp i(\vec{k}-\vec{k}') \cdot \vec{R}_n] \vec{S}_n \cdot \vec{\sigma}_{s,s'} a^+_{\vec{k}'s'} a_{\vec{k},s} -$$

$$- \left(\frac{J}{N}\right)^2 \sum_{\substack{n,\vec{k}_1,\vec{k}_2,\vec{k}_3,\vec{k}_4, \\ S_1,S_2,S_3,S_4, \\ |E_2-E_4|>\frac{\Gamma}{2}}} [\exp i(\vec{k}_1+\vec{k}_2-\vec{k}_3-\vec{k}_4) \cdot \vec{R}_n] \, [\vec{S}_n \cdot \vec{\sigma}_{s_4,s_1} \delta_{s_3,s_2} -$$

$$- \vec{S}_n \cdot \vec{\sigma}_{s_3,s_2} \cdot \delta_{s_1,s_4} a^+_{\vec{k}_4,s_4} a^+_{\vec{k}_3,s_3} a_{\vec{k}_2,s_2} a_{\vec{k}_1,s_1}] \tag{3}$$

where Γ describes the width of the intermediate state due to the interaction between the localized magnetic moments and the itinerant electrons. The operators $\sigma_{s,s'}$ are defined as

$$\begin{aligned} \sigma_{++} &= (0, 0, \tfrac{1}{2}) \\ \sigma_{--} &= (0, 0, -\tfrac{1}{2}) \\ \sigma_{+-} &= (\tfrac{1}{2}, -i/2, 0) \\ \sigma_{-+} &= (\tfrac{1}{2}, i/2, 0) \end{aligned} \tag{4}$$

3. Calculation of resistivity

The electrical resistivity, R is the inverse of the conductivity and is given by:

$$R = \left[\frac{-e^2}{12\pi^3}\int \tau_{\vec{k}} v_{\vec{k}}^2 \frac{df_{\vec{k}}^0}{dE_{\vec{k}}} d^3\vec{k}\right]^{-1} \tag{5}$$

where f_k^0 is the Fermi distribution function. $\tau_{\vec{k}}$ denotes the relaxation time. In the second Born approximation we have

$$\frac{1}{\tau_{\vec{k}}} = \sum_B W(A \to B) \tag{6}$$

where

$$W(A \to B) = \frac{2\pi}{\hbar}(E_A - E_B)\left| H_{B,A} - \sum_C \frac{H_{B,C}H_{CA}}{E_A - E_C}\right|^2 \tag{7}$$

A and B are unperturbed states of the system of noninteracting localized magnetic moments and itinerant electrons with occupied an empty one electron state $|k, s\rangle$, respectively. E_A, E_B and E_C denote the energies of the unperturbed system in the states A, B, C, respectively. The calculation of the transition probability per unit time, $W(A \to B)$, is straightforwards and may be performed as done elsewhere [1, 2, 3].

Using the $s-d$ Hamiltonian (3) as the perturbing term H', and keeping all parts up to fourth order in the exchange parameter, we find that

$$\frac{1}{\tau_k} = \frac{C_1}{N} W_0 \left(1 - Jg(E_k) - \frac{8\pi J^2 \varrho^2}{N^2} \cdot \frac{kT}{\Gamma(S)}\right) A(S) \tag{8}$$

where $W_0 = 3\pi J^2/2hE_f$. C_1 is the number of the interacting pairs of the impurities, and ϱ denotes the density of the conductions band states at the Fermi level E_f. The functions $g(E_k)$, $\Gamma(S)$ and $A(S)$ are given by the expressions:

$$g(E_k) = \frac{1}{N}\sum_{\vec{q}} \frac{f_q^0}{E - E_k} \tag{9}$$

$$\Gamma(S) = \frac{3\pi J^2 \varrho^2}{12E_f}\left(\frac{E_f C_1}{N} + kT\right) A(S) \times$$

$$\times \left[1 + \sqrt{1 + \frac{6kT\left(\frac{2C_1 E_f}{N} + kT\right)}{\left(\frac{E_f C_1}{N} + kT\right)^2 A(S)}}\right], \tag{10}$$

$$A(S) = \frac{\sum_{S'=0}^{2S} S'(S'+1)\,(2S'+1) \exp \frac{J_1}{2kT} S(S+1)}{\sum_{S'=0}^{2S} (2S'+1) \exp \frac{J_1}{2kT} S(S+1)}. \qquad (1$$

Using the relations (5), (6) and (8) one obtains the following expression for the spin dependent part of the electrical resistivity:

$$R_s = R_1 + R_2$$

where

$$R_1 = \frac{C_1 R_0}{N}\left(1 + \frac{3J}{E_f} \ln T\right) A(S),$$

$$R_2 = \frac{\pi}{2} \frac{C_1 R_0}{N} \left(\frac{3J}{E_f}\right)^2 \frac{kT}{\Gamma(S)} A(S). \qquad (12)$$

The total resistivity of the dilute magnetic alloys is the sum of (12), the lattice resistivity and the resistivity arising from the scattering of the conduction electrons on the potential of the impurity. The impurity potential involves many causes, such as the difference of the valence, the redistribution of the conductions electrons around an impurity atom and the lattice distortion around it. Adding all these terms we obtain that the total resistivity of dilute magnetic alloys is given by the expression:

$$R = AT^5 + CR_v + R_r \qquad (13)$$

where AT^5 represents the lattice term (with $A \approx 3 \times 10^{-9}/°K^5$ for AuFe alloys). CR_v is the resistivity arising from the impurity potential, which is proportional to the concentration of the impurities [1].

4. *Discussion*

Let us first examine the behaviour of the spin dependent parts of the electrical resistivity R_1 and R_2 in the low and high temperature regions respectively:

1) In the case of ferromagnetic coupling *i. e.* for $J_1 > 0$ and at the same time $J_1/kT \gg 1$, we find that the magnetic terms in the electrical resistivity are given by

$$R_{1\uparrow\downarrow}(J_1 \gg kT) = 2S(S+1) \frac{C_1 R_0}{N}\left(1 + \frac{3J}{E_f} \ln T\right) \qquad (14)$$

$$R_{2\uparrow\downarrow}(J_1 \gg kT) = 6kT\left(\frac{C_1 E_f}{N} + kT\right) \times$$

$$\times \left[1 + \sqrt{1 + \frac{6kT\left(\frac{2E_f C_1}{N} + kT\right)}{\left(\frac{E_f C_1}{N} + kT\right)^2 2S(S+1)}}\right]. \qquad (15)$$

A closer analysis of (14) tells us that in a wide range of temperatures $R_{1\uparrow\uparrow}$ is greater than analogous term R_{1K} obtained by Kondo for noninteracting magnetic atoms. In the limit $T \to 0$ one obtains

$$\frac{R_{1\uparrow\uparrow}}{R_k} = \frac{2S+1}{S+1}. \tag{16}$$

On the other hand, the influence of the ferromagnetic interactions between localized magnetic moments on the forth-order terms, $R_{2\uparrow\uparrow}$, is negligible. In the high-temperature region, *i. e.* for $J_1/kT \ll 1$, the effect of magnetic coupling is quenched and two magnetic impurities are practically uncorrelated. The resistivity of the pair reduces to the sum of two Kondo resistivities of the isolated impurity. The graphical analysis of formulas (12) and (13) in Figs 1 and 2 shows that the Kondo point goes into the region of the higher temperatures.

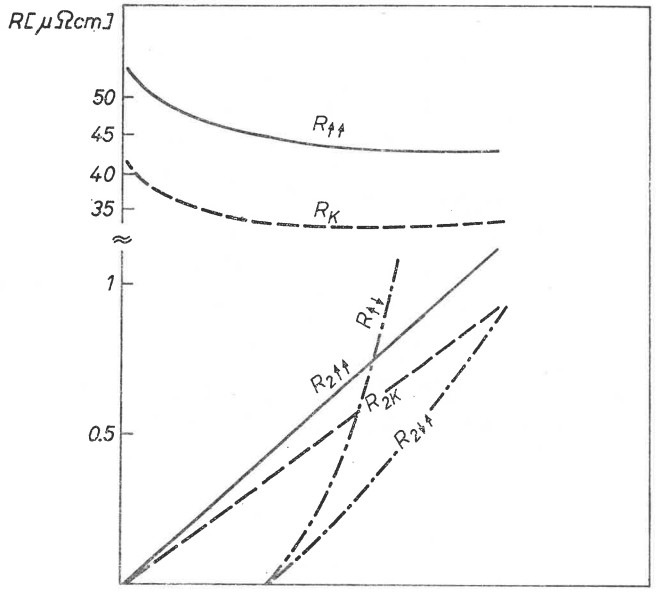

Fig. 1. Resistivity *vs* temperature of $Au_{0.91}$ $Fe_{0.09}$. R_{1K} — third order term for noninteracting magnetic impurities obtained by Kondo [1]. $R_{1\uparrow\uparrow}$, $R_{1\downarrow\downarrow}$— the same terms for parallel and antiparallel magnetic coupling of the localized magnetic moments, respectively. R_{2K}, $R_{2\uparrow\uparrow}$ and $R_{2\downarrow\downarrow}$ — the second divergent parts of the electrical resistivity. R_K, $R_{\uparrow\uparrow}$ and $R_{\downarrow\uparrow}$ — the total temperature-dependent electrical resistivity. The value of S is equal to 1/2

2) If antiparallel interaction between localized moments exists and $J_1/kT \ll -1$, the situation becomes quite different. The spin-dependent electrical resistivity tends exponentially to zero with decreasing temperature and in consequence of this there is no Kondo effect. This first-order term of the electrical resistivity is strongly reduced in a wide range of temperatures. In the zero temperature limit a comparison of the results of formula (12) with Kondo's term shows that

$$R_{1\uparrow\downarrow}(|J_1| \gg kT) = \frac{6C_1R_0}{N}\left[1 + \frac{3J}{E_f}\ln T \exp\left(-\frac{J_1}{kT}\right)\right] \tag{17}$$

$$R_{2\uparrow\downarrow}(|J_1| \gg kT) = 6kT\left(\frac{C_1 E_F}{N} + kT\right) \times$$

$$\times \left[1 + \left\{1 + \frac{kT\left(\frac{2E_f C_1}{N} + kT\right)}{\left(\frac{E_f C_1}{N} + kT\right)^2} \cdot \exp\left(\frac{J_1}{kT}\right)\right\}^{\frac{1}{2}}\right] \tag{18}$$

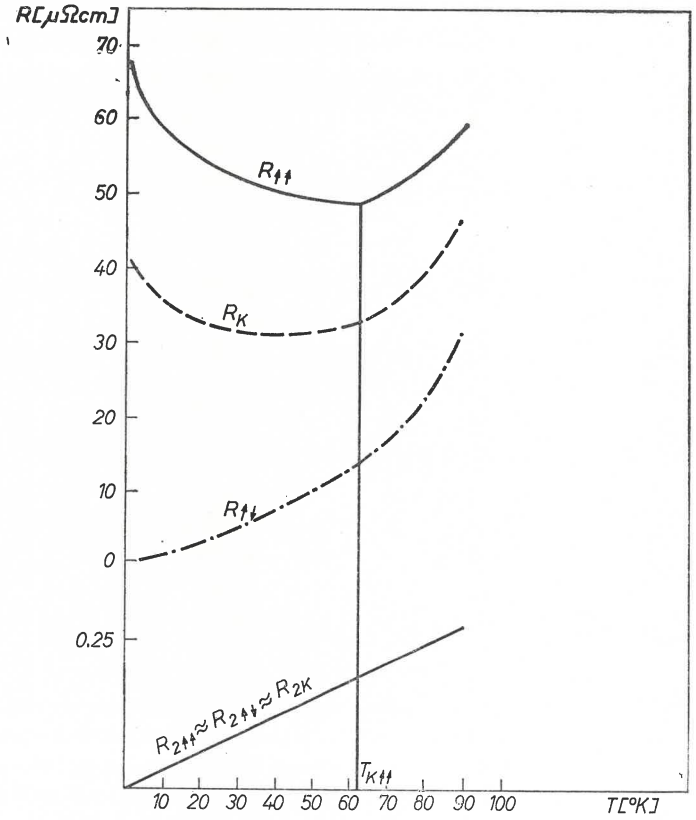

Fig. 2. The electrical resistivity for $S = 2$. The description of the curves is given in the Fig. 1

A more exact discussion of the magnetic parts of the electrical resistivity in the case of the antiferromagnetic coupling is shown in Figs 1 and 2. The values of spin are $\frac{1}{2}$ and 2, the exchange parameter J, corresponds to the Curie temperature of the $Au_{0.86}Fe_{0.14}$ alloy *i. e.* 104°K [7]. In the case of parallel and antiparallel interaction we take $J_1 > 0$ and $J_1 < 0$ respectively. Concentration of magnetic impurities is equal to $\frac{1}{z-1}$, where

z is the number of the first neighbours in the f. c. c. lattice. (At this concentration, assuming uniform distribution of the magnetic atoms, each impurity has in its neighbouring another magnetic atom.) The value of $R_0 = \frac{3\pi m J^2 V}{2e^2 N h E_f}$ is given in the Kondo paper [1] and is equal to 83.33 for $S = 2$. Note that the last value of the spin was found experimentally for the AuFe alloys [7].

5. *Conclusions*

We have calculated the transition probability per unit time for conduction electrons due to modified $s-d$ exchange interactions including the exchange coupling between magnetic impurities. We performed calculations in the second Born approximation up to fourth-order terms in the $s-d$ exchange integral. Using the standard relations for the relaxation time and resistivity we found the spin dependent parts of the electrical resistivity due to a pair of interacting magnetic impurities. In the case of noninteracting magnetic moments these terms give rise to a resistivity minimum. The most important conclusion reached in the present paper is that in the effect of the antiparallel exchange coupling between the magnetic atoms strongly reduces the electrical resistivity and the Kondo minimum rather does not occur. If the temperature decreases in the low temperature region the electrical resistivity due to the pair of antiparallel coupled impurities decreases exponentially. In the case of parallel interactions between the localized magnetic moments the magnitude of the spin-dependent part of the resistivity is generally greater than the Kondo terms. The resistivity minimum is shifted towards the higher temperatures. It is shown in the Fig. 2 that the magnitude of this shift is equal to 8–10°K. At very low temperatures the ratio of the electrical resistivity due to a pair of parallel coupled magnetic impurities to the Kondo term is equal to $\frac{2S+1}{S+1}$.

We believe that our simple model may be of some use in the description of the electrical resistivity of dilute magnetic alloys when the concentration of the magnetic impurities is not small enough for the interactions between the magnetic to be negligible. The exchange short-order interactions were found experimentally in a very large range of impurity concentrations by many authors [7]. The magnetization of the AuFe alloys, as measured by Crangle and Scott, at temperature down to 4.2°K shows that these alloys exhibit superparamagnetic behaviour. On the other hand, linear extrapolation of the curve of magnetic susceptibility suggests that no finite Curie temperature exists for these systems. Crangle and Scott conclude that alloys containing no more than 11% of iron exhibit no long-range ferromagnetism. Borg, Booth and Violet made Mössbauer effect experiments and investigated hyperfine fields using ^{57}Fe atoms [8]. They found evidence of short range magnetic alloys, down to 1% at Fe. The idea of the existence of superparamagnetic cluster regions in AuFe alloys rich in gold is also supported by magnetoresistance measurements carried out by Tournier and Ishikawa down to temperatures of the order 0.05°K. They conclude that the magnetization field curves in the alloys containing 8% Fe (*i. e.* when the concen-

tration of the impurities is about $1/(z-1)$) are consistent with the existance of an array of superparamagnetic particles [9].

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