Resistance Minimum in Dilute Magnetic Alloys

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Based on the s-d interaction model for dilute magnetic alloys we have calculated the scattering probability of the conduction electrons to the second Born approximation. Because of the dynamical character of the localized spin system, the Pauli principle should be taken into account in the intermediate states of the second order terms. Thus the effect of the Fermi sphere is involved in the scattering probability and gives rise to a singular term in the resistivity which involves $c \log T$ as a factor, where c is the concentration of impurity atoms. When combined with the lattice resistivity, this gives rise to a resistance minimum, provided the s-d exchange integral J is negative. The temperature at which the minimum occurs is proportional to $c^{1/5}$ and the depth of the minimum to c, as is observed. The predicted $\log T$ dependence is tested with available experiments and is confirmed. The value of J to have fit with experiments is about -0.2 ev, which is of reasonable magnitude. Our conclusion is that J should be negative in alloys which show a resistance minimum. It is argued that the resistance minimum is a result of the sharp Fermi surface.

§1. Introduction

A minimum in the resistivity-temperature curve of dilute magnetic alloys has been found in a number of alloys, including alloys of Cu, Ag, Au, Mg, Zn with Cr, Mn, Fe, Mo, Re, Os as impurities, and detailed experimental investigations have been made of various properties of these alloys.[1] A criterion of occurrence of the resistance minimum deduced from these observations may be that it is closely connected with the existence of localized magnetic moments of impurity atoms. Thus the only case of alloys of Au with the second series of transition elements which shows a minimum in resistivity is the alloys with Mo, which is also the only case where negative magneto-resistance is found so that localized moments are involved.[2] We also find no resistance minimum in alloys of Al, where no evidence of localized moments is found from the measurements of susceptibility.

~ The most convincing demonstration of the criterion is the observation made by Sarachik[3] that the dilute alloys of Fe (one atomic percent) with the series of NbMo alloys as host metals show a marked resistance minimum in the range of concentration ratio, where the measurements of susceptibility revealed the existence of localized moments of Fe atoms and none where localized moments were not found. Thus when a resistance minimum is found, we invariably find some evidence of localized moments, and inversely when localized moments are revealed from some measurements, we usually observe a minimum in resistivity. This conclusion indicates that the resistance minimum is a direct consequence of

the interaction between spins of the localized and conduction electrons, and is not connected with some specific assumptions concerning the band structures or crystal symmetries of host metals nor with characteristics of the d-levels of magnetic ions, such as the number of d-electrons, the location of d-levels relative to the Fermi surface or the features of splitting of degeneracy due to the crystalline field.

Thus the resistance minimum must be deduced from a simplified model for the system consisting of localized spins and conduction electrons, without introducing any specific assumptions. The model, which we take in the followings, is the s-d exchange model, which is due to Zener,[4] Kasuya[5] and Yosida[6] and has been applied to many magnetic metals and alloys.

Then our subject is to find which effect deduced from this model will lead to the resistance minimum. For this purpose we shall stress the following two experimental observations.

1. The temperature at which the minimum occurs, which we shall denote by T_{min}, is roughly proportional to $c^{1/5}$, where c is the concentration of impurity atoms.[2,7] This fact indicates that T_{min} is quite insensitive to the concentration. It is usually about 10 to 20°K for the concentrations of available experiments. This temperature is usually quite high compared with the magnitude of the exchange interaction between the localized spins. For example in the case of CuFe alloys T_{min} for the smallest concentration $(c = 5 \times 10^{-6})$ ever measured is still 10°K.[2] Thus we see that the localized spins are not correlated (completely paramagnetic) at the temperatures around T_{min} and above.

2. The depth of the minimum, which is defined by the resistivity at absolute zero $(\rho_{T=0})$ minus that at T_{min} (ρ_{min}), is roughly proportional to c.[2] $\rho_{T=0}$ is not strictly defined but usually defined as measured at the lowest temperature or extrapolated to the absolute zero. Since ρ_{min} is also proportional to c, the relative depth of the minimum is indepedent of c. Its magnitude is usually about 10%.

From these two observations it is clear that the appearance of resistance minimum is not due to a correlation between localized spins but is a result of adding contributions from each spin.

It is true that when the concentration is large correlation between spins has some effects on the resistivity at low temperatures where the exchange interaction between spins is not negligible. A maximum following the minimum for some alloys of higher concentrations is evidently related to this effect. However for lower concentrations the maximum moves to lower temperatures rapidly, while the minimum still remains above the helium temperature. Thus our conclusion is that the resistance minimum must be deduced from the s-d exchange model at temperatures where localized spins are not correlated without introducing specific assumptions.

At the paramagnetic range of temperature the resistance due to the s-d interaction is independent of temperature, when the transition probability is calculated in the first Born approximation. In the following sections we shall show that

a temperature-dependent term is obtained when the transition probability is calculated to the second Born approximation and that this term properly accounts for the resistance minimum when combined with the lattice resistivity.

§2. Calculations

We take as the unperturbed hamiltonian the following expression:

$$H_0 = \sum_{\mathbf{k}s} \varepsilon_\mathbf{k} a_{\mathbf{k}s}^* a_{\mathbf{k}s} , \tag{1}$$

where $\varepsilon_\mathbf{k}$ is the one-electron energy of the conduction electron with the wave number $\mathbf{k}$, $a_{\mathbf{k}s}$ and $a_{\mathbf{k}s}^*$ are its annihilation and creation operators, s denoting the component of the spin along the z-direction. According to the statement in the introduction, we have neglected the energy related to the localized spin system, the exchange and anisotropy energy. The perturbing hamiltonian is given by

$$\begin{aligned} H' = &- (J/N) \sum_{n\mathbf{k}\mathbf{k}'} \exp\{i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}_n\} \\ &\times \{(a_{\mathbf{k}'+}^* a_{\mathbf{k}+} - a_{\mathbf{k}'-}^* a_{\mathbf{k}-}) S_{nz} + a_{\mathbf{k}'+}^* a_{\mathbf{k}-} S_{n-} + a_{\mathbf{k}'-}^* a_{\mathbf{k}+} S_{n+}\}, \end{aligned} \tag{2}$$

where $\mathbf{R}_n$ denotes the position vector of the n-th impurity atom, whose spin operator is denoted by $\mathbf{S}_n$. $S_{n\pm}$ is defined by $S_{nx} \pm i S_{ny}$. We may choose any set of three mutually orthogonal space directions as the coordinate axes, because (2) has an isotropic form of the exchange interaction between the localized and conduction electrons. N is the total number of atoms in the crystal.

The expression (2) has been led by Kasuya,[4] who has taken account of the direct exchange interaction between the localized and conduction electrons. J denotes the magnitude of the interaction and may be positive. On the other hand, we have shown[8] that the mixing of the wave functions of the localized and conduction electrons can lead to an interaction of the same form as (2) with J negative. Both effects may exist and in the present paper we regard J as a parameter, of which the sign and magnitude will be determined later to have fit with experiments.

The transition probability per unit time from the initial state a to the final state b is given to the second Born approximation by

$$\begin{aligned} W(a \rightarrow b) = &(2\pi/\hbar)\delta(E_a - E_b) \\ &\times \{H'_{ab} H'_{ba} + \sum_{c \neq a}(H'_{ac} H'_{cb} H'_{ba} + \text{comp. conj.})/(E_a - E_c)\}, \end{aligned} \tag{3}$$

where a, b and c denote total states of the system and E_a, E_b and E_c are their total energies.*[)] The second term in the bracket represents the second Born

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*[)] It should be noted that in a previous paper,[9] where we have deduced the anomalous Hall effect based on the localized d-electron model, we have taken two factors of the matrix elements in the second term of (3) from the s-d exchange interaction (2) and the last factor from the hamiltonian which involves the spin-orbit interaction as a factor. It is what we have neglected in that paper as irrelevant to the anomalous Hall effect which we now take up in the present paper.

approximation for the transition probability and is usually smaller than the first by a factor J/ε_F, where ε_F is the fermi energy. It will be found, however, that it involves a factor which has a singular dependence on the energy of the initial state.

In order to show this, let us first consider the process, in which the electron with the wave number $\mathbf{k}$ and the positive sign of the spin (which will be denoted by $\mathbf{k}+$) is scattered to the final state $\mathbf{k}'$ with the same sign of the spin ($\mathbf{k}'+$). It should be noted that we are considering Slater determinants for the initial, final and intermediate states. The matrix elements in (3) should be taken between them. The intermediate states for this process can be devided into four groups.

1. The electron with $\mathbf{k}+$ is first scattered to the unoccupied state $\mathbf{q}'+$ and then to $\mathbf{k}'+$.

2. The order of the successive processes may be interchanged. One of the occupied electrons, which we shall denote by $\mathbf{q}+$, is first scattered to $\mathbf{k}'+$ and then the electron with $\mathbf{k}+$ fills up the state $\mathbf{q}+$ which is now empty.

It should be noted that the final states obtained by the above two processes have the opposite sign. For the first we may write the process as

$$| \cdots \mathbf{q}+ \cdots \mathbf{k}+ \rangle \rightarrow | \cdots \mathbf{q}+ \cdots \mathbf{q}'+ \rangle \rightarrow | \cdots \mathbf{q}+ \cdots \mathbf{k}'+ \rangle ,$$

whereas for the second as

$$| \cdots \mathbf{q}+ \cdots \mathbf{k}+ \rangle \rightarrow | \cdots \mathbf{k}'+ \cdots \mathbf{k}+ \rangle \rightarrow | \cdots \mathbf{k}'+ \cdots \mathbf{q}+ \rangle .$$

3. The sign of the spin may be changed in the intermediate states. The electron with $\mathbf{k}+$ is scattered to the state $\mathbf{q}'-$, while the z-component of the spin of the n-th atom, which we shall denote by M_n, is increased by unity ($M_n \rightarrow M_n+1$). After that the electron is again scattered to $\mathbf{k}'+$, while the z-component of the n-th spin returns to M_n.

4. The reverse of the above. One of the occupied electrons, denoted by $\mathbf{q}-$, is scattered to the state $\mathbf{k}'+$, M_n being decreased by unity. The electron with $\mathbf{k}+$ fills the state $\mathbf{q}-$, M_n returning to the initial value. The final states of the above two processes also have the opposite sign.

Then the second term in the bracket of (3) can be written as

$$\sum_{\mathbf{q}'} H'_{\mathbf{k}+,\mathbf{q}'+} H'_{\mathbf{q}'+,\mathbf{k}'+} H'_{\mathbf{k}'+,\mathbf{k}+} (1-f^0_{\mathbf{q}'})/(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{q}'})$$

$$-\sum_{\mathbf{q}} H'_{\mathbf{q}+,\mathbf{k}'+} H'_{\mathbf{k}+,\mathbf{q}+} H'_{\mathbf{k}'+,\mathbf{k}+} f^0_{\mathbf{q}}/(\varepsilon_\mathbf{q}-\varepsilon_{\mathbf{k}'})$$

$$+\sum_{n\mathbf{q}} H'_{\mathbf{k}+Mn,\mathbf{q}'-Mn+1} H'_{\mathbf{q}'-Mn+1,\mathbf{k}'+Mn} H'_{\mathbf{k}'+,\mathbf{k}+} (1-f^0_{\mathbf{q}'})/(\varepsilon_\mathbf{k}-\varepsilon_{\mathbf{q}'})$$

$$-\sum_{n\mathbf{q}} H'_{\mathbf{q}-Mn,\mathbf{k}'+Mn-1} H'_{\mathbf{k}+Mn-1,\mathbf{q}-Mn} H'_{\mathbf{k}'+,\mathbf{k}+} f^0_{\mathbf{q}}/(\varepsilon_\mathbf{q}-\varepsilon_{\mathbf{k}'})$$

$$+ \text{comp. conj.} , \tag{4}$$

each term corresponding to the above four processes in the written order. Note the negative sign before the second and fourth term. Here the matrix elements are taken between one-electron states. $f^0_\mathbf{q}$ is the Fermi distribution function for

the electron with the energy ε_q. As mentioned in the introduction, we neglect correlation between any pair of localized spins. Then the above expression becomes

$$2(-J/N)^3 \sum_n M_n^3 \sum_{q'} (1-f^0_{q'})/(\varepsilon_k - \varepsilon_{q'})$$

$$-2(-J/N)^3 \sum_n M_n^3 \sum_q f^0_q/(\varepsilon_q - \varepsilon_{k'})$$

$$+2(-J/N)^3 \sum_n M_n(S-M_n)(S+M_n+1) \sum_{q'} (1-f^0_{q'})/(\varepsilon_k - \varepsilon_{q'})$$

$$-2(-J/N)^3 \sum_n M_n(S+M_n)(S-M_n+1) \sum_q f^0_q/(\varepsilon_q - \varepsilon_{k'}). \qquad (5)$$

Taking account of the energy conservation $(\varepsilon_k = \varepsilon_{k'})$, we see that the first and second term combine to give

$$2(-J/N)^3 \sum_n M_n^3 \sum_q 1/(\varepsilon_k - \varepsilon_q), \qquad (6)$$

which is just what is obtained when we take no account of the Pauli principle.[*] This state of affairs is usually encountered when the perturbing system has no internal degrees of freedom. Thus the problem of potential scattering of the conduction electrons in metals can be treated by the one-electron approximation, the exclusion principle being discarded. The scattering through intermediate states without change of the spin direction (the processes 1 and 2) may be regarded of the same nature as the potential scattering.

As a result of this fact, the expression (6) has little dependence on the initial energy ε_k and we will neglect it. For the same reason we shall discard the term which does not involve f^0_q in the third and fourth term of (5). On the other hand the term involving f^0_q does not vanish there, which becomes

$$(4J^3/N^3) \sum_n M_n^2 \sum_q f^0_q/(\varepsilon_q - \varepsilon_k). \qquad (7)$$

This term has remained because of the difference of matrix elements between the process 3 and 4. In the third process we first increase the z-component of a localized spin and then decrease it, whereas in the fourth we first decrease it and then increase it. The two processes do not give the same answer, which means $S_+ S_- - S_- S_+ \neq 0$. This simply expresses the dynamical character of the localized spin system. It has the internal degrees of freedom.

From the assumption that the localized spins are randomly oriented, we may have

$$\sum_n M_n^2 = \{S(S+1)/3\}cN.$$

Using this equation and introducing a notation

$$g(\varepsilon) = (1/N) \sum_q f^0_q/(\varepsilon_q - \varepsilon), \qquad (8)$$

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[*] The fact that this expression diverges arises from our neglect of the dependence of J on the wave number difference $\mathbf{k} - \mathbf{k'}$. Its proper account makes the expression convergent. But this problem is not our main concern.

we have the following expression for the transition probability from $\mathbf{k}+$ to $\mathbf{k}'+$:

$$W(\mathbf{k}+\to\mathbf{k}'+) = \{2\pi J^2 S(S+1)c/3\hbar N\}\{1+4Jg(\varepsilon_{\mathbf{k}})\}\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'}). \qquad (9)$$

As can easily be shown, this also gives the probability of the transition from $\mathbf{k}-$ to $\mathbf{k}'-$.

In the next place we shall consider the process in which the electron with $\mathbf{k}+$ is scattered to the state $\mathbf{k}'-$, while the n-th spin increases its z-component by unity $(M_n \to M_n+1)$. The intermediate states for this process can also be devided into four groups.

1. The state of the electron is first changed from $\mathbf{k}+$ to $\mathbf{q}'-$, the z-component of the n-th spin being increased to M_n+1, and then from $\mathbf{q}'-$ to $\mathbf{k}'-$, S_{nz} remaining to be M_n+1.

2. One of the occupied states, denoted by $\mathbf{q}-$, is changed to $\mathbf{k}'-$, all the z-components of the localized spins being unchanged. Note that in this process S_{nz} remains to be M_n (not M_n+1 as in the first process). Then the electron with $\mathbf{k}+$ fills up the empty state $\mathbf{q}-$, while M_n is increased to M_n+1. The final state obtained in this way has the opposite sign to that of the first process.

3. The electron with $\mathbf{k}+$ is scattered to the unoccupied state $\mathbf{q}'+$, all the z-components of the localized spins being unchanged, and then to the state $\mathbf{k}'-$, while M_n being increased by unity.

4. One of the occupied state $\mathbf{q}+$ is changed to the state $\mathbf{k}'-$, while M_n being increased by unity. Then the electron with $\mathbf{k}+$ is scattered to the empty state $\mathbf{q}+$, all the z-components of the localized spins being unchanged. Note that in the latter process S_{nz} remains to be M_n+1. The final state obtained in this way also has the opposite sign to that of the third process.

The transition probability is easily calculated as in the first case and is given by

$$W(\mathbf{k}+\ M_n \to \mathbf{k}'-\ M_n+1) = (2\pi J^2/\hbar N^2)(S-M_n)(S+M_n+1)$$
$$\times \{1+4Jg(\varepsilon_{\mathbf{k}})\}\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'}). \qquad (10)$$

Similarly we obtain

$$W(\mathbf{k}-\ M_n \to \mathbf{k}'+\ M_n-1) = (2\pi J^2/\hbar N^2)(S+M_n)(S-M_n+1)$$
$$\times \{1+4Jg(\varepsilon_{\mathbf{k}})\}\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'}). \qquad (11)$$

Summing these expressions over the impurities, we have the following expression for the transition probability of the spin-flip processes:

$$W(\mathbf{k}\pm\to\mathbf{k}'\mp) = \sum_n W(\mathbf{k}\pm\ M_n \to \mathbf{k}'\mp\ M_n\pm1)$$
$$= \{4\pi J^2 S(S+1)c/3\hbar N\}\{1+4Jg(\varepsilon_{\mathbf{k}})\}\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'}), \qquad (12)$$

which is just twice of (9).

We shall investigate the dependence of (9) and (12) on the energy of the initial state, $\varepsilon_{\mathbf{k}}$, which is entirely involved in the function $g(\varepsilon_{\mathbf{k}})$. At the absolute zero of temperature, $f^0_{\mathbf{q}}$ can be replaced by a step function, which is unity when $q < k_0$ and zero when $q > k_0$, where k_0 is the magnitude of the Fermi momentum. Then assuming that $\varepsilon_{\mathbf{k}} = \hbar^2 \mathbf{k}^2 / 2m$, we obtain

$$g(\varepsilon_{\mathbf{k}}) = (3z/2\varepsilon_F) \{1 + (k/2k_0) \log | (k-k_0)/(k+k_0) |\} \quad T = 0, \qquad (13)$$

where z is the number of conduction electrons per atom. The singular nature of this function' is common to the problems concerning the Fermi surface and reflects its sharpness. From this expression and (9) and (12), we see that W increases when the electron approaches the Fermi surface, provided J is negative. Since at $T \neq 0$ the average of $|k-k_0|$ for thermally excited electrons is proportional to T, we can, even at this stage of calculation, expect a term proportional to $\log T$ in the expression of the resistivity.

It is true that $g(\varepsilon_{\mathbf{k}})$ diverges when $\varepsilon_{\mathbf{k}}$ approaches to ε_F. This divergence arises from our neglect of the energy of the localized spin system. The spin levels of the localized system are completely degenerate. Both the exchange interaction between the localized spins and the anisotropy energy for the direction of the spin relative to the crystalline axes can lift the degeneracy and we shall have a convergent result. As stated in the introduction, we are interested in the temperature range much higher than these splittings, where we need not be worried about the divergence.

At a finite temperature it is not legitimate to use (13) for $g(\varepsilon_{\mathbf{k}})$, because we are interested in the electrons in the energy range of kT from the Fermi surface. It makes the calculation easier to retain the definition of $g(\varepsilon_{\mathbf{k}})$, (8), here and to carry out the summation after we get the expression for the resistivity.

The rate of change of the probability $f^{\pm}_{\mathbf{k}}$ with which the state $\mathbf{k} \pm$ is occupied due to the collision with the localized spins is given by

$$(\partial f^{\pm}_{\mathbf{k}}/\partial t)_{\text{coll}} = \sum_{\mathbf{k}'} W(\mathbf{k} \pm \to \mathbf{k}' \pm)(f^{\pm}_{\mathbf{k}'} - f^{\pm}_{\mathbf{k}})$$
$$+ \sum_{\mathbf{k}'} W(\mathbf{k} \pm \to \mathbf{k}' \mp)(f^{\mp}_{\mathbf{k}'} - f^{\pm}_{\mathbf{k}}). \qquad (14)$$

When the electric field E is applied along the x'-direction, we may put

$$f^{\pm}_{\mathbf{k}} = f^0_{\mathbf{k}} - E k_{x'} \, \varPhi(\varepsilon_{\mathbf{k}}), \qquad (15)$$

for the stationary distribution of $f^{\pm}_{\mathbf{k}}$. Then we have

$$(\partial f^{\pm}_{\mathbf{k}}/\partial t)_{\text{coll}} = -(f^{\pm}_{\mathbf{k}} - f^0_{\mathbf{k}}) \sum_{\mathbf{k}'} \{W(\mathbf{k} \pm \to \mathbf{k}' \pm) + W(\mathbf{k} \pm \to \mathbf{k}' \mp)\}$$
$$= -(f^{\pm}_{\mathbf{k}} - f^0_{\mathbf{k}})/\tau_k, \qquad (16)$$

where τ_k is defined by

$$1/\tau_k = \{3\pi z J^2 S(S+1) c/2\varepsilon_F \hbar\} \{1 + 4Jg(\varepsilon_k)\}. \qquad (17)$$

Then from a standard theory we can easily obtain the conductivity:

$$\sigma = -(e^2/12\pi^3)\int \tau_k v_k{}^2 (df^0/d\varepsilon_k) d^3\mathbf{k}, \tag{18}$$

where $v_k = \hbar k/m$. The resitivity is the inverse of σ and is expressed from (17) as

$$\rho_{\rm spin} = c\rho_M \{1 - (\hbar^2 J/\pi m k_0)\int g(\varepsilon_k)(df^0/d\varepsilon_k)d^3\mathbf{k}\}, \tag{19}$$

where ρ_M is defined by

$$\rho_M = 3\pi m J^2 S(S+1)(V/N)/2e^2\hbar\varepsilon_F, \tag{20}$$

V denoting the volume of the crystal.

We shall neglect all the terms of the order $(kT/\varepsilon_F)^2$. The integral in (19) is carried out as follows:

$$\int g(\varepsilon_k)(df^0/d\varepsilon_k)d^3\mathbf{k}$$

$$= (V/8\pi^3 N)\int\int f^0{}_{k'}(df^0{}_k/d\varepsilon_k)/(\varepsilon_{k'}-\varepsilon_k)d^3\mathbf{k}d^3\mathbf{k}'$$

$$= -(4m^3V/\pi N\hbar^6)\int\int (\varepsilon\varepsilon')^{1/2}f^0(\varepsilon)(df^0/d\varepsilon')/(\varepsilon'-\varepsilon)d\varepsilon d\varepsilon'$$

$$= -(8m^3V\varepsilon_F/\pi N\hbar^6)$$

$$-(4m^3V\varepsilon_F/\pi N\hbar^6)\int\int\log|(\sqrt{\varepsilon}-\sqrt{\varepsilon'})/(\sqrt{\varepsilon}+\sqrt{\varepsilon'})|(df^0/d\varepsilon)(df^0/d\varepsilon')d\varepsilon d\varepsilon'.$$

The integral in the last term may be calculated as

$$\cong \int\int\log|(\varepsilon-\varepsilon')/4\varepsilon_F|(df^0/d\varepsilon)(df^0/d\varepsilon')d\varepsilon d\varepsilon'$$

$$= {\rm const.} + \log T.$$

Combining these results together in (19) and neglecting all terms of the order (J/ε_F) except the one involving $\log T$ as a factor, we have

$$\rho_{\rm spin} = c\rho_M \{1 + (3zJ/\varepsilon_F)\log T\}. \tag{21}$$

As we have expected, this contains a singular term involving $\log T$ which increases towards low temperature, if J is negative. This term arises from the second Born approximation as a result of the dynamical nature of the spin system. Its singularity is associated with the sharpness of the Fermi surface. As stated above, the divergence of (21) at $T=0$ does not occur, if a proper account of the splitting of the Zeeman levels of the localized spins is taken. Then $\log T$ should be replaced by $\log T_0$ at $T \ll T_0$, where kT_0 is an energy of the order of the splitting.

§3.   Comparison with experiments

We assume that the total resistivity can be expressed as the sum of three terms: viz., the lattice resistivity ρ_L, the resistivity arising from the impurity potential and that due to the spin scattering (21):

$$\rho = \rho_L + c\rho_A + c\rho_M + c(3zJ\rho_M/\varepsilon_F)\log T, \tag{22}$$

where $c\rho_A$ is the resistivity due to the impurity potential, which involves many causes, such as the difference of valence, the redistribution of the conduction electrons around the impurity atom and the lattice distortion around it.

First we assume a phenomenological expression for the resistivity of alloys which is given by

$$\rho = aT^5 + c\rho_0 - c\rho_1 \log T \tag{23}$$

and see how far it can represent the experimental results. Differentiating it with respect to T, we see that the temperature at which the minimum occurs is given by

$$T_{\min} = (\rho_1/5a)^{1/5} c^{1/5}. \tag{24}$$

This dependence of $T_{\min}$ on the concentration of impurity atoms is a consequence of the logarithmic term in the resistivity.

In the next place, the depth of the minimum is given by

$$\rho_{T=0} - \rho_{\min} = \rho_1 c \{\log(T_{\min}/T_0) - (1/5)\}, \tag{25}$$

where T_0 is as explained in the preceding section. Actually it will be more appropriate to take it as the lowest temperature at which the measurement is carried out for samples of lower concentration. Since $T_{\min}$ is very insensitive to c and is about 20°K and T_0 may be taken below 1°K, the last factor of (25) is practically independent of c and has a value around 3 or so. Thus the depth of the minimum is proportional to c, as is observed.

Knook[2] has experimentally determined $T_{\min}$ and $\rho_{T=0} - \rho_{\min}$ as functions of the concentration of Fe atoms in Cu. For c between 5×10^{-6} and 10^{-3}, his results may approximately be represented by $T_{\min} = 115c^{1/5.3}$ degrees and $\rho_{T=0} - \rho_{\min} = 100c$ $\mu\Omega$cm. From the latter relation and (25) we find that $\rho_1 = 33\ \mu\Omega$cm, when the last factor of (25) is assumed to be 3. Then from (24) with ρ_1 given above and $a = 2.6 \times 10^{-10}$ (the value for pure copper), we have $T_{\min} = 120c^{1/5}$ degrees, which is in good agreement with Knook's result. The choice of the value for the last factor of (25) involves an ambiguity, but since the fifth root of ρ_1 is involved in (24), it causes only a little difference. From (24) we understand the experimental fact that $T_{\min}$ is usually lower for Ag and Au than for Cu, because a for the former are much larger than that for Cu.

Thus we have seen that the phenomenological expression (23) for the resistivity

of alloys well represents the experimental observations. Now our calculation shows that ρ_1 is given by

$$\rho_1 = -\rho_M(3zJ/\varepsilon_F). \tag{26}$$

The residual resistance ρ_0 defined in (23) is the sum of ρ_A and ρ_M. Its value is 1000 $\mu\Omega$ cm for iron in copper. We can expect that both ρ_A and ρ_M are of comparable magnitude and we shall arbitrarily take $\rho_M = 500\ \mu\Omega$ cm. Then from (26) with $\rho_1 = 33$ we have $J/\varepsilon_F = -0.022$ or $J = -0.15$ ev if $\varepsilon_F = 7$ ev. This magnitude of J is quite reasonable.

In order to test the logarithmic dependence on T we have made a comparison of (23) with an experiment[10] made on the alloys of iron with gold, in which the resistivity has been measured to very low temperatures. In Fig. 1 the three curves drawn represent the three functions, $0.20-0.0078 \log T$, $0.077-0.004 \times \log T$ and $0.034-0.0016 \log T$, respectively from the above, in units of $\mu\Omega$ cm. The agreement is quite good, particularly the steep rise at the lowest temperature is well represented by a logarithmic function. From (22) both the constant terms and the coefficients of the logarithmic terms must be proportional to concentrati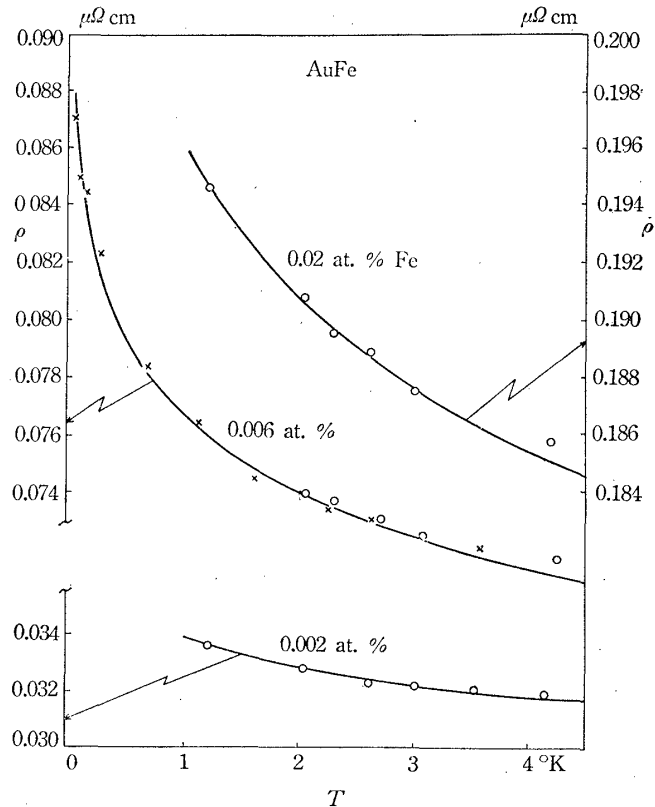on. Nominal solute concentration is indicated in the figure. The indicated values, however, are in proportion neither to the constant terms nor to the logarithmic terms. This may be due to inaccurate experimental determination of concentration. We see, however, that the three constant terms are in the ratio of 2.5 : 1 : 0.44, while the logarithmic terms are in the ratio of 2.0 : 1 : 0.40. This again confirms (23). If we assume that the concentration of the alloy with nominal 0.006 at. % iron is as indicated, we find from (23) that $\rho_1 = 67\ \mu\Omega$ cm. Then again with $\rho_M = 500\ \mu\Omega$ cm and $\varepsilon_F = 5.5$ ev, we have $J = -0.25$ ev, which is also of reasonable magnitude.

For many other alloys including CuMn, CuCr and CuFe we obtain good

Fig. 1. Comparison of experimental and theoretical ρ–T curves for dilute AuFe alloys.

agreements between theoretical and experimental ρ-T curves above the temperature at which the steep rise towards low temperature is suppressed by spin ordering and below the temperature where the lattice resistivity becomes predominant. In the temperature range where the steep rise due to lattice scattering sets in, we always find that theoretical curves (23) show less steep rise than experimental points, if parameters are adjusted at lower temperature and a is chosen for pure copper. At present we have no explanation of this fact.

Recently Coles[11] observed a new type of low-temperature resistivity anomaly in alloys of Rh with Fe. Instead of showing a minimum, the resitivity decreased more rapidly towards low temperatues. We found that this phenomenon can be accounted for by the present theory if J is assumed to be positive. (Although Coles reported that magneto-resistance is normal, the susceptibility measurement by Clogston et al.[12] revealed a localized magnetic moment for this alloy.) The resistivity increase due to alloying (denoted by $\Delta\rho$) is found to be proportional to c. The best fit with experimental results (dashed curves in Fig. 2) is obtained by $\Delta\rho/c = 3.4 + 30\,\log T\,\mu\Omega\,\mathrm{cm}$, which is represented by the continuous curve. Thus we have $\rho_1 = -30\,\mu\Omega\,\mathrm{cm}$, which is of the same magnitude as in the CuFe case but of the opposite sign. The constant term is much smaller here than in the AuFe case. In the present case the perturbation expansion should be carried out to higher orders. It is surprising that a single logarithmic term represents the experimental results rather well over a wide range of temperature.

Fig. 2. Resistivity increase divided by iron concentration.

§4. Conclusion and discussion

We have calculated the scattering probability for the conduction electrons due to the s-d exchange interaction to the second Born approximation without introducing any specific assumptions concerning the band structure and the location of the d-levels and have found a temperature dependent term for resistivity, which gives rise to a minimum in resistivity when combined with the lattice resistivity, provided the exchange interaction J between the conduction and localized electrons is negative (favourable for antiparallel spins). One of the important conclusions reached in this paper is that J should be negative in alloys showing resistance minimum. Our problem is then to find the origin of negative J. This was the subject of a previous paper[8] in which we showed
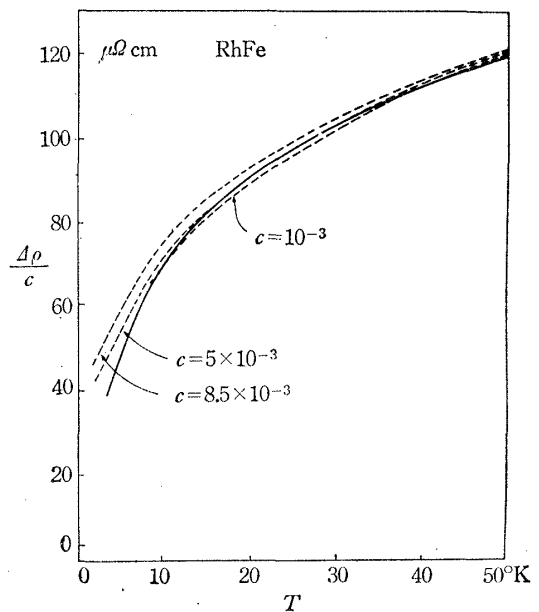
that the effect of mixing wave functions of conduction electrons and d-electrons leads to a negative J. The sign of J is then determined by the balance between the direct exchange interaction and the mixing effect. Our conclusion above shows that the resistance minimum is likely to be observed where the effect of mixing is large. In this connection the experiment by Sarachik[3] is interesting. She observed a resistance minimum in the critical range of concentration ratio of Mo to Nb where magnetic moment begins to appear. Here the effect of mixing may be expected to be so large that J is definitely negative.

We can correctly predict both the magnitude and the concentration dependence of the depth of minimum and the temperature at which the minimum occurs. A logarithmic term in resistivity is predicted and confirmed experimentally.

It must be stressed that the logarithmic term in resistivity can be traced back in its origin to the dynamical character of the localized spin system. Because of this the problem of s-d scattering is not properly treated in one-electron approximation and the effect of other electrons, that is, the effect of the Fermi sphere are involved. We encounter a similar situation in the case of the electron-phonon interaction. Because of the dynamical character of phonon (emission probability is not equal to absorption probability), electrons in phonon field are not properly treated in one-electron approximation but a kind of electron-electron interaction comes into play. This plays a part of attractive potential between electrons and causes superconductivity. It also has an effect of clothing conduction electrons and causes mass shift. This cloud on the conduction electrons is thus quite different in character from that on polaron. It involves a logarithmic function arising from the effect of the Fermi sphere.[13] In our case of the s-d interaction, too, mass shift should occur. This point will be a subject of further publication. An effect of the Fermi sphere is usually reflected in a singular logarithmic function in perturbation calculation as a result of the sharpness of the Fermi surface. Thus our conclusion is that the resistance minimum itself is a result of the sharp Fermi surface.

Acknowledgements

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