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RESISTIVITY DUE TO EXCHANGE SCATTERING IN DILUTE MAGNETIC ALLOYS

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Résumé. — Dans les alliages binaires dilués (jusqu'à des concentrations de 1 %) avec une distribution au hasard du soluté, il faut distinguer entre les atomes dissous, les atomes isolés et les paires de proches voisins. Si les atomes dissous ont un spin résultant et s'il existe une interaction d'échange entre premiers voisins ainsi qu'entre atomes dissous et électrons de conduction, il en résultera un terme dépendant de la température dans la résistivité due aux impuretés. Le signe de ce terme dépend du signe de l'interaction d'échange entre les atomes voisins dissous. Le modèle des paires fournit une explication possible de l'allure de la résistivité des alliages magnétiques dilués, à basse température, comme l'ont suggéré Brailsford et Overhauser, et aussi Dekker. On discute les conséquences de ce modèle.

Abstract. — In dilute binary alloys (up to about one atomic percent) with a random distribution of solute atoms, one may distinguish between isolated solute atoms and pairs of nearest neighbours of such atoms. If the solute atoms have a resultant spin and if there exists an exchange interaction between nearest neighbours as well as between the solute atoms and the conduction electrons, a temperature dependent impurity resistivity results. The sign of the temperature dependence is determined by the sign of the exchange interaction between neighbouring solute atoms, the wave vector of the conduction electrons at the Fermi level and the distance between nearest neighbours. The pair model provides a possible explanation of the behaviour of the resistivity of dilute magnetic alloys at low temperatures, as has been suggested by Brailsford and Overhauser, and by Dekker. Some consequence of this model are discussed.

According to Matthiessen rule, the electrical resistivity of metals and alloys is given by the sum of the resistivities corresponding to the various scattering processes suffered by the conduction electrons. This rule evidently requires the scattering processes to be independent; one may therefore expect it to hold, for example, in the case of dilute alloys. Thus, for a dilute alloy the resistivity may be written in the form

\[ \rho(T) = \rho_{\text{ph}} + \rho_i \]

Here, the first term on the right hand side refers to scattering by phonons; the last term includes scattering by impurities and other imperfections. Since \( \frac{d\rho_{\text{ph}}}{dT} > 0 \), a temperature-independent impurity resistivity implies a positive temperature coefficient of the total resistivity. This is the case, for example, in dilute diamagnetic alloys and this behavior is usually termed "normal". In such alloys the scattering process is caused by a perturbing potential \( V(r) \) produced by the impurity centers at the location of a conduction electron. In contrast to this behavior, many dilute alloys of transition elements in metals such as Cu, Ag and Au exhibit anomalies which may be classified roughly into two categories:

(a) \( \rho(T) \) exhibits a minimum at a temperature of the order of 10°K; an example is Fe in Cu.

(b) \( \rho(T) \) exhibits a minimum at some low temperature, followed by a maximum at a still lower temperature; examples are Cr in Au and Mn in Cu.

The concentrations for which these anomalies are observed are usually of the order of 0.1 atomic per cent. If one assumes that these alloys satisfy Matthiessen rule and that \( \rho_{\text{ph}}(T) \) is not essentially altered by the presence of the impurity atoms, one is faced with the problem of trying to find an explanation for a temperature dependent impurity resistivity. In particular, case (a) requires \( d\rho_i/dT < 0 \) in the low temperature region, whereas case (b) requires that \( \rho_i \) increases with increasing temperature to a maximum value and then decreases. One clue suggesting a possible explanation for these anomalies is provided by the fact that magnetic measurements on these alloys indicate the occurrence of a noticeable coupling between the spins of the impurity atoms even in dilute alloys. This coupling arises via the oscillating charge distributions surrounding the impurity atoms. As a result of the exchange interaction between the spin of an impurity atom and the conduction electrons, the charge distributions for the two possible magnetic quantum numbers of the conduction electrons will be different, resulting in an indirect exchange interaction between the impurity atoms. These arguments then lead to a model in which the impurity scattering differs from that in dilute diamagnetic alloys in two respects: 1° The perturbing Hamiltonian will contain terms corresponding to an exchange interaction between the conduction electrons and the impurity atoms, over and above the usual perturbation \( V(r) \); 2° As a result of the exchange coupling between the impurity atoms, inelastic collisions between the conduction electrons and the system of impurity atoms may occur. Also, one may expect interference between electron waves scattered by different centers at close proximity to become important.

Although the indirect exchange interaction
between the magnetic impurities probably extends over relatively large distances, it is instructive to consider a crude model in which one assumes that the coupling is limited to nearest neighbours. For the low concentrations of interest, it is then sufficient to distinguish between isolated magnetic impurities and pairs of coupled impurities. The isolated impurity-atoms give rise to a temperature independent resistivity, at least in the absence of magnetic fields. However, the scattering caused by the pairs leads to a resistivity which is temperature dependent. Brailsford and Overhauser [1], and the present author [2] have suggested that the scattering by pairs of coupled impurities provides at least a qualitative explanation for the observed anomalies. Without going into details, the essential features of this interpretation will be summarized.

Consider a pair of similar impurities with spin operators \( S_1 \) and \( S_2 \), and assume an exchange Hamiltonian of the form \( H_{\text{pair}} = -(2J_{12}/\hbar^2) S_1 \cdot S_2 \).

Employing the notation \( S_a^2 = S_a^2 = \hbar^2 a(a + 1) \), the energy levels of the pair are given by

\[
E(I) = J_{12}[2a(a + 1) - I(I + 1)]
\]

with \( I = 0, 1, \ldots, 2s \). The magnetic quantum number corresponding to the resultant spin of the pair will be denoted by \( M = -I, \ldots, (I - 1), I \).

Taking atom 1 at the origin of the coordinate system and atom 2 at \( R \), we choose for the perturbation produced by this pair at the position \( r \) of a conduction electron

\[
H'(r) = V \delta(r - 0) + V \delta(r - R) - (2J/\hbar^2) \delta(r - 0) S_a S_1 - (2J/\hbar^2) \delta(r - R) S_a S_2.
\]

The first two terms on the right hand side represents the ordinary Coulomb potential caused by the charge distributions around the impurity atoms, whereas the last two terms represent the exchange interaction between the conduction electron of spin \( S_a \) and the pair. The choice of delta functions is only a matter of expediency; it simplifies the calculations without impairing the essential argument. Let \( m_{\text{ea}} \) represent the magnetic quantum number of a conduction electron; the conduction electrons will be considered as free particles with an isotropic effective mass, independent of the wave vector \( k \).

Suppose the initial and final states of the system (conduction electron plus pair) are defined respectively by \( m_{\text{ea}}, I, M, k \) and by \( m_{\text{ea}}, I', M', k' \). In the Born approximation the probability for this transition is determined by

\[
|< m_{\text{ea}}, I', M' | H'(r) \exp\{ i(k - k') \cdot r \} | m_{\text{ea}}, I, M |^2.
\]

Upon evaluating these matrix elements one finds that the following types of collisions can occur:

**elastic collisions:**

\[ \Delta I = 0; \quad \Delta M = -\Delta m_{\text{ea}} = 0 \text{ or } \pm 1 \]

**inelastic collisions:**

\[ \Delta I = \pm 1; \quad \Delta M = -\Delta m_{\text{ea}} = 0 \text{ or } \pm 1. \]

Collisions corresponding to \( \Delta M = \pm 1 \) involve spinflip. From a knowledge of the matrix elements, the resistivity due to pairs can be found from transport theory, properly extended to include inelastic collisions. The interference of the waves scattered by the two partners of the pair appears in the form of an oscillating function \( F(k_F R) \), where \( k_F \) is the wave vector corresponding to the Fermi energy. Considering only the resistivity arising from exchange interaction (the last two terms in equation (3), one finds for the high temperature region \( kT \ll J_{12} \) the following contributions:

**elastic scattering:**

\[ \rho_{\text{elastic}} = \alpha J^2 [1 + J_{12}k_T] \]

**inelastic scattering:**

\[ \rho_{\text{inelastic}} = \alpha J^2 [1 - J_{12}k_T] \]

where \( \alpha \) is a constant. The total resistivity due to exchange scattering is thus proportional to \( 1 + J_{12}F(2kT) \). Since both anomalies mentioned earlier require \( d\rho/dT < 0 \) at high temperatures, this implies that \( J_{12}F(k_F R) > 0 \). This leads to the following two cases:

(a) \( J_{12} > 0 \) and \( F > 0 \). In this case the ground state of the pair is ferromagnetic. The resistivity due to elastic scattering is then a maximum for \( T = 0 \) and approaches monotonically a lower constant value at high temperatures. The inelastic scattering gives rise to a resistivity which increases from zero at \( T = 0 \) to a constant value at high temperatures. The total exchange resistivity shows a negative temperature coefficient and when combined with \( \rho_{\text{ph}} \) may produce an anomaly of type (a).

(b) \( J_{12} < 0 \) and \( F < 0 \). In this case the ground state of the pair is antiferromagnetic, producing an elastic exchange resistivity which rises from zero at \( T = 0 \) to a constant value at high temperatures. The inelastic scattering is responsible for the occurrence of a maximum in the total impurity resistivity; upon combination with \( \rho_{\text{ph}} \) this may give rise to an anomaly of type (b).

It thus appears that this model can account in principle for the observed anomalies. It must be admitted, however, that the model is only a crude approximation and that a realistic approach should take into account the relatively long range of the indirect exchange interactions between the impurities. Therefore, the present model does not permit a quantitative comparison with experiment.

**REFERENCES**
