ON THE NATURE OF IMPURITY FERROMAGNETISM

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The ferromagnetism arising upon introduction of a small amount of magnetic atoms into a nonmagnetic metal is considered. It is shown that the objection raised in Yosida's paper [3] against explaining this effect by exchange interaction between the admixture atoms and conductivity electrons is unfounded. The main magnetic characteristics of impurity ferromagnets are determined.

MATTHIAS, Suhl, and Corenzwit [1] have observed that ferromagnetism exists in a non-magnetic metal in which a small amount of paramagnetic atoms is dissolved. The idea that this ferromagnetism may be the consequence of exchange interaction between the momenta of the impurity atoms and the conduction electrons was advanced in the literature many times [2]. However, this idea was discarded after the publication of the paper by Yosida [3], where it was stated that such an interaction did not lead to the appearance of homogeneous polarization of the electron spin. Instead, according to Yosida, the polarization of the electron spin takes place near each impurity atom, and this polarization oscillates rapidly and decreases in amplitude with increasing distance from the impurity atom.

Actually, Yosida's statement is in error. Although the decrease in the spin density does indeed take place, it is nevertheless not very fast [proportional to $(p_0R)^{-3} \cos p_0R - (p_0R)^{-4} \sin p_0R$, where R is the distance from the impurity atom]. In view of this, in considering the polarization of the electron spin at a given point, it is necessary to take into account the influence of all the impurity atoms, not only the nearest one. This leads to an entirely different deduction than obtained by Yosida.

Indeed, let us take the Yosida formula for the density of the number of electrons with different spin orientations:

$$\rho_{\pm} = \frac{n}{V} \pm \frac{3}{4} \frac{n}{\varepsilon_F NV} \sum_q J(q) f(q) \sum_n S_n^z \cos [q(r - R_n)], \quad (1)$$

where 2n is the total electron number density, ε_F the Fermi limiting energy, N the number of crystal cells, J(q) the Fourier component of the exchange potential, S_n the spin of the n-th atom of the impurity, and

$$f(q) = 1 + \frac{4p_0^2 - q^2}{4p_0 q} \ln \left| \frac{2p_0 + q}{2p_0 - q} \right|$$

(p_0 is the limiting Fermi momentum).

We assume that the spin of the impurity atoms averaged over all atoms is different from zero, $\bar{S}_n \neq 0$. Since we are interested in homogeneous polarization, we can immediately confine ourselves to an examination of small values of q in (1). We also assume that the impurity atoms are distributed randomly in the lattice. It follows therefore that that sum over the impurity atoms can be replaced by an integral over R_n; as a result we obtain

$$\rho_{\pm} = \frac{n}{V} \left(1 \pm \frac{3}{2} c_i \frac{J(0)}{\varepsilon_F} \bar{S}_z \right), \quad (2)$$

where c_i is the number of impurity atoms per crystal cell. The impurity spin polarization thus gives rise to homogeneous polarization of the electron spin.

It is seen from the foregoing argument that Yosida's objections to attributing the impurity ferromagnetism to exchange interaction of the impurity spins and of the conduction electrons is unfounded. We investigate below the thermodynamic properties of such a model.

The interaction between the electrons and the impurity atoms can be written in the form

$$U = - \int s(r) \sum_i S_i J(r - R_i) d^3r, \quad (3)$$

where S_i and R_i are the spin and coordinate of the i-th impurity atom, J(R) some interaction function, and s(r) the electron spin density at the point r. The interaction (3) can be regarded as the result of the action of some fictitious magnetic field on the electron spin. We assume that the electrons become "magnetized" in this field, i.e.,

at each point of space some average electron moment is produced. In view of the random distribution of the impurity atoms, which in the mean is homogeneous in space, we can assume the average density of the electron spin to be independent of the coordinate. In this case U can be rewritten in the form

$$U = - as \sum_i \mathbf{S}_i,$$ (4)

where the constant is $a = \int J(\mathbf{r}) d^3\mathbf{r}$.

Formula (4) corresponds fully to the energy of the independent moments in the external field and it is possible to obtain from it in the usual fashion the impurity part of the free energy of the system (per unit volume):

$$F_i = - N_i T \left[\ln \operatorname{sh} \frac{as(S + 1/2)}{T} - \ln \operatorname{sh} \frac{as}{2T} \right],$$ (5)*

where N_i is the number of impurity atoms per unit volume.

The addition to the electron free energy for a specified magnetization (i.e., s) can be written in the form

$$F_e = (2\mu_0 s)^2 / 2\chi_0,$$ (6)

where μ_0 is the Bohr magneton and χ_0 the paramagnetic susceptibility of the electrons in the absence of impurities.

The value of s is obtained by minimizing the free energy $F_i + F_e$. The equation obtained is transcendental

$$4\mu_0^2 s/\chi_0 = N_i a S B_S (asS/T).$$ (7)

Here B_S is the known Brillouin function. It can be shown that for this value of s the sum $F_i + F_e < 0$ over the entire range of the temperatures where $s \neq 0$.

Let us consider Eq. (7) in the high-temperature region $T \gg asS$. Here

$$B_S(\alpha) \approx \frac{\alpha}{3} \frac{S + 1}{S} - \frac{\alpha^3}{45} \frac{(S + 1/2)^4 - (1/2)^4}{S^4}.$$

Substituting this in (7) we obtain in first approximation

$$s \left(\frac{4\mu_0^2}{\chi_0} - \frac{N_i a^2 S(S + 1)}{3T} \right) = 0.$$

This equation determines the Curie temperature, starting with which a nonvanishing value of s can appear:

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*sh = sinh.

$$T_c = \frac{N_i S(S + 1) a^2 \chi_0}{12\mu_0^2}.$$ (8)

Thus, the Curie temperature is proportional to the impurity concentration.

The next approximation of (7) in asS/T gives the temperature variation of s near T_c:

$$s^2 = \frac{5}{4\mu_0^2} \frac{S(S + 1) N_i \chi_0}{S^2 + S + 1/2} (T_c - T).$$ (9)

In the opposite limiting-case $T \to 0$, $B_S \to 1$ and we obtain

$$s = N_i a S \chi_0 / 4\mu_0^2.$$ (10)

The total moment comprises the electronic moment and the impurity moment, equal to $N_i \mu_0 g_i S B_S (asS/T)$, where g_i is the gyromagnetic ratio for the impurity atom. Thus, according to (7), the total magnetic moment per unit volume is

$$M = 2\mu_0 (1 + 2g_i \mu_0^2/a\chi_0) s.$$ (11)

From (9) and (11) we see that near T_c we have $M \sim (T_c - T)^{1/2}$. When $T = 0$ we obtain the saturation moment

$$M = \mu_0 N_i S (g_i + a\chi_0/2\mu_0^2).$$ (12)

Depending on the sign of the constant a, which can be different in different cases, the moment of the impurity and the moment of the electrons can be parallel or antiparallel. Note also that in this model we have assumed that there is no noticeable coupling between the impurity spin and the lattice. This means that $g_i \approx 2$ and thus only the new constant a appears in all the formulas.

To take into account the influence of the external magnetic field we must add to the free energy a term $-2\mu_0 sH$, and in addition, replace as in the impurity term of F_i [Eq. (5)] by as + $g_i \mu_0 H$.

As a result we obtain in place of (7)

$$\frac{4\mu_0^2}{\chi_0} s - 2\mu_0 H = N_i a S B_S \left[\frac{(as + g_i \mu_0 H) S}{T} \right].$$ (13)

From this, in particular, we obtain the paramagnetic susceptibility at $T > T_c$:

$$\chi = \chi_0 \left\{ 1 + \frac{T_c}{T - T_c} \left(1 + \frac{2\mu_0^2 g_i}{a\chi_0} \right)^2 \right\}.$$ (14)

The formulas obtained make it also possible to obtain the additional specific heat. Of course, as in all cases of second-order phase transitions, these formulas do not fit the experimentally observed singularity near the transition point as $T \to T_c$.

At temperatures $T \ll T_c$ formula (7) leads to an exponential temperature variation of all the quantities. This conclusion, apparently, cannot be correct for arbitrarily low temperatures, in view of the fact that the collective excitations of the spin systems are not taken into account here.

In conclusion it must be noted that in accordance with the experimental data the ferromagnetism arises not in all weak solutions of magnetic atoms in non-magnetic metals. For example in the case of a weak solution of Mn in Cu we obtain antiferro-magnetism [4]. The nature of this phenomenon is unclear at present. The attempt by Overhauser [5] to construct a theory for the antiferromagnetism of weak solutions appears unconvincing to us.

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