INDIRECT COUPLING BETWEEN LOCALIZED MAGNETIC MOMENTS BY NARROW BAND ELECTRONS

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Starting with the Modified Zener Hamiltonian as a model for a ferromagnetic metal, an effective exchange interaction between localized spins is derived. In contrast to the existing approaches exchange integrals depend via enhanced susceptibility on the actual polarization of itinerant electrons. Equations for magnetization, Curie temperature and susceptibility of the system are presented and compared with other similar approaches.

1. Introduction

The interaction between localized moments and itinerant electrons has attracted much attention for many years. In the simplest approach, itinerant electrons, assumed free, interact with the localized spins by means of the so called *s-d* exchange interaction. For some applications it is convenient to derive from the *s-d* model an effective coupling between localized spins, known as the RKKY interaction. The detailed description and applications of the RKKY interaction to the rare earth metals are given in Ref. [1].

However, in alloys or intermetalic compounds containing transition metals itinerant electrons are no longer purely *s*-like. It is generally accepted that at least a part of the *d* electrons from the transition metal atoms become also itinerant and provide an independent mechanism of interaction between localized rare earth moments. Bloch and Lemaire [2] and Bloch et al. [3] considered a model consisting of spins localized on sites occupied by rare earth metal atoms and a narrow band formed by *d* states from transition metal atoms. They used it to study paramagnetic susceptibility and ordering temperature of RE-Co alloys. Essentially the same model combined with the coherent potential approximation was shown to qualitatively account for mean magnetic moment per atom in some RE-transition metals alloys [4]. Arai and Parrinello [5] and Bartel [6] expressed the opinion (see also Ref. [8]) that the localized electron theory could become a useful (at least

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phenomenological) tool for studying ferromagnetism in transition metals and alloys provided that a small admixture of itinerant d electrons is present in order to introduce an adequate exchange coupling through intra-atomic effects. Some investigations were performed along this line using the so-called modified Zener model (MZM). In this model itinerant electrons are described by the Hubbard Hamiltonian, and they are coupled to the localized spins on each site by the usual exchange interaction. Bartel [6] applied the Green function method to calculate the dynamic susceptibility of such a system. In the following he used it to evaluate the spin-wave spectrum and Curie temperature.

The aim of the present paper is to find the indirect exchange interaction between localized spins. In contrast to the RKKY interaction or to the approach presented in Ref. [7] we do not restrict ourselves to the free electron approximation or to the paramagnetic ground state of itinerant electrons. However, for the sake of simplicity we consider only one narrow band and assume ferromagnetic ordering for the magnetic moments. The effective spin Hamiltonian is calculated by considering the average value of the term describing the interaction between itinerant electrons and localized spins by means of the Green function method. In addition, in Section 3 magnetization of both subsystems, Curie temperature and susceptibility above T_C are discussed.

2. *Derivation of the effective spin Hamiltonian*

In the Bloch representation the Hamiltonian of the MZM is

$$H = H_1 + H_2, \tag{1}$$

where

$$H_1 = \sum_{k,s} \varepsilon_k a_{ks}^{\dagger} a_{ks} + \frac{U}{N} \sum_{k,k',q} a_{k+q+}^{\dagger} a_{k+} a_{k'-q-}^{\dagger} a_{k'-}, \tag{2}$$

and

$$H_2 = -\frac{J}{N} \sum_{n,k,q'} e^{-iqr_n} [S_n^{+} a_{k+q-}^{\dagger} a_{k+} + S_n^{-} a_{k+q+}^{\dagger} a_{k-} + S_n^{z} (a_{k+q+}^{\dagger} a_{k+} - a_{k+q-}^{\dagger} a_{k-})], \tag{3}$$

$a_{ks}^{+}(a_{ks})$ is the creation (annihilation) operator of an electron in Bloch state (ks), $s = +\text{or}-$, $S_n^{+}, S_n^{-}, S_n^{z}$ stand for the operators of a spin localized at site n, U is the Coulomb interaction energy between electrons with opposite spins at the same site, J denotes the energy of exchange interaction between localized and itinerant electrons.

We want to calculate in some approximation the average value of H_2. This can be done by means of spectral theorems applied to the appropriate Green functions. For example, in order to calculate the average value of the first term in Eq. (3) we have to use the following Green function

$$\Gamma_{nkq}^{\dagger} = \langle\langle S_n^{+} a_{k+}; a_{k+q-}^{\dagger} \rangle\rangle = -i\theta(t) \langle \{ S_n^{+}(t) a_{k+}(t), a_{k+q-}^{\dagger}(0) \} \rangle.$$

The Fourier transformed equation for Γ^+_{nkq} is

$$(\omega-\varepsilon_k)\Gamma^+_{nkq}(\omega) = \frac{U}{N}\sum_{k',q'} \langle\langle S_n^+ a_{k-q+} a^\dagger_{k'-q'-} a_{k'-}; a^\dagger_{k+q-}\rangle\rangle$$

$$-\frac{J}{N}\sum_{l,q'} e^{-iq'r_l}[\langle\langle S_n^+ S_l^- a_{k-q'-}; a^\dagger_{k+q-}\rangle\rangle + \langle\langle S_n^+ S_l^z a_{k-q'+}; a^\dagger_{k+q-}\rangle\rangle]$$

$$+\langle\langle [S_n^+, H_2] a_{k+}; a_{k+q-}\rangle\rangle. \tag{4}$$

The following approximations are introduced

$$\langle\langle S_n^+ a_{k-q'+} a^\dagger_{k'-q'-} a_{k'-}; a_{k+q-}\rangle\rangle$$

$$\approx \langle a_{k'-q'-} a_{k'-}\rangle \Gamma^+_{nkq} - \langle S_n^+ a^\dagger_{k'-q'-} a_{k-q'+}\rangle \langle\langle a_{k'-}; a^\dagger_{k+q-}\rangle\rangle,$$

$$\langle\langle S_n^+ S_l^- a_{k-q'-}; a^\dagger_{k+q-}\rangle\rangle \approx \langle S_n^+ S_l^-\rangle \langle\langle a_{k-q'-}; a^\dagger_{k+q-}\rangle\rangle,$$

$$\langle\langle S_n^+ S_l^z a_{k+q'+}; a^\dagger_{k+q-}\rangle\rangle \approx \langle S_l^z\rangle \Gamma^+_{nk-q'q+q'},$$

and finally the last term in Eq. (4) is neglected because its contribution is of higher order when $|J| \ll U$.

The single particle Green function is calculated in the Hartree–Fock approximation and its Fourier transform becomes

$$\langle\langle a_{ks}; a^\dagger_{k's}\rangle\rangle = \delta_{kk'} G_{ks}(\omega), \tag{5}$$

where

$$G_{ks}(\omega) = (\omega-\varepsilon_{ks})^{-1} \tag{6}$$

and

$$\varepsilon_{ks} = \varepsilon_k + \tfrac{1}{2} Un - \tfrac{1}{2} s\Delta. \tag{7}$$

$$\Delta = Um + 2J\langle S^z\rangle \tag{8}$$

is the Stoner splitting and

$$n = n_+ + n_-, \quad m = n_+ - n_-, \tag{9}$$

$$n_s = \frac{1}{N}\sum_k \langle a^\dagger_{ks} a_{ks}\rangle. \tag{10}$$

In the following $G_{ks}(\omega)$ is used to find the thermal averages referring to itinerant electrons. Taking into account the above approximations and Eqs (5) and (6), equation (4) can be rewritten as follows

$$\Gamma^+_{nkq}(\omega) = -\frac{U}{N} G_{k+}(\omega) G_{k+q-}(\omega)\left[\sum_{k'} \langle S_n^+ a^\dagger_{k'+q-} a_{k'-}\rangle \right.$$

$$\left. -\frac{J}{N}\sum_l e^{iqr_l}\langle S_n^+ S_l^-\rangle\right].$$

Applying the spectral theorem we get

$$\sum_k \langle S_n^+ a_{k+q-}^\dagger a_{k+} \rangle = J \frac{\chi_{-+}(q)}{1-U\chi_{-+}(q)} \sum_l e^{iqr_l} \langle S_n^+ S_l^- \rangle,$$

where

$$\chi_{ss'}(q) = -\frac{1}{N} \sum_k \frac{f(\varepsilon_{k+qs}) - f(\varepsilon_{ks'})}{\varepsilon_{k+qs} - \varepsilon_{ks'}} \tag{11}$$

are the components of the static susceptibility of independent itinerant electrons. In a similar way we can express the average values of the other parts of H_2 in terms of the averages of products of localized spins operators. Including the lowest term, linear in spin operators, we get

$$\langle H_2 \rangle = -Jm \sum_i \langle S_i^z \rangle$$

$$- \sum_{ij} [J_{ij}^+ \langle S_i^+ S_j^- \rangle + J_{ij}^- \langle S_i^- S_j^+ \rangle + 2J_{ij}^z \langle S_i^z S_j^z \rangle], \tag{12}$$

where

$$J_{ij}^+ = (J_{ij}^-)^* = \frac{J^2}{N} \sum_q e^{iq(r_i - r_j)} \frac{\chi_{-+}(q)}{1-U\chi_{-+}(q)} \tag{13}$$

and

$$J_{ij}^z = \frac{J^2}{N} \sum_q e^{iq(r_i - r_j)} \frac{\chi_{++}(q) + \chi_{--}(q) + 2U\chi_{++}(q)\chi_{--}(q)}{1-U^2\chi_{++}(q)\chi_{--}(q)}. \tag{14}$$

We can interpret the right hand side of Eq. (12) as the average value of an effective spin Hamiltonian

$$H_S = -Jm \sum_i S_i^z - \sum_{ij} (J_{ij}^+ S_i^+ S_j^- + J_{ij}^- S_i^- S_j^+ + 2J_{ij}^z S_i^z S_j^z). \tag{15}$$

Note that Eq. (15) contains terms with $i = j$ which are neglected in RKKY theory. These terms correspond to an indirect interaction of the spin at the i-th site with itself via the band electrons. Thus we have arrived at the following simplified physical picture. The narrow band electrons interact with localized spins which are treated as a kind of an external quantum field. The Hamiltonian of the band electrons has a form of the Hamiltonian of MZM, Eqs (1) to (3). Properties of the localized spin system are determined by the effective spin Hamiltonian (15) with the exchange integrals J_{ij} dependent on the actual polarization of the band electrons which on the other hand depends on the magnetization of localized spins. So the determination of any physical quantity needs selfconsistent calculations. If the polarization of band electrons is neglected, our H_S is equivalent to an effective Hamiltonian (abandoning contribution from s electrons) derived in Ref. [7], if in addition $U \to 0$, then Eq. (15) reduces to the RKKY interaction.

3. *Magnetization and susceptibility*

As an illustrative example of our approach we will find and discuss the equations for the magnetization of the system. Once the effective spin Hamiltonian is derived it can be found in a straightforward way applying the Hartree–Fock and molecular field approximations to H_e and H_S, which then become

$$H_e = \sum_{ks} \varepsilon_{ks} a_{ks}^{\dagger} a_{ks},$$

$$H_S = -h \sum_i S_i^z,$$

where ε_{ks} is given by Eq. (7) and

$$h = Jm + J^2 \langle S^z \rangle \frac{\delta_+(T) + \delta_-(T) + 2U\delta_+(T)\delta_-(T)}{1 - U^2 \delta_+(T)\delta_-(T)} \tag{16}$$

with

$$\delta_s(T) = \lim_{q \to 0} \chi_{ss}(q) = -\frac{1}{N} \sum_k \frac{\partial f(\varepsilon_{ks})}{\partial \varepsilon_{ks}} \xrightarrow{T \to 0} \mathcal{N}_s(\varepsilon_F), \tag{17}$$

where $\mathcal{N}_s(\varepsilon)$ is the density of states function per spin direction. The magnetic moment per atom

$$M = g\mu_B(\langle S^z \rangle + \tfrac{1}{2} m), \tag{18}$$

has to be calculated from a set of selfconsistent equations

$$\langle S^z \rangle = S B_S\left(\frac{Sh}{k_B T}\right), \tag{19}$$

$$m = \frac{1}{N} \sum_k [f(\varepsilon_{k+}) - f(\varepsilon_{k-})], \tag{20}$$

and

$$n = \frac{1}{N} \sum_k [f(\varepsilon_{k+}) + f(\varepsilon_{k-})], \tag{21}$$

with the help of Eqs (16) and (17). Here $B_S(x)$ is the Brillouin function, n stands for the number of itinerant electrons per atom and Eq. (21) determines the chemical potential of electrons. The conditions for the Curie temperature in our model are

$$\langle S^z \rangle_{T=T_C} = 0, \quad m_{T=T_C} = 0. \tag{22}$$

Expanding the right hand sides of Eqs (19) and (20) to the first order in $\langle S^z \rangle$ and m and substituting into Eq. (22) allows one to write the following equation for T_C

$$\left[U + \tfrac{4}{3} J^2 \frac{S(S+1)}{k_B T_C}\right] \delta(T_C) - 1 = 0, \tag{23}$$

where

$$\delta(T) = \lim_{\Delta \to 0} \delta_s(T) = -\int \mathcal{N}(\varepsilon) \frac{\partial f(\varepsilon)}{\partial \varepsilon} d\varepsilon \tag{24}$$

and $\mathcal{N}(\varepsilon)$ means $\mathcal{N}_s(\varepsilon)$ above T_C. The Curie temperature following from Eq. (23) is about two times higher than that predicted by Bartel [6] because he has neglected the contribution to the molecular field acting on every localized spin due to the presence of the all other spins. This, however, does not change very much the qualitative features of T_C as a function of J, U, and n. In the same approximation, the static susceptibility of the system above the Curie temperature is

$$\frac{\chi(T)}{g^2 \mu_B^2} = \tfrac{1}{2} \bar{\delta}(T) + \frac{S(S+1)}{3k_B} \frac{[1+J\bar{\delta}(T)]^2}{T - T_C \dfrac{\bar{\delta}(T)}{\delta(T)}}, \tag{25}$$

where

$$\bar{\delta}(T) = \frac{\delta(T)}{1 - U\delta(T)}.$$

Qualitatively similar results for T_C and $\chi(T)$ were obtained by Bloch et al. in Refs [2] and [3] from their theory of RE-Co alloys. However, the equation for susceptibility presented in Ref [2] introduces several phenomenological parameters having no direct microscopic interpretation and the equation for the Curie temperature in Ref. [3] bears the same fault as the corresponding equation in Bartel's paper [6] does. It is seen from Eqs (19) and (25) that the magnetization as well as the susceptibility above the Curie temperature are not far from the corresponding results for completely localized spins only. The detailed behaviour of magnetization and susceptibility with temperature depends on the electrons density of states function and has to be found numerically.

4. Conclusions

We have examined a modified Zener model with the aim to separate partially, itinerant electron subsystem from localized spin subsystem in a selfconsistent way. As a result we have got Hamiltonians H_e and H_S describing electron and spin subsystems respectively. H_e is simply MZM Hamiltonian while H_S has a form of an anisotropic indirect exchange interaction which is a generalization of RKKY interaction for: (*i*) the ground state of the itinerant electrons not having to be paramagnetic, (*ii*) the correlations between itinerant electrons being taken into account, (*iii*) selfpolarization of the localized spins being taken into account, and (*iv*) exchange integrals depending on the actual magnetization of the system.

The advantages, in our opinion, of this two-stage approach to the MZM are as follows: (*i*) Once we have derived the effective spin Hamiltonian it provides in the Hartree–Fock and molecular field approximations relatively simple equations for magnetizations of both

subsystems. In particular in our approximation the molecular field acting on localized spins comes both from the itinerant and localized electrons in contrast to the approximation used in Refs [3] and [6], where only the first contribution appears. It accounts, for example, for a Curie temperature different by a factor of two. (*ii*) It allows for the easier development of an alloy theory for the model due to the fact that the itinerancy and localization are separated, but in a selfconsistent manner.

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