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# On the theory of carrier-induced ferromagnetism in diluted magnetic semiconductors

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Two different approaches (presented in the literature as alternative approximations) to the problem of carrier-induced ferromagnetism in the system of disordered magnetic ions of a diluted magnetic semiconductor are analyzed. They are based on a self-consistent procedure for the mean exchange fields and the RKKY interaction. Calculations in the framework of exactly solvable model are carried out, and it shows that these approaches stem from two different contributions to the magnetic susceptibility. One stems from the diagonal part of the carrier–ion exchange interaction and corresponds to mean field approximation. The other one stems from the off-diagonal part of the same interaction and describes the indirect interaction between localized spins via free carriers. These two contributions can be responsible for the different magnetic properties. Thus, the aforementioned contributions are complementary but not alternative to each other. A general approach is proposed and compared with different approximations to the problem under consideration.

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## Introduction

In recent years, there has been a substantial increase of interest in studies of carrier-induced ferromagnetism in diluted magnetic semiconductors (DMS). A number of works (see [1] and references therein) present proof of the existence of a ferromagnetic transition in the DMS $Pb_{1-x-y}Sn_yMn_xTe$, induced by strong exchange interaction of the Mn ions with the band holes. A ferromagnetic phase transition was also found in $p$-doped DMS quantum wells [2]. The carrier-induced ferromagnetism was observed in the DMS $(Ga_{1-x}Mn_x)As$ with $x$ of a few percent, where holes are associated with the Mn ions in these structures [3].

To describe the transition to a ferromagnetic (FM) phase induced by free carriers, all of the authors of the aforementioned works used similar approaches: the role of band carriers was reduced to induction of the indirect interaction between localized spins through electrons (holes) (LeL interaction), known in the physics of metals as the RKKY (Ruderman–Kittel–Kasuya–Yosida) interaction [4]. Then magnetic ions were considered as a subsystem separated from the electrons (holes) because of small free carrier contribution to the total magnetization of the system. The consideration of magnetic ions with the RKKY interaction in terms of the Curie–Weiss field permits one to incorporate in it the spin–spin interactions caused by other mechanisms (LL interaction). The latter mechanisms are assumed to result in a temperature shift $\Theta_{LL}$ in the Curie law for the magnetic susceptibility without free carriers, $\chi^{-1} \propto T - \Theta_{LL}$ . Thus, the electrons (holes) change this dependence by the additional shift $\Theta_{LeL}$ [1]:

$$\chi^{-1} \propto T - \Theta_{LL} - \Theta_{LeL}. \qquad (1)$$

Since for Mn-based DMS $\Theta_{LL} < 0$ while $\Theta_{LeL} > 0$, the FM phase transition can take place if $\Theta_{LeL} > |\Theta_{LL}|$.

Another approach to the problem under consideration was developed in Refs. 5,6, where the carrier–ion exchange interaction was treated in terms of a self-consistent mean field approximation (MFA). Indirect LeL interaction does not appear in this

approach, but the free-carrier contribution to the thermodynamic potentials was taken into account in the MFA. As a result, this theory predicts a temperature shift $\Theta_{MF}$ in the Curie law:

$$\chi^{-1} \propto T - \Theta_{LL} - \Theta_{MF}. \qquad (2)$$

Now we would like to emphasize that the values of temperature shifts $\Theta_{LeL}$ and $\Theta_{MF}$ calculated under some additional (but common enough) assumptions coincide with each other for $3D$, $2D$, and $1D$ electron gases [2]. This coincidence can lead to the spurious conclusion that the MFA and LeL (or RKKY) interaction considered in the Curie–Weiss field approximation are of an identical nature. This paper draws attention to the fact that free carriers still cause a significant contribution to the thermodynamic potentials even after the LeL interaction is taken into account. This contribution treated in the MFA is not identical to the LeL (or RKKY) interaction. This last statement becomes clear if we take into account that the LeL (RKKY) interaction appears after partial diagonalization of the interaction Hamiltonian with respect to space quantum numbers (the electron wave vectors $\mathbf{k}$ in the case of the RKKY interaction). While the MFA approach (Refs.5,6) uses only the diagonal part of the interaction and therefore does not depend on the spatial configuration of the magnetic ions. So, it is possible to imagine a physical situation in which the MFA contribution exceeds the RKKY contribution to the magnetic susceptibility. Thus the main result we want to prove below can be reduced to the statement that the static magnetic susceptibility depends significantly, generally speaking, on both $\Theta_{MF}$ and $\Theta_{LeL}$.

The structure of the paper is following. First we illustrate our approach to the model allowing exact statistical calculations. Then we consider more realistic system that is related to experimental situation in [1–3]. In conclusion we discuss the general approach to the FM phase transitions in the DMS and compare our approach with the works devoted to this problem.

## Magnetic susceptibility in exactly solvable model

The Hamiltonian of our model is similar to that applied in the aforementioned works and comprises a sum of localized spin moments (LSM), electrons, and their interaction Hamiltonians:

$$H = H_m + H_e + H_{em}, \qquad (3)$$

where

$$H_m = g_m \mu_B B \Sigma_j S_Z^j \equiv \omega_m M_L ;$$

$$H_e = \Sigma_{b,k,\sigma} (\varepsilon_b + \omega_e \sigma) \mathbf{a}^+_{b,k,\sigma} \mathbf{a}_{b,k,\sigma} ;$$

$$H_{em} = -(J/N_0) \Sigma_{b,b',k,\sigma} A_{b,b'} (\sigma M) \mathbf{a}^+_{b,k,\sigma} \mathbf{a}_{b',k,\sigma} .$$

Here $S_Z^j$ is the $Z$ component of the $j$th LSM spin, while $M_L = \Sigma_j S_Z^j$, $j = 1, \ldots, N_m$; $N_m$ is the number of LSMs in the system; $g_m$ and $\omega_m = g_m \mu_B B$ are, respectively, the LSM $g$ factor and Zeeman splitting in the field $B$; $\omega_e$ is band electron Zeeman splitting in the field $B$. Three quantum numbers can be attributed to electrons: band number $b$, intraband quantum number $k$, and projection of spin $\sigma = \pm 1/2$; $\mathbf{a}^+_{b,k,\sigma}$ and $\mathbf{a}_{b,k,\sigma}$ are the creation and annihilation operators; $J$ is the carrier–ion exchange interaction constant; the normalization factor $N_0$ equals one-half of the number of electronic states in each of bands $b$, and $A_{b,b'}$ is an interband transition matrix element.

The structure of Hamiltonian (3) is similar to that in [1–3,6]. The difference is both in the dispersions of band carriers, $\varepsilon_{b,k} = \varepsilon_{b',k}$, that corresponds to flat bands, and in the lack of intraband exchange scattering. The exchange scattering between bands $b$ and $b'$ is taken into account by the matrix element $A_{b,b'}$.

If we restrict ourselves to only two electronic bands $b = 1, 2$, the diagonalization of the Hamiltonian becomes trivial. Eigenenergies $E$ are defined by the repopulating of electrons within bands $b = 1$ and 2 as well as by normalized amount of ions spin projection $\mu = M_L/N_m$. For simplicity we assume $A_{b,b'} = 1$. Thus, the energy of unit volume reads:

$$E_b = n_b(\varepsilon_1 + \varepsilon_2)/2 + (G_L + \omega_e)(n_{b+} - n_{b-}) \pm$$

$$\pm (n_b \Delta E/2) \left[ 1 + \left( \frac{G_L}{\Delta E} \right)^2 \right]^{1/2} + \omega_m n_m \mu . \qquad (4)$$

The signs «–» and «+» correspond to $b = 1$ and $b = 2$; $\Delta E$ is the energy interval between these bands; $n_{b+}$ and $n_{b-}$ are concentrations of electrons with spin projection $\sigma = +1/2$ and $-1/2$ in the band $b$, the total electron concentration is $n_b = n_{b+} + n_{b-}$; $G_L = Jx\mu$ is an effective exchange field of magnetic ions acting on electrons, $x = N_m/N_0$ is the fraction of magnetic cations in the crystal, $n_m$ and $N_m$ are their concentration and total number, and $N_0$ is the total number of cations in the crystal.

Since value of $G_L$ is infinitesimal at $T > T_c$ ($T_c$ is the FM phase transition temperature) and $B \to 0$, the square root in Eq. (4) can be expanded in the small parameter $(G_L/\Delta E)^2$ up to the first nonvanishing term. We will also assume that only the lowest energy band $b = 1$ is filled, i.e., $\Delta E >> kT$. Then the energy spectrum assumes following form:

$$E = n_e\varepsilon_1 + n_e G_L \sigma_e + \omega_m n_m \mu - n_e G_L^2/(4\Delta E) \ . \quad (5)$$

For brevity, we introduce the total concentration of electrons $n_e = n_{b=1}$ and the average projection of electronic spins $\sigma_e = (n^+ - n^-)/2(n^+ + n^-)$; the electronic $g$ factor is assumed to be equal to zero.

There are two possible courses of further action. The first one (following Refs. 1–3) is to restrict consideration to the magnetic ions only. This restriction means consideration of only the last two terms in Eq. (5). The second way [5,6] is to consider only the first three terms. Note that last term in Eq. (5) is just the contribution to the energy from the LeL spin–spin interaction induced by the band electrons, because $G_L^2 \propto \Sigma_{jj'} S_Z^j S_Z^{j'}$ while all the rest of the terms are due to the electron and LSM energies with the diagonal part of their interaction. We consider a third approach that takes into account the full energy of the system (5).

For magnetic susceptibility calculations we need the partition function. This function has the form

$$Z = \iint U_{N_m}(M_L) U_{N_e}(M_B)\, \mathrm{e}^{-E/kT}\, dM_B dM_L \ , \quad (6)$$

and can be immediately calculated with the help of Eq. (5). The projections of the total LSM spins $M_L = N_m \mu$ and the band electrons $M_B = N_e\sigma_e$ are introduced in Eq. (6). Beyond magnetic saturation, the statistical weight $U_N(M)$ is given by a Gaussian distribution in the thermodynamic limit $N_m \to \infty$, $N_e \to \infty$ [7]:

$$U_N(M) = \frac{(2S+1)^N}{(\pi\Delta_S)^{1/2}}\, \mathrm{e}^{\frac{M^2}{\Delta_S}} \ , \quad (7)$$

where $\Delta_S = \frac{2}{3}\, S(S+1)N$. Equation (7) is also applicable for band electrons with $S = 1/2$ if the electrons obey Boltzmann statistics. Such an approach is evidently realized in the limit $N_e << N_0$. Thus, the partition function $Z$ is calculated by straightforward integration in Eq. (6) with the aforementioned assumptions. After some algebra we arrive at following final result:

$$\chi_0^{-1} = \chi_{0,L}^{-1} \left( 1 - \frac{\Theta_{LeL}}{T} - \frac{\Theta_{MF}^2}{T^2} \right)$$

with

$$\Theta_{LeL} = \frac{1}{6}\, S(S+1)\, \frac{J^2 x^2}{\Delta E}\, \frac{N_e}{N_m} \ ;$$
$$\Theta_{MF} = \left[ \frac{1}{12}\, S(S+1)\, J^2 \Omega_0^2 n_m n_e \right]^{1/2} \ , \quad (8)$$

where $\chi_{0,L} = \frac{2}{3}\, S(S+1)(g\mu_B)^2 n_m/T$ is the paramagnetic susceptibility of noninteracting LSM, and $\Omega_0$ is the volume of the unit cell.

Calculations in the framework of the approach of Refs. 1–3 reproduce the result (8) but without the last term $(\Theta_{MF}/T)^2$. This term appears as a contribution of free carriers to the thermodynamic potential due to diagonal part of the carrier–ion exchange interaction $H_{em}$ (3). One can see that the diagonalization procedure does not remove this term, and therefore the LeL interaction can never take it into account.

Another approach [5,6] treats the interaction $H_{em}$ in the first order of perturbation theory only. As a result, the last term in Eq. (5) does not appear, and therefore $\Theta_{LeL} = 0$ in Eq. (8) for our model. It is interesting to note that, in spite of the extreme simplicity of the model under consideration, the expression for $\Theta_{MF}$ in Eq. (8) reproduces the result of [6] obtained in terms of self-consistent exchange fields for a more realistic situation.

One can see that expression for $\Theta_{MF}$ (8) is neither quantitatively nor qualitatively similar to $\Theta_{LeL}$ . It permits one to make following general statement. For problems of magnetic phase transitions, magnetic susceptibility, magnetization, etc. it is important to simultaneously take into account the contributions of both the magnetic ions and electron subsystems to the thermodynamic potentials despite the negligible magnetization of the free carriers. In doing so, it is also important to take into account both the diagonal and off-diagonal parts of the carrier–ion exchange interaction. The omission of any of the aforementioned terms in the Hamiltonian leads, generally speaking, to significant inaccuracy or even to qualitative changes.

## General approach to calculation of crytical temperature

We present now the correct consideration of the problem of spontaneous magnetic transitions induced by band carriers in DMS. We choose the Hamiltonian in a form similar to (3) but incorporate the LL spin–spin interaction $H_{LL}$ between the LSM in the magnetic part $H_m$ and the intraband exchange scattering between Bloch electron states

in the interaction $H_{em}$ [8]. In such a case the intraband exchange scattering generates the LeL interaction. To calculate the magnetic susceptibility with the help of the modified Hamiltonian (3), we shall carry out the approximate diagonalization of Eq. (3) by elimination of its off-diagonal (in $\mathbf{k}$ and $\mathbf{k}'$) components by the canonical transformation method [9] in the second order of perturbation theory. As a result, the effective LeL spin–spin interaction operator assumes the form

$$H_{LeL} = \sum_{j,j'} J_{\text{eff}}(\mathbf{R}_{j,j'}) \mathbf{S}^j \mathbf{S}^{j'} , \qquad (9)$$

where $\mathbf{R}_{j,j'}$ is the radius vector joining the pairs of magnetic ions at the crystal lattice sites $j$ and $j'$. The structure of the indirect interaction (9) is similar to the Hamiltonian $H_{LL}$, so they can be joined. The specific form of $J_{\text{eff}}(\mathbf{R}_{j,j'})$ in Eq. (9) depends on the degeneracy of the electron gas [10], the influence of magnetic field [11], the effect of casual anisotropy [12], the structure of the energy band of the semiconductor [1], and the dimensionality of the system [2].

Then, the diagonal part of the operator $H_{em}$ should be written in the form of the Zeeman energy of the LSM in the effective field $B_e = J\Omega_0 \sigma_e / g_m \mu_B$ and added to the Zeeman term of the magnetic Hamiltonian $H_m$. We should emphasize here that $B_e$ is similar to the Knight field in nuclear magnetism. The LSM $g$-factor shift induced by the field $B_e$ has recently been observed in DMS [13].

One more standard step is the transformation of the spin–spin interactions in $H_m$ to the energy in the Curie—Weiss fields. It is well known that such an approach reduces the thermodynamical treatment of interacting spins to consideration of isolated spins with an effective temperature $T_{\text{eff}} = T - \Theta$. The parameter $\Theta = \Theta_{LL} + \Theta_{LeL}$ is determined by both the LL and the LeL interactions (Eq. (9)).

As a result, the free energy can be presented in terms of the electronic and ionic parts only [14]:

$$F = F_e(\sigma_e) + F_m(B + J\Omega_0 \sigma_e / g_m \mu_B , T - \Theta) , \qquad (10)$$

where $F_m(B + B_e , T - \Theta)$ is the contribution of noninteracting (isolated) spins subjected to the uniform magnetic field $B + B_e$ at the temperature $T - \Theta$. Note that Eq. (10) takes into account both diagonal (the term $B_e = J\Omega_0 \sigma_e / g_m \mu_B$) and off-diagonal (the term $\Theta_{LeL}$) parts of the carrier–ion exchange interaction. The electronic spin polarization $\sigma_e$ is found by minimization of the functional (10). Further substitution of $\sigma_e$ obtained in such a manner to Eq. (10) defines completely the thermodynamic characteristics of the system: the magnetization $M_\alpha = -\partial F / \partial B_\alpha$; magnetic susceptibility $\chi_{\alpha,\alpha} = -\partial^2 F / \partial B_\alpha^2$, $\alpha = x, y, z$; and the temperature of the magnetic phase transition $T_c$.

The specific form of the free energy functional (10) depends on the aforementioned and many other peculiarities of our system. As an illustration, we consider now the most popular case of a degenerate electron gas in a simple isotropic band of a semiconductor. We consider the magnetic transition temperature $T_c$ on the basis of the previous results [1–3,6]. Equation (10) permits one to obtain the following equation for the critical temperature point:

$$(T_{\text{eff}})_c - \Theta_{MF} = 0 . \qquad (11)$$

Here $\Theta_{MF}$ is given by the corresponding formulas of [2,6], and $(T_{\text{eff}})_c = T_c - \Theta_{LL} - \Theta_{RKKY}$, where $\Theta_{LeL} = \Theta_{RKKY}$ for the specific case considered. The parameter $\Theta_{RKKY}$ coincides with $\Theta_{MF}$ only under the assumptions mentioned in the introduction to this paper. Parameter $\Theta_{LL}$ should be taken from the experiment, $\Theta_{LL} = -T_0$, where $T_0 > 0$ corresponds to the antiferromagnetic LL exchange interaction realized in the majority of experimental situations for DMS (see Ref. 15 and references therein). We can thus obtain $T_c = 2\Theta_{MF} - T_0$. If one takes into account only self-consistent exchange mean fields or RKKY interactions, the value of $T_c$ is determined by a different expression: $T_c = \Theta_{MF} - T_0$. This difference can be important in the prediction of conditions for the realization of carrier-induced ferromagnetism in different experimental situations.

## Conclusion

We have shown that neglecting the electronic contribution to the free energy (10) leads to a substantial lowering of the predicted $T_c$ value despite the consideration of the indirect interaction via free carriers. Moreover, the example considered shows that such neglect can lead to qualitatively different results in some cases. The present work also shows that different parts of interaction operator are responsible for different mechanisms of FM ordering in DMS. Therefore, both diagonal and off-diagonal (in $\mathbf{k}$) parts of the carrier–ion exchange interaction are important. Nevertheless, the main conclusion of previous works remains valid: carrier-induced FM transition in DMS is possible under high enough carrier concentrations, and reduction of the dimensionality of the system enhances this effect.

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