MECHANISMS OF CARRIER-INDUCED FERROMAGNETISM IN DILUTED MAGNETIC SEMICONDUCTORS

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(January 5, 2022)

Two different approaches to the problem of carrier-induced ferromagnetism in the system of the disordered magnetic ions, one bases on self-consistent procedure for the exchange mean fields, other one bases on the RKKY interaction, used in present literature as the alternative approximations is analyzed. Our calculations in the framework of exactly solvable model show that two different contributions to the magnetic characteristics of the system represent these approaches. One stems from the diagonal part of carrier-ion exchange interaction that corresponds to mean field approximation. Other one stems from the off-diagonal part that describes the interaction between ion spins via free carriers. These two contributions can be responsible for the different magnetic properties, so aforementioned approaches are complementary, not alternative. A general approach is proposed and compared with different approximations to the problem under consideration.

One of the fruitful ideas of microscopic theory of magnetism is to separate the electrons with non-compensated spins into those localized in the crystalline lattice cites and delocalized (band) ones. The exchange interaction between localized and delocalized electrons is then represented as a spin density of band electrons at the cites occupied by magnetic ions. Vonsovskii realized this idea in and calculated band electron energy shift due to $s-d$-exchange interaction. The Hamiltonian of $s-d$-exchange interaction was further used to study the ferromagnetic metals ($\text{FM}$), superfine interactions in the solid solutions of magnetic metals ($\text{SSIM}$), and magnetic impurities in non-magnetic metals ($\text{SINM}$). The detailed review of this material has been made in. Let us note some specific features of the carrier-ion exchange interaction. Its diagonal part is represented by the exchange integral $J(\vec{k}, \vec{k}' = \vec{k})$ over the Bloch functions with wave vectors $\vec{k}, \vec{k}' = \vec{k}$ and describes so-called magnetizing effect (or effect of redistribution of electronic population (repopulation effect)). The latter effect is indeed the variation of total spin of the band carriers in the effective exchange field $\vec{G}_L$ created by localized spin moments (LSM). The value of this variation is defined by Pauli susceptibility with field $\vec{G}_L$. We emphasize that the effect of giant spin splitting (GSS) of band states of $\text{FM}$, discovered in diluted magnetic semiconductors (DMS) has similar nature. From the standpoint of, the repopulation effect in metals may be considered as a redistribution the band electrons populations between spin split subbands in the same way as a single Fermi level is established for electrons with opposite spin orientations. The off-diagonal in $\vec{k}$ part of the carrier-ion exchange interactions with the $J(\vec{k}, \vec{k}' = \vec{k})$ gives rise to the spin density oscillations. These oscillations, however, lead to indirect exchange interaction between the LSMs, known in metal physics as RKKY interaction.

Effective exchange field $\vec{G}_L$ is proportional to LSM magnetization $\vec{M}$. Latter, in turn, is defined by the sum of external magnetic field $\vec{B}$ and effective exchange field $\vec{B}_e$, which acts on the LSM by the polarized electron spins. The LSM magnetization $\vec{M}$ also depends on spin-spin interactions between LSM. Below we will present the arguments that both direct spin-spin interaction which is independent on the free carriers (LL-interactions) and indirect one induced by the free carriers (LeL -interactions) contribute to $\vec{M}$. In turn, the effective field of carriers $\vec{B}_e$ depends on the quantity $\vec{G}_L + g_e \mu_B \vec{B}$, determined by electronic spin polarization, where $g_e$ is electron g-factor, $\mu_B$ is Bohr magneton. So, the effective exchange field $\vec{G}_L$ is expressed through itself in a self consistent manner. Generally speaking, such self consistency increases the magnetic susceptibility $\chi = Const(1 - \Theta)^{-1}$. Thus one can expect a ferromagnetic phase transition if $\Theta > 0$. A possibility of the appearance of dopant-induced ferromagnetism in semiconductors due to exchange interaction of the band and localized electrons was studied for the case of semiconductor with bivalent shallow impurities which reveal both magnetic and electric properties. Later the expressions for critical temperatures of ferromagnetic transition in DMS with deep magnetic ion levels were obtained in. It should be noted that self consistent procedure was essential for determination of the $\vec{B}_e$ and $\vec{B}_L$ exchange fields in both aforementioned works.

In recent years, it has been substantial increase of interest to studies of the carriers induced ferromagnetism in the DMS. A number of works (see and references therein) were devoted to the proof of the existence of fer-
romagnetic transition in the DMS \( Pb_{1-x-y}Sn_xMn_yTe \), induced by strong exchange interaction of the Mn ions with the band holes with wide range (up to \( 2 \cdot 10^2 \text{cm}^{-3} \)) of concentrations. Ferromagnetism of the Mn ions was found to be due to their interaction with band holes confined in two-dimensional quantum wells on the base of DMS \( Cd_{1-x}Mn_xTe \). The carrier-induced ferromagnetism was also observed in the structures \( A_1^p \_xMn_yB^5 \) with \( x \) about few percents. The holes in these structures are also associated with the Mn ions (\( \Theta \)).

However, the approach used in the aforementioned works (\( [17, 20, 21] \)) was different from that of \( [18, 19] \). Namely, the role of the band carriers were reduced to induction of the RKKY interaction only. So, self-consistent contribution of diagonal part of carrier-ion exchange interaction was not taken into account.

Note, that authors of Ref. \( [22] \) carried out a special analysis of magnetic susceptibility peculiarities in the assumption that role of carrier-ion interaction is reduced to the repopulation effect only. In the spirit of the works \( [22, 24] \) they used the self consistent procedure for the exchange fields \( \tilde{G}_L \) and \( \tilde{B}_c \). The ferromagnetic phase transition temperature \( \Theta_{MF} \), obtained by this procedure was shown to coincide with transition temperature \( \Theta_{RKKY} \), calculated with the help of RKKY interaction, considered as the sole reason for carrier-induced ferromagnetism. Such coincidence can be obtained only under following additional assumptions: (i) RKKY interaction can be represented in terms of Curie-Weiss field, (ii) the magnetic ions spatial distribution corresponds to the ideal gas of particles. The obtained coincidence \( \Theta_{RKKY} = \Theta_{MF} \) may cause an illusion of equivalence and interchangeability of these two approaches.

It already follows from aforementioned discussion that the equality \( \Theta_{RKKY} = \Theta_{MF} \) is not strictly asserted for real systems because at least the spatial distribution of magnetic ions corresponds to the lattice gas rather then to ideal gas. Moreover, it is possible to imagine a situation when the non random spatial distribution results in antiferromagnetic transition due to oscillating nature of RKKY interaction with \( \Theta_{RKKY} < 0 \) while inequality \( \Theta_{MF} > 0 \) is true for any spatial distribution of the magnetic ions. Formally speaking, the equality \( \Theta_{RKKY} = \Theta_{MF} \) by itself cannot be used as an evidence of the statement that either self-consistent exchange fields consideration or carriers-induced LeL interaction are the equivalent descriptions of the same interactions. This stems from the aforementioned fact that they are described by different parts of Hamiltonian. Thus, both self-consistent mean exchange fields, leading to the repopulation effect, and Curie-Weiss field stemming from the carriers-induced spin-spin interaction, have to be taken into account simultaneously (\( \Theta \)). In spite of the fact, that latter conclusion follows both from the analysis of classical works (\( \Theta \)) and from detailed discussion of the differences between manifestations of diagonal and off-diagonal parts of interaction (\( \Theta \)), it is not completely clear if it is applicable to the case of carrier-induced ferromagnetism in DMS. In our view this question can be clarified by the calculation of phase transition temperature \( T_c \) from the first principles by means of exactly solvable model. In this case, we avoid the auxiliary models for self-consistent fields \( \tilde{G}_L, \tilde{B}_c \) or Curie-Weiss field of LeL-interacting LSM. We also avoid the discussion of the equivalence or complimentarity of the approaches under consideration.

It is impossible to calculate exact value of \( T_c \) in real experimental case (\( \Theta \)). However, this is not necessary since our aim is to clarify the relative role of aforementioned mechanisms of magnetic phase transitions. Below we give an illustrative example of exact solution for the magnetic phase transition temperature \( T_c \). This example allows also to separate contributions from self-consistent fields \( \tilde{G}_L \) and \( \tilde{B}_c \) and carriers-induced LeL spin-spin interaction.

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

\[
\mathcal{H} = \mathcal{H}_m + \mathcal{H}_e + \mathcal{H}_{em},
\]

where

\[
\mathcal{H}_m = g_m \mu_B B \sum_j S_j^z \equiv \omega_m M_L,
\]

\[
\mathcal{H}_e = \sum_{b,k,\sigma} (\epsilon_b + \omega_e \sigma) a_\dagger b, k, \sigma a_{b,k,\sigma},
\]

\[
\mathcal{H}_{em} = -\frac{J}{N_0} \sum_{b,b',k,\sigma} A_{b,b'} (\sigma M) a_\dagger b, k, \sigma a_{b', k, \sigma}.
\]

Here \( S_j^z \) is \( z \)-component of \( j \)-th LSM spin while \( M_L = \sum_j S_j^z \), \( j = 1...N_m \). \( M_L \) is the number of LSM in the system, \( g_m \) is the LSM g-factor, \( \omega_m = g_m \mu_B B \) is LSM Zeeman splitting in the field \( B \) and \( \omega_e \) is delocalized electrons 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 \( \sigma = \pm 1/2 \); \( a_{b,k,\sigma}^\dagger \) and \( a_{b,k,\sigma} \) are the creation and annihilation operators; \( J \) is a constant of carrier-ion exchange interaction; normalization factor \( N_0 \) corresponds to one half of the number of electronic states in each of bands \( b \); \( A_{b,b'} (\sigma M) \) is an interband transition matrix element.

The structure of Hamiltonian (\( \Theta \)) is similar to that in (\( \Theta \)). The difference is both in the dispersions of band carriers, \( \epsilon_{b,k} = \epsilon_b \), that formally corresponds to flat bands, and in the lack of intraband exchange scattering. The exchange scattering between bands \( b \) and \( b' \) with electron spin flip 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. Eigenergery \( E \) is defined by the redistribution of electrons (with spins projections \( \sigma \)) within bands \( b = 1 \)
and 2 as well as by the magnetic ions (with spin projections $S^b_z$) distribution or, more precisely, by normalized amount of magnetic ions $\mu = M_L/N_m$. For simplicity we assume $A_{b,b'} = 1$. Thus, the energy of unit volume reads:

$$E_b = \frac{1}{2} n_b \Delta E + (G_L + \omega_c) (n_{b+} + n_{b-}) \pm \frac{1}{2} n_b \Delta E \sqrt{1 + \left( \frac{G_L}{\Delta E} \right)^2 + \omega_m n_m \mu}. \quad (2a)$$

The signs "−" or "−+" before the square root sign in the equation (2a) correspond to $b = 1$ or $b = 2$, $\Delta E$ is an energy interval between these bands, the $n_{b+}$ and $n_{b-}$ corresponds to concentration of electrons with the spin projection $\sigma = +1/2$ and $-1/2$ in the band $b$, the total concentration is $n_b = n_{b+} + n_{b-}$, $G_L = J x \mu$, $x = N_m/N_0$ being a fraction of magnetic cations in the crystal, $N_m$ is their concentration.

Since value of $G_L$ is infinitesimal at $T > T_c$, the square root in expression (2a) can be expanded over the small parameter $(G_L/\Delta E)^2$ up to first nonvanishing term. This term is proportional to $\mu^2 = (N_m)^{-2} \sum_{j,j'} S^j_z S^{j'}_z$. Therefore, it can be considered as a contribution to the energy from LeL spin-spin interaction induced by the band electrons. We will also assume, that the only lowest energy band $b = 1$ is filled, i.e. $E \gg kT$. Then, the energy spectrum assumes following form:

$$E = e_n J x \mu \sigma_e - e_n \left( \frac{G_L}{2 \Delta E} \right)^2 + \omega_m n_m \mu. \quad (2b)$$

For brevity, we introduce a total concentration of electrons $n_e = n_{b=1}$ and an average projection of electronic spins $\sigma_e = (n^+ + n^-)/(n^+ + n^-)$. The first term in Eq.(2b) corresponds to diagonal part of interaction $\delta_{ij}$ while the second one represents the contribution from off-diagonal (with respect to quantum number $b$) part; the electronic $g$-factor is assumed to be equal to zero.

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

$$Z = \int \int U_{N_m} (M_L) U_{N_e} (M_B) \exp \left( -\frac{E}{kT} \right) dM_L dM_B \quad (3)$$

and can be immediately calculated with the help of Eq. (2b). The full projections (ne yasno) of LSM $M_L = N_m \mu$ and the band electrons $M_B = N_e \sigma_e$ are introduced in Eq. (2b). Beyond magnetic saturation, the statistical weights $U_N (M)$ are defined by Gaussian distribution in thermodynamic limit ($N_m \rightarrow \infty$, $N_e \rightarrow \infty$) [23]:

$$U_N (M) = \frac{(2S + 1)^N}{\sqrt{\pi} \Delta S} \exp \left( -\frac{M^2}{\Delta S} \right), \quad (4)$$

where $\Delta S = (2/3) S(S+1) N$, $S$ is the LSM value. Eq.(4) is also applicable for band electrons with $S=1/2$ if electrons obey a Boltzmann statistics. Such approach is evidently realized in the limit $N_e \ll N_0$. Thus, the partition function $Z$ and hence the magnetic susceptibility is calculated by straightforward integration in Eq. (3) with aforementioned assumptions. After some algebra we arrive at following final result:

$$\chi_0 = \frac{\chi_0 L}{1 - \frac{2}{3} S(S+1) \frac{J^2 \Delta E N_m}{N_0} - \frac{2}{3} S(S+1) \frac{J^2 \Delta E N_m}{N_0}} \quad (5)$$

where $\chi_0 L = \frac{2}{3} S(S+1) \frac{n_m}{N}$ is a paramagnetic susceptibility of noninteracting LSSM with concentration $n_m$.

Expression (5) allows an easy interpretation. In the case of band electrons absence, $N_e = 0$, the $\chi_0$ describes an ideal paramagnet. If the electrons are present, the interband exchange scattering influence can be excluded from consideration as $\Delta E \rightarrow \infty$. Clearly, this corresponds to taking into account the self-consistent exchange fields $G_L$ and $B_z$ only. General Eq. (5) gives for this case

$$T_c = \Theta_{MF} = \sqrt{\frac{1}{12} S(S+1) J^2 \Omega^2 n_m n_e}, \quad (6)$$

where $\Theta_0$ is a crystal unit cell volume. It is interesting to note that in spite of the extreme simplicity of the model under consideration, Eq. (5) reproduces a result of the obtained in terms of self-consistent exchange fields for more realistic situation. It allows to consider the third term in the denominator of Eq. (5) as a contribution from self-consistent exchange fields $G_e$ and $G_L$.

As it was mentioned above, the second term is due to off-diagonal part of carrier-ion exchange Hamiltonian. With respect to this part only, we have to omit the last term in denominators of Eq. (5). In this case we obtain

$$T_c = \Theta_{ind} = \frac{1}{6} S(S+1) J^2 \Omega^2 n_m n_e \frac{n_m}{\Delta E} \quad (7)$$

It is seen that Eq. (7) is not similar to Eq. (5), neither quantitatively nor qualitatively. It gives possibility to make a following general statement. For the problems of magnetic phase transitions, as well as for the calculations of magnetic susceptibility, magnetization, etc. it is important to simultaneously take into account both parts (diagonal and off-diagonal) of carrier-ion exchange interaction. Therefore, the omission of any one of the aforementioned terms in the Hamiltonian leads, generally speaking, to significant inaccuracy or even to qualitative changes.

We shall present now the way of consideration of the problem of spontaneous magnetic transitions induced by band carriers in DMS, which we consider to be correct. We choose the Hamiltonian in the form (1), but will incorporate there the LL spin-spin interaction between LSM $H_{LL}$ to the $H_m$ and the intraband exchange scattering between Bloch electron states $\pi$.

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To calculate the magnetic susceptibility with the help of modified Hamiltonian (1), we shall carry out the approximate diagonalization of Eq. (1) by elimination of its off-diagonal (in $\bar{k}$ and $\bar{p}$) components by canonical transformation method in second order of perturbation theory. As a result, the operator of effective LeL spin-spin interaction assumes the form:

$$\mathcal{H}_{LeL} = \sum_{j,j'} J_{eff} \left( \bar{R}_{j,j'} \right) \bar{S}^j \bar{S}^{j'},$$

where $\bar{R}_{j,j'}$ is a radius-vector joining the pairs of magnetic ions in the crystal lattice sites $j$ and $j'$. The structure of $\mathcal{H}_{LeL}$ is similar to the $\mathcal{H}_{LL}$ magnetic Hamiltonian $\mathcal{H}_m$ and can be added to it. The specific form of $J_{eff} \left( \bar{R}_{j,j'} \right)$ is defined by electronic gas degeneration, influence of magnetic field, effect to casual anisotropy, structure of energy band of semiconductor, and dimensionality of the system.

The diagonal part of the operator $\mathcal{H}_m$ can be written down in the form of LSM’s Zeeman energy in the effective field $B_e = J_{eff} \mu_0 \sigma_e g_m \mu_B$. This part can be added to the Zeeman term of magnetic Hamiltonian $\mathcal{H}_m$. One more standard step is the transformation of spin-spin interactions in $\mathcal{H}_m$ and $\mathcal{H}_{LeL}$ to Curie-Weiss fields. Such approach, as it is well known, reduces the thermodynamical treatment of interacting spin to the consideration of isolated spins with the effective temperature $T_{eff} = T - \Theta$. The parameter $\Theta = \Theta_{LL} + \Theta_{LeL}$ is defined by both LL interaction and LeL interaction (Eq. (8)).

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

$$F = F_e (\sigma_e) + F_m (B + B_e, T - \Theta),$$

where $F_m (B + B_e, T - \Theta)$ is a contribution of noninteracting (isolated) spins subjected to the uniform magnetic field $B + B_e$ at the temperature $T - \Theta$. Note that the Eq. (8) takes into account both diagonal part of carrier-ion exchange interaction (term $B_e$) and its off-diagonal part (term $\Theta_{ind}$). The electronic polarization $\sigma_e$ is calculated by minimization of functional $\mathcal{H}_e$. Further substitution of $\sigma_e$ obtained in such manner to Eq. (8) defines completely thermodynamic characteristics of the system: magnetization $M_\alpha = -\partial F / \partial B_\alpha$, susceptibility $\chi_{\alpha\beta} = -\partial^2 F / \partial B_\alpha \partial B_\beta$, $\alpha, \beta = x, y, z$ and the temperature of magnetic phase transition.

Specific form of free energy functional (8) depends on aforementioned and many other peculiarities of our system. As an illustration, we consider now the most popular case of degenerated electronic gas in a simple isotropic band of semiconductor. We consider the magnetic transition temperature $T_c$ based on the preceding results (Eq. (9)). The Eq. (8) permits to obtain following equation for the critical temperature point:

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

Here $\Theta_{MF}$ is defined by corresponding formulas of the $\Theta_{LL}$, where only the diagonal part of the interaction operator $\mathcal{H}_{m}$ is taken into account and $(T_{eff})_c = T_c - \Theta_{LL} - \Theta_{RKKY}$. Parameter $\Theta_{RKKY}$ coincides with the $\Theta_{MF}$ only under the conditions, 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 antiferromagnetic LL exchange interaction realized in majority of experimental situations for DMS (Ref. [15,19,20]). So, for $T_c$ we can 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 other expression: $T_c = \Theta_{MF} - T_0$. This difference can be important to the prediction of conditions for carriers-induced ferromagnetism realization in different experimental situations.

We had shown that neglecting of any part of interactions $\mathcal{H}_{em}$ leads to essential reduction of predicted $T_c$ value even in the simplest models. Moreover, considered example shows that such neglecting leads to the qualitatively different results. Present work shows that both diagonal and off-diagonal in $\bar{k}$ parts of carrier-ion exchange interaction are important. Their simultaneous consideration is shown to change the part of earlier results quantitatively or even qualitatively. Nevertheless, main conclusion of preceding works remains valid: carrier induced ferromagnetic transition in DMS is possible under high enough carriers concentrations and reduction of the system dimensionality enhances this effect.

Let us finally note that we have considered the necessity of simultaneous consideration of both contributions of carrier-ion exchange interaction. One can see that effect is essential. It is clear, that similar approach have to be applied to many other cases. For instance, it would be useful to take into account the LeL exchange interaction of the LSM via conduction electrons in the problem of free or bound magnetic polaron.

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22 Not for all situations, however, this is plainly formulated. Sometimes, consideration of no more than one part of a carrier-ion exchange interaction (12–21) leads to qualitatively correct but not quantitatively exact results. In a number of cases, the corresponding corrections are important.

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