Suppression of carrier induced ferromagnetism by composition and spin fluctuations in diluted magnetic semiconductors

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We suggest an approach to account for spatial (composition) and thermal fluctuations in "disordered" magnetic models (e.g. Heisenberg, Ising) with given spatial dependence of magnetic spin-spin interaction. Our approach is based on introduction of fluctuating molecular field (rather than mean field) acting between the spins. The distribution function of the above field is derived self-consistently. In general case this function is not Gaussian, latter asymptotics occurs only at sufficiently large spins (magnetic ions) concentrations n. Our approach permits to derive the equation for a critical temperature Tc of ferromagnetic phase transition with respect to the above fluctuations. We apply our theory to the analysis of influence of composition fluctuations on Tc in diluted magnetic semiconductors (DMS) with RKKY indirect spin-spin interaction. Two new results are obtained. (i) It is shown that composition fluctuations destroy ferromagnetic ordering in DMS if the carrier concentration nc exceeds the critical value nc ≈ 0.1ni.(ii) In the case nc < 0.5ni and magnetic ion spin S = 5/2 the composition fluctuations shift the critical temperature Tc (as compared to its mean field value) by a factor about 1 – 5nc/ni.

Ferromagnetic (FM) ordering in p-doped diluted magnetic semiconductors (DMS) attracts much attention of both experimentalists [1]-[2] and theorists [3]-[4]. Common approach to this problem is a mean field approximation (MFA), corresponding to some mean overall (homogeneous) carrier and magnetic ion magnetization. Once appeared, the exchange fields of carriers and magnetic ions lead to mutual spin splitting that can be stable at low enough temperatures T < Tc [8]. The problem of "high temperature ferromagnetism" in DMS is now very important from the point of view of technical applications. So, the question how to increase the critical temperature Tc has been widely discussed in recent publications (see [9] and references therein). One way to control Tc is to vary the concentrations of magnetic ions and/or carriers. In the case of degenerate carriers, the critical temperature Tc of FM phase transition in the bulk DMS is proportional to n1n2/3 [3], where n1 and n2 are the magnetic ion and carrier concentrations respectively.

Beyond the MFA, we should consider the Friedel oscillations of carrier spin polarization [9] occurring at the scale of inverse Fermi wave vector 1/kF. In this case, the fluctuations of inter-magnetic ion distance r_i,j should be taken into account, if the nearest neighbor mean value r is about 1/kF. Really, in the case of k_F r ≈ 1 the numbers of magnetic ion pairs with FM and AFM RKKY interaction is comparable, which can lead to suppression or complete destruction of long range FM order.

Note, that the limitation of MFA by inequality k_F r << 1 was pointed out earlier [1], but quantitative contribution of disorder in the magnetic ion subsystem has not been considered till present. Moreover, the problem of the existence or non-existence of a finite critical temperature Tc > 0 at arbitrary relation between n_i and n_e has not been resolved also.

Here we develop a theory of magnetic ordering of disordered magnetic system with given spin-spin interaction in terms of fluctuating local molecular field approximation (FFA). The method we propose here is applied to calculations of Tc caused by RKKY interaction.

The Hamiltonian of the problem reads

\[ \mathcal{H} = \sum_{j<j'} J(\vec{r}_{j,j'}) \vec{S}_j \cdot \vec{S}_{j'} + \sum_j \vec{H}_0 \vec{S}_j, \]  

(1)

where magnetic field \( \vec{H}_0 \) and interaction \( J(\vec{r}_{j,j'}) \) is measured in energy units (i.e. \( g \mu = 1, \mu \) is Bohr magneton). The Hamiltonian (1) incorporates two "sources of randomness". First, (the spatial disorder) is that spin can be randomly present or absent in the specific j-th cite of a host semiconductor. Second, (the thermal disorder) is a random projection of a spin (if any) in j-th cite.

Note, that our general formalism is valid for any form of \( J(\vec{r}_{j,j'}) \) so that its specific form will be chosen later.

In a mean field approximation, the Hamiltonian (1) reduces to the sum of Zeeman energies

\[ \mathcal{H} = \sum_i (\vec{H}_i + \vec{H}_0) \vec{S}_i, \]  

(2)
in the external magnetic field $\vec{H}_0$ and local molecular field

$$\vec{H}_i = \sum_{j \neq i} J(\vec{r}_{ij}) \left\langle \vec{S}_j \right\rangle,$$

(3)

where $\left\langle \vec{S}_j \right\rangle$ is a thermal average at a site $j$. Next step of the MFA is to substitute the molecular field (3) by the mean field

$$\vec{H}_{mf} = \sum_{j \neq i} J(\vec{r}_{ij}) \left\langle \vec{S}_j \right\rangle.$$

(4)

The bar means the averaging over spatial disorder, so the $\vec{H}_{mf}$ is homogeneous over crystal volume; $\left\langle \vec{S} \right\rangle$ is an average spin in this mean field,

$$\left\langle \vec{S} \right\rangle = \frac{\text{Tr} \{ \vec{S} e^{-\beta(\vec{H}_{mf} + \vec{H}_0)\vec{S}} \}}{\text{Tr} e^{-\beta(\vec{H}_{mf} + \vec{H}_0)\vec{S}}}.$$

(5)

$\beta = 1/T$. The system of Eqs (4) and (5) gives well-known solution for mean fields $\vec{H}_{mf}$ and magnetization $\vec{M} = n g \mu (\vec{S})$.

This work draws attention that the spatial and thermal fluctuations can be taken into consideration by introduction of random field rather than mean field. In our approach, we consider every spin $\vec{S}_j$ as a source of fluctuating (random) field

$$\vec{H}_f = J(\vec{r}_i - \vec{r}_j) \vec{S}_j$$

(6)

affecting other spins at the sites $\vec{r}_i$. In other words, every spin in our approach is subjected to some random (rather than mean) field, created by the rest of spin ensemble. So, all thermodynamic properties of the system will be determined by the distribution function $f(\vec{H}_f)$ of the random field $\vec{H}_f$. Namely, any spin dependent macroscopic quantity (like magnetization) $<< \vec{A} >>$ reads

$$<< \vec{A} >> = \int < A > \vec{H}_f f(\vec{H}_f) d\vec{H}_f,$$

(7)

where $< A > \vec{H}_f$ is single particle thermal average with effective Hamiltonian (2), where $\vec{H}_i$ is substituted by $\vec{H}_f$. The distribution function $f(\vec{H}_f)$ is defined as

$$f(\vec{H}_f) = \left\langle \delta \left( \vec{H}_f - \sum_{j \neq i} J(\vec{r}_i - \vec{r}_j) \vec{S}_j - \vec{H}_0 \right) \right\rangle.$$

(8)

Our FFA approach is based on micro-canonical statistical theory of magnetic resonance line shape [8]. Latter theory assumes the additivity of local molecular field contributions as well as the non-correlative spatial distributions of spins (magnetic ions).

Latter assumptions with respect to spectral representation of $\delta$ function permit to transform the Eq. (3) to the self-consistent integral equation for $f(\vec{H}_f) \equiv f(\vec{H})$.

Introducing the probability $n_i(\vec{r})d^3r$ for small volume $d^3r$ to be occupied by a particle, we obtain

$$f(\vec{H}) = \int \exp \left[ i \vec{r}(\vec{H} - \vec{H}_0) \right] \exp \left( \int_V << -iJ(\vec{r})\vec{S}_f > - 1 >> n_i(\vec{r}) d^3r \right) \left( \frac{d^3r}{2\pi} \right)^3,$$

(9)

where according to definition (3)

$$<< -iJ(\vec{r})\vec{S}_f > - 1 >> = \int_{-\infty}^{\infty} \frac{\text{Tr} \left\{ \exp \left[ -iJ(\vec{r})\vec{S}_f \right] \exp( -\beta(\vec{H} + \vec{H}_0)\vec{S}) \right\} f(\vec{H}) d^3H - 1}{\text{Tr} \left\{ \exp \left[ -iJ(\vec{r})\vec{S}_f \right] \right\}}.$$

(10)

Eqs (3) and (10) represent the integral equation for distribution function $f(\vec{H})$. This equation can be solved iteratively.

However, in many cases it is possible to avoid the solution of the integral equation since in these cases it is exactly reducible to the set of transcendental equations for macroscopic quantities like $<< \vec{S} >>$ (magnetization),..., $<< \vec{S}_n >>$, $n > 1$ of the system.

This approach can be most easily demonstrated for the Ising case in Hamiltonian (2). Although this case is less general than Heisenberg one, it is perfectly valid either for the case of hole induced ferromagnetism in quantum wells or in the case of host semiconductors with uniaxial symmetry.

If the magnetic fields are directed along OZ axis, the scalar product reduces to $\vec{S}_f = S_{Z \tau} = m \tau$, $m = -S, ..., S$ and (2) takes the form

$$f(\vec{H}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{iH\tau} \mathcal{G}(\tau) d\tau,$$

(11)

$$\mathcal{G}(\tau) = \left\langle \left\langle \int_V n_i(\vec{r}) \left( e^{-iJ(\vec{r})m \tau} - 1 \right) d^3r \right\rangle \right\rangle.$$

(12)

The calculation of average in (12) yields
In this case Eq. (12) assumes the form
\[ G(\tau) = \mathcal{G}\{\{a_m, b_m\}, \tau\} = \sum_{m=1/2}^{S} \{a_m\mathcal{F}_1(m\tau) + ib_m\mathcal{F}_2(m\tau)\}; \] (14)
\[ \mathcal{F}_1(m\tau) = \int_{V} n_i(\vec{r})[\cos(J(\vec{r})m\tau) - 1]d^3r, \]
\[ \mathcal{F}_2(m\tau) = \int_{V} n_i(\vec{r})\sin(J(\vec{r})m\tau)d^3r. \] (15)

Let us return to Eq. (9) for \( f(H) \). Multiplying it by \( A_m(\beta H) \) and \( B_m(\beta H) \) and integrating over \( H \), we obtain the closed system of nonlinear equations for the numbers \( a_m \) and \( b_m \). The explicit form of this system reads
\[ a_m = \int_{-\infty}^{\infty} A_m(\tau, \beta) \exp \left\{ \sum_{m'=1/2}^{S} a_{m'} \mathcal{F}_1(m'\tau) \right\} \cos \left( \sum_{m'=1/2}^{S} b_{m'} \mathcal{F}_2(m'\tau) \right) d\tau; \]
\[ b_m = \int_{-\infty}^{\infty} B_m(\tau, \beta) \exp \left\{ \sum_{m'=1/2}^{S} a_{m'} \mathcal{F}_1(m'\tau) \right\} \sin \left( \sum_{m'=1/2}^{S} b_{m'} \mathcal{F}_2(m'\tau) \right) d\tau, \] (16)
\[ A_m(\tau, \beta) = \frac{1}{\pi} \int_{0}^{\infty} A_m(\beta H) \cos(H\tau)dH; \ B_m(\tau, \beta) = \frac{1}{\pi} \int_{0}^{\infty} B_m(\beta H) \sin(H\tau)dH. \] (17)

To obtain (16), we substitute it into Eq. (14). Solutions of the Eqs (16) allow to obtain magnetization of the system \( M = g_\mu \mathcal{M}_\mathcal{I} \), where \( \mathcal{M}_\mathcal{I} = -\langle S_Z \rangle = -\sum_{m=1/2}^{S} mb_m \) as well as other spin averages
\[ M_k = \langle S^k \rangle = (-1)^k \sum_{m=1/2}^{S} m^k \{a_m; \text{for even } k, \ b_m; \text{for odd } k. \] (18)

The critical temperature \( T_c \) is usually defined as a temperature, where nonzero infinitesimal magnetization appears. In terms of Eqs (16) this means that we can put \( b_m = 0 \) for \( T \geq T_c \). In this case, the parameters \( a_m = a_m(\beta) \) should be found from the equations
\[ a_m = \int_{-\infty}^{\infty} A_m(\tau, \beta) \exp \left\{ \sum_{m'=1/2}^{S} a_{m'} \mathcal{F}_1(m'\tau) \right\} d\tau; \] (19)
that stem from (16) for \( b_m = 0 \).

Parameters \( b_m \) can be obtained by linearization of the second equation (16):
\[ b_m = \sum_{m'=1/2}^{S} K_{m,m'}b_{m'}; \ K_{m,m'} = \int_{-\infty}^{\infty} B_m(\tau, \beta) \mathcal{F}_2(m'\tau) \exp \left\{ \sum_{m''=1/2}^{S} a_{m''}(\beta) \mathcal{F}_1(m''\tau) \right\} d\tau. \] (20)
Thus, near $T_c$ parameters $b_m$ obey the system of homogeneous linear equations. Well-known condition of existence of non-trivial solution of this system gives following equation for $T_c$:

$$\det (K - I) = 0,$$

where $I = \delta_{m,m'}$ is the identity matrix, $m, m' = 1/2, \ldots, S$.

The Eqs (14) and (21) are the main result of theoretical background we develop. We consider the system of these equations from the case of $S = 1/2$. The system (20) reduces to single equation with $a_{1/2} = A_{1/2} = 1; A_{1/2}(\tau, \beta) = \delta (\tau)$ and $B_{1/2} = \tanh \beta H/2$; $B_{1/2}(\tau, \beta) = (\beta \sinh \tau \beta)^{-1}$.

Subsequent calculations require the definition of spatial dependence of $J(r')$. Usually in the problems of carrier-induced ferromagnetism in DMS, the RKKY interaction is considered as an effective spin-spin exchange interaction that results in FM ordering. Recently, the complex valence band structure of III-V and II-VI semiconductors was discussed as a reason for magnetic anisotropy and some enhancement of carrier-induced magnetic interaction [10], [6]. To clarify the role of fluctuations, here we neglect the effects of complex valence band structure, which can be also incorporated in our theory.

In the case of simple one band carrier structure, the RKKY interaction reads

$$J(r) = J_0 \frac{x \cos x - \sin x}{x^4}, \quad x = 2k_F r$$

$$J_0 = \frac{1}{4\pi^3} \frac{J_{ci} \Omega_0^2 m^* k_F^4}{\hbar^2},$$

where $J_{ci}$ is a carrier-ion exchange constant, $\Omega_0$ is a unit cell volume, $m^*$ is the density of states effective mass. The critical temperature is determined now by the equation that follows from the Eq. (20) after the substitution of Eq. (22) to integrals (15):

$$1 = \frac{\theta \nu}{2} \int_{-\infty}^{\infty} \frac{\varphi_2(\xi)}{\sinh (\theta \xi)} \exp \left( -\frac{\pi \nu}{2} \varphi_1(\xi) \right) d\xi.$$  

(24)

Here we introduce the dimensionless parameters $\theta = \pi T_c/2 J_0$ and $\nu = n_i/6 \pi^2 n_e$. The functions $\varphi_1(\xi)$ and $\varphi_2(\xi)$ are related to integrals (15). In the case of homogeneous magnetic ions distribution, $n_i(\vec{r}) = n_i = \text{const}$,

$$\varphi_1(\xi) = \int_0^\infty \left\{ 1 - \cos \left( \frac{\xi x \cos x - \sin x}{x^4} \right) \right\} x^2 dx$$

$$\varphi_2(\xi) = \int_0^\infty \sin \left( \frac{\xi x \cos x - \sin x}{x^4} \right) x^2 dx.$$  

(25)  

(26)

The MFA results (for spin $S = 1/2$) can be recovered from Eqs (2) and (21) by their expansion up to the linear terms $\varphi_1(\xi) \to 0; \varphi_2(\xi) \to \xi$, that leads to the equation for critical temperature in the form

$$\theta^{MF} = \nu \pi^2/4.$$  

Gaussian asymptotics of distribution function can be used to analyze the influence of fluctuations on $T_c$ near MFA. Latter asymptotics corresponds to the next term of expansion of the Eq. (27),

$$\varphi_1(\xi) \to \pi \xi^2/30, \quad \varphi_2(\xi) \to \xi.$$  

(27)

The numerical solution of the Eq. (24) with respect to $\nu$ is presented in the Fig.1. Note the qualitative difference in behavior of $\theta$ as a function of $n_e/n_i$ for MFA and Gaussian distribution function. While MFA predicts the FM ordering at any relation between the electron and magnetic ion concentrations, the fluctuations suppress ferromagnetism and make it impossible if the concentration ratio $n_e/n_i = 1/6 \pi^2 \nu$ exceeds critical value $(n_e/n_i)_c = 1/6 \pi^2 \nu_c$. We find

$$(\nu_c)^{\text{Gauss}} = \pi/15 \approx 0.2094 \quad (28)$$

as a solution of the Eq. (24) in the limit $\theta \to 0$ that corresponds to fluctuation restriction for carrier concentration $(n_{ec})^{\text{Gauss}} = 5/2 \pi^3 \approx 0.08 n_i$.

![FIG. 1. Dimensionless phase transition temperature $T_c/|J_0|$ for spin 1/2 versus ratio $\nu_e/\nu_i$. Here $J_0 = (6\pi^2)^{4/3} J_{ci}^2 m^* \Omega_0^2 / \hbar^2$, $\nu_{i,e} = n_{i,e} \Omega_0$ is unit cell volume. Here we put $\nu_i = 1$. 1- non-Gaussian distribution function, 2- Gaussian distribution function, 3- MFA.](image-url)
We can also take into account the deviations of molecular field fluctuations from their Gaussian asymptotics by numerical solution of the Eq. (24). The results are also reported in the Fig.1. The critical concentration for this case turns out to be

$$\nu_c \approx 0.1366, \quad n_{cc} \approx 0.12 n_i \quad (29)$$

The visible deviation of $T_c$ (24) from that for Gaussian asymptotics is revealed at relatively narrow interval $n_c/n_i$ near the critical value $(n_c/n_i)_c$. It can be shown that this property holds for any other magnetic ion spin $S > 1/2$. This permits to assert that the molecular field fluctuations can be considered with sufficient accuracy with Gaussian asymptotics for distribution function.

Gaussian asymptotics for $S > 1/2$ corresponds to the expansion of Eq. (17) in powers of $t$ up to the second order (it can be shown that this is valid for sufficiently high spin concentration):

$$\mathcal{F}_1(m\tau) = -\sigma^2 m^2 \tau^2,$$

$$\mathcal{F}_2(m\tau) = -\Delta H m \tau \quad (30)$$

with

$$\sigma^2 = \frac{1}{2} \int_V n_i(\vec{r}) J^2(\vec{r}) d^3r, \quad (31)$$

$$\Delta H = \int_V n_i(\vec{r}) J(\vec{r}) d^3r. \quad (32)$$

Approximation (30) permits to find the explicit form for Gaussian asymptotics of distribution function (14). This function depends on only two parameters (18): $M_1$ and $M_2$:

$$f_G(H) = f_{M_1,M_2}(H) = \frac{1}{2\sigma \sqrt{\pi M_2}} \exp \left[ - \frac{(H - \Delta H M_1)^2}{4\sigma^2 M_2} \right]. \quad (33)$$

Thus, instead the $(2S+1)/2$ equations (16) the Gaussian asymptotics "generates" only two of them

$$M_1 = \int_{-\infty}^{\infty} \langle S_Z \rangle_{H_f} f_{M_1,M_2}(H_f) dH_f, \quad (34)$$

$$M_2 = \int_{-\infty}^{\infty} \langle S_Z^2 \rangle_{H_f} f_{M_1,M_2}(H_f) dH_f. \quad (35)$$

Note that the equations (34) and (35) resemble replica-symmetric solution found for Ising (spin 1/2) spin glass (see [3]). The reason we do not use replica formalism here is that the explicit form of $J(\vec{r})$ is important for DMS. The MFA result immediately follows from the Eq. (34) in the limit of zero fluctuations, $\sigma \rightarrow 0$, $f_{M_1,M_2}(H) \rightarrow \delta(H - \Delta H M_1)$:

$$T_c^{MF} = \frac{1}{3} S(S + 1) \int_V n_i(\vec{r}) J(\vec{r}) d^3r. \quad (36)$$

The equation for $T_c$ can be obtained from Eq’s (34) and (35) as $M_1 \rightarrow 0$. Then Eq.(34) can be transformed to following equation for critical temperature

$$T_c = \frac{\Delta H}{\sqrt{\pi}} \int_{-\infty}^{\infty} m'_S \left( \frac{2\sigma \sqrt{M_2}}{T_c} x \right) e^{-x^2} dx, \quad (37)$$

where $m_S(x) = SB_S(Sx)$ is non-normalized Brillouin function, $m'_S(x)$ is its derivative. In the case of small fluctuations, $2\sigma \sqrt{M_2} \ll T$, Eq. (37) is reduced to MFA result (36).

We can find analytically the first fluctuation correction to $T_c^{MF}$ due to Gaussian fluctuations of molecular field. The first term of expansion of (37) in $\sigma/\Delta H$ yields

$$\frac{T_c}{T_c^{MF}} = 1 - \frac{6}{5} \left( 1 + \frac{1}{2S(S+1)} \right) \frac{\sigma^2}{\Delta H^2}. \quad (38)$$

The physical meaning of Eq. (38) is the lowering of $T_c$ by the fluctuations. This means that due to fluctuations, FM ordering occurs at lower temperature or that the fluctuations suppress FM order at a given temperature.

To apply this result to the case under consideration, the parameters (31) and (32) have been evaluated for RKKY interaction (22) and constant spins concentration $n_i(\vec{r}) = n_i = const$:

$$\Delta H = \frac{J_0}{12\pi} \frac{n_i}{n_e}, \quad (39)$$

$$\sigma = \frac{J_0}{6\pi} \sqrt{\frac{n_i}{n_e}}. \quad (40)$$

This gives

$$\left( \frac{T_c}{T_c^{MF}} \right)_{RKKY} = 1 - \frac{12\pi^2}{25} \left( 1 + \frac{1}{2S(S+1)} \right) \frac{n_e}{n_i}. \quad (41)$$

For the important case of the magnetic ions $Mn^{2+}$ with $S = 5/2$ the Eq. (41) yields $1 - 5.0(n_e/n_i)$. 

![Graph](image-url)
FIG. 2. Dimensionless phase transition temperature $T_c/J_{00}$ for spin 5/2 versus ratio $\nu_e/\nu_i$ (also $\nu_i = 1$). The notations are similar to those for spin 1/2. 1- Gaussian distribution function, 2- first fluctuation correction to MFA, 3 - MFA.

In Fig. 2, we reproduce the electron concentration dependence of $T_c$ calculated in MFA, with Gaussian distribution function (37) and with approximation (40) for $S = 5/2$. The dependence $T_c = T_c(n_e/n_i)$ presented in the figure reveals also the presence of critical ratio $(n_e/n_i)_c$. The equation for the $(n_e/n_i)_c$ can be found as $T \rightarrow 0$ in Eq. (37). Equation we obtain $(\Delta H^2 = \pi \sigma^2)$ is independent of spin, so account for the Eqs (39) reproduces the result we discussed for spin $S = 1/2$: $\nu_c = \pi/15$, or $(n_e/n_i)_c = 5/2\pi^3 \approx 0.08$.

In conclusion, here we present a theory of fluctuated molecular field for the spin systems with given dependence $J(\vec{r})$. We derive the equations for order parameters and FM phase transition temperature for Ising model with indirect spin-spin RKKY interaction in DMS. Our analysis show, that fluctuations of transverse spin components (which are present in Heisenberg model) do not reveal new qualitative properties in the problem of FM ordering in DMS.

Naturally, the RKKY interaction is not unique spin-spin interaction in DMS. There is additional background interaction independent of free carrier concentration. In II-VI DMS this AFM exchange interaction is dominant for close magnetic pairs. An intuitive approach to this problem was proposed in Ref [11], where MFA was applied to ensemble of magnetic ion spins with excluded AFM exchange pairs. Our formalism can naturally incorporate this interaction by adding correspondent terms to Hamiltonian (1).

Actually, the parameter of theory that present the fluctuations of RKKY interaction in bulk DMS is a cube of ratio of Fermi wave vector length to mean inter-ion separation. We can expect that similar parameter will be "responsible" for fluctuations in 2D systems. This mean that critical temperature would depend on carrier concentration in accordance with MFA prediction in 2D case.

The detailed analysis of all aforementioned factors will be presented elsewhere.

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