Quasi-Two Dimensional Diluted Magnetic Semiconductors 
with Arbitrary Carrier Degeneracy

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In the framework of the generalized mean field theory, conditions for arising the ferromagnetic state in a \textit{two-dimensional} diluted magnetic semiconductor and the features of that state are defined. RKKY-interaction of magnetic impurities is supposed. The spatial disorder of their arrangement and temperature alteration of the carrier degeneracy are taken into account.

Diluted magnetic semiconductors, such as Ga$_{1-x}$Mn$_x$As \cite{1}, are broadly investigated in connection with their potential for new electronics developments and, especially, spintronics. For explanation of the ferromagnetism in those compounds the known RKKY-mechanism of the indirect exchange interaction is recruited \cite{1,2} which leads to the correct estimating the Curie temperature in the framework of the traditional mean field theory. Mn-atoms (with concentration $N_\mu$) substituting for Ga-atoms introduce in the system the own magnetic moments and, in addition, as acceptors deliver free holes (with concentration $n$). It is precisely those holes become to be carriers responsible for the interaction. However, the equality of the concentrations $n = N_\mu$ keeps only at low Mn-concentrations ($x \lesssim 0.05$) \cite{1,2}, so that the carrier concentration is usually less than the concentration of magnetic impurities: $n = \gamma N_\mu$ where the coefficient of the impurity efficiency $\gamma < 1$ and falls$^1$ with increasing $N_\mu$.

Nevertheless, the concentration of magnetic impurities, delivering carriers, in actual systems is usually so high that the impurity band is formed which at $x \gtrsim 0.01$ merges into the valence band \cite{5}. Even though, the carrier concentration occurs to be not so high that one could consider them as highly degenerated ones within the whole (being of interest) range of relatively high temperatures. Furthermore, it is important that in that range the carrier concentration is almost independent of the temperature: $n = \gamma N_\mu \approx \text{Const}$.

Although two-dimensional structures represent the most natural systems for the embedding in the traditional semiconductor technology, almost all theoretical works are dealt with the three-dimensional systems of degenerate carriers. Similarly, most of experimentally studied systems are three-dimensional ones.

The objective of the present paper is considering magnetic features of \textit{two-dimensional} semiconductor systems with magnetic impurities interacting by RKKY-mechanism via carriers of \textit{arbitrary degeneracy}. That problem has been recently considered in the paper \cite{6} where it has been shown that reducing the system dimension (from 3D to 2D) results in the significant lowering of the Curie temperature (under equivalent parameters). We think though that there are some inexactitudes in the paper. Firstly, authors \cite{6} has neglected of the temperature dependence $\varepsilon_F(T)$ of the carrier Fermi energy that is inadmissible under the intermediate degeneracy when $\varepsilon_F/k_B T \sim 1$ (and, all the more, at $\varepsilon_F < 0$). In addition, the disorder in the arrangement of magnetic impurities has been considered in the framework of the mean

$^1$One could control the relative hole concentration (i.e., the $\gamma$-value) by simultaneous introducing non-magnetic acceptors (for instance, Be \cite{3,4}) or choosing the temperature of the film growth \cite{2}.
field theory (continual or the so called lattice one) where the averaged impurity configuration is ordered one and the scattering of local effective magnetic fields is not considered. As we demonstrate below, taking into account the mentioned things influences the results significantly.

The general expression for RKKY interaction in two-dimensional systems is known [7, 8, 9], however the relevant results refer to the degenerate carriers and have to be generalized for the case of the intermediate degeneracy $|\varepsilon_F/k_BT| \sim 1$ ($\varepsilon_F$ is the carrier Fermi energy).

In a few papers [10, 11] (and later in [12]) the systems with non-degenerate carriers have been considered and it has been shown that the energy of RKKY interaction in the non-degenerate case is exponentially falls with the distance (in contrast to the degenerate case where the fall is power behaved): $J \propto \exp(-r^2/\lambda_T^2)$, where $\lambda_T = \hbar/(2mk_BT)^{1/2}$ is the mean (thermal) de Broglie carrier wave length. However, those papers, first, refer to the three-dimensional systems and, second, they lack of taking into account the random arrangement of magnetic impurities.

It is known that the traditional mean field theory does not provide the adequate description of a disordered (random) system of magnetic moments [13]. In the present paper, we shall use the generalized mean field theory [14] for systems with the indirect interaction of magnetic impurities taking into account the randomness of their spatial arrangement. We shall use Ising approximation and suppose that the indirect coupling between magnetic moments of impurity atoms is realized by means of RKKY interaction which is replaced by the effective magnetic field, whereupon system properties are described with the help of the distribution function of local values of the field arising as a result of magnetic ions’ coupling with their own surroundings. In real systems, the scattering of those fields proves to be so substantial that RKKY interaction makes the magnetic ordering possible at lower temperatures only (as compared to those predicted by the traditional mean field theory). Dependencies of magnetic properties on the carrier concentration turn out to be unusual, as well.

In [7, 8, 9], the expression has been derived for the energy $w(r)$ of indirect RKKY interaction for two parallel spins $S_1$, $S_2$ of magnetic ions spaced at the distance $r$ in the two-dimensional system with degenerated carriers:

$$w(r) = -\frac{m}{4\pi\hbar^2} \left( \frac{J_{\text{ex}}}{N} \right)^2 F(r) S_1 S_2, \quad F(r) = -\int_0^{k_F} k N_0(kr) J_0(kr) dk,$$

(1)

where $J_{\text{ex}}$ is the exchange energy for interaction of a spin with a free charge carrier of the mass $m$, $N$ is the concentration of lattice atoms ($N = 1/a^2$ for the square lattice of the period $a$); $J_{0,1}$ are Bessel functions. To generalize that result to the case of the arbitrary degeneracy (with the Fermi energy $\varepsilon_F$ of any sign and value) it is sufficient to introduce the Fermi distribution function in the integrand (1) and extend the integration over the interval $0 < k < \infty$ [6]:

$$F(r, T) = -\frac{1}{r^2} \int_0^\infty \frac{y N_0(y) J_0(y) dy}{1 + \exp[(h^2 y^2/2mr^2 - \varepsilon_F)/k_BT]}.$$

(2)

The behavior of the function (2) is determined not only by the temperature as such but also by the temperature dependence of the Fermi energy. The latter circumstance has been ignored in [6]. In the framework of the standard two-dimensional band and under the invariable carrier concentration, the ratio $\eta = \varepsilon_F/k_BT$ ($\eta < 0$ refers to the non-degenerated carriers) is defined by the relation

$$e^{\eta(T)} = e^{\pi\hbar^2 n/mkT} - 1$$

(3)
that predicts negative \( \eta \)-values at \( T = 100 \text{ K} \) if \( n \lesssim 10^{12} \text{ cm}^{-2} \). From here, e.g. for the values \( n = 3 \cdot 10^{12} \text{ cm}^{-2}, m = 0.5m_0, a = 4\text{Å} \) corresponding to the experiments \([3,4]\), the temperature dependence \( \eta(T) \) results which is shown in the inset of Fig. 1. In fact, it is seen that in the real experiment, carriers could not be considered as degenerated (in \([3,4]\) actual temperatures amount to 170 K).

Taking into account \([3]\), the expression \((11)\) could be written in the form

\[
-\frac{1}{2}\int_{-\infty}^{\infty} A(\eta) \exp(-iq\eta)d\eta, \quad A(\eta) = \lim_{N_{\text{max}} \to \infty} \left[ \sum_{\xi=\pm 1} \int_{0}^{r_{\text{max}}} e^{iqh_{\xi}(r, \eta)\xi\eta} \eta(r)dr \right]^{N_{\text{max}}},
\]

where \( \eta = r/a \) is the reduced separation between interacting impurities, \( \tau = 2\pi ma^2k_B T/h^2 \) is the reduced temperature\(^2\), \( x = N_\mu/N \) is the relative concentration of magnetic impurities. Under the strong degeneracy \( (\eta \gg 1) \), Eq. \((5)\) reproduces the result \([7]\), while in the case of non-degenerate carriers it is similar to the relevant three-dimensional result obtained in \([10]\). In the most actual case of the intermediate degeneracy (see below), the function \((5)\) has been estimated by numerical calculations.

In Fig. 1, dependencies \( \phi(\rho) \) are shown calculated with \((5)\) for different values of the carrier concentration \( x\eta \) (the actual range of \( x\eta \)-values is determined by conditions of the concrete experiment). Particularly, it is clear that at low enough carrier concentrations (corresponding to \( \eta \ll 1 \), \( \phi(\rho) \) does not depend quantitatively correct results at \( S \geq 1 \), as well. Appropriate generalization does not meet some principal difficulties.

Let the system consisting of randomly arranged and oriented Ising spins be in the state characterized by the average reduced magnetization \( 0 \leq j \leq 1 \). The total interaction energy \( W = \sum_i w_i \) of a given spin \( S_i \) with other spins \( S_j \) \( (i = 2, 3, \ldots) \) is a random value which we shall define by the effective local magnetic field \( H = -W/\mu \) \( (\mu = g\mu_B\sqrt{S(S+1)}) \) and describe by the distribution function \( F(j; H) \) depending on the average concentration \( N_\mu \) of effective magnetic ions and the reduced system magnetization \( j = 2\xi - 1 \) where \( \xi \) is the average fraction of spins of magneto-active ions directed up.

The randomness of the distribution of magneto-active impurities is restricted only by the necessity to place them in the fixed locations (sites) of the matrix lattice. For strongly diluted systems, that restriction is not significant and the distribution function could be found by the Markov’s method \([15]\), according to which

\[
F(j; H) = \frac{1}{2\pi} \int_{-\infty}^{\infty} A(q) \exp(-iqH)dq, \quad A(q) = \lim_{N_{\text{max}} \to \infty} \left[ \sum_{\xi=\pm 1} \int_{0}^{r_{\text{max}}} e^{iqh_{\xi}(r, \eta)\xi\eta} \eta(r)dr \right]^{N_{\text{max}}},
\]

where \( h_{\xi}(r, \eta) = -\xi w(r)/\mu \) is the field generated at the origin by the spin spaced at the random distance \( r \) from it. The random parameter \( \xi \) takes values \( \pm 1 \) (with probabilities \( \xi \)

\(^2\text{For GaMnAs } \tau \approx 10^{-5} T/\text{K}.\)

3
and \((1 - \xi)\), accordingly) and determines the direction of the remote spin, \(\kappa_\zeta(\zeta)\) and \(\kappa_r(r)\) are distribution functions for random values of the parameter \(\zeta\) and the distance \(r\), \(N_{\text{max}} = \pi r_{\text{max}}^2 N_\mu\) is the number of magneto-active impurities in the circle of the radius \(r_{\text{max}}\) over whose area the integration is performed.

In the spirit of the mean field theory, \(\zeta\)-distribution could be written as

\[
\kappa_\zeta(\zeta) = [(1 - \xi)\delta(\zeta + 1) + \xi \delta(\zeta - 1)].
\]  

As for the \(r\)-distribution, \(\kappa_r(r)\), one has, in principle, to account for the clustering effect originating from the mutual attraction experienced by near Mn-atoms in GaAs-lattice and resulting in their non-random (correlated) spatial distribution. Uniform non-correlated \(r\)-distribution would be

\[
\kappa_r(r) = \begin{cases} 
2r/(r_{\text{max}}^2 - r_{\text{min}}^2), & r > r_{\text{min}}, \\
0, & r < r_{\text{min}},
\end{cases}
\]

where the existence of the minimal separation \(r_{\text{min}}\) between magnetic ions (conditioned by the embedding of impurity atoms in the lattice of semiconductor matrix) is accounted for\(^3\). To take into account the correlation of Mn-atoms one has to add the correlation function \(g(r)\) in the right-hand side of that relation. This function could be, in principle, calculated if the spatial dependence of Mn-atoms interaction energy would be known. However, the exact reason for the tendency of Mn-atoms to clustering is unclear\(^{16}\). At the same time, Monte-Carlo calculations\(^{17}\) show that impurity correlations have only small effects on the ferromagnetic transition temperature of Ga\(_{1-x}\)Mn\(_x\)As 3D-system. Thus, though the consideration of the clustering is, in principle, straightforward – it is sufficient to introduce in the integrand\(^8\) (and \((9), (11)\), see below) the correlation function \(g(r)\) – later on, we will use the simple relation\(^8\) that corresponds to \(g(r) = 1\).

Substituting \((7), (8)\) in \((6)\) one finds

\[
A(q) = \exp[-2\pi N_\mu C(q)], \quad C(q) = \int_{r_{\text{min}}}^{\infty} \{1 - \cos[qh(r)] - i \cdot j \sin[qh(r)]\} r \, dr.
\]

Relationships\(^9\) do not lead to a simple analytical expression for the distribution function \(F_\mu(j; H)\). So, to determine the latter we have used the low \(q\) approximation, based on the fact that in the inverse Fourier transform\(^6\) the region of high \(q\)-values is not important. In that approximation

\[
C(q) = Pq^2 - ijQq,
\]

where

\[
P = \frac{1}{2} \int_{r_{\text{min}}}^{\infty} h^2(r) r \, dr = (J_{\text{eff}}/\mu)^2 a^2 \phi_P(\rho_{\text{min}}), \quad \phi_P(\tau) = \frac{1}{2} \int_{\rho_{\text{min}}}^{\infty} \phi^2(\rho) \rho d\rho,
\]

\[
Q = \int_{r_{\text{min}}}^{\infty} h(r) r \, dr = (J_{\text{eff}}/\mu)^2 a^2 \phi_Q(\rho_{\text{min}}), \quad \phi_Q(\tau) = \int_{\rho_{\text{min}}}^{\infty} \phi(\rho) \rho d\rho,
\]

\(^3\)The minimal possible distance between magnetic-active Mn-ions substituting for Ga-atoms in the zincblende AsGa-lattice equals \(a = a_0/\sqrt{2} \approx 4 \text{Å}\) where \(a_0 = 5.7 \text{Å}\) is the side of the cubic cell.
\( \rho_{\text{min}} \equiv r_{\text{min}}/a = 1. \)

Substituting (10), (9) in (6) we find that in the considered approach the distribution \( F(j; H) \) is described by the shifted (relative to \( H = 0 \)) Gauss function:

\[
F(j; H) = \frac{1}{\sqrt{2\pi}\sigma} \exp \left[ -\frac{(H - jH_j)^2}{2\sigma^2} \right],
\]

(12)

\[
H_j = 2\pi N_\mu Q \propto N_\mu, \quad \sigma = (2\pi N_\mu P)^{1/2} \propto N_\mu^{1/2}.
\]

(13)

The position of the maximum (\( H = jH_j \)) of the distribution is determined by the parameter \( Q \) and depends linearly on the system magnetization \( j \) while the distribution width \( \sigma \) is defined by the parameter \( P \) and does not depend on \( j \). The positive sign of \( H_j \) means that the average direction of the effective magnetic field coincides with the direction of the average magnetization, that is the field is, on average, promotes the ferromagnetic ordering of magnetic moments.

Temperature dependencies \( H_j(\tau) \) of the exchange field defined by the relation (13) are shown in Fig. 2. As illustrated in the inset of that Figure, \( H_j \)-value peaks at \( x\gamma \sim 0.01 \). It is just this condition which results in the maximum Curie temperature (see below).

Relations (13) for the shift \( H_j \) of the distribution function \( F(j; H) \) and its broadening \( \sigma \) could be rewritten in the form

\[
H_j = \left( J_{\text{eff}}/\mu \right) [2\pi x \phi_Q(\tau)], \quad \sigma = \left( J_{\text{eff}}/\mu \right) [2\pi x \phi_P(\tau)]^{1/2}.
\]

(14)

It follows herefrom

\[
H_j/\sigma = (2\pi x)^{1/2} \psi(\tau), \quad \psi(\tau) = \frac{\phi_Q(\tau)}{[\phi_P(\tau)]^{1/2}}.
\]

(15)

As is shown below, the ferromagnetic ordering is possible under the condition \( H_j/\sigma > \sqrt{\pi}/2 \) or

\[
x^{1/2} \psi(\tau) > 1/2.
\]

(16)

Thus, in addition to the material parameters \( x = N_\mu/N \) and \( \gamma = n/N_\mu \), the function \( \psi(\tau) \) defining the ratio \( H_j/\sigma \) of Gauss function parameters becomes to be crucial in the considered problem. As an example, temperature dependencies of that ratio for \( x = 0.1 \) and various \( \gamma \)-values are shown in Fig. 3. In the inset, the dependencies of the maximum Curie temperature \( T_{C_{\text{max}}} \) on \( \gamma \)-value following from (16) is displayed (for various \( x \)-values).

In the traditional mean field theory, the distribution function is \( \delta \)-like one for any magnetization \( j \): \( F(j; H) = \delta[H - jH_j] \). It is evident, that the broadening of that distribution in a random system prevents ferromagnetic ordering. The magnetization of such a disordered system has to be calculated taking into account the scattering of local interaction energies \( H \) by means of the straight-forward generalization of the equation \( j = \tanh[\mu H(j)/kT] \) referring to the regular Ising system:

\[
j = \int_{-\infty}^{\infty} \tanh \left[ \frac{\mu H}{kT} \right] F(j; H) dH.
\]

(17)

4The lattice model [13] used in [6] corresponds to \( \delta \)-like distribution function \( F(j, H) = \delta(H - jH_j) \) where \( \mu H_j = \sum_i N_i w(r_i); N_i, r_i \) are the number of \( i \)th nearest neighbors and their distance.
Using the expression (12) for the distribution function $F(j; H)$ one gets the equation generalizing the standard mean field one:

$$j = -\frac{1}{\sqrt{2\pi}} \left( \frac{H_j}{\sigma} \right) \int_{-\infty}^{\infty} \tanh \left( \frac{u}{\theta} \right) \exp \left[ -\frac{1}{2} \left( \frac{H_j}{\sigma} \right)^2 (u - j)^2 \right] \, du,$$

(18)

where \( \theta = kT/\mu H_j = \tau/x I^2 \phi_Q(\tau) \), \( I = \sqrt{\pi}J_{ex}(\hbar^2/ma)^{-1} \) is the reduced strength of the interaction\(^5\). That equation predicts the phase diagram of the system, temperature dependencies of its magnetization (in ferromagnetic phase) and susceptibility (in paramagnetic phase), as well as the dependence of Curie temperature \( \theta_C \) on the interaction strength, the relative magnetic ion concentration \( x = N_\mu/N \) and the relative free carrier concentration \( \gamma = n/N_\mu \).

To clarify under what conditions that equation has the solution corresponding to the ferromagnetic state \((j > 0)\) notice that in the vicinity of Curie temperature where the magnetization is small \((j \to 0)\), it follows from (18)

$$\sqrt{\frac{2}{\pi}} \left( \frac{H_j}{\sigma} \right)^3 \int_0^{\infty} \tanh \left( \frac{u}{\theta} \right) \exp \left[ -\frac{1}{2} \left( \frac{H_j}{\sigma} \right)^2 u^2 \right] u \, du = 1.$$

(19)

The integral in (19) peaks at \( \theta = \tau = 0 \) and its maximum value equals \((\sigma/H_j)^2\). It follows herefrom that the ordered state is only possible under the condition

$$\frac{H_j}{\sigma} > \sqrt{\frac{\pi}{2}},$$

(20)

which means that the effective RKKY-field proportional to \( H_j \) has to overpower not only the thermal disordering but also the scattering of local fields proportional to \( \sigma \). The upper boundary \( \tau_C^{\text{max}} \) of the temperature range where the cited condition is satisfied determines the maximum attainable temperature of the ferromagnetic ordering at infinite interaction energy \((I \to \infty)\). It could be derived from the condition \( 2x^{1/2} \psi(\tau) = 1 \). The existence of the maximum Curie temperature is associated with the lifting of the carrier degeneration at high temperatures and, as a consequence, the finiteness of the effective energy of inter-impurity interaction even at \( I \to \infty \).

Curie temperature at the finite interaction energy could be determined by solving the equation (19). Relevant non-monotone dependencies \( \tau_C(\gamma) \) are displayed in Fig. 4. It is clear that the relative carrier concentration \( \gamma = n/N_\mu \) ambiguously influences Curie temperature in accordance with the non-monotone dependence of the exchange field \( H_j \) on \( \gamma \) (cf. Fig. 2). To compare, the dashed line in Fig. 4 reproduces the dependence \( \tau_C(n) \) presented in \([6]\) and obtained in the framework of the standard mean-field theory.

The optimal carrier concentration occurs to be on the order of \( 10^{12} \text{ cm}^{-2} \), that is significantly lower than the value \( n \sim 10^{14} \text{ cm}^{-2} \) predicted in \([6]\). In addition, Fig. 5 demonstrates there is a threshold value of the interaction strength \( I \) to drive the system in the ferromagnetic state. This is to be contrasted with the result of the standard mean-field theory which predicts no such a threshold.

\(^5\)Presently accepted value \( J_{pol} = 0.15 \text{ eV}\cdot\text{nm}^3 \) \([2]\) (for GaMnAs) results in \( I \approx 1 \).
In conclusion, conditions of establishing the ferromagnetic state and its parameters in quasi-two dimensional semiconductor systems with magnetic impurities coupled via RKKY interaction have been studied in the paper. As distinct from [6], two new important factors have been included in the consideration, allowing for the spatial disarray of interacting magnetic impurities, and the temperature dependence of the carrier degeneracy. It has been demonstrated that both factors complicate transition of the system into the ferromagnetic state: disorder of the impurities arrangement reduces the Curie temperature (as compared to the regular system) while lifting the degeneracy of carriers makes the Curie temperature finite even in the extreme case of the infinitely strong interaction. Besides, the concentration dependence of the transition temperature occurs to be non-monotone and there is a threshold interaction strength to drive the system in the ferromagnetic state.

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Captions

Fig. 1. Spatial dependencies $\phi(\rho)$ of the RKKY interaction energy for magnetic impurities in the two-dimensional system with various carrier concentrations degeneracy determining by the parameter $x\gamma$ (notice, different scales above and under the Y-axis break). In the insert – the temperature dependence $\eta(T)$ corresponding to conditions of the experiments [3, 4].

Fig. 2. Temperature dependencies $H_j(\tau)$ of effective exchange field for the two-dimensional system with various concentrations of magnetic impurities ($x = N_\mu/N$) and free carriers ($\gamma = n/N_\mu$). In the insert – the maximum attainable $H_j$-value.

Fig. 3. Temperature dependencies of the ratio $H_j/\sigma$ of Gauss distribution function parameters for two-dimensional system with the concentration of magnetic impurities $x = 0.05$ at various carrier concentrations $\gamma$. In the insert – maximum Curie temperature $\tau_C^{\text{max}}$ that could be attained at given $x$- and $\gamma$-values.

Fig. 4. Dependencies $\tau_C(n)$ of Curie temperature on the carrier concentration for two-dimensional system with the concentration of magnetic impurities $x = 0.1$ for various interaction strengths $I$. The dashed line is the result having presented in [6] and corresponding to $I \approx 1$.

Fig. 5. Dependencies $\tau_C(I)$ of Curie temperature for two-dimensional system with the concentration of magnetic impurities $x = 0.1$ at various carrier concentrations determined by the parameter $\gamma$. 
\[ \eta = \epsilon_F/kT \]

\[ n = 3 \times 10^{12} \text{ cm}^{-2} \]

\[ m = 0.5m_0 \]

\[ \phi(\rho) \]

\[ x\gamma = 0.1 \]

\[ \rho, \text{ cm}^{-2} = 0.5 \]

\[ \tau = 0.1 \]

\[ \rho \]

\[ \rho \]
