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 the ferromagnetic state in a *two-dimensional* diluted magnetic semiconductor and the features of that state are defined. The Ruderman-Kittel-Kasuya-Yosida interaction of magnetic impurities is supposed. The spatial disorder of their arrangement and temperature alteration of the carrier degeneracy are taken into account.

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Diluted magnetic semiconductors, such as $Ga_{1-x}Mn_xAs$,[1] are broadly investigated in connection with their potential for new electronics developments and, especially, spintronics. For an explanation of the ferromagnetism in those compounds, the known Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism of the indirect exchange interaction is used,[1,2] which leads to the correct estimate of the Curie temperature in the framework of the traditional mean-field theory. Mn atoms (with concentration N_μ) substituting for Ga atoms introduce into the system their own magnetic moments, and, as acceptors do, they deliver free holes (with concentration n). It is precisely those holes that become carriers responsible for the interaction. However, the equality of the concentrations $n=N_\mu$ is maintained only at low Mn concentrations ($x \lesssim 0.05$),[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 with increasing N_μ. (One could control the relative hole concentration (i.e., the γ value) by simultaneously introducing nonmagnetic acceptors [for instance, Be (Refs. 3 and 4)] or choosing the temperature of the film growth.[5])

The concentration of magnetic impurities delivering carriers in actual systems is usually so high that an impurity band is formed that at $x \gtrsim 0.01$ merges into the valence band.[5] Nevertheless, the carrier concentration turns out to be not so high as to be considered highly degenerate within the whole range of interest 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 incorporation into traditional semiconductor technology, almost all theoretical works thus far have dealt with three-dimensional systems of degenerate carriers. Similarly, most of the experimentally studied systems have been three-dimensional.

The objective of the present paper is to consider magnetic features of *two-dimensional* semiconductor systems with magnetic impurities interacting by the RKKY mechanism via carriers of *arbitrary degeneracy*. That problem was considered recently in Ref. 6, where it was shown that reducing the system dimension (from 3D to 2D) results in a significant lowering of the Curie temperature (under equivalent parameters). We think, however, that there are some inexactitudes in the paper. First, the authors[6] neglected the temperature dependence $\varepsilon_F(T)$ of the carrier Fermi energy, which is inadmissible under the intermediate degeneracy when $\varepsilon_F/k_BT \sim 1$ (and, even more, at $\varepsilon_F < 0$). In addition, the disorder in the arrangement of magnetic impurities was considered in the framework of the mean-field theory (the continual or so-called lattice one), where the averaged impurity configuration is *ordered* and the *scattering* of local effective magnetic fields is not considered. As we demonstrate below, taking these things into account influences the results significantly.

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

In a few papers[10,11] (and later in Ref. 12), the systems with nondegenerate carriers were considered and it was shown that the energy of the RKKY interaction in the nondegenerate case falls exponentially with the distance (in contrast to the degenerate case, in which the fall is power-dependent): $J \propto \exp(-r^2/\lambda_T^2)$, where $\lambda_T=\hbar/(2mk_BT)^{1/2}$ is the mean (thermal) de Broglie carrier wavelength. However, first of all, those papers refer to the three-dimensional systems, and secondly, they do not take into account the random arrangement of magnetic impurities.

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

In Refs. 7–9 an expression has been derived for the energy $w(r)$ of the indirect RKKY interaction for two parallel spins $\mathbf{S}_1, \mathbf{S}_2$ of magnetic ions spaced at a distance r in the

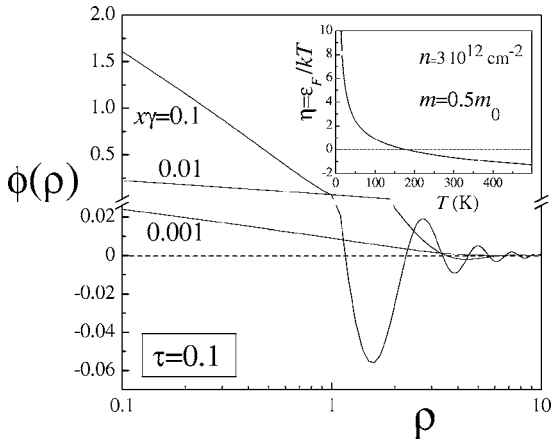

FIG. 1. Spatial dependencies $\phi(\rho)$ of the RKKY interaction energy for magnetic impurities in the two-dimensional system with various carrier concentrations determined by the parameter $x\gamma$ (notice the different scales above and below the y-axis break). Inset: the temperature dependence $\eta(T)$ corresponding to conditions of the experiments (Refs. 3 and 4).

two-dimensional system with *degenerated* carriers,

$$w(r) = -\frac{m}{4\pi\hbar^2}\left(\frac{J_{\mathrm{ex}}}{N}\right)^2 F(r)\mathbf{S}_1\mathbf{S}_2,$$
$$F(r) = -\int_0^{k_F} kN_0(kr)J_0(kr)dk, \tag{1}$$

where J_{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), and $J_{0,1}$ and $N_{0,1}$ are Bessel functions. To generalize that result to the case of arbitrary degeneracy (with the Fermi energy ε_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]

$$F(r,T) = -\frac{1}{r^2}\int_0^{\infty} \frac{yN_0(y)J_0(y)dy}{1+\exp[(\hbar^2y^2/2mr^2-\varepsilon_F)/k_BT]}. \tag{2}$$

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 was ignored in Ref. 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 nondegenerate carriers) is defined by the relation

$$e^{\eta(T)} = e^{\pi\hbar^2 n/mkT} - 1, \tag{3}$$

which predicts negative η values at $T=100$ K if $n \lesssim 10^{12}$ cm^{-2}. From here, e.g., for the values $n=3\times10^{12}$ cm^{-2}, $m=0.5m_0$, and $a=4$ 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 degenerate (in Refs. 3 and 4, the actual temperatures amounted to 170 K).

Taking into account Eq. (3), the expression (1) could be written in the form

$$w(\rho) = -J_{\mathrm{eff}}\phi(\rho,\tau)\mathbf{S}_1\mathbf{S}_2, \tag{4}$$

where $J_{\mathrm{eff}}=(ma^2/4\pi\hbar^2)J_{\mathrm{ex}}^2$,

$$\phi(\rho,\tau) = -\frac{1}{\rho^2}\int_0^{\infty}\frac{yN_0(y)J_0(y)dy}{1+\exp(y^2/\rho^2\tau-\eta)}$$
$$= -\frac{1}{\rho^2}[e^{2\pi^2\gamma x/\tau}-1]\int_0^{\infty}\frac{yN_0(y)J_0(y)dy}{e^{2\pi^2\gamma x/\tau}+e^{y^2/\rho^2\tau}-1}, \tag{5}$$

$\rho=r/a$ is the reduced separation between interacting impurities, $\tau=2\pi ma^2k_BT/\hbar^2$ is the reduced temperature [for GaMnAs $\tau\approx10^{-3}T$ (K)], and $x=N_\mu/N$ is the relative concentration of magnetic impurities. Under the strong degeneracy ($\eta\gg1$), Eq. (5) reproduces the result from Ref. 7, while in the case of nondegenerate carriers it is similar to the relevant *three-dimensional* result obtained in Ref. 10. In the most actual case of the intermediate degeneracy (see below), the function (5) has been estimated by numerical calculations.

In Fig. 1, the dependencies $\phi(\rho)$ are shown calculated with Eq. (5) for different values of the carrier concentration $x\gamma$ (the actual range of $x\gamma$ values is determined by the conditions of the concrete experiment). In particular, it is clear that at low enough carrier concentrations (corresponding to $\eta\leqslant0$), oscillations of the function $\phi(\rho)$ disappear and its sign corresponds to the ferromagnetic interaction.

For simplicity, we use the Ising model corresponding to $S=1/2$ and leading, as is known, to qualitatively correct results at $S\gtrsim1$ as well. Appropriate generalization does not resolve some fundamental difficulties.

Let the system consisting of randomly arranged and oriented Ising spins be in the state characterized by the average reduced magnetization $0\leqslant j\leqslant1$. The total interaction energy $W=\Sigma_i w_i$ of a given spin $\mathbf{S}_1$ with other spins $\mathbf{S}_i$ ($i=2,3,\ldots$) is a random value that 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_μ of effective magnetic ions and the reduced system magnetization $j=2\xi-1$, where ξ is the average fraction of spins of magneto-active ions directed upward.

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 Markov's method,[15] according to which

$$F(j;H) = \frac{1}{2\pi}\int_{-\infty}^{\infty}A(q)\exp(-iqH)dq,$$
$$A(q) = \lim_{N_{\max}\to\infty}\left[\sum_{\zeta=\pm1}\int_0^{r_{\max}} e^{iqh_\zeta(r,\zeta)}\kappa_\zeta(\zeta)\kappa_r(r)dr\right]^{N_{\max}}, \tag{6}$$

where $h_\zeta(r,\zeta)=-\zeta w(r)/\mu$ is the field generated at the origin by the spin spaced at the random distance r from it. The random parameter ζ takes values ± 1 [with probabilities ξ 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 ζ and the distance r, and $N_{\max}=\pi r_{\max}^2 N_\mu$ is the number of magneto-active impurities in the circle of the radius $r_{\max}$ over whose area the integration is performed.

In the spirit of the mean-field theory, ζ distribution could be written as

$$\kappa_\zeta(\zeta)=[(1-\xi)\delta(\zeta+1)+\xi\delta(\zeta-1)]. \tag{7}$$

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 the GaAs lattice and resulting in their nonrandom (correlated) spatial distribution. Uniform noncorrelated r distribution would be

$$\kappa_r(r)=\begin{cases} 2r/(r_{\max}^2-r_{\min}^2), & r>r_{\min}, \\ 0 & r<r_{\min}, \end{cases} \tag{8}$$

where the existence of the minimal separation $r_{\min}$ between magnetic ions (conditioned by the embedding of impurity atoms in the lattice of a semiconductor matrix) is accounted for. (The minimal possible distance between magnetic-active Mn ions substituting for Ga atoms in the zinc-blende AsGa lattice equals $a=a_0/\sqrt{2}\approx 4$ Å, where $a_0=5.7$ Å is the side of the cubic cell.) To take into account the correlation of Mn atoms, one has to add the correlation function $g(r)$ on the right-hand side of that relation. This function could be, in principle, calculated if the spatial dependence of Mn-atom interaction energy were known. However, the exact reason for the tendency of Mn atoms to cluster 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 the $Ga_{1-x}Mn_xAs$ 3D system. Thus, although the consideration of the clustering is, in principle, straightforward—it is sufficient to introduce in the integrand (8) [and Eqs. (9) and (11), see below] the correlation function $g(r)$—later on we will use the simple relation (8) that corresponds to $g(r)=1$.

Substituting Eqs. (7) and (8) into Eq. (6), one finds

$$A(q)=\exp[-2\pi N_\mu C(q)],$$
$$C(q)=\int_{r_{\min}}^{\infty}\{1-\cos[qh(r)]-ij\sin[qh(r)]\}rdr. \tag{9}$$

Relationships (9) do not lead to a simple analytical expression for the distribution function $F_x(j;H)$. 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,

$$C(q)=Pq^2-ijQq, \tag{10}$$

where

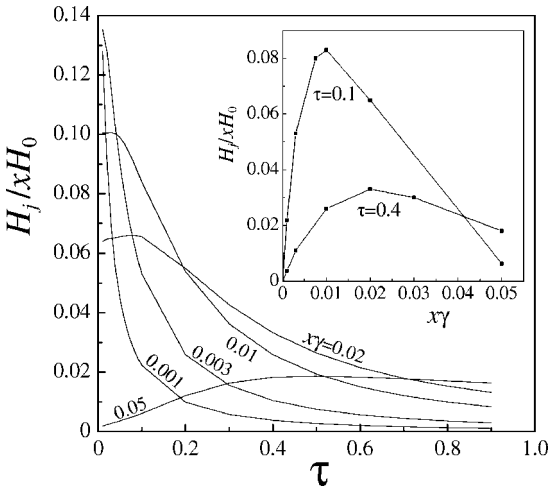

FIG. 2. Temperature dependencies $H_j(\tau)$ of effective exchange field for the two-dimensional system with various concentrations of magnetic impurities ($x=N_\mu/N$) and free carriers ($\gamma=n/N_\mu$). Inset: the maximum attainable H_j value.

$$P=\frac{1}{2}\int_{r_{\min}}^{\infty}h^2(r)rdr=(J_{\text{eff}}/\mu)^2a^2\phi_P(\rho_{\min}),$$
$$\phi_P(\tau)=\frac{1}{2}\int_{\rho_{\min}}^{\infty}\phi^2(\rho)\rho d\rho,$$
$$Q=\int_{r_{\min}}^{\infty}h(r)rdr=(J_{\text{eff}}/\mu)a^2\phi_Q(\rho_{\min}), \tag{11}$$
$$\phi_Q(\tau)=\int_{\rho_{\min}}^{\infty}\phi(\rho)\rho d\rho,$$

$\rho_{\min}\equiv r_{\min}/a=1$.

Substituting Eqs. (10) and (9) into Eq. (6), we find that in the considered approach, the distribution $F(j;H)$ is described by the shifted (relative to $H=0$) Gauss function

$$F(j;H)=\frac{1}{\sqrt{2\pi}\sigma}\exp\left[-\frac{(H-jH_j)^2}{2\sigma^2}\right], \tag{12}$$

$$H_j=2\pi N_\mu Q\propto N_\mu,\quad \sigma=(2\pi N_\mu P)^{1/2}\propto N_\mu^{1/2}, \tag{13}$$

while the lattice model[13] used in Ref. 6 corresponds to the δ-like distribution function $F(j,H)=\delta(H-jH_j)$, where $\mu H_j=\Sigma_i N_i w(r_i)$; N_i and r_i are the number of ith nearest neighbors and their distance.

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 σ 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, on average the field promotes the *ferromagnetic* ordering of magnetic moments.

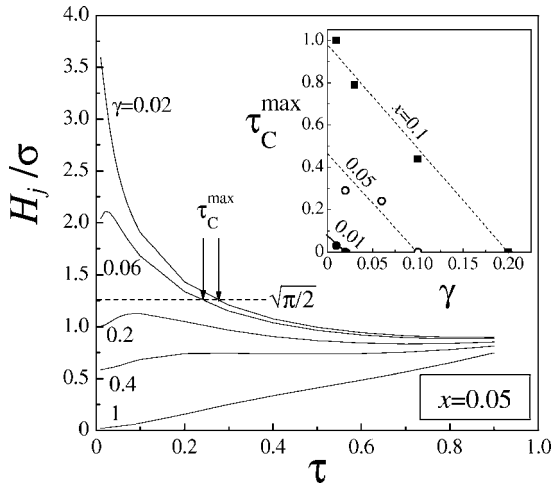

FIG. 3. Temperature dependencies of the ratio H_j/σ of Gauss distribution function parameters for the two-dimensional system with the concentration of magnetic impurities $x=0.05$ at various carrier concentrations γ. Inset: maximum Curie temperature $\tau_C^{\max}$ that could be attained at given x and γ values.

The 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, the H_j value peaks at $x\gamma \sim 0.01$. It is just this condition that 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 σ could be rewritten in the form

$$H_j = (J_{\text{eff}}/\mu)[2\pi x \phi_Q(\tau)], \quad \sigma = (J_{\text{eff}}/\mu)[2\pi x \phi_P(\tau)]^{1/2}. \tag{14}$$

It follows that

$$H_j/\sigma = (2\pi x)^{1/2}\psi(\tau), \quad \psi(\tau) = \frac{\phi_Q(\tau)}{[\phi_P(\tau)]^{1/2}}. \tag{15}$$

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

$$x^{1/2}\psi(\tau) > 1/2. \tag{16}$$

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/σ of Gauss function parameters becomes crucial in the problem considered. As an example, the temperature dependencies of that ratio for $x=0.1$ and various γ values are shown in Fig. 3. In the inset, the dependencies of the maximum Curie temperature $\tau_C^{\max}$ on the γ value following from Eq. (16) are displayed (for various x values).

In the traditional mean-field theory, the distribution function is δ-like 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 straightforward generalization of the equation $j=\tanh[\mu H(j)/kT]$ referring to the regular Ising system,

$$j = \int_{-\infty}^{\infty} \tanh\left[\frac{\mu H}{kT}\right] F(j;H)\,dH. \tag{17}$$

Using the expression (12) for the distribution function $F(j;H)$, one gets the equation generalizing the standard mean-field one,

$$j = -\frac{1}{\sqrt{2\pi}}\left(\frac{H_j}{\sigma}\right)\int_{-\infty}^{\infty} \tanh\left(\frac{u}{\theta}\right)\exp\left[-\frac{1}{2}\left(\frac{H_j}{\sigma}\right)^2 (u-j)^2\right]du, \tag{18}$$

where $\theta=kT/\mu H_j=\tau/xI^2\phi_Q(\tau)$, and $I=\sqrt{\pi}J_{\text{ex}}(\hbar^2/ma^2)^{-1}$ is the reduced strength of the interaction [for GaMnAs, the presently accepted value $J_{\text{pd}}=0.15$ eV nm^3 (Ref. 2) results in $I\approx 1$]. That equation predicts the phase diagram of the system, the temperature dependencies of its magnetization (in ferromagnetic phase) and susceptibility (in paramagnetic phase), as well as the dependence of Curie temperature θ_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 Eq. (18) that

$$\sqrt{\frac{2}{\pi}}\left(\frac{H_j}{\sigma}\right)^3 \int_0^{\infty} \tanh\left(\frac{u}{\theta}\right)\exp\left[-\frac{1}{2}\left(\frac{H_j}{\sigma}\right)^2 u^2\right]u\,du = 1. \tag{19}$$

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

$$\frac{H_j}{\sigma} > \sqrt{\frac{\pi}{2}}, \tag{20}$$

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 σ. The upper boundary $\tau_C^{\max}$ 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 interimpurity interaction even at $I\to\infty$.

The Curie temperature at the finite interaction energy could be determined by solving Eq. (19). Relevant nonmonotone 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 nonmonotone dependence of the exchange field H_j on γ (cf. Fig. 2). To compare, the dashed line in Fig. 4 reproduces the dependence $\tau_C(n)$ presented in Ref. 6 and obtained in the framework of the standard mean-field theory.

The optimal carrier concentration turns out to be on the

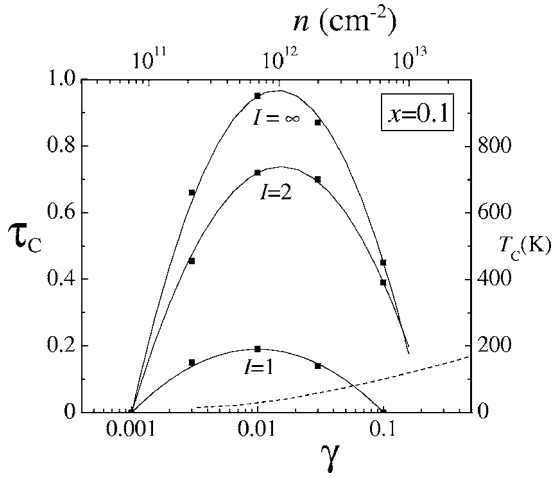

FIG. 4. Dependencies $\tau_C(n)$ of Curie temperature on the carrier concentration for the two-dimensional system with the concentration of magnetic impurities $x=0.1$ for various interaction strengths I. The dashed line is the result presented in Ref. 6 and corresponding to $I \approx 1$.

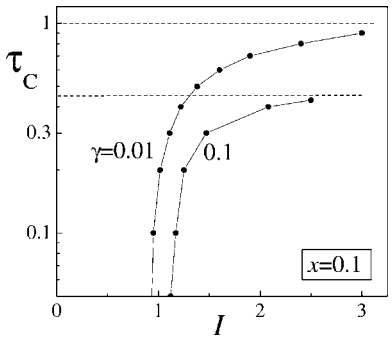

FIG. 5. Dependencies $\tau_C(I)$ of Curie temperature for the two-dimensional system with the concentration of magnetic impurities $x=0.1$ at various carrier concentrations determined by the parameter γ.

order of 10^{12} cm^{-2}, which is significantly lower than the value $n \sim 10^{14}$ cm^{-2} predicted in Ref. 6. In addition, Fig. 5 demonstrates that 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 threshold.

In conclusion, the conditions of establishing the ferromagnetic state and its parameters in quasi-two-dimensional semiconductor systems with magnetic impurities coupled via the RKKY interaction have been studied in this paper. As distinct from Ref. 6, two 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 the 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 turns out to be nonmonotone and there is a threshold interaction strength to drive the system in the ferromagnetic state.

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