Diluted magnetic semiconductors with correlated impurities: Mean-field theory with RKKY interaction

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(Received 18 August 2006; revised manuscript received 31 October 2006; published 17 January 2007)

In the framework of the generalized mean-field theory, the influence of correlating impurities in diluted magnetic semiconductors with indirect RKKY interaction is studied. It is shown that there is the limited range of impurity concentrations x where ferromagnetic ordering is possible and clustering shifts that interval to lower x values. The Curie temperature is a nonmonotone function of x, peaks at $x \sim 0.1$, and its maximum value is slightly influenced by the clustering.

DOI: 10.1103/PhysRevB.75.045204 PACS number(s): 75.50.Pp, 75.10.Nr, 75.30.Hx

I. INTRODUCTION

In most papers dealing with diluted magnetic semiconductors, the distribution of the magnetic atoms (for instance, Mn atoms substituting Ga atoms in GaAs) is considered to be absolutely random.[1–3] That approach ignores the possible correlation of their arrangement in the crystal lattice. However, as calculations show, the interaction of Mn atoms positioned in the neighbor sites of Ga sublattices in $Ga_{1-x}Mn_xAs$ leads to their attraction.[4] The formation of the Mn_2 pair lowers the system energy by the value $\Delta^{(2)} \approx 0.25$ eV (compared to that for the system with two Mn atoms distant from each other). Such a high binding energy promotes associated impurities and prevents their absolute chaotization even in the course of long-term annealing. Thus, in diluted magnetic semiconductors one has always to do with a *correlated* impurity distribution.

The concentration of Mn_2 pairs can be relatively high even without the impurity interaction. In fact, the probability of pair formation equals $p_2=6x(1-x)^5$ where x is the fraction of substituted Ga atoms. Therefrom it follows that already at $x=0.06$ (the typical content of impurities in the magnetic semiconductor) $p_2 \approx 0.3$. The correlation enlarges the concentration of Mn_2 pairs still more (and groups Mn_3 and Mn_4 of three, four atoms, etc., as well) and could even result in the formation of Mn clusters—compact distant complexes of impurity atoms.[5]

In this connection the question arises concerning the influence of the impurity correlation on the properties of diluted magnetic semiconductors. Numerical Monte Carlo calculations of magnetic features of such a system with correlated impurities have been recently carried out in Ref. 6. In that paper, the case of relatively weakly correlated impurities has been considered—the number of atoms in the impurity cluster has not exceeded 4. The calculation has shown that the correlation has a weak influence on the temperature of magnetic ordering. However, to determine the validity limits of that statement analytical model estimations for systems whose parameters are varied in a more broad range are needed. Execution of such calculations forms the purpose of the present work.

II. CORRELATED MAGNETIC MOMENTS

Let one Mn atom be placed in the center of a spherical sample of radius r_{max}. We consider the case of diluted semiconductors ($x \ll 1$) when restrictions applied by the lattice discrecity are not significant and could be reduced to the only requirement—the distance between a given atom and other impurities should be more than a certain minimum spacing r_{min} (the minimal possible distance between magnetic-active Mn ions substituting for Ga atoms in the zinc-blende AsGa lattice equals $r_{min}=a/\sqrt{2} \approx 4$ Å where $a=5.7$ Å is the side of the cubic cell). Thus, impurity atoms could be positioned at any distance $r_{min} \le r \le r_{max}$ from a given atom (placed in the coordinate origin), so the volume of the accessible space equals $V=(4\pi/3)(r_{max}^3-r_{min}^3)$. Then at the random *noncorrelated* arrangement of impurity atoms the distribution function $\kappa_r(r)$ of random interatomic distances is

$$\kappa_r(r) = \frac{4\pi\langle n_{Mn}\rangle r^2}{N_{Mn}}, \tag{1}$$

where $\langle n_{Mn}\rangle$ is the average concentration of Mn atoms and $N_{Mn}=\langle n_{Mn}\rangle V$ is their total number in the sample of radius r_{max}.

Let the average concentration of Mn atoms in the lattice be $\langle n_{Mn}\rangle = x n_{Ga}$ where $n_{Ga}=4/a^3$ is the concentration of Ga sites. It is known[3,7,8] that only certain of them substitute for Ga atoms and introduce in the system their own magnetic moments, so $\langle n_\mu\rangle < \langle n_{Mn}\rangle$. Let the average fraction of those magnetic-active atoms be $x_\mu=\langle n_\mu\rangle/n_{Ga} < x=\langle n_{Mn}\rangle/n_{Ga}$. It is precisely these atoms that are acceptors and deliver mobile charge carriers (holes) with average concentration $\langle p\rangle$ that are responsible for the interaction. However, the equality of average concentrations $\langle n_\mu\rangle=\langle p\rangle$ remains only at low Mn concentrations ($x \lesssim 0.02$) because interstitial Mn atoms and Mn antisite defects are donors.[3,7,8] The resulting compensation leads to a lowering of the carrier concentration compared to that of magnetic-active Mn atoms: $\langle p\rangle = \gamma\langle n_\mu\rangle$ where the coefficient $\gamma < 1$of the impurity efficiency falls with increasing $\langle n_{Mn}\rangle$. One could control the relative hole concentration (i.e., the γ value) by simultaneously introducing nonmagnetic acceptors [for instance, Be (Refs. 9 and 10)] or choosing the temperature of the film growth.[11]

The interaction of magnetic-active impurities could be taken into account completing the distribution (1) by the pair correlation function $g(r)$ (Ref. 12) and making the replacements $\langle n_{Mn}\rangle \to \langle n_\mu\rangle$, $N_{Mn} \to N_\mu$:

$$\kappa_r(r) = C\frac{4\pi\langle n_\mu\rangle r^2 g(r)}{N_\mu}, \quad C = \frac{V}{4\pi\int_{r_{\min}}^{r_{\max}} r^2 g(r)}. \tag{2}$$

As the function $g(r)$ is limited and at $r\to\infty$ $g(r)\to 1$,[12] it could be readily shown that $C\to 1$ at $r_{\max}\to\infty$, so

$$\kappa_r(r) \to \frac{4\pi\langle n_\mu\rangle r^2 g(r)}{N_\mu} \text{ at } r_{\max}\to\infty. \tag{3}$$

The estimation of the correlation function $g(r)$ is a difficult problem requiring knowledge of the spatial dependence of the impurity atom interaction. There are a few successful instances of performing the relevant calculations: the model of hard spheres or the modified Lenard-Jones interaction.[12] But the clear qualitative result for a real attractive potential consists in the following: over the range $r_{\min}<r\lesssim(1-5)r_{\min}$ the correlation function fades with increasing r (not necessarily monotonously) from the initial enhanced value $g=g_0>1$ to the final one $g\approx 1$. Thus, as the *model* correlation function there could be taken, for example, the function

$$g(r) = \begin{cases} 0, & r < r_{\min}, \\ 1 + (g_0 - 1)e^{(r_{\min}-r)/r_c}, & r \geqslant r_{\min}. \end{cases} \tag{4}$$

Here $r_c=(1-5)r_{\min}$ is the correlation radius.

Clustering Mn atoms result in the nonuniformity of the local hole concentration $p(r)$, as well. If the screening length $r_{\mathrm{TF}}\sim r_{\min}$ is shorter than the correlation length r_c, the spatial distribution of holes follows that of Mn atoms. Then the hole concentration near the cluster center should be enhanced by $\approx g_0$ times compared to the average hole concentration $\langle p\rangle$, so

$$p(r) \approx \langle p\rangle \begin{cases} g_0, & r < r_{\min} \\ g(r), & r \geqslant r_{\min}. \end{cases} \tag{5}$$

III. LOCAL MAGNETIC FIELD

It is known that traditional mean-field theory does not provide an adequate description of a disordered system of magnetic moments. In the present paper, we shall use the generalized mean-field theory[13] for systems with an indirect interaction of magnetic impurities taking into account the randomness of their spatial arrangement. We 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 traditional mean-field theory).

For simplicity we use the Ising model corresponding to $S=1/2$ and leading, as is known, to qualitatively correct results at $S\gtrsim 1$, as well. The appropriate generalization does not meet some principal difficulties.

In a uniform system, the energy $w(r)$ of indirect RKKY interaction for two parallel spins $\mathbf{S}_1$ and $\mathbf{S}_2$ of magnetic ions spaced at the distance r is defined by the expression[14,15]

$$w(r) = -J_0\Phi(r)\exp(-r/l),$$
$$\Phi(r) = \left(\frac{a}{r}\right)^4[\varphi(r)\cos\varphi(r) - \sin\varphi(r)], \tag{6}$$

where

$$J_0 = \frac{1}{16\pi^3}\left(\frac{ma^2}{\hbar^2}J_{pd}^2\right), \quad \varphi(r) = 2k_F(r)r, \tag{7}$$

$k_F(r)=[3\pi^2 p(r)]^{1/3}$ is the Fermi momentum of carriers (holes) of concentration p, l is their mean free path, and J_{pd} is the exchange energy for the interaction of a Mn spin with a free charge carrier.[16]

To adjust relation (6) for a nonuniform system, one could replace the phase $\varphi(r)$ by the mean phase

$$\bar{\varphi}(r) = 2\int_0^r k_F(r)dr = 2k_F^0 r_{\min}\left[g_0^{1/3} + \frac{1}{r_{\min}}\int_{r_{\min}}^r g^{1/3}(r)dr\right],$$
$$2k_F^0 r_{\min} = 2(3\sqrt{2}\pi^2 x_\mu\gamma)^{1/3},$$

whereupon the function $\Phi(r)$ assumes the form [taking into account Eq. (4)]

$$\bar{\Phi}(\rho) = \frac{4}{\rho^4}[\bar{\varphi}(\rho)\cos\bar{\varphi}(\rho) - \sin\bar{\varphi}(\rho)],$$
$$\bar{\varphi}(\rho) = 2k_F^0 r_{\min}\left[g_0^{1/3} + \int_1^\rho g^{1/3}(\rho)d\rho\right], \tag{8}$$

where $\rho=r/r_{\min}$.

Let the system consisting of randomly arranged and oriented Ising spins be in the state characterized by the average reduced magnetization $j=2\xi-1$ where ξ is the average fraction of magnetic-active ions with spins directed up. 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 which we shall define by the effective local magnetic field $H=-W/\mu$ (μ is the magnetic moment of the impurity atom) and describe by the distribution function $F(j;H)$ depending on the average concentration n of effective magnetic ions and magnetization j.

Correlation of magnetic impurities and determining the distribution of exchange interactions in spin glasses have attracted a lot of interest. References 18–20 are an example of an ingenious approach to determine that distribution. However, all of them refer to the case of antiferromagnetic interactions of the only sign and are unsuitable for oscillating RKKY interactions. In addition, those methods (typical for percolation models) require a strong (exponential) dependence of the interaction energy on the distance. The method used in the present paper, though more complex and less vivid, is more universal. In particular, it tolerates the clustering of interacting species.

For strongly diluted systems, the distribution function could be found by Markov's method,[21] according to which

$$F(j;H)=\frac{1}{2\pi}\int_{-\infty}^{\infty}A(q)\exp(-iqH)dq,$$

$$A(q)=\lim_{N\to\infty}\left[\sum_{\zeta=\pm1}\int_{r_{\min}}^{r_{\max}}e^{iqh_{\zeta}(r,\zeta)}\kappa_{\zeta}(\zeta)\kappa_r(r)dr\right]^N, \tag{9}$$

where N is the number of magnetic impurities in the integration volume, $h_{\zeta}(r,\zeta)=-\zeta h(r)$, and $h(r)=w(r)/\mu=-(J_0/\mu)\Phi(r)$ is the field generated at the origin by the spin spaced at 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, and $\kappa_{\zeta}(\zeta)$ is the distribution function of the random parameter ζ. In the spirit of mean-field theory, the ζ distribution could be written as

$$\kappa_{\zeta}(\zeta)=[(1-\xi)\delta(\zeta+1)+\xi\delta(\zeta-1)]. \tag{10}$$

As for the r distribution, $\kappa_r(r)$, it is defined by Eq. (3).

Substituting Eqs. (3) and (10) in to Eq. (9) one finds

$$A(q)=\exp[-4\pi\langle n_{\mu}\rangle C(q)],$$

$$C(q)=\int_{r_{\min}}^{\infty}\{1-\cos[qh(r)]-i\cdot j\sin[qh(r)]\}r^2g(r)dr. \tag{11}$$

Relationships (11) 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 (9) the region of high q values is not important. In that approximation,

$$C(q)=Pq^2-ijQq, \tag{12}$$

where

$$P=\frac{1}{2}\int_{r_{\min}}^{\infty}h^2(r)g(r)r^2dr=r_{\min}^3\left(\frac{J_0}{\mu}\right)^2\phi_P(\rho_c,\ell),$$

$$\phi_P(\rho_c,\ell)=\frac{1}{2}\int_1^{\infty}e^{-2\rho/\ell}[1+e^{(1-\rho)/\rho_c}]\bar{\Phi}^2(\rho)\rho^2d\rho,$$

$$Q=\int_{r_{\min}}^{\infty}h(r)r^2g(r)dr=r_{\min}^3\left(\frac{J_0}{\mu}\right)\phi_Q(\rho_c,\ell),$$

$$\phi_Q(\rho_c,\ell)=\int_1^{\infty}e^{-\rho/\ell}[1+e^{(1-\rho)/\rho_c}]\bar{\Phi}(\rho)\rho^2d\rho, \tag{13}$$

and $\rho_c=r_c/r_{\min}$, $\ell=l/r_{\min}$.

Substituting Eqs. (12) and (11) into Eq. (9) we find that in the approach considered the distribution $F(j;H)$ is described by the shifted (relative to $H=0$) Gauss function[22]

$$F(j;H)=\frac{1}{\sqrt{2\pi}\sigma}\exp\left[-\frac{(H-jH_j)^2}{2\sigma^2}\right], \tag{14}$$

$$H_j=-4\pi\langle n_{\mu}\rangle Q,\quad \sigma=(4\pi\langle n_{\mu}\rangle P)^{1/2}. \tag{15}$$

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; i.e., the field promotes, on average, the *ferromagnetic* ordering of magnetic moments.

Relations (15) for the shift H_j of the distribution function $F(j;H)$ and its broadening σ could be rewritten in the form

$$H_j=-4\pi n_{\mu}r_{\min}^3\left(\frac{J_0}{\mu}\right)\phi_Q(\rho_c,\ell),$$

$$\sigma=\left[4\pi n_{\mu}r_{\min}^3\left(\frac{J_0}{\mu}\right)^2\phi_P(\rho_c,\ell)\right]^{1/2}, \tag{16}$$

where $4\pi n_{\min}^3=4\pi x_{\mu}\sqrt{2}$.

It follows herefrom that

$$H_j/\sigma=-(4\pi n_{\mu}r_{\min}^3)^{1/2}\psi(\rho_c,\ell),$$

$$\psi(\rho_c,\ell)=\frac{\phi_Q(\rho_c,\ell)}{[\phi_P(\rho_c,\ell)]^{1/2}}. \tag{17}$$

IV. FERROMAGNETIC-STATE PROPERTIES: MEAN-FIELD APPROACH

Now we will show that the ferromagnetic state is possible at high enough value of the ratio H_j/σ only. In traditional mean-field theory, the distribution function is a δ-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 a straightforward generalization of the equation $j=\tanh[\mu H(j)/kT]$ referring to the regular Ising system:

$$j=\int_{-\infty}^{\infty}\tanh\left[\frac{\mu H}{kT}\right]F(j;H)dH. \tag{18}$$

Using expression (14) for the distribution function $F(j;H)$ one gets an equation generalizing the standard mean-field one:

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

where $\theta=kT/\mu H_j$. That equation predicts the phase diagram of the system and temperature dependences of its magnetization (in the ferromagnetic phase) and susceptibility (in the paramagnetic phase), as well as the dependence of the Curie temperature θ_C on the interaction strength J_0, the relative magnetic ion concentration x_{μ}, and the relative free-carrier concentration $\gamma=\langle p\rangle/n_{\mu}$.

To clarify under what conditions that equation has a solution corresponding to the ferromagnetic state ($j>0$) notice that in the vicinity of the Curie temperature where the mag-

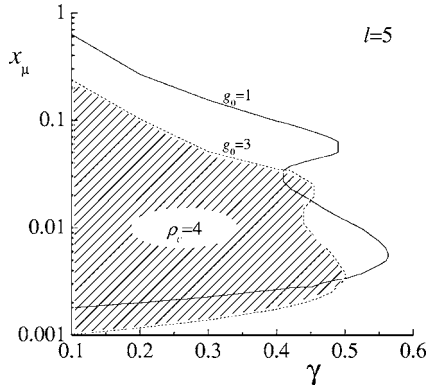

FIG. 1. Bordered areas of the ferromagnetic state in the (x_μ, γ) plane without $(g_0=1)$ and with $(g_0=3, \rho_c=4$, shaded) clustering for $\ell=5$.

netization is small $(j \to 0)$, it follows from Eq. (19) that

$$\sqrt{\frac{2}{\pi}}\left(\frac{H_j}{\sigma}\right)^3 \int_0^\infty \tanh\left(\frac{u}{\theta}\right)\exp\left[-\frac{1}{2}\left(\frac{H_j}{\sigma}\right)^2 u^2\right] u\, du = 1. \tag{20}$$

The integral in Eq. (20) peaks at $\theta=0$, and its maximum value equals $(\sigma/H_j)^2$. It follows herefrom that the ordered state is only possible under the condition

$$\sigma < H_j\sqrt{\frac{2}{\pi}}, \tag{21}$$

which means that in the ferromagnetic state $(j=0)$ the fraction of magnetic ions feeling the antiferromagnetic effective RKKY field should be small enough. (It relates not only to the RKKY interaction but to any alternating-sign interaction, as well.) In fact, under this condition that fraction is smaller than

$$\int_{-\infty}^{0} F(j;H)dH\Big|_{j=1,\sigma=H_j\sqrt{2/\pi}} = \frac{1}{2}[1-\mathrm{Erf}(\sqrt{\pi}/2)] = 0.105;$$

i.e., 10% of unlucky magnetic ions are admissible only.

The condition (21) determines the region of the parameters x_μ and γ whereby the ferromagnetic state is possible. In Fig. 1, those restricted regions are shown for two sets of correlation parameters: $g_0=0$ (no correlation) and $g_0=3$ (with $r_c=3$). For one particular value $\gamma=0.3$, the corresponding dependences of H_j and σ and their ratio are depicted in Fig. 2 (at $\ell=5, \rho_c=4$).

One can see that the correlation moves the region of the ordered magnetic state to significantly lower percentage x_μ of the magnetic ions. The highest x_μ values corresponding, say, to $\gamma=0.3$ equal $x_{\max}\approx 0.15$ for uncorrelated impurities and $x_{\max}\approx 0.06$ for correlated ones.

To confirm our results, one could mention the fact that there are no works where the Curie temperature T_C in GaMnAs would be more than ~170 K whatever the Mn concentration or annealing procedure might be (see the review in Ref. 3 and references therein). Moreover, in Ref. 24 one could see a clear lowering of T_C with increasing $x>0.08$

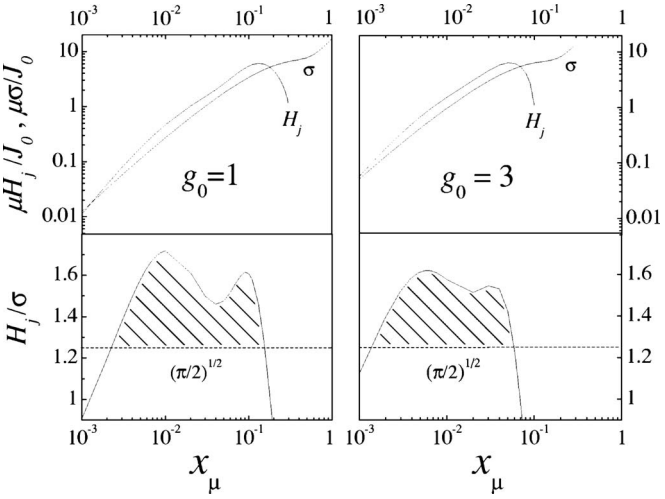

FIG. 2. Parameters H_j and σ of the Gauss distribution function (14) and their ratio at various magnetic ion contents x_μ without $(g_0=1)$ and with $(g_0=3, \rho_c=4)$ clustering for $\ell=5$.

(when the tendency to clustering is enhanced) even after annealing. So one could say it is just the clustering that excludes the ferromagnetism in $Ga_{1-x}Mn_xAs$ at $x \gtrsim 0.1$.

Of course, the latter conclusion is sensitive to variations of the relevant parameters, such as the correlation length ρ_c, the range of RKKY interaction ℓ, and the correlation parameter g_0. Calculations show that increasing either of them results in a shift of both borders of the x_μ area corresponding to the ferromagnetic state to lower values. For instance, the relevant shifts of those borders at $\gamma=0.3$ are demonstrated in Fig. 3.

As for the Curie temperature, one could note that according to Eq. (20) the ratio $k_B T_C/\mu H_j$ is defined by the ratio H_j/σ only. Hence, if the latter ratio is slightly influenced by clustering, then the same is valid for the Curie temperature. As can be seen from Fig. 2, clustering changes those parameters insignificantly, so one should conclude that with the appropriate increase of the concentration x_μ of Mn ions and conservation the γ parameter, T_C in the uniform system is almost the same as in the clustered system. Numerical calculations confirm that conclusion: Figure 4 demonstrates that for the system with, say, $\gamma=0.3$ the Curie temperature is

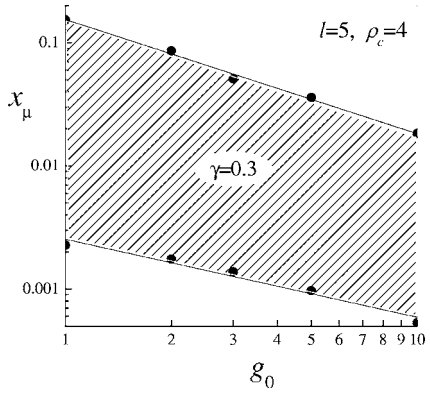

FIG. 3. Shifts of the ferromagnetic area borders for various values of the clustering parameter g_0 $(\rho_c=4)$ at $\gamma=0.3$, $\ell=5$.

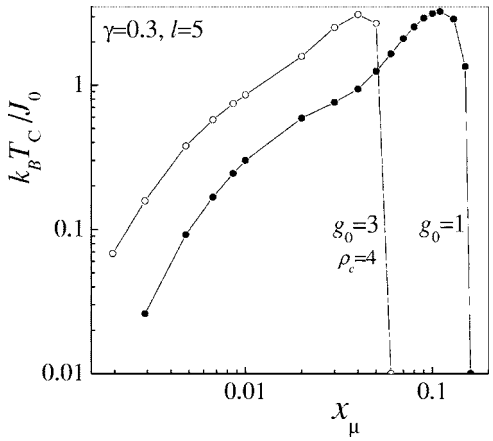

FIG. 4. Concentration dependences of the Curie temperature for the case of $\gamma=0.3$ without ($g_0=1$) and with ($g_0=3$, $\rho_c=4$) clustering at $\ell=5$.

nonmonotone with concentration x_μ. It culminates at $x_\mu \sim 0.1$, and the maximum value equals $T_C \approx 3J_0/k_B$ in both cases (with and without clustering).

In conclusion, the influence of correlating impurities in diluted magnetic semiconductors with indirect RKKY interaction has been studied. It is shown that there is a limited range of impurity concentrations x where ferromagnetic ordering is possible and the clustering shifts that interval to lower x values. The Curie temperature is a nonmonotone function of x, peaks at $x \sim 0.1$, and its maximum value is slightly influenced by the clustering.

ACKNOWLEDGMENTS

This work has been supported by Grants No. 06-02-116313, 07-02-00927 of the Russian Foundation of Basic Researches and the ISTC Project No. G-1335.

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