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A Green's function model for ferromagnetism and spin excitations of (Ga, Mn)As diluted magnetic semiconductors*

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We study (Ga, Mn)As diluted magnetic semiconductors in terms of the Ruderman–Kittel–Kasuya–Yosida quantum spin model in Green's function approach. Random distributions of the magnetic atoms are treated by using an analytical average of magnetic configurations. Average magnetic moments and spin excitation spectra as functions of temperature can be obtained by solving self-consistent equations, and the Curie temperature $T_{\rm C}$ is given explicitly. $T_{\rm C}$ is proportional to magnetic atomic concentration, and there exists a maximum for $T_{\rm C}$ as a function of carrier concentration. Applied to (Ga, Mn)As, the theoretical results are consistent with experiment and the experimental $T_{\rm C}$ can be obtained with reasonable parameters. This modelling can also be applied to other diluted magnetic semiconductors.

Keywords: diluted magnetic semiconductor, Green's function, Ruderman–Kittel–Kasuya–Yosida
PACC: 7540B, 7540M, 7550P

1. Introduction

Diluted magnetic semiconductors (DMSs) have been studied for three decades in order to search for thermodynamically stable materials with both traditional semiconductor characteristics and robust high-temperature ferromagnetism for new multifunctional electronic devices.[1,2] $Ga_{1-x}Mn_xAs$ is one of the most important DMS systems. Recent years have witnessed two breakthroughs in its Curie temperature. A Curie temperature of 110 K ($x=0.053$)[3] was achieved in 1998 and 173 K ($x=0.08$)[4] in 2005. It is reasonably believed that higher Curie temperatures may be achieved in the future.

The ferromagnetism in $Ga_{1-x}Mn_xAs$ systems has been extensively studied.[5,6] Many methods have been used to calculate the magnetic properties, especially the Curie temperature $T_{\rm C}$, of $Ga_{1-x}Mn_xAs$, including the mean field method combined with virtual crystal approximation (MF-VCA),[3,7−11] the self-consistent local random phase approximation method,[12−15] the first-principles method,[16−18] the Monte Carlo method,[19,20] and some others.[21] The famous MF-VCA method gives $T_{\rm C} \propto p^{1/3}$ (p is the carrier concentration of p-type semiconductors). However, Bouzerar *et al*[15] claimed that MF-VCA is not suitable for studying ferromagnetism in diluted magnetic systems because it leads to strong overestimation of $T_{\rm C}$, and the reasonable $T_{\rm C}$ should at first increase and then decrease with the increase of hole concentration p. The widely accepted point of view is that the ferromagnetism stems from some indirect ferromagnetic coupling among local magnetic moments of Mn^{2+} ($S = 5/2$) which is mediated by itinerant carriers ('holes' here). Such effective spin interactions and consequent ferromagnetism in $Ga_{1-x}Mn_xAs$ DMS[3,5,7,8] and some others that are similar can be well described by the Ruderman–Kittel–Kasuya–Yosida (RKKY) quantum spin model.[22]

In this paper, we use Green's function approach to investigate the RKKY model and make configuration average analytically to deal with spin disorder. Using Tyablikov decoupling routine,[23] we can successfully solve self-consistent equations of Green's functions. Average magnetic moment is determined by a self-consistent nonlinear equation set from zero to Curie temperature. 3/2 temperature behaviour is obtained for the low temperature moment. A reasonable average moment is obtained for the whole temperature region up to Curie temperature $T_{\rm C}$. Our $T_{\rm C}$

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of this approach is much better than the mean-field result. With the magnetic atom concentration fixed, the $T_{\rm C}$ increases firstly with the increase of carrier concentration and then decreases to zero when carrier concentration reaches 0.27. With the carrier concentration fixed, the $T_{\rm C}$ is proportional to the magnetic atom concentration. The temperature effect of the carrier concentration is also studied. Applying this approach to the $Ga_{1-x}Mn_xAs$ system and using experimental parameters, we obtain reasonable $T_{\rm C}$ values and experimental trends of magnetic moments with changing temperature and other parameters. This Green's function plus configuration average method can be used to study effective spin models for other DMSs.

The remaining parts of this paper are organized as follows. In Section 2, we shall present our configuration-averaged Green's function approach to the RKKY spin model. In Section 3, we shall work out the spin excitation spectra and the self-consistent equations of the average moments, and furthermore present the phase-transition temperatures. In Section 4, we shall investigate the case of $Ga_{1-x}Mn_xAs$ and compare our calculated results with the experiments concerned. We shall give our conclusion in Section 5. In Appendix we shall give a brief explanation of the configuration average.

2. Green's function approach to the RKKY model

We use the standard spin RKKY Hamiltonian

$$\hat{H} = -\frac{1}{2}{\sum_{i\neq j}}' J_{ij}\hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j, \tag{1}$$

where J_{ij} is the RKKY effective spin exchange interaction[24] between local spin $\hat{\boldsymbol{S}}_i$ and $\hat{\boldsymbol{S}}_j$ ($\hat{\boldsymbol{S}}_i$ and $\hat{\boldsymbol{S}}_j$ are both quantum spin operators),

$$J_{ij} = -\frac{J_{\rm pd}^2 m^* k_{\rm F}^4}{2\pi^3\hbar^2} F(2k_{\rm F}R_{ij}), \tag{2}$$

where $F(x)$ is an oscillating function defined by $F(x) = [x\cos(x) - \sin(x)]/x^4$. The coupling constant $J_{\rm pd}$ in Eq.(2) is the local coupling of the itinerant spin to the local magnetic moment, m^* is the effective mass of the carrier, $k_{\rm F}$ is the Fermi wave vector ($k_{\rm F}^3 = 3\pi^2 p$ under free electron gas approximation), and R_{ij} is the distance between local magnetic moments at $\boldsymbol{R}_i$ and $\boldsymbol{R}_j$. Here, the magnetic atoms are distributed randomly in the full lattice of the undoped semiconductor. The prime for the summation in Eq.(1) means that the summation is done only over the sites occupied by magnetic impurities.

Hence, we use Green's function approach[25] to treat the model Eq.(1). In this scheme one uses a double-time Green's function $\langle\langle \hat{A}(t);\ \hat{B}(t')\rangle\rangle$ which satisfies the following equation of motion:

$$\begin{aligned}\mathrm{i}\,\hbar\frac{\mathrm{d}}{\mathrm{d}t}\langle\langle \hat{A}(t);\hat{B}(t')\rangle\rangle &= \delta(t-t')\langle[\hat{A}(t),\hat{B}(t)]\rangle \\ &\quad + \langle\langle[\hat{A}(t),\hat{H}];\hat{B}(t')\rangle\rangle.\end{aligned} \tag{3}$$

This approach proves successful for various Heisenberg spin models on regular lattices.[25–29] Usually, one needs more than one Green's function when spin S is larger than 1/2.[26–30] For our random spin system we construct double-time spin Green's functions between spin operators at two magnetic sites $\boldsymbol{R}_i$ and $\boldsymbol{R}_j$,

$$\widetilde{G}_{ij}^n(t,t') = \langle\langle \hat{S}_i^+(t);(\hat{S}_j^-(t'))^n(\hat{S}_j^+(t'))^{n-1}\rangle\rangle, \tag{4}$$

where n takes 1, 2, 3, ... , $2S$. As a rule, we needs $2S$ independent Green's functions for spin S. They can be expressed as

$$\widetilde{G}_{ij}^n(t,t') = \frac{1}{2\pi\hbar}\int G_{ij}^n(\omega)\mathrm{e}^{-\mathrm{i}\,\omega(t-t')/\hbar}\mathrm{d}\omega, \tag{5}$$

because the Hamiltonian Eq.(1) is time independent. Assuming that each local spin has the same average z-component $\langle\hat{S}^z\rangle$ and making a Tyablikov cutoff approximation,[23] we derive the equation of motion

$$\left(\omega - \langle\hat{S}^z\rangle{\sum_{l(\neq i)}}' J_{il}\right)G_{ij}^n = \delta_{ij}C_n - \langle\hat{S}^z\rangle{\sum_{l(\neq i)}}' J_{il}G_{lj}^n, \tag{6}$$

where the constant C_n is given by

$$C_n \equiv \langle(2n\langle\hat{S}^z\rangle + n^2 - n)(\hat{S}^-)^{n-1}(\hat{S}^+)^{n-1}\rangle. \tag{7}$$

Generally speaking, the full lattice is always regular and has translational symmetry. As long as all its sites are occupied by local spins (see Fig.A1(a) in appendix), Fourier transformation can be done directly between real space and k space for the Green's functions because the identities

$$\frac{1}{N}\sum_{\boldsymbol{k}\in\mathrm{BZ}}\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot(\boldsymbol{R}_i-\boldsymbol{R}_j)} = \delta_{ij} \tag{8}$$

and

$$\frac{1}{N}\sum_{j=1}^{N}\mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{R}_j} = \delta_{\boldsymbol{k}\boldsymbol{k}'} \tag{9}$$

can be used, where BZ denotes the first Brillouin zone of the full lattice and N is the total number of the

lattice sites. When local spins are distributed randomly in the lattice (see Fig.A1(b) in Appendix), the $\boldsymbol{k}$ vector can still be used. In fact, it can be accessible experimentally although dopants are distributed randomly. As a result, Eq.(8) is still correct, although the summation

$$\frac{1}{N'}\sum_{j=1}^{\prime N'} \mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{R}_j}$$

is not equivalent to $\delta_{\boldsymbol{k}\boldsymbol{k}'}$ because all the $\boldsymbol{R}_j$ ($1 \le j \le N'$) here make only a subset of all the sites of the full lattice. Here N' is the total number of the local spins in a given spin configuration and we always have $N' \le N$.

Rigorously speaking, we have a given spin configuration for a specific sample. It appears that we have to calculate the prime summations over the given spin configuration in order to calculate the Green's functions. Actually, we do not need to do it because the $\boldsymbol{k}$ vector is still usable and experimentally accessible for doped systems. Accordingly, what we need is the average of the prime summation over all the possible spin configurations. More details about this configuration average are given in the appendix.

After using the configuration average, the Fourier transformation in Eq.(6) can be completed and the Green's functions are obtained as

$$G_{ij}^n = \sum_{\boldsymbol{k}} g_{i\boldsymbol{k}}^n \mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{R}_j}, \quad g_{\mathrm{i}\,\boldsymbol{k}}^n = \sum_{\boldsymbol{k}'} g_{\boldsymbol{k}'\boldsymbol{k}}^n \mathrm{e}^{\mathrm{i}\,\boldsymbol{k}'\cdot\boldsymbol{R}_i}. \quad (10)$$

Here $g_{\boldsymbol{k}'\boldsymbol{k}}^n$ is expressed as

$$g_{\boldsymbol{k}'\boldsymbol{k}}^n = \frac{1}{N}\frac{C_n\delta_{\boldsymbol{k},-\boldsymbol{k}'}}{\omega - \langle\hat{S}^z\rangle N'(J_{\boldsymbol{0}} - J_{\boldsymbol{k}'})}, \quad (11)$$

where

$$J_{\boldsymbol{k}} = \frac{1}{N'^2}\left\langle \sum_{i,l=1}^{N'}{}' J_{il}\mathrm{e}^{-\mathrm{i}\,\boldsymbol{k}\cdot(\boldsymbol{R}_i-\boldsymbol{R}_l)}\right\rangle_{\mathrm{c}}. \quad (12)$$

According to Eqs.(11) and (12), the spin excitation energy spectrum is given by

$$E(\boldsymbol{k}) = \langle\hat{S}^z\rangle\frac{1}{N'}\left\langle \sum_{i\neq l}^{N'}{}' J_{il}\left[1-\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot(\boldsymbol{R}_l-\boldsymbol{R}_i)}\right]\right\rangle_{\mathrm{c}} \quad (13a)$$

$$= \langle\hat{S}^z\rangle\frac{1}{N'}\frac{N'(N'-1)}{N(N-1)}\sum_{i\neq l}^{N} J_{il}\left[1-\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot(\boldsymbol{R}_l-\boldsymbol{R}_i)}\right] \quad (13b)$$

$$= \langle\hat{S}^z\rangle\frac{N'-1}{N(N-1)}\sum_{i=1}^{N}\sum_{\boldsymbol{\delta}} J_{i,i+\boldsymbol{\delta}}(1-\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{\delta}}), \quad (13c)$$

in which the equality Eq.(13b) comes from Eq.(A4) in Appendix, and the equality Eq.(13c) is obtained through making a cutoff approximation for the range of the RKKY exchange interaction. $\boldsymbol{\delta}$ means a neighbour of the lattice site $\boldsymbol{R}_i$. The RKKY exchange interaction J_{il} is taken into account only up to the first zero point, $2k_{\mathrm{F}}R = 4.49$, of the oscillating function $F(2k_{\mathrm{F}}R)$ in our calculations.

3. Spin excitations, magnetic moments and Curie temperatures

To evaluate $\langle\hat{S}^z\rangle$ and T_{C}, the key point is to simplify $E(\boldsymbol{k})$ in Eq.(13c) so that specific calculations can be performed. The resulting spin excitation spectra are given by

$$E(\boldsymbol{k}) = \langle\hat{S}^z\rangle\frac{N'-1}{N(N-1)}N\sum_{\boldsymbol{\delta}} J_{0,\boldsymbol{\delta}}(1-\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{\delta}}) \quad (14a)$$

$$= x_m\langle\hat{S}^z\rangle\sum_{\boldsymbol{\delta}} J(|\boldsymbol{\delta}|)(1-\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{\delta}}) + O\left(\frac{1}{N}\right). \quad (14b)$$

Here the equality Eq.(14a) is obtained because the $\boldsymbol{\delta}$ summation is independent of the specific lattice site. x_{m} in Eq.(14b), defined as N'/N, is the dimensionless effective concentration of magnetic impurities. Figure 1 shows this energy spectrum along three different directions: [100], [110] and [111]. The carrier concentration x_p is set to 0.1 and 0.0068 (for simplicity, we define a dimensionless carrier concentration $x_p \equiv p/N_0$, where N_0 is the concentration of primitive cells, being $4/a^3$ for an fcc lattice with lattice constant a). The energy spectra are scaled with a constant $E_0 = e_0 x_{\mathrm{m}}\langle\hat{S}^z\rangle$, where e_0 is defined as $e_0 = J_{\mathrm{pd}}^2 m^*/(a^4\hbar^2)$.

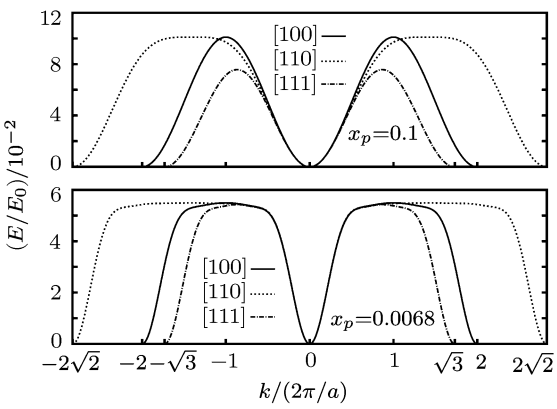


Fig.1. Spin excitation spectra, $E(\boldsymbol{k})/E_0$, along the [100] (solid), [110] (dash) and [111] (dot-dash) directions. The curves are plotted for the ranges $\left[-2\frac{2\pi}{a},\ 2\frac{2\pi}{a}\right]$, $\left[-2\sqrt{2}\frac{2\pi}{a},\ 2\sqrt{2}\frac{2\pi}{a}\right]$ and $\left[-\sqrt{3}\frac{2\pi}{a},\ \sqrt{3}\frac{2\pi}{a}\right]$, respectively. The carrier concentration x_p is taken as 0.1 (upper panel) and 0.0068 (lower panel).

Using the spectrum theorem, we derive the correlation functions

$$\langle(\hat{S}^-)^n(\hat{S}^+)^n\rangle = \Phi(\langle\hat{S}^z\rangle)C_n, \tag{15}$$

where the Φ factor is given by

$$\Phi(\langle\hat{S}^z\rangle) = \frac{1}{N}\sum_{\boldsymbol{k}}\frac{1}{\mathrm{e}^{E(\boldsymbol{k})/k_{\mathrm{B}}T}-1}. \tag{16}$$

Eventually, $\langle\hat{S}^z\rangle$ from zero to the phase transition temperature can be evaluated self-consistently in terms of Eq.(16) and the Callen expression[31]

$$\langle\hat{S}^z\rangle = \frac{(S-\Phi)(1+\Phi)^{2S+1}+(S+1+\Phi)\Phi^{2S+1}}{(1+\Phi)^{2S+1}-\Phi^{2S+1}}. \tag{17}$$

It can be shown that $E(\boldsymbol{k})$ is proportional to k^2 in the neighbourhoods of the Γ point ($k=0$). This leads to the famous low-temperature behaviour,

$$\langle\hat{S}^z\rangle \approx \langle\hat{S}_0^z\rangle - cT^{3/2}, \tag{18}$$

where c is a constant independent of temperature T. While temperature approaches T_{C}, we have $\langle\hat{S}^z\rangle\to 0$, and furthermore $E(\boldsymbol{k})\to 0$ and $\Phi\to\infty$. Expanding Eq.(16) and Eq.(17), we derive

$$\langle\hat{S}^z\rangle^2 = \frac{\frac{4}{3}S^2(S+1)^2\left(\frac{T}{T_{\mathrm{C}}}\right)^2}{\eta(1)\eta(-1)+\frac{4}{5}S(S+1)-\frac{3}{5}}\left(1-\frac{T}{T_{\mathrm{C}}}\right), \tag{19}$$

which means a square-root behaviour

$$\langle\hat{S}^z\rangle \propto \sqrt{1-\frac{T}{T_{\mathrm{C}}}} \tag{20}$$

when temperature approaches T_{C}. Here the Curie temperature T_{C} is defined as

$$T_{\mathrm{C}} = \frac{S(S+1)}{3k_{\mathrm{B}}\eta(-1)}. \tag{21}$$

The constants $\eta(q_i)$ ($q_i=1$ and –1) in the above equations are defined as

$$\eta(q_i) = \frac{1}{N}\sum_{\boldsymbol{k}}\left[\frac{E(\boldsymbol{k})}{\langle\hat{S}^z\rangle}\right]^{q_i}. \tag{22}$$

Equation (21) is a general expression for the Curie temperature T_{c}. Inserting Eqs.(2), (14b) and (22) into Eq.(21), we obtain the following full expression of T_{C} for direct evaluations,

$$T_{\mathrm{C}} = \frac{\frac{S(S+1)e_0x_{\mathrm{m}}(12\pi^2x_p)^{\frac{4}{3}}}{12\pi^3k_{\mathrm{B}}}}{\frac{1}{N}\sum_{\boldsymbol{k}}\frac{1}{-\sum_{\boldsymbol{R}}F\left((12\pi^2x_p)^{1/3}\frac{2R}{a}\right)[1-\mathrm{e}^{\mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{R}}]}}. \tag{23}$$

It can be shown that when x_p is fixed, the phase-transition temperature T_{C} is proportional to $x_{\mathrm{m}}m^*J_{\mathrm{pd}}^2$, which is the same as what was obtained with the MF-VCA method, but this T_{C} is not a simple function of the carrier concentration x_p. Actually, our T_{C} at first increases with x_p and then decreases down to 0 when $x_p>0.27$, and hence has a maximum at a fixed value $x_p=0.03$, as shown in Fig.2. It is easy to understand why ferromagnetism disappears at large x_p within the RKKY scheme. Qualitatively, when x_p is large enough, the oscillation period of the RKKY exchange interaction becomes very small and the effective coupling length of the interaction becomes less than the average distance between two local magnetic moments, which leads to a 'frustrated RKKY'[5] interaction and the breaking of ferromagnetic order. The T_{C} kinks at $x_p=0.052$ and 0.096 are because of the jumps of the total number of the neighbouring sites resulting from the cutoff of $F(2k_{\mathrm{F}}R)$. Quantitatively, while doing the summation in Eq.(14b), we make a cutoff at $2k_{\mathrm{F}}R_{\mathrm{cut}}=4.49$, which means

$$R_{\mathrm{cut}} = 2.245(12\pi^2x_p)^{-1/3}a. \tag{24}$$

It can be clearly seen that R_{cut} decreases with x_p. When $R_{\mathrm{cut}}<\sqrt{2}a/2$ ($\sqrt{2}a/2$ is the distance between the nearest neighbours for the fcc lattice), the total number of the neighbours to be summed becomes zero, that is to say, there is no effective exchange interaction between spins, and hence ferromagnetism disappears. Letting $R_{\mathrm{cut}}=\sqrt{2}a/2$, we obtain the critical value of $x_p=0.27$ shown in Fig.2. Similarly, letting R_{cut} equal the distance between the next-nearest neighbours a and between the third-nearest neighbours $\sqrt{6}a/2$, we obtain the two special values, $x_p=0.096$ and $x_p=0.052$, respectively. They correspond to the two kinks in Fig.2.

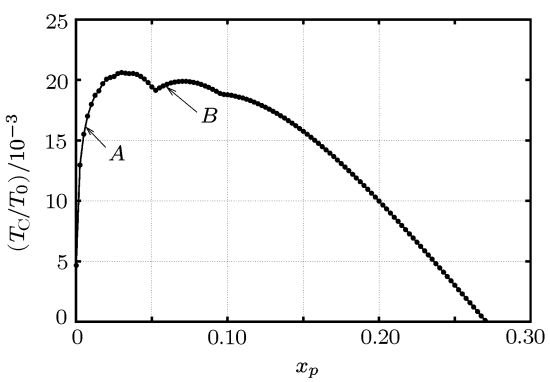


Fig.2. Curie temperature T_{C} vs x_p, where T_{C} is scaled with $T_0=x_{\mathrm{m}}S(S+1)e_0/k_B$. Point A corresponds to $x_p=0.0068$ and point B $x_p=0.064$. T_{C} has a peak at $x_p=0.03$ and vanishes when $x_p>0.27$.

It is known that carrier concentration is a function of temperature in traditional semiconductors. Hence we further investigate the dependence of $T_{\rm C}$ on x_{p_0}. The temperature function of the carrier concentration takes the form

$$p = p_0 \mathrm{e}^{-\frac{\Delta E}{k_{\rm B}T}} \quad \text{or} \quad x_p = x_{p_0} \mathrm{e}^{-\frac{\Delta E}{k_{\rm B}T}}, \qquad (25)$$

where p_0 is the maximal concentration when $T \to \infty$, x_{p_0} is defined as p_0/N_0, and ΔE is the activation energy of the carriers. We use the experimental data $(T, p) = (10\ \mathrm{K}, 1 \times 10^{20}\ \mathrm{cm}^{-3})$ for $x = 0.053$ from Ref.[32] and assume $x_{p_0} = x$, and then we get $\Delta E/k_{\rm B} = 24.6$ K. After considering that x_p varies with T, we have to solve Eqs.(23) and (25) self-consistently to calculate $T_{\rm C}$. Shown in Fig.3 are three curves of $T_{\rm C} - x_{p_0}$ for three different ΔE values: 0, 2.1 and 5.0 meV. By confining $x_{p_0} = \alpha\, x_{\rm m}$ $(0 < \alpha \leq 1)$, we obtain $T_{\rm C} - x_{\rm m}$ relations for $\alpha = 1$ and 0.8, as shown in Fig.4.

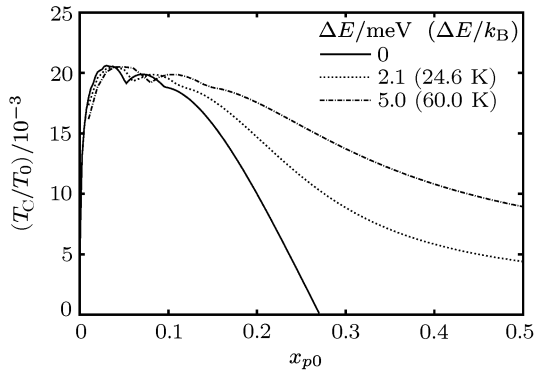


Fig.3. Curie temperature $T_{\rm C}$ vs. x_{p_0} with three different ΔE values: 0, 2.1, and 5.0 meV. $T_{\rm C}$ is scaled with $T_0 = x_{\rm m} S(S+1) e_0/k_{\rm B}$.

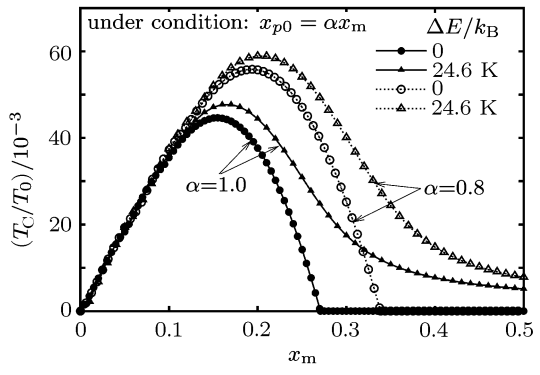


Fig.4. Curie temperature $T_{\rm C}$ vs. $x_{\rm m}$ under the condition $x_{p_0} = \alpha x_{\rm m}$, where α is taken as 1.0 (solid) and 0.8 (dash). $\Delta E/k_{\rm B}$ is set to 0 (circles) and 24.6 K (triangles). $T_{\rm C}$ is scaled with $T_0 = x_{\rm m} S(S+1) e_0/k_{\rm B}$.

4. Application to the (Ga, Mn)As system

As have been shown experimentally, impurities in $Ga_{1-x}Mn_xAs$ are mainly substitutional Mn_{Ga}, along with some interstitial Mn_I and As antisites. The Mn_{Ga} acts as both the local magnetic moment with 5 $\mu_{\rm B}$ and the acceptor for a hole carrier. The Mn_I compensates both carriers and local magnetic moments from Mn_{Ga}, and the As antisite also compensates holes as a double donor like Mn_I. However, the Mn_I can be fully removed by annealing[33] and the As antisite concentration can be reduced effectively by using As dimers instead of As tetramers.[34] Accordingly, the effective concentration $x_{\rm m}$ for magnetically active Mn impurities (substitutional Mn) is less than or equal to the nominal doping concentration x in $Ga_{1-x}Mn_xAs$.

Before calculations, we have to point out that one of the most important parameters in the RKKY interaction is the local coupling constant between Mn magnetic moments and carriers, $J_{\rm pd}$ (or $N_0\beta$).[5,35,36] Its value has several versions due to different authors. Exciton splitting results at low concentration ($x < 0.001$) gave $N_0\beta = 2.5$ eV,[37] magnetotransport results vary from $|N_0\beta| = 3.3$ eV[3] to $|N_0\beta| = 1.5$ eV,[38] a core-level photoemission study showed $N_0\beta = -1.2$ eV[39] and first-principles calculations gave values from $N_0\beta = -2.8$ eV[40] to $N_0\beta = -4.5$ eV.[17] It is clear that concurrence has not been reached until now. As shown below, our model will get reasonable results compared with experiments if we assume a resonable exchange constant $|N_0\beta| = 2.2$ eV (or $J_{\rm pd} = 100$ eV·Å^3).

We calculate $T_{\rm C}$ using Eq.(23) with parameters $x_{\rm m}$ and x_p from Refs.[3] and [4]. Specifically, we use spin $S = 5/2$, effective mass $m^* = 0.5 m_{\rm e}$,[3] and lattice constant $a = 5.65$ Å (the value of undoped GaAs with zincblende structure). For the sample in Ref.[3], we simply take $x_{\rm m} = x = 0.053$, because no specific information about the fraction of substitutional Mn

Table 1. Experimental parameters ($x_{\rm m}$ and x_p), experimental Curie temperatures $T_{\rm C}^{\rm exp}$, and our calculated Curie temperatures $T_{\rm C}^{\rm calc}$ if we use $J_{\rm pd}$=100 eV·Å^3 (or $|N_0\beta|$=2.2 eV). The parameters in case No 1 are from Ref.[3] and those in case No 2 are based on Ref.[4].

No	$x_{\rm m}$	$p/10^{20}\mathrm{cm}^{-3}$	x_p	$T_{\rm C}^{\rm exp}$/K	$T_{\rm C}^{\rm calc}$/K
1	0.053	1.5	0.0068	110	113
2	0.056	12.4	0.0560	173	140
	0.060	13.5	0.0600	173	151
	0.064	14.1	0.0640	173	163

in all Mn impurities was presented. For another sample, Ref.[4] explicitly gave $x = 0.08$, $x_{\rm m}/x = 0.70 \sim 0.80$, and $x_p \sim x_{\rm m}$ and hence we take three values of $x_{\rm m}$: 0.056, 0.060 and 0.064. We can see from Table 1 that calculated Curie temperatures have good agreements with experimental values if we assume $|N_0\beta| = 2.2$ eV.

Figure 5 shows the average magnetic moments versus temperature T, calculated with the parameters (shown by point A and point B in Fig.2) from Refs.[3] and [4], respectively. The curves of the average moments versus temperature qualitatively have the same shape as that in Ref.[4].

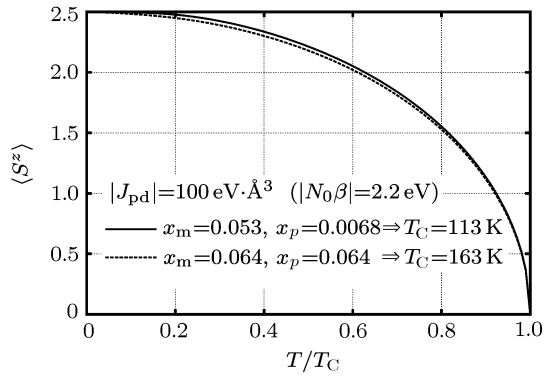


Fig.5. $\langle \hat{S}^z \rangle$ vs $T/T_{\rm C}$ with two sets of the parameters $(x_{\rm m}, x_p) = (0.053, 0.0068)$ (solid) and (0.064,0.064) (dash). The parameters for the two curves are taken from Refs.[3] and [4] (refer to Table 1) respectively.

Our results support the prediction that room temperature ferromagnetism in $Ga_{1-x}Mn_xAs$ will be reached at $x_{\rm m} = 10\%$.[5,11] The results imply that $T_{\rm C}$ can be increased by adjusting x_p toward the peaked value 0.03 (or $p = 6.7 \times 10^{20}$ cm^{-3}) and enlarging the active local magnetic moment concentration. By codoping other elements, i.e., Sn with Mn, the effective local magnetic moment concentration and the carrier concentration can be controlled independently.[41] This may be a possible approach to achieve $T_{\rm C}$ above room temperature.

5. Conclusion

We have used a double-time Green's function approach to study the RKKY model and meanwhile have introduced an analytical configuration average method to treat the random distributions of magnetic moments. Quantum effects are taken into account and spin excitation spectra are explicitly derived in this approach. This is why we can obtain the 3/2 low-temperature moment behaviour. The average magnetic moment is determined self-consistently and is reasonable for the whole temperature range from zero to Curie temperature $T_{\rm C}$. $T_{\rm C}$ is proportional to the active magnetic moment concentration and the squared direct spin exchange constant. It first increases and then decreases with increasing carrier concentration. The thermal-activated effect of the carriers is also investigated.

Applying the Green's function plus configuration average approach to the $Ga_{1-x}Mn_xAs$ system and using the experimental parameters in Refs.[3] and [4], we obtain reasonable Curie temperatures and average magnetic moments if we use $|N_0\beta| = 2.2$ eV. A good agreement with experiments is obtained considering the fact that some parameters cannot be accurately determined at this stage. Our results confirm that the RKKY model is a good description for a carrier mediated DMS such as $Ga_{1-x}Mn_xAs$ and its $T_{\rm C}$ can be boosted further to room temperature by increasing the effective moment concentration and controlling the carrier concentration. It is certain that our Green's function plus configuration average method can be applied to other carrier-mediated DMSs with RKKY effective spin interactions, such as (In, Mn)As,[5] (Ga, Mn)Sb,[5,42] (In, Mn)Sb,[5] (Zn, Cr)Te,[5,43−46] p-(Be, Mn)Te:N,[5,47] and p-(Zn, Mn)Te:N.[5,8,42] Actually, this method can be applied to other effective spin models beyond the RKKY interaction for other ferromagnetic semiconductors.

Appendix: Configuration average

For an independent prime summation f, we can define its configuration average over all the spin configurations as

$$\langle f \rangle_{\rm c} = \frac{1}{\Omega} \sum_{c_i \in \mathbb{C}} f(c_i), \tag{A1}$$

where $\mathbb{C}$ is the set of all the Ω different spin configurations of the same number of magnetic moments. For a configuration average on a prime summation over a single index such as $\left\langle \sum_{i=1}^{N'}{}' A_i \right\rangle_{\rm c}$, we have

$$\begin{aligned} \left\langle \sum_{i=1}^{N'}{}' A_i \right\rangle_{\rm c} &= \frac{1}{\Omega} \left[\sum_{c_j \in \mathbb{C}} \left(\sum_{i=1}^{N'}{}'_{c_j} A_i \right) \right] \\ &= \frac{1}{\Omega} \left[\frac{\Omega N'}{N} \sum_{i=1}^{N} A_i \right] \\ &= \frac{N'}{N} \sum_{i=1}^{N} A_i \end{aligned} \tag{A2}$$

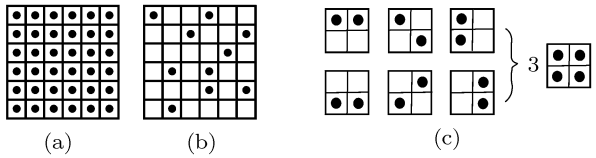


Fig.A1. (a) The lattice formed by the cations of the undoped semiconductor. The total number of the sites is N. The full configuration is defined by occupying each site with a local spin. (b) A given spin configuration. There are N' randomly distributed spins, where $N' < N$. (c) A demonstration of the configuration average of a summation over a single index with $N = 4$ and $N' = 2$. In this case we have six spin configurations, $\Omega = 6$, and they correspond to $\Omega N'/N$ full configurations when the prime summation is changed into the regular summation over the full configuration.

where $\sum'_{c_j}$ means that the summation is done over spin configuration c_j ($c_j \in \mathbb{C}$), as demonstrated in Fig.A1. As a result, we derive

$$\left\langle \sum_{j=1}^{N'}{}' \mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{R}_j} \right\rangle_{\mathrm{c}} = \frac{N'}{N}\sum_{j=1}^{N} \mathrm{e}^{\mathrm{i}(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{R}_j} = N'\delta_{\boldsymbol{k}\boldsymbol{k}'}. \tag{A3}$$

As for a configuration average on a prime summation over double indexes such as $\left\langle \sum'^{N'}_{i\neq j} B_{ij} \right\rangle_{\mathrm{c}}$, the configuration average can be performed as follows

$$\begin{aligned}\left\langle \sum_{i\neq j}^{N'}{}' B_{ij} \right\rangle_{\mathrm{c}} &= \frac{1}{\Omega}\Big[\sum_{c_k\in\mathbb{C}} \Big(\sum_{i\neq j}^{N'}{}'_{c_k} B_{ij} \Big) \Big] \\ &= \frac{1}{\Omega}\left[\Omega \frac{N'(N'-1)}{N(N-1)} \sum_{i\neq j}^{N} B_{ij} \right] \\ &= \frac{N'(N'-1)}{N(N-1)} \sum_{i\neq j}^{N} B_{ij}. \end{aligned} \tag{A4}$$

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