Carrier-induced ferromagnetism in diluted magnetic semiconductors

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We present a theory for carrier-induced ferromagnetism in diluted magnetic semi-conductor (DMS). Our approach treats on equal footing both quantum fluctuations within the random-phase approximation and disorder within the coherent potential approximation (CPA). This method allows for the calculation of T_c, magnetization, and magnon spectrum as a function of hole, impurity concentration, and temperature. It is shown that, sufficiently close to T_c and within our decoupling scheme (Tyablicov type), the CPA for the itinerant electron gas reduces to the virtual crystal approximation. This allows, in the low-impurity concentration and low density of carriers, to provide analytical expression for T_c. For illustration, we consider the case of $Ga_{1-c}Mn_cAs$ and compare our results with available experimental data.

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The discovery of carrier-induced ferromagnetism in diluted magnetic semiconductor (DMS) has attracted considerable attention from both theoreticians and experimentalists. The interest in these material is mainly stimulated by the possible technological applications (e.g. semiconductor spin devices). For example by doping GaAs (Refs. 1 and 2) with magnetic impurities Mn^{2+}, T_c exceeding 100 K has been reached. The doping of a III-V semiconductor compound with Mn impurities introduces simultaneously local magnetic moments ($S=5/2$) and itinerant valence-band carriers ($s=1/2$). One of the important open issues is to find out whether it is possible to reach critical Curie temperature of order 300 K. Thus it is important to understand theoretically how T_c varies with the impurity concentration, effective mass, hole concentration, and exchange integral. Many theoretical approaches have been performed to analyze ferromagnetism in DMS, this includes mean-field theory,[3–5] spin-wave theory,[6] first-principle calculations,[7–9] and Monte Carlo simulations.[10] In contrast to most of the theoretical work, we present a theory that is able to treat the disorder in a more realistic manner (beyond coarse graining). Our theory includes quantum fluctuations within random-phase approximation (RPA) and the disorder is treated within coherent potential approximation (CPA). It should be stressed that in our approach the spin impurities are treated quantum mechanically.

We start with the following minimal Hamiltonian:

$$H=\sum_{ij,\sigma} t_{ij}c^{\dagger}_{i\sigma}c_{j\sigma}+\sum_i J_i\vec{S}_i\vec{s}_i. \tag{1}$$

The first term stands for the tight-binding part of the itinerant free electron gas $t_{ij}=t$ if i and j are nearest neighbor, or 0 otherwise. The second term is the exchange between localized impurities spin and itinerant electron gas, J_i are random variables: $J_i=J$ if site i is occupied by a Mn^{2+} ion or 0. The operator $\vec{s}_i=c^{\dagger}_{i\alpha}(1/2\vec{\sigma}_{\alpha\beta})c_{i\beta}$ is the spin operator at i of the itinerant electron gas and S_i is the spin of the magnetic impurity.

Let us define the Green's function

$$G^{+-}_{ij}(t)=-i\theta(t)\langle[S^+_i(t),S^-_j(0)]\rangle=\langle\langle S^+_i;S^-_j\rangle\rangle. \tag{2}$$

We write the equation of motion and use Tyablicov decoupling,[11] (equivalent to RPA) which is suitable for ferromagnetic systems. It consists in closing the system by approximating the higher-order Green's function $\langle\langle S^z_i s^+_i;S^-_j\rangle\rangle \approx\langle S^z_i\rangle\langle\langle s^+_i;S^-_j\rangle\rangle$. In this approximation, we obtain in frequency space,

$$(\omega+J_i\langle s^z\rangle)G^{+-}_{ij}(\omega)=2\langle S^z_i\rangle\delta_{ij}+J_i\langle S^z_i\rangle\langle\langle s^+_i;S^-_j\rangle\rangle, \tag{3}$$

where $\langle s^z\rangle$ is the magnetization of the itinerant electron gas and $\langle S^z_i\rangle$ is the magnetization of a magnetic ion at site i. It is convenient to rewrite the new Green's function that appears in the right part of the equality in the following form: $\langle\langle s^+_i;S^-_j\rangle\rangle=1/L^2\Sigma_{kq}e^{iqR_i}\Gamma^{k+q,k}_j$, where $\Gamma^{k+q,k}_j=\langle\langle c^{\dagger}_{k+q,\uparrow}c_{k,\downarrow};S^-_j\rangle\rangle$. We obtain

$$\Gamma^{k+q,k}_j=f(k,q,\omega)\sum_l\frac{1}{2}J_l e^{-iqR_l}G^{+-}_{lj}, \tag{4}$$

where

$$f(k,q,\omega)=\frac{(\langle n_{k+q,\uparrow}\rangle-\langle n_{k,\downarrow}\rangle)}{\omega-(\epsilon_k-\epsilon_{k+q})+cJ\langle S^z_A\rangle}, \tag{5}$$

where $\langle n_{k,\sigma}\rangle$ is the occupation number of (k,σ) state. c is the impurity concentration, $\langle S^z_A\rangle$ is the averaged magnetization of Mn^{2+}, and ϵ_k denotes the hole's dispersion. Inserting both Eq. (4) and Eq. (5) into Eq. (3), we immediately find

$$G^{+-}_{ij}=g_i\delta_{ij}+g_i\sum_l\phi_{il}G^{+-}_{lj}, \tag{6}$$

where the T-dependent locator g_i is defined as

$$g_i(\omega)=\frac{2\langle S^z_i\rangle}{\omega+J_i\langle s^z\rangle}; \tag{7}$$

$\phi_{il}=\frac{1}{4}J_iJ_l\chi^0_{il}(\omega)$ and $\chi^0_{il}(\omega)$ is the Fourier transform of the polarized susceptibility $\chi^0(q',\omega)$

$$\chi^0(q',\omega)=\frac{1}{L}\sum_k\frac{(\langle n_{k+q',\uparrow}\rangle-\langle n_{k,\downarrow}\rangle)}{\omega-(\epsilon_k-\epsilon_{k+q'})+cJ\langle S^z_A\rangle}. \tag{8}$$

Note that Eq. (6) *still contains the disorder* through ϕ_{il} and g_i. It is also interesting to mention that the previous

equation can be interpreted as the propagator of a free particle moving on a disordered medium, g_i is the random on-site potential, and ϕ_{il} the long-range-hopping terms. Note also that ϕ_{il} is energy dependent through $\chi^0_{il}(\omega)$. To solve the problem we have to calculate in a self-consistent manner $\langle s^z\rangle$ and $\langle n_{k,\sigma}\rangle$ that appear in Eq. (6). For that purpose we have to write the equation of motion for the Green's function $K_{ij,\sigma}=\langle\langle c_{i,\sigma};c^\dagger_{j,\sigma}\rangle\rangle$. After decoupling we get

$$\left(\omega-\frac{1}{2}z_\sigma J_i\langle S^z_i\rangle\right)K_{ij,\sigma}=\delta_{ij}+\sum_l t_{il}K_{lj,\sigma}. \tag{9}$$

One can recognize the propagator of the Anderson model, with on-site random potential depending on the spin σ: $\epsilon_{i,\sigma}=\frac{1}{2}z_\sigma J_i\langle S^z_i\rangle$. Since in our model the potential is temperature dependent through $\langle S^z_i\rangle$ then, sufficiently close to T_c, we will always be in the metallic regime $k_f l_e\rangle\rangle 1$:[12] $l_e\approx 1/(J\langle S^z\rangle)^2$. This is in contrast with the standard Anderson model where the impurities are static. Equations (6) and (9) ($\sigma=\pm 1$) provide a closed system of equations that have to be solved self-consistently within CPA.

The simplest way is to start with Eq. (9). Indeed, it is straightforward to get the solution with the standard CPA since it contains only diagonal disorder. The averaged Green's function is

$$K_{k,\sigma}=\frac{1}{\omega-\epsilon(k)-\Sigma_\sigma(\omega)}, \tag{10}$$

where the self-energy is

$$\Sigma_\sigma(\omega)=V_\sigma-[\epsilon_{A,\sigma}-\Sigma_\sigma(\omega)]K^{00}_\sigma(\omega)[\epsilon_{B,\sigma}-\Sigma_\sigma(\omega)], \tag{11}$$

where $\epsilon_{A,\sigma}=\frac{1}{2}z_\sigma J\langle S^z_A\rangle$, $\epsilon_{B,\sigma}=0$, and V_σ is the average value $V_\sigma=\frac{1}{2}z_\sigma Jc\langle S^z_A\rangle$ and $K^{00}_\sigma=1/L\Sigma_q 1/[\omega-\epsilon(q)-\Sigma_\sigma(\omega)]$.

The self-energy $\Sigma_\sigma(\omega)$ can be reexpressed as

$$\Sigma_\sigma(\omega)=V_\sigma[1+(1-c)\tfrac{1}{2}z_\sigma J\langle S^z_A\rangle K^{00}(\omega)]. \tag{12}$$

We see that when $T\to T_c$, $\Sigma_\sigma(\omega)\to\Sigma^{VCA}_\sigma(\omega)=V_\sigma$. Thus, in the framework of our decoupling scheme, close enough to T_c, the CPA for Eq. (10) reduces to the VCA.

The final step of the calculation consists in solving Eq. (6). In order to provide analytical form for T_c, we use the similar approximation (VCA) for G^{+-}_{ij} as done above for $K_{ij,\sigma}$. We expect this approximation to be reasonable in the limit of both low-impurity concentration and low density of itinerant carriers. To get the averaged Green's function, we use the well-known Blackman-Esterling-Beck formalism.[13,14] By contrast with standard CPA, this approach is suitable for nondiagonal disorder problems. It is based on a 2×2 matrix Green's-function formalism for binary alloys using locator expansion. Within VCA approximation, one gets for the averaged Green's function of an atom of type A

$$G^{+-}_A(k,\omega)=\frac{c}{g_A^{-1}-c\,\alpha(k,\omega)}, \tag{13}$$

where $\alpha(k,\omega)=\frac{1}{4}J^2\chi^0(k,\omega)$.

Note also that since the Ga atoms have no magnetic moment, it implies $G^{+-}_B(k,\omega)=g_B=0$. The Mn^{2+} ion propagator can be rewritten, $G^{+-}_A(k,\omega)=2/[E-E(q)]$, where $E=\omega/\langle S_z\rangle$. The dispersion $E(q)$ is solution of

$$E(q)=-J\frac{\langle s_z\rangle}{\langle S_z\rangle}+\frac{1}{2}J^2\chi^0(q,E(q)\langle S_z\rangle) \tag{14}$$

According to Ref. 15 the magnetization can be expressed in the following form:

$$\langle S^z_A\rangle=\frac{(s-\phi)(1-\phi)^{2S+1}+(S+1+\phi)\phi^{2S+1}}{(1+\phi)^{2S+1}-\phi^{2S+1}}, \tag{15}$$

where $\phi=1/L\Sigma_{\mathbf{q}}1/[e^{\beta\omega(q)}-1]$.

When $T\to T_c$, $\phi=(k_BT_c/c\langle S^z_A\rangle)(1/L)\Sigma_{\mathbf{q}}1/E(\mathbf{q})$. This implies for T_c the standard RPA form

$$T_c=\frac{1}{3}c\frac{S(S+1)}{\frac{1}{N}\sum_q\frac{1}{E(q)}}. \tag{16}$$

This expression is similar to the one obtained in the clean limit for the Kondo lattice model.[16,17] In the vicinity of T_c the dispersion $E(q)$ is

$$E(q)=\frac{1}{8\pi^2}\frac{J^2}{t}\frac{1}{2}\left[k_f-\frac{1}{q}\left(k_f^2-\frac{q^2}{4}\right)\ln\left(\frac{q+2k_f}{q-2k_f}\right)\right]. \tag{17}$$

Note that, below T_c, the Eq. (14) should be solved numerically in order to get $E(q)$ as a function of the temperature. This is required to calculate $\langle S_z\rangle$ and $\langle s_z\rangle$ as function of T. According to Eq. (16), T_c is given by

$$T_c=\frac{S(S+1)}{24\pi^2}\frac{J^2c}{t}\left(\frac{1}{N}\sum_q\frac{1}{C(q,k_f)}\right)^{-1}, \tag{18}$$

where we define $C(q,k_f)=\frac{1}{2}[k_f-1/q(k_f^2-q^2/4)\ln[(q+2k_f)/(q-2k_f)]]$. This implies that T_c is proportional to J^2 and to the effective mass $(1/t)$. The dependence on the hole concentration is only contained in $C(q,k_f)$. We define the hole concentration as $n_h=\gamma c$, where $\gamma\leqslant 1$. This is the simplest way to take into account the presence of As antisites.[18] In Fig. 1 we show the variation of T_c as a function of γ. We observe that in the low-hole-concentration regime, T_c agrees very well with the mean-field result (this is more clear in the inset log-log plot). In the mean-field regime the magnon excitation spectrum is dispersionless: $E^{MF}(q)=\lim_{q\to\infty}E(q)=(1/8\pi^2)(J^2/t)k_f$ where $k_f=(3\pi^2\gamma c)^{1/3}$. In this limit

$$T_c=\frac{1}{24}\left(\frac{3}{\pi^4}\right)^{1/3}S(S+1)\frac{J^2}{t}\gamma^{1/3}c^{4/3}. \tag{19}$$

When increasing γ, T_c *strongly* deviates from the mean-field results and shows a *broad* maximum. Such a maximum was also observed in Ref. 6. By further increase in γ, the Curie temperature starts to decrease.[19] As we observe it from Fig. 1, for very large γ, T_c agrees very well with the case where the magnon spectrum is approximated by $E(q)=E^{stiff}(q)=Dq^2$ where the stiffness D is given by $D=(1/48\pi^2)(J^2/t)(1/kf)$, this regime is denoted "stiffness" regime. In this regime we find

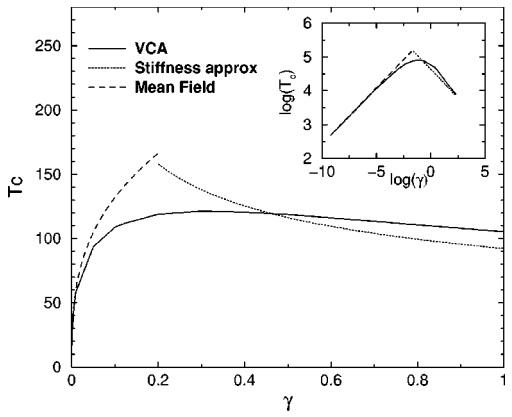

FIG. 1. T_c as a function of γ for $c=0.05$ and $J^2/t=10.5$ eV. The continuous line represent the VCA calculation, the dashed line corresponds to T_c within the mean-field approximation, and the dotted line is obtained by approximating the dispersion by $E(q)=Dq^2$, D is the spin stiffness. The inset shows $\ln(T_c)$ versus $\ln(\gamma)$.

$$T_c=\frac{1}{144}\frac{1}{(18\pi^4)^{1/3}}S(S+1)\frac{J^2}{t}\gamma^{-1/3}c^{2/3}. \tag{20}$$

The existence of a maximum can be understood in the following way: Like in the Ruderman-Kittel-Kasuya-Yosida situation,[20] the exchange oscillates with typically length scale $l_{osc}\propto 1/k_f$. Thus, it is expected that when the length scale gets sufficiently large (larger than the average distance between impurities) some Mn-Mn bonds are coupled antiferromagnetically. The induced frustration leads to the immediate consequence a decrease in T_c. In Fig. 2 we illustrate the previous discussion by showing the dispersion as a function of k/k_c where k_c is chosen in order to conserve the volume of the Brillouin zone $[v=(2\pi)^3]$. The results are shown for the three different regions: ''mean-field,'' ''intermediate,'' and ''stiffness'' regime. We observe that in all cases the dispersion goes to 0 (when $q\to 0$), as expected, when the Goldstone theorem is fulfilled.

In Fig. 3, we show the region for which T_c reaches its maximum as a function of $c[T_c^{max}(c)]$ and the region where

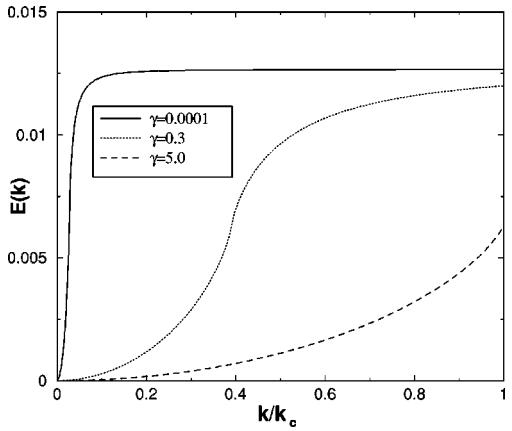

FIG. 2. Magnon dispersion for $c=0.05$ in three different regimes: ''mean-field'' (continuous line), ''intermediate'' (dotted line), and ''stiffness'' regime (dashed line). $E(q)$ is rescaled by a factor $\lambda=J^2/t(3\pi^2\gamma)^{1/3}$

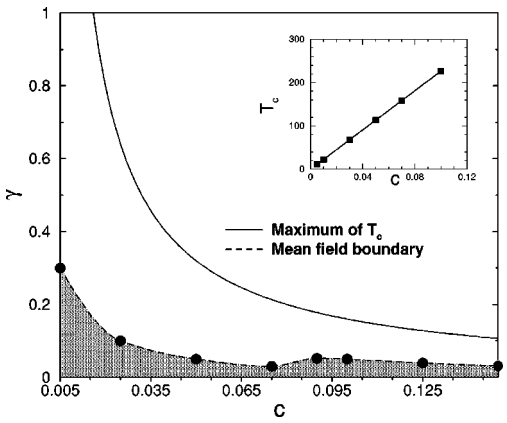

FIG. 3. The dashed area represents region where mean-field result for T_c is valid, the symbols are calculated points and the dashed line a fit. The continuous curve represents values of (γ,c) for which T_c is maximum (T_c^{max}). In the inset we have plotted T_c^{max} as a function of c assuming $J^2/t=10.5$ eV.

MF formula provides a good approximation for T_c, it corresponds to $|(T_c-T_c^{MF})/T_c|\leqslant 0.1$. First we see that the region of validity of the mean-field (MF) result (dashed area) corresponds to a very narrow region typically $\gamma\leqslant 0.05$. A good approximated value of the γ, for which T_c is maximum, can be obtained by taking the intersection point between the MF and ''stiffness'' values. This leads to $\gamma_{max}c=n_{max}=0.016$.

So far, we did not have to specify the values of the parameters t and J for our discussion. In order to check the validity of our theory we compare our results with available experimental data. GaAs is known to have a fcc structure with a lattice constant $a_0\approx 5.6$ Å. For simplification in our calculation we have assumed a simple cubic structure thus the lattice constant which has to be taken in our calculation, is $a_1=a_0/4^{1/3}$ in order to conserve the volume for the unit cell. Also assuming an effective mass for the holes $m=0.5m_e$ one gets $t=0.63$ eV. The remaining free parameter J will be chosen in order to fit the experimental data of Ref. 2. For that purpose we calculate γ for each sample according to the measured experimental values of the hole concentration given in Fig. 2 of Ref. 2. The results are depicted in Fig. 4. As can be seen we find a very good agreement with the experimental data if $J^2/t=10.5$ eV, this implies $|J|=2.58$ eV.[22] Note that the deviations observed at low c are due to the uncertainty on the hole-concentration value (see the huge error bars in Fig. 2 of Ref. 2). From the experimental measurements, there is no clear consensus concerning the correct value of this parameter. Indeed, recent core-level photoemission has provided $J=-1.2\pm 0.2$ eV.[23] Whilst, from Magnetotransport measurements a value of $|J|=2.4\pm 0.9$ eV was suggested.[2,21] And within first-principle calculations Sanvito *et al.*[7] have found $J\approx -4.65\pm 0.25$ eV. In order to proceed to a better estimation of the parameters J, one should compare theoretical calculations with other data, for instance, transport measurements data.[24] However, it is interesting to note that the band splitting at $T=0$ K ($\Delta=JcS$) obtained within our calculations agrees with the experimental value reasonably well.[25] In the inset of Fig. 3, assuming $J^2/t=10.5$ eV we show T_c as function of c taking

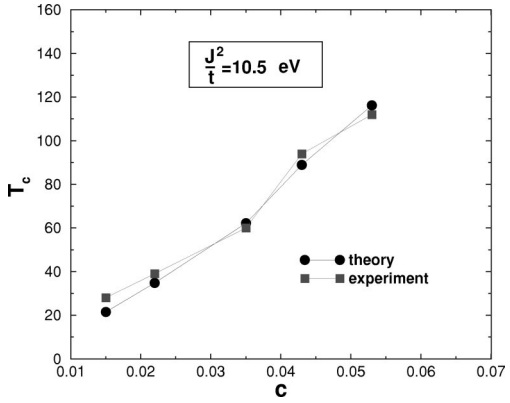

FIG. 4. T_c in Kelvin as a function of c. The filled squares corresponds to experimental values taken from Ref. 2. The filled circles represent the calculated values, the γ's where also taken from the same reference.

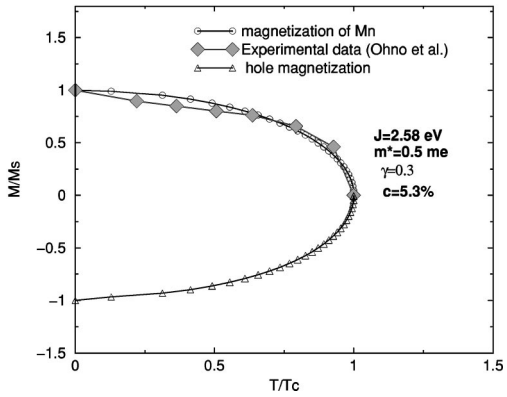

FIG. 5. Normalized magnetization as a function of T/T_c. The experimental data are taken from Ref. 2.

γ on the line of "maximum of T_c." For instance if $c \approx 0.1$ and $\gamma \approx 0.2$ a T_c of order 230 K can be reached.

Let us proceed further on and compare the calculated magnetization with the measured one. In the experimental data the concentration of Mn^{2+} is $c = 0.053\%$ and the parameter γ is estimated to be 0.3 (see Ref. 21) In Fig. 5, we show the magnetization as function of the T where $m^* = 0.5 m_e$ and $J = 2.58$ eV. We observe that for sufficiently high temperature $T \leqslant 0.5 T_c$ there is a very good agreement with the measured Mn^{2+} magnetization. On decreasing T, some deviation appears, which suggests that the VCA treatment is not good enough in this region, which was expected.

To conclude, we have presented a general theory for carrier-induced ferromagnetism in DMS. Our approach allows one to treat the disorder beyond simple coarse graining within full CPA treatment. It goes beyond mean field and includes quantum fluctuations in the RPA approximation. We have shown that, within our decoupling scheme and sufficiently close to T_c, the CPA for the itinerant gas reduces to VCA, which allowed us to provide analytical results for T_c in the low impurity concentration and hole-density regime. We have also discussed its dependence on the hole concentration. We have also shown that the mean-field approximation is only valid for very low carrier concentration. Additionally, for illustration of our theory a comparison with available experimental data on $Ga_{1-c}Mn_cAs$ was made. We find a very good agreement with the experimental results assuming a single band for itinerant carriers and a large exchange constant $J = -2.58$ eV. Finally, this work provides a good starting point for higher decoupling scheme.

Note added. After this work was completed we became aware of Yang *et al.* comment.[17] By analogy with the Kondo lattice model[16] (no disorder) they proposed a similar expression for T_c as the one derived in Eq. (16).

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