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2006 J. Phys.: Condens. Matter 18 1441

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# Ground-state properties and the molecular theory of the Curie temperature in the coherent potential approximation of diluted magnetic semiconductors

Sze-Shiang Feng<sup>1</sup> and Mogus Mochena

Physics Department, Florida A & M University, Tallahassee, FL 32307, USA

E-mail: [shixiang.feng@famu.edu](mailto:shixiang.feng@famu.edu)

Received 31 October 2005

Published 13 January 2006

Online at [stacks.iop.org/JPhysCM/18/1441](http://stacks.iop.org/JPhysCM/18/1441)

## Abstract

Using a spin-1/2 description of valence holes and Kondo coupling between local spins and carriers, GaAs-based III–V diluted magnetic semiconductors (DMS) are studied in the coherent potential approximation (CPA). Our calculated relation between ground-state energy and impurity magnetization shows that ferromagnetism is always favourable at low temperatures. For very weak Kondo coupling, the density of states (DOS) of the host semiconductor is not modified much. Impurity bands can be generated at the bottom of the host band only when Kondo coupling is strong enough. Using Weiss molecular theory, we predict a linear relation of Curie temperature with respect to Kondo coupling and doping concentration  $x$  if the hole density is proportional to  $x$ .

(Some figures in this article are in colour only in the electronic version)

The ferromagnetism of III–V-type diluted magnetic semiconductors (DMS) is not well understood. To explain ferromagnetism in DMS, various models and approaches have been proposed [1–6]. Though the models differ from each other in detail, they all agree that the coupling between the carriers and local spins is of fundamental importance. An issue of debate, however, is how the exchange between localized spins is induced by the carriers. One model for this induced exchange is the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction [1, 2]. Another version which results in conclusions equivalent to RKKY is the Zener model [4] which uses the fact that the valence holes are on p-orbitals. A third model is the double-exchange (DE) mechanism [7], but this model is inconsistent with the charge-transfer properties [4]. Though RKKY can give a Curie temperature in agreement with experiment, some argue that the RKKY model breaks down here [5, 8] because the local coupling between the carrier and the impurity spin is much larger than the Fermi energy and cannot be treated perturbatively.

<sup>1</sup> Author to whom any correspondence should be addressed.

In dealing with the effect of the localized spins, a key issue is whether or not randomness should be taken into consideration. The above models are all mean field approximations (MFA) which assume homogeneity and neglect randomness. But DMSs are disordered systems with positional disorder of Mn impurities. As concluded in [2, 9], disorder has a substantial influence upon the magnetic susceptibility of carriers. Hence, any first principle consideration should take into account the randomness of the impurities.

A classic method of dealing with randomness is the coherent potential approximation (CPA) [10] which has been applied to DMS [11–13]. Based on the formalism of [14] and assuming a very large local spin  $S$  while keeping the product  $IS$  constant (where  $I$  is the Kondo-like interaction), [11] obtained the density of states and the relation between Curie temperature and the doping concentration. Using the averaged carrier Green's function, Bouzerar *et al* [13] arrived at the conclusion that the local coupling between the carrier and the impurity spins must be intermediate in order to acquire ferromagnetism. In this paper we use the formalism of the CPA in [15] to study the ground-state properties of III–V DMS. In contrast to [12], we keep  $S = 5/2$  and treat the impurity spins fully quantum mechanically. Though it is mostly accepted that the effective spin of valence holes is  $3/2$  [16–20], we describe here the holes as spin- $1/2$  fermions. It is usually believed that such a description can still catch the essential physics. Because of spin–orbit interaction, the p-orbitals are split (with split-off  $\simeq 0.34$  eV) into a spin- $3/2$  multiplet and a spin- $1/2$  multiplet [21]. Using a spherical approximation, the kinetic energy of the Luttinger–Kohn Hamiltonian [21] for the spin- $3/2$  multiplet takes the form  $\sum_{\mu} (\hbar^2 \mathbf{k}^2 / 2m_{\mu}) c_{\mathbf{k}\mu}^{\dagger} c_{\mathbf{k}\mu}$  near the valence top after diagonalization, where  $m_{\mu} = m_h \simeq 0.5m$  for  $\mu = \pm 3/2$  and  $m_{\mu} = m_{\ell} \simeq 0.07m$  for  $\mu = \pm 1/2$  ( $m$  is the effective mass of a free hole). The interaction between the spin of holes and local  $5/2$ -spins now takes a  $\mathbf{k}$ -dependent form  $\sum_{\mathbf{k}, \mathbf{k}'} \mathbf{S} \cdot c_{\mathbf{k}}^{\dagger} \mathbf{J}(\mathbf{k}, \mathbf{k}') c_{\mathbf{k}'} \exp[-i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}]$  [18]. Since the DOS for parabolic band is  $g(\varepsilon) = (1/2\pi^2 \hbar^3) (2m)^{3/2} \sqrt{\varepsilon}$ , we have the ratio of DOS  $g_h(\varepsilon)/g_{\ell}(\varepsilon) \simeq 19$  for heavy holes and light holes, i.e. about 95% of valence holes are heavy holes. Therefore, it is a valid approximation to consider only heavy holes. What is more, since the hole density is very small, the Fermi wavevector is supposed to be very small and the it is thus a reasonable approximation to consider those  $\mathbf{k}$  values in the interaction term. And this leads to the usual assumption that the carriers are shallow holes and the coupling of the shallow holes to the  $\text{Mn}^{2+}$  can be described by local Kondo interaction between  $5/2$ -spins and  $1/2$ -spins.

We study here one of the most commonly studied DMS,  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ , where the doping concentration  $x$  varies from 0.015 to 0.075 in the region of interest for ferromagnetism [1]. In  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ , ferromagnetism was first realized at a Curie temperature of 110 K [2]. The carriers are holes originating from randomly distributed Mn. The system is highly compensated [22–24] with a hole density  $p$  only around 10% of the Mn density  $x$ . There are different kinds of randomness, e.g. substitutional randomness, interstitial randomness, antisite randomness and directional randomness of impurity spin. It is commonly agreed now that interstitial Mn atoms and antisite As only reduce the hole densities and do not affect conduction of holes significantly. Therefore we consider only two kinds of randomness, i.e. the random substitution of the Mn atoms and the random direction of the impurity spins. The model Hamiltonian in our description is

$$H = \sum_{\mathbf{i}, \mathbf{j}, \sigma} t_{\mathbf{ij}} c_{\mathbf{i}\sigma}^{\dagger} c_{\mathbf{j}\sigma} + \sum_{\mathbf{i}} u_{\mathbf{i}} \quad (1)$$

where  $u_{\mathbf{i}}$  depends on whether  $\mathbf{i}$  is a Ga or Mn site. For a Ga site  $u_{\mathbf{i}} = u_{\mathbf{i}}^{\text{G}} = E_{\text{G}} \sum_{\sigma} c_{\mathbf{i}\sigma}^{\dagger} c_{\mathbf{i}\sigma}$ , and for a Mn site  $u_{\mathbf{i}} = u_{\mathbf{i}}^{\text{M}} = E_{\text{M}} \sum_{\sigma} c_{\mathbf{i}\sigma}^{\dagger} c_{\mathbf{i}\sigma} + J_{\text{K}} \mathbf{S}_{\mathbf{i}} \cdot \mathbf{s}_{\mathbf{i}}$ .  $\mathbf{S}_{\mathbf{i}}$  is the local spin of Mn at site  $\mathbf{i}$ ,  $\mathbf{s} = (1/2) c_{\sigma}^{\dagger} \boldsymbol{\tau}_{\sigma\sigma'} c_{\sigma'}$  is the spin of a hole where  $c_{\sigma}^{\dagger} (c_{\sigma})$  is the creation(annihilation) operator for holes, spin indices  $\sigma, \sigma' = \uparrow, \downarrow$  and  $\boldsymbol{\tau} = (\tau_1, \tau_2, \tau_3)$  are the three usual Pauli matrices.  $E_{\text{M}}$

and  $E_G$  are the on-site energies for Ga and Mn and are assumed constant. The hopping energy  $t_{ij} = t$  if  $\mathbf{i}, \mathbf{j}$  are nearest neighbours and zero otherwise and  $J_K > 0$  is the local Kondo coupling. The details of the lattice structure are not crucial in the following discussion. According to the general scheme of CPA, the virtual unperturbed Hamiltonian is

$$\mathcal{H}(\varepsilon) = \sum_{\sigma, \mathbf{k}} (t_{\mathbf{k}} + \Sigma_{\sigma}(\varepsilon)) c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}\sigma} \quad (2)$$

where  $\varepsilon$  is the Fourier frequency variable,  $\Sigma_{\sigma}(\varepsilon)$  is the CPA self-energy to be determined self-consistently and  $t_{\mathbf{k}}$  is the Fourier transformation of  $t_{ij}$ . Then the relative perturbation  $V$  is given by

$$V = H - \mathcal{H}(\varepsilon) = \sum_{\mathbf{i}} v_{\mathbf{i}} \quad (3)$$

where  $v_{\mathbf{i}} = v_{\mathbf{i}}^G = \sum_{\sigma} (E_G - \Sigma_{\sigma}) c_{\mathbf{i}\sigma}^{\dagger} c_{\mathbf{i}\sigma}$  for Ga and  $v_{\mathbf{i}} = v_{\mathbf{i}}^M = \sum_{\sigma} (E_M - \Sigma_{\sigma}) c_{\mathbf{i}\sigma}^{\dagger} c_{\mathbf{i}\sigma} + J_K \mathbf{S}_{\mathbf{i}} \cdot \mathbf{s}_{\mathbf{i}}$  for Mn. The reference Green's function is  $\langle \mathbf{i}\sigma | \mathcal{R}(\varepsilon) | \mathbf{j}\sigma' \rangle = \langle 0 | c_{\mathbf{i}\sigma}(\varepsilon - \mathcal{H})^{-1} c_{\mathbf{j}\sigma'}^{\dagger} | 0 \rangle$  where  $|0\rangle$  is the vacuum state of the  $c$  operators, and the associated  $t$  matrices are  $t_{\mathbf{i}}^G = v_{\mathbf{i}}^G / (1 - \mathcal{R}v_{\mathbf{i}}^G)$ ,  $t_{\mathbf{i}}^M = v_{\mathbf{i}}^M / (1 - \mathcal{R}v_{\mathbf{i}}^M)$ . So the CPA equation and DOS are given by

$$(1-x)t_{\mathbf{i}}^G + x \langle t_{\mathbf{i}}^M \rangle_{\text{spin}} = 0 \quad (4)$$

and

$$g_{\sigma}(\varepsilon) = -\frac{1}{\pi} \text{Im} F_{\sigma}(\varepsilon) \quad (5)$$

where  $\langle \dots \rangle_{\text{spin}}$  denotes the average over the configurations of impurity spins and  $F_{\sigma}(\varepsilon) = \langle \sigma \mathbf{i} | \mathcal{R} | \mathbf{i}\sigma \rangle = (1/N) \sum_{\mathbf{k}} [1/(\varepsilon - t_{\mathbf{k}} - \Sigma_{\sigma})]$ , where  $N$  is the number of lattice sites. As usual, the spin-resolved bare DOS (for undoped GaAs) can be approximated by the semicircle DOS

$$g_0(\varepsilon) = \frac{2}{\pi \Delta} \sqrt{1 - \left(\frac{\varepsilon}{\Delta}\right)^2} \quad (6)$$

where  $\Delta$  denotes the half-band width. At zero temperature, the carrier density for spin  $\sigma$  can be expressed as  $n_{\sigma} = \int_{-\infty}^{\varepsilon_F} g_{\sigma}(\varepsilon) d\varepsilon$ , where  $\varepsilon_F$  is the Fermi energy, and the total carrier density  $n = n_{\uparrow} + n_{\downarrow}$ . The total electronic ground-state energy per site is  $\varepsilon_g = \int_{-\infty}^{\varepsilon_F} \varepsilon [g_{\uparrow}(\varepsilon) + g_{\downarrow}(\varepsilon)] d\varepsilon$ . Defining  $V_{\uparrow} = E_M - \Sigma_{\uparrow} + (J_K/2)S^z$ ,  $V_{\downarrow} = E_M - \Sigma_{\downarrow} - (J_K/2)S^z$ ,  $U_{\sigma} = V_{\sigma} - J_K/2$ ,  $W_{\uparrow} = (1/4)J_K^2 S^- S^+$ ,  $W_{\downarrow} = (1/4)J_K^2 S^+ S^-$ , the CPA equations can be written as

$$(1-x) \frac{E_G - \Sigma_{\uparrow}}{1 - F_{\uparrow}(E_G - \Sigma_{\uparrow})} + x \left\langle [V_{\uparrow}(1 - F_{\downarrow}U_{\downarrow}) + F_{\downarrow}W_{\uparrow}] \times \frac{1}{(1 - F_{\uparrow}V_{\uparrow})(1 - F_{\downarrow}U_{\downarrow}) - F_{\uparrow}F_{\downarrow}W_{\uparrow}} \right\rangle_{\text{spin}} = 0 \quad (7)$$

$$(1-x) \frac{E_G - \Sigma_{\downarrow}}{1 - F_{\downarrow}(E_G - \Sigma_{\downarrow})} + x \left\langle [V_{\downarrow}(1 - F_{\uparrow}U_{\uparrow}) + F_{\uparrow}W_{\downarrow}] \times \frac{1}{(1 - F_{\downarrow}V_{\downarrow})(1 - F_{\uparrow}U_{\uparrow}) - F_{\uparrow}F_{\downarrow}W_{\downarrow}} \right\rangle_{\text{spin}} = 0. \quad (8)$$

These relations are given in [15] in another context. For any  $f(S^z)$ , the spin average is given by  $\langle f(S^z) \rangle_{\text{spin}} = \sum_{S^z=-S}^S e^{\lambda S^z} f(S^z) / \sum_{S^z=-S}^S e^{\lambda S^z}$  where  $\lambda$  is determined by the condition  $\langle S^z \rangle_{\text{spin}} = m$ ,  $m$  being the given magnetization of the impurity spins. In our single-particle CPA, the Callen–Shtrikman relation [25] that tells there is a one-to-one correspondence between  $m$  and  $\langle (S^z)^n \rangle_{\text{spin}}$  for  $n > 1$  applies. Corresponding to bare DOS, we have  $F^{(0)}(\varepsilon) = (2/\Delta^2)(\varepsilon - \sqrt{\varepsilon^2 - \Delta^2})$  so  $F_{\sigma}(\varepsilon) = F^{(0)}(\varepsilon - \Sigma_{\sigma})$ . Solving for  $\Sigma_{\sigma}$ , we have

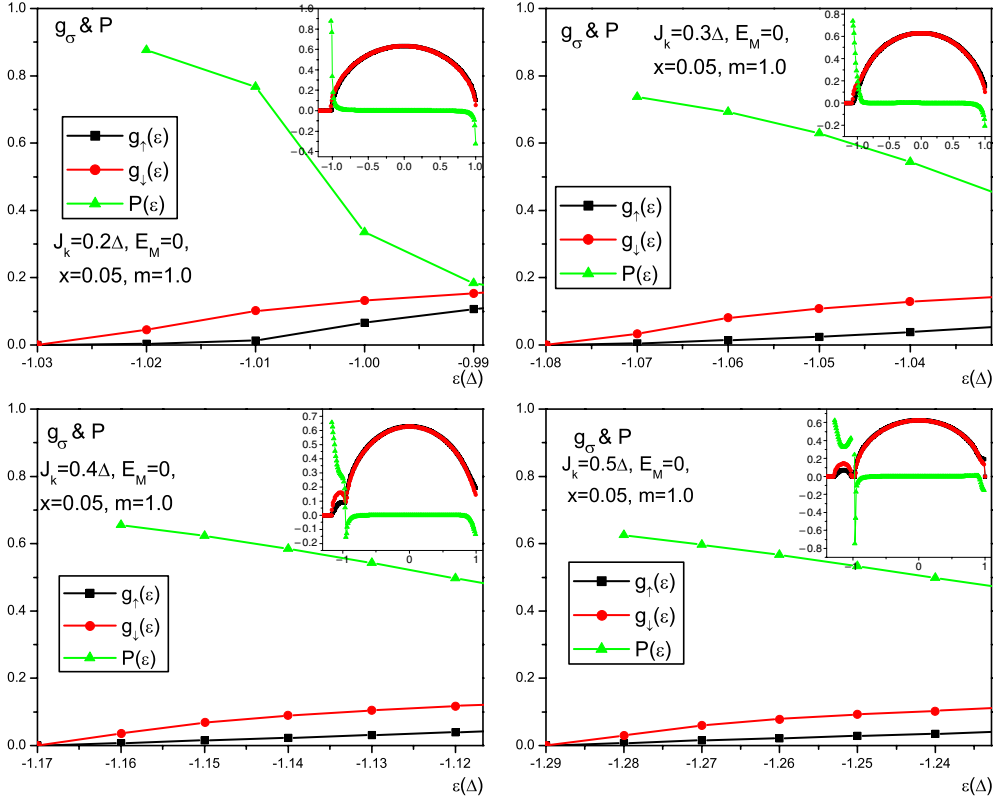
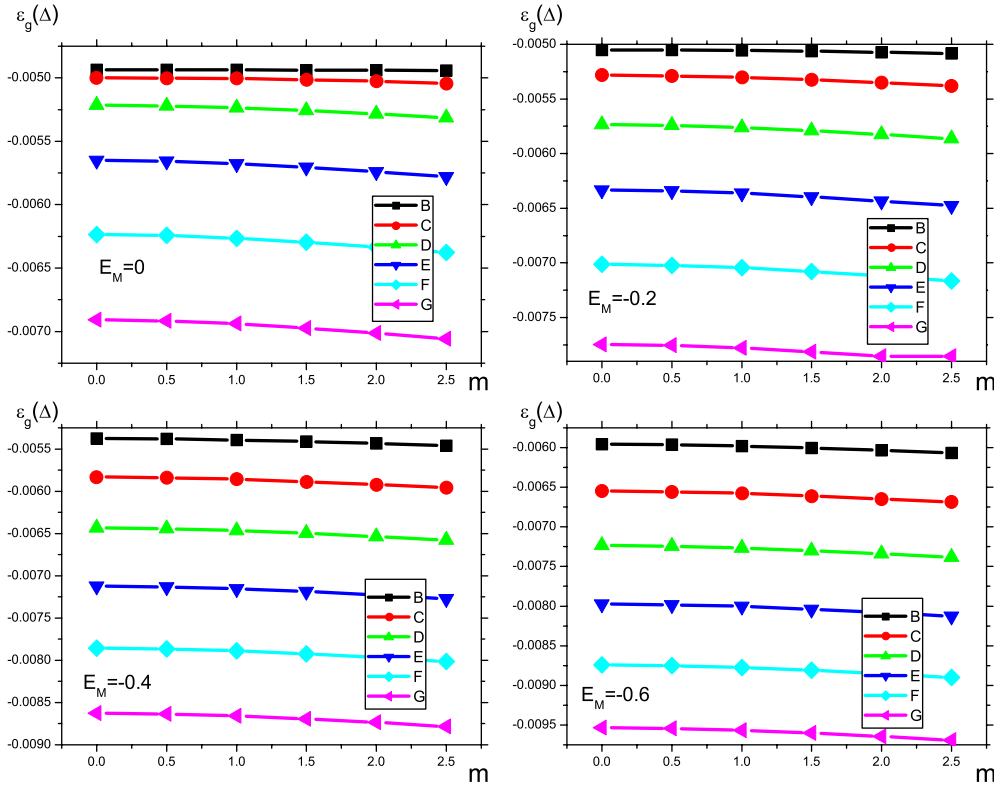


Figure 1. DOS and polarization for a number of model parameters.

$\Sigma_\sigma = \varepsilon - (\Delta^2/4)F_\sigma - 1/F_\sigma$ . Therefore CPA equations can be turned into equations for functions  $F_\sigma$  for a given  $\varepsilon$ . Once  $F_\sigma(\varepsilon)$  are known, DOS  $g_\sigma(\varepsilon)$  and quantities like Fermi energy can be calculated.

To solve CPA equations (7) and (8), we choose  $x = 0.05$ ,  $p = 0.1x$  and set  $E_G = 0$  since we can shift the chemical potential without loss of physics. Energies are normalized so that  $\Delta = 1$ . The solution of  $F_\sigma(\varepsilon)$  is generated by iteration starting from  $F^{(0)}$ . First, we use the small value of  $x$  to generate  $\Sigma_\sigma$  and then use the resulting  $\Sigma_\sigma$  to generate  $F_\sigma$ . We calculated the DOS and the spin polarization  $P(\varepsilon)$  [26] for model parameters  $J_K = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6$ ,  $E_M = 0, -0.2, -0.4, -0.6$  with  $m = 0.05, 1.0, 1.5, 2.0, 2.5$ . As a check for our numerical results, the sum rule  $\int_{-\infty}^{\infty} g_\sigma(z) dz = 1$  is preserved and the relation  $n_\uparrow + n_\downarrow = p$  is also preserved where  $n_\sigma$  is calculated from spin-resolved DOS respectively. In figure 1 we show some of our results. The curves presented in the main panels of figure 1 are from the bottom of the band to Fermi energies at zero temperature for clarity since this portion is important for low-temperature physics. The insets outline the full behaviour. It is seen that for very weak Kondo couplings such as  $J_K = 0.2\Delta, 0.3\Delta$  there are no impurity bands and the DOS is not substantially different from the bare DOS in shape. Only when Kondo coupling becomes strong enough can there be impurity bands and corresponding peaks. In contrast to the conclusion of classical spin approximation [12], the spin polarizations are not constant up to Fermi energies. Figure 1 shows that for  $m > 0$ , there are always more spin-down carriers than spin-up carriers, in compliance with the fact that the local p-d coupling is antiferromagnetic [4].



**Figure 2.** Relation of  $\varepsilon_g$  versus  $m$ : B,  $J_K = 0.1$ ; C,  $J_K = 0.2$ ; D,  $J_K = 0.3$ ; E,  $J_K = 0.4$ ; F,  $J_K = 0.5$ ; G,  $J_K = 0.6$ .

Another substantial difference between our calculated DOS and that in [12] is that there is no impurity peak above the top of the band in our calculation. So the  $\varepsilon \leftrightarrow -\varepsilon$  symmetry of the bare DOS is broken by the impurities.

Figure 2 shows the relation between ground-state energy per site and the impurity magnetization  $m$ . In the actual calculation of ground-state energy, an important issue is the determination of the Fermi energy which is fixed by the integration of an interpolation function of the DOS. If the interval  $\delta\varepsilon$  is not small enough, the integral of the interpolated DOS may vary significantly with the choice of  $\delta\varepsilon$ . To make it stable enough, we choose  $\delta\varepsilon = 5 \times 10^{-4}$  to interpolate the DOS linearly. Our results show that for all the chosen values of model parameters, the ground-state energy per site always decreases, though very slowly, with increase in impurity magnetization. Therefore CPA predicts that at very low temperatures ferromagnetism is always energetically favourable for all the model parameters considered.

In figure 3, we show the dependence of Curie temperature on the model parameters and the doping concentration. Weiss molecular field theory is employed as follows to calculate the Curie temperature. Given  $m$ , one can calculate DOS and then  $\langle s_z \rangle$ . So one can establish the relation  $\langle s_z \rangle = \langle s_z \rangle(m)$ . On the other hand, given  $\langle s_z \rangle$ , each impurity spin feels an effective field  $J_K \langle s_z \rangle$  and thus we have  $m = SB_S(\beta h)$  with  $h = -J_K \langle s_z \rangle(m)$ ,  $\beta = 1/k_B T$  and  $B_S(x)$  the conventional Brillouin function. For very small  $m$ , we have  $\langle s_z \rangle \simeq -Am$  with  $A > 0$  and we have  $\beta h \simeq \beta J_K A m$ . So  $B_S(\beta h) \simeq (S + 1)\beta h/3$  and thus the Curie temperature can be estimated by  $k_B T_C \simeq J_K S(S + 1)A/3$ . For small  $m$ , letting

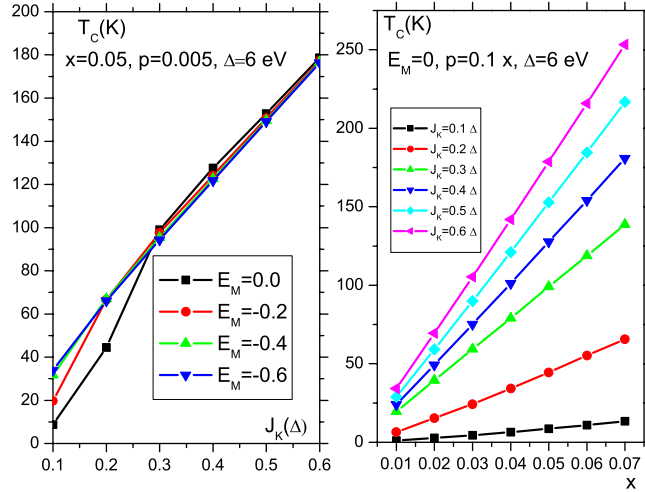


Figure 3. Theoretical estimation of Curie temperature using Weiss molecular theory.

$F_{\uparrow}(z) = F(z) + \psi(z)m$ ,  $F_{\downarrow}(z) = F(z) - \psi(z)m$  where  $F(z)$  is the paramagnetic solution, we have

$$A(J_K, E_M, x, \beta) \simeq \frac{1}{\pi} \int_{-\infty}^{\varepsilon_F} d\varepsilon \text{Im} \psi(\varepsilon) \quad (9)$$

where we have ignored the  $\beta$  dependence. As our numerical results (not shown here) indicate, the chemical potential is very close to the zero-temperature Fermi energy in a wide range of temperatures ( $\beta > 100$ ), showing that the Fermi function can be approximated by the zero-temperature step function. Since the width of the full valence band of GaAs is about 10–12 eV (the width of  $\Gamma_8$  band  $\sim 4$  eV) [27], here we take  $\Delta = 6$  eV. The left panel in figure 3 shows the relation of  $T_C$  versus  $J_K$  for different values of  $E_M$ . The curves exhibit a linear relation. For  $J_K > 0.3$ ,  $T_C$  is almost independent of  $E_M$ . The right panel shows the dependence of Curie temperature on the doping concentration for  $E_M = 0$  and various values of  $J_K$ . Here we still assume that the hole density  $p = 0.1x$ . Again, the relation indicated is linear in the range  $0.01 < x < 0.07$ . Figure 3 suggests that to reach the observed  $T_C = 110$  K, the value of  $J_K$  needs to be  $0.3\Delta - 0.4\Delta$ . For  $\Delta = 6$  eV, this value is much larger than the value 1 eV calculated in [17]. But it is still within the possible range proposed in [28]. Like in the dynamical mean field study [29] which also used a semicircle DOS and bandwidth  $\sim 10$  eV, the crucial issue in our current CPA study is the behaviour of the Curie temperature versus the model parameters. The resulting number of Curie temperatures can be scaled up or down depending on the choice of the bandwidth.

To conclude, we summarize our results here. Using CPA and treating the impurity spins fully quantum mechanically, we have calculated the ground-state energies of GaAs-based III–V DMS for a wide range of model parameters. The results show that ferromagnetism is always preferable at low temperatures. Unlike the classical treatment of the impurity spins, our approach predicts that an impurity band can arise only at bottom of the band of the host system, showing asymmetry caused by doping. With the help of the Weiss molecular theory of ferromagnetism, we obtained a linear relation of Curie temperature with respect to Kondo coupling and doping concentration. Our results agree with those in [30]. As is known from experiments [1, 22],  $T_C$  increases almost linearly with  $x$  for  $x < 0.053$  and starts to drop when  $x$  becomes larger. The contradiction might be reconciled by the dependence on  $x$  of the

exchange integral (usually denoted as  $N_0\beta$ ). It is found in [28] that the absolute value  $N_0\beta$  decreases as  $x$  increases, a behaviour already well known to occur in  $\text{Co}_{1-x}\text{Mn}_x\text{S}$ . Therefore, if there is a relation like  $J_K = -ax + b$  with  $a$  and  $b > 0$ , as suggested by the data given in [28], it can be expected that the resulting Curie temperature may start to decrease at a certain level of doping concentration.

## Acknowledgments

The authors thank Professor Rajagopal for his helpful discussions. SF is also grateful to Professor Takahashi for helpful communications. This work is supported in part by the Army High Performance Computing Research Center (AHPCRC) under the auspices of the Department of the Army, Army Research Laboratory (ARL) under Cooperative Agreement number DAAD19-01-2-0014.

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