Hole-hole correlation effects on magnetic properties of Mn$_x$III$_{1-x}$V diluted magnetic semiconductors

T. Jungwirth$^{a,b,c,1}$, Byoungkak Lee$^{a,c}$, A.H. MacDonald$^{a,c}$

$^a$Department of Physics, Indiana University, Bloomington, IN 47405, USA
$^b$Institute of Physics ASCR, Cukrovarnická 10, 162 00 Prague 6, Czech Republic
$^c$Department of Physics, The University of Texas at Austin, Austin, TX 78712, USA

Abstract

The mean-field theory represents a useful starting point for studying carrier-induced ferromagnetism in Mn$_x$III$_{1-x}$V diluted magnetic semiconductors. A detail description of these systems requires to include correlations in the many-body hole system. We discuss the effects of correlations among itinerant carriers on magnetic properties of bulk Mn$_x$III$_{1-x}$V and magnetic semiconductor quantum wells. Presented results were obtained using parabolic band approximation and we also derive a many-body perturbation technique that allows to account for hole-hole correlations in realistic semiconductor valence bands.

Keywords: Diluted Magnetic Semiconductors, Ferromagnetism, Electronic correlations

1. Introduction

Recent advances [1–3] in the fabrication and control of Mn$_x$III$_{1-x}$V diluted magnetic semiconductors (DMS) have opened up a broad and relatively unexplored frontier for both basic and applied research. We have developed a mean-field theory of free-carrier induced ferromagnetism in DMS which is intended to be useful for bulk materials as well as for any spatially inhomogeneous systems. Special emphasis is placed on interaction effects in the itinerant many-body system whose inclusion is required for a detailed understanding of magnetic properties of these materials. In Section 2 we derive the mean-field theory equations. Correlation effects on magnetic properties of bulk DMS and Mn-doped semiconductor quantum wells are studied in Sections 3 and 4, respectively, using a parabolic band model. In Section 5 we present a many-body perturbation theory that accounts for hole-hole interactions in realistic semiconductor valence bands and evaluate the exchange enhancement of the ferromagnetic critical temperature for typical sample parameters.

2. Mean-field theory

Our theory is based on an envelope function description of the valence band electrons and a spin representation of their kinetic-exchange interaction [4] with $d$-electrons on the $S = 5/2$ Mn$^{++}$ ions:
\[ H = H_m + H_f + J_{pd} \sum_{i,j} \mathbf{S}_i \cdot \mathbf{s}(\mathbf{r}_i - \mathbf{R}_j), \]  

where \( i \) labels a free carrier, \( I \) labels a magnetic ion and the exchange interaction is parametrized by a coupling constant \( J_{pd} \). In Eq. (1) \( H_m \) is the Hamiltonian of the magnetic ions, \( H_f \) is the six-band Luttinger Hamiltonian for free carriers in the valence band, \( \mathbf{S} \) is the magnetic ion spin and \( \mathbf{s} \) is the electron-spin operator projected onto the \( j = \frac{3}{2} \) and \( \frac{1}{2} \) valence band manifold of the Luttinger Hamiltonian.

In the absence of external fields, the mean polarization of a magnetic ion is given by [5]

\[ \langle m \rangle_I = B_S(J_{pd}S[n_{\uparrow}(\mathbf{R}_I) - n_{\downarrow}(\mathbf{R}_I)]/2k_BT), \]  

where \( B_S(x) \) is the Brillouin function,

\[ B_S(x) \approx \frac{S + 1}{3S} x, \quad x \ll 1. \]  

The electron spin-densities \( n_{\sigma}(\mathbf{r}) \) are determined by solving the Schrödinger equation for electrons which experience an electrostatic potential, \( v_{es}(\mathbf{r}) \), a spin-dependent kinetic-exchange potential,

\[ h_{pd}(\mathbf{r}) = J_{pd} \sum_I \delta(\mathbf{r} - \mathbf{R}_I)\langle m \rangle_I, \]  

and a local-spin-density-approximation (LSDA) exchange-correlation potential, \( v_{xc}(\mathbf{r}) \), on which we comment further below. The kinetic-exchange potential is non-zero only in the ferromagnetic state and we assume that the magnetic ions are randomly distributed and dense on a scale set by the free carrier Fermi wavevector and that their density, \( c(\mathbf{r}) \), can be controlled. This allows us to take a continuum limit where

\[ h_{pd}(\mathbf{r}) = J_{pd} c(\mathbf{r}) \langle m \rangle(\mathbf{r}). \]  

### 3. Homogeneous DMS

For homogeneous systems with randomly distributed localized spins the mean-field equations can be solved analytically. The hole spin-density and the kinetic-exchange potential are related as

\[ (n_{\uparrow} - n_{\downarrow})/2 = \frac{\chi_f}{(g^* \mu_B)^2} h_{pd}, \]  

where \( \chi_f \) is the interacting hole magnetic susceptibility. The Curie-Weiss temperature, obtained from Eqs. (2) - (6), is given by

\[ k_B T_c = \frac{cS(S + 1)}{3(g^* \mu_B)^2} \frac{J_{pd}^2 \chi_f}{(g^* \mu_B)^2}. \]  

In Fig. 1 we have plotted the ferromagnetic transition temperature as a function of the total carrier density, \( n \), predicted by this expression for p-type Mn_xGa_{1-x}As with \( J_{pd} = 0.15 \text{ eV mm}^3 \) \( c = 10^{21} \text{ cm}^{-3} \) and for valence bands approximated by a single parabolic band with effective hole mass \( m^* = 0.5 m_e \). When exchange and correlation effects are neglected and \( \chi_f \) is replaced by its zero-temperature value, \( T_c \) is proportional to \( n^{1/3} \). Including the parabolic band exchange-correlation potential [6] enhances \( T_c \) by \( \approx 20\% \) for typical hole densities \( \sim 10^{20} \text{ cm}^{-3} \).

### 4. DMS quantum wells

In the parabolic band approximation, the spin-densities in Mn_xGa_{1-x}As quantum wells are determined by solving the Schrödinger equation

\[ \left[-\frac{1}{2m^*_\parallel}(\partial_x^2 + \partial_y^2) + \frac{1}{2m^*_z} \partial_z^2 + v_{es}(z)\right] \psi_n(x,y,z) = E_n \psi_n(x,y,z), \]
\[+v_{xc,\sigma}(z) = \frac{\alpha}{2} \psi_{k,\sigma}(r) = \epsilon_{k,\sigma} \psi_{k,\sigma}(r);\]
\[n_\sigma(z) = \sum_k f(\epsilon_{k,\sigma})|\psi_{k,\sigma}(z)|^2,\]
where the in-plane effective mass \(m^*_n \approx 0.11m_0\)
and the out-of-plane mass \(m^*_z \approx 0.38m_0\). The electrostatic potential varies along the growth (z) direction and includes band offset and ionized impurity contributions. The LSDA equation for the exchange-correlation potential reads
\[v_{xc,\sigma}(z) = \frac{d[n_{xc}(n_\uparrow, n_\downarrow)\rho]}{dn_\sigma} |_{n_\sigma = n_\sigma(z)} ,\]
where \(\epsilon_{xc}(n_\uparrow, n_\downarrow)\) is the exchange and correlation energy per particle of a spatially uniform system [6].

If hole correlations and subband mixing are neglected the following analytic expression can be found for \(T_c\):
\[T_c = \frac{S(S+1)}{12} \frac{J^2}{k_B} \frac{m^*_n}{\pi \hbar^2} \int dz |\varphi(z)|^4 C(z).\]
In Eq. (10), the critical temperature increases with the subband wavefunction, \(\varphi(z)\), and magnetic impurities overlap, is proportional to \(J^2\) and \(m^*_n\). Similar \(T_c\)-equation was derived [7] using the RKKY theory and its predictions are in agreement with the measured carrier-induced ferromagnetism in II-VI quantum wells [7,8]. Here we emphasize the role of hole-hole interactions and Mn doping profile on magnetic properties of biased III-V DMS quantum wells. A remarkable feature of quasi-2D ferromagnetic DMS is the possibility of tuning \(T_c\) through a wide range in situ, by the application of a gate voltage. In Fig. 2 we illustrate this for the case of a \(w = 10\) nm quantum well with magnetic ions covering only a \(d = 3\) nm portion near one edge. Even for such a narrow quantum well, the critical temperature can be varied over an order of magnitude by applying a bias voltage which draws electrons into the magnetic ion region. Hole-hole interactions substantially increase the critical temperature, as also shown in Fig. 2.

![Fig. 2. Dependence of \(T_c\) on bias voltage applied across the \(w = 10\) nm wide quantum well partially occupied by magnetic ions over a distance of 3 nm near one edge of the quantum well. Circles (squares) represent results of the full numerical self-consistent calculations without (with) the exchange-correlation potential.

5. Exchange and correlations in realistic valence bands

A semiempirical six band model is used for quantitative description of electronic states in cubic semiconductors near the top of the valence band. For systems with spin-orbit coupling the Kohn-Luttinger Hamiltonian [9] is derived in the basis of total angular momentum eigenstates and Kohn-Luttinger Hamiltonian eigenstates:
\[H_I = \sum_{n_1, n_2, k_1, k_2, q} a^{\dagger}_{n_1, k_1, k_2} a^{\dagger}_{n_2, k_2} q a_{n_1, k_1} a_{n_2, k_2} V(q),\]
where \(V(q)\) is the Fourier transform of the Coulomb interaction. Using the following notation for the transformation between creation and annihilation operators for the total angular momentum eigenstates and Kohn-Luttinger Hamiltonian eigenstates:
\[a^{\dagger}_{n, k} = \sum_{\alpha} z_{n,\alpha, k} a^{\dagger}_{\alpha, k}, a_{n, k} = \sum_{\alpha} z_{n,\alpha, k} a_{\alpha, k}\]
the exchange contribution to the hole total energy has a form
where \( n_{\alpha, k} \) is the Fermi distribution function of Kohn-Luttinger eigenstates.

In the random phase approximation (RPA), the correlation energy is related to the wavevector and frequency dependent dielectric function as [10]

\[
E_c = \frac{1}{2} \sum_{q} n_{\alpha_1, \alpha_2, k+q} \times \left| \sum_{n} z_{n, \alpha_2, k+q} \bar{\sigma}_{n, \alpha_1, k} \right|^2 V(q), \quad \text{(13)}
\]

where \( n_{\alpha, k} \) is the Fermi distribution function of Kohn-Luttinger eigenstates.

The Curie-Weiss temperature for realistic valence bands is obtained from Eq. (7) with the magnetic susceptibility given by

\[
\chi_T = \frac{d^2(E_T/\Omega)}{dh^2},
\]

where the itinerant system total energy per volume, \( E_T/\Omega \), has a kinetic (band) contribution and the exchange and correlation contributions given by Eqs. (13) and (14).

For the current experimental values of the hole density \( n = 3.5 \times 10^{20} \text{ cm}^{-3} \), Mn concentration \( c = 1 \times 10^{21} \text{ cm}^{-3} \), and kinetic exchange constant \( J_{pd} = 55 \text{ meV nm}^{-3} \), the six-band model gives mean-field critical temperature for bulk Mn\(_x\)Ga\(_{1-x}\)As, \( T_c = 86 \text{ K} \) when exchange and correlations are neglected. The exchange and correlation contributions to the total energy enhances \( T_c \) by a factor of 1.2. Compared to the experimental \( T_c = 110 \text{ K} \), the mean-field theory accurately predicts the ferromagnetic critical temperature of Mn\(_x\)Ga\(_{1-x}\)As DMS.

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