Transition temperature of ferromagnetic semiconductors: a dynamical mean field study

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We formulate a theory of doped magnetic semiconductors such as Ga\textsubscript{1−x}Mn\textsubscript{x}As which have attracted recent attention for their possible use in spintronic applications. We solve the theory in the dynamical mean field approximation to find the magnetic transition temperature $T_c$ as a function of magnetic coupling strength $J$ and carrier density $n$. We find that $T_c$ is determined by a subtle interplay between carrier density and magnetic coupling.

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Diluted magnetic semiconductors have attracted much recent attention \cite{1} for their potential use in spintronic devices. The prospect of carrying out both information processing and storage on the same chip is an exciting possibility. For applications it is desirable to find materials which are ferromagnetic at as high a temperature as possible, so the discovery \cite{2} of a ferromagnetic transition with $T_c$ as high as 110K in MBE grown Ga\textsubscript{1−x}Mn\textsubscript{x}As, has inspired a great deal of interest. In addition to its potential technological significance, the ferromagnetism of Ga\textsubscript{1−x}Mn\textsubscript{x}As is an important fundamental condensed matter problem. The cause of ferromagnetism in Ga\textsubscript{1−x}Mn\textsubscript{x}As (and similar materials, e.g. In\textsubscript{1−x}Mn\textsubscript{x}As where $T_c \approx 25 − 30K$) is controversial: the different proposed mechanisms \cite{3,4,5,6,7,8,9} do not qualitatively agree with each other. In this paper we present a new theoretical approach which allows calculation of magnetic transition temperatures (and other properties) over a much wider temperature range than had previously been possible and provides new insights into the factors controlling $T_c$.

It is well established that in III-V systems such as Ga\textsubscript{1−x}Mn\textsubscript{x}As, the Mn ions go in substitutionally at the cation (Ga) sites and contribute itinerant holes to the conduction band and act as a partially compensated. The Mn ion has a half filled d-orbitals unrelated to the doped holes, magnetic exchange between Mn spins arising from other orbitals unrelated to the doped holes, $J_{AF}$ is a direct antiferromagnetic exchange between Mn spins arising from other orbitals unrelated to the doped holes, $J$ is the local exchange coupling between the spin of the Mn and the spins of the semiconductor carriers. We normalize the $\delta$ function in the $J$ term to the volume $b^3 = 5.65 Å^3$ per GaAs unit. The large (S = 5/2) value of the Mn spin justifies treating the spins classically, so that the partition function $Z$ may be determined by finding the free energy $F(\{S_i\})$ of holes in a fixed spin configuration and then averaging over spin configurations, i.e.

$$Z = \int \{dS\} e^{-\left(\sum_{i,j} J_{AF}(R_i − R_j)S_i \cdot S_j + F(\{S_i\})\right)/k_B T}$$

The key issue is therefore the evaluation of $F(\{S_i\})$. From Eq. \cite{1} we see that $F$ is the free energy of non-interacting carriers in a spin dependent potential, which may have randomness both from the distribution of Mn positions and from spin disorder. Further, as will be shown explicitly below, the relevant temperatures are small compared to the hole Fermi energy ($E_F$), so that $F$ is to a good approximation simply the carrier ground state energy in the given spin configuration. The crucial quantity governing $T_c$ is the change in $F$ as the spin configuration goes from disordered to ordered. As we will show below, the change in $F$ involves several competing effects not evident in previous calculations of $T_c$ such as the static mean field theory \cite{1}.

$$H_{KL} = \sum_{i,j} J_{AF}(R_i − R_j)S_i \cdot S_j$$
To obtain more detailed information, we consider here the idealized Kondo lattice model, in which \( J_{AF} \), \( V_r \) and \( W \) in Eq.1 are neglected. These may easily be included in our formalism, and will be discussed in a future paper. We absorb the magnitude of the spin into the definition of \( J \) and study first the \( T \to 0 \) limit of the fully polarized ferromagnetic state, \( S_i \parallel \hat{z} \) at \( T = 0 \). In this state the carriers feel a spatially varying spin dependent potential with mean strength \( xJ \) per GaAs unit cell. It leads to a shift in the band offset (upwards for one species and downwards for the other) proportional to \( xJ \). For values of \( J \) less than a critical value \( J_c \), the spin dependent potential does not lead to any bound states. The wave functions are scattering states and the band offset is essentially \( \pm xJ \). The energy is given simply by filling the up and down bands up to the appropriate chemical potential. The critical value \( J_c \) is identical to the local zero field critical value \( J_c^{\text{loc}} \) as the ‘RKKY’ limit although strictly speaking the term RKKY refers to the behavior of \( \chi \) at distances long compared to the spacing between carriers and Eq.3 applies even for spins closer together than this distance. When Eq.3 applies, the ordering wavevector is the one which maximizes \( \chi \) and \( T_c \sim J^2 \), which is also the static mean field result. Note that for \( n \) greater than a (numerically small) critical value \( n_c \), the maximum in \( \chi \) is at a nonzero wavevector, leading to a non-ferromagnetic ordered state.

In the \( J \to \infty \) limit, at all times each carrier is bound to an Mn site with a binding energy proportional to \( J \) and spin parallel to the Mn spin on that site. The dependence of energy on spin configurations arises because in the paramagnetic state some hopping processes are blocked and is therefore set by the impurity band width which is never large (because the Mn are dilute) and vanishes as \( J \to \infty \) due to the contraction of the Bohr radius of the bound state. In this limit \( T_c \) depends crucially on the impurity band filling. For a full impurity band (one carrier per Mn) no low energy hopping processes are possible in a fully polarized ferromagnetic state; the ground state for a nearly filled impurity band is antiferromagnetic or phase separated. The static mean field theory does not capture this physics at all, predicting instead a \( T_c \sim J^2 \) for all \( J \).

We now present a dynamical mean field theory which gives a reasonable account of the small and intermediate \( J \) regime as well as the crossover to the ‘impurity band’ regime. It however does not adequately treat the band narrowing arising from extreme wavefunction localization so breaks down at some \( J \gg J_c \). We model the GaAs:Mn system as a lattice of sites, which are randomly nonmagnetic (with probability \( 1 - x \)) or magnetic (with probability \( x \)). Standard arguments show that the relevant physics may then be determined from the local (momentum-integrated) Green function \( G_{\text{loc}}^{a,b}(\omega) = b^3 \int \frac{d^p r}{(2\pi)^p} \left( \omega - \sum_{a\beta} \delta_{aa}(\omega) - \varepsilon_{pa} \right)^{-1} \). \( G_{\text{loc}} \) is in general a matrix in spin and band (not shown) indices and depends on whether one is considering a magnetic (a) or non-magnetic (b) site. Being a local function, it is the solution of a local problem specified by the partition function \( Z_{\text{loc}} = \int d\text{Se}^{-S_{\text{loc}}} \) with action \( S_{\text{loc}} = g_{\text{loc}}^{a\beta}(\tau - \tau') c_{a\alpha}^+(\tau) c_{\alpha\beta}(\tau') + J S \sum_{a\alpha\beta} c_{a\alpha}^+(x) \sigma_{a\beta} c_{a\beta}(x) \) on the a (magnetic) site and \( S_{\text{loc}} = g_{\text{loc}}^b(\tau - \tau') c_{a\alpha}^+(\tau) c_{\alpha\beta}(\tau') \) on the non-magnetic (b) site. The a-site mean field function \( g_0^a \) can be written as \( g_0^a = a_0 + a_1 \hat{m} \cdot \sigma_{a\beta} \) with \( \hat{m} \) the magnetization direction and \( a_1 \) vanishing in the paramagnetic state. It is specified by the condition that the local Green function computed from \( Z_{\text{loc}} \), namely \( \delta \ln Z_{\text{loc}} / \delta g_0^a = (g_0^a - \Sigma)^{-1} \) is identical to the local Green function computed by performing the momentum integral using the same self energy. The momentum integral requires an upper cutoff because the \( p^2/2m \) dispersion given in Eq.4 applies only near the band edges. We take the density of states as a semicircle, \( N(\varepsilon) = b^3 \int \frac{d^p r}{(2\pi)^p} \frac{1}{\sqrt{4\varepsilon^2 - \varepsilon^2}} = \sqrt{4t^2/2\pi m^2 t^2} \) with parameter \( t \) chosen to match the band-edge density of states, \( t = (4\pi)^{2/3} / 2mb^2 \). This choice of cutoff corresponds to a Bethe lattice in infinite dimensions; the crucial point is that it has the physically correct band edge density of states, this is the only aspect important for our work. Then \( g_0 \) obeys the equation \( g_0^a(\omega) = g_0^a(\omega) = \omega + \mu - x\tilde{t}^2 (g_0^a(\omega) + JS \cdot \sigma_{a\beta})^{-1} - (1 - x)\tilde{t}^2 \delta_0^a(\omega) \) where the angular brackets denote averages performed in the ensemble defined by the appropriate \( Z_{\text{loc}} \).

The solution of the equation depends crucially on \( J/t \), \( x \) and \( T \). The inset to Fig.1 shows the majority-spin density of states corresponding to the \( T = 0 \) ferromagnetic state. For small \( J \) we see the expected shift proportional to \( xJ \). For \( J > J_c \) an impurity band centered at \( -J \) and containing \( x \) states is seen to split off from the main band. The DMFT \( J_c \) is in good numerical agreement with the results of [13]; this and the obviously correct qualitative behavior confirms its reliability in the experimentally relevant regime.
As the temperature is increased, the spins disorder and eventually the magnetic transition temperature is reached. Above this temperature, \( g_0 \) is spin-independent. The main panel of Fig. 1 shows the density of states for \( T > T_c \). For \( xJ^2/(t^2 - J^2) \ll 1 \) there is a small spin independent band offset of size \( xJ^2/(t^2 - J^2) \). For \( J > t \) an impurity band forms, corresponding to carriers locally parallel to Mn spins.

The ferromagnetic transition temperature \( T_c \) may be obtained by linearizing the equation in the magnetic part of \( g_0 \), leading to an implicit equation for \( T_c \).

\[
1 = \sum_n \frac{-2h^2(xJ)^2/3}{(g_0^2 - x^2J^2)^2(1 - t^2/g_0^2) - xJ^2t^2(5/3 - J^2/g_0^2)}
\]

where temperature is contained in the Matsubara sum over the frequency \( \omega_n \) on which \( g_0 \) depends.

**FIG. 1.** Main panel: density of states at \( T > T_c \) for \( J = 0, 0.5t, t \) and \( 2t \). Inset: majority-spin density of states at \( T = 0 \) for the same parameter values.

**FIG. 2.** Calculated transition temperature vs carrier concentration for \( x = 0.05 \) and various \( J \) values as shown.

**FIG. 3.** Calculated \( T_c \) as a function of the local exchange coupling \( J \) for a fixed value (\( x = 0.05 \)) of the Mn concentration and for different values of the hole density per Mn ion (\( n_h = 0.2, 0.5, 0.75, 0.85, 1.0 \) as shown).

Figure. 3 shows the electron density dependence of the magnetic transition temperature for \( J = 0.5t \) (less than the critical value for impurity band formation), \( J = t \) (the critical value for impurity band formation) and \( J = 1.5t \) and \( 2t \) (where the impurity band is well formed). The striking feature, evident in all three curves, is the non-monotonic behavior of the transition temperature. This has different origins in different regimes. For \( xJ^2/(t^2 - J^2) < 1 \) an analytic solution for \( T_c \) may be obtained. The details will be presented elsewhere; one result is that the density \( n_\text{max} \) at which \( T_c \) is maximized is \( n_\text{max} = \frac{1}{5}(2 - \sqrt{3 + 2J^2/t^2 - J^4/t^4} + O(xJ^2/(t^2 - J^2))) \approx 0.04 - O(J/t) \). Thus in this limit the \( T_c \) maximum is a consequence of structure in the underlying electronic susceptibility. The precise position depends on the cutoff, but is very low. For \( J > t \) the physics is dominated by the spin-polarized impurity band. In this limit \( T_c \) is controlled by the delocalization energy in the impurity band, and is therefore maximized when the band is half filled. In a filled impurity band \( (n = x) \) no low energy hopping processes are allowed in a ferromagnetic state whereas in an antiferromagnetic state hopping is allowed with amplitude \( x^{1/2}/|\Delta| \) where \( \Delta \sim J \) is the gap between the impurity and conduction band. This physics implies that very near the filled impurity band limit, the ground state is antiferromagnetic. As \( \Delta \) increases the window of antiferromagnetism decreases.

Fig. 3 shows the magnetic transition temperature as a function of magnetic coupling \( J \) for different hole densities ranging from \( n_h = 0.1/\text{Mn} \) to \( 1/\text{Mn} \) (in our conventions, \( n = x n_h \)). The collapse in \( T_c \) for the filled band is evident. The physically evident decrease of \( T_c \) at very large \( J \) due to decrease of impurity state Bohr radius is not captured by our model, so we expect that for \( J > 2t \)
Our calculation overestimates $T_c$.

Fig. 4 shows the dependence of $T_c$ on Mn concentration $x$ for $J = t$, a value of the order of the LSDA estimate. We see that simultaneous increases in the Mn concentration (by say a factor of 2) and the density (by say a factor of 4) should increase $T_c$ by more than a factor of two.

We compare our results to the predictions of other means of calculation. The ‘mean field theory’ \[5\] applied to our model predicts $T_c = x n^{1/3} J^2 / t$ at all $n, J$. In the limit $x J^2 / (t^2 - J^2) < 1$ and $n_{\text{min}} < n < n_{\text{max}}$ our analytic solution of the equations yields the mean field result but we find deviations either as $J$ approaches $t$, or when $n$ exceeds $n_{\text{max}}$ or in the extremely low density limit $n < (x J^2 / t)^{3/2}$ (not visible in the plots shown here) where $T_c \sim x n$. An alternative approach to $T_c$ involves spin-wave excitations \[6\]. In classical high-spin magnets at $T \sim T_c$ spin waves are excited throughout the Brillouin zone and $T_c$ occurs when the number of excitations (set by $T$ divided by a typical magnon energy) is large enough. The present theory may be thought of as a calculation of a typical (i.e. averaged over the zone) magnon energy (which is itself determined by the changes in electronic energy due to spin disorder) on the assumption that the spin wave excitations have no particular spatial structure. In $d = 3$ for $T$ near $T_c$ this is correct except for small amplitude critical fluctuations of no particular energetic significance. More importantly, our calculation provides detailed access to the experimentally relevant intermediate $J, n, x$ regimes.

We now briefly discuss numerical estimates of $T_c$ for GaAs:Mn. The band theory estimates \[7\] $J = 1 \text{eV}$ and $t = 0.75 \text{eV}$ (corresponding to the heavy hole mass) along with the typical density $n_h = 0.1/\text{Mn}$, implies a $J/t \approx 1.3$ and a $T_c \approx 80 \text{K}$ for $x = 0.05$ whereas the light hole mass ($t = 1.5 \text{eV}$) implies $J/t \approx 0.75$ and a $T_c \approx 50 \text{K}$. These $T_c$ values are for the single band model; the contributions of the two bands add, leading to a $T^{\text{phys}} \approx 130 \text{K}$. Although our theoretical estimates agree well with the experimental $T_c$ \[12\], this agreement should not be taken too seriously in view of the simplifying approximations of our model. Interestingly, for the LSDA $J$ the heavy hole band makes a higher contribution to $T_c$. Increase of $n$ by about 50% will increase $T_c$ by a similar amount for the heavy hole band but less for the light hole band. Increases in $J$ if it can be managed will also increase $T_c$ (although much less rapidly than the quadratic dependence predicted by the mean field theory). The most promising route to a higher temperature ferromagnet is predicted to be a simultaneous increase in $x$ to a value of order 0.1 and $n$ to about 0.02 or about 0.2/Mn.

In summary, we have presented a theory of the magnetic semiconductors which can handle both the weak coupling limit ($xJ$ less than Fermi energy $E_F$) and the intermediate coupling regime ($J > t$ but not too large). This method correctly treats the physically relevant situation in which the carriers are constrained to be locally parallel to the Mn spins and allows, for example, calculation of the resistivity and optical conductivity. Discussion of these quantities will be given elsewhere.

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