A dynamical mean-field approximation study of a tight-binding model for Ga$_{1-x}$Mn$_x$As

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The magneto-optical properties of the ferromagnetic semiconductor Ga$_{1-x}$Mn$_x$As are studied within the dynamical mean-field approximation (DMFA). A material-specific multiband $sp^3$ tight-binding Hamiltonian is employed for the dispersion of the GaAs host. The calculated density of states shows an impurity band and a distorted valence band for large and moderate values of magnetic coupling, respectively. Upon using the more realistic band structure, the ferromagnetic transition temperature is significantly closer to the experimental results than the previous predictions of $k \cdot p$ models. The optical conductivity shows a Drude-like peak at low frequencies which is suppressed by increasing of the magnetic coupling.

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Introduction- The discovery of a new generation of dilute magnetic semiconductors (DMS) with large Curie temperature has led to numerous experimental and theoretical studies. The ultimate goal is to find a DMS with a ferromagnetic transition above room temperature and suitable for use in spintronic devices. One of the most promising candidates is Ga$_{1-x}$Mn$_x$As due to its high $T_c$ and compatibility with current electronic applications. Despite this interest, some of the most basic questions about Ga$_{1-x}$Mn$_x$As are still unanswered after more than a decade of its discovery. Among them are questions about the nature of its underlying magnetic interactions and the role of the impurity band.

In this work we combine the $sp^3$ tight-binding Hamiltonian for the zinc blende GaAs with the self consistent dynamical mean-field approximation (DMFA) to study the magnetic and transport properties of Ga$_{1-x}$Mn$_x$As. We assume that the manganese (Mn) doping does not alter the tight-binding parameters of GaAs and any change in the band structure is due to the many-body effects induced by the magnetic coupling between the Mn local moments and the itinerant holes.

Several previous studies have used the tight-binding approximation for DMS. Tang and Flatté calculated the local density of states for a single Mn and two nearby Mn impurities and the magnetic circular dichroism of bulk Ga$_{1-x}$Mn$_x$As using large supercells; Mašek et al. took the Weiss mean-field approach to study the electronic structure for several DMS and Turek et al. compared both tight-binding approaches. Large-scale Monte-Carlo studies of real-space tight-binding Hamiltonians including only the three valence bands has also been performed.

Our DMFA calculation incorporates quantum self-energy corrections which were not included in most of the studies described above. We also incorporate valence and conduction bands on an equal footing while Yildirim et al. included only valence bands. Because this method is non-perturbative, it allows us to study both the metallic and impurity-band regimes, as well as small and large couplings. Since the strength of the coupling between the magnetic ions and the charge carriers is comparable to the Fermi energy, temporal fluctuations included in our DMFA approach are required in a realistic calculation especially in the vicinity of the critical temperature.

We choose tight-binding parameters according to Chadi and Cohen, which give the correct band features within a sufficiently wide window of $\pm$ 2.0 eV around the Fermi energy, such as effective masses for valence and conduction bands and the gap at the center of the Brillouin zone. Our model also includes the spin-orbit interaction of the parent material which was proved very important in previous studies of Ga$_{1-x}$Mn$_x$As.

The inclusion of the realistic band structure of the parent material leads to more realistic results. For example, the band repulsion between the conduction band and the impurity band, which was absent in some previous calculations based on the $k \cdot p$ approach results in a significant reduction of the bandwidth of the impurity band. Narrowing of the impurity bandwidth increases the localization of the mediating holes, which suppresses the estimated critical temperature. Consequently, we find that the calculated $T_c$ is significantly smaller, and closer to the experimental results.

Model- We model the magnetic interactions in Ga$_{1-x}$Mn$_x$As using the simplified Hamiltonian proposed by Zaránd and Jánko,

$$H = H_0 + J \sum_i S(R_i) \cdot J(R_i),$$

where $H_0$ is a 16×16 matrix including both electronic dispersion and spin-orbit coupling of the $sp^3$ holes of the parent material. Our tight-binding parameters reproduce the correct band structure for the heavy and light bands around the center of the Brillouin zone, the split-off energy gap and the gap between the valence and the conduction bands. The spin-orbit coupling is modeled with a term $\lambda_s L \cdot S$ where $\lambda_{Ga}$ and $\lambda_{As}$ are interaction constants for Ga and As atoms respectively. The second term in Eq. 1 describes the interaction between Mn.
spins and mediating holes, where $J_c$ is the exchange coupling between the localized moments with spin $\mathbf{S}(R_i)$ and the holes total angular momentum density $\mathbf{J}(R_i)$, both at site $R_i$. Since the spins of Mn$^{2+}$s are relatively large ($S = 5/2$) we treat them as classical vectors.

We employ the DMFA algorithm\cite{112313} to calculate the magnetic and transport properties of the material within a Green’s function formalism\cite{121118}. We calculate the average magnetization of the manganese ions:

$$
M = \frac{1}{Z} \int d\Omega e S^z \exp \{-S_{\text{eff}}(S)\} 
$$

with the partition function $Z = \int d\Omega \exp[-S_{\text{eff}}(S)]$, and the effective action $S_{\text{eff}}(S) = -\sum_i \log \det \tilde{\mathcal{G}}_0^{-1}(i\omega_n) + J_c S \cdot J_c$\cite{121118,201121} where $\tilde{\mathcal{G}}_0$ is the cluster excluded Green function.

The average polarization of the charge carriers at the chemical potential is defined as:

$$
P = \frac{\sum_j j^z_i n_i(\mu)}{\sum_i n_i(\mu)}
$$

where $j^z_i$ is the $z$ component of the total angular momentum and $n_i$, the density of holes in the $i$th band.

To compute the optical conductivity we use the Kubo formula\cite{221924}:

$$
\sigma_{ij}(\omega) = \frac{\pi}{\omega} \int \nu [f(\nu) - f(\nu + \omega)] \times
\sum_k Tr[v_i(k)A(k,\nu + \omega)v_j(k)A(k,\nu)],
$$

where $Tr$ is the trace, $v_i(k)$ is the velocity along the $i$ direction, $A(k,\omega)$, the spectral function at momentum $k$ and frequency $\omega$, and $f(\omega)$, the Fermi function. The group velocities involve the hopping terms in the Hamiltonian and are calculated as $v_i(k) = \frac{\delta H(k)}{\delta A_{ij}}|_{A=0}$, where $H$ is the Hamiltonian and $A$ the vector potential.

**Results and Discussion** - We first focus on the density of states (DOS) of the charge carriers at the doping $x = 0.05$ where $T_c$ is among the highest reported.\cite{112313} In Figure 1 and 2 we assume 50% hole compensation due to anti-site and interstitial doping, such that $n_h = x/2 = 0.025$. This corresponds to the optimum filling in the impurity band regime. In any case, the profile of the DOS changes minimally with filling. Figure 1 shows that noticeable changes in $T_c$ happens when the impurity band is forming. Inset: Average magnetization of the ions versus temperature for the same couplings.

![FIG. 1: (color online). Total density of states versus energy at the edge of the valence band for $x = 0.05$, $n_h = x/2 = 0.025$ and several coupling strengths at $T = 58$ K. The edge of the conduction band lies at the right bottom corner. The impurity band is not well separated from the valence band up to large couplings $(J_c \sim 4 \text{ eV})$. The vertical dashed line shows the location of the chemical potential for $J_c = 4.0 \text{ eV}$. Band repulsion between the conduction and impurity bands leads to reduction of the impurity bandwidth. Inset: sp$^2$ total density of states for pure GaAs and Ga$_{0.95}$Mn$_{0.05}$As with $J_c = 4.0 \text{ eV}$. We use a Fourier transform filter to reduce the noise in the calculated density of states. The noise is in regions away from the Fermi energy and is due to the effect of the k mesh in the coarse graining step of the DMFA self-consistency loop. We have used up to 62,000 k points in this calculation.](image1)

![FIG. 2: (color online). Average hole polarization versus temperature for different values of $J_c$ at $n_h = x/2 = 0.025$. The polarization is frustrated since it does not reach its maximum allowed value, $P = 3/2$, at $T = 0$. Comparison with Figure 1 shows that noticeable changes in $T_c$ happens when the impurity band is forming. Inset: Average magnetization of the ions versus temperature for the same couplings.](image2)
The impurity band is not completely separated until couplings around ∼ 4 eV. As we expected, the repulsion between the conduction and the impurity band confines the latter within the band gap and narrows down the impurity bandwidth. This leads to localization of the charge carriers which in return reduces the ferromagnetic transition temperature, $T_c$.

Figure 2 displays the temperature dependence of the hole polarization for different couplings. We choose a narrow energy window (0.1 eV) at the chemical potential and calculate the average $\mathcal{P}$ within this window using Eq. (3). The polarization is frustrated since it does not reach its maximum allowed value, $\mathcal{P} = 3/2$, at $T = 0$, and it is further reduced with decreasing magnetic coupling strength. Comparison with Figure 3 shows that noticeable changes in $T_c$ correspond to couplings where the impurity band is forming and separating from the valence band. On the other hand, $T_c$ does not change once the impurity band is fully separated from the valence band. The maximum $T_c$ we obtained is ∼ 220 K which is closer to experimental results than previous estimates using the $k \cdot p$ model. One can also see the same behavior from the inset which shows the average magnetization of the Mn$^{2+}$ ions versus temperature. However, note that within the DMFA the average magnetization is not frustrated. This is an artifact of DMFA. Inclusion of non-local correlations using cluster methods reduces the value of the magnetization.

Figure 3 displays $T_c$ versus hole filling for $x=5\%$ and different couplings. For large $J_c$, in the impurity-band regime, $T_c$ is maximum at the optimum filling of $n_h = x/2$. This is consistent with studies based on the impurity band picture. For moderate couplings around $J_c \sim 1.5$ eV and fillings $n_h > 2\%$, $T_c$ increases by decreasing the hole concentration. Recent studies in thin films find $T_c$ to be proportional to the hole concentration for a wide range of $n_h$ values. This might indicate a filling larger than the doping, $n_h > x$, in those experiments. The inset in Figure 3 shows $T_c$ versus $J_c$ for fixed hole concentration $n_h = x/2 = 2.5\%$. The plateau at large couplings is due to the absence of non-local correlations within the DMFA.

Finally, we calculate the optical conductivity in the direction perpendicular to the magnetization for different couplings at $x=5\%$, $n_h=2.5\%$ and $T=58$ K. The conductivity is suppressed by increasing of $J_c$ due to the increasing localization of the holes. The inset displays the total density of states versus energy at the edge of the valence band for the same parameters.

Conclusions: We study the magnetic and transport properties of the diluted magnetic semiconductor...
Ga$_{1-x}$Mn$_x$As within the framework of the multi-orbital DMFA. We employ a semi-empirical $sp^3$ tight-binding approximation to model the band structure of the parent material. We choose tight-binding parameters according to Chadi\textsuperscript{[22]} which reproduce the correct band structure within the relevant energy window, including the valence, split-off and conduction bands. The spin-orbit coupling is modeled with a term $\lambda \mathbf{L} \cdot \mathbf{S}$. We find that this more realistic band structure leads to more realistic results.

The density of state shows a distorted valence band for moderate coupling strengths, $J_z \sim 1.5$ eV, and an impurity band for couplings larger than $J_z \sim 3.0$ eV. The band repulsion between the impurity and conduction bands reduces the bandwidth of the impurity band. This in turn leads to localization of the itinerant holes and reduction of $T_c$. The average hole polarization displays a clear magnetic frustration. Moreover, the optical conductivity $\sigma_{xx}$ shows a Drude-like peak at low frequencies which disappears with increasing couplings. While our conductivity results are similar to previous tight-binding calculations\textsuperscript{[22]} they do not display the low energy peak observed experimentally.\textsuperscript{[22]} Despite the inclusion of band repulsion effects and that our impurity band is narrower than found previously, it is still too wide. This wide impurity band smears out any low energy features. We believe that the inclusion of corrections including cavity field and non-local correlations into the dynamical mean-field approximation will result in further suppression of the impurity bandwidth and, possibly, development of the peak in the conductivity.

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