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Magnetism in semiconductors: A dynamical mean field study of ferromagnetism in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$

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We employ the dynamical mean field approximation to perform a systematic study of magnetism in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$. Our model incorporates the effects of the strong spin-orbit coupling on the $J = 3/2$ GaAs valence bands and of the exchange interaction between the randomly distributed magnetic ions and the itinerant holes. The ferromagnetic phase transition temperature T_c is obtained for different values of the impurity-hole coupling J_c and of the hole concentration n_h at the Mn doping of $x = 0.05$. We also investigate the temperature dependence of the local magnetization and spin polarization of the holes. By comparing our results with those for a single band Hamiltonian in which the spin-orbit coupling is switched off, we conclude that the spin-orbit coupling in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ gives rise to frustration in the ferromagnetic order, strengthening recent findings by Zaránd and Jankó (Phys. Rev. Lett. **89**, 047201 (2002)).

The discovery of ferromagnetism in GaAs doped with Mn has renewed interest in the properties of diluted magnetic semiconductors.[1] Since these materials are good sources of polarized holes, they may form the basis of a new field called *spintronics*. [2] Spintronic devices employ both the spin and the charge of the carrier to convey information.

In $\text{Ga}_{1-x}\text{Mn}_x\text{As}$, the Mn ions are in the Mn^{2+} state with a half-filled d shell of total spin $S = 5/2$. [3, 4] Since Mn^{2+} ions primarily replace Ga^{3+} , they act as effective acceptors by supplying holes as well as localized spins. The valence band of pure GaAs is p -like so the strong spin-orbit interaction couples the $l = 1$ angular momentum of the p orbitals to the electron spin ($s = 1/2$), resulting in a total spin $J = l + s = 3/2$ for the valence holes. [5] As discussed by Zaránd and Jankó, [6] the strong spin-orbit coupling also induces an *anisotropic* carrier-mediated interaction between the Mn ions and, as a consequence, frustration in their ferromagnetic order. While the results of Ref. 6 were limited to the metallic regime and to small values of the Mn-hole coupling, a different approach suggests the presence of an impurity band in the dilute limit. [7]

In this Letter, we employ the Dynamical Mean Field Approximation (DMFA) [8, 9, 10, 11] to perform a systematic analysis of ferromagnetism in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$, including the effects of strong spin-orbit coupling on the $J = 3/2$ GaAs valence band. The DMFA includes the spin-split impurity band through quantum self-energy corrections which are not included in other mean-field theories. Because this method is non-perturbative, it allows us to study both the metallic and impurity-band regimes as well as both small and large couplings. We show how the spin-orbit interaction affects the ferromagnetic critical temperature T_c , the hole polarization, and the Mn magnetization. By comparing our results with those for a single band Hamiltonian without spin-orbit coupling, we conclude that strong spin-orbit cou-

pling in $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ produces frustration for all coupling strengths. For carrier concentrations smaller than the doping, both T_c and the polarization of the carriers are reduced for all values of the coupling. For larger carrier densities and large couplings, we unexpectedly find that frustration induces a small but finite T_c , in sharp contrast with the vanishing T_c found when the spin-orbit is neglected.

Our starting point is the simplified Hamiltonian proposed in Ref. 6:

$$H = H_0 - J_c \sum_{R_i} \mathbf{S}_i \cdot \hat{\mathbf{J}}(R_i). \quad (1)$$

The first term includes the electronic dispersion and the spin-orbit coupling of the $J = 3/2$ valence holes within the spherical approximation. [12] The second term is the dominant part of the interaction between the Mn spins and the valence holes, [13] with J_c the exchange coupling and $\hat{\mathbf{J}}(R_i)$ the total $J = 3/2$ spin density of the holes at the site i of a Mn ion with spin \mathbf{S}_i . The relatively large magnitude of the Mn local moment justifies a classical treatment of its spin.

Within the spherical approximation, the Hamiltonian of pure GaAs is rotationally invariant. Hence, H_0 is diagonal in a *chiral* basis,

$$H_0 = \sum_{\mathbf{k}, \gamma} \frac{k^2}{2m_\gamma} \tilde{c}_{\mathbf{k}\gamma}^\dagger \tilde{c}_{\mathbf{k}\gamma}, \quad (2)$$

where $\tilde{c}_{\mathbf{k}, \gamma}^\dagger$ creates a chiral hole with momentum \mathbf{k} parallel to its spin and $\hat{\mathbf{J}} \cdot \hat{\mathbf{k}} = \pm 3/2$ or $\pm 1/2$. The two effective masses $m_\gamma = m_h \approx 0.5m$ and $m_\gamma = m_l \approx 0.07m$ correspond to the heavy and light bands with $\gamma = \pm 3/2$ and $\pm 1/2$ respectively (m is the electron mass).

Unfortunately, the exchange interaction term obtains a rather complicated momentum-dependent form in the chiral basis [6] that is responsible for the frustrated order

of the Mn. The competition between the strong spin-orbit coupling on the hole bands, which aligns the hole spin parallel to its momentum, and the exchange interaction with the local moment, which aligns the hole and local spins, precludes all of the carrier density from mediating the magnetic order.

We develop a DMFA algorithm that takes advantage of the simple diagonal form of H_0 in the chiral basis and the local form of the exchange interaction in the non-chiral basis. The coarse-grained Green function matrix in the non-chiral fermion basis is

$$\hat{G}(i\omega_n) = \frac{1}{N} \sum_{\mathbf{k}} [i\omega_n \hat{I} - \hat{\epsilon}(k) + \mu \hat{I} - \hat{\Sigma}(i\omega_n)]^{-1}, \quad (3)$$

where N is the number of \mathbf{k} points in the first Brillouin zone and $\hat{\epsilon}(k) = \hat{R}^\dagger(\hat{\mathbf{k}}) \frac{k^2}{2m_\gamma} \hat{R}(\hat{\mathbf{k}})$ is the dispersion in the spherical approximation. Here, \hat{R} are spin 3/2 rotation matrices that relate the fermion operator, $c_{\mathbf{k}\gamma}$ to its chiral counterpart, $\tilde{c}_{\mathbf{k}\gamma} = R_{\gamma\nu}(\hat{\mathbf{k}}) c_{\mathbf{k}\nu}$. The mean-field function $\hat{G}(i\omega_n) = [\hat{G}^{-1}(i\omega_n) + \hat{\Sigma}(i\omega_n)]^{-1}$ is required to solve the DMFA impurity problem. At a nonmagnetic site, the local Green function equals the mean-field function, $\hat{G}_{non} = \hat{G}$. At a magnetic site, \hat{G}_{mg} must solve a local problem. By treating disorder in a fashion similar to the *coherent potential approximation* (CPA)[14] for a given local spin configuration, we obtain $\hat{G}_{mg}(i\omega_n) = [\hat{G}^{-1}(i\omega_n) + J_c \mathbf{S} \cdot \hat{\mathbf{J}}]^{-1}$.

Now $\hat{G}_{mg}(i\omega_n)$ must be averaged over all possible spin orientations at the local site and over all possible impurity configurations on the lattice. The former is implemented by using the effective action [15]

$$S_{eff}(\mathbf{S}) = - \sum_n \log \det \left[\hat{G}(i\omega_n) (\hat{G}^{-1}(i\omega_n) + J_c \mathbf{S} \cdot \hat{\mathbf{J}}) \right] \times e^{i\omega_n 0^+} \quad (4)$$

to average over the angular distribution of the local spins:

$$\langle \hat{G}_{mg}(i\omega_n) \rangle = \frac{1}{\mathcal{Z}} \int d\Omega_{\mathbf{S}} \hat{G}_{mg}(i\omega_n) \exp[-S_{eff}(\mathbf{S})], \quad (5)$$

where $\mathcal{Z} = \int d\Omega_{\mathbf{S}} \exp[-S_{eff}(\mathbf{S})]$. If the Mn ions are randomly distributed with probability x , the configurationally-averaged Green function reads $\hat{G}_{avg}(i\omega_n) = \langle \hat{G}_{mg}(i\omega_n) \rangle x + \hat{G}(i\omega_n)(1-x)$.

Finally, the magnetization of the Mn ions can be calculated as:

$$M^z = \frac{1}{\mathcal{Z}} \int d\Omega_{\mathbf{S}} S^z \exp[-(S_{eff}(\mathbf{S}) - \beta \mathbf{S} \cdot \delta \mathbf{H}^z)], \quad (6)$$

where a small magnetic field $\delta \mathbf{H}^z$ is applied to break the symmetry along a preferential direction, i.e., the z axis.[16] By fitting the magnetization $M^z(T)$ in the vicinity of the transition to the Curie-Weiss form, we can extract the value of T_c for each set of parameters studied.

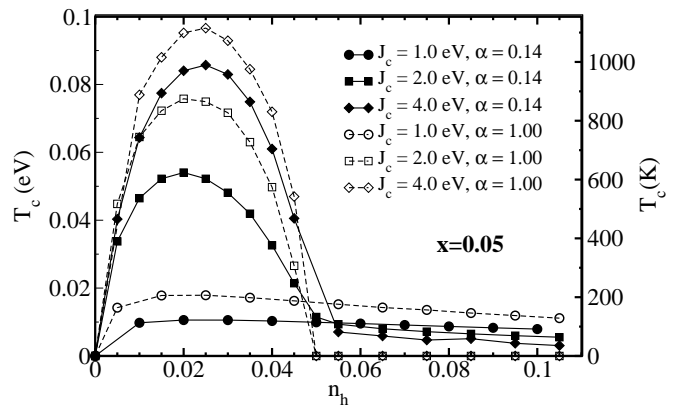


FIG. 1: Phase transition temperature T_c versus carrier concentration n_h for Mn doping $x = 0.05$, various J_c values and effective mass ratios, α .

We focus on the doping of $x = 0.05$ associated with the highest T_c reported.[17, 18, 19, 20, 21] In order to clearly elucidate the role played by the spin-orbit coupling, we introduce two parameters. One is the hole mass of an equivalent system composed of two degenerate bands with the same average kinetic energy as our system: $2m_{eq}^{\frac{3}{2}} = m_h^{\frac{3}{2}} + m_l^{\frac{3}{2}}$. The other is the ratio of light and heavy hole effective masses, $\alpha = \frac{m_l}{m_h}$. The chirality can be switched off by setting $\alpha = 1$ while keeping m_{eq} constant.

Fig. 1 presents our results for the phase transition temperature T_c as a function of the hole concentration n_h for a fixed Mn concentration, $x = 0.05$, and for various values of J_c and α . There are two regimes corresponding to small and large values of the interaction strength, J_c . For large J_c , the holes form bound states with the Mn impurities, an impurity band develops inside the GaAs gap, and the properties of the host are greatly affected. The value of J_c at which the impurity band appears depends upon the value of α .

First consider the non-chiral case with $\alpha = 1.0$ so that the spin-orbit coupling is turned off. For $J_c = 1.0$ eV the system is far from the impurity-band regime and T_c has a relatively slow variation with respect to n_h . For $J_c = 2.0$ eV the system is beyond the threshold for the formation of an impurity band, which then dominates the physics. The maximum T_c occurs when the impurity band is nearly half filled ($n_h \approx x/2$).[22] For $J_c = 4.0$ eV the impurity band is well established and the maximum T_c is large. Because ferromagnetic order restricts the hopping of holes when the impurity band is full, T_c vanishes for $J_c > 1.0$ eV and $n_h > x$. An antiferromagnetic ground state is energetically more favorable in the regime $n_h > x$ [22] because the carriers can then easily hop from one impurity site to another. Magnetization curves for $J = 2.0$ and 4.0 eV and for $n_h > x$ provide evidence for antiferromagnetism. They fit a Curie-Weiss

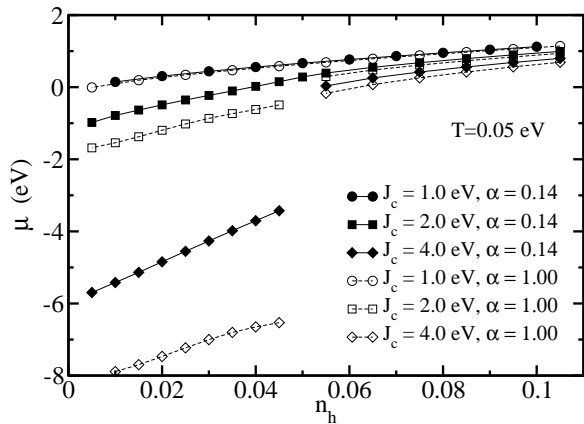


FIG. 2: Chemical potential, μ , versus n_h for Mn doping $x = 0.05$ and $T = 0.05$ eV using the same values of J_c and α as in Fig. 1.

functional form, $M^z = \frac{M_0^z}{1 + T/\Theta}$ with a positive Θ ranging from 40 to 117K, indicating antiferromagnetic order for $\alpha = 1.0$ and $n_h > x$.

We now consider the chiral case with $\alpha = 0.14$ so that the spin-orbit coupling in GaAs is turned on. The effects of frustration on the ferromagnetic critical temperature are readily seen in Fig. 1. For $J_c = 1.0$ eV, T_c consistently lies below its non-chiral counterpart. At $J_c = 2.0$ and 4.0 eV, the difference between the chiral and non-chiral results is even larger. For $n_h < x$, the non-chiral T_c is always higher than the chiral T_c . However, for $n_h > x$, the chiral results continue to yield a finite although greatly diminished T_c in the same regime where T_c vanished when $\alpha = 1.0$. Surprisingly, the ferromagnetic T_c survives at large dopings due to the intrinsic frustration in the system. Strong spin-orbit scattering allows the impurity-band carriers to hop between impurity sites that are ferromagnetically aligned even when the impurity band is full, thereby stabilizing the magnetic order.

The formation of an impurity band is responsible for many of the outstanding properties of this system.[22] Impurity-band formation appears as a discontinuity of the hole chemical potential μ at $n_h = x$. Fig. 2 depicts μ versus n_h for $T = 0.05$ eV (580K) and for the same values of J_c and α used in Fig. 1. The presence of an impurity band depends on both J_c and α . When $J_c = 1.0$, no impurity band is present so the chiral and non-chiral μ are almost identical and neither shows a discontinuity. When $J_c = 2.0$ eV and $\alpha = 1.0$, the impurity band has already split from the valence band. At the same value of $J_c = 2.0$ eV but when $\alpha = 0.14$, the change in slope of μ at $n_h = x$ indicates that the impurity band is present but that it overlaps with the main band. When $J_c = 4.0$ eV and $\alpha = 0.14$, the hole chemical potential is discontinuous at $n_h = x$, signaling the splitting of the impurity band from the valence band. For these larger values of J_c , the non-chiral μ always lies below the chiral value, indicating

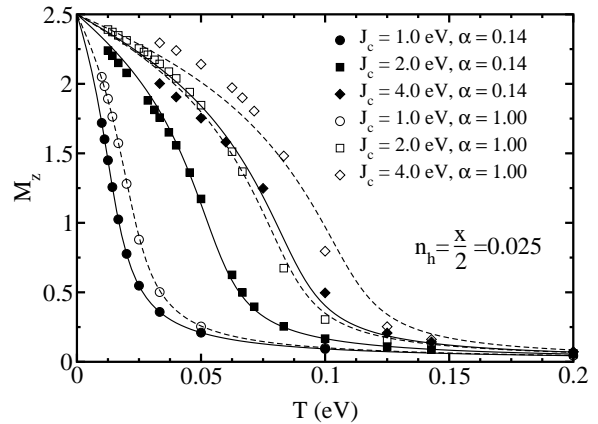


FIG. 3: Magnetization versus temperature for different chirality modes and different values of J_c at $n_h = x/2 = 0.025$ and $\delta H^z = 0.004$ eV. The curves are the static mean field fits with T_c values equal to those given by our results, solid curves fit the results for $\alpha = 0.14$; dashed curves the ones for $\alpha = 1.0$.

that the non-chiral impurity band lies at lower energies than the chiral one. Since the interaction between the impurity moments is mediated by the host of holes, the less pronounced impurity band for $\alpha = 0.14$ is another signature of the frustration produced by chirality.

Optical conductivity measurements[23] show that the impurity band is split from the valence band at the doping of $x = 0.05$. This fact and our results for T_c suggests a value of J_c between the value of 1.2 eV obtained from photoemission techniques[4] and the value of 4.5 ~ 6 eV inferred from infrared spectroscopy.[3]

The magnetization also reveals the effects of frustration. In Fig. 3, the temperature dependence of the magnetization is plotted with $n_h = x/2 = 0.025$. Our results are compared with Curie-Weiss fits for the same T_c values. When $J_c = 1.0$ eV, the mean-field curves perfectly fit the data not only at large temperatures but also below T_c . But for larger J_c values and low temperatures, discrepancies appear between the static mean-field curves and the DMFA results. For $\alpha = 1.0$, the magnetization in our model lies above the mean-field Heisenberg magnetization when $T < T_c$ since the scattering between the itinerant carriers and the localized spins becomes coherent at low temperatures, thereby enhancing the magnetic order of the local ions. This effect has also been seen in the double exchange model.[24] However, for $\alpha = 0.14$, frustration reduces the low-temperature coherence between the carriers and local ions. So when $T < T_c$, the magnetization curves lie below the mean-field Heisenberg predictions.

Lastly, we study the carrier spin polarization, \mathcal{P} , which is constructed from appropriate sums of the Green function. Control of \mathcal{P} is important for *spintronics* applications, since the spin polarization of the holes is required to transport information. Fig. 4 illustrates the temperature dependence of the hole polarization when $n_h = x/2 = 0.025$ for several values of J_c and α . For

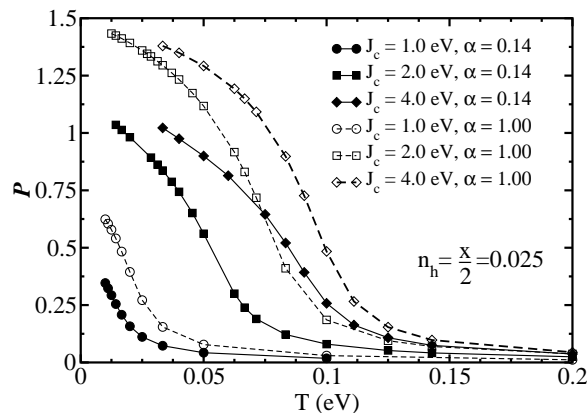


FIG. 4: The hole polarization P versus temperature at $n_h = x/2 = 0.025$ and $\delta H^z = 0.004$ eV.

all J_c values, chirality suppresses the polarization. For $\alpha = 1.0$, large J_c , and low temperatures, P approaches $3/2$ since all holes occupy the lowest energy level with $j_z = +3/2$. But due to frustration even for large J_c and low temperatures, P for $\alpha = 0.14$ is significantly smaller. This agrees with previous calculations of the zero-temperature polarization, where the destructive effects of the spin-orbit interaction were also found.[25]

In summary, we have studied the diluted magnetic semiconductor $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ using a non-perturbative multi-orbital DMFA algorithm incorporating the effects of the strong spin-orbit coupling. We have calculated the ferromagnetic critical temperature, hole chemical potential, local ion magnetization, and hole polarization for a

broad range of model parameters. We find that the spin-orbit coupling leads to frustration and reduced magnetization when the hole concentration n_h is smaller than the impurity concentration x , in agreement with previous perturbative calculations.[6] In addition, we find that this behavior persists for large values of J_c , and that frustration greatly reduces the transition temperature T_c and the polarization of the carriers for all J_c . Finally, when J_c is large, we find the surprising result that frustration induces a region of finite T_c for $n_h > x$.

This approach has promising future ramifications. It can be extended to study other magnetic semiconductors and realistic devices such as semiconducting heterostructures and quantum dot systems, which can be tailored to take full advantage of the intrinsic anisotropy of the ferromagnetic order. More sophisticated approaches, such as the dynamical cluster approximation (DCA),[26] may be used to go beyond the single-site approximation and explore the cooperative and glassy effects of frustration, such as the reduction in the local magnetization at low temperatures.

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