Diluted magnetic semiconductors based on III-V compounds are attracting considerable attention due to their combination of magnetic and semiconducting properties, that may lead to spintronic applications [6], [7]. Ga$_{1-x}$Mn$_x$As is the most studied of these compounds with a maximum Curie temperature $T_C \approx 110$ K at low doping $x$, and with a carrier concentration $p=(n/x)<1$ due to the presence of As antisite defects [6] or Mn interstitials [8]. It is widely believed that this ferromagnetism is carrier-induced, with holes introduced by doping mediating the interaction between $S=5/2$ Mn-spins. This Zener mechanism operates in other materials as well [9].

In spite of the excitement around 110 K DMS, room temperature ferromagnetism should be achieved for potential applications, with logic and memory operations in a single device. For this reason, a goal of the present effort is to analyze the dependence of $T_C$ on the parameters $x$, $p$, and $J/t$, helping in setting realistic expectations for DMS potential technological applications. This goal can only be achieved with good control over the many-body aspects of the problem, and for this purpose lattice Monte Carlo (MC) techniques are crucial, improving on previously employed MF approximations. Our results lead to an optimistic view in this respect, since $T_C$ is found to increase linearly with $x$ up to $x\sim 0.25$.

Our effort builds upon previous important DMS theoretical studies [2], [3], [7], [9], [10]. However, to analyze whether $T_C$ can be substantially increased from current values, techniques as generic as possible are necessary. In particular, both the strong interactions and disorder must be considered accurately, with computational studies currently providing the best available tools. For these reasons, our work differs from previous approaches in important qualitative aspects: (1) Some groups use a continuum six-band description of DMS [9], (2) Other theories assume carriers strongly bounded to impurity sites [9], and employ Hartree-Fock approximations. (3) Dynamical MF theory (DMFT) [8] may not capture the percolative character of DMS, with a random impurity distribution and cluster picture [9], [8], (4) Other approaches use MF uniform states [8], or introduce a reduced basis in simulations [11]. While the previous work is important in describing current DMS materials, our goal is to establish the phase diagram of a DMS model avoiding MF approximations.

For the above mentioned reasons, here a generic MC study of a lattice spin-fermion model for DMS is reported. The Hamiltonian is

$$\hat{H} = -t \sum_{\langle ij \rangle, \sigma} \hat{c}_{i \sigma}^\dagger \hat{c}_{j \sigma} + J \sum_I \hat{S}_I \cdot \hat{\sigma}_I,$$

where $\hat{c}_{i \sigma}^\dagger$ creates a hole at site $i$ with spin $\sigma$, and the hole spin operator interacting antiferromagnetically with the localized Mn-spin $\hat{S}_I$ is $\hat{\sigma}_I = \hat{c}_{I \alpha}^\dagger \sigma_{\alpha,\beta} \hat{c}_{I \beta}$. The carrier can visit any site of the lattice (assumed cubic [11]) by hops introduced by doping mediating the interaction between $S=5/2$ Mn-spins. This Zener mechanism operates in other materials as well [9].

The MC technique used here is as in Mn-oxides investigations [1] show that an intermediate or large $J/t$ plays a role analogous to a Hubbard $U/t$ at any $x$ [9], [11]. At low $x$, the probability of nearest neighbors (NN) Mn-spins is also low (0.0625 at $x=0.25$), justifying the neglect of an antiferromagnetic (AF) Mn-Mn coupling. The hole motion is described by a one-band tight-binding model, while a more realistic model should include many bands as well as spin-orbit interaction [1]. Despite this simplification, our study considers the underlying lattice, absolutely necessary for a qualitative understanding of the DMS phase diagram.

The MC technique used here is as in Mn-oxides investigations [1]: it includes the full Exact Diagonalization (ED) of the hole sector for each MC spin configuration, and density-of-states expansion calculations beyond mean-field approximations.
The latter allows us to reach clusters with up to $8^3 = 512$ sites if up to 40 terms are included, reaching an accuracy comparable to ED for smaller clusters. Both methods are nearly exact, and the error bars of our results mainly arise from intrinsic thermal fluctuations and averages over several random Mn-disorder configurations. Comparing estimations of different clusters and based on previous experience with similar models [9], $T_C$ can be calculated within a $\sim 25\%$ accuracy, sufficient for our purposes [17]. The order parameter for the ferromagnetic-paramagnetic transition was taken to be the absolute value of the magnetization of the Mn-spins normalized to 1, namely $|M| = \frac{1}{N} \sum_{I,R} (\hat{S}_I \cdot \hat{S}_R)$. Size effects are better visualized in the zero-momentum spin structure factor $S(q=0) = \frac{1}{N} \sum_{I,R} (\hat{S}_I \cdot \hat{S}_R)$. Another useful quantity is the spin-spin correlation at distance $d$, $C(d) = \frac{1}{N(d)} \sum_{|I-R|=d} (\hat{S}_I \cdot \hat{S}_R)$, where $N(d)$ is the number of pairs of Mn moments separated by a distance $d$.

Typical results for small and intermediate $J/t$ of our large-scale computational effort are in Fig.1a. There $S(q=0)$ and $|M|$ vs. temperature $T$ are shown for three cluster sizes, $x = 0.1$, and $p = 0.4$. Note the small size dependence of the magnetization (inset), and the volume growth of $S(q=0)$ at fixed $T < T_C$. The estimated $T_C/t$ is $\sim 0.04$, with an uncertainty 0.01 sufficiently small for our purposes. Even with just the $4^3$ cluster, $T_C$ could be estimated fairly well, as shown in Fig.1b. This is important to simplify our computational search for optimal $T_C$’s varying many parameters. In Fig.1b, the temperature where a deviation from the high-$T$ limit is found is slightly larger than the $T_C/t = 0.04$ obtained from larger clusters (indicated). Studying the spin-spin correlation at the largest available distance, a nonzero value characteristic of an ordered ferromagnetic (FM) state was obtained at $T$ just below 0.04. Figure 1c provides another example of our comprehensive $T_C$ study, using just two cluster sizes at the $x-p$ location of our most optimal $T_C$, at fixed $J/t = 2$. Here the use of only $4^3$ and $6^3$ clusters provides once again a fairly accurate value $T_C = 0.08t$.

To understand the qualitative $T_C$ trends, first consider the simplest case: the $p$ dependence at fixed $J$ and $x$. Using the results in Fig.2a contrasted against Fig.1b (same cluster size) $T_C$ is found to change by a factor $\sim 2$, when $p$ varies from 0.1 to 0.4. However, this tendency does not continue with increasing $p$, since at $p = 1$ or beyond, a FM state is not formed: the Pauli principle reduces drastically the carrier kinetic energy, leading instead to an AF state. An example at $p = 3$ and on an $8 \times 8$ cluster (results are qualitatively similar in two and three dimensions) is in Fig.2b, where the oscillations in the spin correlations indicate staggered order. In general, the optimal $p$ is $\sim 0.5$, between the hole empty $p = 0$ and saturated $p = 1$ limits, as found with DMFT [7]. A similar result occurs in Mn-oxide models, recovered from Eq.(1) at $x = 1$. In that context, investigations at large Hund coupling, the analog of $J$ for DMS, have shown that $p = 0.5$ optimizes $T_C$ to a number $\sim 0.11 - 0.13$ [14, 15], likely an upper-bound on the $T_C$ that could be achieved with Eq.(1).
Consider now the $J/t$ dependence of $T_C$. The MF approximation suggests $T_C^{mf} \propto J^2$. However, this does not hold when more accurate methods are used in the calculations. In fact, for $J/t=\infty$ and a Mn dibute system, the holes are trapped in Mn-sites, reducing drastically the conductance and $T_C$. Small FM clusters of spins are formed at a temperature scale $T^*$, but there is no correlation between them, leading to a global vanishing magnetization [3]. These results cannot be obtained within a mean-field approximation. The large-$J/t$ ideas can be tested in our MC simulation by monitoring the short- and long-distance behavior of the spin-spin correlations $C(d)$. In a “clustered” state (large $J/t$), $C(d)$ at the shortest distance can be robust at $T<T^*$, but $C(d)$ at the largest distance vanishes due to the uncorrelated nature of the magnetism between independent clusters (see Fig.2c). This subtle effect explains the incorrect MF prediction, since $T_C^{mf} \sim T^*$, which grows with $J/t$, rather than the true $T_C$ (see also Fig.4a). Since both in the $J/t=0$ and $J/t=\infty$ limits $T_C$ is suppressed, an optimal $J/t|_{opt}$ must exist where $T_C$ is maximized. Simulation results as in Fig.2c indicate that the optimal $J/t$ value is close to 2. This phenomenon is not captured in itinerant [4] or localized [5] limits nor by DMFT [6], but it is observed in the present generic MC simulations.

The existence of a $J/t|_{opt}$ can be illustrated just using two spins and one carrier in a finite cluster at $T=0$. For any fixed angle $\theta$ between the Mn-spins, assumed coplanar, the energy is found exactly. The ground state of this $p=0.5$ system is always at $\theta=0$ (FM), while the energetically worse state is $\theta=\pi$ (AF-configuration). Their energy difference $\Delta E$ is a crude estimation of the FM state stability (Fig.2d). An optimal $J/t$ is found in all dimensions, with stability increasing with the coordination number [22]. The result Fig.2d is understood measuring the electronic density $n(i)$ of the same problem on a chain (Fig.3a). At small $J/t$, the delocalization manifests in the nearly uniform density, leading to weak FM. At large $J/t$, strong localization decouples the Mn-spins, producing again weak FM. However, there is an optimal value where the system takes advantage of $J/t$, but also allows for a nonzero effective coupling among separated classical spins, leading to a stronger FM.

Consider now the $x$ dependence of $T_C$. For simplicity, $J/t=2$ is mainly studied, which is both close to optimal and experimentally realistic [22]. Fig.3b shows $T_C$ vs. $x$ at $p=0.4$, and for two reasonable values of $t$. Experiments [4] indicate a linear growth of $T_C$ up to 5% (shown), as in the numerical results. The slope of $T_C$ vs. $x$ is in remarkable agreement with MC predictions, in a reasonable range of $t$. Regarding $x>0.05$, a reduction of $T_C$ was originally reported in experiments [4]. However, recent data gathered with an optimized annealing treatment [23] indicate a $T_C$ “plateau”. This still seems in contradiction with the linearly growing $T_C$ of the MC results, but it suggests that even more refined thin-films may continue increasing $T_C$ with increasing $x$. The MC results clearly indicate linear behavior up to $x \sim 0.25$ (Fig.3b). To the extent that our model describes DMS quantitatively, higher values of $T_C$ could be expected experimentally. Regarding the presence of a $T_C$ maximum at $x=0.25$: the origin of this effect is the growing probability with $x$ of having both holes and Mn-spins at NN-sites. In this case, AF links are formed since $J/t=2$ is not so strong to keep the link FM, reducing $T_C$ at large $x$ even at $p=0.5$. As $J/t$ grows, the effect diminishes and the maximum in $T_C$ moves toward $x=1$, as naively expected. Reciprocally, as $J/t$ decreases from 2 ($J/t=1$ shown in Fig.3b), the maximum in $T_C$ moves toward smaller $x$’s, and only $t=0.5$ can provide high-$T$ ferromagnetism. This illustrates the key role that the optimization of $J/t$ plays in these models, effect not captured by MF approximations.

$|M|$ at $T=0$ is in Fig.3c. In agreement with experiments, the $x=0.1$ result indicates a magnetization $\sim 50\%$ of its maximum value. This nonsaturated behavior originates in the random distribution of Mn-spins, since Mn-clusters are formed providing a trap to holes. Nonclustered spins are not much visited by those holes, and their spins are not polarized. With growing $x$, holes are more itinerant, polarizing the entire sample (Fig.3c) [24].

In summary, MC investigations of a spin-fermion model for DMS unveils substantial differences with previously reported results employing MF techniques. The subtle regime of intermediate $J/t$ appears the most relevant in these compounds. $T_C\sim 0.08K$ is an upper limit for the FM critical temperature, result close to those accepted for $x=1$ [4,23]. Our main results are summarized in Fig.4, that contain (a,b) the nontrivial $J/t$ dependence of $T_C$ and $T^*$, and (c) $T_C$ with varying $x$ and $p$, at optimal $J/t$. To the extent that the present model is ap-
This assumption is much used in Mn-oxide studies, where experimental value for Ga
between itinerant and localized regimes. (b) Schematic phase
T cluster, are comparable with results of Ref.[10]
T=340 K, and our simulations
refines the “true” transition
the form of uncorrelated clusters. $T_C$ is the “true” transition
temperatures are similar. The optimal $J/t$ is intermediate
between itinerant and localized regimes. (b) Schematic phase
diagram believed to be valid both in 2D and 3D, with the
clustered and FM states indicated. (c) Numerically obtained
cluster, that re-

temperature, defined as the $T$ where
sizes are expected with realistic FCC lattices.

![Image](Image109x546 to 242x635)

**FIG. 4:** (a) MC phase diagram in 2D varying $J/t$, at fixed
$x$ and $p$. At large $J/t$, a broad scale $T^*$ corresponds to the
formation of uncorrelated clusters. $T_C$ is the “true” transition

corresponds to the

$|J/t|_{opt}$∼2 must be intermediate between the itinerant and localized
limits (Fig.4a,b). This $J/t$, or larger, is expected to keep
the semiconducting nature of the state at $T>T_C$. Only
band calculations beyond our model can predict which
particular material will have such an optimal $J/t$. (ii) $x$
should be increased beyond 0.1. At $|J/t|_{opt}$, the best
value is $x$∼0.25. Currently, $x=0.14$ is the experimental
limit [20]. (iii) The number of antisite defects must
be controlled such that $p$∼0.5 ($p$∼1 would be detrimental
due to competing antiferromagnetism). (iv) As the
coordination number grows, $T_C$ grows. (v) The simplest
procedure to increase $T_C$ relies on increasing the
scale $t$. In fact, (Ga,Mn)As and (In,Mn)As have different
hybridization strengths [27], and this should be an
important consideration in studying new materials. Our
work also suggests formal analogies between DMS and
manganate models, with similar $T_C$’s, and a related
clustered state above ordering temperatures.

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