PHYSICAL REVIEW B, VOLUME 65, 241202(R)

Global versus local ferromagnetism in a model for diluted magnetic semiconductors studied with Monte Carlo techniques

Matthias Mayr,[1] Gonzalo Alvarez,[2] and Elbio Dagotto[2]
[1]*Max-Planck-Institut für Festkörperforschung, 70569 Stuttgart, Germany*
[2]*National High Magnetic Field Lab and Department of Physics, Florida State University, Tallahassee, Florida 32310*
(Received 14 March 2002; published 11 June 2002)

A model recently introduced for diluted magnetic semiconductors by Berciu and Bhatt [Phys. Rev. Lett. **87**, 107203 (2001)] is studied with a Monte Carlo technique, and the results are compared with Hartree-Fock calculations. For doping rates close to the experimentally observed metal-insulator transition, a picture dominated by ferromagnetic droplets formed below a T^* scale emerges. The moments of these droplets align as the temperature is lowered below a critical value $T_C < T^*$. Our Monte Carlo investigations provide critical temperatures considerably smaller than Hartree-Fock predictions. Disorder does not seem to enhance ferromagnetism substantially. The inhomogeneous droplet state should be strongly susceptible to changes in doping and external fields.

DOI: 10.1103/PhysRevB.65.241202 PACS number(s): 75.50.Pp, 71.30.+h, 75.40.Mg

Recent advances in experimental techniques have allowed us for the introduction of magnetic elements into semiconducting hosts beyond the solubility limit. This has stimulated the exciting research area of diluted magnetic semiconductors (DMS) with the prospect of manipulating the charge as well as the spin degrees of freedom, and its possible technological applications in such novel fields as ''spintronics.'' Prominent among these new compounds is the III-V system $Ga_{1-x}Mn_xAs$, where ferromagnetic (FM) transition temperatures as high as 110 K were obtained.[1] Although some of the basic ingredients of the physics of these materials such as the local antiferromagnetic (AF) exchange between the Mn spins and the charge carriers are known, the origin of such fairly high critical temperatures is still in question. The ferromagnetism itself can be understood as being carrier induced, in a similar fashion as the FM state in the double-exchange (DE) model for manganites at intermediate doping.[2] However, alternative descriptions based on Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions between the impurity spins are claimed to lead to transition temperatures comparable to those observed.[3] Yet it is not obvious whether RKKY can apply for systems with small Fermi energies as is the case for compensated semiconductors. Additionally, the effects of disorder that accompanies chemical doping was neglected in this approach.[4] Recent annealing studies have shown the relevance of disorder and defects in DMS.[5] It was proposed,[6] based on Hartree-Fock (HF) calculations, that disorder in the dopant position and hopping amplitudes has a strong effect on the magnetic properties of these compounds, and should be incorporated in trying to understand the behavior of DMS.

The above described subtle interplay between localizing and delocalizing tendencies in these models often cannot be described correctly in a mean-field picture, but it requires nearly exact methods such as Monte Carlo (MC) techniques. A well-known example is the manganites in the low-doping regime, where mean-field approximations and exact solutions lead to different conclusions about the nature of the metal-insulator transition (MIT). The cluster formation and percolative picture that has emerged for these systems cannot be captured properly with mean-field approximations.[2] Therefore, here the model introduced in Ref. 6 for DMS in the low-doping limit describing electrons in an impurity band is investigated using MC methods, and the results will be compared to mean-field calculations.

Although the system under study is actually hole doped, it is described in terms of an electron-doped material. Therefore, the acceptor level is treated as a donor level lying approximately 100 meV below the conduction band. Furthermore, only the impurity band itself will be considered, whereas the valence- (conduction-) band states are neglected. This should be a good approximation as long as the Fermi energy of the ''electrons'' is considerably smaller than the gap between the donor state and the lowest conduction level. The simplest such model[6] takes into account the hopping between random Mn sites and the AF interaction between the impurity spins and the charge carriers. The disorder potential and Coulomb interactions are neglected and the model is given by

$$H_{DMS} = \sum_{i,j,\alpha} (t_{ij} - \mu\delta_{ij})(c^\dagger_{i\alpha} c_{j\alpha} + \text{H.c.}) + \sum_{i,j,\alpha,\beta} J_{ij} \left(c^\dagger_{j\alpha} \frac{1}{2} \boldsymbol{\sigma}_{\alpha,\beta} c_{j\beta} \right) \mathbf{S}_i , \quad (1)$$

where $c^\dagger_{i\alpha}$ is the creation operator for an electron at the impurity site i with spin α, and t_{ij} is the hopping amplitude between sites i,j. For two sites at distance $r=|i-j|$ an exponential form, $t(r)=2(1+r/a_B)\exp(-r/a_B)$ Ry, was assumed,[6] with a_B the Bohr radius associated with the impurity site and Ry its corresponding binding energy E_b. $\mathbf{S}_i$ represents the (assumed classical) spin at the Mn ($3d^5$) site with $|\mathbf{S}_i|=5/2$, $\boldsymbol{\sigma}$ denotes the vector of Pauli matrices, μ the chemical potential, and J_{ij} describes the nonlocal interaction between impurity sites and mobile holes. Owing to the localized nature of the Mn spins, it can be expressed as $J_{ij} = J\exp(-2r/a_B)$, with J being an AF coupling constant. J is chosen as the energy unit and periodic boundary conditions are imposed. The effects of an external field can easily be introduced via a Zeeman term.

Following Ref. 6, we assume the hole binding energy $E_b = 112.4$ meV (1 Ry) and, additionally, we have $J = 15$ meV. It was checked that our conclusions below do not depend crucially on the exact values of the exchange coupling and, therefore, the results should be typical for the systems considered. The lattice constant a of the GaAs lattice is known to be 5.65 Å, and $a_B = 7.8$ Å.[7] With these values, the various nonlocal couplings J_{ij} and hoppings t_{ij} can easily be calculated once a random distribution of the impurity ions within the host lattice (of size L^3) has been chosen. In addition, these systems are presumably strongly compensated through As antisite donors resulting in a relatively small effective hole concentration of 10% of the Mn ions.[1]

The previous mean-field study of H_{DMS} resulted in some interesting conclusions about the magnetic behavior of DMS.[6] In particular, an increase in T_C by a factor of ~ 2 was reported when a disorder Mn distribution was studied, compared with a periodic distribution. This is a challenging counterintuitive prediction that deserves to be tested with techniques beyond the HF approach, even more so as it is known that critical temperatures typically are overestimated by mean-field methods, as it happens, e.g., in the DE model.[2] Fortunately, Hamiltonian (1) is especially suited for MC calculations, and the procedure used here has been extensively discussed in the study of the manganites.[2]

The MC calculations were performed on a variety of lattice sizes, but the bulk of our work was done studying systems with $N = 80$ impurity sites and a doping level of $x \approx 0.02$, which lies in the neighborhood of the realistic MIT.[8] Other lattice sizes ($N = 50$–100), with slightly different values of x, yield similar results and lead to the same conclusions. The density p of charge carriers is fixed at $p = 0.1$ (with respect to the number of Mn dopants), unless otherwise noted. The random placement of Mn ions in the GaAs host lattice creates regions of high and low impurity densities, respectively. Some of the results presented below are for only one such configuration of Mn impurities, but it was confirmed that the results for other configurations are within 10% of the data shown. Furthermore, for the configuration selected the HF critical temperature T_C^{HF} is the same as for larger systems ($N = 200,300$) with similar values of x, p, and J. Most interesting is the magnetization behavior, with and without applied magnetic field. It is analyzed by measuring the magnetization per site, $m = M/N$ ($M = \sqrt{\Sigma_{ij} \mathbf{S}_i \cdot \mathbf{S}_j}$), of the Mn spins, neglecting the contribution of the charge carriers. The latter are of much less importance as they have a spin of 1/2 only, and also a density that is only a fraction of that of the Mn spins. Figure 1 compares m for the MC as well as for the HF solution, with both results obtained for the *same* configuration of Mn impurities.

In this particular case $T_C^{HF} \sim 0.85$ (similar to earlier results on larger systems[6]) for the impurity configuration considered, whereas the MC solution suggests an enhancement of FM correlations at $\sim 0.3(\pm 0.1)$. Similar values and comparable deviations are found for other configurations and system sizes as well [Fig. 2(a)], which supports the notion that mean-field treatments generally overestimate T_C's. At this same temperature, M (scaled by $|\mathbf{S}|$) starts to deviate from the totally disordered case $M = 1$ as well and also a pronounced change in dM/dT is observed (Fig. 2)(b), signaling the development of FM correlations. Furthermore, the concept of a disorder-enhanced T_C does not appear in agreement with MC. Figure 1 points out the influence of disorder as it also includes HF and MC results for the periodic case. From the magnetization data as well as the magnetic fluctuation, $\chi \propto \langle M^2 \rangle - \langle M \rangle^2$, a critical behavior around $T_C^{per} \sim 0.25 \pm 0.05$ can be deduced for the periodic system, which is slightly below the critical region for the disordered case. This

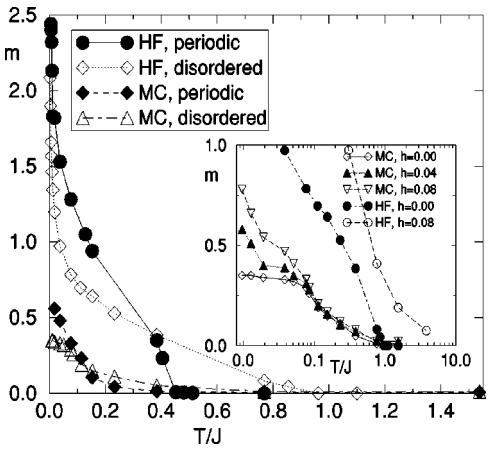

FIG. 1. Magnetization of the Mn spins vs T for model (1) with $x \approx 0.02$ and $N = 80$ impurity sites. Shown are results of MC as well as HF calculations, for either the random system or for impurities in a superlattice (periodic) configuration. Disorder strongly increases T_C in the mean-field description, but much less in the MC approach. The strong influence of a magnetic field at low temperatures is demonstrated in the inset. This is in contrast to HF, where the effect of h is remarkable at all T's (open circles, $h = 0.08$).

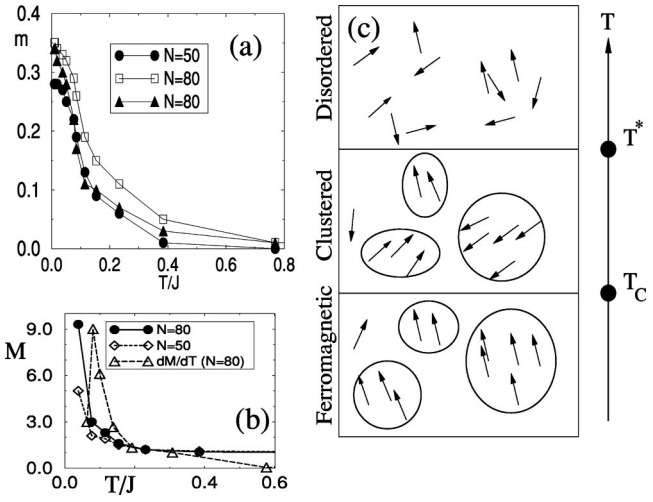

FIG. 2. (a) It shows m vs T for two system sizes, and in the case of the $N = 80$ system, the magnetization function for a different impurity configuration. In all cases, $0.02 < x < 0.025$. (b) M (scaled by $|\mathbf{S}|$) for $N = 50$, 80 and dM/dT (triangles). (c) Schematic representation of the development of FM clusters and subsequent alignment as T is lowered. Ordered domains form at T^*, and align at the critical point T_C.

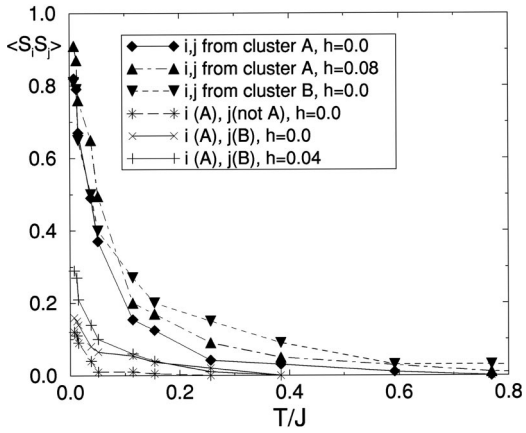

FIG. 3. MC time-averaged spin correlations per site vs T (scaled by $|\mathbf{S}|^2$) for the system shown in Fig. 1 for external fields $h=0.0$, 0.04, and 0.08. Correlations are measured either between sites i,j of the same cluster (filled symbols, cluster A or B), or between sites that do not belong to the same cluster. At high temperatures, the spin-spin correlations are approximately zero at all MC times.

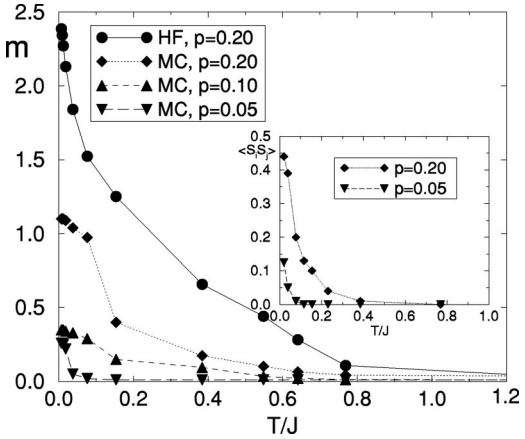

FIG. 4. m vs T for systems with different doping fractions $p=0.05$, 0.1, and 0.2, and a constant Mn density $x=0.02$ at zero external field. Weakened compensation leads to an enhanced charge-carrier rate, with extended FM domains. The inset shows the intercluster (A,B) spin correlations for $p=0.20$ and $p=0.05$, demonstrating the influence of p on the correlations.

appears in agreement with previous HF results,[6] but the reduction in T_C is much less pronounced. It will be argued below, however, that the actual transition temperature is lower than T_C^{per}. A thorough analysis reveals that the disordered system breaks apart into occupied and almost empty domains (in both approaches) and the main contribution to m is entirely due to a single, large cluster, with average particle density $\langle n\rangle^{cl}>p$. This leads to an increased temperature where FM correlations start building up compared to the periodic case since the effective local magnetic field is enhanced in this region. Single sites or very small clusters are virtually unoccupied and do not order at any finite temperature. For this reason the magnetization is never fully saturated in the MC approach as in the HF description, but reaches a plateau at about 15% of the possible value for a sample with the given Mn doping rate. If x is selected considerably lower, it would be very unlikely for such a cluster to exist and there should be no finite saturation magnetization, as it is observed for samples with $x\leqslant 0.005$.[1] The conclusions above are also supported by analyzing the spin correlations between different clusters A,B (Fig. 3).

This reveals that each cluster turns FM as the temperature is reduced,[9] but the overall magnetization will remain low as the magnetization vectors for different domains point in different directions, whereas in HF they are required to be aligned along a chosen S_z axis. In other words, the idea that delocalization of charge carriers leads to a spin alignment of the Mn ions across the whole sample,[6] appears to be an artifact of the approximation employed. It also leads to the unphysical observation of totally aligned impurity spins in the extremely diluted limit $x<0.001$ at very low temperatures. But it is interesting to note from Fig. 3 that the spin correlations between sites of a given cluster and all other sites assume a finite value at temperatures below ~ 0.08 (10 K). In addition, finite correlations develop between A,B at $T\sim 0.2$. The exact value of this temperature likely depends on the relative position between two specific domains, but is in good agreement with the value $T_C=0.08$ quoted above, where all sites were taken into account. Additionally, $M/|\mathbf{S}|$ is proportional to the number of spins below this temperature as it should be for a FM ordered state [Fig. 2(b)]. In any case, these values are *lower* than T_C^{per} and they suggest two different temperature scales in the problem: T^* (~ 0.3), where isolated magnetic domains are formed, and a smaller true critical temperature T_C (~ 0.1) where long-range order is established. Below T^*, spins in the vicinity of a given cluster start to align, i.e., the magnetic domain grows in size as the gain in kinetic energy outweighs the loss in entropy. Further delocalization of the charge carriers then causes the alignment of these domains resulting in finite bulk magnetization. The associated T_C is considerably lower than the one predicted by mean-field theory.[10] This idea is also qualitatively depicted in Fig. 2(c). In this scenario disorder enhances T^*, but *reduces* T_C.

These percolationlike effects are also visible once a magnetic field is applied. As shown in the inset of Fig. 1, small fields of $h\sim 0.04$ (5–10 T) are sufficient to introduce a robust change in the total magnetization of the sample at low temperatures. From Fig. 3, it can be concluded that this happens mostly through the mutual alignment of already preformed FM areas, whereas the magnetization of a given domain remains comparatively unaffected by those small fields. The susceptibilities should be even stronger for smaller values of x, especially in the insulating regime. A pronounced magnetoresistance effect was indeed observed in lightly doped, insulating InAs and GaAs systems in a similar temperature regime.[11] The situation described here is reminiscent of the colossal effects that can be observed in transition metal oxides. In that context it was conjectured that disorder by chemical doping leads to cluster formation and concomitant large responses to small changes in various parameters.[12]

The possible percolative nature of the magnetic transition in DMS has been discussed recently,[13] and it was proposed that the relevant analog is the problem of randomly placed

spheres that overlap and, thus, form an infinite connected area once the radius of the spheres is larger than the percolation radius r_{per}. In $d=3$, $r_{per}=1.33r_o$, with r_o ($\propto x^{-1/3}$) the interspin distance (and percolation radius) in the periodic case. The MC data ($r_{per}\approx 2-3$ lattice units) agree well with this prediction. In this scenario an increase in x will lead to a smaller r_{per} and, therefore, to an increase in T_C. At some point, however, r_{per} will be smaller than the typical cluster size at T^*, which makes T^* the maximum critical temperature possible. This temperature, although slightly lower, is in the range of the observed T_C^{max} and small changes in the parameters of H_{DMS} might be sufficient to result in higher transition temperatures.

This can be demonstrated by altering the compensation rate. Figure 4 shows m for the same impurity configuration as above, but with $p=0.20$ and $p=0.05$, respectively. As expected, a lowered compensation rate leads to enhanced magnetism. The analysis of the spin correlations (inset Fig. 4) shows that this is mostly due to an increased intercluster spin correlation, whereas the m vs T curve for a given domain (not shown) is largely unchanged from the one at $p=0.1$. Thus, the increased number of charge carriers mainly leads to larger droplets, favoring the creation of a coherent FM state. Additionally, the m vs T curve acquires a more Brillouin-like look.[14] The opposite is true for the case of even stronger compensation, $p=0.05$, where a small reduction in T_C is observed.

In summary, a model for DMS has been studied with MC methods at low temperatures. For the doping rates considered, the system can be described in terms of ferromagnetic droplets. A change in parameters, such as temperature or external fields can lead to the mutual alignment of these domains, while T_C is considerably reduced compared to a mean-field analysis. The HF predictions of a disorder-enhanced transition point are not in agreement with nearly exact MC methods. The predicted T_C is somewhat lower than that measured experimentally, but this value depends considerably on parameters that are not very well known. The inhomogeneous state is expected to show large susceptibilities, e.g., a pronounced magnetoresistance effect.

Discussions with M. Berciu, especially about the implementation of the Hartree-Fock method for Eq. (1), are gratefully acknowledged. Part of this work (E.D.) was supported by NSF Grant No. DMR-0122523.

[1] H. Ohno, Science **281**, 951 (1998); J. Magn. Magn. Mater. **200**, 110 (1999).

[2] E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. **344**, 1 (2001).

[3] T. Dietl, A. Haury, and Y.M. d'Aubigné, Phys. Rev. B **55**, R3347 (1997).

[4] J. Schliemann, J. König, and A.H. MacDonald, Phys. Rev. B **64**, 165201 (2001); J. König, J. Schliemann, T. Jungwirth, and A.H. MacDonald, cond-mat/0111314 (unpublished).

[5] S.J. Potashnik, K.C. Ku, S.H. Chun, J.J. Berry, N. Samarth, and P. Schiffer, Appl. Phys. Lett. **79**, 1495 (2001); T. Hayashi, M. Tanaka, K. Seto, N. Nishinga, and K. Ando, *ibid.* **71**, 398 (1997).

[6] M. Berciu and R.N. Bhatt, Phys. Rev. Lett. **87**, 107203 (2001); some effects of on-site disorder are discussed in M. Berciu and R.N. Bhatt, cond-mat/0111045 (unpublished), and will also be considered in a future publication.

[7] B.I. Shklovskii and A.L. Efros, *Electronic Properties of Doped Semiconductors* (Springer-Verlag, Berlin, 1984).

[8] Hamiltonian (1) might not be applicable for higher doping levels, see C. Timm *et al.*, cond-mat/0111504 (unpublished).

[9] m vs T for a specific domain (see, e.g., Fig. 3) can be fitted very well with a Brillouin function.

[10] See also, M.P. Kennet, M. Berciu, and R.N. Bhatt, cond-mat/0203173 (unpublished).

[11] H. Ohno, M. Munekata, T. Penney, S.v. Molnar, and L.L. Chang, Phys. Rev. Lett. **68**, 2664 (1992); F. Matsukura, H. Ohno, A. Shen, and Y. Sugawara, Phys. Rev. B **57**, R2037 (1998).

[12] J. Burgy, M. Mayr, V. Martin-Mayor, A. Moreo, and E. Dagotto, Phys. Rev. Lett. **87**, 277202 (2001).

[13] A. Kaminski and S. Das Sarma, Phys. Rev. Lett. **88,** 247202 (2002).

[14] Within this family of localized hole models it seems difficult to reproduce the recent annealing improved magnetization results (Ref. 5) that present a rather conventional behavior.