Global versus Local Ferromagnetism in a Model for Diluted Magnetic Semiconductors
Studied with Monte Carlo Techniques

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A model recently introduced for diluted magnetic semiconductors by Berciu and Bhatt (PRL 87, 107205 (2001)) is studied with a Monte Carlo technique, and the results are compared to 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.

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Recent advances in experimental techniques have allowed 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 Ga₁₋ₓMnₓAs, where ferromagnetic (FM) transition temperatures as high as 110 K were obtained. 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. 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. 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. Recent annealing studies have shown the relevance of disorder and defects in DMS. It was proposed, 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 are 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. 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 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

$$H_{DMS} = \sum_{i,j,\alpha}(t_{ij} - \mu\delta_{ij})(c_{i\alpha}^\dagger c_{j\alpha} + \text{H.c.}) + \sum_{i,j,\alpha,\beta}J_{ij}(c_{i\alpha}^\dagger \frac{1}{2}\sigma_{\alpha,\beta} c_{j\beta})S_i,$$

where $c_{i\alpha}^\dagger$ is the creation operator for an electron at the impurity site $i$ with spin $\alpha$, 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, with $a_B$ the Bohr radius associated with the impurity site and Ry its corresponding binding energy $E_b$. $S_i$ represents the (assumed classical) spin at the Mn (3d$^5$) site with $|S_i|=5/2$, $\sigma$ denotes the vector of Pauli matrices, $\mu$ the chemical potential, and $J_{ij}$ describes the non-local 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 $J$ an AF coupling constant. $J$ is chosen as the energy unit and periodic boundary conditions (PBC) 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 $E_{bd}=112.4$meV (1Ry) 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 5.65Å, and $a_B=7.8$Å. 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.

The previous mean-field study of $H_{DMS}$ resulted in some interesting conclusions about the magnetic behavior of DMS. 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. Fortunately, Hamiltonian (1) is especially suited for MC calculations, and the procedure used here has been extensively discussed in the study of the manganites.

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 $x=0.02$, which lies in the neighborhood of the realistic MIT. 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 density, 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, $m=M/N$ ($M=\sum_{i<j}S_i \cdot S_j$), 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. Fig. 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) 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 $|S|$) 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), signalling the development of FM correlations. Furthermore, the concept of a disorder-enhanced $T_C$ does not appear in agreement with MC. Fig.3 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, $\chi \propto (M^2) - \langle M \rangle^2$, a critical behavior around $T_{c}^{MF} \sim 0.25 \pm 0.05$ can be deduced for the periodic system, which is slightly below the critical region for the disordered case. This appears in agreement with previous HF results, but the reduction in $T_C$ is much less pronounced. It will be argued below, however, that the actual transition temperature is lower than $T_{c}^{MF}$. 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 $(n)_{cl} > p$. This leads to an increased temperature where FM corre-
lations 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 \( x \leq 0.005 \)\( ^{11} \). The conclusions above are also supported by analyzing the spin correlations between different clusters A, B (Fig. 3).

Additionally, \( M/|S| \) is proportional to the number of spins below this temperature as it should be for a FM ordered state (Fig. 3(b)). In any case, these values are lower than \( T_{C}^{\text{per}} \) and they suggest two different temperature scales in the problem: \( T^* \) (\( \sim 0.3 \)), where isolated magnetic domains are formed, and a smaller true critical temperature \( T_{C} \) (\( \sim 0.1 \)) 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\( ^{11} \). This idea is also qualitatively depicted in Fig. 3(c). In this scenario disorder enhances \( T^* \), but reduces \( T_{C} \).

This reveals that each cluster turns FM as the temperature is reduced, 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 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 \( \sim 0.08 \) (10K). 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.

Fig. 2. (a) 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 \( |S| \)) 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 \( T_{C} \).

These percolation-like effects are also visible once a magnetic field is applied. As shown in the inset of Fig. 3, small fields of \( h \sim 0.04 \) (5-10T) 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.\( ^{12} \) 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.\textsuperscript{13}

The possible percolative nature of the magnetic transition in DMS has been discussed recently,\textsuperscript{8} and it was proposed that the relevant analog is the problem of randomly placed spheres which overlap and, thus, form an infinite connected area once the radius of the spheres is larger than the percolation radius \(r_{\text{per}}\). In \(d=3\), \(r_{\text{per}}=1.33r_o\), with \(r_o\) (\(\propto x^{-1/3}\)) the interspin distance (and percolation radius) in the periodic case. The MC data \((r_{\text{per}}=2-3\text{ lattice units})\) agree well with this prediction. In this scenario an increase in \(x\) will lead to a smaller \(r_{\text{per}}\) and, therefore, to an increase in \(T_C\). At some point, however, \(r_{\text{per}}\) 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 \(T_C^{\text{max}}\) and small changes in the parameters of \(H_{\text{DMS}}\) might be sufficient to result in higher transition temperatures.

This can be demonstrated by altering the compensation rate. Fig.\textsuperscript{\textcopyright} 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 \(\textsuperscript{\textcopyright}\)) 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.\textsuperscript{\textsuperscript{\textcopyright}}

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 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.

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