

Effects of randomness on ferromagnetism in a Ga_{1-x}Mn_xAs diluted magnetic semiconductor

Mark O'Brien¹ and Kevin Ingersent²

¹*Department of Physics, Lycoming College, Williamsport, PA 17701*

²*Department of Physics, University of Florida, Gainesville, FL 32611*

Results are presented in order to gain insight into magnetization curve effects of various *Mn* placement schemes in Ga_{1-x}Mn_xAs. This is done in order to understand the nature of the reported increase of critical temperature that accompanies the increase in *Mn* placement disorder. In the following document, it is shown that a threefold increase of this temperature arises when heavy clumps of *Mn* atoms are placed in an existing GaAs lattice. Results presented below are based on a mean field approach.

I. INTRODUCTION

As the name implies, diluted magnetic semiconductors (DMS) are doped semiconductors that have the possibility of becoming ferromagnetic at sufficiently low temperatures. DMS have the general form A_{1-x}M_xB where AB is either a II-VI or a III-V semiconductor and M a magnetic element. Interest in such materials began in 1989 when molecular beam epitaxy techniques (MBE) were used to introduce quantities of the magnetic material in excess of the usual saturation limit which lead to the existence of ferromagnetism [1]. When the particle represented by A is replaced by a particle represented by M in a DMS, a charge carrier is created based on the valence difference between A and M. This charge carrier interacts antiferromagnetically (AFM) with a nearby M particle which causes the ferromagnetic alignment of the M spins to take place.

In particular, Ga_{1-x}Mn_xAs has received much of the attention due to the observation of ferromagnetic behavior occurring with doping concentrations (x) of 0.053 and Curie temperatures of the order of 110K [2,3]. This critical temperature is significantly higher than that of other III-V DMS[4].

In addition to the obvious effects of charge carrier density, applied magnetic field, and *Mn* concentration (x), disorder has been shown to play a large part in the shape of magnetization curve as well as the value of the Curie temperature[5, 6]. It is believed that the increase in critical temperature is due to pockets of dense clusters of *Mn* that are not present in ordered cases [5, 6]. This increase is attributed to charge carriers preferentially gathering around the clusters which in turn causes spin polarization to occur at higher temperatures.

In this document, we consider the effects of various degrees of clustering in order to gain more insight to the cause of critical temperature increase. We investigate a mean field approach to a Ga_{1-x}Mn_xAs structure in its insulator state in which charge carriers move only in the impurity band while the underlying GaAs only provides structure. Several degrees of

clustering will be discussed and results comparing average impurity spin as well as average charge carrier spin as a function of temperature will be given.¹

II. THEORY

The Hamiltonian used has two major parts which correspond to the two major properties of DMS, spin and charge movement. The first part has the form of a tight binding model which describes the charge carrier movement:

$$\mathcal{H}_{cm} = \sum_{i,j} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} ,$$

where the $c_{i\sigma}^{\dagger}$ is the creation operator (the creation operator can be viewed as an operator responsible for placing a hole at site *i* and with spin σ), t_{ij} is the hopping matrix (reflects the probability of a hole at *Mn* site *i* moving to a *Mn* at site *j*). We take t_{ij} as

$$t_{ij} = 2(1 + r_{ij}) \exp\left(\frac{-r_{ij}}{a_b}\right) \text{Ry} ,$$

where Ry is the Rydberg for the hole, *Mn* system.

The second term is a more complicated and describes the spin interactions of a *Mn* atom at site *i* with a charge carrier at site *j*:

$$\mathcal{H}_{spin} = \sum_{i,j} J_{ij} \vec{S}(i) \cdot \vec{s}(j) ,$$

where J_{ij} is the AFM exchange integral which we take as:

$$J_{ij} = J \exp\left(\frac{-2r_{ij}}{a_b}\right)^2 .$$

¹ While a complete model should include both impurity and valence band states, it is believed that this model is a good approximation at low temperatures due to low Fermi temperatures (of the order of a few tens of meV smaller than that of the gap between impurity and valence band)[6].

² Our hopping and overlap coefficients have been taken to be of the form of a hydrogenic orbital. See [6] for models with different hopping coefficients and overlap integrals.

Other desired terms can be added to the Hamiltonian such as a term which describes an external magnetic field or onsite disorder[6].

Even though the present form of the Hamiltonian only has two terms, it would require a basis that would have a dimension of 24^{N_d} where N_d is the number of Mn sites. This is why a mean-field approach must be used. This entails using the following factorization for the spin interaction term:

$$\vec{S}(i) \cdot \vec{s}(j) \rightarrow \left\langle S^z(i) \right\rangle s^z(j) + S^z(i) \left\langle s^z(j) \right\rangle - \left\langle S^z(i) \right\rangle \left\langle s^z(j) \right\rangle,$$

where the implicit assumption has been made that symmetry is only broken in the z-direction [6].

With this factorization, we now have four total terms to our Hamiltonian:

$$\mathcal{H} = \sum_{i,j} t_{ij} c_{i\sigma}^+ c_{j\sigma} + \sum_{i,j} J_{ij} \left(\left\langle S^z(i) \right\rangle s^z(j) + S^z(i) \left\langle s^z(j) \right\rangle - \left\langle S^z(i) \right\rangle \left\langle s^z(j) \right\rangle \right) - \text{diagonalize (the)}$$

For the calculations needed, we only used the first and second of these terms. We first write the $s^z(j)$ term as $\frac{\sigma}{2} c_{j\sigma}^+ c_{j\sigma}$. This is due to the assumption that charge carriers are only in a spin +1/2 or a spin -1/2 state, and that $c_{j\sigma}^+ c_{j\sigma}$ corresponds to the total number of holes at site j (the so called number operator). Substitution of the above quantity into Eq. () gives us the following two terms:

$$\mathcal{H}_{CC} = \sum_{i,j} t_{ij} c_{i\sigma}^* c_{j\sigma} + \frac{\sigma}{2} \sum_i \left(\sum_j J_{ij} \left\langle S^z(i) \right\rangle \right) c_{i\sigma}^* c_{i\sigma}.$$

This Hamiltonian, which describes charge carrier movement, can be diagonalized when $\left\langle S^z(i) \right\rangle$ is given to obtain the single hole eigenvectors $\psi_{n\sigma}$ and their energy eigenvalues $E_{n\sigma}$. The occupation probability of the state (n, σ) at temperature $T = 1/k_B\beta$ and chemical potential μ is given by the Fermi distribution:

$$f(E_{n\sigma}) = (\exp(\beta(E_{n\sigma} - \mu)) + 1)^{-1}.$$

The chemical potential must be chosen in order to satisfy the condition

$$N_h = \sum_{n\sigma} f(E_{n\sigma})$$

where N_h is the (predetermined) number of holes in the system.

Now with the Fermi distribution and state vectors for the charge carriers at hand, it is straight forward to calculate the expectation value of the hole spin at site i :

$$\left\langle s^z(j) \right\rangle = \frac{\sigma}{2} \sum_{n,\sigma} |\psi_{n,\sigma}|^2 f(E_{n,\sigma}).$$

Finally, the $\left\langle s^z(j) \right\rangle$ determine the expectation value of the Mn spin at each site to be $\left\langle S^z(j) \right\rangle = B_S(x) = (S + 1/2) \coth[(S + 1/2)x] - 1/2 \coth(x/2)$

where $x = \beta \sum_j J_{ij} \left\langle s^z(j) \right\rangle$ and $S=5/2$ is the size of the Mn spin, and $B_S(x)$ is the Brillouin function.

III. TECHNIQUES

We solve all of the above equations using an iterative algorithm. Initially, a guess as to what the Mn 's spin expectation value is made. From there, we diagonalize the matrix to obtain state vectors and energy eigenvalues. Then, we use the Fermi distribution, and the expectation value of the charge carriers spin. Finally, we used the Brillouin function to check our initial guess for the Mn spins. We continue to recalculate values for the Mn spins until self-consistency is reached (self consistency was taken when $\left\langle s^z(j) \right\rangle$ at successive iterations changed by no more than 6×10^{-5} at any site).

IV. RESULTS

For all results obtained, we used a values of 112.4meV for the Rydberg, $J=15\text{meV}$, lattice spacing a of 5.65 Å, and bohr radius $a_B = 5.65$ Å [7]. Furthermore, the lattice was set up as a 24x24x24 fcc lattice with 512 Mn particles ($x=0.00926$), and a 10 to 1 Mn to hole ratio.

A. Cubic lattice

The main focus of this project was to examine the cause of the reported effect of disorder. Initially, an evenly spaced cubic lattice of Mn sites was set up in order to check our results against those obtained by Berciu *et al.* In the cubic case it can be shown that in the ordered cubic lattice that the solutions to the diagonalization process are simple plane waves[5]. This lead to site independent values of charge carrier and Mn spin. The result of which allows the average value of the Mn particles to have their maximum value of spin (2.5) for some of the lower temperatures considered. Although the curves started with higher values, the critical temperature of such particle alignments was shown to be significantly less than that of the other cases considered. The arrangement of particles can be seen in fig. 1, and the spin-spin curves

can be seen in Fig. 4. These results agree with the results obtained by Berciu et. al [5,6].

B. Completely random *Mn* placement

The other case studied that was also considered by Berciu *et al.* was that of a completely random arrangement of *Mn* sites in the GaAs lattice (Fig 2). Although in this case the expectation value for the *Mn* was initially lower, the critical temperature of the system was substantially higher. This is due to migration of holes to sites where the *Mn* density is higher. This causes several *Mn* atoms to be left with a low probability of having a hole available to polarize the site, hence bringing down the averaged *Mn* over all sites down at lower temperatures. However, at higher temperatures, holes located on the clustered area maintain the *Mn* spin polarization.

C. Clustered *Mn* placement

After the random location disorder cases were considered, situations were considered which allowed *Mn* particles to cluster (Fig 3). This started by assigning a set number of clusters a random number of particles. Then, the first particle in each cluster was placed in a random location. From there, other particles were placed around the initial particle in the cluster with a biased weighting factor:

$$w(x) = \coth(i) - 1/2 \coth(i/2) + 1/2 .$$

The weighting factor induced a biased in the following way: $w(0) = 1/2$ of particles were placed on a site $1/2a$ in any direction, $w(1)-w(0)=.231$ of the particles were on a site a away in any direction, $w(2)-w(1)=.150$ were on a site $3/2a$ away in any direction. In general, $w(i)-w(i-1)$ of the total particles were at a distance $(i+1)/2$ away in any direction.

We have been able to show that the critical temperature was highly dependent on the amount of clustering aloud (see Fig 4). The figure points out that as the number of particles in a given cluster increases, the value of the Curie temperature also increases. This is in agreement with the results predicted by Berciu *et al.* Also, Fig 5 depicts to the migration of holes to the largest cluster in a typical five cluster *Mn* distribution. Lastly, Fig 6 shows the details of several runs of a 50 cluster system (10.24 *Mn* atoms per cluster).

V. CONCLUSIONS

In the above document we hope to have given some insight into the nature of the surprising increase in Curie temperatures given by theoretical models presented by Berciu *et al.*[5,6] of $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ with $x = 0.00926$ and 10 to 1 *Mn* to hole ratio. We were able to do this by showing the even further increase in Curie temperatures above that which were obtained for a completely random situation. Furthermore, we were able to confirm nearly complete hole migration to the heavily clustered regions leaving smaller clusters unpolarized, but providing a means for larger clusters to stay polarized for higher temperatures. Future research on the subject could include comparisons of a uniform high density situation to those presented here. Also, a model that included potential interactions among the holes that are usually ignored due to low hole density may effect a system where holes migrate to heavily clustered regions may effect the overall spin polarization curves.

VI. ACKNOWLEDGEMENTS

I would like to thank the national science foundation for the opportunity to conduct this research. I would also like to thank the University of Florida for giving me a home for the summer. Furthermore, I would like to thank Drs. Kim McCall and Dianne Cothran for their assistance this summer. Lastly, I would like to thank Professor Alan Dorsey and Taylor for answering questions when Professor Ingersent was not around to assist me.

VII. REFERENCES

- [1] H. Ohno, Journal of Mag. and Mag. materials 200 (1999) 110-129 (1999).
- [2] F. Matsukura, H. Ohno, A. Shen and Y. Sugawara, Phys. Rev. B **57**, R2037 (1998).
- [3] H. Ohno and F. Matsukura, Solid State Commun. **117**, 179 (2001)
- [4] A. Hauray *et al.*, Phys. Rev. B **56** 13 103 (1997)
- [5] Mona Berciu and R.N. Bhatt, Phys. Rev. **87**, 107203 (2001)
- [6] Mona Berciu and R.N. Bhatt, cond-mat/0009161
- [7] A.K. Bhattacharjee and C. Benoit a la Guillaume, Solid State Commun. **113**, 17 (2000).

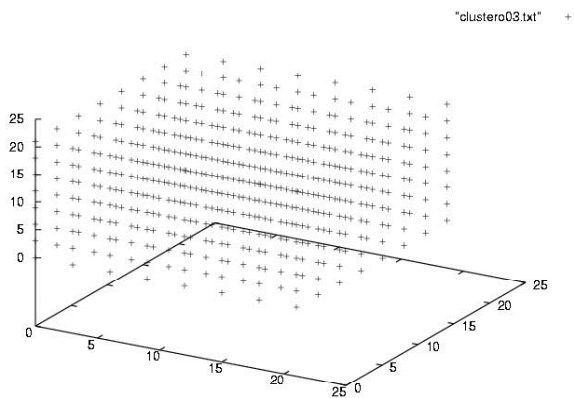


Figure 1. Cubic Mn distribution

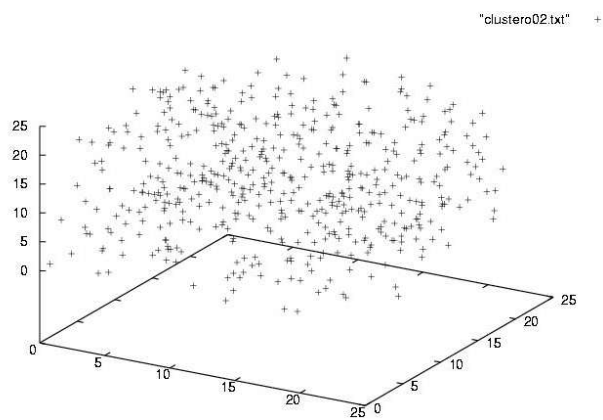


Figure 2. Random Mn distribution

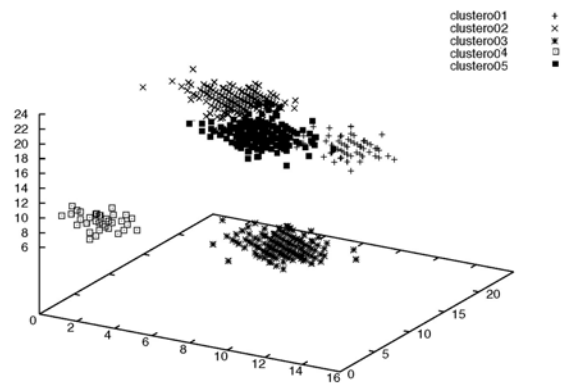


Figure 3. Caption of a typical five cluster Mn site location.

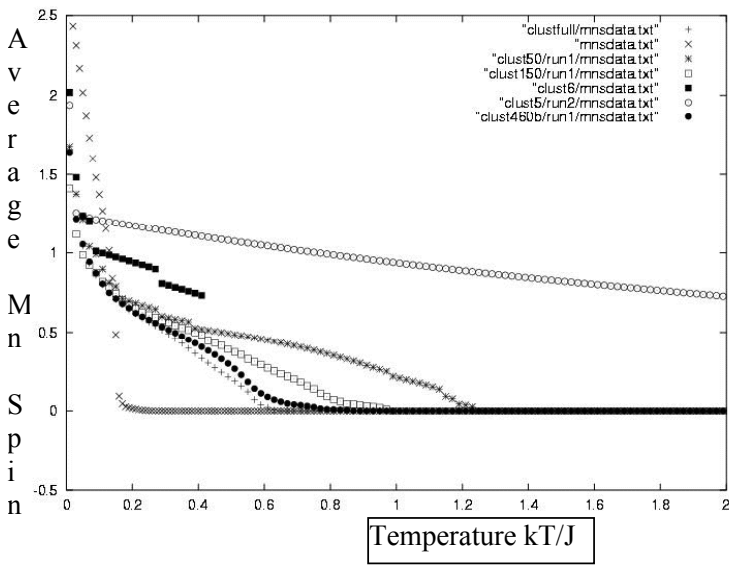


Figure 4. Comparison of the average Mn spin of the entire lattice as a function of temperature. Plots are given for various degrees of clustering.

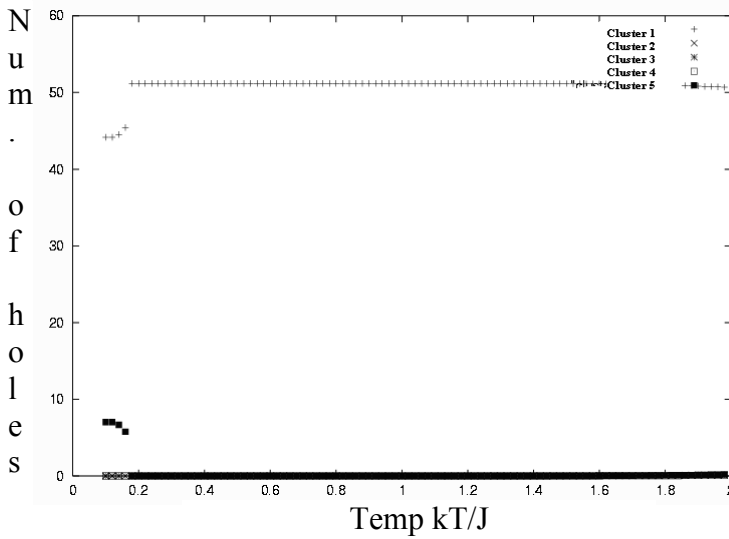


Figure 5. Number of holes at a given temperature for a five cluster system.

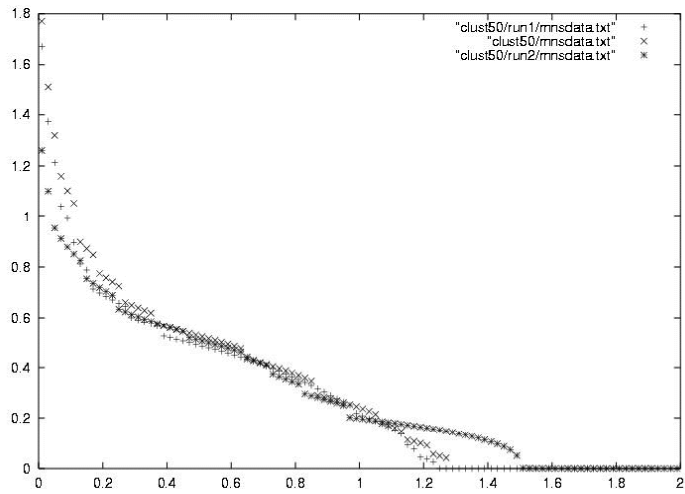


Figure 6. Comparison of various 50 cluster runs.