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Ab-initio study of transition temperature of Mn-doped GaN diluted magnetic semiconductor

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We study the effect of clustering and random metal atoms distribution on the transition temperature of Mn-doped GaN Diluted Magnetic Semiconductors. To calculate the exchange coefficients between the Mn atoms, a Density functional theory-based Locally Self Consistent Multiple Scattering (LSMS) method has been used. Here we present the results of our calculations, which show that random metal distribution increases the calculated transition temperature of the material while the clustering decreases it.

1 Introduction

A new type of material called Diluted Magnetic Semiconductors (DMS) has many potential technological applications. One of these applications is to design semiconductor spin electronics ("spintronics") devices [1]. Ever since Ohno et. al. [2] doped Mn ions into GaAs, and found that (Ga_{1-x}, Mn_x)As DMS has the transition temperature (T_C) above 100 K, the theoretical and experimental research on DMS, and the origin of its ferromagnetism, is continuing. For practical applications, however, DMS materials with transition temperatures well above room temperature are required. By varying its growth and annealing conditions, a system of Mn-doped GaN with transition temperatures in the range of 228 - 370 K has been achieved [3]. The reasons for the wide range of the reported (theoretical and experimental) transition temperatures, however, are not well understood.

Although the theoretical study of DMS has been done for a number of years, it is fair to say that, at the present time, there is no systematic way to predict the transition temperature of a DMS material. To calculate the transition temperature of a DMS system, the three most commonly used techniques are: (i) The Mean Field Approximation, (ii) The Random Phase Approximation, and (iii) The Monte Carlo (MC) simulation. All of these techniques rely on an accurate determination of the exchange coefficients J_{ij} between the atoms.

Recent studies on the clustering effect (or phase separation) of transition metal-doped DMS suggested that the existence of metal clusters could affect the transition temperature of a DMS system. The studies, however, have provided mixed results. Refs. [4]-[5] reported that the existence of metal clusters could increase the transition temperature of Mn-doped GaN. On the other hand, in a recent paper, Sandratskii et. al. [6] reported that Mn clustering decreases the calculated transition temperature of Mn-doped GaN. The decrease in transition temperature, according to Sandratskii et. al., is due to the splitting of the impurity band in the Mn intracluster.

In this paper we report the results of an ab-initio study of the effect of random Mn atoms distribution on the transition temperature of Mn-doped GaN, a widely studied DMS material. The effect of Mn clustering on transition temperature is also reported in this paper. Two steps were used in the present paper. In the first step, we calculated the exchange coefficients between the Mn atoms in the system. Once these coefficients have been determined, the transition temperature was estimated using the Mean Field Approximation (MFA).

2 Computational Method

We used the Density Functional Theory (DFT)-based Locally Self Consistent Multiple Scattering (LSMS) method [7] to calculate the exchange coefficient J_{ij} between metal (Mn) atoms in a supercell. LSMS method is an all-electron, real space, multiple scattering theory approach to calculate the electronic and magnetic properties of periodic systems.

A supercell of 64 Zinc-Blende GaN atoms has been used in our calculations. The lattice constant is 4.5 Angstrom. We only consider substitutional impurities, where one or more Ga atoms in a supercell of the semiconductor crystal are replaced by Mn atoms.

Once the exchange coefficients were determined, the transition temperature, T_C, can be calculated using the MFA formula: $T_C^{MFA} = (2n/3k_B)\Sigma_j J_{0j}$ where k_B is the Boltzmann constant, n is the number of Mn clusters in the system, and J_{0j} is the exchange coefficient of Mn atoms. In this paper we consider $n = 1 - 4$. In order to investigate the effect of Mn clustering, we also calculated the transition temperature for both the Mn intercluster and Mn intracluster. In our definition, Mn intercluster

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Table 1 Calculated transition temperatures (in Kelvin) of Mn-doped GaN for different number of clusters n and concentrations x.

Number of clusters (n)	Concentration (x)	Calculated T_C^{MFA} (in K)
1	0.0625	17.89
2	0.0625	103.16
3	0.09375	164.21
4	0.125	218.95

means that the Mn atoms are not connected through a common N atom, while Mn intracluster means that the Mn atoms are connected through a common N atom.

3 Results

In Table 1 we list the calculated MFA transition temperatures of (Mn_x,Ga_{1-x})N for $x = 0.0625, 0.09375$, and 0.125. From this table we see that the transition temperature increases as the concentration x increases, reaching $T_C = 218.95$ K at $x = 0.125$. Similar trend of increased transition temperature with respect to the increased concentration has also been reported, e.g., in Monte Carlo simulation of (Ga,Mn)N [8]. In Ref. [8], however, the calculated transition temperature slightly decreases when the concentration is increased from 0.10 to 0.15.

For all values of x, however, our calculations predicted higher transition temperatures than those estimated by MC simulation [8]. Using the combination of Coherent Potential Approximation and MFA in a supercell, Ref. [9] reported the transition temperature of 176 K at $x = 0.0625$, higher than our value of 103.16 K.

The difference between MFA and MC simulation is due to the simplification in the evaluation of T_C^{MFA}, which only takes into account the sum of exchange coefficients $(\Sigma_j J_{0j})$. One other reason, as we found out in the course of our calculations, is because the Mn atoms exchange interaction in Mn-doped GaN is short-ranged. The short-ranged interaction also was not taken into account in calculating T_C^{MFA}.

Table 1 also shows that the clustering of Mn atoms has the effect of decreasing the transition temperature. This effect can be seen from the values of T_C given in the first and second rows in Table 1. The case of $n = 1$ corresponds to two Mn atoms sharing a common N atom (thus, performing an Mn intracluster), while the case of $n = 2$ corresponds to two separated Mn atoms. Detail examinations of the individual exchange coefficients of the Mn atoms reveal that the antiferromagnetic coupling between the neighboring Mn atoms is the cause of the smallness of T_C. At the present calculation it is not possible to see the effect of clustering by increasing the Mn concentration as the effect of the nearby supercells cannot be neglected. Calculations using larger number of atoms (256 and 512 atoms) in the supercell are underway and will be reported elsewhere.

4 Conclusion

The transition temperatures of Mn-doped GaN DMS with different concentrations have been calculated with the help of LSMS method. Our calculations show that the transition temperature increases as the concentration is increased. According to our calculations, the random Mn atoms distribution increases the calculated transition temperature of the material while the existence of Mn cluster decreases the transition temperature.

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