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Curie temperatures of cubic (Ga, Mn)N diluted magnetic semiconductors from the RKKY spin model

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Abstract
We explore how much the RKKY spin interaction can contribute to the high-temperature ferromagnetism in cubic (Ga, Mn)N diluted magnetic semiconductors. The usual coupling constant is used and effective carriers are considered independent of doped magnetic atoms, as is shown experimentally. Our Monte Carlo simulated results show that maximal Curie temperature is reached at the optimal carrier concentration for a given Mn concentration, equaling 373 K for 5% Mn and 703 K for 8% Mn. Because such a Monte Carlo method does not overestimate transition temperatures, these calculations indicate that the RKKY spin interaction alone can yield high-enough Curie temperatures in cubic (Ga, Mn)N under optimized conditions.

1. Introduction

Diluted magnetic semiconductors (DMSs) are promising materials for fabricating practical spintronic devices using both the spin and charge of an electron. High Curie temperatures (T_c) above room temperature have been realized in such materials, especially those based on III–V semiconductors. So far, the highest Curie temperature, 173 K, is reached in the case of 8% Mn-doped GaAs [1]. Because the band gap of GaN is wider than that of GaAs, GaN-based DMSs have great potential for room temperature spintronic materials, and have been studied intensively [2–4]. For cubic (Ga, Mn)N, ferromagnetism at room temperature has been reported [5] and local structures around substitutional Mn atoms have been determined by x-ray absorption spectroscopy [6]. The ferromagnetism of (Ga, Mn)As has been widely explored [7, 8], and now it can be explained mainly by a carrier-induced RKKY spin interaction [9, 10]. In contrast, the ferromagnetism of the (Ga, Mn)N is still controversial. The Zener model in mean field treatment was used to describe the ferromagnetism in zinc-blende (ZB) magnetic semiconductors and thus to predict that 5% Mn-doped ZB GaN has T_c higher than room temperature [11], but it was recently shown to overestimate Curie temperatures [12, 13]. On one hand, magnetic percolation was considered crucial for the ferromagnetism in (Ga, Mn)N [13], and on the other hand the transition temperature of the (Ga, Mn)N was studied with Monte Carlo (MC) simulation using exchange integrals determined from first-principles calculations, and the result was used to claim that high T_c was impossible in the (Ga, Mn)N [14]. However, achieving a high T_c is dependent on many factors during the synthesis of cubic (Ga, Mn)N samples. Actually, the T_c varies from 10 to 900 K in different samples [15, 16]. Recently, good progress has been made for partly elucidating these in the case of doped ZnO [17]. Therefore, it is highly desirable to further study the high-temperature ferromagnetism observed in (Ga, Mn)N samples.

Here we explore how much the RKKY spin interaction can contribute to the high-temperature ferromagnetism of cubic (Ga, Mn)N diluted magnetic semiconductors. As is shown experimentally, the effective carrier concentration ρ_c is supposed to be independent of magnetic atomic concentration ρ_m, the spin S is set to $5/2$ [18–21], and the usual coupling constant is used [9]. Our Monte Carlo simulations show

that for a given ρ_m, the maximal Curie temperature T_{cp} is achieved at an optimal carrier concentration ρ_{cp}, and T_{cp} is proportional to ρ_m, reaching 373 K at $\rho_m = 5\%$ and even 703 K at $\rho_m = 8\%$. Because the Monte Carlo method does not overestimate phase transition temperatures, these results imply that the RKKY effective spin interaction can play an important role in the ferromagnetism and high T_c can be achieved by realizing high ρ_m and optimizing ρ_c.

The remaining part of this paper is organized as follows. In section 2 we describe our model and simulation method. In section 3 we shall present our main Monte Carlo simulated results. In section 4 we shall make some discussions toward comparison with experiments concerned and give our conclusion.

2. Model and method

Generally speaking, effective carriers (holes or electrons) can be introduced in a semiconductor by shallow acceptors or donors. For ZB GaN, doped Mn d states are deep in the wide semiconductor gap of 3.2 eV [5, 22] so that the doped Mn impurities themselves cannot introduce effective carriers [23]. In this case, the effective carriers can be introduced by N vacancies [24], hydrogenation [25], co-doping [26, 27], or induced by light or electric field [28, 29]. Therefore, ρ_c can be considered independent of ρ_m. The effective RKKY spin model is defined by the Hamiltonian [30]

$$H = \tfrac{1}{2}\sum_{\alpha\beta} J_{\alpha\beta}\vec{S}_\alpha \cdot \vec{S}_\beta, \tag{1}$$

where Greek letters such as α indicate magnetic atomic sites, and $\vec{S}_\alpha$ is the quantum spin operator on the site α. The RKKY spin exchange constant is defined as

$$J_{\alpha\beta} = -\frac{3J_{pd}^2\rho_c k_F^3}{8\pi E_F}F(2k_F r_{\alpha\beta}), \tag{2}$$

where J_{pd} is the p–d exchange integral between the carrier and the magnetic atom, E_F the Fermi energy, and k_F the Fermi wavenumber. It can be proved that $k_F = (3\pi^2\rho_c)^{1/3}$ and $E_F = \hbar^2 k_F^2/(2m^*)$, where $\hbar$ is Planck's constant divided by 2π and m^* the effective carrier mass. $r_{\alpha\beta}$ is the distance between α and β, and the function $F(x)$ is defined as $F(x) = (\sin x - x\cos x)/x^4$.

MC simulations are performed with the Metropolis algorithm [31]. The size of the lattice is a $20 \times 20 \times 20$ face-center-cubic unit cell with lattice constant a, and each of unit cell consists of four sites. Mn atoms are distributed randomly among the lattice sites and ten magnetic atomic configurations are considered for averaging. Usually, the finite size effect makes it difficult to accurately determine T_c from the temperature dependence of the average magnetic moment $M(T)$. For our simulations on the large lattice, the peak of magnetic susceptibility $\chi(T)$ is used to determine the phase transition temperature T_c with enough accuracy. The infinite length of the RKKY exchange interaction is cut off and the largest $r_{\alpha\beta}$ is determined by the first node of the oscillating function $F(2k_F r_{\alpha\beta})$, because it does not change our main results to take into account the remainder of the RKKY interaction beyond the first node. For each temperature, 100 000 MC steps are calculated, the first 50 000 steps are discarded and the averaging is done every fifth step of the remaining. The accuracy is confirmed by further MC simulations of 200 000 MC steps for temperatures near T_c (20–30 K lower and higher than T_c).

3. Main simulated results

The necessary parameters are J_{pd}, S, a, and m^*. We use $J_{pd} = 150$ eV Å^3 that was obtained from magneto-transport measurements [9] and is taken for all the Mn-doped III–V magnetic semiconductors [11]. Because doped Mn d states are deeply in the wide semiconductor gap of cubic (Ga, Mn)N, effective carriers cannot be introduced by Mn dopants alone. However, there are always impurities such as N or Ga defects in (Ga, Mn)N samples, and the N defects always release electrons in (Ga, Mn)N and change the Mn d^4 state to a half-filled Mn d^5 state [21]. The divalent state $Mn^{2+}(d^5)$ is confirmed by Mn $L_{2,3}$ x-ray absorption from $Ga_{1-x}Mn_xN$ samples grown by molecular beam epitaxy [18, 20]. Therefore, we use $S = 5/2$, $a = 4.45$ Å [23], and $m^* = 1.38m_0$ (an average value of those in the three directions: [100], [110], and [111]) [32].

In figure 1 we present temperature dependences of both $M(T)$ and $\chi(T)$ of Mn-doped ZB GaN in the case of $\rho_m = 5\%$ and $\rho_c = 1.5 \times 10^{20}$ cm^{-3}. The $\chi(T)$ is calculated for ten configurations of Mn distributions in the lattice of ZB GaN. We determine the transition temperature, $T_c = 373$ K (± 5 K), in terms of the peak of $\chi(T)$. For $\rho_m = 5\%$, we present T_c as a function of ρ_c between 0.3×10^{20} and 2.5×10^{20} cm^{-3} in figure 2. T_c at first increases with increasing ρ_c, reaches the maximal value at $\rho_c = 1.5 \times 10^{20}$ cm^{-3}, and then decreases with increasing ρ_c. As a result, there is a maximal value T_{cp} for the transition temperatures and it corresponds to an optimal carrier concentration ρ_{cp}. For $\rho_m = 5\%$, we obtain the maximal Curie temperature $T_{cp} = 373$ K at the optimal carrier concentration $\rho_{cp} = 1.5 \times 10^{20}$ cm^{-3}.

It is highly desirable to determine how high the Curie temperature can be for a given ρ_m in the cubic (Ga, Mn)N. For this purpose, we perform MC simulations under four Mn concentrations ($\rho_m = 1\%, 3\%, 5\%$, and 8%). Figure 3 shows the maximal Curie temperatures T_{cp} and corresponding optimal carrier concentrations ρ_{cp} as functions of the Mn concentration ρ_m. It is obvious that ρ_{cp} increases with ρ_m and T_{cp} is proportional to ρ_m. T_{cp} reaches 703 K when the Mn concentration is $\rho_m = 8\%$. To confirm the accuracy of our simulations, we have done similar calculations for 5.3% Mn-doped GaAs. Taking reasonable experimental parameters [9, 33], we obtain a maximal transition temperature 106 K, which agrees well with the experimental 110 K [9]. Therefore, it is reasonable to conclude that very high Curie temperature can be achieved for Mn-doped ZB GaN by realizing high Mn concentration and optimizing effective carrier concentration.

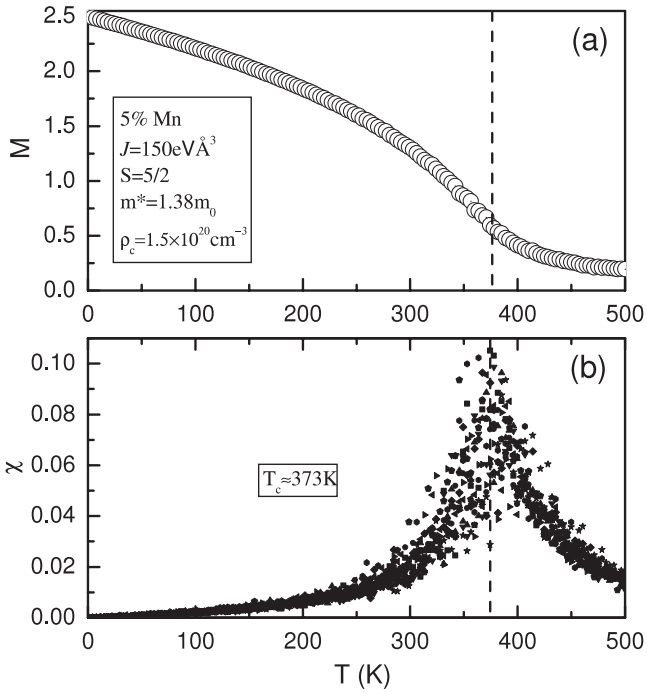

Figure 1. The temperature dependences of spontaneous magnetization $M(T)$ (a) and magnetic susceptibility $\chi(T)$ (b) of Mn-doped ZB GaN for $\rho_m = 5\%$ and $\rho_c = 1.5 \times 10^{20}$ cm^{-3}. The Curie temperature is determined by the peak of $\chi(T)$. Ten different symbols such as stars and triangles are used to present the $\chi(T)$ for the ten Mn distributions.

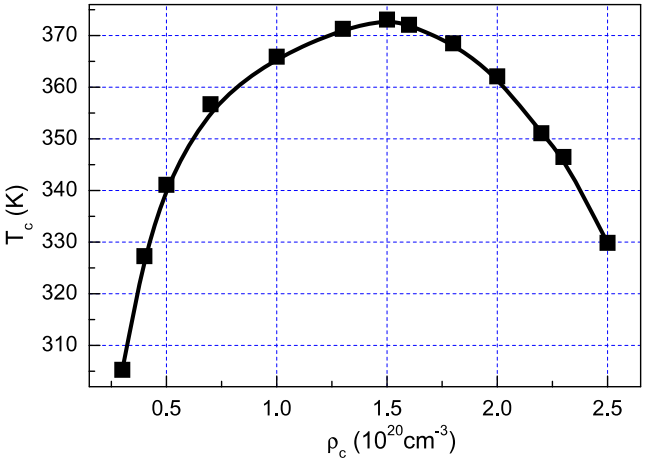

Figure 2. The dependence of the Curie temperature T_c on carrier concentration ρ_c for $\rho_m = 5\%$. The maximal transition temperature $T_{cp} = 373$ K corresponds to the optimal carrier concentration $\rho_{cp} = 1.5 \times 10^{20}$ cm^{-3}.

(This figure is in colour only in the electronic version)

4. Discussions and conclusion

It must be pointed out that many factors in real samples and during their fabrication must be optimized to achieve the maximal Curie temperature for a given ρ_m. Usually, the trivalent state of the Mn ion, Mn^{3+} (d^4, $S = 2$), in (Ga, Mn)N coexists with Mn^{2+} (d^5, $S = 5/2$) [18–21, 34].

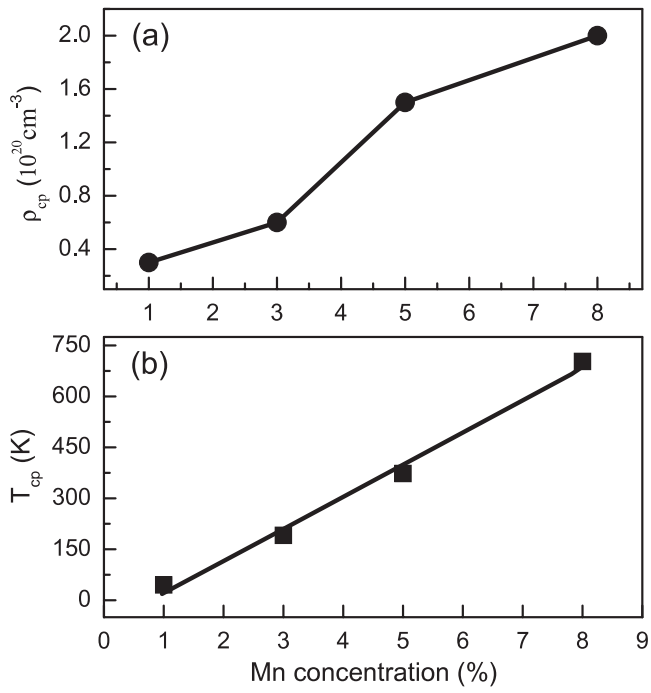

Figure 3. The dependence of the optimal carrier concentration ρ_{cp} (a) and the corresponding maximal Curie temperature T_{cp} (b) on the Mn concentration ρ_m.

Actually, Mn^{2+} ($S = 5/2$) is necessary for high-temperature ferromagnetism [35]. Fortunately, Mn^{2+} can be enhanced, even made dominant, by introducing additional electrons through N vacancies, hydrogenation, and appropriate co-doping [18, 20, 21, 24, 25]. As for effective carriers, both p-type [25, 5, 16, 19, 36] and n-type [15, 24] have been achieved in real samples. Appropriate n-type carriers are useful to the high-temperature ferromagnetism, but too many can fill the empty Mn d states in the gap and thus destroy the ferromagnetism [24]. High concentrations ($\sim 10^{20}$ cm^{-3}) of p-type carriers are useful for enhancing T_c [17, 19, 25] through achieving the optimal concentrations ρ_{cp}. However, the detail of the multiple valence bands is involved in determining the high-temperature ferromagnetism, and the spin–hole coupling can reduce the hole mobility and enhance the hole effective mass in the top of the valence bands [37]. At this stage, we have used a simplified approach in treating the hole carriers. More details should be considered in the future. In some real cases, average effective carrier concentrations are quite low, but their T_c values still reach room temperatures [5, 36]. This phenomenon should be attributed to effective carrier inhomogeneity in the samples. The optimal concentrations are actually achieved in parts of the samples. With all these optimized, high T_c can be obtained by realizing high ρ_m.

In summary, we find out the role the RKKY spin interaction can play in the high-temperature ferromagnetism in cubic (Ga, Mn)N as a diluted magnetic semiconductor. The usual coupling constant is used and effective carriers are considered independent of doped magnetic atoms, as is shown experimentally. Our Monte Carlo simulated results show that maximal Curie temperature is reached at the optimal carrier

concentration for a given Mn concentration, equaling 373 K for 5% Mn and 703 K for 8% Mn. Because such a Monte Carlo method does not overestimate transition temperatures, our results indicate that the RKKY spin interaction alone can yield high-enough Curie temperatures in cubic (Ga, Mn)N under optimized conditions. Besides, this should be of much interest for working out the complete mechanism of the ferromagnetism in (Ga, Mn)N.

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