第 25 卷第 12 期　　半 导 体 学 报　　Vol. 25, No. 12
2004 年 12 月　　CHINESE JOURNAL OF SEMICONDUCTORS　　Dec., 2004

Magnetism and Stability of Diluted Magnetic Semiconductor $(Ga_{1-x}Fe_x)As$*

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Abstract: Magnetism and the stability of $(Ga_{1-x}Fe_x)As$ are investigated using the first principles LMTO-ASA band calculation by assuming supercell structures. Four concentrations of the 3d impurities are studied (x = 1, 1/2, 1/4, and 1/8). The results show the effect of varying Fe concentration on the magnetic and stable properties.

Key words: diluted magnetic semiconductor; electronic band calculation; magnetism; stability
PACC: 8160C; 8160B; 8140
CLC number: TN304.7　　**Document code**: A　　**Article ID**: 0253-4177(2004)12-1586-05

1 Introduction

For more than three decades, diluted magnetic semiconductors (DMS) have attracted a great of interests because of the combination of magnetic and semiconducting properties. In recent years, the attention has been focused on Ⅲ-Ⅴ based DMS since the successful preparation of $(Ga_{1-x}Mn_x)As$ and $(In_{1-x}Mn_x)As$[1,2]. The potential utility of some physical properties of the DMS are numerous. To deeply understand the properties in semiconductors and the practical need for the ferromagnetic diluted magnetic semiconductor materials with high Curie temperature for the realization of the semiconductor spin-electronic devices, much experiments and theories have been done.

The $(Ga_{1-x}Fe_x)As$ was successfully prepared by Haneda *et al.*[3] X-ray results offer direct evidence of Fe substitution for Ga sites in GaAs and $(Ga_{1-x}Fe_x)As$ compound is mainly paramagnetic[3,4]. Then in GaFe-As composite structures, photo-induced magnetization as well as magnetoresistance (MR) effect is observed even at room temperature[5,6]. Photo induced ferromagnetic has been observed[7] too. Moriya *et al.*[8] have investigated the properties of (Ga, Fe, Mn)As as an interesting material. The result shows that the ration of paramagnetic part to ferromagnetic part increases with the increase of Fe content. Park *et al.*[9] have obtained that the ground state is ferromagnetic for $(Ga_{0.937}Fe_{0.063})As$ by using the linear muffin-tin orbital (LMTO) band method with the local density approximation (LDA). To get a further insight of the magnetic nature of $(Ga_{1-x}Fe_x)As$, we studied the homogeneous incorporation $(Ga_{1-x}Fe_x)As$ by using the first-principles band structure calculation with supercell models, where x = 1, 1/2, 1/4, and 1/8.

2 Computational details

We have used self-consistent tight-binding linear muffin-tin orbital (TB-LMTO) method with

* Project supported by Foundation for Outstanding Young Teachers in University of Henan Province, and the Natural Science Foundation of the Education Bureau of Henan Province (Nos. 2003140027, 2004140004)

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Received 27 January 2004, revised manuscript received 15 June 2004

the atomic sphere approximation (ASA) for $(Ga_{1-x}Fe_x)As$[10~12]. This method has been applied to deal with the adsorption properties of Co atoms on the Si (100) surface and Mn on GaAs (001) by Wei[13], Yang[14], respectively, and the obtained results are in good agreement with experiments. $(Ga_{1-x}Fe_x)As$ is modeled by supercells with $x = 1$, 1/2, 1/4, and 1/8. The crystal structure for $x = 1$ is of zinc-blende type (space group T_d^2). For $x = 1/2$, 1/4, and 1/8, we have assumed a supercell structure with a volume of two, four, and eight times of the primitive unit cell of the zinc-blende type structure. In Cartesian for $x = 1$, the primitive translation vectors are (1, 0, 0), (0, 1, 0), (0, 0, $\sqrt{2}$); the same for 1/2, 1/4, and 1/8, the primitive translation vectors are (2, 0, 0), (0, 1, 0), (0, 0, $\sqrt{2}$) and (2, 0, 0), (0, 2, 0), (0, 0, $\sqrt{2}$) and (2, 0, 0), (0, 2, 0), (0, 0, 2), respectively. For studying Fe-doped GaAs, we replace a pair of Ga atoms by Fe atoms. The reason for substituting two Ga atoms by Fe is to allow us to study if the coupling between Fe atoms is ferromagnetic or antiferromagnetic. The structure is then optimized by minimizing the total energy around the raw lattice constant for different magnetic configurations, *e.g.* paramagnetism (PM), ferromagnetism (FM), antiferromagnetism (AFM) phases. In the calculation of partial DOS, we have used the optimized values of lattice parameters. Spin polarized bands are calculated with minimal basis consisting of s-, p-, and d-orbital for Ga, As, and Fe. Apart from the valence states of Ga, Fe, and As, the core orbitals are kept frozen to their isolated atomic form.

3 Results and discussion

From the variation of total energy with lattice constant for different magnetic configurations (PM, FM, AFM), we get that the optimized lattice constants of $(Ga_{1-x}Fe_x)As$ for $x = 1$, 1/2, 1/4, and 1/8 are 0.52, 0.55, 0.56, and 0.565nm, respectively, satisfied with the Vangard's rule. The lattice constant of $(Ga_{1-x}Fe_x)As$ becomes smaller with the increase of Fe composition, reflecting the short bond length of Fe- As than that of Ga- As[3].

Figure 1 shows that the total energy of ferromagnetic state and paramagnetic state are larger than that of antiferromagnetic state with the same composition for $x = 1$, 1/2, 1/4, and 1/8, which tells us that the ground state is antiferromagnetic and the total energy of the antiferromagnetic state is the lowest among the three. Here, we choose the total energy of antiferromagnetic state as the zero point of energy for corresponding concentration x. But in experiment the magnetism of $(Ga_{1-x}Fe_x)As$ compound is mainly paramagnetic[3,4]. The difference between theoretical expectation and experimental results is not clear; it may stem from the local defects, FeAs, Fe_2As, $FeAs_2$[4], the inhomogeneous Fe incorporation or disorder. Figure 1 also shows that the total energy of ferromagnetic state E_1 decreases with x decreasing. When $x = 1/8$, there is the almost negligible energy difference between ferromagnetism and antiferromagnetism. With decreasing x ($< 1/8$), it is possible that the ground state becomes ferromagnetic, which is good agree with the result of Park *et al.*[9].

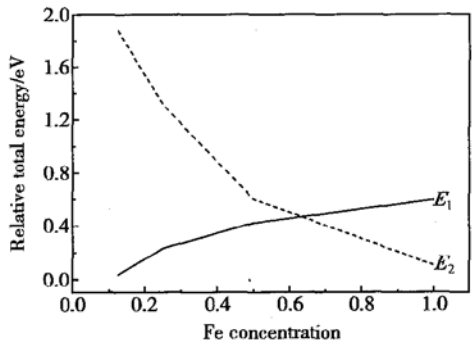

Fig. 1 Total energy of ferromagnetic state (E_1, solid line) and paramagnetic state (E_2, dash line) versus Fe concentration when the system is stable

In III-V semiconductor, a cation vacancy creates three holes in the valence band leaving anion dangling bonds. When Fe occupies the cation site, it donates three electrons to fulfill the bonds. Fe is left with five unpaired d electrons which should

give rise to $5\mu_B$/Fe atom. Dealing with ideal uncompensated systems, we should always obtain a magnetic moment of $5\mu_B$/Fe[15], but our results are not agree with the theory. These results are showed in Table 1, which shows that the magnetic moment of Fe decreases (except $x=1$) with x increasing. The induced moment of the nearest As

Table 1 Magnetic moments of Fe atom and the nearest As atom versus Fe concentration All moments are in units of μ_B.

x	Fe	As
1	2.8593	-0.1044
1/2	2.1325	-0.0189
1/4	2.7113	0.0438
1/8	3.1081	0.0572

atom increases with x decreasing. The induced moment of the nearest As atom is parallel ($x < 1/2$)[16] or antiparallel ($x \geqslant 1/2$) to the moment of Fe[3]. These behaviors can be explained by Figs. 2 (a) and (b). First of all we observe a metallic behavior, namely, the Fermi level exists not only in the spin-up bands but also in the spin-down bands. The presence of both spin-up and spin-down states at the Fermi level leads to strong electron transfer between two spin channels, which may be one of the reason that the moment of Fe is strongly dependent on concentration of Fe. These findings are in good agreement with the result of Ref. [16]. Secondly, in Fig. 2(a) ($x=1/8$), an spin-down impurity band lies in the semiconducting gap of GaAs and is separated from the valence and conduction bands, a spin-up impurity band merges at the top of the valence band and is not completely filled. For valence band, the contribution is mainly from spin-up Fe3d and As4p states, so the magnetic polarization parallel to the Fe moment is mainly induced at the As site. With x increasing, the impurity band becomes broader. Lastly, in Fig. 2(b) ($x=1/2$), the number of spin-down Fe3d and As4p states is obviously increased, so the magnetic moment of Fe ($x \leqslant 1/2$) and the induced moment of the nearest As atom decrease with x increasing. The increasing of spin-down As4p states is larger than that of the

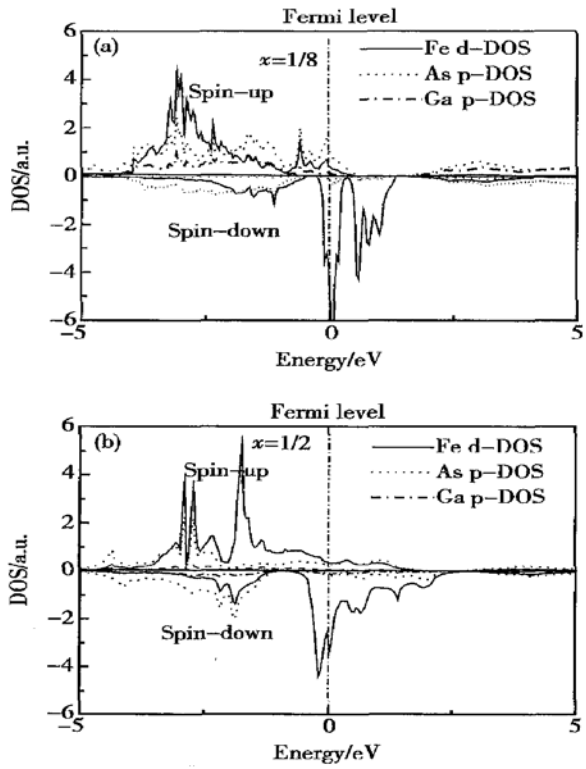

Fig. 2 Partial DOS for (Ga, Fe)As ($x=1/8, 1/2$)

spin-down Fe3d states, so the ferromagnetic hybridization of Fe3d and As4p states turns into antiferromagnetic one. Following along Ref. [16], we consider it may be that the introducing of Fe makes the number of the spin-up holes become very little, with which the number of the As spin-up electrons increases. At the same time, because of a lower energy position of the Fe3d spin-down states, the As spin-down states' hybridizing with empty states makes the number of the As spin-down electrons decrease. The main number of the states at and above the Fermi level is the spin-down type. However the spin-down hole states are strongly localized on the Fe atom, which can not mediate ferromagnetism efficiently[16]. With the decreasing of the concentration of Fe, the number of the spin-down As4p and Fe3d electrons decreases and the number of the spin-down holes increases, which means that the ferromagnetic state may occur in $(Ga_{1-x}Fe_x)As$ at homogeneous low Fe concentration ($x < 1/8$). The mechanism is perhaps similar

to the hole-mediated ferromagnetism. This idea is not fully convincing at present since substitutional Fe acts as a deep acceptor in GaAs[8].

4 Summary

In conclusion, we have used the first-principles LMTO-ASA band calculation for diluted magnetic semiconductor $(Ga_{1-x}Fe_x)As$ (x = 1, 1/2, 1/4, 1/8). By analyzing the variation of total energy with different lattice constants and different magnetic configurations (PM, FM, AFM), we get the optimized lattice constants for x = 1, 1/2, 1/4, and 1/8 are 0.52, 0.55, 0.56, and 0.565nm, respectively, and the ground states are all antiferromagnetic. The difference in total energies of antiferromagnetic and ferromagnetic states increases with increasing x, which shows that ferromagnetic state may occur in $(Ga_{1-x}Fe_x)As$ at homogeneous low Fe concentration. Using the optimized lattice constants we calculated the partial DOS of Fe3d, As4p, and Ga4p for x = 1/8 and 1/2. The results show that $(Ga_{1-x}Fe_x)As$ is metallic and the impurity band becomes broader with increasing x. The calculation of magnetic moment shows that the magnetic moment of Fe decreases (except x = 1) with increasing x. It also shows that the induced moment of the nearest As atom increases with decreasing x. The induced moment of the nearest As atom is parallel ($x < 1/2$) or antiparallel ($x \geqslant 1/2$) to the moment of Fe. For the complex property in practice, more work on theory and experiment are needed in the future.

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稀磁半导体$(Ga_{1-x}Fe_x)As$的磁性及稳定性*

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摘要: 运用第一性原理下的LMTO-ASA方法研究稀磁半导体$(Ga_{1-x}Fe_x)As$(x= 1, 1/2, 1/4和1/8)的电子结构、磁性及其稳定性. 计算了Fe掺杂浓度的变化对$(Ga_{1-x}Fe_x)As$的磁性及稳定性的影响.

关键词: 稀磁半导体; 电子能带计算; 磁性; 稳定性

PACC: 8160C; 8160B; 8140

中图分类号: TN304.7 **文献标识码**: A **文章编号**: 0253-4177(2004)12-1586-05

* 河南省高校青年骨干教师基金和河南省教育厅自然科学基础研究计划(编号: 2003140027, 2004140004)资助项目

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2004-01-27收到, 2004-06-15定稿