Enhancement of Ferromagnetic Coupling in Mn/GaAs Digital Ferromagnetic Heterostructure by free-hole injection

M. C. Qian, C. Y. Fong, and Warren E. Pickett

Department of Physics, University of California, Davis CA 95616-8677

(Dated: December 9, 2005)

Abstract

We have studied the effect of free-hole injection on the ferromagnetic coupling in the Mn/GaAs digital ferromagnetic heterostructure (DFH) using *ab initio* electronic-structure methods. The DFH is modeled by a supercell periodically consisting of a δ-doped layer of MnAs and fifteen layers of GaAs. The injection of free holes is simulated by assigning a range of missing electrons in unit cell. The δ-doped layer of Mn atoms in GaAs introduces three spin-polarized hole bands which are the consequence of hybridization between the d-states of the Mn atoms and the p-states of the nearest neighboring As atoms. These spin-polarized holes are confined to the vicinity of the MnAs layer. There are two closed and one open hole Fermi surfaces around the Γ point. After the injection of free holes, the Fermi energy is lowered, consequently the number of spin-polarized holes in the layer of MnAs increases monotonously. We characterize the ferromagnetic coupling by the total energy difference between the antiferromagnetic and the ferromagnetic phases, E_{FA}, per one pair of Mn atoms. The results of E_{FA} and the projected spin-polarized holes at a Mn atom and the nearest neighboring As atoms are given as functions of the injected free holes. Our results show the enhancement of the ferromagnetic coupling by free-hole injection, which is in agreement with the experimental observation.

PACS numbers: 73.22.-f, 61.48.+c, 73.20.Hb, 71.30.+h

Since it is crucial to have spintronic devices operating at or above room temperature, how to increase the ferromagnetic coupling between the magnetic atoms in a sample has recently attracted much attention. In the Mn dilute doped in GaAs, the Curie temperature T_C can be raised by increasing the Mn concentration. However, it is limited to be 5-7% [1], because the hole concentration will be saturated by the compensation of Mn acceptors due to the defects of As-antisites and Mn interstitials.[2] Postgrowth annealing[3] has been used to increase the Curie temperature to about 150 K. Recently, the concept of increasing hole concentration by the p-type co-doping has been adopted by several groups [4–6] to enhance the ferromagnetic coupling in materials. By injecting free holes into the Mn layer through the two-dimensional hole gas from p-type AlGaAs layers, Nazmul *et al.*[7] achieved remarkably high T_C up to 250 K in the Mn δ-doped in GaAs, digital ferromagnetic heterostructure (Mn/GaAs-DFH), They suggested the mechanism of the spin-polarized holes mediating the magnetic coupling between the Mn atoms so that they can tune the ferromagnetic ordering by controlling the hole concentration. Theoretically, the Mn/GaAs-DFH is predicted to be a half-metal[8], which has 100 % spin polarization at the Fermi energy, E_F. Therefore, the experimental results are extremely encouraging in showing that there is a possibility to increase T_C for this material even though the half-metallicity may dismish somewhat. In this paper, we report our results of simulations using a first-principles method and an ideal model DFH consisting of a δ-layer of Mn in GaAs, (Mn/GaAs-DFH), and confirm that the free-hole injection in fact enhances the ferromagnetic coupling in the Mn layer, thus can increase T_C.

The total energy and electronic structure calculations have been performed using the pseudopotential plane-wave method based on density functional theory[9] within the generalized gradient approximation (GGA).[10] We used the VASP program[11] for our spin polarized calculations in which ultrasoft pseudopotential are used for Mn, Ga and As atoms with their normal valence electron configurations. These pseudopotentials are well tested.[12] A cutoff energy of 450 eV and Monkhorst-Pack[13] mesh of $11 \times 11 \times 1$ **k**-points were used in all calculations. Wigner-Seitz sphere radii of Mn, Ga and As atoms were chosen as 1.35 Å , 1.41 Å and 1.59 Å , respectively, to construct projected densities of states.

The supercell modeling the Mn/GaAs-DFH is shown in Fig. 1 and consists of a δ-doped layer of MnAs and 15 layers of GaAs in the z-direction. The Mn layer substitutes Ga layer to model an ideal digital ferromagnetic heterostructure. The z-direction of the supercell is coincided with a cubic [001] axis while the x- and y-axis are along the [110] and [$\bar{1}$10]

directions of the face-centered cubic structure. The lattice constant is taken as $a_0/\sqrt{2}$, where $a_0 = 5.722$ Å is the optimized lattice constant of the host semiconductor GaAs. Since the ionic relaxation effect will not alter our conclusions about the features of our study on this DFH, in the following, we focus our analysis on the unrelaxed systems. The injection of free holes are modeled by subtracting electrons from the total number of electrons of the DFH and incorporating a uniform compensating background to maintain the charge neutrality of the supercell.

To investigate the enhancement of the magnetic coupling by the free-hole injection, we carried out total energy calculations for ferromagnetic and antiferromagnetic phases without and with the free holes injection. The difference of the total energies E_{FA} between ferromagnetic and antiferromagnetic order is used to characterize the ferromagnetic coupling in the Mn layer.

The total and partial densities of states (PDOS) for the majority- and minority-spin channels are given in Fig. 2. The PDOSs show projections onto the Mn, the nearest neighboring As, and Ga. For the majority spin channel, we find a finite density of states at E_F, as shown in the top panel. In the second and third panels, the antibonding states in a 2.8 eV range around E_F from ~ -1.8 to $+1.0$ eV show strong hybridization between Mn-t_{2g} and As-p states. The bonding states centered at ~ 3.2 eV below E_F also exhibit strong Mn-t_{2g}–As-p character. The nonbonding Mn-e_g states are located at ~ 2.5 eV below E_F, having a width of about 1.0 eV. The conduction states at ~ 1.0 eV above E_F and higher are contributed mainly by the As atoms. For the minority spin channel, a semiconducting gap of ~ 0.42 eV is opened between valence and conduction bands. The valence states have mainly also the Mn-t_{2g}–As-p character and are located at ~ 1.5 eV below E_F. The conduction states originate from both antibonding p–t_{2g} hybrid states and nonbonding e_g states of the Mn, leading to a peak at ~ 1.0 eV. The states coming from the Ga atoms are nearly non-spin-polarized. The results show that the interaction of a Mn atom is confined primarily to its nearest-neighbor As atoms and exhibits a two-dimensional feature.

The band structures of the model Mn/GaAs-DFH without and with 0.25 free-hole injection are presented in Fig. 3. The solid and dotted horizontal lines in Fig. 3 (c) and (d) indicate the E_F with and without injection. Without injection, the half-metallic properties of the DFH are well manifested by the majority spin states intersected by the E_F, while the minority spin states exhibiting the semiconducting behavior. The metallic states at the E_F

are originated from the d-states of the Mn atoms hybridizing with their nearest neighbor As p-states. Because the periodic supercell is used, the hole states form bands. They are indicated by 1, 2 and 3 in Fig 3 (a). After injecting 0.25 free holes per Mn, the Fermi energy is lowered by 0.11 eV. The injection results in two parts of additional holes in system. One part is distributed in the host semiconductor GaAs, another part is located in the δ-doped layer of MnAs, which is fully spin polarized and should contribute to the enhancement of ferromagnetic coupling in the Mn layer. On the other hand, the Fermi energy is below the valence band maximum of host semiconductor GaAs and the half-metallicity is slightly destroyed.

We now turn the discussions to the Fermi surfaces and begin with the case without the free-hole injection based on the band structure. Around the Γ-point, there are three bands intersected by the E_F. The Fermi surfaces of these three bands are given by the dotted lines in Fig. 4. There are two closed Fermi surface and one open Fermi surfaces with the states at the Γ-point unoccupied. It is in this sense, we call the Fermi surfaces as the hole surfaces. When the DFH is injected by free holes, the hole surfaces expand as determined from the self-consistent electronic calculations for the hole concentration increased from zero to 0.25 free holes per Mn. For the injection of 0.25 free holes per Mn, the expanded hole surfaces are shown by the solid lines in Fig. 4. The shape of hole surfaces are almost not changed.

We determined the ferromagnetic coupling between the Mn atoms as a function of the free-hole injection. The E_{FA} is shown in Fig. 5 and exhibits a monotonic increasing behavior as the hole concentration increases. The result suggests the enhancement of the ferromagnetic coupling between the Mn atoms in the δ-doped layer and agrees with experimental results[7]. The additional holes in the Mn and the nearest neighboring As atomic sites are also shown in Fig. 5. At the beginning of free-hole injection, the efficiency is almost 100% to induce the spin-polarized holes in the layer of MnAs. After the Fermi energy is lowered to touch the valence band maximum of host semiconductor, the injected free holes are distributed in both host semiconductor and the layer of MnAs. The injection of 0.25 free holes gives 0.1 spin-polarized holes in the layer of MnAs and the efficiency is about 40 %. These results demonstrate clearly that the hole-mediated mechanism enhances the ferromagnetic coupling between the Mn atoms.

In summary, we examine the effect of free-hole injection on the ferromagnetic coupling in a Mn/GaAs DFH by first-principles calculations. For the DFH without injection, it is

a half-metal with the Fermi surfaces composed of three sheets. The injection of free holes expands the hole Fermi surafces and increase the hole concentration in the layer of MnAs. The enhancement of ferromagnetic coupling between the Mn atoms by the free-hole injection is consistent with the experimental finding. These results also confirm the free-hole mediated mechanism for the magnetic coupling in a Mn/GaAs DFH.

Acknowledgments

This work is partially supported by National Science Foundation with grant No. ESC-0225007, and the San Diego Supercomputer Center.

---

[1] H. Ohno, Science **281**, 951 (1998); K. C. Ku, S. J. Potashnik, R. F. Wang, S. H. Chun, P. Schiffer, N. Samarth, M. J. Seong, A. Mascarenhas, E. Johnston-Halperin, R. C. Myers, A. C. Gossard, and D. D. Awschalom, Appl. Phys. Lett. **82**, 2302 (2003).

[2] P. A. Korzhavyi, I. A. Abrikosov, E. A. Smirnova, L. Bergqvist, P. Mohn, R. Mathieu, P. Svedlindh, J. Sadowski, E. I. Isaev, Yu. Kh. Vekilov, and O. Eriksson, Phys. Rev. Lett. **88**, 187202 (2002); K. M. Yu, W. Walukiewicz, T. Wojtowicz, I. Kuryliszyn, X. Liu, Y. Sasaki, and J. K. Furdyna, Phys. Rev. B **65**, 201303(R) (2002).

[3] M. Adell, L. Ilver, J. Kanski, V. Stanciu, P. Svedlindh, J. Sadowski, J. Z. Domagala, F. Terki, C. Hernandez and S. Charar, Appl. Phys. Lett. **86**, 112501 (2005).

[4] J. A. Schuller, E. Johnston-Halperin, C. S. Gallinat, H. Knotz, A. C. Gossard, and D. D. Awschalom, J. Appl. Phys. 95, 4922 (2004); E. Johnston-Halperin, J. A. Schuller, C. S. Gallinat, T. C. Kreutz, R. C. Myers, R. K. Kawakami, H. Knotz, A. C. Gossard and D. D. Awschalom, Phys. Rev. B **68**, 165328 (2003).

[5] K. Onomitsu, H. Fukui, T. Maeda, Y. Hirayama and Y. Horikoshi, J. Vac. Sci. Technol B **22**, 1746 (2004).

[6] K. M. Yu, W. Walukiewicz, T. Wojtowicz, W. L. Lim, X. Liu, U. Bindley, M. Dobrowolska and J. K. Furdyna, Phys. Rev. B **68**, 041308R (2003);

[7] A. M. Nazmul, S. Sugahara, and M. Tanaka, Phys. Rev. B **67**, 241308R (2003); A. M. Nazmul, T. Amemiya, Y. Shuto, S. Sugahara, and M. Tanaka, Phys. Rev. Lett. **95**, 017201 (2005).

[8] S. Sanvito and N. A. Hill, Phys. Rev. Lett. **87**, 267202 (2001).

[9] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964); W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).

[10] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).

[11] G. Kresse and J. Hafner, J. Phys.: Conden. Matt. **6**, 8245 (1994); G. Kresse and J. Furthmuller, Phys.Rev. B **54**, 11169 (1996).

[12] J. E. Pask, L. H. Yang, C. Y. Fong, W. E. Pickett, and S. Dag, Phys. Rev. B **67**, 224420 (2003).

[13] H. J. Monkhorst and J. D. Pack, Phys. Rev. B **13**, 5188 (1976).

[14] J. H. Zhao, F. Matsukura, T. Takamura, E. Abe, D. Chiba, and H. Ohno, Appl. Phys. Lett. **79**, 2776 (2001).

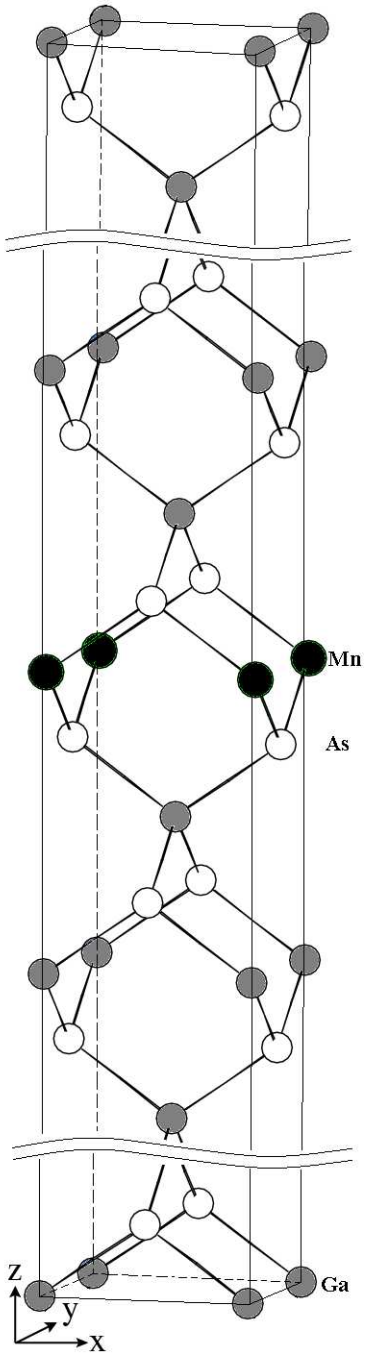

FIG. 1: Mn/GaAs DFH unit cell, consisting of one layer MnAs and 15 layers GaAs in the z-direction. Black, grey and open circles denote Mn, Ga and As atoms, respectively.

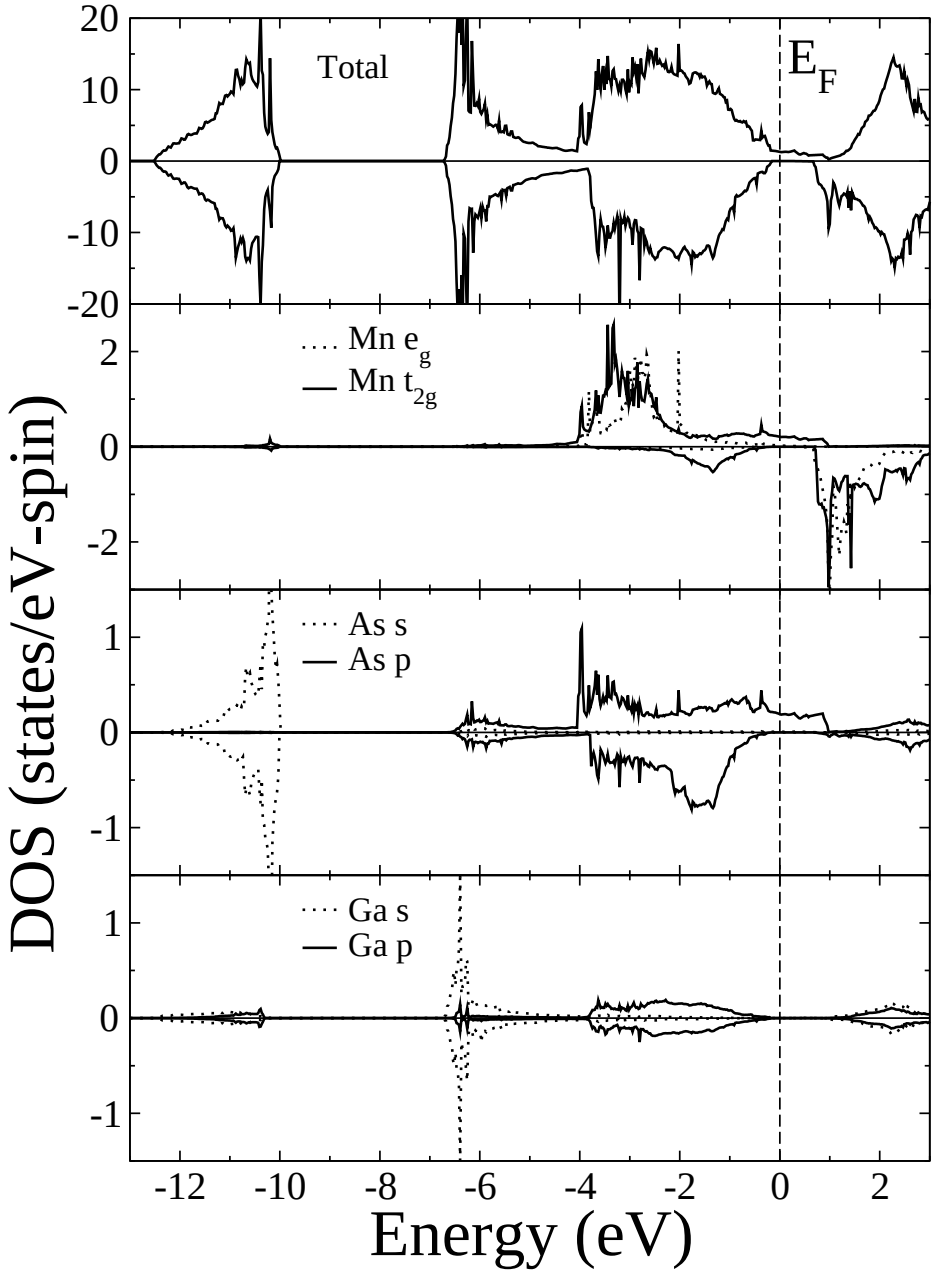

FIG. 2: Calculated total and partial densities of states for Mn δ-doped in GaAs. Majority densities are plotted as positive values; minority densities, as negative. The vertical dashed line indicates the Fermi level.

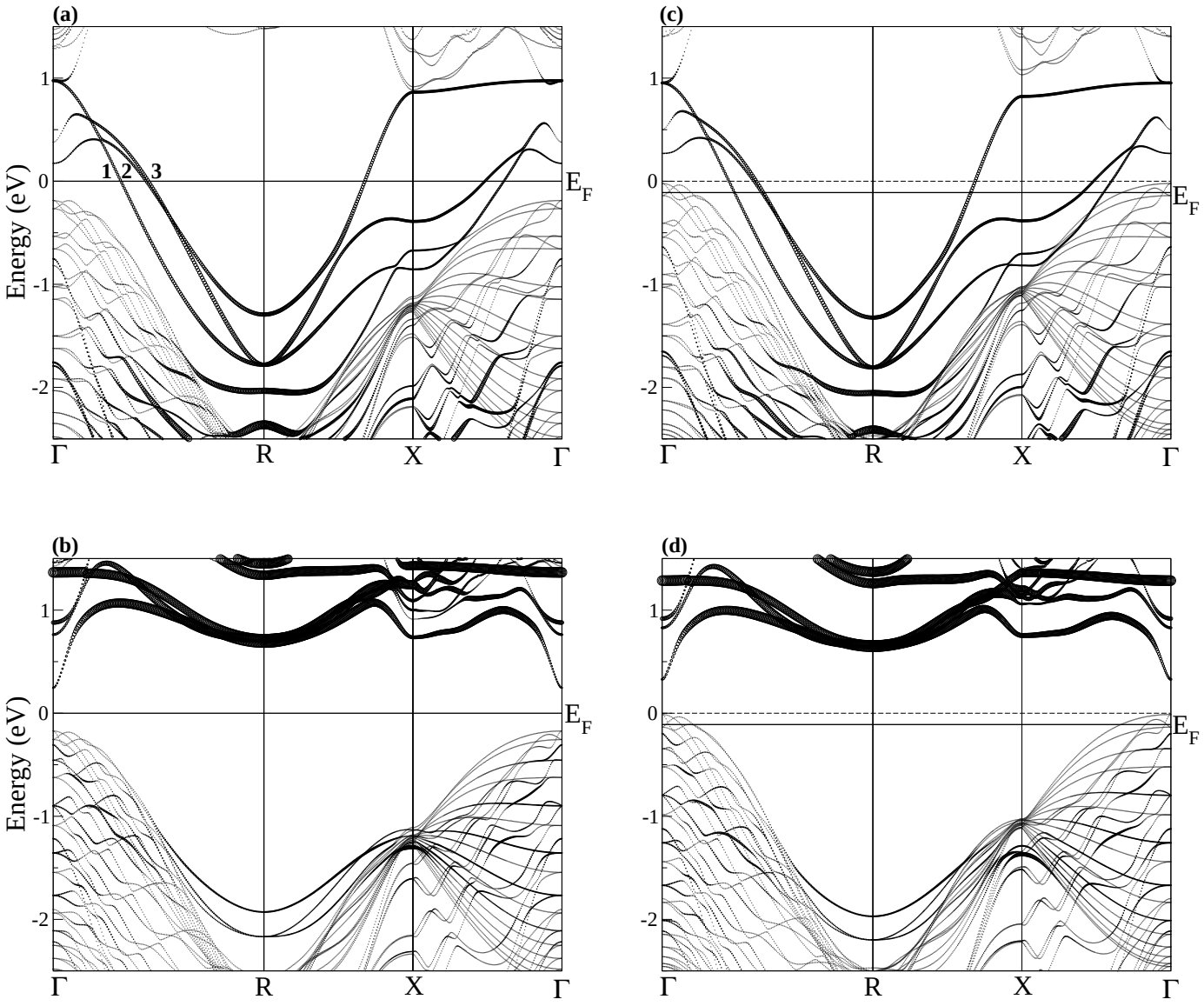

FIG. 3: Calculated band structure along high symmetry lines Γ-R-X-Γ in the $\mathbf{k}_x$-$\mathbf{k}_y$ plane, (a),(c) spin up and (b),(d) spin down for Mn δ-doped in GaAs without and with the 0.25 free-hole injection, respectively. High symmetry points of the Brillouin zone are $\Gamma = (0,0,0)$, R$= (1,1,0)$, X$= (1,0,0)$. The size of circles indicates the fraction of Mn d character. The conduction bands are labeled 1, 2 and 3. The Fermi energy without injection is set to zero. After 0.25 free-hole injection, the Fermi energy is down by 0.11 eV.

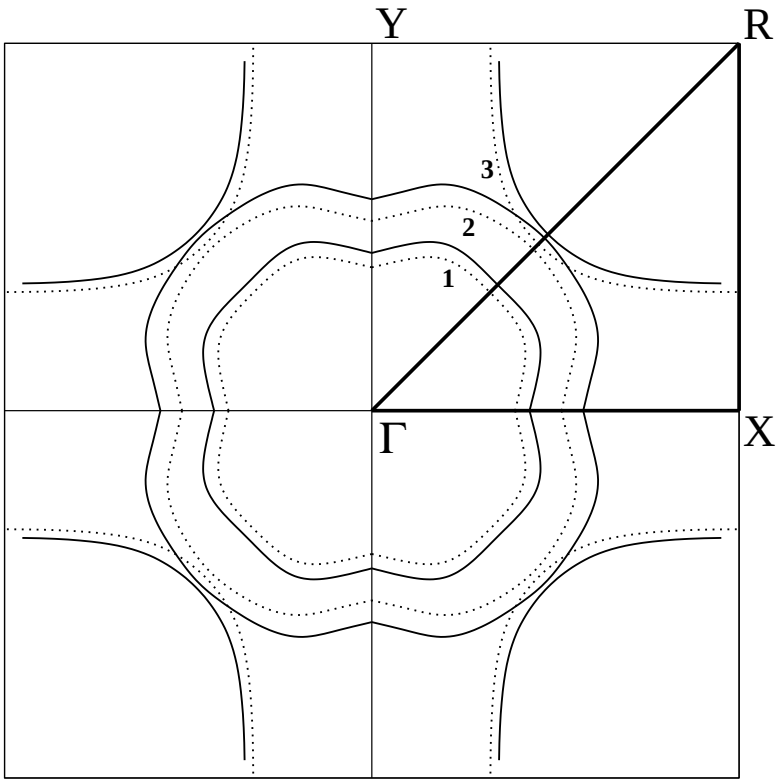

FIG. 4: Calculated two-dimensional Fermi surfaces. The solid and dotted lines refer to the Mn δ-doped in GaAs with and without 0.25 free-hole injection, respectively.

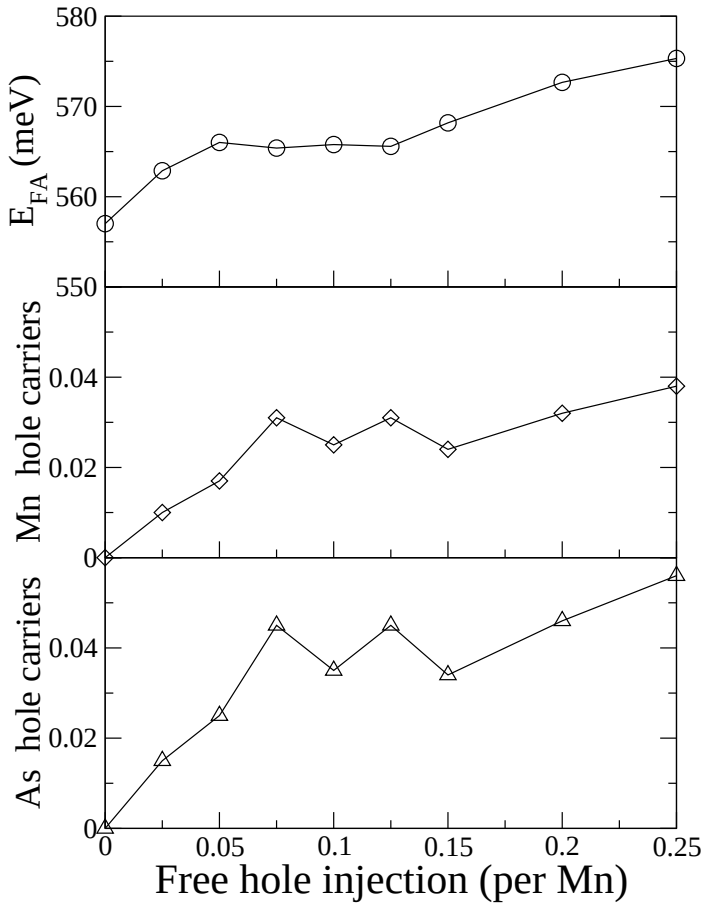

FIG. 5: Calculated ferromagnetic coupling between the Mn atoms and additional spin-polarized hole carriers (in unit of electron) located at the Mn and the nearest neighboring As atomic sites, as a function of the free-hole injection into system.