Uniaxial strain-dependent magnetic and electronic properties of (Ga,Mn)As nanowires*

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Variations in magnetic and electronic properties as a function of uniaxial strain in wurtzite (Ga,Mn)As nanowires (NWs) grown along the [0001] direction were investigated based on density functional theory (DFT). We found that (Ga,Mn)As NWs are half-metal, and the ferromagnetic state is their stable ground state. The magnetism of the NWs is significantly affected by the strain and by the substituent position of Mn impurities. By examining charge densities near the Fermi level, we found that strain can regulate the conductive region of the NWs. More interestingly, the size of spin-down band gap of the NWs is tunable by adjusting uniaxial stress, and the NWs can be converted from indirect to direct band gap under tension.

Keywords: dilute magnetic semiconductors, (Ga,Mn)As, nanowire, strain

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1. Introduction

Over the past several years, extraordinary enthusiasm has developed in the frontier of semiconductor research dealing with spintronic semiconductor nanostructures, especially nanowires. Compared with conventional semiconductor devices, spintronic devices have the potential advantages of nonvolatility, increased data processing speed, decreased electric power consumption, and increased integration densities, owing to the utilization of the spin freedom of electrons.[1] Some spin-dependent devices have already been designed or even fabricated despite the fact that they cannot meet the requirements of practical applications yet because of working temperature and other parameters.[2–5] Since the late 1980s, III–V dilute magnetic semiconductors (DMSs), such as (In,Mn)As[6] and (Ga,Mn)As,[7] have been successfully prepared by low-temperature molecular-beam epitaxy (LT-MBE), which made DMSs a frontier of spintronic research.

In DMSs like (Ga,Mn)As, substituent Mn atoms act as acceptors, introducing both local moments and holes. Recent experiments have demonstrated that ferromagnetism in (Ga,Mn)As is mediated by the impurity band derived from Mn dopants.[8,9] It should be noted that under equilibrium growth conditions the incorporation of magnetic Mn ions into III–As semiconductor crystals is limited to approximately 0.1%.[10] So LT-MBE, very much a nonequilibrium growth technique, was applied to make Mn concentration sufficient to form ferromagnetism and get a high transition temperature. Already, Curie temperatures as high as 200 K[11] have been obtained in (Ga,Mn)As nanostructures.

Because of the importance for both basic science and applications in semiconductor spintronic devices, (Ga,Mn)As-based nanostructures have attracted a lot of attention. Ohno *et al.*[12] reported the fabrication of all-semiconductor, light-emitting spintronic devices using III–V heterostructures based on (Ga,Mn)As, and in 2007, the spin valve effect was successfully realized in all-semiconductor (Ga,Mn)As/p-GaAs-(Ga,Mn)As trilayer devices,[13] which opens a parameter space for studying GMR different from that of metallic systems. Besides, various theoretical studies investigated the properties of (Ga,Mn)As. For instance, Franceschetti *et al.*[14] demonstrated that the magnetic properties of (Ga,Mn)As alloys depend on the spatial configuration of the magnetic impurities as well as the alloy composition. Another study[15] showed that the strength of ferromagnetic interaction between two Mn ions in (Ga,Mn)As depends heavily on the direction of the line connecting them, because of the inhomogeneous spatial distribution of holes. On the other hand, strain engineering has already been used to enhance the performance of electronic devices, e.g., increasing the charge carrier mobility of CMOSFETs,[16] and enhancing the direct-transition photoluminescence of Ge layers[17] and Ge-on-Si films.[18] NWs are ideal materials for the application of high elastic strain due to their enhanced yield strength and other mechanical properties. So it is desirable to apply stress on (Ga,Mn)As NWs experimentally and explore changes in properties, particularly unexpected ones.

In this article we report our use of uniaxial strain to investigate the magnetic and electronic properties of (Ga,Mn)As

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NWs. Our first-principles calculations were performed using self-consistent density functional theory (DFT) as implemented in the SIESTA package.[19] We primarily investigate the magnetically stable ground state of NWs, as well as the transformations of magnetism caused by strain. Then we explore the electronic properties of NWs, including density of states (DOS), charge densities and band structure.

2. Theoretical

On one hand, GaAs NWs are commonly grown by using MBE technique, and their crystal structure is often dominated by hexagonal wurtzite phase. On the other hand, one *ab initio* theoretical study indicates that, for GaAs NWs with diameters of up to 5 nm, the most stable nanowires have the wurtzite [0001] structure.[20] Based on all these findings, we chose Mn-doped GaAs NWs with wurtzite [0001] structure as our computational model. The diameter of the NW in our model is 1.3 nm. For each simulation run, one Mn impurity is substituted for one Ga cation, which is labeled by a certain arbitrary number in Fig. 1(a). Then the elementary cell is doubled along the c direction, to guarantee that there is a pair of Mn ions in each periodic image. Thus, the supercell used in our study contains 96 atoms.

In our calculations, the exchange correlation potential is approximated by the generalized gradient approximation (GGA).[21] SIESTA code uses a linear combination of atomic orbitals (LCAO) as a basis set and the valence states are described using a double-ζ polarized (DZP) basis set. The energy cutoff for the plane waves is 300 Ry and the Brillouin zone is sampled using a $1 \times 1 \times 8$ Monkhorst–Pack grid. We allow the internal coordinates to relax until the Hellmann–Feynman forces are less than 0.03 eV/Å.

3. Results and discussion

To study the magnetism of (Ga,Mn)As NWs, we calculated the energy difference (ΔE) between the total energy of a wire with the Mn pairs at three different locations for parallel (ferromagnetic, FM) and antiparallel (antiferromagnetic, AFM) alignment of Mn magnetic moments

$$\Delta E = E_{\mathrm{AFM}} - E_{\mathrm{FM}}.$$

We successively applied c-axial strains of -5% to 5% to the wire. The results are presented in Fig. 1(b), and the positive ΔE obtained at zero applied stress suggests that ferromagnetic state is the ground state of (Ga,Mn)As NWs. It also can be

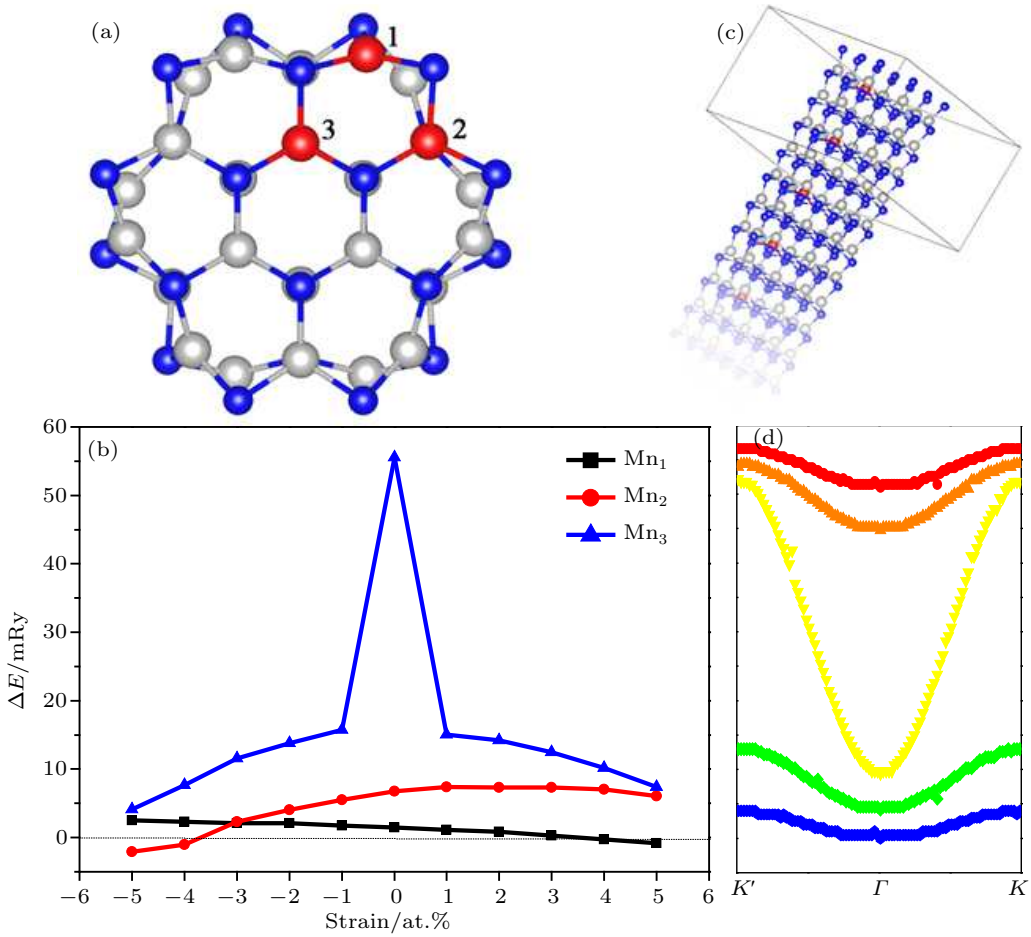


Fig. 1. (color online) (a) Top view of (Ga,Mn)As NW with Mn atoms (red balls) in three different positions. The blue balls denote As; gray represents Ga atoms; (b) the ΔE of wires as a function of uniaxial strain; (c) side view of (Ga,Mn_3)As NW with a supercell outlined by a box Mn_3 means a Mn impurity takes the #3 site; (d) magnon energy spectrum of (Ga,Mn_3)As NWs; the red, orange, yellow, green, and blue lines correspond to -5%, -3%, 0%, 3%, and 5% of strain, respectively.

seen clearly that both tension and compression will significantly weaken the stability of ferromagnetism for $(Ga,Mn_3)As$. In addition, the black line shifts down in energy while the red one shifts up, and excessive stress can make both of them antiferromagnetic. So we conclude that the variation trend of magnetic properties of (Ga,Mn)As NWs depends on the spatial position of the magnetic impurities as well as the amount of stress. Furthermore, the strongest FM coupling is obtained when a Mn atom is substituted for the cation in position 3 (or equivalent), while the weakest is obtained when the Mn atom is in position 1. That is, in (Ga,Mn)As NWs it is more ferromagnetically favorable for Mn to take a site inside the wire than to stay at the lateral surface. Going further, we explored the magnon energy spectrum of $(Ga,Mn_3)As$ NWs (see Fig. 1(d)). The spin wave stiffness constant, which reflects the decay rate of the spin wave, can be determined by examining the curvature of the spectral line at the Γ point. Either tension or compression will cause the line to be flatter and thus decrease the spin wave stiffness constant, making magnon lifetime shorter. Now that figure 1(b) has told us the ideal Mn substitutional site in the wire, all the following calculations were implemented for Mn_3-doped GaAs NWs unless otherwise specified. In fact, our calculations show that although different configurations lead to numerical differences in density of states, band structures, etc., our overall conclusions will not be affected.

Next, we calculated the total density of states (TDOS) of a wire with FM coupling, as is shown in Fig. 2(a). Electrons at the Fermi level show complete spin polarization: the spin up channel has conducting behavior while the spin down evidences insulated behavior. So this (Ga,Mn)As NW is half-metal, which is consistent with a previous investigation of structure-dependent electronic and magnetic properties of (Ga,Mn)As and (In,Mn)As NWs.[22] Besides, we can see that the effect of uniaxial stress on the TDOS is not prominent. The partial density of states (PDOS) of some electron orbitals in (Ga,Mn)As at zero applied stress was also investigated, as is presented in Fig. 2(b). The majority bands around Fermi level are due to 4p electrons of both Ga and As atoms. In the energy range of −4.9 eV−−4.3 eV, there is a strong interaction among As-4p, Ga-4s, and Mn-3d states, and this hybridization will bring about the formation of a chemical bond. Moreover, the magnetic moment of (Ga, Mn)As NWs arises mainly from the asymmetric distribution of Mn-3d states, as we can see in Fig. 2(b), and the PDOS of Mn-3d orbits is influenced by the uniaxial stress, but not dramatically.

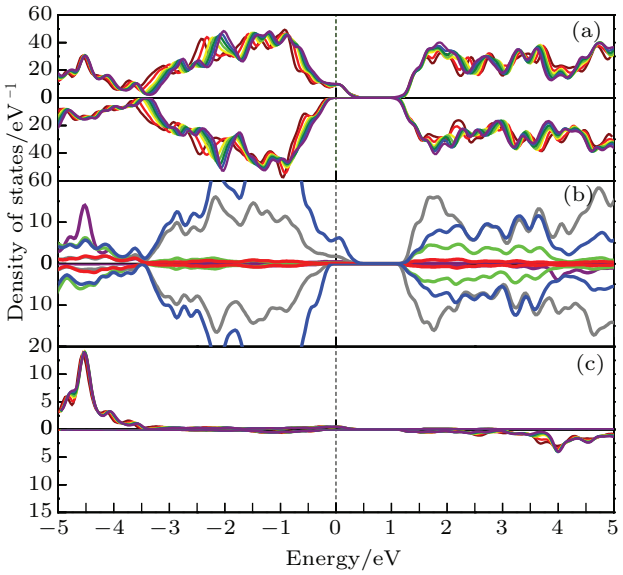


Fig. 2. (color online) (a) TDOS of (Ga,Mn)As NWs without and with strain; (b) PDOS of (Ga,Mn)As NWs at zero applied stress. The purple, green, gray, red, and blue lines correspond to Mn-3d, Ga-4s, Ga-4p, As-4s, and As-4p orbits, respectively; (c) PDOS of Mn-3d orbits without and with strain. In panels (a) and (c), the uniaxial strain takes the value of −5%, −3%, −1%, 0%, 1%, 3%, and 5% successively, presented by burgundy, red, yellow, green, olive, blue, and purple lines, respectively. The vertical dashed line denotes Fermi level in all three, and all the calculations are performed assuming FM coupling.

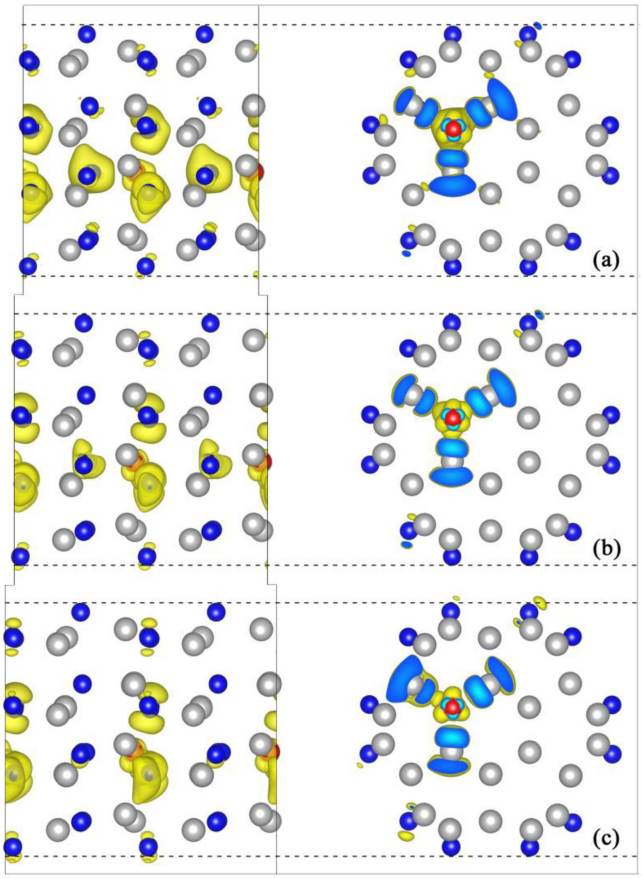


Fig. 3. (color online) Isosurfaces of charge densities of bands corresponding to the energy interval from the Fermi level to 0.1 eV higher; the yellow isosurface represents the charge of 0.001 e/Å^3; (a) −5% of strain; (b) 0% of strain; (c) 5% of strain.

In addition, we examined the charge density near the Fermi level, obtained by summing the charge densities of all the k-points contributed in the small energy region from the Fermi level to 0.1 eV above the Fermi level. The isosurfaces of charge densities shown in Fig. 3 for −5%, 0%, and 5% of strain bring out the nature of the states near E_f. Since conductivity is closely related to charge density near the Fermi

level, one can see that the conductivity of the system is dominated by Mn atoms as well as their neighboring As atoms. Another very striking feature is that compressive strain forces the charge densities to move toward the interior of the wire, while tensile strain makes them approach the edge, which indicates that strain can modulate electronic transport properties of the NWs effectively.

Finally, we analyzed the electronic band structure of (Ga,Mn)As NWs, as depicted in Fig. 4. Certain spin-up valence bands cross the Fermi level, from which we can observe metallic trends, while the spin-down bands act like semiconductors, and Table 1 lists the energy gap (E_g) values with varying applied uniaxial strain. On one hand, when we add tensile stress to the wire, E_g becomes smaller overall and there occurs a remarkable decrease of the band gap of up to 254 meV at 5% of strain. On the other hand, we noticed that the spin-down component can be converted into direct-band-gap from indirect by tensile stress. That is, the size and type of the band gap are stress-dependent, which is intriguing indeed.

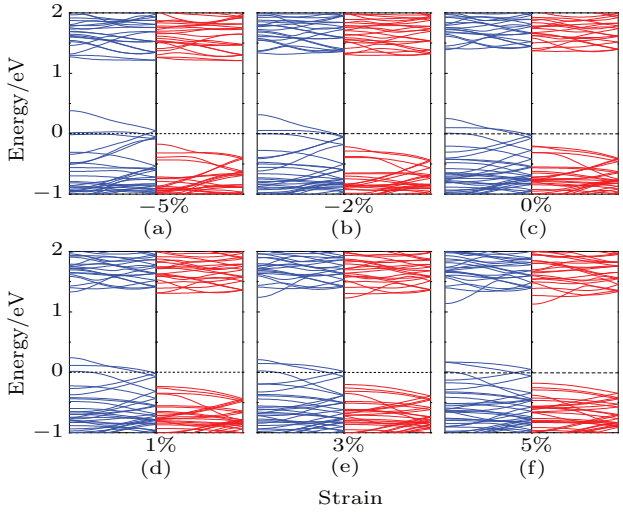


Fig. 4. (color online) Electronic band structure of (Ga,Mn)As NWs plotted successively according to uniaxial strain. Spin-up and spin-down bands are represented by blue and red lines, respectively. The horizontal dashed lines denote the Fermi level. In panels (a)–(f), the uniaxial strain takes the value of −5%, −2%, 0%, 1%, 3%, and 5%, respectively.

Table 1. Energy gap (E_g) of spin-down bands of (Ga,Mn)As NWs, D and I represent direct and indirect, respectively.

Strain	−5%	−4%	−3%	−2%	−1%	0%	1%	2%	3%	4%	5%
E_g/eV	1.382	1.438	1.486	1.513	1.539	1.563	1.543	1.492	1.4321	1.371	1.309
D /I	I	I	I	I	I	I	I	D	D	D	D

4. Conclusions

In our work, DFT methods were employed to explore the magnetic and electronic properties of (Ga,Mn)As NWs as a function of different degrees of uniaxial strain. As we predicted, (Ga,Mn)As NWs are half-metal, and the ferromagnetic state is their stable ground state. Under the *c*-axial stress, the magnetism of NWs can be regulated. By examining the charge densities near the Fermi level, we found that strain can affect the distribution of the conductive region in the wire. More interesting, we also discovered that the spin-down band gap of (Ga,Mn)As NWs is size-tunable by adjusting uniaxial stress, and tensile stress can even convert it from indirect to direct band gap. Actually, uniaxial stress has previously been applied continuously and reversibly on NWs by using, for example, a four-point wafer bending jig[23] or flexible metal substrates coated with polymer supports.[24] Our results could be useful for experimental applications of strained (Ga,Mn)As NWs and fabrication of novel devices.

References

[1] Wolf S A, Awschalom D D, Buhrman R A, Daughton J M, von Molnar S, Roukes M L, Chtchelkanova A Y and Treger D M 2001 *Science* **294** 1488

[2] Chakrabarti S, Holub M A, Bhattacharya P, Mishima T D, Santos M B, Johnson M B and Blom D A 2004 *Nano Lett.* **5** 209

[3] Holub M, Shin J, Chakrabarti S and Bhattacharya P 2005 *Appl. Phys. Lett.* **87** 091108

[4] Kohda M, Ohno Y, Takamura K, Matsukura F and Ohno H 2001 *Jpn. J. Appl. Phys.* **40** 1274

[5] Wang H, Hu G C and Ren J F 2013 *Chin. Phys. B* **22** 58504

[6] Munekata H, Ohno H, von Molnar S, Segmüller A, Chang L L and Esaki L 1989 *Phys. Rev. Lett.* **63** 1849

[7] Ohno H, Shen A, Matsukura F, Oiwa A, Endo A, Katsumoto S and Iye Y 1996 *Appl. Phys. Lett.* **69** 363

[8] Samarth N 2012 *Nat. Mater.* **11** 360

[9] Dobrowolska M, Tivakornsasithorn K, Liu X, Furdyna J, Berciu M, Yu K M and Walukiewicz W 2012 *Nat. Mater.* **11** 444

[10] Jungwirth T, Sinova J, Mašek J, Kučera J and MacDonald A H 2006 *Rev. Mod. Phys.* **78** 809

[11] Chen L, Yang X, Yang F, Zhao J, Misuraca J, Xiong P and von Molnár S 2011 *Nano Lett.***11** 2584

[12] Ohno Y, Young D K, Beschoten B, Matsukura F, Ohno H and Awschalom D D 1999 *Nature* **402** 790

[13] Xiang G, Sheu B L, Zhu M, Schiffer P and Samarth N 2007 *Phys. Rev. B* **76** 035324

[14] Franceschetti A, Zunger A and van Schilfgaarde M 2007 *J. Phys.: Condens. Matter* **19** 242203

[15] Guo X G, Chen X S, Sun Y L, Zhou X H, Sun L Z, Cao J C and Lu W 2004 *Phys. Rev. B* **69** 085206

[16] Chan V, Rengarajan R, Rovedo N, Jin W, Hook T, Nguyen P, Chen J, Nowak E, Chen X D and Lea D 2003 *IEEE International Electron Devices Meeting, IEDM'03 Technical Digest* Decmber 8–10, 2003 Washington, DC, USA, pp. 3.8.1–3.8.4

[17] Huo Y, Lin H, Chen R, Makarova M, Rong Y, Li M, Kamins T I, Vuckovic J and Harris J S 2011 *Appl. Phys. Lett.* **98** 011111

[18] Sun X, Liu J, Kimerling L C and Michel J 2009 *Appl. Phys. Lett.* **95** 011911

[19] Soler J M, Artacho E, Gale J D, García A, Junquera J, Ordejón P and Sánchez-Portal D 2002 *J. Phys.: Condens. Matter* **14** 2745

[20] Galicka M, Bukała M, Buczko R and Kacman P 2008 *J. Phys.: Condens. Matter* **20** 454226

[21] Perdew J P, Chevary J A, Vosko S H, Jackson K A, Pederson M R, Singh D J and Fiolhais C 1992 *Phys. Rev. B* **46** 6671

[22] Galicka M, Buczko R and Kacman P 2011 *Nano Lett.* **11** 3319

[23] Thompson S E, Suthram S, Sun Y, Sun G, Parthasarathy S, Chu M and Nishida T 2006 *IEDM'06 International Electron Devices Meeting*, Decmber 11–13, 2006 San Francisco, CA, USA, pp. 1–4

[24] Signorello G, Karg S, Björk M T, Gotsmann B and Riel H 2012 *Nano Lett.* **13** 917