Anisotropic half-metallic ground state of Mn atomic wire on GaAs(110)

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The authors have investigated magnetic properties of Ga-substituted Mn atomic wires on the GaAs(110) surface, using first-principles calculations based on the spin-density functional theory. The Mn atomic wires are assumed to align in the ⟨110⟩- and ⟨001⟩-directions. The ⟨110⟩-oriented wire is more stable than the ⟨001⟩-oriented one and has the ferromagnetic ground state with the magnetic moment of $4.0\mu_B$ per Mn atom. The band structure has a large dispersion along the wire and exhibits a half-metallic state. The ferromagnetic character of the Mn wire results from the double exchange interaction through the *p-d* hybridization between the Mn-3*d* and the GaAs surface states. © *2009 American Vacuum Society.* [DOI: 10.1116/1.3153285]

I. INTRODUCTION

A collaboration of magnetic and semiconducting materials attracts great interests as actualizations and creations of spintronic devices. A diluted magnetic semiconductor (DMS), the magnetic semiconductor doped with magnetic impurities, is leading candidates for fundamental components of such devices.[1] In particular, the III-V-based DMS is one of the excited targets.[2]

In most cases of recent DMSs, the transition metals, especially Cr or Mn, have been introduced into the III-V semiconductor hosts. The epitaxial layer of the (Ga, Mn)As has a well-aligned ferromagnetic order and allows higher Curie temperature than ever before.[3] The ferromagnetism for the III-V based DMSs originates from the hole-mediated exchange interaction.[4] It has experimentally been confirmed[5] that increase in the hole concentration elevates the Curie temperature, but the excessive Mn atoms break the ferromagnetism by reduction in the hole concentration. The excessive Mn atoms located at the interstitial position act as double donors,[6] which give rise to the self-compensation for holes supplied by the substitutional Mn atoms at the cation (Ga) sites. Recently, the substitution of a Mn atom with the surface Ga atom on GaAs(110) surface can be realized, using the substitution technique by a scanning tunneling microscope tip.[7] The Mn pair substituted using such a technique exhibits the ferromagnetic coupling of the Mn core spins. The scanning tunneling spectra of the Mn pair have two in-gap hole states, but on the other hand, for the single Mn atom, only one in-gap hole state emerges.[8] This splitting of the in-gap state can be explained by the carrier transfer between two Mn sites of the Mn pair.[9] The ferromagnetic alignment of the Mn core spins gives the same site energy on each site and induces a large splitting by coherent transfer. However, the antiferromagnetic alignment does not allow the coherent transfer because of the different site energy owing to the on-site exchange interaction. Furthermore, it has been revealed that the magnitude of the ferromagnetic coupling strongly depends on the orientation of the Mn pair.[8] Such an anistropic interaction has been investigated for the bulk system; the anisotropy of Mn–Mn coupling in bulk GaAs originates from the anisotropy of the hole states caused by the spin-orbit interaction.[10] Without the spin-orbit interaction, first-principles calculations also show the anisotropic ferromagnetic Mn–Mn interaction mediated by the *p-d* hoppings.[11] However, the magnetic interaction in Mn pair on the GaAs(110) surface has not been clarified yet.

In this study, we investigate the magnetic properties of Ga-substituted Mn atomic wires on the GaAs(110) surface. We focus on the anisotropic Mn–Mn interactions in the atomic wire, considering orthogonal two orientations with each other, the ⟨110⟩ and ⟨001⟩ directions on the surface. Additionally, we explore a mechanism of the onset of the ferromagnetic spin coupling between neighboring Mn atoms in the wires.

II. CALCULATION METHODS

We prepare a slab model for the clean GaAs(110)-(1×1) surface with a thickness of 6 atomic layers. Two layers of Ga and As atoms from the back side of the slab are fixed at the bulk position. The back side surface is terminated with H atoms and their bond lengths are fixed at the optimized values. Mn atoms are substituted with the topmost Ga atoms on the GaAs(110)-(1×1) reconstructed surface. We adopt a periodic supercell geometry consisting of a slab and a vacuum region with a width of 12 Å. A (2×2) unit cell is prepared parallel to the surface for the study of magnetic stabilities between ferromagnetic (FM) and antiferromagnetic (AFM) states along the wire and a (1×2) or (2×1) unit cell for the calculation of the FM band structure. As for the wire direction, two orientations, the ⟨110⟩ and

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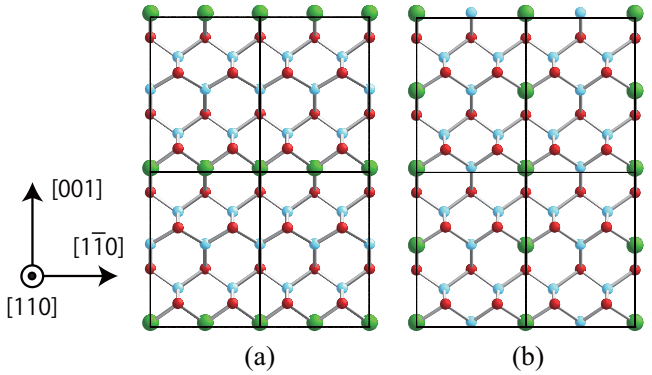

FIG. 1. (Color online) Atomic arrangements of (a) ⟨110⟩- and (b) ⟨001⟩-oriented Mn atomic wires on GaAs(110). Large green (gray), small blue (white), and small red (black) balls indicate Mn, Ga, and As atoms, respectively.

⟨001⟩ directions, on the surface are assumed, as shown in Fig. 1. The in-plane lattice constant is fixed at the calculated bulk value.

We execute total-energy calculations based on the first-principles pseudopotential method within the spin density functional theory.[12–14] The Troullier–Martins type of the pseudopotential[15] is chosen for the Ga, As, and H atoms and the ultrasoft Vanderbilt type[16] for the Mn atom, in which the nonlinear core correction is considered.[17] The generalized gradient approximation[18] is used for the exchange-correlation potentials. The cutoff energy is set to be 36 Ry for a plane wave expansion. 64 **k** points are used for the integration over the two-dimensional (1×1) Brillouin zone.

TABLE I. Energy difference per two Mn atoms ΔE between the ferromagnetic (FM) and the antiferromagnetic (AFM) states for Mn atomic wires on GaAs(110) surface, in bulk GaAs, and the Mn wire in free space. The Mn–Mn separations in free space were assumed to have the same value as those on the GaAs(110). Negative signs of ΔE indicate that the FM states are more stable than the AFM states.

$\Delta E=E_{\mathrm{FM}}-E_{\mathrm{AFM}}$	⟨110⟩ wire	⟨001⟩ wire
On GaAs(110)	−0.410 eV	+0.008 eV
In free-space	−0.003 eV	+0.002 eV
Mn separation	4.05 Å	5.73 Å

The structural relaxation is carried out until the remaining forces acting on atoms are less than 10^{-3} Hartree$/a_B$.

III. RESULTS AND DISCUSSION

First, we show the magnetic properties of Mn atomic wires having the FM and AFM orders along the wire. They were studied for the models with the (2×2) in-plane unit cell including two Mn atoms, and the results are summarized in Table I. The ⟨110⟩-oriented wire on GaAs(110) has a large energy difference of 0.410 eV between the FM and AFM orders. This energy difference is much larger than that of the isolated Mn wire in free space, as shown in Table I. The FM states are the ground state only for the ⟨110⟩-oriented wire and have the magnetic moment of $4.0\mu_B$ per Mn atom, being independent of the wire orientations. Since the ⟨001⟩-oriented wire has few energy difference, the Mn–Mn coupling on the ⟨001⟩-directions is very small. This anisotropy

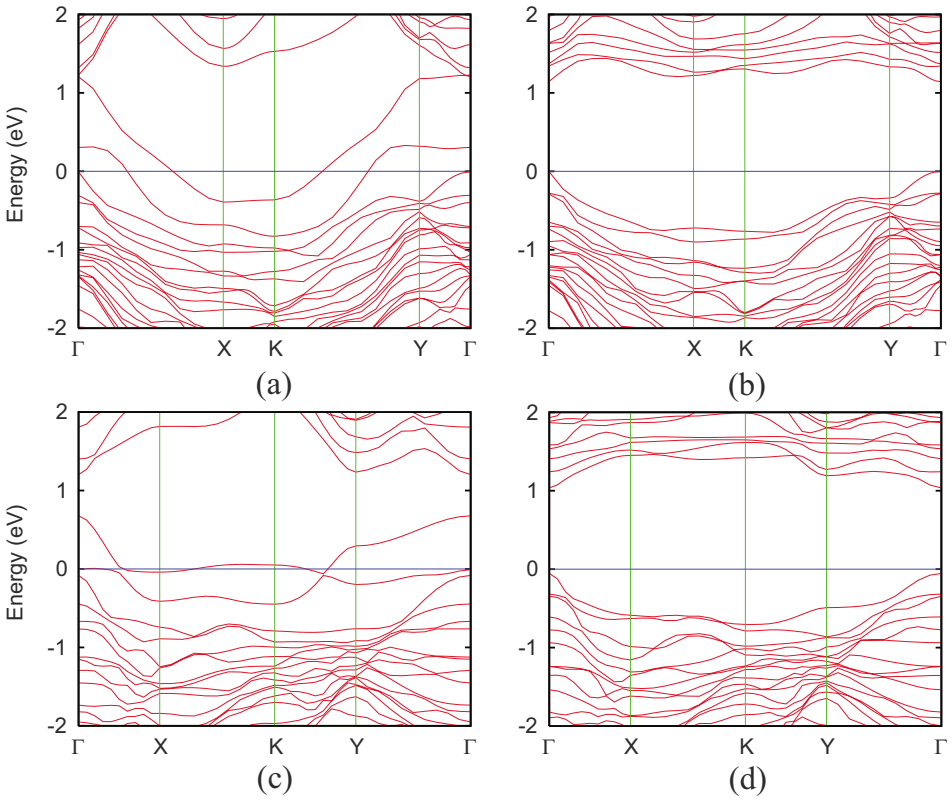

FIG. 2. (Color online) Spin-polarized band structure for (a) the majority spin and (b) the minority spin of ⟨110⟩-oriented Mn atomic wire and for (c) the majority spin and (d) the minority spin of ⟨001⟩-oriented Mn atomic wire on GaAs(110). The Fermi energy is shifted to zero. Γ-X and Γ-Y directions are parallel to the ⟨110⟩- and the ⟨001⟩-oriented wires, respectively.

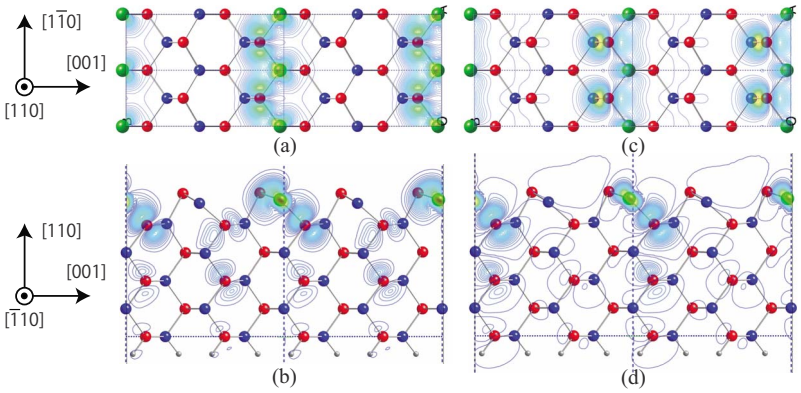

FIG. 3. (Color online) Cross-sectional distributions in the plane crossing Mn sites of the probability density for the half-metallic states of the majority spin at the Fermi level for the ⟨110⟩-oriented wire. (a) and (b) and (c) and (d) show the probability density at the wave vectors of $\mathbf{k}=(0.17,0,0)$ and (0.33, 0, 0), respectively.

of the magnetic coupling within the Mn wires corresponds well to the past experimental results for the surface Mn pairs[8] and theoretical studies[10,11] for the isolated Mn pair in bulk GaAs. Next, we evaluate structural stabilities between the ⟨110⟩- and ⟨001⟩-oriented wires. The FM ⟨110⟩-oriented wires are more stable than the FM ⟨001⟩-oriented ones by the energy of 0.345 eV per unit cell. We can assert that the ⟨110⟩-oriented wire is more stable than the ⟨001⟩ wire and has the stable FM ground state.

Next, we focus on the band structures near the Fermi level of the FM states for both wire orientations. In Fig. 2, the Mn wires have the half-metallic ground states irrespective of the wire orientation. The FM state for the ⟨110⟩-oriented wire has the metallic band structure for the majority spin, as shown in Fig. 2(a). Two metallic bands cross the Fermi energy in the direction parallel to the wire orientation. On the other hand, the minority spin has the semiconducting band structure as shown in Fig. 2(b). Thus, the ⟨110⟩-oriented wire has the one-dimensional half-metallic states along the wire orientation. In the case of the FM state of the ⟨001⟩-oriented wire in Fig. 2(c), the majority spin state has two metallic bands across the Fermi energy, whereas the minority spin state has the semiconducting band structure, as well as the ⟨110⟩-oriented wire. The FM state of the ⟨001⟩-oriented wire also exhibits the half-metallic character. One of the half-metallic states has a large energy dispersion along the ⟨110⟩-direction perpendicular to the wire orientation, but the other has a small dispersion. In contrast to the ⟨110⟩-oriented wire, the one dimensionality is broken for the ⟨001⟩-oriented wire. These results indicate that the ⟨110⟩-oriented wire is a pro-

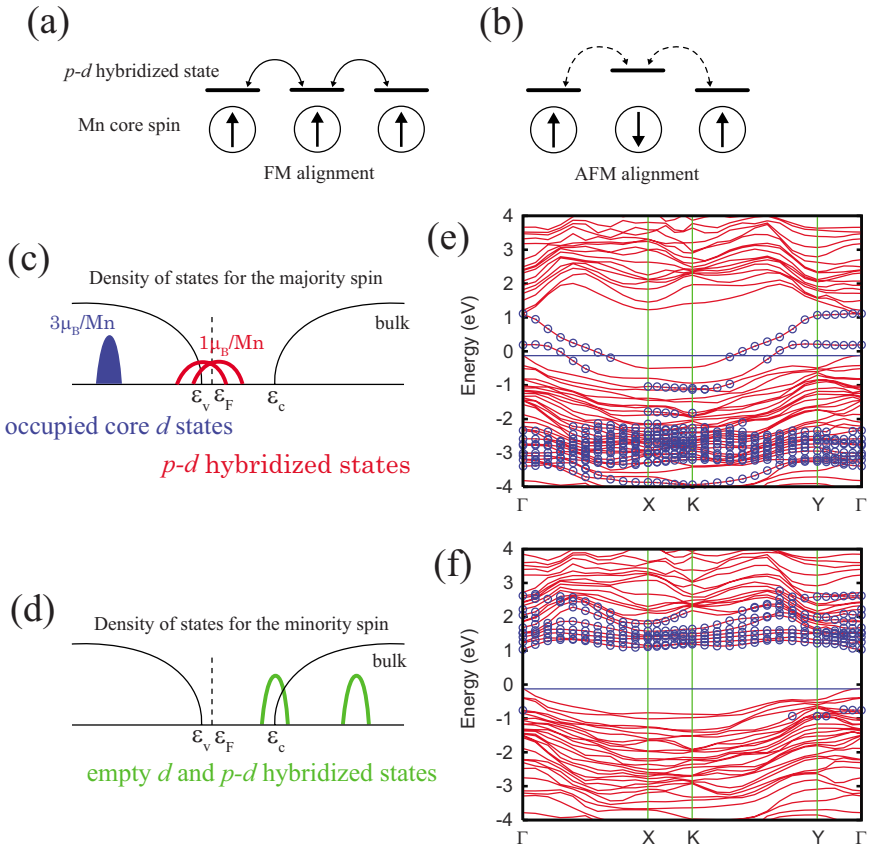

FIG. 4. (Color online) Schematic pictures for the transfer of *p-d* hybridized orbits between the Mn sites; (a) the FM alignment and (b) the AFM alignment of the Mn core spins. Here, the horizontal lines indicate the site energy at Mn sites. (c) and (d) show the schematic density of states attributable to Mn-*d* state for the majority spin and the minority spin, respectively, in addition to the bulk density of states of GaAs. Here, ε_v, ε_c, and ε_F indicate the valence band maximum, the conduction band minimum of GaAs, and the Fermi energy of the ⟨110⟩-oriented Mn wire, respectively. (e) and (f) show the band structures of the majority spin and the minority spin for the ⟨110⟩-oriented Mn wire, respectively, in which closed circles denote the states having large amplitudes at Mn site.

spective candidate for the one-dimensional spin filter because the wire has the half-metallic states with a large energy dispersion parallel to the wire orientation.

Finally, we discuss on the origin of the FM ground state for the ⟨110⟩-oriented wire. Figure 3 shows the distributions of the probability density for the metallic majority bands at the Fermi energy in the Γ-X axis in Fig. 2. The metallic states for the majority spin of the ⟨110⟩-oriented wire consist of the localized surface states which have small interaction with the neighboring wires. The d-orbitals of Mn atom hybridize with the As p-orbitals, parallel to the wire orientation, as shown in Fig. 3. The half-metallic states originate from the p-d hybridizations. The p-d hybridized orbitals near a Mn atom overlap those at the neighbor Mn site. The p-orbitals of As atoms assist the d-states of Mn atoms to interact with the neighboring Mn sites, whereas the d-states of Mn atoms cannot overlap directly with those at the neighboring sites as seen in Table I. When the core spins of the Mn atom are aligned as shown in Fig. 4(a) schematically, the site energies of the p-d hybridized states at the Mn site are the same. This same site energy helps the coherent transfer between p-d hybridized orbitals at each Mn sites. On the other hand, the antiferromagnetic alignment of the Mn core spins introduces the different site energies by the exchange interactions with the Mn core spins, and it is difficult to transfer coherently between Mn sites, as shown in Fig. 4(b). The ferromagnetism of the Mn core spins can be explained by the double exchange interaction with the energy gain of the kinetic energy. Actually, for the majority spin, the d-states included in the Mn cores are located in the vicinity of E_F-3 eV, and the d-states included in the p-d hybridizations appear in two half-metallic bands in the bulk bandgap, as shown in Fig. 4(e). For the minority spin, the d-states of Mn atoms are located in the vicinity of the conduction band bottom and higher energies, as shown in Fig. 4(f). Thus, the FM states have the total magnetic moment of $4.0\mu_B$: five d-orbitals of a Mn atom split into three occupied core states and half-occupied two p-d hybridized states, as schematically shown in Fig. 4(c). On the GaAs(110) surface, a Mn atom substituted into the Ga sites provides one hole as well as in bulk GaAs. On the Mn pair in bulk GaAs, the ferromagnetic coupling of Mn core spins has been explained by the similar p-d hopping mechanism.[11]

IV. SUMMARY

We have calculated the magnetic properties of Ga-substituted Mn atomic wires on the GaAs(110) surface. The ⟨110⟩-oriented wire has a strong intrawire ferromagnetic coupling and is more stable than the ⟨001⟩-oriented wire. The ⟨110⟩-oriented wire has two one-dimensional half-metallic states at the Fermi energy. These half-metallic states consist of the d-orbital of Mn atoms and the p-orbital of As atoms. The ⟨110⟩-oriented Mn atomic wires on the GaAs(110) surface are promising materials for one-dimensional spin filters.

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