Uniaxial strain-modulated conductivity in manganite superlattice (LaMnO$_3$/SrMnO$_3$)

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We have investigated the magnetic ordering and electrical conductivity transitions of (LaMnO$_3$)$_2$/(SrMnO$_3$)$_2$ superlattices grown on SrTiO$_3$ substrate based on density-functional theory. It is found that the uniaxial tensile strain along the z axis of about 1.4% induced a magnetic transition from antiferromagnetic to ferromagnetic ordering. At the interface the orbital order changes from a combination of x^2-y^2 and $3z^2-r^2$ to x^2-y^2 as strain becomes more compressive; as a result the electrical transport is transformed from three-dimensions to two-dimensions at the high uniaxial compressive strain. Our results suggest that the out-of-plane electrical conductivity can be modulated and controlled by uniaxial strain. © *2011 American Institute of Physics.* [doi:10.1063/1.3548675]

With rapid progress in fabrication technology of heterostructures, the perovskite manganite superlattices (SLs) composed of alternating LaMnO$_3$ (LMO) and SrMnO$_3$ (SMO) layers have attracted considerable attention in recent years.[1–7] The systems are regarded as potential candidates for applications in spintronics due to the numerous magnetic and electric properties which arise from the interplay between spin, charge, and orbital degrees of freedom.[8–11] They are sensitive to the change of external conditions such as doping concentration, magnetic field, electric field, temperature, and strain. In particular, strain plays a very important role for tuning the structure and magnetic and transport properties in La$_{1-x}$Sr$_x$MnO$_3$ (LSMO). Previous studies have reported that different types of strain can induce magnetic ordering transition in LSMO.[12–17] One is intrinsic strain arising from lattice misfit by growing on different substrates. Generally, the transition of magnetic ordering originates from the competition between double exchange and superexchange mechanisms.[18,19] Ferromagnetic (FM) ordering is stable in LMO/SMO SL grown on La$_{0.3}$Sr$_{0.7}$Al$_{0.65}$Ta$_{0.35}$O$_3$ substrate with no strain. The magnetic ordering of SL grown on SrTiO$_3$ (STO) with tensile strain favors an A-type antiferromagntic (A-AFM) order. The SL grown on LaAlO$_3$ substrate with compressive strain induces a C-type antiferromagnetic order.[14,15] The other is external strain arising from uniaxial strain or hydrostatic pressure.[16,17] However, all of these theoretical works have only focused on the effects of intrinsic strain on the magnetic properties of alloy or solid solutions. In fact, many interesting and unexpected phenomena have been found in SL structure other than in alloy or solid solution. Recent experiments have observed that the resistivity of the SL structures is far lower than that of the corresponding

alloy structures or solid solutions.[20] Due to the complexity and sensitivity of structure and magnetic and electric properties of LSMO, a complete understanding of strain effect on the intrinsic physical mechanism has not yet been comprehensive up to now.

In this letter, we have investigated the uniaxial strain effect on the magnetic and transport properties in (LMO)$_2$/(SMO)$_2$ SL using first-principles calculations based on density-functional theory. The external uniaxial strain is defined as $\varepsilon=(c-c_0)/c_0\times100\%$ based on the conventional mechanical definition. The AFM-FM magnetic ordering transition occurs when the tensile uniaxial strain is up to 1.4%. More interesting, the out-of-plane conductivity can be modulated by the compressive strain, which leads to the conductivity transition from three-dimensions (3-D) to two-dimensions (2-D). All calculations are performed by using the generalized gradient approximation (GGA+U) method as implemented in the Vienna *ab initio* simulation package (VASP).[21–23] A $5\times5\times2$ Monkhorst–Pack mesh and a 400 eV plane-wave energy cutoff are used in calculations. For different strained systems, all the atom positions are relaxed until the force acting on each atom is less than 0.01 eV/Å. We choose the onsite Coulomb interaction parameters U_{eff} =3 eV to treat the localized d electron states of Mn atoms as assumed in other studies.[17]

The artificial SLs consist of alternating LMO and SMO unit cells grown on STO (001) substrate. Figures 1(a) and 1(b) show the relaxed structures of (LMO)$_2$(SMO)$_2$ superlattice under uniaxial strains $\varepsilon=0\%$ and $\varepsilon=4\%$. The in-plane lattice constant is constrained as the theoretical lattice constant of STO (3.95 Å). By virtue of the in-plane tensile strain from STO substrate, the cubic LMO and SMO undergo the tetragonal distortions of 0.99 and 0.96, respectively. The out-of-plane c of SL is obtained by minimizing the energy. It is found that the Jahn–Teller (JT) distortion ($\mathbf{Q}_2$) of MnO$_6$ oc-

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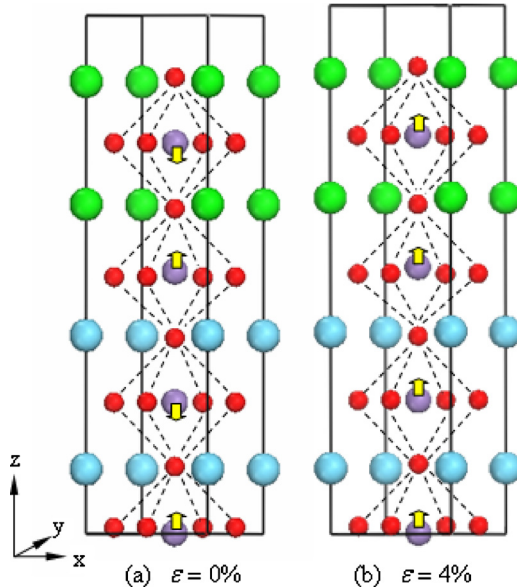
FIG. 1. (Color online) The relaxed ground structures of $(LaMnO_3)_2/(SrMnO_3)_2$ superlattice for (a) $\varepsilon=0\%$ with A-AFM magnetic ordering and (b) for $\varepsilon=4\%$ with FM ordering indicated by yellow arrows. Blue, green, purple, and red spheres denote La, Sr, Mn, and O atoms, respectively. The dashed lines show the MnO_6 octahedrons.

FIG. 3. (Color online) Orbital-resolved density of states for Mn e_g states with different uniaxial compressive strains. The blue and red lines stand for the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals, respectively. Positive and negative values are for majority and minority, respectively. The Fermi energy is set to zero.

tahedron for inner LMO layer is 0.11 Å in the ground state of this SL. As is well known, the JT distortion will be suppressed strongly for the mixed compound $(La,Sr)MnO_3$. Q_2 of MnO_6 octahedron in interface LMO layer is taken as 0.02 Å. We find that the SL system has A-AFM metallic ground states because the special structure of SL with the La and Sr atoms ordered arrangement and interface effect. It is in well agreement with experiment results.[1,12] The charges will leak from Mn^{3+} in LMO block to Mn^{4+} in SMO block mediated by O atoms because of interface effect in SL structure. The tensile in-plane strain results in MnO_6 octahedrons being compressive. The e_g charges prefer to occupy the lower energy orbital x^2-y^2 states than $3z^2-r^2$ states. The FM double exchange is stronger than the AFM superexchange in the MnO_2 plane, which favored the A-AFM order.

In order to investigate the effect of uniaxial strain along the growth direction of SL on the magnetic transition in $(LMO)_2/(SMO)_2$ SL, we show the total energy versus uniaxial strain for two different magnetic structures in Fig. 2. The calculated results indicate the AFM-FM phase transition under uniaxial tensile strain of about 1.4%. As a consequence, only the FM and A-AFM orders are considered. Since the in-plane Mn-O bonds are constrained by the substrate, the in-plane magnetic order stabilized FM order as that of the ground states. With the out-of-plane tensile strain applied on SL increasing, the MnO_6 octahedrons experienced a deformational process from compressive to elongated. The FM order is more stable than the AFM order when the compressive distortions of MnO_6 octahedrons disappear. Therefore, the magnetic phase transition depends strongly on the change of Mn-O bonds.

With the compressive uniaxial strain increasing, a fascinating phenomenon for the electrical conductivity transition of the SL from 3-D to 2-D has been observed. To gain the role of uniaxial strain in modulating the conductivity of this SL system, we studied further electronic properties of SL under different external compressive strains. The SL with the A-AFM ordering remains stable due to the compressive uniaxial strain. As we all know, the occupancy of e_g orbital strongly affects the electrical conductivity of LSMO. The e_g orbitals split into the in-plane x^2-y^2 lower energy orbital and out-of-plane $3z^2-r^2$ higher energy orbital related to the JT effect. Figure 3 shows the orbital-resolved density of states (DOS) of Mn e_g orbitals because the t_{2g} orbitals have no contribution to Fermi energy occupation. It is found that the occupancy of x^2-y^2 orbitals at Fermi level has nearly no change with the increase of the uniaxial strain. It originates from the in-plane Mn-O bonds that maintain the original lengths of the SL ground states. In contrast, the occupancy of $3z^2-r^2$ orbitals at Fermi level is decreasing with the compressive strain increasing. The larger is the compressive strain, the shorter are the out-of-plane Mn-O bonds. It is in favor of the strong hybridization between Mn and O atoms along the z axis. As a consequence, the bonding states move down to lower energy, while antibonding states move up to
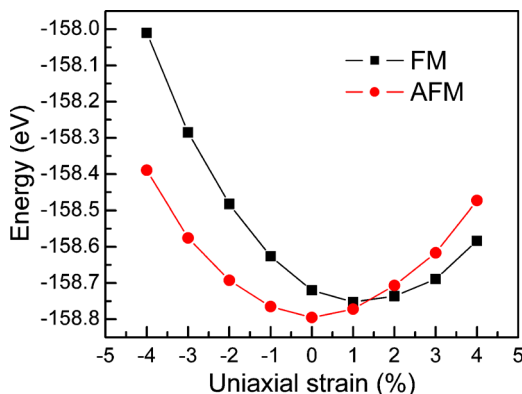

FIG. 2. (Color online) The total energy as a function of the uniaxial strain for the optimized superlattice with AFM and FM ordering, respectively.
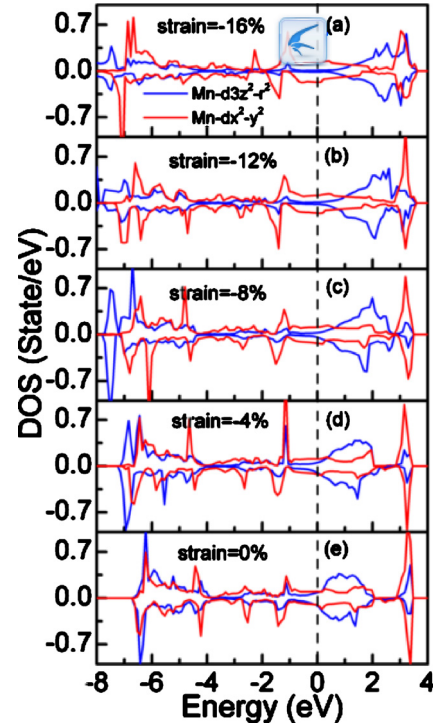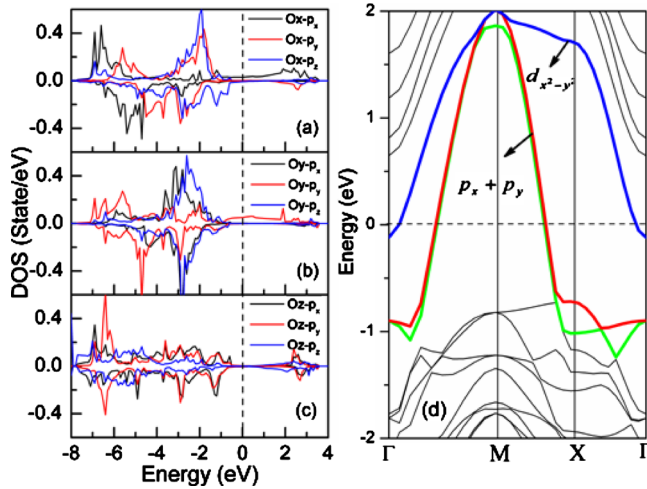
FIG. 4. (Color online) Orbital-resolved density of states for O atoms positioned at different directions under uniaxial strain $\varepsilon=-12\%$ (left panel). The black, red, and blue lines denote p_x, p_y, and p_z orbitals of O atoms, respectively. Positive and negative values are for majority and minority, respectively. The Fermi energy is set to zero. (d) shows the spin-up band structure of SL under uniaxial strain $\varepsilon=-12\%$.

higher energy. There is nearly no contribution of $3z^2-r^2$ orbitals at the region of Fermi level for $\varepsilon=-12\%$, as shown in Fig. 3(b). A band gap along the z axis is clearly visible. Only in-plane x^2-y^2 orbital occupies the Fermi level.

In order to confirm the electrical conductivity character of this SL, we also show the orbital-resolved DOS of O atoms positioned in three different directions around Mn atoms under compressive strain $\varepsilon=-12\%$ in Figs. 4(a)–4(c), respectively. It is found that the states at Fermi level are dominated by the p_x and p_y orbitals of O atoms positioned at x and y directions, respectively. These orbitals are hybridized strongly with the in-plane Mn-$d_{x^2-y^2}$ orbital. There is no contribution of p_z orbitals of all O atoms at Fermi level. Moreover, only Mn and O atoms are responsible for the electrical transport in LMO/SMO SL by scrutinizing the Fermi level occupation from the projected DOS of each kind of atoms. In other words, the metallic states are mainly from the contribution of Mn-$d_{x^2-y^2}$, O-p_x, and O-p_y in-plane states in this SL for $\varepsilon=-12\%$. It is also proved by the electronic band structure, as shown in Fig. 4(d). There are three bands across the Fermi energy. The one with higher energy is dominated by Mn-$d_{x^2-y^2}$ orbital. The other two bands (colored by red and green) are mainly from the contribution of O-p_x and O-p_y orbitals. All these states are lying in the x-y plane. Therefore, the electrical conductivity is transformed from 3-D to 2-D under the high compressive strain condition.

In conclusion, we have investigated that the uniaxial strain leads to the transition of magnetic ordering and electrical conductivity in $(LMO)_2/(SMO)_2$ SL grown on STO substrate. The AFM-FM magnetic order transition is present at the uniaxial tensile strain of about 1.4%. The out-of-plane electrical conductivity is decreasing with the uniaxial compressive strain increasing. At the interface the orbital order changes from a combination of x^2-y^2 and $3z^2-r^2$ orbitals to x^2-y^2 orbital as strain becomes more compressive; as a result the electrical transport is transformed from 3-D to 2-D under the high compressive strain of about -12%. The Mn-$d_{x^2-y^2}$, O-p_x, and O-p_y in-plane states are responsible for the 2-D conductivity. We can modulate and control the out-of-plane conductivity by changing the external uniaxial strain. These fascinating phenomena make it possible that the LMO/SMO SL becomes a potential candidate material for the piezomagnetic or piezoelectric switch.

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