Spin-moment formation and reduced orbital polarization in $LaNiO_3/LaAlO_3$ superlattice: LDA + U study

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Density functional band calculations have been performed to study $LaNiO_3/LaAlO_3$ superlattices. Motivated by recent experiments reporting the magnetic and metal-insulator phase transition as a function of $LaNiO_3$ layer thickness, we examined the electronic structure, magnetic properties, and orbital occupation depending on the number of $LaNiO_3$ layers. Calculations show that the magnetic phase is stabler than the nonmagnetic for finite and positive U values. The orbital polarization is significantly reduced by U even in the magnetic regions. The implications of the results are discussed in comparison to recent experimental and theoretical studies within the limitations of the LDA + U method.

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I. INTRODUCTION

Understanding transition-metal oxides is of perpetual interest and importance in condensed-matter physics and material science.[1] Recent advances in layer-by-layer growth techniques of heterostructures of transition-metal compounds have created particular interest due to their great scientific and technological potential.[2] Exotic material phenomena that are clearly distinctive from the “normal” phases include interface superconductivity,[3] magnetism,[4,5] charge,[6,7] and orbital reconstruction.[8]

One of the most intriguing classes of materials may be the nickelate superlattices.[9–16] A series of recent theoretical studies have created considerable interest, suggesting heterostructuring-induced orbital polarization and possible high-T_c superconductivity in a $LaNiO_3$ (LNO)/$LaAlO_3$ (LAO) superlattice.[12,17] In this picture, the two degenerate Ni e_g orbital states are split by a combination of translational symmetry breaking and on-site Coulomb interaction, leading to a cupratelike band structure. Although this kind of theoretical picture has been challenged by more recent dynamical mean-field theory (DMFT) calculations based on a charge-transfer model including oxygen states explicitly,[15] several experimental papers have found other interesting phenomena in this system. Boris and co-workers[10] reported a metal-to-insulator transition as a function of LNO layer thickness: Even though the bulk LNO is a paramagnetic (PM) metal, the heterostructure $(LNO)_m/(LAO)_n$ becomes insulating and magnetic if m is small, $m \leqslant 2$, while it remains PM and metallic when $m \geqslant 4$.[10] The insulating behavior was also observed by Freeland *et al.*[13] However, the detailed magnetic and electronic structure changes, as well as the other important physical quantities such as orbital polarization, have not yet been clearly understood as a function of layer thickness m.

In this study, we performed a detailed first-principles analysis for the electronic structure, magnetism, and orbital polarization of a LNO/LAO superlattice using the local density approximation with the Hubbard U correction (LDA + U) method.[18–20] Since previous calculations were performed at the LDA[14] or DMFT level[12,15] and assumed the bulklike PM phase, a LDA + U calculation can provide meaningful information, especially regarding the magnetism in this system. Our total energy calculations showed that the Ni spins order ferromagnetically within the LNO layer, and the interlayer couplings are also ferromagnetic (FM). This result may indicate the existence of another ground-state configuration in between $m \sim 2$ and $m \sim 4$ superlattices. The orbital polarization is significantly reduced by U, which is in contrast to the simplified Hubbard-type model prediction,[12] but consistent with the extended charge-transfer model DMFT calculation.[15] The calculated Ni d valence based on LDA + U supports the recently suggested picture for the metal/insulator phase diagram based on the d valency,[21] and demonstrates the importance of the double-counting issue. These results are discussed in comparison to recent theoretical and experimental studies.

II. COMPUTATIONAL DETAILS

For the band-structure calculations, we employed a Troullier-Martins type norm-conserving pseudopotential[22] with a partial core correction and linear combination of the localized pseudoatomic orbitals (LCPAO)[23] as a basis set. In this pseudopotential generation, the semicore $3p$ electrons for transition-metal atoms were included as valence electrons in order to take into account the contribution of the semicore states to the electronic structure. We adopted the local density approximation (LDA) for the exchange-correlation energy functional as parametrized by Perdew and Zunger,[24] and used an energy cutoff of 400 Ry and a k grid of $12 \times 12 \times 6$ per unit superlattice volume. The LDA + U functional is adapted from the formalism of Refs. 25 and 26.[27] The geometry relaxation has been performed with a force criterion of 10^{-3} hartree/bohr. During the relaxation process, the in-plane lattice constant is fixed to the $SrTiO_3$ value (3.905 Å) considering the substrate effect in the experimental situation. The optimized cell volume and the out-of-plane lattice parameter have been used.[14] The tilted structure of the oxygen octahedra is not considered in this study. The orbital polarization P, defined as

$$P = \frac{n_{x^2-y^2} - n_{3z^2-r^2}}{n_{x^2-y^2} + n_{3z^2-r^2}}, \quad (1)$$

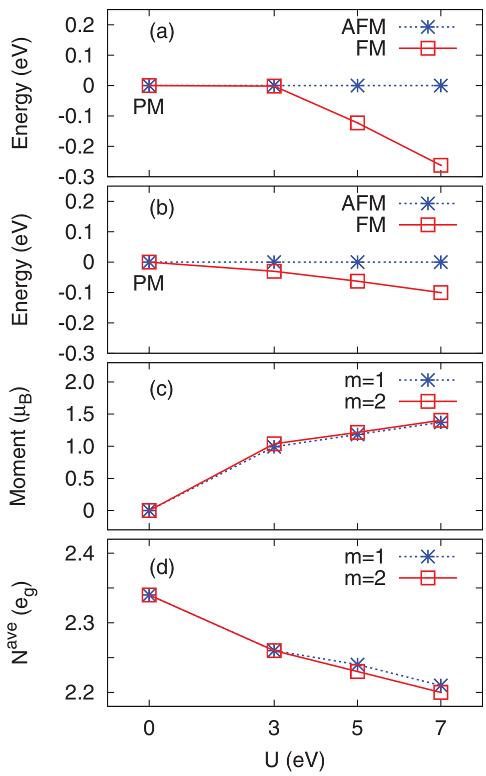

FIG. 1. (Color online) (a) Calculated total energy of $(LNO)_1/(LAO)_1$ with the in-plane spin ordering of FM and AFM as a function of U. (b) Calculated total energy of $(LNO)_2/(LAO)_1$ with the interlayer magnetic coupling of FM and AFM as a function of U where the in-plane spins are set to be FM. (c) Calculated Ni magnetic moment for $(LNO)_1/(LAO)_1$ and $(LNO)_2/(LAO)_1$ as a function of U (FM order considered). (d) Average number of Ni e_g electrons in $(LNO)_1/(LAO)_1$ and $(LNO)_2/(LAO)_1$ as a function of U.

can be calculated by integrating the projected density of states (DOS) up to the Fermi level. The Mulliken charge analysis has been used for the atomic charge decompositions. All the calculations were performed using the density functional theory code OPENMX.[28]

III. RESULT AND DISCUSSION

One natural and evidently important question raised from experimental studies on the magnetic moment formation in this heterostructure[10] is about the ground-state spin structure and its moment size. Figure 1 summarizes our results. Figure 1(a) shows the calculated total energy of the $(LNO)_1/(LAO)_1$ superlattice as a function of U. While at $U = 0$ eV the initial setups by both FM and antiferromagnetic (AFM) spins eventually converge to a PM solution, the magnetic ground states are stabilized at the finite U. As clearly seen in Fig. 1(a), FM spin ordering is energetically favored within the LNO plane. The energy difference between the FM and AFM state is 3, 246, and 525 meV per $(LNO)_1/(LAO)_1$ for $U = 3$, 5, and 7 eV, respectively. The FM spin arrangement is also favored for the interlayer (LNO-LNO) spin couplings as shown in Fig. 1(b), in which we present the total energies of $(LNO)_2/(LAO)_1$; the interlayer couplings are set to be FM or AFM while the in-plane order is FM. The calculated FM-AFM energy difference is 60, 126, and 201 meV per $(LNO)_2/(LAO)_1$ for $U = 3$, 5, 7 eV, respectively. The energy differences between the FM and AFM spin structures becomes larger as U increases in both cases of intralayer and interlayer coupling. That is, the on-site correlations stabilize FM spin ordering, and as U decreases, the two magnetic solutions become more close in their energies, and eventually converge to the PM phase at $U = 0$. The magnetic coupling between LNO layers separated by LAO is found to be two orders of magnitude smaller (in the case of $U = 5$) compared to the coupling strengths presented in Fig. 1. The magnetic moment is also dependent on U. Figure 1(c) shows the calculated magnetic moment for the FM case as a function of U. The moment increases as U increases as in the other typical correlated transition-metal oxide materials.[18,20,25] It is noted that, even at a quite small value of $U = 3$ eV, the Ni moment already becomes $\sim 1\mu_B$, and further increases to be $1.2\mu_B$ and $1.4\mu_B$ at $U = 5$ and 7 eV, respectively.

Our calculation results may seem to suggest that the magnetic moment formed in the thin-LNO superlattice[10] is ordered ferromagnetically. Since the spin-polarized oxide heterostructure could be useful for device applications, there has been active research for finding the structure that produces FM spin order.[29–31] Therefore our result of a FM ground state in LNO/LAO may have a positive implication for such an application. However, it should be noted that the muon-spin rotation (μSR) experiment by Boris *et al.*[10] is not well interpreted in the long-range FM ordering picture even though the μSR is basically a local probe, and that the origin of the metal-insulator phase transition in the nickelate series is not clearly understood yet. Especially regarding the charge disproportionation or ordering in nickelates, the conventional LDA + U has a clear limitation in describing such phenomena.[32,33] Moreover, Figs. 1(a) and 1(b) indicate the possibility of the AFM ground state in the negative U region, which is more or less related to the reported charge disorders in the nickelate systems.[32,33] Therefore one needs to be careful in the interpretation of our LDA + U results on the FM spin ground state as an indication of a long-range-ordered ground state as one may see in the actinide systems, for example.[34,35]

Our results have another interesting implication regarding the phase diagram. Since the thin-LNO superlattice, $(LNO)_{m\leqslant 2}/(LAO)_n$, can have either a FM insulating (FM-I) ground state as predicted by LDA + U calculations, or an AFM insulating (AFM-I) one which is more consistent with the μSR experiment, the system would have a FM metallic (FM-M) or an AFM metallic (AFM-M) region in between the thin-LNO limit ($m \leqslant 2$), and the bulklike thick-LNO limit ($m \geqslant 4$; PM and metallic, PM-M).[10] One may also expect the PM insulating (PM-I) phase stabilized in the same region of m: the intermediate regime, $2 \leqslant m \leqslant 4$ (see Fig. 2). As the LDA + U method is unable to properly describe correlated PM solutions,[36,37] a further pursuit along this line is beyond the scope of our study. For that purpose, one may resort to the dynamical mean-field theory (DMFT) calculations with charge self-consistency,[36,37] and compare the total energy for PM, FM, and AFM configurations as we did in this study within the LDA + U scheme.

To understand the metal-insulator phase transition as a function of LNO thickness,[10] we examined Ni e_g DOS depending on the thickness m. The d-bandwidth change as

FIG. 2. (Color online) The suggested schematic phase diagram (based on the previous experiment and our calculation) of the possible ground-state configuration of $(LNO)_m/(LAO)_n$ as a function of m (see the text).

a function of m may be useful information as U/W plays an important role in the metal-insulator phase transition of rare-earth nickelate.[38] Figure 3 shows the DOS of the PM case with $U = 0$. Even if the bulk LNO locates at the vicinity of the metal-insulator phase boundary and exhibits correlated electron behaviors, the electronic structure of PM LNO has been reasonably well described within the LDA [or the generalzied gradient approximation (GGA)], as shown in previous studies.[14,38,39] Figures 3(a)–3(d) present the evolution of e_g DOS as a function of m. It is noted that the $d_{3z^2-r^2}$ bandwidth notably changes, whereas the $d_{x^2-y^2}$ width remains almost the same across $m = 2$–4. In the $m = 2$ case [Fig. 3(b)], which was reported to be magnetic and insulating,[10] the right edge of the $d_{3z^2-r^2}$ state is fairly similar to that of $d_{x^2-y^2}$, whereas the left edge is reduced. It is not certain, however, that such a relatively small difference in terms of the effective bandwidth can make a significant change in the physical parameter (e.g., p-d hoppings), and lead to a metal-insulator transition (if one performs some other calculations such as DMFT on top of it) It is also found that the $3z^2 - r^2$ states at around -4 eV are gradually reduced as m increases, which is an indication of the reduced hybridization along the out-of-plane direction.

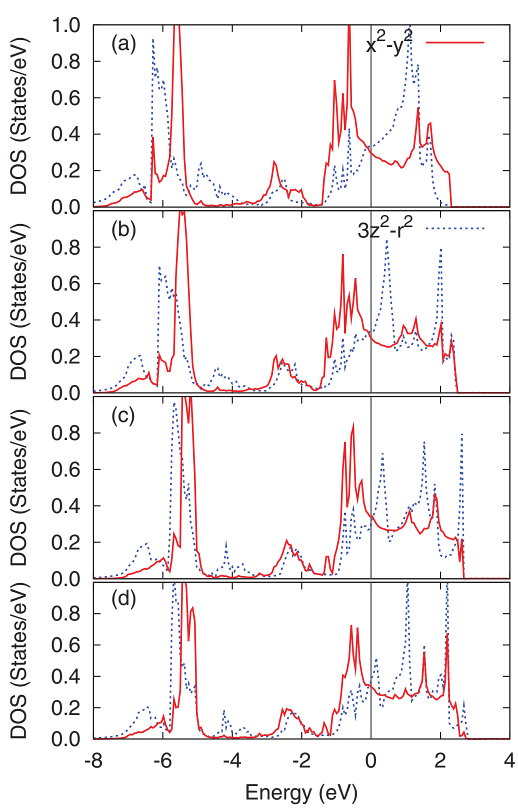

FIG. 3. (Color online) Ni e_g DOS of $(LNO)_m/(LAO)_1$ superlattice geometry with PM spin ($U = 0$): (a) $m = 1$, (b) $m = 2$, (c) $m = 3$, and (d) $m = 4$. Solid (red) and dotted (blue) lines correspond to $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ states, respectively, and the Fermi level is set to be 0 (vertical lines).

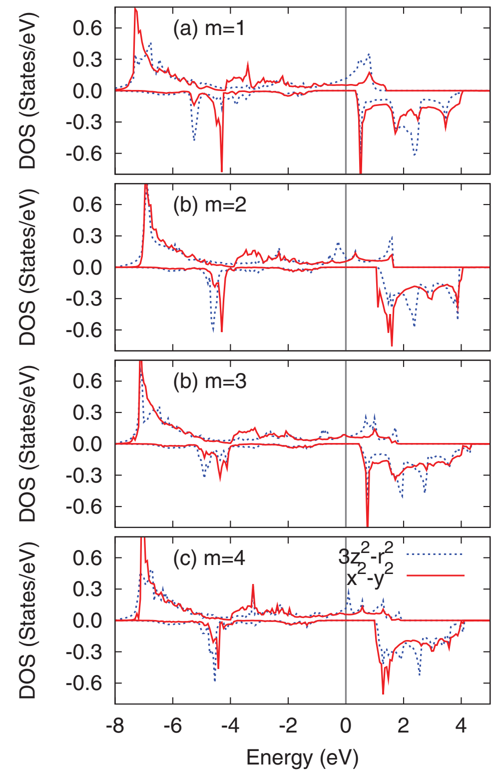

FIG. 4. (Color online) Ni e_g DOS of $(LNO)_m/(LAO)_1$ superlattice geometry with FM spin ($U = 5$ eV): (a) $m = 1$, (b) $m = 2$, (c) $m = 3$, and (d) $m = 4$. Up (down) panels represent the up (down) spin states. Solid (red) and dotted (blue) lines correspond to $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ states, respectively, and the Fermi level is set to be 0 (vertical lines).

A complementary picture can be provided by LDA + U calculations for the magnetic phase. Figures 4(a)–4(d) show the evolution of e_g DOS as a function of m for the FM $(LNO)_1/(LAO)_1$. Once again the notable change is found in the $d_{3z^2-r^2}$ band; for $m = 1$, the up-spin $d_{3z^2-r^2}$ state (upper panel) forms a fairly localized DOS around the Fermi level. This state becomes more and more delocalized as m increases, as seen in Figs. 4(b)–4(d). Once again, however, the amount of the effective bandwidth change depending on m does not seem to be enough to make a metal-insulator phase transition across $m = 2$–4. Origins other than the simple U/W change may be more relevant to the transition.[21,32]

An interesting point observed in Fig. 4 is that, even in the LDA + U calculation, the systems do not become a perfect insulator, but have a finite number of states around the Fermi level.[40] It may partly be attributed to the strong covalency between Ni d and O p states. As O p is much less affected by U, the small amount of DOS is not removed perfectly. Another important factor is the double-counting energy correction for which several functional forms have been suggested,[18–20,25,26] but, to the best of our knowledge, there is no well-defined solution as of yet:[21] As the double-counting energy is typically represented by $\frac{1}{2}UN_d(N_d-1)$, the LDA + U charge self-consistency adjusts N_d (or the effective d-level energy and therefore the charge-transfer energy $\epsilon_p - \epsilon_d$), depending on U. According to a recent DMFT study, which tunes the double-counting term as a parameter, the system eventually becomes insulating at large U.[15,41] Therefore the small states around the Fermi level obtained by LDA + U can be attributed to the double-counting error that is hardly handled within the current formalism of LDA + U.

An interesting recent finding on (three-dimensional) nickelates is that the metal-insulator transition occurs at a very narrow region of N_d and the same holds for the cuprates.[21] From the single-site DMFT calculations with a double-counting energy as another tuning parameter, Wang *et al.*[21] showed that there is a well-defined N_c, that is, the critical value of the d valency of a transition metal; the system is metallic if $N_d \geqslant N_c$ and insulating if $N_d \leqslant N_c$. For nickelates, $N_c^{e_g} \approx 1.3$. That is, for the parameters (implicitly including the double-counting correction) which result in $N_d \geqslant N_c$, the system remains metallic even for the very large U. This conclusion suggests N_d as the critical variable for understanding charge-transfer systems.[21] Now it might be instructive to analyze our LDA + U results within this unique picture of Ref. 21. Even if LDA + U is a lower-level approximation compared to DMFT, a large merit of LDA + U is that it can be performed with the whole charge self-consistency and take magnetism into account while the previous DMFT calculations have dealt with the PM phase.[12,15] Figure 1(d) presents N_d for PM ($U = 0$) and FM ($U = 3,5,7$) calculations of $(LNO)_{m=1}/(LAO)_1$ and $(LNO)_{m=2}/(LAO)_1$. While N_d decreases as U increases, the difference is not significant: $\Delta N_d \approx 0.15$. Therefore the current implementation of LDA + U and the charge self-consistency roughly follow the constant N_d line. Now we note that the value of $N_d^{e_g} \geqslant 2.2$ is far from the $N_c \approx 1.3$ predicted by DMFT. Therefore it is consistent with the metallic ground state and supports the conclusion of Ref. 21.[42]

A relative orbital occupation in the two e_g orbitals is a central quantity to understanding LNO/LAO systems.[9,12,14,15] The orbital polarization can be nonzero due to the translation symmetry breaking induced by heterostructuring while the bulk polarization is 0. According to previous calculations, highly polarized P can possibly drive the system to be a high-T_c superconductor.[12,17] On the other hand, a more recent DMFT calculation based on the realistic model Hamiltonian including oxygen orbitals predicted that the polarization is actually reduced by the on-site correlation.[15] Since both of the previous DMFT calculations assumed a PM phase and did not consider the full charge self-consistency, the LDA + U calculation can give complimentary information for the magnetic solution in spite of its limitation in describing the correlation effect compared to DMFT. The result is presented in Fig. 5. It is noted that the inclusion of U significantly reduces the polarization. There is a large separation between the $U = 0$ and 3 results, while the differences between the $U = 3$ and 5 results are small. Importantly, the calculated polarizations for

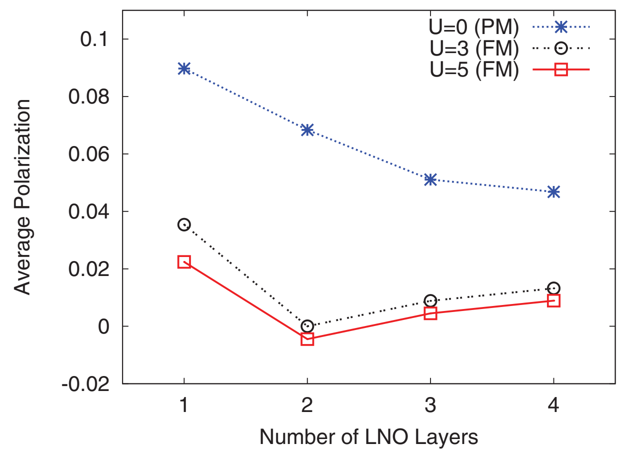

FIG. 5. (Color online) The averaged orbital polarization as a function of LNO layer thickness. Dotted (blue, double cross), double-dotted (black, circle), and solid (red, square) lines represent $U = 0$ (PM), $U = 3$ (FM), and $U = 5$ (FM), respectively.

finite U are an order of magnitude the same compared with the recent DMFT results.[15] Therefore it supports the conclusion of the recent DMFT calculation.[15] The small polarization at finite U can also be seen in the DOS presented in Fig. 4, where no large difference can be found between the two e_g orbital occupations. We note that the reduced orbital polarization by U can be compatible with the ferromagnetic spin order. For cuprates, for example, the fully polarized $x^2 - y^2$ orbital is directly related to the in-plane AFM spin order. In the nickelates, we have one more orbital degree of freedom available and the spin can align ferromagnetically. And actually it is found that the enhanced ferromagnetic trend by increasing U corresponds to the reduced orbital polarization as shown in Fig. 5.

IV. SUMMARY

Using the band-structure calculations based on LDA + U, we examined the magnetic moment, electronic structure, d valence, and orbital polarization as a function of U and m (LNO thickness). The calculated results clearly showed the formation of a magnetic moment at the finite U region being consistent with a recent μSR, but the long-range-ordering pattern is not so clear considering the experiment. While the $d_{3z^2-r^2}$ bandwidth is reduced as m approaches to 1, it may not be enough to be responsible for the metal-insulator transition. The calculated orbital polarization is significantly reduced by U, strongly supporting the conclusion of a recent DMFT calculation and indicating the absence of high-temperature superconductivity in this system.[43]

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[40]We found that this feature is retained up to $U = 9$ eV, and for the larger U, the charge convergency is not well stabilized.
[41]It should be noted that the effective U values, used in the self-consistent LDA + U calculation and in the effective model DMFT one, cannot be directly compared.
[42]The value of $N_c \sim 1.3$ in Ref. 21 is from the cubic LNO calculation. For the case of a superlattice, the almost same value of N_c has been found in the previous calculations of Ref. 15
[43]We found a closely related study has recently been published: A. Blanca-Romero and R. Pentcheva, Phys. Rev. B **84**, 195450 (2011). This work is also trying to address the metal-insulator transition and magnetic phase in the same type of superlattice systems using the GGA + U calculation. A notable difference is that the tilted oxygen octahedra structure has been used in that paper.