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Ab initio study of the electronic structure of $Sr_2FeMoO_6/SrTiO_3$ multilayers with Fe deficient interfaces

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Abstract

The electronic structure and the magnetic properties of $Sr_2FeMoO_6/SrTiO_3$(SFMO/STO) multilayers using the Full Potential Linearized Augmented Plane Wave method are investigated in order to discuss the role played by the Fe deficiency at the interface between SFMO and STO layers. The superlattice cell is built with (i) an STO layer consisting on five atomic layers for which the in plane lattice is taken equal to the one of bulk SFMO, (ii) a bulk-like SFMO layer consisting of seven atomic layers ending with Sr_2O_2 and (iii) an interfacial atomic layer consisting on $FeMoO_4$ (stoichiometric SFMO layer) or Mo_2O_4 (Fe deficient SFMO layer). The present calculations show that the interface has a very limited impact on the electronic and magnetic properties for the stoichiometric SFMO layer. On the other hand, the polarization of the evanescent states into the STO layer is strongly reduced and the gap into the majority spin band is recovered only for the most inner $FeMoO_4$ atomic plane of SFMO for the Fe deficient SFMO layer as compared to the other case.

Keywords: Band structure calculations; Metal–insulator–metal structures; Oxides; Superlattices; Surface and interface states

1. Introduction

Magnetic tunnel junctions (MTJ) using an half-metallic material – acting as an insulator for one spin channel and like a metal for the other one like Sr_2FeMoO_6 (SFMO) [1,2] – as magnetic electrodes are expected to exhibit very large magnetoresistance (MR) signals due to the high spin polarization of the injected current (theoretically, it should be 100% spin polarized). Indeed, with electrodes consisting in a half metal, the current flowing through the MTJ should be exactly equal to zero for antiparallel configuration of the magnetization of the two electrodes.

Epitaxial thin SFMO films have been deposited on $SrTiO_3$ (STO) substrates and it has been shown that they present properties similar to the bulk material [3–5] confirming the potential of this material. In order to investigate the current polarization due to a SFMO electrode, MTJ using an ultrathin STO film as barrier between SFMO and Co thin layers have been realized [6]. At low temperature, a clear positive magnetoresistive signal of 50% is obtained in a SFMO/STO/Co junctions yielding a negative spin polarization (SP) value of 85% for SFMO. This result shows that the SFMO/STO interface preserves the high polarization of the supposed half-metallic SFMO electrode. However, it is not clear if bulk SFMO has this 85% SP or if there is a lowering of the SP due to the interface with the STO layer.

Recently, several studies were focussed on $Sr_2FeMoO_6/SrTiO_3$(SFMO/STO) MTJ but the expected MR was not obtained even for well prepared samples [7]. By combining X-ray magnetic circular dicrhoism and X-ray photoemission spectroscopy, it has been shown that the surface magnetic moment of SFMO is anomalously weak consistent with a lack of Fe at the surface. The absence of MR is consequently ascribed to Fe deficient surfaces and interfaces of the SFMO layer where Fe atoms are replaced by Mo atoms and the polarization is strongly reduced. The aim of the present work is to investigate the electronic structure and the magnetic properties of SFMO/STO superlattices taking into account the interfacial Fe deficiency.

2. Unit cells and calculation method

In the [0 0 1] direction, SFMO consists of an alternance of $FeMoO_4$ and Sr_2O_2 atomic layers whereas STO consists of an alternance of TiO_2 and SrO atomic layers. Consequently, by doubling the in plane cell of STO using a $c(2 \times 2)$ cell, we get an alternance of Ti_2O_4 and Sr_2O_2 atomic layers simi-

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lar to the one of SFMO. In order to have the smallest possible SFMO/STO total cell and to insure that the inner atomic plane of each SFMO and STO layer has a bulk-like environment, the total cell for the stoichiometric case is build by a SFMO layer consisting of a 9 atomic layers $[FeMoO_4/Sr_2O_2]_4FeMoO_4$ stacking (denoted by $SFMO_9$) and of a STO layer consisting of $[Sr_2O_2/Ti_2O_4]_2/Sr_2O_2$ (denoted by STO_5); the total cell corresponds then to $SFMO_9/STO_5$. The Fe deficient cell consists of a $Mo_2O_4/[Sr_2O_2/FeMoO_4]_3/Sr_2O_2/Mo_2O_4$/STO stacking. The bulk in plane lattice parameter for both layers being very similar ($a_{SFMO} = 5.57$ Å, $a_{STO}\sqrt{2} = 5.52$ Å), the one of SFMO has been taken for the whole stack with a tetragonal distortion applied on the STO layer in order to preserve the cell volume. For the interfacial Fe deficient case, the interfacial Fe atoms are replaced by Mo atoms.

We calculate self-consistently the band structure for the considered system in the full potential augmented plane wave formalism (FLAPW) in the FLEUR implementation [8] using the GGA + U method and taking core, semi-core and valence states into account. This approach has been recently applied to bulk SFMO and Sr_2CoMoO_6 [9,10] where it has been exhibited that the GGA + U method is required in order to get a good agreement with experiments. Self-consistency has been obtained using 16 special k-points in the irreducible Brillouin zone and a few hundred of k-points have been used to determine densities of states projected on each atomic layer displayed in Figs. 1 and 2.

3. Stoichiometric case

For this case, the SFMO/STO superlattice is entirely half-metallic: as displayed in Fig. 1, there are no states available around the Fermi level in the majority spin DOS. Only electrons of the minority spin band can flow through this superlattice which corresponds to the parallel configuration of the magnetization of the two electrodes. For the antiparallel configuration, the gap in the majority band of the electrode with positive magnetization will present some states induced by the minority states of the next electrode with negative magnetization through the thin STO layer and reciprocally. Consequently, SFMO is no more strictly half-metallic but the current is certainly extremely weak as compared to the one in the parallel configuration because each electrode acts as an insulator in its bulk for one or the other spin channel.

The interfacial $FeMoO_4$ DOS (Fig. 1d.) is found very similar to the one of the most central $FeMoO_4$ atomic layer (which can be considered as bulk-like). This very limited impact of the interface on the properties of the SFMO layer is also reflected in the magnetic moments profile which shows nearly no variation when considering Fe or Mo atoms from the interfacial to the central atomic planes ($M_{Fe} = 3.99, 3.96, 3.97\ \mu_B$ and $M_{Mo} = -0.40, -0.40, -0.37\ \mu_B$ successively).

Consequently, the STO layer, terminated by the SrO atomic plane, has a weak impact on the electronic structure and the magnetic properties of the SFMO layer. This shows clearly that "perfect" SFMO/STO multilayers are a good candidate for spintronic devices.

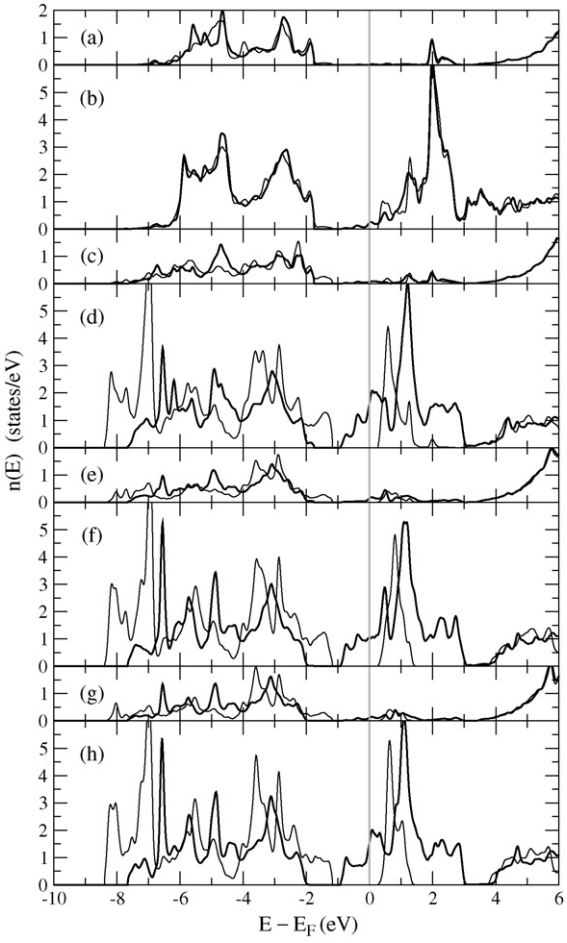

Fig. 1. Densities of states (DOS) projected on each inequivalent atomic layers (AL) for the stoichiometric case: (a) central Sr_2O_2 AL, (b) Ti_2O_4 AL, (c) interfacial Sr_2O_2 AL, (d) interfacial $FeMoO_4$ AL, (e) Sr_2O_2 AL, (f) $FeMoO_4$ AL, (g) Sr_2O_2 AL and (h) central $FeMoO_4$ AL. The majority $n^{(+)}(E)$ (minority $n^{(-)}(E)$) spin DOS corresponds (respectively) to the thin (thick) line.

4. Interfacial Fe deficient case

Replacing the interfacial Fe atoms by Mo atoms has clearly a strong impact. In previous works on bulk SFMO presenting imperfections [10,11], it has been shown that the half-metallic property is lost when Mo antisite are introduced by substituting an Fe atom ($Sr_4FeMo_3O_{12}$ cell) and that the local moment on the Mo antisite has an opposite sign ($+0.26\ \mu_B$) as compared to the one on the regular Mo sites ($-0.39\ \mu_B$). However, this case corresponds to a bulk situation with a high concentration of Mo antisites (half of the Fe sites are occupied by Mo) leading to an overestimation of the role played by such antisites.

As displayed by Fig. 2, the electronic structure of the SFMO/STO superlattice with Fe deficiency at the interface shows significant differences as compared to the "perfect" case. The half-metallic property is lost for the whole cell and the spin polarization of the local DOS at the Fermi level, defined by $P = (n^{(+)}(E_F) - n^{(-)}(E_F))/(n^{(+)}(E_F) + n^{(-)}(E_F))$, even vanishes completely on the interfacial atomic planes (from the a to the h atomic layer, P is equal to $-0.33, -0.21, +0.01, +0.04, -0.37, -0.54, -0.54, -0.99$). Consequently, for this case, the