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Magneto-electric coupling in antiferromagnet/ferroelectric $\text{Mn}_2\text{Au}/\text{BaTiO}_3$ interface

Evgeny Plekhanov,¹ Alessandro Stroppa,² and Silvia Picozzi³

¹*King's College London, Theory and Simulation of Condensed Matter (TSCM),
The Strand, London WC2R 2LS, United Kingdom*

²*Consiglio Nazionale delle Ricerche, Istituto SPIN,
UOS CNR-SPIN L'Aquila, I-67100 L'Aquila, Italy*

³*Consiglio Nazionale delle Ricerche, Istituto SPIN, UOS L'Aquila,
Sede di lavoro CNR-SPIN c/o Univ. "G. D'Annunzio", 66100 Chieti, Italy*

Within the crucial issue of the electric field control of magnetism, the use of antiferromagnets coupled to ferroelectrics is much less explored than the ferromagnets counterpart, although the first choice might lead to better performances and larger stability with respect to external perturbations (such as magnetic fields). Here we explore the possibility to control the magnetic anisotropy of a Mn_2Au layer by reversing the ferroelectric polarization of BaTiO_3 in $\text{Mn}_2\text{Au}/\text{BaTiO}_3$ interfaces. By means of a thorough exploration of many possible geometry configurations, we identify the two most stable, corresponding to compressive and tensile strain at the interface. The first appears to be easy-axis while the second – easy-plane, with a large induced moment on the interface Ti atom. The reversal of ferroelectric polarization changes the anisotropy by approximately 50%, thus paving the way to the control of AFM properties with an electric field.

I. INTRODUCTION

The next generation of random access memory (RAM) devices will necessarily have to overcome the problem of high power consumption needed to maintain the written data, which amounts to approximately 99% of the total power consumed in current RAM. An innovative step in this direction might be taken by exploiting the so called “magneto-electric coupling” (MEC), based on the interaction between the magnetic and ferroelectric order parameters, either in a single phase multiferroic^{1–4} or in a ferroelectric/ferromagnetic heterostructure^{5,6}. Thanks to the MEC, it could be possible to achieve a magnetic reading-process and electrical writing process of the information bits, therefore avoiding energy dissipation coming from significant currents on a small scale and allowing for higher bit density. In addition, the use of ferroelectric materials instead of dielectric ones in the Ferroelectric (FE) RAM (FERAM) dramatically increases the time during which the cell conserves its charge, hence reducing the power consumption. Ferromagnets, usually employed in devices based on MEC, are, however, subject to external magnetic fields. That is why it was recently proposed to use antiferromagnets (AFMs)^{7,8}, which have the advantage of being roughly insensitive to external magnetic fields, while keeping non-volatility. A number of candidate systems were proposed in the past for the electric control of antiferromagnetism: i) exchange-spring^{7–9} or field-cooling¹⁰ reversal of IrMn AFM magnetization, subsequently detectable by a change in tunneling current; ii) electric field switching of the IrMn magnetization through immersion in an ionic liquid¹¹; iii) BaTiO_3 polarization reversal which induces a FM-AFM switch in Fe^{12} or FeRh^{13} thin films.

Coupling ferroelectricity to the magneto-crystalline anisotropy (MCA) might represent an alternative way to electrically control the magnetic properties^{14–16}. For example, one might envisage controlling or even switch-

ing the difference in energy between easy and hard axis (i.e. the magnetic anisotropy energy, MAE) of the magnetic layer as a function of the direction of polarization in the underlying FE substrate. To our knowledge, this coupling between FE and AFM is rather unexplored in the literature, but could lead to significant advantages. Indeed, changing the orientation of the AFM quantization axis (e.g. from in-plane to out-of-plane and viceversa) is a serious technological challenge and would definitely benefit from a possible FE modulation of the MCA, thus facilitating the *electrical* writing process. Successively, the read could be accomplished e.g. via Tunneling Anisotropic Magneto-Resistance (TAMR)¹⁰.

Within this framework, in this manuscript we propose a new interface, *i.e.* [001]-oriented $\text{Mn}_2\text{Au}/\text{BaTiO}_3$, and explore theoretically the MCA modulation upon BaTiO_3 (BTO) polarization switch. As magnetic layer, we considered Mn_2Au , as it was recently shown to be an antiferromagnet with a strikingly high estimated Neel temperature (more than 1000K)^{17,18}. Owing to the strong spin-orbit in Au, it possesses also a sizable MCA, which results in a significant TAMR^{19–21}. We identify the most stable interface for each of the tensile and compressive mismatch types and show that the BTO polarization switch modulates the MCA energy barrier by approximately 50%, when going from the more to the less stable orientations of the sublattice magnetization.

The article is organized as follows: in Sec. II we analyze the magnetic and structural properties of a large number of theoretical $\text{Mn}_2\text{Au}/\text{BaTiO}_3$ interfaces, identifying among them the two most stable configurations. In Sec. III we study the MCA modulation upon polarization switch. Finally, the main results are summarized in Sec. IV.

II. SIMULATION RESULTS

Due to the large lattice mismatch between BTO and Mn_2Au , as discussed in detail in the following paragraph, we addressed the ultrathin limit of Mn_2Au films grown on BaTiO_3 .

The stability range of BTO with respect to the in-plane lattice mismatch was found in Ref.²² to be $\pm 6\%$. The same range for Mn_2Au is difficult to estimate, although in Ref.²⁰ it was shown that thin films of $\text{Mn}_2\text{Au}/\text{Fe}$ were grown on MgO substrate. Assuming the MgO lattice parameter to be $a_{\text{MgO}} = 4.212 \text{ \AA}$ while the experimental²³ $a_{\text{Mn}_2\text{Au}} = 3.328 \text{ \AA}$, we can estimate that in that case the lattice mismatch was either -26.6% (direct match) or $+10\%$ (45° rotated match). In the $\text{Mn}_2\text{Au}/\text{BTO}$ case, considering the bulk thick layer of BTO with the in-plane lattice constant of the order of $4 \text{ \AA} \pm 6\%$, the direct and 45° rotated matches might be at the best -12% and $+10\%$ respectively. Given the high lattice mismatch, we only focus on the ultrathin limit of Mn_2Au films, *i.e.* half-unit-cell.

In order to find out the most stable configuration of the interface between Mn_2Au and BTO, we examine a large number of possible interfaces. We consider three unit cells of barium titanate (with the standard tetragonal distorted-perovskite cell) and a half-cell of Mn_2Au (HCMA). Mn_2Au crystallizes in the tetragonal $I4/mmm$ space group constituted by planes of Au alternated to Mn bilayers. From the magnetic point of view, the ground state shows planes of ferromagnetically aligned Mn atoms, with the adjacent planes coupled antiferromagnetically. On the barium titanate side we consider the TiO_2 termination, normally found as the most stable occurring at interfaces of BTO with metals^{12,24}. In order to explore all the possibilities, we consider three terminations of HCMA: i) Au; ii) Mn and iii) Mn_2 . In addition, each HCMA termination has two possibilities to stack on the TiO_2 layer, *e.g.* for the Au termination, Au atoms can occupy either Ba or Ti position. Finally, we consider both direct and rotated by 45° match, *i.e.* corresponding to tensile and compressive strain. Summarizing, for each direction of BTO polarization ($\uparrow$ and $\downarrow$, corresponding to polarization directed outwards or inwards with respect to the BTO layer), given three possible terminations on HCMA, two different stacking geometries and two different strain conditions, we consider $3 \times 2 \times 2 = 12$, which leads to a total of 24 simulated configurations (including polarization switching). The schematic view of the 12 configurations at fixed BTO polarization is shown in Fig.1. In order to avoid the loss of ferroelectric (FE) polarization during the structural optimization (which might spuriously arise, due to the limited number of BTO unit cells), we used a sort of “constrained” atomic optimization: the farthest layer from the interface are forced to have the same structure as bulk BTO, whereas the layers closer to the junction are allowed to relax. We fix the in-plane BTO lattice constant to its experimental value $a = b = 3.991 \text{ \AA}$.

Figure 1. (Color online) Scheme of 12 possible interfaces between BTO and Mn_2Au . The interfaces 1–3 and 7–9 feature the Mn_2Au subject to tensile strain, while the interfaces 4–6 and 10–12 have Mn_2Au under compressive strain. The Roman numbers label the interface layers, in going from TiO_2 towards Mn_2Au . Ti, O, Mn and Au atoms are shown by means of light-blue, red, yellow and purple spheres.

We perform density functional theory (DFT) simulations using the Vienna Ab initio Simulation Package (VASP)²⁵ and the Generalized Gradient Approximation (GGA)²⁶ in the Perdew-Burke-Ernzerhof (PBE) formalism for the exchange-correlation potential. We use an energy cutoff for the plane wave basis of 500 eV and a $12 \times 12 \times 1$ Monkhorst-Pack k -point mesh²⁷, while the structural optimization is accomplished until the ionic forces become less than $0.01 \text{ eV/\AA}$. In order to evaluate the magneto-crystalline anisotropy, the relativistic spin-orbit coupling (SOC) is self-consistently taken into account in the usual perturbative way. Simulations are done by resorting to the slab geometry, with $20 \text{ \AA}$ of vacuum added along the z -axis in order to avoid the self-interaction through the periodic boundary conditions. In addition, since the BTO layer has a net dipole moment oriented along the z axis, the dipole corrections, as implemented in VASP, were used, so as to make any possible electric field in the vacuum region far from the slab to vanish.

As mentioned, by construction all the interfaces can be divided into two groups, when referring to the strain state: compressive and tensile. The absolute values of the lattice mismatch in both cases are comparable, although the number of Mn_2Au atoms is different in the two strain cases (cfr Fig. 2 below). Therefore, we identify the lowest energy configuration within each group for each direction of polarization $P_{\text{BTO}(\uparrow/\downarrow)}$. The comparison of the interface properties is summarized in Tabs. I, II. Here, the interfaces 1–3 and 7–9 are under tensile strain, while the junctions 4–6 and 10–12 are under compressive strain. It can be seen from Tab. I that the properties of the system crucially depend on the HCMA termination. Remarkably, interface 3, IF-3, the most stable in the tensile group -

Figure 2. (Color online) Side view of the most stable interfaces under tensile and compressive strain and with different ferroelectric polarization direction for the BTO polarization: *i*) IF-3 with $P_{BTO\downarrow}$ (a) and $P_{BTO\uparrow}$ (b) and *ii*) IF-5 with $P_{BTO\downarrow}$ (c) and $P_{BTO\uparrow}$ (d). The blue arrow shows the direction of ferroelectric polarization in BTO.

presents a huge magnetic moment transfer on the interface Ti atom - larger than $0.5\mu_B$. This moment transfer reduces to $0.3\mu_B$ when the polarization is reversed, but it is anyway much larger than it was predicted in similar Fe/BTO interfaces²⁸. Among the compressive group, the lowest energy is found at interface 5 (IF-5), while the energy difference with respect to the next-lowest energy configuration (interface 6) is larger than 1 eV, being the latter much greater than the corresponding difference in the tensile group (0.15eV).

The ground state Mn magnetic pattern in the compressive group is different from bulk Mn_2Au : rather than alternating ferromagnetic layers, we find an alternation of the AFM Mn layers. The stability of this pattern is probably due to the reduced thickness of the Mn_2Au layer. The tensile interfaces exhibit somewhat larger Mn moments, when compared to the compressive case. This difference is a consequence of the geometrical frustration present in the compressive interfaces with AFM boundary Mn layers. Indeed, a large hybridization between Mn and Ti atoms contributes cooperatively to the stabilization of a large induced magnetic moment on Ti, when the Mn moments are parallel, but tend to lower the Mn moments, when they are antiparallel, with a side effect to nullify the Ti induced moment.

The pattern of the Ti-O displacements depends on the interface type as well. In general, the displacement of the interface Ti with respect to the surrounding planar oxygen atoms is such as to move away from the interface (being at most close to zero, *i.e.* as for IF-5) for both $P_{BTO\uparrow}$ and $P_{BTO\downarrow}$ and for all interface types. $P_{BTO\downarrow}$ strongly enhances this displacement so as to even exceed, for certain interface types, the bulk displacement of BTO adopted in our calculations for the third “frozen” Ti layer (having a Ti-O displacement of 0.222 Å). The off-centering of the middle Ti-O layer is almost always negligible for $P_{BTO\uparrow}$, while enhanced for $P_{BTO\downarrow}$.

Table I. Relevant properties of the interfaces with $P_{BTO\uparrow}$ (collinear calculations without SOC, dipole corrections included). The energy difference ΔE_{tot} (in eV per unit cell, second column) is measured relative to the lowest energy configurations in each group (IF-3 and IF-5 respectively, highlighted in bold). The magnetic moment μ is shown for interface Ti and Mn atoms (third and fourth column, respectively) and measured in units of μ_B . The interplanar distance between Ti and planar O-atoms (in Å, fifth column).

IF	ΔE_{tot} (eV)	μ (Ti ₁ & Ti ₂)		μ (Mn _i)		Ti-O displ. (Å)	
1	1.800	-0.07,	-0.17	-4.07,	4.02	-0.12,	0.00
2	1.865	0.08,	0.14	-4.07,	4.09	-0.14,	0.01
3	0.000	0.59,	0.35	-3.89,	3.93	-0.14,	-0.01
4	2.177	0.15,	0.20	3.60,	-3.56	-0.09,	0.01
				-3.25,	3.38		
5	0.000	-0.09,	-0.02	3.23,	-3.12	-0.01,	0.06
				-2.32,	2.83		
6	1.086	0.00,	0.00	3.26,	-3.26	-0.09,	0.03
				-3.74,	3.74		
7	1.822	-0.04,	-0.02	-4.07,	4.07	-0.08,	0.04
8	0.166	-0.61,	-0.41	-4.05,	4.02	-0.20,	-0.02
9	1.926	0.06,	0.22	-3.94,	3.90	-0.16,	0.00
10	2.330	0.02,	0.00	3.54,	-3.51	-0.03,	0.06
				-3.30,	3.35		
11	1.887	-0.21,	-0.01	2.58,	-2.76	0.10,	0.11
				-0.04,	3.79		
12	2.855	-0.52,	-0.34	4.00,	-3.77	-0.18,	-0.02
				-3.76,	3.81		

Table II. Relevant properties of the interfaces with $P_{BTO\downarrow}$ for the most stable configurations, *i.e.* IF-3 and IF-5 (collinear calculations without SOC, dipole corrections included). For the $P_{BTO\uparrow}$ values see rows in bold, as reported Tab. I. The magnetic moment μ is shown for interface Ti and Mn atoms (third and fourth column, respectively) and measured in units of μ_B . The interplanar distance between Ti and planar O-atoms (in Å, fifth column).

IF	μ (Ti ₁ & Ti ₂)		μ (Mn _i)		Ti-O displ. (Å)	
3	0.30,	0.02	-3.94,	3.94	-0.24,	-0.21
			3.27,	-3.09		
5	0.01,	0.00	-2.54,	3.03	-0.19,	-0.16

III. DISCUSSION

A. Magneto-electric coupling

As was mentioned above, compressive and tensile interfaces show very different magnetic properties. Namely, in their respective ground-state configurations, the interface Ti atom in IF-3 bears a large induced magnetic moment, while in IF-5 it is almost non-magnetic. The moment of Ti is caused by the hybridization with surrounding Mn moments, and it is the positions and orientations of these Mn atoms that the magnitude of the Ti moment depends on. In IF-3, the Ti-Mn distance is somewhat larger than in the IF-5 (3.13 Å vs 2.77 Å), however, in the IF-3, the most stable magnetic configuration has the nearest-

Table III. Total energies obtained by changing the spin quantization axis for the most stable interfaces in the compressive and tensile strain (IF-3 and IF-5) as a function of polarization switching. The arrows in the second and fourth columns are related to the orientation of Mn moment and have the following meaning: $\uparrow$ - perpendicular to the interface; $\rightarrow$ - parallel to the interface along the a -axis; $\nearrow$ - parallel to the interface and diagonal in the $a - b$ plane. The total energy difference ΔE_{tot} is measured relative to the lowest energy configuration (highlighted in bold), for each of the strain and polarization conditions.

P_{BTO}	μ	$ \Delta E_{tot}(\text{meV}) $	μ	$ \Delta E_{tot}(\text{meV}) $
$P_{\uparrow}$	IF-3 $\uparrow$	0.86	IF-5$\uparrow$	0.00
	IF-3 $\rightarrow$	0.11	IF-5 $\rightarrow$	1.37
	IF-3$\nearrow$	0.00	IF-5 $\nearrow$	2.46
$P_{\downarrow}$	IF-3 $\uparrow$	2.55	IF-5$\uparrow$	0.00
	IF-3$\rightarrow$	0.00	IF-5 $\rightarrow$	2.36
	IF-3 $\nearrow$	0.02	IF-5 $\nearrow$	2.17

neighbor Mn moments parallel, while in the IF-5 they appear to be antiparallel. Therefore, the contributions to the induced magnetic moment on Ti atom, coming from the two Mn atoms cancel out in the case of IF-5. Focusing on IF-3, we conclude that the switch of P_{BTO} leads to a change of the interface magnetic moment of the order of $0.3\mu_B$. This value is comparable to that calculated in Ref.²⁸ for the Fe/BTO interface. Let us now recall the definition of the surface magneto-electric coefficient, α :

$$\alpha = \frac{\mu_0 \Delta M}{E_c},$$

where ΔM is the increment of the magnetic moment (per 1 BTO unit cell) and E_c is the BTO coercive field. Being BTO in common between Ref.²⁸ and the present work and being ΔM comparable when BTO is interface with Fe or Mn_2Au , the surface magneto-electric coefficient appears to be of the same order of magnitude. This is a rather unexpected result: an interface coupling between a ferroelectric and an antiferromagnet can generate a sizable magneto-electric coupling with a net interface magnetization.

B. Magnetic anisotropy

The results obtained when considering different spin-quantization axes, aimed at evaluating the MCA and its dependence on the direction of BTO ferroelectric polarization, are reported in Tab. III. As evident, IF-3 shows an easy-plane anisotropy, while the IF-5 is characterized by an easy-axis. Upon switching the BTO polarization, the MCA properties of the IF-3 and IF-5 are sizeably affected: for the IF-3 the easy-plane anisotropy changes from 2.6 meV at $P_{BTO\downarrow}$ to 0.8 meV at $P_{BTO\uparrow}$, while for the IF-5 the easy-axis anisotropy varies from

2.2 meV at $P_{BTO\downarrow}$ to 1.4 meV at $P_{BTO\uparrow}$. It follows that the MCA changes by almost 50% upon polarization switch. It is important to note that in both interfaces the $P_{BTO\uparrow}$ polarization corresponds to lower MCA and that these MCA energies, if measured per formula unit of Mn_2Au , are comparable to the MCA of the bulk Mn_2Au ¹⁹ (1.22 meV).

IV. CONCLUSIONS

In this manuscript, we have presented an investigation of the $\text{Mn}_2\text{Au}/\text{BTO}$ interface by considering a large number of interface structural configurations and identifying the most stable junctions under compressive and tensile strain. By a thorough study of their magnetic and ferroelectric properties we show an example (*i.e.* in the IF-3 interface) of a FE-AFM coupling which generates a net FM magnetization at the interface tuned by the FE polarization switch. The FE polarization also modulates the magnetocrystalline anisotropy within the range of approximately 50%. Interestingly, this is an example of an interface containing an AFM ingredient which generates a net magnetic moment in the layer close to the junction. This interface magnetization is strongly affected by the direction of P_{BTO} , *i.e.* with a change of 100%. The magnetic anisotropy of HCMA is also modulated by the P_{BTO} switch, with a change of approximately 50%. Different interfaces (under compressive and tensile strain conditions) could be realised by changing the growth conditions of Mn_2Au on BTO, *i.e.* by varying the lattice spacing of BTO using different substrates.

The physical mechanism, responsible for the MEC presented here, has to be ascribed to a sizable Mn-Ti hybridization, which leads to a significant transfer of magnetic moment on Ti atoms. The switch of FE polarization implies a change of Mn-Ti distance and, hence, of the magnetic moment transfer, thereby reversing a very subtle balance of total energies, leading to magneto-crystalline anisotropy. The MEC studied in this manuscript is a surface effect. As such, we expect it to be most efficient for thin films of Mn_2Au . It should be noted, however, that experimentally, it is quite difficult to control the growth of Mn_2Au thin films, and that due to the richness of intermetallic phases formed by manganese and gold, the result might have a different stoichiometry: *e.g.* MnAu_2 or MnAu . The study of these possibilities goes beyond the scope of the present manuscript, although, thanks to the fact that both MnAu_2 or MnAu are antiferromagnets with interesting properties^{29–32}, once interfaced with BTO, they might well exhibit peculiar magneto-electric effects.

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