Switching Magnetization by 180° with an Electric Field

M. Fechner,[1,2,*] P. Zahn,[3,4] S. Ostanin,[1] M. Bibes,[5] and I. Mertig[1,3]
[1]Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany
[2]Department for Material Theory, ETH Zürich, CH-8093 Zürich, Switzerland
[3]Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, D-06099 Halle, Germany
[4]Helmholtz-Zentrum Dresden-Rossendorf, P.O. Box 51 01 19, D-01314 Dresden, Germany
[5]Unité Mixte de Physique CNRS/Thales, 1 Avenue Augustin Fresnel, Campus de l'École Polytechnique, 91767 Palaiseau, France, and Université Paris-Sud, 91405 Orsay, France
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Magnetoelectric coupling allows for manipulating the magnetization by an external electric field or the electrical polarization by an external magnetic field. Here, we propose a mechanism to electrically induce 180° magnetization switching combining two effects: the magnetoelectric coupling at a multiferroic interface and magnetic interlayer exchange coupling. By means of first-principles methods, we investigate a ferroelectric layer in contact with a Fe/Au/Fe trilayer. The calculations show that the interface magnetism is strongly coupled to the ferroelectric layer. Furthermore, under certain conditions a reversal of polarization causes a sign reversal of the interlayer exchange coupling which is results in a 180° switching of the free layer magnetization. We argue that this magnetoelectric coupling mechanism is very robust and can find applications in magnetic data storage.

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Functional materials are characterized by a robust response to external fields. For instance, the resistivity of magnetoresistive materials may be modified by external magnetic fields, as in mixed-valence manganites. However, the response may be larger (at room temperature) and easier to control in heterostructures combining several materials. A typical example is giant magnetoresistance (GMR) discovered by Grünberg and Fert [1,2] in magnetic multilayers. Although magnetic data storage technology currently uses tunneling magnetoresistance [3] (where the difference in resistance caused by the relative alignment of the layer magnetization, separated by an insulating barrier, is used to read the state of a magnetic bit), GMR drove a large increase of information density in magnetic memories by a factor of 4 and initiated the field of spintronics.

In the last 20 years of miniaturization of magnetic memory cells, it turned out that the reading procedure is scalable down to the nanometer range. However, the writing mechanism driven by a magnetic field appeared as a crucial limiting factor, due to a large power consumption and the magnetic stray fields that may affect neighboring cells. The introduction of current-induced magnetization switching [4], where a spin-polarized current leads to a torque on the magnetization, enabled further down scaling of the memory cells. Nevertheless, the required current density for switching is still large and can cause metal migration. Many new concepts appeared recently related to the magnetoelectric coupling [5] which allow for writing the information by the application of a voltage, with prospective power consumption lower by several orders of magnitudes compared to the state of the art.

In most of the proposed magnetoelectric systems, a rotation of the magnetization by 90° could be achieved. For instance, Pertsev and Kohlstedt [6] proposed a piezoelectric layer in combination with a ferromagnet/nonmagnet/ferromagnet (FM/NM/FM) trilayer in which the magnetization could be rotated in the adjacent ferromagnet by 90° due to magnetostriction [6]. Another approach exploits the magnetoelectric coupling in combination with exchange bias, showing a 90° [7] and recently even 180° [8] rotation in CoFe films on top of a $BiFeO_3$ layer. However, still in most cases the switching is limited to 90° so far or is assisted by a magnetic field or spin transfer torque. Any step beyond these limits would be an important breakthrough towards a new memory cell design.

In this Letter, we propose a hybrid structure for a magnetic 180° switch driven purely by an electric field. It consists of a ferroelectric (FE) thin film in contact with a FM/NM/FM trilayer as shown in Fig. 1(a). This type of heterostructure combines two types of coupling, magnetoelectric coupling at the multiferroic interface and interlayer exchange coupling (IEC) acting in the trilayer. If the heterostructure is well designed, it allows rotation of the magnetization in the layer by 180° under polarization switching. This process would allow for small magnetic solid state memory cells which are writable by an electric field and readable by the GMR effect. Furthermore, this design would require only small voltages, if the thickness of the FE layer is chosen sufficiently small. In the following, we first describe the microscopic modeling of the system. Second, the conditions for the 180° switching are derived. Finally, the influence of layer thickness variations is estimated to be small enough for a proper device performance.

Figure 1(b) shows the proposed hybrid structure at the atomic scale. As a layer, we consider $PbTiO_3$ (PTO), a

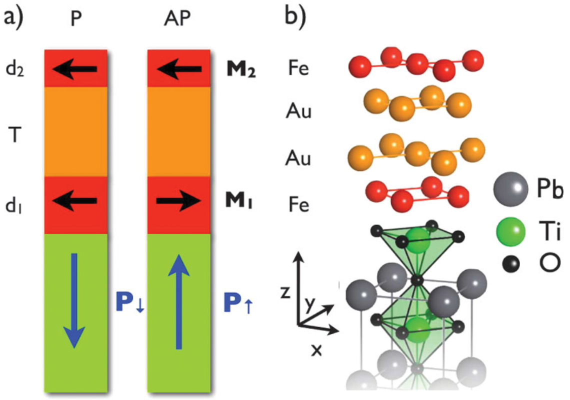

FIG. 1 (color). (a) Hybrid design to allow for switching the interlayer exchange coupling by polarization reversal. If the magnetoelectric coupling at the interface is strong enough, it allows controlling the alignment of magnetizations M_1 and M_2. (b) Hybrid structure on the atomic scale. It consists of a TiO_2 terminated $PbTiO_3$ layer covered by a Fe/Au/Fe trilayer, here with a thickness of 1, 2, and 1 atomic layer, respectively.

strong ferroelectric with a polarization of 0.95 C/m^2 [9]. The trilayer on top consists of Fe, Au, and Fe layers in a bcc, fcc, and bcc structure. We assume epitaxial growth on the PTO fixing the in-plane lattice constant to 3.88 Å. The PTO/Fe interface is TiO_2 terminated with the Fe atoms above the O, which was theoretically and experimentally confirmed [10–12].

To investigate the energetics of the switching process, we performed first-principles calculations based on density functional theory. For geometrical optimization we utilize the Vienna *ab initio* simulation package (VASP) [13] relying on the local density approximation and employing pseudopotentials based on the projector augmented wave method. Considering the fact that the energy differences of the IEC are in the meV range, we carefully checked convergence of our results. Finally, a $10 \times 10 \times 3$ k-point mesh for structural relaxation and a $19 \times 19 \times 3$ k-point mesh for determining the magnetic properties and total energies were used to sample the Brillouin zone. The cutoff energy is 700 eV, and we optimized the structure until the forces were smaller than 0.01 meV/Å. Besides the treatment in VASP, we apply the screened Korringa-Kohn-Rostoker method [14]. This allows for an $O(N)$ scaling for larger layer thicknesses. Furthermore, it permits for local shifts of the spin-dependent potentials to model local changes of charge and magnetization. The charge density is expanded up to angular momenta of $l = 3$, while the potentials are treated in the atomic-sphere approximation. Within the calculation we model the system by supercells including 5 unit cells PTO attached to the Fe/Au/Fe trilayer. The whole layer is separated by 5 nm vacuum from the next stack. We further employ the dipole correction [15] to suppress any interaction between the lowest FE layer and the metal surface. The thicknesses of the individual layers in the trilayer are denoted by $d1/T/d2$ in units of monolayers (ML) as depicted in Fig. 1(a) Since the IEC strongly depends on the three thicknesses, the parameter space was restricted to exemplary values. Furthermore, we denote the magnetizations of the two ferromagnetic layers by M_1 and M_2.

The electric polarization in PTO is characterized by the relative displacement $\delta = z_{\text{cation}} - z_{\text{oxygen}}$ of oxygen atoms and cations in each layer. A positive displacement corresponds to a polarization pointing towards the interface and is denoted as $P_\uparrow$, whereas $P_\downarrow$ is the case of negative displacement. The first three layers of PTO close to the FM (namely, $TiO_2/PbO/TiO_2$ layers) and the Fe/Au/Fe trilayer are relaxed, whereas the remaining PTO layers are fixed to keep the bulk polarization in positive or negative direction, respectively [10].

The structural changes in the trilayer are small during the relaxation. For the system 1/4/1, the Fe-Au interlayer distance shows an expansion of 5% with respect to the mean value of the layer distances in Fe and Au. In the same configuration, the Au layers experience an expansion of 0.5% with respect to the bulk value, which is in agreement by the compressive strain of the bottom layer. For Fe thicknesses of 2 ML, we obtain an interlayer compression of 16% and 13% for the top and bottom layer, respectively. This compression is lifted if the Fe thickness exceeds 2 ML. It is important to note that we did not observe a remarkable change of these values for the different polarization states of PTO. The most pronounced structural change occurs at the interface. For $P_\uparrow$ a moderate enhancement (about 30%) of the buckling of the Ti-O layer with respect to the free surface value is observed at the FE/FM interface, whereas it keeps nearly the free surface value for the opposite polarization $P_\downarrow$. This behavior is related to the missing Ti-O bond at the interface [9].

After structural relaxation, we explore the magnetic properties of the system. From the comparison of the total energies for different magnetic structures, we obtain that within each magnetic layer the ferromagnetic state is preferred independent on the electric polarization or Au thickness. Furthermore, the magnetization M_1 alters upon polarization reversal, due to the hybridization of Ti d states and Fe minority d states [10]. An induced moment of $-0.35\mu_B$ at Ti appears for $P_\uparrow$ and a negligible moment is induced for $P_\downarrow$. The Fe moments at the interface show a similar change from $2.4\mu_B$ for $P_\uparrow$ to $2.7\mu_B$ for $P_\downarrow$. Finally, this gives a net change of $\Delta M_1 = 0.45\mu_B$ per Fe site upon polarization reversal. The size of the magnetic moment M_2 of the top Fe layer is not affected by the electric polarization or the Au thickness as expected.

In the next step we determine the preferred relative alignment of the magnetizations in both FM layers. Since both Fe layers interact via IEC, an oscillation between parallel and antiparallel alignment of the two magnetizations as a function of Au thickness is expected [16]. To this end we perform total energy calculations where either M_1 and M_2 are aligned parallel (P) or antiparallel (AP) aligned. Furthermore, these calculations are performed for both polarization directions $P_\uparrow$ and $P_\downarrow$. For the energy difference we use the definition $\Delta E^{\uparrow(\downarrow)} = E_P^{\uparrow(\downarrow)} - E_{AP}^{\uparrow(\downarrow)}$; thus, a positive energy difference points to an antiferromagnetic coupling.

Figure 2 shows ΔE as a function of Au layer thickness T for polarizations $P_\uparrow$ and $P_\downarrow$ for Fe thicknesses $d1 = d2 = 1$ ML. For both polarizations we observe the expected oscillation as a function of Au layer thickness, while the two curves are split energetically by about 10 meV. Furthermore, the polarization $P_\downarrow$ favors P alignment, whereas $P_\uparrow$ favors AP alignment of layer magnetizations. Thus, we conclude that the orientation of the ferroelectric polarization strongly modifies the IEC energy. The most promising behavior is obtained for 1 and 4 ML thicknesses of Au. Here, we observe an AP alignment for $P_\uparrow$ and P alignment for $P_\downarrow$. Thus, the polarization direction determines the magnetic ground state. In the following, we explain these findings by a detailed analysis of the electronic and magnetic structure to map out the origin of the change of the IEC strength in this system.

To elucidate this very unique coupling phenomenon, we recall the origin of the IEC. The oscillation of the coupling strength of the two FM layers separated by a nonmagnetic spacer was first attributed to a Ruderman-Kittel-Kasuya-Yosida–like interaction [16]. Equivalently, it can also be understood as a quantum interference effect within the NM spacer material [17]. The formation of spin-dependent quantum well states leads to a variation of the density of states and thus a change in total energy. Finally, the energy difference between the P and AP alignment can be approximated by

$$E_P - E_{AP} \approx \frac{1}{\pi^3} \,\mathrm{Im} \int d^2k_\parallel \int_{-\infty}^{\epsilon_F} \Delta r_1 \Delta r_2 e^{2ik_\perp T} d\epsilon, \qquad (1)$$

where $k_\parallel$ and $k_\perp$ are the momentum parallel to the interface and perpendicular to it in the NM layer, respectively [17].

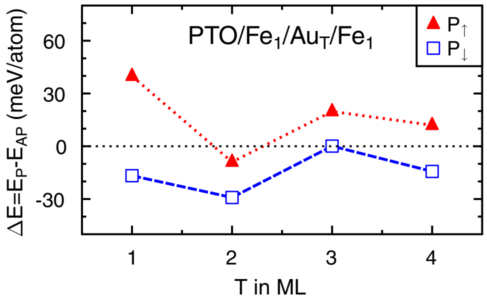

FIG. 2 (color online). The energy difference ΔE between P and AP alignment of the magnetizations in the two Fe layers as a function of Au monolayers separating them. The two curves correspond to the two polarization directions $P_\uparrow$ and $P_\downarrow$.

The complex numbers $\Delta r = \frac{r^\uparrow - r^\downarrow}{2}$ are the differences of the spin-dependent reflectivities of the FM layers 1 and 2 with $r^\uparrow$ ($r^\downarrow$) the reflection coefficient of the majority (minority) electrons. Obviously, the spacer material determines the period of the oscillation, whereas its strength and phase are related to the spin asymmetry of the reflection coefficients.

Exploring this expression, there are two possibilities to change the sign of the IEC strength. The first one is a change in the momentum $k_\perp$ that determines the oscillation period. It turned out that only some stationary points of the spacer material Fermi surface [17] contribute to the period of the oscillation. So, a change of the period requires a change of the Fermi surface, which would require a charging of the NM spacer material, which is strongly hampered by the effective charge screening in metals. To further rule out this mechanism in our hybrid system, we checked the neutrality of our trilayer. In all cases the Au layer stays neutral, and only at the FE/FM interface was a small charging of $0.2e$ as a result of the polarization screening found.

The second approach utilizes a change of the reflectivities and thus the phase and amplitude of the oscillation. Obviously, a modification of the FM/NM interface would be the most straightforward approach. It has been shown by You and Bader [18] that by insertion of an insulating barrier (I) an applied electric field can trigger the transition between P and AP alignment. The observed effect is due to the spin-dependent screening of the induced charge at the FM/I interface, which changes the spin asymmetry of the reflectivities. A similar approach was proposed by Zhuravlev, Vedyayev, and Tsymbal [19] by replacing the NM spacer by a FE. However, the disadvantage of inserting an insulating layer is the very small IEC energy due to the exponential decay of the wave functions in the insulator.

To preserve a large IEC, a change of the outer interfaces of the FM/NM/FM trilayer seems advantageous. As shown by You and Suzuki [20], it is also possible to change the reflectivity of the FM layer by placing a semiconductor on one side of the trilayer. The variation of the Schottky barrier at the interface under an applied voltage causes a sufficient change of reflectivities to switch the alignment of the FM layers. However, the required electric fields are strong and the switching is volatile.

Our approach aims at the change of the reflectivity at the FE/FM interface by means of the FE polarization and the screening of the excess charge in the ferromagnet. Since the polarization charge of PTO is about 1 electron per surface unit cell, it exceeds by far the capability of a semiconductor layer. Furthermore, the screening also influences the magnetic properties of the interface and thus changes the exchange splitting. Because of the remanence of the polarization, this switching mechanism is nonvolatile.

To further explore the origin of the change in IEC under polarization reversal, we investigated if either charging or

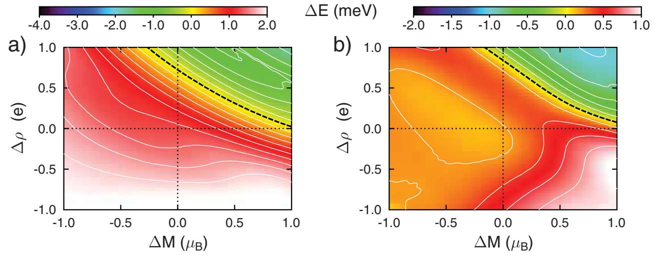

FIG. 3 (color). IEC energy as a function of local changes ΔM and $\Delta\rho$ for the Fe/Au/Fe system for a 5/6/5 trilayer. Units are meV per interface unit cell containing 1 Fe atom. (a) The thicknesses are fixed to 5/6/5; (b) Fe thickness variations of ±1 ML are included additionally.

change of magnetic moments shows a larger influence on the IEC. Therefore we extended the trilayer to a configuration 5/6/5 and used a slab calculation without the FE layer. After a self-consistent calculation, we obtained that the preferred coupling in this configuration is *AP*. Starting from the self-consistent potential, we simulated a potential change in the spirit of a rigid band shift at the outer boundary in one Fe layer, in such a way that we alter systematically the magnetization and the local charge. This allows us to map the strength of the coupling energy ΔE with respect to the change in magnetic moment ΔM and local charge $\Delta\rho$ approximately, as depicted in Fig. 3(a). The boundary between a preferred *P* and *AP* alignment is indicated by the dashed line, and a switching between the two states can be realized by either charging the interface or changing the magnetization, where the first way is in line with Ref. [20]. If both effects can be applied at once, a switching is obviously more likely. The size of the simulated shifts in charge and magnetization corresponds to the size of the calculated shifts at the PTO/Fe interface.

Up to this stage we have shown that for certain combinations $d_1/T/d_2$ a switching of the IEC can be triggered by the FE polarization reversal. To show that this behavior is robust against small variations of the layer thicknesses, we consider the system 5/6/5 with independent variations of the Fe layer thicknesses by ±1 ML. So, the additional contributions to the IEC energy are stemming from the systems 6/6/5 and 5/6/4. For reasons of simplicity, we assume that every configuration occurs in a real sample with a probability of 1/3. Figure 3(b) shows the averaged ΔE map. It is obvious that for the averaged system a sign reversal of the IEC strength can still be obtained for certain changes of the Fe layer with respect to magnetization and charge. Consequently, the observed mechanism is robust even under variations of the individual layer thicknesses.

To return to the GMR effect, we have calculated the expected change in in-plane resistance under the reversal of the magnetization directions within the semiclassical Boltzmann approach applying a constant relaxation time approximation [21]. For all considered systems, a change in resistivity of the order of 30% is expected, which provides a way to detect the magnetization reversal electrically.

In summary, we found a PTO/Fe/Au/Fe heterostructure that shows 180° switching of the magnetization of the free Fe layer by application of an electric field. The switching process is caused by the magnetoelectric coupling at the FE/FM interface amplified by the IEC in the FM/NM/FM trilayer. Finally, we have shown that this switching mechanism is caused by local changes of the charge and magnetization at the outer interfaces of FM/NM/FM trilayers. Others mechanisms beside the discussed magnetoelectric coupling may provide a link to other external fields. We hope that our theoretical prediction stimulates experimental investigations which support our findings.

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*michael.fechner@mat.ethz.ch

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