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Monte Carlo simulation of charge mediated magnetoelectricity in multiferroic bilayers

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ABSTRACT

Simulations of a bilayer ferroelectric/ferromagnetic multiferroic system were carried out, based on the Monte Carlo method and Metropolis dynamics. A generic model was implemented with a Janssen-like Hamiltonian, taking into account magnetoelectric interactions due to charge accumulation at the interface. Two different magnetic exchange constants were considered for accumulation and depletion states. Several screening lengths were also included. Simulations exhibit considerable magnetoelectric effects not only at low temperature, but also at temperature near to the transition point of the ferromagnetic layer. The results match experimental observations for this kind of structure and mechanism.

1. Introduction

Ferroelectric (FE) materials are dielectrics that maintain a net polarization even at zero electric field, property associated to the piezoelectric phenomenon. This characteristic is used in several technological applications, such as movement and pressure sensors, accelerometers, actuators and gyroscopes [1,2]. On the other hand, ferromagnetic materials (FM) retain magnetization even at zero magnetic field. They can be used in data store devices such as flash and hard drives. While ferroelectricity is associated to the charge, ferromagnetism is caused by the spin. Among the known materials, very few exhibit ferroelectricity and ferromagnetism simultaneously. Materials that show a direct relationship between both ferroelectric and ferromagnetic phenomena (magnetoelectric (ME) effect) are yet scarcer [3–5]. A new area in device design focuses on the possibility of building heterostructures that include, high performance ferroelectric and ferromagnetic materials with a considerable ME effect [6]. A first attempt for theoretically explaining the ME phenomenon was carried out in 1959 by Landau and Lifshitz. They proposed the existence of an additional term αHE in the free energy Hamiltonian, where α is a proportionality constant and H and E are magnetic and electric fields, respectively. Although this macroscopic description, followed by other authors [7,8], explains the relationship between the electric and magnetic fields, it lacks a more detailed correlation between the two elements that are involved in these systems, charge and spin. Janssen et al. [9,10] proposed an improved description of the interaction between the FE and FM components of the ME system. Using this model, they simulated the dynamics and configurations of domain walls for the unidimentional case. Given the difficulty of solving this model in greater dimensions, Li et al. [11] implemented an algorithm based on the Monte Carlo method for a two-dimensional lattice, in which magnetic and electric orders were imposed at each lattice point. Their results showed that the ME coupling, as well as the electric field, can produce a weak FM ordering.

Leufke et al. [12] grew $La_{0.87}Sr_{0.13}MnO_3/PbZr_{0.52}Ti_{0.48}O_3$ (PZT/LSMO) heterostructures by performing in situ magnetometry studies. These studies allowed precise measurements of the ME effects present between these materials. Such effects were associated to the charge accumulation at the interface, due to the PZT polarization under the effect of an electric field. This charge accumulation causes electron migration from the FM material toward the interface, creating a magnetic variation similar to that observed in manganite (LSMO) doping. According to the manganite phase diagram [13], this variation leads to greater critical temperatures and lower saturation magnetization. However, the decrease in the magnetization is higher than expected when only the charge accumulation effect is considered. Another study for $BaTiO_3/La_{0.67}Sr_{0.33}\ MnO_3$ (BTO/LSMO) heterostructures allows to conclude that the charge accumulation not only causes effects similar to the doping phenomenon, but also favors a modification of the kind of magnetic coupling in the first two layers of the magnetic material, shifting from the FM interaction in the depletion stage (charge carriers depletion from the interface) to an AF coupling in the accumulation stage (carriers accumulation at the interface).

In this work, Monte Carlo simulations of FE/FM bilayers in three-dimensional heterostructures were carried out. Magnetic and electric orders were individually imposed. Afterwards, the

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system was relaxed using the Metropolis dynamics and a model that included ME effects associated to the charge accumulation at the interface. The proposed Hamiltonian and the conditions given to it allowed to reproduce, for the first time, experimental behaviors through the Monte Carlo method (e.g. the shifting of the critical temperature in the ferromagnetic material and significant changes in the magnetic ordering type, when exposed to an external electric field). These behaviors were observed during accumulation and depletion periods in PZT/LSMO and PZT/BTO real systems.

2. Model and simulations

The simulations, considered a Hamiltonian for a tridimensional cubic system of $L \times L \times d$ dimensions, with periodic boundary conditions in the x–y plane and free boundary conditions out of the plane (z axis). Such a system is composed by two layers with thicknesses of $d1$ and $d2$; FE and FM order parameters were imposed to lower and upper layers respectively, through the electrical displacement ($\vec{u}$) and the Heisenberg spin ($\vec{S}$). Additionally, interactions between electric and magnetic systems were considered. According to the model described in [9,10], the following Hamiltonian was proposed:

$$H = H_{FE} + H_{FM} + H_{ME}, \tag{1}$$

where H_{FE} and H_{FM} represent the contributions of the electric and magnetic systems, respectively, and H_{ME} corresponds to the interaction between both, magnetic and electric layers. For the FE system, the following DIFFOUR Hamiltonian [10] was assumed:

$$H_{FE} = \sum_i \left(\frac{P_0^2}{2m} - \frac{a}{2}\vec{u}_i^{\,2} + \frac{b}{2}\vec{u}_i^{\,4} \right) - \sum_{nn} U_{ij}\left(\vec{u}_i \times \vec{u}_j\right) - \sum_i \vec{u}_i^{\,z} \times \vec{E} \tag{2}$$

Here, $\vec{u}_i$ is the ferroelectric dipole associated to the ith site, $(P_0^2/2m)$ is the kinetic energy, a and b are the double-well potential parameters for the ferroelectric spins, and U_{ij} is the FE exchange coefficient. Being $u\vec{}_i$ constant in magnitude and under a suitable reference system, the Hamiltonian can be rewritten [14] as

$$H_{FE} = -\sum_{nn} U_{ij}\left(\vec{u}_i \times \vec{u}_j\right) - \sum_i \vec{u}_i^{\,z} \times \vec{E}, \tag{3}$$

where $\vec{u}_i^{\,z}$ is the component of $\vec{u}_i$ in the z axis and E represents the electric field acting on the z direction.

For the magnetic system, a Heisenberg Hamiltonian was considered [15], given by

$$H_{FM} = -J\sum_{nn} \vec{S}_i \times \vec{S}_j - h\sum_i S_i^x - K\sum_i (S_i^x)^2, \tag{4}$$

being J the FM exchange constant, K the anisotropy constant, h the external magnetic field, $\vec{S}_i$ and $\vec{S}_j$ the spins associated to the ith and jth sites, respectively, and S_i^x the component of $S\vec{}_i$ in the x direction. In this model, J takes a $J1$ value for the interaction between one spin and its nearest neighbors, as long as this ion is out of the screening length (number of FM layers that provide charge carriers to the interface). Otherwise, J takes a $J2 > J1$ value if the ion is placed into that length and contributes to the screening.

For the ME coupling or interaction between the two subsystems, the subsequent model is proposed:

$$H_{me} = \sum_k g_{me} u_k^z \vec{S}_i \times \vec{S}_j. \tag{5}$$

In this expression, g_{me} is the parameter that fits the ME coupling intensity between the z component of the electrical displacement u_k^z and the magnetic interaction between spins. While the sums in Eqs. (3) and (4) consider nearest neighbor interactions (n.n.), in Eq. (5), $\vec{S}_i$ and $\vec{S}_j$ correspond to neighbor spins of $d1+1$ and $d1+2$ layers, exactly above the electric dipole u_k, which belongs to the $d1$ layer that is closest to the interface. Unlike the Janssen model, spatial and temporal inversion symmetries are not required, since both are broken at the interface between FE and FM materials, allowing the odd power term in u_k^z.

For positive values of U and J, FE and FM interactions are activated, and for positive values of g_{me} and u_k^z, an antiferromagnetic (AF) coupling between $d1+1$ and $d1+2$ layers is favored. This AF coupling competes with the FM one generated by the exchange parameter J. In order to simulate the charge accumulation mechanism at the interface [16], a restriction is imposed to the Hamiltonian. According to this restriction, the ME contribution only occurs when the u_k^z in the FE system of the layer that is nearest to the interface reaches a positive value. The former is justified as the electric polarization at the interface is the source of the screening phenomenon, in which spins coming from the ferromagnetic layer take part. It is assumed that if the electric displacement at the interface is zero or takes negative values, the ME effect is not present, and the magnetic system, in the vicinity of this site, behaves as an independent system, without any coupling to the FE layer. Simulations take into account the carriers accumulation at the interface by means of g_{me}, $J1$, and $J2$ parameters, not only to show an AF interaction in the first two layers of the magnetic system, but also for modeling experimentally observed changes at the critical temperature for this kind of heterostructures [17]. In the system under study, it is assumed that the screening length goes beyond the first two layers, considering a carrier emptying from layers that are more distant from the interface. In the model, the hole generation in these layers, favors an FM ordering through the exchange constant $J2$.

As it has been stated, this change in the magnetic ordering has been identified in PZT/LSMO and BTO/LSMO bilayers. The hole accumulation at the LSMO interface is comparable to a higher Sr doping in the manganite, being responsible for a greater critical temperature and a lower magnetization. To include this behavior in the present model, $J1$ and $J2$ exchange parameters were taken for carrier depletion and accumulation states in the magnetic system. For the depletion process, the constant considered was lower than that for the accumulation one, in order to be consequent with the doping and critical temperature changes mentioned above.

Initially, high temperature or disorder conditions in the spin and dipole directions, were imposed to the system. To simplify the model, an electrical displacement of ± 1 was randomly assigned at each lattice point in the FE layer, in one of the six directions ($\pm x$, $\pm y$, $\pm z$). In the FM layer, spins with $|\vec{S}| = 1$ were also randomly assigned, but in any direction, following a uniform distribution. For this initial temperature, new random $\vec{S}_i$ or $\vec{u}_i$ were chosen, and the energy change caused by this movement was calculated. This change was accepted or rejected according to the Metropolis Algorithm [15]. In order to ensure the convergence in the calculus of the observables, the lattice was scanned 10,000 times, where each time is considered as a Monte Carlo step (mcs) that can be taken as the time scale of simulations. Observables of interest as electric polarization in z, P_z, and magnetization in x, M_x, were averaged over the following 5000 mcs. These quantities were calculated according to (6) and (7):

$$P_z = \sum_i u_i^z \tag{6}$$

$$M_x = \sum_i S_i^x. \tag{7}$$

The process was repeated, lowering the temperature in a range ensuring a disorder–order change in both FE and FM systems.

From the calculation of these observables, it was possible to determine polarization and magnetization curves depending on