AIP | Journal of Applied Physics

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Citation: J. Appl. Phys. **113**, 17C719 (2013); doi: 10.1063/1.4795440
View online: http://dx.doi.org/10.1063/1.4795440
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v113/i17
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JOURNAL OF APPLIED PHYSICS **113**, 17C719 (2013)

Stress-mediated magnetoelectric memory effect with uni-axial $TbCo_2$/FeCo multilayer on 011-cut PMN-PT ferroelectric relaxor

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(Presented 15 January 2013; received 4 November 2012; accepted 7 December 2012; published online 28 March 2013)

We present here the implementation of a magnetoelectric memory with a voltage driven writing method using a ferroelectric relaxor substrate. The memory point consists of a magnetoelastic element in which two orthogonal stable magnetic states are defined by combining uni-axial anisotropy together with a magnetic polarization in the hard axis direction. Using a ferroelectric relaxor substrate, an anisotropic stress is created in the magnetic element when applying a voltage across electrodes. Because of the inverse magnetostrictive effect, the effective anisotropy of the magnetic element is controlled by the applied voltage and used to switch magnetization from one state to the other. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4795440]

I. INTRODUCTION

As the need for energy efficient data processing and data storage booms, magneto-electric memories have raised a tremendous interest among research teams. They could indeed combine the advantages of magnetic storage, such as non-volatility, speed, and endurance, with energy efficient electric writing techniques. Intrinsic magneto-electric materials (with the notable examples of Cr_2O_3 and $BiFeO_3$) have been considered in several designs, but offer only a weak magnetoelectric response at room temperature and therefore require either cryogenic techniques[1] or precise control of the operating temperature.[2] On the other hand, composite materials, introduced by Boomgaard *et al*. in 1976,[3] use strain-coupled piezoelectric and magnetostrictive materials. They can operate at room temperature and offer several design possibilities. With such an interface strain coupling, electric field control of ferromagnetism was shown in iron thin film structures.[4] Most of the composite memory devices proposed up to now are toggle memories, whose initial state must be known prior to writing operation,[5] or involve complex schemes to switch between multiple anisotropy directions defined by crystallographic axes of epitaxially grown materials,[6] or precisely synchronized driving signals.[7]

In 2010, we proposed and patented an innovative memory device called MELRAM (Magneto-ELectric Random Access Memory) based on a composite structure composed of a nanometer sized magnetostrictive material embedded in a piezoelectric matrix.[8–11] In particular, we showed that the competition between an external magnetic field and uni-axial magnetic anisotropy can be used to define two perpendicular equilibrium positions for magnetization[12,13] and that the application of stress can trigger the switch between both positions in a deterministic fashion. A macroscopic device using a commercial piezoelectric stack provided an experimental proof of concept of this memory element[14] and demonstrated the writing principle using piezoelectric stress. The characterization of the magnetic state was performed using either a Vibrating Sample Magnetometer (VSM) or Magneto Optical Kerr Effect (MOKE). However, this approach is not suitable for an integrated device at the sub-micrometer scale. We hereby present the results obtained using a relaxor substrate with adequate symmetry and characteristics. Such a substrate has already been coupled to magnetic layers for memory applications by Wu *et al*.[15] In their model, the information is supposed to be stored in the ferroelectric subsystem due to switchable remanent stress states. In our experiment, the information is stored in the magnetic subsystem and persists without either electrical tension or remanent stress.

II. PRINCIPLE OF THE MEMORY AND CONSIDERATIONS ABOUT STRESS

The magnetic and mechanical configurations of a MELRAM cell are given in Figure 1 and consists of a magnetic element with a defined easy axis (EA) at a 45° angle relative to the X axis, subjected to a polarizing field H_S in the hard axis (HA) direction and a mechanical stress. The EA can be obtained by considering an ellipsoid shaped element or induced during the film preparation (e.g., *sputtering under magnetic field*). The element has a high magnetostriction λ_S. Assuming the magnetization is homogeneous in the film, the magnetic free energy of the system submitted to in-plane stress is described by the following equation:

$$F_{mag} = -MH_s\cos\left(\frac{3\pi}{4}-\varphi\right) - \frac{1}{2}MH_A\cos^2\left(\frac{\pi}{4}-\varphi\right) - \frac{3}{2}\lambda_S(\sigma_{xx}-\sigma_{yy})\cos^2(\varphi) - \frac{3}{2}\lambda_S\sigma_{xy}\cos(\varphi)\sin(\varphi), \quad (1)$$

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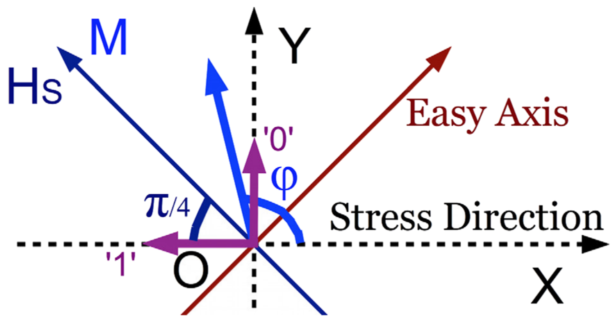

FIG. 1. Magnetic and mechanical configurations of a memory cell. The polarizing field H_S is applied perpendicular to the magnetic easy axis and defines two stable positions. Tensile or compressive stress is applied to switch between states.

where M is the magnetization of the element, making an angle φ with respect to OX, H_S is a polarizing field applied in the hard axis direction, H_A is the value of the effective anisotropy field, and σ_{ij} the mechanical stresses applied to the element in the plane of the film. As shown in the dashed curve of Figure 2, in the absence of applied stress, the competition between the uniaxial anisotropy and the Zeeman interaction leads to the creation of two minima of energy, for values of H_S lower than the anisotropy field H_A. In the case where H_S has a value of about $\sqrt{2}/2$ times the value of H_A, the two stable positions marked "1" and "0" are aligned along the OX− and OY+ axes. When a stress is applied, the energy profile is deformed by the magnetoelastic terms. Switching the magnetization to positions "1" or "0" means favoring a minimum of energy for $\varphi = \pi$ or $\varphi = \pi/2$, respectively. Therefore, the last term of the energy has no influence, since the product $\cos(\varphi)\sin(\varphi)$ is zero in these two positions. Shear stress, thus, have to be avoided. However, the term in $K \cdot \cos^2(\varphi)$ is of interest: depending on the sign and strength of K, the energy profile is suitably modified. It can be also deduced that for a maximum efficiency, σ_{xx} and σ_{yy} have to be of opposite signs, which leads to conditions on the substrate properties. A first approach is to consider a piezoelectric material with an in-plane polarization and electrodes on opposite vertical sides in order to generate an in-plane electric field, as proposed, for instance, by Hu *et al.*[16] The stress is then generated, say along OX using the d_{33} piezoelectric coefficient, and due to the Poisson effect, an opposite stress along OY is created. Unfortunately,

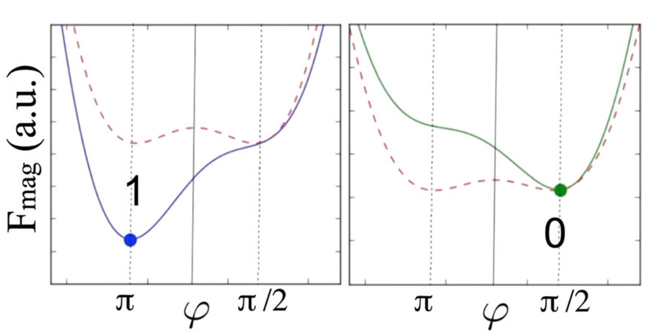

FIG. 2. Magnetic free energy of the system when submitted to tensile (left) or compressive (right) stress. Without stress applied, the energy exhibits two stable positions (dashed curve).

this approach is limited to non conductive magnetic layers as a conductive film induces a strong distortion of the electric field lines, which have to be perpendicular to the conductor, and therefore leads to improper stress generation. In this case, the electric polarization of the electroactive material has, then, to be perpendicular to the plane, and the relevant stress is related to the d_{31} and d_{32} coefficients. For most piezoelectrics, such as PZT, the deformation is isotropic in the plane and those coefficients are equal, leading to no effect on the magnetoelastic system. Suitable materials belong to a class of relaxors such as $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) or $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PZN-PT). For PMN-PT, compositions near the so-called morphotropic phase boundary, 011-cut and poled single crystals exhibit suitable characteristics ($d_{31} \approx -1900 \times 10^{-12}$ C/N and $d_{32} \approx +1000 \times 10^{-12}$ C/N).[17] With such a substrate, the writing procedure is as follows: upon tensile/compressive stress (i.e., $\sigma_{xx} - \sigma_{yy} > 0$), and assuming positive magnetostriction, the free energy exhibits a single minimum for $\varphi = \pi$ (Fig. 2, left). Magnetization will then rotate toward the "1" state whatever its previous state was. Upon the removal of the stress, the state is preserved. Upon compressive/tensile stress (i.e., $\sigma_{xx} - \sigma_{yy} < 0$), the minimum lies at $\varphi = \pi/2$ (Fig. 2, right), corresponding to the "0" state.

III. EXPERIMENTS

A schematic view of the device is shown at the top of Figure 3. As explained above, the stress is generated by a commercial PMN-PT substrate. Before further processing, one side of the substrate was mechanically polished to ensure a surface smooth enough for MOKE measurements. The magnetic element is a $10 \times (TbCo_{2(5nm)}/FeCo_{(5nm)})$ exchange coupled multilayer. This type of structure is used as it combines a fairly high magnetostriction and a well defined uni-axial anisotropy. The film was deposited onto the polished side of the substrate through a shadow mask by RF

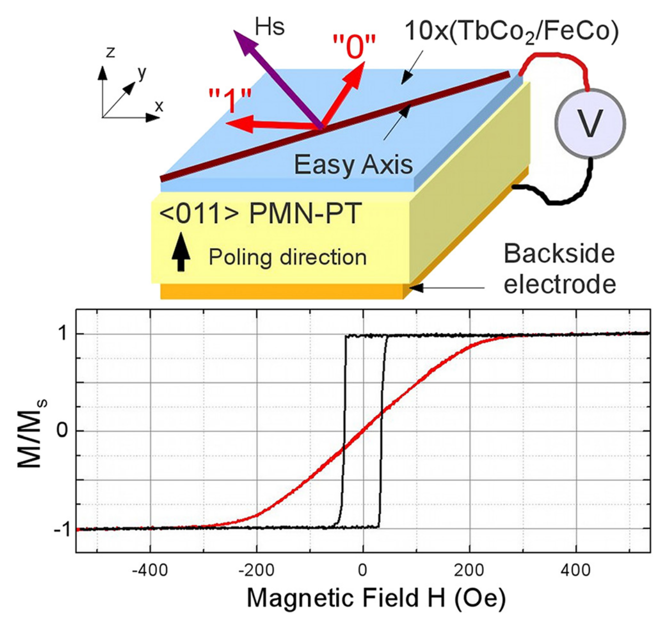

FIG. 3. Top: Schematic of the demonstrator. The magnetoelastic multilayer is deposited on top of a 011-cut and polarized PMN-PT single crystal. Bottom: VSM magnetization characterizations of the deposited layer.

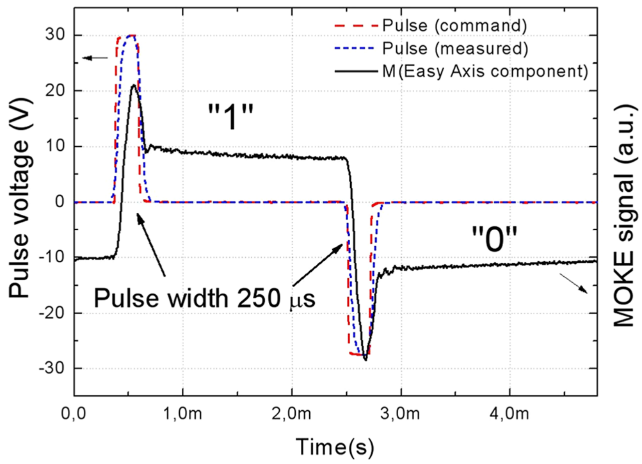

FIG. 4. MOKE evidence of magnetization switching between the two stable states upon application of positive or negative voltage pulses leading, respectively, to $\sigma_{xx} - \sigma_{yy} > 0$ and $\sigma_{xx} - \sigma_{yy} < 0$.

sputtering using a rotary turn table in a Leybold Z550 equipment. The deposition was made under a magnetic field generated by permanent magnets in order to induce a magnetic EA in the desired direction, i.e., with an angle of 45° with respect to the X axis. The obtained film magnetization was characterized with a Vibrating Sample Magnetometer (VSM). The results are shown in Figure 3. It is characteristic of a uni-axial behavior with an anisotropy field of about $H_A = 200$ Oe. For this kind of layer, the magnetostriction λ_S is measured by laser deflectometry with the clamped beam technique and is about 10^{-4}. The switching was evidenced using a MOKE setup to measure the projection of magnetization along the EA, while the polarizing field H_S was applied on the HA, so as to define two stable positions. As shown in Figure 4, the system behaves as expected. When applying either positive or negative voltage pulses, tensile or compressive stress, respectively, is generated leading to a switch to "1" or "0." One can also note that the state is kept upon removal of the voltage. Switching was obtained with 250 μs pulses, which is the shortest time allowed by the experimental setup.

IV. CONCLUSION

We have presented here the concept and experimental implementation of a device for a magnetoelectric memory cell using the effect of stress on anisotropy combined with the definition of 2 perpendicular stable positions in an anisotropic element. The writing of the information in the magnetic system is voltage controlled through the use of a 011-cut and poled PMN-PT relaxor substrate. A positive voltage sets the magnetization in one of the stable positions, whereas a negative voltage sets it in the other position. The position is kept when no voltage is applied. MOKE experiments evidenced the switching with sub-millisecond pulses. Since the information is stored magnetically, the readout can be made using GMR/spin valve techniques. Such an hysteretic giant magnetoresistance effect in a structure with two different values of anisotropy and submitted to mechanical stress was recently demonstrated.[18] Biasing strategies for the magnetic polarization using FePt permanent magnet layers are also investigated.[19]

ACKNOWLEDGMENTS

The authors would like to thank the Direction Générale de l'Armement (DGA-France) for the Ph.D. funding of Dr. Y. Dusch. This work was supported by the Agence Nationale de la Recherche ANR (France) through the PNano NAMAMIS project, by the Russian Federation Ministry of Education and Sciences (Contract No. 11.519.11.3023) and the Nanomaterials RAS Program, and the French Embassy in Moscow (MAE).

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