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Yajie Chen
Northeastern University

Trifon Fitchorov
Northeastern University

Carmine Vittoria
Northeastern University

V. G. Harris
Northeastern University

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Electrically controlled magnetization switching in a multiferroic heterostructure

Yajie Chen,[a)] Trifon Fitchorov, Carmine Vittoria, and V. G. Harris
Department of Electrical and Computer Engineering, Center for Microwave Magnetic Materials and Integrated Circuits, Northeastern University, Boston, Massachusetts 02115, USA

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A demonstration of magnetization reversal via the application of electric field across a multiferroic heterostructure, consisting of a FeCoV ribbon bonded to a lead magnesium niobate-lead titanate crystal, is presented. The magnetization switching occurs by an abrupt change in magnetization near ferromagnetic coercivity, coinciding with an electrical field-induced magnetic anisotropy field. Experiments reveal a converse magnetoelectric coupling of $\alpha=\mu_0(\mathrm{dM/dE})=1.6\times10^{-7}$ s m^{-1} upon magnetization reversal in the strain-mediated heterostructure. The frequency dependence of magnetization switching is presented and explained within the framework of a relaxation model for the multiferroic heterostructure. © *2010 American Institute of Physics*. [doi:10.1063/1.3475417]

Development of modern electronic devices increasingly trends toward miniaturization, multifunctionality, and improved performance, the latter often with regards to operating frequency, bandwidth, power handling, power consumption, gain, loss, etc. The development of such enhanced devices, assemblies, and systems is often driven by timely advances in the discovery and refinement of distinct materials and processing.[1,2] In recent years, there has been great interest shown in mulifterroic (MF) materials as either single phase[3] or as heterostructures.[4] In these systems electric and magnetic degrees of freedom are intimately coupled leading to the potential of controlling either magnetic polarization by application of electric fields or electric polarization by means of applied magnetic fields.

Electric field control of magnetization, often referred to as the converse magnetoelectric effect, has received considerable attention.[5] A key motivating factor is that magnetic fields are not required for magnetization switching. Techniques for electric control of magnetization include the spin-torque effect in spin-valves[6] and magnetic tunnel junctions,[7] and electric field control of magnetization in single phase multiferroic materials[8,9] and in ferromagnetic/ferroelectric heterostructures.[10,11]

An attractive approach toward electric field control of magnetization takes advantage of elastic coupling[12–15] either as magnetostrictive coupling or as elastostrictive coupling. Here, we show that piezomagnetism in a FeCoV alloy film/piezoelectric lead magnesium niobate-lead titanate (PMN-PT) crystal heterostructure can be used to manipulate the magnetic easy axis in the ferromagnetic film by simply changing the amplitude of the voltage applied to the piezoelectric crystal. Importantly, the highly efficient coupling between electric, magnetic, and strain degrees of freedom in the ferromagnet/piezoactuator heterostructure allows reversible switching of the magnetization, other than a simple change in amplitude of the magnetization as has been repeatedly demonstrated.[13,16,17] As such, this effect, demonstrated at room temperature, is of particularly fundamental and technological interest and utility.

In this letter, we demonstrate electric field control of magnetization reversal of a layered multiferroic heterostructure consisting of a ferromagnetic magnetostrictive FeCoV ribbon affixed to a piezoelectric PMN-PT single crystal substrate. The $Fe_{48}Co_{48}V_2$ ribbon has a thickness of 120 μm, saturation magnetization of $4\pi M_s=20$ kG, and saturation magnetostriction coefficient of $\lambda_s=60$ ppm. It is noteworthy that the $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$ single crystal, with $x=28-32\%$ PT, features anisotropic in-plane piezoelectric coefficients, d_{31} and d_{32}, where $d_{31}=-1500$ pC/N and $d_{32}=900$ pC/N when poled in the $\langle011\rangle$ direction. We employed a $\langle011\rangle$-type PMN-PT crystal of dimensions L10 $\times$W5$\times$T0.5 mm^3 coated with Au electrodes as a means of applying electric bias. This multiferroic heterostructure was designed to operate in the L-T ME coupling mode (i.e., longitudinal magnetized/transverse polarized) and consisted of a laminated structure of FeCoV/0.7PMN-0.3PT poled along the $\langle011\rangle$ direction. The two components were bonded with quick curing ethyl cyanoacrylate-based adhesive. Measurement of the converse magnetoelectric coupling was performed in the geometry depicted as the inset to Fig. 1. Specifically, an external magnetic field (H) was applied along d_{32} of the PMN-PT crystal, while the d_{33} direction aligned perpendicular to the heterostructure plane. For the heterostructure of the present study, a thickness ratio of 0.24, where t=120 μm for the FeCoV ribbon and t=500 μm for the PMN-PT crystal, was used.

Magnetic properties were measured using a vibrating sample magnetometer (VSM, Lakeshore Model 7400) with the magnetic field aligned parallel to d_{31}. The applied voltage ranged from −400 to 400 V across the PMN-PT crystal corresponding to an electric field strength (E) of −8 to +8 kV/cm. Low frequency magnetoelectric response was measured by the VSM while alternating square wave electric fields were applied across the PMN-PT crystal (BK Precision 4011A Functional Generator with High-Voltage Power Amplifier, Trek Model 609B-3). Electric field dependence of polarization was measured by a ferroelectric measurement system (Radiant Technologies, Inc.).

Figure 1 presents a representative magnetic hysteresis loop of the MF heterostructure under the application of electric voltages across the PMN-PT crystal. The magnetic rib-

[a)]Author to whom correspondence should be addressed. Electronic mail: y.chen@neu.edu.

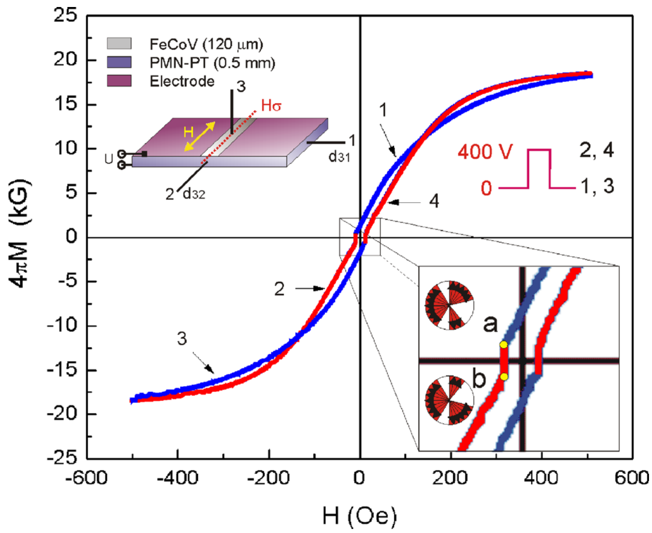

FIG. 1. (Color online) Magnetic hysteresis loop controlled by an applied electric voltage for an FeCoV/PMN-PT heterostructure. Upper left inset: Sketch of field configuration and geometries of the heterostructure. Right lower inset: Enlargement of the magnetization switched by an electric field and spin configurations in two magnetization states.

bon, affixed to the ferroelectric slab, showed a coercivity of ~10 Oe while the applied magnetic field was 500 Oe. There appeared a relatively abrupt step in the demagnetizing curve when a reverse magnetic field was applied near the value of the magnetic coercivity, H_c. The net magnetization vector undergoes a reversal from positive to negative through H_c. This experiment also demonstrated a reduction in coercivity by ~20% when an electric voltage of 300 V was applied. Thus, we can take advantage of the abrupt change in magnetization in the vicinity of coercivity in order to achieve a magnetization reversal, other than a simple change in magnitude of the magnetization as has been shown previously.[17] The latter is sometimes called magnetization switching but is more accurately described as a change in magnetization.

For the next experiment, a saturation magnetic field of 500 Oe was applied. The sample returned to its remanent state and a voltage of 400 V was applied as the reverse magnetic field approached −4 Oe. As shown in the inset to Fig. 1, point *a* on magnetization curve 1 abruptly drops to point *b* on segment 2 due to the application of voltage. More importantly, the precipitous drop in magnetization implies an apparent magnetization reversal that is evidenced by the change in sign of magnetization. In this instance, an electric field was insufficient to complete reversal of all spins unless a reverse saturation magnetization occurred. However, the detectable change in sign of magnetization arises from the reorientation of spins. The inset to Fig. 1 illustrates possible configurations of spins in two magnetization states, corresponding to points *a* and *b*. This switching reflects a converse magnetoelectric coupling coefficient of $\alpha=\mu_0(dM/dE)=1.6\times10^{-7}$ s m^{-1} which is comparable to the largest direct ME coupling with sharp domain switching, $\Delta P/\Delta H\sim10^{-9}$ s m^{-1},[3,18] and the recently reported room temperature value of $\alpha\sim2\times10^{-8}$ s m^{-1} for a $La_{0.67}Sr_{0.33}MnO_3/BaTiO_3$ film heterostructure.[4] Additionally, this result compares favorably with other laminated heterostructures with direct ME coupling without magnetization switching, such as $Pb(Zr,Ti)O_3$/terfenol-D, ~10^{-8} s m^{-1}.[19–21] Since the strength of ME coupling is a

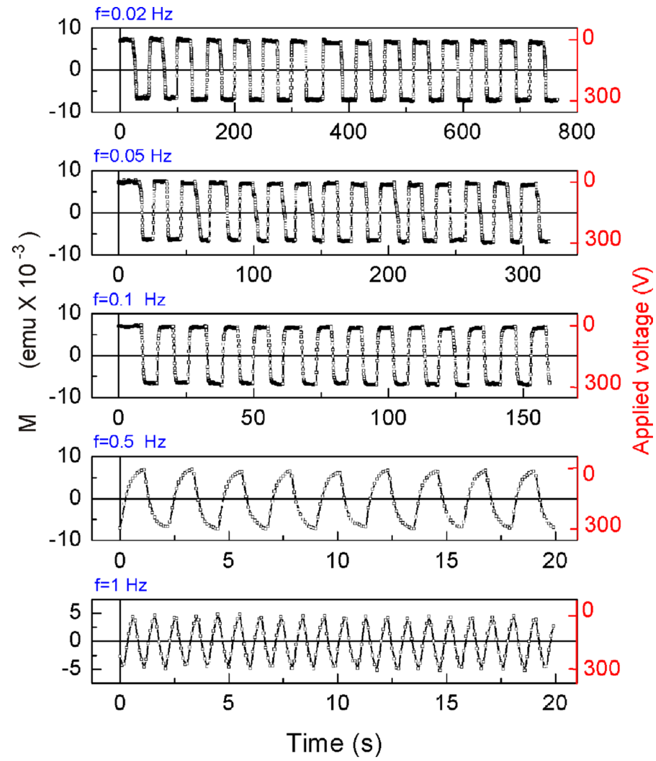

FIG. 2. (Color online) Magnetization switching under application of a square waveform electric field at different frequencies.

function of external magnetic field, this heterostructure yields a maximum CME coupling, $\alpha\sim2.2\times10^{-7}$ s m^{-1}, under the application of a magnetic field ~160 Oe. However, the CME response does not occur coherently with magnetization reversal, but rather as an abrupt change in magnitude of the magnetization.

Similarly, curves 3 and 4 in Fig. 1 represent voltage off and on states, respectively. It is unsurprising that the switching between curves 3 and 4 is similar to that between curves 1 and 2 due to the symmetric nature of the hysteresis loop. As a result, magnetization switching is reversible in the multiferroic heterostructure. Reversible or irreversible switching is affected by design considerations that include field configuration and geometry of the heterostructure, as well as the intrinsic properties of the piezoelectric and piezomagnetic materials.

The strength of the magnetization switching is proportional to the converse magnetoelectric coupling coefficient, α, which depends upon the slope of the demagnetizing curve near the magnetic coercive field. In the present case, a stress-induced anisotropy field is assumed to be aligned along the direction of the applied magnetic field. Thus, the stress-induced anisotropy field is expressed as,[22]

$$H_\sigma=\sigma\left\{\frac{\partial}{\partial M}\left[\frac{3}{2}\lambda+(e_{zz}^{me})_0\right]\right\}(\cos^2\varphi-\upsilon\sin^2\varphi), \qquad (1)$$

where σ and λ are the induced stress and magentostriction coefficient in the magnetic material, respectively, υ is the Poisson's ratio, $(e_{zz}^{me})_0$ is the magnetostrain in the demagnetized state, while φ represents the angle between the magnetization and the stress axes. The stress produced by an applied electric field is $\sigma=d_{32}YU/t$, where Y, U and t denote Young's modules, applied voltage and thickness of PMN-PT crystal, respectively. Given an ideal transformation of strain

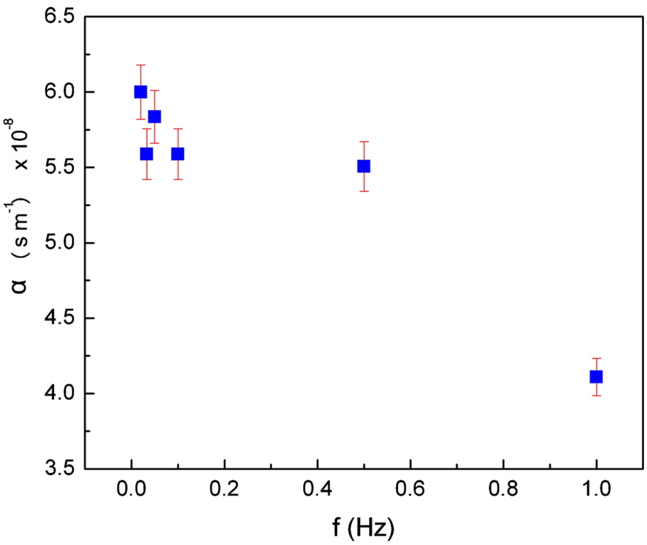

FIG. 3. (Color online) Frequency dependence of the converse magnetoelectric coupling coefficient in a FeCoV/PMN-PT heterostructure.

from the PMN-PT crystal to the magnetic film, the stress-induced anisotropy field due to the applied electric voltage is expressed as,

$$H_{\sigma}^{E} = \frac{3 \cdot d_{32} \cdot Y \cdot U}{2t} \frac{\partial \lambda}{\partial M} (\cos^2 \varphi - \upsilon \sin^2 \varphi). \quad (2)$$

This expression relates the derivative of the magnetostriction with respect to magnetization to the applied magnetic field. These experiments show a reduction in coercivity due to the application of an electric field, which suggests that an external electric field enables reversal of the magnetization. When the magnetic field strength approaches the coercive field, spins become frustrated and easily flipped, as sketched in the inset to Fig. 1. The response of magnetization switching is triggered by an induced magnetic field, of presumably merely a few Oersted. This is due to substantive values of λ and M under the application of low bias fields.

Low frequency switching, 0.02–1 Hz, of magnetization in this heterostructure was investigated. Accordingly, while the magnetization switching is driven by a pulsed electric field a small reverse bias magnetic field of $H_b = -4$ Oe is required. Subsequently, a square wave of amplitude 300 V was applied to the MF heterostructure, with no change in polarity of electric voltage (see Fig. 2). The electric field-induced magnetization is shown to switch in response to the pulsed electric field. This indicates a switching of the magnetization vector, though it does not imply a 180° rotation of spins. In fact, the reversal of the measured magnetization is generated by a stress-induced magnetic anisotropy field. Note that the switching is reversible, which is determined by the vector configuration of internal anisotropy field, bias magnetic field and stressed-induced magnetic field. Investigation of the underlying physical mechanisms of this effect is underway.

Moreover, we discuss two phenomena observed in these measurements. One, the induced signal evolves to a triangle-like waveform with increasing frequency, typically higher than 0.5 Hz. Second, the change in amplitude of induced magnetization decreases slightly with frequency. Correspondingly, the CME coupling coefficient reduces from 6×10^{-8} s m^{-1} at $f = 0.02$ Hz to 4.2×10^{-8} s m^{-1} at $f = 1$ Hz, as shown in Fig. 3. Both effects result from ferroelectric relaxation in the piezoelectric crystal and are dependent upon the applied E. This has been verified in previous experiments of the time and frequency domain response of a PMN-PT based heterostructure.[23,24] However, an electric field sufficient to saturate the electric polarization in the piezoelectric crystal can effectively reduce the relaxation time.

In summary, we have demonstrated an alternative path to magnetization reversal by the application of electric field in a multiferroic heterostructure. The switching of magnetization, i.e., the change in sign of magnetization, is shown to occur near magnetic coercivity of the magnetostrictive layer, in this case FeCoV. With the assistance of a small magnetic bias field of merely a few Oersted, one observes magnetization switching having a CME coupling of $\alpha \sim 1.7 \times 10^{-7}$ s m^{-1}. Importantly, this demonstration of magnetization reversal in a multiferroic heterostructure enables the development of a generation of magnetic devices dynamically controlled by electric fields.

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