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# Strain-controlled nonvolatile magnetization switching

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ABSTRACT

We investigate different approaches towards a nonvolatile switching of the remanent magnetization in single-crystalline ferromagnets at room temperature via elastic strain using ferromagnetic thin film/piezoelectric actuator hybrids. The piezoelectric actuator induces a voltage-controllable strain along different crystalline directions of the ferromagnetic thin film, resulting in modifications of its magnetization by converse magnetoelastic effects. We quantify the magnetization changes in the hybrids via ferromagnetic resonance spectroscopy and superconducting quantum interference device magnetometry. These measurements demonstrate a significant strain-induced change of the magnetization, limited by an inefficient strain transfer and domain formation in the particular system studied. To overcome these obstacles, we address practicable engineering concepts and use a model to demonstrate that a strain-controlled, nonvolatile magnetization switching should be possible in appropriately engineered ferromagnetic/piezoelectric actuator hybrids.



## 1. Introduction

In magnetoelectric multiferroics, where the ferromagnetic and ferroelectric order parameters are coupled, an electric-field control of the magnetic properties becomes possible [1–3]. This opens the way for appealing novel magnetization control schemes in future spintronic devices [4]. Unfortunately, single-phase multiferroics with strong magnetoelectric coupling remain rare [5,2]. Attractive alternatives are composite material systems made from ferroelectric and ferromagnetic compounds [6–9]. In such systems, an electric-field control of magnetism can be realized using electric field effects in carrier-mediated ferromagnets [10,11], or exchange coupling at ferromagnetic/multiferroic interfaces [12,13]. A third, powerful approach relies on strain-mediated, indirect magnetoelectric coupling in ferromagnetic/ferroelectric hybrid systems. In recent years, these hybrids were mostly fabricated by depositing ferromagnetic thin films on ferroelectric substrates [14–25]. Another approach to realize a strain-mediated control of the magnetization is to fabricate ferromagnetic thin film/piezoelectric actuator hybrids by either depositing or cementing ferromagnetic thin films onto commercially available $Pb(Zr_xTi_{1-x})O_3$ (PZT) multilayer piezoelectric actuator stacks [cf. Fig. 1(a)] [26–31]. In these hybrids, the application of a voltage to the piezoelectric actuator results in a deformation, which is transferred to the overlaying ferromagnetic thin film, changing its magnetic anisotropy due to the converse magnetoelastic effect.

In this paper, we report on two different experimental approaches towards a strain-mediated, nonvolatile, voltage-controlled magnetization switching in the complete absence of magnetic fields. They are based on ferromagnetic thin film/piezoelectric actuator hybrids using $Fe_3O_4$ as the ferromagnet. Our experiments show that a significant modification of the magnetic anisotropy is possible via voltage-controlled strain. This work extends our previous studies on ferromagnetic/ferroelectric hybrids [26,27,30,20], where we achieved a reversible reorientation of the magnetization by up to 90° in Ni based hybrids. However, a true switching of the magnetization between two (or more) remanent states solely by means of an electric field induced strain has not been realized experimentally up to now [32–37].

## 2. The spin-mechanics concept

The orientation of a well-defined homogeneous magnetization in a ferromagnet depends on external mechanical stress due to magnetostriction [38,39]. We exploit this so-called spin-mechanics scheme to control the magnetic anisotropy in $Fe_3O_4$ thin films cemented on $Pb(Zr_xTi_{1-x})O_3$ (PZT) multilayer piezoelectric actuator stacks [cf. Fig. 1(a)]. In particular, we compare different hybrids fabricated by cementing $Fe_3O_4$ thin films with different angles $\alpha$ between the crystallographic axes $\{\mathbf{x},\mathbf{y}\}$ of the film and the principal elongation axes $\{\mathbf{x}',\mathbf{y}'\}$ of the actuator with $\mathbf{z}\|\mathbf{z}'$.

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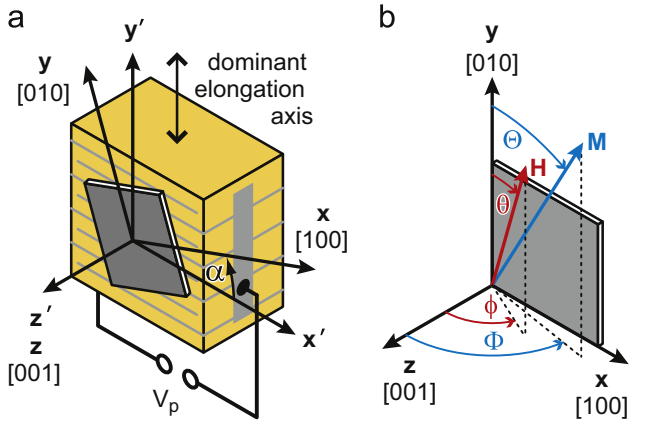


**Fig. 1.** (Colour online) (a) Schematic illustration of a $Fe_3O_4$ thin film/piezoelectric actuator hybrid. The coordinate system of the thin film and the actuator enclosing an angle $\alpha$ are denoted by $\{\mathbf{x},\mathbf{y},\mathbf{z}\}$ and $\{\mathbf{x}',\mathbf{y}',\mathbf{z}'\}$, respectively. (b) Orientation of the magnetic field $\mathbf{H}(H,\theta,\phi)$ and the magnetization $\mathbf{M}(M_s,\Theta,\Phi)$ with respect to the crystallographic axes $\langle 100\rangle$ of the $Fe_3O_4$ thin film.

The application of a voltage $V_p>0$ ($V_p<0$) to the piezoelectric actuator causes an elongation $\epsilon_2'>0$ (contraction $\epsilon_2'<0$) along the actuator's dominant elongation axis $\mathbf{y}'$, which is due to elasticity accompanied by a contraction (elongation) along the two orthogonal directions $\mathbf{x}'$ and $\mathbf{z}'$ [cf. Fig. 1(a)]. This leads to a change of the strain state $\epsilon$ of the $Fe_3O_4$ thin film elastically clamped onto the piezoelectric actuator. This causes a modification of the magnetic anisotropy, and thus alters the direction of the magnetization $\mathbf{M}$. In a macrospin model, the magnetization $\mathbf{M}$ of the $Fe_3O_4$ thin film described by $\mathbf{M}(M_s,\Theta,\Phi)=M_s\mathbf{m}(\Theta,\Phi)$ aligns in such a way that the free energy density $F$ takes its minimum value in equilibrium. Here, $m_x=\sin\Theta\sin\Phi$, $m_y=\cos\Theta$, and $m_z=\sin\Theta\cos\Phi$ [cf. Fig. 1(b)] are directional cosines and $M_s$ the saturation magnetization. The orientation of the magnetization $\mathbf{m}(\Theta,\Phi)$ can be calculated in the framework of a single domain model by using a phenomenological thermodynamic model based on the free energy density

$$F=F_{\text{Zeeman}}+F_{\text{u,eff}}^{001}+F_{\text{mc}}+F_{\text{el}}+F_{\text{me}} \tag{1}$$

with the Zeeman energy density $F_{\text{Zeeman}}=-\mu_0 M_s\mathbf{m}(\Theta,\Phi)H\mathbf{h}(\theta,\phi)$, the effective uniaxial anisotropy contribution $F_{\text{u,eff}}^{001}=\frac{1}{2}\mu_0 M_s^2 m_z^2+K_u^{001}m_z^2$, which comprises the demagnetization contribution and the uniaxial contribution $K_u^{001}$ resulting from the pseudomorphic growth of the ferromagnetic thin film, the first-order magnetocrystalline anisotropy contribution $F_{\text{mc}}=K_c(m_x^2m_y^2+m_y^2m_z^2+m_z^2m_x^2)$ with the cubic anisotropy constant $K_c$, the elastic energy density [40] $F_{\text{el}}=\frac{1}{2}c_{11}(\epsilon_1^2+\epsilon_2^2+\epsilon_3^2)+c_{12}(\epsilon_1\epsilon_2+\epsilon_2\epsilon_3+\epsilon_1\epsilon_3)+\frac{1}{2}c_{44}(\epsilon_4^2+\epsilon_5^2+\epsilon_6^2)$, and the magnetoelastic contribution

$$F_{\text{me}}=B_1\left[\epsilon_1\left(m_x^2-\frac{1}{3}\right)+\epsilon_2\left(m_y^2-\frac{1}{3}\right)+\epsilon_3\left(m_z^2-\frac{1}{3}\right)\right]+B_2(\epsilon_4m_ym_z+\epsilon_5m_xm_z+\epsilon_6m_xm_y). \tag{2}$$

The magnetoelastic coupling coefficients $B_1$ and $B_2$ can be written as a function of the magnetostrictive constants $\lambda_{100}$ and $\lambda_{111}$, which yields $B_1=-\frac{3}{2}\lambda_{100}(c_{11}-c_{12})$ and $B_2=-3\lambda_{111}c_{44}$, respectively. Here we use bulk values for the magnetostrictive constants ($\lambda_{100}=-19.5\times10^{-6}$ and $\lambda_{111}=+77.6\times10^{-6}$) as well as for the elastic stiffness constants $c_{ij}$ ($c_{11}=27.2\times10^{10}$ N/m$^2$, $c_{12}=17.8\times10^{10}$ N/m$^2$, and $c_{44}=6.1\times10^{10}$ N/m$^2$) [41–43].

To determine the modification of the magnetic anisotropy caused by strain effects induced by the piezoelectric actuator, we first derive the strain tensor $\epsilon$ of the $Fe_3O_4$ thin film. In the $\{\mathbf{x}',\mathbf{y}',\mathbf{z}'\}$ coordinate system, the strain components $\epsilon_4'$, $\epsilon_5'$, and $\epsilon_6'$ vanish, since no shear strains are present. Furthermore, as the thin film is clamped to the piezoelectric actuator, the in-plane strains $\epsilon_1'$ and $\epsilon_2'$ are not independent. Due to the actuator's elastic properties, these strain components are related via the Poisson ratio $\nu=0.45$ according to $\epsilon_1'=-\nu\epsilon_2'$. To obtain the strain tensor $\epsilon$ in the coordinate system of the $Fe_3O_4$ thin film $\{\mathbf{x},\mathbf{y},\mathbf{z}\}$, we apply a tensor transformation as described in detail in Refs. [38,44]. The strain components $\epsilon_i$ ($i=3,\ 4,\ 5$) can then be deduced according to the mechanical equilibrium condition $\sigma_i=\partial F/\partial\epsilon_i=0$ ($i=3,4,5$). With this relation, we finally obtain $\epsilon$ as a function of $\epsilon_2'$, neglecting comparably small magnetoelastic terms

$$\epsilon=\begin{pmatrix}-\frac{1}{2}[-1+\nu+(1+\nu)\cos(2\alpha)]\epsilon_2'\\ \frac{1}{2}[1-\nu+(1+\nu)\cos(2\alpha)]\epsilon_2'\\ -\frac{c_{12}}{c_{11}}(1-\nu)\epsilon_2'\\ 0\\ 0\\ (1+\nu)\sin(2\alpha)\epsilon_2'\end{pmatrix}. \tag{3}$$

Now we are in a position to derive the magnetization orientation $\mathbf{m}(\Theta,\Phi)$ by tracing the minimum of the total free energy density $F$ as a function of $\epsilon_2'$, which can be controlled by $V_p$. The corresponding evolution is calculated by minimizing Eq. (1) with respect to the orientation of the magnetization $\Theta$. Since the strain induced in the ferromagnetic thin film is of the order of $10^{-3}$ in our hybrid structures, the magnetoelastic energy contribution $F_{\text{me}}$ will not overcome the demagnetization energy in $Fe_3O_4$ thin films. Thus, the magnetization remains in-plane in case of zero magnetic field, which results in $\Phi=90^\circ$.

To illustrate the concept of a strain-induced, nonvolatile magnetization switching in zero magnetic and electric fields, Fig. 2 exemplary shows free energy density $F(\Theta,\epsilon_2')$ contours within the film plane for $\alpha=0^\circ$ [Fig. 2(b)–(d)] and $\alpha=45^\circ$ [Fig. 2(f)–(h)]. In both cases, the induced uniaxial strain is symmetric with respect to the crystallographic axes of the cubic ferromagnetic thin film. This results in two energetically equivalent minima in the free energy density $F$, which forces domain formation. To lift this degeneracy a small uniaxial magnetic anisotropy contribution in the film plane is introduced in the simulations given by $F_u^{ip}=K_u^{ip}(m_x\sin\Theta_u+m_y\cos\Theta_u)^2$ with the uniaxial anisotropy constant $K_u^{ip}$. For illustration purposes, we here use $\Theta_u=10^\circ$ and $K_u^{ip}>0$ with $|K_u^{ip}/K_c|=1/15$. To meet the experimental conditions of $Fe_3O_4$ thin films, we choose $K_u^{001}>0$ and $K_c<0$.

In case of $\alpha=0^\circ$, the ferromagnetic thin film is elongated and contracted along the cubic axes ($\mathbf{x}'\|\mathbf{x}$ and $\mathbf{y}'\|\mathbf{y}$) [cf. Fig. 2(a)] and thus no shear strains appear ($\epsilon_6=0$) [cf. Eq. (3)]. Starting at $\epsilon_2'=0$ ($V_p=0$ V) [cf. black line in Fig. 2(b)], the magnetization orientation $\Theta$ is aligned along a magnetically easy axis, e.g., at $\Theta=47^\circ$ (point A). Upon increasing $\epsilon_2'$ ($V_p>0$ V), the magnetically easy axis and thus the magnetization orientation $\Theta$ continuously rotates towards $\Theta=98^\circ$ (point B). The corresponding free energy density contour for $\epsilon_2'=+\epsilon_{\max}$ [cf. red line in Fig. 2(b)] is calculated assuming $B_1\epsilon_{\max}/K_c=3/5$, which corresponds to $\epsilon_{\max}=1\times10^{-3}$ in case of $Fe_3O_4$ thin films. By decreasing $\epsilon_2'$ back to 0, the magnetization orientation continuously rotates to the energetically stable direction $\Theta=133^\circ$ (point C) at $\epsilon_2'=0$. This demonstrates that a reorientation of the magnetization by about 86° is feasible. To check the possibility to reorient the magnetization orientation to the initial configuration (point A), $\epsilon_2'$ is inverted. Fig. 2(c) discloses that the easy axis gradually rotates from $\Theta=133^\circ$ (point C) to $\Theta=165^\circ$ (point D) by inducing $\epsilon_2'=-\epsilon_{\max}$. However, upon reducing $\epsilon_2'$ back to 0, the easy axis rotates back to $\Theta=133^\circ$ (point C). Thus, the magnetization remains in point C