Giant magnetoelectric effect via strain-induced spin reorientation transitions in ferromagnetic films

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(Received 3 October 2008; published 16 December 2008)

It is shown theoretically that a giant magnetoelectric susceptibility exceeding 10^{-6} s/m may be achieved in the ferromagnetic/ferroelectric epitaxial systems via the magnetization rotation induced by an electric field applied to the substrate. The predicted magnetoelectric anomaly results from the strain-driven spin reorientation transitions in ferromagnetic films, which take place at experimentally accessible misfit strains in $CoFe_2O_4$ and Ni films.

DOI: 10.1103/PhysRevB.78.212102 PACS number(s): 75.80.+q, 75.70.Ak, 77.65.−j

The magnetoelectric effect defined broadly as a coupling between magnetic and electric properties of a material system currently attracts great interest of physicists.[1–3] In particular, the switching of magnetization direction by an electric field represents the subject of cutting-edge research[4–7] owing to potential applications in novel magnetoelectric devices such as electric-write magnetic-read memories.[8] Since the direct magnetoelectric coupling is usually weak,[2] the switching caused by the strain-mediated indirect coupling between electric field and magnetization becomes an attractive possibility. This scenario may be realized via the strain-induced spin reorientation transitions (SRTs) in ferromagnetic films.

SRTs were observed in thin films of many ferromagnetic substances.[9–15] They represent a change in magnetization orientation at a critical film thickness ranging from several monolayers[9,13] to hundreds of nanometers.[15] The size-driven SRTs in ultrathin films are explained by a thickness-independent contribution to the film free energy, which results from the surface magnetocrystalline anisotropy[16] and competes with the energy of the film volume. Another important contribution is due to the magnetoelastic energy associated with the coupling between magnetization and lattice strains.[17,18] The relaxation of misfit strains with increasing film thickness also leads to SRTs in some epitaxial films.[11,13,15] This experimental observation indicates that the magnetization reorientation can be induced by tuning the film strains externally. When such strain-induced transition is caused by the application of electric field to a piezoelectric substrate, the sought magnetoelectric switching takes place.

In this Brief Report, the strain-driven SRTs are described theoretically using a nonlinear thermodynamic approach[19–21] developed earlier. The calculations are performed for relatively thick (001)-oriented films of cubic ferromagnets, where the surface effect on the magnetization orientation may be neglected. It is shown that, in $CoFe_2O_4$ and Ni films, the magnetization reorientation occurs at experimentally accessible critical strains of small magnitude. For the ferromagnetic/ferroelectric heterostructures, where the substrate has a high piezoelectric response, a giant magnetoelectric susceptibility is predicted.

Consider a thin ferromagnetic film grown on a dissimilar thick nonmagnetic substrate. Since there are no mechanical forces acting on the upper film surface, the Helmholtz free energy may be used to determine its equilibrium thermodynamic state. For cubic ferromagnets, the contribution to the Helmholtz free-energy density, which is associated with the energy of magnetocrystalline anisotropy, can be written as[19]

$$U_{\rm an} = K_1(m_1^2m_2^2 + m_1^2m_3^2 + m_2^2m_3^2) + K_2m_1^2m_2^2m_3^2, \qquad (1)$$

where K_1 and K_2 are the anisotropy constants at fixed strains, and m_i $(i=1,2,3)$ are the direction cosines of the magnetization $\mathbf{M}_s$ relative to the principal cubic axes. The elastic energy contribution equals

$$U_{\rm el} = \frac{1}{2}c_{11}(u_{11}^2 + u_{22}^2 + u_{33}^2) + c_{12}(u_{11}u_{22} + u_{11}u_{33} + u_{22}u_{33}) + \frac{1}{2}c_{44}(u_{12}^2 + u_{13}^2 + u_{23}^2), \qquad (2)$$

where c_{11}, c_{12}, and c_{44} are the elastic stiffnesses at constant magnetization, and u_{ij} (i and $j=1,2,3$) are the lattice strains defined in the crystallographic reference frame (x_1, x_2, x_3) with the x_3 axis orthogonal to the film surfaces. Owing to the coupling between the magnetization and lattice strains, a magnetoelastic contribution $U_{\rm me}$ also exists,[17,18] which is usually approximated by the formula

$$U_{\rm me} = B_1\left[\left(m_1^2 - \frac{1}{3}\right)u_{11} + \left(m_2^2 - \frac{1}{3}\right)u_{22} + \left(m_3^2 - \frac{1}{3}\right)u_{33}\right] + B_2(m_1m_2u_{12} + m_1m_3u_{13} + m_2m_3u_{23}), \qquad (3)$$

involving two magnetoelastic coefficients B_1 and B_2. The lattice strains u_{ij} in Eqs. (2) and (3) can be calculated via mechanical boundary conditions of the problem using the magnetoelastic equations of state $\sigma_{ij}=\partial(U_{\rm el}+U_{\rm me})/\partial u_{ij}$, where σ_{ij} are the mechanical stresses. When the ferromagnetic layer is in a single-domain state and there are no misfit dislocations in the film/substrate system, the strain and stress fields inside the film may be assumed to be homogeneous. In this case, the lattice matching at the interface gives $u_{11}=u_{m1}$, $u_{22}=u_{m2}$, and $u_{12}=0$, where $u_{m1}=(b_1-a)/a$ and $u_{m2}=(b_2-a)/a$ are the misfit strains defined by the differences between the substrate lattice parameters b_1 and b_2 measured along the x_1 and x_2 axes and the lattice constant a of a free-standing film.[22] The conditions $\sigma_{13}=\sigma_{23}=\sigma_{33}=0$ fulfilled on the film free surface further give $u_{13}=-B_2m_1m_3/c_{44}$, $u_{23}=-B_2m_2m_3/c_{44}$, and $u_{33}=-[B_1(m_3^2-1/3)+c_{12}(u_{m1}+u_{m2})]/c_{11}$. The substitution of calculated strains into Eqs. (2) and (3)

and the summation of all contributions including the magnetostatic energy yield the following expression for the energy density ΔF of a homogeneously magnetized film:

$$\Delta F = B_1(u_{m1}m_1^2 + u_{m2}m_2^2) + \left[\frac{1}{2}\mu_0 M_s^2 - \frac{B_1^2}{6c_{11}} - \frac{c_{12}}{c_{11}} \times B_1(u_{m1}+u_{m2})\right]m_3^2 + K_1 m_1^2 m_2^2 + \left(K_1 + \frac{B_1^2}{2c_{11}} - \frac{B_2^2}{2c_{44}}\right) \times (m_1^2+m_2^2)m_3^2 + K_2 m_1^2 m_2^2 m_3^2, \quad (4)$$

where terms independent of the magnetization direction were omitted. The single-domain state is assumed here in order to evaluate the remnant magnetization appearing after the application of a strong magnetic field (this magnetization is used to determine SRTs experimentally). It should be noted that the magnetocrystalline coefficient K_1 involved in Eq. (4) differs from the bulk anisotropy constant $K_{1\sigma}$, which is measured at constant stresses, but can be easily calculated as $K_1 = K_{1\sigma} + B_1^2 c_{11}/[(c_{11}-c_{12})(c_{11}+2c_{12})] + B_2^2/(2c_{44})$. Equation (4) also represents a good approximation for the mean energy density of a thick film with large densities ρ_1 and ρ_2 of misfit dislocations at the film/substrate interface. Indeed, when the dislocation spacing is much smaller than the film thickness, the influence of dislocation arrays on the mean lattice strains can be described by replacing the substrate lattice parameters b_1 and b_2 in the relations for u_{m1} and u_{m2} by effective parameters $b_1^* = b_1(1-\rho_1)$ and $b_2^* = b_2(1-\rho_2)$.[23] It should be emphasized that the strain relaxation caused by the generation of misfit dislocations can be employed to tune the lattice strains additionally by varying the thickness of an epitaxial film grown on an appropriate buffer layer deposited on a ferroelectric substrate.

The most remarkable feature of Eq. (4) is the presence of terms linearly dependent on the misfit strains u_{m1} and u_{m2}. Since these strains may be both positive and negative, the mechanical substrate effect may change the direction of the easy axis of magnetization. The resulting strain-induced SRT can be described by calculating the equilibrium orientation of magnetization as a function of strains u_{m1} and u_{m2} via the minimization of ΔF. Since the direction cosines m_i satisfy the relation $m_1^2+m_2^2+m_3^2=1$, the energy ΔF appears to be a function of only two variables.

Consider first the case of equal misfit strains ($u_{m1}=u_{m2}=u_m$), which corresponds to a cubic or tetragonal substrate (or thick buffer layer) with the (001)-oriented surface. In order to describe various possible situations, the calculations were performed for cobalt ferrite and iron, where the spontaneous magnetization $\mathbf{M}_s$ is oriented in bulk crystals along one of the edges of the unit cell ($K_{1\sigma}>0$), and for nickel, where $\mathbf{M}_s$ is directed along the cube diagonal ($K_{1\sigma}<0$).[19] In $CoFe_2O_4$ films, an abrupt SRT takes place at a critical misfit strain u_m^* defined by the formula

$$u_m^* = \frac{c_{11}}{B_1(c_{11}+2c_{12})}\left(\frac{1}{2}\mu_0 M_s^2 - \frac{B_1^2}{6c_{11}}\right). \quad (5)$$

With $M_s = 3.5\times10^5$ A/m, $B_1 = 5.9\times10^7$ J/m^3 (Ref. 5), $c_{11} = 2.7\times10^{11}$ N/m^2, and $c_{12} = 1.6\times10^{11}$ N/m^2 (Ref. 24), Eq. (5) gives a small tensile strain of $u_m^* \cong 0.06\%$. In the range of positive misfit strains $u_m > u_m^*$, the magnetization is orthogonal to the film surfaces owing to $B_1>0$; whereas at $u_m < u_m^*$ $\mathbf{M}_s$ is parallel to one of the in-plane crystallographic axes [100] or [010]. This result agrees with the experimental observations of magnetization orientations in $CoFe_2O_4$ films of different thicknesses,[15] which were epitaxially grown on MgO. Indeed, the out-of-plane magnetization appears at thicknesses $t \le 240$ nm ($u_m \ge 0.25\%$), whereas at $t = 400$ nm ($u_m = 0.012\%$) the preferential direction of $\mathbf{M}_s$ lies in the film plane.[15] Hence the prediction of the strain-induced SRT at $u_m^* \cong 0.06\%$ is consistent with the experimental data.

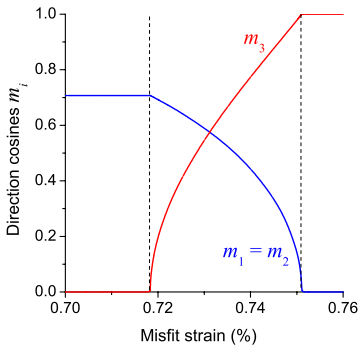

FIG. 1. (Color online) Direction cosines m_i of the spontaneous magnetization in a thick Ni film as a function of the misfit strain u_m in the heterostructure.

In iron, the magnetization is much larger than in $CoFe_2O_4$ ($M_s = 1.7\times10^6$ A/m),[25] whereas the magnetoelastic constant B_1 has much smaller absolute value [$B_1 = -3.3\times10^6$ J/m^3 in bulk crystals[25] and $B_1 \sim 1\times10^6$ J/m^3 in highly strained films;[26] $c_{11} = 2.42\times10^{11}$ N/m^2 and $c_{12} = 1.465\times10^{11}$ N/m^2 (see Ref. 27)]. As a result, the strain-induced SRT could take place in thick Fe films only at large strains above 20%, which are not accessible experimentally. However, such transition is expected to be possible in ultrathin Fe films with thicknesses close to the critical thickness at which the size-induced SRT occurs.[10,13]

In contrast to $CoFe_2O_4$ and Fe films, a gradual strain-driven magnetization rotation should take place in Ni films. Indeed, the minimization of the free energy (4) shows that between $u_m^* \cong 0.718\%$ and $u_m^{**} \cong 0.751\%$ the angle between $\mathbf{M}_s$ and the substrate normal gradually changes from 90° to 0°.[28] The dependence of the direction cosines m_i on the misfit strain is shown in Fig. 1. This behavior is similar to the strain-driven rotation of spontaneous polarization in ferroelectric thin films.[29] It should be noted that in the intermediate strain range $u_m^* < u_m < u_m^{**}$, where $m_1 = m_2 \neq 0$ and $m_3 \neq 0$, the shear lattice strains u_{13} and u_{23} differ from zero so that the phase state of the Ni film is formally monoclinic.

Suppose now that the ferromagnetic film is deposited on a thick ferroelectric substrate with two opposite faces covered by continuous electrodes. For simplicity, we assume that in the absence of electric field applied to the substrate the misfit strains u_{m1} and u_{m2} are equal to each other ($u_{m1}=u_{m2}=u_m^0$). Owing to the converse piezoelectric effect inherent in ferroelectric materials, the applied electric field $\mathbf{E}$ creates macroscopic strains $u_{ij} = d_{kij}E_k$ in the substrate having piezoelectric coefficients d_{kij}. Via the interfacial coupling in the film/

substrate system, the field-induced substrate deformations change the in-plane lattice strains u_{11} and u_{22} in a ferromagnetic film, which may result in the magnetization reorientation. This *electric-field-driven* SRT can be described using Eq. (4) and taking into account variations in the misfit strains u_{m1} and u_{m2} with the field intensity E. To maximize the influence of electric field on the film lattice strains, relaxor ferroelectrics, such as $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PZN-PT) or $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) having ultrahigh piezoelectric coefficients,[30] should be used as a substrate.

If the electric field is applied along the x_3 axis (substrate with the top and bottom electrodes), the misfit strains may be written in the linear approximation as $u_{m1}(E)=u_m^0+d_{31}E$ and $u_{m2}(E)=u_m^0+d_{32}E$. (The matrix notation is employed for piezoelectric coefficients here and below.) In the symmetric case ($d_{32}=d_{31}$), the influence of electric field on the magnetization orientation becomes similar to the strain effect discussed above. Therefore, to induce an SRT in the $CoFe_2O_4$ film magnetized along the [001] axis ($u_m^0>u_m^*$), the misfit strain $u_m(E)=u_m^0+d_{31}E$ should decrease below the critical value u_m^* given by Eq. (5). Since the out-of-plane magnetization changes by M_s at the transition,[31] the maximum absolute value of the magnetoelectric susceptibility $\alpha=\mu_0\Delta M/\Delta E$ equals

$$\alpha_{\max}=\left|\frac{\mu_0 M_s d_{31}}{u_m^*-u_m^0}\right|. \tag{6}$$

It should be emphasized that the field-induced reduction in the misfit strain may be rather large ($\sim 1\%$ at the piezoelectric coefficient $d_{31}\sim -1000$ pm/V given in Ref. 32). Indeed, it is created by the field directed *along* the substrate polarization, which is limited only by the high dielectric breakdown field $E_b>100$ kV/cm of PZN-PT or PMN-PT,[30] but not by the ferroelectric coercive field $E_c\sim 2$ kV/cm.

In contrast to $CoFe_2O_4$ and other ferromagnetic films with an abrupt SRT, for which the susceptibility α formally diverges as $u_m^0\to u_m^*$, the material systems involving Ni films display only a limited magnetoelectric anomaly because here the magnetization rotates gradually in a finite strain range. To evaluate the upper bound of α, we studied the electric-field-induced magnetization reorientation in Ni films for a special heterostructure, where the initial strain u_m^0 is equal to the critical strain $u_m^{**}\cong 0.751\%$ corresponding to the right boundary of SRT in Fig. 1. Using the dependence $m_3(u_m)$ plotted in Fig. 1 and the relation $E=(u_m-u_m^0)/d_{31}$ between the electric field and the misfit strain with $d_{31}=-1000$ pm/V, we calculated the magnetoelectric susceptibility as $\alpha(E)=\mu_0 M_s[m_3(E)-1]/E$. The results of calculations (see Fig. 2) demonstrate two remarkable features: (i) the susceptibility reaches very high level of 10^{-6} s/m already at a small field $E\approx 20$ V/cm and (ii) the magnitude of α weakly depends on the field intensity up to $E\sim 5$ kV/cm.[33] The maximum theoretical value $\alpha_{\max}=1.86\times 10^{-6}$ s/m is almost ten times larger than the magnetoelectric susceptibility $\alpha\sim 2.3\times 10^{-7}$ s/m achieved recently in the $La_{0.67}Sr_{0.33}MnO_3/BaTiO_3$ heterostructure.[7] This value also greatly exceeds the susceptibility reported for the $La_{0.7}Sr_{0.3}MnO_3$ film grown on PMN-PT in Ref. 6 and is several orders of magnitude larger than the susceptibilities of magnetoelectric crystals.[1]

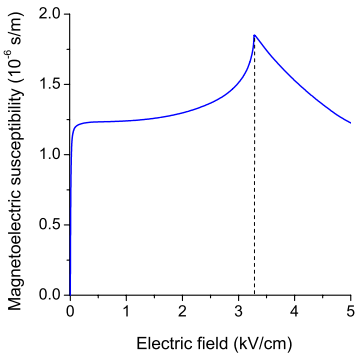

FIG. 2. (Color online) Magnetoelectric susceptibility of the Ni/PZN-4.5% PT heterostructure as a function of the electric field applied to the substrate in the direction orthogonal to film surfaces. The initial misfit strain is taken to be $u_m^0=0.7511\%$ and the substrate piezoelectric coefficient $d_{31}=-1000$ pm/V. The peak of susceptibility (dashed line) corresponds to the end of SRT at $u_m\cong 0.718\%$. The maximum susceptibility $\alpha_{\max}$ reduces to 1.46×10^{-6} s/m at $u_m^0=0.76\%$ and down to 9.88×10^{-5} s/m at $u_m^0=0.78\%$.

The electric field can be applied to the substrate in a direction parallel to the film surfaces as well. In this case, the strains $u_{m1}(E)$ and $u_{m2}(E)$ become very different so that the magnetoelectric effect acquires new features. If the field is directed along the [100] crystallographic axis of the film, the misfit strains can be evaluated as $u_{m1}(E)=u_m^0+d_{33}^*E$ and $u_{m2}(E)=u_m^0+d_{31}^*E$, where d_{in}^* are the substrate piezoelectric coefficients defined in the reference frame (x_1^*,x_2^*,x_3^*) with the x_3^* axis oriented along the field direction and the x_1^* axis parallel to the interface. For $CoFe_2O_4$ films, the calculation shows that the SRT can be induced at $u_m^0>u_m^*$ by an in-plane electric field directed along the substrate polarization. The critical field intensity $E_{\rm cr}$ equals

$$E_{\rm cr}=-\frac{(c_{11}+2c_{12})(u_m^0-u_m^*)}{[(c_{11}+c_{12})d_{31}^*+c_{12}d_{33}^*]}, \tag{7}$$

where the denominator is negative since $d_{31}^*<0$ and its magnitude is close to $d_{33}^*/2$.[32] When the applied field exceeds $E_{\rm cr}$,

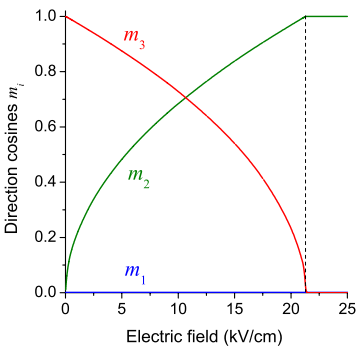

FIG. 3. (Color online) Direction cosines m_i of the spontaneous magnetization in a thick Ni film as a function of electric field applied to the PZN-4.5% PT substrate along the x_1 axis parallel to the interface. The initial misfit strain in the heterostructure is assumed to be $u_m^0=0.7511\%$.

the magnetization flips onto the film plane and becomes parallel to the x_2 axis. The maximum magnetoelectric susceptibility $\alpha_{\max}=\mu_0 M_s/E_c$ is proportional to $(u_m^0-u_m^*)^{-1}$ and exceeds 10^{-6} s/m at $u_m^0-u_m^*<0.056\%$ in the case of $CoFe_2O_4$ film grown on PZN-4.5% PT [$d_{33}^*=2000$ pm/V and $d_{31}^*=-1000$ pm/V (see Ref. 32)].

For Ni films, the minimization of the energy density (4) demonstrates that a gradual magnetization rotation takes place in these films when an in-plane electric field is applied to the ferroelectric substrate. Figure 3 shows the direction cosines of magnetization as a function of the field intensity for the special heterostructure introduced above ($u_m^0=u_m^{**}$). Remarkably, the magnetization rotates in the (100) crystallographic plane ($m_1=0$), but not in the $(1\bar{1}0)$ one ($m_1=m_2$), as it happens under the influence of electric field orthogonal to the film/substrate interface. The variation in the magnetoelectric susceptibility $\alpha(E)=\mu_0 M_s[m_3(E)-1]/E$ with the field intensity is similar to the dependence shown in Fig. 2. However, the maximum theoretical susceptibility $\alpha_{\max}=0.285\times10^{-6}$ s/m reached at the end of SRT is considerably smaller than in the case of electric field orthogonal to the interface.

Thus, the spin reorientation transitions may be induced in epitaxial ferromagnetic films by a moderate electric field applied to a ferroelectric substrate. The resulting magnetoelectric effect, which is mediated by the mechanical film/substrate interaction, increases dramatically when the misfit strain in the heterostructure becomes close to a critical value corresponding to the strain-induced SRT.[34] The magnetoelectric susceptibility of such ferromagnetic/ferroelectric heterostructures may reach giant values exceeding 10^{-6} s/m.

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