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THE TEMPERATURE-SENSITIVITY OF A CRITICAL ELECTRIC FIELD INDUCED MAGNETIC EASY-AXIS REORIENTATION FERROMAGNETIC/FERROELECTRIC LAYERED HETEROSTRUCTURES

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Abstract. *We have investigated the effects of temperature on the magnetic easy axis reorientation (EARs) in polycrystalline ferromagnetic (FM) nano-scale thin films, i.e. (001)-oriented cubic films such as Fe, Ni, CFO and (0001)-oriented hexagonal films such as Co, grown on ferroelectric (FE) substrates, for example PZT, BTO substrates. The model of FM/FE bilayered heterostructures has been applied to study the total free energy of the FM films. By minimizing this energy, we have shown that temperature has a significant effect on a critical electric field induced magnetic reorientation.*

Keywords: magnetoelectric effect, multiferroic materials, magnetic reorientation.

I. INTRODUCTION

The multiferroic materials have been the subject of intense experimental and theoretical investigations, motivated by the coexistence of at least two ferroic phases that these systems display [1, 2]. In multiferroic materials, the coupling interaction between the different order parameters, indeed, is governed by the magnetoelectric (ME) effect discovered long time ago by Landau and Lifshitz from thermodynamics and symmetry considerations [3]. Interestingly, neither of the constituent phases owns ME effect, but the coupling interaction between these phases can induce a remarkable ME effect. Hence, this phenomenon has recently enjoyed a great interest in the context of a search for the novel functionalities, i.e. a controllable possibility of the magnetization (or electric polarization) by applying an electric (or magnetic) field [4, 5]. In particular, the

electric-field control of magnetization known as converse ME effect has attracted the attention of the physics community due to its potential applications, such as electrically tunable microwave magnetic devices [2], electric- write magnetic memories [6]. Given the potential applications, converse ME effect of the multiferroic materials has been therefore broadly investigated. In practice, those devices are desired to work in a regular condition, i.e. room temperature, as well as in an unstable and a severe environment, for example, a very hot or very cold area. The temperature sensitivity therefore plays an important role in the device's functions. One of the main themes is to understand the physics mechanisms of the coupling between the magnetic order and the electric one [7, 8]. Indeed, Pertsev has considered the lattice misfit at the FM/FE interface caused the moderate strain-induced out-of-plane easy axis reorientation (EAR) in FM films grown on relaxor ferroelectrics [7]. Expecilaly, Hu *et al.* has also calculated the electric-field-induced EARs in FM/FE layered heterostructures, based on a phenomenological approach, taken into account the residual strains as well as different modes of applied electric fields [8]. Most of the previous studies had been, however, carried out at the fixed room temperature $T_R = 300$ K, little is known yet about the physics mechanism of ME effect as the temperature away from T_R . The purpose of this study is to provide a simple theoretical framework of the temperature effect on the EARs reorientation away from room temperature. We present a detailed discussion on the electric-field-induced EARs in FM/FE heterostructure with two structures of the magnetic films, e.g. a cubic structure (Fe, Ni, CFO) and a hexagonal structure (Co) grown on FE substrates, i.e. Pb(Zn,Ti)O₃(PZT), PZN-PT, BaTiO₃(BTO).

In this work, we investigate systematically the critical electric field induced the magnetic easy-axis reorientation in the wide range of temperature from 250 K to the Curie temperature, T_C , for different FM films, i.e. Fe(1043K), Ni(631K), CFO(793K) and Co(1350 K) [9]. For the practical applications, we however pay our attention to the temperature range from 250K up to 350K which is far from T_C . In order to compare with the previous studies at room temperature $T_R = 300$ K, the physics properties, i.e. the critical electric field, at temperature away from T_R are considered.

This paper is organized as the follows: In Sec. II, we describe the model Hamiltonian and briefly provide the calculation for EARs reorientation condition, i.e. the effect of temperature on the critical electric field induced EARs while the more detailed discussion for the particular systems are presented in Sec. III. Finally, we outline our conclusions in Sec. IV.

II. THEORETICAL MODEL

Our starting point for the studies of a FM/FE bilayered heterostructure with a FM thin film grown on a FE substrate is the well-known model for the Helmholtz free energy of the FM films in a single domain state [7, 8] written as the follows:

$$F = F_{mc} + F_{shape} + F_{me} + F_S \quad (1)$$

where the first term of F_{mc} is the magneto crystalline anisotropy energy, F_{shape} is the shape anisotropy, F_{me} is the magneto elastic anisotropy energy, and F_S is the surface anisotropy energy. The exchange anisotropy energy between domains has been neglected in this model.

The first term in (1), F_{mc} is the contribution to the total free energy associated with the magnetocrystalline anisotropy energy. Here, we assumed that the change in the magnetocrystalline anisotropy energy [3] does not contribute to the total free energy 1, i.e. $\Delta F_{mc} = 0$.

The coupling between electric-field and magnetization are driven by both of elastic strains, namely the strain-mediated ME coupling, and interfacial chemical bonding or the accumulation of spin-polarized screening charges at the interface called the interface-charged mediated ME effect [10]. For our particular interest, we however consider here the physics mechanism of FM/FE heterostructure driven by the former one which is more significant at large thickness, i.e. the film thickness is greater than a critical value [11]. In other words, we have not taken into account an interface-charge mediated ME effect, hence we will ignore the last term in (1), $F_S \approx 0$, in our calculation.

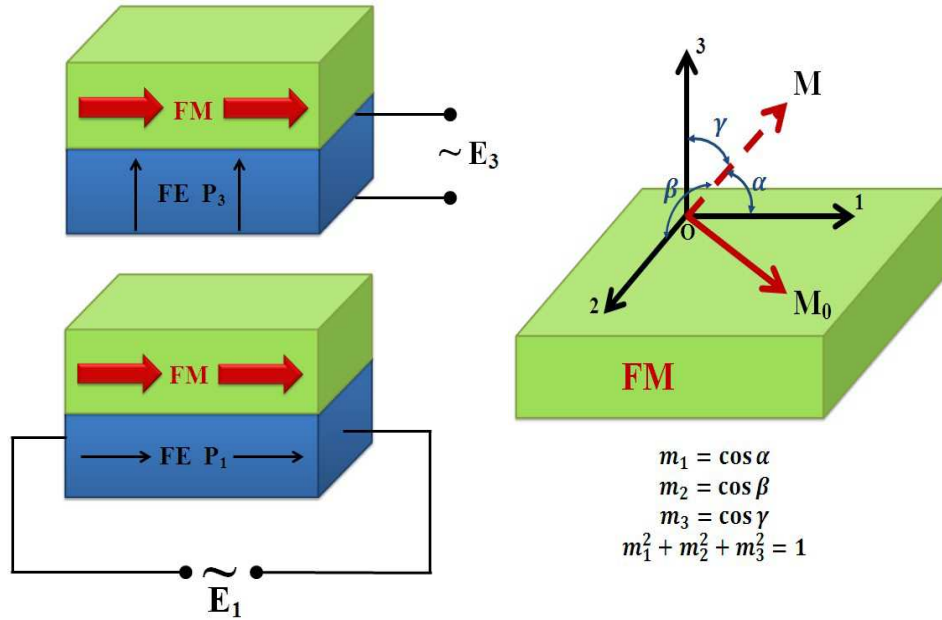


Fig. 1. A model of FM/FE layered heterostructure in two electric field modes: (a) a longitudinal electric field E_3 mode: an out of plane polarization P_3 can be generated by an external electric field E_3 ; (b) a transverse electric field E_1 mode: in plane polarization P_1 can be generated by an external electric field E_1 . The arrows denote the orientation of magnetization (polarization) in FM (FE) layer. (c) A diagram describes the spontaneous magnetization (magnetic easy axis) rotates from its initial in-plane direction \vec{OM}_0 to another direction \vec{OM} in FM film. 1, 2, and 3 are the elastic strains defined in the crystallographic reference frame; m_1 , m_2 , and m_3 are the direction cosines of the magnetic easy axis with respect to the principal cubic axis.

In the FE layer, an external electric field has been applied along the out-of-plane (i.e., longitudinal electric field E_3) or in-plane (i.e., transverse electric-field E_1) direction, respectively, as shown in Fig. 1. The in-plane strains are caused by the electric field through the converse magnetoelectric (ME) effect in both case. The total energy (1) may be to determine the thermodynamic equilibrium of the systems. In general, the spontaneous magnetization of the FM layer (easy axis)

rotates from its initial in-plane $\overrightarrow{OM_0}$ to another direction along with \overrightarrow{OM} (Fig. 1) above the critical external electric field [7, 8]. As we will show below, interestingly, temperature can assist this reorientation, indeed. For the sake of simplicity, the change in the Helmholtz free energy can be written as:

$$\Delta F(T) = F_H^M(T) - F_H^{M_0}(T) = \Delta F_{shape}(T) + \Delta F_{me}(T) \quad (2)$$

Indeed, the rotation process can only happen if the condition $\Delta F(T) < 0$ is satisfied. The effect of temperature on the EAR can be described through the study of the reorientation process of the magnetization at different electric field modes, i.e. E_3, E_1 . In the presence of an applied electric field, in plane elastic strains induced by the FE layer can be expressed as:

$$\varepsilon_{11} = \varepsilon_{22} = \varepsilon_0 + d_{31}E_3 + \alpha_T \Delta T \quad (3)$$

for a longitudinal electric field E_3 (see Fig. 1) and

$$\varepsilon_{11} = \varepsilon_0 + d_{33}E_1 + \alpha_T \Delta T \quad (4)$$

$$\varepsilon_{22} = \varepsilon_0 + d_{31}E_1 + \alpha_T \Delta T \quad (5)$$

for a transverse electric field E_1 (see Fig. 1). In which ε_0 is the original residual strain in the films without applying electric field.

This idea is motivated by the fact that temperature has a significant effect on the ME mechanical deformation [12–14]. The temperature, hence, appeared in (3), (4), (5) describes its impact on an elastic interaction which induces coupling between electricity and magnetic phenomenon. For simplicity, a perfect interface between FE and FM layers has been assumed, i.e. $\varepsilon_{11,22}^{FE} = \varepsilon_{11,22}^{FM}$. It is well-known that the critical electric field E^{cr} has been derived from the minimizing ΔF_{tot} with respect to direction cosine m_3 .

The detailed calculation for each term in (2) is referred to Appendix. We come up with the total free energy change ΔF_{tot} for both of the cubic as well as the hexagonal cases which can be written in a simple form as:

$$\Delta F_{tot} = -2td_{31}[E_3 - E_3^{cr}(T)]m_3^2 + K_B m_3^4 + \frac{1}{3}B_1 \Delta T \quad (6)$$

in the E_3 mode, and

$$\Delta F_{tot} = -t(d_{31} + d_{33})[E_1 - E_1^{cr}(T)]m_3^2 + K_B m_3^4 + \frac{1}{3}B_1 \Delta T \quad (7)$$

in the E_1 mode. By minimizing the energy change with respect to m_3 , we can derive the electric critical values, i.e. $E_3^{cr}(T), E_1^{cr}(T)$, which explicitly depends on temperature as follows:

$$E_3^{cr}(T) = -\frac{\varepsilon_0}{d_{31}} + \frac{K_A(T)}{2td_{31}} + \frac{(2t + B_1)\alpha_T}{2td_{31}} \Delta T$$

in the E_3 mode, and

$$E_1^{cr}(T) = -\frac{2\varepsilon_0}{(d_{31} + d_{33})} + \frac{K_A(T)}{t(d_{31} + d_{33})} + \frac{(2t + B_1)\alpha_T}{t(d_{33} + d_{31})} \Delta T$$

in the E_1 mode.

In the above equations $\Delta T = T - T_R$ describes the changing in temperature with respect to the room temperature $T_R = 300$ K; the constants $K_B = \frac{B_2^2}{c_{44}} - \frac{B_1^2}{c_{11}}, t = \frac{B_1}{2} + \frac{c_{12}B_1}{c_{11}}$ are for the cubic

FM and $K_B = \frac{B_2^2}{c_{44}} - \frac{B_1^2}{c_{33}}$, $t = \frac{B_1}{2} + \frac{c_{13}B_1}{c_{33}}$ are the constants in case of the hexagonal FM films. The material parameters, such as B_1 , B_2 , c_{11} , c_{33} , c_{44} have been taken from experiments [8]. Whereas, the temperature dependent term $K_A(T)$ is associated with the shape energy (Appendix). In the limit of $T \rightarrow T_R$, all the above results agree well with the well-known results calculated at T_R [7, 8].

As we have shown above, the critical electric field can be determined explicitly in temperature. In other words, the direction cosine m_3 plays a role as an order parameter. Moreover, we are here interested in showing the critical electric field change as the temperature moving away from the room temperature $T_R = 300$ K. For our better comparison, we therefore define the critical electric field change with respect to the one valued at room temperature as follows:

$$\Delta E^{cr}(\%) = \frac{|E^{cr}(T) - E^{cr}(T_R)|}{|E^{cr}(T_R)|} \quad (8)$$

We have found that the critical electric field change (8) is the same for both of a transverse electric field E_3^{cr} and the longitudinal electric field E_1^{cr}

$$\begin{aligned} \Delta E_1^{cr}(\%) &= \Delta E_3^{cr}(\%) \\ &= \frac{\left| -\frac{1}{2}\mu_0 M_S^2(0) \left[1 - \left(\frac{T_R}{T_C} \right)^{\frac{3}{2}} \right]^2 + \frac{1}{2}\mu_0 M_S^2(0) \left[1 - \left(\frac{T}{T_C} \right)^{\frac{3}{2}} \right]^2 - \left(\frac{c_{12}}{c_{11}} + 1 \right) B_1 \alpha_T \Delta T \right|}{\left| -\frac{1}{2}\mu_0 M_S^2(0) \left[1 - \left(\frac{T_R}{T_C} \right)^{\frac{3}{2}} \right]^2 + \frac{2B_1^2}{3c_{11}} - \frac{B_2^2}{c_{44}} \right|} \end{aligned} \quad (9)$$

$$\begin{aligned} &= \frac{\left| -\frac{1}{2}\mu_0 M_S^2(0) \left[1 - \left(\frac{T_R}{T_C} \right)^{\frac{3}{2}} \right]^2 + \frac{1}{2}\mu_0 M_S^2(0) \left[1 - \left(\frac{T}{T_C} \right)^{\frac{3}{2}} \right]^2 - 2 \left(\frac{c_{13}}{c_{33}} + 1 \right) B_1 \alpha_T \Delta T \right|}{\left| -\frac{1}{2}\mu_0 M_S^2(0) \left[1 - \left(\frac{T_R}{T_C} \right)^{\frac{3}{2}} \right]^2 + \frac{2B_1^2}{3c_{33}} - \frac{B_2^2}{c_{44}} \right|} \end{aligned} \quad (10)$$

As shown above, strikingly, $\Delta E_1^{cr}(\%)$ and $\Delta E_3^{cr}(\%)$ do not depend on the FE substrates parameters but the FM film parameters. In other words, the change of the critical electric field with temperature is universal for all three FE layers, i.e. Pb(Zn,Ti)O₃ (PZT), PZN-PT, BaTiO₃ (BTO) under the range of our interest. Therefore, for an illustration, the calculations are performed for two structures of the FM films: iron (Fe), nickel (Ni) and CFO - the cubic case and cobalt (Co) - the hexagonal case. The materials parameters of the FM films have been taken from the Ref. [8, 11].

III. THE EFFECT OF TEMPERATURE ON A CRITICAL INDUCED ELECTRIC FIELD

III.1. (001)-oriented cubic cases: Fe, Ni, CFO films

At room temperature $T_R = 300$ K (Fig. 2a), all features calculated for the electric field induced EAR in three FM layers (Fe, Ni, CFO) agree well with the previous studies [8]. For example, the quartic coefficient K_B has play a role as an order parameter, example $K_B > 0$, m_3 switches smoothly from zero to one whereas electric field induced EAR will suffer the discontinuous transition from zero to one at $E = E^{cr}$ if $K_B < 0$. When the temperature change occurs,

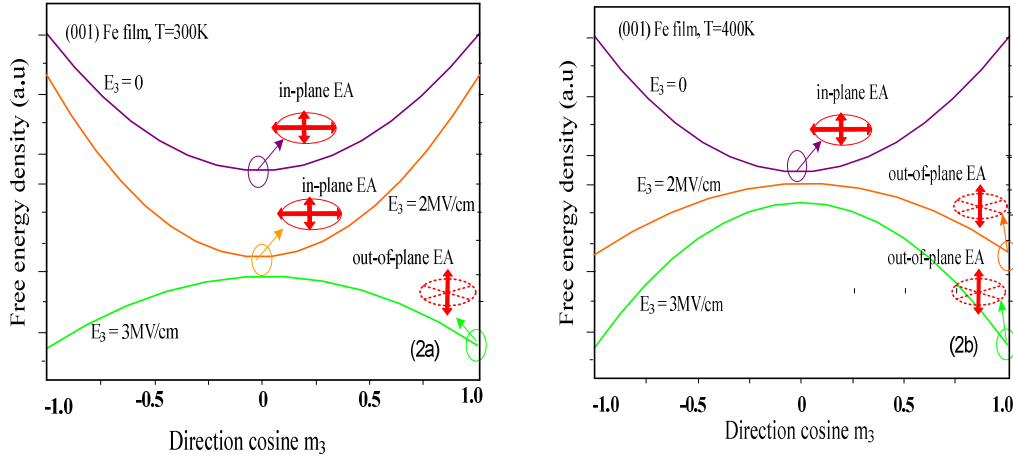


Fig. 2. Free energy change is plotted for a typical case: (001)-oriented cubic Fe-film under a longitudinal electric field E_3 as a function of the direction cosine m_3 with three different cases of $E_3 = 0$, $E_3=2\text{MV/cm}$ and $E_3=3\text{MV/cm}$: (a) at room temperature $T_R = 300\text{K}$ and a critical electric field $E^{cr}=2.2\text{MV/cm}$ (b) at temperature $T = 350\text{K}$ with a critical electric field $E^{cr}=2.0\text{MV/cm}$. The arrows within ellipses denote the magnetic easy axis and the opened ellipses with arrows indicate the minimum of free energy change.

the competition between the two terms (the second term and the third term) in the numerator of $E = E_1^{cr}$ and $E = E_3^{cr}$ lead to the two different physics mechanism: if the second term dominates, then the change in a critical electric field is negative. Otherwise, its change will be opposite if the third term dominates.

We start our investigation with Fe-film: Fig. 2 shows the electric field induced out of plane EAR in Fe films at two values of temperature, i.e. the room temperature $T_R = 300\text{K}$ and $T = 350\text{K}$. The stable orientation of the magnetic easy axis is indicated by the minima of the total energy change with respect to m_3 . At room temperature, when the electric field is less than the critical electric field E_3^{cr} , the favored direction of m_3 is the in-plane direction. Interestingly, the temperature has reduced the critical electric field, for example Fe-cubic films, from 2.2MV/cm (at $T_R = 300\text{K}$) [8] to 2.0MV/cm (at $T = 350\text{K}$) which is about 9% shown in Fig. 2b. In other words, at $T = 350\text{K}$ EAR has indeed occurred at the electric field $E_3 = 2\text{MV/cm}$ (the orange curve) whereas under the same electric field, EAR has not happened at the room temperature. This has demonstrated that an increase of temperature above T_R can assist the EAR, i.e. the critical electric field is lowered with an increase of temperature in the range of our interest. In the other side, i.e. temperature is reduced below T_R , we have observed the opposite physics mechanism which temperature has hindered the EAR. This has been confirmed by a similar study of ΔF_{tot} at temperature below T_R (not shown here). Significantly, the higher the temperature is, the lower the critical electric field is. For example, at $T = 400\text{K}$ the critical electric field has been reduced by 20% comparing to the one valued at T_R .

We conclude that the critical electric field E^{cr} is sensitive to the temperature change. Another evidence to support the above conclusion is illustrated in Fig. 3a. In this figure, we have

plotted a change of E^{cr} (the same for E_3^{cr} and E_1^{cr}) in the full range of temperature from room temperature 300 K up to the Curie temperature $-T_C$ for ferromagnetic thin films. It has been confirmed by a similar study of effect of the E^{cr} on temperature below T_R (not shown here). As we approach the Curie temperature, the change of curve displays a non-linear effect. However, we just pay our attention to the temperature range in which the linear behavior change of the critical electric field can be approximated. As we show in Fig. 3a for Fe film, the change of E^{cr} away from T_R is significant. In particular, $\Delta E^{cr}(\%)$ is about 9% at $T = 350$ K and 8.5% at $T = 250$ K, respectively. However, it becomes larger if temperature increases, for example $\Delta E^{cr}(\%)$ is about 9% at $T = 350$ K.

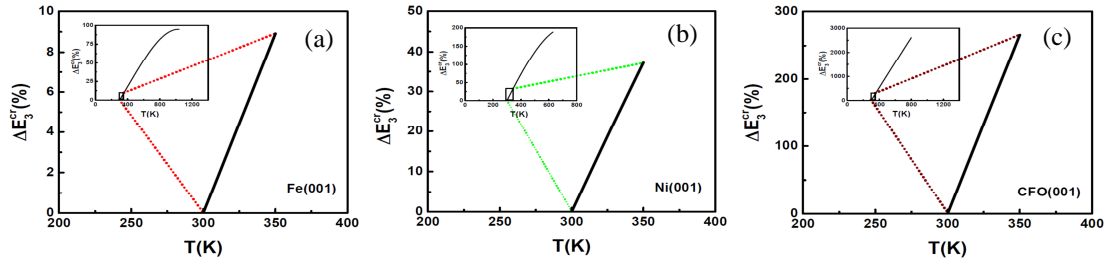


Fig. 3. A change of critical electric field with temperature for (a) (001)-oriented cubic Fe-film. The inset is a change of E^{cr} in the range of temperature from 300 K up to Curie temperature $T_C = 1043$ K indicated by an arrow, the rectangle narrows the range of temperature from 300 K to 350 K. (b) (001)-oriented cubic Ni-film. The inset is a change of E^{cr} in the full range of temperature from 300K up to Curie temperature $T_C = 631$ K indicated by an arrow. The rectangle narrows the range of temperature from 300 K to 350 K. (c) A negative change of critical electric field E^{cr} with temperature for (001)-oriented cubic CFO-film. The inset is a change of E^{cr} in the full range of temperature from 300 K up to Curie temperature $T_C = 793$ K indicated by an arrow. The rectangle narrows the range of temperature from 300 K to 350 K.

A surprised result has been seen as we study the electric field induced EAR of Ni and CFO films shown in Fig. 3b and Fig. 3c. The more obvious change has been even observed for Ni-films. In this figure, we see that the change of E^{cr} at $T = 350$ K is about 37% and 38% at $T = 250$ K, respectively. This change of E^{cr} is much larger than the change of E^{cr} observed in Fe-film. Indeed, the high magnetostrictive constants of Ni-films ($\lambda_S = -32,9 ppm$) is responsible for this observation. We should note that the magnetostrictive constants of Fe-films ($\lambda_S = -7.0$ ppm) is much lower than the one of Ni-films. Not surprisingly, the large change has been observed for CFO-films due to the very large magnetostrictive constant ($\lambda_S = -110$ ppm).

III.2. (0001)-oriented hexagonal cases: Co films

In this case, we have also studied a critical electric field induced EARs applied for the (0001)-oriented hexagonal cases, e.g. Co-films under the change of temperature. Interestingly, we have observed the similar features as found in single crystal FM film, i.e. Fe and Ni-films. As we shown in Fig. 4, we have illustrated that a temperature also assisted to the EARs reorientation as we increase temperature from T_R . Surprisingly, temperature has a minor effect on a E^{cr} , i.e. the critical electric change is about 5% at $T = 350$ K and 4% at $T = 250$ K, respectively. The

large but negative magnetoelastic coupling $B_1 = -8.1 \text{ MJ/m}^3$ [11] which can be described by the magnetostrictive constants λ_S is responsible for this surprised result.

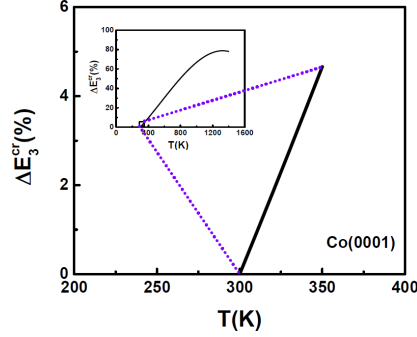


Fig. 4. A change of critical electric field with temperature for (0001)-oriented hexagonal Co-film. The inset is a change of E^{cr} in the range of temperature from 300K up to the Curie temperature $T_C = 1043\text{K}$ indicated by an arrow, the rectangle narrows the range of temperature from 300 K to 350 K.

IV. CONCLUSIONS

The temperature has a significant effect on the critical electric field induced easy axis reorientation in FM layer. We have investigated systematically these effects on two types of FM films which are the cubic film structure such as Fe, Ni, CFO as well as the hexagonal film structure such as Co. In the temperature range of our interest (250 - 350K), we have observed the linear effect of temperature on the critical electric field. Depending on the magnetoelastic coupling associated with the magnetostrictive constants of the FM films, the significant change of critical electric field has been observed. The higher the magnetoelastic coupling, the more sensitive the temperature dependence of the critical electric field is. The non-linear behavior starts to play an important role at the very high temperature which is close to T_C .

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APPENDIX

The contribution from shape anisotropy energy to the total free energy change can be written as:

$$\Delta F_{shape}(T) = F_{shape}^M(T) - F_{shape}^{M_0}(T) = \frac{1}{2} \mu_0 M_0^2 \left[1 - \left(\frac{T}{T_c} \right)^{\frac{3}{2}} \right]^2 m_3^2$$

The last term in (1) associated with the magneto elastic energy contributes to the total free energy change usually approximated by:

$$\begin{aligned} \Delta F_{me} = & B_1 \left(m_1^2 - \frac{1}{3} \right) (\epsilon_{11} + \alpha_T \Delta T) + B_1 \left(m_2^2 - \frac{1}{3} \right) (\epsilon_{22} + \alpha_T \Delta T) + B_1 \left(m_3^2 - \frac{1}{3} \right) \epsilon_{33} \\ & + B_2 (m_1 m_2 \epsilon_{12} + m_1 m_2 \epsilon_{13} + m_1 m_2 \epsilon_{23}) \end{aligned}$$

in which α_T is the thermal expansion coefficient, B_1 and B_2 are magneto elastic coupling coefficients, and ϵ_{ij} (i and $j = 1, 2, 3$) are elastic strains defined in the crystallographic reference frame. The thermal expansion coefficient in 3-direction has been ignored since the thermal expansion in 1-, 2- direction is much larger than 3-direction in the FM layer. Moreover, we have assumed that these coefficients are approximately constant and associated with the magnetostrictive constants, $\lambda_S = \lambda_{100} = \lambda_{111}$ of the polycrystalline FM films since $B_1 = -\frac{3}{2} \lambda_S (c_{11} - c_{12})$ and $B_2 = -3 \lambda_S c_{44}$ in which c_{11} , c_{12} and c_{44} are the elastic stiffness constants of the FM films. We have also used the pure elastic energy to derive the mechanical equilibrium conditions [8].

The change in elastic contribution ΔF_{el}^C (for the cubic films) and ΔF_{el}^H (for the hexagonal films) to the total free energy is written respectively as the follows:

$$\begin{aligned} \Delta F_{el}^C = & \frac{1}{2} c_{11} (\epsilon_{11}^2 + \epsilon_{22}^2 + \epsilon_{33}^2) + c_{12} (\epsilon_{11} \epsilon_{22} + \epsilon_{11} \epsilon_{33} + \epsilon_{22} \epsilon_{33}) \\ & + \frac{c_{44}}{2} (\epsilon_{12}^2 + \epsilon_{13}^2 + \epsilon_{23}^2) - \alpha_T \Delta T (3c_{12} + 2c_{44}) (\epsilon_{11} + \epsilon_{22}) \\ \Delta F_{el}^H = & \frac{1}{2} c_{11} (\epsilon_{11}^2 + \epsilon_{22}^2) + \frac{1}{2} c_{33} \epsilon_{33}^2 + c_{12} \epsilon_{11} \epsilon_{22} + c_{13} (\epsilon_{11} \epsilon_{33} + \epsilon_{22} \epsilon_{33}) + \frac{c_{44}}{2} (\epsilon_{13}^2 + \epsilon_{23}^2) \\ & + \frac{1}{4} (c_{11} - c_{12}) \epsilon_{12}^2 - \alpha_T \Delta T [(c_{11} + c_{12} + c_{13}) (\epsilon_{11} + \epsilon_{22})] \end{aligned}$$

in which c_{11} , c_{12} and c_{44} are the elastic stiffness constants of the FM films.