Voltage-induced strain control of the magnetic anisotropy in a Ni thin film on flexible substrate

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Voltage-induced magnetic anisotropy has been quantitatively studied in polycrystalline Ni thin film deposited on flexible substrate using microstrip ferromagnetic resonance. This anisotropy is induced by a piezoelectric actuator on which the film/substrate system was glued. In our work, the control of the anisotropy through the applied elastic strains is facilitated by the compliant elastic behavior of the substrate. The in-plane strains in the film induced by the piezoelectric actuation have been measured by the digital image correlation technique. Non-linear variation of the resonance field as function of the applied voltage is found and well reproduced by taking into account the non linear and hysteretic variations of the induced in-plane strains as function of the applied voltage. Moreover, we show that initial uniaxial anisotropy attributed to compliant substrate curvature is fully compensated by the voltage induced anisotropy.

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I. INTRODUCTION

Prospect of controlling local magnetization using electric fields, for low power and ultra fast new electronics has resulted in an amount of new research domains mainly focused on artificial engineered materials. Two decades ago, the condensed matter community began studying the interactions between a spin polarized current [1, 2] and magnetic domain walls in magnetic thin films. Percolation and domain wall excitation (precessional and/or translational) by spin angular momentum transfer from the current to the magnetic system pushed the study of new magnetic materials and advanced lithographed magnetic devices [3–5]. Since then, new magnetic configurations have been engineered by properly shaping magnetic nanostructures (for instance: U-shaped nanowires, focused ion beam nanobridges, giant magnetoresistance spin valve nanowires systems, magnetic racetrack memory devices). Unfortunately many experimental studies have indicated extremely low current-driven domain wall velocity and also non-adiabatic contributions to current-magnetization interaction, which introduce intrinsic threshold current for domain wall motion. Clearly, either the maximum possible speed of domain wall based driven recording and storage systems or power dissipation (directly linked to the current threshold) are not suitable for ultra-fast future electronics.

In order to overcome these important factors limiting the future miniaturization of integrated recording circuits based on simple magnetic nanostructured material, latest approaches start to involve intrinsic multiferroic and magnetoelectric artificial materials. Natural multiferroics have quite immediately shown a too weak magnetic order at room temperature making them useless for applications [6–8]. Using artificial nanostructures combining the two different orders (magnetic and electric one) in a unique system seems to be the only promising route. The easiest artificial architecture for a voltage control of the magnetization in those kinds of materials, appears to be a piezoelectric/magnetostrictive bilayers presenting a good strain-mediated coupling at the interface [9–11]. In order to achieve the highest magnetoelectric coupling for an efficient voltage manipulation of the magnetization, the magnetic device has to be encapsulated inside a piezoelectric environment which lead to two main difficulties. First, deposition of piezoelectric material generally needs high temperature and the most of the time also post annealing treatment under severe condition of oxidation. This latter could degrade the good magnetic properties of the magnetic layer. Second, the clamping effect due to the substrate, limits the stress applicable to the magnetization by the piezoelectric environment reducing perspective on applications. Indeed, in this kind of bilayers (piezoelectric/magnetostrictive), a significant strained induced magnetoelectric coupling is obtained only in the presence of non negligible in-plane strains. In order to avoid both latters issues, inverse magnetoelectric effect has been studied by depositing ferromagnetic thin films on a piezoelectric substrate [12–14] or by directly gluing (via an epoxy glue) a (thin film, rigid substrate) magnetic system onto a piezoelectric actuator [20–23].

Noticeably, Brandlmaier et al. [20] have shown that film strains are much smaller (50%) than the expected values in case of a complete strain transmission through the cement and the rigid substrate.

In this paper, voltage induced in-plane magnetic anisotropy has been quantitatively studied in a magnetostrictive film deposited on a flexible substrate and glued onto a piezoelectric actuator. For this study, Nickel has been chosen due to its well-known negative effective magnetostriction coefficient at saturation even in a polycrys-
talline film with no preferred orientations. Digital Image Correlation (DIC) and Micro-Strip FerroMagnetic Resonance (MS-FMR) techniques have been employed to i) quantitatively measure the in-plane strains transmitted to the ferromagnetic film responsible of the inverse magnetoelastic effect occurring in the fabricated biferroic structure and to ii) experience the voltage-driven changes of the in-plane magnetic anisotropy.

II. EXPERIMENTAL DETAILS

A 200 nm-thick Nickel film was deposited on a flexible (Kapton® [24]) substrate (125 µm) by radio frequency sputtering at room temperature. The Ni film was found to be polycrystalline with no strong preferred orientations by x-ray diffraction. After deposition, the film/substrate system was glued onto a piezoelectric actuator. A flexible compliant substrate (Young’s modulus \( E = 4 \) GPa) has been chosen in order to avoid clamping effects which can occur with rigid substrates (for example, \( E = 180 \) GPa for Silicon) and thus to have maximum strain transmission in between the piezoelectric actuator and the film [20, 21]. Figure 1 shows a sketch of the Ni thin film and of the structure allowing voltage-control of the in-plane magnetic anisotropy. Two different axes systems are defined. The first one \((x, y, z)\) is attached to the Ni film while the second one \((X, Y, Z)\) describes the piezoelectric actuator geometry. \( \phi_H \) (resp. \( \phi_{HH} \)) is the angle between \( x \) (resp. \( X \)) axis and the applied magnetic field \( \vec{H} \); the magnetization \( \vec{M} \) is defined by its polar angles \((\theta, \varphi)\) as usual [25].

All the measurements presented here were performed at room temperature using both microstrip ferromagnetic resonance (MS-FMR) and a digital image correlation (DIC) technique. MS-FMR was employed to probe the influence of the applied voltage on the magnetic properties via the resonance field of the uniform precession mode. The DIC was used to quantitatively measure the in-plane strains \((\varepsilon_{XX} \text{ and } \varepsilon_{YY})\) induced by the piezoelectric actuator at the surface of the ferromagnetic film.

MS-FMR experiments allowed the resonance field \( H_{res} \) probing by sweeping the applied magnetic field in presence of a fixed pumping Radio Frequency (RF) field \( H_{RF} \). In this kind of RF-resonance technique, a weak modulation of the applied field \((\sim 2 - 5 \text{ Oe at } 1480 \text{ Hz})\) is performed in order to optimize the signal to noise ration for a better lock-in detection of the signal. Thus, this technique gives access to the first field derivative of the RF absorption as function of the applied field. A more detailed presentation can be found elsewhere [26].

DIC is a technique which allows displacement and strain fields measurements on an object surface by capturing images of the object surface at different states [27, 28]. One state is recorded before applying voltage, i.e. the reference image, and the other states are subsequent images of the deformed object. DIC generally uses random patterns of gray levels of the sample surface to measure the displacement via a correlation of a pair of digital images. In this study, the film surface was spray-painted with a speckle pattern to generate a contrast in the uniform specimen face. Hence, from the measured strain fields we can estimate the mean in-plane strains \( \varepsilon_{XX} \text{ and } \varepsilon_{YY} \), that cannot be straightforwardly attained with other techniques such as x-ray diffraction [29].

III. RESULTS AND DISCUSSION

In the first paragraph of this section we present the magnetic parameters of the Ni film at zero applied voltage. The second part is devoted to a quantitative measure of the in-plane strains \((\varepsilon_{XX} \text{ and } \varepsilon_{YY})\) at the surface of the Ni film using the DIC technique. In the third we present the analytical model allowing the conversion of the applied voltage into strains, which we developed to analyze the influence of the applied voltage on the magnetic properties of the film observed by MS-FMR.

A. Magnetic parameters at zero applied voltage

MS-FMR experiments in absence of applied voltage were performed in order to determine the magnetic parameters of the Ni thin film. Indeed, at zero voltage, the frequency of the uniform mode is expected to only depend on the gyromagnetic factor \( \gamma \), on the magnetization at saturation \( M_S \) and on the possible presence of magnetic anisotropies [30]. Figure 2a shows the frequency variation of the uniform mode as function of the magnetic field applied either along \( x \) direction (open symbols) either along \( y \) direction (filled symbols). The angular dependence of the resonance field of the uniform mode at 10 GHz is also presented in Figure 2b. A non negligible uniaxial anisotropy of magnitude \( \sim 310 \text{ Oe} \) is
observed along x direction (i.e., \( \varphi_H = 0 \)) corresponding to a minimum resonance field. Since the studied Ni film is polycrystalline with no preferred crystallographic orientations, no macroscopic magnetostrictive effect is expected, thus this uniaxial anisotropy should have a magnetoelastic origin. Indeed, as the film is deposited on a flexible substrate, a slight curvature along a given direction during the elaboration process should lead to a non-zero magnetoelastic anisotropy of the maintained flat sample during FMR measurements, as suggested by Zhang et al. [33]. In this work, this uniaxial anisotropy will be fitted by using an ad hoc uniaxial anisotropy characterized by an anisotropy constant \( K_U \). From now, easy axis will refer to x direction since the angle between x and the uniaxial anisotropy is close to zero. Thus, the lines (continuous and dashed) in Figure 2 correspond to fits using the following expression obtained with the assumption of an in-plane magnetization (i.e., \( \theta = \frac{\pi}{2} \)):

\[
f^2 = \left( \frac{\gamma}{2\pi} \right)^2 \left( H_{\text{res}} \cos(\varphi - \varphi_H) + \frac{2K_U}{M_S} \cos 2\varphi \right) \left( H_{\text{res}} \cos(\varphi - \varphi_H) + \frac{2K_U}{M_S} \cos^2 \varphi + 4\pi M_S \right)
\]

(1)

Where \( f \) is the microwave driven frequency. It results from the analysis of the data that the magnetic parameters are close to the one of bulk Ni [32]. A magnetization at saturation (\( M_S \)) of 480 emu.cm\(^{-3}\) (4\( \pi \)\( M_S \) \( \approx \) 6030 G) with a gyromagnetic factor (\( \gamma \)) of 1.885 \( \times \) 10\(^7\) Hz.Oe\(^{-1}\) are found. The uniaxial anisotropy constant is estimated to \( K_U = 7.3 \times 10^4 \) erg.cm\(^{-3}\) (we define \( H_U = 2K_u/M_S \approx 315 \) Oe). These measurements performed at zero voltage will serve as reference in order to evaluate the changes related to the in-plane strains induced by the piezoelectric actuator.

Figure 2: a) Frequency variation of the uniform mode as function of the in-plane applied magnetic field at zero applied voltage. Open symbols are obtained with \( \varphi_H = 0 \) (\( \vec{H} \parallel \hat{x} \)) and the filled ones are obtained with \( \varphi_H = \pi/2 \) (\( \vec{H} \parallel \hat{y} \)). b) In-plane angular dependence of the resonance field at 10 GHz. The solid and dashed lines in a) and b) are best fits to the experimental data using the following parameters: \( \gamma = 1.885 \times 10^7 \) Hz.Oe\(^{-1}\), \( M_S = 480 \) emu.cm\(^{-3}\), \( K_U = 7.3 \times 10^4 \) erg.cm\(^{-3}\) and equation (1).

B. Quantitative in-plane strains measurements by DIC

DIC technique has been used to measure the in-plane strains \( (\varepsilon_{XX} \text{ and } \varepsilon_{YY}) \) at the surface of the film. The measurements have been performed by varying the external voltage from 0 V to 100 V (filled symbols) and back to 0 V (open symbols) with steps of \( \sim 5 \) V. Note that the first 0 V image was taken after applying a “saturating” voltage of 100V in order to avoid hysteresis effects which can occur after several hours at zero voltage. The measured \( \varepsilon_{XX} \) and \( \varepsilon_{YY} \) as function of the applied voltage are shown in Figure 3 and reach respectively \( 10^{-3} \) and \( -0.5 \times 10^{-3} \) at 100 V. Indeed \( \varepsilon_{XX} \) is found to be positive while \( \varepsilon_{YY} \) is found to be negative with a ratio \( \frac{\varepsilon_{YY}}{\varepsilon_{XX}} \approx 0.5 \). Non linear variations for both \( \varepsilon_{XX} \) and \( \varepsilon_{YY} \) with the voltage is observed due to the intrinsic properties of the ferroelectric material used in the fabrication of the actuator [33]. This behavior is well reproducible after “saturating” the actuator a first time (at 100 V in this work). It should be noted that these values correspond to mean values calculated in area close to \( 4 \times 4 \) mm\(^2\) (area of the thin film). However, the heterogeneities of \( \varepsilon_{XX} \) and \( \varepsilon_{YY} \) can be neglected compared to their “absolute” values and even at the edge of the \( 4 \times 4 \) mm\(^2\) area. Consequently, the measured \( \varepsilon_{XX} \) and \( \varepsilon_{YY} \) can be considered as homogeneous in \( XY \) plane. Moreover, the estimated in-plane shear strains by DIC were found to be negligible.

In addition, the analysis of \( \varepsilon_{XX} \) and \( \varepsilon_{YY} \) on an uncoated area of the actuator gives same values. Thus, the
transmission of the in-plane strains between the actuator and the surface of the film is close to 100%. Therefore, the values of $\varepsilon_{XX}$ and $\varepsilon_{YY}$ shown in Figure 3 should be homogeneous in the thickness, \textit{i.e.} the strains are well transmitted through the cement and through the film/substrate interface. This result is consistent with previous studies performed on metallic films of comparable thicknesses deposited onto flexible substrates and analyzed both by DIC and x-ray diffraction techniques \cite{28, 34}.

C. Voltage induced magnetic anisotropy probed by MS-FMR

The voltage induced anisotropy has been studied in two different configurations defined by the angle between the easy axis and $X$ direction (see Figure 4). In those configurations, the easy axis is either aligned along $Y$ (first configuration, Figure 4a) either along $X$ (second configuration, Figure 4b). As previously mentioned, the Ni film is polycrystalline with no preferred orientations. This allows to define an effective isotropic magnetostrictive coefficient at saturation $\lambda$ \cite{25}. In these conditions and because of the opposite sign of $\varepsilon_{XX}$ and $\varepsilon_{YY}$ values, an additional in-plane anisotropy field parallel to $Y$ direction is expected (in the range 0-100V). Consequently, the initial easy axis is expected to be increased in the first configuration while a competition between the initial in-plane anisotropy and the voltage induced one will takes place in the second configuration.

The influence of the applied voltage on the magnetic properties of the thin film has been probed by MS-FMR technique. Figures 5a and 5b show typical MS-FMR experimental spectra recorded at 10 GHz with an applied magnetic field along the initial hard axis ($i.e.$ $\phi_H = 0$ for the first configuration and $\phi_H = \frac{\pi}{4}$ for the second configuration) and for different applied voltages (0, 30, 60 and 100 V). Note that the experimental conditions (in term of applied voltage) were same than for the DIC study. Moreover, in order to avoid non linear variations of $\lambda$ as function of the applied magnetic field \cite{35}, the experimental spectra have been recorded at 10 GHz. In these conditions, the deduced resonance fields are in a magnetic saturating regime (as shown in Figure 2).

It clearly appears that the resonance field increases (resp. decreases) with the applied voltage in the first (resp. second) configuration. This indicates that the initial uniaxial anisotropy is either reinforced in the first configuration either softened in the second one. This is consistent with a negative magnetostriction coefficient \cite{17}. The deduced resonance fields as function of the applied voltage are presented in Figures 5c and 5d. Here, filled symbols represent the upswEEP (0 to 100 V) and open symbols the downsweep (100 to 0 V). The solid and dashed lines are best fits to the experimental data by using an analytical expression deduced from equations 2-4. An isotropic magnetostriction coefficient of $-26 \times 10^{-6}$ is deduced.

In order to quantitatively bind the resonance field variations to the in-plane strains induced by the applied voltage, we can calculate the uniform precession mode
Figure 6: Resonance field as function of the applied voltage at 10 GHz for different $\varphi_H$ values ($0, \pi/4$ and $\pi/2$). Filled symbols correspond to the experimental data obtained from the first configuration (Figure 4a) while open symbols are obtained from the second configuration (Figure 4b). The dashed and continuous lines are fits to the experimental data obtained by using an analytical expression deduced from equations 2-4. A magnetostrictive coefficient at saturation of magnitude $\lambda = -26 \times 10^{-6}$ is deduced.

frequency as function of the applied voltage (strain) by adding a magnetoelastic energy term $F_{ME}$ to the total magnetic energy density $F$ of the Ni film. In an isotropic case, which is closely ours (non-textured Ni film), $F_{ME}$ energy term can be written as:

$$F_{ME} = -\frac{3}{2} \lambda \left( (\gamma_x^2 - \frac{1}{3}) \sigma_{xx} + (\gamma_y^2 - \frac{1}{3}) \sigma_{yy} \right)$$  \hspace{1cm} (2)

$\sigma_{xx}$ and $\sigma_{yy}$ being the in-plane principal stress tensor components while $\gamma_x$ and $\gamma_y$ correspond to the direction cosines of the in-plane magnetization. Moreover, the relation between the principal components of stress and strain tensors is given by the following equations where $E$ is the Young’s modulus and $\nu$ is the Poisson’s ratio:

$$\sigma_{xx} = \left( \frac{E}{1+\nu} \right) \left( \frac{1}{1-\nu} \varepsilon_{xx} + \frac{\nu}{1-\nu} \varepsilon_{yy} \right)$$ \hspace{1cm} (3)

$$\sigma_{yy} = \left( \frac{E}{1+\nu} \right) \left( \frac{1}{1-\nu} \varepsilon_{yy} + \frac{\nu}{1-\nu} \varepsilon_{xx} \right)$$ \hspace{1cm} (4)

The voltage dependence of the uniform mode resonance field is calculated by using the following relation

$$\left( \frac{2\pi f_0}{\gamma} \right)^2 = \frac{1}{(M \sin \theta)^4} \left( \frac{\partial^2 F}{\partial \sigma^2} \sigma^2 - (\frac{\partial^2 F}{\partial \sigma \sigma^2})^2 \right).$$ \hspace{1cm} (5)

Assuming a Young’s modulus of $2 \times 10^{11}$ dyn.cm$^{-2}$ ($\approx 200$ GPa) and a Poisson’s ratio of 0.3, the only undetermined parameter is $\lambda$. Introducing the experimentally found voltage dependence of $\varepsilon_{XY}$ and $\varepsilon_{YY}$, the best fits of all the experiments performed in the both studied configurations gives $\lambda = -26 \times 10^{-6}$. It should be noted that this value is consistent with commonly found values for non-textured polycrystalline Ni film (about $-30 \times 10^{-6}$) and validate our overall approach. It should be noted here that the polycrystalline theoretical value can be found by applying an arithmetic average of the analytical solutions given by Reuss and Voigt models [23]:

$$\lambda_{Reuss} = \frac{2}{5} \lambda_{100} + \frac{3}{5} \lambda_{111} \hspace{1cm} (5)$$

$$\lambda_{Voigt} = \frac{2}{2+3A} \lambda_{100} + \frac{3A}{2+3A} \lambda_{111} \hspace{1cm} (6)$$

Where $\lambda_{100}$ and $\lambda_{111}$ are respectively the magnetostriction coefficients of $<100>$ and $<111>$ directions families (respectively equal to $-51 \times 10^{-6}$ and $-22.4 \times 10^{-6}$), $A$ is the Zener anisotropy index ($A = 2C_{44}/(C_{11}-C_{12}) \approx 2.6$ for Ni). [23]

Note that the hysteretic and non-linear variations of the resonance field as function of the applied voltage is well reproduced by the analytical model proposed.

Figure 6 shows variations of the resonance field as function of the applied voltage for different $\varphi_H$ angles ($0, \pi/4$ and $\pi/2$). Here, open squares correspond to experimental data obtained from the first configuration while filled circles are obtained from the second configuration. The measurements coming from the first configuration indicate that the voltage induced anisotropy is parallel to the initial uniaxial one and consequently the magnitude of the easy axis along $Y$ is increased. This behavior is observed via the difference between the resonance fields measured at $\varphi_H = 0$ and $\varphi_H = \pi/2$ with $2H_U \sim 600$ Oe at zero voltage and around 1400 Oe at 100 V. Thus, the magnitude of the easy axis at 100V is equal to more than twice time the measured value in the absence of voltage. The second configuration is more interesting since a competition between the initial uniaxial anisotropy and the voltage induced one takes place. It results that the initial anisotropy is totally compensated at around 80 V (i.e. $H_{ME} = H_U$, where $H_{ME}$ being the induced magnetoelastic anisotropy field) which leads to an isotropic in-plane behavior of the resonance field. Interestingly, at 100V a non zero easy axis perpendicular to the initial one at 0 V is observed.

The solid and dashed lines in Figure 6 are fits to the experimental data calculated by using the parameters previously given. One can note the good agreement between the experimental measurements and the calculated lines.

Note that a non-negligible overestimation of the magnetostrictive coefficient can occurs if we neglect the influence of $\varepsilon_{YY}$ (i.e. assumption of a uniaxial strain). Indeed, the calculated lines (not shown here) in this approximation are quasi similar to the one previously calculated. However, an overestimation of $\lambda$ is performed
(λ = −40 × 10^{-6} instead of −26 × 10^{-6}). Thus, the present study also demonstrates that a full characterization of the in-plane strains is needed if we want to quantitatively analyzed the inverse magnetoelectric effect occurring inside such structures.

IV. CONCLUSION

- Magnetic anisotropy in Nickel thin films grown on Kapton® substrate has been studied by microstrip ferromagnetic resonance. An “initial” uniaxial anisotropy has been found and is most probably related to the non-flatness of the compliant substrate during deposition.

- Piezo-actuation of Ni/polyimide system has been performed allowing a voltage-induced strain control leading to a change of the total magnetic anisotropy.

- An accurate study by digital image corelation technique allowed a good knowledge of in-plane macroscopic strains at the surface of the film as function of applied voltage to the piezoelectric actuator. This technique that allows high precision (about 5 × 10^{-5}) is found to be relevant for determining in-plane strains in the elastic domain.

- A comparison of strains of the film with that of the piezoelectric actuator demonstrates the full transmission of in-plane elastic strains across the whole system, which is due to the compliant behavior of the polyimide substrate.

- A simple analytical model taking into account of the measured strains as function of applied voltage well matches the experimental data. The only adjusted parameter is the effective magnetostrictive coefficient at magnetic saturation of Ni film and is found to be close to previously published values.

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