Optimization of indirect magnetoelectric effect in thin-film/substrate/piezoelectric-actuator heterostructure using polymer substrate

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Indirect magnetoelectric effect has been studied in magnetostrictive-film/substrate/piezoelectric-actuator heterostructures. Two different substrates have been employed: a flexible substrate (Young’s modulus of 4 GPa) and a rigid one (Young’s modulus of 180 GPa). A clear optimization of the indirect magnetoelectric coupling, studied by micro-strip ferromagnetic resonance, has been highlighted when using the polymer substrate. However, in contrast to the rigid substrate, the flexible substrate also leads to an a priori undesirable and huge uniaxial anisotropy which seems to be related to a non equibiaxial residual stress inside the magnetostrictive film. The “strong” amplitude of this non equibiaxiality is due to the large Young’s modulus mismatch between the polymer and the magnetostrictive film which leads to a slight curvature along a given direction during the elaboration process and thus to a large magnetoelastic anisotropy.

Prospect of local magnetization control using an electric field or a pure voltage, for low power and ultrafast new electronics has resulted in an amount of new research domains mainly focused on artificial engineered materials[1–4]. Artificial magnetoelectric systems seems to be a promising route for such control. The easiest artificial architecture for a magnetization voltage control in those kinds of materials appears to be the piezoelectric/magnetostrictive bilayers presenting a good strain-mediated coupling at the interface [4–12]. However, in these systems, clamping effects due to the substrate limits the strain applicable by the piezoelectric environment to the magnetization and therefore reduces perspective on applications. Indeed, in this kind of bilayers, a significant magnetoelectric coupling is obtained only in the presence of negligible in-plane stresses in the magnetic media. Therefore, the elastic strains are transferred at the interfaces of such systems, more this indirect magnetoelectric effect is optimized. Generally, the magnetic thin films are first deposited on a thick substrate (~hundreds of microns), which is then cemented on a piezoelectric actuator[9–10][13–14]. This process limits the desired phenomenon, especially when the substrate is stiff such as for commonly used wafer (Si, GaAs, ...). This often reported limitation[14] (a few ten percents of losses in best cases) can be avoided by depositing the magnetic thin film on a compliant substrate such as polyimides[10] that are more and more used in flexible spintronics[16].

The present study presents a quantitative comparison of the obtained effective MagnetoElectric (ME) coupling in two different artificial magnetoelectric heterostructures characterized by the presence of a flexible or a rigid substrate between a ferromagnetic film and a piezoelectric actuator. The effective ME coupling will be deduced from in situ MicroStrip FerroMagnetic Resonance (MS-FMR). The artificial ME heterostructures are composed of a 200 nm Nickel film deposited by radio frequency sputtering either on a rigid substrate (Silicon of thickness 500 µm) or on a flexible one (Kapton® of thickness 125 µm) and then glued onto a piezoelectric actuator as presented on Figure 1. It is worth mentioning that Ni material has been chosen because of its well-known negative effective magnetostriiction coefficients at saturation even in a polycrystalline film with no preferred orientations, which is closely the situation for both systems. In this condition, only one magnetostriction coefficient at saturation (λ) is sufficient to characterize the magnetoelastic anisotropy. Moreover, the two substrates have been chosen because of their Young’s modulus (~4 GPa for Kapton® as compared to ~180 GPa for Si).

MS-FMR experiments have been performed at a fixed frequency by sweeping the applied magnetic field from 0 to 3 kOe[14]. The microwave driven frequency was fixed at 8 GHz in all the presented experiments, where the resonance fields are sufficiently higher than the in-plane magnetic anisotropies fields present in the structures, avoiding undesirable magnetic and magnetoelastic hysteresis effects. The in situ applied voltage experiments have been done by varying the external voltage from 0V to 100V and back to 0V with step of around 5 V. Figure 2 shows typical MS-FMR spectra for two different applied voltages (0 and 100V). The magnetic field is applied along y direction which corresponds to a...
negative induced in-plane strains. Indeed, in the studied range of applied voltage [0-100V], the piezoelectric actuator presents a main positive in-plane strain along x direction ($\varepsilon_{xx} > 0$) and a smaller negative one along y direction ($\varepsilon_{yy} < 0$ with $\varepsilon_{xx} \sim -2\varepsilon_{yy}$) [11]. We observed that in both structures the resonance field decreases as a function of the applied voltage. This behavior is consistent with the negative magnetostriction coefficient at saturation of Ni material ($\lambda < 0$). Indeed, in first approximation, the magnetoelastic anisotropy can be viewed as a voltage-induced uniaxial magnetoelastic anisotropy field $\vec{H}_{ME}(V)$ along y direction. Thus, since the in situ MS-FMR experiments are performed along y, this y-axis will be easier (when increasing the applied voltage) for the magnetization direction leading to lower values of the resonance field. Figure 2 shows a clear shift of the resonance field $\delta H_R$ (defined as $\delta H_R = H_R(0) - H_R(V)$) between 0 V and 100 V for both heterostructures: it is close to 50 Oe for Si substrate and reaches a value around 350 Oe when a flexible substrate is used.

Figure 2: Experimental spectra recorded at 8 GHz for an in-plane magnetic field at 90 degree with respect to the main positive strain axis of the actuator (along y-axis). The largest shift of the resonance is clearly put into evidence for the Ni film deposited onto Kapton®.

The complete $\delta H_R$ variations as function of the applied voltage are reported in Figure 3 where circles (resp. squares) refer to results obtained for Si (resp. Kapton®) substrate. A non linear and hysteretic variation is put into evidence when using Kapton® as a substrate whereas an almost linear dependence is found in the second structure indicating a difference of the strain transmission efficiency. This non linear and hysteretic behavior is due to the intrinsic properties of the ferroelectric material used during piezoelectric actuator fabrication [11]. However, in first approximation, if this non linear and hysteretic variation of $\delta H_R$ is neglected and adjusted by a linear fit, an effective magnetoelastic coupling $\alpha_{ME}$ (in V.cm$^{-1}$.Oe$^{-1}$ being given the width of the piezoelectric actuator (0.7 cm)) can be estimated. The linear adjustments to the experimental data are presented in Figure 3. A strong magnetoelectric coupling is found for the Ni/Kapton®/Piezoelectric system with an effective magnetoelectric coefficient $\alpha_{ME} \sim 0.2$ V.cm$^{-1}$.Oe$^{-1}$. Interestingly, in the case of the Ni/Si/Piezoelectric system, it can be conclude that the magnetoelectric coupling is roughly seven times less efficient because of its higher magnetoelectric coefficient value ($\alpha_{ME} \sim 1.35$ V.cm$^{-1}$.Oe$^{-1}$). The relatively weak coupling found in the second system is due to a weaker transmission of the in-plane elastic stresses. The strain loss of a factor of almost 7 cannot be explained only by the stiffness of Silicon but also by the often reported imperfection of cementation of the epoxy glue[14]. Obviously these imperfections are insignificant when the substrate is highly stretchable like polymers.

However, depositing a magnetic thin film on a flexible substrate can lead to an a priori undesirable effects related to its possible anisotropic non-flatness during elaboration process. Moreover, because of the large contrast between the polymer and the Ni Young’s modulus (~4 GPa and ~200 GPa, respectively), a more or less pronounced additional curvature due to growth stress develops during film deposition. These phenomena lead to a stress state in the magnetic film with a possible in-plane non-equibiaxiality[17]. This behavior is illustrated in Figure 4 showing in-plane angular dependencies (thereafter $\varphi_H$ is the angle between the in-plane applied magnetic field and x-axis) of the resonance field of the as-deposited structures. A huge uniaxial anisotropy (anisotropy field close to 300 Oe) characterized by a horizontal peanut shape of the resonance field variation is observed when the Ni film grown on Kapton® whereas a very weak one (anisotropy field around 30 Oe) is found when using the Si substrate. Since the present Ni films are polycrystalline with no preferential crystallographic...
constant shift in the resonance field baseline because these fits allow an evaluation of the in-plane non-
residual stress tensor components while effects are expected in both cases. Thus this uniaxial
orientations, no in-plane macroscopic magnetocrystalline
anisotropy should have a magnetoelastic origin. The
fluence of in-plane residual stresses on the magnetic properties of the as deposited films can be modeled using an
isotropic magnetoelastic anisotropy energy term $F_{ME}$:

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

$$\sigma_{xx}^{\text{residual}}$$ and $\sigma_{yy}^{\text{residual}}$ being the in-plane principal residual stress tensor components while $\gamma_x$ and $\gamma_y$ correspond to the direction cosines of the in-plane magnetization. In this condition the resonance field expression can be written as $H_R = H_1 + H_2$ with:

$$H_1 = \left[ \left( 2\pi M_S + \frac{3\lambda}{2M_S} \sigma_{xx}^{\text{residual}} \sin^2 \varphi_H \right. \right.$$

$$+ \sigma_{yy}^{\text{residual}} \cos^2 \varphi_H \left. \right)^2 + \left( \frac{2\pi f}{\gamma} \right)^2 \right]^{0.5} - 2\pi M_S$$

$$H_2 = -\frac{3\lambda}{4M_S} \left[ \sigma_{xx}^{\text{residual}} (1 + 3 \cos 2\varphi_H) \right.$$

$$+ \sigma_{yy}^{\text{residual}} (1 - 3 \cos 2\varphi_H) \right]$$

In the above expressions $H_1$ essentially represents a constant shift in the resonance field baseline because $2\pi M_S$ ($M_S$ being the saturation magnetization) and $\frac{2\pi f}{\gamma}$ ($\gamma$ is the gyromagnetic factor) are found to be larger than the equivalent magnetoelastic anisotropy field. The influence of in-plane residual stresses on the magnetic properties is included in $H_2$ term. It should be noted that this expression is obtained in the assumption of a uniform magnetization (macropin approximation) aligned along the applied magnetic field. This assumption is well fulfilled at the 8 GHz driven frequency chosen for this study. It clearly appears that an equibiaxial residual stress ($\sigma_{xx}^{\text{residual}} = \sigma_{yy}^{\text{residual}}$) leads to an isotropic variation of the resonance field with a slight increase or decrease of the mean value depending on the sign of the residual stress. However, a non equibiaxial residual stress ($\sigma_{xx}^{\text{residual}} \neq \sigma_{yy}^{\text{residual}}$) leads to an anisotropic angular variation of the resonance field. Thanks to this model, the experimental angular variations of the resonance field have been fitted (see continuous lines in Figure 4) by using usual Ni material magnetic parameters: $M_S = 480$ emu.cm$^{-3}$, $\gamma = 1.885 \times 10^7$ Hz.Oe$^{-1}$ and $\lambda = -26 \times 10^{-6}$ [10]. The deduced non-equbiaxiality is $|\sigma_{xx}^{\text{residual}} - \sigma_{yy}^{\text{residual}}| = 200$ MPa and $|\sigma_{xx}^{\text{residual}} - \sigma_{yy}^{\text{residual}}| = 15$ MPa for the Ni/Kapton® deposits onto flexible and rigid substrate, respectively. These results show that MS-FMR experiments can also be used for the determination of the non-equibiaxial residual stress in magnetostrictive films. It should be noted here that this kind of knowledge is not straightforward to get with standard techniques (sample deflection, X-ray diffraction), for which an equibiaxial stress state is generally assumed. Indeed, this parameter cannot be neglected since it determines the initial magnetization direction in the magnetic thin film being considered.

In conclusion, an optimization of the indirect magnetoelastic coupling in thin-film/piezoelectric system/substrate/piezoelectric actuator heterostructure has been performed by employing a polymer as a substrate. This optimization is due to a better strain transmission between the piezoelectric actuator and the Ni film which is due to the weak Young’s modulus of the Kapton®. Furthermore, at zero applied voltage, a huge magnetoelastic anisotropy is evidenced in the Ni/Kapton®/Piezoelectric system. It is attributed to a “residual” non-equibiaxial stress state due to a slight curvature along a given direction which generally appears when depositing a metallic film onto a flexible substrate. However, this effect could be a limitation for some applications where weak in-plane magnetic anisotropies are required.

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