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### Thin film magnetoelectric composites near spin reorientation transition

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### ABSTRACT

We report the use of a magnetic instability of the spin reorientation transition type to enhance the magnetoelectric sensitivity in magnetostrictive–piezoelectric structures. We present the theoretical study of a clamped beam resonant actuator composed of a piezoelectric element on a passive substrate actuated by a magnetostrictive nanostructured layer. The experiments were made on a polished 150  $\mu$ m thick 18  $\times$  3 mm<sup>2</sup> lead zirconate titanate (PZT) plate glued to a 50  $\mu$ m thick silicon plate and coated with a giant magnetostrictive nanostructured Nx(TbCo<sub>2 5nm</sub>/FeCo<sub>5nm</sub>) layer. A second set of experiments was done with magnetostrictive layer deposited on PZT plate. Finally, a film/film structure using magnetostrictive and aluminium nitride films on silicon substrate was realized, and showed ME amplitudes reaching 30 V Oe<sup>-1</sup> cm<sup>-1</sup>. Results agree with analytical theory.

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### 1. Introduction

The magnetoelectric (ME) effect, predicted by Curie in 1894 is the change of electric polarization as a response to an applied magnetic field [1,2]. With the development of new techniques, the interest for ME effect grew again (see Fiebig's reviews [3,4]): new "stress mediated" composite materials mainly associating magnetostrictive and piezoelectric or electrostrictive based materials were created and displayed ME effects several orders of magnitude higher than in natural ME materials. ME coefficient values as high as 10 V Oe<sup>-1</sup> cm<sup>-1</sup> could be obtained in Terfenol-D/PMN-PT laminate composites. Higher values can be found in the case of resonant devices [5-8]. Most of these composites are still bulk materials, and therefore, hard to integrate in micro-electromechanical-systems (MEMS) or microelectronic devices. Recent works on ME epitaxial nanostructures show very promising features [9]. In the present work, we investigated RF-sputtered thin film based solutions combined with the use of a magnetic instability which highly increases the sensitivity of the magnetomechanical interaction [10] and as a result enhancement of ME effect.

## 2. Spin reorientation transition in giant magnetostrictive nanostructured multilayers

The magnetic multilayer technology enables the preparation of nanostructures with tailored magneto-elastic properties. In particular, the (TbFe<sub>2</sub>/Fe) or (TbCo<sub>2</sub>/FeCo) nanostructured multilayers with giant magnetostriction and almost perfect uni-axial magnetic anisotropy were elaborated [11,12]. These structures are of interest because of brightly expressed spin reorientation transition (SRT) in a transversal bias magnetic field. In the vicinity of the SRT the magnetic subsystem becomes strongly nonlinear and anomalously sensitive to the external driving field [13–17]. Integration of these materials into MEMS has been shown [18,19]. For a given uni-axial magnetostrictive layer with a magnetic configuration such as the one shown in Fig. 1, the magnetic free energy of the system can simply be written as the sum of the Zeeman energy and the anisotropy energy restricted at its first term:

$$F = -M(H_S \cos(\varphi) + h \sin(\varphi) + \frac{1}{2}H_A \sin^2(\varphi))$$
(1)

where  $H_A$  is the in plane anisotropy field. This can then be used to understand the behaviour of the magnetization angle  $\varphi$ .

When the bias field  $H_S$  is perpendicular to the easy axis (EA) and strong enough (i.e.  $H_S > H_A$ ), magnetization is parallel to  $H_S$ and this orientation is stable. Application of an alternating field with an amplitude of a few Oersteds perpendicularly to  $H_S$  leads to a linear oscillation of the magnetization with a small angular amplitude. The magneto-elastic vibrations are linearly excited in such conditions. In the point of SRT ( $H = H_A$ ), the magnetic subsystem becomes unstable: thanks to the flat shape of the free energy, the amplitude of magnetic oscillations increases (Fig. 2) and the response to h is strongly nonlinear. When  $H_S < H_A$ , the equilibrium angle of the magnetization is rotated towards the anisotropy axis (angular phase) and linear oscillation around this position takes place when for low amplitudes of h. As expected for

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**Fig. 1.** Magnetic configuration of the uni-axial multilayer. Orientations of the bias  $H_S$  and alternating h magnetic fields and magnetization M relatively to the easy axis (EA) are shown.



**Fig. 2.** Free energy of the system as a function of the magnetization angle  $\varphi$ , calculated at the SRT point. Calculations were made here with a value of  $H_A = 244$  Oe and an alternative excitation amplitude of 2 Oe. The resulting magnetization angle amplitude is about 0.5 rad.

the SRT, this kind of behaviour is consistent with the Landau theory of second order phase transitions [20].

In the case of giant magnetostrictive films, the maximum deformation of the material is obtained when the magnetization rotates homogeneously with a  $90^{\circ}$  angle in the plane of the layer. Thus, the extreme sensitivity of the magnetic system at the SRT point is reflected on the magneto-elastic system and can be, in turn, exploited in stress mediated hybrid magnetostrictive/piezo-electric ME materials.

### 3. Theory of ME effect for vibrating substrate/film structures

With the configuration given in Fig. 3, we already showed [21] that for a given resonance mode number n, the amplitude of ME voltage generated by an excitation field h at a frequency  $\omega$  could be written as

$$V = \frac{2B(e_{31} - e_{33}C_{12}/C_{11})}{\epsilon\rho(\Omega_n^2 - \omega^2 + 2i\delta_n\omega)} \frac{d_m d_p}{4d} (2d_1 - d_m)(2d_1 - 2d_m - d_p) \times \frac{f_n\chi h}{S^2}$$

with the dimensionless function  $f_n = S\mu_n\gamma_n / \int U_n^2 dS$ .

*B* is the magnetomechanical coupling constant, *h* is the magnetic excitation field,  $\chi = \partial \varphi / \partial h$  is the sensitivity of the magnetization angle  $\varphi$  with respect to *h*, *A<sub>n</sub>* is the amplitude of vibration of the considered mode.



**Fig. 3.** Side schematic view of the considered magnetostrictive actuator with integrated piezoelectric material stacked on a non-piezoelectric (passive) substrate.

 $\mu_n$  and  $\gamma_n$ , respectively, are the magnetostrictive and piezoelectric form factors that can be expressed as

$$\mu_n = \iint \left( \frac{\partial^2 U_n}{\partial x^2} - \frac{\partial^2 U_n}{\partial y^2} \right) dS$$

for bending vibration modes,

$$\mu_n = \iint \left( 2 \frac{\partial^2 U_n}{\partial x \partial y} \right) dS$$

for torsion vibration modes,

$$y_n = \iint \left( \frac{\partial^2 U_n}{\partial x^2} + \frac{\partial^2 U_n}{\partial y^2} \right) dS$$

where  $U_n(x,y)$  represents the envelope of the elastic displacement along *z* axis for the considered vibration mode. The  $e_{ij}$  are the piezoelectric constants,  $C_{ij}$  are the elastic moduli,  $\delta_n$  is the damping coefficient,  $\Omega_n$  is the renormalized resonance frequency,  $\rho$  the substrate density and  $\varepsilon$  is the dielectric coefficient of the piezoelectric layer.

From this, it can be calculated that for the film/film ME layer on a thicker substrate, bending vibration modes can result in ME voltage whereas torsion vibration will result in no effect. It can be also deduced that if there is a thin magnetic film on a thick piezoelectric layer and no substrate, neither bending non torsion vibration will lead to generation of ME voltage.

In the case of 'in plane' modes, such as longitudinal vibration modes, similar calculations can be done leading to

$$V = \frac{d_m d_p}{d} \frac{B(e_{31} - e_{33}C_{12}/C_{11})^2}{\epsilon \rho} \frac{f_n}{(\Omega_n^2 - \omega^2 + 2i\delta_n \omega)} \chi h$$

with

$$f_n = \mu_n \gamma_n / S \cdot \int (U_x^{n2} + U_y^{n2}) \, dS$$

Magnetostrictive and piezoelectric form factors  $\mu_n$  and  $\gamma_n$ , respectively, are, for a longitudinal vibration, mode

$$\mu_n = \int dS \left( \frac{\partial U_x^n}{\partial x} - \frac{\partial U_y^n}{\partial y} \right) \text{ and } \gamma_n = \int dS \left( \frac{\partial U_x^n}{\partial x} + \frac{\partial U_y^n}{\partial y} \right)$$

This time  $U_x^n$  and  $U_y^n$  are the envelopes of elastic displacement along the *x* and *y* axes of the vibration mode *n*. Other parameters are the same as previously defined.

The expression of *V* in this case shows that for a cantilever type device (beam clamped at one end), the first longitudinal vibration mode will lead to generation of ME voltage.

### 4. Experiments and discussion

Three configurations have been realized and characterized.

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