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Nicolas Tiercelin
Vladimir Preobrazhensky
Olivier BouMatar
Abdelkrim Talbi
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Alexey Klimov
Théo Mathurin
Omar Elmazria
Michel Hehn
Philippe Pernod

Invited Paper

# From non-linear magnetoacoustics & spin reorientation transition to magnetoelectric micro/nano-systems

Nicolas Tiercelin[a], Vladimir Preobrazhensky[a,b], Olivier BouMatar[a], Abdelkrim Talbi[a], Stefano Giordano[a], Yannick Dusch[a], Alexey Klimov[c,d], Théo Mathurin[a], Omar Elmazria[e], Michel Hehn[e], and Philippe Pernod[a]

[a]Joint International Laboratory LIA LICS : Univ. Lille, CNRS, Centrale Lille, ISEN, Univ. Valenciennes, UMR 8520 - IEMN, 59651 Villeneuve d'Ascq, France
[b]Joint International Laboratory LIA LICS: Wave Research Center, A.M.Prokhorov GPI, RAS, ul. Vavilova 38, Moscow, 119991 Russia
[c]Joint International Laboratory LIA LICS: Moscow Technological University (MIREA) Vernadsky Avenue 78, Moscow, 119454 Russia
[d]Joint International Laboratory LIA LICS: V. A. Kotel'nikov Inst. of Radioeng. and Electronics (IRE RAS), ul. Mokhovaya 11/7, Moscow, 125009 Russia
[e]Institut Jean Lamour UMR 7198, Université de Lorraine - CNRS, 54506 Vandœuvre-lès-Nancy, France

## ABSTRACT

The interaction of a strongly nonlinear spin system with a crystalline lattice through magnetoelastic coupling results in significant modifications of the acoustic properties of magnetic materials, especially in the vicinity of magnetic instabilities associated with the spin-reorientation transition (SRT). The magnetoelastic coupling transfers the critical properties of the magnetic subsystem to the elastic one, which leads to a strong decrease of the sound velocity in the vicinity of the SRT, and allows a large control over acoustic nonlinearities. The general principles of the non-linear magneto-acoustics (NMA) will be introduced and illustrated in 'bulk' applications such as acoustic wave phase conjugation, multi-phonon coupling, explosive instability of magneto-elastic vibrations, etc. The concept of the SRT coupled to magnetoelastic interaction has been transferred into nanostructured magnetoelastic multilayers with uni-axial anisotropy. The high sensitivity and the non-linear properties have been demonstrated in cantilever type actuators, and phenomena such as magneto-mechanical RF demodulation have been observed. The combination of the magnetic layers with piezoelectric materials also led to stress-mediated magnetoelectric (ME) composites with high ME coefficients, thanks to the SRT. The magnetoacoustic effects of the SRT have also been studied for surface acoustic waves propagating in the magnetoelastic layers and found to be promising for highly sensitive magnetic field sensors working at room temperature. On the other hand, mechanical stress is a very efficient way to control the magnetic subsystem. The principle of a very energy efficient stress-mediated magnetoelectric writing and reading in a magnetic memory is described.

**Keywords:** NonLinear Magnetoacoustics, Spin Reorientation Transition, nanostructured magnetoelastic films, SAW, Magnetoelectric Devices

## 1. INTRODUCTION

The interaction of a strongly nonlinear spin system with a crystalline lattice results in a significant modification of the acoustic properties of those magnetic materials near the spin-reorientation transition (SRT).[1] The magnetoelastic coupling transfers the critical properties of the magnetic subsystem to the elastic one: the sound velocity decreases dramatically in the vicinity of the SRT[2] and the acoustic nonlinearity reaches giant and controllable values. Such a strong acoustic nonlinearity allows for the experimental observation in solids of a great

Further author information: (Send correspondence to N. Tiercelin.)
E-mail: nicolas.tiercelin@iemn.univ-lille1.fr, Telephone: +33 3 20 19 79 46

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number of nonlinear wave phenomena such as Brillouin scattering of sound by sound, acoustic bistability, collective oscillations of phonons, elasticity control by acoustic excitations, etc... The references on original papers can be found in.[1,3] A wide range of parametric effects is observable near the SRT due to controllability of the acoustic parameters by an external electromagnetic field. One of them is the supercritical parametric wave phase conjugation (WPC) of ultrasounds caused by the electromagnetic modulation of the sound velocity. The applications of the magnetoacoustic WPC in ultrasonic diagnostics and imaging are demonstrated in.[4–6]

It is also possible to induce a SRT in anisotropic magnetoelastic thin films and transfer some of the aforementioned concepts into microactuators and sensors. Thanks to the SRT, the magnetic system of the films is highly sensitive and non-linear.[7,8] Phenomena such as the magneto-mechanical demodulation of RF signals have been demonstrated.[9] The properties of the SRT in hybrid piezoelectric-magnetostrictive devices were also studied and an enhancement of the magnetoelectric effect was clearly evidenced.[10–12]

The alteration of the elastic system in magnetostrictive films can also be used to design new kind of microdevices using acoustic waves: for instance, in surface acoustic waves (SAW) based delay lines, the velocity of the propagating waves can be modulated by a magnetoelastic film placed between the interdigital transducers (IDT) submitted to an external magnetic field.[13] This effect can be used to devise efficient magnetic field micro-sensors.

The contol of the electronic, magnetic and dielectric properties of materials *via* elastic strains/stresses has recently formed a new direction of functional electronics called "straintronics".[14] Incidentally, the SRT in the magnetic subsystem can be induced by the application of an external stress that can be conveniently generated by a piezoelectric crystal mechanically coupled to the magnetic material. The magnetic state in such coupled systems is controlled by the electric field and conversely, the application of magnetic field induces a variation of the dielectric polarization. Such a kind of stress- or strain-mediated interaction is the basic principle of the multiferroics composites,[15,16] and the stress induced SRT was proposed as a concept for a magneto-electric random access memory (MELRAM) with ultra-low energy consumption.[17]

In the present paper we briefly discuss the mechanism of dramatic modification of the acoustic properties of magnetic materials near SRT and describe some phenomena in bulk magnetoelastic materials. Then we show how the concepts of SRT have also been transposed to magnetoelastic thin films with uni-axial anisotropy and applied to microactuators and sensors. In the last part we discuss an extension of this work on the extra-low energy control of magnetization in magnetoelectric memories.

## 2. MAGNETOELASTIC INTERACTION NEAR THE SPIN REORIENTATION INSTABILITY

The crucial factor in the formation of magnetoacoustic anomalies near the SRT is the instability of the magnetic system at the transition point. This kind of instability is inherent "by nature" in antiferromagnetic crystals exhibiting a magnetic anisotropy of the "easy plane" type (AFEP) and belonging to the $D_{3d}^6$ symmetry group such as $\alpha - Fe_2O_3$, $FeBO_3$, $MnCO_3$, etc. The magnetic configuration of the AFEP crystals in a magnetic field H that is applied in the basal plane is shown on fig.1.

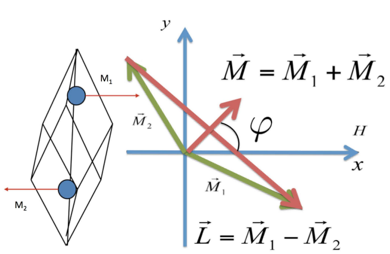

Figure 1. Magnetic configuration of the AFEP crystals.

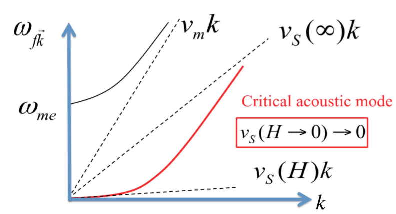

Figure 2. Spectra of magnetoelastic waves in AFEP crystals.

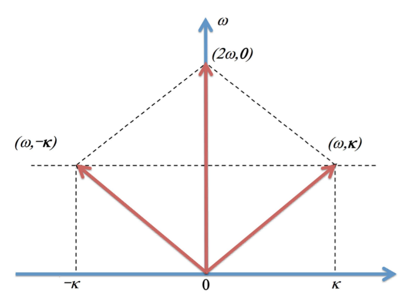

Figure 3. Diagram of acoustic parametric resonance.

The thermodynamic potential of the magnetic subsystem is the following function of the angular position $\varphi$ of the magnetization vector M relative to the applied magnetic field:

$$F_m = \frac{M_0}{2H_E}\left[\gamma^{-2}\nu_m^2\left(\nabla\varphi\right)^2 - \left(H\cdot\cos\varphi + H_D\right)\right], \tag{1}$$

where $M_0$ is the sublattice magnetization, $\nu_m$ is the spin wave velocity, $\gamma$ is the magneto-mechanical ratio, $H_E$ and $H_D$ are respectively the exchange and Dzyaloshinsky-Moriya effective fields. The homogeneous magnetization for $H = 0$ corresponds to the indifferent equilibrium with a potential energy that is independent of the in-plane orientations of magnetic moments. The magnetic subsystem becomes unstable relatively to the spin reorientation in the basal plane. This critical state is reflected in the spin dynamics by the rise of a soft mode with the $\omega_{fk} \Rightarrow \nu_m k$ spectrum.

The spontaneous magnetostriction introduces a magnetoelastic anisotropy in the spin dynamics. As a result, the spin wave spectrum gets actuation $\omega_{me} = \gamma\sqrt{2H_E H_{me}}$ with $H_{me}$ proportional to the spontaneous magnetostrictive deformation $\lambda_S$ of the crystalline lattice. The critical properties caused by the indifferent equilibrium in the spin subsystem are transferred to the acoustic excitations. One mode of the acoustic branch of the magnetoelastic spectrum becomes soft: long-wave sound velocity tends to zero. The spectrum of coupled magnetoelastic waves is shown schematically on fig.2 by dashed lines. For a linear magnetostriction, the magnetoelastic energy is a linear function of deformations $\widehat{u}$ and a nonlinear function of the deviation angle $\varphi$:

$$F_{me} = \left[\widehat{B}_1\cos 2\varphi + \widehat{B}_2\sin 2\varphi\right]\widehat{u}, \tag{2}$$

where $\widehat{B}_{1,2}$ are the magnetoelastic constants matrixes. For long-wave acoustic excitations, the angle $\varphi$ can be deduced from the quasi equilibrium equation $\frac{\partial(F_m+F_{me})}{\partial\varphi} = 0$. It is convenient to represent the nonlinear solution of this equation as a power series of deformations:

$$\varphi \approx -2\frac{H_E}{M_0}\frac{\widehat{B}_2\widehat{u}}{\left(\omega_{f0}/\gamma\right)^2} + 2\left(\frac{H_E}{M_0}\right)^2\frac{\left(2\widehat{B}_1\widehat{u}\right)\left(2\widehat{B}_2\widehat{u}\right)}{\left(\omega_{f0}/\gamma\right)^4} + \cdots \tag{3}$$

where $\omega_{f0} = \gamma\sqrt{H\left(H + H_D\right) + 2H_E H_{me}}$ is the antiferromagnetic resonance frequency. This substitution of the solution (3) of eqs. (1) and (2) allows for the presentation of the acoustic excitations energy in the conventional form with effective elastic moduli that are dependent on the magnetic field:

$$F_e^{eff} = \sum_n \frac{1}{n!}\widehat{C}_{(H)}^{(n)}\widehat{u}^n \tag{4}$$

where : $\widehat{C}_{(H)}^{(2)} = \widehat{C}^{(2)} - \frac{H_E}{M_0}\left(\frac{2\widehat{B}_2}{\omega_{f0}/\gamma}\right)^2$

$\widehat{C}_{(H)}^{(3)} = -6\left(\frac{H_E}{M_0}\right)^2\frac{\left(2\widehat{B}_1\right)\left(2\widehat{B}_2\right)^2}{\left(\omega_{f0}/\gamma\right)^2} \approx 10^{15} \div 10^{16} dN/cm^2$;

$\widehat{C}_{(H)}^{(4)} = 12\left(\frac{H_E}{M_0}\right)^3\frac{\left(2\widehat{B}_2\right)^4}{\left(\omega_{f0}/\gamma\right)^6}\left(1 + \frac{1}{4}\frac{\gamma^2 H H_D}{\omega_{f0}^2}\right) - 48\left(\frac{H_E}{M_0}\right)^3\frac{\left(2\widehat{B}_1\right)^2\left(2\widehat{B}_2\right)^2}{\left(\omega_{f0}/\gamma\right)^6} \approx 10^{20} \div 10^{21} dN/cm^2$

The second order moduli describe the linear properties of acoustic waves. In particular, for the velocity of the critical mode one can find: $\nu_S(H) = \nu_S(\infty)\sqrt{1 - \zeta_{me}^2(H)}$, where the magnetoelastic coupling coefficient is equal to:

$$\zeta_{me}(H) = \sqrt{\frac{2H_E H_{me}}{H(H + H_D) + \left(\omega_{me}/\gamma\right)^2}}. \tag{5}$$

Near the spin reorientation instability ($H \to 0$), the coupling coefficient tends to its fundamental limit $\zeta_{me} \to 1$ and the sound velocity decreases anomalously: $\nu_S(H \to 0) \to 0$.[2] The third and fourth order moduli respectively describe the quadratic and cubic acoustic nonlinearity. The important feature of the magnetoacoustic nonlinearity is the rapid increase of the nonlinear moduli with increase of their order: $\widehat{C}_{(H)}^{(n+1)}/\widehat{C}_{(H)}^{(n)} \approx \zeta_{me}^2(H)/\lambda_S$. For

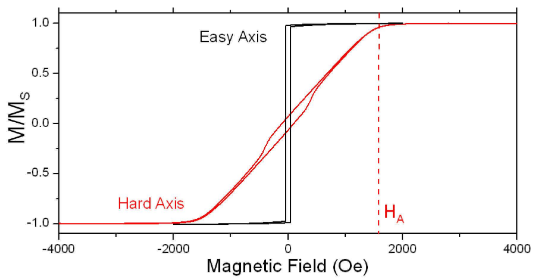


Figure 4. Normalized magnetization loops for a $25 \times (TbCo_2(4nm)/FeCo(4nm))$ nanostructured magnetoelastic layer deposited by RF-Sputtering under a magnetic field.

$\zeta_{me} \approx 1$, the nonlinearity becomes strong even for relatively small acoustic deformations that are comparable with the typical value of the spontaneous magnetostriction $\lambda_S \approx 10^{-5}$. These theoretical conclusions are confirmed by a great number of experimental results,[1] and are valid for all kinds of spin reorientation transitions developed as a second order phase transition.

## 3. PARAMETRIC MAGNETOELASTIC INTERACTIONS

The strong dependence of the elastic moduli on magnetic field offers an efficient mechanism for parametric interactions of acoustic waves with electromagnetic pumping. the application of a RF magnetic field $\overrightarrow{h_p}(t) = \overrightarrow{h_p} cos\omega_p t$ results in parametric interactions with energy:

$$F_{int} = \sum_n \frac{1}{n!} \frac{\partial \widehat{C}^{(n)}}{\partial \overrightarrow{H}} \widehat{u}^n \overrightarrow{h_p} \cos \omega_p t \tag{6}$$

The second order process (n=2) corresponds to an electromagnetic modulation of the sound velocity. In the resonance conditions $\omega_p = \omega_1 + \omega_2$, $\overrightarrow{k_1} + \overrightarrow{k_2} = 0$, where the indexes 1,2 denote the parameters of acoustic waves, the process is known as "wave phase conjugation" (WPC). The resonance diagram for the magnetoacoustic WPC is shown on fig.3.

Over the threshold of the parametric instability $h_{pc} = \left( \frac{1}{\nu_S} \frac{\partial \nu_S}{\partial H} kL \right)^{-1}$, where L is the length of the interaction zone under electromagnetic pumping, the WPC is accompanied by an exponential amplification of the phase conjugate waves. Various applications of the magnetoacoustic WPC with giant (more then 80 dB) amplification in ultrasonic imaging and diagnostics were demonstrated experimentally (see reviews[5,6] and references within). Moreover, the combination of the parametric interaction with quadratic nonlinearities allows for a cascade process in which, at the first step, an acoustic wave generates the second acoustic harmonics according to the resonance condition $2\omega_k = \omega_{2k}$ and then instantaneously, the second harmonic is phase conjugated by the electromagnetic pumping at the frequency $2\omega_p = \omega_{2k} + \omega_{-2k}$. This phenomenon was observed in $\alpha - Fe_2O_3$ single crystals.[18]

## 4. SPIN REORIENTATION TRANSITION IN MAGNETOELASTIC THIN FILMS

The SRT concepts exploited in bulk materials have also been transferred into thin film based applications. As shown on fig.4, magnetoelastic nanostructured layers that have been RF-Sputtered under a magnetic field exhibit a clear uni-axial anisotropy along the chosen bias field direction.

For such layers, and in the magnetic configuration described on fig.5a, the magnetic free energy density of the system can be written:

$$F_m = -\mu_0 M \left( H_S \cos(\varphi) + \frac{1}{2} H_A \sin^2(\varphi) + h \sin\varphi \right), \tag{7}$$

that is, in a power series at the 4th order:

$$F_m = -\mu_0 M H_S + \frac{1}{2}\mu_0 M \left(H_S - H_A\right)\varphi^2 + \frac{1}{8}\mu_0 M H_A \varphi^4 - \mu_0 M h \varphi, \tag{8}$$

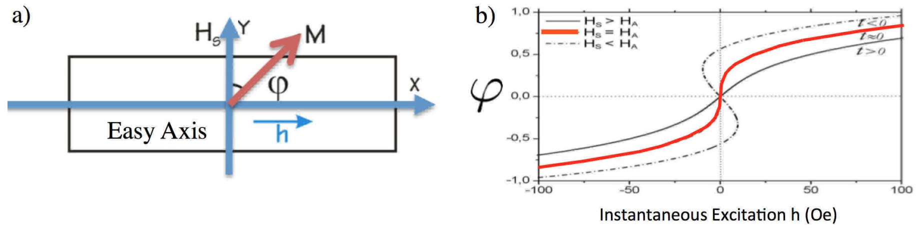


Figure 5. a) Magnetic configuration of a uni-axial magnetoelastic layer submitted to an in-plane magnetic polarization $H_S$ and excitation $h$. b) Example of a calculated magnetization response to the excitation $h$ for 3 cases $H_S > H_A$ (above the SRT), $H_S < H_A$ (bistable zone) and $H_S = H_A$ (SRT point).

which is characteristic of the free energy of systems exhibiting a second order phase transition as theorized by Landau:[19]

$$\Omega = \Omega_0 + \alpha t\eta^2 + b\eta^4 - \eta h_e, \tag{9}$$

where $\eta$ is the order parameter and $h_e$ the external field. Here, $\eta$ corresponds to the magnetization angle $\varphi$, $h_e = \mu_0 M h$, $\Omega_0 = -\mu_0 M H_S$, $\alpha = \frac{1}{2}\mu_0 M$, $b = \frac{1}{8}\mu_0 M H_A$ and $t = H_S - H_A$. As earlier, when $t \to 0$, the susceptibility of the order parameter with respect to the external field tends to infinity. Here, when the polarizing field $H_S$ is close to $H_A$, the magnetization susceptibility to the external field $h$ becomes very high and as shown on fig.5b, its response is also higly non-linear. Since the magnetoelastic effect is directly linked to the rotation of magnetization it is therefore possible to induce a high magnetomechanical susceptibility in micro-actuators as well as non-linear driving techniques.[7,8,20–22]

In particular, the strong non-linear response in the vicinity of the SRT allowed the demonstration of a magnetomechanical demodulation of signals : as shown on fig. 6, the vibration of a clamped beam type actuator was monitored with a laser/PSD technique while it was excited by an amplitude modulated RF signal using a microstrip waveguide. On the right hand side of the figure, a torsion vibration of the actuator was clearly induced with a maximum of amplitude near the SRT condition. An enhancement of these effects was also observed and studied when considering RF frequencies close to the ferromagnetic resonance (FMR) conditions of the magnetic layer.[9]

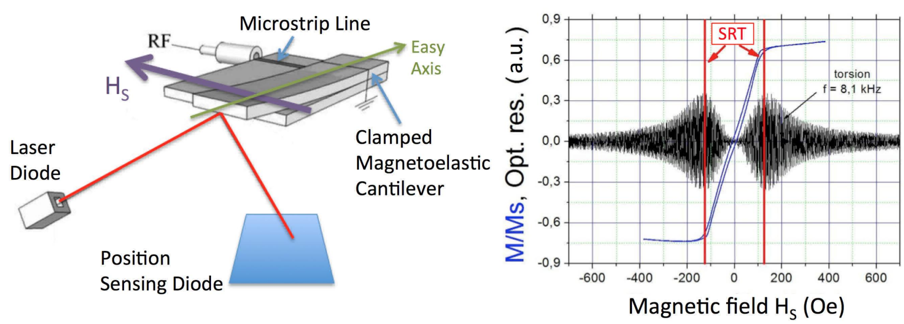


Figure 6. left: configuration for the excitation of a clamped magnetoelastic beam type actuator by a modulated RF signal. Right: Measured mechanical vibration for the first torsion mode of the beam at $f_T = 8.1$Khz, when excited with a 4GHz carrier with an amplitude modulation at $f_T$.

The properties of SRT have been later exploited in magnetoelectric (ME) hybrid magnetoelastic films/piezoelectric substrate[10] and film/film structures.[11,12] For the latter, a polished $50\mu m$ silicon substrate was coated by an electrode, aluminum nitride, and magnetostrictive nanostructured layer. A ME coefficient of $30\,\mathrm{V}\cdot\mathrm{Oe}^{-1}\cdot\mathrm{cm}^{-1}$ at a 35 KHz longitudinal resonance was measured. Nonlinear excitation of this mode showed a "nonlinear" dynamic ME coefficient of $4\,\mathrm{V}\cdot\mathrm{Oe}^{-1}\cdot\mathrm{cm}^{-1}$. The theoretical study also showed that since the symmetries of stress and deformation for piezoelectric and magnetoelastic materials are fundamentally different, one has to chose properly the geometries of the ME devices with respect to the considered mode of operation whether it be static or using a given resonance mode.

## 5. MAGNETIC FIELD SENSORS WITH SURFACE ACOUSTIC WAVES

As stated earlier, thanks to the magnetoelastic coupling, we can of course expect that the magnetization susceptibility and the elastic properties are linked, which means any change on the magnetic system will be reflected on the acoustic waves propagation. In particular, in the vicinity of the SRT, where the magnetic susceptibility is amplified, the acoustic waves velocity can be greatly affected. In order to take advantage of this effect, we realized High Frequency Surface Acoustic Wave (SAW) devices in which a part of the acoustic waves travels in a magnetostrictive film. A device example is shown on fig. 7: it is a SAW-based delay line composed of a pair of interdigital transducers (IDTs) on a piezoelectric lithium niobate (LNO) substrate. Using a lift-off process, a $N \times (TbCo_2/FeCo)$ nanostructured magnetoelastic layer is locally deposited by RF-Sputtering under a magnetic field in order to induce the uni-axial anisotropy.

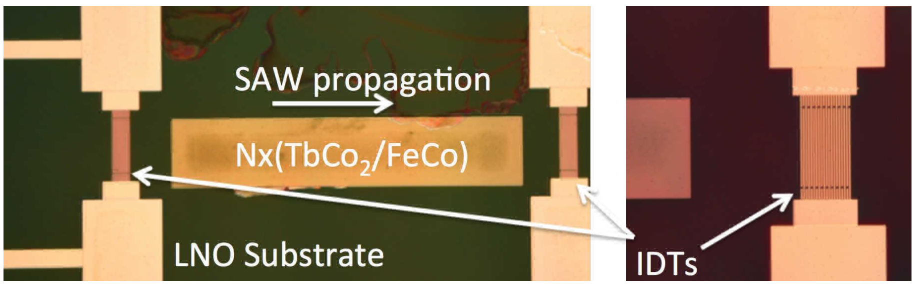


Figure 7. Magnetic field sensor based on a surface acoustic wave delay line on LNO substrate. A magnetoealastic multilayer is deposited in-between the IDTs.

One of the devices consisted in a 300 $\mu$m wide and 200 nm $20 \times (\mathrm{TbCo_2(5nm)/FeCo(5nm)})$ film with an easy axis along the propagation direction of SAWs generated by IDTs with 16 $\mu$m digits on a Y- cut $LiNbO_3$ substrate. The frequency response of the signal transmitted between the two IDT, *i.e.* the $S_{21}$ characteristic, was measured with a network analyzer (Agilent 8753). Rayleigh (R) and shear horizontal modes (SH) could be clearly identified at 232 and 271 MHz respectively. Upon application of a magnetic field perpendicularly to the easy-axis, a variation of the wave velocity was observed for both modes and reached 0.2% for the first SH mode.[13]

A theoretical description of the tunability of the SAW velocities was also developed based on the derivation of an equivalent piezomagnetic material of a magnetostrictive thin film, which allowed to compute mode shapes of elastic waves in layered piezoelectric-piezomagnetic composites deposited on a substrate.[23] In this model, the magnetization curves measured on real magnetic thin films can be introduced in calculations, and a very good agreement between the model and measurements of the SAW device was evidenced for both modes. It was found that only a small part of the acoustic wave is actually travelling in the magnetoelastic layer as most of if propagates in the LNO, thus impeding the effect. Simulations show that fully confining the wave in the magnetic layer could lead to huge variations of up to 20%.

Other configurations of SAW devices such as resonators allowing a remote/wireless interrogation are currently investigated[24] and are the subject of another communication in the same session of this conference proceedings.

## 6. MAGNETOELECTRIC CONTROL OF MAGNETIZATION IN STRESS-MEDIATED ARTIFICIAL MULTIFERROICS

Another aspect that is widely studied in magnetoelastic/piezoelectric coupled systems is the possibility for the control of magnetization with an electric field through stress. Since there are very few intrinsic multiferroic materials that could allow an efficient electric control of the magnetic properties, the so-called strain- or stress-mediated magnetoelectric coupling is of high interest, especially for magnetic memories applications.[15,25]

Based on our previous studies on SRT, we devised a new concept for a stress-mediated magneto-electric random access memory (MELRAM) with ultra-low energy consumption.[26] The magnetic configuration for this memory cell is given on fig. 8. It is based on a magnetoelastic element possessing a uni-axial magnetic anisotropy with an anisotropy field $H_A$, and submitted to a polarizing field $H$. A mechanical stress $\sigma_{ij}$ can be applied in

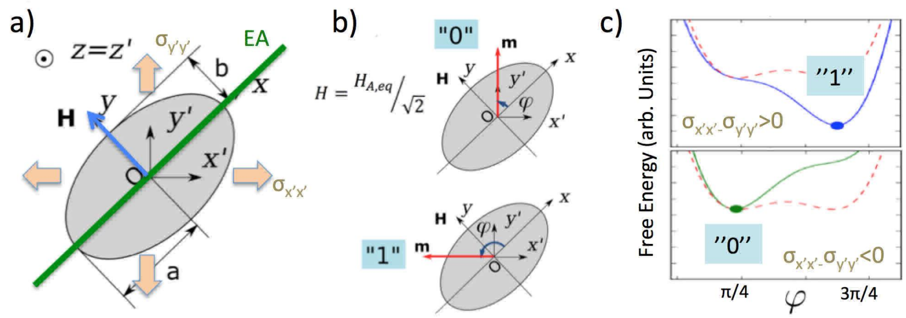


Figure 8. a) MELRAM configuration: magnetoelastic element with an easy axis and a polarising field. b) stable positions for magnetization without any applied stress. c) shape of the free energy for 'tensile' or 'compressive' stress.

the plane of the element. This time, the magnetic free energy of the system, given by eq. 10 is the sum of the Zeeman interaction, the anisotropy energy and the magnetoelastic energy.

$$F_m = -\mu_0 M_S H \sin(\varphi) - \frac{1}{2}\mu_0 M_S H_A \cos^2(\varphi) - \frac{3}{2}\lambda_S(\sigma_{x'x'} - \sigma_{y'y'})cos^2\left(\varphi + \frac{\pi}{4}\right) \tag{10}$$

It is noticeable here that the magnetoelastic energy has a similar dependence on the magnetization angle $\varphi$ to that of an anisotropy energy and therefore, because of the magnetoelastic coupling, any applied stress will affect the overall anisotropy of the magnetic layer. As shown on fig.8b, in the absence of a stress, two stable positions for the magnetization exist for $H < H_A$ (bistable mode as shown on fig. 5b), and they are aligned along the $x'$ and $y'$ axis for $H = \sqrt{H_A}/2$. The application of tensile or compressive stress along the $x$ axis modifies the shape of the free energy (fig.5b) and induces the switch of magnetic moments from one stable position to the other via the magnetoelastic interaction. The stress can be very efficiently generated with an electric field applied to a piezoelectric material elastically coupled with a magnetic one.[27] The magneto-electric switch between the magnetic states ensures a nonvolatile writing of information with a high energy efficiency. The dynamics of the commutation have been extensively studied using micro-magnetic simulations as well as with the Eshelby formalism.[28–30] They show that switching times below the nanosecond can be achieved in nanoscale devices with realistic stress pulses, and with an extremely low energy consumption of less than 2000 $k_BT$, *i.e.* about a hundred-fold less than state of the art in magnetic memories. Macroscopic prototypes using PZT actuators[31] or PMN-PT ferroelectric relaxors substrates[17] have been proposed and validated the switching concept.

As we saw, magnetic memory cells associated to the stress-mediated magnetoelectric effect promise extremely low bit-writing energies. Most investigations have focused on the process of writing information in the memory cell, and very few on the readout schemes. The usual assumption is that the readout will be achieved using magnetoresistive structures such as Giant Magneto-Resistive stacks[32] or Magnetic Tunnel Junctions.[33] Since the writing energy is very low in the ME systems, the readout energy using magnetoresistive approaches becomes non negligible. We recently demonstrated that incidentally, the magneto-electric interaction itself contains the potentiality of the readout of the information encoded in the magnetic subsystem. The principle of a magnetoelectric readout of the information by an electric field in a composite multiferroic heterostructure was considered theoretically and demonstrated experimentally using $[N \times (TbCo_2/FeCo)]$/PMN-PT stress-mediated ME heterostructures.[34] Since the magnetoelastic layer is mechanically coupled to the ferroelectric crystal, the dynamic reorientation of the magnetization induces variations of the electric polarization in the ferroelectric subsystem. Upon the application of a voltage pulse, *e.g.* positive, aiming at writing "1", the current response depends on the stored state: if the stored state was "0", then the magnetization rotates and a ME polarization is created, whereas if it was "1" already, there is no rotation and no extra polarization. The ME signal can be detected through the means of a Wheatstone bridge-like circuit and used to probe the stored state of the memory. Simulations show that for sub-micrometric devices, this reading operation will cost only a few attojoules, which is again much lower that the energy needed with a state of the art MTJ.

## SUMMARY

In this paper, we explained how the Spin Reorientation Transition can be induced and taken advantage of. In conjunction with magnetostrictive materials, the high sensitivity and highly non-linear response of the magnetic system can be reflected on the elastic properties thanks to the magnetoelastic coupling, and give rise to exciting properties leading to applications in acoustics, micro-sensors, micro-actuators and straintronics.

## ACKNOWLEDGMENTS

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