This document is the accepted manuscript version of the following article:

Finizio, S., Foerster, M., Vaz, C. A. F., Mix, C., Mawass, M. A., Tkach, A., ... Kronast, F. (2013). Electrical-field control of magnetism mediated by strain in Ni nanostructures fabricated on pre-poled PMN-PT (011). SPIN, 3(3), 1340008 (7 pp.).

<https://doi.org/10.1142/S2010324713400080>

Electrical-field control of magnetism mediated by strain in Ni nanostructures fabricated on pre-poled PMN-PT (011)

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We investigate the effects of piezoelectric-generated strain on the magnetization configuration of Ni nanostructures grown on pre-poled piezoelectric (011) $[\text{Pb}(\text{Mg}_{0.33}\text{Nb}_{0.66})\text{O}_3]_{0.68} - [\text{PbTiO}_3]_{0.32}$ (PMN-PT) by high resolution x-ray microscopy. We observe a strong uniaxial anisotropy in the Ni nanostructures, due to the relaxation of the substrate following the deposition of the Ni. The anisotropy can be modified by the application of an electric field to the piezoelectric substrate (thus generating a piezoelectric strain in the system) through the magneto-elastic effect. By applying an electric field to the PMN-PT, the magnetization configuration in nanostructured Ni squares and rings was reversibly manipulated with a high reproducibility.

Keywords: Magneto-elastic effect, Piezoelectric substrate, Domain structures, Magnetic nanostructures, Electrical field manipulation of magnetization

1. Introduction

While a key advantage of current magnetic storage technologies is that the magnetic system is designed to host a stable and predictable magnetic state, thus allowing for long-term data retention, this implies also that the manipulation of the magnetic state (i.e. the "writing" of a magnetic bit) conventionally requires the application of a large external magnetic field to achieve the switching of the magnetization (e.g. by the use of the Oersted field resulting from injected currents). Such energy intensive approaches lead to an unwanted energy consumption (and resulting heating) of devices.

Several approaches for reducing the energy consumption of such magnetic devices have become objects of interest in magnetism research. An example of such approaches is switching of the magnetization by the injection of a spin-polarized current^{1; 2; 3}. However, the large current densities needed for such spin torque switching lead to high power consumption, as only a small part of the energy is used for creating a spin torque that results in the actual switching of the magnetization, while most of the energy is expended in ohmic losses.

For this reason, voltage-controlled switching of the magnetization has recently been object of interest, as this approach does not require the use of large electrical currents that Oersted fields and spin-polarized current switching require. Examples of voltage controlled manipulation of the magnetization include using direct magneto-electric coupling by changing the charge carrier properties⁴, and the inverse magnetostriction effect^{5; 6}.

Here, we focus on the latter effect, which relies on the fact that the application of a strain influences, through the magneto-elastic interaction, the anisotropy in magnetic structures⁷. The application of strain generates an anisotropy term (magneto-elastic anisotropy), the size of which is governed by the magneto-strictive constant λ_s of the material and the applied strain^{7; 8}. This can then be exploited to overcome the other energy terms which contribute to the Landau free energy, such as the shape anisotropy due to dipolar coupling. This effect is of particular interest in the current studies of magnetic nanostructures, where shape anisotropy plays a key role in the determination of the magnetic configuration of the system^{9; 10}, and where the switchable magneto-elastic anisotropy can be utilized to control the magnetization of the system¹⁰.

By utilizing a piezoelectric substrate, it is possible to generate an electric-field induced strain through the piezoelectric effect. This effect, combined with the

magneto-elastic interaction, can thus be exploited to achieve voltage controlled switching of the magnetization, which is of particular interest for the development of magnetic devices with low energy consumption.

While there are different approaches for the generation of strain, piezoelectric substrates have been mostly utilized¹¹. These substrates can be used in their virgin unpoled or in a pre-poled state. The pre-poled state is often used, as it allows for a well-defined application of the strain, which is linear with the applied voltage for a sizeable voltage range. Furthermore, as magnetic nanostructures can be utilized as memory bits, it is of particular interest to analyze the effect of the magneto-elastic interaction on such systems deposited on pre-poled piezoelectric substrates.

2. Experimental

In this article, an experimental study of the effects on the magnetization of Ni nanostructures deposited on a relaxor piezoelectric substrate $[\text{Pb}(\text{Mg}_{0.33}\text{Nb}_{0.66})\text{O}_3]_{0.68} - [\text{PbTiO}_3]_{0.32}$ (PMN-PT) close to the morphotropic phase boundary, which allows for strong piezoelectric effects¹², is presented. To apply a piezoelectric strain to the Ni nanostructures, top and bottom electrodes have been deposited on the substrate, as shown in Fig. 1(a). The cutting direction of the substrate is (011), which allows for the generation of a piezoelectric strain in the in-plane direction (i.e. along the surface of the substrate) when applying an electric field along the out-of-plane direction. The piezoelectric strain presents a hysteretic behavior with respect to the applied electric field, as shown in Fig. 1(b) but, by applying selectively voltages of a certain range, a reversible, non-hysteretic, strain can be generated.

The Ni was deposited by electron-beam evaporation on top of a 50 nm thick sputter-deposited Pt electrode (with an adhesion layer of 5 nm of Ti between the Pt electrode and the Ni), deposited on a 0.5 mm thick pre-poled PMN-PT (011) substrate (provided by Atom Optics Co., Ltd., Shanghai, China). For details about the fabrication and deposition, refer to¹⁰. The substrate was pre-poled, i.e. an electric field was applied, which resulted in the crossing of the peak in the hysteresis loop shown in Fig. 1(b) (adapted with permission from¹⁶), prior to the deposition of the Ni.

Nanostructuring of the sample was accomplished by electron-beam lithography, with a lift-off technique, to obtain nanostructures of various geometries and dimensions (from 500 nm to 4 μm), as shown in Fig. 1(c), where a scanning electron microscopy (SEM) im-

age of the structures is presented. In the work presented here, the chosen geometries were square and ring structures, which have been suggested for memory device applications^{13; 14}, as well as for sensors¹⁵.

Figure 1. (a) Schematic of the experimental setup; (b) In-plane strain vs. out-of-plane applied electric field for a (011) PMN-PT substrate; (c) SEM micrograph of some of the analyzed ring structures.

The magnetization configuration of the Ni nanostructures has been imaged by photoemission electron microscopy (PEEM) with X-ray magnetic circular dichroism (XMCD) as the magnetic contrast mechanism¹⁷. The sample was illuminated with circularly polarized x-rays of 852.5 eV (Ni L_3 -edge, corresponding to the highest dichroic contrast). All the measurements presented here were carried out at room temperature.

Imaging was carried out at the UE49-PGM-SPEEM beamline at BESSY II and at the SIM (X11-MA) beamline at SLS, using an Elmitec PEEM setup (type LEEM III¹⁸).

The piezoelectric strain was applied to the sample by contacting the top and bottom electrode to a floating voltage generator placed inside the high voltage environment of the PEEM control unit (in order to allow for the extraction of the secondary electrons from the sample, a high voltage - typically 10 to 20 kV - has to be applied to the sample), as shown in Fig. 1(a). In order to allow for the measurement with an applied piezoelectric strain, the top electrode has been directly contacted to the sample high voltage, whilst the bottom electrode has been connected to the other output of the floating voltage generator. Electric fields up to 800 kV/m have been

applied across the substrate (corresponding to voltages up to 400 V). To avoid the cracking of the substrate, all voltages were applied slowly (all ramps were applied at a maximum rate of 100 V/s).

The magnetic state of the Ni nanostructures was initialized by the application of a magnetic field (with an electromagnet integrated within the PEEM sample cartridge, which can generate magnetic fields up to 50 mT, along the direction of the incoming x-rays - parallel to the contrast direction indicated in Figs. 2, 3, and 5). The samples were imaged prior, during and after the application of various piezoelectric strains. Due to the pre-poling of the substrate, part of the hysteresis cycle is probed during the measurements to ensure a monotonous (ideally linear) dependence of the strain on the applied voltage (refer to Fig. 1(b)). The usable electric field ranges from ca. -140 kV/m to +800 kV/m (the positive direction denotes a positive voltage applied to the bottom electrode, with respect of the top electrode).

3. Results

Using the above described setup, squares (widths between 500 nm and 2 μm) and rings (diameters varying from 1 μm to 4 μm, and widths varying from 200 nm to 350 nm) have been imaged. Using XMCD-PEEM imaging, square structures were first studied before applying any voltage. These structures are expected to exhibit the magnetic Landau state if shape anisotropy dominates¹⁹. Here, however, a bi-domain magnetic state was found, as can be seen in Fig. 2(a).

As previously stated, the piezoelectric material reacts to the application of an electric field with the generation of a piezoelectric strain. This strain is composed of a compressive and a tensile component, perpendicular to each other, as indicated in Fig. 2(b). For electric fields below a threshold value of about 340 kV/m, an increase of the area of domains with the magnetization parallel to the applied compressive strain direction is observed, as shown in Fig. 2(b). As the threshold value of about 340 kV/m is overcome, the magnetic configuration of the square changes from the Landau-like state shown in Fig. 2(b) to a more complex multi-domain state, with most of the magnetization parallel to the compressive strain direction, as shown in Fig. 2(c). Upon relaxation of the applied electric field back to 0 V/m, the magnetic configuration reversibly switches back to the original bi-domain state, as shown in Fig. 2(d) (cf. Fig. 2(a)). The magnetic state with an applied electric field above about 340 kV/m presents a characteristic vortex-like configuration (Fig. 2(c)), albeit the exact details of all the domains can vary slightly from cycle to cycle. The key

observation that the magnetization parallel to the compressive strain direction increases holds, however, for all cycles.

Figure 2. XMCD-PEEM images of a $2\ \mu\text{m}$ wide Ni square as function of applied piezoelectric strain. (a) Initialized condition, showing a bi-domain state; (b) Magnetization configuration with an applied electric field of $320\ \text{kV/m}$, showing the increase of domains with the magnetization parallel to the applied compressive strain direction; (c) Magnetization configuration with an applied electric field of $400\ \text{kV/m}$, showing that most of the magnetization in the magnetic structure with a vortex multi-domain pattern now points in the direction parallel to the compressive strain (up and down); (d) Magnetization configuration of the sample after relaxation of the electric field to $0\ \text{V/m}$, showing the reversibility of the changes shown in (b) and (c). The grayscale bar indicates the direction of the magnetic contrast, also indicated by the red arrows. The blue and yellow arrows in (b) denote the direction of the compressive and tensile strain components respectively.

XMCD-PEEM imaging of the ring structures has also been carried out, as shown in Fig. 3. Upon initialization with an external magnetic field, the rings are in the magnetic state shown in Fig. 3(a) and (d), with the magnetization pointing preferentially along the direction of the initial applied field.

Upon the application of a piezoelectric strain, an irreversible nucleation of domains pointing in the opposite direction with respect to the original magnetic configuration is observed, as shown in Fig. 3(b), (c), and (e). The nucleation of the domains occurs both during the application of the strain (Fig. 3(b) and (d)) and after the strain is relaxed to the zero field condition (Fig. 3(c)). The nucleation of the domains occurs along preferential sites, marked in Fig. 3(e), which lie at 45° with respect to the direction of the applied piezoelectric strain.

Figure 3. XMCD-PEEM images of Ni ring structures as function of applied piezoelectric strain. (a) Initialized condition (rings of $4\ \mu\text{m}$ of diameter and $350\ \text{nm}$ of width), showing a domain state with the magnetization pointing either parallel or anti-parallel to the incoming x-rays; (b) Magnetization configuration during the application of an $800\ \text{kV/m}$ electric field, showing the nucleation of inverted magnetic domains (indicated by the red arrows); (c) Magnetization configuration after the relaxation of the field to $0\ \text{V/m}$, showing the further nucleation of inverted magnetic domains (indicated by the red arrows); (d) Initialized condition (ring of $4\ \mu\text{m}$ of diameter and $350\ \text{nm}$ of width); (e) Magnetization configuration during the application of an $800\ \text{kV/m}$ electric field, showing the nucleation of inverted magnetic domains (indicated by the red circles). The grayscale bar indicates the direction of the magnetic contrast, also indicated by the red arrows in (d). The blue and yellow arrows denote the direction of the compressive and tensile strain components respectively. The arrows in (a) and (d) indicate the direction of the initialization field. The black marks in (e) indicate the position of the sites where the nucleation of inverted domains is observed.

An irreversible nucleation of domains pointing in the opposite direction with respect to the initialized magnetic state is observed. The original magnetic state can then be retrieved only by applying the initialization magnetic field again.

A statistical analysis of switching by domain nucleation was conducted, on sets of 4 rings each (thus 16 total nucleation sites were analyzed for each available

geometry). The occupation of the 4 sites marked in Fig. 3(e) was analyzed for each one of the available geometries, and the observed occupation of each site as a function of the applied piezoelectric strain (electric field) is given in Table 1 and Fig. 4. Rings with a larger diameter exhibit a higher probability of strain-induced nucleation of reverse domains, while, for a given diameter, the narrower rings show a higher probability of switching.

Table 1. Total occupation of the 4 nucleation sites shown in Fig. 3(e) as function of the ring geometry (outer diameter and width of the ring) and of the applied electric field (the determination was carried out after relaxation to zero field). 100% denotes the presence of an inverted domain in all 4 nucleation sites for all rings. The occupation has been calculated by counting the number of reversed domains observed in a series of rings of the same geometry divided by the total number of available nucleation states.

Diam [μm]	Width [nm]	Init	500 kV/m	800 kV/m
2	200	44%	66%	88%
2	300	22%	38%	81%
3	250	38%	81%	88%
3	300	44%	75%	81%
4	250	50%	88%	100%
4	350	56%	88%	88%

Figure 4. Total occupation of the 4 nucleation sites shown in Fig. 3(e), for the case of rings of 4 μm diameter, with widths of 250 and 350 nm, as function of differently applied electric fields to the piezoelectric substrates. In this case, imaging was carried out with the electric field applied to the piezoelectric. The occupation has been calculated with the same method used in Table 1.

For comparison, and to ascertain the influence of the pre-poling, preliminary analyses of Ni nanostructures deposited on un-poled PMN-PT substrates were also carried out. We only investigated the magnetization configuration at no externally applied electric field of a square Ni structure with the same geometry as the one

shown in Fig. 2. As presented in Fig. 5(b), it is possible to observe that the magnetic state of the square is in a quasi-Landau state for the un-poled substrate and in a bi-domain state for the pre-poled substrate.

Figure 5. XMCD-PEEM images of a 2 μm wide Ni square at no externally applied electric field. (a) Ni deposited on a pre-poled substrate, showing a bi-domain state; (b) Ni deposited on an un-poled substrate, showing a magnetic Landau state. The grayscale bar indicates the direction of the magnetic contrast, and the spin structure is also visualized by the red arrows.

4. Discussion

The analysis of the magnetization configuration of square structures (Fig. 2) reveals the presence of a uniaxial anisotropy induced by strain when an electric field is applied to the piezoelectric substrate. The magnetic configuration of the square does not exhibit the Landau state pattern, which is expected for a zero strain and shape-anisotropy dominated magnetization configuration. Instead, a bi-domain state with the magnetization pointing along the direction of the easy axis of a uniaxial anisotropy is observed. This bi-domain state is the typical signature of a strong uniaxial anisotropy¹⁹.

The origin of the observed anisotropy in the Ni nanostructures can be attributed to the use of a pre-poled substrate during the deposition of the material (as shown in Fig. 5). The substrate is a relaxor ferroelectric, which exhibits a relaxation of the strained state with time. The evaporation of the Ni leads to the growth of a fully relaxed film on a pre-strained PMN-PT substrate. The PMN-PT then relaxes with time to its original low strain condition, causing a strain in the deposited Ni film, thus generating through the magneto-elastic effect the observed uniaxial anisotropy that leads to the magnetic state shown in Fig. 5(a).

As observed in previous works⁸, the negative sign of the magnetostrictive constant of Ni ($\lambda_s \sim -33$ ppm for Ni⁸), implies that the magneto-elastic induced anisotropy generates an easy axis which points along the compressive direction of the applied strain. In the case described in this article, the piezoelectric strain is ap-

plied to the material in the same directions as the original pre-poling strain. The increasing strain in the substrate then reduces the strain of the Ni film until, at an electrical field strength between 320 kV/m and 340 kV/m, the original pre-strained condition of the substrate, at which the Ni deposition was conducted, is recovered. This then leads to the Ni exhibiting a vortex-type state, typical of shape anisotropy-dominated magnetic states of an unstrained Ni film structure. Upon further increasing the electric field, the additional uniaxial strain then favors the perpendicular magnetization direction. This leads to the state shown in Fig. 2(c). The square nanostructures could thus be reversibly switched between two different magnetic states (bi-domain and vortex states). However, only one of the states is stable for large and small applied electric fields, thus this is not useful to store information, since there is no bi-stability at zero applied electric field.

The study of the Ni ring structures has allowed us to observe (Fig. 4 and Table 1) that larger and narrower rings show a higher probability of nucleation of a reversed magnetic domain, suggesting the existence of a lower energy barrier for these structures. In this case, a bi-stability of the magnetic state at zero applied electric field has been observed.

The observed behavior is highly reproducible and the obtained magnetic states are stable in time (i.e. no changes in the attained magnetic states were observed after waiting times of several hours). In case of square structures, the original configuration is fully recovered upon the relaxation of the applied strain, whilst in the case of ring structures, the original configuration is recovered upon the re-application of the initialization field. This allowed for the reliable repetition of the presented experiments, and constitutes a positive point for device applications based on such system.

5. Conclusions

The influence of an applied electric field on the magnetization configuration of square and ring Ni nanostructures on pre-poled (011) PMN-PT substrates was analyzed.

The existence of a strong uniaxial anisotropy prevailing over shape anisotropy, and attributed to the relaxation of the PNM-PT after the deposition of the Ni, was observed.

The study of square nanostructures allowed us to observe the existence of a strong uniaxial anisotropy term, which, after comparison with nanostructures grown on un-poled PMN-PT substrates, can be attributed to the pre-existing strain conditions of the sub-

strate during the deposition of the magnetic material. The magnetic anisotropy can be influenced by the application of a piezoelectric strain, and the magnetization configuration could be switched between two different magnetic states. However, as there is no bi-stability at zero applied electric field (no applied piezoelectric strain), the system cannot be utilized for non-volatile applications such as data storage.

The analysis of ring nanostructures revealed the possibility to switch the magnetic state at precise nucleation sites. The reversed domains remained in that condition upon reducing the electric field to zero, showing that, here, a bi-stability can be achieved.

The observed phenomena show the possibility to achieve a current-less control of the magnetization by exploitation of the magneto-elastic interaction. However, the high uniaxial anisotropy present in the systems poses a challenge for further applications in devices.

Acknowledgments

Part of this work was performed at the Swiss Light Source, Paul Scherrer Institut, Villigen, Switzerland and at the Helmholtz Zentrum Berlin, Germany. The authors further acknowledge the financial support by the EU's 7th Framework Programme IFOX (NMP3-LA-2010 246102), the European Research Council through the Starting Independent Researcher Grant MASPIC (ERC-2007-StG 208162), the Swiss National Science Foundation, the Graduate School of Excellence "Materials Science in Mainz" (GSC 266), and the Deutsche Forschungsgemeinschaft (DFG).

Bibliography

1. V. Uhlir et al., *Physical Review B* **81**, 224418 (2010).
2. M. Kläui et al., *Physical Review Letters* **94**, 106601 (2005).
3. J.A. Katine et al., *Physical Review Letters* **84**, 3149 (2000).
4. C.A.F. Vaz et al., *Physical Review Letters* **104**, 127202 (2010).
5. F. Zavaliche et al., *Nano Letters* **7**, 1586 (2007).
6. J.-M. Hu, Z. Li, L.-Q. Chen and C.-W. Nan, *Nature Communications* **2**, 553 (2011).
7. E. Du Tremolet de Lacheisserie, *Magnetostriction: theory and applications of magnetoelasticity* (CRC Press, 1993).
8. M. Weiler et al., *New Journal of Physics* **11**, 013021 (2009).
9. A. Bur et al., *Journal of Applied Physics* **109**, 123903 (2011).
10. J.L. Hockel et al., *Applied Physics Letters* **100**, 022401 (2012).

11. C.A.F. Vaz, J. Hoffman, C.H. Ahn and R. Ramesh, *Advanced Materials* **22**, 2900 (2010).
12. K. Uchino, *Ferroelectrics* **151**, 321 (1994).
13. M. Kläui et al., *Journal of Physics: Condensed Matter* **15**, R985 (2003).
14. J.-G. Zhu, Y. Zheng and G.A. Prinz, *Journal of Applied Physics* **87**, 6668 (2000).
15. M.M. Miller et al., *Applied Physics Letters* **81**, 2211 (2002).
16. T. Wu et al., *Journal of Applied Physics* **109**, 124101 (2011).
17. J. Stöhr et al., *Science* **258**, 658 (1993).
18. Elmitec GmbH, <http://www.elmitec-gmbh.com/>.
19. A. Hubert and R. Schäfer, *Magnetic Domains: The Analysis of Magnetic Microstructures* (Springer-Verlag, Berlin, 1998).