Modification of magnetic anisotropy in Ni thin films by poling of (011) PMN-PT piezosubstrates

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ABSTRACT

This study reports the magnetic and magnetotransport properties of 20 nm thick polycrystalline Ni films deposited by magnetron sputtering on unpoled piezoelectric (011) $[PbMg_{1/3}Nb_{2/3}O_3]_{0.68}$-$[PbTiO_3]_{0.32}$ (PMN-PT) substrates. The magnetoresistance (MR), as well as the magnetization reversal, is found to depend on the polarization state of the piezosubstrate. Upon poling the PMN-PT substrate, which results in a transfer of strain to the Ni film, the MR value decreases by a factor of 12 at room temperature and a factor of 21 at 50 K for the current direction along the PMN-PT [100] direction, and slightly increases for the $[01\bar{1}]$ current direction. Simultaneously, a strong increase in the room temperature coercive field value is observed, while the ratio between the remnant and saturation magnetization shows a pronounced minimum for the [100] direction, indicating it as a hard axis, induced by poling the piezosubstrate.

Introduction

The conventional control of magnetization by a current-generated magnetic field is disadvantageous for electronic devices and sensors, due to difficulties in reducing power consumption and realizing device miniaturization. The non-volatile voltage control of the magnetization, resistivity or magnetoresistance (MR) in multiferroic heterostructures is one of the most promising schemes for achieving energy-efficient electronic applications [1]. Such artificial multiferroics consist of ferro(i)magnetic and ferroelectric layers, and the parameters of the former are controlled by a polarization of the latter, switched by an external electric field with maximum energy efficiency [2]. Recently, much attention has been paid to strain-mediated magnetoelectric coupling between room-temperature ferromagnetic Ni films and (011)-oriented $[PbMg_{1/3}Nb_{2/3}O_3]_{1-x}$-$[PbTiO_3]_x$ (PMN-PT) ferroelectric substrates with high piezoelectric coefficients [3–8]. In such a structure, upon application of the

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electric field, the piezoactive substrate induces a strain in the ferromagnetic film, and hence modifies its magnetic properties due to the magnetostriction effect. For the substrate, a (011) cut is chosen because of the possibility to get a high anisotropy by inducing simultaneously compressive and tensile strains in orthogonal [100] (x) and [01$\bar{1}$] (y) in-plane directions, due to the different signs of d_{31} and d_{32} piezocoefficients. However, while the electric field control of magnetization has been widely studied for Ni films on (011) PMN-PT [3–7], and the effect of the electric field on the MR of Ni films on (011) PMN-PT was recently reported [8], a report is still missing on the angular dependence of the poling modified magnetization, and on the temperature dependence of the poling modulated MR in Ni films on (011) PMN-PT.

This study reports these features of magnetotransport and magnetic properties of 20 nm thick polycrystalline Ni films deposited by magnetron sputtering on unpoled piezoelectric (011) PMN-32PT substrates. In particular, the effect of polling on the magnetoresistive response is determined for different crystallographic directions.

Experimental procedure

The samples were prepared on polished (011)-oriented $[PbMg_{1/3}Nb_{2/3}O_3]_{1-x}$-$[PbTiO_3]_x$ ($x = 0.32$) substrates (Atom Optics Co., LTD, Shanghai, China) of 5 × 5 × 0.5 mm^3 size, which were annealed at 300°C for 30 min. First, top (10 nm) and bottom (50 nm) Pt layers were DC-magnetron sputtered on both sides of the PMN-PT substrate using 5 nm thick adhesion layers of Cr. Then, polycrystalline Ni films with a thickness of 20 nm were deposited on the top side by DC-magnetron sputtering using an Ar pressure of 10^{-2} mbar.

Magnetoresistance measurements were performed in a van der Pauw configuration within a He cryostat (Oxford Instruments, MicrostatHe), using a system electrometer (Keithley 6514) as a current source and a 6.5-digit multimeter (Agilent 34411A) as a voltmeter with an error for our measurement range of ~0.02%. A voltage supply (FuG MCN 35-200) was used for application of the electric fields up to 4kV/cm between the top (Ni film) and bottom (Pt) electrodes of PMN-PT. The magnetic field up to 0.3 T was generated by an electromagnet (GMW 3470) powered by a bipolar power supply (Kepco BOP 36–6M). The same electromagnet and power supply together with a sample rotator were used for magneto-optic Kerr effect (MOKE) measurements. A nearly linearly polarized beam pointed to the sample was produced by a HeNe laser system (CVI Melles Griot) with a wavelength of $\lambda = 632.8$ nm and an output power of 5 mW, passing through a Glan–Thompson polarizer (Thorlabs) with an extinction coefficient of 10^{-5}. The beam reflected from the sample was periodically modulated at 50 kHz between left and right circularly polarized light by a photoelastic modulator (Hinds Instruments PEM-100), transmitted through an analyzer (polarizer with the transmission axis rotated by 45 degrees) and finally detected by a photosensitive fast responding diode (Hinds Instruments DET-200), connected to a lock-in amplifier (Signal Recovery 7225 DSP) with the modulation signal used as a reference input.

Results and discussion

Figure 1 shows the magnetization loops of Ni films on (011) PMN-PT, obtained at room temperature by the MOKE technique along different in-plane directions and normalized to the values obtained at maximum applied magnetic field in order to investigate the magnetic

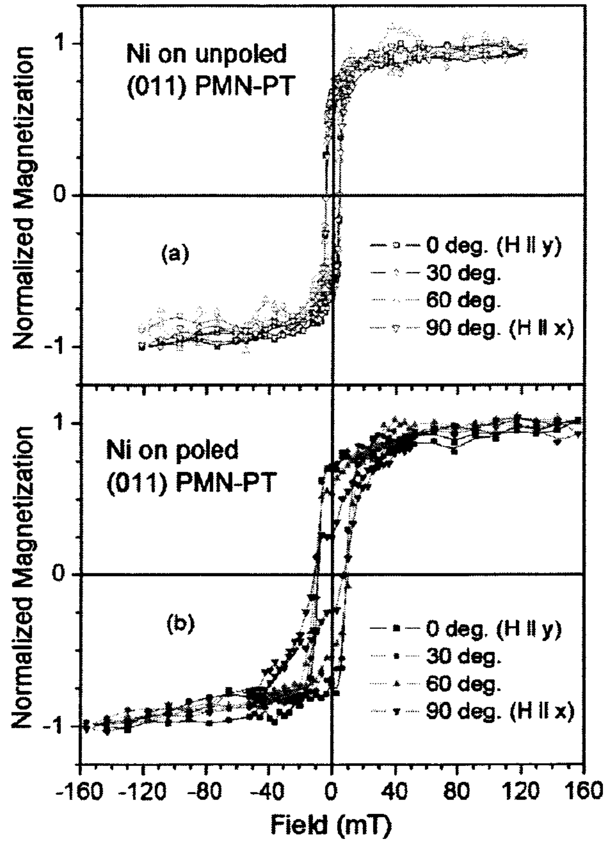

Figure 1. Room-temperature magnetization loops of the Ni films on (011) PMN-PT in unpoled (a) and poled (b) states, measured along different in-plane directions by the MOKE technique. Lines connecting points are a guide to the eye.

anisotropy. The loops obtained on these samples by the SQUID technique along $[01\bar{1}]$ (*y*) and [100] (*x*) directions were recently reported, revealing a saturation magnetization of 255 ± 15 kA/m [8]. Clear hysteresis is observed, indicating the ferromagnetic state of the films at room temperature. In the unpoled state (Fig. 1a), the magnetization loops for all the magnetic field directions are similar, suggesting that the films are mainly polycrystalline without strong magnetic anisotropy. However, after application of an electric field of −4 kV/cm (Fig. 1b), the loops become distinctly wider, and dependent on the crystallographic orientation of the (011) PMN-PT substrate. While in the *y* direction the loop is rectangle-shaped, it is slanted and slim in the *x* direction, indicating a hard axis behavior.

The magnetization loops measured in the intermediate directions demonstrate a continuous variation from the rectangle-like to the slanted shape at the sample rotation angles from 0° to 90° with respect to the *y* direction, as shown in Figure 1b. It is seen also that the coercive field H_c, where the magnetization crosses zero value, and the ratio between the remnant and saturation magnetization M_r/M_S, become dependent on the angle after poling. Analyzing the loops, an angular dependence of H_c and M_r/M_S from 0° to 180° is deduced and plotted in Figure 2. From the angular dependent magnetometry study of Ni films on (011) PMN-PT, we find that when PMN-PT is unpoled, H_c is 4.2 ± 0.4 mT for all the magnetic field

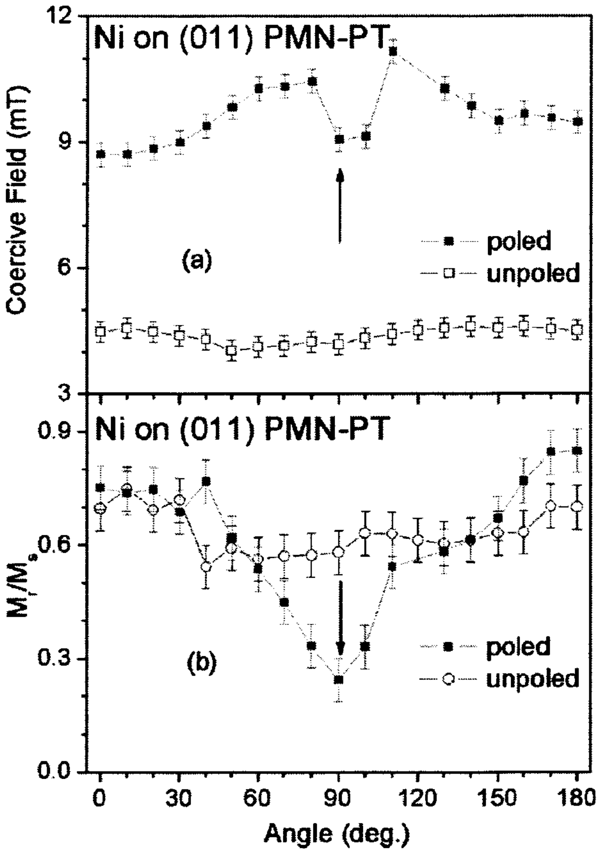

Figure 2. Angular dependence of the coercive field H_c and the ratio between the remnant and saturation magnetization M_r/M_s, deduced from MOKE magnetization loops of the Ni films on (011) PMN-PT, pointing to an induced hard axis. The line is a guide to the eye.

directions. However, when it is poled, the coercive field value increases more than twice, revealing also a dependence on the magnetic field direction. H_c smoothly increases when the film is rotated from the y and equivalent directions (0° and 180°) for an angle up to 80° but then there is a sharp decrease approaching 90°, corresponding to the x axis being a magnetic hard axis, as shown by an arrow in Figure 2a.

As shown in Figure 2b, the M_r/M_s ratios change significantly by poling as well. In the unpoled state, they vary around 0.65 ± 0.10. However, after poling, M_r/M_s ratios strongly change from about 0.9 around the y and equivalent directions (0° and 180°) to about only 0.25 around 90°, corresponding to the magnetically hard x axis. Such behavior of Mr/Ms is in accordance with the Stoner-Wohlfarth model [9]. Thus, the change of the coercive field and normalized magnetization when the sample is rotated in-plane shows that we could induce a strong uniaxial in-plane anisotropy by poling the PMN-PT, which induced different remnant strains in orthogonal [100] (x) and $[01\bar{1}]$ (y) in-plane directions.

We have also measured the MR of the Ni film on (011) PMN-PT, before and after poling the piezosubstrate by the application of −4 kV/cm, as shown in Figure 3a and 3b, respectively, for representative temperatures of 50 and 250 K. The measurements were performed in both y and x current directions, and both longitudinal and transverse geometries. Both

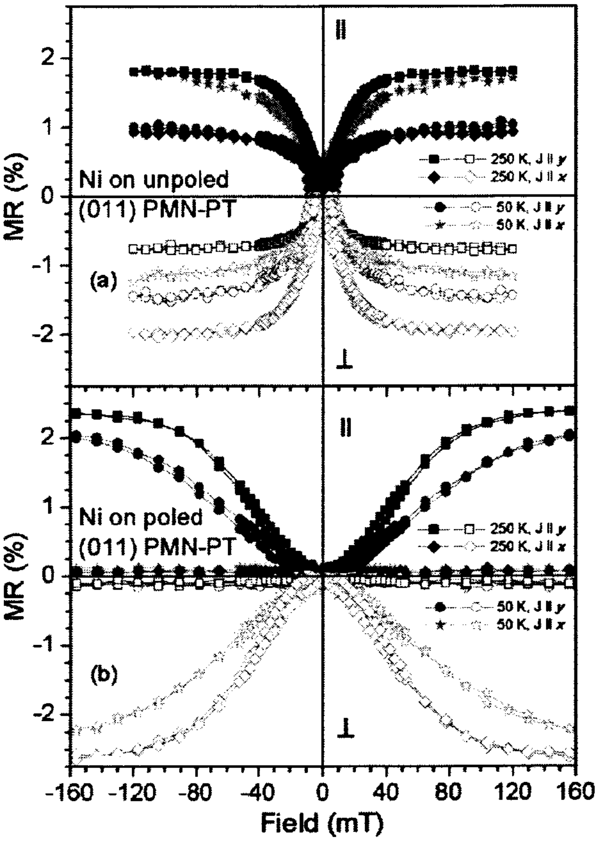

Figure 3. Magnetoresistance ratio ΔR/R0 in longitudinal (∥, solid symbols) and transverse (⊥, open symbols) geometries of the Ni film on (011) PMN-PT in unpoled state (a) and after poling with application of −4 kV/cm electric field (b) as a function of magnetic field, measured with the current along the *x* (diamonds and stars) and *y* (squares and circles) in-plane directions at 250 K (squares and diamonds) and 50 K (circles and stars). The lines connecting points are guides to the eye.

directions are found to exhibit a typical anisotropic magnetoresistance (AMR) curve. The resistivity of the Ni film in the longitudinal (transverse) direction shows minima (maxima) near the coercive field, and it increases (decreases) and saturates to a constant value MR_{max} above the saturation field H_S. However, the value of MR_{max} depends greatly on the magnetic field direction and temperature. In the unpoled state, shown in Figure 3a, the longitudinal MR at 250 K is higher for the *y* direction, while at 50 K it is higher for the *x* direction. In the poled state, presented in Figure 3b, the longitudinal MR is always higher along *y*, because when magnetic field is along the *x* direction, MR is suppressed by poling both near room temperature (by a factor of 12) and on cooling (by a factor of 21 at 50 K)."

Figure 4 summarizes the temperature dependence of the MR in the Ni film on (011) PMN-PT, showing the longitudinal and transverse MR_{max} values as a function of temperature between 5 and 295 K. In the unpoled state, the longitudinal MR decreases on cooling after a peak around 200 K for the *y* direction, while for the *x* direction the peak is around 50 K and the MR mainly increases on cooling. However, after poling the longitudinal MR along *x* diminishes without any significant variation on cooling. In contrast the longitudinal MR increases along *y*, revealing a peak of 2.6% around 150 K. The transverse MR as a

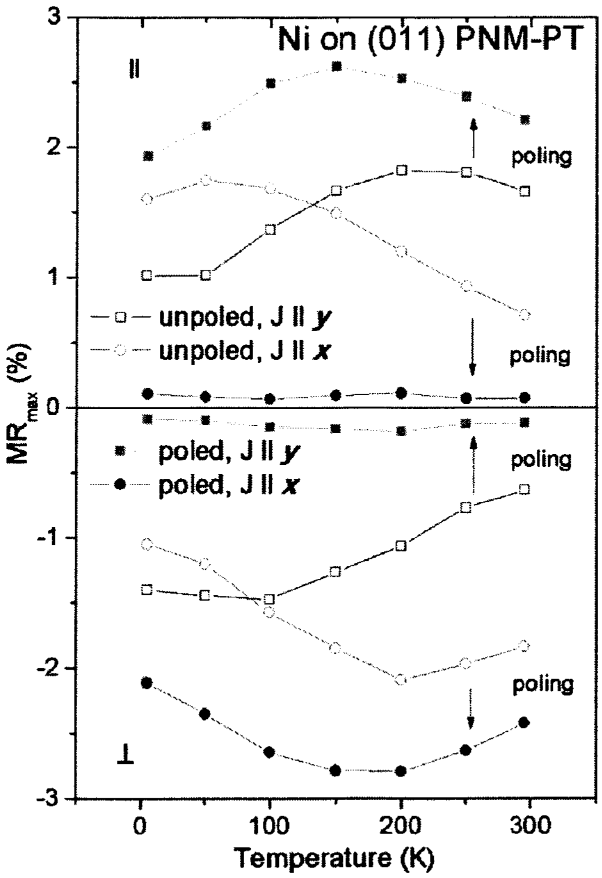

Figure 4. Saturation longitudinal (||) and transverse (⊥) magnetoresistance values MRmax of the Ni film on (011) PMN-PT in unpoled (open symbols) and poled (solid symbols) states with current along the *y* (squares) and *x* (circles) in-plane directions as a function of temperature. The lines connecting points are guides to the eye.

function of temperature nearly reflects that of the longitudinal MR in respect to the magnetic field direction.

A peak between 100 and 200 K in the temperature dependences of the longitudinal MR has previously been reported for Ni thin films and nanowires [10–12]. Maximum absolute values of the MR ratio between 1.8% [10] and 2.6% [11] were obtained, being comparable to those observed in this work, excluding the MR ratio along the *x* direction in the poled state. The low-temperature increase of the MR ratio on heating was explained by the contribution of electron-defect scattering to MR, before MR saturates at temperatures where electron-phonon scattering dominates [11]. The decrease of the MR ratio at higher temperatures was attributed to electron-magnon scattering [11]. Thus, the difference in the peak temperature for longitudinal MR along the *x* and *y* directions in the unpoled state should be related to the defect density variation along these directions. On the other hand, the variation of the peak MR ratio, together with the slight shift of the peak temperature after poling, indicates the change in the electron-phonon scattering that is due to the unit cell deformation by the residual strain.

Conclusions

In conclusion, we have demonstrated the very large effect of the poling of (011) PMN-PT piezosubstrates on the magnetic anisotropy and magnetotransport properties of Ni films, both at room temperature and at low temperatures. The coercive field is increased by the poling more than 2 times, while the M_r/M_s ratio becomes strongly dependent on the magnetic field direction, showing a deep minimum along x. At the same time, the longitudinal magnetoresistance is reduced by a factor of ~20 for the [100] (x) current direction, indicating an induced hard axis, and increased for the $[01\bar{1}]$ (y) current direction, indicating an easy axis. The modification of the MR response by the applied electric field was attributed to a change in the strain-induced anisotropy of the films, induced by the PMN-PT substrate with piezocoefficients of different signs in orthogonal in-plane x and y directions, and found to exist not only at room temperature but also on cooling down to 5 K.

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