Modulation of magnetic coercivity in Ni thin films by reversible control of strain

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ABSTRACT

In this study, we demonstrated the magnetoelectric control of magnetic thin films. (111)-textured Pd/Ni/Pd thin films were prepared on mica/lead zirconium titanate (PZT) substrates for the investigation. The reversible modulation of magnetic coercivity in Ni films was observed through the electric-voltage-controlled strain variation from the PZT substrate. For 14 nm Ni film, the applied electric field of ± 350 V/m led to ± 0.5% strain variation of PZT, which was transferred to ± 0.4% strain variation of Pd/Ni/Pd thin films on mica, and resulted in ± 17 Oe (± 5% of the preliminary magnetic coercivity). The reversible modulation of magnetic coercivity is supposed to be caused by the voltage-controlled strain through the magneto-elastic effect.

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1. Introduction

Due to the potential in technical applications, the electric field control of magnetic devices has long been demanded and its development is still challenging. Conventional magnetic field or spin-torque-triggered magnetization switching methods yield high power consumption and energy dissipation, while electric-field-controlled or assisted switching methods, which may be applicable in spintronic devices, provide the possibility of low-power-consumption [1–4]. Accordingly, many studies have been carried out at the aim of electric field control of magnetism in the last decade. Up to now, several kinds of device structure have been proposed and demonstrated, for example, the utilization of single magnetic layer on oxide [3–13], magnetic semiconductors [14–19], multiferroic materials [20], and other hybrid systems [4,20]. These reports proposed various physical mechanism for the electric-field-control of magnetism as follows: the modulation of carrier density, d-state electron occupation-induced change of interface magnetic anisotropy [21], and the coupling between ferroelectricity and ferromagnetism.

In magnetic semiconductors, the local electric field can substantially change the carrier density because of the limited number of conducting electrons; this severely modulates the magnetic properties [14–19]. However, the voltage control of magnetic semiconductors is not typically applicable in ambient conditions because of the low Curie temperature. Besides, numerous recent studies have focused on ferromagnetic transition metals combined with oxide thin films, such as MgO [3–11], AlO x [9] and ZnO [12,13], in which the interface charge accumulation may modulate the interface magnetic anisotropy and thus changes the magnetic behavior. However, this interface effect can be applied only to thin films of limited thickness in which the interface magnetic anisotropy dominates the magnetic behavior.

Multiferroics and magnetoelectrics also provide other possibilities for the electric-field-control of magnetism. For example, bismuth ferrite (BiFeO 3 ) reveals both ferroelectricity and antiferromagnetism in single material [20]. However, the application of its antiferromagnetism, instead of ferromagnetism, is still hard to accomplish. Furthermore, the coupling between ferromagnetic metal and ferroelectric oxide, named artificial multiferroics, has currently attracted much attention and actually provides more possibilities for functionality design [22–36]. The coupling between ferromagnetism and ferroelectricity is mediated by the magneto-elastic effect through the reversible electric-field-controlled strain variation [22–26]. The ferroelectric materials of BaTiO 3 (BTO) [32–35] and Pb(Zr,Ti 1−x )O 3 (PZT) [26–31] have been widely used in the previous studies. For example, Lahtinen et al. reported on the coupling between ferromagnetic and ferroelectric domains in CoFe/BTO heterostructures [33]. Li et al. investigated the Ni/PZT bi-layered films and the measured Kerr-signal to voltage loops show explicitly butterfly-shaped features for thick Ni layers, dominated by the strain mediated magneto-electric coupling [29]. Besides, Chung et al. also observed the reversible single-domain evolution from an initial single-domain state to a transitional S-shape domain state with an electric field [28].
Nevertheless, due to the significant difference between the surface properties of the ferroelectric ceramics and the metallic ferromagnetic films, the questions about how to get well-ordered crystalline structure in the magnetic thin films on ferroelectric ceramics, and how is the strain transferring through the interface are essential and worth studying. In this experiment, mica sheets were adopted for the deposition of (111)-texture Ni films. We combined the magnetic thin films Pd/Ni/Pd/mica with the PZT substrate. The electric field control of magnetic coercivity was demonstrated. Through the application of bias voltage to PZT, the strain transfer from PZT to the thin film was investigated, and accordingly the magnetic coercivity of Ni films was systematically modulated. The detailed mechanism of magneto-elastic anisotropy change through the strain variation is discussed in the text.

2. Experimental method

The 2.7 nm Pd/Ni/14 nm Pd trilayer films were grown on mica sheets (∼10 μm) by electron-beam-heated evaporation in a vacuum chamber with a base pressure of 3 × 10⁻⁹ Torr [37]. The thickness of Ni ranged over few tens nm, while the thicknesses of the top and the bottom Pd layer were always fixed. After being cleaved in air, mica substrates were put into a vacuum chamber and then annealed at 400 K for 12 h. The thin film deposition was performed while the substrate was at room temperature. The crystalline structure was characterized by X-ray diffraction (XRD). For the XRD measurements, X-ray from a Cu Kα anode was used to obtain the Bragg reflections from the samples. The film thickness and the Pd/Ni/Pd trilayer structure were calibrated by transmission electron microscopy (TEM). After deposition, the Pd/Ni/Pd/mica films were glued to PZT substrates using epoxy resin (PZT: APC International, Ltd.). For the investigation of strain variation of the samples, strain gauges (Omega Engineering, Inc. KFH-3-350-C1-11L1M2R) were attached to both the bottom-side of PZT and the top-side of the Pd/Ni/Pd films for comparison. The magneto-optical Kerr effect (MOKE) was measured using a 670 nm semiconductor laser with electromagnets of the maximum magnetic field of 4000 Oe in the in-plane direction.

3. Experimental results

Fig. 1 illustrates the sample structure prepared in this experiment. The Pd/Ni/Pd trilayers were sequentially deposited on a newly cleaved mica surface. The 14 nm Pd buffer layer was adopted for improving the crystalline orientation of Ni films. The 2.7 nm Pd capping layer was added for the protection of Ni films from contamination and oxidation. The thicknesses of the Pd capping layer and buffer layer were invariant in the series of samples, i.e. only the Ni thickness was changed. After thin film deposition, the Pd/Ni/Pd/mica sample was glued to a PZT plate of length = 10 mm, width = 5 mm, and thickness = 2 mm. The top and bottom surfaces of PZT plate were coated with Ag films as electrodes for applying voltage. Fig. 2(a) shows the TEM cross section image of the sample. The Pd/Ni/Pd trilayer structures were clear to observe. Besides, from the TEM image, we confirmed the flat mica surface, which ensures the subsequent uniform and flat thin film growth. The energy-dispersive X-ray spectroscopy (EDS) line profiles were plotted in Fig. 2(b), confirming the element composition of the sample.

3.1. Crystalline structure

Fig. 3 shows the X-ray diffraction patterns of the following three samples: a mica sheet, 2.7 nm Pd/14 nm Ni/14 nm Pd/mica/PZT,
and 2.7 nm Pd/42 nm Ni/14 nm Pd/mica/PZT. The comparison between these data helps us to identify the observed XRD peaks. The XRD pattern of a mica sheet reveals main peaks at $2\theta=26.9^\circ$, 36.0$^\circ$, and 45.4$^\circ$ [38]. These feature peaks are still observable after the deposition of Pd/Ni/Pd films. At $2\theta=38.2^\circ$, there is the PZT (111) peak, which is consistent with the literature [39]. Because the Pd/Ni/Pd/mica did not fully cover the PZT substrate and some PZT area was at the surface top, the PZT(111) peak was observed in our measurement. As indicated in Fig. 3, the peaks of Pd(111) and Ni(111) appear at $2\theta=40.1^\circ$ and 44.2$^\circ$, respectively, showing the preference for the (111)-texture for both Ni and Pd layers. Actually previous works have reported that Pd thin film preferred (111)-orientation when grown on mica [40,41]. The matching between Ni(111) and Pd(111) has also been suggested in Pd/Ni(111) and Ni/Pd(111) systems [42,43]. Combining the information from previous studies, the (111)-texture of Pd/Ni/Pd/mica observed in our measurement is reasonable. The above TEM and XRD patterns reveal that the mica substrate not only provides a flat surface for the thin film deposition, but also bring about the highly oriented crystalline structure of (111)-texture for the Pd/Ni/Pd films.

### 3.2. Magnetoelastic response

Fig. 4 shows the MOKE hysteresis loops of 2.7 nm Pd/14 nm Ni/14 nm Pd/mica/PZT, measured while applying a bias voltage to PZT (from 0 V to ±700 V). The shape of the MOKE hysteresis loops remains invariant, and the squareness of hysteresis loop is always nearly 100%. Only the magnetic coercivity is modified by the bias voltage. The magnetization reversion curves reveal systematic variation with the bias voltage. The shift of the reversion curves close to the coercivity field region is magnified, as shown in the insets. As shown in Fig. 4(a), the magnetic coercivity ($H_c$) monotonically increased with the increasing positive bias voltage. Inversely, as shown in Fig. 4(b), when applying a negative bias voltage, the $H_c$ decreased monotonically. Fig. 5(a) and (b) summarizes the voltage-induced $H_c$ variation of 22 nm and 14 nm Ni samples, respectively. The $H_c$ is plotted as a function of the applied electric field on PZT. Note that the thickness of PZT is 2 mm, and the effective electric field $E$ is deduced by $E$=voltage/ thickness of the PZT substrate. For 22 nm Ni, the $H_c$ is around 242 Oe while $E=0$ kV/m. When applying $E=+350$ kV/m and $-350$ kV/m, the $H_c$ changes to 250 Oe and 235 Oe, respectively. The variation of $H_c$ is ±3% of the preliminary magnetic coercivity. For 14 nm Ni, as shown in Fig. 5(b), the $H_c$ is around 350 Oe while $E=0$ kV/m. When applying $E=+350$ kV/m and $-350$ kV/m, the $H_c$ changes to nearly 370 Oe and 330 Oe, respectively. The variation of $H_c$ is ±5% of the preliminary magnetic coercivity. In comparison between Fig. 5(a) and (b), a thinner Ni film revealed a larger $\Delta H_c$, i.e. a larger magneto-elastic effect than that in a thicker film. This might be due to the fact that the thicker films accumulate more defects in the whole film, which might prevent the transfer of strain control, and thus lead to a minor magneto-elastic effect [44]. Besides, in the thin film region, the magneto-elastic coupling coefficient $B$ may not be a constant but a function of strain or thickness, i.e. $B=B$(strain) or $B$(thickness). These effects also should be considered.
In order to figure out the detailed physical mechanism, in Fig. 5(c) the strain variation of the PZT and Pd/Ni/Pd film was recorded as a function of the applied electric field $E$ for comparison. Note that the strain data is measured from the sample of 2.7 nm Pd/14 nm Ni/14 nm Pd/mica/PZT, the same as Fig. 5(b). For the PZT substrate, an electric field of $\pm 350$ kV/m induced the strain of $\pm 0.5\%$. Correspondingly, a smaller strain of $\pm 0.4\%$ was measured on the surface of Pd/Ni/Pd/mica. This measurement indicates that around 80% of the strain from the PZT substrate was transferred to the Pd/Ni/Pd thin film. The loss of strain in the mechanical transfer might be due to the layer-wise structure of mica, which may allow a gradient of strain in the micrometer-scale-thick mica sheet.

4. Discussion

The main objective of this study is to demonstrate the magnetoelectric control of magnetic thin films. We combined the magnetic Ni thin films and the ferroelectric PZT substrate in the samples. The deposition on mica helps us to achieve the (111)-texture in the Pd/Ni/Pd films. The magneto-elastic effect is expected to mediate the coupling between ferromagnetism and ferroelectricity, since the strain of Ni and PZT is correlated, as shown in Fig. 5(c). Applying the voltage (electric field) to PZT induced the size variation of PZT, and the strain of Ni films will be modified as well. The strain variation of Ni film triggers the modification of magneto-elastic anisotropy energy, and thus changes the magnetic coercivity $H_c$. In Fig. 5(c), the experimental data exhibits that the negative strain in the film plane direction caused the decrease of $H_c$. Inversely, a positive strain resulted in the increase of $H_c$. This observation can be explained by introducing the magneto-elastic anisotropy energy. Actually, we performed the strain measurement in different directions in the surface plane and confirmed that the in-plane strain is isotropic. In a cubic-(111)-orientated magnetic thin film, the magneto-elastic anisotropy $E_{me}$ can be expressed as follows [45]:

$$E_{me} = \Delta (E_1 - E_\perp) = B_2 (\varepsilon_1 - \varepsilon_\perp)$$  \hspace{1cm} (1)

where $E_{me}$ is the magneto-elastic anisotropy, i.e. the change of energy difference between in-plane magnetization ($E_1$) and perpendicular magnetization ($E_\perp$). $B_2$ denotes the magneto-elastic coupling coefficient. For face-centered-cubic (FCC) Ni, $B_2 = 10 \text{ (MJ/m}^3\text{)}$ [45]. $\varepsilon_1$ and $\varepsilon_\perp$ denote isotropic in-plane film strain and perpendicular film strain, respectively. With the given elastic constants $c_{ij}$ of FCC Ni from the literatures, the perpendicular strain can be deduced from the in-plane strain [45]:

$$\varepsilon_\perp = -2\varepsilon_1 \frac{C_{11} + 2C_{12} - 2C_{44}}{C_{11} + 2C_{12} + 4C_{44}} \approx \varepsilon_1 - 0.62\varepsilon_1$$  \hspace{1cm} (2)

After combining Eqs. (1) and (2), we obtain the following equation and thus the $E_{me}$ can be deduced from the measured in-plane film strain $\varepsilon_1$:

$$E_{me} \approx 16.2\varepsilon_1 \text{ (MJ/m}^3\text{)}$$  \hspace{1cm} (3)

According to Eq. (3), a positive (negative) in-plane strain $\varepsilon_1$ will lead to the positive (negative) $E_{me}$, indicating the tendency towards the perpendicular (in-plane) magnetization. This idea of strain-induced magneto-elastic anisotropy is schematically illustrated in Fig. 6. The total magnetic anisotropy energy (MAE) is expressed as a function of magnetization angle $\theta$; $\theta$ is the angle between the surface normal and the magnetization direction. For the ease of comparison, the MAE at $\theta = 0^\circ$ is aligned to the same value, i.e. MAE ($\theta = 0^\circ$) is taken as the reference energy. Since the easy axis of the Ni film lies in surface plane, the MAE reaches the minimum value at $\theta = \pm 90^\circ$. MAE at $\theta = 0^\circ$, i.e. the surface normal direction, is an energy barrier during the magnetization switching. Actually the MAE is composed of magnetic crystalline anisotropy, magnetic shape anisotropy, magnetic surface anisotropy, magneto-elastic anisotropy, etc. When the strain is modulated, the change of magneto-elastic anisotropy $E_{me}$ will thus affect the MAE. For example, while a positive strain is applied, a positive $E_{me}$ will be added to MAE. This positive $E_{me}$ ($\Delta (E_1 - E_\perp) > 0$) will reduce the MAE barrier, and thus a smaller magnetic field is required for magnetization reversal, i.e. $\Delta H_c < 0$. On the contrary, while a negative strain is applied, a negative $E_{me}$ will be added to MAE. This negative $E_{me}$ ($\Delta (E_1 - E_\perp) < 0$) will enhance the MAE barrier, and thus a larger magnetic field is required for magnetization reversal, i.e. $\Delta H_c > 0$. Accordingly, the correlation between the strain variation and the modulation of magnetic coercivity $\Delta H_c$, as shown in Fig. 5, is successfully explained by the magneto-elastic effect. Furthermore, based on the picture illustrated in Fig. 6, the strain modulation may even induce the spin reorientation transition from in-plane to perpendicular direction, when the MAE is small and the strain is large enough. If the magneto-elastic anisotropy dominates the MAE, for example in the ultrathin Ni film, then the electric-field-controlled strain can be an effective means to modulate the magnetic behavior, i.e. the positive (negative) strain will orient the magnetic easy axis to the perpendicular (in-plane) direction. In the previous studies, the magnetic easy axis rotated in the film plane by applying a unidirectional strain to the sample [24–26]. In contrast, the Ni films prepared in our current experiment reveal isotropic magnetic hysteresis loops in the film plane. This is due to the (111)-texture of crystalline structure and the weak anisotropy in the surface plane. The $H_c$ variation was driven by the isotropic strain $\varepsilon_1$ transferred from PZT. Besides, one should note that bulk magnetoelasticity does often not apply to nm thin films. Both thickness and strain may induce a deviation of the respective $B$ from its bulk value. Thus the proposed magneto-elastic coupling shown in Fig. 6 can only qualitatively explain our
experimental observation. As shown in Fig. 7, which summarizes the shift of the $H_c$ for 14 nm Ni film plotted as a function of the in-plane strain, a positive in-plane strain decreases the $H_c$ and a negative in-plane strain increases the $H_c$. The linear fitting indicates a slope of $-38 \pm 4$ Oe/strain (%). Further quantitative explanation about the correlation between $H_c$ and strain needs consideration of the thickness dependence of magnetoelasticity, magnetic domain nucleation and magnetic domain wall motion in the thin film.

In the previous study of Wang et al. on Co/MgO/Co magnetic tunnel junctions (MTJ), the electric-field-controlled $H_c$ variation was reported [3]. Because of the different polarity of interface charge accumulation, the top and bottom magnetic Co layers in-creased and decreased, respectively, while a bias voltage was applied to the junction. This observation has been proposed for fu-ture application of electric-field-controlled magnetic memory storage [3]. According to our successful demonstration in this re-port, magneto-elastic effect is another suitable choice for the electric-field-controlled magnetic device. In Eqs. (1)-(3), the re-lation between the magneto-elastic anisotropy $E_{me}$ and the strain $\epsilon$ is dominated by the magneto-elastic constants $B_1$ and $B_2$, espe-cially their polarity. For example, $B_1$ and $B_2$ are positive for FCC Ni, but negative for hexagonal-close-packed (HCP) Co. It implies that if the electric-field-induced positive strain occurred in a MTJ made of HCP-Co/oxide/FCC-Ni, then the $H_c$ of Co and Ni may change in the opposite ways, and vice versa. These demonstrated or pro-posed ideas for the manipulation of magneto-elastic anisotropy will be valuable in the future design of spintronic devices.

5. Summary

In this experiment, Pd/Ni/Pd trilayer thin films were deposited on cleaved mica sheets. By attaching to a PZT substrate, the strain variation of Pd/Ni/Pd thin films on mica, and resulted in $\pm 17$ Oe (±5% of the preliminary magnetic coercivity). The correlation between the electric-field-controlled strain variation and the corre-sponding change of $H_c$ can be qualitatively explained by the modulated magneto-elastic anisotropy. Due to the positive mag-neto-elastic constant $B_2$ of Ni, positive strain in Ni film reduced the energy difference between the in-plane magnetization and per-pendicular magnetization, i.e. the potential barrier for magnetism reversion was reduced. Accordingly the $H_c$ was decreased with a positive strain and vice versa. To extend the demonstration of the present experiment, the strain-mediated electric-field-control of MTJ with magnetic layers of opposite sign of magneto-elastic constants is proposed for future studies and applications.

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