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Magneto-optical Kerr effect investigation on magnetoelectric coupling in ferromagnetic/antiferroelectric multilayer thin film structures

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Magneto-optical Kerr effect investigation on magnetoelastic coupling in ferromagnetic/antiferroelectric multilayer thin film structures

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Magnetoelastic (ME) membranes comprising soft ferromagnetic Ni and antiferroelectric (AFE) (Pb,La)(Zr,Sn,Ti)O3 (PLZST) layers were proposed and fabricated through a bulk micromachining process on silicon wafers. An AC-mode magneto-optical Kerr effect technique was proposed to examine the magnetoelastic coupling in the multilayer membranes, in which the electric field-induced magnetization rotation was analyzed for understanding the underlying coupling mechanisms. The AFE to ferroelectric phase transformation of PLZST induced a rotation of magnetization of about 0.5° in Ni, persuaded by strain-induced anisotropy of about −0.5 kJ/m³.

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Magnetoelastic (ME) thin films with coupled electric and magnetic order parameters (magnetoelastic coupling) have attracted tremendous interest with the potential in realizing new multifunctional devices.1–6 In contrast to single-phase magnetoelectrics, which have weak magnetoelastic coupling and very low Curie or Neel temperatures,7 multilayer thin film structures consisting of ferroelectric (FE) and ferromagnetic components could exhibit magnetoelastic coupling at room temperature with the promise for technological applications.

Magnetoelastic coupling in multilayer thin films is thought to be mediated by strain transfer between the ferroelectric and ferromagnetic films8 or by accumulation of spin-polarized charges at the interface of the ferroelectric and an ultrathin ferromagnetic layer.9,10 When the ferromagnetic film is thicker than 10–15 nm, the later mechanism is thought to become ineffective and the former mechanism becomes the main factor.

Compared to traditional FEs, such as Pb(Zr,Ti)O3 (PZT) and Pb(Mg1/3,Nb2/3)O3-PbTiO3 (PMN-PT) which have strong piezoelectric responses, some antiferroelectric (AFE) thin film materials could exhibit even larger strains (of the order of 0.5%) during field-induced AFE-FE phase transformation.11 Since the phase transformation occurs very fast (in about 90 ns),12 the strain transfer could be more efficient than the conventional piezoelectric strain. Thus, we are considering that the replacement of the FE component by an AFE material may enhance the magnetoelastic coupling.

In this study, such an AFE material is proposed as an alternative to the FE component in a multilayer thin film structure comprising AFE (Pb0.97,La0.02)(Zr0.90,Sn0.05,Ti0.05)O3 (PLZST) and ferromagnetic Ni thin films. The magnetoelastic coupling of the multilayer structure is studied using our established AC-mode magneto-optical Kerr effect (MOKE) method.

510 nm-thick PLZST thin film was deposited on commercially available Pt/Ti/SiO2/Si substrate using a chemical solution deposition method described in Ref. 13. A 50 nm-thick Ni thin film was then deposited on the PLZST using a Denton DC sputtering system (sputtering power 100 W, base pressure 10⁻⁷ Torr, and Ar pressure 10⁻⁵ Torr) and patterned by a lift-off process. The Ni layer was used as a top electrode as well, besides the ferromagnetic function. The crystalline structure and morphology of multilayer film are presented and discussed in supplementary material.14

In the common multiferroic multilayer thin films, the massive substrate could substantially suppress the elastic coupling hindering any magnetoelastic coupling.15–17 To overcome this, researchers used vertical, composite nanostructures where the elastic strain is normal to the substrate surface,18–21 or used ferroelectric substrates that played an active role in the system.22,23 The former suffers from high leakage current (since the magnetic component is usually conducting) while the latter requires costly and very special substrates, which are not suitable for device integration and applications.

To reduce the substrate clamping effect, we removed the Si substrate under the active layers by micro-machining to fabricate free standing structures (see supplementary material).14 Although, cantilevers are more preferred to reduce the substrate clamping effect, their voltage-induced deflection was too large for convenient measurements by the proposed magneto-optical testing method. Hence, membrane structure was chosen as they are more mechanically stable and permit precise control of the measurements. To illustrate the substrate clamping effect by comparison, membranes without Si and with a residual Si layer of 15 μm in thickness were fabricated.

Room temperature out-of-plane polarization-electric field (P-E) characteristics of the membrane was determined by a standard RT66A (Radiant Technologies, USA) testing unit. In-plane and out-of-plane magnetic field dependant magnetization (M-H) loops were measured using a MicroMag™, which was designed to avoid the substrate clamping effect. The membrane was placed on the sample stage of the MicroMag™ and a lock-in technique was used for detecting the maximum field-induced magnetization rotation.

PVDF sensors were used to measure the total charge generated due to piezoelectric and magnetoelastic strain contribution. The output signal was amplified using a charge amplifier and then digitized for data acquisition. The MOKE signal were acquired using a commercial system equipped with a He-Ne laser (λ = 632.8 nm) and a photodetector. The signal was amplified using a lock-in amplifier and then digitized for data acquisition.

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2900 alternating gradient magnetometer (AGM). A laser scanning vibrometer (OFV-056, PolyTech GmbH, Germany) was used to measure electric field induced displacement of the membrane.

The $P-E$ hysteresis loop (Fig. 1(a)) shows the characteristic double loops of AFEs, with a maximum polarization of $33.7 \mu C/cm^2$ and a small remnant polarization of $2.1 \mu C/cm^2$. The forward (AFE $\rightarrow$ FE) and the backward (FE $\rightarrow$ AFE) switching fields were $162 \text{ kV/cm}$ and $86 \text{ kV/cm}$, respectively. The $M-H$ curve (Fig. 1(b)) shows the soft magnetic nature of the Ni film with a saturation magnetization and a coercive field of about $5.3 \times 10^5 \text{ A/m}$ and $9 \text{ mT}$, respectively. It also demonstrates that the in-plane direction is the easy magnetization direction for the Ni film.

The electric-field-induced displacement of the membranes was measured under a sine-wave voltage at a frequency of $250 \text{ Hz}$ and is shown in Fig. 1(c). At low electric fields, below the AFE to FE switching field, the AFE phase was stable and thus the membrane displacement was very small. With an increasing electric field, the AFE to FE transformation was initiated and accompanied by a large strain in the PLZST film. This event is reflected in the figure as a significant increase in displacement of the membrane, from about $250 \text{ kV/cm}$. With further increase in electric field, the displacement saturated (at about $79.8 \text{ nm}$ for free standing membranes and at about $50.2 \text{ nm}$ for membrane clamped by $15 \mu m$-Si) when the transformation was completed.

The converse magnetoelectric coupling in multilayer thin films is usually detected via a change in the $P-E$ sensitivity. With further increase in electric field, the AFE to FE transition was measured using conventional magnetometers or a MOKE system. Among them, the MOKE system is preferred as it is fast, non-destructive and has a higher sensitivity.

To prove magnetoelectric coupling using the conventional methods, an optical study based on the MOKE was first undertaken. The membrane was placed perpendicular to the magnetic field and a plane polarized light beam was incident at a small angle (\(\angle < 10^\circ\)) to the film plane normal, in the “polar MOKE geometry” (Fig. 2(a)). The polarization plane of the reflected light will get rotated by the magnetization of Ni film. This rotation, $\delta$, is proportional to the normal component of the magnetization vector of Ni, $M \cdot \cos \gamma$, where $M$ and $\gamma$ denote the magnetization vector and the angle between this vector and the normal to the film (Fig. 2(b)). The polarization rotation was measured using a polarization sensitive detector. To validate the proposed MOKE setup, out-of-plane displacement loops were obtained and compared to that measured by AGM. The loop obtained by MOKE was in good agreement with that obtained by AGM, as shown in Fig. 1(b), suggesting that the proposed MOKE setup is reliable.

Comparison of the in-plane and out-of-plane $M-H$ curves of Ni (Fig. 1(b)) revealed that, the magnetization vector of Ni was in the plane of the film at zero magnetic field. When the applied magnetic field was increased normal to the surface, the magnetization vector rotated towards the normal, so that at every value of the field $H$, its direction $\gamma$ corresponds to the minimum of the energy density, $E$,

$$E = \left( K + \frac{\mu_0 \cdot M_s^2}{2} \right) \cdot \cos^2 \gamma - \mu_0 \cdot H \cdot M_s \cdot \cos \gamma, \quad (1)$$

where the quantity $\mu_0 \cdot M_s^2/2$ is shape anisotropy energy ($\mu_0$ and $M_s$ are the permeability of free space and saturation magnetization, respectively), $K$ is the anisotropy induced by strain and the term $-\mu_0 \cdot H \cdot M_s \cdot \cos \gamma$ is Zeeman energy. The magnetization of Ni lies in a direction to minimize the total energy $E$. By equating derivative of Eq. (1) over $\gamma$ to zero to find the minimum value of $E$, one obtains

$$\cos \gamma = \frac{\mu_0 \cdot H \cdot M_s}{\mu_0 \cdot M_s^2 + 2K}, \quad (2)$$

where $\cos \gamma$ is proportional to the MOKE signal $\delta$. Equation (2) is valid while $H < H_{SAT}$; at higher $H$, the magnetization is

FIG. 2. (a) Setup for magneto-optical Kerr effect in polar geometry. (b) Notation of angles of magnetization vector of Ni. Strain induced by AC electric field causes the magnetization vector of Ni to rotate by the angles $+\delta_1$ and $-\delta_2$; $N$ and $M$ denote the normal to the film plane and magnetization vector of Ni.
is parallel to the field, hence $\gamma = 0$, and the MOKE signal reaches saturation. From the saturation magnetization obtained by AGM in Fig. 1(b), $M_S = 5.3 \times 10^5$ A/m, the corresponding shape anisotropy energy can be calculated as $\mu_0 M_S^2/2 = 176.4$ kJ/m$^3$. From the dependence of the $M$-$H$ hysteresis loop on the applied DC electric field, it is possible to determine $K$, the anisotropy induced by strain.

However, we could not find a significant change in the $M$-$H$ curve of Ni under an applied DC bias to PLZST, suggesting that the value of $K$ was rather small. This might be due to the reason that only a small portion of strain is coupled to Ni under DC condition. In order to estimate strain induced anisotropy, we developed an AC-mode MOKE method which provides high sensitivity to study the magnetoelectric coupling of the membrane. A magnetization rotation model was developed to quantify the magnetoelectric coupling and explain the underlying physics.

The MOKE system was adapted to measure the oscillation of the magnetization vector ($\Delta \gamma$) of Ni due to an AC electric field applied to the PLZST layer of the sample at each applied steady magnetic field. The AC voltage created oscillation of strain induced anisotropy energy $\Delta K$, resulting in oscillation of magnetization direction of Ni. $\Delta \gamma$, which appeared as the AC component of the MOKE signal, $\Delta \delta/\delta$, and was recorded using a phase lock amplifier.

The experiment was conducted at various electric fields and the AC component of the MOKE signal was measured. The MOKE AC signal can be converted to $\Delta \gamma$ by considering the MOKE signal magnitude at saturation of $M$-$H$ loop where magnetization vector of Ni rotates by 90$^\circ$ from in-plane direction to out-of-plane direction ($\Delta \gamma = 90^\circ$).

$\Delta \gamma$ was plotted against magnetic field at every applied AC voltage. To minimize the experimental error, at least 5 curves were recorded for each voltage and the average curves were used for analysis. As shown in Fig. 3(a), the dependence of $\Delta \gamma$ on the magnetic field for different amplitudes of the AC voltage is an odd function of the magnetic field in all cases. At high AC voltages, when AFE-FE transformation occurs, by increasing $H$ to $H_{SAT}$, the value of $\Delta \gamma$ increases to its maximum of about 0.5$^\circ$ and then by further increasing $H$ above $H_{SAT}$, it decreases to zero.

This dependence can be explained by calculation of $\Delta \gamma$ from Eq. (2) (see supplementary material14):

$$\Delta \gamma = \cos^{-1} \left( \frac{\mu_0 \cdot H \cdot M_s}{\mu_0 \cdot M_s^2 + 2 \Delta K} \right) - \cos^{-1} \left( \frac{H}{M_s^2} \right). \quad (3)$$

It is clear from Eq. (3) that at $H = 0$, $\Delta \gamma = 0$, consistent with experimental results in Fig. 3(a). For $0 < H < H_{SAT}$ and $-H_{SAT} < H < 0$, one needs to estimate the value of $\Delta K$ as described below.

Since the MOKE signal is proportional to $\cos \gamma$, the MOKE AC signal, for small $\Delta K$, from Eq. (2), can be written as (see supplementary material14):

$$\frac{\Delta \delta}{\delta} = -2\Delta K \mu_0 \cdot M_s^2. \quad (4)$$

The measured value $\Delta \delta/\delta \approx 0.01$, gives $\Delta K \approx -0.5$ kJ/m$^3$ at 18 V (350 kV/cm, saturation electric field) applied to the structure. Setting the calculated $\Delta K$ in Eq. (3), the dependence of $\Delta \gamma$ on magnetic field ($H$) for $-H_{SAT} < H < H_{SAT}$ can be plotted as shown in Fig. 3(b). It is seen that the experimental results are in good agreement with data calculated from the model. At higher field $H > H_{SAT}$, the effect of external magnetic field is much larger than the effect of strain induced anisotropy. Consequently, there are no oscillations of $\gamma$ and the MOKE AC signal and $\Delta \gamma$ return to zero.

Since the strain response due to the PLZST phase transformation is non-linear and magnetoelectric coupling of the multilayer is strain-driven, the multilayer exhibits a non-linear magnetoelectric coupling. The effective magnetoelectric coupling coefficient ($\varepsilon_{eff}$) of the multilayer can be estimated as $\varepsilon_{eff} = \Delta M_{max}/V$. To estimate $\varepsilon_{eff}$, it is necessary to calculate the change of magnetization of Ni due to the 0.5$^\circ$ rotation of magnetization induced by the applied electric field ($\Delta M_{max}$). Since the saturation magnetization of Ni (530 kA/m) corresponds to $\Delta \gamma = 90^\circ$, the 0.5$^\circ$ rotation of magnetization of Ni can cause a change of magnetization of 3 kA/m ($\sim$38 G in Gaussian c.g.s. system). Hence, the effective magnetoelectric coupling coefficient of the multilayer is about 2.1 G/V.

The $\Delta \gamma$ of the membranes clamped to the 15 $\mu$m-thick Si layer was about 60% of that of the membrane without Si (Fig. 3(c)), which is attributed to the substrate clamping effect and smaller electric field induced displacement of
membrane as proven by electric field induced displacement measurements as shown in Fig. 1(b).

Compared to hybrid magnetoelectric heterostructures with magnetic thin films on bulk ferroelectric substrates, the magnetoelectric coupling observed in our Ni/PLZST multilayer thin film structure turned out to be weaker. By using bulk ferroelectric substrate, the DC voltage dependence of $M-H$ loops in hybrid magnetoelectric heterostructures has revealed rotation of easy axis of magnetization from in-plane to the out-of-plane direction, equivalent to $\Delta \gamma = 90^\circ$. Compared to them, and the earlier MOKE techniques measuring remnant magnetization changing with external magnetic field, our proposed AC-mode MOKE method, with minimized leakage current induced heating effects, can give fast, accurate, and quantitative information about the voltage-induced changes in the orientation of magnetization vector as well as the enforced anisotropy and their dependence on external magnetic field.

In conclusion, ME membranes comprising soft ferromagnetic Ni and AFE PLZST layers were proposed and fabricated through a bulk micro-machining process on silicon wafers. A sensitive AC-mode MOKE technique was proposed to quantify the magnetoelectric coupling in the multilayer membranes, in which the electric field-induced magnetization rotation was determined for understanding the underlying coupling mechanisms. The AFE to FE phase transformation of PLZST induced a rotation of magnetization of about $0.5^\circ$ in Ni, persuaded by strain-induced anisotropy of about $-0.5 \text{kJ/m}^3$.

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