Modulation of Spin Dynamics in Ni/Pb(Mg1/3Nb2/3)O3-PbTiO3 Multiferroic Heterostructure

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ABSTRACT

Motivated by the fast-developing spin dynamics in ferromagnetic/piezoelectric structures, this work attempts to manipulate magnonics (spin dynamics) by the converse magnetoelectric (ME) coupling. Herein, electric field (E-field) tuning magnetism, especially the surface spin wave, is accomplished in Ni/0.7Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}-0.3PbTiO\textsubscript{3} (PMN-PT) multiferroic heterostructures. The Kerr signal (∝ magnetization) changes of Ni film are observed when direct current (DC) or alternative current (AC) voltage is applied to PMN-PT substrate, where the signal can be modulated breezily even with no extra magnetic field (H-field) is needed in AC-mode measurement. Deserved to be mentioned, an “1” (i.e., “on”) and “0” (i.e., “off”) surface spin wave switch upon applying an E-field is created at room temperature. In addition, the magnetic anisotropy of heterostructures has been investigated by E-field induced
ferromagnetic resonance (FMR) shift, and a large 490 Oe shift of FMR is determined at the angle of 45° between $H$-field and heterostructure plane.

**Keywords:** spin dynamics, multiferroic heterostructure, non-volatile, magnetic anisotropy, spin wave switch

1. Introduction

Multiferroic heterostructures constricted from ferroelectric (FE) and ferromagnetic (FM) phases are widely applied in new functionalities and devices, such as microwave devices, sensors, logic circuits, nonvolatile memory and spintronics devices [1-3]. In recent years, with the ever-increasing demand for fast, small, energy-efficient and non-volatile electronic devices, the investigation of $E$-field manipulating spin degree of freedom and magnetic states launch a blast of the upsurge in FM/FE heterostructures [4-7]. Compared with conventional current trigger and magnetic field ($H$-field) tuned devices, these $E$-field-manipulatable multiferroic devices exhibit ultra-low power consumption, fast computing, more compact and lightweight [8-10]. Artificial FM/FE multiferroic heterostructures are of great potential for devices due to their strong magnetoelectric (ME) coupling, that is, $E$-field regulates magnetism or vice versa. Various characteristics related to magnetics can be dominated by $E$-field in the multiferroic system, such as Curie temperature ($T_c$) [11,12], magnetic domain [13-15], magnetoresistance [16-18], ferromagnetic resonance (FMR) [9,19], etc.

To realize the ME coupling effect in multiferroics, strain/stress, interfacial charge, and exchange bias interplay can be adjusted in FM/FE multiferroic heterostructures. Recently, the $E$-field control of spin wave excitation in transition based on strain/stress mental oxides has been proposed at low temperatures [20,21]. Clearing the competition and coexistence of those mechanisms also presents an arresting diversity in metal and
metallic oxide. As the typical magneto resistive material, Ni is usually chosen to be used as the ferromagnetic phase since it is a prototype 3d itinerant ferromagnet with a high Curie temperature \( T_c = 631 \text{ K} \) [22]. Moreover, Ni is also an ideal material for extensive applications in sensors due to its high temperature stability, anticorrosion, and abrasive resistance [23-27]. Ni/0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3 (PMN-PT) single-crystal is a well-known piezoelectric material, which is desired for FM/FE heterostructures because of its giant anisotropic piezoelectric coefficients and low loss tangents [28-32]. Enlightened by the accomplishment of above, we devoted exploring \( E \)-field dominate magnonics. In this work, it was observed that the applied voltage has deformed lattice, leading to an FMR shift and spin wave excitation at room temperature in Ni/PMN-PT. The tunable multiferroic heterostructures represent great opportunities for new electronic devices.

2. Experimental

In this experiment, the Ni/PMN-PT multiferroic heterostructures were fabricated by direct sedimentation of Ni films onto the commercial and polished 011-oriented PMN-PT using a magnetron sputtering method. The vacuum of the main chamber reached \( 7 \times 10^{-7} \) Torr before growing Ni film, and the temperature of PMN-PT was maintained at 200 \( \text{o} \)C and sputtering power was 80 W with the constant argon pressure of \( 4 \times 10^{-2} \) Torr during deposition.

The surface and cross-sectional morphologies of the Ni/PMN-PT heterostructures were characterized by atomic force microscopy (AFM, Asylum Research, MFP-3D) and focused ion beam scanning electron microscope (FIB-SEM, Thermo Scientific, Scios 2), respectively. The Kerr hysteresis loops were determined by magneto-optical Kerr effect (MOKE, Durham Magneto Optics Ltd, Nano MOKE\(^\text{TM} \) 3) magnetometer, and the FMR was performed with an electron spin resonance (ESR, JEOL, JES-FA200).
system. The direct current (DC) voltage was applied by a Keithley 2410, and the alternative current (AC) voltage was applied by a Keysight 6804A.

3. Results and discussion

In this work, a reversible $E$-field tuning FMR shift in Ni/PMN-PT heterostructures was achieved by using the pulsed laser deposition (PLD) method. The back of PMN-PT (5×4×0.3 mm$^3$) was coated by a thin layer of platinum as the base electrode, the Ni layer as the top electrode. Fig. 1a shows the schematic diagram of $E$-fields control ESR measurement in Ni/PMN-PT heterostructures at out-of-plane orientation, i.e., $H$-field perpendicular to the sample plane (90°), where in-plane orientation is $H$-field parallel to the sample plane (0°). The PMN-PT single crystal produced strain when a vertical applied $E$-field across it, the $P$-$E$ loop and $\varepsilon_{33}$-$E$ curve of PMN-PT are displayed in Fig. 1b, which presents the PMN-PT with a coercive field of 2.6 kV/cm, and a remnant and saturation polarization of 52 and 62 μC cm$^{-2}$ under 10 kV/cm.

**Fig. 1.** (a) Schematic illustration of ESR measurement in the Ni/PMN-PT multiferroic heterostructure (out-of-plane). (b) The polarization and out-of-plane strain as a function of the $E$-field across the PMN-PT (011) single crystal.

As shown in Fig. 2a, the Ni film exhibits a smooth and dense surface at the range of 2×2 μm$^2$. The cross-sectional morphology image presents that the thickness of Ni
film is about 140 nm in Fig. 2b.

**Fig. 2.** (a) Surface and (b) cross-sectional morphology images of Ni thin film grown on PMN-PT.

Fig. 3a shows the schematic diagrams of MOKE measurement under varying applied voltages. A laser beam was focused on the surface of Ni film, and then DC or AC voltage will be applied to the PMN-PT substrate, where no external $H$-field was used in AC-mode measurement. Fig. 3b and Fig. 3c display the in-plane Kerr hysteresis loops of Ni/PMN-PT under different $E$-fields when DC voltages were applied perpendicularly to substrate. It can be calculated that when the $E$-field increases to 6.7 and -6.7 kV/cm, the coercive field decreases by 10 and 20 Oe, respectively. Fig. 3d presents a butterfly-shaped Kerr-$E$ curve, which is similar to the $\varepsilon_{33}$-$E$ loop of PMN-PT (Fig. 1b), providing specific evidence for stain-mediated converse ME coupling in Ni/PMN-PT, which arose from $E$-field-induced strain in PMN-PT and then delivered to Ni layer, attributing to the magnetoelastic effect.
Fig. 3. (a) Schematic illustration of Ni/PMN-PT multiferroic heterostructure by MOKE measurements. (b) In-plane Kerr hysteresis loops under positive and (c) negative voltage. (d) $E$-field dependence of Kerr signal without external $H$-field in Ni/PMN-PT multiferroic heterostructure.

From the above result, it can be inferred that the magnetic anisotropy of the Ni/PMN-PT is due to the strain-mediated converse ME coupling. To prove it, the voltage-regulated ESR tests are compared to theoretical calculation value. The $E$-field-induced effective magnetic field ($H_{\text{eff}}$) along different orientations is expressed as

$$H_{\text{eff},[011]} = \frac{3\lambda Y}{M_s(1+\nu)} (d_{31} - d_{32})E$$  \hspace{1cm} (1)

$$H_{\text{eff},[100]} = \frac{-3\lambda Y}{M_s(1+\nu)} (d_{31} - d_{32})E$$  \hspace{1cm} (2)

$$H_{\text{eff},[011]} = \frac{-3\lambda Y}{M_s(1+\nu)} (d_{31} + d_{32})E$$  \hspace{1cm} (3)

where $Y$ is Young’s modulus of Ni film ($2\times10^{12}$ dyne/cm$^2$) and $\nu$ is Poisson’s ratio of
0.3 [33,34], and \( \lambda \) is the magnetostriction constant of Ni film (~24 ppm). Based on Eq. (1), the calculated effective magnetic field is

\[
H_{\text{eff},[0\overline{1}1]} = 469 \text{ Oe}
\]

(4)

Under a cyclic \( E \)-field of ±6.7 kV/cm, FMR spectra were obtained at the angles of 0° (i.e., in-plane direction), 45° and 90° (i.e., out-of-plane direction) between \( H \)-field and heterostructure plane, which are shown in Fig. 4. The maximum shifts of FMR field (\( \Delta H_t \)) are 410, 490 and 210 Oe at 0°, 45° and 90° as shown in Fig. 4a, 4b and 4c, corresponding to mean ME coupling coefficients \( \alpha = \Delta H_t / \Delta E \) come to 102.5, 122.5 and 52.5 Oe · cm · kV\(^{-1} \), respectively. The complete spectra of FMR varying with \( E \)-field at the three angles are presented in Fig. S1, S2 and S3. It is worth mentioning that the observed value of 410 Oe is approximate to the calculated value of 469 Oe (Eq. (4)).

At the above experimental stage, not only a large FMR shift of 490 Oe is observed, but reversible \( E \)-field tunability also can be shown completely in Ni/PMN-PT multiferroic heterostructure.

Moreover, the relationship between FMR and \( E \)-field are also plotted in Fig. 4d, 4e and 4f. Interestingly, the tunable FMR by \( E \)-field exhibited characteristic butterfly shape, further reveal this converse ME coupling caused by strain/stress mechanism in Ni/PMN-PT multiferroic heterostructure. Meanwhile, the FMR changes with \( E \)-field appear opposite trend at 0°, 45° and 90°, which indicating that magnetic anisotropy exists in the Ni thin film, and this opposite trend maybe resulting from strain response in different crystalline directions of the 011 cut PMN-PT substrate.
Fig. 4. $E$-field dependence of FMR at the angles of 0°, 45° and 90° between $H$-field and heterostructure plane in Ni/PMN-PT multiferroic heterostructure.

It’s worth mentioning that both FMR and surface spin wave modes are observed under $E$-fields (as shown in Fig. 5a) at the angle of 80° between $H$-field and heterostructure plane in Ni/PMN-PT heterostructure at room temperature. Clearly, the surface spin wave is excited by a small $E$-field of 3.3 kV/cm, which has never been reported. This is probably owing to the unequal upper and lower interface conditions of the Ni layer, that is, one free surface and one faying surface with the PMN-PT. At this angle, $E$-fields can regulate the surface spin wave “1” and “0” as shown in Fig. 5b. Therefore, switching control can be realized in spin dynamics devices.
Fig. 5. (a) $E$-field dependence and (b) control of spin wave in Ni/PMN-PT multiferroic heterostructure at room temperature.

4. Conclusions

In summary, $E$-field-tuning non-volatile magnetic anisotropy and surface spin wave switch has been successfully certified in Ni/PMN-PT multiferroic heterostructures, wherein a conspicuous $E$-field regulated FMR shift of ca. 490 Oe arose from strain/stress effect was observed, and the ME coupling coefficient $\alpha$ comes to 122.5 Oe $\cdot$ cm $\cdot$ kV$^{-1}$. Furthermore, at the angle of 80° between $H$-field and heterostructure plane, the surface spin wave mode can be switched “1” or “0” via applied $E$-field particularly was achieved at room temperature, which might be enabled for $E$-field controllable logic device. The achievements of this work make the Ni/PMN-PT multiferroic heterostructures promising candidates for developing novel $E$-field tunable magnonics or spintronics devices.

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References

[1] Guo Q, Xu X, Wang F, et al. In-plane electric field controlled ferromagnetism and anisotropic magnetoresistance in a LSMO/PMN-PT heterostructure. *Nanotechnology* 2018, **29**: 224003.

[2] Hu JM, Li Z, Wang J, et al. Electric-field control of strain-mediated magnetoelectric random access memory. *J Appl Phys* 2010, **107**: 093912.

[3] Zhao S, Zhou Z, Peng B, et al. Quantitative determination on ionic-liquid-gating control of interfacial magnetism. *Adv Mater* 2017, **29**: 1606478.

[4] Cui J, Hockel J L, Nordeen P K, et al. Giant electric-field-induced magnetic anisotropy reorientation with patterned electrodes on a Ni thin film/lead zirconate titanate heterostructure. *J Appl Phys* 2014, **115**: 17C711.

[5] Chen C, Barra A, Mal A, et al. Voltage induced mechanical/spin wave propagation over long distances. *Appl Phys Lett* 2017, **110**: 072401.

[6] Gomez JE, Vargas JM, Aviles-Felix L, et al. Magnetoelectric control of spin currents. *Appl Phys Lett* 2016, **08**: 242413.

[7] Zhao YL, Sun Y, Pan LQ, et al. Probing ferromagnetic/ferroelectric interfaces via spin wave resonance. *Appl Phys Lett* 2013, **102**: 042404.

[8] Liu W, Liu M, Ma R, et al. Mechanical strain-tunable microwave magnetism in flexible CuFe$_2$O$_4$ epitaxial thin film for wearable sensors. *Adv Funct Mater* 2018, **28**: 1705928.

[9] Liu M, Howe BM, Grazulis L, et al. Voltage-impulse-induced non-volatile ferroelastic switching of ferromagnetic resonance for reconfigurable
magnetoelectric microwave devices. *J Adv Mater* 2013, **25**: 4886-4892.

[10] Figerez SP, Tadi KK, Sahoo KR, *et al*. Molybdenum disulfide–graphene van der Waals heterostructures as stable and sensitive electrochemical sensing platforms. *Tungsten* 2020, **2**: 411-422.

[11] Thiele C, Dorr K, Bilani O, *et al*. Influence of strain on magnetization and magnetoelectric effect in La$_{0.7}$A$_{0.3}$MnO$_3$/PMN-PT(001) (A=Sr; Ca). *Phys Rev B* 2006, **75**: 054408.

[12] Molegraaf HJA, Hoffman J, Vaz CAF, *et al*. Magnetolectric effects in complex oxides with competing ground states. *Adv Mater* 2009, **21**: 3470-3474.

[13] Zhang Y, Chen XY, Xie B, *et al*. Leakage current characteristics of SrTiO$_3$/LaNiO$_3$/Ba$_{0.67}$Sr$_{0.33}$TiO$_3$/SrTiO$_3$ heterostructure thin films. *Rare Met* 2021, **40**: 961-967.

[14] Liou YD, Chiu YY, Hart RT, *et al*. Deterministic optical control of room temperature multiferroicity in BiFeO$_3$ thin films. *Nat Mater* 2019, **18**: 580-587.

[15] Li C, Huang L, Li T, *et al*. Ultrathin BaTiO$_3$-based ferroelectric tunnel junctions through interface engineering. *Nano Lett* 2015, **15**: 2568-2573.

[16] Garcia V, Bibes M, Bocher L, A, *et al*. Ferroelectric control of spin polarization. *Science* 2010, **327**: 1106-1110.

[17] Hu JM, Li Z, Chen LQ, *et al*. Design of voltage-controlled magnetic random access memory based on anisotropic magnetoresistance in a single magnetic layer. *Adv Mater* 2012, **24**: 2869-2873.

[18] Oh N, Park S, Kim Y, *et al*. Magnetic properties of M-type strontium ferrites with different heat treatment conditions. *Rare Met* 2021, **39**: 84-88.

[19] Zhu M, Nan T, Peng B, M, *et al*. Advances in magnetics epitaxial multiferroic heterostructures and applications. *IEEE T Magn* 2017, **53**: 1-16.
[20] Xu H, Feng M, Liu M, et al. Strain-mediated converse magnetoelastic coupling in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ multiferroic heterostructures. *Cryst growth des* 2018, **18**: 5934-5939.

[21] Lou J, Liu M, Reed D, et al. Giant electric field tuning of magnetism in novel multiferroic FeGaB/lead zinc niobate–lead titanate (PZN-PT) heterostructures. *Adv Mater* 2009, **21**: 4711-4715.

[22] Behroozfar A, Bhuiyan MEH, Daryadel S, et al. Additive printing of pure nanocrystalline nickel thin films using room environment electroplating. *Nanotechnology* 2019, **31**: 055301.

[23] Luo JK, Flewitt AJ, Spearing SM, I. et al. Young's modulus of electroplated Ni thin film for MEMS applications. *Mater Lett* 2004, **58**: 2306-2309.

[24] Ito T, Ushiyama T, Yanagisawa Y, et al. Growth of highly insulating bulk single crystals of multiferroic BiFeO$_3$ and their inherent internal strains in the domain-switching process. *Cryst Growth Des* 2011, **11**: 5139-5143.

[25] Luo JK, Pritschow M, Flewitt AJ, et al. Effects of process conditions on properties of electroplated Ni thin films for microsystem applications. *J Electrochem Soc* 2006, **153**: D155.

[26] Gilbert I, Chavez AC, Pierce DT, et al. Magnetic microscopy and simulation of strain-mediated control of magnetization in PMN-PT/Ni nanostructures. *Appl Phys Lett* 2016, **109**: 162404.

[27] Bai H, Li J, Hong Y, et al. Enhanced ferroelectricity and magnetism of quenched (1-x)BiFeO$_3$-xBaTiO$_3$ ceramics. *J Adv Ceram* 2020, **9**, 511-516.

[28] Han PD, Yan WL, Tian J, et al. Cut directions for the optimization of piezoelectric coefficients of lead magnesium niobate-lead titanate ferroelectric crystals. *Appl Phys Lett* 2005, **86**: 052902.
[29] Zheng L, Jing Y, Lu X, et al. Temperature and electric-field induced phase transitions, and full tensor properties of [011]C-poled domain-engineered tetragonal 0.63Pb(Mg_{1/3}Nb_{2/3})O-0.37PbTiO_3 single crystals. *Phys Rev B* 2016, **93**: 094104.

[30] Wu T, Zhao P, Bao M, et al. Domain engineered switchable strain states in ferroelectric (011)[Pb(Mg_{1/3}Nb_{2/3})O_3](1-x)-[PbTiO_3]x (PMN-PT, x≈0.32) single crystals. *J Appl Phys* 2011, **109**: 124101.

[31] Jung J, Lee W, Kang W, et al. Review of piezoelectric micromachined ultrasonic transducers and their applications. *J Micromech Microeng* 2017, **27**: 113001.

[32] Ye F, Dai H, Peng K, et al. Effect of Mn doping on the microstructure and magnetic properties of CuFeO_2 ceramics. *J Adv Ceram* 2020, **9**: 444-453.

[33] Liu M, Obi O, Cai Z, et al. Electrical tuning of magnetism in Fe_3O_4/PZN-PT multiferroic heterostructures derived by reactive magnetron sputtering. *J Appl Phys* 2010, **107**: 073916.

[34] Handley RC. Modern magnetic materials: principles and applications. New York, *Wiley* 2000.
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