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Electric field (E-field) tunable multiferroic devices such as tunable RF devices and memory storage mediums require strong magnetoelectric interactions. In this paper, we demonstrated a Terfenol-D/ lead zinc niobate-lead titanate composite bilayer structure with an E-field-induced magnetic anisotropy field of 3500 Oe and a magnetoelectric coefficient of 580 Oe cm/kV. This is the largest E-field induced anisotropy reported to date, resulting in a strong dependence of magnetization process and tunable ferromagnetic resonance in both amorphous and crystalline Terfenol-D films. In addition, the magnetostriction constants of crystalline and amorphous Terfenol-D were estimated to be 420 ppm and 320 ppm, respectively, which are close to report values. Electrically manipulating magnetic performance holds great potential for future ultra-low power, lightweight, tunable magnetic devices, and memory storage. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4754424]

I. INTRODUCTION

Control of magnetic film properties by E-field is of fundamental and technological importance for realizing faster, smaller, and ultra-low power electronics.[1,2] Multiferrroic materials that simultaneously exhibit ferroelectric, (anti)ferromagnetic, and ferroelastic ordering have attracted widespread interest over the past years.[3–9] In particular, multiferroic composites possessing magnetostrictive and piezoelectric phases lead to a strain-mediated magnetoelectric (ME) interaction that allows E-field control of magnetism. Strong ME coupling was recently demonstrated in layered composites of ferrite and ferromagnetic metals in combination with ferroelectric relaxors of PZN-PT (lead zinc niobate-lead titanate) and PMN-PT (lead magnesium niobate-lead titanate).[10,11] Applying an E-field on Fe_3O_4/PZN-PT, a magnetic anisotropy of 860 Oe was produced, resulting in a ME coefficient of $\alpha = dH/dE = 108$ Oe·cm/kV.[11] This ME coupling strength, however, is considerably lower than theoretical prediction.[12] Strong ME coupling, large magnetostriction and high piezoelectric coefficients are critical for enhanced performance of these materials, and more efforts are needed to develop new types of ultralow power spintronics, memory devices and tunable microwave devices.[6,12–23] In this study, we report a significant E-field-induced effective magnetic anisotropy up to 3500 Oe in Terfenol-D/PZN-PT which is four times more than that we demonstrated in Fe_3O_4/PMN-PT heterostructures.

This giant ME coupling is comparable with the recent report where a latent magnetic anisotropy was produced with applying a large electric field in $BiFeO_3$ crystal and enabled a significant manipulation of spin wave.[24]

Terfenol-D ($Tb_{0.7}Dy_{0.3}Fe_2$) exhibits the highest room-temperature magnetostriction value and has been widely used as actuator and sensor in either bulk or thin film forms.[25,26] Based on our previous model,[11] the ME coefficient is proportional to the product of magnetostriction coefficient (λ) and piezoelectric coefficient (d), which would enable a giant E-field induced magnetic anisotropy once incorporating with ferroelectric materials,. In this work, amorphous and crystalline Terfenol-D films were grown on single crystal (011) cut PZN-PT substrates that possess giant in-plane piezoelectric coefficients ($d_{31} = -3000$ pC/N [100]; $d_{32} = 1100$ pC/N [01$\bar{1}$]). A substantial effective magnetic field of 3500 Oe was produced by applying an E-field of 6 kV/m on polycrystalline Terfenol-D/PZN-PT, corresponding to a ME coefficient of 580 Oe cm/kV. This strong E-field-induced magnetic anisotropy has many applications with tunable magnetic devices by applying electric fields rather than magnetic fields generated by electromagnets, which are bulky, noisy, slow, and energy consuming. Such ME devices could also be miniaturized and integrated with semiconductor processing technology.

II. EXPERIMENT

Amorphous and crystalline Terfenol-D films were deposited on Pt coated Si substrates and PZN-PT slabs, respectively, (10 mm (L) × 5 mm (W) × 0.5 mm (T)) by

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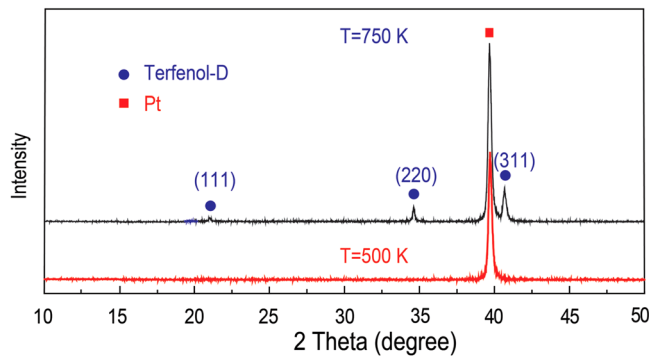
FIG. 1. XRD profiles of Terfenol-D prepared on Pt/Si substrate at 500 K and 750 K.

magnetron sputtering under a variety of temperatures. The base pressure of chamber was 1×10^{-8} Torr while the deposition pressure was set at 3 mTorr. Accounting for the relative sputtering yields of two independent targets $Tb_{0.3}Dy_{0.7}$ and Fe, we prepared Terfenol-D films by co-sputtering with an estimated composition of $Tb_{0.3}Dy_{0.7}Fe_2$ and thickness of approximately 100 nm. The microstructure of the film was investigated by using x-ray diffraction (XRD) measurements. Room-temperature magnetization characterization was measured by a vibrating sample magnetometer (VSM). E-field dependence of microwave performance was quantitatively measured by a ferromagnetic resonance test system with the working frequency of 9.3 GHz.[22]

III. RESULTS AND DISCUSSION

Figure 1 presents x-ray diffraction profiles of Terfenol-D films grown on Pt/Si substrates under temperature of 500 K and 750 K. Amorphous phase of Terfenol-D prepared at 500 K was identified without the presence of any diffrac-

tion peaks except from the substrates and Pt. The crystal structure of Terfenol-D grown at 750 K was confirmed by the existence of several broad polycrystalline diffraction peaks, indicating the right composition of Terfenol-D. No evidence of any secondary phase was found from x-ray measurements.

Figure 2 shows the growth temperature dependence of magnetic hysteresis loops for Terfenol-D films prepared on Pt/Si substrates. The film prepared at 500 K (Fig. 2(a)), characterized as amorphous phase, exhibits a well-defined magnetic easy axis perpendicular to the film, which has been reported in many studies and could attribute from the growth condition induced magnetic anisotropy.[25] As the substrate temperature increased to 600 K (Fig. 2(b)), the magnetic property still shows amorphous behavior with the coercive field less than 50 Oe, however, the magnetization process now exhibits an easy in-plane and an out-of-plane hard axis implying a tensile stress was produced as the deposition temperature increase, with the magnetic anisotropy compressed from out-of-plane to in-plane. For crystalline Terfenol-D film grown at 750 K (Fig. 2(c)), an open magnetic hysteresis loop is observed with the coercive field above 2000 Oe. In-plane ferromagnetic resonance (FMR) was measured by our FMR test system, showing a large FMR linewidth of 1200 Oe. The growth temperature dependence of magnetic properties in Terfenol-D film suggests that the magnetostriction in amorphous or crystalline phases of Terfenol-D is considerably large, indicating stress-induced magnetic anisotropy is extremely sensitive to the growth conditions. This provides more opportunities to prepare Terfenol-D films with tailored magnetic properties for specific applications by adjusting the growth conditions.

Strain-engineered E-field tuning of magnetic properties of the Terfenol-D/PZN-PT film-substrate system was examined by E-field-induced changes in magnetization process as
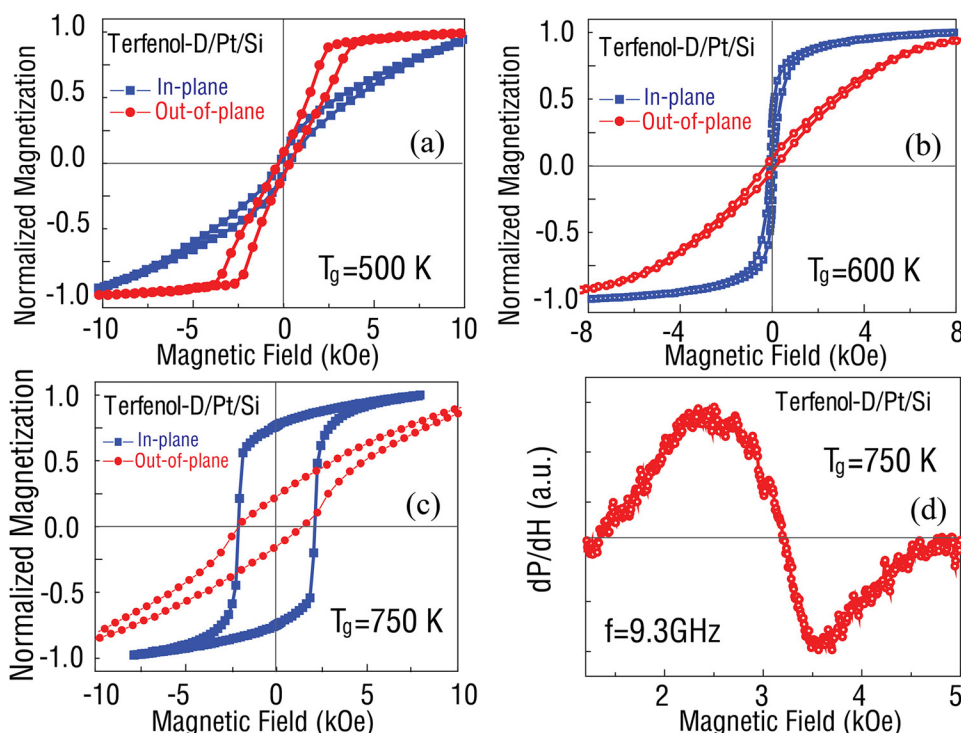

FIG. 2. (a,b,c) Magnetic Hysteresis loops of Terfenol-D/Pt/Si deposited at T = 500 K (a), 600 K (b), and 750 K (c). (d) Ferromagnetic resonance profile of Terfenol-D/Pt/Si prepared at 750 K.
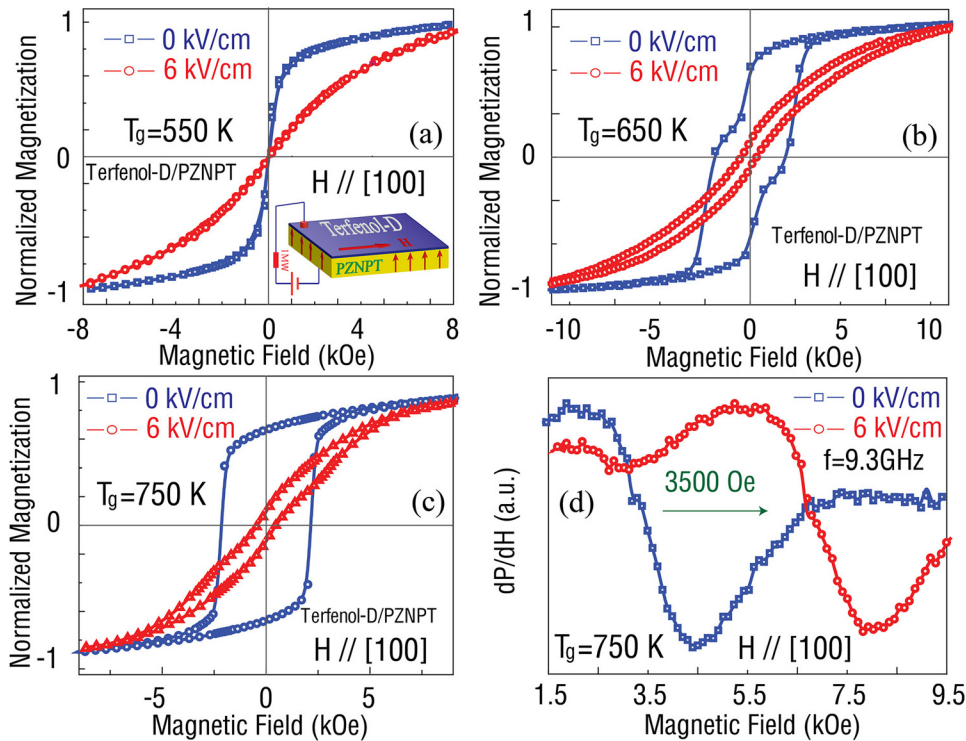
FIG. 3. E-field dependence of in-plane magnetic hysteresis loops of Terfenol-D/PZN-PT deposited at 550 K (a), 650 K (b), and 750 K (c). Inset in (a) is the measurement configuration: E-field is applied through the thickness direction and magnetic field is applied along in-plane [100] direction. (d) FMR spectra of polycrystalline Terfenol-D/PZN-PT at E = 0 kV/cm (blue) and E = 6 kV/cm (red).

shown in Figure 3. For both amorphous (Fig. 3(a)) and crystalline (Fig. 3(c)) or intermediate (Fig. 3(b)) Terfenol-D films, applying an E-field through the thickness direction of PZN-PT, in-plane magnetization process along [100] direction becomes increasingly more difficult, indicating a magnetic anisotropy was produced perpendicular to in-plane [100] orientation. This phenomenon can be interpreted as strong magnetoelastic and piezoelectric effects. A dc field applied across the PZN-PT substrate results in a compressive stress along the in-plane [100] direction caused by the inverse piezoelectric effect. Once this mechanical strain coherently transfers to the Terfenol-D film, the magnetic moment undergoes a rotation away from the [100] direction due to the magnetoelastic effect. Given the complicated nature of the magnetization process involving domain wall motion, domain rotation, and nonlinear behaviors, E-field-induced magnetic hysteresis loop change is not sufficient to quantitatively determine this magnetic anisotropy which is essential for fundamental and technologic study. In the present study, a field-swept ferromagnetic resonance measurement was performed to quantitatively determine the magnetic anisotropy produced by an E-field.[23] For magnetic thin films, the in-plane resonance frequency as a function of resonance field can be expressed as

$$f = \gamma \sqrt{(H_r + H_k + H_{eff})(H_r + H_k + H_{eff} + 4\pi M_s)}, \quad (1)$$

where γ is the gyromagnetic ratio ~ 2.8 MHz/Oe, H_r is the FMR field, H_k is the anisotropic field in plane, and $4\pi M_s$ is the magnetization of Terfenol-D film. H_{eff} is the E-field-induced effective magnetic anisotropy. Through observing E-field-induced resonance field change, the effective magnetic anisotropy can be quantitatively confirmed by $H_{eff} = \Delta H_r$. According to the E-field dependence of FMR spec-

trum plotted in Fig. 3(d) for crystalline Terfenol-D/PZN-PT, the resonance field H_r is shifted upward by 3500 Oe as applying an electric field of 6 kV/m, indicating a record-high E-field-induced magnetic anisotropy of 3500 Oe is achieved, corresponding to a magnetoelectric coefficient of 580 Oe cm/kV. Such E-field-induced FMR shift is consistent with E-field-induced hard magnetization process previously observed. Figure 4 shows E-field dependence of ferromagnetic resonance field for both amorphous and polycrystalline Terfenol-D/PZN-PT composites. As increasing electric fields, the resonance fields or effective magnetic fields were dramatically increased up to 3500 Oe and 2700 Oe for polycrystalline and amorphous Terfenol-D/PZN-PT, respectively. By taking account of the mode in Ref. 22, the magnetostriction constants of Terfonel-D crystalline and amorphous films are estimated to be 420 ppm and 320 ppm, respectively, which is
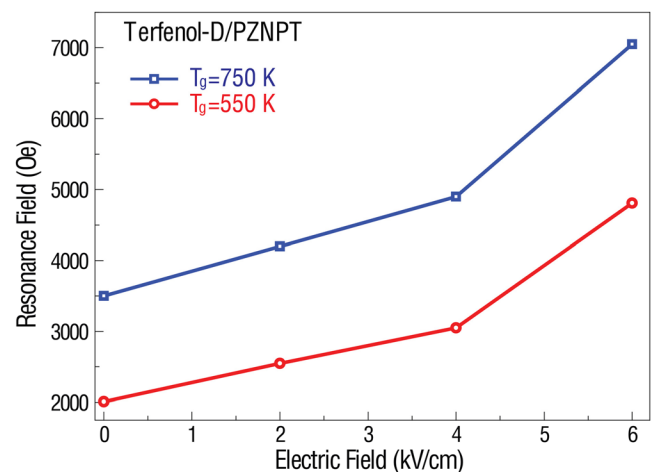

FIG. 4. E-field dependence of resonance field for polycrystalline (red) and amorphous (blue) Terfenol-D/PZN-PT.

close to other reports.[23] In addition, the negligible change in FMR linewidth implies a homogeneous displacement in magnetic films caused by electric fields.

IV. CONCLUSION

An energy-efficiency technique of E-field tuning of magnetic properties has been demonstrated in amorphous and crystalline Terfenol-D/PZN-PT multiferroic heterostructure. A record-high E-field-induced magnetic anisotropy of 3500 Oe, corresponding to a magnetoelectric coefficient of 580 Oe cm/kV, was quantitatively determined through the shift of FMR profiles under various electric fields. The magnetostriction constants of polycrystalline and amorphous Terfenol-D films are estimated to be 420 ppm and 320 ppm, respectively, which are much less than those observed in bulk but close to those reported for thin films. Efforts are underway to design and fabricate a tunable dual-channel transducer device.

ACKNOWLEDGMENTS

[1]W. Eerenstein, N. D. Mathur, and J. F. Scott, Nature (London) **442**, 759 (2006).

[2]W. Eerenstein, M. Wiora, J. L. Prieto, J. F. Scott, and N. D. Mathur, Nature Mater. **6**, 348 (2007).

[3]C. W. Nan, M. I. Bichurin, S. X. Dong, D. Viehland, and G. Srinivasan, J. Appl. Phys. **103**, 031101 (2008).

[4]Y. H. Chu, L. W. Martin, M. B. Holcomb, M. Gajek, S. J. Han, Q. He, N. Balke, C. H. Yang, D. Lee, W. Hu, Q. Zhan, P. L. Yang, A. Fraile-Rodri-guez, A. Scholl, S. X. Wang, and R. Ramesh, Nature Mater. **7**, 678 (2008).

[5]X. He, Y. Wang, N. Wu, A. N. Caruso, E. Vescovo, K. D. Belashchenko, P. A. Dowben, and C. Binek, Nature Mater. **9**, 579 (2010).

[6]M. Bibes and A. Barthelemy, Nature Mater. **7**, 425 (2008).

[7]E. Y. Tsymbal and H. Kohlstedt, Science **313**, 181 (2006).

[8]C. A. F. Vaz, J. Hoffman, C. H. Anh, and R. Ramesh, Adv. Mater. **22**, 2900 (2010).

[9]N. A. Spaldin, S. W. Cheong, and R. Ramesh, Phys. Today **63**, 38 (2010).

[10]J. Lou, M. Liu, D. Reed, Y. H. Ren, and N. X. Sun, Adv Mater. **21**, 4711 (2009).

[11]M. Liu, O. Obi, J. Lou, Y. J. Chen, Z. H. Cai, S. Stoute, M. Espanol, M. Lew, X. Situ, K. S. Ziemer, V. G. Harris, and N. X. Sun, Adv. Funct. Mater. **19**, 1826 (2009).

[12]G. Srinivasan, in *Annual Review of Materials Research* (2010), Vol. 40, p. 153.

[13]Y. K. Fetisov and G. Srinivasan, Appl. Phys. Lett. **88**, 143503 (2006).

[14]J. H. Lee, L. Fang, E. Vlahos, X. L. Ke, Y. W. Jung, L. F. Kourkoutis, J. W. Kim, P. J. Ryan, T. Heeg, M. Roeckerath, V. Goian, M. Bernhagen, R. Uecker, P. C. Hammel, K. M. Rabe, S. Kamba, J. Schubert, J. W. Freeland, D. A. Muller, C. J. Fennie, P. Schiffer, V. Gopalan, E. Johnston-Halperin, and D. G. Schlom, Nature (London) **476**, 114 (2011).

[15]M. Liu, X. Li, H. Imrane, Y. J. Chen, T. Goodrich, Z. H. Cai, K. S. Ziemer, J. Y. Huang, and N. X. Sun, Appl. Phys. Lett. **90**, 152501 (2007).

[16]M. Liu, S. D. Li, O. Obi, J. Lou, S. Rand, and N. X. Sun, Appl. Phys. Lett. **98**, 222509 (2011).

[17]J.-M. Hu and C. W. Nan, Phys. Rev. B **80**, 224416 (2009).

[18]T. Wu, A. Bur, K. Wong, J. Leon Hockel, C.-J. Hsu, H. K. D. Kim, K. L. Wang, and G. P. Carman, J. Appl. Phys. **109**, 07D732 (2011).

[19]S. X. Dong, J. Y. Zhai, J. F. Li, and D. Viehland, Appl. Phys. Lett. **88**, 082907 (2006).

[20]J. Das, Y. Y. Song, N. Mo, P. Krivosik, and C. E. Patton, Adv. Mater. **21**, 2045 (2009).

[21]P. Zhao, Z. L. Zhao, D. Hunter, R. Suchoski, C. Gao, S. Mathews, M. Wuttig, and I. Takeuchi, Appl. Phys. Lett. **94**, 243507 (2009).

[22]D. A. Filippov, G. Srinivasan, and A. Gupta, J. Phys. Condens. Matter **20**, 425206 (2008).

[23]V. M. Petrov, G. Srinivasan, M. I. Bichurin, and A. Gupta, Phys. Rev. B **75**, 224407 (2007).

[24]P. Rovillain, R. de Sousa, Y. Gallais, A. Sacuto, M. A. Méasson, D. Colson, A. Forget, M. Bibes, A. Barthélémy, and M. Cazayous, Nature Mater. **9**, 975 (2010).

[25]P. I. Williams, D. G. Lord, and P. J. Grundy, J. Appl. Phys. **75**, 5257 (1994).

[26]A. Speliotis, O. Kalogirou, and D. Niarchos, J. Appl. Phys. **81**, 5696 (1997).