

MAGNETOELECTRIC COUPLING OF MULTIFERROIC $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3\text{-CoFe}_2\text{O}_4$ COMPOSITE THIN FILM WITH SANDWICH STRUCTURE

Shuhong Xie^{*}, Yun Ou^{*}, Feiyue Ma^{**} & Jiangyu Li^{**a)}

^{*} Faculty of Materials, Optoelectronics and Physics, and Key Laboratory of Low Dimensional Materials & Application Technology of Ministry of Education, Xiangtan University, Hunan, 411105, China

^{**} Department of Mechanical Engineering, University of Washington, Seattle, WA 98195, U.S.A.

PCP laminated composite thin film was synthesized by sol-gel process and spinning coating. The ME coupling is produced by mechanical interactions arising from magnetostrictive of CFO and piezoelectric effects of PZT. The ME coupling has been confirmed by evolution of piezoresponse under an external magnetic field and switching spectroscopy piezoresponse force microscopy, showing in plane field magnetically induced ferroelectric domain evolution, increased coercive field and decreased the piezoelectric response.

Multiferroic materials with two or more types of ferroic orders have attracted a great deal of attention in the last decade. In particular, the coexistence of electric and magnetic ordering makes it possible to manipulate the electric state of a multiferroic material through a magnetic field, or vice versa, and thus opens door for a variety of applications [1], such as multiple-state memory devices that can be electrically written and magnetically read, electrically controlled microwave phase shifters, magnetically controlled electro-optic or piezoelectric devices, and so on. However, the magnetoelectric (ME) couplings in single-phase multiferroic materials are usually extremely small [2]. In order to enhance the ME coupling for practical applications, hybrid multiferroic composites consisting of ferroelectric and ferromagnetic phases have been widely explored [3], wherein the two phases are coupled together through mechanical interactions arising from magnetostrictive and piezoelectric effects, the so-called product property. In recent years, both theory and experiment research indicated that the sandwich structure has the higher magnetoelectric coefficient in multiferroic composite thin film. Motivated by these, we synthesized $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3\text{-CoFe}_2\text{O}_4$ (PZT- CFO) composite thin film with sandwich structure (PCP).

ME coupling is a significant parameter to verify the performance of multiferroic PCP thin film. To characterize the ME coupling of PCP thin film, an Asylum Research Variable Field Module (VFM) and switching spectroscopy piezoresponse force microscopy (SSPFM) are used to apply an in-plane magnetic field of 2000 Oe to the specimen during PFM characterization to examine the evolution of ferroelectric domain and polarization switching induced by the external magnetic field.

The schematic of ME coupling setup is shown in Fig. 1(a). An in-plane magnetic field is applied to the PCP thin film using VFM, then examine the induced changes in vertical and lateral PFM to verify the ME coupling in the layer film. Dual frequency resonance tracking technique is used in vertical PFM [4], the amplified and intrinsic vertical piezoresponse of the film can be determined by correcting the quality factor Q , as shown in Fig. 1(b)-(c). The resulting vertical and lateral PFM phase and amplitude mappings before (not shown here) and after the application of magnetic field confirm the ME coupling in PCP thin film. In the absence of external magnetic field, the average magnified and intrinsic vertical PFM amplitude are 857 and 55 pm, and average lateral PFM amplitude is 3.80 nm, respectively. After the application of external magnetic field of 2000 Oe, the corresponding vertical and lateral amplitudes decrease to 479, 29 and 939 pm, respectively. Such changes are caused by the stress constraint on PZT layers owing to CFO layer magnetostriction, which makes it harder for ferroelectric PZT domains to switch, resulting in smaller PFM amplitude. The changes in PFM phase mapping are also observed in Fig. 1(d), more obvious phase contrast with some initially disconnected domains connected together, and some domain separate into several domains, suggesting ferroelectric domain evolution in thin film induced by external magnetic field. Such evolution is induced indirectly through the mechanical interactions between magnetostrictive and piezoelectric strains. The corresponding mappings of quality factor before and after the application of external magnetic field by VFM indicate that the quality factor is clearly reduced in areas where domains are switched, confirming domain switching is a dissipation process. A large variation in quality factor Q is shown in Fig. 1(e), ranging between 10 to 40.

Local switching behavior of PCP thin film bare surface is studied by SSPFM. Fig.2 (a)-(d) are coercive bias mappings and representative hysteresis loops under 0 and 2000 Oe magnetic field. As shown in Fig.2(a) and Fig.2(b), without magnetic field the average coercive voltage of mapping is 10.8 V, with 2000 Oe magnetic field the average coercive voltage increase to 18.5 V, confirming the ME coupling of composite thin film, and indicating that magnetic field increase the coercive bias voltage owing to vertical compressive stress. As seen in Fig. 2(c) and (d), in the absence of external magnetic field, the maximum PFM amplitude is measured to be around 250 nm, and the corresponding coercive voltage is about 14 V. After the application of external magnetic field of 2000 Oe, the maximum PFM amplitude decreases to 120 nm, and the corresponding coercive voltage increases to about 18 V, further confirming the ME coupling in PCP thin film. The observation is also consistent with change in PFM phase and amplitude mappings.

^{a)} Corresponding author. Email:jjli@uw.edu

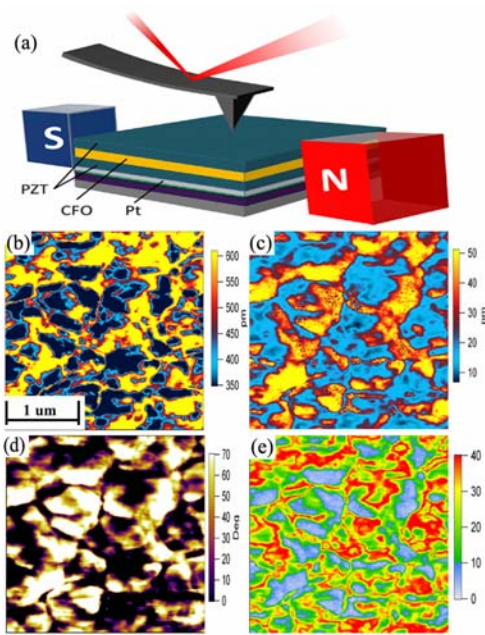


Fig. 1 Dual frequency vertical PFM mapping of PCP thin film after the application of external magnetic field by VFM; (a) the schematic ME coupling setup; (b) magnified amplitude; (c) amplitude; (d) phase and (e) quality factor

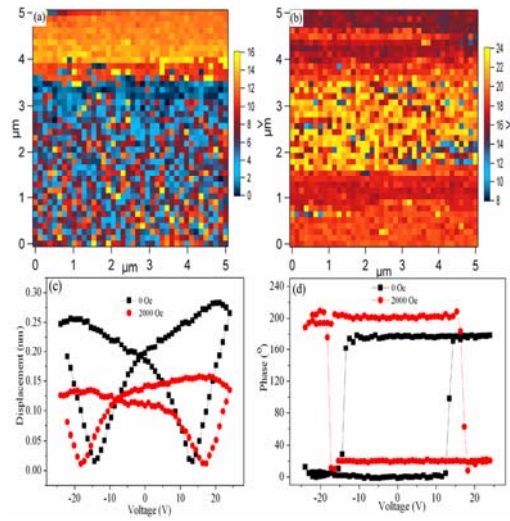


Fig. 2 SSPFM mapping of PCP thin film before and after the application of external magnetic field by VFM; (a) coercive voltage mapping without external magnetic field; (b) coercive voltage mapping under 2000 Oe magnetic field; (c) amplitude-voltage butterfly and (d) phase-voltage hysteresis loops

References

- [1] T. Lottermoser, T. Lonkai, U. Amann, et al, Nature, 2004, 430, 541-544.
- [2] H. Zheng, J. Wang, S. E. Lofland, et al, Science, 2004, 303, 661-663.
- [3] C. W. Nan, M. I. Bichurin, S. X. Dong, et al, J. Appl. Phys., 2008, 103, 031101-35
- [4] S. H. Xie, A. Gannepalli, Q. N. Chen, et al, Nanoscale., 2012, 4, 408-413.