

## Ameliorated Magnetoelectric coupling in Free-Standing Multiferroic Thin Film

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Magnetoelectric characteristics of  $\text{CoFe}_2\text{O}_3$ (CFO)/ $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) thin film on substrate decreased by tensile stress. The free-standing structure can relax the constraint from the substrate, because its hemispherical space separates the film and substrate and thus could improve the ME response to the multiferroic film. Fig. 1(a), (b) show the free-standing (CFO/PZT/LaNiO<sub>3</sub>(LNO)/NiO multiferroic thin film. The magnetoelectric effect in multiferroic materials, electrically or magnetically induced and it is mathematically described by the ME coupling coefficient ( $\alpha$ ),

The ME effect is the change in the electric polarization (P) of the sample due to the application of a magnetic field (H):

$$\alpha = \partial P / \partial H$$

Fig. 3 (a) shows the relative remnant polarization curve corresponding to the applied electric field. The remnant polarization arises corresponding to the increasing applied voltage linearly for planar and non-linearly for free-standing film. In the case of free-standing thin film, the change of remnant polarization value nine times ( $4.5 \mu\text{C}/\text{cm}^2$ ) larger than the planar film ( $0.5 \mu\text{C}/\text{cm}^2$ ) and also these changes of remnant polarization is nonlinear. It causes the dipole alignment along the out-of-plane direction, which can increase the remnant polarization. Fig. 3 (b) shows the relative remnant polarization curve corresponding to the applied magnetic field. These planar and free-standing thin films do not show the remnant polarization below 2 kOe. In the multiferroic composites, the electric polarization in the PZT film induced due to the magnetostriction of the CFO phase, involving the domain-wall motion and domain rotation. Since the free-standing and planar film have the coercivity of 2 kOe (fig.2 (a), (b)). So, it could not induce the strain on the PZT film up to the limit of coercivity. In the case of planar multiferroic film, a constant remnant polarization is 0.06, develops after exposure to the magnetic field of 2 kOe. In the case of free-standing thin film, the change of remnant polarization value nearly four times ( $3.5 \mu\text{C}/\text{cm}^2$ ) larger than the planar film ( $0.9 \mu\text{C}/\text{cm}^2$ ) and these changes of remnant polarization are not constant, and can further linearly increases with increasing the magnetic field owing to the two reasons. One is the enhancement in the magnetostriction of the CFO layer and the other reason is that the relaxation of tensile stress in the PZT layer.

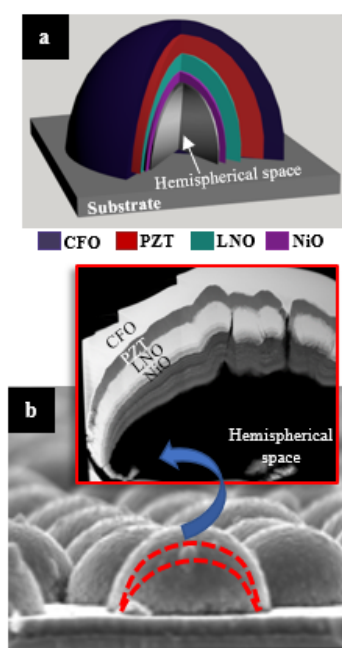


Figure 1. Multiferroic free-standing (CFO/PZT/LNO/NiO) thin film. (a) Schematic Illustration (b) SEM image. Inset: Cross-sectional STEM image

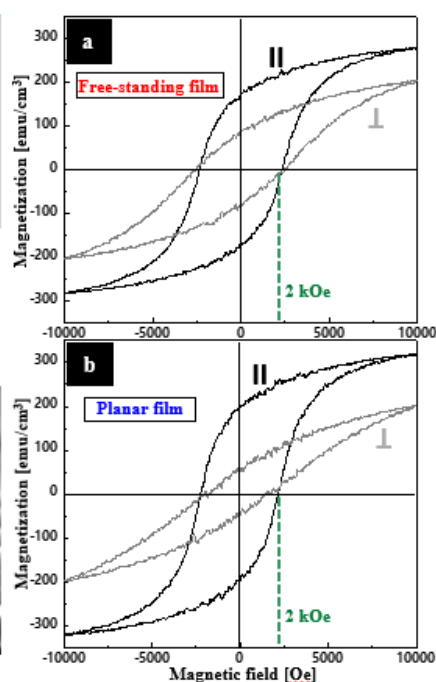


Figure 2. Magnetic hysteresis loop of the multiferroic (CFO/PZT/LNO/NiO) thin film. (a) Planar film (b) Free-standing film

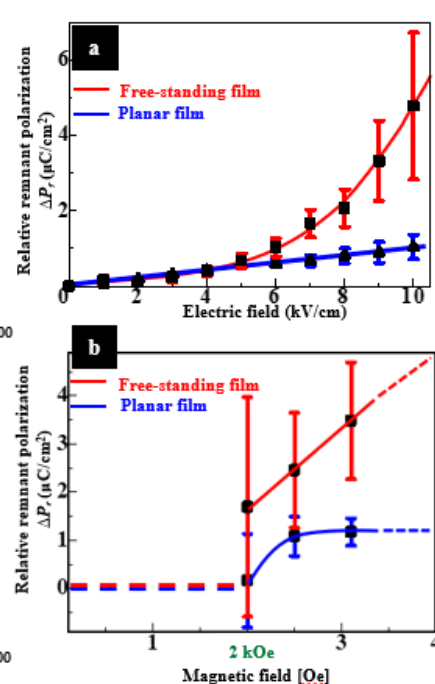


Figure 3. (a) Electric field dependence (b) Magnetic field dependence of the relative remnant polarization between planar and free-standing multiferroic (CFO/PZT/LNO/NiO) thin film