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Voltage impulse induced bistable magnetization switching in multiferroic heterostructures

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We report on voltage impulse induced reversible bistable magnetization switching in FeGaB/lead zirconate titanate (PZT) multiferroic heterostructures at room temperature. This was realized through strain-mediated magnetoelectric coupling between ferroelectric PZT and ferromagnetic FeGaB layer. Two reversible and stable voltage-impulse induced mechanical strain states were obtained in the PZT by applying an electric field impulse with its amplitude smaller than the electric coercive field, which led to reversible voltage impulse induced bistable magnetization switching. These voltage impulse induced bistable magnetization switching in multiferroic heterostructures provides a promising approach to power efficient bistable magnetization switching that is crucial for information storage.

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Magnetic random access memory (MRAM) based on magnetic tunnel junction (MTJ) has attracted a lot of recent interests due to its nonvolatile property, fast speed, and high read and write endurance.^{1,2} Writing the magnetic bits in MRAM has been challenging, which has been traditionally achieved by the Oersted fields produced by large electric currents,³ or more recently with the spin torque effect by passing a spin polarized current through the magnetic film.^{4,5} Both methods need large current or current density, leading to high energy consumption during the writing process.

Voltage control of logic bits in semiconductor logic systems has been a very energy efficient and fast approach, and has been widely used. Similar voltage control of bistable magnetization switching, if achieved, would enable nonvolatile and energy efficient MRAM. Several different approaches have been proposed for achieving voltage control of bistable magnetization switching, including dilute magnetic semiconductors,^{6,7} electric field induced toggle switching of nanomagnets,^{8–10} and strain-mediated converse magnetoelectric coupling in ferromagnetic (FM)/ferroelectric (FE) heterostructures,^{11–13} etc. E-field control of magnetization in dilute magnetic semiconductors has been demonstrated, which, however, is limited to very low temperatures.^{6,7} More recently, electric-field induced toggle switching of nanoscale thin-film magnets also shows great potential in MRAM devices using the electric field induced magnetocrystalline anisotropy (MCA) change.^{8–10}

Voltage control of the bistable magnetization switching has been expected in magnetoelectric random access memory (MERAM) based on magnetoelectric (ME) multiferroic heterostructures.^{14–18} Most recently, Hu *et al.* proposed nanoscale MERAM devices with an ultrahigh storage capacity and a low power dissipation of 0.16 fJ per bit by simulations. Recent demonstration of strong converse magnetoelectric coupling through voltage control of magnetism has shown great potential for voltage tunable RF/microwave

magnetic devices.^{17–19} However, reversible and nonvolatile bistable magnetization switching by voltage impulses in ME multiferroic heterostructures has been elusive. In this paper, we demonstrate a nonvolatile and reversible voltage impulse induced bistable magnetization switching in FeGaB/lead zirconate titanate (PZT) multiferroic ME heterostructures. This power efficient bistable magnetization switching by voltage impulses shows great potential in enabling power efficient MERAM.

Multiferroic heterostructures of FeGaB/PZT were prepared by magnetron-sputtering. 15 nm thick Permalloy (Ni₈₀Fe₂₀) film and 100 nm thick FeGaB film were sequentially deposited onto PZT ceramic slab [10 mm (L) × 2 mm (W) × 0.25 mm (T)] which was pre-poled along its thickness direction, followed by 5 nm Cu for passivation. The Ni₈₀Fe₂₀ thin film was used as a seed layer to increase the magnetic softness and improve adhesion of the FeGaB thin film.²¹ The FeGaB film has a composition of Fe₇₅Ga₁₀B₁₅ and the deposition condition of the FeGaB film was reported elsewhere.^{19,20} Magnetic properties of FeGaB thin film were measured by vibrating-sample magnetometer. Piezoelectric strain of PZT slab was measured by a photonic sensor.

Fig. 1 presents the electric polarization P as a function of the electric field E applied along the thickness direction of the PZT slab, indicating a remnant polarization of $32 \mu\text{C}/\text{cm}^2$ and an electric coercive field E_c of about 8 kV/cm. The curves of in-plane strain ϵ of the PZT slab measured by cycling sinusoidal electric field E with amplitude of 8 kV/cm and 16 kV/cm are also shown in Fig. 1. When $E > E_c$ (i.e., at 16 kV/cm), the whole butterfly curve was observed due to the complete ferroelectric domains switching. On the other hand, only ferroelectric domain wall motion was induced when $E < E_c$ (i.e., at 8 kV/cm), which resulted in a strain ϵ - E hysteresis loop with a large remnant strain. This ϵ - E hysteresis loop generated two different strain states at $E = 0$ depending on the E -field history at $E < E_c$, leading to bistable magnetization states in the FeGaB film through converse magnetoelectric coupling in the FeGaB/PZT heterostructures. Fig. 2 demonstrates the magnetic hysteresis loops under positive and negative E -field of

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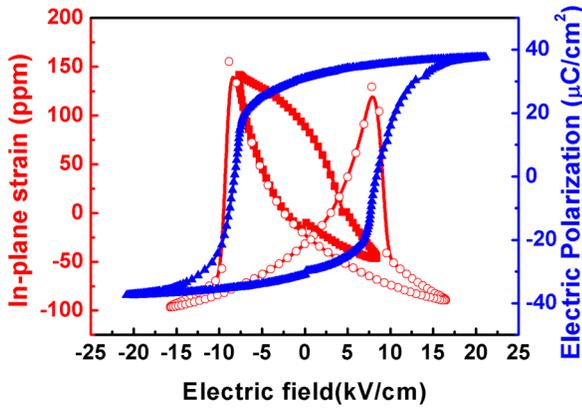


FIG. 1. The polarization-electric field loop of the PZT ceramics and in-plane ϵ -E loops under application of E-field of 16 kV/cm and 8 kV/cm.

± 8 kV/cm measured along the length direction of the FeGaB/PZT heterostructure, respectively. The remnant magnetization ratio shows a large change from 70% to 54% when the E-field was varied from -8 kV/cm to 8 kV/cm; moreover, the magnetic coercive field was also found to be reduced from 15 Oe to 6 Oe, respectively.

The ϵ -E hysteretic loop with large remnant strain enables two stable strain states under zero E-field. More specifically, a positive E-field on the PZT led to a contraction in the length direction of the PZT slab through the inverse piezoelectric effect, which resulted in a compressive strain in FeGaB thin film that led to a reduced uniaxial anisotropy along the length direction of the PZT, while a negative E field would results in a tensile strain along the length of the PZT slab and an enhanced uniaxial anisotropy of the FeGaB film along the length direction of the heterostructure. The schematic of the heterostructure was presented in Fig. 2 inset to better understand the concept of the bistable magnetization switching. The strain in the width direction (X direction) was very small compared to the length direction (Y direction) since the length of the PZT slab is 5 times larger than its width ($\sigma_y \gg \sigma_x$). In that case, the total free energy that consists of the Zeeman energy, shape anisotropy, and magnetoelastic energy can be expressed as

$$F_{\text{free}} = -\vec{H} \cdot \vec{M}_s + 2\pi M_s^2 \cos^2 \theta - \frac{3}{2} \lambda (\sigma_y - \sigma_x) \sin^2 \theta \sin^2 \varphi,$$

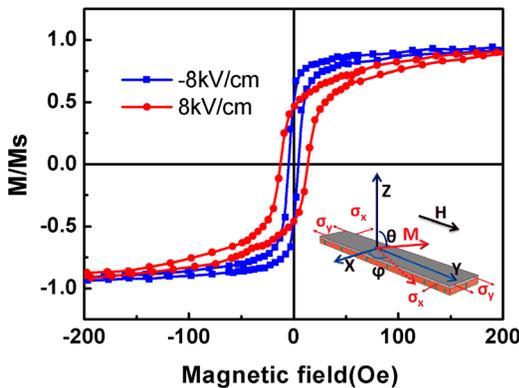


FIG. 2. Magnetic hysteresis loops of FeGaB/PZT heterostructure measured in length direction (Y direction) under application of E-field of -8 kV/cm and 8 kV/cm. Inset shows the schematic of FeGaB/PZT multiferroic heterostructure.

where θ , φ are the angles defined in Fig. 2 inset and λ is the in-plane magnetostriction coefficient of PZT ceramic. A compressive strain would be produced ($\sigma_y < 0$) when a positive E-field (i.e., at 8 kV/cm) was applied on the PZT slab; the magnetic easy axis would be forced to rotate toward the x direction by minimizing the free energy. A large energy barrier was formed due to the remnant strain difference leading to the bistable magnetization states.

Fig. 3 demonstrates the magnetization ratio M/M_s as a function of E-field with the amplitude varied within $-8 \sim 8$ kV/cm and $-16 \sim 16$ kV/cm. The applied E-field was generated by the sinusoidal voltage with a frequency of 0.04 Hz, and the magnetization was measured under a bias field of 3 Oe. The M-E curve was observed with the butterfly shape when E-field was larger than E_c (i.e., at 16 kV/cm); while a loop-like M-E curve was observed when the amplitude of E-field was 8 kV/cm, close to E_c . In the loop-like M-E curve, the magnetization ratio was varied from 71% to 56% with the change of E-field from -8 kV/cm to 8 kV/cm, leading to bistable magnetization that was consistent with the magnetic hysteresis loop in Fig. 2.

The electric field leads to a large hysteresis in the $\epsilon \sim E$ loop and bistable magnetization states at $E < E_c$. In comparison, further increasing E-field to 16 kV/cm would completely switch the FE domain leading to the relaxation of strain in the interface and a mono state of magnetization. We can define the two magnetization states defined by different signs of E-field as “1” and “0” in order to understand the magnetization control process by continuous E-field. As mentioned above, a positive E-field would induce a reduced effective magnetic anisotropy along the length direction; this magnetic state is defined as “0,” which could be maintained when the E-field is decreased to zero. By further decreasing the applied E-field, the magnetization started to increase and was stabilized at -8 kV/cm which forms another magnetic state defined as “1.” State “1” remained unchanged when the E-field was increased from -8 kV/cm to zero as well. The whole process can be traced by following the arrow in Fig. 3.

Voltage impulse control of magnetization would lead to further reduced energy consumption compared to continuous voltage or electric field. The voltage impulse induced magnetization control in the FeGaB/PZT multiferroic heterostructures was also measured, and the stability of the two magnetic states was tested in open circuit rather than close-

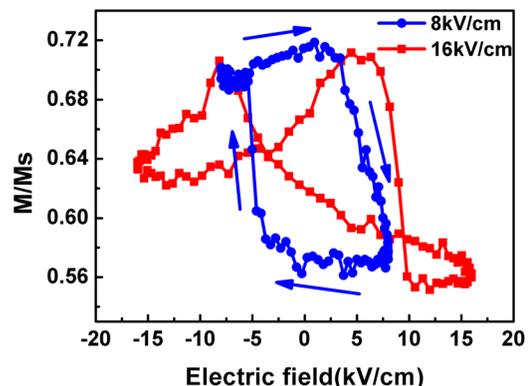


FIG. 3. Magnetization ratio versus E-field with a magnetic bias field (i.e., 3 Oe) under application of E-field of 16 kV/cm and 8 kV/cm.

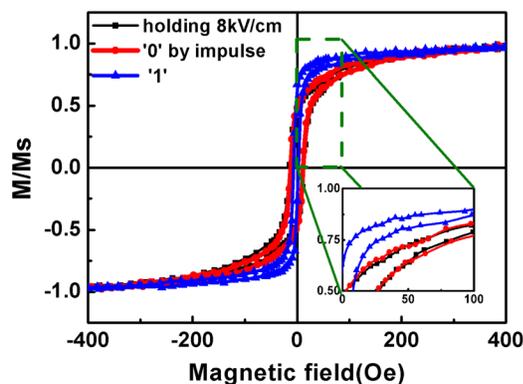


FIG. 4. Magnetic hysteresis loops of FeGaB/PZT heterostructure in state “1”; state “0” which switched by voltage impulse, and holding an E-field of cm. Inset shows the enlarged area.

loop at a fixed voltage. Fig. 4 shows the magnetic hysteresis loops presenting this process. First, magnetic state “1” was manipulated by increasing E-field from -8 kV/cm to zero. Then an 8 kV/cm voltage impulse with duration of about 100 ms was applied on the multiferroic heterostructure. The duration of the voltage impulse in our experiment is controlled by a relay and limited by the comparatively large scale of our samples. Magnetic hysteresis loop of state “0” was maintained after open circuit for 30 min, which was basically the same as another magnetic hysteresis loop measured by holding the E-field at 8 kV/cm. The slight change of the magnetization can be clearly seen in enlarged area (Fig. 4, inset). In other words, the nonvolatile reversible control of bistable magnetization can be achieved by using voltage impulses.

In order to simulate the real memory write process, the impulse induced bistable magnetization switching was measured continuously. Fig. 5 represents magnetization ratio under the same bias field (i.e., 3 Oe) together with the applied voltage impulse in time-domain measurement. A

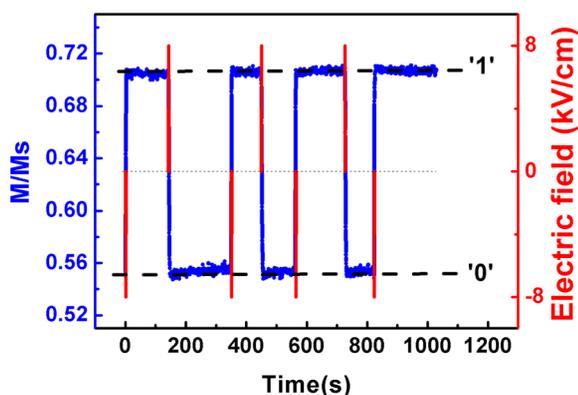


FIG. 5. Magnetization ratio of FeGaB/PZT heterostructure switched by voltage impulse measured in length direction with a magnetic bias field (i.e., 3 Oe) in time-domain measurement.

positive impulse remarkably decreases the magnetization ratio from 70% to 55% which means a switch from state “1” to state “0.” Similarly, a negative impulse then alters the state from “0” to “1.” The two distinct states keep stable in open circuit, which is consistent with Fig. 4 and demonstrates that nonvolatile voltage impulse control of magnetization can be realized in magnetoelectric multiferroic heterostructures.

In summary, we have demonstrated voltage-impulse controlled nonvolatile, reversible bistable magnetization switching in FeGaB/PZT heterostructures. Two distinct strain and magnetization states were induced by positive and negative sign of E-fields close to coercivity E_c , which remained stable in open circuit. The reversible and nonvolatile change of magnetization is due to the strain-induced ME coupling between FM and FE layers and the large remnant strain states in ϵ -E loop. Our results show the feasibility of strain-mediated magnetoelectric multiferroic heterostructures for application in voltage impulse writable MRAMs.

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