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Large reversible electric-voltage manipulation of magnetism in NiFe/BaTiO3 heterostructures at room temperature

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Abstract
NiFe/BaTiO3 heterostructures were grown on (0 0 1)-SrTiO3 single crystal substrates with SrRuO3 as a buffer layer via pulsed laser deposition and off-axis magnetron sputtering. The magneto-optical Kerr effect measurement on the heterostructures demonstrated that the magnetism of the NiFe thin film can be reversibly manipulated by switching the electric voltage applied on the BaTiO3 layer, with a large change in the magnetic coercive field and saturation magnetization of the NiFe thin films. An analogous on–off switch of magnetism in the NiFe thin films was demonstrated and a novel way for Curie point writing based on the heterostructure was also proposed. Our results may provide a useful way for manipulating magnetism of bits in high-density information storage with better thermal performance and reduced power consumption.

(Some figures in this article are in colour only in the electronic version)

Multiferroic materials with coexistence of both electric and magnetic orderings have attracted ever-increasing interest due to their multi-functionality, which provides significant potential for the next-generation multi-functional devices. Artificially structured multiferroic magnetoelectric (ME) composites which combine magnetic and ferroelectric systems have been found to exhibit a large ME response at room temperature and at low bias field [1, 2]. More recently, ME nanocomposite thin films of ferroelectric and magnetic substances, such as combinations of ferroelectric oxides (e.g. BaTiO3 and Pb(Zr,Ti)O3) with ferrites (e.g. NiFe2O4, CoFe2O4 and Fe3O4) [3–5], manganates (e.g. La0.67Sr0.33MnO3) [6] or magnetic metals (e.g. Fe) [7], have become new routes to such multiferroic ME composites. The ME coupling effect in these ME composites includes two aspects, i.e. magnetic field control of electric polarization and electric-field control of magnetization.

Electric-field control of magnetic behaviour can be realized by the ME coupling effect in multiferroic ME composite systems, either through the strain-induced ME effect across the interface [7, 8], or bias exchange in multiferroic BiFeO3-based heterostructures [9], or charge-driven ME effect in multiferroic ME heterostructures containing ultrathin ferromagnetic films [10–12]. Such an electric-field control of magnetism has attracted tremendous attention due to its potential applications in the design of next-generation spintronic devices such as high-density electric-field controlled magnetic random access memories.
(MRAMs). By modulating the carrier concentration in ferromagnetic semiconductors via applying external electric fields, Ohno and co-workers [13] demonstrated electric-field control of magnetic phase transition and coercivity in several systems, such as Mn-doped GaAs and InAs. For example, in a metal–insulator–semiconductor field-effect transistor structure, application of an external electric field to the thin (In,Mn)As channel layer has been shown to modify the ferromagnetic transition temperature and the coercive field of the channel layer. On this basis, Ohno demonstrated an electrically assisted magnetization reversal without changing the applied magnetic fields or temperature which showed the possibility of a new scheme for Curie point writing. The so-called Curie point writing means that magnetization reversal is assisted by elevating the film temperature locally above the Curie temperature or any suitable transformation temperature [14], typically achieved by heating the region of interest using laser-light irradiation in both cases of magneto-optical memory disks and ultrahigh-density magnetic recording [15]. However, this new scheme process was carried out at a very low temperature of about 32 K, which limits its potential application. In this work, we report an alternative way for such Curie point writing at room temperature in NiFe/BaTiO3 (BTO) multiferroic ME composite films grown on (001)-SrTiO3 (STO) substrates. Our results demonstrate that the magnetism of the ferromagnetic NiFe thin film can be manipulated by switching the electric voltage applied on the BaTiO3 layer. In this NiFe/BTO heterostructure, an analogous on–off switch of magnetism in the NiFe layer has also been demonstrated by means of an applied electric field.

In our study, the BTO thin films were first prepared on (001)-oriented SrTiO3 single crystal substrates with SrRuO3 (SRO) as a buffer layer using pulsed laser deposition (KrF laser, λ = 248 nm). The sputtering targets were SRO and BTO single phase ceramic discs fabricated by conventional solid state reaction and sintering. A laser density of about 1.5 J cm−2 and a flowing oxygen pressure of 13 Pa were maintained during deposition. The substrate temperature was kept at 600 °C and 800 °C for SRO and BTO deposition, respectively. After the deposition, the films were cooled to room temperature in 1000 Pa ambient oxygen. The thicknesses of the SRO and BTO are both about 100 nm. Finally, a continuous NiFe thin film was grown on the BTO/SRO/STO heterostructure (figure 1) with a rectangular mask (2 × 3 nm2), using off-axis magnetron sputtering. Permalloy Ni80Fe20 was chosen as the sputtering target. The thickness of the NiFe thin layer was kept at 10 nm.

To investigate the epitaxial quality of the heterostructure, x-ray diffraction (XRD) was carried out using a Rigaku D2500 diffractometer with Cu Kα. As seen from a typical θ–2θ scan shown in figure 1(b), no additional or intermediate phase peaks apart from BTO and SRO appeared, and there were only the (00l) peaks in the diffraction patterns as marked in the figure, indicating epitaxially c-axis oriented SRO and BTO layers, normal to the substrate surface. The NiFe layer was not detected in the XRD patterns due to its very low diffraction intensity excited by its very small thickness. The morphology of the heterostructure, as shown in figure 2, was characterized using an atomic force microscope (AFM, PSIA XEI100), which clearly demonstrates a uniform surface morphology with fine grains. The root-mean-square (RMS) roughnesses of the surface with or without the NiFe film are 3.3 and 2.4 nm, respectively. The surface grain size of BTO is about 200 nm. The comparison in figure 2 shows that the thin NiFe layer just copies the morphology of the BTO layer by simply covering.

The polarization–electric voltage (P–E) measurement was carried out on a TF analyzer2000 (aixACCT Co.) with the top NiFe layer and the SRO layer as the top and the bottom electrodes, respectively. The NiFe/BTO/SRO/STO heterostructures exhibit an evident ferroelectric hysteresis loop (figure 3). The saturation polarization Ps and remanent polarization Pr are about 40 μC cm−2 and 21 μC cm−2, respectively, which are larger than those (i.e. Ps = 30 μC cm−2 and Pr = 12 μC cm−2) for the pure BTO film deposited on STO without a SRO buffer layer [3]. The electric coercive voltage for the BTO film on SRO/STO is about 2.8 V, i.e. 28 MV m−1, which is also remarkably smaller than that (i.e. 47 MV m−1) for the BTO film directly deposited on a STO substrate [3]. It is reasonable according to Tagantsev and Stolichnov’s model [16] that SRO with a better conductivity decreases the thickness of the interfacial dielectric layer between the ferroelectric layer and the electrode layer, resulting in a smaller coercive field.

To detect the local magnetism variations of the NiFe layer, magneto-optical Kerr effect (MOKE) magnetometry (Durham
Figure 2. AFM topography images of (a) the BTO film without NiFe layer deposited and (b) the surface of NiFe film on the BTO.

Figure 3. Ferroelectric hysteresis loop of the NiFe/BTO/SRO/STO heterostructure at 2000 Hz.

NanoMOKE2 was utilized. The magnetic field was applied in the plane of the heterostructure (figure 1(a)), the same direction as that of the remanent magnetization. For characterizing the magnetization values, a physical property measurement system (PPMS-9T) was also used to calibrate the MOKE signal. Typically, the amplitude of the measured Kerr signal is proportional to the magnetization. As seen in figure 4(a), the squared magnetic hysteresis loop measured by MOKE at zero external electric field shows the well-defined ferromagnetic properties of the NiFe layer in the heterostructures and the saturation magnetization $M_s$ is about 120 emu cm$^{-3}$.

For applying the voltage on the heterostructure, the film samples were electrically connected with silver wires bonded on the top NiFe layer and the bottom SRO layer (figure 1(a)). The laser beam was focused on the bare surface of the NiFe film. The direct current voltage is applied using a voltage sourcemeter (Keithley 2410) from 0 up to 20 V in ascending steps, then descending to $-20$ V, and finally ascending back to 0 along the arrows marked in figure 4(b). The MOKE measurements focused on different regions in the samples presented similar behaviour. All the measurements were performed at room temperature. Figure 4(a) shows the variations in the longitudinal Kerr loops with the voltage from 0 to 20 V. The Kerr loops remain square in shape but shrink with increasing electric voltage bias until the bias reaches 20 V. At 20 V, the Kerr loop displays a hard magnetization state and the coercive field finally seems to vanish, where the easy axis of magnetization is away from the in-plane direction. On reversing the bias voltage to a negative one, similar results were observed. The large voltage-induced magnetization variations in figures 4(a) and (b) clearly indicate that the bias voltage affects the distribution of domain orientations in the magnetic layer, i.e. a reorientation of the magnetization directions, which is a complex combination of domain rotation and domain wall movement. However, the details of the underlying mechanism, whether it is due to the strain-induced ME effect across the interface [7, 8, 17] or the charge-driven ME effect [10, 11], or both, remain to be investigated.

The coercive field $H_c$ at zero voltage bias is about 12 Oe and decreases with an increase in the bias voltage no matter whether the voltage is positive or negative, i.e. $H_c$ is modified by a factor of 48, from 12 Oe at zero bias to about 0.25 Oe at $\pm20$ V. As seen from figure 4(b), the electric voltage manipulation of $H_c$ is reversible and it displays nearly no hysteretic behaviour. The changes in the coercive field $H_c$ in the positive and negative voltage range illustrate good symmetry with nearly no difference.

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Obviously, the applied electric voltage bias also causes a significant change in the saturation magnetization amplitude $M_s$. $M_s$ also decreases with an increase in the bias voltage no matter whether the voltage is positive or negative. When applying a bias voltage of $\pm20$ V, the $M_s$ decreases to about 40 emu cm$^{-3}$ which is only about one-third of 120 emu cm$^{-3}$ at zero electric bias. Since the magnitude of $M_s$ is a function of the external electric voltage and a high or low $M_s$ appears with a change in the electric voltage, a reversible on-off switch of magnetism in the NiFe layer can be achieved. As shown in figure 4(c), at zero electric bias applied on BTO ($t = 0$), the NiFe layer is in an ‘on’ state with a high value of $M_s$, but the NiFe layer becomes in an ‘off’ state with a low value of $M_s$ as an electric voltage (e.g. 16 V) is applied ($t = 5$ min). When the electric voltage is subsequently switched back to 0, the NiFe layer returns to the ‘on’ state again ($t = 10$ min). The magnetic state of the NiFe layer can still be reversibly and reproducibly controlled through an applied electric voltage. When a dc magnetic field bias of about 16 Oe is applied on the sample, there is no change in the Kerr signal of the sample and the state of the NiFe layer...
can still be reversibly changed by the electric bias as discussed above. The interesting variations of the coercive force $H_c$ and magnetization amplitude controlled by the electric voltage might be due to the electric voltage modulated total anisotropy energy of the NiFe layer and the spin reorientation transition away from the in-plane magnetization direction as mentioned above.

As the magnitude of $H_c$ is changed with the external electric voltage, a similar magnetization-reversal process can also be electrically assisted, as Ohno and co-workers [15] demonstrated at a very low temperature. First, we apply a sufficiently large positive magnetic field to get the magnetization of the NiFe layer saturated under 0 V and then decrease the magnetic field to a small magnetic field, e.g. $-6$ Oe, as marked as the initial state by point A in figure 4(d). Because the magnitude of $H_0 = -6$ Oe is smaller than the coercive field $H_c = -12$ Oe at zero electric voltage, this state A is maintained until an electric voltage of 18 V is applied. In response to this applied voltage, the value of $M_s$ changes from point A to point B, illustrating that the magnetization reversal is triggered by the electric voltage. Once the magnetization is reversed, the negative value of $M_s$ can be maintained and shows a variation only when the electric voltage is switched between 18 and 0 V, as denoted by points C and D. This electrically assisted magnetization reversal without changing the applied electric fields or temperature offers a novel way for Curie point writing where the magnetization reversal is assisted by heating the local region of interest using laser-light irradiation. It is worth noting that the Curie point writing demonstrated by Ohno and co-workers [15] was carried out at a very low temperature of about 32 K. Comparably, this process can be executed at room temperature in the NiFe/BTO heterostructure.

In summary, NiFe (10 nm)/BaTiO$_3$ (100 nm)/SrRuO$_3$ (100 nm) heterostructures have been grown on (0 0 1)-SrTiO$_3$ substrates. A large reversible electric-voltage manipulation of the coercive force $H_c$ and saturation magnetization $M_s$ of the NiFe layer can be achieved at room temperature, demonstrating an analogous on–off switch of magnetism of the NiFe layer and a novel way for Curie point writing at room temperature. Such multiferroic ME heterostructures may offer an alternative to heat-assisted magnetic recording for future high-density information storage with better thermal performance and lower power consumption.

Acknowledgments

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