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## Electric field control of magnetic properties of Nd<sub>2</sub>Fe<sub>14</sub>B thin films grown onto PMN-PT substrates

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Nd<sub>2</sub>Fe<sub>14</sub>B films with easy magnetization axis perpendicular to the film plane were grown onto Ta-buffered piezoelectric (011)-0.7Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.3PbTiO<sub>3</sub> (PMN-PT) substrates using a magnetron sputtering technique. The film retains good permanent magnetic properties, with coercivities up to 1.4 T and spin reorientation near 150 K. Our investigations demonstrated that the converse piezoelectric effect generated by an electric field across the PMN-PT substrate can rotate the angle of spin reorientation of the Nd<sub>2</sub>Fe<sub>14</sub>B films, resulting in a considerable manipulation of magnetization by the electric field at temperatures below 150 K. The ratio of magnetization change reaches 30% in a magnetic field of 2 T when an electric field of 6 kV/cm is applied at 30 K. With increasing temperature, the electric field-induced tunability of magnetization decreases. These observations of permanent magnetic properties and electrically controlled magnetization effects make the Nd<sub>2</sub>Fe<sub>14</sub>B/PMN-PT heterostructure attractive for exploring multifunctional magnetic/electric devices. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4973755>]

Nd<sub>2</sub>Fe<sub>14</sub>B is one of the most mature and widely used permanent magnetic materials. Continuous efforts to investigate the intrinsic properties and promote performance in applications as permanent magnetic materials have been made.<sup>1-4</sup> The fabrication of Nd<sub>2</sub>Fe<sub>14</sub>B thin films has also been the focus of research, noting that permanent magnetic films with specialized magnetic performance are required in key applications of microstructural devices.<sup>5-13</sup> This applies in various fields, such as magnetic microelectromechanical devices, magnetic sensors, and magnetic storage. For this purpose, it is necessary to make permanent magnetic thin films. Research on thin films of Nd<sub>2</sub>Fe<sub>14</sub>B also aids in understanding the intrinsic nature of the materials.<sup>8-10</sup> For maximizing the performance of thin films with high remanence and coercivity, fabricating Nd<sub>2</sub>Fe<sub>14</sub>B thin films with anisotropy is essential. Many efforts have been dedicated to the growth of Nd<sub>2</sub>Fe<sub>14</sub>B films on a number of substrates with various buffer layers,<sup>7-12</sup> such as Ta-buffered Al<sub>2</sub>O<sub>3</sub> (0001)<sup>7</sup> and Mo-buffered MgO(100),<sup>9</sup> and good performance of permanent magnetic properties have been observed. However, studies of thin films, particularly on ferroelectric/piezoelectric substrates, for the purpose of electrically controlled magnetization are rare, and the magnetoelectric (M-E) effect in Nd<sub>2</sub>Fe<sub>14</sub>B thin films has not been studied.

The converse magnetoelectric (M-E) effect, i.e., the control of magnetism via an electric field, has attracted much attention because of its potential application in the area of information storage. Among many studies, an artificial structure combining piezoelectric and ferromagnetic

materials is one of the main foci.<sup>15-23</sup> The electric field-induced manipulation of magnetization has been studied in ferromagnetic (ferrimagnetic)/piezoelectric heterostructures, involving a number of heterostructures composed of different ferromagnetic or ferrimagnetic films, such as CoFe<sub>2</sub>O<sub>4</sub>/PMN-PT,<sup>16</sup> La<sub>2/3</sub>Sr<sub>1/3</sub>MnO<sub>3</sub>/PMN-PT,<sup>17</sup> Pr<sub>0.7</sub>Sr<sub>0.3</sub>MnO<sub>3</sub>/PMN-PT,<sup>18</sup> Co<sub>40</sub>Fe<sub>40</sub>B<sub>20</sub>/PMN-PT,<sup>19</sup> Ni/PMN-PT,<sup>20</sup> and Fe/BaTiO<sub>3</sub>(BTO),<sup>22</sup> where the piezoelectric PMN-PT and BTO crystals produce a sizable in-plane strain under an electric field. However, we noticed that these studies primarily focused on soft ferromagnetic or ferrimagnetic materials, and no one has ever studied a possible change in magnetization for hard magnetic materials, which may help in the exploitation of the converse M-E effect and interest more researchers in the properties of hard magnetic materials.

Here, we report the growth of Nd<sub>2</sub>Fe<sub>14</sub>B films with thicknesses of 100 nm and 200 nm, with the easy magnetization axis perpendicular to the film plane, on Ta-buffered (011)-oriented 0.7Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.3PbTiO<sub>3</sub> (PMN-PT) substrates. The PMN-PT substrate was chosen because of its excellent converse piezoelectric effect. The films show good permanent magnetic properties and spin reorientation at approximately 150 K. The converse piezoelectric effect generated by an electric field across the PMN-PT substrate can rotate the spin reorientation angle of the Nd<sub>2</sub>Fe<sub>14</sub>B film, resulting in a considerable manipulation of magnetization by the electric field. The simultaneous appearance of electronically controlled magnetization and hard magnetic properties can aid in exploring multifunctional microelectronic devices and promote the comprehensive application of Nd<sub>2</sub>Fe<sub>14</sub>B films.

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$\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films were prepared in an ultra-high vacuum chamber with base pressure less than  $10^{-6}$  Pa using a magnetron sputtering technique. A Ta buffer layer with a thickness of 20 nm was sputtered on a (011)-oriented PMN-PT substrate prior to the sputtering of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films to avoid diffusion of the substrate material to the film and promote the textured or even epitaxial growth of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films. After the sputtering of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films, a capping layer of 100 nm of Ta was sputtered to prevent oxidation of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films. During sputtering, Ar pressure was kept at 0.5 Pa, and a DC sputtering power of 120 W was used. The substrate temperature was kept at  $350^\circ\text{C}$  for the growth of both the Ta buffer and capping layer, while  $550^\circ\text{C}$  was used for the growth of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films. The thickness of the films was controlled by the sputtering time. The target was manufactured using the induction-melting technique, and the composition of  $\text{Nd}_{16}\text{Fe}_{73}\text{B}_{11}$  with rich Nd was chosen for the formation of the Nd-rich phase to realize good permanent magnetic properties,<sup>13</sup> and compensate for the possible loss of Nd during the deposition process. After the deposition, the films were sealed in an evacuated quartz tube with a vacuum less than  $10^{-4}$  Pa and then processed by rapid thermal annealing at  $700^\circ\text{C}$  for 1–3 min, followed by cooling down to room temperature in air.

The magnetic properties were measured using Quantum Design superconducting quantum interference devices (SQUID VSM). The magnetization under an electric field was measured in situ, where an electrometer (6517B Keithly) provided the electric field. X-ray diffraction (XRD) using Cu-K $\alpha$  radiation was carried out to identify the structure and orientations of the films.

The XRD patterns of the heterostructures with 100-nm and 200-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films are shown in Figs. 1(a) and 1(b), respectively. Except for the peaks from the (011)-PMN-PT substrate and the (011)-Ta layer, only the (00 $l$ ) peaks of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  appear, without any other impure phases, particularly for the 200-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , demonstrating the excellent c-axis orientation of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films. For the 100-nm

$\text{Nd}_2\text{Fe}_{14}\text{B}$ , the (00 $l$ ) peaks become weak because of the (011)-Ta capping layer, but the (006) and (008) peaks can still be clearly identified (Fig. 1(b)). A Ta layer as the buffer is most widely used to fabricate textured or even epitaxial  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films. Ta not only has the effect of isolation from oxidation, but it also matches well with the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  lattice. Bulk  $\text{Nd}_2\text{Fe}_{14}\text{B}$  shows a tetragonal structure with lattice parameters  $a = 8.8 \text{ \AA}$  and  $c = 12.20 \text{ \AA}$ , while Ta has a cubic structure with a lattice parameter of  $3.303 \text{ \AA}$ , whose face diagonals, i.e., the (011) plane of Ta, matches well with the basal plane of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ ; thus, textured or even the epitaxial growth of (001) $\text{Nd}_2\text{Fe}_{14}\text{B}$ |(011)Ta is strongly favored. Because of the excellent c-axis orientation of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films, the obtained heterostructures show good permanent magnetic properties. Figs. 1(c) and 1(d) present magnetic hysteric loops (M-H) along the in-plane [100] and out-of-plane directions for the heterostructures with 100-nm and 200-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films, respectively. The room temperature out-of-plane coercivities reach 1.4 T and 0.5 T for 200-nm and 100-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films, respectively, and increase significantly with decreasing temperature. As a typical display, Fig. 1(c) also presents the out-of-plane M-H loops of the heterostructure with a 100-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  film measured at 30 K, where the coercivity increases to 1.3 T. The different coercivities in films with different thicknesses may come from the specific grain boundaries and intergranular phases, which provide pinning sites for domain-wall motion. The imperfect squareness of the loops is probably related to the oxidation of the films during the fabrication process.

We also measured the dependence of magnetization on temperature (M-T curves) in the remnant state of the film with 100-Oe magnetic fields applied along both the in-plane and out-of-plane directions. Bulk-like spin reorientation occurs in 100-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films. As shown in Fig. 2, there is an obvious upturn in the magnetization for the out-of-plane M-T curve and a downturn in the magnetization for the in-plane M-T curve. This result indicates the occurrence of spin reorientation near  $T_{\text{SR}} \sim 150 \text{ K}$ ,<sup>8–10</sup> which means the films change from the easy axis to the easy cone structure at  $T_{\text{SR}}$ . The magnetization starts canting from the [001] direction (c axis) of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films toward the in-plane direction,

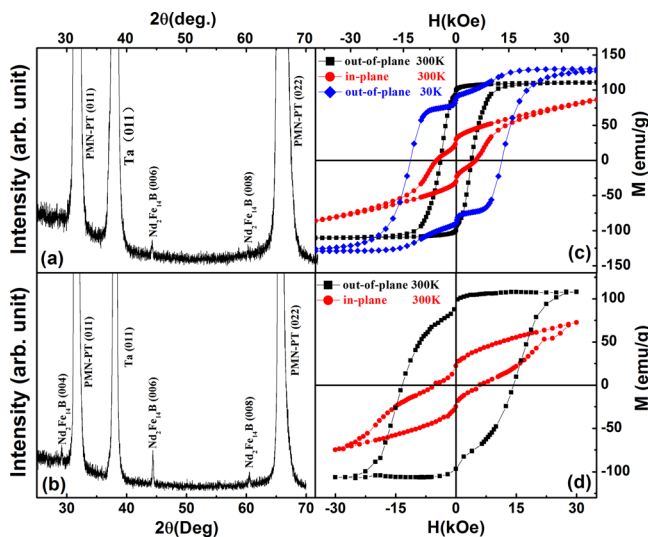


FIG. 1. XRD patterns of the heterostructures with (a) 100-nm and (b) 200-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films, and magnetic hysteric loops (M-H) along the in-plane [100] and out-of-plane directions for the heterostructures with (c) 100-nm and (d) 200-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films.

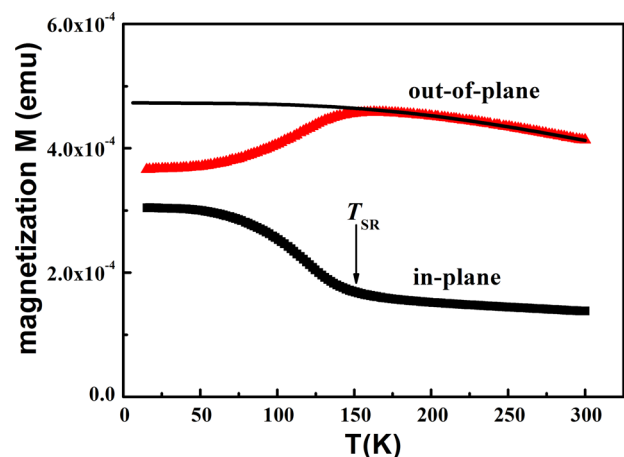


FIG. 2. Magnetization dependence on temperature of the in-plane [100] and out-of-plane directions for the heterostructure with 100-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films measured in the remnant state with a 100-Oe magnetic field applied.

and the tilt angle monotonously increases with decreasing temperature. The calculated maximal canting angle is  $\sim 39^\circ$  based on the difference between the simulated total magnetization and the out-of-plane magnetization (Fig. 2), which is nearly the same as that of a previously reported film ( $\sim 40^\circ$ ) grown on Mo-buffered (100)-MgO,<sup>9</sup> but largely exceeds the single-crystal case ( $\sim 30^\circ$ ) at 4.2 K.<sup>14</sup> Compared with the single crystal, strain may be introduced from the substrate to the textured or epitaxial films grown on it. Although the exact reason for the enlarged spin reorientation angle is not clear, possible strain from the substrate cannot be excluded for the textured or epitaxial films grown on (011)-PMN-PT or (001)-MgO. In bulk  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , the spin reorientation phenomenon has been previously studied. It was reported that bulk  $\text{Nd}_2\text{Fe}_{14}\text{B}$  shows a spin reorientation near 135 K, and the spin reorientation temperature differs with different particle sizes in polycrystalline  $\text{Nd}_2\text{Fe}_{14}\text{B}$ .<sup>14</sup> This phenomenon has also been observed in  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin films.<sup>8,9</sup> The occurrence of spin reorientation is evidence of the good crystallization for the present  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films as thin as 100 nm. Moreover, the growth of such a thin film on PMN-PT provides a powerful platform for studying the M-E effect through strain transfer enforced by an electric field, which has never been demonstrated before for  $\text{Nd}_2\text{Fe}_{14}\text{B}$  permanent magnetic films.

To study the influence of an electric field on the magnetic properties of the films, the dependence of magnetization on the electric field (M-E curves) at constant magnetic fields and the hysteretic loops under a constant electric field were measured. The upper image in Fig. 3 shows a sketch of the heterostructures. As can be seen, a Au layer acts as the

bottom electrode, and the Ta capping layer acts as the top electrode, so that an electric field can be applied across the PMN-PT substrate. Because of the excellent piezoelectric effect, applying an electric field along the out-of-plane [011] direction can induce a sizable anisotropic in-plane strain in the PMN-PT substrate. We measured the room-temperature strain along both the in-plane [100] and [01-1] directions under the electric field,<sup>15</sup> and found that the [100] direction undergoes a compressive strain as large as  $\sim -0.31\%$ , while the [01-1] direction undergoes a very small tensile strain of 0.018% when an electric field of 6 kV/cm is applied along the out-of-plane [011] direction of the PMN-PT substrates; we therefore chose the in-plane [100] direction to study the M-E effect.

The temperature we chose to measure the M-E effect varied from 30 K to 300 K. The samples investigated here were first brought to the target temperature, at which the M-E curves of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films were measured under different constant magnetic fields applied along the in-plane [100] direction of the (011)-PMN-PT substrate. The  $[M(E)-M(0)]/M(0)$  versus E curves at various temperatures and magnetic fields are displayed in Fig. 3. The electric field exhibits a great influence on the magnetization of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  film along the [100] direction of the PMN-PT substrate. The magnetization at a constant magnetic field notably decreases with the electric field applied across the substrate, and the converse M-E effect becomes more noticeable at higher magnetic fields. When a bipolar electric field of 6 kV/cm was applied, there was a 30% decrease in magnetization at 30 K with a magnetic field of 2 T applied along the [100] direction of the PMN-PT (011) substrate. The most interesting

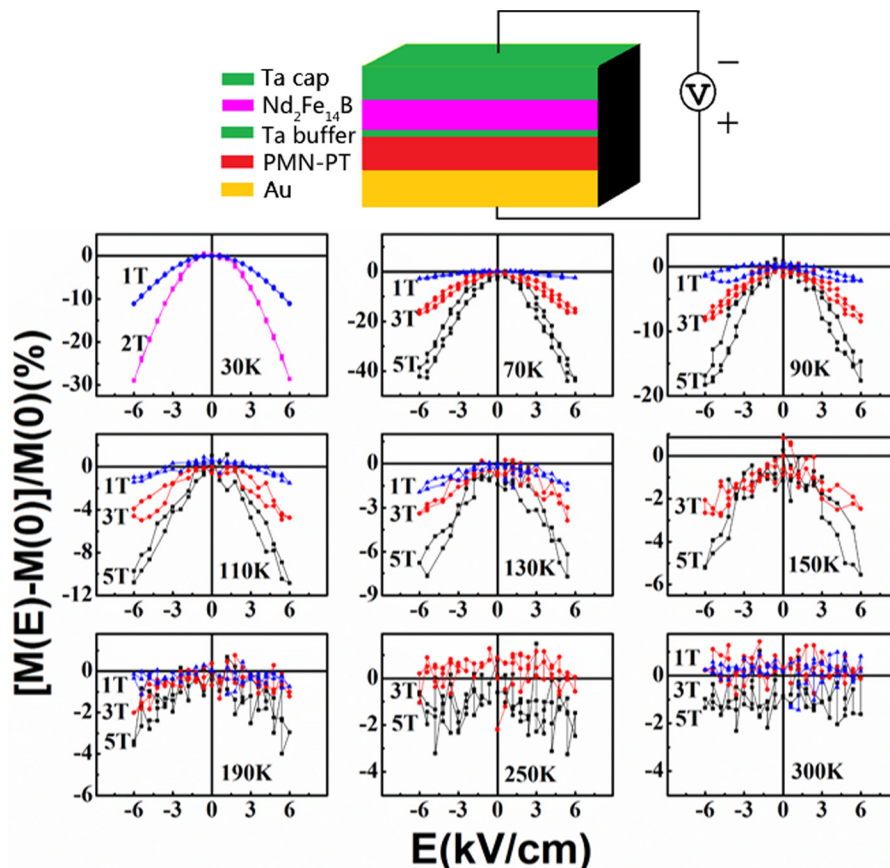


FIG. 3. Magnetization of the heterostructure with a 100-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin film as a function of electric field measured with different magnetic fields along the in-plane [100] direction. For comparison, the measurements were carried out at various temperatures from 30 K up to 300 K. The upper image demonstrates a sketch of the heterostructures during measurement.



phenomenon is that the film only shows a notable change of magnetization below the temperature of 150 K, which happens to be the spin reorientation temperature of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  film here, and the M-E effect is enhanced with decreasing temperature. The magnetic hysteresis loops at different temperatures were also studied under voltage application in the sequence of 0 V, 6 kV/cm, and 0 V (after the removal of the electric field). As a typical display, Figs. 4(a) and 4(b) present the magnetic loops measured at 30 K and 300 K, respectively. Consistent with the M-E curves, the regulation of magnetization by the electric field is remarkable and critically dependent on the magnetic field at temperatures well below 150 K. For the case at 30 K (Fig. 4(a)), visible regulation by the electric field starts from 0.4 T and then shows a monotonous increase with increasing magnetic field. After the removal of the electric field, the hysteretic loops return to their initial states before the electric field was applied. In contrast, at temperatures well above 150 K, such as 300 K (Fig. 4(b)), the magnetization is insensitive to the electric field, even when a high magnetic field of 3 T is applied.

The strain-induced M-E effect for the combination of a ferromagnetic metal with piezoelectric materials has been widely studied.<sup>19–22</sup> Investigations of the manipulation of the magnetization by an electric field in a Ni/BaTiO<sub>3</sub> (BTO) hybrid structure determined that the tensile strain induced by the BTO substrate causes a large uniaxial magnetic anisotropy because of the inverse magnetostriction, changing the measured projection of magnetization to the direction of the magnetic field. For those parts of the Ni thin film, the magnetization is clamped to the *a*-domains of the BTO substrate.<sup>21</sup> The modulation of remnant magnetization for multiferroic  $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}/(011)\text{-PMN-PT}$  heterostructures was also studied. An anisotropic strain on the film changes the in-plane easy magnetization axis. The easy axis rotates approximately 22° from the easy axis [100] to the [01-1] direction, inducing considerable changes in the remnant magnetization.<sup>17</sup> Moreover, in a study of electric tuning of magnetism in  $\text{Fe}_3\text{O}_4/(011)\text{-PZN-PT}$  multiferroic heterostructures,<sup>23</sup> the dependence of magnetic anisotropy on the electric field was investigated both theoretically and experimentally. The theoretical prediction, which introduces the magnetoelastic energy and an effective anisotropy field, matches well with the experimental results, and the  $\text{Fe}_3\text{O}_4$  film shows a large change in the squareness ratio of 44% along the in-plane [100] direction of the substrate.

However, the situation is quite different for the present  $\text{Nd}_2\text{Fe}_{14}\text{B}/(011)\text{-PMN-PT}$  heterostructures. At temperatures

higher than the spin reorientation temperature ( $\sim 150$  K) of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  films, particularly around room temperature, we did not detect any change in coercivity and remnant magnetization when an electric field was applied. This result indicates that the easy axis along the out-of-plane *c* direction is robust enough that a strain as large as  $\sim -0.31\%$  cannot shake it. The large coercivities in  $\text{Nd}_2\text{Fe}_{14}\text{B}$  primarily originate from the grain boundaries and intergranular phases, which provide pinning sites for domain-wall motion. The insensitivity of coercivity to an electric field indicates that the strain induced by the electric field has little effect on the grain boundaries and pinning sites, and the strain-induced change in microstructure is not enough to change the easy axis at high temperature or even around room temperature. In this situation, the magnetization is totally aligned with the out-of-plane direction, and the anisotropic energy introduced by the piezoelectric strain can be neglected, as there is no projection of the magnetization in the in-plane direction, so we cannot observe any obvious magnetization change in this case.

As the temperature approaches 150 K, temperature-induced spin reorientation occurs. The magnetic structure of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin film begins to change from the easy-axis to an easy cone. In other words, the magnetization gradually reorients from the out-of-plane to the in-plane direction with decreasing temperature, starting from a point near 150 K. The maximal tilt angle,  $\sim 39^\circ$ , which is larger than that of bulk  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , is probably due to the strain induced by the substrate. In this situation, the in-plane magnetization will be the projection of the easy cone magnetization, and strain-induced magnetic anisotropy will thus influence the projection of the magnetization along the in-plane [100] direction of the heterostructure. However, a significant M-E effect only appears under the application of a magnetic field, and it increases with an increasing magnetic field. This phenomenon can be understood by considering that the application of a magnetic field along the in-plane [100] direction favors the rotation of the easy cone away from the *c* axis, resulting in the observed M-H loops shown in Fig. 4(a) (black curve, 0 kV/cm). When an electric field of 6 kV/cm is applied, the induced large compressive strain along the in-plane [100] direction may release the strain, which favors the rotation of the easy cone forward to the *c* axis, resulting in the reduction of the projection of the magnetization along the in-plane [100] direction, as shown in the M-H loops in Fig. 4(a) (red curve, 6 kV/cm) and the M-E curves shown in Fig. 3. With decreasing temperature, the converse M-E effect becomes more apparent because, as the opening angle between the

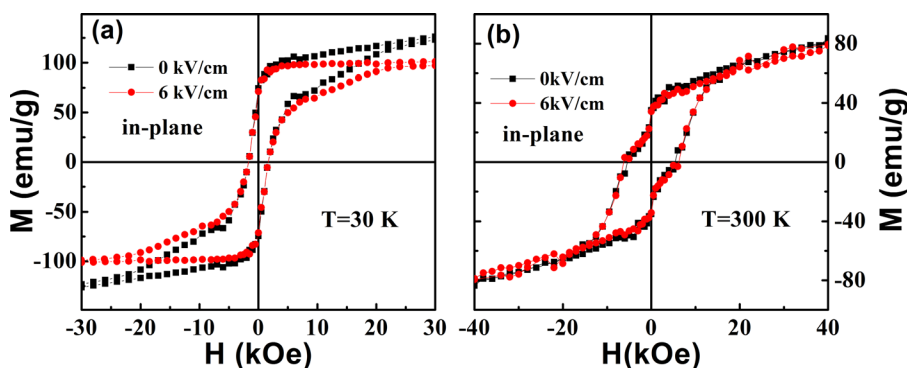


FIG. 4. Magnetization loops measured as a function of the magnetic field along the in-plane [100] direction for the heterostructure with a 100-nm  $\text{Nd}_2\text{Fe}_{14}\text{B}$  thin film at (a) 30 K and (b) 300 K under applied electric fields of 0 kV/cm and 6 kV/cm.

magnetization and the perpendicular direction increases, the effect is amplified.

By introducing a Ta buffer layer, we have fabricated *c* axis-oriented Nd<sub>2</sub>Fe<sub>14</sub>B thin films over (011)-PMN-PT piezoelectric substrates. Good permanent magnetic properties and a notable magnetoelectric effect were simultaneously observed in the heterostructures. The coercivity can be as large as 1.4 T near room temperature, and it clearly increases with decreasing temperature. Moreover, spin reorientation was demonstrated near 150 K, below which the strain induced by an electric field can rotate the easy-axis with the assistance of the magnetic field, resulting in a considerable modulation of the magnetization along the in-plane [100] direction of the heterostructure. The modulation ratio increases noticeably with decreasing temperature because of the opening up of the spin reorientation angle, which shows a completely reversible behavior after the electric field was withdrawn. A 30% change in magnetization occurred when an electric field of 6 kV/cm was applied at 30 K. These observations can assist in the exploration of multifunctional microelectronic devices and promote the comprehensive application of Nd<sub>2</sub>Fe<sub>14</sub>B films.

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