Nonvolatile ferroelectric domain wall memory integrated on silicon

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Abstract

Ferroelectric domain wall memories have been proposed as a promising candidate for nonvolatile memories, given their intriguing advantages including low energy consumption and high-density integration. Perovskite oxides possess superior ferroelectric properties but perovskite-based domain wall memory integrated on silicon has rarely been reported due to the technical challenges in the sample preparation. Here, we demonstrate a domain wall memory prototype utilizing freestanding BaTiO_3 membranes transferred onto silicon. While as-grown BTO films on (001) SrTiO_3 substrate are purely c -axis polarized, we find they exhibit distinct in-plane multidomain structures after released from the substrate and integrated onto silicon due to the collective effects from depolarizing field and strain relaxation. Based on the strong in-plane ferroelectricity, conductive domain walls with reading currents reaching 2 nA are observed and can be created artificially, highlighting the great potential of the integration of perovskite oxides with silicon for ferroelectric domain wall memories.

Introduction

Ferroelectric domain walls (DWs), the low-dimensional structures that separate differently polarized regions, have been shown to exhibit appealing properties in recent years^{1–5}, including metallic conduction^{6–11}, magnetoresistance¹² and photovoltaic behaviors^{13–15}. Conductive DWs, mainly appearing as head-to-head (H-H) or tail-to-tail (T-T) DWs^{6, 16–19}, have been hitherto detected in various types of materials, such as proper ferroelectric perovskites^{8, 16–18, 20}, non-perovskites^{10, 21, 22} and improper ferroelectrics^{11, 23, 24}. Due to the nanoscale space distribution of DWs and non-destructive switching of polarization states, ferroelectric DWs hold great potentials for the applications in high-density and low power nonvolatile memories^{16, 25, 26}. For practical industrial applications in complementary metal oxide semiconductor (CMOS)-based technology, however, it is highly desired to integrate these DW memories with silicon (Si). To our knowledge, such attempts have only been demonstrated in LiNbO_3 ²⁷ and $\alpha\text{-In}_2\text{Se}_3$ ²⁸ and no success has been achieved in ferroelectric perovskite oxides up to date although they exhibit excellent ferroelectric properties to facilitate the exploitation of DW memory.

Integrating perovskite oxides with Si has long been a challenging task since the high growth temperature and oxidant environment for the growth of high crystalline quality perovskite oxides can easily oxidize the surface of Si wafer, hindering the subsequent high-quality integration of these two types of materials^{29–31}. Adopting special growth recipes, SrTiO_3 (STO) can be epitaxially grown on Si with high crystalline quality and showing ferroelectricity³². Using STO as a buffer layer, many ferroelectric oxides like BaTiO_3 (BTO), PbTiO_3 have also been integrated with Si^{33, 34}. Nonetheless, these ferroelectric oxides are mostly under compressive strain and exhibit out-of-plane ferroelectricity, while H-H or T-T conductive DWs normally require in-plane polarization components. Recently, high quality freestanding ferroelectric perovskite oxides have been achieved using water-soluble $\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) sacrificial layer^{35, 36}, providing

a new route to integrate high crystalline quality functional oxides, such as BTO³⁷ and BiFeO₃ (BFO)³⁸, with Si and other technologically important semiconductors. Moreover, without the clamping effect imposed by the substrate, freestanding oxide membranes have exhibited great tunability in strain engineering and dimensionality^{37, 39–41}. By a combination of strain relaxation and depolarizing field^{42–44}, the in-plane ferroelectric domain and H-H or T-T DW structures can be managed and may allow the integration of conductive DW memories on Si.

Here, through BTO/SAO/STO heterostructures grown by molecular beam epitaxy (MBE), we realized the in-plane multidomain configuration in BTO/Si structure and thus the integration of conductive DWs in BTO on the Si substrates. The ferroelectric polarization rotation from out-of-plane *c*-axis to in-plane *a*-(*b*) axis in BTO membrane emerges as a combined effect of strain relaxation and depolarizing field during release from STO substrate. The nA-scale reading current and the systematic performance of ‘write and read’ process on our prototype signifies a promising step forward in achieving Si-based nonvolatile conductive DW memory.

Results

DW memory prototype fabrication. As-grown BTO films were synthesized on TiO₂-terminated (001) STO substrates after growing 6 u.c. SAO buffer layers by molecular beam epitaxy (see Methods), with the thicknesses ranging from 6 u.c. to 160 u.c. The reflection high energy electron diffraction (RHEED) oscillations and diffraction patterns are displayed in Supplementary Fig. 1a. High-quality single crystallinity and smooth step-and-terrace surfaces are both confirmed (Supplementary Fig. 1b). As schematically shown in Fig. 1, we fabricated the ferroelectric DW memory prototype by transferring freestanding BTO membranes onto Au-coated Si. The micrometer-size and well-preserved smooth surfaces of BTO membranes promote the subsequent fabrication.

90° polarization rotation during release. To guarantee that conductive DWs in BTO/Si would form as we desired, freestanding BTO membranes should be strongly in-plane polarized. We made systematic PFM measurements to investigate the domain configurations in BTO membranes, since in-plane ferroelectric domain patterns for fully relaxed BTO membranes have never been experimentally observed.

The *c*-axis oriented ferroelectric polarization of epitaxially compressive-strained BTO grown on SAO/STO was confirmed for our samples in the first place⁴⁵. Bistable switching was managed by applying biases of ± 20 V. Fig. 2a demonstrates the domain configurations for 30 u.c. as-grown BTO acquired after domain switching, including both out-of-plane (vertical PFM) and in-plane (lateral PFM) results. The presence of explicit 180° phase contrast in vertical PFM (VPFM) phase image together with the absence in lateral PFM (LPFM) phase image indicates as-grown BTO being purely out-of-plane polarized, which is further affirmed by the out-of-plane phase hysteresis and butterfly-shape amplitude loop in Fig. 2b. In addition, in VPFM phase image, the unbiased area exhibits evident monodomain structure. The consistency between outermost unbiased area and innermost positive-biased area further illustrates the

existence of built-in field, which points down to the STO substrate and brings about spontaneous out-of-plane downward polarization in BTO layers, consistent with the previous report⁴⁵.

The same domain writing process was then performed on 30 u.c. freestanding BTO membrane transferred onto the Si (the BTO/Si heterostructure), with bias voltages of ± 13 V. The VPFM images and LPFM images obtained after switching are respectively shown in Fig. 2c. For VPFM, the created phase contrast is barely visible, implying the weak out-of-plane ferroelectric ground state of relaxed BTO membranes. The faint phase contrasts persist in freestanding BTO membranes of thicknesses lower than 30 u.c. (Supplementary Fig. 2a), sometimes even vanishing about an hour after switching (Supplementary Fig. 2b). However, thick BTO membranes, such as 160 u.c. BTO, show sharp phase contrast in poled regimes (Supplementary Fig. 2c), which is consistent with previous observations³⁷. This instability of out-of-plane polarization in thinner freestanding BTO membranes can be understood as a result of size effect due to strain release and the existence of depolarizing field. The phase matching of the unpoled perimeter and positively poled regime in BTO/Si sample further suggests the intrinsic polarization oriented downwards to Si. Recalling that samples were upended onto Si, the seemingly same downward polarization before and after transferring actually reflect a 180° distinction. LPFM scans for 30 u.c. BTO/Si show completely different results from the as-grown ones. The in-plane box-in-box pattern is switched, while the unbiased region sustains pristine high contrast multidomain structure. From the in-plane hysteresis loop in Fig. 2d, it is apparent that BTO membranes generate in-plane ferroelectricity after released onto Si. Similar behaviors are also observed in other BTO/Si samples, with thicknesses varying from 4 u.c. to 160 u.c. (Supplementary Fig. 3). Angle-resolved PFM measurements were carried out (Supplementary Fig. 4), revealing that in-plane polarization inside the randomly selected domain areas are mostly *a*-axis or *b*-axis equivalently oriented. It is also noteworthy that the amplitudes of LPFM scanning results are even larger than those of VPFM scanning at the same driving voltage, evidencing the strong in-plane polarization in ferroelectric phase of BTO/Si.

Structural transition and the corresponding origin. To study the polarization rotation after film releasing, systematic structural characterization has been performed by XRD on BTO films before and after transferring. By conducting high-resolution XRD 2θ - ω scans and reciprocal space mapping (RSM) measurements (Supplementary Fig. 5, 6) for BTO samples of various thicknesses, we obtained their lattice constants and the corresponding *c/a* ratios, as shown in Fig. 3a, b. Compared with tetragonal phase in bulk BTO ($a = b = 3.992$ Å, $c = 4.036$ Å), the as-grown BTO films of all thicknesses show larger out-of-plane constants and smaller in-plane constants, which is resulted from the epitaxial compressive strain imposed by STO substrates. Sharing the same in-plane constant with STO substrates, the 15 u.c. as-grown BTO proved to be coherently strained, with its *c/a* ratio reaching 1.06, much larger than the bulk BTO *c/a* ratio (1.011). However, as the as-grown films become thicker, the relaxation effect becomes prominent, leading to the increase of *a* values and the decrease of *c* values. Hence, the *c/a* ratios decrease monotonously from 1.06 to 1.024 as thicknesses increase from 15 u.c. to 160 u.c., gradually approaching the *c/a* ratio (1.011) for bulk BTO (Fig. 3a). In spite of this, for as-grown BTO not thicker

than 160 u.c., the c/a ratios are always larger than 1.024, suggesting the stable c -axis tetragonal phase of the crystal structures.

The freestanding BTO membranes on Si display strikingly different structural information from the as-grown ones. Freestanding BTO presents sharp increments in a values and reductions in c values compared with as-grown films. And the measured c/a ratios of all-thickness BTO membranes are close to 1.00. Such variations indicate the strong strain relaxation of the freestanding membranes after release from substrates, and give rise to crystal structure collapses in c direction and expansions in a direction in general. In addition, without effective screening from electrodes during transfer, the large depolarizing field in freestanding BTO membranes also tends to induce the formation of in-plane polarization^{42–44} to preserve polar order⁴⁶. Therefore, through the transferring process, under the combined influence of strain relaxation and depolarization field effect, BTO membranes transform from c -axis tetragonal phase to a mixture of a -axis, b -axis and c -axis tetragonal phases to stabilize ferroelectric polarization. In light of that each tetragonal phase (a -axis, b -axis or c -axis) possesses especially elongated a , b or c values, the mixture of three tetragonal phases leads to the overall c/a ratios about 1.00, which were exactly acquired by XRD measurements.

As structural transition occurred from pure c -axis phase to a combination of three tetragonal phases, the potential landscapes will be notably altered. The changes of potentials are schematically illustrated in Fig. 3c-f. In out-of-plane direction, the as-grown BTO layers prefer to polarize towards the STO substrates (called downward polarization), which may be attributed to the interface effect determined by the termination surface⁴⁵ (Fig. 3c). Nevertheless, during release, without clamping effect, the strong depolarizing field caused by initial out-of-plane polarization cannot be stabilized anymore, resulting in weakened c -axis ferroelectricity (Fig. 3d and Supplementary Fig. 2c). Such depolarization, together with strain relaxation, prompts remarkable alteration in the potential profile of (001) plane and the generation of in-plane ferroelectricity. Compared to the as-grown BTO clamped on the substrate (Fig. 3e), freestanding BTO membrane possesses favorable in-plane polarization pointing along [100] and equivalent directions with minimum potential, as schematically shown in Fig. 3f. Such local in-plane ferroelectric instability comes from the formation of a -axis and b -axis tetragonal phases, implying the coexistence of polarization along a -axis and b -axis, which is in accordance with our data from angle-resolved PFM measurements (Supplementary Fig. 4).

Identification and manipulation of conductive DWs. The pronounced in-plane ferroelectricity indicates BTO/Si an optimal platform for exploration and manipulation of the expected conductive DWs. Using c-AFM (Fig. 4a), we identified the intrinsic conductive DWs in BTO/Si and further accomplished the controllable creation of conductive DWs through scanning probe techniques.

To gain further insight into the electrical transport characteristics of the prototype DW memory, current-voltage (I-V) curves collected on the DW (ON state) and off the DW (OFF state) are shown in Fig. 4b. The acquired I-V curve of ON state displays an onset of conductivity at 1.5 V, the necessary bias to annihilate the potential barriers between the top electrode (tip) and the conductive DWs^{6, 47}. Above 1.5 V, the current

increases linearly along the voltage by fitting, which is a typical ohmic conduction behavior. Further, these I-V curves reveal a relatively high-current contrast of about three orders of magnitude between OFF and ON states ($\text{ON/OFF}=10^3$) at a read bias of +4V. Such ON/OFF ratio is around the same level of DW memories grown on perovskite substrates^{8, 16, 17, 19, 20, 25, 48}, signifying the comparable performance of our Si-based prototype as a candidate for nonvolatile memories. These results were repeatable for randomly picked spots.

Fig. 4c-e show the identification of intrinsic conductive DWs in freestanding BTO, representing the 'read-out' process of memory. In the rectangular area, domain patterns were examined by LPFM while conductivities were tested by c-AFM. Polarization orientations for in-plane domains were verified by domain switching process. Phase pattern in Fig. 4c displays distinct DWs, the regarding conductivities of which are demonstrated in c-AFM images (Fig. 4d). By overlapping two graphics, Fig. 4e reveals that only H-H DWs present notable conductivity ($\sim\text{nA}$), compared with those T-T DWs and domain regions ($\sim\text{pA}$), akin to many other complex oxides^{11, 22, 48, 49}. Such selectivity manifests the n-type conductivity in the relaxed BTO membrane on Si, just like bulk BTO⁶. Free electrons as charge carriers may be provided by intrinsic excitation, biased tip injection or charged defects such as oxygen vacancies⁴⁸. In order to screen the positive bound charges, the electrons accumulate at H-H DWs, bringing out the detected conductivity.

The ultimate step for memory functionality realization is to achieve artificial creation of conductive DWs through external electric field, which represents the 'writing' process of memory. By scanning the negative-bias tip (from -6 V to -10 V, amplitudes vary by thickness) over a small horizontal stripe region, we were able to switch polarization orientations and artificially create H-H conductive DWs by the electric trailing field of the tip^{50, 51}. Fig. 4e and Fig. 4f respectively demonstrate the domain structures before and after domain switching. Evident H-H DWs appeared after switching, and was verified to be conductive in the later c-AFM measurement (Fig. 4g, h). The successful manipulation of the conductive DWs decisively displayed the potential industrial applicability of perovskite/Si ferroelectric DW memories.

Discussion

Apart from ON/OFF ratio, integration density is also a necessary concern in the application of nonvolatile DW memories. From the thickness-dependent LPFM results of freestanding BTO membranes, in-plane DW densities of our BTO/Si prototype are also analyzed. As shown in Supplementary Fig. 7, domain sizes tend to reduce as films become thinner, and thus the fractions of DWs in the whole material are also higher in thinner films. This relation between thickness and DW density suggests a potential path to improve integration density of industrial DW memory. As the film thickness shrinks down, it is possible to obtain high-density conductive DWs as we elevate the number of DWs through dimensionality engineering.

It is also worth mentioning that the widely reported ferroelectric DW memory with the architecture of coplanar metal electrodes^{25, 26, 48} is also supposed to be applicable to our BTO/Si prototype. In that case,

two top electrodes bridging the conductive DW can be utilized for both 'writing' and 'reading' process, through a coplanar switching bias, as also illustrated in Fig. 1a.

In summary, the in-plane ferroelectric multidomain structures have been realized and systematically revealed in BTO membranes after transferred onto Si, the origin of which proved to be the collaboration of the strain relaxation and the depolarizing field effect during release. With the in-plane ferroelectricity being the foundation, functionality of conductive DW memory was realized by the integrated prototype BTO/Si, as conductive DWs can both be 'read' and 'written' in the heterostructure. The revealed 90° polarization rotation and our manipulation of conductive DWs in BTO/Si heterostructure offers a promising candidate for high-density non-volatile DW memories and provides a brand-new strategy for fabricating perovskite/Si alternatives to next-generation electronics including FETs and nanosensors.

Online Content

Any methods, references, data availability, acknowledgements, author contributions, competing interests, supplementary information and any associated accession codes are available in the online version of this paper.

Declarations

Data availability

The data that support the findings of this study are available from the authors on reasonable request.

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Author contributions

Y. F. N. conceived and directed the experiments. Assisted by T. Y. G and J. H. G, H. Y. S. grew the BTO/SAO/STO heterostructures. Using clamping device made by J. F. Y., H. Y. S., J. R. W. and Y. W. L. integrated the prototype through transferring. Y. S. W. and Y. F. H. completed electrodes deposition by electron beam lithography. H. Y. S. and J. R. W. performed the ferroelectricity characterization. J. R. W. analyzed the PFM data with the help from H. Y. F. H. Y. S., J. R. W and J. H. G conducted XRD measurements, with H. Y. S analyzing the data. J. R. W., H. Y. S. and Y. W. L. completed c-AFM tests. Y. F.

N., J. R. W. and H. Y. S. wrote the manuscript. All authors discussed the data and contributed to the manuscript.

Competing financial interests

The authors declare no competing financial interests.

Additional information

Supplementary information is available for this paper at XXX. Correspondence and requests for materials should be addressed to Y.F.N. Reprints and permissions information is available at XXX.

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Figures

Figure 1

Schematics of DW memory prototype and fabrication process. a, OFF (high resistance, left panel) and ON (low resistance, right panel) states for conductive DW memory prototype. Arrows represent polarization orientations and the red interface denotes the position of DW. b, Schematics of operations for transferring the as-grown BTO film directly onto the Si. The optical image shows the released 30 u.c. BTO membrane on Si substrate.

Figure 2

Ferroelectric evolution of BTO before and after transferring. a,c, Vertical (out-of-plane) and lateral (in-plane) PFM domain switching results for as-grown 30 u.c. BTO/SAO/STO and 30 u.c. BTO/Si. Bias voltages of ± 20 V and ± 13 V are applied in a and c, respectively. The blue box and green box areas are scanned by a dc bias tip with positive and negative voltages, respectively. Schematics below c illustrate the trailing field method for in-plane polarization switching^{50, 51}. b,d, Local phase hysteresis loops and amplitude butterfly curves obtained from out-of-plane signal of 30 u.c. BTO/SAO/STO in a and in-plane signal of 30 u.c. BTO/Si in c, respectively.

Figure 3

Structural transition of BTO. a,b, Lattice constants and c/a ratios of BTO at different thicknesses before and after transferring. Bulk BTO lattice constants are offered for comparison in a. The insets in b schematically illustrate the overall structural deformation on a unit cell scale. Error bars are drawn considering instrument precision and accidental errors. c,d, Qualitative potential diagram along [001] for BTO/SAO/STO in c and BTO/Si in d. Out-of-plane polarization directions are shown for both cases to clarify the c-axis polarization variation. e,f, Qualitative in-plane potential diagram for BTO/SAO/STO in e and BTO/Si in f.

Figure 4

'Reading' and 'Writing' of conductive DWs. a, Schematics of c-AFM measurement circuit. b, I-V curves on and off the conductive DWs. c-e, Identification of the intrinsic conductive DWs. Current signals from c-AFM test (c) and in-plane phase signals from LPFM test (d) are overlapped for comparison (e). Arrows represent the polarization orientations in d. f-i, Creation of the new conductive DWs. In-plane domain structures measured by LPFM before (f) and after domain switching (g) are displayed with arrows represent the polarization. Current signals measured by c-AFM (h) after domain switching are overlapped over in-plane phase result for comparison (i).

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