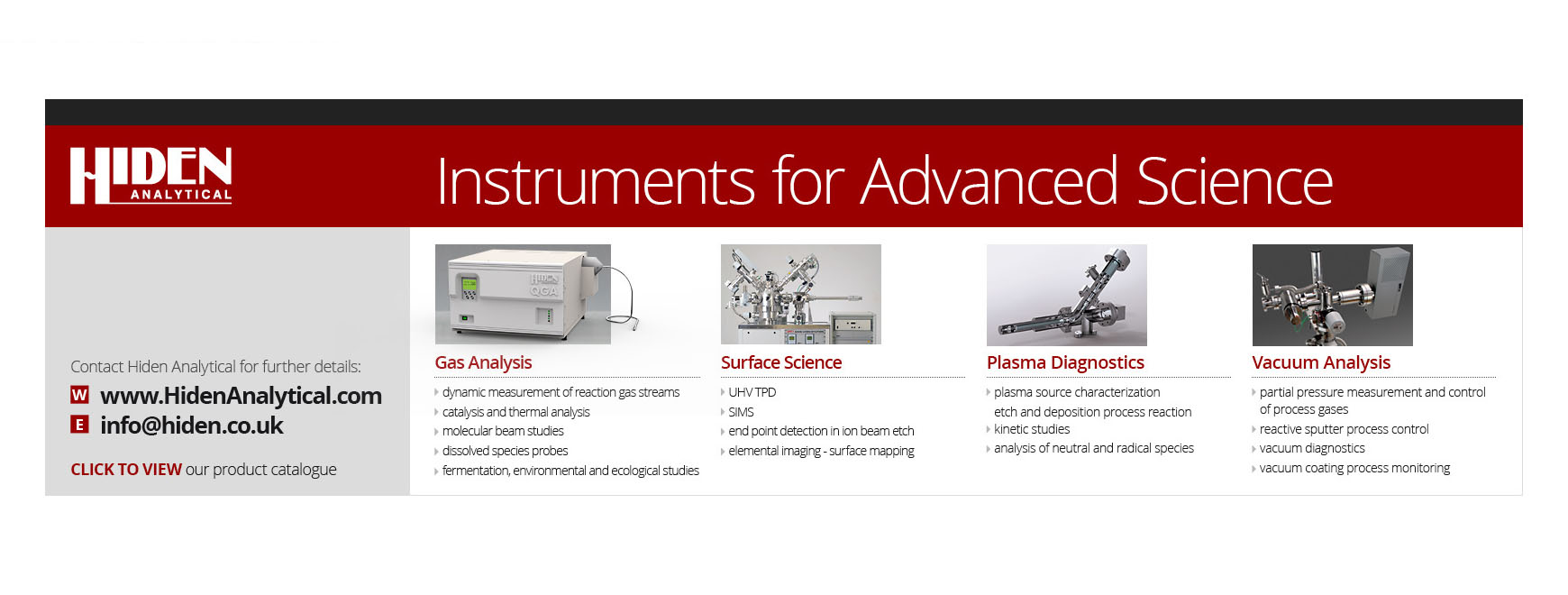
**Evolution of ferroelectric HfO2 in ultrathin region down to 3nm**   
[Xuan Tian](http://aip.scitation.org/author/Tian%2C+Xuan), [Shigehisa Shibayama](http://aip.scitation.org/author/Shibayama%2C+Shigehisa), [Tomonori Nishimura](http://aip.scitation.org/author/Nishimura%2C+Tomonori), [Takeaki Yajima](http://aip.scitation.org/author/Yajima%2C+Takeaki), [Shinji Migita](http://aip.scitation.org/author/Migita%2C+Shinji), and [Akira Toriumi](http://aip.scitation.org/author/Toriumi%2C+Akira)

Citation: Appl. Phys. Lett.**112**, 102902 (2018); doi: 10.1063/1.5017094 View online: <https://doi.org/10.1063/1.5017094>  
View Table of Contents: <http://aip.scitation.org/toc/apl/112/10>  
Published by the [American Institute of Physics](http://aip.scitation.org/publisher/)

**Articles you may be interested in**   
[Ferroelectricity in hafnium oxide thin films](http://aip.scitation.org/doi/abs/10.1063/1.3634052)   
[Applied Physics Letters](http://aip.scitation.org/doi/abs/10.1063/1.3634052) **[99](http://aip.scitation.org/doi/abs/10.1063/1.3634052)**[, 102903 (2011](http://aip.scitation.org/doi/abs/10.1063/1.3634052)); 10.1063/1.3634052  
 [Enhancing ferroelectricity in dopant-free hafnium oxide](http://aip.scitation.org/doi/abs/10.1063/1.4973928)   
[Applied Physics Letters](http://aip.scitation.org/doi/abs/10.1063/1.4973928) **[110](http://aip.scitation.org/doi/abs/10.1063/1.4973928)**[, 022903 (2017); 10.1063/1](http://aip.scitation.org/doi/abs/10.1063/1.4973928).4973928  
 [Ferroelectric and piezoelectric properties of Hf1-xZrxO2 and pure ZrO2 films](http://aip.scitation.org/doi/abs/10.1063/1.4983031) Applied Physics Letters **110**, 182905 (2017); 10.1063/1.4983031  
 [Kinetic pathway of the ferroelectric phase formation in doped HfO2 films](http://aip.scitation.org/doi/abs/10.1063/1.5003918) Journal of Applied Physics **122**, 124104 (2017); 10.1063/1.5003918   
[Effects of high pressure nitrogen annealing on ferroelectric Hf0.5Zr0.5O2 films](http://aip.scitation.org/doi/abs/10.1063/1.5003369) Applied Physics Letters **112**, 092906 (2018); 10.1063/1.5003369   
[Tunneling current in HfO2 and Hf0.5Zr0.5O2-based ferroelectric tunnel junction](http://aip.scitation.org/doi/abs/10.1063/1.5016823) Journal of Applied Physics **123**, 094501 (2018); 10.1063/1.5016823





APPLIED PHYSICS LETTERS 112, 102902 (2018) 

[Evolution of ferroelectric HfO2 in ultrathin region down to 3](https://doi.org/10.1063/1.5017094) [nm](https://doi.org/10.1063/1.5017094)

Xuan Tian,1,a)Shigehisa Shibayama,1Tomonori Nishimura,1Takeaki Yajima,1Shinji Migita,2 and Akira Toriumi1   
1Department of Materials Engineering, The University of Tokyo, Hongo, Tokyo 113-8656, Japan   
2National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8569, Japan

(Received 24 November 2017; accepted 23 February 2018; published online 6 March 2018)

The ferroelectric properties of ultrathin Y-doped HfO2 films were investigated. Ferroelectricity was

demonstrated experimentally in 3 nm-thick Y-doped HfO2 via direct detection of displacement cur-

rents during polarization switching. The dependence on the HfO2 thickness within the 30 to 3 nm

range revealed that the ferroelectric properties decrease rapidly below a critical thickness. In the ultra-

thin HfO2 region, methods such as higher Y doping or metal capping annealing were required to fur-

ther stabilize the ferroelectric phase. These methods could be used to enhance the switchable polarization (Psw) to 35 lC/cm2in 5 nm- and 10 lC/cm2in 3 nm-thick Y-doped HfO2. This paper indi-

cates that HfO2 ferroelectricity is scalable even in the ultrathin region. Published by AIP Publishing.

<https://doi.org/10.1063/1.5017094>

Ultrathin ferroelectric films are required for high-density ferroelectric random-access memory (FeRAM)1,2 and non-volatile resistive memory with ferroelectric tunnel-ing junctions (FTJs).3–5However, the spontaneous polariza-tion of ferroelectric films tends to degrade as the thickness is scaled down due to intrinsic size effects,6,7interfacial mechanical stresses,8,9chemical effects,10and potentially increased intrinsic depolarization field effects11–13from unscreened charges at electrode interfaces. Therefore, the thickness dependence of the ferroelectric properties is of vital importance with regard to both materials science and practical applications.

Since the discovery of ferroelectric HfO2, well-behaved ferroelectric properties have been demonstrated in polycrys-talline HfO2 films by doping them with cations (Y, Sr, Al, Si, Ge, Gd, or Zr)14–20or anions (N).21Since HfO2 is fully compatible with conventional complementary metal-oxide-semiconductor (CMOS) processes and can be manufactured using common atomic layer deposition (ALD), chemical vapor deposition (CVD), or physical vapor deposition (PVD) methodologies, it may prove superior to traditional perov-skite ferroelectric materials with regard to applicability to ferroelectric devices. Scaled ferroelectric field-effect-transis-tors (FeFETs) with thin ferroelectric HfO2 layers have been fabricated using 28 nm22,23and 22 nm24technology node metal gate/high-k CMOS processes. It is thought that the fer-roelectricity in HfO2 originates from one of the orthorhom-bic phases (Pca21). This is not the ground state but rather is a metastable phase of HfO2. Therefore, it is expected that the resulting ferroelectricity can be influenced by external condi-tions such as stress,25–27doping,17,28thermal treatment,29,30 and film thickness.25,30,31It has been reported that decreasing the film thickness or particle size of HfO2 or structurally sim-ilar ZrO2 stabilizes high symmetric phases more than mono-clinic ones due to the lower surface energies in high symmetric phases.32–34However, most studies have focused on relatively thick ferroelectric HfO2 regions (>6 nm). No

a)Telephone: þ81-3-5841-1907. E-mail: [xuan@adam.t.u-tokyo.ac.jp](mailto:xuan@adam.t.u-tokyo.ac.jp)

systematic study of ultrathin ferroelectric HfO2 has been reported thus far.

In this paper, the evolution of ferroelectricity in Y-doped HfO2 samples with thicknesses from 30 nm to 3 nm is reported. We unambiguously demonstrate stable ferroelectric properties in 3 nm-thick Y-doped HfO2 via pulsed polariza-tion current measurements. Heavily doped p-type germa-because it improved the ferroelectric properties by forming nium (NA ¼ 5 � 1018cm�3) was used as the bottom electrode less of an interfacial layer than heavily doped Si or TaN elec-trodes.35Y-doped HfO2 films were deposited via rf co-sputtering of HfO2 and Y2O3 targets. The Y2O3 plasma power was controlled for adjusting the Y doping concentra-tion, while that of HfO2 was fixed to 80 W in all of the films. The atomic concentration of Y was estimated via x-ray pho-toelectron spectroscopy (XPS). Most of the films were proc-essed via post-deposition annealing (PDA) in N2 at 600�C for 30 s. However, the 4 nm and 3 nm samples used a PDA temperature of 650�C to enhance film crystallization. The HfO2 film thickness was measured via x-ray reflectivity (XRR). Au was deposited via vacuum evaporation through a

|  |
| --- |
| stencil mask to make a top electrode (TE) with an �43 lm radius. Samples that were processed by post-metallization |
| annealing (PMA) included a top TiN layer (�25 nm) depos-ited via rf sputtering, before they were subject to the previ- |

|  |  |
| --- | --- |
| |  | | --- | | ously discussed PDA thermal annealing process. | |

Ferroelectric properties were measured using polarization-voltage (P-V), current-voltage (I-V), and the double pulse mea-surements using a ferroelectric analyzer (FCE-1A, TOYO Corporation). A schematic of the sample structure and mea-surement configuration is shown in Fig. 1(a). Polarization was measured using P-V and double-pulse measurements five times and averaged. The first poling process was ignored in the P-V measurement. No field cycling was used in these measure-ments. Figure 1(b) shows both typical P-E and I-E measure-ment results from a 30 nm-thick Y-doped HfO2 film. Since the remanent polarization (Pr) in HfO2 is dependent on the Y doping concentration,14the Y2O3 sputtering power was con-trolled to induce the largest available Pr. The strongest Pr of

|  |  |  |
| --- | --- | --- |
| 0003-6951/2018/112(10)/102902/5/$30.00 | 112, 102902-1 | Published by AIP Publishing. |

|  |  |  |
| --- | --- | --- |
| 102902-2 | Tian et al. | Appl. Phys. Lett. 112, 102902 (2018)  FIG. 1. (a) Schematic views of a device with a top electrode (TE)/Y-doped HfO2/pþ-Ge structure and ferro-electricity measurement system. (b) P-E (black) and I-E (gray) characteris-tics of a 30 nm-thick Y-doped HfO2 capacitor measured at 10 kHz. The Y concentration is 0.53 at. %. |
|  | |

approximately 11 lC/cm2was produced using a Y2O3 sputter-ing power of 8 W. This corresponded to doping HfO2 with

more accurately.41,42The measurement step is briefly illus-trated in Fig. 2(c). A negative pulse is used to switch the

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| 0.53 at. % of Y. | polarization | up | before | the | double | pulse | measurement. |

Figure 2(a) shows a cross-sectional TEM image of a fer-roelectric capacitor with a 3 nm-thick Y-doped HfO2 layer. The film exhibits a polycrystalline structure (also indicated in the XRD results in Fig. 3) and sits on a pþ-Ge substrate with an interfacial layer of �0.4 nm. Piezoelectric-force micros-copy (PFM) is commonly used to characterize ferroelectricity in ultrathin films36,37but is prone to inclusion of artifacts that exhibit ferroelectric-like properties.38,39Therefore, direct dis-placement current measurements of ferroelectric switching were used in this study.

Experimental P-V and I-V results from 3 nm-thick HfO2 films are shown in Fig. 2(b). Compared to perovskite ferro-electrics,40the leakage current is well suppressed in the 3 nm HfO2 layer. This can be attributed to the fact that the HfO2 has a large barrier height with both Au and Ge. Although the P-V result includes a hysteresis loop, it is narrow and slanted.

Therefore, the first recorded current peak (IPþC) includes both polarization switching and capacitor charging currents, while the third current peak (IC) includes only the capacitor charg-ing current (no polarization occurs during the second pulse). Thus, the polarization switching current (IP) can be estimated from the difference between IPþC and IC. Figure 2(d) shows the double pulse measurement result from a 3 nm-thick HfO2 layer. The Psw value is estimated to be �2.8 lC/cm2based on integrating the IP. This is substantially comparable to the value obtained from the P-V curve. In addition, an ultrathin paraelectric Y2O3 measurement is illustrated in the inset of Fig. 2(d), in which exactly the same currents are shown for two pulses with no corresponding IP curve. This further sup-ports the idea that the current difference in the double pulses originates from ferroelectric polarization switching. Thus, fer-roelectricity in a 3 nm-thick Y-doped HfO2 layer has been

Therefore, it is difficult to determine whether it stems from confirmed directly.

ferroelectric characteristics. Displacement current peaks that directly indicate ferroelectric switching are clearly observed in the I-V result. The Psw obtained from the P-V hysteresis loop is �3.0 lC/cm2for 3 nm Y-doped HfO2 with the same Y concentration as a 30 nm layer. Since leakage current in thin films is intrinsically related to the P-V curve, double pulse measurements help to characterize the ferroelectric properties

The thickness dependence of Psw in 30 nm to 3 nm Y-doped HfO2 films prepared with a fixed Y doping concen-tration of 0.53 at. % is summarized in Fig. 3(a). As the HfO2 film thickness decreases, Psw gradually increases and then sharply decreases as the film thickness passes below a criti-

cal value of �8 nm. XRD results indicating the strongest HfO2 diffraction peaks are shown in Fig. 3(b). The results

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | FIG. 2. (a) TEM image of a ferroelec-tric capacitor with a Au/3-nm-thick Y-doped HfO2/pþ-Ge structure. (b) P-V (black) and I-V (gray) characteristics of a Au/3 nm-thick Y-doped HfO2/pþ-Ge ferroelectric capacitor. (c) Double pulse measurement diagram, where IPþC is the polarization switching cur-rent plus the capacitor charging cur- | | | | | | | | |
| rent, | IC | | is | the | capacitor | | charging | |
| current, IDC is the capacitor discharge current, and IL is the leakage current. (d) Double pulse measurement result | | | | | | | | |
| from | a | 3 nm-thick | | | | Y-doped | | HfO2 |
| capacitor. Results from an ultrathin (�4 nm) paraelectric Y2O3 capacitor are shown for comparison in the inset. | | | | | | | | |

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| 102902-3 | Tian et al. | Appl. Phys. Lett. 112, 102902 (2018)  FIG. 3. (a) Thickness dependence of | | | |
|  | |
| Psw | in | 30 nm–3 nm-thick | Y-doped |
| HfO2 films. The co-sputtering condi-tion was fixed for all of the samples.  (b) Grazing incidence x-ray diffraction (GIXRD) patterns from 27�to 33�for 20, 8, 6, and 4 nm thick samples. (c)  Thickness dependence of ro/t/c as deter-mined from the GIXRD results. (d)  The relationship between Psw and ro/t/c in Y-doped HfO2 samples thinner than 8 nm. | | | |

indicate the presence of both monoclinic and high symmetric phases. The high symmetric phase peak consists primarily of three phases: orthorhombic, tetragonal, and cubic (o/t/c), which are difficult to distinguish from each other.14,21,30We can define the high symmetric phase ratio (ro/t/c) using ro/t/c ¼ Io/t/c/(Io/t/c þ Im11-1 þ Im111) in the diffraction intensities in 27�–33�. The film thickness dependence of ro/t/c is shown in Fig. 3(c). This quantity also exhibits a critical thickness around 8 nm. Figure 3(d) indicates the relationship between Psw and ro/t/c below 8 nm. The Psw reduction in the ultrathin region may result from decreases in the ferroelectric phase.

Since interfacial effects should be more significant in thinner films, an internal driving force might be needed to recover their ferroelectric properties. Therefore, the effects of doping concentration and annealing methods on the crys-talline structure and ferroelectric properties of 5 nm Y-doped HfO2 were investigated. Either higher Y doping or PMA processing was expected to promote the high symmetric phase. The 5 nm-thick HfO2 in Fig. 3 exhibits a significantly reduced Psw of approximately 8 lC/cm2. Increasing the Y doping concentration to 1.51 at. % increases the Psw to approximately 24 lC/cm2, as shown in Fig. 4(b). When the PMA process with TiN capping is applied, no significant increase in the high symmetric phase is noted via XRD [Fig. 4(a)]. However, Psw improves significantly to 35 lC/cm2, as shown in Fig. 4(b). It is inferred that higher Y doping increases the portion of both the ferroelectric phase and the non-ferroelectric high symmetric phase. In Fig. 4(b), both PDA and PMA samples exhibit asymmetric Ec values, which may stem from a voltage polarity dependent asymmetric voltage drop in the pþ-Ge substrate. In addition, the differ-ence between the work functions of Au and pþ-Ge in PDA samples is larger than that between TiN and pþ-Ge in PMA samples. These are likely reasons for the asymmetric P-V curve shown in Fig. 4(b). Furthermore, the average Ec is

reduced in the PMA sample. This might be due to a lack of interfacial layer formation during the PMA process (data not shown).

The 5 nm-thick film requires a large driving force toward the ferroelectric phase in order to maintain ferroelec-tric polarization comparable to that of a thicker film. The properties of the 3 nm-thick Y-doped HfO2 film were investi-gated by changing the Y doping concentration and annealing method. Figure 4(c) shows a double pulse measurement of a 3 nm Y-doped HfO2 film with a Y concentration of 1.51 at. %. The Psw increases to �10 lC/cm2in the 3 nm-thick HfO2 film, demonstrating that ferroelectricity can be scaled down to the ultrathin region. Figure 4(d) summarizes the Y con-centration dependence of Psw in 3 nm HfO2 films prepared via PDA and PMA and shows that application of PMA to a 3 nm film is not as effective as using a 5 nm film with regard to maintaining a high Psw.

Figure 5(a) shows the dependence of Psw on the thick-nesses of samples produced via PMA and PDA processes. High Psw values are demonstrated with layers as thin as 5 nm. A sharp drop in Psw is observed with the 3 nm film, which implies that other effects may destabilize the ferro-electric state in the ultrathin region below 5 nm. The relation-ship between Psw and ro/t/c is plotted in Fig. 5(b) for different HfO2 thicknesses and both PDA and PMA processes. A 5 nm film prepared via a PMA process exhibits a high Psw under a high ro/t/c. This is the same trend as is observed with thicker films. On the other hand, a 3 nm film exhibits a weaker rela-tionship between Psw and ro/t/c. A combination of a high ro/t/c and low Psw is shown in Fig. 5(b) for a 3 nm Y-doped HfO2 device. It is inferred that the low Psw in 3 nm HfO2 might stem from two causes. One potential cause is that more regions may remain amorphous in the 3 nm film because it is difficult to crystallize thinner films. The other is that more

|  |
| --- |
| phases might transfer from orthorhombic to tetragonal when |

|  |  |  |
| --- | --- | --- |
| 102902-4 | Tian et al. | Appl. Phys. Lett. 112, 102902 (2018)  FIG. 4. (a) GIXRD results from 25�to 35�and (b) P-V characteristics in 5 nm-thick Y-doped HfO2 samples fab-ricated using various processing condi-  tions: 0.53 at. % (gray) and 1.51 at. %  (blue) Y doping with PDA, and 0.53 at.  % Y doping with PMA (red). (c)  Double pulse measurement of a 3 nm-  thick Y-doped HfO2 (1.51 at. %) sam-ple fabricated using PDA. (d) The  relationship between Psw and the Y doping concentration in 3 nm-thick Y-  doped HfO2 samples fabricated via PDA and PMA.  FIG. 5. (a) Thickness dependence of  Psw in 30 nm-thick to 3 nm-thick Y-doped HfO2 films prepared via PDA and PMA. Y doping is maintained at  8 W (0.53 at. %) for all samples. (b)  The relationship between Psw and ro/t/c in Y-doped HfO2 with PDA and PMA. The Y doping concentration (0.53 at.  %) is the same for all films except the  solid red circle (1.51 at. %). |
|  | |

the film is less than 3 nm thick.32,43Since Psw is on the trend from the thicker side down to 5 nm HfO2, the depolarization field effect will not be significant. In fact, preliminary reten-tion experimental results from PMA samples have shown that 5 nm films exhibit weak Psw decreases comparable to those of 30 nm films over the course of 103s, while 3 nm films exhibit significant reductions in relative polarization as time passes (data not shown). This might result from the presence of a stronger depolarization field in the 3 nm film, but ferroelectric orthorhombic phase reduction should also be considered. Further study of the depolarization field effect is in progress.

In summary, ultrathin ferroelectric HfO2 films as thin as 3 nm have been unambiguously demonstrated. A rapid Psw decrease below a critical thickness of approximately 8 nm was observed upon systematically testing the thickness depen-dence of HfO2 ferroelectricity. To overcome this challenge, either the Y doping concentration was increased or a PMA process with top capping was employed. Both approaches are useful for stabilizing the ferroelectric phase in ultrathin HfO2. Experimental results revealed that a 5 nm HfO2 exhibited a Psw that was comparable to those of thick films. Psw values of

up to 10 lC/cm2were achieved in 3 nm HfO2 films. However, the lower Psw values observed in 3 nm films should be investi-gated further. This observation of HfO2 ferroelectricity evolution in the ultrathin region supports the scalability of nano-scale ferroelectric HfO2 devices.

This research was supported by JST-CREST (Grant No. JPMJCR14F2). X.T. was financially supported by the China Scholarship Council (CSC).

1K. Kim and S. Lee, [J. Appl. Phys.](https://doi.org/10.1063/1.2337361) 100(5), 051604 (2006).

2J. Muller, T. S. Boscke, S. Muller, E. Yurchuk, P. Polakowski, J. Paul, D. Martin, T. Schenk, K. Khullar, A. Kersch, W. Weinreich, S. Riedel, K. Seidel, A. Kumar, T. M. Arruda, S. V. Kalinin, T. Schl€osser, R. Boschke, R. van Bentum, U. Schr€oder, and T. Mikolajick, in IEEE International Electron Devices Meeting, Washington, DC, USA (IEEE, 2013), pp. 280–283.

3A. Chanthbouala, A. Crassous, V. Garcia, K. Bouzehouane, S. Fusil, X. Moya, J. Allibe, B. Dlubak, J. Grollier, S. Xavier, C. Deranlot, A. Moshar, R. Proksch, N. D. Mathur, M. Bibes, and A. Barthelemy, [Nat. Nanotechnol.](https://doi.org/10.1038/nnano.2011.213) 7(2), 101–104 (2012).

4A. Gruverman, D. Wu, H. Lu, Y. Wang, H. W. Jang, C. M. Folkman, M. Ye Zhuravlev, D. Felker, M. Rzchowski, C. B. Eom, and E. Y. Tsymbal, [Nano Lett.](https://doi.org/10.1021/nl901754t) 9(10), 3539–3543 (2009).

|  |  |  |
| --- | --- | --- |
| 102902-5 | Tian et al. | Appl. Phys. Lett. 112, 102902 (2018) |

5Z. Wen, C. Li, D. Wu, A. Li, and N. Ming, [Nat. Mater.](https://doi.org/10.1038/nmat3649) 12(7), 617–621 (2013).

6W. Cochran, [Adv. Phys.](https://doi.org/10.1080/00018736000101229) 9(36), 387–423 (1960).

7V. Fridkin and S. Ducharme, Ferroelectricity at the Nanoscale: Basics and Applications (Springer, 2014), p. 11.

8A. G. Zembilgotov, N. A. Pertsev, H. Kohlstedt, and R. Waser, [J. Appl. Phys.](https://doi.org/10.1063/1.1427406) 91(4), 2247–2254 (2002).

9F. Griggio, S. Jesse, A. Kumar, O. Ovchinnikov, H. Kim, T. N. Jackson, D. Damjanovic, S. V. Kalinin, and S. Trolier-McKinstry, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.108.157604) 108(15), 157604 (2012).

10J. F. Ihlefeld, P. G. Kotula, B. D. Gauntt, D. V. Gough, G. L. Brennecka, P. Lu, and E. D. Spoerke, [J. Am. Ceram. Soc.](https://doi.org/10.1111/jace.13576) 98(7), 2028–2038 (2015). 11N. Sai, A. M. Kolpak, and A. M. Rappe, [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.72.020101) 72(2), 020101 (2005).

12G. Gerra, A. K. Tagantsev, N. Setter, and K. Parlinski, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.96.107603)  96(10), 107603 (2006).

13J. Junquera and P. Ghosez, [Nature](https://doi.org/10.1038/nature01501) 422(6931), 506–509 (2003).

14T. Olsen, U. Schr€oder, S. M€uller, A. Krause, D. Martin, A. Singh, J. M€uller, M. Geidel, and T. Mikolajick, [Appl. Phys. Lett.](https://doi.org/10.1063/1.4747209) 101(8), 082905 (2012).

15T. Schenk, S. Mueller, U. Schroeder, R. Materlik, A. Kersch, M. Popovici, C. Adelmann, S. Van Elshocht, and T. Mikolajick, in 2013 Proceedings of the European Solid-State Device Research Conference (ESSDERC) (2013), pp. 260–263.

16S. Mueller, J. Mueller, A. Singh, S. Riedel, J. Sundqvist, U. Schroeder, and T. Mikolajick, [Adv. Funct. Mater.](https://doi.org/10.1002/adfm.201103119) 22(11), 2412–2417 (2012).

17T. S. B€oscke, J. M€uller, D. Br€auhaus, U. Schr€oder, and U. B€ottger, [Appl.](https://doi.org/10.1063/1.3634052)  [Phys. Lett.](https://doi.org/10.1063/1.3634052) 99(10), 102903 (2011).

18L. Xu, T. Nishimura, S. Shibayama, T. Yajima, S. Migita, and A. Toriumi, [J. Appl. Phys.](https://doi.org/10.1063/1.5003918) 122(12), 124104 (2017).

19S. Mueller, C. Adelmann, A. Singh, S. Van Elshocht, U. Schroeder, and T.

Mikolajick, [ECS J. Solid State Sci. Technol.](https://doi.org/10.1149/2.002301jss) 1(6), N123–N126 (2012). 20J. Muller, T. S. Boscke, U. Schroder, S. Mueller, D. Brauhaus, U. Bottger, L. Frey, and T. Mikolajick, [Nano Lett.](https://doi.org/10.1021/nl302049k) 12(8), 4318–4323 (2012).

21L. Xu, T. Nishimura, S. Shibayama, T. Yajima, S. Migita, and A. Toriumi, [Appl. Phys. Express](https://doi.org/10.7567/APEX.9.091501) 9(9), 091501 (2016).

22J. Muller, E. Yurchuk, T. Schlosser, J. Paul, R. Hoffmann, S. Muller, D. Martin, S. Slesazeck, P. Polakowski, J. Sundqvist, M. Czernohorsky, K. Seidel, P. Kucher, R. Boschke, M. Trentzsch, K. Gebauer, U. Schroder, and T. Mikolajick, in Symposium on VLSI Technology (VLSIT), Honolulu, HI, USA (IEEE, 2012), pp. 25–26.

23M. Trentzsch, S. Flachowsky, R. Richter, J. Paul, B. Reimer, D. Utess, S. Jansen, H. Mulaosmanovic, S. M€uller, S. Slesazeck, J. Ocker, M. Noack, J. M€uller, P. Polakowski, J. Schreiter, S. Beyer, T. Mikolajick, and B. Rice, in 2016 IEEE International Electron Devices Meeting (IEDM), San Francisco, CA, USA (IEEE, 2016), pp. 294–297.

24S. D€unkel, M. Trentzsch, R. Richter, P. Moll, C. Fuchs, O. Gehring, M. Majer, S. Wittek, B. M€uller, T. Melde, H. Mulaosmanovic, S. Slesazeck,

T. Mikolajick, J. H€ontschel, B. Rice, J. Pelleri, and S. Beyer, in 2017 IEEE International Electron Devices Meeting (IEDM), San Francisco, CA, USA (IEEE, 2017), pp. 485–488.

25P. Polakowski and J. M€uller, [Appl. Phys. Lett.](https://doi.org/10.1063/1.4922272) 106(23), 232905 (2015). 26M. H. Park, H. J. Kim, Y. J. Kim, T. Moon, and C. S. Hwang, [Appl. Phys.](https://doi.org/10.1063/1.4866008)  [Lett.](https://doi.org/10.1063/1.4866008) 104(7), 072901 (2014).

27T. S. B€oscke, St. Teichert, D. Br€auhaus, J. M€uller, U. Schr€oder, U. B€ottger, and T. Mikolajick, [Appl. Phys. Lett.](https://doi.org/10.1063/1.3636434) 99(11), 112904 (2011).

28J. M€uller, U. Schr€oder, T. S. B€oscke, I. M€uller, U. B€ottger, L. Wilde, J. Sundqvist, M. Lemberger, P. K€ucher, T. Mikolajick, and L. Frey, [J. Appl. Phys.](https://doi.org/10.1063/1.3667205) 110(11), 114113 (2011).

29M. Hoffmann, U. Schroeder, T. Schenk, T. Shimizu, H. Funakubo, O. Sakata, D. Pohl, M. Drescher, C. Adelmann, R. Materlik, A. Kersch, and T. Mikolajick, [J. Appl. Phys.](https://doi.org/10.1063/1.4927805) 118(7), 072006 (2015).

30M. H. Park, H. J. Kim, Y. J. Kim, W. Lee, T. Moon, and C. S. Hwang, [Appl. Phys. Lett.](https://doi.org/10.1063/1.4811483) 102(24), 242905 (2013).

31E. Yurchuk, J. M€uller, S. Knebel, J. Sundqvist, A. P. Graham, T. Melde, U. Schr€oder, and T. Mikolajick, [Thin Solid Films](https://doi.org/10.1016/j.tsf.2012.11.125) 533, 88–92 (2013). 32M. Shandalov and P. C. McIntyre, [J. Appl. Phys.](https://doi.org/10.1063/1.3243077) 106(8), 084322 (2009).

33S. V. Ushakov, A. Navrotsky, Y. Yang, S. Stemmer, K. Kukli, M. Ritala, M. A. Leskel€a, P. Fejes, A. Demkov, C. Wang, B. Y. Nguyen, D. Triyoso, and P. Tobin, [Phys. Status Solidi B](https://doi.org/10.1002/pssb.200404935) 241(10), 2268–2278 (2004).

34M. W. Pitcher, S. V. Ushakov, A. Navrotsky, B. F. Woodfield, G. Li, J. B.

Goates, and B. M. Tissue, [J. Am. Ceram. Soc.](https://doi.org/10.1111/j.1551-2916.2004.00031.x) 88(1), 160–167 (2004). 35P. D. Lomenzo, Q. Takmeel, C. M. Fancher, C. Zhou, N. G. Rudawski, S. Moghaddam, J. L. Jones, and T. Nishida, [IEEE Electron Device Lett.](https://doi.org/10.1109/LED.2015.2445352)  36(8), 766–768 (2015).

36V. Garcia, S. Fusil, K. Bouzehouane, S. Enouz-Vedrenne, N. D. Mathur, A. Barthelemy, and M. Bibes, [Nature](https://doi.org/10.1038/nature08128) 460(7251), 81–84 (2009).

37A. Chernikova, M. Kozodaev, A. Markeev, D. Negrov, M. Spiridonov, S. Zarubin, O. Bak, P. Buragohain, H. Lu, E. Suvorova, A. Gruverman, and A. Zenkevich, [ACS Appl. Mater. Interfaces](https://doi.org/10.1021/acsami.5b11653) 8(11), 7232–7237 (2016). 38N. Balke, P. Maksymovych, S. Jesse, A. Herklotz, A. Tselev, C. B. Eom, I. I. Kravchenko, P. Yu, and S. V. Kalinin, [ACS Nano](https://doi.org/10.1021/acsnano.5b02227) 9(6), 6484–6492 (2015).

39R. K. Vasudevan, N. Balke, P. Maksymovych, S. Jesse, and S. V. Kalinin, [Appl. Phys. Rev.](https://doi.org/10.1063/1.4979015) 4(2), 021302 (2017).

40Y. S. Kim, D. H. Kim, J. D. Kim, Y. J. Chang, T. W. Noh, J. H. Kong, K. Char, Y. D. Park, S. D. Bu, J. G. Yoon, and J. S. Chung, [Appl. Phys. Lett.](https://doi.org/10.1063/1.1880443) 86(10), 102907 (2005).

41D. Y. Chen, M. Azuma, L. D. McMillan, and C. A. Paz de Araujo, in Proceedings of 1994 IEEE International Symposium on Applications of Ferroelectrics (1994), pp. 25–28.

42J. M€uller, T. S. B€oscke, D. Br€auhaus, U. Schr€oder, U. B€ottger, J. Sundqvist, P. K€ucher, T. Mikolajick, and L. Frey, [Appl. Phys. Lett.](https://doi.org/10.1063/1.3636417) 99(11), 112901 (2011).

43R. Materlik, C. K€unneth, and A. Kersch, [J. Appl. Phys.](https://doi.org/10.1063/1.4916707) 117(13), 134109

S. M€uller, J. Ocker, M. Noack, D.-A. L€ohr, P. Polakowski, J. M€uller, (2015).