Comparing Crystallography and Ferroelectric Properties of *a*-Axis Oriented $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ versus Non-*c*-Axis Oriented $\text{SrBi}_2\text{Ta}_2\text{O}_9$ Thin Films on Si(100)

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Growth, structure and properties of *a*-axis oriented ferroelectric $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin films on SrRuO_3 -electroded, buffered Si(100) substrates are compared with those of (116)-oriented ferroelectric $\text{SrBi}_2\text{Ta}_2\text{O}_9$ thin films on the same substrate. It is shown how the ferroelectric properties of (116)-oriented $\text{SrBi}_2\text{Ta}_2\text{O}_9$ films suffer from certain inherent crystallographic properties of the films, whereas *a*-axis oriented $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin films are free from such shortcomings. In result, the latter have a very large remanent polarization in amount of $32 \mu\text{C}/\text{cm}^2$ —a new record for bismuth-layered perovskite thin films on substrates—pointing to the superiority of *a*-axis oriented $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin films with respect to memory applications.

Keywords: Bismuth-layered perovskites; thin films; epitaxy; ferroelectricity

INTRODUCTION

Ferroelectric films of bismuth-layered compounds like $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT) or $(\text{Bi},\text{La})_4\text{Ti}_3\text{O}_{12}$ (BLT) are relevant to non-volatile ferroelectric random access memories (NV-FRAMs) [1–3]. Though commercial low-density memories made of polycrystalline SBT films are already in use, the integration of Bi-layered type ferroelectric films into the silicon technology and the increase of memory density (up to the Gbit-size range) remain a challenge. In particular, the growth of *uniformly oriented* Bi-layered films on *Si(100)* substrates is significant, because such films will be required, if the lateral size of ferroelectric memory cells will drop down to below 100 nm for Gbit memories [2, 4]. This cell size being of the order of the grain size of polycrystalline films, the arising non-uniformity problems with randomly oriented films [5] can most probably only be solved by using uniformly oriented films.

The presence of a non-zero polarization component *perpendicular* to the film plane is essential, if the film is to be used in a ferroelectric thin-film capacitor geometry. However, this presence is not trivial, because in many bismuth-layered compounds, the vector of the spontaneous polarization is perpendicular to the *c*-axis. This means, that *c*-axis oriented SBT films, the growth of which is easy, do not have a polarization component perpendicular to the film plane. As a consequence *non-c-axis oriented* SBT or BLT films should be grown on Si(100).

The growth of non-*c*-axis oriented (or textured) SBT, $\text{SrBi}_2\text{Nb}_2\text{O}_9$ (SBN) or $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ -based bismuth-layered perovskite films on *complex-oxide single crystal* substrates has been reported by several groups [6–14]. Though one can obtain useful information on growth-structure-property relations from such films, the latter suffer from their inapplicability within silicon microelectronics. The growth of non-*c*-axis oriented Bi-layered perovskite films on *electroded Si(100) substrates* was, however, achieved only very recently [15–22]. Whereas in our group, buffered Si(100) substrates were covered with SrRuO_3 bottom electrodes of different crystallographic orientations [15–20] yielding, e.g., (116)-oriented SBT films [15, 16, 18], other groups used Pt-coated Si(100) [21, 22]. Pt-coated SrTiO_3 was also used [23].

In the following, we will demonstrate that bismuth-layered perovskite films with an inclined polarization vector, like (116)-oriented SBT films, suffer from certain shortcomings of crystallographic origin, whereas uniformly *a*-axis oriented films are free from such shortcomings. In particular, structure and properties of the recently achieved *a*-axis oriented BLT films on Si(100) are considered, which have a very large perpendicular polarization component due to their uniform orientation [19, 20].

EXPERIMENTAL

The YSZ buffer layers, the SrRuO_3 electrode layers, and the bismuth-layered perovskite films were all grown by pulsed laser deposition (PLD) not breaking the controlled atmosphere conditions in between. An excimer laser (KrF; wavelength 248 nm) was used at a repetition rate of 5 or 10 Hz to ablate corresponding pressed powder targets in a pure oxygen atmosphere. Targets used for $\text{SrBi}_2\text{Ta}_2\text{O}_9$ and $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ film deposition were rich in bismuth. Substrate temperatures between 650°C and 825°C, laser fluences between 1.7 and 4.0 J/cm², and oxygen pressures between 30 and 100 Pa were applied. The deposition conditions for each of the layers or films were carefully optimized. For details, see [15–20]. The thickness of the YSZ buffer layer was 60 nm or 80 nm, the one of the SrRuO_3 electrode layer ranged from

10 nm (in case of *a*-axis oriented BLT films) up to 50 nm . . . 80 nm (in case of (116)-oriented SBT films). The thickness of the ferroelectric films ranged from 250 nm to 1 μ m. For the electrical characterization, Pt top electrodes with a diameter of 100 μ m were r.f. sputter-deposited at R.T. After top electrode deposition, the samples were annealed at 450 to 650°C for 15 to 30 min to stabilize the contact between the ferroelectric film and the Pt electrode.

The entire heterostructures were structurally characterized by X-ray diffraction (XRD) using a Philips X'Pert MRD four-circle diffractometer, by atomic force microscopy (AFM) using a Digital Instruments D5000 microscope in tapping mode, and by transmission electron microscopy (TEM) using a Philips CM20 T (at 200 keV primary energy) and a JEOL 4010 (at 400 keV primary energy) electron microscopes. Local piezoelectric hysteresis curves were recorded by probing the sample with a conductive AFM tip and measuring the piezoelectric constant vs. a direct current voltage applied between the tip and the electrode underneath the sample (piezoresponse-AFM, PFM), cf. [24]. The ferroelectric properties were evaluated using a ferroelectric tester (TF Analyzer 2000, AixACCT). Polarization vs. electric field curves were recorded at a frequency of 100 Hz or 200 Hz, and fatigue endurance was determined at a frequency of 0.1 to 10 MHz.

RESULTS AND DISCUSSION

Crystallographic Background

Crystallographic indexing in this work is based on the following unit cells, keeping in mind that two of them (Bi₄Ti₃O₁₂; SrRuO₃) are approximatives of the proper unit cells. (Data are based on well-known work of other authors.)

- SrBi₂Ta₂O₉: orthorhombic unit cell, $a = 0.5531$ nm, $b = 0.5534$ nm, and $c = 2.4984$ nm;
- Bi₄Ti₃O₁₂: pseudo-orthorhombic unit cell, $a = 0.545$ nm, $b = 0.541$ nm, and $c = 3.283$ nm;
- SrRuO₃: pseudo-cubic unit cell, $a = 0.393$ nm;
- YSZ: cubic unit cell, $a = 0.514$ nm;
- Si: cubic unit cell, $a = 0.5431$ nm.

(116)-Oriented SrBi₂Ta₂O₉ Films

Figure 1 shows a XRD θ - 2θ scan of the SBT/SrRuO₃/YSZ/Si structure. The peaks correspond to well-defined (116)-oriented SBT, (110)-oriented

Figure 1 X-ray θ - 2θ diffraction pattern of the (116) SBT film system. The peak labeled as ($\square$) is an unidentified phase. The inset shows ω -scans for the (a) SBT (2212), (b) SrRuO_3 (110), and (c) YSZ (200) peaks.

SrRuO_3 , and (100)-oriented YSZ. The epitaxial relationship, based on the XRD θ - 2θ results, is

$$\text{SBT}(116) \parallel \text{SrRuO}_3(110) \parallel \text{YSZ}(100) \parallel \text{Si}(100).$$

The full-width at half maximum (FWHM) values of the SBT (2212), SrRuO_3 (110), and YSZ(200) peaks (inset in Fig. 1) are 1.7° , 0.96° , and 1.07° , respectively, in a rocking curve. Figure 2 shows a ferroelectric hysteresis loop, revealing a remanent polarization of only $P_r = 3.4 \mu\text{C}/\text{cm}^2$ and a coercive field of $E_c = 71 \text{ kV}/\text{cm}$, and Fig. 3 indicates a very good fatigue endurance.

AFM topography images of these $\text{SrBi}_2\text{Ta}_2\text{O}_9$ films reveal long grains of four different azimuthal orientations, with a characteristic azimuthal difference angle of $\sim 20^\circ$ frequently occurring (Fig. 4). In comparison, (116)-oriented SBT films on (110) SrTiO_3 do not show this feature. An azimuthal difference angle of $\sim 20^\circ$ has also been systematically observed in XRD ϕ scans, revealing that this angle is present already in the SrRuO_3 electrode

Figure 2 Ferroelectric hysteresis loop of the Pt/(116)SBT/(110)SrRuO₃/(100)YSZ/(100)Si heterostructure.

layer, but not in the YSZ buffer layer. After evaluating various pole figures (*cf.* [15]), the explanation is schematically given in Fig. 5: Due to its (110) orientation, the SrRuO₃ thin film consists of domains of four different azimuthal orientations, with an azimuthal difference angle of 19.5° regularly occurring (see Figure). The (116)-oriented SBT film growing on these four azimuthal domains of the (110)-oriented SrRuO₃ film turns out to consist of eight corresponding azimuthal domains (or multiple twins), *viz.* two for each azimuthal SrRuO₃ domain, with corresponding angles of ~20° [15]. TEM cross-sectional investigations (Fig. 6) indeed revealed that both the SrRuO₃

Figure 3 Fatigue endurance of the Pt/(116)SBT/(110)SrRuO₃/(100)YSZ/(100)Si heterostructure.

Figure 4 AFM topography image of the (116)SBT film showing characteristic angles of $\sim 20^\circ$ between grains.

and the SBT film consist of azimuthal domains about 50 nm in lateral size, and that the pattern of azimuthal domains in the SBT film replicates the pattern in the SrRuO_3 electrode layer: Each SrRuO_3 domain of a specific azimuthal orientation induces the growth of a region of a corresponding azimuthal orientation in the SBT film. While the other interfaces are plane and sharp, the $\text{SrRuO}_3/\text{SrBi}_2\text{Ta}_2\text{O}_9$ interface has a characteristic roof-like morphology (Fig. 6).

All the azimuthal SBT domains, however, have their (116) plane parallel to the substrate surface, so that the vector of the remanent polarization is at an angle of $\sim 30^\circ$ with respect to the film surface in all these domains. Accordingly, the film could be uniformly polarizable over its entire area. The maximum possible perpendicular component of the remanent polarization amounts to $|P_r| \cdot \sin(30^\circ) = 0.5 |P_r|$, which is of the order of $5 \dots 8 \mu\text{C}/\text{cm}^2$ depending on stoichiometry [25]. The measured value of $3.4 \mu\text{C}/\text{cm}^2$ thus

Figure 5 Schematic of the rectangle-on-cube epitaxy of (110) SrRuO₃ on (100) YSZ: (a) (100) plane of YSZ; (b) (110) plane of SrRuO₃; (c) four epitaxial variants.

indicates that either non-switching ferroelectric domains or probably some specific defects are present in the films.

In addition, and as a rule, hysteresis curves of the (116)-oriented SBT films were asymmetric (*cf.* Fig. 2). To establish the origin of this asymmetry, local piezoelectric hysteresis curves were recorded from individual SBT grains (azimuthal domains) in a PFM (Fig. 7), and HRTEM cross section images were taken from a region containing a few azimuthal domains only (Fig. 8). It turns out that microscopically the domain structure is related to the facetting of the SrRuO₃ layer. The latter is overall (110)-oriented, but the surface of it consists of {100} facets. (The facets are also visible in Fig. 6, but there the viewing direction is at about 45° to the one of Fig. 8 due to a different sample tilt.) One may consider the (116)-oriented growth of the BLT film as a (001)-oriented (“*c*-axis oriented”) growth of each domain on its underlying {100} facet. In result, the spontaneous ferroelectric polarization follows an up-down scheme as schematically indicated in Fig. 8. As long as the vectors of two neighbouring domains are

Figure 6 TEM cross section micrograph of the (116) SBT film system. A and B point to two SBT domains. The individual grains (azimuthal domains) are about 50 nm in lateral size.

head-tail-coupled, no charges accumulate at the domain boundaries. But the reversal of the polarization vector of one individual domain during switching (Fig. 7) results in a head-head coupling at one domain boundary and a tail-tail coupling at the other domain boundary. This corresponds to a thermodynamically unfavourable configuration. Switching into this unfavourable configuration is difficult, and returning back to the low-energy configuration is easy. Overall, an asymmetry of the local hysteresis curve

Figure 7 Local piezoelectric hysteresis curve of one single SBT grain (azimuthal domain) of about 50 nm in lateral size in a (116)-oriented SBT film.

Figure 8 HRTEM cross section image of an interface-near region of a (116)-oriented SBT film on a (110)-oriented, faceted SRO electrode on the YSZ buffer. The two arrows indicate possible directions of the polarization vector. Note that these vectors are not in the paper plane, but rather make an angle of $\approx 45^\circ$ with the latter.

results (Fig. 7) which also affects the symmetry of the global hysteresis curve (Fig. 2).

It is obvious that this inherent asymmetry of the hysteresis curves, the origin of which is the particular crystallography of the films, is a serious shortcoming of the (116)-oriented SBT films. As long as (110)-oriented SrRuO₃ films consisting of several growth domains (Fig. 5) are used as bottom electrodes for bismuth-layered perovskite thin films, this asymmetry of the switching properties seems to be unavoidable. This is not only true for (116)-oriented SBT and (118)-oriented BLT films growing on (110)-oriented SrRuO₃ bottom electrodes, but in modified and even more serious form also for (103)-oriented SBT and (104)-oriented BLT films on (111)-oriented SrRuO₃ bottom electrodes, *cf.* [17]. To avoid these problems, the growth of uniformly *a*-axis oriented films is necessary.

***a*-Axis Oriented BLT Films**

La-substituted bismuth titanate $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ (BLT), a rather new bismuth-layered perovskite material [3], promises to have both a high fatigue resistance and a high remanent polarization, thus potentially being able to replace SBT and PZT. Its growth in the form of *a*-axis oriented thin films on Si(100) could have been considered an important step forward towards NV-FRAMS.

Recently we succeeded in growing uniformly *a*-axis oriented $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin films on buffered and electroded Si(100) wafers by PLD under very well specified conditions [19, 20]. The key experience was the observation that during PLD of $\text{SrBi}_2\text{Ta}_2\text{O}_9$ films onto the model substrate SrLaGaO_4 (110), part of the films surprisingly were growing in the (100) orientation, this orientation “competing” with the common (116) orientation [12]. Analyzing this observation and making reasonable assumptions on conditions favouring the (100) orientation, we were able to grow *a*-axis oriented $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin films covering Si(100) substrates with very thin, strained (110)-oriented SrRuO_3 electrode layers on (100)-oriented YSZ buffer layers and applying “kinetic,” oxygen-rich growth conditions resulting in a high growth rate [19, 20].

As we knew before, BLT films grow in the (118) orientation on non-strained, 50 nm thick SrRuO_3 (110) bottom electrodes [13]. A calculation showed that epitaxially stretched SrRuO_3 (110) bottom electrodes are able to promote the nucleation of (100)-oriented BLT nuclei. It turned out that 10 nm thin SrRuO_3 bottom electrodes on 60 nm thick YSZ buffer layers indeed fulfilled this role. However, only part of the nuclei were (100)-oriented, the others still being (118)-oriented. In order to further promote the growth of the (100)-oriented nuclei into an entirely *a*-axis oriented film, a particularly high oxygen background pressure and a high deposition rate had to be used. The SrRuO_3 thickness ($t = 10$ to 50 nm), substrate temperature ($T = 650$ to 825°C), laser fluence ($J = 1.7$ to 4.0 J/cm²), laser repetition rate ($f = 2$ to 10 Hz), and oxygen pressure ($P = 30$ to 100 Pa) were all systematically varied while the YSZ thickness (60 nm) and the number of laser pulses (3000) were fixed. The volume fractions of the resulting (100) and (118) orientations were evaluated by XRD ϕ scans and pole figures. In addition, they could also be estimated from the morphology of the grains visible in AFM images. As a result of a rather extensive optimization process [20], the following optimum conditions were found to enable the growth of uniformly *a*-axis oriented BLT films: Substrate temperature $T = 765^\circ\text{C}$, laser fluence $J = 3.4$ J/cm², repetition rate $f = 7$ Hz, and oxygen background pressure

Figure 9 Pole figure of an *a*-axis oriented BLT film (117 peak of BLT). The center of the circle corresponds to $\psi = 0^\circ$, the rim to $\psi = 90^\circ$.

$P = 100$ Pa. The specific roles of these conditions in attaining the required film orientation have been discussed [19, 20].

Figure 9 shows a pole figure of the BLT 117 reflection, recorded from a BLT film of uniform *a*-axis orientation, demonstrating the entire (100) orientation of the film: All peaks are at $\psi \approx 57^\circ$, corresponding to the angle $\angle(100); (117) = 56.9^\circ$. (The peaks marked with asterisks originate from a shoulder of the YSZ (111) peak.) Each set of peaks consists of two sub-sets (of two peaks each), indicating four different azimuthal domains due to the specific crystallography of the system: Again, the four azimuthal domains of the SrRuO₃ bottom electrode result in the presence of azimuthal domains in the *a*-axis oriented BLT films. However, in this case they do not have serious consequences for the electrical properties, see below. The corresponding epitaxy relationship is

$$\text{BLT}(100) \parallel \text{SrRuO}_3(110) \parallel \text{YSZ}(100) \parallel \text{Si}(100);$$

$$\text{BLT}[001] \parallel \text{SrRuO}_3[001], \text{ where } \text{SrRuO}_3[\bar{1}11] \parallel \text{YSZ}(011) \parallel \text{Si}(011)$$

including four azimuthal domains. TEM investigations confirmed the *a*-axis orientation. High-resolution TEM images (Fig. 10) are in very good agreement with BLT structure models (superimposed in Fig. 10).

Figure 11 shows a plan-view TEM image of three neighbouring azimuthal domains. The *c*-axes of these domains are clearly in the plane of the film,

Figure 10 HRTEM cross section image of an *a*-axis oriented BLT grain, and superimposed $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ structure model.

Figure 11 Plan-view TEM image of three neighbouring azimuthal domains in a uniformly *a*-axis oriented BLT film.

Figure 12 P-E hysteresis loop of a Pt-BLT(100)-SrRuO₃(110) capacitor.

but they differ by azimuthal angles of 20°, 70°, and 90°, respectively. The reason for the presence of these azimuthal domains is again the structure of the underlying SrRuO₃ electrode layer, *cf.* Fig. 5. However, the *a*-axis of *all* the azimuthal domains being perpendicular to the film plane, all these domains have identical electrical properties along the film normal, and no more extensive asymmetry of the hysteresis curve occurs (see Fig. 12). As another consequence, these films have a high remanent polarization.

Figure 12 represents a ferroelectric polarization-electric field (*P-E*) hysteresis loop of a 1 μm thick *a*-oriented BLT film (recorded at 200 Hz at a maximum applied field of 740 kV/cm, with Pt top electrodes 100 μm in diameter). A remanent polarization of 32 μC/cm² has been attained, which is a new record for bismuth-layered perovskite films on substrates. Provisional fatigue measurements up to 10⁹ switching cycles showed almost no fatigue [20].

CONCLUSIONS

The ferroelectric properties of non-*c*-axis oriented bismuth-layered perovskite thin films grown on Si(100) and having an inclined polarization vector—like (116)- or (103)-oriented SrBi₂Ta₂O₉ or (118)- or (104)-oriented Bi_{3.25}La_{0.75}Ti₃O₁₂ films—suffer from inherent crystallographic properties of the films, whereas *a*-axis oriented Bi_{3.25}La_{0.75}Ti₃O₁₂ thin films are free from such shortcomings. In result, the latter have a very large remanent polarization in amount of 32 μC/cm²—a new record for bismuth-layered perovskite thin films on substrates—pointing to the superiority of *a*-axis oriented Bi_{3.25}La_{0.75}Ti₃O₁₂ thin films with respect to memory applications.

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REFERENCES

- [1] C. A. P. De Araujo, J. D. Cuchiaro, L. D. McMillan, M. C. Scott, and J. F. Scott, "Fatigue-free ferroelectric capacitors with platinum electrodes," *Nature* **374**, 627–629 (1995).
- [2] J. F. Scott, *Ferroelectric Memories*. Berlin: Springer; 2000.
- [3] B. H. Park, B. S. Kang, S. D. Bu, T. W. Noh, J. Lee, and W. Jo, "Lanthanum-substituted bismuth titanate for use in non-volatile memories," *Nature* **401**, 682–684 (1999).
- [4] M. Alexe, C. Harnagea, D. Hesse, and U. Gösele, "Patterning and switching of nanosize ferroelectric memory cells," *Appl. Phys. Lett.* **75**, 1793–1795 (1999).
- [5] A. Gruverman, "Scaling effect on statistical behavior of switching parameters of ferroelectric capacitors," *Appl. Phys. Lett.* **75**, 1452–1454 (1999).
- [6] J. Lettieri, Y. Jia, M. Urbanik, C. I. Weber, J.-P. Maria, D. G. Schlom, H. Li, R. Ramesh, R. Uecker, and P. Reiche, "Epitaxial growth of (001)-oriented and (110)-oriented $\text{SrBi}_2\text{Ta}_2\text{O}_9$ thin films," *Appl. Phys. Lett.* **73**, 2923–2925 (1998).
- [7] K. Ishikawa and H. Funakubo, "Electrical properties of (001)- and (116)-oriented epitaxial $\text{SrBi}_2\text{Ta}_2\text{O}_9$ thin films prepared by metalorganic chemical vapor deposition," *Appl. Phys. Lett.* **75**, 1970–1972 (1999).
- [8] S. E. Moon, T. K. Song, S. B. Back, S.-I. Kwun, J.-G. Yoon, and J. S. Lee, "Controlled growth of a-/b- and c-axis oriented epitaxial $\text{SrBi}_2\text{Ta}_2\text{O}_9$ ferroelectric thin films," *Appl. Phys. Lett.* **75**, 2827–2829 (1999).
- [9] J. Lettieri, M. A. Zurbuchen, Y. Jia, D. G. Schlom, S. K. Streiffer, and M. E. Hawley, "Epitaxial growth of non-c-oriented $\text{SrBi}_2\text{Nb}_2\text{O}_9$ on (111) SrTiO_3 ," *Appl. Phys. Lett.* **76**, 2937–2939 (2000).
- [10] H. N. Lee, A. Visinoiu, S. Senz, C. Harnagea, A. Pignolet, D. Hesse, and U. Gösele, "Structural and electrical anisotropy of (001)-, (116)-, and (103)-oriented epitaxial $\text{SrBi}_2\text{Ta}_2\text{O}_9$ thin films on SrTiO_3 substrates grown by pulsed laser deposition," *J. Appl. Phys.* **88**, 6658–6664 (2000).
- [11] H. N. Lee, S. Senz, A. Pignolet, and D. Hesse, "Epitaxial growth of non-c-oriented ferroelectric $\text{SrBi}_2\text{Ta}_2\text{O}_9$ thin films on SrTiO_3 substrates," *J. Europ. Ceram. Soc.* **21**, 1565–1568 (2001).
- [12] H. N. Lee, D. N. Zakharov, S. Senz, A. Pignolet, and D. Hesse, "Epitaxial growth of ferroelectric $\text{SrBi}_2\text{Ta}_2\text{O}_9$ thin films of mixed (100) and (116) orientation on SrLaGaO_4 (110)," *Appl. Phys. Lett.* **79**, 2961–2963 (2001).
- [13] H. N. Lee and D. Hesse, "Anisotropic ferroelectric properties of epitaxially twinned $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin films grown with three different orientations," *Appl. Phys. Lett.* **80**, 1040–1042 (2002).
- [14] T. Watanabe, H. Funakubo, K. Saito, T. Suzuki, M. Fujimoto, M. Osada, Y. Noguchi, and M. Miyayama, "Preparation and characterization of a- and b-axis-oriented epitaxially grown $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ -based thin films with long-range lattice matching," *Appl. Phys. Lett.* **81**, 1660–1662 (2002).

- [15] H. N. Lee, S. Senz, A. Visinoui, A. Pignolet, D. Hesse, and U. Gösele, "Epitaxial growth of non-*c*-oriented ferroelectric SrBi₂Ta₂O₉ thin films on Si(100) substrates," *Appl. Phys. A: Mater. Sci. Proc.* **71**, 101–104 (2000).
- [16] H. N. Lee, S. Senz, N. D. Zakharov, C. Harnagea, A. Pignolet, D. Hesse, and U. Gösele, "Growth and characterization of non-*c*-oriented epitaxial ferroelectric SrBi₂Ta₂O₉ films on buffered Si(100)," *Appl. Phys. Lett.* **77**, 3260–3262 (2000).
- [17] H. N. Lee, S. Senz, A. Pignolet, and D. Hesse, "Epitaxial growth of (103)-oriented ferroelectric SrBi₂Ta₂O₉ thin films on Si(100)," *Appl. Phys. Lett.* **78**, 2922–2924 (2001).
- [18] H. N. Lee, D. Zakharov, S. Senz, A. Pignolet, and D. Hesse, "Growth and characterization of non-*c*-axis-oriented SrBi₂Ta₂O₉ epitaxial thin films on Si(100) substrates with SrRuO₃ bottom electrodes," *Integr. Ferroelectrics* **39**, 73–80 (2001).
- [19] H. N. Lee, D. Hesse, N. D. Zakharov, and U. Gösele, "Ferroelectric Bi_{3.25}La_{0.75}Ti₃O₁₂ films of uniform *a*-axis orientation on silicon substrates," *Science* **296**, 2006–2009 (2002).
- [20] H. N. Lee, D. Hesse, N. Zakharov, S. K. Lee, and U. Gösele, "Growth of uniformly *a*-axis oriented ferroelectric lanthanum-substituted bismuth titanate films on silicon substrates," *Submitted to J. Appl. Phys.* (2002).
- [21] G. Asayama, J. Lettieri, M. A. Zurbuchen, Y. Jia, S. Trolier-McKinstry, D. G. Schlom, S. K. Streiffer, J.-P. Maria, S. D. Bu, and C. B. Eom, "Growth of (103) fiber-textured SrBi₂Nb₂O₉ films on Pt-coated silicon," *Appl. Phys. Lett.* **80**, 2371–2373 (2002).
- [22] R. R. Das, P. Bhattacharya, and R. S. Katiyar, "Enhanced ferroelectric properties in laser-ablated SrBi₂Nb₂O₉ thin films on platinumized silicon substrate," *Appl. Phys. Lett.* **81**, 1672–1674 (2002).
- [23] J. R. Duclère, M. Guilloux-Viry, A. Perrin, E. Cattin, C. Soyer, and D. Rèmeis, "Ferroelectric (116) SrBi₂Nb₂O₉ thin films epitaxially grown by pulsed laser deposition on epitaxial (110)Pt/(110)SrTiO₃ electrode," *Appl. Phys. Lett.* **81**, 2067–2069 (2002).
- [24] C. Harnagea, A. Pignolet, M. Alexe, D. Hesse, and U. Gösele, "Quantitative ferroelectric characterization of single submicron grains in Bi-layered perovskite thin films," *Appl. Phys. A: Mater. Sci. Proc.* **70**, 261–267 (2000).
- [25] E. C. Subbarao, "Family of ferroelectric bismuth compounds," *J. Phys. Chem. Sol.* **23**, 665–669 (1962).