Configuration and local elastic interaction of ferroelectric domains and misfit dislocation in PbTiO$_3$/SrTiO$_3$ epitaxial thin films

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Abstract
We have studied the strain field around the 90° domains and misfit dislocations in PbTiO$_3$/SrTiO$_3$ (001) epitaxial thin films, at the nanoscale, using the geometric phase analysis (GPA) combined with high-resolution transmission electron microscopy (HRTEM) and high-angle annular dark field—scanning transmission electron microscopy (HAADF-STEM). The films typically contain a combination of a/c-mixed domains and misfit dislocations. The PbTiO$_3$ layer was composed from the two types of the a-domain (90° domain): a typical a/c-mixed domain configuration where a-domains are 20–30 nm wide and nano sized domains with a width of about 3 nm. In the latter case, the nano sized a-domain does not contact the film/substrate interface; it remains far from the interface and stems from the misfit dislocation. Strain maps obtained from the GPA of HRTEM images show the elastic interaction between the a-domain and the dislocations. The normal strain field and lattice rotation match each other between them. Strain maps reveal that the a-domain nucleation takes place at the misfit dislocation. The lattice rotation around the misfit dislocation triggers the nucleation of the a-domain; the normal strains around the misfit dislocation relax the residual strain in a-domain; then, the a-domain growth takes place, accompanying the introduction of the additional dislocation perpendicular to the misfit dislocation and the dissociation of the dislocations into two pairs of partial dislocations with an APB, which is the bottom boundary of the a-domain. The novel mechanism of the nucleation and growth of 90° domain in PbTiO$_3$/SrTiO$_3$ epitaxial system has been proposed based on above the results.

Keywords: HRTEM, geometric phase analysis, strain, elastic interaction, 90° domain, misfit dislocation, PbTiO$_3$, ferroelectric thin film

1. Introduction
A typical ferroelectric material with a domain structure, PbTiO$_3$ also has a ferroelastic nature. Its 90° domains interact with the local strain fields. Many sources of the strain field, such as the lattice mismatch, thermal mismatch and phase transition, and high density of the misfit dislocations exist at the interface of an epitaxial thin film and its substrate. Many
microscopy studies of domain structure and misfit dislocation of ferroelectric thin films have been conducted. However, most of them only documented the structure and configuration as images and did not analyze the strain fields and their mutual elastic interaction [1–4].

The local strain fields are accompanied with the 90° domain and misfit dislocation. The residual strain in the film is well known to control the domain configuration: for example, the domain wall can be pinned by the dislocation [5]. Recently, Nakaki et al have reported that the 90° domain configuration depends strongly on the thickness of PbTiO$_3$ and PZT thin films [6–8]. Strain fields also affect the nucleation and growth of the 90° domain [9]. Consequently, the residual strain in the film controls the configuration and properties of the 90° domain.

To understand the elastic interaction between the 90° domain and misfit dislocation, it is important to elucidate the effect of the residual strain on each domain by measuring the local strain field at the nanoscale. To date, many approaches have been used to evaluate the residual strain or stress. For example, x-ray diffraction and Raman spectroscopy are used for micrometer–millimeter-scale analyses. However, these methods cannot be used to probe local strain fields around each domain or dislocation. Convergent beam electron diffraction (CBED) is a useful characterization technique of the strain field at the nanoscale. However, it requires a complex analysis at every measurement point and, therefore, is too time-consuming for strain mapping. Recently, quantitative high-resolution transmission electron microscopy (QHRTEM) has been developed, which uses Fourier transform mapping (FTM) [10], peak-fitting method [11], geometric phase analysis (GPA) [12] and so on. Among these approaches, GPA is advantageous for the spatial resolution and robustness of image quality. GPA has been applied to various materials such as metals, semiconductors and oxides [12–20].

The 90° domain structures of the bulk PbTiO$_3$ and BaTiO$_3$ crystals have been examined in earlier studies [11–14], but only for the simple 90° domain wall. Chu et al studied the interaction between the 90° domain and misfit dislocation of a lead-zirconate-titanate (PZT) nano island [15, 16]. It is yet unclear how the strain fields interact and affect the domain structure formation. Kiguchi et al applied GPA to a high-angle annular dark field—scanning transmission electron microscopy (HAADF-STEM) image and demonstrated that the technique can be used to characterize the local strain in PbTiO$_3$ epitaxial thin films. However, the details remain unclear [17].

In this study, we elucidate the strain fields and elastic interaction between the domain and misfit dislocation in PbTiO$_3$/SrTiO$_3$ epitaxial thin films. We propose a nucleation and growth mechanism of the nano sized 90° domain from the GPA of data obtained with aberration-corrected HRTEM and HAADF-STEM at the atomic scale.

2. Experimental procedures

PbTiO$_3$ films were grown at 600°C on SrTiO$_3$ (001) single-crystal substrates, using pulsed metalorganic chemical vapor deposition as described elsewhere [21, 22]. The film orientation and phases were identified by x-ray diffraction (XRD), using a 2θ–ω geometry with a two-dimensional detector, and a Cu-Kα line (0.15406 nm wavelength) monochromated by a (220) Ge four-crystal monochromator with a Göebel mirror (D8 Discover; Bruker AXS GmbH). Cross-sectional TEM specimens were prepared by low-angle Ar ion polishing (2–5 kV, 2–4˚, PIPS model 691; Gatan, Inc., USA) with epoxy as an adhesive agent. Final ion polishing was conducted with a low-energy ion milling system (0.3–1 kV, 4˚, Gentle Mill IV5; Technolog Linda Ltd, Hungary). The nanostructure and interfaces of the films were investigated using TEM and HAADF-STEM methods (TITAN 80–300 with an aberration corrector, 300 kV, Cs (TEM) ~1 μm, Cs (STEM) ~1.2 mm; FEI Co., Netherlands). HRTEM images were processed with a 2D Wiener filter (HREM-Filters Pro; HREM Research Inc., Japan). The imaging conditions for HAADF-STEM were as follows: convergent semi angle α = 10 mrad, collection semi angles β_in = 76 mrad, β_out = 200 mrad. The HAADF-STEM images were deconvoluted using the instrumental function, i.e. the probe function used in the HAADF-STEM, using the maximum entropy method (DeconvHAADF; HREM Research Inc., Japan). A two-dimensional strain field in the film was mapped with a nanometer spatial resolution using geometric phase analysis (GPA Phase; HREM Research Inc., Japan).

3. Results and discussion

3.1. Phase and orientation of PbTiO$_3$/SrTiO$_3$ thin film

Figure 1 shows a two-dimensional XRD pattern of a PbTiO$_3$/SrTiO$_3$ thin film. Each spot is identified to 00L (L = 1, 2) reflections of PbTiO$_3$ and SrTiO$_3$ as well as H00 (H = 100) satellite spots of PbTiO$_3$ along the χ-axis. The 00L and H00 reflections correspond to the c-domain and a-domain whose tetragonal c-axis of the PbTiO$_3$ unit cell orients out-of-plane and nearly in-plane, respectively. The PbTiO$_3$ thin film has a typical a/c-mixed domain configuration. The additional line around 29° is identified with the 222 reflection of the pyrochlore Pb$_2$Ti$_2$O$_6$ phase (ICDD Card No. 26-0142).

3.2. Cross-sectional structure of PbTiO$_3$/SrTiO$_3$ thin film

3.2.1 Overview and 90° domain configuration. Figure 2(a) shows a cross-sectional bright-field (BF) image of a PbTiO$_3$/SrTiO$_3$ thin film taken under the two-beam condition with the diffraction vector g = 002. Under the diffraction condition, the c-domain shows dark contrast attributable to the Bragg condition; the opposite is true for the a-domain. The PbTiO$_3$ layer shows a typical a/c-mixed domain configuration where the a-domains are about 10–30 nm wide. The bottom 100 nm layer shows a strong diffraction contrast so that the morphology details are unclear. Figure 2(b) is a cross-sectional BF-STEM image of the film, taken along the [010] zone axis and clearly showing the film morphology. In the bottom layer, very short and thin a-domains (contrast A) are visible in addition to the typical a-domain (contrast B) in...
Figure 1. Two-dimensional XRD pattern of a PbTiO$_3$/SrTiO$_3$ thin film.

Figure 2. (a) Cross-sectional bright field (BF) image of PbTiO$_3$/SrTiO$_3$ thin film taken under the two beam condition with the diffraction vector of $g = 002$; (b) cross-sectional BF-STEM image taken from the zone axis [010] direction. The contrast A is the nano sized $a$-domain, B is the normal $a$-domain, C is the threading dislocation, and D is the 180° domain.

3.2. Strain mapping and elastic interaction

3.2.1 Geometric phase analysis of PbTiO$_3$/SrTiO$_3$ thin film. First, we consider the applicability limits of GPA using a homogeneous region around the PbTiO$_3$/SrTiO$_3$ interface. Figures 3(a)–(c), respectively, show a cross-sectional HRTEM image, its magnification, and a HAADF-STEM image around a PbTiO$_3$/SrTiO$_3$ interface with no misfit dislocations or $a$-domains. Here, PbTiO$_3$ has a [001] orientation relative to the SrTiO$_3$ substrate, i.e. the $c$-domain. The interface is atomically sharp. In the HAADF-STEM image, the intensity at an atomic column is related to the average atomic number in the column. The background intensity, which is the intensity between atomic columns in the HAADF-STEM image, is related to the average atomic number of the substance. The background intensity is higher in PbTiO$_3$ than in SrTiO$_3$ because of the higher average atomic number. Figure 3(c) reveals that the bottom layer of PbTiO$_3$ is the Pb–O layer. In contrast, the top layer of the SrTiO$_3$ substrate is a Ti–O layer as follows from the background contrast. Figure 4 shows that phase maps of figure 3(a) correspond to the reciprocal vectors $g_1 = 101$ (P$_{g1}$) and $g_2 = 10\bar{1}$ (P$_{g2}$). These maps were calculated taking a reference region in the SrTiO$_3$ substrate. The phase corresponds to the local deviation of the lattice spacing of the (101) and (10\bar{1}) planes in the PbTiO$_3$ film from that of the SrTiO$_3$ reference at every point in the image. Figure 5 presents strain maps of figure 3(a), namely, the normal strain in the in-plane direction $\varepsilon_{xx}$ (a), the normal strain in the out-of-plane direction $\varepsilon_{yy}$ (b), the sharing strain $\varepsilon_{xy}$ (c), and the rigid rotation $\omega_{xy}$ (d). The line profiles of figures 5(e) and (f) were taken along the lines marked in (a) and (b), respectively. The strain maps of figures 5(a), (c) and (d) are almost flat; the profile of figure 5(e) reveals a deviation of less than 0.01 and the standard deviation of 0.003. There is a slight gradient in the out-of-plane direction in profile (e), which is directed along the wedge of the
TEM specimen. It is related to the thickness variation across the specimen and is sufficiently small for the mapping of strain around lattice defects, which is presented in the next section. Figure 5(a) shows that the normal strain $\epsilon_{xx}$ is within 0.01. The bulk lattice parameters are $c = 0.4153$ nm and $a = 0.3899$ nm for PbTiO$_3$ and $a = 0.3905$ nm for SrTiO$_3$ [1]. The lattice mismatch of the $a$-axis between those crystals is 0.0016 at room temperature. It is sufficiently small for the GPA, and thus, the normal strain map of $\epsilon_{xx}$ shows a flat image. However, the $\epsilon_{yy}$ strain component shows a distinct change between PbTiO$_3$ and SrTiO$_3$. The profile (f) reveals a strain of about 0.055, indicating that the tetragonality of the PbTiO$_3$ layer is 1.055. The lattice mismatch between the $c$-axis of PbTiO$_3$ and $a$-axis of SrTiO$_3$ is 0.063. The lattice parameters of this film are $c = 0.4097$ nm and $a = 0.3900$ nm [6, 7]. The difference between the thin film and bulk crystals can be ascribed to the lattice and thermal mismatches as well as the domain configuration. The $a$-axis spacing is almost identical to that of the bulk crystal, but the $c$-spacing is smaller. The value of the true normal strain $\epsilon_{yy} = 0.049$ agrees within the measurement accuracy with that deduced by the GPA.

Figure 3 shows an atomically sharp PbTiO$_3$/SrTiO$_3$ interface. However, the interface profiled in figure 5(f) is blurred and has a 1 nm width. This blurring is a GPA artifact in the GPA, a mask is placed on a diffraction spot of the HRTEM image, so that only a part of the reciprocal space is used for calculating the phase map. A point in the real space is expressed using infinite reciprocal space, and a limited volume of the reciprocal space corresponds to a finite volume in the real space. Therefore, phase and strain GPA maps have a limited spatial resolution, which is about 0.85 nm in this study.

In summary, it is possible to image strain with the GPA at a spatial resolution of less than 1 nm and with a strain sensitivity of better than 0.01. Care should be exercised when interpreting local structures, such as an atomically sharp interface.

### 3.3.2 Elastic interaction between the 90° domain and misfit dislocation

#### 3.3.2.1 Nanostructure around the 90° domain and misfit dislocation

Figure 6 shows an HRTEM image (a) magnified in (b) around an $a$-domain near the interface with edge-type dislocations (rectangular regions), an HAADF-STEM image (c) around two edge dislocations with an $a$-domain, and an edge-type dislocation without an $a$-domain in a separate region (d). Dashed lines in (a) and (b) mark the PbTiO$_3$/SrTiO$_3$ interface. The arrows in (c) mark an ant
Figure 5. Strain maps of figure 4: (a) the normal strain in the in-plane direction $\varepsilon_{xx}$, (b) the normal strain in the out-of-plane direction $\varepsilon_{yy}$, (c) the sharing strain $\varepsilon_{xy}$ and (d) the rigid rotation $\omega_{xy}$. The line profiles in (e) and (f) were taken along the lines marked in (a) and (b), respectively.

Figure 6. (a) An HRTEM image around the PbTiO$_3$/SrTiO$_3$ interface, (b) magnifies the boxed region in (a) around an $\alpha$-domain near the interface with edge-type dislocations, (c) is an HAADF-STEM image around edge dislocations with an $\alpha$-domain and (d) shows an edge-type dislocation without an $\alpha$-domain in a separate region. Dashed lines in (a) and (b) mark the PbTiO$_3$/SrTiO$_3$ interface. The arrows in (c) mark an anti phase boundary (APB) along the (101) plane.
phase boundary (APB) along the (101) plane. Figure 6 shows (a) an HRTEM image around the PbTiO$_3$/SrTiO$_3$ interface, (b) magnifies the boxed region in (a) around an $a$-domain near the interface with edge-type dislocations, (c) is an HAADF-STEM image around edge dislocations with an $a$-domain, (d) shows an edge-type dislocation without an $a$-domain in a separate region. The dashed line in (a) marks the PbTiO$_3$/SrTiO$_3$ interface. The arrows in (c) mark an anti phase boundary (APB) along the (101) plane, where the atomic columns are shifted by half of the lattice period. The $a$-domain is about 3 nm wide. The lattice plane in the $a$-domain is strongly deflected within the 3 nm band because of the large tetragonality of PbTiO$_3$ (1.055). The HAADF-STEM image of figure 6(c) clearly shows that the lattice rotation takes place across the 90° domain wall, and that it stems from the lattice rotation around the core of dislocations. The 90° domain wall is almost parallel to the (101) plane, as determined theoretically from the strain-compatible condition between the adjacent domains in the tetragonal symmetry [28].

The HRTEM image of figures 6(a) and (b) shows a weak contrast at the PbTiO$_3$/SrTiO$_3$ interface. The HAADF-STEM image of figure 6(c) clearly shows the relationship between the interface and the dislocations. In an early study, Stemmer et al reported that in the PbTiO$_3$/SrTiO$_3$ heteroepitaxial system, the misfit dislocation is often the edge-type perfect dislocation with the Burgers vector $\mathbf{b} = a[100]$ [1]. Figure 6(d) shows that a misfit dislocation without an $a$-domain is the perfect dislocation with the Burgers vector $\mathbf{b} = a[001]$ corresponding to a side of the unit cell. The extra double-plane is composed of a pair of Pb–O and Ti–O layers. However, the misfit dislocation with an $a$-domain in figures 6(a)–(c) is dissociated into a couple of edge-type partial dislocations at the bottom end of the $a$-domain (No. 1 and No. 2). Furthermore, additional edge-type partial dislocations No. 3 and No. 4 are introduced after the $a$-domain growth. This result implies that the edge-type dislocation with the Burgers vector $\mathbf{b} = a[001]$ was introduced and also dissociated into a pair of edge-type partial dislocations. Extra half-planes of Pb–O or Ti–O layers are inserted at the substrate and the left sides parallel to (100) and (001) planes, respectively. Considering the contrast of the atomic columns in figure 6(c), extra half-planes of Ti–O are inserted in the partial dislocations No. 1 and No. 3, and those of Pb–O are in the partial dislocations No. 2 and No. 4. Figure 6(b) also shows that the extra half-plane of the partial dislocation No. 1 includes two Pb–O atomic columns above the interface. Then, these two Pb–O atomic layers, i.e. one unit cell layer of PbTiO$_3$ (about 0.4 nm) just above the interface, are strained compressively. The extra half-plane of the partial dislocation No. 2 is distinct from a three-unit-cell layer of PbTiO$_3$ (about 1.2 nm) immediately above the interface.

The atomic arrangement on the (101) plane between these partial dislocations is shifted by half of the PbTiO$_3$ unit cell. In our observations, all the $a$-domains immediately above the PbTiO$_3$/SrTiO$_3$ interface are accompanied with two pair of partial dislocations with an APB.

### 3.3.2.1 Strain map around the 90° domain and misfit dislocation.

Figure 7 shows the GPA results for figure 6(a), namely, $\varepsilon_{xx}$ (a), $\varepsilon_{yy}$ (b), $\varepsilon_{xy}$ (c) and $\omega_{xy}$ (d); panels (e)–(h) present the line profiles taken along the lines in the corresponding maps (a)–(d).

**Figure 7.** GPA of figure 6(a) around an $a$-domain near the film/substrate interface, namely $\varepsilon_{xx}$ (a), $\varepsilon_{yy}$ (b), $\varepsilon_{xy}$ (c) and $\omega_{xy}$ (d) maps. The insets in (a)–(d) magnify strain fields around dislocations. Panels (e)–(h) present the line profiles taken along the lines in the corresponding maps (a)–(d).
clearly illustrates the \(a\)-domain. The \(a\)-axis of the \(c\)-domain is almost equivalent to the SrTiO\(_3\) substrate; the \(c\)-axis of the \(a\)-domain fluctuates locally. The strain field \(\varepsilon_{xx}\) around the partial dislocation corresponds to that of the dislocation No. 1 and No. 2. The tensile side of the strain field \(\varepsilon_{xx}\) around the partial dislocation is coupled with the \(a\)-domain. The \(\varepsilon_{yy}\) map shows that the lattice parameter along the \(a\)-axis in the \(a\)-domain is almost the same as in the SrTiO\(_3\) substrate, and it fluctuates locally. The strain field \(\varepsilon_{yy}\) around the partial dislocation corresponds to that of the dislocation No. 3 and No. 4. The compressive side of the strain field \(\varepsilon_{yy}\) is coupled with the \(a\)-domain. The \(\varepsilon_{xy}\) map shows no strain around the \(a\)-domain, but around the misfit dislocation. There is no elastic interaction between the \(a\)-domain and the dislocations through the sharing strain. The \(\omega_{xy}\) map shows that the lattice of the \(a\)-domain rotates counterclockwise against the \(c\)-domain and the substrate. The lattice rotation around the dislocations is coupled with that of the \(a\)-domain. In these maps, the fluctuation of the measured strain is large due to the contribution of defects such as the \(a\)-domain, dislocations and the interface.

4. Discussion

4.1. Elastic interaction between the 90° domain and misfit dislocation

Figure 6(b) shows that the partial misfit dislocations No. 1 and No. 2 have extra half-planes in the direction towards the substrate, because of the smaller \(a\)-axis of SrTiO\(_3\) than PbTiO\(_3\) at the film deposition temperature. The partial dislocations No. 3 and No. 4 have no relationship with the lattice mismatch between the film and the substrate. However, the partial dislocations No. 3 and No. 4 are necessary for lattice matching between the \(a\)- and \(c\)-domains by lattice rotation.

Figure 7(a) shows that the strain field \(\varepsilon_{xx}\) around the partial dislocations is compressive and tensile in nature at the substrate and surface side, respectively. The tensile side of the normal strain \(\varepsilon_{xx}\) and the compressive side of the normal strain \(\varepsilon_{xy}\) around the misfit dislocations agree well with those of the \(a\)-domain. These normal strain components orient the principal axis (the direction of maximum and minimum values) of the edge dislocations. On the other hand, figure 7(d) shows that this axis rotates clockwise by 45°, i.e. it orient to [101] direction. This means that the obtained lattice rotation component is composed with that of each partial dislocation, i.e. the No. 1 and No. 3, and the No. 2 and No. 4. The lattice rotation \(\omega_{xy}\) around the dislocations coincides well with those of the \(a\)- and \(c\)-domains. These results show that the introduction of the edge-type dislocation with the Burgers vector \(b = a[001]\) is inevitable to relax the lattice mismatch accompanied with the nucleation and growth of the \(a\)-domain. Consequently, the strain fields \(\varepsilon_{xx}\) and \(\varepsilon_{yy}\) of the \(a\)-domain interact attractively with those of misfit dislocations through the lattice rotation \(\omega_{xy}\).

4.2. Nucleation and growth mechanism of the 90° domain

Theis and Schlom proposed that the \(a\)-domain of the PbTiO\(_3\) thin film nucleates from the step edge on the SrTiO\(_3\) substrate [29]. However, the nucleation site of the \(a\)-domain has no clear relationship with the step edge in our experiments. Here, we consider the nucleation and growth mechanism of an \(a\)-domain in the PbTiO\(_3\)/SrTiO\(_3\) epitaxial thin film based on figures 8 and 9.

Figure 8 shows the temperature dependence of the lattice parameter and of the lattice mismatch between PbTiO\(_3\) and SrTiO\(_3\) [1]. Figure 9 shows a schematic model of the formation of an \(a\)-domain at a misfit dislocation. The lattice mismatch (1.3%) is relaxed at 600 °C by introducing the misfit dislocation with the Burgers vector \(b = a[100]\) (figures 9(a) and (b)). An extra double-plane is inserted at the SrTiO\(_3\) side. Figure 6(d) shows that the misfit dislocation introduced at a high temperature remains at room temperature, based on the direction of the extra double-plane.

Upon cooling to the phase transition temperature of 490 °C, because of the thermal mismatch between PbTiO\(_3\) and SrTiO\(_3\), the mismatch of PbTiO\(_3\) in the \(a\)-axis direction decreases to −0.5% of the 600 °C value (figure 8(b)). Then, the tensile stress develops in the PbTiO\(_3\) layer (figure 9(c)). In particular, the tensile stress is concentrated around the misfit dislocation and is accompanied with the tensile strain field at the surface side. The \(a\)-domain has a larger lattice parameter than the \(c\)-domain in the in-plane direction, which means that the nucleation of the \(a\)-domain can compensate for the tensile strain field in the in-plane direction (figure 9(d)).
Figure 9. Schematic model of the formation of an \textit{a}-domain at a misfit dislocation. (a) Ideal PbTiO$_3$ and SrTiO$_3$ lattices. (b) After contacting PbTiO$_3$ and SrTiO$_3$ at 600 °C, a misfit dislocation is formed with an extra double-plane at the substrate side. (c) Cooling to below 490 °C introduces tensile stress at the PbTiO$_3$ side indicated by arrows. (d) Tensile stress around a misfit dislocation induces the nucleation of an \textit{a}-domain. The arrows show tensile stress resulting from the thermal strain. (e) Introduction of additional edge-dislocation with \( b = [001] \), and dissociation of both of the dislocation into two pairs of partial dislocations with an APB as a result of the \textit{a}-domain growth.

Consequently, the strain field of the misfit dislocation interacts attractively with that of the \textit{a}-domain.

Here, we have focused on the lattice rotation around dislocations. The lattice rotation angle at the left side of the dislocation in figure 6(d) is about 3°. This value is close to the tilt angle of the \textit{a}-domain owing to the tetragonality expressed as: \( \alpha = 2 \arctan(c/a) - 90° \) \cite{6}; for \( c/a = 1.055 \), \( \alpha = 3.1° \). The lattice rotation around the misfit dislocation has an advantage for the nucleation of the \textit{a}-domain.

Thus, the tensile strain arising from the lattice mismatch and thermal mismatch strains function as a driving force for the \textit{a}-domain nucleation and growth, and the lattice rotation around the edge-type mismatch dislocation triggers this nucleation.

At the initial stage, the misfit dislocation of the PbTiO$_3$/SrTiO$_3$ system is a perfect dislocation with the Burgers vector \( b = a[100] \), as shown in figure 6(d). Then, the additional edge-dislocation with the Burgers vector \( b = a[001] \) is also introduced for the lattice rotation around the \textit{a}-domain. After that, these full dislocations dissociate into a pair of partial dislocations with an APB under the process of growth of the \textit{a}-domain, as shown in figures 6(a)–(c). The APB comprises a bottom boundary of the \textit{a}-domain parallel to the (101) plane (figure 9(e)). The APB has excess energy, according to its width, which inhibits the growth of the \textit{a}-domain. Consequently, the nano sized \textit{a}-domain is surrounded by domain walls and an APB. A pair of partial dislocations with an APB would inhibit the further growth and domain switching of a nano sized \textit{a}-domain.

The mechanism of \textit{a}-domain nucleation at a misfit dislocation agrees well with the phase-field simulation of the PbTiO$_3$/SrTiO$_3$ system \cite{9}. However, our results differ from those obtained through simulation in three important points: firstly, the \textit{a}-domain nucleates on a perfect dislocation located at the PbTiO$_3$/SrTiO$_3$ interface; however, it would not make surface contact with the interface; secondly, the additional edge-type dislocation with the Burgers vector perpendicular to the misfit dislocation; finally, the dislocations dissociates into a pair of partial dislocations with the APB. Thus, our atomic-scale observation provides direct information on the elastic interaction between an \textit{a}-domain and a misfit dislocation, and on the domain nucleation and growth mechanism.

5. Conclusions

We studied the elastic interaction between the 90° domain and misfit dislocation in PbTiO$_3$/SrTiO$_3$ (001) epitaxial thin films using aberration-corrected HRTEM and conventional HAADF-STEM techniques combined with the GPA. The films show a complex diffraction contrast originating from the usual 90° domain, the nano sized 90° domain at the bottom regions, the 180° domain, and the misfit and threading dislocations. Quantitative information was extracted from strain mapping by GPA from the atomic-resolution HRTEM images. We have established that the nano sized 90° domain stems from two pair of partial dislocations, and that its bottom end is an APB. In particular, we imaged and analyzed the nanoscale strain fields around an \textit{a}-domain and two pair of partial misfit dislocations. The results elucidated the elastic interaction between the \textit{a}-domain and the partial misfit dislocations, showing that they attract each other and
that the elastic interaction would cause domain pinning at the misfit dislocation. In particular, the lattice rotation around the misfit dislocation would trigger the nucleation of the \( \alpha \)-domain under the driving force thermal stress. Based on these results, it has been proposed that the novel mechanism for the nucleation and growth of 90° domain in PbTiO\(_3\)/SrTiO\(_3\) epitaxial system. Firstly, 90° domains nucleate at misfit dislocations through the lattice rotation. Then, during the growth of the \( \alpha \)-domain, an additional full dislocation is introduced for the lattice matching between the \( \alpha \)- and \( c \)-domains through the lattice rotation. After that, two full dislocations dissociate into two pair of partial dislocations with an APB.

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