Effect of Growth Defects on Microwave Properties in Epitaxial Ba_{0.5}Sr_{0.5}TiO₃ Thin Films Grown on (001) MgO by Pulsed Laser Deposition

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Abstract

$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ (BSTO) films have been grown heteroepitaxially on (001) MgO substrates by pulsed laser deposition (PLD) to fabricate microwave phase shifters for the wide frequency range 45 MHz – 50 GHz. Both as-grown and ex-situ annealed films have a cube on cube epitaxial relationship with $\langle 100 \rangle \text{BSTO} // \langle 100 \rangle \text{MgO}$. Threading dislocations are the dominant defects, mostly with Burgers vectors $\mathbf{b} = \langle 101 \rangle$. Growth at 10^{-1} mbar oxygen pressure resulted in significantly better properties than that at 10^{-4} mbar. Ex-situ annealing of the film grown at 0.1 mbar resulted in a reduction of 40% in threading dislocation density and a 40% increase in dielectric tunability.

Keywords: Ferroelectric thin films; TEM; dislocations; tunability

Introduction

Perovskite ferroelectric films are suitable for frequency-agile microwave applications [1, 2]. For the practical operation of tunable devices, a high tunability of the permittivity and a low dielectric loss are essential. The dielectric loss and tunability of epitaxial thin films are generally much worse than those of bulk BSTO [3]. Dislocations are the common line defects found in ferroelectric thin films [4 - 7]. The highly localized strain fields associated with the misfit dislocations (MDs) may suppress ferroelectricity, due to the strong coupling of the polarisation to the strain; this results in polarisation gradients and thus an internal electric field which degrades the ferroelectric properties [8].

BSTO thin films can be grown epitaxially on a single crystal substrate by PLD: this allows good stoichiometric transfer of the cationic species from the target to the substrate [9]. Magnesium oxide is a suitable substrate for microwave applications because of its low permittivity and low loss [10]. However, the large lattice mismatch ($\sim 6\%$) and the difference in thermal expansion lead to the formation of defects such as dislocations and low angle grain boundaries [11]. The existence of these defects will deteriorate the tunability and increase the loss of the films [12]. Some attempts have been made to reduce the number of defects [12 - 14], but the exact effect of the defects on the dielectric behaviour has not been completely clarified. The manipulation of dislocation density is one of the key issues for achieving a high tunability and a low loss.

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In this work, we varied the oxygen pressure during film growth and used post-deposition annealing to influence the dislocation density in BSTO films grown on (001) MgO. The defects in the different films were studied by transmission electron microscopy and correlated with the microwave properties.

Experiment

Epitaxial BSTO thin films with a nominal thickness of 350 nm were grown on a (001) MgO substrate by PLD. A laser fluence of 1.5 J cm^{-2} at 5 Hz pulse rate was used. The oxygen pressure for the deposition ranged from 10^{-4} to 10^{-1} mbar. After growth all films were soaked in 800 mbar of static oxygen for one hour at 650°C . Additional ex-situ annealing in a flowing oxygen furnace at 1150°C for 24 hours was performed on some selected samples. For measurements of the dielectric properties, silver co-planar transmission lines were patterned onto the top surface of the films. The scattering parameters for microwaves (2-40 GHz) propagating along the lines were measured in a variable-temperature probe station under dc bias fields from $0\text{-}1.6 \text{ V } \mu\text{m}^{-1}$. The sample patterning and extraction of the dielectric properties of the films are described in detail elsewhere [15]. The use of co-planar lines means that the measured dielectric response is dominated by the in-plane properties.

To examine the crystallinity and epitaxial growth, θ - 2θ , asymmetric ω - 2θ and ω scans were performed in a six-circle X-ray diffractometer using $\text{CuK}\alpha$ radiation. For defect analysis, the film was examined by TEM in both cross section and plan view. Plan view and cross-sectional transmission electron microscopy (TEM) specimens were prepared by the tripod polishing technique [16]. The TEM specimens were thinned by mechanical grinding using diamond discs with a particle size from $15 \text{ }\mu\text{m}$ to $0.5 \text{ }\mu\text{m}$, followed by syton silicon colloidal suspension polishing down to electron transparency. The tripod wedge polishing method was chosen over ion milling because it gives a larger thin area, avoids ion contamination of the sample and causes less chemical smearing of the specimen surface. The TEM investigations were carried out using a Tecnai F20 microscope operated at 200 kV.

Results

X-ray diffraction scans showed that all of the BSTO films had good crystallinity and had grown epitaxially on top of the (001) MgO. Table 1 summarises the in-plane and out-of-plane lattice parameters and full width at half maximum (FWHM) of the (002) rocking curves. Compared with the BSTO target, the films grown at 10^{-4} mbar have a larger unit cell than the films grown at 10^{-1} mbar. The films grown at 10^{-4} mbar, have an out-of-plane lattice parameter larger than the in-plane lattice parameter (i.e. the crystal structure is slightly tetragonal with the tetragonal axis parallel to the normal to the film). Films grown at 10^{-1} mbar showed an improved crystallinity which was further improved by ex-situ annealing. The slight in-plane tetragonality of these films as-grown was relieved by the ex-situ annealing.

The microwave properties of the BSTO films are summarised in Table 2; a detailed description is reported elsewhere [15]. The highest tunability was obtained for films grown at an oxygen pressure of 10^{-1} mbar; when such a film was ex-situ annealed, there was a further 40% increase in dielectric tunability as shown in Table 2. Ex-situ annealing of the films grown at 10^{-1} mbar also sharpened the transition from the ferroelectric to the paraelectric state compared with the as-grown films and reduced the temperature at which the permittivity obtained its maximum value. There was no hysteresis in the permittivity versus voltage curves at room temperature, although there was hysteresis on cooling to 80 K (i.e. on cooling into the ferroelectric state). Ex-situ annealing had little effect on the (poor) dielectric properties of the films grown at 10^{-4} mbar oxygen.

Figure 1a shows a cross-sectional bright field (BF) image of a BSTO film which was grown at an oxygen pressure of 10^{-4} mbar. The BSTO film is epitaxial on the (001) MgO substrate (e.g. figure 1(b)) but with a 4° to 5° mosaic spread measured from the angular spread of the diffraction spots as marked in figure 1(b). The film has a uniform thickness of 350 nm and a sharp interface with the substrate. Threading dislocations (TDs) run from the interface to the top of the film. In addition to the TDs, figure 1(c) shows vertical and horizontal moiré fringes. The mismatch stress has been relaxed largely but not completely by the formation of misfit dislocations (MDs) in the film at the interface. Figure 2 shows a high resolution image of the same film. The interface contains irregularly spaced misfit dislocations whose average separation is about 4 nm. The sample was unstable under the electron beam due to charging of the substrate, but, from the best Fast Fourier Transformed (FFT) HRTEM image (Figure 2b), it was possible to measure the projected Burgers vector (**b**) of dislocation 1. In working out the Burgers circuit it was assumed that: $[100]_{\text{BSTO}} \approx [100]_{\text{MgO}}$. Then, denoting the thickness of the interface as x , the number of atom planes involved was counted as shown by the dots in figures 2. $\mathbf{b} = 5[001]_{\text{BSTO}} - 9[100]_{\text{BSTO}} - 4.5[001]_{\text{BSTO}} - x - 4[001]_{\text{MgO}} + 8.5[100]_{\text{MgO}} + 4[001]_{\text{MgO}} + x = \frac{1}{2} [\bar{1} 0 1]$.

Figure 3 shows a cross-sectional BF image of an as-grown BSTO film grown at an oxygen pressure of 10^{-1} mbar oxygen. The density of TDs, of the order of 10^{10} cm^{-2} , in this growth condition is similar to that in the film grown at 10^{-4} mbar but there are no vertical and horizontal moiré fringes as were seen in figure 1(c); these figures were only observed in the film grown at low oxygen pressure. This suggests an absence of low angle boundaries in the films grown at 10^{-1} mbar. Comparing Figures 3a and b, most of the TDs are in contrast both when $g = 002$ and $g = 200$ which means the threading dislocations are not pure edge or screw type. The contrast of the TDs is clearer when $g = 002$. Figure 4 shows the contrast experiment carried out on the TDs. The results of the contrast experiment are summarised in table 3. Most of the dislocations have Burgers vectors which are in a $\langle 101 \rangle$ direction where $\langle 101 \rangle$ is not lying in the interface (thus $[101]$, $[10\bar{1}]$, $[011]$ and $[0\bar{1}1]$, but not $[110]$ or $[\bar{1}\bar{1}0]$). They are probably $\langle 101 \rangle$. Due to the high density of TDs and unclear contrast in several g vectors, the exact **b** was difficult to determine.

The HRTEM image in figure 5 reveals MDs in the interface plane. The MD separation distances are quite regular compared to those in the film grown at 10^{-4} mbar and have an average separation of 3 nm. Burgers circuits drawn around MD 1 show that the projected Burgers vector is $\mathbf{b}_1 = 5[001]_{\text{BSTO}} - 8[100]_{\text{BSTO}} - 5.5[001]_{\text{BSTO}}$

- $x - [001]_{\text{MgO}} + 7.5 [100]_{\text{MgO}} + 2[001]_{\text{MgO}} + x = \frac{1}{2} [\bar{1}01]$. MD 2 is a dislocation with projected $\mathbf{b}_2 = 4[001]_{\text{BSTO}} - 6[100]_{\text{BSTO}} - 3[001]_{\text{BSTO}} - x - 2.5[001]_{\text{MgO}} + 5.5 [100]_{\text{MgO}} + 1.5[001]_{\text{MgO}} + x = \frac{1}{2} [\bar{1}00]$.

Figure 6 shows a cross-section image of the ex-situ annealed film grown at 10^{-1} mbar. The density of TDs is drastically reduced by the ex-situ annealing as compared with the as-grown film illustrated in figures 3-5. Most of the TDs are out of contrast when $g = 002$ as shown in figure 6 a, b and c. However, the density of the pure edge or mixed $\langle 101 \rangle$ type TDs is similar with and without ex-situ annealing as shown in figures 3b and 6b when the TD densities are compared using $g = 200$. Using the $g \cdot b = 0$ criterion, it can be concluded that the TDs which have a Burgers vectors with a screw component were annihilated during the ex-situ annealing process, while the edge dislocations with Burgers vector component = $[100]$ or $[010]$ still exist in the ex-situ annealed sample. The better crystallinity compared with the films grown at 10^{-4} mbar is evidenced by the inset SAD pattern in figure 6b: a smaller mosaic spread is recorded when compared to the film grown at 10^{-4} mbar oxygen pressure. This is consistent with the X-Ray diffraction data presented in Table 1. The HRTEM image shown in figure 7 shows a regular array of MD's with a separation similar to that observed in the as-grown films (Figure 5). In the films grown at 10^{-1} mbar oxygen pressure, the interface dislocations are, unlike the TDs, in good contrast when $g = 200$ as shown in figure 8c. The as-grown films deposited at 10^{-4} mbar have a slightly smaller MD density (MD separation ~ 4 nm, Figure 8a) than the films grown at high oxygen pressure (average MD separation ~ 3 nm, Figure 8c). Ex-situ annealing did not change the density of MD's. However, the interfaces decorated with MDs appear different after ex-situ annealing. Ex-situ annealing gave a curved interface as shown in figure 8b and 8d while the as-grown films have straight interfaces as shown in figure 8a and 8c. Moiré fringes are only observed in the as-grown film grown at 10^{-4} mbar (figure 8a). After ex-situ annealing of the films grown at 10^{-4} mbar, some recrystallisation of the film occurs leading to higher angle grain boundaries (figure 8b).

Figures 9a and 9b show plan view bright field images of the top surface of the as-grown and ex-situ annealed films grown at 10^{-1} mbar under near two beam conditions, with $g = 110$. The plan view clearly shows a decrease in dislocation density due to the ex-situ annealing. The densities of TDs reaching the top surface of the BSTO films before and after ex-situ annealing are $3.0 \times 10^{10} \text{ cm}^{-2}$ and $1.7 \times 10^{10} \text{ cm}^{-2}$ respectively. There is thus a 40% reduction in TD density due to the ex-situ annealing.

Discussion

1) Epitaxy and crystallinity

Both X-ray diffraction and TEM results show that the films grown at 10^{-1} mbar have a good quality of epitaxial growth and crystallinity (Table 1 and the inset in figure 6b) and that ex-situ annealing further improves the crystallinity (Table 1). Strains in the ex-situ annealed film grown at 10^{-1} mbar are fully relaxed as indicated by the calculation of the lattice mismatch from the x-ray diffraction measurements. The FWHM of the ex-situ annealed sample implies excellent crystallinity of the film.

2) Low angle grain boundaries

In films grown at 10^{-4} mbar, moiré fringes are observed and some of them may be caused by low angle grain boundaries (LAB) (Figure 1b and 8a). The moirés are only observed in the films grown at 10^{-4} mbar. The existence of low angle boundaries between columns in films has been associated with low tunability [12]. Previous work has shown the area close to a LAB to have a crystal structure different from that of a relaxed structure, since the boundary usually has altered composition and bond lengths [17, 18]. As shown in Table 2, the dielectric tunability of the films grown at 10^{-4} mbar is five times smaller than that as-grown film grown at 10^{-1} mbar. The high density of LABs may not be the only source of the degradation in tunability in films grown at 10^{-4} mbar oxygen pressure; these films are also more strained and are likely to be oxygen deficient.

3) Misfit dislocations

The film-substrate interface of all the films has atomic sharpness without any interfacial reaction layer (Figures 2, 5, 7 and 8). The interfaces of the as-grown films are straight while those after ex-situ annealing become wavy regardless of the growth pressure. The MD separation in the films grown at 10^{-1} mbar (3 nm) is smaller than in the film grown at 10^{-4} mbar oxygen pressure (4 nm) (Figures 2, 5 and 8), irrespective of ex-situ annealing. This is consistent with the expected larger lattice parameter in oxygen deficient films, leading to a smaller mismatch between the film and substrate. Normally, it is expected that $[100]$ and $[010]$ type pure edge misfit dislocations relieve the stress in the film arising from the mismatch of the lattice parameters of the film and substrate. The mismatch between bulk BSTO and MgO is of the order of 6.6 % which would produce one pure edge type MD in every 15 unit cells of MgO, i.e. every 6 nm. However, the measured average separation of MDs is smaller in all growth conditions. Therefore the MDs formed at the interface cannot be pure edge type dislocations. Indeed, from the Burgers circuit analysis, the MDs have projected Burgers vectors equal to $\frac{1}{2} \langle 101 \rangle$ are shown in figures 2b and 5. The MDs do not have $\mathbf{b} = \frac{1}{2} [110]$ or $\frac{1}{2} [1\bar{1}0]$, vectors which would accommodate the misfit more efficiently. It is known that $[100]$ misfit dislocations may dissociate into two partial dislocations according to the reaction $[100] \rightarrow \frac{1}{2}[101] + \frac{1}{2}[10\bar{1}]$ in order to decrease the dislocation energy [4] i.e. one edge dislocation may split into two partials incorporating a Burgers vector component $[001]$. Such a component would not relieve any mismatch strain. All of the films grown under the different conditions have similar arrays of MDs which suggests that the substrate is important in determining the $\mathbf{b} = \frac{1}{2} \langle 101 \rangle$ nature of the MDs. It is known that formation of partial dislocations can be associated either with the perimeter of monolayer-height islands that nucleate during growth or with the steps on a miscut substrate [19]. The projected vector of the smallest step in MgO is $\frac{1}{2} \langle 101 \rangle$. If $[001]$ BSTO is deposited on such a $\frac{1}{2} [101]$ step, the resultant mismatch vector is: $[001]_{\text{BSTO}} - \frac{1}{2}[101]_{\text{MgO}} \approx \frac{1}{2}[\bar{1}01]_{\text{BSTO}}$. In this case a dislocation will be formed with Burgers vector $\mathbf{b} = \frac{1}{2}[\bar{1}01]_{\text{BSTO}}$. This dislocation has a component along $[001]$ and examples are shown in figures 2b and 5 which suggest

that $\mathbf{b}$ is indeed equal to $\frac{1}{2}[\bar{1}01]_{\text{BSTO}}$. Thus $\mathbf{b}$ reflects a translation in the substrate lattice rather than that of the film.

An array of such defects with $\mathbf{b} = \frac{1}{2} \langle 101 \rangle$ would accommodate the misfit if the spacing were sufficiently small, and the formation of them might be preferred to the array of MDs with $\mathbf{b} = \frac{1}{2} \langle 110 \rangle$ if the initial density of steps on the MgO surface were sufficiently high. However, to relax fully the misfit strain, the separation of MDs with $\mathbf{b} = \frac{1}{2} \langle 101 \rangle$ would need to be only 2 nm, while the observed separation is 3 – 4 nm. The relaxation of the misfit strain is thus not achieved fully by this type of MD.

MD's probably do not contribute significantly to the microwave dielectric properties of these films, because the films are of the order of 300 nm in thickness yet the MD's are located at the film-substrate interface. Furthermore, in studies which evidenced a correlation between MD's and dielectric response [20], localised charges on the cores of the misfit dislocations were associated with hysteresis in the capacitance-voltage characteristics: no such hysteresis is seen in the present films [15].

4) Threading dislocations

If the number of MDs is not sufficient to relieve the strain, TDs are formed by the glide of half-loops generated in response to the remaining misfit stresses at the interface. The configuration which consists of an MD segment and two TD segments that thread from the interface to the deposit surface as described by Matthews [22]. The dislocations mostly reported for the cubic SrTiO_3 perovskite structure have $\mathbf{b}$ of the $\langle 100 \rangle$ or $\langle 110 \rangle$ types at room temperature [23]. In BaTiO_3 , which has a tetragonal perovskite structure at room temperature, $\langle 110 \rangle$ dislocations are also frequently observed [24]. In the present case, the TDs in the BSTO film are mostly $\langle 101 \rangle$ type. Table 3 provides a summary of the possible Burgers vectors of the TDs. The density of the TDs is too high for it to be likely that these defects originate in the substrate and are then copied to the film. The Burgers vectors of some of the TDs are not the same as the MDs (e.g. $[110]$, or $[011]$), which means they are not formed by the forcing out of MDs during the coalescence of island growth nuclei. As mentioned in the previous section, it is believed that the mismatch stress has not been fully relaxed by the MDs: the origin of these TDs may therefore be the introduction of half-loops of dislocation at the deposit surface in response to the residual misfit. The half-loops would expand to the interface giving the misfit-relieving segments.

The TD density is more or less the same in films grown at both 10^{-4} mbar and 10^{-1} mbar (Figures 1 and 3). Ex-situ annealing reduces the density of TDs in the 10^{-1} mbar film by 40 % (e.g. fig. 9). Figures 1a, 3a and 6 show dislocations in strong contrast with $g = 002$ indicating the existence of a $[001]$ component, which is the screw component for dislocations parallel to the film normal. Tilting experiments suggest that the TDs left after ex-situ annealing are mainly of edge or mixed type. The screw type TDs, which have higher mobility, are annihilated during the ex-situ annealing process. For the half-loops to be glissile, $\mathbf{b}$ should be $\langle 101 \rangle$ and indeed most of the TDs have $\mathbf{b} = \langle 101 \rangle$ although some of them are ambiguous (Figure 4 and Table 3). The dislocation annihilation may be due to the interaction between two threading segments and shrinkage of the resulting half-loops. The average spacing of the TD segments in our films is less than 50 nm. This small distance between the TDs will

create significant interaction forces between the threading segments. There are two cases to consider [25]: when the coplanar half loops have opposite $\mathbf{b}$ they will constrict and annihilate, leaving two half-loops shrinking towards the substrate interface and film surface; when, two TDs have opposite $\mathbf{b}$ components parallel to $\pm[001]$ but a different component in the plane they will form two half loops and a constriction segment with a smaller $\mathbf{b}$. Under such circumstances, TDs formed with other values of $\mathbf{b}$ are also common because of the strong interaction. Dislocations 1, 2, 4 in figure 6 a show the half-loop shrinking towards the film surface while dislocation 3 is shrinking towards the interface.

Because of the small distance between the TDs, the magnitudes of the elastic interactions between adjacent TDs will be significant. The overlapping of stress fields may cause local variation of the polarisation and hence internal electric fields strong enough broaden and shift the ferroelectric transition and so reduce the permittivity and tunability at a particular temperature of measurement [8, 13]. The decrease in TD density due to ex-situ annealing is correlated with an increase in the permittivity, tunability and dielectric loss and also a sharpening of the ferroelectric to paraelectric transition (i.e. a reduction of heterogeneity) [15]. Since the TDs are present throughout the volume of the film, it is more likely that they, rather than the MDs, are influencing the dielectric properties.

5) Point defects

Although our TEM studies provide no direct evidence of the presence of point defects in our films, they may nevertheless be present in large numbers. Oxygen vacancies, the most common point defects in titanate [26], may cause ferroelectric polarisation in a very local area and indeed a Raman study of the lattice-dynamical properties of $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ films has provided evidence for the existence of polarised nanoregions [26]. Thus, a large number of point defects would decrease the tunability of the films. We would expect the films grown at low oxygen pressure to have more oxygen vacancies and thus be less tunable (Table 2).

Conclusion

In heteroepitaxial BSTO films, point defects, low angle grain boundaries, threading dislocations and misfit dislocations may all be present, depending on the growth conditions. The films with the best microwave properties have the lowest density of threading dislocations.

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Table 1. Summary of out-of-plane and in-plane lattice parameters and the FWHMs of BSTO films grown at different oxygen pressures and after ex-situ annealing. In the last two columns, the lattice parameters are compared with the lattice parameters of the bulk target BSTO ($a = 0.39531 \text{ nm}$). The lattice parameter of MgO is 0.4212 nm .

	Oxygen Pressure (mbar)	FWHM	In-plane lattice parameter (nm)	Out-of-plane lattice parameter (nm)	Strain of the in-plane lattice parameter (%)	Strain of the out-of-plane cell parameter (%)
As grown	10^{-4}	0.936°	0.3976 ± 0.0004	0.3995 ± 0.0003	0.5717	1.0700
As grown	10^{-1}	0.866°	0.3959 ± 0.0001	0.3949 ± 0.0001	0.1482	-0.1161
Ex-situ annealed	10^{-1}	0.265°	0.3960 ± 0.0001	0.3955 ± 0.0001	0.1829	0.0604

Table 2. Summary of microwave properties of BSTO films grown at different oxygen pressures and annealing conditions. The “effective loss tangent” $= \frac{\epsilon''_{eff}}{\epsilon'_{eff}}$, includes both

dielectric and conductor losses. The figure of merit is obtained from the ratio of the differential phase shift to the insertion loss of the co-planar waveguide transmission line devices [15].

	Oxygen pressure (mbar)	Tunability (%) at 20 GHz and in a bias field of $1.6 \text{ V}\mu\text{m}^{-1}$	Permittivity (ϵ') at 20 GHz	Effective dielectric loss (tan ξ_{eff})	Figure of Merit (deg dB^{-1}) at 20 GHz and $1.6 \text{ V}\mu\text{m}^{-1}$
As grown	10^{-4}	4.5	240	0.08	-
	10^{-1}	22	870	0.25	5.5
Ex-situ annealed	10^{-4}	Not tunable	N/A	N/A	N/A-
	10^{-1}	31	2000	0.3	7.2

Table 3. Result of contrast experiment: $\checkmark$, visible. $\times$ invisible.

Dislocations	g					Direction of b
	$00\bar{2}$	200	111	$\bar{1}11$	$\bar{2}0\bar{2}$	
1	$\checkmark$	$\times$	$\times$	$\times$	$\checkmark$	$01\bar{1}$ or $0\bar{1}1$
2, 14, 18, 21, 24	$\checkmark$	$\times$	$\checkmark$	$\checkmark$	$\checkmark$	$0\bar{1}\bar{1}$ or 011 or 001 or $00\bar{1}$
4, 9, 10,	$\checkmark$	$\checkmark$	$\checkmark$	$\times$	$\checkmark$	$\bar{1}0\bar{1}$
8	$\checkmark$	$\checkmark$	$\times$	$\checkmark$	$\times$	$\bar{1}01$
12, 19, 25	$\times$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	100 or $\bar{1}00$
16	$\checkmark$	$\checkmark$	$\times$	$\times$	$\times$	$10\bar{1}$
3, 5, 6, 7, 11, 13, 15,						Not clear

17, 20, 22, 23						
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For Peer Review Only

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Figure 1. (a) Cross-sectional bright field image of the as-grown film grown at 10^{-4} mbar taken with a beam direction near $[010]$ and a diffraction vector $g=002$. (b) Diffraction pattern from the interface between the film and substrate. (c) Dark field image showing moiré fringes.

Figure 2. Cross-sectional HRTEM image of interface of the as-grown film deposited at 10^{-4} mbar showing (a) the misfit dislocations marked by arrows ($BD \sim [010]$). (b) FFT image of misfit dislocations 1 and 2; Burgers circuit is drawn around dislocation 1; S and F are the start and finish points respectively.

Figure 3. Cross-sectional bright field image of the as-grown film deposited at 10^{-1} mbar taken with beam direction $[010]$ zone. (a) $g = 002$, $s > 0$, (b) $g = 200$, $s > 0$. The insets with dislocations numbered are taken at the Bragg condition with $s = 0$. The TD contrast is much stronger with $g = 002$.

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Figure 7. High resolution image showing the interface of the ex-situ annealed film grown at 10^{-1} mbar.

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Figure 9. Plan-view image near the surface of the film taken when $g = (110)$. Films grown at 10^{-1} mbar (a) as-grown film (b) ex-situ annealed.

Figure 1. (a) Cross-sectional bright field image of the as-grown film grown at 10^{-4} mbar taken with a beam direction near $[010]$ and a diffraction vector $g=002$. (b) Diffraction pattern from the interface between the film and substrate. (c) Dark field image showing moiré fringes.

33x15mm (600 x 600 DPI)

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21x41mm (600 x 600 DPI)

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31x61mm (600 x 600 DPI)

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19x12mm (600 x 600 DPI)

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141x42mm (600 x 600 DPI)

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41x11mm (600 x 600 DPI)

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41x12mm (600 x 600 DPI)**

Figure 4. Contrast experiment on dislocations in the as-grown film deposited at 10^{-1} mbar with dislocations numbered. The dark field images have higher and better resolution so are shown to help identify the visibility of the dislocations.
137x254mm (600 x 600 DPI)

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144x87mm (600 x 600 DPI)

Figure 6. Cross-sectional bright field image of the film grown under 10^{-1} mbar oxygen pressure followed by ex-situ annealing: (a) Low magnification BF image $BD \sim [010]$, $g = 002$ and dislocation half loops are marked as 1, 2, 3, 4, (b) $g = 00$. Inset is the SAD pattern of the interface taken at $BD \sim [010]$. c) Dark filed image of dislocations marked with numbers. Most of the dislocation are out of contrast in $g = 002$ and in contrast with $g = 200$.

45x13mm (600 x 600 DPI)

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52x18mm (600 x 600 DPI)

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49x20mm (600 x 600 DPI)

Figure 7. High resolution image showing the interface of the ex-situ annealed film grown at 10^{-1} mbar.

120x80mm (600 x 600 DPI)

Figure 8. Two beam bright field cross-section TEM image showing the interface, BD ~ [010]. $g = 200$. (a) as-grown film grown at 10^{-4} mbar, (b) ex-situ annealed film grown at 10^{-4} mbar, (c) as-grown film grown at 10^{-1} mbar; the inset to c is a dark field image showing a clear misfit dislocation contrast and (d) ex-situ annealed film grown at 10^{-1} mbar.
68x36mm (600 x 600 DPI)

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90x59mm (600 x 600 DPI)

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59x44mm (600 x 600 DPI)

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74x70mm (600 x 600 DPI)

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78x74mm (600 x 600 DPI)