Surface Termination of Oxide Single Crystal Substrates by Laser Annealing

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Abstract

Before thin film growth, oxide single crystal substrates must be treated to produce chemically homogeneous terraces with a uniform terrace height. It has been shown that laser annealing can achieve surface terminations similar to those achieved by conventional substrate preparation methods, with the advantage of being an *in situ* technique that does not use wet chemicals. Here we show that laser annealing substrates in an MBE chamber at a background pressure of 10^{-6} Torr of 10% O_3 yields uniform terrace heights on Al_2O_3 $(1\bar{1}02)$, Al_2O_3 $(11\bar{2}0)$, $TbScO_3$ (110), and $DyScO_3$ (110). Although the desired terrace structure was achieved on $SrTiO_3$ (001), further work is needed to minimize the effects of chemical reduction during the anneal. In addition, further optimization is necessary to achieve uniform terrace heights on Al_2O_3 $(10\bar{1}0)$ and Al_2O_3 (0001).

Introduction

The surface of the substrate plays an important role in the growth and properties of oxide thin films. To achieve high quality films and observe interface phenomena, atomically sharp interfaces and chemically homogeneous terraces are required.[1,2,3] Since as-received substrates do not meet these requirements, they must be prepared prior to film growth. Conventional substrate preparation involves a combination of etching in acidic solutions and annealing at around 1000 °C.[2] However, these techniques suffer from multiple issues, including limited chemical selectivity[2] and residue from chemical etching.[4] The etchants can also be hazardous (e.g. HF) and these conventional techniques are *ex situ*, meaning that there is little control over the surface of the substrate in the time between substrate preparation and film growth. To address some of these issues, a laser annealing substrate preparation method has been developed for a PLD chamber with a background pressure of 10^{-2} Torr of O_2.[5] This method is *in situ* and does not involve wet chemicals, minimizing the risk of substrate contamination. In this work, we show that a uniform terrace structure can be achieved using this laser annealing method with a background pressure of 10^{-6} Torr of 10% O_3, an accessible environment for the MBE at PARADIM.

Methods

The substrates were annealed in the PARADIM MBE chamber, using the 10.6 μm laser substrate heater. The background pressure was set to 10^{-6} Torr of 10% O_3. A representative temperature profile throughout the annealing process is shown in Fig. 1. The substrates were annealed at high temperatures for 200 seconds. The rate of temperature increase was lowered above 1000 °C to minimize the risk of cracking the substrates.

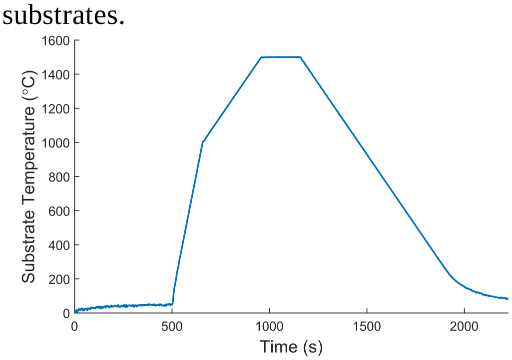

Fig. 1 Representative substrate temperature profile throughout the annealing process

Atomic force microscopy (AFM) in tapping mode at a scan rate of 3.91 Hz was used to characterize the surface morphology of the substrates before and after annealing. It should be noted that Al_2O_3 (0001) and $(10\bar{1}0)$ substrates were wiped with isopropanol and lint-free wipes before annealing to remove large particles. This was not necessary for the other substrates.

Results

The AFM scans of Al_2O_3 $(1\bar{1}02)$ before and after annealing are shown in Fig. 2. Although there is a terrace structure on this as-received substrate (Fig. 2a), the terrace edges are rough and there are small particles on the surface. After annealing (Fig. 2b), the terrace edges are smooth and there are no visible particles. Fig. 2c shows the line profile perpendicular to the terraces (Profile 1) and along a single terrace (Profile 2). Profile 1 shows an average terrace height of 0.34 ± 0.02 nm while Profile 2 shows that there is very little height variation along a single terrace.

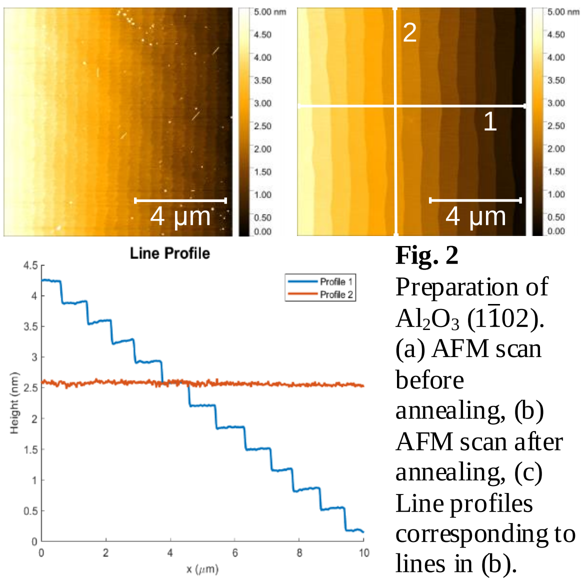

Fig. 2 Preparation of Al_2O_3 $(1\bar{1}02)$. (a) AFM scan before annealing, (b) AFM scan after annealing, (c) Line profiles corresponding to lines in (b).

A similar terrace structure after annealing was observed for Al_2O_3 $(11\bar{2}0)$ annealed at 1400 °C, $TbScO_3$ (110) annealed at 1470 °C, $DyScO_3$ (110) annealed at 1400 °C, and $SrTiO_3$ (001) annealed at 1400 °C. There was little height variation along a terrace, and the average terrace height was measured to be 0.27 ± 0.04 nm, 0.38 ± 0.03 nm, 0.39 ± 0.02 nm, and 0.41 ± 0.02 nm respectively.

When annealing $SrTiO_3$ at 1400 °C, the substrate reduces. Since this oxygen deficiency in $SrTiO_3$ cause electrical conduction,[6] further work must be done to minimize the reduction of $SrTiO_3$ during the annealing process.

Before annealing, Al_2O_3 $(10\bar{1}0)$ does not display any terrace structure. No change was observed after annealing for 200 seconds at temperatures below 1250 °C. After annealing at 1300 °C, terracing was observed, but multiple terrace heights were present, and annealing at temperatures above 1350 °C caused this terrace structure to disappear entirely. Therefore, more work needs to be done to achieve a uniform terrace height in the annealing of Al_2O_3 $(10\bar{1}0)$.

After annealing Al_2O_3 (0001) at 1500 °C, terracing was observed, with terrace heights corresponding to 1/6 of the unit cell height. When annealed at 1650 °C, many of the neighboring terraces fused together to form new terraces with heights corresponding to 1/3 of the unit cell height. Avoiding terrace heights that correspond to 1/6 of the unit cell is desirable, since this terrace structure can cause twinning in films.[7] Further work must be done to establish an annealing recipe that yields a surface that consists entirely of terraces with a height that corresponds to 1/3 of the unit cell.

Conclusions

In this project, it was shown that substrates can be prepared for thin film growth by laser annealing in an environment of 10^{-6} Torr of 10% O_3. The annealing recipes for Al_2O_3 $(1\bar{1}02)$, Al_2O_3 $(11\bar{2}0)$, $TbScO_3$ (110), and $DyScO_3$ (110) were determined, and further work must be done to establish the annealing recipes for $SrTiO_3$ (001), Al_2O_3 $(10\bar{1}0)$, and Al_2O_3 (0001).

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