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Screw-like structures in ex situ $Tl_2Ba_2CuO_6$ films

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Abstract. The growth of ex situ $Tl_2Ba_2CuO_6$ films on single-crystal $LaAlO_3$ was investigated using atomic force microscopy and X-ray diffraction. Most of the film surface consisted of flat areas separated by single or several unit-cell steps. This morphology suggests that the films grow predominantly by a step-flow mechanism. A Tl-2201 film grown using a precursor film deposited at 700 °C exhibited stacks of planes arranged in a screw-like manner on a flat background. Islands of 1–3 μm size and a height of 2–20 unit cells were dominant structures on the surface. Possible causes for the formation of the screw-like structures are discussed.

Understanding the growth of high-T_c superconducting films is important from the point of view of fundamental science as well as for applications. Recently, some interest has been directed towards the $Tl_2Ba_2CuO_6$ (Tl-2201) system in order to study the fundamental properties of high-T_c superconductors [1]. The reason for the interest in this compound is its apparent simplicity. Thus, the structure exhibits single CuO_2 planes, it can be prepared with tetragonal symmetry and it does not undergo phase transformations at low temperature.

In most Tl-cuprate thin film processes the superconducting film is formed by annealing a precursor film in Tl_2O vapor at high temperature. The formation of an epitaxial Tl-2201 film involves the diffusion of Tl_2O through the Ba_2CuO_x precursor film, causing nucleation of Tl-2201 at the substrate/precursor film interface. Film growth is expected to be influenced by precursor film properties as well as by temperature and Tl_2O(g) vapor pressure during synthesis.

1 Experimental

The Tl-2201 films were prepared on single crystal $LaAlO_3$ substrates by an ex situ process, treating thallium-free precursor films with Tl_2O(g) [2, 3]. The precursor films had a thickness of 150–200 nm and were deposited by laser ablation of a Ba_2CuO_x target. The precursor films were annealed in a lidded crucible made from polycrystalline Tl-2201, acting as the source of Tl_2O(g). The Tl_2O annealing involved heating the sample and the Tl_2O(g) source rapidly to 780 °C. The temperature was then increased by 2 °C/h to 800 °C. Subsequently, the sample was cooled by 60 °C/h to room temperature. Two Tl-2201 films, (1) and (2), were investigated, the precursor films being deposited at 300 and 700 °C, respectively. To study early stages of precursor film growth, 2 nm Ba_2CuO_x films were deposited at 200, 400, 600 and 800 °C. A Nanoscope III atomic force microscope (AFM) in contact mode was used for surface imaging. The measurements were carried out in air. Silicon nitride and silicon tips were used.

2 Results and discussion

X-ray diffraction showed that the Tl-2201 films were c-axis oriented. The omega scan of (0,0,10) had a FWHM of 0.33°. The onset of the transition to the superconducting state as measured by susceptibility was between 74 and 76 K for both films.

The surfaces of both Tl-2201 films consisted of flat areas with terraces separated by steps corresponding to one or several unit cells (Figs. 1a,b). Both films contained a small number of a-axis particles (Fig. 2a). These particles exhibited single (film 1) or several (film 2) unit cell steps, the particle thickness corresponding to 10–20 unit cells. Structures reminiscent of grain boundaries could be observed, also showing the presence of unit cell high steps (Fig. 2b).

Film 1 (precursor deposition temperature 700 °C) exhibited screw-like structures on the flat background, while no such features were present in film 2 (precursor deposition temperature 300 °C). Individual unit-cell-high “screw heads” on the flat terraces were the simplest screw-like structures on the surface of film 1 (Fig. 3a). The screw merges

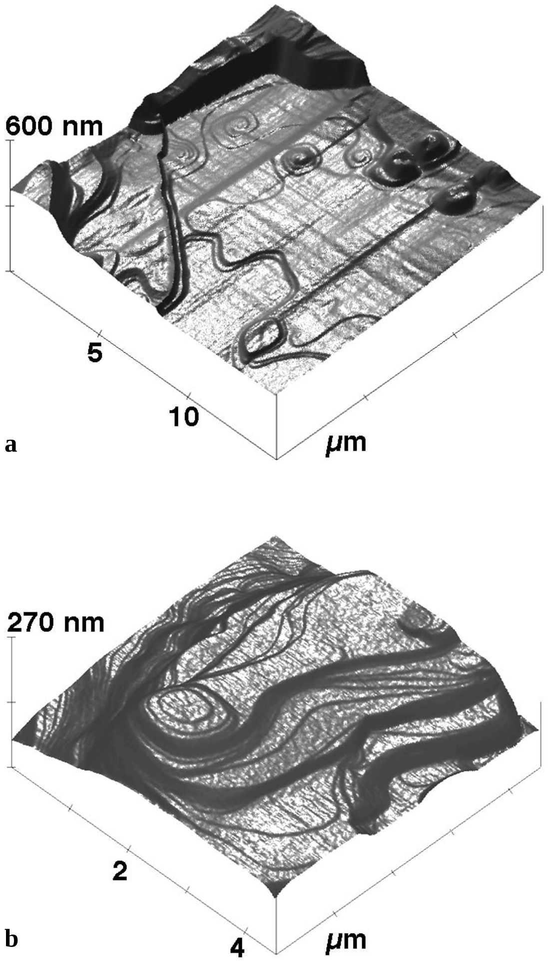

Fig. 1a,b. AFM images of film 1. **a** The surface consists of atomically flat terraces separated by steps of one or several unit cells. **b** Higher magnification showing a monolayer-high 2D island on a terrace

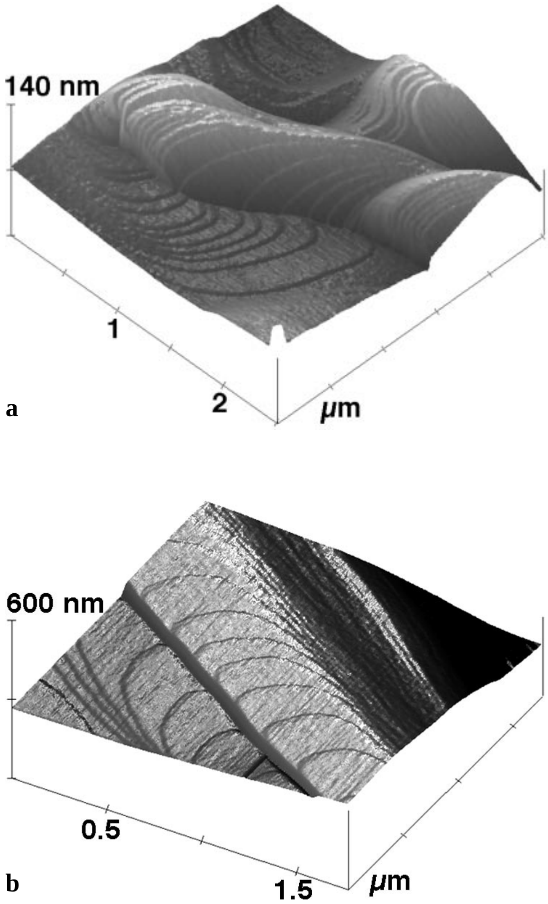

Fig. 2a,b. AFM images of film 1. **a** A few *a*-axis particles are seen, exhibiting single or several unit cell high steps. **b** Structures reminiscent of grain boundaries with unit cell high steps

with the terrace after about one turn. Screw-like structures forming islands of 1–3 μm diameter and a height of 2–20 unit cells were common (Figs. 3b–d). These structures grow in the (001) direction. On top of the islands, *c*-axis-oriented step trains were found, the steps being of single unit cell height.

The screw-like structures observed in film 1 exhibit some similarities with screws found in sputtered YBCO and BiSSCO films [4–6] and in melt-grown YBCO single crystals [7, 8]. The density of 3D islands in the Tl-2201 film is about $3–5 \times 10^6\,\mathrm{cm}^{-2}$, being approximately 1000 times less than the reported density of screws in sputtered $YBa_2Cu_3O_{7-x}$ films ($6 \times 10^9\,\mathrm{cm}^{-2}$ [4–6]). The screw-like structures observed in film 1 and in sputtered $YBa_2Cu_3O_{7-x}$ films exhibit unit-cell steps, while in melted YBCO samples the steps are much coarser, reaching 10–100 nm height or more [7, 8]. In YBCO films, the typical terrace width in screws is reported to be 20 nm [4–6]. In the Tl-2201 3D islands the terrace width reaches 200–300 nm. The diameter of the 3D islands in film 1 was 10 times larger compared with screws in sputtered $YBa_2Cu_3O_{7-x}$ films [4–6], but were only of the order of one tenth of that found in melt-grown structures [7, 8].

The screw-like structures in film 1 are formed by the turning of stacks consisting of 2–10 layers. In contrast, the screws observed in $YBa_2Cu_3O_{7-x}$ films exhibit single-layer turns [4–6]. Early stages of screw growth, involving the simultaneous movement of four-layer heads, are shown in Fig. 3e. The screw-like structures in film 1 are cylindrical, in contrast with the pyramidal shape typical for screw features in sputtered YBCO films.

The screw-like structures in film 1 exhibited symmetrical as well as elongated shapes, Fig. 3b,c. The height of the islands approximately corresponds to the number of unit cell steps displayed on the top of the islands. Often, an edge is formed parallel to the long axis of the island, dividing the 3D island into two roughly equal parts. A train of single-layer steps is developed around the edge (Fig. 3b,c). In some cases a central core was visible, around which the screw has developed (Fig. 3d).

To investigate possible causes for the observed differences between films 1 and 2 we studied the early stages of precursor film growth by depositing 2 nm thick films at several temperatures. At deposition temperatures in the range 200–600 °C, corresponding to film 2, the substrate was densely covered by 1–4 nm high clusters (Fig. 4a). On the film deposited at 800 °C there appeared well-separated (sep-

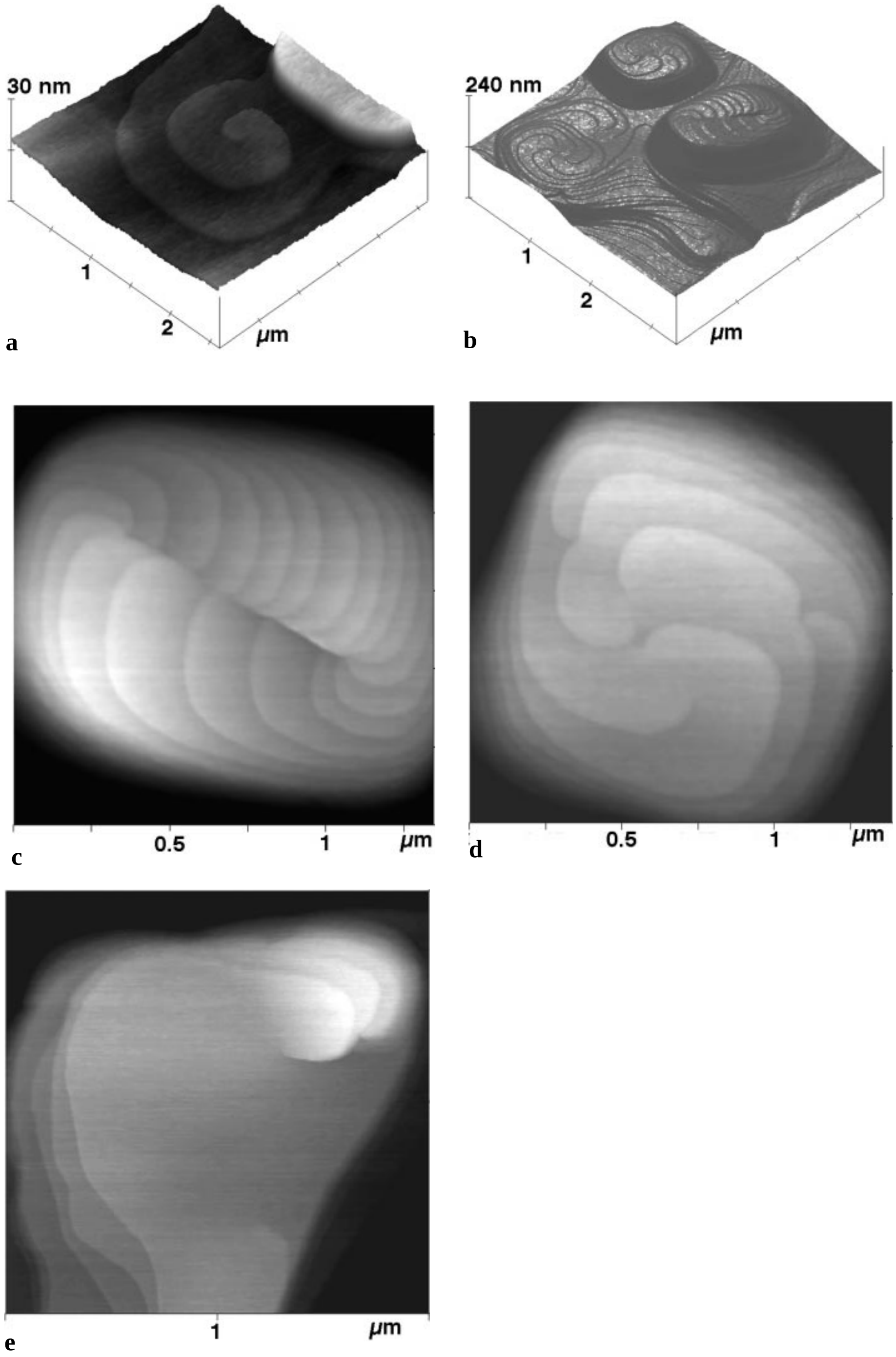

Fig. 3a–e. Screw-like structures in film 1. **a** Individual unit-cell-high screw-like structure on a terrace. **b** Different types of screw-like 3D structure. Upper left corner: symmetrical; right: elongated. **c** Screw developed around edge. **d** Screw developed around a central core. **e** Possible starting point of the formation of a screw

aration 50–200 nm) clusters of up to 40 nm height (Fig. 4b). The number density of clusters was $4 \times 10^{10}\,\mathrm{cm}^{-2}$ and $4 \times 10^{7}\,\mathrm{cm}^{-2}$ at 200–600 °C and 800 °C, respectively. X-ray analysis of a 200 nm thick precursor film deposited at 700 °C showed the presence of small amounts of BaO and CuO, while no crystalline phases appeared in a precursor film deposited at 300 °C.

The terraced nature of the *c*-axis-oriented Tl-2201 films and the frequent occurrence of steps of unit cell height indicates that the films grow predominantly by a step-flow mechanism. The observed differences in morphology between film 1 and film 2 are connected to the different deposition temperatures employed in precursor film deposition. Thus the increased roughness of the precursor surface combined with inclusions of crystalline phases are suggested to cause the formation of the screw-like structures in film 1. Two Tl-2201 films on $SrTiO_3$ grown using precursor films deposited at 300 °C [9] exhibited features similar to film 2. This confirms the importance of precursor film properties for the appearance of the screw-like features. It is suggested that screws form as steps flowing over the surface are diverted by obstacles in the form of second-phase inclusions in the precursor film. The appearance of elongated screw-like islands may be caused by the interaction of two or more

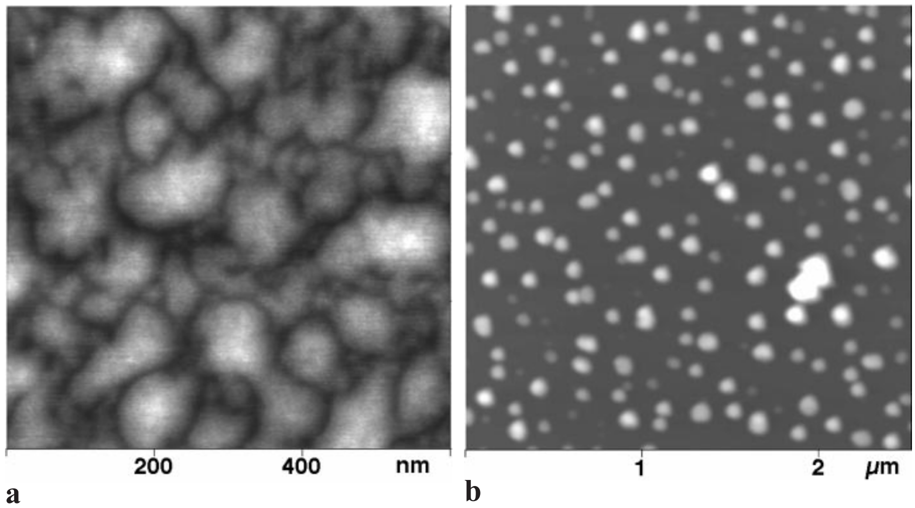

Fig. 4a,b. AFM images of 2 nm thick precursor films. **a** Deposition temperature < 600 °C resulted in a substrate coated by densely spaced 1–4 nm high clusters. **b** Deposition temperature 800 °C, resulting in up to 40 nm high well-separated clusters

stacks of layers. More complicated structures observed on the surface may be explained by the interaction of a number of steps.

3 Conclusions

Tl-2201 films on $Tl_2Ba_2CuO_6$ grow by a step-flow mechanism. A relatively low density of screw-like features occurred in one Tl-2201 film made using a precursor film deposited at 700 °C. It is suggested that these features are connected to the increased roughness of the precursor film and to inclusions of crystalline phases caused by the high deposition temperature.

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