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Giant step bunching in epitaxial $SrRuO_3$ films on vicinal $SrTiO_3(001)$

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Abstract

We report here on the giant step bunching, with steps reaching 20–40 unit cells high, observed in $SrRuO_3$ epitaxial films on vicinal $SrTiO_3(001)$. Bunching develops in films thicker than tens of nanometers, whereas three-dimensional (3D) islands were found in thinner films. We show that bunching forms when these islands coalesce. The growth of the 3D islands on substrates having varied miscut angles is investigated. In spite of the formation of bunching from initial nucleation of islands, the final morphology of the film surface is highly ordered as statistical analysis confirmed. The influence of processing parameters on the bunching morphology was investigated, and it has been demonstrated that lateral and vertical dimensions can be separately adjusted with a proper combined selection of film growth rate and substrate miscut angle.

Keywords: Step bunching; Ferromagnetic oxide; Film growth; Self-organization; Epitaxy

1. Introduction

$SrRuO_3$ (SRO) is ferromagnetic oxide below 160 K, and it is a conductor with a room temperature electrical resistivity of about 200–300 μΩ cm [1]. It has an orthorhombic structure that can be indexed as a distorted perovskite having a lattice parameter of 0.393 nm. The high compatibility of its crystal structure with other perovskite oxides, its high thermal stability [2], and the high electrical conductivity makes SRO suitable as electrode, mainly in ferroelectric devices [3].

SRO films deposited on vicinal, (001) oriented, $SrTiO_3$ (STO) substrates have the lowest electrical resistivity [4–6]. The surface of these films develops a remarkable step bunching [7–9], which contrasts to the morphology of terraces and monolayer steps observed in films on low vicinal substrates [2,9–12]. Bunching of steps implies an increased roughness of the surface, so its control is important, particularly when considering the use of SRO as metallic electrode in some applications. Moreover, surfaces with bunched steps are of interest as they could be used as templates for growth of low-dimensional structures [13]. Therefore, understanding and controlling the formation of bunching in SRO films is of the highest interest.

To date, bunching has been mainly studied in some detail, in semiconducting and metallic systems, being the number of monolayer steps in a bunch generally below ten [14–16]. Although less usual, bunches with more than ten monolayer steps were also reported [17–20]. It is commonly accepted that when growth mode is step flow, bunching results from differences in step velocity. However, we show here that bunching in SRO does not form in such a one dimensional process, but from the coalescence of three-dimensional (3D) islands. Details of the bunching formation process are reported elsewhere [8]. Here we present statistical analysis of both, lateral (in-plane) and vertical (out-of-plane) dimensions in SRO films displaying bunched morphology. We have also investigated the influence of the STO miscut angle, growth rate and the 3D islands-grown at initial stage, on bunching formation.

2. Experimental

SRO thin films were grown by pulsed laser deposition using a KrF excimer laser (248 nm wavelength and 34 ns pulse duration). The laser beam was focused, with a fluence around 2 J/cm^2, on a stoichiometric target of $SrRuO_3$ (SCI Engineered Materials). The target was rotated during the ablation process to reduce non-uniform erosion. Films of varied thickness were prepared on $SrTiO_3(001)$ substrates (CrysTec), misoriented

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towards the [100] direction by controlled vicinality θ_v up to ~4°. The films were deposited under 10 Pa of pure oxygen and at a substrate temperature of 750 °C. Atomic force microscopy (AFM) working in tapping mode was used to characterize the surface morphology of substrates and films. AFM image processing and analysis was done using the WSxM software [21]. AFM height profiles and X-ray diffractometry were used to determine the vicinality of the substrates. Additional details of the deposition conditions, as well as on epitaxial nature, lattice strain, and magnetotransport properties can be found elsewhere [5,6,22]. Films here reported were grown at rates of ~0.018 or ~0.0042 nm per laser pulse as deduced from proper calibration using X-ray reflectivity. The nominal thickness (t) of the films ranges from 1.7 to 100 nm.

3. Results and discussion

Fig. 1 shows the morphology of a SRO film grown on a vicinal ($\theta_v \geq 1°$) STO substrate, displaying flat terraces separated by bunches of steps. Within the vertical and lateral resolutions of the microscope, islands are not observed. Steps are parallel and, remarkably, they can be above 8 nm high. These films are epitaxial, with a (001) out-of-plane orientation (pseudocubic lattice, 1 unit cell (u.c.)~0.4 nm). Therefore, bunches can include more than 20 monolayer steps. This is illustrated in the height profile displayed in Fig. 1b, where u.c. units are used in the height axis. Bunching with so many monolayer steps is unusual, and thus it can be properly defined as giant bunching. The surface of the substrate used does not show any similar pre-existing morphology. In the inset of Fig. 1b we include an AFM image (1 × 1 μm^2 scan size, 2D view) of the θ_v~2° substrate after annealing in air during 2 h at 800 °C, a temperature higher than the used to grow the films. There is a well defined morphology of steps, one or half u.c. high, running along a [100] direction and separating terraces around 10 nm wide. It implies that bunching in SRO films is not a replica of a similar topography in the substrate, and thus it develops during the epitaxial growth. The high size of bunching in SRO films is well appreciated when comparing the AFM image of the substrate with a 2D view of the film

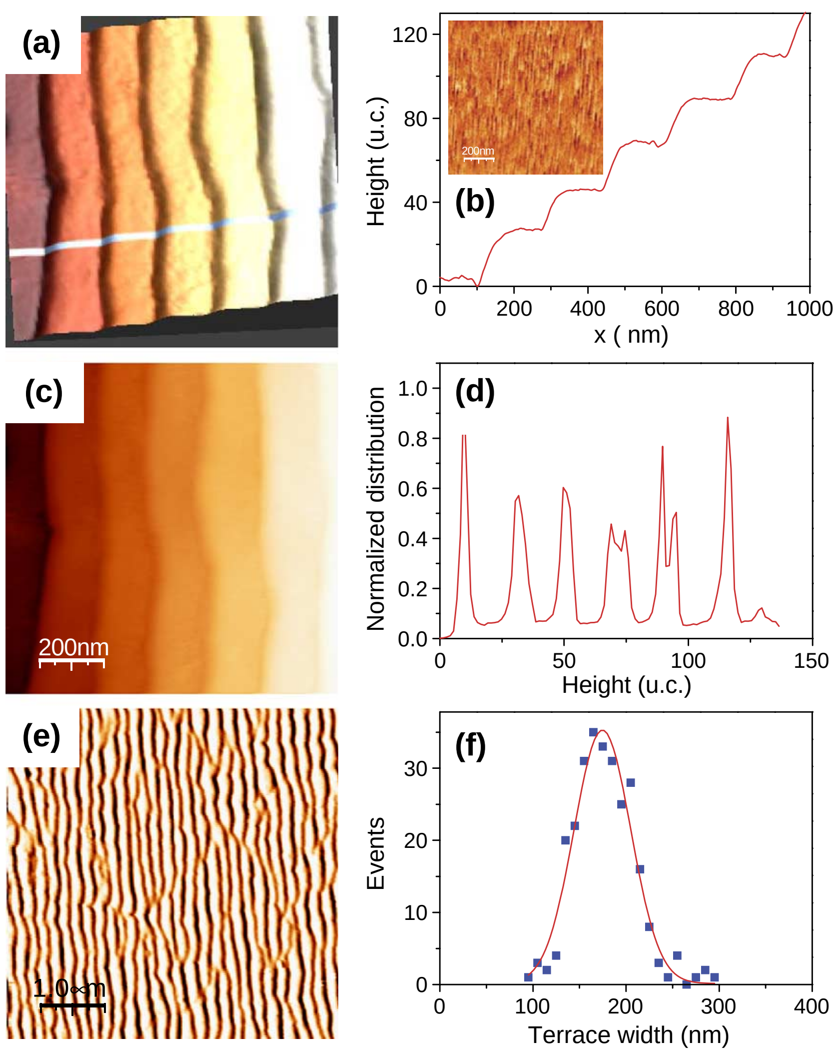

Fig. 1. Atomic force microscopy topographic images of a t = 100 nm film on θ_v~2° STO. (a) 3D view (1 × 1 μm^2 scan size) and, (b) height profile (height in SRO lattice unit cell units) along the line drawn in the image. Inset: topographic image of the substrate (1 × 1 μm^2 scan size); (c) 2D view (1 × 1 μm^2 scan size) and, (d) histogram of heights; (e) derivative view (5 × 5 μm^2 scan size) and, (f) histogram of terrace widths.