

# Ordered SrTiO<sub>3</sub> Nanoripples Induced by Focused Ion Beam

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**Abstract:** Ordered nanoripples on the niobium-doped SrTiO<sub>3</sub> surfaces were fabricated through focused ion beam bombardment. The surface morphology of the SrTiO<sub>3</sub> nanoripples was characterized using in situ focused ion beam/scanning electron microscopy. The well-aligned SrTiO<sub>3</sub> nanostructures were obtained under optimized ion irradiation conditions. The characteristic wavelength was measured as about 210 nm for different ion beam currents. The relationship between the ion irradiation time and current and SrTiO<sub>3</sub> surface morphology was analyzed. The presented method will be an effective supplement for fabrication of SrTiO<sub>3</sub> nanostructures that can be used for ferroelectric and electronic applications.

**Keywords:** SrTiO<sub>3</sub>; focused ion beam; nanoripple; self-assembly

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## Introduction

The performance for nano-optoelectronic and nano-electronic devices are expected to be improved through the use of ordered nanostructures [1-4]. Focused ion beam (FIB) bombardment has recently been of considerable interest in self-assembly due to its capability of forming well-ordered nanostructures via self-organization as well as templates for the nanomaterial growth. Ion-beam-induced spatially aligned nanoripples, nanodots, and nanowires have been reported [5-7]. For example, the periodic ripples or terrace-like patterns introduced by FIB have been obtained on metals [8, 9], semiconductors [10-12] and insulators [7, 13]. The self-organization introduced by FIB is attributed to the dynamic competition between the roughening and smoothing process under ion bombardment [13, 14]. All of these efforts demonstrate the potential of the FIB technique in nanofabrication.

In this letter, we report the observation of self-organized SrTiO<sub>3</sub> (STO) nanoripples under the influence of FIB irradiation. For many years, oxide materials have received great attention for electronic, dielectric, and thermoelectric applications [15]. STO is one of these important oxide materials with high dielectric constant, low dielectric loss, large carrier effective mass, and high chemical stability. Due to these appealing properties, a wide range of methods have been studied to fabricate STO nanomaterials, including the sol-gel method [16], hydrothermal synthesis [17], and sonochemical method [18]. Although self-organized nanostructures on oxides such as LaAlO<sub>3</sub> nanoripples introduced by FIB have been reported [7], few works has been reported on self-assembly of STO nanostructures by FIB.

This paper demonstrates that ordered STO nanoripples can be created on STO surfaces through FIB irradiation. Many experiments have been done to ob-

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serve the orientation, propagation, terrace surface details, and wavelength of the ripple structures. In this paper, different beam current and sputtering time are used to investigate the effects of ion bombardment on the morphology of the nanoripples. Well-ordered STO ripples are observed with a beam current of 1.0 nA for about 269 s.

## Experiment

The niobium-doped STO (Nb-STO) (111) surfaces were used in this study. The ion bombardment experiments were performed using a field emission scanning electron microscopy (SEM)/FIB dual-beam system (FEI Nova 200 NanoLab). A focused 30 keV  $\text{Ga}^+$  beam was used for ion sputtering with varying beam currents from 0.1 nA to 3.0 nA for different sputtering time. Spot sizes from  $10 \times 10 \mu\text{m}^2$  to  $20 \times 20 \mu\text{m}^2$  were sputtered with an overlap of 50%, and the dwell time of each spot is 1  $\mu\text{s}$ . The surface morphology was characterized by *in situ* SEM and atomic force microscopy (AFM) at room temperature. Many reports have pointed out that the surface material will be efficiently sputtered under off-incident angle. Thus, the experiments were all carried out at an incident angle of  $52^\circ$ .

Another important factor in FIB sputtering is the accelerate voltage. High-accelerate voltage provides high-energy of incident ions, which is necessary for some wide bandgap materials. High voltage will induce high local heating to the sample surface, which will influence the deposition and growth of the sputtered materials. For example, ordered Ga droplets can only be obtained under relatively lower voltages (e.g. 5 keV) [6]. We have compared some samples irradiated by 20 keV and 30 keV  $\text{Ga}^+$  with different beam currents and it is found that the STO surface is hardly sputtered under 20 keV  $\text{Ga}^+$  even for a beam current up to 3 nA. Therefore, the sputtering is all carried out using 30 keV  $\text{Ga}^+$ .

## Results and Discussion

Figure 1 shows the Nb-STO surface morphology after being irradiated under different ion beam currents (or ion flux). Each spot of  $10 \times 10 \mu\text{m}^2$  area is sputtered under the current of 0.1 nA, 0.3 nA, 0.5 nA, and 1.0 nA for five minutes. It can be seen that the surface is clearly better sputtered with some ripple structures with increasing ion beam current. In Fig. 1(b), the Nb-STO surface is characterized by the ion-beam microscopy, where some black shadow is induced by the ion beam. It is shown from Figs. 1 (c) and (d) that the terrace-like structures are obtained through the ion beam irradiation. As shown in Fig. 1, the morphology of the nanoripples is strongly dependent on ion flux.

The wavelength of the nanoripples decreases to a minimum at the beam current of 0.3 nA and then increases with the beam current. However, the ordering of the nanoripples has not been observed. In Fig. 1, the sputtering depth is different between each area; the sputtering depths are 230 nm and 400 nm for beam currents 0.5 nA and 1.0 nA, respectively. The sputtering also depends on the ion fluence instead of ion flux. Therefore, in order to investigate the effects of ion fluence on the spatial ordering of the Nb-STO nanoripples, the Nb-STO surfaces are irradiated with the same ion beam current but for different durations.

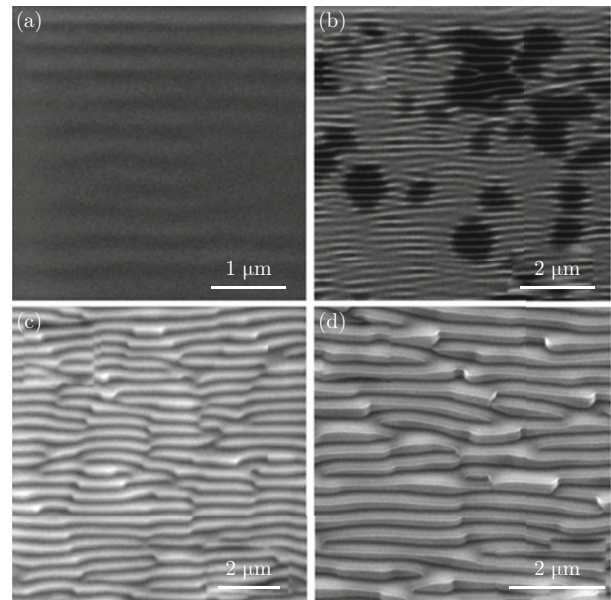


Fig. 1 The SEM images of the Nb-STO surface morphology sputtered by FIB on under different ion beam currents, incident angle  $52^\circ$ , dwell time 1  $\mu\text{s}$ , pattern time five minutes. (a) 0.1 nA, (b) 0.3 nA, (c) 0.5 nA and (d) 1.0 nA.

In order to investigate the effects of ion influence on STO nanoripples introduced by FIB, three  $10 \times 10 \mu\text{m}^2$  squares were milled with the milling depth of 150 nm, 200 nm, and 230 nm at 0.5 nA for 200 s, 267 s, and 300 s, as shown in Fig. 2 (a), (b) and (c). With the increase of sputtering time, the surface sputtering depth and wavelength increases which clearly leads to nanoripples. After sputtering for 267 s (ion fluence  $8.33 \times 10^{17}$  ions/ $\text{cm}^2$ ), some ordered nanoripple structures appear, while the surface morphology is disordered with a sputtering time of 200 s and 300 s. Secondly, the Nb-STO surfaces were irradiated under 1.0 nA beam current for different durations, 200 s, 269 s, and 300 s. The irradiation area for each current is  $10 \times 10 \mu\text{m}^2$ ,  $10 \times 20 \mu\text{m}^2$ , and  $10 \times 10 \mu\text{m}^2$ , respectively. The SEM images are shown in Fig. 2 (d), (e), and (f). Interestingly, the surface morphology with a sputtering time of 269 s (ion fluence  $8.4 \times 10^{17}$  ions/ $\text{cm}^2$ ) is more ordered than that with a sputtering time of 300 s (ion fluence  $1.87 \times 10^{18}$

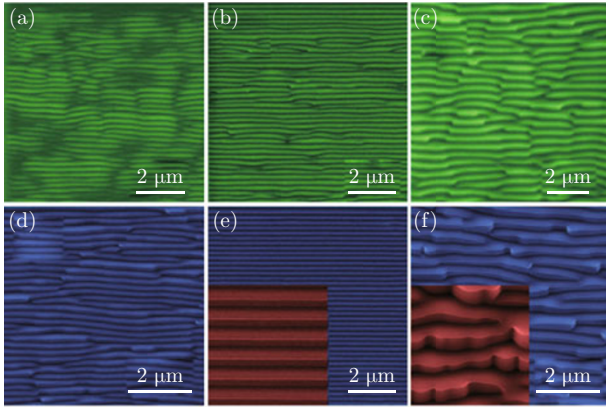


Fig. 2 The SEM images of surface morphology patterned by FIB under different current and time. (a) 0.5 nA (200 s), (b) 0.5 nA (267 s), (c) 0.5 nA (300 s), (d) 1.0 nA (200 s), (e) 1.0 nA (269 s) and (f) 1.0 nA (300 s). The insets in (e) and (f) are magnified SEM images.

ions/cm<sup>2</sup>). It is notable that the periodic structures obtained with a sputtering time of 269 s have clear edges. The ordered nanostructures are wave-like ripples (Fig. 2(e)) instead of terrace-shaped ripples (Fig. 2(f)). However, with a sputtering time of 200 s, the surface morphology also shows terrace-shaped ripples without ordering (Fig. 2(d)).

Figure 3(a) and (b) shows the three-dimensional AFM image and the line profiles of the ordered nanoripples, respectively, thus confirming that the well-aligned Nb-STO ripple structures are created by the FIB irra-

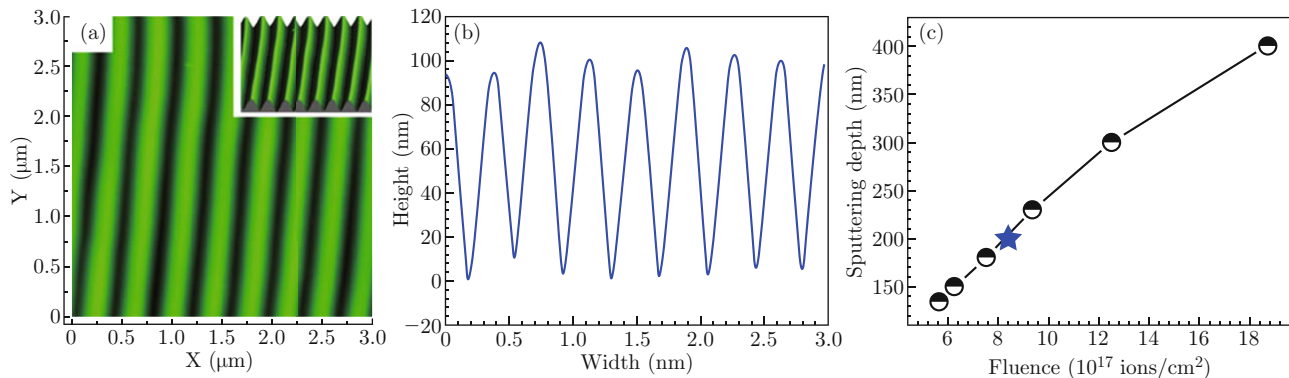


Fig. 3 (a) AFM image of the surface morphology of ordered STO nanoripples. The inset is the three-dimensional view. (b) The AFM line profile of the ordered STO nanoripples. (c) Ion milling depth versus ion fluence. The star indicates the optimum ion fluence for fabrication of ordered STO nanoripples.

## Conclusion

Nanoripples on the Nb-STO surface are obtained through self-assembly using FIB irradiation. The effects of different ion flux and ion influence on the surface morphology of nanoripples are investigated. The well-aligned STO nanoripples can be achieved with an ion fluence of  $8.4 \times 10^{17}$  ions/cm<sup>2</sup>. The ordered ripple structure has the characteristic wavelength of 375 nm

and amplitude of about 115 nm. These ordered Nb-STO nanostructures are similar to the FIB-introduced ripples reported on 3c-SiC and LaAlO<sub>3</sub> surfaces, where the amplitude of the ripple wave is slightly smaller [7, 12]. It is found that the sputtered atoms can reach an average range of 30 nm at each bombardment process simulated by the SRIM-2010 code. During the ion sputtering process, the surface will be irradiated repeatedly, and thus the sputtered atoms will be accumulated to form the terrace-shaped or saw-tooth-shaped structures. However, it is only with the proper ion fluence that ordered ripple structures can be obtained. The ion fluence can be obtained from sputtering time, beam current, and sputtering area. Figure 3(c) summarizes the milling depth versus the ion fluence under different operation parameters. The best ordering of Nb-STO nanoripples is observed with fluence of  $8.4 \times 10^{17}$  ions/cm<sup>2</sup>. Less ordered ripples are obtained with either higher or lower ion fluence. It is very interesting that similar ion fluences have been reported for fabrication of ordered nanoripples on LaAlO<sub>3</sub> and SiC surfaces [7, 12]. The previous reports on 3c-SiC and LaAlO<sub>3</sub> surfaces have demonstrated that the formation of ordered nanoripples is a balance between the surface roughening process and surface smoothing process [7, 12]. Because the sputtering process is a dynamic equilibrium between sputter and deposition, the ordered ripple structure could only be obtained when the ion fluence satisfies the balance condition.

and amplitude of about 100 nm. This method presents an effective means of fabrication of ordered ferroelectric nanomaterials for advanced electronic devices.

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