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The effects of carbon coating on nanoripples induced by focused ion beam

Guang Ran,[1,2] Jiaming Zhang,[2,5] Qiangmin Wei,[2] Shengqi Xi,[1] Xiaotao Zu,[4] and Lumin Wang[2,3,a)]

[1]*State Key Laboratory for Mechanical Behavior of Materials, School of Materials Science and Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, People's Republic of China*
[2]*Department of Materials Science and Engineering, University of Michigan, Ann Arbor, Michigan 48109, USA*
[3]*Department of Nuclear Engineering and Radiological Science, University of Michigan, Ann Arbor, Michigan 48109, USA*
[4]*Department of Applied Physics, University of Electronic Science and Technology of China, Chengdu, 610054, People's Republic of China*
[5]*Departments of Geological Sciences, University of Michigan, Ann Arbor, Michigan 48109, USA*

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The morphology and topography of self-assembled nanoripple structures on $LaAlO_3$ (100) surface with and without carbon coating were characterized using focused ion beam (FIB)/scanning electron microscope, as well as *ex situ* atomic force microscopy and transmission electron microscopy. When the surface was not covered by carbon coating and had low roughness, well-ordered and highly uniform nanoripples self-assembled on the surface bombarded by FIB. In contrast, disordered nanoripples formed after carbon coating. The deposited carbon significantly influences the morphology of surface nanoripples due to its effect on the surface roughness that affect the dynamic competition between the roughening and smoothing processes. This discovery suggests a means for fabricating well-ordered and highly uniform nanoripples for nanoscale devices application. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3054641]

Focused ion beam (FIB) bombardment is an attractive, alternative, and controlled fabrication method for preparing self-assembled periodic structures with nanometer dimensions which become very important in nanoscale devices. This technology offers the functions of high-energy ion beam (typically 30 keV Ga^+) sputtering and ion assisted chemical vapor deposition in the size of nanometer scale, and thus allows localized and controlled material removal and deposition.[1,2] The ion-induced nanoscale ripple structures via self-assembly processes have been of a particular interest. Ion-induced ripple patterns have been obtained on a variety of materials, including insulators [e.g., SiO_2 (Ref. 3) and $Cd_2Nb_2O_7$ (Ref. 1)], semiconductors [e.g., Ge,[4] Si,[5] diamond,[6] and SiC, (Ref. 2)] and metals [e.g., Ag,[7] Cu (Ref. 8)]. The ripples or wavelike patterns with a spatial periodicity varying from the nanometer to the micrometer range depend on many parameters including FIB system parameters (e.g., ion flux and fluence, ion energy, incident angle, and temperature) and target material parameters (e.g., composition, crystal structure, and orientation).[2,9–15]

Large numbers of previous experimental results indicate the nanoripples induced by FIB bombardment have many defects and exhibit certain disordered state on specimen surface.[1,16–20] However, the well-ordered and highly uniform nanoripples are more important for nanoscale device applications because of the controlled spacing and periodicity. When the surface of the dielectric and semiconducter materials is bombarded by FIB, there will be an induced charge on the specimen surfaces. In order to avoid this problem, conducting materials such as carbon or gold have often been deposited on the specimen surface prior to FIB milling. As shown later in this report, the deposited carbon significantly influence the morphology and topography of surface nanoripples created by FIB bombardment.

In this work, FIB induced self-assembled nanoripples on the $LaAlO_3$ (100) surface were studied. In particular, the difference of the ripple morphologies on the surfaces with or without carbon coating before the FIB milling was investigated while keeping the ion energy, ion beam current, and incident angle constant.

Commercially available wafers of $LaAlO_3$ single crystal with the (100) orientation were used in the present work. The ion bombardment experiments were carried out using a field emission SEM/FIB dual-beam system (FEI Nova 200 NanoLab). A 30 keV focused Ga^+ beam was used. During the FIB bombardment, a predesigned rectangular area (e.g., $10\times 10\ \mu m^2$) was selected, and the dwell time was 1 μs for every FIB patterning. *Ex situ* atomic force microscopy (AFM) characterization of the patterned samples was performed in a Nanoscope IIIa AFM in air, operating in a tapping mode using phosphorus doped Si cantilevers. Three-dimensional (3D) topography images were obtained by processing the original AFM data. The transmission electron microscope (TEM) specimens were prepared using the FIB "lift-out" technique. High resolution TEM (HRTEM) observations were performed using a JEOL 2010F field emission gun analytical electron microscope (200 kV).

When the surface of insulating $LaAlO_3$ sample is bombarded by FIB, there is a charge buildup on the specimen surface. Carbon coating on the surface is required to avoid the charging effects. However, the roughness of $LaAlO_3$ surface increases after the carbon coating process. Disordered nanoripples formed by FIB bombardment on the $LaAlO_3$ (100) surface with carbon coating as shown in Fig. 1(a). Previous experimental results showed that low surface roughness and no ripples after normal incident angle of FIB bombardment.[2] In the present study, a FIB with 5 nA ion

[a)]Author to whom correspondence should be addressed. Electronic mail: lmwang@umich.edu. Tel: 1-734-647-8530. FAX: 1-734-647-8531.

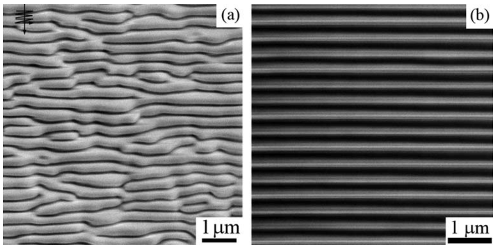

FIG. 1. SEM images showing the nanoripples on the single crystal $LaAlO_3$ (100) surface induced by 30 keV Ga^+ FIB bombardment. The wave vector is parallel with the ion beam direction as marked by the arrow. (a) Irregular nanoripples on the $LaAlO_3$ specimen surface with carbon deposited before FIB bombardment. (b) The well-ordered and high uniform nanoripples on the $LaAlO_3$ specimen surface without carbon coating.

current was used to scan on the specimen surface at normal incident angle to remove the deposited carbon, which produced a smooth surface area with rms roughness less than 2.5 nm as shown in Fig. 2(b). The carbon eliminated area is about $30\times30\ \mu m^2$. The elimination depth was selected according to the depth of carbon coating on the surface. The area without coated carbon is free from the charging effect since some Ga^+ ions are bombarded into the surface serving as an conducing medium between ion beam and the area.

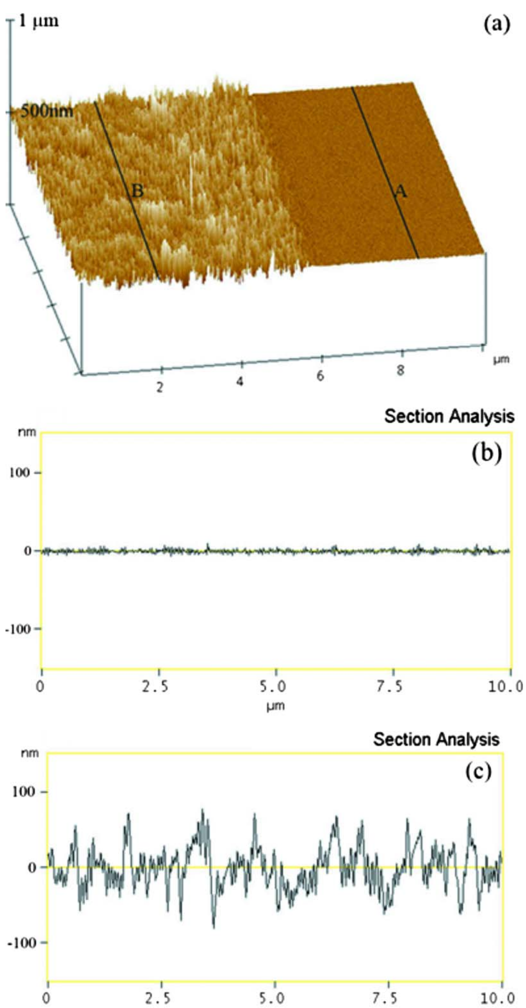

FIG. 2. (Color online) (a) AFM 3D image showing the morphology and topography of $LaAlO_3$ (100) surface before FIB bombardment. The area on the right was without carbon coating as marked A, the area on the left was with carbon coating as marked B; [(b) and (c)] the section analysis of AFM image along lines A and B, respectively.

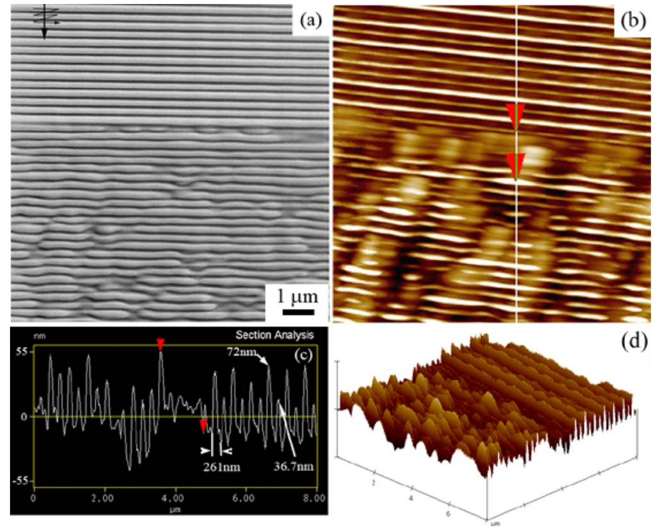

FIG. 3. (Color online) (a) SEM image showing the nanoripples on the single crystal $LaAlO_3$ (100) surface induced by 30 keV Ga^+ FIB bombardment. The wave vector is parallel with the ion beam direction as marked by the vertical arrow; (b) AFM image showing the morphology of nanoripple structure on the $LaAlO_3$ (100) surface; (c) the section analysis of nanoripples; and (d) AFM 3D image showing topography of nanoripples structure.

However, it is very important that there is a large surface area with deposited carbon around the FIB scanned area for avoiding the surface charging.

Figure 1 is a SEM images showing the nanoripples on the single crystal $LaAlO_3$ (100) surface induced by 30 keV Ga^+ FIB bombardment. The process was proceeded with ion current of 0.5 nA, incident angle of 52°, ion beam scanned area of $16.2\times10\ \mu m^2$, ion flux of 1.92×10^{15} ions/cm^2 s, and ion fluence of 7.97×10^{17} ions/cm^2. The wave vectors are parallel to the ion beam trajectory [as marked by the vertical arrow in Fig. 1(a)]. Well-ordered and highly uniform nanoripples can be induced by FIB bombardment on the $LaAlO_3$ (100) surface without prior carbon coating as shown in Fig. 1(b). In contrast, the disordered nanoripples shown in Fig. 1(a) were formed under the same ion beam bombardment condition if carbon coating remain on the $LaAlO_3$ (100) surface. The only explanation for different results is the change in surface roughness due to the carbon coating which affects the dynamic competition between the roughening and smoothing process under ion bombardment.

Figure 2(a) is an AFM 3D image showing the topography of $LaAlO_3$ (100) surface before FIB bombardment. It is evident that the surface without carbon coating is very smooth. The cross-sectional profiles in the AFM image are shown in Fig. 2(b) and 2(c). The roughness of surface with carbon coating is about 29 nm, while the roughness of surface without carbon coating is less than 2.5 nm. After FIB bombardment, it can be seen that well-ordered and highly uniform nanoripples formed on the surface without carbon coating and disordered nanoripples appeared on the surface with carbon coating as shown in Fig. 3. The AFM 3D image and the cross-sectional analysis of AFM image is shown in Figs. 3(c) and 3(d). The wavelength of well-ordered ripples is approximately 261 nm. The average amplitude of well-ordered ripples is approximately 55 nm.

Real time observation from the recorded movies during FIB bombardment indicates that the well-ordered and highly uniform nanoripples on surface without carbon coating are

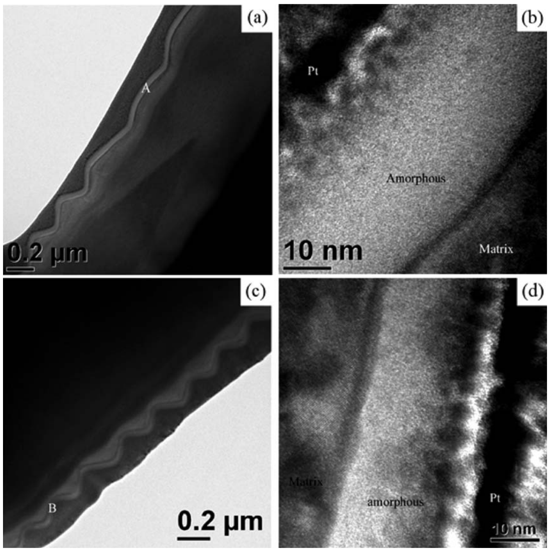

FIG. 4. TEM images and HRTEM images showing the cross section of nanoripples on the single crystal $LaAlO_3$ (100) surface induced by 30 keV Ga^+ FIB bombardment. (a) TEM image showing the cross-sectional morphology of disordered nanoripple on the specimen surface with carbon coating; (b) HRTEM image of area A in (a); (c) TEM image showing the cross-sectional morphology of well-ordered nanoripples on the specimen surface without carbon coating; (d) HRTEM image of area B in (c).

formed at an early stage and then the amplitude of nanoripples is increased with the time of ion bombardment. However, the disordered nanoripples on the surface with carbon coating were formed from the beginning of FIB bombardment, and always present disorder state even with the extended bombardment time. The time for formation of nanoripples on the surface with carbon coating is shorter than that on surface without carbon coating. It took only several seconds to form disordered ripples.

TEM specimens were prepared perpendicularly to the surface of nanoripples. The cross-sectional morphology of nanoripples on the single crystal $LaAlO_3$ (100) was shown in Fig. 4. It can be seen that the cross section of nanoripples are irregular for the specimen with carbon coating as shown in Fig. 4(a), but are well ordered for the specimen without carbon coating as shown in Fig. 4(c). From TEM images, the amorphous layer covering the nanoripples were observed. The thickness of amorphous lay is about 35 nm as shown in Figs. 4(b) and 4(d). Each nanoripple shows axial symmetry on the specimen surface without carbon coating as shown in Fig. 4(c), which indicates that the nanoripples are self-assembled under FIB bombardment.

Several theories have been developed to describe the ion-beam-induced surface ripples.[21–23] The spontaneous formation of self-assembled rippled patterns can be understood as a competition between the roughening induced by ion bombardment and the smoothing caused by surface diffusion. Beyond a critical incident angle (50°), the balance of curvature-dependent sputtering and surface diffusion leads to the ripple formation.[2,23] The rough carbon surface will affect the dynamic competition between the roughening and smoothing process.

Well-ordered and high uniform nanoripples were also observed on the commercially available wafer of $SrTiO_3$ (100) single crystal and Al_2O_3 (001) single crystal surface without carbon coating and with low surface roughness. The differences among these results are the values of wavelength and amplitude of nanoripples induced by the same FIB bombardment parameters.

In summary, predeposited carbon on the specimen surface significantly influences the morphology and topography of surface nanoripples induced by FIB bombardment because the carbon coating increases the roughness of specimen surface. Under off-normal bombardment without sample rotation, well-ordered and highly uniform nanoripples can be induced by FIB bombardment on $LaAlO_3$ (100) surface without prior carbon coating. However, only disordered nanoripples were obtained on the same surface with carbon coating prior to the FIB milling. The method of ion beam bombardment to produce well-ordered and highly uniform nanoripples on smooth surfaces may be applied to the fabrication of nanodevices. The dimensions of the ripple structures make them ideal for applications as subwavelength photonic devices.[24–26]

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