Applied Surface Science 268 (2013) 270–273

Contents lists available at SciVerse ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Roughness scaling in titanium thin films: A three-dimensional molecular dynamics study of rotational and static glancing angle deposition

Matilda Backholm [a,*], Morten Foss [a], Kai Nordlund [b]

[a] Interdisciplinary Nanoscience Center (iNANO), Gustav Wieds Vej 14, Aarhus University, 8000 Aarhus C, Denmark
[b] Department of Physics, University of Helsinki, P.O. Box 64, 00014 Helsinki, Finland

ARTICLE INFO

Article history:
Received 20 September 2012
Received in revised form
11 December 2012
Accepted 11 December 2012
Available online 21 December 2012

Keywords:
Glancing angle deposition
Surface roughness
Molecular dynamics simulations

ABSTRACT

Three-dimensional molecular dynamics simulations of the glancing angle deposition of titanium was performed both with and without substrate rotation for different deposition angles ($\alpha = 85^\circ$, 80°, 55°, and 0°). The surface roughness of the final films, all consisting of 10,000 deposited atoms, was calculated at different length scales of the substrate. The roughness scaling was shown to be, within error, identical for the rotational and static glancing angle deposited thin films.

1. Introduction

Glancing angle deposition (GLAD) is a physical vapour deposition technique performed at oblique angles, α, of incidence [1,2]. The deposition angle is defined as the angle between the direction of the incoming atoms and the normal to the surface, and is in GLAD typically considered to be $\alpha > 70^\circ$ [3]. As atoms are deposited, islands will nucleate on the substrate [4]. Due to the grazing angle of incidence, these islands will "shadow" part of the substrate. This phenomenon is known as self-shadowing [5], or geometrical/ballistic shadowing [6,7], and gives rise to the growth of isolated columns on the surface.

In static GLAD, the substrate is kept fixed during the deposition, producing columns tilted towards the incoming flux of atoms [8–10]. By rotating the substrate, vertical columns will be grown [6]. The different substructures of these columns, e.g., helical or zig–zag shapes, will depend on a range of experimental parameters, such as rotational speed and continuity of rotation. In both static and rotational GLAD, the resulting thin films have characteristic topographical features on the nanometer scale [7], including a roughness which is easily tuned by changing the deposition angle [11]. The roughness scaling in GLAD thin films has been of the greatest interest experimentally, as these nano-rough films have the potential to be used as, e.g., bio-compliant coatings on medical implants [12,13].

The roughness of a two-dimensional surface depends on how much the film thickness, $h(\mathbf{r})$, deviates from the mean thickness, $\bar{h}$, at different points on the surface, described by the position vector $\mathbf{r}$ [11]. This can be expressed by the so-called interface width, which is defined as the root-mean-square (rms) fluctuation of the height [14]

$$w(L) = \sqrt{\frac{1}{L^2}\sum [h(\mathbf{r}) - \bar{h}]^2}, \tag{1}$$

where L is the linear size of the surface and the mean value of the height is defined as

$$\bar{h} = \frac{1}{L^2}\sum h(\mathbf{r}). \tag{2}$$

For the case of self-affine surfaces, such as a GLAD film [11], the roughness is generally described as [15,16,14]

$$w(L) \sim \begin{cases} w_{sat}, & \text{if } L \gg L_{crossover} \\ L^{\gamma}, & \text{if } L \ll L_{crossover} \end{cases} \tag{3}$$

where $L_{crossover}$ is the length at which saturation of the roughness, w_{sat}, occurs, and γ is the so called roughness exponent, describing the power-law-like increase of the roughness at smaller length scales [11]. Furthermore, the saturated roughness has been shown to scale as a power law function of the deposition angle [17]

$$w_{sat} \propto w_0 + \alpha^{\kappa}, \tag{4}$$

* Corresponding author. Tel.: +1 905 525 9140.
E-mail address: backhomm@mcmaster.ca (M. Backholm).

0169-4332/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.apsusc.2012.12.077

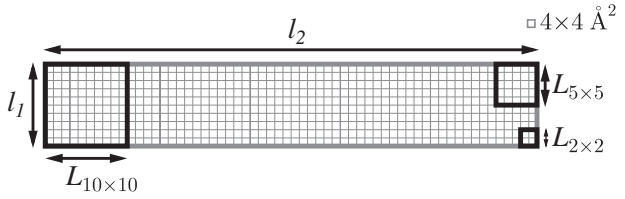

Fig. 1. A schematic illustration of the roughness calculation procedure for the static GLAD film. In each of the smallest grids, with a size of 4×4 Å^2, the highest z-coordinate was determined, giving rise to a height map with atomic resolution in all directions. The roughness was then calculated by using Eq. (1) in differently sized regions. Three examples of different section sizes, L, are shown. All of these regions have a side length that is a fraction of both the width, l_1, and length, l_2, of the surface.

where κ is the saturated roughness exponent, and w_0 is the saturated roughness of a reference film grown at an angle normal to the surface.

Two-dimensional molecular dynamics (MD) simulations of GLAD have previously been performed [18–20], and other simulation techniques have also been used to study the GLAD process on a theoretical level [21–23]. In previous work, we have compared a complete, three-dimensional MD simulation of rotational GLAD with experimental findings [17]. Here we present the results from three-dimensional MD simulations of both rotational and static glancing angle deposition of titanium. The roughness scaling of the resulting films was investigated, and a comparison between the two cases was made. The results presented here contribute with new insights into the nano-topographical features of GLAD thin films.

2. Molecular dynamics simulations

The code PARCAS ([24], see also [27,28]) was used to perform three-dimensional molecular dynamics (MD) simulations of both static and rotational glancing angle deposition. The Ti part of the Sabochick–Lam CuTi-potential [25] was used to deposit titanium atoms onto a four monolayer thick titanium substrate. To simulate an infinite surface of a bulk material, periodic boundaries were applied to the substrate in the directions parallel to its surface. Furthermore, temperature control was applied to the borders of the cell, as well as to the two top layers of the substrate. The two bottom layers were fixed. All of the simulations were performed at 300 K.

In this work, GLAD was studied at four different deposition angles: $\alpha = 85°$, 80°, 55°, and 0°, the last corresponding to deposition performed at an angle normal to the surface. A total number of 10,000 atoms were deposited in each separate case. To account for statistical variations, three simulations with different random seed numbers were performed per angle, summing up to 24 different simulations in total.

To simulate rotational GLAD, a square shaped substrate with the side length 123.12 Å and width 121.79 Å, corresponding to 2016 atoms per monolayer and 8064 atoms in total, was created. The simulation of a deposition event was performed by creating a titanium atom 6.5 Å above the highest point of the surface at a randomly chosen position in the xy-plane. An energy of 0.04 eV was given to the atom at an angle α in the z-direction, directing the atom towards the surface at a chosen deposition angle. The simulation time for the deposition of one atom was 30 ps, with a variable time step [26] of about 4 fs. After each deposition event, the direction of the subsequent atom was rotated one degree in the xy-plane, simulating the planar rotation of the substrate. This is equivalent to a rotational speed of 0.03° ps^{-1}, or 5.6×10^9 rpm. The computational deposition flux was 2.24×10^8 atoms nm^{-2} s^{-1}, giving a similar ratio between rotational speed and deposition flux as used experimentally [17].

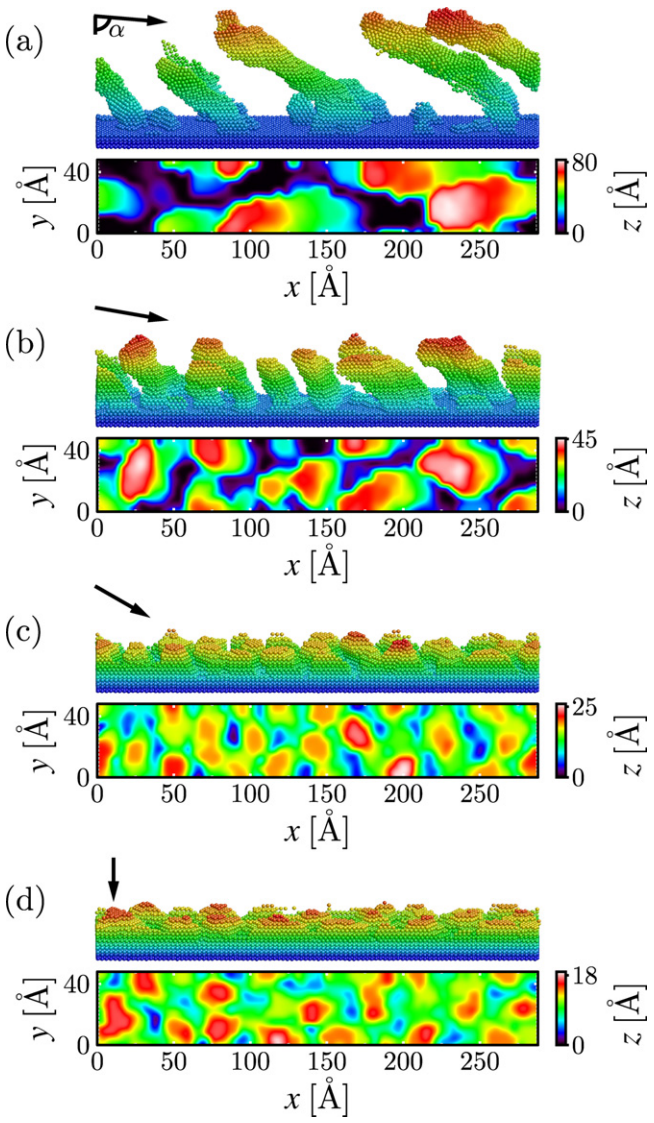

Fig. 2. The final structures of the thin films produced with static GLAD with the deposition angles (a) $\alpha = 85°$, (b) $\alpha = 80°$, (c) $\alpha = 55°$, and (d) $\alpha = 0°$. The length and width of the substrate is 294 Å and 51 Å, respectively, and the colour scheme illustrate the height variations of the films. The arrows indicate the direction of the deposited atoms. The height map of each film is shown below the snapshots, and have a colour scale specific to each case. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

In the static GLAD simulations, a substrate with the side length and width of 293.73 Å and 50.65 Å, respectively, was created. This size corresponds to a similar substrate area as in the rotational simulations, with a total of 8000 titanium substrate atoms. The rectangular shape of this substrate was chosen in accordance to the columnar growth direction, as most of the columns would be aligned along the direction of the incoming flux of atoms. With this substrate shape, the self-shadowing effect could therefore be captured in a more reliable way than what would have been possible with the square shaped substrate used for the rotational GLAD simulation. The deposition was performed in the same manner as described above, but without the rotational step. All of the atoms were deposited in the xy-direction parallel to the longer side of the substrate.

To calculate the roughness of the final structure, the substrate was divided into a 4×4 Å^2 grid in the xy-direction, as illustrated in Fig. 1 for the case of the static GLAD simulations. The size of the grid was chosen to be as small as possible, while not giving rise to any