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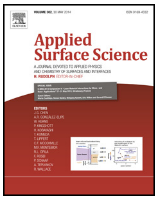

Temporal evolution of ripple pattern on silicon surface: An ion induced solid flow approach

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ABSTRACT

In this paper, we study temporal evolution of low energy ion-beam induced parallel-mode ripple patterns and explore the possibility of applying ion induced solid flow model on our experimental data. Experiments were performed by using 500 eV argon ions at a fixed ion flux over an angular window of 51° to 72.5°—where parallel-mode ripple patterns evolve. It is observed that evolution of ripples at all angles remains in the linear regime (where the ripple wavelength remains constant and the roughness evolution follows an exponential growth) up to a certain crossover time beyond which the non-linear effects get into the act (in the form of ripple coarsening at lower incidence angles and faceting at higher ones). The intrinsic timescale, beyond which transition from the linear to the non-linear regime takes place at all angles, matches quite well with those predicted by the solid flow model. It is also observed that the non-linear regime sets in quickly at higher incidence angles.

Introduction

Ion beam sputtering (IBS) leads to formation of spontaneously arising patterns of corrugations, holes, and dots [1,2] at the surfaces of different materials viz. metals, semiconductors, and insulators [1,3–7]. This process is cost-effective and shown to be useful for thin film/nanoscale magnetism and plasmonics. In fact, patterned substrates, mostly in the form of ripples, are being used as templates to grow and tune magnetic and plasmonic properties of thin films by using their morphological anisotropy [8–11]. However, in spite of its potential future applications, complete control over tailoring IBS induced self-organized patterns with desired properties is yet to be achieved. One of the major reasons behind this has been the lack of complete understanding of the mechanisms that govern the pattern formation.

Ripple formation is known to result from off-normal ion bombardment of materials. In the linear stability analysis of Bradley and Harper (BH), patterns originate from destabilizing (roughening) effect in which the erosion rate is enhanced at regions of high concave curvature and the stabilizing (smoothening) effect of surface diffusion [12]. This theory explains many experimental observations although significant contradiction exists. One of the important predictions of BH theory is that a flat surface is always unstable under ion erosion. In contrast, experimental studies on Si and SiO_2 [13–16] show the existence of flat surfaces, up to incident angles of 50° and 45°, respectively, from where ripple formation starts. Such a sharp boundary between a flat (stable) and a rippled (unstable) silicon surface was identified by Madi *et al.* as bifurcation in control parameter (incident angle, θ and ion energy, E) space [17]. It was concluded that the experiments reported by them, together with a careful analysis of the dynamics near the bifurcation point (flat surface-to-ripples phase transition), show the inadequacy of all existing models and provides important constraints on the relevant physical processes in ion beam sputtering. In a later work, Madi *et al.* [15] showed that stability at low angles and topographical instability at high angles can be determined by the effects of ion impact-induced prompt atomic redistribution (calculated from Carter–Vishnyakov (CV) model) [13] and that the erosive component (calculated from BH model) is essentially irrelevant.

In a more recent work, Castro *et al.* [18] put forward an ion induced solid flow model to explain formation of ripple patterns at high angles. This theory is based on the continuum description of the surface flow that is driven by surface confined stress-induced

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viscous flow of a thin amorphous layer that forms during ion bombardment. It has been pointed out by the same authors that both the mass redistribution and the viscous flow descriptions agree with the experiments with respect to the transition from flat to rippled surface at a particular incidence angle and the dependence of the pattern wavelength on incident angle of ions [19]. However, molecular dynamics (MD) based calculations cannot probe the required timescales associated with pattern evolution whereas the solid flow model offers the advantage of time dependent (dynamic) evolution of ripple patterns [15,20].

In this paper, we identify the hitherto unexplored angular window in which the solid flow model may be invoked to explain 500 eV Ar-ion induced ripple formation. Since, in general, it is observed that the ion incidence angle (in our case 51°) at which transition from flat to rippled surface takes place remains constant at different energies (in the range of 0.25–1.5 keV) [17,19,21], we have chosen an Ar-ion energy of 0.5 keV as a representative one and tested the applicability of the solid flow model on silicon over an angular window of 51° to 72.5°—wherever ripple patterns evolve. We found out the experimental timescale at an arbitrary angle of incidence – the reference angle (60°) – up to which the system evolved linearly. This time was used to predict the intrinsic timescale (by invoking the solid flow model) for other ion incidence angles and subsequently systematic experiments were carried out to check their proximities at each of those angles. These angles included the transition point of flat to rippled surface and beyond which parallel-mode ripples disappear. We also show that the temporal evolution of ripples at all these angles remains in the linear regime although the non-linear regime sets in quickly at higher incidence angles.

Theoretical approach

The solid flow model is based on the assumption that due to the impact of the ions and the subsequent release of energy within the target, defects are created inside the material. These events occur within a few picoseconds after the impact. Relaxation of some amount of defects leads to sputtering of target atoms which is associated with the generation of a residual stress that is confined within a thin amorphous layer that builds up beneath the surface [22] and reaches a stationary value. This ion-induced (compressive) stress is manifested as a highly viscous flow of the incompressible amorphous layer. Using a hydrodynamic description of the solid flow with a linear approximation in perturbations around a flat target profile, Castro *et al.* [23] obtained a real part for the linear dispersion relation (i.e. amplification rate of wave vector, q) that is given by

$$\omega_q = \frac{-\left[f_E d^3 \phi(\theta) q^2 + \sigma d^3 q^4\right]}{3\mu} \tag{1}$$

where $\phi(\theta) = \partial/\partial\theta\left(\Psi(\theta)\sin\ \theta\right)$, d is the average thickness of the amorphous layer, μ is (ion-induced) viscosity, and σ is the interface surface tension. The parameter f_E can be understood as the gradient of residual stress induced by the ions across the amorphous layer, whose angular dependence is described through the function $\Psi(\theta)$. Since this angular function needs to be prescribed, we take $\Psi(\theta) = \cos\theta$ as the simplest geometrically motivated choice that shows a good agreement [23]. According to the solid flow model [18], the smallest timescale associated with ripple formation in the linear regime can be written as

$$\tau(\theta, E) \sim \frac{\mu}{f_E^2 d^3 \phi^2(\theta)} \tag{2}$$

The most general prediction that can be obtained from this theory is that it allows finding the characteristic scale for the exponential growth of the pattern amplitude occurring at short times where linear approximation holds. For a given pair of angle and energy reference values, $(\theta_{\mathrm{ref}}, E_{\mathrm{ref}})$, one can extrapolate the value of the intrinsic timescale, τ, for any other pair (θ, E) through

$$\tau(\theta, E) = \tau(\theta_{\mathrm{ref}}, E_{\mathrm{ref}}) \frac{J_{\mathrm{exp}}(\theta_{\mathrm{ref}}, E_{\mathrm{ref}}) E_{\mathrm{ref}}{}^{-7/3+2m} \phi^2(\theta_{\mathrm{ref}})}{J(\theta, E) E^{-7/3+2m} \phi^2(\theta)} \tag{3}$$

where $J_{\mathrm{exp}}(\theta_{\mathrm{ref}}, E_{\mathrm{ref}}) = J_{\mathrm{exp}}(E)\cos\theta$ is the flux used in a particular experiment for energy E and angle θ, with $J_{\mathrm{exp}}(E)$ being the flux at normal incidence and m can be assumed to be in the range 1/3–1/2 [18].

In case of solid flow model, based on detailed binary-collision simulations, it is assumed that the generated stress depends on the ion energy (<2 keV) [24,25]. However, there is no direct measurement which quantifies ion induced stress. Thus, it would be a good strategy if the parameter, f_E, can be avoided while calculating the intrinsic timescale at any angle. In order to do so, instead of using Eq. (2), we experimentally determined the intrinsic timescale (for a reference ion incidence angle) up to which the growth of roughness follows an exponential behaviour and the ripple wavelengths remain constant. This intrinsic timescale has been utilized to verify the validity of solid flow model at other ion incidence angles.

Experimental

The substrates used in the experiments were sliced into small pieces from a *p*-type Si(1 0 0) wafer (B-doped, resistivity 0.01–0.02 Ω cm). A UHV-compatible experimental chamber (Prevac, Poland) was used which is equipped with a 5-axes sample manipulator and an electron cyclotron resonance (ECR) based broad beam, filament-less ion source (GEN-II; Tectra GmbH, Germany). The chamber base pressure was below 5×10^{-9} mbar and the working pressure was maintained at 2.5×10^{-4} mbar by using a differential pumping unit. Silicon samples were fixed on a sample holder which was covered by a sacrificial silicon wafer of the same lot to ensure a low impurity environment [14]. This was further confirmed by quantifying Rutherford backscattering spectrometric data (using 2 MeV He$^+$-ions) which did not show the presence of any impurity above the detection limit ($\sim 10^{15}$ at cm^{-2}). The beam diameter and the ion flux were measured to be 3 cm and 1.3×10^{14} ions cm^{-2} s^{-1}, respectively. The experiments reported here were performed at this fixed flux value and fluences in the range of 1×10^{16} to 1×10^{18} ions cm^{-2} (corresponding to the exposure times of 1 to 120 min) for four incidence angles, viz. 51°, 60°, 65°, and 72.5°.

Following Ar-ion exposure the samples were imaged by *ex-situ* atomic force microscopy (AFM) (MFP-3D; Asylum Research, USA). Silicon probes were used having diameter ~10 nm. Root mean square (rms) surface roughness, w, was calculated for all AFM images by using the WSxM software [26]. Transmission electron microscopic (TEM) study was performed for one sample exposed to Ar-ion beam at 60° incident angle.

Results and discussion

Fig. 1 presents a high-resolution cross-sectional TEM image of a silicon sample which was exposed to argon ions at 60° for the duration of 11 min. The presence of ion-induced surface corrugation in the form of ripples (wavelength, $\lambda \sim 30$ nm) is clearly seen from Fig. 1. In addition, a thin amorphous silicon layer at the top is also evident which forms during ion bombardment and subsequently acts as a highly viscous and incompressible medium. This meets one of the pre-requisites for invoking solid flow model in the present case.

To further justify the invoking of the solid flow model, we performed systematic experiments at 60° which was chosen to be the

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