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Transition from smoothing to roughening of ion-eroded GaSb surfaces

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During ion sputtering of GaSb(100) surfaces a transient behavior from initial smoothing to roughening accompanied by self-organized pattern formation has been observed using *in situ* x-ray reflectivity and grazing incidence small angle scattering. The induced patterns show hexagonally ordered nanodot arrays with a spatial periodicity of 30 nm. The correlation length of the pattern increases with ion fluence. In the framework of the Bradley–Harper model [R. M. Bradley and J. M. E. Harper, J. Vac. Sci. Technol. A **6**, 2390 (1988)], where the dot pattern formation results from an interplay of surface roughening due to sputtering and surface smoothing due to diffusion, the initial smoothing behavior is explained by the same surface diffusion processes as the pattern formation. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3136765]

The roughness of surfaces and interfaces strongly influences many properties of thin films and surfaces, among them the electrical, optical and magnetic properties.[1] Therefore controlling the roughness is an important issue in a broad variety of technological applications. In addition, by controlling the roughness down to the nanoscale the structure and properties of films deposited on these surfaces can be tuned. For example, nanostructured surfaces can be used as templates for the growth of thin magnetic films with a predefined anisotropy[2] and metallic thin films exhibiting dichroitic plasmonic properties.[3] When materials are bombarded with energetic ions the surface is eroded by the sputtering process. Ion beam sputtering (IBS) is often used to modify the roughness of solid surfaces on lateral scales ranging from a few nanometers up to micrometers.[4] Under certain sputtering conditions (energy, ion mass, incidence angle, fluence, etc.), IBS leads to a smoothing of the surface, whereas at other parameters the surface roughness is increased, leading eventually to periodic patterns, i.e., ripple or dot patterns.[5] Recently, it has also been shown that the pattern formation might be accompanied by an actual decrease of the surface roughness.[6]

In this letter we report on *in situ* x-ray scattering measurements of the morphology of GaSb (100) surfaces during Ar^+ IBS at normal incidence. Especially, the early time regime is addressed, which gives insight into the detailed mechanisms governing the dynamics in the beginning of the pattern formation process. The roughness and morphology of the sample surface are measured by *in situ* x-ray reflectivity (XRR) and grazing incidence small angle x-ray scattering (GISAXS). This experimental approach, i.e., *in situ* x-ray scattering, has proven to be a valuable tool to investigate the surface evolution and transient behaviors in the early time regime of the ion erosion process, which would be difficult to access with other *ex situ* techniques.[7,8] In contrast to earlier investigations of the roughening behavior of ion eroded GaSb surfaces[9] where only an increase in the roughness with the formation of a dot pattern has been measured, we observe a transition behavior of the surface morphology: an initial smoothing regime is followed by a subsequent roughening of the GaSb surface and the formation of a dot pattern. Simulations of the dynamics of the surface morphology with a continuum approach show that the initial smoothing results from the same relaxation mechanism leading to the formation of the periodic dot structure.

Ion erosion was performed in a compact high vacuum chamber with a base pressure of 10^{-7} mbar with a Kaufman type ion source. The chamber, equipped with a 360° beryllium window for the x-ray scattering measurements,[10] is mounted on a goniometer at the ID01 synchrotron beam line at the ESRF in Grenoble (France). Sputtering was done with Ar^+ ions at normal incidence with an energy of 450 eV and ion flux of 1×10^{15} $cm^{-2}\,s^{-1}$. XRR and GISAXS spectra were measured at an x-ray energy of 8 keV after consecutive steps of sputtering, stopping the erosion process during the measurements. The sample temperature was always below 60 °C during each sputtering step. At these conditions the GaSb surface should be amorphized and in sputter equilibrium after an ion fluence of approx. 1×10^{16} cm^{-2}.[11]

Figure 1(a) shows XRR measurements of the initial, untreated surface and after sputtering with fluences between 3.6×10^{16} and 6.8×10^{17} cm^{-2}. In general, the decay of specular reflected intensity with increasing incidence angle α above the critical angle $\alpha_c \approx 0.32°$ reflects the penetration of the x-ray beam in the material and is, for a perfectly flat surface, given by the Fresnel equations. Surface roughness leads to offspecular diffuse scattered intensity and hence to a faster decay of the specular signal with incidence angle. Finally, the presence of a layer with different optical properties

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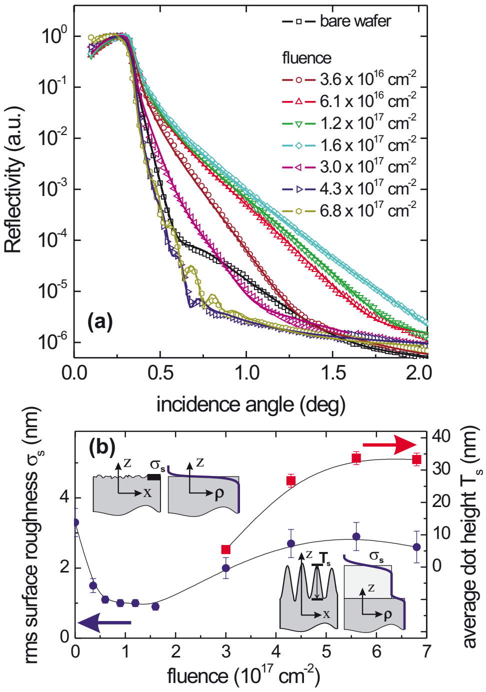

FIG. 1. (Color online) (a) XRR (symbols) of the GaSb surface sputtered with 450 eV Ar$^+$ ions at different ion fluences. The rough initial surface is smoothed in the beginning, followed by a strong roughening process. (b) Evolution of the roughness σ_s and the thickness T_s of the transition layer, respectively, as obtained from a simulation [solid lines in (a)] of the reflectivity curves. Insets illustrate the nature of the transition layer at low and high fluences, respectively.

on top of the surface leads to interference fringes whose angular distance is inversely proportional to the layer thickness.

Prior to sputtering, the XRR measurements reveal a rather rough surface with a thin surface layer which probably corresponds to the native oxide on top of the GaSb.[19] During ion erosion up to a fluence of 1.6×10^{17} cm^{-2} the reflectivity at angles $\alpha > \alpha_c$ increases significantly indicating a smoothing of the surface due to the ion irradiation. The roughening observed for higher fluence can be associated with the onset of a dot pattern formation as visible by GISAXS measurements (see text below). Moreover, the interference fringes observed in the XRR curves in this fluence region indicate the development of a layer with an electron density reduced with respect to the bulk value of GaSb. In order to quantify the surface evolution the reflectivity data are analyzed assuming a transition layer on top of a GaSb substrate that is characterized by a constant (lower) electron density, by its thickness T_s, and by introducing a surface roughness σ_s as a Gaussian-like density gradient into the Fresnel formula [insets in Fig. 1(b)]. Figure 1(b) shows the evolution of σ_s and of T_s, respectively.

At the beginning of sputtering the oxide layer is removed almost immediately and the GaSb surface can be modeled by the surface roughness only, i.e., $T_s=0$. The initial decrease of σ_s reflects the reduction of the surface roughness. With the onset of pattern formation at a fluence of 3×10^{17} cm^{-2} T_s starts to grow until it reaches a stable value of 30 nm, giving a measure for the averaged height of the dot pattern. The simultaneous increase of σ_s can be interpreted as height fluctuations of the dot pattern.

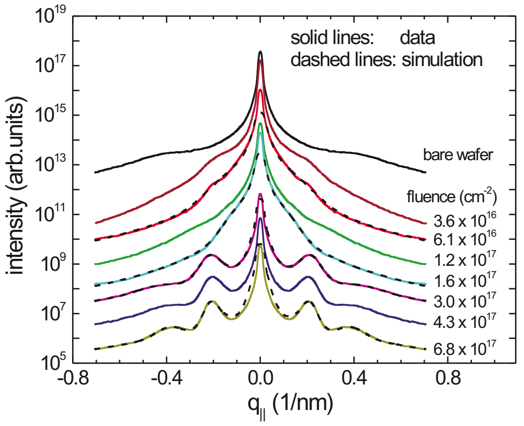

FIG. 2. (Color online) GISAXS spectra of the GaSb sample at different ion fluences.

In addition to XRR, GISAXS spectra were recorded between the sputtering cycles (Fig. 2). Complementary to XRR, these measurements contain information about the lateral height correlations along the surface. At the beginning of the erosion process, the strong diffuse scattering of the substrate is reduced, reaching a minimum at a fluence of 1.6×10^{17} cm^{-2}. At a fluence of 3×10^{17} cm^{-2}, corresponding to the increase of surface roughness as obtained by XRR, satellite peaks appear, indicating the onset of the formation of a periodic pattern. The position of these peaks is related to the mean interdot distance (wavelength) l, whereas number and width of the satellite peaks are related to the correlation length ξ.[12] The dashed lines in Fig. 2 show simulations of the scattered intensity. During the smoothing regime, the diffuse scattering can be modeled taking into account the nonspecular component of the structure factor of a rough surface.[13] For a quantitative analysis of the GISAXS spectra from the periodic dot pattern, the data has been fitted with the program ISGISAXS,[14] using a model of cone-shaped particles and the two-dimensional hexagonal paracrystal model to describe their correlation in a short-range order scenario.[14] The fitting results for the mean wavelength l and the correlation length ξ (not shown) revealed that the wavelength increases only slightly from 30 to 32 nm, whereas the correlation length increases with fluence by about a factor of 2 from roughly 10 to about 20 periods. This increase of ξ is reflected in the decrease of FWHM of the first-order satellite peak and the appearance of second-order peaks in Fig. 2.

In the Bradley–Harper (BH) model of IBS, pattern formation is explained by the interplay of two processes: first, roughening by the curvature dependent sputter yield and second, surface relaxation by surface diffusion.[15] In order to elucidate the smoothing and pattern formation mechanisms at the beginning of the ion sputtering, numerical integrations of a nonlinear extension of the BH model, the Kuramoto–Sivashinsky (KS) equation,[16]

$$\frac{\partial h}{\partial t} = -v_0 + \nu\nabla^2 h - D_{\mathrm{eff}}\nabla^4 h + \frac{\lambda}{2}(\nabla h)^2, \tag{1}$$

have been performed. Here, v_0 is the constant erosion velocity of the planar surface. The second term represents the curvature dependence of the erosion velocity and the third term the effective surface relaxation. The nonlinear fourth term incorporates the dependence of the local erosion velocity on the surface slopes and is responsible for the formation

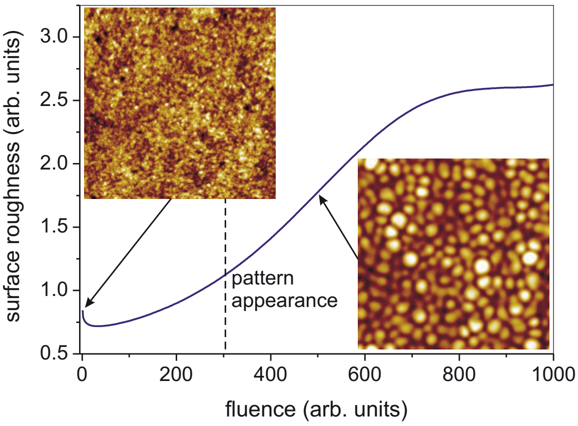

FIG. 3. (Color online) Root mean square roughness and surface morphology obtained by numerical integration of the KS Eq. (1). Note the good qualitative agreement with the measured results [Fig. 1(b)]. X and y scales depend only on simulation parameters and can be rescaled arbitrarily. The size of the AFM type images is $500\times500\ \text{nm}^2$.

of ordered dot patterns at intermediate times.[17,18] However, for long sputtering times, in the nonlinear regime the pattern evolves to kinetic roughening which is not observed experimentally for GaSb. In our integration of the continuum equation a real atomic force microscopy (AFM) image of the virgin GaSb(100) wafer was used as the starting surface (left inset in Fig. 3). The coefficients $\nu=0.15$, $D_{\text{eff}}=1$, and $\lambda=0.1$ were chosen such that the periodicity l is close to the experimental measured periodicity of 30 nm and that the initial dynamics of the pattern evolution can be followed easily. In this way the scales of the initial surface are mapped to the simulation.

Figure 3 shows the evolution of the surface roughness in the simulation. In the initial phase of the simulation the diffusion term leads to a reduction of the roughness at length scales smaller than the wavelength l of the system, whereas at larger length scales roughness is not affected. Therefore, the net effect of the diffusion term is to reduce the rms roughness, calculated as the integral of the power spectral density (PSD), in the beginning. At longer simulation times, the KS instability starts to grow and the pattern exhibits dot features with the wavelength l (right inset in Fig. 3). At fluences around 300 an ordered dot pattern appears which can be identified by a pronounced peak in the PSD. Now the surface is dominated by the pattern and the roughness increases again due to the formation of dots. The wavelength l is given by the ratio between the roughening coefficient ν and the diffusion coefficient D_{eff}. Thus, the same diffusion mechanism that determines the formation of periodic patterns is also responsible for the initial surface smoothing.

In summary, we presented *in situ* measurements of the nanodot formation on an Ar-eroded GaSb(100) surface using XRR and GISAXS. We showed that the sputtering process leads to an initial smoothing of the surface, followed by a roughening process associated with the formation of nanodots. Both the smoothing and roughening behavior can be modeled using the KS equation and are the result of the same diffusion mechanisms.

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[19] The initial root mean square (rms) roughness of commercially available GaSb(100) has a value of ~0.8 nm and is thus much higher than that of, e.g., commercial Si(100) with ~0.2 nm.