

Surface Science Letters

# Ion beam sputtering induced ripple formation in thin metal films

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## Abstract

We have observed the formation of ripples in a number of thin metal films, e.g. Au, Pt, Ag, Cu and Co under  $\text{Ar}^+$  ion beam sputtering at grazing incidence. The structures are found to be quite stable under ambient conditions. The results show that the ripple formation in polycrystalline metallic films relies on the erosion-induced surface instability similar to that in amorphous materials.

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## 1. Introduction

One of the experimental techniques for fabricating submicron-sized well-defined pattern on solid surfaces is the low energy ion beam sputtering. Among the various ion beam induced morphologies, the formation of periodic ripple structures has attracted much research interest in recent years both for understanding the underlying physical mechanisms [1] as well as for potential applications in the field of nanotechnology, e.g. ripple pattern might be exploited as a template for quantum dot formation [2]. Although a large number of works on the ripple morphology have

been carried out in semiconductor materials, little work has been done on metal surfaces [1]. The group of Valbusa et al. [3] was pioneer in observing nanoscale-ripples on single crystals of Cu and Ag, where they utilized the presence of Erlich–Schwoebel (ES) barrier at crystalline step edges to develop ripples along energetically favoured crystallographic directions. Since the diffusion-biased ripples are highly sensitive to the substrate temperature, the ripple morphology is found to be unstable at room temperature, because of the low ES barrier heights [4]. Very recently, Sekiba et al. [5] have shown that in situ oxidation of the rippled surface immediately after the formation may provide a long term stability at room temperature or higher.

Polycrystalline metal thin films have wide industrial applications in electronic, magnetic, and optical devices [6]. It will, therefore, be interesting

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to explore the possibility of formation of correlated surface features such as wavelength selected ripple structures, at sub-micrometre length scales, in these systems. In this paper, we report the development of ripple topography at grazing ion beam sputtering on a number of thin metallic films at ambient temperature.

## 2. Experimental

Thin films of Au, Pt, Ag, Cu and Co were deposited by d.c. magnetron sputtering (Pfeiffer, PLS 500) onto commercially available polished Si(100) wafers, previously degreased and cleaned. The base pressure in the deposition chamber was  $2 \times 10^{-6}$  mbar. The films were deposited in  $2.5 \times 10^{-2}$  mbar Ar gas pressure with a discharge power of 25 W. The film thicknesses were in the range of 30–200 nm. These samples were then sputtered with mass analysed 5–27 keV  $\text{Ar}^+$  ions in a low energy ion beam (LEIB) system developed in the laboratory [7]. The angle of ion incidence with respect to the surface normal was varied from  $10^\circ$  upto  $80^\circ$ . The beam current through a 4 mm diameter aperture was in the range of 2–3  $\mu\text{A}$ . The samples were exposed to ion fluences in the range  $5 \times 10^{15}$ – $5 \times 10^{17}$  ions/ $\text{cm}^2$ , which were measured by a current integrator (Danfysik, model 554) after suppression of the secondary electron emission. The base pressure in the target chamber was less than  $5 \times 10^{-8}$  mbar. The surface morphology of the ion-irradiated samples was examined by a Park Scientific AFM (Auto Probe CP) in the contact mode. All the measurements were carried out in air at room temperature.

## 3. Results and discussion

Atomic force microscopy of the as-deposited films shows that the initial surface topography contains characteristic bump-like structures, which are typical for thin films grown by the process of sputtering [8]. The evolution of surface morphology as a function of the angle of ion incidence for a given ion fluence and energy shows first the development of mound structure which tends to

grow upto the angle of incidence  $\theta \sim 50^\circ$ . Further increase of the incidence angle shows the beginning of distinct changes of the surface morphology which ultimately ends up with regular ripple structures at grazing ion incidence. Fig. 1(a)–(t) show some selected AFM images of the sputtered Co, Cu, Ag, Pt and Au films, respectively, at the angles of  $60^\circ$ ,  $70^\circ$ , and  $80^\circ$ , because at these angles the gradual morphological transitions are clearly visible and this is a general feature for all the metallic films studied in the present sputtering conditions. At  $60^\circ$  weakly pronounced ripples with wave vector parallel to the ion beam direction, especially in Co and Cu films, appear. At  $70^\circ$  the morphology shows the development of arrays of tiny cones aligned along the projection of the ion beam direction. Finally, at  $80^\circ$  regular ripple-like surface instability with the wave vector perpendicular to the ion beam direction is developed. In passing we mention that the AFM images of the ripples including that of the other morphological structures are found to be quite reproducible even after several weeks of bombardment.

For the quantitative analysis of the ripple morphology, we have calculated numerically the height–height autocorrelation function  $C(\mathbf{r}) = \langle [h(\mathbf{r})h(0)] \rangle$ , where  $h(\mathbf{r})$  is the relative surface height at the position  $\mathbf{r}$  and  $\langle \rangle$  denotes an average over all positions and directions. As an illustration, Fig. 2 shows a typical AFM image of the rippled structure on a Au film together with the corresponding 2D-autocorrelation function. The ripple wavelength  $\lambda$  is defined as the separation between the central peak and the first correlation maximum while taking linear scans of  $C$ . Fig. 3 shows a typical set of data for the ripple wavelength  $\lambda$  as a function of the substrate material when sputtered with the same total ion fluence and energy.

Valbusa et al. [3,4] showed that sputtering of metal surfaces involves two types of surface instabilities depending on the angle of ion incidence,  $\theta$ : the first one arising from erosion process and the other deriving from anisotropic surface diffusion. The erosion-induced surface instability dominating at grazing incidence  $\theta > \theta_c$  ( $\theta_c \approx 50$ – $70^\circ$  [9]) leads to ripple structures aligned parallel to the ion beam projection, independent of the sur-

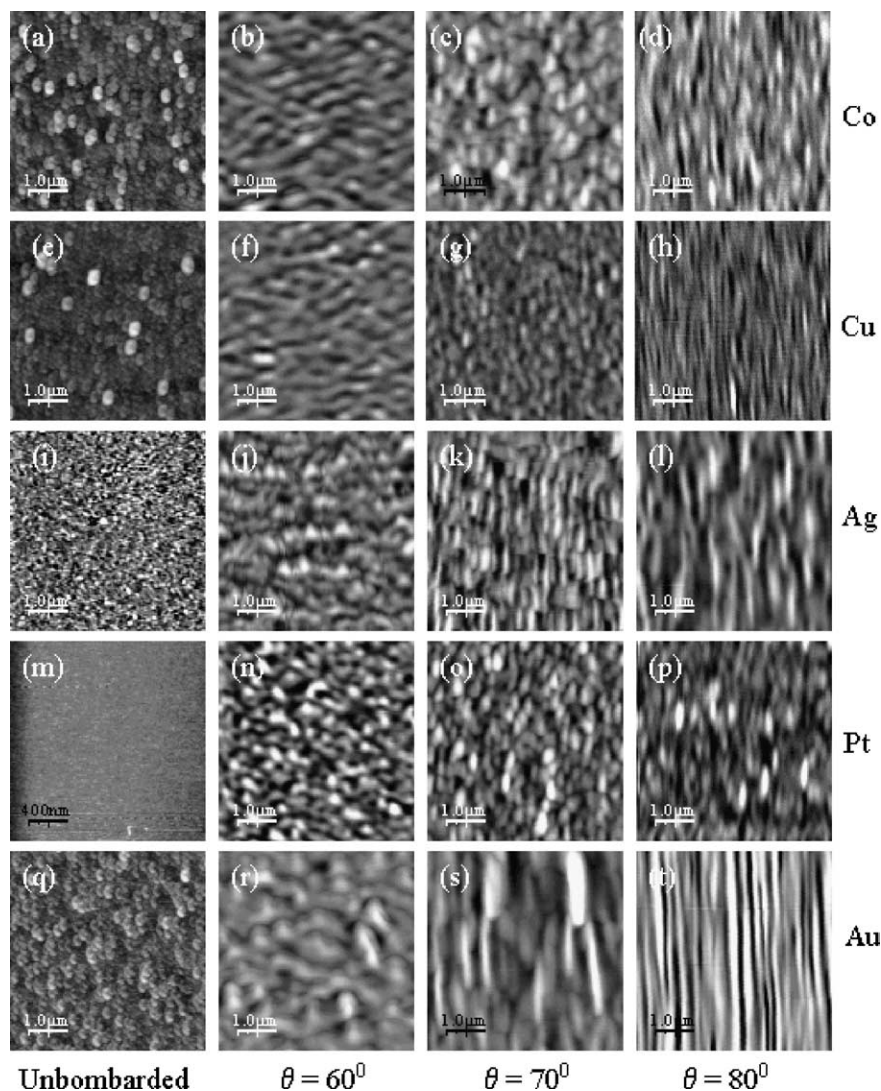


Fig. 1. AFM images of the unbombarded and 16.7 keV  $\text{Ar}^+$  sputtered Co, Cu, Ag, Pt and Au surfaces at different angles of incidence  $\theta$  as indicated. The bombarding fluence  $\phi$  for Co, Cu, Ag, and Au is  $1 \times 10^{17}$  ions/cm<sup>2</sup>, while that for Pt is  $5 \times 10^{16}$  ions/cm<sup>2</sup>. The ion beam direction is from the bottom to the top.

face crystallinity or orientation. The erosive regime is supposed to govern by the Bradley and Harper (BH) theory [10], where the surface height evolution  $h(x, y, t)$  can be described as [4,10]

$$\frac{\partial h}{\partial t} = -v_0 + \gamma \frac{\partial h}{\partial x} + v_x \frac{\partial^2 h}{\partial x^2} + v_y \frac{\partial^2 h}{\partial y^2} - K \nabla^4 h + \eta, \quad (1)$$

where  $v_0$  is the surface erosion rate of the flat surface at normal incidence,  $\gamma$  is related to the derivative of the sputtering yield with respect to the angle of ion incidence,  $v_x$  and  $v_y$  are the effective surface tensions generated by the surface erosion process, the constant  $K$  is related to the surface diffusion which is activated by different physical processes, namely, thermal or ion beam

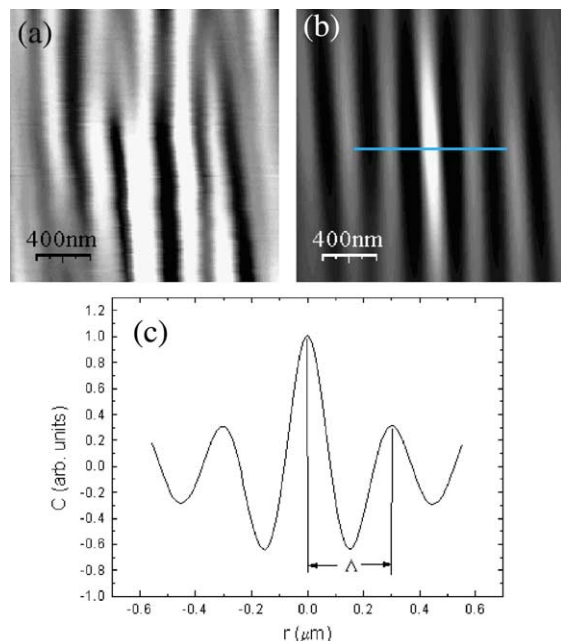


Fig. 2. (a) AFM image of the rippled surface on Au film after 16.7 keV  $\text{Ar}^+$  ion sputtering at  $\theta = 80^\circ$  and  $\phi = 1 \times 10^{17}$  ions/ $\text{cm}^2$ ; the ion beam direction is from the bottom to the top. (b) showing the corresponding 2D-autocorrelation function. (c) showing the 1D-autocorrelation function along the marked line in (b) in order to determine  $\Delta$ .

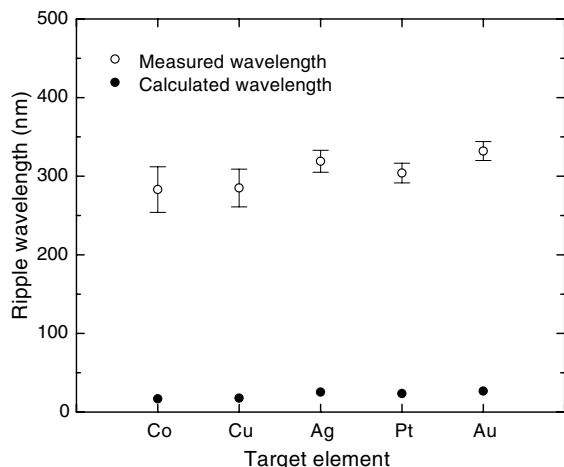


Fig. 3. The ripple wavelength  $\Delta$  versus the target element after 16.7 keV  $\text{Ar}^+$  ion sputtering at  $\theta = 80^\circ$  and  $\phi = 1 \times 10^{17}$  ions/ $\text{cm}^2$ .

induced or both [1] and, finally  $\eta$  is the noise term associated with the randomness of the bombarding ions.

Eq. (1) can further be extended to crystalline materials by including the effects of anisotropic diffusion in different crystallographic directions as well as the existence of the ES barrier at the step edges [11]. One of the consequences of the presence of the ES barrier is that the ripple structure in metal surfaces could be formed even at normal ion beam incidence ( $\theta \approx 0^\circ$ ) by tuning the surface temperature [11]. However, for polycrystalline thin films, the grains are mainly randomly oriented and the grain sizes are usually much smaller compared to the film thickness [12,13]. For such a system, the existence of ES barrier is improbable because of the lack of well-defined atomic steps at the surface [12]. Therefore, the approximation of isotropic diffusivity as in amorphous materials seems to be well held in polycrystalline metallic films. The stability of the ripple structure at room temperature indicates that the thermally activated diffusion energy barriers in thin polycrystalline films are comparatively higher than that in monocrystalline metal surfaces. Rossnagel and Robinson [14] measured the activation energy for adatom surface diffusion on various polycrystalline materials from the Arrhenius plot of the sputter cone spacings for several temperatures. The barrier heights lie typically in the range of 0.3–1 eV. Similar plots for ripple wavelengths on Ag single crystals yield activation energy around 0.15 eV [4].

At room temperature the surface diffusion is driven by the collisional effects rather than pure thermal effect [15]. The mobility of adatoms is believed to originate from the overlapping collision cascades due to multiple ion impact [16]. Carter and Vishnyakov [17] proposed to add a ballistic smoothing term of the form  $|A(E, \theta)| \nabla^2 h$  in Eq. (1) in order to account the effect of recoiling-adatom diffusion induced by ion irradiation at a given energy  $E$ . More recently, Makeev et al. [1] showed that fourth-order derivatives of the surface height function  $h(x, y)$  may also cause the smoothing effect which does not involve real mass transport. For such a case the wavelength of the ripples, in the present experimental conditions,

can be derived as  $\Lambda = 2\pi\sqrt{\frac{2D_{yy}}{|v_y|}}$ , where  $D_{yy}$  is the ion induced smoothing coefficient in the  $y$  direction as defined in Eq. 52 of Ref. [1]. The ratio  $D_{yy}/v_y$  can be estimated from the values of the ion penetration depth  $a$  and the longitudinal and lateral straggling widths  $\sigma$  and  $\mu$ , respectively, using the computer code SRIM [18]. Although the experimental data at fixed bombarding ion energy and fluence follow the same trend as the theoretically calculated  $\Lambda$  in different target elements, the theory underestimates substantially the experimental wavelength values (cf., Fig. 3). One of the reasons for this discrepancy is that the ripple wavelength  $\Lambda$  coarsens with the ion fluence  $\phi$  as a power law  $\Lambda \sim \phi^n$  with the exponent  $n = 0.53$ , e.g. measured for Pt films [19], whereas the BH theory predicts a fixed ripple wavelength. Nevertheless, such a large discrepancy simply indicates the failure of the model which is based on curvature-dependent sputtering for the calculation of  $\Lambda$ . Recently, several authors [20–22] have reported that the characteristic wavelengths of the generated sputtered patterns on various semiconductor targets are a factor of 4–10 higher than the theoretically expected value.

It is worth mentioning that the ripple formation does not depend on the initial surface topography, i.e. whether the initial surface is atomically flat or rough, the surface is always characterised by ripples at grazing ion beam sputtering. Such a result is due to the effect of the noise term in Eq. (1), which operates at the early time of bombardment. The random arrival of the bombarding ions destroys the old surface morphology followed by the development of new morphology evolving with the erosion time [23].

Finally, we have also bombarded clean Si(100) wafers in the angular range of 10–80° under identical conditions. Here, ripples are formed only at the ion incidence angle of 60° and the wave vector of the ripples is found to be parallel to the ion beam direction (Fig. 4). The most interesting observation, however, is that the Si ripples are generated at fluences  $>10^{17}$  ions/cm<sup>2</sup> in agreement with that of others, e.g. [24], in contrast to the metallic ripples which begin to develop at much lower fluences of about  $10^{15}$  ions/cm<sup>2</sup>.

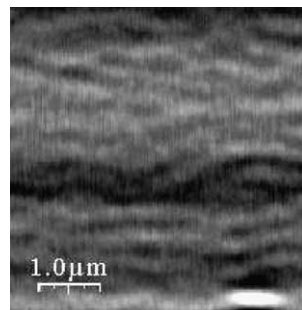


Fig. 4. AFM image of the ripples formed on a Si(100) surface after 16.7 keV Ar<sup>+</sup> ion sputtering at  $\theta = 60^\circ$  and  $\phi = 5 \times 10^{17}$  ions/cm<sup>2</sup>. The ion beam direction is from the bottom to the top.

#### 4. Conclusion

In conclusion, grazing ion beam sputtering at room temperature can induce ripple structures on thin metallic films similar to that reported on single crystal metal surfaces at low temperature [4]. Unlike the latter, such ripples are found to be quite stable at ambient conditions. The present experiment also indicates that the ripples do not form, and the surface undergoes kinetic roughening so long as  $\theta < \theta_c$ , where  $\theta_c \simeq 50$ – $60^\circ$ . Finally, it is noted that, independent of the initial morphology of the surface, the ripples in metallic films start to generate at ion fluences as low as  $10^{15}$  ions/cm<sup>2</sup> which is roughly two orders of magnitude smaller than that for the ripples formed on Si surfaces.

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