Scaling Laws of the Ripple Morphology on Cu(110)

S. Rusponi, G. Costantini, C. Boragno, and U. Valbusa

INFM—Unità di Ricerca di Genova e Centro di Fisica delle Superfici e delle Basse Temperature del CNR, Dipartimento di Fisica dell' Università, Genova, Italy

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The evolution of the Cu(110) surface morphology during low temperature (180 K) ion sputtering was studied as a function of the incident ion beam angle θ by means of scanning tunneling microscopy. The morphology was dominated by a ripple structure with the wave vector parallel or perpendicular to the direction of the incident beam. The time evolution of the interface shows that the ripple wavelength increases in time following a scaling law $\lambda \propto t^z$, with $z = 0.26 \pm 0.02$. These results are ascribed to the effect of a Schwoebel barrier on the interlayer diffusion of the recoiling atoms produced during ion sputtering. [S0031-9007(98)07515-2]

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The understanding of the erosion process induced by ion sputtering on surfaces is of great interest since this method is commonly used in analytical techniques or to produce structures on the nanometer scale. Quite recently there has been a great effort in performing both experimental as well as computer simulation studies in order to explain the appearance of peculiar morphologies created by ion bombardment, such as ripples. It has been known for a long time that such ripples can develop after ion bombardment (usually at room temperature and at off-normal incidence) on glass [1], amorphous films [2], and semiconductors [3–5], but satisfactory models explaining this effect have been developed only in recent years [4,6–9]. In all these models the interface is described by a continuum function (the height h in respect to a reference plane) and its variation in space and time is studied without reference to the atomistic processes involved during sputtering. The structures are interpreted as interface instabilities caused by the sputtering erosion, dependent on the local surface curvature, which competes with the (low) surface diffusion. Many experiments on metals have been reported, all performed in quite similar experimental conditions (ion energy 1–5 keV and ion flux 5–50 μA/cm^2). Hexagonal pits on Pt(111) [10] and Cu(111) [11] or square holes on Ag(001) [12] and on Cu(001) [13] have been observed, reflecting the atomic surface symmetry without any evidence of ripple structures. Similar results have been found also on crystalline Fe films [14]. In a recent Letter [15] we observed a ripple structure on a Ag(110) single crystal (1 keV Ar^+ ion bombardment). Three features distinguish this surface morphology from those observed on semiconductor or amorphous materials [16]: (a) It appears after sputtering at normal incidence [$\theta = 0°$, cf. Fig. 2(a)]; (b) it is always aligned along $\langle 1\bar{1}0\rangle$, independently on the ion beam direction at least for $\theta \leq 30°$ and for different surface orientations δ [cf. Fig. 2(a)]; (c) it appears only in a restricted temperature range (280–320 K). We attributed this difference to the fact that, in metals, surface diffusion prevails over erosion in determining the final morphology [15], while in nonmetals the morphology is mainly caused by erosion [8]. Since the substrate temperature can be varied in order to suppress different diffusion terms governing the surface evolution, as previously shown by Chason *et al.* [4], we expect that also in metals it should be possible to induce a ripple structure *having the same characteristics* observed in amorphous and semiconductor materials simply depressing diffusion rates by lowering the temperature. In order to check this possibility we performed ion sputtering experiments on Cu(110) in a wide temperature range.

The experimental setup as well as the method used has been described elsewhere [15,17]. The Cu(110) sample, with a miscut $<0.15°$ [18], presents terraces of the order of 100 nm or larger. The crystal has been sputtered by 1 keV Ar^+ ions at a temperature T_S in the range 180–450 K and subsequently observed with an ultrahigh vacuum-scanning tunneling microscope (UHV-STM) after a fast cooling to $T \cong 100$ K. After each measurement the substrate was reprepared by annealing at 900 K. The surface morphology has been analyzed as a function of the ion flux Φ, the sputtering time t, ion incidence angle θ, and surface orientation δ [cf. Fig. 2(a)]. For $\theta = 0°$, $T_S = 300$–320 K and $\Phi = 7$ μA/cm^2, the morphology is similar to that observed on Ag(110) [15], with ripples aligned along the fast diffusion direction $\langle 1\bar{1}0\rangle$ (Fig. 1). On the contrary, for $T_S \leq 180$ K, we observe a ripple structure whose orientation depends on the ion beam direction. In Fig. 2, the morphology of the Cu(110) surface after ion sputtering at different incidence angles θ is reported. For all images $T_S = 180$ K, $\Phi = 5$ μA/cm^2, $t = 15$ min, and $\delta = 0°$. At normal incidence [Fig. 2(b)] the surface appears rough, without evidence of any periodic structure. At $\theta = 15°$ [Fig. 2(c)] the morphology changes significantly. A regular pattern of ripples is now present on the surface, having the wave crests aligned (within 10°) to the $\langle 001\rangle$ direction and almost perpendicular to the direction of the incident ion beam. The ripple wavelength λ can be calculated from the two-dimensional map of the height-height correlation

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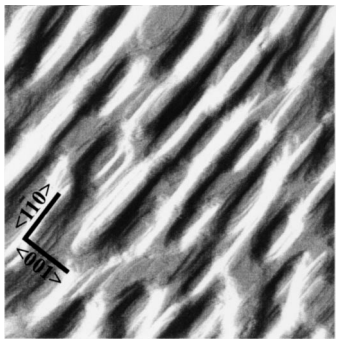

FIG. 1. Surface morphology (size 400×400 nm^2) after ion sputtering at normal incidence ($\theta = 0°$). The values of the experimental parameters are $T_S = 320$ K, $\Phi = 7\ \mu$A/cm^2, and $t = 15$ min. A ripple structure aligned along the $\langle 1\bar{1}0 \rangle$ direction is clearly visible.

function $G(\mathbf{r})$ as described in [19]. $G(\mathbf{r})$ is defined as [7]

$$G(r) = \langle h(r_1)h(r_1 + r)\rangle - \langle h(r_1)\rangle^2, \qquad (1)$$

where $h(\mathbf{r})$ is the height of the interface at position $\mathbf{r}$ and $\langle\ \rangle$ indicates a spatial average. For each image, we set $\langle h(r)\rangle = 0$ by subtraction of the best-fit plane. The 2D $G(\mathbf{r})$ can be easily calculated via the 2D fast-Fourier transform algorithm and its power spectrum [19,20]. For $\theta = 15°$, λ results 13 ± 2 nm, while the mean peak-to-peak depth amplitude of the ripples is about 0.7 nm, corresponding to more than 5 Cu layers. Increasing the incidence angle up to 55° [Fig. 2(d)] the periodic pattern does not change its characteristics; at $\theta \cong 60°$ the periodic structure disappears [Fig. 2(e)]. Finally for $\theta = 70°$ [Fig. 2(f)] the ripple structure rotates by 90°, with the wave crests parallel to the ion beam direction. Now the spatial period is 16 ± 4 nm, while the depth is of about 0.4 nm. The ripple rotation, observed varying θ, does not depend on the ion flux or ion fluence, at least in the range studied in the present work ($1.5 \leq \Phi \leq 16\ \mu$A/cm^2 and $1 \leq t \leq 240$ min).

In order to quantitatively characterize the observed morphology, we study the scaling properties of the interface as done in [21–23] for surfaces with a periodic (mound) morphology. First we followed the surface evolution *versus* the sputtering time. In Fig. 3 we report four images taken at different times, in the following experimental conditions: $\Phi = 16\ \mu$A/cm^2, $T_S = 180$ K, $\theta = 45°$, and $\delta = 0°$. The interface roughness W, defined as the root mean square of the surface height, scales as $W \propto t^{\beta}$, with $\beta = 0.43 \pm 0.08$ and does not saturate in more than two decades (Fig. 4). Also for $\theta = 70°$ no saturation has been found ($\beta = 0.20 \pm 0.05$), while for normal incidence W seems to saturate at a low value ($\cong$4 atomic layers) in about 100 sec with $\beta \cong 0.6$ [24]. We also calculated the dependence of W on the system size L, finding the scaling law $W \propto L^{\alpha}$. We determined α from our STM images following the methods reported in [14,25]. For all the values of θ considered, we found an algebraic scaling of the interface characterized by α that increases with time from 0.5 to 0.9. Finally, we observed a time scaling behavior also for the ripple wavelength $\lambda \propto t^z$ with z depending on θ. We found $z = 0.26 \pm 0.02$ at $\theta = 45°$ (Fig. 4) and $z = 0.13 \pm 0.03$ at $\theta = 70°$.

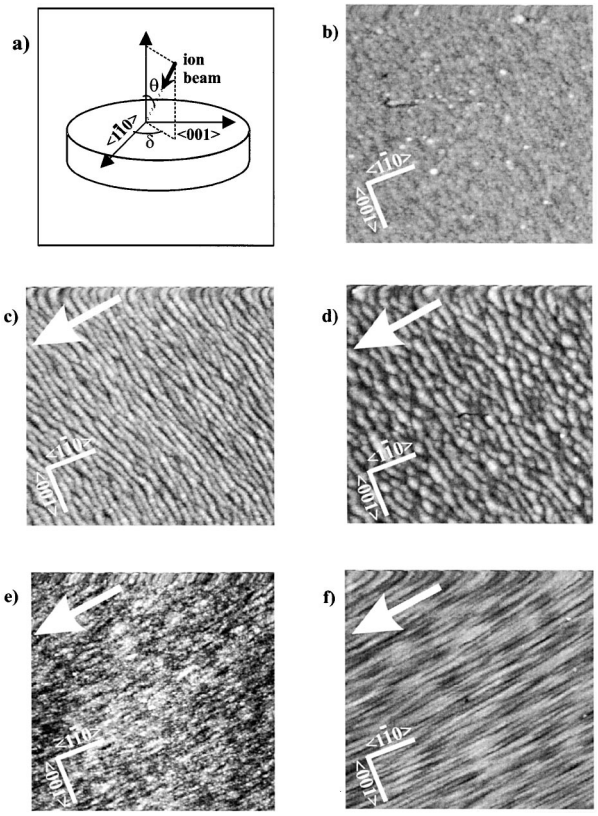

FIG. 2. Five images (size 400×400 nm^2) of the Cu(110) surface after ion sputtering at different impact angles θ. The values of θ are 0°, 15°, 55°, 65°, and 70°, respectively, for (b), (c), (d), (e), and (f). For all images $T_S = 180$ K, $\Phi = 5\ \mu$A/cm^2, $t = 15$ min, and $\delta = 0°$. The white arrow indicates the ion beam direction. The inset (a) shows the experimental geometry.

The results we presented indicate that ion sputtering *at low temperature* can induce also on a single crystal metal surface a ripple structure similar to that produced on amorphous or semiconductor materials. In fact, our results show that at $\theta = 0°$ no ripples are present on the surface, while for $\theta > 0°$ their orientation is related to the ion beam direction. For $\delta = 0°$, an abrupt rotation by 90° occurs if θ is greater than a critical value ($\theta_c \cong 60°$), according to the continuum theory for amorphous systems developed by Cuerno and Barabasi [8] (in the following referred to as the CB model). However, the observed scaling exponents are different from the values calculated in the CB model which does not predict any coarsening effect in time ($z = 0$) and foresees $\alpha \cong 0$ for $\theta > \theta_c$.

Barabasi and Stanley [26] have pointed out that sputtering erosion and atom deposition are two processes with a number of common features. This allows us to describe

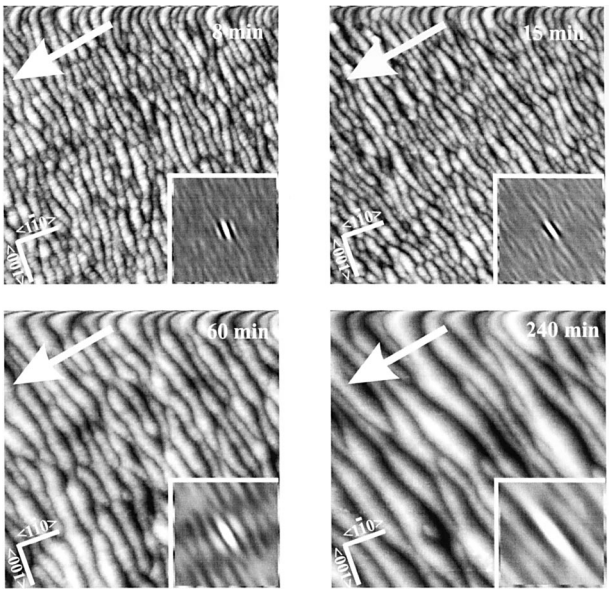

FIG. 3. Four images (size 400×400 nm^2) of the Cu(110) surface after ion sputtering at different sputtering times. For all images, $T_S = 180$ K, $\theta = 45°$, $\delta = 0°$, and $\Phi = 16\ \mu$A/cm^2. The insets report the corresponding $G(\mathbf{r})$ (area 100×100 nm^2).

the surface evolution during sputtering by means of equations formally identical to those describing surface growth, as shown in [8]. In particular, in both cases, the surface evolution is determined by atomistic processes involving deposited adatoms (during growth) or adatoms and vacancies created by the ion impacts (during sputtering). In fact it has been demonstrated by Teichert *et al.* [27] that a large number of adatoms are created on a surface during ion sputtering. They estimated that, on Pt(111), each 5 keV Xe^+ ion projectile produces about 65 adatoms located within an average distance of 25 Å from the impact point, but in some cases they found islands containing up to 500 adatoms. They also noted that these adatoms nucleate in monatomic height islands, indicating that they diffuse significantly on the surface and contribute to the final surface morphology. Also in the present case the adatoms created by ion impacts represent the main diffusing species, while vacancies do not contribute significantly to the mass redistribution as their energy diffusion barrier is too high [28]. Looking for an analogous result in growth experiments, we note that the observed scaling laws appear very similar to those measured on substrates with a Schwoebel barrier at the step edges [21,23,29,30]. In fact a relevant mound coarsening with an exponent $z \cong 0.25$ has been observed during homoepitaxial growth on Fe(001) [29,30] while $\alpha \cong 1, \beta \cong 0.5$, and $z \cong 0.25$ have been reported for Cu deposition on Cu(001) [21,23]. Identical results have been also obtained in computer simulations [31,32]. At grazing impact angles ($\theta > \theta_c$), we found that λ still follows a time scaling law but with a smaller value of

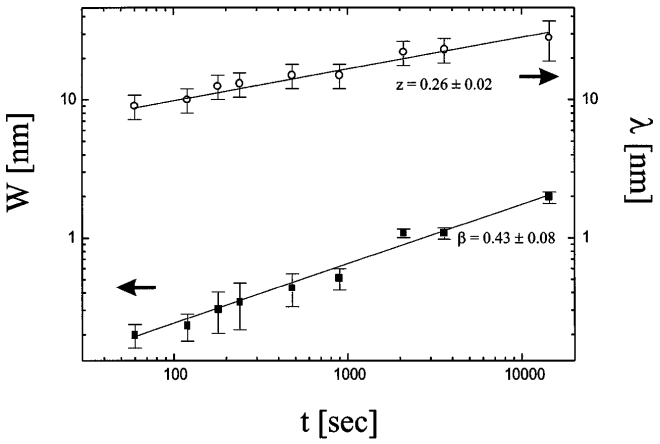

FIG. 4. The ripple wavelength λ and the surface roughness W as a function of the sputtering time t. $T_S = 180$ K, $\theta = 45°$, $\delta = 0°$, and $\Phi = 16\ \mu$A/cm^2.

the z coefficient ($z = 0.13$). Similar values have been found, at the initial growth stages, both in a growth experiment [30] and in computer simulations [32,33]. The previous resemblance suggests that a Schwoebel barrier has to have a leading effect also during sputtering. In fact the adatoms that reach a step edge feel a Schwoebel barrier ($E_{S001} = 0.27$ eV and $E_{S110} = 0.19$ eV, respectively, along $\langle 001 \rangle$ and $\langle 1\bar{1}0 \rangle$ [28]) which, limiting the interlayer mass transport, produces an uphill adatom current that leads to an increase of the local slope, i.e., to a surface instability. In order to account for this effect we suggest to introduce in the CB model the term

$$-S_{110}\,\frac{\partial^2 h}{\partial x^2} - S_{001}\,\frac{\partial^2 h}{\partial \gamma^2} + \text{nonlinear terms}, \qquad (2)$$

where $S_{001} \propto 1 - \exp(-E_{S001}/kT)$ and $S_{110} \propto 1 - \exp(-E_{S110}/kT)$ are positive coefficients [31,34,35].

The linear part of Eq. (2), having the same functional dependence as the erosion terms in the CB model, determines the growth of surface instabilities and their temperature evolution as shown in [36]. The nonlinear terms in Eq. (2), as discussed in [31,34,35], control the time evolution of the ripple structure inducing a coarsening effect [37].

For normal sputtering we do not observe any surface instability. A similar result has been obtained by Carter and Vishnyakov [38] during high energy, near-normal incidence Xe^+ sputtering of Si. They explain the absence of periodic structures by including in the CB model a smoothing term, effective only for a small value of θ, accounting for the atomic displacement induced by irradiation. We think that for $\theta = 0°$ this sputtering-induced diffusion term can dominate both previous instabilities and thus does not allow the formation of periodic features [36].

Finally, we are not able to measure the effects of the implanted ions on the final surface morphology. However, since the implanted ion number as well as the radiation damage are expected to be temperature independent and,

on the contrary, Figs. 1 and 2 show very different morphologies, we can exclude any relevant role of the ion implantation on the surface evolution, at least in first approximation.

In conclusion, the nature and behavior of the ripple morphology induced by ion sputtering at low temperature on Cu(110) has been studied. The results indicate that for a full description of the erosion process on single crystal metals it is necessary to introduce in the CB model [8] a most realistic diffusion term, taking into account the presence of a Schwoebel barrier.

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