

Temperature and Time Evolution of Ripple Structure Induced by Ion Sputtering on Cu(110)

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

INFM - Unita' di Ricerca di Genova e Centro di Fisica delle Superfici e delle Basse Temperature del CNR

Dipartimento di Fisica dell' Universita', Genova Italy

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Abstract

The surface morphology generated by ion sputtering on a Cu(110) crystal has been investigated by Scanning Tunneling Microscopy (STM). A ripple structure is observed for all the considered values of the incident ion beam angle θ ($0^\circ < \theta < 70^\circ$). In particular normal sputtering produces a well defined ripple structure whose wave vector rotates from $\langle 001 \rangle$ to $\langle 1 \bar{1} 0 \rangle$ by increasing the substrate temperature. Moreover for $\theta = 45^\circ$ the ripple wavelength λ increases in time following a scaling law $\lambda \propto t^z$, with $z=0.26 \pm 0.02$. These results are described by a continuum equation which includes, in addition to the surface curvature dependent erosion terms, a diffusion term that takes into account both the surface anisotropy and the effect of an Ehrlich-Schwoebel barrier.

The evolution of surface morphology during ion sputtering is a complex phenomenon which includes roughening and smoothing processes. On amorphous materials [1,2] and semiconductors [3-5] off normal ion sputtering at room temperature generates a modulation of the surface (ripples) with a wave vector that depends on the ion incidence angle θ , while for normal incidence ($\theta \cong 0^\circ$) no periodic structures are observed. These results have been theoretically explained in terms of a linear instability caused by the surface curvature dependent sputtering, which competes and dominates the smoothing due to the thermal surface diffusion [6,7]. On single crystal metals, on the contrary, surface diffusion seems to be the leading process since ion sputtering, in quite similar experimental conditions, produces features that reflect the substrate symmetry without any relationship with the ion beam direction: square pits have been observed on Cu(001) [8] and Ag(001) [9], hexagonal ones on Pt(111) [10], Au(111) [11] and Cu(111) [12], ripples with crests oriented along the fast diffusion direction $\langle 1 \bar{1} 0 \rangle$ on Ag(110) [13].

In order to clarify the characteristics of the competition between surface diffusion and ion erosion we performed ion sputtering experiments on Cu(110) in a wide temperature range. The experimental set-up as well as the preparation and measure methods have been described elsewhere [13,14]. The Cu(110) sample, with a miscut $< 0.15^\circ$, 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 variable over the range $180 \div 450$ K and subsequently observed with an Ultra High Vacuum STM after a fast cooling to $T \cong 100$ K. After each measurement the substrate was re-prepared by annealing up to 900 K. The surface morphology has been analyzed as a function of ion flux Φ , sputtering time t , ion incidence angle θ and surface azimuthal orientation δ (fig. 1).

*Corresponding author. Tel. ++39-10-3536356 ; Fax ++39-10-3622790 ; email :boragno@server1.fisica.unige.it

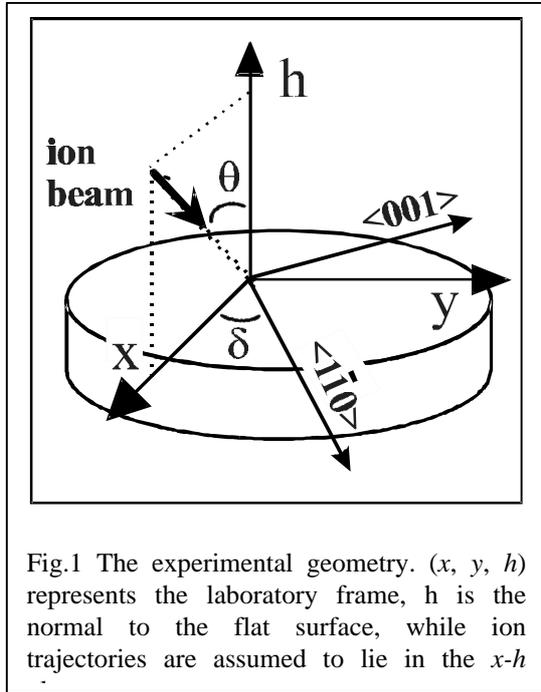


Fig.1 The experimental geometry. (x, y, h) represents the laboratory frame, h is the normal to the flat surface, while ion trajectories are assumed to lie in the x - h

Off-normal ion bombardment ($\theta = 45^\circ$) at low temperature ($T_s = 180$ K) produces a ripple structure similar to that observed on amorphous or semiconductor materials (fig. 2a). In this experimental conditions we have followed the dependence of the surface evolution versus the sputtering time. We observed a time scaling behavior for the ripple wavelength $\lambda \propto t^z$ with $z = 0.26 \pm 0.02$ (fig. 2b), while 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. Differently to the experiments on amorphous materials, a periodic ripple structure is observed also for normal sputtering. In fig.3 we show the results obtained for $\theta = 0^\circ$, $\delta = 0^\circ$ and different T_s . At the lowest temperature considered ($T_s = 180$ K) the surface is rough (fig. 3b). For T_s in the range 250 - 270 K the surface morphology is characterized by a well defined ripple structure (wavelength $\cong 20$ nm) with the crests aligned along $\langle 001 \rangle$ (fig. 3c). A little increase in the sputtering temperature ($T_s = 320$ K) produces a degradation of this structure (fig. 3d). If T_s is further increased in the range 350 - 360 K a second ripple structure (wavelength $\cong 100$ nm) appears (fig. 3e), with the crests along $\langle 1 \bar{1} 0 \rangle$ (90° ripple rotation). Finally, at higher temperatures a quasi layer-by-layer erosion is observed (fig. 3f). Similar results have been also reported on Ag(110) [13].

Both the ripple coarsening (fig. 2b) and the ripple rotation observed for normal sputtering (fig. 3) are not predicted by the continuum model recently proposed by Cuerno and Barabasi [7] in order to account for the experimental observations on amorphous material. The coarsening exponent $z = 0.26 \pm 0.02$ is very close both to the value 0.25 predicted by continuum models for mound formation in molecular beam epitaxy (MBE) [15,16] and to values experimentally measured on substrates endowed with a step-edge Schwoebel barrier [17-20]. In fact a relevant mound coarsening with an exponent $z \cong 0.25$ has been observed during homoepitaxial growth on Fe(001) [19,20] while $\beta \cong 0.5$ and $z \cong 0.25$ have been reported for Cu deposition on Cu(001) [17,18]. Identical results have been also obtained in computer simulations [21]. The previous resemblance suggests that a Schwoebel barrier has to have a leading effect also during sputtering. A Schwoebel barrier [22], limiting the interlayer mass transport (adatom and/or vacancy), produces an uphill adatom current that leads to an increase of the local slope, i.e. to a surface instability. This diffusion instability can overcome the one due to ion erosion and thus determines the final surface morphology. A similar explanation was recently proposed also by Ramana Murty et al. [11] to account for pit coarsening in sputtered Au(111).

Following this idea, in order to explain the present data and, more generally, the morphology of all the metal sputtered surfaces, we suggest to modify the diffusive term $-D\nabla^2(\nabla^2 h)$ in the Cuerno-Barabasi model [7]. In fact this term describes only an isotropic diffusion on a flat surface and does not take into account that the adatom (or vacancy) mobility on a single crystal metal surface is actually biased both in the vertical direction, by a Schwoebel barrier at the step edges, and in the surface plane, by a diffusion rate which, in the case of (110), is different along the two crystallographic directions. In the linear approximation and in the simple case of $\delta = 0^\circ$ ($\langle 1 \bar{1} 0 \rangle$ parallel to the x -axis (fig. 1)), the diffusion term has the following form:

$$-S_{001} \frac{\partial^2 h}{\partial y^2} - S_{\bar{1}\bar{1}0} \frac{\partial^2 h}{\partial x^2} - D_{001} \frac{\partial^4 h}{\partial y^4} - D_{\bar{1}\bar{1}0} \frac{\partial^4 h}{\partial x^4} \quad (1)$$

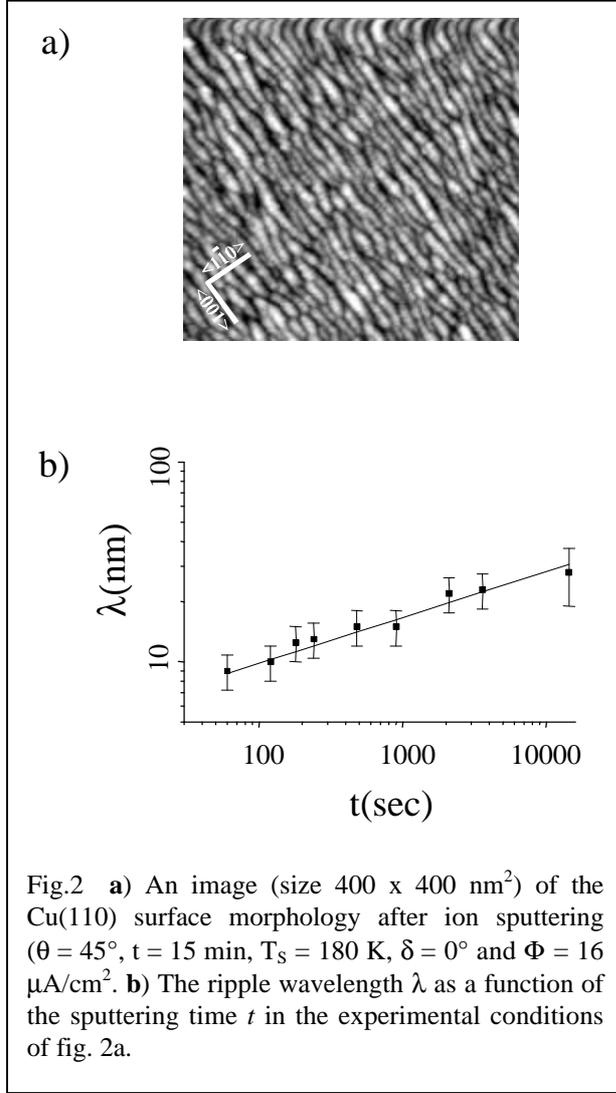


Fig.2 a) An image (size 400 x 400 nm²) of the Cu(110) surface morphology after ion sputtering ($\theta = 45^\circ$, $t = 15$ min, $T_s = 180$ K, $\delta = 0^\circ$ and $\Phi = 16$ $\mu\text{A}/\text{cm}^2$). b) The ripple wavelength λ as a function of the sputtering time t in the experimental conditions of fig. 2a.

where all the coefficients are positive. $S_{001} \propto 1 - R_{001}$ and $S_{\bar{1}\bar{1}0} \propto 1 - R_{\bar{1}\bar{1}0}$ [23] where $R_{001} = e^{-E_{S001}/kT}$ and $R_{\bar{1}\bar{1}0} = e^{-E_{S\bar{1}\bar{1}0}/kT}$ are the ratio of the probability to hop to a lower layer versus the probability to bound back on the terrace; $D_{001} \propto e^{-E_{D001}/kT}$ and $D_{\bar{1}\bar{1}0} \propto e^{-E_{D\bar{1}\bar{1}0}/kT}$ [6,24] where E_{D001} and $E_{D\bar{1}\bar{1}0}$ represent the energy barriers for surface diffusion respectively along $\langle 001 \rangle$ and $\langle \bar{1} \bar{1} 0 \rangle$. Since in Cu(110) the activation energy for vacancy diffusion is higher than the one for adatoms [25], we limit ourselves to consider only the mobility of adatoms generated by ion impacts. In this case $E_{S001} = 0.27$ eV, $E_{S\bar{1}\bar{1}0} = 0.19$ eV, $E_{D001} = 0.42$ eV and $E_{D\bar{1}\bar{1}0} = 0.29$ eV [25]. Following ref. [26] we have assumed a constant prefactor for all the atomistic processes $\cong 10^{12} \text{ sec}^{-1}$. The first two terms in (1) account for the asymmetry in the inter-layer diffusion generated by the presence of a Schwoebel barrier at the step edges [15,16,23,27] while the last two describe the diffusion on a flat surface [24,28,29].

With this change, the linear equation for the time evolution of the surface height h [7] during *normal sputtering* ($\theta = 0^\circ$) becomes

$$\frac{\partial h}{\partial t} = -v_0 + \gamma \frac{\partial h}{\partial x} + (v - S_{\bar{1}\bar{1}0}) \frac{\partial^2 h}{\partial x^2} + (v - S_{001}) \frac{\partial^2 h}{\partial y^2} + |A(E, \theta)| \nabla^2 h - D_{\bar{1}\bar{1}0} \frac{\partial^4 h}{\partial x^4} - D_{001} \frac{\partial^4 h}{\partial y^4} + \eta \quad (2)$$

since, for $\theta = 0^\circ$, $v_x = v_y = v$. The term $|A(E, \theta)| \nabla^2 h$ has been suggested by Carter and Vishnyakov [30] to account for the smoothing effect due to the recoiling adatom diffusion induced by irradiation at a given energy E , while the coefficients γ and v depend on Φ , θ and the deposited energy distribution (see eq. (5) of ref. [7]). Similarly to what is discussed in ref. [7], a Laplacian term with a negative coefficient leads to a ripple-like instability, in the time evolution of h , characterized by a wave vector k oriented along the direction (x or y) for which the absolute value of the coefficient of the Laplacian term is the largest one. As a consequence, when eq. (2) holds, the surface morphology will be determined by a competition among the two roughening terms $v - S_{\bar{1}\bar{1}0}$, $v - S_{001}$ and the smoothing term $|A(E, \theta)|$. Since the S coefficients, contrary to v and A , are temperature dependent, T_s determines which term dominates among the three. At the lowest temperature ($T_s = 180$ K) $D_{\bar{1}\bar{1}0}/D_{001} \cong 4 \cdot 10^3$, implying that adatom diffusion is activated only along $\langle \bar{1} \bar{1} 0 \rangle$, and as a consequence D_{001} and S_{001} have a negligible effect.

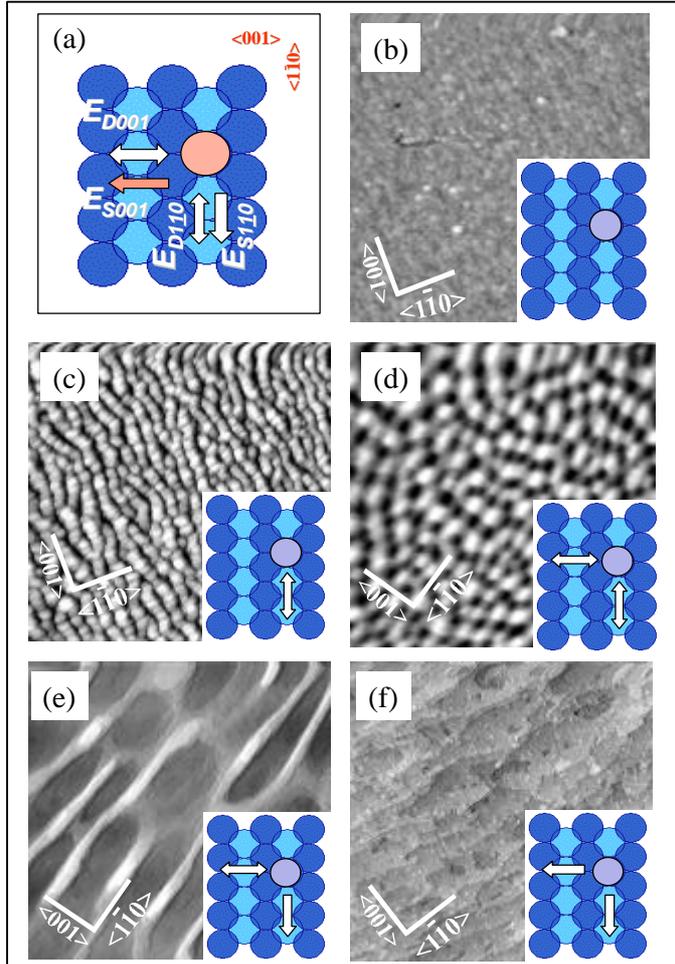


Fig.3 Five images (size $400 \times 400 \text{ nm}^2$ for **b** and **c**, while for **d**, **e** and **f** size is $800 \times 800 \text{ nm}^2$) of Cu(110) after ion sputtering at normal incidence ($\theta = 0^\circ$) for different temperatures T_s : 180 K (**b**), 250 K (**c**), 320 K (**d**), 360 K (**e**) and 400 K (**f**). $\Phi = 16 \mu\text{A}/\text{cm}^2$, $t = 15 \text{ min}$ and $\delta = 0^\circ$. The insets show the diffusion processes that are responsible of the observed surface morphology at each substrate temperature (the double white arrow indicates the adatom reflection at the step edge due to the Schwoebel barrier in its direction, while the single filled arrow indicates the direction along which the adatom can freely diffuses). Fig. 3a shows the energy barriers corresponding to the activated processes.

at $T_s = 350 \text{ K}$, only inter-layer motion in the $\langle 001 \rangle$ direction is inhibited ($R_{001} \cong 5 \cdot 10^{-2} R_{110}$) and the surface shows a ripple structure with k along the y direction (fig. 3e). Finally if $T_s \geq 380 \text{ K}$, adatoms can overcome also the second step edge barrier and the smoothing effect due to the 4th-order terms in (2) prevails (fig. 3f).

The linear model presented has a general validity for (110) surfaces in the presence of a Schwoebel barrier for adatoms (or vacancies) and is able to explain all the morphologies observed after ion sputtering. In case of (111) or (100) surfaces, as those described in ref. [8-12], equation (1) has to be

Thus the previous three terms become $v - S_{110}$, v and $|A(E, \theta)|$. Experimental data for sputtering at this temperature show a non-periodic surface (fig. 3b) characterized by a low value (≈ 4 atomic layers) [31] of the roughness W . Both these results indicate a predominance of the smoothing term. For $T_s = 250 \text{ K}$ the diffusion rate along $\langle 1 \bar{1} 0 \rangle$ increases ($-D_{110} \partial^4 h / \partial x^4$ term) and thus, respect to the $T_s = 180 \text{ K}$ case, the number of adatoms that try to diffuse to a lower layer is multiplied by a factor $D_{110}(T_s = 250 \text{ K}) / D_{110}(T_s = 180 \text{ K}) \cong 200$.

However, when $R_{110} \cong 10^{-4}$, the adatom thermal energy is not large enough to allow inter-layer diffusion. Adatoms that reach a step edge rebound back on the terrace producing an uphill current some order of magnitude bigger than the one that occurs at $T_s = 180 \text{ K}$ ($-S_{110} \partial^2 h / \partial x^2$). In the continuum equation this effect is represented by the predominance of $v - S_{110}$ on the temperature independent terms A and v , generating a surface instability with k parallel to the x direction (fig. 3c). At higher temperatures ($300 \leq T_s \leq 320 \text{ K}$), $D_{110} / D_{001} \cong 200$; as a consequence adatoms begin to diffuse also along $\langle 001 \rangle$ and, being $E_{S110} + E_{D110} \cong E_{D001}$ [25], overcome the Schwoebel barrier for the inter-layer motion along $\langle 1 \bar{1} 0 \rangle$. Consequently the instability due to $-S_{001} \partial^2 h / \partial y^2$ starts to take on effectiveness to detriment of that due to $-S_{110} \partial^2 h / \partial x^2$. Thus both x and y instabilities are presents and the surface is characterized by rectangular mounds randomly arranged (fig. 3d). For sputtering

modified in order to account for the different symmetry directions and energy diffusion barriers [28]. We also note that equation (2), being in a linear form, describes only the ripple formation with a specific direction and wavelength but does not predict any coarsening effect. The time dependence of λ shown in fig. 2b can be described only in a non-linear model [15,16,23]. In particular the second order derivative terms, which in eq. (1) express the action of a Schwoebel barrier on diffusion, have to be substitute by the more general terms

$$-S_{110} \frac{\partial}{\partial x} \left(f(\partial h/\partial x) \frac{\partial h}{\partial x} \right) - S_{001} \frac{\partial}{\partial y} \left(f(\partial h/\partial y) \frac{\partial h}{\partial y} \right) \quad (3)$$

where the different expressions for the f function can be found in refs. [15,16,23].

In conclusion we have shown that on a single crystal metal the surface morphology produced by ion sputtering depends both on ion erosion and surface diffusion processes. We have also proposed a linear equation able to explain the experimental results. This equation is a modified version of the one proposed by Cuerno and Barabasi for amorphous materials [7] in which we have included a diffusive term that accounts both for the presence of a Schwoebel barrier at the step edges and for the anisotropic surface diffusion.

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