

Ripple Structure on Ag(110) Surface Induced by Ion Sputtering

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

Centro di Fisica delle Superfici e Basse Temperature del CNR and INFN, Dipartimento di Fisica dell'Universita', Genova, Italy
(Received 4 December 1996)

The morphology of the Ag(110) surface after Ar⁺ ion sputtering has been studied by a variable temperature scanning tunneling microscope. For ion energies greater than 800 eV and in the temperature range $270 < T_S < 320$ K (where T_S is the sputtering temperature), a peculiar ripple structure well aligned along the $\langle 1\bar{1}0 \rangle$ direction and independent of the ion beam angle has been observed. A simple model of this effect, based on the anisotropy and on the temperature hierarchy of the Ag adatom and vacancy surface diffusion, explains the observed features. [S0031-9007(97)02931-1]

PACS numbers: 68.55.Jk, 68.35.Bs, 79.20.Rf

The evolution of surface morphology during ion sputtering is a complex phenomenon which includes, generally speaking, roughening and smoothing processes, and the actual morphology is determined by a balance between them. In many cases, it has been found that the roughness of a sputtered surface evolves following scaling laws [1–3] as observed in graphite samples [4] and that also a transition between dynamic scaling regimes can occur, as recently observed in Ge [5]. The driving mechanism for all these phenomena is the dependence of the sputtering yield on the local surface curvature [4,6,7]. A continuum equation for the height of the interface well describes the time evolution of the system [4] and, in particular conditions, some spatial components can be amplified so that a ripple structure can appear, with well-defined orientation and amplitude [8]. This structure has been observed in SiO₂ films [6] and in Ge(001) [7]. In both cases, it is perpendicular to the ion beam direction, with a periodicity of 260 and 2000 Å, respectively. It appears after sputtering at off normal incidence ($\vartheta = 55^\circ$), and it is strongly dependent on the ion fluence and the crystal temperature. In a recent Letter, Cuerno *et al.* [9] introduced a stochastic model to explain the formation of this structure induced by ion sputtering. They show that the time evolution of the surface roughness crosses different scaling regimes and in a precise time interval, *determined by a balance diffusion and erosion rates*, the height of the interface is almost periodic. The above model is of a general character, where the appearance of a ripple structure is independent of the nature of the sample. Although many experiments of ion sputtering have been carried out on metals, none of them have reported the appearance of a periodic structure. Recently, Krim *et al.* [10] measured by scanning tunneling microscopy (STM) the surface roughness after ion sputtering in Fe(100) films. They found that the surface roughness scales with the scan size indicating a self-affine fractal surface, however, they did not observe the periodic structure because they did not measure the height of the interface under the appropriate experimental conditions. As stated above in fact, the time interval where it is possible to observe such an effect is

strongly dependent on the experimental conditions as well as on the physical properties of the surface, and it can be particularly short in metals as the diffusion mobility of vacancies and adatoms is high. In light of the above considerations, we decided to investigate the morphology of a metal surface after ion sputtering, in order to verify whether a ripple structure can be induced also in metal surfaces. Such a study is conveniently performed in a variable temperature scheme: As the surface diffusion is strongly temperature dependent, for a fixed ion flux a particular temperature must exist at which the diffusion and erosion rates have the correct relation, and in these conditions a periodic structure should appear on the surface. The experiment was carried out by using a variable temperature STM [11], located in an UHV chamber (base pressure $\approx 10^{-10}$ mbar) with standard facilities. The sample temperature T_S can be varied in the 60–500 K temperature range. To perform this study, we used an Ag(110) crystal where the surface diffusion is highly anisotropic, being faster along the $\langle 1\bar{1}0 \rangle$ direction [12]. In this case the formation of a ripple structure should be favored, as the mass transport is enhanced in a well-defined direction. In addition, the adatom and vacancy mobility is high enough to allow us to search for the effect exploring a temperature range around room temperature. The Ag crystal is aligned to better than 0.2° along the (110) direction. In Fig. 1, the Ag(110) surface after the standard cleaning procedure (several cycles of sputtering and annealing, as reported in [13]) is shown, and in the inset the atomic structure is resolved. The experiment consists of sputtering the surface by Ar⁺ ions, with an energy E_i between 500 eV and 1.5 keV, while T_S is in the range 230–400 K. After the sputtering process, the sample is cooled in few minutes down to $T_0 = 200$ K, in order to inhibit the interlayer mass transport. The surface morphology is then investigated by STM as a function of the angle of incidence of the ion beam ϑ , as well as of the ion flux Φ and of the ion fluence Ψ ($\Psi = \Phi t$, where t is the sputtering time). All images presented in this Letter have been acquired at constant current in

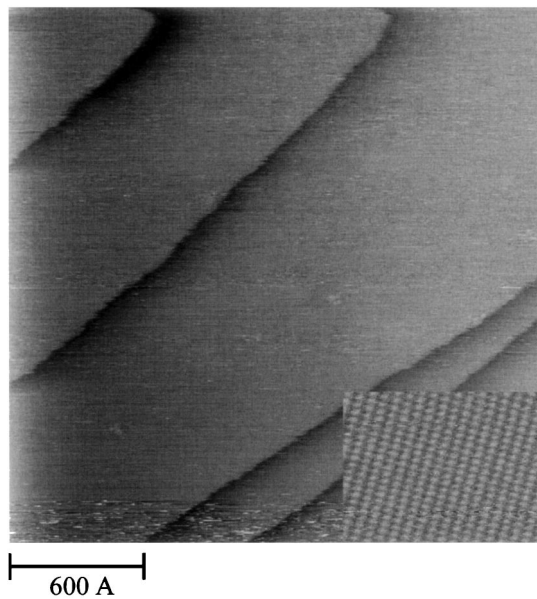


FIG. 1. The Ag(110) after several sputtering and annealing cycles, showing that large terraces are developed on our sample (image size $2400 \times 2400 \text{ \AA}^2$). In the inset the atomic structure is resolved.

differential mode, so that it is not possible to extract an absolute calibration of the surface roughness; however, in many cases it is possible to count directly the exposed layers. The tip-sample voltage is in the 1–1.5 V range, while the tunneling current is about 1 nA. In these conditions, the effect of the scanning on the sample morphology is negligible, as directly observed by comparing subsequent images. In Fig. 2, a series of six images obtained at different T_S is presented. For all images $\vartheta = 0^\circ$ (i.e., normal incidence), $E_i = 1 \text{ keV}$, $t = 15 \text{ min}$, $\Phi = 3 \cdot 10^{13} \text{ ions cm}^{-2} \text{ sec}^{-1}$, and $\Psi = 2.7 \cdot 10^{16} \text{ ions cm}^{-2}$. The ion flux and fluence can be expressed in terms of the Ag(110) surface density, resulting in $\Phi = 0.035 \text{ ML sec}^{-1}$ and $\Psi = 32 \text{ ML}$ (1 ML = $8.45 \cdot 10^{14} \text{ atoms cm}^{-2}$), respectively. It is evident that a ripple structure is induced on the Ag(110) surface, if $T_S = 300 \text{ K}$ [cf. Fig. 2(d)]. At lower temperatures, the surface morphology does not present any long range order structure, as in Figs. 2(a) and 2(b), while for $T_S = 320 \text{ K}$ the periodicity starts to vanish and disappears completely at higher temperatures [cf. Figs. 2(e) and 2(f)]. The structure reported in Fig. 2(c) is a particular case, which will be described in detail later.

The ripple morphology has been reported several times, and more than 200 images have been acquired. From these images, we can derive some general features of this phenomenon:

(1) When the structure is developed as in Fig. 2(d), the “channels” are aligned along the $\langle 1\bar{1}0 \rangle$ direction, and more than 20 layers are exposed. The spatial periodicity of the structure is $630 \pm 130 \text{ \AA}$ [14]. The orientation and the periodicity are independent on the ion beam incidence

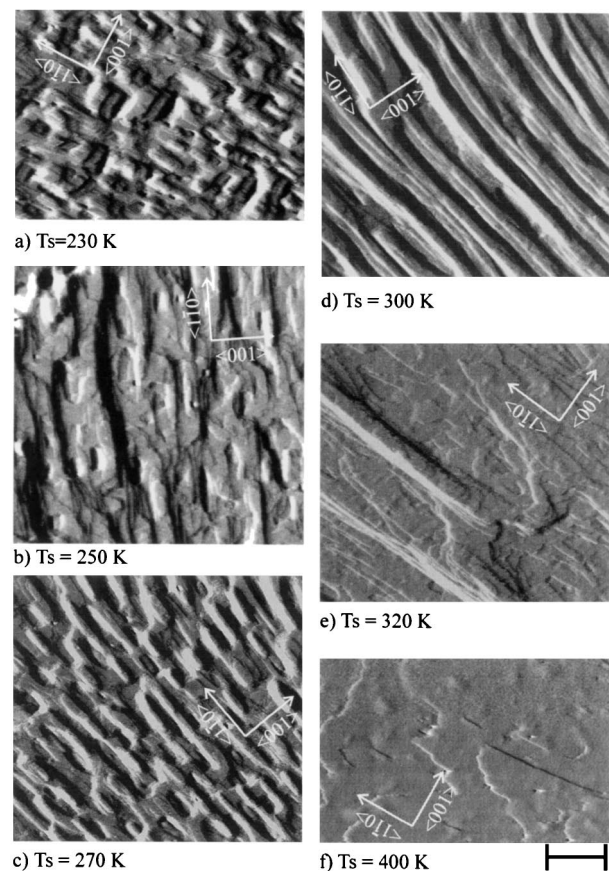


FIG. 2. Six images of the Ag(110) after sputtering at different temperatures T_S : 230, 250, 270, 300, 320, and 400 K, respectively. For all images the sputtering conditions are $E_i = 1 \text{ keV}$, $\Phi = 0.035 \text{ ML sec}^{-1}$, $\Psi = 32 \text{ ML}$. The bar corresponds to a length of 960 \AA for the images (a)–(e) and to a length of 480 \AA for the image (f).

angle, as well as from the azimuthal angle which have been varied during the experiment from 0° to 30° and from 0° to 60° , respectively.

(2) The ripple structure sets in for $E_i > 800 \text{ eV}$ at 300 K, and it is present everywhere on the crystal with a coherence of several thousand Angstrom [see Fig. 2(d)]. Above this energy, the resulting structure does not change significantly. This is related to energy dependence of the sputtering yield Y_S which remains about constant for energies in the range 1–10 keV [15]. Following the method described in [16], we measured $Y_S = 2.5 \pm 0.3$ at an ion energy of 1 keV, in agreement with the value 2.9 reported in the literature [15].

(3) It is unstable at room temperature. In Fig. 3 we show the same surface region after ion sputtering at $T_S = 300 \text{ K}$, 3(b) being acquired 140 min after 3(a). In the case, the sample was not cooled down to 200 K. A substantial smoothing of the surface is evident: Few channels are present on the same area, and they are not as deep. The exposed layers are about 21 in (a) and 17 in (b).

(4) The structure depends on the ion flux and not on the fluence, at least at 300 K. In Fig. 4 we report

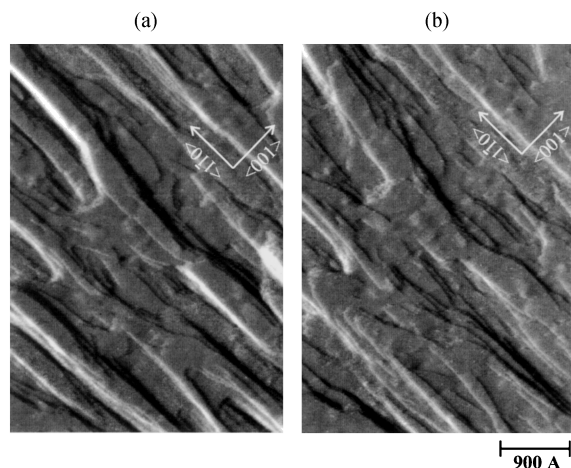


FIG. 3. Two successive images of the same sample zone (b) being acquired about 140 min after (a). The sputtering conditions are $T_S = 300$ K, $E_i = 1$ keV, $\Phi = 0.035$ ML sec^{-1} , and $\Psi = 32$ ML.

two images of the Ag(110) crystal after ion sputtering in the following conditions: $T_S = 300$ K, $E_i = 1$ keV, $\vartheta = 0^\circ$, $\Psi = 10.5$ ML, but at different fluxes, $\Phi = 0.035$ ML sec^{-1} for 4(a) and $\Phi = 0.01$ ML sec^{-1} for 4(b). While in 4(a) the ripple structure is well developed with the channels elongated along the compact direction, in 4(b) the surface does not present any evidence of periodicity, and fewer layers are exposed.

A complete analysis of time and temperature evolution of the ripple structure, as well as the ion energy, ion flux, and fluence dependence, will be presented elsewhere [17].

In order to develop a qualitative model of this phenomenon, we believe that two observations are fundamental: The ripple structure is well oriented along the $\langle 1\bar{1}0 \rangle$ direction, and it appears only in a restricted temperature range. Previous studies on the surface morphology of Cu(001) and Pt(111) after ion sputtering [18–20] clearly show that the diffusion processes of adatoms and vacancies play the major role in determining the surface structure. However, in these isotropic faces, no ripple structures have

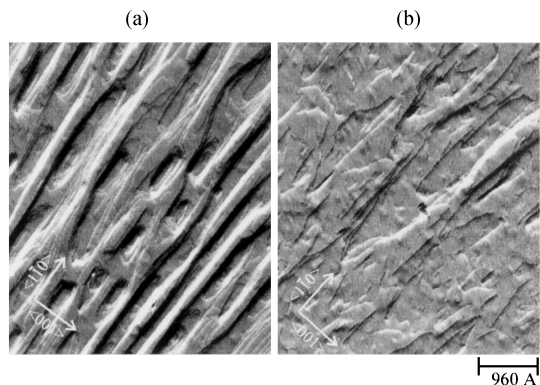


FIG. 4. Two images acquired at $T_0 = 200$ K after sputtering at equal fluence (10.5 ML) but at different ion fluxes, 0.035 ML sec^{-1} (a) and 0.01 ML sec^{-1} (b), respectively. $T_S = 300$ K and $E_i = 1$ keV.

been found. On the contrary, it is well known that for (110) metal surfaces both intralayer and interlayer diffusion processes are strongly anisotropic, enhancing the mass transport along the $\langle 1\bar{1}0 \rangle$ direction. From these considerations, we infer that in our case the *anisotropy* and the *temperature hierarchy* of the different surface diffusion processes are responsible for the formation of the ripple structure. The energy barriers for the adatom diffusion on the Ag(110) surface have been recently calculated by quenched molecular dynamics [12]. The intralayer diffusion along the $\langle 1\bar{1}0 \rangle$ direction has an energy barrier $\Sigma_1 = 0.28$ eV (in-channel jump), while for the corresponding process along the $\langle 001 \rangle$ direction $\Sigma_2 = 0.38$ eV (cross-channel exchange); among the interlayer processes, the jump down a $\langle 1\bar{1}0 \rangle$ step (jump descent) has an energy barrier of $\Sigma_3 = 0.35$ eV, significantly less than the corresponding process at a $\langle 001 \rangle$ step (exchange descent) $\Sigma_4 = 0.56$ eV. Despite the absolute values, the calculation reported in [12] establishes a precise hierarchy for the predominant diffusion process at a fixed temperature.

By means of these results, we can explain our observation as follows: (a) At low temperatures ($T_S < 250$ K), the dominant process is the in-channel jump: The interlayer mass transport is inhibited or less effective and the surface morphology does not present any periodicity or structure. Only the shape of holes and islands recalls the surface symmetry. (b) When $250 < T_S < 320$ K, the interlayer diffusion along the $\langle 1\bar{1}0 \rangle$ direction becomes more and more important: The two mechanisms (in-channel jump and jump descent) act together to create the deep holes [cf. Fig. 2(c)] and the “hills and valleys” structures [cf. Fig. 2(d)] as the mass redistribution is always along the $\langle 1\bar{1}0 \rangle$ direction. (c) Finally, at $T_S > 320$ K the interlayer mass transport along the $\langle 001 \rangle$ direction (i.e., perpendicular to the channels) is also activated, and the surface recovers a smooth morphology.

From our experimental results, it is not possible to deduce the real values of the energy barriers involved. However, the ratio between the energy barriers for the two competing interlayer processes can be estimated in a rough approximation following the method reported in [21]. Assuming that the two processes become important at the temperature $T_{S3} = 250$ K and $T_{S4} = 320$ K, respectively, a rough evaluation of the ratio can be inferred from the relation $\Sigma_4/\Sigma_3 \approx T_{S4}/T_{S3}$. From our results $\Sigma_4/\Sigma_3 \approx 1.3$, lower than but of the same order as 1.6 calculated by molecular dynamics. The value 1.3 corresponds to the rough approximation that the prefactors for the two processes are equal. Overcoming this approximation and by using realistic values for the prefactors [22], the ratio is always in the range 1.3–1.6.

Two other observations support the proposed model: the instability of the ripple structure at room temperature and its dependence on the ion flux. While at low temperature the ripple structure is stable (see Fig. 2), at room temperature the surface is smoothed significantly on a time scale of an hour (see Fig. 3): According to

our picture this effect can occur because mass transport along the $\langle 001 \rangle$ direction becomes high enough. On the other hand, the different morphology which appears at low ion fluxes (see Fig. 4) can be understood considering that the surface has sufficient time, between an ion hit and the following, to rearrange itself via the diffusion processes.

In spite of the goodness of these qualitative and intuitive arguments to explain our results, the transition between the “holes” [cf. Fig. 2(c)] to the hills and valleys [cf. Fig. 2(d)] regime must involve a more complicated mechanism. A crucial role could be played by the cluster of vacancies created by the sputtering in deeper layers (not in the first). These clusters have been recently observed by Schmid *et al.* [23], and the fingerprint of their presence is small bumps visible on the surface. As we observe similar features on Ag(110) after the sputtering procedure [cf. Fig. 2(f)], we propose that these clusters are present also in our surface after sputtering and that they can move, at sufficient temperature, and coalesce to produce the hills and valleys morphology.

In conclusion, we report on a new effect induced on Ag(110) surface by ion sputtering. In a restricted temperature range $270 < T_S < 320$ K, the sputtering produces a ripple structure, having a periodicity of about 600 Å. Its orientation and periodicity are independent of the geometrical parameters of the experiment. In a simple model, this ripple structure is the result of the balance between the erosion rate and the anisotropic surface diffusion. We want to emphasise that the ripple structure appears on the Ag(110) surface after ion sputtering *at normal incidence*, while in other cases [6,7] it has been observed only at larger incidence angles. At normal incidence the dependence of the sputtering yield on the surface curvature and the shadowing effect are negligible, so they cannot trigger the appearance of the surface instability. The same effect should appear on all metal surfaces (at the correct temperature), in which the adatom diffusion is strongly anisotropic. Our experiment not only confirms the generality of the model proposed in [9], but also demonstrates a new possibility to modify a metal surface on a nanometer scale.

We are indebted to R. Ferrando, H. Brune, and P. Politi for many elucidating discussions. D. Pagnotta and E. Giudice must be thanked for their help in the STM measurements.

*Corresponding author; electronic address: boragno@ge.infn.it

[1] R. Cuerno and A.L. Barabasi, Phys. Rev. Lett. **74**, 4746 (1995).

- [2] A.L. Barabasi and H.E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge England, 1995).
- [3] T. Halpin-Healey and Y.C. Zhang, Phys. Rep. **254**, 215 (1995).
- [4] E.A. Eklund, E.J. Snyder, and R. Stanley Williams, Surf. Sci. **285**, 157 (1993).
- [5] D.M. Smilgies, P.J. Eng, E. Landemark, and M. Nielsen (to be published).
- [6] T.M. Mayer, E. Chason, and A.J. Howard, J. Appl. Phys. **76**, 1633 (1994).
- [7] E. Chason, T.M. Mayer, B.K. Kellerman, D.T. McIlroy, and A.J. Howard, Phys. Rev. Lett. **72**, 3040 (1994).
- [8] We note that also in a recent simulation on film growth by MBE, a ripple structure of the interface has been found; see P. Politi and J. Villain, Phys. Rev. B **54**, 5114 (1996).
- [9] R. Cuerno, H.A. Makse, S. Tomassone, S.T. Harrington, and H.E. Stanley, Phys. Rev. Lett. **75**, 4464 (1995).
- [10] J. Krim, I. Heyvaert, C. Van Haesendonck, and Y. Bruynseraede, Phys. Rev. Lett. **70**, 57 (1993).
- [11] R. Conti, S. Rusponi, D. Pagnotta, and U. Valbusa (to be published).
- [12] F. Hontinfinde, R. Ferrando, and A.C. Levi, Surf. Sci. **366**, 306 (1996).
- [13] L. Vattuone, M. Rocca, C. Boragno, and U. Valbusa, J. Chem. Phys. **101**, 713 (1994).
- [14] About the observed periodicity, we note that a miscut of 0.14° from the (110) direction corresponds to an average terrace width of the order of 600 Å. Despite terraces larger than 600 Å that have been routinely observed in a well prepared surface, we cannot exclude *a priori* that the observed period of 600 Å is in some way related to the miscut of our sample.
- [15] H.E. Roosendal, in *Sputtering by Particle Bombardment*, edited by R. Behrisch (Springer Verlag, Heidelberg, 1981).
- [16] T. Michely and G. Comsa, Nucl. Instrum. Methods Phys. Res., Sect. B **82**, 207 (1993).
- [17] S. Rusponi, C. Boragno, and U. Valbusa (to be published).
- [18] T. Michely and G. Comsa, Surf. Sci. **256**, 217 (1991).
- [19] C. Teichert, M. Hohage, T. Michely, and G. Comsa, Phys. Rev. Lett. **72**, 1682 (1994).
- [20] J.C. Girard, Y. Samson, S. Gauthier, S. Rousset, and J. Klein, Surf. Sci. **302**, 73 (1994).
- [21] R. Stumpf and M. Scheffler, Phys. Rev. Lett. **72**, 254 (1994).
- [22] C.L. Liu, J.M. Cohen, J.B. Adams, and A.F. Voter, Surf. Sci. **253**, 334 (1991); see also R. Ferrando and G. Treglia, Phys. Rev. Lett. **76**, 2109 (1996); Ref. [21].
- [23] M. Schmid, W. Hebenstreit, P. Varga, and S. Crampin, Phys. Rev. Lett. **76**, 2298 (1996).