Phase boundaries of nanodots and nanoripples over a range of collision cascades

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The nonlinear continuum model proposed by Cuerno and Barabasi is the most successful and widely acceptable theoretical description of oblique incidence ion sputtered surfaces to date and is quite robust in its predictions of the time evolution and scaling of interfaces driven by ion bombardment. However, this theory has thus far predicted only ripple topographies and rough surfaces for short and large scales, respectively. As a result, its application to the interpretation and study of nanodots, predicted by Monte Carlo simulations for, and observed in experiments of, oblique incidence sputtering is still unclear and, hence, an open problem. In this paper, we provide a new insight to the theory, within the same length scale, that explains nanodot formation on off-normal incidence sputtered surfaces, among others, and propose ways of observing the predicted topographies of the MC simulations, as well as possible control of the size of the nanodots, in the framework of the continuum theory.

For some time now, the scientific community has been captivated by possible cooperative behaviour exhibited by surfaces sputtered by energetic ions, in which patterns are not only formed in the process of random ejection of surface particles, but that the orientation of the patterns can be tuned by varying the experimental sputtering parameters [4]. Coupled with their nature as nanostructures, these nanopatterns are of high technological importance due to obvious opto-electronic and other nanodevice applications. Among the earliest theories put forward to explain the time evolution of ion eroded surfaces was the linear continuum theory of Bradley and Harper [1], which describes the surface topography at any time instant as the result of an interplay or competition between the destabilizing erosion process, that creates an instability in which troughs are eroded in preference to crests, and the stabilizing processes of surface diffusion. Although, this explains the short length scale properties of sputtered surfaces observed in the period, it does not explain the larger length scale roughening behaviour.

According to Cuerno and Barabasi, the latter case is the result of nonlinear effects and the sputter noise in the sputtering process [5]. While this defines new scaling regimes absent from the linear theory, as well as ripple topographic regimes agreeing with the linear theory, a number of unresolved problems persist. For instance, the scaling properties of the different scaling regimes are unknown. Furthermore, the nonlinear theory, like the linear theory, has thus far predicted mainly ripple topography at short length scales. Also, calculations are usually performed for isotropic distribution of the energy of the impinging ion, mainly for ease of exposition of the theory, and a few cases of anisotropic energy distribution. These are inconclusive for certain important cases as we shall see below.

Meanwhile, a number of experiments of normal incidence sputtering have demonstrated the existence of a different kind of surface morphology, dot topographies, on semiconductor and amorphous surfaces at nanometre length scales with the presence of a characteristic length scale in the system [6–9]. Moreover, recent Monte Carlo simulations [10, 11] reported the existence of these dot topographies for off-normal incidence sputtering of amorphous substrates at collision cascade parameters different from the existing continuum theory calculations.

In Ref. [10], six different topographic regions were reported as to be expected for early times in the sputtering process. When considering the topographies at later times as well as the nature of the nanostructures, this six regions reduce to three, through a merger of the first four (I, II, III, IV).

Although, a number of targeted theoretical descriptions focussed at accounting for nanodot characteristics have been proposed, [12–14] the actual formation of these nanodots from oblique incidence sputtering remain unclear and is not yet understood, since the continuum theory has so far not predicted anything other than ripple topographies for off-normal incidence. This is still an open problem, which we investigate in this paper by providing phase diagram calculations of the continuum theory yet unreported for anisotropic distribution of the energy of the impinging ion. In particular, we provide those necessary to resolve the unanswered questions about the prediction of the continuum theory as regards non-ripple morphologies.

We propose a different interpretation of the continuum theory which, among other explanations, indicates that for anisotropic distribution of the energy of the impinging ions, the presence of a characteristic length scale may not predict the separation of ripple crests or troughs but that of dots. We discuss the accessibility of any of the regions studied to experimental probing; propose an explanation for the formation of the dots that arise from oblique incidence ion sputtering on the basis of the continuum theory, and a possible way of achieving or controlling required dot size.
The continuum theoretical description of interface morphology in terms of deterministic and stochastic partial differential equations is a powerful and successful tool for understanding the behaviour of diverse interface phenomena. For the specific case of ion sputtered surfaces, the distribution \( E(x) \) of the energy \( E \) of the incident ion to a surface particle located at position \( x = (x_1, x_2, x_3) \) is assumed in the continuum theory to be of the Gaussian form [13]:

\[
E(x) = \frac{E}{\sqrt{2\pi} \alpha \rho^2} \exp \left( -\frac{x_1^2}{2\alpha^2} - \frac{x_2^2}{2\rho^2} \right),
\]

where \( \alpha \) and \( \rho \) are the widths of the distribution parallel and perpendicular to the ion beam direction, respectively. The erosion velocity \( v \propto \partial_t h \), by definition, following which the dynamic evolution of the surface height, \( h(r, t) \), at nanometre length scales is, for most cases, governed by a Kuramoto-Sivashinsky type stochastic partial differential equation [5]

\[
\partial_t h(r, t) = -v_0 + \zeta \partial_x h(r, t) + \varsigma_x \partial_{xx} h(r, t) + \varsigma_y \partial_{yy} h(r, t) + \eta_x [\partial_x h(r, t)]^2 + \eta_y [\partial_y h(r, t)]^2 - \mathcal{D} \nabla^4 h(r, t) + \beta. \tag{2}
\]

\( v_0 \) is the erosion velocity of a flat surface, \( \zeta \) is a proportionality constant related to the local surface slope along the x-direction, \( \nu_x \) and \( \nu_y \) are the (linear) surface tension coefficients, \( \lambda_x \) and \( \lambda_y \) are the nonlinear coefficients, \( \mathcal{D} \) is the surface diffusion coefficient, and \( \beta \) is the normal noise term, which is assumed to have a Gaussian distribution with zero mean.

Using the convenient notation,

\[
a_\alpha = \frac{a}{\alpha}, a_\rho = \frac{a}{\rho}, \kappa = \cos \theta, \sigma = \sin \theta, \omega = a_\alpha^2 \sigma^2 + a_\rho^2 \kappa^2,
\]

\[
\Upsilon = \frac{FEPa}{\alpha \rho \sqrt{2\pi} \omega} \exp \left( -a_\alpha^2 a_\rho^2 \kappa^2 / 2\omega \right),
\]

where \( F \) is the ion flux and \( J \) is the proportionality constant between the power deposition and the rate of erosion, we provide the coefficients, for ease of reference, as follows. [5, 14]:

\[
c_\alpha = \Upsilon a_\alpha^2 \alpha^2 \left( 2a_\alpha^4 \sigma^4 - a_\alpha^2 a_\rho^2 \sigma^2 \kappa^2 + a_\alpha^2 a_\rho^2 \sigma^2 \kappa^2 - a_\rho^4 \kappa^4 \right),
\]

\[
\varsigma_x = \Upsilon \frac{\kappa}{2\omega^2} \left[ a_\alpha^2 a_\rho^2 \sigma^4 \left( 3 + 2\kappa^2 \right) + 4a_\alpha^4 a_\rho^4 \sigma^2 \kappa^2 - a_\alpha^4 a_\rho^4 \kappa^4 \left( 1 + 2\sigma^2 \right) \right] - \omega^2 [2a_\alpha^4 \sigma^2 - a_\alpha^2 a_\rho^2 \kappa^2 - \omega^2]
\]

\[
\varsigma_y = \Upsilon \frac{\kappa}{2\omega^2} \left( a_\alpha^4 \sigma^2 + a_\alpha^2 a_\rho^2 \kappa^2 - a_\alpha^2 a_\rho^2 \kappa^2 - \omega^2 \right)
\]

Non-linear effects are believed to be irrelevant at nanoscales, hence, at such length scales Eq. \(2\) predicts the presence of a characteristic length scale \( \Gamma = \sqrt{\mathcal{D} / |\varsigma|} \) in the system [4, 5] which manifests as periodic structures (e.g. in the separation of ripple crests/troughs); where \(|\varsigma|\) is the largest absolute value of the negative surface tension coefficients. Thus, if neither of \(|\varsigma_x|\) and \(|\varsigma_y|\) is less than zero, the characteristic length scale is absent. And, since the ripple wavelength is \( \lambda = 2\pi \sqrt{\mathcal{D} / |\varsigma|} \), no ripples are formed. In other words, the present continuum theory interpretation is that we either have ripples or not for oblique incidence, whereas reports of the possibility of other topographies have emerged.

In order to clarify this we obtain the phase diagrams of the continuum theory for the asymmetric energy distribution cases \( \alpha = 0 \rightarrow 5 \) and \( \rho = 0 \rightarrow 5 \), reported in Refs. [10] and [11]. The phase diagrams have been obtained from the variations of \( \varsigma_x, \varsigma_y, \eta_x, \eta_y \) as functions of \( \alpha \) and \( \rho \) (Figs. [1] and [2]); and as functions \( a \) and \( \theta \) (Figs. [3] and [4]). A calculation of the coefficients (as functions of \( \alpha \) and \( \rho \)) have revealed the same number (three) of topographic regions (Fig. [1]), as in the simulations at later times. Since these coefficients are also functions of \( a \) and \( \theta \) it is necessary to obtain the phase diagram in these regard as well. Indeed, as we show below, the phase diagram in terms of \( a \) and \( \theta \) allow for a wide range of possibilities of the coefficients and, hence, a large number of topographic regions. The regions to be encountered below in any of the two cases are as defined in Table [II]. As will be seen in what follows, if we ignore the nonlinear coefficients then there are only three possible regions, two of which describe ripples with either of the two possible orientations and the remaining one describing the situation in which neither of the surface tension coefficients is negative.

In the phase diagram presented in Fig. [1] obtained from the continuum theory, ripples are oriented along the y-axis in the three regions 2, 4, and 9. Whereas, in the simulation ripples are oriented perpendicular to the ion beam direction for the same sputtering parameters. Assuming that the projection of the ion beam direction
coefficients must be dominant and we explain their role
in the simulation corresponds, in the reference frame of
the continuum theory, to a straight line segment par-

to the x-axis, we have an agreement between the
continuum and the discrete theories that enables us to
interprete the results of the continuum theory as regards
dot topographies. For instance, since the disagreement
between the results of the continuum theory as regards
the continuum theory, to a straight line segment par-

in the simulation results. To investigate this we perform

calculations and on their basis propose an-
other interpretation of the result of an application of Eq.

is to further enhance the distinction between boundaries,
and not coded for any specific region.

Until now, phase diagram calculations have not been
done for this oblique incidence region and the only contin-

uum theory interpretation of the results of applications of
Eq. 2 to an understanding of the time evolution of sput-
tered surfaces is one that merges two relatively different
length scales, one short (of the order of 1 µm, the other
long (of the order of tens of µm). This interpretation
is, of course, very crucial to an understanding of transitions
that do occur, has been experimentally verified, and
are exhibited in the surface topography and the scaling
behaviour reported by diverse experiments at different
length scales. Here, we provide the results of these yet
unreported calculations and on their basis propose an-
other interpretation of the result of an application of Eq.

within the same length scale, which, we argue, accounts
for the unexplained phenomenon of oblique incidence dot
formation.

Note that the values of the collision cascade param-
eters at the phase boundaries are a bit different to those
in the simulation results. To investigate this we perform
calculations for the phase diagram at different θ [see Fig.

to observe shifts in these boundaries, which indicate the
possibility of a quantitative agreement with the val-
ues reported in the simulation. The general result for the
three regions of Fig. 1 when considering both a and θ, is
shown in Figs. 2 and 3(a) and (b), for α = 3.3, ρ = 4.5;
α = 1.6, ρ = 3.3; and α = 2.0, ρ = 1.0, respectively. A
transition from region 2 to 4 of Fig. 1 when considering
a fixed α and varying ρ, is due to the change of sign of
the nonlinear coefficient ς_x, associated to increasing local
surface slopes along the x-axis, which is a precursor to
the change from region 4 to 9, in which case there is no
directional change in nonlinearity but only a change of
sign of the surface tension coefficient ς_x. This is impor-

TABLE I. Definition of the regions found in the calculations,
as described in the text.

| Region | Relative Signs of the Coefficients |
|--------|-----------------------------------|
| 1      | ς_x < ς_y ≤ 0; ς_x < 0, ς_y < 0    |
| 2      | ς_y < ς_x ≤ 0; ς_x < 0, ς_y < 0    |
| 3      | ς_x < ς_y ≤ 0; ς_x > 0, ς_y < 0    |
| 4      | ς_y < ς_x ≤ 0; ς_x > 0, ς_y < 0    |
| 5      | ς_x < ς_y ≤ 0; ς_x > 0, ς_y > 0    |
| 6      | ς_y < ς_x ≤ 0; ς_x < 0, ς_y > 0    |
| 7      | ς_x < ς_y ≤ 0; ς_x > 0, ς_y > 0    |
| 8      | ς_y < ς_x ≤ 0; ς_x > 0, ς_y > 0    |
| 9      | ς_x > 0, ς_y < 0; ς_x > 0, ς_y > 0 |
| 10     | ς_x > 0, ς_y < 0; ς_x < 0, ς_y < 0 |
| 11     | ς_x > 0, ς_y < 0; ς_x > 0, ς_y > 0 |
| 12     | ς_x > 0, ς_y < 0; ς_x > 0, ς_y = 0 |

FIG. 1. Phase diagram for collision cascade parameters α and ρ ranging from 0 to 5. θ = 50°, α = 6.0. The three regions 2, 4, and 9, are as defined in the text.

FIG. 2. Boundary shifts in the phase diagram, arising from varying θ, for the same parameters as in Fig. 1 Left: θ = 70°; Right: θ = 30°.
FIG. 3. Phase diagram for the isotropic case $\alpha = 3.3$ and $\rho = 4.5$, representative of region 9 of Fig. 1, for varying $\theta$ and $a$; $\theta$ (horizontal axis) ranging from 0 to 90, and $a$ (vertical axis) ranging from 0 to 6. The regions found are as labelled in the figure, and defined in the text.

FIG. 4. Phase diagram for representative collision cascade parameters of the two remaining regions of Fig. 1 and for varying $\theta$ and $a$; $\theta$ (horizontal axis) ranging from 0 to 90, and $a$ (vertical axis) ranging from 0 to 6. (a) $\alpha = 1.6$ and $\rho = 3.3$; (b) $\alpha = 2.0$ and $\rho = 1.0$. The regions found are as labelled in the figure, and defined in the text.

A negative surface tension coefficient is representative of the instability arising from the sputtering process in which troughs are eroded in preference to crests. A positive surface tension coefficient would then imply a neglect of troughs in the erosion process. On the other hand, a negative nonlinear coefficient implies that the height evolution increases as local surface slopes increase, and vice-versa. This means that the interplay that leads to ripple formation is enhanced or countered depending on the relative signs of the nonlinear coefficients which is capable of creating a further instability that disturbs the interplay. Following this, pattern formation in the 12 regions highlighted above are as tabulated below in Table II. Note that due to the interplay between $\nu_x$ and $\nu_y$ periodic structures with either of two possible orientations are always present, except if one of $\nu_x$ or $\nu_y$ is zero. Details of how to calculate the quantities (e.g. ripple amplitude, growth rate, etc.) in Table II can be found in Refs. [3, 16].

In particular, for an explanation of region 9, trough erosion along the x-axis is not favoured and the height evolution along the x-axis decreases. Since, the erosion is a stochastic process, and erosion along the x-axis, for this region, is much reduced in comparison to that along the y-axis, the continuity of eroded troughs along the y-axis is broken and instead of long grooves we have pits interspersed with isolated protrusions which together make dot topography.

Based on this explanation, then dot size depends on the relative magnitudes of the nonlinear coefficients, which again is dependent on the sputtering parameters. Thus, for a preferred dot size and growth with time, one would need to strike the right balance between the appropriate choices of material (which influences $a$, $\alpha$, $\rho$, etc) and sputtering conditions such as ion incidence, temperature, etc, according to the phase diagram of Figs. 1, 2, 3, and 4. There are a few cases with $\zeta_x > 0$, $\zeta_y < 0$, $\eta_x < 0$, $\eta_y = 0$ in Figs. 2 and $\zeta_x > 0$, $\zeta_y > 0$, $\eta_x = \eta_y = 0$ in Fig. 4 and a few case of region 12 in Figure ???. These do not appear in the phase diagram because they are exceptional cases which are too tiny to be noticed.

On the accessibility of some of the regions studied here to probing by experiments, we recall the assumption of the continuum [4, 5] and discrete [10, 11, 17, 18] theories that the impinging ion penetrates a distance $a$ into the material, after which it distributes its energy according to a Gaussian distribution, which is a simple representation of the geometry defined by the collision cascades triggered by the impinging ion. Thus, it is possible to design the shape of this geometry through controlled defect creation or doping in the first few surface layers of
TABLE II. Definition of the regions found in the calculations, as described in the text.

| Region | Topography |
|--------|------------|
| 1      | ripples oriented along x with much less prominent underlying periodic structure along y |
| 2      | ripples oriented along y with much less prominent underlying periodic structure along x |
| 3      | ripples oriented along x with shorter amplitude and (possibly prominent, depending on the relative size of $\varsigma_y$) periodic structure along y |
| 4      | almost pure ripples (i.e. very little or no sign of an underlying periodic structure) oriented along y with normal amplitude growth |
| 5      | almost pure ripples oriented along x with normal amplitude growth |
| 6      | ripples oriented along y with shorter amplitude and (possibly prominent, depending on the relative size of $\varsigma_x$) periodic structure along x |
| 7      | possibly a rough surface, or low amplitude ripples oriented along x; depending on the relative strengths of the competing factors |
| 8      | ripples oriented along y with slow or no amplitude growth; or rough surface if the nonlinearities cancel out or soften the surface tension instability |
| 9      | dots with underlying periodic structure oriented along y |
| 10     | short ripples oriented along y, or dots of lower growth rate and underlying structure oriented along y; depending on the relative strengths of the competing factors |
| 11     | ripples oriented along x with slow or no amplitude growth; or rough surface if the nonlinearities cancel out or soften the surface tension instability |
| 12     | dots with less prominent or no underlying periodic structure |

width roughly about the penetration depth $a$.

In summary, we have proposed a new interpretation of the continuum theory that explains surface topographies yet unaccounted for and also agrees with the previous conceptual framework of the theory. This new insight considers the stochastic time evolution equation within the same lengthscale that is of general interest in experimental studies and characterization of the nanostructures. A possible means of achieving the sputtering parameters necessary for experimental observation of the topographic regions of present interest, e.g. the nanodot regions, is discussed.

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[1] E. A. Eklund, R. Bruinsma, J. Rudnick, and R. S. Williams, Phys. Rev. Lett. 67, 1759 (1991)
[2] E. Chason, T. M. Mayer, B. K. Kellerman, D. T. McLroy, and A. J. Howard, Phys. Rev. Lett. 72, 3040 (1994)
[3] E. O. Yewande, Modelling and simulation of surface morphology driven by ion bombardment, PhD dissertation, University of Göttingen, Faculty of Mathematics and Natural Sciences (2006), this is for further details.
[4] R. M. Bradley and J. M. E. Harper, J. Vac. Sci. Technol. A 6, 2390 (1988)
[5] R. Cuerno and A. L. Barabási, Phys. Rev. Lett. 74, 4746 (1995)
[6] S. Facsko, T. Dekorsy, C. Koerdt, C. Trappe, H. Kurz, A. Vogt, and H. L. Hartnagel, Science 285, 1551 (1999)
[7] F. Frost, A. Schindler, and F. Bigl, Phys. Rev. Lett. 85, 4116 (2000)
[8] R. Gago, L. Vázquez, R. Cuerno, M. Varela, C. Balles- teros, and J. M. Albella, Appl. Phys. Lett. 78, 3316 (2001)
[9] B. Ziberi, F. Frost, and B. Rauschenbach, Appl. Phys. Lett. 88, 173115 (2006)
[10] E. O. Yewande, A. K. Hartmann, and R. Kree, Phys. Rev. B 73, 115434 (2006)
[11] E. O. Yewande, R. Kree, and A. K. Hartmann, Phys. Rev. B 75, 155325 (2007)
[12] M. Castro, R. Cuerno, L. Vázquez, and R. Gago, Phys. Rev. Lett. 94, 016102 (2005)
[13] B. Kahng, H. Jeong, and A. L. Barabási, Appl. Phys. Lett. 78, 805 (2001)
[14] J. Muñoz-Garcia, M. Castro, and R. Cuerno, Phys. Rev. Lett. 96, 086101 (2006)
[15] P. Sigmund, Phys. Rev. 184, 383 (1969)
[16] M. Makeev, R. Cuerno, and A. L. Barabási, Nucl. Instrum. Methods Phys. Res. B 197, 185 (2002)
[17] A. K. Hartmann, R. Kree, U. Geyer, and M. Köbel, Phys. Rev. B 65, 193403 (2002)
[18] E. O. Yewande, A. K. Hartmann, and R. Kree, Phys. Rev. B 71, 195405 (2005)