Quantum dot and hole formation in sputter erosion

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Recently, it was experimentally demonstrated that sputtering under normal incidence leads to the formation of spatially ordered uniform nanoscale islands or holes. Here, we show that these nanostructures have inherently nonlinear origin, first appearing when the nonlinear terms start to dominate the surface dynamics. Depending on the sign of the nonlinear terms, determined by the shape of the collision cascade, the surface can develop regular islands or holes with identical dynamical features, and while the size of these nanostructures is independent of flux and temperature, it can be modified by tuning the ion energy. © *2001 American Institute of Physics.*
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The fabrication and physical properties of quantum dots (QDs) are topics of high current interest due to their potential applications in optoelectronics.[1] However, despite the high interest in the subject, the methods available for the fabrication of such dots are rather limited. Lithographic techniques[2] cannot produce small and dense enough dots necessary for device applications. While much research has focused on strained QD formation,[3] there is continued high demand for alternative methods that would allow low cost and efficient mass fabrication of QDs. In the light of these technological driving forces, the recent demonstration that sputter erosion can lead to uniform nanoscale islands, that exhibit quantum confinement, will undoubtly capture the interest of the scientific community.[4] Ion beam sputtering has long been a leading candidate for surface patterning. While ripple formation on sputter eroded surfaces has been observed already in the 1970s,[5] in the last decade much work has been devoted to understand both the experimental and theoretical aspects of this fascinating self-organized phenomena. However, most experiments have focused on off-normal incidence, that leads to ripple formation. While it was expected that under normal incidence the ripples should be replaced by some periodic cellular structures, such surface features have not been observed experimentally. Recently, two groups have obtained simultaneous advances in this direction. Facsko *et al.*, investigating low-energy normal incident Ar$^+$ sputtering of GaSb (100) surfaces,[4] observed that as erosion proceeds, nanoscale islands appear on the surface, that are remarkably well ordered, and have a uniform size distribution. On the other hand, recent experiments of Ar$^+$ sputtering of Cu (110), and Ne$^+$ sputtering of Ag (001) under normal incidence lead to relatively uniform depressions or holes.[6] These experiments raise a number of questions regarding the mechanisms responsible for the formation of these nanostructures. While it is tempting to interpret these structures as periodic perturbations expected by the linear theory of

sputtering,[7] a careful analysis of the experimental results indicates that such an approach is less than satisfactory. To mention only a few discrepancies, Facsko *et al.* reported that the obtained island size is independent of temperature, while, according to the linear theory their size should decrease exponentially with T.[7] Also, the linear theory would predict at best a cellular structure displaying a square lattice, in contrast with the hexagonal ordering observed in experiments.[4]

In this letter we present the detailed theory addressing the formation of sputter-induced QDs and holes. We find that regular nanostructures first appear as the nonlinear terms become relevant, allowing us to predict the characteristic time, τ, necessary for their formation, and calculate the dependence of τ on the physical parameters characterizing the ion bombardment process. Furthermore, we show that the QDs and holes, observed in different experiments, could be governed by the same physical phenomena. Finally, we predict that the size of these islands and holes, while independent of flux and temperature, depends on the ion energy.

A successful description of the morphological evolution of sputter eroded surfaces has been proposed by Bradley and Harper (BH),[7] based on Sigmund's theory of sputtering,[8] predicting that the height $h(x,y,t)$ of the eroded surface is described by the linear equation

$$\partial_t h = \nu_x \partial_x^2 h + \nu_y \partial_y^2 h - K \partial^4 h, \tag{1}$$

where ν_x and ν_y are effective surface tensions, generated by the erosion process, and K is the surface diffusion constant induced by thermal diffusion. The balance of the unstable erosion term $(-|\nu|\partial^2 h)$ and the surface diffusion term $(-K\partial^4 h)$ acting to smooth the surface, generates ripples with wavelength

$$\ell_i = 2\pi\sqrt{2K/|\nu_i|}, \tag{2}$$

where i refers to the direction (x or y) along which ν_i (ν_x or ν_y) is the largest. While predicting the ripple wavelength and orientation,[9] the linear theory fails to explain a number of experimental features, such as the saturation of the ripple
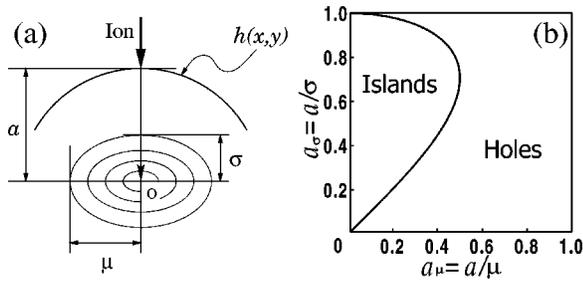
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FIG. 1. (a) The characteristics of the collision cascade generated by an ion. According to Sigmund's theory of sputtering (see Ref. 8), an ion penetrates the surface until a penetration depth a, where it radiates its energy out, such that the kinetic energy generates damage, decays exponentially with the distance from O. For most systems the energy distribution is anisotropic, being characterized by the characteristic decay length scales σ and μ along or perpendicular to the ion direction, respectively. (b) Phase diagram showing the parameter regimes corresponding to island (QD, $\lambda > 0$) and hole ($\lambda < 0$) formation.
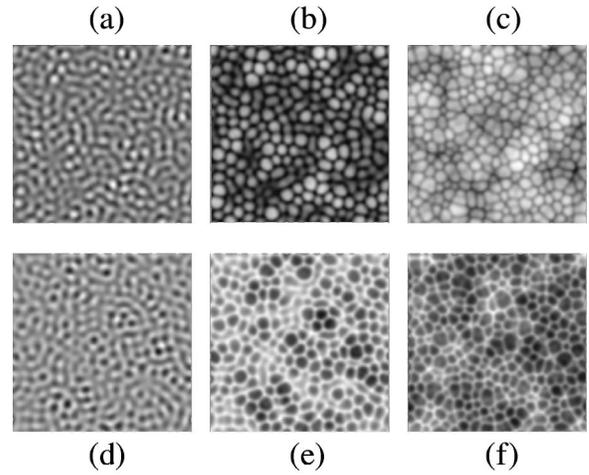

FIG. 2. (a)–(c) Surface morphologies predicted by Eq. (3), for $\lambda = 1$ at different stages of surface evolution. The pictures correspond to (a) $t = 4.0$, (b) 5.8, (c) 8.0×10^4. (d)–(f) The same as in (a)–(c), but for $\lambda = -1$. In all cases we used $\nu = 0.6169$, $D = 2$, and system size 256×256.

amplitude,[10–12] or the appearance of kinetic roughening.[13,14] It has been proposed[15] that these shortcomings can be cured by the inclusion of nonlinear terms and noise, derived from Sigmund's theory of sputtering. Consequently, Eq. (1) has to be replaced by the nonlinear equation

$$\partial_t h = \nu_x \partial_x^2 h + \nu_y \partial_y^2 h - D_x \partial_x^4 h - D_y \partial_y^4 h - D_{xy} \partial_x^2 \partial_y^2 h$$
$$+ \frac{\lambda_x}{2}(\partial_x h)^2 + \frac{\lambda_y}{2}(\partial_y h)^2 + \eta(x,y,t), \qquad (3)$$

where λ_x and λ_y describe the tilt-dependent erosion rate, depending on flux f and the penetration depth a, and $\eta(x,y,t)$ is an uncorrelated white noise with zero mean, mimicking the randomness resulting from the stochastic nature of ion arrival to the surface.[15,16] Furthermore, the sputtering process always generates ion induced effective diffusion that, together with thermal diffusion, is incorporated in D_x, D_y, and D_{xy}.[16] Under normal incidence, the coefficients in Eq. (3) are isotropic, given by Refs. 16 and 17

$$\nu \equiv \nu_x = \nu_y = -f a a_\sigma^2 / 2 a_\mu^2, \qquad (4)$$

$$D \equiv D_x = D_y = f a^3 a_\sigma^2 / 8 a_\mu^4, \qquad (5)$$

$$\lambda \equiv \lambda_x = \lambda_y = (f/2a_\mu^2)(a_\sigma^2 - a_\sigma^4 - a_\mu^2), \qquad (6)$$

where $a_\mu = a/\mu$ and $a_\sigma = a/\sigma$ and μ and σ, defined in Fig. 1(a), characterize the shape of the collision cascade of the bombarding ion.

To investigate the origin of the QDs and the dynamics of QD formation under normal incident ion sputtering, we integrated numerically the continuum Eq. (3), using isotropic coefficients as expected for normal incidence, the methods being described in Ref. 18. To improve the uniformity of the QDs, the numerical simulations were carried out without noise ($\eta = 0$), using instead a random initial surface configuration.

Our main result, presenting the morphology of the ion sputtered surface at three different stages of their time evolution, is shown in Fig. 2. Let us first concentrate on the $\lambda > 0$ case (upper panels in Fig. 2). In the early stages of the sputtering process the surface is dominated by small, wavy perturbations [Fig. 2(a)] generated by the interplay between the ion induced instability and surface relaxation. However,

since the system is isotropic in the (x,y) plane, these ripple precursors are oriented randomly, generating short wormlike morphologies on the surface. After some characteristic time, τ, these structures turn into isolated but closely packed islands, reminiscent of the QDs reported experimentally [Fig. 2(b)]. Note that upon a closer inspection one can observe the emergence of hexagonal order in the island positions. As the sputtering proceeds, the supporting surface develops a rough profile, destroying the overall uniformity of the islands [Fig. 2(c)]. A similar scenario is observed for $\lambda < 0$, the only difference being that now the islands are replaced by holes [Figs. 2(d)–2(f)]. Thus the development of QDs and holes is governed by the same underlying physical phenomena, the only difference being that for QDs $\lambda > 0$, and for holes $\lambda < 0$. Indeed, this morphological change is expected from the nonlinear continuum theory, Eq. (3) being symmetric under the simultaneous transformation $\lambda \rightarrow -\lambda$ and $h \rightarrow -h$, indicating that changing the sign of λ does not affect the dynamics of the surface evolution, but simply turns the islands into mirrored holes. Since, according to Eq. (6) the sign of λ is determined only by the relative magnitude of a_σ and a_μ, whether islands or holes appear is determined by the shape of the collision cascade, shown in Fig. 1(a). Consequently, using Eq. (6) we can draw a phase diagram in terms of the reduced penetration depths a_σ and a_μ that separates the regions displaying QDs versus holes [Fig. 1(b)]. These results also indicate that the QDs and holes are inherently nonlinear objects, since, should the linear terms be responsible for their formation, the surface morphology should not depend on the sign of λ [Eq. (1) has a full $h \rightarrow -h$ symmetry].

The crossover behavior from the linear to the nonlinear regimes can be monitored through the surface width, $W^2(L,t) \equiv 1/L^2 \sum_{x,y} h^2(x,y,t) - \bar{h}^2$. As Fig. 3 shows, this quantity exhibits a sharp transition at a characteristic time τ. for $t < \tau$, the width W increases exponentially as $W \sim \exp(\nu t/\ell^2)$, while for $t > \tau$, W still increases but at a considerably smaller rate than an exponential.[18] The crossover time is given by $\tau \sim \sigma^2/fa$ in terms of the experimental parameters,[18] indicating its dependence on the flux and ion
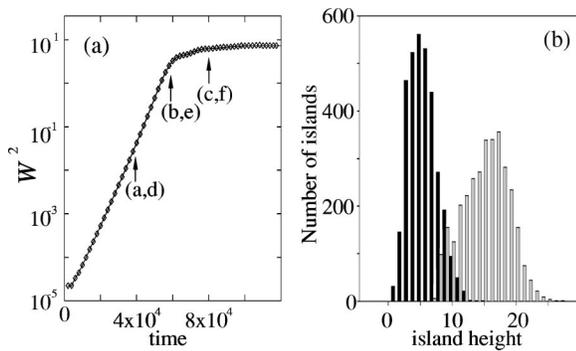
FIG. 3. (a) Time evolution of the surface width W^2 for the parameters $\nu = -0.6169$, $D=2$, $\lambda=1$ or -1, and system size 256×256. The crossover time is estimated to be $\tau\approx5.8\times10^4$. The arrows denote the moments when the surface snapshots shown in Fig. 2 were recorded. (b) Island height distribution right at the crossover time $t=\tau$ (black columns), and at a later time, $t=8.0\times10^4$ (gray columns), indicating that the width of the distribution is smaller at the crossover time, and broadens as we go beyond τ.

beam energy. Correlating these results with the observed surface morphologies, we find that the QDs first appear at $t \approx \tau$. Indeed, in Fig. 3 we marked with arrows the time when the morphologies in Figs. 2(a)–2(c) were recorded, indicating that no QDs exist before τ [Fig. 2(a)], but they are fully developed at τ [Fig. 2(b)], and their uniformity rapidly vanishes after τ [Fig. 2(c)]. This implies that it is desirable to stop the erosion process right at τ to obtain the most uniform QDs.

Based on the results presented in Figs. 2 and 3, the following scenario emerges for QD and hole formation. In the early stages of the erosion process the linear theory correctly describes the surface evolution, and thus we observe the cellular structure predicted by the BH theory. However, as time increases, the nonlinear terms *turn on* breaking the up-down symmetry of the surface.[19] The sign of λ (governed by the shape of the collision cascade) determines whether QDs or holes form, these structures appearing at a characteristic time τ, that depends on the flux and the ion energy, and their characteristic size is given by the ripple period ℓ [Eq. (2)]. As time increases beyond τ, the nonlinear terms lead to kinetic roughening of the surface at large length scales,[15,19] and while the QDs and holes do not disappear, the substrate on which they exist becomes rough, destroying the overall island/hole uniformity and ordering. Indeed, as Fig. 3(b) shows, the island height distribution has a larger width after τ than right at τ. Thus, ordered and uniform QDs and holes can be obtained only at the crossover between the linear and nonlinear regimes emerging at the characteristic time τ.

Since the mechanism generating the QDs is the ion induced instability, balanced by ion induced diffusion, their typical separation is given by the most unstable wavelength, $\ell=2\pi\sqrt{2D/\nu}$. The simulations indicate that the islands (or holes) first appear as small surface perturbations. However, as their amplitude increases, around τ they get in contact with each other, and their growth saturates. Thus their final diameter coincides with ℓ. Indeed, we determined the diameter of the QDs in simulations for $\nu=-0.6169$, $D=2$ and $\lambda=1$, observing approximately 16 QDs aligned along the side of the system of the size 256×256, in agreement with

the prediction $L/\ell\approx16$. Using Eqs. (4) and (5), we find that $\ell=\sqrt{2}\pi\mu$, i.e., from the *average separation* of the islands one can *determine the size of the horizontal width* of the collision cascade [Fig. 1(a)]. Furthermore, since typically we have $\mu\sim a\sim\epsilon^{2m}$, when ϵ is the ion energy and m is a constant that weakly depends on ϵ ($m=1/2$ for $\epsilon\approx10 \sim100$ keV),[8] we predict that one can tune the size of the QDs by changing the ion energy ϵ, while the size is independent of the flux and the temperature.

Finally, the conclusion $\ell\sim\mu$ holds only when ion induced effective diffusion[16] is the main relaxation mechanism. While the temperature independent QD size reported by Facsko *et al.*[4] indicates that for GaSb this is the main relaxation mechanism, at high temperatures or in other systems thermal diffusion could be more relevant.[16,17,20] Note, however, that the scenario presented above for QD and hole formation is not conditional on ion induced diffusion. Should thermal diffusion be the dominating relaxation mechanism it will change only our prediction for ℓ and τ. In this case we expect $\ell\sim2\pi\sqrt{2D_0\exp(-E/2kT)/\nu}$, thus the QD size will depend exponentially on T, and we also have $\ell\sim\epsilon^{-1/2}$, and $\ell\sim f^{-1/2}$. For the crossover time we obtain $\tau\sim D_0^2 \times\exp(-2E/kT)/\nu$, $\tau\sim\epsilon^{-1}$, and $\tau\sim f^{-1}$. However, the phase diagram [Fig. 1(b)] and the expected dynamical evolution (Figs. 2 and 3) will not be sensitive to the nature of the relaxation mechanism.

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