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.

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The fabrication and physical properties of quantum dots (QDs) are topics of high current interest due to their applications in optical devices and as potential construction blocks of novel computer architectures. However, despite the high interest in the subject, the methods available for the fabrication of such dots are rather limited. Lithographic techniques, while undergoing rapid improvements in resolution, still cannot produce small and dense enough dots necessary for device applications. While much research has focused on self-assembled QD formation, that generates dots through the unique combination of strain and growth kinetics, these techniques offered relatively uniform islands only for a few material combinations and still have to find their way into devices. Consequently, 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 undoubtedly capture the interest of the scientific community. 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 70s, 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, by breaking the symmetry along the surface, leads to anisotropic structures, such as ripples. While theoretically 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, 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. These experiments, while represent significant technological breakthroughs, 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, a careful analysis of the experimental results indicates that such 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 $T$. Also, the linear theory would predict at best a cellular structure displaying a square lattice, in contrast with the hexagonal ordering observed in experiments.

In this paper we present the first detailed theory addressing the formation of sputter-induced QDs and holes. We demonstrate that these structures are the result of inherently nonlinear phenomena, and thus they cannot be accounted for in the context of the linear theory. We find that regular nanostructures first appear as the nonlinear terms become relevant, allowing us to predict the characteristic time, $\tau$, necessary for their formation, and calculate the dependence of $\tau$ on the physical parameters characterizing the ion bombardment process. Furthermore, we show that the QDs and holes, observed in different experiments, are governed by the same physical phenomena, the difference between them coming from the shape of the ion cascade characterizing the interaction of the bombarding ions with the substrate. Finally, we predict that the size of these islands and holes, while independent of flux and temperature, depends on the ion energy.

A particularly successful description of the morphological evolution of sputter eroded surfaces has been proposed by Bradley and Harper (BH), based on Sigmund’s theory of sputtering, 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 \( \nu_x \) and \( \nu_y \) 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 \( \nu_i \) (\( \nu_x \) or \( \nu_y \)) is the largest. While successful in predicting the ripple wavelength and orientation \[6\], the linear theory fails to explain a number of experimental features, such as the saturation of the ripple amplitude \[16\] \[17\], or the appearance of kinetic roughening \[14\] \[15\]. To address these questions it has been proposed \[10\] that these shortcomings can be cured by the inclusion of nonlinear terms and noise, derived from the Sigmund’s theory of sputtering. Consequently, Eq. (1) has to be replaced by the nonlinear equation,
\[
\begin{align*}
\partial_t h &= \nu_x \partial_x^2 h + \nu_y \partial_y^2 h - D_x \partial_x^4 h - D_y \partial_y^4 h - D_{xy} \partial_x^2 \partial_y^2 h \\
&\quad + \frac{\lambda_x}{2} (\partial_x h)^2 + \frac{\lambda_y}{2} (\partial_y h)^2 + \eta(x, y, t), \tag{3}
\end{align*}
\]
where \( \lambda_x \) and \( \lambda_y \) 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 \[10\] \[14\]. Furthermore, the sputtering process always generates ion induced effective diffusion, that together with thermal diffusion, are incorporated in \( D_x, D_y \), and \( D_{xy} \) \[17\]. Under normal incidence, the coefficients in (3) are isotropic, given by \[17\] \[18\]
\[ \nu \equiv \nu_x = \nu_y = -fa \sigma^2 /2a^2 \mu, \tag{4} \]
\[ D \equiv D_x = D_y = fa^3 a^2 /8a^4 \mu, \tag{5} \]
\[ \lambda \equiv \lambda_x = \lambda_y = (f /2a^2 \mu)(a^2 - a^4 - a^2), \tag{6} \]
where \( a_{\mu} = a / \mu \) and \( a_{\sigma} = a / \sigma \) and \( \mu \) and \( \sigma \) defined in Fig. 1a, 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 equation (3), using standard discretization techniques \[14\], and isotropic coefficients as expected for normal incidence. We choose a temporal increment \( \Delta t = 0.01 \) and impose periodic boundary conditions \( h(x, y, t) = h(x + \ell, y, t) = h(\ell_t, y, t) = \ell_t + \ell, t) \) where \( L \times L \) is the size of the substrate. To improve the uniformity of the QDs, the numerical simulations were carried out without noise \( (\eta = 0) \), using instead a random initial surface configuration. However, we repeated the simulations for the noisy case as well, finding that the size uniformity of QDs is enhanced as the noise amplitude decreases.

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. 2a) 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, \( \tau \), these structures turn into isolated but closely packed islands, reminiscent of the QDs reported experimentally (Fig. 2b). Note that upon a closer inspection one can observe the emergence of hexagonal order in the island positions. As the sputtering proceeds, while the islands do not disappear, the supporting surface develops a rough profile, destroying the overall uniformity of the islands (Fig. 2c). A similar scenario is observed for \( \lambda < 0 \), the only difference being that now the islands are replaced by holes (Figs. 2d-f), reminiscent of the morphologies observed experimentally by Rusponi et al. \[6\]. The first conclusion we can draw from these results is that the development of QDs and holes is governed by the same underlying physical phenomena, the only difference being that for QDs we have \( \lambda > 0 \), 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 \( \lambda \) does not affect the dynamics of the surface evolution, but simply turns the islands into mirrored holes. Since, according to (6) the sign of \( \lambda \) is determined only by the relative magnitude of \( a_{\sigma} \) and \( a_{\mu} \), whether islands or holes appear is determined by the shape of the collision cascade, shown in Fig. 1a. Consequently, using (6) we can draw a phase diagram in terms of the reduced penetration depths \( a_{\sigma} \) and \( a_{\mu} \) that separates the regions displaying QDs versus holes (Fig. 1b). These results also indicate that the QDs and holes are inherently nonlinear objects, that can be explained only by the nonlinear theory, since, should the linear terms be responsible for their formation, the surface morphology should not depend on the sign of \( \lambda \) (indeed, Eq. (1) has a full \( h \rightarrow -h \) symmetry). In the following, we investigate the dynamics of QD and hole formation, providing further proof of their common nonlinear origin.

The crossover behavior from the linear to the nonlinear regimes can be monitored through the surface width,
\( W^2(L, t) \equiv \frac{1}{4} \sum_{x,y} \bar{h}^2(x, y, t) - \bar{h}^2 \). As Fig. 3 shows, this quantity exhibits a sharp transition at a characteristic time \( \tau \): for \( t < \tau \), the width \( W \) increases exponentially as \( W \sim \exp(\mu t/\ell^2) \), while for \( t > \tau \), \( W \) still increases but at a considerably smaller rate than an exponential \[24\]. The crossover time is given by \( \tau \sim \sigma^2 f a \) in terms of the experimental parameters \[23\], indicating its dependence on the flux and ion 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 Fig. 2a-c were recorded, indicating that no QDs exist before \( \tau \) (Fig. 2a), but they are fully developed at \( \tau \) (Fig. 2b), and their uniformity rapidly vanishes after \( \tau \) (Fig. 2c).

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 cell structure predicted by the BH theory. However, as time increases, the nonlinear terms turn on breaking the up-down symmetry of the surface. Such delayed effect of the nonlinear terms is a well known feature of the class of surface evolution equations which \( \ell \) belongs to \[21\]. The sign of \( \lambda \) determines whether QDs or holes form, these structures appearing at a characteristic size of the collision cascade (Fig. 1). Furthermore, while the temperature independent QD size reported by Facsko et al. \[8\] indicates that for GaSb this is the main relaxation mechanism, at high temperatures or in other systems thermal diffusion could be more relevant \[17,18,22\]. 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 (certainly true in any system at high temperatures) it will change only our prediction for \( \ell \) and \( \tau \). In this case we expect \( \tau \sim 2\pi \sqrt{2D_0} \exp(-E/2kT)/\nu \), 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 \( \tau \sim D_0^2 \exp(-2E/kT)/\nu \), \( \tau \sim \epsilon^{-1} \), and \( \tau \sim f^{-1} \). However, the phase diagram (Fig. 1b) and the expected dynamical evolution (Figs. 2 and 3) will not be sensitive to the nature of the relaxation mechanism.

In conclusion, we demonstrated that the nonlinear theory can fully explain the recently observed quantum dots and holes generated by sputter erosion. We showed that these nanostructures appear only when the nonlinear terms become effective, stabilizing the surface amplitude, and that the difference in the islands and holes comes in the sign of the nonlinear terms. Furthermore, we are able to predict the dependence of the characteristic time for QD formation, and find that their saturation size is given by the size of the collision cascade. We believe that these results will lead to a better understanding of the formation and evolution of these fascinating nanostructures, thus they will guide further experiments aiming to better control these systems.

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FIG. 1. (a) The characteristics of the collision cascade generated by an ion. According to Sigmund's theory of sputtering [8], an ion penetrates the surface until a penetration depth $a$, where it radiates its energy out, such that the kinetic energy generated damage, decays exponentially with the distance from $O$. For most systems the energy distribution is anisotropic, being characterized by the characteristic decay lengthscales $\sigma$ and $\mu$ 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) $8.0 \times 10^4$. (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 $256 \times 256$.

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 $256 \times 256$. The crossover time is estimated to be $\tau \approx 5.8 \times 10^4$. 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, $t=8.0 \times 10^4$ (gray columns), indicating that the width of the distribution is smaller at the crossover time, and broadens as we go beyond $\tau$. 

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