Redeposition of sputtered material is a nonlinear effect

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(Received 1 September 2010; revised manuscript received 5 December 2010; published 4 February 2011)

It is demonstrated that redeposition of sputtered material is a nonlinear effect in experiments on pattern formation induced by ion bombardment of solid surfaces. As a result, redeposition is not the physical mechanism responsible for the formation of the highly regular hexagonal arrays of nanodots sometimes produced by normal-incidence ion sputtering.

DOI: 10.1103/PhysRevB.83.075404 PACS number(s): 79.20.Rf, 81.16.Rf, 68.35.Ct

I. INTRODUCTION

Bombarding a solid surface with a broad ion beam produces a remarkable variety of self-assembled nanoscale patterns,[1] including periodic height modulations or "ripples," as well as mounds arranged in hexagonal arrays of surprising regularity. The spontaneous emergence of these patterns is not just fascinating in its own right, since in the future ion bombardment may prove to be an important tool in the fabrication of nanostructures.

The first type of pattern formation to be discovered was the ripples that often develop when the nominally flat surface of a solid is subjected to oblique-incidence ion bombardment.[2] According to the Bradley-Harper (BH) theory,[3] these ripples are the result of a surface instability caused by the curvature dependence of the sputter yield.

In 1999, experiments by Facsko *et al.* revealed that normal-incidence ion bombardment (NIIB) of GaSb can result in the formation of nanoscale mounds or "nanodots" arranged in a densely packed, highly regular hexagonal array.[4] Well-ordered hexagonal nanodot arrays can also be produced by oblique-incidence ion bombardment of InP if the sample is rotated while it is bombarded.[5] These observations are not just of academic interest: Ion bombardment is a fast and reproducible means of producing a nearly regular array of quantum dots on a semiconductor surface in a single process step. If NIIB is to be optimized as a nanofabrication tool, an understanding of how it engenders pattern formation is essential.

In the BH theory, a solid surface subject to NIIB is unstable, and the amplitude of the surface disturbance increases exponentially in time. To account for the saturation in the amplitude of the nanodots that is observed experimentally, the leading-order nonlinear term must be added to the linear BH equation of motion.[6] This gives the Kuramoto-Sivashinsky (KS) equation

$$\frac{\partial h}{\partial t} = -A\nabla^2 h - B\nabla^2\nabla^2 h + \frac{1}{2}\lambda(\nabla h)^2, \qquad (1)$$

where $h(x,y,t)$ is the height of the surface above the point (x,y) in the x-y plane at time t, $A > 0$ and λ are constants that have been computed using the Sigmund theory of sputtering,[3,7,8] and B is the surface diffusivity. Although the KS equation yields saturation, it produces no hint of short-range hexagonal order and exhibits spatiotemporal chaos.[9]

In the experiments of Facsko *et al.*,[4] the nanodot size distribution was sharply peaked and the dot arrays had short-range hexagonal order that extended over six or more lattice spacings. These observations indicate that there was a narrow band of unstable wavelengths, according to the modern theory of pattern formation.[9] By contrast, all ripple wavelengths that exceed a critical value are unstable in the linear BH theory and in theories that add nonlinear terms to the BH equation of motion.[6,10]

In an *ad hoc* attempt to explain how NIIB produces short-range hexagonal order, Facsko *et al.* added the term $-ah$ to the right-hand side of Eq. (1), yielding the so-called damped KS equation.[11] For appropriate values of the damping parameter a, this term leads to a narrow band of unstable wavelengths and to short-range hexagonal ordering. However, the equation of motion must be invariant under the transformation $h \to h + h_0$, where h_0 is a constant, and so this modification of the KS equation is not physically acceptable. Facsko *et al.* were aware of this deficiency of their model, and so proposed replacing the term $-ah$ in the damped KS equation by $-a(h - \langle h\rangle)$, where $\langle h\rangle$ denotes the spatial average of h. This yields

$$\frac{\partial h}{\partial t} = -a(h - \langle h\rangle) - A\nabla^2 h - B\nabla^2\nabla^2 h + \frac{1}{2}\lambda(\nabla h)^2. \qquad (2)$$

This modification restores the $h \to h + h_0$ invariance but does not alter the nature of the pattern formation.

It has been unclear whether or not there is any plausible physical origin for the damping term $-a(h - \langle h\rangle)$ in Eq. (2). Facsko and co-workers suggested that this term could model redeposition of sputtered material,[11] but this issue has not been resolved. In spite of this, the implications of the model of Facsko *et al.* have been studied in considerable detail.[12]

In this paper, we show that redeposition leads to the addition of a *nonlinear* term to the equation of motion for normal-incidence ion bombardment. Therefore, redeposition does not produce a narrow band of unstable wavelengths, nor does it lead to short-range hexagonal order. In addition, redeposition cannot give rise to the linear damping term added to the KS equation by Facsko *et al.*

Although redeposition must be ruled out as the causal factor for the experimentally observed pattern formation, there are other theories for the genesis of the short-range hexagonal order induced by NIIB. In the theory of Castro *et al.*, a mobile layer at the surface of the solid modifies the dynamics, leading to the addition of a second nonlinear term to the KS equation.[10] This theory accounts for the coarsening of the nanodot arrays that is sometimes observed in experiments,[4,13,14] and so represents an important contribution to the field. At first, it seemed that the Castro *et al.* nonlinearity might also produce short-range hexagonal order,[10] but more recent numerical work strongly suggests that this is not the case.[15] This is to be expected, since the equation of motion introduced by Castro *et al.* does not have a narrow band of unstable wavelengths.

Very recently, Bradley and Shipman have advanced a theory of pattern formation induced by NIIB of binary compounds.[16] In their theory, the coupling between a surface layer of altered composition and the surface topography leads to a narrow band of unstable wavelengths and to short-range hexagonal order for a certain range of the parameters. The starting point of their fully nonlinear theory is the linear theory of Shenoy *et al.*[17]

This paper is organized as follows. In Sec. II, we show how the equation of motion for the solid surface is modified by redeposition. The implications of the resulting equation of motion are explored in Sec. III. We close the paper with our conclusions in Sec. IV.

II. THE EFFECT OF REDEPOSITION

Consider the planar surface of an amorphous solid composed of a single atomic species. We place the origin on the solid surface and orient the z axis so that it is normal to the surface.

The surface is now perturbed, and so its height h above the x-y plane becomes a function of $\boldsymbol{x} \equiv x\hat{\boldsymbol{x}} + y\hat{\boldsymbol{y}}$. We assume that the surface height is a slowly varying function of position. Thus, $|\nabla h| \ll 1$ at all points on the surface, where $\nabla \equiv \hat{\boldsymbol{x}}\partial_x + \hat{\boldsymbol{y}}\partial_y$.

The perturbed surface is now subjected to normal-incidence ion bombardment. In the vacuum above the solid, the flux of ions is taken to be $-J\hat{z}$, where J is a constant independent of both position and time. The surface of the solid moves as a result of sputtering, redeposition, surface diffusion, and ion-induced surface flow,[18–20] and so h now depends on t as well as $\boldsymbol{x}$. The small slope approximation will remain valid for all $t > 0$ if the flat surface is stable. If the flat surface is unstable, on the other hand, this approximation is valid only for early times.

In modern experiments on pattern formation induced by ion bombardment, an ultrahigh vacuum is maintained: Typically, the working pressure is less than 10^{-6} bars and can be as low as 10^{-10} bars.[21] In these conditions, a sputtered atom that strikes the sample surface almost certainly traveled ballistically between its point of origin and the point where it contacts the surface once more. Indeed, in studies on the effects of ion bombardment on large amplitude surface structures like trenches and asperities, it is universally assumed that the trajectory of a sputtered atom is a straight line—see, for example, Refs. 22–26. The sputtered atoms will therefore be taken to travel ballistically in the present work. We will also make the simplifying assumption that a sputtered atom that strikes the surface is redeposited there with probability one, although this assumption does not affect our final conclusions.

Consider a point $\boldsymbol{r} = \boldsymbol{x} + h(\boldsymbol{x},t)\hat{z}$ on the surface (see Fig. 1). The projection of this point onto the x-y plane is $\boldsymbol{x}$. Let the unit normal to the surface at $\boldsymbol{r}$ be $\hat{\boldsymbol{n}}$. We wish to find $F(\boldsymbol{r},t)dA$, the number of atoms redeposited on a surface area element $d\boldsymbol{A} \equiv \hat{\boldsymbol{n}}\, dA$ centered on the point $\boldsymbol{r}$ per unit time. We will work to lowest nontrivial order in the spatial derivatives of h. As a result, we may take the sputter yield Y to depend only on the local angle of incidence α for the purpose of computing $F(\boldsymbol{r},t)dA$—the curvature dependence of the sputter yield introduces a higher-order correction to $F(\boldsymbol{r},t)dA$, which we will neglect.

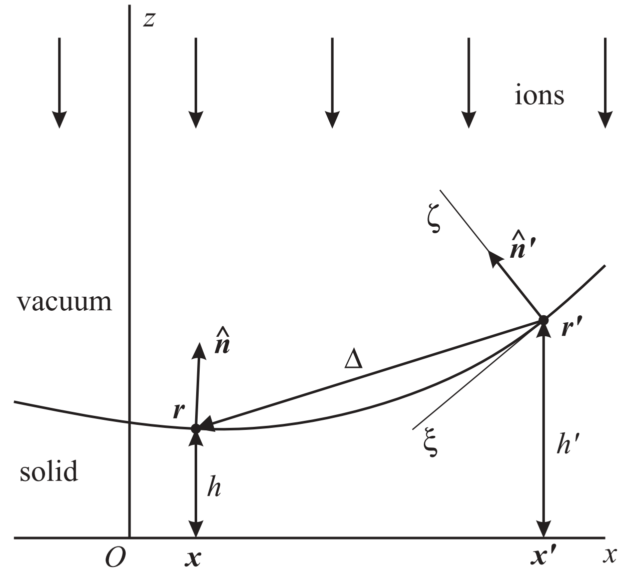

FIG. 1. Normal-incidence ion bombardment of the solid surface. For the purpose of drawing this figure, the surface height $h(x,y,t)$ has been taken to be independent of y. The curve is the surface of the solid. The surface point $\boldsymbol{r}$ is at a height $h = h(\boldsymbol{x},t)$ above the point $\boldsymbol{x}$ on the x-y plane. The unit normal to the surface at $\boldsymbol{r}$ is $\hat{\boldsymbol{n}}$. The coordinate system (ξ,η,ζ) has its origin at the point $\boldsymbol{r}'$. The ζ axis lies along $\hat{\boldsymbol{n}}'$, while the η axis points out of the page.

We begin by determining $\Phi(\boldsymbol{r},\boldsymbol{r}',t)dA\,dA'$, which is defined to be the number of atoms incident on $d\boldsymbol{A}$ per unit time that come from sputtering from the surface element $d\boldsymbol{A}' \equiv \hat{\boldsymbol{n}}'\, dA'$ centered on the point $\boldsymbol{r}'$. The ion flux incident upon $d\boldsymbol{A}'$ is $J\, d^2x'$, where $d^2x' \equiv dx'\, dy'$. We introduce a local coordinate system (ξ,η,ζ) with its origin at the point $\boldsymbol{r}'$ and with the ζ axis along $\hat{\boldsymbol{n}}'$ (see Fig. 1). The coordinate axes will be oriented so that the η axis lies along $\hat{z} \times \hat{\boldsymbol{n}}'$. The angle between $\hat{\boldsymbol{n}}'$ and $\hat{z}$ is the local angle of incidence α, which, to lowest order, is $|\nabla' h(\boldsymbol{r}',t)|$.

The atoms sputtered from $d\boldsymbol{A}'$ do not have an isotropic angular distribution. Let $f(\alpha,\theta,\phi)d\Omega$ be the fraction of the total yield sputtered into the element of solid angle $d\Omega = \sin\theta\, d\theta\, d\phi$ with polar and azimuthal angles θ and ϕ measured relative to the axes ξ, η, and ζ. Of course, the angular distribution function $f(\alpha,\theta,\phi)$ does not depend on ϕ for $\alpha = 0$. In the linear collision-cascade regime, $f(0,\theta,\phi)$ is proportional to $\cos\theta$, but experiments have revealed significant deviations from this form for both high and low ion energies.[27] For nonzero α, on the other hand, the angular distribution of the sputtered atoms is peaked around the specular direction.[27]

The precise form of f does not matter for our purposes, except for one key attribute: $f(\alpha,\theta,\phi)$ tends to zero as $\theta \to \pi/2$ for all values of α and ϕ.[28,29] Physically, this means that the sputtered atoms never graze the surface of the solid, no matter what the local angle of incidence α is. It is simple to see why this is so. Let $n(\boldsymbol{v})d^3v$ be the number density of atoms just beneath the surface of the solid with velocity $\boldsymbol{v}$ in the volume element d^3v in velocity space. Immediately below the surface, the current density of atoms with velocity $\boldsymbol{v}$ lying in the volume element d^3v is $n(\boldsymbol{v})\boldsymbol{v}\,d^3v$. Thus, the flux of atoms through $d\boldsymbol{A}$ with velocity $\boldsymbol{v}$ in d^3v is $n(\boldsymbol{v})\boldsymbol{v}\cdot d\boldsymbol{A}\,d^3v$. If $\boldsymbol{v}$ is tangent to the sample surface and so is orthogonal to $d\boldsymbol{A} = \hat{\boldsymbol{n}}\,dA$, this flux vanishes.

The solid angle that $d\boldsymbol{A}$ subtends relative to the point $\boldsymbol{r}'$ is

$$d\Omega = -d\boldsymbol{A}\cdot\boldsymbol{\Delta}/\Delta^3, \tag{3}$$

where $\boldsymbol{\Delta} \equiv \boldsymbol{r} - \boldsymbol{r}'$ is the vector leading from $\boldsymbol{r}'$ to $\boldsymbol{r}$. The number of atoms incident on dA per unit time that came from the surface element dA' is

$$\Phi(\boldsymbol{r},\boldsymbol{r}',t)dA\,dA' = JY(\alpha)f(\alpha,\theta,\phi)d\Omega\,d^2x', \tag{4}$$

provided that there is a line of sight between $\boldsymbol{r}$ and $\boldsymbol{r}'$. [Naturally, if there is no line of sight between $\boldsymbol{r}$ and $\boldsymbol{r}'$, no atoms sputtered from $d\boldsymbol{A}'$ will be redeposited on $d\boldsymbol{A}$ and so $\Phi(\boldsymbol{r},\boldsymbol{r}',t) = 0$.] Inserting Eq. (3) into Eq. (4) and again working to lowest nontrivial order in the derivatives of h, we obtain

$$\Phi(\boldsymbol{r},\boldsymbol{r}',t)dA\,dA' = -JY(0)f(\alpha,\theta,\phi) \times \frac{h - h' - (\boldsymbol{x}-\boldsymbol{x}')\cdot\nabla h}{|\boldsymbol{x}-\boldsymbol{x}'|^3}d^2x\,d^2x', \tag{5}$$

where $h \equiv h(\boldsymbol{x},t)$ and $h' \equiv h(\boldsymbol{x}',t)$.

We can simplify Eq. (5) because $\alpha \cong |\nabla' h'|$ is small and θ is close to $\pi/2$. Since

$$\hat{\boldsymbol{n}}' = \frac{\hat{z} - \nabla' h'}{\sqrt{1 + (\nabla' h')^2}}, \tag{6}$$

we have

$$\cos\theta = \frac{\boldsymbol{\Delta}\cdot\hat{\boldsymbol{n}}'}{\Delta} \cong \frac{h - h' - (\boldsymbol{x}-\boldsymbol{x}')\cdot\nabla' h'}{|\boldsymbol{x}-\boldsymbol{x}'|}. \tag{7}$$

Let $\beta \equiv \pi/2 - \theta$. Equation (7) shows that $\cos\theta = \sin\beta$ is small, and so $\beta \cong \sin\beta = \cos\theta$ is small as well. As a consequence,

$$f(\alpha,\theta,\phi) = f\left(\alpha,\frac{\pi}{2}-\beta,\phi\right) \cong f\left(\alpha,\frac{\pi}{2},\phi\right) - \beta\frac{\partial}{\partial\theta}f(\alpha,\theta,\phi)\bigg|_{\theta=\pi/2}. \tag{8}$$

$f(\alpha,\pi/2,\phi)$ vanishes because $f(\alpha,\theta,\phi)$ tends to zero as $\theta \to \pi/2$. Moreover, to lowest order we can replace α by zero in the surviving term on the right-hand side of Eq. (8). Since $f(0,\theta,\phi)$ is independent of ϕ,

$$\gamma \equiv -\frac{\partial}{\partial\theta}f(0,\theta,\phi)\bigg|_{\theta=\pi/2} \tag{9}$$

is simply a positive constant. Equation (8) therefore reduces to

$$f(\alpha,\theta,\phi) = \gamma\frac{h - h' - (\boldsymbol{x}-\boldsymbol{x}')\cdot\nabla' h'}{|\boldsymbol{x}-\boldsymbol{x}'|}. \tag{10}$$

Returning to Eq. (5), we have at last

$$\Phi(\boldsymbol{r},\boldsymbol{r}',t) = JY(0)\gamma\frac{[h - h' - (\boldsymbol{x}-\boldsymbol{x}')\cdot\nabla h][h' - h - (\boldsymbol{x}'-\boldsymbol{x})\cdot\nabla' h']}{|\boldsymbol{x}-\boldsymbol{x}'|^4}, \tag{11}$$

provided that there is a line of sight between $\boldsymbol{r}$ and $\boldsymbol{r}'$.[30]

To obtain $F(\boldsymbol{r},t)dA$, we need only sum $\Phi(\boldsymbol{r},\boldsymbol{r}',t)dAdA'$ over all surface area elements $d\boldsymbol{A}'$. It follows that

$$F(\boldsymbol{r},t) = \int \Phi(\boldsymbol{x}+h\hat{z},\boldsymbol{x}'+h'\hat{z},t)d^2x'. \tag{12}$$

Therefore, including the effects of redeposition in Eq. (1), we obtain

$$\frac{\partial h}{\partial t}(\boldsymbol{x},t) = -A\nabla^2 h(\boldsymbol{x},t) - B\nabla^2\nabla^2 h(\boldsymbol{x},t) + \frac{1}{2}\lambda[\nabla h(\boldsymbol{x},t)]^2 + \Omega_0 F(\boldsymbol{x}+h(\boldsymbol{x},t)\hat{z},t), \tag{13}$$

where Ω_0 is the atomic volume. Equation (13) is the desired equation of motion.[31]

At first sight, it appears that $\Phi(\boldsymbol{r},\boldsymbol{r}',t)$ diverges as $|\boldsymbol{x}-\boldsymbol{x}'| \to 0$, and so the validity of Eq. (13) might be questioned. In fact, $\Phi(\boldsymbol{r},\boldsymbol{r}',t)$ tends to a finite limit as $|\boldsymbol{x}-\boldsymbol{x}'|$ tends to zero. To see this, let us look at the behavior of $\Phi(\boldsymbol{r},\boldsymbol{r}',t)$ for $\boldsymbol{x}' = \boldsymbol{x} + l\hat{\boldsymbol{e}}$ in the limit that $l \to 0$; here $\hat{\boldsymbol{e}}$ is an arbitrary unit vector in the x-y plane. $\Phi(\boldsymbol{r},\boldsymbol{r}',t)$ is zero for sufficiently small l if

$$\sigma \equiv \frac{\partial^2}{\partial l^2}h(\boldsymbol{x}+l\hat{\boldsymbol{e}},t)\bigg|_{l=0} \tag{14}$$

is negative, and so we may confine our attention to the case in which $\sigma \geqslant 0$. After expanding $h' = h(\boldsymbol{x}',t) = h(\boldsymbol{x}+l\hat{\boldsymbol{e}},t)$ in a Taylor series in l in Eq. (11) and taking the $l \to 0$ limit, we obtain

$$\lim_{l\to 0}\Phi(\boldsymbol{r},\boldsymbol{r}',t) = \tfrac{1}{4}JY(0)\gamma[(\hat{\boldsymbol{e}}\cdot\nabla)^2 h(\boldsymbol{x},t)]^2, \tag{15}$$

and this is well defined and finite for a smooth surface.

III. DISCUSSION

There are obvious difficulties *a priori* with the idea that the damping term $-a(h - \langle h\rangle)$ models redeposition in high vacuum. Clearly, very little material sputtered from a surface point $\boldsymbol{r}'$ will be redeposited at $\boldsymbol{r}$ if the two points are remote from one another. Moreover, *none* of the material sputtered from $\boldsymbol{r}'$ will be redeposited at $\boldsymbol{r}$ if there is no line of sight between the two points. In contrast, all surface points $\boldsymbol{r}' \neq \boldsymbol{r}$ contribute equally to the damping term $-a[h(\boldsymbol{x},t) - \langle h\rangle]$. This is clearly unphysical.

These observations do not permit us to conclude that redeposition is not responsible for the experimentally observed pattern formation, since it could be argued that redeposition might contribute a linear term to the equation of motion with a form different from the damping term of Facsko *et al.*, and that this term could produce a narrow band of unstable wavelengths. However, it is readily apparent from Eqs. (11) and (12) that $F(\boldsymbol{r},t)$ is a nonlinear functional of h. Because redeposition is a nonlinear effect, it cannot produce a narrow band of unstable wavelengths or short-range hexagonal order.

Our detailed analysis yields further evidence that the linear damping term $-a(h - \langle h\rangle)$ cannot be used to model redeposition: Redeposition contributes a nonlinear term to the equation of motion, not a linear one. In addition, we found that $\Phi(\boldsymbol{r},\boldsymbol{r}',t)$ decays like $|\boldsymbol{x}-\boldsymbol{x}'|^{-4}$ as $|\boldsymbol{x}-\boldsymbol{x}'| \to \infty$, which makes precise our observation that little of the material sputtered from a surface point $\boldsymbol{r}'$ will be redeposited at $\boldsymbol{r}$ if the two points are widely separated.

Our study of redeposition does not completely rule out the validity of the equation of motion (2) for sputtering in high vacuum, since it could be argued that some physical effect other than redeposition might be responsible for the presence of the damping term $-a(h - \langle h \rangle)$. However, this damping term is highly nonlocal, and this severely restricts the possibilities. Shadowing and ion reflection are nonlocal phenomena, but their effects are entirely negligible for normal-incidence ion bombardment.

Anspach and Linz recently performed Monte Carlo simulations of the effects of redeposition of sputtered material.[32] Their simulations were confined to two dimensions, and the effects of surface diffusion, the curvature dependence of the sputter yield, and ion-induced surface flow were omitted. In addition, the angular distribution of sputtered material Anspach and Linz adopted does not vanish in the $\theta \to \pi/2$ limit, except when the local angle of incidence α is zero. Their simulations are therefore not expected to accurately model normal-incidence ion bombardment of a real three-dimensional solid. Although Anspach and Linz touched on the question of whether or not redeposition could lead to a damping term of the kind introduced by Facsko *et al.*, they were unable to give a definite answer. Even if they had been able to provide one, the limitations of their model would have left it open to dispute.

The fact that $\Phi(\boldsymbol{r},\boldsymbol{r}',t)$ rapidly tends to zero as $|\boldsymbol{x} - \boldsymbol{x}'| \to \infty$ suggests that it might be possible to approximate $F(\boldsymbol{r},t)$ by a term that only depends on the derivatives of h at $\boldsymbol{x}$, i.e., there might be a good, purely local approximation to $F(\boldsymbol{r},t)$. This is not the case, though. To see this, consider the scaling properties of $F(\boldsymbol{r},t)$. We set

$$h(\boldsymbol{x},t) = \langle h \rangle + W g(\boldsymbol{x}/L), \tag{16}$$

where g is a scaling function with $\langle g \rangle = 0$ and W and L are the vertical and lateral scales of the surface disturbance, respectively. This yields $F(\boldsymbol{r},t) \propto (W/L)^2$. The only term that depends solely on the derivatives of h at $\boldsymbol{x}$, that has the correct scaling properties, and that is invariant under rotations of the surface about the vertical line passing through $\boldsymbol{x}$ is $\rho[\nabla h(\boldsymbol{x},t)]^2$, where ρ is a constant. However, there is a problem with this potential local approximant to $F(\boldsymbol{r},t)$. A cursory glance at Eqs. (11) and (12) suggests that F is invariant under the transformation $h \to -h$, but this is not correct, as we can establish by considering the inverted surface $\tilde{h} \equiv -h$. (The solid fills the space beneath the inverted surface.) If there is a line of sight between the points $\boldsymbol{r} = \boldsymbol{x} + h(\boldsymbol{x},t)\hat{z}$ and $\boldsymbol{r}' = \boldsymbol{x}' + h(\boldsymbol{x}',t)\hat{z}$ on the original surface, then there is no line of sight between the points $\tilde{\boldsymbol{r}} = \boldsymbol{x} + \tilde{h}(\boldsymbol{x},t)\hat{z}$ and $\tilde{\boldsymbol{r}}' = \boldsymbol{x}' + \tilde{h}(\boldsymbol{x}',t)\hat{z}$ on the inverted surface. (Naturally, the converse of this statement is also true.) As a result, $F(\boldsymbol{r},t) = c(W/L)^2$, where the value of the constant of proportionality c depends on the sign of W. Since the term $\rho[\nabla h(\boldsymbol{x},t)]^2$ does not share this property, it is at best a crude approximation to $F(\boldsymbol{r},t)$.

Two simple physical examples are sufficient to show that $\rho[\nabla h(\boldsymbol{x},t)]^2$ can be a poor approximation to $F(\boldsymbol{r},t)$ indeed. First, consider a one-dimensional sinusoidal disturbance $h(\boldsymbol{x},t) = A \cos(kx)$, where A and k are constants. A point at the base of a trough receives redeposited material. In contrast, a point P at the top of a crest does not, because only points at the top of the adjacent crests have a line of sight to P, and sputtered atoms do not graze the surface. Thus, F vanishes at the top of a crest but not at the bottom of a trough. In contrast, $\rho(\nabla h)^2$ vanishes at both of these points.

A second example is provided by the tilted planar surface $h(\boldsymbol{x},t) = sx$, where s is a constant. In this case, F is zero at any point $\boldsymbol{r}$ on the surface, regardless of the value of the surface slope s. The term $\rho(\nabla h)^2 = \rho s^2$, on the other hand, depends on the value of s and is nonzero for $s \neq 0$.

If, despite these deficiencies, redeposition is modeled by replacing F with $\rho(\nabla h)^2$, its effect on the dynamics is simple: The value of the constant λ in the KS equation (1) is merely altered. The resulting modified equation of motion is just the KS equation, which does not produce short-range hexagonal order.

We have restricted our analysis to normal-incidence ion bombardment because that is the case of greatest experimental interest, and for the sake of simplicity. However, an analysis that is completely analogous to the one in Sec. II would show that redeposition contributes a nonlinear term to the equation of motion for oblique-incidence bombardment, just as it does for normal incidence.

IV. CONCLUSIONS

In this paper, we established that in high vacuum, redeposition of sputtered material is a nonlinear effect. Therefore, redeposition cannot produce a narrow band of unstable wavelengths and does not lead to arrays of nanodots with short-range hexagonal order. In addition, redeposition cannot give rise to the linear damping term added to the Kuramoto-Sivashinsky equation by Facsko *et al.*[11] in their attempt to model the development of hexagonal ordering during normal-incidence ion bombardment. Finally, we showed that in general the effect of redeposition at an arbitrary surface point is poorly modeled by a term that depends only on the form of the surface in the immediate vicinity of that point.

ACKNOWLEDGMENT

I would like to thank Peter Sigmund for helpful correspondence.

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[30]If the curvature dependence of the sputter yield had been taken into account in our analysis of the effect of redeposition, a correction term would appear on the right-hand side of Eq. (11). This correction is a factor of $(\nabla')^2 h'$ smaller than the term that has been retained.
[31]In Ref. 10, a thin, mobile surface layer is assumed to alter the dynamics of the solid surface. To leading order, the effect of such a layer is to introduce a term proportional to $\nabla^2(\nabla h)^2$ on the right-hand side of Eq. (13). None of the conclusions of this paper are altered if this nonlinear term is included.
[32]N. Anspach and S. J. Linz, J. Stat. Mech.: Theory Exp. **(2010)** P06023.