Comment on "Kinetic Roughening of Ion-Sputtered Pd(001) Surface: Beyond the Kuramoto-Sivashinsky Model"

Recently, Kim *et al.* [1] studied experimentally and theoretically kinetic roughening of Pd(001) surfaces eroded by ion-beam sputtering (IBS), in which large-scale roughness coexists with submicrometric moundlike structures. To describe properties of the surface height $h(\mathbf{x}, t)$, the following stochastic equation is proposed in [1]:

$$\frac{1}{c}\frac{\partial h}{\partial t} = -1 - \nu\nabla^2 h - D\nabla^4 h + \lambda_1(\nabla h)^2 + \lambda_2\nabla^2(\nabla h)^2 + \eta, \tag{1}$$

generalizing previous models of erosion by IBS [2,3] through appearance of the $\lambda_2\nabla^2(\nabla h)^2$ term. Unfortunately, the application of Eq. (1) to the experiments in [1] is hampered by mathematical and physical inconsistencies:

(i) By extending previous perturbative approaches [2,3] to a higher order, the authors obtain λ_2 as the following function of ion penetration length (a) and cascading sizes in transverse (μ) and longitudinal (σ) directions:

$$\lambda_2 = \mu^2/2 + (3/8)(\mu/\sigma)^4(\sigma^2 - a^2). \tag{2}$$

Equation (1) is linearly unstable for a band of Fourier modes $h_{\mathbf{k}}(t)$. If λ_1 and λ_2 have the same signs, the corresponding terms cancel each other in the time evolution of the Fourier mode $h_{\mathbf{k}_c}(t)$ with $k_c = (\lambda_1/\lambda_2)^{1/2}$. If k_c lies within the unstable band, this Fourier mode becomes *nonlinearly unstable*, and the continuum description breaks down [4]. Since moundlike patterns are observed in [1], necessarily [5] $\lambda_1 > 0$, thus requiring $\lambda_2 < 0$ for mathematical well-posedness. Using that [3,5]

$$\lambda_1 = (f\mu^2/2a^2)(a^2/\sigma^2 - a^4/\sigma^4 - a^2/\mu^2), \tag{3}$$

with f a positive constant, it is straightforward to see that such condition is unattainable; namely, Eqs. (2) and (3) take the same signs for any choice of parameters a, σ, and μ. Using the absolute values [6] of λ_1, λ_2 as reported on Table I in [1], $1/k_c$ is in the range 5 Å to 10 Å; hence, the nonlinearly unstable mode $h_{\mathbf{k}_c}(t)$ occurs in the experiments in [1], and Eq. (1) breaks down as a continuum description of this physical system.

(ii) For the three values of the average ion energy ε studied in [1], λ_1 is reported to be *negative*, as computed by the TRIM package. As shown in [5] and confirmed by numerical integration of Eq. (1), this would lead to production of *holes*, rather than the observed *mounds* [1]. Thus, for parameters of Table I, Eq. (1) does not produce the type of morphologies found experimentally.

(iii) Finally, the authors argue that the experimental exponents are in good agreement with those of the conserved Kardar-Parisi-Zhang (CKPZ) equation (1) with $\nu = \lambda_1 = 0$, namely [7], $\alpha \simeq 2/3$, $\beta \simeq 0.2$, and $1/z \simeq 10/3$. Although this value of β agrees with the one reported, such is not the case for α or $1/z$. E.g., for $\varepsilon = 0.5$ keV, the observed values are $\alpha_{\text{exp}} \simeq 1$, $z_{\text{exp}}^{-1} \simeq \beta_{\text{exp}} \simeq 0.2$. It is stressed in [1] that α must be close to 1 due to the moundlike structures formed, and then $\beta = \alpha/z = 1/z$, but this property does not hold for the CKPZ equation, nor does it for Eq. (1), as can be checked by numerical integration using parameters in [1].

In summary, Eq. (1), as derived within the approximations in [1–3], is not a well-defined continuum description of the experiments in [1]. Further morphological analysis in [1] is hampered by physical inconsistencies. Recently, a related equation has been derived to describe nanopatterning by IBS [8]. However, to achieve a mathematically consistent framework, additional physical mechanisms are needed over those previously considered [1–3].

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