

Stability of a strongly anisotropic thin epitaxial film in a wetting interaction with elastic substrate

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received 16 October 2010; accepted in final form 31 December 2010

published online 4 February 2011

PACS 68.55.J- – Morphology of films

PACS 81.15.Aa – Theory and models of film growth

PACS 81.16.Dn – Self-assembly

Abstract – The linear dispersion relation for surface perturbations, as derived by LEVINE *et al.*, *Phys. Rev. B*, **75** (2007) 205312, is extended to include a smooth surface energy anisotropy function with a variable anisotropy strength (from weak to strong, such that sharp corners and slightly curved facets occur on the corresponding Wulff shape). Through detailed parametric studies it is shown that a combination of a wetting interaction and strong anisotropy, and even a wetting interaction alone results in complicated linear stability characteristics of strained and unstrained solid films.

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Introduction. – Studies of the morphological instabilities of thin solid films are a first step towards understanding complex phenomena such as the formation of three-dimensional nanoscale islands in strained alloy heteroepitaxy. Such studies became common after the pioneering works of Asaro and Tiller [1], Grinfeld [2] and Srolovitz [3]. The classical Asaro-Tiller-Grinfeld instability is one of a uniaxially stressed solid film on a rigid infinite substrate. Its variants for the single-component and alloy films on rigid as well as on deformable substrates have been studied and this research continues. Reviews of works on the single-component films have been published, see for instance ref. [4].

Film-substrate wetting interaction is a relatively new concept in the field of research on morphological instability and evolution. When surface slopes are not very large, this additional wetting energy can be considered a function of the local film thickness h , but not of the slopes of h [5–7]. In refs. [8–11] and others it has been shown that wetting interaction damps long-wave instability modes in a certain range of film thickness, thus changing the instability spectrum from long-wave type to short-wave type. The latter mode of instability is more relevant to the process of formation of

island arrays [12]. In ref. [9] it is recognized that in the presence of wetting interaction, the boundary conditions that describe the stress balance at the film free surface and at the film-substrate interface must be augmented by *wetting stress* terms—that are proportional to the rate of change of the surface energy with h (see footnote ¹). Wetting stress and lattice-mismatch stress have different origin, and the former may be present even when the latter is absent. For wetting (non-wetting) films, the solution of the elastic free-boundary problem with boundary conditions that include wetting stress terms, results in additional destabilizing (stabilizing) contributions in the dispersion relation. Some stability characteristics have been analyzed in ref. [9] within the framework of long-wave approximation, where in addition the surface energy is assumed isotropic. This letter extends that work by adding strong anisotropy and considering not only wetting films, but also non-wetting films. Here we recognize that the film thickness and the wetting length are two *independent* characteristic lengths, *i.e.* the former length is determined by film deposition, while the latter

¹Wetting stress is called conjoining (or disjoining) pressure when studying the dynamics of thin *liquid* films on substrates [13,14]. This pressure is partially responsible for the so-called spinodal instability, which typically leads to film dewetting (rupture); for discussions of spinodal instability, see for instance ref. [15] and references therein.

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one is determined by the molecular structure and properties of the film-substrate interface. Since the wetting length may be, and normally is, less than the deposited film thickness, the perturbation wavelengths may be comparable to the film thickness but still much larger than the wetting length. In this case the long-wavelength approximation may hold with respect to the wetting length, but not with respects to the film thickness. In the fourth section we show that this approach reveals linear stability features that, we believe, went unnoticed in prior publications.

Problem statement. – Following refs. [9,16], we consider a dislocation-free, one-dimensional, single-crystal, epitaxially strained thin solid film in a wetting interaction with a solid, semi-infinite elastic substrate. The film surface $z = h(x, t)$ evolves due to surface diffusion. This evolution is described by

$$\frac{\partial h}{\partial t} = \frac{\Omega DN}{kT} \frac{\partial}{\partial x} \left[\left(1 + \left(\frac{\partial h}{\partial x} \right)^2 \right)^{-1/2} \frac{\partial \mathcal{M}}{\partial x} \right], \quad (1)$$

where Ω is the atomic volume, D is the adatoms diffusivity, N is the adatoms surface density, kT is the Boltzmann factor, and \mathcal{M} is the surface chemical potential [17]. The latter has contributions from the elastic energy in the film, the anisotropic surface energy, and a wetting interaction:

$$\mathcal{M} = \mathcal{E}(h) + \Omega \left[\left(\gamma + \frac{\partial^2 \gamma}{\partial \theta^2} \right) \kappa - \delta \left(\frac{\kappa^3}{2} + \frac{\partial^2 \kappa}{\partial S^2} \right) + \left(\frac{\partial \gamma}{\partial h} - \text{sign}(h_x) \frac{\partial^2 \gamma}{\partial h \partial \theta} \right) \cos \theta \right], \quad (2)$$

where θ is the angle that the unit surface normal makes with the reference crystalline direction, say [01] (chosen along the z -axis, which is normal to the substrate), $\gamma(h, \theta)$ is the height- and orientation-dependent surface energy, κ is the curvature of the surface, S is the arclength along the surface and h_x is the surface slope (note, $\partial/\partial S = (\cos \theta) \partial/\partial x = (1 + h_x^2)^{-1/2} \partial/\partial x$). The term proportional to the small positive parameter δ is the regularization that is required in view of ill-posedness of eq. (1) for strong anisotropy, that is when $\epsilon_\gamma \geq 1/(m^2 - 1)$ in eq. (4) below [18–21]. Note also that the mixed derivative term in eq. (2) is nonlinear and thus it has no impact on linear stability.

In this letter we consider the two-layer exponential model for the surface energy [5,7]:

$$\gamma(h, \theta) = \gamma_t(\theta) + (\gamma_s - \gamma_t(\theta)) \exp(-h/\ell), \quad (3)$$

where γ_s is the surface energy of the substrate when there is no film, ℓ is the characteristic wetting length, and $\gamma_t(\theta)$ is the anisotropic surface energy of a thick film:

$$\gamma_t(\theta) = \gamma_0(1 + \epsilon_\gamma \cos m\theta), \quad \epsilon_\gamma \geq 0. \quad (4)$$

Here γ_0 is the mean surface energy, ϵ_γ is the strength of anisotropy and m is the integer parameter specifying

anisotropy type (*i.e.*, four-fold, six-fold, etc.) By comparison with experimental and *ab initio* computational studies the two-layer exponential model has been shown the most accurate to-date [22,23]. In the absence of anisotropy, $\gamma_t = \gamma_0 = \text{const}$, and δ vanishes. δ is taken zero also at weak anisotropy, $\epsilon_\gamma < 1/(m^2 - 1)$. In reality, the maximum of $\gamma_t(\theta)$ might occur at $\theta = \beta$, where the non-zero angle β is a misorientation from the reference direction. Without significant loss of generality we assume $\beta = 0$ in eq. (4).

The expression for the elastic energy $\mathcal{E}(h)$ in eq. (2) is derived in ref. [16] without accounting for wetting interaction. Wetting interaction is considered in several papers, including refs. [8–11,22,24–26] (in ref. [26] the wetting effect arises not from the dependence of surface energy on thickness, but from the thickness-dependent elastic energy, which cannot be calculated from linear elasticity theory). To this end, by combining expressions derived by us in refs. [9,25] we state the dimensionless linear growth rate *in the longwave limit*, $kh_0 \ll 1$ (where k is the perturbation wave number and h_0 is the uniform thickness of unperturbed planar film):

$$\begin{aligned} \omega(h_0, k, \mu, \epsilon, \epsilon_\gamma) &= A\epsilon^2 (\mu + A_1 h_0 k) k^3 \\ &- B\epsilon [\mu (h_0 - 1) + h_0 (B_1 h_0 - A_1) k] k^3 a \exp(-h_0) \\ &+ F [(\Lambda - (G + \Lambda) \exp(-h_0)) k^4 - \Delta k^6 \\ &- ak^2 \{\exp(-h_0) - B_2 a \exp(-2h_0)\}]. \end{aligned} \quad (5)$$

ℓ has been chosen as the length scale and ℓ^2/D as the time scale. Here ϵ is the misfit strain in the film, and $\mu = \mu_f/\mu_s$ is the ratio of the film shear modulus to the substrate shear modulus. Other parameters are:

$$A = \frac{8N\Omega^2 \alpha_s (1 + \nu_f)^2 \mu_f}{kT\ell \alpha_f^2}, \quad B = \frac{4N\Omega^2 \gamma_0 \alpha_s (1 + \nu_f) \nu_f}{kT\ell^2 \alpha_f^2}, \quad (6)$$

$$A_1 = \frac{2C_1}{\alpha_f \alpha_s}, \quad B_1 = \frac{\nu_f C_2 - \alpha_f}{2\alpha_f \alpha_s \nu_f}, \quad B_2 = \frac{\beta_f \gamma_0}{\alpha_f \mu_f \ell}, \quad (7)$$

$$C_1 = \alpha_f + \alpha_f \beta_s \mu - \alpha_s^2 \mu^2, \quad C_2 = 4\alpha_f + 3\alpha_f \beta_s \mu - 4\alpha_s^2 \mu^2, \quad (8)$$

$$\alpha_{f(s)} = 2(1 - \nu_{f(s)}), \quad \beta_{f(s)} = 1 - 2\nu_{f(s)}, \quad (9)$$

$$F = \frac{N\Omega^2 \gamma_0}{kT\ell^2}, \quad \Delta = \frac{\delta}{\gamma_0 \ell^2}, \quad (10)$$

$$\Lambda = (m^2 - 1) \epsilon_\gamma - 1, \quad G = \gamma_s/\gamma_0, \quad a = G - 1 - \epsilon_\gamma. \quad (11)$$

In eqs. (6)–(9) ν is Poisson’s ratio. Note the coupling of wetting interaction and misfit strain through the term proportional to ϵ (the second line of eq. (5)). This term, responsible for breaking symmetry between compressive and tensile stress states, drops out of the growth rate in the absence of wetting interactions, $h_0 \rightarrow \infty$. In the square brackets of this term, $-\mu$ and $-A_1 h_0 k$ are the contributions from the wetting stress; see also refs. [9,25]. Another contribution from the wetting stress is the term proportional to $\exp(-2h_0)$ in the square brackets in the last line of the equation.

Our goal is to elucidate the roles of anisotropy, wetting interaction and wetting stress and to characterize film stability in the space of dimensionless parameters h_0 , k , μ , ϵ and ϵ_γ . Other material parameters will be fixed to their most characteristic values. We choose the following values: $D = 1.5 \times 10^{-6}$ cm²/s, $N = 10^{15}$ cm⁻², $\Omega = 2 \times 10^{-23}$ cm³, $kT = 1.12 \times 10^{-13}$ erg, $\gamma_0 = 2 \times 10^3$ erg/cm², $\nu_f = 0.198$, $\nu_s = 0.217$, $\mu_f = 10^{12}$ erg/cm³, $\delta = 5 \times 10^{-12}$ erg, and $\ell = 3 \times 10^{-8}$ cm. The value of the characteristic wetting length is of the order of 1 ML thickness for InAs or Ge film [22]. We assume strong anisotropy, *i.e.* $\epsilon_\gamma > 1/(m^2 - 1) \equiv \epsilon_\gamma^{(c)}$ and thus $\Lambda > 0$. For strained films considered in the fourth section, we choose $m = 32$ as the most characteristic value [27]. However, as far as the effect of anisotropy on linear stability is of interest, similar results are obtained for other common values such as $m = 4$ or $m = 6$. That is, choosing larger m simply means that smaller values of ϵ_γ are above the critical value $\epsilon_\gamma^{(c)}$. *Wetting films* require $a > 0$ [25], thus we choose $\gamma_s = 2\gamma_0$, $\epsilon_\gamma^{(c)} < \epsilon_\gamma < 1$. For the analysis of *non-wetting films* ($a < 0$), we choose $\gamma_s = \gamma_0/2$, $\epsilon_\gamma > \epsilon_\gamma^{(c)}$. It is clear that wetting stress terms (pointed out above) are destabilizing (stabilizing) in wetting (non-wetting) films.

Films with wetting interaction and zero misfit strain and wetting stress. – When misfit strain and wetting stress are not present, eq. (5) reduces to

$$\omega(h_0, k, \epsilon_\gamma) = F [(\Lambda - (G + \Lambda) \exp(-h_0)) k^4 - \Delta k^6 - a k^2 \exp(-h_0)]. \quad (12)$$

First, we consider wetting films. It follows that the perturbations with wave numbers larger than $k_c = \sqrt{\Lambda/\Delta}$ cannot destabilize a film of any thickness. (Here, k_c is not the customary cut-off wave number, but is determined from the condition $\omega < 0$ for any h_0 .) However, in the opposite case $k < k_c$ only the films of thickness that is less than the critical, $h_0^{(c1)}$, are stable:

$$h_0 < h_0^{(c1)} = -\ln \frac{\Lambda k^2 - \Delta k^4}{a + (G + \Lambda) k^2}. \quad (13)$$

With $\Delta = 25/9$ corresponding to the material parameters stated above, $m = 4$, and $\epsilon_\gamma = 0.1$, we obtain $k_c = 0.42$. Taking typical $k = 0.1k_c$ in eq. (13) gives $h_0^{(c1)} = 6.94$, which translates to the dimensional value of 7 ML. Figure 1 shows the contour plot of $h_0^{(c1)}(k, \epsilon_\gamma)$. It can be seen that stronger anisotropy decreases $h_0^{(c1)}$. We notice also that strong anisotropy destabilizes (that is, the contribution proportional to k^4 in the square bracket of eq. (12) is positive) only relatively thick films, such that

$$h_0 > h_0^{(c2)} = -\ln \frac{\Lambda}{G + \Lambda}. \quad (14)$$

For the chosen values, $h_0^{(c2)} = 1.6$ ML. Such threshold-type influence of strong anisotropy is distinctly different

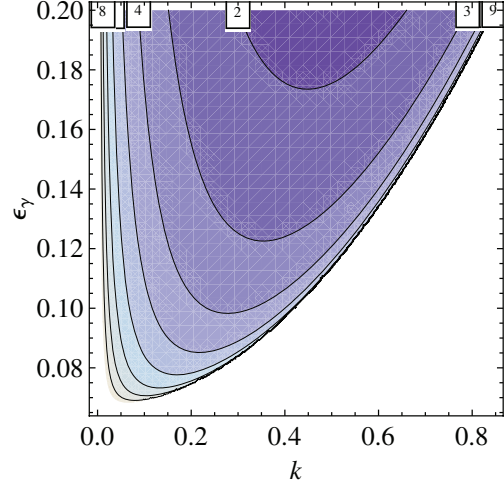


Fig. 1: (Colour on-line) Contour plot of the critical thickness $h_0^{(c1)}$ for strong anisotropy, $\epsilon_\gamma > \epsilon_\gamma^{(c)} = 1/15$.

from the simplified model in which wetting interaction is absent. The latter model can be obtained by taking $h_0 \rightarrow \infty$ in eq. (12), and thus this equation becomes $\omega(k, \epsilon_\gamma) = F(\Lambda k^4 - \Delta k^6)$, from which it is clear that strong anisotropy has destabilizing influence on a film of arbitrary thickness. These findings to some extent echo refs. [22,26], where the existence of the critical perturbation amplitude that is necessary to destabilize a film in the presence of a cusp in the surface energy $\gamma(\theta)$ (which is the case below the roughening temperature), has been demonstrated. Thus if a film is thin, critical amplitude may be unattainable and the film will not be destabilized. However note that models of refs. [22,26] do not allow straightforward separation of the effects of surface energy and mismatch stress, and thus our results cannot be easily compared to these papers.

Next, we consider non-wetting films. One example of such material system may be the energetically driven dewetting of silicon-on-insulator [28,29]. Repeating the analysis and referring to the critical values shown above, it follows that a film of any thickness is stable with respect to perturbations with wave numbers larger than $\max(k_c, k_c^{(u)})$, where $k_c^{(u)} = \sqrt{-a/(G + \Lambda)}$. If $k_c < k < k_c^{(u)}$, then the film is stable if $h_0 > h_0^{(c1)}$ and unstable otherwise. If $k_c^{(u)} < k < k_c$, then the film is stable if $h_0 < h_0^{(c1)}$ and unstable otherwise. Finally, if $k < \min(k_c, k_c^{(u)})$, then the film of any thickness is unstable. With $G = 0.5$ and $\epsilon_\gamma = 0.1$, $k_c^{(u)} = 0.77 > k_c$, and therefore the second possibility, $k_c^{(u)} < k < k_c$, must be dismissed. Typically, the first scenario ($k_c < k < k_c^{(u)}$) holds, and thus there is a critical thickness below which the film is unstable [10].

Results similar to those shown above for wetting and non-wetting films can be obtained (numerically) with non-zero wetting stress, since the negative exponent $\exp(-2h_0)$

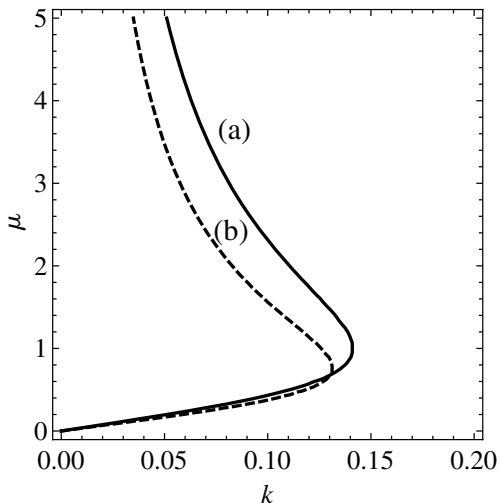


Fig. 2: Zero level curve of $f(k, \mu, h_0) = \mu(h_0 - 1) + h_0(B_1 h_0 - A_1)k$. (a) $h_0 = 2$, (b) $h_0 = 3$. To the left of each curve, the symmetry-breaking term in the long-wave growth rate (5) (second line) is stabilizing, to the right —destabilizing (when $a, \epsilon > 0$).

decays fast compared to the terms in eq. (12) that are proportional to $\exp(-h_0)$.

Wetting films with non-zero misfit strain and wetting stress. — The situation presented in this section is common for Stranski-Krastanov growth of epitaxial thin films.

As we pointed out in the second section, in the presence of misfit strain and wetting interaction, eq. (5) contains the term that is proportional to the first power of misfit strain. Whether this term is destabilizing or stabilizing (for, say, $\epsilon > 0$) depends on the sign of the expression $f(k, \mu, h_0) = \mu(h_0 - 1) + h_0(B_1 h_0 - A_1)k$. Only for sufficiently small k and large μ this is positive. Then the second term in eq. (5) is stabilizing as shown in fig. 2. Increasing h_0 makes the domain of stabilization smaller. As μ is in the range 0.5–1.0 for a typical heteroepitaxial semiconductor system, the coupling of misfit strain and wetting interaction has stabilizing effect on an ultrathin film of thickness of the order of several wetting lengths, for *long-wave perturbations*. Note that the standard ϵ^2 -term is always destabilizing for all perturbation wavelengths [16].

In order to demonstrate some effects of arbitrary relation between wetting length, film thickness and the perturbation wavelength, in conjunction with strong anisotropy, we use next the full *dimensional* growth rate expression involving hyperbolic functions of kh (see footnote ²), where k, h are now the dimensional wave number and mean thickness, respectively. (Equation (5) emerges upon expansion of this growth rate in powers of small dimensionless parameter kh , retaining the dominant terms of the expansion

²Available on request from authors.

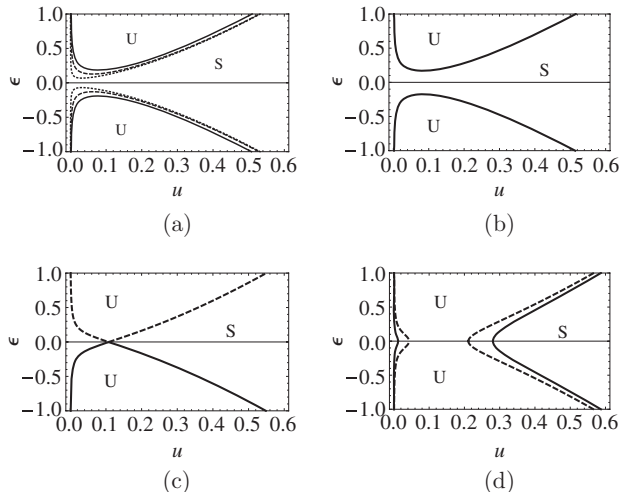


Fig. 3: Neutral stability curves. (a) $\epsilon_\gamma = 0$. $r = 0.1$ (solid), $r = 1$ (dash), $r = 3$ (dot). (b)–(d): $\epsilon_\gamma = 0.01$. (b) $r = 0.1$, (c) $r = 0.518$, (d) $r = 1$ (dashed), $r = 3$ (solid). Domains of surface stability (instability) are marked by S (U).

(long-wave approximation), and non-dimensionalization.) As in eq. (5), the full growth rate is quadratic in ϵ , allowing one to explicitly determine the boundaries of neutral stability, $\omega = 0$, in the r - ϵ or u - ϵ planes. Here r and u (dimensionless) are defined by $h = r\ell$, $k = u(2\pi/\ell)$.

Figure 3 shows neutral stability curves, in the u - ϵ plane, for $\epsilon_\gamma = 0$ and 0.01. (For the value $m = 32$ used in this section, $\epsilon_\gamma^{(c)} = 0.001$.) For all three values of a film thickness in the former (isotropic) case, and for the smallest value in the latter (strongly anisotropic) case, the film is destabilized by *short-wavelength* perturbations, $u > 0.02$, above some critical value of the misfit parameter ϵ . Increasing film thickness in the isotropic case to values as large as 50ℓ only makes the domain of stability shrink. However, for larger film thickness in the strongly anisotropic case (figs. 3(c), (d)) two stability domains emerge separated by the domain of instability. The splitting of a single domain into two domains occurs at $r = 0.5$. The size of stability domains decreases with increasing film thickness. Overall, the film is less stable with increasing anisotropy (as expected). Note that instability in figs. 3(c), (d) is present for some u even when misfit is zero. Responsible for this is the combined destabilizing effect of anisotropy and wetting stress, which together outweigh the stabilizing effect of the wetting layer; also see the third section. Similar behaviour is observed for increasing μ while keeping the thickness fixed. We also notice that only the $r = 0.1$ case can be (probably) captured by the long-wave approximation, as $kh = 2\pi r u = 0.38 \sim 1$ for $r = 0.1, u = 0.6$, and is even larger for other values of r in fig. 3. In order to characterize the horizontal spacing between two stability domains in figs. 3(c), (d), in fig. 4 we plot the neutral stability curve corresponding to the level $\epsilon = 0$. It can be seen that for all reasonable r this

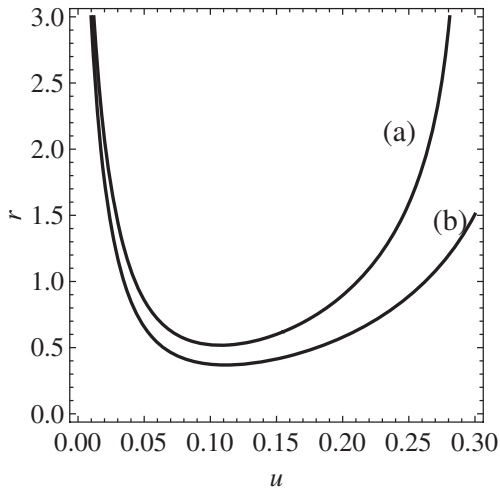


Fig. 4: Neutral stability curves (see text). (a) $\epsilon_\gamma = 0.01$, (b) $\epsilon_\gamma = 0.014$. The film is stable below each curve.

spacing does not exceed 0.3. For comparison, the case of slightly larger anisotropy is also shown.

To summarize, we considered all combinations of wetting interaction (through the exponential two-layer model), lattice-mismatch and wetting strains, and strong anisotropy. Our results demonstrate the complicated linear stability of ultrathin films ($h \sim 1:5$ wetting lengths). In particular, we show that extremely thin ($h \sim 1:2$ wetting lengths), unstressed wetting films are not destabilized by arbitrarily strong anisotropy. Anisotropic, stressed wetting films are destabilized by any level of mismatch stress, but only in the narrow range of perturbation wave numbers. Such films can remain stable with respect to short-wavelength perturbations when they are very thin and at any reasonable mismatch stress level. Our final remark concerns two-dimensional surfaces and corresponding surface energy anisotropies $\gamma_t(h_x, h_y)$ of the generic form (4) (where contribution in the y -direction is additive, as is commonly assumed). We conjecture that, if the surface orientation (of a thick film) is still one of the high-symmetry crystallographic orientations, such as [001] or [111], then the effect of such in-plane anisotropy is *nonlinear* and thus the latter anisotropy will not affect the results. This can be qualitatively understood, for instance, by following the analysis leading to eq. (15) in ref. [8] while accounting for the nonlinear nature of the mixed derivative term in eq. (2) and the form of eq. (3), in this letter. Due to the complexity of formulation and derivation, the exact proof is beyond the scope of this note. The results are also unchanged if the surface is two dimensional but the in-plane anisotropy is zero.

MK acknowledges the support of WKU Faculty Scholarship Council via grants 10-7016 and 10-7054, and thanks

B. J. SPENCER (University at Buffalo) for comments on the first draft of the manuscript.

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