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Surface island formation with localized stresses

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Motivated by recently reported experimental observations, the role of surface structures in the formation of surface islands by the Asaro–Tiller–Grinfeld–Srolovitz instability was investigated via computer simulations using the phase field method. These structures caused a spontaneous ordering in the form of a wavelike marching of surface islands during instability. The simulation results were used to explain a particular feature of the previously reported experimental observations, namely the existence of regular bands of nanoislands on yttria-stabilized zirconia/gadolinia-doped ceria heteroepitaxial surfaces.

Keywords: Surface structure; Thin films; Self-organization and patterning; Simulation; Phase field models

Recently, Rauscher et al. [1] reported on the discovery of nanoisland formation in gadolinia-doped ceria (GDC)/yttria-stabilized zirconia (YSZ) hetero-epitaxial systems. They found that after deposition of GDC on YSZ single-crystal surfaces spallation would sometimes occur, leaving behind a surface of unknown composition decorated by large regions of remnant GDC pads (Fig. 1). Upon heat treatment of these surfaces, pseudo-periodic arrays of nanoislands were observed – often forming randomly in the spalled region, but always occurring in bands at the border between remnant pads and spalled surfaces (Fig. 1). These islands were spatially ordered, formed pseudo-periodic square lattices and were nearly uniform in size.

The formation of nanoislands is generally understood to proceed by one of two mechanisms: dewetting [2] or strain-induced surface instability [3]. In dewetting, island formation is driven by a reduction in interfacial energy; in strain-induced instability, a reduction in strain energy is the driving force. For this work, we have assumed that the strain driven mechanism is operative. Our basis for this assumption is the nature of the observed island morphologies, where a very strong alignment along the elastically "soft" directions of the substrate and a sharply uniform size distribution is found [1]. Both of these observations are highly suggestive of a strain-induced island formation mechanism [4–6], and may be contrasted with the tendency of dewetted islands to form without spatial correlation and to possess a broad size distribution [2].

Surface instability due to the presence of stresses is described by the Asaro–Tiller–Grinfeld–Srolovitz (ATGS) instability [7–9], where strain energy is minimized at the expense of interfacial energy via island formation [10–14]. However, even given the assumption that island formation proceeds via the ATGS instability, understanding the striking self-organization of this system invites further study. Rauscher et al. [1] speculated that band formation occurred due to a catalytic effect of the remnant pad edges, possibly due to variations in the local stress.

Computational models, and in particular phase-field models, have been extensively employed to investigate the ATGS instability [13,15–18]. For the most part, this work has focused on the stability of homogeneous films against roughening in the presence of stress. We are aware of two publications which have discussed the effect of heterogeneities on surface instability. Eggleston and Voorhees [19] showed that the presence of surface mesas could engender a wavelike marching of surface instability in a growing thin film during deposition. This effect was attributed to a capillary instability due to the geometrical tendency of surface mesa corners to accumulate excess deposited material relative to planar regions, and to surface energy anisotropy. Wise et al. [20] demonstrated a similar wavelike ordering during deposition which was induced by strain patterning because of the presence of misfitting inclusions in the substrate.

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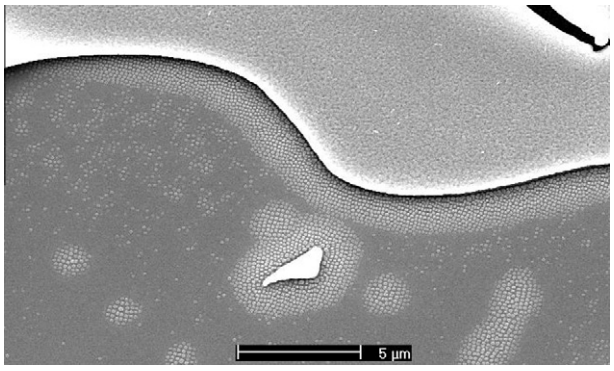

Figure 1. SEM micrograph of island bands surrounding remnant GDC patches (top and center).

In the context of these observations, we wish to investigate the local stress state associated with remnant surface patches and its effect on the formation of island bands. We construct a phase field model to study stress-induced surface instabilities in a thin film with arbitrary surface morphology and arbitrary distribution of local strain. The results of Eggleston and Voorhees and Wise et al. are extended to the case of a static film with inhomogeneous stress distributions. By choosing our model parameters to reflect the essential features of the experimental system [1], we show that island banding is due to the presence of structural heterogeneities in the film surface.

The experimental system [1] can be characterized by three principal material divisions: the bulk YSZ substrate, remnant surface patches of sputtered GDC and the surface exposed by spallation. In this investigation, only the effects of elastic strain energy will be considered. Remnant patches of deposited material are interpreted as surface inhomogeneities and possible local sources of stress beyond coherency strains (i.e., deposition stress [21,22]), neglecting any chemical inhomogeneity.

Thus, a simple two-dimensional system is considered, as shown in Figure 2. We first distinguish between solid and void. The solid is further subdivided into a substrate region, a film region and remnant patches. Stress may or may not be present from two distinct sources: deposition stress in the remnant patches, and stress due to lattice misfit between the substrate and the film. With these simplifications we investigate the self-organization observed in Ref. [1].

We employ the phase field method developed in Ref. [17], using the Cahn–Hilliard (CH) equation and the phase field microelasticity (PFM) method of Wang et al. [23], as these methods permit a large degree of

Figure 2. Schematic of the computational cell as it is employed in this work. Values for the important numerical fields vary within different regions: in the vapor, $c(\mathbf{r}) = 0$ and $\epsilon_{ij}^{T0}(\mathbf{r}) = 0$; in the substrate. $c(\mathbf{r}) = 1$ and $\epsilon_{ij}^{T0}(\mathbf{r}) = 0$; in the patch. $c(\mathbf{r}) = 1$ and $\epsilon_{ij}^{T0}(\mathbf{r}) = 0$; and in the film. $c(\mathbf{r}) = 1$ and $\epsilon_{ij}^{T0}(\mathbf{r}) = 0$.

variation in the local strain distribution. The CH equation describes the time-evolution of a conserved field, such as concentration:

$$\frac{\partial c}{\partial t} = \nabla\left[M\nabla\frac{\delta F}{\delta \eta}\right] \tag{1}$$

where M is the chemical mobility and F is the coarse-grained free energy. In this work, c refers to concentration in a two-phase (solid and void) system, varying between zero and one. Free energy F consists of separate chemical and elastic components.

The first bracketed term on the right-hand side contains the local chemical free energy and the gradient energy term, where κ is a constant. We define f_c:

$$F = \int_V \left[f_c(c) + \frac{1}{2}\kappa(\nabla_c(\mathbf{r}))^2\right] + E^{elast}(c,\epsilon) \tag{2}$$

$$f_c(c) = \begin{cases} Ac^2(1-c)^2 & \text{in the void and film} \\ Ac^2 & \text{in the substrate and patch} \end{cases} \tag{3}$$

In the void and film regions, the double-well polynomial in Eq. (3) yields a two-phase equilibrium system. We assign one minimum of c as the "solid" phase, and the other as a "void" or "vapor" phase. In the substrate and patch regions, we employ a single-well potential to restrict evolution. This ensures that the non-film regions will remain constant in shape, which is convenient for the specification of $\epsilon_{ij}^{T0}(\mathbf{r})$.

For the treatment of elastic energy, the PFM method developed by Wang et al. [17], specifically their method for the treatment of voids, is employed. The PFM method is a technique used in the context of phase field modeling to solve equations of linear elasticity for a field of arbitrary structural and modulus inhomogeneities. Structural variations are described via the stress-free transformation strain $\epsilon_{ij}^{T0}(\mathbf{r})$. In this work, ϵ_{ij}^{T0} will be used to characterize deposition and misfit stresses in the model thin-film system. A full derivation is provided in the series of papers by Wang et al., and thus further details are deferred [23].

In the scope of this communication, it is only necessary to note that we represent the free surface (i.e., the void/solid interface) by explicitly linking the spatially dependent elastic modulus tensor to the phase field $c(\mathbf{r})$. We define ρ, the material density distribution, consistent with the previous definition of c, such that density is vanishing in the void:

$$\rho(\mathbf{r}) = \frac{1}{2}\left[\tanh\frac{2c(\mathbf{r})-1}{2\tau} + 1\right] \tag{4}$$

where τ is a parameter that controls the sharpness of the transition between solid and void ($\tau = 0.1$ in this work). This, in turn, defines the spatially variant elastic modulus tensor field: $C_{ijkl}(\mathbf{r}) = C^0_{ijkl}\rho(\mathbf{r})$. It is necessary to adopt $\rho(\mathbf{r})$ rather than make C_{ijkl} vary directly with c in order to prevent numerical instability [17]. $C_{ijkl}(\mathbf{r})$ is then used in the solution of the elastic equilibrium equation at each time step.

Lastly, the dimensionless parameters governing the evolution of these equations are determined. Within the CH equation, these are κ and A, which together govern the interfacial energy of the model. In order to