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Current-driven morphological evolution of single-layer epitaxial islands on crystalline substrates

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ABSTRACT

We develop and validate a nonlinear model for the current-driven dynamics of single-layer epitaxial islands on crystalline substrates. Simulations based on the model show that the dependence of the stable steady island migration speed v_m on the inverse of the island size is not linear for larger-than-critical island sizes. In this nonlinear regime, we report morphological transitions, Hopf bifurcations, and instabilities for various surface crystallographic orientations and island misfit strains. Proper rescaling of v_m gives a universal linear relationship for its dependence on island size.

The driven assembly of confined quantum structures is of special importance to nanoelectronics and nanofabrication technologies. A particularly interesting problem is the current-driven dynamical response of single-layer adatom and vacancy clusters, i.e., islands and voids of single-layer thickness/depth, on substrate surfaces. Several theoretical studies have been conducted to address this problem. Mehl and coworkers [1] demonstrated a monotonic decrease in the island drift velocity with island size in kinetic Monte Carlo (KMC) simulations of electromigration-induced motion of single-layer islands on the Cu(001) surface. They found edge diffusion, i.e., adatom migration along the island edge, to be the dominant mass transport mechanism [1], while other diffusion mechanisms also have been observed in a recent molecular-dynamics study of Ag island diffusion on Cu(001) [2]. Pierre-Louis and Einstein [3] studied theoretically and with KMC simulations the electromigration-induced dynamics of single-layer atom and vacancy clusters on surfaces of isotropic media, including the mass-transport regimes of periphery diffusion, terrace diffusion, and evaporation–condensation (attachment/detachment) and examined the morphological stability of the islands. In the limit of periphery diffusion, they found the drift velocity of steady islands, v_m, to be inversely proportional to the island size, R_s, i.e., $v_m \propto 1/R_s$ [3]. Hausser and coworkers reported an electromigration-induced fingering instability of the circular steady state of single-layer vacancy islands on crystal surfaces in a continuum model in the terrace diffusion regime [4]. Complex morphological evolution has been reported for electromigration-driven single-layer adatom islands on solid surfaces, reaching oscillatory and even chaotic asymptotic states [5] based on a continuum model in the periphery (edge) diffusion regime that accounted for edge diffusional anisotropy [5–7]. The continuum model results were consistent with oscillatory dynamics in the morphological evolution of such homoepitaxial islands that has been demonstrated by atomic-scale KMC simulations on Cu(001) surfaces [8].

Experimental studies of electromigration-driven dynamics of islands on surfaces [9] have been sparse. A recent experimental study [10] based on *in situ* scanning tunneling microscopy (STM) examined the effects of thermally excited defects on the current-biased displacement of single-layer Ag islands on single crystalline Ag(111) substrates with island radii R_s over the range from 2 to 50 nm. The study found that the driving force for island drift is in the same direction as the electron flow acting on atomic defect sites on the island edge and established an island migration velocity varying inversely with the island radius, i.e., $v_m \propto 1/R_s$ [10].

The purpose of this Letter is to develop and implement a fully nonlinear continuum model for the systematic analysis of the current-driven morphological evolution of single-layer epitaxial islands on crystalline elastic substrates. We have extended the model of Ref. [5] to include in the analysis the driven dynamics of coherently strained heteroepitaxial islands and validated the model by comparison of its predictions with the experimental data of Ref. [10]. We focus on the exploration of migration dynamics of morphologically stable steady islands over a broad range of misfit strains and kinetic parameters and find that the islands' driven translational speed v_m is inversely proportional to their size R_s up to a critical island size. For larger-than-critical sizes, the $v_m(1/R_s)$ relation becomes nonlinear and the island dynamics

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in the nonlinear regime is characterized by morphological transitions, Hopf-bifurcation transitions to oscillatory asymptotic states, and morphological instabilities. We derive a universal linear relationship that can describe this complex nonlinear behavior through rescaling v_m with an island morphological metric.

The model follows a two-dimensional (2D) approximation for the evolution of one-monolayer-thick (1-ML-thick) coherently strained epitaxial islands on elastically deformable crystalline substrates of infinite thickness. In this 2D representation, mass transport due to curvature-driven diffusion, stress-driven diffusion, and electromigration is limited only to the island boundary (edge), i.e., periphery diffusion dominates mass transport consistently with the experimental study of Ref. [10] and the atomic-scale computational study of Ref. [1]. According to the model, the normal velocity component at a point on the island boundary, v_n, is expressed using mass conservation through the continuity equation, $v_n = -\Omega\partial J_s/\partial s$ with J_s being the total mass flux along the island edge, as:

$$v_n = \frac{\partial}{\partial s}\left\{\frac{\Omega D_s(\theta,\varepsilon_m)}{k_B T}\left[\Omega(\tilde{\gamma}+\tilde{\gamma}_{el})\frac{\partial \kappa}{\partial s} - q_s^* E_s\right]\right\}, \quad (1)$$

where s is the arc length along the island edge. In Eq. (1), $D_s(\theta, \varepsilon_m) = D_{s,\max}(\varepsilon_m) f(\theta)$ is the anisotropic diffusivity for edge diffusion, with $f(\theta)$ being the anisotropy function and ε_m the misfit strain ($\varepsilon_m = 0$ and $\varepsilon_m \neq 0$ for homoepitaxial and heteroepitaxial islands, respectively), Ω is the atomic area (in 2D), k_B is Boltzmann's constant, T is the temperature, $\tilde{\gamma}$ is the island's edge free energy per unit length (edge stiffness), and q_s^* is the effective charge of an edge atom. $E_s = E_0 \cos\theta$ is the local component of the electric field tangent to the island's edge, with E_0 being the strength of the applied electric field that is aligned with the Cartesian x-axis; θ is the edge orientation angle formed between the local normal to the island edge and the Cartesian y-axis. $\Delta\mu = \Omega(\tilde{\gamma}+\tilde{\gamma}_{el})\kappa$ is the chemical potential difference of an edge atom from that of an atom on a straight edge of an unstrained island [11–13], where $\kappa = d\theta/ds$ is the local edge curvature, and $\tilde{\gamma}_{el} = 2Y\varepsilon_m^2\Omega$ is the elastic contribution to the edge stiffness [14,15] with Y being the island's elastic modulus; the edge gradient of $\Omega\tilde{\gamma}_{el}\kappa$ is the driving force for strain-induced edge diffusion. The edge stiffness $\tilde{\gamma}$ is assumed to be isotropic, meaning simply that this thermodynamic anisotropy is much weaker than the diffusional (kinetic) anisotropy expressed by the function $f(\theta)$, as is typically the case in fcc metals [16].

For epitaxial islands on surfaces of face-centered cubic (fcc) crystalline solids, we use the 3-parameter function $f(\theta) = \{1 + A\cos^2[m(\theta+\phi)]\}/(1+A) \leq 1$ [5]; A is a dimensionless parameter that expresses the strength of the anisotropy, ϕ is the misorientation angle formed between a fast edge diffusion direction and the direction of the externally applied field, and m is an integer parameter that expresses symmetry due to surface crystallographic orientation; $m = 1$, 2, and 3 corresponds to <110>-, <100>-, and <111>-oriented surfaces, respectively [16]. It should be mentioned that this anisotropy function, in conjunction with an isotropic surface stiffness, has led to simulation predictions of electromigration-driven void dynamics in fcc metallic thin films in excellent agreement with experimental measurements [16]. The edge adatom diffusion barrier depends linearly on the misfit strain ε_m, $E_b = E_b' + \varepsilon_m\alpha'$, and, hence, $D_{s,\max}(\varepsilon_m) = D_0 \exp[-(E_b' + \varepsilon_m\alpha')/k_BT] = D_{0,\max}\exp(-\varepsilon_m\alpha)$ [17,18], where $D_{0,\max} \equiv D_0\exp(-E_b'/k_BT)$ and $\alpha \equiv \alpha'/k_BT$.

Within this mass transport regime, we have simulated directly the driven morphological evolution of individual (isolated) single-layer epitaxial islands starting with an elliptical (perturbed circular) shape as the initial configuration [19], employing a well-tested front tracking method [16]. The results reported in this study, unless specified otherwise, are for diffusional anisotropy parameters $A = 10$ and $\phi = 0°$ and material properties typical of Ag; the characteristic length scale $l_E = (\tilde{\gamma}\Omega/|q_s^* E_0|)^{1/2}$ and the diffusional time scale $\left(\tau = l_E^4/\left[(D_{0,\max}/k_BT)\tilde{\gamma}\Omega^2\right]\right)$ are used as the units of length and time, respectively.

First, we simulated the current-driven motion of single-layer homoepitaxial islands under the conditions of the experimental study of Ref. [10] in order to carry out systematic comparisons and validate our model in terms of its predictive capabilities [20]. Fig. 1 shows a comparison of our simulation predictions with the experimental measurements of Ref. [10]. The predicted stable island morphology at steady state is compared with the experimental observation (STM image) in Fig. 1(a); this is a *moving island that translates in the electric-field direction at constant speed*. The measured migration speed, v_m, of such driven morphologically stable islands as a function of the island radius R_s is compared with our simulation predictions in Fig. 1(b). It should be pointed out that the STM image of Fig. 1(a) represents an instantaneous (including shape fluctuations) stable island configuration moving under the action of the electric field and does not reflect the equilibrium shape of the island. The comparisons demonstrate good quantitative agreement between our modeling predictions and the experimental findings of Ref. [10].

We conducted a systematic parametric study of current-driven motion of stable single-layer epitaxial islands on crystalline substrates of fcc metals. To quantify the effects of strain due to lattice mismatch on the island periphery diffusion, we used results of atomic-scale simulations reported in the literature [17,18]. Fig. 2 shows representative results from our computer simulations for the dependence of the island migration speed, v_m, on the island size, $R_s = \sqrt{A_{isl}}$ with A_{isl} being the island area, for current-driven epitaxial island motion on <110>-, <100>-, and <111>-oriented surfaces of single-crystalline fcc metals that leads

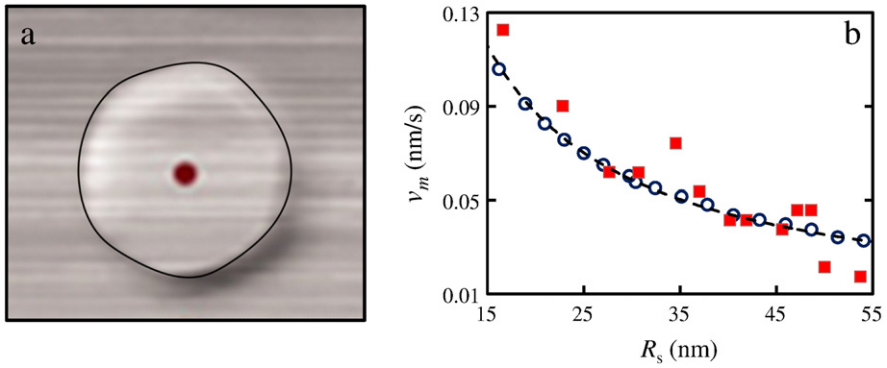

Fig. 1. Comparison of simulation predictions with experimental observations and data for current-driven migration of morphologically stable 1-ML-thick homoepitaxial islands on Ag(111). (a) Faceted (nearly hexagonal) island morphology. The gray shading corresponds to an STM image from Fig. 2 of Ref. [10] and the solid line is our simulation prediction of the stable island morphology under the experimental conditions. (b) Dependence of the current-driven island migration speed, v_m, on the island radius R_s under the conditions of Fig. 3C in Ref. [10]. Open circles and solid squares denote our simulation predictions and the experimental data of Fig. 3C in Ref. [10], respectively. The dashed curve is a least-squares fit to the simulation results according to the relationship $v_m \propto 1/R_s$.