Surface Science 669 (2018) 25–33

Contents lists available at ScienceDirect

Surface Science

journal homepage: www.elsevier.com/locate/susc

Analysis of current-driven oscillatory dynamics of single-layer homoepitaxial islands on crystalline conducting substrates

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ARTICLE INFO

Keywords:
Homoepitaxial islands
Surface electromigration
Edge morphological evolution
Nonlinear dynamics
Bifurcations
Shape oscillations

ABSTRACT

We report results of a systematic study on the complex oscillatory current-driven dynamics of single-layer homoepitaxial islands on crystalline substrate surfaces and the dependence of this driven dynamical behavior on important physical parameters, including island size, substrate surface orientation, and direction of externally applied electric field. The analysis is based on a nonlinear model of driven island edge morphological evolution that accounts for curvature-driven edge diffusion, edge electromigration, and edge diffusional anisotropy. Using a linear theory of island edge morphological stability, we calculate a critical island size at which the island's equilibrium edge shape becomes unstable, which sets a lower bound for the onset of time-periodic oscillatory dynamical response. Using direct dynamical simulations, we study the edge morphological dynamics of current-driven single-layer islands at larger-than-critical size, and determine the actual island size at which the migrating islands undergo a transition from steady to time-periodic asymptotic states through a subcritical Hopf bifurcation. At the highest symmetry of diffusional anisotropy examined, on {111} surfaces of face-centered cubic crystalline substrates, we find that more complex stable oscillatory states can be reached through period-doubling bifurcation at island sizes larger than those at the Hopf points. We characterize in detail the island morphology and dynamical response at the stable time-periodic asymptotic states, determine the range of stability of these oscillatory states terminated by island breakup, and explain the morphological features of the stable oscillating islands on the basis of linear stability theory.

1. Introduction

The controlled motion and dynamical behavior of atoms or atomic clusters adsorbed on crystalline substrate surfaces under the action of externally applied macroscopic forces is an important and intriguing area of study from the perspective of nanoscience and nanotechnology. Toward this end, obtaining a comprehensive fundamental understanding of the complex nonlinear dynamics of such surface adsorbates and their features driven by externally applied macroscopic forcing is of great significance. It has been demonstrated that atoms adsorbed on crystalline substrate surfaces, as well as collective surface dynamical behavior, can be driven in a controlled manner by electric fields or thermal gradients through the transport phenomena of electromigration [1–10] or thermomigration [11–14]. In general, the ability to drive mass transport and pattern formation on surfaces through use of macroscopic forces has been demonstrated at both mesoscopic and nanoscopic length scales in various material systems ranging from metal conductors [2] to colloids [15] and block copolymers [16–18] to liquid droplets [19]. These driven nonlinear dynamical phenomena have inspired recent studies to explore externally applied macroscopic forcing as a convenient and efficient tool to create nanopatterns on crystalline solid surfaces [20–24].

Among such macroscopic-force-driven assembly processes, the electromigration-driven assembly of crystalline conducting surface features, such as single-layer islands and voids, have been investigated extensively through both atomistic simulation and continuum-domain simulation studies [8–10,25,26]. Theoretical studies have examined various mechanisms of mass transport in single-layer epitaxial islands [27–32], with atomic transport through periphery or edge diffusion identified as the dominant of these diffusional transport processes. An important experimental study [2] based on *in situ* scanning tunneling microscopy has shown that such homoepitaxial islands on metallic substrates move in the direction of electron flow through edge atomic diffusion with a migration speed that is inversely proportional to the island size, R_s (where $R_s = \sqrt{\text{island area}}$). In a previous study on current-driven single-layer island dynamics on crystalline conducting substrates [25], we reported that there exist two critical island sizes, $R_{c,1}$ and $R_{c,2}$ with $R_{c,1} < R_{c,2}$, which set an island size range within which complex

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https://doi.org/10.1016/j.susc.2017.10.027
Received 30 August 2017; Received in revised form 27 October 2017; Accepted 28 October 2017
Available online 28 October 2017
0039-6028/

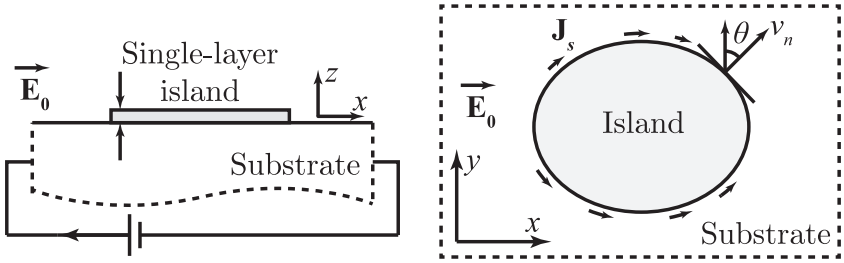

Fig. 1. Schematic representation of a single-layer epitaxial island with a rounded edge morphology on a crystalline conducting substrate under the action of an electric field. The $x - y$ plane of a Cartesian frame of reference corresponds to the substrate surface plane.

island edge morphological evolution occurs. Specifically, we found that, under the action of an externally applied electric field, islands with size $R_s < R_{c,1}$, migrate on the substrate surface retaining a stable rounded morphology with a migration speed that is linearly proportional to the inverse of the island size $1/R_s$. At island sizes $R_s > R_{c,1}$, the electric field, in conjunction with edge diffusional anisotropy, triggers a morphological instability on the edge of the migrating islands, resulting in formation of protrusions, facets, fingers, and necks on the island's edge. For sizes $R_s > R_{c,2}$, these protrusions, facets, and fingers grow and lead, through formation of thin necks, to the breaking up of the parent island into daughter islands. For island sizes between $R_{c,1}$ and $R_{c,2}$, the current-driven migrating islands can reach a time-periodic oscillatory state through a Hopf bifurcation transition [25]. The occurrence of such current-driven complex oscillatory single-layer island dynamics was first reported in the theoretical study of Ref. [8]. In spite of these interesting theoretical analyses, the intriguing oscillatory states reached by current-driven epitaxial islands have not been characterized fully and their range of complexity and stability have not been established.

In this article, we conduct a systematic study to identify, demonstrate, and characterize the complex oscillatory current-driven dynamics of single-layer homoepitaxial islands and the effects on this dynamics of important physical parameters, including island size, substrate surface orientation, and direction of externally applied electric field. The initial island morphology is shown in Fig. 1; this is a rounded morphology constituting a low-amplitude perturbation from the equilibrium (circular) morphology in the absence of electric field action. Using a linear theory of edge morphological stability [33–36] developed in Ref. [21], we determine the critical island size $R_{c,1}$ at which the island's rounded edge shape becomes unstable and, hence, bracket the size range over which the time-periodic oscillatory states could occur. We also examine the dependence of $R_{c,1}$ on the (misorientation) angle the electric field makes with the fast edge diffusion direction for {110}, {100}, and {111} face-center cubic (fcc) crystalline substrate surfaces. Next, using direct dynamical simulations, we study the edge morphological dynamics of current-driven single-layer islands with sizes R_s larger than that required to destabilize the rounded edge morphology and as large as required to cause island breakup, *i.e.,* $R_{c,1} < R_s < R_{c,2}$. We show that there exists a critical island size R_{HP} such that $R_{c,1} < R_{HP} < R_{c,2}$, at which the migrating islands undergo a transition from a steady to a time-periodic asymptotic state mediated by a subcritical Hopf bifurcation. We also determine the dependence of R_{HP} on the substrate surface orientation and the direction of the electric field with respect to fast edge diffusion directions. We characterize in detail the island morphology at the stable time-periodic asymptotic states and explain the morphological stability of these oscillating islands on the basis of linear stability theory.

The rest of this article is structured as follows: In Section 2, we give a brief description of our model of driven morphological evolution of single-layer epitaxial islands. In Section 3, the linear stability theory (LST) is presented for determining the critical island size $R_{c,1}$ for the onset of current-driven island edge instability and the dependence of $R_{c,1}$ on the direction of the applied electric field is predicted. In Section 4, we present the results of direct dynamical simulations, based on our model, of islands undergoing morphological oscillations and characterize their driven complex oscillatory dynamics. In Section 5, we explain through a linear theory of island edge stability the morphological features of stable islands undergoing edge shape oscillations. Finally, the conclusions of our study are summarized in Section 6.

2. Model of island dynamics

The initial configuration of a single-layer (*i.e.*, monolayer-thick) rounded epitaxial island is shown in Fig. 1 on a crystalline solid substrate surface that corresponds to the $x - y$ plane of a Cartesian frame of reference; as detailed in Section 3, the initial shape of the island's edge is a low-amplitude perturbation from a circular shape. The current-driven evolution of an island with such a morphology is monitored by time stepping the continuity equation, according to which the normal velocity, v_n, at any point on the island edge is proportional to the edge divergence of the mass flux along the island edge, J_s. The dominant mode of mass transport in such single-layer islands is edge diffusion [27–31]. Terrace diffusion and evaporation-condensation kinetics are neglected for the size of the metallic islands and the temperature range considered in this study (several tens of degrees above room temperature) [2,27–31]. The applied electric field that drives island electromigration [1–10] is aligned with the Cartesian x axis and has strength E_0. The flux J_s is expressed by a Nernst-Einstein equation [8,25] resulting in the continuity equation

$$v_n = -\Omega \frac{\partial}{\partial s}\left\{ \frac{D_s(\theta,\varepsilon_m)}{k_B T}\left[-\frac{\partial \mu}{\partial s} + q_s^* E_s \right] \right\}. \tag{1}$$

In Eq. (1), Ω is the atomic area, s is the arc length along the island's edge, $D_s(\theta, \varepsilon_m)$ is the edge atomic diffusivity, where θ is the edge orientation (Fig. 1) and ε_m is the misfit strain ($\varepsilon_m = 0$ and $\varepsilon_m \neq 0$ for homoepitaxial and heteroepitaxial islands, respectively), k_B is the Boltzmann constant, T is temperature, μ is the chemical potential of an edge atom, q_s^* is the effective charge of an atom at the island edge, and $q_s^* E_s$ is the local component of the electromigration force, tangent to the island's edge, as typically expressed in all phenomenological models of driven atomic transport due to electromigration [1–4,7,8]. The chemical potential of an edge atom is expressed as $\mu = (\tilde{\gamma} + \tilde{\gamma}_{\mathrm{el}})\Omega\kappa$, where $\tilde{\gamma}$ is the edge stiffness that is taken to be isotropic [8,25], $\tilde{\gamma}_{\mathrm{el}}$ is the elastic contribution to the island edge tension [25], and $\kappa = d\theta/ds$ is the local edge curvature. It should be emphasized that elastic effects are absent in homoepitaxial islands ($\varepsilon_m = 0$ and $\tilde{\gamma}_{\mathrm{el}} = 0$) and that isotropic edge stiffness implies a circular island morphology at equilibrium. The local approximation $E_s = E_0 \cos\theta$ [8] is justified by the 2D nature of the single-layer islands, which renders nonlocal effects, such as current crowding, negligible. The edge atomic diffusivity is taken to be anisotropic and expressed as $D_s(\theta, \varepsilon_m) = D_{s,\max}(\varepsilon_m) f(\theta)$, where $f(\theta)$ is the corresponding anisotropy function. From the dimensional analysis of Eq. (1), we derive the characteristic length scale l_E as $l_E = \sqrt{\tilde{\gamma}\Omega/|q_s^* E_0|}$ and the characteristic time scale τ as $\tau = l_E^4/[D_{s,\max}\tilde{\gamma}\Omega^2/(k_B T)]$. From the physical parameters reported in Ref. [2], we determine l_E and τ to be 13.6 nm and 32.5 s, respectively.