Surface Electromigration as a Moving Boundary Value Problem

M. Schimschak and J. Krug
Fachbereich Physik, Universität GH Essen, D-45117 Essen, Germany
(Received 6 September 1996)

Mass transport through surface electromigration strongly affects the stability and morphology of metal surfaces. Here the problem is treated within a continuum theory which takes full account of the nonlocal coupling between the electromagnetic bulk potential and the surface evolution. Key results of the numerical solution of the resulting moving boundary value problem are a scale-dependent drift of surface features and the absence of stable selected facet orientations. These effects cannot be reproduced within approximate local theories. [S0031-9007(96)02097-2]

PACS numbers: 66.30.Qa, 05.70.Ln, 68.35.Ja, 81.65.–b

Surface electromigration refers to the directed motion of adsorbed atoms (*adatoms*) on a solid surface which is caused by an electric current in the bulk of the material. It has long been recognized [1] that the associated mass transport can lead to pattern formation on a mesoscopic scale, through a mechanism related to the formation of ripples on wind blown sand [2]. A manifestation of recent interest is the current-induced step bunching observed on surfaces vicinal to Si(111) [3]. Electromigration along interfaces and grain boundaries is also believed to play a crucial role in the failure of metallic interconnects in integrated circuits [4–6].

Two different mechanisms are identified to be responsible for the coupling between the adatoms and the electric field. One is the direct electrostatic interaction, which requires that the adatoms are ionized and carry some effective charge. The other, the so-called "wind force," is due to the scattering of the electrons off the migrating adatoms and appears to be dominant at least for certain metals [7]. The wind force can also be characterized by an effective charge, leading to the same kind of force law as for the direct electrostatic interaction. This already gives a hint of the principal difficulty in describing surface electromigration on mesoscopic scales. The movement of the adatoms under the applied electric field induces a shape change of the specimen, which changes the electric field along the surface. It is this feedback between the surface evolution and the driving field which gives rise to a nonlocal *moving boundary value* problem.

In this Letter we present a selection of results from a numerical solution of the nonlocal continuum equations [8]. Our main purpose is to display the richness of intriguing phenomena associated with this type of surface dynamics, which is quite comparable to the extensively studied moving boundary problems of solidification [9]. A second goal is to delineate the validity of various *local* approximations which have been used in previous work [2,5,10,11].

Continuum theory.—We propose a continuum description of the surface evolution that takes into account electromigration and capillarity-driven surface diffusion *á la* Mullins [12]; desorption which might be important at high temperatures is neglected. Moreover we restrict ourselves to one-dimensional modulations which can be parametrized by a height function $h(x,t)$. The one-dimensional geometry is both convenient and directly relevant to the modeling of shape changes at the edge of an effectively two-dimensional conductor line [6,8]; the description through a single-valued height function, however, becomes inappropriate if the dynamics creates overhangs; see below. Volume conservation implies that h satisfies the continuity equation

$$\partial_t h + \partial_x J_s = 0\,, \tag{1}$$

where the mass current J_s along the surface is the product of the (orientation dependent) adatom mobility σ [2,10,13] and a driving force. In equilibrium the driving force is the derivative of the chemical potential with respect to the arclength. The external electric field contributes, as indicated above, a term qE_s where q is the effective charge density of the adatoms [14] and $E_s(x)$ the electric field along the surface. Recalling that the chemical potential is the product of the surface curvature and the surface stiffness $\tilde{\gamma}$, here assumed to be isotropic, we get the following expression for the current

$$J_s = \sigma(\partial_x h)\left(\frac{\tilde{\gamma}}{g^{1/2}}\partial_x \frac{\partial_x^2 h}{g^{3/2}} + qE_s(x)\right), \tag{2}$$

where $g = 1 + (\partial_x h)^2$. Dimensional analysis reveals $l_E = \sqrt{\tilde{\gamma}/|qE_0|}$ as characteristic length scale [5], where E_0 is a *typical* value of the electric field specified below. Assuming $qE_s > 0$ for simplicity and rescaling all lengths with $1/l_E$, time with $\sigma(0)\tilde{\gamma}/l_E^4$, the electric field with $1/E_0$ and the adatom mobility with $1/\sigma(0)$, we end up with the following dimensionless equation of motion:

$$\partial_{\tilde{t}}\tilde{h} = -\partial_{\tilde{x}}\left[\tilde{\sigma}(\partial_{\tilde{x}}\tilde{h})\left(\frac{1}{g^{1/2}}\,\partial_{\tilde{x}}\frac{\partial_{\tilde{x}}^2\tilde{h}}{g^{3/2}} + \tilde{E}_s(\tilde{x})\right)\right]. \tag{3}$$

The caret, which we will omit from now on, indicates scaled quantities.

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Next the local electric field $E_s(x)$ has to be specified. Since the time scale for electrodynamic processes is much shorter than that for diffusion processes involving adatoms, a quasistatic treatment of the electrodynamics is appropriate. Then the existence of an electric potential U in the bulk is a consequence of Maxwell's law $\mathbf{E} = 0$. Since the potential difference applied to the specimen is a constant of motion, the *typical* electric field mentioned above is just the negative potential difference divided by the length of the sample. Combining Ohm's law $\mathbf{J} \sim \mathbf{E}$ with the stationarity of the electric current, it follows that the potential has to obey Laplace's equation $\nabla^2 U = 0$ in the bulk with metallic boundary conditions on the surface; that is, the component of the electric field normal to the surface vanishes, while the tangential electric field which enters (3) is given by $E_s = -\partial_s U|_{\text{surface}}$. This shows that surface electromigration defines a new class of "Laplacian" interface dynamics, which differs from the familiar type [9] both in the boundary conditions for the Laplacian field U, and in the way the field couples to the interface motion.

Clearly the problem simplifies enormously if the electric field in (3) is replaced by some local function of h and its derivatives; however, there does not seem to be any systematic justification for such an approximation. Some aspects of void migration [5] can evidently be captured by assuming a field that is constant in the x direction, corresponding to $E_s(x) = E_0/\sqrt{g}$ [5,10,11]. We will instead compare our results to a local model in which the field is constant along the surface, $E_s = \text{const}$. This has the advantage of reproducing at least the linear stability properties of the full problem, which we address next.

Linear stability analysis.—We consider a metal slab of thickness δ in the h direction and infinitely extended in the x direction. One can think of the lower boundary as being attached to a flat substrate, whereas the upper boundary is the actual surface, subject to some sinusoidal disturbance $h(x,t) = \epsilon \exp(ikx + \omega t)$. The resulting electric field is then given by $E_s = [1 - \epsilon k \exp(ikx + \omega t)/\tanh(k\delta)]$, leading to the dispersion relation

$$\omega(k) = -k^4 + \sigma'(0)k^2 + i\frac{k^2}{\tanh(\delta k)}. \qquad (4)$$

The linear stability for small k is determined by the sign of the second term, i.e., the sign of $\sigma'(0)$ in relation to the direction of the electric force [2,10]. This also defines a characteristic length $l_i = 2\pi\sqrt{2/\sigma'(0)}\, l_E$ which determines the initial length scale of modulations in the case of linear instability $\sigma'(0) > 0$. The imaginary term gives rise to a drift of the surface modulations with velocity $v = k/\tanh(\delta k)$ in the direction opposite to that of the electric force [15]. While the real part of (4) is reproduced by the local approximation $E_s = \text{const}$, the imaginary part is intrinsically nonlocal. A drift can be induced in a local theory by setting $E_s \sim -h$; however, in that case the drift velocity is independent of k. Note also the dependence of the drift term on the sample thickness δ, which reflects the lack of translational invariance in the h direction for the nonlocal problem.

Numerical results.—In our numerical investigation of Eq. (3), we treat systems of lateral size L with periodic boundary conditions. We solve the Laplace equation in the domain shown in Fig. 1 by a boundary element method and extract the tangential electric field. The finite difference analog of (3) is then iterated by a variable-order, variable-step Adams method.

We assume an anisotropic adatom mobility of the form

$$\sigma(h') = \frac{1 + S\cos^2\{N[\arctan(h') + \varphi]\}}{1 + S\cos^2(N\varphi)}, \qquad (5)$$

where N is the number of symmetry axes and φ is the angle between a symmetry direction and the average surface orientation. S is a parameter determining the strength of the anisotropy. Throughout this paper we present results for $N = 3$ or 4 and $S = 1$. Orientations are chosen such that a flat surface would be linearly unstable, i.e., $-\pi/(2N) < \varphi < 0$.

Figure 2 shows the time evolution of a surface that is initially flat, with a small random perturbation. The number of symmetry axes N is chosen to be four. We compare the full problem (right) with the local approximation (left). Subsequent surface profiles have been displaced in the vertical direction in order to illustrate the evolution in time. At early times in both cases the instability sets in on the scale $l_i = 7.2$, followed by a coarsening regime in which the typical length scale of the modulation increases. The most striking difference between the two cases is due to the drift of surface structures in the nonlocal problem. One observes solitonlike features which follow curved trajectories, reflecting the fact that the drift velocity decreases with increasing wavelength [see (4)]. Occasionally this leads to collisions during which one of the features disappears. On a quantitative level the drift is therefore found to *speed up* the coarsening process. At time $t = 5000$, the feature wavelength in the nonlocal simulation exceeds that of the local approximation by a factor of two. The precise coarsening law is difficult

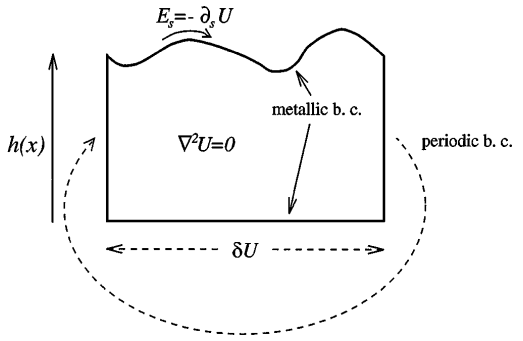

FIG. 1. Sketch of the domain used in the numerical solution of (3).

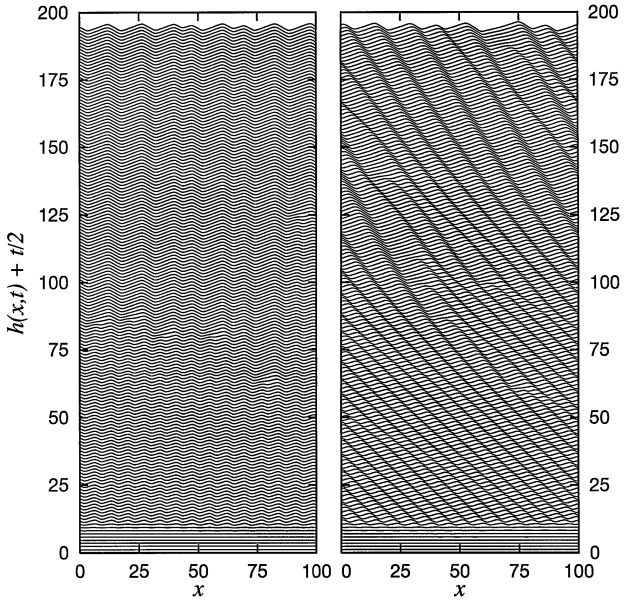

FIG. 2. Time evolution of systems with random initial surface profiles. Left: approximation with constant electric force along the surface; right: full problem. Subsequent surface profiles have been displaced by $t/2$ ($N = 4$, $\varphi = -\pi/32$, $S = 1$, and $l_i = 7.2$).

to ascertain, but it appears that the wavelength increases roughly logarithmically with time, as is typical in one-dimensional systems without fluctuations [16].

In the local theory the late stage morphology is characterized by well-defined, stable facets appearing at orientations which can be computed from the orientation dependence of the adatom mobility [2,10]. To investigate whether a similar morphology selection mechanism operates in the nonlocal case, we carried out a second set of simulations in which coarsening effects were avoided by starting with surface configurations that are already distorted on the scale of the system size. The equations of motion were then integrated until the surface width saturates, indicating that a steady state had been reached. For small initial distortions the systems exhibit the exponentially growing surface width and the drift velocity predicted by the linear stability analysis. Before the steady state is reached, we observe a crossover regime in which the width may, for certain orientations, increase *faster* than in the linear regime. The final steady state solution attains a constant drift velocity which does not seem to be simply related to the velocity in the linear regime.

Figure 3(a) shows scaling plots of late time profiles for various system sizes in comparison to those of the local approximation; Figure 3(b) shows the corresponding orientations $\theta(x) = \arctan(h') + \varphi$ of the surfaces. Here, the number of symmetry axes $N = 3$. For the local case the convergence to well-defined facet solutions, with facet orientations that are independent of the average tilt [2,10], is clearly observed. For the full problem no such convergence is found. First, as can be seen in Fig. 3(a),

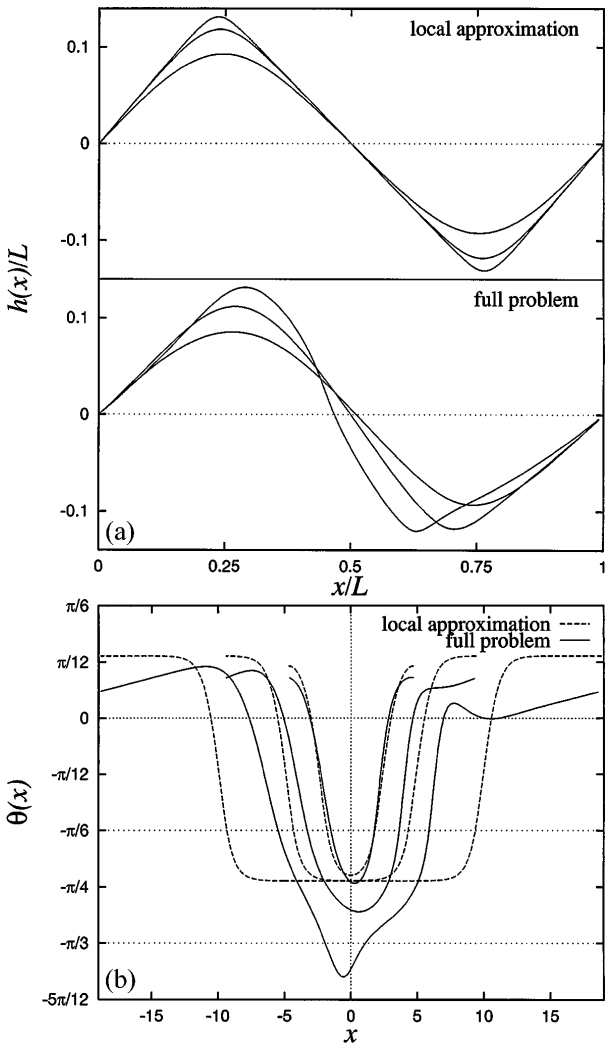

FIG. 3. Late time surface configurations. (a) shows scaling plots of surface profiles for the local (upper part) and nonlocal (lower part) problems, and (b) shows the corresponding surface orientations. The plots contain data for system sizes $L = 1.5l_i$, $3l_i$, and $6l_i$, with $N = 3$, $S = 1$, and $\varphi = -\pi/12$. All systems are in the steady state, except the largest one corresponding to the full problem, which is unstable. The dotted lines in (b) indicate orientations where the adatom mobility has an extremum; the line $\theta = -\pi/3$ indicates the limit of linear stability.

the $h \longrightarrow -h$ symmetry of the local theory is broken. Second, in Fig. 3(b) strong distortions of the local orientation appear near the minimum, which become more pronounced with increasing system size until the surface becomes unstable.

Observations such as these suggest the following scenario: An increase of the lateral length scale of surface modulations goes along with an increase in their amplitude, and therefore with a decrease of the electric field on the hills and an increase in the valleys. Consequently, the rate at which adatoms move from one hillside to the other at the bottom of the valleys increases. This gives

rise to a steepening of one hillside, which is apparent from Fig. 3(b). If the lateral length scale of the modulation becomes so large that the steep hillside reaches a linearly unstable orientation, which, in the case shown in Fig. 3(b), is the case for $\theta < -\pi/3$, the entire hill-valley structure becomes unstable. This manifests itself in diverging slopes. Since our algorithm applies so far only to surfaces that can be parametrized by a height function, we cannot investigate the behavior of the surface beyond this point; however, the formation of overhangs and slitlike voids, as observed in [8], seems very likely. For $N = 3$ the same type of instability was also observed in simulations of large systems, as in Fig. 2, at long times ($t \approx 1500$). For $N = 4$ the surfaces show the same tendency to steepen one hillside as described in the above scenario, but so far we cannot confirm that they, in the end, actually become unstable.

We note in this context that in the local approximation with $E_s(x) = E_0/\sqrt{g}$ the facet selection mechanism was found to fail under certain conditions [10]; presumably also in this case the breakdown reflects a tendency of the morphology to develop overhangs and, possibly, slitlike solutions [5].

In conclusion, we have demonstrated that the inclusion of the nonlocal feedback between surface evolution and bulk current qualitatively changes the electromigration-induced instability of a metal surface. The most intriguing result of our investigation is that the surface, under some circumstances, develops overhangs, which could subsequently evolve into voids or slits. To further address this issue, our algorithm needs to be extended to allow for arbitrary surface conformations.

We wish to thank the IFF, Forschungszentrum Jülich, where most of this work was done, for its hospitality, and Bill Mullins for helpful conversations. Partial support by DFG within SFB 237 *Unordnung und grosse Fluktuationen* is gratefully acknowledged.

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