# Ratchet Effect in Surface Electromigration: Smoothing Surfaces by an ac Field

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We demonstrate that for surfaces that have a nonzero Schwoebel barrier the application of an ac field parallel to the surface induces a net electromigration current that points in the descending step direction. The magnitude of the current is calculated analytically and compared with Monte Carlo simulations. Since a downhill current smoothes the surface, our results imply that the application of ac fields can aid the smoothing process during annealing and can slow or eliminate the Schwoebel-barrier-induced mound formation during growth. [S0031-9007(97)05220-4]

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Growing epitaxial films with smooth surfaces is one of the ongoing challenges of the thin film community. However, this goal is hampered by a series of basic physical effects that lead to the development of unavoidable surface roughness during growth. In particular, there is abundant experimental and theoretical evidence that during deposition the diffusion bias generated by the Schwoebel barrier (see Fig. 1) results in a net *uphill current*, which in turn leads to the formation of mounds and to a fast and unwanted increase in the interface roughness [1]. As Fig. 1 demonstrates, the Schwoebel barrier introduces spatial asymmetry in the otherwise symmetric lattice potential. Interestingly, in recent years it has been recognized that in such periodic and spatially asymmetric systems (ratchets) nonequilibrium fluctuations can *rectify* Brownian motion and induce a nonzero net current [2]. This *fluctuation driven transport* is believed to play an essential role in the operation of motor proteins or molecular motors, and might result in new separation techniques [3]. In this paper we propose the first nanoscale application of this transport mechanism based on the atomic electromigration on vicinal surfaces induced by alternating electric fields. We demonstrate that the Schwoebel barrier induced asymmetry in the lattice potential can be used to generate a *downhill current,* aiding the smoothing of surfaces during growth or annealing.

Atom diffusion on crystal surfaces is a thermally activated process: atoms can hop from their position to a neighboring one by overcoming a potential barrier $\Delta E$. The hopping rate is given by the Arrhenius law $k = \nu_0 \exp(-\Delta E/k_\mathrm{B}T)$, where $T$ is the temperature and $\nu_0$ is the vibration frequency of the surface atoms. Figure 1 illustrates the lattice potential of a vicinal surface that consists of long flat terraces separated by monatomic steps. The barrier height for diffusion on a flat surface is denoted by $E_0$. Near a step atoms form additional lateral bonds of energy $E_1$ with the step atoms, leading to a deeper potential valley. Finally, jumping over a step requires passing an additional potential barrier, the Schwoebel barrier, $E_\mathrm{b}$ [4].

For most metals and semiconductors the otherwise random surface diffusion of the atoms can be biased by an external electric field applied parallel to the surface, a phenomenon known as surface electromigration [5]. The effective force, $\boldsymbol{F}$, acting on the surface atoms is proportional to the field $\boldsymbol{E}$, $\boldsymbol{F} = Ze\boldsymbol{E}$, where $e$ ($> 0$) is the elementary charge and $Z$ is the effective charge number which consists of two terms, $Z = Z_\mathrm{d} + Z_\mathrm{w}$. The

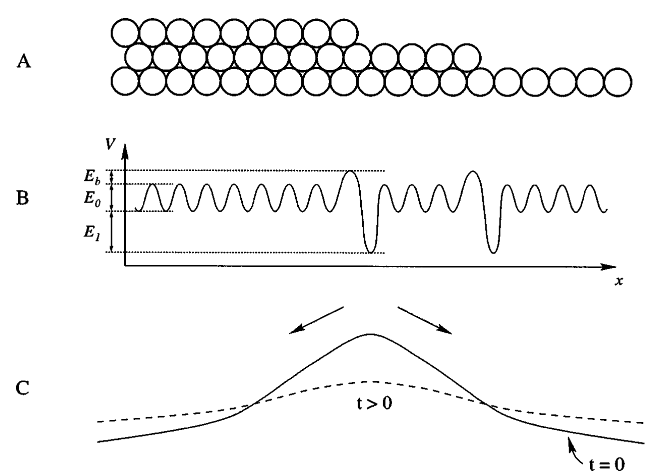


FIG. 1. Schematic illustration of the cross section of a vicinal surface containing two monatomic steps (A) and the asymmetric lattice potential (B) experienced by an atom diffusing on this surface. Note that if the Schwoebel barrier $E_b = 0$, the lattice potential near a step is spatially symmetric, while this symmetry is broken for $E_b \neq 0$. A downhill (uphill) current points to the right (left) on this figure. As (C) illustrates, if there is a mound on the surface at $t = 0$, a downhill current indicated by the arrows will tend to decrease its height; i.e., it smoothes the film by the reorganization of the material on the surface.

  

"direct" term $Z_d$ ($> 0$) is associated with the electrostatic interaction between the atom and the electric field, while the "wind" term $Z_w$ ($< 0$) is generated by the scattering of the current carrying electrons on the surface atoms. The competition between these two terms can result in either positive or negative effective charge [6].

The constant electric field induces a surface current parallel to the field. However, it is expected that a *periodic field* with zero mean does *not* create a net current over a full period, since it simply amounts to biasing in an equal manner the motion of the atoms in two opposing directions. In contrast, next we demonstrate that for systems that have a nonzero Schwoebel barrier the asymmetry induced by the barrier rectifies the diffusion process, generating a net current along the surface, even if the external field has a zero mean value. Most important, the induced current is *always downhill;* i.e., it points towards the descending step direction, independent of the step orientation or the effective charge. Since the downhill current acts to smooth the surface, it has the potential to accelerate the smoothing process during annealing and to slow or eliminate the Schwoebel barrier induced mound formation during growth. Consequently, this nanoscale ratchet effect can have important technological applications for thin film growth.

Consider a vicinal semiconductor surface with terraces of equal width $w$ in an alternating external electric field (with zero mean) perpendicular to the steps and parallel to the surface. The lattice potential of this system is periodic and *spatially asymmetric*, and the alternating field acts as a zero-mean nonequilibrium fluctuating force. In the presence of such field (or ac current) a directed net flow of the diffusing atoms on the surface is expected from the theory of fluctuation driven transport [2]. A simple explanation of the effect responsible for this current is given in Fig. 2. To evaluate the net current we first consider the motion of a *single* atom on a *fixed* vicinal surface with terraces of width $w$. Since the diffusion parallel to the steps is not affected by the electric field we can neglect the transverse direction and consider the motion in one dimension, perpendicular to the steps. The one-dimensional lattice potential for the diffusing atom is similar to the potential of Fig. 1, but it is periodic with period $w$, each period containing $\ell$ valleys and barriers, where $\ell = w/a$ is the dimensionless size of the terraces measured in the units of the lattice constant $a$. Associated with the steps, every $(\ell - 1)$th $E_0$ barrier is followed by a higher Schwoebel barrier $E_b$ and a deeper valley $E_1$ representing binding to the step. The system can be reduced to one period of the potential with periodic boundary conditions, in which the motion of the atom is described by the master equation

$$\frac{\partial P_i(t)}{\partial t} = k_{i-1}^{+}(t)P_{i-1}(t) + k_{i+1}^{-}(t)P_{i+1}(t) - [k_i^{+}(t) + k_i^{-}(t)]P_i(t)\,, \qquad (1)$$

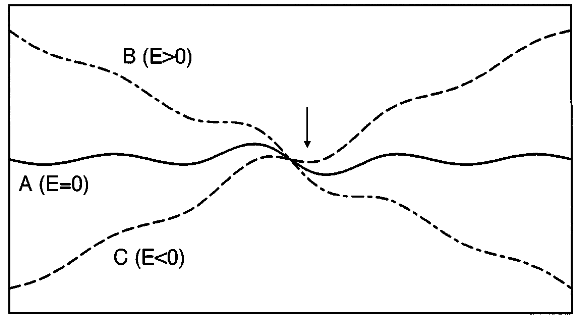


FIG. 2. The effect of a uniform electric field on the potential landscape experienced by an atom diffusing on the surface. The line $A$ corresponds to the lattice potential in the vicinity of a step for $E = 0$ [see also Fig. 1(B)]. Assuming $Z > 0$, a large enough field to the right ($E > 0$) "tilts" the potential landscape, eliminating the minima of the potential, biasing the adatom motion to the right (see $B$). On the other hand, as $C$ illustrates, an inverse field ($E < 0$) of the same magnitude does not eliminate the minima in the vicinity of the step (marked by an arrow), locally trapping the particle. Consequently, the current to the left in $C$ is smaller than the current to the right in $B$, and a periodic external field will result in a net current to the right, i.e., a downhill current. For smaller fields there are minima in both cases, but the minimum corresponding to $C$ is deeper than that of $B$, again resulting in a more efficient trapping of the atoms moving uphill, and to a net downhill current. Note that the direction of the net current is determined only by the step orientation, and not by the sign of $Z$.

where $1 \leq i \leq \ell$ with periodic boundary conditions, $P_i(t)$ denotes the probability that the atom is situated in the $i$th valley at time $t$. $k_i^-$ and $k_i^+$ denote the hopping rates from the $i$th valley to the left and to the right, respectively, where $k_i^-(t) = \nu_0 e^{-[\Delta E_i^- + \delta E(t)]/k_B T}$, and $k_i^+(t) = \nu_0 e^{-[\Delta E_i^+ - \delta E(t)]/k_B T}$, $\Delta E_1^- = E_0$, $\Delta E_1^+ = E_0 + E_b$, $\Delta E_2^- = E_0 + E_1 + E_b$, $\Delta E_2^+ = E_0 + E_1$, and $\Delta E_j^- = \Delta E_j^+ = E_0$ for $3 \leq j \leq \ell$ are the heights of the barriers of the lattice potential. Finally, $\delta E(t) = F(t)a/2$ is the change in the barrier heights associated with the force $F(t) = ZeE(t)$ induced by the electric field $E(t)$.

If the period of the electric field is much larger than the relaxation time $\tau \approx \ell^2 \exp(E_0/k_B T)/\nu_0$ of the probability distribution $P_i(t)$, $P_i(t)$ is "slaved" by the electric field and the solution of the master equation reduces to the stationary solution with constant electric field $E$. If the period of the field is much smaller than $\tau$, $P_i(t)$ has no time to adjust to the field, resulting in a very small net current. Thus $f_u = 1/\tau$ is the upper limit of the frequency, since exceeding it the current goes to zero. Therefore, in the following we assume that the frequency $f$ of the alternating field is below the upper limit $f_u$, so that the stationary solution of the master equation applies. In this case Eq. (1) reduces to a system of $(\ell - 1)$ independent linear equations with the normalization condition $\sum_{i=1}^{\ell} P_i = 1$, giving the $\ell$th equation. The solution provides the stationary current as

$$J_{\rm st}(F) = k_1^+(F)P_1(F) - k_2^-(F)P_2(F)$$
$$= \frac{2\sinh(\ell\delta E/k_{\rm B}T)\,\nu_0 e^{-E_0/k_{\rm B}T}}{(e^{E_b/k_{\rm B}T}-1)(e^{E_1/k_{\rm B}T}-1)e^{(1-\ell)\delta E/k_{\rm B}T} + [\ell + (e^{E_b/k_{\rm B}T}-1) + (e^{E_1/k_{\rm B}T}-1)]\frac{\sinh(\ell\delta E/k_{\rm B}T)}{\sinh(\delta E/k_{\rm B}T)}}. \quad (2)$$

For simplicity we restrict the calculation to the case when the field is as a symmetric square wave; i.e., the force alternates between $+F$ and $-F$ at constant intervals. In this case, when $f \ll f_u$, the net current $J_0$ can be calculated from (2) as $J_0 = [J_{\rm st}(+F) + J_{\rm st}(-F)]/2$.

If the interaction between the diffusing atoms is neglected, the average number of atoms that are able to move (i.e., that are not part of a terrace) is $1 + N_a$ in each period. The first term indicates the edge of the terrace, and the second term, $N_a = (\ell - 1)/[1 + \exp(E_1/k_{\rm B}T)]$, is the average number of diffusing atoms (adatoms) on the surface. Thus the net particle current is

$$J = J_0(F)(1 + N_a) = J_0(F)\left(1 + \frac{\ell - 1}{1 + e^{E_1/k_{\rm B}T}}\right). \quad (3)$$

Equation (3) provides the downhill current generated by the interplay between the ac field and the Schwoebel barrier. The terrace size dependence of this current for two different temperatures is shown in Fig. 3(A).

The previous calculation, while correctly describing the nature and the qualitative features of the net current, neglects the atom-atom interaction and the step fluctuations. Since the source of atoms are the steps (adatoms detach from step edges), the step length is not fixed, but it fluctuates. To incorporate these effects and to check the validity of the analytical prediction, we performed Monte Carlo (MC) simulations with activated diffusion along the surface. In the simulations we start with a series of steps of length $\ell$. Every surface atom that has less than two neighbors in the same layer is allowed to diffuse with a probability $P \sim \exp(-\Delta E/k_BT)$. In the presence of an electric field $\delta E$ the activation energy is reduced or increased with $\delta E$, depending on whether the direction of the hop is along or against the field. In the simulations we use a periodic square wave with frequency $f \ll f_u$, and determine the net current defined as the number of excess hops in the downhill direction in the unit time, normalized to the system size.

As Fig. 3(A) indicates, the net current obtained in the Monte Carlo simulation qualitatively agrees with the prediction (3) of the master equation. Furthermore, there is excellent quantitative agreement at low temperature (900 K) while (3) overestimates the current at higher $T$. Note that the fit does not require any fitting parameter.

To pinpoint the temperature range for which the analytical solution provides a good approximation, in Fig. 3(B) we plot the temperature dependence of the net current for $\ell = 20$ and $\ell = 200$. We find excellent agreement between the theory and the MC simulations for $T < T^*$ ($\simeq$1000 K), while for $T > T^*$ the current predicted by (3) systematically exceeds the numerical result. Indeed, increasing the temperature also increases the adatom density on the surface, and consequently the role of the neglected atom-atom interactions also increases [7]. The value of $T^*$ depends only on the energy barriers $E_0$, $E_1$, and $E_b$. In some materials the bonding energies might be higher than the quoted values, which further increases $T^*$ and the range of applicability of the analytical solution. Note that the agreement between the analytical predictions and the MC simulations is better for $\ell = 200$ than $\ell = 20$, underlying the larger impact of step fluctuation on shorter steps [8].

Since in the typical electromigration experiments [5] $\delta E$ is much smaller than $k_BT$, we can expand the current

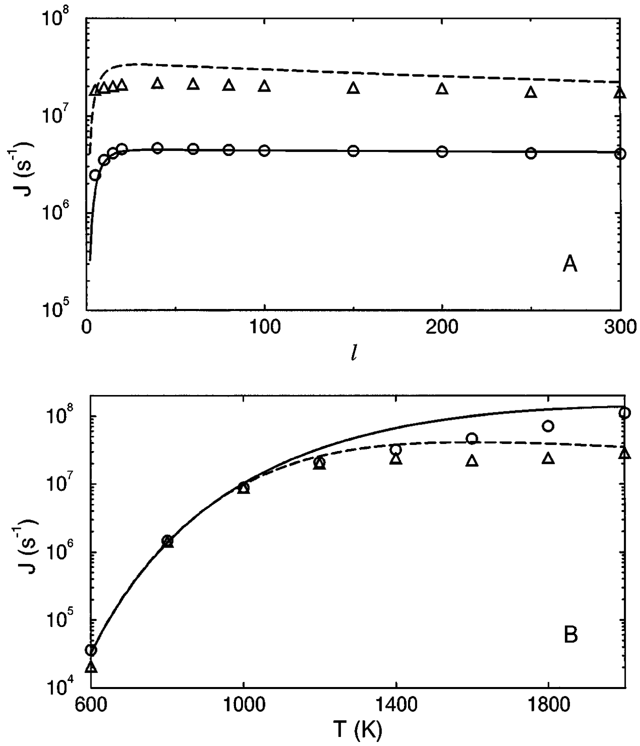


FIG. 3. (A) The ac field induced downhill current as a function if the average step size $\ell$. The lines correspond to the analytical results (2) and (3) for $T = 900$ K (continuous line) and $T = 1200$ K (dashed line), while the circles (900 K) and triangles (1200 K) are the currents predicted by the MC simulations. (B) The temperature dependence of the net current for average step sizes $\ell = 20$ and $\ell = 200$. The continuous ($\ell = 20$) and dashed ($\ell = 200$) lines correspond to the analytical predictions, while the circles ($\ell = 20$) and triangles ($\ell = 200$) are the result of the MC simulations. In the simulations we used the parameters $E_0 = 0.3$ eV, $E_1 = 0.6$ eV, $E_b = 0.15$ eV, $Z = 0.5$, $E = 10^8$ V/m, and $\nu_0 = 10^{13}$ s$^{-1}$. The system size is $L = 2000$, and the results were averaged over 20 independent runs.

$J_0(F)$ into Taylor series in terms of $(\delta E/k_B T)$, obtaining that the net current is a second order effect, proportional to $(Z\delta E)^2$. The MC simulations confirm this prediction, providing a quantitative expression for tuning the current with $E$ [9].

In conclusion, we have demonstrated both analytically and numerically that the Schwoebel barrier, in the presence of a periodic external electric field, leads to a downhill current. Since most metal and semiconductor surfaces do have a nonzero Schwoebel barrier and display electromigration, we expect that the appearance of such a net current is relevant for a large class of technologically important materials. Thus the application of an ac current during either growth or annealing can lead to a nontrivial smoothing effect, and aid the growth of smooth surfaces. This consequence of the ratchet effect can thus have important practical applications in the growth and processing of high quality thin films.

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[8] When a small step emits (absorbs) an atom, its relative change in size will be much larger than that of a larger step. Thus one expects a better agreement between the theory and the simulation for large $\ell$. On the other hand, fluctuation in the step sizes for small $\ell$ are smaller due to step-step interaction, balancing this effect.

[9] In order to amplify the effect in the time span allowed by the MC simulation, in Fig. 3 we used $ZE = 5 \times 10^7$ V/m, larger than the typical values used in current experiments [5], that are of order $10^5$ V/m. However, using the expression for the field dependence, we can calculate the current for arbitrary electric fields.