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journal or publication title	Physical Review Letters
volume	80
number	19
page range	4233-4266
year	1998-01-01
URL	http://hdl.handle.net/2297/25292

doi: 10.1103/PhysRevLett.80.4233

Control of Chaotic Wandering of an Isolated Step by the Drift of Adatoms

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(Received 30 October 1997)

The drift of adatoms strongly influences the wandering pattern of an isolated step moving in a surface diffusion field. When the drift velocity has a component against the step motion and exceeds a critical value, the straight step becomes unstable with long wavelength fluctuations, and wanders. This wandering pattern can be controlled by changing the direction of the drift. When the drift has no component parallel to the step edge, the unstable step obeys the Kuramoto-Sivashinsky equation and shows a chaotic pattern. When the drift has a component parallel to the step edge, the step obeys the Benney equation. If the parallel component is sufficiently large, the step shows a regular pattern. [S0031-9007(98)05966-3]

PACS numbers: 81.10.Aj, 05.70.Ln, 47.20.Hw, 68.35.Fx

To provide a perfectly oriented surface on a crystal, molecular beam epitaxy (MBE) and other similar methods are widely used. In these methods, atoms adsorbed on the crystal surface diffuse on it and are incorporated into atomic steps. For a vicinal face which consists of equidistant straight steps, the step-flow mode is expected to produce a perfectly oriented interface when the steps move uniformly. However, kinetic instabilities of the step morphology interfere with this ideal behavior. Steps wander wildly or they bunch to form macrosteps, and the uniformity of the surface will be deteriorated.

The wandering (or meandering) [1] is a step instability in response to a fluctuation along the step edge, and steps cannot remain straight. They deform chaotically in space and time [2]. The instability exists even for an isolated step and is caused by an asymmetry in the surface diffusion field of adsorbed atoms (adatoms): The net rate of adatom incorporation in the step from the upper terrace is different from that of the lower terrace. Contrary to the wandering, the bunching [3] is an instability perpendicular to the step edge, and the step separation or the step density along the vicinality cannot remain homogeneous. The surface takes the form of a regular array of pulses or macrosteps [4]. This is also caused by the asymmetry in the surface diffusion field [5]. The strong contrast between the chaotic morphology under the wandering instability and the periodic pulses under the bunching instability is caused by the symmetry difference along the step edge and orthogonal to it. The direction to the right along the step is equivalent to the left, and the parity symmetry exists along the step edge. On the contrary, the direction forward from the step is different from that backward, and there is no inversion symmetry orthogonal to the steps.

The asymmetry in the diffusion field results either from the energy barrier suppressing the interlayer transport (the Ehrlich-Schwoebel effect [6–10]) or from the drift of adatoms due to the external field (for instance, the electromigration [11,12]). In Si(111), a direct electric current is

shown to induce the bunching instability [13,14], and this is attributed to the drift of adatoms perpendicular to the steps [3,15–19]. The adatoms encounter a force $\mathbf{f}$ proportional to the external electric field $\mathbf{E}$ as [20] $\mathbf{f} = Ze\mathbf{E}$ (Z is the effective valence of an adatom), and drift on the surface with a drift velocity $\mathbf{v} = (D_s Ze/k_B T)\mathbf{E}$ with the diffusion constant D_s at the temperature T . The same electromigration effect is theoretically shown to cause a wandering instability and chaotic behavior for an isolated step [21]. The advantage of electromigration is its ability to alter the direction of the drift and then the parity symmetry of the system by varying the orientation of the external field. This in principle leads to control over the behavior and the profile of an unstable step. The purpose of this paper is to explicitly show an ability of the external field in controlling the morphological transition of steps from a chaotic to an orderly one.

We use the modified version [3,15–19,21] of the standard step-flow model [22] to study the effect of adatom drift on the step morphology. While adatoms diffuse on the surface, they drift with a constant velocity $\mathbf{v} = (v_x, v_y)$ due to the external direct electric field. Simultaneously, they also evaporate at a rate of $1/\tau$. We first examine the case where the deposition of atoms from the ambient vapor phase is neglected. The situation has been extensively studied in Si [13]. The diffusion equation of the adatom density $c(\mathbf{r}, t)$ is then written as

$$\frac{\partial c(\mathbf{r}, t)}{\partial t} = D_s \nabla^2 c(\mathbf{r}, t) - \mathbf{v} \cdot \nabla c(\mathbf{r}, t) - \frac{1}{\tau} c(\mathbf{r}, t). \quad (1)$$

For simplicity of presentation, we consider the stability of an isolated step. The step runs on average in x direction, and its local configuration is denoted by $y = \zeta(x, t)$. Because of evaporation, it recedes the step-up, i.e., $-y$ direction. Further from the step, the density of adatoms released from the receding step vanishes: $c(\mathbf{r}, t) \rightarrow 0$ at $y \rightarrow \pm\infty$. We assume that adatom incorporation at the step from the upper and the lower terraces is so

fast that the local equilibrium is realized at the step edge [1,2,21]: $c|_{y=\zeta} = c_{\text{eq}}^0(1 - \Gamma\kappa)$. Here c_{eq}^0 is the equilibrium adatom density of a straight step, Γ is the capillary length [1] given by $\Gamma \equiv \Omega\tilde{\beta}/k_B T$ (Ω : atomic area, $\tilde{\beta}$: step stiffness), and κ is the curvature. In solving Eq. (1) we use the stationary approximation, $\partial c(\mathbf{r}, t)/\partial t = 0$, which is valid for a slow step motion. The mass conservation at the step implies that the normal step velocity obeys the relation

$$\frac{\partial \zeta / \partial t}{\sqrt{1 + (\partial \zeta / \partial x)^2}} = \Omega[(D_s \nabla c - \mathbf{v}c) \cdot \hat{\mathbf{n}}]_{\zeta+0} - \Omega[(D_s \nabla c - \mathbf{v}c) \cdot \hat{\mathbf{n}}]_{\zeta-0}, \quad (2)$$

where $\hat{\mathbf{n}}$ is the unit vector normal to the step and is directed to the lower terrace. Hereafter in the analytical calculation, we measure time and space in units of τ and the surface diffusion length $x_s = \sqrt{D_s \tau}$, respectively, and denote a dimensionless variable A as $\tilde{A}$.

We first investigate the linear stability of a straight step moving in the y direction. When the step is straight, the adatom density depends only on the y coordinate, and the velocity is determined as $\tilde{V}_0 = -\Omega c_{\text{eq}}^0(4 + \tilde{v}_y^2)^{1/2}$. If there is a small sinusoidal perturbation of the step profile, $\tilde{\zeta}(\tilde{x}, \tilde{t}) = \tilde{V}_0 \tilde{t} + \tilde{\zeta}_1 e^{i\tilde{q}\tilde{x} + \tilde{\omega}_{\tilde{q}}\tilde{t}}$, the adatom density responds with a modulation with the same wave number $\tilde{q}$ as $c(\tilde{x}, \tilde{y}, \tilde{t}) = c_0(\tilde{y}) + c_1(\tilde{y})e^{i\tilde{q}\tilde{x} + \tilde{\omega}_{\tilde{q}}\tilde{t}}$. The linear dispersion relation is obtained from Eq. (2) as

$$\begin{aligned} \frac{\tilde{\omega}_{\tilde{q}}}{\Omega c_{\text{eq}}^0} &= -\tilde{v}_y \sqrt{1 + \frac{\tilde{v}_y^2}{4}} \\ &\quad + (\tilde{v}_y - 2\tilde{\Gamma}\tilde{q}^2) \sqrt{1 + \frac{\tilde{v}_y^2}{4} + \tilde{q}^2 + i\tilde{q}\tilde{v}_x} \\ &\approx i \frac{\tilde{v}_x \tilde{v}_y}{2} \tilde{q} - i \frac{\tilde{v}_x}{4} (\tilde{v}_y + 4\tilde{\Gamma}) \tilde{q}^3 \\ &\quad + \frac{1}{2} (\tilde{v}_y - 4\tilde{\Gamma}) \tilde{q}^2 - \frac{1}{8} (\tilde{v}_y + 8\tilde{\Gamma}) \tilde{q}^4. \end{aligned} \quad (3)$$

The approximation holds, if the wavelength of the perturbation is longer than the surface diffusion length, $\tilde{q} \ll 1$ or $q \ll x_s^{-1}$, and the drift velocity is so small that terms of order $\tilde{v}_y^2$ are negligible compared to unity. When the drift velocity perpendicular to the step $\tilde{v}_y$ exceeds the critical value $\tilde{v}_c = 4\tilde{\Gamma}$, the straight step becomes unstable against the long wavelength fluctuation, as signaled by the positive coefficient of the $\tilde{q}^2$ term. The wave number of the fastest growing mode is determined by the real part of $\tilde{\omega}_{\tilde{q}}$ as $\tilde{q}_{\text{max}} = [2(\tilde{v}_y - 4\tilde{\Gamma})/(\tilde{v}_y + 8\tilde{\Gamma})]^{1/2}$, and is small near the critical velocity. The wave pattern shifts along the step edge with the velocity $\tilde{V}_x = \Omega c_{\text{eq}}^0 \tilde{v}_x [-\tilde{v}_y/2 + (\tilde{v}_y + 4\tilde{\Gamma})\tilde{q}_{\text{max}}^2/4]$. The characteristic feature of $\tilde{V}_x$ is its proportionality to the drift velocity of adatoms in the x direction, $\tilde{v}_x$. Near the threshold of instability, the step pattern shifts in the opposite direction of the adatom drift.

To predict the step behavior as the fluctuation grows, we must take nonlinear effects into account. The nonlinear evolution equation near the threshold of the instability can be derived formally with the reductive perturbation method [2,21]. Here we give an intuitive explanation of its derivation. The linear part of the evolution equation is obtained from Eq. (3) by replacing $\tilde{\omega}_{\tilde{q}}$ and $i\tilde{q}$ with $\partial/\partial \tilde{t}$ and $\partial/\partial \tilde{x}$. The translational symmetry in the y direction implies that nonlinear terms significant in the long wavelength instability are $\tilde{\zeta}_{\tilde{x}}^2$, $\tilde{\zeta}_{\tilde{x}}\tilde{\zeta}_{\tilde{x}\tilde{x}}$, $\tilde{\zeta}_{\tilde{x}\tilde{x}}^2$, To find the most important term, we introduce the distance from the critical velocity as a small parameter; $\epsilon \equiv 4(\tilde{v}_y - \tilde{v}_c)/3\tilde{v}_y$. Since $\tilde{q}_{\text{max}}$ is proportional to $\sqrt{\epsilon}$, the most important nonlinear term should be the lowest in the x derivatives, and it has the form $\gamma \tilde{\zeta}_{\tilde{x}}^2$. The coefficient γ is determined from a simple consideration of the velocity of a straight step tilted from the x axis [23]. With a tilting angle θ the step position is expressed as $\tilde{\zeta} = \tilde{x} \tan \theta + \tilde{V}_y(\theta)\tilde{t}$. By combining the velocity of the linear analysis and the nonlinear term $\gamma \tilde{\zeta}_{\tilde{x}}^2$, the step velocity in the y direction $\tilde{V}_y(\theta)$ is expressed as $\tilde{V}_y(\theta) = \tilde{V}_0 + (\Omega c_{\text{eq}}^0 \tilde{v}_x \tilde{v}_y \tan \theta)/2 + \gamma \tan^2 \theta$ [the second term corresponds to the linear term of Eq. (3)]. On the other hand, the rotation of the coordinate system gives the velocity of a straight step as $\tilde{V}_y(\theta) = -(\Omega c_{\text{eq}}^0 / \cos \theta) \sqrt{4 + (\tilde{v}_y \cos \theta - \tilde{v}_x \sin \theta)^2}$. By comparing the coefficients of the θ^2 terms in both equations, γ is determined as $\gamma = -\Omega c_{\text{eq}}^0$, which is correct up to the first order in $\tilde{v}_x$ and $\tilde{v}_y$ [the accurate expression is $\gamma = -\Omega c_{\text{eq}}^0(1 + \tilde{v}_x^2 + \tilde{v}_y^2)(1 + \tilde{v}_y^2)^{-3/2}$]. In a scaled moving frame, $X \equiv \sqrt{\epsilon}[\tilde{x} + \Omega c_{\text{eq}}^0 \tilde{v}_x \tilde{v}_y \tilde{t}/2]$, $T \equiv 3\epsilon^2 \Omega c_{\text{eq}}^0 \tilde{\Gamma} \tilde{t}/2$, $H \equiv \tilde{\zeta}/3\epsilon \tilde{\Gamma}$, the nonlinear equation is expressed generically as

$$\frac{\partial H}{\partial T} + \frac{\partial^2 H}{\partial X^2} + \frac{\partial^4 H}{\partial X^4} + \delta \frac{\partial^3 H}{\partial X^3} + \frac{1}{2} \left(\frac{\partial H}{\partial X} \right)^2 = 0, \quad (4)$$

where δ is defined by $\delta \equiv -4\tilde{v}_x/3\sqrt{\epsilon}$. Equation (4) is a form of the Benney equation [24].

The Benney equation was originally introduced to describe the instability of a film flow [24]. Without the third derivative term or $\delta = 0$ in Eq. (4), we have the Kuramoto-Sivashinsky (KS) equation [25,26]. It is known to reveal a chaotic behavior spatially and temporally in the profile evolution of $H(X, T)$. On the other hand, when the second and the fourth derivative terms vanish or $\delta \rightarrow \infty$ in Eq. (4), we have the Korteweg-de Vries (KdV) equation, and a regular array of solitons is realized. The Benney equation thus interpolates between chaotic and regular patterns. It was studied numerically under a periodic boundary condition by Kawahara and co-workers [27]. When δ is sufficiently large ($|\delta| > 1.2$), equidistant solitonlike pulses are observed. Thus, near the critical point where ϵ is small and δ is large, the step

would show a regular periodic pattern propagating along the step edge with a constant velocity.

To test the above analysis we perform Monte Carlo simulation of a lattice model. The system size is 256×64 with periodic boundary conditions. The simulation algorithm is similar to that of Ref. [28] except that there is no Schwoebel effect but rather the drift of adatoms. The drift is incorporated as a bias in the diffusion trial. In a time interval Δt , an adatom occupying a site (i, j) moves with transition probabilities $(1 \pm p_x)/4$ to the neighboring sites $(i \pm 1, j)$, and with probabilities $(1 \pm p_y)/4$ to the sites $(i, j \pm 1)$. Also during this interval an adatom can evaporate back into the ambience with the probability, p_{ev} , but no deposition takes place. In the continuum and the low density limit, the prescription corresponds to the diffusion equation (1) with $D_s = 1/4\Delta t$, $v_x = p_x/2\Delta t$, $v_y = p_y/2\Delta t$, and $\tau^{-1} = p_{ev}/\Delta t$. We choose our units in such a way as to make D_s unity. The values of the other parameters are as follows: the equilibrium adatom density $c_{eq}^0 = 0.18$, the capillary length $\Gamma = 0.54$, the lifetime of adatoms $\tau = 256$, and $x_s = 16$. With these parameter values the instability is expected to occur at $8.4 \times 10^{-3} \leq v_y \leq 1.94$ from Eq. (3).

Figure 1 shows the time evolution of a step pattern in an extended frame, $0 \leq x \leq 256$ and $-600 \leq y \leq 0$, at various adatom drift velocities. The step is initially prepared straight: $\zeta(x, 0) = 0$. Without the adatom drift, the receding step shows only thermal fluctuation, but no indication of macroscopic instability for the pattern formation, as shown in Fig. 1(a). Figure 1(b) shows the step motion with a large drift in the uphill direction ($v_y = -0.8$). Since directions of the step retardation and of the adatom drift coincide, the step fluctuation is suppressed. A similar kinetic smoothing has been found for a receding step with the Schwoebel effect [28]. Here, the electromigration decreases the coefficient of the q^2 term in Eq. (3) and enhances the effective stiffness [29]. On the contrary, when the drift of adatoms is in the downhill direction, the fluctuation is enhanced. Above the critical value $v_c = 8.4 \times 10^{-3}$, wandering instability occurs as shown in Fig. 1(c). With $v_y = 0.2$ the wavelength of the most unstable mode is estimated by using the full expression of Eq. (3) as $\lambda_{max} = 27.5$ which is the order of the mean interval of the step protrusions. On closer inspection, however, one finds that protrusions appear at irregular intervals and that they collide and annihilate chaotically, as is expected from the analysis of the KS equation [2]. When a transversal component $v_x = 0.3$ is added to the vertical drift $v_y = 0.2$, the step morphology changes, as is shown in Fig. 1(d). The wandering step now shows a regular pattern as expected from the solution of the Benney equation with a large dispersion coefficient [4,18,19], estimated here as $\delta = -5.6$. The step wandering is thus proven to be controlled by the adatom drift. The most unstable wavelength λ_{max} is here estimated as $\lambda_{max} = 29.0$ which roughly agrees

FIG. 1. Time evolution of a receding step with adatom drift velocities; (a) $(v_x, v_y) = (0, 0)$; (b) $(v_x, v_y) = (0, -0.8)$; (c) $(v_x, v_y) = (0, 0.2)$; and (d) $(v_x, v_y) = (0.3, 0.2)$.

with the characteristic wavelength in the early stage of evolution in Fig. 1(d).

In this paper we study the effect of adatom drift on the wandering pattern of an isolated step. The wandering of the step is shown to be described generically by the Benney equation. When the drift has no component parallel to the step edge, the system has the inversion symmetry along the step, $x \rightarrow -x$, and the step obeys the KS equation, a limited case ($\delta = 0$) of the Benney equation. When the drift has a component parallel to the step edge, the parity symmetry is broken, and the wandering step shows a regular pattern at a large δ . Theoretically the simple continuum equation can be justified near the critical point as we have assumed $\epsilon \ll 1$ in the derivation. The result of the Monte Carlo simulation, however, suggests that the applicability of our equation is much wider.

We have considered, so far, the case without deposition, but the result can easily be extended to the case with a finite deposition flux F . For small $\tilde{v}_x$ and $\tilde{v}_y$, Eq. (3) is modified by replacing $\tilde{v}_y$ to $(1 - F/F_{eq})\tilde{v}_y$. Here F_{eq} is the equilibrium flux of deposition: $F_{eq} = c_{eq}^0/\tau$. When the overlayer is growing, $F > F_{eq}$, the instability

takes place when the drift is opposite to the step motion and the drift speed is larger than the critical value $\tilde{v}_c = 4\tilde{\Gamma}/(F/F_{eq} - 1)$. If the drift by the electromigration is in the same direction as the step motion, the kinetic smoothing takes place, which might be useful to manipulate a straight step.

Si(111) is one of the systems, where the wandering instability by electromigration is expected. From the critical drift velocity, we can calculate the critical electric field as $E_c = k_B T v_c / D_s Z e = 4\Omega\tilde{\beta}/x_s^2 Z e$. By using the experimental values $x_s \approx 2.5 \mu\text{m}$ [30] and $\tilde{\beta} \approx 2 \times 10^{-10} \text{J m}^{-1}$ [31], E_c is estimated at the order of 10^2V m^{-1} with $|Z| \approx 0.1$ [32], which is the common voltage for heating Si. The wandering is observed in Si(111) in MBE growth [8] without electric field, and might be caused by the Schwoebel effect. The wandering pattern there is strongly orientation dependent [33] due to the 7×7 reconstruction at the low temperature ($\leq 750^\circ\text{C}$). Therefore, the wandering instability we have studied is expected only at high temperatures where the step anisotropy and the Schwoebel effect become small. However, the derivation of Eq. (4) based on the expansion by a parameter $qx_s \ll 1$ is then rather difficult to justify, since the diffusion length x_s on Si(111) is extremely large there. But there are many other materials and combinations of adsorbates and substrates which show surface electromigration [12], and once the wandering instability takes place, the Benney equation may be applicable since the basic structure of the equation is determined by the symmetry of the system. Tilting of the electric field can then control the regularity and the periodicity of patterns formed on the crystal surface.

This work is supported by the Grant-in-Aid from the Japanese Ministry of Education, and by JSPS Research for the Future Program. The authors have also benefited from the interuniversity cooperative research program of the Institute for Materials Research at Tohoku University. They are also grateful to C. Misbah for careful reading of the manuscript.

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