

Linear smoothing of GaAs(1 0 0) during epitaxial growth on rough substrates

Michael B. Whitwick*, T. Tiedje, Tian Li

Physics and Astronomy, University of British Columbia, AMPEL, 2355 East Mall, Vancouver, British Columbia, Canada V6T 1Z4

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ABSTRACT

The smoothing of weakly roughened GaAs (100) substrates is measured with elastic light scattering during homoepitaxial growth of GaAs buffer layers. The smoothing measurements are used to determine the coefficient of the linear term in the continuum growth equation for GaAs, as a function of growth rate and temperature. The temperature and growth rate dependence are in good agreement with theoretical predictions from an atomistic description of the growth process. The density of atomic steps on the surface, which is a key parameter in the continuum growth equation, is measured independently using atomic force microscopy. The linear smoothing coefficients computed from the experimental values for the step density, are found to be in good agreement with the smoothing rates measured with light scattering. These experiments provide experimental support for the continuum growth model that has been derived analytically from basic atomic-level phenomena in epitaxial film growth.

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With the increasing interest in nanoscale semiconductor devices there is a need to understand and control surface and interface structure in epitaxial semiconductor films in a more quantitative way. A simple example of a structure-related phenomenon that takes place during epitaxial growth of semiconductors is the smoothing of an initially rough substrate during growth of an epitaxial buffer layer. In the case of GaAs (100) surfaces, the roughness produced by the oxide evaporation is smoothed out by the deposition of a few hundred nanometers of material. Atomically smooth interfaces are normally preferred for electronic and optical devices. Therefore, it is of interest to develop a more quantitative description of the surface morphology evolution of semiconductors not only from the perspective of understanding the underlying mechanisms but also for learning how to make better devices.

The continuum growth equation approach provides a simple way to describe the time evolution of the surface morphology [1–3]. For GaAs this approach is found experimentally to give a good description of the surface morphology on mesoscopic length scales greater than ~ 30 nm, and for small-angle surface

topography on the order of a few degrees or less [4]. In the growth of GaAs by molecular beam epitaxy, As is supplied in excess and the growth rate is limited by the flux of Ga. In this case it is common to treat the growth as being controlled by the dynamics of a single species. The time dependence of the surface morphology in the presence of a flux of deposited atoms can be described by a non-linear continuum growth equation of the form

$$\frac{\partial h}{\partial t} = v\nabla^2 h - \lambda_2 \nabla^2 (\nabla h)^2 + \text{noise} \quad (1)$$

where $h(x,t)$ is the surface height with respect to a flat reference plane that moves up as the surface grows. Following the experimental data we assume in this equation that the growth is stable ($v > 0$). The noise is associated with the random arrival, attachment and detachment of adatoms at the surface and produces a small surface roughness for the steady-state growth conditions [2]. The coefficients in the growth equation can be evaluated in terms of atomistic parameters, in the context of the classic Burton–Cabrera–Frank picture [5] of crystal growth in which adatoms diffuse on atomically flat terraces, nucleate monolayer islands on the terraces and incorporate into step edges [6–8]. In this picture the coefficients of the linear and non-linear

* Corresponding author. Tel.: +1 604 822 5425; fax: +1 604 822 4750.

E-mail address: whitwick@gmail.com (M.B. Whitwick).

terms are given by

$$v = \zeta F/S_0, \quad \lambda_2 = F/(\beta a^2 S_0^4) \quad (2)$$

respectively, where ζ is a dimensionless parameter that depends on the values of the potential barriers for adatom migration at step edges (Ehrlich–Schwoebel effect), F is the deposition rate in nm/s, S_0 is the step density, β is a dimensionless parameter of order unity and a is the crystal lattice constant. In this analysis the substrate is assumed to be isotropic in the plane of the surface, although in practice the GaAs (100) surface is not isotropic. Since the coefficients in the growth equation are proportional to the adatom flux, the growth equation describes kinetic smoothing, and not the smoothing in equilibrium due to surface diffusion associated with gradients in the surface chemical potential, such as is described by the Mullins equation [2,9].

Although the form of Eq. (1) is in agreement with a rather large body of experimental data on the surface morphology of GaAs [4,10–13], the expressions for the coefficients in the growth equation have not been tested as a function of temperature and growth rate for GaAs(100), or any other system. The purpose of this paper is to compare the theoretical expression for v , the coefficient of the linear term in the growth equation, with experimental data obtained from light scattering experiments, as a function of temperature and growth rate.

Surface roughness measurements were carried out using ultraviolet (UV) light scattering during homoepitaxial growth of GaAs on (100) oriented substrates in a VG molecular beam epitaxy system, for growth rates ranging from 0.01 to 6 ML/s at a constant As_2 flux of 8 ML/s and substrate temperatures between 400 and 590 °C. The substrate temperature was measured with bandgap thermometry [14], and the growth rates were determined by comparing ion gauge flux measurements in the growth chamber with X-ray diffraction measurements on the grown films. Before growth, the polished (100) on-axis ($\pm 0.5^\circ$) GaAs substrates were outgassed at 400 °C for 1 h before growth. The surface oxide was desorbed thermally by ramping the substrate temperature to 610 °C for about 10 min. Thermal oxide desorption results in a roughening of the surface (RMS roughness ~ 5 nm) through the explosive evaporation of the Ga oxide, creating random pits typically about 400 nm in diameter, 30 nm deep and 1 μm apart [15]. The samples were cooled rapidly at the end of the growth by turning off the substrate heater (~ 50 °C/min initial cooling rate), in order to freeze the step density in place. The grown films were imaged in air with an AFM equipped with 1 or 10 nm radius tips. The density of atomic steps was determined from the AFM images, with Matlab edge-finding software. An example of the edge-finding method is shown in the insert of Fig. 4.

In the light scattering experiments, a beam of 244 nm light was directed onto the substrate with a ~ 5 mm diameter spot. The substrate was oriented with the [110] or $[1\bar{1}0]$ directions parallel to the plane of incidence. The diffusely scattered light is measured at a non-specular viewport in the plane of incidence, using a photomultiplier with a line filter to block the background light. The angles of the incident and backscattered paths are 55° and -25° (backscattering) relative to the substrate surface normal, respectively. The specularly reflected UV beam passes through an optical port to a beam block outside the MBE chamber, in order to minimize background light that is scattered inside the growth chamber. The optical viewports were heated to 300 °C to prevent arsenic coating of the windows. The UV light is produced by frequency doubling the 488 nm line of a 2 W argon ion laser and carried to the MBE system through 15 m of UV grade optical fiber. The net UV power delivered to the substrate is typically 15 mW.

The intensity of the diffusely scattered light is proportional to the power spectral density of the surface morphology [16] at a spatial

frequency of $32 \mu\text{m}^{-1}$, which is set by the wavelength and the angles of the incident and scattered light. An example of the diffusely scattered light signal as a function of time during growth is shown Fig. 1. The starting GaAs substrate is smooth but becomes rough when the surface oxide is removed through thermal desorption. The clean surface smoothes slowly while it is annealed in an As_2 flux at the oxide desorption temperature and later at the growth temperature. Introducing a Ga flux starts the GaAs growth and causes rapid smoothing of the surface as indicated by the sharp drop in the intensity of the diffusely scattered light in Fig. 1. In agreement with the theoretical results in Eq. (2), the smoothing rate in Fig. 1 is much faster ($\sim 100\times$) when material is being deposited, than during annealing with no growth. The smoothing is characterized by two regimes: an initial rapid smoothing followed by a slower exponential decay of the power spectral density, as indicated in the semilog plot in Fig. 1. All of the light scattering measurements showed the characteristic fast initial decay followed by a slower exponential decay at long times. The background scattering is due to the residual steady-state roughness associated with the noise term in Eq. (1) or to scattering from small particles that contaminate the surface. In the following we will neglect the noise term in Eq. (1) and assume that it only contributes to the steady-state roughness at long times.

The experiments show that the surface morphology at the spatial frequency $q = 32 \mu\text{m}^{-1}$ is dominated by the initial roughness associated with the oxide desorption and decays during growth to a steady-state value. In the limit of weak surface topography, the non-linear term in Eq. (1) can be neglected and the surface smoothing will be dominated by the linear term. In this case, the time dependence of the scattered light intensity, which is proportional to the power spectral density (PSD) of the surface, will have the form

$$I(t) - I_0 \propto \exp(-2\nu q^2 t) \quad (3)$$

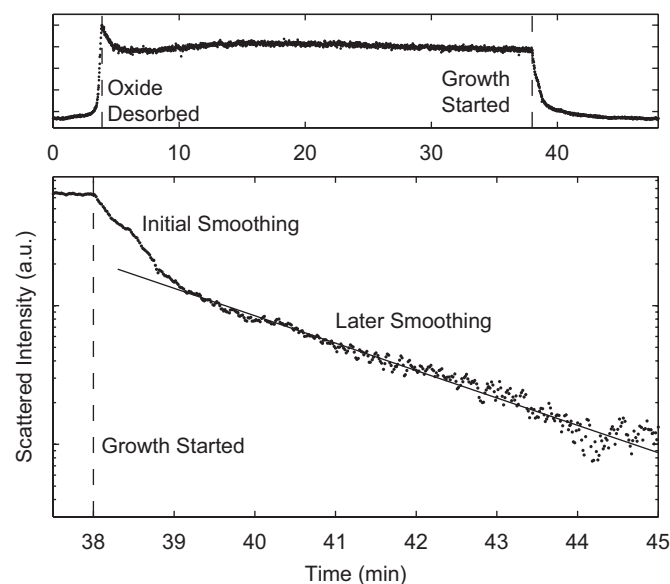


Fig. 1. Diffuse light scattering showing the effect of the desorption of the surface oxide and the start of growth of a GaAs buffer layer at 1 ML/s and 550 °C. The substrate was ramped up to 610 °C where it was held for 15 min to remove the surface oxide, then lowered to 550 °C, and kept at this temperature for 10 min before opening the Ga shutter at 38 min. The oxide evaporation corresponds to the abrupt increase in the scattered light signal at 3 min. Growth starts at 38 min when the Ga shutter is opened, which causes an abrupt decrease in the intensity of the diffusely scattered light. The semilog plot at the bottom shows the decrease in the scattered light intensity at the start of growth in more detail with the background scattering at long times subtracted.

where I_0 is the steady-state light intensity due to scattering off previous optical elements and the surrounding chamber. The factor of two in the exponent comes from the fact that the PSD is equal to the square of the spatial Fourier transform of the surface. The predicted exponential decay in Eq. (3) is consistent with experiments which show that the scattered light intensity decays exponentially at long times. By measuring the exponential decay rate of the scattered light intensity we can determine the coefficient ν of the linear term in the growth equation. The temperature and flux dependence of ν , inferred from the light scattering data, are shown in Figs. 2 and 3 for the scattering vector aligned parallel to the [110] and [1 $\bar{1}$ 0] directions. Since the [110] direction in the GaAs(100) surface is not equivalent to the [1 $\bar{1}$ 0] direction the surface smoothing rate is different in these two directions. Smoothing along the [1 $\bar{1}$ 0] direction is observed to be faster than in the [110] direction by a factor of about 2.5 in Figs. 2

and 3. The temperature and flux have little effect on the anisotropy in the smoothing rates. In the figures the values of ν are determined from the slower, long-time exponential decay, of the scattered light intensity. This is appropriate as the linear term dominates the surface smoothing for small amplitude surface topography and the amplitude is smallest at long times.

The step density is set by a balance between the rate of filling of terraces during growth and the nucleation of new monolayer islands [2]. It is expected to have the following power-law flux dependence:

$$S_0 \sim \left(\frac{F}{D}\right)^{x/(2x+4)} \quad (4)$$

where D is the adatom diffusion coefficient and x is the size of the critical nucleus for formation of a stable island on the surface. As shown in Fig. 2, the smoothing coefficient ν also has a power-law dependence on flux for Ga fluxes less than 1.5 ML/s. Combining Eq. (4) with the expression for ν (Eq. (2)) allows one to predict the exponent in the power-law dependence of ν on F . If we define the exponent z in the power-law expression for the smoothing parameter $\nu \sim F^z$, then for critical nucleus sizes of, 3 and 4 atoms the exponents z are $\frac{3}{4}$, $\frac{7}{10}$, and $\frac{2}{3}$ respectively. The experimental value for z from Fig. 2 is 0.68, which is consistent with critical nucleus sizes of 3 or 4.

For fluxes greater than 1.5 ML/s the coefficient ν decreases with increasing flux. The reason for this behavior is not known. The GaAs (100) surface reconstruction is known to depend on the As₂/Ga flux ratio. The flux ratio where the smoothing coefficient begins to drop in Fig. 2 is close to where one would expect a transition from the (2 × 4) reconstructed As-rich surface to one of the Ga-rich surface reconstructions [17–19]. Therefore the drop in the smoothing rate may be associated with the surface changing from As-rich to Ga-rich. Also at low Ga flux, we would expect the (2 × 4) reconstruction to change, to the more As-rich c(4 × 4) reconstruction [20–22]. However, there is no evidence in Fig. 2 of any corresponding change in the flux dependence of the smoothing rate at low Ga fluxes.

In Fig. 3, ν at 1 ML/s deposition rate shows a rather weak temperature dependence that can be fit to an Arrhenius form with activation energy 0.56 eV. According to the theoretical expression for ν the temperature dependence is associated with the ratio ζ/S_0 . Without more information it is not possible to assign the temperature dependence to one or the other of these two parameters. However, we can estimate the temperature dependence of S_0 from the temperature dependence of the diffusion constant, D . RHEED experiments indicate that the activation energy for the adatom diffusion constant in GaAs is between 1.5 and 2 eV [23,24]. Taking a critical nucleus size of 3.5, the activation energy for S_0 will in this case be between 0.48 and 0.64 eV according to the expression for S_0 in Eq. (4). Therefore the temperature dependence of the step density alone can explain the temperature dependence of ν . This interpretation is supported by direct measurements of the step density using AFM shown in Fig. 4, which has temperature dependence similar to $1/\nu$. In general, one would expect the dimensionless parameter ζ , which describes the downhill diffusion bias, to be temperature dependent in its own right. However, the temperature dependence may be rather weak. For example, if the diffusion bias were caused by a negative Ehrlich–Schwoebel (ES) barrier [25], lowering the barrier to adatom hopping at step edges by a few kT or more, will ensure that the adatoms hop in the direction of the lowered barrier with high probability, independent of temperature [7,8]. This will produce a temperature independent ζ equal to 0.5. Larger values for ζ can be expected in the presence of a barrier to attachment of adatoms at step edges from below (inverse ES barrier) [8].

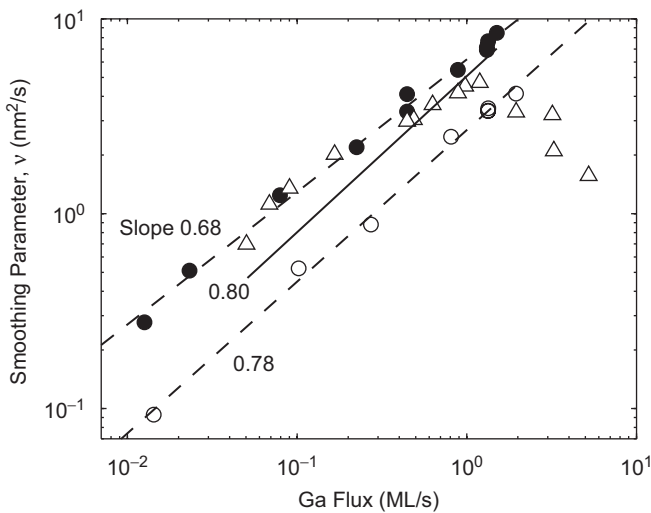


Fig. 2. Linear smoothing coefficient ν , inferred from light scattering data as a function of Ga flux at fixed As₂ overpressure. The symbols and lines have the following meanings: growth temperature of 550 °C (□) and 590 °C (●) with momentum transfer in the light scattering along the [1 $\bar{1}$ 0] direction; GaAs grown with a substrate temperature of 590 °C (○), with the scattering vector parallel to the [110] direction; the dashed lines are least square fits; the solid line is the linear smoothing coefficient calculated from Eq. (4) using $\zeta = 0.5$ and the fit to the step density data shown in Fig. 4.

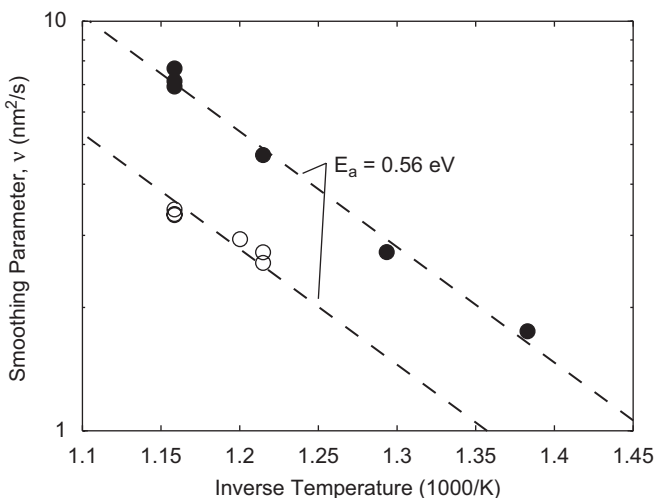


Fig. 3. Linear smoothing coefficient ν , as a function of temperature at a growth rate of 1 ML/s. The symbols indicate that the light scattering was measured parallel to the [1 $\bar{1}$ 0] direction (●), and the [110] direction (○).

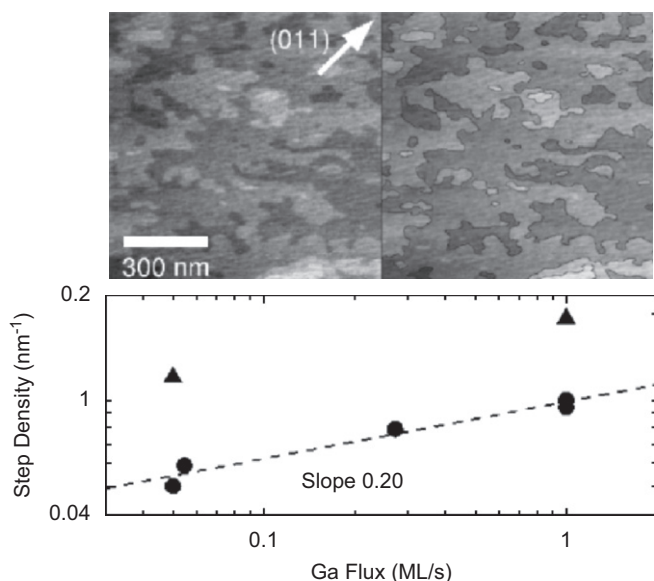


Fig. 4. Top: An AFM image used to determine the step density. In the right hand image the step edges are highlighted with an edge-finding routine. The average step density is 0.079 nm^{-1} . Bottom: Step density obtained from AFM images of samples grown with different Ga fluxes. The GaAs layer is grown to a thickness of 500 nm. The symbols indicate growth temperatures of 590 °C (●) and 400 °C (▲).

The step density measured by AFM after growth at different fluxes and growth temperatures is shown in Fig. 4. The flux dependence of the step density is consistent with Eq. (4). Specifically, for critical island sizes of 2 and 3, the flux dependences of the step density should have the form $S_0 \sim F^{0.25}$ and $S_0 \sim F^{0.30}$, respectively. Both expressions are consistent with the AFM data, which gives an exponent of 0.25, but with an uncertainty that encompasses 0.30. One can turn the problem around and use the AFM measurements of the step density to estimate the smoothing coefficient. To estimate the smoothing coefficient we substitute the flux dependence of the step density found from the AFM data in Fig. 4 (solid line), into the expression for ν in Eq. (2) with $\zeta = 0.5$. The inferred flux dependence of the smoothing coefficient is indicated by the solid line in Fig. 2. Of course the smoothing rates measured by light scattering depend on the direction relative to the crystallographic axes due to the anisotropy of the GaAs(100) surface. We have not attempted to include the anisotropy in the model. Nevertheless the smoothing coefficient ν inferred from the step density is in rather good agreement with the values for ν obtained from light scattering. From results in the literature we expect the sample in Fig. 4 grown at 400 °C to have the (4×4) reconstruction while the sample grown at 590 °C is expected to have the (2×4) reconstruction [17–22].

The comparison between the model and the experiments suggests that the diffusion bias parameter ζ is close to unity, and that it has a weak temperature dependence. A negative ES barrier and/or a small incorporation barrier, on the order of kT , will produce a diffusion bias of this magnitude. The physical significance of $\zeta = 1$ is that adatoms deposited on a vicinal terrace, will experience, on average, a displacement of one interatomic distance in the downhill direction. Small values for the step edge barriers, on the order of 0.03–0.07 eV, are physically reasonable for GaAs. The ES barrier and incorporation barriers are similar to next nearest neighbor interactions and therefore should be significantly smaller than the lateral binding energy of an adatom at a step edge. Experimentally the lateral binding of an adatom at a step edge is estimated to be $\sim 0.3 \text{ eV}$ in GaAs(100) [26]. It is remarkable that the small energy associated with the

asymmetry in the interatomic potential near a step edge, makes the difference between stable growth as observed in GaAs(100), and unstable growth leading to the formation of surface mounds, as commonly observed in metals [27]. The small step edge barriers can be compared with the much larger 1.5–2 eV energy barrier for adatom hopping on a flat terrace [26].

We now estimate the magnitude of the non-linear term in Eqs. (1 and 2). The typical rms roughness for a thermally deoxidized surface $h_{\text{rms}} \sim 5 \text{ nm}$, which corresponds to a 25 nm peak-to-peak roughness for Gaussian noise [4]. If the characteristic length of the surface roughness is $1 \mu\text{m}$, then the step density will be 0.25 nm^{-1} from the surface topography alone, assuming atomic steps are 0.4 nm high. At the end of the rapid smoothing the power spectral density typically drops by a factor of five (see Fig. 1). The surface amplitude will decrease by the square root of the change in the PSD from 25 to 11 nm peak to peak, and the step density decreases by the same factor to 0.11 nm^{-1} . This step density is similar to the measured value after growth in Fig. 1. We can use this value to estimate the size of the non-linear smoothing term. To compare the non-linear smoothing with the linear smoothing we linearize the non-linear term in Eq. (1) and obtain a linearized rate $\lambda_2 q^2 h_{\text{rms}}$, analogous to that for ν . According to the model $\lambda_2 = F/(\beta a^2 S_0^4) \sim 9 \times 10^2 \text{ nm}^3/\text{s}$ for $\beta = 5$ and a growth rate of 0.27 ML/s as in Fig. 2. This gives a linearized smoothing term, analogous to ν , of $2 \text{ nm}^2/\text{s}$. This is of the same order as the linear smoothing at the same flux and growth rate. Since it depends on the fourth power of the step density this estimate will vary significantly with small changes in the step density. The non-linear term is clearly important since AFM measurements of the shape of the smoothed surface show rounded mounds separated by V-shaped valleys [10]. This lack of inversion symmetry in the surface topography is characteristic of a non-linear growth equation. We cannot be sure that the linear term dominates the smoothing rate, from the theoretical estimates for the relative magnitude of the two terms alone. However, the temperature and flux dependence of the smoothing rates are consistent with the smoothing being dominated by the linear term and are not consistent with it being dominated by the non-linear term. The observed exponential decay is another indication that the linear term dominates at long times. Due to the dependence of λ_2 on the inverse fourth power of the step density we would expect a much weaker flux dependence, and stronger temperature dependence for the smoothing rate, than is observed, if the non-linear smoothing were dominant. In fact, depending on the size of the critical nucleus, x , the non-linear coefficient λ_2 , will have almost the same temperature dependence as the diffusion constant, which is much larger than the observed temperature dependence of the smoothing rate.

In conclusion, we have used UV light scattering to measure the growth rate and temperature dependence of the smoothing of rough GaAs(100) substrates during MBE growth. The temperature and flux dependence of the smoothing rate are consistent with a continuum growth equation description, in which the parameters in the growth equation are derived from a Burton-Cabrera-Frank type atomistic model for epitaxial growth. In this model the step density is the principle experimental parameter, which controls the surface smoothing. The experimental results suggest that the linear term in the growth equation dominates the smoothing rate in the case of the small amplitude surface roughness studied in the experiments. In the growth model we assume that there are potential barriers at atomic steps that favor downhill adatom migration. The measured smoothing rate indicates that adatoms deposited from the vapor onto a stepped surface migrate in the downhill direction by an average of one interatomic distance, with only a weak dependence on the substrate temperature. This behavior can be explained by a negative Ehrlich–Schwoebel

barrier at step edges and/or by a weak inverse Ehrlich–Schwoebel barrier to adatom incorporation from the lower terrace. The smoothing rates measured in the light scattering experiments are in good agreement with theoretical predictions based on independent experimental measurements of the step density obtained from atomic force microscopy images.

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