Smoothing of textured GaAs surfaces during molecular beam epitaxy growth

M. Adamczyk, A. Ballestad, and T. Pinnington  
Advanced Materials and Process Engineering Lab, Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia V6T 1Z4  

T. Tiedje  
Advanced Materials and Process Engineering Lab, Department of Electrical and Computer Engineering, University of British Columbia Vancouver, British Columbia V6T 1Z4  

M. Davies and Yan Feng  
Institute for Microstructural Sciences, National Research Council of Canada, Ottawa, Ontario K1A 0R6, Canada  

(Received 10 October 1999; accepted 20 December 1999)  

The surface morphology of homoepitaxial GaAs layers grown by molecular beam epitaxy on random and periodically textured substrates has been measured by atomic force microscopy and elastic light scattering. The random texture was obtained by thermal evaporation of the surface oxide and the periodic texture consisted of one-dimensional grating patterns fabricated by holographic lithography. The time evolution of the surface morphology was simulated numerically with a nonlinear growth equation that includes deposition noise and anisotropy in the surface diffusion. The surface of the random substrate develops shallow mounds as the large amplitude initial texture smooths out, an effect that has previously been attributed to unstable growth. © 2000 American Vacuum Society. [S0734-211X(00)01103-3]

I. INTRODUCTION

Historically, most epitaxial film growth has been carried out on planar substrates in order to make uniform layers such as superlattices and one-dimensional quantum wells. In order to make higher dimensional nanostructures and certain kinds of optoelectronic devices such as distributed feedback lasers it is of interest to be able to predict and control the shape of films grown on patterned substrates. The surface morphology of patterned substrates evolves during growth due to migration of the deposited atoms on the surface and to noise inherent in the deposition process. Ideally one would like a growth equation that describes how the surface morphology evolves during growth on patterned substrates. The same equation will also describe the roughening of initially flat surfaces in the presence of deposition noise. The presence of mounds on the surface of molecular beam epitaxy (MBE) grown GaAs layers has been interpreted as evidence that the growth equation for GaAs is unstable, although this may be controversial.

The scaling behavior of random surface structures during film growth has been explored theoretically for a variety of different growth equations in the presence of deposition noise. Pinnington et al. have succeeded in interpreting measurements of surface roughening during strain relaxation using the Edwards Wilkinson equation. In this article, we describe the results of experiments on the growth of GaAs films on textured GaAs substrates in which we measure the surface morphology as a function of time during the film growth. We compare the experimental results with numerical simulations of the time evolution of the surface morphology using an anisotropic Kardar–Parisi–Zhang (AKPZ) equation.

The GaAs homoepitaxial growth experiments were carried out in a VG-V80H MBE deposition system, equipped with solid-source effusion cells for both group III and group V elements. The substrate temperature was monitored throughout growth using an optical band gap thermometer, with an absolute accuracy of about 5 °C. Prior to loading into the MBE system, the substrates were oxidized by exposure to ultraviolet ozone, to reduce carbon contamination of the surface. The wafers were then transferred to the MBE growth chamber for a final oxide removal. Depending on the sample, either thermal cleaning or atomic hydrogen etching was used for surface oxide removal prior to growth as discussed later. Thermal oxide evaporation is a widely used method for cleaning GaAs substrates prior to MBE growth. After growth, the samples were annealed at the growth temperature for 10 min under an As2 flux and then cooled to 450 °C in about 10 min before interrupting the As2 flux. The substrate heater was then shut off to quench the sample by radiative cooling to the liquid nitrogen filled cryoshroud.

The surface morphology of the films was monitored during the growth using elastic light scattering with both 250 nm wavelength light from a 100 W Hg arc lamp and 488 nm light from a 10 mW Ar ion laser. The light scattering experiment is described in more detail in a previous publication. The light scattering signal is proportional to the power spectral density (PSD) of the surface roughness at a spatial frequency q defined by the scattering geometry and the wavelength of the incident light. In this way two different spatial frequencies corresponding to roughness on length scales of 154 nm (41 μm−1) and 393 nm (16 μm−1) were monitored.

Electron mail: adamcyk, aballest, pinn, tiedje@physics.ubc.ca
simultaneously throughout the growth experiments. The in-plane scattering vector for the 16 μm⁻¹ scattering is oriented at 45° with respect to the [011] direction. No changes in surface structure were observed by light scattering during the cool down. Ex situ characterization of the samples carried out with a Digital Instruments Multimode atomic force microscope (AFM) indicate that the postgrowth anneal does not affect the surface structure on micron length scales, in accordance with the findings of Coluci et al.⁶

**II. RANDOMLY TEXTURED SUBSTRATE**

Randomly textured substrates were produced by thermal desorption of the surface oxide in ultrahigh vacuum, starting with polished (001) oriented, on-axis (±0.5°), GaAs substrates. The oxide was desorbed by a thermal treatment in which the substrate temperature was ramped, under As₂ overpressure, at a constant rate of (5±1) °C/min up to a final temperature of 600 °C and subsequently annealed for approximately 20 min. During the temperature ramp there is an explosive evaporation of the native oxide⁷,₈ which leaves an oxide free surface with shallow pits up to about 30 nm deep, as shown in the AFM image in Fig. 1(a). This roughened surface was then used as a substrate for studying the smoothing that takes place during film growth. Nominally 1-μm-thick GaAs layers were deposited at a growth rate of 2.1 Å/s and a substrate temperature of 553±5 °C. The As₂ beam-equivalent pressure was 2.5×10⁻⁶ Torr.

In Fig. 2, we show the time evolution of the two different wavelength scattered light signals during the buffer layer growth. The scattered light intensity decays exponentially with time which confirms that the surface smooths during the film deposition. Remarkably the decay rate is independent of spatial frequency for q = 16 and 41 μm⁻¹ [see Fig. 2(a)], and is also independent of temperature from 553 to 585 °C as shown in Fig. 2(b). These results are inconsistent with the predictions of standard growth equations.¹ For example, in the case of the Edwards–Wilkinson (EW) equation

\[
\frac{\partial h(r,t)}{\partial t} = \nu \nabla^2 h(r,t) + \text{noise},
\]

where \( h(r,t) \) is the surface height at \((r,t)\) relative to a flat reference plane, one would expect a q-dependent surface smoothing rate. In the present situation, where there is a large initial roughness, the deposition noise can be neglected and the EW equation predicts that the surface power spectral density will decay exponentially with a q-dependent decay rate

\[
\hat{h}(q,t)^2 \propto \exp(-2\nu q^2 t).
\]  

The absence of a q dependence in the experimental smoothing rate means that the surface morphology dynamics of GaAs cannot be described by an equation of the EW type or by any similar equations with higher order spatial derivatives at least for the relatively rough surface produced by the thermal desorption of the oxide. A possible explanation for the observed behavior is that the deposited atoms are preferentially captured by the abundance of step sites that exist in the oxide desorption pits on the surface thereby reducing the amplitude of the surface roughness without affecting the q dependence of the PSD.

In spite of the discrepancy with the light scattering data on the thermally cleaned substrate noted earlier, the EW equation has been used successfully to model the surface roughening of initially smooth InGaAs/GaAs heteroepitaxial layers during strain relaxation through formation of misfit dislocations.⁴ In this case the parameter \( \nu \) was found to be temperature dependent. In the present experiment, if the smoothing were described by Eq. (2), then one would expect a temperature dependent smoothing rate, also contrary to

---

**Fig. 1.** 5×5 μm AFM images of GaAs surfaces (a) after thermal oxide removal (b) after growth of a 1-μm-thick GaAs buffer layer on a thermally cleaned substrate and (c) 1×1 μm blow up of preceding image. Grayscale values are (a) 30, (b) 4, and (c) 4 nm. The black line is aligned along the [011] crystal direction, which we define as the x direction.
what we observe. The different behavior in the InGaAs strain relaxation experiment may be caused by a difference in the amplitude of the surface roughness in the two experiments. In the present experiment, the peak initial roughness of the starting surface is about 30 nm, which is an order of magnitude larger than the surface roughness in the InGaAs experiments. Accordingly, one might expect a transition to EW-like behavior as the surface roughness is smoothed out. The change in slope of the 16 μm⁻¹ data in Fig. 2(a) at long times may be an indication of such a transition. From the slope of the solid line in Fig. 2(a) we infer that ν in the EW equation is equal to 2.6 nm²/s. This is in the same range as the ν values determined from the simulations discussed later. We test the continuum growth equations by comparing the final surface morphology of a buffer layer with the predictions of a simulation. Since the buffer layer is much smoother than the thermally cleaned surface an EW-like growth model may be applicable in this case even if it does not correctly describe the behavior for large amplitude surface roughness.

The similarity between the lateral length scale of the mounds in the buffer layer in Fig. 1(b) with the spacing of the oxide desorption pits on the starting substrate, suggest that the morphology of the buffer layer is a remnant of the structure of the starting surface. Furthermore, the buffer layer in Fig. 1(b) shows two features that are not accounted for by Eq. (1): first, the surface structure is anisotropic in the plane, and second, the inversion symmetry is broken in that the valleys have a cusp-like shape while the ridge tops are smoothly rounded. The simplest way to incorporate these features in the model is to add a quadratic, nonlinear term to Eq. (1) and modify the spatial derivatives as follows:

\[
\frac{\partial h(r,t)}{\partial t} = \left( \nu_x \frac{\partial^2 h}{\partial x^2} + \nu_y \frac{\partial^2 h}{\partial y^2} \right) h(r,t) + \frac{1}{2} \left[ \lambda_x \frac{\partial}{\partial x} + \lambda_y \frac{\partial}{\partial y} \right] h(r,t) + \text{noise.} \quad (3)
\]

This is the anisotropic version of the KPZ equation. The quadratic term is nonconservative—it violates mass conservation—and therefore cannot be applicable in MBE growth where typically all of the deposited group III atoms are incorporated into films. Nevertheless, we use the quadratic term as a substitute for more complex conservative nonlinear terms with higher order derivatives which are much more difficult to simulate.

We use a finite-difference, time-domain numerical scheme to solve Eq. (3). The equation is discretized on a 512×512 grid so that we can use data obtained by AFM as our initial condition. We label the discretized points \( H^n_{i,j} \) at time \( n\Delta t \) for spatial site \((i\Delta x,j\Delta y)\). The iteration rule then becomes

\[
H^{n+1}_{i,j} = H^n_{i,j} + \Delta t \left[ \nu_x \frac{H^n_{i+1,j} - 2H^n_{i,j} + H^n_{i-1,j}}{\Delta x^2} + \nu_y \frac{H^n_{i,j+1} - 2H^n_{i,j} + H^n_{i,j-1}}{\Delta y^2} \right]
+ \Delta t \left( \lambda_x \frac{\partial}{\partial x} h(r,t) + \lambda_y \frac{\partial}{\partial y} h(r,t) \right) + \text{noise.} \quad (4)
\]

where the last term represents the deposition noise for deposition rate \( F \), and lattice constant \( a \); \( \eta(t) \) is a random number with a uniform distribution in the range \([-0.5, 0.5]\).²

Although the light scattering signals return to the level of the starting polished substrate during the GaAs growth, the AFM images suggest that a correlation between the surface

---

**Fig. 2.** In situ light scattering measurement during GaAs growth on thermally cleaned substrate. The background level of the light scattering signals has been subtracted from the light scattering data in order to isolate the change in the power spectral density due to the smoothing effect of the growth. (a) Simultaneously acquired signals at two different spatial frequencies: 16 and 41 μm⁻¹ and (b) scattered intensity at 41 μm⁻¹ acquired during two different growth runs performed at 553 and 585 °C.
structure of the deposited film and the initial substrate roughness remains after growth of the 1 μm buffer layer. AFM images of a 1-μm thick GaAs epilayer grown on a thermally cleaned GaAs substrate are shown in Figs. 1~b! and 1~c!.

Comparing this surface with the thermally cleaned substrate shown in Fig. 1~a!, we find that the rms surface roughness has been reduced from 5 to 1 nm. The high spatial frequency roughness of the thermally cleaned surface has been replaced by a more slowly varying surface structure. However, the length scale of the mounds present on the postgrowth surface is similar to the spacing of the pits observed in the thermally cleaned substrate. To test whether the growth equation can correctly describe the observed morphology of the buffer layer, we simulated the time evolution of the surface morphology using Eq. (3). The oxide desorbed surface in Fig. 1~a! was taken as the initial condition for the computation. The parameters $n$ and $\lambda$ are adjusted by trial and error, for a visual match to the AFM image of the buffer layer in Fig. 1~b!. The result is shown in Fig. 3, with parameters as indicated in the figure caption.

The simulation reproduces the length scale and anisotropy of the mounds in the buffer layer reasonably well. The valleys in the simulation show a more cusp-like shape than the tops of the ridges, similar to the buffer layer. The density of mounds is not reproduced as well. Based on the similarity between the structures in the simulation due to the initial condition, and the features that are observed in the sample, we conclude that the mounds on the surface of the buffer layer are due to the incomplete smoothing of the initial surface roughness. This conclusion is further supported by the fact that buffer layers grown on substrates cleaned with atomic hydrogen without the oxide desorption pits, do not show mounds. This is consistent with Coluci et al.’s interpretation of similar phenomena occurring in chemical beam epitaxy growth. 

In Fig. 4, we show the PSD parallel to the [011] and [01] crystal directions, for the buffer layer surface as well as for the simulation surface from Fig. 3. The PSD for the simulation is shifted to slightly lower spatial frequency compared with the sample. This is apparent visually as the simulation has a more smoothly varying topography [compare Figs. 1~b! and 3]. Note also that the PSD drops off at high spatial frequencies approximately as the 4th power of $q$. This is another indication of the role of the initial conditions as the long time PSD will decay as $q^{-1.58}$ for surfaces described by the KPZ equation. 

### III. PERIODICALLY TEXTURED SUBSTRATES

Experiments were also carried out on wafers patterned by photolithography. One-dimensional grating masks were defined by holographic lithography and etching of a SiO$_2$ mask on the semiconductor. The SiO$_2$ mask is 100 nm thick and tends to create straighter side walls than masks defined in photoresist, which preserves the uniformity along the length of the gratings for deep etches. The grating has a a pitch of 400 nm. After a 2 min etch in an electron cyclotron resonance etcher using a Cl$_2$/BCl$_3$/Ar gas mixture, we obtain almost perfectly square, 100 nm deep gratings as shown in Fig. 5. The gas flow rates were 0.5 sccm Cl$_2$, 1.0 sccm BCl$_3$, and 10 sccm Ar at a pressure of 10 mTorr. The radio frequency power was 17 W. The temperature of the sample was measured during etch using an optical band gap thermometer and found to be about 35±5°C. Upon completion of the etch, the sample is immersed in dilute HF to remove the remaining SiO$_2$. 

---

Fig. 3. KPZ simulation with the same orientation and scale as Fig. 1(b), using the surface in Fig. 1(a) as the initial condition. The parameters used in the simulation are $n_x=10$, $n_y=1$ nm$^2$/s, $\lambda_x=1$, and $\lambda_y=5$ nm/s.

Fig. 4. Power spectral density of the surface of a GaAs buffer layer obtained by Fourier transforming the AFM image in Fig. 1(b) (dotted lines) and power spectral density obtained by Fourier transforming the simulated surface shown in Fig. 3 (solid lines).
The native oxide was removed in the MBE growth chamber with atomic hydrogen to avoid the surface damage produced by thermal cleaning. The atomic hydrogen cleaning was performed by introducing molecular hydrogen into the growth chamber via a feed line mounted on a retractable flux gauge manipulator in front of the substrate. The hydrogen is cracked using a W filament placed at the end of the manipulator. During the etch, the MBE growth chamber pressure is $3.5 \times 10^{-6}$ Torr. The etch is stopped after a streaky reflection high-energy electron diffraction (RHEED) pattern is observed thereby indicating that the native oxide on the sample has been removed. During the etch, the substrate heater is adjusted to provide a temperature of 150 °C, however, radiant heating from the filament causes the substrate temperature to rise above this set point.

In Fig. 5, we show AFM cross sections of a lithographically defined grating on a GaAs substrate and a similar cross section after depositing a 0.5-μm thick film. Also shown in Fig. 5 is a simulated cross section obtained by solving Eq. (3) using the AFM image of the grating pattern as the initial condition. The simulation is in excellent agreement with the experimental data. The cusp-like valley cross sections clearly demonstrate the need for a nonlinear term in the growth equation. Both the $\nu$ and $\lambda$ parameters are smaller than the parameters used in the simulation in Fig. 3 ($2 \text{ nm}^2/\text{s}$, $0.05 \text{ nm}/\text{s}$ compared with $10 \text{ nm}^2/\text{s}$, $1 \text{ nm/s}$ for Fig. 3). The reason for this difference is not understood as one would expect the higher temperature growth on the grating to show a larger value for $\nu$.

### IV. CONCLUSIONS

We find that conventional growth equations cannot describe the smoothing of large amplitude/high aspect ratio surface roughness on GaAs. However, once the surface roughness has been reduced to a small value, on the order of 1 nm, by growing a buffer layer, we find that simulations based on an AKPZ equation describe the observed surface shapes reasonably well. The mounds observed in GaAs buffer layers deposited on thermally cleaned substrates are interpreted as being due to incomplete smoothing of the starting substrate wafer rather than unstable growth as has been suggested earlier. The nonlinear continuum growth equation describes the development of the mounds from the initial conditions, but in the end after very long deposition times the surface should evolve to a roughness level governed by the deposition noise.

### ACKNOWLEDGMENTS

The authors thank NSERC, NRC, and Rogers Canadian Cable Labs Fund for financial support. One of us (M.A.) thanks the BC Science Council for a graduate fellowship.

---