Two-component simulation for molecular beam epitaxy growth of GaAs

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Presented is a two-component simulation for molecular beam epitaxy growth of GaAs. The key aspects of the simulation are the high surface mobility of one component (Ga) and the inclusion of a second, volatile component (As). Simulations show how the interplay of the substrate temperature and As overpressure determines the supersaturation, and therefore whether the growth occurs in the kinetically limited or near-equilibrium regime. Qualitative comparisons of the simulations and experiment are presented.

Numerical simulations have led to a better understanding of molecular beam epitaxy (MBE) growth.[1–11] MBE growth is an extremely complicated mix of surface chemistry, reaction diffusion, and bulk incorporation. It is impossible to include all details, or study all aspects of MBE growth in one model. In order to extract useful information and gain new knowledge from simulations, some simplifications must be made. For III-V MBE growth, it has become common to eliminate certain details such as the crystal structure[2–4] and surface reconstruction.[5–9] The current state of the art uses first-principal total-energy calculations to extract material parameters and then input these into kinetic Monte Carlo simulations.[11] This technique holds much promise for the future, but has so far been used only for small lattice sizes and short deposition times.

While actual GaAs growth involves two elemental components, some models only consider one species (Ga). The one-component models have generated interesting results, and at times striking agreement with reflection high-energy electron diffraction (RHEED) and scanning tunnel microscopy (STM) experiments.[12,13] However, the limitations to single-component modeling are many. One of the issues that cannot be described correctly by a one-component simulation is: does the growth flux of Ga atoms represent a significant perturbation to the equilibrium thermal background of Ga atoms on the surface? Equivalently, what is the supersaturation, the ratio of the Ga adatom concentration during growth to that during equilibrium, under typical growth conditions? This is a very important issue for all growth phenomena. If the supersaturation is high, then growth is in a regime where irreversible kinetic effects dominate; conversely, if the supersaturation is low then growth is near equilibrium with reversible dynamics and entropy playing a large role.

To develop a simulation model to explore this question the role of substrate temperature and Ga, As fluxes needs to be considered. These factors are the primary controls of the Ga adatom concentration. In one-component models there is simply no role for the As overpressure. In this paper, we present a simple two-component model for GaAs growth. This model allows for physical processes that are missing from one-component simulations, i.e., the decomposition of GaAs on the surface and the subsequent desorption of As atoms from the surface.[14] Results from this model show that it is possible for MBE growth to be either kinetically or entropically controlled and that the smoothest growth occurs at the crossover between these two conditions.

It is important to note that we are not the first group to explicitly treat the multicomponent nature of MBE growth. The early paper by Ghaisas and Madhukar presented simulations examining the surface reactions necessary to explicitly treat the molecular nature of As_2 and its decomposition.[3] Itoh *et al.* considered in detail the effects of the As stabilized 2×4 surface reconstruction on the dynamics of island nucleation and growth.[10] More recently, Scheffler's group has used a combination of DFT and kinetic Monte Carlo simulations to examine GaAs and InAs/GaAs growth.[11] These works show the power of a first-principle calculation. Our work takes a different approach to the inclusion of both growth species. We have tried to make the fewest alterations to the standard SOS model and yet capture the new surface dynamics that multiple growth species exhibit. To this end we have chosen not to include the detailed surface chemistry of the two components.[3,11,14]

Our two-component model for MBE growth of GaAs is based on a cubic lattice with no surface reconstruction.[15] Each lattice site can be occupied by either a Ga atom or a GaAs unit. Driven by experimental observations, the Ga atoms on the surface are given a high mobility by treating them in a mean-field approximation, effectively conferring rapid diffusion over large distances. In this model the surface GaAs unit is immobile; however, it can decompose into a Ga atom, which, as described above, diffuses rapidly on the surface and an As atom, which leaves the surface via desorption.[14] The possibility of Ga adatom diffusion normal to the surface is ignored. These simple rules define the substance of the simulation. They were developed to include two specific features of the MBE process: rapid surface diffusion of Ga and decomposition of GaAs into mobile Ga and desorbed As.

Throughout the simulations an As flux is used. This is meant to simulate the As overpressure present during MBE growth. Impinging As atoms adhere to the surface with probability equal to 1 if they land on a lattice site occupied by a Ga adatom, otherwise they will not adsorb on the surface. To initiate film growth a Ga flux is added to the surface. The sticking coefficient of the incoming Ga atoms is unity. For
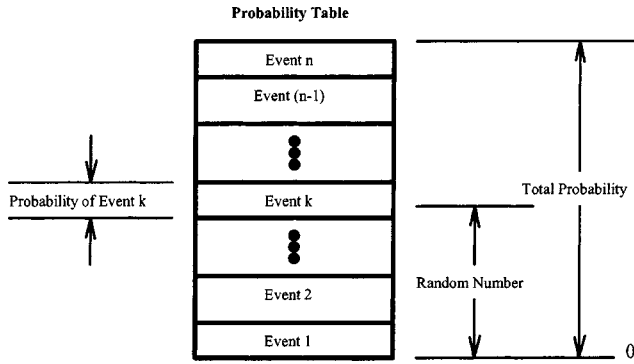
FIG. 1. Schematic illustration of the simulation event table and the means of selecting a random event to occur. This technique increases the speed of the simulations.

FIG. 2. Plot of the Ga concentration as a function of substrate temperature and arsenic overpressure.

the simulations presented in this paper the ratio of the As flux to Ga flux during growth is fixed at 10:1.

There are four possible generic types of events that can occur during MBE growth: Ga diffusion, incoming As atoms combining with Ga atoms to form GaAs, GaAs decomposition followed by As desorption, and Ga arrival. The event probabilities are calculated for each lattice site for the possible outcomes and placed in a stack. Then a random number is chosen to determine which event will occur (Fig. 1). The probability stack gets updated after every new event.

Due to the high mobility of the Ga atoms on the surface, it is computationally inefficient to explicitly track each one of them individually. Instead, a mean-field approach is adopted. We treat the Ga adatoms as having infinite mobility and replace their occupation with spatially varying probabilities. The probability depends on whether the lattice site is at a step edge or on a terrace. The probabilities of finding Ga atoms at a step-edge site (P_{edge}) and on a terrace site (P_{terr}) are determined by the following two simultaneous equations:

$$P_{edge}N_{edge} + P_{terr}(N_{site} - N_{edge}) = N_{Ga}, \quad (1)$$

$$P_{edge}/P_{terr} = \exp(T_c/T), \quad (2)$$

where N_{edge} is the number of step-edge sites, N_{site} is the total number of lattice sites, N_{Ga} is the total number of Ga atoms on the surface, T is the substrate temperature, and T_c is the temperature at which the ratio P_{edge}/P_{terr} is e. The probability of finding a Ga adatom at a step-edge site is higher than that at a terrace site and this is reflected in a nonzero T_c. This accounts for the higher binding energy of Ga atoms at step-edge sites. Ga atoms reach a thermodynamic equilibrium rapidly between step edges and terrace sites. At lower temperatures, because most Ga atoms cannot break away from the relatively stronger binding at step edges, very few Ga atoms are found on the terraces. On the other hand, at higher temperatures more Ga atoms can be found on the terraces because the Ga atoms have enough thermal energy to break away from step edges. T_c is used as a free parameter in the model. It was adjusted so that the probability of a Ga atom to be located at a step edge was twice as large as on a
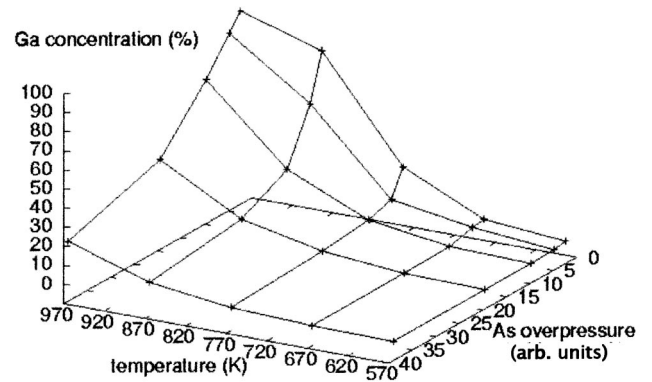
terrace site. The inclusion of this effect is reasonable, but its presence is not essential to any of the simulation results or conclusions drawn.

When an impinging As atom lands on a site occupied by a Ga atom, the As atom combines with the Ga atom to form a GaAs unit on the surface at that lattice site. If the As atom lands on a GaAs site, it simply leaves the surface, consuming no time. The probability $P_{As\downarrow}$ of an As atom coming down and combining with a Ga atom at a certain site is

$$P_{As\downarrow} = \frac{R_{As}}{\text{sites}} \times P_{Ga}(\text{site}), \quad (3)$$

where R_{As} is the As flux rate in number of As atoms per time unit, sites is the total number of lattice sites on the surface, and $P_{Ga}(\text{site})$ is the probability of finding a Ga atom at the site, determined by Eqs. (1) and (2).

The probability $P_{As\uparrow}$ for a GaAs unit to decompose into a surface Ga and a desorbed As atom is determined by bond counting and temperature. The explicit function is given by the following equation:

$$P_{As\uparrow}(\text{site}) = [1 - P_{Ga}(\text{site})] \times e^{-[E_0 + \text{nn(site)} \times E_{nn}]/k_B T}, \quad (4)$$

where $P_{Ga}(\text{site})$ is the probability of finding a Ga atom at a site; E_0 is the binding to the substrate, 0.25 eV; nn(site) is the number of nearest neighbors at a site, E_{nn} is the binding energy between nearest neighbors (0.15 eV), k_B is the Boltzmann constant, and T the temperature. The possibility for GaAs decomposition on the surface is the central different feature of this model.

Simulations are performed on a 128×128 lattice unless otherwise indicated. The computation starts with a smooth GaAs surface and an As flux, but no Ga flux. With the probability discussed earlier, thermal GaAs decomposition and As desorption can occur producing Ga adatoms on the surface. All the while, As atoms in the flux can combine with liberated Ga adatoms on the surface to form GaAs units. With these dynamics the system is allowed to come into equilibrium where the mean Ga adatom concentration no longer varies in time.

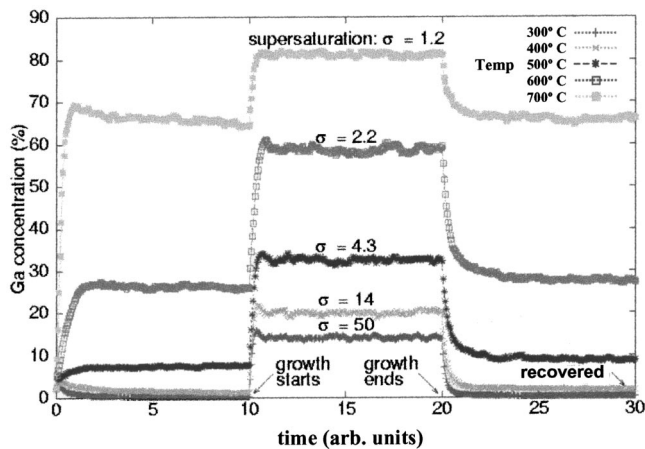The equilibrium Ga adatom concentration is a function of both substrate temperature and As overpressure (Fig. 2). As

FIG. 3. Plot of the Ga adatom concentration verses time for five substrate temperatures ranging from 300 to 700 °C. The initial change in the Ga concentration is due to the surface coming to equilibrium at the corresponding temperature. At time equals 10 deposition is started and the surface Ga adatom concentration increases abruptly. After ten time units the deposition is ended and the adatom concentration relaxes to the equilibrium value. The supersaturation σ is listed for each of the growth sequences. Images of the surfaces at three temperatures and times are shown in Fig. 5.

FIG. 4. Plot of the number of step-edge sites verses time for five substrate temperatures ranging from 300 to 700 °C. The initial change in the surface is due to it coming into equilibrium at the corresponding temperature. At time equals 10 deposition is started and the number of step edges increase abruptly. After ten time units the deposition is ended and the surface relaxes an equilibrium configuration. Note that the temperature that produced the smoothest surface was an intermediate temperature, 500 °C.

the substrate temperature increases, so does the Ga adatom concentration. This is because at higher substrate temperatures As atoms are more likely to break away from the GaAs units and desorb from the surface. With a higher As overpressure, the Ga adatom concentration is decreased. This is due to the fact that there are more incoming As atoms to combine with Ga atoms to form GaAs on the surface.

Once the system reaches a steady state, a Ga flux is added to allow GaAs film growth. As stated earlier, the As and Ga fluxes are fixed at a ratio of 10:1. The overall growth rate is determined by the Ga flux as long as there is a sufficient As overpressure. In these simulations, after ten layers of GaAs growth, the Ga flux is turned off, and the surface is allowed to recover at the growth temperature.

The Ga adatom concentration as a function of time for five such simulations is shown in Fig. 3. These simulations are performed at nominal temperatures ranging from 300 to 700 °C. In each case it is seen that the surface Ga adatom concentration during growth is higher than that during equilibrium. This is reasonable and indicates that the system is driven out of equilibrium by the Ga beam flux. The supersaturation σ is a measure of the degree to which the system is out of equilibrium. For the curves in Fig. 3, the supersaturation ranges from 50 for 300 °C to 1.2 for 700 °C. Thus this sequence of curves shows a transition from supersaturated growth at lower substrate temperatures to near equilibrium growth at higher. As expected, Fig. 3 also shows that the Ga adatom concentration drops back to its pre-growth level after some time of recovery for each temperature. This is because the same number of Ga atoms before and after growth are needed to maintain equilibrium between the surface and the As flux.

Figure 4 shows the number of step-edge sites on the surface as a function of time for the same temperatures as in
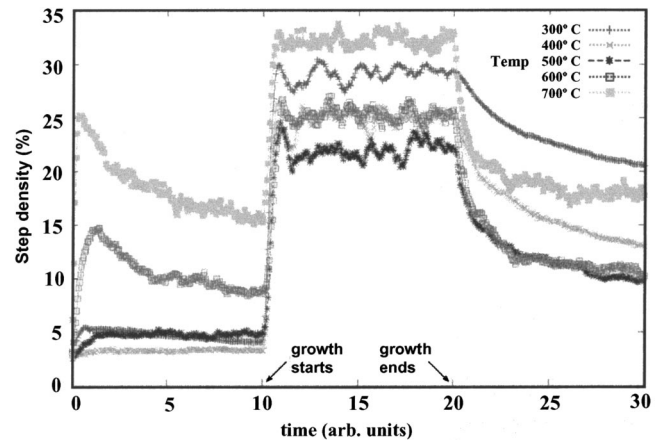
Fig. 3. In the simulation, a site is a step edge if it has two or three nearest neighbors (not counting the bond to the site below). The number of step-edge sites is a measure of the roughness of the surface, i.e., the higher the number, the rougher the surface. Before the growth (Ga flux) is initiated, the equilibrium number of step-edge sites is a monotonically increasing function of the substrate temperature. The surface at 300 °C has the lowest number of step-edge sites while the surface at 700 °C has the highest. This is not true during growth. While depositing Ga it is not the lowest temperature, 300 °C, surface, but the intermediate 500 °C substrate that has the fewest step-edge sites. This is an important result unique to this two-component simulation; smoother growth occurs at mid-range substrate temperatures.

The explanation for this result is straightforward. During growth, there are more impinging As atoms adsorbed to the surface because of the increased concentration of Ga atoms on the surface. At lower temperatures, there is not enough GaAs decomposition and As desorption to smooth out the increased roughness due to kinetic limitations. On the other hand, at high temperatures there is too much GaAs decomposition and As desorption causing a thermal roughening. Between these low and high temperature extremes, there is an optimum at which the GaAs molecules that protrude above the smooth surface have enough energy to decompose; yet the GaAs molecules that form the flat surface are stable. At this intermediate temperature the simulation results in optimum layer-by-layer growth.

Snapshots of the model surfaces at 300, 500, and 700 °C at three different stages of the simulation: immediately before growth starts (surface in equilibrium with the As flux), immediately before the growth ends (after about ten layers deposited), and at the end of the simulation (after recovery at the growth temperature) are shown in Fig. 5. It is clear that the surface grown at 500 °C is smoother than those grown at
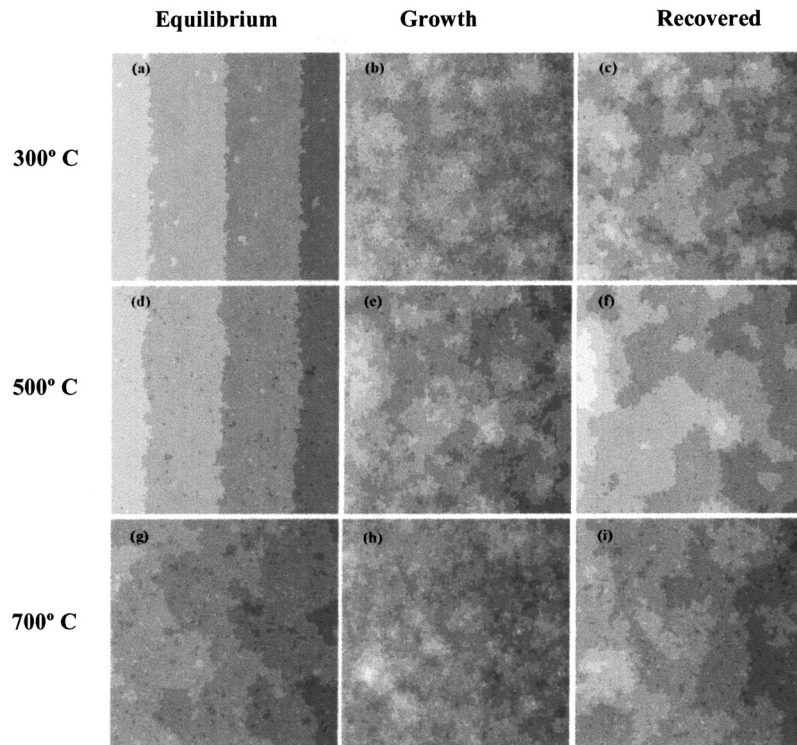
Equilibrium **Growth** **Recovered**

FIG. 5. Set of simulated images of GaAs surface grown at three temperatures: 300, 500, and 700 °C. The images correspond to temperature and time points from Figs. 3 and 4. The images progress left to right as a function of time. The left most column is the surface after thermal equilibration ($t=9$), the center column shows the surfaces at the end of the deposition ($t=20$), and the right column shows the surface after recovery ($t=30$).

300 and 700 °C. This is due to the reasons discussed above. After recovery, the 500 °C surface becomes even smoother, whereas the 300 and 700 °C surfaces are not improved as much. This is because at 300 °C there is not enough As desorption to smooth out the surface, while at 700 °C there is too much thermal roughening. The 500 °C surface has the correct kinetics for smooth growth: not too much thermal roughening and yet enough thermal energy for GaAs decomposition and As desorption to smooth asperities formed by kinetic limitations of diffusion.

These results can be compared to a conventional one-component model for GaAs. In such a model there is no optimal temperature for smooth growth. The higher the temperature of the substrate, the smoother the growth in the simulation. This follows from the increased diffusion length of the adatoms and the very high energy of binding at step edges used in these simulations. (In a one-component model if the binding energy of the step attachment is lowered so that adatom attachment becomes reversible all agreement with experiment is lost. Therefore the one-component models for MBE are parametrized to be in the highly kinetically limited regime.) Thus the experimentally verified existence of an optimum substrate temperature for smooth growth is much more successfully modeled by the two-component simulation than by the single species model.

It is instructive to compare other experimental findings to the different types of simulations to determine their relative strengths and weaknesses. Consider a thermal quenching experiment with no growth occurring. In a one-component simulation if two surfaces are quenched from different initial high temperatures to a low temperature, the surface with the higher initial temperature will be smoother than the other because of its higher surface atom mobility. This is not consistent with experimental findings, where the opposite is

found.[16] However, in the two-component model the number of free Ga atoms needed to maintain equilibrium between the surface and the As flux is smaller after quenching. Where do the excess Ga atoms on the surface go? They combine with the impinging As atoms to form GaAs islands. For the same final temperature, the substrate with the higher initial temperature (with more Ga adatoms) will have the rougher surface.

Simulations of such quenching experiments have been performed. Figure 6 shows the simulation surfaces with initial temperatures 500, 600, and 700 °C at three different stages: right before quenching (equilibrated surface), right after quenching to 400 °C, and at the end of the simulation (after recovery at 400 °C). The surface with the 500 °C initial temperature is the smoothest at the end of the run. These results are in excellent agreement with experiment and can be directly compared to the STM images presented in Ref. 16.

Another interesting experiment to examine was conducted by Deluca and Barnett. They used an ion beam scattering technique to measure two-dimensional islands during MBE growth.[17] In these experiments they measured the specularly reflected ion beam during a short growth (1.5 monolayers) and surface recovery. They reasoned that the decrease in specularly reflected ion current during recovery is directly proportional to the decrease in surface roughness. There are principally two sources for ion scattering, scattering from Ga adatoms and from the step edges of islands. (The scattering of the ion beam can be treated, to a much higher degree than electrons, within a kinematical approximation.) These two different origins for the scattering, Ga adatom and step edge, possess very different relaxation dynamics. The Ga adatom population decreases exponentially during the recovery;[18] and the step-edge length, assuming Ostwald ripening, can be

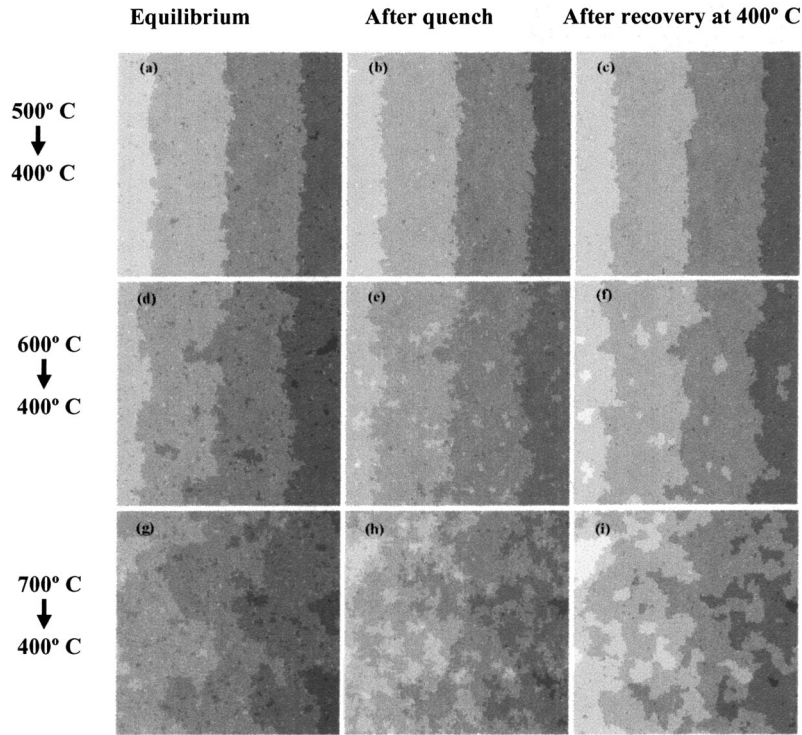**Equilibrium**   **After quench**   **After recovery at 400° C**

FIG. 6. Simulated images of the progression of GaAs surfaces quenched from an elevated initial temperature to 400 °C. The quenching process allows the adatoms to coalesce and form islands. During recovery the island edges smoothen.

described by a power-law expression.[19] Using these results, Deluca and Barnett fitted the specularly reflected ion current with the following expression:[17]

$$I_S = I_0[1 - C_1 \exp(-t/\tau_A) - C_2(C_3 + t^m)^{-1/2}], \quad (5)$$

where I_S is the specularly reflected ion current, I_0 is the ion current prior to growth, C_1, C_2, and C_3 are independent fitting parameters, t is time, τ_A is the lifetime of the Ga adatoms prior to capture at step edges, $m = \frac{2}{3}$ for coarsening kinetics limited by surface diffusion, and $m = 1$ for coarsening kinetics limited by attachment to steps.[13] Fits from the experiments allowed determination of the parameters and the power-law dependence for step-edge relaxation.[17,19]

Turning away from the experiment and back to the two-component model, we have simulated these short growth and recovery experiments. In the simulation, we grow 1.5 layers of GaAs on a 500 °C equilibrated surface. After turning off the Ga flux the surface is was allowed to recover at the growth temperature. Unlike the experiment, the simulation can track the adatom concentration and the surface step density separately. Figure 7 shows the Ga adatom concentration as a function of time during recovery. The fitted curve is an exponential decay. This curve shows that the growth flux increased the Ga adatom concentration by a factor of 2.3 over its equilibrium value. When the deposition was stopped the relaxation time for the adatoms to return to their equilibrium value was 0.32 time units.

Figure 8 shows the step density during recovery and the corresponding fit to a power law. The power-law fit is the third term in Eq. (5). In the fit, $m = 0.4$. This is not consistent with either coarsening kinetics limited by surface diffusion (for which $m = \frac{2}{3}$), or with coarsening kinetics limited by attachment (for which $m = 1$).[20] Note that even in the simu-

lation MBE growth is a series of complicated processes and the use of an Ostwald ripening model is very much a simplification.

Nevertheless, the simulation naturally produces two time constants for recovery, just as observed experimentally: a short time associated with relaxation of the Ga adatom concentration, and a longer time associated with the reduction in step density. The reduction in Ga adatom concentration is a result of As atoms in the flux impinging on the surface and combining with Ga atoms. The exponential dependence on time is natural. The decrease in step density is the result of
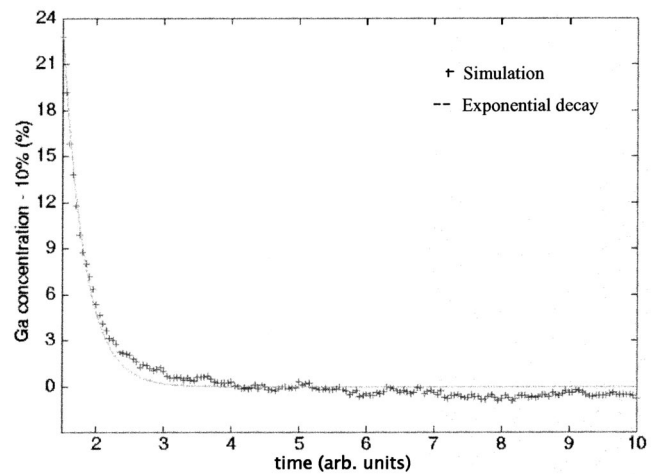

FIG. 7. Plot of the change in the Ga adatom concentration (crosses) during recovery from a 1.5 layer growth at 500 °C. (A 10% thermal background has been subtracted.) The exponential fit (dashes) describes the decay of Ga adatom concentration after growth is terminated. The exponential is the second term in Eq. (5). It gives a time constant for Ga adatom decay (τ_A) of 0.32.
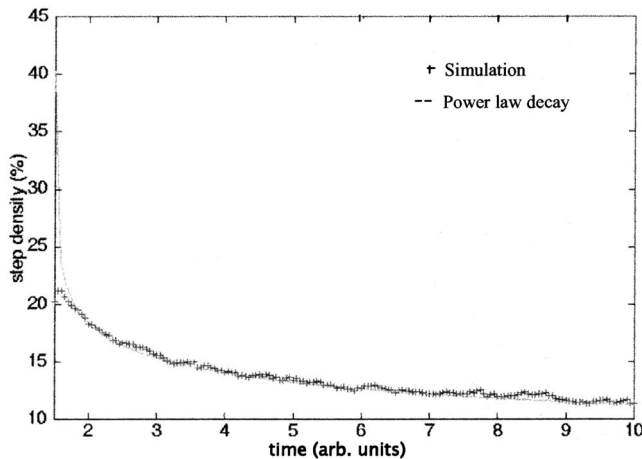
FIG. 8. Plot of the change in the step density (crosses) during recovery from a 1.5 layer growth at 500 °C. The power-law fit (dashes) describes the decay of the step density after growth is terminated. The power law is the third term in Eq. (5). The best fit has a power-law decay with the exponent equal to 0.4.

island coalescence produced by the breakup of GaAs units, the Ga entering the surface probability distribution and then readsorption of As in a different location. It is important to note that these surface dynamics are quite different than simple diffusion of a single species and explains the breakdown of the power-law predictions for ripening.

The most commonly used *in situ* diagnostic for molecular beam epitaxy is reflection high-energy electron diffraction (RHEED). During deposition of GaAs when the growth mode is layer by layer, the intensity of the specularly reflected spot of the reflection pattern oscillates.[21,22] The two-component model is capable of reproducing RHEED oscillations in the form of oscillations in Ga adatom concentration and step density. Although it is difficult to establish a direct relationship between RHEED oscillations and the Ga adatom concentration and/or step density, these quantities are good indications of surface roughness, i.e., the higher the Ga adatom concentration and step density, the rougher the surface. Figure 9(a) shows the Ga adatom concentration before and during growth on the terrace of a 256×256 square lattice with periodic boundary conditions. The growth rate is approximately one layer per time unit. Figure 9(b) is an enlargment of the rectangle in Fig. 9(a). Oscillations are clearly visible after the initiation of growth. Similarly, Fig. 10(a) shows the step density before and during growth. Figure 10(b) is an enlargement of the rectangle in Fig. 10(a). Both curves in Figs. 9(b) and 10(b) show oscillations with a period of ~1 time unit, which is consistent with the growth rate. As growth continues, the amplitudes of Ga adatom concentration and step density oscillations decrease, and the curves appear less periodic. This decay is similar to experiments, and is caused by the statistical disorder in the growth surface that accumulates over time. Eventually the surface arrives at a dynamical steady state and the oscillations vanish.[23]

It is instructive to examine the actual simulation surfaces from which the data in Figs. 9 and 10 was derived. Figure 11 shows the simulation surfaces corresponding to the enumer-
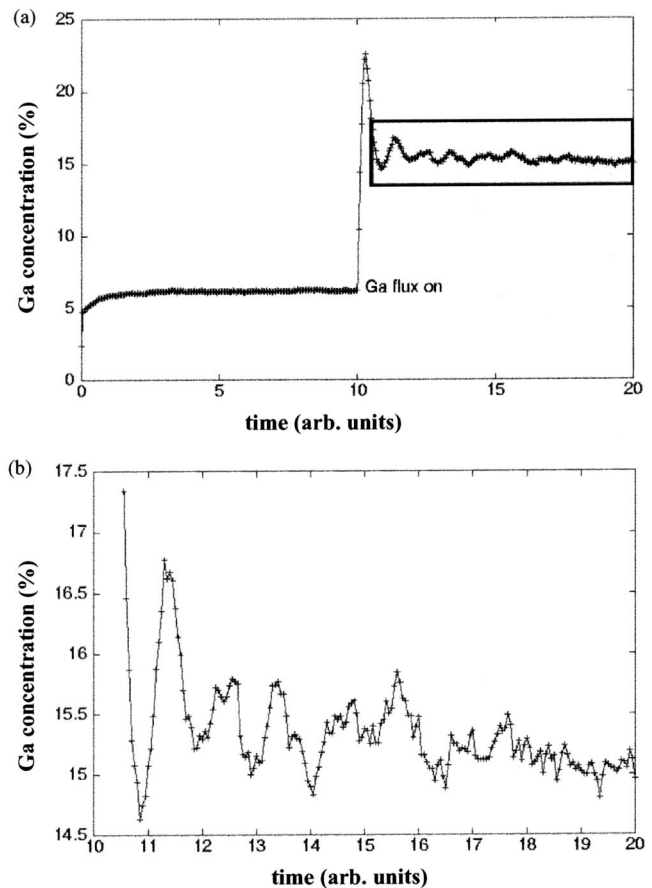

FIG. 9. Ga adatom concentration for a simulation performed at 500 °C. The lattice size was 256×256. For the first ten time units the system is coming to equilibrium. At time equals 10 deposition is started with a growth rate of 1. (a) shows the adatom concentration for the entire simulation. The inset in (a) is shown magnified as (b). Oscillations with a period of one time unit are seen to decay during the first ten layers of growth.

ated peaks and valleys of the step density curve in Fig. 10(b). Surfaces (1) and (3) correspond to step density maximum (indicating a rough surface), and show 50% coverage; surfaces (2) and (4) correspond to step density minimums (indicating a smooth surface), and are close to being fully covered.

Conventionally, the disappearance of RHEED oscillations as a function of increasing temperatures is explained by a transition from "layer-by-layer" to "step-flow" growth mode. During step-flow growth, the adatoms acquire enough thermal energy to diffuse a distance larger than the typical terrace size allowing the particles to attach to step edges. This leads to the image of a train of steps moving across the sample in unison. Thus statistically the surface does not change and therefore there are no RHEED oscillations.

The two-component model provides an alternative explanation for the disappearance of RHEED oscillations during higher temperature MBE growth. As shown earlier, there is a thermal background of Ga adatoms on the surface during equilibrium. With the initiation of growth, the added Ga atoms from the source are a perturbation to the equilibrium
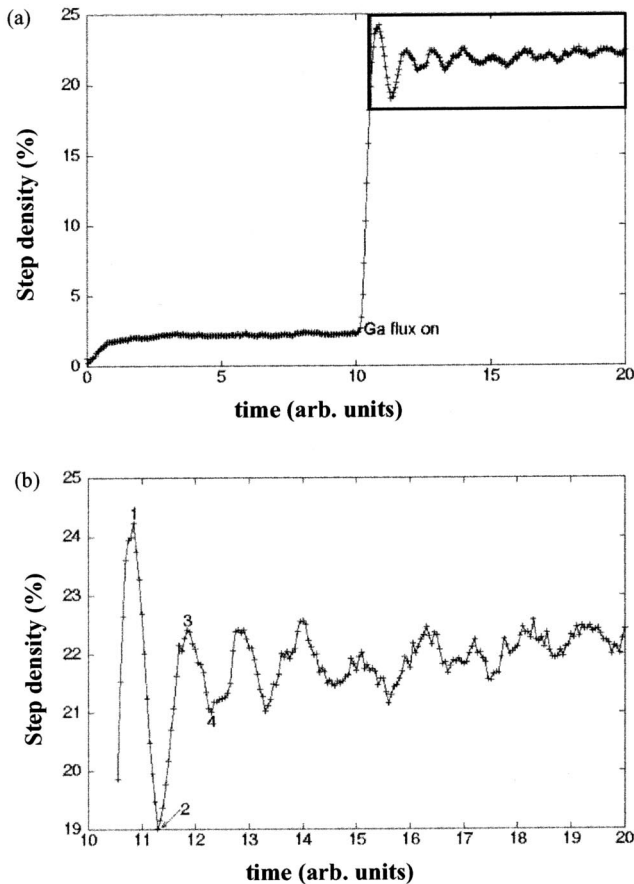
FIG. 10. Step density for a simulation performed at 500 °C. The lattice size was 256×256. For the first ten time units the system is coming to equilibrium. At time equals 10 growth is started with a growth rate of 1. (a) shows the step density for the entire simulation. The inset in (a) is shown magnified as (b). Oscillations with a period of one time unit are seen to decay during the first ten layers of growth. Note that the step density oscillations are slightly delayed with respect to the adatom concentration oscillations shown in Fig. 9. The location marked 1, 2, 3, 4 refer to the surfaces shown in Fig. 11.

FIG. 11. Simulation surfaces (256×256 lattice sites) corresponding to the marked peaks and valleys of the step density curve in Fig. 10(b). (1) and (3) correspond to maximum step densities and rough surfaces. (2) and (4) correspond to minimum step densities and smooth surfaces.

adatom concentration (this is seen as the large jump in concentration in Fig. 3). As the substrate temperature is elevated, the equilibrium concentration of adatoms rapidly increases (Figs. 2 and 3). This leads to a relative decrease in the magnitude of the perturbation upon initiation of growth and the amplitude of the oscillations of the Ga adatom concentration is much reduced and eventually vanishes. In other words, the perturbations due to the Ga flux are much smaller for higher substrate temperatures and as a result, the RHEED oscillations disappear.

This explanation does not invoke a changing of the diffusion length for the Ga adatoms. Remember that in these simulations the effective diffusion length for Ga adatoms is very large, i.e., the size of the system, and this is true for *all* substrate temperatures. Therefore the elimination of RHEED oscillations for this model does not depend on a transition to "step-flow" growth; it depends on a transition to near-equilibrium growth. If samples with different vicinal surface or plan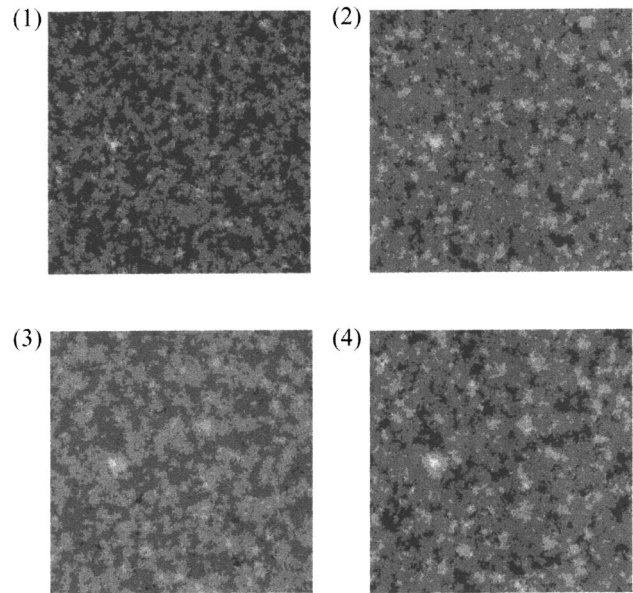es are examined then experimentally it is found that with larger miscut (smaller average terrace size) the transition temperature for the disappearance of RHEED oscillations is lowered.[21] This is explained by the two-component model in noting that with larger miscut (larger number of steps) the equilibrium concentration of adatoms on the surface will be larger, compared with a singular surface at the same temperature. Therefore the crossover point to where no oscillations are observable will occur at lower temperatures for higher miscut. To provide truly quantitative comparison with experiment would require a detailed knowledge of the surface chemistry and reaction dynamics. It is thought that the reaction for MBE with solid source As requires two As_4 molecules and two Ga adatoms to form GaAs. Thermal dissociation of GaAs from the surface produces As_2 vapor.

In conclusion, this two-component model of MBE growth incorporates the high surface mobility of one species and the volatility of the other. The model allows us to consider the Ga adatom concentration before, during, and after growth. Simulations indicate that at typical growth conditions, the surface Ga adatom concentration may be high even before the Ga flux is turned on. If this is so, during growth the Ga flux acts as a *small* perturbation to the Ga atoms on the surface; therefore the supersaturation is low enough to call the growth "near equilibrium." The simulations also show that near equilibrium (low supersaturation) growth produces smoother surfaces, as long as the surfaces are not kinetically limited.

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