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Correlation between island-formation kinetics, surface roughening, and RHEED oscillation damping during GaAs homoepitaxy

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The basic mechanisms controlling surface roughening and its correlation to the damping behavior of reflection high energy electron diffraction (RHEED) intensity oscillations are studied. The experimental observations are compared to results of Monte Carlo simulations of atomistic processes on the growing surface, such as surface diffusion of both Ga atoms as well as of slowly migrating GaAs molecules. With the simulation model, quantitative reproduction of the temperature and III/V flux-ratio dependence of RHEED oscillation damping by simulated step density oscillations is demonstrated. Systematic simulation studies are performed to examine the relationship between oscillation decay, surface roughening, nucleation rate, surface kinetics, and the prevalent growth conditions. The nucleation rate is identified as the central quantity which determines both the oscillation damping as well as the surface roughening. [S0163-1829(97)03844-7]

I. INTRODUCTION

Molecular beam epitaxy (MBE) has been well established as a production tool for devices such as lasers[1] or high mobility transistors.[2] Furthermore, much effort has been invested in optimizing low-dimensional structures for basic research as, for example, to study the electronic properties of two-, one-, and zero-dimensional systems, where MBE often acts as a basic technology.[3] For optimum performance of such structures, the crystalline quality as well as the morphological sharpness of the interfaces are of primary importance. Roughness leads to broadening of quantization energies and to increased carrier scattering in active regions, which lowers electron and hole mobilities. As interface smoothness is ultimately determined by that of the growing surface, it is technologically important to understand the mechanisms controlling morphology.[4] Basic atomistic processes determining the crystal growth and, thus, the morphological evolution are arrival, desorption, reaction, dissociation, surface diffusion, step edge attachment as well as detachment, and nucleation. These kinetic processes are controlled by the prevalent growth parameters, of which the most important are the growth temperature, the growth rate, and the flux ratio in the case of compound semiconductors.

To obtain stochiometric incorporation of gallium and arsenic, arsenic must be present in excess. This is due to the condensation behavior of both species. The sticking coefficient of gallium turns out to be unity at usual substrate temperatures, whereas arsenic only sticks via reaction with gallium. If the arsenic flux is too low, excessive gallium will be incorporated resulting in unstoichiometric films.[5] On the other hand, if arsenic flux is too high, surface roughness increased significantly[6] and photoluminescense spectra indicated enhanced formation of group-III vacancies.[7] The substrate temperature during GaAs homoepitaxy strongly determines the mobility of the diffusing species[8] as well as the surface roughness.[6]

Measurements of the reflection high energy electron diffraction (RHEED) intensity showed an increasing oscillation damping, when the arsenic flux is increased[5,9,10] as well as when the growth temperature is decreased.[10] A qualitative model for the oscillating behavior of the RHEED specular beam was proposed by Neave *et al.*,[11,12] but the origin of oscillation decay is not yet identified unequivocally. van der Wagt *et al.*[13] attribute oscillation damping to macroscopic growth rate variations due to gallium flux nonuniformities. Since in our experiments we use a constant gallium flux, this effect is expected not to be important. Several authors proposed that RHEED oscillation decay is due to the evolution of the surface towards a steady state with time-independent step density.[4,14] Dobson *et al.*[14] addressed the surface-diffusion length as to determine this steady state step density. However, yet, no definite conclusions can be drawn. Therefore, the question still remains, how the arsenic population on the surface as well as the substrate temperature influences the RHEED oscillation decay and the roughness evolution.

Lewis *et al.*[9] point out that with increasing arsenic flux the probability increases that the diffusing Ga atoms are reactively incorporated before they reach a surface step. As an extension to this model, we assume a three-step incorporation mechanism: (1) reaction of mobile Ga atoms with As to form strongly bound GaAs molecules, (2) surface diffusion of these GaAs molecules, but with significantly higher diffusion barrier than of diffusing Ga atoms, (3) incorporation of Ga atoms and GaAs molecules in step-, kink-, and 2D island positions. Previous simulation results give strong evidence for mobility of GaAs molecules. A computer simulation of GaAs homoepitaxy, which is based on the above assumptions, was succesfully used to reproduce experimental data of the flux-ratio dependent As_4 sticking coefficient, the time evolution during surface recovery after growth stops, and surface topologies.[15] In this work, we focus on the influence of the flux ratio and the growth temperature on surface kinetics with emphasis on RHEED oscillation decay and roughness evolution.

II. EXPERIMENTAL SETUP

Most of the growth experiments are performed in an especially designed MBE system which is described in detail in Ref. 16. As a peculiarity, a quadrupole mass spectrometer allows precise control of the As_4 flux and of the Ga/As flux ratio. Some additional films are grown in a second MBE system (Riber 32 P) in order to check the influence of the growth environment. Growth is carried out on nominally planar (001) GaAs substrates. After thermal oxide removal under arsenic pressure, a 300 nm thick buffer layer is grown in order to smooth the surface. The subsequent layers are grown at substrate temperatures between 550 °C and 680 °C. The gallium flux is kept constant corresponding to a low temperature growth speed of 0.3 monolayers (ML) per second, and the arsenic flux is varied to achieve flux ratios $J_{\mathrm{Ga}}/J_{\mathrm{As}_4}$ from 0.4 up to 1.4. During growth, the peak intensity of the RHEED specular beam intensity along the [110] azimuth is recorded by a sensitive CCD camera in combination with a frame grabber and a personal computer for image processing. The diffraction conditions are chosen to correspond to in-phase diffraction, for which reflection from adjacent layers add constructively and the kinematic theory yields a constant intensity. Thus, the modulation of the specular beam intensity during growth can be traced back to variations of the surface step density.[17]

III. SIMULATION MODEL

The simulation used here closely resembles a model presented by Clarke and Vvedensky.[18] With this model, Shitara *et al.* successfully simulated epitaxial growth on GaAs (001) surfaces.[17] Basic assumptions are a cubic lattice, a solid-on-solid configuration[19] (that means no overhangs and no vacancies), random deposition as well as surface migration of gallium. As an extension to this model, our simulation includes surface kinetics of Ga and As atoms, and of GaAs molecules. The incorporation of arsenic is modeled by a process in which every impinging As_4 molecule dissociates into atoms. Two As atoms desorb resulting in a maximum As_4 sticking coefficient of 0.5.[20] The two remaining As atoms perform a random search for a free gallium atom and either react to GaAs or desorb as well.

After impinging, the adsorbants move randomly by isotropic nearest-neighbor hopping. The hopping rate of an adsorbant is given by

$$k(T)=k_d \exp\left(-\frac{E_{\mathrm{dif}}}{k_B T}\right), \tag{1}$$

where $k_d=1\times10^{13}\ \mathrm{s}^{-1}$ is assumed as vibrational frequency for diffusion, k_B is Boltzmann's constant, T is the substrate temperature, and the surface-diffusion barrier is[17]

$$E_{\mathrm{dif}}=E_S+nE_N, \tag{2}$$

with the surface contribution E_S, the number of nearest-neighbors $n=0\ldots4$, and the nearest-neighbor contribution E_N.

Surface diffusion of Ga atoms, As atoms, and GaAs molecules is taken into account. As a simplification, the migration of GaAs molecules is modeled by the same mechanism as for diffusion of single atoms. Just the diffusion barrier is supposed to be significantly higher.[15] The surface contributions E_S for Ga, As, and GaAs are denoted as $E_{\mathrm{S,Ga}}$, $E_{\mathrm{S,As}}$, and $E_{\mathrm{S,GaAs}}$, respectively. To reduce the number of simulation parameters, the same value of the nearest-neighbor contribution E_N is used for Ga atoms and GaAs molecules. Arsenic migration is simulated in a special manner. Due to the short lifetime of arsenic on the surface,[21] no arsenic-arsenic interactions are considered. Thus, every migrating As atom can be treated separately. Interaction of As atoms with GaAs molecules is neglected, whereas interaction with Ga atoms results in reaction to GaAs molecules. Arsenic atoms may desorb when they find no Ga atom within their surface lifetime. Desorption of Ga atoms and GaAs molecules is neglected, since experiments showed a gallium desorption rate of zero up to growth temperatures of about 650 °C.[22]

The characteristic quantity to describe migration of As atoms is the surface-diffusion length up to desorption: $\lambda_{\mathrm{As}}=\sqrt{D_s\tau_{\mathrm{des}}}$, with the surface-diffusion coefficient $D_s=(\frac{1}{4})(a_0)^2k_d\exp(-E_{\mathrm{dif}}/k_BT)$, the distance of a nearest-neighbor hopping a_0, the lifetime up to desorption $\tau_{\mathrm{des}}=(1/k_a)\exp(E_{\mathrm{des}}/k_BT)$, the vibrational frequency k_a for desorption, and the activation energy for desorption E_{des}. With the above assumptions we get

$$\lambda_{\mathrm{As}}=\left(\frac{a_0}{2}\right)\exp\left(\frac{dE_{\mathrm{As}}}{2k_BT}\right), \tag{3}$$

with $dE_{\mathrm{As}}=E_{\mathrm{des}}-E_{\mathrm{dif}}$ and $\sqrt{k_d/k_a}\approx1$.

The simulation is thus characterized by four parameters: $E_{\mathrm{S,Ga}}$, $E_{\mathrm{S,GaAs}}$, E_N, and dE_{As}. These quantities are to be regarded as effective diffusion barriers that incorporate in an average way local fluctuations, fast processes not included explicitly, and other factors such as surface reconstruction.[17] As described in a former paper,[15] detailed comparisons to RHEED measurements are made to determine the model parameters: $E_{\mathrm{S,Ga}}=1.39$ eV, $E_{\mathrm{S,GaAs}}=1.74$ eV, $E_N=0.435$ eV, and $dE_{\mathrm{As}}=0.868$ eV. Due to the [110] azimuth of the RHEED experiments, these numerical values mainly reflect surface migration perpendicular to the orientation of arsenic dimers and bonds to gallium terminated steps, respectively.[23]

In the simulation runs presented here, the temperature is varied from 550 °C up to 650 °C. The growth rate $G=0.3$ ML/s is fixed by the constant gallium flux. The flux ratio $J_{\mathrm{Ga}}/J_{\mathrm{As}_4}$ is varied from 0.4 up to 1.4 by changing the arsenic flux. The simulations are started on a 230×230 matrix with periodic boundary conditions and without initial steps.

IV. RESULTS AND DISCUSSION

A. Damping behavior of RHEED oscillations

According to Shitara *et al.*[17] an approximately linear relationship between the measured intensity I_{sp} of the RHEED specular beam and the simulated surface step density

$$n_{st}=N^{-1}\sum_{x,y}\{[1-\delta(z_{x,y},z_{x+1,y})]\cos\phi +[1-\delta(z_{x,y},z_{x,y+1})]\sin\phi\} \tag{4}$$

can be assumed, where $\delta(x,y)$ is the Kronecker δ function, $z_{x,y}$ is the height at the lattice site (x,y), N is the number of lattice sites, and ϕ is the azimuthal angle ($\phi=0$ at the [110] direction). Thus, simulation runs can be compared directly to real growth processes. An example of the experimentally ob-

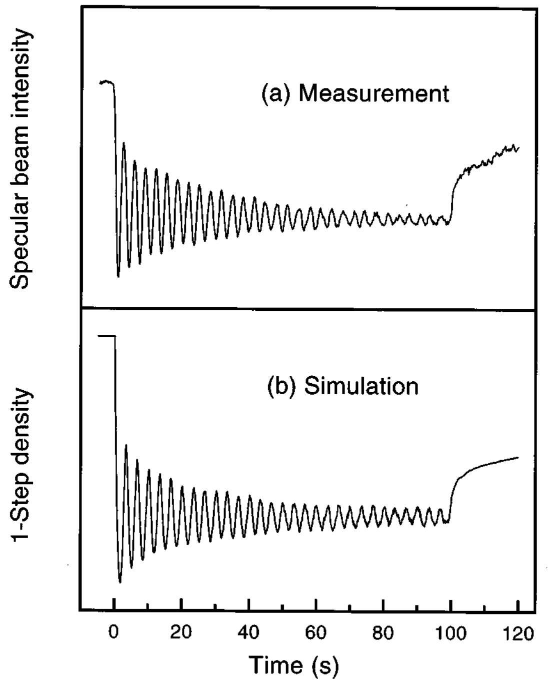

FIG. 1. Typical measured RHEED intensity oscillations (a) and simulated step density oscillations (b), with growth parameters: temperature $T = 600$ °C, growth rate $G = 0.3$ ML/s, flux ratio $J_{\mathrm{Ga}}/J_{\mathrm{As}_4} = 0.65$, and simulation parameters: $E_{\mathrm{S,Ga}} = 1.39$ eV, $E_{\mathrm{S,GaAs}} = 1.74$ eV, $E_N = 0.435$ eV, and $dE_{\mathrm{As}} = 0.868$ eV as described in the text.

served time evolution of the RHEED specular beam intensity during growth and subsequent surface recovery is plotted in Fig. 1(a). The simulation of the corresponding surface step density is shown in Fig. 1(b). As can be seen, simulation and real growth yield nearly identical oscillations. A model for the oscillating intensity of the RHEED specular beam was proposed by Neave *et al.*[11,12] One main difference between real growth and simulation is noticeable: the initial fast decrease after growth starts is smaller for RHEED oscillations than in the simulated step densities. This may reflect a change of reconstruction during growth which is not included in our simulation model.

As a measure of surface evolution, the damping behavior of RHEED intensity oscillations is analyzed quantitatively as a function of flux ratio and temperature. The oscillation damping is characterized by a damping constant ξ given as the number of deposited monolayers (ML) at which the oscillation amplitude is reduced to the fraction of $1/e$ of the initial amplitude. As shown in Fig. 2(a), a decrease of the As_4 flux results in a reduction of the RHEED oscillation damping with a minimum decay at a flux ratio $J_{\mathrm{Ga}}/J_{\mathrm{As}_4}$ of about 1.1. Growth at higher flux ratios yields strongly damped oscillations. A similar behavior is found when the growth temperature is increased [Fig. 2(b)]. Up to about 630 °C, the RHEED oscillation damping is reduced, whereas at higher temperatures, the damping increases again. To check the influence of the growth environment on the RHEED measurements, the temperature-dependent values of the oscillation damping constant are compared to those, which are determined with a second MBE system under identical diffraction and growth conditions. We find agreement within ±6% indicating that RHEED oscillation damping is controlled only by the respective growth conditions.

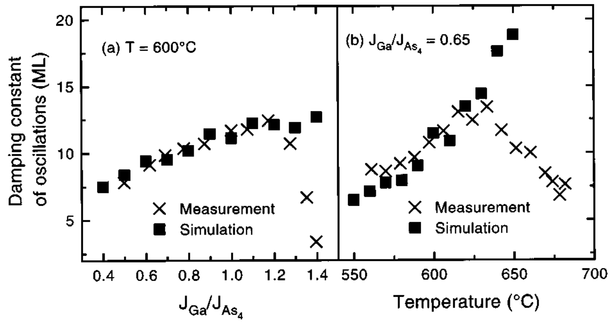

FIG. 2. Damping constant of measured RHEED intensity oscillations as well as of simulated step density oscillations dependent on the flux ratio and on growth temperature. The damping constant is given by the number of monolayers (ML) which can be grown until the oscillation amplitude is reduced to the value $1/e$ of the initial amplitude.

In Fig. 2(a) together with RHEED data, we depict the damping behavior of simulated step density oscillations. Quantitative agreement between real growth and simulation is obtained up to flux ratios $J_{\mathrm{Ga}}/J_{\mathrm{As}_4}$ of about 1.1 and up to growth temperatures of about 630 °C. In this regime, our simulation and the chosen parameters appear to realistically model the surface morphology. Therefore, the simulation provides access to atomic scale processes, which are difficult to deduce from experiments alone.

However, for values of $J_{\mathrm{Ga}}/J_{\mathrm{As}_4} > 1.1$ or temperatures > 630 °C, we find deviations between experiment and simulation. Excessively strong RHEED oscillation damping at $J_{\mathrm{Ga}}/J_{\mathrm{As}_4} > 1.1$ is attributed to formation of three-dimensional gallium droplets.[5] Since this is not implemented in our model, no such damping is observed during simulation runs. Furthermore, at growth temperatures higher than 630 °C, RHEED patterns indicate a change of surface reconstruction from (2×4) to (3×1). The related modification of energy barriers for surface diffusion seems to result in an increasing RHEED oscillation decay. Our simulation parameters only account for a (2×4) reconstructed surface and, thus, the experimental behavior is not found in simulated step densities.

B. Formation of two-dimensional growth islands

The flux ratio of impinging Ga atoms and As_4 molecules directly controls the ratio of mobile Ga atoms and GaAs molecules $n_{\mathrm{Ga}}/n_{\mathrm{GaAs}}$ on the surface. From the simulation, an approximately linear dependence is found, as demonstrated in Fig. 3(a). Figure 3(b) shows the temperature dependence of $n_{\mathrm{Ga}}/n_{\mathrm{GaAs}}$. At higher temperatures, the rate of formation of GaAs molecules is reduced, due to the shorter surface-diffusion length up to desorption of As atoms [see Eq. (3)]. To characterize the surface mobility, the average number of surface hops h_s per impinging Ga atom is determined. Hops of both Ga atoms and GaAs molecules are taken into account. In Fig. 3(c), a linear increase of h_s is observed as function of flux ratio, which is attributed to the increasing

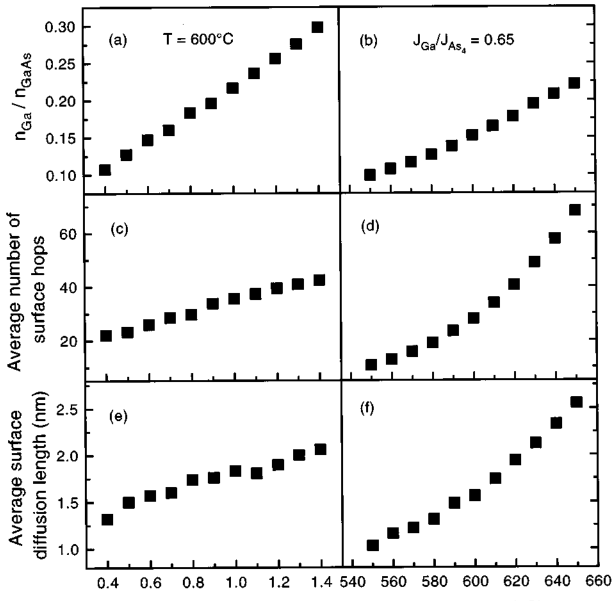

FIG. 3. Simulated steady-state ratio $n_{\rm Ga}/n_{\rm GaAs}$ of mobile Ga atoms and GaAs molecules on the surface (a), (b), average number of surface hops h_s per impinging Ga atom (c), (d), and average surface-diffusion length $\bar{\lambda}$ up to incorporation (e), (f) versus flux ratio and temperature.

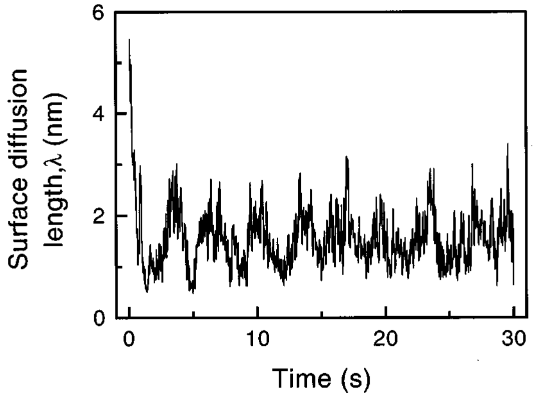

FIG. 4. Typical evolution of the surface-diffusion length λ up to incorporation during growth with parameters as indicated in Fig. 1.

number of mobile Ga atoms. At 600 °C, a single Ga atom performs approximately a hundred times more hops per unit of time than the GaAs molecule. Variations of the growth temperature [Fig. 3(d)] affect h_s by two mechanisms. First, the surface hopping rate $k(T)$ is directly determined by the temperature as is indicated in Eq. (1), and, second, the temperature dependence of $n_{\rm Ga}/n_{\rm GaAs}$ yields an additional contribution. As a consequence, a correct model of the temperature dependence of growth kinetics must include the reaction of gallium and arsenic to GaAs molecules.

The surface-diffusion length λ up to incorporation is a very important quantity which characterizes the surface kinetics. To determine values of λ from simulation runs, single diffusing particles are traced from the point of impinging towards the point of incorporation. The straight distance between these points gives the surface-diffusion length. A particle is assumed as incorporated, when the lifetime at a specific site is longer than the time for deposition of one monolayer. In the temperature range discussed here, this is the case for Ga atoms with at least three nearest neighbors and GaAs molecules with two nearest neighbors. During simulation runs, oscillations of λ are observed with frequency as for the step density oscillations (Fig. 4). From these oscillating values of λ, the average surface-diffusion length $\bar{\lambda}$ is calculated. The dependence of $\bar{\lambda}$ on the flux ratio [Fig. 3(e)] and on temperature [Fig. 3(f)] closely resembles that of h_s. As for h_s, the flux ratio influences $\bar{\lambda}$ by the ratio $n_{\rm Ga}/n_{\rm GaAs}$, and the temperature affects $\bar{\lambda}$ by both the hopping rate $k(T)$ as well as by $n_{\rm Ga}/n_{\rm GaAs}$.

Ohta *et al.*[24] experimentally determined the surface-diffusion coefficient along [110] and [$1\bar{1}0$] direction from the growth mode transition between two-dimensional nucleation and step flow growth. At 600 °C and $J_{\rm Ga}/J_{\rm As_4}=0.67$ they found a diffusion coefficient of about 8×10^{-14} cm^2 s^{-1} along [110], which corresponds to a surface-diffusion length of 6 nm. From the simulation, we get an average diffusion length of 1.6 nm at the above growth conditions. To discuss these different values, the respective growth mode must be considered. Ohta *et al.* deduced the surface diffusion length during step flow growth on misoriented surfaces, whereas our simulation reflects growth by two-dimensional nucleation on surfaces without misorientation steps. A misoriented surface can approximately be regarded as a regular arrangement of quasiparallel steps with monolayer height. During step flow growth, no additional growth islands are generated and the surface-diffusion length equals the average step distance.[8] In the case of two-dimensional nucleation, particles are incorporated via periodic formation and coalescense of monolayer height growth islands and the surface-diffusion length is determined by the oscillating density and size of these islands. Therefore, the different growth modes may explain the disagreement between our value of $\bar{\lambda}$ and the value given by Ohta *et al.*

The surface mobility of the diffusing particles strongly determines the rate of formation of two-dimensional growth islands. As a measure of the nucleation rate, the number $n_{i,\theta=0.1}$ of islands at a coverage θ of 0.1 ML (monolayer) is taken, assuming negligible coalescence at this coverage. A reduced rate of island formation is found, when the flux ratio [Fig. 5(a)] or the growth temperature [Fig. 5(b)] is increased. Obviously, the enhancement of surface mobility lowers the nucleation rate. Growth islands are generated via collisions between diffusing particles. With increasing mobility, the probability increases that the particles attach to existing islands resulting in the reduced formation of new islands.

Periodic *in-phase* generation and coalescence of two-dimensional growth islands cause the oscillatory behavior of step density and RHEED specular beam intensity. We attribute the oscillation decay of both quantities to increasing *out-of-phase* nucleation. Out-of-phase nucleation means that growth islands are formed on top of existing islands. These islands remain even on a completed layer and result in the gradual reduction of the specular beam intensity at integer coverages. Therefore, the number $n_{i,\theta=1}$ of islands remaining after completing one monolayer is taken as a measure of

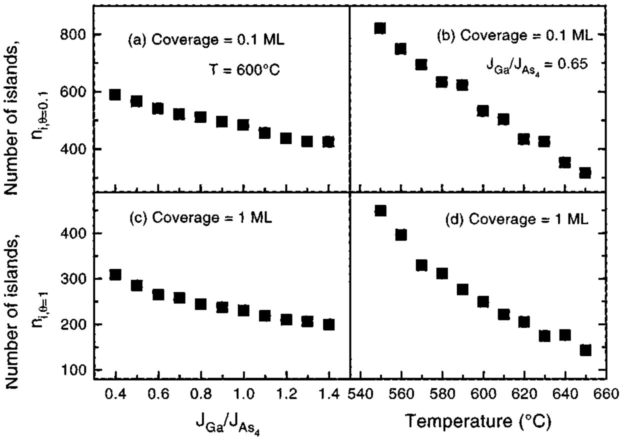

FIG. 5. Number of simulated growth islands with a size of at least two Ga atoms after deposition of 0.1 ML (a), (b), and after deposition of one complete monolayer (c), (d). The data in (a) and (b) are taken as measure of the absolute nucleation rate and the data in (c) and (d) as measure of out-of-phase nucleation.

out-of-phase nucleation. The dependence of $n_{i,\theta=1}$ on the flux ratio and on temperature [Fig. 5(c) and Fig. 5(d)] equals that of $n_{i,\theta=0.1}$, but the absolute number of islands is considerably smaller at $\theta=1$. These results demonstrate that with decreasing flux ratio as well as with decreasing growth temperature the rate of *out-of-phase* nucleation increases. This is in agreement with a reduced surface mobility, a higher nucleation rate, and, thus, more strongly damped oscillations.

C. Evolution of surface roughness

In the following section, the evolution of surface roughness will be discussed. The root mean square (RMS) roughness r_{RMS} of simulated surfaces is calculated using the expression $r_{\mathrm{RMS}}=N^{-1}[\Sigma_{x,y}(z_{x,y}-\overline{z})^2]^{1/2}$ where $z_{x,y}$ is the local height. The simulation yields oscillations of r_{RMS} similar to step density oscillations, as demonstrated in Fig. 6. As for n_{st}, periodic formation and coalescence of two-dimensional growth islands cause the oscillating roughness. But in contrast to step density oscillations with an approximately constant mean value, the average RMS roughness increases during deposition of the first few monolayers. This behavior reflects the different meaning of both quantities. The step density as defined above counts all steps irrespective of the layer, where they are found, whereas r_{RMS} is more sensitive to local variations of the height of the particles. Therefore, the increasing average value of r_{RMS} indicates that an increasing number of layers is involved. Sudijono *et al.*[4] used scanning tunneling microscopy (STM) to study the evolution of surface roughness. They observed no significant increase of roughness during growth. Probably, the small roughness variations found here are below the detection limit of the STM method.

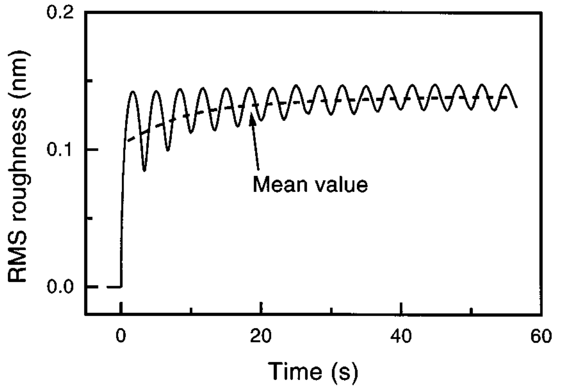

FIG. 6. Typical evolution with time of simulated RMS roughness $r_{\mathrm{RMS}}=N^{-1}[\Sigma_{x,y}(z_{x,y}-\overline{z})^2]^{1/2}$ (with $z_{x,y}$ is the height) during growth with parameters as indicated in Fig. 1. In contrast to the step density evolution, the average RMS roughness increases with deposition time.

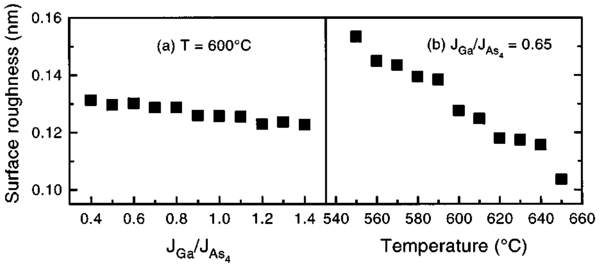

FIG. 7. Simulated surface roughness $r_{\mathrm{RMS},\theta=15}$ after deposition of 15 ML as function of the flux ratio and of temperature.

To quantitatively characterize the surface roughening, we analyze the absolute value of the surface roughness $r_{\mathrm{RMS},\theta=15}$ after deposition of 15 ML. As shown in Fig. 7, the surface roughness decreases with increasing flux ratio as well with increasing growth temperature. This behavior is in qualitative agreement with measurements of Baeta Moreira *et al.*[6] who studied the influence of flux ratio and temperature on the surface morphology with RHEED and microscopic methods. Obviously, the smoothest surfaces are obtained during growth with high surface mobility, which is given either by an increased temperature or by a reduction of the arsenic flux. We would like to note again that we expect our simulation data to represent the real surface evolution only for flux ratios $J_{\mathrm{Ga}}/J_{\mathrm{As}_4}$ up to 1.1 and growth temperatures up to 630 °C.

V. CONCLUSIONS

The damping behavior of measured RHEED and simulated step density oscillations as well as the evolution of surface roughness are studied. We discuss the results in terms of surface mobility and nucleation rate. Using a simulation model which accounts for surface diffusion of both Ga atoms as well as of GaAs molecules, we achieve quantitative reproduction of the flux ratio and temperature-dependent behavior of RHEED oscillation damping by simulated step densities. The parameters used for the simulation are obtained from independent experiments such as the measurement of the surface recovery time after growth stops and measurements of the As_4 sticking coefficient.[15] The high degree of agreement between real growth and simulations as demonstrated here is a further strong indication for surface mobility of GaAs molecules.

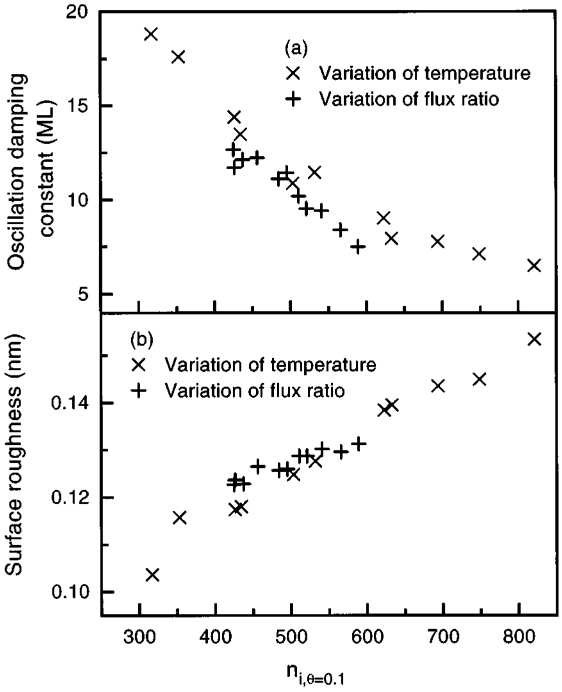

FIG. 8. Simulated damping constant of step density oscillations (a) and surface roughness $r_{\mathrm{RMS},\theta=15}$ (b) dependent on the number of islands $n_{i,\theta=0.1}$ after deposition of 0.1 ML. The values of $n_{i,\theta=0.1}$ are taken as a measure of nucleation rate.

The nucleation rate is identified as the central quantity which determines oscillation damping as well as surface roughening. As shown in Fig. 8(a), a high rate of formation of two-dimensional growth islands corresponds to a high rate of *out-of-phase* nucleation and to strongly damped oscillations. Furthermore, a high nucleation rate causes a larger number of layers which contribute to the surface roughness [Fig. 8(b)]. To present a variety of growth conditions, both the flux ratio and the growth temperature are varied. Weakly damped oscillations as well as smooth surfaces are found to correspond to a small nucleation rate and, very importantly, these correlations are independent on which parameter is varied. Thus, the nucleation rate seems to be more important than the flux ratio or growth temperature. This central importance of the nuclation rate for crystal growth is well known, but the particular properties of the respective materials may differ drastically. Therefore, for a detailed understanding of the nucleation behavior every material needs to be studied separately. Furthermore, we conclude that changes of either the flux ratio or the growth temperature seem to result in the same kind of modification of surface morphology evolution.

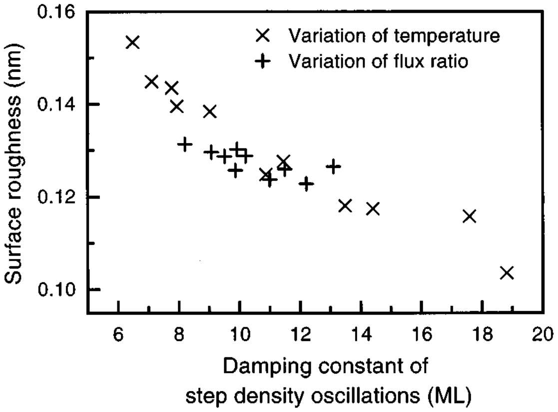

FIG. 9. Simulated surface roughness $r_{\mathrm{RMS},\theta=15}$ versus the damping constant of step density oscillations during variations of both the flux ratio as well as the temperature.

In Fig. 9 the simulated surface roughness $r_{\mathrm{RMS},\theta=15}$ is plotted as a function of the damping constant of step density oscillations. Smooth surfaces correspond to weakly damped oscillations and again the correlation does not depend on which parameter is varied. To obtain a minimum surface roughness during growth of GaAs, high values of the substrate temperature as well as of the flux ratio are required. However, the maximum growth temperature is limited by a change of reconstruction which seems to lower the surface mobility. The maximum flux ratio is also limited, since high flux ratios yield the unstoichiometric incorporation of excessive gallium and formation of gallium droplets.[5]

ACKNOWLEDGMENTS

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[*] Deceased.

[1] See, for example, H. Tanaka and M. Mushiage, J. Cryst. Growth **111**, 1043 (1991).

[2] See, for example, K. Kondo, J. Saito, T. Igarashi, K. Nanbu, and T. Ishikawa, J. Cryst. Growth **95**, 309 (1989).

[3] See, for example, *Nanostructures and Quantum Effects*, edited by H. Sakaki and H. Noge (Springer, Berlin, 1994).

[4] J. Sudijono, M. D. Johnson, M. B. Elowitz, C. W. Snyder, and B. G. Orr, Surf. Sci. **280**, 247 (1993).

[5] Ch. Heyn and M. Harsdorff, J. Cryst. Growth **150**, 117 (1995).

[6] M. V. Baeta Moreira, M. A. Py, and E. Tuncel, J. Cryst. Growth **112**, 14 (1991).

[7] G. Wicks, W. I. Wang, C. E. C. Wood, L. F. Eastman, and L. Rathbun, J. Appl. Phys. **52**, 5792 (1981).

[8] J. H. Neave, P. J. Dobson, B. A. Joyce, and J. Zhang, Appl. Phys. Lett. **100**, 47 (1985).

[9] B. F. Lewis, F. J. Grunthaner, A. Madhukar, T. C. Lee, and R. Fernandez, J. Vac. Sci. Technol. B **3**, 1317 (1985).

[10] J. M. Van Hove, P. R. Pukite, and O. I. Cohen, J. Vac. Sci. Technol. B **3**, 563 (1985).

[11] J. H. Neave, B. A. Joyce, P. J. Dobson, and N. Norton, Appl. Phys. A: Solids Surf. **31**, 1 (1983).

[12] J. H. Neave, B. A. Joyce, and P. J. Dobson, Appl. Phys. A: Solids Surf. **34**, 1 (1984).

[13] J. P. van der Wagt, K. L. Bacher, G. S. Solom, and J. S. Harris, Jr., J. Vac. Sci. Technol. B **10**, 825 (1992).

[14] P. J. Dobson, B. A. Joyce, J. H. Neave, and J. Zhang, J. Cryst. Growth **81**, 1 (1987).

[15] Ch. Heyn and M. Harsdorff, Phys. Rev. B **55**, 7034 (1997).

[16] Ch. Heyn and M. Harsdorff, J. Cryst. Growth **133**, 241 (1993).

[17] T. Shitara, D. D. Vvedensky, M. R. Wilby, J. Zhang, J. H. Neave, and B. A. Joyce, Phys. Rev. B **46**, 6815 (1992).

[18] S. Clarke and D. D. Vvedensky, Phys. Rev. Lett. **58**, 2235 (1987).

[19] J. D. Weeks and G. H. Gilmer, Adv. Chem. Phys. **40**, 157 (1979).

[20] C. T. Foxon and B. A. Joyce, Surf. Sci. **50**, 434 (1975).

[21] C. T. Foxon and B. A. Joyce, Acta Electron. **21**, 139 (1978).

[22] A. H. Kean, C. R. Stanley, M. C. Holland, J. L. Martin, and J. N. Chapman, J. Cryst. Growth **111**, 189 (1991).

[23] T. Shitara, D. D. Vvedensky, M. R. Wilby, J. Zhang, J. H. Neave, and B. A. Joyce, Phys. Rev. B **46**, 6825 (1992).

[24] K. Ohta, T. Kojima, and T. Nakagawa, J. Cryst. Growth **95**, 71 (1989).