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Step-edge barriers on GaAs(001)

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We investigate the growth kinetics on vicinal GaAs(001) surfaces by making detailed comparisons between reflection high-energy electron-diffraction specular intensity measured near in-phase diffraction conditions and the surface step density obtained from simulations of a solid-on-solid model. Only by including a barrier to interlayer transport and a short-range incorporation process of freshly deposited atoms can the simulations be brought into agreement with the measurements both during growth and during post-growth equilibration of the surface.

The roughness of growing surfaces has become one of the main topics in the study of surface processes. One reason for this is the evident importance of minimizing surface roughness for applications. Aside from its relevance for the fabrication of narrow layered structures (quantum wells, lateral superlattices, magnetic multilayers, etc.), surface roughness has recently been suggested as triggering the transition from epitaxial to amorphous growth during Si, Ge, and GaAs homoepitaxy.[1] Provided the roughness can be controlled, this opens up the exciting prospect of low-temperature epitaxy and doping. Another reason for the intense interest in surface roughness is purely theoretical. The roughness of growing surfaces has been observed to exhibit asymptotic dynamical scaling behavior[2] which has led to the classification of growth models into various universality classes.

In a majority of the theoretical studies, the focus has been on the evolution of the surface morphology due to fluctuations in the incoming flux of particles and surface diffusion. Studies of metal epitaxy have revealed the importance of two processes which directly influence the evolution of surface roughness. The first is the way freshly deposited atoms are incorporated into the growing material ("transient mobility,"[3] "downward funneling,"[4] etc.). Such incorporation mechanisms smoothen the growth front and lead to quasi-layer-by-layer growth at temperatures low enough that thermal mobility is negligible.[3–8] The second process is the interlayer transport of material and, in particular, the effect of activation barriers to adatom hopping between layers. These barriers lead to the opposite trend of the incorporation just described, namely, a rapid roughening of the growing *singular* surface.[7–11]

While the effects of an incorporation process and hopping barriers are clearly manifested on metal surfaces, the question arises as to whether these processes might also play an observable role in the growth of semiconductor surfaces. In this paper we address this question by investigating the growth kinetics on vicinal GaAs(001) surfaces. We make direct comparisons between the evolution of the measured reflection high-energy electron-diffraction (RHEED) specular-beam intensity and the step density of simulated surfaces. RHEED is an *in situ* real-time probe of surface morphology and, as numerous studies have shown,[12] can be a sensitive measure of the presence of various surface processes. We find that *quantitative* agreement between the RHEED specular intensity and the step density can be achieved *both* during growth and post-growth equilibration of the surface if our model includes barriers to interlayer transport and an incorporation process for arriving atoms.

We first describe the basic simulation model[13] before including the refinements just described. The growing crystal is assumed to have a simple cubic structure with neither vacancies nor overhangs (the solid-on-solid model[14]). Growth is initiated by the random deposition of atoms onto the substrate at a rate determined by the flux. The subsequent migration of surface adatoms is taken as a nearest-neighbor hopping process with the rate $k(T)=k_0\exp(-E_D/k_BT)$, where T is the substrate temperature, $k_0=2k_BT/h$, k_B is Boltzmann's constant, h is Planck's constant, and E_D is the hopping barrier. The latter is comprised of a term, E_S, from the substrate, and a contribution, E_N, from each lateral nearest neighbor. Thus, the hopping barrier of an atom with n lateral nearest neighbors is $E_D=E_S+nE_N$, where $n=0,\dots,4$. Thermal desorption is neglected.

In attempting to develop a model with as few free parameters as possible while retaining the essential features of the growth kinetics, some simplifying assumptions have been introduced. Those that require the closest examination are as follows. (i) The mobility and nearest-neighbor bonding of adatoms are isotropic. (ii) The group-V kinetics are not included explicitly in the model, since it is assumed that under normal growth conditions the group-V species is present in sufficient quantities to ensure microscopic stoichiometry. (iii) The effects of the surface reconstruction on mobility can be incorporated as part of the effective migration parameters (E_S and E_N).

In the experiments reported by Shitara *et al.*,[15] growth and diffraction conditions were chosen to conform as closely as possible to these assumptions. In particular, to satisfy (i) the surfaces were misoriented toward the

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[010] direction to reduce the effect of the anisotropy, to satisfy (ii) the As/Ga (atomic) ratio was held at approximately 2.5 to maintain the 2×4 reconstruction in a fairly wide temperature range near the temperature at which growth becomes dominated by step flow. This also addresses assumption (iii). For the reasons discussed in Ref. 15, the diffraction conditions were chosen as "in-phase," or "Bragg" conditions for which the maxima in the specular-beam intensities on the corresponding singular surface occur at monolayer (ML) increments of material deposited. Since for in-phase diffraction conditions the kinematic theory yields a constant intensity, regardless of the surface configuration, the density of surface steps was used to model the *variations* of the RHEED specular-beam intensity during growth. The most striking result to emerge from this study is the extent of *quantitative* agreement between the RHEED specular-beam intensity and the step density evolutions during growth to the extent that the relative changes of the magnitudes of the two quantities with temperature are the same.

On the other hand, there was also clear disagreement during post-growth recovery (smoothing of the surface), which was most evident at lower substrate temperatures. The simulations generally showed a much more rapid recovery than the measured RHEED profiles.[15] More important, these simulations were unable to reproduce the systematic dependencies of the time constant τ_1, Ref. 16, for the initial stage of the recovery (cf. Refs. 17 and 18). These are serious discrepancies because the equilibration of a surface after a period of growth is a more discriminating test of kinetic models than growth alone. During growth under typical conditions, the maximum time scale is set by the deposition flux and processes occurring over longer time scales are "frozen out." However, during recovery, these processes can come into play even though their effect during growth can be safely omitted.[17]

To more accurately model the equilibration of the surface, we introduce an activation barrier to hopping at step edges, E_B, of the same form as that used in our simulations of the reentrant oscillations seen in thermal-energy atom scattering measurements on Pt(111).[8] This barrier is expected to provide a strong signature in the recovery profile by inhibiting the interlayer transport of adatoms and thus slowing down the recovery.[19] However, even a quite small barrier to interlayer hopping has important consequences during growth as well. The surface quickly roughens and complete disagreement between simulations and experiment during growth *and* recovery is observed. We are thus led to the conclusion that if step-edge barriers are present, then some compensating smoothing mechanism must also be present. We have therefore included an additional process that an arriving atom undergoes before being incorporated on the surface. A search is carried out within a square of a fixed linear size L, centered upon the original site, for the site that maximizes the number of nearest neighbors. The effect of this process is a smoothing of edges of preexisting steps and islands created on the terraces. A similar

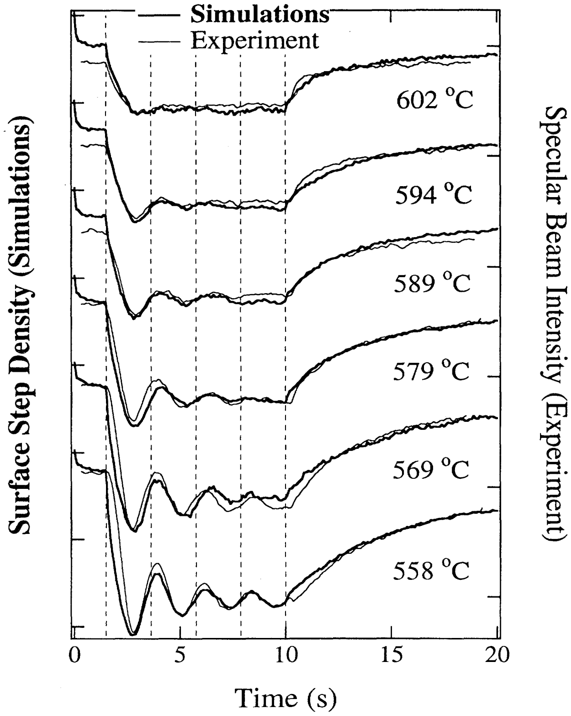

FIG. 1. Direct comparison between measured RHEED intensity and the simulated surface step density on a GaAs(001) vicinal surface misoriented by 2° toward the [010] direction at the growth rate of 0.47 ML/s.

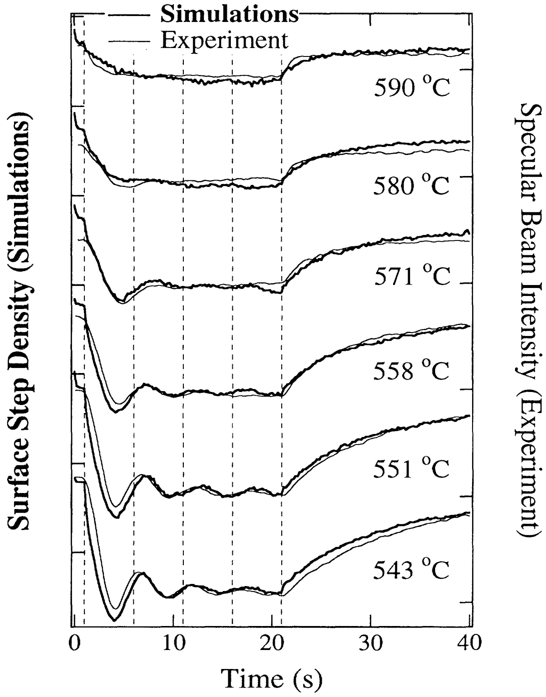

FIG. 2. Direct comparison between measured RHEED intensity and the simulated surface step density on a GaAs(001) vicinal surface misoriented by 2° toward the [010] direction at the growth rate of 0.20 ML/s.

mechanism has been invoked to explain the monolayer to bilayer transition in the growth of group-IV materials,[20] though we can only speculate about its possible microscopic origins for the case of GaAs(001). The simulations reported below were carried out on 232×232 lattices with steps running diagonally across the lattice (a miscut toward the [010] direction)[15] with the parameters $E_S=1.54$ eV, $E_N=0.23$ eV, $E_B=0.175$ eV, and $L=7$.

A comparison of our modified model with the data of Ref. 15 for two different Ga fluxes is shown in Figs. 1 and 2. The step densities show a much better agreement with the RHEED curves over the entire growth and recovery periods than those produced with the original model. The incorporation process brings the simulations and the measurements into closer agreement at the onset of growth and during growth, where the improvement is manifested particularly in a much better reproduction of the first maximum delay phenomenon.[21] This is due to an increase in the number of atoms incorporated into preexisting steps immediately after they are deposited. In Fig. 3 is an Arrhenius plot of the temperature dependence of τ_1, which now *does* exhibit an Arrhenius dependence, in agreement with measurements.[16]

The comparisons in Figs. 1 and 2 are compelling not least because the effects of the incorporation process and step-edge barrier act in *opposition* during growth, as discussed above, but in *concert* during recovery. The step-edge barrier slows down the equilibration process by inhibiting interlayer transport while the incorporation process produces a smoothing of step edges and a decrease in the number of free adatoms, which leads to a high coordination of most of the adatoms forming clusters on the uppermost layer. This reduces the influence of fast processes such as free adatoms migrating to coordinated sites and the elimination of sites with low coordination, both of which were overemphasized in the original model. Therefore, according to our simulations, both step-edge barriers and the incorporation process are important factors for correctly reproducing the recovery curves.

A question naturally arises: are there any other experimental hints of there being barriers at step edges or an incorporation process on semiconductor surfaces? The role of step-edge barriers has been examined in the regularization of terrace width distributions on vicinal surfaces.[22] The existence of step-edge barriers has also been suggested recently as being responsible for the unexpectedly rapid increase in the surface roughness in low-temperature growth of Si (Ref. 10). The "forbidden temperature window" in the growth of AlAs(001) (Ref. 23) is very suggestive of reentrant layer-by-layer growth behavior caused by the step-edge barriers. Alternatively, the observations of epitaxial growth at very low temperatures[1,24] could be explained as the effect of an incorporation process.

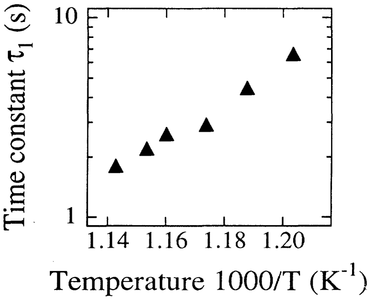

FIG. 3. The Arrhenius temperature dependence of the time constant τ_1 of the fast stage of post-growth recovery.

A final comment is in order concerning the results presented here with regard to the presence of As. Shitara *et al.*[15] observed that the areas of disagreement between the RHEED measurements and the original model are in transient regimes of growth before the As/Ga ratio has attained a steady state value.[18] The explanation of the recovery effect presented in this paper relies on the value of the step-edge barrier (and also other model parameters) being the same during growth and recovery. In particular, if the step edges are the preferred sites for adsorbed As, then maintaining a steady state As/Ga ratio over the entire surface is not required, though the absence of As at these sites would be expected to have an observable effect. In fact, at the highest temperatures, where As desorption becomes most appreciable, the step density recovers more slowly than the RHEED specular-beam intensity (Figs. 1 and 2), suggesting that a step edge without As has a lower barrier to interlayer hopping than the same edge with an adsorbed As.

In conclusion, we have studied growth and post-growth recovery on GaAs(001) surfaces using computer simulations of a solid-on-solid model. To the best of our knowledge, we have achieved for the first time *quantitative* agreement between the evolution of the RHEED specular-beam intensity and its theoretical counterpart, in our case the surface step density. Our results support strongly the idea of there being step-edge barriers to interlayer transport as well as an incorporation process for arriving atoms on GaAs(001) surfaces.

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