Evaporation and Step Edge Diffusion in MBE

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Using kinetic Monte–Carlo simulations of a Solid–on–Solid model we investigate the influence of step edge diffusion (SED) and evaporation on Molecular Beam Epitaxy (MBE). Based on these investigations we propose two strategies to optimize MBE–growth. The strategies are applicable in different growth regimes: during layer–by–layer growth one can reduce the desorption rate using a pulsed flux. In three–dimensional (3D) growth the SED can help to grow large, smooth structures. For this purpose the flux has to be reduced with time according to a power law.

Key words: Evaporation and Sublimation; Growth; Surface Diffusion; Surface structure, morphology, roughness, and topography

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The growth of high quality compound semiconductors is of great technological importance. In order to optimize growth a detailed knowledge of microscopic processes is very important. We will follow this line of thought and will report on two new approaches of growth which will be presented in more detail in a forthcoming publication. We will exploit macroscopic effects of two distinct microscopic mechanisms. The term “microscopic” refers to events at the atomic scale: e.g. a single diffusion step of an adatom or evaporation of an atom. These are the ingredients of the computer model. This is contrasted to the term “macroscopic” for effects which are typically measurable in experiments: e.g. the overall mass desorption, form and distribution of three–dimensional structures, or the growth rate.

The model used here is the conventional solid–on–solid model on a simple cubic lattice. All simulations will start on a singular surface. The system size will be at least 300×300 lattice constants with periodic boundary conditions. Even

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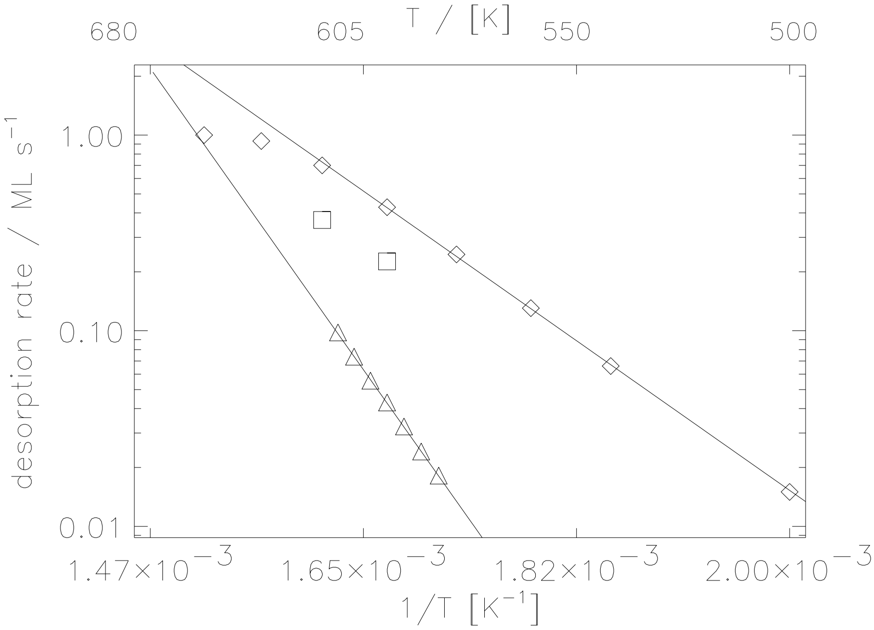

Fig. 1. The diamonds (◇)represent measured desorption rates during growth. At high temperatures the desorption rate saturates and equals the flux of impinging particles (1 ML/s). The desorption rate is considerably higher than the sublimation rate of the same model (△, sublimation rate is multiplied by ten). Using the proposed "flush-technique" we obtain lower desorption rates (□). We used a mean flux of 1 ML/s - a constant flux of 0.77 ML/s plus additional 0.23 ML in 0.003 s at the beginning of each second.

though the model includes only one species of particles it has been successfully applied to reproduce quantitatively RHEED-oscillations during the growth of GaAs(001) [1]. In this paper we want to study qualitative effects rather than to derive a model for a specific material. For this purpose we set the vibration frequency to $\nu_0 = 10^{-12} s^{-1}$ which is of the order of typical Debye frequencies. The activation energy for the different microscopic processes is parameterized in a simple manner. We choose a barrier for diffusion of $E_B = 0.9$ eV, $E_D = 1.1$ eV for desorption and at step edges an additional Ehrlich–Schwoebel barrier E_S of 0.1 eV. Each in–plane neighbour contributes the binding energy of $E_N = 0.25$ eV. This set of parameters is loosely connected to CdTe(001) and reproduces some features of sublimation [2,3] and annealing.

Two of the most important compound semiconductors decrease their growth rate with increasing temperature in MBE–growth (CdTe(001) [4–6] and GaAs (001) [7]). The desorption rate was found to be activated with a low activation energy. This should be contrasted to sublimation with considerably higher activation energies (c.f. [8] for CdTe(001)). The same qualitative difference in activation energies is observed in simulations [9,2]. As can be seen in fig. (1) desorption during growth (flux of arriving particles F = 1 ML/s) is an activated process with 0.9 eV which is considerably lower than E_D. This is due to a negative contribution of E_B to the effective energy: high diffusion barriers reduce the diffusion length more than the typical island separation. Hence, the desorption rate increases. On the contrary, sublimation (F = 0 ML/s) shows an activation with 1.7 eV. Here, the freely diffusing adatoms (which evaporate easily) must be created through the detachment from steps or the creation of vacancies. Thus the sublimation energy is much higher than E_D.

Our findings imply a simple way to reduce desorption during growth: short flushes of particles at the beginning of each monolayer result in a great density of islands. Afterwards with a low flux the particles most likely hit islands to stick to. This reduces the adatom density and hence yields a low overall desorption rate. As can be seen in fig. (1) such simulations yield desorption rates reduced by a factor 2. In addition this method leads to prolonged layer–by–layer growth as will be shown in a forthcoming publication.

Quite generally, layer–by–layer growth is not attainable forever [10]. To optimize MBE–growth it could thus be advantageous to study the growth of 3D–structures. Using a simplified model [11] we have found a strong influence of SED on the properties of the growing surface. Clearly, a strong SED leads to structures with smooth step edges. However, a second consequence of strong SED is a fast coarsening process. If the structures are becoming too large compared to the SED–length ℓ_{SED} the coarsening process slows down and the step edges become rough. Hence, MBE–growth always drives the surface in the latter regime with comparably weak SED. This can be avoided by *e.g.* reducing the flux during growth which in turn increases ℓ_{SED}. The flux–dependence of the typical extension of mounds as well as of the SED are power laws in time. Hence the flux should be reduced according to a power law $F(t) \propto 1/t^{\omega}$. A detailed calculation and determination of ω will be published in a forthcoming publication.

We applied this strategy to the growth of the model at $560K$ which is a reasonable growth temperature for CdTe(001). For a first qualitative investigation we choose $\omega = 0.1$. To prevent the inference of evaporation we inhibited this process. However, we checked that even with desorption, the strategy is still applicable and useful. After the deposition of 300 ML under constant flux the structures are small and of irregular shape. With the adaption of the flux (in the end $F = 0.11$ ML/s) the structures are considerably larger (SED assisted coarsening) and the steps are smoother.

To summarize, we have shown how the detailed knowledge of underlying microscopic (atomistic) processes can lead to an optimization of MBE–growth. If desorption is present a flush–technique can decrease the desorption rate during layer–by–layer growth. Besides the improved growth rates such experiments allow to decide whether physisorption (as speculated in [6]) or the

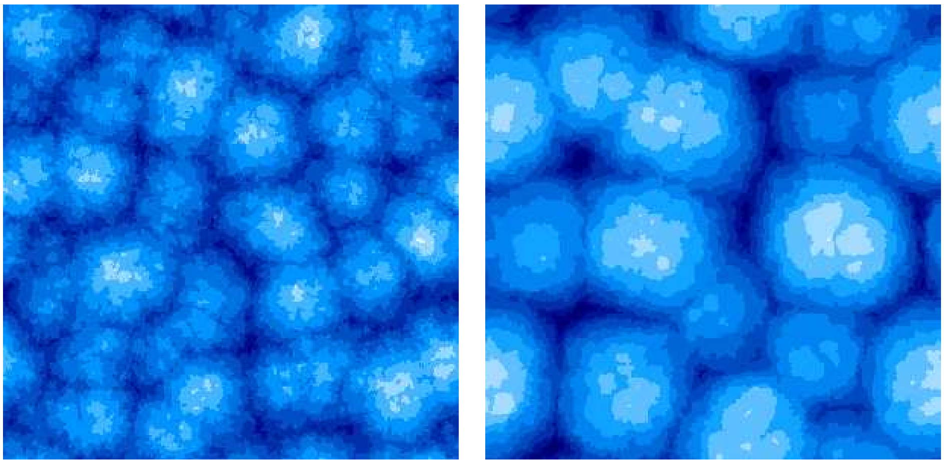

Fig. 2. Snapshots of surfaces after the deposition of $\langle h \rangle = 300$ ML. Both simulations started with a flux $F_0 = 1$ ML/s. The right surface corresponds to the simulation where the flux was reduced during growth according to $F = F_0/(t/10s)^{0.1}$ after the deposition of 10 ML's. The system size is 300×300 in both cases.

above mechanism is dominant. To obtain larger structures in conventional 3D–growth one just has to grow for longer times. The step edges will become smooth due to the equilibration after growth stops. However, during growth a larger probability for the creation of vacancies or other crystal defects will be present. After growth stops these faults cannot be eliminated. This is the point where our second strategy improves the growth. The step edge diffusion remains always strong enough to maintain smooth step edges.

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