Epitaxy using Total−energy DFT Calculations in Combination with

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Modeling Molecular Beam Epitaxy using Total−energy DFT Calculations in Combination with kinetic Monte Carlo Simulations

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When modeling epitaxial growth, the properties of interest (e.g. the growth morphology) develop over time scales of the order of seconds and involve several thousands of atoms, while the ruling microscopic processes operate in the length and time domains of 0.1 − 1 nm, and femto− to pico−seconds. Even with a further increase in computer power, such time and length scales will not be accessable to simulations using *ab initio* molecular dynamics. We are thus challenged by the need to bridge this gap in length and time scales by many orders of magnitude using multiscale modeling techniques. Here, kinetic Monte Carlo (kMC) simulations offer an efficient and accurate simulation tool. While earlier kMC simulations employing empirically derived effective parameters lacked predictive power, we show that the inherent reliability of the *ab initio* approach can be extended to experimentally relevant situations, provided that the rates entering in a kMC simulation are directly derived from DFT calculations.

Using the results of DFT calculations [1] for the atomic−scale processes of growth on the β2 (2×4)−reconstructed GaAs(001) surface, including adsorption, desorption [2], surface diffusion [3] and nucleation [4], we show how the molecular beam epitaxy of GaAs can be modeled in atomistic detail. The main results of these investigations are:

- As_2 molecules adsorb without dissociation and become surface As dimers. Adsorbed Ga atoms incorporate themselves by splitting these As surface dimers.
- Non−activated As_2 adsorption into strongly bound adsorption sites is possible only when the As_2 can attach to a square array of three or four Ga dangling bonds.
- Bound pairs of Ga adatoms that could act as island nuclei originate either from dimerization of Ga atoms or from an indirect interaction mediated through the substrate reconstruction.

In order to understand the consequences of the microscopic physics for the growth properties, it is required to investigate the interplay of these various processes during growth. By performing kinetic Monte Carlo simulations on the basis of the DFT results, we are in position to assess the importance of specific processes at various growth temperatures and to explore the statistical aspects of island nucleation and growth.[5] We extract the saturated island density from the simulations, and compare its temperature dependence with the scaling laws predicted from traditional nucleation theory. The simulated island morphology is also compared with STM images of 2D islands obtained from growth experiments after submonolayer deposition.

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