Ab-Initio Based Multiscale Calculations Of Extended Defects In And On Group III – Nitrides

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Preface

In late 1993 S. Nakamura of Nichia Chemicals introduced the first high efficient GaN-based blue light-emitting diode. This year is widely regarded as the turning point in the field of III-Nitride semiconductors. Since then group III-Nitrides are a hot topic in the semiconductors research. While there has been a tremendous progress in materials quality and and device fabrication our understanding concerning fundamental properties and growth phenomena is often still limited.

One of the controversial issues is the effect dislocations have on the electronic properties of the GaN epilayers. Dislocations are characterized by two different length scales: The core structure of the defect is rather localized, while the strain field introduced by the defect is significant even far away from the core. *Ab-initio*-based methods are a reliable tool for atomic scale calculations. However, they are characterized by a rather limited (limited by the available computational power) ability to describe systems consisting of large numbers of atoms. Thus, it is essential to develop methods which allow to describe physical systems on all relevant length scales. We have therefore developed *ab-initio*-based multiscale approaches which combine first-principles calculations with mesoscopic concepts. A first approach combines first-principles, empirical potentials and elasticity theory and links the microscopic with the macroscopically relevant length scales. The accuracy of this approach is in principle limited only by the accuracy of the *ab-initio* calculations. A second approach, which has been applied to study growth is *ab-initio atomistic thermodynamics*: First-principles calculations are combined with the thermodynamic concept of chemical potentials and enable to derive the thermodynamic stability of the various systems as function of the growth conditions.

In the first part of the thesis, the focus will be on the growth of GaN surfaces. A variety of step/vicinal surface configurations on the technologically most relevant GaN (0001) orientation are considered and their thermodynamic stability under different growth conditions is examined. An important conclusion from these studies is that steps may spontaneously form under N-rich conditions, while under more metal-rich conditions they become thermodynamically unfavorable against a Ga rich laterally contracted bilayer structure. Next, we spotlight the Ga-rich growth conditions limit and we study the formation of small Ga islands on the laterally contracted bilayer structure. Our calculations reveal that the islands can be assumed as precursors of Ga droplet formation. Based on these results we explain recent controversially discussed growth studies within a unified model. Our calculations further reveal that structures exceeding 2 MLs of Ga are feasible and thermodynamically stable under extreme Ga rich conditions even on intrinsically non-polar surfaces such as the GaN $(1\bar{1}00)$ as important for growing field-free structures.

Next, we move from surfaces to interfaces and we study monatomic-scale ordering in AlGaN alloys. In agreement with recent experiments we find that ordering results in bandgap reduction. The origin of the bandgap reduction is shown to be a carrier localization in mono-atomic GaN quantum wells embedded in AlN barriers. Finally, from the two dimensional features (surface/interfaces) we move to one dimensional extended defects in GaN. i.e., dislocations. Based on our multiscale approach we investigate the properties of a variety of different core configurations. We find that the giant local strain-field around the dislocation core, in combination with the small lattice constant of GaN, causes deep defect states and thus electrically active edge dislocations even when all core atoms are fully coordinated.

In summary, in this thesis we developed a multiscale approach and applied it to address a number of hitherto unsolved problems and to get a microscopic understanding of extended defects in and on group III-Nitrides. This thesis has been performed during the period July 2000 - December 2004 in the Independent-Junior-Research Group of the Fritz-Haber-Institut der Max-Planck-Gesellschaft.

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