Stony Brook University
The Graduate School

Doctoral Defense Announcement

Abstract

Theoretical and Computational Studies Related to Solar Water Splitting with Semiconductor Alloys

By

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The $(Ga_{1-x}Zn_x)(N_{1-x}O_x)$ solid solution (or, alloy) is a visible-light-driven photocatalyst for water splitting. Its reduced band gap is a main advantage for harvesting solar energy. Because the synthesized samples are in the powder form, the understanding of the bulk structures and the surfaces are hindered. In this thesis, we address both the bulk and the surfaces of this material through simulations based on the density-functional theory (DFT) version of quantum electronic structure theory.

The ordering of the atoms in the alloy is the key information to understand the bulk properties, especially the band gap reduction mechanism. Using the cluster expansion formalism, we construct an accurate model from DFT calculations. The subsequent Monte Carlo simulation reveals a phase diagram which has a wide miscibility gap and an $x = 0.5$ ordered compound. The disordered phase displays strong short-range order (SRO) at synthesis temperatures. To study the influences of SRO on the lattice and the electronic properties, we conduct DFT calculations on snapshots from the Monte Carlo simulation. We find that the lattice parameters deviate from Vegard's law. The bond lengths show unusually large relaxations. The band gap bowing depends on both the SRO and the concentration of ZnO in the alloy.

Recent experiments showed that the semi-polar $(10\underline{1}1)/(10\underline{1}\underline{1})$ surfaces dominate the powder samples. To search for stable reconstructions of these two surfaces, we use a genetic algorithm to explore the surface structures. To simplify the study, we only consider the pure GaN bulk with various numbers of Ga, N, and O atoms allowed to bond to surfaces. A few stable reconstructions at different Ga, N, and O chemical potentials are found. The consequences for the water splitting catalysis are discussed.

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