Stony Brook University
The Graduate School

Doctoral Defense Announcement

Abstract

First Principle Study of Water: From Fundamental Properties to Phtotocatalytic Reactions

By

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Ab initio molecular dynamics (AIMD) simulations of liquid water, based on density functional theory (DFT), yield structural and diffusive properties in reasonable agreement with experiment when artificially high temperatures are used in the simulations for gradient-corrected (GGA) functionals, with NVT ensemble. A series of simulations with GGA functionals is performed and underestimation of equilibrium density, at ambient conditions, is found. Electronic density of water molecules with several GGA functionals is examined and used to explain structural and dynamical property differences. Dispersive non-local electron-electron correlations are studied with a van der Waals density functional (vdW-DF) of Dion et al. When a vdW-DF is used, the density improves drastically and the experimental diffusivity is reproduced without the need of thermal corrections. The origin of the density differences between all the functionals is analyzed. It has been shown the vdW-DF increases the population of non-H-bonded interstitial sites, at distances between the first and second coordination shells. However, it excessively weakens the H-bond network, collapsing the second coordination shell.

GaN/ZnO alloy semiconductors have been shown to be promising materials to serve as photo-anode in photocatalytical fuel cells. A thorough AIMD study of the non polar GaN(1010) surface with aqueous interface is carried out. It has been shown that water dissociation happens very fast within 1ps. Interacting OH dimers are found to account for around 80% of the dissociated water molecules through a hydrogen bond. A detailed study of the OH dimers shows their distinguishable structural, dynamical and electronic properties. Solvent water molecules interacting with a dissociating wet surface is also studied in detail, especially the hydrogen bond network. Proton diffusion is found starting from non-dissociated surface water molecules. The effective free energy barrier of proton diffusion is shown to be near room temperature thermal energy.

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