First-principles studies on silicon nanowire surfaces

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First-principles studies on silicon nanowire surfaces
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Abstract

Silicon nanowires (SiNWs) are extremely important in nanoscience and nanotechnology because Si-based nanoelectronics is completely compatible with Si-based microelectronics. One of SiNWs' main advantages stems from their tunable properties for various functional applications. Due to the large surface-to-volume ratio, surface effects become significant and promising for property tuning of SiNWs. The objective of this thesis work is to theoretically study several aspects of SiNW surfaces. Using a first-principles approach based on density functional theory, it is revealed that significant surface effects are critically important for the functionalization and application of SiNWs.

This thesis began with a clean surface of bulk silicon, which was useful for the understanding the surface of SiNWs. Silicon dimer along the $[01\bar{1}]$ direction is the basic reconstruction configuration on a Si(100) surface. But the details of the dimer arrangement are complicated and remain controversial. In Chapter 3, we show that both the energetics and vibrational dynamics reveal the sensitivity of surface reconstructions to initial surface morphologies. Consequently, domains of different reconstructions are induced by local structures may be introduced by temperature fluctuations or other disturbances. The p(2×2) is the minority in the product compared to p(2×1) and c(4×2), and hence rarely observed in experiments, even though it is energetically favorable. Additionally, spontaneous dimer-flipping in an asymmetric p(2×1) surface around 100 K in the dynamics simulation provides a new insight into the dominative c(4×2) geometry observed in experiments from 80 K to

200 K.

In Chapter 4, we study the surfaces of SiNWs with and without any passivation. Without any passivation, dangling bonds on the surface involve strong interactions, and can induce metallic characteristics in SiNWs. These dangling bonds are chemically very active, and are commonly saturated by hydrogen atoms for stability. Upon passivation, the configurations of symmetric and canted silicon dihydrides (SiH_2) on (100) facets are found to be significant in the electronic and optical properties of <110> SiNWs. We reveal that surface terminations with canted SiH_2 result in a dramatic widening of the band gap, with increments as large as 20%. The valence band maximum diffuses in the surface layer, which enhances the electronic activity of surface defects. The imaginary part of the dielectric function of the wire with canted SiH_2 is, in general, leans more towards the blue shift than to the wire with symmetric SiH_2.

Generating lattice strain is a possible approach for improving the performance of silicon-based devices. The strain also plays an important role in determining the electronic and optical properties of SiNWs. For instance, it can be generated in SiNWs by introducing surface stresses or lattice mismatching with the surrounding medium or substrate. However, such transverse strains have not been studied in simulations. In Chapter 5 we present a new strategy, namely a “cyclic replacement” approach, to simulate the transverse deformations of SiNWs. We find that a tensile strain within the surface of a <112> SiNW induces an indirect-to-direct band-gap transition, while a compressive strain preserves their indirect characteristics. In cases

of <110> SiNWs, the band gap maintains a direct band-gap feature with the tensile strain within the surface; however, a direct-to-indirect band-gap transition could be induced by a compressive strain. Due to the surface strains, the state of the valence-band minimum (VBM) and the conduction-band minimum (CBM) could be spatially separated. The CBM of the <112> SiNW is pushed to the tensed surface, while the VBM of the <110> SiNW is located within the compressed surface. This effect may be beneficial for SiNW applications in solar cells because thermal excitation and internal electric fields could separate the electron and hole spatially into core and surface.

The large surface-to-volume ratio of SiNWs could potentially be important in determining their transport properties. Using first-principles calculations and experiments we show, in Chapters 6 and 7, that surface effects allow the effective doping of SiNWs via electron transfer across the surface layer, which provides a considerable concentration of majority carriers in SiNWs that are induced by surface passivants such as hydrogen. Surface passivation doping could be used for a wide range of nanodevice fabrications, including the diode array that was fabricated in this work. This was achieved by terminating the different sections of a SiNW with different passivants. Furthermore, the transport properties of SiNWs can be modulated by additional adsorption via the transfer of fractional electrons to or from adsorbates.

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