

# Ab-initio calculations of electronic states in nano-crystalline Si quantum dots

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## Introduction

Nanocrystalline Si Quantum Dots (QDs) has widely been studied as fundamental building blocks for the bottom-up approach in order to construct nm-scale device structures. Strongly coupled Si QDs are particularly interested for quantum information device applications. Theoretical analysis has recently been reported on the stable structures for QDs with the size of under a few nanometers [1][2]. Experimental studies have also been reported on the structures for extremely small Si QDs with diameter less than 2 nm [1][3]. In addition, the wave function interaction has been recently studied for double Si QDs, and the quasi molecular states, which are the bonding-like states and the antibonding-like states have been observed at 4.2K [4]. In this paper we use the ab-initio method to demonstrate the electronic states of strongly coupled double Si QDs with diameter down to 1 nm. We analyze the formation of a two level system in an atomistic manner and discuss how the tunnel splitting between the two levels depends on the structural configuration of the double dots.

## Simulation Models

In the present analysis we used the well tested SIESTA, which is the LCAO based DFT simulation package [5][6]. First we studied various possible crystal structures for a single Si QD with diameter around 1 nm. It has been reported theoretically and experimentally that there are various stable structures for 1-nm-scale Si QDs, and we chose the four structures with hydrogen terminated surfaces (see Fig. 1): (a) Bulk-like, (b) 29Si-24H [1], (c) Icosahedral [2], (d) Fullerene-like. Figure 2 shows the bare eigenenergy spectrum calculated for each of these structures. The energy levels become discrete because of the quantum size effect, and the numbers listed in the figure shows the number of eigenvalues that consist each energy subband. The density of the subbands differs depending on the structure. The Icosahedral structure shows the most discrete nature which is almost like an atomic energy spectrum. Structures (a) and (c) can be scaled up, but the other two structures are unique. The band gaps were calculated by taking the difference between the HOMO and LUMO states which is shown in Fig.3. The band gaps show quantum size effects. The four structures at about 1nm show that the band gaps also differ

from structural differences.

## Quasi-Molecular States

Theoretical work for multiple QDs have not been accomplished so far, compared to the number of researches done on isolated QDs. We hereafter use the Icosahedral structure because of its most atomic like energy structure and the symmetry it possesses to calculate wave function coupling between two QDs.

We first parted the Icosahedral QDs by vacuum and calculated the Total Density of States (TDOS) for the system. Figure 4 shows the splitting of each state, and the bonding and antibonding states can be observed. As mentioned above the normal subbands are consisted of a number of eigenvalues so the subbands will not break into just two states. However the normal LUMO state is composed of just one eigenstate and is fairly isolated from the other eigenstates. For this reason we focused on the LUMO's bonding and antibonding states. The Localized Density of States (LDOS) is visualized in Fig.5 to observe the orbitals that consist these states. We have changed the orientation of the Icosahedral QDs and found that the orbitals can be classified into two categories. The conventional type of quasi molecular state (QMS) (Fig. 5(a)), and the quasi localized state (QLS) (Fig. 5(b)). The bonding- antibonding splitting is found 112 meV for QMS and 78 meV for QLS. Although the overall size of the LUMO split is found larger for QMS, the QLS can also form bonding and antibonding states. We also found that by bridging the QDs via oxygen, the size of the LUMO splits are enhanced and the relative positions are fixed.

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## References

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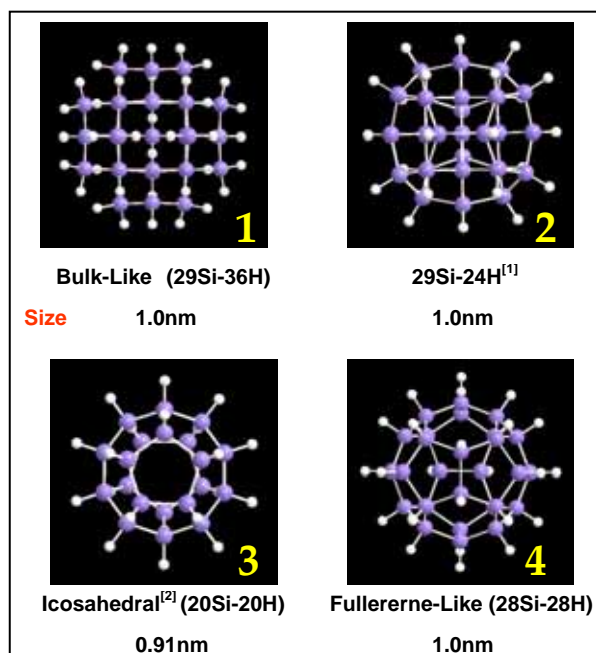


Fig. 1: Structural variety in the 1 nm regime

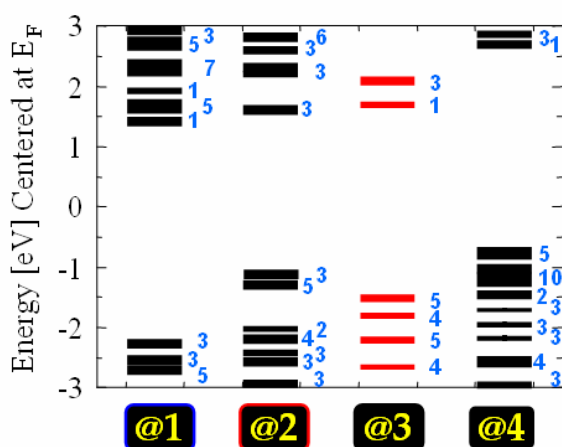


Fig. 2: Bare eigenenergy spectra for the four structures

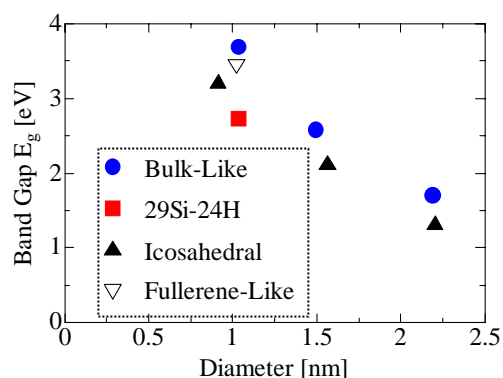


Fig. 3: Bandgap calculated for the four structures as a function of QD diameter

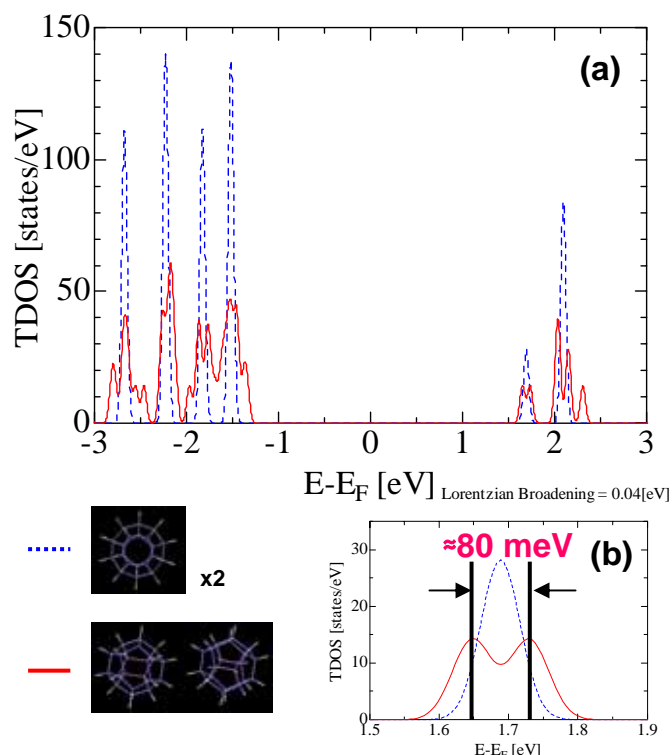


Fig. 4: (a) Total Density of States (TDOS)  
(b) Close-up view of the split LUMO state

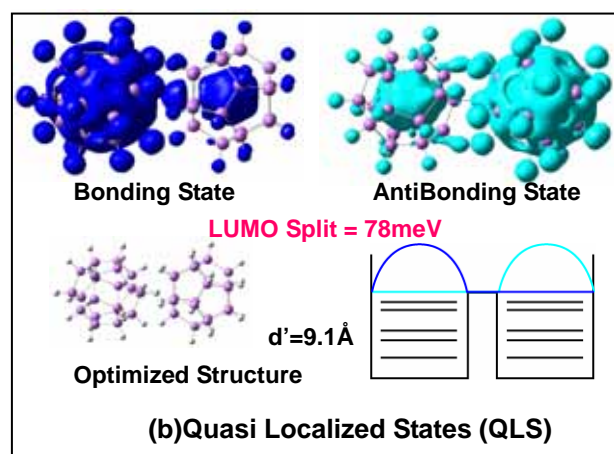
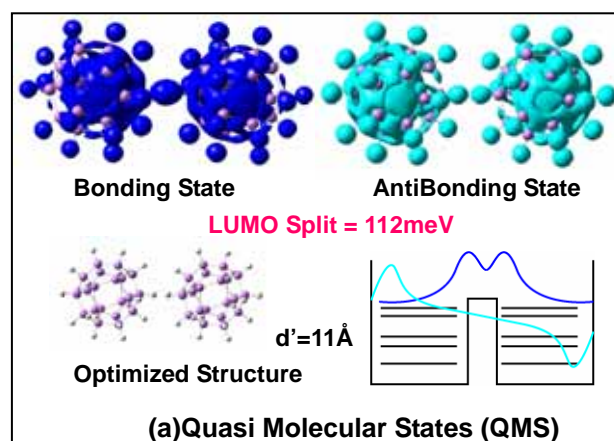


Fig. 5 Visualization of the Local Density of States (LDOS) for (a) QMS and (b) QLS

## Abstract's Abstract or maybe summary

By using the Icosahedral QDs we have obtained the bonding and antibonding states by ab-initio calculations which may lead to a very clear two level system. By visualizing the states, not only the conventional quasi molecular states but also the quasi localized states were observed as two level systems.

In this paper we use the ab-initio method to demonstrate the electronic states of strongly coupled double Si QDs with diameter down to 1 nm. We have analyzed the formation of a two level system in an atomistic manner. By changing the orientation of the QDs we have found that the orbitals can be classified into two categories. The conventional type of quasi molecular state (QMS) and the quasi localized state (QLS). Overall splitting is found larger in QMS, but the QLS can also form two level states. By bridging the QDs via oxygen, we have shown the possibility of forming a very clear two level system.