Motivations: towards quantum control of quantum dots

Quantum dots (QDs) are nanoscale semiconductor crystalline structures where the electron/hole wavefunctions are confined in all three directions of space, resulting in a quantized energy spectrum, as opposed to bulk semiconductors where the absorption spectrum is continuous above the band gap. The size of the dot will define the spacing between excited states, therefore allowing manipulation of the nanocrystal to the desired optical properties.[1]

The potential of such devices is enormous, ranging from transistors, lasers or biological imaging to medical diagnosis.[2] In all these domains, the experimental developments have been astonishing. Yet, from a theoretical point of view, the microscopic description of these devices is rendered difficult by the frontier between molecules and solid state. This requires knowledge of different aspects such as the band structure, the nature of the lattice vibrations and the details of the electron-phonon interactions and of the transport physics.

One of the potential applications of QDs is of particular interest to the future of computer technology: the use of excitonic states for quantum computing. In this scheme, the presence (absence) of excitons localized in the QDs represent the 1 (0) of binary language, while laser pulses perform the quantum operations. The idea is to control the laser pulses pursuing these operations through the use of optimal control theory. The goal of this work is then to get a better understanding of the interaction between the field and the QDs.

The considered QDs are made of silicon, which has been the cornerstone of the growth of microelectronics for decades, because of its abundance on Earth and the ease with which its properties can be modified.

The first step of this work consists in obtaining accurate ab initio calculations of the structural parameters involved in the model:

- energy levels
- transition dipole moments
- ...

Methodology

The quantum dots were modeled using the DENEH program[1] to cut out a sphere in bulk silicon then passivating those spheroids with hydrogen atoms, thus resulting in the nanoclusters shown below:

<table>
<thead>
<tr>
<th>QD</th>
<th>Cutting diameter</th>
<th>Si₂H₄</th>
<th>Si₃H₆</th>
<th>Si₅H₁₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si₂H₄</td>
<td>0.8 nm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si₃H₆</td>
<td>1 nm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si₅H₁₂</td>
<td>1.25 nm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si₇H₁₈</td>
<td>1.5 nm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si₉H₂₆</td>
<td>1.75 nm</td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

These structures were optimized using two different approaches:

- Plane-wave DFT via VASP
  - with the PBE & B3LYP functionals, PAW potentials and an energy cutoff of 250 eV
  - since there is no large-scale periodicity in our system, calculations were done using the Γ point as the only k-point.
- “Classic” DFT via ORCA
  - with the PBE & B3LYP functionals and the 6-31G* basis set

This way we were able to obtain the HOMO-LUMO gap of the clusters through Kohn-Sham eigenvalues.

TD-DFT calculations were also performed with ORCA with the same functionals and basis set, as it is a more suitable method for the description of excited states. This way we were able to obtain the optical gap of the clusters. Unfortunately, to our knowledge, VASP doesn’t seem to support TD-DFT.

Results

Results of our DFT and TD-DFT calculations are shown in the above graph. When compared to the literature[4-7], we can see that:

- The B3LYP functional seems better suited for this type of calculation, presenting, at the TD-DFT level, a rather good agreement with the literature.
- The difference between plane-wave basis set and localized basis set is very small (~0,1 eV), except for smaller clusters (~0,35 eV).

Perspectives: towards quantum dot arrays

Using a single quantum dot as a support for quantum computing would not prove to be very effective. Indeed, the lifetime of higher excited states would probably be too short to allow us to implement any noticeable logical operation.

- Alternative: build an array of quantum dots
  - A quantum dot represents a qubit: 0 in the absence of an exciton, 1 in the other case.

The next step of this work is then to model a pair of quantum dots and apply our optimal control codes to it, similarly to what Raisanen et al. have done[8].

- This way we should be able to pursue a controlled population transfer and populate a dark exciton state, much like we did with acetylene in one of our previous works[9].
- We could also control the “communication” between quantum dots with the help of an external tunable electric field, affecting the permanent dipole-dipole interaction.

Concerning the optical addressing, one idea could be to establish a quantum dot “matrix”, similar to the one developed by Bao and Bawendi for their quantum dot spectrometer (see Figure below)[10].

This way, we could distribute quantum dots of different sizes along the matrix, allowing only one quantum dot at a time to react to the laser excitation.

References