QUANTUM CHEMICAL SIMULATIONS OF DOPED TiO_2 NANOTUBES FOR PHOTOCATALYTIC HYDROGEN GENERATION

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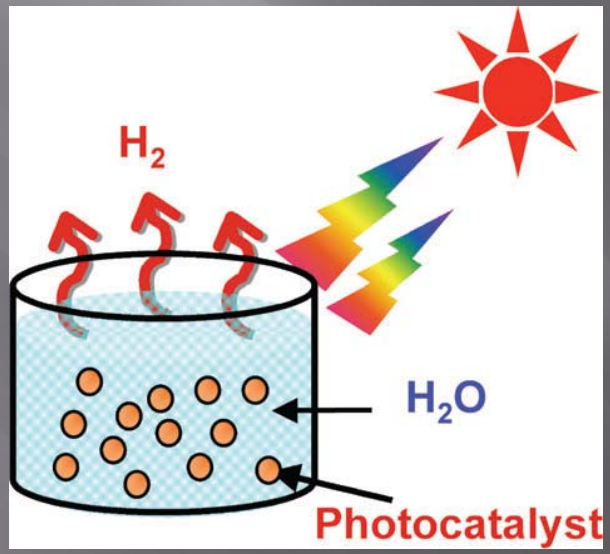
Problem

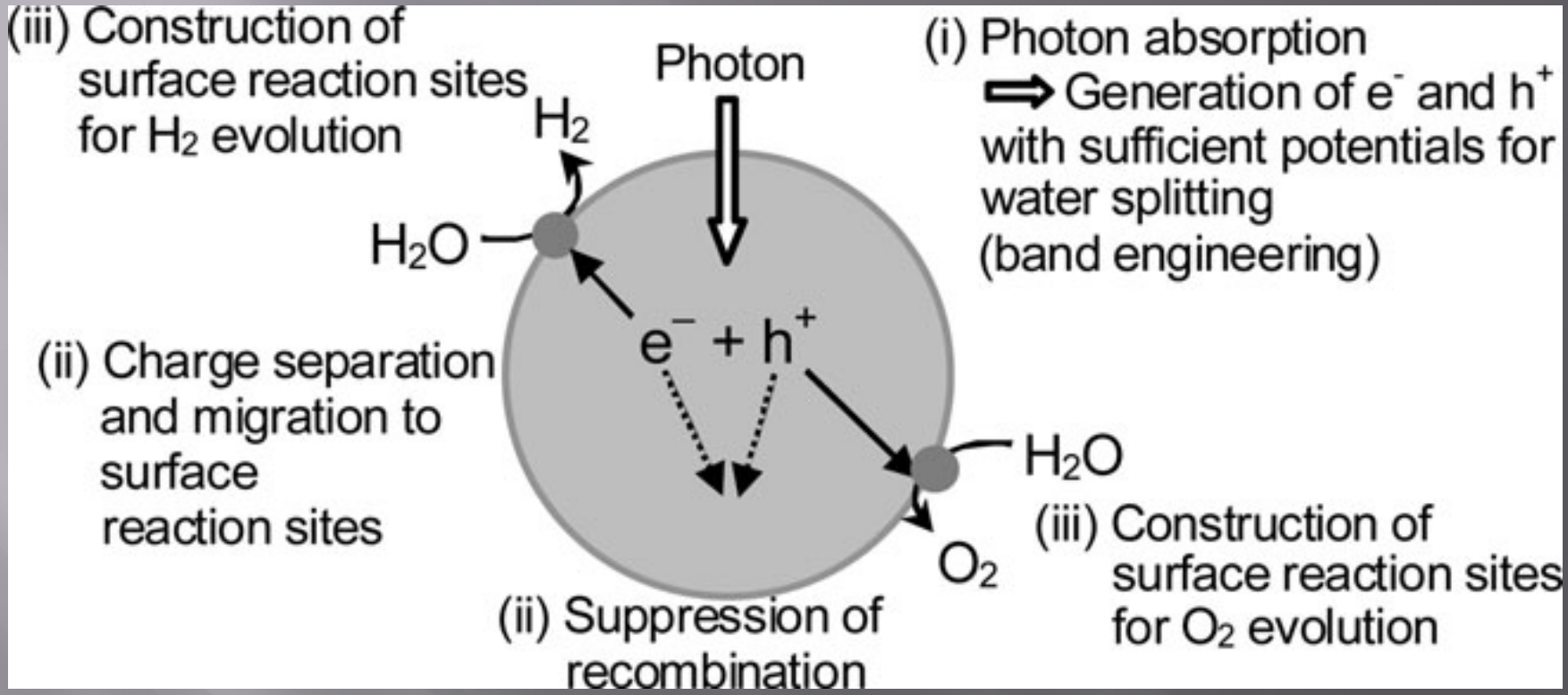
- Issues of alternative energy sources and environment protection are one of the most important topics of today.
- Hydrogen is a extraordinary environment-friendly energy source; in addition it is widely used in chemical industry
- Today hydrogen is produced from fossil fuels – such as natural gas

One of the ways how to generate H_2 by means of solar energy

- Photocatalytic water splitting; a superfical scheme

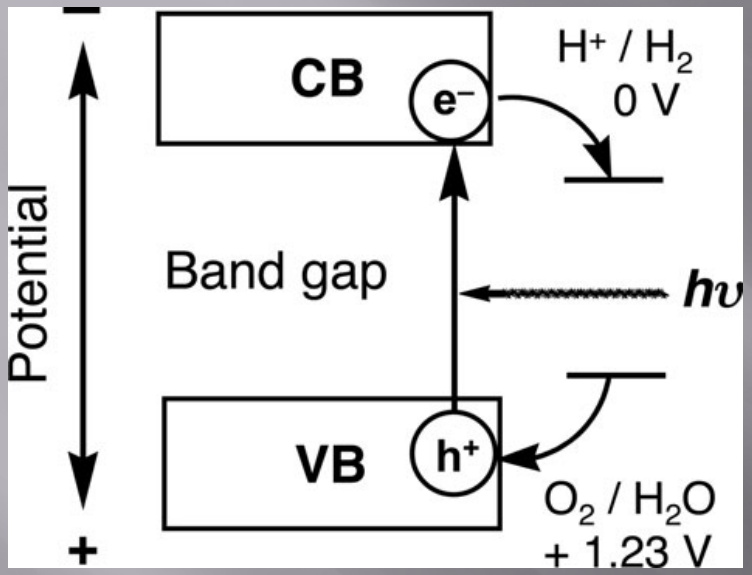


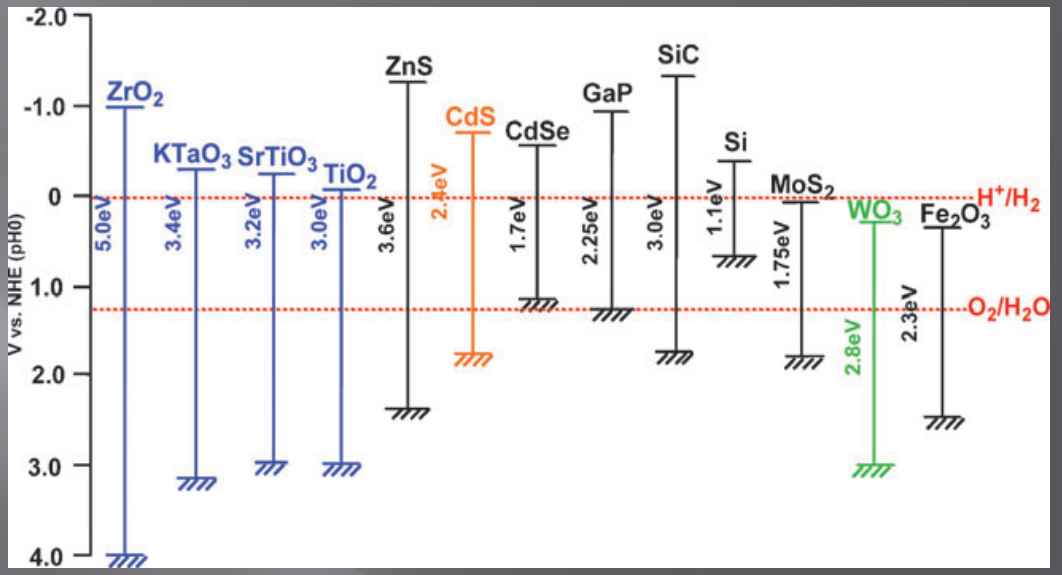
Hydrogen photocatalytic generation



- Photon absorption, e^- un h^+generation
- Charge separation and migration, avoidance of recombination
- Generation of reaction centers

Hydrogen photocatalytic generation





- Band gap: 1.23 – 2.5 eV interval
- Positions of valence band and conduction band
- Choice of catalyst
- Catalyst modifying

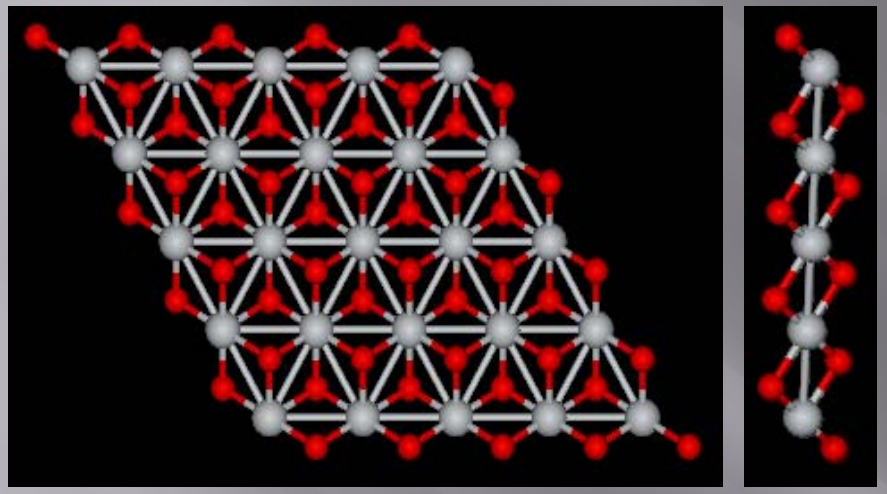
Calculations

- *ab initio* – LCAO - GTF – hybrid exchange-correlation functionals – DFT
- Commercial periodic code CRYSTAL
- Latvian SuperCluster (LASC, 222 CPU cores)

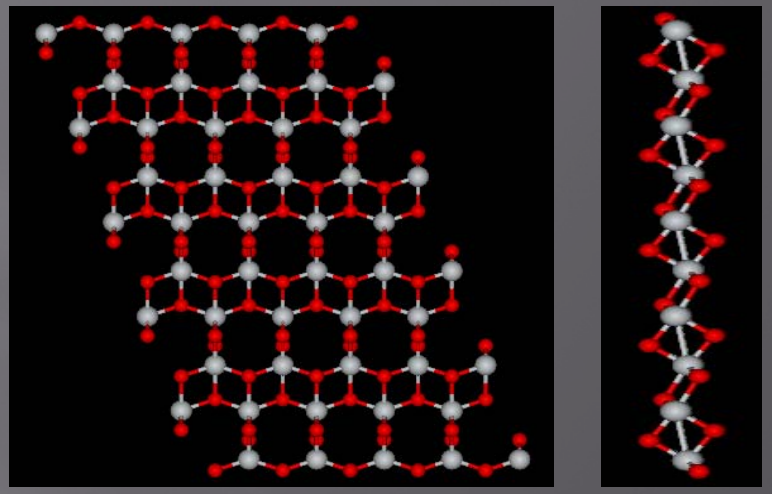
Stages of work I

- E_{xc} selection (PBE0, B3PW, B3LYP, SOGGAXC)
- Analysis of results – based on experimentally found band gap TiO_2
- B3LYP – the most reliable results, further precision - corrections for HF non-local exchange part

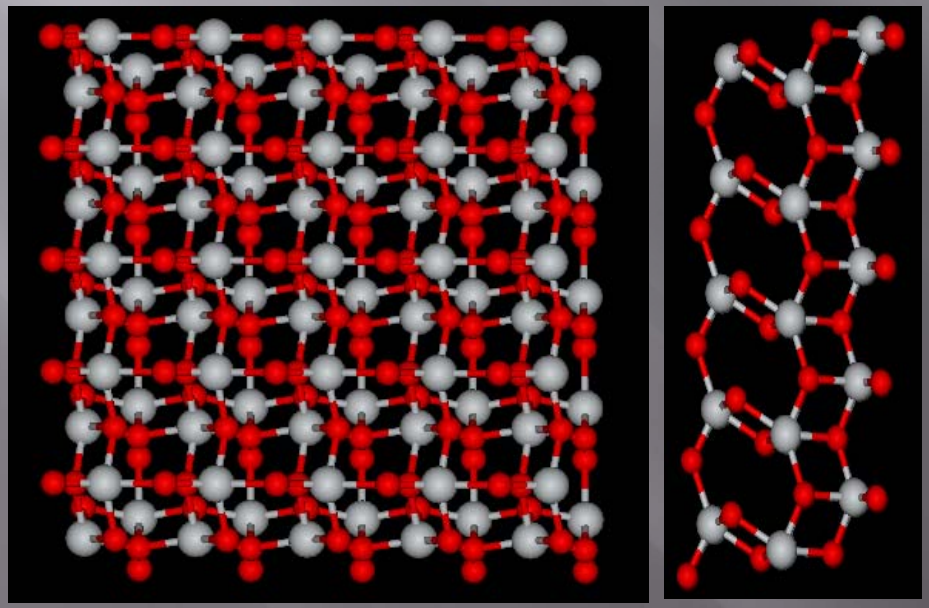
Stages of work II. 2D-structures.



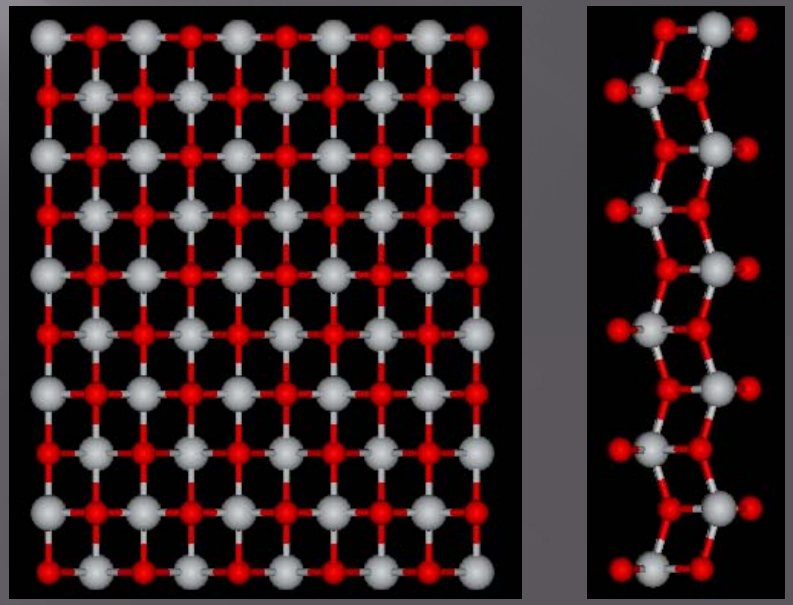
Anatase (101), 3 layers



Anatase (101), 6 layers



Anatase (001), 9 layers



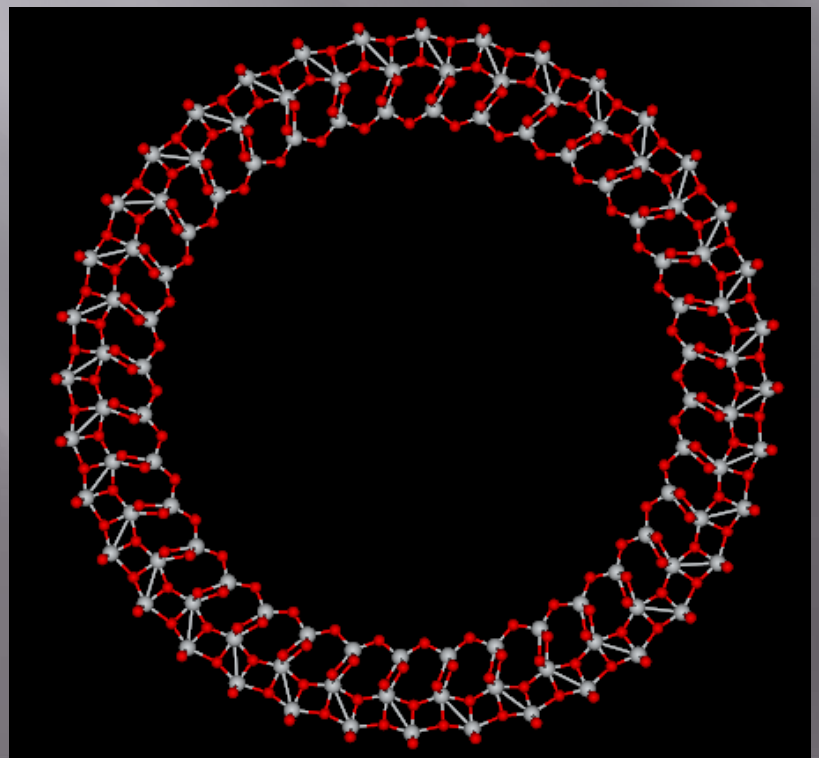
Anatase (001), 6 layers

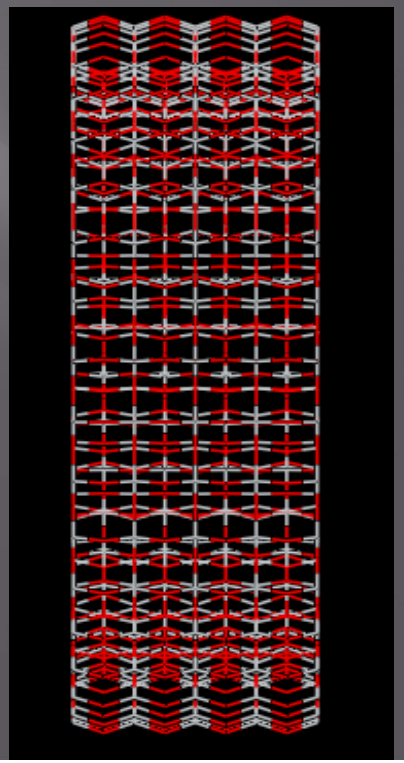
Stages of work IV. Nanotubes.

- Anatase (101), 3 layers (n,n) un (n,0)
- Anatase (101), 6 layers (n,0) un (0,n)
- Anatase (001), 6 layers (n,0) un (0,n)
- Anatase (001), 9 layers (n,0) un (0,n)

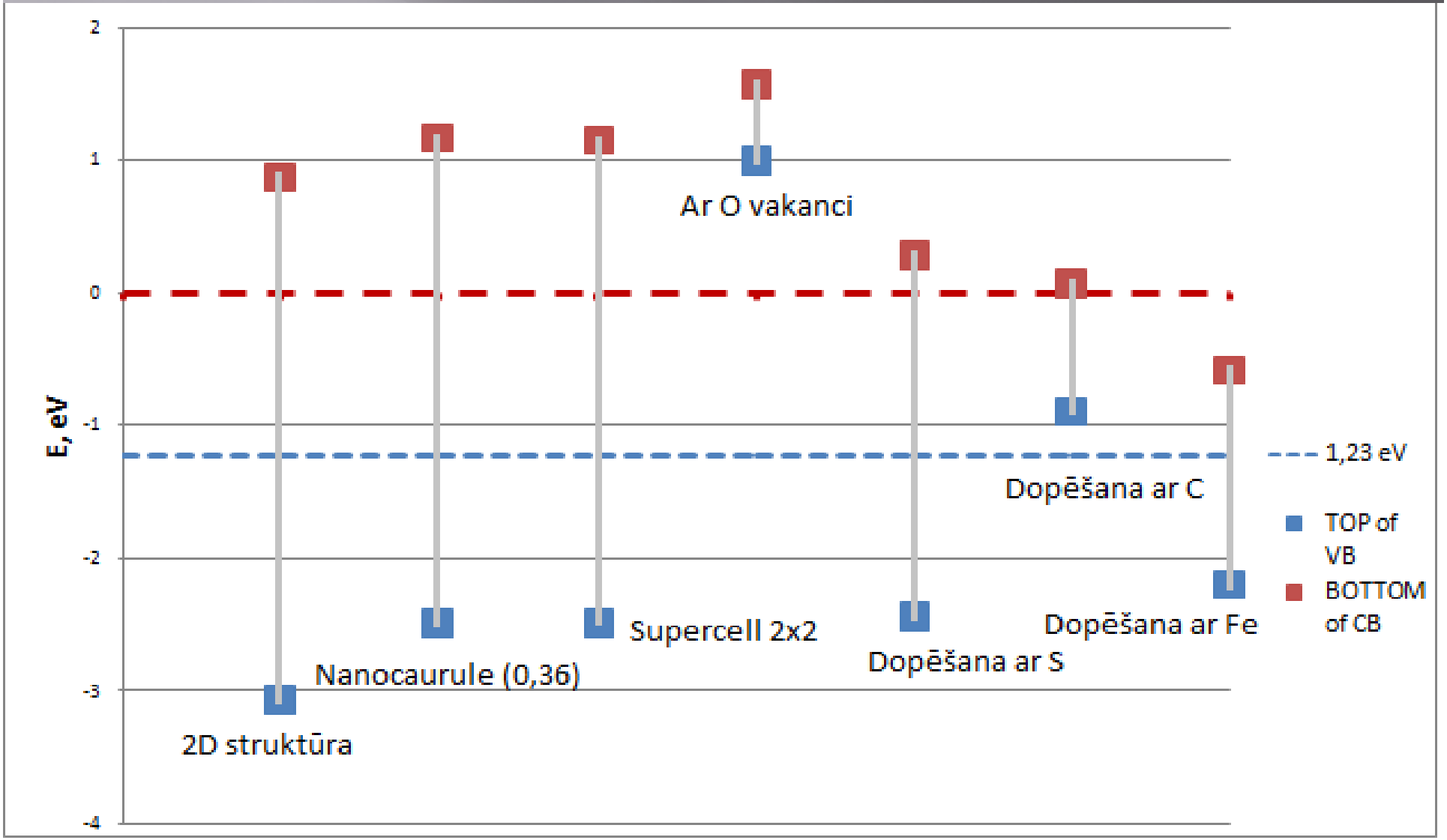
Most advantageous configurations.

- Anatase (001), 9 layers (0,n)
- The result agrees with ones published earlier (for instance, Ferarri paper - J. Phys. Chem. Lett. 2010, 1, 2854–2857)





Stage V. Positions of Valence Bands and Conduction Bands



Conclusions

- The most suitable exchange-correlation functional is chosen and optimized for calculations on TiO_2 nanotubes
- The most stable configuration of TiO_2 nanotubes is found – 9-layer anatase (001) structure with chirality indexes (0,n)
- Nanotubes with S dopants are predicted to possess the highest photocatalytic activity

Thank you for attention!

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