Atomic-scale modeling for solar energy materials

By: Merid Legesse, Michael Nolan, and Giorgos Fagas

Tyndall National Institute

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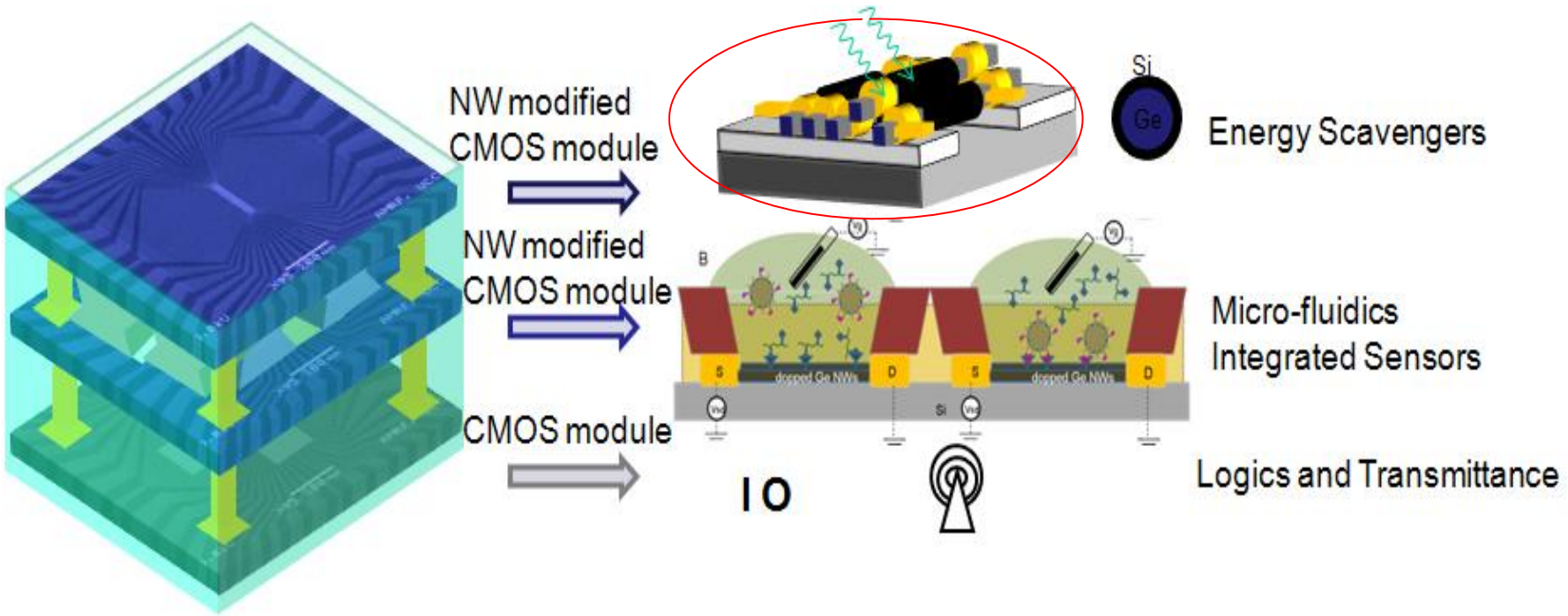
SEVENTH FRAMEWORK PROGRAMME
ucc
Coláiste na hOllscoile Corcaigh, Éire
University College Cork, Ireland

Outline

- The SiNAPS project
- Methods
- Preparation of amorphous silicon(a-Si) and hydrogen amorphous silicon(a-S:H)
- Results
  - Model of (a-Si) and (a-S:H)
  - Density of state of a-Si and (a-S:H)
  - Estimation of band gap
  - Crystalline –amorphous interface (cSi-aSi)
- Summary

Aim of SiNAPS Project

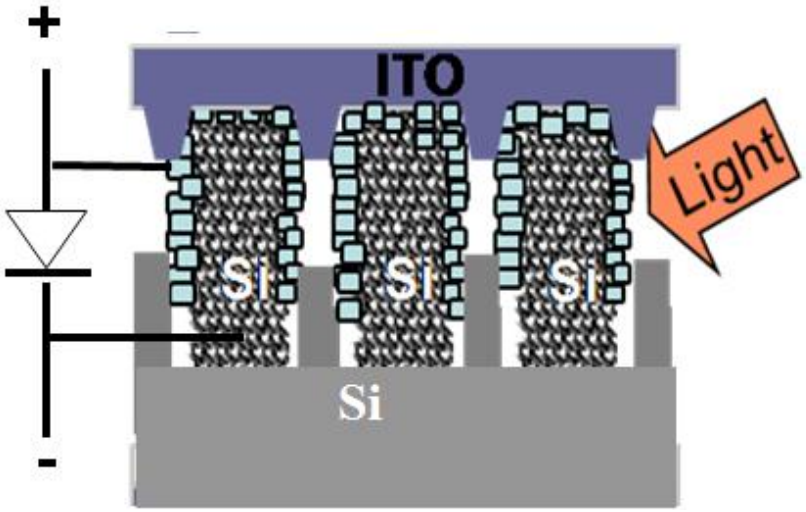
- To develop dust-size chemical sensing platforms that harvest energy from light and miniaturisation below $4mm^3$.

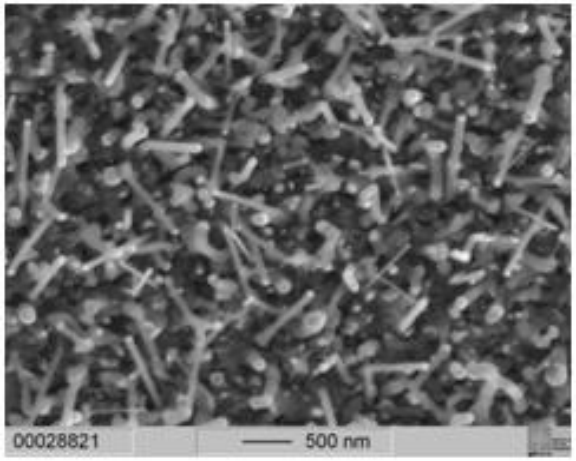


- A cost-effective technology enabling material platform
- Solar energy harvesting at the nanoscale
- Chemical sensing at the nanoscale

Photovoltaic cell

Vertically integrated NW-based PVs





CVD grown Si nanowires with gold caps

- High and uniform charge separation
- Uniform core shell structure
- High efficiency

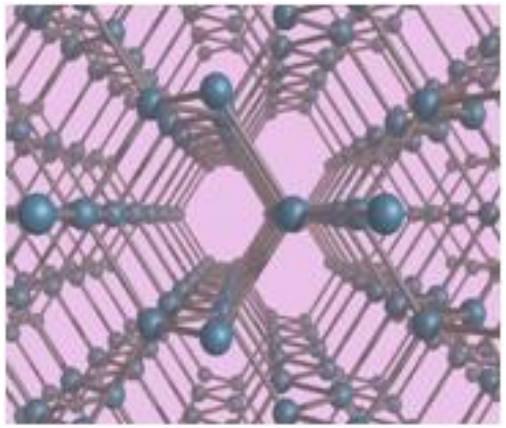
Amorphous silicon solar cell

http://www.ehow.com/how_8747941_make-amorphous-solar-cell.html

Amorphous solid

- Amorphous Solids are made up of randomly orientated atoms , ions, or molecules that do not form defined patterns or lattice structures.

Crystalline Si	Amorphous Si
1.1 eV indirect bandgap	1.4 eV direct bandgap
4-fold coordination	≤ 4-fold coordination; 3-fold and 5-fold
Fixed bond length and angles	Variable bond lengths and angles
Short and long range order	Short range order
Electron mobility 1000 cm^2/Vs	Electron mobility ~1 cm^2/Vs



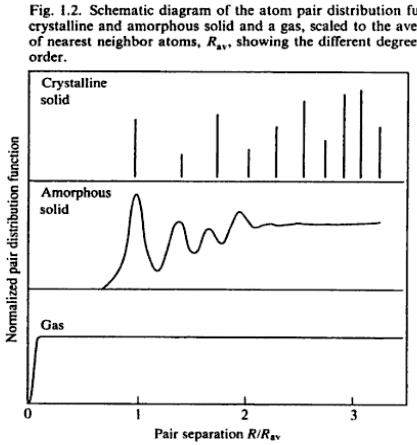
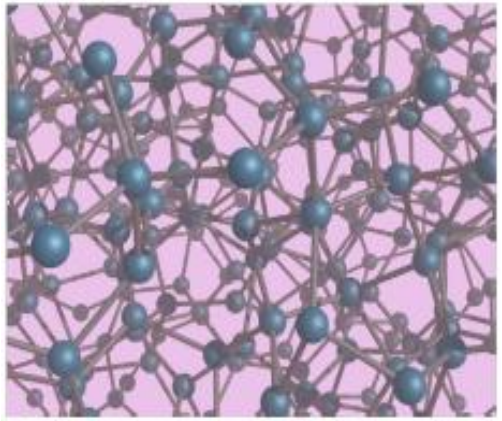


Fig. 1.2. Schematic diagram of the atom pair distribution functions for a crystalline and amorphous solid and a gas, scaled to the average separation of nearest neighbor atoms, R_{av}, showing the different degree of structural order.



"Hydrogenated Amorphous Silicon" By R. A. Street"

Consequence of disorder

STRUCTURE		ELECTRONIC PROPERTIES
❖ Bonding disorder	→	Band tails, localization
❖ Structural defects	→	Electronic states in the band gap

Fig. 1.6. Schematic density of states distribution for an amorphous semiconductor showing the bands, the band tails, and the defect states in the band gap. The dashed curves are the equivalent density of states in a crystal.

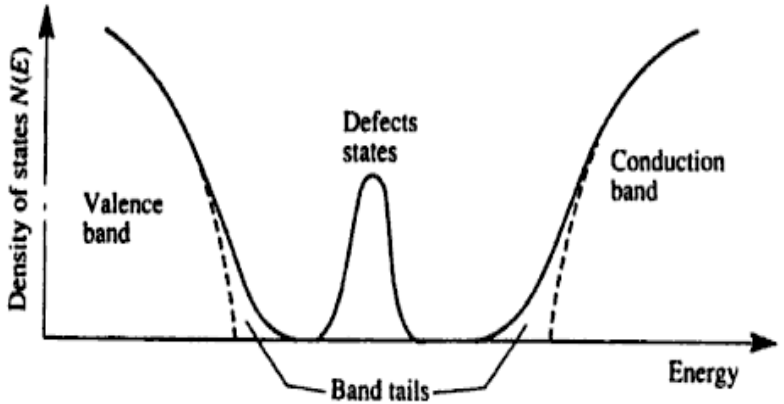
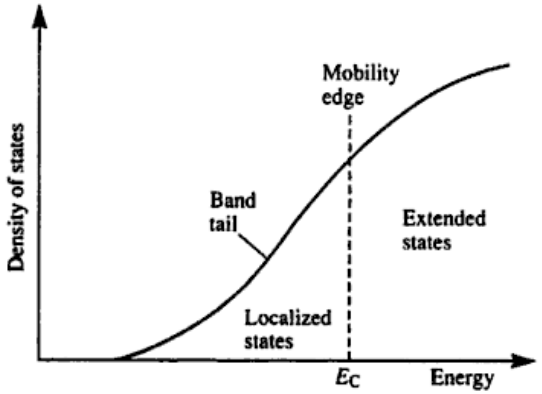


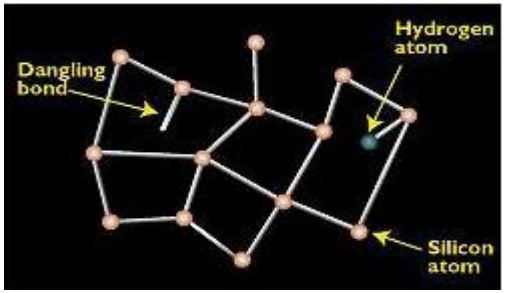
Fig. 1.9. The density of states distribution near the band edge of an amorphous semiconductor, showing the localized and extended states separated by the mobility edge.



"Hydrogenated Amorphous Silicon" By R. A. Street"

Hydrogenated amorphous silicon

- It is generally accepted that hydrogen plays an important role in passivating defects in amorphous silicon.
- It produces an electronic quality material with a high photoconductivity.



Advantages

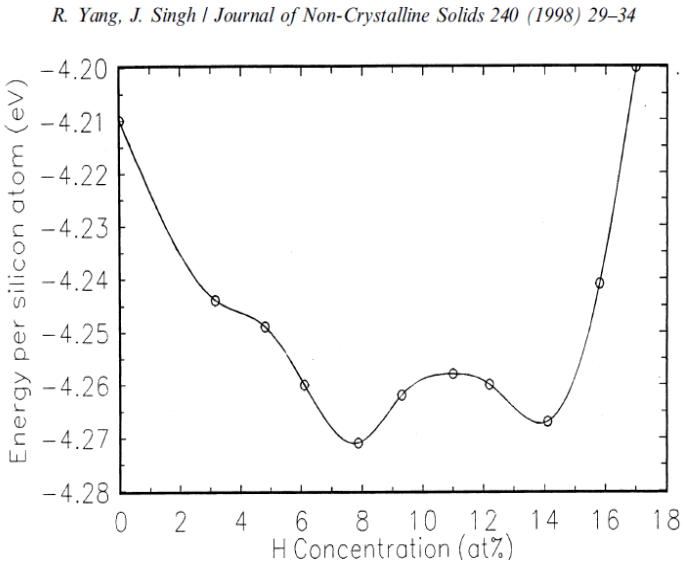
- Low cost glass substrate
- Capability of being deposited on a large area
- Low temperature deposition & fabrication processes

Disadvantages

➡ Electron mobility

$1 cm^2/Vs$

Hydrogenated amorphous silicon



Hydrogen contents in the range 8-15%

However,

- The effect of hydrogen in a-Si:H
- The relationship between hydrogen content and electronic properties a-Si:H
- local bonding environment of hydrogen

Still not well understood

Methods

- We are using General utility lattice programme(GULP) for generating amorphous model.
  - Tersoff Potential

- Vienna Ab initio Simulation Package VASP and Quantum espresso for electronic structure calculation.
  - Density functional theory.

Preparation of amorphous silicon

Optimized structure of Bulk silicon

250 ps
3500 K

Melted structure of Silicon

10^{12} K/s-10^{14} K/s

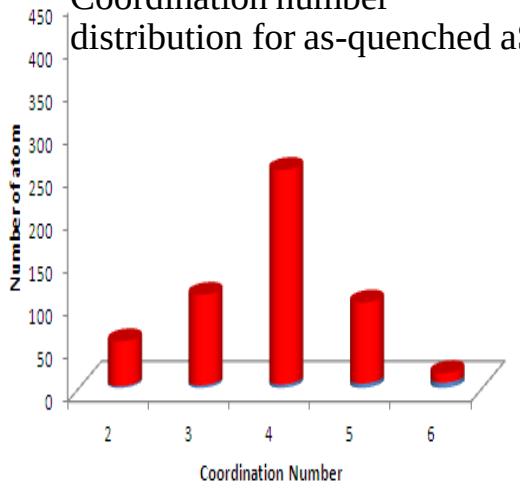
Quenched structure

300 K
100 PS

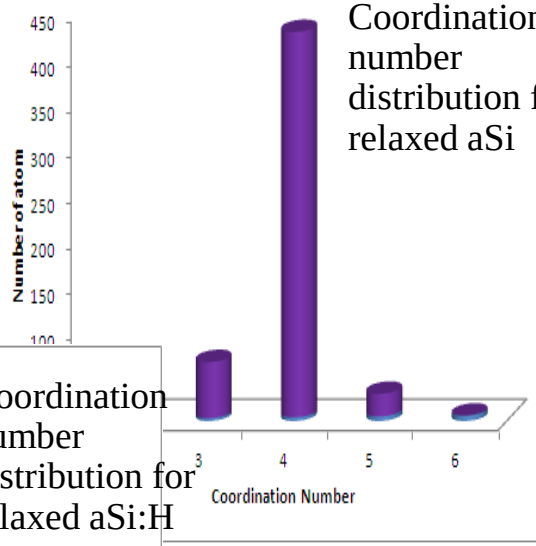
Relax Amorphous Silicon

Relax Hydrogenated amorphous Silicon

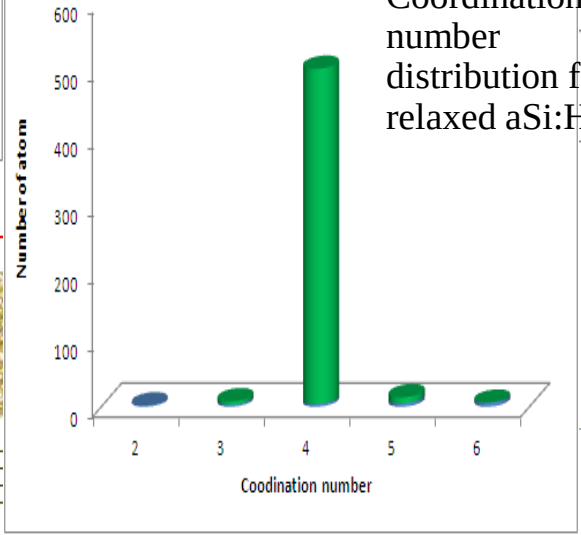
Coordination number distribution for as-quenched aSi



Coordination number distribution for relaxed aSi



Coordination number distribution for relaxed aSi:H



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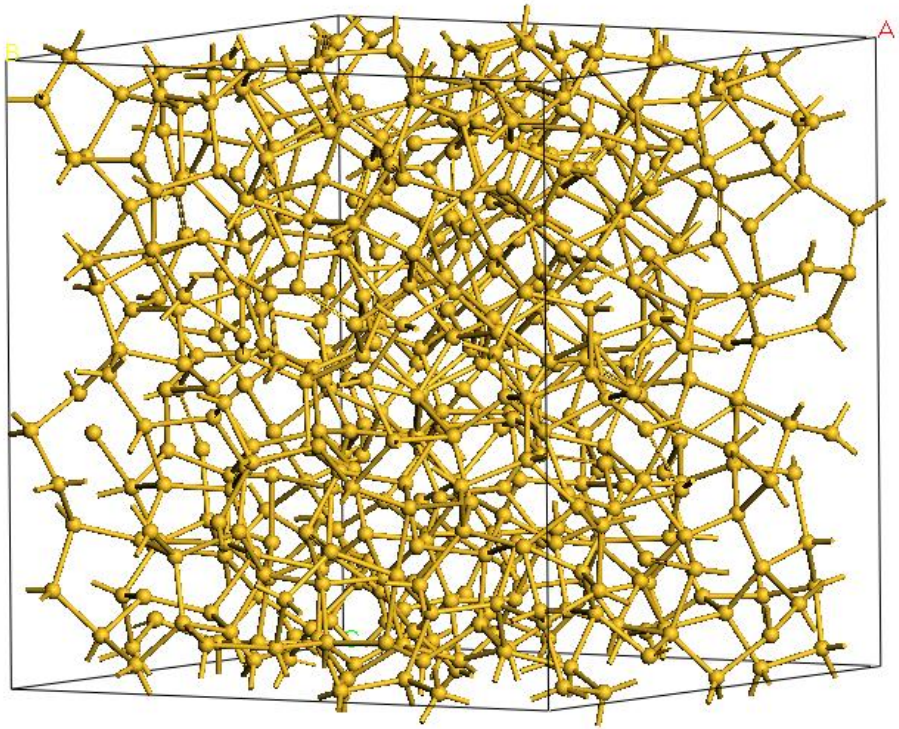
Structure

a-Si



a-Si:H

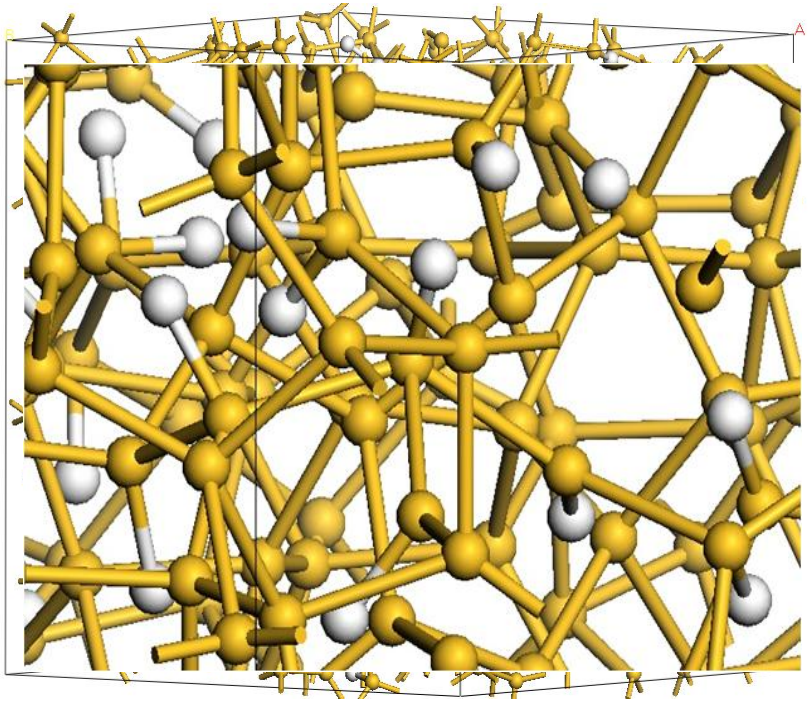


Fig. 1.2: (a) 512 atoms of bulk a-Si and (b) 512 and 62 H atoms of bulk a-Si:H after quenching to 300 K from a melt at 3500 K.

Radial distribution function (RDF)

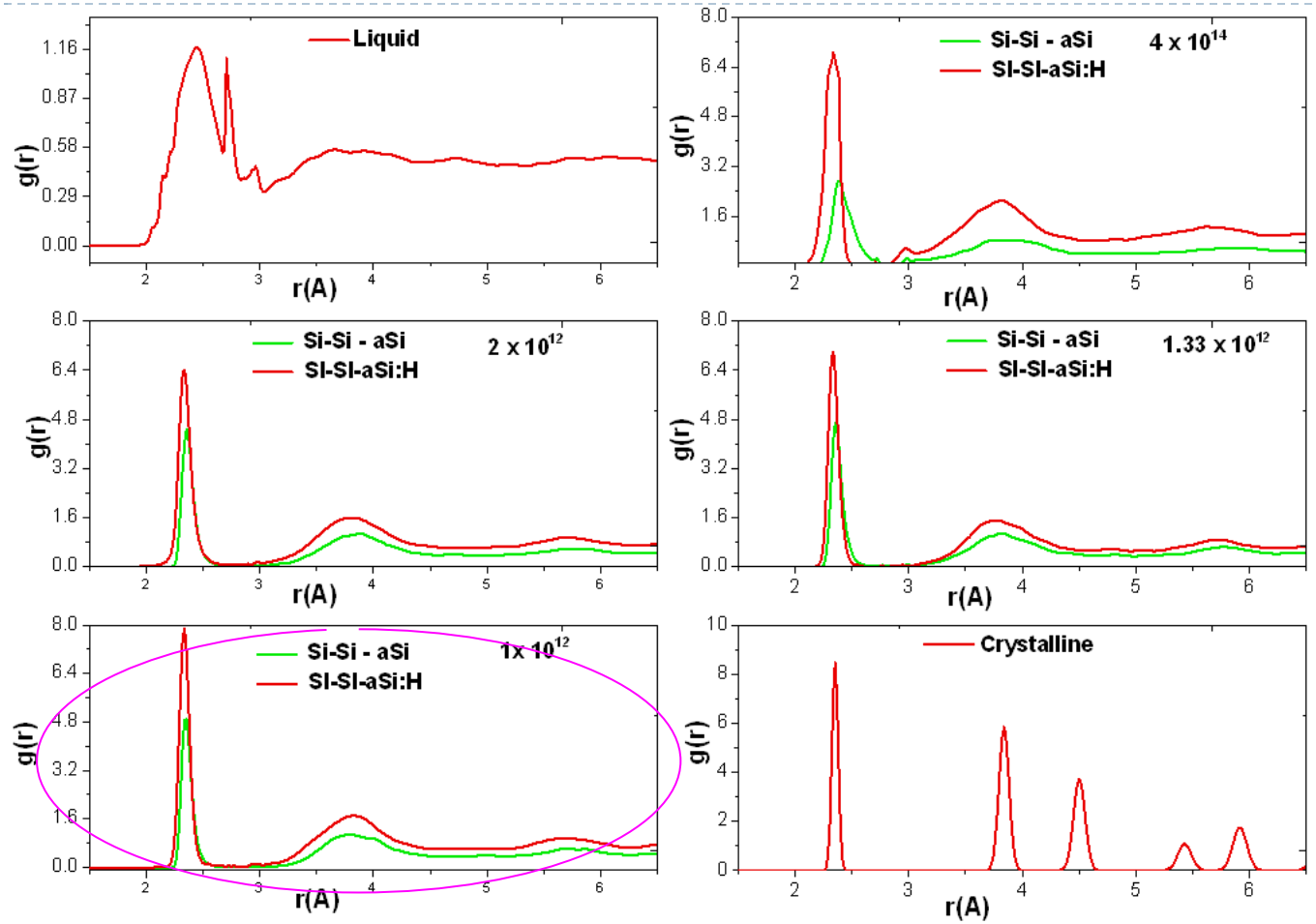
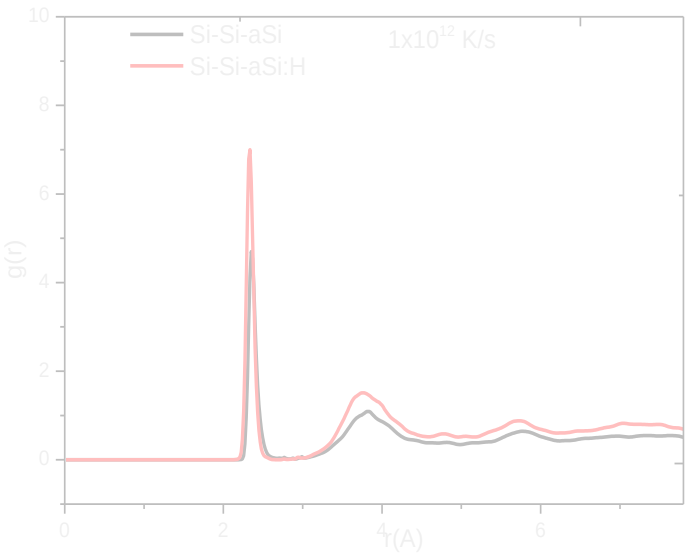
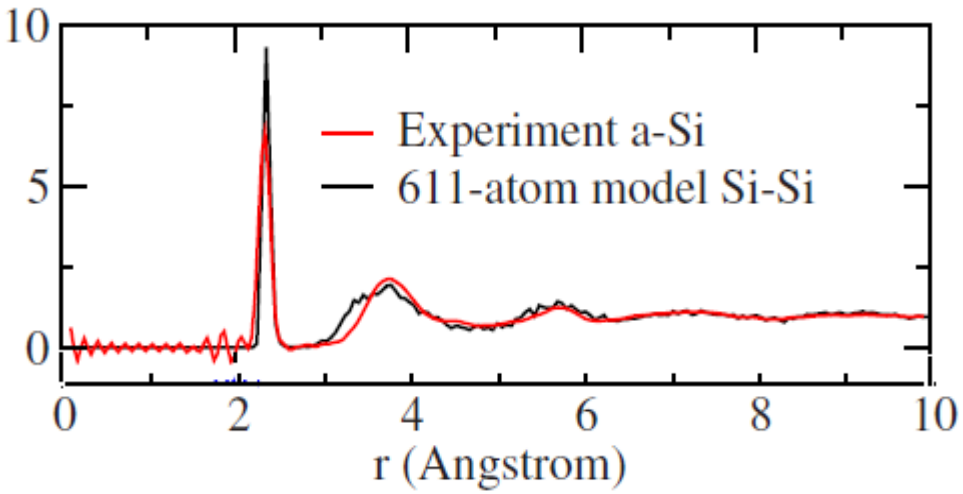


Fig. 1.3: Quench rate dependent RDF for 512 atom bulk Si (a) melting temperature (b) quench rate $4x10^{14}$ K/s (c) quench rate $2x10^{12}$ K/s (d) $1.3x10^{12}$ K/s (e) quench rate $1x10^{12}$ K/s (f) crystalline.

Radial distribution function (RDF)

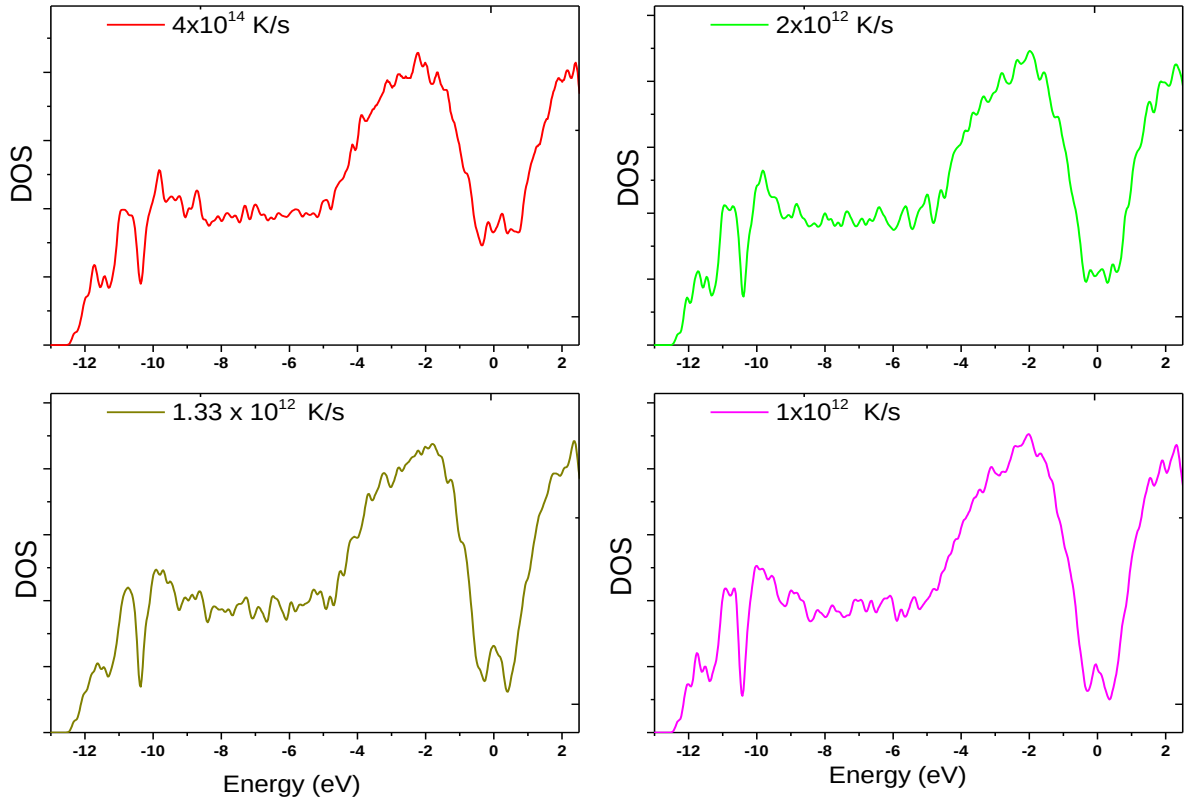


RDF of a-Si:H at the quench rate of $1X10^{12}$ K/s



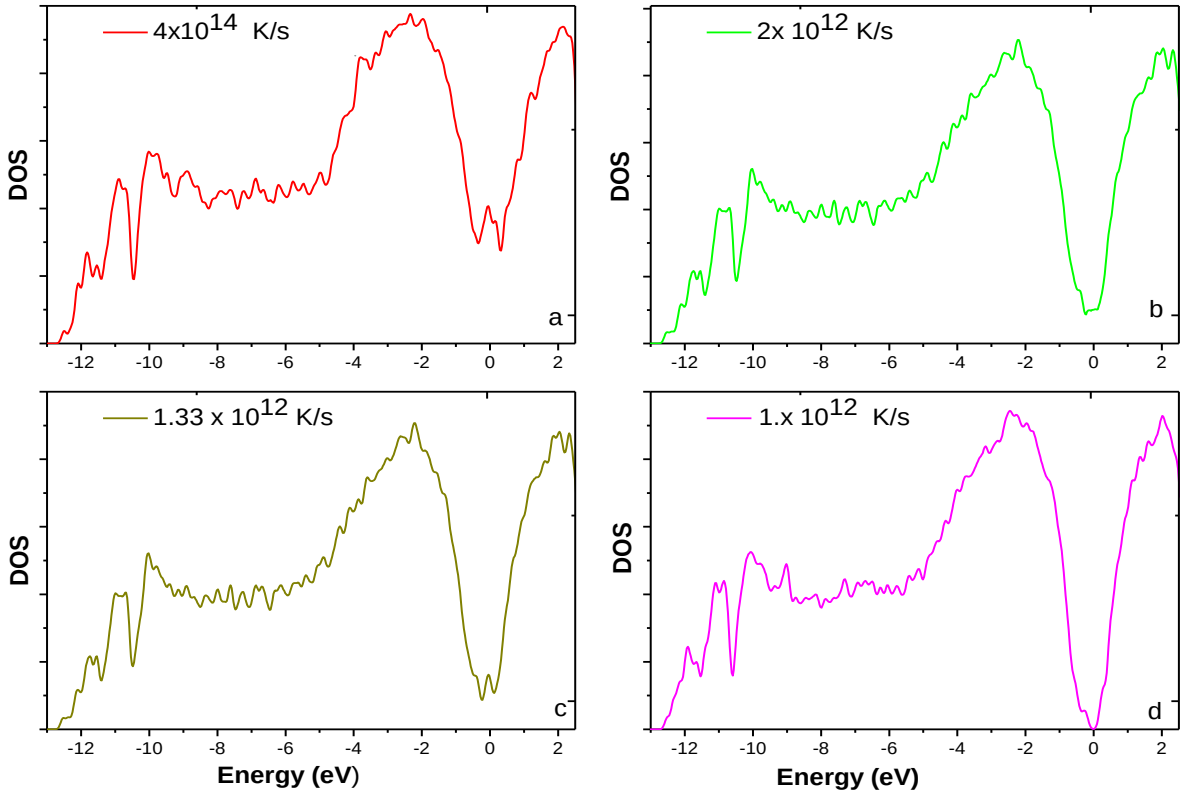
Parthapratim Biswas, and D. A. Drabold
PHYSICAL REVIEW B **76, 125210 (2007)**

DOS of a-Si after Relaxation



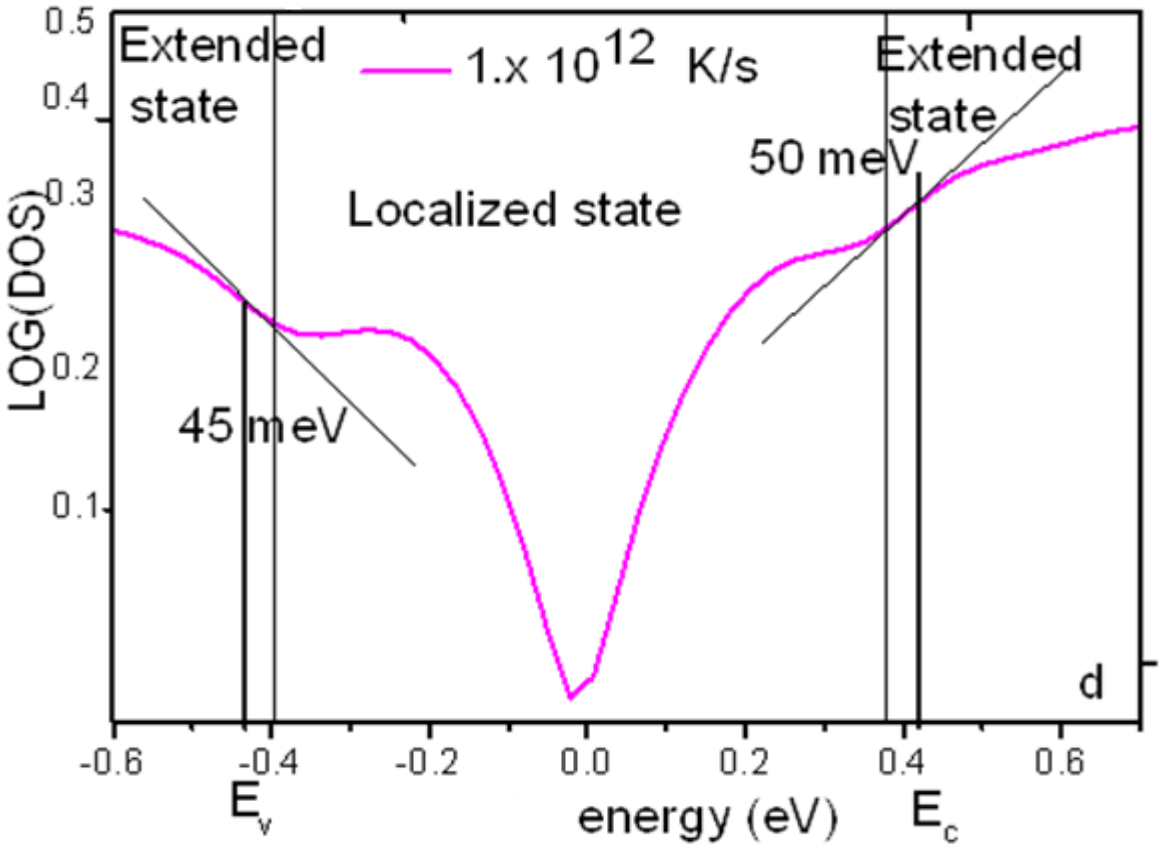
Total density of states of 512 atom bulk a-Si at different quench rates after VASP relaxation. The Fermi level is at 0 eV.

DOS of a-Si:H



Total DOS for a-Si:H from structure generated from aSi produced a different quenched rates (a) 512 atom aSi and 21% H (b) atom aSi and 18% H (c) atom a Si and 15% H (d) atom aSi and 12% H. The Fermi level is shifted to 0 eV.

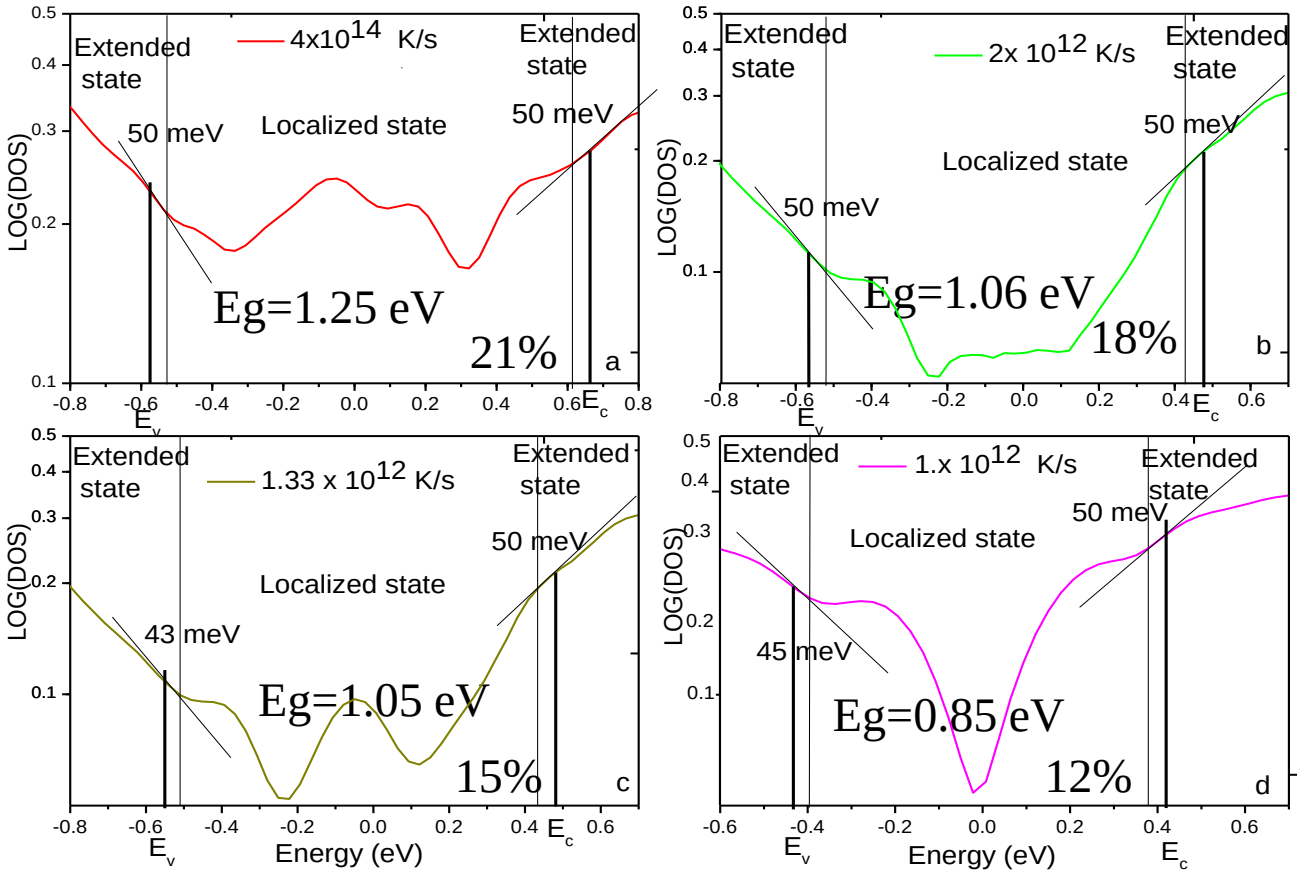
Band tails of a-Si:H



E_g=0.85 eV (DFT)

Semi-log plot of the total DOS in the gap region for a-Si:H. Valance band energy (E_v) and conduction band energy (E_c) mobility edge. The Bold solid lines indicate the edge of valance and conduction band.

Band tails of a-Si:H

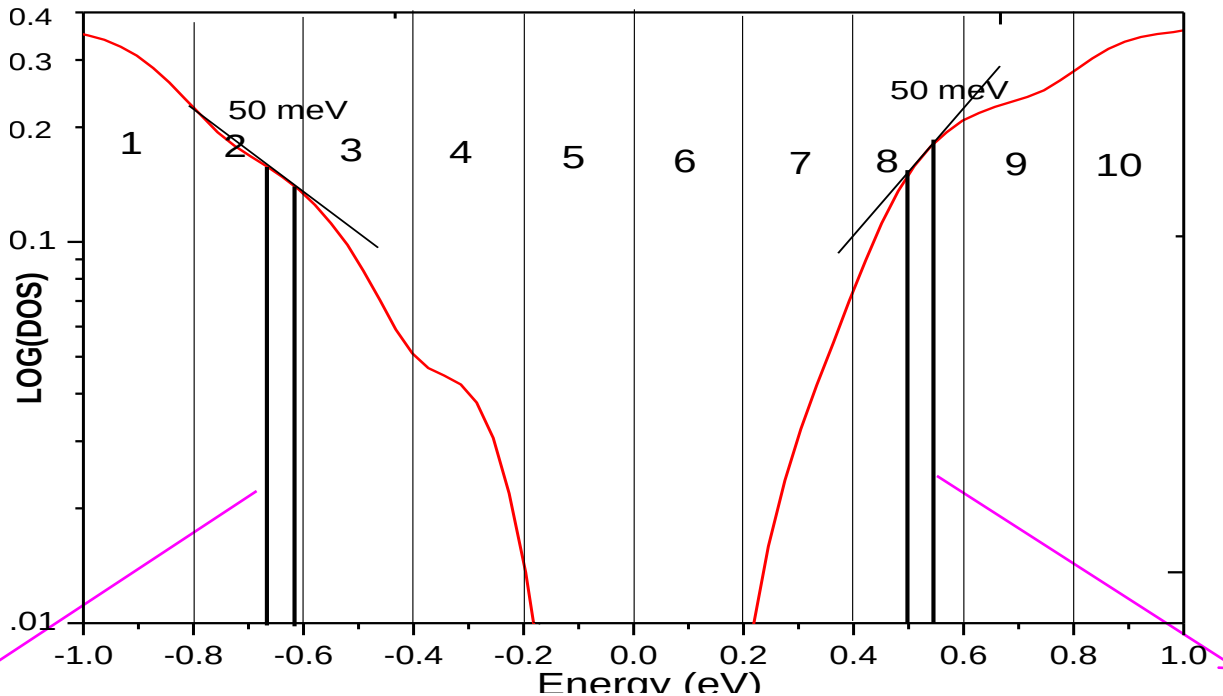


Semi-log plot of the total DOS in the gap region for a-Si:H. Valance band energy (E_v) and conduction band energy (E_c) mobility edge. The Bold solid lines indicate the edge of valance and conduction band.

Semi-log plot of the total DOS of aSi:H

Quench-rate = 1.0 X 10^{12} K/s 512-a-Si +14%H

Energy gap =1.2 eV + 0.4 eV(DFT error) =1.6 eV

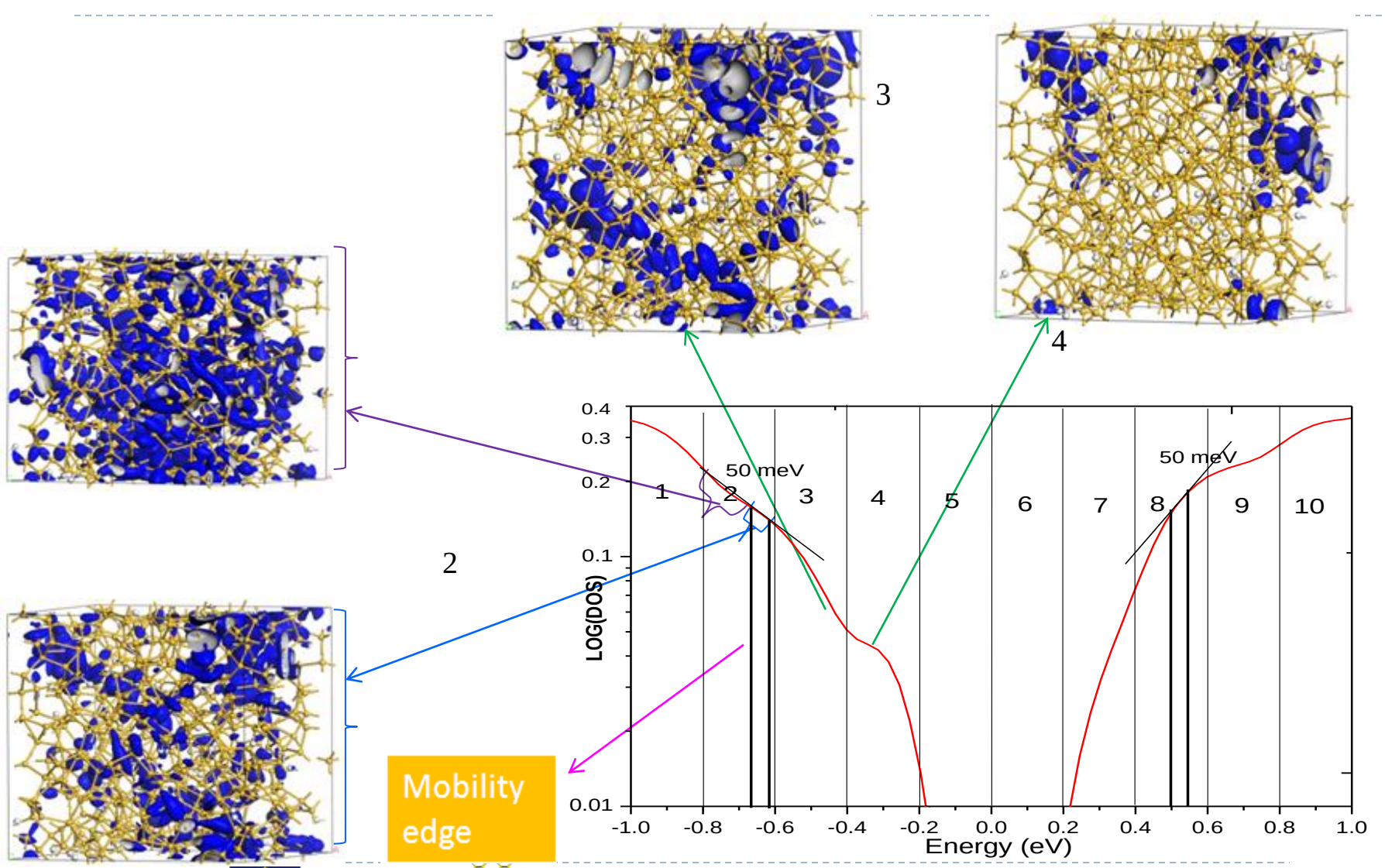


Mobility edge

Mobility edge

1.2 eV

Charge density analysis

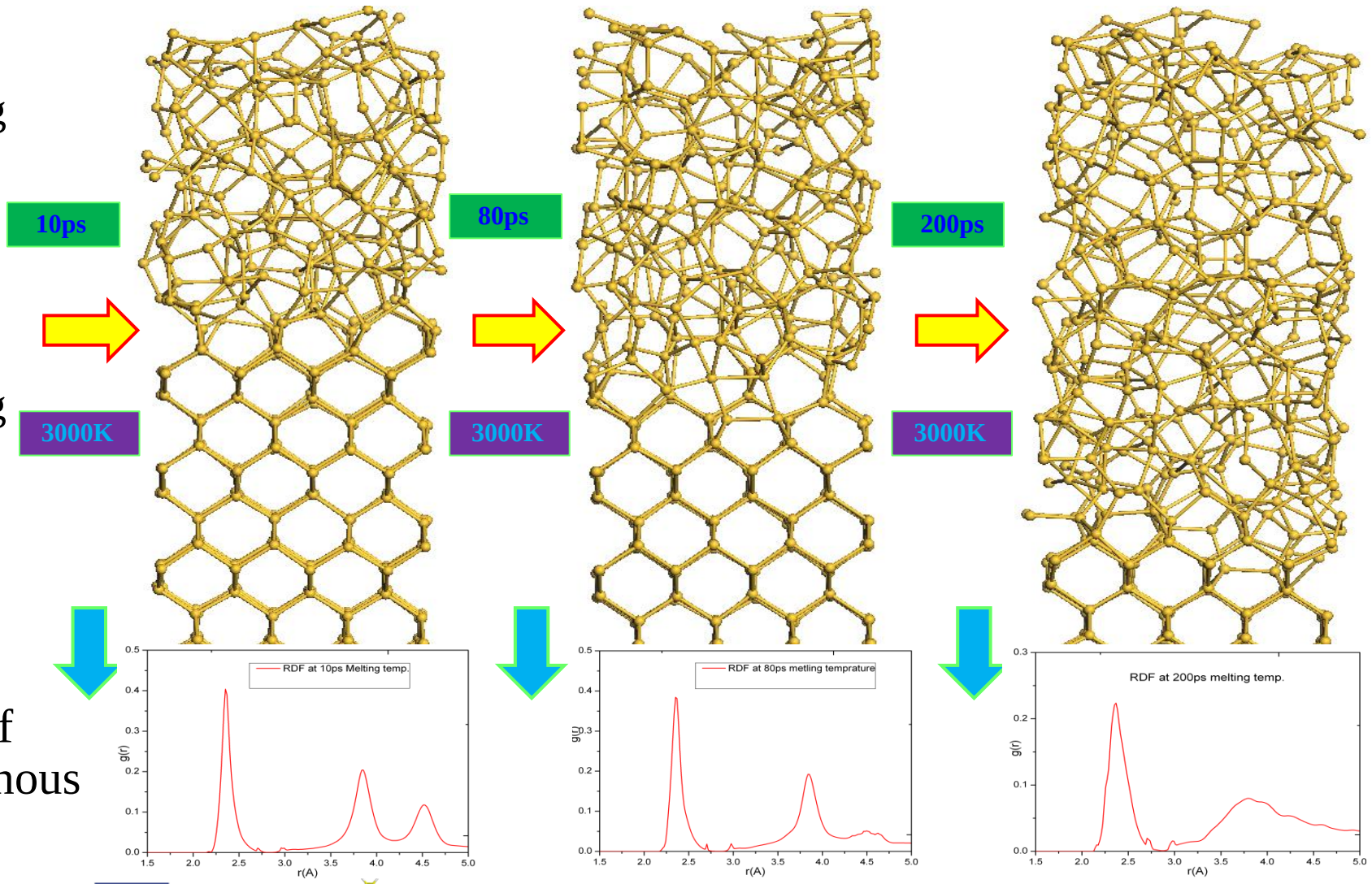


Different thickness layer of amorphous on crystalline Si

Melting time

Melting Temp.

RDF of amorphous region



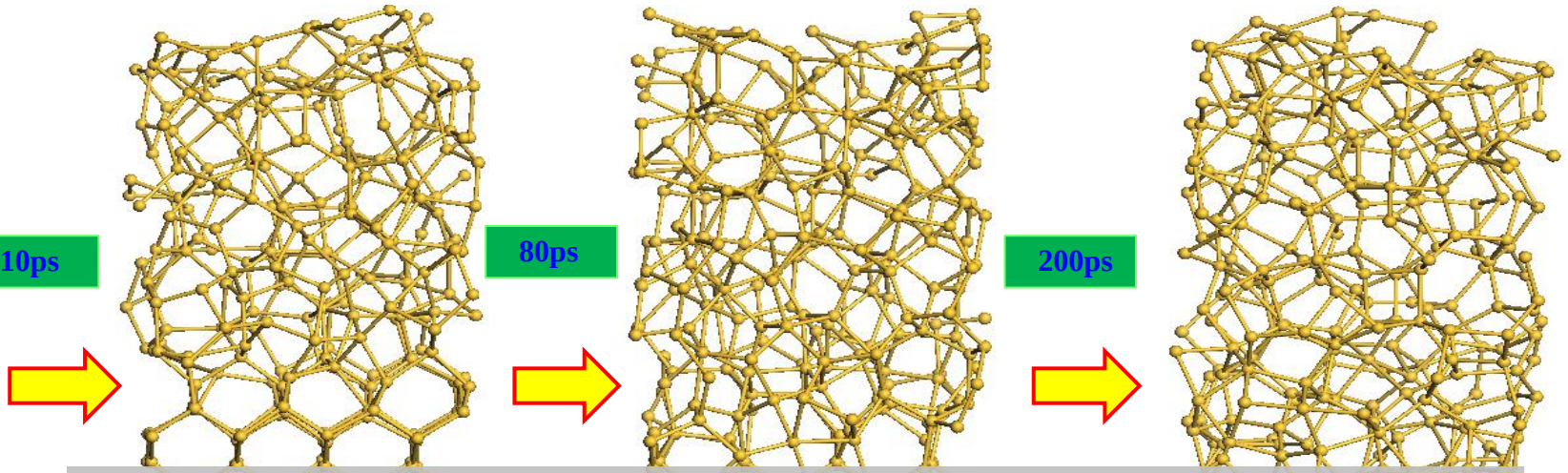
Different thickness layer of amorphous on crystalline Si

Melting time

10ps

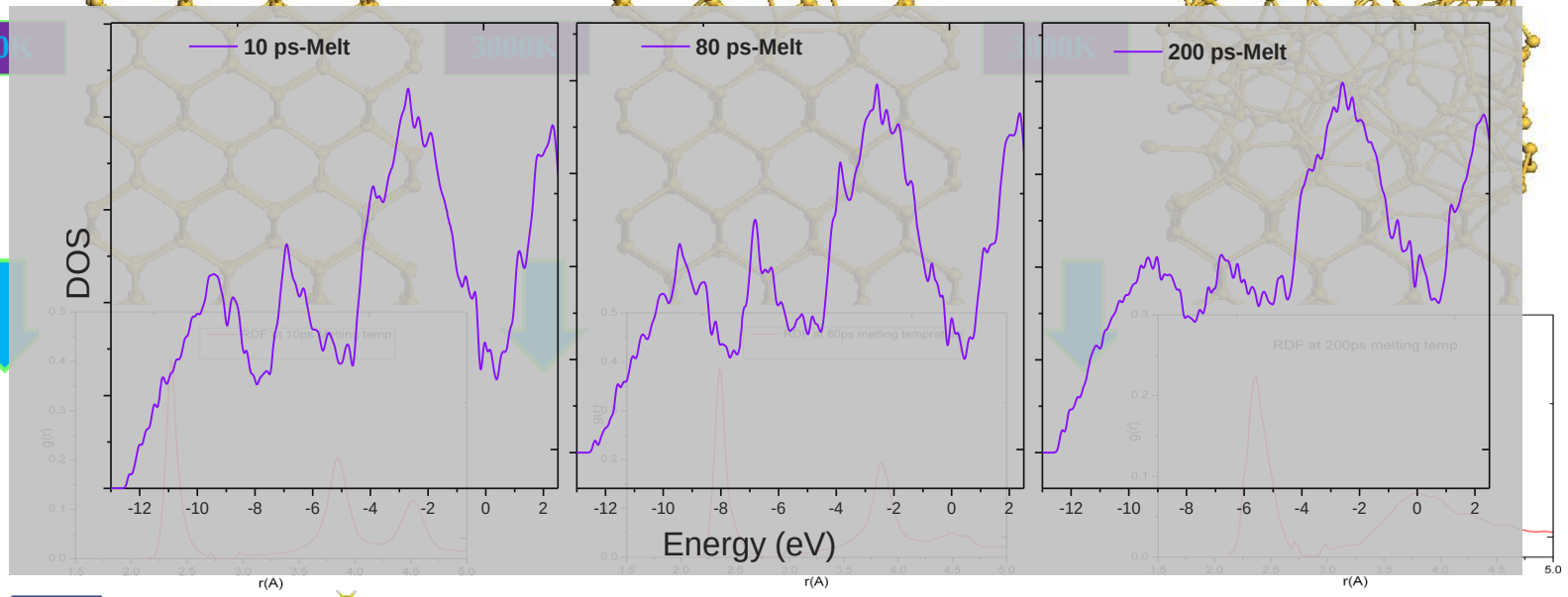
80ps

200ps



Melting Temp.

3000K



RDF of amorphous region

Summary

- we reproduced the experimental result of RDF
- The DOS of a-Si shows the introduction of new electronic states in the band gap arising from formation of dangling silicon and distortion of angles and bonds.
- The amount of new states depends on the cooling rate in the quenching simulations.
- The DFT relaxed structure gives less defective amorphous Si
- Hydrogen atoms saturate the dangling silicon bonds
- The slowest quench rate ($1x10^{12}$ K/s) gives good quality amorphous silicon and 14% H concentration saturates all the dangling bonds.

Acknowledgement

- Dr. Michael Nolan
- Dr. Giorgos Fagas
- Dr. Fritz Falk IPHT

- SiNAPS Project
- ICHEC and SFI for computing

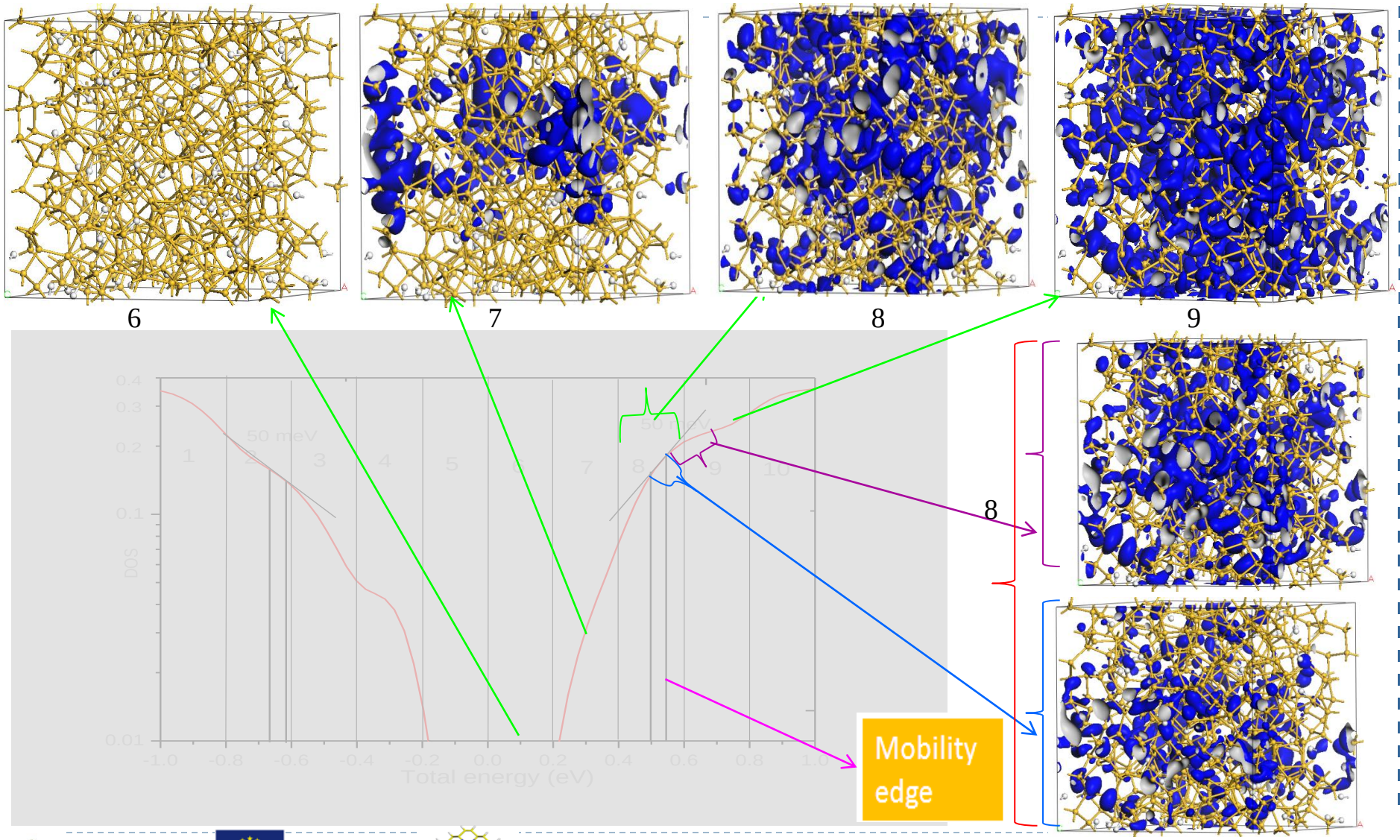
Thank you for your attention

Coordination number

Quench rate (K/s) a-Si	3-fold Coordinated (%)	4-fold Coordination (%)	5-fold coordinated (%)	Average coordination Number Expt. {3.90}
4×10^{14}	21	51	23	3.81
2×10^{12}	18	75	12	3.87
1.3×10^{12}	15	78	12	3.88
1×10^{12}	12	83	5	3.92
1×10^{12} (a-Si:H)	Less than 1%	98	1	3.96

Table: 1 *Average coordination number of Si for different quench rates for 512 atom bulks amorphous Si supercell*

Electronic Structure



Amorphous-crystalline Si Interface

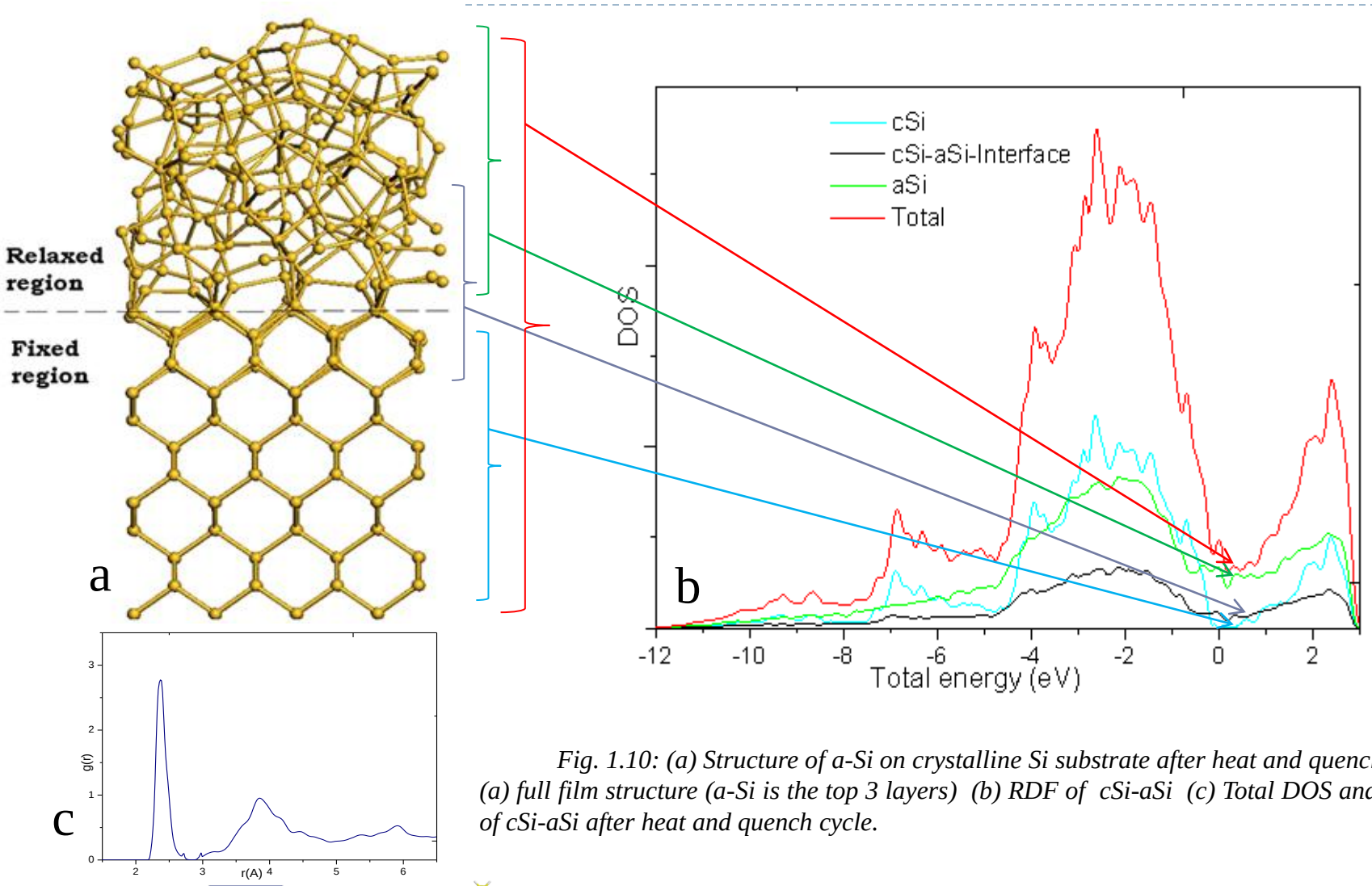


Fig. 1.10: (a) Structure of a-Si on crystalline Si substrate after heat and quench cycle. (a) full film structure (a-Si is the top 3 layers) (b) RDF of cSi-aSi (c) Total DOS and PDOS of cSi-aSi after heat and quench cycle.

DOS of Different H concentration of aSi:H

Quench-rate
=1.0 X 10^{12} K/s