From Femtoseconds to Gigaseconds: The SolDeg Project to Analyze Si Heterojunction Cell Degradation with Machine Learning

I. Degradation in Si-only heterojunctions
II. Experimental degradation study
III. Development of a Machine-Learning based Si-H interatomic potential
IV. Degradation in c-Si/a-Si:H heterojunctions

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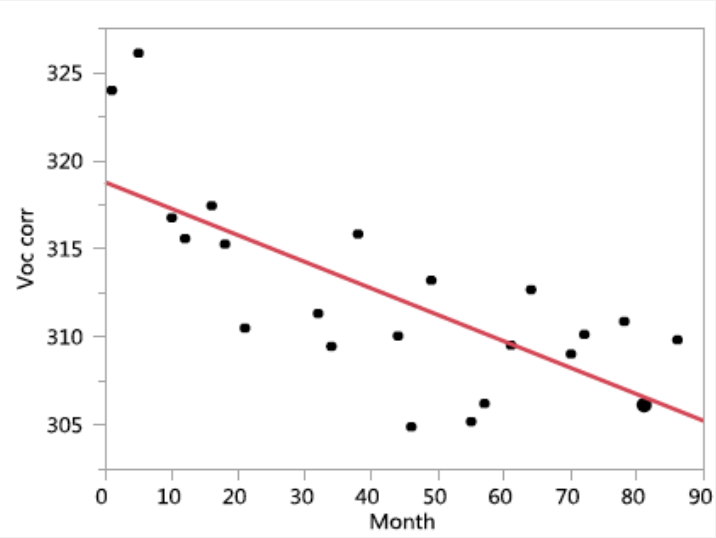
Motivation

1. Si Heterojunction cells hold Si world record efficiency of 26.7%

2. Some groups reported efficiency degradation of 1%/yr, twice the usual. 0.5%/yr was attributed to Voc.

3. Eliminating this 0.5%/yr degradation would have the effect of increasing the efficiency by close to 2% in terms of LCOE, based on the System Advisor Model



D. Jordan et al., IEEE J. of Photovoltaics, 8, 177 (2018)

4. Bertoni: "The defects did it!"
Investigated c-Si/a-Si stacks. Reported strong increase of SRV over 28 months. Attributed it to the increase of defect density at the interface.

On the other hand, the modeling reveals that during the 28-month storage period the density of defect states at the interface, N_S, has increased by an order of magnitude, that is $(5.5 \pm 1.5) \times 10^{7}\ cm^{-2}$. This N_S variation results in a strong SRV

Bernardini and Bertoni Phys. Status Solidi A 2018, 1800705

I. The SolDeg platform

We developed the SolDeg platform to analyze the formation of defects at the c-Si/a-Si interface
This requires

- connecting extreme time scales from femtoseconds to gigaseconds (20 years)
- simulation of large number of large samples with extreme precision

1. Create c-Si/a-Si stacks

2. Generate shocked clusters at the c-Si/a-Si interface as likely hosts of electronic defects

3. Identify shocked clusters that actually host electronic defects

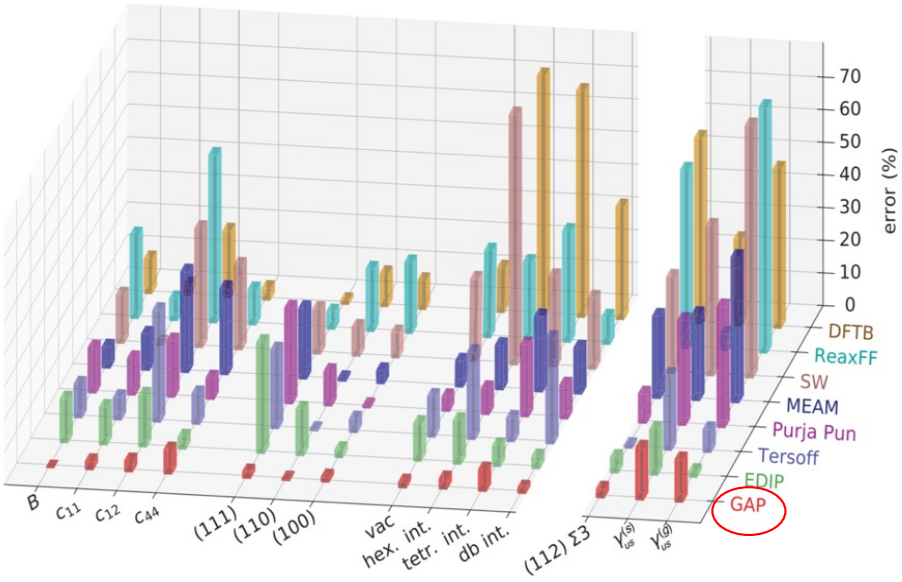
4. Determine the energy barriers that control the generation of these electronic defects

5. Determine the distribution of these barriers

6. Determine the defect generation from the energy barrier distribution

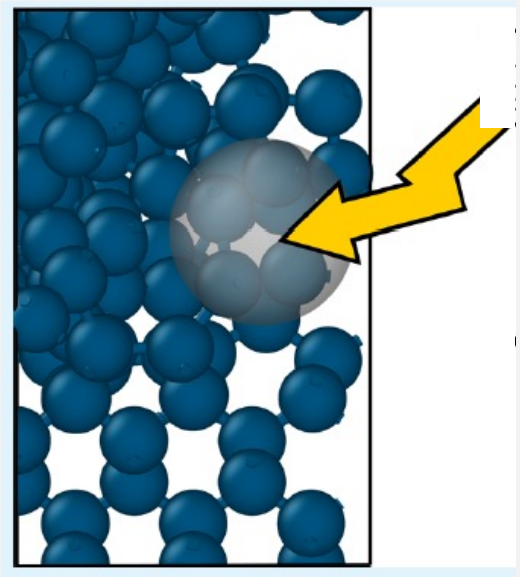
Extreme Precision by Machine Learning-developed Potentials

1. Create c-Si/a-Si stacks using Molecular Dynamics with Machine-Learning (ML)-developed Si-Si potential: Gaussian Approximation Potential Si GAP.



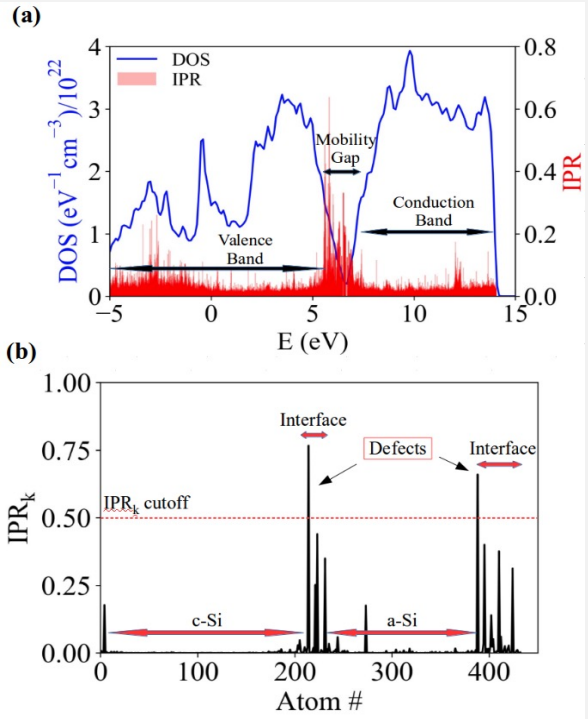
Comparison of GAP with other interatomic potentials.

2. Generate shocked clusters at the c-Si/a-Si interface with "cluster-blaster" as likely hosts of electronic defects



Connect decades of time scales: Nudged Elastic Band method

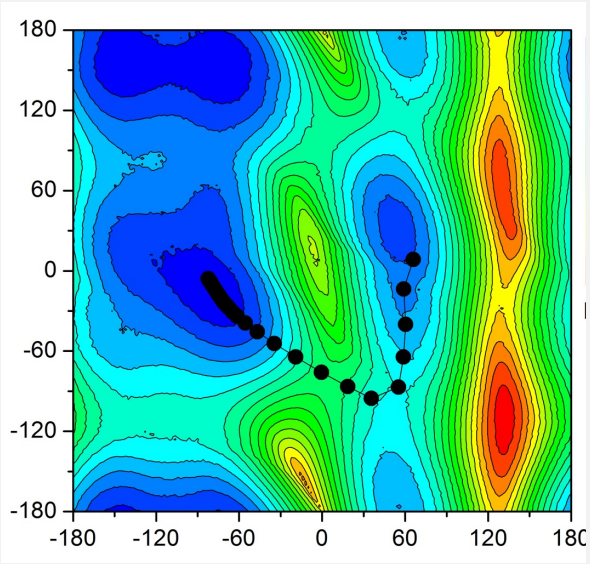
3. Identify shocked clusters that actually host electronic defects: IPR



(a) IPR as a function of energy, (b) IPR as a function of z-depth

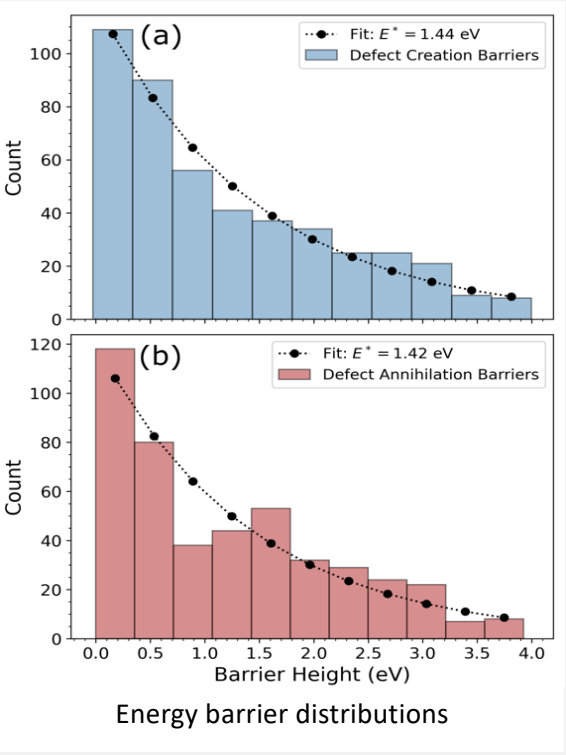
4. Determine energy barriers that control the generation of the electronic defects

Nudged Elastic Band method to determine energy barriers



exp $^{(1eV/300K)}$ ~ 10^{13}

5. Determine the barrier distribution

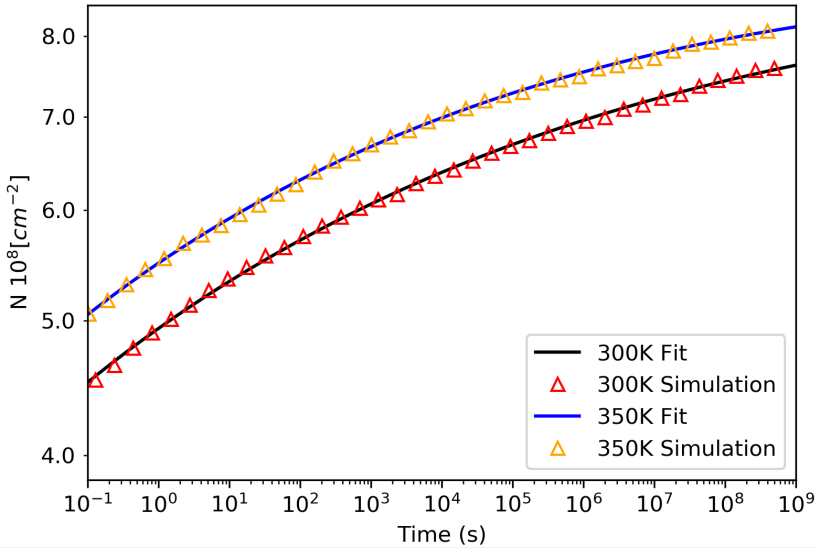


Energy barrier distributions

Determine and Analyze Dynamics of Defect Generation

6. Determine the defect generation from the energy barrier distribution

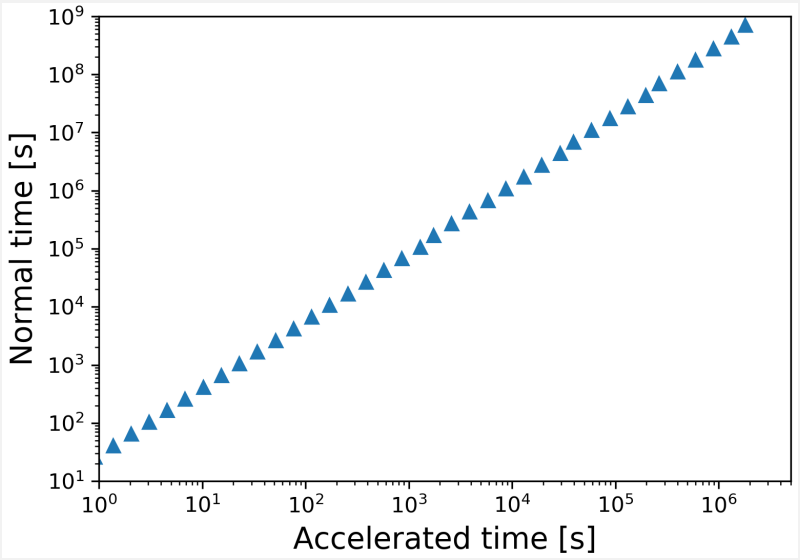
Accelerated Superbasin Kinetic Monte Carlo



$$N(t) = N_{sat}(1 - \exp[-(t/\tau_0)^{\beta}])$$

Stretched exponential fits very well

Accelerated testing by elevated temperature:
Quantitative correspondence?
Stretched exponential predicts:
t(norm) ~ t(acc)$^{(T(acc)/T(norm)}$



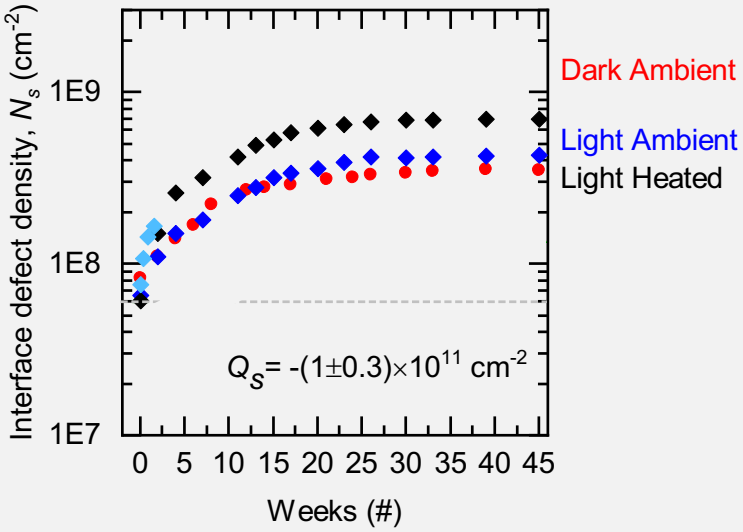
t(norm) ~ t(acc)^s
s = 0.85 = T(acc)/T(norm)

Remarkable confirmation

II. Experimental analysis of SHJ degradation

Bertoni, Mazoor:

- Created c-Si/a-Si:H stacks with 12% H
- Extracted surface recombination velocity using the temperature- and injection-dependent lifetime spectroscopy technique over 45 weeks

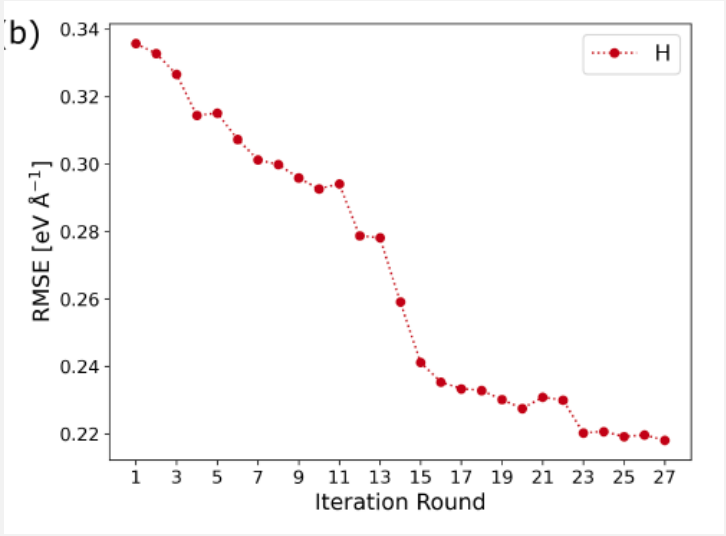


- N(t) well-fitted with stretched exponential
- Stretching exponent bigger than in Si-only simulation. Probably because barrier for H migration is much smaller than for Si migration.

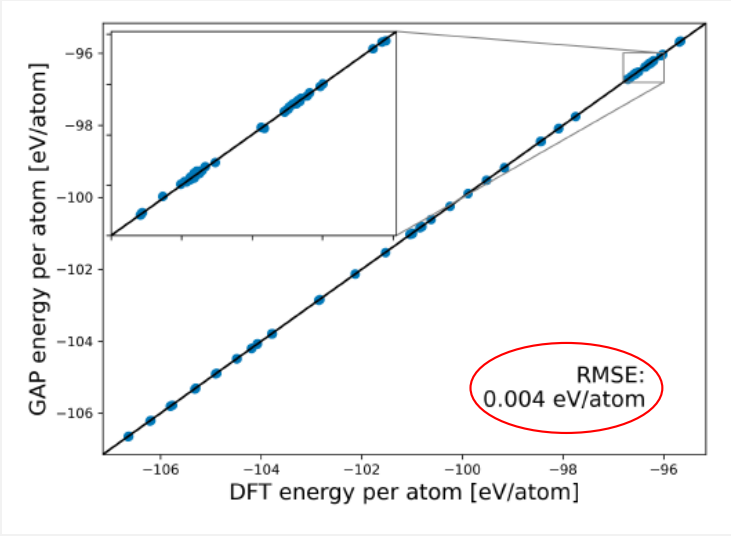
III. For SolDeg for c-Si/a-Si:H – Need Machine-Learning based Si-H GAP

Problem: no Machine Learning-based Si-H (GAP) potential
We developed the world's first Si-H Machine Learning-based GAP potential

Iteration	Structure Type
1	Optimized structures (all phases)
2	Optimized structures (all phases)
3	Low T anneal of a-Si:H
4	High T anneal of liq-Si:H
5	High T anneal of liq-Si:H
6	Med T anneal (1100K) of a-Si:H
7	Heating a-Si:H from 500K to 800K at 10^{13} K/s
8	Heating a-Si:H from 800K to 1100K at 10^{13} K/s
9	Heating a-Si:H from 1100K to 1400K at 10^{13} K/s
10	Heating a-Si:H from 1100K to 1400K at 10^{13} K/s
11	Heating a-Si:H from 800K to 1400K at 10^{12} K/s
12	Added new a-Si:H structures
13	Add new a-Si:H structures
14	Added c-Si/a-Si:H interface structures
15	Added c-Si/a-Si:H interface structures
16	Added new c-Si divacancy structures
17	Added new liq-Si:H structures
18	Added new c-Si vacancy structures
19	Added new c-Si interstitial structures
20	Low T anneal of c-Si/a-Si:H interface structures
21	Optimization of c-Si/a-Si:H interface structures
22	NPT high T anneal of liq-Si:H structures
23	NVT high T anneal of liq-Si:H structures
24	Quenching liq-Si:H from 2000K to 1500K at 10^{13} K/s
25	Annealing quenched liq-Si:H structures at 1500K
26	Quenching liq-Si:H from 1500K to 1400K at 10^{12} K/s
27	Added hydrogen passivated c-Si surface (100) and c-Si surface (111) structures

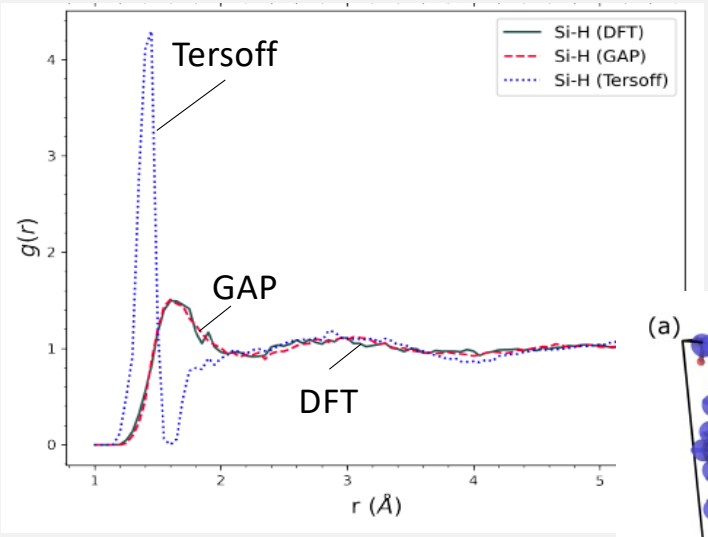


Forces: Deviation from DFT substantially reduced



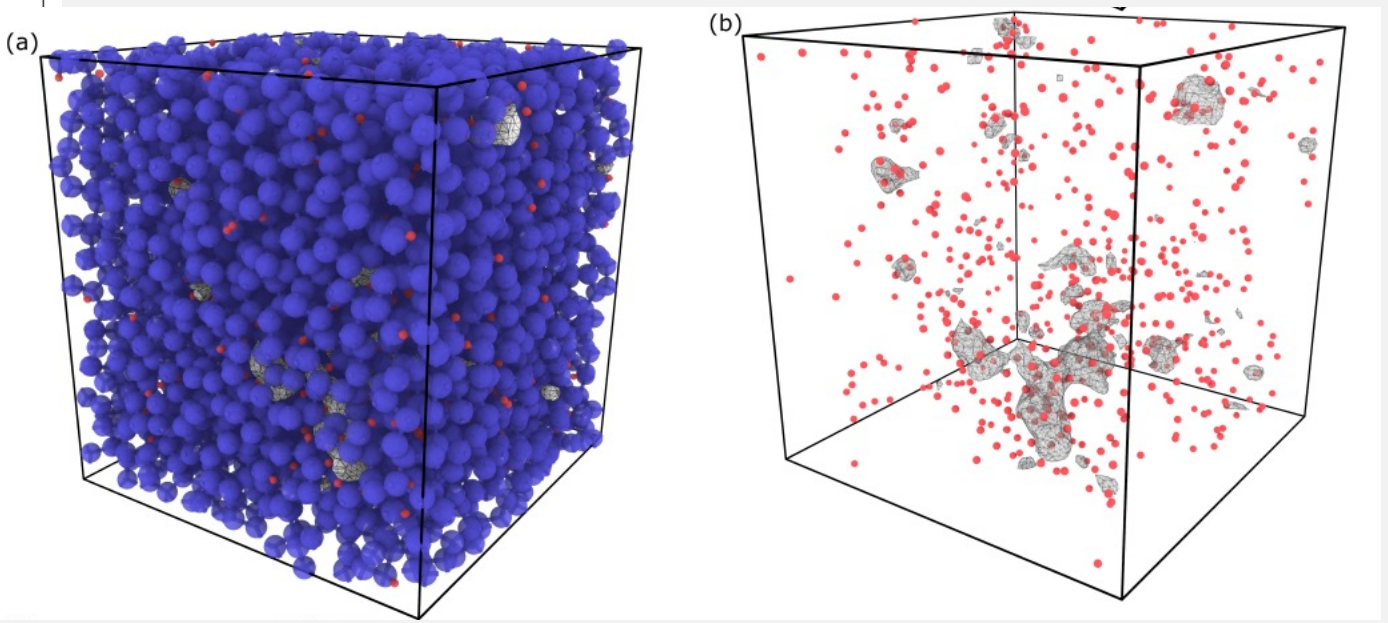
Energies: reproduces DFT within 4 meV/atom

Machine-Learning based Si-H GAP



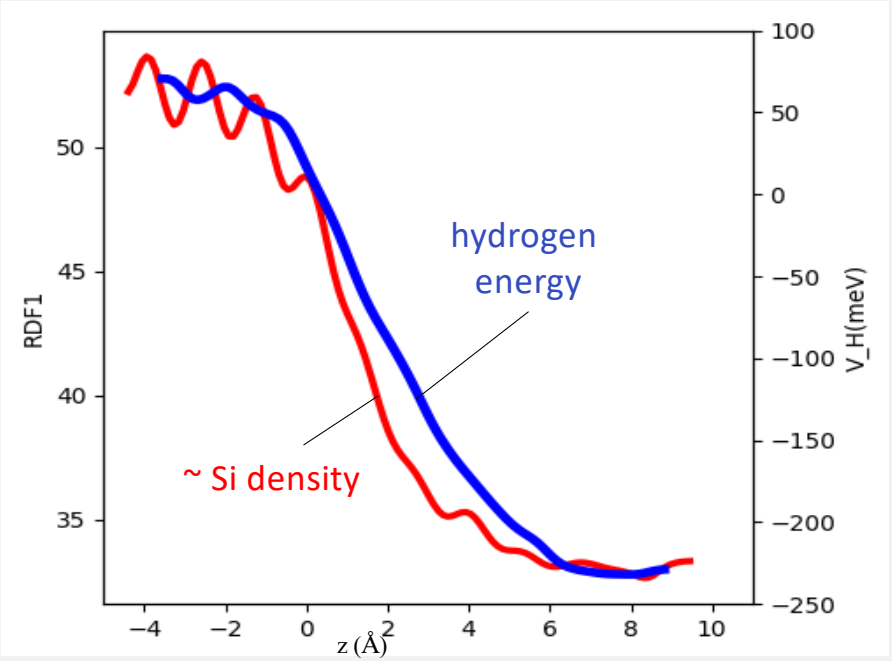
Run time scales with N, not with N^3, like DFT
Thus, here is a simulation ~ 4,600 Si and H atoms, completely unreachable for DFT

Our Si-H GAP reproduces DFT correlator qualitatively better than Tersoff



IV. Repeat SolDeg for c-Si/a-Si:H with Si-H GAP

1. Created 60 c-Si/a-Si:H stacks.
2. Inserted H to ~500 positions into each of the 60 stacks and measured its energy (blue). This represents 30,000 calculations.
3. Gradient of hydrogen energy creates a force that drives H atoms away from SHJ interface!
4. What created H energy gradient? We measured Si density (red).
5. The Si density gradient correlates with, and likely causes, the hydrogen energy gradient

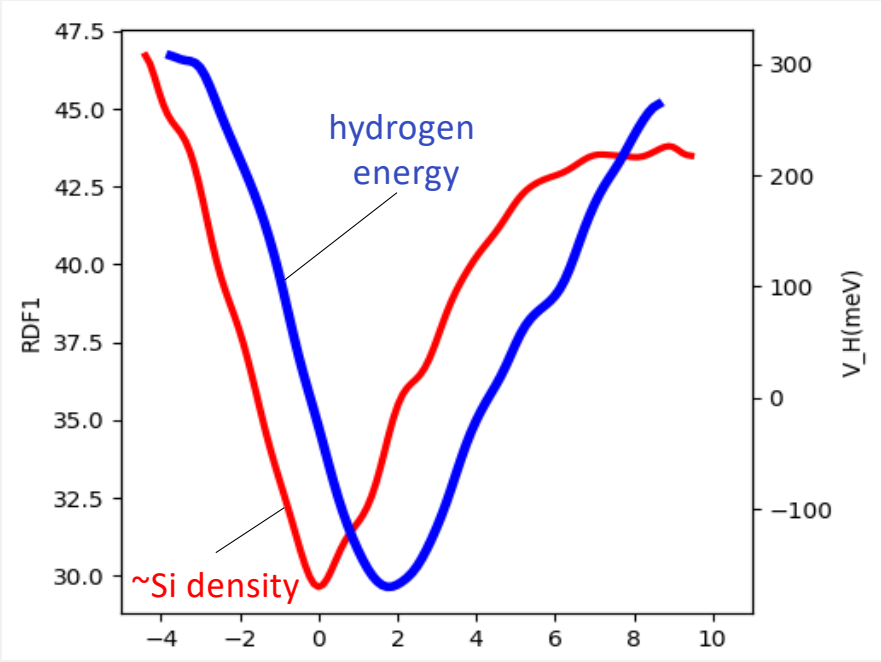


The Si density gradient across interface creates a force that drives H away from interface, degrading the SHJ cell passivation

How to stop H degradation? NoDeg cells

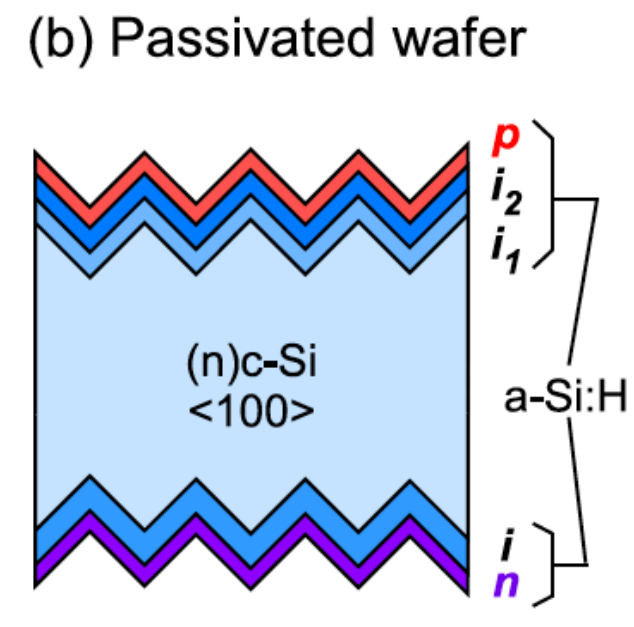
1. The H energy gradient was created by the Si density gradient.
2. Therefore, we expect that if we create a minimum for the Si density at the interface, the H energy gradient can be changed into an H energy valley, stabilizing the passivation.
3. We created 60 new c-Si/a-Si:H stacks where the Si density had a minimum at the interface. Again measured the H energy at 30,000 positions. Found that the H energy gradient has been eliminated!

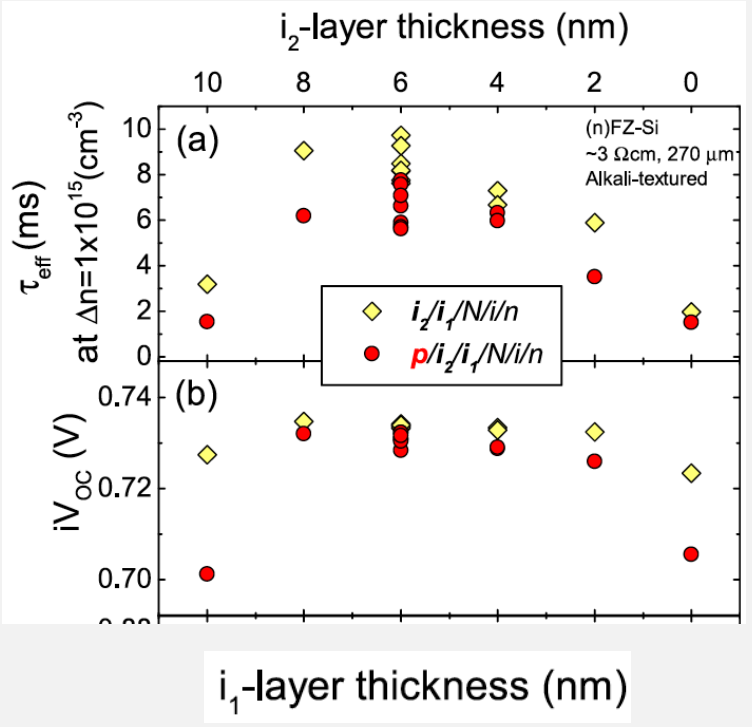
Creating a Si density minimum at the interface preserve the H passivation at the interface.



Possibly relevant experiments

Sai et al. J. Appl. Phys.
124, 103102 (2018)





"Surface passivation at the a-Si:H/c-Si interface is significantly improved by using an intrinsic a-Si:H bilayer, i.e., a stack of an interfacial layer with a large R* and an additional dense layer"

SUMMARY

1. Developed SolDeg platform to analyze defect dynamics that bridges 24 orders of magnitude in time

2. In Si-only SHJs found that defect generation follows a stretched exponential

3. Discovered scaling law of accelerated testing: $t(norm) \sim t(acc)^{(T(acc)/T(norm)}$

4. Reported experimental analysis of SHJ degradation

5. Developed Machine-Learning-based Si-H GAP potential for most accurate Molecular Dynamics simulation of c-Si/a-Si:H SHJs with many thousands of of atoms

6. Performed SolDeg simulation of c-Si/a-Si:H SHJs

7. Determined H energy that drives H away from SHJ interface, thereby degrading the cell passivation

8. Proposed modified SHJ architecture where the H-drift degradation channel is blocked

- ACS Applied Materials & Interfaces 2021, 32424
- arxiv 2106.02946 - submitted to Phys. Rev. Mat.
- To be submitted this week