A novel method to predict densities of amorphous materials from first-principles calculations

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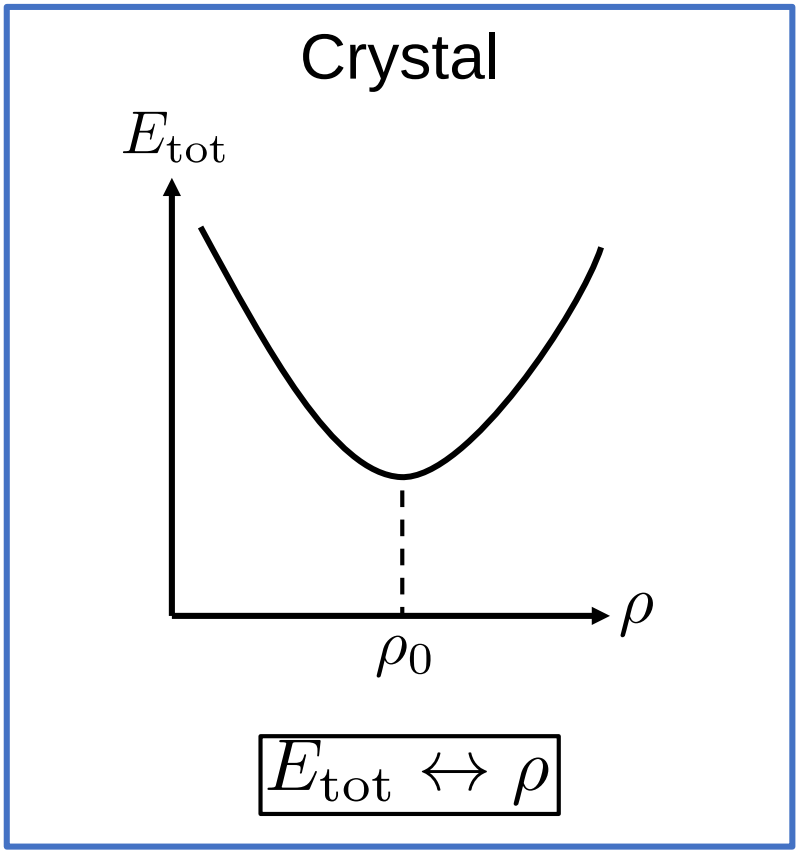
(arxiv:1704.06107)

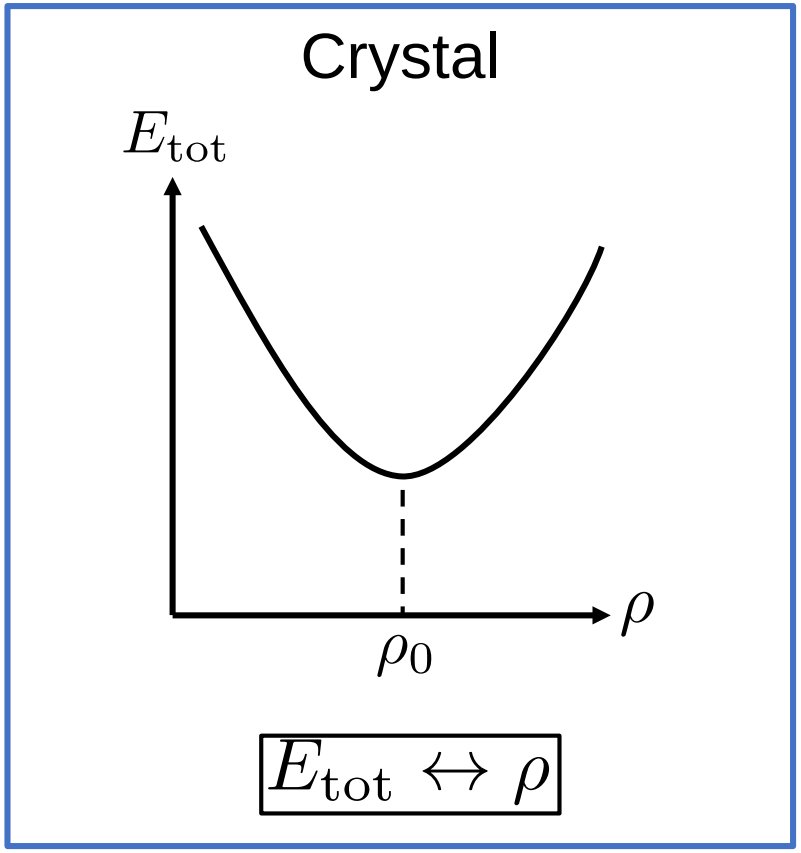
Determining the densities of materials

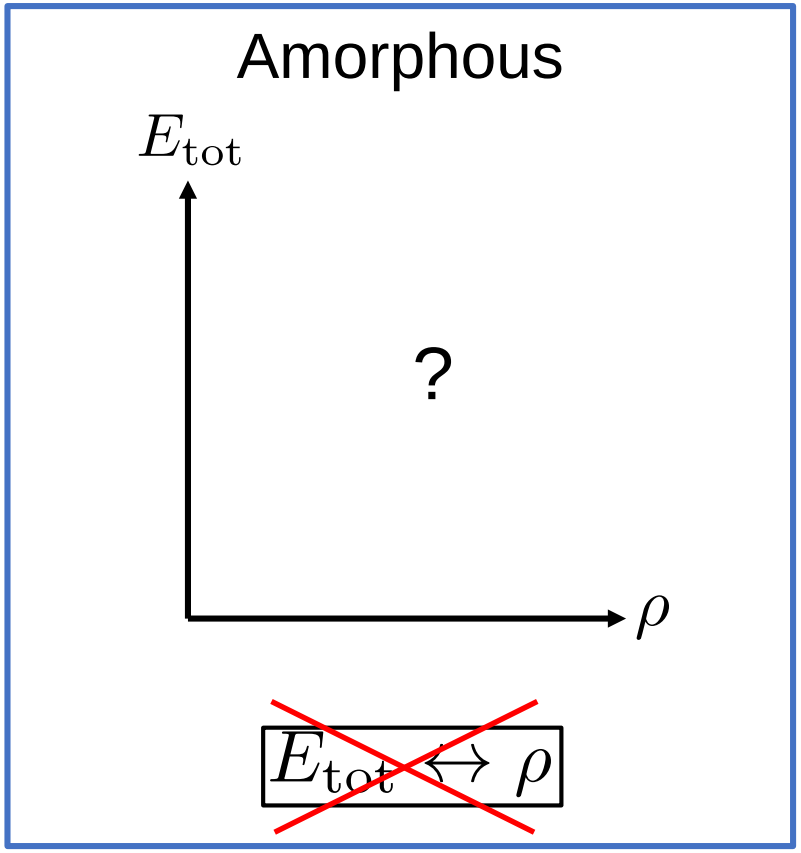
The densities (ρ) of crystalline materials are determined by calculating the total energy (E_{tot}), since E_{tot} corresponds to ρ



Determining the densities of materials

Not for amorphous materials, because the structures are disordered



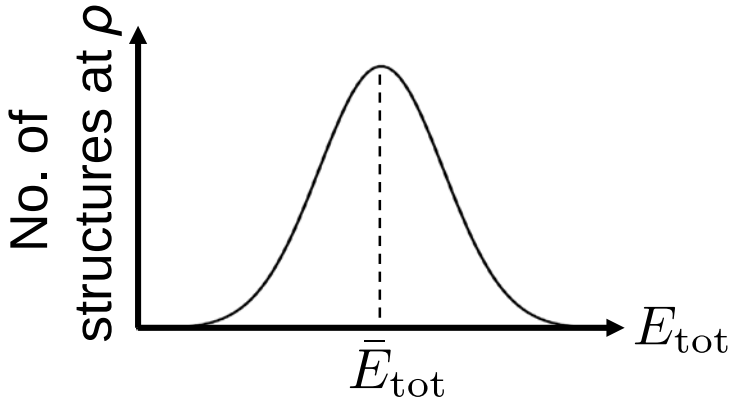


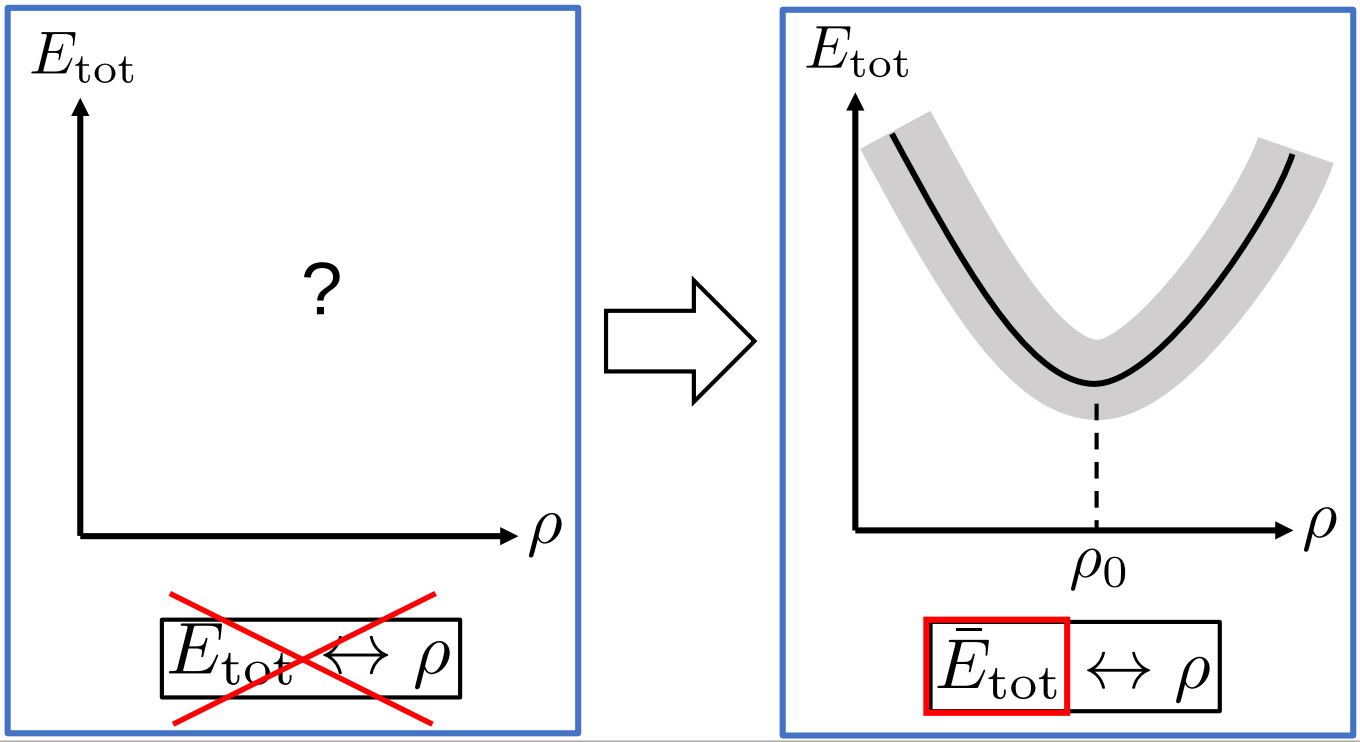
Any solution for amorphous materials? → our work

An assumption

The canonical distribution of the amorphous samples form a Gaussian distribution

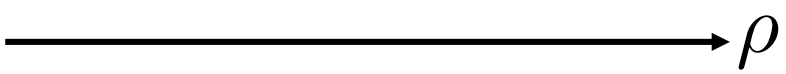
→Then, the averaged energy corresponds to the density





Procedure to determine the density

1. Generate multiple samples (1,…,M) at different densities (ρ_1,…,ρ_K)
2. Obtain their total energies from DFT
3. Average the total energies at each density
4. Find the density that minimizes the average $\rightarrow \rho_0$



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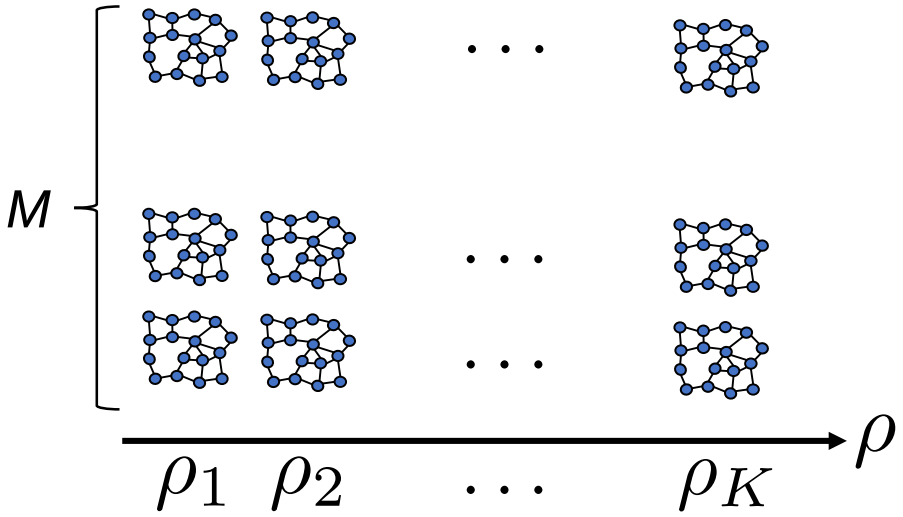

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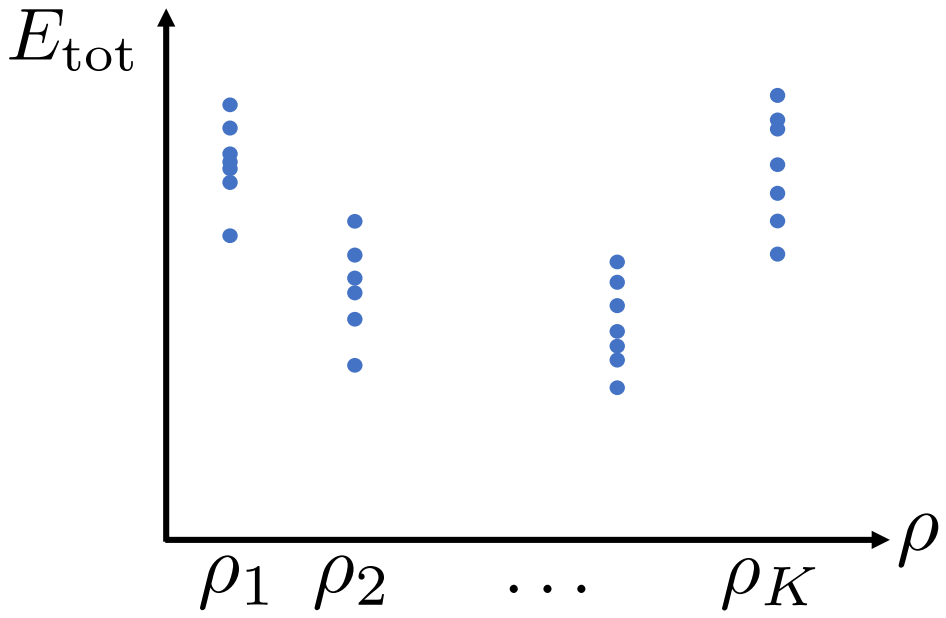

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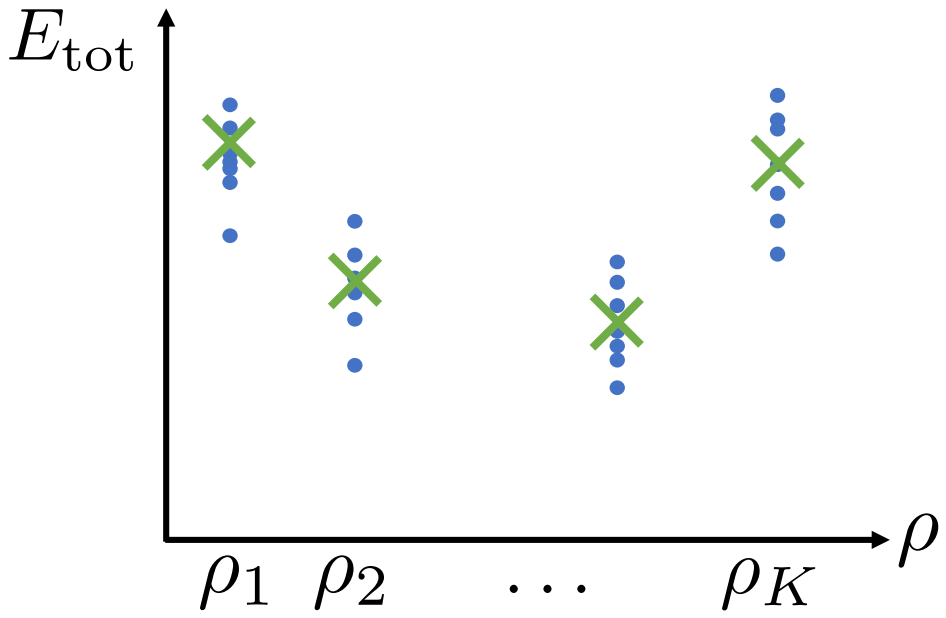

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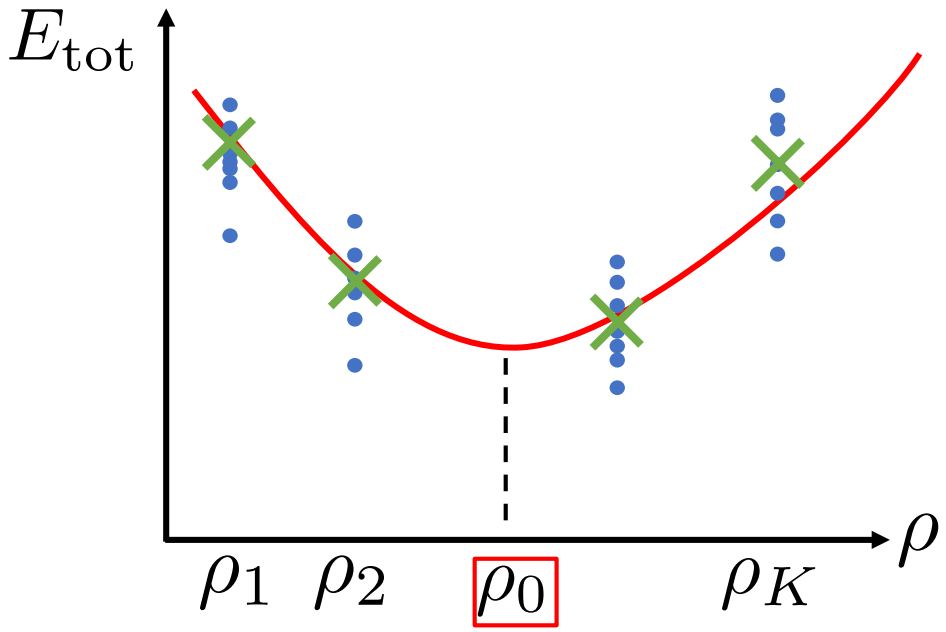
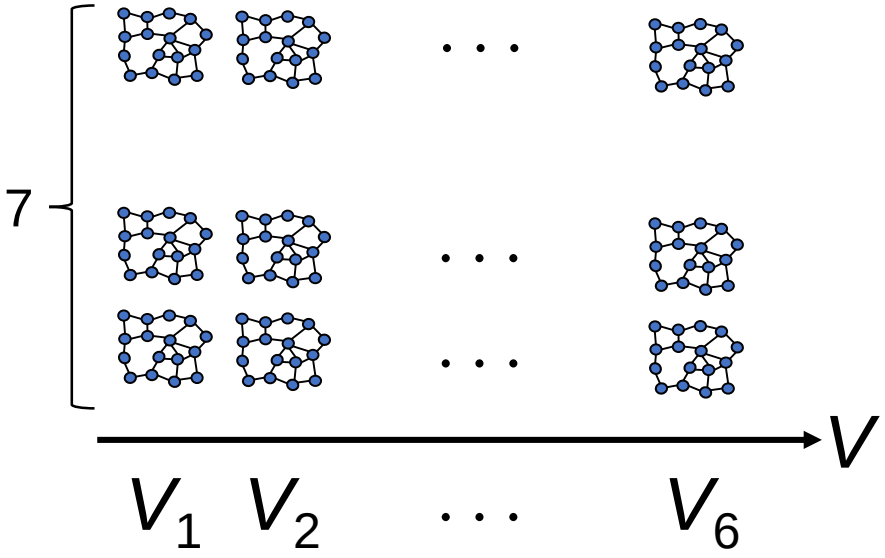


→ examined with amorphous Si (a-Si)

Generating a-Si samples

1. Generate 7 samples at 6 different volumes *V*

- V/V_C = 0.86 - 1.16 (V_C: volume of crystal)
- System: 54 Si atoms/cell with periodic boundaries

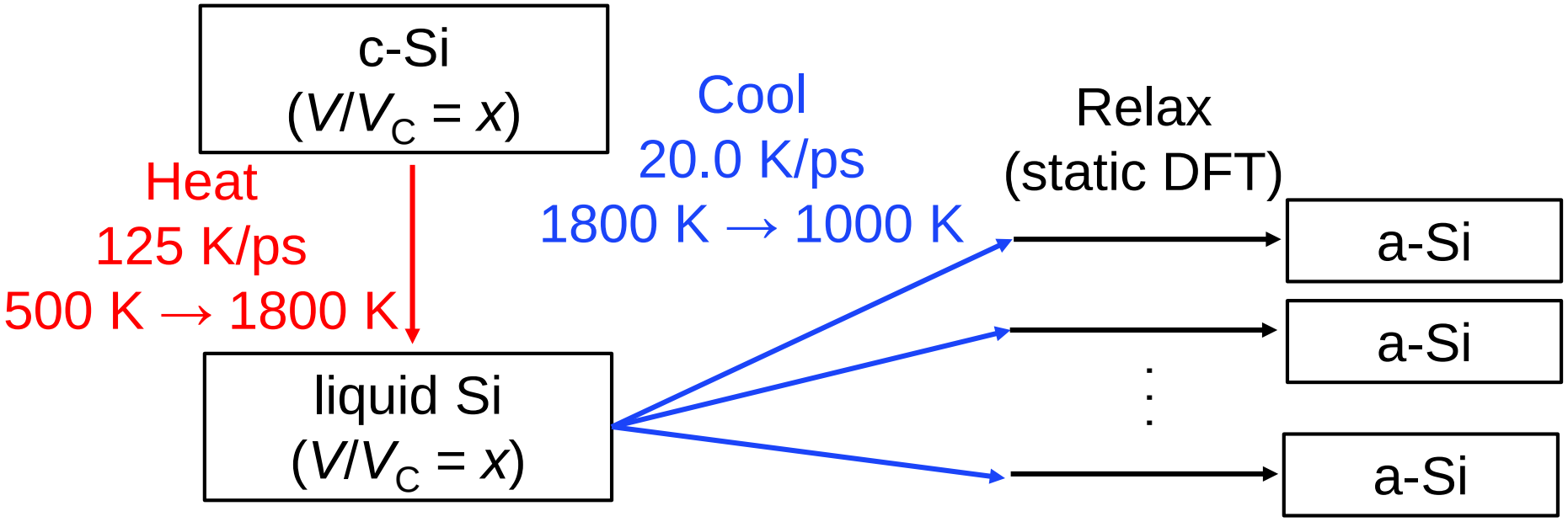


Generating a-Si samples

1. Generate 7 samples at 6 different volumes *V*

- V/V_C = 0.86 - 1.16 (V_C: volume of crystal)
- System: 54 Si atoms/cell with periodic boundaries
- Melt-quench method by Car-Parrinello molecular dynamics (CPMD)

(functional=PBE, cutoff=40 Ry, BZ sampling=Γ point)

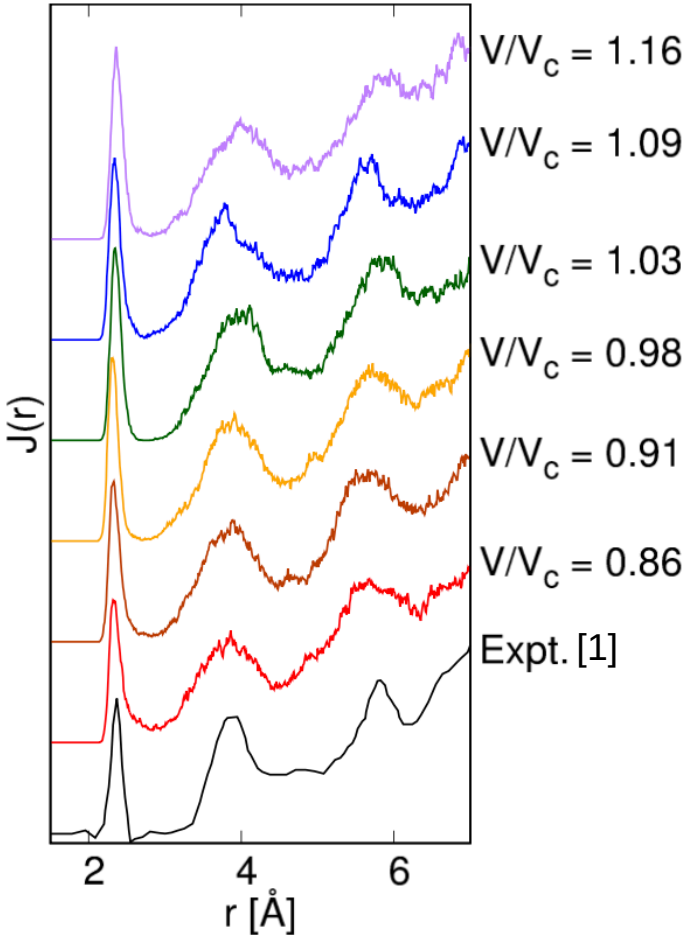


Radial distributions of samples

Good agreement with experiments:

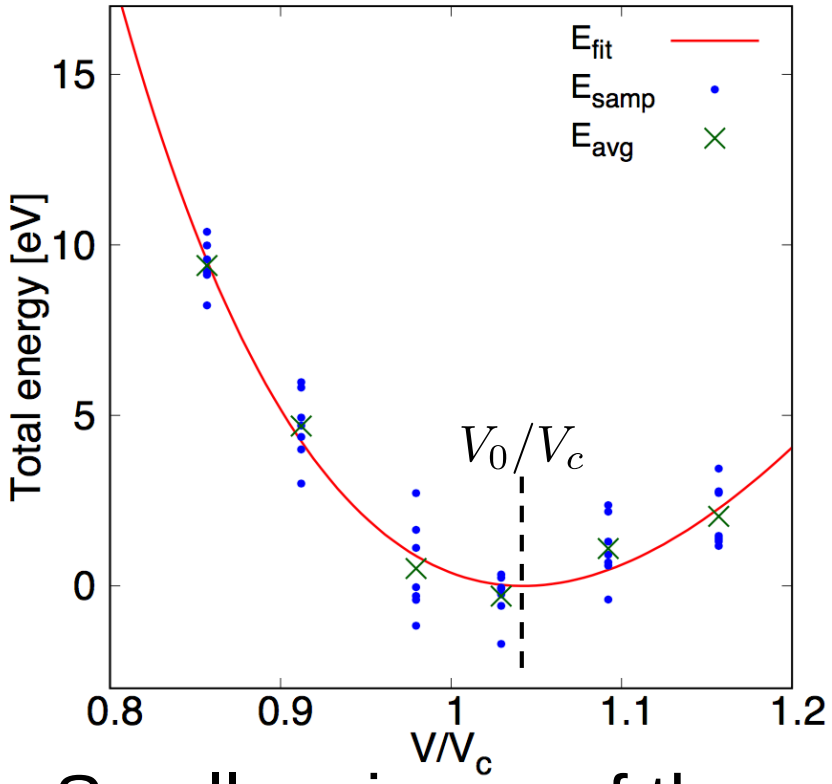
- First peak at ~2.3 Å
- Broader peaks subsequently

→Obtained samples are certainly valid a-Si



[1] PRB 60, 19 (1999)

Total energies



	Calc.	Expt.
V_0/V_C	1.042	1.017 - 1.019 [1]
B_0 (a-Si)	61.27	36 – 95 [2]
B_0 (c-Si)	89.59	97.6 [3]

- Small variance of the samples' energies (< 1.6 eV)
- Both V_0/V_C and B_0 (bulk modulus) agrees with expt.

→Our method works for a-Si

[1] Appl. Phys. Lett. **64**, 437 (1994)

[2] "*Thin film materials*", Cambridge University Press, (2003); PRB **58**, 8941 (1998)

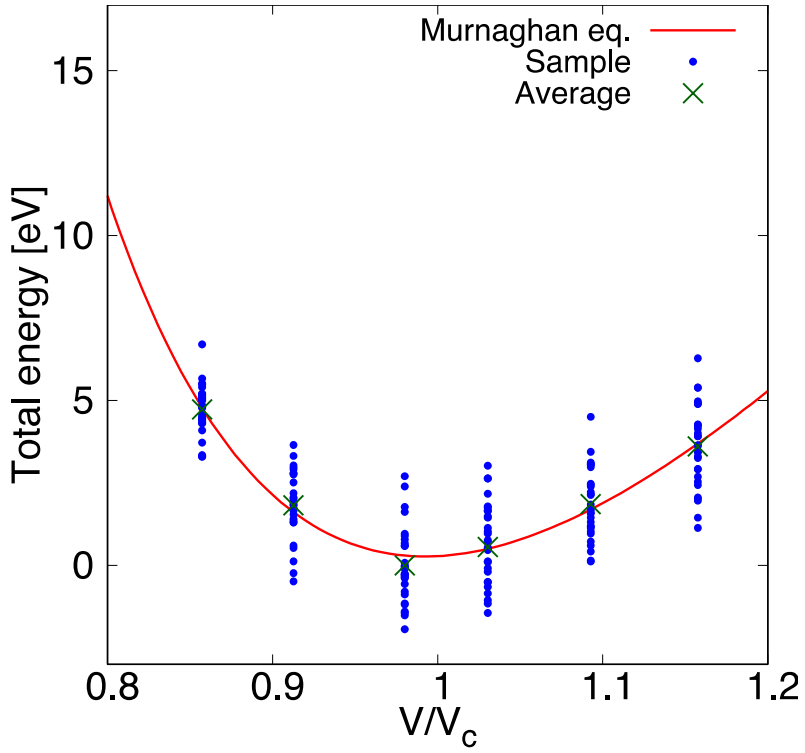
[3] J. Microelectromech. Syst. **19**, 2 (2010)

Results from MD simulations

What if we generate samples by MD instead of CPMD?

Conditions:
- Tersoff potential
- 30 samples at each volume

Result:
- V_0/V_C = 0.992
- B_0 = 49 GPa

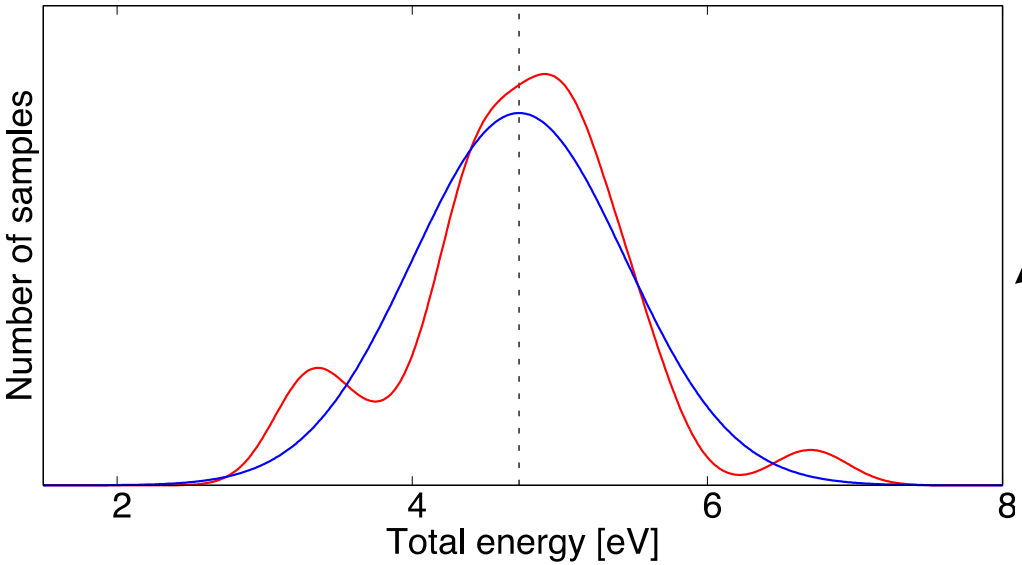


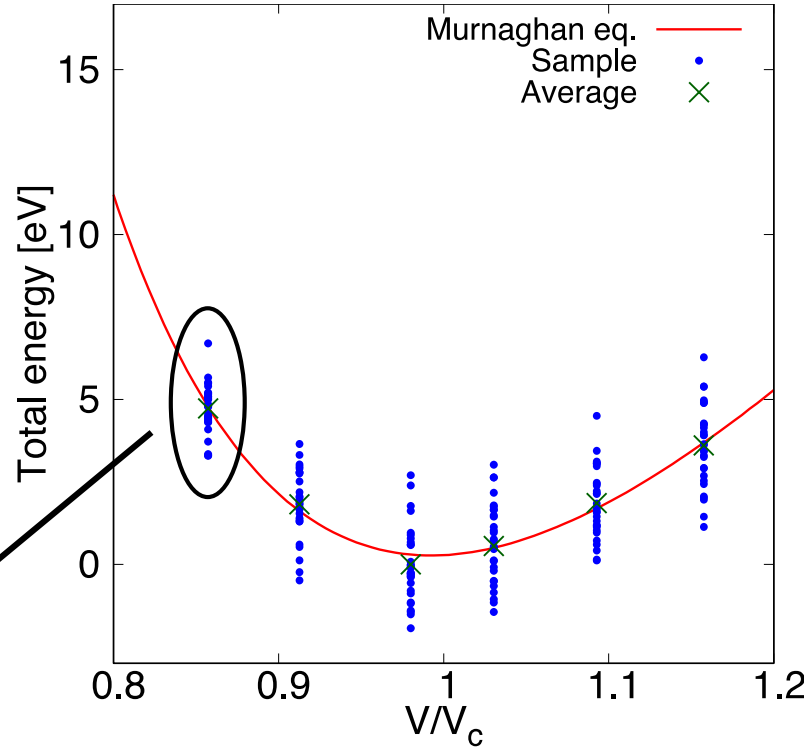
→It works, but the total energy is higher (3 eV/cell)
→CPMD-generated structures are energetically preferable

Results from MD simulations

A finding: the sample distribution is close to Gaussian distribution

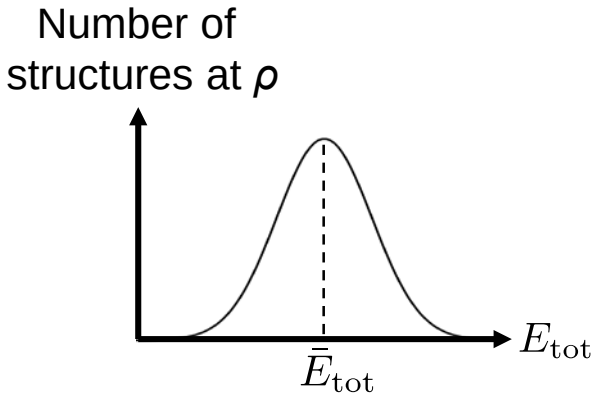
→Supports our assumption

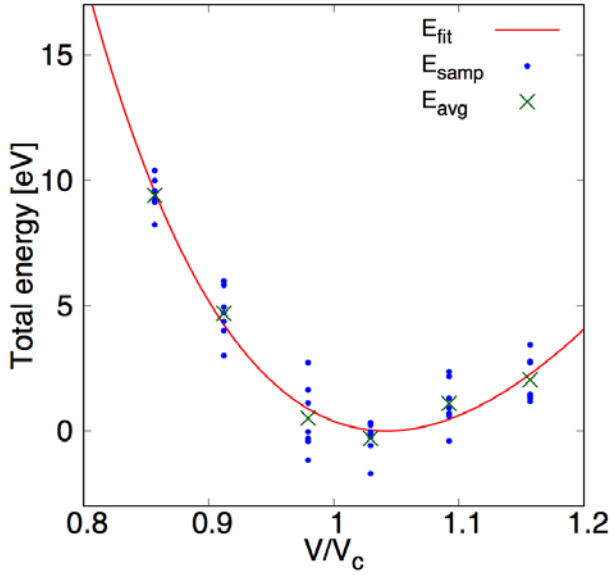




Summary

- A method to computationally determine the density of amorphous materials is proposed
- For a-Si, both the density (volume) and the bulk modulus have been well reproduced
- Results from MD simulations imply the validity of our underlying assumption





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