Formation Energies of Silicon Interstitials: DMC vs. DFT

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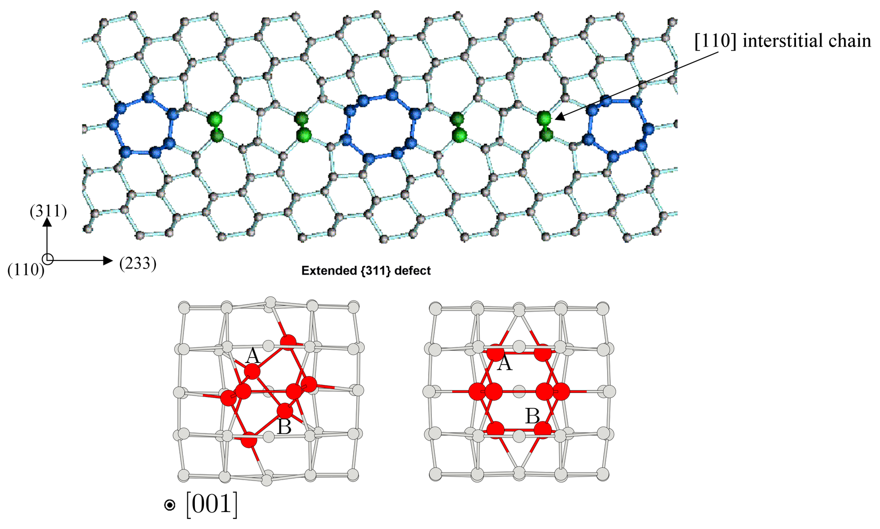
Interstitial Silicon Defects

From point defects to extended {311} defect

- Ion implanted Si interstitials nucleate {311} defects limiting device fabrication and performance
- Defects identified in TEM/EELS experiment [1]
- Defects from TB-MD and DFT relaxations
- Several dozen new stable interstitial clusters [2]



Quantum Monte Carlo Method

Trial Wave Function and Jastrow Factor

$$\Psi_T = \underbrace{\mathcal{D}^{\uparrow}\mathcal{D}^{\downarrow}}_{\text{Slater determinant}} \times \underbrace{\mathcal{J}(r_i, r_j, r_{ij})}_{\text{Jastrow factor}}$$

$$\mathcal{J}(r_i, r_j, r_{ij}) = \underbrace{\prod_{\alpha i} \exp(A_{\alpha i})}_{\text{electron-ion correlation}} \times \underbrace{\prod_{ij} \exp(B_{ij})}_{\text{electron-electron correlation}}$$

Variance Minimization

- Optimize Jastrow by variance minimization with Levenberg-Marquardt
- More stable and requires less configurations than energy minimization

Diffusion Monte Carlo

- Stochastic method of solving many-body Schrödinger equation
- Projection of ground state

Density-Functional Calculations

- Use CPW2000 and VASP

Approximations

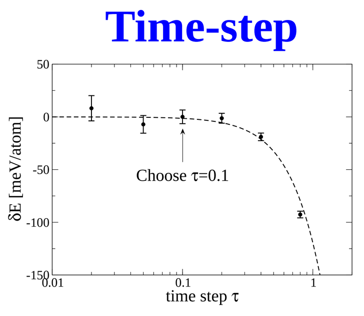
Controlled

- Statistical (increase MC steps)
- Finite-size (larger systems)
- Time-step (smaller time step)
- Population control (more walkers)
- Grid-size (decrease grid spacing)

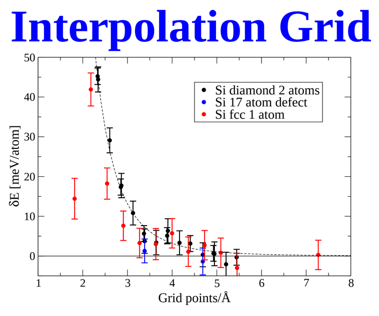
Uncontrolled

- Fixed node
- Pseudopotential
- Pseudopotential locality

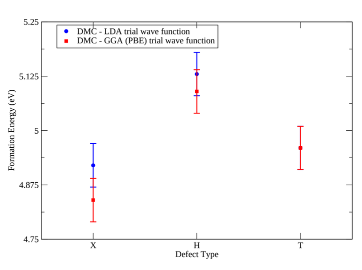
Time-step



Interpolation Grid

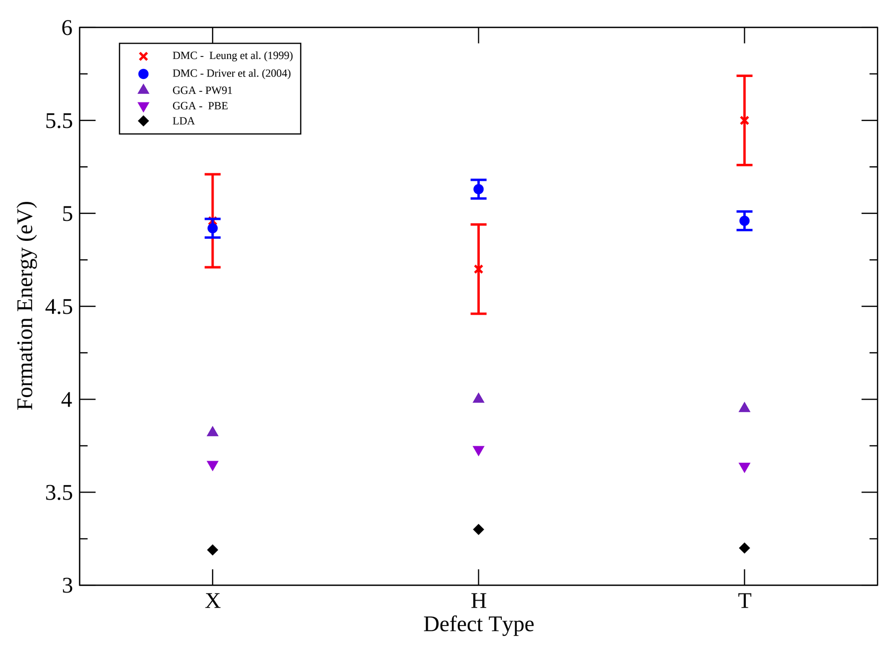


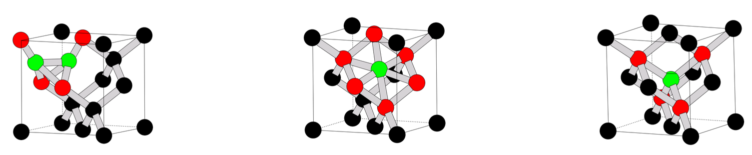
Fixed Node



Formation Energies of Interstitial Silicon Defects

Single interstitial in 16-atom cell

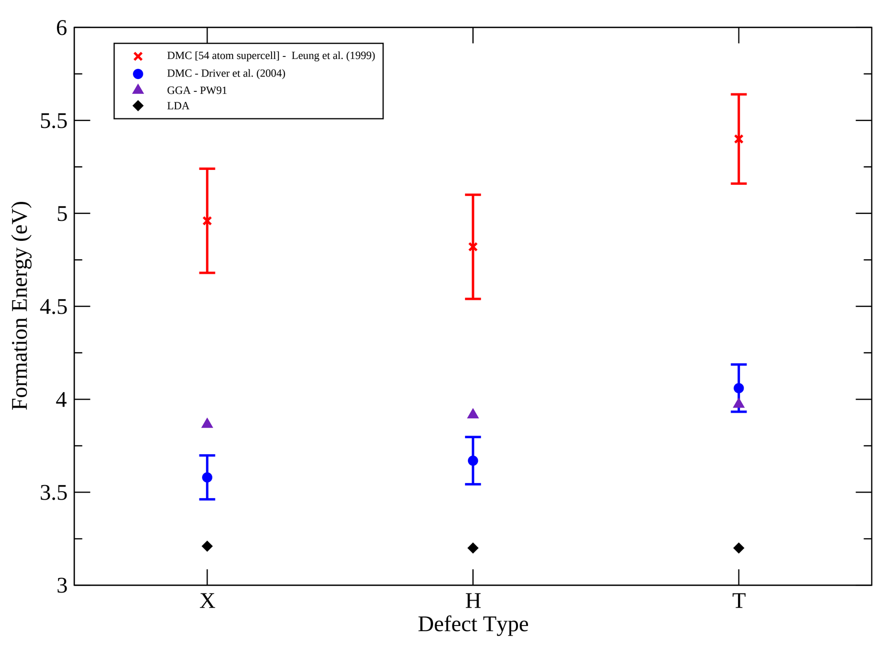


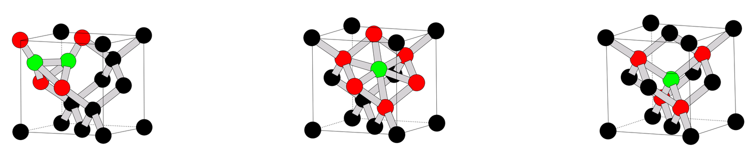


	X	H	T
DMC	**4.92(5)**	**5.13(5)**	**4.96(5)**
DMC[3]	4.96(24)	4.70(24)	5.50(24)
GGA-PW91	3.82	4.00	3.94
GGA-PBE	3.65	3.73	3.64
LDA-CA	3.19	3.30	3.20

- 16-atom cell results are similar to earlier DMC by Leung et al.
- DFT underestimates defect energies:
  - LDA underestimates by about 1.5 eV
  - GGA (PW91 and PBE) underestimates by about 1 eV

Single interstitial in 64-atom cell

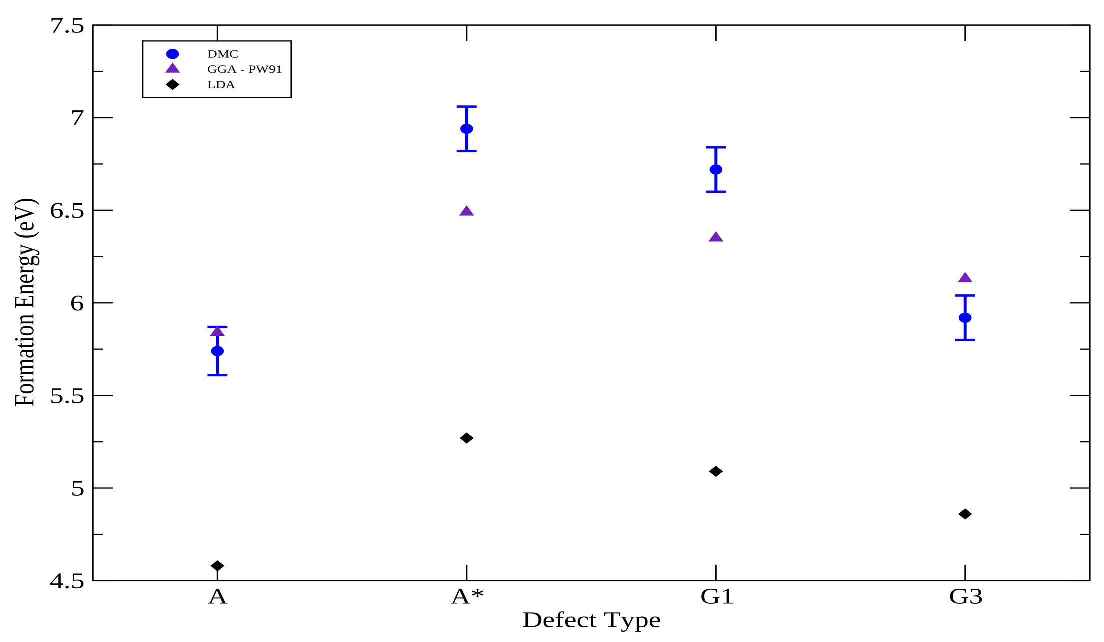




	X	H	T
DMC	**3.58(12)**	**3.67(13)**	**4.06(13)**
DMC[3]	4.96(28)	4.82(28)	5.40 (28)
GGA-PW91	3.87	3.92	3.97
LDA-CA	3.21	3.20	3.33

- In 64-atom cells we find the DMC energies to be close to GGA and LDA values. This contrasts with Leung et al. observations in 54- atom cells.
- These results are unexpected. Further study and analysis is needed to fully understand their importance.
- Energy versus volume curves of bulk Si suggest dependence of energy on pseudopotential.

Di-interstitial in 64-atom cell



	A	A-star	G1	G3
DMC	**5.73(12)**	**6.94(12)**	**6.72(12)**	**5.92(12)**
GGA-PW91	5.84	6.49	6.35	6.13
LDA-CA	4.58	5.27	5.09	4.86

- Di-interstitial results appear to be consistent with the single intersitial 64-atom cell results.
- GGA values lie within about 0.5 eV of DMC values
- LDA underestimates defect energies by about 1 eV.

Bibliography

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Acknowledgements

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