Structural simplicity as a restraint on the structure of amorphous silicon

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1 Full list of configurations

All configurations are 512 atoms, in a 21.7 Å cubic box. All RMC refined configurations were refined for 128 million attempted moves.

Random A random configuration, with no other constraints.

Hard sphere A random configuration, generated with the constraint that no atom be placed within 2.2 Å of another.

RMC A random configuration refined against PDF data taken from a 4096 atom WWW configuration using RMC, with no other constraints.

RMC relax The RMC configuration optimised with DFT using the PBE functional until convergence.

INVERT A random configuration refined against PDF data taken from a 4096 atom WWW configuration using RMC, with the INVERT PDF variance restraint applied[S1].

INVERT relax The INVERT configuration optimised with DFT using the PBE functional until convergence.

SPH A random configuration refined against PDF data taken from a 4096 atom WWW configuration using RMC, with the INVERT PDF variance restraint, the spherical harmonic bond-orientational order parameter variance constraint, and the spherical harmonics local symmetry restraint applied[S1, S2].

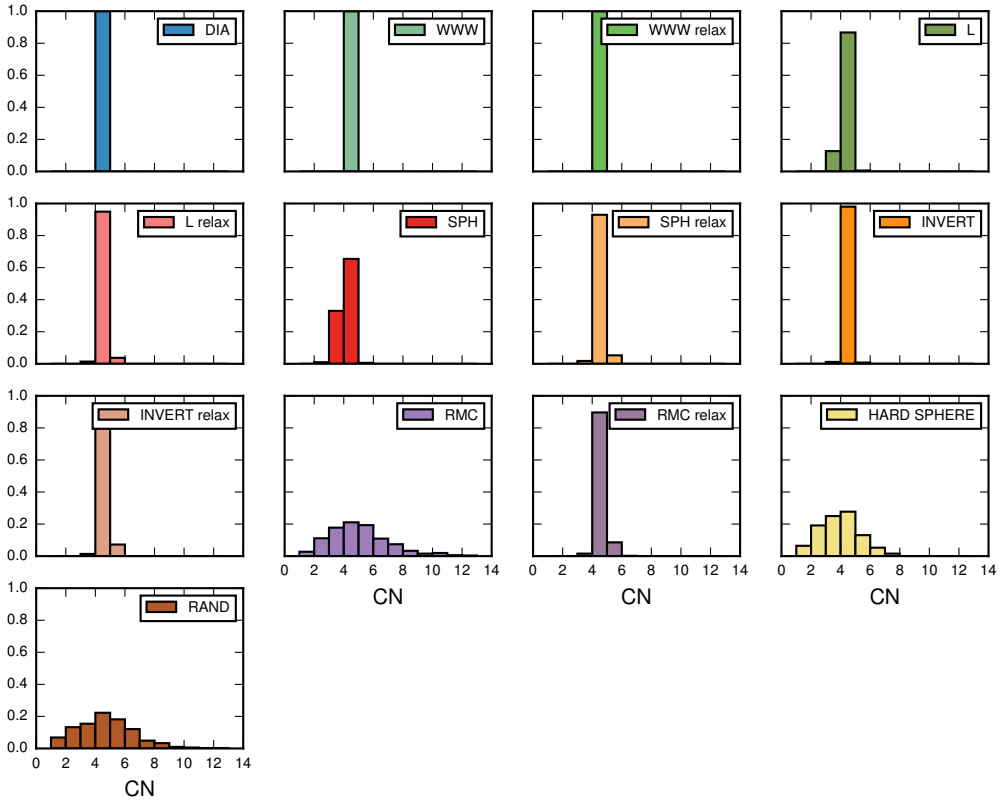
SPH relax The SPH configuration optimised with DFT using the PBE functional until convergence.

L A random configuration refined against PDF data taken from a 4096 atom WWW configuration using RMC, with the SOAP self-similarity restraint L also applied[S3].

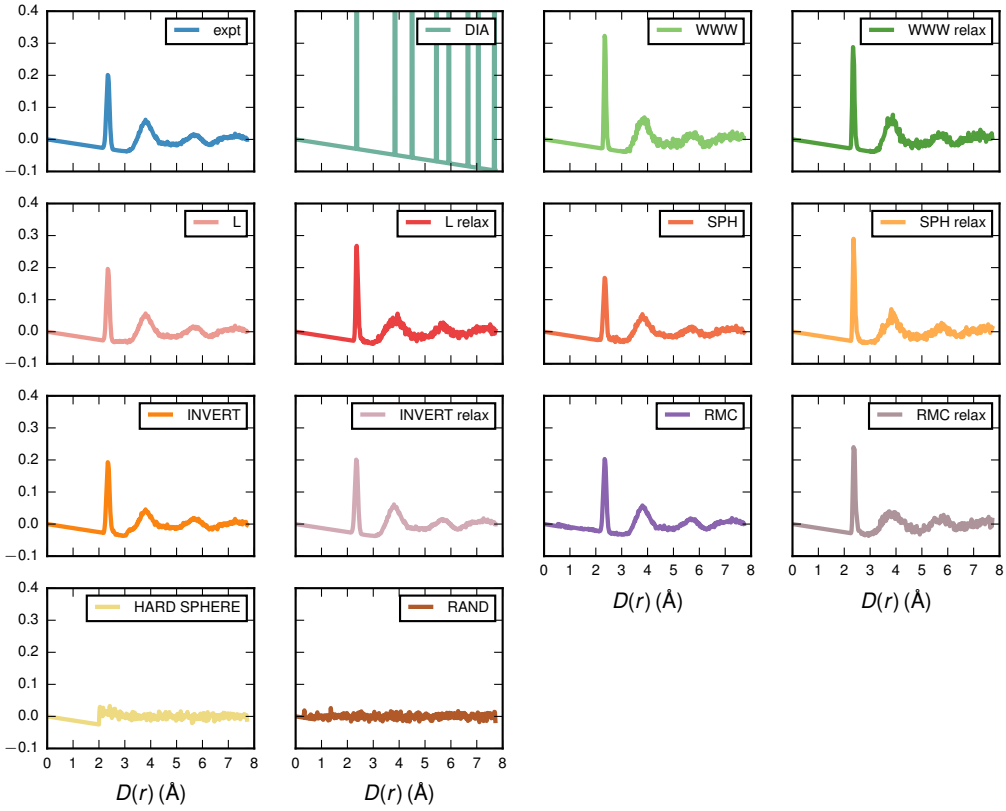
L **relax** L optimised with DFT using the PBE functional until convergence.

WWW A configuration generated using the WWW algorithm[S4].

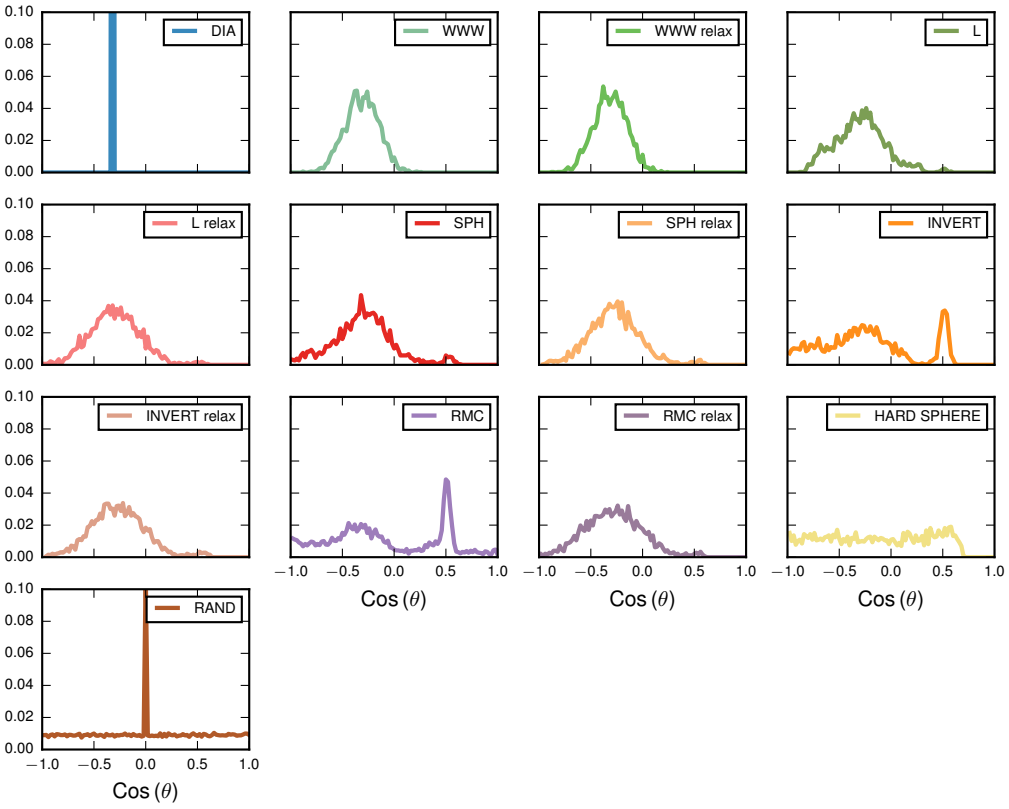
WWW relax The WWW configuration optimised with DFT using the PBE functional until convergence.



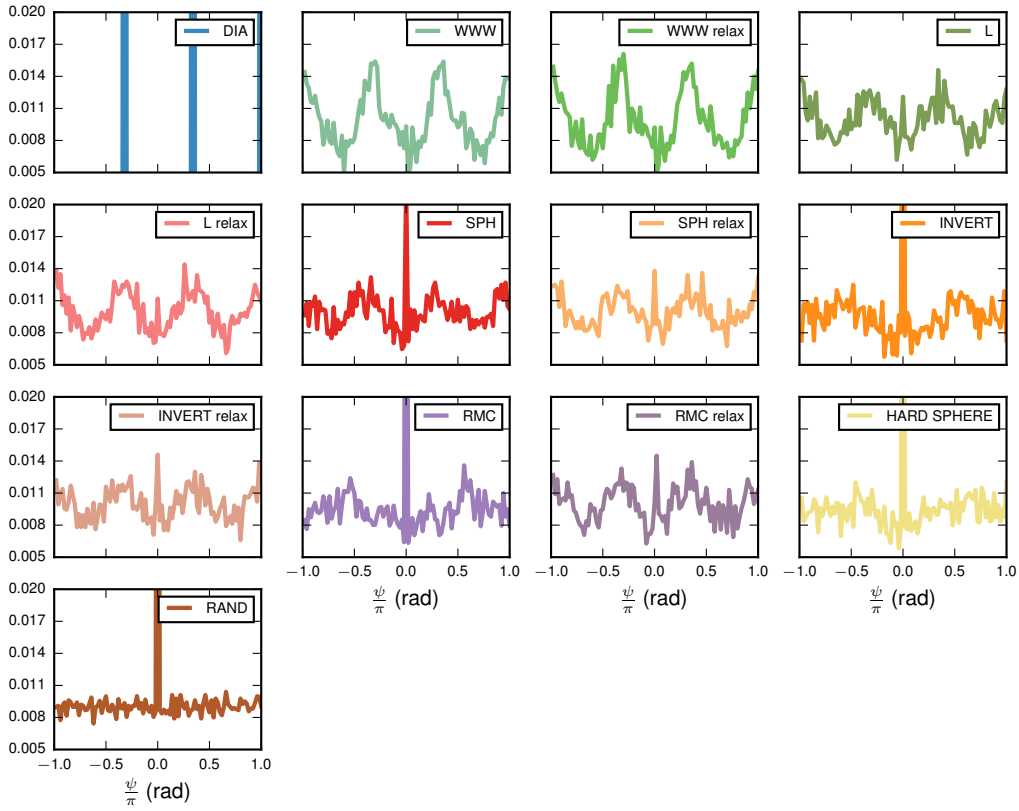
ESI Fig. 1: Histograms of the coordination number distributions for all configurations



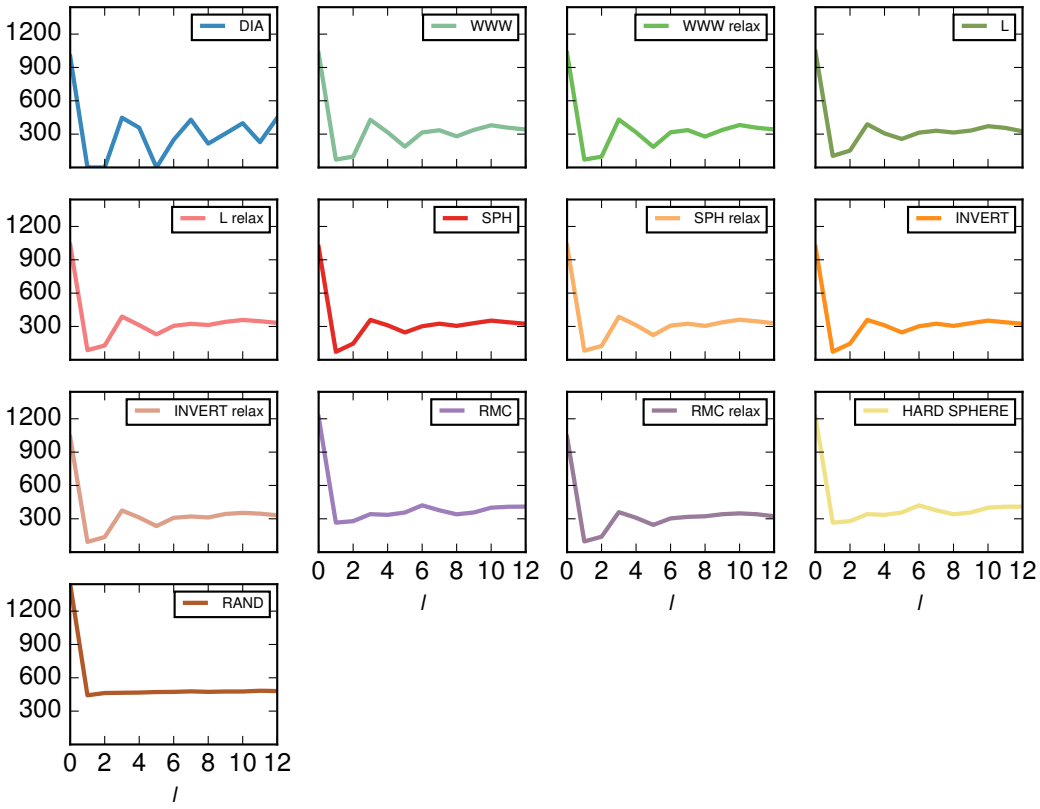
ESI Fig. 2: Pair distribution functions for all configurations



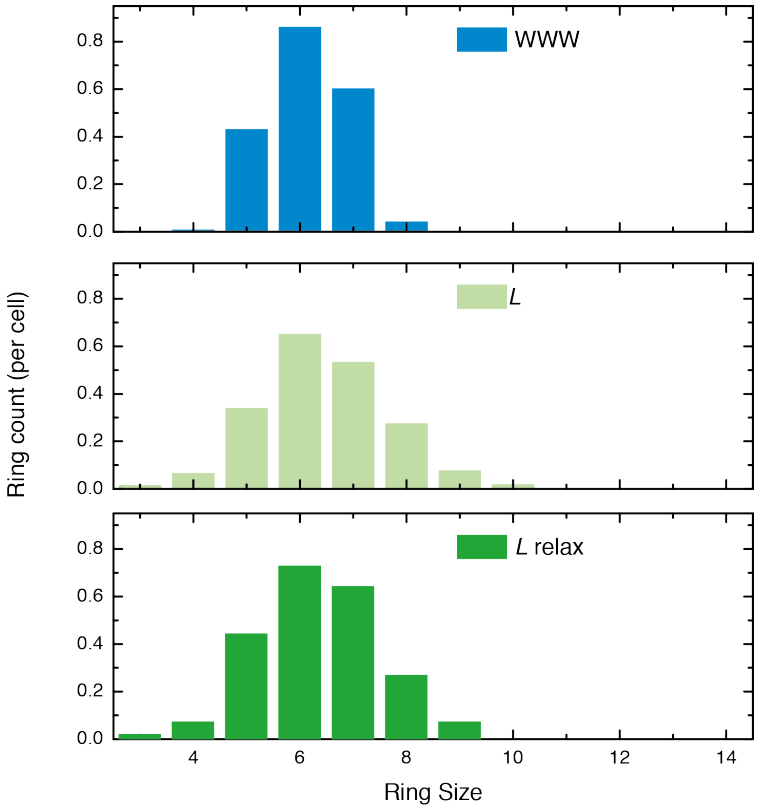
ESI Fig. 3: Histograms of the bond angle distributions for all configurations. The large spike at $\cos(\theta) = 0$ for the random configuration results from near overlapping atoms.



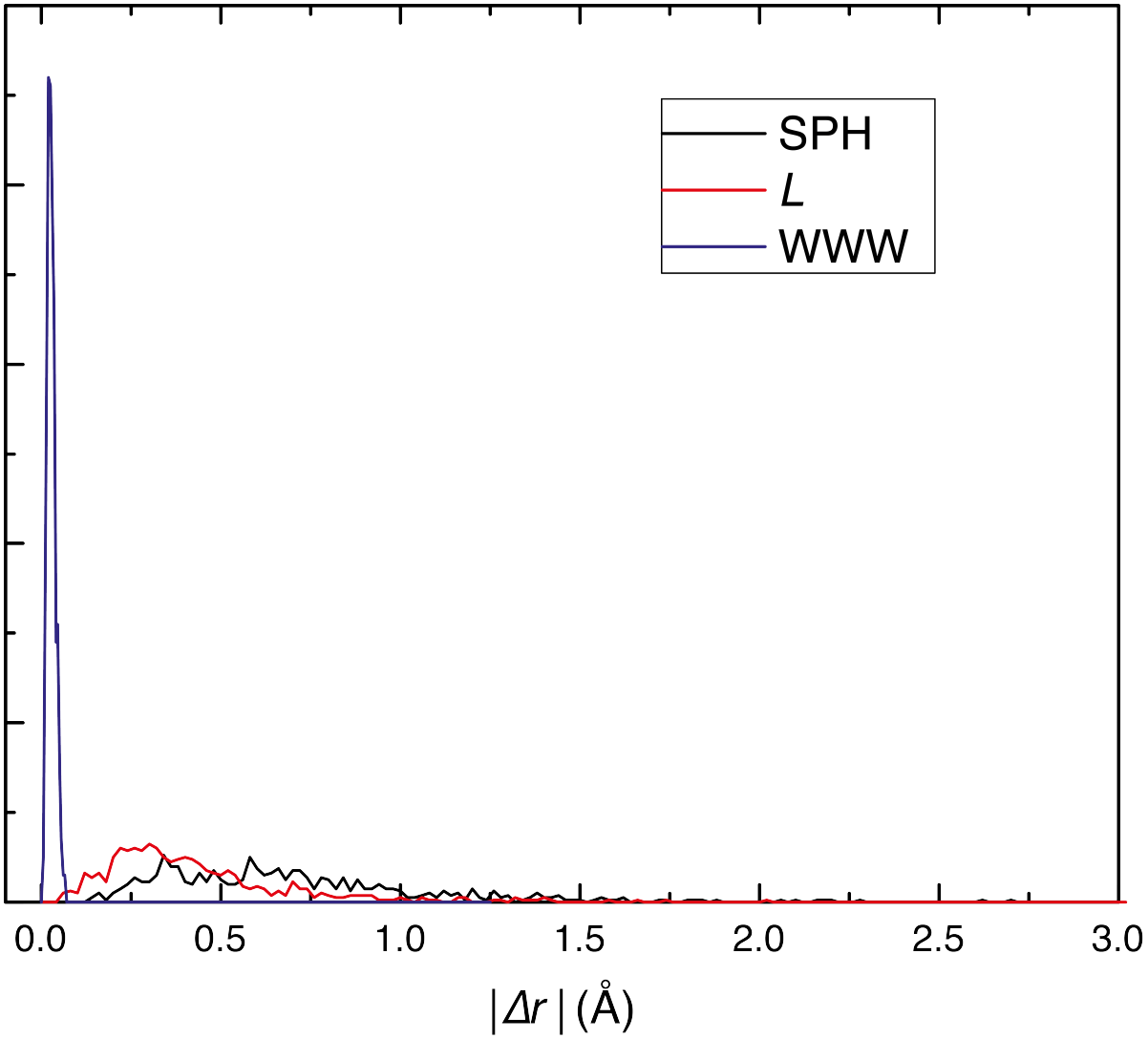
ESI Fig. 4: Histograms of the dihedral angle distributions for all configurations. The spike at $\psi = 0$ results from three-membered rings, which are necessarily completely planar.



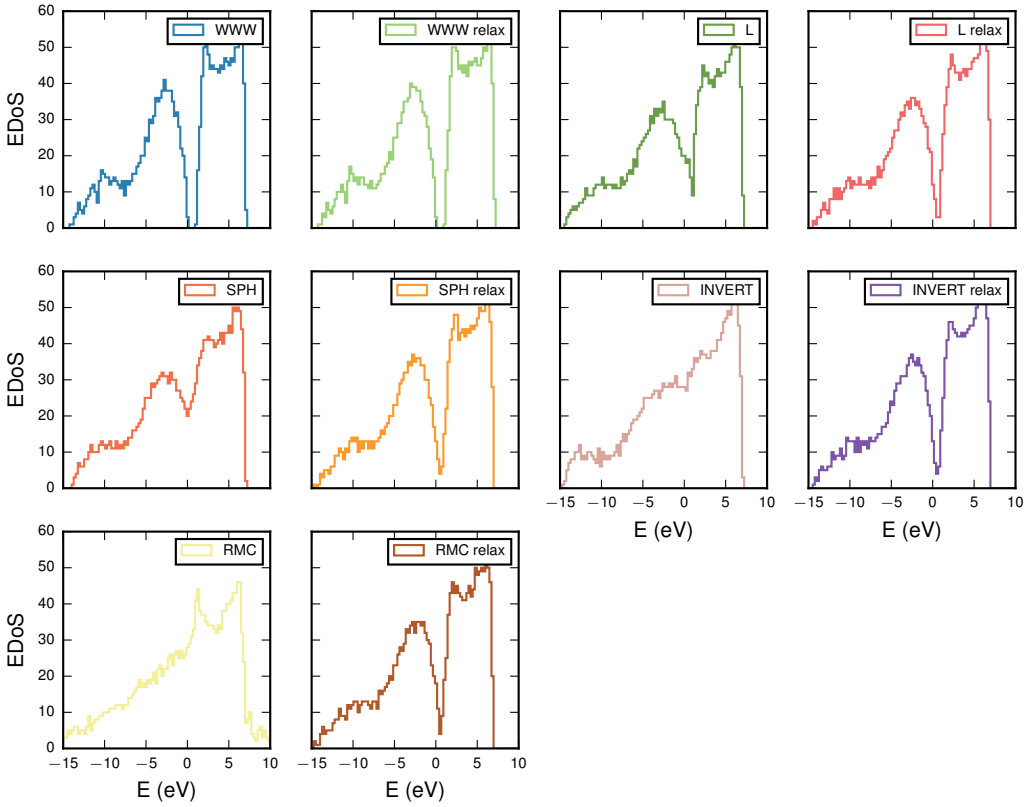
ESI Fig. 5: Calculated average Q_l coefficients for all configurations.



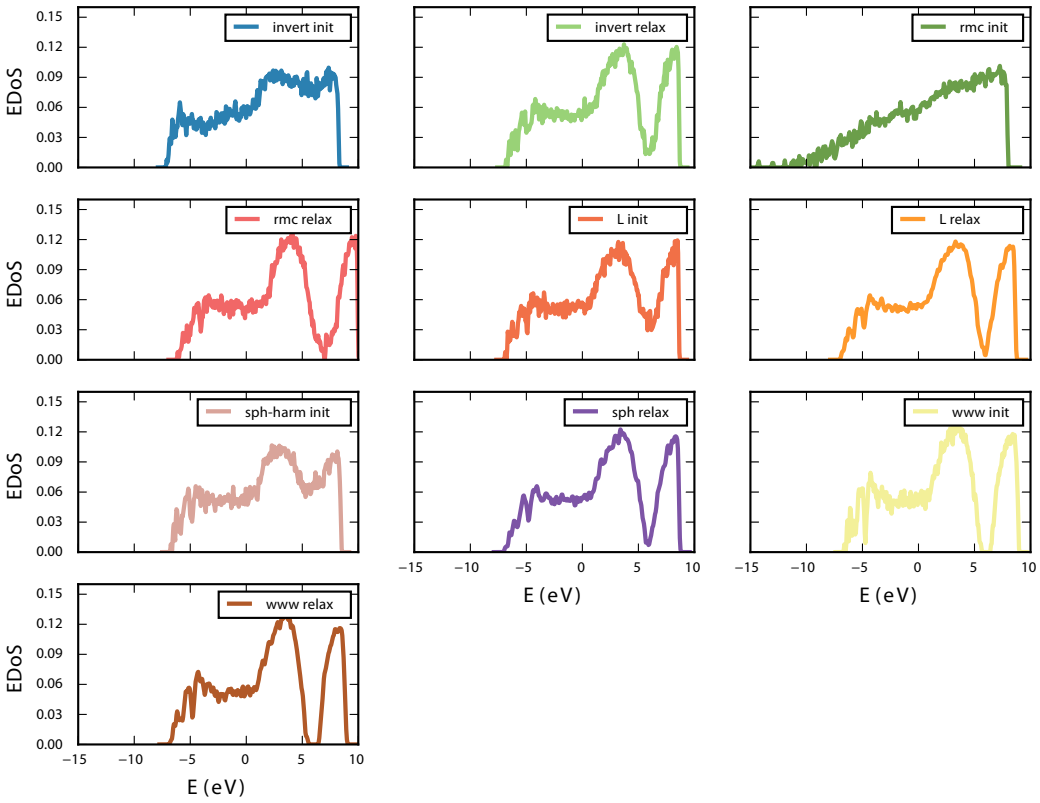
ESI Fig. 6: Ring statistics (using the King's shortest path convention and normalised by number of rings per cell) for the WWW configuration and PDF+L refined configuration before and after relaxation. Calculated using ISAACS[S5].



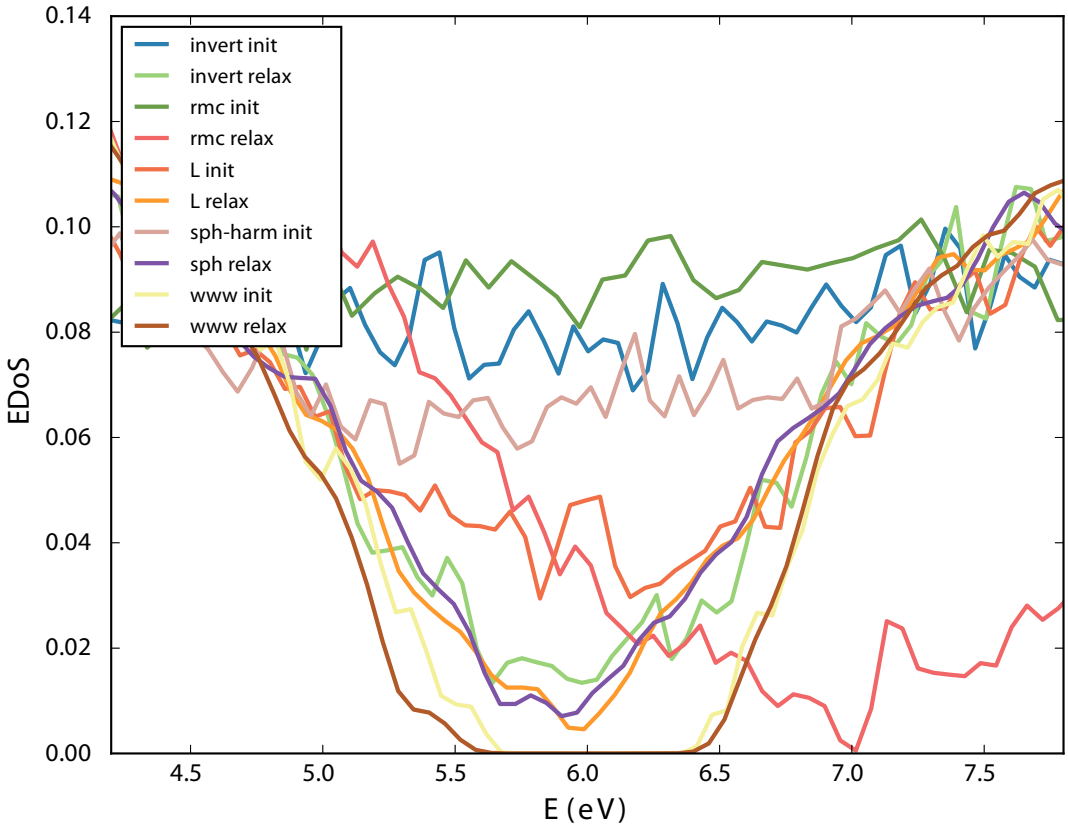
ESI Fig. 7: Histogram of the root square displacement during optimisation for the WWW, SOAP and SPH configurations.



ESI Fig. 8: Electronic density of states calculated using the GSP tight-binding Hamiltonian[S6].



ESI Fig. 9: Electronic density of states calculated using DFT and the MBJ functional.



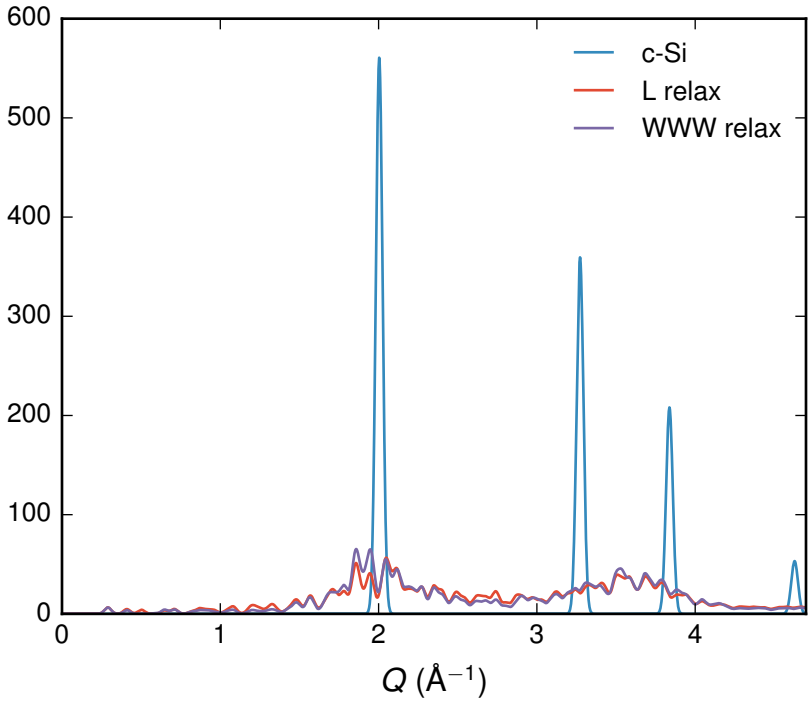
ESI Fig. 10: Electronic density of states calculated using DFT and the MBJ functional, around the gap.

ΔE (eV/atom)	WWW	L	SPH	INVERT	RMC
PBE unoptimised	0.15818068	0.401097615	0.515364201	0.698205189	6.361609732
PBE optimised	0.157081969	0.271789	0.260181578	0.277312328	0.299999896

ESI Table 1: Calculated DFT energies (using the PBE functional) for the optimised and unoptimised configurations of Si, relative to a configuration of crystalline Si.

$\log_{10}(\chi^2)$	WWW	WWW relax	L	L relax	SPH	SPH relax	INVERT	INVERT relax	RMC	RMC relax	hard sphere	random
χ^2_{PDF}	−4.58	−4.77	−6.08	−4.65	−6.18	−4.53	−5.35	−4.46	−5.95	−4.39	−3.89	−3.86
χ^2_{INVERT}	−1.52	−1.52	−1.31	−1.39	−1.28	−1.42	−1.90	−1.37	−0.42	−1.33	−0.95	−0.30
$\mathrm{Var}(Q_l)$	−1.11	−1.10	−1.03	−0.99	−0.99	−0.97	−0.94	−0.97	0.20	−0.91	−0.41	0.35
$1/S$	−0.82	−0.82	−0.63	−0.70	−0.61	−0.73	−0.50	−0.67	−0.40	−0.66	−0.46	−0.18
L	−2.19	−2.16	−1.95	−1.67	−1.82	−1.62	−1.31	−1.60	−0.79	−1.54	−0.90	−0.67

ESI Table 2: Calculated metrics for the full range of configurations (presented as logarithms for ease of comparison.) The metrics are: the fit to the PDF calculated from a 4096 atom configuration generated using the WWW algorithm; the PDF variance measured using the INVERT metric; the variance of the bond orientational order coefficients, Q_l; the reciprocal of the symmetry measure S and the SOAP similarity measure L. The anomalously high values of χ^2_{PDF} for the WWW and relaxed configurations arise from misfits in the width of the nearest neighbour peak relative to the reference 4096 WWW derived configuration.



ESI Fig. 11: Calculated powder X-ray diffraction patterns created using the CrystalDiffract software of three configurations, illustrating the lack of long-range order for the L and WWW configurations.

References

(S1) M. J. Cliffe, M. T. Dove, D. A. Drabold, A. L. Goodwin, *Phys. Rev. Lett.* **104**, 125501 (2010).
(S2) M. J. Cliffe, A. L. Goodwin, *Phys. Status Solidi B* **250**, 949 (2013).
(S3) A. P. Bartók, R. Kondor, G. Csányi, *Phys. Rev. B* **87**, 184115 (2013).
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(S6) L. Goodwin, A. J. Skinner, D. G. Pettifor, *Europhys. Lett.* **9**, 701 (1989).