Structural simplicity as a constraint on the structure of amorphous silicon

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March 13, 2017

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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 constraint 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 constraint, the spherical harmonic bond-orientational order parameter variance constraint, and the spherical harmonics local symmetry constraint applied[S1, S2].

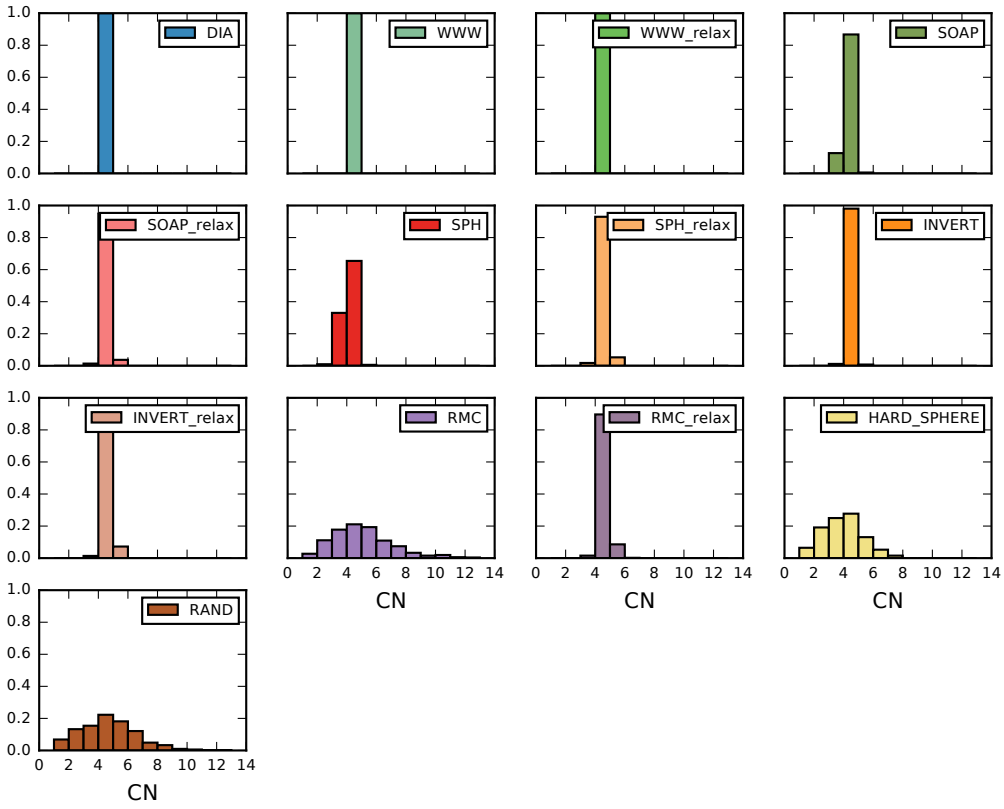
SPH relax The SPH configuration optimised with DFT using the PBE functional until convergence.

SOAP A random configuration refined against PDF data taken from a 4096 atom WWW configuration using RMC, with the SOAP variance constraint also applied[S3].

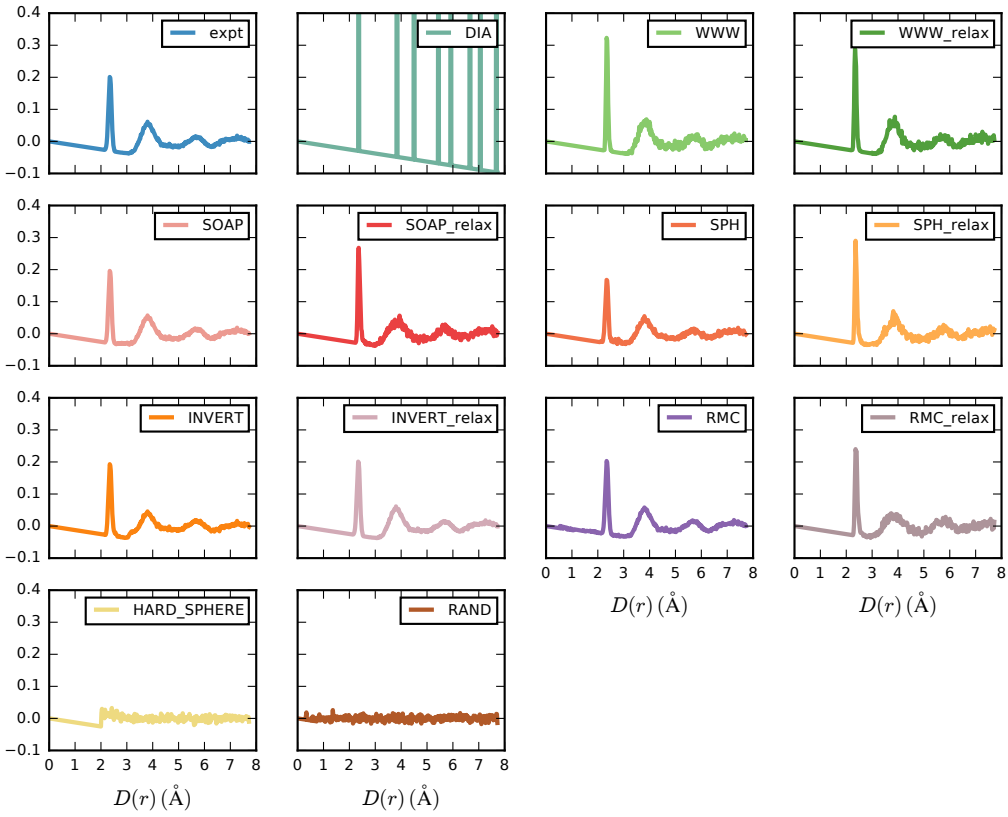
SOAP relax The SOAP configuration optimised with DFT using the PBE functional until convergence.

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

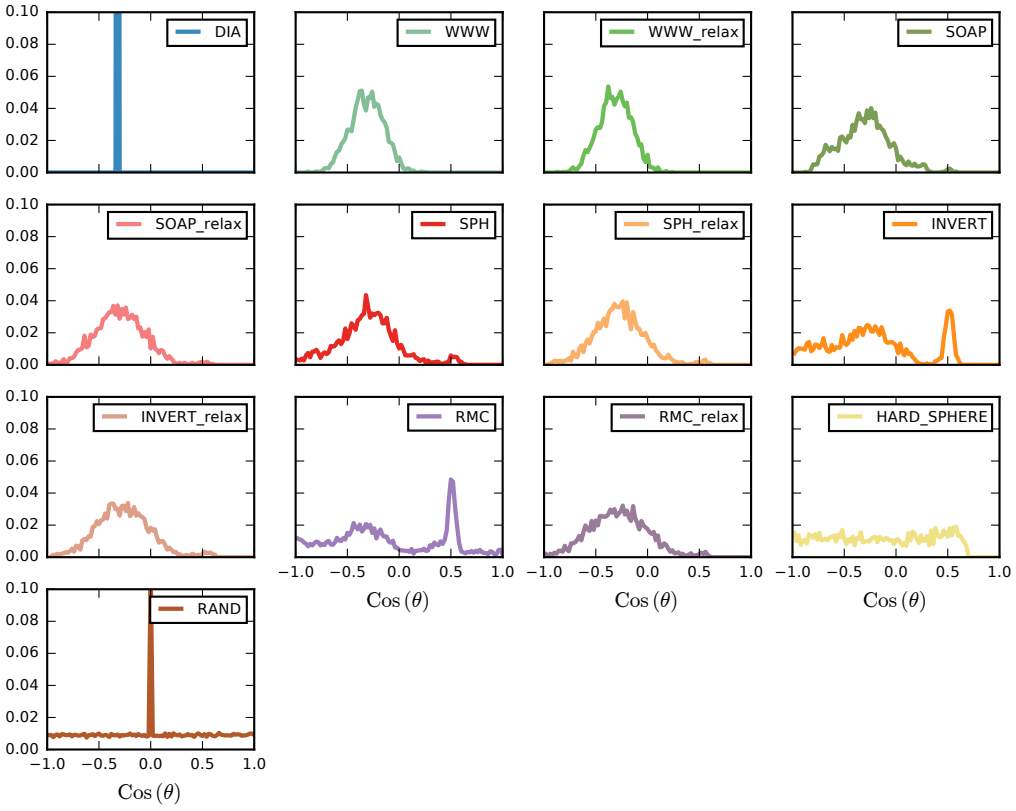
WWW relax The WWW configuration optimised with DFT using the PBE functional until convergence.



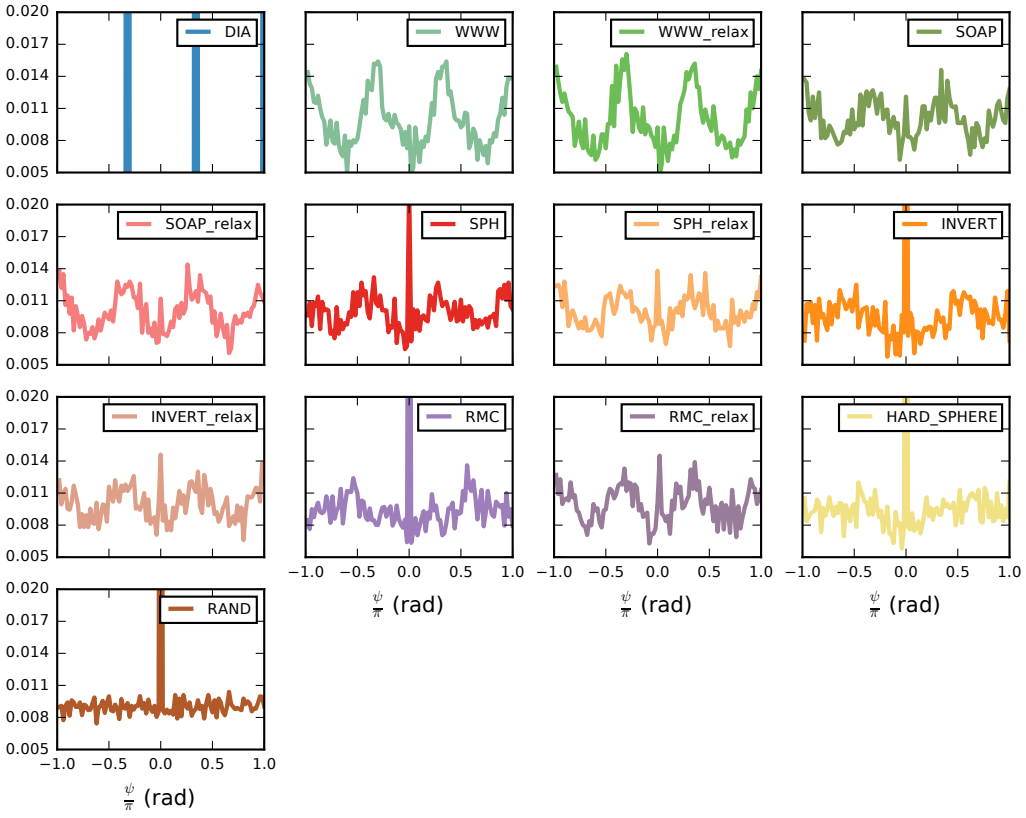
ESI Fig. 1: Histograms of the coordination number distributions for all configurations



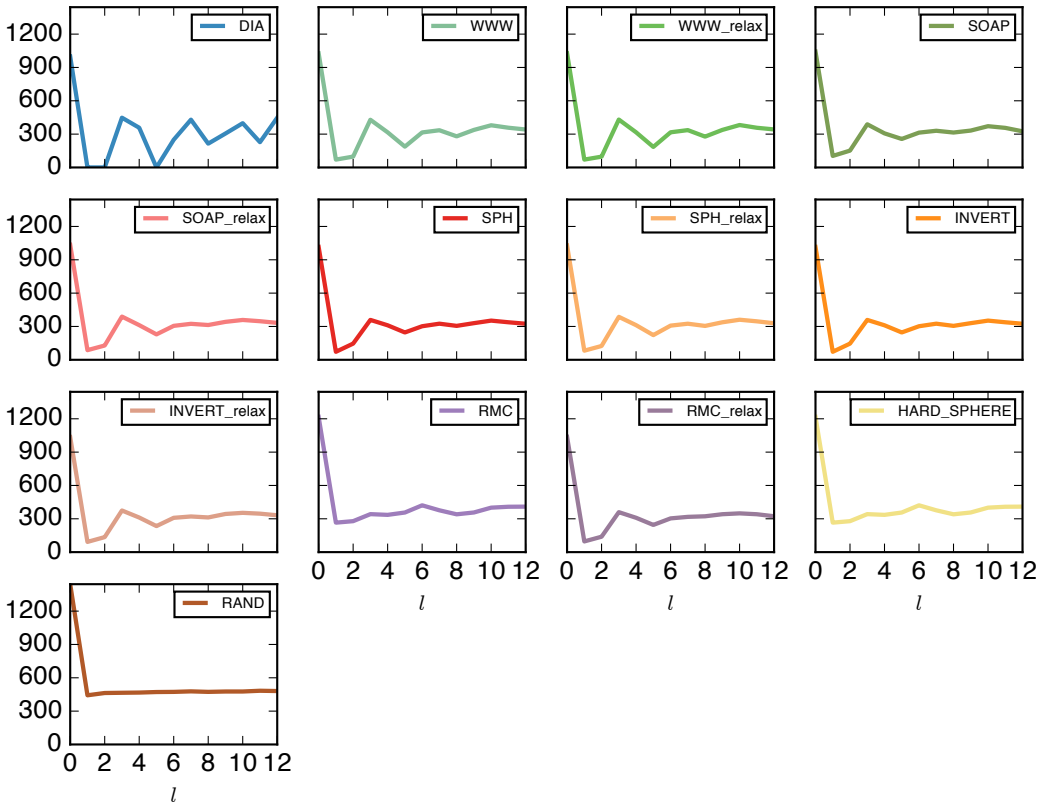
ESI Fig. 2: Pair distribution functions for all configurations



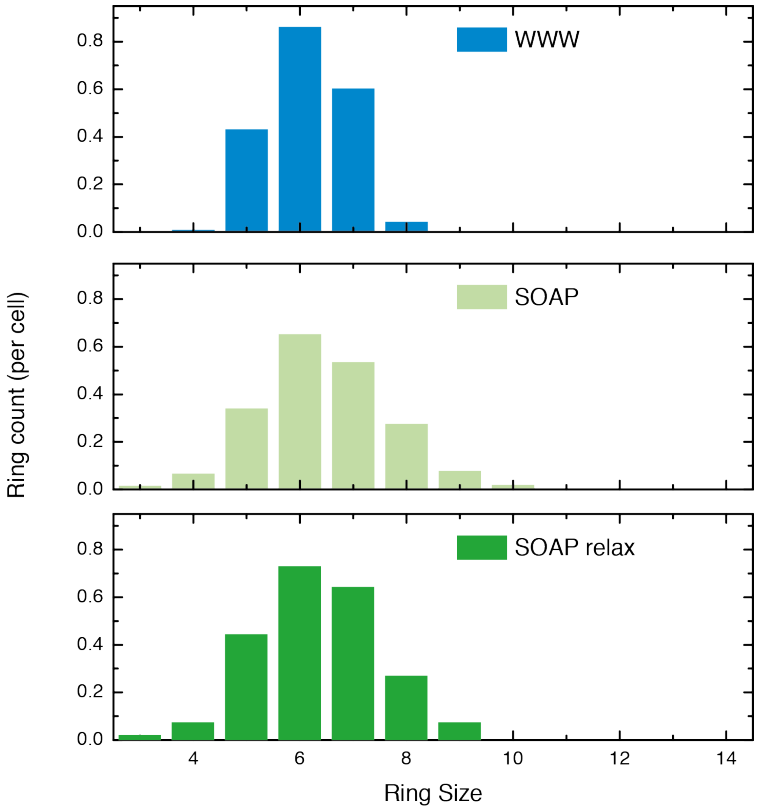
ESI Fig. 3: Histograms of the bond angle distributions for all configurations. The large spike at $\cos(\theta) = 0$ for the random configuration results from near overlapping atoms.



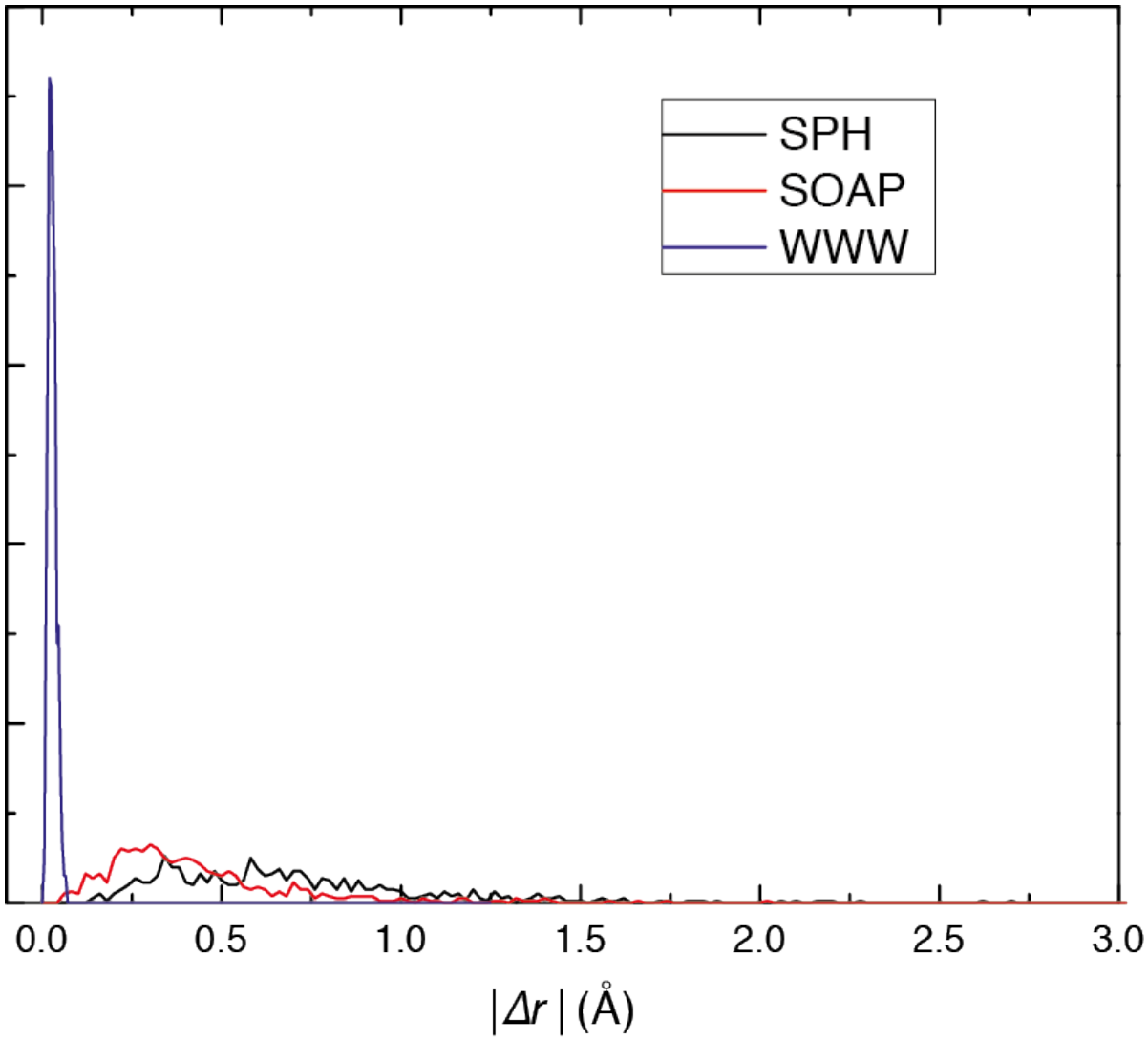
ESI Fig. 4: Histograms of the dihedral angle distributions for all configurations. The spike at $\psi = 0$ results from three-membered rings, which are necessarily completely planar.



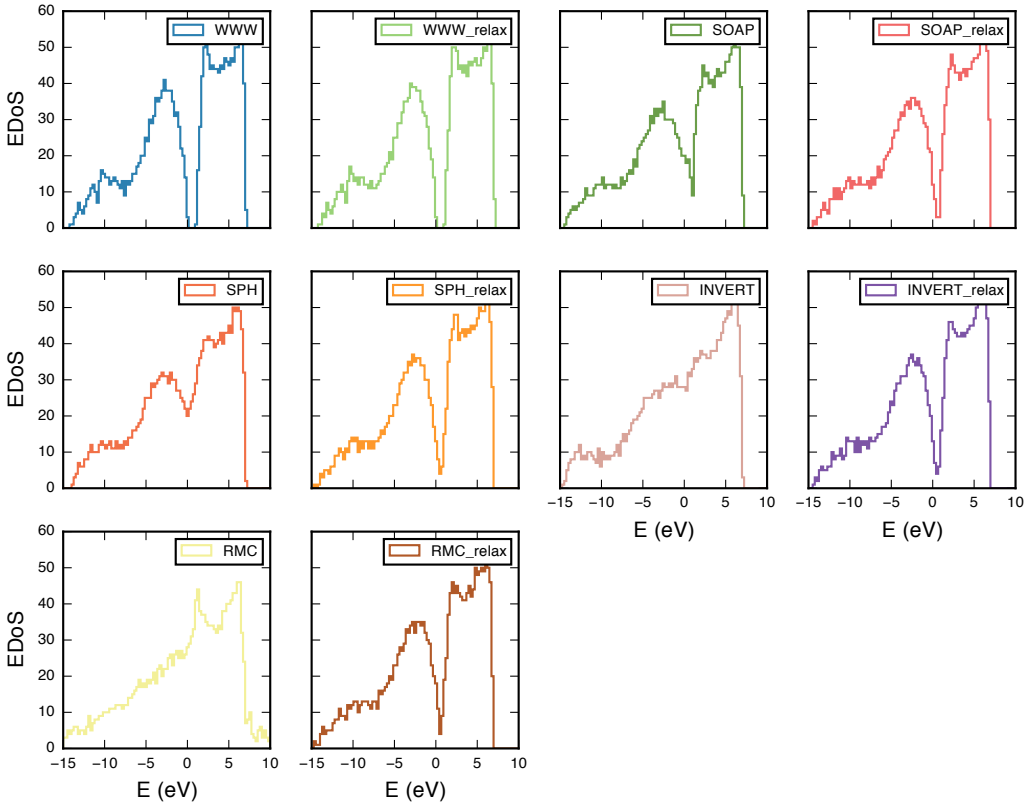
ESI Fig. 5: Calculated average Q_l coefficients for all configurations.



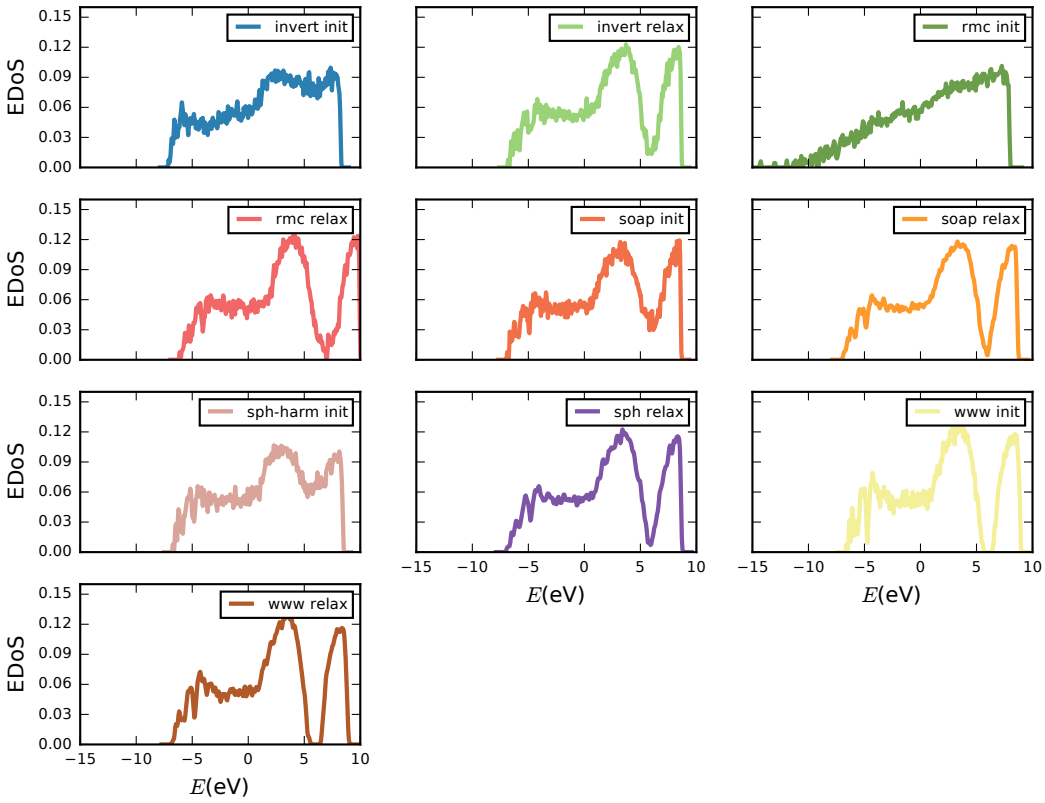
ESI Fig. 6: Ring statistics (using the King's shortest path convention and normalised by number of rings per cell) for the WWW configuration and the SOAP configuration before and after relaxation. Calculated using ISAACS[S5].



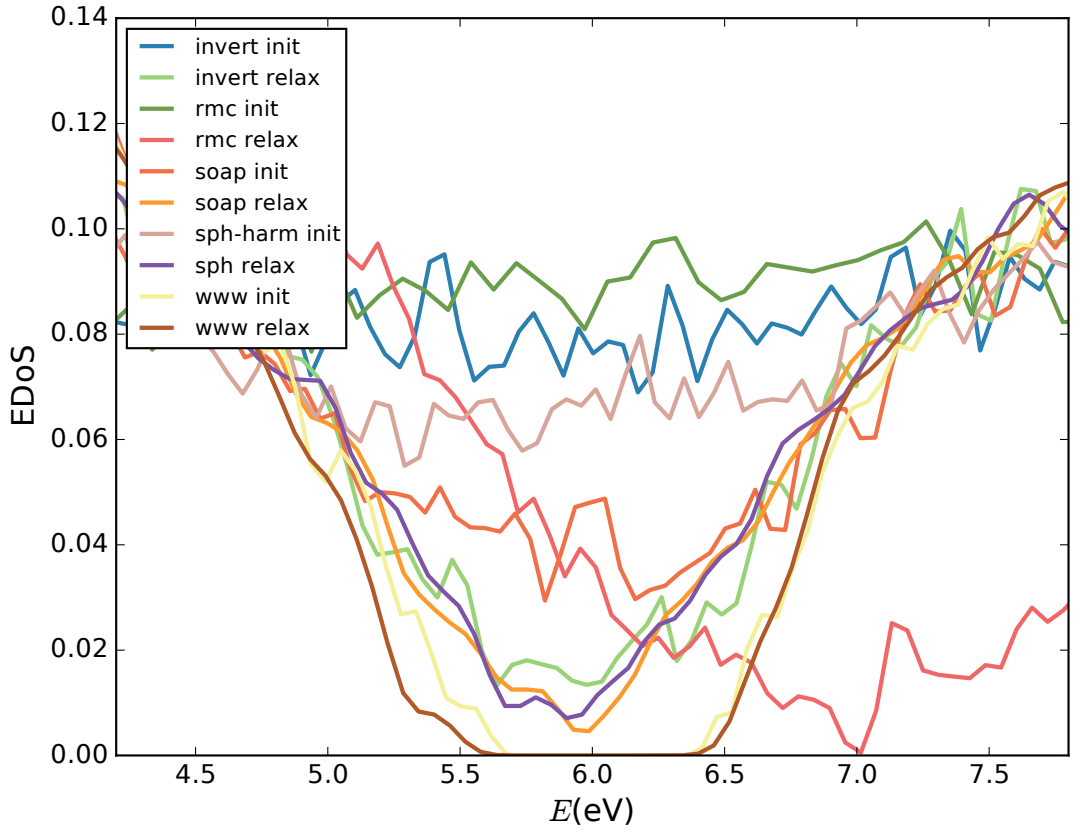
ESI Fig. 7: Histogram of the root square displacement during optimisation for the WWW, SOAP and SPH configurations.



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	SOAP	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	SOAP	SOAP 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.

References

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(S5) S. Le Roux, V. Petkov, *J. Appl. Crystallogr.* **43**, 181 (2010).
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