Molecular Properties of the Ice Ih Basal Surface

An Application of the SCME Water Interaction Potential

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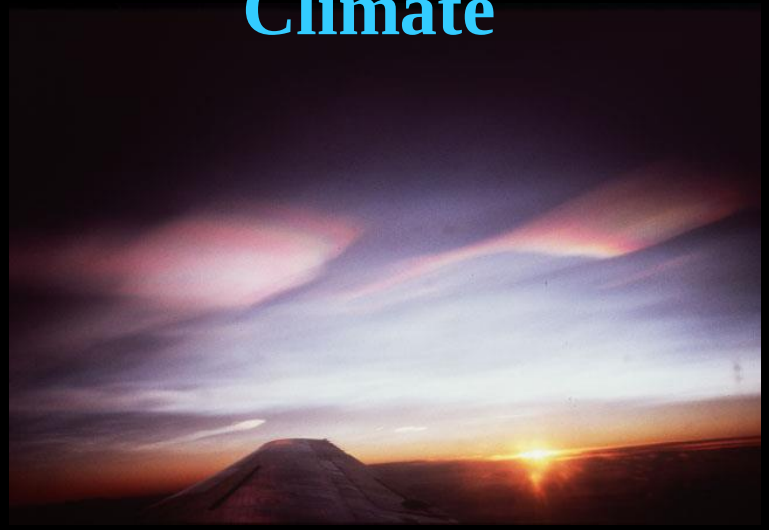
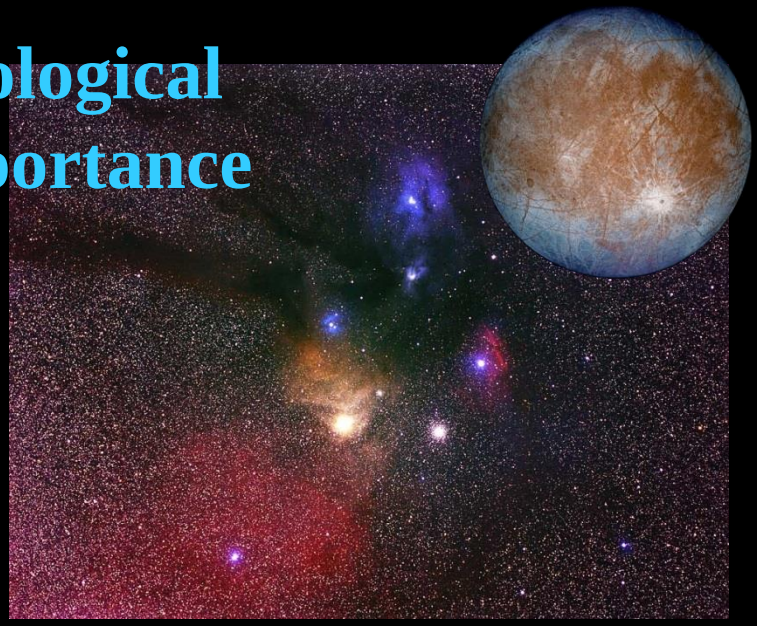
Why Study Ice?

Environment, Weather and Climate



Chemical Significance

Biological Importance



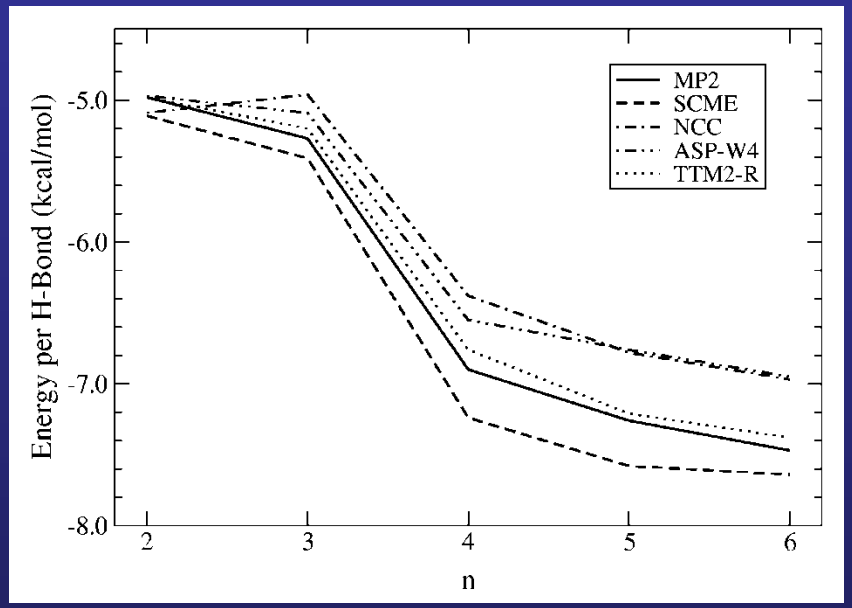
The SCME Potential

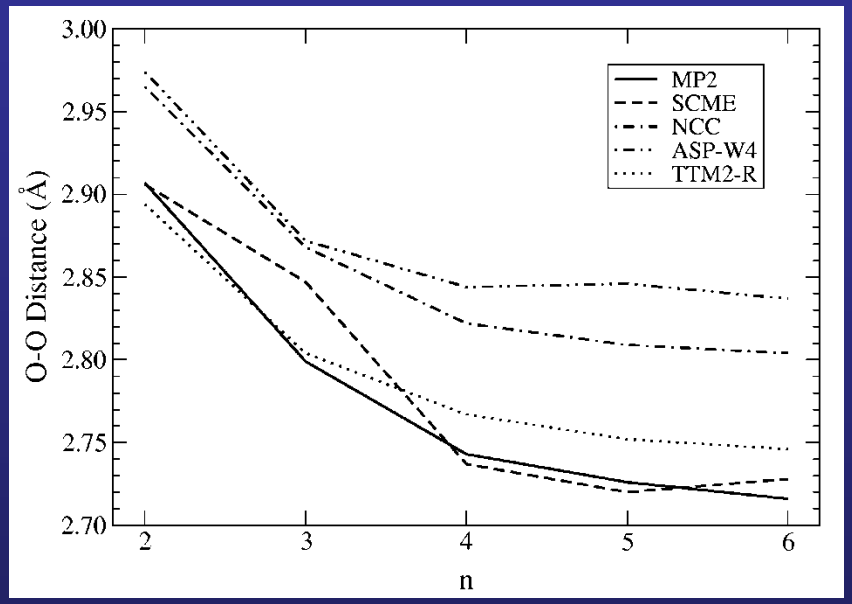
- Based on a Single Center Multipole Expansion
- Does not use point charges
- Rigid
- Transferable
- Parametrized using a mixture of *ab initio* and empirical values

Total Energy:

$$E_{\mathrm{tot}} = E_{\mathrm{es+ind}} + E_{\mathrm{disp}} + E_{\mathrm{rep}}$$

$(H_2O)_{n=2-6}$ ring water clusters:

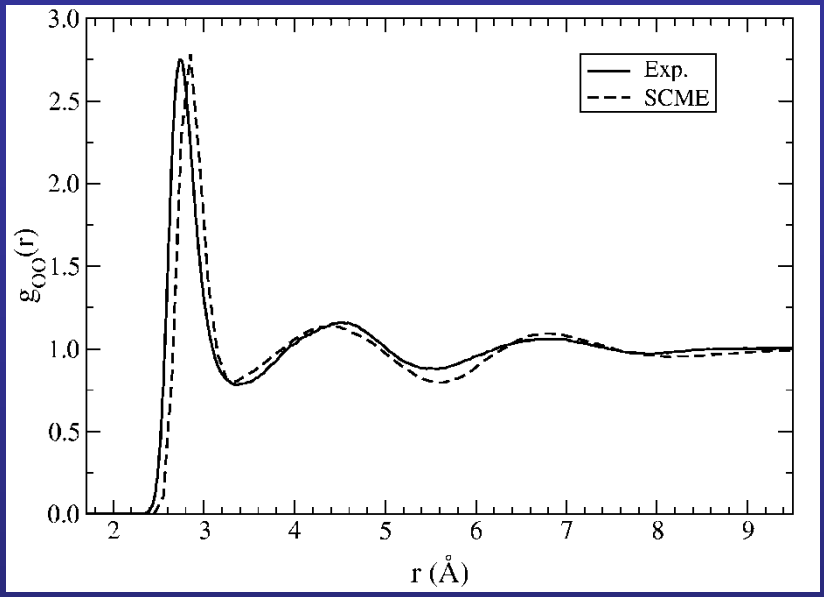


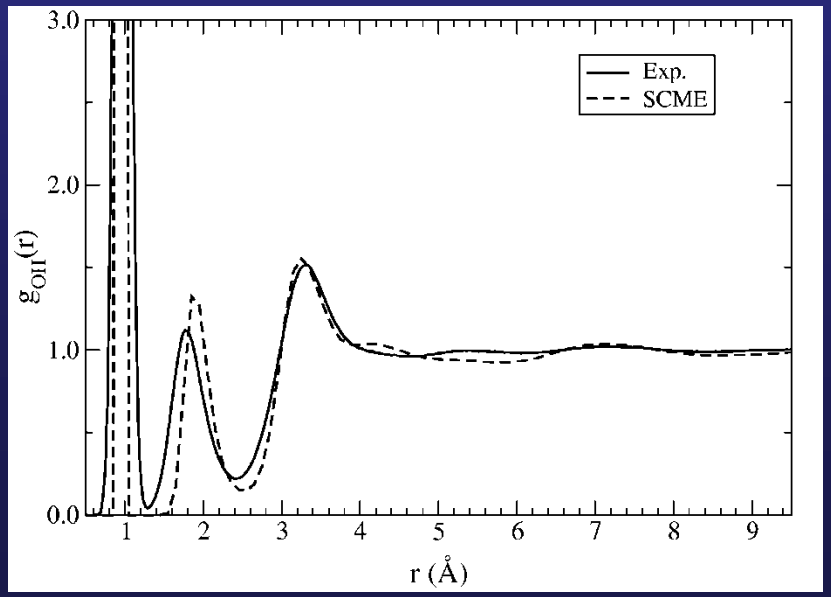


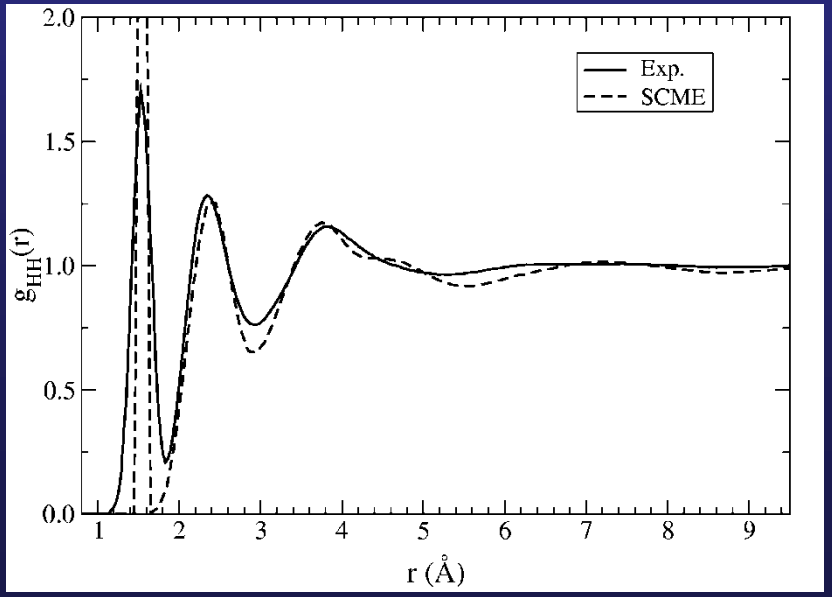
Interaction Energy per H-Bond

Average O-O Distance

Radial Distribution Functions for Liquid Water:







Bulk ice Ih:

Property	Exp.[a]	GGA	PW91[b]	SCME	TIP4P	RWK2[c]	DC	TTM2-R
Lattice Energy[d]	−0.6110		−0.55	−0.6109±0.0049	−0.634	−0.555	−0.550	−.6370
$\langle r_{OO} \rangle$[e]	2.751	2.70		2.742±0.004	2.683		2.738	
a[e]	4.4969	4.41		4.470±0.025				4.478
b[e]	7.7889	7.63		7.747±0.052				7.756
c[e]	7.3211	7.20		7.287±0.029				7.314
ρ[f]	0.933	0.989	0.954	0.948±0.004	1.009	0.942	0.960	0.942
V_{molec}[g]	32.05	30.3	31.35	31.55±0.15	29.62	31.73	31.14	31.75
Bulk Modulus[h]	10.9		13.5	11.4±0.3	16.6	18.0		
μ_{molec}[i]	2.90		2.8	3.50±0.07	2.35		3.02	2.86[j]

[b] All values taken from Ref.[9] except the bulk modulus, taken from Ref.[78].
[b] From Ref.[79]
[c] From Ref.[75]
[d] In eV/molec
[e] In Å
[f] In Å^3/molec
[g] In g/cm^3
[h] In MPa
[i] In Debye
[j] Calculated at 100K

Simulation of the Ice Ih Basal Surface

General Properties of the Cells:

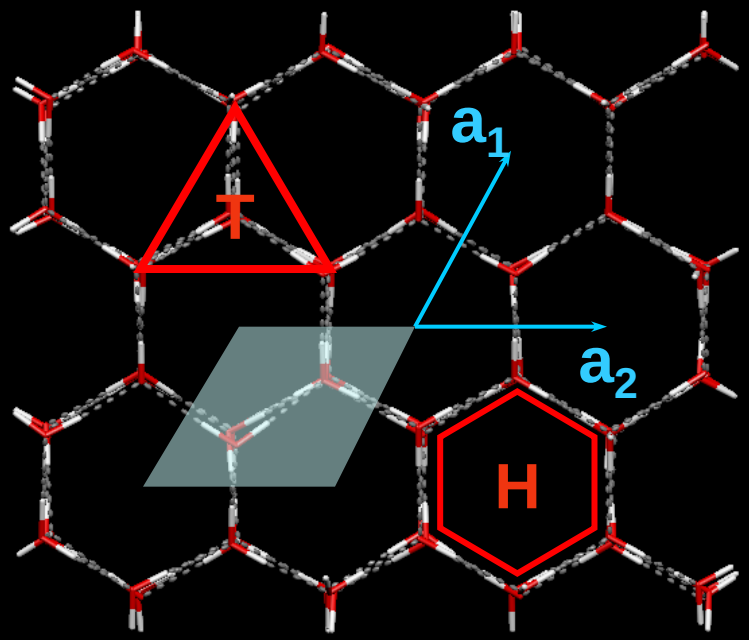
- Proton-disordered
- Null total dipole
- Four bilayers
- Bottom bilayer frozen at bulk conformation

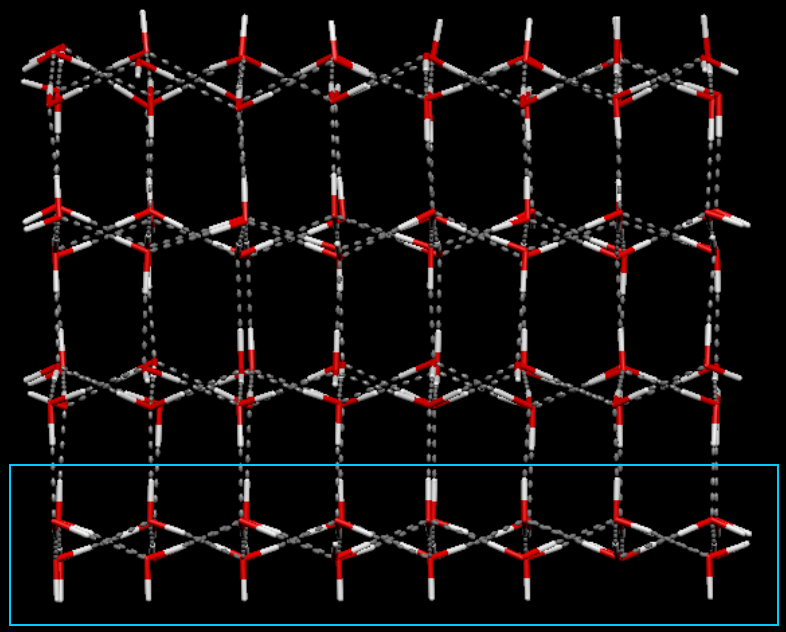
5 Small Cells:

- 3 x 2 x 2 repetitions of the orthogonal cell
- 96 molecules per cell

10 Large Cells:

- 4 x 2 x 2 repetitions of the orthogonal cell
- 128 molecules per cell


T
H
a_1
a_2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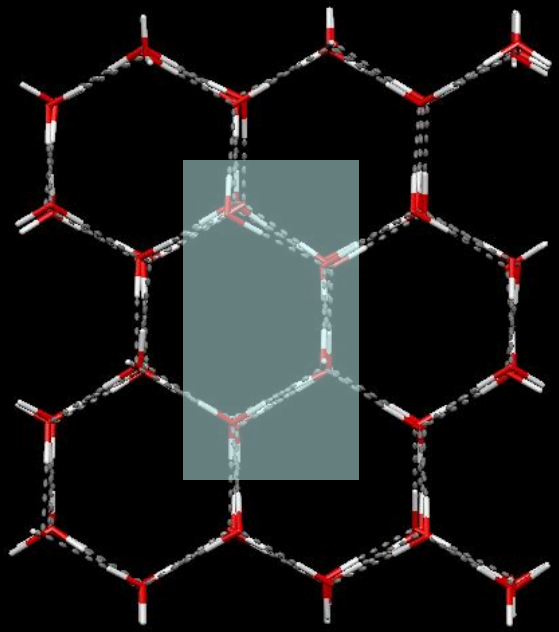
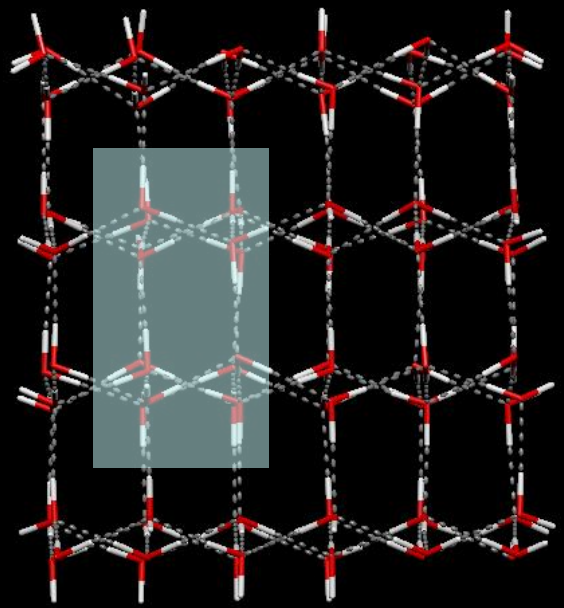




Binding energy (in eV) for the different types of minima:

	All			Large			Small		
	#	E	ΔE	#	E	ΔE	#	E	ΔE
DFAAH	61	-0.612	0.097	40	-0.606	0.110	21	-0.623	0.074
DDA-T	52	-0.607	0.078	38	-0.611	0.081	14	-0.597	0.072
DDA-H	20	-0.590	0.070	13	-0.593	0.071	7	-0.586	0.068
DFAAT	24	-0.578	0.066	13	-0.570	0.071	11	-0.585	0.061
DFA-I	41	-0.539	0.074	33	-0.543	0.076	8	-0.525	0.066

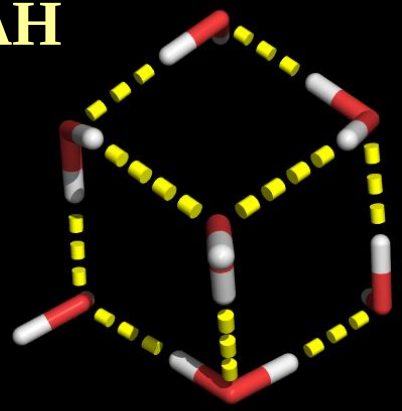
One accepted Hydrogen-Bond
One "free" lone pair

DFA-H

One donated hydrogen
One free Hydrogen

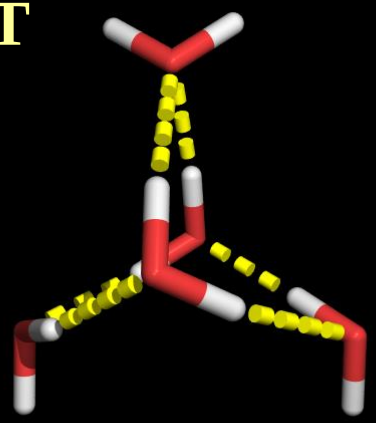
Over a hexagonal site

DFAAH



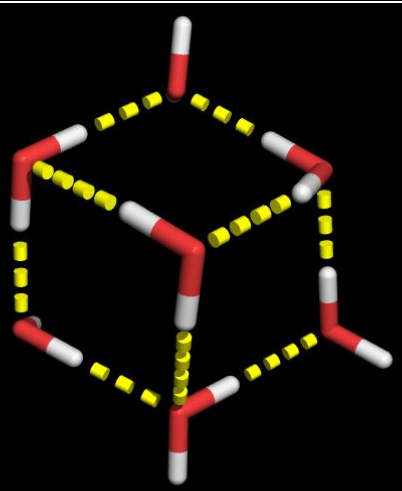
E_b = -0.612 ± 0.097 eV

DDA-T



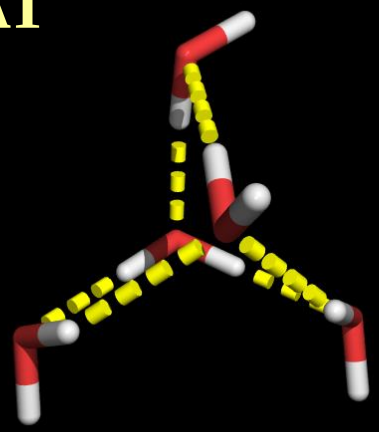
E_b = -0.607 ± 0.078 eV

DDA-H



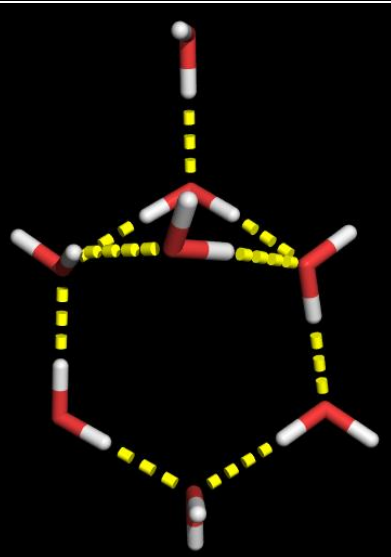
E_b = -0.590 ± 0.070 eV

DFAAT



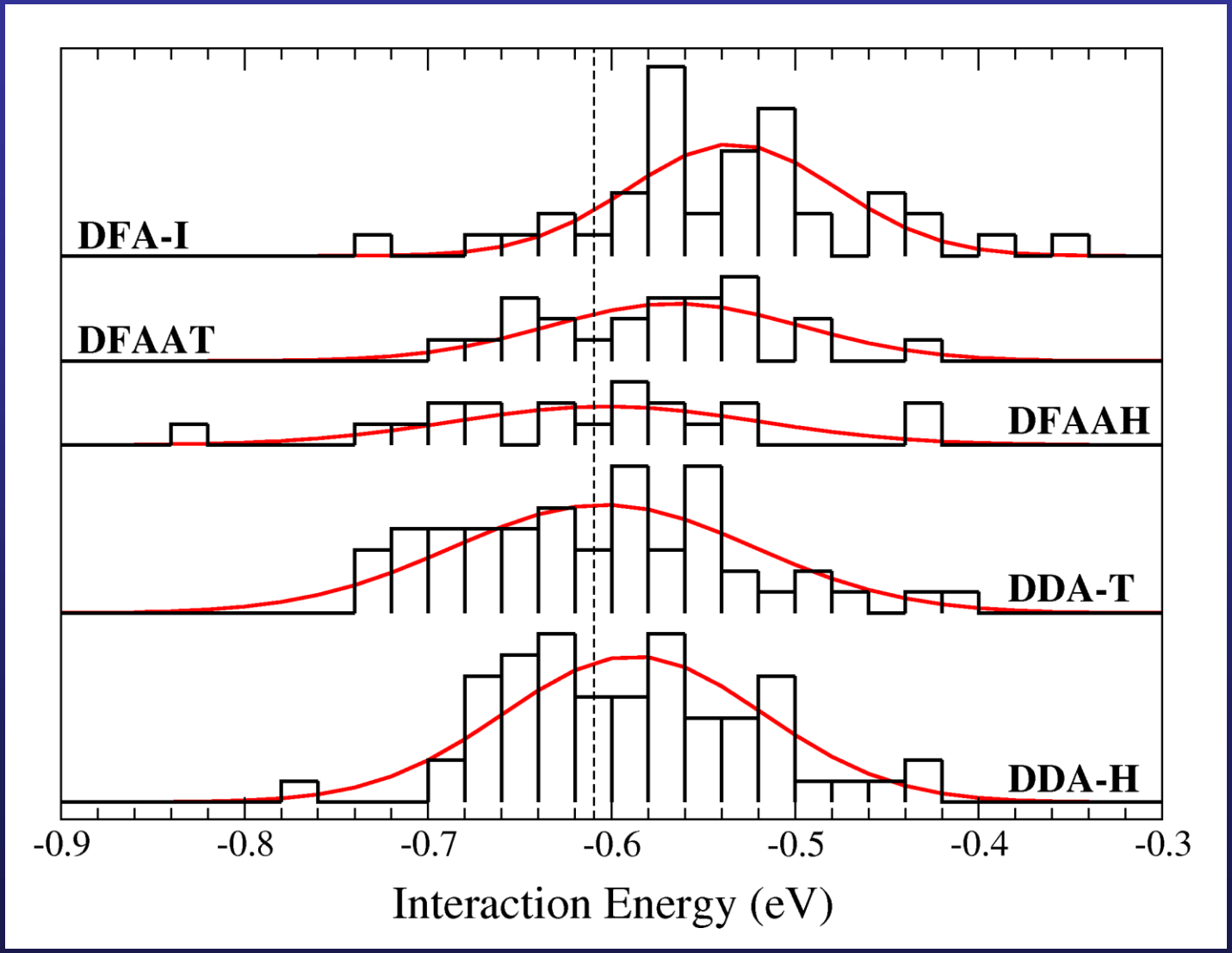
E_b = -0.578 ± 0.066 eV

DFA-I

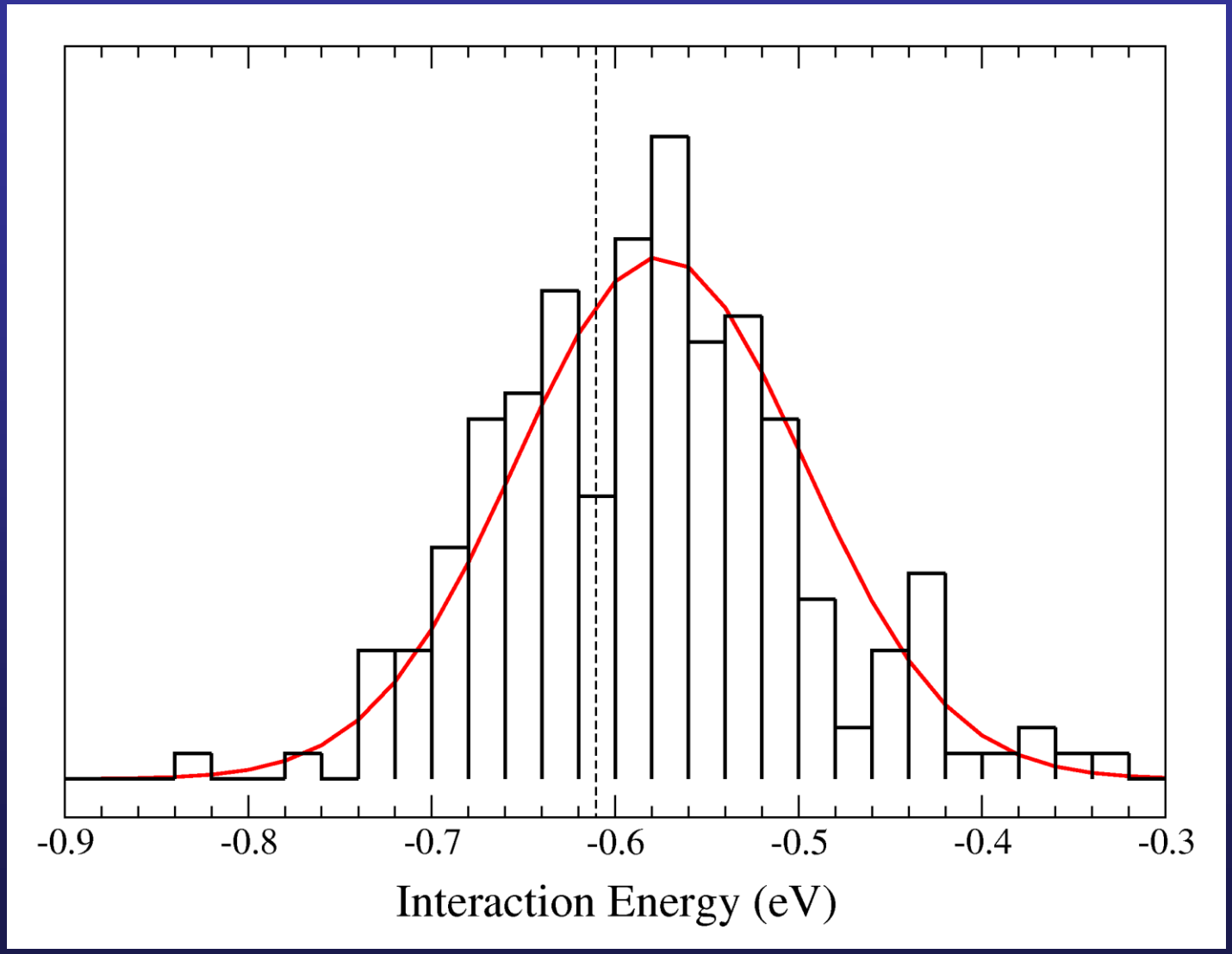


E_b = -0.539 ± 0.074 eV

Energy histogram for each type of minima:



Full histogram:



Rounding up..

SCME:

- Good behavior over a wide range of conditions: water clusters, liquid water and bulk ice
- Fast
- Able to predict properties that were not included in the parametrization

Adsorption:

- There are five prevalent types of sites for adsorption
- The distributions of the binding energies for each type of site are very similar
- Approximately 40% of the sites have a binding energy larger than the lattice energy of bulk ice Ih

Future

SCME:

- Improved version of the potential that fixes some of the problems
- Faster coding and better software distribution

Adsorption:

- Study of site-to-site transitions
- Use results to estimate surface diffusion rate
- Study multiple adsorptions/surface interactions

“Pet” proyect:

- Modeling of the surface acoustic phonon modes
- Use results to estimate surface diffusion rate
- Study multiple adsorptions/surface interactions