Figure Captions

Fig. 1: Global minimum energy structures for $(H_2O)_n$, $n=12$, for the TIP4P potential (left panel) and the TTM2-F potential (right panel), with corresponding total energies after local optimization with each potential. For a better understanding of the three-dimensional structures, the following "color" coding is used for the oxygen atoms: light grey = in front or intermediate distance, dark grey = in the back, white = non-surface internal molecules.

Fig. 2: As Fig. 1, but for $n=13$.

Fig. 3: Global minimum energy structures for $(H_2O)_n$, $n=14,15,16$, with the TTM2-F potential. The best structures for TIP4P show a slightly different orientational pattern for $n=15,16$, see text. Color coding as in Fig. 1.

Fig. 4: As Fig. 1, but for $n=17$.

Fig. 5: Global minimum energy structures for $(H_2O)_n$, $n=18,19,20$, with the TTM2-F potential. The best structure for TIP4P shows a slightly different orientational pattern for $n=19$, see text. Color coding as in Fig. 1.

Fig. 6: As Fig. 1, but for $n=21$.

Fig. 7: Several important minimum structures for $n=22$. The centered-cage structure 1 is the global minimum for both TTM2-F and TIP4P. The all-surface structure 2 is a very low-energy local minimum for both potentials. The centered-cage isomer 3 is more preferred by TTM2-F than by TIP4P. Color coding as in Fig. 1.

Fig. 8: Selected TTM2-F minimum structures between $n=24$ and $n=30$, suggesting a transition from one to two internal molecules in the cage at $n=28$, and a cage growth pattern via cuboid and prismoid extrusions. The structures for $n=24,26,28$ are true proposals for global minima. The structure for $n=27$ is a low-energy local minimum. For $n=29,30$, we are not sure to have captured the global minima yet.

Fig. 9: Measures for cluster stability as a function of cluster size n. "Fit deviation" gives the deviations from a least-squares fit of cluster binding energies to a quadratic form. "Stability" is the commonly used stability function. Both graphs are arbitrarily scaled to the same value at $n=20$. The additional outlier point at $n=20$ is the fit deviation of the "four cubes" local minimum structure.

Fig. 10: The global minimum energy cage structure of $n=17$ with the TTM2-F potential. Lengths and directions of the arrows attached to the water monomers show relative size and orientation of the total induced dipoles for each monomer. In this cluster projection, deviations of these dipoles from the HOH plane and from the H-O-H angle bisector are clearly visible for the two molecules in the top right corner of the graph.

Fig. 11: Simulation of IR spectra of the TIP4P all-surface (top) and the TTM2-F centered-cage (bottom) forms of $n=17$, with assignments of a few characteristic peaks. The two spectra can be told apart rather easily by the different peak pattern in the region $3050 - 3350$ cm^{-1}.
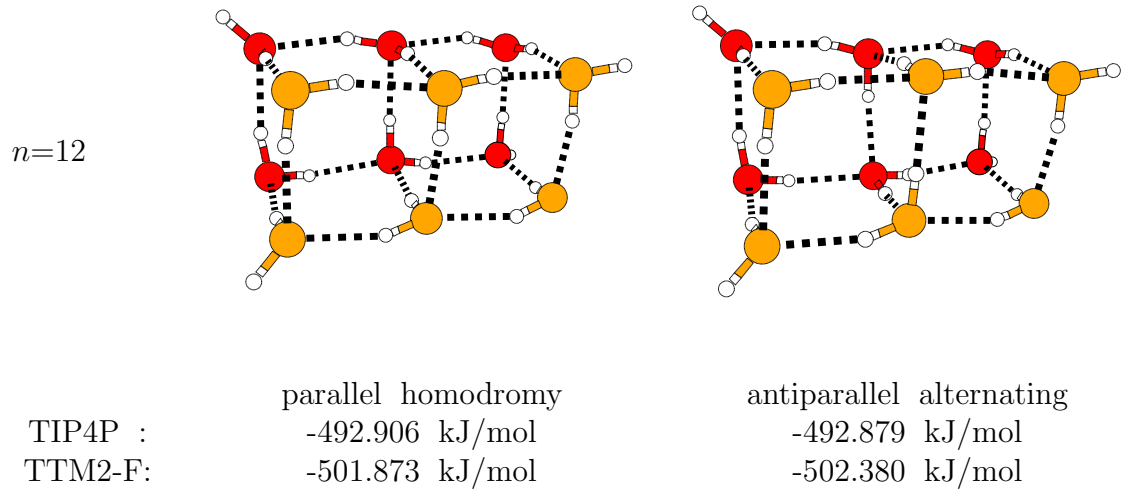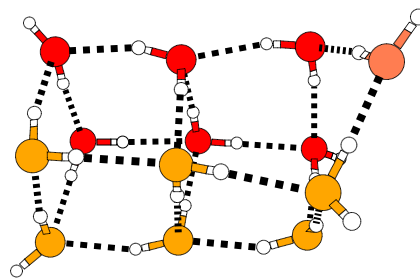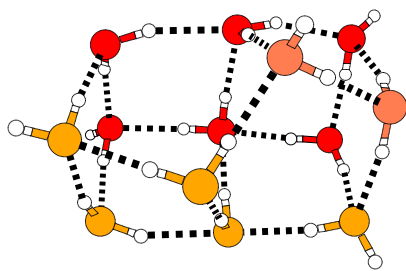
Fig. 12: As Fig. 11, but for $n=21$.

$n=12$

	parallel homodromy	antiparallel alternating
TIP4P :	-492.906 kJ/mol	-492.879 kJ/mol
TTM2-F:	-501.873 kJ/mol	-502.380 kJ/mol

Figure 1: Hartke

$n{=}13$

	middle corner dimerized	terminal edge capped
TIP4P :	-532.971 kJ/mol	-532.969 kJ/mol
TTM2-F:	-541.502 kJ/mol	-545.733 kJ/mol

Figure 2: Hartke
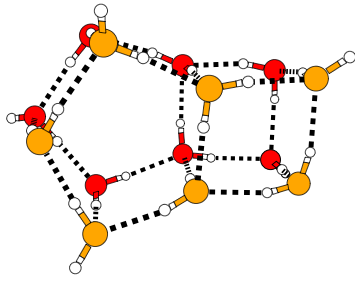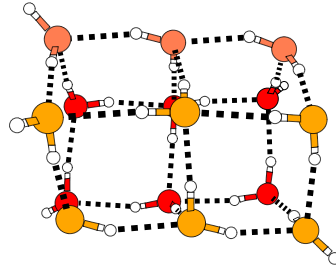
$n = 14$: cube & pent.prism $n = 15$: pent.prism stack $n = 16$: cube stack
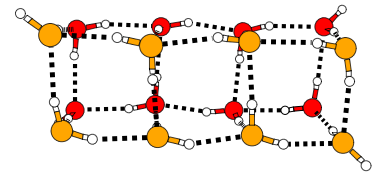
Figure 3: Hartke

$n=17$
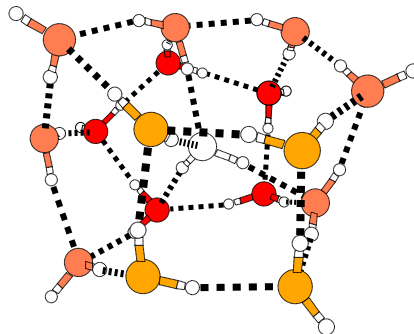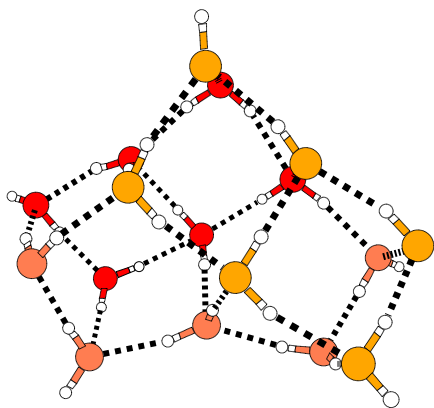
	all surface	interior water
TIP4P :	-723.808 kJ/mol	-721.743 kJ/mol
TTM2-F:	-742.062 kJ/mol	-746.734 kJ/mol

Figure 4: Hartke
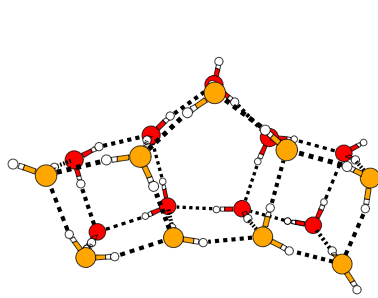
$n = 18$: 2 cubes & pent.prism $n = 19$: water-centered cage $n = 20$: 3 pent.prisms
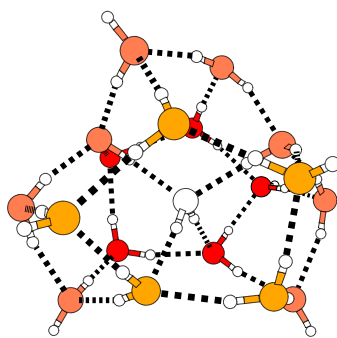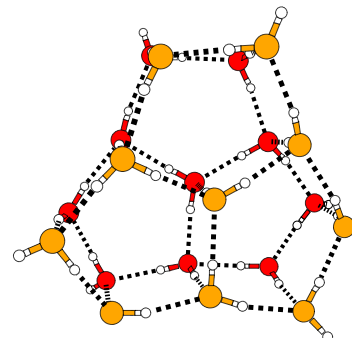
Figure 5: Hartke

$n=21$
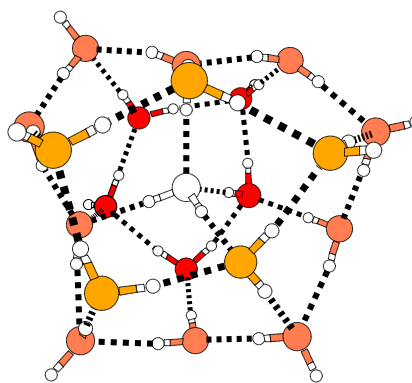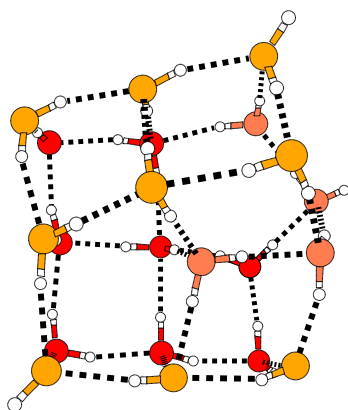
	all surface	centered dodecahedron
TIP4P :	-916.708 kJ/mol	-915.230 kJ/mol
TTM2-F:	-944.712 kJ/mol	-952.266 kJ/mol

Figure 6: Hartke
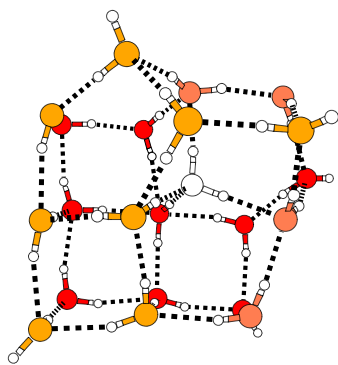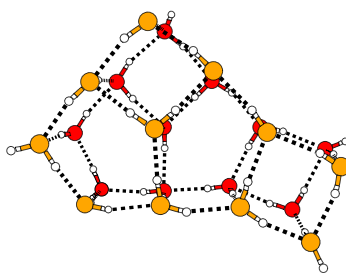
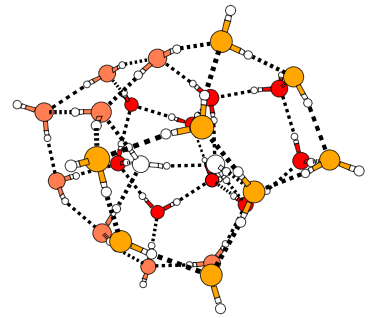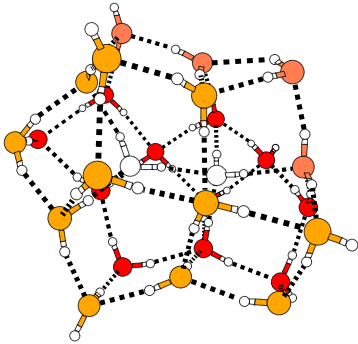$n = 22$: structure 1 $n = 22$: structure 2 $n = 22$: structure 3
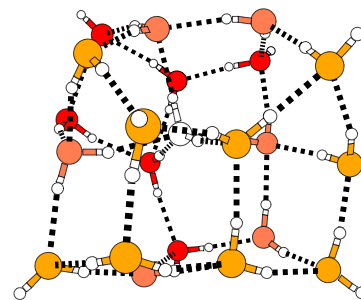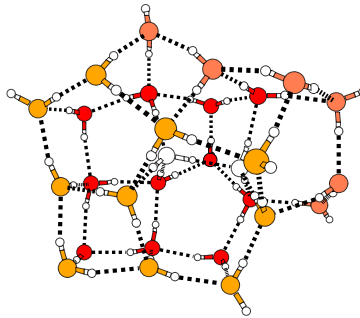
Figure 7: Hartke
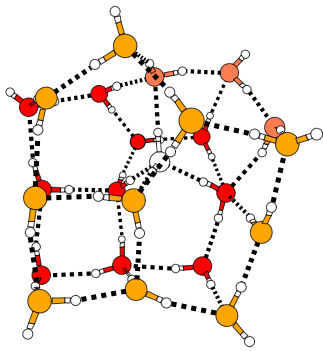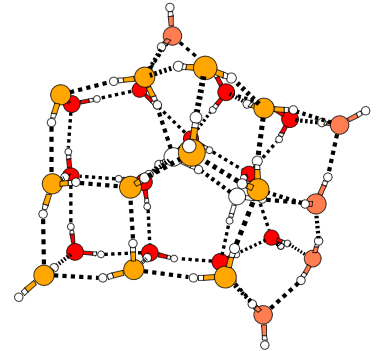
$n = 24$: cube & cage
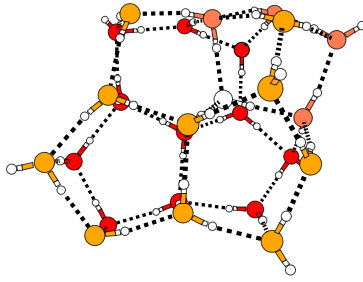
$n = 26$: pent.prism & cage

$n = 27$: two molecules in cage

$n = 28$: two molecules in cage

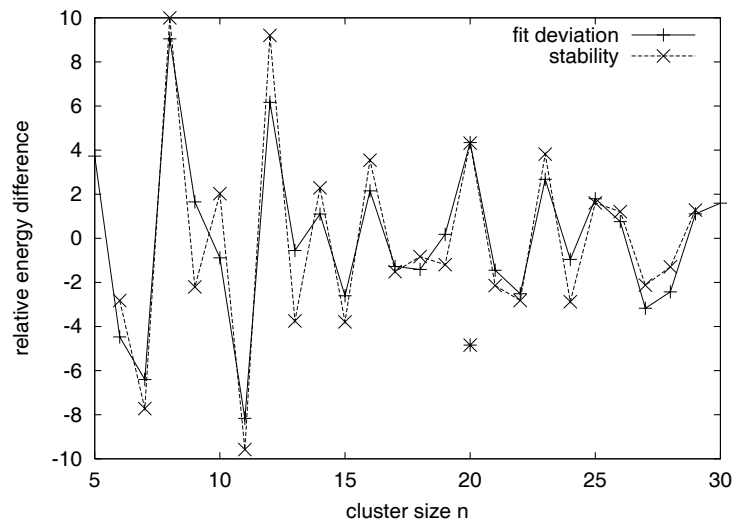$n = 29$: cube & cage
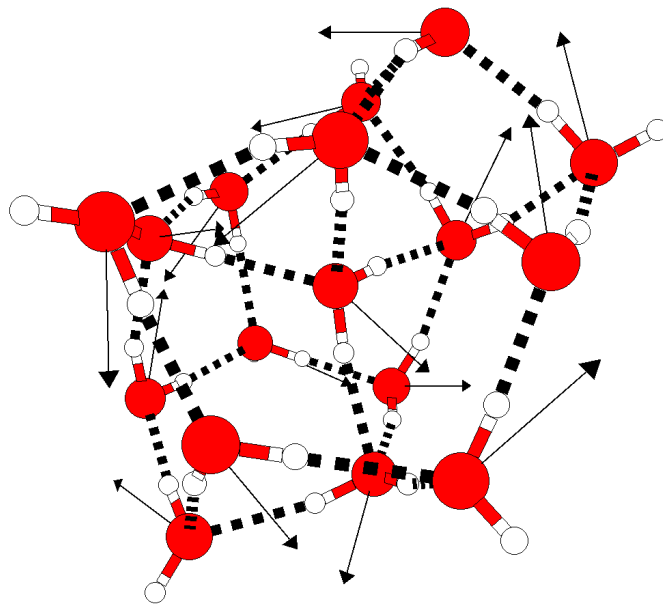
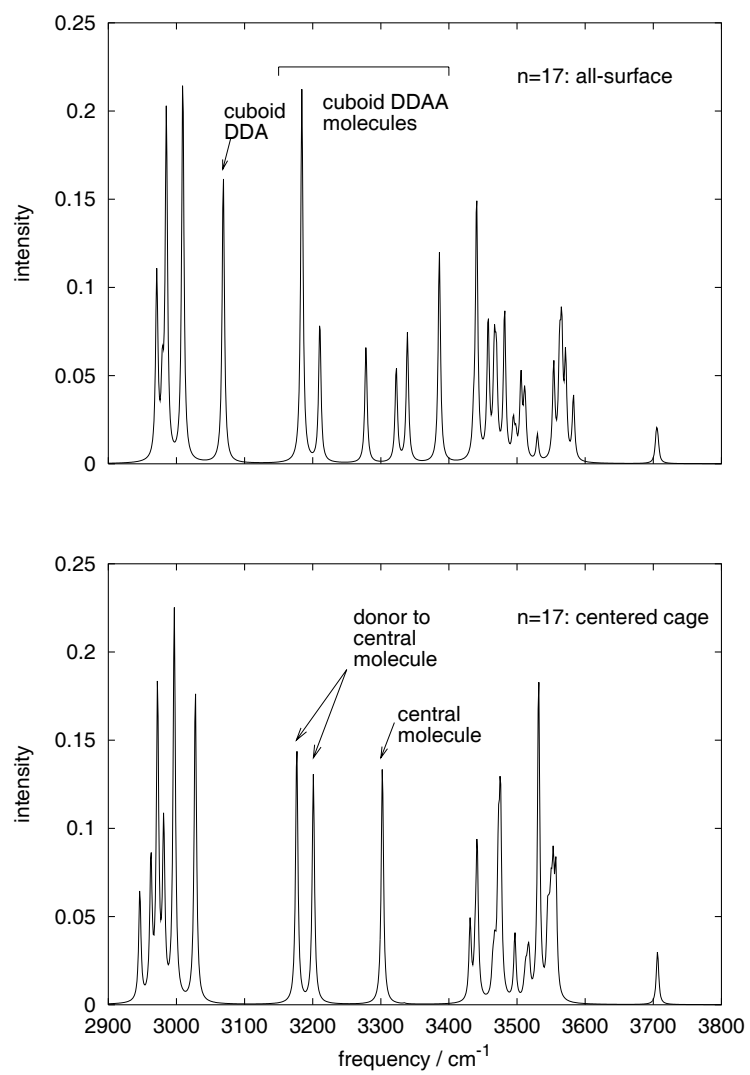$n = 30$: cube & cage

Figure 8: Hartke

Figure 9: Hartke
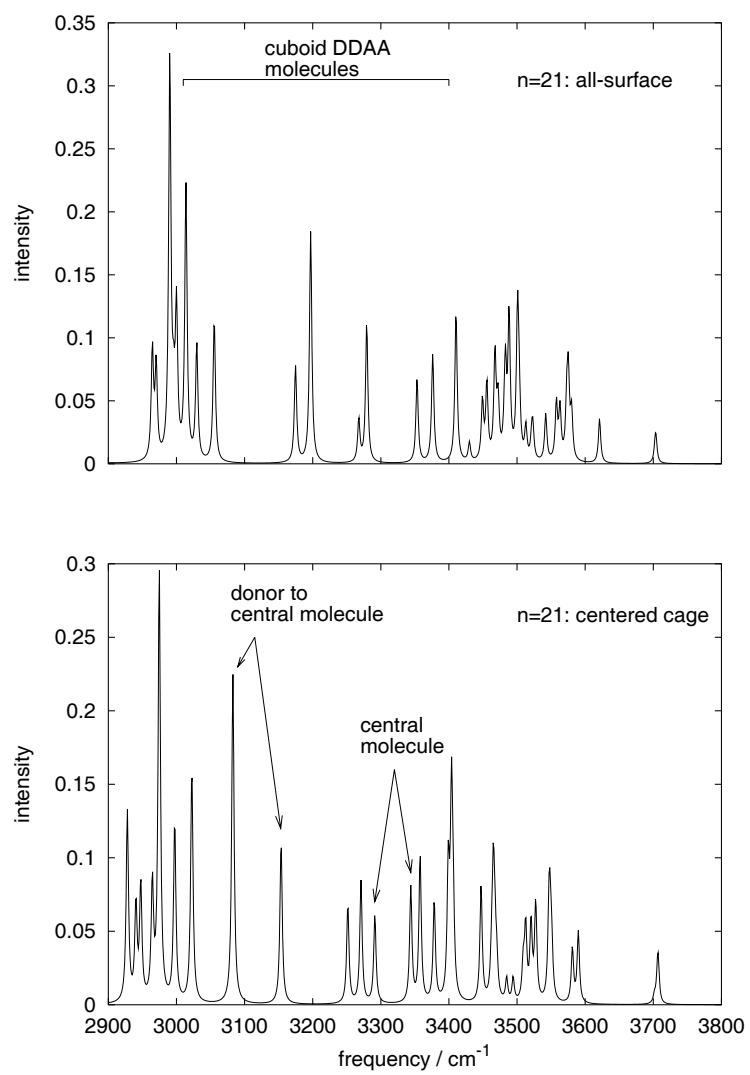
Figure 10: Hartke

Figure 11: Hartke

Figure 12: Hartke