Supporting Information

ADF-specific implementational details

Normal mode vectors are expressed by ADF in machine accuracy in the `TAPE21` (`*.t21`) binary dump format. This file can be converted into an ASCII-formatted file *via* the `DMPKF` utility distributed with ADF. We implemented a post-processor to obtain the normal mode vectors from ADF frequency calculations in a form amenable to our numerical differentiation scheme.

We accelerated computation of the dipole derivatives and encourage facile SCF convergence by preconditioning the SCF *via* reuse of the molecular orbital composition obtained from geometry optimization. This is necessary to obtain good performance due to the overhead of the characteristically slow recomputation of the initial SCF for each finite difference step.

Discussion of anomalous CPU-time of heptane MBH region 4

In the case of heptane, it is noted that the MBH normal mode computation for region 4 took an anomalously long time with respect to other regions, with consequent effects for the total calculation time. The cause of this anomaly is unknown, however in light of the fact that the calculation was performed using 4 concurrent threads on a cluster (the *Raijin* cluster housed at the NCI National Facility at the Australian National University) consisting of 16-core compute nodes, it is plausible that Von Neumann bottlenecking occurred due to competition with other memory-intensive calculations simultaneously allocated to the remaining 12 cores on the node.