Supplementary Information for "Impurity effects on solid-solid transitions in atomic clusters"

B. E. Husic,[1,*] D. Schebarchov,[1,†] and D. J. Wales[1,‡]

[1] *University Chemical Laboratories, Lensfield Road, Cambridge CB2 1EW, UK*

(Dated: September 19, 2016)

GENERALISED BASIN-HOPPING

Generalised[1] basin-hopping[2,3] (GBH) is a global optimisation method, which can be described as a Markov chain with the canonical Metropolis[4] acceptance criterion in the space of *multiminima* (configurations that are locally optimal with respect to multiple metrics). For homoatomic clusters considere in this study, each trial configuration in the Markov chain is generated from the last accepted one by first randomly displacing the Cartesian coordinates, with the maximum displacement fixed at about half the atomic spacing in the corresponding bulk ground state. The perturbed coordinates are quenched using a modified L-BFGS[5] algorithm,[6] as implemented in GMIN,[7] and then iteratively refined using a dynamic lattice search (described elsewhere[1]). For doped clusters each trial configuration was also subjected to systematic biminimisation,[8] yielding a permutational isomer (a homotop[9]) that is locally optimal with respect to atom-identity swaps. Since each swap (and each iteration of the dynamic lattice search) is accompanied by a quench, the average computational cost of locating a multiminimum for a cluster with one impurity atom is around N quenches. Our GBH runs were limited to between 10^6 and 10^7 quenches, depending on system size, and during each run a list of local minima (not just multiminima) was accumulated and sorted by potential energy. On rare occasions the L-BFGS algorithm converged to a stationary point that is not a local minimum (but a saddle point), and these configurations were removed during post-processing. Finally, the temperature parameter in the Metropolis criterion was adaptively adjusted to maintain the (local average) acceptance ratio close to 0.5, and each run was restarted from a random configuration whenever 50 consecutive trials in the Markov chain failed to produce a lower-energy multiminimum.

The database of minima for LJ_N (N=31,38,75) as well as $\mathrm{Ni}_{74}\mathrm{X}$ and $\mathrm{Au}_{54}\mathrm{X}$ (X = Au, Ag, Al, Cu, Ni, Pd, Pt, Pb) model clusters are included in this Supplementary Information (in the file `data.zip`). Each line of the `min.data` files (and the comment line for each frame in the XYZ files) contains the potential energy (U), the log-product of normal modes ($\ln \bar{\nu}^{\kappa}$), the point-group order, and in some cases also the determinant of the inertia tensor.

* bhusic@stanford.edu; Department of Chemistry, Stanford University, Stanford, CA 94305
† Dmitri.Schebarchov@gmail.com
‡ dw34@cam.ac.uk

[1] D. Schebarchov and D. J. Wales, Phys. Chem. Chem. Phys. **17**, 28331 (2015).
[2] Z. Li and H. A. Scheraga, Proc. Natl. Acad. Sci. USA **84**, 6611 (1987).
[3] D. J. Wales and J. P. K. Doye, J. Phys. Chem. A **101**, 5111 (1997).
[4] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. **21**, 1087 (1953).
[5] C. G. Broyden, J. Inst. Math. Appl. **6**, 76 (1970); R. Fletcher, Comput. J. **13**, 317 (1970); D. Goldfarb, Math. Comput. **24**, 23 (1970); D. F. Shanno, *ibid.* **24**, 647 (1970).
[6] J. Nocedal, Math. Comput. **35**, 773 (1980).
[7] D. J. Wales, "GMIN: A program for basin-hopping global optimisation, basin-sampling, and parallel tempering," `http://www-wales.ch.cam.ac.uk/GMIN/`.
[8] D. Schebarchov and D. J. Wales, Phys. Rev. Lett. **113**, 156102 (2014).
[9] J. Jellinek and E. Krissinel, Chem. Phys. Lett. **258**, 283 (1996).