Phase transitions and thermodynamic properties of dense assemblies of truncated nanocubes and cuboctahedra Supplementary Information

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This supplementary information contains additional details about Wang-Landau algorithm used in the present work, figures illustrating convergence of the algorithm and more snapshot images of the simulated systems.

In the original formulation of the Wang-Landau algorithm [1] it was suggested that a uniform histogram distribution should be attained at each iteration, and after that a new iteration begins with increment Δf (in terms of the present paper) scaled by factor $\alpha = 0.5$. Later it was shown that such a scheme may lead to convergence problem with increasing system size [2]. In paper [3] it was demonstrated that increase of the scaling factor to $\alpha = 0.7 - 0.9$, while making the initial stages of the process slower, allows to cover a substantially wider range of states having large entropy difference. In another work [4] it was argued that Wang-Landau algorithm attains optimal convergence if the WL increment Δf decreases continuously proportionally to inverse first power of the simulation time (or the number of MC steps): $\Delta f \sim 1/t$. In the present work we took into account our and others experience from the previous WL simulations.

At the first iterations of the WL procedure we typically used initial value of the increment $\Delta f = 0.1$ and the number of MC steps 100000. Also, we decreased the increment Δf by factor $\alpha = 0.7$ at each iteration which gives to the WL algorithm more time (comparing to value $\alpha = 0.5$ in the original formulation) to tune the weights f_i and discover more densely packed states. The number of MC steps was increased at each iteration as $1/\sqrt{\alpha}$, with an additional requirement that the system performs a full walk at least once between states of the highest and lowest density. We found that waiting too long for attaining a flat histogram at initial iterations (when increment Δf is not enough small) may distort the free energy profile so that it will be difficult to correct it at later iterations with lower value of increment Δf. A relaxed criteria, that the system just makes a full walk in between the earlier reached lowest and highest volumes turned out to give a more stable behavior of the algorithm. An example of tuning $f(V/n)$ dependence is shown in Fig. 1 SI. One can see that the free energy profile is essentially stabilized after 30 iterations, including the ordered region ($V/n < 1.5$ for this case). The values of f_i at this stage are however still noisy for numerical differentiation for computation of the pressure.

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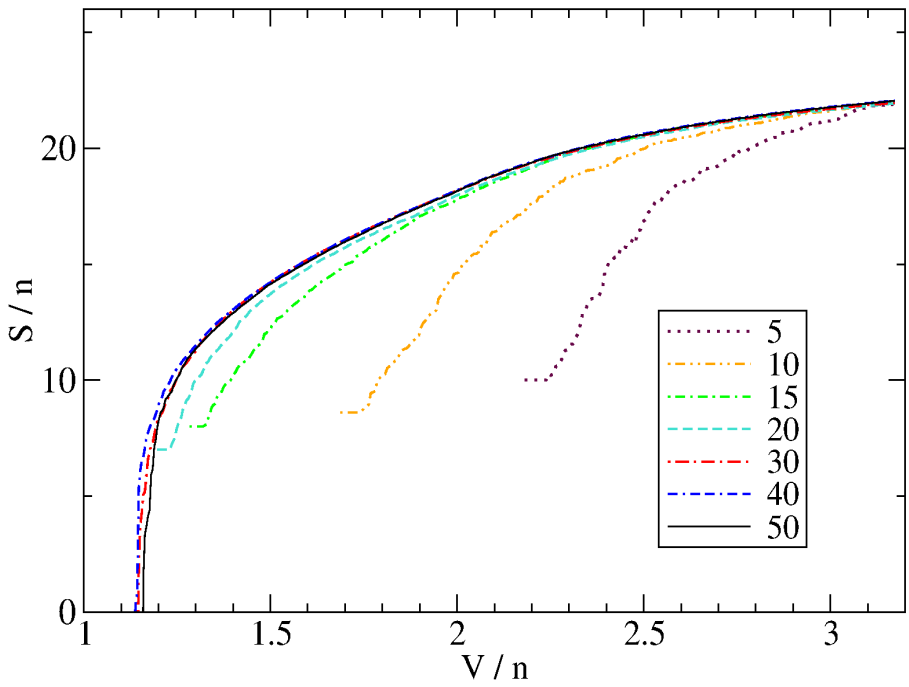

Figure 1: Accumulated entropy at the corresponding iteration of the WL procedure. Case $n = 64$ and $\tau = 0.6$. All the curves are brought to the same value of entropy at the maximum volume.

That is why we run additional 20 iterations decreasing the increment each time by factor $\alpha = 0.9$ and increasing the number of MC steps at each iteration by factor $1/\alpha$. Note that the later regime implements effectively the criteria suggested in paper [4], that the increment decreases proportionally to the inverse power of the MC steps made. Figure 1 SI demonstrates that the free energy profile remains stable at last 20 iterations.

Fig 2 SI shows how the system changes during the last five iterations; one can see that the system repeatedly visit both densely packed ordered states and low density liquid state (see example of a liquid state configuration in Fig. 3S h). The length of the last WL iteration was $4.8 \cdot 10^9$ MC steps.

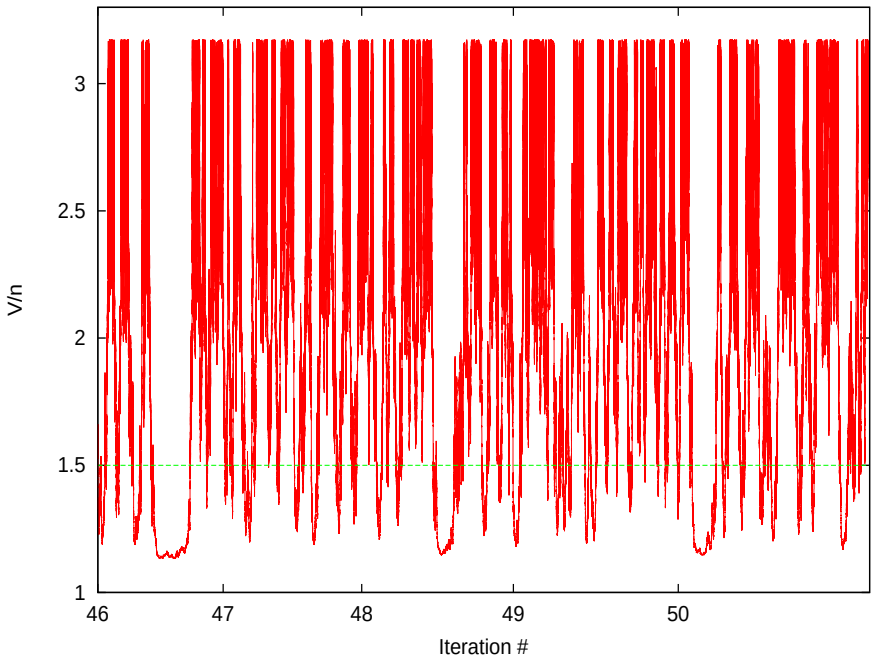

Figure 2: Example of system traveling between high and low density states during the five last iterations of the WL procedure. Case $n = 64$ and $\tau = 0.6$. The horizontal green line shows approximately the border of the ordered crystal phase.

References

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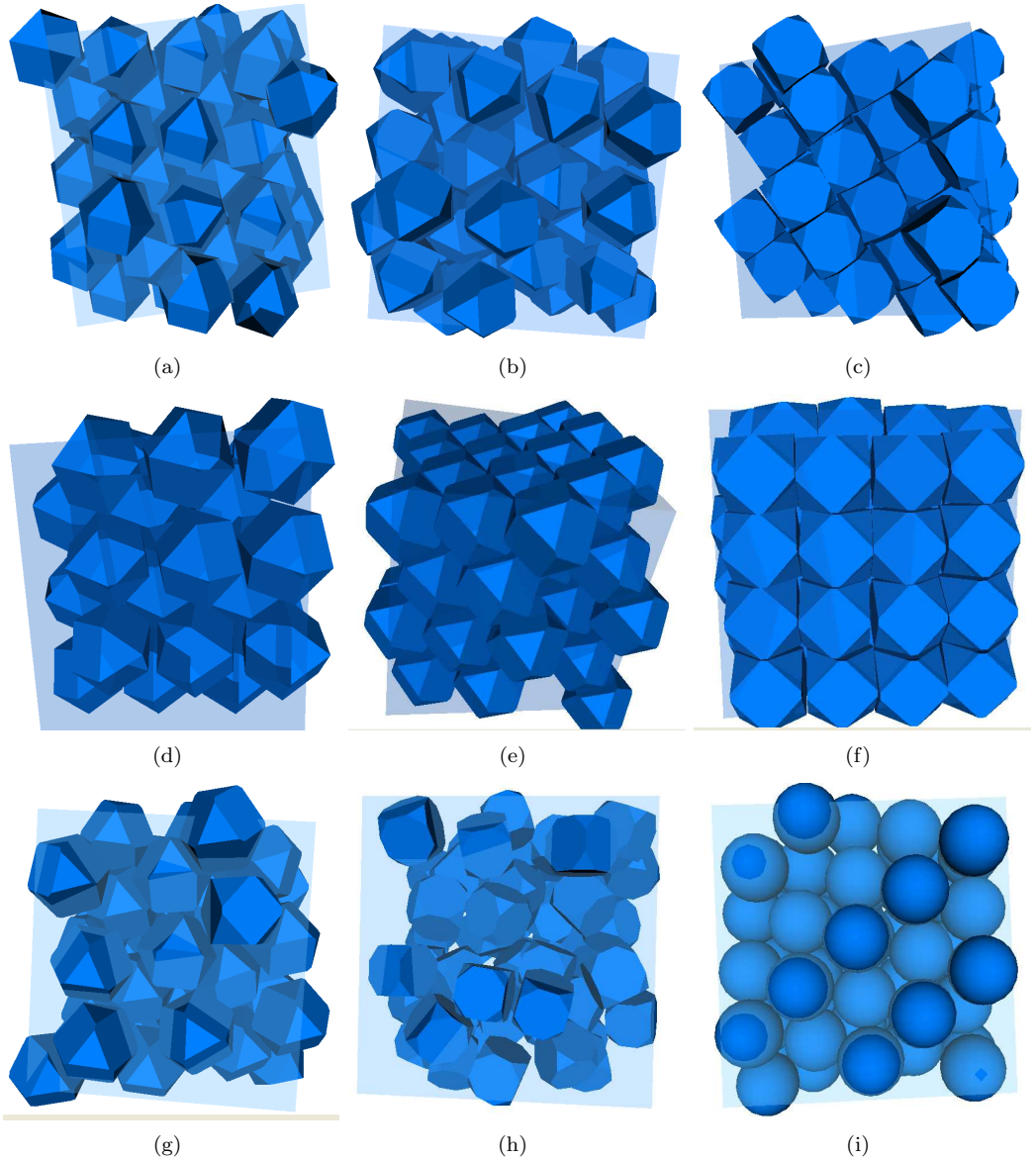

Figure 3: Examples of system configurations: a) $n = 90, \tau = 1, \rho = 0.88$ (dense packing); b) $n = 90, \tau = 0.9, \rho = 0.81$ (dense packing); c) $n = 90, \tau = 0.6, \rho = 0.81$ (dense packing); d) $n = 54, \tau = 1, \rho = 0.91$ (dense packing); e) $n = 64, \tau = 0.9, \rho = 0.89$ (dense rhombohedral-type packing); f) $n = 64, \tau = 0.9, \rho = 0.89$ (dense cubic-type packing; g) $n = 64, \tau = 0.9, \rho = 0.78$ (jammed liquid structure); h) $n = 64, \tau = 0.6, \rho = 0.32$ (low density unordered state); i) hard spheres, $n = 90, \rho = 0.9$ (dense packing).