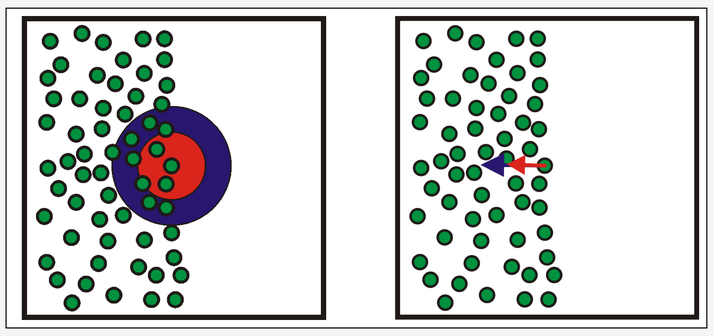
Multilevel Summation for Dispersion: A Linear-Time Algorithm for r^{-6} Potentials

Daniel Tameling[1,2,*] **Paul Springer**[1] **Paolo Bientinesi**[1] **Ahmed E. Ismail**[1,2]

[1] Graduate School AICES, RWTH Aachen University [2] Aachener Verfahrenstechnik (AVT), RWTH Aachen University [*] E-Mail: tameling@aices.rwth-aachen.de

Motivation

- Dispersion interactions are non-bonded pairwise interactions, which are proportional to r^{-6} as found in the Lennard-Jones and Buckingham potential
- For inhomogeneous systems, such as interfacial systems and systems with sharp concentration gradients, using a cutoff method is inaccurate



The use of different cutoffs for inhomogeneous systems can lead to significant differences in the forces

- Long-range methods represent dispersion interactions accurately
- Currently, the most popular long-range methods in molecular dynamics are the Ewald mesh methods, which have a complexity of $\mathcal{O}(N \log N)$ and do not scale well, because of their communication patterns
- The Multilevel Summation (MLS) is a linear method, which was recently developed, and we extend this method to dispersion interactions

The Multilevel Summation method for Dispersion

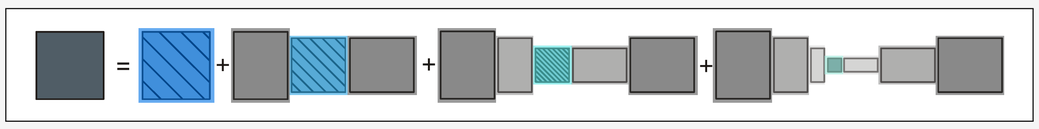
The dispersion potential can be written as

$$V = \xi^T \; \mathbf{G} \; \xi$$

e.g. $\xi_i = \sqrt{2}\sqrt{\varepsilon_{ii}}\sigma_{ii}^3$ (Lennard-Jones potential with geometric mixing)

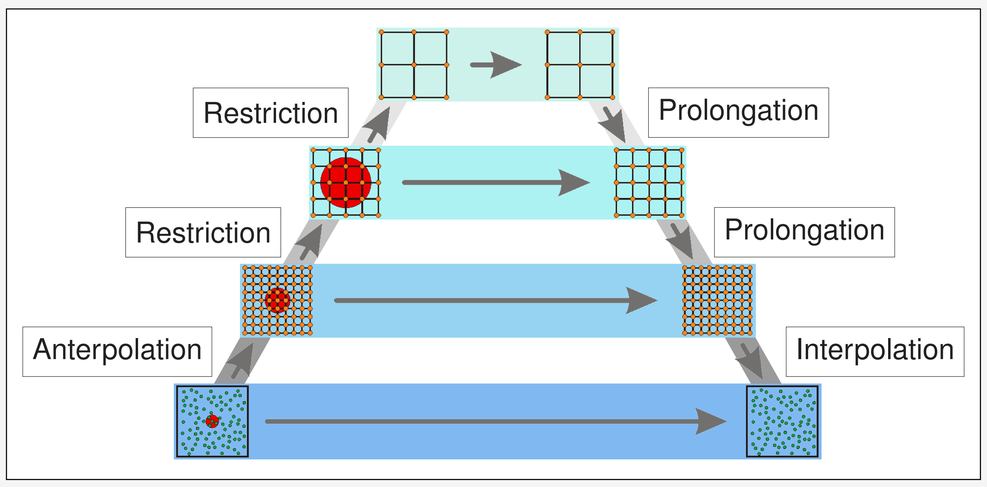
$G_{ij} = \frac{1}{r_{ij}^6}$ for $i \neq j$

The MLS algorithm improves the complexity to $\mathcal{O}(N)$ by splitting the dense matrix of the potential into a sequence of matrices, starting with a sparse one, and proceeding with increasingly denser ones that are, additionally, reduced in size, resulting in smaller matrices as the density increases. The smaller matrices yield through interpolation an approximation for their full-sized equivalents.



Multilevel Summation approximation of the dense matrix of the dispersion potential with 3 grid levels

Physically, the size reduction and interpolation of the matrices is an approximation of the potential on a hierarchy of grids, where the sparsity is induced by a growing cutoff, with exception of the last grid, where no cutoff can be applied.



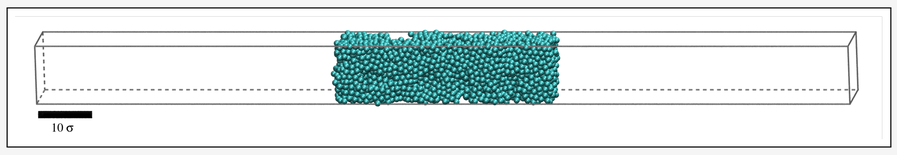
Schematic representation of one step of the Multilevel Summation algorithm with 3 grid levels

The general formula for approximating r^{-6} with l grid levels is

$$\frac{1}{r^6} \approx g_0 + \mathcal{I}_1 \left[g_1 + \mathcal{I}_2 \left[g_2 \ldots + \mathcal{I}_l \left[g_l \right] \ldots \right] \right]$$

Simulations

We created a prototype implementation of the MLS for dispersion. Our main test system was a Lennard-Jones gas consisting of 4000 particles in a slab of dimensions $11.01\sigma \times 11.01\sigma \times 176.16\sigma$ with geometric mixing and periodic boundary conditions.

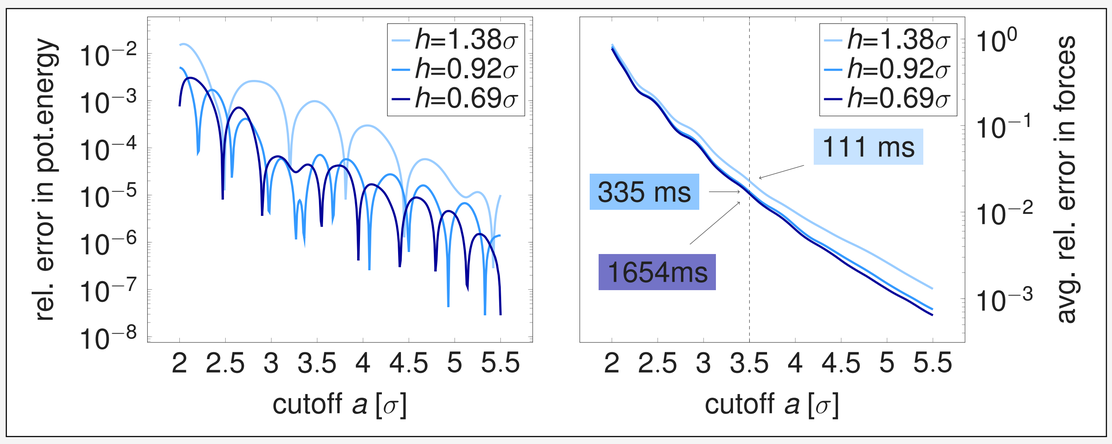


Snapshot of a simulation setup in a slab geometry with 4000 particles.

Moreover, in order to test the scalability of the algorithm, slab systems with 500, 32,000 and 256,000 particles were simulated, where the domain size was adjusted, while the aspect ratios were preserved.

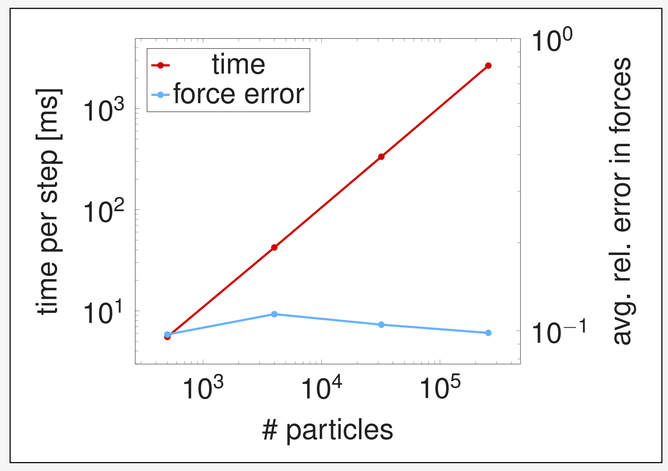
Numerical Results

The results of our MLS solver for dispersion are compared with a high-precision calculation done in LAMMPS. From a theoretical analysis we find that the accuracy is governed by the cutoff a and the finest grid spacing h; increasing the former and decreasing the latter produces more accurate results.



Error in the potential energy. Error in the forces.

A theoretical investigation of the influence of a and h on the performance of the algorithm shows that the run time of the grid-based part is proportional to a^3/h^6.
Finally, the serial prototype implementation scales linearly:



Total timings per step and error in forces for 500, 4000, 32000 and 256,000 particles.

Conclusions

- We have extended the Multilevel Summation method to dispersion interactions
- The accuracy of the Multilevel Summation is influenced by the cutoff a and the spacing of the finest grid h
- The run time of the grid-based part is proportional to a^3/h^6
- The linear complexity has been demonstrated with a serial, prototype implementation

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