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

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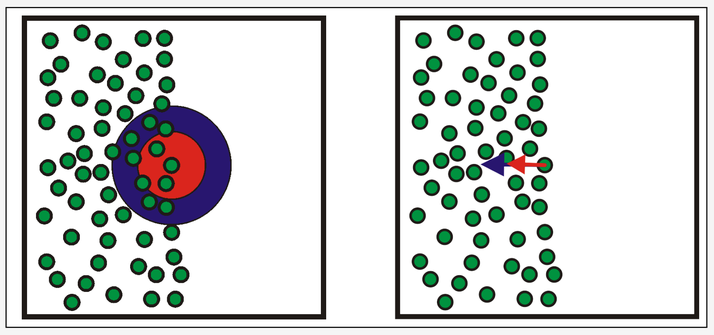
Motivation

- 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

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

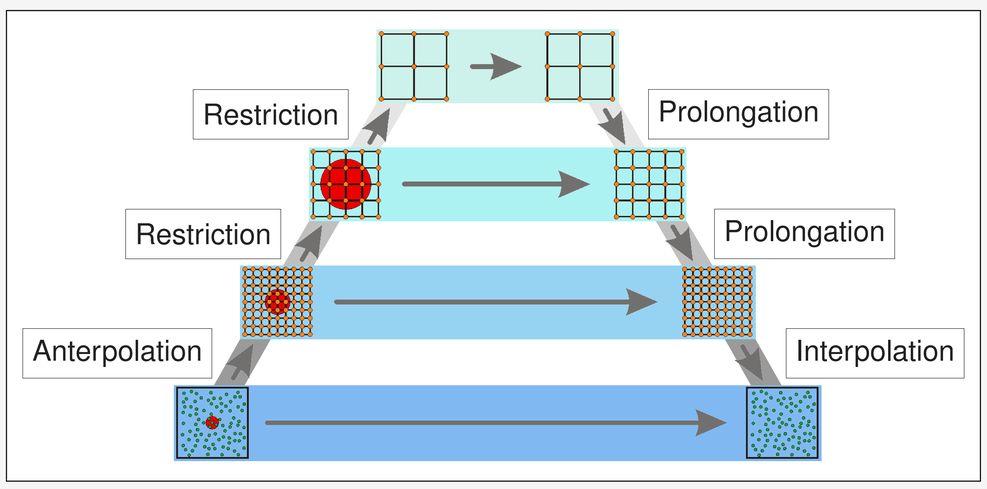
e.g. $\xi_i = \sqrt{2}\sqrt{\varepsilon_{ii}}\sigma_{ii}^3$ (Lennard-Jones potential with geometric mixing)

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

The MLS algorithm improves the complexity to $\mathcal{O}(N)$ by interpolating the potential on a hierarchy of increasingly sparse grids, where a growing cutoff is used on every grid except the last one.

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

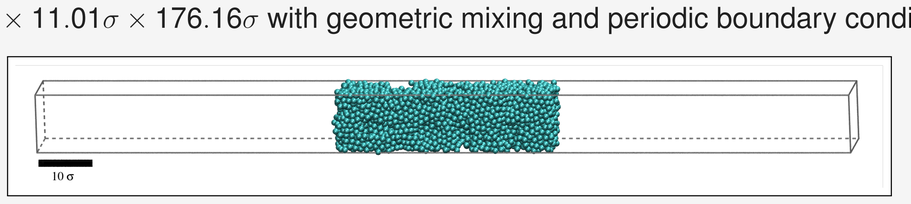
$$\frac{1}{r^6} \approx g_0 + \mathcal{I}_1 \left[g_1 + \mathcal{I}_2 \left[g_2 \ldots + \mathcal{I}_l \left[g_l \right] \ldots \right] \right]$$



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

Simulations

We created an implementation of the MLS for dispersion for LAMMPS. 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.

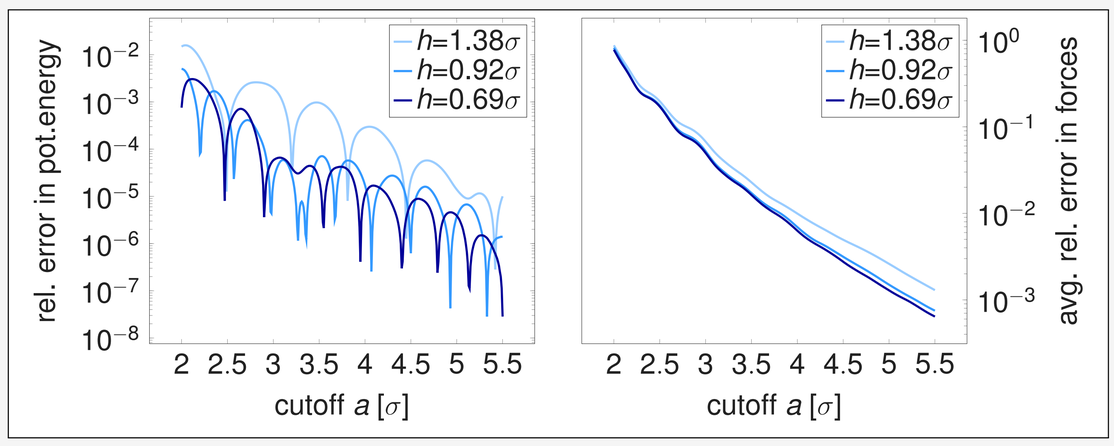


Snapshot of a simulation setup in a slab geometry with 4000 particles.

Moreover, to test the scalability of the algorithm, similar slab systems were simulated, where the domain size was adjusted, while the aspect ratios were preserved.

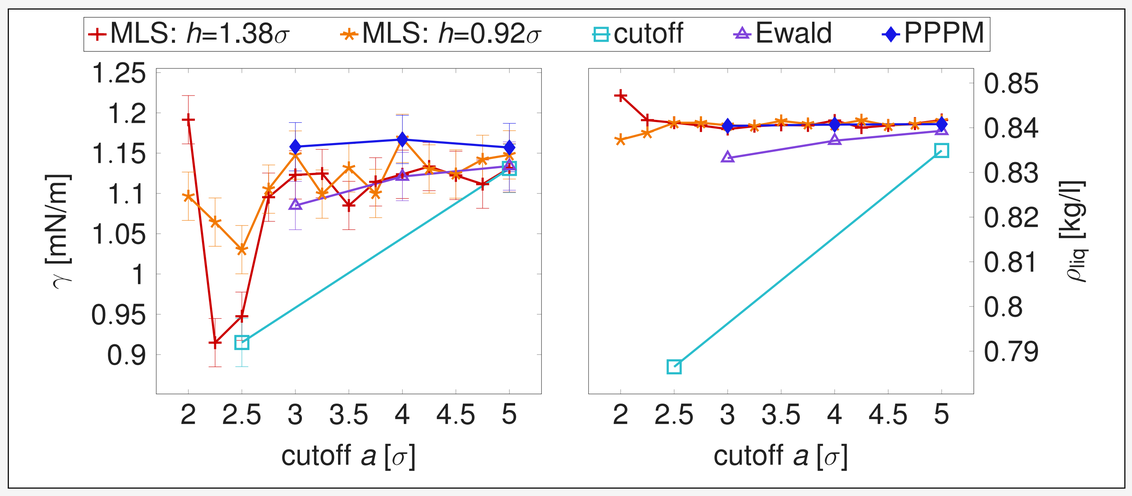
Numerical Results

The results of our MLS solver for dispersion are compared with a high-precision calculation. 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.

Results for the surface tension and the liquid density yield comparable results to other standard methods: (Results for cutoff-, Ewald-, and PPPM- method are from [2])

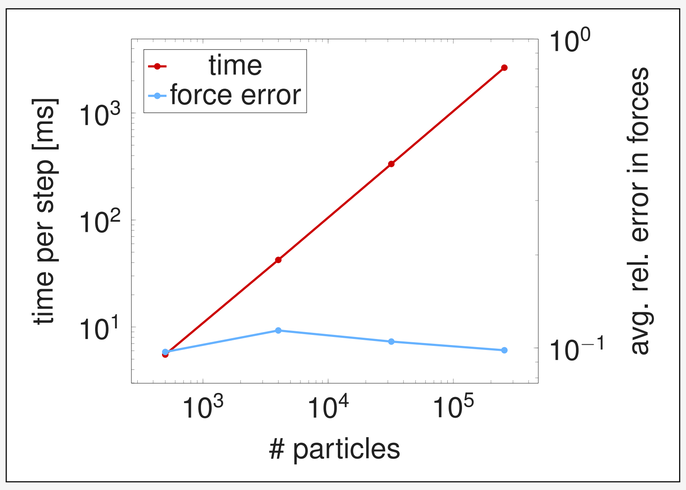


Surface tension. Liquid density.

(cutoff-method with tail correction)

Finally, we investigated the performance of the implementation:

MLS(h=1.38σ):	a=3σ	9.0 ms
MLS(h=0.92σ):	a=3σ	46.8 ms
Cutoff:	a=5σ	7.3 ms
Ewald:	a=4σ	37.7 ms
PPPM:	a=3σ	6.2 ms



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

Acknowledgements

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References

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