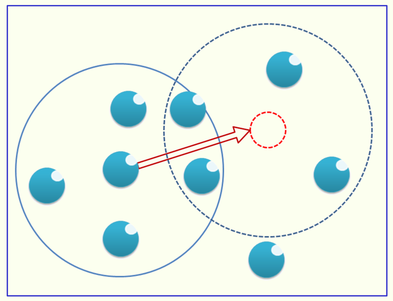
GPU-BASED MONTE CARLO SIMULATIONS FOR CANONICAL AND GIBBS ENSEMBLES

Eyad Hailat[a], Vincent Russo[a], Jason Mick[b], Kamel Rushaidat[a], Loren Schwiebert[a], and Jeffrey Potoff[b]

Departments of Computer Science[a] and Chemical Engineering[b], Wayne State University, Detroit, MI

Overview

Markov Chain Monte Carlo (MCMC) simulation of chemical systems allows examination of nanoscopic thermodynamics and associated behavior at small time scales. These simulations tend to be computationally expensive, requiring days or more of CPU time to collect data. In order to remedy the inherent time complexity of these simulations and allow for easier exploration of physical phenomena, optimization work is essential. NVIDIA's Compute Unified Device Architecture (CUDA) allows one to exploit the parallel nature of these scientific algorithms.



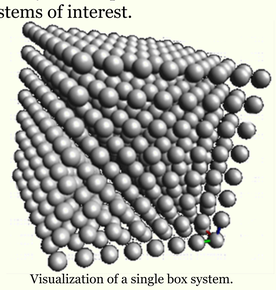
Motivation

In chemistry, computer simulations are considered a valid substitute to lab experiments to get information on the liquid state of material. However, there is a class of problems that cannot be simulated using current methodologies. These problems require an open system, which employs an algorithm that allows for fluctuation in the number of molecules in the system. For such systems, Monte Carlo methods must be used.

Typical applications of Monte Carlo simulation include:

- Prediction of physical properties and phase behavior
- Prediction of adsorption isotherms for gases in porous materials
- Simulation of biological systems at constant chemical potential
- Simulation of nanoparticles to stabilize drug dispersion

Our open source software provides an ensemble driven Monte Carlo engine for the simulation of chemical systems on the GPU. This software serves as a new tool in the field of Monte Carlo simulation of chemical systems to gain new insights into dynamic processes in nanoscopic and biological systems of interest.



Visualization of a single box system.

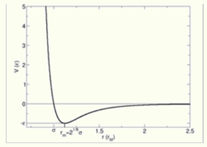
Canonical and Gibbs Ensembles

Monte Carlo simulations require one to select a particular type of statistical thermodynamic ensemble to simulate the system. Molecular systems consist of several important correlated variables – volume, temperature, number of particles, system energy, and pressure. For each ensemble, specific variables are fixed, and others remain independent.

The **canonical ensemble** is commonly employed to equilibrate and study the equilibrium structure and energy of small-molecular systems. Once equilibrated, the Widom insertion method allows for the direct calculation of chemical potential regions of system components.

The **Gibbs ensemble** offers a way to directly simulate coexisting phases and calculate phase equilibria properties. This ensemble consists of two simulation boxes, where the canonical is limited to one.

$$V_{LJ} = 4\varepsilon\left\{\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right\}$$



The Lennard-Jones algorithm aside a plot of energy vs. radial distance.

$$U^*(r_c) = \frac{8}{9}\pi\rho^*\epsilon\sigma^3\left[\left(\frac{\sigma}{r_c}\right)^9 - 3\left(\frac{\sigma}{r_c}\right)^3\right]$$

$$P^*(r_c) = \frac{32}{9}\pi\rho^{*2}\epsilon\sigma^3\left[\left(\frac{\sigma}{r_c}\right)^9 - \frac{3}{2}\left(\frac{\sigma}{r_c}\right)^3\right]$$

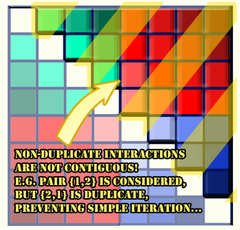
Long range corrections were applied in standard fashion for homogenous fluid.

Computational Methods

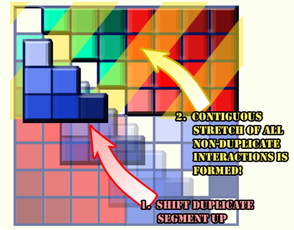
Remapping Pairwise Interactions

Volume transfer requires a complete recalculation of all unique particle pairwise interactions. Representing particles as a matrix creates a problem of having a non-contiguous block of interactions. To overcome this problem, a remapping of the matrix is made by shifting the lower triangle region up and remapping that part to create a contiguous block of unique particle interactions.

PROBLEM:
NON-CONTIGUOUS UNIQUE INTERACTIONS

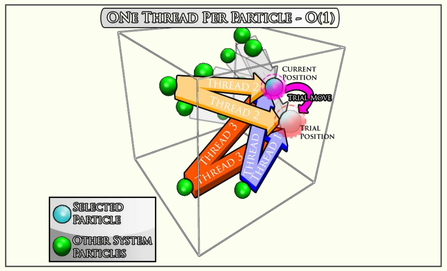


SOLUTION:
REMAPPING VIA SELECTIVE SHIFT OF DUPLICATE BOTTOM REGION



Multi-Threading Scheme

Each thread is devoted to one particle, excluding the selected particle. The thread calculates the change in the particle interaction with the trial particle moving from its current position to the trial position. The sum of all particles is then compiled.

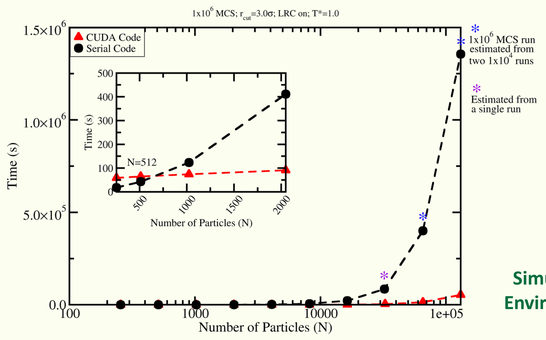


Performance Results

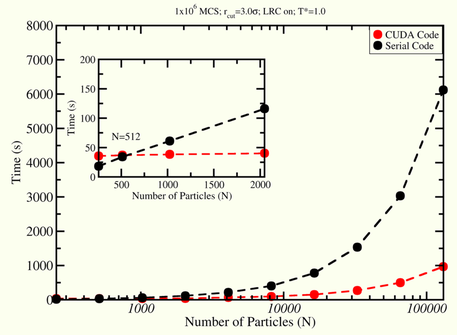
The results show performance gain in terms of **execution time** when using our CUDA implementation over the serial CPU implementation, especially on large sized systems with many particles. As the system size increases, the performance gap between the serial and parallel algorithms also increases. However, there is no speed up noticed when the system size is less than 128 particles for the NVT and for a system size of 512 particles in each box for the Gibbs ensemble, due to the parallel algorithm overhead and memory latency.

The Gibbs ensemble parallel algorithm gains **14.4** times speed up and the parallel Canonical ensemble gains **6.3** times speed up over the serial algorithms. The two figures show rapid increase in the serial algorithms execution times, while the parallel algorithms are doing much better.

Gibbs ensemble on the GPU



Canonical ensemble on the GPU



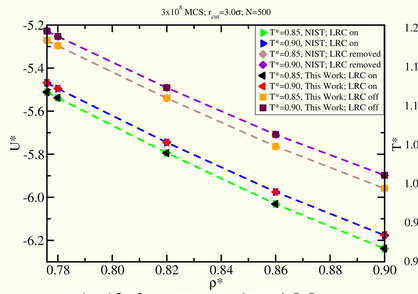
Simulation Environment

- NVIDIA GeForce GTX 560: 1.70 GHz engine clock speed.
- Intel Core i5-2500k CPU 3.30 GHz, 8 GB RAM
- CUDA version 4.0

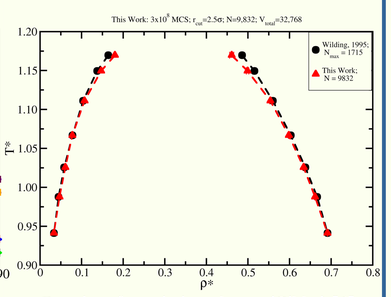
Code Validation

The engine code was compared with the literature [5-6].

- Canonical ensemble: evaluated liquid and gas phase reduced internal energies (U*) for various reduced densities (ρ*) and compared with data from NIST [5].
- Gibbs ensemble: evaluated vapor-liquid coexistence curve at different reduced temperatures (T*) and compared with histogram reweighted simulations of the grand canonical ensemble [6].
- The results showed good agreement with earlier work.
  - Slight deviation near the critical point.



Liquid Phase Energy (NVT) [5]



Density of coexisting phases (Gibbs) [6]

References

[1] J. Anderson, C. Lorenz, and A. Travvesset. "General purpose molecular dynamics simulations fully implemented on graphics processing units," *Journal of Computation Physics*, 227(10):5342-5359, (2008).
8[2] K. Binder, "Monte Carlo and molecular dynamics simulations in polymer science," *Oxford University Press*, USA, (1995).
[3] J.J Potoff and A.Z. Panagiotopoulos, "Surface tension of the three-dimensional Lennard-Jones fluid from histogram-reweighting Monte Carlo simulations," *Journal of Chemical Physics*, 112(14):6411-6415 (2000).
[4] D. Frenkel and B. Smit "Understanding molecular simulation: from algorithms to applications," volume 1, *Academic press*, (2002).
[5] *Benchmark Results for the Lennard-Jones Fluid*. Available from: http://www.cstl.nist.gov/srs/LJ_PURE/index.htm.
[6] Wilding, "Critical-Point and Coexistence-Curve Properties of the Lennard-Jones Fluid - A Finite-Size-Scaling Study," Physical Reviews E, 52(1):602, (1995).

Acknowledgments

This work has been supported in part by Wayne State University's Research Enhancement Program (REP) and National Science Foundation grant NSF CBET - 0730768. We also thank NVIDIA for the generous donation of some of the graphics cards used in this study.

Conclusion

- We have demonstrated the application of CUDA and GPUs to simulations in two ensembles, which show a "break even" point of around 500 particles.
- The effectiveness of the GPU was shown to improve with increasing system size.
- Gibbs ensemble Monte Carlo appears to be particularly well suited to the GPU, as the GPU provides a means to flatten the volume move, which scales as $O(N^2)$.