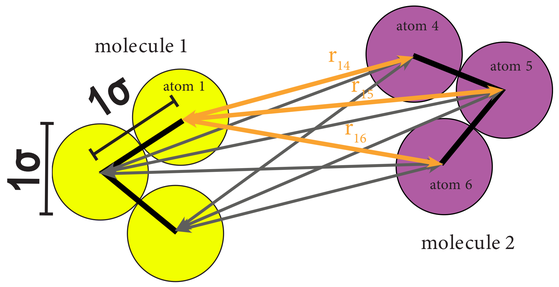
6-fold orientational ordering of supercooled liquids in two dimensions

Andrew Ton and Richard M. Stratt
Brown University, Department of Chemistry and Department of Physics
andrew_ton@brown.edu, richard_stratt@brown.edu
Providence, RI 02912

Abstract

Supercooled liquids are liquid systems cooled below their freezing temperature. We demonstrate a robust supercooled regime in simulations of two-dimensional one-component molecular liquids. We discover that in the supercooled liquid-glass regime, molecules achieve a six-fold orientational symmetry.

Molecular Dynamics of Soft Disk Molecules



Molecular dynamics is a computer simulation technique to model physical systems. We solved a classical many-body problem by numerical integration of Newton's equations. In our model, molecules were composed of three rigid sites which interact via Lennard-Jones potentials. Simulations were carried out in the NVE ensemble.

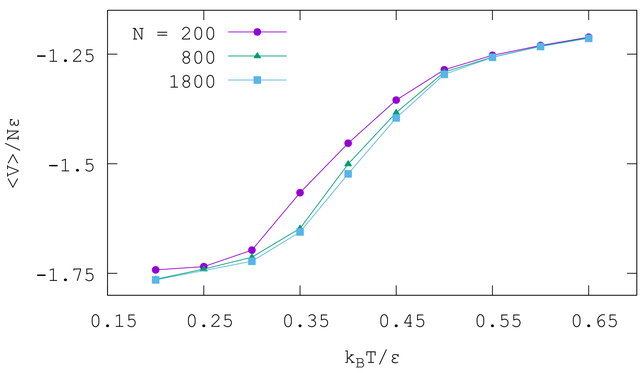


Figure: Potential energy per particle as a function of temperature plotted for various system sizes N. Each energy is an average over $3x10^5$ configurations.

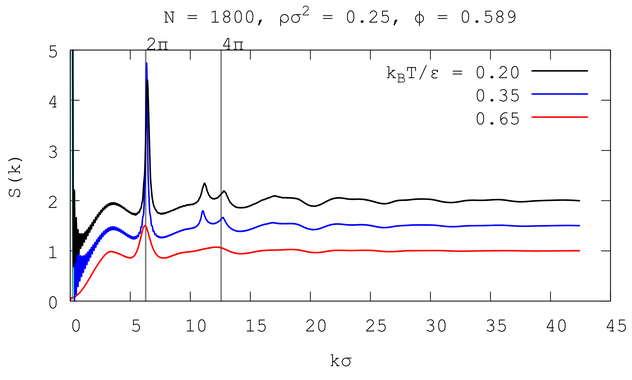
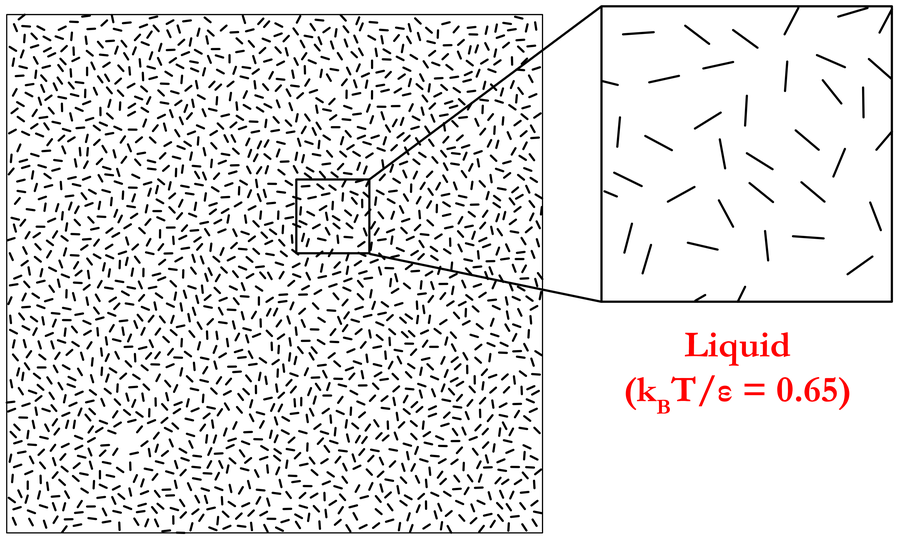
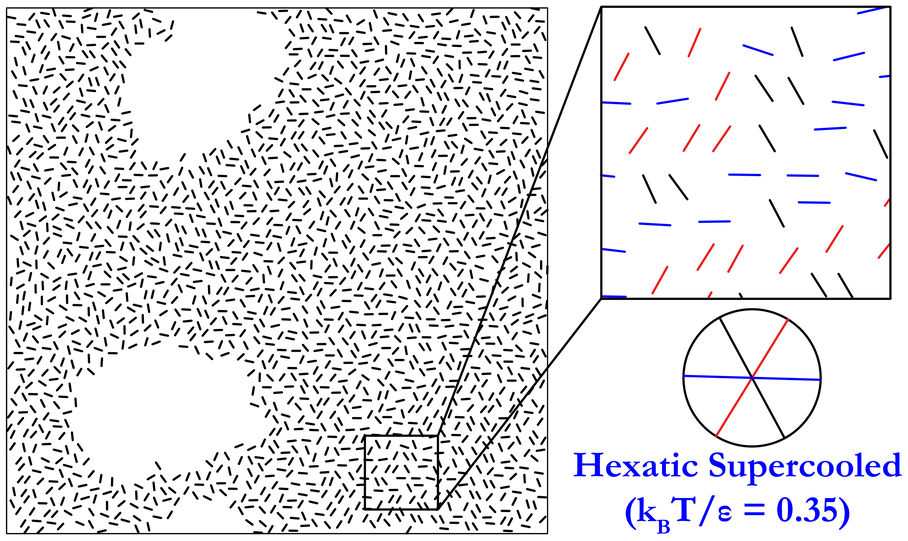
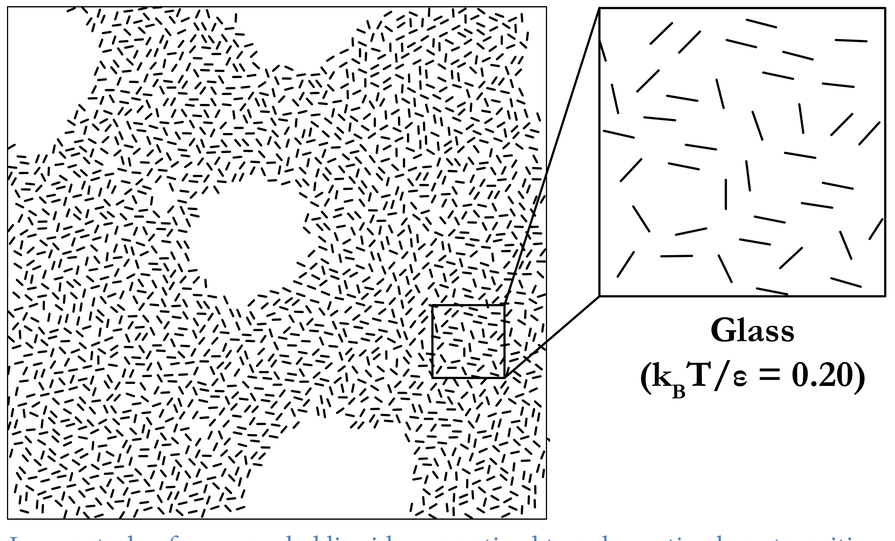


Figure: Static structure factor S(k) via Fourier transform of the pair correlation function $g_2(r)$. Curves are averages over $3x10^5$ configurations.

A Different Kind of Hexatic







In our study of supercooled liquids, we noticed two dramatic phase transitions. The first is a violent loss of spatial homogeneity known as the cavitation transition. The other has not been seen before, and we are calling it the hexatic phase (reminiscent of nematic order in liquid crystals). Here, there are six preferred directions for molecular orientations. This hexatic orientational order appears to be long range while translational order is not.

Order Parameter s_6

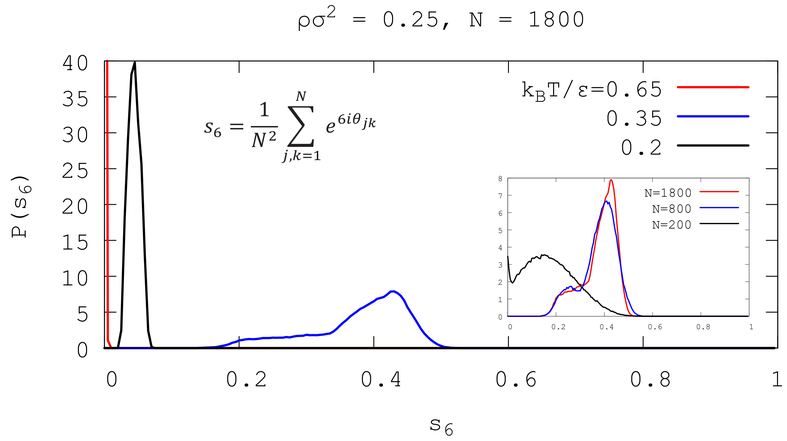


Figure: The hexatic orientational order parameter s_6 describes the degree of six-fold symmetry in molecular orientations. θ_{jk} is the angle between orientation vectors of molecules j and k. Probability distributions of s_6 are averaged over $3x10^5$ configurations. Inset: Hexatic order parameter at constant temperature and density for different system sizes N=1800, 800, 200.

Two Step Relaxation (Caging)

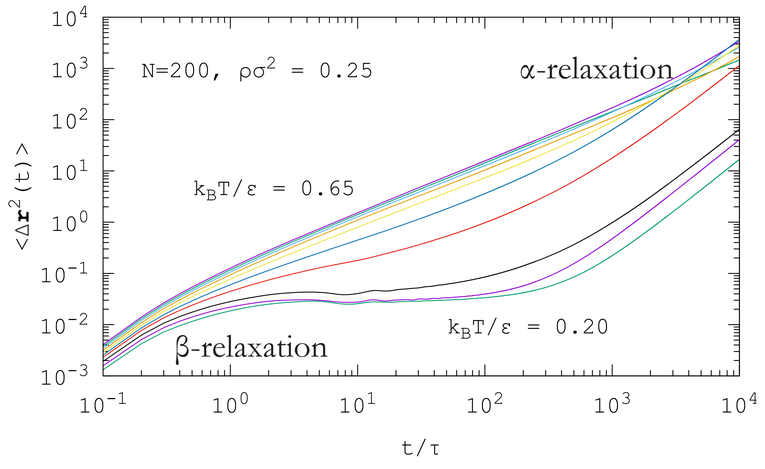


Figure: Mean squared displacement as a function of time plotted for a range of temperatures. τ is on the order of picoseconds (10^{-12} seconds). Averaging is over $2x10^5$ trajectories of N=200 molecules.

Open Questions

Finding a hexatic orientational phase leaves us with a lot of questions. What are its properties, and can we find other structural and dynamical signatures of this phase? Do we have some degree of underlying quasicrystalline structure? Finally, does this orientational symmetry in 2-d supercooled liquids admit any analogs in 3-d supercooled liquids?

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