Pseudospinodal interpretation of the glass transition

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Viewgraphs posted at
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Near-mean-field interpretation of glass transition

Glass transition in Lennard-Jones type glasses corresponds to a liquid-solid *pseudospinodal*.

1. In the mean-field limit, the liquid-solid instability is well defined and is a critical point. For near-mean-field systems, the instability is broadened and divergences are replaced by apparent divergences.

2. Apparent divergence of static structure function, $S(k_0 \neq 0)$.
   - In mean-field limit $S(k_0) \sim \epsilon^{-\gamma}$ with $\gamma = 1$ and $\epsilon = (T - T_s)/T_s$.

- In near-mean-field systems, the divergence is suppressed:

$$S(k_0) \sim \epsilon^{-\tilde{\gamma}}$$

  $\tilde{\gamma} = 1$ in $d = 1$, $\tilde{\gamma} = 1/2$ in $d = 2$, and $\tilde{\gamma} = 0$ in $d = 3$.

3. Growing length scale.

4. Scaling of cluster size distribution, n_s.

   - $n_s \sim s^{-3/2}$, mean-field critical fluctuations near the pseudospinodal.

   - $n_s \sim s^{-2}$, frustrated spinodal nucleation closer to pseudospinodal.

Molecular dynamics simulations of 3D, two-component Lennard-Jones system

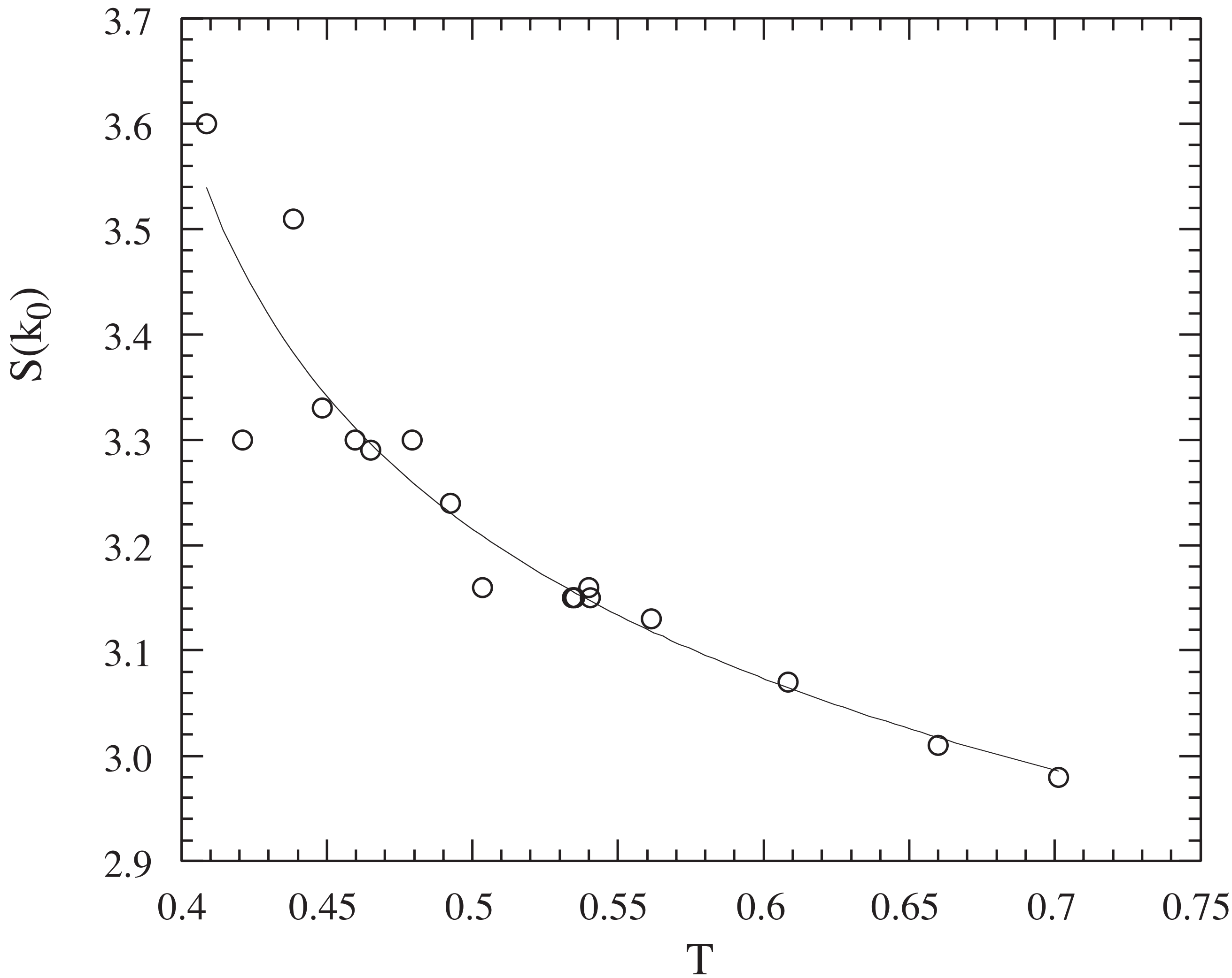
Kob-Andersen potential, model glass former

$$\begin{aligned} \rho &= 1.2 \\ N &= 8000 \\ \Delta t &= 0.005 \\ \text{equilibration time} &= 50\,000\tau \\ \text{length of runs} &= 50\,000\tau \end{aligned}$$

Results for D same as Kob-Andersen.

$T_{\text{mc}} = 0.435$, mode coupling temperature

$\mathbf{S(k_0)}$



$$S(k_0) \sim (T - 0.35)^{-0.08}$$

$T_{\mathrm{mc}} = 0.435$ mode coupling temperature

Consistent with predictions of near-mean-field theory.

Clusters

Theory predicts correlated clusters near liquid-solid pseudospinodal in analogy to their existence near Ising critical point and spinodal.

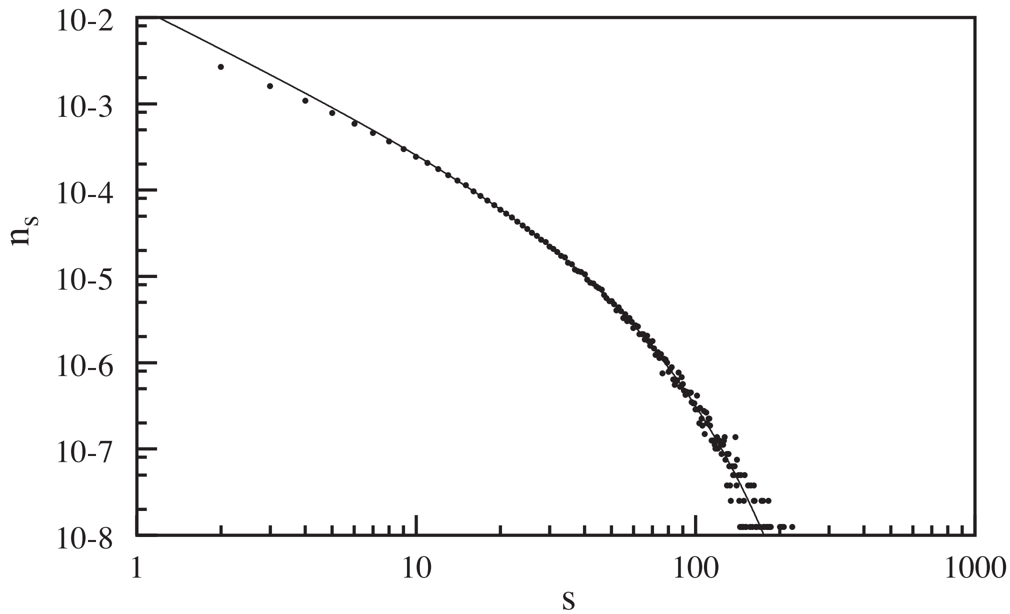
1. Look for groups of four particles that form regular tetrahedra.

2. Particles that are members of a minimum number of regular tetrahedra are “solid-like.”

3. Solid-like particles that are nearest neighbors are part of the same cluster.

Cluster Size Distribution

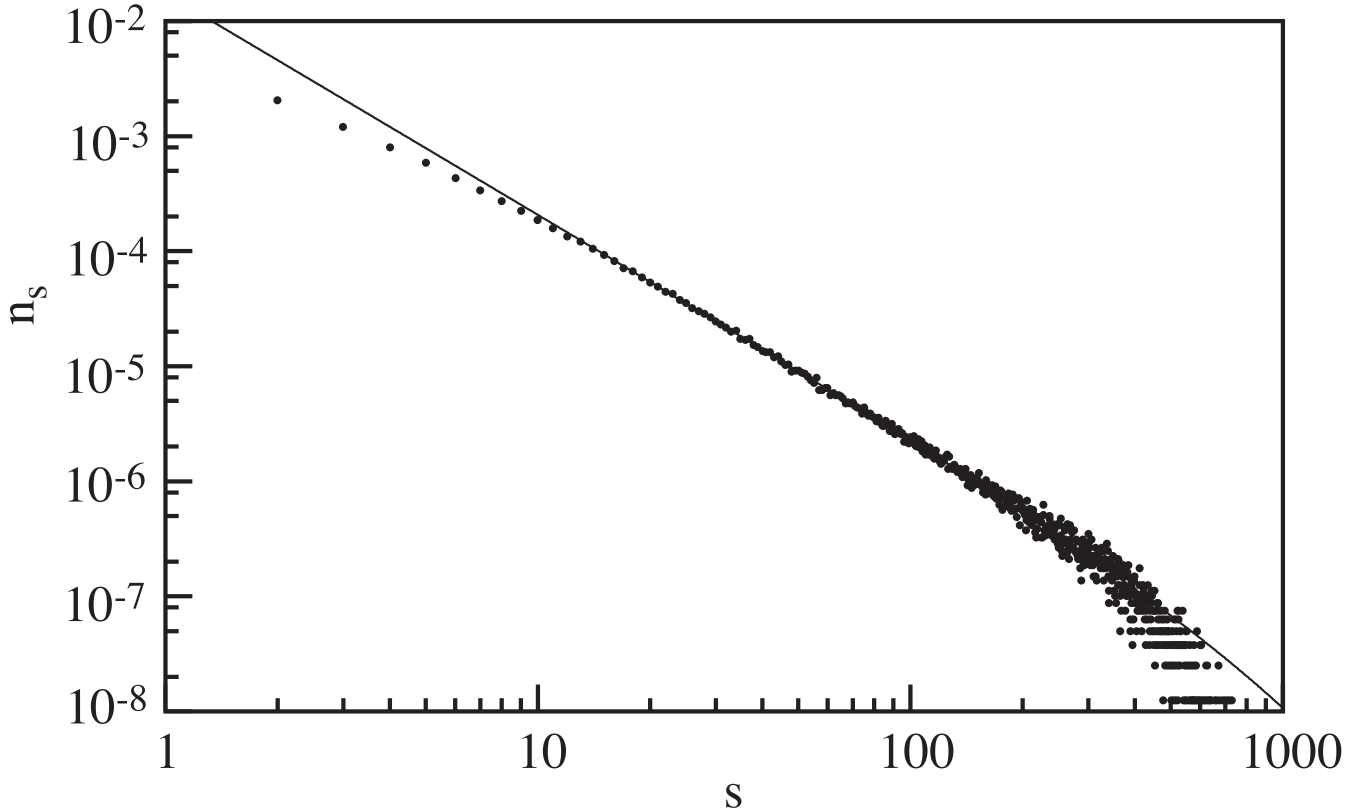
$$T = 0.60$$



$$n_s = 0.014 s^{-1.59} e^{-s/29.7}$$

Cluster Size Distribution

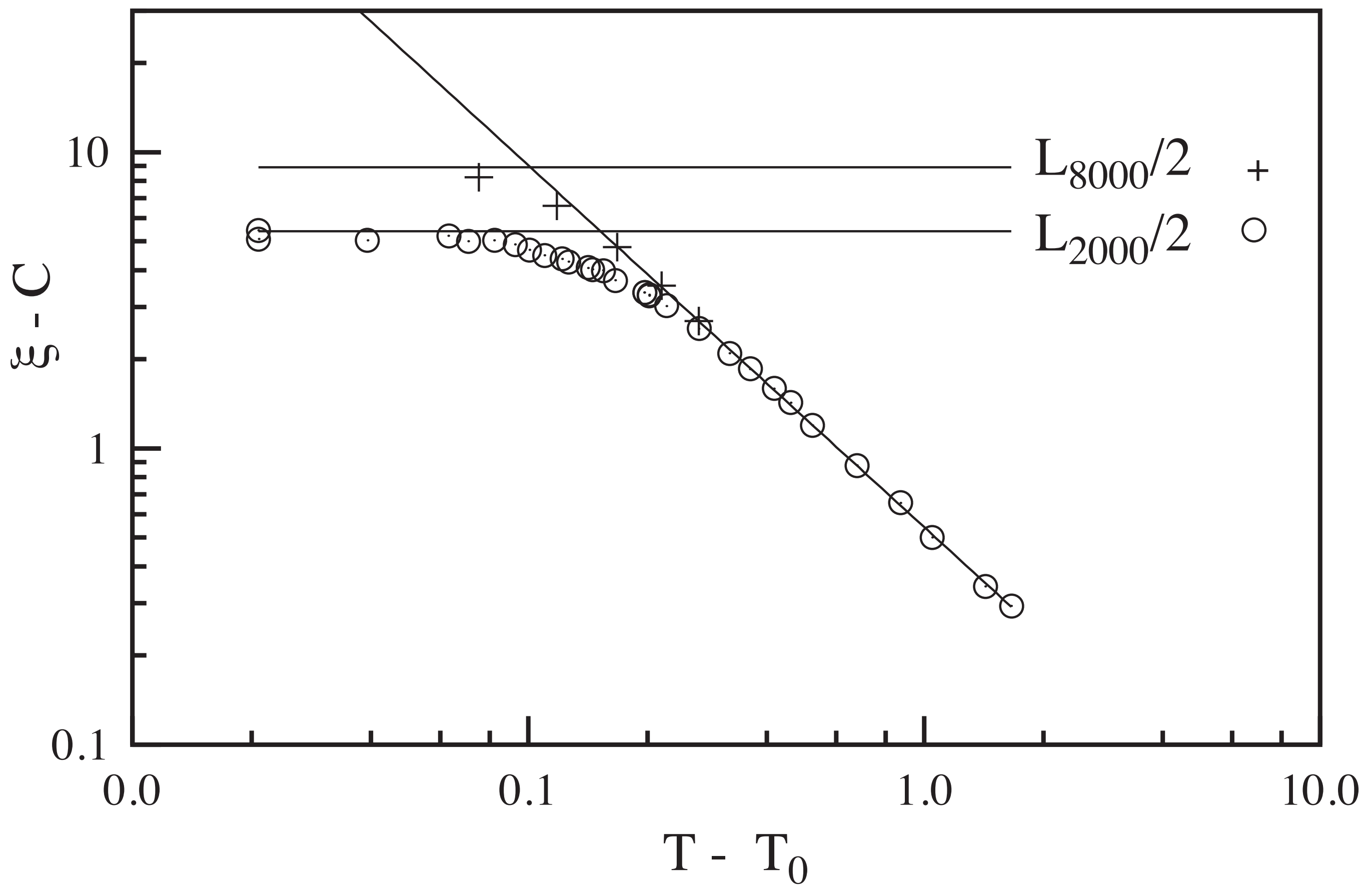
$T = 0.458$



$$n_s = 0.017 s^{-1.91} e^{-s/960}$$

A simple power law $n_s = As^{-x}$ fits well over the range $20 \leq s \leq 300$ with $A = 0.019$ and $x = 1.96$.

Increasing Length



The connectedness length ξ is plotted so that $\xi = A(T-T_0)^{-\tilde{\nu}}+C$ appears as a straight line. $A = 0.54$, $T_0 = 0.34$, $\tilde{\nu} = 1.2$, and $C = 0.53$. The horizontal lines are at half the system size $L/2$ for $N = 2000$ and 8000. For $N = 2000$ finite-size effects appear at $T = 0.60$. For $N = 8000$, finite-size effects appear at $T = 0.458$.

Summary

- Glass transition associated with a liquid-solid pseudospinodal.
- Growing length associated with clusters.
- Why is the effective interaction in dense liquids effectively long-range?
- Other evidence for influence of pseudospinodal in Lennard-Jones systems: nucleation for deep quenchs and in spinodal decomposition.
- There is probably no universal mechanism for the glass transition.

Related Publications

H. Gould, G. Johnson, H. Wang, and W. Klein, "Near-mean-field effects in a three-dimensional model glass former," in preparation.

W. Klein, H. Gould, J. Tobochnik, F. Alexander, M. Anghel, and G. Johnson, "Clusters and fluctuations at mean-field critical points and spinodals," Phys. Rev. Lett. **85**, 1270–1273 (2000)

N. Gulbahce, H. Gould, and W. Klein, "Zeros of the partition function and pseudospinodals in long-range Ising models," cond-mat/0308418 and Phys. Rev. E.

G. Johnson, A. I. Mel'cuk, H. Gould, W. Klein, and R. Mountain, "Molecular-dynamics study of long-lived structures in a fragile glass-forming liquid," Phys. Rev. E **57**, 5707–5718 (1998).

W. Klein, "Fractals and multifractals in early-stage spinodal decomposition and continuous ordering," Phys. Rev. Lett. **65**, 1462-1465 (1990)

J. Yang, H. Gould, W. Klein, and R. D. Mountain, "Molecular dynamics study of supercooled liquids," J. Chem. Phys. **93**, 711 (1990).

F. J. Cherne, M. I. Baskes, R. B. Schwarz, S. G. Srinivasan, and W. Klein, "Non-clasical nucleation in supercooled Nickel," cond-mat/0302491