Segregation, jamming, and glass-forming ability of binary hard spheres

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NSF MRSEC DMR-1119826

NIH-GM065835


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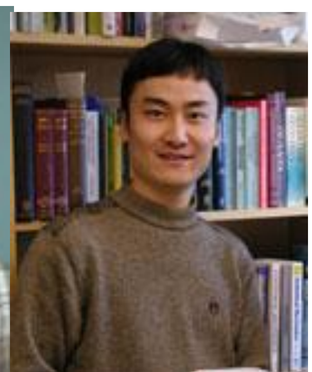
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Christine Jacobs-Wagner


Bradley Parry


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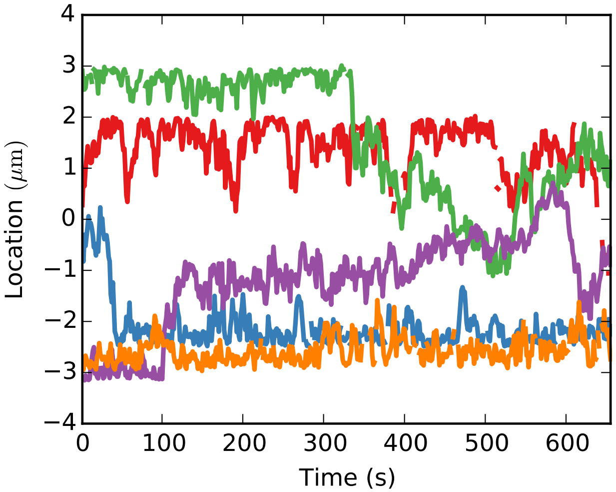
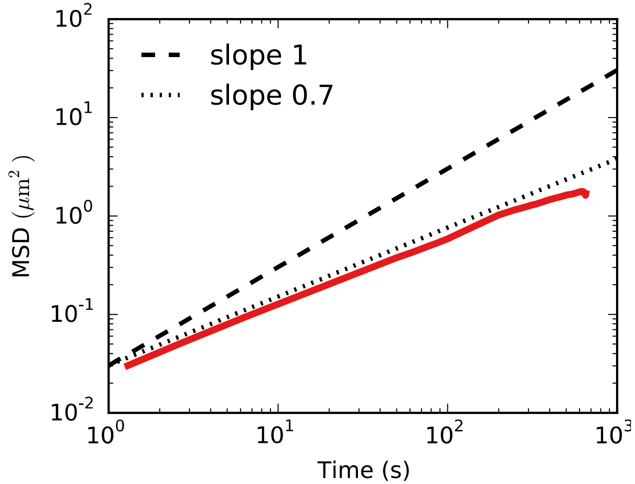
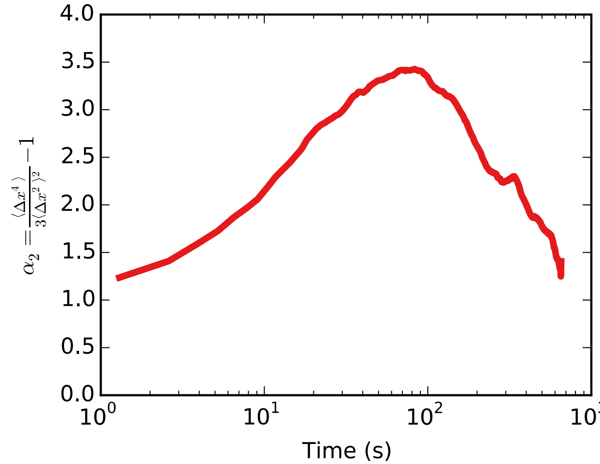
Mystery `Glassy' Dynamics



Subdiffusive behavior



Non-Gaussian parameter ≈3.5



What types of models capture subdiffusive motion and large α_2 ≈3.5?
How do we discriminate among models?

B. R. Parry, I. V. Surovtsev, M. T. Cabeen, CSO, E. R. Dufresne, C. Jacobs-Wagner, Cell 156 (2014) 1.

Confocal imaging of colloidal hard spheres

E. R. Weeks, J. C. Crocker, A. C. Levitt, A. Schofield, and D. A. Weitz, *Science* **287** (2000) 628.

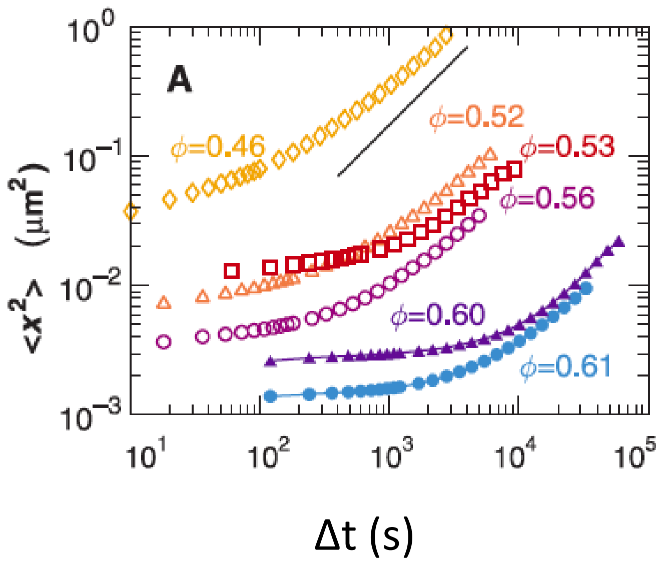


Fig. 1. Relaxation behavior. (**A**) Mean square displacement $\langle \Delta x^2(\Delta t) \rangle$ for several volume fractions ϕ. Open symbols are "supercooled fluids," which form crystals after a few hours (except for $\phi = 0.46$, which remains a fluid). Closed symbols are "glasses," which do not form crystals even after several weeks. Although the particles are tracked in three dimensions, only the one-dimensional $\langle \Delta x^2 \rangle$ is shown because the z resolution is poorer. The straight line shows a slope of 1. There is inherent uncertainty in these data due to the difficulty in averaging over the temporally and spatially inhomogeneous relaxation processes; see, for example, the data for $\phi = 0.53$. (**B**)

Plateau in MSD; Cage size ≈ $<\Delta x^2>^{1/2}$; diffusive at long lag times, $\Delta t > \tau_r$

Cages and Jumps

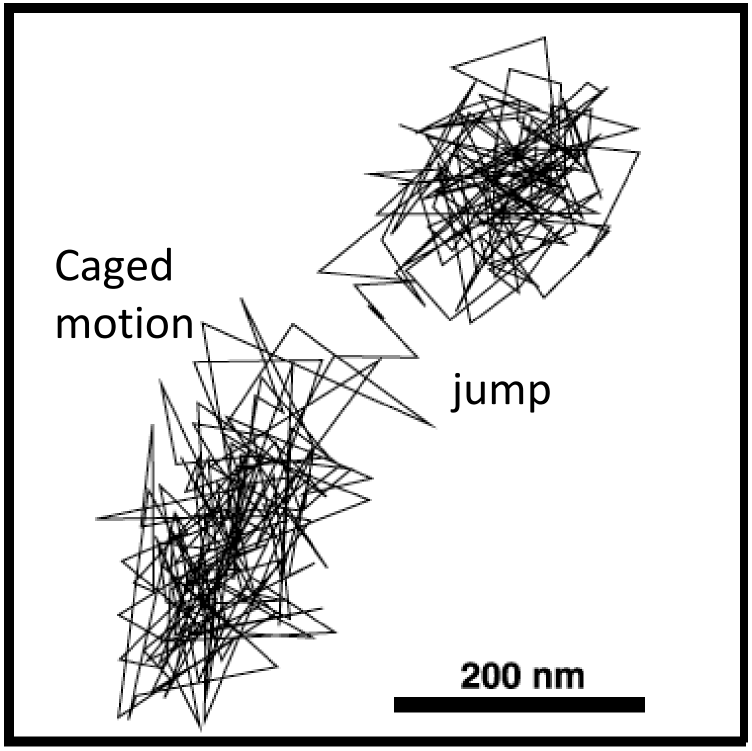
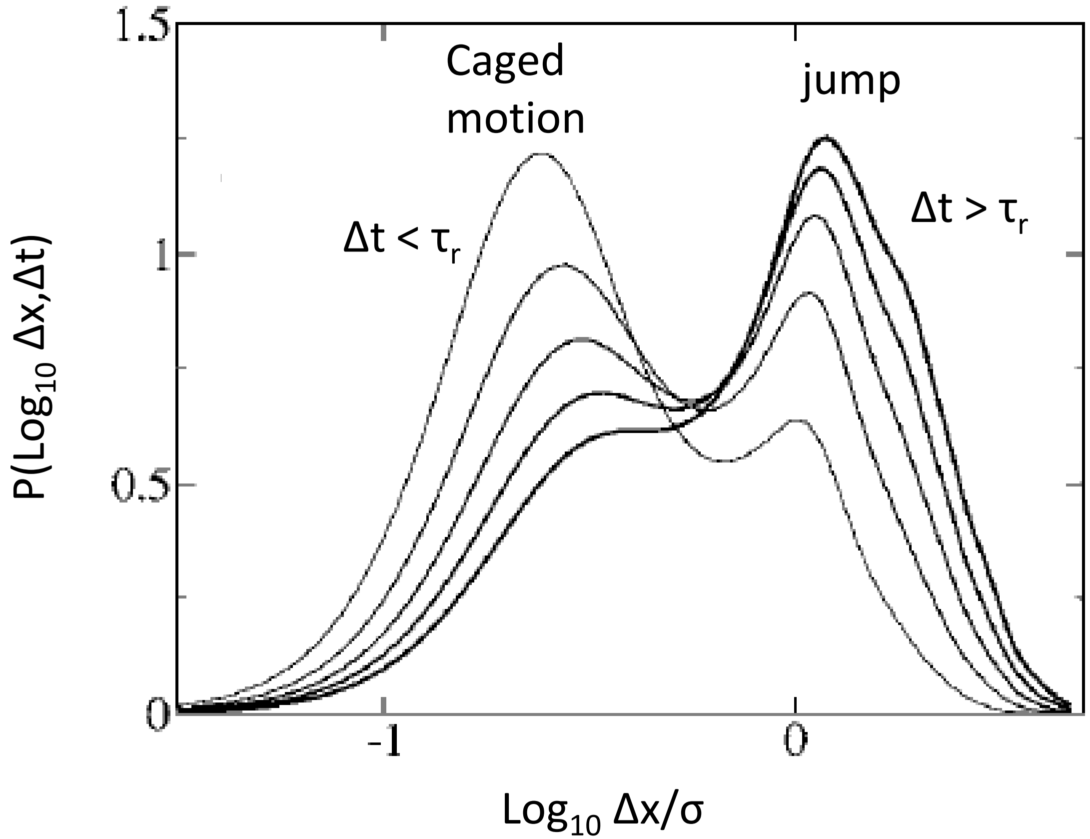


Fig. 2. A typical trajectory for 100 min for $\phi = 0.56$. Particles spent most of their time confined in cages formed by their neighbors and moved significant distances only during quick, rare cage rearrangements. The particle shown took ~500 s to shift position. The particle was tracked in 3D; the 2D projection is shown.

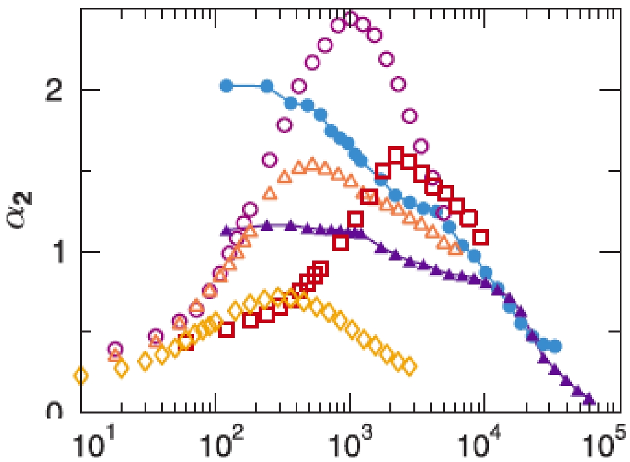
Displacement Probability Distribution



E. Flenner and G. Szamel, PRE 72 (2005) 011205

Non-Gaussian Parameter

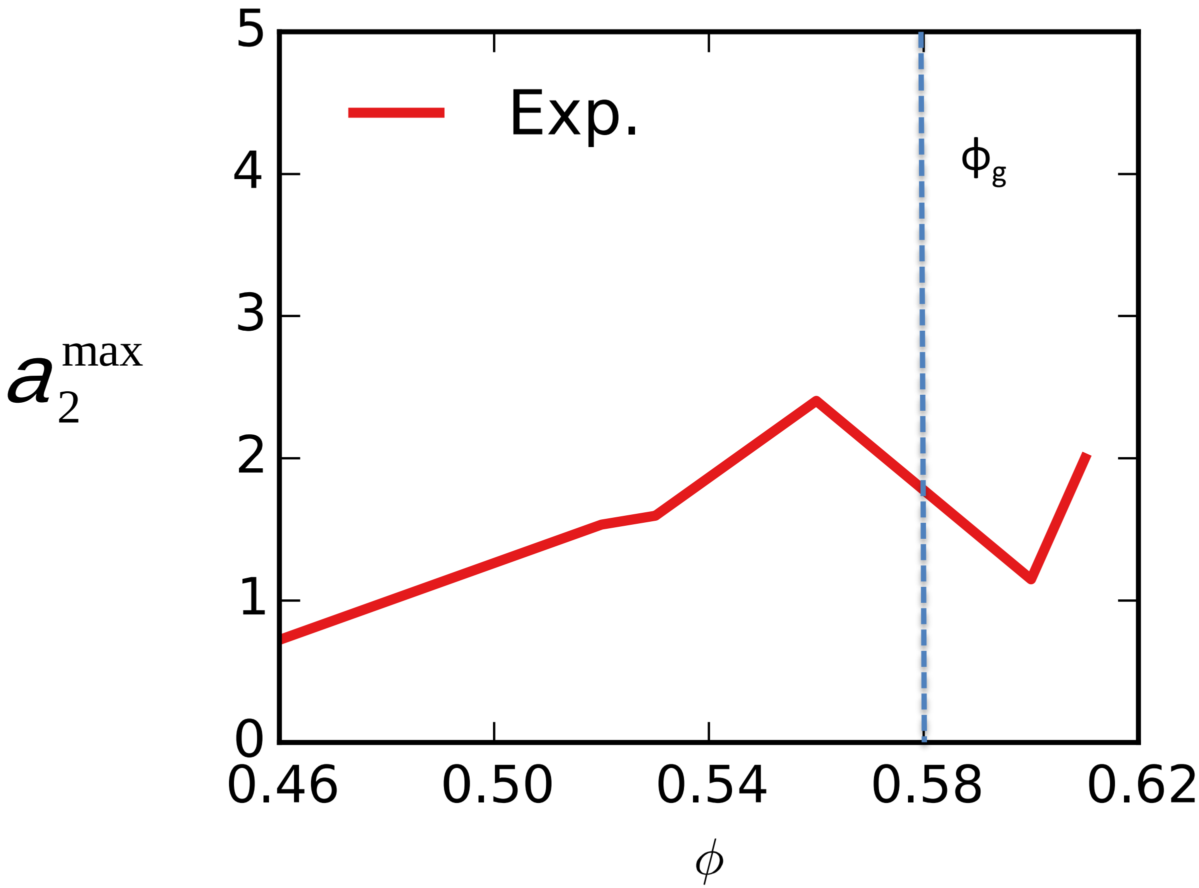
$\alpha_2 < 2.5$



Δt (s)

$$\alpha_2(\Delta t) = \frac{\left\langle (\Delta x)^4 \right\rangle}{3\left\langle (\Delta x)^2 \right\rangle^2} - 1$$

ϕ=0.46, 0.52, 0.53, 0.56, 0.60, 0.61

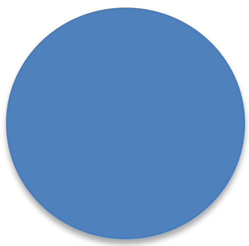


Exp.
ϕg
a2max
ϕ
0.46
0.50
0.54
0.58
0.62
0
1
2
3
4
5

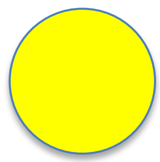
What is the best glass-forming bidisperse hard sphere system?

$x_S=N_L/N_S$

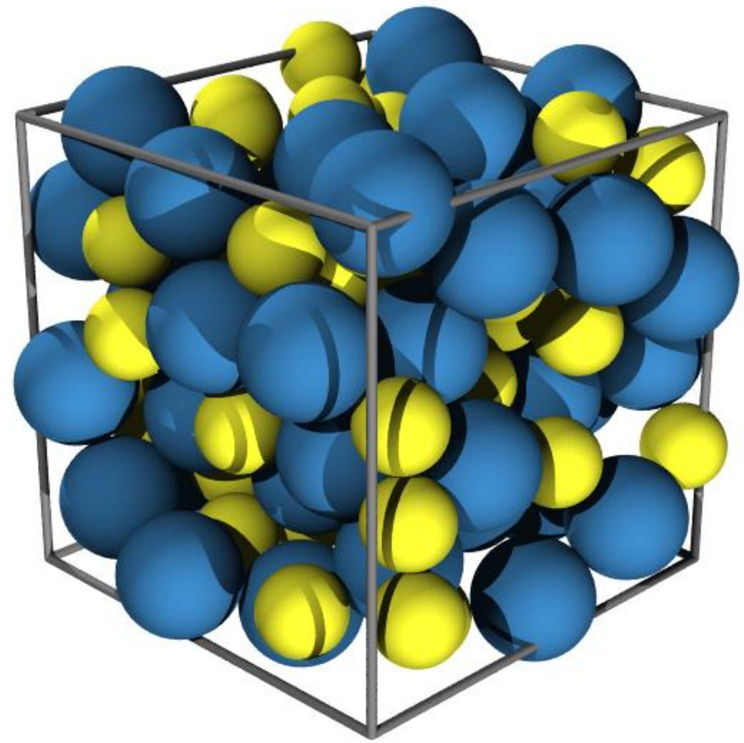
σ_S/σ_L



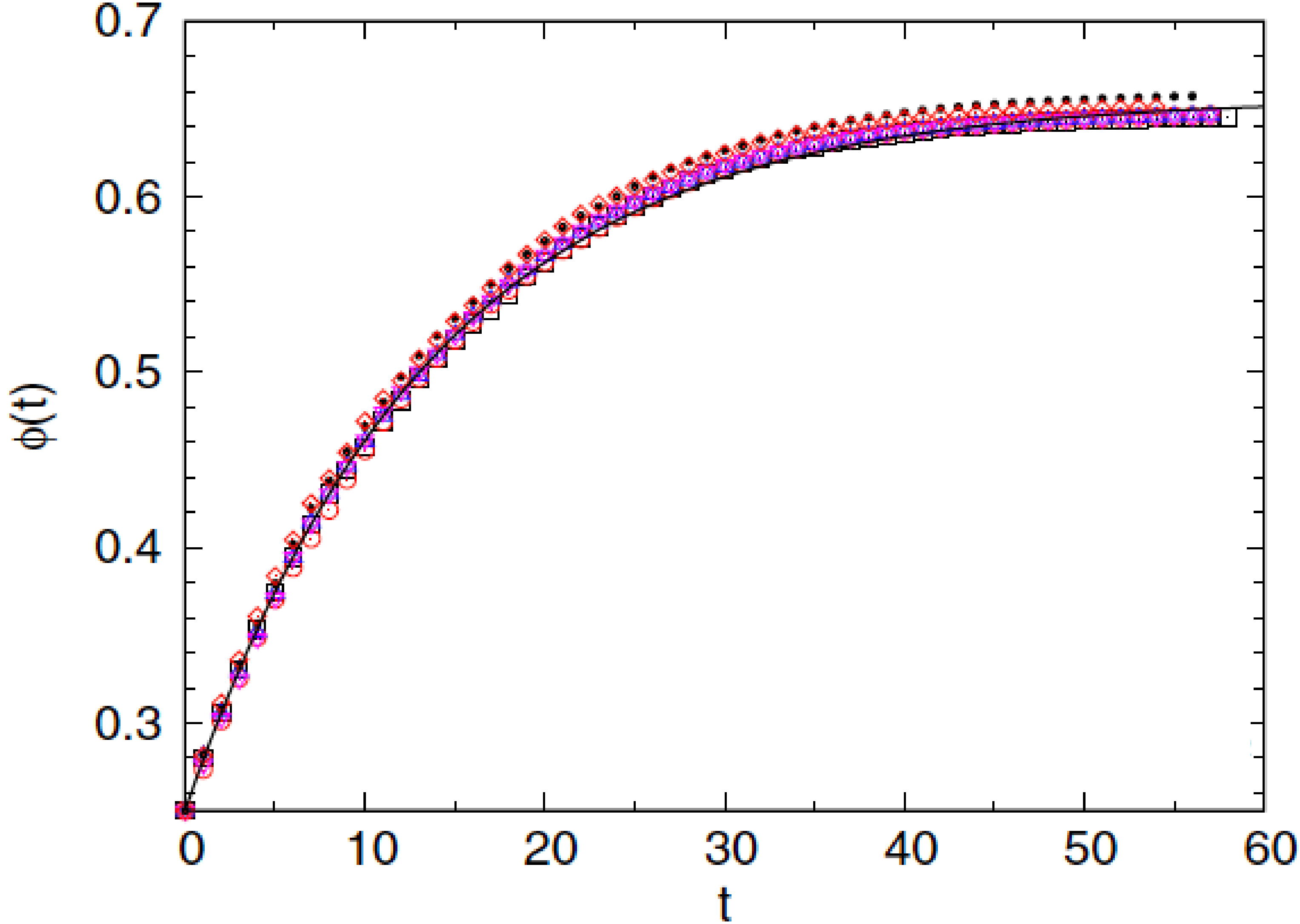
σ_L

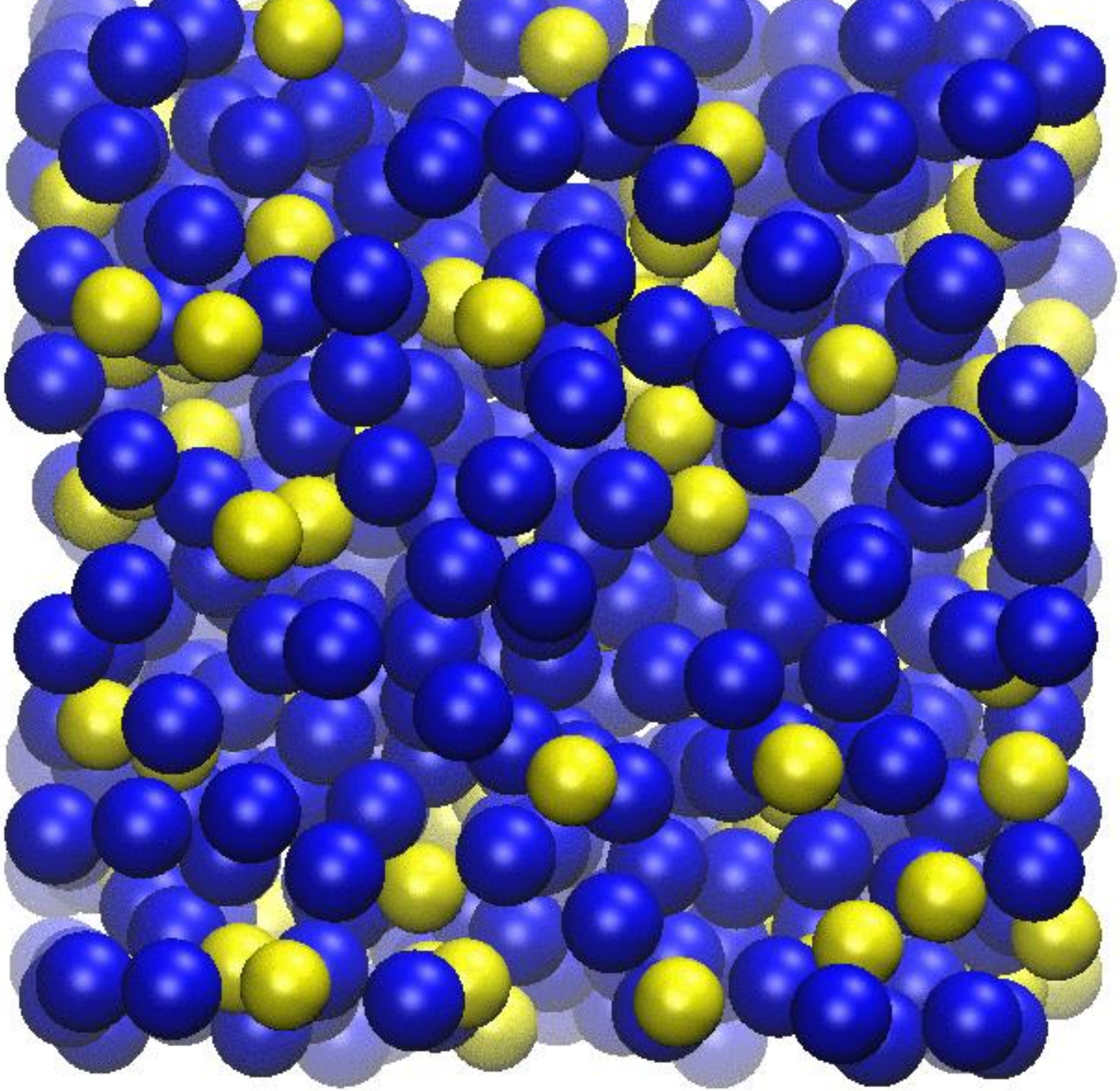


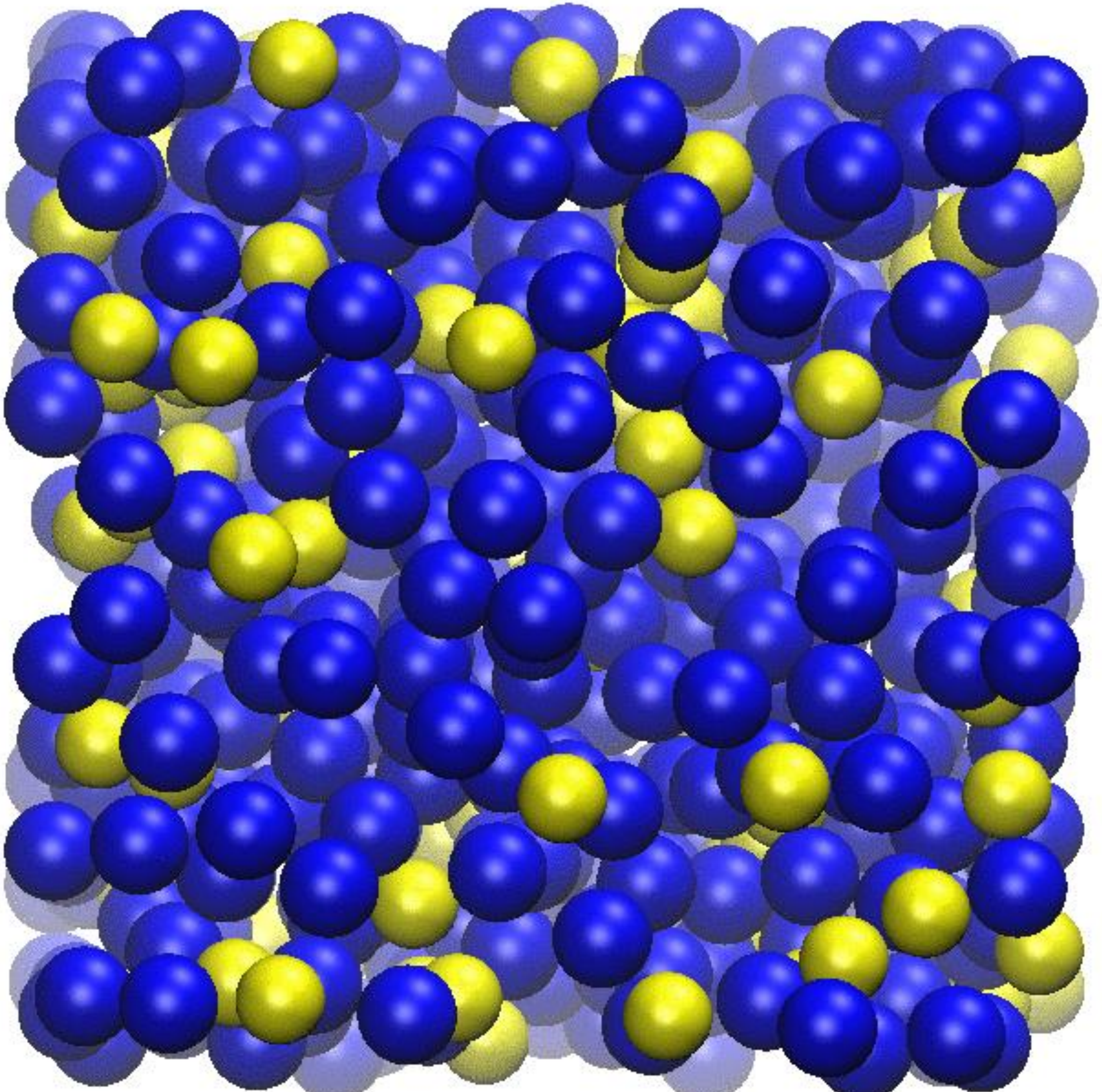
σ_S



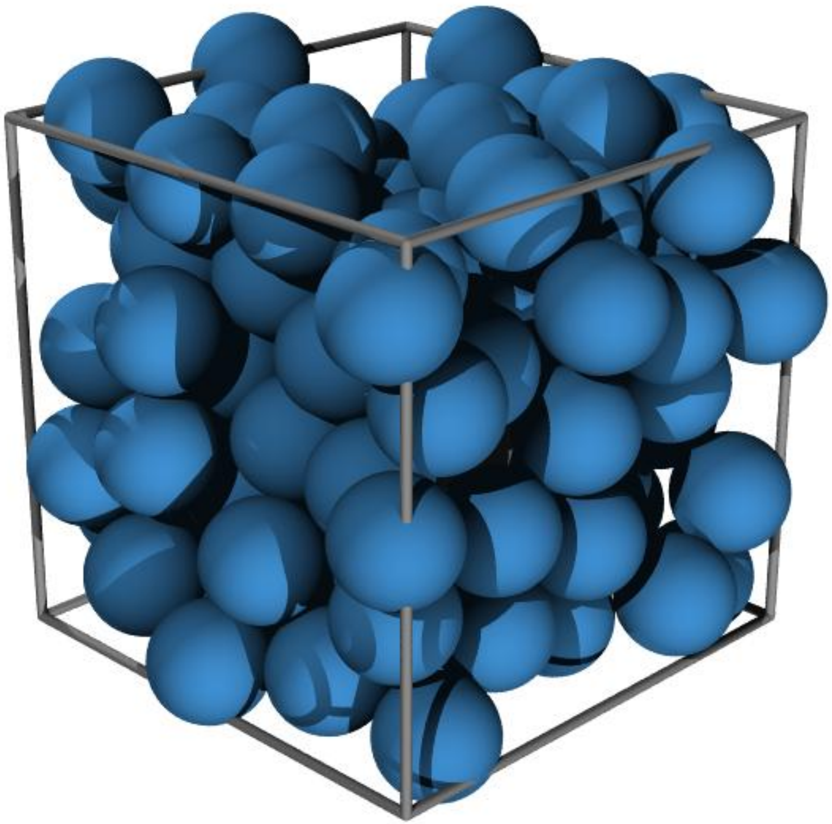
$$f_J - f(t) = (f_J - f_0)e^{-kRt}$$



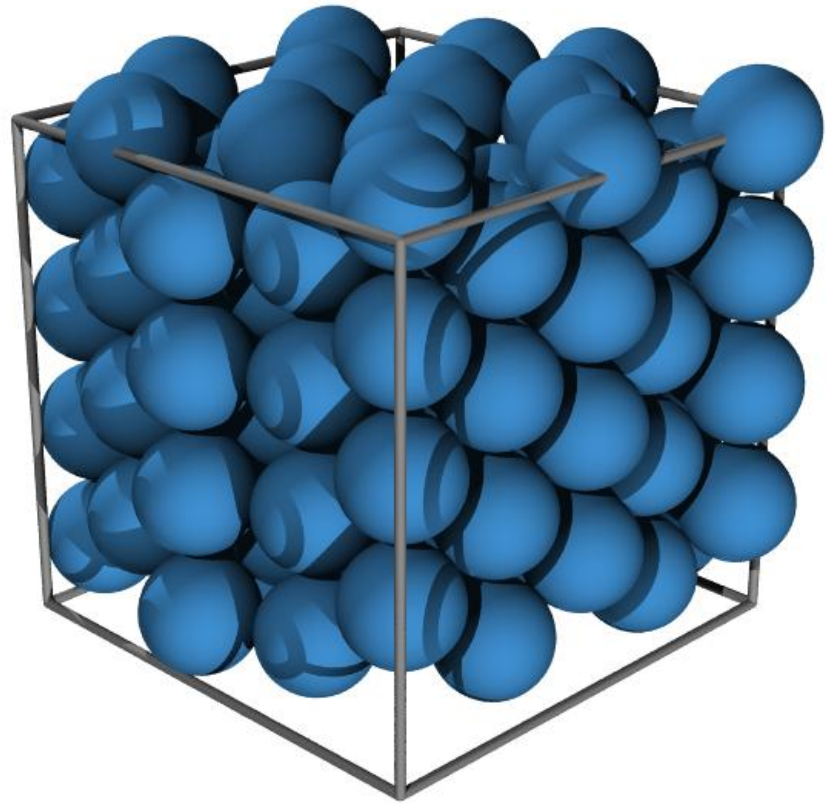




Critical Compression Rate R_c



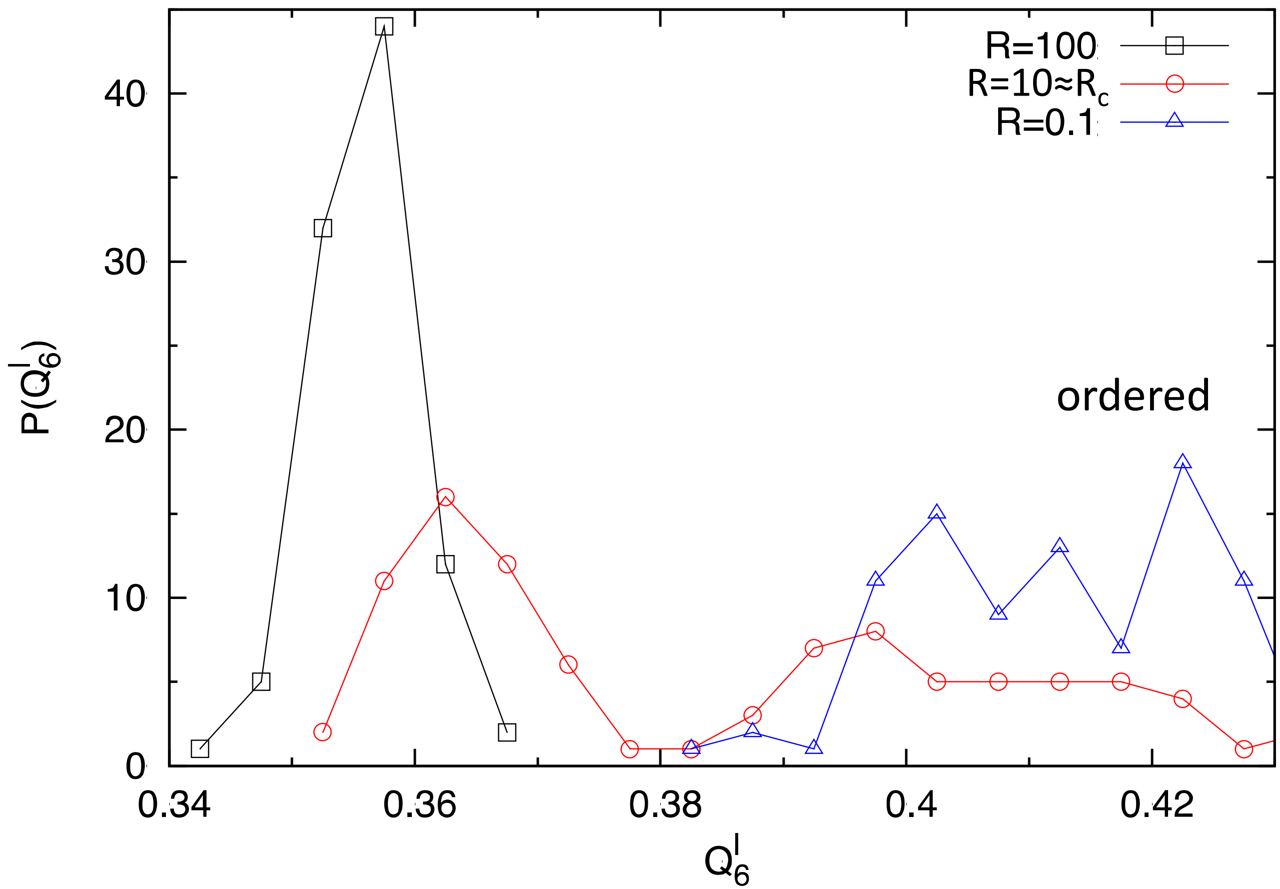
$R>R_c$

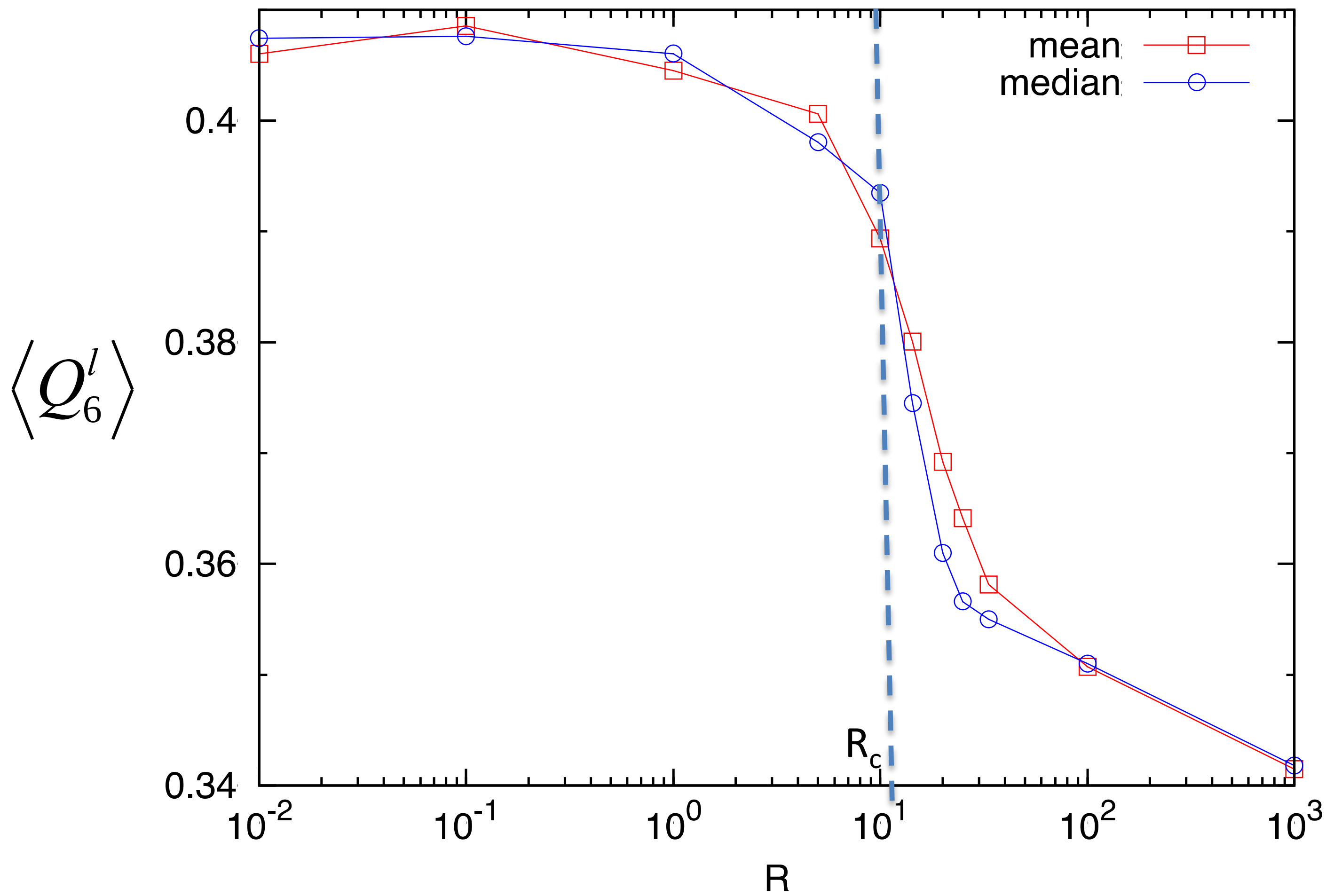


$R<R_c$

K. Wang, W. W. Smith, M. Wang, Y. Liu, J. Schroers, MDS, CSO, Phys. Rev. E 90 (2014) 032311

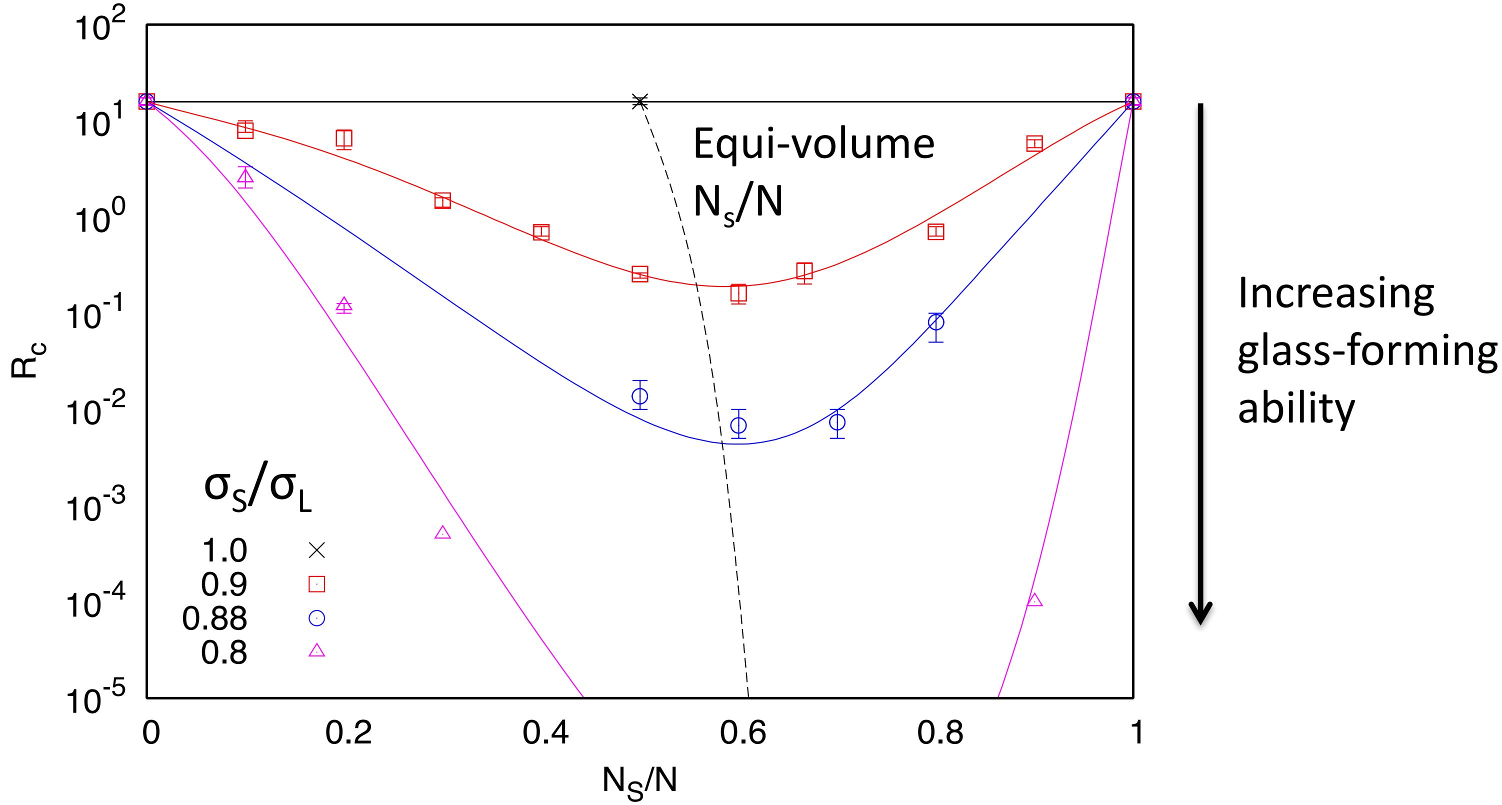
Bond orientational order parameter





mean
median
0.4
0.38
0.36
0.34
$\langle Q_6^l \rangle$
R_c
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
R

Critical Compression Rate



Does R_c continue to decrease with decreasing σ_S/σ_L?

GFA for binary hard spheres

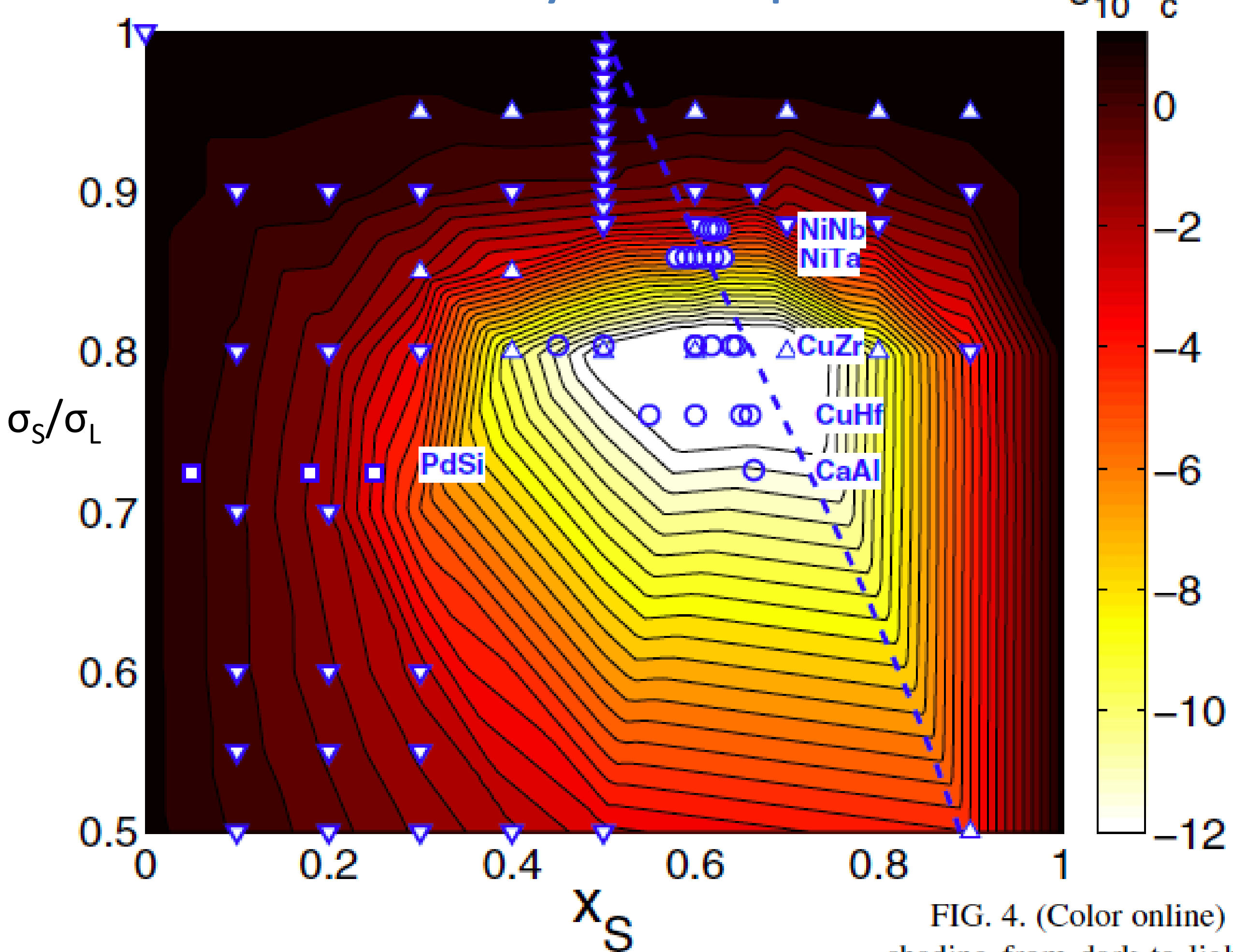


FIG. 4. (Color online) Contour plots of R_c versus α and x_S. The shading from dark to light indicates decreasing R_c on a (base-10) logarithmic scale. The downward triangles are from MD simulations, the upward triangles are obtained by fitting R_c to Eq. (1), and the circles and squares correspond to known metal-metal (e.g., NiNb, NiTa, CuZr, CuHf, and CaAl [30–32]) and metal-metalloid (e.g., PdSi [14]) binary BMGs, respectively. The dashed line satisfies $x_S^* = (1+\alpha^3)^{-1}$, at which the large and small particles occupy the same volume. R_c contours in the central region are extrapolated down to $R_c \sim 10^{-4}$ (a) and 10^{-12} (b).

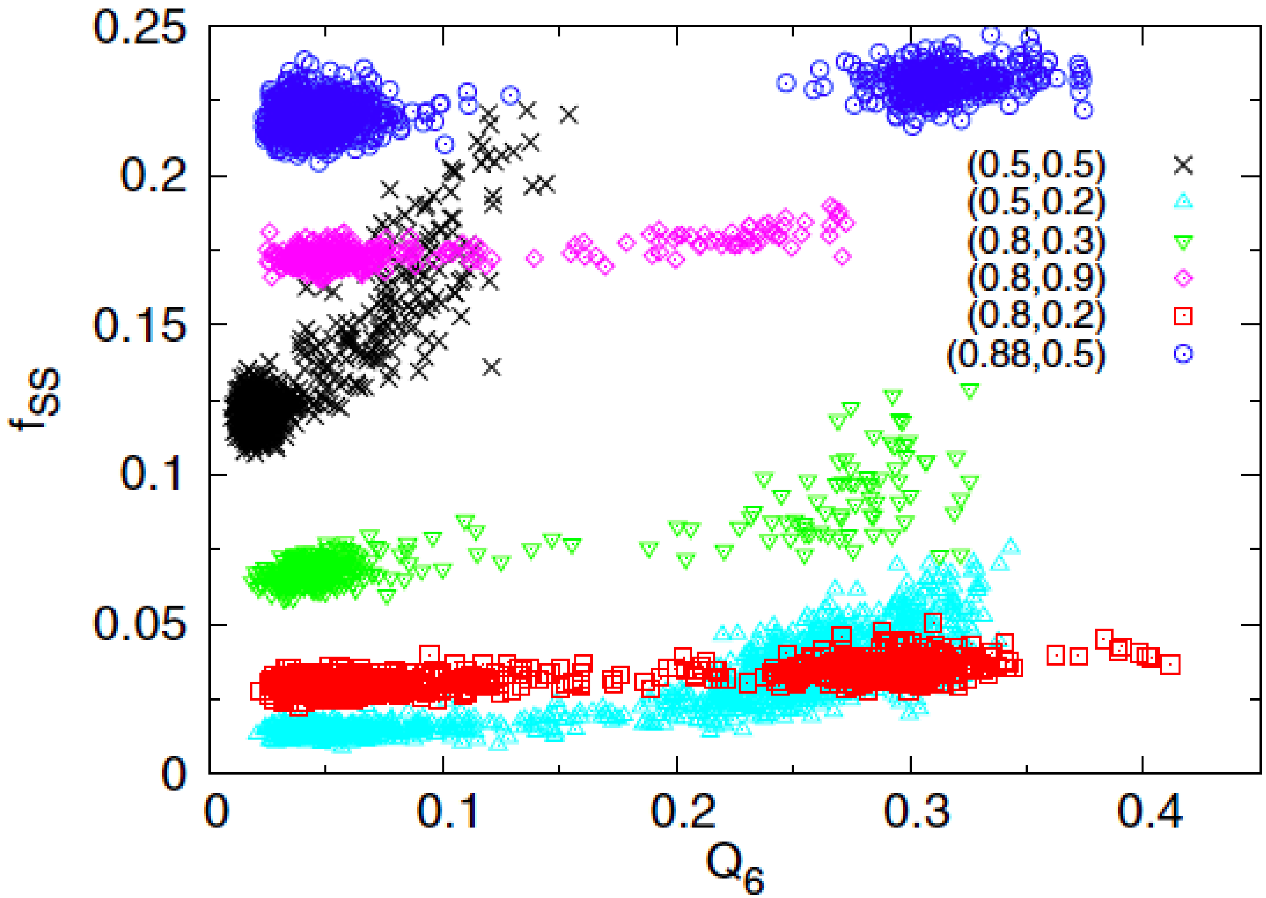
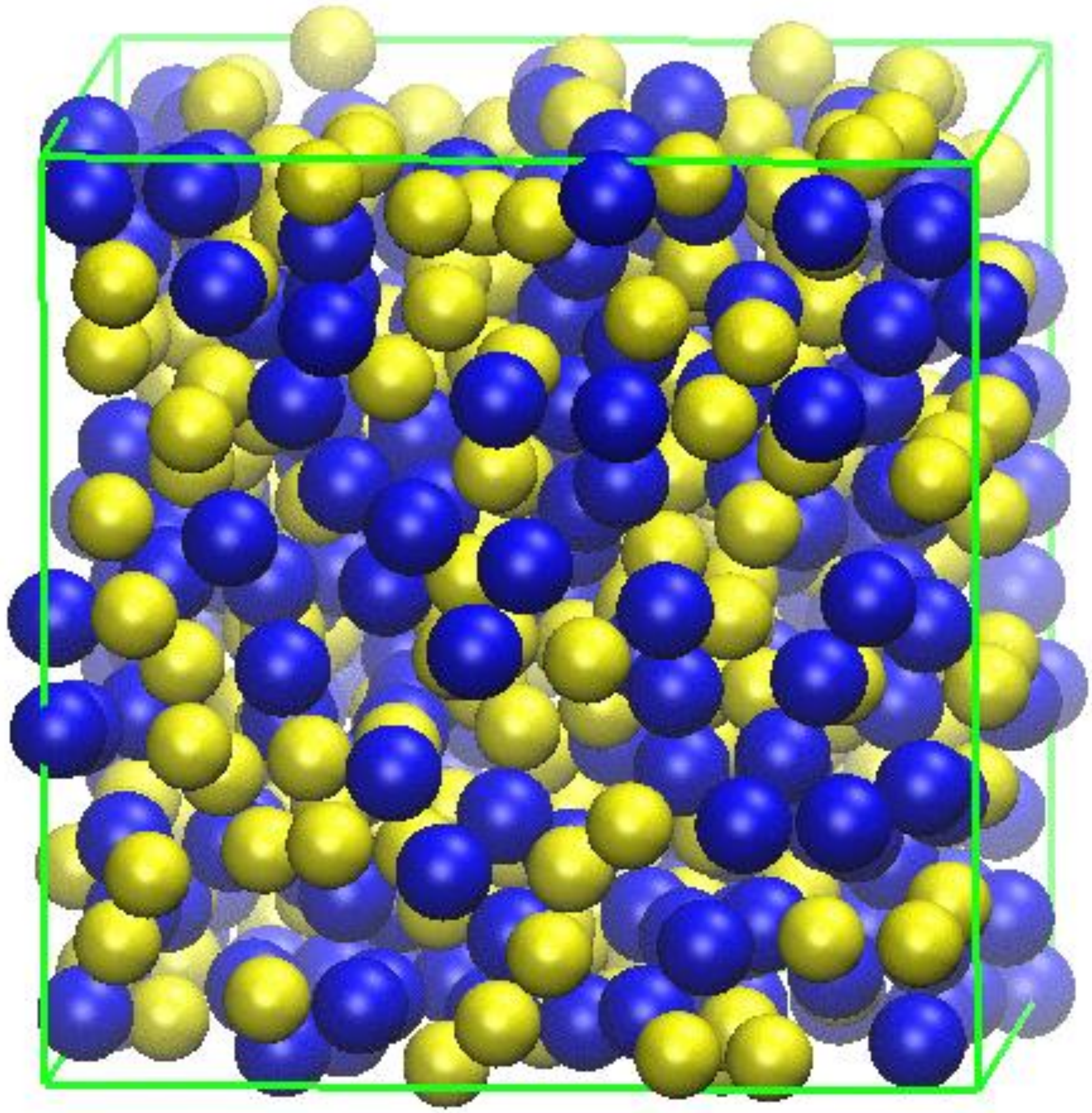
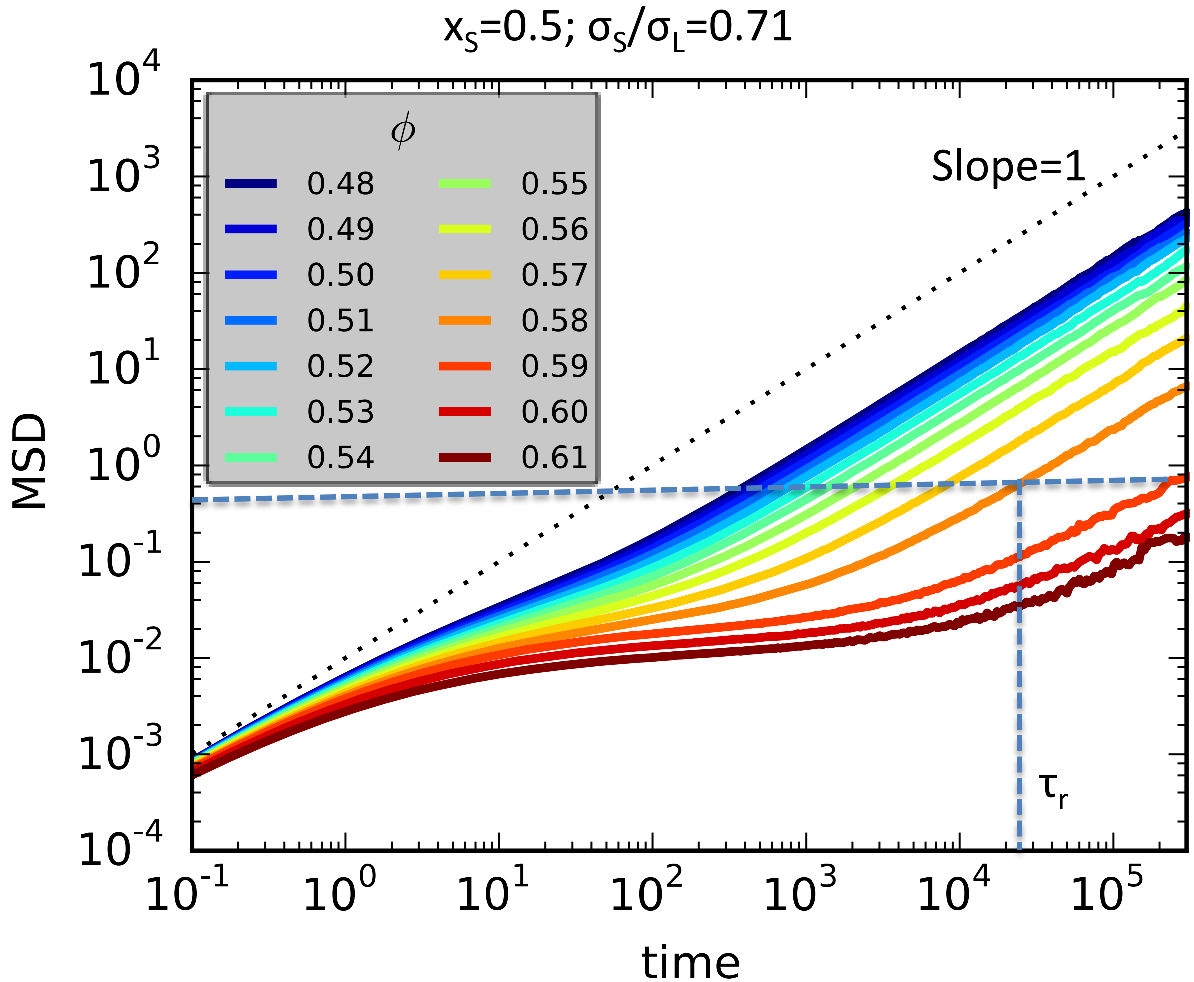


FIG. 6. (Color online) Fraction of small-small nearest neighbors f_{SS} versus Q_6 for six (α, x_S) values: (0.5,0.5) (crosses), (0.5,0.2) (upward triangles), (0.8,0.3) (downward triangles), (0.8,0.9) (diamonds), (0.8,0.2) (squares), and (0.88,0.5) (circles). f_{SS} for (0.8,0.9) has been shifted downward by 0.6 to enable comparison with the other systems. The systems with (0.5,0.5), (0.5,0.2), and (0.8,0.3) partially demix as indicated by the increase in f_{SS} with increasing Q_6.

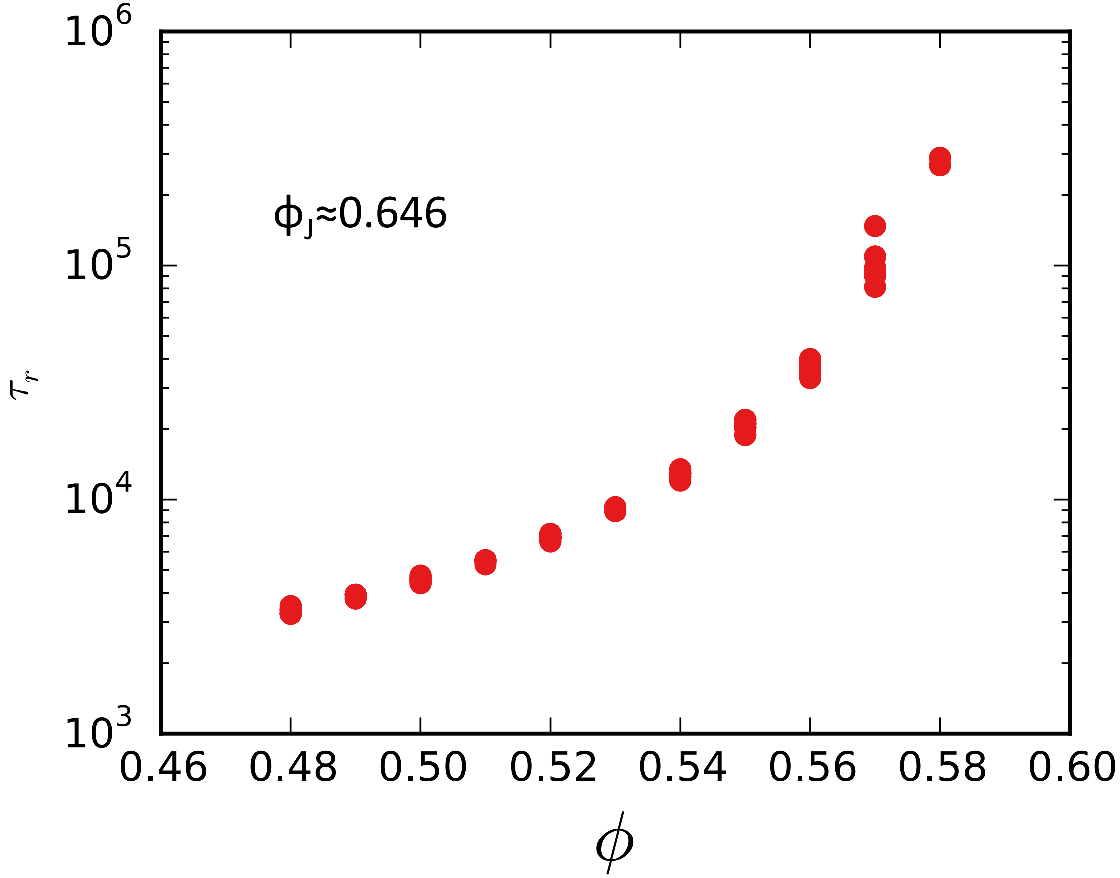
Brownian dynamics of binary hard spheres at fixed packing fraction ϕ



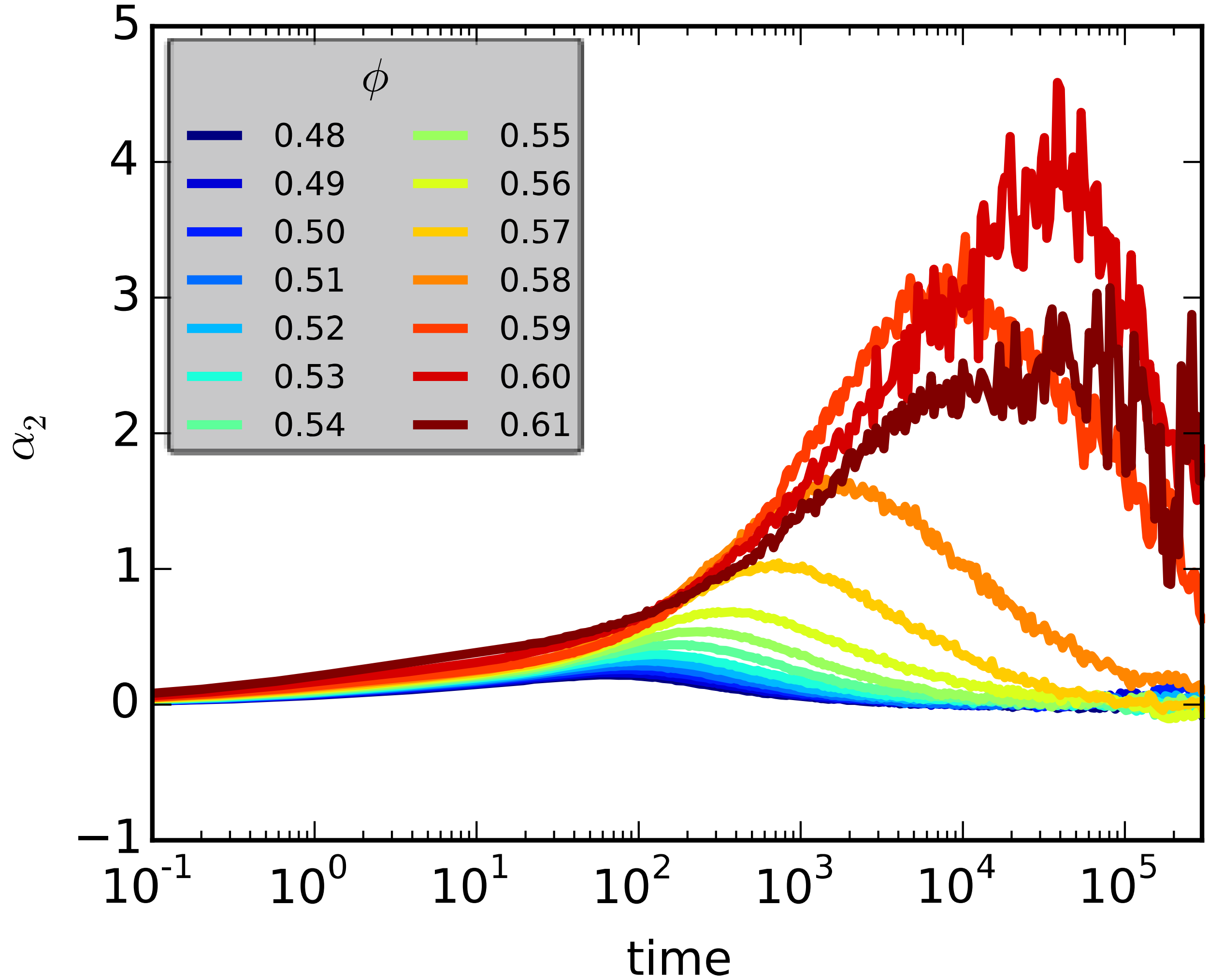


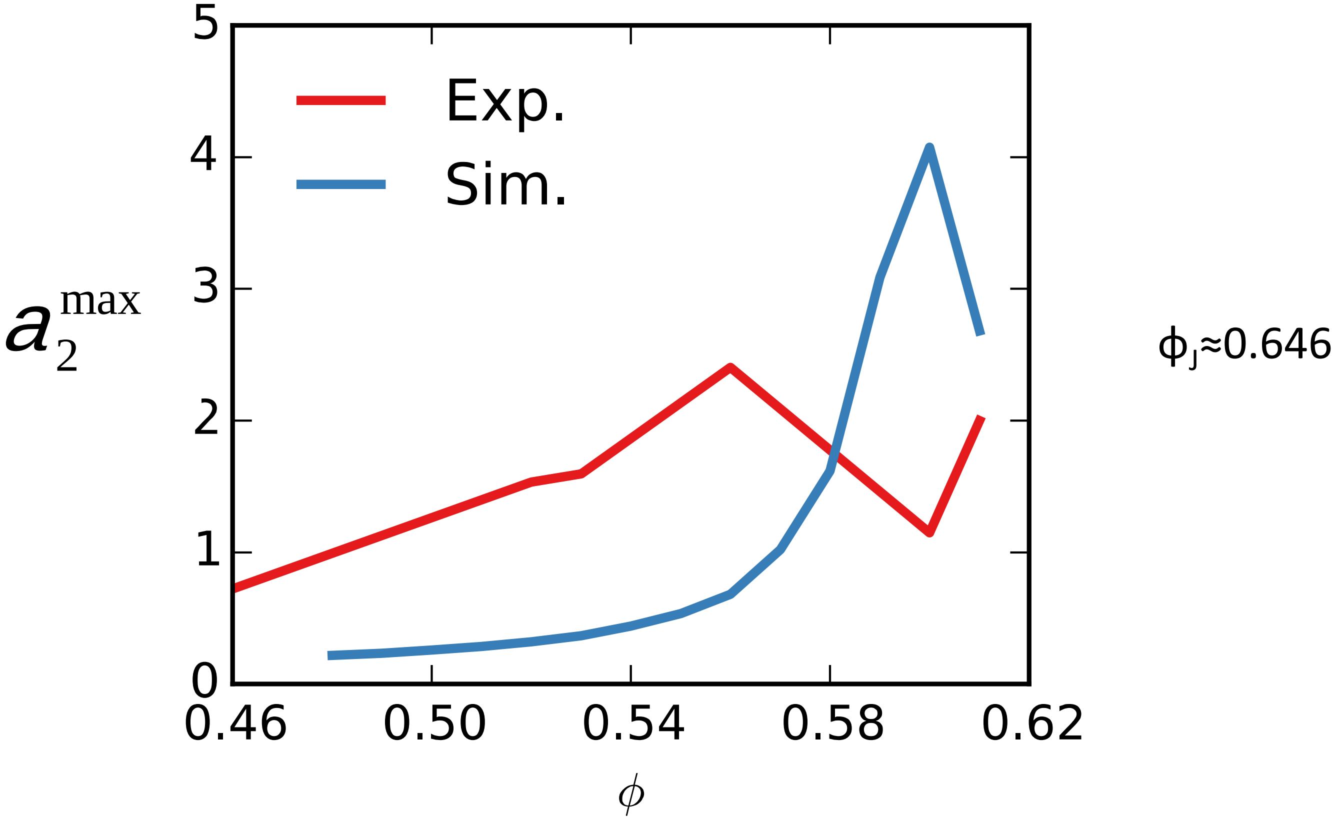
x_S=0.5; σ_S/σ_L=0.71
ϕ
0.48
0.49
0.50
0.51
0.52
0.53
0.54
0.55
0.56
0.57
0.58
0.59
0.60
0.61
Slope=1
τ_r
MSD
time
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^-1
10^0
10^1
10^2
10^3
10^4
10^5

Structural relaxation time



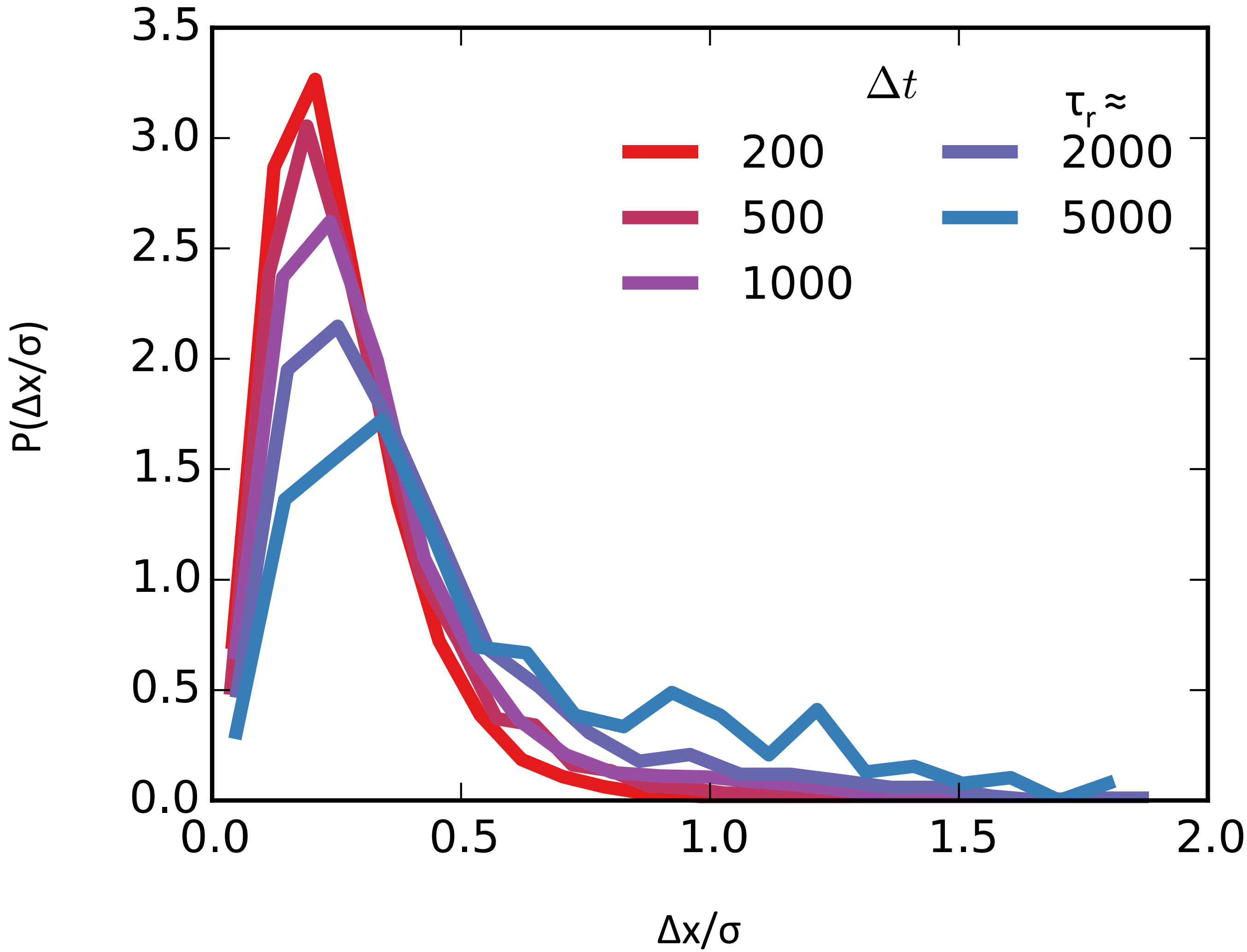
Non-Gaussian Parameter





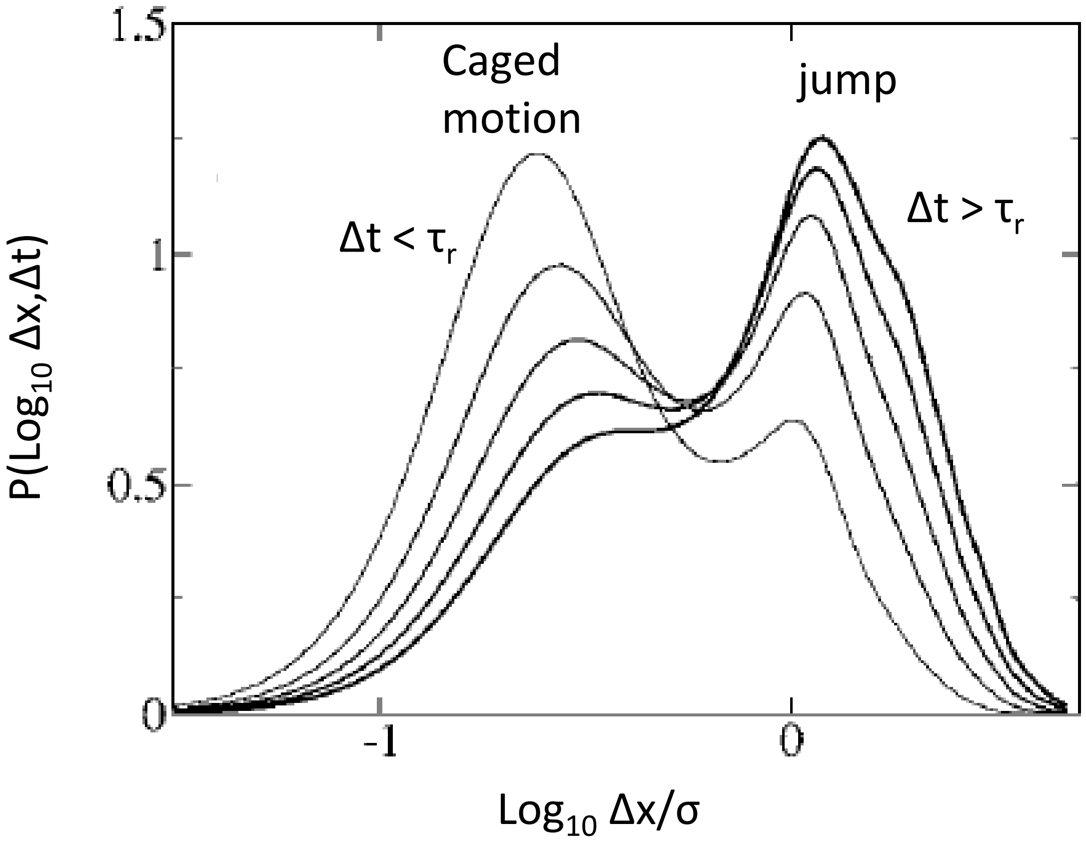
Exp.
Sim.
$a_2^{\max}$
ϕ
0
1
2
3
4
5
0.46
0.50
0.54
0.58
0.62
$\phi_J \approx 0.646$

Displacement Distribution at ϕ=0.58



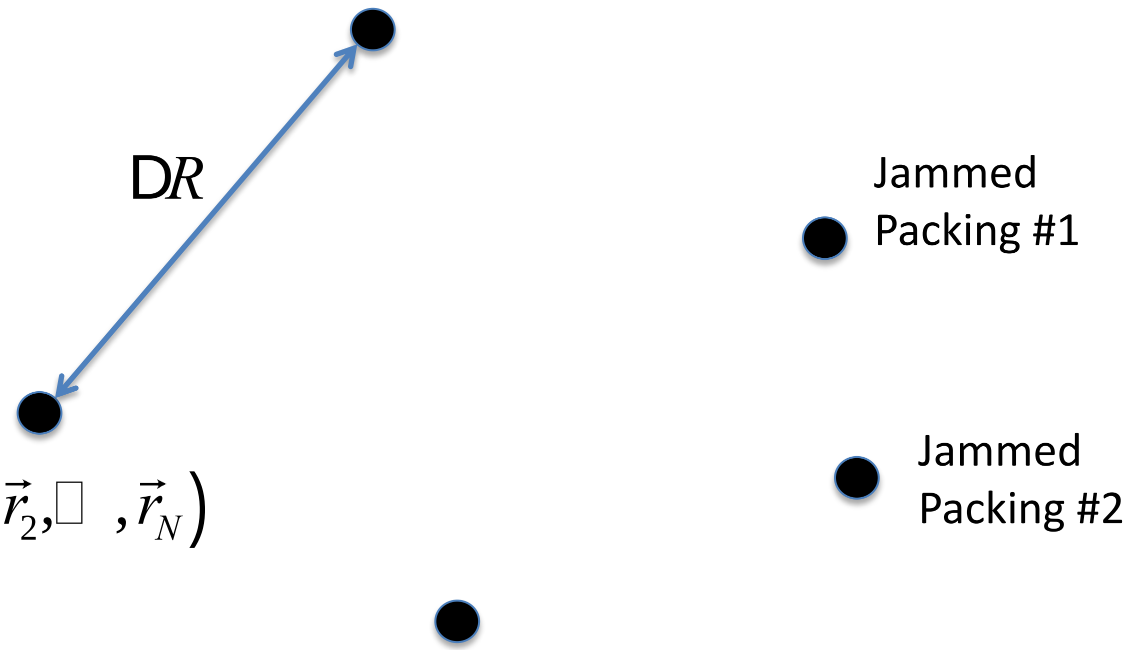
One strong peak representing cage motion

Displacement Probability Distribution



E. Flenner and G. Szamel, PRE 72 (2005) 011205

Jumps in Configuration Space



$$\vec{R} = \left(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N\right)$$

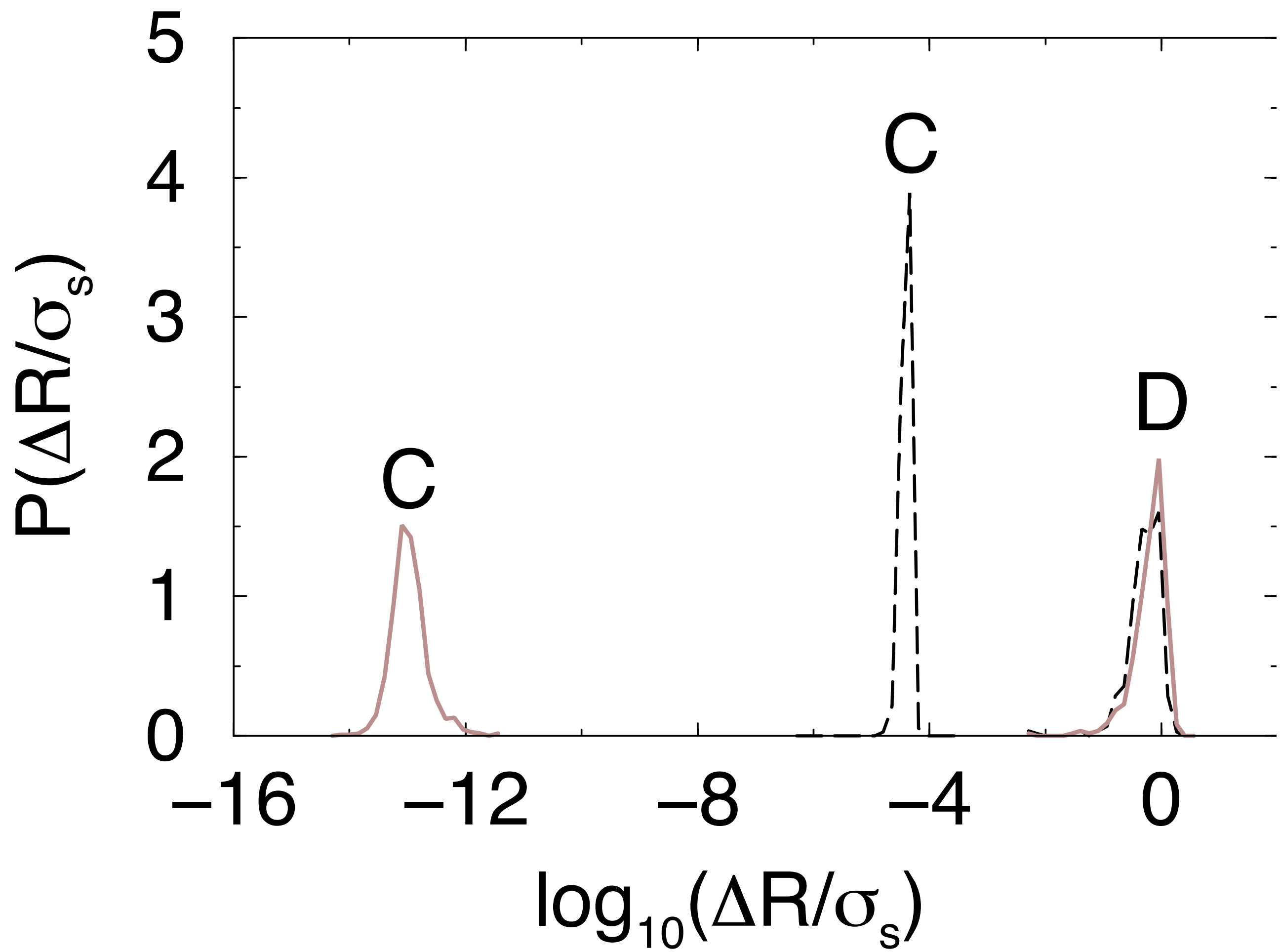
$$\frac{\langle \Delta R \rangle}{\sigma} \approx 0.1$$

If N particles jump, jump size λ=<ΔR>/N

If one particle jumps, jump size λ=<ΔR>

G.-J. Gao, J. Blawzdziewicz, CSO, MDS, PRE 80 (2009) 061304.

Distribution of separations in configuration space between jammed states



Conclusions and Open Questions

- Binary glass-forming metal-metal alloys occur in the best GFA region for binary hard spheres

- Can we create `classes' of glassy materials based on $a_2^{\max}$?

- Do hard sphere systems have different displacement distributions than Lennard-Jones systems?

- Does jump size approach cage size as $f \to f_J$?

- How do avalanches differ for hard spheres vs. Lennard-Jones systems?