Releasing Frustration For Better Relaxation

An investigation of a supercooled liquid on the 3-sphere

Phys. Rev. Lett. 118, 215501 (2017)

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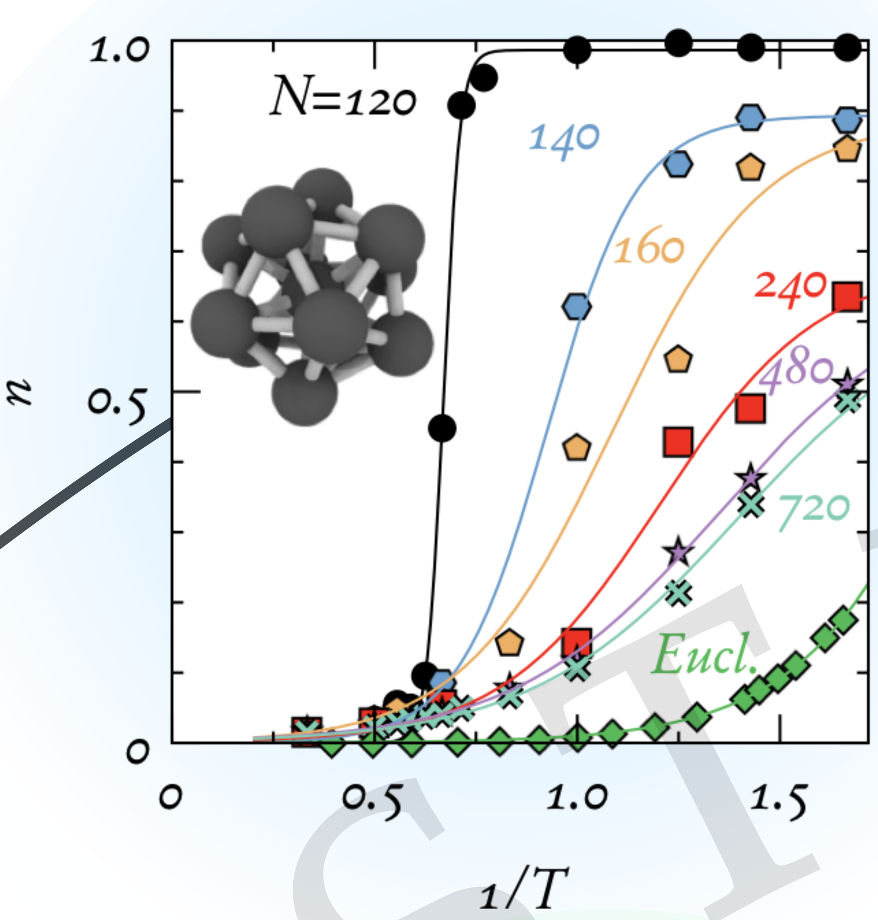
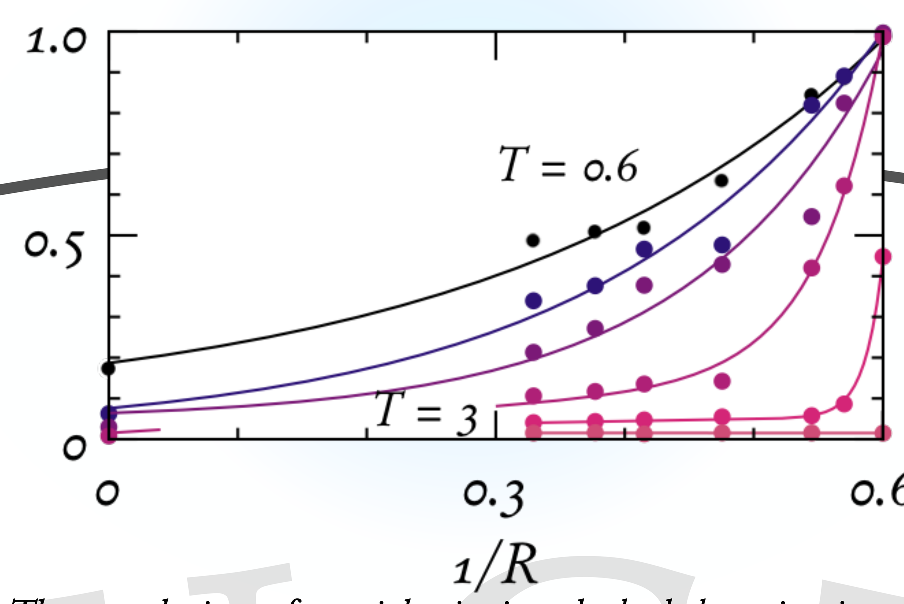
GEOMETRIC FRUSTRATION. *Several theories for dynamic arrest and the glass transition exist. An often invoked mechanism for slow relaxation common to several approaches involves the emergence of energetically preferred geometric motifs that cannot tessellate the Euclidean flat space and therefore induce a "frustrated" dynamics in the liquid. Frustration-limited domain growth theory implies that as we lower the temperature the regions involved in such motifs grow in size, collective behaviour takes place on larger scales and the relaxation gets slower in a super-Arrhenius fashion. According to the theory, this behaviour should be more evident as frustration is reduced, meaning that fragility should increase as frustration decreases. However, were the slowing down dominated by geometric motifs, the size of arrested regions should correspond to the size of geometrically frustrated domains.*

3-SPHERE. *We want to test geometric frustration controlling curvature in 3 dimensions in order to release frustration. Previous studies have only be performed in 2D, where frustration is introduced by curvature (and not released). The surface of a sphere embedded in 4 dimensions is a 3D manifold called 3-sphere. It is possible to tesselate a 3-sphere with icosahedra employing exactly 120 particles (unfrustrated ordered polytope). However, 3-spheres can never tessellate Euclidean space (they are frustrated). We perform Monte-Carlo calculations with local moves that sample both the structural and dynamical properties of a popular Lennard-Jones binary mixture which is known to form frustrated icosahedral domains in Euclidean space. We fix the density of the liquid and change the curvature: for small numbers of particles N we obtain small 3-spheres of high curvature and low frustration; large 3-spheres approach the Euclidean limit.*

STRUCTURE

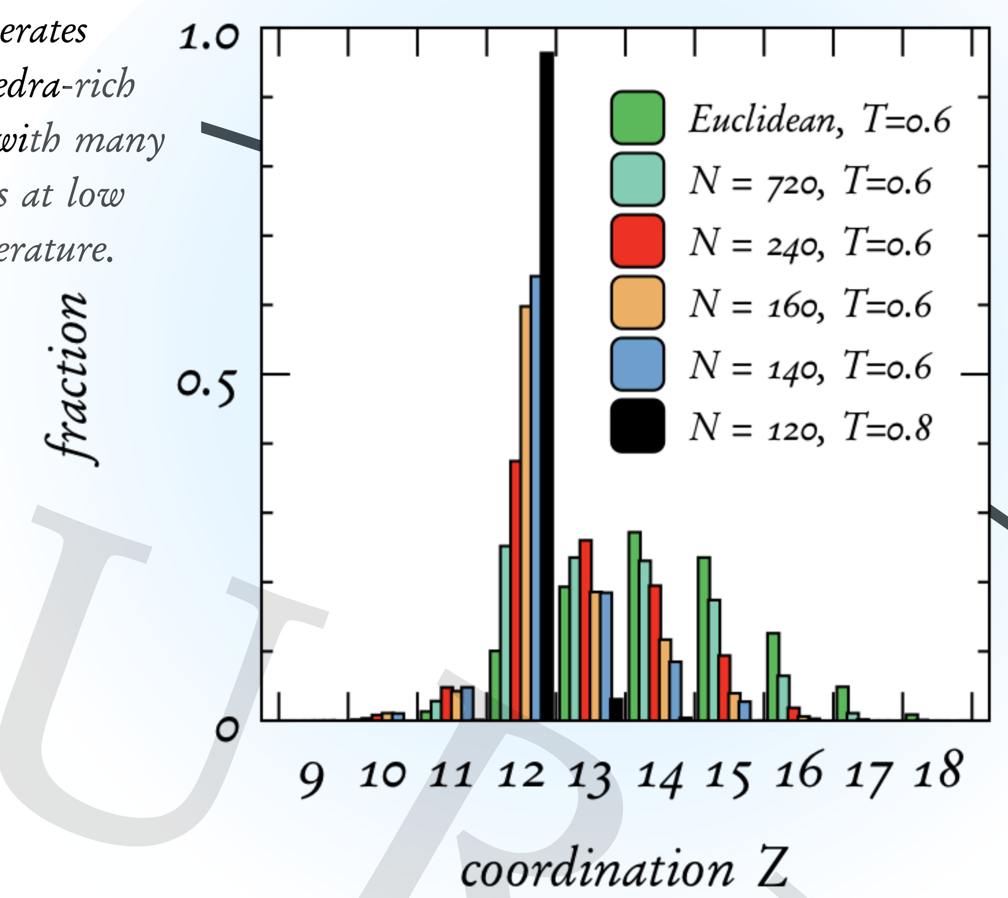


The concentration of particles involved in icosahedral domains increases at low temperature. Sharp transitions at small N, broad crossover in the Euclidean limit.

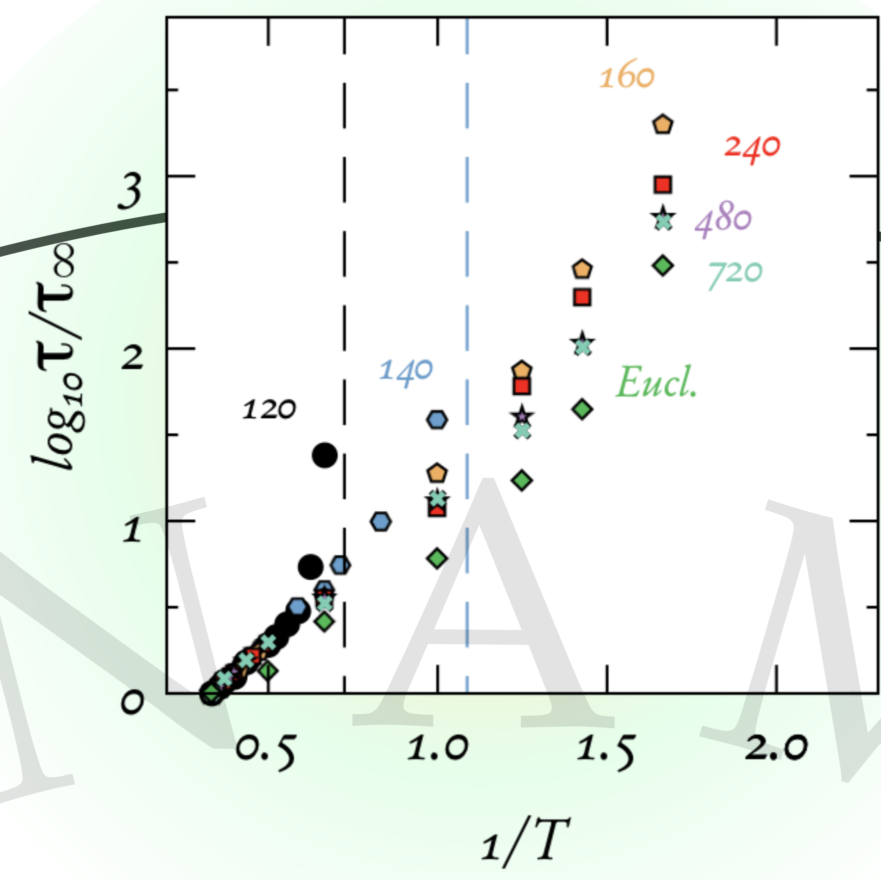


The population of particles in icosahedral domains increases sharply with the curvature 1/R, meaning that frustration decreases markedly only in the limit case of high curvatures.

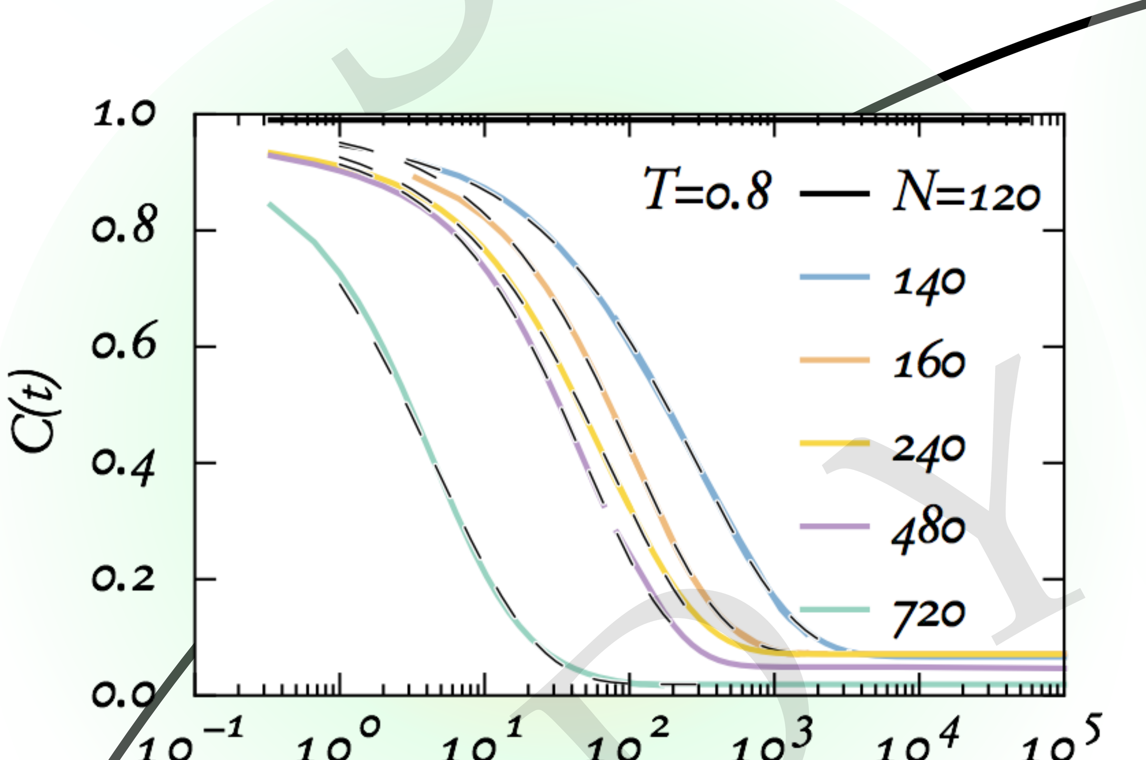
A mild degree of frustration generates icosahedra-rich liquids with many defects at low temperature.



DYNAMICS



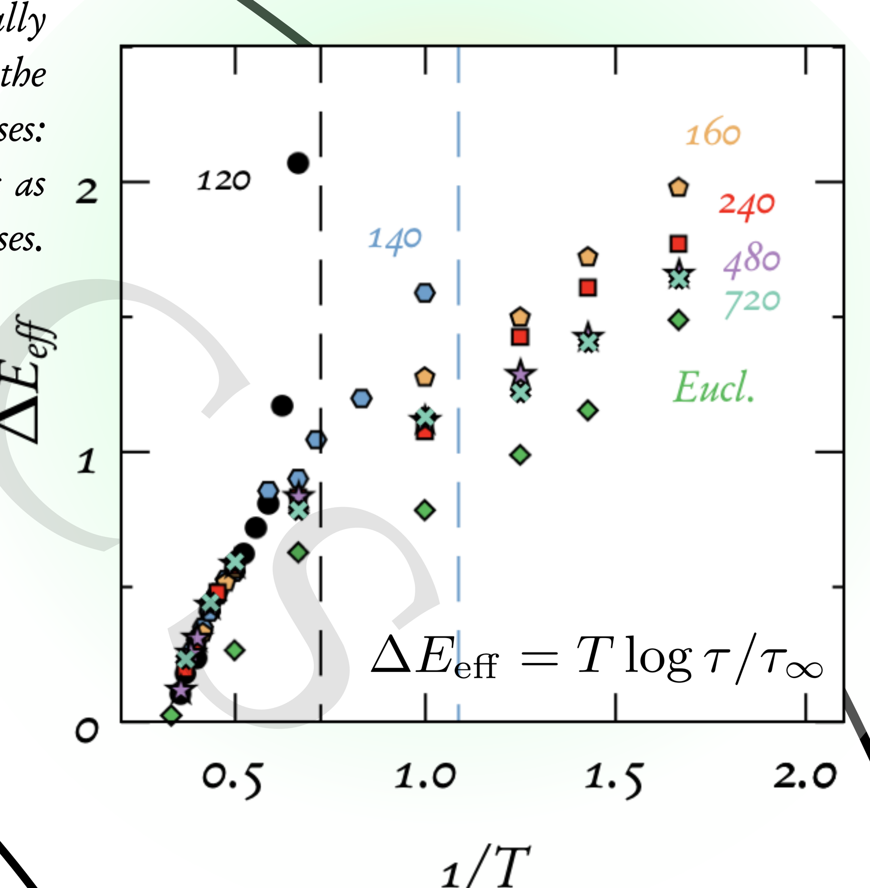
The relaxation times truly diverge only for N=120,140.



Decorrelation of particle neighborhoods is measured to extract relaxation times and define mobile and immobile particles.

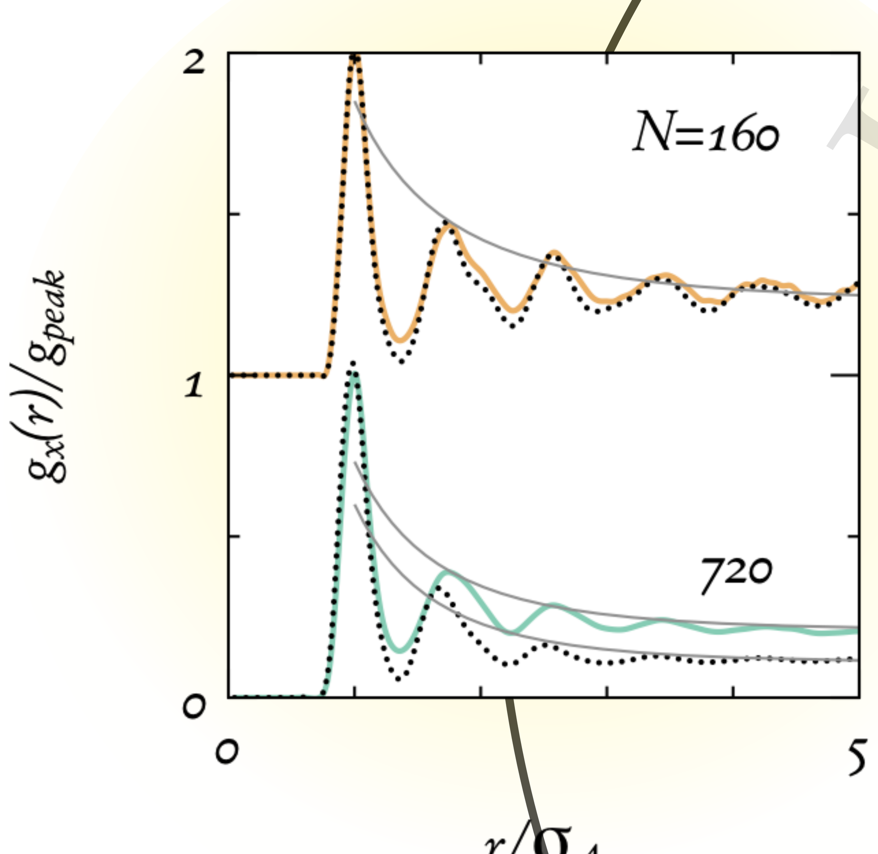
$$C(t)=\left\langle \frac{1}{N}\sum_{i=1}^{N}\frac{v_i(t_0+t)\cdot v_i(t_0)}{v_i^2(t_0)}\right\rangle_{t_0}$$

The activation barrier ΔE_{eff} increases continuously as the temperature is decreased and it monotonically decreases as the curvature decreases: fragility decreases as frustration increases.



LENGTHS

Lengths are extracted in real space from the decay of suitable radial distribution functions.

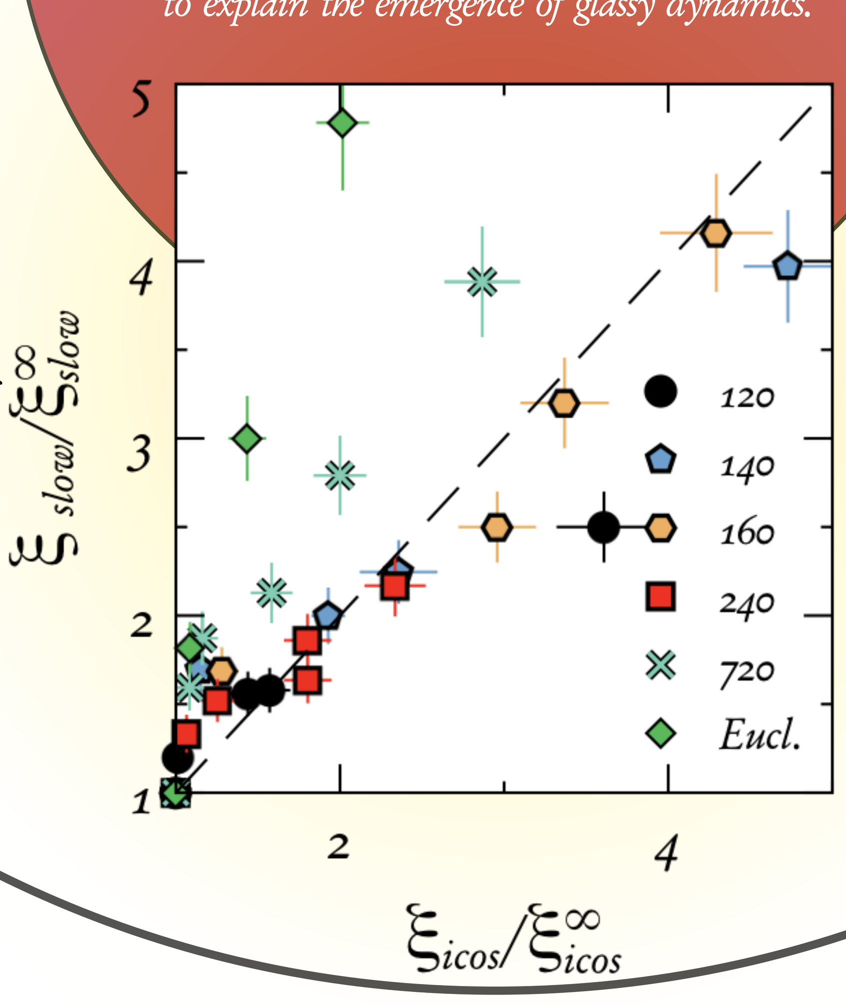


Pair distribution functions for the slow domains (continuous coloured lines) and icosahedral domains (dotted lines) for two systems with different curvatures at the same reduced temperature T=0.8.

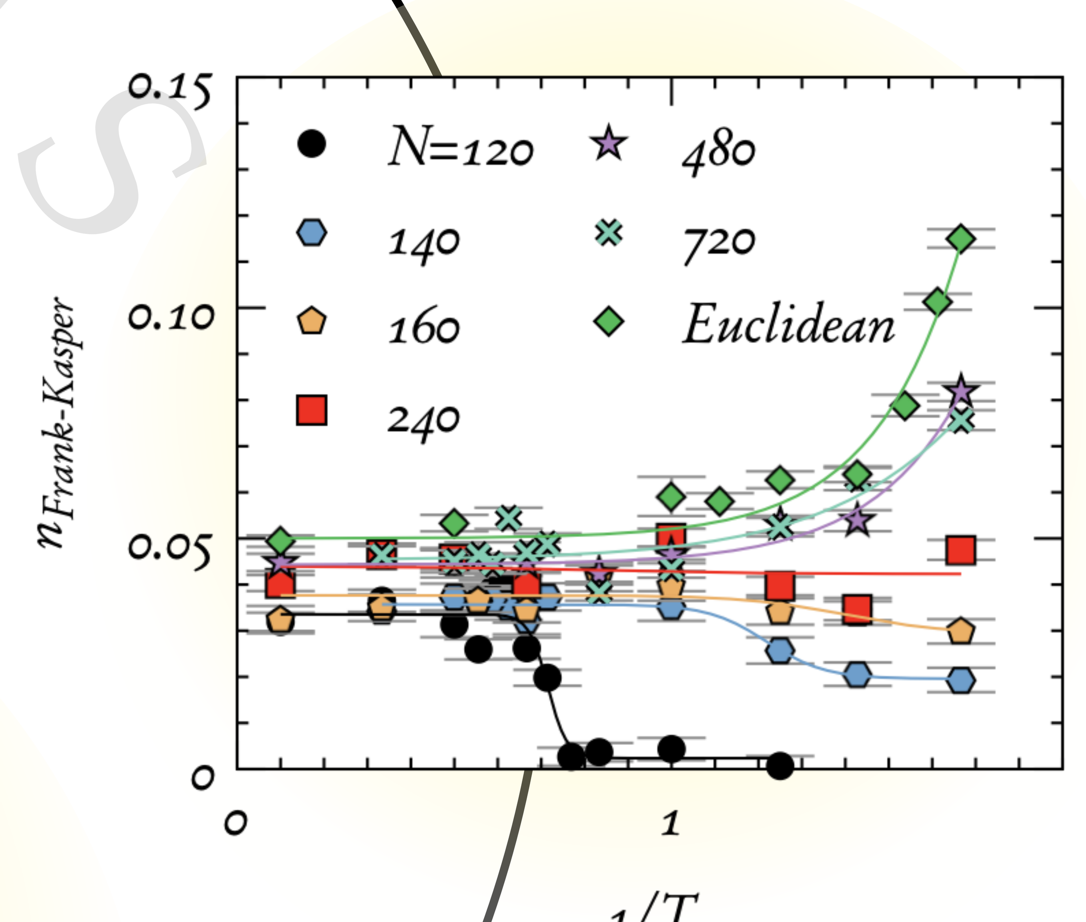
At high curvature, frustration is suppressed, and an ordered state emerges where structure and dynamics are coupled. In the Euclidean limit, structure and dynamics become decoupled.

The frustration of geometric motifs is not sufficient to explain the emergence of glassy dynamics.

The size of slow domains is decoupled to the size of icosahedral domains for large spheres and the Euclidean limit.



The size of slow domains is tightly coupled to the size of icosahedral domains for small, highly curved spheres.



Possible additional sources of frustration: the emergence of so-called Frank-Kasper bonds (related to Laves phases) in the Euclidean limit

FURTHER READING

This work: F. Turci, G. Tarjus, C. P. Royall, Phys. Rev. Lett. 118, 215501, (2017).

On geometric frustration: G. Tarjus, S. A. Kivelson, Z. Nussinov, and P. Viot, J. Phys.: Condens. Matter 17, R1143 (2005).

On 2D numerical tests: F. Sausset, G. Tarjus, and P. Viot, Phys. Rev. Lett. 101, 155701 (2008)
F. Sausset and G. Tarjus, Phys. Rev. Lett. 104, 065701 (2010).
J.-P. Vest, G. Tarjus, and P. Viot, Molecular Physics 112, 1330 (2014).