Slow or Fast, Strong or Fragile

Generalized Mode-Coupling Theory of the Glass Transition for Hard Spheres

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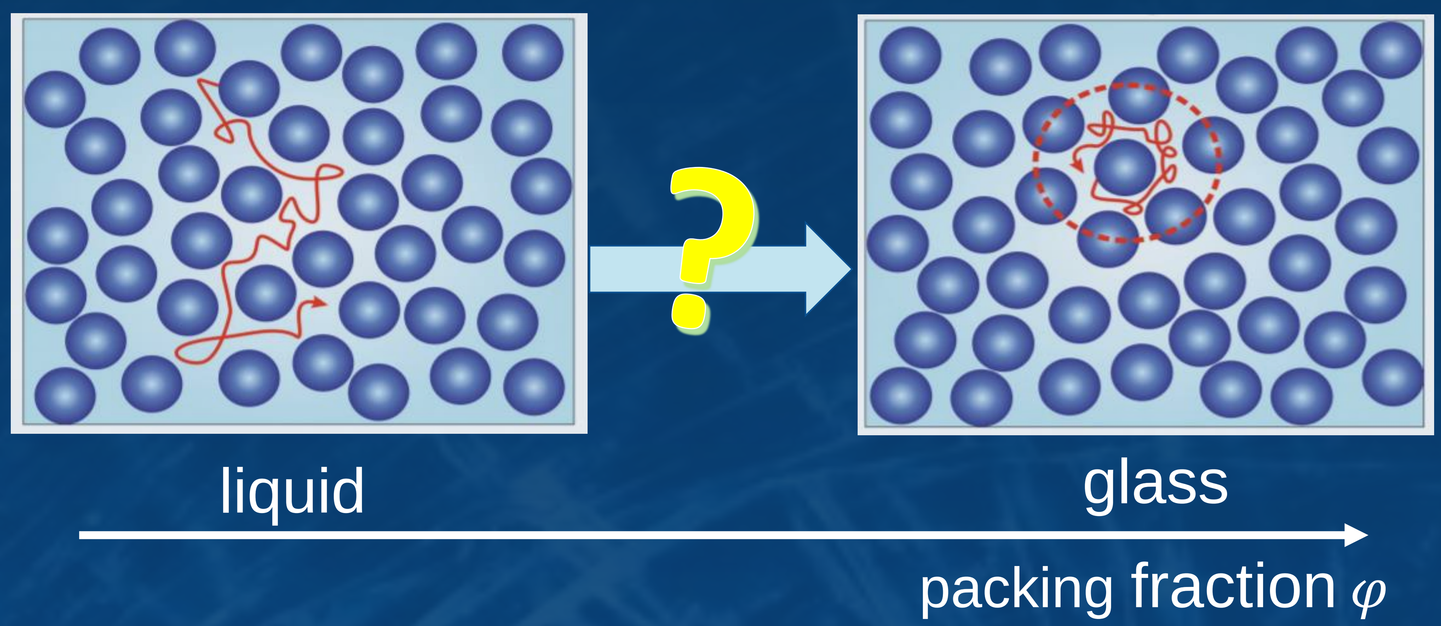
Motivation



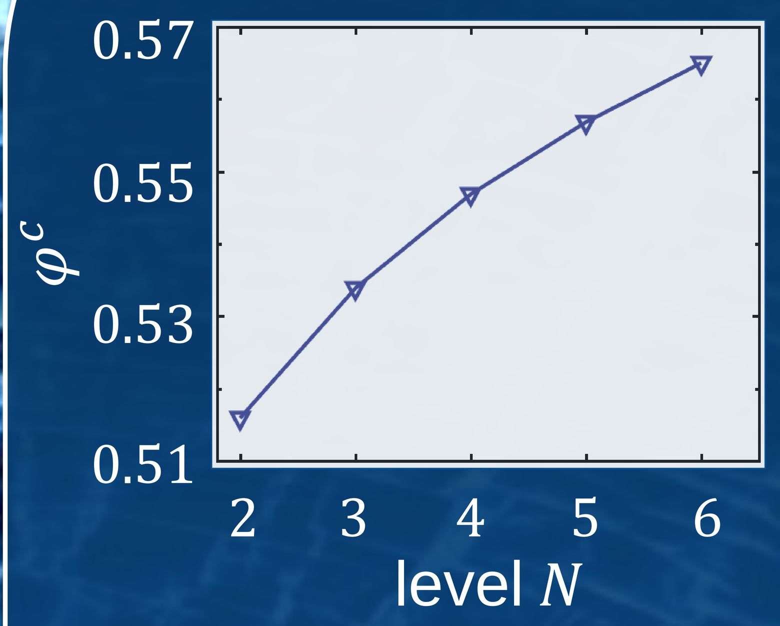
Q: The structures of liquid and glass are very similar but the relaxation of glass is tremendously slower than the liquid. How to explain it? What happened from liquid to glass?

A: Generalized mode-coupling theory (GMCT).
→ *first-principles based & determined by structure*

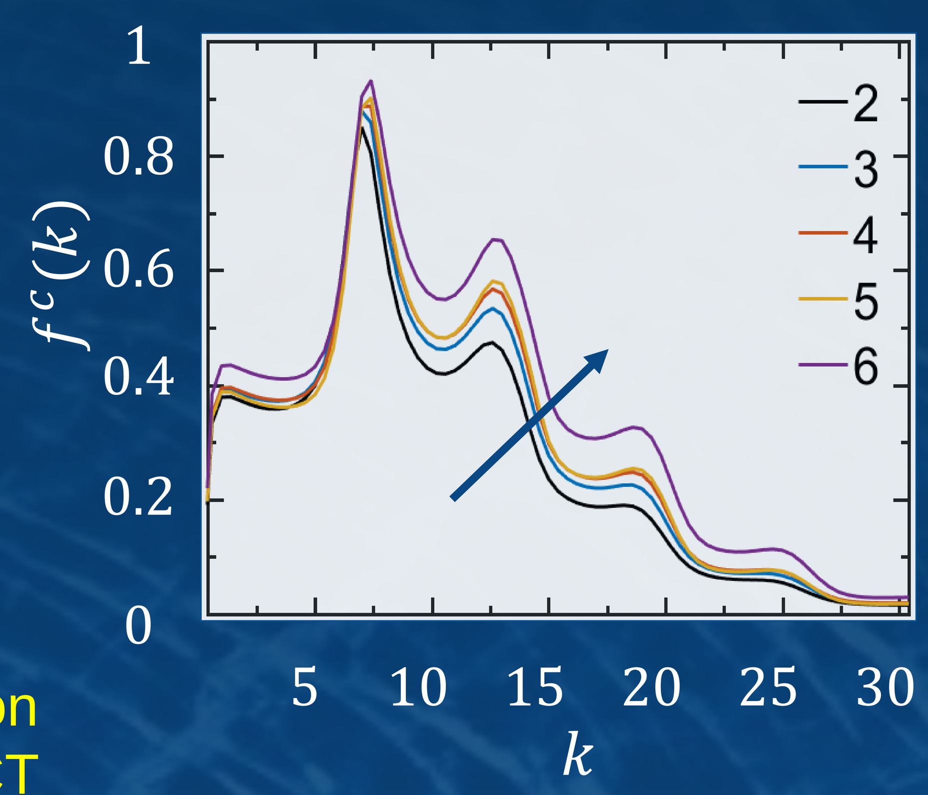
Here we show that GMCT for Percus-Yevick hard spheres predicts the glass transition with **higher critical density, slower dynamics near the ciritical point,** and **lower fragility**. All come closer to empirical observations.

GMCT can be applied to design advanced glass-forming materials in the future.

Critical Point

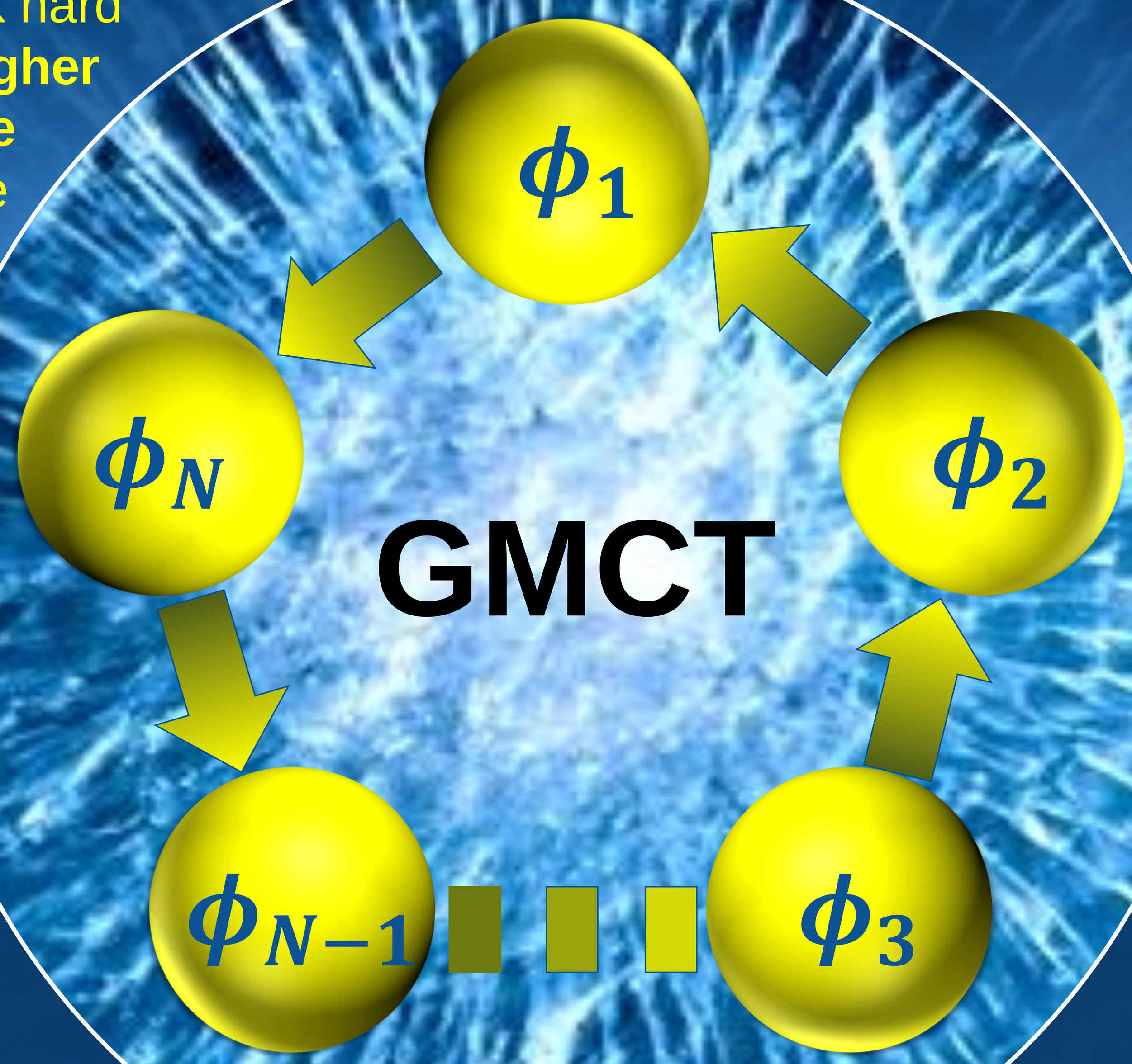


The critical packing fraction φ^c grows with the GMCT level N, and approaches the experimental value 0.563.



The nonergodicity parameters $f^c = \lim_{t\to\infty} \phi_1(k,t)$ increase with the GMCT level N, implying the system is more solid-like.

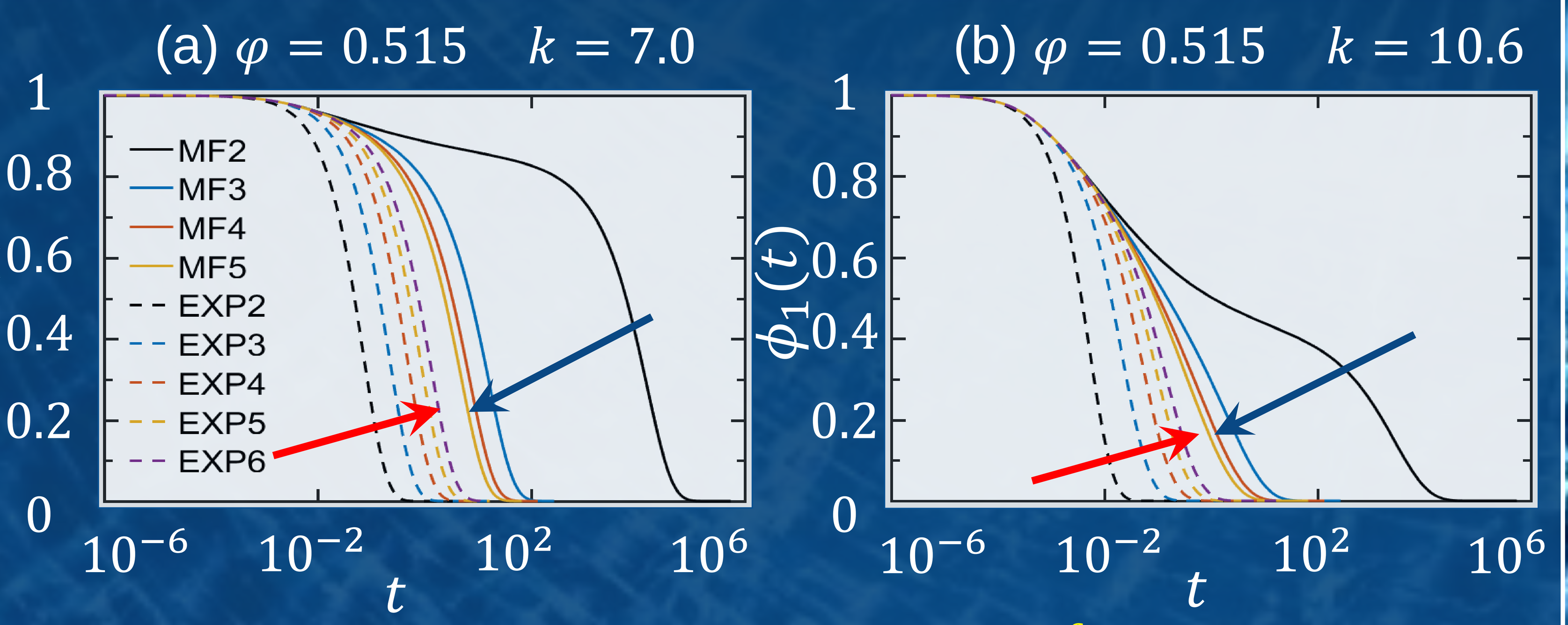
The values seem to converge for higher levels. The exception of the 6th level is due to numerical inaccuracy.



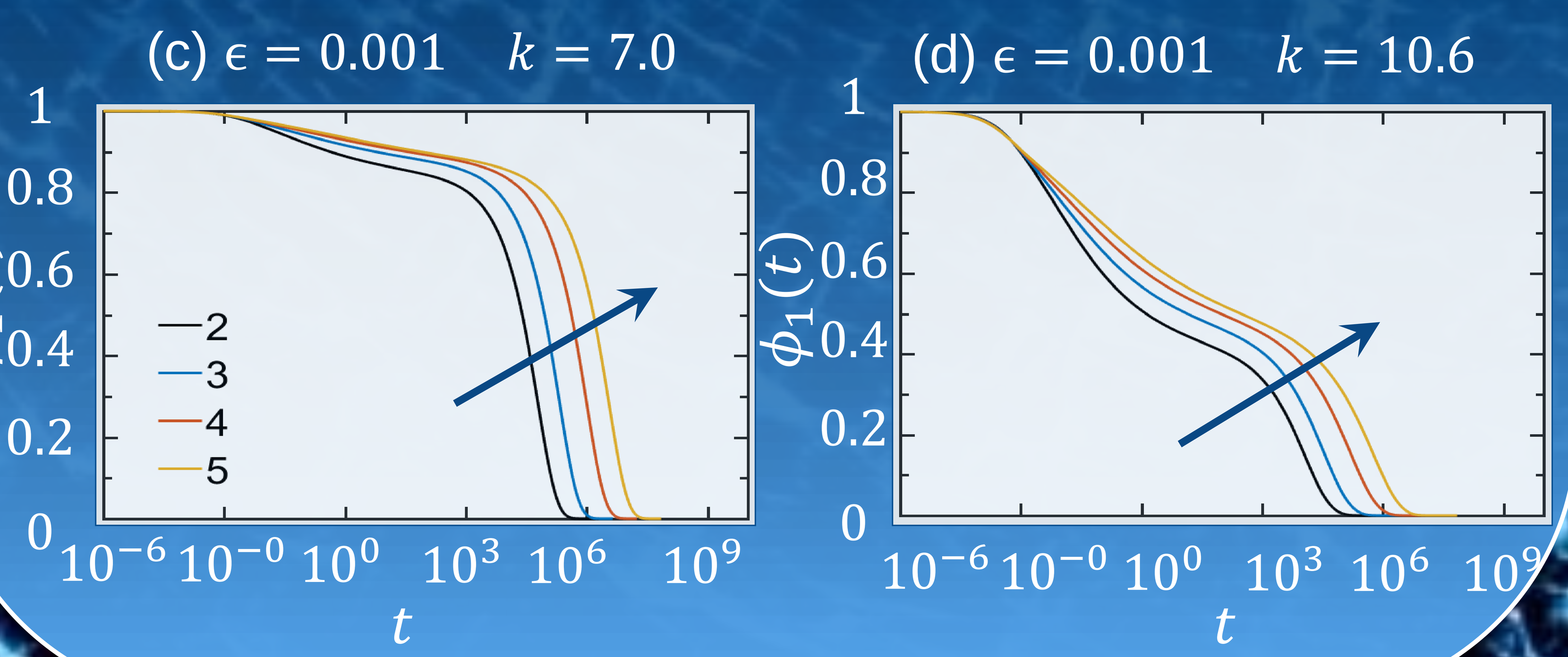
Full-Time Relaxation

Mean field (MF) closures provide an upper bound to the dynamics and exponential (EXP) closures give a lower bound. When the level increases, the two kinds of closures will converge.

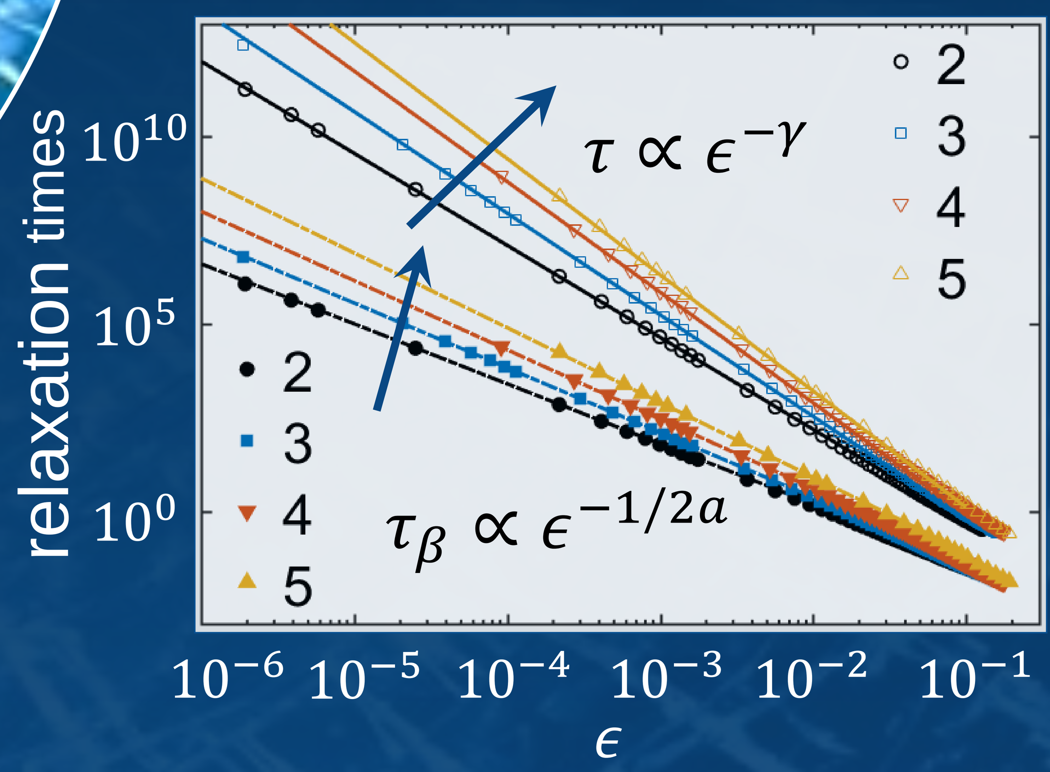
For a given **packing fraction φ**, the higher MF closure level gives **faster relaxations** at all wave vectors.



For a given ***reduced* packing fraction $\epsilon = \frac{\varphi^c - \varphi}{\varphi^c}$**, the higher MF closure level gives **slower relaxations** at all wave vectors.



Scaling Laws



$\tau_\beta \propto \epsilon^{-1/2a}$ corresponds to the β relaxation regime, the plateau at intermediate times. a decreases with higher level N.

$\tau \propto \epsilon^{-\gamma}$ corresponds to the final-decay α relaxation regime. γ increases with higher level N.

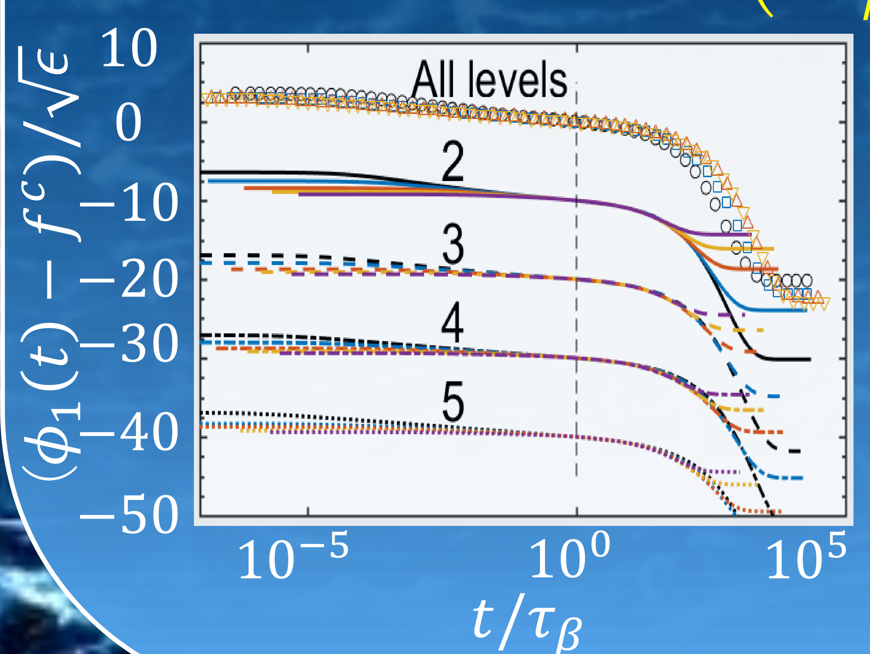
The larger γ, the less fragile the system.

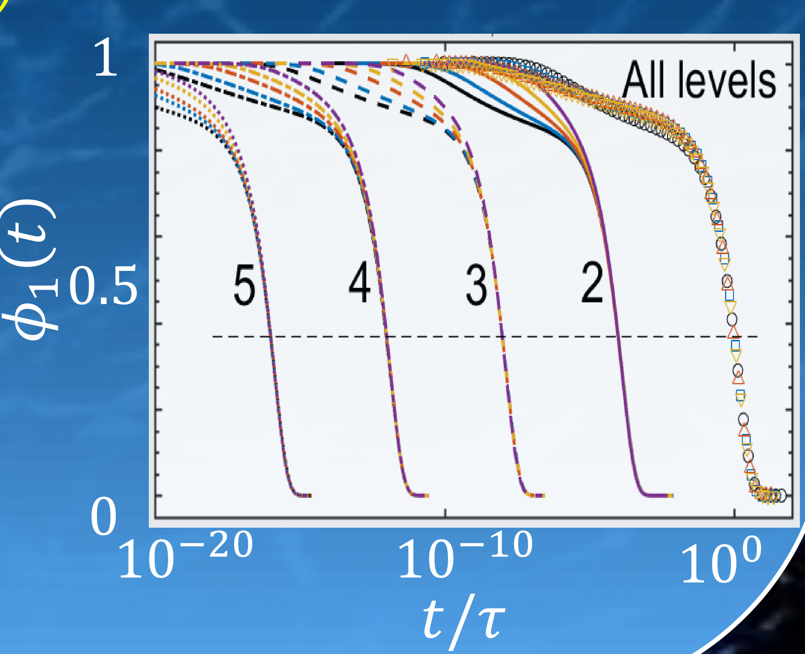
β scaling law:

$$\phi_1(k,t) = f^c(k) + \sqrt{\epsilon}\, g\left(k, \frac{t}{\tau_\beta}\right)$$

α scaling law:

$$\phi_1(k,t) = u\left(k, \frac{t}{\tau}\right) \quad \forall \epsilon$$





[1] Liesbeth M. C. Janssen and David R. Reichman. "Microscopic dynamics of supercooled liquids from first principles." Phys. Rev. Lett., 115, 205701 (2015)
[2] Liesbeth M. C. Janssen. "Mode-coupling theory of the glass transition: a primer." Front. Phys., 6, 97 (2018)