Onset of hierarchical relaxation and its connection with boson peak in 2D glassy system

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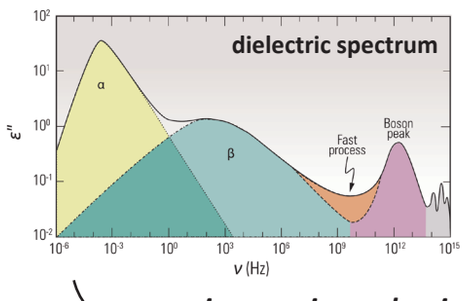
Introduction

Background

Glassy system exhibit ***hierarchical*** and heterogeneous relaxations when approaching ***glass transition***[2]:

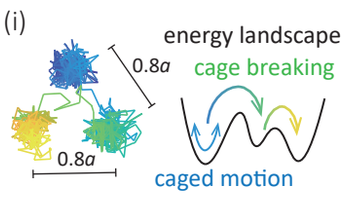


long-time α relaxation
short-time β relaxation
faster process

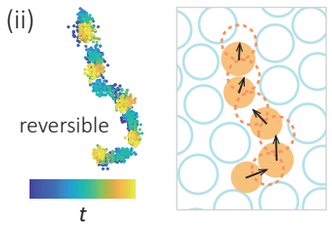
→ ***microscopic mechanism*** and origin of growing hierarchy with lower T ?

Difficulty

single-particle motion

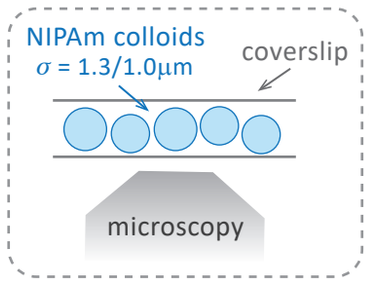


string-like cooperative motion



Methods *Colloidal experiment & Mode analysis method*

Research Process



Correlation matrix of displacement

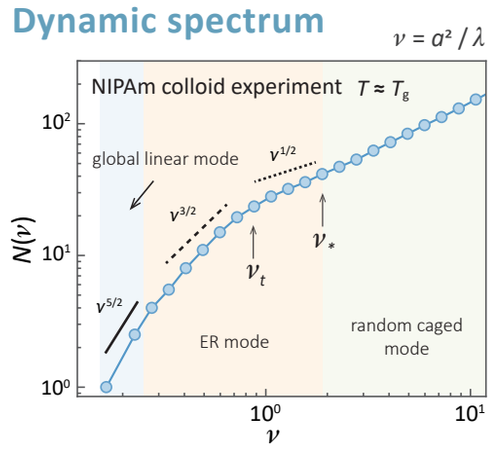
high-dimension version of correlation function:

$$D_{ij} = \langle (\mathbf{r}_i(t+\Delta t) - \mathbf{r}_i(t)) \cdot (\mathbf{r}_j(t+\Delta t) - \mathbf{r}_j(t)) \rangle_{t,t_a}$$

Diagonalizing matrix D

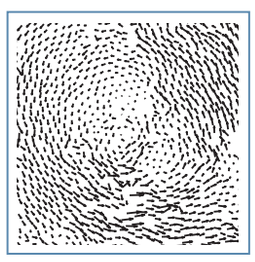
eigenvalue λ = amplitude of dynamic behaviors
eigenvector (***eigenmodes***) = correlation form of motion

Dynamic spectrum

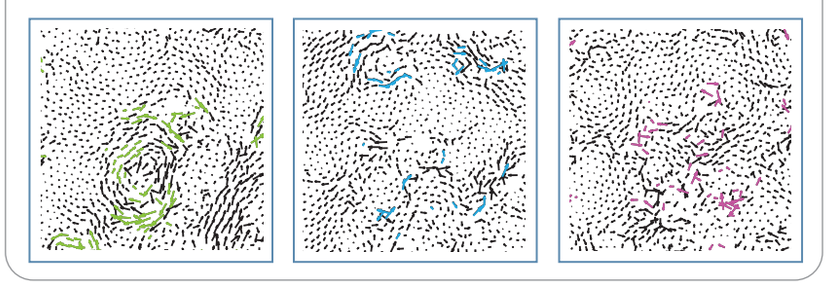


Dynamic eigenmodes

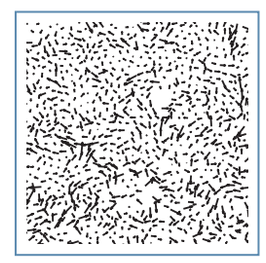
global linear mode
largest λ, phonon-like



elementary relaxation(ER) mode
large λ, spacially localized



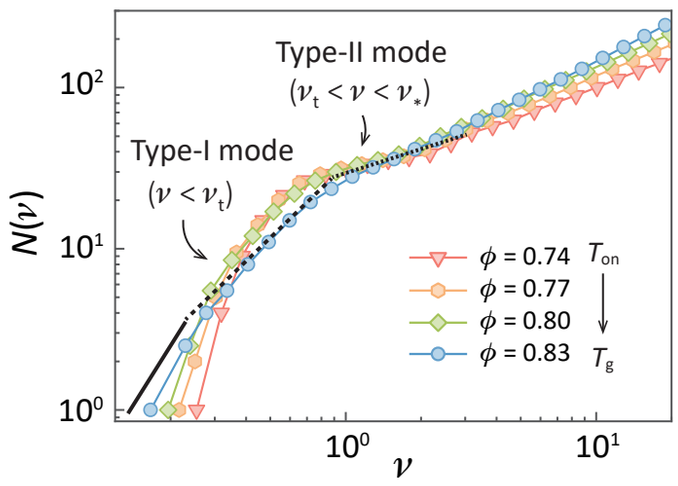
random caged mode
small λ, random



Results *Two types of elementary relaxation modes*

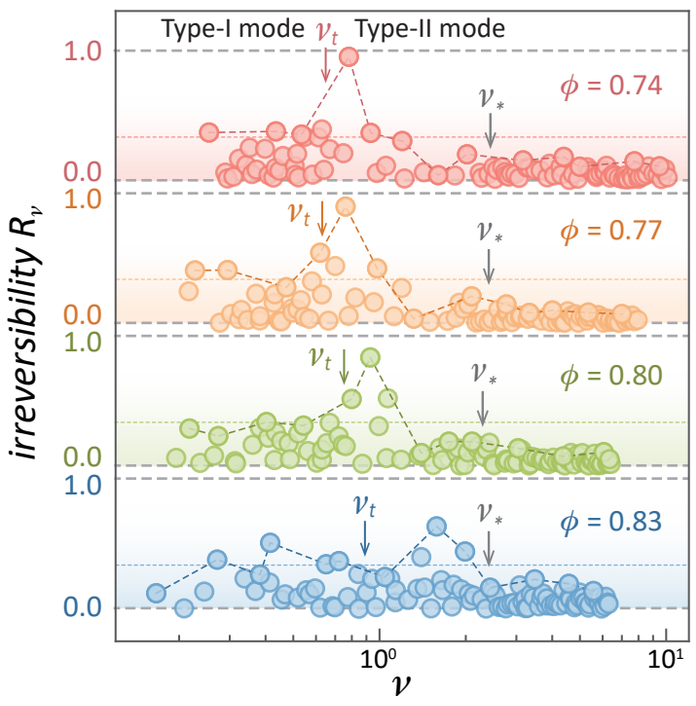
T-dependent spectrum

1. ***Convergence*** to vibrational spectrum at low T
2. Two types of ER modes separated by ν_t



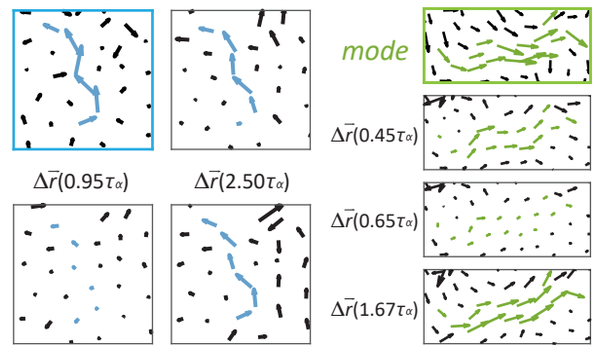
Reversibility of eigenmodes

Type-I mode: highly reversible - slow β relaxation
Type-II mode: irreversible - conventional flow
small-amplitude, reversible - fast β relaxation

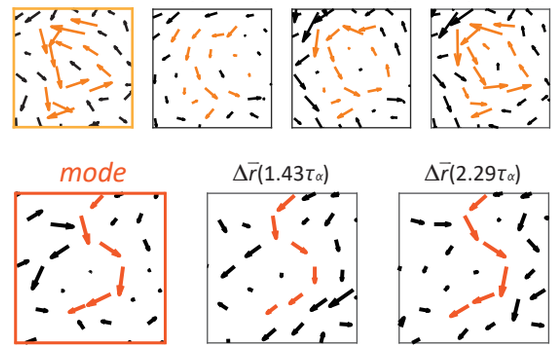


Real-space visualization of ER

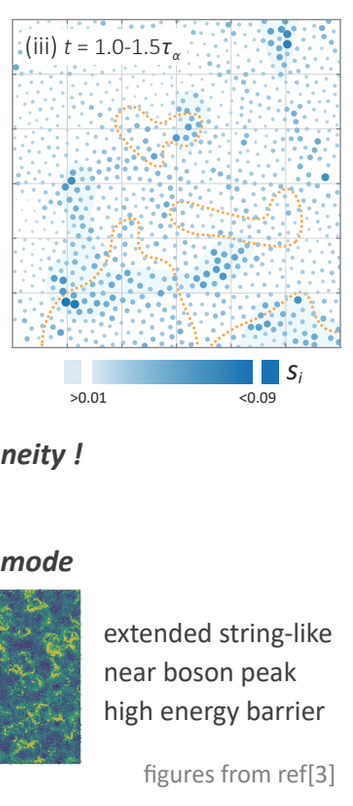
Type-I mode: reversible hopping motion



Type-II mode: unidirectional flow



dynamic facilitation (DF)



1. ER in Type-I modes accumulate through DF to become α diffusion (***hierarchy*** & ***heterogeneity***)
2. ER in Type-II modes serve as conventional flow like liquid (no such contribution)

Dynamic facilitation: a single relaxation can trigger another excitation in its vicinity →

Temperature-dependent dynamic features

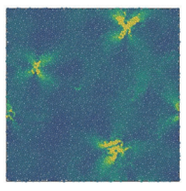
With lower temperature, Type-I modes dominate the relaxation dynamics → ***hierarchy and heterogeneity !***

Origin of ER connected to boson peak

boson peak: excess vibrational density of states over ω^{d-1}

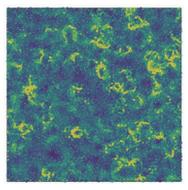
Diversity of ER originates from two types of dynamical 'defects' in vibrational modes

Type-I mode



T=0: quasi-localized
T>0: string-like
low frequency
low energy barrier

Type-II mode



extended string-like
near boson peak
high energy barrier

figures from ref[3]

Conclusion

We find two types of elementary relaxations and their origin:

	Relaxation mode	***Behaviors***	***Origin***	***Hierarchy & Heterogeneity***
Type-I mode	slow process	large-amplitude hopping	low-frequency QLM	Yes
Type-II mode	fast process	flow / fast reversible motion	string-like BP mode	No

References

[1] Corresponding manuscript. To be submitted.
[2] Chandler, D. & Garrahan, J. P. *Annu. review physical chemistry* **61**, 191–217 (2010).
[3] Hu, Y.-C. & Tanaka, H. *Nat. Phys.* **18**, 669–677 (2022).