Excitations, Emergent Facilitation and Glassy Dynamics in Supercooled Liquids

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A Dynamics-Based Perspective: Dynamical Facilitation Theory

DF theory (Chandler and Garrahan, *Annu. Rev. Phys. Chem*, 2010) can explain glassy dynamics **without knowledge of local structure** with two key ideas:

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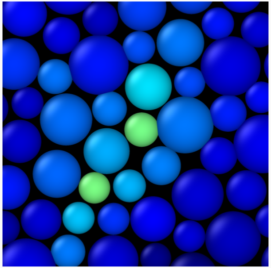

Energy Cost J_σ

Below the onset temperature T_o, localized excitations drive glassy dynamics! (Keys, et al. *Phys. Rev. X* 2011)

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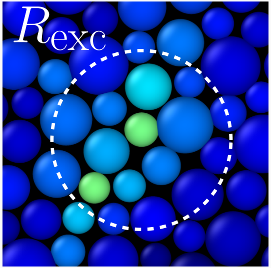

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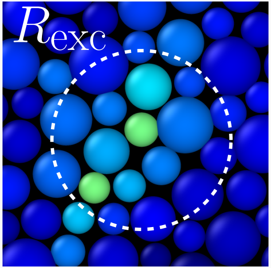

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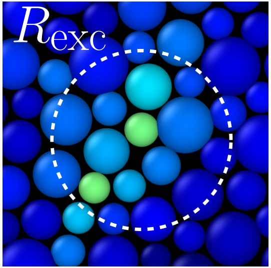

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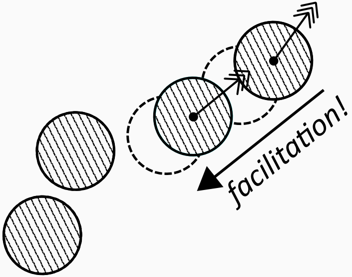

Excitations facilitate the creation and relaxation of another excitation close by.

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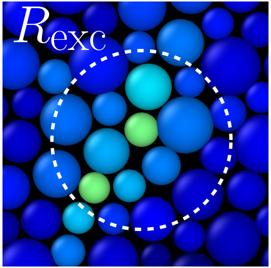

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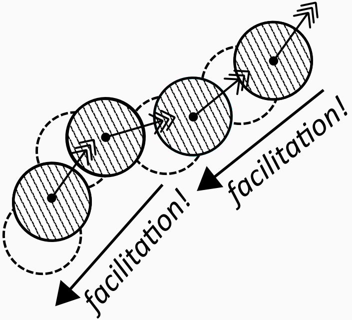

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A Theory of Onset Temperature in 2D

Inherent–State Melting and the Onset of Glassy Dynamics in Two-Dimensional Supercooled Liquids

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(Dated: April 18, 2022)

Below the onset temperature T_o, the equilibrium relaxation time of most glass-forming liquids exhibits glassy dynamics characterized by super-Arrhenius temperature dependence. In this supercooled regime, the relaxation dynamics also proceeds through localized elastic excitations corresponding to hopping events between inherent states, i.e., potential-energy minimizing configurations of the liquid. Despite its importance in distinguishing the supercooled regime from the high-temperature regime, the microscopic origin of T_o is not yet known. Here, we construct a theory for the onset temperature in two dimensions and find that inherent-state melting transition, described by the binding-unbinding transition of dipolar elastic excitations, delineates the supercooled regime from the high-temperature regime. The corresponding melting transition temperature is in good agreement with the onset temperature found in various two-dimensional atomistic models of glass formers. We discuss the predictions of our theory on the displacement and density correlations of two-dimensional supercooled liquids, which are consistent with observations of the Mermin-Wagner fluctuations in recent experiments and molecular simulations.

Keywords: Two-dimensional glassy dynamics, Kosterlitz-Thouless transition, excitations, geometric charges

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A Theory of Dynamical Facilitation: The Role of Revertibility

$\beta J_\sigma = 1$ (High-T)

Origin of Glassy Behavior

A combination of **emergent constraints** and **revertibility** of dynamics.

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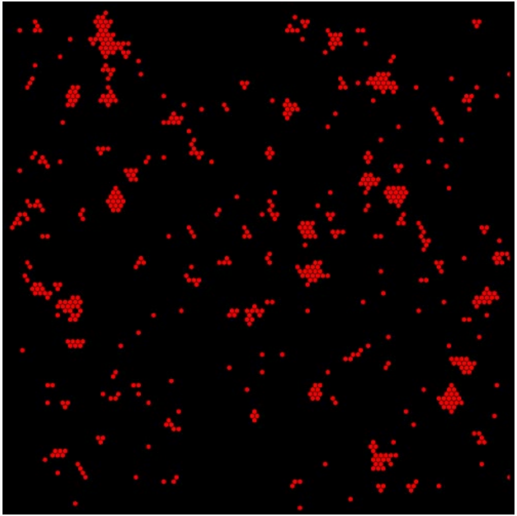

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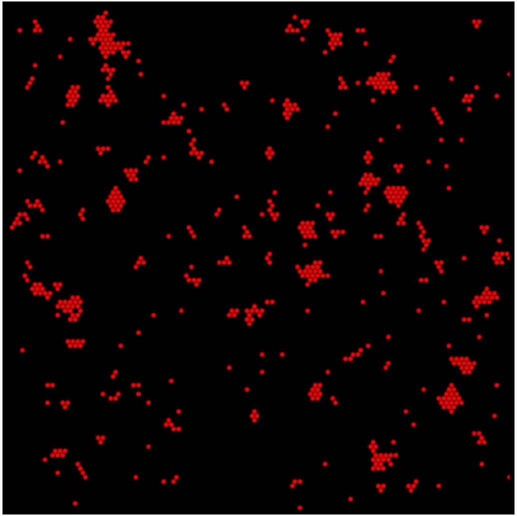

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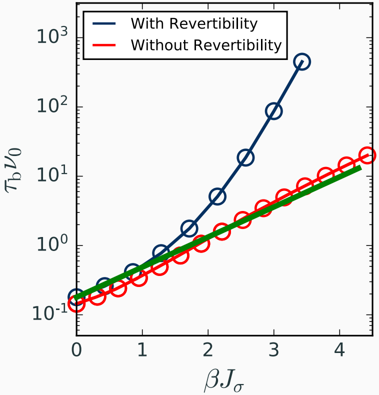

Relaxation time data when revertibility is turned off (red markers) along with an Arrhenius timescale $\tau_{\mathrm{arr}} \sim e^{\beta J_\sigma}$.

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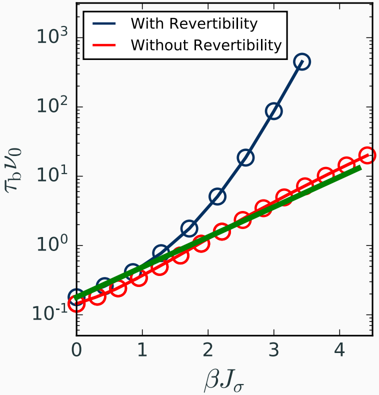

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No revertibility, no super-Arrhenius behavior.