Kinetic fragility directly correlates with the many-body static amorphous order in glass-forming liquids

Indrajit Tah and Smarajit Karmakar

Phys. Rev. Materials **6**, 035601 — Published 7 March 2022

DOI: 10.1103/PhysRevMaterials.6.035601

Kinetic Fragility Directly Correlates with the Many-body Static Amorphous Order in Glass-Forming Liquids

Indrajit Tah[1,2*] and Smarajit Karmakar[2†]
[1] *Department of Physics and Astronomy, University of Pennsylvania 209 South 33rd Street Philadelphia, PA, USA and*
[2] *Tata Institute of Fundamental Research, 36/P, Gopanpally Village,*
Serilingampally Mandal,Ranga Reddy District, Hyderabad, 500107, India

The term "fragility" describes the rate at which viscosity grows when a supercooled liquid approaches its putative glass transition temperature. The field of glassy materials is actively searching for a structural origin that governs this dynamical slowing down in the supercooled liquid, which occurs without any discernible change in structure. Our work shows clear evidence that growing many-body static amorphous order is intimately correlated with the kinetic fragility of glass-forming liquids. It confirms that the system's dynamical response to temperature is concealed in its microstructures. This finding may pave the way for a deeper understanding of the different temperature dependence of the relaxation time or viscosity in a wide variety of glass-forming liquids.

Introduction: The dramatic rise in viscosity or relaxation time upon supercooling is a universal hallmark feature across all glass-forming liquids. Despite extensive investigations [1–6], one of the fundamentally unsolved challenges in condensed matter physics is understanding of microscopic origin of rapid rise in viscosity (η) with relatively small temperature changes while approaching the calorimetric glass transition temperature (T_g), defined as the temperature at which η becomes $10^{14} Poise$. In this context, it's also worth noting that, while near-diverging growth of viscosity is universal in all glass-forming liquids, the rate at which viscosity grows at low temperatures is quite non-universal and varies significantly across liquids.

The term "fragility" was first coined in Ref. [7] to characterize this rapid non-universal changes in viscosity near T_g. Although, the word "fragility" is used to describe the dynamical properties, several theoretical and experimental studies [8–13] demonstrate that the fragility is fundamentally connected to thermodynamic properties of the liquid, like the excess entropy and the specific heat which are directly linked with the microscopic structure of a liquid. This connection led to a search for the structural or thermodynamic origin of fragility. However, the correlation between thermodynamics and dynamical properties in supercooled liquids remains somewhat controversial, leading to inherent uncertainty, which prevents researchers from reaching a consensus. In this article, we have addressed the connection between growing static amorphous order and fragility by performing extensive molecular dynamics simulations of a model glass-forming liquid, showing very large variation in fragility with increasing density.

The temperature dependence of viscosity and relaxation time for a wide variety of supercooled liquids can be well fitted by the Vogel-Fulcher-Tamann (VFT) formula [14–16]:

$$\tau_\alpha(T) = \tau_0 \exp\left[\frac{1}{K_{VFT}\ (T/T_{VFT} - 1)}\right], \qquad (1)$$

where τ_α is the structural relaxation time (defined later), τ_0 is the viscosity at infinite high temperature, T_{VFT} is the apparent divergence temperature for relaxation time and K_{VFT} denotes the "Kinetic fragility". The fragility index provides a unifying framework for the classification of a broad range of systems, from molecular liquids [7] to colloidal [17] and biological systems [18, 19]. Additionally, material functionality and manufacturability are directly linked with their fragility index. For example, the system that have low fragility index, generally has a wide glass transition temperature range which enhances the flexibility of material moulding. Moreover, material bulk properties depend on its molecular mechanisms and an important question is how the molecular mechanisms of glass forming liquids differ from each other in such a way that their dynamic properties vary so widely. Fragility also plays important role in bio-preservation[20–22]. Empirical evidence suggests that the larger the fragility of a liquid, the better it will be in preserving biomacromolecule when used as a medium. Although, there are deviations from this hypothesis but this remained as a rule of thumb in bio-preservation industry. Thus, a better understanding of fragility may lead to better understanding of the physics of biopreservation.

Models and Methods: We have performed extensive computer simulations of soft repulsive particles [23–25] by varying the density, (ρ), and temperature, (T), which cover a broad spectrum of fragility. This model (referred as Harmonic Potential model) shows a crossover from strong glass to fragile glass with varying density (ρ) beyond the jamming density (ρ_J) [25]. This provides a elegant way to tune fragility without changing the particle composition, interaction potential or curving the configurational space [26]. Simulations are done in three-dimensions in the density range $\rho \in [0.65, 0.82]$ with $N = 108000$ particles. More detailed information about the models and simulations is provided in SI [27]. For dynamical characterisations, we have computed the average relaxation timescale, τ_α, via the two-point overlap correlation function ($Q(t)$) (defined in the SI [27]) at each equilibrium density, ρ, and temperature, T state points. The relaxation time τ_α is defined as $\langle Q(\tau_\alpha)\rangle = 1/e$, where $\langle\cdots\rangle$ refers to ensemble average. The calorimetric glass transition temperature, T_g for this model in simulations is defined as $\tau_\alpha(T_g) = 5 \times 10^6$.

Results: Fig. 1 shows the Angell plot of the relaxation time (τ_α) as a function of rescaled inverse temperature. To have a proper comparison, the data has been normalized by respective T_g at different density. At low density the relaxation time exhibits Arrhenius dependence, indicating a behaviour of a strong glass-forming liquid, whereas at high density the relaxation time display super-Arrhenius temperature dependence, often referred as “Fragile glass-former”. The observed rise in fragility with increasing density is consistent with a previous theoretical model [28] in which increased fragility was linked to rising cooperative rearrangement volume (V_c). In these models, fragility can be tuned over a broad range, by only increasing the density This gives us an opportunity to look into the precise molecular mechanisms that cause such large change in fragility. In the inset of Fig. 1, we plot the fragility index (K_{VFT}) against density. One can see that increasing the bulk density by a factor of 1.26 ($\rho = 0.65$ to 0.82) changes the kinetic fragility by a factor of 17 ($K_{VFT} = 0.0295$ to 0.5002). Thus this model is an ideal test-bed for deciphering the microscopic origin of fragility and its possible connection to growing amorphous order as envisaged in Random First Order Transition (RFOT) Theory [29, 30]. In the subsequent paragraphs, we will discuss the strong correlation between changing fragility and the two important growing length scales in the systems; namely dynamic heterogeneity length scale (ξ_d) and static amorphous order length scale (ξ_s).

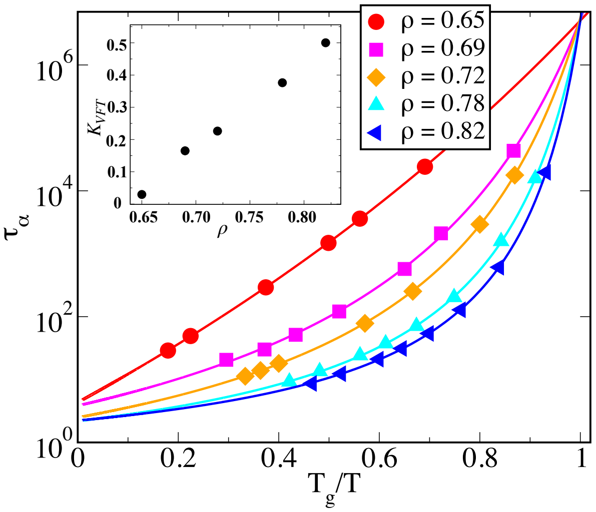

FIG. 1. Angell plot of (τ_α) as a function of rescaled temperature, T_g/T at different densities (ρ). At higher density the relaxation time exhibits a sharp growth upon decreasing temperature, a feature of a fragile glass-former, while at lower density the relaxation time displays Arrhenius behaviour with temperature, suggesting a crossover to strong glass-former. Lines are the fit to VFT form. Inset shows the fragility index against density.

Dynamic Length Scale: Since, fragility is measured from dynamical properties like relaxation time, it is natural to investigate the temperature dependence of ξ_d for these model systems and try to understand a possible correlation between them. Recent work [31] pointed out that growth of dynamic heterogeneity (DH) in strong liquids is different than in fragile liquids. Thus, a systematic study of DH [32] in this current set up is indeed warranted to have a detailed understanding of how heterogeneity gets affected by changing fragility in the system. DH can be quantified via the fluctuation of Q(t) [33] and the length scale associated with DH, can be measured from the spatial correlation of four-point structure factor (fluctuation of two point correlation function in Fourier space, $S_4(q, t = \tau_\alpha)$) [34, 35]. In SI [27], the determination of ξ_d from $S_4(q, t = \tau_\alpha)$ has been elaborated. Recently, in Ref. [36], another method of extracting dynamic length scale has been proposed. In this method the dynamical properties of the systems is measured at a smaller sub volume of the systems by dividing the whole systems in to smaller blocks of length, L_B. This method can be easily implemented both in experiments and in numerical simulations and termed as “block analysis” method. The method has also been shown to significantly improve the statistical averaging of the data as well as include all possible fluctuations (*e.g* density, temperature, composition, *etc.*) that are important in measuring four-point correlation functions (see SI [27] for further details).

In Fig. 2(a), we show a comparison of ξ_d obtained using two different methods for systems which reside on the two extreme ends of the spectrum in the Angell plot; first one being the most fragile liquid with $\rho = 0.82$ (see Fig. 2(a)), while the other is at the strong liquid end (Fig. 2(a) inset) with $\rho = 0.65$. The quite good agreement between these two ways of estimations of ξ_d for all state points provides us confidence on the measured heterogeneity length scale. An interesting question that naturally comes up is the following: if the two extreme systems (strong and fragile) have very similar structural relaxation times, do they have similar dynamic heterogeneity? To investigate this we plot χ_4 as a function of time in Fig. 2(b) for strong and fragile liquid (τ_α for these two systems are close to each other) at roughly the same block size (L_B). We find that fragile liquid show stronger dynamic heterogeneity than strong liquid for a given block size. In the inset of Fig. 2(b), we plot the peak value of $\chi_4(t)$ as a function of L_B for these two extreme systems at the same τ_α. Fragile liquid shows stronger dynamic heterogeneity at each length scale than the strong glass-former. This observation is in stark contrast with a recent finding [37] of possible decoupling of relaxation time and dynamic heterogeneity in active glass-forming

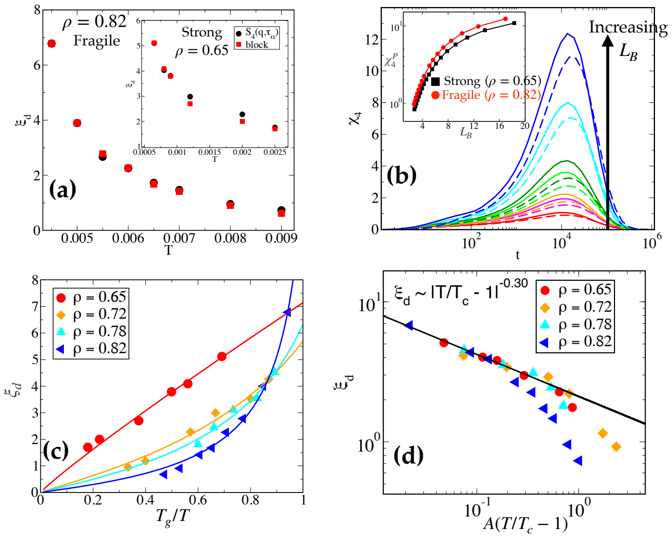

FIG. 2. **(a).** Dynamics length scale as a function of temperature for fragile liquid. The legend shows the method of extraction of length scale. Similar plot for strong glass forming liquid is shown in the inset. **(b).** χ_4 as a function of time for various block size (L_B). Fragile liquids ($\rho = 0.82$, solid lines), shows stronger growth of dynamic heterogeneity compare to strong liquids ($\rho = 0.65$, dotted lines). We display $\chi_4(t)$ for the strong and fragile liquids at roughly the same block scale. The maximum of $\chi_4(t)$ (χ_4^P) for fragile and solid liquids at the same relaxation time (τ_α) is shown in the inset as a function of block size (L_B). **(c).** Dynamic length scales as a function of rescaled temperature by T_g. Solid lines are fits to the form $\xi_d \sim |T/T_{VFT} - 1|^{-\nu}$. **(d).** Dynamic length scales are plotted as a function of $|T/T_c - 1|$, where T_c is the MCT divergence temperature. The length scale data are rescaled to make it more comparable to other data sets. The black line is a power fit with an exponent $\nu = 0.30$ (see text for discussion).

liquids. Next we focus on the temperature dependence of ξ_d with changing fragility. Fig. 2(c) shows the growth of dynamic length scale as a function T_g/T. It can be seen that for the fragile glass former, ξ_d shows a sharp growth upon supercooling, whereas strong glass former produces a gentle growth resembling the Angell like plot for τ_α (see Fig.1).

Inhomogeneous mode coupling theory (IMCT) [38] predicts that three-point density correlation function, $\chi_3(t)$, which can be obtained by measuring the response of the system under an external perturbation, is intimately related to the four-point susceptibility $\chi_4(t)$ and shows similarly scaling behaviour near the MCT transition temperature (T_c). Thus, according to IMCT, ξ_d should have a critical like behaviour: as $\xi_d \sim |T - T_c|^{-\nu}$ with $\nu = 1/4$ being the predicted critical exponent. Recently, Tah et.al, [39] showed for few model glass forming liquids that the exponent ν is in agreement with IMCT prediction for temperature near T_c. However, validity of this result across a wide range of systems with changing fragility is not studied before. In Fig. 2(d) we show the dependence of ξ_d vs $(T - T_c)/T_c$ for different fragile systems. The black line shows the power law fit to the few low temperature data points. We find for all the different fragile systems the value of the exponent is ~ 0.30 (near T_c) which is not very different from the exponent $\nu = 0.25$ expected by IMCT. The small difference in exponent may be attributable to the finite dimensions, and understanding how the exponent shifts in higher dimensions (e.g. near the upper critical dimension $d_u = 8$), will be of considerable interest[40–43].

Amorphous order and Fragility: The lack of a priori knowledge of the nature of the structural order in disorder glass forming liquids makes it difficult to measure the relevant degree of order. However, existing results show that the system has domains of different mobility near the glass transition temperature and these domains have relaxation rates that are substantially faster or slower than the average relaxation rate. These different domains often called as "cooperatively rearranging regions (CRRs)", are the primary building blocks of the phenomenological Adam-Gibbs theory [44] of glass transition and its subsequent development, i.e., the random first-order transition (RFOT) theory [29, 30]. Different mobility domains have different patch entropy, and the mean patch correlation length can be easily extracted from the largest groups of congruent patches which are considered to have a similar local order [45]. This hints that the origins of heterogeneous dynamics may be buried in the local amorphous structure. In [46] a non-trivial correlation function known as point-to-set (PTS) correlation [47] is proposed to calculate the structural or thermodynamic correlation length scale in supercooled glass-forming liquids in an order-agnostic way as envisaged in RFOT theory. Growth of this static length scale gives a notion of emerging thermodynamic order that may be connected to the dynamical slowing down of the system, even when traditional structural features (e.g. "pair correlation function") are blind to capture the dramatic slowing down of the relaxation time. Here, we address the crucial question of whether the growing thermodynamic amorphous order can universally explain the origin of a wide spectrum of fragility.

To address this, we have computed ξ_s using point-to-set correlation function in cavity geometry (see the SI [27]) [4, 47–50]. Fig. 3(a) and 3(b) show the nature of the correlation function with increasing cavity radius, R for various studied temperatures for a strong and a fragile liquid. The correlation functions for the strong liquid decay at the same rate for all temperatures, indicating no growth in the amorphous order (see Fig. 3(a)) with decreasing temperature. However, for fragile liquid these correlation functions show significantly slower decay as a function of R with decreasing temperature. ξ_s can be obtained by simply fitting the correlation functions (see SI for details). We also extracted ξ_s using block analysis

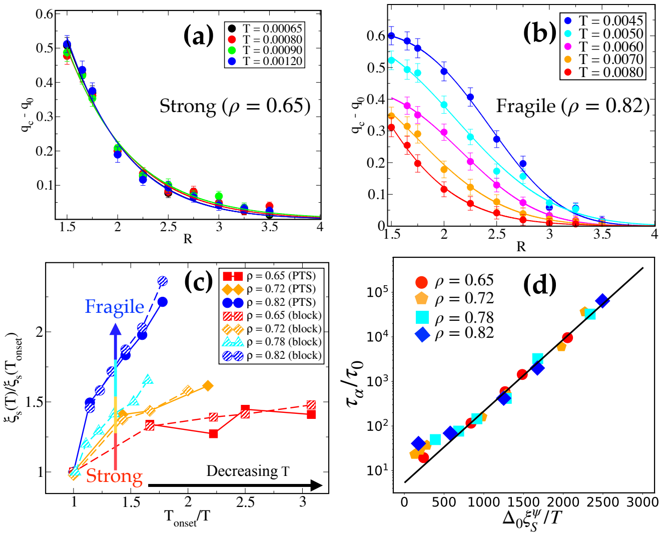

FIG. 3. Overlap correlation as a function of cavity radius R for strong glass former **(a)** and fragile glass former **(b)** at various temperatures. **(c)** Rescaled static length scale (ξ_s) as a function of reduced temperature (re-scaled by onset temperature (T_{onset})). Here, $\xi_s(T_{onset})$ is the value of static length scale at onset temperature. **(d)** Rescaled relaxation time (τ_α/τ_0) as a function of ξ_s^ψ/T yielded a good data collapse for all the different fragile liquids.

of relaxation time as proposed in Ref. [36] (see SI [27] for details). The temperature dependence of the length scale for different fragile liquids determined by point-to-set method and block analysis method are shown and compared in Fig. 3(c). We find that the length scales obtained by these two methods are very similar for all the studied systems. For better representation, we rescaled ξ_s by its value at onset temperature (temperature at which the thermodynamic and dynamic properties begin to depart from its high temperature behaviour [51]) and show the data only for temperatures below the onset temperature (see SI). For strong glass forming liquid ξ_s ("red squares") does not grow much with decreasing temperature, but for fragile glass forming liquid it ("blue circles") grows sharply as system approaches the glass transition temperature.

Within the RFOT scenario, the structural relaxation time of a system is related to ξ_s as

$$\tau_\alpha(T) = \tau_0 \exp\left[\frac{\Delta_0\ \xi_s^\psi(T)}{T}\right], \tag{2}$$

where typical free energy barrier is $\Delta(T) = \Delta_0\ \xi_s^\psi(T)$ with ψ being an apriori unknown scaling exponent. Δ_0 and τ_0 set the energy scale and microscopic time scale of the system. Discussion regarding causal relation between ξ_s and τ_α is elaborated in SI. Now, according to Fig. 3(c), the typical free energy barrier for a strong glass former remains nearly constant as ξ_s (red squares) stays nearly constant, but rises (blue circles) sharply for a fragile liquid as the temperature is lowered towards T_g, resulting in super-Arrhenius behaviour. We believe this is the first numerical evidence that conclusively suggest that ξ_s controls the relaxation process including the drastic change in fragility with changing density in supercooled liquids. We also believe the above results are very general and constitute a first clear demonstration that order agnostic thermodynamic order (applicable to all generic glass formers) is strongly correlated to fragility, rather than some specific locally favored structure (LFS) [52–54]. This result supports the previous theoretical model [28] by demonstrating that the rise in fragility with increasing density is linked to the increase in amorphous order.

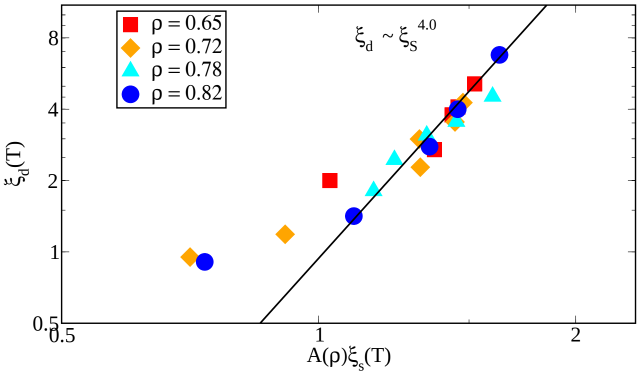

FIG. 4. Dynamic length scale (ξ_d) as a function of static length scale (ξ_s) for all different degrees of fragile liquids. $A(\rho)$ is a density dependent scale parameter to bring all the data together and to highlight their universal relationship.

Now, if τ_α is solely depended on the thermodynamics length scales as given in Eq.2, then one should be able to collapse all the data of relaxation time for all the systems with varying fragility on to a master curve by plotting against ξ_s^ψ/T. To validate the above argument we plotted the re-scaled relaxation time (τ_α/τ_0) (re-scaled by time scale (τ_0) at very high temperature where $\xi_s \to 0$) as a function of $\Delta_0\xi_s^\psi(T)/T$ and find that data for all the systems with different fragility collapses on to a straight line as shown in Fig. 3 (d). We keep the exponent $\psi = 2.6$ same while varying Δ_0. Good data collapse implies an encouraging universality between the relaxation time and the static length scale across systems with large change in fragility. Encouraged by this finding, we investigate if DH [55–57] is linked to the underlying microscopic structure, i.e. whether static and dynamic length scales are universally connected. We plot ξ_d against ξ_s in Fig. 4 for all the fragile liquids and find that ξ_d has a universal relationship with ξ_s, as $\xi_d \sim \xi_s^4$ at least for the studied model system. Universality of this power-law relationship across various model systems can not be ascertained from this study alone.

Conclusion: Taken together, our findings show that by fine-tuning the system's packing fraction, one might achieve a bigger change in kinetic fragility in soft repulsive systems for delving deeper into the relationship between their kinetic fragility and the microscopic structural ordering. This could be easily tested experimentally by using colloidal suspensions [58]. The heterogeneity length scale for fragile liquid increases significantly faster as the system approaches the glass transition temperature, whereas strong liquid shows much smaller increase. All liquids show MCT critical like behavior near the MCT temperature (T_c), with an exponent $\nu = 0.30$ that is close to the exponent anticipated by IMCT theory. We show that the distinct temperature dependence of the order agnostic thermodynamics length scale controls the temperature dependence of the structural relaxation time for all the fragile liquids indicting that widely different temperature dependency of structural relaxation in various glass-forming liquids is simply concealed in the local amorphous order, whose growth determines whether a system will be strong to fragile glass formers. Since the material durability is an intrinsic property that is tightly linked to its fragility index, we expect that our findings will be useful to better understand the role of microscopic structure in the emergence of fragility in future. Finally, our observation that dynamic length scale is universally related to static length scale also hints that *probably* static length scale is the sole player in the physics of glass transition.

IT thanks S. Ridout and A. J. Liu for discussions. IT wants to acknowledge use of Extreme Science and Engineering Discovery Environment (XSEDE) [59], which is supported by National Science Foundation grant number TG-BIO200091. SK would like to acknowledge funding by intramural funds at TIFR Hyderabad from the Department of Atomic Energy (DAE). Support from Swarna Jayanti Fellowship grants DST/SJF/PSA-01/2018-19 and SB/SFJ/2019-20/05 are also acknowledged

---

[*] itah@sas.upenn.edu

[†] smarajit@tifrh.res.in

[1] L. Berthier, G. Biroli, J.-P. Bouchaud, L. Cipelletti, D. E. Masri, D. L'Hôte, F. Ladieu, and M. Pierno, "Direct experimental evidence of a growing length scale accompanying the glass transition," *Science*, vol. 310, no. 5755, pp. 1797–1800, 2005.

[2] L. Berthier and G. Biroli, "Theoretical perspective on the glass transition and amorphous materials," *Rev. Mod. Phys.*, vol. 83, pp. 587–645, Jun 2011.

[3] S. Karmakar, C. Dasgupta, and S. Sastry, "Growing length scales and their relation to timescales in glass-forming liquids," *Annual Review of Condensed Matter Physics*, vol. 5, no. 1, pp. 255–284, 2014.

[4] I. Tah, S. Sengupta, S. Sastry, C. Dasgupta, and S. Karmakar, "Glass transition in supercooled liquids with medium-range crystalline order," *Phys. Rev. Lett.*, vol. 121, p. 085703, Aug 2018.

[5] R. Das, I. Tah, and S. Karmakar, "Possible universal relation between short time β-relaxation and long time α-relaxation in glass-forming liquids," *The Journal of Chemical Physics*, vol. 149, no. 2, p. 024501, 2018.

[6] I. Tah, A. Mutneja, and S. Karmakar, "Understanding slow and heterogeneous dynamics in model supercooled glass-forming liquids," *ACS Omega*, vol. 6, pp. 7229–7239, 03 2021.

[7] C. A. Angell, "Formation of glasses from liquids and biopolymers," *Science*, vol. 267, no. 5206, pp. 1924–1935, 1995.

[8] S. Sastry, P. G. Debenedetti, and F. H. Stillinger, "Signatures of distinct dynamical regimes in the energy landscape of a glass-forming liquid," *Nature*, vol. 393, no. 6685, pp. 554–557, 1998.

[9] L. M. Martinez and C. A. Angell, "A thermodynamic connection to the fragility of glass-forming liquids," *Nature*, vol. 410, no. 6829, pp. 663–667, 2001.

[10] K. Ito, C. T. Moynihan, and C. A. Angell, "Thermodynamic determination of fragility in liquids and a fragile-to-strong liquid transition in water," *Nature*, vol. 398, no. 6727, pp. 492–495, 1999.

[11] P. G. Debenedetti and F. H. Stillinger, "Supercooled liquids and the glass transition," *Nature*, vol. 410, no. 6825, pp. 259–267, 2001.

[12] N. A. Mauro, M. Blodgett, M. L. Johnson, A. J. Vogt, and K. F. Kelton, "A structural signature of liquid fragility," *Nature Communications*, vol. 5, no. 1, p. 4616, 2014.

[13] C. Dalle-Ferrier, A. Kisliuk, L. Hong, G. Carini, G. Carini, G. DâĂŹAngelo, C. Alba-Simionesco, V. N. Novikov, and A. P. Sokolov, "Why many polymers are so fragile: A new perspective," *The Journal of Chemical Physics*, vol. 145, no. 15, p. 154901, 2016.

[14] H. VOGEL, "Das temperaturabhangigkeitsgesetz der viskositat von flussigkeiten," *Phys. Z.*, vol. 22, pp. 645–646, 1921.

[15] G. S. Fulcher, "Analysis of recent measurements of the viscosity of glasses," *Journal of the American Ceramic Society*, vol. 8, no. 6, pp. 339–355, 1925.

[16] G. Tammann and W. Hesse, "Die abhÃďngigkeit der viscositÃďt von der temperatur bie unterkÃijhlten flÃijssigkeiten," *Zeitschrift fÃijr anorganische und allgemeine Chemie*, vol. 156, no. 1, pp. 245–257, 1926.

[17] J. Mattsson, H. M. Wyss, A. Fernandez-Nieves, K. Miyazaki, Z. Hu, D. R. Reichman, and D. A. Weitz, "Soft colloids make strong glasses," *Nature*, vol. 462, no. 7269, pp. 83–86, 2009.

[18] D. M. Sussman, M. Paoluzzi, M. C. Marchetti, and M. L. Manning, "Anomalous glassy dynamics in simple models of dense biological tissue," *EPL (Europhysics Letters)*, vol. 121, p. 36001, feb 2018.

[19] I. Tah, T. A. Sharp, A. J. Liu, and D. M. Sussman, "Quantifying the link between local structure and cellular rearrangements using information in models of biological tissues," *Soft Matter*, 2021.

[20] K. Fox, "Biopreservation. putting proteins under glass," *Science*, vol. 267, no. 5206, pp. 1922–1923, 1995.

[21] Y. ROOS and M. KAREL, "Plasticizing effect of water on thermal behavior and crystallization of amorphous food models," *Journal of Food Science*, vol. 56, no. 1, pp. 38–

43, 1991.

[22] M. Mukherjee, J. Mondal, and S. Karmakar, “Role of α and β relaxations in collapsing dynamics of a polymer chain in supercooled glass-forming liquid,” *The Journal of Chemical Physics*, vol. 150, no. 11, p. 114503, 2019.

[23] C. S. O’Hern, S. A. Langer, A. J. Liu, and S. R. Nagel, “Random packings of frictionless particles,” *Phys. Rev. Lett.*, vol. 88, p. 075507, Jan 2002.

[24] A. J. Liu and S. R. Nagel, “The jamming transition and the marginally jammed solid,” *Annual Review of Condensed Matter Physics*, vol. 1, no. 1, pp. 347–369, 2010.

[25] L. Berthier and T. A. Witten, “Compressing nearly hard sphere fluids increases glass fragility,” *EPL (Europhysics Letters)*, vol. 86, p. 10001, apr 2009.

[26] F. m. c. Sausset, G. Tarjus, and P. Viot, “Tuning the fragility of a glass-forming liquid by curving space,” *Phys. Rev. Lett.*, vol. 101, p. 155701, Oct 2008.

[27] “See supplemental material at [url will be inserted by publisher] for additional detail on our methodologies, which includes refs,”

[28] J. Krausser, K. H. Samwer, and A. Zaccone, “Interatomic repulsion softness directly controls the fragility of supercooled metallic melts,” *Proceedings of the National Academy of Sciences*, vol. 112, no. 45, pp. 13762–13767, 2015.

[29] T. R. Kirkpatrick, D. Thirumalai, and P. G. Wolynes, “Scaling concepts for the dynamics of viscous liquids near an ideal glassy state,” *Phys. Rev. A*, vol. 40, pp. 1045–1054, Jul 1989.

[30] V. Lubchenko and P. G. Wolynes, “Theory of structural glasses and supercooled liquids,” *Annual Review of Physical Chemistry*, vol. 58, no. 1, pp. 235–266, 2007. PMID: 17067282.

[31] H. Staley, E. Flenner, and G. Szamel, “Reduced strength and extent of dynamic heterogeneity in a strong glass former as compared to fragile glass formers,” *The Journal of Chemical Physics*, vol. 143, no. 24, p. 244501, 2015.

[32] M. D. Ediger, “Spatially heterogeneous dynamics in supercooled liquids,” *Annual Review of Physical Chemistry*, vol. 51, no. 1, pp. 99–128, 2000. PMID: 11031277.

[33] N. Lačević, F. W. Starr, T. B. Schrøder, V. N. Novikov, and S. C. Glotzer, “Growing correlation length on cooling below the onset of caging in a simulated glass-forming liquid,” *Phys. Rev. E*, vol. 66, p. 030101, Sep 2002.

[34] R. Yamamoto and A. Onuki, “Dynamics of highly supercooled liquids: Heterogeneity, rheology, and diffusion,” *Phys. Rev. E*, vol. 58, pp. 3515–3529, Sep 1998.

[35] N. LaÄŊeviÄĞ, F. W. Starr, T. B. SchrÃÿder, and S. C. Glotzer, “Spatially heterogeneous dynamics investigated via a time-dependent four-point density correlation function,” *The Journal of Chemical Physics*, vol. 119, no. 14, pp. 7372–7387, 2003.

[36] S. Chakrabarty, I. Tah, S. Karmakar, and C. Dasgupta, “Block analysis for the calculation of dynamic and static length scales in glass-forming liquids,” *Phys. Rev. Lett.*, vol. 119, p. 205502, Nov 2017.

[37] K. Paul, S. K. Nandi, and S. Karmakar, “Dynamic heterogeneity in active glass-forming liquids is qualitatively different compared to its equilibrium behaviour,” 2021.

[38] G. Biroli, J.-P. Bouchaud, K. Miyazaki, and D. R. Reichman, “Inhomogeneous mode-coupling theory and growing dynamic length in supercooled liquids,” *Phys. Rev. Lett.*, vol. 97, p. 195701, Nov 2006.

[39] I. Tah and S. Karmakar, “Signature of dynamical heterogeneity in spatial correlations of particle displacement and its temporal evolution in supercooled liquids,” *Phys. Rev. Research*, vol. 2, p. 022067, Jun 2020.

[40] P. Charbonneau, A. Ikeda, J. A. van Meel, and K. Miyazaki, “Numerical and theoretical study of a monodisperse hard-sphere glass former,” *Phys. Rev. E*, vol. 81, p. 040501, Apr 2010.

[41] P. Charbonneau, A. Ikeda, G. Parisi, and F. Zamponi, “Dimensional study of the caging order parameter at the glass transition,” *Proceedings of the National Academy of Sciences*, vol. 109, no. 35, pp. 13939–13943, 2012.

[42] S. Franz, H. Jacquin, G. Parisi, P. Urbani, and F. Zamponi, “Quantitative field theory of the glass transition,” *Proceedings of the National Academy of Sciences*, vol. 109, no. 46, pp. 18725–18730, 2012.

[43] M. Adhikari, S. Karmakar, and S. Sastry, “Spatial dimensionality dependence of heterogeneity, breakdown of the stokesâĂŞeinstein relation, and fragility of a model glass-forming liquid,” *The Journal of Physical Chemistry B*, vol. 125, no. 36, pp. 10232–10239, 2021. PMID: 34494429.

[44] G. Adam and J. H. Gibbs, “On the Temperature Dependence of Cooperative Relaxation Properties in Glass-Forming Liquids,” *J. Chem. Phys.*, vol. 43, pp. 139–146, July 1965.

[45] F. m. c. Sausset and D. Levine, “Characterizing order in amorphous systems,” *Phys. Rev. Lett.*, vol. 107, p. 045501, Jul 2011.

[46] J.-P. Bouchaud and G. Biroli, “On the adam-gibbs-kirkpatrick-thirumalai-wolynes scenario for the viscosity increase in glasses,” *The Journal of Chemical Physics*, vol. 121, no. 15, pp. 7347–7354, 2004.

[47] G. Biroli, J. P. Bouchaud, A. Cavagna, T. S. Grigera, and P. Verrocchio, “Thermodynamic signature of growing amorphous order in glass-forming liquids,” *Nature Physics*, vol. 4, no. 10, pp. 771–775, 2008.

[48] G. M. Hocky, T. E. Markland, and D. R. Reichman, “Growing point-to-set length scale correlates with growing relaxation times in model supercooled liquids,” *Phys. Rev. Lett.*, vol. 108, p. 225506, Jun 2012.

[49] K. Hima Nagamanasa, S. Gokhale, A. K. Sood, and R. Ganapathy, “Direct measurements of growing amorphous order and non-monotonic dynamic correlations in a colloidal glass-former,” *Nature Physics*, vol. 11, no. 5, pp. 403–408, 2015.

[50] S. Yaida, L. Berthier, P. Charbonneau, and G. Tarjus, “Point-to-set lengths, local structure, and glassiness,” *Phys. Rev. E*, vol. 94, p. 032605, Sep 2016.

[51] S. Sastry, “Onset temperature of slow dynamics in glass forming liquids,” *PhysChemComm*, vol. 3, pp. 79–83, 2000.

[52] D. Coslovich and G. Pastore, “Understanding fragility in supercooled lennard-jones mixtures. i. locally preferred structures,” *The Journal of Chemical Physics*, vol. 127, no. 12, p. 124504, 2007.

[53] J. E. Hallett, F. Turci, and C. P. Royall, “Local structure in deeply supercooled liquids exhibits growing lengthscales and dynamical correlations,” *Nature Communications*, vol. 9, no. 1, p. 3272, 2018.

[54] H. Tong and H. Tanaka, “Revealing hidden structural order controlling both fast and slow glassy dynamics in supercooled liquids,” *Phys. Rev. X*, vol. 8, p. 011041, Mar 2018.

[55] T. Kawasaki and H. Tanaka, “Structural signature of slow dynamics and dynamic heterogeneity in two-

dimensional colloidal liquids: glassy structural order," *Journal of Physics: Condensed Matter*, vol. 23, p. 194121, apr 2011.

[56] H. Tong and H. Tanaka, "Structural order as a genuine control parameter of dynamics in simple glass formers," *Nature Communications*, vol. 10, no. 1, p. 5596, 2019.

[57] H. Tanaka, H. Tong, R. Shi, and J. Russo, "Revealing key structural features hidden in liquids and glasses," *Nature Reviews Physics*, vol. 1, no. 5, pp. 333–348, 2019.

[58] J. Mattsson, H. M. Wyss, A. Fernandez-Nieves, K. Miyazaki, Z. Hu, D. R. Reichman, and D. A. Weitz, "Soft colloids make strong glasses," *Nature*, vol. 462, no. 7269, pp. 83–86, 2009.

[59] J. Towns, T. Cockerill, M. Dahan, I. Foster, K. Gaither, A. Grimshaw, V. Hazlewood, S. Lathrop, D. Lifka, G. D. Peterson, R. Roskies, J. R. Scott, and N. Wilkins-Diehr, "Xsede: Accelerating scientific discovery," *Computing in Science & Engineering*, vol. 16, pp. 62–74, Sept.-Oct. 2014.