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RAPID COMMUNICATION

A dynamic crossover with possibly universal dynamic signatures in simple glass-forming liquids

Yiming Zheng(郑一鸣)[1], Mingyu Zhu(朱明宇)[1], Licun Fu(付立存)[1], Pengfei Guan(管鹏飞)[2,3,†], and Lijin Wang(王利近)[1,‡]

[1] *School of Physics and Optoelectronic Engineering, Anhui University, Hefei* 230601, *China*
[2] *Advanced Interdisciplinary Science Research Center (AiRCenter), Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo* 315201, *China*
[3] *Beijing Computational Science Research Center, Beijing* 100193, *China*

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On approaching the glass transition, the structural relaxation of glass-forming liquids slows down drastically, along with a significant growth of dynamic heterogeneity. Recent studies have achieved substantial advancements in elucidating the quantitative correlations between structural relaxation and dynamic heterogeneity. Here, we present the discovery of a novel dynamic crossover with possibly universal dynamic signatures by investigating the relationship between structural relaxation and dynamic heterogeneity. Specifically, the structural relaxation time at the dynamic crossover τ_c is equal to the time scale for the maximum non-Gaussian parameter, which could serve as a quantitative characterization of dynamic heterogeneity. The degree of dynamic heterogeneity at the crossover is approximately equivalent across all investigated glass-forming liquids, leading to a scaling collapse between structural relaxation and dynamic heterogeneity. Moreover, the mean squared displacement at the structural relaxation time is nearly constant across different temperatures as long as the structural relaxation time does not exceed τ_c. We further observe that the temperature at the dynamic crossover is lower than the onset temperature of slow dynamics. Our findings thus suggest the existence of a novel dynamic crossover with possibly universal dynamic signatures in glass-forming liquids, which merits in-depth investigations.

Keywords: glass-forming liquids, dynamic heterogeneity, structural relaxation, glassy dynamics

PACS: 61.20.Lc, 64.70.Q–, 71.55.Jv

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1. Introduction

Glasses are ubiquitous in nature and technology. However, the nature of glasses remains one of the most challenging topics.[1,2] Upon rapid cooling or compression, nearly all liquids can undergo a transformation to the glassy state, accompanied by a dramatic slowdown of dynamics. Although the essence of the glass transition remains an unresolved issue, it is widely acknowledged that glass-forming liquids often display remarkable dynamics that distinguishes them from simple liquids.[3–15] In particular, as they approach the glass transition, glass-forming liquids typically undergo complex relaxation processes. Substantial evidence suggests that the dynamics of relaxation and diffusion in liquids at high temperatures follows an Arrhenius temperature dependence. In contrast, at low temperatures, most glass-forming liquids exhibit super-Arrhenius behavior. The terminology fragility[16] $m = \partial(\log\tau)/\partial(T_g/T)|_{T=T_g}$, with T_g and τ being the glass transition temperature and structural relaxation time, respectively, has been introduced to quantify to what extent the dynamics deviates from the Arrhenius behavior. Specifically, more fragile glass-forming liquids exhibit a steeper super-Arrhenius increase with decreasing temperatures, and hence a larger value of m.

During the past few decades, substantial experimental and theoretical efforts have unveiled a new dimension of relaxation behavior in glass-forming liquids, known as dynamic heterogeneity,[2,3,15,17,18] which pertains to the spatio-temporal fluctuations of local dynamics. Interestingly, dynamic heterogeneity, as well as other dynamic features such as the two-step relaxation observed in glass-forming liquids, may remain detectable even in the glassy state,[19–22] although the manner in which these features are characterized differs between glasses and glass-forming liquids. The phenomenon of dynamic heterogeneity now occupies a central position in the contemporary understanding of the dynamics of glass-forming liquids.[15] In some studies,[23,24] the fragility is claimed to be closely linked to the dynamic heterogeneity. Moreover, a substantial body of research[2,25–35] has been conducted, attempting to establish a general relationship between the structural relaxation and dynamic heterogeneity. Of these studies, one well-documented feature is that there is a nearly universal de-

†Corresponding author. E-mail: pguan@nimte.ac.cn
‡Corresponding author. E-mail: lijin.wang@ahu.edu.cn

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coupling between the time scale, $\tau_{\alpha_{2,\max}}$, corresponding to the maximum non-Gaussian parameter, and the structural relaxation time upon cooling.[25,26,36] Note that the non-Gaussian parameter could be employed to quantify the strength of the dynamic heterogeneity, and hence $\tau_{\alpha_{2,\max}}$ is the characteristic time pertaining to the dynamic heterogeneity. Furthermore, it has been reported that τ consistently exceeds $\tau_{\alpha_{2,\max}}$ [37] in glass-forming liquids, whereas one recent study[25] suggests that $\tau_{\alpha_{2,\max}}$ could also surpass τ under certain circumstances. Here, we contribute to these advancements by demonstrating that more novel insights can be derived from the decoupling relationship between τ and $\tau_{\alpha_{2,\max}}$.

In this study, we delve into the dynamics of different types of model glass-forming liquids across a large range of pressures. We determine a dynamic crossover by probing the decoupling relation between $\tau_{\alpha_{2,\max}}$ and τ. Specifically, $\tau_{\alpha_{2,\max}} = \tau$ at the crossover. Interestingly, we observe that there are several universal dynamic features pertaining to this crossover. First, the crossover divides the behavior of the dependence of $\tau_{\alpha_{2,\max}}$ on τ into two distinctive scenarios. Second, the dynamic heterogeneity quantified by $\alpha_{2,\max}$ at the crossover is nearly constant, leading to scaling collapses both between τ and $\tau_{\alpha_{2,\max}}$ and between $\alpha_{2,\max}$ and τ. Moreover, the crossover could result in a bifurcation in the temperature dependence of the mean squared displacement $\Delta^2(t)$ at $t = \tau$, $\Delta^2(\tau)$. Specifically, above the temperature corresponding to the crossover T_c, $\Delta^2(\tau)$ remains almost constant. In contrast, below T_c, $\Delta^2(\tau)$ exhibits a monotonic increase as the temperature decreases. However, no such behavior could be observed in the analysis of the temperature dependence of the mean squared displacement at $t = \tau_{\alpha_{2,\max}}$, $\Delta^2(\tau_{\alpha_{2,\max}})$.

2. Simulation details

We considered three model systems: two utilizing the soft-core potential $V^{\mathrm{S}}(r_{ij})$ and one employing the hard-core potential $V^{\mathrm{H}}(r_{ij})$. The interaction between particles i and j is either $V^{\mathrm{S}}(r_{ij})$ or $V^{\mathrm{H}}(r_{ij})$ when their separation r_{ij} is less than the cutoff distance r_{ij}^{c}, with no interaction occurring otherwise. The soft-core potential is defined by

$$V^{\mathrm{S}}(r_{ij}) = \frac{\epsilon_{ij}}{\alpha}\left(1 - \frac{r_{ij}}{\sigma_{ij}}\right)^{\alpha}. \tag{1}$$

In $V^{\mathrm{S}}(r_{ij})$, r_{ij}^{c} is set to σ_{ij}; $\alpha = 2$, corresponding to the harmonic potential (hereafter referred to as Harm), whereas $\alpha = 2.5$, the Hertzian potential (Hertz). We implement the hard-core potential through the typical Lennard–Jones potential

$$V^{\mathrm{H}}(r_{ij}) = g(r_{ij}) - g(r_{ij}^{\mathrm{c}}) - (r_{ij} - r_{ij}^{\mathrm{c}})g'(r_{ij}^{\mathrm{c}}), \tag{2}$$

where

$$g(r_{ij}) = 4\epsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]. \tag{3}$$

The r_{ij}^{c} in $V^{\mathrm{H}}(r_{ij})$ is set to $2.5\sigma_{ij}$.

For both soft- and hard-core potentials, our model systems are composed of two species of particles, A and B, with equal mass $M = 1$. The ratio of the number N_{A} of A particles to the number N_{B} of B particles is 4:1. We set $\epsilon_{\mathrm{AB}} = 1.5\epsilon_{\mathrm{AA}}$, $\epsilon_{\mathrm{BB}} = 0.5\epsilon_{\mathrm{AA}}$, $\sigma_{\mathrm{AB}} = 0.8\sigma_{\mathrm{AA}}$, and $\sigma_{\mathrm{BB}} = 0.88\sigma_{\mathrm{AA}}$. Therefore, the hard-core potential in this study corresponds to the well-known Kob–Anderson Lennard–Jones model.[38] The energy, length, and mass are given in units of ϵ_{AA}, σ_{AA}, and M, respectively, and hence the time and temperature are in units of $(M\sigma_{\mathrm{AA}}^2/\epsilon_{\mathrm{AA}})^{1/2}$ and $\epsilon_{\mathrm{AA}}/k_{\mathrm{B}}$, respectively, with k_{B} (set to one) the Boltzmann constant.

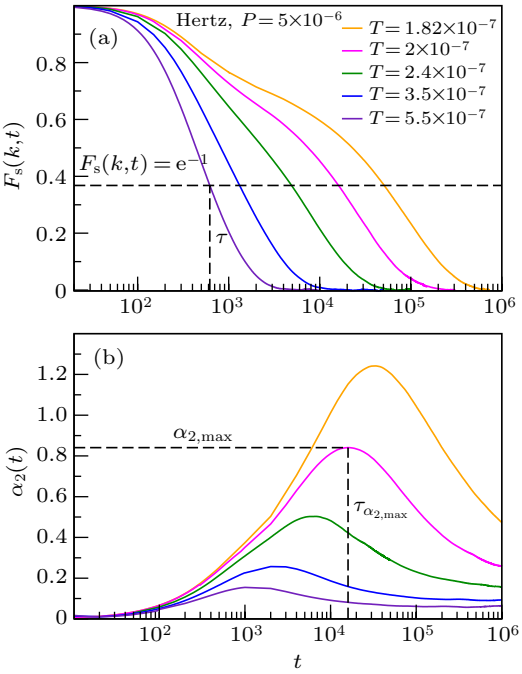


Fig. 1. Illustration of how we determine (a) τ from the self-part of the intermediate scattering function $F_s(k,t)$, as well as (b) $\alpha_{2,\max}$ and $\tau_{\alpha_{2,\max}}$ from the non-Gaussian parameter $\alpha_2(t)$. Lines in (a) and (b) have the same meanings.

We performed extensive molecular dynamics simulations in three-dimensional systems in the ensemble of NPT (particle number N, pressure P, and temperature T). Periodic boundary conditions are employed in all directions during the simulation. In our simulation, we numerically solve the equations of motion of the following form:[39]

$$\frac{\mathrm{d}\boldsymbol{r}_i}{\mathrm{d}t} = \boldsymbol{v}_i + \lambda \boldsymbol{r}_i, \tag{4}$$

$$\frac{\mathrm{d}\boldsymbol{v}_i}{\mathrm{d}t} = \frac{1}{M}\sum_{j\neq i}\boldsymbol{F}_{ij} - (\lambda + \xi)\boldsymbol{v}_i, \tag{5}$$

$$\frac{\mathrm{d}L}{\mathrm{d}t} = L\lambda. \tag{6}$$

Here, $\boldsymbol{r}_i$ and $\boldsymbol{v}_i$ are the position and velocity of particle i, respectively, $\boldsymbol{F}_{ij} = -\nabla U_{ij}$ is the force exerted on particle i by particle j, L is the simulation box length, and λ and ξ are Lagrange multipliers to maintain constant P and T.

To characterize the structural relaxation, we calculate the self-part of the intermediate scattering function,[2] which is defined as $F_s(k,t) = \frac{1}{N}\langle\sum_{j=1}^{N}\exp\{i\boldsymbol{k}\cdot[\boldsymbol{r}_j(t)-\boldsymbol{r}_j(0)]\}\rangle$, where $\boldsymbol{r}_j(t)$ is the location of particle j at time t, $|\boldsymbol{k}|$ takes approximately the value at the first peak of the static structure factor,[39] and $\langle\cdot\rangle$ denotes the time average. The structural relaxation time τ is such that $F_s(k,\tau) = e^{-1}$, as illustrated in Fig. 1(a).

We use the non-Gaussian parameter,[17,26,37,38] which characterizes the degree of the deviation of particle displacements from the Gaussian distribution, to quantify the dynamic heterogeneity. It is defined as $\alpha_2(t) = \frac{3}{5}\langle\Delta r^4\rangle/\langle\Delta r^2\rangle^2 - 1$, where Δr is the displacement of one particle over time t. In glass-forming liquids, $\alpha_2(t)$ typically exhibits a non-monotonic dependence on t at each temperature, as demonstrated in Fig. 1(b). We use the maximum of $\alpha_2(t)$, $\alpha_{2,\max}$, to quantify the strength of dynamic heterogeneity, and the time corresponding to $\alpha_{2,\max}$, $\tau_{\alpha_{2,\max}}$, as a characteristic time scale for the dynamic heterogeneity.

The integration time step in our simulation ranged from 0.001 to 0.01, depending on the temperature. The data were collected after the systems had been equilibrated for at least 10τ (depending on the temperature and pressure). The time for the production run was also at least 10τ (depending on the temperature and pressure). While our following results primarily focus on systems comprising $N = 1000$ particles, we have also conducted partial verification in systems with $N = 10000$ to confirm the absence of finite-size effects that could influence our findings.

3. Results and discussion

Figure 2(a) shows the correlation between τ and $\tau_{\alpha_{2,\max}}$. Although both τ and $\tau_{\alpha_{2,\max}}$ increase upon cooling,[17,26,37] they decouple, i.e., τ is not linearly correlated with $\tau_{\alpha_{2,\max}}$, as demonstrated in Fig. 2(a). In particular, the τ versus $\tau_{\alpha_{2,\max}}$ curves could exhibit significant deviations across different systems, which has garnered some attraction in attempts to elucidate their quantitative correlation. A single power law $\tau \sim \tau_{\alpha_{2,\max}}^{\gamma}$ was proposed originally;[26,36] however, a recent study demonstrated that a combination of two different power laws provides a more reasonable description across the entire range of τ or $\tau_{\alpha_{2,\max}}$.[25] Therefore, it is intriguing to investigate whether there are any distinctive features pertaining to the crossover between the two different power laws. Moreover, although not explicitly stated, it is implicit from several studies that $\tau_{\alpha_{2,\max}} < \tau$.[37] Only recently, it has been reported that it is also possible to observe $\tau_{\alpha_{2,\max}} > \tau$.[25] Therefore, another intriguing aspect to investigate is whether there is anything special with respect to the point where $\tau_{\alpha_{2,\max}} = \tau$. Alternatively, is it possible that the crossover between the two distinct power laws coincides with the point where $\tau_{\alpha_{2,\max}} = \tau$?

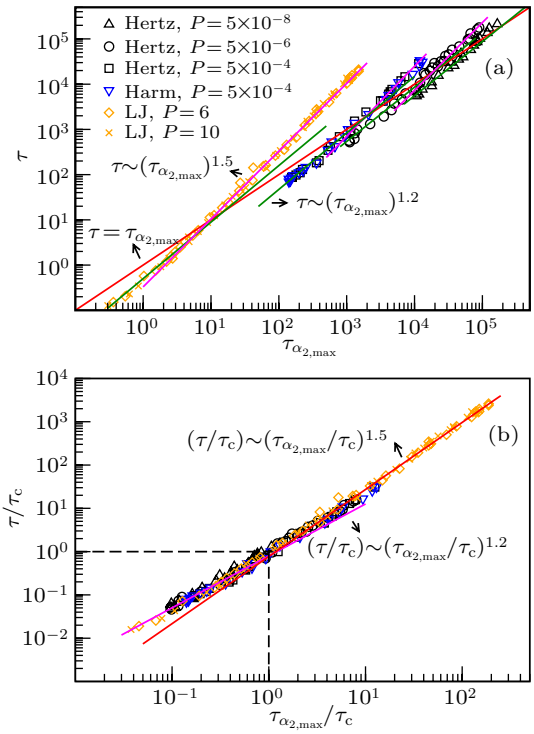


Fig. 2. (a) Correlation between τ and $\tau_{\alpha_{2,\max}}$. The red line indicates $\tau = \tau_{\alpha_{2,\max}}$. Each green line represents $\tau \sim \tau_{\alpha_{2,\max}}^{1.2}$, whereas each pink line corresponds to $\tau \sim \tau_{\alpha_{2,\max}}^{1.5}$. (b) Scaling collapse between τ and $\tau_{\alpha_{2,\max}}$. τ_c is determined as when $\tau = \tau_{\alpha_{2,\max}}$. The solid lines are fits to $\tau/\tau_c \sim (\tau_{\alpha_{2,\max}}/\tau_c)^{\gamma}$, where $\gamma = 1.2$ (pink line) and $\gamma = 1.5$ (red line). Symbols in (a) and (b) represent the same meanings.

Here, we derive an affirmative answer to the aforementioned question by reporting a dynamic crossover with physical significance, corresponding to where $\tau_{\alpha_{2,\max}} = \tau$ in all glass-forming liquids under study in this work. To facilitate the subsequent discussion, we will denote the crossover as either the crossover relaxation time τ_c or the crossover temperature T_c, depending on specific contexts. In the following, we will demonstrate that the dynamic crossover is accompanied by universal features that were not anticipated previously.

First, we observe two different power-law regimes separated by the crossover τ_c in all systems shown in Fig. 2(a). Specifically, $\tau \sim \tau_{\alpha_{2,\max}}^{1.2}$ and $\tau \le \tau_{\alpha_{2,\max}}$ when $\tau \le \tau_c$, whereas $\tau \sim \tau_{\alpha_{2,\max}}^{1.5}$ and $\tau > \tau_{\alpha_{2,\max}}$ when $\tau > \tau_c$. In particular, in our glass-forming liquids at $P = 5\times10^{-8}$ under the Hertzian potential, an obvious regime was observed where $\tau \sim \tau_{\alpha_{2,\max}}^{1.2}$ and $\tau < \tau_{\alpha_{2,\max}}$, whereas in our systems at $P = 6$ or $P = 10$ under the LJ potential, a distinct regime was identified where $\tau \sim \tau_{\alpha_{2,\max}}^{1.5}$ and $\tau > \tau_{\alpha_{2,\max}}$. Therefore, our different glass-forming liquids investigated are complementary in convincingly revealing the existence of two distinct power laws separated by the dynamic crossover that describe reasonably the correlation between τ and $\tau_{\alpha_{2,\max}}$.

The curves of τ versus $\tau_{\alpha_{2,\max}}$ could exhibit either deviation or overlap, as demonstrated in Fig. 2(a) and reported in other studies.[26,36] This raises an important issue: whether there exists a universal description between τ and $\tau_{\alpha_{2,\max}}$.

There have been some efforts attempting to address this issue. Initially, a scaling collapse of τ ($\tau_{\alpha_{2,\max}}$) was achieved by adjusting a system-dependent scaling factor to rescale $\tau_{\alpha_{2,\max}}$,[26] however, the physical significance of the scaling factor remains unclear. Recently, it has been proposed to achieve a universal description between τ and $\tau_{\alpha_{2,\max}}$ by invoking a characteristic time scale τ^*concealed within state points exhibiting identical dynamic heterogeneity in different model glass-forming liquids.[25] Specifically, when plotting τ/τ^* against $\tau_{\alpha_{2,\max}}/\tau^*$, different τ versus $\tau_{\alpha_{2,\max}}$ curves could collapse onto the same master curve.[25] Note that τ^* is system dependent, which is very similar to τ_c reported in this study. This motivates us to check whether τ_c plays the same role as τ^* in achieving a universal description between τ and $\tau_{\alpha_{2,\max}}$. As demonstrated in Fig. 2(b), when both τ and $\tau_{\alpha_{2,\max}}$ are scaled by τ_c, a scaling collapse could be achieved as well. Moreover, it is obvious that the obtained master curve can be adapted well by two power laws with distinct scaling exponents of 1.2 and 1.5, and lines representing these two power laws intersect around the dynamic crossover $\tau = \tau_{\alpha_{2,\max}} = \tau_c$. However, based on a single τ versus $\tau_{\alpha_{2,\max}}$ curve, it's hard to discern so convincingly the two distinct power laws, as demonstrated in Fig. 2(a). It should be noted that we employed power-law relations to fit the data in Fig. 2; however, alternative methods for describing the data cannot be ruled out. What is more intriguing is the hidden physical significance of the possible descriptions, which warrants further theoretical investigation.

Now we have demonstrated that, like τ^*, τ_c could also lead to the scaling collapse of $\tau(\tau_{\alpha_{2,\max}})$. Note that τ^* is defined in a manner that ensures $\alpha_{2,\max}(\tau^*) = C_1$ with C_1 a constant, across different glass-forming systems, and hence different values of C_1 will lead to different τ^*. This motivates us to check how $\alpha_{2,\max}(\tau_c)$ varies. Figure 3(a) shows the correlation between $\alpha_{2,\max}$ and τ for the same systems shown in Fig. 2(a). For each system, $\alpha_{2,\max}$ increases with increasing τ, indicating that dynamic heterogeneity grows as structural relaxation slows down during vitrification.[18,40–42] Interestingly, we could observe that $\alpha_{2,\max}(\tau_c) \approx 0.75$ in all systems displayed in Fig. 3(a), which is another important dynamic feature pertaining to the dynamic crossover. Therefore, a more straightforward method emerges for identifying the dynamic crossover. Specifically, the crossover $\alpha_{2,\max}$, $\alpha_{2,\max,c} \approx 0.75$, regardless of glass-forming liquids and system sizes examined. From the perspective of the constant dynamic heterogeneity,[25] τ_c could be considered a subset of τ^*. Taken together, $\alpha_{2,\max} = f(\tau/\tau^*)$, as demonstrated in Ref. [25], it is expected that $\alpha_{2,\max} = f(\tau/\tau_c)$. This is demonstrated in Fig. 3(b), where the $\alpha_{2,\max}$ versus τ curves for all investigated systems can collapse onto the same master curve when τ is scaled by τ_c.

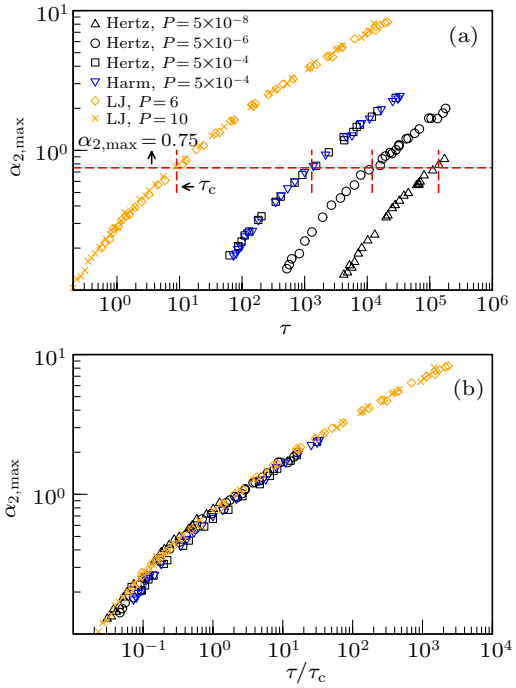


Fig. 3. (a) Correlation between τ and $\alpha_{2,\max}$. Each vertical dashed line marks approximately the crossover τ_c. The horizontal line represents $\alpha_{2,\max} = 0.75$. (b) Scaling collapse between $\alpha_{2,\max}$ and τ. Symbols in (a) and (b) represent the same meanings.

Additionally, there have been several studies[43,44] attempting to achieve a universal description between the structural relaxation and vibrational dynamics in various glass-forming liquids. The vibrational dynamics in these studies is quantified by the well-known mean squared displacement $\Delta^2(t)$ at a very short yet characteristic time. Here, $\Delta^2(t) = \langle \frac{1}{N}\sum_{i=1}^{N} |\boldsymbol{r}_i(t) - \boldsymbol{r}_i(0)|^2 \rangle$. This motivates us to examine the behavior of $\Delta^2(t)$ at $t = \tau$ and $t = \tau_{\alpha_{2,\max}}$. In particular, as T decreases, both τ and $\tau_{\alpha_{2,\max}}$ increase, as demonstrated in Fig. 1. Do $\Delta^2(\tau_{\alpha_{2,\max}})$ and $\Delta^2(\tau)$ exhibit similar behavior?

We show the evolution of $\Delta^2(\tau)$ with $\alpha_{2,\max}$ in Fig. 4(a) and the evolution of $\Delta^2(\tau_{\alpha_{2,\max}})$ in Fig. 4(b) for all systems investigated. Firstly, scaling collapses between $\Delta^2(\tau)$ and $\alpha_{2,\max}$ as well as between $\Delta^2(\tau_{\alpha_{2,\max}})$ and $\alpha_{2,\max}$ could be observed. Interestingly, we observe that the dynamic crossover bifurcates the dependence of $\Delta^2(\tau)$ on $\alpha_{2,\max}$, as demonstrated in Fig. 4(a). Specifically, one could observe an approximate plateau in each $\Delta^2(\tau)$ versus $\alpha_{2,\max}$ plot at low $\alpha_{2,\max}$ up to around $\alpha_{2,\max,c} = 0.75$. This suggests that, despite the difference in relaxation times, particles exhibit nearly identical mean squared displacements when the temperature exceeds the crossover temperature. After the low-$\alpha_{2,\max}$ plateau, $\Delta^2(\tau)$ increases monotonically with $\alpha_{2,\max}$. However, the same scenario does not apply to $\Delta^2(\tau_{\alpha_{2,\max}})$, as demonstrated in Fig. 4(b). In particular, one could observe that $\Delta^2(\tau_{\alpha_{2,\max}})$ decreases with increasing $\alpha_{2,\max}$, which is significantly distinct from the manner in which $\Delta^2(\tau)$ depends on $\alpha_{2,\max}$. It would be interesting to investigate the origin of such a significant disparity in future work.

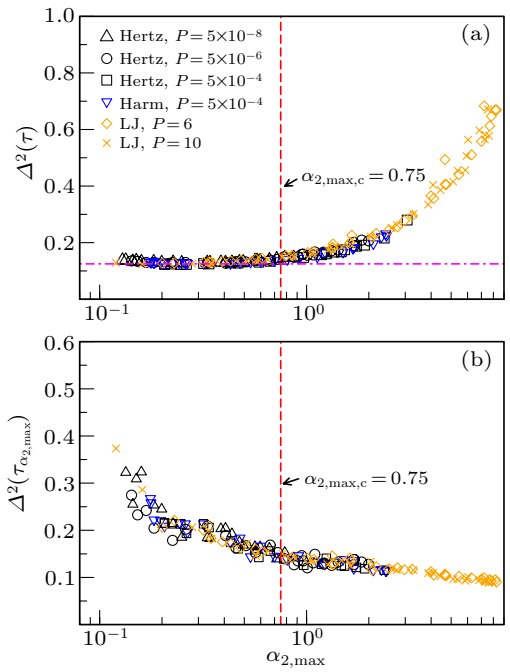


Fig. 4. The $\alpha_{2,\max}$ dependence of $\Delta^2(\tau)$ in (a) and $\Delta^2(\tau_{\alpha_{2,\max}})$ in (b). The vertical dashed lines in (a) and (b) both indicate the crossover $\alpha_{2,\max,c}=0.75$. Symbols in (a) and (b) represent the same meanings.

4. Summary and outlook

In summary, we conduct extensive numerical simulation studies of the relationship between structural relaxation and dynamic heterogeneity in glass-forming liquids under varying pressures and/or interaction potentials. Our results suggest the existence of a novel dynamic crossover where τ equals $\tau_{\alpha_{2,\max}}$. Several features accompany the dynamic crossover. First, the dynamic crossover bifurcates the quantitative relation between τ and $\tau_{\alpha_{2,\max}}$. Specifically, $\tau \sim \tau_{\alpha_{2,\max}}^{1.2}$ and $\tau \leq \tau_{\alpha_{2,\max}}$ when $\tau \leq \tau_c$, whereas $\tau \sim \tau_{\alpha_{2,\max}}^{1.5}$ and $\tau > \tau_{\alpha_{2,\max}}$ when $\tau > \tau_c$. Second, the dynamic heterogeneity quantified by $\alpha_{2,\max}$ at the dynamic crossover is almost the same in all glass-forming liquids investigated. It is this feature that could lead to scaling collapses between τ and $\tau_{\alpha_{2,\max}}$ as well as between $\alpha_{2,\max}$ and τ, in line with the main claims in Ref. [25]. Finally, we observe that the dynamic crossover bifurcates the $\alpha_{2,\max}$ dependence of the mean squared displacement at τ into two distinct regimes. However, such a bifurcation is not observed in the investigation of the $\alpha_{2,\max}$ dependence of the mean squared displacement at $\tau_{\alpha_{2,\max}}$.

In a literal sense, the deviation of the particle displacement distribution from a Gaussian distribution reaches its maximum at the time $\tau_{\alpha_{2,\max}}$. We observe that $\tau_{\alpha_{2,\max}} = \tau$ at the dynamic crossover, which is accompanied by several universal dynamic features. However, the physical mechanism underlying the dynamic crossover remains unclear and warrants further investigation for a more comprehensive understanding.

One interesting question is whether the dynamic crossover, in particular the crossover temperature T_c, coincides with any known characteristic temperatures. Here, we make a preliminary comparison by examining the onset temperature T_{on} associated with slow dynamics, i.e., the temperature where the dynamics begins to deviate from the Arrhenius behavior, as illustrated in Fig. 5(a). Importantly, figure 5(b) indicates that T_c is consistently lower than T_{on} across all systems investigated in this study; correspondingly, the time scale τ_c corresponding to T_c is larger than τ_{on} corresponding to T_{on}, as shown in Fig. 5(c). Additionally, we find that T_c in each system is higher than the corresponding mode-coupling temperature T_{mc},[2] as demonstrated in Fig. 6. Note that a systematic comparison of all known characteristic temperatures in the literature with our reported crossover temperature falls outside the scope of this study. Nevertheless, we leave the comparison and the possible correlation of the crossover temperature with other characteristic temperatures for future investigations.

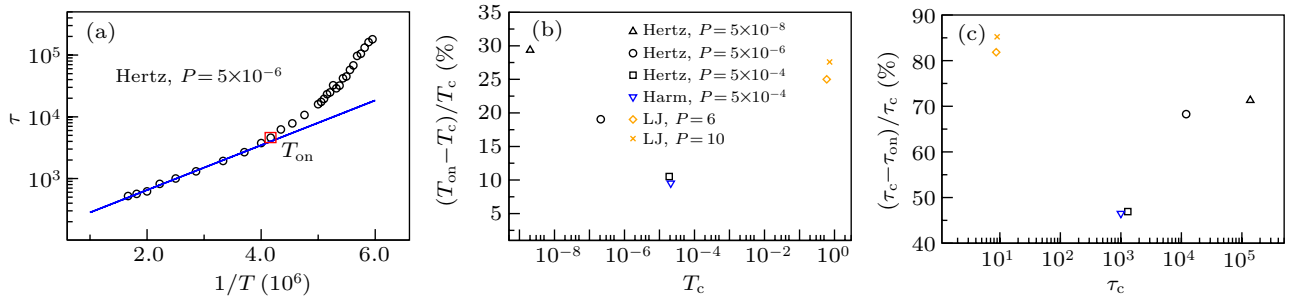


Fig. 5. (a) Illustration of the determination of the onset temperature T_{on}. The red line is a fit to the Arrhenius function $\tau \sim \exp(E_A/T)$ with $E_A = 3.32\times10^{-7}$. (b) The difference between T_{on} and T_c in each system investigated. (c) The difference between τ_{on} (time corresponding to T_{on}) and τ_c. Symbols in (b) and (c) represent the same meanings.

Additionally, the dynamic heterogeneity is more commonly characterized by the four-point dynamic susceptibility χ_4.[2] One natural question is whether such a dynamic crossover reported in this work by studying the non-Gaussian parameter could be obtained as well when comparing τ with the time scale, $\tau_{\chi_{4,\max}}$, corresponding to the maximum χ_4. Actually, this has been addressed implicitly in previous studies.[25,32] Here, we reproduce the relation between τ and $\tau_{\chi_{4,\max}}$ in Fig. 7. It suggests that $\tau \sim \tau_{\chi_{4,\max}}$, and the τ versus $\tau_{\chi_{4,\max}}$ curves could collapse without the need of rescaling τ or $\tau_{\chi_{4,\max}}$, which is consistent with previous studies.[25,32] Therefore, the dynamic crossover is not obvious at all simply from

comparing τ and $\tau_{\chi_{4,\max}}$. This also suggests the distinction between characterizing dynamic heterogeneity through the non-Gaussian parameter and dynamic susceptibility, a topic that warrants further investigation.

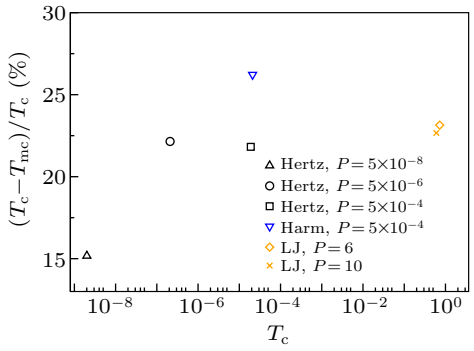


Fig. 6. Comparison between T_{mc} and T_c. T_{mc} denotes the mode-coupling temperature obtained by fitting the relaxation time data to the relation $\tau \sim (T - T_{mc})^{\gamma}$, where γ is a fitting parameter.

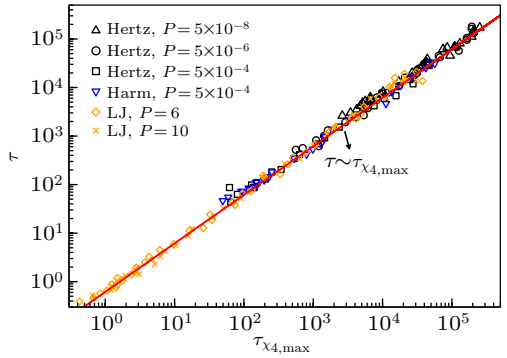


Fig. 7. Correlation between τ and $\tau_{\chi_{4,\max}}$. $\tau_{\chi_{4,\max}}$ is the time scale corresponding to the maximum four-point dynamic susceptibility whose detailed definition could be found in Ref. [25].

Finally, one recent study has reported certain dynamic properties of glass-forming liquids to be a unique function of excess entropy.[12] Therefore, future investigations will aim to determine whether scaling collapses as demonstrated in Figs. 3(b) and 4 exhibit any correlation with excess entropy.

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