Screening and collective effects in randomly pinned fluids: a new theoretical framework

Anh D Phan

Faculty of Materials Science and Engineering, Phenikaa Institute for Advanced Study, Phenikaa University, Hanoi 12116, Vietnam
E-mail: anh.phanduc@phenikaa-uni.edu.vn

Received 9 June 2022, revised 4 August 2022
Accepted for publication 19 August 2022
Published 30 August 2022

Abstract
We propose a theoretical framework for the dynamics of bulk isotropic hard-sphere systems in the presence of randomly pinned particles and apply this theory to supercooled water to validate it. Structural relaxation is mainly governed by local and non-local activated process. As the pinned fraction grows, a local caging constraint becomes stronger and the long range collective aspect of relaxation is screened by immobile obstacles. Different responses of the local and cooperative motions results in subtle predictions for how the alpha relaxation time varies with pinning and density. Our theoretical analysis for the relaxation time of water with pinned molecules quantitatively well describe previous simulations. In addition, the thermal dependence of relaxation for unpinned bulk water is also consistent with prior computational and experimental data.

Keywords: relaxation, glass transition, random pinning, glasses, non-equilibrium physics, collective effects

(Some figures may appear in colour only in the online journal)

1. Introduction
After several decades of intensively attempting to understand fully physical mechanisms underlying glass transition, this phenomenon has still challenged scientists all over the world as an unsolved and fascinating problem [1–3]. When molten materials are cooled down by fast rate to avoid crystallization, the systems fall out of equilibrium. The structural relaxation time of these non-equilibrium substances dramatically increases many orders of magnitudes and exceeds both simulation and experimental time scales. While recent breakthrough developments in glass theory enable to access low temperatures far from the glass transition temperature, \( T_g \), and analyze roles of decisive factors responsible for kinetic slowdown. Understanding how to manipulate the glass transition would open revolution in a wide range of technological applications and fundamental physics.

It is well-known that spatial confinement effects substantially vary glassy dynamics of a system compared to its bulk counterpart [4, 5]. There are many ways to design confinement: pinning molecules and/or particles [6–11], quenching network [12] or using surfaces and interfaces [13–16]. Dynamics of confined systems can be slower [13–16] or faster [17–19] since molecular mobility strongly depends on properties of surfaces, boundaries and finite-size effects. Although much effort has been dedicated to obtaining a universal description for the dynamical property, it is still poorly understood and a debate topic. Among such confinements, influences of random pinning process on the structural relaxation of glass formers have received particular attention due to imitability of quenched disorder porous media and the slowing down dynamics of mobile particles without changing the structure.

Recently, elastically collective nonlinear Langevin equation (ECNLE) theory has described qualitatively and
quantitatively the alpha and beta relaxation, fragility, diffusion constant, and mechanical properties of various single-component materials, composites, and films [19–31]. Basically, the theory views a relaxation event as a combination of two distinctive but strongly related processes: cage-scale hopping and the long range elastically collective rearrangement of surrounding particles. Local cageing constraints play a main role on structural relaxation at high temperatures but the elastic deformation becomes dominant in deeply supercooled regime. The presence of confinement changes correlation between local and collective dynamics. Thus, The relaxation and other related properties are affected. Since the ECNLE theory predicts relaxation time in the range of 0.1 ps to 10^3 s, it can be exploited to explain physical mechanisms underlying measurements in simulations and experiments.

In a prior work [21], we extended the ECNLE theory to qualitatively elucidate trends seen in simulations of slow dynamics in randomly pinned fluids. Immobile particles occupy available space and restrict motion of mobile particles. The presence of pinned particles reduces the mobility of particle cage and greatly enhances local trapping effects. However, pinned particles introduce inhomogeneity of the fluid and are unresponsive to cooperative rearrangement. These features raise an idea of screening or spatial localization of the distortion/displacement field associated with cooperative motions beyond the local cage scale. However, for simplification, we completely ignored the screening effect on the glass transition in [21]. The missing physics leads to a large quantitative deviation between theory and simulations. Thus, with screening effect of elasticity, key intriguing and open questions include: (a) how does the elastic displacement field behave? (b) how does a role of collective motions on the glass transition change with pinning? (c) can new theory provide a better description for simulations and experiments? Answers for these questions are not trivial, and touch on interesting fundamental physics issues.

In this work, we develop a new framework of the ECNLE theory to capture the screening effect caused by immobile components on the structural relaxation of randomly-pinned particle systems. After formulating a mathematical form of the screened displacement field, the longer range collective elastic part of the barrier is calculated. Contribution and competition of the local cage effects and collective rearrangement to the dynamical relaxation become subtle. This behavior totally different from the previous study [21]. To evaluate validity of the new theory, we use this approach to model dynamics of supercooled water and compare results with previous simulation and experimental studies.

### 2. Theoretical background

We consider glassy dynamics of a single-component hard-sphere system but a finite fraction of particles are randomly pinned to provide neutral confinement. This pinning process has no significant effect on the structure. The idealized treatment is consistent with prior simulations [7, 8] which Vogel and his coworkers simulated a system of pinned water molecules and showed that the radial distribution functions, g(r), are insensitive to pinning. The direct correlation function of mobile particles is identical to that of pinned particles c11(r) = c12(r) = c22(r) = c(r) [21], where c(r) is the direct correlation function of the one-component hard sphere fluid, and the subscript 1 and 2 denote mobile and pinned particles, respectively. For a fluid of unpinned bulk hard spheres, the Percus–Yevick integral equation theory can be used to compute structural correlations.

For sufficiently large density, a mobile particle is dynamically arrested within a particle cage formed by its nearest neighbors and a barrier emerges in the dynamic energy profile. The dynamic free energy of a tagged mobile particle in the pinned-mobile sphere systems quantifying its interactions with nearest neighbors is calculated by [21, 32]

\[
\frac{F_{\text{dyn}}(r)}{k_B T} = -\frac{\alpha}{\beta} \frac{\rho c(q)^3}{(1 - \rho(c(q))2 q^2)} - 3 \ln (\frac{L}{d}),
\]

where r is the displacement of the particle, d is the particle diameter, q is the wavevector, k\_B is the Boltzmann constant, T is the temperature, \rho is the density number of a hard-sphere fluid or the number of hard spheres per volume, \alpha is the pinning fraction, c(q) is the Fourier transform of c(r), and the static structure factor S_{12}(q) is given by

\[
S_{12}(q) = \frac{\sqrt{\rho_1 \rho_2 c_{12}(q)}}{[1 - \rho_1 c_{11}(q)][1 - \rho_2 c_{22}(q)] - \rho_1 \rho_2 c_{12}(q)c_{21}(q)},
\]

where \rho_1 = \rho(1 - \alpha) and \rho_2 = \rho(1 + \alpha) are the density number of mobile and pinned particles, respectively. The leading term in equation (1) corresponds to a dynamic mean-field trapping potential favoring localization and the second term favors the fluid state.

Numerical calculations of F_{\text{dyn}}(r) provide information of characteristic length and energy scales of local dynamics as shown in figure 1(a). The local minimum and maximum of F_{\text{dyn}}(r) are the localization length, r_L, and the barrier position, r_B, respectively. The local barrier height is F_B = F_{\text{dyn}}(r_B) - F_{\text{dyn}}(r_L) and the jump distance from the localized position to the barrier position is \Delta r = r_B - r_L.

When \alpha = 0, the dynamic free energy of two-component system reduces to that of one-component system

\[
\frac{F_{\text{dyn}}(r)}{k_B T} = -3 \ln (\frac{r}{d}) - \frac{\rho c(q)^3}{(2\pi)^3} \frac{\rho c(q)^3}{(1 - \rho c(q))(2 - \rho c(q))} \times \exp \left[ -\frac{q^2 r^2}{6} \right],
\]

The pinning process induces slower particle dynamics due to a stronger local caging constraint. Apart from local dynamics, the structural relaxation is also governed by long-range collective motions. Diffusion of a particle from its cage
requires cooperative rearrangement of particles in the first coordination shell to create space for a large amplitude hop and displacement field, \( u(r) \), outside the cage along radial direction. Physically, we expect that pinned particles act as obstacles which can ‘screen’ the displacement field associated with the collective elastic component of the activation barrier. The long range displacements are sufficiently small, so we model the motion around their localized position as harmonic oscillations. The spring constant in ECNLE theory is calculated using \( \kappa \equiv \kappa_0(\alpha) = \frac{\partial^2 E_{\text{kin}}(r)}{\partial r^2} \bigg|_{r=0} \). Pinned particles are made stationary by applying a hypothetical forces to balance the internal elastic force on the particle. The structure of pinned-mobile particle system is identical to that of a one-component system, and the Hookean restoring force acting on a mobile particle \( K_0(\alpha)u(\alpha) \) is the same as the elastic force exerting on the pinned particles. Thus, the external force \( F_{\text{ext}} \) has to be \(-K_0(\alpha)u(\alpha)\). The ‘-’ sign illustrates the opposite direction of \( F_{\text{ext}} \) relative to the elastic force. Additionally, the equilibrium equations and boundary conditions for the continuum elastic treatment of collective barrier remain unchanged.

The continuum elastic equation for the displacement field beyond the cage scale is

\[
\left(K_B + \frac{G}{3}\right) \nabla (\nabla \cdot u) + G \nabla^2 u + \rho \alpha F_{\text{ext}} = 0,
\]

where \( K_B \) and \( G \) are the bulk and dynamic shear modulus, respectively, and \( \rho \alpha F_{\text{ext}} \) is the body force (or force per unit volume). Solving equation (4) with boundary conditions of \( u(r = r_{\text{cage}}) = \Delta u_{\text{eff}} \) and \( u(r \to \infty) = 0 \) gives an analytical result of the displacement field

\[
u(r) = \frac{1 + kr}{1 + kr - r_{\text{cage}}} e^{-\kappa(r-r_{\text{cage}})} \Delta u_{\text{eff}} r_{\text{cage}}^2,
\]

where \( r_{\text{cage}} \) is the cage radius estimated by the first minimum position of \( g(r) \) and an amplitude of the cage expansion, \( \Delta u_{\text{eff}} \), is [21, 33]

\[
\Delta u_{\text{eff}} = \frac{3}{r_{\text{cage}}} \left[ \frac{r_{\text{cage}}^2 \Delta r^2}{32} - \frac{r_{\text{cage}} \Delta r^3}{192} + \frac{\Delta r^4}{3072} \right],
\]

and \( \kappa \) is the inverse screening length of the displacement field given by

\[
\kappa = \sqrt{\frac{60 \Phi}{\pi d^3} \frac{K_0}{K_B + 4G/3}},
\]

where \( \Phi = \rho \pi d^3 / 6 \) is the volume fraction and the bulk modulus of the hard-sphere fluid is [33]

\[
K_B = \frac{6 \Phi k_B T (1 + 2 \Phi)^2}{\pi d^3} (1 - \Phi)^2.
\]

Based on the systematic analysis of the dynamic shear modulus in the ultra-local limit [34], \( G \) can be approximated as

\[
9 \Phi k_B T (1 - \alpha)/5 \pi d r_{\text{cage}}^2. \]

The displacement field is screened in the same manner as the Yukawa-like function.

The elastic barrier is

\[
F_e = 4 \pi \int_{r_{\text{cage}}}^{\infty} dr r^2 \rho (1 - \alpha) g(r) K_0 u^2(r) / 2 = 12 \Phi (1 - \alpha) K_0 \Delta u_{\text{eff}}^2 \left( \frac{r_{\text{cage}}}{d} \right)^3 \left( 1 + \frac{\kappa r_{\text{cage}}}{1 + \kappa r_{\text{cage}}} \right)^2,
\]

where the analytical expression in equation (9) adopts \( g(r) \approx 1 \) (relatively accurate for \( r > r_{\text{cage}} \)), and the factor \((1 - \alpha)\) arises since only mobile particles are responsible for elastic expansion.

Based on Kramer’s theory [20, 33], the structural relaxation time is

\[
\frac{\tau_\alpha}{\tau_s} = 1 + \frac{2 \pi}{\sqrt{\kappa B} \kappa e B} \left( \frac{d}{2 \pi} \right)^3 \frac{\kappa}{k_B T} \exp \left( \frac{F_B + F_e}{k_B T} \right),
\]

where \( B \equiv B_0(\alpha) = \frac{\partial^3 F_{\text{kin}}(r)}{\partial r^3} \bigg|_{r=0} \) is the barrier curvature and \( \tau_s \) is a short relaxation time scale which is [20]

\[
\tau_s = \frac{g(d)}{24 \rho d^2} \left[ \frac{M}{\pi k_B T} \left[ 1 + \frac{1}{36 \Phi} \int_0^{\infty} dq q^2 \frac{S(q) - 1}{S(q) + b(q)} \right] \right],
\]

where \( b^{-1}(q) = 1 - j_0(q) + 2j_2(q) \), \( j_0(x) \) is the spherical Bessel function of order \( n \), \( g(d) \) is a contact number among molecules, \( M \) is the molar mass of particle, and \( S(q) = 1/[1 - \rho c(q)] \). We assume that \( \tau_s \) is unaffected by pinning. For water molecule, \( d = 3 \AA \) and \( M = 3 \times 10^{-20} \) kg.

3. Results and discussions

Figure 1(a) shows a variation of the dynamic free energy with pinning. Although the structure of the pinned hard-sphere fluid remains unchanged with the pinning process, immobile particles strengthens the local caging constraint. Thus, the localization length decreases, and the barrier position and local barrier increase as \( \alpha \) grows. Our results in figure 1(b) suggest that the local barrier raises at least two times when \( \alpha \) changes from 0 to 0.5. It means that pinning particles induces slower cage-scale dynamics compared to the unpinned system.

Equations (9) and (10) indicate how the alpha relaxation depends the elastic barrier. One can expect that the dynamics with and without the screening effect of elasticity deviate with each other by many orders of magnitude. When the screening effect is ignored in the same way as [21], \( \kappa = 0 \) and the analytical form of the elastic barrier becomes

\[
F_e = 12 \Phi (1 - \alpha) K_0 \Delta u_{\text{eff}}^2 \left( \frac{r_{\text{cage}}}{d} \right)^3.
\]

\( K_0(\alpha) \Delta u_{\text{eff}}^2(\alpha) \sim K_0(\alpha) \Delta u^2(\alpha) \) and the jump distance \( \Delta r(\alpha) \) increase with \( \alpha \). Although the factor \((1 - \alpha)\) is reduced as pinning more particles, the unscreened elastic barrier monotonically increases with \( \alpha \) as shown in figure 2(a). This universal variation was already found in [21].
When the screening effect is considered, $\kappa \neq 0$ and the displacement field decays with distance by both exponential function and inverse power laws of distance (as seen in equation (5)). The elasticity or collective effect on glassy dynamics is localized. The localization of elasticity is characterized by an exponential decay length, $\kappa^{-1}/d$. Figure 2(b) shows a weak dependence of the decay length on the volume fraction. Increasing the number of pinned particles leads to greater screening effect of the long range cooperative rearrangement, shortens $\kappa^{-1}$, and significantly decreases $F_e$. At the same pinning fraction, the screened $F_e$ calculated using equation (9) is much smaller than the unscreened counterpart. Interestingly, the screened $F_e$ non-monotonically varies with $\alpha$. After a considerable drop at $\alpha \leq 0.06$, the elastic energy has a small increase but it cannot compare to a rise of the local barrier.

Note that the alpha time calculated using equations (10) and (11) is density-dependent. To validate our screened ECNLE theory, it is necessary to compare the temperature dependence of the predicted alpha time of water with simulation. The comparison requires a thermal mapping to convert from an effective hard-sphere volume fraction into temperature. In prior works [22–27], we formulated a universal correlation between density and temperature for various amorphous materials based on thermal expansion process. The thermal mapping is

$$T = T_{g,\text{bulk}} + \frac{\Phi_g - \Phi}{\beta \Phi_0},$$

(13)

where $T_{g,\text{bulk}}$ and $\Phi_g$ are the bulk glass transition temperature and the glass transition volume fraction, respectively, defined by $\tau_\alpha(\Phi_g) = \tau_\alpha(T_{g,\text{bulk}}) = 100 \text{ s}$, $\beta = 12 \times 10^{-4} \text{ K}^{-1}$.
is an effective thermal expansion coefficient, and \( \Phi_0 = 0.5 \) is a characteristic volume fraction \([22–27]\). Equation \((13)\) is a minimalist conversion and all chemical/conformational complexities of molecules are encoded in \( T_g,_{\text{bulk}} \).

For unpinned bulk water, we find \( T_g = 0.6191 \) and \( T_g,_{\text{bulk}} \approx 136 \) K is the most commonly accepted experimental value \([35–38]\). We now can use the ENCLE theory to calculate \( \tau_\alpha(T) \) for pinned-mobile water and compare with thermal simulations \([7, 8]\) in figure \(3(a)\). The screened ENCLE approach provides a good description with simulation. Particularly, the predicted relaxation time for \( T = 210 \) K perfectly overlaps the simulated relaxation time over a wide range of pinning fractions. However, quantitative agreement between theory and simulation are not obtained at all temperatures. A possible reason is that we assume a universal coupling between the cage-scale hopping and molecular cooperative rearrangements in our calculations, thus relatively simplifying geometrical and chemical effects. This treatment allows us to predict \( \tau_\alpha(T) \) without any adjustable parameter. In \([22, 39]\), one material-specific numerical parameter \( a_\alpha \) is introduced to scale the collective elastic barrier as \( F_e \rightarrow a_\alpha F_e \). The parameter characterizes for a non-universal local-collective correlation in polymers and other amorphous materials. By adjusting \( a_\alpha \), the best fit to experimental \( \tau_\alpha(T) \) and dynamic fragility are achieved. But when experimental/simulation data is not available, the parameter \( a_\alpha \) cannot be determined and analysis is not predictive.

Ignoring the screening effect leads to poor agreement with simulation as shown in figure \(3(b)\) since \( \tau_\alpha \) grows more rapidly with \( \alpha \). Particularly for \( \alpha \geq 0.2 \), a separation between \( \tau_\alpha \) computed using simulation and the ENCLE theory without screening becomes remarkable. For \( \alpha \leq 0.1 \), \( \tau_\alpha \) in figure \(3(a)\) has a slight drop because of a decrease of the elastic barrier but substantially increases when \( \alpha \geq 0.1 \). Since \( \tau_\alpha(T) \) non-monotonically varies with pinning, one can expect the same behavior for the \( T_g \) variation. It is hard to know whether the former trend is correct because the simulations does not study the low \( \alpha \) regime. The reduction may be a true nature of the phenomenon or an artifact of our mean field like screening analysis at low \( \alpha \).

Equations \((10)\) and \((13)\) can be also exploited to determine the temperature dependence of \( \tau_\alpha \) for unpinned bulk water and contrast numerical results with a prior simulation \([40]\) and experimental study \([41]\). In \([41]\), authors measure the self diffusion constant in transiently heated water films as a function of temperature, \( D(T) \). In the framework of ENCLE theory \([25–27]\), the relation between the structural relaxation time and diffusion constant is \( D(T) = \Delta r^2/6\tau_\alpha(T) \). Thus, by approximating \( \Delta r \approx 0.5d = 1.5 \) Å, it is possible for us to deduce \( \tau_\alpha(T) \) from diffusion data. Figure \(4\) shows that our predicted \( \tau_\alpha \) agrees qualitatively well with experimental data for \( T > 252 \) K. This is because at high temperatures, the elastic barrier slightly contributes to the relaxation and the liquid-air interface has a minor effect on the diffusion measurement \([30, 31]\). Thus, the film-averaged diffusion is approximately identical to the bulk value. At low temperatures, interfacial effects on the diffusion and relaxation process become larger \([30, 31]\). The diffusion constant measured in film systems is quantitatively different from the bulk diffusion. While the temperature-dependent diffusion obtained using TIP4P/ICE model \([40]\) nearly overlaps our theoretical curve when reduced by a factor of \( \sim 2 \). Deviation between these curves becomes significant at low temperatures since the ENCLE theory does not capture physics of possible emergence of crystallization and fragile-to-strong transition reported in \([38, 42]\).

The thermal dependence of \( \tau_\alpha \) is also analyzed using the dynamic fragility \( m = (\partial \log \tau_\alpha/\partial (T_g/T))_{T=T_g} \). From ENCLE data in figure \(4\), we find that the theoretical fragility is about 46, which is much higher than the experimental analog \( (m \sim 18) \) \([41]\). This difference can be considered as a consequence of the super-Arrhenius dynamics. Both the ENCLE theory and Dyre’s phenomenological elastic shoving model \([43, 44]\) indicate that the strong temperature dependence of the elastic collective dynamics with cooling is solely responsible for the super-Arrhenius behavior. However, in \([42]\), Shi et al proposed a two-state theory to describe the water dynamics.

![Figure 3.](image-url)
Two states (so-called fast and slow water state) co-exist in different structures and obey Arrhenius law. The view contradicts with our initial assumption that only one structure exists at a given temperature/density. This may be another missing physics for the ECNLLE theory to predict the fragile-to-strong crossover besides nonuniversal coupling of cage scale hopping and collective elastic distortion.

4. Conclusions

We have formulated a new theoretical approach based on the ECNLLE theory to understand physical mechanisms underlying slow dynamics in randomly pinned particles. The random pinning constructs neutral confinement which has no influence on pair structure but the dynamics is dramatically changed. Our theoretical results show that pinning particles induces slower cage-scale dynamics via strengthening a local caging constraint on mobile particles. An expansion of the first coordination shell is required to create space for a large amplitude local structural relaxation and generate a long range displacement field outside the cage via elastically cooperative motions of particles. Since the pinned particles are not allowed to move, the displacement field nucleated from a cage surface cannot propagate through them. Thus, we propose a new screening type of effect for the elastic displacement field proposed in this work is relatively sensible. The approach could reveal a true nature of slow dynamics in randomly pinned particle fluids.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

This research was funded by the Vietnam National Foundation for Science and Technology Development (NAFOSTED) under Grant Number 103.01-2019.318.

ORCID ID

Anh D Phan  https://orcid.org/0000-0002-8667-1299

References

[1] Debenedetti P G and Stillinger F H 2001 Nature 410 259–67
[2] Ngai K 2007 J. Non-Cryst. Solids 353 709–18
[3] Berthier L and Biroli G 2011 Rev. Mod. Phys. 83 587
[4] Kremer F 2014 Dynamics in Geometrical Confinement (Berlin: Springer)
[5] Schweizer K S and Simmons D S 2019 J. Chem. Phys. 151 240901
[6] Gokhale S, Hima Nagamanasa K, Ganapathy R and Sood A K 2014 Nat. Commun. 5 4685
[7] Klameth F and Vogel M 2015 J. Phys. Chem. Lett. 6 4385
[8] Klameth F, Henritzi P and Vogel M 2014 J. Chem. Phys. 140 144501
[9] Kob W and Berthier L 2013 Phys. Rev. Lett. 110 245702
[10] Scheidler P, Kob W and Binder K 2004 J. Phys. Chem. B 108 6673–86
[11] Kob W and Coslovich D 2014 Phys. Rev. E 90 052305
[12] Jadrich R B and Schweizer K S 2014 Phys. Rev. Lett. 113 208302
[13] Sarangapani P S, Schofield A B and Zhu Y 2011 Phys. Rev. E 83 030502
[14] Fakhraai Z and Forrest J A 2005 Phys. Rev. Lett. 95 025701
[15] Glor E C, Composto R J and Fakhraai Z 2015 Macromolecules 48 6682
[16] Lang R J, Merling W L and Simmons D S 2014 ACS Macro Lett. 3 758
[17] Paeng K and Ediger M D 2011 Macromolecules 44 7034
[18] Paeng K, Richert R and Ediger M D 2012 Soft Matter 8 819–26
[19] Mirigian S and Schweizer K S 2014 J. Chem. Phys. 141 161103
[20] Mirigian S and Schweizer K S 2014 J. Chem. Phys. 140 194506
[21] Phan A D and Schweizer K S 2018 J. Chem. Phys. 148 054502
[22] Phan A D, Wakabayashi K, Paluch M and Lam V D 2019 RSC Adv. 9 40214–21
[23] Phan A D, Zacccone A, Lam V D and Wakabayashi K 2021 Phys. Rev. Lett. 126 025502
[24] Phan A D, KnapiK-Kowalczyk J, Paluch M, Hoang T X and Wakabayashi K 2019 Mol. Pharm. 16 2992–8
[25] Phan A D, Koperwas K, Paluch M and Wakabayashi K 2020 Phys. Chem. Chem. Phys. 22 24365
[26] Ngan N K, Phan A D and Zaccone A 2021 Phys. Status Solidi 15 2100235

[27] Phan A D, Ngan N K, Nga D T, Le N B and Ha C V 2022 Phys. Status Solidi 16 2100496

[28] Zhang R and Schweizer K S 2018 J. Phys. Chem. B 122 3465–79

[29] Phan A D and Schweizer K S 2019 Macromolecules 52 5192–206

[30] Phan A D and Schweizer K S 2018 Macromolecules 51 6063–75

[31] Ghanekarade A, Phan A D, Schweizer K S and Simmons D S 2021 Proc. Natl Acad. Sci. 118 e2104398118

[32] Viehman D C and Schweizer K S 2008 J. Phys. Chem. 128 084509

[33] Mirigian S and Schweizer K S 2014 J. Chem. Phys. 140 194507

[34] Schweizer K S and Yatsenko G 2007 J. Chem. Phys. 127 164505

[35] Bertolini D, Cassettari M and Salvetti G 1982 J. Chem. Phys. 76 3285–90

[36] Kringle L, Thornley W A, Kaya B D and Kimmel G A 2021 Proc. Natl Acad. Sci. 118 e2022884118

[37] Amann-Winkel K, Gainaru C, Handle P H, Seidl M, Nelson H, Bohmer R and Loerting T 2013 Proc. Natl Acad. Sci. 110 17720–5

[38] Swenson J 2018 Phys. Chem. Chem. Phys. 20 30095–103

[39] Xie S-J and Schweizer K S 2016 Macromolecules 49 9655–64

[40] Espinosa J R, Navarro C, Sanz E, Valeriani C and Vega C 2016 J. Chem. Phys. 145 211922

[41] Xu Y, Petrik N G, Smith R S, Kay B D and Kimmel G A 2016 Proc. Natl Acad. Sci. 113 14921–5

[42] Shi R, Russo J and Tanaka H 2018 Proc. Natl Acad. Sci. 115 9444–9

[43] Dyre J C, Christensen T and Olsen N B 2006 J. Non-Cryst. Solids 352 4635–42

[44] Dyre J C 2006 Rev. Mod. Phys. 78 953