Quantum mode-coupling theory for binary mixtures

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We extend the quantum mode-coupling theory of neat liquids to the case of binary mixtures, in order to study supercooled liquids where quantum fluctuations may compete with thermal fluctuations. We apply the theory to a generic model of a binary mixture of Lennard-Jones particles. Our treatment may be used to study quantum aging and exotic glass melting scenarios in structural supercooled quantum liquids. © 2005 American Institute of Physics. [DOI: 10.1063/1.1832593]

I. INTRODUCTION

When a liquid is rapidly cooled below its melting temperature, the system enters a supercooled state, where the relaxation is characterized by very slow dynamics. For classical liquids, the description of supercooled and glassy states remain a challenge. Even the numerical simulation of deeply supercooled liquids is not feasible due to the dramatic slowing of dynamical fluctuations near the glass transition. The crossover temperature for slow relaxation for most glass forming materials, such as polymer glasses, spin glasses, and structural glasses, occurs at a temperature where quantum fluctuations are negligible. Several exceptions, where quantum fluctuations compete with thermal fluctuations, have been studied, including the so called Coulomb glasses and quantum spin glasses. However, a similar slowing that may occur in a structural quantum glass is virtually unexplored.

State of the art quantum simulation methods, such as semiclassical, path-integral centroid, and analytic continuation methods, are still limited to short time dynamics, and thus are not applicable to study the slowing down of the supercooled quantum state. In principle, the quantum mode-coupling theory (QMCT) developed by Reichman and Rabani for neat (single component) quantum liquids can be used to study a range of problems related to the dynamics of supercooled quantum systems. But, in practice, single-component liquids do not show glassy behavior because nucleation occurs rapidly, and the system crystallizes even on the time scales of computer simulations.

Unlike single-component liquids, binary mixtures provide a simple route to study glassy phenomena, since particle dispersion prevents crystallization. Thus, classical binary mixtures have been used to explore and understand the dynamics near the glass transition. In this paper we adopt a similar approach to study quantum glassy systems, and take a first step towards the development of a quantum mode-coupling theory suitable for supercooled binary mixtures. In Sec. II we formulate a QMCT for binary mixtures starting from a quantum generalize Langevin equation (QGLE). Applications of the QMCT for binary mixtures is provided in Sec. III for supercooled liquids somewhat above the onset temperature of slow dynamics. We focus on a generic model of Lennard-Jones binary mixtures, sometimes referred to as the Kob–Andersen model, and discuss the dynamics in the normal and slightly supercooled regimes. Finally, in Sec. IV we conclude.

II. QUANTUM MODE-COUPLING THEORY FOR BINARY MIXTURES

We consider a two-component system of quantum particles with concentrations \( x_j = N_j/N \), where \( N_j \) is the number of particles for species \( j = 1, 2 \). We focus on density fluctuations described by the partial intermediate scattering function

\[
F_{ij}^{\alpha}(q,t) = \frac{1}{N} \langle \hat{\rho}_i^\alpha(q) \hat{\rho}_j(q,t) \rangle.
\]

where \( \hat{\rho}_j(q) = \sum_{\alpha=1}^{N_j} e^{i q \cdot r_\alpha} \) is the density operator at wave vector \( q \) for species \( j \). In the above equation, the notation \( \kappa \) implies that the quantity under consideration involves the Kubo transform given by

\[
\hat{\rho}_j(q) = \frac{1}{\beta \hbar} \int_0^{\beta \hbar} d\lambda e^{-\lambda \hat{H}} \hat{\rho}_j(q) e^{\lambda \hat{H}}.
\]

where \( \hat{H} \) is the Hamiltonian of the system, \( \beta = 1/(k_B T) \) is the inverse temperature, and \( \langle \cdot \cdot \cdot \rangle \) denotes a quantum mechanical ensemble average.

To obtain the corresponding real-time partial intermediate scattering function (non-Kubo), the standard relation in frequency space must be applied

\[
S_{ij}(q,\omega) = \frac{\beta \hbar \omega}{2} \left[ \coth \left( \frac{\beta \hbar \omega}{2} \right) + 1 \right] S_{ij}^{\alpha}(q,\omega),
\]

where \( S_{ij}^{\alpha}(q,\omega) \) is the Fourier transform of the Kubo transform of the partial intermediate scattering function \( F_{ij}^{\alpha}(q,t) \) given by

\[
S_{ij}^{\alpha}(q,\omega) = \frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} dt e^{i \omega t} F_{ij}^{\alpha}(q,t).
\]
and \( S_{ij}(q,\omega) \) is the partial dynamic structure factor (also the corresponding Fourier transform of the non-Kubo partial intermediate scattering function).

Following the standard projection operator procedure, the time evolution of the Kubo transform of the partial intermediate scattering function is given by the exact QGLE in the matrix form

\[
\frac{d^2 F^\alpha(q,t)}{dt^2} + \Omega_{\alpha}^2(q) F^\alpha(q,t) + \int_0^t dt' K^\alpha(q,t-t') \frac{d F^\alpha(q,t')}{dt'} = 0,
\]

(5)

where \( F^\alpha(q,t) \) is a 2x2 matrix with components \( F_{ij}^\alpha(q,t) \). In the above equation the frequency matrix \( \Omega_{\alpha}^2(q) \) (for \( n = 1 \)) is also a 2x2 matrix given by

\[
\Omega_{\alpha}^2(q) = \Omega_{\alpha}^2(q)[S^\alpha(q)]^{-1},
\]

(6)

where \( [S^\alpha(q)]^{-1} \) is the inverse of the matrix \( S^\alpha(q) \), whose elements are given by the Kubo transform of the partial static structure factor \( S_{ij}^\alpha = F_{ij}^\alpha(q,0) \), and \( \Omega_{\alpha}^2 \),

\[
\Omega_{\alpha}^2(q) = \left\langle \frac{d^2 \tilde{p}_i^\alpha(q)}{dt^2} \right\rangle = \frac{1}{2} \left\langle \frac{d^2 \tilde{p}_i^\alpha(q)}{dt^2} \right\rangle.
\]

Following the derivation for a quantum mode-coupling theory of neat fluids, the 2x2 memory kernel matrix \( K^\alpha(q,t) = K_{ij}^\alpha(q,t) + K_{mc}^\alpha(q,t) \) is approximated by a sum of two matrices, a fast decaying binary portion \( K_{ij}^\alpha(q,t) \) and a slower decaying mode-coupling portion \( K_{mc}^\alpha(q,t) \). The former is obtained from a short time expansion of the exact memory kernel, and is given by

\[
K_{ij}^\alpha(q,t) = K_{ij}^\alpha(q,0) \exp \left[ -\Lambda^\alpha(q) t^2 \right],
\]

(7)

where the 2x2 matrix \( \Lambda^\alpha(q) \) is given by

\[
\Lambda^\alpha(q) = -[\Omega_{\alpha}^2(q,0)]^{-1} \Omega_{\alpha}^2(q,0).
\]

(8)

In the above equations, \( K_{ij}^\alpha(q,0) \) and \( K_{mc}^\alpha(q,0) \) are given by

\[
K_{ij}^\alpha(q,0) = -\Omega_{\alpha}^2(q)[\Omega_{\alpha}^2(q)]^{-1} + \Omega_{\alpha}^2(q)[\Omega_{\alpha}^2(q)]^{-1} = 0,
\]

(9)

and

\[
K_{mc}^\alpha(q,0) = \Omega_{\alpha}^2(q)[\Omega_{\alpha}^2(q)]^{-1} - \Omega_{\alpha}^2(q),
\]

(10)

respectively, and \( \Omega_{\alpha}^2(q) \) is given by Eq. (6). In the applications reported below we approximate the matrix \( \Lambda^\alpha(q) \) by the empirical expression

\[
\Lambda^\alpha(q) = \frac{1}{4} \Omega_{\alpha}^2(q,0).
\]

(11)

This expression does not involve higher order terms such as \( \Omega_{\alpha}^2(q) \). Although the calculation of \( \Omega_{\alpha}^2(q) \) is possible, it involves higher order derivative of the interaction potential and thus becomes a tedious task for the path integral Monte Carlo technique. Interestingly, for neat fluids, we find that the approximation to the lifetime given by Eq. (11) is accurate to within 5% for classical fluids. It also provides quantitative results for classical binary mixtures.

Turning to discuss the slow portion of the memory kernel, the partial mode-coupling matrix elements are given by

\[
[K_{mc}^\alpha(q,t)]_{ij} \approx \frac{k_B T}{2m_j \pi (2\pi)^n} \sum_{\alpha',\beta} \int dk \times V_{ia\beta}^\alpha(q,k) V_{ja'}^\alpha(q,|q-k|) \\
\times [F_{k\alpha}^\beta(k,t) F_{b\beta}^\alpha(|q-k|)]_{ib} \\
- F_{b\alpha}^\beta(k,t) F_{k\beta}^\alpha(|q-k|),
\]

(12)

where \( n \) is the liquid number density and \( m_i \) is the mass of species \( i \). The exact definition of the vertex \( V_{ia\beta}^\alpha(q,k) \) appearing in the Eq. (12) is described elsewhere (for monatomic case), and its generalization for mixtures is straightforward. In the applications reported below we adopt an approximation to the vertex similar to the one used for neat fluids. Namely, for the three-point density terms we use the (Kubo) convolution approximation and for the two-point density-current terms we have used the fact that the matrix \( J_{ij}^\alpha \) can be approximated by a diagonal form with elements given by \( [J_{ij}^\alpha]_{ij} = (k_B T/m_i) \delta_{ij} \) (\( \delta_{ij} \) is Kronecker’s delta function). This approximation for the two point density-current terms is accurate within an error that is less than 1% for the relevant \( q \) values for the binary mixture studied in this work. Based on these approximations, the matrix elements of the vertex are given by

\[
V_{ia\beta}^\alpha(q,k) \approx \frac{q \cdot k}{q} [S_{\alpha}^{-1}(k)]_{i\alpha} \delta_{\beta \beta} \\
+ \frac{(q-k) \cdot q}{q} [S_{\alpha}^{-1}(q)]_{ij} \delta_{\beta \alpha} \\
- \frac{q}{x_j} [S(k)S_{\alpha}^{-1}(k)]_{ij} \delta_{\alpha \beta}. \]

(13)

In the above equation, \( [S_{\alpha}^{-1}(q)]_{ij} \) is the \( ij \) element of the inverse matrix of \( S_{\alpha}(q) \) and as before \( \delta_{ij} \) is Kronecker’s delta function.

The subtraction of the product of terms in Eq. (12) involving the matrix elements of \( F_{ij}^\alpha(q,t) \) is done to prevent overcounting the total memory kernel at short times, namely, to ensure that the even time moments of the total memory kernel are exact to forth order in time. The binary term of the Kubo transform of the partial intermediate scattering function appearing in Eq. (12) is obtained from a short time expansion of \( F^\alpha(q,t) \), and is given by

\[
F_{ij}^\alpha(q,t) = S_{ij}^\alpha(q) \exp \left[ -\frac{1}{4} \Omega_{\alpha}^2(q,t^2) \right].
\]

(14)

The symbol “*” in the above equation designates the transpose of the matrix \( \Omega_{\alpha}^2(q) \).

The three-dimensional integral in Eq. (12) can be reduced to a two-dimensional integral by making use of the isotropy of the system. The result is given by
easily obtained. is a diagonal matrix so that the function 

\[ M \] 

where 

\[ V_{ij} \] 

have diagonalized these matrices with the appropriate unitary 

transformation, and evaluated the function of these matrices 

\[ A \] 

as follows:

\[ A \text{transformation, and evaluated the function of these matrices as follows:} \]

\[ f(M) = \mathcal{U} [f(\mathcal{U}^{-1} M \mathcal{U})] \mathcal{U}^{-1} , \] 

where \( M \) is a matrix, \( f \) is an analytic function, and \( \mathcal{U}^{-1} M \mathcal{U} \) is a diagonal matrix so that the function \( f \) of the latter can be easily obtained.

\[ \left[ K_{\alpha \beta}^\epsilon (q,t) \right]_{ij} \]

\[ \approx \frac{k_B T}{32 \pi m \xi} \sum_{\alpha, \beta} \sum_{i,j} \times \int_0^\infty dk \int_{|q-k|}^{|q+k|} dk' \left[ V_{i \alpha \beta}(q,k,k') V_{j \alpha' \beta'}(q,k',k) \right] 

\times \left[ F_{\alpha \alpha}(k,t) F_{\beta \beta}(k',t) - F_{\alpha \beta}(k,t) F_{\beta \alpha}(k',t) \right], \]

(15)

where

\[ V_{i \alpha \beta}(q,k,k') \]

\[ \approx (q^2 + k^2 - k'^2) \delta_{ij} \left( \frac{Q_{ja}(k)}{x_j} - [S^{-1}(k)]_{ja} \right) \]

\[ + (q^2 + k'^2 - k^2) \delta_{ij} \left( \frac{Q_{j\alpha}(k')}{x_j} - [S^{-1}(k')]_{ja} \right) \]

(16)

and

\[ Q_{\alpha \beta}(k) = [(S(k) S^{-1}(k))]_{\alpha \beta} \delta_{\alpha \beta} . \]

(17)

To perform the calculation of the exponential function of the matrices \( A^\epsilon(q) \) and \( \Omega^\epsilon(q) \) [cf. Eqs. (7) and (14)], we have diagonalized these matrices with the appropriate unitary transformation, and evaluated the function of these matrices as follows:

\[ f(M) = \mathcal{U} [f(\mathcal{U}^{-1} M \mathcal{U})] \mathcal{U}^{-1} , \]

(18)

\[ \text{III. RESULTS FOR QUANTUM KOB–ANDERSEN MODEL} \]

We focus on a 80:20 mixture \((x_1=0.8 \text{ and } x_2=0.2)\) of Lennard-Jones particles with a mass equal to that of a hydrogen molecule \((m_1=m_2=m_{12})\). The pair interaction potential is given by \( V_{ij} = 4 \epsilon \sigma_j / \left( \sigma_j / r \right)^{12} - \left( \sigma_j / r \right)^6 \), \( i,j=1,2 \). The parameters for the interactions between the \( i = 1 \) particles were chosen to mimic that of the Silvera-Goldman \( \text{para-hydrogen pair potential; } \epsilon_{11} = 30 \text{ K and } \sigma_{11} = 3 \text{ Å} \). The remaining interactions were identical to those used by Kob and Andersen for classical systems.\(^{54,55}\) For the \( i = 2 \) particles we took \( \epsilon_{22} = 0.5 \epsilon_{11} \) and \( \sigma_{22} = 0.88 \sigma_{11} \), and the cross terms were \( \epsilon_{12} = 1.5 \epsilon_{11} \) and \( \sigma_{12} = 0.8 \sigma_{11} \).

The results reported below were obtained for liquid number densities that are somewhat above the triple-point density of liquid \text{para-hydrogen} \((n = 0.8 \sigma_1^3 \text{ and } 0.9 \sigma_1^3)\), and for a range of temperatures above and slightly below the melting temperature of the mixture \((0.3 \leq k_B T / \epsilon_{11} \leq 1)\). These densities were chosen to ensure that the onset temperature of slow relaxation is low enough to overlap with the onset of quantum fluctuations.

To obtain the static quantities required for the solution of the QGLE for the binary mixtures, we have performed path integral Monte Carlo (PIMC) simulations using the NVT ensemble with \( N = 256 \) particles. In the path integral simulation the imaginary-time interval was discretized into \( P \) Trotter slices of size \( \epsilon = \beta / P \) with \( P = 100 \) (this value was sufficient to converge the results at the lowest temperature studied). The staging algorithm\(^ {70}\) combined with centroid moves was employed for the Monte Carlo moves. The number of beads that were staged was 50 above \( k_B T / \epsilon_{11} = 0.25 \) and 25 below this temperature. The PIMC simulations were started from the face-centered cubic (fcc) lattice configuration at the highest
temperature studied ($k_B T / \epsilon_{11} = 1.0$). The number of Monte Carlo passes at each temperature was sufficient to equilibrate the system at that given temperature. The final configuration at each temperature was used as the initial configuration for the next temperature.

In Figs. 1 and 2 we show the partial static structure factor $S_{ij}^\epsilon(q)$, the partial frequency factor $[ \Omega^\epsilon_2(q)]_{ij}$, and the zero time partial memory kernel $[ K^\epsilon(q,0)]_{ij}$ for the two reduced densities studied. We focus on the $q$ component, since it is related to the 12 term:

$$S_{ij}^\epsilon(q) = S_{ij}^\epsilon(q) = (m_1 / x_1) [ \Omega^\epsilon_n ]_{ij}, \quad (m_2 / x_2) [ \Omega^\epsilon ]_{ij}, \quad (m_3 / x_3) [ K^\epsilon(q,0)]_{ij}.$$  

Several interesting features, some common to classical binary mixtures, were observed for the static input. In the limit $q \rightarrow 0$, the value of $S_{ij}^\epsilon(q)$ is much lower than that of $S_{22}^\epsilon(q)$, reflecting the lower compressibility of larger particles. Furthermore, for $q \leq q_{11}^{\max}$, $[ q_{ij}^{\max} \leq q_{ij} ]$ is the value of $q$ where $S_{ij}^\epsilon(q)$ reaches its first maximum. We observed two different length scales for the 22 component, while only one is observed for the 11 component. This is a result of the stronger cross-particle interactions ($\epsilon_{12} > \epsilon_{11} > \epsilon_{22}$) and the lower molar fraction of $i=2$ particles, which results in an additional, slightly larger, length scale for component 22. Unlike the classical case, the Kubo transform of the partial structure factor does not reach the plateau value of 1 at high values of $q$, however, the positions of its peaks are identical to those of the non-Kubo partial structure factor.

We find that the temperature dependence of $S_{ij}^\epsilon(q)$ is very smooth. As the temperature decreases (or density increases), the height of the first peak increases, similar to the classical case. At higher values of $q > q_{11}^{\max}$ the Kubo transformed partial static structure factor decreases with decreasing temperature, unlike the classical case. This is a result of the increase of quantum fluctuations relative to thermal fluctuations, giving rise to lower values of the imaginary-time density-density correlations entering the Kubo transformation [cf. Eq. (2)].

In Figs. 1 and 2 we also plot the Kubo transform of the partial frequency factor $[ \Omega^\epsilon_2(q)]_{ij}$, which has a shape that is very similar to that of a classical liquid mixture. The weak, nearly linear, temperature dependence of $[ \Omega^\epsilon_2(q)]_{ij}$ is consistent with the result that $J^\epsilon_2(q)$ can be approximated by a diagonal matrix with elements equal to $[ \Omega^\epsilon_2(q)]_{ij}$. Several interesting features, some common to classical binary mixtures, were observed for the static input. In the limit $q \rightarrow 0$, the value of $S_{ij}^\epsilon(q)$ is much lower than that of $S_{22}^\epsilon(q)$, reflecting the lower compressibility of larger particles. Furthermore, for $q \leq q_{11}^{\max}$, $[ q_{ij}^{\max} \leq q_{ij} ]$ is the value of $q$ where $S_{ij}^\epsilon(q)$ reaches its first maximum. We observed two different length scales for the 22 component, while only one is observed for the 11 component. This is a result of the stronger cross-particle interactions ($\epsilon_{12} > \epsilon_{11} > \epsilon_{22}$) and the lower molar fraction of $i=2$ particles, which results in an additional, slightly larger, length scale for component 22. Unlike the classical case, the Kubo transform of the partial structure factor does not reach the plateau value of 1 at high values of $q$, however, the positions of its peaks are identical to those of the non-Kubo partial structure factor.

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$q = 0$. However, since momentum is not conserved for individual components in a binary mixture, the memory kernel can take a finite value at $q = 0$.

The results for the Kubo transform of the partial intermediate scattering function at the two densities studied are shown in Figs. 3 and 4. The results for each component are plotted at $q = q_{ij}^{\text{max}}$ and at $q = q_{ij}^{\text{max}}/2$ for that component. Time is measured in units of $\sqrt{m\sigma_{11}^2/48\epsilon_{11}}$.

The short time decay of the partial intermediate scattering function, which is weakly temperature dependent for both densities, can be approximated by a quadratic decay given by Eq. (14) for both wave vectors shown. This short time decay is dominated by ballistic motion. At intermediate times, the dynamic response of the binary liquid depends on the value of $q$. At low values of $q$ the Kubo transform of the intermediate scattering function develops a shoulder for the 11 com-
ponent at both densities. The shoulder signifies the existence of collective coherent excitations and becomes more pronounced at lower temperatures or at higher densities. As $q$ approaches the value of $q_{ij}^{\text{max}}$, the shoulder disappears. At long times, the decay of the partial intermediate scattering function is characterized by a single relaxation time that can be fit to a simple exponential form for all values of $q < q_{ij}^{\text{max}}$. As the temperature decreases or the density increases the relaxation becomes slower.

At the lowest temperature studied for $n = 0.8/s^{1.3}$ the behavior at intermediate times for $q = q_{ij}^{\text{max}}$ is slightly different compared to the higher temperature case. The intermediate and long time decay show hints for a two step relaxation. The two step relaxation is typical of classical supercooled binary liquids. The standard interpretation of this for classical systems is that the intermediate time relaxation is associated with motion of particles inside the cage (known as the $\beta$ relaxation), while the longer time decay is associated with hopping of particles outside their cage (known as the $\alpha$ relaxation). Our results suggest that it is possible that supercooled quantum liquids will show a similar relaxation behavior.

In order to provide a more conclusive picture of slowing down in supercooled quantum liquids we must solve the quantum mode-coupling equations at lower temperatures. However, several approximations built into the treatment of the quantum mode-coupling theory presented here limit the application to a relatively high temperature supercooled state. The major approximation involves the simplified version of the quantum vertex given by Eq. (14), which is based on the (Kubo) convolution theory, and leads to unphysical divergence at high values of $q$. It is sufficient for the description of density fluctuations in normal quantum liquids where a cutoff in $q$ provides quantitative results. For the deep supercooled state, couplings to higher $q$ values are necessary to properly describe the slowing down of dynamical susceptibilities. However, at these higher $q$ values the divergent behavior of the simplified vertex steps in, and (at this point) prohibits an accurate calculation of the dynamic response of the liquid. The exact temperature at which our approach fails is hard to predict a priori. However, we find that a numerical solution of the QGLEs becomes unstable at low temperatures for any reasonable choice of the cutoff value in $q$.

The Kubo transformed partial memory kernel at the same value of $q$ used for the partial intermediate scattering function and at the same thermodynamic state is plotted in Figs. 5 and 6. The decay of the memory kernel is characterized by two relaxation regimes. At short times the quadratic decay observed is dominated by the binary portion. This decay is nearly temperature independent. At long times the decay is dominated by the mode-coupling portion of the memory kernel. Unlike the binary portion, the decay of the mode-coupling portion is temperature dependent. As the temperature is lowered the long time decay of the memory kernel becomes slower. The fact that only the mode-coupling portion of the memory kernel is temperature sensitive is a result of the self-consistent nature of the mode-coupling equation. These give rise to the nonergodic behavior expected at very lower temperatures.

IV. CONCLUSIONS

In the past, we have studied dynamical susceptibilities of neat quantum liquids. These simple liquids are not expected to show glassy behavior due to the fact that even on the time scale of computer simulations, when the liquid is rapidly cooled below its melting temperature, the system simply

FIG. 5. Plots of the Kubo transform of the partial memory kernel at $n = 0.8/s^{1.3}$. Left, middle, and right panels are for 11, 22, and 21 components, respectively. The upper panels are the results for $q = q_{ij}^{\text{max}}$ and the lower panels are the results for $q = q_{ij}^{\text{max}}/2$. The dotted-dashed, solid, dashed, and dotted curves are for $k_B T/\varepsilon_1 = 0.3, 0.4, 0.5$, and 0.67, respectively. Log-log scale is used only for the diagonal elements, since the off-diagonal assume negative values.
Our study has focused on the formulation of a quantum mode-coupling theory for density fluctuations, although other correlations can be studied following similar lines. We have applied the theory to study density fluctuations in a quantum version of the Kob–Andersen model for binary mixtures. In this respect, only predictions of the theory can be made, since experimental results are not available, and all other computational methods are limited to equilibrium short-time dynamics, far from the relevant time scales of slow dynamics expected for the supercooled quantum state.

Our study has addressed several open questions regarding the dynamics and thermodynamics of structural quantum glass forming liquids. We showed that it is possible to supercool a quantum mixture. Furthermore, there are indications that the onset temperature for slow relaxation occurs at temperatures higher than the predicted superfluid transition for this mixture. In this work we have only focused on mildly supercooled liquids. This has allowed us to stay within the framework of our previous approximations to the various vertices that appear in the quantum mode-coupling theory. In future work we will calculate these vertices in a numerically exact fashion via quantum Monte Carlo. This will allow us to go to much lower temperatures where the interplay between quantum fluctuations and glassy behavior is expected to be quite rich. Indeed, understanding the interplay between glassy fluctuations and quantum fluctuations in systems that exhibit rich quantum behavior is largely unexplored in systems without quenched disorder. One possible scenario is the interplay between the glass transition and a competing transition to a superfluid state. This will be investigated in future work.

Finally, we would like to point out one possible experimental realization of a structural quantum glass: a mixture of ortho-deuterium and para-hydrogen. Indeed, preliminary results described elsewhere indicate that it is possible to supercool a quantum mixture of ortho-deuterium and para-hydrogen, while a similar classical system crystallizes. This is due to quantum fluctuations that stabilize the mixture. Semiclassically, these fluctuations lead to an effective size dispersion that arises from the different masses of the particles. Furthermore, the onset temperature for slow relaxation seems to occur at temperatures higher than the predicted transition to the superfluid phase. Indeed, the hydrogen mixture may thus be an ideal playground to understand the interplay between supercooling and superfluidity.

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