Role of the Pair Correlation Function in the Dynamical Transition Predicted by Mode Coupling Theory

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In a recent study, we have found that for a large number of systems the configurational entropy at the pair level S_{c2}, which is primarily determined by the pair correlation function, vanishes at the dynamical transition temperature T_c. Thus, it appears that the information of the transition temperature is embedded in the structure of the liquid. In order to investigate this, we describe the dynamics of the system at the mean field level and, using the concepts of the dynamical density functional theory, show that the dynamical transition temperature depends only on the pair correlation function. Thus, this theory is similar in spirit to the microscopic mode coupling theory (MCT). However, unlike microscopic MCT, which predicts a very high transition temperature, the present theory predicts a transition temperature that is similar to T_c. This implies that the information of the dynamical transition temperature is embedded in the pair correlation function.

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The structure of a liquid at the pair level, described by the pair correlation function, which is directly accessible in experiments, plays an important role in the theory of the liquid state. For systems with pair additive potentials, thermodynamic quantities can be expressed in terms of the pair correlation function. However, it is not clear [1,2] whether the information contained in the pair correlation function is sufficient to predict the dynamics of the liquid. To explore this issue, some of us considered in a recent work [3] the two-body contribution S_2 to the excess entropy [4–7], which is given in terms of the pair correlation function, and the pair configurational entropy S_{c2}, which can be obtained from S_2 and the vibrational entropy S_{vib}. We showed that both S_2 and S_{c2} are sensitive to small changes in the structure and S_{c2} appears to vanish at the dynamical transition temperature T_c [3,8,9]. The temperature T_c is obtained by fitting the temperature dependence of the α relaxation time obtained from simulations [8,10,11] or experiments [12,13] to a power-law form predicted by both the standard mode coupling theory (MCT) F_2 model [14] and the schematic form of the generalized MCT [15].

The connection between the vanishing of S_{c2} and dynamical transition is not understood. The fact that we have observed this connection for multiple systems also at different state points rules out the spurious nature of this connection [3,8,9]. Note that in the calculation of S_{c2} we primarily require the information of the pair correlation function. Surprisingly identical information, when fed into standard microscopic MCT, predicts a transition temperature T_c^{micro} which is much higher than T_c [10,16]. Thus, although the dynamical transition is predicted by MCT, it fails to predict the transition point. This discrepancy between T_c and T_c^{micro} is attributed to the approximations made in the standard MCT. There have been several important studies to improve the theoretical framework [15,17–19]. There have also been studies to attribute a physical meaning to the dynamical transition and power-law behavior observed in simulation and experimental studies [8,10–13]. The p-spin model where the free energy barriers for activated motion diverge in the thermodynamic limit shows a MCT-like power-law behavior and a similar transition temperature [20]. The concepts of the p-spin model have then been extended to a real system [21], where it has been shown that at T_c the saddle order in the landscape appears to vanish [22,23] and the activated dynamics starts to play a dominant role [24].

In this Letter, we attempt to verify if, indeed, there is any information of the dynamical transition temperature embedded in the pair correlation function, as suggested by the vanishing of S_{c2} value [3,8,9]. In analogy with mean field descriptions of spin systems, in which the thermodynamics of a system of interacting spins is approximated by that of a single spin in an effective field, we formulate a description of the dynamics of a collection of interacting particles in terms of that of a single particle in an effective potential, which may be viewed as the “caging potential” created by the neighbors of the particle being considered. We obtain the effective potential in terms of the equilibrium pair correlation function and calculate the mean first passage time for activated escape from this potential. Unlike other studies [15,17–19], the present study does not attempt to develop a better theoretical framework to study the full dynamics of a supercooled liquid. The aim is to study the role of the structure on the dynamics only at the pair level, so by construction, the theory does not take into account the effects of higher order correlations and is not expected to provide the full dynamics.

Using mean field approximation, the time evolution of the density can be written in terms of the following Smoluchowski equation [25]

$$\frac{\partial\rho(\mathbf{r},t)}{\partial t} = \boldsymbol{\nabla}\cdot\left(\frac{1}{\xi}[k_BT\boldsymbol{\nabla}\rho(\mathbf{r},t)+\rho(\mathbf{r},t)\boldsymbol{\nabla}\Phi(\mathbf{r})]\right). \quad (1)$$

Here $\rho(r,t)$ is not the average liquid density but describes the density of each particle in an effective potential $\Phi(r)$, which leads to its caging. In the spirit of mean field theory, Eq. (1) can also describe the dynamics of a set of noninteracting particles in an external potential $\Phi(r)$.

To calculate the potential, we use the dynamic density-functional theory (DDFT) approach, where $\Phi(r) = \{[\delta F_{\mathrm{ex}}[\rho(r)]]/\delta\rho(r)\}$. There have been earlier studies using the DDFT approach, where F_{ex}, the excess part of the free energy functional, is obtained from the Ramakrishnan-Yussouff (RY) form and the $\rho(r,t)$ was numerically solved using iterative techniques. In these studies, the time evolution of the density showed caging and glassy behavior [29,30].

In the present work, we do not use the iterative technique but follow a set of arguments given in Refs. [31,32] to obtain the approximate form of the potential. The standard RY form for excess free energy is [33] $F_{\mathrm{ex}} \simeq -\frac{1}{2}\int d\mathbf{R}\int d\mathbf{R}'\rho(\mathbf{R})C(|\mathbf{R}-\mathbf{R}'|)\rho(\mathbf{R}') = -\frac{1}{2}\int[d\mathbf{q}/(2\pi)^3]\times C_q\rho_{\mathbf{q}}^2$, where $\rho(\mathbf{R}) = \langle\sum_i\delta(\mathbf{R}-\mathbf{R}_i)\rangle$ and $C(|\mathbf{R}'|)$ is the direct correlation function. To dynamically close the theory, we make the Vineyard approximation and write $\rho^s(R,t) \simeq [3/2r_i^2(t)]^{3/2}e^{[-3R^2/2r_i^2(t)]}$, thus $\rho_q^s = e^{[-q^2r_i^2(t)/6]}$. Next we drop the particle index and assume $r_i^2(t) = r^2$. Then the approximate form of the potential becomes [25]

$$\begin{aligned}\Phi(r) &\simeq -\frac{1}{2}\int\frac{d\mathbf{q}}{(2\pi)^3}C_q\left(N^{-1}\sum_{i\neq j}e^{-i\mathbf{q}\cdot\mathbf{R}_{\mathbf{ij}}^{(0)}}\right)e^{-q^2r^2/3}\\ &= -\frac{1}{2}\int\frac{d\mathbf{q}}{(2\pi)^3}\rho C_q^2S_qe^{-q^2r^2/3}. \quad (2)\end{aligned}$$

In Eq. (2), we neglect the self-term ($i = j$) [31,32,34], as it is known to produce unphysical results [35] and also note that we are interested in obtaining the interaction only due to the other particles. Here S_q is the static structure factor of the system. Thus, the mean field potential $\Phi(r)$ is described only in terms of the pair structure of the liquid. Note that, since Eq. (1) describes the dynamics of independent particles in this potential $\Phi(r)$, the dynamics does not have any contribution from higher order correlations beyond two body.

The form of F_{ex} in the present formalism is similar to that in the earlier studies of Schweizer and co-workers [32,34]. However, there is a difference in the way the dynamics is calculated. Schweizer and co-workers used Kramers theory to calculate the dynamics, where the information of only the free energy barrier height is required. In the present formalism, we use the mean first passage time (MFPT) formalism, where the information of the full potential energy surface is required [36]. The MFPT τ_{mfpt} is the time taken to escape the potential $\Phi(r)$ and is given by [25]

$$\tau_{\mathrm{mfpt}} = \frac{1}{D_0}\int_0^{r_{\max}}e^{\beta\Phi(y)}dy\int_0^y e^{-\beta\Phi(z)}dz, \quad (3)$$

where $D_0 = k_BT/\xi$ and $r_{\max}$ is the range of the localization potential $\Phi(r)$.

Since our systems are binary [37], we calculate τ_{mfpt} from the binary form of Eq. (3) [25]. The first interesting result is that, for all the systems [37], $1/D_0\tau_{\mathrm{mfpt}}$ shows a power-law divergence (Fig. 1) and can predict the transition temperature T_{mfpt} (Table I). Note that T_{mfpt} is similar to the dynamical transition temperature T_c (also given in Table I), obtained by fitting the simulated diffusion coefficient values (Fig. 1, right panel) to the power-law behavior. Interestingly, the temperature regime over which $1/D_0\tau_{\mathrm{mfpt}}$ shows power-law behavior is similar to that of the diffusion (Fig. 1) and relaxation time [8].

To show that the present analysis is sensitive to small changes in structure, we compare the results for the Kob-Andersen binary Lennard-Jones (KALJ) system and its repulsive counterpart the Weeks-Chandler-Andersen (KAWCA) system at $\rho = 1.2$. In Fig. 2, we plot $\Phi(r)$, which is used to calculate τ_{mfpt} [via Eq. (3)] and show that, although the structures are similar, the potentials are different. The strength of the present formulation is that this difference is enough to predict the difference in the dynamics and in T_{mfpt} (see Table I). Berthier and Tarjus [2] used the scheme of Schweizer and co-workers [32,34] and found that, at $\rho = 1.2$, it failed to show the difference in the dynamics between KALJ and KAWCA systems. As mentioned earlier, although their expression of F_{ex} is similar to ours, in their scheme, the information of only the free energy barrier height was used for the calculation of the dynamics. Also, the calculation was performed for an effective one component description of the binary system [2]. Note that, in the present formalism, to observe the difference in the dynamics, it is important to consider a full binary system at the level of partial structure factors. However, we make an approximation that the displacement of the two species are the same [see Ref. [25] for the derivation of the binary form of Eq. (2)]. The MCT, on the other hand, even at the binary level, fails to predict the difference in the dynamics [1]. In microscopic MCT, the information of the difference in the structure enters through the vertex [14]. It is possible that, due to the strong temperature dependence of the vertex [8] and the well-known feedback mechanism [14], the MCT is not sensitive to these differences in the structure entering through the vertex and thus predicts the dynamics to be closer. However, identifying the exact cause of failure of MCT [2] and of Schweizer's scheme [32] in predicting the

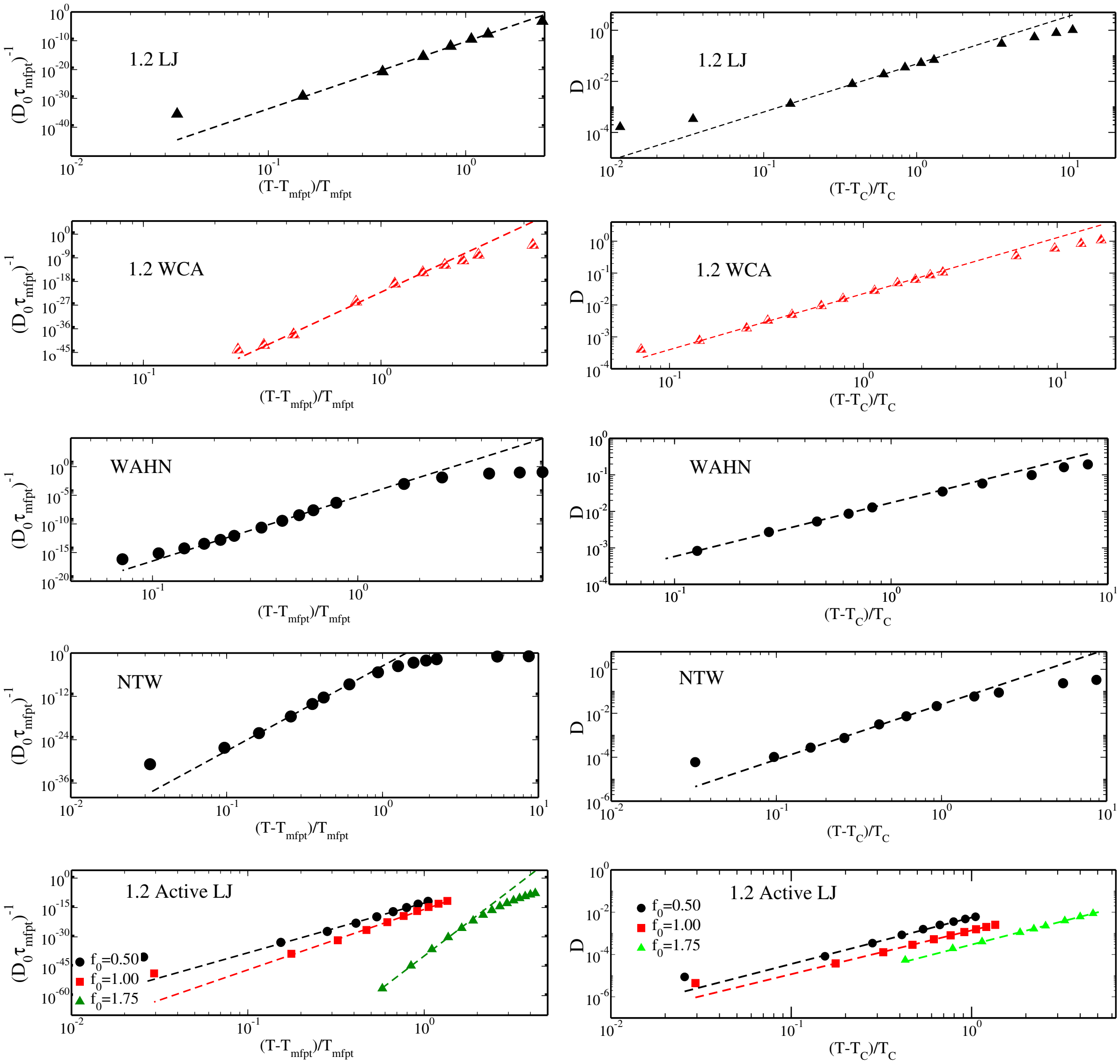

FIG. 1. The power-law dependence of $(D_0\tau_{\mathrm{mfpt}})^{-1}$ predicts a transition temperature T_{mfpt} (left) and the same for total diffusivity (right). The dashed lines are the power-law fits. For active systems, f_0 is the activity as described in Ref. [36]. For clarity, the ln D plot is shifted for $f_0 = 1.0$ by -1. Here, for all the systems, we take $D_0 = 1$.

difference in the dynamics for KALJ and KAWCA systems will require further investigation.

We find that this present method of mean first passage time calculation can also predict the density effect. In Fig. 3, we plot the $1/D_0\tau_{\mathrm{mfpt}}$ for KALJ and KAWCA systems for three different densities. We also plot the corresponding diffusion coefficients in the inset. Note that the $1/D_0\tau_{\mathrm{mfpt}}$ show similar behavior as the diffusion

TABLE I. Transition temperatures (T^*) are shown. T_{mfpt} values are obtained by fitting $1/D_0\tau_{\mathrm{mfpt}}$ to power law. T_c values are obtained by fitting the simulated diffusion values to power-law KALJ [1], KAWCA [1], active LJ [38], WAHN [39], and NTW [40]. T_c^{micro} is calculated from solving the microscopic MCT equation [1,8,10] [Eq. (6)].

	KALJ			KAWCA			Active LJ				
T^*	$\rho = 1.2$	$\rho = 1.4$	$\rho = 1.6$	$\rho = 1.2$	$\rho = 1.4$	$\rho = 1.6$	$f_0 = 0.50$	$f_0 = 1.00$	$f_0 = 1.75$	NTW	WAHN
T_{mfpt}	0.428 ±0.022	0.94 ±0.029	1.757 ±0.042	0.283 ±0.005	0.824 ±0.04	1.691 ±0.018	0.38 ±0.004	0.335 ±0.006	0.196 ±0.013	0.308 ±0.012	0.566 ±0.013
T_c	0.435	0.93	1.76	0.28	0.81	1.69	0.39	0.34	0.19	0.31	0.56
T_c^{micro}	0.887	1.868	3.528	0.76	1.771	3.33	0.768	0.761	0.747	0.464	0.87

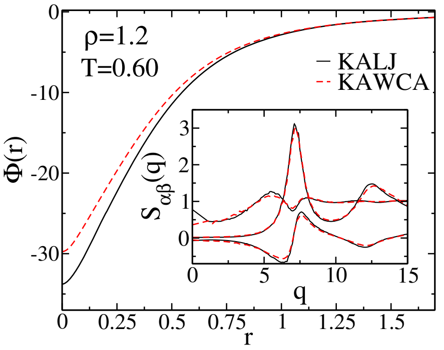

FIG. 2. Plot of potential energy surface $\Phi(r)$ vs r at temperature $T = 0.60$ and density $\rho = 1.2$ for KALJ and KAWCA systems. (Inset) The plots for partial structure factor $S_{\alpha\beta}(q)$ vs q.

coefficients. At low density, the values for the two systems are apart and at high density they overlap.

Next we show that we can derive the standard microscopic MCT equation from our present formalism. Equation (1) is expressed in terms of the fluctuation of density, $\delta\rho$ [$\delta\rho(r) = \rho(r) - \rho_0$, where ρ_0 is the average density] and $\Phi(r)$ is replaced in terms of F_{ex}. We use the RY expression of F_{ex} [33], and after taking the functional derivative and then the gradient of it, we can write

$$\xi\frac{\partial\delta\rho_{\mathbf{k}}(t)}{\partial t} = -k_B T k^2(1 - \rho_0 C_k)\delta\rho_{\mathbf{k}}(t) + \frac{k_B T}{2}\int_q d\mathbf{q}[\mathbf{k}\cdot\mathbf{q}C_q + \mathbf{k}\cdot(\mathbf{k}-\mathbf{q})C_{|\mathbf{k}-\mathbf{q}|}] \times \delta\rho_{\mathbf{q}}(t)\delta\rho_{\mathbf{k}-\mathbf{q}}(t). \quad (4)$$

Following Kawasaki's arguments that the last term on the right-hand side of the above equation contains the products of two ρ's at different wave vectors, so it behaves in an irregular manner like thermal noise, this equation can be considered as the Langevin equation for the density field [41]. In non-Markovian limit applying fluctuation dissipation relation (FDR) [41], the friction ξ can be replaced by a short time part of friction γ and a memory function $\mathcal{M}_k(t)$. Thus, we can rewrite Eq. (4) as

$$\gamma\frac{\partial\delta\rho_{\mathbf{k}}(t)}{\partial t} + \frac{k_B T k^2}{S_k}\delta\rho_{\mathbf{k}}(t) + \int_0^t ds\mathcal{M}(k, t-s)\frac{\partial\delta\rho_{\mathbf{k}}(t)}{\partial s} + \mathcal{R}_{\mathbf{k}}(t) = 0, \quad (5)$$

where $\mathcal{R}_{\mathbf{k}}(t)$ is the new thermal noise and $\mathcal{M}_k(t) = \{[\langle\mathcal{R}_k(t)\mathcal{R}_{-k}(0)\rangle]/k_B TV\} = [(k_B T\rho_0)/16\pi^3]\int d\mathbf{q}\{\hat{k}\cdot[\mathbf{q}C_q + (\mathbf{k}-\mathbf{q})C_{|\mathbf{k}-\mathbf{q}|}]\}^2 S_q(t)S_{k-q}(t)$ [25]. Here we use Gaussian decoupling and Wick's theorem to treat the four-point correlation function. From Eq. (5), we can write a mode coupling theory equation in the overdamped limit for density-density correlation $S_k(t) = \langle\delta\rho_k(t)\delta\rho_{-k}(0)\rangle$

$$\gamma\frac{\partial S_k(t)}{\partial t} + \frac{k_B T k^2}{S_k}S_k(t) + \int\mathcal{M}_k(t-\tau)\dot{S}_k(t)d\tau = 0. \quad (6)$$

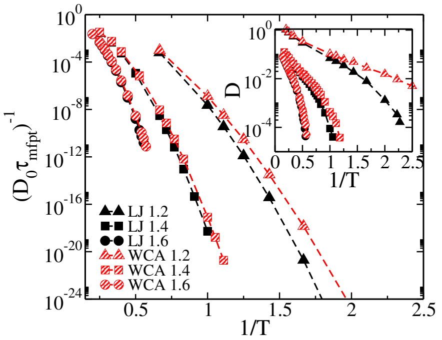

FIG. 3. The plot of $(D_0\tau_{\mathrm{mfpt}})^{-1}$ against inverse temperature for KALJ and KAWCA systems at different densities. We take $D_0 = 1$. (Inset) The plot for diffusion coefficients with inverse temperature.

In Table I, we present the transition temperatures predicted by the binary form of Eq. (6) [42]. We find that for all the systems $T_c^{\mathrm{micro}} \gg T_c$. This higher value of T_c^{micro} can be connected to the Gaussian decoupling approximation used to arrive at Eq. (6). In a recent work, we have also shown that the form of the vertex function in the theory that depends on the structure factor might also be responsible for this premature divergence [8].

In a similar spirit as presented here and also earlier by Kawasaki [41], Zaccarelli *et al.* [43] have mapped the Newtonian dynamics for the density on a Langevin dynamics, where they have divided the force into dissipative term and noise term. The dissipative term was then calculated using the FDR, and similar Gaussian approximations were made. However, in their formalism, only after assuming that the interaction between the densities is given by C_q they could arrive at the MCT expression. Note that this assumption is already present in the RY form of free energy functional used in the present study. Thus, the study of Zaccarelli *et al.* clearly demonstrates the approximations present in MCT and the possible routes of its improvement.

In the present work, in analogy with mean field description of spin systems, we express the dynamics of an interacting particle system in terms of a noninteracting particle system in an effective potential, where the latter provides the effect of the interaction only at the two-body level. Thus, in our formalism, by construction we do not have any effect of correlation beyond two body. We then obtain the mean first passage time, which now depends only on the pair structure of the liquid. The temperature dependence of it can predict a transition temperature that is similar to T_c. This result is similar to the earlier observation of the vanishing of S_{c2} at T_{k2}, where $T_{K2} \simeq T_c$ [8,9]. Thus, our study provides a physical meaning to the dynamical transition temperature and shows that the structure at the pair level has the information of the transition, which is now predicted by two independent theoretical frameworks. We also show that the formalism is sensitive to small changes in structure and can predict that the dynamics is different even for systems having similar structures [1,3,8]. We further show that, starting from the mean field equation, we can also derive the microscopic MCT equation. However, the transition temperature predicted by the microscopic MCT is much higher than the T_c value. Thus, the break down of microscopic MCT in predicting the transition temperature can be connected to the Gaussian decoupling approximation.

However, although our present formalism predicts the correct transition temperature, unlike the microscopic MCT, the power-law exponent is not universal. Also note that this present method of deriving the transition temperature is intimately connected to the structure of the liquid. Thus, for systems like pinned particles, as the structure remains same, this theory will not be able to predict different transition temperatures for different pinning densities [44].

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