



Phenomenological theory of a renormalized simplified model based on time-convolutionless mode-coupling theory near the glass transition



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HIGHLIGHTS

- Time-convolutionless mode-coupling theory is used to describe the dynamics near the glass transition.
- Renormalized simplified model is proposed.
- Renormalized recursion equation is solved for fragile liquids and strong liquids.
- A novel difference between the dynamics of fragile liquids and that of strong liquids is discussed.

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ABSTRACT

The renormalized simplified model is proposed to investigate indirectly how the static structure factor plays an important role in renormalizing a quadratic nonlinear term in the ideal mode-coupling memory function near the glass transition. The renormalized simplified recursion equation is then derived based on the time-convolutionless mode-coupling theory (TMCT) proposed recently by the present author. This phenomenological approach is successfully applied to check from a unified point of view how strong liquids are different from fragile liquids. The simulation results for those two types of liquids are analyzed consistently by the numerical solutions of the recursion equation. Then, the control parameter dependence of the renormalized nonlinear exponent in both types of liquids is fully investigated. Thus, it is shown that there exists a novel difference between the universal behavior in strong liquids and that in fragile liquids not only for their transport coefficients but also for their dynamics.

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1. Introduction

Study of glass transition must be one of last pioneering works left in condensed matter physics [1–4]. Despite decades of many researches on various glass-forming materials, the mechanism of the glass transition is not fully elucidated theoretically yet. The main purpose of the present paper is to propose a phenomenological theory to understand the glass transition phenomena from a unified point of view. This is done by introducing a renormalized simplified model based on the time-convolutionless mode-coupling theory (TMCT) proposed recently by the present author [5]. Then, it is shown indirectly how the static structure factor plays an important role in renormalizing the quadratic nonlinear term contained in the ideal mode-coupling memory function [6]. Thus, this approach is successfully applied to investigate a novel difference between two types of glass-forming liquids classified by Angell [7], strong liquids such as SiO_2 and GeO_2 and fragile liquids such as Toluene and Salol, not only for their transport coefficients but also for their dynamics near the glass transition.

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In order to study the dynamics of supercooled liquids, we have recently proposed the time-convolutionless mode-coupling theory (TMCT) and derived the TMCT equation for the intermediate scattering function $f_\alpha(q, t)$ [5], where $\alpha = c$ for collective case and $\alpha = s$ for self case. Starting from the TMCT equation and introducing the cumulant function $K_\alpha(q, t)$ by $K_\alpha(q, t) = -\ln(f_\alpha(q, t))$, we have then derived the second order differential equation for $K_\alpha(q, t)$ [8]

$$\frac{\partial^2 K_\alpha(q, t)}{\partial t^2} = \frac{q^2 v_{th}^2}{S_\alpha(q)} - \gamma_\alpha \frac{\partial K_\alpha(q, t)}{\partial t} - \int_0^t \Delta\varphi_\alpha(q, t-s) \frac{\partial K_\alpha(q, s)}{\partial s} ds \quad (1)$$

with the ideal MCT nonlinear memory function $\Delta\varphi_\alpha(q, t)$ [6], where γ_α is a positive constant, and v_{th} an average thermal velocity. Here $S_c(q) = S(q)$ and $S_s(q) = 1$, where $S(q)$ is a static structure factor. This equation has the same form as that of the ideal MCT equation for $f_\alpha(q, t)$ [6], except the first term. As discussed in the previous papers [8,9], therefore, all the mathematical predictions proposed in the ideal MCT equation [6,10] can be directly applicable to the ideal TMCT Eq. (1). One of such examples is an existence of a critical point. In fact, similarly to MCT, there exists an ergodic to non-ergodic transition at a critical point, above which $f_\alpha(q, t)$ reduces to the following non-zero solution $f_\alpha(q)$ for a long time:

$$f_\alpha(q) = \exp\left[-\frac{1}{\mathcal{F}_\alpha(q)}\right], \quad (2)$$

where $\mathcal{F}_\alpha(q)$ the long-time limit of the memory function [5,8]. Since this solution $f_\alpha(q)$ is different from that of MCT, the critical point is expected to be quite different from that of MCT. In the previous paper [8], this has been first investigated clearly by employing the simplified model proposed in the original MCT paper [6]. Then, in Ref. [11] it has been also checked directly by solving Eq. (1) numerically for the Percus–Yevick model [12]. Thus, the critical point of TMCT has been shown to coincide with the singular point of the master curve for the diffusion coefficient [13,14]. As is shown later, this relation is in general true even for other systems.

The memory function $\Delta\varphi_\alpha(q, t)$ contains the static structure factors. Once they are known, therefore, it is easy to solve Eq. (1) numerically. Thus, one can compare the numerical solutions with the simulation results and the experimental data one by one. Even though those solutions are obtained numerically, however, it is still not clear how the wavevector dependence of the static structure factors plays an important role in the dynamics of supercooled liquids. In the present paper, therefore, we investigate this from a viewpoint based on the simplified model. Under the original simplified model proposed in MCT, the memory functions can be written as [8]

$$\Delta\varphi_\alpha(q_m, t) = B_\alpha f_c(q_m, t) f_\alpha(q_m, t), \quad (3)$$

$$\mathcal{F}_\alpha(q) = \kappa_\alpha f_c(q_m) f_\alpha(q_m), \quad (4)$$

where κ_α is a coupling parameter, $B_\alpha (= q_m^2 v_{th}^2 \kappa_\alpha / S_\alpha(q_m))$ a dynamic coupling parameter, and q_m a peak position of $S(q)$. In the previous paper [8], we have first transformed Eq. (1) into the recursion equation for $K_\alpha(q_m, t)$ and solved it by iteration under the initial conditions obtained from the simulation results [15] for the binary mixtures with the Stillinger–Weber potential [16]. Thus, we have shown that the simplified model can describe the simulation results very well only in a liquid state but not in a supercooled state at all. In this sense, TMCT based on the simplified model is just a mean-field theory to describe the dynamics in a liquid state. Thus, the integration of the static structure factors over wavevector in the memory function is turned out to be indispensable to describe the dynamics in a supercooled state. In order to check how such an integration affects the dynamics, we assume that the quadratic nonlinear terms given in Eqs. (3) and (4) can be replaced by the renormalized nonlinear terms as

$$\Delta\varphi_\alpha(q_m, t) = B_\alpha [f_c(q_m, t) f_\alpha(q_m, t)]^{1+w_\alpha}, \quad (5)$$

$$\mathcal{F}_\alpha(q) = \kappa_\alpha [f_c(q_m) f_\alpha(q_m)]^{1+w_\alpha}, \quad (6)$$

respectively, where w_α is a renormalized nonlinear exponent to be determined. Here $w_\alpha > 0$ in a supercooled state and a glass state, while $w_\alpha = 0$ in a liquid state. The renormalized exponent w_α is then obtained by comparing the numerical solutions of the recursion equation for $K_\alpha(q_m, t)$ with the simulation results. In fact, by adjusting the value of w_α appropriately, the numerical solutions are shown to agree with the simulation results well within error.

In the previous papers [13,14,18–22], we have proposed the master curve for the diffusion coefficient and shown how strong liquids (S) are different from fragile liquids (F) not only quantitatively but also qualitatively. Thus, we have found the renormalized exponent ε_i to distinguish both liquids from each other, where $i = F$ for (F) and $i = S$ for (S). In fact, ε_i is given by $\varepsilon_F \simeq 4/3$ and $\varepsilon_S \simeq 5/3$. We note here that the renormalized exponent ε_i depends only on types of liquids but not on the details of materials. Although the difference between the exponents in both liquids is small, this suggests that there exist qualitatively different mechanisms between (F) and (S). In fact, in Ref. [18], we have shown that the dynamics of self-diffusion process in (S) is quite different from that in (F) by analyzing the simulation results. In this paper, we apply the present approach to investigate such a difference consistently and to check whether a universality on ε_i also holds on w_α or not. In Fig. 1, the exponent w_c is plotted versus scaled temperature T_c/T for both types of liquids, where T_c is a singular temperature of the master curve. As a typical example of (F), we here take the simulation results [15,18] obtained for the Stillinger–Weber binary mixtures, while for (S) we take the simulation results [18,22] obtained for SiO_2 with the Nakano–Vashishta potential [17]. Thus, the temperature dependence of w_c in (S) is shown to be quite

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