

Mode-Coupling Theory of the Glass Transition

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The **Mode-Coupling Theory (MCT)** is the only known theory about glass transition that are first-principles-based [7, 9]. It uses the Mori-Zwanzig formalism [11] to integrate out unnecessary degrees of freedom and focuses on quantities that characterize glasses.

1 A review of Mori-Zwanzig formalism

First we have a brief review of the Mori-Zwanzig formalism. It says that any time-dependent quantity A obeying the (generalized) Heisenberg equation

$$dA/dt = i\mathcal{L}A \quad (1)$$

also obeys the closed-form equation

$$\dot{A}(t) = i\Omega A(t) - \int_0^t ds K(s)A(t-s) + F(t). \quad (2)$$

The three terms on the RHS are named as the **frequency matrix**, the **memory function**, and the **fluctuating force**, respectively. The fluctuating force collects all “fast” variables that are orthogonal to A , and the memory function is the time autocorrelation function of the fluctuating force. These two terms represent how A gets connected to (in the case in a quantum theory, entangled with) the degrees of freedom that are ignored. Assuming we already have an inner product defined on physical quantities, which is usually

$$(A, B) = \langle A^* B \rangle, \quad (3)$$

The complex conjugate is motivated by the same argument in quantum field theories, i.e. if A, B are Fourier components of real variables φ, ψ , then $\langle \varphi \psi \rangle$ can be expanded into a sum of $\langle A^* B \rangle$ terms, where A and B are indexed by the same frequency. We have

$$i\Omega = (A, i\mathcal{L}A)(A, A)^{-1}, \quad (4)$$

$$F(t) = e^{it(1-\mathcal{P})\mathcal{L}}i(1-\mathcal{P})\mathcal{L}A = e^{it(1-\mathcal{P})\mathcal{L}}(\dot{A} - i\Omega A), \quad (F(t), A(0)) = 0, \quad (5)$$

and

$$K(t) = -(i\mathcal{L}F(t), A)(A, A)^{-1} = (F(0), F(t))(A, A)^{-1}, \quad (6)$$

where

$$\mathcal{P}X = (A, A)^{-1}(X, A)A. \quad (7)$$

Note that the convention of notation varies in the literature, and the two expressions of $K(t)$ in (6) can both be seen. We require A to be “slow” variables (or satisfy other conditions that somehow separate it from other degrees of freedom), or otherwise fluctuation is too strong for A to be a useful quantity.

Of course we can have several slow variables and several fast variables. Therefore, we may replace A by a vector. Note that at this time, Ω, F and K are matrices, and it can be found that (3) should be replaced by

$$\langle A, B \rangle = \langle A^* B^\top \rangle, \quad (8)$$

instead of

$$\langle A, B \rangle = \langle A^\dagger B \rangle.$$

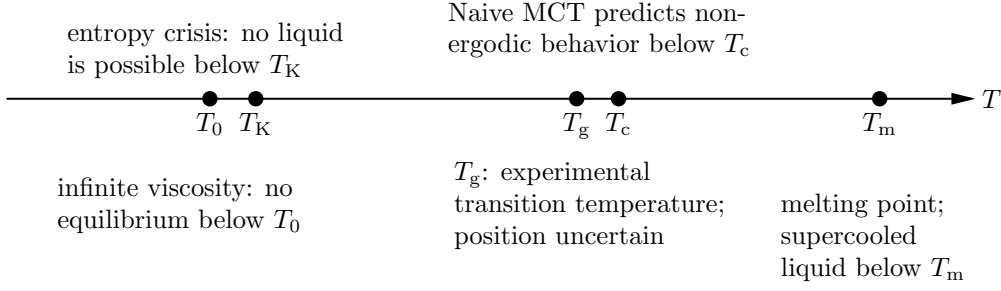


Figure 1: Important temperature points. As the temperature goes down, first the liquid becomes overcooled, and if the liquid is fragile, two-step relaxation occurs. The T_c is a dynamic phase transition

2 Motivation of MCT

Now we go back to derive a theory about glass transition. **Mode coupling theory** is a so-called *generalized* hydrodynamic theory for glasses [3, 4] in that it still only considers density fluctuations in the system, but the density fluctuations are not smoothened. We will find a closed equation about the density-density correlation function, and observe its time evolution.

Here we consider some motivation of MCT. We already know that glasses are like liquids in that there is no broken spacial translational symmetry, but glasses are not just (simple) liquids in that there are some weird things happening. That is the reason we need a *hydrodynamic* theory which focuses on density modes, which can be measured directly using scattering experiments [9], but the theory must be more than ordinary Navier-Stokes equations to include microscopic details. We know glass transition is of kinetic nature, and therefore a quantitative theory of it must be a dynamic one. A hydrodynamic theory is a dynamic theory, but it does not capture essential microscopic details of a supercooled liquid and therefore cannot explain glass transition. A microscopic thermodynamic theory (or a theory invoking thermodynamic concepts to explain the dynamic behavior of a system, for example the Adam-Gibbs theory [10]) captures the microscopic details relevant in glass transition, but is unable to capture some dynamic details like two-step relaxation. We hope a generalized hydrodynamic theory can mix the advantages of the two approaches together.

Note that the spacial translation symmetry gives

$$\langle \rho(0, 0) \rho(\mathbf{r}, t) \rangle = \frac{1}{V} \int \frac{d^3 \mathbf{k}}{(2\pi)^3} e^{-i\mathbf{k} \cdot \mathbf{r}} \langle \rho_{-\mathbf{k}}(0) \rho_{\mathbf{k}}(t) \rangle, \quad (9)$$

and since no valuable information is provided when $|\mathbf{r}| \rightarrow 0$, we will work on the correlation function in the momentum space to separate different spatial scales. What we are going to do is to find a self-consistent equation about the density-density correlation function in the small momentum region (or the large $|\mathbf{r}|$ region). We denote the correlation function as

$$F(k, t) = \frac{1}{N} \langle \rho_{-\mathbf{k}}(0) \rho_{\mathbf{k}}(t) \rangle = \frac{1}{N} \sum_{ij} \left\langle e^{-i\mathbf{k} \cdot \mathbf{r}_i(0)} e^{i\mathbf{k} \cdot \mathbf{r}_j(t)} \right\rangle, \quad (10)$$

where

$$\begin{aligned} \rho_{\mathbf{k}}(t) &= \int d^3 \mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} \rho(\mathbf{r}, t) \\ &= \sum_i \int d^3 \mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} \delta(\mathbf{r} - \mathbf{r}_i(t)) \\ &= \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i(t)}. \end{aligned} \quad (11)$$

Specifically, we have

$$F(k, 0) = S(k), \quad (12)$$

which is the static structural factor.

Note

If we are discussing a quantum system, we need to be more clear here about whether the density modes we are talking about are operators or expectations. If we are talking about a theory of density operators, this is known as (a hydrodynamic flavor of) bosonization. If we are talking about a theory of expectation of density operators, this is known as (hydrodynamic approach) of kinetic theory or (dynamic) density functional theory. All these approaches are different in a quantum many-body theory context, but since we are talking about a system where quantum fluctuation is not important, we can consider these approaches being equivalent, and call them *hydrodynamic* or *generalized hydrodynamic* ones.

3 The exact time-evolution equation of the correlation function

The derivation shown below is mainly based on [9], but the notation is from [4].

We are going to apply the Mori-Zwanzig formalism to $F(k, t)$. We need to find some slow variables and apply (2) to them to find their dynamics, and then we are able to find the dynamics of $F(k, t)$. It can be easily noticed that since we are interested in the small k region, the time derivative

$$\dot{\rho}_{\mathbf{k}} = \sum_i \frac{i\mathbf{k} \cdot \mathbf{p}_i}{m} e^{i\mathbf{k} \cdot \mathbf{r}_i}$$

is also small, and therefore $\rho_{\mathbf{k}}(t)$ is a slow variable. Then we also find that

$$i|\mathbf{k}| j_{\mathbf{k}}^L = i\mathbf{k} \cdot \underbrace{\sum_i \frac{\mathbf{p}_i}{m} e^{i\mathbf{k} \cdot \mathbf{r}_i}}_{j_{\mathbf{k}}}$$

is a slow variable. So the slow variable set is

$$\mathbf{A} = \begin{pmatrix} \delta\rho_{\mathbf{k}} \\ j_{\mathbf{k}}^L \end{pmatrix}, \quad (13)$$

where

$$\delta\rho_{\mathbf{k}} = \rho_{\mathbf{k}} - \langle \rho_{\mathbf{k}} \rangle = \sum_i e^{i\mathbf{q} \cdot \mathbf{r}_i} - (2\pi)^3 \rho \delta(\mathbf{q}). \quad (14)$$

Applying (2) to \mathbf{A} , we have

$$\dot{\mathbf{A}}(t) = i\Omega\mathbf{A}(t) - \int_0^t ds \mathbf{K}(s)\mathbf{A}(t-s) + \mathbf{F}(t),$$

and since

$$\langle \mathbf{A}(0), \mathbf{F}(t) \rangle = \langle \mathbf{A}(0)^* \mathbf{F}(t)^\top \rangle = 0,$$

which is a result in the Mori-Zwanzig formalism, we have

$$\dot{\mathbf{C}} = i\Omega\mathbf{C}(t) - \int_0^t ds \mathbf{K}(s)\mathbf{C}(t-s), \quad (15)$$

where we have

$$\mathbf{C}(t) = \langle \mathbf{A}(0)^* \mathbf{A}(t)^\top \rangle = \begin{pmatrix} \langle \delta\rho_{-\mathbf{q}}(0) \delta\rho_{\mathbf{q}}(t) \rangle & \langle \delta\rho_{-\mathbf{q}}(0) j_{\mathbf{q}}^L(t) \rangle \\ \langle -j_{-\mathbf{q}}^L(0) \delta\rho_{\mathbf{q}}(t) \rangle & \langle -j_{-\mathbf{q}}^L(0) j_{\mathbf{q}}^L(t) \rangle \end{pmatrix}, \quad (16)$$

$$\begin{aligned} i\Omega &= \langle \mathbf{A}, \dot{\mathbf{A}} \rangle \langle \mathbf{A}, \mathbf{A} \rangle^{-1} \\ &= \begin{pmatrix} \langle \delta\rho_{-\mathbf{q}} \delta\dot{\rho}_{\mathbf{q}} \rangle & \langle \delta\rho_{-\mathbf{q}} \delta\dot{j}_{\mathbf{q}}^L \rangle \\ \langle -\delta j_{-\mathbf{q}}^L \delta\dot{\rho}_{\mathbf{q}} \rangle & \langle -\delta j_{-\mathbf{q}}^L \delta\dot{j}_{\mathbf{q}}^L \rangle \end{pmatrix} \begin{pmatrix} \langle \delta\rho_{-\mathbf{q}} \delta\rho_{\mathbf{q}} \rangle & \langle \delta\rho_{-\mathbf{q}} \delta j_{\mathbf{q}}^L \rangle \\ \langle -\delta j_{-\mathbf{q}}^L \delta\rho_{\mathbf{q}} \rangle & \langle -\delta j_{-\mathbf{q}}^L \delta j_{\mathbf{q}}^L \rangle \end{pmatrix}^{-1} \end{aligned} \quad (17)$$

Note that unitarity guarantees that the $\langle \mathbf{A}, \mathbf{A} \rangle$ factor in (17) is constant, and therefore we have

$$\langle \mathbf{A}, \mathbf{A} \rangle = \begin{pmatrix} \langle \delta \rho_{-\mathbf{q}} \delta \rho_{\mathbf{q}} \rangle & \langle \delta \rho_{-\mathbf{q}} \delta j_{\mathbf{q}}^{\text{L}} \rangle \\ \langle \delta j_{-\mathbf{q}}^{\text{L}} \delta \rho_{\mathbf{q}} \rangle & \langle \delta j_{-\mathbf{q}}^{\text{L}} \delta j_{\mathbf{q}}^{\text{L}} \rangle \end{pmatrix} = \begin{pmatrix} NS(q) & 0 \\ 0 & \frac{Nk_{\text{B}}T}{m} \end{pmatrix}, \quad (18)$$

where

$$\langle \delta \rho_{-\mathbf{q}} \delta j_{\mathbf{q}}^{\text{L}} \rangle \propto \mathbf{q} \cdot \langle \mathbf{p} \rangle = 0,$$

and

$$\begin{aligned} \langle -j_{-\mathbf{q}}^{\text{L}} j_{\mathbf{q}}^{\text{L}} \rangle &= \sum_{i,j} \langle (\hat{\mathbf{q}} \cdot \mathbf{v}_i)(\hat{\mathbf{q}} \cdot \mathbf{v}_j) \rangle e^{i\mathbf{q} \cdot (\mathbf{r}_j - \mathbf{r}_i)} \\ &= \sum_{i,j} \delta_{ij} \mathbf{q} \cdot \frac{k_{\text{B}}T}{m} \cdot \mathbf{q} e^{i\mathbf{q} \cdot (\mathbf{r}_j - \mathbf{r}_i)} \\ &= \frac{Nk_{\text{B}}T}{m}. \end{aligned}$$

Here and below we calculate all expectations with the assumption that the translational symmetry is not broken, and therefore the probabilistic distributions of positions and momenta are independent. We also invoke the equipartition theorem. We also should keep in mind the fact that velocities at one time step may be correlated to the past configuration, and therefore *non*-equal time correlation functions $\langle \rho_{-\mathbf{q}}(0) j_{\mathbf{q}}^{\text{L}}(t) \rangle$ is not necessarily zero (or otherwise some weird facts will occur, like a variable is constant but its time derivative is not, etc.). We can also evaluate the $\langle \mathbf{A}, \dot{\mathbf{A}} \rangle$ factor. A variable is always orthogonal to its time derivative, and we have

$$\langle \delta \rho_{-\mathbf{q}} \delta \dot{\rho}_{\mathbf{q}} \rangle = \langle \delta \rho_{\mathbf{q}} \delta \dot{\rho}_{\mathbf{q}} \rangle = 0, \quad \langle -\delta j_{-\mathbf{q}}^{\text{L}} \delta \dot{j}_{\mathbf{q}}^{\text{L}} \rangle = 0,$$

and

$$\begin{aligned} \langle \delta \rho_{\mathbf{q}} \dot{j}_{\mathbf{q}}^{\text{L}} \rangle &= \sum_i \left(\langle \delta \rho_{-\mathbf{q}} \hat{\mathbf{q}} \cdot \dot{\mathbf{v}}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle + \underbrace{\langle \delta \rho_{-\mathbf{q}} \cdot i\mathbf{q} \cdot \mathbf{v}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle}_{\propto \langle \mathbf{v}_i \rangle = 0} \right) \\ &= \sum_i \langle \delta \rho_{-\mathbf{q}} \hat{\mathbf{q}} \cdot \dot{\mathbf{v}}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle \\ &= \sum_{i,j} \langle e^{-i\mathbf{q} \cdot \mathbf{r}_j} \delta \rho_{-\mathbf{q}} \hat{\mathbf{q}} \cdot \dot{\mathbf{v}}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle \\ &= \frac{d}{dt} \sum_{i,j} \underbrace{\langle e^{-i\mathbf{q} \cdot \mathbf{r}_j} \delta \rho_{-\mathbf{q}} \hat{\mathbf{q}} \cdot \mathbf{v}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle}_{\propto \langle \mathbf{v}_i \rangle = 0} - \sum_{i,j} \langle -i\mathbf{q} \cdot \mathbf{v}_j e^{-i\mathbf{q} \cdot \mathbf{r}_j} \delta \rho_{-\mathbf{q}} \hat{\mathbf{q}} \cdot \mathbf{v}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle \\ &= i \sum_{i,j} \langle \mathbf{q} \cdot \mathbf{v}_j e^{-i\mathbf{q} \cdot \mathbf{r}_j} \delta \rho_{-\mathbf{q}} \hat{\mathbf{q}} \cdot \mathbf{v}_i e^{i\mathbf{q} \cdot \mathbf{r}_i} \rangle \\ &= i \sum_{i,j} \mathbf{q} \cdot \frac{k_{\text{B}}T}{m} \hat{\mathbf{I}} \cdot \hat{\mathbf{q}} e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \\ &= \frac{iqNk_{\text{B}}T}{m}, \end{aligned}$$

and the same derivation works for $\langle -\delta j_{-\mathbf{q}}^{\text{L}} \delta \dot{\rho}_{\mathbf{q}} \rangle$. Therefore, (17) evaluates to be

$$i\Omega = \begin{pmatrix} 0 & \frac{iqNk_{\text{B}}T}{m} \\ \frac{iqNk_{\text{B}}T}{m} & 0 \end{pmatrix} \begin{pmatrix} NS(q) & \frac{Nk_{\text{B}}T}{m} \end{pmatrix}^{-1} = \begin{pmatrix} 0 & iq \\ \frac{iqk_{\text{B}}T}{mS(q)} & 0 \end{pmatrix}. \quad (19)$$

Then we need the memory matrix, which in turn requires us to find the fluctuation force. Note that if we are able to evaluate $\mathbf{F}(0)$ into a linear function of $\mathbf{A}(0)$, then we have already obtained $\mathbf{F}(t)$ by replacing $\mathbf{A}(0)$ with $\mathbf{A}(t)$. We have

$$\mathbf{F}(0) = \dot{\mathbf{A}} - i\Omega \mathbf{A} = \begin{pmatrix} \delta \dot{\rho}_{\mathbf{q}} \\ \dot{j}_{\mathbf{q}}^{\text{L}} \end{pmatrix} - \begin{pmatrix} 0 & iq \\ \frac{iqk_{\text{B}}T}{mS(q)} & 0 \end{pmatrix} \begin{pmatrix} \delta \rho_{\mathbf{q}} \\ j_{\mathbf{q}}^{\text{L}} \end{pmatrix} = \begin{pmatrix} 0 \\ j_{\mathbf{q}}^{\text{L}} - \frac{iqk_{\text{B}}T}{mS(q)} \delta \rho_{\mathbf{q}} \end{pmatrix} \Big|_{t=0} =: \begin{pmatrix} 0 \\ R_{\mathbf{q}}(0) \end{pmatrix},$$

and hence we have

$$\mathbf{F}(t) = \begin{pmatrix} 0 \\ j_{\mathbf{q}}^{\text{L}} - \frac{iqk_{\text{B}}T}{mS(q)} \delta \rho_{\mathbf{q}} \end{pmatrix} = \begin{pmatrix} 0 \\ R_{\mathbf{q}} \end{pmatrix}. \quad (20)$$

Now the memory matrix can be obtained straightforwardly:

$$\begin{aligned}\mathbf{K}(t) &= \langle \mathbf{F}(0)^* \mathbf{F}(t)^\top \rangle \langle \mathbf{A}, \mathbf{A} \rangle^{-1} = \begin{pmatrix} 0 & 0 \\ 0 & \langle R_{-\mathbf{q}}(0) R_{\mathbf{q}}(t) \rangle \end{pmatrix} \begin{pmatrix} NS(q) & 0 \\ 0 & \frac{Nk_{\text{B}}T}{m} \end{pmatrix}^{-1} \\ &= \begin{pmatrix} 0 & 0 \\ 0 & \frac{m}{Nk_{\text{B}}T} \langle R_{-\mathbf{q}}(0) R_{\mathbf{q}}(t) \rangle \end{pmatrix}.\end{aligned}\quad (21)$$

Now (15), (16), (19), (21) and the definition of $R_{\mathbf{q}}$ in (20) form a closed group of equations. Actually we are only interested in $F(q, t)$, and we can focus on the left bottom element of (15), which is

$$\begin{aligned}\frac{d}{dt} \langle -j_{-\mathbf{q}}^{\text{L}}(0) \delta \rho_{\mathbf{q}}(t) \rangle &= \frac{iqk_{\text{B}}T}{mS(q)} \langle \delta \rho_{-\mathbf{q}}(0) \delta \rho_{\mathbf{q}}(t) \rangle \\ &\quad - \int_0^t d\tau \frac{m}{Nk_{\text{B}}T} \langle R_{-\mathbf{q}}(0) R_{\mathbf{q}}(\tau) \rangle \langle -j_{-\mathbf{q}}^{\text{L}}(0) \delta \rho_{\mathbf{q}}(t - \tau) \rangle.\end{aligned}\quad (22)$$

Note that the time translation symmetry guarantees

$$\langle \delta \rho_{-\mathbf{q}}(-t) \delta \rho_{\mathbf{q}}(0) \rangle = \langle \delta \rho_{-\mathbf{q}}(0) \delta \rho_{\mathbf{q}}(t) \rangle,$$

and by taking time derivative we have

$$\begin{aligned}\frac{d}{dt} \langle \delta \rho_{-\mathbf{q}}(0) \delta \rho_{\mathbf{q}}(t) \rangle &= \frac{d}{dt} \langle \delta \rho_{-\mathbf{q}}(-t) \delta \rho_{\mathbf{q}}(0) \rangle \\ &= \frac{d}{dt} \sum_i \langle e^{-i\mathbf{q} \cdot \mathbf{r}_i(-t)} \delta \rho_{\mathbf{q}}(0) \rangle \\ &= \sum_i \langle -i\mathbf{q} \cdot (-\mathbf{v}_i(-t)) e^{-i\mathbf{q} \cdot \mathbf{r}_i(-t)} \delta \rho_{\mathbf{q}}(0) \rangle \\ &= iq \langle -j_{-\mathbf{q}}^{\text{L}}(-t) \delta \rho_{\mathbf{q}}(0) \rangle \\ &= iq \langle -j_{-\mathbf{q}}^{\text{L}}(0) \delta \rho_{\mathbf{q}}(t) \rangle,\end{aligned}$$

which means

$$\begin{aligned}\langle -j_{-\mathbf{q}}^{\text{L}}(0) \delta \rho_{\mathbf{q}}(t) \rangle &= \frac{1}{iq} \frac{d}{dt} \langle \delta \rho_{-\mathbf{q}}(0) \delta \rho_{\mathbf{q}}(t) \rangle \\ &= \frac{N}{iq} \frac{dF(q, t)}{dt}.\end{aligned}$$

Substitution of the above equation into (22), we get

$$\frac{d^2 F(q, t)}{dt^2} + \frac{q^2 k_{\text{B}}T}{mS(q)} F(q, t) + \frac{m}{Nk_{\text{B}}T} \int_0^t d\tau \langle R_{-\mathbf{q}}(0) R_{\mathbf{q}}(\tau) \rangle \frac{dF(q, t)}{dt} = 0, \quad (23)$$

where

$$R_{\mathbf{q}} = j_{\mathbf{q}}^{\text{L}} - \frac{iqk_{\text{B}}T}{mS(q)} \delta \rho_{\mathbf{q}}. \quad (24)$$

(23) is exact under the conditions that at least the supercooled liquid is locally in thermal equilibrium, so we have a definition of temperature, that equipartition theorem works for the momenta and that spacial and temporal translational symmetries are not broken, which is obviously correct for glass. However, (23) is *not* closed in that $R_{\mathbf{q}}$ should be evaluated using $\rho_{\mathbf{q}}$ and $j_{\mathbf{q}}^{\text{L}}$ instead of $\langle \delta \rho_{-\mathbf{q}}(0) \delta \rho_{\mathbf{q}}(t) \rangle$. Therefore, we need to derive an explicit expression of $R_{\mathbf{q}}$ using $F(q, t)$. If such an expression is not available, then we are unable to reach a generalized hydrodynamic theory of glass.

4 Closing the MCT equation (23)

Now we need some intuition to have an approximation of $\langle R_{-\mathbf{q}}(0) R_{\mathbf{q}}(\tau) \rangle$.

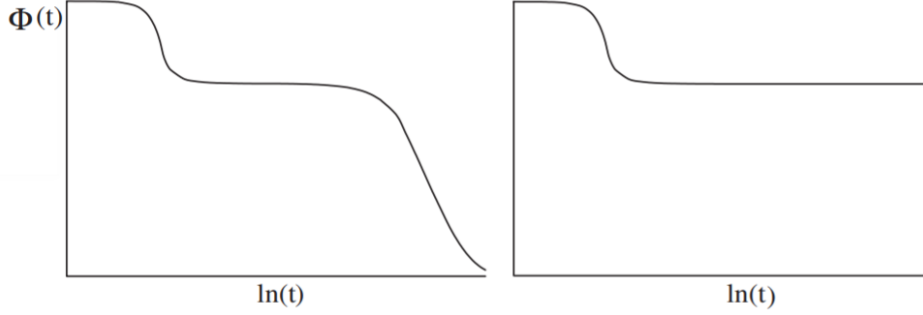


Figure 2: Figure 5 from [9].

5 MCT's solution, its success and its failure

Now we review what can and cannot be captured with MCT.

The largest problem is probably that MCT predicts that there is almost a dynamic phase transition¹ at $T = T_c$ [1], with divergent viscosity and a clear ergodic-non-ergodic distinction. We already know this is not the case. First, we already know that T_g is dependent to the time scale of the observation. Some may argue that because the viscosity is too large to be measured near T_g , but experimentalists doubt that [8]. Even if there is a clear liquid-glass distinction, considering usually we have $T_c > T_g$ [9], the transition temperature predicted by MCT is wrong, because obviously when $T < T_c$ the system is still able to relax and therefore is ergodic. The divergent viscosity formula still works well when we are not too close to T_c . The reason it fails near $T = T_c$ can be attributed to its mean-field nature [2, 7]. (To be exact, MCT is not a typical mean-field theory, either, because it fails with infinite dimensions [6].)

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¹Dynamic phase transition does happen from time to time in non-equilibrium systems. See [5], for example.

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