Mode-Coupling Theory of the Glass Transition

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December 10, 2021

The **Mode-Coupling Theory (MCT)** is the only known theory about glass transition that are first-principles-based [2, 3]. It uses the Mori-Zwanzig formalism [4] 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

$$\mathrm{d}A/\mathrm{d}t = \mathrm{i}\mathcal{L}A \tag{1}$$

also obeys the closed-form equation

$$\dot{A}(t) = \mathrm{i}\Omega A(t) - \int_0^t \mathrm{d}s\, K(s)A(t-s) + F(t). \tag{2}$$

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

$$(A, B) = \langle A^* B \rangle\,, \tag{3}$$

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

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

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

and

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

where

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

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

$$\langle A, B \rangle = \langle A^* B^\top \rangle\,, \tag{8}$$

instead of

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

2 The exact MCT equation

Now we go back to derive a theory about glass transition. **Mode coupling theory** is a so-called *generalized* hydrodynamic theory for glasses [1] in that it still only considers density fluctuations in the system, but the density fluctuations are not smoothened. The derivation shown below is mainly based on [3], but the notation is from [1]. Note that the spacial translation symmetry gives

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

and since no valuable information is provided when $|\boldsymbol{r}| \to 0$, 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 $|\boldsymbol{r}|$ region). We denote the correlation function as

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

where

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

Specifically, we have

$$F(k,0) = S(k), \tag{12}$$

which is the static structural factor.

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

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

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

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

is a slow variable. So the slow variable set is

$$\mathbf{A} = \begin{pmatrix}\delta\rho_{\boldsymbol{k}}\\ j_{\boldsymbol{k}}^{\mathrm{L}}\end{pmatrix}, \tag{13}$$

where

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

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

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

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

$$\dot{\mathbf{C}} = \mathrm{i}\boldsymbol{\Omega}\mathbf{C}(t) - \int_0^t \mathrm{d}s\,\mathbf{K}(s)\mathbf{C}(t-s), \tag{15}$$

where we have

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

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

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

$$\langle \mathbf{A}, \mathbf{A}\rangle = \begin{pmatrix} \langle \delta\rho_{-\boldsymbol{q}}\, \delta\rho_{\boldsymbol{q}}\rangle & \langle \delta\rho_{-\boldsymbol{q}}\, \delta j^{\mathrm{L}}_{\boldsymbol{q}}\rangle \\ \langle \delta j^{\mathrm{L}}_{-\boldsymbol{q}}\, \delta\rho_{\boldsymbol{q}}\rangle & \langle \delta j^{\mathrm{L}}_{-\boldsymbol{q}}\, \delta j^{\mathrm{L}}_{\boldsymbol{q}}\rangle \end{pmatrix} = \begin{pmatrix} NS(q) & 0 \\ 0 & \frac{Nk_{\mathrm{B}}T}{m} \end{pmatrix}, \tag{18}$$

where

$$\langle \delta\rho_{-\boldsymbol{q}}\, \delta j^{\mathrm{L}}_{\boldsymbol{q}}\rangle \propto \boldsymbol{q}\cdot\langle \boldsymbol{p}\rangle = 0,$$

and

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

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 $\langle \rho_{-\boldsymbol{q}}(0) j^{\mathrm{L}}_{\boldsymbol{q}}(t)\rangle$ 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

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

and

$$\begin{aligned} \langle \delta\rho_{\boldsymbol{q}}\, \dot{j}^{\mathrm{L}}_{\boldsymbol{q}}\rangle &= \sum_i \Big(\langle \delta\rho_{-\boldsymbol{q}}\, \hat{\boldsymbol{q}}\cdot\dot{\boldsymbol{v}}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle + \underbrace{\langle \delta\rho_{-\boldsymbol{q}}\cdot\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{v}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle}_{\propto\langle \boldsymbol{v}_i\rangle=0}\Big) \\ &= \sum_i \langle \delta\rho_{-\boldsymbol{q}}\, \hat{\boldsymbol{q}}\cdot\dot{\boldsymbol{v}}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle \\ &= \sum_{i,j} \langle \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_j}\, \delta\rho_{-\boldsymbol{q}}\, \hat{\boldsymbol{q}}\cdot\dot{\boldsymbol{v}}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle \\ &= \frac{\mathrm{d}}{\mathrm{d}t}\sum_{i,j} \underbrace{\langle \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_j}\, \delta\rho_{-\boldsymbol{q}}\, \hat{\boldsymbol{q}}\cdot\boldsymbol{v}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle}_{\propto\langle \boldsymbol{v}_i\rangle=0} - \sum_{i,j} \langle -\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{v}_j \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_j}\, \delta\rho_{-\boldsymbol{q}}\, \hat{\boldsymbol{q}}\cdot\boldsymbol{v}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle \\ &= \mathrm{i}\sum_{i,j} \langle \boldsymbol{q}\cdot\boldsymbol{v}_j \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_j}\, \delta\rho_{-\boldsymbol{q}}\, \hat{\boldsymbol{q}}\cdot\boldsymbol{v}_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}\rangle \\ &= \mathrm{i}\sum_{i,j} \boldsymbol{q}\cdot\frac{k_{\mathrm{B}}T}{m}\overset{\leftrightarrow}{\boldsymbol{I}}\cdot\hat{\boldsymbol{q}}\mathrm{e}^{\mathrm{i}\boldsymbol{q}(\boldsymbol{r}_i-\boldsymbol{r}_j)} \\ &= \frac{\mathrm{i}qNk_{\mathrm{B}}T}{m}, \end{aligned}$$

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

$$\mathrm{i}\boldsymbol{\Omega} = \begin{pmatrix} 0 & \frac{\mathrm{i}qNk_{\mathrm{B}}T}{m} \\ \frac{\mathrm{i}qNk_{\mathrm{B}}T}{m} & 0 \end{pmatrix} \begin{pmatrix} NS(q) & \\ & \frac{Nk_{\mathrm{B}}T}{m} \end{pmatrix}^{-1} = \begin{pmatrix} 0 & \mathrm{i}q \\ \frac{\mathrm{i}qk_{\mathrm{B}}T}{mS(q)} & 0 \end{pmatrix}. \tag{19}$$

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

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

and hence we have

$$\mathbf{F}(t) = \begin{pmatrix} 0 \\ \dot{j}_{\boldsymbol{q}}^{\mathrm{L}} - \frac{\mathrm{i}qk_{\mathrm{B}}T}{mS(q)}\,\delta\rho_{\boldsymbol{q}} \end{pmatrix} = \begin{pmatrix} 0 \\ R_{\boldsymbol{q}} \end{pmatrix}. \tag{20}$$

Now the memory matrix can be obtained straightforwardly:

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

Now (15), (16), (19), (21) and the definition of $R_{\boldsymbol{q}}$ 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

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}\left\langle -j_{-\boldsymbol{q}}^{\mathrm{L}}(0)\,\delta\rho_{\boldsymbol{q}}(t)\right\rangle &= \frac{\mathrm{i}qk_{\mathrm{B}}T}{mS(q)}\left\langle \delta\rho_{-\boldsymbol{q}}(0)\,\delta\rho_{\boldsymbol{q}}(t)\right\rangle \\ &\quad - \int_0^t \mathrm{d}\tau\, \frac{m}{Nk_{\mathrm{B}}T}\langle R_{-\boldsymbol{q}}(0)R_{\boldsymbol{q}}(\tau)\rangle \left\langle -j_{-\boldsymbol{q}}^{\mathrm{L}}(0)\,\delta\rho_{\boldsymbol{q}}(t-\tau)\right\rangle. \end{aligned} \tag{22}$$

Note that the time translation symmetry guarantees

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

and by taking time derivative we have

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

which means

$$\begin{aligned} \left\langle -j_{-\boldsymbol{q}}^{\mathrm{L}}(0)\,\delta\rho_{\boldsymbol{q}}(t)\right\rangle &= \frac{1}{\mathrm{i}q}\frac{\mathrm{d}}{\mathrm{d}t}\langle \delta\rho_{-\boldsymbol{q}}(0)\,\delta\rho_{\boldsymbol{q}}(t)\rangle \\ &= \frac{N}{\mathrm{i}q}\frac{\mathrm{d}F(q,t)}{\mathrm{d}t}. \end{aligned}$$

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

$$\frac{\mathrm{d}^2F(q,t)}{\mathrm{d}t^2} + \frac{q^2k_{\mathrm{B}}T}{mS(q)}F(q,t) + \frac{m}{Nk_{\mathrm{B}}T}\int_0^t \mathrm{d}\tau\,\langle R_{-\boldsymbol{q}}(0)R_{\boldsymbol{q}}(\tau)\rangle\frac{\mathrm{d}F(q,t)}{\mathrm{d}t} = 0. \tag{23}$$

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