Abstract

In this set of lecture notes we review the mode-coupling theory of the glass transition from several perspectives. First, we derive mode-coupling equations for the description of density fluctuations from microscopic considerations with the use the Mori-Zwanzig projection operator technique. We also derive schematic mode-coupling equations of a similar form from a field-theoretic perspective. We review the successes and failures of mode-coupling theory, and discuss recent advances in the applications of the theory.

1 Important Phenomenology for MCT

Since our objective will be to sketch a derivation of what we will call Mode-Coupling Theory (MCT), we will focus our attention on one observable in particular, namely density fluctuations. For this we will first define some of the concepts needed to do so.

We want to calculate a specific time correlation function. In general, such a function is expressed as follows,

$$C(t) = \langle A(t)A(0) \rangle.$$  \hfill (1)

It is an ensemble average of the evolution of the fluctuations of a variable in time, at equilibrium. As seen in Fig. 1, A(t) fluctuates around its average value in equilibrium, while C(t) measures the correlation of A at one time with the value of A at another time.

The density or particles in a liquid can be one example of A(t),

$$\rho(r,t) = \sum_i \delta(r - r_i(t)),$$  \hfill (2)

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Figure 1: Left: time evolution of the instantaneous fluctuations of the quantity $A$. The product of fluctuations separated by time $t'$, averaged over all $t'_0$'s gives the correlation function $C(t')$, at equilibrium. Right: fluctuations of the density on lengthscale $L \sim 2\pi/k$; if $k$ is small, the area of density fluctuations is large.

which we can Fourier transform,

$$
\rho_k(t) = \sum_i \int dr e^{i k \cdot r} \delta (r - r_i(t)),
= \sum_i e^{i k \cdot r_i(t)}.
$$

(3)

In this case the correlation function will be labelled $F(k, t)$, which is can be expressed as follows.

$$
F(k, t) = \frac{1}{N} \langle \rho_{-k}(0) \rho_k(t) \rangle = \frac{1}{N} \sum_{ij} \langle e^{-i k \cdot r_i(0)} e^{i k \cdot r_j(t)} \rangle.
$$

(4)

Note that we need to have $\sum_i k_i = 0$ (i.e. $-k + k = 0$!) to conserve momentum, otherwise the correlation function is equal to zero.

The variables labeled by $k$ measure density fluctuations in reciprocal ("$k = |k|$") space, which can be thought as the inverse length. When $k$ is small we are looking at long lengthscales, as we can see in Fig. 1. When it is large, we are probing very short scales.

The function $F(k, t)$ is essentially what scattering experiments measure. At $t = 0$,

$$
F(k, t = 0) = \frac{1}{N} \langle \rho_{-k}(0) \rho_k(0) \rangle \equiv S(k),
$$

(5)

where $S(k)$ is called the static structure factor of the liquid. Why that name? Consider the radial distribution function of a liquid $g(r)$. The function $g(r)$ is proportional to the probability that a particle is a distance $r$ away from a particle at the origin. In a dense liquid $g(r)$ shows the structure of the solvation shells as depicted in Fig. 2. Also, it can be shown that [1][2][3]

$$
S(k) = 1 + \rho \int dr e^{-i k \cdot r} g(r),
$$

(6)
Figure 2: Left: radial distribution function \( g(r) \) for a simple liquid of size \( \sigma \). Right: the corresponding structure factor \( S(k) \). A sample structure is also depicted where the solvation shells are indicated by the dotted lines. The exclusion radius can be seen in the absence of amplitude of \( g(r) \) for \( r \ll \sigma \).

where \( \rho = N/V \) is the density of the system, and thus \( S(k) \) is also indicating something about the liquid structure. An example for a simple liquid is depicted in Fig. 2.

But how do we expect \( F(k, t) \) to behave? For high temperatures – above the melting point – \( F(k, t) \) will decay like a single exponential function in time for for \( k \geq 2\pi/\sigma \) as plotted in Fig. 3. For supercooled liquids, the situation is different and a characteristic decay pattern can also be seen in Fig. 3. We observe a multi-step relaxation.

1. At short times decay is coming from free and collisional events that involve local particle motion. Consistent with a short-time expansion, \( F(k, t) \sim S(k) - A(k)t^2 + \ldots \) in this regime. This will be true at any temperature. We will not be concerned much with this part of the decay.

2. Intermediate times encompass a period during which particles appear trapped in cages formed by other particles. This regime is the \( \beta \)-relaxation regime. The decay to the plateau (IIa) may be fitted as \( f + At^{-a} \) and the decay from the plateau (IIb) as \( f - Bt^{b} \). Also, the exponents have a scaling consistent with the relationship

\[
\frac{\Gamma(1-a)^2}{\Gamma(1-2a)} = \frac{\Gamma(1+b)^2}{\Gamma(1+2b)}.
\]  

3. At long times, in the \( \alpha \)-relaxation regime, the decay may be fitted to a stretched exponential law

\[
F(k, t) \sim e^{-(\frac{t}{\tau})^\beta}.
\]

with \( 0 < \beta < 1 \). Do not be confused with the notation. It is the \( \beta \) power that appears in the \( \alpha \)-relaxation regime! In general \( \beta \) and \( \tau \) will be \( k \) and temperature dependent.
Figure 3: Left: $F(k, t)$ exhibiting exponential $e^{-t/\tau}$ decay for a normal liquid. Right: supercooled liquids do not have such a simple decay. The various temporal regimes are described in the text. Notice the logarithmic scale.

In a later section, we will return to this kind of phenomenology. For now, we just make a few superficial remarks about things that will be covered in more depth by others in these lectures.

The constant $\tau$ that appears in the stretched-exponential decay law is strongly temperature dependent. All transports coefficients — $D$ (diffusion), $\eta$ (viscosity), etc. — are strongly temperature dependent as well. Over a rather wide range of temperatures, a fit to this temperature dependence may be

$$\eta \sim e^{\frac{E}{T - T_0}}.$$  \hspace{1cm} (9)

Clearly, as $T_0$ is approached, relaxation times become so large that the system cannot stay in equilibrium. Other fitting forms, some that do not imply a divergence at finite temperatures, may be used to fit the data as well.

Some systems, hard-spheres for example, are not characterized by temperature, but by density of packing fraction $\phi = \frac{4}{3} \pi a^3 \rho$, where $a$ is the particle radius. For such systems, one may fit with

$$\eta \sim e^{\frac{B}{\phi - \phi_c}},$$  \hspace{1cm} (10)

or with other forms.

2 The Mode-Coupling Theory of density fluctuations

Our strategy will be to derive an exact equation of motion for $F(k, t)$ and then to make approximations that allow us to solve them. The approximations are uncontrolled, and we will judge them by their success or failure.
2.1 Memory functions

Consider some classical function of phase space variables $A(t)$, where the time dependence originates from that of the positions $r_i$ and of the momenta $p_i$ for a $N$-particle system. We know from Hamilton’s equations that

$$\frac{dA(t)}{dt} = \{A(t), \mathcal{H}\} \equiv i\mathcal{L}A(t),$$

where $\{,\}$ is a classical Poisson bracket, which can be expressed as follows,

$$\{A, B\} \equiv \sum_i \left( \frac{\partial A}{\partial r_i} \cdot \frac{\partial B}{\partial p_i} - \frac{\partial A}{\partial p_i} \cdot \frac{\partial B}{\partial r_i} \right).$$

Also, for liquids of interest, $\mathcal{H}$ is a classical Hamiltonian with pairwise interactions $\phi(r)$ between the particles,

$$\mathcal{H} = \sum_i \frac{p_i^2}{2m} + \frac{1}{2} \sum_{i,j \neq i} \phi(r_{ij}).$$

We can thus identify the following,

$$i\mathcal{L} = \frac{1}{m} \sum_i \left( p_i \cdot \frac{\partial}{\partial r_i} \right) - \sum_{i,j \neq i} \left( \frac{\partial \phi(r_{ij})}{\partial r_i} \right) \frac{\partial}{\partial p_i}.$$  

It would be possible to integrate the differential equation to find $A(t) = e^{i\mathcal{L}t} A(0)$, but this is not useful by itself.

We also need to define a scalar product of the variables as

$$(A, B) \equiv \langle A^* B \rangle.$$

Now, consider an operator called a projection operator $\mathcal{P}$,

$$\mathcal{P} \equiv (A, \ldots, A)^{-1} A.$$  

If $A$ is a vector, $(A, A)^{-1}$ is thus the inverse of a matrix. Note also that $\mathcal{P}^2 A = \mathcal{P} A = A$. In geometrical terms, the projection operator finds the component of some variable $B$ along the chosen direction $A$, as depicted in Fig. 4.

This is useful as we can extract from an arbitrary $B$ how much “character” of $A$ it has. In particular, the operator $A$ may be a slowly varying (quasi-hydrodynamic) variable. Consider the density as defined in Eq. 4,

$$\rho_k(t) = \sum_i e^{ik \cdot r_i(t)},$$

and then

$$\dot{\rho}_k(t) = i k \cdot \sum_i \frac{p_i(t)}{m} e^{ik \cdot r_i(t)} = i k \cdot j_k(t) = i |k| j_k^L(t),$$

where $j_k^L(t)$ is the longitudinal current. If $k$ is small (large length scales), then $\dot{\rho}_k(t)$ is approximately small. This is what is meant by slow.
Figure 4: A two-dimensional version of the projection operator $\mathcal{P}B$. The quantity $B$ is projected unto the space $A$, which extracts the $A$ component of $B$ (indicated by a thick dashed line).

limit $k = 0$, then $\rho_{k=0} = 0$, and the density is strictly conserved. As Fig. [1] indicates, if $k$ is small, the area of density fluctuations if large, i.e. the rate at which the number of particles fluctuates is small.

We now want to find the exact equation of motion for a correlation function

$$\frac{dA(t)}{dt} = e^{i\mathcal{L}_t \mathcal{P}} A(t) + e^{i\mathcal{L}_t (1 - \mathcal{P})} i\mathcal{L}_A,$$

(18)

where

$$i\Omega = (A, i\mathcal{L}_A) \cdot (A, A)^{-1}.$$  

(19)

Now, writing

$$e^{i\mathcal{L}_t} = e^{i\mathcal{L}_t \mathcal{O}(t)} + e^{i\mathcal{(1-P)}\mathcal{L}_t},$$

(20)

$\mathcal{O}(t)$ can be obtained by differentiating both sides of the equation

$$i\mathcal{L} e^{i\mathcal{L}_t} = i\mathcal{L} e^{i\mathcal{L}_t \mathcal{O}(t)} + e^{i\mathcal{L}_t \hat{\mathcal{O}}(t)} + i [(1 - \mathcal{P}) \mathcal{L}] e^{i\mathcal{(1-P)}\mathcal{L}_t},$$

$$i\mathcal{L} \left( e^{i\mathcal{L}_t \mathcal{O}(t)} + e^{i\mathcal{(1-P)}\mathcal{L}_t} \right) = i\mathcal{L} e^{i\mathcal{L}_t \mathcal{O}(t)} + e^{i\mathcal{L}_t \hat{\mathcal{O}}(t)} + i [(1 - \mathcal{P}) \mathcal{L}] e^{i\mathcal{(1-P)}\mathcal{L}_t},$$

$$i\mathcal{L} \mathcal{O}(t) + e^{i\mathcal{(1-P)}\mathcal{L}_t} = e^{i\mathcal{L}_t \hat{\mathcal{O}}(t)} + i [(1 - \mathcal{P}) \mathcal{L}] e^{i\mathcal{(1-P)}\mathcal{L}_t},$$

$$e^{i\mathcal{L}_t \hat{\mathcal{O}}(t)} = i\mathcal{P} \mathcal{L} e^{i\mathcal{(1-P)}\mathcal{L}_t},$$

$$\mathcal{O}(t) = i \int_0^t d\tau e^{i\mathcal{L}_\tau} \mathcal{P} \mathcal{L} e^{i\mathcal{(1-P)}\mathcal{L}_\tau},$$

(21)

where the last equality follows from the fact $\mathcal{O}(0) = 0$. As a result, we may write

$$e^{i\mathcal{L}_t \mathcal{P}} = \int_0^t d\tau e^{i\mathcal{L}_\tau (1 - \mathcal{P})} i\mathcal{P} \mathcal{L} f(\tau) + f(t),$$

(22)

where $f(t)$ is called the fluctuating force.

$$f(t) \equiv e^{i\mathcal{(1-P)}\mathcal{L}_t} \mathcal{P} \mathcal{L} \mathcal{A}.$$  

(23)
What does this mean? The fluctuating force is obtained by taking the
time derivative of $\mathbf{A}$, using the complimentary projection operator $(1 - P)$
to remove the “$A$” character – perhaps the slow character – and is then
propagated in the orthogonal – fast – space. To put it another way, if $P$
removes the slow character from a variable, then the fluctuating force is
the remaining fast force. We will come back to this later.

It can be shown that $(\mathbf{A}, f(t)) = 0$, by noticing that the definition of
$f(t)$ contains the $(1 - P)$ factor. This means that $f(t)$ is orthogonal to $\mathbf{A}$,
in accord with the discussion above. Noting that
\[
i(\mathbf{A}, \mathbf{L} f(t)) = i(\mathbf{L} \mathbf{A}, f(t)) = i((1 - P)\mathbf{A}, f(t)) = -(f(0), f(t)),
\]
the first term on the RHS of Eq. 22 allows the equation of motion to be
rewritten as
\[
\frac{dA(t)}{dt} = \mathbf{\Omega} \cdot \mathbf{A}(t) - \int_0^t d\tau K(\tau) \cdot \mathbf{A}(t - \tau) + f(t),
\]
where we define the memory function $K(t)$
\[
K(t) \equiv (f, f(t)) \cdot (\mathbf{A}, \mathbf{A})^{-1}.
\]
This is a fundamental and exact equation for the time dependence of $\mathbf{A}(t)$.

Defining the correlation matrix
\[
\mathbf{C}(t) \equiv \langle \mathbf{A}^\ast(0) \mathbf{A}(t) \rangle = (\mathbf{A}, \mathbf{A}(t))
\]
and using the equality $(\mathbf{A}, f(t)) = 0$, we get
\[
\frac{d\mathbf{C}(t)}{dt} = \mathbf{\Omega} \cdot \mathbf{C}(t) - \int_0^t d\tau K(\tau) \cdot \mathbf{C}(t - \tau)
\]
as an exact equation for the matrix of correlation functions $\mathbf{C}(t)$ that we
will want to compute. The problem with computing $\mathbf{C}(t)$ is embodied in
the difficulty of determining $K(t)$.

Now, we want to focus this general framework on density fluctuations
with respect to the bulk density $\rho$, which will allow us to get an expression
for the intermediate scattering function. Consider
\[
\mathbf{A} = \begin{bmatrix} \delta \rho q \\ j^L_q \end{bmatrix},
\]
where
\[
\delta \rho_q = \sum_i e^{iq \cdot r_i} - (2\pi)^3 \rho \delta(q),
\]
\[
j^L_q = \frac{1}{m} \sum_i (\hat{q} \cdot \mathbf{p}_i) e^{iq \cdot r_i},
\]
and therefore
\[
\mathbf{C}(t) = \begin{bmatrix} \langle \mathbf{A}^\ast(0) \mathbf{A}(t) \rangle \\ \langle \delta \rho_q \delta \rho_q(t) \rangle \\ \langle j^L_q \delta \rho_q(t) \rangle \\ \langle j^L_q j^L_q(t) \rangle \end{bmatrix}.
\]
For the purpose of this demonstration, we will concentrate on the element in the lower left corner of the matrix, which is in this case \( \frac{N q d^2 F(q, t)}{m^2} \). At \( t=0 \), the matrix reduces to

\[
C(0) = \begin{bmatrix}
NS(q) & 0 \\
0 & \frac{N k_B T}{m}
\end{bmatrix}.
\]

(32)

Also,

\[
i\Omega = \begin{bmatrix}
\langle A^* A \rangle & \langle A^* A \rangle \\
\langle A^* A \rangle & \langle A^* A \rangle
\end{bmatrix}^{-1},
\]

\[
= \begin{bmatrix}
\langle \delta\rho_q \rangle & \langle \delta\rho_q \rangle \\
\langle \delta\rho_q \rangle & \langle \delta\rho_q \rangle
\end{bmatrix}^{-1}.
\]

(33)

To obtain the last two equations, we used integration by parts, the property that the correlation of an observable and its derivative is always zero, and the statistical thermodynamics result that follows:

\[
\langle j^L_q \rangle = \frac{i q}{m} \sum_{i,j} \langle \hat{q} \cdot \hat{p}_i \rangle e^{-i q \cdot \hat{r}_j} e^{i q \cdot \hat{r}_j}.
\]

(34)

The random force \( f(0) \) is expressed as

\[
f(0) = (1 - P) \hat{A}.
\]

(35)

We will now look at the equation of motion term by term. First,

\[
\frac{dC(t)}{dt} = \begin{bmatrix}
\frac{d}{dt} \langle \delta\rho_q \rangle & \frac{d}{dt} \langle \delta\rho_q \rangle \\
\frac{d}{dt} \langle \delta\rho_q \rangle & \frac{d}{dt} \langle \delta\rho_q \rangle
\end{bmatrix}.
\]

(36)

Note that the lower left corner term equals \( \frac{N q d^2 F(q, t)}{m^2} \). Second,

\[
i\Omega \cdot C(t) = \begin{bmatrix}
\langle \delta\rho_q \rangle & \langle \delta\rho_q \rangle \\
\langle \delta\rho_q \rangle & \langle \delta\rho_q \rangle
\end{bmatrix}.
\]

(37)

Notice the lower left corner term is \( -\frac{N k_B T}{mNS(q)} \) \( F(q, t) \) this time. Lastly, the
memory matrix is

\[
K(t) = \left[ \begin{array}{cc} 0 & R_q(t) \\ \frac{\delta^2}{\delta \phi^2} & \frac{1}{m N k_B T} \end{array} \right],
\]

Concentrating on the lower left corner, using the equation of motion from Eq. 28, we find

\[
\frac{d^2 F(q, t)}{dt^2} + \frac{q^2 k_B T}{m S(q)} F(q, t) + \frac{m}{N k_B T} \int_0^t d\tau \langle R_{-q} R_q(\tau) \rangle \frac{d}{dt} F(q, t - \tau) = 0.
\]

This equation is exact, but impossible to solve. To make approximations, we will look at \(\langle R_{-q} R_q(t) \rangle\) using some intuition. Recall that

\[
R_q = \frac{\partial_q}{\partial q} - \frac{q k_B T}{m S(q)} \delta \rho \delta \phi
\]

and that

\[
\frac{d \partial_q^L}{dt} = \frac{d}{dt} \left\{ \frac{1}{m} \sum_i (\hat{q} \cdot \hat{p}_i) e^{i q \cdot \pi_i} \right\},
\]

Also, note that in this last equation \(\frac{d \pi}{dt}\) is a force and therefore

\[
\frac{d \pi}{dt} \sim - \sum_{i \neq j} \nabla \phi(\|\pi_i - \pi_j\|) = \sum_k i k \delta \rho_k \delta \rho_{-k},
\]

where we made the momentum-space transformation \(\phi_k = \int dr e^{i k \cdot r} \phi(r)\). We then see that hidden in the fluctuating force is a pair of densities. This illustrates an important point: at first, we may have suspected that the fluctuating force is a fast variable and that it decays on a short timescale because we removed the slow modes \(\delta \rho_k\) from it - but we see that it contains, at leading order, a slow character (at least if \(\delta \rho_k\) is slow) from the product of slow modes \(\delta \rho_k \delta \rho_{-k}\). Overall the time derivative of the current has the symmetry of \(\delta \rho_k \delta \rho_{-k}\) with the \(q\) factor comes from \(\sum \delta \rho_k \delta \rho_{-k}\), that multiplies the force in Eq. 41.

We will now approximate \(\langle R_{-q} e^{i Q L} R_q \rangle\). As a convention, note that \(Q \equiv 1 - P\).

1. Replace \(e^{i Q L} \rightarrow \mathcal{P}_2 e^{i Q L} \mathcal{P}_2\), where we define the new projection operator

\[
\mathcal{P}_2 \equiv \sum_{k_1, k_2, k_3, k_4, k_5, k_6} A_{k_1, k_2, k_3, k_4} \langle A_{k_5, k_6}^* \rangle A_{k_1, k_2} A_{k_3, k_4}^{-1}
\]
and where $A_{k_1,k_2} = \delta \rho_{k_1} \delta \rho_{k_2}$. The $\mathcal{P}_2$ operator simply projects $R_q$ onto its dominant slow product mode. We neglect the $Q$ in the exponent, simply because it is hard to compute anything keeping it there. However, to $\mathcal{O}(q^2)$, it may be shown that this neglect is not consequential.

2. Factorize four-point density terms into products of two-point ones.

Using this algorithm, we get

$$\mathcal{P}_2 R_q = \sum_{k_1,k_2} V_q(k_1,k_2) \delta \rho_{k_1} \delta \rho_{k_2},$$

where

$$V_q(k_1,k_2) \equiv \sum_{k_3,k_4} \langle \delta \rho_{k_1} \delta \rho_{k_2} R_q \rangle \cdot \langle \delta \rho_{k_3} \delta \rho_{k_4} \rangle^{-1}.$$  \hspace{1cm} (45)

The denominator has a product of four density variables that will be factorized into products of two structure factors. The numerator has terms like:

$$- \langle \delta \dot{\rho}_{-\vec{k}} \delta \rho_{k-q} \rangle = -\langle \delta \dot{\rho}_{-\vec{k}} \delta \rho_{k-q} \rangle^L - \langle \delta \rho_{-\vec{k}} \delta \dot{\rho}_{k-q} \rangle^L,$$  \hspace{1cm} (46)

where the result was obtained by integration by parts, and

$$-\frac{iqkB_T}{mS(q)} (\delta \rho_{-\vec{k}} \delta \rho_{k-q}).$$

Let’s calculate one of the terms in Eq. 46

$$\langle \delta \dot{\rho}_{-\vec{k}} \delta \rho_{k-q} \rangle^L = i \left\langle \sum_i (k \cdot p_i) e^{-i(k \cdot r_i)} \sum_j e^{i(q \cdot r_j)} \sum_l \langle \hat{q} \cdot p_l \rangle e^{i(q \cdot r_l)} \right\rangle,$$

$$= i \sum_{ij} \langle e^{i(k-q) \cdot r_j} e^{i(q-k) \cdot r_i} \rangle \frac{k_B T}{m} (k \cdot \hat{q})$$

$$= i (k \cdot \hat{q}) \frac{k_B T}{m} N S(|k-q|),$$

where we used the result of Eq. 34 to complete the calculation. The other term similarly gives

$$- \langle \delta \rho_{-\vec{k}} \delta \dot{\rho}_{k-q} \rangle = i (\hat{q} \cdot (q-k)) \frac{k_B T}{m} N S(k).$$

The term in Eq. 47 is hard to compute directly, but within the convolution approximation, it can be reduced as follows [1, 5]:

$$\langle \delta \rho_{-\vec{k}} \delta \rho_{k-q} \rangle \approx N S(k) S(q) S(|k-q|).$$

After treating all static density fluctuations within the Gaussian (and convolution) approximations, we find that the vertex $V_q(k_1,k_2)$ can be expressed as a function of only two wavevectors. As a consequence of translational invariance, we would be left only with terms involving the difference of wavevectors $k \equiv k_1 - k_2$, which allows us to write $V_q(k_1,k_2) = V_{k,q-k}$.
Also note that the summation is now only over $k$. Combining all terms gives
\[
V_{k,q-k} = \frac{ik_BT}{2mN} \left\{ \frac{\langle \hat{q} \cdot k \rangle}{S(k)} + \hat{q} \cdot (q - k) - (q \cdot \hat{q}) \right\},
\]
\[
= \frac{ipk_BT}{2mN} \{ (\hat{q} \cdot k)c(k) + \hat{q} \cdot (q - k)c(|k - q|) \},
\] (51)
where we have rewritten the result using the direct correlation function $c(k) \equiv \frac{1}{2} \left( 1 - \frac{1}{S(k)} \right)$. So, piecing this together,
\[
\langle (R_qP_2)(P_2R_q(t)) \rangle \approx \sum_{k,k'} |V_{k,q-k}V_{k',q-k'}| \langle \delta \rho_{-k'} \delta \rho'_{-q} \delta \rho_k(t) \delta \rho_{q-k}(t) \rangle,
\]
\[
\approx \sum_{k,k'} |V_{k,q-k}V_{k',q-k'}| N^2 F(k,t)F(|q - k|,t)(\delta_{k,k'} + \delta_{k'-q,k}),
\]
\[
= \frac{\rho^2(k_BT)^2}{2m^2} \sum_k |\tilde{V}_{q-k,k}|^2 F(k,t)F(|q - k|,t),
\] (52)
where we used Wick’s factorization and where
\[
\tilde{V}_{q-k,k} \equiv \{ (\hat{q} \cdot k)c(k) + \hat{q} \cdot (q - k)c(|q - k|) \}.
\] (53)
We need to convert the discrete sum to the continuous integral, $\sum_k \to \frac{V}{(2\pi)^3} \int dk$, and multiply by the $\frac{m}{Nk_BT}$ prefactor as obtained in Eq. 39 to get the final MCT equation.
\[
0 = \frac{d^2F(q,t)}{dt^2} + \frac{q^2k_BT}{mS(q)} F(q,t) + \int_0^t d\tau K(q,t-\tau) \frac{\partial F(q,\tau)}{\partial \tau} (54)
\]
with
\[
K(q,t) = \frac{\rho k_BT}{16\pi^3m} \int dk |\tilde{V}_{q-k,k}|^2 F(k,t)F(|k - q|,t)
\] (55)

2.2 Some Properties of the Solution(s) of the MCT Equation

2.2.1 Schematic MCT

Via the approximation discussed by Bengtzelius et al. we can reduce our MCT equation to a schematic form,
\[
\frac{\partial^2 \Phi(t)}{\partial t^2} + \Omega_0^2 \Phi(t) + \lambda \int_0^t d\tau \Phi^2(t - \tau) \frac{\partial \Phi(\tau)}{\partial \tau} = 0,
\] (56)
where $\Phi(t) \sim F(k,t)$, before we neglect the coupling wavevectors. The solutions of Eq. 56 have been discussed by Leutheusser [7] and Bengtzelius et al. [9]. The most striking feature of this equation, and of the full MCT equation from which we “derived” it, is that there is a transition to a completely non-ergodic phase for particular $\Omega_0^2$ and $\lambda$ (or $T$ and $\rho$ for the real MCT equation). The two cases are depicted in Fig. 5.

The transition from ergodic to non-ergodic at a sharp, well-defined set of parameters may be interpreted as the transition from a liquid to a solid.
Figure 5: Left: $\Phi(t)$ decay in the ergodic (supercooled) case. The correlation vanishes on a finite timescale. Right: in the non-ergodic (glassy) case, that same function remains finite even for infinite times.

The fact that correlations do not decay as $t \to \infty$ is indicative of this. However, no information of an ordered state was used or imposed. Thus, the solid could only be a disordered one, i.e. a glass.

### 2.2.2 Solutions of Full MCT Equations

If we denote by $T_c$ the temperature where MCT predicts a glass transition, the relaxation times $\tau$ scales as \[ \tau(q, T) \sim A_q(T - T_c)^{-\gamma}, \] (57)

which means that when $T \to T_c$, $\tau$ diverges as a power law with a universal exponent $\gamma$. This form may fit data, but only over a limited temperature range.

Also, the decay in the $\beta$-relaxation regime is indeed given by \[ f + At^{-a}, \] \[ f - Bt^b, \]

with

\[ \frac{\Gamma(1 - a)^2}{\Gamma(1 - 2a)} = \frac{\Gamma(1 + b)^2}{\Gamma(1 + 2b)}, \] (58)

at least for $T$ very close to $T_c$, i.e. when $\frac{T - T_c}{T} \ll 1$. This is a great triumph of the MCT equations and is fully consistent with simulations and experiments [5, 6]. Furthermore, the power $\gamma$ is related to the exponents $a$ and $b$ as

\[ \gamma = \frac{1}{2a} + \frac{1}{2b}. \] (59)

For the $\alpha$-relaxation regime, an approximate solution of the full MCT equations is indeed approximately given by

\[ F(k, t) \sim e^{-\frac{\beta}{\tau_k^*}} \tau_k^{ak}, \] (60)
and this stretched exponential function describes well experiments and simulations. The schematic equation, however, only exhibits exponential decay.

More generally, MCT predicts that for a correlator at temperature $T$ a time-temperature superposition holds $[5, 6]$:

$$C(t, T) = \hat{C}(t/\tau(T)),$$  

(61)

where $C(t, T)$ is a correlation function, $\hat{C}$ is some master function and $\tau(T)$ is the $\alpha$-relaxation time. This is also generally consistent with experiments and simulations.

2.2.3 Redux: An Assessment of the Successes and Failures of MCT

Even a scientist who is opposed to the spirit and approximations that go into MCT ought to be impressed by its success, where it succeeds. Furthermore, it is essentially the only first-principle theory of glassy liquids. Namely, from the structure of the liquid alone ($S(k)$, the structure factor) a detailed set of dynamical predictions emerge. We will now spell out where MCT works and where (we think) it does not.

Successes

1. MCT makes some remarkable predictions that are correct. For example, the remarkable scaling properties in the $\beta$-relaxation regime that are predicted are essentially correct and so is the time-temperature superposition in the $\alpha$-relaxation regime. This is similarly accurate for other predictions that we will not discuss here $[5, 6]$.

2. MCT has predicted novel relaxation patterns correctly. One recent striking example is the behavior of colloidal suspensions with induced short-ranged attractions$[8]$. Here, MCT has predicted that adding attractions may melt the glass from hard-spheres, and that for certain parameters, logarithmic relaxation may be observed. Both predictions have been confirmed again by computer simulations and experiments.

3. Although we will not discuss it here, there exist models with quenched disorder (spin-glass models) for which the schematic MCT is exact $[10, 11]$. These models make connections between MCT and energy landscape theories possible, as well as extensions of the MCT approach to situations that are out-of-equilibrium (aging). This has been very fruitful and has led to new insights into glassy systems.

Failures

1. The best-known failure of MCT is that it predicts a sharp glass transition at a temperature $T_c$, but $T_c > T_g$. This means that MCT predicts kinetic arrest to a non-ergodic phase at temperatures where the system is still ergodic and liquid.
Figure 6: The non-Gaussian parameter as depicted here is one of the features that MCT cannot reproduce accurately. The curves increase in magnitude and spread with decreasing temperature (from left to right).

2. MCT also predicts power-law divergence of transport coefficients and relaxation times as in Eq. [57], but this is less accurate over a wide range of temperatures than the temperature dependence of transport coefficients given in Eq. [9]. It is a reasonable fitting form over several decades of relaxation time in mildly supercooled liquids. In addition, the parameters $\beta$, $a$, $b$, ... are predicted to be constant in MCT, at least for temperatures for $T - T_c \ll 1$. But in actuality, they are mildly temperature dependent. One should be careful, however, not to take asymptotic predictions of MCT and apply them to cases where $T - T_c$ is not small.

3. Another failure of MCT is in the prediction of certain indicators of collective relaxation. In general, timescales and lengthscales of such heterogeneous motion can be probed by multi-point correlations. A simple, non-multi-point function that seems to correlate crudely with the timescale of such motion is the non-Gaussian parameter

$$\alpha_2(t) = \frac{3\langle r^4(t) \rangle}{5\langle r_2(t) \rangle^2} - 1,$$

where

$$\langle r^2(t) \rangle = \langle [r(t) - r(0)]^2 \rangle$$

for a tagged particle and similarly for the other term. Usually, the behavior of $\langle \alpha^2(t) \rangle$ is similar to what is depicted in Fig. 6. However, MCT predictions of $\alpha_2(t)$ are quite inaccurate. In general, MCT fails to accurately depict properties such as the non-Gaussian parameter and the breakdown of the Stokes-Einstein relation.

One may take from this last result that MCT is not capable of saying anything about dynamically heterogeneous motion in supercooled liquids, but perhaps this statement is too strong. We will explore this further in the next section.
2.3 Field-Theoretic Description

Before finishing this section, we provide a sketch of the field-theoretic approach to schematic MCT [10, 11]. In some sense, the memory function approach can be thought of as arising from coupled Langevin equations for the modes $\delta \rho_q$ and $j_q^L$. For example,

\[
\dot{\delta \rho_q} = i q j_q^L(t), \tag{64}
\]

\[
\frac{\partial}{\partial t} j_q^L(t) = -i q k_B T \frac{S(q)}{m} \dot{\delta \rho_q}(t) - k_B T \frac{1}{m} \int dk (i \delta_q - k) c(k) \delta \rho_{q-k}(t) \delta \rho_k(t) \text{ from the fluctuating force}
\]

\[
- \eta_0 \frac{\dot{j}_q^L(t)}{m} + \eta_q(t) \text{ viscosity term noise term}. \tag{65}
\]

As a toy model for this (forgetting vector labels and wavevectors), we get

\[
\dot{\phi}(t) = -\mu(t)\phi(t) - \frac{g}{2}\phi(t)^2 + \eta(t) \tag{66}
\]

with $\langle \eta(t)\eta(0) \rangle = 2T \delta(t)$. We also define

\[
G_0(t, t') \equiv e^{-\int_t^{t'} dt' \mu(t')}
\]

and

\[
(G \otimes f)(t) \equiv \int_0^t dt' G_0(t, t') f(t'), \tag{68}
\]

where $G_0$ is the bare response function, or propagator. It must be zero if $t < t'$. The solution for $\phi(t)$, with $\phi(0) = 0$ is, in graphical terms,

\[
\phi(t) = \left( G_0 \otimes \eta \right) - \frac{g}{2} \left( G_0 \otimes \eta \right) \left( G_0 \otimes \eta \otimes G_0 \otimes \eta \right) + \ldots. \tag{69}
\]

where an arrow represents $G_0$, an $\times$ represents the noise, and a factor of $g/2$ is associated with each branching point. These terms are simply obtained as a solution from integrating Eq. 66. In a more compact notation,

\[
\phi(t) = G_0 \otimes \eta - \frac{g}{2} G_0 \otimes \left( G_0 \otimes \eta \cdot G_0 \otimes \eta \right) + \ldots. \tag{70}
\]

where "\otimes" is a simple product.

We define two kinds of functions,

\[
C(t, t') \equiv \langle \phi(t)\phi(t') \rangle\tag{71}
\]

and

\[
G(t, t') \equiv \langle \frac{\partial \phi(t)}{\partial \eta(t')} \rangle = \frac{1}{2T} \langle \phi(t)\eta(t') \rangle, \tag{72}
\]

where the last equality is true for Gaussian noise. Again, $C(t, t')$ can be defined on the entire $(t, t')$ plane but $G(t, t')$ only for $t > t'$. Let’s
construct a series for the two functions. The zeroth-order contribution to $C(t,t')$ is given as follows:

$$
\langle \rangle = \left( \int_0^t dt_1 G_0(t,t_1) f(t_1) \int_0^{t'} dt_2 G_0(t_2, t_2) f(t_2) \right),
$$

$$
= \int_0^t \int_0^{t'} dt_1 dt_2 G_0(t, t_1) G_0(t_2, t_2) 2T \delta(t_1 - t_2).
$$

(73)

The bracket average implies connecting the $\times$-vertices to form diagrams with none of these left. Diagrams that do not pair up all noise vertices (i.e., those with an odd number of $\times$-vertices) average to zero.

Going beyond zeroth-order, we get the following:

$$
C(t,t') = + \langle \rangle + \ldots = + \ldots,
$$

$$
≡ \int_0^t dt_1 \int_0^{t'} dt_2 G_0(t, t_1) D(t_1, t_2) G_0(t_2, t_2),
$$

(74)

where to obtain the last line we defined

$$
D(t_1, t_2) \equiv 2T \delta(t_1 - t_2) + \frac{g^2}{2} C_0(t_1, t_2) C_0(t_1, t_2) + \ldots
$$

(75)

What about $G(t,t')$?

$$
G(t,t') = + \left( \frac{1}{2T} \right) + \ldots,
$$

(76)

where the lone $\times$ has to be attached to a $\times$ on the tree and then the remaining diagram is closed. Thus,

$$
G(t,t') = + \ldots,
$$

$$
= G_0(t,t') + \int_{t'}^t dt_1 \int_{t'}^{t_1} dt_2 G_0(t_1, t_1) \Sigma(t_1, t_2) G_0(t_2, t'),
$$

(77)

where we similarly defined

$$
\Sigma(t_1, t_2) \equiv g^2 C_0(t_1, t_2) G_0(t_1, t_2) + \ldots
$$

(78)

This is an exact, formal representation of the perturbations series. In fact, in some sense it is simply a definition of the kernels $D$ and $\Sigma$. We can appeal to the structure of the perturbation series to justify this.

Also, the lower limit of the second integration insures that $t_2 > t'$. In fact, let’s take a closer at the limits of integration for a sample diagram, the second term in Eq. (77) which is reproduced on the left-hand side of Fig. 4 with additional labels. The incoming branch imposes $t > t_1$, the central loop $t_1 > t_2$, and the outgoing branch $t_2 > t'$, for an overall $t > t_1 > t_2 > t'$. The resulting integration limits are thus $\int_{t'}^t dt_1 \int_t^{t_1} dt_2$.

If you have followed so far, you might be asking yourself what happened to the diagrams like the one appearing on the right in Fig. 4. Such closed loops are called tadpoles. They do not contribute to the time dependence of $C$ or $G$, and we assume that their contribution is absorbed into $\mu(t)$. 16
Figure 7: Left: As an exercise, work out the integration limits for this sample diagram. The time pairings indicate the beginning and end times of a given segment. There are in this diagram two internal vertices, \( t_1 \) and \( t_2 \), and two external ones, \( t \) and \( t' \). Right: a simple tadpole diagram.

The next lowest-order terms are then the diagrammatic forms we last drew in Eq. [74] and [77].

To make a self-consistent approximation we replace \( G_0 \) and \( C_0 \) in our second-order approximations for \( D \) and \( \Sigma \) by \( G \) and \( C \). We will call this the Mode-Coupling Approximation, a name that will be clear in meaning at the end \[10, 11\]. This is Eq. [74] and [77] with

\[
\Sigma(t_1, t_2) = g^2 C(t_1, t_2) G(t_1, t_2), \tag{79}
\]

\[
D(t_1, t_2) = 2T \delta(t_1, t_2) + \frac{g^2}{2} C(t_1, t_2)^2. \tag{80}
\]

These two equations can be further manipulated by noting that

\[
G_0 = \left( \mu(t) + \frac{\partial}{\partial t} \right)^{-1}, \tag{81}
\]

and so we can multiply both sides of Eq. [74] and [77] by \( G^{-1} \).

\[
G^{-1}_0 \otimes G = I + \Sigma \otimes G, \tag{82}
\]

\[
G^{-1}_0 \otimes C = G^{-1}_0 \otimes \left\{ G_0 + G_0 \otimes \Sigma \otimes G \right\} \otimes D \otimes G, \]

\[
= D \otimes G + \Sigma \otimes \left\{ G \otimes D \otimes G \right\}, \tag{83}
\]

\[
= D \otimes G + \Sigma \otimes C,
\]

where \( I \) is the identity operator. In other notation,

\[
\left\{ \frac{\partial}{\partial t} + \mu(t) \right\} G(t, t') = \delta(t - t') + \int_{t'}^{t} dt'' \Sigma(t, t'\prime') G(t', t'\prime'), \tag{84}
\]

\[
\left\{ \frac{\partial}{\partial t} + \mu(t) \right\} C(t, t') = \int_{0}^{t} dt'' D(t, t''') G(t', t''') + \int_{0}^{t} dt''' \Sigma(t, t''') C(t''', t') \tag{85}
\]

This still does not look like the MCT equations derived before. We will manipulate the RHS of the second equation to achieve this. Taking the first term, we substitute

\[
\int_{0}^{t} dt'' D(t, t''') G(t', t''') \rightarrow \int_{0}^{t'} D(t, t''') \left. \frac{1}{T} \frac{\partial}{\partial t'''} C(t', t''') \right|_{t'''}^{t''} \tag{86}
\]

We can replace \( t \rightarrow t' \) because of the restriction on time arguments in \( G(t', t''') \) and we can substitute \( G \) for \( C \) as written by using the fluctuation-dissipation theorem (FDT) and assuming that the system is at equilibrium. The result can now be integrated by parts,

\[
\left. \frac{1}{T} \left[ D'(t, t''') C(t'', t') \right] \right|_{0}^{t'} - \frac{1}{T} \int_{0}^{t'} dt''' \frac{\partial}{\partial t'''} D'(t, t''') C(t'', t'). \tag{87}
\]
Note that we can neglect the $\delta$-function part of $D$ here, since it provides no contribution. So, we denote the regular part of $D$ as $D'$. Again, using the FDT, we can substitute
\[
\frac{\partial}{\partial t'} D'(t, t'') \rightarrow T\Sigma(t, t''),
\]
yielding an integral which can be combined with the other term of Eq. 83 to obtain
\[
- \int_0^t dt'' \Sigma(t, t'') C(t'', t') + \int_0^t dt'' \Sigma(t, t'') C(t'', t') = \int_0^t dt'' \Sigma(t, t'') C(t'', t').
\]
Now, using the FDT in the reverse direction and integrating by parts, we get
\[
\left. \frac{1}{T} D'(t, t'') C(t', t'') \right|_t = \frac{1}{T} \int_0^t dt'' D'(t, t'') \frac{\partial}{\partial t'} C(t'', t').
\]
Combining all terms on the right-hand side of Eq. 85, we get
\[
\left. \frac{1}{T} \left[ D'(t, t'') C(t', t'') \right] \right|_t - \frac{1}{T} \int_0^t dt'' D'(t, t'') \frac{\partial}{\partial t'} C(t'', t').
\]
Using the fact that, in equilibrium, these functions are time-translation invariant,
\[
D'(t, t'') = D'(t - t''),
C(t, t') = C(t - t'),
C(t'', t') = C(t'' - t'),
C(0, t') = C(t'),
D'(t, 0) = D'(t),
D'(t, t) = D'(0),
\]
and making the transformation,
\[
t'' - t' \equiv \tau',
\]
\[
t - t' \equiv \tau,
\]
\[
t' \rightarrow \infty,
\]
we find
\[
\left( \frac{\partial}{\partial t} + \tilde{\mu}(t) \right) C(t) + \frac{1}{T} \int_0^t d\tau D'(t - \tau) \frac{\partial C(\tau)}{\partial \tau} = 0,
\]
where
\[
\tilde{\mu}(t) = \mu(t) - \frac{1}{T} D'(0),
\]
\[
D'(t) = \frac{g^2}{2} C(t - \tau)^2.
\]
This is just the schematic model with
\[
\Omega_0^2 \leftrightarrow \tilde{\mu}(t),
\]
\[
\lambda \leftrightarrow \frac{g^2}{2T}.
\]
The only difference is that $\frac{\partial C(t)}{\partial t}$ appears instead of $\frac{\partial^2 \phi(t)}{\partial t^2}$. This actually makes no difference as far as the glassy properties are concerned. In fact, for models of overdamped systems, such as Brownian colloidal spheres, $\frac{\partial \phi(t)}{\partial t}$ is what appears naturally in the reduction of the full MCT equations of the schematic model. This completes the relationship between the memory function/projection operator MCT derivation and a field-theoretic approach.

3 Looking Ahead: Beyond “Simple” MCT

In this section we will outline some thoughts on attempts to do better than the MCT derived thus far. This is not an exhaustive discussion, but is meant to give an idea about what can be done.

3.1 Coupling to Currents

Götze and Sjögren [12] as well as Das and Mazenko [13] have developed theories that remove the sharp transition at $T = T_0$. In both cases, it is the coupling to certain current modes that are ignored in the expressions we have just derived that restore ergodicity below $T_c$.

In the theory of Götze and Sjögren, the Laplace transform of our exact equation of motion for $F(k, t)$ is given as

$$F(k, z) = \frac{-1}{z - \frac{k^2}{\omega(k)^2}} \Omega(k),$$

where $\omega(k) = \frac{k^2}{mS(k)}$ and $M(k, z)$ is the Laplace transform of the memory function

$$M(k, z) = \int_0^\infty dt e^{-zt} M(k, t),$$

and similarly,

$$F(k, z) = \int_0^\infty dt e^{-zt} F(k, t).$$

Essentially, within the “extended” MCT of Götze and Sjögren,

$$M(k, z) \sim \frac{K(k, z)}{1 - \delta h(k, z) K(k, z)},$$

where $K(k, z)$ is the ordinary MCT memory function given in Eq. 39. The expression for $\delta h(k, z)$ is complicated, but to see what it does, note that:

1. if $\delta h = 0$, we recover exactly the ordinary MCT that we have derived before. This can be checked by applying a Laplace transform to the old expressions.

2. if $\delta h(k, z)$ has no singularities as $z \to 0$, the strict transition at $T_c$ in the MCT we have previously derived is removed since relaxation is governed by $M \sim 1/|\delta h|$ at long times and not $K(k, z)$, which yields a pole singularity in z-space.
In the theory of Das and Mazenko, a hydrodynamic approach is used. The kinetic energy of the free energy functional in terms of current $j$ and density $\rho$ has the form

$$K.E.[j, \rho] \sim \frac{j^2}{\rho},$$

(103)

This is like the usual $p^2/2m$ kinetic energy. However, the $1/\rho$ part coupled to the current rounds off the strict singularity at $T_c$, like in the Götze and Sjögren theory.

Note that in both theories, we need currents to restore ergodicity. For some systems, like (simulated) colloidal hard spheres undergoing Brownian motions, these currents do not exist! Thus, the Götze and Sjögren and the Das and Mazenko theories cannot be used to improve ordinary MCT there.

### 3.2 New Closures

As mentioned above, the extended MCT of Götze and Sjögren and of Das and Mazenko cannot tell us anything (beyond ordinary MCT) for Brownian hard-sphere systems. An interesting proposal was recently put forward by Szamel [14]. The main idea is not to factorize the memory function expression leading to the ordinary MCT given in Eq. 39, but to write an exact equation of motion for it, and then factorize the memory function for the new equation. Here is a sketch of the idea.

Recall our old approach to MCT,

$$\frac{\partial^2 F(k, t)}{\partial t^2} + \frac{k^2 k_B T}{m S(k)} F(k, t) + \int_0^t d\tau K(k, t - \tau) \frac{\partial F(k, \tau)}{\partial \tau} = 0,$$

(104)

where essentially $K(k, t) \sim \langle \delta \rho \delta \rho \rangle$ is a four-point function of density variables. In the old approach, the closure involved

$$K(k, t) \sim \langle \delta \rho \delta \rho \rangle \frac{\partial F(q, t)}{\partial q} = \sum_q F(q, t) F(|k - q|, t)$$

(105)

and this allowed to solve for $F(k, t)$.

Instead of factorizing the four-point memory kernel, let’s write an exact equation of motion for it, following the same lines of reasoning as before,

$$\frac{\partial^2 K(t)}{\partial t^2} + \Gamma K(t) + \int_0^t d\tau R(t - \tau) \frac{\partial K(t)}{\partial \tau} = 0,$$

(106)

The wavevector indices are suppressed to simplify the notation in order to clarify idea behind the manipulations. This has the same form as before, but with new frequencies $\Gamma$ and a new memory function $R(t - \tau)$.

Schematically, $R \sim \langle \delta \rho \delta \rho \rangle$ is a six-point function! We can close the equation for $K$ and $F$, by approximating $R \approx K \cdot F$ the product of a four-point and a two-point function. This yields two coupled sets of integro-differential equations that may be solved self-consistently yielding a converged $F(k, t)$.

This approach has not been considered for the full dynamics of $F(k, t)$, but yields a better estimate for $T_c$, (i.e. the $T_c$ that is extracted is closer to the measured glass transition).
3.3 Four-point correlations and dynamical heterogeneities

It was mentioned in Section 2 that MCT does not describe well

$$\alpha_2(t) \equiv \frac{3\langle r^4(t) \rangle}{5\langle r^2(t) \rangle^2} - 1$$

(107)

and that $\alpha_2(t)$ seems to correlate well with the timescale of maximal dynamical heterogeneity $^{16,17}$. It turns out that this timescale is in the late $\beta$-regime. This highlights the fact $\alpha_2(t)$ yields information on transiently mobile particles that jump due to the destruction of cages. It should be noted that there is no lengthscale dependence in $\alpha_2(t)$.

To gain some information about a growing (dynamical) lengthscale, a multipoint dynamical generalization of the static structure factor may be studied $^{18,19,20,21}$. The $k \rightarrow 0$ limit of this structure factor, as Sharon Glotzer discusses in her lectures, is the susceptibility

$$\chi_4(t) \sim \int d\mathbf{r}_1 \ldots d\mathbf{r}_4 \theta_a(|\mathbf{r}_1 - \mathbf{r}_2|) \theta_a(|\mathbf{r}_3 - \mathbf{r}_4|) \times$$

$$\langle \rho(\mathbf{r}_1,0) \rho(\mathbf{r}_2,t) \rho(\mathbf{r}_3,0) \rho(\mathbf{r}_4,t) \rangle,$$

(108)

where the function $\theta_a(|\mathbf{r}_1 - \mathbf{r}_2|)$ equals one when $|\mathbf{r}_1 - \mathbf{r}_2| \leq a$, and zero otherwise $^{15}$. The timescale at which $\chi_4(t)$ peaks is generally in the $\alpha$-regime. The growing lengthscale associated with dynamic heterogeneity is associated with slow moving, transiently caged particles.

Given the superficial similarity with $\alpha_2(t)$, as shown in figure 8, one might conclude that MCT cannot compute objects like $\chi_4$, but this is not the case. Recent work by Biroli and Bouchaud $^{19}$, motivated by the earlier insight of Franz and Parisi $^{20}$ and Kirkpatrick and Thirumalai $^{21}$, shows that MCT may make quantitative statements about the scales of length and time associated with dynamical heterogeneity. So far, absolute lengthscales have not been computed, but dynamical exponents $z$ relating timescales $\tau$ and lengthscales $\xi$ have,

$$\tau \sim \xi^z,$$

(109)
where \( z = 2\gamma \) and \( \gamma \) is given in Eq. 59.

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