Power-law Decay and the Ergodic-Nonergodic Transition in Simple Fluids

Abstract It is well known that mode coupling theory (MCT) leads to a two-step power-law time decay in dense simple fluids. We show that much of the mathematical machinery used in the MCT analysis can be taken over to the analysis of the systematic theory developed in the Fundamental Theory of Statistical Particle Dynamics [1]. We show how the power-law exponents can be computed in the second-order approximation where we treat hard-sphere fluids with statics described by the Percus-Yevick solution.

1 Introduction

The long time dynamics of dense classical fluids is complicated. There is experimental evidence of power-law decays as a function of time. One approach used to describe this evolution is Mode-Coupling Theory (MCT). MCT has been an extremely controversial theory. The problem is that there is no systematic derivation. MCT has been cobbled together piece by piece leading to its present form. There has been no systematic way of deriving, correcting, or extending the basic description.

Despite these criticisms MCT has led to a long time scenario which seems to match a lot of the physical operations in a dense fluid. It has been argued by [2] that MCT is the “mean field” theory governing glassy behavior and the basic results are more general than found in MCT.

The most contentious issue in judging MCT is the existence of an ergodic-nonergodic (ENE) transition. It is commonly believed that there are no ENE transitions in the classes of models studied. However there is almost a transition. In the ideal MCT scenario there is an ENE transition at some critical packing fraction $\eta^*$. MCT implies a rather elaborate slow dynamics on either side of the critical density. There is, for $\eta < \eta^*$, a three step process [3].

The equilibrium density autocorrelation function first exhibits power-law decay

$$G_{\rho \rho}(q,t) = \langle \rho_{-q}(0)\rho_{q}(t) \rangle_c \sim f_q + A_q t^{-a},$$

followed by the von Schweidler decay

$$\langle \rho_{-q}(0)\rho_{q}(t) \rangle_c \sim f_q - B_q t^b;$$

and finally exponential (possibly stretched) decay

$$\langle \rho_{-q}(0)\rho_{q}(t) \rangle_c \sim e^{-(t/t_0)^\beta}.$$

A full analysis of the theory gives expressions for the exponents

$$\frac{\Gamma(1-a)^2}{\Gamma(1-2a)} = \lambda = \frac{\Gamma(1+b)^2}{\Gamma(1+2b)},$$

where $\lambda$ is a material dependent quantity.
In FTSPD\cite{1} and SDENE\cite{4} one of us introduced a new theory for the dynamics of collections of classical particles. It is shown in \cite{3} that at second order in perturbation theory one is led to a description which has similarities with MCT.

In this paper we take the analysis of the kinetics beyond the analysis of the location of the transition. We explore the slow kinetics for the regimes below and above the transition. It was shown in \cite{3,5} that the density-density cumulant $G_{pp}(q,t)$ satisfies the kinetic equation

$$\frac{\partial G_{pp}(q,t)}{\partial t} = -Dq^2 \bar{\rho} S(q)^{-1} G_{pp}(q,t) + \bar{D}q^2 \int_0^t ds \tilde{\beta} \Sigma_{BB}(q,t-s) \tilde{G}_{pp}(q,s),$$

(1.1)

where $\beta = 1/k_B T$ is the inverse temperature, $\bar{\rho}$ is the average density, $D$ is the diffusion coefficient, $S(q)$ is the static structure factor, and $\Sigma_{BB}$ is the B-B component of the self-energy matrix for the theory of the two fields $\rho$ and $B$ where $B$ is a response field. The important point is that $\Sigma_{BB}$ can be determined systematically in perturbation theory and, at second order in the potential, is given by

$$\beta^2 \bar{\rho}^4 \Sigma_{BB}(q_1,t) = -\frac{1}{2} \int \frac{dk}{(2\pi)^3} k_3 dt' \delta(q_1-k_2-k_3) \delta(t-t') \left[ 1 + 2\tilde{K}_{12} \frac{\partial}{\partial t} + 2\tilde{K}_{13} \frac{\partial}{\partial t'} + \tilde{K}_{12}^2 \frac{\partial^2}{\partial t^2} \right] \left[ G_{pp}^{(0)}(k_2,t) G_{pp}(k_3,t') + G_{pp}(k_2,t') G_{pp}(k_3,t) \right],$$

where $\tilde{K}_{12} = (q_1 \cdot k_2)/q_1^2 k_2^2$, $\tilde{K}_{13} = (q_1 \cdot k_3)/q_1^2 k_3^2$, $(G_{pp}^{(0)})$ is the non-interacting density autocorrelation function and $\tilde{G}_{pp}$ and $\tilde{G}_{pp}$ are dressed propagators \cite{5}. Recently it has been suggested \cite{6} that it is important to include information in addition to the static structure factor to make accurate quantitative predictions about glassy dynamics. In principle, a systematic extension of FTSPD to higher orders in the effective potential can take such data into account.

There are a number of differences between the MCT kernel and $\Sigma_{BB}$ although the overall structure is similar. In the FTSPD the self-energy has the basic structure represented by the one-loop diagrams shown in Figure 1. The triangles at the ends of the graph are the lowest order vertices. We study here the cases where the vertex functions are given by the non-interacting values. These vertices have a frequency dependence. Between the vertices we have several sets of products of matrix propogators. An interesting feature is that all of these propagators $G_{ij}^{(0)}$, $G_{ij}$, $\tilde{G}_{ij}$, satisfy similar fluctuation-dissipation theorems. For a full discussion see \cite{7}.

It has been understood for some time that there are corrections to MCT that consist of higher numbers of loops. The two loop correction is of the general form

$$\Sigma_{BB}^{(2)}(1,2) = 1 \quad \text{where now one has 4-point vertices and a product of these dressed matrix propagators.}$$

Our work here is to explore the long time solutions of the kinetic equations. Near an ENE transition we can assume certain analytic forms for the time dependence of $G_{pp}(q,t)$. For MCT a rather elaborate mathematical machinery has been developed for performing this asymptotic analysis \cite{8}. While none of this is new for MCT, we find it useful to present these procedures in some detail. The results of an analysis for MCT are well known; for the case of the FTSPD the results are all new and comparable to the MCT. We begin with some background information on FTSPD and MCT. We follow this by a review of the asymptotic MCT analysis. Finally, we apply the analysis to FTSPD and numerically determine the power-law and von Schweidler exponents for hard spheres using the Percus-Yevick approximation for the static structure factor. We compare these results to those of the corresponding mode coupling theory.

Fig. 1 Generic one loop contribution to $\Sigma_{BB}(1,2)$. 

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2 Reviews of FTSPD and MCT

In this section we shall briefly review the origins of FTSPD and MCT and how one can use them to construct kinetic equations like (1.1) for correlation functions of interest.

FTSPD

The Fundamental Theory of Statistical Particle Dynamics is a framework from which mode-coupling-like kinetic equations can be derived, including systematic corrections to them. In Smoluchowski dynamics, FTSPD is developed as a field theory whose fundamental field is the position variable, \( \mathbf{R}_i(t) \). Starting with the equations of motion,

\[
\dot{\mathbf{R}}_i(t) = \mathbf{D}_i + \mathbf{n}_i(t)
\]

together with the variance of the Gaussian noise term

\[
\langle \mathbf{n}_i(t) \mathbf{n}_j(t') \rangle = 2k_B T \delta_{ij} \delta(t - t')
\]

we obtain a generating functional for a path

\[
\mathcal{Z}(H) = \langle e^{\mathbf{R}_i H_i} \rangle = \int [d \mathbf{n}] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \exp \left\{ -\frac{1}{4k_B T} \int_{t_0}^{\infty} \mathbf{n}_i^2 dt + \mathbf{R}_i \cdot H_i \right\}
\]

where \( \mathbf{R} \cdot H \) is shorthand for

\[
\int_{t_0}^{\infty} \mathbf{R}(t) \cdot H(t) \, dt
\]

and the integrated-over initial conditions are

\[
\mathbf{R}_i(t_0) = \mathbf{R}_i(0)
\]

with probability distribution given by \( P_0(\mathbf{R}_i(0)) \). We can obtain the Martin-Siggia-Rose action for this system by exploiting the functional delta-function identity

\[
\int [d \mathbf{n}_i] \delta(\mathbf{R}_i - \mathbf{DF}_i - \mathbf{n}_i) = \int [d \mathbf{R}_i] \det \mathbf{M}_i \delta(\mathbf{R}_i - \mathbf{DF}_i - \mathbf{n}_i) = 1
\]

where \( \det \mathbf{M}_i \) is the Jacobian

\[
\det \mathbf{M}_i = \det \frac{\delta \mathbf{n}_i}{\delta \mathbf{R}_i}
\]

Inserting this identity into the generating functional and using the functional Fourier representation of the delta-function we obtain

\[
\mathcal{Z}(H) = \int [d \mathbf{n}_i] [d \mathbf{R}_i] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \exp \left\{ -\frac{1}{4k_B T} \int_{t_0}^{\infty} \mathbf{n}_i^2 dt + \mathbf{R}_i \cdot H_i \right\}
\]

\[
\quad = \int [d \mathbf{n}_i] [d \mathbf{R}_i] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \det \mathbf{M}_i \delta(\mathbf{R}_i - \mathbf{DF}_i - \mathbf{n}_i) \exp \left\{ -\frac{1}{4k_B T} \int_{t_0}^{\infty} \mathbf{n}_i^2 dt + \mathbf{R}_i \cdot H_i \right\}
\]

\[
\quad = \int [d \mathbf{n}_i] [d \mathbf{R}_i] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \det \mathbf{M}_i \exp \left\{ i \int \mathbf{R}_i \cdot (\mathbf{DF}_i - \mathbf{n}_i) dt \right\} \exp \left\{ -\frac{1}{4k_B T} \int_{t_0}^{\infty} \mathbf{n}_i^2 dt + \mathbf{R}_i \cdot H_i \right\}
\]

We now combine the arguments of the two exponentials into a single integral:

\[
\mathcal{Z}(H) = \int [d \mathbf{n}_i] [d \mathbf{R}_i] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \det \mathbf{M}_i \exp \left\{ \int_{t_0}^{\infty} \left( i \mathbf{R}_i \cdot (\mathbf{DF}_i - \mathbf{n}_i) - \frac{\mathbf{n}_i^2}{4k_B T} \right) dt + \mathbf{R}_i \cdot H_i \right\}
\]

\[
\quad = \int [d \mathbf{n}_i] [d \mathbf{R}_i] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \det \mathbf{M}_i \exp \left\{ \int_{t_0}^{\infty} \left( i \mathbf{R}_i \cdot (\mathbf{DF}_i - \mathbf{n}_i) \right) dt + \mathbf{R}_i \cdot H_i \right\}
\]

\[
\quad = \int [d \mathbf{n}_i] [d \mathbf{R}_i] [d^3 \mathbf{R}_i] P_0(\mathbf{R}_i(0)) \det \mathbf{M}_i \times \exp \left\{ \int_{t_0}^{\infty} \left( i \mathbf{R}_i \cdot (\mathbf{DF}_i) - \frac{(\mathbf{n}_i^2 + 2ik_B T \mathbf{DF}_i)^2}{4k_B T} - \mathbf{R}_i \cdot \mathbf{D}_i \right) dt + \mathbf{R}_i \cdot H_i \right\}
\]
Now all other variables are independent of $\eta$, so we can perform the Gaussian integral and absorb the constant into the measure. We obtain

$$Z(H) = \int [dR_i][d\tilde{R}_i] d^3 \tilde{R}_i^{(0)} P_0(\tilde{R}_i^{(0)}) \det M_i \exp \left\{ - \int [\tilde{R}_i] k_B T D\tilde{R}_i - i\tilde{R}_i(\tilde{R}_i - DF_i) dt + R_i \cdot H \right\}.$$ 

We use this to build the generating functional for all the paths:

$$Z_N(H) = \int [dR_i][d\tilde{R}_i] d^3 \tilde{R}_i^{(0)} P_0(\tilde{R}_i^{(0)}) \exp \left\{ - \int dt \sum_{i=1}^N [\tilde{R}_i] k_B T D\tilde{R}_i - i\tilde{R}_i(\tilde{R}_i - DF_i) - A'_j + \phi \cdot H \right\},$$

where we have rewritten the Jacobians as $A'_j = \ln \det M_i$ and introduced the symbol $\phi$ to represent the fields (constructed from $R$, $\tilde{R}$) that we wish to couple to $H$. A more complete derivation with a treatment of the Jacobian is available in [4]. From the generating functional of the paths we obtain the grand canonical partition function

$$Z_T[H] = \sum_{N=0}^{\infty} \frac{Z_N(H)}{N!}$$

from which we construct the generator of cumulants

$$W[H] = \log Z_T[H].$$

The source fields $H_\rho$ and $H_B$ are coupled to the density

$$\rho(1) = \sum_i \delta(x_1 - R_i(t_1)),$$

and response field

$$B(1) = D \sum_i \{\tilde{R}_i V_1 + \theta(0) V_1^2\} \delta(x_1 - R_i(t_1)),$$

respectively. The value of $\theta(0)$ is chosen based upon the discretization scheme we adopt for the path integral. The standard Stratonovich discretization results in $\theta(0) = 1/2$. We generate cumulants by functional differentiation:

$$G_{\rho \cdots \rho \cdots B \cdots B}(1, \ldots, \ell, \ell + 1, \ldots, n) = \frac{\delta}{\delta H_\rho} \cdots \frac{\delta}{\delta H_\rho} \frac{\delta}{\delta H_B} \cdots \frac{\delta}{\delta H_B} W[H]\bigg|_{H_\rho = H_B = 0}.$$ 

From the Legendre transform of $W[H]$ one generates the irreducible vertices

$$\Gamma_{ij \cdots k} = \frac{\delta}{\delta G_i} \frac{\delta}{\delta G_j} \cdots \frac{\delta}{\delta G_k} W[G].$$

The two-point vertices satisfy the Dyson equation

$$\sum_k \Gamma_{ik} G_{kj} = \delta_{ij}.$$ 

Expanding the vertex to second order in the potential we have $\Gamma^0_{ik} = y_{ik}^{(0)} + \gamma_{ik}^{(1)} - \Sigma_{ik}$. Plugging into the $B \cdot \rho$ component of Dyson’s equation we construct the kinetic equation

$$\frac{\partial G_{\rho \rho}}{\partial t}(q,t) = -D q^2 \tilde{\rho} S(q)^{-1} G_{\rho \rho}(q,t) + D q^2 \int_0^t ds \beta^2 \tilde{\rho} \Sigma_{BB}(q,t - s) \tilde{G}_{\rho \rho}(q,s),$$

where $\tilde{D} = k_B T D$ and $G_{\rho \rho}(q,0) = S(q)$. It is shown in [4] that approximations for $\Sigma_{BB}$ can be generated as a power series in an effective interaction potential.

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1 Using the Ito prescription is equivalent to making $\theta(0) = 0$, resulting in a trivial Jacobian. The Stratonovich prescription is usually preferable since the ordinary chain rule of calculus remains unmodified when dealing with multiplicative noise. Either way, in FTSPD the Fokker-Planck equation and all correlation functions resulting from the path integral are independent of the choice of discretization since the noise is not multiplicative.
MCT

The most common path to Mode Coupling Theory is through the projection operator formalism of Zwanzig and Mori. We briefly summarize the development given in [9]. We start from the fundamental relationship between a function of phase-space variables $A(r, p)$ and the Hamiltonian $\mathcal{H} = \sum p_i^2 / 2m + \frac{1}{2} \sum_{i \neq j} V(r_{ij})$:

$$\frac{dA}{dt} = \{A, \mathcal{H}\} \equiv i\mathcal{L}A(t), \quad (2.1)$$

where we have used the Poisson bracket

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

This equation defines the Liouville operator $\mathcal{L}$:

$$i\mathcal{L} = \sum_i \frac{p_i}{m} \frac{\partial}{\partial r_i} - \sum_{i \neq j} \frac{\partial V}{\partial r_i} \frac{\partial}{\partial p_i}.$$ 

It follows that we may formally express the time evolution of $A$ as

$$A(t) = e^{i\mathcal{L}t}A(0).$$

The next step in the analysis is to identify $A$ with a set of functions that we are interested in tracking. These are referred to as “slow variables.” For the derivation of the basic MCT equation these are the density fluctuations and longitudinal current: $A(t) = (\delta \rho_0(t), j_x^L(t))$. We also define an inner product

$$(A, B) = \langle A^\dagger B \rangle,$$

which allows us to define the projection operator

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

Note that $A = A(0)$ above. The projections perpendicular to the slow variables are referred to as “fast variables.” Separating the left-hand side of (2.1) into components parallel and perpendicular to $A(0)$ allows us to express the time evolution as

$$\frac{dA}{dt} = i\Omega A(t) + \int_0^t e^{i\mathcal{L}(t-\tau)} i\mathcal{P} \mathcal{L} f(\tau) d\tau + f(t),$$

where

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

and

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

Exploiting the orthogonality between $f(t)$ and $A(0)$ allows us to write

$$\frac{dA}{dt} = i\Omega A - \int_0^t K(t-\tau)A(\tau) d\tau + f(t),$$

where the kernel is given by

$$K(t) = (f(0), f(t))(A(0), A(0))^{-1}.$$ 

Introducing the correlation matrix $C(t) = \langle A^\dagger(0)A(t) \rangle = (A(0), A(t))$ we have

$$\frac{dC}{dt} = i\Omega C(t) - \int_0^t d\tau K(t-\tau)C(\tau).$$

\footnote{The projection operator retains the same form when $A$ is generalized to a set of functions. In this case, $(A, A)$ refers to the matrix $A_{ij} = (A_i, A_j)$ and $\mathcal{P} B = \sum_i (A, A)^{-1}_{ij} (A, B) A_i$.}
Specializing to our choice of slow variables, \( A = (\delta p_0(t), j_k^0(t)) \), we obtain
\[
\frac{\partial^2 G_{pp}(q,t)}{\partial t^2} + \frac{q^2 k_B T}{mS(q)} G_{pp}(q,t) + \frac{m}{N k_B T} \int_0^t d\tau \langle R_{-q}(0) R_q(t-\tau) \rangle G_{pp}(q,\tau) = 0
\] (2.2)
from the \( \rho \cdot j \) component of our correlation matrix, where
\[
G_{pp}(q,t) = \langle \delta p_{-q}(0) \delta p_q(t) \rangle
\]
is the density-density cumulant\(^3\) and
\[
R_q = \frac{d j_q^0}{dt} - i \frac{q k_B T}{mS(q)} \delta p_q.
\]
So far all of these manipulations have been exact. However, computing with (2.2) is not practical without simplifying the kernel \( K(t) = \langle R_{-q}(0) R_q(t) \rangle \). This is the point where the uncontrolled approximations of MCT are introduced. They consist of:

- Replacing the time evolution operator \( e^{(1-\rho) q^2 t} \rightarrow \mathcal{P}_2 e^{|q|^2 t} \mathcal{P}_2 \), where \( \mathcal{P}_2 \) projects onto the dominant “slow” mode of \( R_q \).
- Gaussian and convolution approximations for the correlation functions which allow them to be factorized into simpler correlation functions proportional to the static structure factor.

In contrast to the FTSPD, it is not clear how one could go about systematically improving the MCT approximations for the memory function kernel.

3 Review of MCT Asymptotic Analysis

These approximations define mode coupling theory and lead to the following equation\(^3\) for the density autocorrelation function, \( G_{pp}(q,t) \):
\[
\frac{\partial^2}{\partial t^2} G_{pp}(q,t) + \Omega^2(q) G_{pp}(q,t) + \Omega^2(q) S(q) \int_0^t dt' M(q,t-t') \frac{\partial}{\partial t'} G_{pp}(q,t') = 0,
\]
where
\[
\Omega^2(q) = \frac{q^2 k_B T}{mS(q)},
\]
\[
M(q,t) = \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} V^2(q,k) G_{pp}(k,t) G_{pp}(|q-k|,t)
\]
\[
V^2(q,k) = \frac{\rho c}{q^2} \{\hat{q} \cdot k c(k) + \hat{q} \cdot (q-k) c(|q-k|)\}^2
\]
\[
\rho c(k) = 1 - \frac{1}{S(k)}
\]
The important point is that \( M \) is a non-linear function of the density autocorrelation function. \( V(q,k) \) is the vertex in the theory. We may rewrite the equation in terms of the normalized density autocorrelation function \( \phi(k,t) \) which is related to the autocorrelation function by \( G_{pp}(q,t) = S(q) \phi(q,t) \). The kinetic equation can then be written as
\[
\frac{\partial^2}{\partial t^2} \phi(q,t) + \Omega^2(q) \phi(q,t) + \Omega^2(q) \int_0^t dt' M'(q,t-t') \frac{\partial}{\partial t'} \phi(q,t') = 0.
\] (3.1)

\(^3\) It should be noted that the convention used in this section (and most of the MCT literature) for the cumulants is different from the convention we have employed in the discussion of FTSPD: \( G_{pp}^{\text{MCT}}(q,t) = \lim V^{-1}(\delta p_{-q}(0) \delta p_q(t)) \) while \( G_{pp}^{\text{FTSPD}}(q,t) = \lim N^{-1}(\delta p_{-q}(0) \delta p_q(t)) \), where \( V \) and \( N \) are the volume and number of particles, respectively. Accordingly the static structure factors are related in the same way: \( S^2(q) = p S^{\text{MCT}}(q) \). We continue to use the MCT convention when we discuss the MCT kinetic equation below, and switch back to the original convention when we discuss the FTSPD kinetic equation.

\(^4\) We are presenting the Newtonian MCT equation. For Smoluchowski dynamics only one time derivative appears.
The corresponding kernel $M'(q,t)$ is given by

$$M'(q,t) = \frac{1}{2} \int \frac{d^3 k}{(2\pi)^3} V^2(q,k) \phi(k,t)\phi(|q-k|,t)$$

$$V^2(q,k) = \frac{\rho}{q^2} S(q)S(k)S(|q-k|) \{\hat{q} \cdot k c(k) + \hat{q} \cdot (q-k)c(|q-k|)\}^2 .$$

The only relevant parameter for hard spheres is the number density, $\rho$. We are interested in extracting the long-time slow dynamics for this model. In the following sections all numerical calculations and plots are found using the Percus-Yevick approximation for the hard sphere structure factor. It is useful to work using the Laplace transform which diagonalizes [34] [10, 11]. This leads to

$$\hat{z}^2 \phi(q,z) - z + \Omega^2 \phi(q,z) + \Omega^2 \mathcal{L}[M(q,t)](z\phi(q,z) - 1) = 0 ,$$

where we have applied the initial conditions $\phi(q,0) = 1$ and $\dot{\phi}(q,0) = 0$. Rearranging terms we have

$$\frac{\Omega^2 \phi(q,z)}{1 - z\phi(q,z)} = \mathcal{L}[M(q,t)] + z .$$

(3.2)

3.1 Heuristic Analysis of MCT: Time Dependence at Transition

In the long-time regime we assume $\phi(q,z) \to \infty$ as $|z| \to 0$. In this regime ($|z| \ll 1$) we may ignore the $z$ on the right hand side of (3.2) and thereby cancel the factors of $\Omega$. This gives us

$$\frac{\phi(q,z)}{1 - \phi(q,z)} = \mathcal{L}[M(q,t)] .$$

(3.3)

Next we apply the ansatz $\phi(q,t) = f(q) + (1 - f(q))^2 \psi_q(t)$ where $\psi_q(t)$ is assumed small and $f(q)$ is a non-ergodicity parameter. The Laplace transform of the ansatz is given by

$$\phi(q,z) = \frac{f(q)}{z} + (1 - f(q))^2 \psi_q(z) .$$

We now plug this into (3.3) and expand the left hand side to second order in $\psi_q(z)$:

$$\frac{1}{1 - f_q} \left( \frac{f_q}{z} + (1 - f_q)\psi_q(z) + z(1 - f_q)^2 \psi_q^2(z) \right) = \mathcal{L}[M(q,t)] .$$

To apply the ansatz to the right hand side note that

$$\phi(k,t)\phi(|q-k|,t) = \{f_k + (1 - f_k)^2 \psi_k(t)\}\{f_{k-k} + (1 - f_{k-k})^2 \psi_{k-k}(t)\}$$

$$= f_k f_{k-k} + f_k(1 - f_{k-k})^2 \psi_k(t) + f_{k-k}(1 - f_k)^2 \psi_k(t) + (1 - f_k)^2 (1 - f_{k-k})^2 \psi_k(t) \psi_{k-k}(t) .$$

The two middle terms above may be combined into a single term in the integral. We now employ the notation of [8] to make the following more compact. Let

$$F_q[f_k] = \frac{\rho S(q)}{2q^2} \int \frac{d^3 k}{(2\pi)^3} V^2(q,k) f_k f_{k-k}$$

$$C_q = \rho \frac{\delta F_q[f_k]}{\delta f_k}$$

$$C_{qk} = \frac{\delta F_q[f_k]}{\delta f_k} (1 - f_k)^2$$

$$C_{qkp} = \frac{1}{2} \frac{\delta^2 F_q[f_k]}{\delta f_k \delta f_p} (1 - f_k)^2 (1 - f_p)^2 .$$

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5 Our conventions are defined in Appendix B.

6 Note that in MCT both Newtonian dynamics and Smoluchowski dynamics satisfy the same asymptotic equation.
We obtain
\[
\frac{1}{z} \left( \frac{f_q}{1-f_q} - F_q[f_k] \right) + \left( \psi_q(z) - C_{qk} \psi_k(z) \right) + z(1-f_q) \psi^2_q(z) - C_{qkp} \mathcal{Z}[\psi_q(t) \psi_p(t)] = 0 \quad (3.5)
\]
where it is understood that we are integrating over repeated indices: \(C_{qk} f_k \equiv \int d^3k C_{qk} f_k / (2\pi)^3\), etc. This expansion yields three conditions for the ideal metastable state. The first one, given by
\[
\frac{f_q}{1-f_q} = F_q[f_k]
\]
determines \(f_q\). For hard spheres, this equation can be solved by iteration. A plot of \(f_q\) using the Percus-Yevick approximation for the hard-sphere structure factor is available in Figure 2. The second and third conditions can be found by considering a further expansion of \(\psi_q(z) = \psi^{(1)}_q + \psi^{(2)}_q\), with \(\psi^{(2)}_q / \psi^{(1)}_q \to 0\) as \(z \to 0\). We plug the expansion into our equation and match orders. To the order of \(\psi^{(1)}_q\), we obtain
\[
\psi^{(1)}_q(z) = C_{qk} \psi^{(1)}_k(z),
\]
where \(C_{qk}\) is defined by (3.4). This equation determines the spatial dependence of \(\psi^{(1)}_q(z)\). We can solve this equation for \(\psi^{(1)}_q(z)\) by finding an eigenvector of the operator
\[
C_{qk} = \frac{\rho S(q)}{q^2} V^2(q,k) f_{q-k}(1-f_k)^2
\]
with unit eigenvalue. This eigenvector is found at the critical density \(\eta_{\text{MCT}} = 0.515\). Below the critical density (3.6) cannot support a nontrivial solution. Above the critical density the eigenvalue moduli are all less than one. Denote the right eigenvector associated with the unit eigenvalue by \(e_k\), and the corresponding left eigenvector by \(\hat{e}_k\). Furthermore, impose the conditions \(\sum_k \hat{e}_k e_k = 1\) and \(\sum_k \hat{e}_k e_k e_k (1-f_k) = 1\). With this notation we have
\[
\psi^{(1)}_q = Ae_q \phi_v(z),
\]
Fig. 2 This plot shows \(f^{\text{MCT}}_k\), the non-ergodicity parameter, at the critical density \(\eta_{\text{MCT}} = 0.515\).
with $A$ a thus far undetermined constant and $\phi_v(z)$ a function of $z$ only. Plots of $e_k$ and $\hat{e}_k$ for the hard sphere system are available in Figures 3 and 4, respectively. Next, we match the second order terms in (3.5). We obtain

$$\left( \delta_{qk} - C_{qk} \right) \phi_k^{(2)} = -\left( 1 - f_k \right) z \left[ \psi_q^{(1)} \right]^2 + C_{qkp} \mathscr{L} \left[ \psi_k^{(1)} \psi_p^{(1)} \right].$$

Now multiply both sides by the left eigenvector, $\hat{e}_q$. We obtain

$$0 = -\hat{e}_q \left( 1 - f_q \right) z \left[ \psi_q^{(1)} \right]^2 + \hat{e}_q C_{qkp} \mathscr{L} \left[ \psi_k^{(1)} \psi_p^{(1)} \right]$$

$$= -\hat{e}_q \left( 1 - f_q \right) z A^2 e_q e_q \phi_q^{(2)}(z) + A^2 \hat{e}_q C_{qkp} e_k e_p \mathscr{L} \left[ \phi_k^{(2)}(t) \right]$$

$$= -A^2 \phi_q^{(2)}(z) + A^2 \lambda \mathscr{L} \left[ \phi_q^{(2)}(t) \right],$$

where we have used the condition $\sum_k \hat{e}_k e_k (1 - f_k) = 1$ and $\lambda = \hat{e}_q C_{qkp} e_k e_p$ in the third line. This gives us an equation for the $z$-dependent part of $\psi^{(1)}$:

$$z \phi_q^{(2)}(z) = \lambda \mathscr{L} \left[ \phi_q^{(2)}(t) \right].$$

Using $\phi_v(t) = t^{-a}$, which implies $\phi_v(z) = \frac{\Gamma(1-a)}{z^{1-2a}}$, we obtain a quantitative expression for the power law index

$$\frac{z \Gamma^2(1-a)}{z^{2-2a}} = \lambda \frac{\Gamma(1-2a)}{z^{1-2a}}.$$

This implies

$$\frac{\Gamma^2(1-a)}{\Gamma(1-2a)} = \lambda = \hat{e}_q C_{qkp} e_k e_p,$$

where $\lambda$ has to be determined numerically. This analysis only applies near the transition density. The assumption of $\psi = \psi^{(1)} + \psi^{(2)}$ and matching the orders might fail in other circumstances. A question remaining in this analysis is the nature of the separation of $\psi_q(z)$ into first and second order terms.

---

The amplitude of the power law is absorbed in the parameter $A$. 

---

Fig. 3 This plot shows $e_k^{MCT}$, the right eigenvector of $C_{qk}^{MCT}$, at the critical density $\eta_{MCT} = 0.515$. The eigenvector is normalized according to the convention $\sum_k e_k e_k = 1$. 

---
3.2 Ergodic-Nonergodic Transition

Having demonstrated the approach corresponding to slow power-law decay, we can push harder for a fuller description at the transition as well as above and below. We take first the case of working at the transition. To see if the mode-coupling approach supports an ergodic-nonergodic transition, we write

$$\phi(q, t) = f_q + g_q(t),$$

with $g_q(t) \to 0$ as $t \to \infty$. This implies $\phi(q, z) = f_q/z + g_q(z)$, with $g_q(z)$ less singular than $1/z$. Let us make this substitution in (3.2):

$$\frac{\Omega^2(f_q/z + g_q(z))}{1 - f_q} = \Omega^2 \mathcal{L}[M(q, t)] + z.$$

Ignoring terms less singular than $1/z$ in the long time limit ($z \to 0$) we obtain

$$\frac{f_q}{1 - f_q} = F_q(f_k). \quad (3.8)$$

Therefore, we find that there is a non-ergodic state when the above equation supports a nontrivial solution for $f_q$.

Solving this equation by iteration reveals that there is indeed an ergodic/non-ergodic transition for hard spheres at $\eta \approx 0.52$, using the Percus-Yevick approximation for the static structure factor.

Next let us determine how the non-ergodicity parameter changes when we perturb the density:

$$\rho \to \rho + \delta \rho.$$ 

Write $f_q = f_q^\rho + (1 - f_q^\rho)^2 g_q$, where $g_q$ is “small”, and substitute into the left hand side of (3.8):

$$\frac{f_q}{1 - f_q} = f_q^\rho + (1 - f_q^\rho)^2 g_q = f_q^\rho + g_q + (1 - f_q^\rho)g_q^2 + O(g^3)$$

The right hand side of (3.8) expands to

$$F_q^\rho + \delta F_q^\rho \frac{f_k^\rho + \delta f_k^\rho}{f_k^\rho} + \delta f_k^\rho + \frac{1}{2} \delta f_k^\rho \delta f_p^\rho \delta f_p + O(g^3, g \rho, \rho^2),$$

where $\delta f_k = (1 - f_k^\rho)^2 g_k$ and $F_q^\rho$ is evaluated at $f_k^\rho$. Using notation introduced in Section 3.1 we obtain

$$F_q^\rho + \delta F_q^\rho \frac{f_k^\rho + \delta f_k^\rho}{f_k^\rho} = F_q^\rho + C_\rho \delta + C_{\rho k} g_k + C_{\rho k p} g_k g_p + O(g^3, g \rho, \rho^2).$$
Combining both sides we obtain an equation for $g_q$:

$$g_q + (1 - f_q^0)g_q^2 = C_q \epsilon + C_q k g_k + C_{q k p} g_k g_p + O(g^3, g \rho, \rho^2).$$

Rewriting in a more suggestive form,

$$(\delta_{qk} - C_{qk})g_k = -(1 - f_q^0)g_q^2 + C_q \epsilon + C_{q k p} g_k g_p + O(g^3, g \rho, \rho^2)$$

(3.9)

At the transition density, $\rho = \rho_c$, $C_{qk}$ has unit eigenvalue. Let us expand $g_k = g_k^{(1)} + g_k^{(2)}$ and assume that $g_k^{(1)}$ is the unit eigenvalue of $C_{qk}$. In that case, we can write $g_k^{(1)} = g_{qk}$, with $g$ a thus far undetermined small parameter. Plugging this form into (3.9) allows us to relate $g$ and the separation parameter $\epsilon = (\rho - \rho_c) / \rho_c$. Expanding to second order in $g$, we obtain

$$(\delta_{qk} - C_{qk})g_k^{(2)} = -(1 - f_q^0)(g_q^{(1)})^2 + C_q \epsilon + C_{q k p} g_k^{(1)} g_p^{(1)} + O(g^3, g \rho, \rho^2).$$

Multiplying both sides by the left eigenvector of $C_{qk}$ to eliminate $g_k^{(2)}$ yields

$$0 = -g^2 \hat{\epsilon}_q (1 - f_q^0) \epsilon_q g + \hat{\epsilon}_q C_q \epsilon + g^2 \hat{\epsilon}_q C_{q k p} \epsilon_k \epsilon_p + O(g^3, g \rho, \rho^2).$$

Recalling the convention for the eigenvectors, $\hat{\epsilon}_q (1 - f_q^0) \epsilon_q g = 1$, and the definition of $\lambda = \hat{\epsilon}_q C_{q k p} \epsilon_k \epsilon_p$, we find

$$g^2 = \frac{\hat{\epsilon}_q C_q \epsilon}{1 - \lambda}.$$

We also know that $C_q$ is positive, since to leading order in the separation parameter $F_q^0 + \delta \rho = (1 + \epsilon)F_q^0$. Therefore our assumptions are consistent near the transition density as long as $\lambda < 1$ and the small parameter in $g_k^{(1)}$ scales as $\sqrt{\epsilon}$. Putting all of this together, the non-ergodicity parameter near the transition density can be written as

$$f_q = f_q^0 + g(1 - f_q^0)^2 \epsilon_q.$$

A plot demonstrating the proper scaling behavior is shown in Figure 5. A similar analysis implies that for the largest eigenvalue $E_0$ of $C_{qk}$, assuming the eigenvalue is one at the critical density, $E_0 = 1 - 2g(1 - \lambda)$, agreeing with the statements in [13]. A plot of the maximum eigenvalue of $C_{qk}$ as a function of the packing fraction is shown in Figure 6.

3.3 Time regimes

In this section we will analyze the asymptotic long-time behavior of $\phi(q, t)$. First we need to find out what happens at the critical density so we can match the other solutions to it in the limit $\epsilon \to 0$. The analysis of $\phi(q, t)$ at the critical density is essentially the same as in Section 3.1 but we include it here for completeness. In the non-ergodic (glass) case we have only one large time to consider. In the ergodic (liquid) case there are two time regimes; the power law decay as in the solid case followed by the von Schweidler decay. To analyze these regimes we rescale the time $t$ by introducing a large constant $\omega$, such that $\tau = \omega \epsilon_q t$. In this rescaling the power law decay corresponds to $\tau \ll 1$ and the von Schweidler decay to $\tau \gg 1$.

3.3.1 Critical Density

Here we are interested in the asymptotic behavior of $\phi(q, t)$ at large times. First we consider what happens at the critical density. As in Section 3.1, we apply the ansatz $\phi(q, t) = f(q) + (1 - f(q))^2 \psi_q(t)$. We obtain

$$\frac{1}{z} \left( \frac{f_q}{1 - f_q} - f_q |f_k| \right) + (\psi_q(z) - C_q k \psi_k(z)) + z(1 - f_q) \psi_q^2(z) - C_{q k p} \mathcal{L} [\psi_k(t) \psi_p(t)] = 0,$$

where the first term (proportional to $1/z$) vanishes by definition. Again, considering a further expansion of $\psi_q(z) = \psi_q^{(1)} + \psi_q^{(2)}$ with $\psi_q^{(2)} / \psi_q^{(1)} \to 0$ as $z \to 0$, we plug into our equation and match orders. To the order of $\psi_q^{(1)}$, we obtain

$$\psi_q^{(1)}(z) = C_{q k} \psi_k^{(1)}(z).$$
Therefore, we may write \( \psi_q^{(1)}(z) = Ae_q \phi_\nu(z) \). Plugging this back into (3.10) and multiplying by the left eigenvector of \( C_q \) to eliminate \( \psi_q^{(2)} \), we obtain an equation for \( \phi_\nu(z) \):

\[
z \phi^2_\nu(z) = \lambda L[\phi^2_\nu(t)].
\]

### 3.3.2 Glass Side

Next let us consider what happens when we are slightly away from the critical density on the glass side. Here we use the ansatz \( \phi(q, z) = f_q / z + (1 - f_q)g \psi(q, z) \), with \( \psi(q, t) \to e_q \) as \( t \to \infty \). \( g \) was computed in Section 3.2. Starting from (3.2) and expanding to second order in \( g \),

\[
\frac{1}{z} \left( \frac{f_q}{1-f_q} - F_q[f_k] - C_q \frac{\delta \rho}{\rho} \right) + g \left( \psi_q(z) - C_q \psi_k(z) - \delta C_q \psi_k(z) \right) + g^2 \left( z(1-f_q) \psi_q^2(z) - C_q \rho L[\psi_k(t) \psi_p(t)] \right) = 0.
\]

According to our assumptions the first two terms in the \( 1/z \) factor cancel. We are left with

\[
-\frac{1}{z} \left( C_q \frac{\delta \rho}{\rho} \right) + g \left( \psi_q(z) - C_q \psi_k(z) - \delta C_q \psi_k(z) \right) + g^2 \left( z(1-f_q) \psi_q^2(z) - C_q \rho L[\psi_k(t) \psi_p(t)] \right) = 0.
\]

Let us re-write \( \delta \rho/\rho \) as \( g^2B \). \( B \) can be determined from the expression for \( g^2 \) in Section 4. Note that \( \epsilon = \delta \rho/\rho \). Also note that \( \delta C_q \rho = gD_q \) since \( \delta C_q \rho \) scales as \( \sqrt{E} \). We obtain

\[
g \left( \psi_q(z) - C_q \psi_k(z) \right) + g^2 \left( z(1-f_q) \psi_q^2(z) - C_q \rho L[\psi_k(t) \psi_p(t)] - D_q \psi_k(z) - \frac{C_q B}{z} \right) = 0.
\]

Using the same method as before, we split \( \psi_k = \psi^{(1)}_k + g \psi^{(2)}_k \). We find

\[
\psi^{(1)}_k(z) = Ae_k \psi_\nu(z).
\]

Using this result and eliminating \( \psi^{(2)}_k \) by multiplying by the left eigenvector we obtain an equation for \( \psi_k(z) \):

\[
z \psi^2_k(z) - \lambda L[\psi^2_k(t)] - D' \psi_k(z) - \frac{B'}{z} = 0,
\]

\[3.11\]
which is slightly different from (3.7). Now change frequency variables to \( \zeta = z/\omega_c \), with \( \varepsilon \to 0 \). \( \omega_c / \varepsilon \to 0 \) \( z \to 0 \) and \( \zeta \) constant. This implies \( \mathcal{L}[f(t)](z) \to \omega_c^{-1} \mathcal{L}[f(\tau)](\zeta) \), where \( \tau = \omega_c t \). Rewriting according to this change of variables:

\[
\frac{1}{\omega_c} \left\{ \zeta \psi_\nu^2(\zeta) - \lambda \mathcal{L}[\psi_\nu^2(\tau)](\zeta) - D' \psi_\nu(\zeta) - B' \zeta \right\} = 0 \tag{3.12}
\]

We see that we obtain the same equation as before, but with \( z \to \zeta \). Now let us make the ansatz \( \psi_\nu(\tau) = 1 + \tau^{-a} \). This implies \( \psi_\nu(\zeta) = 1/\zeta + \Gamma(1-a)/\zeta^{1-a} \). Substituting this in (3.12),

\[
\frac{1}{\omega_c} \left\{ \zeta \psi_\nu^2(\zeta) - \lambda \mathcal{L}[\psi_\nu^2(\tau)](\zeta) - D' \psi_\nu(\zeta) - B' \zeta \right\} = 0.
\]

In the limit \( \zeta \gg 1 \), \( \tau \ll 1 \), we find that

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

provided \( 0 < a < 1/2 \). In the limit \( \zeta \ll 1 \) the solution goes to its final asymptotic value. Plugging in above allows us to obtain an equation for \( A \) in \( \psi_\nu^{(1)}(t) = Ae^{r_\nu}(t) \), removing some ambiguity from our solution. Evaluating the limit as \( \varepsilon \to 0 \) and matching the solution with what we found at the critical density implies that

\[
\lim_{\varepsilon \to 0} g(1 - f_k)^2 e^{\lambda(1 + \tau^{-a})} = (1 - f_k)^2 Ae^{-\varepsilon^{1/2} t^{-a}}.
\]

Since \( g \sim \varepsilon^{1/2} \), we must have

\[
\varepsilon^{1/2} \tau^{-a} = \text{constant} \times \varepsilon^{-a}.
\]

From the definition \( \tau = \omega_c t \),

\[
\varepsilon^{1/2} \omega_c^{-a} = \text{constant}.
\]

From this we find that \( \omega_c \) scales as \( \varepsilon^{1/2a} \).
3.3.3 Liquid Side

Finally we consider what happens when we are slightly below the critical density (liquid side). Here we need to assume that a slight change from the critical density results in a long plateau region which is close to $f_0^\ast$. Following the same procedure as before we obtain a slightly different equation:

$$\zeta \psi_r(\zeta) - \lambda S(\psi_r(t)(\zeta)) + D' \psi_v(\zeta) + \frac{B'}{\zeta} = 0 .$$

Note the change in sign of the final two terms. This is due to the separation parameter being negative on the liquid side of the transition, implying $g$ is negative there as well. However, the analysis for $\zeta \gg 1$ is basically the same as on the glass side, and we obtain

$$\lambda = \frac{\Gamma(1-a)^2}{\Gamma(1-2a)} .$$

This solution is valid as long as $t_0 \ll t \ll \omega_z^{-1}$, where $t_0$ is a timescale governing the microscopic dynamics and $\omega_z$ was found in the previous section to be $\Omega_0 \times t^{1/2}$ with $\Omega_0$ constant.

For $\zeta \ll 1$ we consider the ansatz $\psi_v(\tau) = 1 - \tau^b$, which implies $\psi_v(\zeta) = 1/\zeta - \Gamma(1+b)/\zeta^{1+b}$. Plugging this into our equation,

$$\zeta \left(1 - 2\frac{\Gamma(1+b)}{\zeta^{1+b}} + \frac{(1+b)^2}{\zeta^{2+b}}\right) - \lambda \left(1 - 2\frac{\Gamma(1+b)}{\zeta^{1+b}} + \frac{(1+2b)}{\zeta^{1+2b}}\right) + D' \left(1 - \frac{\Gamma(1+b)}{\zeta^{1+b}}\right) + \frac{B'}{\zeta} = 0 .$$

Taking the limit, and considering only the most singular terms, we obtain

$$\lambda = \frac{\Gamma(1+b)^2}{\Gamma(1+2b)} .$$

provided $b > 0$. The validity of our solution is based on the assumption that $|z\psi(q,z)| \ll 1$. We used this assumption when we expanded the fraction in $\mathbf{3.2}$ as a geometric series. Since our solution is singular as $z \to 0$, this inequality will eventually be violated rendering our solution invalid. This will happen when

$$g \zeta \frac{1}{\zeta^{1+b}} = g \zeta^{-b} \sim 1 \implies e^{1/2} \omega_z z^{-b} \sim 1 \implies z \sim e^{1/2} \zeta^{-1/2} = \epsilon^\gamma ,$$

where $\gamma = 1/2a + 1/2b$. Therefore this von Schweidler part of the solution is valid when

$$\epsilon^{-1/2a} \ll \Omega_0 \times t \ll \epsilon^{-\gamma} .$$

Thus we have analytically found the famous two step process of a power law to a plateau followed by the von Schweidler decay.

4 Application to FTSPD

It turns out that the asymptotic analysis of the FTSPD is remarkably similar to the MCT analysis. The FTSPD yields the following kinetic equation for the evolution of the normalized density autocorrelation function in the case of Smoluchowski Dynamics:

$$\frac{\partial \phi}{\partial t} + \frac{\partial q^2}{S(q)} \phi(q,t) + \frac{\partial q^2}{2} \int_0^t ds \rho \beta^2 \Sigma_{bb}(q,t-s) \frac{\partial \phi(q,s)}{\partial s} = 0$$

where the memory function kernel is given at second order by

$$\rho^4 \beta^2 \Sigma_{bb}(q,t) = \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} \left[ \tilde{G}_{pp}(k,t)G_{pp}^{(0)}(|q-k|,t) + \tilde{G}_{pp}(k,t)\tilde{G}_{pp}(|q-k|,t) \right] .$$

In writing down $\mathbf{4.1a}$ we have already assumed that the frequency dependence of the 3-point vertices can, reasonably, be dropped in the long-time limit. This amounts to dropping time derivative terms acting on the density-related quantities $G_{pp}^{(0)}$, $G_{pp}$, $\tilde{G}_{pp}$, $G_{pp}$ where $G_{pp}^{(0)}$ is the non-interacting correlation function, and the dressed propagators
\( \mathcal{G}_{pp} \) and \( \dot{\mathcal{G}}_{pp} \) are discussed in some detail in Appendix A. In the following section we demonstrate that this frequency dependence is irrelevant for our asymptotic analysis. To analyze the asymptotic behavior of this equation using the methods of MCT we must examine the Laplace transform of \( \phi(q,t) \):

\[
z\phi(q,z) - 1 + \frac{Dq^2}{S(q)} \phi(q,z) + \dot{Dq}^2 \mathcal{L}[p\beta^2 \Sigma_{BB}(q,t)](z\phi(q,z) - 1) = 0,
\]

where we have used the initial condition \( \phi(q,0) = 1 \) and \( \mathcal{L}[\Sigma_{BB}(q,t)] \) is the Laplace transform of \( \Sigma_{BB}(q,t) \). Dividing by \( z\phi(q,z) - 1 \) we obtain

\[
\dot{D}q^2 \mathcal{L}[\phi(q,z)] = 1 + \dot{D}q^2 \mathcal{L}[p\beta^2 \Sigma_{BB}(q,t)]. \quad (4.1b)
\]

We see that we obtain essentially the same equation as in MCT but with a different kernel.

### 4.1 Long-time behavior of \( \Sigma_{BB}(q,t) \)

For the asymptotic analysis of our kinetic equation \( (4.1b) \), we must determine which terms in the kernel \( \Sigma_{BB}(q,t) \) are relevant at long times. We begin the analysis with an examination of \( \mathcal{G}_{pp} \), defined in the frequency domain by

\[
\mathcal{G}_{pp}(q,\omega) = G_{pp}(0) \sigma_{ij} G_{jk} \sigma_{kl} G_{lp}^0 = V(q)^2 \left\{ G_{pp}^0 G_{BP} G_{BP}^0 + G_{pp}^0 G_{pp} G_{BP}^0 + G_{pp}^0 G_{pp} G_{pp}^0 \right\},
\]

where \( \sigma_{ij} = V(k) \) if \( i \neq j \) and is zero otherwise \([4]\). In the time domain this becomes

\[
\dot{\mathcal{G}}_{pp}(q,t) = V(q)^2 \int_{-\infty}^{\infty} ds \int_{-\infty}^{\infty} ds' \left\{ G_{pp}^0(t-s') G_{BP}(s) G_{BP}^0(s') + G_{pp}^0(t-s') G_{pp}(s) G_{BP}^0(s') \right. \
+ \left. G_{pp}^0(t-s') G_{pp}(s) G_{pp}^0(s') \right\},
\]

where we have suppressed the wave-number dependence on the right-hand side. We shall now examine each of the three terms in the integrand in turn. Using the fluctuation dissipation theorem, we have for the first term

\[
\beta^{-2} \int_{-\infty}^{\infty} ds \int_{-\infty}^{\infty} ds' G_{pp}^0(t-s') G_{BP}(s) G_{BP}^0(s') = \int_{-\infty}^{\infty} ds \int_{-\infty}^{\infty} ds' G_{pp}^0(t-s') \theta(-s) \dot{G}_{pp}(s) \theta(-s') \dot{G}_{pp}^0(-s') \
= \int_{-\infty}^{0} ds \int_{-\infty}^{\infty} ds' G_{pp}^0(t-s') G_{pp}(s) G_{pp}^0(-s') \
= \frac{1}{2} G_{pp}^0(0) \int_{-\infty}^{0} ds G_{pp}^0(t-s) G_{pp}(s),
\]

where we have performed the integral over \( s' \) in the final line. For the second term we have

\[
\beta^{-2} \int_{-\infty}^{\infty} ds \int_{-\infty}^{\infty} ds' G_{pp}^0(t-s') G_{pp}(s) G_{BP}^0(s') \
= \int_{-\infty}^{0} ds \int_{-\infty}^{\infty} ds' \theta(t-s') G_{pp}^0(t-s') G_{pp}(s) \theta(-s') G_{pp}^0(-s') \
= \frac{1}{2} G_{pp}^0(0) \int_{-\infty}^{0} ds G_{pp}^0(t-s') G_{pp}(s) G_{pp}^0(-s') \
= \frac{1}{2} G_{pp}^0(0) \int_{-\infty}^{0} ds G_{pp}^0(t-s') G_{pp}(s) G_{pp}^0(-s') \
= -Dq^2 \int_{-\infty}^{0} ds \int_{-\infty}^{\infty} ds' G_{pp}^0(t-s') G_{pp}(s) G_{pp}^0(s') + G_{pp}^0(0) \int_{t}^{\infty} G_{pp}^0(t-s) G_{pp}(s) ds
\]

\[+ G_{pp}^0(0)^2 G_{pp}(t). \]
Finally, the third term becomes

\[ \beta^{-2} \int_{-\infty}^{\infty} ds \int_{-\infty}^{\infty} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s') = \int_{0}^{\infty} ds \int_{-\infty}^{\infty} ds' \theta(t-s')G_{pp}^{(0)}(t-s') \theta(s)G_{pp}(s)G_{pp}^{(0)}(s') \]

\[ = \int_{0}^{\infty} ds \int_{-\infty}^{t-s} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s') \]

\[ = -Dq^2 \int_{0}^{\infty} ds \int_{-\infty}^{t-s} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s'). \]

Next, using the fact that \( G_{pp}^{(0)}(t) = \pm Dq^2 G_{pp}(t) \) to add up the double integrals from the second and third terms (with the sign in front of \( q^2 \) depending on the sign of \( t \)), we obtain

\[ -Dq^2 \int_{0}^{t} ds \int_{-\infty}^{t-s} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s') - Dq^2 \int_{0}^{\infty} ds \int_{-\infty}^{t-s} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s') \]

\[ = -Dq^2 \int_{0}^{t} ds \int_{0}^{t-s} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s') - \int_{0}^{\infty} ds G_{pp}(t)G_{pp}^{(0)}(t)G_{pp}(s)ds \]

\[ = \frac{1}{2} G_{pp}(0) \int_{0}^{\infty} ds G_{pp}^{(0)}(t-s)G_{pp}(s). \]

Adding all three terms,

\[ \tilde{G}_{pp}(q,t) = [\beta V(q)]^2 \left\{ -Dq^2 \int_{0}^{t} ds \int_{0}^{t-s} ds' G_{pp}^{(0)}(t-s')G_{pp}(s)G_{pp}^{(0)}(s') - G_{pp}(0) \int_{0}^{t} ds G_{pp}(t-s)G_{pp}(s) + G_{pp}(0)^2 G_{pp}(t) \right\} \]

\[ = [\beta V(q)]^2 G_{pp}(0) \left\{ G_{pp}(0)G_{pp}(t) - \int_{0}^{t} ds [1 + Dq^2(t-s)]G_{pp}(t-s)G_{pp}(s) \right\}. \]

From here we compute the Laplace transform,

\[ \tilde{G}_{pp}(q,z) = [\beta V(q)]^2 G_{pp}(0) \left\{ G_{pp}(0)(t=0)G_{pp}(z) - \frac{z+2Dq^2}{(z+Dq^2)^2}zG_{pp}(z) - G_{pp}(0)(t=0) \right\}. \]

We see that the only singularities as \( z \to 0 \) come from \( G_{pp}(z) \). Therefore, in the \( z \to 0 \) limit we find that the most dominant term is

\[ \tilde{G}_{pp}(q,z) \to [\beta V(q)p_0]^2 G_{pp}(q,z), \]

where we have replaced \( G_{pp}^{(0)}(0) \) with \( p_0 \). Taking the inverse transform,

\[ \tilde{G}_{pp}(q,t) = [\beta V(q)p_0]^2 G_{pp}(q,t), \]

where \( t_0 \) is a timescale governing the early-time dynamics. Plugging in our ansatz, \( G_{pp}(q,t) = S(q)(f_q + h_q t)^{1/\alpha} \), and multiplying by \( G_{pp}^{(0)}(t) \),

\[ \mathcal{L}[\tilde{G}_{pp}(q,t)G_{pp}^{(0)}(q,t)](z) = [\beta V(q)p_0]^2 S(q) \mathcal{L}[f_q + h_q t^{1/\alpha}]G_{pp}(t) \]

\[ = [\beta V(q)p_0]^2 S(q)p_0 \left\{ f_q - \frac{1}{z+Dq^2} + \frac{\Gamma(1+\alpha)}{h_q(z+Dq^2)^{1+\alpha}} \right\}. \]

Since this term is not singular as \( z \to 0 \) it does not affect the asymptotic analysis in the lowest order limit. Thus we can ignore the “single particle” contribution to \( \Sigma_{pp} \) in (4.1a). Note that the derivative terms in the kernel add multiples of \( z \) to our Laplace transformed expressions which implies that they go to zero in the limit. It follows that the dominant contribution to the asymptotic equation from the kernel is the “collective” \( \tilde{G}_{pp}(q-k,t) \tilde{G}_{pp}(k,t) \) term.
4.2 Ergodic/Non-ergodic Transition

To determine if an ENE transition occurs we set $\phi(q,z) = f_q/z$ and let $z \to 0$. Plugging into (4.1b), we obtain

$$\frac{D_q^2 f_q/z}{S(q)} \frac{1}{1 - f_q} = 1 + \frac{D_q^2}{z} \mathcal{L}[\rho \beta^2 \Sigma_{BB}(q,t)]$$

with

$$\mathcal{L}[\rho \beta^2 \Sigma_{BB}] = \frac{1}{z} \beta^2 \int \frac{d^3 k}{(2\pi)^3} V(k) S(k) V(q - k) S(q - k) f_k f_{q-k}.$$ 

Putting this form into the equation, and setting

$$F_q[f_k] = S(q) \frac{\beta^2}{2p} \int \frac{d^3 k}{(2\pi)^3} V(k) S(k) V(q - k) S(q - k) f_k f_{q-k}$$

we obtain

$$\frac{1}{z} \frac{f_q}{1 - f_q} = \frac{S(q)}{D_q^2} + \frac{1}{z} F_q[f_k]$$

which becomes

$$\frac{f_q}{1 - f_q} = F_q[f_k]$$

in the $z \to 0$ limit. From here it is apparent that the determination of the ENE transition is identical to MCT, except that we have a different “vertex” or potential.

4.3 Time Dependence at Transition

Ignoring the constant term (which becomes negligible in the $z \to 0$ limit), equation (4.1b) reduces to

$$\frac{\phi(q,z)}{1 - z \phi(q,z)} = \mathcal{L}[\rho \beta^2 \Sigma_{BB}].$$

We apply the ansatz $\phi(q,t) = f(q) + (1 - f(q))^2 \psi_q(t)$. The Laplace transform of the ansatz is given by

$$\phi(q,z) = \frac{f(q)}{z} + (1 - f(q))^2 \psi_q(z).$$

We now plug this into the equation and expand the left hand side to second order in $\psi_q(z):

$$\frac{1}{1 - f_q} \left( \frac{f_q}{z} + (1 - f_q) \psi_q(z) + z(1 - f_q)^2 \psi_q^2(z) \right) = \mathcal{L}[\rho \beta^2 \Sigma_{BB}(q,t)].$$

To apply the ansatz to the right hand side we must analyze $\tilde{G}_{pp}$. Ultimately, we need to compute the Laplace transform of $G_{pp}(q,t) \tilde{G}_{pp}(p,t)$. A prerequisite for this computation is $G_{pp}(q,t)$. Our ansatz will be

$$G_{pp}(q,t) = S(q) (f_q + h_q |t|^{\alpha}).$$

This implies we set $\psi_q(t) = h_q |t|^{\alpha} / (1 - f_q)^2$.

$$\tilde{G}_{pp}(q, \omega) = \frac{V(q)}{2} \left[ (G_{pp}^{(0)}(q, \omega))(G_{pp}(q, \omega) + G_{pp}(q, \omega)) + G_{pp}(q, \omega)(G_{pp}^{(0)}(q, \omega) + G_{pp}^{(0)}(q, \omega)) \right].$$

In the time domain, we have (suppressing wave-number arguments)

$$\tilde{G}_{pp}(t) = \frac{V(q)}{2} \int_{-\infty}^{\infty} ds \left[ (G_{pp}^{(0)}(s)(G_{pp}(t-s) + G_{pp}(t-s)) + G_{pp}(s)(G_{pp}^{(0)}(t-s) + G_{pp}^{(0)}(t-s)) \right]$$

$$= \frac{\beta V(q)}{2} \left\{ \int_t^{\infty} \tilde{G}_{pp}(s-t)G_{pp}^{(0)}(s) ds + \int_{-\infty}^{t} \tilde{G}_{pp}(t-s)G_{pp}^{(0)}(s) ds + \beta^{-1} \int_{-\infty}^{\infty} G_{pp}(t-s)(G_{pp}^{(0)}(s) + G_{pp}^{(0)}(s)) ds \right\}$$

(4.2)
where we have invoked the fluctuation dissipation theorem (FDT) in the second line. Using the FDT on one of the terms in the third integral we obtain
\[ \beta^{-1} \int_{-\infty}^{\infty} G_{pp}(t-s) G_{pp}^{(0)}(s)\, ds = \int_{-\infty}^{0} G_{pp}(t-s) G_{pp}^{(0)}(-s)\, ds \]
\[ = -G_{pp}(t) G_{pp}^{(0)}(0) - \int_{-\infty}^{0} G_{pp}(t-s) G_{pp}^{(0)}(-s)\, ds \]
\[ = -G_{pp}(t) G_{pp}^{(0)}(0) - \int_{-\infty}^{0} \dot{G}_{pp}(t-s) G_{pp}^{(0)}(s)\, ds , \]
where we have used the properties of \( G_{pp}^{(0)}(t) \) in the second and third lines. A similar computation reveals that the other term in the third integral of \( \{4.2\} \) reduces to
\[ \beta^{-1} \int_{-\infty}^{\infty} G_{pp}(t-s) G_{pp}^{(0)}(s)\, ds = -G_{pp}(t) G_{pp}^{(0)}(0) + \int_{0}^{\infty} \dot{G}_{pp}(t-s) G_{pp}^{(0)}(s)\, ds . \]
Adding all the pieces together,
\[ \vec{G}_{pp}(q,t) = \frac{\beta V(q)}{2} \left\{ \int_{0}^{t} G_{pp}(s-t) G_{pp}^{(0)}(s)\, ds + \left[ \int_{-\infty}^{0} + \int_{0}^{\infty} - \int_{-\infty}^{0} \right] \dot{G}_{pp}(t-s) G_{pp}^{(0)}(s)\, ds - 2G_{pp}(t) G_{pp}^{(0)}(0) \right\} \]
\[ = \beta V(q) \left\{ -G_{pp}(z) G_{pp}^{(0)}(t = 0) + (z G_{pp}(z) - G_{pp}(t = 0)) G_{pp}^{(0)}(z) \right\} . \]
Taking the Laplace transform,
\[ \vec{G}_{pp}(q,z) = \beta V(q) \left\{ -G_{pp}(z) G_{pp}^{(0)}(t = 0) + (z G_{pp}(z) - G_{pp}(t = 0)) G_{pp}^{(0)}(z) \right\} . \]
Since \( G_{pp}^{(0)}(z) \) is not singular as \( z \to 0 \), we find that the non-integral term of \( \vec{G}_{pp}(t) \) dominates in the long-time limit. Therefore, we may replace
\[ \vec{G}_{pp}(q,t) \to -\beta \rho_{0} V(q) S(q) (f_{q} + h_{q}|t|^{q}) \]
in this limit. At this point the analysis becomes identical to the MCT case, except that we have a different vertex function. Using the notation of Section 3, let
\[ F_{q}[f_{q}] = \frac{\beta^{2} S(q)}{2p} \int \frac{d^{3}k}{(2\pi)^{3}} V(k) S(k) V(q-k) S(q-k) f_{q} f_{q-k} \]
\[ C_{q} = \rho \frac{\delta F_{q}[f_{q}]}{\delta f_{q}} \]
\[ C_{qk} = \frac{\delta F_{q}[f_{q}]}{\delta f_{k}} (1 - f_{k})^{2} \]
\[ C_{qkp} = \frac{1}{2} \frac{\delta^{2} F_{q}}{\delta f_{k} \delta f_{p}} (1 - f_{k})^{2} (1 - f_{p})^{2} \]
where \( V(q) \) is a pseudo-potential determined in \([4]\). Putting all this together, we find
\[ \lim_{z \to 0} \mathcal{Z}[\Sigma_{BB}(q,t)](z) = \frac{F_{q}[f_{q}]}{z} + C_{qk} \psi_{k}(z) + C_{qkp} \mathcal{Z}[\psi_{k}(t) \psi_{p}(t)] . \]
Therefore,
\[ \frac{1}{z} \left( \frac{f_{q}}{1-f_{q}} - F_{q}[f_{q}] \right) + \left( \psi_{q}(z) - C_{qk} \psi_{k}(z) \right) + z (1 - f_{q}) \psi_{q}^{2}(z) - C_{qkp} \mathcal{Z}[\psi_{k}(t) \psi_{p}(t)] = 0 , \]
and the analysis for the ergodic and non-ergodic regimes now proceeds identically to the mode coupling case, the only difference being the “vertex” in MCT and the pseudo-potential in FTSPD.
Fig. 7 The product $V(k)S(k)$ appearing in the FTSPD kinetic equation kernel $\Sigma_B(q,t) = \frac{\pi}{15q^3} \int \frac{dk}{(2\pi)^3} V_k S_{q-k} S_{k} \phi(q,t) \bar{\phi}(q-k,t)$ at the transition. The negative values of this combination imply that $C^{FT}_q$ is not a positive matrix. This is why we cannot choose $\hat{e}_k$ such that $\hat{e}_k > 0$ for all $k$ as we can in Mode Coupling Theory. Nevertheless, the maximum eigenvalue of $C^{FT}_q$ approaches unity as the packing fraction tends to the transition point implying the analysis is still valid.

5 Numerical Results for Hard Spheres

Solving for $f$ and $\psi$ as suggested by the preceding MCT analysis yields results in close agreement with [13]. We find that with the Percus-Yevick approximation for the structure factor of hard spheres the transition happens at a packing fraction of $\eta_{MCT} = 0.515$ and $\lambda_{MCT} = 0.734$. Solving for the exponents, we find $a = 0.312$ and $b = 0.584$. We find that $\lambda$ increases quite sharply in the vicinity of the critical density. Barrat et. al. report $\eta_{MCT} = 0.525$ and $\lambda_{MCT} = 0.758$ with the Verlet-Weiss approximation.

While conducting these numerical experiments it became clear that convergence near the critical density requires integrating out to fairly high ($k \sigma \sim 100$) wavenumbers. Increasing the range beyond this does not have a noticeable effect on the calculations. In addition, the number of iterations required for the convergence of the non-ergodicity parameter increases quite rapidly as the transition density is approached. For instance, convergence to three digits in the eigenvalue occurs after 60 iterations for $\eta_{MCT} = 0.52$, but it takes over 700 iterations at $\eta_{MCT} = 0.515$.05.

The vertex in MCT has some special properties which allow one to draw conclusions about the eigenvalues and eigenvectors. Because $C^{MCT}_q$ is a positive matrix it can be proved that there is a largest eigenvalue and that it becomes unity in the continuum limit. In addition, it can be shown that the both the right and left eigenvectors, $e_k$ and $\hat{e}_k$, must be positive. In FTSPD the situation is more complicated - as can be seen from Figure 7 the pseudopotential no longer guarantees that $C_q^{FT} \geq 0$. Therefore the same proofs from MCT cannot be carried over to the FTSPD case. However, despite the complication with the pseudopotential, numerical evidence (see Figure 8) indicates that the largest eigenvalue of $C_q^{FT}$ is unity. In addition, the right eigenvector $e_k$ is found to be positive, as in MCT. We also observe the same $\sqrt{\varepsilon}$ scaling behavior in $f_q^{FT}$ that we see in MCT (see Figure 9).

Using the first-order result for the pseudopotential for simplicity, which is essentially the direct correlation function [1, 4], solving the FTSPD for hard spheres with the Percus-Yevick approximation appears to be computationally easier than MCT. Convergence only requires integration to at most $k \sigma \sim 80$ compared to $k \sigma \sim 100$ for MCT. This is expected because the decay of the vertex function is much faster for large wavenumbers in the FTSPD. Convergence requires more iteration closer to the transition similarly to MCT. We find that the ENE transition occurs at a packing fraction of $\eta_{FT} = 0.602$ with a corresponding $\lambda_{FT} = 0.553$. The non-ergodicity parameter at the transition is plotted in Figure 10 while the right and left eigenvectors are plotted in Figures 11 and 12, respectively. Solving for the exponents, we obtain $a = 0.386$ for the power law relaxation and $b = 0.935$ for the von Schweidler relaxation.
Fig. 8 The eigenvalues of $C_{qk}^{FT}$ sorted by eigenvalue modulus. The largest eigenvalue is real, non-degenerate, and approaches unity as the density approaches the transition point. This graph was constructed from $C_{qk}^{FT}$ computed on a $800 \times 800$ grid with a spacing $dk = 0.1$. The spectra for grids down to $400 \times 400$ with $dk = 0.2$ and up to $1600 \times 1600$ with $dk = 0.05$ are essentially identical.

6 Conclusion

We have demonstrated that, to second order in the effective potential, the FTSPD essentially reduces to a mode coupling theory. This shows that FTSPD is a framework from which MCT can be derived and systematic corrections introduced. We find that an ergodic / non-ergodic transition occurs at a packing fraction of $\eta_{FT} = 0.602$, which is in good agreement with some recent simulations and experiments [14]. This agreement prompts us to investigate the effectiveness of the pseudopotential approach for other systems. In addition, we plan to confirm this asymptotic analysis by a direct integration of the FTSPD kinetic equation. Although we have gathered extensive numerical evidence for their veracity, proofs that the largest eigenvalue $E_0$ of $C_{qk}^{FT}$ is real, positive, and nondegenerate and $E_0 \to 1$ as $\eta \to \eta^*$ are still unavailable to the best of our knowledge. In a forthcoming paper we extend this analysis to a treatment of binary mixtures. An analysis of Newtonian dynamics within the framework of FTSPD is given in [7].

A Properties of $\tilde{G}$ and $\bar{G}$

In this appendix we prove that the dressed propagators $\tilde{G}$ and $\bar{G}$ individually satisfy the same fluctuation dissipation theorem that $G$ satisfies:

$$G_{pp}(q,\omega) = \frac{2}{\beta \omega} \text{Im} G_{Bp}(q,\omega).$$

In the time domain,

$$G_{Bp}(q,t) = \beta \theta(-t) G_{pp}(q,-t).$$

This result holds at all orders of perturbation theory. The non-interacting cumulants are given by

$$G_{pp}^{(0)}(q,\omega) = \frac{2 \rho_0 \tilde{D} q^2}{\omega^2 + (\tilde{D} q^2)^2}$$

and

$$G_{Bp}^{(0)}(q,\omega) = G_{Bp}^{(0)}(q,\omega)^* = \frac{\beta \rho_0 \tilde{D} q^2}{i \omega - \tilde{D} q^2}. $$
Fig. 9 This plot shows $f_{\text{FT}}(k\sigma = 7.4)$, the non-ergodicity parameter at wavenumber $k\sigma = 7.4$, as a function of the packing fraction. The dashed line shows a least squares fit of $A\sqrt{\epsilon} + B$ to the data.

Fig. 10 This plot shows $f_{\text{FT}}^k$ at the critical density $\eta_{\text{FT}} = 0.602$.

In the following subsections it will be useful to decompose the cumulants into real and imaginary components:

\[ G_{\rho\rho}^{(0)} = R_0 + iI_0 \quad G_{\rho\rho} = R + iI \]
\[ G_{\rho B}^{(0)} = R_0 - iI_0 \quad G_{\rho B} = R - iI \]
\[ G_{\rho\rho}^{(0)} = \frac{2}{\beta\omega}I_0 \quad G_{\rho\rho} = \frac{2}{\beta\omega}I \]
Fig. 11 This plot shows $\Phi_{\text{FT}}$, the right eigenvector of $C_{\text{FT}}$, at the critical density $\eta_{\text{FT}} = 0.602$. The eigenvector is normalized according to the convention $\sum k \hat{e}_k \hat{e}_k = 1$.

A.1 $\tilde{G}$ FDS

Recall the form of $\tilde{G}_{ij}$ given by

$$\tilde{G}_{ij} = \frac{1}{2} (G_{ix}^{(0)} \sigma_{xy} G_{yj} + G_{ix} \sigma_{xy} G_{yj}^{(0)})$$

Explicitly, this yields

$$\tilde{G}_{\rho B} = \frac{1}{2} (G_{\rho B}^{(0)} \beta V G_{\rho B} + G_{\rho B} \beta V G_{\rho B}^{(0)}) = G_{\rho B}^{(0)} \beta V G_{\rho B},$$

$$\tilde{G}_{B\rho} = \frac{1}{2} (G_{B\rho}^{(0)} \beta V G_{B\rho} + G_{B\rho} \beta V G_{B\rho}^{(0)}) = G_{B\rho}^{(0)} \beta V G_{B\rho},$$

and

$$\tilde{G}_{\rho \rho} = \frac{1}{2} (G_{\rho \rho}^{(0)} \beta V G_{\rho \rho} + G_{\rho \rho}^{(0)} \beta V G_{\rho \rho} + G_{\rho \rho} \beta V G_{\rho \rho}^{(0)} + G_{\rho \rho} \beta V G_{\rho \rho}^{(0)}) .$$

If we write out our contributing terms as real and imaginary components then we have for the imaginary part of $\tilde{G}_{\rho B}$.

$$\text{Im} \tilde{G}_{B\rho} = \frac{1}{2i} (\tilde{G}_{B\rho} - \tilde{G}_{\rho B})$$

$$= \frac{\beta V}{2i} [(R_0 + i I_0)(R + iI) - (R_0 - i I_0)(R - iI)]$$

$$= \beta V (I R_0 + I_0 R) .$$

Looking next at $\tilde{G}_{\rho \rho}$, we have

$$\tilde{G}_{\rho \rho} = \frac{2V}{\omega} (R_0 I + R I_0) .$$

Finally, we obtain the fluctuation dissipation relation

$$\tilde{G}_{\rho \rho} = \frac{2}{\beta \omega} \text{Im} \tilde{G}_{B\rho} .$$
Fig. 12 This plot shows $\tilde{e}_k^T$, the left eigenvector of $C_{FT}^T$, at the critical density $\eta_{FT} = 0.602$. We see that this eigenvector is not positive for all $k$, in contrast to MCT. The reason for this is that $C_{FT}^T$ is not a positive matrix, rendering the Perron-Frobenius theorems inapplicable. The eigenvector is normalized according to the convention $\sum_k \tilde{e}_k \tilde{e}_k = 1$.

A.2 $\tilde{G}$ FDS

We may repeat the same procedure for $\tilde{G}_{ij}$, which is given by

$$\tilde{G}_{ij} = G_{\alpha \alpha}^{(0)} \sigma_{\gamma \gamma} G_{\gamma \gamma}^{(0)} \sigma_{\epsilon \epsilon} G_{\epsilon \epsilon}^{(0)} \delta_{\alpha \gamma \gamma \epsilon \epsilon}.$$

This implies

$$\tilde{G}_{\rho \beta} = G_{\rho \beta}^{(0)} \beta \sqrt{V} G_{\rho \beta} \sqrt{V} G_{\rho \beta}^{(0)}$$

and

$$\tilde{G}_{\rho \rho} = G_{p p}^{(0)} \beta \sqrt{V} G_{\rho \beta} \sqrt{V} G_{\rho \beta}^{(0)} + G_{p p}^{(0)} \beta \sqrt{V} G_{\psi \beta} \sqrt{V} G_{\rho \beta}^{(0)} + G_{p p}^{(0)} \beta \sqrt{V} G_{\rho \beta} \sqrt{V} G_{p p}^{(0)}.$$

Using the same decomposition into real and imaginary components, we have

$$\text{Im } \tilde{G}_{\rho \beta} = \frac{1}{2i} \left( \tilde{G}_{\rho \beta} - \tilde{G}_{\rho \beta}^{(0)} \right)$$

and

$$\tilde{G}_{\rho \rho} = \frac{2 \beta V^2}{\omega} \left[ i_0 (R + iI) (R + iI) (R_0 + iI) (R_0 + iI) - (R_0 - iI) (R_0 + iI) (R_0 - iI) (R_0 + iI) \right]$$

and

$$\tilde{G}_{\rho \rho} = 2 \beta \omega \text{Im } \tilde{G}_{\rho \rho}.$$
We define the Fourier transform of a function \( f(t) \) as
\[
\mathcal{F}[f(t)](\omega) = f(\omega) = \int_{-\infty}^{\infty} e^{i\omega t} f(t) dt
\]
with inverse transform
\[
\mathcal{F}^{-1}[f(\omega)](t) = f(t) = \int_{-\infty}^{\infty} e^{-i\omega t} f(\omega) \frac{d\omega}{2\pi}.
\]
One of the tools we have used extensively is the Laplace transform:
\[
\mathcal{L}[f(t)](z) = \int_{0}^{\infty} e^{-zt} f(t) dt.
\]
We define the convolution of two functions \( f \) and \( g \) as:
\[
(f \ast g)(t) \equiv \int_{0}^{t} f(t-s)g(s)ds
\]
The Laplace transform of a convolution is then
\[
\mathcal{L}[(f \ast g)(t)] = \mathcal{L}[f(t)] \mathcal{L}[g(t)].
\]
In addition we have employed a “discrete” notation for various integral kernels. This is convenient since we are only considering spherically symmetric functions for which the notation corresponds to the discretized form used in the numerical routines. For example,
\[
C_{qk}f_k \equiv \int \frac{d^3k}{(2\pi)^3} C_{qk}f_k
\]
and
\[
\delta_{qk} \equiv (2\pi)^3\delta(q - k).
\]
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