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Hierarchy in the Static Fluctuation-Dissipation Theorem of One-Component Plasmas

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**HIERARCHY IN THE STATIC FLUCTUATION-DISSIPATION
THEOREM OF ONE-COMPONENT PLASMAS**

A Dissertation
Presented to
The Department of Mathematics and Statistics

by

Joshuah T. Heath

In Partial Fulfillment
of the Requirements for the Degree of
Bachelor of Science
in the subject of
Applied Mathematics

University of Vermont
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**HIERARCHY IN THE STATIC FLUCTUATION-DISSIPATION
THEOREM OF ONE-COMPONENT PLASMAS**

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Contents

DEDICATION	iv
ACKNOWLEDGEMENTS	vi
I INTRODUCTION	1
II DEVELOPMENT OF THE FLUCTUATION-DISSIPATION THEOREM 2	
III HIERARCHY OF STATISTICAL MECHANICAL RESPONSE FUNC- TIONS IN ONE-COMPONENT PLASMAS	9
3.1 Introduction to the Nomenclature and Derivation of the Linear and Quadratic FDT	9
3.2 Derivation of the Cubic Static Fluctuation-Dissipation Theorem and the Quadrangle Symmetry	16
3.3 Extention to Higher Orders: The Quartic FDT and the General FDT Formula	20
IV CONCLUSION AND FUTURE WORK	28

Dedicated to the memory of Dr. Ryogo Kubo, whose work on fluctuation phenomena shall always be considered the crowning jewel of non-equilibrium statistical mechanics

*"At quite uncertain times and places,
The atoms left their heavenly path,
And by fortuitous embraces,
Engendered all that being hath.
And though they seem to cling together,
And form associations here,
Yet, soon or late, they burst their tether,
And through the depths of space career.*

*So we who sat, oppressed with science,
As British asses, wise and grave,
Are now transformed to wild Red Lions,
As round our prey we ramp and rave.
Thus, by a swift metamorphosis,
Wisdom turns wit, and science joke,
Nonsense is incense to our noses,
For when Red Lions speak, they smoke.*

*Hail, Nonsense! dry nurse of Red Lions,
From thee the wise their wisdom learn,
From thee they cull those truths of science,
Which into thee again they turn.
What combinations of ideas,
Nonsense alone can wisely form!
What sage has half the power that she has,
To take the towers of Truth by storm?*

*Yield, then, ye rules of rigid reason!
Dissolve, thou too, too solid sense!
Melt into nonsense for a season,
Then in some nobler form condense.
Soon, all too soon, the chilly morning,
This flow of soul will crystallize,
Then those who Nonsense now are scorning,
May learn, too late, where wisdom lies."*

-James Clerk Maxwell, "Molecular Evolution"

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Hierarchy in the Static Fluctuation-Dissipation Theorem of One-Component Plasmas

Joshuah T. Heath

31 Pages

Directed by Kenneth I. Golden, Ph.D.

Fluctuation-dissipation theorems (FDTs) link transport coefficients (density response functions, conductivities, electric susceptibilities, etc.) to equilibrium n -point correlation functions. Of special importance to us is the applications of the FDT to one component plasmas and binary ionic mixtures. When applied to such systems, the fluctuation-dissipation theorem provides invaluable insight into response functions and transport coefficients across the non-equilibrium spectrum. After an in-depth review of the FDT and linear response theory, we focus upon expanding the work of K.I. Golden and G. Kalman (J. Stat. Phys. **3**, 87 (1972); Annals of Phys. **141**, 160 (1982)) , which proposes a nonlinear response theory for magnetic field-free classical plasmas. We attempt to re-formulate the hierarchy of static fluctuation-dissipation relations in terms of external density response functions. This provides a systematic formalism for calculating higher order correlation functions in terms of lower-order ones. In future studies, we plan to derive relationships between the screened and external response functions in the RPA (or any suitable approximation method which takes account of particle correlation effects beyond the RPA). This will then provide insight into the hierarchy of static structure functions and their correlation functions.

Chapter I

INTRODUCTION

In this dissertation, I will focus upon a mathematical investigation into the realm of a classical, one-component plasma in zero magnetic field. In short, this thesis will investigate the mathematical relationship between fluctuations about equilibrium and the responses to external forces in the plasma species afore-mentioned. This theorem, known as the **fluctuation dissipation theorem**, relates the density-response function to the structure function of our multilayer plasma and began as a generalization of the Langevin equation in Brownian motion. Indeed, the fluctuation dissipation theorem (or FDT) can be used to describe the fluctuations about equilibrium of any fluid. Nevertheless, it has immense promise when applied to the realm of plasma physics, and can accurately describe the dynamical and static properties of multilayer plasmas via statistical mechanical perturbation theory. This is of immense importance to astrophysics and technological fabrication due to the fact that the modeling of binary ionic mixtures will help us better understand the stellar interiors in carbon-oxygen white dwarfs, as well as the layered charge particle systems that arise in layered multiple-quantum-well structures. [15]

In the pages that follow, I will mainly look into the **static fluctuation dissipation theorem**, which relates the response function to the equilibrium pair correlation function. The main focus of this work will be to expand the work done by K.I. Golden and G. Kalman on the nonlinear fluctuation dissipation theorem, an expansion upon the static FDTs, a generalization of the second law of thermodynamics, and a major contribution to the statistical study of Coulomb systems. Of particular interest will be the Golden-Kalman-Silevitch paper published in Vol 6 of the 1972 issue of the Journal of Statistical Physics. Whereas the conventional FDT relates the equilibrium correlations of a system and the linear response of said system to an external perturbation, the nonlinear FDT relates said correlations and the *nonlinear* response of the system. This is to be expected—the response of a certain system should not be restricted to merely the linear, and indeed the quadratic and even cubic response functions have been studied before in, not only plasma physics, but also crystal physics and nonlinear optics. [28][2] By utilizing a velocity-average-approximation in addition to fluctuation-dissipation relations, one might be able to establish a hierarchy of response functions of steadily increasing nonlinearity for the static one-component plasma, as investigated in the paper "Plasma response functions, fluctuation-dissipation relations and the Velocity Average Approximation", published in the 1982 edition of Annals of Physics by K.I. Golden and G. Kalman. Their derivation will be recast in a spatial Fourier-transform formalism and expanded, resulting in a general hierarchy of static FDT's.

Chapter II

DEVELOPMENT OF THE FLUCTUATION-DISSIPATION THEOREM

The science of statistical physics began in 1738, when Daniel Bernoulli first established the "billiard ball" model of the gas laws derivation, which relied upon the theory that gas consists of large amounts of high-velocity particles [19]. This embryonic stage of modern kinetic theory, together with Bernoulli's insight into the connection between pressure and temperature via the principle of conservation of mechanical energy, proved to be a forshadow of the revolution against the caloric theory of heat to come a century later with the publication of "On Matter, Living Force, and Heat" by Joule in 1847, which first stated that kinetic energy is converted into heat via frictional forces. Caloric theory, which stated that an increase in heat produced an increased quantity of the repulsive liquid "caloric" being added to each matter particles (and thus explaining thermal expansion), finally gave way to kinetic theory with the publication of a paper by German chemist Karl Kronig, who assumed that particles moved in straight lines until they collided with other matter. This paper paved the way for such men as Rudolf Clausius and James Clerk Maxwell to advance kinetic theory into the latter 19th century, the former introducing the mean-free-path theory of gas molecule dynamics and the latter first theorizing a random locomotion of gas molecules as well as deriving the well-known velocity distribution function.

In 1872, kinetic theory reached its zenith as scientists began to look beyond equilibrium processes and into the realm of **nonequilibrium thermodynamics**. Maxwell's distribution of molecular velocities, published in the paper "Illustration of the Dynamical Theory of Gases" and now given by

$$f(\mathbf{v}) = \left(\frac{m}{2\pi kT}\right)^{3/2} \exp\left(-\frac{m\mathbf{v}^2}{2kT}\right) dv \quad (2.0.0.1)$$

(where m is the mass of each particle, k is Boltzmann's constant, and T is the absolute temperature), we notice that this distribution only holds if we are at thermal equilibrium [9]. To fully understand the transition gases experience when exiting the state of equilibrium, Austrian physicist Ludwig Boltzmann came up with the expression known as the Boltzmann equation. By relating macroscopic laws in terms of microscopic forces between molecular collisions, Boltzmann's equation laid the groundwork for non-equilibrium statistical mechanics (NESM) and is a perfect illustrator of the key idea behind the philosophy of NESM: to understand macroscopic phenomena in a real (i.e., non-ideal) system via looking

from a microscopic vantagepoint.

Nevertheless, one important piece was still missing from statistical physics at the time of Boltzmann that was crucial to the understanding of such non-ideal systems: the consideration of the atom. Though the atom's (and, thus, the molecule's) existence was first postulated near the dawn of the 1800's by French scientist Antoine Lavoisier, the century closed with scientists still less than positive of the atom's existence, or at the very least the role of atoms in macroscopic phenomenon. Many influential physicists, Ernest Mach included, have gone so far as to claim that it was quite possible to completely ignore the role of atomic influences in the world of physical study, and even Boltzmann has been quoted with stating that the fluctuations of large numbers of atoms would vanish on the macroscopic scale; i.e., that such phenomena would be generally invisible to contemporary instruments:

In the molecular theory, we assume that the laws of the phenomena found in nature do not essentially deviate from the limits that they would approach in the case of an infinite number of infinitesimally small molecules...It is indispensable for any application of the infinitesimal calculus to molecular theory; indeed, without it, our model which strictly deals always with a large finite number, would not be applicable to apparently continuous quantities...

–Ludwig Boltzmann, *Lectures on Gas Theory*, translated to English by Stephen G. Brush

Of course, this didn't stop many young physicists at the turn of the century to try and find conclusive evidence for atoms. One such physicist (Albert Einstein) attempted to solve this problem via a study of Brownian motion ("On a Motion of Small Particles in a Stationary Liquid, as Required by the Molecular Kinetic Theory of Heat").

The study of Brownian motion began in 1828, when the Scottish-born botanist Robert Brown began studying the motion of miniscule particles in pollen-infested water [5][8]. These particles exhibited an irregular, seemingly random motion that could not be explained by contemporary science, and their movement soon became known as Brownian motion. The locomotion of these particles was characterized to be independent of each other, to become more rapid in the smaller particles than the larger, to be independent of the density or general composition of the particle in question, to be directly proportional to temperature, and to be perpetual. Only in the mid-19th century did scientists begin to believe that Brownian motion was caused by collisions with the molecules in the fluid. Such men as Christian Wiener, Giovanni Cantoni, Joseph Delsaulx, and Ignace Carbonelle proposed that Brownian motion was caused by some sort of molecular disturbance, either by "ether atoms" in the case of Wiener or the thermal motion of the liquid itself by Cantoni. However, such a theorem faced opposition by the cytologist Karl von Nageli and even soon-to-be Nobel

laureate William Ramsey, who believed there was some other explanation for the random walk of Brownian motion, such as a dichotomy between attractive or repulsive forces felt by the moving particle itself.

The theory of atoms and molecules was finally brought into the forefront of Brownian motion research in the late 19th century, when French physicist Jean Baptiste Perrin suggested that Brownian motion could result from the perpetual collision of elastic molecules. Only in 1905 did a mathematical description come to scientific attention, derived by Albert Einstein and, independently, William Sutherland and Marian von Smoluchowski. In Einstein's paper, classical hydrodynamics was used to determine the effect of molecules on the viscosity of a solvent, and comes to two important results: the connection between the coefficients of viscosity of a liquid with suspended particles (η^*) and that without (η):

$$\eta^* = \eta\left(1 + \frac{5}{2}\phi\right) \quad (2.0.0.2)$$

(where ϕ is the volume fraction occupied by the molecules); and the determination of the coefficient of diffusion (D) of the molecules:

$$D = \mu kT \quad (2.0.0.3)$$

where T is the absolute temperature of the fluid in which the Brownian particles are moving, k is Boltzmann's constant, and μ is the measurement of the mobility of the Brownian particles. The latter result is of great importance to us, as we will see in the derivation of this coefficient to follow.

Einstein's original calculation of the coefficient (commonly known as the Einstein-Sutherland derivation) relies heavily upon two opposing physical theories: Stokes' hydrodynamic theory (which stated that the Brownian particle is much larger than the molecules of the liquid, and is thus unaffected by said molecules' motion) and van't Hoff's osmotic theory (which stated that the particle suspended in the liquid behaves in a similar fashion to the liquid molecules, and is thus bound by the same physical laws as molecular agitation). Though Einstein was able to reconcile these two theories, there was a major problem in Einstein and Sutherland's original derivation. In said derivation, one finds that the average displacement of the Brownian particle as a function of time is

$$\langle \bar{x}^2 \rangle = 2Dt \quad (2.0.0.4)$$

where D is the diffusion coefficient, t is the time interval in question, and the angular brackets denote an equilibrium average. This leads to the conclusion that the mean squared

displacement of the Brownian particle is temporally linear. Unfortunately, such a theory disagreed with experimental results to try and measure the instantaneous velocity of the particles, which yielded inconsistent measurements. Though Einstein was one of the first scientists to give us a mathematical description of Brownian motion and thus accurately determine the existence of molecules, the theory was fundamentally flawed when considering the mean squared displacement of the Brownian particles at short times. The theory wouldn't be corrected until 1908, when French physicist Paul Langevin first derived the famous **Langevin equation**, the first known example of a **stochastic equation**, an equation which describes the time-evolution of a random process. This equation might be derived from looking at the equation of motion of N particles of mass m and radius r , given by

$$m\ddot{x} = f_s(t) + f_b(t) = -\alpha\dot{x}(t) + f_b(t) \quad (2.0.0.5)$$

Here, f_s is the viscous drag force (or Stokes' force) experienced by a particle in the liquid of viscosity η , f_b is the random fluctuating force of the particles, and $\alpha = 6\pi\eta r$ is a constant. By imposing a Markovian approximation, the Langevin equation might be derived as

$$\langle \bar{x}(t)^2 \rangle = \frac{6kT}{\alpha} \left(t - \frac{m}{\alpha} \left(1 - \exp\left(-\frac{\alpha}{m}t\right) \right) \right) \quad (2.0.0.6)$$

This equation agrees with experimental results, and leads to the final form of the Einstein diffusion coefficient D :

$$D = \frac{1}{N} \int_0^\infty dt \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle \quad (2.0.0.7)$$

Note that we no longer have any direct temperature or mobility dependence. Instead, we have the diffusion coefficient, a kinetic coefficient which describes the diffusion of the particles, expressed in terms of the autocorrelation function of the velocity, a dynamic quantity which measures the fluctuation of the Brownian particles' velocity. Thus, we see clearly from the above equation that there is a connection between the macroscopic diffusion of the particles and the microscopic fluctuations, and may indeed be considered one and the same physical phenomenon. Therefore, by examining the equilibrium state of the particles, we may determine properties of the particles outside equilibrium, and we thus come to the conclusion of this exercise: *Einstein's corrected diffusion coefficient for the diffusion of Brownian particles is the first example of a fluctuation-dissipation theorem* [22]. Because of its status, eqn. 2.2.3.24 is also known as a **fluctuation dissipation theorem of the first kind**, and, as we will see later, it is also the first example of a **Green-Kubo formula**. Similarly, we may derive the fluctuation-dissipation theorem of the second kind by utilizing

the generalized Langevin equation, which gives us

$$D = \frac{1}{2\pi\beta^2\tilde{C}_s(0)} \quad (2.0.0.8)$$

where $\tilde{C}_s(\omega)$ is the temporal Fourier transform of the correlation function $C_s(t)$ and where I have taken $\tilde{x}(\omega = 0) = 0$. This is the main result of this section and tells us important information about the first fluctuation dissipation theorem. Basically, the relation above states that the Einstein diffusion coefficient D is only sensitive to long time, equilibrium behavior of $\tilde{C}_s(\omega)$, due to the fact that

$$2\pi\tilde{C}_s(\omega = 0) = \int_{-\infty}^{\infty} dt' C_s(t') = \mathcal{D} \quad (2.0.0.9)$$

where \mathcal{D} is the steady-state drag coefficient. Hence, unless the integral of the first fluctuation-dissipation relation diverges as a result, the dynamic memory of $\tilde{C}_s(\omega)$ are averaged within D . This furthers the deep connection between fluctuations at equilibrium and diffusion, and is thus a statement of the second fluctuation dissipation relation.

In the above, we have looked at the first two relations which relate thermal fluctuations to the diffusion of the system in the example of Brownian motion. Such relations weren't expanded upon until the mid-20th century, when the works of Hazime Mori, Robert Zwanzig, and others generalized the theory of Brownian motion to encompass the evolution of any thermodynamic system via analysis of the random observable X . This value may be any parameter, and the key to its advancement of nonequilibrium statistical mechanics is its relation to the irreversible behavior of the thermodynamical system as a whole. Such a result will lead us to a derivation of most-quoted format of the fluctuation-dissipation theorem, a generalization of the relations stated above to any macroscopic system [31].

The theorem to be described is quite peculiar in the sense that it was developed after a thirty to forty year hiatus after the application of the Langevin equation to Brownian motion. Though some kind of the fluctuation-dissipation theorem was theorized by Harry Nyquist in the 1920's, the now accepted theorem developed by Callen and Welton was published in 1951—over four decades after Einstein introduced the diffusion coefficient. This theorem is given by

$$\Im(\tilde{X}(\omega)) = -\frac{1}{2}\omega\beta S(\omega) \quad (2.0.0.10)$$

where X is an observable and $S(\omega)$ is the power spectrum. The theorem above perfectly relates the response function of the frequency of the system to the correlation function of a pair of observables, and is thus a more general discription of the FD relations mentioned

earlier. Now, the Brownian motion of particles in a liquid solvent can be generalized to the perturbation of any macroscopic system. Furthermore, it is important to note that, whereas the results from the fluctuation dissipation theorem of the second kind related frictional to random forces, (3.3.0.74) relates the response function describing a perturbation to a correlation function. Thus, the above is a fluctuation-dissipation theorem of the first kind. More importantly, recall that the correlation function describes the system at equilibrium, and the response function describes the linear response beyond equilibrium. Thus, the fluctuation-dissipation theorem, a theory which finds its genesis in an attempt to relate microscopic fluctuations to macroscopic dissipative phenomenon, might be considered a bridge between equilibrium and non-equilibrium thermodynamic processes, and was thus a major advancement in contemporary statistical mechanics.

The fluctuation-dissipation theorem might be generalized beyond Brownian motion if we consider the theorem of linear response, which states that, in the presence of a perturbing agent $F(t')$, the change in the average of the observable χ is

$$\Delta\langle X(t)\rangle = \int_{-\infty}^t dt' F(t') \chi_{AX}(t-t') \quad (2.0.0.11)$$

where $\chi_{AX}(t-t')$ is the response function of observables A and X , which is given by

$$\chi_{AX}(t-t') = \langle \{A(0), X(t-t')\} \rangle = \beta \langle \dot{A}(0); X(t-t') \rangle \quad (2.0.0.12)$$

This expression is a generalized fluctuation-dissipation theorem. No longer are we considering the motion of Brownian particles; instead, we have derived a formalism that describes the response to an external perturbation of a general system of particles, and related said response to the canonical correlation. It is important to note where the fluctuation and dissipation come into the picture: here, the fluctuation is a result of the correlation function, and the imaginary part of the dissipation is described by the Fourier transform of the correlation $\chi(\omega)$, where the imaginary and real components of the susceptibility ($\chi''(\omega)$ and $\chi'(\omega)$) might be related via the Kramers-Kronig relations

$$\chi'(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \frac{\chi''(\omega')}{\omega' - \omega} \quad \chi''(\omega) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega' \frac{\chi'(\omega')}{\omega' - \omega} \quad (2.0.0.13)$$

These relations might then lead to integral identities known as sum rules.

In this general framework of susceptibilities, the classical, dynamical FDT is given as

$$\chi''(\omega) = -\frac{1}{2} \omega \beta S(\omega) \quad (2.0.0.14)$$

which becomes

$$S(\mathbf{k}, \omega) = -\frac{2}{\beta n \omega} \chi''(\mathbf{k}, \omega) \quad (2.0.0.15)$$

in a nonuniform medium, where $S(\mathbf{k}, \omega)$ is the Fourier transform of the autocorrelation function, known as the structure factor. Integrating over all possible ω values, we obtain the classical static FDT

$$S(\mathbf{k}) = 1 + n g_{\mathbf{k}} = \frac{1}{\beta n} \hat{\chi}(\mathbf{k},) \quad (2.0.0.16)$$

where $g_{\mathbf{k}}$ is the one-particle distribution function.

The main application of the FDT to be investigated in this work is to electrodynamic systems. The study of electrodynamic linear response found its foundations in the development of the Kramers Konig relations above. [12] The sum rules which resulted from these relations could then be applied to better understand conductivity and dielectric functions, in both the long wavelength and short wavelength (i.e., \mathbf{k} dependent) regimes [36]. The macroscopic consequences of the former have been the topic of intense interest in the scientific community, with such scientists as Klimontovich, Rukhadze, and Silin spending much time on the topic. However, of particular historical interest to us is the work of Golden and Kalman on the relations between the external and the internal conductive and dielectric quantities in the framework of linear response. Though Landau might be considered the father of many of the advancements pertaining to linear response [25], it is Golden and Kalman who presented the groundbreaking idea of a tensor fluctuation-dissipation theorem for multicomponent coulomb systems pervaded by an external magnetic field in 1969.

Chapter III

HIERARCHY OF STATISTICAL MECHANICAL RESPONSE FUNCTIONS IN ONE-COMPONENT PLASMAS

3.1 Introduction to the Nomenclature and Derivation of the Linear and Quadratic FDT

Here, we begin the main bulk of the thesis and presentation of new material. However, before we dive into the original work, let us first briefly review the essential nomenclature of our system—i.e., the classical plasma. We begin by defining the particle density $n(\mathbf{r})$ and its Fourier transform, $n(\mathbf{k})$:

$$n(\mathbf{r}) = \sum_i \delta(\mathbf{r} - x_i) = n_0 + \delta n(\mathbf{r}) \quad n(\mathbf{k}) = \sum_i e^{i\mathbf{k}\cdot x_i} = N\delta_{\mathbf{k}} + \delta n(\mathbf{k}) \quad (3.1.0.17)$$

If we are to consider the static fluctuation-dissipation theorem, then we must consider our system in a state of equilibrium. This system consists of one-component plasma particles, each carrying a charge $\hat{Z}e$ immersed in an external potential $\hat{\phi}(r) = \hat{Q}/r \rightarrow \hat{\phi}(\mathbf{k}) = 4\pi\hat{Q}/\mathbf{k}^2$. Therefore, it is trivial to then define the external potential energy as

$$\hat{U}(\mathbf{k}') = Ze\hat{\phi}(\mathbf{k}') = \frac{4\pi Ze\hat{Q}}{k'^2} \quad (3.1.0.18)$$

it is important to note that $\mathbf{k}' \neq 0$ to preclude the possibility of a divergence in the perturbation. The system itself is described by the Liouville distribution function

$$\Omega = \frac{\exp(-\beta H)}{\int d\Gamma \exp(-\beta H)} \quad (3.1.0.19)$$

In the presence of an external perturbation, the Hamiltonian for our system is perturbed via

$$H = H^{(0)} + \hat{H} \quad (3.1.0.20)$$

where the unperturbed Hamiltonian for a system of N electrons in an electron background is given by

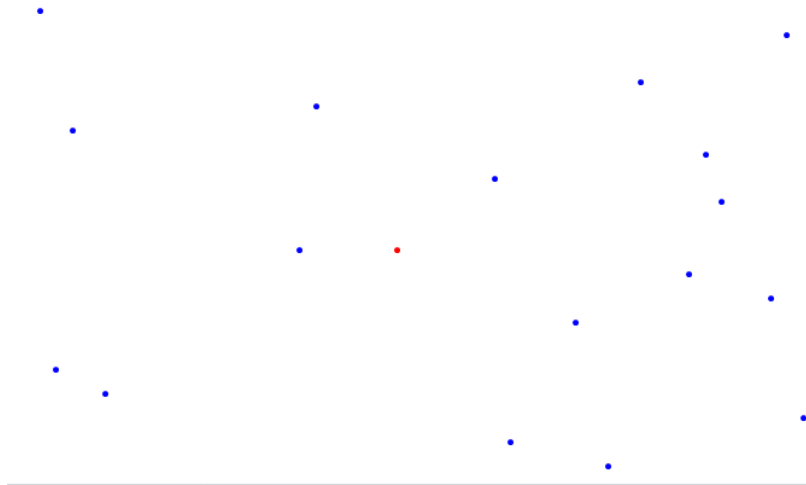


Figure 1: A diagram of our system, where all plasma particles are represented as point charges. The plasma particles (blue) are immersed in a weak external potential energy originating from an external charge Q (red).

$$H^0 = \underbrace{\sum_{i=1}^N \frac{p_i^2}{2m}}_{\text{kinetic energy of particles}} + \frac{1}{2}e^2 \underbrace{\sum_{i,j=1, i \neq j}^N \phi_{ij}(\mathbf{x}_i - \mathbf{x}_j)}_{\text{particle-particle interactions}} + H_{pb} + H_{bb} \quad (3.1.0.21)$$

where H_{pb} and H_{bb} is the Hamiltonian for the particle-background interaction and the background-background interaction, respectively, and

$$\hat{H} = \frac{1}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') n(-\mathbf{k}') \quad (3.1.0.22)$$

with the microscopic density defined as

$$n(-\mathbf{k}') = \sum_i \exp(i\mathbf{k}' \cdot \mathbf{x}_i) \quad (3.1.0.23)$$

which follows directly from (3.1.0.18). Notice that, in (3.1.0.22), we have specifically excluded the $k' = 0$ term. This is due to the fact that, at $k' = 0$, the potential will diverge, and thus is an un-physical solution. Therefore, we now take the equilibrium Liouville distribution as

$$\Omega^{(0)} = \frac{\exp(-\beta H^{(0)})}{\int d\Gamma \exp(-\beta H^{(0)})} \quad (3.1.0.24)$$

and, likewise, define the average microscopic density at equilibrium as

$$\langle \delta n(\mathbf{k}) \rangle^{(0)} = \int d\Gamma \Omega^{(0)} n(\mathbf{k}) - N \delta_{\mathbf{k}} \quad (3.1.0.25)$$

If we are to define the general form of the average microscopic density, we have to introduce the external density response function $\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \dots, \mathbf{k}^q)$, in which the equation of density response becomes a constitutive relation:

$$\langle \delta n(\mathbf{k}) \rangle = \frac{1}{V^{q-1}} \sum_{\mathbf{k}', \mathbf{k}'' \dots \mathbf{k}^q} \hat{\chi}(\mathbf{k}', \mathbf{k}'', \dots, \mathbf{k}^q) \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \dots \hat{U}(\mathbf{k}^q) \delta_{\mathbf{k}'+\mathbf{k}''+\dots+\mathbf{k}^q-\mathbf{k}} \quad (3.1.0.26)$$

where we have taken our sum response out to q th order.

We may similarly define the triangle symmetry for the quadratic structure factor and response function, respectively:

$$S(\mathbf{k}', \mathbf{k}''; -\mathbf{k}) = S(-\mathbf{k}, \mathbf{k}'; \mathbf{k}'') = S(\mathbf{k}'', -\mathbf{k}; \mathbf{k}') \quad (3.1.0.27)$$

$$\hat{\chi}(\mathbf{k}', \mathbf{k}'') = \hat{\chi}(-\mathbf{k}, \mathbf{k}') = \hat{\chi}(\mathbf{k}'', -\mathbf{k}) \quad (3.1.0.28)$$

The hierarchy of static fluctuation-dissipation theorems for a classical, one-component plasma was first tackled by K.I. Golden and G. Kalman in their 1982 *Annals of Physics* paper [14]. In this paper, the static structure factors are expressed in terms of the one, two, and three-particle distribution function g , h , and i :

$$\begin{aligned} S(\mathbf{k}') &= 1 + n g_{\mathbf{k}'} \\ S(\mathbf{k}', \mathbf{k}'') &= 1 + n g_{\mathbf{k}'} + n g_{\mathbf{k}''} + n g_{\mathbf{k}'+\mathbf{k}''} + n^2 h_{\mathbf{k}'\mathbf{k}''} \\ S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') &= 1 + n g_{\mathbf{k}'} + n g_{\mathbf{k}''} + n g_{\mathbf{k}'''} + n g_{\mathbf{k}'+\mathbf{k}''} + n g_{\mathbf{k}''+\mathbf{k}'''} \\ &\quad + n g_{\mathbf{k}'+\mathbf{k}'''} + n g_{\mathbf{k}'+\mathbf{k}''+\mathbf{k}'''} + n^2 h_{\mathbf{k}'\mathbf{k}''} \\ &\quad + n^2 h_{\mathbf{k}''\mathbf{k}'''} + n^2 h_{\mathbf{k}'\mathbf{k}'''} + n^2 h_{\mathbf{k}'+\mathbf{k}''\mathbf{k}'''} + n^2 h_{\mathbf{k}''+\mathbf{k}'''\mathbf{k}'} + n^2 h_{\mathbf{k}'''+\mathbf{k}'\mathbf{k}''} + n^3 i_{\mathbf{k}'\mathbf{k}''\mathbf{k}'''} \end{aligned} \quad (3.1.0.29)$$

From these relations, one may establish the linear, quadratic, and cubic static FDTs, but the mathematical complexity leading to the end result makes it nearly impossible to establish higher orders, such as the quartic. What one needs is a simpler derivation of the above, which might be done by inputting our perturbed Hamiltonian into the Liouville distribution function. This gives us

$$\begin{aligned}
\Omega &= \frac{\exp(-\beta H)}{\int d\Gamma \exp(-\beta H)} \\
&= \frac{\exp(-\beta(H^{(0)} + \hat{H}))}{\int d\Gamma \exp(-\beta(H^{(0)} + \hat{H}))} \\
&= \frac{\exp(-\beta H^{(0)}) \exp(-\beta \hat{H})}{\int d\Gamma \exp(-\beta H^{(0)}) \exp(-\beta \hat{H})}
\end{aligned} \tag{3.1.0.30}$$

Now, if we assume that the perturbation is small enough, we may expand the exponential in the numerator and denominator to obtain

$$\begin{aligned}
\Omega &= \Omega^{(0)} \frac{1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \dots}{\int d\Gamma \Omega^{(0)} \left(1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \dots\right)} \\
&= \Omega^{(0)} \frac{1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \dots}{1 - \beta \langle \hat{H} \rangle + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle - \dots}
\end{aligned} \tag{3.1.0.31}$$

If one wishes to derive the linear static FDT, one need only look at those terms that go as far as \hat{H} , therefore giving us

$$\Omega = \Omega^{(0)} \frac{1 - \beta \hat{H}}{1 - \beta \langle \hat{H} \rangle} = \Omega^{(0)} \left(1 - \beta \hat{H}\right) \left(1 + \beta \langle \hat{H} \rangle\right) \tag{3.1.0.32}$$

where we have taken

$$\frac{1}{1 - \beta \langle \hat{H} \rangle} \approx 1 + \beta \langle \hat{H} \rangle \tag{3.1.0.33}$$

Therefore, we obtain from (3.1.0.32) the following form of the linear Liouville distribution:

$$\begin{aligned}
\Omega^{(1)} &= \Omega^{(0)} \left(1 - \beta \hat{H}\right) \left(1 + \beta \langle \hat{H} \rangle\right) = \Omega^{(0)} \left(1 + \beta \langle \hat{H} \rangle - \beta \hat{H} + \beta^2 \hat{H} \langle \hat{H} \rangle\right) \\
&\approx \Omega^{(0)} + \beta \Omega^{(0)} \left(\langle \hat{H} \rangle - \hat{H}\right) = \Omega^{(0)} + \Omega^{(1)}
\end{aligned} \tag{3.1.0.34}$$

where

$$\Omega^{(1)} = -\beta \Omega^{(0)} \left(\hat{H} - \langle \hat{H} \rangle\right) \tag{3.1.0.35}$$

Now, if one notes that $\langle n(-\mathbf{k}) \rangle = 0$ for $\mathbf{k} \neq 0$, one obtains

$$\langle \hat{H} \rangle = \frac{1}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') \langle n(-\mathbf{k}') \rangle = 0 \quad (3.1.0.36)$$

Therefore,

$$\Omega^{(1)} = \frac{-\beta \Omega^{(0)}}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') n(-\mathbf{k}') \quad (3.1.0.37)$$

Now that we have the first-order Liouville distribution, we calculate the first order density response to obtain our final result. First, a trivial calculation gives

$$\begin{aligned} \langle \delta n(\mathbf{k}) \rangle^{(1)} &= \int d\Gamma \Omega^{(1)} \delta n(\mathbf{k}) \\ &= \int d\Gamma \Omega^{(1)} n(\mathbf{k}) - N \delta_{\mathbf{k}} \int d\Gamma \Omega^{(1)} \\ &= \int d\Gamma \Omega^{(1)} n(\mathbf{k}) \end{aligned} \quad (3.1.0.38)$$

where we have deleted the second term due to the fact that we have taken $\mathbf{k} \neq 0$. This gives us

$$\begin{aligned} \langle \delta n(\mathbf{k}) \rangle^{(1)} &= \int d\Gamma \Omega^{(1)} n(\mathbf{k}) \\ &= \int d\Gamma \left(\frac{-\beta \Omega^{(0)}}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') n(-\mathbf{k}') \right) n(\mathbf{k}) \\ &= -\frac{\beta}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') \int d\Gamma \Omega^{(0)} n(-\mathbf{k}') n(\mathbf{k}) \\ &= -\frac{\beta}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') \langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \\ &= -\frac{\beta}{V} \sum_{\mathbf{k}' \neq 0} \hat{U}(\mathbf{k}') \langle \delta n(\mathbf{k}) \delta n(-\mathbf{k}') \rangle \\ &= -\beta n S(\mathbf{k}) \hat{U}(\mathbf{k}) \end{aligned} \quad (3.1.0.39)$$

The last line of (3.1.0.39) follows from the consequence of the invariance of the homogenous system under spatial translation. However, we know that

$$\langle \delta n(\mathbf{k}) \rangle^{(1)} = \hat{\chi}(\mathbf{k}) \hat{U}(\mathbf{k}) \quad (3.1.0.40)$$

Hence, one obtains the linear external response function as

$$\boxed{\hat{\chi}(\mathbf{k}) = -\beta n S(\mathbf{k})} \quad (3.1.0.41)$$

Note that (3.1.0.41) reproduces the linear fluctuation-dissipation theorem in [14], but recast in the language of a more popular density response function formalism.

Now, let us proceed to the somewhat more involved derivation of the quadratic static FDT. We begin this derivation by expanding the Liouville distribution function to quadratic terms—namely,

$$\Omega = \Omega^{(0)} \frac{1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2}{1 - \beta \langle \hat{H} \rangle + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle} \quad (3.1.0.42)$$

To simplify this, let us start by calculating

$$\frac{1}{1 - \beta \langle \hat{H} \rangle + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle} \quad (3.1.0.43)$$

This might be greatly simplified if we recall that we made the simplification that $\langle \hat{H} \rangle = 0$, thus giving us

$$\frac{1}{1 - \beta \langle \hat{H} \rangle + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle} = \frac{1}{1 + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle} \approx 1 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle \quad (3.1.0.44)$$

Therefore,

$$\begin{aligned} \Omega &= \Omega^{(0)} \left(1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 \right) \left(1 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle \right) \\ &= \Omega^{(0)} \left(1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle + \frac{\beta^3}{2} \hat{H} \langle \hat{H}^2 \rangle - \frac{\beta^4}{4} \hat{H}^2 \langle \hat{H}^2 \rangle \right) \\ &\approx \Omega^{(0)} \left(1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle \right) \\ &= \Omega^{(0)} - \beta \Omega^{(0)} \hat{H} + \frac{\beta^2}{2} \Omega^{(0)} \left(\hat{H}^2 - \langle \hat{H}^2 \rangle \right) \\ &= \Omega^{(0)} + \Omega^{(1)} + \Omega^{(2)} \end{aligned} \quad (3.1.0.45)$$

Hence, we write the quadratic Liouville distribution as

$$\begin{aligned}
\Omega^{(2)} &= \frac{\beta^2}{2} \Omega^{(0)} \left(\hat{H}^2 - \langle \hat{H} \rangle^2 \right) \\
&= \frac{\beta^2}{2V^2} \Omega^{(0)} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \left(n(-\mathbf{k}') n(-\mathbf{k}'') - \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \right) \quad (3.1.0.46)
\end{aligned}$$

Carrying out the second order density response, one obtains

$$\begin{aligned}
\langle \delta n(\mathbf{k}) \rangle^{(2)} &= \int d\Gamma \Omega^{(2)} \delta n(\mathbf{k}) \\
&= \int d\Gamma \Omega^{(2)} n(\mathbf{k}) - N \delta_{\mathbf{k}} \int d\Gamma \Omega^{(2)} \\
&= \int d\Gamma \Omega^{(2)} n(\mathbf{k}) \\
&= \int d\Gamma \left(\frac{\beta^2}{2V^2} \Omega^{(0)} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \left(n(-\mathbf{k}') n(-\mathbf{k}'') - \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \right) \right) n(\mathbf{k}) \\
&= \frac{\beta^2}{2V^2} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \left(\int d\Gamma \Omega^{(0)} n(-\mathbf{k}') n(-\mathbf{k}'') n(\mathbf{k}) - \int d\Gamma \Omega^{(0)} \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle n(\mathbf{k}) \right) \\
&= \frac{\beta^2}{2V^2} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \left(\langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle - N \delta_{\mathbf{k}} \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \right) \\
&= \frac{\beta^2}{2V^2} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \Big|_{\mathbf{k} \neq 0} \quad (3.1.0.47)
\end{aligned}$$

where $\mathbf{k} = \mathbf{k}' + \mathbf{k}''$. Now, we may represent the above in terms of the structure factor via

$$\begin{aligned}
\langle n(\mathbf{k}) \rangle^{(2)} &= \frac{\beta^2}{2V^2} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \Big|_{\mathbf{k} \neq 0} \\
&= \frac{\beta^2 n}{2V} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') S(\mathbf{k}', \mathbf{k}'') \delta_{\mathbf{k}' + \mathbf{k}'' - \mathbf{k}} \\
&= \frac{1}{V} \sum_{\mathbf{k}', \mathbf{k}'' \neq 0} \hat{\chi}(\mathbf{k}', \mathbf{k}'') \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \delta_{\mathbf{k}' + \mathbf{k}'' - \mathbf{k}} \quad (3.1.0.48)
\end{aligned}$$

Eqs. (3.1.0.47) and (3.1.0.48) provide the quadratic FDT:

$$\boxed{\hat{\chi}(\mathbf{k}', \mathbf{k}'') = \frac{\beta^2 n}{2} S(\mathbf{k}', \mathbf{k}'')} \quad (3.1.0.49)$$

which is the same result obtained by Golden, Kalman, and Silevitch in their earlier paper, but again recast here in the more popular density response function formalism [14].

3.2 Derivation of the Cubic Static Fluctuation-Dissipation Theorem and the Quadrangle Symmetry

One of the most important implications of the above derivations of the linear and quadratic static FDTs is its simplicity. The derivation described above is easily used to extend the static FDT to third order. To do this, let us first expand the numerator and denominator of the Liouville distribution function to the cubic order, giving us

$$\Omega = \Omega^{(0)} \frac{1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^3}{6} \hat{H}^3}{1 - \beta \langle \hat{H} \rangle + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle - \frac{\beta^3}{6} \langle \hat{H}^3 \rangle} \quad (3.2.050)$$

Because we have taken $\langle \hat{H} \rangle = 0$ (see above), we ultimately are left to simplify

$$\frac{1}{1 + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle - \frac{\beta^3}{6} \langle \hat{H}^3 \rangle} = 1 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle + \frac{\beta^3}{6} \langle \hat{H}^3 \rangle \quad (3.2.051)$$

Therefore, we now obtain the Liouville distribution out to cubic order as

$$\begin{aligned} \Omega &= \Omega^{(0)} \left(1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^3}{6} \hat{H}^3 \right) \left(1 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle + \frac{\beta^3}{6} \langle \hat{H}^3 \rangle \right) \\ &= \Omega^{(0)} \left(1 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle + \frac{\beta^3}{6} \langle \hat{H}^3 \rangle - \beta \hat{H} + \frac{\beta^3}{2} \hat{H} \langle \hat{H}^2 \rangle - \frac{\beta^4}{6} \hat{H} \langle \hat{H}^3 \rangle \right. \\ &\quad \left. + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^4}{4} \hat{H}^2 \langle \hat{H}^2 \rangle + \frac{\beta^5}{12} \hat{H}^2 \langle \hat{H}^3 \rangle - \frac{\beta^3}{6} \hat{H}^3 + \frac{\beta^5}{12} \hat{H}^3 \langle \hat{H}^2 \rangle \right. \\ &\quad \left. - \frac{\beta^6}{36} \hat{H}^3 \langle \hat{H}^3 \rangle \right) \\ &\approx \Omega^{(0)} \left(1 - \frac{\beta^2}{2} \langle \hat{H}^2 \rangle + \frac{\beta^3}{6} \langle \hat{H}^3 \rangle - \beta \hat{H} + \frac{\beta^3}{2} \hat{H} \langle \hat{H}^2 \rangle + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^3}{6} \hat{H}^3 \right) \\ &= \Omega^{(0)} - \beta \Omega^{(0)} \hat{H} + \frac{\beta^2}{2} \Omega^{(0)} \left(\hat{H}^2 - \langle \hat{H}^2 \rangle \right) - \frac{\beta^3}{6} \Omega^{(0)} \left(\hat{H}^3 - 3 \hat{H} \langle \hat{H}^2 \rangle - \langle \hat{H}^3 \rangle \right) \\ &= \Omega^{(0)} + \Omega^{(1)} + \Omega^{(2)} + \Omega^{(3)} \end{aligned} \quad (3.2.052)$$

Hence, we write the cubic Liouville distribution as

$$\begin{aligned} \Omega^{(3)} &= -\frac{\beta^3}{6} \Omega^{(0)} \left(\hat{H}^3 - 3 \hat{H} \langle \hat{H}^2 \rangle - \langle \hat{H}^3 \rangle \right) \\ &= -\frac{\beta^3}{6V^3} \Omega^{(0)} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \\ &\quad \times \left(n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') - 3n(-\mathbf{k}') \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle - \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \right) \end{aligned} \quad (3.2.053)$$

For the third order density response, one then obtains

$$\begin{aligned}
\langle \delta n(\mathbf{k}) \rangle^{(3)} &= \int d\Gamma \Omega^{(3)} \delta n(\mathbf{k}) \\
&= \int d\Gamma \Omega^{(3)} n(\mathbf{k}) - N \delta_{\mathbf{k}} \int d\Gamma \Omega^{(3)} \\
&= \int d\Gamma \Omega^{(3)} n(\mathbf{k}) \\
&= \int d\Gamma \Omega^{(0)} \left(-\frac{\beta^3}{6V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \left(n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') \right. \right. \\
&\quad \left. \left. - 3n(-\mathbf{k}') \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle - \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \right) \right) n(\mathbf{k}) \\
&= -\frac{\beta^3}{6V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \left(\int d\Gamma \Omega^{(0)} n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(\mathbf{k}) \right. \\
&\quad \left. - 3 \int d\Gamma \Omega^{(0)} n(-\mathbf{k}) n(-\mathbf{k}') \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle - \int d\Gamma \Omega^{(0)} \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(\mathbf{k}) \rangle \right) \\
&= -\frac{\beta^3}{6V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \left(\langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(\mathbf{k}) \rangle \right. \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle - \langle n(\mathbf{k}) n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}''') \rangle \\
&\quad \left. - \langle n(\mathbf{k}) n(-\mathbf{k}''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle - N \delta_{\mathbf{k}} \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \right) \\
&= -\frac{\beta^3}{6V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \left(\langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(\mathbf{k}) \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} - \langle n(\mathbf{k}) n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad \left. - \langle n(-\mathbf{k}) n(-\mathbf{k}''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \Big|_{\mathbf{k} \neq 0} \right) \tag{3.2.0.54}
\end{aligned}$$

where $\mathbf{k} = \mathbf{k}' + \mathbf{k}'' + \mathbf{k}'''$. Notice that we have utilized the triangle symmetry above to expand the three-fold correlation function $3n(-\mathbf{k}') \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle$. This thus allows us to represent the above in terms of the respective structure factors:

$$\begin{aligned}
\langle n(\mathbf{k}) \rangle^{(3)} &= -\frac{\beta^3 n}{6V^2} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \left(S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') \delta_{\mathbf{k}'+\mathbf{k}''+\mathbf{k}'''-\mathbf{k}} \right. \\
&\quad - NS(\mathbf{k}')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}'}\delta_{\mathbf{k}''+\mathbf{k}'''} - NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}'''}\delta_{\mathbf{k}'+\mathbf{k}''} \\
&\quad \left. - NS(\mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}''}\delta_{\mathbf{k}'+\mathbf{k}'''} \right) \\
&= -\frac{\beta^3 n}{6V^2} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \delta_{\mathbf{k}'+\mathbf{k}''+\mathbf{k}'''-\mathbf{k}} \left(S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') \right. \\
&\quad - NS(\mathbf{k}')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}'} - NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}'''} \\
&\quad \left. - NS(\mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}''} \right) \\
&= \frac{1}{V} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''' \neq 0} \hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \delta_{\mathbf{k}'+\mathbf{k}''+\mathbf{k}'''-\mathbf{k}} \quad (3.2.0.55)
\end{aligned}$$

where we have defined the cubic density response function as

$$\begin{aligned}
\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') &= \\
&- \frac{\beta^3 n}{6} \left(S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') - NS(\mathbf{k}')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}'} - NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}'''} - NS(\mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}''} \right) \quad (3.2.0.56)
\end{aligned}$$

Note that (3.2.0.56) is a far more transparent version of the cubic static FDT derived in [14], where the Kronecker delta contributions in (3.2.0.55) were absorbed in the definition of the four-point structure function. However, we may simplify this greatly if we introduce the hierarchal correction coefficient P , which we define in the cubic case as

$$P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \left(NS(\mathbf{k}')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}'} + NS(\mathbf{k}')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}'''} + NS(\mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}''} \right) \quad (3.2.0.57)$$

Therefore giving us the cubic static FDT as

$$\boxed{\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = -\frac{\beta^3 n}{6} S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \quad (3.2.0.58)$$

Notice that, with the above derivation, we obtain a somewhat different result than that obtained by Golden and Kalman in their 1982 article. It has the same general form, but this derivation makes explicit the factor P . This will prove useful when we go onto higher orders in the next subsection.

In the same vein as the quadratic density response function, we note the cubic response obeys a cyclic rotation of its wave vector arguments—a symmetry known as the **quadrangle symmetry**. Recall that, by definition, the triangle symmetry tells us that

$$S(\mathbf{k}', \mathbf{k}''; -\mathbf{k}) = S(\mathbf{k}'', -\mathbf{k}; \mathbf{k}') = S(-\mathbf{k}, \mathbf{k}'; \mathbf{k}'') \quad (3.2.0.59)$$

where it is understood that $\mathbf{k} = \mathbf{k}' + \mathbf{k}''$, thereby guaranteeing the same triangle symmetry for the external response function:

$$\hat{\chi}(\mathbf{k}', \mathbf{k}'') = \hat{\chi}(\mathbf{k}'', -\mathbf{k}) = \hat{\chi}(-\mathbf{k}, \mathbf{k}') \quad (3.2.0.60)$$

via (3.1.0.49). Therefore, we can similarly define the quadrangle symmetry as

$$S(\mathbf{k}', \mathbf{k}'', \mathbf{k}'''; -\mathbf{k}) = S(\mathbf{k}'', \mathbf{k}''', -\mathbf{k}; \mathbf{k}') = S(\mathbf{k}''', -\mathbf{k}, \mathbf{k}'; \mathbf{k}'') = S(-\mathbf{k}, \mathbf{k}', \mathbf{k}''; \mathbf{k}''') \quad (3.2.0.61)$$

The quartic analog of (3.2.0.60) is not as trivial as the above due to the hierarchal correction coefficient $P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')$. Therefore, to illustrate the relation

$$\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = \hat{\chi}(\mathbf{k}'', \mathbf{k}''', -\mathbf{k}) = \hat{\chi}(\mathbf{k}''', -\mathbf{k}, \mathbf{k}') = \hat{\chi}(-\mathbf{k}, \mathbf{k}', \mathbf{k}'') \quad (3.2.0.62)$$

we must first show that the quadrangle symmetry is obeyed in the P -coefficient; that is, that

$$P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = P(\mathbf{k}'', \mathbf{k}''', -\mathbf{k}) = P(\mathbf{k}''', -\mathbf{k}, \mathbf{k}') = P(-\mathbf{k}, \mathbf{k}', \mathbf{k}'') \quad (3.2.0.63)$$

To show this, we first recall (3.2.0.57), and let $\mathbf{k}' \rightarrow \mathbf{k}'', \mathbf{k}'' \rightarrow \mathbf{k}''', \mathbf{k}''' \rightarrow -\mathbf{k}$, and $-\mathbf{k} \rightarrow \mathbf{k}'$. This gives us

$$\begin{aligned} & P(\mathbf{k}'', \mathbf{k}''', -\mathbf{k}) \\ &= 1 - \frac{1}{S(\mathbf{k}'', \mathbf{k}''', -\mathbf{k})} \left(NS(\mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}'+\mathbf{k}''} + NS(\mathbf{k}'')S(-\mathbf{k})\delta_{-\mathbf{k}'+\mathbf{k}} + NS(\mathbf{k}''')S(-\mathbf{k})\delta_{\mathbf{k}'+\mathbf{k}'''} \right) \\ &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \left(NS(\mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}'''} + NS(\mathbf{k}'')S(-\mathbf{k})\delta_{\mathbf{k}-\mathbf{k}'} + NS(\mathbf{k}''')S(-\mathbf{k})\delta_{\mathbf{k}-\mathbf{k}''} \right) \\ &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \left(NS(\mathbf{k}')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}'''} + NS(\mathbf{k}'')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}'} + NS(\mathbf{k}''')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}''} \right) \\ &= P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') \end{aligned} \quad (3.2.0.64)$$

by virtue of noting that $\mathbf{k} = \mathbf{k}' + \mathbf{k}'' + \mathbf{k}'''$ in the Kronecker delta and (3.2.0.59). Similar cyclic permutations yield the other terms in the quadrangle symmetry of the correction factor:

$$\begin{aligned}
& P(\mathbf{k}''', -\mathbf{k}, \mathbf{k}') \\
&= 1 - \frac{1}{S(\mathbf{k}''', -\mathbf{k}, \mathbf{k}')} \left(NS(\mathbf{k}''')S(-\mathbf{k})\delta_{\mathbf{k}''+\mathbf{k}'''} + NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}''+\mathbf{k}'} + NS(-\mathbf{k})S(\mathbf{k}')\delta_{\mathbf{k}''-\mathbf{k}} \right) \\
&= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \left(NS(\mathbf{k}'')S(-\mathbf{k})\delta_{\mathbf{k}-\mathbf{k}'} + NS(\mathbf{k}''')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}''} + NS(\mathbf{k}'')S(\mathbf{k}')\delta_{\mathbf{k}'+\mathbf{k}''} \right) \\
&= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \left(NS(\mathbf{k}'')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}'} + NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}''} + NS(\mathbf{k}''')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}''} \right) \\
&= P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = P(\mathbf{k}'', \mathbf{k}''', -\mathbf{k}') \tag{3.2.0.65}
\end{aligned}$$

$$\begin{aligned}
& P(-\mathbf{k}, \mathbf{k}', \mathbf{k}'') \\
&= 1 - \frac{1}{S(-\mathbf{k}, \mathbf{k}', \mathbf{k}'')} \left(NS(-\mathbf{k})S(\mathbf{k}')\delta_{\mathbf{k}''-\mathbf{k}} + NS(-\mathbf{k})S(\mathbf{k}'')\delta_{\mathbf{k}''+\mathbf{k}''} + NS(\mathbf{k}')S(\mathbf{k}'')\delta_{\mathbf{k}''+\mathbf{k}'} \right) \\
&= 1 - \frac{1}{S(-\mathbf{k}, \mathbf{k}', \mathbf{k}'')} \left(NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}''-\mathbf{k}} + NS(-\mathbf{k})S(\mathbf{k}'')\delta_{-\mathbf{k}+\mathbf{k}'} + NS(\mathbf{k}''')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}''} \right) \\
&= 1 - \frac{1}{S(-\mathbf{k}, \mathbf{k}', \mathbf{k}'')} \left(NS(\mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}''-\mathbf{k}} + NS(\mathbf{k}')S(\mathbf{k}'')\delta_{-\mathbf{k}+\mathbf{k}'} + NS(\mathbf{k}''')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}''} \right) \\
&= P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = P(\mathbf{k}'', \mathbf{k}''', -\mathbf{k}) = P(\mathbf{k}''', -\mathbf{k}, \mathbf{k}') \tag{3.2.0.66}
\end{aligned}$$

3.3 Extention to Higher Orders: The Quartic FDT and the General FDT Formula

In the above subsections, we have rederived the static FDT for one-componet plasmas via an expansion of the Liouville distribution function. Such a derivation is much simpler than the route as expressed in 5.2.2., and might be applied to higher orders with ease. This is shown below for the quartic case; i.e., $\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}''')$. This is the

First, we extend the Liouville distribution function to quartic order, giving us

$$\Omega = \Omega^{(0)} \frac{1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^3}{6} \hat{H}^3 + \frac{\beta^4}{24} \hat{H}^4}{1 - \beta \langle \hat{H} \rangle + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle - \frac{\beta^3}{6} \langle \hat{H}^3 \rangle + \frac{\beta^4}{24} \langle \hat{H}^4 \rangle} \approx \Omega^{(0)} \frac{1 - \beta \hat{H} + \frac{\beta^2}{2} \hat{H}^2 - \frac{\beta^3}{6} \hat{H}^3 + \frac{\beta^4}{24} \hat{H}^4}{1 + \frac{\beta^2}{2} \langle \hat{H}^2 \rangle - \frac{\beta^3}{6} \langle \hat{H}^3 \rangle + \frac{\beta^4}{24} \langle \hat{H}^4 \rangle} \tag{3.3.0.67}$$

Noting that

$$\frac{1}{1 + \frac{\beta^2}{2}\langle\hat{H}^2\rangle - \frac{\beta^3}{6}\langle\hat{H}^3\rangle + \frac{\beta^4}{24}\langle\hat{H}^4\rangle} = 1 - \frac{\beta^2}{2}\langle\hat{H}^2\rangle + \frac{\beta^3}{6}\langle\hat{H}^3\rangle - \frac{\beta^4}{24}\langle\hat{H}^4\rangle + \frac{\beta^4}{4}\langle\hat{H}^2\rangle\langle\hat{H}^2\rangle \quad (3.3.0.68)$$

Therefore,

$$\begin{aligned} \Omega &\approx \Omega^{(0)} \frac{1 - \beta\hat{H} + \frac{\beta^2}{2}\hat{H}^2 - \frac{\beta^3}{6}\hat{H}^3 + \frac{\beta^4}{24}\hat{H}^4}{1 + \frac{\beta^2}{2}\langle\hat{H}^2\rangle - \frac{\beta^3}{6}\langle\hat{H}^3\rangle + \frac{\beta^4}{24}\langle\hat{H}^4\rangle} \\ &= \Omega^{(0)} \left(1 - \beta\hat{H} + \frac{\beta^2}{2}\hat{H}^2 - \frac{\beta^3}{6}\hat{H}^3 + \frac{\beta^4}{24}\hat{H}^4\right) \left(1 - \frac{\beta^2}{2}\langle\hat{H}^2\rangle + \frac{\beta^3}{6}\langle\hat{H}^3\rangle - \frac{\beta^4}{24}\langle\hat{H}^4\rangle + \frac{\beta^4}{4}\langle\hat{H}^2\rangle\langle\hat{H}^2\rangle\right) \\ &= \Omega^{(0)} \left(1 - \frac{\beta^2}{2}\langle\hat{H}^2\rangle + \frac{\beta^3}{6}\langle\hat{H}^3\rangle - \frac{\beta^4}{24}\langle\hat{H}^4\rangle + \frac{\beta^4}{4}\langle\hat{H}^2\rangle\langle\hat{H}^2\rangle - \beta\hat{H} + \frac{\beta^3}{2}\hat{H}\langle\hat{H}^2\rangle - \frac{\beta^4}{6}\hat{H}\langle\hat{H}^3\rangle + \frac{\beta^2}{2}\hat{H}^2 \right. \\ &\quad \left. - \frac{\beta^4}{4}\hat{H}^2\langle\hat{H}^2\rangle - \frac{\beta^3}{6}\hat{H}^3 + \frac{\beta^4}{24}\hat{H}^4\right) \\ &= \Omega^{(0)} - \beta\Omega^{(0)}\hat{H} + \frac{\beta^2}{2}\Omega^{(0)}\left(\hat{H}^2 - \langle\hat{H}^2\rangle\right) - \frac{\beta^3}{6}\Omega^{(0)}\left(\hat{H}^3 - 3\hat{H}\langle\hat{H}^2\rangle - \langle\hat{H}^3\rangle\right) \\ &\quad + \frac{\beta^4}{24}\Omega^{(0)}\left(\hat{H}^4 - 6\hat{H}^2\langle\hat{H}^2\rangle - 4\hat{H}\langle\hat{H}^3\rangle + 6\langle\hat{H}^2\rangle\langle\hat{H}^2\rangle - \langle\hat{H}^4\rangle\right) \\ &= \Omega^{(0)} + \Omega^{(1)} + \Omega^{(2)} + \Omega^{(3)} + \Omega^{(4)} \end{aligned} \quad (3.3.0.69)$$

Therefore, we can now extract the quartic Liouville distribution function, and perform density response:

$$\begin{aligned} \Omega^{(4)} &= \frac{\beta^4}{24}\Omega^{(0)}\left(\hat{H}^4 - 6\hat{H}^2\langle\hat{H}^2\rangle - 4\hat{H}\langle\hat{H}^3\rangle + 6\langle\hat{H}^2\rangle\langle\hat{H}^2\rangle - \langle\hat{H}^4\rangle\right) \\ &= \frac{\beta^4}{24V^3}\Omega^{(0)}\sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0}\hat{U}(\mathbf{k}')\hat{U}(\mathbf{k}'')\hat{U}(\mathbf{k}''')\hat{U}(\mathbf{k}'''') \\ &\quad \times \left(n(-\mathbf{k}')n(-\mathbf{k}'')n(-\mathbf{k}''')n(-\mathbf{k}'''') - 6n(-\mathbf{k}')n(-\mathbf{k}'')\langle n(-\mathbf{k}''')n(-\mathbf{k}'''')\rangle \right. \\ &\quad \left. - 4n(-\mathbf{k}')\langle n(-\mathbf{k}'')n(-\mathbf{k}''')n(-\mathbf{k}'''')\rangle + 6\langle n(-\mathbf{k}')n(-\mathbf{k}'')\rangle\langle n(-\mathbf{k}''')n(-\mathbf{k}'''')\rangle \right. \\ &\quad \left. - \langle n(-\mathbf{k}')n(-\mathbf{k}'')n(-\mathbf{k}''')n(-\mathbf{k}'''')\rangle\right) \end{aligned} \quad (3.3.0.70)$$

$$\begin{aligned}
\langle \delta n(\mathbf{k}) \rangle^{(4)} &= \int d\Gamma \Omega^{(4)} \delta n(\mathbf{k}) \\
&= \int d\Gamma \Omega^{(4)} n(\mathbf{k}) - N \delta_{\mathbf{k}} \int d\Gamma \Omega^{(4)} \\
&= \int d\Gamma \Omega^{(4)} n(\mathbf{k}) \\
&= \int d\Gamma \left(\frac{\beta^4}{24V^3} \Omega^{(0)} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \right. \\
&\quad \times \left(n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') - 6n(-\mathbf{k}') n(-\mathbf{k}'') \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \right. \\
&\quad - 4n(-\mathbf{k}') \langle n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle + 6 \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \\
&\quad \left. \left. - \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \right) \right) n(\mathbf{k}) \\
&= \frac{\beta^4}{24V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \\
&\quad \times \left(\int d\Gamma \Omega^{(0)} n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') n(\mathbf{k}) \right. \\
&\quad - 6 \int d\Gamma \Omega^{(0)} n(-\mathbf{k}') n(-\mathbf{k}'') \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle n(\mathbf{k}) \\
&\quad - 4 \int d\Gamma \Omega^{(0)} n(-\mathbf{k}') \langle n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle n(\mathbf{k}) \\
&\quad + 6 \int d\Gamma \Omega^{(0)} \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle n(\mathbf{k}) \\
&\quad \left. - \int d\Gamma \Omega^{(0)} \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') n(\mathbf{k}) \rangle \right) \\
&= \frac{\beta^4}{24V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \\
&\quad \times \left(\langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') n(\mathbf{k}) \rangle \right. \\
&\quad - 6 \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \\
&\quad - 4 \langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \\
&\quad + 6N \delta_{\mathbf{k}} \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \\
&\quad \left. - N \delta_{\mathbf{k}} \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \right) \\
&= \frac{\beta^4}{24V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \\
&\quad \times \left(\langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') n(\mathbf{k}) \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad - 6 \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad \left. - 4 \langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right)
\end{aligned} \tag{3.3.0.71}$$

$$\begin{aligned}
&= \frac{\beta^4}{24V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \\
&\times \left(\langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') n(\mathbf{k}) \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}'') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}'') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}'''') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') n(-\mathbf{k}'') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') n(-\mathbf{k}') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}'''') n(-\mathbf{k}') \rangle \Big|_{\mathbf{k} \neq 0} \\
&\quad - \langle n(\mathbf{k}) n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}'''') n(-\mathbf{k}') \rangle \Big|_{\mathbf{k} \neq 0}
\end{aligned}
\tag{3.3.0.72}$$

where we have used the quadrangle permutation symmetry to expand the six- and four- fold correlation function. Representing this in terms of the static structure factors, we finally

obtain the quartic static FDT for one-component plasmas:

$$\begin{aligned}
\langle n(\mathbf{k}) \rangle^{(4)} &= \frac{\beta^4}{24V^3} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \delta_{\mathbf{k}'+\mathbf{k}''+\mathbf{k}'''+\mathbf{k}''''-\mathbf{k}} \\
&\times \left(S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') - NS(\mathbf{k}', \mathbf{k}'') S(\mathbf{k}''') \delta_{\mathbf{k}'''+\mathbf{k}''''} \right. \\
&- NS(\mathbf{k}', \mathbf{k}''') S(\mathbf{k}'') \delta_{\mathbf{k}''+\mathbf{k}''''} - NS(\mathbf{k}', \mathbf{k}'''') S(\mathbf{k}'') \delta_{\mathbf{k}'+\mathbf{k}''''} \\
&- NS(\mathbf{k}'', \mathbf{k}'''') S(\mathbf{k}') \delta_{\mathbf{k}'+\mathbf{k}''''} - NS(\mathbf{k}''', \mathbf{k}'''') S(\mathbf{k}') \delta_{\mathbf{k}'+\mathbf{k}''} \\
&- NS(\mathbf{k}'', \mathbf{k}''') S(\mathbf{k}') \delta_{\mathbf{k}'+\mathbf{k}''''} - NS(\mathbf{k}''', \mathbf{k}'''') S(\mathbf{k}') \delta_{\mathbf{k}-\mathbf{k}'} \\
&- NS(\mathbf{k}''', \mathbf{k}'''') S(\mathbf{k}'') \delta_{\mathbf{k}-\mathbf{k}''} - NS(\mathbf{k}'', \mathbf{k}'''') S(\mathbf{k}'') \delta_{\mathbf{k}-\mathbf{k}''''} \\
&\left. - NS(\mathbf{k}'', \mathbf{k}''') S(\mathbf{k}'''') \delta_{\mathbf{k}-\mathbf{k}''''} \right) \\
&= \frac{1}{V} \sum_{\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''' \neq 0} \hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') \hat{U}(\mathbf{k}') \hat{U}(\mathbf{k}'') \hat{U}(\mathbf{k}''') \hat{U}(\mathbf{k}'''') \delta_{\mathbf{k}'+\mathbf{k}''+\mathbf{k}'''+\mathbf{k}''''-\mathbf{k}}
\end{aligned} \tag{3.3.0.73}$$

whence the quartic response function is given by

$$\begin{aligned}
\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') &= \\
&\frac{\beta^4 n}{24} \left(S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') - NS(\mathbf{k}', \mathbf{k}'') S(\mathbf{k}''') \delta_{\mathbf{k}'''+\mathbf{k}''''} \right. \\
&- NS(\mathbf{k}', \mathbf{k}''') S(\mathbf{k}'') \delta_{\mathbf{k}''+\mathbf{k}''''} - NS(\mathbf{k}', \mathbf{k}'''') S(\mathbf{k}'') \delta_{\mathbf{k}'+\mathbf{k}''''} \\
&- NS(\mathbf{k}'', \mathbf{k}'''') S(\mathbf{k}') \delta_{\mathbf{k}'+\mathbf{k}''''} - NS(\mathbf{k}''', \mathbf{k}'''') S(\mathbf{k}') \delta_{\mathbf{k}'+\mathbf{k}''} \\
&- NS(\mathbf{k}'', \mathbf{k}''') S(\mathbf{k}') \delta_{\mathbf{k}'+\mathbf{k}''''} - NS(\mathbf{k}''', \mathbf{k}'''') S(\mathbf{k}') \delta_{\mathbf{k}-\mathbf{k}'} \\
&- NS(\mathbf{k}''', \mathbf{k}'''') S(\mathbf{k}'') \delta_{\mathbf{k}-\mathbf{k}''} - NS(\mathbf{k}'', \mathbf{k}'''') S(\mathbf{k}'') \delta_{\mathbf{k}-\mathbf{k}''''} \\
&\left. - NS(\mathbf{k}'', \mathbf{k}''') S(\mathbf{k}'''') \delta_{\mathbf{k}-\mathbf{k}''''} \right) \\
&= \frac{\beta^4 n}{24} S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')
\end{aligned} \tag{3.3.0.74}$$

with the quartic hierarchal correction coefficient given by

$$\begin{aligned}
P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')} \left(NS(\mathbf{k}', \mathbf{k}'')S(\mathbf{k}''')\delta_{\mathbf{k}'''+\mathbf{k}''''} \right. \\
&\quad + NS(\mathbf{k}', \mathbf{k}''')S(\mathbf{k}'')\delta_{\mathbf{k}''+\mathbf{k}''''} + NS(\mathbf{k}', \mathbf{k}'''')S(\mathbf{k}'')\delta_{\mathbf{k}''+\mathbf{k}''''} \\
&\quad + NS(\mathbf{k}'', \mathbf{k}'''')S(\mathbf{k}')\delta_{\mathbf{k}'+\mathbf{k}''''} + NS(\mathbf{k}''', \mathbf{k}'''')S(\mathbf{k}')\delta_{\mathbf{k}'+\mathbf{k}''''} \\
&\quad + NS(\mathbf{k}'', \mathbf{k}''')S(\mathbf{k}')\delta_{\mathbf{k}'+\mathbf{k}''''} + NS(\mathbf{k}''', \mathbf{k}'''')S(\mathbf{k}')\delta_{\mathbf{k}-\mathbf{k}'} \\
&\quad + NS(\mathbf{k}''', \mathbf{k}'''')S(\mathbf{k}'')\delta_{\mathbf{k}-\mathbf{k}''} + NS(\mathbf{k}'', \mathbf{k}'''')S(\mathbf{k}''')\delta_{\mathbf{k}-\mathbf{k}''} \\
&\quad \left. + NS(\mathbf{k}'', \mathbf{k}''')S(\mathbf{k}'''')\delta_{\mathbf{k}-\mathbf{k}''''} \right) \tag{3.3.0.75}
\end{aligned}$$

We remind the reader that $P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')$, like $S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')$ is identical to $P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}''''; -\mathbf{k})$, where $\mathbf{k} = \mathbf{k}' + \mathbf{k}'' + \mathbf{k}''' + \mathbf{k}''''$. Furthermore, we observe that P is invariant under cyclic rotation of its wave vector arguments, a symmetry which we refer to as the pentangle symmetry.

Note that the quartic static FDT given above is an entirely new result. By proposing a new derivation of the FDT hierarchy, we have thus been able to expand to higher orders. More importantly, by examining the first four terms, we can propose a general combinatorical formula for the q th order of the static FDT. Such a general equation might be easily derived via taking into consideration the Taylor expansion of the Liouville distribution function, which gives us

$$\boxed{\chi(\mathbf{k}', \mathbf{k}'', \dots, \mathbf{k}^{(q)}) = (-1)^q \frac{\beta^q n}{q!} S(\mathbf{k}', \mathbf{k}'', \dots, \mathbf{k}^{(q)}) P(\mathbf{k}', \mathbf{k}'' \dots \mathbf{k}^{(q)})} \tag{3.3.0.76}$$

Of course, the hierarchal correction coefficient is a little-bit less trivial. I propose the general form of the coefficient, up to q th order, as

$$P(\mathbf{k}', \mathbf{k}'' \dots \mathbf{k}^{(q)}) = \begin{cases} 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'' \dots \mathbf{k}^{(q)})} \sum_{i=1}^{q-2} \sum_{|i|} \mathcal{U}_{\mathbf{k}} \langle Q_q^{[i]} \rangle, & q \leq 3 \\ 1, & q < 3 \end{cases} \tag{3.3.0.77}$$

Here, $Q_q^{[i]}$ is the i -subset of the set Q_q of all densities $n(-\mathbf{k}')$, $n(-\mathbf{k}'')$, $\dots, n(-\mathbf{k}^{(q)})$, $\sum_{|i|}$ is the set over all i -subsets, and the horseshoe operator $\mathcal{U}_{\mathbf{k}}$ is defined as

$$\mathcal{U} \langle j \rangle_{\mathbf{k}} = \langle n(\mathbf{k}) \ell \rangle \langle j \rangle \Big|_{\mathbf{k} \neq 0}, \quad \ell \neq j \tag{3.3.0.78}$$

where j and ℓ are some product of densities.

To test this theory, we first note that the linear and quadratic cases are trivial, because they have a hierarchal correction coefficient of P . Therefore, let us look at the cubic relation. Setting $q = 3$ in (3.3.0.76), we obtain

$$\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') = -\frac{\beta^3 n}{6} S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') \quad (3.3.0.79)$$

where $P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')$ is given by (3.3.0.77) as

$$\begin{aligned} P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''') &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \sum_{|i|} \mathcal{U}_{\mathbf{k}} \langle Q_3^{|i|} \rangle \\ &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \sum_{|i|} \mathcal{U}_{\mathbf{k}} \langle (n(-\mathbf{k}'), n(-\mathbf{k}''), n(-\mathbf{k}'''))^{|i|} \rangle \\ &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \mathcal{U}_{\mathbf{k}} \left(\langle n(-\mathbf{k}') \rangle + \langle n(-\mathbf{k}'') \rangle + \langle n(-\mathbf{k}''') \rangle \right) \\ &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')} \left(\langle n(\mathbf{k})n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}'')n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\ &\quad \left. + \langle n(\mathbf{k})n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}')n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} + \langle n(\mathbf{k})n(-\mathbf{k}''') \rangle \langle n(-\mathbf{k}')n(-\mathbf{k}'') \rangle \Big|_{\mathbf{k} \neq 0} \right) \end{aligned} \quad (3.3.0.80)$$

which is identical to the form found in the previous subsection, except in terms of the density correlation functions. A similar result might be found for the quartic relationship by setting $q = 4$ in (3.3.0.76) and (3.3.0.77), yielding

$$\hat{\chi}(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}''''') = \frac{\beta^4 n}{24} S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}''''') P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}''''') \quad (3.3.0.81)$$

$$\begin{aligned}
P(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''') &= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')} \sum_{i=1}^2 \sum_{|i|} \bar{U}_{\mathbf{k}} \langle Q_4^{|i|} \rangle \\
&= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')} \left(\sum_{|1|} \bar{U}_{\mathbf{k}} \langle Q_4^{|i|} \rangle + \sum_{|2|} \bar{U}_{\mathbf{k}} \langle Q_4^{|i|} \rangle \right) \\
&= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')} \left(\sum_{|1|} \bar{U}_{\mathbf{k}} \langle (n(-\mathbf{k}'), n(-\mathbf{k}''), n(-\mathbf{k}'''), n(-\mathbf{k}''''))^{|i|} \rangle \right. \\
&\quad \left. + \sum_{|2|} \bar{U}_{\mathbf{k}} \langle (n(-\mathbf{k}'), n(-\mathbf{k}''), n(-\mathbf{k}'''), n(-\mathbf{k}''''))^{|i|} \rangle \right) \\
&= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')} \left(\bar{U}_{\mathbf{k}} \left(\langle n(-\mathbf{k}') \rangle + \langle n(-\mathbf{k}'') \rangle + \langle n(-\mathbf{k}''') \rangle \right) \right. \\
&\quad \left. + \bar{U}_{\mathbf{k}} \left(\langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle + \langle n(-\mathbf{k}') n(-\mathbf{k}''') \rangle \right. \right. \\
&\quad \left. \left. + \langle n(-\mathbf{k}') n(-\mathbf{k}'''') \rangle + \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle + \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \right) \right) \\
&= 1 - \frac{1}{S(\mathbf{k}', \mathbf{k}'', \mathbf{k}''', \mathbf{k}'''')} \left(\langle n(\mathbf{k}) n(-\mathbf{k}') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}'') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \langle n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}''') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}'') n(-\mathbf{k}''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}'''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}'') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}''') \rangle \Big|_{\mathbf{k} \neq 0} \right. \\
&\quad \left. + \langle n(\mathbf{k}) n(-\mathbf{k}''') n(-\mathbf{k}'''') \rangle \langle n(-\mathbf{k}') n(-\mathbf{k}'') \rangle \Big|_{\mathbf{k} \neq 0} \right) \tag{3.3.0.82}
\end{aligned}$$

which is identical to the form derived via an expansion of the Liouville distribution, only in terms (once again) of the density correlation functions.

Chapter IV

CONCLUSION AND FUTURE WORK

The purpose of this dissertation was two-fold. The first was to review the current literature in the FDT and compile a concise summary of its history and development. The second purpose of this thesis, however, was to diverge from this past work and develop a new theorem based upon the development of the nonlinear static FDT for the one-component plasma done by Kenneth Golden, Gabor Kalman, and Michael Silevitch in the 1970s and 1980s. In particular, from the linear, quadratic, and cubic FDT relations developed by Golden and Kalman in 1982, the author and K. Golden then developed a general hierarchy for the q th relations via a Taylor expansion of the Boltzmann factor in the numerator and denominator of the Liouville distribution function and an exploitation of density response theory. From this, (3.3.0.74) and (3.3.0.76) were introduced, which are the main products of this thesis and completely new results.

At this point, one question still remains, and that is where do we go from here? Here, the answer is three-fold. The first path that the authors will take in this present research is to the case of binary ionic mixtures. In the pages covered, we have limited our view to a single-component plasma system. If we expand to the case of binary ionic mixtures, then we would be able to use our model to describe the two-component plasmas found in carbon-oxygen white dwarfs. The second path would be to derive a hierarchy in the dynamical FDT for one-component plasmas. Such a hierarchy could be tested by taking the static limit and observing whether or not we obtain the static relations just described. We may also utilize the general formula derived above to relate the screened density response functions to the external response, and thus generate a formalism for obtaining the cluster expansion for the $q + 1$ -point correlation function for the 3D one-component plasma. Now, we are finally well positioned to expand the constitutive relations for the response function out to cubic order.

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