Higher-order fluctuation-dissipation relations in plasma physics: Binary Coulomb systems

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A recent approach that led to compact frequency domain formulations of the cubic and quartic fluctuation-dissipation theorems (FDTs) for the classical one-component plasma (OCP) [Golden and Heath, J. Stat. Phys. **162**, 199 (2016)] is generalized to accommodate binary ionic mixtures. Paralleling the procedure followed for the OCP, the basic premise underlying the present approach is that a $(\mathbf{k},\omega)$ 4-vector rotational symmetry, known to be a pivotal feature in the frequency domain architectures of the linear and quadratic fluctuation-dissipation relations for a variety of Coulomb plasmas [Golden *et al.*, J. Stat. Phys. **6**, 87 (1972); **29**, 281 (1982); Golden, Phys. Rev. E **59**, 228 (1999)], is expected to be a pivotal feature of the frequency domain architectures of the higher-order members of the FDT hierarchy. On this premise, each member, in its most tractable form, connects a single $(p+1)$-point dynamical structure function to a linear combination of $(p+1)$-order p density response functions; by definition, such a combination must also remain invariant under rotation of their $(\mathbf{k}_1,\omega_1),(\mathbf{k}_2,\omega_2),\ldots,(\mathbf{k}_p,\omega_p)$, $(\mathbf{k}_1+\mathbf{k}_2+\cdots+\mathbf{k}_p,\,\omega_1+\omega_2+\cdots+\omega_p)$ 4-vector arguments. Assigned to each 4-vector is a species index that corotates in lock step. Consistency is assured by matching the static limits of the resulting frequency domain cubic and quartic FDTs to their exact static counterparts independently derived in the present work via a conventional time-independent perturbation expansion of the Liouville distribution function in its macrocanonical form. The proposed procedure entirely circumvents the daunting issues of entangled Liouville space paths and nested Poisson brackets that one would encounter if one attempted to use the conventional time-dependent perturbation-theoretic Kubo approach to establish the frequency domain FDTs beyond quadratic order.

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I. INTRODUCTION

Recently, a rotation-symmetry-based procedure was proposed as a methodology for generating tractable frequency domain formulations of nonlinear fluctuation-dissipation relations for the magnetic field-free one-component plasma (OCP) [1,2]. In the present work, the formalism underlying this procedure will be established on a more rigorous footing and substantially generalized to accommodate multi-ionic plasma mixtures.

Over the decades, the fluctuation-dissipation theorem (FDT) has become a powerful tool in statistical physics. The conventional FDT provides a link between the linear response of a system to a weak external perturbation and equilibrium two-point correlations of the system's fluctuating quantities [3–5]. However, the system response is by no means restricted to being linear. In the family of nonlinear response functions, the properties of quadratic and cubic response functions have been extensively studied in condensed matter physics [6,7], plasma physics [8–12], and nonlinear optics [13–15].

The natural extension of the Kubo time-dependent perturbation-theoretic approach leads to the notion of the hierarchy of dynamical fluctuation-dissipation theorems (FDTs). This is a topic that has been studied by scientists representing a wide range of disciplines, most notably, plasma physics [16–20], nonlinear optics [14,21], chemistry [22,23], and statistical physics [24–28].

The conventional nonlinear dynamical FDTs, in their most commonly accepted, yet most primitive forms, link a single pth- order $(p=2,3,\ldots)$ response function to a combination of $(p+1)$-point correlation functions interfering with each other through their entangled Liouville space paths [21]. For the quadratic FDT [16,17,29] featuring equilibrium three-point correlation functions, this means that two of the three microscopic number or current densities are nested inside Poisson brackets, and, as such, are not so easily amenable to practical computations. Ultimately, a way was found to eliminate the unwieldy Poisson bracket terms through repeated applications of Poisson bracket identities, making it possible to further develop the resulting time domain quadratic FDT into a tractable and concise frequency domain formula. This latter formula provides the link between a single three-point dynamical structure function and a manifestly "triangle symmetric" combination of quadratic response function terms. For the magnetic field-free OCP subjected to an external scalar potential perturbation, this combination remains invariant under rotation of its $(\mathbf{k}_1,\omega_1),(\mathbf{k}_2,\omega_2),(\mathbf{k}_1+\mathbf{k}_2,\omega_1+\omega_2)$ 4-vector arguments.

Looking beyond quadratic order, the conventional Kubo time-dependent perturbative-theoretic approach, its rigor notwithstanding, becomes all the more unwieldy in terms of dealing with the daunting issues of Poisson brackets nested inside Poisson brackets, etc.

To remedy this situation, the procedure, recently proposed by the author and J. T. Heath for the one-component plasma [1,2], entirely circumvents the issue of the nested Poisson brackets and entangled Liouville paths encountered in the conventional Kubo approach. Their procedure is based on

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the premise that the same kind of rotational symmetry that prevails for the frequency domain linear and quadratic FDTs is a sacrosanct feature underlying the frequency domain architectures of the higher-order members of the FDT hierarchy. When applied to the magnetic field-free OCP, this procedure has provided compact and tractable cubic ($p = 3$) and quartic ($p = 4$) frequency domain FDTs. Each features a single $(p + 1)$-point dynamical structure function, now free of interference in the Liouville space paths, expressed as a rotation-invariant symmetric combination of the pth-order density response function terms which, in turn, can be calculated from model equations of motion.

In the present study, the formalism underlying the Golden-Heath procedure is suitably generalized to accommodate the derivations of the higher-order frequency domain FDTs for magnetic field-free multi-ionic plasmas, most notably, binary ionic mixture (BIM) plasmas consisting of two mobile classical ion species immersed in a uniform, inert, and neutralizing background. The extreme conditions of density and temperature for such a configuration are typical of stellar matter where the electrons are highly degenerate and rigid, and where the positive nuclei are fully pressure ionized. Examples are the interiors of carbon-oxygen stars in their helium shell-burning phase [30] and certain type-I presupernova cores [31].

The plan of the paper is as follows: The relevant partial response and structure functions for the binary ionic mixture plasmas are introduced in Sec. II. In Sec. III the cubic and quartic static FDTs are derived via a traditional time-independent perturbation-theoretic approach; each static FDT provides the connection between equal-time structure factors and zero-frequency density response functions. In Sec. IV, the companion frequency domain FDTs are formulated in accordance with the architecture suggesting that the $(p + 1)$-point dynamical structure factor is proportional to a rotation-symmetric linear combination of pth-order density response functions. The proportionality constants are determined by matching the static limits of the resulting frequency domain FDTs to their Sec. III counterparts. The derivations culminate in the Sec. IV development of cubic and quartic *combined* FDTs, each in a form that links a spectral correlation of *combined* microscopic charge densities to a rotation-symmetric linear combination of nonlinear external polarizabilities. Conclusions are drawn in Sec. V.

II. PARTIAL STATIC RESPONSE FUNCTIONS AND STRUCTURE FACTORS

The binary ionic mixture (BIM) or binary Coulomb plasma under consideration here can be described as a two-component mixture of N_A and N_B mobile point ions immersed in a uniform neutralizing background of rigid degenerate electrons; the ions obey classical statistics. Each ion member of species $\sigma(= A,B)$ is endowed with mass m_σ and positive charge $Z^\sigma e$. The entire system occupies the large but bounded volume V. The relevant microscopic particle densities for each species and their spatial Fourier transforms are

$$n_\sigma(\mathbf{r},t) = \sum_{i=1}^{N_\sigma} \delta\big[\mathbf{r} - \mathbf{x}_i^\sigma(t)\big], \quad n_\sigma(\mathbf{k},t) = \sum_{i=1}^{N_\sigma} \exp\big[-i\mathbf{k}\cdot\mathbf{x}_i^\sigma(t)\big], \quad \delta n_\sigma(\mathbf{r},t) = n_\sigma(\mathbf{r},t) - n_{\sigma 0},$$
$$\delta n_\sigma(\mathbf{k},t) = n_\sigma(\mathbf{k},t) - N_\sigma \delta_{\mathbf{k}} \ \ (\sigma = A,B). \tag{1}$$

$n_{\sigma 0} = N_\sigma/V$ and $\delta_{\mathbf{k}}$ is the Kronecker delta.

To prepare the framework for the development of the static FDT hierarchy in Sec. III, we may suppose that the equilibrium system is pervaded by an external Coulomb potential $\hat{\Phi}(\mathbf{r}) = \hat{Q}/r$ originating from the introduction of a weak external charge $\hat{Q}$ located at the origin. The resulting potential $\hat{U}^\sigma(\mathbf{x}_i^\sigma) = Z^\sigma e\hat{\Phi}(\mathbf{x}_i^\sigma)$ of the external force acting on ion i belonging to species $\sigma = A$ or B produces density excitations (to all orders in $\hat{U}^\sigma$). The latter are linked to the former by wave-vector-dependent partial density response functions, defined through the hierarchy of constitutive relations,

$$\langle n_\sigma(\mathbf{k})\rangle^{(1)} = \sum_{\sigma'=A,B}\sum_{\mathbf{k}_1} \hat{\chi}_{\sigma'\sigma}(\mathbf{k}_1)\hat{U}^{\sigma'}(\mathbf{k}_1)\delta_{\mathbf{k}_1-\mathbf{k}} = \sum_{\sigma'} \hat{\chi}_{\sigma\sigma'}(\mathbf{k})\hat{U}^{\sigma'}(\mathbf{k}), \tag{2}$$

$$\langle n_\sigma(\mathbf{k})\rangle^{(2)} = \frac{1}{V}\sum_{\sigma'=A,B}\sum_{\sigma''=A,B}\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2} \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{k}_1,\mathbf{k}_2)\hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\delta_{\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}}, \tag{3}$$

$$\langle n_\sigma(\mathbf{k})\rangle^{(3)} = \frac{1}{V^2}\sum_{\sigma'=A,B}\sum_{\sigma''=A,B}\sum_{\sigma'''=A,B}\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\sum_{\mathbf{k}_3} \hat{\chi}_{\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)\hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\hat{U}^{\sigma'''}(\mathbf{k}_3)\delta_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3-\mathbf{k}}, \tag{4}$$

$$\langle n_\sigma(\mathbf{k})\rangle^{(4)} = \frac{1}{V^3}\sum_{\sigma'=A,B}\sum_{\sigma''=A,B}\sum_{\sigma'''=A,B}\sum_{\sigma^{(4)}=A,B}\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\sum_{\mathbf{k}_3}\sum_{\mathbf{k}_4} \hat{\chi}_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4)$$
$$\times \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\hat{U}^{\sigma'''}(\mathbf{k}_3)U^{\sigma^{(4)}}(\mathbf{k}_4)\delta_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4-\mathbf{k}}, \tag{5}$$

etc. The angular brackets denote an ensemble-averaged quantity: $\langle\cdots\rangle^{(p)} = O(\hat{U}^p)$ denotes ensemble averaging over the perturbed system for $p \geqslant 1$ and over the unperturbed system for $p = 0$. Partial static structure factors are customarily defined via

$$[N_\sigma N_{\sigma'} N_{\sigma''} \cdots N_{\sigma^{(p)}}]^{\frac{1}{p+1}} S_{\sigma'\sigma''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p)\delta_{\mathbf{k}_1+\mathbf{k}_2+\cdots+\mathbf{k}_p-\mathbf{k}}$$
$$= \langle \delta n_{\sigma'}(\mathbf{k}_1)\delta n_{\sigma''}(\mathbf{k}_2)\cdots\delta n_{\sigma^{(p)}}(\mathbf{k}_p)\delta n_\sigma(-\mathbf{k})\rangle^{(0)} \ \ (p = 1,2,3,4,\cdots). \tag{6}$$

The presence of the Kronecker delta in (6) is a consequence of the translational invariance of the homogeneous system in its equilibrium state. For $p = 1$, (6) generates the familiar relation between the two-point static structure factor and pair correlation function:

$$S_{\sigma'\sigma}(k) = \frac{1}{\sqrt{N_{\sigma'}N_\sigma}}\langle \delta n_{\sigma'}(\mathbf{k})\delta n_\sigma(-\mathbf{k})\rangle^{(0)} = \frac{1}{\sqrt{N_{\sigma'}N_\sigma}}\langle n_{\sigma'}(\mathbf{k}) n_\sigma(-\mathbf{k})\rangle^{(0)} - \sqrt{N_{\sigma'}N_\sigma}\,\delta_{\mathbf{k}} = \delta_{\sigma'\sigma} + \sqrt{n_{\sigma'0}n_{\sigma 0}}\,h_{\sigma'\sigma}(k). \tag{7}$$

The invariance of the equilibrium density correlation function (6) under permutation of its microscopic densities,

$$\langle \delta n_{\sigma'}(\mathbf{k}_1)\delta n_{\sigma''}(\mathbf{k}_2)\cdots\delta n_{\sigma^{(p)}}(\mathbf{k}_p)\delta n_\sigma(-\mathbf{k})\rangle^{(0)}$$
$$= \langle \delta n_\sigma(-\mathbf{k})\delta n_{\sigma'}(\mathbf{k}_1)\cdots\delta n_{\sigma^{(p)}}(\mathbf{k}_p)\rangle^{(0)} = \langle \delta n_{\sigma^{(p)}}(\mathbf{k}_p)\delta n_\sigma(-\mathbf{k})\cdots\delta n_{\sigma^{(p-1)}}(\mathbf{k}_{p-1})\rangle^{(0)}$$
$$= \cdots = \langle \delta n_{\sigma''}(\mathbf{k}_2)\delta n_{\sigma'''}(\mathbf{k}_3)\cdots\delta n_\sigma(-\mathbf{k})\delta n_{\sigma'}(\mathbf{k}_1)\rangle^{(0)} \quad (\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 + \cdots + \mathbf{k}_p), \tag{8}$$

implies the invariance under simultaneous rotation of the wave-vector–species index arguments of the corresponding structure factors, viz.,

$$S_{\sigma'\sigma''\sigma'''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p) = S_{\sigma\sigma'\sigma''\cdots\sigma^{(p-1)}\sigma^{(p)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_{p-1}) = S_{\sigma^{(p)}\sigma\sigma'\cdots\sigma^{(p-2)}\sigma^{(p-1)}}(\mathbf{k}_p,-\mathbf{k},\mathbf{k}_1,\ldots,\mathbf{k}_{p-2})$$
$$= \cdots = S_{\sigma''\sigma'''\sigma^{(iv)}\cdots\sigma^{(p)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,\ldots,\mathbf{k}_p,-\mathbf{k}). \tag{9}$$

Note how the species indices corotate in lock step with their companion wave vectors.

III. STATIC FLUCTUATION-DISSIPATION RELATIONS

The routine derivation of the static FDT hierarchy begins with the development of the macrocanonical Liouville density in powers of the perturbing Hamiltonian $\hat{H}$:

$$\Omega = \frac{\exp(-\beta H)}{\int d\mathbf{x}d\mathbf{p}\exp(-\beta H)}, \tag{10a}$$

$$\Omega^{(0)} = \frac{\exp(-\beta H^{(0)})}{\int d\mathbf{x}d\mathbf{p}\exp(-\beta H^{(0)})}, \tag{10b}$$

$$H = H^{(0)} + \hat{H}, \tag{11}$$

$$\hat{H} = \sum_{\sigma'=A,B}\sum_{i=1}^{N_{\sigma'}} \hat{U}^{\sigma'}\big(\mathbf{x}_i^{\sigma'}\big) = \frac{1}{V}\sum_{\sigma'=A,B}\sum_{\mathbf{k}'\neq 0}\hat{U}^{\sigma'}(\mathbf{k}')n_{\sigma'}(-\mathbf{k}') = \frac{1}{V}\sum_{\sigma'=A,B}\sum_{\mathbf{k}'\neq 0}\hat{U}^{\sigma'}(\mathbf{k}')\delta n_{\sigma'}(-\mathbf{k}'). \tag{12}$$

The (0) superscript refers to the unperturbed BIM; $d\mathbf{x}d\mathbf{p}$ is a differential volume element in the $6(N_A + N_B)$-dimensional phase space. Both Ω and $\Omega^{(0)}$ are normalized to unity. The deletion of the $\mathbf{k}' = 0$ contribution to (12) is due to the presence of the neutralizing uniform background, which also interacts with the external charge $\hat{Q}$. The first-stage calculation of the average density response to arbitrary order in $\hat{U}$, which proceeds by expanding (10a) in powers of $\hat{H}$, results in the perturbed Liouville densities,

$$\Omega^{(1)} = -\beta\Omega^{(0)}\hat{H}, \tag{13}$$

$$\Omega^{(2)} = \tfrac{1}{2}\beta^2\Omega^{(0)}[\hat{H}^2 - \langle\hat{H}^2\rangle^{(0)}], \tag{14}$$

$$\Omega^{(3)} = -\tfrac{1}{6}\beta^3\Omega^{(0)}[\hat{H}^3 - 3\hat{H}\langle\hat{H}^2\rangle^{(0)} - \langle\hat{H}^3\rangle^{(0)}], \tag{15}$$

$$\Omega^{(4)} = \tfrac{1}{24}\beta^4\Omega^{(0)}[\hat{H}^4 + 6\langle\hat{H}^2\rangle^{(0)}\langle\hat{H}^2\rangle^{(0)} - 6\hat{H}^2\langle\hat{H}^2\rangle^{(0)} - 4\hat{H}\langle\hat{H}^3\rangle^{(0)} - \langle\hat{H}^4\rangle^{(0)}]. \tag{16}$$

Then with the aid of (12), Eqs. (13)–(16) become

$$\Omega^{(1)} = -\frac{\beta}{V}\Omega^{(0)}\sum_{\sigma'=A,B}\sum_{\mathbf{k}_1\neq\mathbf{0}}\hat{U}^{\sigma'}(\mathbf{k}_1)n_{\sigma'}(-\mathbf{k}_1), \tag{17}$$

$$\Omega^{(2)} = \frac{\beta^2}{2!V^2}\Omega^{(0)} \sum_{\sigma'=A,B}\sum_{\sigma''=A,B}\sum_{\mathbf{k}_1\neq\mathbf{0}}\sum_{\mathbf{k}_2\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\big[n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2) - \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)}\big], \tag{18}$$

$$\begin{aligned}\Omega^{(3)} = {}& \frac{\beta^3}{3!V^3}\Omega^{(0)} \sum_{\sigma'=A,B}\sum_{\sigma''=A,B}\sum_{\sigma'''=A,B}\sum_{\mathbf{k}_1\neq\mathbf{0}}\sum_{\mathbf{k}_2\neq\mathbf{0}}\sum_{\mathbf{k}_3\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\hat{U}^{\sigma'''}(\mathbf{k}_3)[n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\\ &- n_{\sigma'}(-\mathbf{k}_1)\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)} - n_{\sigma''}(-\mathbf{k}_2)\langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma'}(-\mathbf{k}_1)\rangle^{(0)} - n_{\sigma'''}(-\mathbf{k}_3)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)}\\ &- \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}],\end{aligned} \tag{19}$$

$$\begin{aligned}\Omega^{(4)} = {}& \frac{\beta^4}{4!V^4}\Omega^{(0)} \sum_{\sigma'=A,B}\sum_{\sigma''=A,B}\sum_{\sigma'''=A,B}\sum_{\sigma^{(4)}=A,B}\sum_{\mathbf{k}_1\neq\mathbf{0}}\sum_{\mathbf{k}_2\neq\mathbf{0}}\sum_{\mathbf{k}_3\neq\mathbf{0}}\sum_{\mathbf{k}_4\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\hat{U}^{\sigma'''}(\mathbf{k}_3)\hat{U}^{\sigma^{(4)}}(\mathbf{k}_4)\\ &\times [n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4) - \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &+ 2\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)}\langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} + 2\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &+ 2\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)} - n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &- n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} - n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &- n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)} - n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}\\ &- n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)} - n_{\sigma'}(-\mathbf{k}_1)\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &- n_{\sigma''}(-\mathbf{k}_2)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} - n_{\sigma'''}(-\mathbf{k}_3)\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &- n_{\sigma^{(4)}}(-\mathbf{k}_4)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}].\end{aligned} \tag{20}$$

As a further check, one can readily verify that (17)–(20) satisfy the normalization requirements,

$$\int d\mathbf{x}\; d\mathbf{p}\Omega^{(p)} = 0, \;\; (p = 1,2,3,4,\cdots). \tag{21}$$

The calculation of the average density response then proceeds according to the prescription,

$$\langle n_\sigma(\mathbf{k})\rangle^{(p)} = \int d\mathbf{x}\; d\mathbf{p}\Omega^{(p)} n_\sigma(\mathbf{k}) \;\; (p = 1,2,3,4,\cdots) \;\; (\sigma = A,B), \tag{22}$$

keeping in mind that the emergent $(p+1)$-point equilibrium density correlation functions can differ from zero if, and only if, their wave-vector arguments satisfy the homogeneity requirement $\mathbf{k} = \mathbf{k}_1 + \cdots + \mathbf{k}_p$. One readily obtains

$$\langle n_\sigma(\mathbf{k})\rangle^{(1)} = -\frac{\beta}{V}\sum_{\sigma'}\sum_{\mathbf{k}_1\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\langle n_{\sigma'}(-\mathbf{k}_1)n_\sigma(\mathbf{k})\rangle^{(0)}, \tag{23}$$

$$\langle n_\sigma(\mathbf{k})\rangle^{(2)} = \frac{\beta^2}{2!V^2}\sum_{\sigma'}\sum_{\sigma''}\sum_{\mathbf{k}_1\neq\mathbf{0}}\sum_{\mathbf{k}_2\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_\sigma(\mathbf{k})\rangle^{(0)}\Big|_{\mathbf{k}\neq\mathbf{0}}, \tag{24}$$

$$\begin{aligned}\langle n_\sigma(\mathbf{k})\rangle^{(3)} = {}& \frac{\beta^3}{3!V^3}\sum_{\sigma'}\sum_{\sigma''}\sum_{\sigma''}\sum_{\mathbf{k}_1\neq\mathbf{0}}\sum_{\mathbf{k}_2\neq\mathbf{0}}\sum_{\mathbf{k}_3\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\hat{U}^{\sigma'''}(\mathbf{k}_3)[\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_\sigma(\mathbf{k})\rangle^{(0)}\\ &- \langle n_{\sigma'}(-\mathbf{k}_1)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)} - \langle n_{\sigma''}(-\mathbf{k}_2)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma'}(-\mathbf{k}_1)\rangle^{(0)}\\ &- \langle n_{\sigma'''}(-\mathbf{k}_3)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)} - \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}N_\sigma\delta_{\mathbf{k}}],\end{aligned} \tag{25}$$

$$\begin{aligned}\langle n_\sigma(\mathbf{k})\rangle^{(4)} = {}& \frac{\beta^4}{4!V^4}\sum_{\sigma'}\sum_{\sigma''}\sum_{\sigma'''}\sum_{\sigma^{(4)}}\sum_{\mathbf{k}_1\neq\mathbf{0}}\sum_{\mathbf{k}_2\neq\mathbf{0}}\sum_{\mathbf{k}_3\neq\mathbf{0}}\sum_{\mathbf{k}_4\neq\mathbf{0}} \hat{U}^{\sigma'}(\mathbf{k}_1)\hat{U}^{\sigma''}(\mathbf{k}_2)\hat{U}^{\sigma'''}(\mathbf{k}_3)\hat{U}^{\sigma^{(4)}}(\mathbf{k}_4)\\ &\times \Big[\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)n_\sigma(\mathbf{k})\rangle^{(0)}\Big|_{\mathbf{k}\neq\mathbf{0}} + 2N_\sigma\delta_{\mathbf{k}}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)}\langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &+ 2N_\sigma\delta_{\mathbf{k}}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} + 2N_\sigma\delta_{\mathbf{k}}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}\\ &- \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} - \langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}\\ &- \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} - \langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}\\ &- \langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma^{(4)}}(-\mathbf{k}_4)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)} - \langle n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)\rangle^{(0)}\end{aligned}$$

$$- \langle n_{\sigma'}(-\mathbf{k}_1)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} - \langle n_{\sigma''}(-\mathbf{k}_2)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma'''}(-\mathbf{k}_3)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)}$$
$$- \langle n_{\sigma'''}(-\mathbf{k}_3)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma^{(4)}}(-\mathbf{k}_4)\rangle^{(0)} - \langle n_{\sigma^{(4)}}(-\mathbf{k}_4)n_\sigma(\mathbf{k})\rangle^{(0)}\langle n_{\sigma'}(-\mathbf{k}_1)n_{\sigma''}(-\mathbf{k}_2)n_{\sigma'''}(-\mathbf{k}_3)\rangle^{(0)}\big]. \tag{26}$$

The equilibrium density correlation functions in (23)–(26) are next traded for static structure functions via Eq. (6). Subsequent comparisons with constitutive relations (2)–(5) then lead to the first four equations of the static FDT hierarchy:

$$\hat{\chi}_{\sigma'\sigma}(\mathbf{k}_1) = -\beta\sqrt{n_{\sigma'0}n_{\sigma0}}S_{\sigma'\sigma}(\mathbf{k}_1) \quad (\sigma,\sigma' = A,B) \quad (\mathbf{k} = \mathbf{k}_1), \tag{27}$$

$$\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{k}_1,\mathbf{k}_2) = \frac{\beta^2}{2!}\sqrt[3]{n_{\sigma'0}n_{\sigma''0}n_{\sigma0}}S_{\sigma'\sigma''\sigma}(\mathbf{k}_1,\mathbf{k}_2) \quad (\sigma,\sigma',\sigma'' = A,B) \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2), \tag{28}$$

$$\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) = -\frac{\beta^3}{3!}\sqrt[4]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma0}}[S_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) - R_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)]$$
$$(\sigma,\sigma',\sigma'',\sigma''' = A,B) \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3), \tag{29}$$

$$\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) = \frac{\beta^4}{4!}\sqrt[5]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma^{(4)}0}n_{\sigma0}}[S_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) - R_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4)]$$
$$(\sigma,\sigma',\sigma'',\sigma''',\sigma^{(4)} = A,B) \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4), \tag{30}$$

where the static (zero-frequency) $\hat{\chi}'s$ are real, and where

$$R_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) = \sqrt[4]{N_{\sigma'}N_{\sigma''}N_{\sigma'''}N_\sigma}[S_{\sigma'\sigma}(\mathbf{k}_1)S_{\sigma''\sigma'''}(\mathbf{k}_2)\delta_{\mathbf{k}-\mathbf{k}_1}\delta_{\mathbf{k}_2+\mathbf{k}_3} + S_{\sigma'''\sigma}(\mathbf{k}_3)S_{\sigma'\sigma''}(\mathbf{k}_1)\delta_{\mathbf{k}-\mathbf{k}_3}\delta_{\mathbf{k}_1+\mathbf{k}_2}$$
$$+ S_{\sigma''\sigma}(\mathbf{k}_2)S_{\sigma'\sigma'''}(\mathbf{k}_3)\delta_{\mathbf{k}-\mathbf{k}_2}\delta_{\mathbf{k}_1+\mathbf{k}_3}] \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3), \tag{31}$$

$$R_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) = \sqrt[5]{N_{\sigma'}N_{\sigma''}N_{\sigma'''}N_{\sigma^{(4)}}N_\sigma}\big[S_{\sigma'\sigma''\sigma}(\mathbf{k}_1,\mathbf{k}_2)S_{\sigma'''\sigma^{(4)}}(\mathbf{k}_3)\delta_{\mathbf{k}_3+\mathbf{k}_4} + S_{\sigma'\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_3)S_{\sigma''\sigma^{(4)}}(\mathbf{k}_2)\delta_{\mathbf{k}_2+\mathbf{k}_4}$$
$$+ S_{\sigma'\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_4)S_{\sigma''\sigma'''}(\mathbf{k}_2)\delta_{\mathbf{k}_2+\mathbf{k}_3} + S_{\sigma''\sigma'''\sigma}(\mathbf{k}_2,\mathbf{k}_3)S_{\sigma'\sigma^{(4)}}(\mathbf{k}_1)\delta_{\mathbf{k}_1+\mathbf{k}_4} + S_{\sigma''\sigma^{(4)}\sigma}(\mathbf{k}_2,\mathbf{k}_4)S_{\sigma'\sigma'''}(\mathbf{k}_1)\delta_{\mathbf{k}_1+\mathbf{k}_3}$$
$$+ S_{\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_3,\mathbf{k}_4)S_{\sigma'\sigma''}(\mathbf{k}_1)\delta_{\mathbf{k}_1+\mathbf{k}_2} + S_{\sigma'\sigma}(\mathbf{k}_1)S_{\sigma'''\sigma^{(4)}\sigma''}(\mathbf{k}_3,\mathbf{k}_4)\delta_{\mathbf{k}-\mathbf{k}_1} + S_{\sigma''\sigma}(\mathbf{k}_2)S_{\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_3,\mathbf{k}_4)\delta_{\mathbf{k}-\mathbf{k}_2}$$
$$+ S_{\sigma'''\sigma}(\mathbf{k}_3)S_{\sigma''\sigma^{(4)}\sigma'}(\mathbf{k}_2,\mathbf{k}_4)\delta_{\mathbf{k}-\mathbf{k}_3} + S_{\sigma^{(4)}\sigma}(\mathbf{k}_4)S_{\sigma''\sigma'''\sigma'}(\mathbf{k}_2,\mathbf{k}_3)\delta_{\mathbf{k}-\mathbf{k}_4}\big] \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4). \tag{32}$$

Note the expected invariance of (31) and (32) with respect to rotation on the $(p+1)$-sided polygons ($p = 3,4$) formed by corotating the wave vectors $\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p,\mathbf{k}$ and their conjugal species indices $\sigma',\sigma'',\ldots,\sigma^{(p)},\sigma$ in lock step. From FDTs (29) and (30), it then follows that the companion static external response functions exhibit the same rotation symmetries:

$$\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) = \hat{\chi}_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2) = \hat{\chi}_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1) = \hat{\chi}_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k}) \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3), \tag{33}$$

$$\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) = \hat{\chi}_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) = \hat{\chi}_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2) = \hat{\chi}_{\sigma'''\sigma^{(4)}\sigma\sigma'\sigma''}(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1)$$
$$= \hat{\chi}_{\sigma''\sigma'''\sigma^{(4)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k}) \quad (\mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4). \tag{34}$$

The partial cubic and quartic FDTs (29) and (30) with their respective rotation symmetries (33) and (34) are the expected multi-ionic generalizations of their OCP counterpart FDTs (18) and (19) with (22) and (23) reported in Ref. [1].

The permutation symmetries that the density response functions inherit from their companion structure factors through the static FDTs for binary Coulomb systems and for the OCP [1,2] suggest that the ubiquitous permutation symmetry that underlies the architectures of the static FDT hierarchy should also underlie the architectures of their companion dynamical frequency domain fluctuation-dissipation relations [1,2]. This is the main topic of the present work to be taken up in the next section.

Then contemplating the upcoming derivations of the corresponding higher-order dynamical FDTs, their counterpart static FDTs (29) and (30) can be made more tractable at a relatively modest cost by confining the present study to wave vectors whose partial sums are never allowed to equal zero:

$$\mathbf{k}_i + \mathbf{k}_j \neq \mathbf{0}; \quad i \neq j; \quad i,j = 1,2,3 \ \ \text{(cubic)},$$
$$\mathbf{k}_i + \mathbf{k}_j \neq \mathbf{0}; \quad i \neq j; \quad i,j = 1,2,3,4 \ \ \text{(quartic)},$$
$$\mathbf{k}_i + \mathbf{k}_j + \mathbf{k}_\ell \neq \mathbf{0}; \quad i \neq j,l; j \neq l; \quad i,j,l = 1,2,3,4 \ \ \text{(quartic)}. \tag{35}$$

The exclusions (35) in no way compromise the rotational invariance symmetries (33) and (34). The resulting partial FDTs,

$$\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) = -\frac{\beta^3}{3!}\sqrt[4]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma 0}}S_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) \quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3), \tag{36}$$

$$\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) = \frac{\beta^4}{4!}\sqrt[5]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma^{(4)}0}n_{\sigma 0}}S_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) \quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4), \tag{37}$$

remain exact since the exclusions (35), of course, apply to both sides of each equation.

This concludes the derivations of the static partial FDTs and concomitant rotational symmetry rules for the binary ionic mixture plasmas.

IV. FREQUENCY DOMAIN FLUCTUATION-DISSIPATION RELATIONS

We turn now to the derivations of the cubic ($p=3$) and quartic ($p=4$) frequency domain dynamical FDTs, each in an especially tractable form that features the connection between a single $(p+1)$-point dynamical structure function,

$$\begin{aligned}&\sqrt[p+1]{N_{\sigma'}N_{\sigma''}N_{\sigma'''}\cdots N_{\sigma^{(p)}}N_{\sigma}}\,S_{\sigma'\sigma''\sigma'''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\ldots,\omega_p)2\pi\,\delta_{\mathbf{k}_1+\mathbf{k}_2+\cdots+\mathbf{k}_p-\mathbf{k}}\delta(\omega_1+\omega_2+\cdots+\omega_p-\omega)\\ &\quad=\langle n_{\sigma'}(\mathbf{k}_1,\omega_1)n_{\sigma''}(\mathbf{k}_2,\omega_2)\cdots n_{\sigma^{(p)}}(\mathbf{k}_p,\omega_p)n_\sigma(-\mathbf{k},-\omega)\rangle^{(0)} \quad (\mathbf{k}_1\neq\mathbf{0},\mathbf{k}_2\neq\mathbf{0},\ldots,\mathbf{k}_p\neq\mathbf{0}),\end{aligned} \tag{38}$$

and a rotation-invariant combination of pth-order density response function terms. The partial density response functions are defined through the constitutive relation

$$\begin{aligned}\langle n_\sigma(\mathbf{k},\omega)\rangle^{(p)} &= \frac{1}{(2\pi V)^{p-1}}\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\cdots\sum_{\mathbf{k}_p}\int_{-\infty}^{\infty}d\omega_1\int_{-\infty}^{\infty}d\omega_2\cdots\int_{-\infty}^{\infty}d\omega_p\,\hat{\chi}_{\sigma'\sigma''\sigma'''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p;\ \omega_1,\omega_2,\ldots,\omega_p)\\ &\quad\times\hat{U}^{\sigma'}(\mathbf{k}_1,\omega_1)\hat{U}^{\sigma''}(\mathbf{k}_2,\omega_2)\cdots\hat{U}^{\sigma^{(p)}}(\mathbf{k}_p,\omega_p)\delta_{\mathbf{k}_1+\mathbf{k}_2+\cdots+\mathbf{k}_p-\mathbf{k}}\delta(\omega_1+\omega_2+\cdots+\omega_p-\omega),\end{aligned} \tag{39}$$

The architectures of the cubic and quartic FDTs will be based on the ubiquitous rotational symmetry requirement that emerges in the derivations of the linear ($p=1$) and quadratic ($p=2$) frequency domain FDTs,

$$\mathrm{Im}\,i^0\left[\frac{\hat{\chi}_{\sigma'\sigma}(\mathbf{k}_1,\omega_1)}{\omega_1}-\frac{\hat{\chi}_{\sigma\sigma'}(-\mathbf{k},-\omega)}{\omega}\right] = -\beta\sqrt{n_{\sigma 0}n_{\sigma'0}}S_{\sigma'\sigma}(\mathbf{k}_1,\omega_1) \quad (\mathbf{k}=\mathbf{k}_1;\omega=\omega_1), \tag{40}$$

$$\begin{aligned}&\mathrm{Im}\,i\left[\frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{k}_1,\mathbf{k}_2;\omega_1,\omega_2)}{\omega_1\omega_2}-\frac{\hat{\chi}_{\sigma\sigma'\sigma''}(-\mathbf{k},\mathbf{k}_1;-\omega,\omega_1)}{\omega\omega_1}-\frac{\hat{\chi}_{\sigma''\sigma\sigma'}(\mathbf{k}_2,-\mathbf{k};\omega_2,-\omega)}{\omega_2\omega}\right]\\ &\quad=-\frac{\beta^2}{2!2}\sqrt[3]{n_{\sigma'0}n_{\sigma''0}n_{\sigma 0}}S_{\sigma'\sigma''\sigma}(\mathbf{k}_1,\mathbf{k}_2;\omega_1,\omega_2) \quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2,\ \omega=\omega_1+\omega_2),\end{aligned} \tag{41}$$

established quite some time ago [3–5,16,17] using the conventional perturbation-theoretic approach that begins with the development of the time-dependent Liouville density in powers of the perturbing scalar potential. We observe that the linear FDT (40) links a single two-point dynamical structure factor to a linear combination of linear external response function terms; the quadratic FDT (41) links a single three-point dynamical structure factor to a linear combination of quadratic external response function terms. The linear architecture remains invariant under (σ'; $\mathbf{k}_1,\omega_1 \leftrightarrow \sigma$; $-\mathbf{k},-\omega$) simultaneous interchange of its 4-vector–species arguments. Similarly, the quadratic architecture remains invariant with respect to rotation on the triangle formed by the 4-vectors $(\mathbf{k}_1,\omega_1),(\mathbf{k}_2,\omega_2),(\mathbf{k},\omega)$; the species indices corotate in lock step. It therefore seems reasonable to expect that, with the conditions (35) imposed on the wave-vector partial sums, each successive higher-order equation in the family of FDTs can be cast in a similar form that connects a single $(p+1)$-point dynamical structure factor to a rotation-symmetric combination of pth-order external density response function terms.

The above discussion, especially its emphasis on the centrality of the rotational invariance symmetry and the guidance provided by the rigorously established linear and quadratic FDTs [3–5,16,17,29], suggests that the frequency domain cubic and quartic dynamical FDTs are well represented by the architectures,

$$\begin{aligned}&\mathrm{Im}\,i^2\left[\frac{\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)}{\omega_1\omega_2\omega_3}-\frac{\hat{\chi}_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;-\omega,\omega_1\omega_2)}{\omega\omega_1\omega_2}\right.\\ &\quad\left.-\frac{\hat{\chi}_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\omega_3,-\omega,\omega_1)}{\omega_3\omega\omega_1}-\frac{\hat{\chi}_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega)}{\omega_2\omega_3\omega}\right]\\ &\quad=-K_3S_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3) \quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3;\ \omega=\omega_1+\omega_2+\omega_3),\end{aligned} \tag{42}$$

$$\mathrm{Im} i^3\Bigg[\frac{\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)}{\omega_1\omega_2\omega_3\omega_4}-\frac{\hat{\chi}_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega,\omega_1,\omega_2,\omega_3)}{\omega\omega_1\omega_2\omega_3}$$
$$-\frac{\hat{\chi}_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)}{\omega_4\omega\omega_1\omega_2}-\frac{\hat{\chi}_{\sigma'''\sigma^{(4)}\sigma\sigma'\sigma''}(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)}{\omega_3\omega_4\omega\omega_1}$$
$$-\frac{\hat{\chi}_{\sigma''\sigma'''\sigma^{(4)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)}{\omega_2\omega_3\omega_4\omega}\Bigg]$$
$$=-K_4S_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)\quad(\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4;\omega=\omega_1+\omega_2+\omega_3+\omega_4). \tag{43}$$

The proportionality constants K_4 and K_5 are to be determined by evaluating (42) and (43) in their static limits and matching the results to their respective static FDT counterpart equations (36) and (37). The rather intricate passage to the static limits of (42) and (43) begins with the conversion of their right-hand-side dynamical structure functions into static (equal-time) structure functions via

$$S_{\sigma'\sigma''\sigma'''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p)\equiv S_{\sigma'\sigma''\sigma'''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p;t_1=0,t_2=0,\ldots,t_p=0)$$
$$=\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\cdots\int_{-\infty}^{\infty}\frac{d\omega_p}{2\pi}S_{\sigma'\sigma''\sigma'''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\ldots,\omega_p). \tag{44}$$

Cubic fluctuation-dissipation relation. Addressing first the proposed frequency domain cubic FDT equation (42), our task consists in evaluating the integrals,

$$I=\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_{3,};\omega_1,\omega_2,\omega_3)}{\omega_1\omega_2\omega_3}$$
$$-\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_{2,};-\omega,\omega_1,\omega_2)}{\omega\omega_1\omega_2}$$
$$-\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_{1,};\omega_3,-\omega,\omega_1)}{\omega_3\omega\omega_1}$$
$$-\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega)}{\omega_2\omega_3\omega}$$
$$=K_3S_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) \tag{45}$$

(single and double prime notations denote real and imaginary parts, respectively). Before embarking on those calculations, however, there is the question of the boundedness of the left-hand-side of (42) in the $\omega_1=\omega_2=\omega_3=0$ limit,

$$\lim_{\omega_1\to0}\lim_{\omega_2\to0}\lim_{\omega_3\to0}\Bigg[\frac{\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)}{\omega_1\omega_2\omega_3}-\frac{\hat{\chi}''_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;-\omega,\omega_1,\omega_2)}{\omega\omega_1\omega_2}$$
$$-\frac{\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\omega_3,-\omega,\omega_1)}{\omega_3\omega\omega_1}-\frac{\hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}\big(\mathbf{k}_2,\mathbf{k}_{3,}-\mathbf{k};\omega_2,\omega_3,-\omega\big)}{\omega_2\omega_3\omega}\Bigg], \tag{46}$$

to be addressed. We will see that it is most likely the intervention of a low-frequency rotational symmetry requirement that rules out the occurrence of isolated singular behavior at the null point $\omega_1=\omega_2=\omega_3=0$ which would otherwise compromise the viability of (42). In that limit, one certainly expects the combination of response function terms in (42) to be bounded, consistent with the guarantee that its right-hand-side zero-frequency structure factor $S_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3:\omega_1=0,\omega_2=0,\omega_3=0)$ is bounded on physical grounds.

In compliance with the reality condition and invariance under spatial reflection,

$$\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)=-\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega_1,-\omega_2,-\omega_3), \tag{47}$$

only terms having net odd frequency parity are allowed in the multivariable expansion about $\omega_1=\omega_2=\omega_3=0$, viz.,

$$\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1\to0,\omega_2\to0,\omega_3\to0)$$
$$=[a_1\omega_1+a_2\omega_2+a_3\omega_3]+\big[b_1\omega_1^3+b_2\omega_2^3+b_3\omega_3^3+b_4\omega_1\omega_2\omega_3+b_5\omega_1\omega_2^2+b_6\omega_1\omega_3^2+b_7\omega_1^2\omega_2+b_8\omega_1^2\omega_3+\omega_2^2\omega_3\big]+\cdots. \tag{48}$$

Now, the question of boundedness would never arise in a scenario where all three linear Taylor coefficients turn out to be identically zero, in which case the expansion (48) would begin with third-order terms, resulting in harmless removable singularities in (46).

In the far more likely scenario and the one I presume prevails here, the issue of isolated singular behavior at $\omega_1 = \omega_2 = \omega_3 = 0$ is entirely circumvented by imposing the low-frequency rotation-symmetry requirement:

$$\begin{aligned}\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1\to 0,\omega_2\to 0,\omega_3\to 0) &= \hat{\chi}''_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;-\omega\to 0,\omega_1\to 0,\omega_2\to 0)\\ &= \hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\omega_3\to 0,-\omega\to 0,\omega_1\to 0)\\ &= \hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2\to 0,\omega_3\to 0,-\omega\to 0),\end{aligned} \tag{49}$$

whence (46) contracts to a form that vanishes identically:

$$\lim_{\omega_1\to 0}\lim_{\omega_2\to 0}\lim_{\omega_3\to 0}\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)\lim_{\omega_1\to 0}\lim_{\omega_2\to 0}\lim_{\omega_3\to 0}\left[\frac{1}{\omega_1\omega_2\omega_3}-\frac{1}{\omega\omega_1\omega_2}-\frac{1}{\omega_3\omega\omega_1}-\frac{1}{\omega_2\omega_3\omega}\right]\equiv 0. \tag{50}$$

With the boundedness of the cubic FDT (42) assured by the intervention of (49), one may assume that the Eq. (45) combination of integrals involving causal response functions can be reformulated into a combination of Cauchy principal value integrals, amenable to Hilbert transform operations:

$$\begin{aligned}I = {} & PPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_{3,};\omega_1,\omega_2,\omega_3)}{\omega_1\omega_2\omega_3}\\ &-PPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_{2,};-\omega,\omega_1,\omega_2)}{\omega\omega_1\omega_2}\\ &-PPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_{1,};\omega_3,-\omega,\omega_1)}{\omega_3\omega\omega_1}\\ &-PPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\frac{\hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega)}{\omega_2\omega_3\omega}.\end{aligned} \tag{51}$$

With no rotation operations contemplated in the mathematical steps that follow, the response functions will sometimes be displayed, for the sake of brevity, without showing their species subscripts until the very end of each series of steps where the subscripts are restored.

By repeated Hilbert transform operations, one obtains for the first two right-hand-side integrals:

$$\begin{aligned}I_1 &= \int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}P\frac{1}{\omega_1}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}P\frac{1}{\omega_2}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}P\frac{1}{\omega_3}\hat{\chi}''_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)\\ &= -\frac{1}{8}\hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1=0,\omega_2=0,\omega_3=0)\equiv -\frac{1}{8}\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3),\end{aligned} \tag{52}$$

$$\begin{aligned}I_2 &= -\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}P\frac{1}{\omega_1}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}P\frac{1}{\omega_2}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}P\frac{1}{\omega}\hat{\chi}''_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;-\omega,\omega_1,\omega_2)\\ &= -\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}P\frac{1}{\omega_1}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}P\frac{1}{\omega_2}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}P\frac{1}{\omega_1+\omega_2+\omega_3}\hat{\chi}''(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;-\omega_1-\omega_2-\omega_3,\omega_1,\omega_2)\\ &= \int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}P\frac{1}{\omega_1}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}P\frac{1}{\omega_2}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}P\frac{1}{\bar{\omega}_3}\hat{\chi}''(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\bar{\omega}_3,\omega_1,\omega_2)\\ &= -\frac{1}{8}\hat{\chi}'_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega=0,\omega_1=0,\omega_2=0)\equiv -\frac{1}{8}\hat{\chi}_{\sigma\sigma'\sigma''\sigma'''}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2),\end{aligned} \tag{53}$$

where the single prime notation [in the last line of Eq. (53)] denotes the real part. Note, however, that the third and fourth integrals of (51), as they now stand, are not amenable to similar simplification. This can be remedied by first invoking the Poincaré-Bertrand theorem [16,17,32,33] suitably generalized to handle the multiple integrals of the present work. The calculation then proceeds as follows:

$$\begin{aligned}I_3 &= -\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\omega_3,-\omega,\omega_1)PPP\frac{1}{\omega_3\omega\omega_1}\\ &= -\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\hat{\chi}''(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\omega_3,-\omega_1-\omega_2-\omega_3,\omega_1)PPP\frac{1}{\omega_3(\omega_1+\omega_2+\omega_3)\omega_1}.\end{aligned}$$

Now make the change of variables $\nu = \omega_3, d\nu = d\omega_3, \bar{\mu} = -\omega_1 - \omega_2, d\bar{\mu} = -d\omega_2$ so that

$$I_3 = \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\bar{\mu}}{2\pi} \int_{-\infty}^{\infty} \frac{d\nu}{2\pi} \hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\nu,\bar{\mu}-\nu,\omega_1) P P\frac{1}{\nu(\bar{\mu}-\nu)}$$
$$= \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1}\left[\int_{-\infty}^{\infty} \frac{d\nu}{2\pi} P\frac{1}{\nu} \int_{-\infty}^{\infty} \frac{d\bar{\mu}}{2\pi} \hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\nu,\bar{\mu}-\nu,\omega_1) P\frac{1}{\bar{\mu}-\nu} + \frac{1}{4}\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;0,0,\omega_1)\right],$$

in virtue of the Poincaré-Bertrand theorem. Subsequent Hilbert transform operations then provide

$$I_3 = \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1}\left[-\frac{1}{4}\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;0,0,\omega_1) + \frac{1}{4}\hat{\chi}''_{\sigma'''\sigma\sigma'\sigma''}(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;0,0,\omega_1)\right] = 0. \tag{54}$$

The evaluation of the remaining integral requires a somewhat different treatment:

$$I_4 = -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega) P P\frac{1}{\omega_3\omega}$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega) P P\frac{1}{\omega_2\omega}$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_{3,},-\omega) P P\frac{1}{\omega_2\omega}$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3}\left[\int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega) P\frac{1}{\omega}\right.$$
$$\left. -\frac{1}{4}\hat{\chi}''_{\sigma''\sigma'''\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};0,\omega_3,0)\right] = 0. \tag{55}$$

The last two lines of (55) again follow from the application of a somewhat more generalized form of the Poincaré-Bertrand theorem tailored by the author to this particular calculation. They have been brought into a form that allows for the application of the Kramers-Kronig relation resulting in the cancellation effect similar to the one in (54). Summing (52)–(55) and exploiting the rotation-symmetry rule (33), one obtains

$$K_3 S_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;0,0,0) = I = I_1 + I_2 + I_3 + I_4 = -\tfrac{1}{4}\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;0,0,0), \tag{56}$$

whence

$$K_3 = \beta^3 \sqrt[4]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma 0}}/24 = \beta^3 \sqrt[4]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma 0}}/(3!2^2) \tag{57}$$

follows from matching (56) to (36). This completes the formulation of the frequency domain cubic fluctuation-dissipation relation.

Quartic fluctuation-dissipation relation. Addressing next the proposed frequency domain quartic FDT (43), we wish to evaluate the integrals:

$$J = \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)}{\omega_1\omega_2\omega_3\omega_4}$$
$$- \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega,\omega_1,\omega_2,\omega_3)}{\omega\omega_1\omega_2\omega_3}$$
$$- \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)}{\omega_4\omega\omega_1\omega_2}$$
$$- \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma'''\sigma^{(4)}\sigma\sigma'\sigma''}(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)}{\omega_3\omega_4\omega\omega_1}$$
$$- \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma''\sigma'''\sigma^{(4)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)}{\omega_2\omega_3\omega_4\omega}$$
$$= K_4 S_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;t=0,t=0,t=0,t=0). \tag{58}$$

To set the stage for this somewhat more involved calculation, I begin as before by first showing that the occurrence of isolated singular behavior in (43) at $\omega_1 = \omega_2 = \omega_3 = \omega_4 = 0$ is ruled out by the intervention of the static quartic rotational symmetry rule (34); it is never an issue. Indeed, one expects that the combination of response functions in (43) will be bounded, consistent with the bounded behavior, guaranteed on physical grounds, of the right-hand-side five-point structure function in the dc (zero-frequency) limit. This is borne out by the following analysis.

In compliance with the reality condition and invariance under spatial reflection,

$$\hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4) = \hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;-\omega_1,-\omega_2,-\omega_3,-\omega_4), \tag{59}$$

only terms having net even frequency parity are allowed in the multivariable expansion about the null point $\omega_1 = \omega_1 = \omega_3 = \omega_4 = 0$. Accordingly, the expansions through quadratic order are

$$\begin{aligned}
&\hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1\to 0,\omega_2\to 0,\omega_3\to 0,\omega_4\to 0)\\
&= \hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;0,0,0,0) + \big[a_{11}\omega_1^2 + a_{22}\omega_2^2 + a_{33}\omega_3^2 + a_{44}\omega_4^2\\
&\quad + a_{12}\omega_1\omega_2 + a_{13}\omega_1\omega_3 + a_{14}\omega_1\omega_4 + a_{23}\omega_2\omega_3 + a_{24}\omega_2\omega_4 + a_{34}\omega_3\omega_4\big] + \cdots, \qquad (60a)\\
&\hat{\chi}'_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega\to 0,\omega_1\to 0,\omega_2\to 0,\omega_3\to 0)\\
&= \hat{\chi}'_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;0,0,0,0) + \big[b_{00}\omega^2 + b_{11}\omega_1^2 + b_{22}\omega_2^2 + b_{33}\omega_3^2\\
&\quad - b_{01}\omega\omega_1 - b_{02}\omega\omega_2 - b_{03}\omega\omega_3 + b_{12}\omega_1\omega_2 + b_{13}\omega_1\omega_3 + b_{23}\omega_2\omega_3\big] + \cdots, \qquad (60b)\\
&\hat{\chi}'_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4\to 0,-\omega\to 0,\omega_1\to 0,\omega_2\to 0)\\
&= \hat{\chi}'_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;0,0,0,0) + \big[c_{44}\omega_4^2 + c_{00}\omega^2 + c_{11}\omega_1^2 + c_{22}\omega_2^2\\
&\quad - c_{40}\omega_4\omega + c_{41}\omega_4\omega_1 + c_{42}\omega_4\omega_2 - c_{01}\omega\omega_1 - c_{02}\omega\omega_2 + c_{12}\omega_1\omega_2\big] + \cdots, \qquad (60c)
\end{aligned}$$

etc.

The first right-hand-side (rhs) members of Eqs. (60a)–(60c) are identified as the members of the zero-frequency rotation-invariant symmetry rule (34); the second rhs members in square brackets represent all the $O(\omega_i\omega_j)$ quadratic contributions to the expansions; the fourth-order contributions are not displayed for brevity. In evaluating (43) in the zero-frequency limit, only the surviving zeroth-order term in each of the above Taylor expansions really matters, that is,

$$\begin{aligned}
&\lim_{\omega_1\to 0}\lim_{\omega_2\to 0}\lim_{\omega_3\to 0\infty}\lim_{\omega_4\to 0}\Bigg[\frac{\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)}{\omega_1\omega_2\omega_3\omega_4}\\
&\quad - \frac{\hat{\chi}_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega,\omega_1,\omega_2,\omega_3)}{\omega\omega_1\omega_2\omega_3} - \frac{\hat{\chi}_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)}{\omega_4\omega\omega_1\omega_2}\\
&\quad - \frac{\hat{\chi}_{\sigma'''\sigma^{(4)}\sigma\sigma'\sigma''}(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4-\omega,\omega_1)}{\omega_3\omega_4\omega\omega_1} - \frac{\hat{\chi}_{\sigma''\sigma'''\sigma^{(4)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)}{\omega_2\omega_3\omega_4\omega}\Bigg]\\
&= \lim_{\omega_1\to 0}\lim_{\omega_2\to 0}\lim_{\omega_3\to 0}\lim_{\omega_4\to 0}\Bigg[\frac{\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;0,0,0,0)}{\omega_1\omega_2\omega_3\omega_4}\\
&\quad - \frac{\hat{\chi}_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;0,0,0,0)}{\omega\omega_1\omega_2\omega_3} - \frac{\hat{\chi}_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;0,0,0,0)}{\omega_4\omega\omega_1\omega_2}\\
&\quad - \frac{\hat{\chi}_{\sigma'''\sigma^{(4)}\sigma\sigma'\sigma''}(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;0,0,0,0)}{\omega_3\omega_4\omega\omega_1} - \frac{\hat{\chi}_{\sigma''\sigma'''\sigma^{(4)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};0,0,0,0)}{\omega_2\omega_3\omega_4\omega}\Bigg]\\
&= \hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;0,0,0,0)\\
&\quad \times \lim_{\omega_1\to 0}\lim_{\omega_2\to 0}\lim_{\omega_3\to 0}\lim_{\omega_4\to 0}\left[\frac{1}{\omega_1\omega_2\omega_3\omega_4} - \frac{1}{\omega\omega_1\omega_2\omega_3} - \frac{1}{\omega_4\omega\omega_1\omega_2} - \frac{1}{\omega_3\omega_4\omega\omega_1} - \frac{1}{\omega_2\omega_3\omega_4\omega}\right] \equiv 0, \qquad (61)
\end{aligned}$$

in virtue of the rotation-symmetry rule (34) and invariance of the stationary system under temporal translation ($\omega = \omega_1 + \omega_2 + \omega_3 + \omega_4$). Thus, the intervention of (34) rules out the possibility of occurrence of isolated singular behavior at $\omega_1 = \omega_2 = \omega_3 = \omega_4 = 0$.

This allows one to recast the Eq. (58) combination of integrals involving causal response functions into the following combination of Cauchy principal part integrals amenable to Hilbert transform operations:

$$\begin{aligned}
J &= PPPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_4}{2\pi}\,\frac{\hat{\chi}'_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)}{\omega_1\omega_2\omega_3\omega_4}\\
&\quad - PPPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_4}{2\pi}\,\frac{\hat{\chi}'_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega,\omega_1,\omega_2,\omega_3)}{\omega\omega_1\omega_2\omega_3}\\
&\quad - PPPP\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_3}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_4}{2\pi}\,\frac{\hat{\chi}'_{\sigma^{(4)}\sigma\sigma'\sigma''\sigma'''}(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)}{\omega_4\omega\omega_1\omega_2}
\end{aligned}$$

$$- PPPP \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma'''\sigma^{(4)}\sigma\sigma'\sigma''}(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)}{\omega_3\omega_4\omega\omega_1}$$
$$- PPPP \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} \frac{\hat{\chi}'_{\sigma''\sigma'''\sigma^{(4)}\sigma\sigma'}(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)}{\omega_2\omega_3\omega_4\omega}. \quad (62)$$

For brevity, species indices are sometimes omitted. By repeated applications of Hilbert transform operations, one obtains the following for the first two right-hand-side integrals:

$$J_1 = \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} P\frac{1}{\omega_4} \hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)$$
$$= \frac{1}{16}\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;0,0,0,0), \quad (63)$$

$$J_2 = -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} P\frac{1}{\omega} \hat{\chi}'(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega,\omega_1,\omega_2,\omega_3)$$
$$= \frac{1}{16}\hat{\chi}_{\sigma\sigma'\sigma''\sigma'''\sigma^{(4)}}(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;0,0,0,0). \quad (64)$$

The evaluations of the remaining three integrals call for the intervention of the Poincaré-Bertrand theorem in order to bring each into a form that will be amenable to Hilbert transform operations:

$$J_3 = -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} PP\frac{1}{\omega_4\omega} \hat{\chi}'(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \left[\int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} P\frac{1}{\omega_4} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega} \hat{\chi}'(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)\right.$$
$$\left. - \frac{1}{4}\hat{\chi}'(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;0,0,\omega_1,\omega_2)\right] = 0, \quad (65)$$

$$J_4 = -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} PP\frac{1}{\omega_4\omega} \hat{\chi}'(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} PP\frac{1}{\omega_4\omega} \hat{\chi}'(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega_1} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \left[\int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} P\frac{1}{\omega_4} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega} \hat{\chi}'(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)\right.$$
$$\left. - \frac{1}{4}\hat{\chi}'(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,0,0,\omega_1)\right] = 0, \quad (66)$$

$$J_5 = -\int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} PP\frac{1}{\omega_4\omega} \hat{\chi}'(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} PP\frac{1}{\omega_4\omega} \hat{\chi}'(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} PP\frac{1}{\omega_4\omega} \hat{\chi}'(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)$$
$$= -\int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} P\frac{1}{\omega_2} \int_{-\infty}^{\infty} \frac{d\omega_3}{2\pi} P\frac{1}{\omega_3} \left[\int_{-\infty}^{\infty} \frac{d\omega_4}{2\pi} P\frac{1}{\omega_4} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} P\frac{1}{\omega} \hat{\chi}'(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)\right.$$
$$\left. - \frac{1}{4}\hat{\chi}'(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,0,0)\right] = 0. \quad (67)$$

Summing contributions (63)–(67) and invoking the rotation-symmetry rule (34), one readily obtains

$$K_4 S_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) = J = J_1 + J_2 + J_3 + J_4 + J_5 = \tfrac{1}{8}\hat{\chi}_{\sigma'\sigma''\sigma'''\sigma^{(4)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4), \quad (68)$$

whence

$$K_4 = \beta^4 \sqrt[5]{n_{\sigma'0}n_{\sigma''0}n_{\sigma'''0}n_{\sigma^{(4)}0}n_{\sigma0}}/(4!2^3) \tag{69}$$

follows from matching (68) with (37). This completes the formulation of the frequency domain quartic fluctuation-dissipation theorem.

Combined fluctuation-dissipation relations. It is especially instructive to derive the fluctuation-dissipation relations for the spectral correlations of *combined* microscopic charge densities:

$$P(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\omega_3,\ldots,\omega_p) = e^{p+1}\sum_\sigma\sum_{\sigma'}\sum_{\sigma''}\ldots\sum_{\sigma^{(p)}} Z^\sigma Z^{\sigma'}Z^{\sigma''}\cdots Z^{(p)}\sqrt[p+1]{n_{\sigma0}n_{\sigma'0}n_{\sigma''0}\cdots n_{\sigma^{(p)}0}}$$
$$\times S_{\sigma\sigma'\sigma''\cdots\sigma^{(p)}}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\omega_3,\ldots,\omega_p), \tag{70}$$

where $p = 1,2,3,4,\cdots$.

Here the spectral correlations of combined microscopic charge densities quite naturally partner with external polarizability response functions, $\hat{\alpha}'s$, defined through the constitutive relations

$$\langle\rho(\mathbf{k},\omega)\rangle^{(p)} = \sum_{\sigma=A,B} Z^\sigma e\langle n_\sigma(\mathbf{k},\omega)\rangle^{(p)}$$
$$= \frac{(-i)^{p+1}}{4\pi}\frac{1}{(2\pi V)^{p-1}}\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\sum_{\mathbf{k}_3}\cdots\sum_{\mathbf{k}_p} kk_1k_2k_3\cdots k_p\int_{-\infty}^{\infty}d\omega_1\int_{-\infty}^{\infty}d\omega_2\int_{-\infty}^{\infty}d\omega_3\cdots\int_{-\infty}^{\infty}d\omega_p$$
$$\times\hat{\alpha}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\omega_3,\ldots,\omega_p)\hat{\phi}(\mathbf{k}_1,\omega_1)\hat{\phi}(\mathbf{k}_2,\omega_2)\hat{\phi}(\mathbf{k}_3,\omega_3)\cdots\hat{\phi}(\mathbf{k}_p,\omega_p)$$
$$\times\delta_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\cdots+\mathbf{k}_p-\mathbf{k}}\delta(\omega_1+\omega_2+\omega_3+\cdots+\omega_p-\omega)\quad (p=1,2,3,4,\ldots). \tag{71}$$

On the other hand, constitutive relation (39) provides

$$\langle\rho(\mathbf{k},\omega)\rangle^{(p)} = \sum_{\sigma=A,B} Z^\sigma e\langle n_\sigma(\mathbf{k},\omega)\rangle^{(p)}$$
$$= \frac{1}{(2\pi V)^{p-1}}\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\sum_{\mathbf{k}_3}\cdots\sum_{\mathbf{k}_p}\int_{-\infty}^{\infty}d\omega_1\int_{-\infty}^{\infty}d\omega_2\int_{-\infty}^{\infty}d\omega_3\cdots\int_{-\infty}^{\infty}d\omega_p\hat{\phi}(\mathbf{k}_1,\omega_1)\hat{\phi}(\mathbf{k}_2,\omega_2)\cdots\hat{\phi}(\mathbf{k}_p,\omega_p)$$
$$\times\delta_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\cdots+\mathbf{k}_p-\mathbf{k}}\delta(\omega_1+\omega_2+\omega_3+\cdots+\omega_p-\omega)\sum_\sigma\sum_{\sigma'}\sum_{\sigma''}\cdots\sum_{\sigma^{(p)}}Z^\sigma Z^{\sigma'}Z^{\sigma''}\cdots Z^{\sigma^{(p)}}e^{p+1}$$
$$\times\hat{\chi}_{\sigma\sigma'\sigma''\cdots\sigma^{(p)}}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\omega_3,\ldots,\omega_p)\quad (p=1,2,3,4,\ldots), \tag{72}$$

in a form that reconciles with (71) by also featuring the potential $\hat{\phi}$ as the external agency which simultaneously drives both species. Comparing (72) with (71), one obtains

$$\hat{\alpha}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\omega_3,\ldots,\omega_p)$$
$$= \frac{4\pi(ie)^{p+1}}{kk_1k_2\cdots k_p}\sum_\sigma\sum_{\sigma'}\sum_{\sigma''}\cdots\sum_{\sigma^{(p)}}Z^\sigma Z^{\sigma'}Z^{\sigma''}\cdots Z^{\sigma^{(p)}}\hat{\chi}_{\sigma\sigma'\sigma''\cdots\sigma^{(p)}}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\omega_3,\ldots,\omega_p). \tag{73}$$

Equation (73) dictates the charge species summation protocol for generating the pth-order external polarizability from the external density response functions. The protocol, along with Eq. (70), when applied to FDTs (40)–(43), generates the following first four equations in the hierarchy of FDTs, each linking a rotationally invariant combination of polarizabilities to a single spectral correlation of combined microscopic charge densities:

$$\frac{\hat{\alpha}''(\mathbf{k}_1;\omega_1)}{\omega_1} - \frac{\hat{\alpha}''(-\mathbf{k},-\omega)}{\omega} = \frac{4\pi\beta}{kk_1}P(\mathbf{k}_1;\omega_1)\quad (\mathbf{k}=\mathbf{k}_1,\omega=\omega_1), \tag{74}$$

$$\frac{\hat{\alpha}''(\mathbf{k}_1,\mathbf{k}_2;\omega_1,\omega_2)}{\omega_1\omega_2} - \frac{\hat{\alpha}''(-\mathbf{k},\mathbf{k}_1;-\omega,\omega_1)}{\omega\omega_1} - \frac{\hat{\alpha}''(\mathbf{k}_2,-\mathbf{k};\omega_2,-\omega)}{\omega_2\omega} = \frac{4\pi}{kk_1k_2}\frac{\beta^2}{2!2}P(\mathbf{k}_1,\mathbf{k}_2;\omega_1,\omega_2)\quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2;\omega=\omega_1+\omega_2), \tag{75}$$

$$\frac{\hat{\alpha}''(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)}{\omega_1\omega_2\omega_3} - \frac{\hat{\alpha}''(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;-\omega,\omega_1,\omega_2)}{\omega\omega_1\omega_2} - \frac{\hat{\alpha}''(\mathbf{k}_3,-\mathbf{k},\mathbf{k}_1;\omega_3,-\omega,\omega_1)}{\omega_3\omega\omega_1} - \frac{\hat{\alpha}''(\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k};\omega_2,\omega_3,-\omega)}{\omega_2\omega_3\omega}$$
$$= \frac{4\pi}{kk_1k_2k_3}\frac{\beta^3}{3!2^2}P(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;\omega_1,\omega_2,\omega_3)\quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3;\omega=\omega_1+\omega_2+\omega_3), \tag{76}$$

$$\frac{\hat{\alpha}''(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4)}{\omega_1\omega_2\omega_3\omega_4} - \frac{\hat{\alpha}''(-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3;-\omega,\omega_1,\omega_2,\omega_3)}{\omega\omega_1\omega_2\omega_3} - \frac{\hat{\alpha}''(\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1,\mathbf{k}_2;\omega_4,-\omega,\omega_1,\omega_2)}{\omega_4\omega\omega_1\omega_2}$$
$$- \frac{\hat{\alpha}''(\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k},\mathbf{k}_1;\omega_3,\omega_4,-\omega,\omega_1)}{\omega_3\omega_4\omega\omega_1} - \frac{\hat{\alpha}''(\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,-\mathbf{k};\omega_2,\omega_3,\omega_4,-\omega)}{\omega_2\omega_3\omega_4\omega}$$
$$= \frac{4\pi}{kk_1k_2k_3k_4}\frac{\beta^4}{4!2^3}P(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;\omega_1,\omega_2,\omega_3,\omega_4) \quad (\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4;\omega=\omega_1+\omega_2+\omega_3+\omega_4). \tag{77}$$

Linear FDT (74) is, of course, well known for the OCP, and here is displayed in rotation-symmetric form. The quadratic FDT (75) for the BIM was established in 1982 [17]; here we report cubic and quartic FDTs (76) and (77).

V. CONCLUDING REMARKS

In this paper tractable frequency domain cubic ($p = 3$) and quartic ($p = 4$) fluctuation-dissipation theorems (FDTs) have been established for magnetic field-free plasmas consisting of two or more ion species immersed in a uniform inert background of rigid degenerate electrons. Each FDT features a single $(p+1)$-point partial dynamical structure factor, entirely free of entangled Liouville space paths, linked to a rotation-invariant combination of pth-order partial density response functions. This latter architecture is dictated by the invariance of the dynamical structure factor under permutation of its $(p+1)$ microscopic particle densities, or equivalently, under rotation of its wave-vector-frequency–species arguments (referred to in the present work as "rotation-invariance symmetry").

In the course of evaluating the resulting frequency domain FDTs (42) and (43) in their static limits, repeated Hilbert transform operations and applications of the Poincaré-Bertrand theorem bring (42) and (43) into forms that can be precisely matched to their independently derived static counterpart equations (36) and (37). Thus, consistency is assured and one ultimately obtains

$$\mathrm{Im}\, i^{p-1}\left[\frac{\hat{\chi}_{\sigma'\sigma''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p;\omega_{11},\omega_2,\ldots,\omega_p)}{\omega_1\omega_2\cdots\omega_p} - \cdots - \frac{\hat{\chi}_{\sigma''\sigma'''\cdots\sigma^{(p)}\sigma\sigma'}(\mathbf{k}_2,\ldots,\mathbf{k}_p,-\mathbf{k};\omega_2,\ldots,\omega_p,-\omega)}{\omega_2\cdots\omega_p\omega}\right]$$
$$= -\frac{\beta^p}{p!2^{p-1}}\frac{1}{V}\sqrt[p+1]{N_{\sigma'}N_{\sigma''}\cdots N_{\sigma^{(p)}}N_\sigma}\, S_{\sigma'\sigma''\cdots\sigma^{(p)}\sigma}(\mathbf{k}_1,\mathbf{k}_2,\ldots,\mathbf{k}_p;\omega_1,\omega_2,\ldots,\omega_p) \quad (p=1,2,3,4)$$
$$(\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\cdots+\mathbf{k}_p;\ \omega=\omega_1+\omega_2+\cdots+\omega_p). \tag{78}$$

The formulations of the static and frequency domain cubic and quartic fluctuation-dissipation relations (36), (37), and (76)–(78) are the main accomplishments of the present work. The alternative rotation-symmetry-based approach is the centerpiece.

With the FDTs presented here, we are now well positioned to generate a wealth of sum rules adding to the pioneering contributions of Vieillefosse [34] and Alastuey [35] at the static level and to the frequency moment sum rules for the quadratic response functions [36] at the dynamical level. Details are given in Ref. [1]. Such an enterprise, while outside the scope of the present work, could merit further exploration in the future.

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