Analytical properties of the quadratic density response and quadratic dynamical structure functions: Conservation sum rules and frequency moments

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The quadratic density response function and the quadratic dynamical structure function (the Fourier transform of the equilibrium three-point density correlations) contain important information about a many body system; they are also ingredients for an improved dynamical mean field theory for strongly coupled Fermi systems. We examine the analytic properties of the density response function and establish new single frequency and double frequency moment sum rules. We relate the sum rule coefficients to the high frequency expansion of the response function. Next we invoke the quadratic fluctuation-dissipation theorem to relate these frequency moments to weighted frequency moments of the dynamical structure function. These latter reduce to straight frequency moments in the high temperature classical and zero temperature degenerate limits. [S1063-651X(96)09010-1]

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I. INTRODUCTION

Sum rules for the linear response functions have played an important role throughout the development of electron-liquid theory. The knowledge of the compressibility sum rule and the ω^3 moment of the density-density response function $\chi(\mathbf{q},\omega)$ has led to much improved local field corrections. Less explored are possible sum rules relating to quadratic response functions. With a little reflection it is easy to realize that the fundamental physical effects that operate in the generation of the linear sum rules—namely, symmetry and conservation laws—must also be responsible for creating sum rules for quadratic response functions. Some of these sum rules for the quadratic density-density response function $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ have already been identified: Golden, Kalman, and Datta [1] have shown the existence of a static compressibility sum rule and established the high frequency behavior of $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$; more recently Tao and Kalman [2] have derived a frequency moment sum rule, analogous to the linear f-sum rule.

In this paper we will systematically establish and analyze a number of sum rules for the quadratic density-density response and the quadratic dynamical structure function. The precise definition of the former is the relation

$$\langle \varrho_{-\mathbf{q}_0}(-\omega_0)\rangle^{(2)}=\frac{1}{V}\sum_{\mathbf{q}_1,\mathbf{q}_2}\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\times\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)\Phi(\mathbf{q}_1,\omega_1)\Phi(\mathbf{q}_2,\omega_2)\times\delta\left(\sum\omega_i\right)\delta\left(\sum\mathbf{q}_i\right), \quad (1)$$

where the two Φ are external fields. For simplicity in writing we assume throughout this paper all $\mathbf{q}_i$ to be nonzero and that always $\omega_0+\omega_1+\omega_2=0$ as well as $\mathbf{q}_0+\mathbf{q}_1+\mathbf{q}_2=\mathbf{0}$. The superscript (2) denotes second order in the external fields.

$\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ is related to the quadratic dynamical structure function through the quadratic fluctuation-dissipation theorem [3]. Thus, somewhat similarly to the linear case, frequency moment sum rules for $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ entail frequency moment sum rules for the quadratic dynamical structure function. There are, however, deep structural differences between the linear and the quadratic fluctuation-dissipation theorems which prevent the analogy to be carried too far.

In analogy to the linear time-dependent two point function $S(\mathbf{q},t)=(1/N)\langle\varrho_{\mathbf{q}}(t)\varrho_{-\mathbf{q}}(0)\rangle^{(0)}$ the equilibrium dynamical three point function is defined as

$$S(\mathbf{q}_0,t_0;\mathbf{q}_1,t_1;\mathbf{q}_2,t_2)=\frac{1}{N}\langle\varrho_{\mathbf{q}_0}(t_0)\varrho_{\mathbf{q}_1}(t_1)\varrho_{\mathbf{q}_2}(t_2)\rangle^{(0)}. \quad (2)$$

While in the classical limit the density operators commute, their ordering is of obvious relevance in the quantum case. This is reflected in the chosen ordering of the arguments for the three point S. The superscript (0) indicates that the average is taken at equilibrium. Hence only the time differences between t_1, t_2, and t_3 matter and one can always shift the time so that one of the time arguments becomes zero. The time Fourier transform of Eq. (2) will be referred to as the quadratic dynamical structure function $S(\mathbf{q}_0,\omega_0;\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)\equiv S(012)$. The six different $S(abc)$ fall into two cycles. Cycling the operators leads to a phase factor $S(bca)=e^{-\beta\hbar\omega_a}S(abc)$ [4]. The two different cycles can be related through $S(abc)=S(-c-b-a)$ where the sign is of relevance only for the frequency arguments since the quadratic structure function is even in the combined wave-vector arguments. The linear analog of all these relations is the simple $S(\mathbf{q},\omega)=e^{\beta\hbar\omega}S(\mathbf{q},-\omega)$.

As in the linear case, the quadratic density response function and the quadratic structure function are connected through a fluctuation-dissipation relation. The quadratic fluctuation-dissipation theorem (QFDT) was established by Golden, Kalman, and Silevitch for a classical system [5] and later by Kalman and Gu for a quantum system [3]. But while the linear fluctuation-dissipation theorem (FDT)

$$\{S(\mathbf{q},\omega)\}_{\mathrm{odd}}=S(\mathbf{q},\omega)-S(\mathbf{q},-\omega)=S(\mathbf{q},\omega)(1-e^{-\beta\hbar\omega})$$
$$=\frac{-2\hbar}{n}\chi''(\mathbf{q},\omega)$$

relates the odd part of the structure function to the imaginary part of the density response, the QFDT links the *even* projection in the combined frequency arguments of the quadratic structure function to the *real* part of the quadratic density response. The QFDT can be written in three different combinations. We will use the following forms:

$$\{S(210)-S(201)\}_{\mathrm{even}}=\{S(210)-S(201)\}(1-e^{-\beta\hbar\omega_2})$$
$$=-\frac{4\hbar^2}{n}\{\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$$
$$-\chi'(\mathbf{q}_2,\omega_2;\mathbf{q}_0,\omega_0)\}, \tag{3a}$$

$$\{S(120)-S(102)\}_{\mathrm{even}}=\{S(120)-S(102)\}(1-e^{-\beta\hbar\omega_1})$$
$$=-\frac{4\hbar^2}{n}\{\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$$
$$-\chi'(\mathbf{q}_0,\omega_0;\mathbf{q}_1,\omega_1)\}. \tag{3b}$$

[We define the static structure factor as the integral $S_{\mathbf{q}}=\int_{-\infty}^{\infty}(d\omega/2\pi)S(\mathbf{q},\omega)$. Our $S(\mathbf{q},\omega)$ differs from the one used in Ref. [6] by a factor $2\pi/n$. Our notation is also slightly different from that of Ref. [4] where the symbol $\hat{\chi}$ is used for χ. χ' and χ'' stand for real and imaginary parts, respectively. The linear and the quadratic versions of χ and S are distinguished in this paper by the number of their arguments.] While it is $\chi'(a,b)$, the *real* part of the quadratic density response, that appears in the QFDT, it should be kept in mind that it is related to the *imaginary* part $\alpha''(a,b)$ of the quadratic polarizability through $\alpha(1,2)=(i4\pi e^3/q_0q_1q_2)\,\chi(1,2)$. We note that causality requirements imply that the quadratic density response function obeys Kramers-Kronig relations for both frequency arguments independently.

The usefulness of sum rules in designing, controlling and checking approximation schemes for the calculation of response functions is well established. The frequency moments calculated through the sum rules play another formal role: they are the coefficients of the high frequency expansion of the response function. Very little is known about the analytic structure of the quadratic quantities, the guideline provided by the sum rules in this connection should be especially valuable. The quadratic response functions govern the response of electron gases to finite amplitude electromagnetic perturbations; therefore the constraints imposed by the sum rules are expected to have a significant bearing on the description of these processes. Somewhat less obviously, the quadratic response and structure functions are fundamentally linked to the correlational properties of interacting electrons [7] and thus the quadratic sum rules can play a role in the determination of the *linear* response of a strongly correlated electron liquid. In particular, as in such calculations perturbative approaches are to be avoided, the sum rules can be exploited to construct nonperturbative approximation schemes.

The plan for this paper is as follows: In Sec. II we recalculate the known, and derive frequency moment sum rules for the quadratic density response. We then discuss how the frequency moments relate to the high frequency expansion of the response function. Section III discusses sum rule relations for the quadratic dynamical structure function. Finally we look at the high temperature, classical, and the zero temperature limits of these sum rules. The Appendix provides details on the derivation of classical frequency moments of the quadratic structure function.

II. SUM RULES FOR THE QUADRATIC RESPONSE FUNCTION

In this section we will analyze the $\omega_a^{r_a}$-$\omega_b^{r_b}$ frequency moments for the quadratic density response function beginning with the moments of even combined power r_a+r_b. The r_a-r_b moment will be designated by X_{r_a,r_b}.

$$X_{r_a,r_b}[a,b]:=\int\frac{d\omega_b}{2\pi}\int\frac{d\omega_a}{2\pi}\,\omega_a^{r_a}\omega_b^{r_b}\chi(\mathbf{q}_a,\omega_a;\mathbf{q}_b,\omega_b). \tag{4}$$

The notation $[a,b]$ refers to the order of integration which will be understood as given in the square brackets and is, in general, relevant (i.e., $X_{r_b,r_a}[b,a]\neq X_{r_a,r_b}[a,b]$). On the other hand, renaming the variables $a\leftrightarrow b$ simply amounts to changing $\mathbf{q}_a\leftrightarrow\mathbf{q}_b$ in the result and does not change the type of moment. Note that $\chi(a,b)=\chi(b,a)$, i.e., the quadratic density response function is symmetric in its two arguments. Normally we will consider $a=1$ and $b=2$ but some other combinations will be needed, too.

Sum rules will be obtained when X_{r_a,r_b} can be expressed in terms of ''known'' or simpler quantities. We will follow Tao and Kalman [2] in the derivation of the first single frequency moment of the quadratic χ and then establish the higher moments.

A. Single frequency moments

We can start with the trivial zeroth moment. It follows from the plus-function character of $\chi(\omega_1,\omega_2)$—causality requires that $\chi(\omega_1,\omega_2)$ is a regular function in the upper ω_1 and ω_2 half-planes [8]—and from its sufficiently fast vanishing for $\omega_1\to\infty$, $\omega_2\to\infty$ [cf. Eq. (13) below], that

$$\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=0. \tag{5}$$

We can evaluate higher frequency moments directly by using the Kubo procedure. The starting point is the density response function as given by the fluctuation-dissipation theorem in the time domain. For comparison, we quote the linear relations as well.

$$\chi(\mathbf{q},t)=-i\frac{\Theta(t)}{\hbar V}\langle[\varrho_{\mathbf{q}}(0),\varrho_{-\mathbf{q}}(-t)]\rangle,$$

$$\chi(\mathbf{q}_1,t_1;\mathbf{q}_2,t_2)=-\frac{\Theta(t_1)\Theta(t_2)}{2\hbar^2V}\{\Theta(t_2-t_1)\times\langle[[\varrho_{-\mathbf{q_0}}(0),\varrho_{-\mathbf{q_1}}(-t_1)],\varrho_{-\mathbf{q}_2}(-t_2)]\rangle+\Theta(t_1-t_2)\times\langle[[\varrho_{-\mathbf{q_0}}(0),\varrho_{-\mathbf{q}2}(-t_2)],\varrho_{-\mathbf{q_1}}(-t_1)]\rangle\}. \quad (6)$$

Here $\Theta(t)$, etc., are step functions ensuring the causal behavior of the response functions. Taking now the derivative of Eq. (6) with respect to t_1 at $t_1=0$, we find that the right-hand side has only one nonvanishing contribution. All other terms vanish because the δ distributions as the derivatives of Θ and the product $\Theta(t_2)\Theta(-t_2)$ leave us with vanishing equal-time commutators. The result contains an additional factor $\frac{1}{2}$ which comes from the step function $\Theta(t_1)$ taken at $t_1=0$.

$$\left.\frac{\partial}{\partial t_1}\chi(\mathbf{q}_1,t_1;\mathbf{q}_2,t_2)\right|_{t_1=0}=-i\frac{\Theta(t_2)}{4\hbar^3V}\times\langle[[\varrho_{-\mathbf{q_0}}(0),[\varrho_{-\mathbf{q_1}}(0),H]],\varrho_{-\mathbf{q}_2}(-t_2)]\rangle^{(0)}. \quad (7)$$

So far no specification of the many body Hamiltonian has been necessary. To proceed further we assume that the system is an electron gas with a neutralizing background [a quantum OCP (one component plasma)]. With this Hamiltonian now the inner, equal time double commutator can be evaluated exactly, resulting in

$$[\varrho_{-\mathbf{q_0}}(0),[\varrho_{-\mathbf{q_1}}(0),H]]=\hbar^2\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{m}\varrho_{\mathbf{q}_2}$$

and leading to the relation

$$\left.\frac{\partial}{\partial t_1}\chi(\mathbf{q}_1,t_1;\mathbf{q}_2,t_2)\right|_{t_1=0}=\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{4m}\chi(\mathbf{q}_2,t_2). \quad (8)$$

In the frequency domain this amounts to

$$\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=i\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{4m}\chi(\mathbf{q}_2,\omega_2). \quad (9)$$

This is the basic sum rule established by Tao and Kalman [2].

The second derivative of Eq. (6) with respect to t_1 can be calculated directly as well. Using the δ distribution's properties $x\delta(x)=0$ and $x\delta'(x)=-\delta(x)$ we find the form

$$\frac{\partial^2}{\partial t_1^2}\chi(\mathbf{q}_1,t_1;\mathbf{q}_2,t_2)=-\delta(t_1)\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\chi(\mathbf{q}_2,t_2)-\delta(t_2-t_1)\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}\chi(-\mathbf{q}_0,t_1)+\frac{\Theta(t_1)\Theta(t_2)}{2\hbar^2V}\{\Theta(t_2-t_1)\times\langle[[\varrho_{-\mathbf{q}_0}(0),\ddot{\varrho}_{-\mathbf{q}_1}(-t_1),\varrho_{-\mathbf{q}_2}(-t_2)]^{(0)}+\Theta(t_1-t_2)\langle[[\varrho_{-\mathbf{q}_0}(0),\varrho_{-\mathbf{q}_2}(-t_2)],\ddot{\varrho}_{-\mathbf{q}_1}(-t_1)]\rangle^{(0)}. \quad (10)$$

Now letting t_1 approach 0, the second term on the right and the second term inside the curly brackets vanish and the remaining inner commutator can be evaluated:

$$[\varrho_{-\mathbf{q}_0},\ddot{\varrho}_{-\mathbf{q}_1}]=-2\hbar^2\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{m}\sum_{\mathbf{p}}\frac{\mathbf{p}\cdot\mathbf{q}_1}{m}c^+_{\mathbf{p}+\mathbf{q}_2/2}c_{\mathbf{p}-\mathbf{q}_2/2}.$$

Recalling that the particle-current density is $\mathbf{j}_{q_2}=\Sigma_{\mathbf{p}}(\mathbf{p}/m)c^+_{\mathbf{p}+\mathbf{q}_2/2}c_{\mathbf{p}-\mathbf{q}_2/2}$, the result can be written as

$$\left.\frac{\partial^2}{\partial t_1^2}\chi(\mathbf{q}_1,t_1;\mathbf{q}_2,t_2)\right|_{t_1=0}=-\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\left[\chi(\mathbf{q}_2,t_2)\delta(t_1)-\frac{\Theta(t_2)}{V}\times\langle[\mathbf{q}_1\cdot\mathbf{j}_{q_2}(0),\varrho_{-\mathbf{q}_2}(-t_2)]\rangle^{(0)}\right]_{t_1=0}. \quad (11)$$

The delta distribution $\delta(t_1)$ on the right constitutes an infinite constant at $t_1=0$, but when in the next subsection we take $t_2=0$, the term in question will vanish due to $\chi(\mathbf{q}_2,t_2=0)=0$. The second term on the right contains the linear density-current-density response function $\chi_{\mathbf{j}\varrho}$, a vector.

A Fourier transformation into the frequency domain yields

$$\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1^2\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\{C\chi(\mathbf{q}_2,\omega_2)-i\mathbf{q}_1\cdot\boldsymbol{\chi}_{\mathbf{j}\varrho}(\mathbf{q}_2,\omega_2)\}, \quad (12)$$

where C is the infinite constant of dimension t^{-1} resulting from the delta distribution $\delta(t_1)$.

The origin of this divergent term can be understood by contemplating the exact asymptotic high frequency behavior of $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ that is of the form [1] (χ is dominated by χ' as $\omega_1\to\infty$, $\omega_2\to\infty$)

$$\chi'(\mathbf{q}_1,\omega_1\to\infty;\mathbf{q}_2,\omega_2\to\infty)=\frac{n}{2m^2}\frac{1}{\omega_0\omega_1\omega_2}\times\left\{\frac{q_0{}^2}{\omega_0}(\mathbf{q}_1\cdot\mathbf{q}_2)+\frac{q_1{}^2}{\omega_1}(\mathbf{q}_0\cdot\mathbf{q}_2)+\frac{q_2{}^2}{\omega_2}(\mathbf{q}_0\cdot\mathbf{q}_1)\right\}. \quad (13)$$

It is clear that for fixed ω_2 the leading term in the integrand goes as ω_1^{-2}, generating a divergent ω_1 integral in Eq. (12). This consideration also shows that more severe divergences would arise from higher than second order single frequency moments. On the other hand, as it is obvious from Eq. (9), there is no divergence problem for the ω_1 moment which, for parity reasons, combines only with the ω_1^{-3} term in Eq. (13). More will be said on this later.

We can now proceed to find the double frequency moments by either taking the derivative of Eqs. (8) and (11) with respect to t_2 at $t_2=0$ or multiplying Eqs. (9) and (12) by and then integrating over ω_2.

B. Double frequency moments

The double frequency moments fall into two classes, depending on whether r_1+r_2 is even or odd. For parity reasons only $\chi'(1,2)$ contributes to the former and only $\chi''(1,2)$ contributes to the latter. First we review the even moments.

Starting from Eq. (4) with $r_1=r_2=-1$ one finds as a direct result of the Kramers-Kronig relation

$$X_{-1,-1}[1,2]=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\frac{1}{\omega_1\omega_2}\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=-\frac{1}{4}\chi(\mathbf{q}_1,0;\mathbf{q}_2,0)\equiv-\frac{1}{4}\chi_{\mathbf{q}_1,\mathbf{q}_2}. \quad (14)$$

In the long wavelength limit ($\mathbf{q}_1\to0$, $\mathbf{q}_2\to0$) the static $\chi_{\mathbf{q}_1,\mathbf{q}_2}$ is subject to a quadratic compressibility sum rule [1], from which follows that

$$\chi_{\mathbf{q}_1,\mathbf{q}_2}\to\frac{n}{2}\frac{(\partial p/\partial n)_T-n(\partial^2p/\partial n^2)_T}{(\partial p/\partial n)_T^3}.$$

Combining now Eq. (9) and the Kramers-Kronig relations for $\chi(1,2)$ we find the frequency quotient sum rule

$$X_{1,-1}[1,2]=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\frac{\omega_1}{\omega_2}\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=-\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{8m}\chi_{\mathbf{q}_2}. \quad (15)$$

The order of integration can be changed in this case (i.e., $X_{1,-1}[1,2]=X_{-1,1}[2,1]$), so that we find the moment $X_{-1,1}$ by just interchanging the indices of the arguments in Eq. (15).

$$X_{-1,1}[1,2]=-\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{8m}\chi_{\mathbf{q}_1}. \quad (16)$$

In the long wavelength limit the static $\chi_{\mathbf{q}_1}$and $\chi_{\mathbf{q}_2}$ are subject to the routine linear compressibility sum rule.

These latter frequency moments are of zeroth order in the combined power of ω_1 and ω_2. The remaining zero order moment is $X_{0,0}$, a mere integration over $\chi(1,2)$. But in view of Eq. (5), the contour integral vanishes and so does $X_{0,0}$.

$$X_{0,0}[1,2]=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=0. \quad (17)$$

Turning now to the second order moments, we note that the moment $X_{1,1}$ is the quadratic equivalent of the well known f-sum rule [2]

$$X_{1,1}[1,2]=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\omega_1\omega_2\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=\frac{n}{2}\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\frac{q_2{}^2}{2m}. \quad (18)$$

The order of integration is relevant here and this explains why the right side is not symmetric in the indices. Renaming the variables gives the moment $X_{1,1}[2,1]$ with changed order of integration. The noncommutability of the ω_1 and ω_2 integration results in a δX term, which will play a role in Sec. III. More details are given in Appendix A.

$$X_{1,1}[1,2]-X_{1,1}[2,1]=\delta X_{1,1}=\frac{n}{2}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}\frac{q_1^2-q_2^2}{2m}. \quad (19)$$

The $X_{2,0}$ moment we find from Eq. (11). After setting $t_2=0$, the divergent term vanishes, the commutator can be evaluated and Fourier transformation into the ω_1-ω_2 domain produces the $X_{2,0}$ moment. Alternatively, we can integrate Eq. (12) over ω_2. In this case the plus-function character of $\chi(\omega_2)$ causes the first term in Eq. (12) to vanish and one is left with

$$X_{2,0}[1,2]=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\omega_1^2\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=n\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}. \quad (20)$$

Renaming the variables here gives the $X_{2,0}[2,1]$ moment. Changing the order of integration in (20) creates $X_{0,2}[2,1]$ which vanishes according to Eq. (5). Therefore

$$\delta X_{2,0}=X_{2,0}[1,2]-X_{0,2}[2,1]=X_{2,0}[1,2]. \quad (21)$$

By making use of Puff's ω^3-moment sum rule [9] for the linear χ we can write down one of the fourth order moments of $\chi(1,2)$, namely,

$$X_{1,3}[1,2]=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\omega_1\omega_2{}^3\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=\frac{n}{2}\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\frac{q_2{}^2}{2m}\left\{\omega_p{}^2+\frac{q_2{}^2}{2m}\left[\frac{\hbar^2q_2{}^2}{2m}+4\langle\varepsilon_{kin}\rangle\right]+\frac{\omega_p{}^2}{N}\sum_k\frac{(\mathbf{k}\cdot\mathbf{q}_2)^2}{k^2q_2{}^2}[S_{\mathbf{q}_2-\mathbf{k}}-S_{\mathbf{k}}]\right\}, \quad (22)$$

where $\omega_p{}^2 = 4\pi ne^2/m$ and $\langle \varepsilon_{kin} \rangle$ is the expectation value of the kinetic energy per electron in the interacting system.

The other fourth order moments $X_{2,2}$ and $X_{3,1}$ contain diverging contributions, that can be understood in view of the remarks made earlier; cf. Eq. (13).

Turning now to the odd moments we note that the two moments $X_{0,-1}[1,2]$ and $X_{-1,0}[1,2]$ vanish:

$$X_{0,-1}[1,2] = \int \frac{d\omega_2}{2\pi} \int \frac{d\omega_1}{2\pi} \frac{1}{\omega_2} \chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2) = 0 \tag{23}$$

because of Eq. (5) and

$$X_{-1,0}[1,2] = \int \frac{d\omega_2}{2\pi} \int \frac{d\omega_1}{2\pi} \frac{1}{\omega_1} \chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2) = i\frac{1}{2} \int \frac{d\omega_2}{2\pi} \chi(\mathbf{q}_1,0;\mathbf{q}_2,\omega_2) = 0 \tag{24}$$

for similar reasons.

The first order moments, both $X_{1,0}$ and $X_{0,1}$, again trivially vanish because of the plus-function behavior of $\chi(\omega_1,\omega_2)$ and $\chi(\omega_2)$.

The situation is different for the two third order moments $X_{1,2}$ and $X_{2,1}$: both lead to divergent integrals, as is obvious from Eqs. (9) and (12).

All the moments listed, with the exception of $X_{1,3}[1,2]$, are coupling independent and thus are exhausted both by $\chi_0(1,2)$, the quadratic response function of the noninteracting gas and by $\chi_{RPA}(1,2)$, the response calculated in the random phase approximation:

$$\chi_{RPA}(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2) = \frac{\chi_0(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)}{\epsilon_0(\mathbf{q}_0,\omega_0)\epsilon_0(\mathbf{q}_1,\omega_1)\epsilon_0(\mathbf{q}_2,\omega_2)},$$

where $\epsilon_0(\mathbf{q},\omega)$ is the Lindhard dielectric function. The assertion concerning $\chi_0(1,2)$ can be verified by direct calculation.

In summary, there are five nontrivial sum rules for $X_{-1,1}$, $X_{1,-1}$, $X_{1,1}$, $X_{2,0}$, and $X_{1,3}$, all of them pertaining to the real part $\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$. This is in contrast to the linear situation where one deals with the frequency moments of the imaginary part $\chi''(\mathbf{q},\omega)$. While $\chi''(\mathbf{q},\omega)$ vanishes very fast for high ω values and thus the construction of the linear frequency moments is never problematic, the high frequency behavior of $\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ is governed by the inverse power of ω_1 and ω_2 and its leading terms are given by the exact high frequency expansion, Eq. (13).

In order to infer the limitations on the existence of the frequency moments of $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ one has to let $\omega_1 \to \infty$ for fixed ω_2 in Eq. (13):

$$\chi'(\mathbf{q}_1,\omega_1\to\infty;\mathbf{q}_2,\omega_2) \cong + \frac{\Omega_{2,2}}{\omega_1^2\omega_2^2} + \frac{\Omega_{3,1}}{\omega_1^3\omega_2} + \frac{\Omega_{4,0}}{\omega_1^4}, \tag{25}$$

$$\Omega_{2,2} = -2n \frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m} \frac{q_2^2}{2m},$$

$$\Omega_{3,1} = -4n \frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m} \frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m},$$

$$\Omega_{4,0} = 2n \frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m} \frac{\mathbf{q}_0\cdot(3\mathbf{q}_1+\mathbf{q}_2)}{2m}.$$

The resulting expression Eq. (25) shows that the integral generating the ω_1^1 moment (to which the $\Omega_{2,2}$ term, for parity reasons, does not contribute) exists, while that generating the ω_1^2 moment is divergent, in accordance with Eqs. (9) and (12).

The high frequency expansion Eq. (13) and its offspring Eq. (25) are based on the asymptotic behavior of $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ in the hydrodynamic limit. A more general approach can be generated, again in some analogy with the linear case, through an expansion in terms of the frequency moments X_{r_a,r_b}. This is made possible by exploiting that $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ is its own double Hilbert transform:

$$\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2) = -P\int \frac{d\overline{\omega_2}}{\pi} \times P\int \frac{d\overline{\omega_1}}{\pi} \frac{\chi(\mathbf{q}_1,\overline{\omega_1};\mathbf{q}_2,\overline{\omega_2})}{(\omega_1-\overline{\omega_1})(\omega_2-\overline{\omega_2})}. \tag{26}$$

Letting $\omega_1\to\infty$ and $\omega_2\to\infty$ (in this order), the high frequency expansion of the real part can be evaluated by the steps indicated below:

$$\begin{aligned}\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2) &= -P\int \frac{d\overline{\omega_2}}{\pi} P\int \frac{d\overline{\omega_1}}{\pi} \times \frac{\chi'(\mathbf{q}_1,\overline{\omega_1};\mathbf{q}_2,\overline{\omega_2})}{(\omega_1-\overline{\omega_1})(\omega_2-\overline{\omega_2})} \\ &\cong -P\int \frac{d\overline{\omega_2}}{\pi}\int \frac{d\overline{\omega_1}}{\pi} \frac{\chi'(\mathbf{q}_1,\overline{\omega_1};\mathbf{q}_2,\overline{\omega_2})}{\omega_1(\omega_2-\overline{\omega_2})} \times\left[1+\frac{\overline{\omega_1}}{\omega_1}+\left(\frac{\overline{\omega_1}}{\omega_1}\right)^2+\cdots\right] \\ &= \frac{1}{\omega_1}\int \frac{d\overline{\omega_1}}{\pi}\left[1+\frac{\overline{\omega_1}}{\omega_1}+\left(\frac{\overline{\omega_1}}{\omega_1}\right)^2+\cdots\right] \times \chi''(\mathbf{q}_1,\overline{\omega_1};\mathbf{q}_2,\omega_2).\end{aligned} \tag{27}$$

The resulting $\omega_1{}^n$ moments of $\chi''(1,2)$ for $n=0,1,2$ can be evaluated with the aid of Eqs. (5), (9), and (12), and their high frequency expansion can be calculated from the known asymptotic behavior of the linear response function. For these lower order moments it is also permissible to expand *both* Kramers-Kronig denominators which then provides

$$\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)\cong-\int\frac{d\overline{\omega_2}}{\pi}\int\frac{d\overline{\omega_1}}{\pi}\frac{\chi'(\mathbf{q}_1,\overline{\omega_1};\mathbf{q}_2,\overline{\omega_2})}{\omega_1\omega_2}\times\left[1+\frac{\overline{\omega_1}}{\omega_1}+\cdots\right]\left[1+\frac{\overline{\omega_2}}{\omega_2}+\cdots\right]=-4\left\{\frac{X_{1,1}}{\omega_1^2\omega_2^2}+\frac{X_{2,0}}{\omega_1^3\omega_2}+\frac{X_{0,2}}{\omega_1\omega_2^3}+\frac{X_{1,3}}{\omega_1^2\omega_2^4}+\frac{X_{2,2}}{\omega_1^3\omega_2^3}+\cdots\right\}. \quad (28)$$

The moments $X_{1,1}$, $X_{2,0}$, and $X_{0,2}$ as given in Eqs. (18), (20), etc. are indeed in agreement with the corresponding $\Omega_{m,n}$ coefficients in Eq. (25). As to the $n\geqslant 3$ moments of $\chi''(1,2)$, no general result is available; nevertheless the $n=3$ moment for the noninteracting $\chi_0(1,2)$ can be calculated, yielding the correct $\Omega_{4,0}$ coefficient in Eq. (25).

One can contemplate the more general high frequency expansion of $\chi'(1,2)$ expressed as $\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)=\Sigma_{m,n(m+n\geqslant 4,even;m\geqslant 2)}\Omega_{m,n}/\omega_1^m\omega_2^n$. As it is clear from the foregoing the expansion for $m+n=4$ is unaffected by correlations and therefore would be exhausted both by $\chi_0'(1,2)$ and $\chi_{RPA}'(1,2)$. Correctional contributions are to show up in $\Omega_{2,4}$, $\Omega_{3,3}$, and $\Omega_{4,2}$. The first two can be derived from Eqs. (22) and (12); the calculation of $\Omega_{4,2}$ hinges upon the evaluation of the ω_1^3 moment of $\chi'(1,2)$. (Details of the expansion will be discussed elsewhere.)

Moments of the (frequency and wave-number) shifted $\chi(\mathbf{q}_0,\omega_0;\mathbf{q}_1,\omega_1)$ and $\chi(\mathbf{q}_0,\omega_0;\mathbf{q}_2,\omega_2)$ are also of interest and will be, in fact, needed in the next section. To transform such moments into the form (4) we can substitute, shift, and rename the integration variables. The procedure is illustrated below for the case $r_1=r_2=1$:

$$\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\omega_1\omega_2\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_0,\omega_0)$$
$$=\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\omega_1\omega_2\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_0,\omega_0)+\delta X_{1,1}$$
$$=\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_0}{2\pi}\omega_1(-\omega_0-\omega_1)\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_0,\omega_0)+\delta X_{1,1}$$
$$=-X_{1,1}[0,1]-X_{0,2}[0,1]+\frac{n}{2}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}\frac{q_1^2-q_2^2}{2m}. \quad (29)$$

The change of the order of integration in the second step is necessary in order to facilitate the substitution from the set $(\omega_1\omega_2)$ to the set $(\omega_0\omega_1)$: in the original order the replacement of ω_2 by $\omega_0=-(\omega_1+\omega_2)$ *after* the integration over ω_1 would not be allowed. $\delta X_{1,1}$ is taken from Eq. (19). That this can be done is not quite obvious, since in (19) the integration variables are ω_1 and ω_2, while in (29) ω_1 and ω_0. The justification is detailed in Appendix A.

III. SUM RULES FOR THE QUADRATIC S

The knowledge of the X_{r_1,r_2} moments allows one to generate similar frequency moments for the quadratic dynamical structure function S. Since the ordering of the density operators within the S functions is important, one has to adhere to a convention as to the chosen ordering for the definition of the moments. We choose the moments of $S(201)$ and $S(102)$ in the following; moments for other orderings or of symmetrized combinations can, however, be generated by a similar procedure. Let us define Z_{r_1,r_2} as

$$Z_{r_1,r_2}:=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1^{r_1}\omega_2^{r_2}\times[1-(-1)^{r_1+r_2}e^{-\beta\hbar\omega_2}]S(201). \quad (30)$$

The exponential in the integrand stems from the inclusion of both S cycles; cf. Eq. (3a). Its sign is determined by the combined parity of the frequencies. The other moment we will use is $\overline{Z}_{r_1,r_2}$, defined as

$$\overline{Z}_{r_1,r_2}:=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1^{r_1}\omega_2^{r_2}\times[1-(-1)^{r_1+r_2}e^{-\beta\hbar\omega_1}]S(102)=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1^{r_1}\omega_2^{r_2}\times[1-(-1)^{r_1+r_2}e^{+\beta\hbar\omega_1}]S(201). \quad (31)$$

To find such Z and $\overline{Z}$ moments we multiply the QFDT, Eqs. (3a) and (3b), respectively, by $\omega_1^{r_1}\omega_2^{r_2}$ and integrate over both frequencies. Under the integral the two different S cycles can be combined by flipping the signs of all ω_i for one S. For even r_1+r_2 Z_{r_1,r_2} then is given in terms of X moments as

$$Z_{r_1,r_2}=\frac{2\hbar^2}{n}\left\{X_{r_1,r_2}[1,2]-(-1)^{r_1}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_0}{2\pi}\times(\omega_0+\omega_2)^{r_1}\omega_2^{r_2}\chi'(\mathbf{q}_0,\omega_0;\mathbf{q}_2,\omega_2)\right\}. \quad (32)$$

For $\overline{Z}_{r_1,r_2}$ we find accordingly

$$\overline{Z}_{r_1,r_2}=\frac{2\hbar^2}{n}\left\{X_{r_1,r_2}[1,2]-(-1)^{r_2}\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\times\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1^{r_1}\omega_2{}^{r_2}\chi'(\mathbf{q}_0,\omega_0;\mathbf{q}_1,\omega_1)\right\}, \quad (33)$$

where special care has to be taken with the double integral. See Appendix A for more discussion.

Now we can immediately write down the $Z_{1,1}$ moment using $X_{1,1}$ and the fact that $X_{0,2}$ vanishes. Note that due to the symmetry of Eq. (18) in the indices 0 and 1 the moments $X_{1,1}[1,2]$ and $X_{1,1}[0,2]$ are identical.

$$Z_{1,1}=\frac{2\hbar^2}{n}\{X_{1,1}[1,2]+X_{1,1}[0,2]\}$$
$$=2\hbar^2\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\frac{q_2^2}{2m}. \tag{34}$$

The 2-0 moment is given in terms of $X_{2,0}$ and $X_{1,1}$.

$$Z_{2,0}=\frac{2\hbar^2}{n}\{X_{2,0}[1,2]-X_{2,0}[0,2]-2X_{1,1}[0,2]-X_{0,2}[0,2]\}$$
$$=4\hbar^2\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}. \tag{35}$$

The moment $Z_{0,2}$ is composed of two $X_{0,2}$ moments and thus vanishes. More generally it can be seen from Eq. (5) and the QFDT, Eq. (3a), that all Z moments with $r_1=0$ vanish.

The corresponding $\overline{Z_{1,1}}$ moment is found according to Eq. (29):

$$\overline{Z_{1,1}}=\frac{2\hbar^2}{n}\{X_{1,1}[1,2]+X_{1,1}[0,1]+X_{0,2}[0,1]-\delta X_{1,1}\} \tag{36}$$

which reduces to

$$\overline{Z_{1,1}}=2\hbar^2\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{2m}\frac{q_1^2}{2m}. \tag{37}$$

In calculating $\overline{Z_{0,2}}$ and $\overline{Z_{2,0}}$ we encounter integrals similar to (29) but creating a $\delta X_{0,2}$ an $\delta X_{2,0}$, respectively, when the order of integration is changed:

$$\overline{Z_{0,2}}=\frac{2\hbar^2}{n}\Big\{-X_{0,2}[1,2]-\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\times\omega_2^2\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_0,\omega_0)\Big\}$$
$$=-\frac{2\hbar^2}{n}\Big\{\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_0}{2\pi}(-\omega_0-\omega_1)^2\times\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_0,\omega_0)+\delta X_{0,2}\Big\}$$
$$=-\frac{2\hbar^2}{n}\Big\{X_{2,0}[0,1]+2X_{1,1}[0,1]+X_{0,2}[0,1]-n\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{2m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}\Big\}=4\hbar^2\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{2m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m} \tag{38}$$

and

$$\overline{Z_{2,0}}=\frac{2\hbar^2}{n}\{X_{2,0}[1,2]-X_{0,2}[0,1]-\delta X_{2,0}\}=0. \tag{39}$$

Observe that $\overline{Z_{r_1,r_2}}(\mathbf{q}_1,\mathbf{q}_2)=Z_{r_2,r_1}(\mathbf{q}_2,\mathbf{q}_1)$, which shows that the ω_1 and ω_2 integrations commute as far as the Z moments are concerned. This is true, even though the individual X moments which combine into a particular Z moment are obviously sensitive to the order of integration. The reason for the difference between the behavior of the Z and X moments has to be sought in the Z moments' expected much faster vanishing for $\omega_1\to\infty$, $\omega_2\to\infty$, and rendering the ω_1-ω_2 integrals unconditionally convergent (cf. Appendix A for details).

To find the odd $Z_{1,0}$ moment we divide the QFDT, Eq. (3a), by $(1-e^{-\beta\hbar\omega_2})$ before integrating over ω_2. At this point we can employ the single frequency moment Eq. (9) and the linear FDT to find the additional relationship

$$\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1\{S(201)-S(210)\}=\frac{\hbar}{m}\mathbf{q}_0\cdot\mathbf{q}_1S(\mathbf{q}_2,\omega_2). \tag{40}$$

An equivalent expression which has the indices 0 and 2 exchanged can be derived in the same manner.

If we now integrate Eq. (40) over ω_2, we are again able to combine the two different S cycles by flipping the signs of all ω_i for one S. The result relates the frequency integral of the dynamical quadratic structure function to the static linear structure function $S_{\mathbf{q}}$:

$$Z_{1,0}=\hbar\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{m}S_{\mathbf{q}_2}. \tag{41}$$

The above procedure is not applicable to the QFDT, Eq. (3b), with our chosen order of integration. Nevertheless, exploiting that the order of integration is not relevant we can write down the corresponding $\overline{Z_{0,1}}$ moment:

$$\overline{Z_{0,1}}=\hbar\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{m}S_{\mathbf{q}_1}. \tag{42}$$

If we first multiply Eq. (40) by ω_2 and then integrate the f-sum rule for the linear dynamical structure function can be used to find the $Z_{1,1}$ moment, Eq. (34), again.

The definition of the static quadratic structure function [3]

$$S_{\mathbf{q}_1,\mathbf{q}_2}:=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\frac{1}{3}\{S(012)+S(120)+S(201)\}$$

could be viewed as another frequency moment if the different cycles of S are combined:

$$\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\frac{1}{3}(1+e^{+\beta\hbar\omega_1}+e^{-\beta\hbar\omega_2})S(201)=S_{\mathbf{q}_1,\mathbf{q}_2}. \tag{43}$$

The moments (34), (35), (37), (38), and (41) constitute sum rules for the quadratic dynamic quantum structure function. One should keep in mind that the exponential in the integrand is a consequence of the fact that the density operators do not commute for a quantum system. It is also interesting to note that the first order moment is at least $O(\hbar)$ and correlation-dependent while the second order moments are $O(\hbar^2)$ and correlation-independent. The sum rules derived in this section can provide guidance in the construction of an approximation for a dynamical density response function. This will be discussed elsewhere.

A. Classical limits

The high temperature, classical limits ($\beta\hbar \to 0$) of the Z moments can be calculated directly and lead to pure frequency moments of $S_{classic}(012)$,

$$M_{r_1,r_2} := \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \omega_1^{r_1} \omega_2^{r_2} S_{classic}(012).$$

Some of these have been derived earlier [5]. The method is recapitulated and extended to higher moments in Appendix C. Here we show how the $\beta\hbar \to 0$ limit can be obtained directly from the results above. It is obvious that the classical M moments satisfy $M_{r_1,r_2}(\mathbf{q}_1,\mathbf{q}_2) = M_{r_2,r_1}(\mathbf{q}_2,\mathbf{q}_1)$. A comparison with the direct classical calculation is given in Appendix C.

First we need to expand $S(201)$ in $\hbar$. An expression for S in terms of $\chi'(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ has been derived in [3]:

$$S(201) = -\frac{4\hbar^2}{nD}\{(1-e^{-\beta\hbar\omega_1})[\chi'(12)-\chi'(20)] - (1-e^{+\beta\hbar\omega_2})[\chi'(12)-\chi'(01)]\}. \tag{44a}$$

D stands for

$$D = -2[\sinh(\beta\hbar\omega_0) + \sinh(\beta\hbar\omega_1) + \sinh(\beta\hbar\omega_2)] = -\hbar^3\beta^3\omega_0\omega_1\omega_2 + O(\hbar^5). \tag{44b}$$

In the limit $\hbar \to 0$ S can be expanded as

$$S(201) = S(012)|_{\hbar=0} + \hbar\,\delta S(201) + O(\hbar^2). \tag{45}$$

Then, to lowest order in $\hbar$, one finds from Eq. (44a)

$$S(012)|_{\hbar=0} \equiv S_{classic}(012) = -\frac{4}{n\beta^2}\left[\frac{\chi'(12)}{\omega_1\omega_2} + \frac{\chi'(20)}{\omega_2\omega_0} + \frac{\chi'(01)}{\omega_0\omega_1}\right]. \tag{46}$$

This is the known classical QFDT expression [10]. The first quantum correction, linear in $\hbar$, is found to be

$$\delta S(201) = \frac{2}{n\beta}\left[\frac{\omega_1\chi'(20)}{\omega_2\omega_0} - \frac{\omega_2\chi'(01)}{\omega_0\omega_1} + \frac{(\omega_1-\omega_2)\chi'(12)}{\omega_1\omega_2}\right]. \tag{47}$$

Here χ' is understood to stand for $\chi'(\hbar=0)$. In contrast to $S_{classic}(012)$ the ordering of the arguments in $\delta S(201)$ is of relevance and specifically we have $\delta S(201) = -\delta S(102)$. The quantum correction to χ' can be ignored for it is at least of order $\hbar^2$. This can be inferred from the observation that the occurrence of $\hbar$ is linked to the frequencies and/or wave vectors and that $\chi'(a,b)$ is an even function in the combined frequency and the combined wave-vector arguments. Because various ω_1-ω_2 moments of δS contribute to the classical limits we will define

$$\Delta_{r_1,r_2} := \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \omega_1^{r_1} \omega_2^{r_2} \delta S(201). \tag{48}$$

For the calculation of the Δ_{r_1,r_2} we refer to Appendix B. Note that $\Delta_{m,n}(\mathbf{q}_1,\mathbf{q}_2) = -\Delta_{n,m}(\mathbf{q}_2,\mathbf{q}_1)$.

Before taking the classical limit of the various Z moments we observe that upon taking the classical limit of expression (40), the two $S_{classic}$ terms on the left-hand side cancel and to order $\hbar$ only the two δS of different orderings contribute. The integration over ω_1 reduces the quadratic χ' to linear χ'' by virtue of Eq. (9) and reproduces the classical linear FDT.

Since $S_{classic}$ is also an even function in its combined ω_1-ω_2 arguments, all moments with odd combined powers in ω_1 and ω_2 vanish due to parity [1]. Starting with the classical limit of $Z_{1,0}$ Eq. (41), to $O(\hbar^0)$ and $O(\hbar^2)$ the resulting equations yield vanishing odd $M_{1,0}$ and $M_{1,2}$ moments. The equation linear in $\hbar$ yields

$$M_{1,1} = \frac{2}{\beta}\Delta_{1,0} - \frac{\mathbf{q}_0\cdot\mathbf{q}_1}{\beta m} S_{\mathbf{q}_2},$$

which using (B1) gives the known classical frequency moment

$$M_{1,1} = \frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m} S_{\mathbf{q}_0}. \tag{49}$$

It is the terms stemming from δS that restore the 1-2 symmetry in the classical expression. The vanishing $Z_{0,1}$ provides to order $\hbar$ an equation for $M_{0,2}$

$$M_{0,2} = \frac{2}{\beta}\Delta_{0,1},$$

which using (68) becomes

$$M_{0,2} = -\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{\beta m} S_{\mathbf{q}_1} - \frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m} S_{\mathbf{q}_0}. \tag{50}$$

Turning now to the classical limits of $Z_{1,1}$ and $Z_{2,0}$, Eqs. (34) and (35), to order $\hbar^2$ they lead to the $M_{1,3}$ and $M_{2,2}$ moments of $S_{classic}$ in terms of Δ moments:

$$M_{1,3} = \frac{2}{\beta}\Delta_{1,2} - \frac{\mathbf{q}_0\cdot\mathbf{q}_1}{\beta m}\frac{q_2^2}{\beta m}, \tag{51}$$

$$M_{2,2} = \frac{2}{\beta}\Delta_{2,1} - 2\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{\beta m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m}. \tag{52}$$

The moment $Z_{0,2}$ provides the relation $M_{0,4} = (2/\beta)\Delta_{0,3}$.

By taking the $\hbar \to 0$ limit of $Z_{1,1}$ and $Z_{0,2}$ we find to order $\hbar^2$ the moments $M_{3,1}$ and again $M_{2,2}$,

$$M_{3,1} = -\frac{2}{\beta}\Delta_{2,1} - \frac{\mathbf{q}_0\cdot\mathbf{q}_2}{\beta m}\frac{q_1^2}{\beta m}, \tag{53}$$

$$M_{2,2} = -\frac{2}{\beta}\Delta_{1,2} - 2\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{\beta m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m}. \tag{54}$$

Furthermore the moment $\overline{Z}_{0,2}$ provides the relation $M_{4,0} = -(2/\beta)\Delta_{3,0}$. The comparison with direct classical calculations is given in Appendix C. Note that the symmetry of $S_{classic}(012)$ in all three arguments implies the relationships

$$M_{2,0}(q_0,q_1)=M_{2,0}(q_0,q_2)$$
$$=M_{2,0}(q_1,q_2)+2M_{1,1}(q_1,q_2)+M_{0,2}(q_1,q_2) \tag{55}$$

and

$$M_{4,0}(q_0,q_1)=M_{4,0}(q_0,q_2)$$
$$=M_{4,0}(q_1,q_2)+M_{0,4}(q_1,q_2)+4M_{3,1}(q_1,q_2)$$
$$+4M_{1,3}(q_1,q_2)+6M_{2,2}(q_1,q_2). \tag{56}$$

B. Zero temperature limit

The sum rules given in previous sections all hold for arbitrary temperature. Now taking the limit $T\rightarrow 0$ we can see immediately from the relation (43) that $S(201)$ must vanish if $\omega_1>0$ or $\omega_2<0$. This comes as no surprise, since in $S(201)$ we have a creation operator $c^+(\omega_2)$ acting on the ground state from the right and an annihilation operator $c(\omega_1)$ acting on the ground state from the left. A similar argument holds for all the six different S functions at $T=0$ and makes each of them vanish in all but two adjacent domains of the ω_1-ω_2 plane which is divided into six domains by the two axes and the line $\omega_1=\omega_2$. In turn, in any of these six domains only two of the S functions are not vanishing [3]. This can be compared with the linear case, where $S(\mathbf{q},\omega)$ vanishes for $\omega<0$ at $T=0$. In Eq. (3a) now one can concentrate on domains where only one of the S functions survives. For example, in the quadrant $\omega_1<0$, $\omega_2>0$, where $S(201)$ does not vanish, we can distinguish two cases and find that Eq. (3a) reduces to the much simpler expressions for $S(201)$:

$$0<\omega_2<-\omega_1: S(201)=\frac{4\hbar^2}{n}\{\chi'(12)-\chi'(20)\}, \tag{57a}$$

$$0<-\omega_1<\omega_2: S(201)=\frac{4\hbar^2}{n}\{\chi'(12)-\chi'(01)\}. \tag{57b}$$

The simplified relationships do not allow one, however, to obtain simpler sum rule expressions. This is due to the domain restrictions in (57a) and (57b). The integration over the unrestricted ω_1-ω_2 plane which generates the sum rule expressions sweeps over all six domains and picks up contributions by various combinations of the S functions. Thus the formal definitions of the moments Eqs. (30) and (31) remain unaffected, although it is more convenient to eliminate the exponential factor in the $T=0$ limit and rewrite the definitions of Z_{r_1,r_2} and $\overline{Z_{r_1,r_2}}$ as

$$Z_{r_1,r_2}=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\,\omega_1^{r_1}\omega_2^{r_2}\{S(012)|_{3,4}+S(210)|_{1,6}$$
$$-S(201)|_{5,6}-S(102)|_{2,3}\} \tag{58}$$

and

$$\overline{Z_{r_1,r_2}}=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\,\omega_1^{r_1}\omega_2^{r_2}\{S(021)|_{4,5}+S(120)|_{1,2}$$
$$-S(201)|_{5,6}-S(102)|_{2,3}\}. \tag{59}$$

The $|_i$ indicates that the integration is to be carried out in that domain only. The numbering of domains starts with the first quadrant and proceeds clockwise (cf. Ref. [3]).

IV. CONCLUSIONS

In this paper we have studied the analytic properties of the longitudinal quadratic response function and the related quadratic dynamical structure function for Fermi systems at arbitrary temperatures. Although the basic relations are independent of the details of the interaction we have focused an the electron liquid (a quantum OCP). For the response function we have systematically established single frequency moment and double frequency moment sum rules, which bear some resemblance to the linear f-sum rule. The analytic properties of the response function make the frequency moments sensitive to the order of integration. We have shown that the coefficients of the high frequency expansion of the real part of the response function can be expressed in terms of its own frequency moments. This is in marked contrast to what happens in the linear case where the similar expansion links the real part to the moments of the imaginary part. For the quadratic dynamical structure function we have defined weighted frequency moments and have used the quadratic fluctuation-dissipation theorem to relate them to the frequency moments of the density response function. In the high temperature classical limit these weighted moments reduce to straight frequency moments that agree with the moments obtained by the straightforward extension of the Kubo procedure to the three point correlations.

It is not within the scope of this paper to exploit possible applications of the newly established sum rules. In view of the increasing interest in the use of nonlinear response functions and multipoint correlations in the description of many body systems there is little doubt that these sum rules, similarly to their linear counterparts, will turn out to be effective theoretical tools. Thus some indication of the possible directions these applications may take seems to be useful.

We have already alluded to the intimate relationship between linear and quadratic response functions. It is via the quadratic fluctuation-dissipation theorem that one can show that quadratic response $\langle\varrho\rangle^{(2)}$, correlational linear response $\langle\varrho\varrho\rangle^{(1)}$, and equilibrium three point correlation $\langle\varrho\varrho\varrho\rangle^{(0)}$ determine each other [4]. A recently established nonperturbative scheme [11] exploits this by showing that a simple-minded approximation for the quadratic response leads to a rather sophisticated improvement in the calculation of the linear response of a classical OCP.

The use of the static quadratic response in the description of liquid and solid metals (in the effective one component approximation) has been well established at least since the seminal paper by Lloyd and Sholl [12]. It has, however, been pointed out recently by Ashcroft and collaborators [13] that the interactions induced by fluctuations require the description in terms of the dynamical quadratic response function.

In a slightly different context, in the calculation of the energy loss of a charged particle moving through a metal the contribution to the Z^3-dependent portion of the energy loss (the so-called Barkas correction, discriminating, e.g., between the energy loss of a proton and that of an antiproton) comes from quadratic response (the linear response provides only a charge-symmetric term) [14]. It is well-known that the linear contribution to the stopping power is controlled by the linear f-sum rule: in analogy, a quadratic frequency moment sum rule is expected to provide an important constraint for the first nonlinear contribution.

Another increasingly important use for nonlinear sum rules originates from the growing popularity of numerical simulations of correlated many body systems [15]. The sum rules provide one of the few exact monitoring controls for checking the accuracy and reliability of the simulation.

Contentionally, most prominently featured is the quadratic response function in the field of nonlinear optics. $\chi(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ is at the heart of sum frequency generation (SFG) and second harmonic generation (SHG, for $\omega_1=\omega_2$). Although in the infinite wave length limit ($\mathbf{q}_i=\mathbf{0}$) it takes an anisotropic medium for SHG to occur, for finite wave lengths ($\mathbf{q}_i\neq\mathbf{0}$) SHG occurs also in an isotropic system. It should be noted, however, that the description of the interaction of electromagnetic waves with a material medium requires the knowledge of more than the single longitudinal component of the full $\chi_{ijk}(\mathbf{q}_1,\omega_1;\mathbf{q}_2,\omega_2)$ response tensor. The generalization of the sum rules derived here to the remaining components needs a further extension of the analysis presented here.

As to the direct use of the multipoint functions and the nonlinear fluctuation-dissipation theorem, it has been pointed out recently by Mukamel *et al.* [16] that the nonlinear response provides a probe for the stability matrix and the Lyapunov exponents of a chaotic system.

The application and exploitation of our sum rules in the contexts discussed above will be subject of future publications. Work on formulating an improved dynamical local field based on the quadratic response is currently in progress.

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APPENDIX A: ORDER OF INTEGRATION FOR *X* MOMENTS

As mentioned above, the order of integration in Eq. (4) is in general relevant. The moments $X_{-1,-1}$, $X_{1,-1}$ and $X_{-1,1}$ are exceptions in this regard because of their better high frequency behavior. Using the Kramers-Kronig relations and (9) one can readily verify that the order of integration can be changed for these moments. For moments of higher combined order this is not the case. To better understand this fact we examine moments of the quadratic density response for a noninteracting system, which is known explicitly:

$$\chi_0(1,2)=\sum_{\mathbf{p}}\frac{n_{\mathbf{p}}}{2}\sum_{s=\pm1}\{(\omega_1+io+a_1)^{-1}\times[(\omega_1+\omega_2+io+b)^{-1}-(\omega_1+\omega_2+io+c_1)^{-1}]+(\omega_2+io+a_2)^{-1}[(\omega_1+\omega_2+io+b)^{-1}-(\omega_1+\omega_2+io+c_2)^{-1}]\}. \tag{A1}$$

Here $o\to0^+$, $a_l=(1/2m)(sq_l^2-2\mathbf{q}_l\cdot\mathbf{p})$, $b=(1/2m)(sq^2-2\mathbf{q}\cdot\mathbf{p})$, and $c_l=(1/2m)(sq_l^2-sq_{\bar{l}}^2-2\mathbf{q}\cdot\mathbf{p})$ with $\bar{l}=1$ if $l=2$ and $\bar{l}=2$ if $l=1$. Again $\mathbf{q}_1+\mathbf{q}_2=\mathbf{q}$. The sum over $s=\pm1$ is just a means of abbreviating the expression.

Because only the real even X moments are of interest to us we first separate the real part of $\chi_0(1,2)$ by applying the relation $1/(\omega\pm io)\to P/\omega\mp i\pi\delta(\omega)$. We encounter integrals of two different types:

$$I=\mathrm{Re}\int_{-\infty}^{\infty}d\omega_2\int_{-\infty}^{\infty}d\omega_1\frac{\varphi(\omega_1,\omega_2)}{\omega_2+io+a}[(\omega_1+\omega_2+io+b)^{-1}-(\omega_1+\omega_2+io+c)^{-1}], \tag{A2}$$

$$J=\mathrm{Re}\int_{-\infty}^{\infty}d\omega_2\int_{-\infty}^{\infty}d\omega_1\frac{\varphi(\omega_1,\omega_2)}{\omega_1+io+a}[(\omega_1+\omega_2+io+b)^{-1}-(\omega_1+\omega_2+io+c)^{-1}]. \tag{A3}$$

The integral I can be calculated in a straightforward fashion:

$$\begin{aligned}I&=\mathrm{Re}\int_{-\infty}^{\infty}\frac{d\omega_2}{\omega_2+io+a}\int_{-\infty}^{\infty}d\omega_1\varphi(\omega_1,\omega_2)\times[(\omega_1+\omega_2+io+b)^{-1}-(\omega_1+\omega_2+io+c)^{-1}]\\&=P\int_{-\infty}^{\infty}\frac{d\omega_2}{\omega_2+a}P\int_{-\infty}^{\infty}d\omega_1\left[\frac{\varphi(\omega_1,\omega_2)}{\omega_1+\omega_2+b}-\frac{\varphi(\omega_1,\omega_2)}{\omega_1+\omega_2+c}\right]\\&\quad-\pi^2[\varphi(a-b,-a)-\varphi(a-c,-a)].\end{aligned} \tag{A4}$$

The π^2 term in the last line provides the difference between I and J. To see this observe that to be able to apply $1/(\omega\pm io)\to P/\omega\mp i\pi\delta(\omega)$ in (A3) we first have to use partial fraction decomposition to eliminate the double pole. As a result the π^2 terms of the two contributions cancel out.

$$\begin{aligned}J&=\mathrm{Re}\int_{-\infty}^{\infty}d\omega_2\int_{-\infty}^{\infty}d\omega_1\bigg\{\frac{\varphi(\omega_1,\omega_2)}{\omega_2+io+b-a}\times[(\omega_1+io+a)^{-1}-(\omega_1+\omega_2+io+b)^{-1}]\\&\quad-\frac{\varphi(\omega_1,\omega_2)}{\omega_2+io+c-a}[-(\omega_1+io+a)^{-1}+(\omega_1+\omega_2+io+c)^{-1}]\bigg\}\\&=P\int_{-\infty}^{\infty}d\omega_2P\int_{-\infty}^{\infty}d\omega_1\frac{\varphi(\omega_1,\omega_2)}{\omega_1+a}\times[(\omega_1+\omega_2+b)^{-1}-(\omega_1+\omega_2+c)^{-1}].\end{aligned} \tag{A5}$$

Interchanging the order of integration in the integrals (A2) and (A3) also changes the type of the integral. Therefore, depending on the order of integration we do or do not encounter the π^2 term.

It is important to realize that—although these are double-principal-value integrals—the Poincaré-Bertrand theorem does not apply here because with $\varphi(\omega_1,\omega_2)=\omega_1^{r_1}\omega_2^{r_2}$ and $r_1,r_2>0$ the integrands under consideration do not unconditionally satisfy the required convergence condition [17]. In fact, for the $X_{1,1}$, $X_{0,2}$, $X_{2,0}$, and the $X_{1,3}$-moments of $\chi_0(1,2)$ one finds that the double-principal-value integrals all vanish and the π^2 term alone contributes to the moment. Using (A2) and (A3) we can now calculate the δX's directly for the $r_1+r_2=2$ cases:

$$\int \frac{d\omega_2}{2\pi}\int \frac{d\omega_1}{2\pi}\omega_1^{r_1}\omega_2^{r_2}\chi_0'(a,b) = \int \frac{d\omega_1}{2\pi}\int \frac{d\omega_2}{2\pi}\omega_1^{r_1}\omega_2^{r_2}\chi_0'(a,b)+\delta X_{r_1,r_2}. \tag{A6}$$

Interestingly, the $\delta X_{r_1,r_2}$ turn out to be identical for $(a,b)=(1,2)$, (0,1), and (0,2):

$$\delta X_{1,1}=\frac{n}{2}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}\frac{q_1^2-q_2^2}{2m}, \tag{A7a}$$

$$\delta X_{2,0}=n\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}, \tag{A7b}$$

$$\delta X_{0,2}=n\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{2m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}. \tag{A7c}$$

APPENDIX B: FREQUENCY MOMENTS OF δS

Based on the definition of Δ_{r_1,r_2} in Eq. (48) it is straightforward to calculate the following moments:

$$\Delta_{1,0}=\frac{2}{n\beta}\{X_{1,-1}[0,2]+X_{-1,1}[0,2]+X_{-1,1}[0,1]+X_{1,-1}[1,2]\} = \frac{\mathbf{q}_0\cdot\mathbf{q}_1}{2m}S_{\mathbf{q}_2}+\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}S_{\mathbf{q}_0}, \tag{B1}$$

$$\Delta_{0,1}=-\frac{2}{n\beta}\{X_{-1,1}[0,2]+X_{-1,1}[0,1]+X_{1,-1}[0,1]+X_{-1,1}[1,2]\} = -\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{2m}S_{\mathbf{q}_1}-\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{2m}S_{\mathbf{q}_0}, \tag{B2}$$

$$\Delta_{1,2}=\frac{2}{n\beta}\{X_{1,1}[0,2]+X_{-1,3}[0,2]+X_{1,1}[1,2]-\Gamma\}, \tag{B3}$$

$$\Delta_{2,1}=\frac{2}{n\beta}\{-X_{2,0}[0,2]-3X_{1,1}[0,2]-X_{-1,3}[0,2]+X_{2,0}[0,1]+2X_{1,1}[0,1]+\delta X_{0,2}+\Gamma+X_{2,0}[1,2]-X_{1,1}[1,2]\}. \tag{B4}$$

Here once more the δX term results from the change of the order of integration. The term abbreviated with Γ occurs in both these Δ moments with opposite sign.

$$\Gamma:=\int\frac{d\omega_2}{2\pi}\int\frac{d\omega_1}{2\pi}\frac{\omega_2^3}{\omega_0}\chi'(01). \tag{B5}$$

Even though we cannot calculate the moments $X_{-1,3}$ and $X_{3,-1}$, using the fact that $X_{2,0}[1,2]=X_{2,0}[1,0]$, $X_{2,0}[0,1]=X_{2,0}[0,2]$, and $X_{1,1}[1,2]=X_{1,1}[0,2]$ we can obtain the sum of $\Delta_{1,2}$ and $\Delta_{2,1}$:

$$\Delta_{1,2}+\Delta_{2,1}=\frac{2}{n\beta}\{2X_{1,1}[0,1]-2X_{1,1}[0,2]+X_{2,0}[1,2]+\delta X_{0,2}\} = \beta\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m}\frac{(q_2^2-q_1^2)}{\beta m}. \tag{B6}$$

This result allows one to cross check the $M_{2,2}$ moment as the classical limit of either $Z_{2,0}$ or $Z_{0,2}$, Eqs. (52) and (54), respectively. Furthermore it enables us to verify classical limits, by matching the sum of the moments $M_{2,2}$ and $M_{3,1}$ from Eqs. (53) and (54) with the direct classical result Eq. (C5).

APPENDIX C: CALCULATION OF CLASSICAL FREQUENCY MOMENTS

For calculating mixed frequency moments of $S_{classic}(012)$ we use the Kubo procedure which is based on the relation

$$M_{r_1,r_2}=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1^{r_1}\omega_2^{r_2}S_{classic}(012) = (i)^{r_1+r_2}\left.\frac{\partial^{r_1+r_2}S_{classic}(012)}{\partial t_1{}^{r_1}\partial t_2{}^{r_2}}\right|_{t_1=t_2=0}. \tag{C1}$$

Only the $r_1+r_2=$ *even* terms contribute. The case $r_1=r_2=0$ just gives the static quadratic structure function. The $r_1=r_2=1$ case was recovered in Eq. (49). For $r_1=2$, $r_2=0$ one finds

$$M_{2,0}=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1{}^2S_{classic}(012) = -\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m}S_{\mathbf{q}_0}-\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{\beta m}S_{\mathbf{q}_2}. \tag{C2}$$

In contrast to χ, $S_{classic}(abc)$ is symmetric in all three arguments. Therefore, $\omega_0^{r_0}\omega_1^{r_1}$ or $\omega_0^{r_0}\omega_2^{r_2}$ moments can be obtained by the rotation of the arguments. Furthermore, since $\omega_0^2=\omega_1^2+2\omega_1\omega_2+\omega_2^2$ and $\omega_0\omega_1=-\omega_1^2-\omega_1\omega_2$ all the second order moments of $S_{classic}$ are connected and can be easily related to each other [cf. Eqs. (55) and (56)]. For all $r_1+r_2=2$ cases the classical quadratic dynamic structure functions sum up to linear static structure functions.

For the fourth order moments of $S_{classic}(012)$ the results are considerably more involved. The simplest is the $r_1=r_2=2$ moment, given by:

$$M_{2,2}=\int_{-\infty}^{\infty}\frac{d\omega_2}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\omega_1{}^2\omega_2{}^2 S_{classic}(012)$$

$$=\left(i\frac{\partial}{\partial t_1}\right)^2\left(i\frac{\partial}{\partial t_2}\right)^2\frac{1}{N}\langle\varrho_{\mathbf{q}_0}(t_0)\varrho_{\mathbf{q}_1}(t_1)\varrho_{\mathbf{q}_2}(t_2)\rangle^{(0)}\Big|_{t_1=t_2=0}$$

$$=\frac{1}{N}\langle\varrho_{\mathbf{q}_0}(0)\ddot{\varrho}_{\mathbf{q}_1}(0)\ddot{\varrho}_{\mathbf{q}_2}(0)\rangle^{(0)}. \tag{C3}$$

For the 3-1 and 4-0 moments the higher time derivatives can be avoided by shifting the time argument under the equilibrium average before taking a particular derivative. For instance, the $r_1=3$, $r_2=1$ classical moment can then be written as:

$$M_{3,1}=\left(i\frac{\partial}{\partial t_1}\right)\frac{1}{N}\langle\varrho_{\mathbf{q}_0}(-t_1)\ddot{\varrho}_{\mathbf{q}_1}(0)\dot{\varrho}_{\mathbf{q}_2}(t_2-t_1)\rangle^{(0)}|_{t_1=t_2=0}$$

$$=-\frac{1}{N}[\langle\dot{\varrho}_{\mathbf{q}_0}(0)\ddot{\varrho}_{\mathbf{q}_1}(0)\dot{\varrho}_{\mathbf{q}_2}(0)\rangle^{(0)}$$

$$+\langle\varrho_{\mathbf{q}_0}(0)\ddot{\varrho}_{\mathbf{q}_1}(0)\ddot{\varrho}_{\mathbf{q}_2}(0)\rangle^{(0)}]. \tag{C4}$$

In addition to $\langle\varrho\ddot{\varrho}\ddot{\varrho}\rangle^{(0)}$ a term $\langle\dot{\varrho}\ddot{\varrho}\dot{\varrho}\rangle^{(0)}$ comes up. For the other fourth order moments we also have index permutations of these two, such as $\langle\ddot{\varrho}\varrho\ddot{\varrho}\rangle^{(0)}$.

The two correlations can be calculated by using Hamilton's equations to replace the time derivatives and then integrating by part. We find that $\langle\dot{\varrho}\ddot{\varrho}\dot{\varrho}\rangle^{(0)}$ leads to a quite simple expression:

$$M_{3,1}+M_{2,2}=-\frac{1}{N}\langle\dot{\varrho}_{\mathbf{q}_0}(0)\ddot{\varrho}_{\mathbf{q}_1}(0)\dot{\varrho}_{\mathbf{q}_2}(0)\rangle^{(0)}$$

$$=-\frac{1}{N}\sum_{i,j,l}\int d\Gamma e^{-\beta H}(\mathbf{q}_0\cdot\mathbf{x}_i)$$

$$\times[(\mathbf{q}_1\cdot\dot{\mathbf{x}}_j)^2+i(\mathbf{q}_1\cdot\ddot{\mathbf{x}}_j)](\mathbf{q}_2\cdot\dot{\mathbf{x}}_l)$$

$$\times e^{-i(\mathbf{q}_0\cdot\mathbf{x}_i+\mathbf{q}_1\cdot\mathbf{x}_j+\mathbf{q}_2\cdot\mathbf{x}_l)}$$

$$=-2\frac{\mathbf{q}_0\cdot\mathbf{q}_1}{\beta m}\frac{\mathbf{q}_1\cdot\mathbf{q}_2}{\beta m}-\frac{\mathbf{q}_0\cdot\mathbf{q}_2}{\beta m}\frac{q_1{}^2}{\beta m}. \tag{C5}$$

Bearing in mind that through the continuity equation $\dot{\varrho}$ is related to the particle velocity and that the velocities of two classical particles are not correlated, it is indeed expected that Eq. (C5) be correlation independent. On the other hand, $\ddot{\varrho}$ is linked to the equation of motion and thus contains a force term. In $M_{2,2}$ the Coulomb interaction between particles gives rise to force-kinetic energy and force-force correlation in $\langle\varrho\ddot{\varrho}\ddot{\varrho}\rangle^{(0)}$.

$$M_{2,2}=\frac{1}{N}\sum_{i,j,l}\int d\Gamma e^{-\beta H}e^{-i(\mathbf{q}_0\cdot\mathbf{x}_i+\mathbf{q}_1\cdot\mathbf{x}_j+\mathbf{q}_2\cdot\mathbf{x}_l)}$$

$$\times\{(\mathbf{q}_1\cdot\dot{\mathbf{x}}_j)^2(\mathbf{q}_2\cdot\mathbf{x}_l)^2-(\mathbf{q}_1\cdot\ddot{\mathbf{x}}_j)(\mathbf{q}_2\cdot\ddot{\mathbf{x}}_l)$$

$$+i(\mathbf{q}_1\cdot\dot{\mathbf{x}}_j)^2(\mathbf{q}_2\cdot\ddot{\mathbf{x}}_l)+i(\mathbf{q}_2\cdot\dot{\mathbf{x}}_l)^2(\mathbf{q}_1\cdot\ddot{\mathbf{x}}_j)\}$$

$$=\frac{1}{\beta^2m^2}\Bigg\{q_1^2q_2^2+2(\mathbf{q}_1\cdot\mathbf{q}_2)^2+ng_{\mathbf{q}_0}[(\mathbf{q}_1\cdot\mathbf{q}_2)$$

$$\times(\mathbf{q}_1\cdot\mathbf{q}_2-2q_1^2-2q_2^2)-2q_1^2q_2^2]+2n$$

$$\times(\mathbf{q}_1\cdot\mathbf{q}_2)[g_{\mathbf{q}_1}q_1^2+g_{\mathbf{q}_2}q_2^2]-2n^2h_{\mathbf{q}_1,\mathbf{q}_2}q_1^2q_2^2$$

$$+\frac{\beta}{V}\sum_{\mathbf{p}}(\mathbf{q}_1\cdot\mathbf{p})(\mathbf{q}_2\cdot\mathbf{p})\phi_{\mathbf{p}}[S_{\mathbf{q}_1+\mathbf{p},\mathbf{q}_2-\mathbf{p}}-S_{\mathbf{q}_1+\mathbf{q}_2-\mathbf{p},\mathbf{p}}]\Bigg\}. \tag{C6}$$

Here $\phi_{\mathbf{p}}=4\pi e^2/p^2$ and $g_{\mathbf{q}_i}$ and $h_{\mathbf{q}_1,\mathbf{q}_2}$ are the Fourier transforms of the pair and triplet correlation functions. The last term in (C6) is the analog of the third frequency moment coefficient for the linear dynamical structure function, $(\beta/V)\Sigma_{\mathbf{p}}(\mathbf{q}\cdot\mathbf{p})^2\phi_{\mathbf{p}}\{S_{\mathbf{q}-\mathbf{p}}-S_{\mathbf{p}}\}$: similarly to this coefficient the two contributions can be interpreted in terms of the forces acting on a particle oscillating in a local potential well generated by the environment of other particles [18]. $M_{3,1}$ can be obtained from Eqs. (C5) and (C6).

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