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Quadratic fluctuation-dissipation theorem: The quantum domain

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A derivation of a general relationship between the frequency- and wave-number-dependent longitudinal quadratic response functions and the three-point dynamical structure function (the Fourier transform of the equilibrium three-point density-density correlations) is presented. This relationship is the full quadratic equivalent, valid for quantum systems of arbitrary degeneracy, of the customary (linear) fluctuation-dissipation theorem. The high-temperature limit reproduces earlier classical results, while its zero-temperature limit is characterized by a piecewise analytic behavior in the $(\omega_1\omega_2)$ plane, generated by the respective dominance of the differently ordered triplets of density operators. There are two main versions of the theorem. In the first the completely symmetrized combination of the three-point functions is expressed in terms on a "triangle-symmetrized" [in the $(\mathbf{k}\omega)$ space] combination of the response functions. In the second "inverted" form, each of the six differently ordered three-point functions is expressed in terms of combinations of response functions, where the triangle symmetry is broken.

I. INTRODUCTION

The relationships commonly known as "fluctuation-dissipation theorems" (FDT) establishing a link between the linear response (a nonequilibrium property) of the system and equilibrium correlations of fluctuating quantities, have become a powerful tool in modern statistical physics and many-body theory. The primitive idea was due to Nyquist,[1] who studied the relationship between the resistivity and noise of electrical networks. About two decades later Callen and Welton[2] generalized Nyquist's observation to a number of physical systems and conjectured the existence of an underlying general physical principle. Callen and Welton were also the first to use the phrase fluctuation-dissipation theorem in their paper.[2] The establishment of the FDT in its modern form is, however, due to Kubo.[3]

While the Callen-Welton-Kubo formalism focuses on the linear-response of the system, it is clear that, in general, the system's response is not restricted to be linear. Thus, in addition to the well-explored linear-response functions, one can examine the properties of higher-order (quadratic, cubic, etc.) response functions, which relate the system's response to higher powers of the perturbing field. Moreover, once one relaxes the restriction of concentrating on the simplest response characteristics of the system, the very concept of "response" can be generalized.

The conventional response functions, to which we will refer as "response functions of the first kind," relate the perturbed averages of physical quantities (density, current, etc.) at a given space-time point to the perturbing field. The effect of the perturbation on the system is, however, further characterized by the perturbation of averages of correlated physical quantities taken at two, three, etc., space-time points. The relationships between these perturbed two-, three-, etc., point functions, which also can exhibit both linear and higher-order behavior, and the perturbing field define[4] "response functions of the second kind," "response functions of the third kind," etc. That all these higher-order response functions, and also the response functions of higher kind, would satisfy some kind of fluctuation-dissipation-like theorem, i.e., should be related to averages of equilibrium correlations, is a rather obvious expectation. Even a cursory reflection over the derivation of the linear FDT should suggest a correlation, e.g., between the quadratic response function or the linear-response function of the second kind, on the one hand, and the equilibrium three-point function, on the other.

Over the last 15 years, a series of quadratic FDT's has been established along these lines. The basic relationship between the quadratic conductivity and the three-point current-current correlations for a one-component classical plasma was derived by Golden, Kalman, and Silevitch[5–7] (GKS) and independently by Sitenko.[8] Relationships for the current-current response function of the second kind were given by Golden and Kalman.[9,10] Generalizations to the surface layer of electron gas and to binary ionic mixtures were derived by Golden and Lu in Ref. 11 and Ref. 12, respectively. Apart from providing a satisfactory extension of the entire concept of the fluctuation-dissipation relations, the formalism of the nonlinear FDT's has turned out to lend itself to formulating useful approximation schemes for strongly coupled classical Coulomb systems.[9,10,12–15] The hierarchy of linear, quadratic, etc., FDT's also generates a convenient alternative to establishing perturbation schemes. This is due to the "order-raising" property of the FDT's: if an expansion in the plasma parameter γ $[\gamma=(4\pi e^2 n\beta)^{3/2}/4\pi n]$ exists, a response function of rank m ($m=1$ linear, $m=2$ quadratic, etc.) relates to an $(m+1)$-point function by the latter being of $O(\gamma^{m+n})$, if the former is $O(\gamma^n)$.

A different line of research on the nonlinear properties of fluctuations of quantum-mechanical systems has been pursued by Soviet workers. The works of Efremov,[16,17] of Stratonovich,[18–20] and of Bochkov and Kozlov[21] aimed at establishing relationships of various orders of nonlinearity between space-time representations of fluctuating quantities.

The present paper, and others representing a sequel to it, address themselves to the problem of establishing a quadratic FDT relation for quantum (for a preliminary account, see Ref. 22) systems of arbitrary degeneracy. Our goals and results overlap, to some extent, with those of Efremov[17] and Stratonovich.[20]

Nevertheless both our approach and our conclusions are different. Our derivation is representation free and follows the singular integral equation method established by GKS.[5] In our results, in contrast to Refs. 16–21, we focus on relationships in momentum-energy $(\mathbf{k},\omega)$ representation between precisely defined response functions and three-point current-current-current or density-density-density correlations.

A digression on a point on which much attention has been focused in connection with the very idea of quadratic—and higher-order—responses, may be in order. A quadratic perturbation necessarily generates a secular contribution in the response of the system. The consequences of this are twofold. First, the manifestation of the secular time growth in the ω representation is a divergence of certain response functions and of the related three-point correlation functions at $\omega=0$. Second, the heat generated by the perturbation also accumulates in time and by raising the temperature it destroys the isothermal character of the system. In their instructive and illuminating paper Tremblay, Patton, Martin, and Maldague (TPMM) (Ref. 23) demonstrated (on a specific model) how these two effects originate from the same source. They also showed that even though the system can be kept at a stationary temperature by coupling it to a heat bath, the way this is done affects the response of the system and therefore, in this sense, quadratic response functions are not uniquely defined.

Important as these aspects of the nonlinear response theory are, they do not have a significant bearing on the analysis presented in this paper (or on earlier studies written in a similar vein[5,20,21]) neither are they, of course, specifically related to the quantum character of the system. This work focuses on relationships between frequency- and wave-number-dependent response functions and correlation functions. These relationships are problem free for all frequencies, except for a possible divergence at $\omega=0$. Thus the relationships derived are certainly valid and unaffected at finite ω values. Actually, a pathological behavior at $\omega=0$ is not a unique feature of the nonlinear responses: the linear conductivity, for example, also exhibits (admittedly, for different physical reasons) a divergence at $\omega=0$ in the RPA. This, however, does not affect the validity and usefulness of the linear FDT, even in the approximation characterized by the random-phase approximation (RPA).

The (rather trivial) answer to the problem of heating is to try to live with the secular terms and to restrict the analysis of the system to "sufficiently short" times. The real question is whether the condition of the time of the analysis being sufficiently short can be met. In this regard, one has to carefully distinguish between two aspects of the problem. The formal question is whether the heating time ($\approx T/\dot{T}$) is sufficiently longer than any other characteristic time of the system and thus whether the "momentary thermal equilibrium" model, implicit in the nonequilibrium perturbation calculation, is a reasonable assumption. For the experimentalist, however, the question that poses itself is whether in a given experimental or observational situation the time of the observation is short enough to justify the same assumption. If this is not the case, the mechanism of cooling becomes of crucial importance indeed, and the question as to the precise nature of the fluctuations and responses one contemplates to measure has to be addressed with extreme care. The guidance provided by TPMM in this respect is again very valuable.

This paper addresses only the formal aspect of the problem. Thus one has to be satisfied that the first weaker condition is met. This, in principle, depends on the strength of the perturbing field, but the reader can easily convince himself that for any reasonable model and for any reasonable value of the perturbing field, the macroscopic heating time is many orders of magnitudes longer than any of the characteristic microscopic times, such as those related to collective excitations, decay of correlations, and the like. TPMM provides more detailed analysis and numerical examples for this.

The model we adopt in this paper is that of a one-component plasma, although generalization to other situations should not present any difficulty. The derivation emulates the classical approach of GKS;[5] it nevertheless deviates from the classical pattern, primarily because of the noncommutability of the different density (or current) operators at displaced arguments, leading to the appearance of six differently ordered three-point functions. In Sec. II the quantum Liouville equation for the statistical operator Ω is solved to second order in the perturbing scalar potential, and the second-order average current is calculated. The result is the primitive form of the FDT, expressing a certain combination of the three-point functions in terms of the quadratic external conductivity as the solution of a singular integral equation. In Sec. III, the primitive form undergoes a series of transformations which ultimately lead to an algebraic relationship between the symmetrized real part of the conductivity [consisting of three pieces, exhibiting a "triangle symmetry" in the $(\mathbf{k}\omega)$ space], and the fully symmetrized three-point function.

In Sec. IV we present an "inversion" of the FDT relation, i.e., we express each of the six three-point functions in terms of a (rather involved) combination of the response functions. This inversion—which is certainly not a trivial consequence of the primitive form of the FDT—makes possible the further applications of the theorem which will be discussed in subsequent publications. The rest of the paper is devoted to examining various limits of the general expression. In Secs. V and VI, the zero-temperature limit and the high-temperature classical limit are calculated, respectively. The interesting structural feature of the former is the piecewise analytic behavior of the three-point functions in different domains in the plane of the perturbing frequencies ω_1, ω_2. The latter, as expected, reproduces the earlier results of GKS. Finally, in Sec. VI, the static limit of the QFDT is investigated: similar to the case of linear FDT and to the classical QFDT, an explicit expression for the static structure function in terms of the quadratic response function can be found.

II. PRELIMINARIES

Consider an equilibrium system described by the Hamiltonian $H^{(0)}$ and by the canonical statistical operator

$$\Omega^{(0)}=Z^{-1}e^{-\beta H^{(0)}} . \tag{1}$$

We are interested in the second-order response of the system under the effect of an external perturbation $\hat{\phi}$,

$$H^{(1)}=\frac{1}{2V}\sum_{\mathbf{k}}[\hat{\phi}_{\mathbf{k}}(t)n_{\mathbf{k}}^{\dagger}+\hat{\phi}_{\mathbf{k}}^{*}(t)n_{\mathbf{k}}]=\frac{1}{V}\sum_{\mathbf{k}}\hat{\phi}_{\mathbf{k}}(t)n_{\mathbf{k}}^{\dagger} , \tag{2}$$

where $n_{\mathbf{k}}$ is the Fourier transform of the local density operator.

The total Hamiltonian $H^{(0)}+H^{(1)}$ generates the Liouville operator

$$\mathcal{L}\equiv\frac{1}{\hbar}[H, \cdots] ,$$
$$\mathcal{L}\equiv\mathcal{L}^{(0)}+\mathcal{L}^{(1)} . \tag{3}$$

The time evolution of the system is conveniently described with the aid of the Liouville equation for the perturbed statistical operator

$$\frac{\partial\Omega}{\partial t}+i\mathcal{L}\Omega=0 . \tag{4}$$

This latter can be expanded in the external perturbation

$$\Omega=\Omega^{(0)}+\Omega^{(1)}+\Omega^{(2)}+ \cdots . \tag{5}$$

The quadratic longitudinal external conductivity $\hat{\sigma}_2$, however, relates the average second-order current density

$$\langle j_{\mathbf{k}}\rangle^{(2)}(t)=\mathrm{Tr}[\Omega^{(2)}(t)j_{\mathbf{k}}] \tag{6}$$

to the external perturbing field $\hat{E}$ through Ohm's law

$$\langle j_{\mathbf{k}}\rangle^{(2)}(t)=\frac{1}{V}\sum_{\mathbf{k}_1\mathbf{k}_2}\int_{-\infty}^{+\infty}d\tau_1\int_{-\infty}^{+\infty}d\tau_2\,\hat{\sigma}(\mathbf{k}_1,\tau_1;\mathbf{k}_2\tau_2)\hat{E}_{\mathbf{k}_1}(t-\tau_1)\hat{E}_{\mathbf{k}_2}(t-\tau_2) . \tag{7}$$

The formal solution to the perturbed statistical operator is

$$\Omega^{(1)}(t)=-i\int_{-\infty}^{t}dt_1\,U(t,t_1)\mathcal{L}^{(1)}(t_1)\Omega^{(0)} , \tag{8}$$

$$\begin{aligned}\Omega^{(2)}(t)&=(-i)^2\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\,U(t,t_1)\mathcal{L}^{(1)}(t_1)U(t_1,t_2)\mathcal{L}^{(1)}(t_2)\Omega^{(0)}\\&=-\frac{1}{\hbar^2V^2}\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\sum_{\mathbf{k}_1\mathbf{k}_2}U(t-t_1)[n_{-\mathbf{k}_1}(t), U(t_1-t_2)[n_{-\mathbf{k}_2}(t), \Omega^{(0)}]]\hat{\phi}_{\mathbf{k}_1}(t_1)\hat{\Phi}_{\mathbf{k}_2}(t_2)\end{aligned} \tag{9}$$

with

$$U(t,t_1)=e^{-i\mathcal{L}^{0}(t-t_1)}=U(t-t_1) \tag{10}$$

being the time evolution operator.

A transformation of (9) can be accomplished through a number of steps. First we use the identity

$$[x,yz]=y[x,z]+[x,y]z \tag{11}$$

and shift the operators $n_{-\mathbf{k}_1}$, $n_{-\mathbf{k}_2}$ from the arbitrary reference time t to t_1 and t_2. Next we note that the application of the Liouville operator $\mathcal{L}^{(1)}$ leads to the commutator $[n_{\mathbf{k}}, e^{-\beta H^0}]$, which can be evaluated with the aid of the Hausdorff-Campbell[24,25] formula, discussed in Appendix A. The result is

$$[n_{\mathbf{k}},e^{-\beta H^0}]=-i\beta\hbar e^{-\beta H^0}\psi\left[-i\beta\hbar\frac{d}{dt}\right]\frac{d}{dt}n_{\mathbf{k}} \tag{12}$$

with

$$\psi(x)=(e^{x}-1)/x . \tag{13}$$

Finally, employing Eq. (12), Eq. (9) is simplified as

$$\Omega^{(2)}(t)=-\frac{1}{V^2}\frac{1}{\hbar^2}\sum_{\mathbf{k}_1\mathbf{k}_2}\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\{n_{-\mathbf{k}_1}(t_1),[n_{-\mathbf{k}_2}(t_2),\Omega^{(0)}]\}\hat{\Phi}_{\mathbf{k}_1}(t_1)\hat{\Phi}_{\mathbf{k}_2}(t_2)$$

$$=-\frac{\Omega^{(0)}}{V^2}\sum_{\mathbf{k}_1\mathbf{k}_2}\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\left\{\frac{\beta}{\hbar}k_2\int\frac{d\omega_2}{2\pi}\psi(\beta\hbar\omega_2)[n_{-\mathbf{k}_1}(t_1),j_{-\mathbf{k}_2}(-\omega_2)e^{i\omega_2t_2}]+\beta^2k_1k_2\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\psi(\beta\hbar\omega_1)\psi(\beta\hbar\omega_2)\times e^{i(\omega_1t_1+\omega_2t_2)}j_{-\mathbf{k}_1}(-\omega_1)j_{-\mathbf{k}_2}(-\omega_2)\right\}\hat{\Phi}_{\mathbf{k}_1}(t_1)\hat{\Phi}_{\mathbf{k}_2}(t_2)\ . \qquad (14)$$

According to Eqs. (6) and (14), $\langle j_{\mathbf{k}}\rangle^{(2)}(t)$ now becomes

$$\langle j_{\mathbf{k}}\rangle^{(2)}(t)=-\frac{1}{V^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}\left[\int_{-\infty}^{t}dt_1\int_{-\infty}^{t_1}dt_2\left\{\frac{\beta k_2}{\hbar}\int\frac{d\omega_2}{2\pi}\psi(\beta\hbar\omega_2)\langle[n_{-\mathbf{k}_1}(t_1),j_{-\mathbf{k}_2}(-\omega_2)e^{i\omega_2t_2}]j_{\mathbf{k}}(t)\rangle+\beta^2k_1k_2\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\psi(\beta\hbar\omega_1)\psi(\beta\hbar\omega_2)\times\langle e^{i(\omega_1t_1+\omega_2t_2)}j_{-\mathbf{k}_1}(-\omega_1)j_{-\mathbf{k}_2}(-\omega_2)j_{\mathbf{k}}(t)\rangle\right\}\right]\times\hat{\Phi}_{\mathbf{k}_1}(t_1)\hat{\Phi}_{\mathbf{k}_2}(t_2)\ . \qquad (15)$$

The asymmetry in the time variables t_1 and t_2 in (15) can be removed by inverting the order of integration, interchanging the variables $(\mathbf{k}_1,t_1,\omega_1)$ and $(\mathbf{k}_2,t_2,\omega_2)$, and then symmetrizing the resulting expression. In addition, the time variables of the averaged products can be shifted as long as their time differences are preserved. We now introduce a notation for the crucially important three-point current correlations and three-point density correlations,

$$\frac{1}{2\pi}\langle n_{\mathbf{k}_1}(\omega_1)n_{\mathbf{k}_2}(\omega_2)n_{-\mathbf{k}}(-\omega)\rangle=N\delta_{\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2}\delta(\omega-\omega_1-\omega_2)\{S(120)+N[\delta_{\mathbf{k}_1}\delta(\omega_1)S(\mathbf{k}_2\omega_2)+\delta_{\mathbf{k}_2}\delta(\omega_2)S(\mathbf{k}_1\omega_1)+\delta_{\mathbf{k}}\delta(\omega)S(\mathbf{k}_1\omega_1)]+N^2\delta_{\mathbf{k}_1}\delta_{\mathbf{k}_2}\delta(\omega_1)\delta(\omega_2)\}\ , \qquad (16a)$$

with

$$\frac{1}{2\pi}\langle n_{\mathbf{k}_1}(\omega_1)n_{-\mathbf{k}_2}(-\omega_2)\rangle=N\delta_{\mathbf{k}_1-\mathbf{k}_2}\delta(\omega_1-\omega_2)[S(\mathbf{k}_1,\omega_1)+N\delta_{\mathbf{k}_1}\delta(\omega_1)]$$

and

$$\frac{1}{2\pi}\langle j_{\mathbf{k}_1}(\omega_1)j_{\mathbf{k}_2}(\omega_2)j_{-\mathbf{k}}(-\omega)\rangle=N\delta_{\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2}\delta(\omega-\omega_1-\omega_2)Q(120)\ . \qquad (16b)$$

If one exploits the relations between cycles for the S or Q functions, as discussed in Appendix C, such as

$$\langle j_{-\mathbf{k}_1}(-\omega_1)j_{-\mathbf{k}_2}(-\omega_2)j_{\mathbf{k}}(\omega)\rangle=\langle j_{-\mathbf{k}}(-\omega)j_{\mathbf{k}_2}(\omega_2)j_{\mathbf{k}_1}(\omega_1)\rangle\ ,$$

the quadratic conductivity can be written as

$$\hat{\sigma}(\mathbf{k}_1,\tau_1;\mathbf{k}_2,\tau_2)=\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_2-\tau_1)\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1\omega_2}(e^{\beta\hbar\omega_2}-1)[Q(021)-Q(012)]$$
$$+\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_1-\tau_2)\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1\omega_2}(e^{\beta\hbar\omega_1}-1)[Q(012)-Q(021)]$$
$$+\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1\omega_2}(e^{\beta\hbar\omega_1}-1)$$
$$\times(e^{\beta\hbar\omega_2}-1)[\Theta(\tau_1-\tau_2)Q(012)+\Theta(\tau_2-\tau_1)Q(021)]\ . \tag{17}$$

However, the latter can be formulated in a simpler way by observing relations that exist among Q functions with differently ordered arguments. As discussed in Appendix B, one can shift frequency arguments within a cycle with the aid of the relationship [where (abc) stands for any combination of the arguments $(\mathbf{k}_1\omega_1)$, $(\mathbf{k}_2\omega_2)$, $(\mathbf{k}_0\omega_0)$, with $(\mathbf{k}_0\omega_0)\rightarrow(-\mathbf{k},-\omega)$]

$$e^{-\beta\hbar\omega_a}Q(abc)=Q(bca)\ . \tag{18}$$

With these relations in mind, Eq. (17) becomes

$$\hat{\sigma}(\mathbf{k}_1,\tau_1;\mathbf{k}_2,\tau_2)$$
$$=\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_2-\tau_1)\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1\omega_2}\{[Q(012)+Q(210)]-[Q(102)+Q(201)]\}$$
$$+\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_1-\tau_2)\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1\omega_2}\{[Q(021)+Q(120)]-[Q(102)+Q(201)]\}\ , \tag{19}$$

while its Fourier transform is

$$\hat{\sigma}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)=\frac{n}{2\hbar^2}\int d\omega_1'\int d\omega_2'\,\delta_+(\omega_2-\omega_2')\delta_+(\omega_1+\omega_2-\omega_1'-\omega_2')\frac{1}{\omega_1'\omega_2'}\{[Q(012)+Q(210)]-[Q(102)+Q(201)]\}$$
$$+\frac{n}{2\hbar^2}\int d\omega_1'\int d\omega_2'\,\delta_+(\omega_1-\omega_1')\delta_+(\omega_1+\omega_2-\omega_1'-\omega_2')\frac{1}{\omega_1'\omega_2'}$$
$$\times\{[Q(021)+Q(120)]-[Q(102)+Q(201)]\}\ . \tag{20}$$

Equation (20) is the primitive form of the QFDT. Even though it provides an explicit expression for the quadratic conductivity $\hat{\sigma}_2$ in terms of the three-point current correlations, its usefulness is limited.

III. DYNAMIC FLUCTUATION-DISSIPATION THEOREMS

Equation (20) provides a link between the two principal objects, the quadratic conductivity $\hat{\sigma}_2$, and the three-point current correlations, whose relationships are to be the central statement of the QFDT. Nevertheless, Eq. (20) is still not of the form that could constitute the desired formulation of the QFDT. The main reason for this is that the right-hand side (rhs) of Eq. (20) is an integral relationship which, in fact, generates an integral equation for the combination of the Q functions that appear under the integral. Only the solution of this integral equation would provide an explicit connection between the quantities in question. Linked to this problem, although not in an obvious way, is the structural defect of Eq. (20) which manifests itself through the lack of symmetry in the variables ω_1,ω_2, and ω in the projection operation generated by the δ_+ functions. The origin of this lack of symmetry can be traced back to the entirely different symmetry properties with respect to these variables of $\hat{\sigma}_2$ on the one hand, and of the Q functions on the other. $\hat{\sigma}(\mathbf{k}_1,\omega_1;\mathbf{k}_2\omega_2)$ is symmetric in the pairs $(\mathbf{k}_1,\omega_1)$ and $(\mathbf{k}_2,\omega_2)$ [but not in $(\mathbf{k},\omega)$], while the Q functions possess an intrinsic symmetry with respect to any of the three pairs, which is, however, broken by the different orderings of the arguments of Q's (but is completely restored in the classical limit). Thus to arrive at the desired form of the FDT we proceed to eliminate the asymmetry. This can be accomplished in two stages. First, a particular symmetrized combination of Eq. (19) is constructed so as to generate a completely symmetric kernel of the integral equation for the three-point functions. In the second stage the asymmetry in the frequency arguments of the conductivity is eliminated by rotating the arguments in the $(\mathbf{k},\omega)$ space along the sides of the triangle spanned by the $(\mathbf{k}_1,\omega_1)$-$(\mathbf{k}_1,\omega_1)$-$(\mathbf{k},\omega)$ triad. This operation will, at the same time, provide a solution of the integral equation.

The first step is most easily done in the time domain. We change time arguments, but preserve the response-function character of the $\hat{\sigma}$'s with the new time arguments by requiring them to vanish both for $\tau_1<0$ and $\tau_2<0$. Starting with Eq. (19), we generate

$$\Theta(\tau_1)\hat{\sigma}(\mathbf{k}_1,\tau_2-\tau_1;-\mathbf{k},\tau_2)$$
$$=\frac{-n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_2-\tau_1)\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{e^{+i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1(\omega_1+\omega_2)}\{[Q(012)+Q(210)]-[Q(120)+Q(021)]\}\ . \tag{21}$$

It is easily shown that $\hat{\sigma}$ has odd parity under spatial reflection: $[\hat{\sigma}(-\mathbf{k}_1,-\mathbf{k}_2)=-\hat{\sigma}(\mathbf{k}_1,\mathbf{k}_2)]$, and a combined $(\mathbf{k}_1,\omega_1)\rightarrow(-\mathbf{k}_1,-\omega_1)$ and $(\mathbf{k}_2,\omega_2)\rightarrow(-\mathbf{k}_2,-\omega_2)$ transformation, leads to the change of cyclic order of the arguments of the Q functions $[Q(abc)\rightarrow Q(cba)]$. Applying these operations to (21) we find

$$\Theta(\tau_1)\hat{\sigma}(\mathbf{k}_1,\tau_2-\tau_1;-\mathbf{k},\tau_2)$$
$$=\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_2-\tau_1)\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_1(\omega_1+\omega_2)}\{[Q(012)+Q(210)]-[Q(021)+Q(120)]\} \tag{22}$$

and similarly

$$\Theta(\tau_2)\hat{\sigma}(-\mathbf{k},\tau_1;\mathbf{k}_2,\tau_1-\tau_2)$$
$$=\frac{n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\Theta(\tau_1-\tau_2)\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{e^{-i(\omega_1\tau_1+\omega_2\tau_2)}}{\omega_2(\omega_1+\omega_2)}\{[Q(021)+Q(120)]-[Q(012)+Q(210)]\}\ . \tag{23}$$

We now combine (19), (22), and (23) and construct the symmetrized function

$$\Psi(\mathbf{k}_1,\tau_1;\mathbf{k}_2,\tau_2)\equiv\hat{\sigma}(\mathbf{k}_1,\tau_1;\mathbf{k}_2,\tau_2)-\Theta(\tau_1)\hat{\sigma}(\mathbf{k}_1,\tau_2-\tau_1;-\mathbf{k},\tau_2)-\Theta(\tau_2)\hat{\sigma}(-\mathbf{k},\tau_1;\mathbf{k}_2,\tau_1-\tau_2)$$
$$=\frac{-n}{2\hbar^2}\Theta(\tau_1)\Theta(\tau_2)\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}e^{-i(\omega_1\tau_1+\omega_2\tau_2)}\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}, \tag{24}$$

where $\Theta(\tau_1)$ and $\Theta(\tau_2)$ are step functions. A more useful form is obtained by expressing (24) in the Fourier domain

$$\Psi(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\equiv\hat{\sigma}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)+\int_{-\infty}^{+\infty}d\mu\,\delta_+(\omega_1-\mu)\hat{\sigma}^*(\mathbf{k}_1,\mu;-\mathbf{k},-\omega_2-\mu)$$
$$+\int_{-\infty}^{+\infty}d\mu\,\delta_+(\omega_2-\mu)\hat{\sigma}^*(\mathbf{k}_2,\mu;-\mathbf{k},-\omega_1-\mu)$$
$$=-\frac{n}{2\hbar^2}\int_{-\infty}^{+\infty}d\mu_1\int_{-\infty}^{+\infty}d\mu_2\,\delta_+(\omega_1-\mu_1)\delta_+(\omega_2-\mu_2)\underline{R}\left\{\frac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2,\mu_2)}{\mu_1\mu_2}\right\}. \tag{25a}$$

Here $\underline{R}\{\ \}$ represents full symmetrization with respect to the permutation of the arguments,

$$\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}=\frac{Q(102)+Q(201)}{\omega_1\omega_2}-\frac{Q(012)+Q(210)}{\omega\omega_2}-\frac{Q(021)+Q(120)}{\omega\omega_1}\ . \tag{25b}$$

The Q functions are real, as shown in Appendix C; thus taking the real part of (25) leads to a more explicit form of the integral equation,

$$\Psi'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=-\frac{n}{2\hbar^2}\int_{-\infty}^{+\infty}d\mu_1\int_{-\infty}^{+\infty}d\mu_2\left[\tfrac{1}{4}\delta(\omega_1-\mu_1)\delta(\omega_2-\mu_2)-\frac{1}{4\pi^2}\mathbf{P}\left[\frac{1}{(\omega_1-\mu_1)(\omega_2-\mu_2)}\right]\right]$$
$$\times\underline{R}\left\{\frac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2\mu_2)}{\mu_1\mu_2}\right\}$$
$$=-\frac{n}{8\hbar^2}\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}+\frac{n}{8\hbar^2\pi^2}\mathbf{P}\int_{-\infty}^{+\infty}d\mu_1\int_{-\infty}^{+\infty}d\mu_2\frac{\underline{R}\left\{\dfrac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2,\mu_2)}{\mu_1\mu_2}\right\}}{(\omega_1-\mu_1)(\omega_2-\mu_2)}\ . \tag{26}$$

We note in passing that the importance of the symmetrization procedure outlined above is especially clear in the classical limit[5] where the unsymmetrized kernel (22) is expressible only in terms of unwieldy Poisson brackets.

$$\underline{R}[\Psi(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]\equiv\Psi(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)+\Psi(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)+\Psi(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)\ , \tag{27}$$

which now does possess the "triangle symmetry." Note that $\underline{R}[\ ]$ acts on a function already symmetric in (1,2) and therefore is different (by a factor of $\frac{1}{2}$) from $\underline{R}\{\ \}$. In contrast to the previous definition [Eq. (24)] $\underline{R}[\ ]$, acting on a function already symmetric in its (1,2) arguments, generates only a rotation on the $(\mathbf{k}_1,\omega_1)$, $(\mathbf{k}_2,\omega_2)$, $(-\mathbf{k},-\omega)$ triangle.

Noting the fact that the real part of σ_2 has odd parity and the imaginary part has even parity with respect to the simultaneous sign reversals of its frequency arguments, we obtain from (25)

$$\begin{aligned}\Psi'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=&\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,-\omega_1;-\mathbf{k},\omega)-\tfrac{1}{2}\hat{\sigma}'(-\mathbf{k},\omega;\mathbf{k}_2,-\omega_2)\\&+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\omega_1+\mu}\hat{\sigma}''(\mathbf{k}_1,\mu;-\mathbf{k},\omega_2-\mu)+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\omega_2+\mu}\hat{\sigma}''(-\mathbf{k},\omega_1-\mu;\mathbf{k}_2,\mu)\\=&\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,-\omega_1;-\mathbf{k},\omega)-\tfrac{1}{2}\hat{\sigma}'(-\mathbf{k},\omega;\mathbf{k}_2,-\omega_2)\\&+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\omega-\mu}\hat{\sigma}''(\mathbf{k}_1,\omega_2-\mu;-\mathbf{k},\mu)+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\omega-\mu}\hat{\sigma}''(-\mathbf{k},\mu;\mathbf{k}_2,\omega_1-\mu)\ ,\end{aligned}\tag{28}$$

$$\begin{aligned}\Psi'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)=&\hat{\sigma}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)+\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,-\omega_1;-\mathbf{k},\omega)\\&+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{-\omega_1+\mu}\hat{\sigma}''(\mathbf{k}_1,\mu;\mathbf{k}_2,\omega-\mu)+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\mu-\omega}\hat{\sigma}''(\mathbf{k}_1,\omega_2-\mu;-\mathbf{k},\mu)\\=&\hat{\sigma}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)+\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,-\omega_1;-\mathbf{k},\omega)\\&+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\omega_2-\mu}\hat{\sigma}''(\mathbf{k}_1,\omega-\mu;\mathbf{k}_2,\mu)+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\mu-\omega}\hat{\sigma}''(\mathbf{k}_1,\omega_2-\mu;-\mathbf{k},\mu)\ ,\end{aligned}\tag{29}$$

$$\begin{aligned}\Psi'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)=&\hat{\sigma}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)-\tfrac{1}{2}\hat{\sigma}'(-\mathbf{k},\omega;\mathbf{k}_2,-\omega_2)+\tfrac{1}{2}\hat{\sigma}'(\mathbf{k}_1,\omega_1;k_2,\omega_2)\\&+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{\mu-\omega}\hat{\sigma}''(-\mathbf{k},\mu;\mathbf{k}_2,\omega_1-\mu)+\frac{1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}d\mu\frac{1}{-\omega_2+\mu}\hat{\sigma}''(\mathbf{k}_1,\omega-\mu;\mathbf{k}_2,\mu)\ .\end{aligned}\tag{30}$$

By combining (28)–(30), according to (27), we find

$$\begin{aligned}[\Psi'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]&=2[\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)+\hat{\sigma}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)+\hat{\sigma}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\\&\equiv 2\underline{R}[\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]\ .\end{aligned}\tag{31}$$

Now we concentrate on the effect of the $\underline{R}[\ ]$ operation on the rhs of Eq. (26). While it leaves invariant $\underline{R}\{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)/\omega_1\omega_2\}$, it affects the Hilbert transform term,

$$\begin{aligned}\Psi'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)=-\frac{n}{8\hbar^2}\Bigg[&\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}\\&-\frac{1}{\pi^2}\int_{-\infty}^{+\infty}d\mu_1\int_{-\infty}^{+\infty}d\mu_2\,\mathrm{P}\left[\frac{1}{-\omega+\mu_1+\mu_2}\right]\mathrm{P}\left[\frac{1}{\omega_1-\mu_1}\right]\underline{R}\left\{\frac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2,\mu_2)}{\mu_1\mu_2}\right\}\Bigg]\end{aligned}\tag{32a}$$

and

$$\begin{aligned}\Psi'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)=-\frac{n}{8\hbar^2}\Bigg[&\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}\\&-\frac{1}{\pi^2}\int_{-\infty}^{+\infty}d\mu_2\int_{-\infty}^{+\infty}d\mu_1\,\mathrm{P}\left[\frac{1}{\omega_2-\mu_2}\right]\mathrm{P}\left[\frac{1}{-\omega+\mu_1+\mu_2}\right]\underline{R}\left\{\frac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2,\mu_2)}{\mu_1\mu_2}\right\}\Bigg]\ .\end{aligned}\tag{32b}$$

In order to be able to combine (32a) and (32b) with (31), we have to interchange the order of integration in (32b), which can be done with the aid of the Poincaré-Bertrand theorem,[26]

$$\Psi'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)=-\frac{n}{8\hbar^2}\left[\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}\right.$$

$$-\frac{1}{\pi^2}\int_{-\infty}^{+\infty}d\mu_1\int_{-\infty}^{+\infty}d\mu_2\left[\mathrm{P}\left[\frac{1}{\omega_2-\mu_2}\right]\mathrm{P}\left[\frac{1}{-\omega+\mu_1+\mu_2}\right]\right.$$

$$\left.\left.-\pi^2\delta(\omega_2-\mu_2)\delta(\omega-\mu_1-\mu_2)\right]\underline{R}\left\{\frac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2,\mu_2)}{\mu_1\mu_2}\right\}\right]$$

$$=-\frac{n}{8\hbar^2}\left[2\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}-\frac{1}{\pi^2}\int_{-\infty}^{+\infty}d\mu_1\int_{-\infty}^{+\infty}d\mu_2\mathrm{P}\left[\frac{1}{\omega_2-\mu_2}\right]\mathrm{P}\left[\frac{1}{-\omega+\mu_1+\mu_2}\right]\right.$$

$$\left.\times\underline{R}\left\{\frac{Q(\mathbf{k}_1,\mu_1;\mathbf{k}_2,\mu_2)}{\mu_1\mu_2}\right\}\right]. \tag{32c}$$

Now combining (26) and (32) into $\underline{R}[\Psi'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]$ we find that the Hilbert transform terms cancel each other, leading to the final form of the QFDT,

$$\underline{R}[\hat{\sigma}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]=-\frac{n}{4h^2}\underline{R}\left\{\frac{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}$$

$$=\frac{n}{4h^2}\frac{e^3}{kk_1k_2}\underline{R}\left\{\omega S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\right\}. \tag{33}$$

Alternative forms of the QFDT can be obtained by trading, on the one hand, the conductivity for the polarizability α or for the density response function χ,[7] and, on the other hand, the current three-point function for the density three-point functions $S(120)$, etc., defined in (16), and by working in terms of the internal response functions.[7,9] Then

$$\underline{R}\{\omega S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\}=-\frac{\hbar^2}{n\pi}\frac{k_1k_2k}{e^3}\,\mathrm{Im}\frac{1}{\epsilon(\mathbf{k}_1,\omega_1)\epsilon(\mathbf{k}_2,\omega_2)\epsilon^*(\mathbf{k},\omega)}\underline{R}[\omega\alpha(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]$$

$$=\frac{4\hbar^2}{n}\,\mathrm{Re}\left[\frac{1}{\epsilon(\mathbf{k}_1,\omega_1)\epsilon(\mathbf{k}_2,\omega_2)\epsilon^*(\mathbf{k},\omega)}\right]\underline{R}[\omega\chi(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]. \tag{34}$$

IV. INVERSION OF THE FLUCTUATION-DISSIPATION RELATION

Equation (34) constitutes a useful form of the FDT. It is probably also the simplest form into which the relationship can be cast. Nevertheless, Eq. (34) provides only the symmetrized combination of the S functions. Because of the appearance of both cycles in the symmetrized product, it is impossible to invert (34) in such a way that an explicit expression for $S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ results. It is possible, however, to obtain such an explicit representation. In this section we present an alternate form of the FDT relation, which, while it lacks the simplicity of (34), leads to an expression for a single $S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ in terms of the quadratic response functions. The derivation rests on the observation that combinations of Eq. (19) and its counterparts generated by shifting arguments can be combined to yield a linear system of equations for the two cycles of S, from which each cycle can independently be extracted.[18] First we trade $\hat{\sigma}$ and Q for $\hat{\chi}$ and S in Eq. (19),

$$\hat{\chi}(\mathbf{k}_1,\tau_1;\mathbf{k}_2,\tau_2)$$

$$=-\frac{n}{2\hbar^2}\{\Theta(\tau_1)\Theta(\tau_2-\tau_1)\underline{F}_{12}[S(012)+S(210)]$$

$$+\Theta(\tau_2)\Theta(\tau_1-\tau_2)\underline{F}_{12}[S(120)+S(021)]$$

$$-\Theta(\tau_1)\Theta(\tau_2)\underline{F}_{12}[S(201)+S(102)]\} \tag{35}$$

with $\underline{F}_{12}$ representing the Fourier-transform operator

$$\underline{F}_{12}\equiv\underline{F}(\tau_1,\tau_2\,|\,\omega_1,\omega_2)$$

$$=\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}e^{-i(\omega_1\tau_1+\omega_2\tau_2)}\cdots. \tag{36}$$

Changing the arguments of $\hat{\chi}$ one generates a second equation,

$$\hat{\chi}(\mathbf{k}_2,\tau_2-\tau_1;-\mathbf{k},-\tau_1)$$
$$=-\frac{n}{2\hbar^2}\{\Theta(\tau_2-\tau_1)\Theta(-\tau_2)\underline{F}_{12}[S(120)+S(021)]$$
$$+\Theta(-\tau_1)\Theta(\tau_2)\underline{F}_{12}[S(201)+S(102)]$$
$$-\Theta(\tau_2-\tau_1)\Theta(-\tau_1)\underline{F}_{12}[S(021)+S(210)]\}. \tag{37}$$

Here we have exploited relations such as

$$\underline{F}(\tau_2-\tau_1,-\tau_1\mid\omega_1,\omega_2)S(012)=\underline{F}(\tau_1,\tau_2\mid\omega_1,\omega_2)S(120). \tag{38}$$

Two additional relations can be generated by $\tau_1\rightarrow-\tau_1,\tau_2\rightarrow-\tau_2$ time reflection, observing $S(cba)=S(-a\,-b\,-c)$ [cf. Eq. (C9)]. Adding now these four relationships one can note that for the surviving terms the Θ functions combine into a full coverage of the τ_1-τ_2 plane, thus leading to the result

$$\underline{F}_{12}[S(012)+S(210)-S(201)-S(102)]$$
$$=-\frac{2\hbar^2}{n}\hat{\chi}[(\mathbf{k}_1,\tau_1;\mathbf{k}_2,\tau_2)+\hat{\chi}(\mathbf{k}_1,-\tau_1;\mathbf{k}_2,-\tau_2)$$
$$-\hat{\chi}(\mathbf{k}_2,\tau_2-\tau_1;-\mathbf{k}_1,-\tau_1)$$
$$-\hat{\chi}(\mathbf{k}_2,\tau_1-\tau_2;-\mathbf{k}_1,-\tau_1)] \tag{39}$$

or

$$S(012)+S(210)-S(201)-S(102)$$
$$=-\frac{4\hbar^2}{n}[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)]. \tag{40}$$

Interchanging 1 and 2 yields a similar, but independent relationship,

$$S(021)+S(120)-S(201)-S(102)$$
$$=-\frac{4\hbar^2}{n}[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]. \tag{41}$$

Observing now the relationships within the cycles, Eq. (B4), we can express, say, $S(012)$ and $S(210)$,

$$S(012)=-\frac{4\hbar^2}{n}\frac{1}{D}\{(1-e^{-\beta\hbar\omega_1})e^{-\beta\hbar\omega_2}[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)]$$
$$+(1-e^{-\beta\hbar\omega_2})[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\}, \tag{42a}$$

$$S(210)=\frac{4\hbar^2}{n}\frac{1}{D}\{(1-e^{+\beta\hbar\omega_1})e^{+\beta\hbar\omega_2}[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)]$$
$$+(1-e^{+\beta\hbar\omega_2})[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\}, \tag{42b}$$

with

$$D=2[\sinh(\beta\hbar\omega)-\sinh(\beta\hbar\omega_1)-\sinh(\beta\hbar\omega_2)]. \tag{43}$$

Equations (42a) and (42b) are the desired results of this section.

It is also useful to list the expression for the completely symmetrized combination $\underline{R}\{S\}$,

$$\underline{R}\{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\}$$
$$=-\frac{8\hbar^2}{n}\frac{1}{D}\{[\sinh(\beta\hbar\omega)+2\sinh(\beta\hbar\omega_1)-\sinh(\beta\hbar\omega_2)][\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)]$$
$$+[\sinh(\beta\hbar\omega)-\sinh(\beta\hbar\omega_1)+2\sinh(\beta\hbar\omega_2)][\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\}. \tag{44}$$

V. ZERO-TEMPERATURE LIMIT

At zero temperature, the system is in a pure quantum state (its ground state). As a consequence, each of the S functions is different from zero only within a certain frequency domain. Therefore, the FDT develops a much simpler structure, though it has to be formulated differently for different frequency regions. In order to find the appropriate frequency domains, one develops $\langle 0\,|\,jjj\,|\,0\rangle$ or $\langle 0\,|\,nnn\,|\,0\rangle$ as a sum over a complete set of intermediate states, say $|\,\alpha\rangle$, $|\,\beta\rangle$, etc., and finds

$$\begin{aligned}\langle 0|n_{\mathbf{k}_a}(\omega_a)n_{\mathbf{k}_b}(\omega_b)n_{\mathbf{k}_c}(\omega_c)|0\rangle &= 2\pi\sum_{\alpha,\beta}\int d\tau_1\int d\tau_2\, e^{-i(\omega_a\tau_2+\omega_c\tau_1)}\langle 0|n_{\mathbf{k}_a}(-\tau_2)|\alpha\rangle\langle\alpha|n_{\mathbf{k}_b}(0)|\beta\rangle\langle\beta|n_{\mathbf{k}_c}(-\tau_1)|0\rangle\delta(\omega_a+\omega_b+\omega_c)\\ &= 8\pi^3\sum_{\alpha,\beta}\langle 0|n_{\mathbf{k}_a}(\omega_a)|\alpha\rangle\langle\alpha|n_{\mathbf{k}_b}(\omega_b)|\beta\rangle\langle\beta|n_{\mathbf{k}_c}(\omega_c)|0\rangle\delta(\omega_a+\omega_b+\omega_c)\delta(\omega_a-\omega_{\alpha 0})\delta(\omega_c+\omega_{\beta 0})\end{aligned} \tag{45}$$

with

$$\begin{aligned}\omega_{\alpha 0}&=E_\alpha-E_0\ ,\\ \omega_{\beta 0}&=E_\beta-E_0\ ,\end{aligned} \tag{46}$$

and a,b,c representing any permutation of the indices 1,2,0 $[(\omega_0,\mathbf{k}_0)\equiv(-\omega,-\mathbf{k})]$.

The domains where a particular $Q(abc)$ survives are now determined by the conditions

$$\omega_a>0\ ,$$
$$\omega_c<0\ .$$

On this basis we can divide the frequency space into six regions. In Table I each of the S's and their domains of existence are listed.

First we formulate the zero-temperature limit of the FDT, Eq. (34), for the six different (ω_1,ω_2) regions,

$$\frac{4\hbar^2}{n}\,\mathrm{Re}\left[\frac{1}{\epsilon(\mathbf{k}_1,\omega_1)\epsilon(\mathbf{k}_2,\omega_1)\epsilon^*(\mathbf{k},\omega)}\right]\underline{R}[\omega\chi(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)] = \begin{cases}\omega_1 S(210)+\omega_2 S(120) & \text{for } \omega_1>0,\omega_2>0 \text{ (region 1)}\\ -\omega S(102)+\omega_2 S(120) & \text{for } \omega_1>0,\omega_2<0,\ |\omega_1|>|\omega_2| \text{ (region 2)}\\ -\omega S(102)+\omega_1 S(012) & \text{for } \omega_1>0,\omega_2<0,\ |\omega_1|<|\omega_2| \text{ (region 3)}\\ \omega_2 S(021)+\omega_1 S(012) & \text{for } \omega_1<0,\omega_2<0 \text{ (region 4)}\\ \omega_2 S(021)-\omega S(201) & \text{for } \omega_1<0,\omega_2>0,\ |\omega_1|>|\omega_2| \text{ (region 5)}\\ \omega_1 S(210)-\omega S(201) & \text{for } \omega_1<0,\omega_2>0,\ |\omega_2|>|\omega_1| \text{ (region 6)}\ .\end{cases} \tag{47}$$

Since the left-hand side of (47) is a continuous function of its frequency arguments, so must be the right-hand side. That it indeed is can be seen by examining, for example, the behavior on the boundary between regions 1 and 2 where $\omega_2=0$. The "jump" on the right-hand side is $\Delta=\omega_1[S(102)+S(210)]_{\omega_2=0}$. However, it is easily seen that both $S(102)$ and $S(210)$ are zero on the boundary. Indeed, consider the matrix element $\langle\beta|n_{\mathbf{k}_2}|0\rangle$ in $S(102)$ at $-\omega_2=E_\beta-E_0=0$. Since the ground state is nondegenerate $E_\beta=E_0$ implies $|\beta\rangle=|0\rangle$; the matrix element $\langle 0|n_{\mathbf{k}_2}|0\rangle$, however, vanishes for all $\mathbf{k}_2\neq 0$ because of momentum conservation and $\mathbf{k}_2=0$ has been excluded by virtue of the defining Eq. (16). Similar considerations apply to $\langle 0|n_{\mathbf{k}_2}|\alpha\rangle$ in $S(210)$. Thus $\Delta=0$. The continuity on the other boundaries is ensured in the same fashion.

It is also of interest to consider the zero-temperature limit of the inverted relationships (42) and (43). Different limits are obtained in the six different domains listed above which can conveniently be summarized in the form given in Table II. Here we have used the abbreviations

$$\begin{aligned}a&=\frac{4\hbar^2}{n}\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\ ,\\ b&=\frac{4\hbar^2}{n}\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)\ ,\\ c&=\frac{4\hbar^2}{n}\hat{\chi}(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)\ .\end{aligned} \tag{48}$$

The apparent discontinuities on the boundaries are again easily shown to vanish. For example, between domains 1 and 2, $S(120)$ has a jump a-b; on the boundary, however, $\omega_2=0$ and since χ' is an even function, $a(\omega_2=0)=b(\omega_2=0)$.

VI. CLASSICAL LIMIT

The classical QFDT has been known for some time.[5,8] We now demonstrate that the classical $\hbar\to 0$ limit (which is manifestly equivalent to the high-temperature $\beta\to 0$ limit) of Eq. (34) reproduces the known GKS classical result. In order to accomplish this, we need an expansion of Eq. (34) to order $\hbar^2$, which then yields

$$\underline{R}[\omega S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)]=-\omega\omega_1\omega_2\left[\frac{e^{\beta\hbar\omega_2}S(012)+e^{\beta\hbar\omega_1}S(021)}{\omega_1\omega_2}-\frac{e^{\beta\hbar\omega}S(021)+S(012)}{\omega\omega_2}-\frac{e^{\beta\hbar\omega}S(012)+S(021)}{\omega\omega_1}\right]$$
$$=\frac{\beta^2\hbar^2}{2}\omega\omega_1\omega_2[S(012)+S(021)] \tag{49}$$

and thus

$$\underline{R}\left[\frac{\hat{\alpha}''(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2}{\omega_1\omega_2}\right]=\frac{\hat{\alpha}''(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}-\frac{\hat{\alpha}''(\mathbf{k}_1,\omega_1;-\mathbf{k},-\omega)}{\omega\omega_1}-\frac{\hat{\alpha}''(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)}{\omega\omega_2}$$
$$=\frac{\pi\beta^2e^3n}{kk_1k_2}S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\ . \tag{50}$$

This is exactly the classical result given by Golden, Kalman, and Silevitch in Ref. 5. In terms of the density response function χ,

$$S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=-\frac{4}{\beta^2n}\,\mathrm{Re}\frac{1}{\epsilon(\mathbf{k}_1,\omega_1)\epsilon(\mathbf{k}_2,\omega_2)\epsilon^*(\mathbf{k},\omega)}\underline{R}\left[\frac{\chi(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right]$$
$$=-\frac{4}{\beta^2n}\,\mathrm{Re}\left[\frac{1}{\epsilon(\mathbf{k}_1,\omega_1)\epsilon(\mathbf{k}_2,\omega_2)\epsilon^*(\mathbf{k},\omega)}\right]$$
$$\times\left[\frac{\chi'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}-\frac{\chi'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)}{\omega\omega_2}-\frac{\chi'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)}{\omega\omega_1}\right]. \tag{51}$$

Equation (51) above reproduces the result given by Kalman,[7] with the exception of the coefficient $4/\beta^2n$ which is incorrectly given as $2/\beta^2$ in Ref. 7.

VII. STATIC QFDT

The linear static FDT can be formulated in two different ways. The first more customary relationship arises when one generates the static structure function $S_{\mathbf{k}}=\int(d\omega/2\pi)S(\mathbf{k}\omega)$ in terms of a frequency integral involving $\hat{\chi}''(\mathbf{k},\omega)$. To obtain the second relationship one integrates $(1/\omega)\hat{\chi}''(\mathbf{k},\omega)$ to obtain, via the Kramers-Kronig relations $\hat{\chi}(\mathbf{k})\equiv\hat{\chi}'(\mathbf{k},0)$; this, in turn, is expressed as a frequency integral involving $S(\mathbf{k}\omega)$. In the classical limit there is a confluence of the two relationships, and $\hat{\chi}(\mathbf{k})$ becomes directly related to $S_{\mathbf{k}}$. An analogous procedure can be followed in the case of the QFDT. It is possible to obtain an expression for the static $\hat{\chi}(\mathbf{k}_1,\mathbf{k}_2)\equiv\hat{\chi}'(\mathbf{k}_1,0;\mathbf{k}_2,0)$ and also for $S_{\mathbf{k}_1\mathbf{k}_2}=\int d\omega_1/2\pi\int d\omega_2/2\pi\underline{R}\{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\}$ in terms of the response function although the two relationships are different. Again, in the classical limit, as it has been already shown,[5] the situation is special and there is a direct relationship between $\chi(\mathbf{k}_1,\mathbf{k}_2)$ and $S_{\mathbf{k}_1\mathbf{k}_2}$.

In order to obtain the first version of the static QFDT we consider Eq. (34),

$$\underline{R}\left[\frac{\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right]=\frac{n}{4\hbar^2}\underline{R}\left\{\frac{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}. \tag{52}$$

To obtain the desired result, a careful limiting procedure is needed. First, we prove that both sides of Eq. (52) are finite in the static ($\omega_1\to0,\omega_2\to0$) limit. The boundness of the rhs becomes obvious, by expanding $\underline{R}\{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)/\omega_1\omega_2\}$ as it is done for $\underline{R}\{Q(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)/\omega_1\omega_2\}$ in Sec. V, and realizing that the three-point-density correlations $S(210)$, etc., are bounded for physical reasons. The demonstration for the lhs, however, is more involved. In the subsequent derivation we follow the pattern of Ref. 5.

Let us split $\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ as follows:

$$\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)+\bar{\hat{\chi}}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2) \tag{53}$$

with

$$\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)=\hat{\chi}'(\mathbf{k}_1,0;\mathbf{k}_2,0)$$

being the static quadratic response function. By combining Eq. (52) and Eq. (53) and taking the static limit, we find

$$\lim_{\omega_1\to0}\lim_{\omega_2\to0}\left[\frac{\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)-\hat{\chi}'(-\mathbf{k},\mathbf{k}_1)}{\omega_1}+\frac{\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)-\hat{\chi}'(\mathbf{k}_2,-\mathbf{k})}{\omega_2}\right]$$
$$=-\lim_{\omega_1\to0'}\lim_{\omega_2\to0}\left[\omega\left[\frac{\bar{\hat{\chi}}'(\mathbf{k}_1,\omega_1;-\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}-\frac{\bar{\hat{\chi}}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)}{\omega\omega_2}-\frac{\bar{\hat{\chi}}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)}{\omega\omega_1}\right]\right]$$
$$+\frac{n}{4\hbar^2}\lim_{\omega_1\to0}\lim_{\omega_2\to0}\omega R\left\{\frac{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}. \tag{54}$$

The second term on the rhs of Eq. (54) is zero because of the boundness of $\underline{R}\{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)/\omega_1\omega_2\}$. Furthermore, the first term on its rhs vanishes too due to the even parity of $\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ with respect to the sign change of all frequency arguments

$$\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=\hat{\chi}'(\mathbf{k}_1,-\omega_1;\mathbf{k}_2,-\omega_2) ,$$

which allows one to expand χ' as

$$\begin{aligned}\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)&=\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)+\left[\frac{\partial^2\hat{\chi}'}{\partial\omega_1\partial\omega_2}\right]_{0,0}\omega_1\omega_2\\&\quad+\frac{1}{2!}\left[\frac{\partial^2\hat{\chi}'}{\partial\omega_1^2}\right]_{0,0}\omega_1^2\\&\quad+\frac{1}{2!}\left[\frac{\partial^2\hat{\chi}'}{\partial\omega_2^2}\right]_{0,0}\omega_2^2+\cdots\\&=\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)+\bar{\hat{\chi}}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2) .\end{aligned} \tag{55}$$

Consequently, $\bar{\hat{\chi}}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)/\omega_1\omega_2$ is finite at the static limit. Therefore, the lhs of Eq. (54) is also zero, which requires

$$\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)=\hat{\chi}'(\mathbf{k}_2,-\mathbf{k})=\hat{\chi}'(-\mathbf{k},\mathbf{k}_1) . \tag{56}$$

Thus one obtains

$$\begin{aligned}\underline{R}\left[\frac{\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right]&=\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)\left[\frac{1}{\omega_1\omega_2}-\frac{1}{\omega\omega_2}-\frac{1}{\omega\omega_1}\right]\\&\quad+\underline{R}\left[\frac{\bar{\hat{\chi}}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right]\\&=\underline{R}\left[\frac{\bar{\hat{\chi}}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right] ,\end{aligned} \tag{57}$$

which indicates that the lhs of Eq. (52) is finite in the static limit.

Next, we integrate both sides of Eq. (52) over ω_1 and ω_2,

$$\begin{aligned}&\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\underline{R}\left[\frac{\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right]\\&\quad=\frac{n}{4\hbar^2}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\underline{R}\left\{\frac{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\} .\end{aligned} \tag{58}$$

Since the integrands on the lhs are regular, the integrals can be replaced by their principal values,

$$\begin{aligned}I&\equiv\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\underline{R}\left[\frac{\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right]\\&=\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\\&\quad-\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)}{\omega_1\omega}\\&\quad-\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)}{\omega_2\omega} .\end{aligned} \tag{59}$$

The integration can easily be carried out by using the Kramers-Kronig relation repeatedly,

$$\begin{aligned}I_1&=-\tfrac{1}{4}\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2) ,\\I_2&=-\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(-\mathbf{k},-\omega_1-\omega_2;\mathbf{k}_1,\omega_1)}{\omega_1(\omega_1+\omega_2)}\\&=\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(-\mathbf{k},\omega_1-\omega_2;\mathbf{k}_1,-\omega_1)}{\omega_1(\omega_2-\omega_1)}\\&=-\tfrac{1}{4}\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2) ,\\I_3&=-\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k}_1-\mathbf{k}_2,-\omega_1-\omega_2)}{\omega_2(\omega_1+\omega_2)}\\&=\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{\hat{\chi}'(\mathbf{k}_2,-\omega_2;-\mathbf{k}_1-\mathbf{k}_2,-\omega_1+\omega_2)}{\omega_2(\omega_1-\omega_2)}\\&=\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\frac{\hat{\chi}'(\mathbf{k}_2,-\omega_2;-\mathbf{k}_1-\mathbf{k}_2,-\omega_1+\omega_2)}{\omega_2(\omega_1-\omega_2)}+\tfrac{1}{4}\hat{\chi}'(\mathbf{k}_2,-\mathbf{k})\\&=0 .\end{aligned} \tag{60}$$

TABLE I. Division of the frequency space in six domains of existence.

$S(abc)$	Domain of existence	
$S(201)$	5,6	$(\omega_1<0,\ \omega_2>0)$
$S(102)$	2,3	$(\omega_1>0,\ \omega_2<0)$
$S(210)$	1,6	$(\omega>0,\ \omega_2>0)$
$S(012)$	4,3	$(\omega<0,\ \omega_2<0)$
$S(120)$	1,2	$(\omega>0,\ \omega_1>0)$
$S(021)$	4,5	$(\omega<0,\ \omega_1<0)$

TABLE II. Zero-temperature limits [with a, b, and c defined in Eq. (48)] for the six different (ω_1,ω_2) domains of existence listed in Eq. (47).

$S(abc)$ domain	1	2	3	4	5	6
$S(210)$	b-a					
$S(120)$	c-a	c-b				
$S(102)$		a-b	a-c			
$S(012)$			b-c	b-a		
$S(021)$				c-a	c-b	
$S(201)$					a-b	a-c

Finally, the substitution of (60) for (59) and (58), with

$$F(12)=\omega_1+\omega_2 e^{\beta\hbar\omega}-\omega e^{\beta\hbar\omega_2} \tag{61}$$

provides the QFDT for the static response function,

$$\begin{aligned}\hat{\chi}'(\mathbf{k}_1,\mathbf{k}_2)&=-\frac{n}{2\hbar^2}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\underline{R}\left\{\frac{S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)}{\omega_1\omega_2}\right\}\\&=\frac{n}{2\hbar^2}\int_{-\infty}^{+\infty}\frac{d\omega_1}{2\pi}\int_{-\infty}^{+\infty}\frac{d\omega_2}{2\pi}\frac{1}{\omega\omega_1\omega_2}[F(12)S(012)+F(21)S(021)]\ .\end{aligned} \tag{62}$$

In order to derive the relationship for the static structure function one can integrate any of the S functions or any of their combinations, since the ordering of the static operators is irrelevant. However, the most useful combination is the completely symmetrized one, given in Eq. (44), which yields

$$\begin{aligned}S_{\mathbf{k}_1\mathbf{k}_2}&=\tfrac{1}{6}\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\underline{R}\{S(\mathbf{k}_1,\omega;\mathbf{k}_2,\omega_2)\}\\&=-\frac{4\hbar^2}{n}\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}\frac{1}{D}\{\sinh(\beta\hbar\omega_1)[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)]\\&\qquad+\sinh(\beta\hbar\omega_2)[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\}\ .\end{aligned} \tag{63}$$

In writing down (63) we have exploited the vanishing of the integrals

$$\int d\omega_1\int d\omega_2[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega;-\mathbf{k},-\omega)]$$

and

$$\int d\omega_1\int d\omega_2[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\ ,$$

which follows from (39) and (40). The classical limits both of Eqs. (62) and (63) reduce to the well-known[5]

$$S_{\mathbf{k}_1\mathbf{k}_2}=\frac{2}{n\beta^2}\hat{\chi}(\mathbf{k}_1,\mathbf{k}_2) \tag{64}$$

expression. In the zero-temperature limit (63) simplifies considerably. One finds

$$\begin{aligned}S_{\mathbf{k}_1\mathbf{k}_2}=-\frac{12\hbar^2}{n}\Bigg[&\int_{-\infty}^{+\infty}d\omega_1\int_{-\omega_1}^{0}d\omega_2[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(\mathbf{k}_2,\omega_2;-\mathbf{k},-\omega)]\\&+\int_{-\infty}^{+\infty}d\omega_1\int_{-\infty}^{-\omega_1}d\omega_2[\hat{\chi}'(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)-\hat{\chi}'(-\mathbf{k},-\omega;\mathbf{k}_1,\omega_1)]\Bigg]\ .\end{aligned} \tag{65}$$

Equations (63) and (65) are especially useful as providing a way to calculate the three-particle correlation function $[h(\mathbf{r}_1-\mathbf{r}_2;\mathbf{r}_2-\mathbf{r}_3)$ with Fourier transform $h_{\mathbf{k}_1\mathbf{k}_2}]$ for a degenerate system through

$$1+ng_{\mathbf{k}_1}+ng_{\mathbf{k}_2}+ng_{\mathbf{k}}+n^2h_{\mathbf{k}_1\mathbf{k}_2}=S_{\mathbf{k}_1\mathbf{k}_2}\ . \tag{66}$$

VIII. CONCLUSIONS

In this paper we have derived a general relationship between the three-point density or current correlations and quadratic response functions. This is an obvious, but certainly not trivial, extension of the well-known linear fluctuation-dissipation theorem; it is also the generalization of a similar relationship derived earlier for classical systems.[5] The formal and structural differences between the classical and quantum theorems stem largely from the noncommutability of the density (or current) operators taken at different space-time points. One of the consequences of this noncommutability is the existence of six different three-point functions, distinguished by the ordering of the density operators. In one version of the theorem [Eq. (34)] only the fully symmetrized combination of these three-point functions enters the theorem. In contrast to the classical situation, the Fourier transform of the three-point function $S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ does not exhibit any definite parity property in its frequency argument. As a result of the symmetrization, however, only the even projection of $S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ is connected through the QFDT to the symmetrized combination of the quadratic response function. Thus, in contrast both to the general linear FDT and to the classical QFDT, the theorem in this form does not provide sufficient information to determine the fluctuation spectrum from the knowledge of the response functions. A different version of the theorem [Eq. (42)] allows one to express each of the six three-point functions in terms of the response functions; in this case, however—in contrast to the classical theorem—the three-point function is given not in terms of the fully symmetrized triangle-symmetric combination of the response functions.

Similarly both to the general linear FDT and to the classical QFDT, useful explicit relationships exist between the static structure function

$$S_{\mathbf{k}_1\mathbf{k}_2}=\int\frac{d\omega_1}{2\pi}\int\frac{d\omega_2}{2\pi}S(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$$

and the response functions, on the one hand, and between the static response functions $[\chi(\mathbf{k}_1,\mathbf{k}_2)=\chi(\mathbf{k}_1,\ \omega_1=0;\ \mathbf{k}_2,\ \omega_2=0)$ etc.] and the three-point functions on the other.

As to the response function [say, the conductivity $\sigma(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$], that is also the fully symmetrized combination constructed out of σ-s with arguments rotated along the sides of the triangle $(\mathbf{k}_1,\omega_1)(\mathbf{k}_2,\omega_2)(-\mathbf{k}_1-\mathbf{k}_2,-\omega_1-\omega_2)$ (triangle symmetry) that appears in the first version of the theorem. This feature, however, is already present in the classical version and can be traced to the fact that the Joule dissipation

$$\sum_{\mathbf{k}_1,\mathbf{k}_2}\int d\omega_1\int d\omega_2\,\mathrm{Re}\,j(\mathbf{k},\omega)E^*(\mathbf{k},\omega)\times\delta_{\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2}\delta(\omega-\omega_1-\omega_2)$$

projects out precisely this combination.

We expect that the QFDT will turn out to be useful on the theoretical level both as a calculational tool for the quadratic response functions and for three-point equilibrium correlations, as well as for exact relationships between them, and as a vehicle to formulate new approximation schemes[9] for systems where many point correlations are significant.

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APPENDIX A: EVALUATION OF COMMUTATORS

Let Ω be the exponential operator

$$\Omega=e^{B(s,t,u,\dots)}\ ,\tag{A1}$$

where $s,t,u,\dots$ are parameters. The Campbell[24] and Hausdorff[25] formula provides that

$$\frac{\partial\Omega}{\partial s}=\Omega_s=\Omega\psi(-\mathrm{Ad}[B])B_s\ ,\tag{A2}$$

$$[n,\Omega]=\Omega\psi(-\mathrm{Ad}[B])[n,B]\ ,\tag{A3}$$

where

$$\psi(x)=\frac{e^x-1}{x}=1+\frac{x}{2!}+\frac{x^2}{3!}+\cdots\ ,\tag{A4}$$

$$\mathrm{Ad}[B]=[B,\ \cdots]\ .\tag{A5}$$

By using (A3)–(A5) we obtain

$$\begin{aligned}[n,e^{-\beta H_0}]&=e^{-\beta H_0}\psi(-\mathrm{Ad}[-\beta H_0])[n,-\beta H_0]\\&=-i\beta he^{-\beta H_0}\psi(-i\beta hD)\frac{dn(t)}{dt}\end{aligned}\tag{A6}$$

with $D=d/dt$.

APPENDIX B: EVALUATION OF RELATIONS FOR Q AND S FUNCTIONS WITHIN A CYCLE

Corresponding to the six possible permutations of the $j(n)$ operators with different arguments, there are six different Q (S) functions. They fall in two cycles ("clockwise" and "counterclockwise"). Different relationships apply to two Q (S) functions, depending whether they are within the same cycle or belong to different cycles. In this appendix we display the relationships for Q's within the same cycle.

Consider $\langle j_{\mathbf{k}_1}(\tau_1)j_{\mathbf{k}_2}(\tau_2)j_{\mathbf{k}}(0)\rangle^{(0)}$. By using the definition of the average

$$\begin{aligned}\langle j_{\mathbf{k}_1}(\tau_1)j_{\mathbf{k}_2}(\tau_2)j_{-\mathbf{k}}(0)\rangle^{(0)}&=\frac{1}{Z}\mathrm{Tr}[e^{-\beta H^0}j_{\mathbf{k}_1}(\tau_1)j_{\mathbf{k}_2}(\tau_2)j_{-\mathbf{k}}(0)]\\&=\langle j_{-\mathbf{k}}(-i\beta\hbar)j_{\mathbf{k}_1}(\tau_1)j_{\mathbf{k}_2}(\tau_2)\rangle^{(0)}\end{aligned}\tag{B1}$$

we can find the relations between the Fourier transforms,

$$\langle j_{\mathbf{k}_1}(\omega_1)j_{\mathbf{k}_2}(\omega_2)j_{-\mathbf{k}}(-\omega)\rangle^{(0)}=e^{\beta\hbar\omega}\langle j_{-\mathbf{k}}(-\omega)j_{\mathbf{k}_1}(\omega_1)j_{\mathbf{k}_2}(\omega_2)\rangle^{(0)}\ .\tag{B2}$$

Thus, according to the defining equation (16b), we have

$$e^{\beta\hbar\omega}Q(012)=Q(120)\ .$$

Similarly, the following relationships can be shown to be valid:

$$\begin{aligned}
&e^{-\beta\hbar\omega_2}Q(201)=Q(012)\ ,\\
&e^{-\beta\hbar\omega_1}Q(120)=Q(201)\ ,\\
&e^{\beta\hbar\omega}Q(021)=Q(210)\ ,\\
&e^{-\beta\hbar\omega_1}Q(102)=Q(021)\ ,\\
&e^{-\beta\hbar\omega_2}Q(210)=Q(102)\ .
\end{aligned}\tag{B3}$$

These can be summarized as

$$e^{-\beta\hbar\omega_a}Q(abc)=Q(bca)\ ,\tag{B4}$$

where (a,b,c) stand for any combination of the arguments $(\mathbf{k}_1,\omega_1)$, $(\mathbf{k}_2,\omega_2)$, $(\mathbf{k}_0,\omega_0)$ with $(\mathbf{k}_0,\omega_0)=(-\mathbf{k},-\omega)$.

The same technique can be used for S functions. First, we recall

$$\langle n_{\mathbf{k}_2}(\omega_2)n_{-\mathbf{k}}(-\omega)\rangle^{(0)}=e^{\beta\hbar\omega}\langle n_{-\mathbf{k}}(-\omega)n_{\mathbf{k}_2}(\omega_2)\rangle^{(0)}\ .\tag{B5}$$

Next, we use the relation similar to (B1),

$$\begin{aligned}
&\langle n_{\mathbf{k}_1}(\omega_1)n_{\mathbf{k}_2}(\omega_2)n_{-\mathbf{k}}(-\omega)\rangle^{(0)}\\
&\qquad=e^{\beta\hbar\omega}\langle n_{-\mathbf{k}}(-\omega)n_{\mathbf{k}_1}(\omega_1)n_{\mathbf{k}_2}(\omega_2)\rangle^{(0)}\ .
\end{aligned}\tag{B6}$$

Combining Eqs. (B4) and (B5) with the defining equation (16a), it follows immediately that

$$e^{-\beta\hbar\omega_a}S(abc)=S(bca)\ .\tag{B7}$$

APPENDIX C: EVALUATION OF RELATIONS FOR Q's BETWEEN THE CYCLES

First we demonstrate that the functions $Q(abc)$ [where a,b,c stand for any combination of the arguments $(\mathbf{k}_1,\omega_1),(\mathbf{k}_2,\omega_2),(\mathbf{k}_0,\omega_0)$ with $(\mathbf{k}_0,\omega_0)\equiv(-\mathbf{k},-\omega)$] are real. To see this, compare

$$\begin{aligned}
Q(\xi_1,\tau_1;\xi_2,\tau_2)&=\mathrm{Tr}[\Omega J(\xi_1,\tau_1;\xi_2,\tau_2)]\\
&=\sum_m(\psi_m^*,\Omega J\psi_m)\ ,
\end{aligned}\tag{C1}$$

where J is any combination of the three current operators $j(\xi_1\tau_1),j(\xi_2\tau_2),j(00)$ with the time reversed $Q(\xi_1,-\tau_1;\xi_2,-\tau_2)$ calculated with the aid of the antilinear time-reversal operator T,

$$\begin{aligned}
Q(\xi_1,-\tau_1;\xi_2,-\tau_2)&=\sum_m(T\psi_m^*,\Omega JT\psi_m)\\
&=\sum_m(\psi_m^*T^+,\Omega JT\psi_m)^*\\
&=-(\mathrm{Tr}[\Omega J(\xi_1,\tau_1;\xi_2\tau_2)]^*\\
&=-Q^*(\xi_1,\tau_1;\xi_2,\tau_2)\ ,
\end{aligned}\tag{C2}$$

in view of the negative time parity of the J operator. Moreover, due to the spatial reflection invariance of the system,

$$Q(-\xi_1,\tau_1;-\xi_2,\tau_2)=-Q(\xi_1,\tau_1;\xi_2,\tau_2)\ ,\tag{C3}$$

which can be proved along the line of the derivation (C2), exploiting the negative spatial parity of the J operator.

The Fourier transform of $Q(\xi_1,\tau_1;\xi_2,\tau_2)$, $Q(abc)$ is now easily shown to be real,

$$Q^*(abc)=Q(abc)\ .\tag{C4}$$

Second, we compare

$$Q(abc)=\mathrm{Tr}(\Omega j_aj_bj_c)\tag{C5}$$

with $Q^*(abc)$ expressed as

$$\begin{aligned}
Q^*(abc)&=\mathrm{Tr}[\Omega j_aj_bj_c)^\dagger]=\mathrm{Tr}(j_c^\dagger j_b^\dagger j_a^\dagger\Omega)\\
&=\mathrm{Tr}[\Omega j_{-c}j_{-b}j_{-a}]\\
&=Q(-c\ -b\ -a)\ .
\end{aligned}\tag{C6}$$

Here $-a$, etc., stand for the pair $(-\mathbf{k}_a,-\omega_a)$, etc., and the trivial $\delta_{\mathbf{k}}$, $\delta(\omega)$, etc., factors in front of Q of Eq. (16) have been omitted. We have used the property of the Fourier transformed $j_{\mathbf{k}}$ operators

$$j^\dagger_{\mathbf{k}\omega}=j_{-\mathbf{k}-\omega}\ .\tag{C7}$$

Combining now (C4) and (C6) we can conclude that two Q functions belonging to different cycles can be related to each other by the

$$\begin{aligned}
&(\mathbf{k}\omega)\rightarrow(-\mathbf{k},-\omega)\ \text{transformation}\ ,\\
&Q(cba)=Q(-a\ -b\ -c)\ .
\end{aligned}\tag{C8}$$

Similar considerations apply to the S functions, except that both the time and space parity of the corresponding N operator [combination of the density operators $n(\xi_1,\tau_1),n(\xi_2,\tau_2),n(0,0)$] is positive,

$$S(cba)=S(-a\ -b\ -c)\ .\tag{C9}$$

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