FLUCTUATION DISSIPATION THEOREMS FOR COULOMB SYSTEMS*

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I have learned To look on nature, not as in the hour Of thoughtless youth; but hearing often-times The still, sad music of humanity, Nor harsh nor grating, though of ample power To chasten and subdue. And I have felt A presence that disturbs me with the joy Of elevated thoughts; a sense sublime Of something far more deeply interfused Whose dwelling is the light of setting suns, And the round ocean and the living air, And the blue sky, and in the mind of man.

WORDSWORTH

Summary

Even though engineering and physics start out with fundamentally different approaches to nature—one wants to synthetize, the other to analyze—the closer they get to a concrete problem—as every practicing scientist knows—the less this difference becomes discernible. In their methods, engineering and physics are closely related: so closely, indeed, that methods, schemas, theorems are shared to the extent that their origins are obscured by the routine usage.

The origin of a set of relationships commonly known as Fluctuation-Dissipation Theorems, and widely used in many-body and plasma physics date back to a paper by Nyquist [1], in which he studied the noise generated in electric circuits. Few of the authors of the papers on the application of fluctuation-dissipation theorem, however, would be acquainted with the Nyquist Theorem. Since Nyquist's early paper, the FDT-s have been derived, re-derived and extended innumerable times, and have acquired a central role in many areas of physics. Thus no historical justification is needed in discussing and reviewing them. Nevertheless, such a discussion also provides, for the interested, a glance at one of the manifestations of the confluence of engineering and physical sciences.

* Dedicated to Professor Károly Simonyi on the occasion of his Seventieth Birthday

Introduction

The basic idea of the Fluctuation-Dissipation Theorems is that the equilibrium properties of a physical system can be probed by testing its response to an external perturbation. This is a remarkable fact. A more technical but equally noteworthy feature is that the "testing" of the system can be done to lower degree of accuracy (lower "order" in a perturbation expansion), than the accuracy of the equilibrium information provided.

Amongst the many papers on FDT-s for physical systems Kubo's [2] seminar paper, the early paper by Callen and Welton [3], and Martin's paper [4] written in the more contemporary language of modern many-body physics may be mentioned in particular. In this paper our emphasis is on plasmas, or more generally, on Coulomb systems. The linear FDT-s for classical plasmas were reviewed by Golden and Kalman [5]. The derivation of the nonlinear FDT is relatively new and is due to Golden, Kalman and Silevitch [6] and Sitenko [7]. Applications of the FDT-s—both linear and nonlinear—to plasmas, in particular to strongly coupled dense plasmas, are reviewed by Kalman [8, 9] and by Golden and Kalman [10].

Extensions to multicomponent systems [11, 12] and to surface plasmas [13] are quite recent and due to Kalman and Golden [11] and to Golden and Lu [12, 13]. Finally the important problem of the nonlinear FDT for quantum systems is presently investigated by Gu and Kalman [14]: the results presented in this paper are new.

Fluctuation-Dissipation Theorems establish relationships between two sets of quantities. The first set is the hierarchy of response functions characterizing the system response to an external perturbation. There is a great deal of freedom in choosing suitable "perturbing" and "responding" quantities. We will adopt $\hat{\phi}$, the potential (energy) and *n*, the density, as the fundamental pair, although we will also need the longitudinal electric current density *j* and the longitudinal electric field \hat{E} . The potential can be either "external", ($\hat{\Phi}$) i.e. induced by external sources only, or "total" (Φ), i.e. including also the induced plasma potential. Then, rather symbolically, the two sets of response functions, $\hat{\chi}$ (external) and χ (total), can be defined through the relations

$$n = \hat{\chi} \hat{\Phi} + \hat{\chi} \hat{\Phi} \hat{\Phi}$$
(1a)

$$n = \chi \Phi + \chi \Phi \Phi .$$
 (1b)

Alternatively,

$$\hat{j} = \hat{\sigma} \underbrace{E}_{1} \underbrace{E}_{2} \underbrace{EE}_{2}$$
(2)

etc.

The notation can be made more explicit by using Fourier representation. E.g.,

$$n(\mathbf{k}\omega) = \chi(\mathbf{k}\omega)\Phi(\mathbf{k}\omega) + \sum_{\substack{\mathbf{p}\mathbf{q} \ 2}} \chi(\mathbf{p}\mu, \mathbf{q}\nu)\Phi(\mathbf{p}\mu)\Phi(\mathbf{q}\nu) + \dots$$
(3)
$$\mathbf{p} + \mathbf{q} = \mathbf{k}$$

$$\mu + \nu = \omega$$

$$\sum_{\mathbf{k}\omega} = \frac{1}{2\pi V} \int d\omega \sum_{\mathbf{k}} \qquad (V = \text{volume}) \,.$$

The longitudinal dielectric function ε is related to χ by

$$\varepsilon(\mathbf{k}\omega) = 1 - \varphi_{\mathbf{k}}\chi(\mathbf{k}\omega) \tag{4}$$

with

$$\varphi_{\mathbf{k}} = \frac{4\pi e^2}{k^2} \tag{5}$$

being the Fourier transforms of the Coulomb-potential.

The relationships between the "external" and "total" response functions $\hat{\chi}$ and χ are

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$$\hat{\chi}(\mathbf{k}\omega) = \frac{\chi(\mathbf{k}\omega)}{\varepsilon(\mathbf{k}\omega)}$$

$$\hat{\chi}(\mathbf{p}\mu, \mathbf{q}\nu) = \frac{\chi(\mathbf{p}\mu, \mathbf{q}\nu)}{\varepsilon(\mathbf{p}\mu)\varepsilon(\mathbf{q}\nu)\varepsilon(\mathbf{k}\omega)}.$$
(6)

The second set of quantities are two-point, three-point, etc. functions (i.e. equilibrium averages of fluctuating quantities taken a two, three, etc. spacetime points) and their Fourier transforms, linear, quadratic, etc., (dynamical) structure factors:

$$S(\mathbf{k},\tau) = \frac{1}{N} \langle n_{\mathbf{k}}(0)n_{-\mathbf{k}}(-\tau) \rangle$$

$$Q(\mathbf{k},\tau) = \frac{1}{N} \langle j_{\mathbf{k}}(0)j_{-\mathbf{k}}(-\tau) \rangle$$

$$S(\mathbf{k}_{1} \quad \tau_{1}; \mathbf{k}_{2}, \tau_{2}) = \frac{1}{N} \langle n_{\mathbf{k}_{1}+\mathbf{k}_{2}}(0)n_{-\mathbf{k}_{1}}(-\tau_{1})n_{-\mathbf{k}_{2}}(-\tau_{2}) \rangle$$

$$\equiv S(012)$$

$$S(\mathbf{k}\omega) = \int d\tau e^{i\omega\tau} S(\mathbf{k},\tau) \qquad (7)$$

etc. The precise definition of the fluctuating quantities are

$$n_{\mathbf{k}} = \sum_{i}^{N} e^{-i\mathbf{k}\cdot\mathbf{x}_{i}} - N\delta_{\mathbf{k}}$$
$$j_{\mathbf{k}} = \sum_{i}^{N} \frac{\mathbf{k}\cdot\mathbf{v}_{i}}{k} e^{i\mathbf{k}\cdot\mathbf{x}_{i}}.$$
(8)

Equally important are the static (equilibrium) structure factors, which are linked to the Fourier transforms of the pair and three-body correlation functions g_k and $h_{k_1k_2}$:

$$S_{\mathbf{k}} = \frac{1}{N} \langle n_{\mathbf{k}}(0) n_{-\mathbf{k}}(0) \rangle$$

= 1 + ng_k
$$S_{\mathbf{k}_{1}\mathbf{k}_{2}} = \frac{1}{N} \langle n_{\mathbf{k}_{1}}(0) n_{\mathbf{k}_{2}}(0) n_{-\mathbf{k}}(0) \rangle$$

= 1 + ng_{k1} + ng_{k2} + ng_{k1+k2} + nh_{k1k2}. (9)

Linear fluctuation-dissipation theorem

We consider [5] a one-component plasma under the influence of a perturbing potential $\hat{\Phi}_k(t)$. The equilibrium system is described by the Hamiltonian

$$H^{(0)} = \sum_{i} \frac{p_i^2}{2m} + \frac{1}{2} \sum_{i \neq j} \varphi(\mathbf{x}_i - \mathbf{x}_j).$$
(10)

The perturbation adds to it a term

$$H^{(1)} = \frac{1}{V} \sum_{\mathbf{k}} \sum_{i} \hat{\Phi}_{\mathbf{k}}(t) e^{i\mathbf{k}\cdot\mathbf{x}_{i}}$$
$$= \frac{1}{V} \sum_{\mathbf{k}} \Phi_{\mathbf{k}}(t) n_{-\mathbf{k}}.$$
(11)

The total Hamiltonian

$$H = H^{(0)} + H^{(1)}$$

generates the Liouville-operator

$$L = L^{(0)} + L^{(1)}$$

$$L^{(0)} = -i[H^{(0)}, \dots]$$

$$L^{(1)} = -i[H^{(1)}, \dots]. \qquad (12)$$

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The equilibrium Liouville operator can be used to construct the time evolution operator for the equilibrium system

$$U(t, t') = e^{iL^{(0)}(t, t')} = U(t - t').$$
(13)

The state of the system is described through the phase space distribution function $\Omega({\mathbf{p}_i} {\mathbf{x}_i}, t)$ in the 6N—dimensional phase space; it satisfies the Liouville equation

$$\frac{\partial \Omega}{\partial t} = -iL\Omega \,. \tag{14}$$

A formally similar, but not identical, equation is obeyed by any dynamical variable, say A:

$$\frac{\mathrm{d}A}{\mathrm{d}t} = +iLA\,.\tag{15}$$

Thus the time evolution operator plays a dual role. In equilibrium

$$\Omega(t) = U(t', t)\Omega(t') \tag{16}$$

and

$$A(t) = U(t', t) A(t)$$
. (17)

The equilibrium $\Omega = \Omega^{(0)}$ exhibits the canonical form

$$\Omega^{(0)} = Z^{-1} e^{-\beta H^{(0)}} \tag{18}$$

while its general form under the effect of the perturbation can be expressed as an expansion in $\hat{\varPhi}_{\bf k}$:

$$\Omega = \Sigma \Omega^{(n)}(t) \,. \tag{19}$$

We concentrate now on $\Omega^{(1)}$. It can be written down as the formal solution of the Liouville equation (14):

$$\Omega^{(1)}(t) = -i \int_{-\infty}^{t} dt' U(t, t') L(t') \Omega^{(0)}.$$
 (20)

A series of simple steps leads to

$$\Omega^{(1)}(t) = \frac{-i\beta}{V} \Omega^{(0)} \sum_{\mathbf{k}} \int_{0}^{\infty} d\tau \, \mathbf{k} \cdot \mathbf{j}_{-\mathbf{k}}(t-\tau) \hat{\Phi}_{\mathbf{k}}(t-\tau).$$
(21)

We now can use (21) to evaluate averages of dynamical quantities.

Consider the longitudinal particle current density $\mathbf{j}_{\mathbf{k}}$:

$$\langle j_{\mathbf{k}}(t) \rangle^{(1)} = \int d\{\mathbf{p}_{i}\} d\{\mathbf{x}_{i}\} \Omega^{(1)}(t) j_{\mathbf{k}}$$
$$= -i \frac{\beta k}{V} \int_{0}^{\infty} d\tau \langle j_{\mathbf{k}}(t) j_{-\mathbf{k}}(t-\tau) \rangle^{(0)} \widehat{\Phi}_{\mathbf{k}}(t-\tau) .$$
(22)

For future reference, we note that (22) is equivalent to

$$\hat{\sigma}(\mathbf{k},\tau) = \beta \Theta(\tau) Q(\mathbf{k},\tau) \tag{23}$$

with $\Theta(\tau)$ being the step function.

Shifting to Fourier transform language and trading $j_{\mathbf{k}}(\omega)$ for $n_{\mathbf{k}}(\omega)$, one finds

$$\langle n_{\mathbf{k}}(\omega) \rangle^{(1)} = -i \frac{\beta \omega}{V} \int_{-\infty}^{+\infty} d\omega' \int_{-\infty}^{+\infty} d\tau e^{i\omega'\tau}.$$
 (24)

Taking now the real part of Eq. (24) and using the definitions of Eq. (1) and (7), one obtains

$$S(\mathbf{k}\omega) = -\frac{2}{\beta n\omega} \operatorname{Im} \hat{\chi}(\mathbf{k}\omega).$$
⁽²⁵⁾

This is the linear dynamical FDT. The static FDT is obtained through integration over ω , which yields

$$\frac{1}{\beta n}\chi(\mathbf{k}0) = S_{\mathbf{k}} = 1 + ng_{\mathbf{k}}.$$
(26)

The fact that g_k is expressible in term of $\hat{\chi}(\mathbf{k}\omega)$ at $\omega = 0$ (a result of the application of the Kramers-Kronig relations) is a remarkable feature of classical systems and does not survive once quantum effects are taken into account.

Nonlinear fluctuation-dissipation theorem

While the linear FDT has been known for some time [2, 5], its extension into the nonlinear domain is relatively new, and is due to Golden, Kalman and Silevitch (GKS) [6] and to Sitenko [7]. These authors have established a quadratic FDT, relating quadratic response functions to three-point correlations.

Although the derivation (we follow the more general approach of GKS) is similar to the derivation of the linear FDT, it is much more involved and presents some special problems. Second order perturbation calculation leads to

$$\Omega^{(2)}(t) = -\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 U(t, t_1) L^{(1)}(t_1) U(t_1, t_2) L^{(1)}(t_2) \Omega^{(0)}$$
(27)

the application of which yields the equivalent of (23):

$$\hat{\sigma}(\mathbf{k}_{1},\tau_{1};\mathbf{k}_{2},\tau_{2}) = -\frac{\beta}{2} \Theta(\tau_{1})\Theta(\tau_{2}) \cdot \{\beta Q(120) + \Theta(\tau_{2}-\tau_{1})Z(120) + \Theta(\tau_{1}-\tau_{2})Z(210)\}$$
(28)

with

$$Z(120) = -\frac{i}{k_1 N} \langle [n_{-\mathbf{k}_1}(-\tau_1), j_{-\mathbf{k}_2}(-\tau_2)] j_{\mathbf{k}}(0) \rangle .$$
 (29)

The rest of notation has been explained in (2) and (7).

The problem with Eq. (28) lies in the presence of the unwieldy Poissonbracket terms. Further progress can be made only after their elimination. This can be accomplished through the following steps. First, observe that Z and Qare related to each other by

$$Z(120) + Z(102) = -\beta Q(120)$$

$$Z(210) + Z(201) = -\beta Q(210)$$

$$= -\beta Q(120).$$
(30)

Substituting Eq. (31) into Eq. (28), one finds

$$\hat{\sigma}(\mathbf{k}_{1}, \tau_{1}; \mathbf{k}_{2}, \tau_{2}) = -\frac{\beta}{2} \Theta(\tau_{1}) \Theta(\tau_{2}) \cdot \{\Theta(\tau_{2} - \tau_{1}) Z(102) + \Theta(\tau_{1} - \tau_{2}) Z(201)\}.$$
(31)

Re-labelling the argument of $\hat{\sigma}$ in Eq. (31) in two different ways and exploiting the time translation invariance property of Z, one can construct

$$\hat{\sigma}(\mathbf{k}_{1}, \tau_{2} - \tau_{1}; -\mathbf{k}, \tau_{2}) + \hat{\sigma}(-\mathbf{k}, \tau_{1}; \mathbf{k}_{2}, \tau_{1} - \tau_{2})
= -\frac{\beta}{2} \left\{ \Theta(\tau_{1})\Theta(\tau_{2}) \left[\Theta(\tau_{2} - \tau_{1})Z(120) \right. \\ \left. + \Theta(\tau_{1} - \tau_{2})Z(210) \right] \right. \\ \left. + \Theta(-\tau_{1})\Theta(\tau_{2})\Theta(\tau_{2} - \tau_{1})Z(021) \right. \\ \left. + \Theta(\tau_{1})\Theta(-\tau_{2})\Theta(\tau_{1} - \tau_{2})Z(012) \right\}.$$
(32)

which when combined with Eq. (28) cancels all the Z-terms except the ones multiplied by $\Theta(-\tau_1)$ and $\Theta(-\tau_2)$, respectively. These latter can, however, be eliminated by projecting out the $\tau_1 > 0$, $\tau_2 > 0$ causal part of the combined response functions. This leads to the desired result, which can conveniently be formulated in Fourier transform language. The central object is the causal symmetrized combination of quadratic response functions,

$$\Xi(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2}) = \omega\hat{\chi}(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2}) -\omega_{1}\int d\mu\delta_{+}(\omega_{1}-\mu)\hat{\chi}(-\mathbf{k}_{1}-\mu;\mathbf{k}\omega_{2}+\mu) -\omega_{2}\int d\mu\delta_{+}(\omega_{2}-\mu)\hat{\chi}(\mathbf{k}\omega_{1}+\mu;-\mathbf{k}_{2}-\mu)$$
(33)

which then appears to be related to the quadratic dynamical structure function through

$$\Xi(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2}) = -\frac{\beta^{2}}{2}\int d\mu \int d\nu \delta_{+}(\omega_{1}-\mu)\delta_{+}(\omega_{2}-\nu)$$

$$\mu\nu\omega S(\mathbf{k}_{1}\mu;\mathbf{k}_{2}\nu)$$
(34)

Eq. (34) can be inverted, so that an explicit relationship for S results:

$$S(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2}) =$$

$$= -\frac{2}{\beta^{2}}\operatorname{Re}\left\{\frac{\hat{\chi}(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2})}{\omega_{1}\omega_{1}} - \frac{\hat{\chi}(\mathbf{k}\omega;-\mathbf{k}_{1}\omega_{1})}{\omega_{1}\omega} - \frac{\hat{\chi}(\mathbf{k}\omega;-\mathbf{k}_{2}\omega_{2})}{\omega_{2}\omega}\right\}.$$
 (35)

The above relationship constitutes the quadratic dynamical FDT and it is analogue of the linear Eq. (25). The equivalent of the static Eq. (26) is

$$\hat{\chi}(\mathbf{p}0; \mathbf{q}0) = \beta^2 n (1 + ng_{\mathbf{p}} + ng_{\mathbf{q}} + ng_{\mathbf{k}} + nh_{\mathbf{pq}}).$$
(36)

Quantum fluctuation-dissipation theorems

The quantum version of the linear FDT is well known and is widely used in many body theory [2, 4]. A quantum equivalent of the quadratic theorem has, however, been established only quite recently [14], whether it is the most convenient generalization, is still an open question.

The formal difference between classical and quantum FDT formalisms stems from the non-commutability of the microscopic density (or current) operator taken at two different times:

$$\langle n(\tau)n(0)\rangle \neq \langle n(0)n(\tau)\rangle$$
 (37)

and from the more complex way the quantum Liouville operator operates.

The density matrix Ω obeys the formally identical Liouville Equation Eq. (14) as its classical counter-part. Also, the solution of the Liouville- equation, is formally unchanged in terms of the evolution operator $U = e^{-iL^{(0)}\tau}$. What becomes different is the quantity $L^{(1)}\Omega^{(0)}$. Evaluating the latter, one encounters the commutator $[n_k, e^{-\beta H^{(0)}}]$, which becomes (We use units with $\hbar = 1$)

$$[n_{\mathbf{k}}, e^{-\beta H^{(0)}}] = -i\beta e - \beta H^{(0)} \Psi\left(-i\beta \frac{\mathrm{d}}{\mathrm{d}t}\right) \frac{\mathrm{d}n_{\mathbf{k}}}{\mathrm{d}t}$$
(38)

with

$$\Psi(x) = \frac{e^x - 1}{x}.$$
(39)

Note that for $x \to 0$, $\Psi(x) \to 1$ and the classical limit is recovered. We define $S(\mathbf{k}, \tau)$ now as

$$S(\mathbf{k},\tau) = \frac{1}{N} \langle n_{-\mathbf{k}}(-\tau) n_{\mathbf{k}}(0) \rangle$$

$$\neq \frac{1}{N} \langle n_{\mathbf{k}}(0) n_{-\mathbf{k}}(-\tau) \rangle \equiv S(\mathbf{k},-\tau).$$
(40)

However, one can easily relate the latter to the former by exploiting the invariance of the trace under cyclic permutation

$$S(\mathbf{k}, -\tau) = S(\mathbf{k}, \tau - i\beta)$$

$$S(\mathbf{k} - \omega) = e^{-\beta\omega} S(\mathbf{k}\omega)$$
(41)

and the dynamical FDT takes the form

$$(1 - e^{-\beta\omega})S(\mathbf{k}\omega) = -\frac{1}{n\pi}\operatorname{Im}\hat{\chi}(\mathbf{k}\omega)$$
(42)

while the static FDT becomes (cf. Eq. (26))

$$-\frac{1}{2\pi n}\operatorname{cth}\frac{\beta\omega}{2}\operatorname{Im}\hat{\chi}(\mathbf{k}\omega) = S\mathbf{k} = 1 + ng_{\mathbf{k}}.$$
(43)

Establishing the quadratic quantum FDT is a much more difficult matter than any of the preceding cases. Again, the formal solution for Ω [2] is still given by Eq. (27). The expression replacing Eq. (28), however, becomes

$$\sigma(\mathbf{k}_{1}, \tau_{1}; \mathbf{k}_{2}, \tau_{2}) = -\frac{\beta}{2} \Theta(\tau_{1}) \Theta(\tau_{2}) \int d\omega_{1} \int d\omega_{2} e^{i(\omega_{1}\tau_{1}+\omega_{2}\tau_{2})}$$

$$\{ [\Theta(\tau_{2}-\tau_{1})\Psi(\omega_{2})Z(120) + \Theta(\tau_{1}-\tau_{2})\Psi(\omega_{1})Z(210) + \beta\Psi(\omega_{1})\Psi(\omega_{2}) [\Theta(\tau_{2}-\tau_{1})Q(120) + \Theta(\tau_{1}-\tau_{2})Q(120)] \}$$

$$(44)$$

or

with the understanding that Z and Q are now given in ω -representation. With considerable algebra the symmetrization procedure and the solution of the integral equation outlined in the classical case can be carried through, at the expense, however, of generating all the possible permutations of the three density fluctuation correlations appearing in S. Thus one finds,

$$I(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2}) =$$

$$= -2 \operatorname{Re} \left\{ \frac{\hat{\chi}(\mathbf{k}_{1}\omega_{1};\mathbf{k}_{2}\omega_{2})}{\omega_{1}\omega_{2}} - \frac{\hat{\chi}(\mathbf{k}\omega_{1};-\mathbf{k}_{1}-\omega_{1})}{\omega\omega_{1}} - \frac{\hat{\chi}(\mathbf{k}\omega;-\mathbf{k}_{2}-\omega_{2})}{\omega\omega_{2}} \right\} (45)$$

where T is the totally symmetrized quantity

$$T = -\frac{S(102) + S(201)}{\omega_1 \omega_2} + \frac{S(021) + S(120)}{\omega_1 \omega} + \frac{S(012) + S(210)}{\omega_2 \omega}.$$
 (46)

It is instructive to see how the classical limit of Eq. (46) is recovered. In the classical limit all the S-s are identical and the r.h.s. of Eq. (46) is zero; it is however also divided by an (unexhibited) \hbar^2 . Thus an expansion of the r.h.s. is required. Using the relations between the two cycles of the permutations

$$S(210; \omega_1 \omega_2) = S(012; -\omega_1, -\omega_2)$$
(47)

etc., and also the relations within a cycle

$$S(201) = e^{-\beta \omega_2} S(012)$$

$$S(120) = e^{-\beta \omega_1} S(201)$$
(48)

and expanding to order β^2 (i.e. \hbar^2), Eq. (46) reduces to

$$T = \beta^2 S(012) \tag{49}$$

which, then, when combined with the r.h.s. of Eq. (45) yields Eq. (35).

A final note on the usefulness of Eq. (45). While in all the previously discussed cases the FDT relation can be used either to determine S in terms of χ or vice versa, here only the even projection of $S(\omega) (S(\omega) + S(-\omega))$ can be determined from Eq. (45). Whether an independent relation for the odd projection, that would help one to find the full $S(\omega)$, can be found, is still an open question.

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