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Generalization of the fluctuation-dissipation theorem to classical nonequilibrium systems with anisotropic temperatures

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The fluctuation-dissipation theorem (FDT) in its kinetic form is generalized to anisotropic nonequilibrium states with more than one temperature. In a pseudoequilibrium, expressions for the generalized response functions and for the generalized correlation functions can be defined that preserve the form of a FDT in the classical limit.

The fluctuation-dissipation theorem (FDT) relates the macroscopic response of a system to its microscopic fluctuations. The original formulations by Nyquist and by Callen and Welton[1] use the canonical ensemble and apply therefore only to systems in thermal equilibrium. Later Kubo derived the FDT in the framework of a linear-response theory,[2] and this treatment has been generalized by Salpeter and Akhiezer *et al.* to certain nonequilibrium states like plasmas with different temperatures T_i and T_e for the ions and electrons, respectively.[3]

In this Brief Report we want to show a further extension to classical nonequilibrium systems which can be described in terms of nonisotropic temperature distributions. For this purpose we first examine the very general approach of Klimontovich towards a kinetic form of the FDT (Ref. 4) and rederive the earlier results for the two-temperature plasma.[3] This procedure illuminates the possibility of formulating the FDT for nonisotropic systems.

We consider an ensemble of N-particle systems with Hamiltonians $\hat{H}+\delta\hat{H}(t)$, where $\delta\hat{H}(t)$ is small and fluctuates with vanishing mean value. One might think of a plasma where the fluctuating part accounts for the coupling of the microscopic current to the local field while the principal system $\hat{H}$ describes the particles and the nonfluctuating interactions.[4] The density operator $\hat{f}$ of the total system obeys a Liouville–von Neumann equation with $\hat{H}+\delta\hat{H}$. Its time-dependent matrix elements with the eigenstates $|n\rangle$ and $|m\rangle$ of $\hat{H}$ are similarly divided into their ensemble mean values and into a small fluctuating part

$$f_{nm}(t)=\langle f_{nm}(t)\rangle+\delta f_{nm}(t)=\delta_{nm}f_m(t)+\delta f_{nm}(t)\ . \quad (1)$$

Here the angular brackets denote an ensemble average with respect to the density matrix of the *total system*. We assume that the relaxation of $f_m(t)$ towards equilibrium is slow compared to other time scales in the system like the inverse widths of the transitions $n\to m$ so that the assumption $\langle f_{nm}(t)\rangle=\delta_{nm}f_m$ holds, independent of time. In the energy representation the linearized Liouville–von Neumann equation in the small quantities $\delta\hat{f}$ and $\delta\hat{H}$ then reads

$$(\partial_t+i\omega_{nm})\delta f_{nm}=-\frac{i}{\hbar}\delta H_{nm}(f_m-f_n)\ , \quad (2)$$

with $\omega_{nm}=(E_n-E_m)\hbar$ and $\hat{H}|n\rangle=E_n|n\rangle$. The solution of this inhomogeneous equation will give the matrix A of the linear response of the total system to its internal fluctuations $\delta\hat{H}$. The Fourier components of these induced fluctuations denoted by the superscript "ind" are

$$\delta f_{nm}^{(\mathrm{ind})}(\omega)=-\sum_{n'm'}A_{nmn'm'}(\omega)\delta H_{n'm'}(\omega)\ . \quad (3)$$

Since A is diagonal, the sum collapses and one finds for the imaginary, dissipative part

$$\mathrm{Im}\,A_{nmn'm'}(\omega)=(\pi/\hbar)\delta(\omega-\omega_{nm})\delta_{nn'}\delta_{mm'}(f_m-f_n)\ . \quad (4)$$

As usual one may add a solution of the homogeneous part of Eq. (2) to the solution (3). As we are only aiming towards the average two-point correlator of the fluctuations δf, it is advantageous to multiply by $\delta f_{n'm'}(t')$ before averaging. The homogeneous part of Eq. (2) then yields the equation

$$(\partial_t+i\omega_{nm})\langle\delta f_{nm}\delta f^*_{n'm'}\rangle^{(s)}(t-t')=0 \quad (5)$$

for the so-called source correlation, which depends in a quasistationary state only on the relative time $t-t'$. It has been argued[4] that the initial condition at equal times can be obtained from the quantum analog of the classical formula for a fluctuation $\delta f = f - \langle f \rangle$,

$$\langle \delta f(X,t)\delta f^*(X',t')\rangle^{(s)}_{t'=t} = \delta(X-X')\langle f(X,t)\rangle - \langle f(X,t)\rangle\langle f(X',t)\rangle \ . \qquad (6)$$

This relation is obvious if $f(X,T)=\delta(X-Y(t))$ is the microscopic N-particle phase-space density in $X=(R,k)$ with the $6N$-dimensional solution $Y(\hat{H},t)$ of Hamilton's equations of motion and will be used here also for the extended system. In quantum mechanics, however, the set of coordinates R and set of momenta k in $X=(R,k)$ do not commute. One employs the Wigner transform,

$$\langle f(X)f^*(X')\rangle = \int dr\, dr' \exp[-i(rk-r'k')]\left\langle \hat{f}\left[R+\frac{r}{2},R-\frac{r}{2}\right] f^\dagger\left[R'+\frac{r'}{2},R'-\frac{r'}{2}\right]\right\rangle$$
$$= \int dr\, dr' \exp[-i(rk-r'k')]\langle \psi^\dagger(r_2)\hat{\psi}(r_1)\hat{\psi}^\dagger(r_1')\hat{\psi}(r_2')\rangle \ , \qquad (7)$$

with $r_{1,2}=R\pm r/2$, etc., the N-particle density operator $\hat{f}$ and the N-tuple creation and annihilation operators $\hat{\psi}^\dagger$ and $\hat{\psi}$, respectively. The time arguments t have been suppressed for simplicity. With the (anti) commutation relation

$$[\hat{\psi}(r_1),\hat{\psi}^\dagger(r_1')]_\mp = \delta(r_1 - r_1') \qquad (8)$$

at equal times, one obtains

$$\langle f(X)f^*(X')\rangle = \int dr\, dr' \exp - i(rk-r'k')(\mp)\langle \hat{\psi}^\dagger(r_2)\hat{\psi}^\dagger(r_1')\hat{\psi}(r_1)\hat{\psi}(r_2')\rangle$$
$$+ \int dr\, dr' \exp - i(rk-r'k')\delta(r_1'-r_1)\langle \hat{\psi}^\dagger(r_2)\hat{\psi}(r_2')\rangle$$
$$= 0 + \int dr\, dr' \exp - i(rk-r'k') \sum_{m,m'} \langle n'|r_1'\rangle\langle r_1|n\rangle\delta_{nn'} \sum_{m,m'} \langle m|r_2\rangle\langle f_{mm'}\rangle\langle r_2'|m'\rangle \ . \qquad (9)$$

Here the first term vanishes as the angular brackets refer to an ensemble of N-particle systems while the operator $\hat{\psi}(r_1)\hat{\psi}(r_2')$ annihilates $2N$ particles. The remaining term yields after symmetrization the desired quantum initial condition

$$\langle \delta f_{nm}\delta f^*_{n'm'}\rangle^{(s)}(t'=t) = \tfrac{1}{2}\delta_{nn'}\delta_{mm'}(f_m+f_n) - \delta_{nm}\delta_{n'm'}f_n f_m \ , \qquad (10)$$

where it has been assumed again that $\langle f_{nm}\rangle = \delta_{nm}f_n$ is diagonal. The right-hand sides of the correlators (6) and (10) may be regarded as a measure of incompleteness of the description of the total system in terms of the distribution $\hat{f}$. They vanish for $\delta\hat{H}=0$, because the density operators become idempotent in pure states. The Fourier-transformed solution of Eq. (5) for $n\neq m$, i.e., $\omega\neq 0$, is

$$\langle \delta f_{nm}\delta f^*_{n'm'}\rangle^{(s)}(\omega) = \pi\delta(\omega-\omega_{nm})\delta_{nn'}\delta_{mm'}(f_m+f_n) \ , \qquad (11)$$

and a comparison with Eq. (4) shows for $\omega=\omega_{nm}$ the kinetic form of the FDT:

$$\langle \delta f_{nm}\delta f^*_{n'm'}\rangle^{(s)}(\omega) = \hbar\frac{f_m+f_n}{f_m-f_n}\,\mathrm{Im}\,A_{nmn'm'}(\omega) \ . \qquad (12)$$

One may insert the equilibrium distribution $f_n \propto \exp(-\beta E_n)$. This yields a prefactor on the right-hand side of Eq. (12) which is independent of the quantum states $|n\rangle$ and $|m\rangle$ since

$$\delta(\omega-\omega_{nm})\frac{f_m+f_n}{f_m-f_n} = \delta(\omega-\omega_{nm})\frac{\exp(-\beta E_m)+\exp(-\beta E_n)}{\exp(-\beta E_m)-\exp(-\beta E_n)}$$
$$= \delta(\omega-\omega_{nm})\coth\left[\frac{\beta}{2}\hbar\omega\right] \ , \qquad (13)$$

and thus

$$\langle \delta f_{nm}\delta f^*_{n'm'}\rangle^{(s)}(\omega) = \hbar\coth\left[\frac{\beta}{2}\hbar\omega\right]\mathrm{Im}\,A_{nmn'm'}(\omega) \qquad (14)$$

We note that the proportionality factor is independent of the states. Equation (12) is also valid in nonequilibrium situations, provided that the relaxation of f_n towards the equilibrium temperature T is slow compared to the other time scales in the system, as was already assumed above. Obviously this can be generalized to a quasiequilibrium situation where the system consists of several components a, each characterized by a different inverse temperature β_a:

$$\sum_a \langle \delta f_{nm}\delta f^*_{n'm'}\rangle^{(s),(a)}(\omega) = \hbar\sum_a \coth\left[\frac{\beta_a}{2}\hbar\omega\right]\mathrm{Im}\,A^{(a)}_{nmn'm'}(\omega) \ . \qquad (15)$$

An example is the FDT between the local source current fluctuations,

$$\delta\mathbf{j}^{(s)}(\omega,\mathbf{k}) = \sum_{n,m}\mathbf{j}_{nm}(\mathbf{k})\delta f^{(s)}_{mn}(\omega) \ , \qquad (16)$$

and the imaginary part of the dielectric tensor in a two-component plasma with different temperatures for the electrons ($a=e$) and the ions ($a=i$),

$$\langle \delta \mathbf{j} \delta \mathbf{j}^\dagger \rangle^{(s)}(\omega,\mathbf{k}) = \sum_{a=e,i} \langle \delta \hat{\mathbf{j}} \delta \hat{\mathbf{j}}^\dagger \rangle^{(s),(a)}(\omega,\mathbf{k}) = \frac{\hbar\omega^2}{4\pi} \sum_{a=e,i} \coth\left[\frac{\beta_a}{2}\hbar\omega\right] \mathrm{Im}\underline{\epsilon}^{(a)}(\omega,\mathbf{k}) \quad (17)$$

where $\mathbf{k}$ is the wave number of the perturbation. Here the usual electromagnetic coupling has been taken for $\delta\hat{H}$ and the $\mathbf{j}_{nm}(\mathbf{k})$ in Eq. (16) are the matrix elements of the current. We note that the dielectric tensor is additive in the particle components in the mean-field approximation.[5]

Another interesting nonequilibrium situation occurs in the electron cooling of ions beams in storage rings.[6] The electrons emerge with an isotropic velocity distribution from the cathode. During the acceleration to the mean velocity of the circulating ions this distribution is quenched along the direction of the beam. More generally, we assume that the principal system $\hat{H}$ [but not necessarily the fluctuating coupling $\delta\hat{H}(t)$] is separable along the axes defining the anisotropy. The distribution of the principal system is parametrized according to

$$f_n \propto \exp(-\boldsymbol{\beta}\cdot\mathbf{E}_\mathbf{n}) \equiv \exp\left[-\sum_i \beta_i (E_i)_{n_i}\right] \quad (18)$$

with the vectors

$$\boldsymbol{\beta} = (T_1^{-1}, T_2^{-1}, T_3^{-1}) , \quad \mathbf{E}_\mathbf{n} = ((E_1)_{n_1}, (E_2)_{n_2}, (E_3)_{n_3}) , \quad (19)$$

and $\mathbf{n}=(n_1,n_2,n_3)$. Here $(E_j)_{n_j}$ is the eigenvalue of the Hamiltonian $\hat{H}_j$ in the direction j. For such an anisotropic distribution an FDT cannot be derived, as Eq. (13) does not hold except for the trivial case of an isotropic distribution in two or even one dimensions, e.g., $T_3=0$ and $T_1=T_2$ or $T_3=T_2=0$, respectively. One can, however, formulate a generalized FDT, which applies for an anisotropic system in the classical limit $\hbar\omega \ll \min(T_i)$. We transform with $\mathbf{n}=\boldsymbol{\nu}+\boldsymbol{\kappa}/2$, $\mathbf{m}=\boldsymbol{\nu}-\boldsymbol{\kappa}/2$ to new classical continuous variables $\boldsymbol{\nu}$ and $\boldsymbol{\kappa}$. As the energy transfer is small, one may replace in Eqs. (4) and (11) differences with differentials, which yields

$$\mathrm{Im}\, A_{\kappa\nu\kappa'\nu'}(\omega) = -(\pi/\hbar)\delta_{\kappa\kappa'}\delta_{\nu\nu'}\delta\left[\omega - \sum_j \kappa_j \partial_{\nu_j} E_j/\hbar\right] \sum_i \kappa_i \partial_{\nu_i} f_\nu , \quad (20)$$

$$\langle \delta f_{\kappa\nu} \delta f^*_{\kappa'\nu'} \rangle^{(s)}(\omega) = 2\pi\delta_{\kappa\kappa'}\delta_{\nu\nu'}\delta\left[\omega - \sum_j \kappa_j \partial_{\nu_j} E_j/\hbar\right] f_\nu . \quad (21)$$

Now for the anisotropic distribution (18) the differentiation yields

$$\partial_{\nu_i} f_\nu = -(\beta_i \partial_{\nu_i} E_i) f_\nu . \quad (22)$$

In contrast to this, the argument of the δ function only involves the unweighted sum of the derivatives of the components of the energy. This suggests a decomposition of Eq. (20) into its additive components $i=1,2,3$. We **define**

$$\mathrm{Im}[A_{\kappa,\nu\kappa'\nu'}(\omega)]_i \equiv (\pi/\hbar)\delta_{\kappa,\kappa'}\delta_{\nu\nu'}\delta\left[\omega - \sum_j \kappa_j \partial_{\nu_j} E_j/\hbar\right] \beta_i \kappa_i \partial_{\nu_i} E_i f_\nu \quad (23)$$

and

$$\langle \delta f_{\kappa\nu} \delta f^*_{\kappa'\nu'} \rangle_i^{(s)}(\omega) \equiv 2\pi\delta_{\kappa\kappa'}\delta_{\nu\nu'}\delta\left[\omega - \sum_j \kappa_j \partial_{\nu_j} E_j/\hbar\right] \frac{\kappa_i \partial_{\nu_i} E_i}{\hbar\omega} f_\nu \quad (24)$$

and obtain the decomposed FDT:

$$\langle \delta f_{\kappa\nu} \delta f^*_{\kappa'\nu'} \rangle_i^{(s)}(\omega) = \frac{2T_i}{\omega} \mathrm{Im}[A_{\kappa\nu\kappa'\nu'}(\omega)]_i . \quad (25)$$

The relation between the source current fluctuations and the electronic dielectric tensor in the classical limit $\hbar \to 0$ is now

$$\langle \delta \hat{\mathbf{j}} \delta \hat{\mathbf{j}}^\dagger \rangle^{(s),(a)}(\omega,\mathbf{k}) = \sum_{i=1,2,3} \langle \delta \hat{\mathbf{j}} \delta \hat{\mathbf{j}}^\dagger \rangle_i^{(s),(a)}(\omega,\mathbf{k}) = \frac{\omega}{2\pi} \sum_{i=1,2,3} T_i^{(a)} \mathrm{Im}\underline{\epsilon}_i^{(a)}(\omega,\mathbf{k}) , \quad (26)$$

where i sums over the three spatial directions and $a=e,i$ denotes the component. Here the decomposed permittivity tensor is

$$\mathrm{Im}\underline{\epsilon}_i^{(a)}(\omega,\mathbf{k}) = \frac{4\pi^2}{\omega^2} \beta_i^{(a)} \sum_\mathbf{p} \frac{N_a e^2}{V} \mathbf{v}\mathbf{v} f_\mathbf{p}^{(a)} \delta(\omega - \mathbf{k}\cdot\mathbf{v}) k_i v_i . \quad (27)$$

Here V is the volume of the system, N_a the number of particles, and e their charge. Naturally, the additivity (17) in respect to the components holds also in the present case. It must be realized, however, that the three components of the FDT [(25) and (26)] cannot be measured independently because both the correlator and the response function involve the full anisotropic distribution function f_ν. The existence of a decomposed FDT for classical anisotropic systems has nevertheless been shown to be useful in a calculation of the recombination rate during electron cooling.[6]

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