Stochastic processes II: Response theory and fluctuation theorems[1]

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Abstract. Linear and nonlinear response theory are developed for stationary Markov systems describing systems in equilibrium and nonequilibrium. Generalized fluctuation theorems are derived which relate the response function to a correlation of nonlinear fluctuations of the unperturbed stationary process. The necessary and sufficient stochastic operator condition for the response tensor, $\chi(t)$, of classical nonlinear stochastic processes to be linearly related to the two-time correlations of the fluctuations in the stationary state (fluctuation theorems) is given. Several classes of stochastic processes obeying a fluctuation theorem are presented. For example, the fluctuation theorem in equilibrium is recovered when the system is described in terms of a mesoscopic master equation. We also investigate generalizations of the Onsager relations for non-equilibrium systems and derive sum rules. Further, an exact nonlinear integral equation for the total response is derived. An efficient recursive scheme for the calculation of general correlation functions in terms of continued fraction expansions is given.

1. Introduction

The purpose of this work on stochastic Markov processes is to develop a general scheme for the calculation of transport coefficients for systems whose unperturbed time-dependence is described by a master equation of a stationary Markov process. We study the response of stationary Markov systems to various external test forces. The response function contains valuable information about the dynamics of the system, in particular, the linear response function can be used to investigate the stability and the normal mode frequencies [1]. A common method for calculation of nonequilibrium transport quantities is via the kinetic equation for the *averaged* molecular distribution function, the Boltzmann equation [2, 3]. But within such a description one neglects the statistical fluctuations in the molecular distribution functions, which may be of importance in critical regimes. Furthermore, the derivation of the Boltzmann equation cannot be outlined without certain restrictions which are rather strict (dilute system, weak short-range interactions, binary collisions, etc.) and often not satisfied [2, 3]. To a certain extent the lack of knowledge about the exact state of the system (i.e. the fluctuations) can be described quite naturally by stochastic Boltzmann–Langevin equations [4–6] or more generally by the use of a

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coarse-grained (mesoscopic) Markov master equation for the process defined over a set of marcrovariables $\mathbf{x}(t) = \{x_1(t), \ldots, x_n(t)\}$ forming the adequate state space Σ of the physical system under consideration.

In Sections 2 and 3 we show that the calculation of the linear response in the theory of stochastic processes [7, 8, 9] is related to a general correlation of fluctuations of the unperturbed stationary system. By the measurement of the response function we obtain information for both: the unperturbed system and the actual transport coefficient. Of special interest are those nonlinear classical stochastic processes for which the linear response tensor $\chi(t)$ is linearly related to the two-time correlations between the fluctuations of the state variables $\mathbf{x}(t)$ (fluctuation theorem). Using the results given in reference [10][3]) we investigate in Section 4 several classes of stochastic processes obeying such a theorem. All these theorems hold independently of the magnitude of the fluctuations of the state variables. Such theorems play an important role in statistical mechanics of conservative systems in thermal equilibrium (where one has the famous fluctuation–dissipation theorem [11–12]) and also in the theory of linear irreversible thermodynamics [13]. In Section 5 we give generalizations of Onsager relations for stationary nonequilibrium systems and derive some sum rules. Based on the integral equations derived in Section 1, we study in Section 6 the nonlinear response. An exact nonlinear integral equation for the nonlinear response is presented. The results obtained are discussed briefly in Section 7. In the Appendix we present a convenient numerical procedure for the calculation of correlation functions of stationary stochastic processes in terms of continued fraction expansions.

2. Linear response theory for Markov processes and Markov–Field processes

We want to study the response of a macroscopic system, which is not necessarily in thermodynamic equilibrium, to an external dynamic perturbation. We assume that the unperturbed system can be described by a time-homogeneous Markov process with a stochastic dissipative generator Γ(I (2.20), I (3.1)). We further assume that the process remains of Markov character in the presence of the perturbation, so that the perturbed macroscopic system is described by a stochastic generator $\hat{\Gamma}(t)$ of a nonstationary Markov process [10]

$$\hat{\Gamma}(t) = \Gamma + \Gamma^{\text{ext}}(t). \tag{2.1}$$

Here $\Gamma^{\text{ext}}(t)$ is the stochastic generator, which represents the effect of the perturbation. All the linear stochastic operators defined on Σ act on the space Π, the linear manifold of probabilities. In terms of the unperturbed 'free' time-homogeneous propagator

$$R(t - t_0) = \exp\{\Gamma(t - t_0)\}, \quad t > t_0 \tag{2.2}$$

we obtain in terms of usual linear operator notation in $\Pi(\Sigma)$ for the perturbed nonstationary propagator, $\hat{R}(t \mid t_0)$, the Dyson equation

$$\hat{R}(t \mid t_0) = R(t - t_0) + \int_{t_0}^{t} R(t - s)\Gamma^{\text{ext}}(s)\hat{R}(s \mid t_0)\, ds, \quad t \geq s \geq t_0. \tag{2.3}$$

[3]) This reference will be denoted in the following by I.

By use of the 'proper self energy' Γ^{ext}

$$\Gamma^{ext}(s \mid r) = \Gamma^{ext}(s)\,\delta(s - r^{+}), \tag{2.4}$$

Equation (2.3) can more simply be written in terms of multiplications ($*$) of operators of $\Pi(T)$, where T denotes the time-parameter space:

$$\begin{aligned} \hat{R} &= R + R * \Gamma^{ext} * \hat{R} \\ \blacktriangle &= \uparrow + \overset{\blacktriangle}{\underset{\uparrow}{\diamond}}\,. \end{aligned} \tag{2.5}$$

An alternative form for the Dyson equation is obtained if the relation between the 'proper self energy', Γ^{ext}, and the 'self energy', Λ, is used [14]

$$\begin{aligned} \Lambda &= \Gamma^{ext} + \Gamma^{ext} * R * \Lambda. \\ \blacklozenge &= \diamond + \overset{\blacklozenge}{\underset{\diamond}{\uparrow}}\,. \end{aligned} \tag{2.6}$$

Hence, we have an alternative exact equation in closed form for the perturbed propagator $\hat{R}$

$$\begin{aligned} \hat{R} &= R + R * \Lambda * R. \\ \blacktriangle &= \uparrow + \overset{\uparrow}{\underset{\uparrow}{\blacklozenge}}\,. \end{aligned} \tag{2.7}$$

The perturbation, $\Gamma(t)^{ext}$, is applied after the system has been prepared at time t_0 in a given stationary state described by the stationary probability $p_{st}(\mathbf{x})$. Without loss of generality the perturbation operator $\Gamma^{ext}(t)$ is expanded in terms of time-dependent external forces $F_i(t)$ and linear stochastic operators Ω_i, which may be enlarged to form a Lie algebra

$$\Gamma^{ext}(t) = \mathbf{F}(t)\cdot\mathbf{\Omega}. \tag{2.8}$$

Thus to first order in the self energy Λ we obtain from equation (2.7) for the perturbed probability $\hat{p}(t)$ of the nonstationary Markov process

$$\hat{p}(t) = p_{st} + \int_{t_0}^{t} R(t - \tau)\mathbf{F}(\tau)\ \mathbf{\Omega} p_{st}\, d\tau. \tag{2.9}$$

The linear response tensor $\chi(t - \tau)$ is then defined by the relation of the response of the state functions $\boldsymbol{\psi}(\mathbf{x}) = \{\psi_1(\mathbf{x}), \psi_2(\mathbf{x}), \ldots, \psi_n(\mathbf{x})\}$

$$\begin{aligned} \langle \delta\boldsymbol{\psi}(t)\rangle^{\text{perturbed}} &= \langle \boldsymbol{\psi}(t)\rangle^{\text{perturbed}} - \langle \boldsymbol{\psi}(t)\rangle^{\text{unperturbed}} \\ &= \int \boldsymbol{\psi}(\mathbf{x})\{\hat{p}(\mathbf{x}t) - p_{st}(\mathbf{x})\}\, d\mathbf{x}, \end{aligned} \tag{2.10}$$

to the external test forces $\mathbf{F}(t)$

$$\langle \delta\boldsymbol{\psi}(t)\rangle^{\text{perturbed}} = \int_{t_0}^{t} \chi(t - \tau)\mathbf{F}(\tau)\, d\tau. \tag{2.11}$$

From equations (2.8–2.11) we find by use of the unit step function $\theta(\tau)$

$$\chi(\tau) = \theta(\tau)\int \psi(\mathbf{x})R(\mathbf{x}\tau \mid \mathbf{y}o)[\mathbf{\Omega}p_{st}]_{\mathbf{y}}\, d\mathbf{y}\, d\mathbf{x}. \tag{2.12}$$

The same procedure can be used if stochastic field variables $\mathbf{X}(\mathbf{r}) = (X_1(\mathbf{r}), \ldots, X_n(\mathbf{r}))$ must be chosen as the system variables. The system is then described in terms of a functional master equation. The linear response is given with space dependent force densities $\mathbf{F}(\mathbf{r}t)$ by

$$\langle \delta\psi(\mathbf{r}t)\rangle^{\text{perturbed}} = \int_{t_0}^{t}\int \chi(\mathbf{r},\mathbf{r}';t-\tau)\mathbf{F}(\mathbf{r}';\tau)\, d\mathbf{r}'\, d\tau. \tag{2.13}$$

In terms of functional integrations we obtain for the linear response tensor

$$\chi(\mathbf{r},\mathbf{r}';\tau) = \theta(\tau)\int \psi(\mathbf{r})R(\psi(\mathbf{r}),\tau \mid \psi'(\mathbf{r}'),0)\cdot[\mathbf{\Omega}p_{st}]_{\psi(\mathbf{r}')}\mathscr{D}\psi\mathscr{D}\psi'. \tag{2.14}$$

For Markov field processes which are homogeneous both in time, t, and space, $\mathbf{r}$, the linear response in equation (2.13) is given by a convolution in time and space or in Fourier space

$$\langle \delta\psi(\mathbf{q},\omega)\rangle^{\text{perturbed}} = \chi(\mathbf{q},\omega)\mathbf{F}(\mathbf{q},\omega). \tag{2.15}$$

The problems with in general non-Gaussian stochastic fields (functional integration) can be eliminated if a cell description for the space is introduced or if the stochastic Fourier components for the corresponding vectorial Markov process are used. In the following we restrict the discussion to real vectorial Markov processes $\mathbf{x}(t)$. We will also specialize the discussion by setting the state functions $\psi(\mathbf{x})$ in equation (2.10) equal to the state variables $\mathbf{x}$ unless explicitly stated otherwise.

3. Generalized fluctuation-theorems

In this section we will express the response tensor $\chi(\tau)$ via correlation functions. We may define a vector valued state function $\phi(\mathbf{x})$

$$\phi(\mathbf{x}) = \frac{[\mathbf{\Omega}p_{st}]_{\mathbf{x}}}{p_{st}(\mathbf{x})}, \tag{3.1}$$

so that the response tensor in equation (2.12) of the state variables $\mathbf{x}(t)$ can be written as a correlation function over the *unperturbed* system

$$\chi(\tau) = \theta(\tau)\langle \mathbf{x}(\tau)\phi(\mathbf{x}(o))\rangle. \tag{3.2}$$

Since the perturbation cannot change the normalization of the probabilities we obtain

$$\int [\mathbf{\Omega}p_{st}]_{\mathbf{x}}\, d\mathbf{x} = \langle \phi(\mathbf{x})\rangle = \mathbf{0}. \tag{3.3}$$

Therefore, the state variables $\mathbf{x}$ may be replaced by its fluctuations $\xi = \mathbf{x} - \langle \mathbf{x}\rangle^{\text{unperturbed}}$ and equation (3.2) has the form of a generalized fluctuation theorem:

$$\chi(\tau) = \theta(\tau)\langle \xi(\tau)\phi(\mathbf{x}(o))\rangle. \tag{3.4}$$

By use of a mixing property for the random variables $\mathbf{x}(t)$ we have in the asymptotic limit the property

$$\lim_{\tau \to +\infty} \chi(\tau) = 0. \tag{3.5}$$

An alternative form for the linear response tensor $\chi(t)$ can be derived by using the 'accompanying stationary solution' $p_a(t)$ (accompanying zero eigenvalue solution) which fulfills for a fixed t

$$\hat{\Gamma}(t) p_a(t; \mathbf{F}(t)) = 0, \tag{3.6}$$

with normalization

$$\int p_a(\mathbf{x}t)\, d\mathbf{x} = 1, \quad \forall t \in [t_0, +\infty). \tag{3.7}$$

By use of the relations

$$\mathbf{\Omega} = \frac{\partial}{\partial \mathbf{F}} \hat{\Gamma}(t; \mathbf{F}(t)) \,|_{\mathbf{F}=0}, \tag{3.8}$$

$$p_a(t; \mathbf{F}(t) = 0) = p_{st}, \tag{3.9}$$

we get from

$$\frac{\partial}{\partial \mathbf{F}} \{(\Gamma + \mathbf{F}(t)\mathbf{\Omega}) p_a(t; \mathbf{F}(t))\} = 0, \tag{3.10}$$

for the fluctuation $\boldsymbol{\phi}(\mathbf{x})$ (in general nonlinear)

$$\begin{aligned} \boldsymbol{\phi}(\mathbf{x}) &= \left[\frac{\partial}{\partial \mathbf{F}} \hat{\Gamma}(t; (\mathbf{F}(t)) \,|_{\mathbf{F}=0} p_{st} \right]_{\mathbf{x}} / p_{st}(\mathbf{x}) \\ &= -\left[\Gamma \frac{\partial}{\partial \mathbf{F}} p_a(t; \mathbf{F}(t)) \,|_{\mathbf{F}=0} \right]_{\mathbf{x}} / p_{st}(\mathbf{x}) \\ &= -[\Gamma \boldsymbol{\varphi}]_{\mathbf{x}} / p_{st}(\mathbf{x}). \end{aligned} \tag{3.11}$$

Introducing the fluctuation

$$\boldsymbol{\eta}(\mathbf{x}) = \boldsymbol{\varphi}(\mathbf{x}) / p_{st}(\mathbf{x}), \tag{3.12}$$

the generalized fluctuation theorem, equation (3.4), can be rewritten in the form

$$\chi(\tau) = -\theta(\tau) \frac{\partial}{\partial \tau} \langle \boldsymbol{\xi}(\tau) \boldsymbol{\eta}(\mathbf{x}(o)) \rangle. \tag{3.13}$$

If we are interested in the response to general state functions $\boldsymbol{\psi}(\mathbf{x})$, we have simply to introduce in equations (3.4) and (3.13) the fluctuation $\boldsymbol{\xi}_{\psi}(\mathbf{x}) = \boldsymbol{\psi}(\mathbf{x}) - \langle \boldsymbol{\psi}(\mathbf{x}) \rangle^{\text{unperturbed}}$

4. Fluctuation theorems

The linear response theory for general time-homogeneous Markov processes developed in Sections 2 and 3 shows that the response function can be expressed in terms of a correlation function of, in general, nonlinear fluctuations of the unperturbed stochastic system. A measure of the linear response function yields therefore information for both: the transport coefficient and the unperturbed stationary system.

Next we will derive the conditions for the validity of *fluctuation theorems* (FT) relating the response tensor to the correlation matrix of the fluctuations of the state variables $\mathbf{x}(t)$. In order to obtain a fluctuation theorem the vector valued state functions $\boldsymbol{\phi}(\mathbf{x})$ in equation (3.1) and $\boldsymbol{\eta}(\mathbf{x})$ in equation (3.12) must be linear in the fluctuations. Hence, we see that the general condition for the validity of a fluctuation theorem depends on the form of the external perturbation as well as on the form of the stationary probability $p_{st}(\mathbf{x})$ or the accompanying probability $p_a(\mathbf{x}t)$. A necessary and sufficient condition for the validity of a fluctuation theorem (FT) in terms of the linear stochastic operators $\boldsymbol{\Omega}_i$ and the stationary probability p_{st} is [15, 16]: whenever we have a stochastic system, obeying

$$[\boldsymbol{\Omega} p_{st}]_{\mathbf{x}} = \sum_{n=0} [(\Gamma^n \mathbf{y} \boldsymbol{\alpha}^{(n)} + \mathbf{c}) p_{st}]_{\mathbf{x}}, \tag{4.1}$$

we obtain by using equations (2.2, 2.9, 3.4, 3.13) the fluctuation theorem

$$\boldsymbol{\chi}(\tau) = \Theta(\tau) \sum_{n=0} \frac{\partial^n}{\partial \tau^n} \langle \boldsymbol{\xi}(\tau) \boldsymbol{\xi}(o) \rangle \boldsymbol{\alpha}^{(n)}, \tag{4.2}$$

or in components

$$\chi_{ij}(\tau) = \theta(\tau) \sum_{n=0} \sum_{l} \frac{\partial^n}{\partial \tau^n} \langle \xi_i(\tau) \xi_l(o)\, d \rangle \alpha_{lj}^{(n)}. \tag{4.3}$$

From equation (4.2) one may derive various sum rule theorems. The formulation in Fourier space is straightforward [12] and will not be outlined here. It is clear that the usual analyticity properties, Kramers–Kronig relations and symmetry properties for the real and imaginary parts of the response tensor in Fourier space hold. Next we will investigate classes of stochastic systems satisfying the condition in equation (4.1).

(I) Let us first discuss the important case of a conservative classical system H_0 in thermal equilibrium which is perturbed by external forces $\mathbf{F}(t)$. Then the perturbation Hamiltonian $H^{ext}(t)$ is

$$H^{ext}(t) = -\mathbf{F}(t) \cdot \mathbf{x}. \tag{4.4}$$

The total anti-symmetric stochastic operator is then given in terms of the Poisson bracket $\{\ ,\ \}$

$$\hat{\Gamma}(t) = \{H_0, \ \} - \mathbf{F}(t)\{\mathbf{x}, \ \}. \tag{4.5}$$

By use of the stationary probability

$$p_{st}(\mathbf{x}) = \frac{1}{Z} \exp - \beta H_0(\mathbf{x}), \tag{4.6}$$

with

$$\beta = \frac{1}{kT}, \tag{4.7}$$

we obtain

$$[\boldsymbol{\Omega} p_{st}]_{\mathbf{x}} = \{p_{st}, \mathbf{y}\}_{\mathbf{x}} = -\beta [\Gamma \mathbf{y} p_{st}]_{\mathbf{x}}, \tag{4.8}$$

where

$$\Gamma = \{H_0, \}. \tag{4.9}$$

From equation (4.8) we obtain for the response tensor $\chi(\tau)$ the well-known Kubo fluctuation–dissipation theorem [17, 18] of classical conservative systems in thermal equilibrium

$$\chi(\tau) = -\theta(\tau)\beta \frac{\partial}{\partial \tau} \langle \xi(\tau)\xi(o)\rangle. \tag{4.10}$$

From equation (4.10) we find further that

$$\chi(\omega = o) = \beta\langle \xi(o)\xi(o)\rangle. \tag{4.11}$$

(II) As a second class we consider a general open stochastic system, whose unperturbed time-dependence is described by a general dissipative time-homogeneous generator Γ and whose accompanying probability, $p_a(\mathbf{x}t)$, after an appropriate coupling of the system to the external forces, is of the following form

$$p_a(\mathbf{x}t) = \frac{1}{Z} \exp -[\Phi(\mathbf{x}) - \mathbf{F}(t)\cdot\alpha\mathbf{x}]. \tag{4.12}$$

By use of equation (3.11) we obtain for the state function $\eta(\mathbf{x})$

$$\eta(\mathbf{x}) = \alpha\xi, \tag{4.13}$$

so that $\chi(\tau)$ takes on the form of a fluctuation theorem:

$$\chi(\tau) = -\theta(\tau) \frac{\partial}{\partial \tau} \langle \xi(\tau)\alpha\xi(o)\rangle. \tag{4.14}$$

In particular, the accompanying probability for a stochastic system in a thermal environment is the generalized canonical probability

$$p_a(\mathbf{x}t) = \frac{1}{Z} \exp -\beta[H_0(\mathbf{x}) - \mathbf{F}(t)\cdot\mathbf{x}]. \tag{4.15}$$

The fluctuation–dissipation theorem for stationary Markov processes describing thermal equilibrium is then given by

$$\chi(\tau) = -\theta(\tau)\beta \frac{\partial}{\partial \tau} \langle \xi(\tau)\xi(o)\rangle, \tag{4.16}$$

or using the antisymmetric part

$$\chi''(\tau) = -\frac{i}{2}[\chi(\tau) - \chi^t(-\tau)] \tag{4.17}$$

$$i\chi''(\tau) = -\tfrac{1}{2}\beta \frac{\partial}{\partial \tau} \langle \xi(\tau)\xi(o)\rangle. \tag{4.18}$$

The notation (^{t}) in equation (4.17) denotes the transpose. Formally the results in equations (4.10) and (4.16) coincide; but the two results differ in the following points. In place of the Liouvillian L in class (I) we have in class (II) a 'coarse-grained' or mesoscopic master equation with a stochastic generator Γ which is *not* anti-symmetric. Hence, the motion of a dynamic quantity $f(\mathbf{x}(t))$ in class (II) is not given in terms of a unitary propagator exp-Lt as it is for a conservative system! The correlation in equation (4.16) is calculated using the mesoscopic joint probability whereas in equation (4.10) the fine grained microscopic joint probability is used. Typical examples of physical systems with a stochastic generator belonging to class (II) are the stochastic Ising models in thermal equilibrium [7, 19, 20] and the hard sphere Brownian motion problems [21].

(III) For the important case of a gradient-type perturbation

$$\Gamma^{ext}(t) = -\mathbf{F}(t)\cdot\nabla, \tag{4.19}$$

we obtain from equation (3.4) the generalized fluctuation theorem

$$\boldsymbol{\chi}(\tau) = -\theta(\tau)\langle\boldsymbol{\xi}(\tau)\nabla \ln p_{st}(\mathbf{x}(o))\rangle. \tag{4.20}$$

Whenever the 'potential' $\ln p_{st}(\mathbf{x})$ is of the form

$$\ln p_{st}(\mathbf{x}) = \phi(\mathbf{x}') + x_j\left(\frac{\alpha_{jj}}{2}x_j + \sum_{\substack{i\\ i\neq j}} \alpha_{ji}x_i\right), \tag{4.21}$$

where $\mathbf{x}'$ denotes the state vector without x_j, we get for the components $\chi_{ij}(\tau)$ a FT

$$\chi_{ij}(\tau) = \theta(\tau)\sum_l \alpha_{jl}\langle\xi_i(\tau)\xi_l(o)\rangle, \quad i = 1,\ldots,n. \tag{4.22}$$

If and only if the stationary probability $p_{st}(\mathbf{x})$ is a Gaussian

$$p_{st}(\mathbf{x}) = \frac{\exp - \frac{1}{2}(\mathbf{x}-\mathbf{a})\boldsymbol{\sigma}^{-1}(\mathbf{x}-\mathbf{a})}{[\det(2\pi\boldsymbol{\sigma})]^{1/2}}, \tag{4.23}$$

$$\boldsymbol{\sigma} = \langle(\mathbf{x}-\mathbf{a})(\mathbf{x}-\mathbf{a})\rangle, \tag{4.24}$$

we obtain a FT for all components of $\boldsymbol{\chi}(\tau)$

$$\boldsymbol{\chi}(\tau) = \theta(\tau)\langle\boldsymbol{\xi}(\tau)\boldsymbol{\xi}(o)\rangle\boldsymbol{\sigma}^{-1} \tag{4.25}$$

with the property

$$\boldsymbol{\chi}(o^+) = \mathbb{1}. \tag{4.26}$$

In particular for a continuous Markov process (see [10]) with a Fokker–Planck generator Γ_{FP}

$$\Gamma_{FP} = -\nabla\cdot\mathbf{A}(\mathbf{x}) + \nabla\nabla : \mathbf{D}(x) \tag{4.27}$$

and a perturbation operator of the form in equation (4.19), the fluctuation theorem in equation (4.25) holds for Gauss–Markov processes, where the drift $\mathbf{A}(\mathbf{x})$ and diffusion $\mathbf{D}(\mathbf{x})$ are

$$\mathbf{A}(\mathbf{x}) = (\mathbf{x} - \langle\mathbf{x}\rangle)\boldsymbol{\alpha} = \boldsymbol{\xi}\boldsymbol{\alpha}, \tag{4.28}$$

$$\mathbf{D}(\mathbf{x}) = \mathbf{D}. \tag{4.29}$$

In this case the covariance matrix $\boldsymbol{\sigma}$ fulfills a fluctuation–dissipation theorem of the second kind [8, 11]

$$\boldsymbol{\alpha}\boldsymbol{\sigma} + \boldsymbol{\sigma}\boldsymbol{\alpha}^t = -2\mathbf{D}. \tag{4.30}$$

(IV) Up to this point the symmetry of generalized detailed balance discussed in I has not been used explicitly. As a first class obeying the above symmetry we study the systems described by the (Ito)-stochastic differential equation [10]

$$d\mathbf{x} = \frac{\partial H(\mathbf{x}, \mathbf{y})}{\partial \mathbf{y}} dt \tag{4.31}$$

$$d\mathbf{y} = -\frac{\partial H(\mathbf{x}, \mathbf{y})}{\partial \mathbf{x}} dt - \mathbf{K} \frac{\partial H(\mathbf{x}, \mathbf{y})}{\partial \mathbf{y}} dt + \mathbf{B}\, d\mathbf{w}, \tag{4.32}$$

where

$$K_{ij} = cV_{ij}, \tag{4.33}$$

$$\mathbf{V} = \tfrac{1}{2}\mathbf{B}\mathbf{B}^t. \tag{4.34}$$

These systems fulfil the usual detailed balance condition with respect to time-reversal symmetry. Thus the damping terms and the noise terms do not destroy the detailed balance condition valid for the pure Hamiltonian system. The stationary probability is given by

$$p_{st}(\mathbf{x}, \mathbf{y}) = \frac{1}{Z} \exp - cH(\mathbf{x}, \mathbf{y}). \tag{4.35}$$

Thus if we use a Hamiltonian of the form

$$H(\mathbf{x}, \mathbf{y}) = G(\mathbf{x}) + \frac{1}{2m} \sum_{i=1}^{N} y_i^2,$$

and perturb the system by adding a term $\mathbf{F}(t)\, dt$ to the right-hand side of equation (4.32) we have a gradient type perturbation, equation (4.19); and from equation (4.22) we obtain

$$\chi_{ij}(\tau) = \theta(\tau) \frac{c}{m} \langle \xi_i(\tau) \xi_j(o) \rangle, \quad \text{for all } i. \tag{4.36}$$

Thus class (IV) is fully contained in class (III). Note that the continuous process $(\mathbf{x}(t), \mathbf{y}(t))$ possesses a singular diffusion matrix, $\mathbf{D}$, and even the sub-matrix $\mathbf{V}$ of $\mathbf{D}$ is in general singular. In physics, stochastic differential equations of the form in equations (4.31–4.32) have found application in turbulence theory and Laser theory [8, 22].

Assuming a nonsingular Diffusion matrix, $\mathbf{D}(\mathbf{x})$, the generalized potential conditions in equations (I(4.47)–I(4.49)) yield the relations

$$[\Gamma_{FP}\, y p_{st}]_{\mathbf{x}} = (\mathbf{A}^+(\mathbf{x}) - \mathbf{A}^-(\mathbf{x}))p_{st}(\mathbf{x}), \tag{4.37}$$

$$\nabla \ln p_{st}(\mathbf{x}) = \mathbf{D}^{-1}(\mathbf{x})(\mathbf{A}^+(\mathbf{x}) - \nabla\, \mathbf{D}(\mathbf{x})). \tag{4.38}$$

By using these relations we find further classes of Fokker–Planck systems obeying a generalized detailed balance symmetry (GDB) that yield for a gradient-type perturbation, equation (4.19), a fluctuation theorem.

(V) If we deal with x-independent diffusion coefficients D_{ij} and a linear irreversible damping

$$\mathbf{A}^+(\mathbf{x}) = \boldsymbol{\alpha}\mathbf{x} + \mathbf{c} \tag{4.39}$$

we obtain

$$\chi_{ij}(\tau) = -\theta(\tau) \sum_{k,l} D_{jk}^{-1} \alpha_{kl} \langle \xi_i(\tau) \xi_l(o) \rangle. \tag{4.40}$$

This form has first been discussed in the case of a GDB-symmetry with respect to the usual time reversal symmetry in References [23, 24].

(VI) An important class is obtained using $\mathbf{x}$-independent diffusion coefficients and drift components $A_i^-(\mathbf{x})$, where as a consequence of the GDB-symmetry (equation I 4.44)

$$\begin{aligned} A_i^-(\mathbf{x}) &= \sum_j \tfrac{1}{2}(1 - \varepsilon_i\varepsilon_j)c_{ij}A_j^+(\mathbf{x}), \quad \varepsilon_i\text{: parity of } x_i \\ &= \sum_j \gamma_{ij}A_j^+(\mathbf{x}). \end{aligned} \tag{4.41}$$

With

$$\boldsymbol{\alpha} = (\mathbb{1} - \boldsymbol{\gamma})^{-1}, \tag{4.42}$$

we obtain, using equations (4.37, 4.38), the fluctuation theorem

$$\chi_{ij}(\tau) = -\theta(\tau)\sum_{k,l} D_{jk}^{-1}\alpha_{kl}\frac{\partial}{\partial\tau}\langle\xi_i(\tau)\xi_l(o)\rangle. \tag{4.43}$$

For the special case, $\boldsymbol{\gamma} = \mathbf{0}$, we find the results derived by several authors [24–29]. A physical example with a nonvanishing $\boldsymbol{\gamma}$ is given by the Risken–Fokker–Planck equation for a single mode Laser with a detuning parameter $c_{ij} \equiv \delta$ [30], such that

$$\boldsymbol{\alpha} = \begin{pmatrix} 1 & -\delta \\ \delta & 1 \end{pmatrix}\frac{1}{1 + \delta^2}. \tag{4.44}$$

(VII) A class of Fokker–Planck systems, where in general two terms with $n = 0, 1$ in the FT of the general form given in equation (4.1) occur, is obtained if we consider $\mathbf{x}$-independent diffusion coefficients and linear drift components $\mathbf{A}^-(\mathbf{x})$

$$\mathbf{A}^-(\mathbf{x}) = \boldsymbol{\alpha}\mathbf{x} + \mathbf{c} \tag{4.45}$$

with

$$\alpha_{ii} = 0 \quad \text{for all } i. \tag{4.46}$$

The GDB-symmetry then implies the consequence (equation I (4.49))

$$\mathbf{A}^-(\mathbf{x})\cdot\boldsymbol{\nabla}\ln p_{st}(\mathbf{x}) = 0, \tag{4.47}$$

and for the response tensor $\boldsymbol{\chi}(\tau)$ we have with equations (4.1, 4.37, 4.38)

$$\chi_{ij}(\tau) = -\theta(\tau)\left\{\sum_{k,l}(D_{jk}^{-1})\alpha_{kl}\langle\xi_i(\tau)\xi_l(o)\rangle + \sum_k (D_{jk}^{-1})\frac{\partial}{\partial\tau}\langle\xi_i(\tau)\xi_k(o)\rangle\right\}. \tag{4.48}$$

(VIII) Finally we consider Fokker–Planck systems with nonlinear diffusion coefficients. Special examples are of course all the one-dimensional Fokker–Planck processes with x-dependent drift and diffusion coefficients yielding a stationary Gaussian probability so that

$$-\boldsymbol{\nabla}\ln p_{st}(x) = \frac{1}{\sigma}(x - \langle x\rangle) = -\frac{A(x)}{D(x)} + \frac{d}{dx}\ln D(x). \tag{4.49}$$

For example, we get with

$$A(x) = c_0\xi\, e^{-\alpha\xi^2}, \quad \alpha > 0, c_0 < -2\alpha\cdot c_1 \tag{4.50}$$

$$D(x) = c_1\, e^{-\alpha\xi^2}, \quad c_1 > 0 \tag{4.51}$$

for σ^{-1} in equation (4.25)

$$\sigma^{-1} = -(c_0/c_1 + 2\alpha) > 0. \tag{4.52}$$

Furthermore, for the nonlinear Brownian motion with

$$A(\xi) = -[\xi + \gamma\xi^3], \tag{4.53}$$

$$D(\xi) = [1 + \gamma(2 + \xi^2)], \tag{4.54}$$

we obtain from equations (4.25) and (4.49)

$$\chi(\tau) = \theta(\tau)\langle\xi(\tau)\xi(o)\rangle. \tag{4.55}$$

As a nontrivial example for a vector valued Fokker–Planck process with $\mathbf{x}$-dependent diffusion coefficients we study the (Ito)-stochastic differential equation

$$\begin{aligned} dx_1 &= x_2\, dt, \\ dx_2 &= (-x_1 + x_2 - x_2^3)\, dt + x_2\, dw_2. \end{aligned} \tag{4.56}$$

The stationary probability is found to be

$$p_{st}(x_1 x_2) = \frac{1}{\pi} \exp -(x_1^2 + x_2^2), \tag{4.57}$$

so that by adding a perturbation $\mathbf{F}\, dt$ to the right-hand side of equations (4.56, 4.57) a fluctuation theorem of the form in equation (4.25) holds, with

$$\boldsymbol{\sigma}^{-1} = \begin{pmatrix} 2 & 0 \\ 0 & 2 \end{pmatrix}. \tag{4.58}$$

Note that the Fokker–Planck classes (IV) and (VIII) are fully contained in the more general class (III). But among the Fokker–Planck classes obeying a GBD-symmetry, (IV–VIII), none of the discussed classes is contained fully as a subclass in another class.

All the response functions considered so far can be expressed via a correlation function over the unperturbed system, calculated through the stationary joint-probability $p^{(2)}(\mathbf{x}, \tau; yo)$. Hence, one needs the complete eigenvalue analysis for the stochastic generator Γ [10]. In practice this may become in general very intractable. An approximation procedure requiring the minimum human and computer effort is therefore very desirable. Such a procedure based on continued fractions is sketched in the Appendix.

5. Generalized Onsager relations and sum rules

In this section we discuss some further consequences for the response function in the presence of symmetries considered in I. If S denotes a symmetry transformation, $S \in G$, [10] in the state space Σ belonging to the symmetry group G of $\mathbf{x}(t)$

$$O_S \Gamma O_S^{-1} = \Gamma, \quad \forall S \in G \tag{5.1}$$

we obtain for the response tensor of the transformed ergodic process [10], $\hat{\mathbf{x}}(t) = S\mathbf{x}(t)$, when the perturbation remains of the same form as in the original process $\mathbf{x}(t)$, the useful relation

$$\begin{aligned}\tilde{\chi}(\tau) &= \theta(\tau)\langle\hat{\xi}(\tau)\boldsymbol{\phi}(\hat{\mathbf{x}}(o))\rangle \\ &= \theta(\tau)\langle\xi(\tau)\boldsymbol{\phi}(\mathbf{x}(o))\rangle \\ &= \chi(\tau), \quad \forall S \in G. \end{aligned} \tag{5.2}$$

Moreover, if $\mathbf{x}(t)$ obeys a GDB-symmetry with respect to a transformation T_0 [10]:

$$T_0^2 = \mathbb{1} \tag{5.3}$$

and

$$\phi_j(\varepsilon\mathbf{x}) = \varepsilon_j\phi_j(\mathbf{x}), \tag{5.4}$$

we have from equation (I 4.16)

$$\begin{aligned}\chi_{ij}(\tau, \boldsymbol{\lambda}) &= \theta(\tau)\langle\xi_i(\tau)\phi_j(\mathbf{x}(o))\rangle_\lambda \\ &= \theta(\tau)\varepsilon_i\varepsilon_j\langle\phi_j(\mathbf{x}(\tau))\xi_i(o)\rangle_{\varepsilon\lambda}. \end{aligned} \tag{5.5}$$

Here $\boldsymbol{\lambda}$ denotes a set of external parameters and $\varepsilon\boldsymbol{\lambda} = (\varepsilon_1\lambda_1, \ldots, \varepsilon_n\lambda_n)$. In particular, we obtain in case of equation (4.1) with diagonal matrices

$$\alpha^{(n)} = \operatorname{diag}(c^{(n)}) \tag{5.6}$$

$$\chi_{ij}(\tau) = \theta(\tau)\sum_{n=0} c^{(n)}\frac{\partial^n}{\partial\tau^n}\langle\xi_i(\tau)\xi_j(o)\rangle, \tag{5.7}$$

generalized Onsager relations for stationary Markov processes obeying a fluctuation theorem of the form in equation (5.7) with $\alpha^{(n)}$ given in equation (5.6) and a GDB-symmetry with respect to a transformation T_0:

$$\chi_{ij}(\tau, \boldsymbol{\lambda}) = \varepsilon_i\varepsilon_j\chi_{ji}(\tau. \varepsilon\boldsymbol{\lambda}). \tag{5.8}$$

For example, considering the fluctuation theorem in equation (4.43) with $\mathbf{A}^-(\mathbf{x}) = \mathbf{0}$ and $\mathbf{D} = \operatorname{diag}(1/\beta^{\text{eff}})$ we obtain for the response tensor the Onsager relations

$$\chi_{ij}(\tau) = \varepsilon_i\varepsilon_j\chi_{ji}(\tau) = -\theta(\tau)\beta^{\text{eff}}\frac{\partial}{\partial\tau}\langle\xi_i(\tau)\xi_j(o)\rangle. \tag{5.9}$$

In contrast to equilibrium situations the relations in equation (5.8) may hold in off-equilibrium situations, e.g. in a Laser system, with an effective Boltzmann factor β^{eff}.

Introducing the Fourier transform $\chi(\omega)$ of the response tensor

$$\chi(\tau) = \theta(\tau)\langle\xi(\tau)\boldsymbol{\phi}(\mathbf{x}(o))\rangle = \theta(\tau)\mathrm{C}(\tau), \tag{5.10}$$

$$\chi(\omega) = \operatorname*{Lim}_{\varepsilon\to 0^+}\int_0^\infty \mathrm{C}(\tau)\, e^{i(\omega + i\varepsilon)\tau}\, d\tau = \chi'(\omega) + i\chi''(\omega), \tag{5.11}$$

we obtain for the in general nonlinear correlation $C_{ij}(\tau)$, depending on whether $C_{ij}(\tau)$ is even or odd in τ

$$C_{ij}(\tau) = \frac{1}{\pi}\int_{-\infty}^{+\infty}\chi'_{ij}(\omega)\, e^{-i\omega\tau}\, d\omega\text{: even} \tag{5.12}$$

$$C_{ij}(\tau) = \frac{i}{\pi}\int_{-\infty}^{\infty} \chi''_{ij}(\omega)\, e^{-i\omega\tau}\, d\omega\text{: odd.} \tag{5.13}$$

The relation in equation (5.12) always holds for a stochastic system which fulfils a strong detailed balance condition due to the even property of the stationary joint probability $p^{(2)}(\mathbf{x}\tau;\mathbf{y}o)$ [10]

$$p^{(2)}(\mathbf{x}\tau;\mathbf{y}o) = p^{(2)}(\mathbf{x} - \tau;\mathbf{y}o), \tag{5.14}$$

and for autocorrelation functions.

If we denote the k-th time-derivative of the correlation function $C_{ij}(\tau)$ by $C_{ij}^{(k)}(\tau)$ we obtain from equations (5.12–5.13) the sum rules for stochastic macroscopic systems that are in general in a stationary nonequilibrium state:

$$(-1)^n C_{ij}^{(2n)}(0^+) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \omega^{2n}\chi'_{ij}(\omega)\, d\omega, \quad n = 0, 1, \ldots \tag{5.15}$$

if $C_{ij}(\tau)$ even, and

$$(-1)^n C_{ij}^{(2n+1)}(0^+) = \frac{1}{\pi}\int_{-\infty}^{+\infty} \omega^{2n+1}\chi''_{ij}(\omega)\, d\omega, \quad n = 0, 1, \ldots \tag{5.16}$$

if $C_{ij}(\tau)$ is odd.

In terms of the stochastic generator Γ and the stationary probability $p_{st}(\mathbf{x})$, the static moments are given by

$$\begin{aligned} C_{ij}^{(n)}(0^+) &= \int \xi_i(o)[\Gamma^n \phi_j p_{st}]_{\mathbf{x}}\, d\mathbf{x} \\ &= \langle\langle \xi_i \Gamma^n \phi_j(\mathbf{x})\rangle\rangle, \quad n = 0, 1, \ldots, \end{aligned} \tag{5.17}$$

or using the backward operator Γ^+, $\Gamma^+(\mathbf{x}, \mathbf{y}) = \Gamma(\mathbf{y}, \mathbf{x})$ [10], by the stationary expectation

$$C_{ij}^{(n)}(0^+) = \langle[\Gamma^{+n}\xi_i]_{\mathbf{x}}\phi_j(\mathbf{x})\rangle, \quad n = 0, 1, \ldots \tag{5.18}$$

The above relations represent a generalization of the sum rule theorem of Kubo [11]. It states that the moments of the frequency distribution of the dissipative intensities in stochastic systems are related to the static moments of the corresponding (in general) nonlinear fluctuations given in equations (5.17, 5.18).

6. Nonlinear response

In this section we calculate the response of the system to external forces of arbitrary strength. The exact perturbed probability $\tilde{p}(xt)$ is obtained from the solution of equations (2.3) or (2.5)

$$\tilde{p}(t) = \tilde{R}(t \mid t_0)p_{st}. \tag{6.1}$$

By use of the vector valued state function $\Xi(\mathbf{x}(t), t)$

$$\Xi(\mathbf{x}(t); t) = \frac{[\Omega\tilde{p}(\mathbf{x}(t), t)]_{\mathbf{x}}}{p_{st}(\mathbf{x})} \tag{6.2}$$

with

$$\langle \Xi(\mathbf{x}(t), t)\rangle_{st} = \int [\Omega\hat{p}(t)]_{\mathbf{x}}\, d\mathbf{x} = \mathbf{0}, \tag{6.3}$$

the total response tensor $\chi^{\text{total}}(t, s)$ can be written in the form of a generalized fluctuation theorem. From

$$\langle \xi(t)\rangle^{\text{perturbed}} = \int_{t_0}^{t} \chi^{\text{total}}(t, s)\mathbf{F}(s)\, ds, \tag{6.4}$$

χ^{total} is calculated as a correlation over the unperturbed stationary system

$$\chi^{\text{total}}(t, s) = \theta(t - s)\langle \xi(t)\Xi(\mathbf{x}(s), s)\rangle. \tag{6.5}$$

According to the explicit time-dependence of the nonlinear fluctuation $\Xi(\mathbf{x}(t), t)$ the total response becomes nonstationary. In most practical cases the nonlinear fluctuation $\Xi(t)$ can rarely be calculated since one needs the exact probability solution for the perturbed system. More useful is an iterative solution of

$$\delta\hat{p}(t) = \hat{p}(t) - p_{st} = \sum_{i=1}^{\infty} f_i(t). \tag{6.6}$$

The quantities $f_i(t)$ obey from equation (2.3) the recursion relation

$$f_n(t) = \int_{t_0}^{t} R(t - \tau)\Gamma^{\text{ext}}(\tau) f_{n-1}(\tau)\, d\tau, \tag{6.7}$$

where

$$f_0 = p_{st}. \tag{6.8}$$

The n-th order response is then defined by

$$\langle \xi(t)\rangle^{(n)\text{perturbed}} = \int \mathbf{x} f_n(\mathbf{x}t)\, d\mathbf{x}. \tag{6.9}$$

In terms of the n-th order response tensor $\chi^{(n)}$,

$$\chi^{(n)}(t - t_1, \ldots, t - t_n) = \left.\frac{\delta^n\langle \xi(t)\rangle^{\text{perturbed}}}{\delta\mathbf{F}(t_n)\ldots\delta\mathbf{F}(t_1)}\right|_{\mathbf{F}=0} \tag{6.10}$$

where all functional derivatives are taken at $\mathbf{F}(t) = \mathbf{0}$, equation (6.9) for the n-th order response can be written in the form

$$\langle \xi_i(t)\rangle^{(n)\text{perturbed}} = \int_{t_0}^{t}\int_{t_0}^{t_1}\cdots\int_{t_0}^{t_{n-1}} dt_1\ldots dt_n \chi^{(n)}_{i\underline{l}_1\ldots\underline{l}_n}\mathbf{F}^{\underline{l}_1}(t)\ldots\mathbf{F}^{\underline{l}_n}(t^n). \tag{6.11}$$

Here the underlining denotes summation over all the force components. For the n-th order response tensor $\chi^{(n)}$ one could derive expressions in terms of high order correlation functions. Of practical importance are the dispersion relations (generalized Kramers–Kronig relations) for the response tensor $\chi^{(n)}$, $n = 2, 3, \ldots$ [31].

From the structure of equation (6.11) together with Fourier transform techniques [31], we can see that the response of the system to an input frequency ω_0 with amplitude A_0 leads for the n-th order response to output frequencies $\omega_{\text{out}} = \{0, 2\omega_0, 4\omega_0, \ldots, n\omega_0\}$ when n is even and to frequencies $\omega_{\text{out}} = \{\omega_0, 3\omega_0, 5\omega_0, \ldots, n\omega_0\}$

when n is odd; each with an amplitude proportional to A_0^n. Note that for an unperturbed, but nonstationary process we obtain a response at an *arbitrary* frequency ω_{out} even in the linear response [32]. Hence, the measurement of the linear response can be used to decide if the initial system is sta ionary or not.

Finally, we derive an exact nonlinear integral equation for the total response $\langle \xi(t) \rangle^{\text{perturbed}}$ (not the total perturbed probability $\tilde{p}(t)$) using the technique of functional integration and functional derivatives. With the functional derivative taken at *finite* $\mathbf{F}(s)$

$$\mathbf{Z}(t, s; \mathbf{F}(s)) = \frac{\delta \langle \xi(t) \rangle^{\text{perturbed}}}{\delta \mathbf{F}} \bigg|_{\mathbf{F}(s)}, \tag{6.12}$$

the total response is written

$$\langle \xi(t) \rangle^{\text{perturbed}} = \int_{t_0}^{t} ds \int \mathbf{Z}(t, s; \mathbf{F}(s)) \, \mathscr{D}\mathbf{F}(s). \tag{6.13}$$

The quantity $\mathbf{Z}$ fulfills from equation (6.4) the Dyson equation

$$\begin{aligned} \mathbf{Z}(t, t'; \mathbf{F}(t')) = {} & \chi^{\text{total}}(t; t') \theta(t - t') \theta(t' - t_0) \\ & + \int_{t_0}^{t} ds \, \mathbf{F}(s) \int_{t_0}^{s} dr \, \Sigma(t, s, r) \mathbf{Z}(s, t'; \mathbf{F}(t')), \end{aligned} \tag{6.14}$$

with the self energy Σ

$$\Sigma(t, s, r) = \frac{\delta \chi^{\text{total}}(t, s)}{\delta \langle \xi(r) \rangle^{\text{perturbed}}}. \tag{6.15}$$

In zero order in $\mathbf{F}$ we have for $\mathbf{Z}$

$$\mathbf{Z}(t, s; \mathbf{F}(s)) = \chi^{(1)}(t - s) \theta(s - t_0), \tag{6.16}$$

giving again the linear response result in equation (2.11)

$$\langle \xi(t) \rangle^{(1)\text{perturbed}} = \int_{t_0}^{t} \chi^{(1)}(t - s) \mathbf{F}(s) \, ds. \tag{6.17}$$

7. Conclusions

We have derived generalized fluctuation theorems for stationary Markov processes and Markov field-processes. In Section 4 we have developed the necessary and sufficient conditions for stochastic systems under which the theorem reduces to an ordinary fluctuation theorem. Several classes of stochastic processes describing equilibrium and non-equilibrium systems have been described which fulfil a fluctuation theorem independent of the magnitude of the fluctuations. For example, the fluctuation–dissipation theorem in thermal equilibrium has been derived if the system is described in terms of a mesoscopic master equation. The existence of fluctuation theorems simplifies considerably the renormalized perturbation scheme for nonlinear classical stochastic processes [33–35]. They may also find wide application in the theory of critical dynamics [35–36]. The generalized Onsager relations given in Section 5 also hold in non-equilibrium systems where they simplify the calculation of the response tensor considerably. All results in this paper, except the nonlinear response results in Section 6, can be evaluated by use of the recursive calculation scheme given in the Appendix provided the stationary probability $p_{st}(\mathbf{x})$ and the generator Γ of the time-homogeneous Markov process are known.

Appendix: Efficient calculation of general correlation functions

We present an efficient procedure for the calculation of response functions or correlation function. The response function $\chi_{ij}(\tau) \equiv \chi(\tau)$ always has the form of a generalized fluctuation theorem equation (3.4)

$$\chi(\tau) = \theta(\tau)\langle g(\mathbf{x}(\tau)f(\mathbf{x}(o))\rangle. \tag{A.1}$$

Using the Taylor series expansion

$$\chi(\tau) = \theta(\tau)\sum_{n=0}^{\infty}\frac{p_n}{n!}\tau^n, \tag{A.2}$$

we can construct from the short time behaviour a type of analytical continuation using continued fraction expansions. The method will only require the explicit form of the stochastic generator Γ and the knowledge of the stationary probability $p_{st}(\mathbf{x})$. By use of equations (5.17) or (5.18) we have for the static moment in equation (A.2) the expressions

$$p_n = \left.\frac{d^n\chi(\tau)}{d\tau^n}\right| \tau = 0^+, \tag{A.3}$$

$$= \langle\langle g(\mathbf{x})\Gamma^n f(\mathbf{x})\rangle\rangle, \tag{A.4}$$

$$= \langle[\Gamma^{+n}g]_{\mathbf{x}}f(\mathbf{x})\rangle. \tag{A.5}$$

For the Fourier transform $\chi(\omega)$

$$\chi(\omega) = \operatorname*{Lim}_{\varepsilon\to 0^+}\int_0^{\infty} e^{i(\omega+i\varepsilon)\tau}\chi(\tau)\,d\tau \tag{A.6}$$

we obtain with $z = -i\omega$ from equation (A.2) the sum rule expansion

$$\chi(\omega) = \sum_{n=0}^{\infty}\frac{p_n}{z^{n+1}}. \tag{A.7}$$

The series in equation (A.7) are in general asymptotic series. Next we construct a continued fraction expansion which serves as an analytical continuation of the series in equation (A.7). The corresponding continued fractions to equation (A.7) are given by

$$\chi(\omega) = \frac{c_1}{z+}\,\frac{c_2}{1+}\,\frac{c_3}{z+}\cdots \tag{A.8}$$

$$= \frac{b_1}{z-a_1+}\,\frac{b_2}{z-a_2+}\,\frac{b_3}{z-a_3+}\cdots \tag{A.9}$$

A general evaluation method for the coefficients in equations (A.8, A.9) is the requirement that a formal expansion in powers of $1/x$ has the same coefficients as those appearing in the asymptotic series equation (A.7). A very convenient method for the calculation of the coefficients in the continued fractions consists in a *recursive* calculation scheme:

Starting with

$$
\begin{aligned}
c_1 &= D_1 & D_1 &= p_0 \\
c_2 &= \frac{-D_2}{D_1} & D_2 &= p_1 \\
c_3 &= \frac{-D_3}{D_2} & D_3 &= p_2 + p_1 c_2 \\
c_4 &= \frac{-D_4}{D_3} & D_4 &= p_3 + p_2(c_2 + c_3)
\end{aligned}
\tag{A.10}
$$

one proceeds from $n = 4$ to the higher terms in the following way: using the auxiliary vector $\mathbf{x}$ of dimension L

$$L = 2 \text{ integer } [(n - 1)/2]$$

$$
\left.
\begin{array}{ll}
x(2) = c_2 + c_3, & x(1) = c_2 \\
\text{interchange} & \\
x(2) \rightarrow x(1) & x(1) \rightarrow x(2)
\end{array}
\right] n = 4
\tag{A.11}
$$

and $x(L - 1) = 0$; we work upwards:

$$
\left.
\begin{array}{l}
x(k) = x(k - 1) + c_{n-1}x(k - 2) \\
\qquad k = L, L - 2, \ldots, 4 \\
x(2) = x(1) + c_{n-1} \\
\text{interchange after each recursion step} \\
\text{the odd and even components} \\
x(2) \rightarrow x(1) \qquad x(4) \rightarrow x(3) \\
x(1) \rightarrow x(2) \qquad x(3) \rightarrow x(4) \text{ etc.}
\end{array}
\right] n \geq 5.
\tag{A.12}
$$

The continued fraction coefficient c_n is then given by

$$c_n = \frac{-D_n}{D_{n-1}}, \tag{A.13}$$

and

$$D_n = p_{n-1} + \sum_{i=1}^{L/2} p_{n-i-1} x(2i - 1). \tag{A.14}$$

With the c_n evaluated by equation (A.13) the coefficients in the contracted form, equation (A.9), are simply given by

$$b_1 = c_1 \qquad a_1 = -c_2, \tag{A.15}$$

$$b_{n+1} = -c_{2n}c_{2n+1} \qquad a_{n+1} = -(c_{2n+1} + c_{2n+2}). \tag{A.16}$$

A Fourier inversion yields the response function in time space. In practice one has to break off the usually infinite continued fractions at a finite order. So far the consequences and the quality of this approximation has not been discussed quantitatively. Hence, the construction of error bounds is very desirable. In systems obeying a strong detailed balance condition the error bounds for autocorrelation functions and their time-derivatives have been discussed elsewhere [37, 38].

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