Generalized Fluctuation-Dissipation Theorem for Steady-State Systems

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(Received 11 February 2009; revised manuscript received 13 March 2009; published 24 August 2009)

The fluctuation-dissipation theorem is a central result of statistical physics, which applies to any system at thermodynamic equilibrium. Its violation is a strong signature of nonequilibrium behavior. We show that for any system with Markovian dynamics, in a nonequilibrium steady state, a proper choice of observables restores a fluctuation-response theorem identical to a suitable version of the equilibrium fluctuation-dissipation theorem. This theorem applies to a broad class of dynamical systems. We illustrate it with linear stochastic dynamics and examples borrowed from the physics of molecular motors and Hopf bifurcations. Finally, we discuss general implications of the theorem.

DOI: 10.1103/PhysRevLett.103.090601 PACS numbers: 05.70.Ln, 05.40.−a, 87.16.Nn

The fluctuation-dissipation theorem, derived from fundamental postulates first by Callen and Welton [1], is very useful in investigating the physical properties of systems at thermodynamic equilibrium: the same information is obtained by measuring the response function of a system or its fluctuations, and depending on convenience one can choose one or the other. This choice is useful in many situations and has led, for example, to the development of microrheology, which brings valuable information on complex viscoelastic fluids [2,3]. Systems which are not at thermodynamic equilibrium do not obey the fluctuation-dissipation theorem. Clear departures have been documented in glasses and biological systems for instance [4–7]. One of the most spectacular cases is that of systems close to a Hopf bifurcation, for which a frequency dependent effective temperature can be defined, which diverges close to the oscillation critical frequency [6,8,9].

Generalizing the fluctuation-dissipation theorem to out of equilibrium systems has already been achieved in a number of cases [10–12]. We propose here a simple generalization based on an identity derived by Hatano and Sasa [13] for systems evolving between two steady states and following Markovian dynamics. It is different from the generalization proposed in Ref. [11], which is limited to systems with thermal noise and energy conserving nonequilibrium dynamics, and which is obtained for time dependent observables. Our theorem is obtained under more general conditions with steady-state variables and is valid, in particular, for non-energy-conserving dynamics. We consider a classical system defined by a set of variables collectively denoted by c in a given configuration. These variables could be positions, velocities, concentrations, order parameters, etc. The state of the system is controlled by a set of parameters λ_α. For fixed values of the control parameters, we assume that there exists a steady state of the system characterized by its probability distribution function normalized to unity $\rho_{\rm ss}(c;\lambda)$. Equivalently, we define the potential $\phi(c;\lambda) = -\log[\rho_{\rm ss}(c;\lambda)]$.

For such a system with Markovian dynamics, Hatano and Sasa derive the following identity [13]:

$$\left\langle \exp\left\{-\int_{t_i}^{t_f} dt\dot{\lambda}_\alpha(t)\frac{\partial\phi(c(t);\lambda(t))}{\partial\lambda_\alpha}\right\}\right\rangle = 1, \qquad (1)$$

where summation convention is used and the dot denotes time derivative. The average is taken over a large number of realizations of a given dynamical process defined by the variation of the nonfluctuating control parameters $\lambda_\alpha(t)$, between an initial time t_i and a final time t_f.

We now consider small variations of the control parameters $\delta\lambda(t) = \lambda(t) - \lambda^{\rm ss}$ around a steady-state value $\lambda^{\rm ss}$ which are vanishing at the initial time $\delta\lambda(t_i) = 0$ and such that, for any time t between t_i and t_f, $\int_{t_i}^{t} dt\dot{\lambda}_\alpha(t)\times\frac{\partial\phi(c(t);\lambda(t))}{\partial\lambda_\alpha} \ll 1$.

Expanding Eq. (1) to second order in the integrand, differentiating with respect to the final time t_f, and noting that the equality must be satisfied for any $\lambda_\alpha(t_f)$, we obtain

$$\left\langle\frac{\partial\phi(t_f)}{\partial\lambda_\alpha}\right\rangle = \int_{t_i}^{t_f} dt\delta\dot{\lambda}_\gamma(t)\left\langle\frac{\partial\phi(t_f)}{\partial\lambda_\alpha}\frac{\partial\phi(t)}{\partial\lambda_\gamma}\right\rangle, \qquad (2)$$

where $\phi(t) \equiv \phi(c(t);\lambda(t))$. The left-hand side of this equation can be expanded, up to linear terms in $\delta\lambda$, as

$$\left\langle\frac{\partial\phi(c(t_f),\lambda^{\rm ss})}{\partial\lambda_\alpha}\right\rangle + \left\langle\frac{\partial^2\phi(c,\lambda^{\rm ss})}{\partial\lambda_\alpha\partial\lambda_\gamma}\right\rangle_{\rm ss}\delta\lambda_\gamma(t_f), \qquad (3)$$

where the average $\langle\cdot\rangle_{ss}$ is calculated in the stationary state $\rho_{ss}(c, \lambda^{ss}) = \exp[-\phi(c, \lambda^{ss})]$. The normalization of the stationary density implies: $\langle \frac{\partial^2\phi}{\partial\lambda_\alpha\partial\lambda_\gamma}\rangle_{ss} - \langle\frac{\partial\phi}{\partial\lambda_\alpha}\frac{\partial\phi}{\partial\lambda_\gamma}\rangle_{ss} = 0$. Up to linear terms in $\delta\lambda$, the average in the right-hand side of Eq. (2) can be taken over ρ_{ss}. Then, integrating by parts and combining Eqs. (2) and (3), we obtain the fluctuation-response theorem

$$\left\langle\frac{\partial\phi(c(t);\lambda^{ss})}{\partial\lambda_\alpha}\right\rangle = \int_{t_i}^{t}\chi_{\alpha\gamma}(t-t')\delta\lambda_\gamma(t')dt', \qquad (4)$$

where the response function $\chi_{\alpha\gamma}(t-t')$ is related to the correlation function as

$$\chi_{\alpha\gamma}(t-t') = \frac{d}{dt}C_{\alpha\gamma}(t-t') = \frac{d}{dt}\left\langle\frac{\partial\phi(c(t);\lambda^{ss})}{\partial\lambda_\alpha}\frac{\partial\phi(c(t');\lambda^{ss})}{\partial\lambda_\gamma}\right\rangle_{ss}. \qquad (5)$$

Equations (4) and (5) constitute the central result of this Letter. They are significantly more general than the fluctuation-dissipation theorem for equilibrium systems, since their validity requires only Markovian dynamics and the existence of a steady-state continuum for each value of the control parameter. There is no need for the existence of a temperature. Note also that the correlation function at equal times is $C_{\alpha\gamma}(t-t'=0) = \langle\frac{\partial^2\phi(c;\lambda^{ss})}{\partial\lambda_\alpha\partial\lambda_\gamma}\rangle_{ss}$. The relation between the correlation function and the response function is slightly more complex in Fourier space:

$$\tilde{\chi}_{\alpha\gamma}(\omega) - \tilde{\chi}_{\gamma\alpha}(-\omega) = i\omega\tilde{C}_{\alpha\gamma}(\omega), \qquad (6)$$

with $\tilde{f}(\omega) = \int f(t)\exp[-i\omega(t)]dt$.

An additional difference with equilibrium dynamics is that, in general, the observables $\frac{\partial\phi(t)}{\partial\lambda_\alpha}$ do not have any particular signature with respect to time reversal, detailed balance does not hold, and neither the response function nor the correlation function have particular symmetries. The observables $\frac{\partial\phi(t)}{\partial\lambda_\alpha}$ have been defined in Ref. [14] as the conjugate of the variables λ_α.

In the following, we illustrate via explicit examples interesting differences between the classical fluctuation-dissipation theorem and our generalization. Close to equilibrium, there are systematic ways to define forces and the related coarse-grained variables from conservation laws and broken symmetries. Far from equilibrium, there are no prescriptions for defining control parameters and identifying coarse-grained variables. In particular, the number of control parameters can exceed that of coarse-grained variables defining the state of the system. We illustrate this statement by an example inspired by the physics of molecular motors. One can also make various choices of control parameters, involving nonlinear transformations between them and not necessarily conserving their numbers. All choices are acceptable as long as the number of "slow" coarse-grained variables is correctly identified. On the contrary, omitting a "slow" variable may result in non-Markovian dynamics and then lead to the violation of our fluctuation-response theorem. In many cases, extending the variable space allows the restoration of Markovian dynamics, and of the theorem. We show this possibility in our second example. As discussed in the introduction, there are many examples, such as that of Hopf bifurcations, which have been shown experimentally and theoretically to break the conventional fluctuation-dissipation theorem. When the systems have Markovian dynamics, the current theorem applies. This is our third example. The second and third examples are specific cases of linear stochastic dynamics that we first discuss in a general way.

Although the fluctuation-response theorem is valid for nonlinear dynamics, in many physical situations, steady states are characterized by a Gaussian probability distribution of the configurations, centered around the average value of a particular observable. This is the case for an "infinitely processive" molecular motor moving in an optical trap, when the trapping zone is large compared to the period of the filament on which the motor moves. The trap acts as an elastic spring opposing the motor motion with a force increasing linearly with the distance to the center of the trap: at some distance x_s from the trap center, the force reaches the motor stall force. Around this position one can linearize the average motor velocity as a function of force. Furthermore, the stochasticity of motor stepping results in a diffusive contribution to the long time, large distance dynamics that we consider here. As a result, the motor behavior is described by the simple Langevin equation,

$$\dot{x}(t) = -k[x(t) - x_s] + \eta(t), \qquad (7)$$

where x is the motor position at time t. The inverse relaxation time k is the product of the trap spring constant and the effective mobility defined as the derivative of the average velocity with respect to external force at stall force. The time k^{-1} is assumed here to be large compared to the motor characteristic time scales. The noise $\eta(t)$ includes both the stochasticity of the motor and thermal noise. It is such that $\langle\eta(t)\rangle = 0$, $\langle\eta(t)\eta(t')\rangle = 2D\delta(t-t')$.

The probability distribution function at steady state and the potential read:

$$\phi(x) = -\log\rho_{ss} = \frac{(x-x_s)^2}{2\sigma^2} + \frac{1}{2}\log(2\pi\sigma^2), \qquad (8)$$

where $\sigma^2 = D/k$. There are many control parameters in such an experiment: one can move the trap center keeping every other parameter fixed, change the trapping power, change adenosine triphosphate (ATP) or adenosine diphosphate (ADP) concentrations, etc. All these choices are perfectly licit and obey the fluctuation-response theorem, although the motor state is defined by a single coarse-grained variable, its position $x(t)$. If we choose as control parameters the trap spring constant k, the trap stall position

x_s, and the motor diffusion constant D, one defines the three observables $X_s = \frac{\partial \phi}{\partial x_s} = -\frac{k(x-x_s)}{D}$, $X_k = \frac{\partial \phi}{\partial k} = \frac{(x-x_s)^2}{2D} - \frac{1}{2k}$, $X_D = \frac{\partial \phi}{\partial D} = -\frac{k(x-x_s)^2}{2D^2} + \frac{1}{2D}$. One has nine response and nine correlation functions; all nonvanishing functions can be expressed in terms of only two of them. It is a straightforward exercise to check that relations (4) and (5), are indeed satisfied. In this simple example, the conjugate variables of the noise intensity D and the spring constant k are proportional to each other. However, they convey different informations relevant to different experimental situations: X_D determines the response of the system to changes of temperature or motor noise and X_k the response to changes in the spring constant. Note however that changes in temperature or in the motor noise that can be due to change in ATP conditions, for example, will also affect other parameters and that in practice one might have to use combinations of the current variables to make use of the fluctuation-response theorem.

We now analyze a generic N-dimensional linear system described by a vector $\vec{x}(t)$ obeying the equation

$$\dot{\vec{x}}(t) = \mathbb{A}\vec{x}(t) + \vec{f}(t) + \vec{\xi}(t), \tag{9}$$

where $\mathbb{A}$ is a matrix whose eigenvalues have negative real part to ensure stability. The external perturbation consists of the set of forces $f_i(t)$ $(i = 1, 2, \ldots, N)$. Finally, Markovian dynamics requires $\xi_i(t)$ to be white noises with zero mean value and arbitrary correlation matrix $\mathbb{B}$: $\langle \xi_i(t)\xi_j(t')\rangle = \mathbb{B}_{ij}\delta(t-t')$. Our reference state is the stationary solution of Eq. (9) with zero force. The solution of Eq. (9) for constant forces is a set of Gaussian variables with average $\langle \vec{x}\rangle_{\rm ss} = -\mathbb{A}^{-1}\vec{f}$ and correlation matrix $\Sigma_{ij} = \langle x_i x_j\rangle_{\rm ss}$, obeying $\Sigma\mathbb{A} + \mathbb{A}^T\Sigma + \mathbb{B} = 0$, where $\mathbb{A}^T$ denotes the transpose of $\mathbb{A}$. Notice that Σ does not depend on the forces $\vec{f}$. The equilibrium potential reads

$$\phi(\vec{x}; \vec{f}) = \frac{(\vec{x} + \mathbb{A}^{-1}\vec{f})^T\Sigma^{-1}(\vec{x} + \mathbb{A}^{-1}\vec{f})}{2} + \frac{1}{2}\log|\Sigma| + \frac{N}{2}\log(2\pi). \tag{10}$$

From this potential we obtain the conjugated variables to the external forces:

$$\vec{X} = \frac{\partial \phi}{\partial \vec{f}}\bigg|_{\vec{f}=0} = [\mathbb{A}^{-1}]^T\Sigma^{-1}\vec{x}. \tag{11}$$

To analyze the validity of our theorem, we need the correlation functions in the stationary state for $\vec{f} = 0$, as well as the response functions for small forces. It is convenient to calculate first these functions for the original variables $\vec{x}(t)$. The solution of Eq. (9) is

$$\vec{x}(t) = e^{\mathbb{A}t}\vec{x}(0) + \int_0^t e^{\mathbb{A}(t-t')}[\vec{f}(t') + \vec{\xi}(t')]dt'. \tag{12}$$

The response function follows immediately:

$$\chi_{\vec{x}\vec{x}}(t) = e^{\mathbb{A}t}. \tag{13}$$

For the correlation function, we take $\vec{f} = 0$ and stationary initial conditions in (12), yielding

$$C_{\vec{x}\vec{x}}(t) = \langle \vec{x}(t)\vec{x}(0)^T\rangle_{\rm ss} = e^{\mathbb{A}t}\Sigma. \tag{14}$$

Since $de^{\mathbb{A}t}/dt = \mathbb{A}e^{\mathbb{A}t} = e^{\mathbb{A}t}\mathbb{A}$, Eqs. (13) and (14), obey a fluctuation-response relation only if Σ is proportional to $\mathbb{A}^{-1}$, i.e., if $\mathbb{A}$ is symmetric and $\mathbb{B}$ proportional to identity (equilibrium case). On the other hand, for any linear combination $\vec{y} = \mathbb{U}\vec{x}$ of the original variables, we have

$$C_{\vec{y}\vec{y}}(t) = \mathbb{U}\langle \vec{x}(t)\vec{x}(0)^T\rangle\mathbb{U}^T = \mathbb{U}e^{\mathbb{A}t}\Sigma\mathbb{U}^T \qquad \chi_{\vec{y}\vec{y}}(t) = \mathbb{U}e^{\mathbb{A}t}. \tag{15}$$

Variables $\vec{y}$ obey the fluctuation-response relation if and only if $\mathbb{A}\Sigma\mathbb{U}^T = \mathbb{I}$, or $\mathbb{U} = [\mathbb{A}^{-1}]^T\Sigma^{-1}$, exactly the conjugated variables found in Eq. (11). The nonequilibrium potential ϕ therefore defines by Eq. (11) the appropriate linear transformation of the original variables $\vec{x}$ which restores the fluctuation-response relation.

We now extend our analysis of molecular motors to time ranges comparable to the slowest motor relaxation time. We split the noise into a delta correlated thermal part and a correlated part describing motor force fluctuations. This choice corresponds to the experiments of Ref. [7] in active gels. The equation of motion reads

$$\dot{x}(t) = -k[x(t) - x_s] + \eta_m(t) + \eta_b(t), \tag{16}$$

where $\eta_m(t)$ and $\eta_b(t)$ are, respectively, the motor and the Brownian noises which are uncorrelated and $\eta_b(t)$ obeys the same relations as in our first example (7), where D is replaced by D_b. The motor noise has zero mean $\langle \eta_m(t)\rangle = 0$ and $\langle \eta_m(t)\eta_m(t')\rangle = \frac{D_m}{\tau_m}\exp(-\frac{t-t'}{\tau_m})$.

The system described by Eq. (16) has *a priori* no reason to obey either the fluctuation-dissipation theorem or the fluctuation-response theorem, since it has both nonequilibrium noise and non-Markovian dynamics. Reference [7] shows clearly how the conventional fluctuation dissipation is broken. One can, however, introduce a new dynamical variable η_m and the associated equation leading to Markovian dynamics, namely,

$$\tau_m\dot{\eta}_m(t) = -\eta_m(t) + \zeta_m(t). \tag{17}$$

With $\langle \zeta_m(t)\rangle = 0$ and $\langle \zeta_m(t)\zeta_m(t')\rangle = 2D_m\delta(t-t')$, the set of equations (16) and (17) becomes Markovian, and the fluctuation-response theorem must apply with any choice of control parameters and suitable definitions of the corresponding observables. We choose as control parameters the forces f_x and f_η:

$$\begin{aligned}\dot{x}(t) &= -k[x(t) - x_s] + f_x + \eta_m(t) + \eta_b(t)\\ \tau_m\dot{\eta}_m(t) &= -\eta_m(t) + f_{\eta_m} + \zeta_m(t).\end{aligned} \tag{18}$$

This is a special case of the general linear stochastic dynamics introduced in Eq. (9). Using the observables defined by Eq. (11), one can straightforwardly check the theorem. Experiments testing the corresponding predictions for the correlation and response functions would be very hard to do on linear molecular motors that are never infinitely processive and detach after a finite time from their filament. However they could be performed on rotatory motors such as F1-ATPase [15], angles and torques replacing displacements and forces.

We now turn to our third example: Hopf bifurcations. With a suitable choice of variables, they are described by a two-variable dynamical system of the form

$$\begin{aligned} \frac{dx}{dt} &= -rx - \omega_0 y + f_x + \eta_x \\ \frac{dy}{dt} &= \omega_0 x - ry + f_y + \eta_y. \end{aligned} \tag{19}$$

A complete study requires the appropriate nonlinear terms [6,8,9], but Eqs. (19) hold for any system close to a Hopf bifurcation on the nonoscillating side, provided one is interested only in the linear response and two point correlation functions, and that coefficients and noise in Eqs. (19) are renormalized quantities. For $r > 0$ the system is in a steady state, all observables having a constant average value, while for $r < 0$, the system is in an oscillating state.

With the conventional choice of x, y as observables and f_x, f_y as forces, the fluctuation-dissipation theorem is broken in a spectacular way best illustrated in the frequency domain. At low frequency the system gives energy to the external world, and as a result the imaginary part $\tilde{\chi}''_{xx}$ of the response function $\tilde{\chi}_{xx}$ is negative. At high frequency, the system has a passive behavior, energy is dissipated, and $\tilde{\chi}''_{xx}$ is positive. It changes sign at the frequencies $\omega^2 = \omega_0^2 - r^2$. Since the power spectrum is positive for all frequencies, an effective temperature, defined as $T^{\mathrm{eff}} = \frac{\omega C xx(\omega)}{k_B \tilde{\chi}''_{xx}(\omega)}$, takes negative values at low frequencies, diverges when $\tilde{\chi}''_{xx}(\omega)$ vanishes, and becomes positive at higher frequencies [6]. Using the observables X and Y defined by Eq. (11), the corresponding response and correlation functions satisfy as expected the fluctuation-response theorem. The real time expressions do not give any clue as to why the equality possibly holds for all frequencies. This is possible if the imaginary part of $\tilde{\chi}_{XX}$ changes sign only at $\omega = 0$ so that $\tilde{\chi}''_{XX}(\omega)/\omega > 0$ and finite. Indeed Eq. (6) insures that there is no sign change of $\tilde{\chi}''_{XX}(\omega)/\omega$.

This fluctuation-response theorem is useful in many ways. First, it is much easier to test than the full identity Eq. (1). Second, although we have given examples with linear dynamics and Gaussian statistics, it holds irrespective of the existence of nonlinearities and of the spatial dimension, provided the dynamics is Markovian. When all slow variables are identified, it will be of similar use as the fluctuation-dissipation theorem.

Our example on correlated noise suggests an additional role: in a given experiment, one could start by observing a violation of the theorem and subsequently increase the number of measured variables up to the point where the dynamics becomes Markovian. In the linear response regime, the variables can be redefined with respect to their steady-state values, and our treatment of linear stochastic dynamics proves that a suitable linear combination of these new variables satisfies the fluctuation-response theorem. This procedure is currently achievable in many experimental situations and thus could be used to identify the number of slow variables in a given dynamical system.

Many equations describing chaotic systems and the "noisy" Navier-Stokes equation correspond to Markovian dynamics: whenever steady states are reached, they obey the fluctuation-response theorem, which could be used to identify the slow variables. This is the case of turbulence in finite geometries [16]. Eventually, the dynamics of quantum systems being Markovian, they should also satisfy the real time version of the theorem.

We are grateful to D. Lacoste for pointing out to us Ref. [11]. J. M. R. P. acknowledges financial support from grant MOSAICO (Ministerio de Educación y Ciencia, Spain).

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