Sum rule for response function in nonequilibrium Langevin systems

Tatsuro Yuge*

IIAIR, Tohoku University, Aoba-ku, Sendai 980-8578, Japan

(Received 9 August 2010; published 24 November 2010)

We derive general properties of the linear-response functions of nonequilibrium steady states in Langevin systems. These correspond to extension of the results which were recently found in Hamiltonian systems [A. Shimizu and T. Yuge, J. Phys. Soc. Jpn. **79**, 013002 (2010)]. We discuss one of the properties, the sum rule for the response function, in particular detail. We show that the sum rule for the response function of the velocity holds in the underdamped case, whereas it is violated in the overdamped case. This implies that the overdamped Langevin models should be used with great care. We also investigate the relation of the sum rule to an equality on the energy dissipation in nonequilibrium Langevin systems, which was derived by Harada and Sasa.

DOI: 10.1103/PhysRevE.82.051130 PACS number(s): 05.70.Ln, 05.40.−a

I. INTRODUCTION

Responses to weak perturbations contain important information on the physical properties of systems and are often used to characterize their states. General properties of the response function are useful in investigating the consistency of obtained results both in theories and in experiments and in making prediction from them. For equilibrium states many general properties are well known, which are based on the linear-response theory [1]. Much interest has been devoted recently to extending the properties to nonequilibrium steady states (NESSs) [2–12]. Several properties of the linear-response function of NESSs were found by Shimizu and the author in general Hamiltonian systems [12]. These properties consist of experimentally measurable quantities only, which is a distinguished feature from the formal results in many other works. One of the general properties is the sum rule for the response function [12]:

$$\int_{-\infty}^{\infty} \mathrm{Re}\ \tilde{\Phi}_B^A(\omega;F)\frac{d\omega}{\pi} = \langle \{B,A\}_{\mathrm{P}}\rangle_{F,0}, \tag{1}$$

where A is a physical observable, $-B$ is a perturbation potential, and $\tilde{\Phi}_B^A$ is the Fourier-Laplace transform of the linear response function of A. $\langle\cdots\rangle_{F,0}$ represents the average in a NESS with a driving force (pump field) F and without the perturbation (probe force), and $\{\bullet,\bullet\}_{\mathrm{P}}$ the Poisson bracket in classical systems (or the commutator divided by $i\hbar$ in quantum systems), which often reduces to an easily measurable quantity.

In Ref. [12] they derived these properties assuming that a large system (which is composed of the system of interest, its environments and a driving source) is a (deterministic) Hamiltonian system. The applicable range of the properties is quite large since almost no assumptions were imposed. It is not trivial, however, that the properties hold in stochastic systems. There are two points to examine the validation in such systems. One is that explicit derivation of the properties in stochastic systems extends the universality of the properties. The other is as follows. Some classes of stochastic systems may be regarded as approximate models of certain large Hamiltonian systems, where the degrees of freedom in the environments are eliminated (although in some cases the connections between these models are not sufficiently clear). Therefore the validation is utilized as a criterion for effectiveness of the approximate models because the properties should remain valid if the approximations in reducing the original Hamiltonian models to the stochastic models are good.

In this paper we show that the sum rule holds in the stochastic systems described by the Langevin equations. It is valid in highly nonequilibrium steady states as well as in equilibrium states. Here, "highly nonequilibrium" is used in the following two senses: (1) the driving force may be arbitrarily large and (2) the intensity of the Langevin noise is allowed not to satisfy the second fluctuation-dissipation relation (as in the case of nonequilibrium process in laser physics [13]). We also derive the asymptotic behavior, another general property shown in Hamiltonian systems [12], of response function in the Langevin systems. Furthermore we show that the sum rule for the velocity response function is violated in the overdamped cases. This implies that the overdamped model sometimes gives results inconsistent with those in larger Hamiltonian systems especially when they are related to small time scale phenomena (which are correctly described by the Hamiltonian systems). We also discuss a relation between the sum rule and the Harada-Sasa equality [2,3], which is an expression of the energy dissipation in the Langevin models.

II. MODEL

We define a general form of the Langevin model. The examples are shown in Sec. IV.

We consider a system the state of which is specified by a set of n stochastic variables denoted by $\boldsymbol{\zeta}=(\zeta_1,\zeta_2,\ldots,\zeta_n)$ (called system variables hereafter). The dynamics of the system is assumed to be characterized by the following stochastic differential equation [14] of a general form:

*yuge@m.tains.tohoku.ac.jp

$$d\zeta_i(t) = M_i(\boldsymbol{\zeta}(t);F)dt + \sum_{j=1}^{n} \mathcal{N}_{ij}(\boldsymbol{\zeta}(t);F)\cdot dW_j(t) + \varepsilon f_i^p(t)K_i(\boldsymbol{\zeta}(t))dt. \quad (2)$$

The first two terms in the right-hand side describe the unperturbed dynamics of the system. F represents a pump field, which determines the degree of nonequilibrium of the system (e.g., an external driving force or a temperature difference). The first term is a deterministic part (M_i is a certain known function of $\boldsymbol{\zeta}$ and F). The second one represents the noise term, where $W_i(t)$ is a Wiener process. We assume that the mean of $W_i(t)$ is zero for any i and that $W_i(t)$ and $W_j(t)$ are uncorrelated if $i\neq j$. The symbol "·" implies the multiplication in the sense of Itô. The noise intensity is determined by $\mathcal{N}_{ij}$ (which is a known function of $\boldsymbol{\zeta}$ and F). The third term represents the probe force (perturbation) the response to which is of our interest (f_i^p and K_i are known functions of t and of $\boldsymbol{\zeta}$, respectively). We assume that in the unperturbed system (i.e., in the case of $\varepsilon=0$) is realized a certain steady state which is stable against perturbations.

A situation to be supposed is as follows. The system of our interest is driven to a NESS by the pump field F. The steady state may be in a nonlinear response regime, in a linear-response regime, or an equilibrium state, because F is allowed to be arbitrary (may be very large) and the noise intensity is arbitrary. To measure the response of the NESS, the probe force $\varepsilon f_j^p K_j$ is applied to the system in addition to F. (This is sometimes called pump-probe experiment.) The linear-response function to the perturbation $\varepsilon f_j^p K_j$ is defined by

$$\Phi_{K_j}^A(t-s;F) = \left.\frac{\delta\langle A(\boldsymbol{\zeta}(t))\rangle_{F,\varepsilon}}{\varepsilon\,\delta f_j^p(s)}\right|_{\varepsilon=0}. \quad (3)$$

Here A is a physical observable which is a function of the system variables $\boldsymbol{\zeta}$, and $\langle\cdots\rangle_{F,\varepsilon}$ represents the average in a state with the parameter F and the perturbation. It should be noted that the response function thus defined is that of a NESS which may be in a nonlinear response regime because F is allowed to be large (irrespective of ε). [The meaning of "linear" in the linear-response function of the NESS is the linear order in $\varepsilon f_j^p K_j$ (not F).]

III. GENERAL RESULT

A. Sum rule for response function

We now show the sum rule for the response function in the Langevin system. For this purpose we consider the stochastic differential equation for the observable A. This is given by the Itô formula [14]:

$$dA(\boldsymbol{\zeta}(t)) = \hat{\Lambda}A(\boldsymbol{\zeta}(t))dt + \sum_{i=1}^{n}\sum_{j=1}^{n} \mathcal{N}_{ij}(\boldsymbol{\zeta}(t);F)\frac{\partial A}{\partial \zeta_i}(\boldsymbol{\zeta}(t))\cdot dW_j(t) + \varepsilon\int_{i=1}^{n} f_i^p(t)K_i(\zeta(t))\frac{\partial A}{\partial \zeta_i}(\boldsymbol{\zeta}(t))dt, \quad (4)$$

where the backward operator $\hat{\Lambda}$ is defined by

$$\hat{\Lambda} = \sum_{i=1}^{n} M_i(\boldsymbol{\zeta};F)\frac{\partial}{\partial \zeta_i} + \frac{1}{2}\sum_{i=1}^{n}\sum_{j=1}^{n}\sum_{k=1}^{n}\mathcal{N}_{ik}(\boldsymbol{\zeta};F)\mathcal{N}_{jk}(\boldsymbol{\zeta};F)\frac{\partial^2}{\partial\zeta_i\,\partial\zeta_j}. \quad (5)$$

Employing a method similar to that in Ref. [3], we transform this differential equation into an integral form:

$$A(\boldsymbol{\zeta}(t)) = e^{(t-t_0)\hat{\Lambda}}A(\boldsymbol{\zeta}(t_0)) + \sum_{i=1}^{n}\sum_{j=1}^{n}\int_{t_0}^{t}\mathcal{N}_{ij}(\boldsymbol{\zeta}(t');F)\frac{\partial}{\partial\zeta_i}e^{(t-t')\hat{\Lambda}}A(\boldsymbol{\zeta}(t'))\cdot dW_j(t') + \varepsilon\sum_{i=1}^{n}\int_{t_0}^{t} f_i^p(t')K_i(\boldsymbol{\zeta}(t'))\frac{\partial}{\partial\zeta_i}e^{(t-t')\hat{\Lambda}}A(\boldsymbol{\zeta}(t'))dt', \quad (6)$$

where t_0 is an initial time. For completeness we provide the detail of the transformation in the Appendix.

The average of the second term in the right-hand side of Eq. (6) vanishes because the integrand is a nonanticipating function (the mean value formula of the Itô stochastic integral) [14]. Taking the average of Eq. (6), we thus obtain

$$\langle A(\boldsymbol{\zeta}(t))\rangle_{F,\varepsilon} = e^{(t-t_0)\hat{\Lambda}}A(\boldsymbol{\zeta}(t_0)) + \varepsilon\sum_{i=1}^{n}\int_{t_0}^{t} f_i^p(t') \times\left\langle K_i(\boldsymbol{\zeta}(t'))\frac{\partial}{\partial\zeta_i}e^{(t-t')\hat{\Lambda}}A(\boldsymbol{\zeta}(t'))\right\rangle_{F,\varepsilon} dt'. \quad (7)$$

By a functional differentiation of Eq. (7) with respect to $f_j^p(s)$, an expression of the response function is derived:

$$\Phi_{K_j}^A(t-s;F) = \left\langle K_j(\boldsymbol{\zeta}(s))\frac{\partial}{\partial\zeta_j}e^{(t-s)\hat{\Lambda}}A(\boldsymbol{\zeta}(s))\right\rangle_{F,0}, \quad (8)$$

for $t>s$ ($>t_0$), and $\Phi_{K_j}^A(t-s;F)=0$ otherwise. Note that the Langevin system satisfies the causality condition. If $s\gg t_0$, this expression does not depend on s but on $t-s$ only because a steady state is realized in the unperturbed system. Thus we finally obtain the sum rule:

$$\int_{-\infty}^{\infty}\mathrm{Re}\,\tilde{\Phi}_{K_j}^A(\omega;F)\frac{d\omega}{\pi} = \Phi_{K_j}^A(+0;F) = \left\langle K_j(\boldsymbol{\zeta})\frac{\partial A}{\partial\zeta_j}(\boldsymbol{\zeta})\right\rangle_{F,0}, \quad (9)$$

where $\tilde{\Phi}_{K_j}^A(\omega;F)$ is the Fourier-Laplace transform of $\Phi_{K_j}^A(\tau;F)$ and where we have used $\int_{-\infty}^{\infty}\mathrm{Im}\,\tilde{\Phi}_{K_j}^A(\omega;F)d\omega=0$ (this is because $\Phi_{K_j}^A(\tau;F)$ is a real-valued function). This is the main result of the present paper.

B. Asymptotic behavior

In Ref. [12], in addition to the sum rule, an asymptotic behavior of $\tilde{\Phi}_{K_j}^A(\omega;F)$ was derived. We here show another derivation of it using the sum rule. We first note that the response function satisfies a dispersion relation if the system satisfies the causality condition;

$$\mathrm{Im}\,\tilde{\Phi}^A_{K_j}(\omega;F) = -\mathcal{P}\int_{-\infty}^{\infty}\frac{1}{\omega'-\omega}\mathrm{Re}\,\tilde{\Phi}^A_{K_j}(\omega';F)\frac{d\omega'}{\pi}, \tag{10}$$

where $\mathcal{P}$ denotes the principal value. By multiplying ω to the both sides of this equation and by taking the $\omega\to\infty$ limit, we obtain the asymptotic behavior:

$$\lim_{\omega\to\infty}\omega\,\mathrm{Im}\,\tilde{\Phi}^A_{K_j}(\omega;F) = \int_{-\infty}^{\infty}\mathrm{Re}\,\tilde{\Phi}^A_{K_j}(\omega';F)\frac{d\omega'}{\pi} = \left\langle K_j(\zeta)\frac{\partial A}{\partial \zeta_j}(\zeta)\right\rangle_{F,0}, \tag{11}$$

where we have exchanged the integration and the limit procedure in the first equality and have used the sum rule in the second equality. Hence the asymptotic behavior is valid in the systems (including the Langevin systems) where the sum rule and the dispersion relation hold. The asymptotic value is the same as the sum value in the sum rule.

C. Remarks

Here we make remarks on the results.

First, Eq. (9) should be interpreted as a prediction on the sum value (integral value) of $\tilde{\Phi}^A_{K_j}(\omega;F)$ at many different values of ω [therefore Eq. (9) is called sum rule]. $\tilde{\Phi}^A_{K_j}(\omega;F)$ for each ω is easily measured by experiments or numerical simulations, e.g., in the following way: apply a sinusoidal probe force with frequency ω [i.e., $f^p(t)=\sin\omega t$], measure the observable A for sufficiently long time ($\gg 1/\omega$), and calculate the Fourier component $\langle\tilde{A}_\omega\rangle_{F,\varepsilon}$ at ω from the time-series data of A ($\tilde{\Phi}^A_{K_j}(\omega;F)=\lim_{\varepsilon\to 0}\langle\tilde{A}_\omega\rangle_{F,\varepsilon}/\varepsilon$). The sum rule [Eq. (9)] states that the sum value measured in the system with a small perturbation equals the average value of $S(\zeta)\equiv K_j(\zeta)\,\partial A(\zeta)/\partial\zeta_j$ measured in the system without the perturbation. In many cases S is a quantity that is easy to measure. Therefore it is possible to test the sum rule in experiments and simulations. (In Ref. [12] we demonstrated the validity of a sum rule by a numerical simulation.)

Equation (9) also seems to be a prediction on the value of the response function immediately after the probe force is applied, as seen in the middle of Eq. (9). In contrast to the measurement of $\tilde{\Phi}^A_{K_j}(\omega;F)$, however, it is very difficult to measure $\Phi^A_{K_j}(+0;F)$ in experiments and numerical simulations. Therefore it is more natural to interpret Eq. (9) as a sum rule for $\tilde{\Phi}^A_{K_j}(\omega;F)$.

Second, the sum rule holds for any steady states (if they are stable) including the states in a nonlinear response regime (at large F) as well as an equilibrium state ($F=0$). Although the *form* of the rule is the same for all the states, the *value* of the sum may vary as F is changed. This is because the steady state distribution function, which appears in averaging $S(\zeta)$ in Eq. (9), is dependent on F.

Third, the expression of the response function [Eq. (8)] in more concrete examples was derived in some literatures [9,11], although they did not derive the sum rule from it. The expression itself is not so useful because it is hard to calculate in analytic way unless one knows the explicit form of the steady-state distribution function and because it is hard to measure in experiments and numerical simulations due to the complicated factor $e^{(t-s)\hat{\Lambda}}$. In contrast, the rightmost side of Eq. (9) is much easier to measure since it does not contain such factors, and tests of the validity of Eq. (9) are possible as mentioned in the first remark.

Fourth, the sum rule is different from the moment sum rule (e.g., Ref. [5]). The statement of the moment sum rule is that the sum value of $\tilde{\Phi}^A_{K_j}(\omega;F)$ [and $\omega^\lambda\tilde{\Phi}^A_{K_j}(\omega;F)$] converges to a certain value. However, the dependence of this value on F and the other parameters is not given by it, although the dependence on F is most interesting point in nonequilibrium statistical mechanics. In contrast, the statement of the sum rule is not only the convergence of the sum value but also its equivalence to the average value of $S(\zeta)$, which gives the dependence of the sum on several parameters (including F).

S is independent of the state and the system (without probe). (It depends on the probe force and the observable of our interest, both of which may be chosen irrespective of the state and the system.) Therefore there are no differences in S between equilibrium states and NESSs and between noninteracting systems and interacting systems. The dependence on the state and the system appears only in averaging it, which leads to the dependence of the sum on F in general. When S is independent of ζ, in particular, this dependence disappears and the sums are the same for any steady states and for any systems. This fact cannot be predicted by the moment sum rule.

IV. EXAMPLES

In this section we describe some examples of the Langevin model defined in the general form [Eq. (2)] in Sec. II, and see the concrete forms of the sum rule in the examples. It should be noted that in Eq. (2) the relevant quantities to the sum rule are K_is.

Furthermore we show that the sum rule for the velocity response function is violated in the overdamped case.

A. Underdamped case

One of the simplest examples is the single-particle underdamped Langevin model in one dimension which is described by the following equations:

$$\frac{dp(t)}{dt} = -\frac{\gamma}{m}p(t) - \frac{\partial U}{\partial x}(x(t)) + F + \xi(t) + \varepsilon f^p(t)\frac{\partial B}{\partial x}(x(t)), \tag{12}$$

$$\frac{dx(t)}{dt} = \frac{p(t)}{m}. \tag{13}$$

Here, p, x, m, and γ are the momentum, position, mass, and friction coefficient of the particle, respectively. $U(x)$ is a potential, F is an external driving force, and $-B(x)$ is a perturbation potential of the probe. $\xi(t)$ is a white Gaussian noise with zero mean and satisfies $\langle \xi(t)\xi(t')\rangle = 2D\delta(t-t')$. F is allowed to be arbitrarily strong. A nonequilibrium steady state in the nonlinear response regime is realized for large F, while an equilibrium state is realized for $F=0$.

More precise forms of Eqs. (12) and (13) are given by Eq. (2) where $n=2$, $\zeta_1=p$, and $\zeta_2=x$, and where

$$M_1(p,x;F) = -\frac{\gamma}{m}p - \frac{\partial U}{\partial x}(x) + F, \quad M_2(p,x;F) = \frac{p}{m},$$

$$\mathcal{N}_{11}(p,x;F) = \sqrt{2D}, \quad \mathcal{N}_{ij\neq 11}(p,x;F) = 0,$$

$$K_1(p,x) = \frac{\partial B}{\partial x}(x), \quad K_2(p,x) = 0.$$

In this case the sum rule reads

$$\int_{-\infty}^{\infty} \mathrm{Re}\, \tilde{\Phi}_B^A(\omega;F)\frac{d\omega}{\pi} = \left\langle \frac{\partial B}{\partial x}(x)\frac{\partial A}{\partial p}(p,x)\right\rangle_{F,0}. \tag{14}$$

This form is the same as that in the classical Hamiltonian models where the systems of interest are single-particle systems. In particular, when considering the momentum response to the spatially homogeneous probe force (i.e., $A=p$ and $B=x$), we have the F-independent sum value:

$$\int_{-\infty}^{\infty} \mathrm{Re}\, \tilde{\Phi}_x^p(\omega;F)\frac{d\omega}{\pi} = 1. \tag{15}$$

The second example is a three-dimensional many-particle underdamped Langevin model described by

$$\frac{d\vec{p}_\mu(t)}{dt} = -\frac{\gamma_\mu}{m_\mu}\vec{p}_\mu(t) - \frac{\partial U}{\partial \vec{r}}(\vec{r}_\mu(t)) - \frac{\partial V}{\partial \vec{r}_\mu}(\{\vec{r}_\nu(t)\}) + \vec{F} + \vec{\xi}_\mu(t) + \varepsilon f^p(t)\frac{\partial B}{\partial \vec{r}}(\vec{p}_\mu(t),\vec{r}_\mu(t)), \tag{16}$$

$$\frac{d\vec{r}_\mu(t)}{dt} = \frac{\vec{p}_\mu(t)}{m_\mu} - \varepsilon f^p(t)\frac{\partial B}{\partial \vec{p}}(\vec{p}_\mu(t),\vec{r}_\mu(t)), \tag{17}$$

for $\mu=1,2,\ldots,N$. Here, $\vec{a}=(a^1,a^2,a^3)$ represents a three-dimensional vector. $\vec{p}_\mu$, $\vec{r}_\mu$, m_μ, and γ_μ are the momentum, position, mass, and friction coefficient of the μth particle, respectively. $U(\vec{r})$ is a single-particle potential and $V(\{\vec{r}_\nu\})$ is an interparticle potential. $\vec{F}$ is an external driving force, the strength of which is arbitrary. $-B(\vec{p},\vec{r})$ is a probe potential (perturbation) which is assumed to depend on a momentum as well as on a position. (An example is an interaction of an electron with an electromagnetic field.) $\vec{\xi}_\mu(t)$ is a white Gaussian noise with zero mean and satisfies $\langle \xi_\mu^\alpha(t)\xi_{\mu'}^{\alpha'}(t')\rangle = 2D\delta_{\alpha\alpha'}\delta_{\mu\mu'}\delta(t-t')$ $(\alpha,\alpha'=1,2,3)$. More precise forms of these equations are given by Eq. (2) with $n=6N$, $\zeta_{6\mu-6+\alpha}=p_\mu^\alpha$, and $\zeta_{6\mu-3+\alpha}=r_\mu^\alpha$ $(\alpha=1,2,3)$. The relevant quantities K_is to the sum rule are written as

$$K_{6\mu-6+\alpha}(\{\vec{p}_\nu,\vec{r}_\nu\}) = \frac{\partial B}{\partial r^\alpha}(\vec{p}_\mu,\vec{r}_\mu),$$

$$K_{6\mu-3+\alpha}(\{\vec{p}_\nu,\vec{r}_\nu\}) = \frac{\partial B}{\partial p^\alpha}(\vec{p}_\mu,\vec{r}_\mu).$$

We thus obtain the sum rule for the response function $\Phi_B^A(t-s;F) = \delta\langle A(\{\vec{p}_\mu(t),\vec{r}_\mu(t)\})\rangle_{F,0}/\varepsilon\delta f^p(s)|_{\varepsilon=0}$

$$\int_{-\infty}^{\infty} \mathrm{Re}\, \tilde{\Phi}_B^A(\omega;F)\frac{d\omega}{\pi} = \left\langle \sum_{\mu,\alpha}\left[\frac{\partial B}{\partial r_\mu^\alpha}(\{\vec{p}_\nu,\vec{r}_\nu\})\frac{\partial A}{\partial p_\mu^\alpha}(\{\vec{p}_\nu,\vec{r}_\nu\}) - \frac{\partial B}{\partial p_\mu^\alpha}(\{\vec{p}_\nu,\vec{r}_\nu\})\frac{\partial A}{\partial r_\mu^\alpha}(\{\vec{p}_\nu,\vec{r}_\nu\})\right]\right\rangle_{F,0}. \tag{18}$$

This form is the same as that in the classical Hamiltonian models [Eq. (1)]. This reduces to Eq. (14) if $N=1$, $\partial B/\partial p=0$ and the motion is restricted to one dimension.

In Langevin models the noise intensity D is usually assumed to be equal to $\gamma k_B T$ [which is called the second fluctuation-dissipation relation (FDR) [1]], where T is the temperature of the environment and k_B is the Boltzmann constant. However, as seen in the general result and in the above two examples, this assumption is not necessary for the validity of the sum rule because the noise intensity does not explicitly contribute to the rule. This indicates that the sum rule holds in a very wide range of systems from highly nonequilibrium systems (in the sense that the second FDR is violated) to purely deterministic systems ($D=0$). The violation of the second FDR is often seen in the treatment of nonequilibrium process (far from equilibrium) in light-emitting devices by quantum Langevin equation [13].

For the same reason the noise intensity D may depend on F. Therefore the treatment in the present paper is applicable also to the systems in which the second FDR is gradually violated as F increases. Also D may depend on the position of the particles in the two examples and on μ and α in the second example. One of the consequences from this fact is that the sum rule is valid also in heat conducting nonequilibrium systems driven by temperature difference. It should be noted that the *validity* of the sum rule is independent of the noise intensity whereas the *value* of the sum depends on it because the steady states depend on it in general.

Finally we make a brief comment. In some Langevin systems the natural interpretation of the Langevin equations used in physics is the Stratonovich-type stochastic differential equations [15]. In such cases we should first interpret the given Langevin equations to the corresponding Stratonovich-type equations, then transform them into the Itô-type equations [Eq. (2)], and finally apply the general form of the sum rule. Note that the *form* of the sum rule is identical irrespective of the interpretations, although the *value* of the sum depends on them because the steady states may be different in different interpretations. In the above examples (with position-independent D), the values as well as the forms of the sum rules are independent of the interpretations because the forms of the Stratonovich-type and Itô-type equations are the same except for the senses of the multiplications in these cases.

B. Overdamped case

In this subsection we discuss the sum rule in the overdamped case. For simplicity we consider a single-particle model (the extension to many-particle models is straightforward). We consider the single-particle overdamped Langevin model in one dimension which is described by

$$\gamma \frac{dx(t)}{dt} = -\frac{\partial U}{\partial x}(x(t)) + F + \xi(t) + \varepsilon f^p(t)\frac{\partial B}{\partial x}(x(t)). \quad (19)$$

Here the notations are the same as those in Eqs. (12) and (13). $\xi(t)$ is a white Gaussian noise with zero mean and satisfies $\langle \xi(t)\xi(t')\rangle = 2D\delta(t-t')$. A more precise form of Eq. (19) is given by Eq. (2) where $n=1$ and $\zeta_1 = x$ and where

$$M_1(x;F) = -\frac{1}{\gamma}\left[\frac{\partial U}{\partial x}(x) + F\right],$$

$$\mathcal{N}_{11}(x;F) = \frac{\sqrt{2D}}{\gamma},$$

$$K_1(x) = \frac{1}{\gamma}\frac{\partial B}{\partial x}(x).$$

Even in this model the sum rule is valid *if the observable A* is a function of only x;

$$\int_{-\infty}^{\infty} \mathrm{Re}\, \tilde{\Phi}_B^A(\omega;F)\frac{d\omega}{\pi} = \frac{1}{\gamma}\left\langle \frac{\partial B}{\partial x}(x)\frac{\partial A}{\partial x}(x)\right\rangle_{F,0}. \quad (20)$$

One sometimes considers as an observable of interest the velocity v, which is defined by $v(t)dt = dx(t)$ in the overdamped model. Because we have not derived the sum rule in the cases that the observable is a function of the differentials of the system variables, the general result [Eq. (9)] does not ensure that the sum rule holds for the response function Φ_B^v of the velocity in this model. However, from the viewpoint that the overdamped model is regarded as a coarse-grained model of the underdamped one, Φ_B^v must satisfy the sum rule if the coarse-graining procedure is good.

To investigate this point we again consider the underdamped model described by Eqs. (12) and (13). Because the momentum of the particle is included in the system variables in the underdamped model, we can safely consider the sum rule for the response function of the velocity p/m, which reads

$$\int_{-\infty}^{\infty} \mathrm{Re}\, \tilde{\Phi}_B^{p/m}(\omega;F)\frac{d\omega}{\pi} = \frac{\gamma}{m}\frac{1}{\gamma}\left\langle \frac{\partial B}{\partial x}\right\rangle_{F,0}. \quad (21)$$

The right-hand side diverges in the overdamped limit $(m/\gamma \to 0)$. Therefore a necessary condition for the validity of the sum rule for Φ_B^v is that $\Phi_B^v(+0;F)$ is divergent in the overdamped model. We next examine this condition directly in the overdamped model. Φ_B^v is calculated by averaging Eq. (19) and by functionally differentiating the result with respect to $f(s)$:

$$\Phi_B^v(t-s;F) = \frac{\delta}{\varepsilon\,\delta f(s)}\left\langle \frac{dx(t)}{dt}\right\rangle_{F,\varepsilon}\Bigg|_{\varepsilon=0}$$
$$= \frac{1}{\gamma^2}\left\langle \frac{\partial B}{\partial x}[x(s)]\frac{\partial}{\partial x}e^{(t-s)\hat{\Lambda}}\frac{\partial U}{\partial x}[x(s)]\right\rangle_{F,0}$$
$$+ \frac{1}{\gamma}\left\langle \frac{\partial B}{\partial x}(x)\right\rangle_{F,0}\delta(t-s), \quad (22)$$

where we have used the expression [Eq. (8)] in the first term in the rightmost side. Owing to the second term, $\Phi_B^v(+0;F)$ is divergent [16]. Thus the sum rule for Φ_B^v seems to be valid in the overdamped model in the sense that the both sides of it are divergent. More careful analysis, however, reveals that the diverging behaviors are different between Eq. (22) and the overdamped limit of Eq. (21). The diverging behavior of Eq. (22) is dominated by the delta function in the second term in the rightmost side. This comes from the functional derivative $\delta f(t)/\delta f(s)$, the order of which is estimated as $O(1/\Delta t)$. Here Δt is the smallest time scale of our observation on the system. On the other hand, the diverging behavior of the overdamped limit of Eq. (21) is dominated by the factor γ/m in the right-hand side. Since the overdamped Langevin model should be interpreted as an effective description of the underdamped one in the time scale $\Delta t \gg m/\gamma$, one must first take the $m/\gamma \to 0$ limit and then take the $\Delta t \to 0$ limit to have continuous time limit of the overdamped model. Therefore the diverging behavior is stronger in the overdamped limit of Eq. (21) $[O(\gamma/m)]$ than in Eq. (22) $[O(1/\Delta t)]$. In this sense the sum rule for Φ_B^v is violated in the overdamped model. This is consistent with the fact that the overdamped Langevin model is valid (as a coarse-grained model of the underdamped one) only in the frequency range of $\omega \ll \gamma/m$ (which would result in an incorrect contribution to the sum from the higher frequency region).

V. RELATION TO THE HARADA-SASA EQUALITY

In Refs. [2,3], Harada and Sasa derived an equality (the Harada-Sasa equality) on the energy dissipation rate in nonequilibrium Langevin systems. We here discuss the relation between this equality and the sum rule.

For simplicity we consider the single-particle underdamped Langevin model described by Eqs. (12) and (13). In this model the energy dissipation rate J from the system to the environment is defined by [18,15]

$$J(t)dt = \left(\frac{\gamma}{m}p(t) - \xi(t)\right) \circ dx(t), \quad (23)$$

where the symbol "$\circ$" represents the multiplication in the sense of the Stratonovich interpretation. The Harada-Sasa equality is an expression of the average of J:

$$\langle J\rangle_{F,0} = \frac{\gamma}{m^2}\langle p\rangle_{F,0}{}^2 + \frac{\gamma}{m^2}\int_{-\infty}^{\infty}[\tilde{C}^p(\omega;F)$$
$$- 2mk_{\mathrm{B}}T\, \mathrm{Re}\, \tilde{\Phi}_x^p(\omega;F)]\frac{d\omega}{2\pi}, \quad (24)$$

where the usual assumption (the second FDR) $D = \gamma k_{\mathrm{B}} T$ on the noise intensity is imposed. $\tilde{C}^p(\omega;F)$ is the Fourier transform of the time-correlation function $C^p(\tau;F)$ of the momentum; $C^p(\tau;F)=\langle(p(\tau)-\langle p\rangle_{F,0})(p(0)-\langle p\rangle_{F,0})\rangle_{F,0}$.

We show that this equality is derived with the help of the sum rule. First we note that Eq. (23) is rewritten in the Itô type: $J(t)dt=[\gamma p^2(t)/m^2-D/m]dt-(\sqrt{2D}/m)p(t)dW(t)$. This is derived by substituting "$\xi(t)=\sqrt{2D}dW(t)/dt$" and $dx(t)=p(t)dt/m$ into Eq. (23) and by using the Stratonovich-Itô transformation [14]. Since the average of the last term vanishes due to the mean value formula, the average dissipation rate is written in a simple form [19,15]:

$$\langle J\rangle_{F,0}=\frac{\gamma}{m^2}\langle p^2\rangle_{F,0}-\frac{D}{m}. \tag{25}$$

By multiplying $\int_{-\infty}^{\infty}\mathrm{Re}\,\tilde{\Phi}_x^p(\omega;F)d\omega/\pi$, which is equal to 1 owing to the sum rule [see Eq. (15)], to the last term, and by noting $\langle p^2\rangle_{F,0}=\langle p\rangle_{F,0}{}^2+\int_{-\infty}^{\infty}\tilde{C}^p(\omega;F)d\omega/2\pi$, we obtain

$$\langle J\rangle_{F,0}=\frac{\gamma}{m^2}\langle p\rangle_{F,0}{}^2+\frac{\gamma}{m^2}\int_{-\infty}^{\infty}\left[\tilde{C}^p(\omega;F)-\frac{2mD}{\gamma}\mathrm{Re}\,\tilde{\Phi}_x^p(\omega;F)\right]\frac{d\omega}{2\pi}. \tag{26}$$

This becomes the Harada-Sasa equality if $D=\gamma k_B T$ [20].

As seen in the above derivation, the validation of the Harada-Sasa equality requires that Eqs. (15) and (25) hold. The former holds in a wide range of nonequilibrium systems (even in systems other than Langevin systems) because it is a specific form of the sum rule. In contrast, the validity range of the latter is not so large. The meaning of Eq. (25) with $D=\gamma k_B T$ is that the ratio of the average dissipation rate and the difference between the kinetic energy of the system and the environment temperature, $\langle J\rangle_{F,0}/(\langle p^2\rangle_{F,0}/m-k_BT)$, is a constant, γ/m, which is independent of F. In some systems other than Langevin systems, however, this does not hold, especially in states far from equilibrium. For example, in a numerical simulation of a model of electrical conduction [21,22], it is clearly seen that $\langle J\rangle_{F,0}/(\langle p^2\rangle_{F,0}/m-k_BT)$ does depend on F [23]. In this sense the validity range of the Harada-Sasa equality is restricted to the systems in which the ratio is constant. A sufficient condition for this might be the distinct separation of time scales, as they mentioned in Ref. [3]. It should be noted, however, that even when the ratio is not constant there remains another possibility. That is, it might be possible that one can define an F dependent $\gamma(F)$ in a certain way and that $\gamma(F)/m$ equals the ratio $\langle J\rangle_{F,0}/(\langle p^2\rangle_{F,0}/m-k_BT)$ in the steady state at each F. Whether this is true or not should be tested in systems (e.g., in the model in Refs. [21,22]) which cannot be described by Langevin equations.

VI. SUMMARY

In this paper we extended the validity range of the sum rule (and the asymptotic behavior) for the linear-response function of steady states to a class of stochastic models described by a general form of the Langevin equations. This holds for a wide range of the steady states (if they are stable) including highly nonequilibrium states as well as equilibrium states because the driving force is allowed to be large (e.g., to be in a nonlinear response regime) and the noise intensity may be arbitrary (e.g., not to satisfy the second fluctuation-dissipation relation). The sum rule is a property which normal nonequilibrium models should have and therefore is used as a touchstone to examine the correctness of results in experiments and theories of NESSs. In the overdamped Langevin model the sum rule for the velocity response function does not hold, which suggests that results in the overdamped model should be treated with care if they are concerned with small time scales.

We also showed the relation of the sum rule to the Harada-Sasa equality. The equality is reduced to a simpler form when one uses the sum rule with a specific choice of observable.

Further extension of the validity range of the sum rule remains as theoretical issues. Extension to time-dependent case where M_i and $\mathcal{N}_{ij}$ in Eq. (2) explicitly depend on t is straightforward. It would be also interesting to examine the validation of the sum rule in non-Markovian models. It is easily extended to the non-Markovian models which become Markovian if appropriate dynamical variables are added to the original system variables. For other non-Markovian models, the method used in Refs. [24,25]. to generalize the Harada-Sasa equality to non-Markovian cases might give hints. Finally, it is also important to investigate higher-order responses of NESSs in stochastic systems as studied in Hamiltonian systems recently [26].

ACKNOWLEDGMENTS

The author thanks A. Shimizu for helpful discussions and critical reading of the manuscript. This work was supported by the Grant-in-Aid for the GCOE Program "Weaving Science Web beyond Particle-Matter Hierarchy."

APPENDIX: DERIVATION OF EQ. (6)

We introduce the time-evolution operator $\hat{\Omega}(t)$ corresponding to Eq. (4); for an arbitrary function C of ζ, the value at $\zeta(t)$ is given by $C(\zeta(t))=\hat{\Omega}(t)C(\zeta(t_0))$, where t_0 is the initial time and $\hat{\Omega}(t_0)=1$. Then Eq. (4) is rewritten as

$$d\hat{\Omega}(t)A(\boldsymbol{\zeta}(t_0))=\hat{\Omega}(t)(\hat{\Lambda}A)(\boldsymbol{\zeta}(t_0))dt+\sum_{i=1}^{n}\sum_{j=1}^{n}\hat{\Omega}(t)\left(\mathcal{N}_{ij}\frac{\partial A}{\partial\zeta_i}\right)(\zeta(t_0))\cdot dW_j(t)+\varepsilon\sum_{i=1}^{n}f_i^p(t)\hat{\Omega}(t)\left(K_i\frac{\partial A}{\partial\zeta_i}\right)(\zeta(t_0))dt. \tag{A1}$$

Because A is arbitrary the above equation is regarded as a stochastic differential equation for $\hat{\Omega}$. In order to transform this equation into an integral equation, we introduce an operator, $\check{\Omega}(t)=\hat{\Omega}(t)e^{-(t-t_0)\hat{\Lambda}}$. From Eq. (A1) the differential equation for $\check{\Omega}$ is given by

$$d\check{\Omega}(t) = \sum_{i=1}^{n}\sum_{j=1}^{n}\hat{\Omega}(t)\left[\mathcal{N}_{ij}\frac{\partial}{\partial\zeta_i}e^{-(t-t_0)\hat{\Lambda}}\right]\cdot dW_j(t) + \varepsilon\sum_{i=1}^{n}f_i^p(t)\hat{\Omega}(t)\times\left[K_i\frac{\partial}{\partial\zeta_i}e^{-(t-t_0)\hat{\Lambda}}\right]dt. \tag{A2}$$

Then formally integrating this equation from t_0 to t with the initial condition $\check{\Omega}(t_0)=1$ and multiplying $e^{(t-t_0)\hat{\Lambda}}$ from the right, we obtain

$$\hat{\Omega}(t) = e^{(t-t_0)\hat{\Lambda}} + \sum_{i=1}^{n}\sum_{j=1}^{n}\int_{t_0}^{t}\hat{\Omega}(t')\left[\mathcal{N}_{ij}\frac{\partial}{\partial\zeta_i}e^{(t-t')\hat{\Lambda}}\right]\cdot dW_j(t') + \varepsilon\sum_{i=1}^{n}\int_{t_0}^{t}f_i^p(t')\hat{\Omega}(t')\left[K_i\frac{\partial}{\partial\zeta_i}e^{(t-t')\hat{\Lambda}}\right]dt'. \tag{A3}$$

Acting this equation on $A(\zeta(t_0))$, we finally get Eq. (6).

[1] R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II: Nonequilibrium Statistical Mechanics* (Springer-Verlag, Berlin, 1991).
[2] T. Harada and S.-i. Sasa, Phys. Rev. Lett. **95**, 130602 (2005).
[3] T. Harada and S.-i. Sasa, Phys. Rev. E **73**, 026131 (2006).
[4] T. Speck and U. Seifert, Europhys. Lett. **74**, 391 (2006).
[5] V. Lucarini, J. Stat. Phys. **131**, 543 (2008).
[6] J. Prost, J.-F. Joanny, and J. M. R. Parrondo, Phys. Rev. Lett. **103**, 090601 (2009).
[7] M. Baiesi, C. Maes, and B. Wynants, Phys. Rev. Lett. **103**, 010602 (2009).
[8] M. Baiesi, C. Maes, and B. Wynants, J. Stat. Phys. **137**, 1094 (2009).
[9] M. Baiesi, E. Boksenbojm, C. Maes, and B. Wynants, J. Stat. Phys. **139**, 492 (2010).
[10] D. Villamaina, A. Baldassarri, A. Puglisi, and A. Vulpiani, J. Stat. Mech.: Theory Exp. (2009), P07024.
[11] M. Krüger and M. Fuchs, Phys. Rev. Lett. **102**, 135701 (2009).
[12] A. Shimizu and T. Yuge, J. Phys. Soc. Jpn. **79**, 013002 (2010).
[13] H. Fujisaki and A. Shimizu, Phys. Rev. A **57**, 3074 (1998).
[14] C. W. Gardiner, *Handbook of Stochastic Methods for Physics, Chemistry, and the Natural Sciences* (Springer-Verlag, Berlin, 2004).
[15] K. Sekimoto, *Stochastic Energetics* (Springer, Berlin, 2010).
[16] From the viewpoint that the overdamped model is an approximate one of a certain Hamiltonian model, the infinite value of the sum in the overdamped model implies the violation of the sum rule because it must be finite in Hamiltonian systems [12]. Furthermore, since all the systems in experiments are described by Hamiltonian systems (if the degrees of freedom in sufficiently large environments are included), it should be experimentally verified (e.g., in experiments of the Brownian motion of a bead in air [17]) that the overdamped model gives an incorrect result about the sum rule.
[17] T. Li, S. Kheifets, D. Medellin, and M. G. Raizen, Science **328**, 1673 (2010).
[18] K. Sekimoto, J. Phys. Soc. Jpn. **66**, 1234 (1997).
[19] H. Hasegawa and T. Nakagomi, J. Stat. Phys. **21**, 191 (1979).
[20] This equivalence was pointed out also in their original paper [3].
[21] T. Yuge, N. Ito, and A. Shimizu, J. Phys. Soc. Jpn. **74**, 1895 (2005).
[22] T. Yuge and A. Shimizu, Prog. Theor. Phys. Suppl. **178**, 64 (2009).
[23] T. Yuge (unpublished).
[24] J. M. Deutsch and O. Narayan, Phys. Rev. E **74**, 026112 (2006).
[25] T. Ohkuma and T. Ohta, J. Stat. Mech.: Theory Exp. (2007), P10010.
[26] A. Shimizu, J. Phys. Soc. Jpn. **79**, 113001 (2010).