General Properties of Response Functions of Nonequilibrium Steady States

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We derive general properties, which hold for both quantum and classical systems, of response functions of nonequilibrium steady states. We clarify differences from those of equilibrium states. In particular, sum rules and asymptotic behaviors are derived, and their implications are discussed. Since almost no assumptions are made, our results are applicable to diverse physical systems. We also demonstrate our results by a molecular dynamics simulation of a many-body interacting system.

KEYWORDS: sum rule, asymptotic behavior, reciprocal relation, fluctuation-dissipation relation

Stable states of macroscopic systems can be well characterized by their responses to external probe fields. When the probe fields are weak, the responses are linear functions of the probe fields. General properties of the linear response functions are well-known for equilibrium states.1, 2 In contrast, those for nonequilibrium steady states (NESSs) are not well understood yet, although many attempts have been made.3–13 For example, perturbation expansion of the density operator $\hat{\rho}_F$ of NESSs in terms of a driving field $F$ has often been employed.4 This gives linear and higher-order ($n = 2, 3, \cdots$) response functions (denoted by $\Phi^{(n)}_{\text{eq}}$ and $\Phi^{(n)}_{\text{eq}}$, respectively) of equilibrium states. General properties, such as symmetries, of $\Phi^{(n)}_{\text{eq}}$ were thus derived.4 Similar results were also obtained from the fluctuation theorems.6 However, it is generally hard to obtain linear response functions $\Phi_F$ of NESSs from $\Phi^{(n)}_{\text{eq}}$ because such expansion converges only slowly (or does not converge) for $F$ that is large enough to drive NESSs of interest. Another approach is to utilize some general expression of $\Phi_F$. Such an expression was derived, e.g., in ref. 7. However, it contains the expectation value of a function which is unknown except for simple cases.14 To derive physical results for $\Phi_F$ from such a formal expression, simplifying assumptions were made,7 at the expense of generality. Furthermore, one may expect that $\Phi_F$ could be expressed by small fluctuation in NESSs. However, refs. 8 and 9 showed that $\Phi_F$ of finite macroscopic systems is not a universal function of the fluctuation and temperature, i.e., $\Phi_F$ depends also on another system-dependent parameter(s). Because of these difficulties, general properties of $\Phi_F$ were not clarified.

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In this paper, we derive general properties of $\Phi_F$, which hold for diverse physical systems. We clarify which properties are common or different between $\Phi_F$ and $\Phi_{eq}$. We also illustrate some of the properties by a molecular dynamics (MD) simulation of a many-body system.

Response function of NESS – Suppose that a strong static field $F$ is applied to the target system (the macroscopic system of interest), and a NESS is realized for a sufficiently long time, i.e., for $[t_{in}, t_{out}]$, where $t_{out} - t_{in}$ is macroscopically long. In such a NESS every macroscopic variable $A$ takes a constant value $\langle A \rangle_F$ in the sense that its expectation value at time $t$ behaves as

$$\langle A \rangle^t_F = \langle A \rangle_F + o((A)_{_tp}).$$

(1)

Here, $\langle A \rangle_{tp}$ denotes a typical value of $A$, and $o((A)_{tp})$ represents a (time-dependent) term which is negligibly small in the sense that $o((A)_{tp})/(A)_{tp} \rightarrow 0$ as $V \rightarrow \infty$, where $V$ denotes the volume of the target system. When $A$ is the energy $U$, for example, $\langle U \rangle_{tp} = O(V)$ and $\langle U \rangle^t_F = \langle U \rangle_F + o(V)$.

Suppose that a weak and time-dependent probe field $f(t)$ is applied, in addition to $F$, to the target system for $t \geq t_0$, where $t_{in} < t_0 \leq t < t_{out}$. We are interested in the response of the NESS to $f(t)$. Specifically, we focus on the response,

$$\Delta A(t) \equiv \langle A \rangle^t_{F+f} - \langle A \rangle_F,$$

(2)

of a macroscopic variable $A$ of the target system. To the linear order in $f$, $\Delta A(t)$ can be expressed as

$$\Delta A(t) = \int_{t_0}^{t} \Phi_F(t - t')f(t')dt',$$

(3)

which we call the linear response relation. This and the causality relation

$$\Phi_F(\tau) = 0 \text{ for } \tau < 0,$$

(4)

define the response function $\Phi_F(\tau)$ of the NESS, as in the case of $\Phi_{eq}(\tau)$.

Microscopic expression of $\Phi_F$ – Equations (3) and (4) do not refer to microscopic physics at all – they are phenomenological equations which are closed in a macroscopic level. We now relate them to microscopic physics by deriving a microscopic expression of $\Phi_F(\tau)$.

Since we are interested in general properties of NESSs, we do not employ perturbation expansion with respect to $F$, which, for large $|F|$ of interest, converges only slowly or does not converge except in limited physical situations. To treat $F$ non-perturbatively, we consider a large system which includes the target system, a driving source that generates $F$, and a heat reservoir(s). We call this large system the total system, and denote its Hamiltonian by $\hat{H}^{tot}$. When the target system is an electrical conductor, for example, the driving source may be a battery, the heat reservoir may be the air, and the total system is the one that includes them all, as shown in Fig. 1. On the other hand, we do not include the source of the probe
Fig. 1. An example of a large system which we call the total system. It includes an electrical conductor, a battery, a heat reservoir, and so on.

field $f$, such as a microwave generator, in the total system. We assume that $f$ gives rise to the interaction term $-\hat{B} f(t)$, where $\hat{B}$ is a macroscopic variable of the target system. Hence, the total system is an isolated system except that it is subject to an external weak field $f$.

Therefore, the density operator of the total system $\hat{\rho}^{\text{tot}}_{F+f}(t)$ evolves as

$$i\hbar \frac{\partial}{\partial t} \hat{\rho}^{\text{tot}}_{F+f}(t) = \left[ \hat{H}^{\text{tot}} - \hat{B} f(t), \hat{\rho}^{\text{tot}}_{F+f}(t) \right]. \quad (5)$$

We denote $\hat{\rho}^{\text{tot}}_{F+f}(t)$ with $f = 0$ by $\hat{\rho}^{\text{tot}}_F(t)$. When $f = 0$, the reduced density operator of the target system is

$$\hat{\rho}_F \equiv \text{Tr}' \left[ \hat{\rho}^{\text{tot}}_F(t) \right], \quad (6)$$

where $\text{Tr}'$ denotes the trace operation over the degrees of freedom other than those of the target system. For a time interval $[t_{\text{in}}, t_{\text{out}}]$, a NESS is realized in the target system (while the driving source such as a battery is not in a steady state), and, according to eq. (1), we can regard $\hat{\rho}_F$ as being independent of $t$ as far as macroscopic variables are concerned. Unlike the equilibrium case, however, an explicit form of $\hat{\rho}_F$ is unknown.

When $f \neq 0$, $\hat{\rho}^{\text{tot}}_F(t)$ is changed into $\hat{\rho}^{\text{tot}}_{F+f}(t)$. We assume that the NESS is stable against small perturbations. That is, after small perturbations are removed the target system returns to the same NESS as that before they were applied. Except for NESSs of some soft matters and glass near a melting point, most NESSs, including those of nonlinear optical materials\textsuperscript{4,5} and electrical conductors, satisfy this assumption.

For such NESSs, we can evaluate $\Phi_F$ by evaluating the solution of eq. (5) using a first-order perturbation expansion with respect to $f$. For an observable of interest, $\hat{A}$, of the target system, its response $\Delta A(t) \equiv \text{Tr}[\hat{\rho}^{\text{tot}}_{F+f}(t)\hat{A}] - \text{Tr}[\hat{\rho}^{\text{tot}}_F(t)\hat{A}] = \text{Tr}[\hat{\rho}^{\text{tot}}_{F+f}(t)\hat{A}] - \text{Tr}[\hat{\rho}_F\hat{A}]$ is evaluated to the linear order in $f$ as

$$\Delta A(t) = \int_{t_0}^{t} \frac{1}{i\hbar} \text{Tr} \left( \frac{\partial}{\partial t'} \hat{\rho}^{\text{tot}}_F(t') \left[ \hat{B}, \hat{A}(t-t') \right] \right) f(t') dt',$$ \quad (7)

where the symbol ‘ $\cdot \cdot$ ’ denotes an operator in the interaction picture, i.e., $\hat{A}(t) \equiv$
\[ e^{i \hat{H}_{\text{tot}} t} \hat{A} e^{-i \hat{H}_{\text{tot}} t}. \]  

From consistency with the macroscopic physics, eq. (3), \( t' \) in \( \hat{\rho}_F^{\text{tot}}(t') \) in eq. (7) must be irrelevant. Hence, we can take \( t' \) to be an arbitrary time (such as \( t_0 \)) in \([t_{\text{in}}, t_{\text{out}}]\), and simply write \( \hat{\rho}_F^{\text{tot}}(t) \) as \( \hat{\rho}_F^{\text{tot}} \). We thus obtain a general formula;

\[ \Phi_{AB}(\tau) = \frac{1}{i \hbar} \text{Tr} \left( \hat{\rho}_F^{\text{tot}} \left[ \hat{B}, \hat{A}(\tau) \right] \right) \text{ for } \tau \geq 0. \]  

(8)

Here, we denote \( \Phi_F(\tau) \) by \( \Phi_{AB}^F(\tau) \) to designate variables \( A \) and \( B \). For classical systems, \([\hat{B}, \hat{A}(\tau)]/i \hbar \) (and similar expressions in the following equations) should be replaced with the corresponding Poisson bracket.

The right-hand side (rhs) of eq. (8) represents some correlation in the NESS. If it could reduce to the symmetrized time correlation,\(^1\) it would be equivalent to fluctuation (in the classical regime, \( k_B T \gg \hbar \omega \)). However, as will be discussed shortly, this is not the case when \( F \neq 0 \). Hence, we do not call eq. (8) a fluctuation-dissipation relation (FDR). We call it the response-correlation relation (RCR). Among similar formulas, eq. (8) has the most convenient form to derive useful properties which will be presented below.

**Fluctuation-dissipation and reciprocal relations** – Before deriving useful properties, we use the RCR to discuss why the FDR and the reciprocal relations (including those for finite frequencies\(^1\)) are violated in NESSs.\(^8,9\)

When \( F = 0 \), \( \hat{\rho}_F^{\text{tot}} \) and \( \Phi_{AB}^{\text{eq}} \) of eq. (8) reduce to the equilibrium state \( \hat{\rho}_{\text{eq}} \) and its response function \( \Phi_{eq}^{AB} \), respectively, and the RCR reduces to the equilibrium one\(^1,2\) (which is not customarily called the RCR, though). When the canonical ensemble, e.g., is employed, both \( \hat{\rho}_{\text{eq}} \propto e^{-\beta \hat{H}_{\text{tot}}} \) and \( e^{i \hat{H}_{\text{tot}} t} \) (which defines \( \dot{A}(\tau) \)) are exponential functions of \( \hat{H}_{\text{tot}} \). Using this fact, we can recast the equilibrium RCR as

\[ \Phi_{eq}^{AB}(\tau) = \frac{1}{k_B T} \langle \dot{\hat{B}}(0); \hat{A}(\tau) \rangle_{\text{eq}}, \]  

(9)

where \( \langle \cdot ; \cdot \rangle_{\text{eq}} \) denotes the canonical correlation.\(^1\) This result is known as the Kubo formula, from which one can derive the reciprocal relations.\(^1,2\) In the classical regime (\( k_B T \gg \hbar \omega \)), the canonical correlation reduces to the symmetrized time correlation,\(^1\) and hence to fluctuation,\(^15\) and one obtains the FDR.\(^1,2\)

When \( F \neq 0 \), in contrast, \( \hat{\rho}_F^{\text{tot}} \) (although its explicit form is unknown) cannot be an exponential function of \( \hat{H}_{\text{tot}} \) only. As a result, the RCR cannot be rewritten into a form similar to eq. (9). That is, the RCR holds both for equilibrium states and for NESSs, whereas it is equivalent to the Kubo formula only for the former. As a result, the FDR and the reciprocal relations are violated in NESSs. The difference between the rhs of eq. (8) and the symmetrized time correlation divided by \( k_B T \) is the violating term.

**Properties derived from the phenomenological equations** – Equations (3) and (4) take the same forms as those for \( \Phi_{eq} \). Therefore, among many properties of \( \Phi_{eq} \), those derivable only from eqs. (3) and (4) (without using the Kubo formula) hold also for \( \Phi_F \). For completeness, we mention such properties, although some of them may be rather obvious.
For stable NESSs, we expect that $|\Phi_F(\tau)|$ is integrable over $(-\infty, +\infty)$.\textsuperscript{16} Hence, the Fourier transform
\begin{equation}
\Xi_F(\omega) \equiv \int_{-\infty}^{\infty} \Phi_F(\tau) e^{i\omega \tau} d\tau = \int_{0}^{\infty} \Phi_F(\tau) e^{i\omega \tau} d\tau
\end{equation}
should be a continuous function of $\omega$. As in the case of the Fourier transform $\Xi_{eq}(\omega)$ of $\Phi_{eq}(\tau)$, we can easily show that $\Xi_F(\omega)$ satisfies the dispersion relations,
\begin{equation}
\text{Re} \Xi_F(\omega) = \int_{-\infty}^{\infty} \frac{P}{\omega' - \omega} \text{Im} \Xi_F(\omega') \frac{d\omega'}{\pi},
\end{equation}
\begin{equation}
\text{Im} \Xi_F(\omega) = -\int_{-\infty}^{\infty} \frac{P}{\omega' - \omega} \text{Re} \Xi_F(\omega') \frac{d\omega'}{\pi},
\end{equation}
and the moment sum rules.\textsuperscript{1} We also see that $\text{Re} \Xi_F(\omega)$ is even, whereas $\text{Im} \Xi_F(\omega)$ is odd.

Properties derived from the RCR – We now present the most important results of this paper. For $F = 0$, many properties were previously derived for $\Phi_{eq}$ from the Kubo formula.\textsuperscript{1,2} As discussed above, some of them (such as the FDR) are violated for $\Phi_F$ when $F \neq 0$. However, the other properties of $\Phi_{eq}$ can actually be derived from the RCR without using the Kubo formula, although they were often derived from the Kubo formula (or similar expressions) in the literature. Such properties hold also for $\Phi_F$ if some quantities are replaced with those of a NESS (see below) because the RCR holds even when $F \neq 0$. We now present them.

Note that although their forms are similar to those for $\Phi_{eq}$,\textsuperscript{1,2} their values are often different from those of $\Phi_{eq}$, as will be illustrated later.

Integration of $\Xi_{AB}^F(\omega)$ yields
\begin{equation}
\int_{-\infty}^{\infty} \Xi_{AB}^F(\omega) \frac{d\omega}{\pi} = \Phi_{AB}^F(+0) = \frac{1}{i\hbar} \text{Tr} \left( \hat{\rho}_{F}^{tot} \left[ \hat{B}, \hat{A} (0) \right] \right).
\end{equation}
Since $\hat{A}(0) (= \hat{A})$ and $\hat{B}$ are operators of the target system,
\begin{equation}
\text{Tr} \left( \hat{\rho}_{F}^{tot} \left[ \hat{B}, \hat{A} (0) \right] \right) = \text{Tr} \left( \hat{\rho}_{F} \left[ \hat{B}, \hat{A} \right] \right) = \left\langle \left[ \hat{B}, \hat{A} \right] \right\rangle_F,
\end{equation}
where $\left\langle \cdot \right\rangle_F \equiv \text{Tr} (\hat{\rho}_F \cdot)$ denotes the expectation value in the NESS. Noting also that $\text{Im} \Xi_{AB}^F(\omega)$ is an odd function, we obtain the following sum rule for $\text{Re} \Xi_{AB}^F$;
\begin{equation}
\int_{-\infty}^{\infty} \text{Re} \Xi_{AB}^F(\omega) \frac{d\omega}{\pi} = \left\langle \frac{1}{i\hbar} \left[ \hat{B}, \hat{\dot{A}} (0) \right] \right\rangle_F.
\end{equation}
Since the rhs is the expectation value of a known operator, it can easily be measured experimentally.\textsuperscript{14}

Moreover, by integrating eq. (10) by parts, multiplying the result with $\omega$, and integrating the resultant equation, we obtain the following sum rule for $\text{Im} \Xi_{AB}^F$;
\begin{equation}
\int_{-\infty}^{\infty} \left\{ \omega \text{Im} \Xi_{AB}^F(\omega) - \left\langle \frac{1}{i\hbar} \left[ \hat{B}, \hat{\dot{A}} \right] \right\rangle_F \right\} \frac{d\omega}{\pi} = \left\langle \frac{1}{i\hbar} \left[ \hat{B}, \hat{A} (0) \right] \right\rangle_F.
\end{equation}
In the second line, we have replaced $\hat{\rho}_F^{\text{tot}}$ with $\hat{\rho}_F$ because $\hat{A}(0) = [\hat{A}, \hat{H}^{\text{tot}}]/i\hbar$ is localized in the target system. (Recall that all physical interactions in $\hat{H}^{\text{tot}}$ should be local interactions.) The expectation values in this sum rule can also be measured experimentally.

From these sum rules we can see the asymptotic behavior of $\Xi_{AB}^{+}(\omega)$. As $|\omega|$ is increased, $\Xi_{AB}^{+}(\omega)$ should decay quickly enough such that the integrals of eqs. (13) and (14) converge. In particular, we find

$$\lim_{\omega \to \infty} \omega \text{Im} \Xi_{AB}^{+}(\omega) = \left\langle \frac{1}{i\hbar} \left[ \hat{B}, \hat{A} \right] \right\rangle_F. \tag{15}$$

As discussed above, the reciprocal relation for $\Phi_{eq}^{AB}(t)$ does not hold for $\Phi_{F}^{AB}(t)$, i.e., for $\Xi_{F}^{AB}(\omega)$ for each $\omega$. However, eq. (13) yields

$$\int_{-\infty}^{\infty} \text{Re} \Xi_{F}^{AB}(\omega)d\omega = - \int_{-\infty}^{\infty} \text{Re} \Xi_{F}^{BA}(\omega)d\omega, \tag{16}$$

i.e., a reciprocal relation holds for the integrated values.

**Implications** – The left-hand side of eq. (13) equals to $\Phi_{F}^{AB}(+0)$ (representing instantaneous response), which however is hardly measurable in real physical systems. In contrast, $\text{Re} \Xi_{F}^{AB}(\omega)$ is measurable in a certain finite range of $\omega$. For higher $\omega$, which is out of such a range, $\text{Re} \Xi_{F}^{AB}(\omega)$ decays quickly, as mentioned above. Hence, one does not necessarily have to measure it for higher $\omega$. Therefore, eq. (13) should be considered as a prediction on $\text{Re} \Xi_{F}^{AB}(\omega)$ in a certain finite range of $\omega$. For similar reasons, eqs. (14)-(16) become important when one wants to get information on $\Xi_{F}^{AB}(\omega)$.

These equations, like the corresponding ones for $\Xi_{eq}^{AB}(\omega)$, are very useful for measuring or theoretically calculating $\Xi_{F}^{AB}(\omega)$. For example, one can check experimental or theoretical results against them. Using them and eqs. (11) and (12), one can also estimate $\Xi_{F}^{AB}(\omega)$ in some range of $\omega$ from existing data of $\Xi_{F}^{AB}(\omega)$ in another range. Moreover, as will be illustrated for a Langevin model later, our results can show that some equality is identical to another equality, which were previously treated as independent equalities.

Furthermore, we can see the following. According to eq. (13), the sum value (integral) of $\text{Re} \Xi_{F}(\omega)$ equals to the expectation value $\langle C \rangle_F$ of the Hermitian operator $\hat{C} \equiv [\hat{B}, \hat{A}]/i\hbar$. Since this is an equal-time commutator, $\hat{C}$ depends neither on the Hamiltonian nor on the state. There is no difference in $\hat{C}$ between free particles and interacting ones or between equilibrium states and NESSs. Only through $\hat{\rho}_F$ the sum value can be affected by these factors.

When $\hat{A}$ and $\hat{B}$ are linear functions of canonical variables, in particular, $\hat{C} \propto \hat{1} \ (\text{identity operator})$ and hence the sum takes the same value for every state. More generally, we can say the same when $\text{Tr}[\hat{\rho}_F^{\text{tot}}(t)\hat{C}]$ is conserved during evolution from an equilibrium state to NESSs of interest.

For example, suppose that the target system is an electrical conductor of length $L$. A static electric field $F$ is applied in the $x$ direction (along the conductor). Let $q^i_x$ and $\hat{p}^i_x$ be the $x$
components of the position and momentum, respectively, of the \( j \)th electron in the conductor. Then, \( \hat{B} = \sum_j e \hat{q}^j_x \), and the electric current averaged over the \( x \) direction may be given by

\[
\hat{I} = \frac{1}{L} \sum_j \frac{e}{m} \hat{p}^j_x,
\]

where \( m \) is electron’s mass. If one is interested in \( \hat{I} \), putting \( \hat{A} = \hat{I} \) yields \( \hat{C} = (e^2 N_e/mL)\hat{I} \), where \( N_e \) is the number of electrons in the conductor. We thus find

\[
\int_{-\infty}^{\infty} \text{Re} \Xi_F^B(\omega) \frac{d\omega}{\pi} = \frac{e^2 N_e}{mL},
\]

which is independent of \( F \). Generally, \( \text{Re} \Xi_F^B(\omega) \) at low \( \omega \) depends strongly on \( F \) for large \( |F| \). At high \( \omega \), on the other hand, it is expected that \( \text{Re} \Xi_F^B(\omega) \) would be insensitive to \( F \) because each particle would not collide with other particles in a short time period \( \sim 1/\omega \). (These facts will be illustrated later.) From these viewpoints, eq. (18) may be counterintuitive, and therefore is useful.

Regarding \( \text{Im} \Xi_F^B \), we can apply eqs. (14) and (15). For example, the latter yields

\[
\lim_{\omega \to \infty} \omega \text{Im} \Xi_F^B(\omega) = \frac{e^2 N_e}{mL},
\]

which is also independent of \( F \).

For more general cases where \( \text{Tr}[\hat{\rho}_F^{\text{tot}}(t)\hat{C}] \) is not conserved during evolution from an equilibrium state to NESSs of interest, the sum value generally depends on \( F \). For example, if one is interested in \( \hat{I}^2 \) (to investigate, e.g., current fluctuation) in the above example, putting \( \hat{A} = \hat{I}^2 \) yields \( \hat{C} = (2e^2 N_e/mL)\hat{I} \). We thus find

\[
\int_{-\infty}^{\infty} \text{Re} \Xi_F^{I^2}(\omega) \frac{d\omega}{\pi} = \lim_{\omega \to \infty} \omega \text{Im} \Xi_F^{I^2}(\omega) = \frac{2e^2 N_e}{mL} \langle I \rangle_F,
\]

which depends strongly on \( F \). This fact demonstrates that although the forms of eqs. (13) and (15) are similar to the corresponding ones for \( \Phi^{1,2}_{\text{eq}} \), their values can be very different.

**Non-Hamiltonian systems** – We have assumed that the total system, such as Fig. 1, is a Hamiltonian system. In studies of NESSs, non-Hamiltonian models, such as stochastic models, are often employed. The general properties of \( \Xi_F^{AB} \) must hold also in such models if the models are physically reasonable ones, because every existing physical system is believed to be a Hamiltonian system if a sufficiently large system (such as Fig. 1) is considered.

For example, a nonlinear Langevin model

\[
\dot{p} = -\frac{\gamma}{m} p - U'(x) + F + f(t) + \xi(t),
\]

where \( p = mx \) and \( U(x) \) is a potential, may be derived from a Hamiltonian model by making projection and by approximating \( \gamma \) and \( \xi(t) \) as a constant and white noise, respectively. If these approximations are physically reasonable, eq. (13) must hold in this Langevin model because it holds in the original Hamiltonian model. We can prove that this is the case for any value of \( F \). Hence, the Langevin model is physically reasonable in view of eq. (13).
the other hand, it is well-known that the Langevin model gets worse as $|F|$ is increased. This illustrates that eq. (13), and the other general properties derived above, are not sufficient but necessary conditions for good nonequilibrium models. In this respect, they are similar to the charge conservation, which is also a universal and necessary condition for good models.

As an illustration of significance of eq. (13) on the Langevin model, we can show using eq. (13) that equality (5) of ref. 10 on dissipation $\langle J \rangle_F$ is identical to a simple relation, $\langle J \rangle_F = (\gamma/m)(\langle p^2 \rangle_F/m - k_B T)$, of refs. 17 [eq. (82)] and 18 [eq. (4.13)].

**Numerical example** — Finally, we illustrate the validity of eqs. (18) and (19) by an MD simulation of a model of a classical two-dimensional electrical conductor. The model includes $N_e$ electrons ($e$), $N_p$ phonons ($p$) and $N_i$ impurities ($i$), where $N_e, N_p, N_i \gg 1$. The $e-e$, $e-p$, $e-i$, $p-p$ and $p-i$ interactions are all present, whereas the static electric field $F$ acts only on electrons. The energy of this many-body system is dissipated through thermal walls (which simulate a heat reservoir) for phonons, and a NESS is realized for each value of $F$. This model is a mechanical model supplemented by the thermal walls for phonons. For NESSs at large $|F|$, we previously found the following: (i) $\langle I \rangle_F$ is nonlinear in $F$, (ii) the long-time tail is strongly modified, and (iii) the FDR is significantly violated.

To compute $\Xi^{IB}_F(\omega)$, we take the probe field $f(t)$ to be monochromatic; $f(t) \propto \sin(\omega t)$. We apply it in the $x$ direction in addition to $F$, and perform an MD simulation, in which we here take $e = m = 1, N_e = N_p = 1500, N_i = 500$, and $L = 750$. By calculating the classical counterpart of $\hat{I}(t)$ of eq. (17) for a sufficiently long time ($\gg 1/\omega$), we obtain $\Xi^{IB}_F(\omega)$. This procedure is repeated for various values of $\omega$ and $F$.

Figure 2 shows $\omega$-dependence of $\text{Re} \, \Xi^{IB}_F(\omega)/\pi$ for three different values of $F$. At low frequencies $\text{Re} \, \Xi^{IB}_F(\omega)$ depends strongly on $F$, implying that the response to $F$ is nonlinear. At high frequencies, the $F$-dependence looks quite weak. However, since the horizontal axis is in the logarithmic scale and $\int \Xi(\omega) d\omega = \int \omega \, \Xi(\omega) d(\ln \omega)$, small differences in $\text{Re} \, \Xi^{IB}_F(\omega)$ at high frequencies contribute significantly to the $\omega$-integral of eq. (18). As a result, the integral over all $\omega$ (i.e., the left-hand side of eq. (18)) becomes independent of $F$ (within possible numerical errors), as shown in the inset of Fig. 2. Moreover, the value of the integral agrees with that predicted by eq. (18).

Furthermore, Fig. 3 shows $\omega$-dependence of $\omega \text{Im} \, \Xi^{IB}_F(\omega)$ for three different values of $F$. We observe that as $\omega$ is increased $\omega \text{Im} \, \Xi^{IB}_F(\omega)$ approaches the same asymptotic value, which agrees with that predicted by eq. (19), for all values of $F$.

We have thus confirmed eqs. (18) and (19), which have been derived from the general results, eqs. (13) and (15), respectively. Conversely, as discussed above, the agreement of our numerical results with the general results indicates the following: (i) this model is physically reasonable, and (ii) our MD simulation well describes NESSs and their responses. That is, validity of the numerical results have been checked against the general results even for large
Summary – We have derived general properties of linear response functions $\Phi_F$ (and their Fourier transform $\Xi_F$) of NESSs, which are driven by a strong field $F$. For completeness, we have presented all the basic properties (including rather obvious ones, such as the dispersion relations) which correspond to those of response functions $\Phi_{eq}$ ($\Xi_{eq}$) of equilibrium states.\textsuperscript{1,2} Specifically, we have derived the response-correlation relation, eq. (8), which however cannot be recast into the form of the Kubo formula when $F \neq 0$. As a result, the FDR and reciprocal relations are violated in NESSs, although the latter holds for the integrated values, eq. (16). In contrast, the dispersion relations, eqs. (11) and (12), and the moment sum rules hold even when $F \neq 0$ because they come from the phenomenological equations, eqs. (3) and (4). Furthermore, the sum rules and asymptotic behaviors, eqs. (13)-(15), hold even when $F \neq 0$ if the expectation values in an equilibrium state, $\text{Tr}(\hat{\rho}_{eq} \cdot \cdot)$, are replaced with those in a NESS, $\text{Tr}(\hat{\rho}_F \cdot \cdot)$. We have illustrated some of these results by an MD simulation of an electrical conductor.

These results are quite general, which apply to diverse physical systems because no assumption has been made except that the NESSs are stable. Further generalization to the case $|F|$. In contrast, a typical conventional method is to check results against the FDR (see ref. 11 and references cited therein), which holds only for small $|F|$. \textsuperscript{11}
Fig. 3. $\omega \Im \Xi_{FB}(\omega)$ for $F = 0$ (circles), 0.06 (squares) and 0.1 (triangles). The dashed line represents the rhs of eq. (19).

of time-dependent and/or spatially-varying $F$ (and/or $f$) is straightforward.

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14) The expectation value of a *known* function of canonical variables, such as the rhs's of eqs. (13)-(15), can be measured experimentally, although the density operator of the NESS is unknown. In contrast, the expectation value of an *unknown* function, which often appeared in the literature, cannot be measured in general.
15) In the quantum regime $k_B T \lesssim \hbar \omega$, the symmetrized time correlation is not necessarily the correct quantum-mechanical expression of time correlation, which means correlation of outcomes of subsequent measurements. See, e.g., Sec. 4.8.1 of K. Koshino and A. Shimizu: Physics Reports **412** (2005) 191.
16) Although the upper limit of $\tau$ is $t_{\text{out}} - t_{\text{in}}$, one can make it arbitrarily large by increasing the size of the source of $F$.
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