Multidimensional measures of response and fluctuations in stochastic dynamical systems

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A class of experiments is proposed which involve multiple measurements combined with multiple perturbations of a nonlinear classical complex system. A family of multipoint $n + m - 1$ dimensional measures $R^{(n,m)}$ that provide complementary information on complex systems is obtained by combining m nonlinear stimuli and n measurements. They represent the combined effect of causal response and noncausal spontaneous fluctuations. The proposed signals can be measured either in the frequency domain or in the time domain. Generalized fluctuation-dissipation relations which hold in the nonlinear regime restrict the number of independent techniques. Two-dimensional correlation plots derived from such measurements can reveal various types of couplings among collective modes.

I. INTRODUCTION

Study of dynamical phenomena in nonlinearly coupled systems is of paramount importance in many branches of physics [1,2]. In many cases, analysis of dynamical behavior is often complicated by the presence of fluctuations caused by interactions with a noisy environment or by inherent stochasticity of the system of interest. Dynamics of such systems can be characterized by calculating or measuring averages of some dynamical quantities over different realizations of these fluctuating systems. Such averaged quantities, in which time enters as a parameter, are invariant objects and thus serve as time-dependent measures of fluctuating dynamical systems. One such measure is produced by n-point correlation functions, obtained by performing n measurements of a variable $A(t)$ at times $\tau_1,...,\tau_n$ and constructing the n-point average $\langle A(\tau_n) \ldots, A(\tau_1) \rangle$. More generally one can measure cross correlations of different quantities A_j at different times, yet to simplify the notation we consider a single quantity A, while the extension is straightforward. In a stationary system, these quantities depend on $n - 1$ time intervals and thus constitute an $n - 1$ dimensional characteristic of the system [3–8].

Another way to investigate the properties of a dynamical system is studying how it responds to controlled external perturbations [9–11]. By subjecting a system to $n - 1$ impulsive perturbations at times $\tau_1, \ldots, \tau_{n-1}$ one can record some property $B(\tau_n)$ of the system as a function of the various time delays. The expectation value of $B(\tau_n)$ can be calculated within the scopes of response theory [11], which brings in another type of $(n - 1)$ dimensional characteristic given by the $(n - 1)$th-order response function [12–18] $(-1)^n \langle \{ \cdots \{ B(\tau_n), A(\tau_{n-1}) \} \ldots, A(\tau_1) \} \rangle$, where $\{ \ldots, \ldots \}$ are the Poisson brackets and A is the physical quantity through which external perturbation interacts with the system, $H'(t) = E(t)A$. In case of perturbations by impulsive electric fields, A

could be the dipole moment. Nonlinear $(n - 1)$th-order optical response functions can be measured directly in multidimensional time domain spectroscopic experiments. The response function corresponding to $n = 2$, i.e., the linear response function, is related to the two-point correlation function via the well-known fluctuation-dissipation theorem [19,20], and thus does not carry additional information on the microscopic dynamics of a system beyond what is already contained in the two-point correlation function. A significant research effort has been made in the past to seek for a relation between the nonlinear response functions and multipoint correlation functions, and to derive a generalized fluctuation-dissipation theorem [21–30]. However, for $n > 2$, no simple relation between the $(n - 1)$th-order nonlinear response function and the n-point correlation function was found, implying that the nonlinear response function provides additional information to the n-point correlation function. Both n-point correlation functions and $(n - 1)$th-order response functions depend on $(n - 1)$ time intervals and thus both can serve as complementary $(n - 1)$ dimensional measures of fluctuating dynamical systems.

In the present paper we show that a broader class of dynamical $(n - 1)$ dimensional measures can be introduced by combining m impulsive, but weak, perturbations with $n - m$ measurements done on the system. Indeed, an n-point correlation function corresponds to n consecutive measurements of a dynamical quantity, while n-point response functions represent $(n - 1)$ perturbations followed by a single measurement of some dynamical quantity. It is interesting to explore a generalized class of experiments that consist of m perturbations and $(n - m)$ measurements.

We define a k-point correlation function in which k measurements on a system at times τ_i are mixed with m perturbations of the system at times τ'_j:

$$\langle A(\tau_k) \ldots, A(\tau_1) \rangle_{k,m}$$
$$= \int d\tau'_m \int d\tau'_{m-1} \ldots \int d\tau'_1 R^{(k,m)}(\tau_k, \ldots, \tau_1, \tau'_m, \ldots, \tau'_1)$$
$$\times E_m(\tau'_m) \cdots E_1(\tau'_1), \tag{1.1}$$

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with $k + m = n$. The time variables τ_j and τ_i' are ordered according to the order in which perturbations and measurements are applied. τ_1 is the first measurement, τ_1' is the first perturbation, etc. For instance, the time ordering for an experiment that consists of one perturbation, one measurement, another perturbation, and a second measurement is $\tau_2 > \tau_2' > \tau_1 > \tau_1'$. Note that the chronologically last time must be a τ, not τ', otherwise $R^{(k,m)}$ vanishes, which is a signature of causality. $R^{(k,m)}$ is the response of a k-point correlation function to m impulsive perturbations. We note, however, that for fixed k and m, one will have $(k + m - 1)!/(k - 1)!m!$ different realizations of time domain experiments with particular time ordering of measurements and perturbations, each having its own $R^{(k,m)}$. In this notation $R^{(1,n-1)}$ is an ordinary response function, while $R^{(n,0)}$ is the ordinary n-point correlation function. Similarly we can write frequency domain analogs of these quantities:

$$
\langle \widetilde{A}(\omega_k) \ldots \widetilde{A}(\omega_1) \rangle_{k,m}
$$
$$
= \frac{1}{(2\pi)^m} \int d\omega_m' \ldots \int d\omega_1' \chi^{(k,m)}(\omega_k, \ldots, \omega_1, \omega_m', \ldots, \omega_1')
$$
$$
\times \widetilde{E}_m(\omega_m') \ldots \widetilde{E}_1(\omega_1'), \tag{1.2}
$$

where $\chi^{(k,m)}$ is a generalized susceptibility. Here the system is subjected to m periodic perturbations and k Fourier components of the response are measured. In the frequency domain, time ordering is irrelevant, and thus a frequency domain experiment that consists of m perturbations and k measurements corresponds to a single quantity $\chi^{(k,m)}$ that combines contributions of all $(k + m - 1)!/(k - 1)!m!$ realizations of time ordered experiments. Time domain experiments thus carry more detailed information since different sequences can be separated. We discuss these details in the following sections.

For weak perturbations $E_j(t) = \varepsilon_j \delta(t - \tau_j')$ or $E_j(t) = \varepsilon_j e^{\iota \omega_j' t}$ one can determine the respective $R^{(k,m)}$ and $\chi^{(k,m)}$, respectively, by differentiation:

$$
R^{(k,m)}(\tau_k, \ldots, \tau_1, \tau_m', \ldots, \tau_1') = \frac{\partial^m}{\partial \varepsilon_1 \ldots \varepsilon_m} \langle A(\tau_k) \ldots A(\tau_1) \rangle_{k,m},
$$

$$
\chi^{(k,m)}(\omega_k, \ldots, \omega_1, \omega_m', \ldots, \omega_1') = \frac{\partial^m}{\partial \varepsilon_1 \ldots \varepsilon_m} \langle \widetilde{A}(\omega_k) \ldots \widetilde{A}(\omega_1) \rangle_{k,m},
$$

which in practical applications may be obtained by finite differences.

In the coming sections we discuss the introduced generalized response functions (GRF) $R^{(k,m)}$ and susceptibilities $\chi^{(k,m)}$ in greater detail and analyze them within the scope of classical mechanics in phase space. We concentrate our discussion on systems initially at thermal equilibrium, although other systems such as systems at steady state will be an interesting future application. The paper is organized as follows. In Sec. II we provide explicit expressions for the general class of n-point time domain quantities $R^{(k,m)}$. In Sec. III we show that for systems at thermal equilibrium there exist $n - 1$ independent quantities $R^{(k,m)}$, with $k + m = n$. In Sec. IV we discuss how $R^{(k,m)}$ can serve as new $(n - 1)$ dimensional characteristics of dynamical systems. Several numerical examples are given in Sec. V. The considered models are linearly coupled Morse oscillators, each coupled to a thermal bath of harmonic oscillators, and nonlinearly

coupled harmonic oscillators each coupled to a thermal harmonic bath. There we also consider an exactly solvable model of uncoupled constant-energy harmonic oscillators to simplify subsequent analysis of multidimensional signals of systems at thermal equilibrium. We discuss frequency domain experiments and derive explicit expressions of generalized susceptibilities $\chi^{(k,m)}$ in Sec. VI. We conclude with discussion in Sec. VII.

II. MULTIPLE PERTURBATIONS AND MEASUREMENTS

To set the stage for our discussion, we first review nonlinear response theory, and introduce notations and models. Response theory evaluates the expectation value of a physical quantity B after n perturbations done on the system initially at equilibrium. Let us consider application of this theory to the case when pulses of light are used as external perturbations. Suppose we have a classical system with the Hamiltonian $H(\mathbf{x}, \mathbf{p})$, which we perturb by an external electric field $E(t)$ consisting of a sequence of n pulses $E(t) = \sum E_j(t)$, interacting with the system via the dipole coupling $H_{\text{int}} = -E(t)V$, where V is the system's dipole moment. Vectors $\mathbf{x}$ and $\mathbf{p}$ denote, respectively, a set of N coordinates and their N conjugate momenta of the N-dimensional system of interest. We assume that the dipole moment $V(t) = V(\mathbf{x}(t), \mathbf{p}(t))$ is a function of the system's coordinates $\mathbf{x}$ and momenta $\mathbf{p}$, and implicitly depends on time through $\mathbf{x}(t)$ and $\mathbf{p}(t)$.

The expectation value of $B \equiv V$ at time t can be evaluated via exact expression [31]:

$$
\langle V(t) \rangle = \left\langle T V(t) \exp\left[-\int_{-\infty}^{t} d\tau E(\tau) V_-(\tau) \right] \right\rangle, \tag{2.1}
$$

where $\langle A \rangle = \text{Tr}[A \rho_{eq}]$ denotes averaging with respect to the initial equilibrium distribution density $\rho_{eq} = \rho_{eq}(\mathbf{x}, \mathbf{p})$, and the tracing operation $\text{Tr}[\ldots] \equiv (1/2\pi\hbar)^N \int d\mathbf{x} d\mathbf{p} \ldots$ is over the $(\mathbf{x}, \mathbf{p})$ phase space. The "-" subscript denotes a special type of differential operator, which represents a Poisson bracket [32]:

$$
V_- A \equiv \{V, A\} = \frac{\partial V}{\partial \mathbf{x}} \frac{\partial A}{\partial \mathbf{p}} - \frac{\partial A}{\partial \mathbf{x}} \frac{\partial V}{\partial \mathbf{p}}. \tag{2.2}
$$

T is a time ordering operation. Although position and momentum canonical coordinates $(\mathbf{x}, \mathbf{p})$ are used in the present analysis, it should be noted, however, that both the trace operation and the Poisson bracket are invariant under the canonical transformation of phase space coordinates $(\mathbf{x}, \mathbf{p}) \rightarrow (\mathbf{q}', \mathbf{p}')$. The latter allows one in some cases to chose a convenient set of canonical coordinates $(\mathbf{q}', \mathbf{p}')$ for these operations (such as action-angle coordinates for systems with periodic dynamics, which we use in Sec. V to analyze one of the models). Different nonlinear contributions of the incoming electric field are obtained by expanding the exponent in the right-hand side of Eq. (2.1) in powers of $E(\tau)$. The nth-order contribution reads

$$
\langle V^{(n)}(\tau_{n+1}) \rangle = \int_{-\infty}^{\tau_{n+1}} d\tau_n \int_{-\infty}^{\tau_n} d\tau_{n-1} \ldots \int_{-\infty}^{\tau_2} d\tau_1
$$
$$
\times E(\tau_n) \ldots E(\tau_1) R^{(n)}(\tau_{n+1}, \ldots, \tau_1), \tag{2.3}
$$

where

$$R^{(n)}(\tau_{n+1}, \ldots, \tau_1)$$
$$= (-1)^n \langle V(\tau_{n+1}) V_-(\tau_n) \ldots V_-(\tau_1) \rangle$$
$$= (-1)^n \int \frac{d\mathbf{x} d\mathbf{p}}{(2\pi\hbar)^N} V(\tau_{n+1}) \{V(\tau_n), \ldots \{V(\tau_1), \rho(\mathbf{x}, \mathbf{p})\} \ldots\}$$
$$(2.4)$$

is the nth-order nonlinear response function.

Alternatively, the expression for the nth-order response function can be obtained nonperturbatively [31] from Eq. (2.1) by subjecting the system to n impulsive delta pulses $E(t) = \sum_{i=1}^n \varepsilon_i \delta(t - \tau_i)$, which reduces Eq. (2.1) to

$$\langle V(\tau_{n+1}) \rangle = \langle V(\tau_n) e^{-\varepsilon_n V_-(\tau_n)} \ldots e^{-\varepsilon_1 V_-(\tau_1)} \rangle \qquad (2.5)$$

and Eq. (2.3) to

$$\langle V^{(n)}(\tau_{n+1}) \rangle = \varepsilon_n \varepsilon_{n-1} \ldots \varepsilon_1 R^{(n)}(\tau_{n+1}, \ldots, \tau_1). \quad (2.6)$$

Equation (2.5) serves as a generating function for the response function. From Eqs. (2.5) and (2.6) one can easily find that

$$R^{(n)}(\tau_{n+1}, \ldots, \tau_1) \equiv \frac{\partial^n}{\partial \varepsilon_n \ldots \partial \varepsilon_1} \langle V(\tau_{n+1}) \rangle |_{\varepsilon_n = \varepsilon_1 = 0}$$
$$= (-1)^n \langle V(\tau_{n+1}) V_-(\tau_n) \ldots V_-(\tau_1) \rangle. \quad (2.7)$$

While Eqs. (2.4) and (2.7) are equivalent, Eq. (2.5), that is used in the derivation of Eq. (2.7), has a clearer physical meaning; i.e., it means that the response function can be calculated by imposing n weak impulsive perturbations on the system and performing a measurement of V at some time τ_{n+1} after the last perturbation. The mathematical operation that corresponds to a perturbation is V_- defined in Eq. (2.2). Although the exact form of the perturbation operator is $\exp[-\varepsilon_i V_-]$, it always appears in the expression of the response function Eq. (2.7) together with the derivative $\partial/\partial\varepsilon_i \exp[-\varepsilon_i V_-]|_{\varepsilon_i=0} = -V_-$; we therefore refer to both operators $\exp[-\varepsilon_i V_-]$ and V_- as perturbation operators; i.e., the V_- operator represents infinitely small perturbation.

To simplify further discussions, we introduce a subscript notation for the ordinary product:

$$V_+ A \equiv V A. \qquad (2.8)$$

In this notation, the n-point correlation function reads

$$\langle V_+(\tau_n) V_+(\tau_{n-1}) \ldots V_+(\tau_1) \rangle, \qquad (2.9)$$

and the $(n-1)$th-order response function becomes

$$(-1)^{n-1} \langle V_+(\tau_n) V_-(\tau_{n-1}) \ldots V_-(\tau_1) \rangle. \qquad (2.10)$$

The structural similarity of the expressions in Eqs. (2.9) and (2.10) suggests that we can define a broader class of correlation functions, in which m perturbations V_- are mixed with $k = n - m$ measurements V_+:

$$R_{+,\pm\ldots\pm}^{(k,m)} \equiv (-1)^m \langle V_+(\tau_n) V_\pm(\tau_{n-1}) \ldots V_\pm(\tau_1) \rangle. \quad (2.11)$$

The only requirement is that chronologically the last operation should be V_+, and not V_-, since the trace $\text{Tr}[V_- B] = \text{Tr}[\{V, B\}]$ vanishes, which is also a signature of causality; i.e., there is no reason to make a perturbation if it will not be followed by a measurement. Keeping track of the possible time ordering of $V_+(\tau_i)$ and $V_-(\tau_j')$, the total number of the

generalized response functions $R_{+,\pm\ldots\pm}^{(k,m)}$ in Eq. (2.11) will be 2^{n-1}. Yet, not all of them are independent for systems initially at thermal equilibrium. This will be shown in the following section.

III. SYSTEMS AT THERMAL EQUILIBRIUM

A distinctive feature of the Boltzmann distribution density, $\rho_{\text{eq}}(\mathbf{x}, \mathbf{p}) = (1/Z) \exp[-\beta H(\mathbf{x}, \mathbf{p})]$, is its exponential form with linear dependence on the system's Hamiltonian in its exponent. This allows us to simplify Poisson bracket expressions containing the Boltzmann distribution function using the chain rule:

$$\{V(t), \rho_{\text{eq}}\} = -\beta \{V(t), H\} \rho_{\text{eq}} = -\beta \frac{dV(t)}{dt} \rho_{\text{eq}}. \quad (3.1)$$

For the linear response, this results in the well-known fluctuation-dissipation relation between the two-point time correlation function $R_{++} = \langle V_+(\tau_2) V_+(\tau_1) \rangle$ and the first-order, linear response function $R_{+-} = -\langle V_+(\tau_2) V_-(\tau_1) \rangle$:

$$-\langle V_+(\tau_2) V_-(\tau_1) \rangle = -\text{Tr}[V(\tau_2)\{V(\tau_1), \rho_{\text{eq}}\}]$$
$$= \beta \text{Tr}[V(\tau_2) \dot{V}(\tau_1) \rho_{\text{eq}}]$$
$$= \beta \frac{d}{d\tau_1} \langle V_+(\tau_2) V_+(\tau_1) \rangle. \quad (3.2)$$

Making use of Eq. (3.1) we can write down explicit expressions of several lowest-order generalized response functions from Eq. (2.11). For third-order quantities $n = 3$ we have

$$R_{+++}(\tau_3, \tau_2, \tau_1) = \langle V(\tau_3) V(\tau_2) V(\tau_1) \rangle,$$
$$R_{++-}(\tau_3, \tau_2, \tau_1) = -\langle V(\tau_3) V(\tau_2) V_-(\tau_1) \rangle$$
$$= \beta \frac{d}{d\tau_1} \langle V(\tau_3) V(\tau_2) V(\tau_1) \rangle,$$
$$R_{+-+}(\tau_3, \tau_2, \tau_1) = -\langle V(\tau_3) V_-(\tau_2) V(\tau_1) \rangle$$
$$= -\langle V(\tau_3) \{V(\tau_2), V(\tau_1)\} \rangle$$
$$+ \beta \frac{d}{d\tau_2} \langle V(\tau_3) V(\tau_2) V(\tau_1) \rangle,$$
$$R_{+--}(\tau_3, \tau_2, \tau_1) = \langle V(\tau_3) V_-(\tau_2) V_-(\tau_1) \rangle$$
$$= -\beta \frac{d}{d\tau_1} \langle V(\tau_3) \{V(\tau_2), V(\tau_1)\} \rangle$$
$$+ \beta^2 \frac{d^2}{d\tau_1 d\tau_2} \langle V(\tau_3) V(\tau_2) V(\tau_1) \rangle. \quad (3.3)$$

From Eq. (3.3) one can see that the generalized response functions are actually related to each other through

$$R_{++-}(\tau_3, \tau_2, \tau_1) = \beta \frac{d}{d\tau_1} R_{+++}(\tau_3, \tau_2, \tau_1),$$
$$R_{+--}(\tau_3, \tau_2, \tau_1) = \beta \frac{d}{d\tau_1} R_{+-+}(\tau_3, \tau_2, \tau_1). \quad (3.4)$$

This implies that once $R_{+++}(\tau_3, \tau_2, \tau_1)$ and $R_{+-+}(\tau_3, \tau_2, \tau_1)$ are determined, the other two correlation functions $R_{++-}(\tau_3, \tau_2, \tau_1)$ and $R_{+--}(\tau_3, \tau_2, \tau_1)$ can be found by differentiation. Clearly, from Eq. (3.3) it follows that there is no way to express R_{+-+} in terms of derivatives of R_{+++}. It is well known from the classical theory of nonlinear response functions that

it is not possible to express a nonlinear classical response function, R_{+--}, in terms of classical correlation functions R_{+++} [33]. Therefore a pair of quantities, in which one is either R_{+++} or R_{++-} and the other one is either R_{+-+} or R_{+--}, is sufficient to calculate all four three-point generalized measures in Eq. (2.11), and thus contains all the (independent) information that can be extracted from experiments that consist of m perturbations and $3 - m$ measurements.

Equations (3.4) may also be viewed as generalized fluctuation dissipation relations. We can also write them in the frequency domain by introducing the quantity

$$S_{+,\pm,\ldots,\pm}(\Omega_{n-1},\ldots,\Omega_1)$$
$$= \int_0^\infty dt_n \ldots \int_0^\infty dt_1 R_{+,\pm,\ldots,\pm}(t_n,\ldots,t_1)$$
$$\times \exp(i\Omega_1 t_1 + \cdots \iota\Omega_n t_n), \quad (3.5)$$

which is a Fourier transform of the time-ordered generalized response function $R_{+,\pm,\ldots,\pm}(t_n,\ldots,t_1) = (-1)^m \langle V_+(t_n + \cdots + t_1) \ldots V_\pm(t_2 + t_1) V_\pm(t_1) \rangle$ over the time intervals $t_j \equiv \tau_j - \tau_{j-1}$ between the consecutive perturbations or measurements. We then have

$$S_{+-}(\Omega_1) = i\beta\Omega_1 S_{++}(\Omega_1),$$
$$S_{++-}(\Omega_2,\Omega_1) = i\beta\Omega_1 S_{+++}(\Omega_2,\Omega_1), \quad (3.6)$$
$$S_{+--}(\Omega_2,\Omega_1) = i\beta\Omega_1 S_{+-+}(\Omega_2,\Omega_1).$$

Note that these Fourier transforms of time domain signals are different from the frequency domain signals given by susceptibilities. The connection will be made in Sec. VI.

A. The number of independent GRF

In the previous section we have seen that only one of the two GRF $R_{+,\pm}$ and only two of the four measures $R_{+,\pm,\pm}$ are independent. Before we make a generalization to the case of arbitrary n, we consider one more case, $n = 4$, for clearer illustration. There are $2^{4-1} = 8$ four-point measures $R_{+,\pm,\pm,\pm}$; these are listed in the Appendix. One can see that all these expressions depend only on five distinct GRF:

$$\langle V(\tau_4)V(\tau_3)V(\tau_2)V(\tau_1)\rangle, \quad \langle V(\tau_4)V(\tau_3)\{V(\tau_2),V(\tau_1)\}\rangle,$$
$$\langle V(\tau_4)V(\tau_2)\{V(\tau_3),V(\tau_1)\}\rangle, \quad \langle V(\tau_4)V(\tau_1)\{V(\tau_3),V(\tau_2)\}\rangle,$$
$$\langle V(\tau_4)\{V(\tau_3),\{V(\tau_2),V(\tau_1)\}\}\rangle. \quad (3.7)$$

The second, the third, and the fourth correlation functions in Eq. (3.7) differ only by time ordering and therefore their Fourier transforms can be obtained from another one by permutation of the corresponding frequencies, thus carrying essentially the same information (see Appendix). In Sec. VI, where we discuss frequency domain measurements, time ordering becomes irrelevant, and a measurement of an appropriate nonlinear susceptibility is sufficient to construct the second, the third, and the fourth terms in Eq. (3.7) with no additional information needed beyond the susceptibility. Therefore, the number of independent correlation functions in Eq. (3.7) reduces to only 3:

$$\langle V(\tau_4)V(\tau_3)V(\tau_2)V(\tau_1)\rangle, \quad \langle V(\tau_4)V(\tau_3)\{V(\tau_2),V(\tau_1)\}\rangle,$$
$$\langle V(\tau_4)\{V(\tau_3),\{V(\tau_2),V(\tau_1)\}\}\rangle. \quad (3.8)$$

We can now write down the Poisson bracket operation explicitly. The Poisson bracket of canonical variables reads

$$\{V(\tau_1),V(\tau_2)\} = \epsilon_{ij}M_{jk}(\tau_2,\tau_1)V_i'(\tau_2)V_k'(\tau_1), \quad (3.9)$$

where $V_j' \equiv \frac{\partial V}{\partial x_j}, x_1 \equiv \mathbf{p}, x_2 \equiv \mathbf{x}, \epsilon_{ii} \equiv 0,$ and $\epsilon_{12} = -\epsilon_{21} = 1$. The quantity $M_{jk}(\tau_2,\tau_1) = \frac{\partial x_k(\tau_1)}{\partial x_j(\tau_2)}$ is the *stability matrix* defined as the derivative of a small deviation $\delta x_k(\tau_1)$ at time τ_1 with respect to a small deviation $\delta x_k(\tau_2)$ at time τ_2. A consecutive action of several Poisson brackets can lead to higher-order stability matrices $M_{jk}^{(n)}(\tau_2,\tau_1) = \frac{\partial^n x_k(\tau_1)}{\partial x_{j_1}(\tau_2)\ldots\partial x_{j_n}(\tau_2)}$. The stability matrix is a *new* dynamical quantity that is not contained in correlation functions and describes the classical coherence between the two (or more, for higher-order stability matrices) nearby classical trajectories. Therefore, each Poisson bracket operation in Eq. (3.8) introduces a new type of dynamical variables, the stability matrix, into the expressions of multipoint correlation functions. The expressions in Eq. (3.8) can be rewritten in terms of stability matrices:

$$\langle V(\tau_4)V(\tau_3)V(\tau_2)V(\tau_1)\rangle,$$
$$\langle \epsilon_{ij}V(\tau_4)V(\tau_3)M_{jk}(\tau_2,\tau_1)V_k'(\tau_2)V_i'(\tau_1)\rangle,$$
$$[\langle \epsilon_{ij}\epsilon_{ab}M_{bc}(\tau_4,\tau_3)V_c'(\tau_4)V_a'(\tau_3)M_{ij}(\tau_2,\tau_1)V_k'(\tau_2)V_i'(\tau_1)\rangle$$
$$-\beta\langle \epsilon_{ij}V(\tau_4)\dot{V}(\tau_3)M_{jk}(\tau_2,\tau_1)V_k'(\tau_2)V_i'(\tau_1)\rangle], \quad (3.10)$$

where summation over repeating indexes is implied. These expressions contain zero, one, and two stability matrices, respectively. It is the number of correlation functions with a different number of stability matrices and their combinations that determine the number of independent n-point quantities $R_{+,\pm,\ldots,\pm}$ for systems at thermal equilibrium. By listing all possible combinations of stability matrices, i.e., all possible combinations of Poisson brackets in n-point correlation functions $R_{+,\pm,\ldots,\pm}$, we can count the number of independent n-point quantities $R_{+,\pm,\ldots,\pm}$ [one, however, should check that some combinations may be the same due to the rule $\text{Tr}(A\{B,C\}) = \text{Tr}(\{A,B\}C)$]. The results are given in Table I. We note that the number of independent quantities is $n - 1$. Each of these independent quantities potentially carries some extra information on the underlying dynamics. In the following section, we discuss what new information they can provide.

IV. THE ROLE OF PERTURBATIONS IN CORRELATION FUNCTIONS

In the previous section we have seen that an impulsive perturbation introduces new type of dynamical variables, stability derivatives, which thus should carry additional information on the system's dynamics. A generalized response function $R_{+,\pm,\ldots,\pm}^{(n)}$ consists of an ensemble average of a product of ordinary dynamical quantities $V(\tau)$ or $V_j'(\tau)$ and stability derivatives $\partial x_k(\tau)/\partial x_j(0)$. The major difference between an ordinary dynamical stochastic quantity $x(\tau)$ and its stability derivative $\partial x(\tau)/\partial x(0)$ is that the former has some random initial phase $\varphi_0 = \text{random}(0,2\pi)$, while the latter has zero initial phase $\varphi_0 = 0$. Since any correlation function contains averaging over the total random phase, it is this difference that can provide new surviving components of correlation functions beyond what is contained in ordinary correlation

TABLE I. The number of independent n-point measures $R_{+,\pm,\ldots,\pm}^{(n)}$ in systems with Boltzmann distribution.

n	The number of all possible $R_{+,\pm,\ldots,\pm}^{(n)}$	The number of independent correlation functions
2	2	$\langle VV \rangle \Rightarrow 1$
3	4	$\left.\begin{array}{l}\langle VVV \rangle \\ \langle V\{V,V\}\rangle\end{array}\right\} \Rightarrow 2$
4	8	$\left.\begin{array}{l}\langle VVVV \rangle \\ \langle VV\{V,V\}\rangle \\ \langle V\{V,\{V,V\}\}\rangle\end{array}\right\} \Rightarrow 3$
5	16	$\left.\begin{array}{l}\langle VVVVV \rangle \\ \langle VVV\{V,V\}\rangle \\ \langle V\{V,V\}\{V,V\}\rangle \\ \langle\{V,V\}\{V,\{V,V\}\}\rangle\end{array}\right\} \Rightarrow 4$
6	32	$\left.\begin{array}{l}\langle VVVVVV \rangle \\ \langle VVVV\{V,V\}\rangle \\ \langle VV\{V,V\}\{V,V\}\rangle \\ \langle V\{V,V\}\{V,\{V,V\}\}\rangle \\ \langle\{V,\{V,V\}\}\{V,\{V,V\}\}\rangle\end{array}\right\} \Rightarrow 5$

functions. We can consider the following simple example. Suppose that our observable of interest, $V(t)$, is a harmonic mode $x(t)$ with frequency ω and some random initial phase φ_0. One wants to compare a regular product $V_+(t)V(0) \equiv x(t)x(0)$ with a Poisson bracket expression $V_-(t)V(0) \equiv \{x(t),x(0)\}$ for this system. Expressing $x(t) = A\cos(\omega t + \varphi_0)$, the product $x(t)x(0)$ and the Poisson bracket $\{x(t),x(0)\} = \frac{\partial x(t)}{\partial x(0)}\frac{\partial x(0)}{\partial p(0)} - \frac{\partial x(t)}{\partial p(0)}\frac{\partial x(0)}{\partial x(0)}$ then read

$$x(t)x(0) = \frac{A^2}{2}\cos(\omega t) + \frac{A^2}{2}\cos(\omega t + 2\phi_0),$$
$$\{x(t),x(0)\} = -\frac{1}{\omega}\sin(\omega t). \tag{4.1}$$

One can see that while the former expression contains terms with random phases $\exp(\pm\iota 2\varphi_0)$, the latter expression, the Poisson bracket, contains no such terms. This implies that the overall phase of the product $V(t_n)V(t_{n-1})\cdots V(t)V(0)$ can be different from the overall phase of $V(t_n)V(t_{n-1})\cdots\{V(t),V(0)\}$ and thus can result in a different number of nonvanishing terms in various generalized response functions $R_{+,\pm,\ldots,\pm}^{(n)} \equiv \langle V_+(\tau_n)\cdots V_-(\tau_1)\rangle$. More examples for other types of dynamical variable $V(t)$ for a two-oscillator system are given in Table II.

In weakly anharmonic systems the Poisson bracket operation removes some of the stochastic material phases, and thus determines which terms in n-point correlation functions will survive, i.e., what peaks will be present in multidimensional spectra and what microscopic couplings will contribute to their intensities. The latter can provide extra information on the microscopic dynamics and thus the generalized response functions $R_{+,\pm,\ldots,\pm}^{(n)}$ can serve as complementary $(n-1)$ dimensional measures of stochastic dynamical systems. As shown in the previous section, there should be $n-1$ such independent measures.

The same phase-cancelation approach is used in bispectral and bicoherence stochastic analysis [34,35] (see also Sec. VI). In bispectral analysis, one calculates expectation values of three-point products $E[\widetilde{V}(\omega_1)\widetilde{V}(\omega_2)\widetilde{V}^*(\omega_1+\omega_2)]$ of Fourier transforms of a fluctuating signal $V(t)$, in which only the terms with zero overall random phase survive, thus providing information on wave interactions in quadratically nonlinear systems. This approach is equivalent to constructing a three-point measure $R_{+++}^{(3)}$. The additional insight provided by the present approach is that, by subjecting a system to perturbations, i.e., introducing Poisson brackets, we vary the overall random phase, and thus change or filter the resulting multidimensional spectra. Several numerical examples are given in Sec. V.

V. NUMERICAL EXAMPLES

In this section we calculate different three-point measures $R_{+,\pm,\pm}^{(3)}(t_2+t_1,t_1,0)$ and their Fourier transforms $S_{+,\pm,\pm}^{(3)}(\Omega_2,\Omega_1)$ for several model systems: linearly coupled anharmonic oscillators and nonlinearly coupled harmonic oscillators, which are in equilibrium with thermal bath.

Before we proceed to modeling systems at thermal equilibrium, we provide analytical results for exactly solvable

TABLE II. Overall phases of two-point quantities for a two-oscillator system.

$V(t)$	Phases of terms in $V(t)V(0)$	Phases of terms in $\{V(t),V(0)\}$
Q_1^2	$0, \pm 2\phi_1, \pm 4\phi_1$	$0, \pm 2\phi_1$
$Q_1 + Q_2$	$0, \pm 2\phi_1, \pm 2\phi_2, \pm(\phi_1-\phi_2), \pm(\phi_1+\phi_2)$	0
$Q_1 Q_2$	$\pm 2\phi_1, \pm 2\phi_2, \pm 2(\phi_1-\phi_2), \pm 2(\phi_1+\phi_2)$	$0, \pm 2\phi_1, \pm 2\phi_2$

microcanonical system, which will simplify further analysis of spectra of systems at thermal equilibrium. Expressions for GRF in a microcanonical ensemble can be easily obtained by replacing β in Eq. (3.3) with $-\partial/\partial E$ [36]:

$$R_{+++}(\tau_3, \tau_2, \tau_1) = \langle V(\tau_3)V(\tau_2)V(\tau_1) \rangle,$$

$$R_{++-}(\tau_3, \tau_2, \tau_1) = -\frac{\partial}{\partial E} \frac{d}{d\tau_1} R_{+++}(\tau_3, \tau_2, \tau_1),$$

$$R_{+-+}(\tau_3, \tau_2, \tau_1) = -\langle V(\tau_3)\{V(\tau_2), V(\tau_1)\} \rangle$$
$$- \frac{\partial}{\partial E} \frac{d}{d\tau_2} R_{+++}(\tau_3, \tau_2, \tau_1),$$

$$R_{+--}(\tau_3, \tau_2, \tau_1) = -\frac{\partial}{\partial E} \frac{d}{d\tau_1} R_{+-+}(\tau_3, \tau_2, \tau_1). \quad (5.1)$$

We consider a model of uncoupled harmonic oscillators $Q_1(t)$, $Q_2(t)$ with frequencies $\omega_1 = 10$, $\omega_2 = 4.2$, respectively, and an observable $V(t)$ that is nonlinear in Q_1 and Q_2 given by

$$V = f_1 Q_1 + f_2 Q_2 + f_{11} Q_1^2 + f_{22} Q_2^2$$
$$+ f_{12} Q_1 Q_2 + f_{122} Q_1 Q_2^2. \quad (5.2)$$

Analysis of oscillating microcanonical systems can be efficiently performed in a special type of canonical coordinates: action-angle variables (J, φ). Using these variables, harmonic oscillations $x(t)$ are expressed as $x(t) = \sqrt{2J/\omega} \cos(\omega t + \varphi)$. In Eq. (5.2), a reduced form $Q_j(t) = \sqrt{J_j} \cos(\omega_j t + \varphi_j)$ is implied, with constant factors $\sqrt{2/\omega_j}$ absorbed into coefficients f. In our numerical calculations, these coefficients were taken as $f_1 = f_2 = f_{12} = f_{122} = f_{11} = f_{22} = 1$, and the energies of harmonic oscillators were $J_1 = J_2 = 1$. The averaging in classical correlation functions in Eq. (5.1) is therefore reduced to the averaging over the initial phases φ_1 and φ_2. In Fig. 1 we present two-dimensional (2D) signals $S_{+++}^{(3)}(\Omega_2, \Omega_1)$, $S_{+--}^{(3)}(\Omega_2, \Omega_1)$, $S_{+-+}^{(3)}(\Omega_2, \Omega_1)$, and $S_{+--}^{(3)}(\Omega_2, \Omega_1)$, defined in Eq. (3.5), and in Table III provide a list of positions and intensities of diagonal and cross peaks of $S_{+±±}^{(3)}$, which can be obtained analytically for this model. First, we notice that the information contained in the spectrum $S_{+++}^{(3)}(\Omega_2, \Omega_1)$ is not complete, and it is not possible to determine the coefficients of Eq. (5.2) from this spectrum alone (since all the intensities depend on the products of coefficients). On the other hand, the 2D signal $S_{+-+}^{(3)}(\Omega_2, \Omega_1)$, for instance, allows us to determine the absolute values of the coefficients f_{11} and f_{22} at frequencies $(2\omega_1, 2\omega_1)$ and $(2\omega_2, 2\omega_2)$, respectively, and thus obtain all remaining coefficients f_i, f_{ij}, f_{ijj}. However, if the quadratic anharmonicities f_{11} and f_{22} are small, the peaks at $(2\omega_1, 2\omega_1)$ and $(2\omega_2, 2\omega_2)$ may not appear in $S_{+++}^{(3)}(\Omega_2, \Omega_1)$ or $S_{+-+}^{(3)}(\Omega_2, \Omega_1)$, since their intensities are proportional to the third power of f_{ii}. One can then refer to the spectrum $S_{+--}^{(3)}(\Omega_2, \Omega_1)$, in which the peak $(2\omega_1, 2\omega_1)$ is four times more intensive than in $S_{+-+}^{(3)}(\Omega_2, \Omega_1)$. Otherwise, one can compare $S_{+-+}^{(3)}(\Omega_2, \Omega_1)$ with, for instance, $S_{+++}^{(3)}(\Omega_2, \Omega_1)$; both should have an intensive peak at the frequency (ω_2, ω_2). Either the ratio of the intensities of these diagonal peaks or the ratio of intensities of the cross peaks at (ω_2, ω_2) and $(\omega_2, 2\omega_2)$ in $S_{+++}^{(3)}(\Omega_2, \Omega_1)$ can give us the ratio f_{11}/f_{22} and thus provide a way to express all the remaining coefficients f_i, f_{ij}, f_{ijj} in terms of f_{11}. Once any of these coefficients are determined from some other experiment, from a one-dimensional signal,
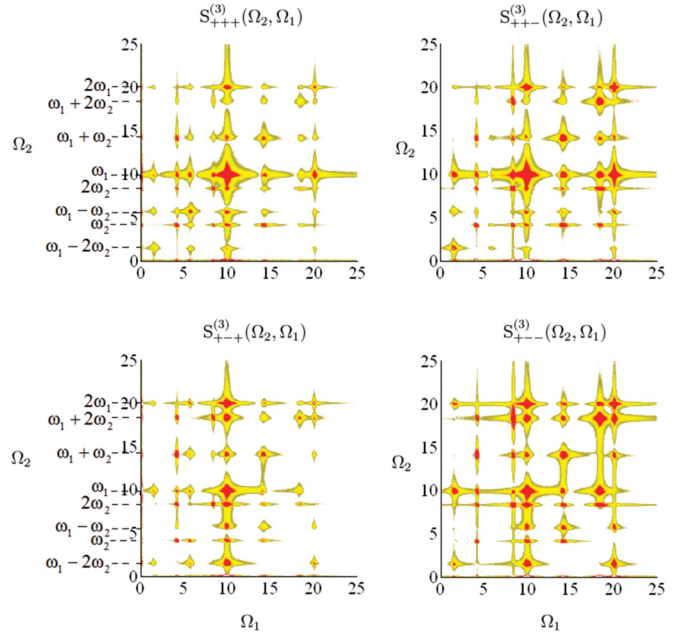

FIG. 1. (Color online) Absolute values of Fourier transforms given by Eq. (3.5) of the correlation functions $R_{+++}^{(3)}(t_2 + t_1, t_1, 0)$, $R_{+-+}^{(3)}(t_2 + t_1, t_1, 0)$, $R_{++-}^{(3)}(t_2 + t_1, t_1, 0)$, and $R_{+--}^{(3)}(t_2 + t_1, t_1, 0)$ for the model of two uncoupled constant-energy harmonic oscillators with $V(t)$ from Eq. (5.2). The two normal mode frequencies are $\omega_1 = 10$ and $\omega_2 = 4.2$.

for instance, the rest are found automatically. We therefore conclude that the complementary spectra $S_{+±±}^{(3)}(\Omega_2, \Omega_1)$ can simplify the analysis of a dynamical system.

We now turn to a system at thermal equilibrium: two coupled dissipative Morse oscillators $U_i(x_i) = D_i(1 - \exp(-\alpha_i x_i))^2$ with a bilinear coupling $U_{12}x_1x_2$. Each oscillator is individually coupled to the Langevin thermostat at temperature T. The friction coefficient is small such that the vibrational dynamics remained underdamped. The fundamental frequencies of Morse oscillators were $\omega_{01} \approx 2\omega_{02}$ so that a Fermi 1 : 2 resonance is possible. The dynamics of coupled oscillators as well as time evolution of stability matrix elements were obtained by solving stochastic Langevin equations. The details of numerical simulations will be published elsewhere. We calculated numerically the three-point quantities $R_{+±±}^{(3)}(t_2 + t_1, t_1, 0)$ with $V = x_1$. Physically $V = x_1$ can correspond to a single optically active mode. The Fourier-transformed spectra $S_{+±±}^{(3)}(\Omega_2, \Omega_1)$ are shown in Fig. 2. By comparing these spectra with Table III, one can deduce on the underlying microscopic dynamics. First, the cross peak at $(\omega_1 + \omega_2, \omega_2)$ in $S_{+--}^{(3)}(\Omega_2, \Omega_1)$ indicates the Fermi-resonance, f_{122}. The latter conclusion could not be made for the same peak of the $S_{+++}^{(3)}(\Omega_2, \Omega_1)$ 2D spectrum, since multiple couplings contribute to its intensity (see Table III). Second, the $(\omega_2, \omega_1 + \omega_2)$ peak in $S_{+-+}^{(3)}(\Omega_2, \Omega_1)$ indicates a bilinear mode-mode coupling f_{12}. Third, as one would expect from the linear dependence of V on x_1, no peaks can be resolved at $(2\omega_1, 2\omega_1)$ and $(2\omega_2, 2\omega_2)$, implying weak quadratic anharmonicities f_{11}, f_{22}. Yet, the peaks at (ω_2, ω_2) in both 2D spectra, or (ω_2, ω_2) and $(\omega_2, 2\omega_2)$ in the $S_{+++}^{(3)}(\Omega_2, \Omega_1)$ spectrum, allow us to estimate the ratio of f_{11}/f_{22}. We also note

TABLE III. 2D spectra for the model in Eq. (5.2) for two modes Q_1 and Q_2 with microcanonical distribution $J_1 = J_2 = 1$. The asterisk in 0^* means that the term is proportional to $J_1 - J_2$.

| (Ω_1,Ω_2)	$|S^{(3)}_{+++}(\Omega_2,\Omega_1)|$	$|S^{(3)}_{+-+}(\Omega_2,\Omega_1)|$	$|S^{(3)}_{++-}(\Omega_2,\Omega_1)|$	$|S^{(3)}_{+--}(\Omega_2,\Omega_1)|$
(ω_1,ω_1)	$\frac{1}{8}(f_{11}+f_{22})(f_1+\frac{f_{122}}{2})^2$	$\frac{1}{8}f_{11}(f_1+\frac{f_{122}}{2})^2$	$\frac{1}{4}(f_{11}+\frac{f_{22}}{2})(f_1+\frac{f_{122}}{2})^2$	$\frac{1}{8}f_{11}(f_1+\frac{f_{122}}{2})^2$
$(\omega_1,2\omega_1)$	$\frac{1}{16}f_{11}(f_1+\frac{1}{2}f_{122})^2$	$\frac{1}{8}f_{11}(f_1+\frac{f_{122}}{2})^2$	$\frac{1}{8}f_{11}(f_1+\frac{f_{122}}{2})^2$	$\frac{1}{8}f_{11}(f_1+\frac{f_{122}}{2})^2$
(ω_1,ω_2)	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$
$(\omega_1,\omega_1-\omega_2)$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$
$(\omega_1,\omega_1+\omega_2)$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$
$(\omega_1,2\omega_2)$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$
$(\omega_1,\omega_1-2\omega_2)$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$
$(\omega_1,\omega_1+2\omega_2)$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$
(ω_2,ω_1)	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_1f_{12}f_2$	$\frac{1}{16}f_{12}f_2(f_1+f_{122})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$
$(\omega_2,\omega_1+\omega_2)$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{16}f_{12}f_2(f_1+f_{122})$	$\frac{1}{16}f_{12}f_2(f_1+f_{122})$	$\frac{1}{16}f_{12}f_2(f_1+\frac{3f_{122}}{2})$
(ω_2,ω_2)	$\frac{1}{8}f_2^2(f_{11}+f_{22})$	$\frac{1}{8}f_2^2f_{22}$	$\frac{1}{8}f_2^2(f_{11}+2f_{22})$	$\frac{1}{8}f_2^2f_{22}$
$(\omega_2,2\omega_2)$	$\frac{1}{16}f_2^2f_{22}$	$\frac{1}{8}f_2^2f_{22}$	$\frac{1}{8}f_2^2f_{22}$	$\frac{1}{8}f_2^2f_{22}$
$(\omega_2,\omega_1-\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	$\frac{1}{64}f_{12}f_{122}f_2$	$\frac{1}{32}f_{12}f_{122}f_2$	0^*
$(\omega_2,\omega_1+2\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	$\frac{3}{64}f_{12}f_{122}f_2$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{1}{16}f_{12}f_{122}f_2$
$(2\omega_1,\omega_1)$	$\frac{1}{16}f_{11}(f_1+\frac{f_{122}}{2})^2$	0	$\frac{1}{4}f_{11}(f_1+\frac{f_{122}}{2})^2$	0
$(2\omega_1,2\omega_1)$	$\frac{1}{32}f_{11}^2(f_{11}+f_{22})$	$\frac{1}{16}f_{11}^3$	$\frac{1}{8}f_{11}^2(f_{22}+\frac{3}{2}f_{11})$	$\frac{1}{4}f_{11}^3$
$(2\omega_1,\omega_1-\omega_2)$	$\frac{1}{64}f_{11}f_{12}^2$	$\frac{1}{64}f_{11}f_{12}^2$	$\frac{1}{16}f_{11}f_{12}^2$	$\frac{1}{16}f_{11}f_{12}^2$
$(2\omega_1,\omega_1+\omega_2)$	$\frac{1}{64}f_{11}f_{12}^2$	$\frac{1}{64}f_{11}f_{12}^2$	$\frac{1}{16}f_{11}f_{12}^2$	$\frac{1}{16}f_{11}f_{12}^2$
$(2\omega_1,\omega_1-2\omega_2)$	$\frac{1}{256}f_{11}f_{122}^2$	$\frac{1}{64}f_{11}f_{122}^2$	$\frac{1}{64}f_{11}f_{122}^2$	$\frac{1}{16}f_{11}f_{122}^2$
$(2\omega_1,\omega_1+2\omega_2)$	$\frac{1}{256}f_{11}f_{122}^2$	$\frac{1}{64}f_{11}f_{122}^2$	$\frac{1}{64}f_{11}f_{122}^2$	$\frac{1}{16}f_{11}f_{122}^2$
$(2\omega_2,\omega_2)$	$\frac{1}{16}f_2^2f_{22}$	0	$\frac{1}{4}f_2^2f_{22}$	0
$(2\omega_2,2\omega_2)$	$\frac{1}{32}f_{22}^2(f_{11}+f_{22})$	$\frac{1}{16}f_{22}^3$	$\frac{1}{8}f_{22}^2(f_{11}+\frac{3}{2}f_{22})$	$\frac{1}{4}f_{22}^3$
$(2\omega_2,\omega_1+\omega_2)$	$\frac{1}{64}f_{12}^2f_{22}$	$\frac{1}{64}f_{22}f_{12}^2$	$\frac{1}{16}f_{12}^2f_{22}$	$\frac{1}{16}f_{12}^2f_{22}$
$(2\omega_2,\omega_1+2\omega_2)$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{3f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{3f_{122}}{4})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{7f_{122}}{4})$
$(2\omega_2,\omega_1)$	$\frac{1}{64}f_{122}f_{22}(f_1+\frac{f_{122}}{2})$	$\frac{1}{64}f_{122}f_{22}(f_1-\frac{f_{122}}{2})$	$\frac{1}{16}f_{122}f_{22}(f_1+\frac{3f_{122}}{4})$	$\frac{1}{16}f_{122}f_{22}(f_1-\frac{f_{122}}{4})$
$(\omega_1+\omega_2,\omega_1)$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{1}{8}f_{12}f_2(f_1+\frac{3f_{122}}{4})$	$\frac{1}{16}f_{12}f_{122}f_2$
$(\omega_1+\omega_2,\omega_2)$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{1}{8}f_{12}f_2(f_1+\frac{3f_{122}}{4})$	$\frac{1}{16}f_{12}f_{122}f_2$
$(\omega_1+\omega_2,2\omega_1)$	$\frac{1}{64}f_{11}f_{12}^2$	$\frac{1}{32}f_{11}f_{12}^2$	$\frac{3}{64}f_{11}f_{12}^2$	$\frac{1}{16}f_{11}f_{12}^2$
$(\omega_1+\omega_2,2\omega_2)$	$\frac{1}{64}f_{22}f_{12}^2$	$\frac{1}{32}f_{22}f_{12}^2$	$\frac{3}{64}f_{22}f_{12}^2$	$\frac{1}{16}f_{22}f_{12}^2$
$(\omega_1+\omega_2,\omega_1+\omega_2)$	$\frac{1}{32}f_{12}^2(f_{11}+f_{22})$	$\frac{1}{32}f_{12}^2(f_{11}+f_{22})$	$\frac{3}{32}f_{12}^2(f_{11}+f_{22})$	$\frac{1}{16}f_{12}^2(f_{11}+f_{22})$
$(\omega_1+\omega_2,\omega_1-\omega_2)$	$\frac{1}{64}f_{22}f_{12}^2$	$\frac{1}{32}f_{22}f_{12}^2$	$\frac{3}{64}f_{22}f_{12}^2$	$\frac{1}{16}f_{22}f_{12}^2$
$(\omega_1+\omega_2,\omega_1+2\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{3}{64}f_{12}f_{122}f_2$	$\frac{1}{16}f_{12}f_{122}f_2$
$(\omega_1-\omega_2,\omega_1-\omega_2)$	$\frac{1}{32}f_{12}^2(f_{11}+f_{22})$	$\frac{1}{32}f_{12}^2(f_{11}-f_{22})$	$\frac{1}{32}f_{12}^2(f_{11}-f_{22})$	0^*
$(\omega_1-\omega_2,2\omega_1)$	$\frac{1}{64}f_{11}f_{12}^2$	$\frac{1}{32}f_{11}f_{12}^2$	$\frac{1}{64}f_{11}f_{12}^2$	0^*
$(\omega_1-\omega_2,\omega_1-2\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{1}{64}f_{12}f_{122}f_2$	0^*
$(\omega_1-\omega_2,\omega_1)$	$\frac{1}{16}f_{12}f_2(f_1+\frac{f_{122}}{2})$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{1}{32}f_{12}f_{122}f_2$	0^*
$(\omega_1-\omega_2,\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	$\frac{1}{32}f_{12}f_{122}f_2$	$\frac{1}{64}f_{12}f_{122}f_2$	0^*
$(\omega_1-\omega_2,\omega_1+\omega_2)$	$\frac{1}{64}f_{12}^2f_{22}$	$\frac{1}{32}f_{12}^2f_{22}$	$\frac{1}{64}f_{12}^2f_{22}$	0^*
$(\omega_1-2\omega_2,\omega_1-\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	0	$\frac{3}{64}f_{12}f_{122}f_2$	0
$(\omega_1-2\omega_2,\omega_1-2\omega_2)$	$\frac{1}{128}f_{122}^2(f_{11}+f_{22})$	$\frac{1}{128}f_{122}^2(f_{11}-2f_{22})$	$\frac{1}{128}f_{122}^2(2f_{11}+5f_{22})$	$\frac{3}{128}f_{122}^2(f_{11}-2f_{22})$
$(\omega_1-2\omega_2,2\omega_1)$	$\frac{1}{256}f_{11}f_{122}^2$	$\frac{1}{128}f_{11}f_{122}^2$	$\frac{1}{128}f_{11}f_{122}^2$	$\frac{3}{128}f_{11}f_{122}^2$
$(\omega_1-2\omega_2,\omega_1)$	$\frac{1}{128}f_{122}f_{22}(f_{122}+2f_1)$	$\frac{1}{64}f_{22}f_{122}^2$	$\frac{1}{128}f_{122}f_{22}(5f_{122}+6f_1)$	$\frac{3}{64}f_{22}f_{122}^2$
$(\omega_1+2\omega_2,\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	0	$\frac{5}{64}f_{12}f_{122}f_2$	0
$(\omega_1+2\omega_2,\omega_1+\omega_2)$	$\frac{1}{64}f_{12}f_{122}f_2$	0	$\frac{5}{64}f_{12}f_{122}f_2$	0
$(\omega_1+2\omega_2,2\omega_1)$	$\frac{1}{256}f_{11}f_{122}^2$	$\frac{1}{128}f_{11}f_{122}^2$	$\frac{3}{128}f_{11}f_{122}^2$	$\frac{5}{128}f_{11}f_{122}^2$
$(\omega_1+2\omega_2,\omega_1+2\omega_2)$	$\frac{1}{128}f_{122}^2(f_{11}+f_{22})$	$\frac{1}{128}f_{122}^2(f_{11}+2f_{22})$	$\frac{1}{128}f_{122}^2(6f_{11}+7f_{22})$	$\frac{5}{128}f_{122}^2(f_{11}+2f_{22})$
$(\omega_1+2\omega_2,\omega_1)$	$\frac{1}{128}f_{122}f_{22}(f_{122}+2f_1)$	$\frac{1}{64}f_{22}f_{122}^2$	$\frac{1}{128}f_{122}f_{22}(7f_{122}+10f_1)$	$\frac{5}{64}f_{22}f_{122}^2$
$(\omega_1+2\omega_2,2\omega_2)$	$\frac{1}{128}f_{122}f_{22}(f_{122}+2f_1)$	$\frac{1}{64}f_{22}f_{122}^2$	$\frac{1}{128}f_{122}f_{22}(7f_{122}+10f_1)$	$\frac{5}{64}f_{22}f_{122}^2$
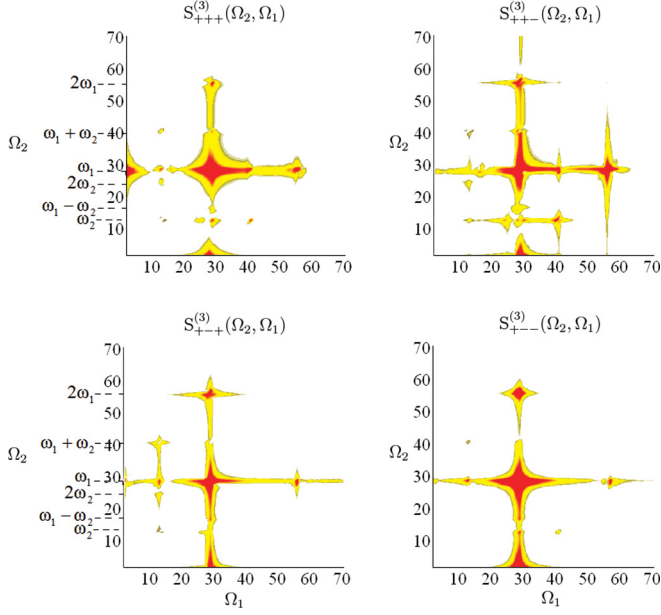
FIG. 2. (Color online) Absolute values of the Fourier transform given by Eq. (3.5) of the correlation functions $R^{(3)}_{+++}(t_2 + t_1, t_1, 0)$, $R^{(3)}_{+-+}(t_2 + t_1, t_1, 0)$, $R^{(3)}_{++-}(t_2 + t_1, t_1, 0)$, and $R^{(3)}_{+--}(t_2 + t_1, t_1, 0)$ for the model of two linearly coupled dissipative Morse oscillators with the fundamental frequencies $\omega_1 = 27.3$ and $\omega_2 = 13.0$.
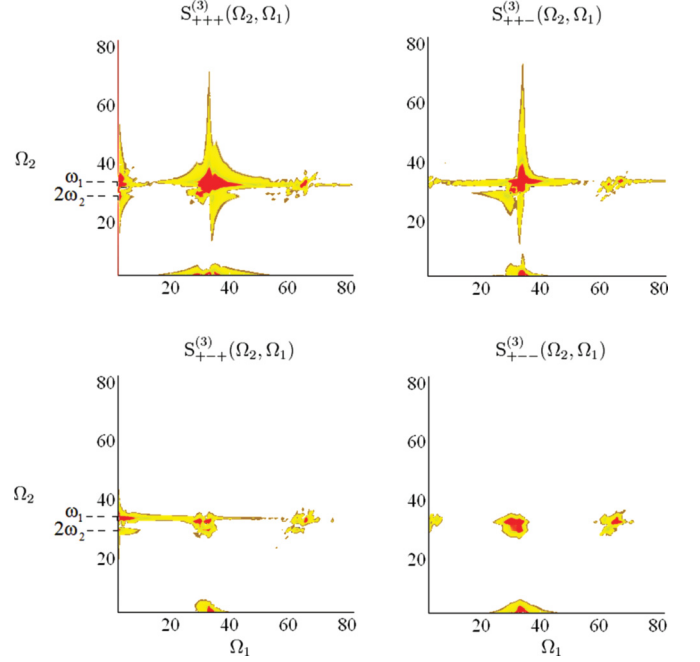

FIG. 3. (Color online) Absolute values of the Fourier transform given by Eq. (3.5) of the correlation functions $R^{(3)}_{+++}(t_2 + t_1, t_1, 0)$, $R^{(3)}_{+-+}(t_2 + t_1, t_1, 0)$, $R^{(3)}_{++-}(t_2 + t_1, t_1, 0)$, and $R^{(3)}_{+--}(t_2 + t_1, t_1, 0)$ for the model of two nonlinearly coupled dissipative harmonic oscillators with the fundamental frequencies $\omega_1 = 31.9$ and $\omega_2 = 14.9$.

that the $(2\omega_1, \omega_1)$ peak in $S^{(3)}_{+-+}(\Omega_2, \Omega_1)$ and $S^{(3)}_{+--}(\Omega_2, \Omega_1)$ in Fig. 2 is the result of anharmonicity of Morse oscillators and is produced due to the temporal growth of stability derivatives. This peak is absent in harmonic systems (see Table III).

Another model system at thermal equilibrium that we consider is two coupled harmonic oscillators $U_i(x_i) = \omega_i^2 x_i^2 / 2$ with a nonlinear coupling $U_{12} x_1 x_2^2$, each in thermal equilibrium with environment, and a linear observable $V = x_1$. The frequencies ω_1 and ω_2 are close to $2:1$ resonance. Figure 3 represents the numerical 2D Fourier transforms of the corresponding GRF calculated in this system. One can observe rather strong diagonal peaks at $(2\omega_2, 2\omega_2)$. By referring to Table III, one can conclude that these peaks are due to the significant quadratic f_{22} contribution of the second mode x_2 to the signal $V(t) = x_1(t)$ due to the nonlinear coupling between the harmonic modes. For the same reason the diagonal peaks (ω_1, ω_1) are strong in $S^{(3)}_{+++}(\Omega_2, \Omega_1)$ and $S^{(3)}_{++-}(\Omega_2, \Omega_1)$ and weak in $S^{(3)}_{+-+}(\Omega_2, \Omega_1)$ and $S^{(3)}_{+--}(\Omega_2, \Omega_1)$, because the latter two 2D signals do not depend on f_{22}. Additionally, the strong diagonal (ω_1, ω_1) peaks in $S^{(3)}_{+++}(\Omega_2, \Omega_1)$ and $S^{(3)}_{++-}(\Omega_2, \Omega_1)$ do not allow one to resolve the weak cross peaks $(\omega_1, 2\omega_2)$ and $(2\omega_2, \omega_1)$, which are much easier to resolve in $S^{(3)}_{+-+}(\Omega_2, \Omega_1)$ and $S^{(3)}_{+--}(\Omega_2, \Omega_1)$ 2D signals, and which thus provide useful complementary information. These cross peaks are indicators of the $1:2$ Fermi resonance as it follows from Table III.

As one can see either from Eq. (3.6) or from Figs. 2 and 3, while the 2D signal $S^{(3)}_{++-}(\Omega_2, \Omega_1)$ contains the same information as $S^{(3)}_{+++}(\Omega_2, \Omega_1)$, and $S^{(3)}_{+--}(\Omega_2, \Omega_1)$ contains the same information as $S^{(3)}_{+-+}(\Omega_2, \Omega_1)$, the former spectra are amplified versions of the latter. For instance, from Eq. (3.6) we have $|S^{(3)}_{++-}(\Omega_2, \Omega_1)| = \beta \Omega_1 |S^{(3)}_{+++}(\Omega_2, \Omega_1)|$, which means

that the low intensity peaks at higher values of Ω_1 that are poorly resolved in the spectra $S^{(3)}_{+++}(\Omega_2, \Omega_1)$ will be much better resolved in the spectra $S^{(3)}_{++-}(\Omega_2, \Omega_1)$. And opposite, the strong peaks at lower values of Ω_1 in $S^{(3)}_{+++}(\Omega_2, \Omega_1)$ will be suppressed in $S^{(3)}_{++-}(\Omega_2, \Omega_1)$ (since they will be multiplied by small Ω_1). The latter provides a significant flexibility for the exploration of multidimensional spectra and investigation of complex dynamics by designing a proper experiment with multiple perturbations and measurements.

VI. FREQUENCY DOMAIN MEASUREMENTS: SUSCEPTIBILITIES

In the previous sections we discussed time domain signals $R^{(n)}_{+,\pm,...,\pm}(\tau_1, ..., \tau_k, \tau'_1, ..., \tau'_m)$, that keep track of time ordering of perturbations $V_-(\tau'_j)$ and measurements $V_+(\tau_i)$. However, in some cases it can be convenient to carry the experiment in the frequency domain. Frequency domain experiments which combine multiple measurements of an observable with multiple perturbations of the system can be performed in the following way. One can perturb a system with frequencies $\omega'_1, ..., \omega'_m$, while measuring k observables $V(\omega)$ at frequencies $\omega_1, ..., \omega_k$ and constructing a k-point correlation function $\langle V(\omega_1) \cdots V(\omega_k) \rangle$. For this type of experiments, time ordering of measurements and perturbations loses its sense. The correlation function then depends on all possible permutations of m perturbations and $k-1$ measurements (except for the last one, which cannot be substituted by a perturbation due to causality), where $m + k = n$.

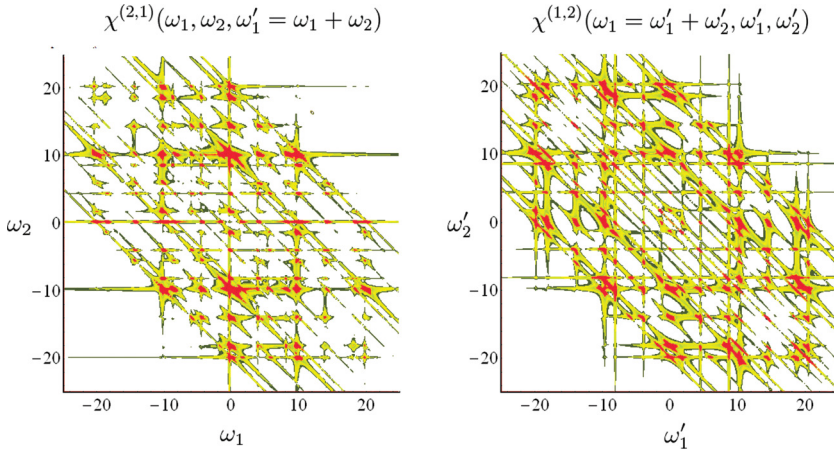First, let us consider the Fourier transform of the outputs of the time domain experiments with one

FIG. 4. (Color online) Generalized susceptibilities for a system of two uncoupled harmonic oscillators with V(t) from Eq. (5.2).

perturbation and two measurements, which are ordered in time. These experiments will be governed by the corresponding generalized response functions $R_{++-}^{(3)}(\tau_3,\tau_2,\tau_1) = \theta(\tau_3-\tau_2)\theta(\tau_2-\tau_1)\langle V_+(\tau_3)V_+(\tau_2)V_-(\tau_1)\rangle$ and $R_{+-+}^{(3)}(\tau_3,\tau_2,\tau_1) = \theta(\tau_3-\tau_2)\theta(\tau_2-\tau_1)\langle V_+(\tau_3)V_-(\tau_2)V_+(\tau_1)\rangle$. The two-point correlation functions that can be measured in these experiments, and which are defined in Eq. (1.1), are, respectively,

$$\langle V(\tau_3)V(\tau_2)\rangle_{++-} = \int d\tau_1 R_{++-}^{(3)}(\tau_3,\tau_2,\tau_1)E(\tau_1),$$
$$\langle V(\tau_3)V(\tau_1)\rangle_{+-+} = \int d\tau_2 R_{+-+}^{(3)}(\tau_3,\tau_2,\tau_1)E(\tau_2). \tag{6.1}$$

Introducing the Fourier transform

$$F(\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\Omega \widetilde{F}(\Omega)e^{-\iota\Omega\tau}, \quad \widetilde{F}(\Omega) = \int_{-\infty}^{\infty} d\tau F(\tau)e^{\iota\Omega\tau}, \tag{6.2}$$

and using the definition in Eq. (3.5), one gets

$$\langle \widetilde{V}(\Omega_3)\widetilde{V}(\Omega_2)\rangle_{++-}$$
$$= \int d\Omega_1 \widetilde{E}(\Omega_1)S_{++-}(\Omega_3,\Omega_3+\Omega_2)\delta(\Omega_3+\Omega_2-\Omega_1),$$
$$\langle \widetilde{V}(\Omega_3)\widetilde{V}(\Omega_1)\rangle_{+-+}$$
$$= \int d\Omega_2 \widetilde{E}(\Omega_2)S_{+-+}(\Omega_3,\Omega_3-\Omega_2)\delta(\Omega_3+\Omega_1-\Omega_2). \tag{6.3}$$

As we discussed in the beginning of this section, in the frequency domain, the measured correlation function is the combination of the two time-ordered correlation functions, i.e.,

$$\langle \widetilde{V}(\omega_2)\widetilde{V}(\omega_1)\rangle_{2,1}$$
$$= \tfrac{1}{2}[\langle \widetilde{V}(\omega_2)\widetilde{V}(\omega_1)\rangle_{++-} + \langle \widetilde{V}(\omega_2)\widetilde{V}(\omega_1)\rangle_{+-+}]. \tag{6.4}$$

Substituting Eq. (6.3) into Eq. (6.4), we get

$$\langle \widetilde{V}(\omega_2)\widetilde{V}(\omega_1)\rangle_{2,1} = \frac{1}{2\pi}\int d\omega_1' \chi^{(2,1)}(\omega_1,\omega_2,\omega_1')\widetilde{E}(\omega_1'), \tag{6.5}$$

where the generalized susceptibility $\chi^{(2,1)}$ is

$$\chi^{(2,1)}(\omega_1,\omega_2,\omega_1')$$
$$= 2\pi\delta(\omega_2+\omega_1-\omega_1')\tfrac{1}{2}[S_{++-}(\omega_2,\omega_2+\omega_1)$$
$$+ S_{+-+}(\omega_2,\omega_2-\omega_1')]. \tag{6.6}$$

The latter result can be generalized to the case of k measurements and m perturbations, with the corresponding susceptibility $\chi^{(k,m)}$, $m+k=n$, introduced in Eq. (1.2):

$$\chi^{(k,m)}(\omega_1,\ldots,\omega_k,\omega_1',\ldots,\omega_m')$$
$$= 2\pi\delta\left(\sum_{i=1}^{k}\omega_i - \sum_{j=1}^{m}\omega_j'\right)\frac{1}{(n-1)!}\sum_{p}$$
$$\times S_{+,\nu_1,\ldots,\nu_{n-1}}(\omega_k,\omega_k+\omega_{\nu_1},\ldots,\omega_k$$
$$+ \omega_{\nu_1}+\cdots+\omega_{\nu_{n-2}}). \tag{6.7}$$

The summation in Eq. (6.7) is over the $(n-1)!$ permutations of indices $\nu_j = +,-$ (m of these indices are "$-$", and $k-1$ indices are "$+$"), with the following rule $\omega_+ \equiv \omega_j$ and $\omega_- \equiv -\omega_j'$. Using the rule of Eq. (6.7), the two remaining three-point susceptibilities $\chi^{(3,0)}(\omega_1,\omega_2,\omega_3)$ and $\chi^{(1,2)}(\omega_1,\omega_1',\omega_2')$, corresponding, respectively, to three measurements (i.e., the symmetrized three-point correlation function in the frequency domain), and two perturbations with one measurement (i.e., the second-order susceptibility from the response theory) read

$$\chi^{(3,0)}(\omega_1,\omega_2,\omega_3)$$
$$= 2\pi\delta(\omega_3+\omega_2+\omega_1)\tfrac{1}{2}[S_{+++}(-\omega_2-\omega_1,-\omega_2)$$
$$+ S_{+++}(-\omega_2-\omega_1,-\omega_1)], \tag{6.8}$$

$$\chi^{(1,2)}(\omega_1,\omega_1',\omega_2')$$
$$= 2\pi\delta(\omega_1-\omega_1'-\omega_2')\tfrac{1}{2}[S_{+--}(\omega_2'+\omega_1',\omega_2')$$
$$+ S_{+--}(\omega_2'+\omega_1',\omega_1')], \tag{6.9}$$

where $S_{+,\pm,\pm}$ were defined in Eq. (3.6), and in both cases the arguments of the S functions were slightly rearranged by expressing one frequency in terms of the two others within the constraints of the delta functions. We can also rewrite Eq. (6.6) in a similar form using the delta-function constraint $\omega_2 = \omega_1' - \omega_1$:

$$\chi^{(2,1)}(\omega_1,\omega_2,\omega_1')$$
$$= 2\pi\delta(\omega_2+\omega_1-\omega_1')\tfrac{1}{2}[S_{++-}(-\omega_1+\omega_1',\omega_1')$$
$$+ S_{+-+}(-\omega_1+\omega_1',-\omega_1)]. \tag{6.10}$$
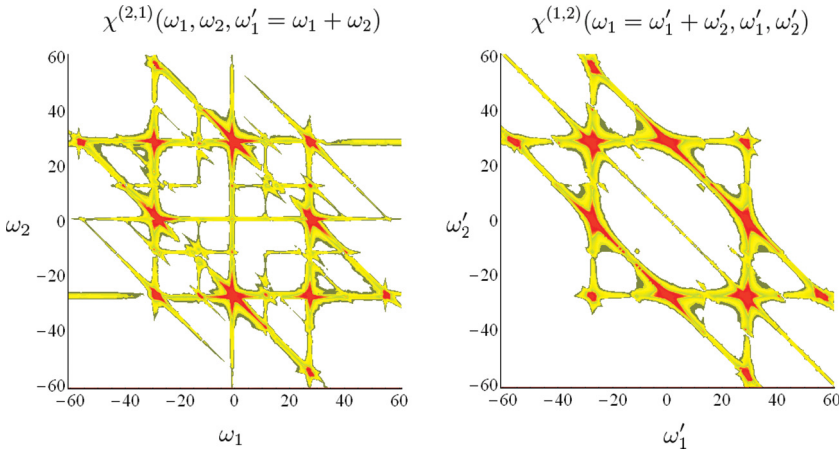
FIG. 5. (Color online) Generalized susceptibilities for a system of two bilinearly coupled dissipative Morse oscillators with the fundamental frequencies $\omega_1 = 27.3$ and $\omega_2 = 13.0$ and $V(t) = x_1(t)$.

The susceptibilities given by Eqs. (6.9) and (6.10) were calculated numerically for the model systems discussed in Sec. V and are shown in Figs. 4 and 5.

It should be noted that the symmetrization procedure given by Eq. (6.7), that is required for frequency domain measurements, reduces the number of independent n-point GRF to $n-1$ due to the loss of information on time ordering. While for systems at thermal equilibrium the number of independent n-point measures in the time domain is equal to $n-1$, this number is expected to be larger for nonthermal systems, for which there exists no simple relation between the distribution density $\rho(E)$ and its derivative $\partial\rho(E)/\partial E$.

VII. DISCUSSION

We proposed and analyzed a class of experiments in which multiple measurements of a dynamical variable are combined with multiple perturbations of the corresponding dynamical system. The $n-1$ time intervals between m perturbations and $k = n - m$ measurements constitute $(n-1)$ dimensional space and result in a family of $(n-1)$ dimensional measures $R^{(k,m)}(t_{n-1}, \ldots, t_1)$. These objects can be directly measured by subjecting a system to weak impulsive perturbations. In the latter family of $(n-1)$ dimensional measures, $n-1$ of them are independent and thus can provide additional information on dynamics to the one already contained in the usual multipoint correlation function, $R^{(n,0)}(t_{n-1}, \ldots, t_1)$, and response function, $R^{(1,n-1)}(t_{n-1}, \ldots, t_1)$.

Each perturbation introduces a new dynamical quantity, the stability derivative, to an n-point correlation function $R^{(k,m)}(t_{n-1}, \ldots, t_1)$, which carries information on classical coherence, i.e., coherence of the nearby classical trajectories. Various combinations of perturbations and measurements thus lead to different time correlation functions between classical dynamical quantities and their classical coherences. On the other hand, in case of weakly anharmonic systems we have shown that a perturbation of a dynamical quantity cancels some of its random phase by launching two classical trajectories with infinitely close initial conditions. The latter allow us to manipulate the total random phase of the n-point time correlation functions, and thus to change the resulting $(n-1)$ dimensional spectra. These differences result in new

spectral peaks or contributions of different mode couplings to their intensities. Additionally, one can amplify or suppress different parts of multidimensional spectra by choosing the appropriate combination of perturbations and measurements. In particular, we have shown that while the 2D signals $S_{+++}(\Omega_2, \Omega_1)$ and $S_{+--}(\Omega_2, \Omega_1)$ contain the same information as $S_{+-}(\Omega_2, \Omega_1)$ and $S_{+--}(\Omega_2, \Omega_1)$, respectively, for systems at thermal equilibrium, the former provide better resolution of spectral peaks at lower Ω_1, while the latter provide better resolution at higher Ω_1.

In the present paper we only discussed applications of this class of multidimensional measures to Hamiltonian systems, yet the approach can be extended to non-Hamiltonian dynamical systems such as chemical reactions, electric currents, currents in membranes, biological systems, and systems initially at a steady state. For instance, the method of multiple perturbations and measurements can be used to obtain new information on structure and connectivity of complex biological networks. A limited number of methods is currently available to study complex reaction mechanisms [37]. The introduced GRF, with their special type of correlations between multiple perturbations, are believed to serve as a useful tool to study correlations in complex reaction networks and thus help to extract information on their topology. The same ideas can be applied to investigate topology of electric currents in biological systems. Thus, the proposed method of multiple perturbations and measurements can provide tools to study complex behavior of general dynamical systems and will be analyzed in future extensions.

APPENDIX

In this Appendix we provide classical expressions of the four-point measures $R_{+,\pm,\pm,\pm}$ for systems with Boltzmann

distribution:

$$R^c_{++++}(\tau_4,\tau_3,\tau_2,\tau_1) = \langle V(\tau_4)V(\tau_3)V(\tau_2)V(\tau_1)\rangle,$$

$$R^c_{+++-}(\tau_4,\tau_3,\tau_2,\tau_1) = \beta\frac{d}{d\tau_1}R^c_{++++}(\tau_4,\tau_3,\tau_2,\tau_1),$$

$$R^c_{++-+}(\tau_4,\tau_3,\tau_2,\tau_1) = -\langle V(\tau_4)V(\tau_3)\{V(\tau_2),V(\tau_1)\}\rangle$$
$$+ \beta\frac{d}{d\tau_2}R^c_{++++}(\tau_4,\tau_3,\tau_2,\tau_1),$$

$$R^c_{++--}(\tau_4,\tau_3,\tau_2,\tau_1) = \beta\frac{d}{d\tau_1}R^c_{++-+}(\tau_4,\tau_3,\tau_2,\tau_1),$$

$$R^c_{+-++}(\tau_4,\tau_3,\tau_2,\tau_1) = -\langle V(\tau_4)V(\tau_1)\{V(\tau_3),V(\tau_2)\}\rangle$$
$$-\langle V(\tau_4)V(\tau_2)\{V(\tau_3),V(\tau_1)\}\rangle$$
$$+ \beta\frac{d}{d\tau_3}R^c_{++++}(\tau_4,\tau_3,\tau_2,\tau_1),$$

$$R^c_{+-+-}(\tau_4,\tau_3,\tau_2,\tau_1) = \beta\frac{d}{d\tau_1}R^c_{+-++}(\tau_4,\tau_3,\tau_2,\tau_1),$$

$$R^c_{+--+}(\tau_4,\tau_3,\tau_2,\tau_1) = -\beta\frac{d}{d\tau_3}\langle V(\tau_4)V(\tau_3)\{V(\tau_2),V(\tau_1)\}\rangle$$
$$-\beta\frac{d}{d\tau_2}\langle V(\tau_4)V(\tau_2)\{V(\tau_3),V(\tau_1)\}\rangle$$
$$-\beta\frac{d}{d\tau_2}\langle V(\tau_4)V(\tau_1)\{V(\tau_3),V(\tau_2)\}\rangle$$
$$+\langle V(\tau_4)\{V(\tau_3),\{V(\tau_2),V(\tau_1)\}\}\rangle$$
$$+ \beta^2\frac{d^2}{d\tau_2 d\tau_3}R^c_{++++}(\tau_4,\tau_3,\tau_2,\tau_1),$$

$$R^c_{+---}(\tau_4,\tau_3,\tau_2,\tau_1) = \beta\frac{d}{d\tau_1}R^c_{+--+}(\tau_4,\tau_3,\tau_2,\tau_1). \quad (A1)$$

The Fourier transforms given by Eq. (3.5) of the above quantities are

$$S_{++++}(\Omega_3,\Omega_2,\Omega_1) = \int \tilde{C}_1(\omega_3,\omega_2,\omega_1)$$
$$\times \tilde{\theta}(\Omega_1 - \omega_1)\tilde{\theta}(\Omega_2 - \omega_2)$$
$$\times \tilde{\theta}(\Omega_3 - \omega_3)d\omega_1 d\omega_2 d\omega_3$$

$$S_{+++-}(\Omega_3,\Omega_2,\Omega_1) = i\beta\Omega_1 S_{++++}(\Omega_3,\Omega_2,\Omega_1)$$

$$S_{++-+}(\Omega_3,\Omega_2,\Omega_1) = -\int \tilde{C}_2(\omega_3,\omega_2,\omega_1)$$
$$\times \tilde{\theta}(\Omega_1 - \omega_1)\tilde{\theta}(\Omega_2 - \omega_2)$$
$$\times \tilde{\theta}(\Omega_3 - \omega_3)d\omega_1 d\omega_2 d\omega_3$$
$$+ i\beta(\Omega_2 - \Omega_1)S_{++++}(\Omega_3,\Omega_2,\Omega_1)$$

$$S_{++--}(\Omega_3,\Omega_2,\Omega_1) = i\beta\Omega_1 S_{++-+}(\Omega_3,\Omega_2,\Omega_1)$$

$$S_{+-++}(\Omega_3,\Omega_2,\Omega_1) = -\int \tilde{\theta}(\Omega_1 - \omega_1)\tilde{\theta}(\Omega_2 - \omega_2)$$
$$\times \tilde{\theta}(\Omega_3 - \omega_3)d\omega_1 d\omega_2 d\omega_3$$
$$\times [\tilde{C}_2(\omega_3,\omega_3 - \omega_1,\omega_2 - \omega_1)$$
$$+ \tilde{C}_2(\omega_3,\omega_1 - \omega_2 + \omega_3,\omega_1)]$$
$$+ i\beta(\Omega_3 - \Omega_2)S_{++++}(\Omega_3,\Omega_2,\Omega_1)$$

$$S_{+-+-}(\Omega_3,\Omega_2,\Omega_1) = i\beta\Omega_1 S_{+-++}(\Omega_3,\Omega_2,\Omega_1)$$

$$S_{+--+}(\Omega_3,\Omega_2,\Omega_1) = \int \tilde{\theta}(\Omega_1 - \omega_1)\tilde{\theta}(\Omega_2 - \omega_2)$$
$$\times \tilde{\theta}(\Omega_3 - \omega_3)d\omega_1 d\omega_2 d\omega_3$$
$$\times [i\beta(\omega_2 - \omega_3)\tilde{C}_2(\omega_3,\omega_2,\omega_1)$$
$$+ i\beta(\omega_1 - \omega_2)\tilde{C}_2(\omega_3,\omega_1 - \omega_2 + \omega_3,\omega_1)$$
$$- i\beta(\omega_2 - \omega_1)\tilde{C}_2(\omega_3,\omega_3 - \omega_1,\omega_2 - \omega_1)$$
$$+ \tilde{C}_3(\omega_3,\omega_2,\omega_1)]$$
$$+ \beta^2(\Omega_2 - \Omega_3)(\Omega_1 - \Omega_2)$$
$$\times S_{++++}(\Omega_3,\Omega_2,\Omega_1)$$

$$S_{+---}(\Omega_3,\Omega_2,\Omega_1) = i\beta\Omega_1 S_{+--+}(\Omega_3,\Omega_2,\Omega_1) \quad (A2)$$

where $\tilde{C}_1(\Omega_3,\Omega_2,\Omega_1) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\langle V(t_3+t_2+t_1)V(t_2+t_1)$ $V(t_1)V(0)\rangle\ e^{i\Omega_3 t_3}e^{i\Omega_2 t_2}e^{i\Omega_1 t_1}dt_1 dt_2 dt_3$ and $\tilde{C}_2(\Omega_3,\Omega_2,\Omega_1)$, $\tilde{C}_3(\Omega_3,\Omega_2,\Omega_1)$ are defined similarly by replacing the correlation function under the integral with $\langle V(t_3 + t_2 + t_1)V(t_2 + t_1),\{V(t_1),V(0)\}\rangle$ and $\langle V(t_3 + t_2 + t_1)\{V(t_2 + t_1),\{V(t_1),V(0)\}\}\rangle$, respectively; $\tilde{\theta}(\Omega) = \pi\delta(\Omega) + \frac{i}{\Omega}$ is the Fourier transform of the Heaviside step function.

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