Thermal response of nonequilibrium RC circuits

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We analyze experimental data obtained from an electrical circuit having components at different temperatures, showing how to predict its response to temperature variations. This illustrates in detail how to utilize a recent linear response theory for nonequilibrium overdamped stochastic systems. To validate these results, we introduce a reweighting procedure that mimics the actual realization of the perturbation and allows extracting the susceptibility of the system from steady state data. This procedure is closely related to other fluctuation-response relations based on the knowledge of the steady state probability distribution. As an example, we show that the nonequilibrium heat capacity in general does not correspond to the correlation between the energy of the system and the heat flowing into it. Rather, also non-dissipative aspects are relevant in the nonequilbrium fluctuation response relations.

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INTRODUCTION

Understanding how a system responds to variations of its parameters is one of the basic features of science. It is well known that systems in thermodynamic equilibrium when slightly perturbed find their way back to a new steady regime by dissipation. The spontaneous correlations in the unperturbed system between this transient entropic change and an observable anticipates us how that observable would react to the actual perturbation. This is at the basis of the fluctuation-dissipation theorem and of related response relations, which hold in great generality in equilibrium [1, 2].

Out of equilibrium, in contrast, there are multiple linear response theories [2–4], some based on the manipulation of the density of states [5–9], some on dynamical systems techniques for evolving observables [10–12], and some on a path-weight approach for stochastic systems [13–17]. The latter has revealed that entropy production is not sufficient for understanding the linear response of nonequilibrium systems. There are rather non-dissipative aspects of the system vs perturbation relation that are equally relevant.

Within the linear response theory one finds recent approaches focusing on temperature perturbations [18–26], which lead for instance to a formulation of nonequilibrium heat capacity [19], a notion that should be useful for constructing a steady state thermodynamics [27–31]. The question is how a system far from equilibrium reacts to a change of one or many of its bath temperatures. For example, one could be interested in the response to temperature variations of a glassy system undergoing a relaxation process [32, 33]. Alternatively, a nonequilibrium steady state may be imposed by putting the system in contact with two reservoirs at different temperatures [34–36]. It is the case of an experiment recently realized with a simple desktop electric circuit in which one resistor was kept at room temperature while the other was maintained at a lower temperature [34, 35].

In this paper, we analyze the experiments of the thermally unbalanced electric circuit [34, 35]. The primary goal of this work is to show how to apply in practice a fluctuation-response relation [25, 26] for computing the susceptibility of the system to a change of one temperature. This is a stand-alone procedure for predicting the thermal linear response of the system. Just to validate its results, we compare them with an alternative estimate of the susceptibility, which is introduced here to exploit the knowledge of the steady state data (which is accessible for the simple system analyzed), used by us in a reweighted form to replace the actual application of the perturbation. This useful procedure constitutes a new result of this work. We also show the connection of this reweighting procedure with another fluctuation-response relation based on the steady state distribution, put forward by Seifert and Speck [8].

In the following section we describe the experimental setup, then we recall the structure of the fluctuation-response relation and we specialize it to our system. In Sec. III we introduce the reweighting procedure, and in Sec. IV we show how to compute a nonequilibrium version of the heat capacity. The conclusions are followed by an appendix in which we recall in detail the steps to compute the Gaussian steady state distribution of linear stable systems and we specify its form for the electrical circuit.

I. EXPERIMENTAL SET-UP

Our experimental set-up is sketched in Fig.1(a). It is constituted by two resistors R_1 and R_2, which are kept at different temperature, T_1 and T_2, respectively. These temperatures are controlled by thermal baths and T_2 is kept fixed at $296K$ whereas T_1 can be set at a value between $296K$ and $88K$ using the stratified vapor above a liquid nitrogen bath. The coupling capacitor C controls the electrical power ex-

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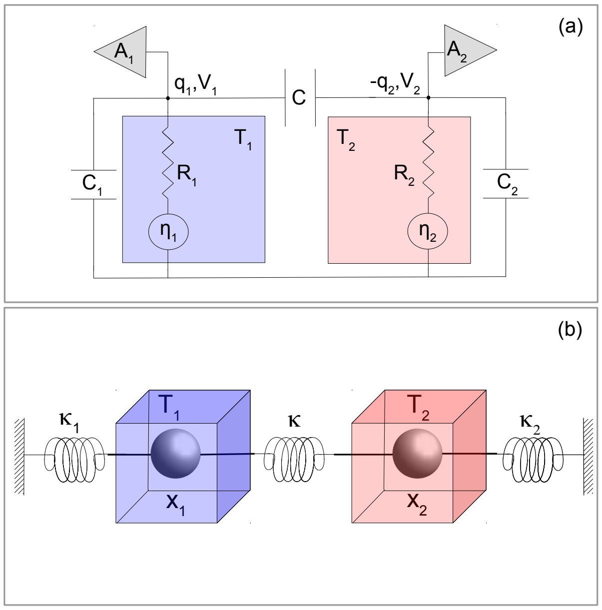

Figure 1. (Color online) (a) Diagram of the circuit. The resistor R_1 is kept at temperature T_1 while R_2 is always at room temperature $T_2 = 296K$. They are coupled via the capacitor C. The capacitors C_1 and C_2 account for the capacitance of the wiring, etc. The voltage generators η_1 and η_2 represent thermal fluctuations of the voltage that the resistors undergo. (b) Equivalent mechanical system with two Brownian particles moving in fluids at different temperatures T_1 and T_2, but trapped and coupled by harmonic springs.

changed between the resistors and as a consequence the energy exchanged between the two baths. No other coupling exists between the two resistors, which are inside two separated screened boxes. The quantities C_1 and C_2 are the capacitances of the circuits and the cables. Two extremely low noise amplifiers A_1 and A_2 [37] measure the voltage V_1 and V_2 across the resistors R_1 and R_2, respectively. All the relevant quantities considered in this paper can be derived by the measurements of V_1 and V_2, as discussed below. In particular, the relationships between the measured voltages and the charges are

$$q_1 = (V_1 - V_2)\, C + V_1\, C_1\,, \tag{1}$$
$$q_2 = (V_1 - V_2)\, C - V_2\, C_2\,. \tag{2}$$

Assuming an initially neutral circuit, we denote by q_1 the charge that has flown through the resistor R_1 into the node at potential V_1, and by q_2 the charge that has flown through R_2 *out of* the node at V_2. By analyzing the circuit one finds that the equations of motion for these charges are

$$R_1 \dot{q}_1 = -\frac{C_2}{X} q_1 + \frac{C}{X}(q_2 - q_1) + \eta_1 \tag{3a}$$
$$R_2 \dot{q}_2 = -\frac{C_1}{X} q_2 + \frac{C}{X}(q_1 - q_2) + \eta_2 \tag{3b}$$

where

$$X = C\, C_1 + C\, C_2 + C_1\, C_2 \tag{4}$$

and $\eta_i(t)$ is a white noise satisfying $\langle \eta_i(t) \eta_j(t') \rangle = 2\delta_{ij} k_{\rm B} T_i R_i \delta(t - t')$. Indeed, in Fig.1(a) the two resistances have been drawn with their associated thermal noise generators η_1 and η_2, whose power spectral densities are given by the Nyquist formula $|\tilde{\eta}_m|^2 = 4 k_B R_m T_m$, with $m = 1, 2$.

More details on the experimental set-up can be found in Refs. [34, 35]. For the data used for the analysis discussed in the following section, the values of the components are: $C = 100$pF, $C_1 = 680$pF, $C_2 = 420$pF and $R_1 = R_2 = 10$MΩ. The longest characteristic time of the system is $Y = (C_1 + C) R_1 + (C_2 + C) R_2$, which for the mentioned values of the parameters is $Y = 13$ ms.

II. THERMAL RESPONSE

The system has $N = 2$ degrees of freedom. Eqs. (3) can be mapped onto the mechanical system in Fig. 1(b) involving two Brownian particles coupled by harmonic springs,

$$\dot{x}_1 = \mu_1 F_1(\boldsymbol{x}) + \sqrt{2 \mu_1 k_{\rm B} T_1}\, \xi_1 \tag{5a}$$
$$\dot{x}_2 = \mu_2 F_2(\boldsymbol{x}) + \sqrt{2 \mu_2 k_{\rm B} T_2}\, \xi_2 \tag{5b}$$

Here $\boldsymbol{x} = (x_1, x_2)$ are the two positions with $x_i = 0$ when the springs are at rest, $\boldsymbol{T} = (T_1, T_2)$ the temperatures, and the (harmonic) forces $\boldsymbol{F} = (F_1, F_2)$ are derived from the potential

$$U(\boldsymbol{x}) = \frac{1}{2} \left[\kappa_1 x_1^2 + \kappa (x_2 - x_1)^2 + \kappa_2 x_2^2 \right] \,. \tag{6}$$

The detailed mapping between the electrical and mechanical models is summarized in Table I; for instance, the admittance $1/R_1$ is mapped to the mobility μ_1. Again, each Gaussian white noise ξ_j is uncorrelated from the other,

$$\langle \xi_j(t) \xi_{j'}(t') \rangle = \delta(t - t') \delta_{jj'}. \tag{7}$$

This recasting in the form (5) allows us to use some recently introduced thermal response formulas [25, 26]. They predict the linear response of an overdamped stochastic system with additive noise, in general nonequilibrium conditions, when the perturbation is a change of one or more temperatures. In accordance with the presentation of that approach, we choose natural units ($k_{\rm B} = 1$) in the following, taking temperatures to have dimensions of energy.

The thermal susceptibility of a state observable $\mathcal{O}(\boldsymbol{x})$ is defined as the response to a step variation $\boldsymbol{T} \to \boldsymbol{\Theta}$ of the set of temperatures, parametrized by a function $\theta(t) = 0$ for times $t < 0$ and $\theta(t) = \theta$ constant for $t \geqslant 0$. In particular, with indicators ϵ_i ($1 \leqslant i \leqslant N$) that specify which temperatures receive the perturbation, here we write

$$\boldsymbol{\Theta} \equiv (T_1 + \epsilon_1 \theta, T_2 + \epsilon_2 \theta) \,. \tag{8}$$

The susceptibility as a function of time t is then

$$\chi^\theta_{\mathcal{O}}(t) = \lim_{\theta \to 0} \frac{\langle \mathcal{O}(\boldsymbol{x}(t)) \rangle_{\boldsymbol{T},\boldsymbol{\Theta}} - \langle \mathcal{O}(\boldsymbol{x}(t)) \rangle_{\boldsymbol{T},\boldsymbol{T}}}{\theta} \,. \tag{9}$$

In the averages $\langle \ldots \rangle_{\boldsymbol{T},\boldsymbol{T'}}$, the first subscript $\boldsymbol{T}$ represents the initial ($t < 0$) temperatures, while the second subscript $\boldsymbol{T'}$ represents the temperatures under which the observed dynamics ($t \geqslant 0$) takes place.

A recent fluctuation-response relation [25, 26] expresses the susceptibility (9) of the state observable $\mathcal{O}(\boldsymbol{x})$ as a sum,

$$\chi_{\mathcal{O}}(t) = S_1 + S_2 + K_1 + K_2 \tag{10}$$

where the terms are

$$S_1 = -\left\langle \mathcal{O}(t) \sum_i \frac{\epsilon_i}{2T_i^2} \int_0^t \dot{x}_i(t') F_i(t')\,\mathrm{d}t' \right\rangle \tag{11a}$$

$$S_2 = \left\langle \mathcal{O}(t) \sum_i \frac{\epsilon_i}{4T_i^2} \sum_{j=1}^{N} \left(\frac{T_i}{T_j} - 1\right) \int_0^t [x_i \dot{x}_j \partial_j F_i](t')\,\mathrm{d}t' \right\rangle \tag{11b}$$

$$K_1 = \left\langle \mathcal{O}(t) \sum_i \frac{\epsilon_i}{4T_i^2} \int_0^t \left[\mu_i F_i^2 + x_i \mathbb{L}^{(T_i)} F_i\right](t')\,\mathrm{d}t' \right\rangle \tag{11c}$$

$$K_2 = \frac{\mathrm{d}}{\mathrm{d}t'} \left\langle \mathcal{O}(t) \sum_i \frac{\epsilon_i}{8\mu_i T_i^2} x_i^2(t') \right\rangle \Bigg|_{t'=0}^{t'=t} \tag{11d}$$

with the shorthand $\partial_j = \partial/\partial x_j$, and $\langle \ldots \rangle = \langle \ldots \rangle_{\boldsymbol{T},\boldsymbol{T}}$ denoting unperturbed averages which have an understood dependence on the distribution $\rho_0(\boldsymbol{x}(0))$ at the time when the perturbation is turned on. (Let us stress that the labels $1, 2$ of these S and K terms have nothing to do with the index of the resistors, particles, etc.) Integrals are in the Stratonovich sense, hence in their discretized version one performs midpoint averages, such as $\dot{x}(t)F(t)\mathrm{d}t \to [x(t+\mathrm{d}t) - x(t)]\frac{1}{2}[F(t+\mathrm{d}t) + F(t)]$. (However, temperatures and mobilities do not depend on the coordinates and the interpretation of the stochastic equation is free.)

The term S_1 is a standard correlation between observable and entropy production, but it contains a prefactor $1/2$ not present in the equilibrium version (Kubo formula). The term S_2 instead correlates the observable with another form of entropy production and clearly it is relevant only if $T_j \neq T_i$ for some (i, j). The terms K_1 and K_2, previously called the *frenetic* terms [3, 20, 24–26], instead correlate the observable with time-symmetric aspects of the dynamics. These are necessarily non-dissipative in nature. In all cases it is understood that we are dealing with quantities in excess due to the perturbation. The generalized generator

$$\mathbb{L}^{(T_i)} = \sum_j \frac{T_i}{T_j} \left[\mu_j F_j(\boldsymbol{x}) \partial_j + \mu_j T_j \partial_j^2\right] \tag{12}$$

was introduced to describe the evolution of the degrees of freedom in terms of the j-th *thermal time* as dictated by the i-th temperature (see [26] for more details). It differs from the backward generator of the dynamics (5),

$$\mathbb{L} = \sum_{j=1}^{N} \left[\mu_j F_j(\boldsymbol{x}) \partial_j + \mu_j T_j \partial_j^2\right] , \tag{13}$$

whose action on a state function inside an average is expressed as $\frac{\mathrm{d}}{\mathrm{d}t} \langle \mathcal{O}(\boldsymbol{x}(t)) \rangle = \langle \mathbb{L}\mathcal{O}(\boldsymbol{x}(t)) \rangle$. The definition of a thermal time permits to recast (5) as isothermal dynamics. For example, if $i = 1$ and hence T_1 is taken as a reference, then the thermal time $\tau_2 = tT_2/T_1$ yields for x_2

$$\frac{dx_2}{d\tau_2} = \frac{T_1}{T_2} \mu_2 F_2(\boldsymbol{x}) + \sqrt{2\mu_2 k_{\mathrm{B}} T_1}\, \xi_2(\tau_2), \tag{14}$$

where the different intensity of the noise ξ_2 (it has now T_1 in the prefactor) is associated with a rescaling $\sim T_1/T_2$ of the mechanical force F_2.

In our analysis we work with experimental trajectory data collected in steady states, where $d/dt' \left\langle \mathcal{O}(t) x^2(t') \right\rangle = -d/dt \left\langle \mathcal{O}(t) x^2(t') \right\rangle = -\left\langle \mathbb{L}\mathcal{O}(t) x^2(t') \right\rangle$ for $t \geqslant t'$. Hence we rather use the alternative form

$$K_2^s = \left\langle \mathbb{L}\mathcal{O}(t) \sum_i \frac{\epsilon_i}{8\mu_i T_i^2} [x_i^2(0) - x_i^2(t)] \right\rangle \tag{15}$$

because it is numerically more stable than K_2 [26]. Since in the given experimental setup it is natural to manipulate T_1 (while the room temperature, T_2, remains unperturbed), we show examples with $\epsilon_1 = 1$ and $\epsilon_2 = 0$. This leads to the susceptibility $\chi_{\mathcal{O}}(t)$ being composed of the specific terms

$$S_1 = -\frac{1}{2T_1^2} \left\langle \mathcal{O}(t) \int_0^t \dot{x}_1(t') F_1(t')\,\mathrm{d}t' \right\rangle \tag{16a}$$

$$S_2 = \frac{1}{4T_1^2} \left\langle \mathcal{O}(t) \left(\frac{T_1}{T_2} - 1\right) \int_0^t [x_1 \dot{x}_2 \partial_2 F_1](t')\,\mathrm{d}t' \right\rangle \tag{16b}$$

$$K_1 = \frac{1}{4T_1^2} \left\langle \mathcal{O}(t) \int_0^t \left[\mu_1 F_1^2 + x_1 \mathbb{L}^{(T_1)} F_1\right](t')\,\mathrm{d}t' \right\rangle \tag{16c}$$

$$K_2 = \frac{1}{8\mu_1 T_1^2} \left\langle \mathbb{L}\mathcal{O}(t) [x_1^2(0) - x_1^2(t)] \right\rangle \tag{16d}$$

with $\mathbb{L}^{(T_1)} = \mu_1[F_1\partial_1 + T_1\partial_1^2] + \mu_2[\frac{T_1}{T_2} F_2 \partial_2 + T_1 \partial_2^2]$. Note that we have dropped the superscript "s" from K_2.

The susceptibility is found as the sum of these correlations with fluctuating trajectory functionals, *predicting* the susceptibility without actually performing perturbations. Before showing examples, in the next section we describe a second procedure aimed at computing the response in a more *direct* way. The latter will then be compared with the fluctuation-response results above.

electrical	mechanical
q_1	x_1
q_2	x_2
$1/R_1$	μ_1 (mobility)
$1/R_2$	μ_2
C_2/X	κ_1 (spring constant)
C_1/X	κ_2
C/X	κ

Table I. Mapping between electric quantities and mechanical ones. Note the inversion of indices for $C_2/X \to \kappa_1$ and $C_1/X \to \kappa_2$.

III. REWEIGHTING

In the analysis via the fluctuation-response relation exposed in the previous section, we deal with experimental data collected in steady states at various temperatures $T = (T_1, T_2)$. Next we show that the same data can be used to extract a form of the susceptibility that is equivalent to Eq. (9). This means that we can bypass once again the step of the actual perturbation of the system in the laboratory.

In the definition (9) what is not useful is that one average is over trajectories under the perturbed $\boldsymbol{\Theta}$, while the other is over unperturbed trajectories. In steady state experiments, trajectories of the former kind are not available. To sidestep this, we find it convenient to consider the alternative formula[1]

$$\chi_{\mathcal{O}}^{\theta}(t) = \lim_{\theta\to 0} \frac{\langle \mathcal{O}(\boldsymbol{x}(t))\rangle_{\boldsymbol{\Theta},\boldsymbol{T}} - \langle \mathcal{O}(\boldsymbol{x}(t))\rangle_{\boldsymbol{\Theta},\boldsymbol{\Theta}}}{-\theta} \ , \qquad (17)$$

because with this form, we can re-express both averages above in terms of steady state averages at $\boldsymbol{T}$, by the following arguments.

First, take the steady state average

$$\langle \mathcal{O}(\boldsymbol{x}(t))\rangle_{\boldsymbol{\Theta},\boldsymbol{\Theta}} = \langle \mathcal{O}(\boldsymbol{x})\rangle_{\boldsymbol{\Theta},\boldsymbol{\Theta}} = \int \mathrm{d}\boldsymbol{x}\, \rho_{\boldsymbol{\Theta}}(\boldsymbol{x})\mathcal{O}(\boldsymbol{x}) \qquad (18)$$

$$= \int \mathrm{d}\boldsymbol{x}\, \rho_{\boldsymbol{T}}(\boldsymbol{x}) \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x})}{\rho_{\boldsymbol{T}}(\boldsymbol{x})}\mathcal{O}(\boldsymbol{x})$$

$$= \left\langle \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x})}{\rho_{\boldsymbol{T}}(\boldsymbol{x})}\mathcal{O}(\boldsymbol{x}) \right\rangle_{\boldsymbol{T},\boldsymbol{T}} \ . \qquad (19)$$

Second, by denoting the probability measure of path $[\boldsymbol{x}]$ under temperatures $\boldsymbol{T}$ by $\mathcal{D}\boldsymbol{x} P_{\boldsymbol{T}}[\boldsymbol{x}]$ [where $P_{\boldsymbol{T}}[\boldsymbol{x}]$ is the path-weight, given that it starts from $\boldsymbol{x}(0)$], take the transient average

$$\langle \mathcal{O}(\boldsymbol{x}(t))\rangle_{\boldsymbol{\Theta},\boldsymbol{T}} = \int \mathcal{D}\boldsymbol{x}\, P_{\boldsymbol{T}}[\boldsymbol{x}] \rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(0)) \mathcal{O}(\boldsymbol{x}(t)) \qquad (20)$$

$$= \int \mathcal{D}\boldsymbol{x}\, P_{\boldsymbol{T}}[\boldsymbol{x}] \rho_{\boldsymbol{T}}(\boldsymbol{x}(0)) \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(0))}{\rho_{\boldsymbol{T}}(\boldsymbol{x}(0))} \mathcal{O}(\boldsymbol{x}(t)) \qquad (21)$$

$$= \left\langle \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(0))}{\rho_{\boldsymbol{T}}(\boldsymbol{x}(0))}\mathcal{O}(\boldsymbol{x}(t)) \right\rangle_{\boldsymbol{T},\boldsymbol{T}} \ . \qquad (22)$$

Thus, by this reweighting via stationary distributions, both averages appearing in Eq. (17) have been reformulated as steady state averages at $\boldsymbol{T}$, and the susceptibility becomes

$$\chi_{\mathcal{O}}^{\theta}(t) = \lim_{\theta\to 0} \frac{-1}{\theta} \left(\left\langle \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(0))}{\rho_{\boldsymbol{T}}(\boldsymbol{x}(0))}\mathcal{O}(\boldsymbol{x}(t)) \right\rangle_{\boldsymbol{T},\boldsymbol{T}} - \left\langle \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x})}{\rho_{\boldsymbol{T}}(\boldsymbol{x})}\mathcal{O}(\boldsymbol{x}) \right\rangle_{\boldsymbol{T},\boldsymbol{T}} \right) . \qquad (23)$$

[1] Eq. (17) is a rewriting of (9) with $\boldsymbol{T}$ and $\boldsymbol{\Theta}$ interchanged, which is equivalent to (9) when the limit of $\boldsymbol{T} \to \boldsymbol{\Theta}$ (i.e. $\theta \to 0$) is taken.

The second single-time average can be written at any instant of time due to time-translation invariance. As such, substituting the particular points $\boldsymbol{x}(0)$ or $\boldsymbol{x}(t)$ one obtains, respectively,

$$\chi_{\mathcal{O}}^{\theta}(t) = \lim_{\theta\to 0} \frac{-1}{\theta} \left\langle [\mathcal{O}(\boldsymbol{x}(t)) - \mathcal{O}(\boldsymbol{x}(0))] \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(0))}{\rho_{\boldsymbol{T}}(\boldsymbol{x}(0))} \right\rangle \qquad (24)$$

or

$$\chi_{\mathcal{O}}^{\theta}(t) = \lim_{\theta\to 0} \frac{1}{\theta} \left\langle \mathcal{O}(\boldsymbol{x}(t)) \left[\frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(t))}{\rho_{\boldsymbol{T}}(\boldsymbol{x}(t))} - \frac{\rho_{\boldsymbol{\Theta}}(\boldsymbol{x}(0))}{\rho_{\boldsymbol{T}}(\boldsymbol{x}(0))} \right] \right\rangle \ . \qquad (25)$$

Again, $\langle \ldots \rangle$ means the steady state average $\langle \ldots \rangle_{\boldsymbol{T},\boldsymbol{T}}$ with the available data. Both formulas can be used to extract the response of the system to a step change of temperature(s) performed at $t = 0$. In our analysis we chose to use Eq. (24).

It is interesting to connect these expressions with previous response relations based on the knowledge of the steady state distribution. One notes that in the limit $\theta \to 0$, the reweighting factor

$$\frac{\rho_{\boldsymbol{\Theta}}}{\rho_{\boldsymbol{T}}} \simeq \frac{\rho_{\boldsymbol{T}} + \theta\, \partial_\theta \rho_{\boldsymbol{\Theta}}}{\rho_{\boldsymbol{T}}} = 1 + \theta\, \partial_\theta \ln \rho_{\boldsymbol{\Theta}} \ . \qquad (26)$$

Substituting this limit, and dropping for simplicity the temperature indices, the second expression (25) for susceptibility above becomes

$$\chi_{\mathcal{O}}(t) = \langle \mathcal{O}(\boldsymbol{x}(t)) \left[\partial_\theta \ln \rho(\boldsymbol{x}(t)) - \partial_\theta \ln \rho(\boldsymbol{x}(0)) \right] \rangle \ , \qquad (27)$$

implying that it comes from a response function [$\chi_{\mathcal{O}}(t) = \int_0^t \mathrm{d}s\, R_{\mathcal{O}}(t-s)$]

$$R(t-s) = \frac{\mathrm{d}}{\mathrm{d}s} \langle \mathcal{O}(\boldsymbol{x}(t)) \partial_\theta \ln \rho(\boldsymbol{x}(s)) \rangle \ . \qquad (28)$$

Equivalently, defining the stochastic entropy $\mathcal{I} = -\ln \rho$,

$$R(t-s) = -\frac{\mathrm{d}}{\mathrm{d}s} \langle \mathcal{O}(\boldsymbol{x}(t)) \partial_\theta \mathcal{I}(\boldsymbol{x}(s)) \rangle \ , \qquad (29)$$

which is Speck and Seifert's response formula [8] for steady states, with the only difference that θ carries a physical dimension while usually the perturbation was expressed in terms of a dimensionless parameter h.

While an analytical expression such as (29) is more elegant than (24) or (25), on the practical side the former may be less convenient. First of all, an analytical expression for the stationary distribution may not be known or calculable, in which case it must be actually measured at two different temperatures and the θ derivative will have to be performed discretely, which is equivalent to using the expressions prior to Eq. (26). Secondly, even if an analytical expression for the stationary distribution is available (as it is for the present system of interest; details in the appendix), its θ derivative might be too unwieldy to work with, from an implementation point of view. A discrete approximation for the derivative, such as (24), is simpler to handle. We have indeed followed this path, using analytical expressions for the distributions $\rho_{\boldsymbol{\Theta}}$ and $\rho_{\boldsymbol{T}}$, choosing $\boldsymbol{\Theta} = (T_1 + \theta, T_2)$ with $\theta = T_1/100$.

IV. NONEQUILIBRIUM HEAT CAPACITY

In this section we show the analysis of experimental data, which show that the fluctuation-response relation $\chi_{\mathcal{O}}(t) = S_1 + S_2 + K_1 + K_2$ with terms listed in (16), can reliably compute the susceptibility of the system. The knowledge of the steady state distribution allows us to use the formula introduced in (24), and to compute the response independently. The two versions turn out to yield results in good agreement with each other.

The averages used to compute susceptibilities are performed over trajectories that extend over ≈ 50 ms, with time steps of length $\Delta t = 1/8192\,\mathrm{s} \approx 0.122\,\mathrm{ms}$. Each trajectory is extracted by choosing a different starting point from the steady state sampling. Three cases are considered, one in the equilibrium condition $T_1 = T_2 = 296$ K ($\approx 7 \times 10^7$ trajectories) and two far from equilibrium, $T_1 = 140$ K ($\approx 2.6 \times 10^7$ trajectories) and $T_1 = 88$ K ($\approx 4.6 \times 10^7$ trajectories).

As an observable $\mathcal{O}(\boldsymbol{x})$ – where we recall that $\boldsymbol{x} = (x_1, x_2) = (q_1, q_2)$ – we consider the total electrostatic energy of the system, Eq. (6), in accord with the mapping of Table I between electrical and mechanical quantities. The backward generator acting on this observable, $\mathbb{L}\mathcal{O}$ appearing in (16d), becomes

$$\begin{aligned}\mathbb{L}U(\boldsymbol{x}) =& \kappa_1\mu_1(F_1(\boldsymbol{x})x_1 + T_1) + \kappa_2\mu_2(F_2(\boldsymbol{x})x_2 + T_2)\\ &+ \kappa[\mu_1(F_1(\boldsymbol{x})(x_1 - x_2) + T_1)\\ &+ \mu_2(F_2(\boldsymbol{x})(x_2 - x_1) + T_2)]\end{aligned} \tag{30}$$

where we remind that $k_B = 1$ and temperatures have dimensions of energy. The response of the energy to a change of temperature becomes the nonequilibrium version of the heat capacity if $T_1 \neq T_2$ (a different definition of heat capacity for nonequilibrium systems can be found in Ref. [19]). The following analysis confirms that in general this heat capacity cannot be computed only from the correlation between energy and heat flowing into the system [19, 20, 24–26], unless this is in equilibrium.

The susceptibility χ_U of the internal energy to a change of T_1 is shown in Fig. 2 as a function of time for the three values of T_1. It correctly converges to a constant value for large times, though its single terms may be extensive in time in nonequilibrium conditions. We have also an analytical argument predicting that such constant value should be $1/2$. It is based on recently proposed mesoscopic virial equations [38]. For each degree of freedom i in an overdamped system subject to multiple reservoirs we have

$$-\langle x_i F_i(\boldsymbol{x})\rangle = T_i\,. \tag{31}$$

In our system with quadratic potential energy this implies $\langle U(\boldsymbol{x})\rangle = T_1/2 + T_2/2$ in a steady state. Therefore, it is expected that the susceptibility $\chi_U(t) = \partial\langle U\rangle/\partial T_1 \to 1/2$ as $t \to \infty$. This is indeed observed in the top panel of Fig. 2, where the steady state is an equilibrium state, with $T_1 = T_2$. The asymptotic value of $1/2$ for the susceptibility is also fairly well reached by the data in the lower panels of Fig. 2; a possible explanation of the slight disagreement is given in the next paragraph. In equilibrium (top panel), K_1 and S_2 vanish while K_2 is equal to S_1, that is, the response is given by twice S_1. This is essentially the Kubo formula, stating that in equilibrium the response of an observable is given by its correlations with the entropy produced in the environment (heat flow divided by reservoir temperature), which is confirmed by the form (16a) of S_1. (The extra factor of $1/T_1$ in Eq. (16) has to

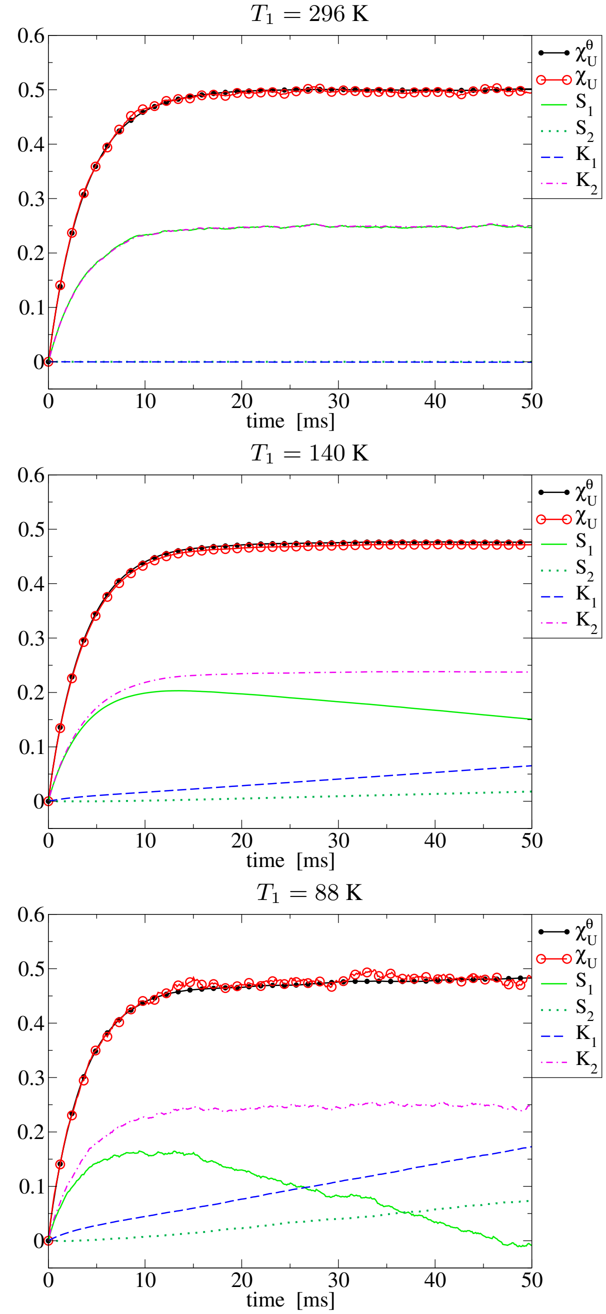

Figure 2. (Color online) Response of the total energy U to a change of T_1, for equilibrium ($T_1 = 296$ K $= T_2$) and for nonequilibrium ($T_1 = 140$ K and $T_1 = 88$ K). The susceptibility χ_U computed with the fluctuations-response relation (10) and its terms S_1, S_2, K_1, K_2 are shown. The susceptibility χ_U^θ computed with the reweighting formula (24) agrees with χ_U.

do with the units of susceptibility.) On the other hand, out of equilibrium the equality between S_1 and K_2 is lost in addition to K_1 and S_2 no longer vanishing, as demonstrated by the two bottom panels of Fig. 2: All the terms S_1, S_2, K_1, and K_2 of Eq. (16) composing $\chi_{\mathcal{O}}$ are all relevant. The correlation S_1 between the observable and the heat flow is not sufficient anymore in nonequilibrium systems. The frenetic terms K_1, K_2 and the new entropy production term S_2, are also relevant for predicting the nonequilibrium response.

While the susceptibility at equilibrium ($T_1 = T_2$) attains the expected asymptotic value of $1/2$ fairly closely, the susceptibility out of equilibrium ($T_1 \neq T_2$) seems to fall a bit short. We argue that this has to do with the inevitable limitation on the time resolution of the trajectory measurements, since numerical simulations of an equivalent system also exhibit the same feature when the time discretization becomes coarse. Indeed, the sampling interval in the experiments (≈ 0.1 ms) is not *much* smaller than the dynamical time scale $Y = 13$ms in the circuit, which one can confirm visually from the plots in Fig. 2. The reason why it is the nonequilibrium susceptibilities which suffered more from this quantization error is likely as follows: Out of equilibrium, trajectory functionals like entropy production are numerically larger than in equilibrium, amplifying any error in the trajectory.

In all examples we also plot the susceptibility χ_U^θ computed with (24). Clearly there is a very good agreement between this estimate and χ_U for all times, including the deviation from the asymptotic value $1/2$ for large times. This suggests that both approaches work well and corroborates our explanation of the slight offset in the asymptotic value, as also χ_U^θ should be affected by the time-step discretization.

CONCLUSION

We have shown that experimental steady state data can be used to predict the thermal linear response of an electric circuit, even if it works in a thermally unbalanced nonequilibrium regime due to a cryogenic bath applied to one of the two resistors. We have used a recent nonequilibrium response relation for our analysis. This approach requires the knowledge of the forces acting on each degree of freedom, an information easily available in our case. The nonequilibrium version of the heat capacity provides a simple demonstration of the fact that in general one cannot expect to predict the response of the energy to thermal variations just from the unperturbed correlations between energy and fluctuating heat flows, as one would do by using the standard fluctuation-dissipation theorem for equilibrium systems. Also non-dissipative aspects play a crucial role: The response includes correlations between the observable and the so-called frenesy of the system [3, 4], which is a measure of how frantically the system wanders about in phase space. Our example of generalised heat capacity, in terms of the response of the total energy of the system, is relevant in the context of steady state thermodynamics [27–31]. Other definitions of heat capacity are possible, for example in terms of excess heat flow with respect to the housekeeping heat flow [19].

In order to have a comparison with an independent method for computing the susceptibility, we also introduced a reweighting procedure that has the advantage of needing no more than the same steady state data. The second method estimates the susceptibility of the system in a more direct sense, namely mimicking actual finite perturbations of the system. This procedure is simple to implement and is related to a linear response formula also based on the knowledge of the steady state distribution.

Appendix A: Gaussian steady states distributions

We review the procedure used to obtain the steady state distribution for linear overdamped stochastic systems with additive noise.

Consider a process given by the stochastic differential equation

$$\dot{\boldsymbol{x}} = -A\boldsymbol{x} + \sqrt{2D}\boldsymbol{\xi} \;, \tag{A1}$$

with $\boldsymbol{\xi}$ being N-dimensional uncorrelated noise and $\boldsymbol{x}$ the N-dimensional state. Here, A and D are $N \times N$ positive-definite constant matrices. The ensemble current corresponding to these degrees of freedom follows as

$$\boldsymbol{J} = -\rho A\boldsymbol{x} - D\boldsymbol{\nabla}\rho \;, \tag{A2}$$

with the Fokker-Planck equation $\partial_t \rho + \boldsymbol{\nabla} \cdot \boldsymbol{J} = 0$. Thus, stationarity implies (index notation hereafter)

$$\begin{aligned} 0 =& -\partial_i J_i && \text{(A3)}\\ =&\partial_i(\rho A_{ij}x_j) + D_{ij}\partial_i\partial_j\rho && \text{(A4)}\\ =&\rho A_{ii} + A_{ij}x_j\partial_i\rho + D_{ij}\partial_i\partial_j\rho \;. && \text{(A5)}\end{aligned}$$

Clearly an exponential quadratic form would satisfy this equation and the ansatz

$$\rho(\boldsymbol{x}) = \sqrt{\frac{\det G}{(2\pi)^N}}\, e^{-\frac{1}{2}x_i G_{ij} x_j} \;, \tag{A6}$$

with G positive-definite, yields

$$\begin{aligned} 0 =&A_{ii} - A_{ij}x_j G_{ik}x_k + D_{ij}(G_{ik}x_k G_{jl}x_l - G_{ij}) && \text{(A7)}\\ =&A_{ii} - D_{ij}G_{ij} - x_k x_l(A_{il}G_{ik} - G_{ik}D_{ij}G_{jl}) \;. && \text{(A8)}\end{aligned}$$

Using the symmetry of G_{ij} and matrix notation, this can be rewritten as

$$\mathrm{Tr}(A - DG) = \boldsymbol{x}^\dagger(GA - GDG)\boldsymbol{x} \;. \tag{A9}$$

Since this is supposed to hold for any $\boldsymbol{x}$, both sides must vanish. For the right-hand side, this implies that the matrix $GA - GDG$ is skew-symmetric, which means that it has vanishing symmetric part,

$$GA + A^\dagger G = 2GDG \;, \tag{A10}$$

or, equivalently,

$$AG^{-1} + G^{-1}A^\dagger = 2D \;. \tag{A11}$$

The left-hand side of (A9) also vanishes, as required, when a G_{ij} satisfying (A11) is found.

Being a linear equation in the unknown entries of G^{-1}, one can imagine rewriting (A11) so as to treat those unknowns as a vector (likewise the right-hand side), and afterwards inverting the matrix equation. This is achieved by resorting to the Kronecker product, denoted by $\otimes$, and a "vectorization" operation, denoted as "vec", which amounts to stacking the columns of a matrix into a single column. Eq. (A11) is recast in the form

$$(I \otimes A + A \otimes I)\,\mathrm{vec}\,G^{-1} = 2\,\mathrm{vec}\,D\;. \tag{A12}$$

Hence we find G^{-1} via

$$\mathrm{vec}\,G^{-1} = 2(I \otimes A + A \otimes I)^{-1}\,\mathrm{vec}\,D \tag{A13}$$

followed by an "un-vec", *i.e.* a procedure reverting back from vectorized matrices to actual ones.

Circuit experiments

In the electric circuit experiments, the equation of motion for the charges is of the form

$$\dot{\boldsymbol{q}} = -R^{-1}\mathcal{C}^{-1}\boldsymbol{q} + R^{-1}\sqrt{2RT}\boldsymbol{\xi}\;, \tag{A14}$$

where

$$\mathcal{C} = \begin{bmatrix} C + C_1 & C \\ C & C + C_2 \end{bmatrix} \tag{A15}$$

$$\mathcal{C}^{-1} = \frac{1}{X}\begin{bmatrix} C + C_2 & -C \\ -C & C + C_1 \end{bmatrix} \tag{A16}$$

$$R = \begin{bmatrix} R_1 & 0 \\ 0 & R_2 \end{bmatrix} \tag{A17}$$

$$T = \begin{bmatrix} T_1 & 0 \\ 0 & T_2 \end{bmatrix} \tag{A18}$$

[$X = \det\mathcal{C}$ was defined in (4)]. Thus, we identify $A = R^{-1}\mathcal{C}^{-1}$ and $D = R^{-1}T$. Through (A13) and inverting the resulting matrix G, we have

$$G = \frac{Y}{Z}\begin{bmatrix} g_{11} & g_{12} \\ g_{21} & g_{22} \end{bmatrix} \tag{A19}$$

with

$$\begin{aligned} Y =& R_1R_2(\det\mathcal{C})(\mathrm{Tr}\,A) \\ =& (C + C_1)R_1 + (C + C_2)R_2 \end{aligned} \tag{A20}$$

$$Z = X[Y^2T_1T_2 + R_1R_2C^2(T_1 - T_2)^2] \tag{A21}$$

and

$$g_{11} = T_2Y(C + C_2) + (T_1 - T_2)R_1C^2 \tag{A22}$$

$$g_{12} = -(C + C_1)CR_1T_1 - (C + C_2)CR_2T_2 \tag{A23}$$

$$g_{21} = g_{12} \tag{A24}$$

$$g_{22} = T_1Y(C + C_1) + (T_2 - T_1)R_2C^2 \tag{A25}$$

We have thus all the elements for computing the steady state distribution (A6) analytically at any combination of temperatures T_1, T_2.

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