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We consider steady state heat conduction across a quantum harmonic chain connected to reservoirs modelled by infinite collection of oscillators. The heat, Q , flowing across the oscillator in a time interval τ is a stochastic variable and we study the probability distribution function $P(Q)$. We compute the exact generating function of Q at large τ and the large deviation function. The generating function has a symmetry satisfying the steady state fluctuation theorem regardless of the specifics of the system. The distribution $P(Q)$ is nongaussian with clear exponential tails. The effect of finite τ and nonlinearity is considered in the classical limit through Langevin simulations. We also obtain predictions of heat current fluctuations at low temperatures in clean wires.

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A lot of interest has been generated recently in fluctuations in entropy production in nonequilibrium systems. Several definitions of entropy production have been used and these give some measure of “second law violations”. A number of authors have looked, both theoretically [1–4] and in experiments [5, 6], at fluctuations of quantities such as work, power flux, heat absorbed, etc. during nonequilibrium processes and these have been generically referred to as entropy production. The new results, referred to as the fluctuation theorems, make general predictions on the probability distribution $P(S)$ of the entropy S produced during a nonequilibrium process [1, 2]. Specifically these theorems quantify the probability of negative entropy producing events which become significant if one is looking at small systems or at small time intervals. There are two different theorems, the transient fluctuation theorem (TFT) and the steady state fluctuation theorem (SSFT). The TFT looks at the entropy produced in a finite time τ in a non-steady state. In SSFT, one looks at a nonequilibrium steady state and the average entropy production rate over a long time interval τ is examined. The precise statement of SSFT is:

$$\lim_{\tau \rightarrow \infty} \frac{1}{\tau} \ln \left[\frac{P(S = \sigma\tau)}{P(S = -\sigma\tau)} \right] = \sigma. \quad (1)$$

In the context of SSFT a quantity of great interest is the large deviation function $h(\sigma)$ which specifies the asymptotic form of the distribution function $P(S)$ through the relation $P(S) \sim e^{\tau h(\sigma)}$ [2, 7]. An equivalent statement of SSFT can be made in terms of a special symmetry of $h(\sigma)$ which is: $h(\sigma) - h(-\sigma) = \sigma$. Remarkably, this relation has been shown to lead to linear response results such as Onsager reciprocity and the Green-Kubo relations [2, 8, 9]. Furthermore it leads to predictions for properties in the far from equilibrium regime.

Heat conduction is a natural example where one talks of entropy production. The standard result from nonequilibrium thermodynamics is that when an amount

of heat Q is transferred from a bath at temperature T_L to a bath at temperature T_R ($< T_L$) the entropy produced S is given by $S = (T_R^{-1} - T_L^{-1}) Q$. However, in general S is a stochastic variable with a distribution $P(S)$. The distribution $P(S)$ for a nonlinear chain connected to Nose-Hoover baths was studied numerically in [10] where they verified that it satisfied SSFT. Refn. [11] studied heat conduction in a nonlinear chain connected to free phonon reservoirs. Based on strong ergodicity properties of the model, it was proved that $P(S) \sim e^{\tau h(\sigma)}$ where $h(\sigma)$ satisfied the SSFT symmetry. A transient version of the heat exchange fluctuation theorem, valid both for classical and quantum systems, was proved in [12].

While the SSFT clearly presents a powerful theorem for nonequilibrium systems, its validity has been established only in specific systems and so far only classically. This letter presents the first explicit calculation of $h(\sigma)$ and demonstration of SSFT in heat conduction in a quantum system. In this letter we study steady state of a harmonic chain connected to baths which are modelled by infinite oscillator sets. We use the method of full-counting statistics [13] to compute the generating function of Q in the limit of large τ . We then show that the corresponding large deviation function satisfies the SSFT symmetry condition. For finite τ we consider heat transport across small chains and study the classical limit through Langevin simulations. We also consider the effect of introducing nonlinearity in the oscillator potential.

We note that independent of the relation to the FTs the study of nonequilibrium fluctuations is of interest especially in the context of transport in nanosystems where such fluctuations are expected to be large. Indeed, a number of recent papers have studied the problem of fluctuations in charge current in mesoscopic systems, both theoretically [13–16] and experimentally [17], and many aspects in higher order fluctuations have been clarified. On the other hand, as regards thermal transport in dielectric wires, nonequilibrium fluctuations have not been

discussed so far even though experimental measurements of heat conduction in nanowires has now become possible [18, 19]. At very low temperatures, the quantized thermal conductance $g_0(T) = \pi k_B^2 T / (6\hbar)$ was predicted [20] and was measured in experiments [19]. Our analysis provides a theoretical basis to study heat current fluctuations in the mesoscopic regime.

Model. — Our model consists of a harmonic chain coupled to two heat baths kept at temperatures T_L and T_R respectively. For the heat baths we assume the standard model of an infinite collection of oscillators. The full Hamiltonian is given by

$$\begin{aligned} \mathcal{H} = & \sum_{n=1}^N \left(\frac{p_n^2}{2m_n} + \frac{k_n}{2} x_n^2 \right) + \sum_{n=2}^N \frac{k}{2} (x_n - x_{n-1})^2 \\ & + \sum_{\ell} \left[\frac{p_{\ell}^2}{2m_{\ell}} + \frac{m_{\ell}\omega_{\ell}^2 x_{\ell}^2}{2} - \lambda_{\ell} x_{\ell} x_1 + \frac{\lambda_{\ell}^2 x_1^2}{2m_{\ell}\omega_{\ell}^2} \right] \\ & + \sum_r \left[\frac{p_r^2 x_r^2}{2m_r} + \frac{m_r\omega_r^2 x_r^2}{2} - \lambda_r x_r x_N + \frac{\lambda_r^2 x_N^2}{2m_r\omega_r^2} \right], \end{aligned} \quad (2)$$

where $\{m_n, x_n, p_n, k_n, k\}$ refer to the system degrees of freedom, $\{x_{\ell}, p_{\ell}, m_{\ell}, \omega_{\ell}\}$ refers to the left reservoir while $\{x_r, p_r, m_r, \omega_r\}$ refers to the right reservoir. The coupling constants between the system and the bath oscillators $\{\lambda_{\ell}, \lambda_r\}$ is switched on at time $t = -\infty$. The initial density matrix is assumed to be of the product form $\rho(-\infty) = \rho_S \otimes \rho_L \otimes \rho_R$, where S, L, R refer respectively to the system and left and right reservoirs. The left and right density matrices are equilibrium distributions corresponding to the respective temperatures: $\rho_{\alpha} = e^{-\beta_{\alpha} \mathcal{H}_{\alpha}} / \text{Tr}[e^{-\beta_{\alpha} \mathcal{H}_{\alpha}}]$ for $\alpha = L, R$ and $\beta_{\alpha} = 1/(k_B T_{\alpha})$. It can be shown [21] that eliminating the bath degrees of freedom leads to an effective quantum Langevin equation for the system given by:

$$\begin{aligned} m_1 \ddot{x}_1 &= -k_1 x_1 - k(x_1 - x_2) \\ & - \int_{-\infty}^t dt' \gamma_L(t-t') \dot{x}_1(t') + \eta_L(t) \\ m_n \ddot{x}_n &= -k_n x_n - k(2x_n - x_{n-1} - x_{n+1}) \quad 1 < n < N \\ m_N \ddot{x}_N &= -k_N x_N - k(x_N - x_{N-1}) \\ & - \int_{-\infty}^t dt' \gamma_R(t-t') \dot{x}_N(t') + \eta_R(t). \end{aligned} \quad (3)$$

The effect of the baths is to produce noise, given by $\eta_{L,R}$, and dissipative effects controlled by the memory kernels $\gamma_{L,R}$. The properties of the noise and dissipation are completely determined by the initial condition of the baths at $t = -\infty$. We now make a few definitions. Let $\tilde{\gamma}_{L,R}(\omega) = \int_0^{\infty} dt \gamma_{L,R}(t) e^{i\omega t}$ and let $\Sigma_{L,R}^r(\omega) = i\omega \tilde{\gamma}_{L,R}(\omega)$, which, as we will see later, gives the self energy correction coming from the baths to the Green's function of the harmonic chain. We also define the spectral function $J_L(\omega) = \frac{\pi}{2} \sum_{\ell} \frac{\lambda_{\ell}^2}{m_{\ell}\omega_{\ell}} \delta(\omega - \omega_{\ell})$ for the left reservoir and a similar function $J_R(\omega)$ for the

right reservoir. Then the dissipation kernels and noise correlations are given by:

$$\begin{aligned} \gamma_{\alpha}(t) &= \frac{2}{\pi} \int_0^{\infty} d\omega \frac{J_{\alpha}(\omega)}{\omega} \cos \omega t \\ \langle \eta_{\alpha}(\omega) \eta_{\alpha}(\omega') \rangle &= 4\pi \hbar \delta(\omega + \omega') \Gamma_{\alpha}(\omega) (1 + f_{\alpha}(\omega)) \end{aligned} \quad (4)$$

for $\alpha = L, R$ and where $\Gamma_{\alpha}(\omega) = \text{Im}\{\Sigma_{\alpha}^r(\omega)\} = J_{\alpha}(\omega)\Theta(\omega) - J_{\alpha}(-\omega)\Theta(-\omega)$ and $f_{\alpha}(\omega) = 1/(e^{\beta_{\alpha}\hbar\omega} - 1)$. All higher noise correlations can be obtained from the two-point correlator. Using the quantum Langevin approach it is straightforward to derive the Landauer type result for average heat current $\langle \hat{\mathcal{I}} \rangle$ [21]:

$$\langle \hat{\mathcal{I}} \rangle = \frac{1}{4\pi} \int_{-\infty}^{\infty} d\omega \hbar \omega \mathcal{T}(\omega) [f_L(\omega) - f_R(\omega)], \quad (5)$$

$$\begin{aligned} \mathcal{T}(\omega) &= 4\Gamma_L(\omega)\Gamma_R(\omega) |G_{1,N}^r(\omega)|^2, \\ G^r(\omega) &= [-\mathbf{M}\omega^2 + \mathbf{K} - \Sigma_L^r(\omega) - \Sigma_R^r(\omega)]^{-1}, \end{aligned} \quad (6)$$

where \mathbf{M}, \mathbf{K} are the mass and the force constant matrix and $\Sigma_{L,R}^r(\omega)$ are self-energy correction matrices with elements $[\Sigma_L^r(\omega)]_{m,n} = \Sigma_L^r(\omega) \delta_{m,n} \delta_{m,1}$ and $[\Sigma_R^r(\omega)]_{m,n} = \Sigma_R^r(\omega) \delta_{m,n} \delta_{m,N}$. Note that $\mathcal{T}(\omega)$ is the transmission coefficient for phonons while $G^r(\omega)$ is the phonon Green's function for the chain.

Statistics of phononic heat transfer. — The heat transfer in time τ is given by $\langle \hat{\mathcal{I}} \rangle \tau$. Here we are interested in the statistics of heat transfer in the nonequilibrium steady state and so need to calculate higher moments of the heat transfer. For this it is necessary to calculate higher order correlations such as $\langle \hat{\mathcal{I}}(t_1) \hat{\mathcal{I}}(t_2) \rangle$, etc. The quantum Langevin approach can in principle be used to compute correlation functions at any order but this becomes increasingly cumbersome. Instead we will use the Keldysh approach which, as we will show, gives the full generating function of the heat transfer.

Several definitions of $\hat{\mathcal{I}}$ are possible depending on where we evaluate the current. Here we consider the current from the left reservoir into the system (obtained by taking a time-derivative of energy in the left reservoir $\mathcal{H}_L = \sum_{\ell} [p_{\ell}^2/(2m_{\ell}) + m_{\ell}\omega_{\ell}^2 x_{\ell}^2/2]$):

$$\hat{\mathcal{I}} = - \sum_{\ell} \frac{\lambda_{\ell}}{m_{\ell}} p_{\ell} x_1. \quad (7)$$

We also define the average heat transfer operator $\hat{\mathcal{Q}} = \int_{-\tau/2}^{\tau/2} dt \hat{\mathcal{I}}(t)$. In order to get the generating function of current, we introduce the modified Hamiltonian $\mathcal{H}_{\pm}(t)$, where only interaction part in (2) is modified as

$$\lambda_{\ell} x_{\ell} x_1 \rightarrow \lambda_{\ell} \left[x_{\ell} \cos(\omega_{\ell} \varphi_{\pm}(t)) + \frac{p_{\ell}}{m_{\ell} \omega_{\ell}} \sin(\omega_{\ell} \varphi_{\pm}(t)) \right] x_1.$$

Here the counting field $\varphi_{\pm}(t)$ is defined as $\varphi_{\pm}(t) = \mp(\hbar\xi)/2$ for $-\tau/2 \leq t \leq \tau/2$ and zero elsewhere. Let us compute the quantity:

$$Z(\xi) = \left\langle \overleftarrow{\mathbb{T}} e^{\frac{i}{\hbar} \int_{-\infty}^{\infty} dt \mathcal{H}_{-}(t)} \overrightarrow{\mathbb{T}} e^{-\frac{i}{\hbar} \int_{-\infty}^{\infty} dt \mathcal{H}_{+}(t)} \right\rangle,$$

where $\langle \dots \rangle$ denotes an average over the initial state, \overleftarrow{T} and \overrightarrow{T} denote forward and reverse time ordering. It can be shown that $\ln Z(\xi)$ is the cumulant generating function for the heat operator:

$$\ln Z(\xi) = \sum_{n=1}^{\infty} \frac{(i\xi)^n}{n!} \langle \hat{Q}^n \rangle_c, \quad (8)$$

where $\langle \hat{Q}^n \rangle_c$ is the n^{th} order cumulant at large τ . Hence the probability distribution of measuring a heat transfer Q is obtained by taking the Fourier transform $P(Q) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\xi Z(\xi) e^{-i\xi Q}$. For large τ one obtains $Z(\xi) \sim e^{\tau \mathcal{G}(\xi)}$ and $P(Q) \sim e^{\tau \tilde{h}(q)}$ with $\tilde{h}(q) = \mathcal{G}(\xi^*) - i\xi^* q$ and where ξ^* is the solution of the saddle-point equation $d\mathcal{G}(\xi^*)/d\xi^* - iq = 0$. We evaluate $\mathcal{G}(\xi)$ using standard path integral and Green's function techniques along the Keldysh contour. The final result is the following form:

$$\begin{aligned} \mathcal{G}(\xi) = & \frac{-1}{4\pi} \int_{-\infty}^{\infty} d\omega \ln \left\{ 1 + \mathcal{T}(\omega) [f_R(-\omega) f_L(\omega) (e^{i\xi \hbar \omega} - 1) \right. \\ & \left. + f_R(\omega) f_L(-\omega) (e^{-i\xi \hbar \omega} - 1)] \right\}. \end{aligned} \quad (9)$$

Phonons convey energy in units $\hbar\omega$ and this appears in the exponential form with the factor ξ . It is easily verified that Eq. (9) reproduces the correct first moment of $\hat{\mathcal{I}}$ given in Eq. (5). The second moment is given by

$$\begin{aligned} \frac{\langle \hat{Q}^2 \rangle_c}{\tau} = & \frac{1}{4\pi} \int_{-\infty}^{\infty} d\omega (\hbar\omega)^2 \left\{ \mathcal{T}^2(\omega) [f_L(\omega) - f_R(\omega)]^2 \right. \\ & \left. - \mathcal{T}(\omega) [f_L(\omega) f_R(-\omega) + f_L(-\omega) f_R(\omega)] \right\}. \end{aligned} \quad (10)$$

We have verified this also with the Langevin approach. This bosonic fluctuation is similar to the optical one [14].

Symmetry.— We note the following symmetry of \mathcal{G} :

$$\mathcal{G}(\xi) = \mathcal{G}(-\xi + i\mathcal{A}), \quad (11)$$

where $\mathcal{A} = \beta_R - \beta_L$. Using the identification $\sigma = Aq$ and the relation between $h(\sigma) = \tilde{h}(q)$ and $\mathcal{G}(\xi)$ immediately leads to the SSFT relation Eq. (1).

The symmetry (11) contains information regarding transport coefficients [9]. For fixed $\beta_L + \beta_R$ let us make the expansion $\langle \hat{Q}^n \rangle_c / \tau = \sum_m L_{n,m} \mathcal{A}^m / m!$. The nonlinear response coefficients $L_{n,m}$ are then given by $L_{n,m} = \partial^{n+m} \mathcal{G}(\xi) / \partial (i\xi)^n \partial \mathcal{A}^m |_{\xi=\mathcal{A}=0}$. The symmetry (11) gives the general relations between the coefficients:

$$L_{n,m} = \sum_{k=0}^m \binom{m}{k} (-1)^{(n+k)} L_{n+k, m-k}, \quad (12)$$

with $L_{0,m} = 0$. For example we get $L_{2,0} = 2L_{1,1}$ and $L_{4,0} = 2L_{3,1} = 6L_{2,2} - 4L_{1,3}$. The first relation is simply the Green-Kubo formula relating the linear current response to equilibrium fluctuations while the second leads to relations between nonlinear response coefficients.

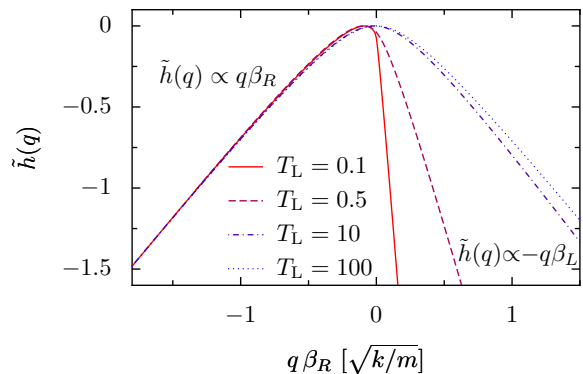


FIG. 1: Plot of $\tilde{h}(q)$ for various temperature regime with $T_R - T_L$ fixed to 1.0 [$\hbar k_B^{-1} \sqrt{k/m}$]. The parameters: $N = 2$, $m_1 = m_2 = m$, $k_1 = k_2 = k$, and $\gamma = 1.0$ [\sqrt{mk}].

Typical distributions.— We present some results on the form of the distribution $P(Q) \sim e^{\tilde{h}(q)}$ for a small chain ($N = 2$) connected to ohmic reservoirs ($\tilde{\gamma}_{L,R}(\omega) = \gamma$). In Fig. 1 we plot $\tilde{h}(q)$, which is numerically obtained, for different temperatures T_L with fixed temperature difference $T_R - T_L$. In all temperature regimes, $\tilde{h}(q)$ shows a clear linear dependence at large q , and those are well fitted by $\beta_R q$ and $-\beta_L q$ for $q < 0$ and $q > 0$ respectively. This exponential tail is one of the characteristics in $P(Q)$.

We now study the effects of a finite τ and nonlinear potential using the classical system. We evaluate $P(Q)$ from direct simulations of the Langevin equations Eq. (3) with white noise. In Fig. 2 we compare the simulation results for different values of τ with the asymptotic function $\tilde{h}(q)$ (obtained for $\hbar \rightarrow 0$). It is clear from Fig. 2 that convergence to the asymptotic distribution function takes place on a rather large time scale. The nonlinear case is also plotted for the same system with an onsite potential $V(x_n) = \alpha x_n^4 / 4$. In the inset, the function $\ln[P(Q)/P(-Q)]/\tau$ is plotted for three cases. The distribution for the nonlinear case deviates from the harmonic cases, and both the average heat current and its fluctuations are suppressed. However, as the inset shows, SSFT is satisfied in the nonlinear case, which indicates the symmetry (11) and the relation (12) hold too.

Heat current fluctuations in a pure wire.— Using Eq.(9), we can derive the heat current fluctuations for a homogeneous wire connected to reservoirs through non-reflecting contacts, a case for which the quantized thermal conductance has been measured [19]. Consider a pure wire with all masses and spring constants equal. If we consider that the heat reservoirs themselves are semi-infinite wires (*i.e.*, the Rubin model of a heat bath) then it is easy to show that the contacts are perfect and we get $\mathcal{T}(\omega) = 1$ for all ω within the allowed bandwidth. At low temperatures and for small $\Delta T = T_L - T_R$, Eq. (5) leads to the quantized heat conductance $g_0(T) = \langle \hat{\mathcal{I}} \rangle / \Delta T = \pi k_B^2 T / (6\hbar)$. From Eq. (10) we now also get the thermal

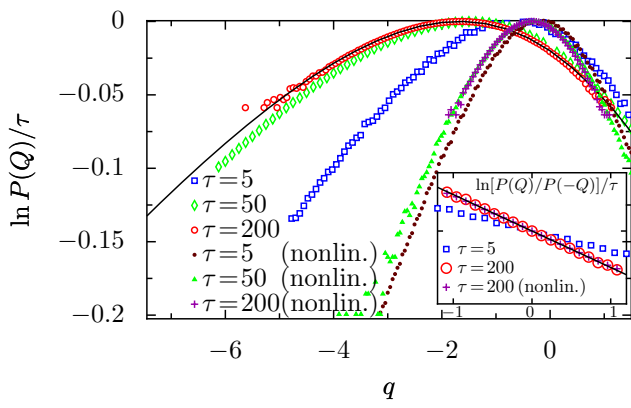


FIG. 2: Plot of $\ln[P(Q)]/\tau$ from Langevin simulations for the same system as in Fig. 1 for various τ . Here $P(Q)$ is normalized so that its maximum is one. The large deviation function $\hat{h}(q)$ is also shown by black solid line. The parameters are $N = 2, m = k = 1, k_B T_L = 10$, and $k_B T_R = 20$. The nonlinear case has $\alpha = 2$. The inset shows $\ln[P(Q)/P(-Q)]/\tau$, and SSFT line $q(\beta_R - \beta_L)$ (black solid line). Heat transfer is measured at the contact to the left reservoir.

noise power at zero frequency $S_0 = \langle \hat{Q}^2 \rangle / \tau$:

$$S_0 = k_B T_L^2 g_0(T_L) + k_B T_R^2 g_0(T_R). \quad (13)$$

This is valid for $T_{L,R}$ in the temperature regime where $g_0(T)$ can be measured [19]. The noise power is also independent of details of system. Independent contributions from T_L and T_R are obtained since there are no scattering process between phonons. Eq. (9) with $\mathcal{T}(\omega) = 1$ gives us the generating function valid in the same regime. While our results have been derived for a one-dimensional wire with scalar displacement variables they are easy to generalize. Similar results can be obtained for realistic models [22] of nanowires and nanotubes.

Summary.— We have derived the explicit distribution for fluctuations in phononic heat transfer across a harmonic chain. We then obtained the first proof of SSFT in quantum heat conduction. We have also made predictions of heat current fluctuations for a clean wire. The measurement of fluctuations of phononic heat transfer in experiments is an important challenging problem. A modification of the set up used in [19] should be able to make a measurement of fluctuations in heat transfer. One possibility is to use heaters with some feed-back mechanism so as to maintain the two reservoirs at fixed temperatures. The fluctuations in the power from the heater would be related to the fluctuations in the heat transfer through the wire.

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