

Nonequilibrium field theory with a temperature gradient: Thermal current in a nanowireYuan Gao and K. A. Muttalib *Department of Physics, University of Florida, P.O. Box 118440, Gainesville, Florida 32611-8440, USA* (Received 28 September 2023; revised 12 December 2023; accepted 26 March 2024; published 8 April 2024)

A perturbative framework is developed within the standard nonequilibrium field theory techniques to incorporate a temperature gradient across a thermoelectric device. The framework uses a temperature-dependent pseudo-Hamiltonian generated from the exact density matrix in the presence of nonuniform temperatures. We develop a perturbation theory for small temperature gradients in long wires and obtain the nonlinear thermal conductance as a function of temperature difference. The framework should be adaptable to more general cases of temperature inhomogeneities in either fermionic or bosonic systems.

DOI: [10.1103/PhysRevB.109.155409](https://doi.org/10.1103/PhysRevB.109.155409)**I. INTRODUCTION**

Thermoelectric current is generated by converting heat to electrical energy [1]. A typical thermoelectric device is a good electrical conductor with its two ends kept at two different fixed temperatures, generating a difference in chemical potential between the two ends [2–4]. However, the device also needs to be a poor thermal conductor in order to have a large efficiency. It has been proposed [5] that improved efficiency and power output in thermoelectric devices require two independent considerations: taking advantage of an interplay between the material and the thermodynamic parameters available to increase the charge transport in the nonlinear regime [6,7], and simultaneously decreasing the heat current by using surface-disordered nanowires [8–15]. Because of the temperature difference between its two ends, there exists a temperature gradient across the nanowire. In the linear-response regime, the temperature difference is assumed to be small and one can treat the entire device at one fixed average temperature. In this case one can use the standard nonequilibrium field theory (NEFT) techniques [16,17], valid for a thermally homogeneous system, to evaluate both the electrical and the thermal current in the device. In the nonlinear regime mentioned above, the temperature difference between the ends can be large and the temperature of the wire is different at different distances from the two ends. The standard NEFT techniques break down in such cases; studying charge or heat currents requires a generalization of the currently available NEFT methods to incorporate thermal inhomogeneity.

In this paper we generalize the existing NEFT techniques in the presence of a temperature gradient and evaluate the heat current across a thermoelectric device. For simplicity we consider a uniform wire of length $L = Na$, a being the lattice spacing and the number of sites $N \gg 1$. One end of the wire is kept at a fixed hot temperature T_H and the other end at a colder temperature T_C . We use a perturbation theory valid for small temperature gradients. The framework divides the system into slices of subsystems at different temperatures connected together and allows us to describe the nonequilibrium Green's functions in terms of the temperature

at the midpoint and additional temperature-dependent perturbation terms proportional to the thermal gradient parameter γ/L , with $\gamma \equiv \ln T_H/T_C$. The phonon Green's functions can be explicitly written down in terms of the same parameter and the temperature of the midpoint of the conductor. We show that for $\gamma \ll 1$ and $N \gg 1$, a systematic and consistent perturbation theory can be developed that allows us to study the effects of the temperature gradient on the thermal current in the absence of any other perturbation. Further perturbations can then be added in this inhomogeneous thermal system.

While we consider a long nanowire as the simplest inhomogeneous thermal system for illustrative purposes, it should be possible to generalize the framework to include arbitrary thermal inhomogeneities. The method involves reinterpreting the density matrix of a given Hamiltonian with different subsystems at different temperatures in terms of an effective density matrix of a temperature-dependent pseudo-Hamiltonian at one arbitrary but fixed temperature. In the following sections we first introduce the framework for general inhomogeneous systems. We exploit the fact that Wick's theorem can be applied if we incorporate the thermal inhomogeneity in the density matrix via a pseudointeraction term added to our Hamiltonian. We consider the case of phonons in detail which is relevant for thermoelectric devices. While the inhomogeneity generates several additional (pseudo)interaction terms, we illustrate our method by choosing one featured term and evaluating the corresponding effects on the nonequilibrium Green's functions as well as the thermal current within our perturbative framework. We evaluate the frequency-dependent thermal current as well as the nonlinear thermal conductivity for finite-temperature differences to leading order in γ . These effects should be observable in thermal transport measurements in nanowires.

The paper is organized as follows. We introduce the general framework for thermally inhomogeneous systems in Sec. II and obtain the correction to the phonon Green's function to first order in the parameter γ due to a thermal gradient across a nanowire in Sec. III. The resulting thermal current and the corresponding thermal conductance are obtained in Sec. IV. Section V contains a summary and discussion.

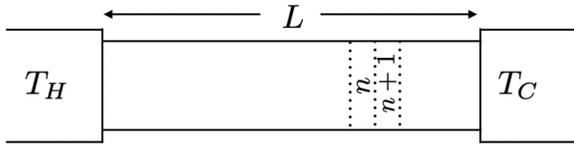


FIG. 1. A wire of length L attached to two leads, hot and cold, with temperatures T_H and T_C , respectively. The temperature inside the wire has a gradient along the length. We imagine dividing the wire into N number of thin slices, the n th slice being at inverse temperature β_n .

II. FRAMEWORK FOR THERMALLY INHOMOGENEOUS SYSTEMS

For simplicity, we consider a wire of length L as shown in Fig. 1 with two ends connected to perfect leads fixed at two different temperatures T_H and T_C ; the subscripts refer to hot and cold, respectively. The temperature difference creates a stationary thermal gradient inside the wire. We now imagine dividing the wire into N number of thin slices, the n th slice being at temperature T_n . The Hamiltonian of the system can be written as a collection of N subsystems,

$$H = \sum_n [H_n^0 + H_n^I + H_{n,n+1}^I], \quad (2.1)$$

where H_n^0 and H_n^I are the free (quadratic) and the interaction parts of the n th slice, respectively, and $H_{n,n+1}^I$ is the interaction between connected slices. The corresponding density matrix is given by

$$\rho(t_0) = \frac{\exp[-\sum_n \beta_n (H_n^0 + H_n^I + H_{n,n+1}^I)]}{\text{Tr}\{\exp[-\sum_n \beta_n (H_n^0 + H_n^I + H_{n,n+1}^I)]\}}. \quad (2.2)$$

The Green's function can be written as

$$G(1, 2) = -i \text{Tr}[\rho(t_0) \mathcal{T} \psi_H(1) \psi_H^\dagger(2)], \quad (2.3)$$

where \mathcal{T} is the time-ordering operator.

The fact that each slice has a different temperature makes the evaluation of the Green's functions difficult because a compact time ordering is not available and the Wick's theorem cannot be applied directly. The fact that the density matrix in (2.2) does not commute with the Hamiltonian (2.1) suggests that a thermally homogeneous Hamiltonian is not a good description of the time evolution of a thermally inhomogeneous system. We take this into account by rewriting the numerator of the density matrix as

$$\sum_n \beta_n [H_n^0 + H_n^I + H_{n,n+1}^I] = \beta_0 H_p, \quad (2.4)$$

$$H_p \equiv \sum_n \left[H_n^0 + \frac{\beta_n - \beta_0}{\beta_0} H_n^0 + \frac{\beta_n}{\beta_0} (H_n^I + H_{n,n+1}^I) \right].$$

We now observe that the density matrix of H defined in (2.1) with different inverse temperatures β_n at different slices n is the same as the density matrix of a pseudointeraction Hamiltonian H_p in (2.4) at an arbitrary but fixed inverse temperature β_0 . We choose to work with the temperature-dependent pseudo-Hamiltonian H_p as our starting point, which means we do all the traces on eigenstates of H_p ; the perturbation theory for this pseudo-Hamiltonian can then be worked out within the standard NEFT framework, albeit with additional

nonstandard temperature-dependent interaction terms. In the following section, we will illustrate the method with a simple but important example, taken from thermoelectricity, where a clear small expansion parameter can be identified and the perturbation theory becomes well defined in the large N limit.

We note that a different series expansion for a thermally inhomogeneous system was proposed in Ref. [18] and Wick's theorem seems to work for an interacting system within the equilibrium Green's function (EGF) framework. There are also exact solutions available for some expectation values in one-dimensional models that can be mapped on to conformal field theories [19,20]. Our framework should be more generally applicable for both EGF and NEFT.

III. NANOWIRE WITH A TEMPERATURE GRADIENT

As a simple illustrative example, we consider phonon propagation across a nanowire. We assume that the free phonon dispersion relation is given by

$$\omega_k^2 = \frac{K}{\rho a^3} \sum_{i=x,y,z} \cos k_i a, \quad (3.1)$$

where ρ is the mass density, and K is the spring constant. We model the temperature profile in the wire in the continuum limit given by

$$\beta_n \rightarrow \beta(x) = \beta_0 e^{\gamma x/L}, \quad (3.2)$$

with

$$T_0 = \sqrt{T_H T_C}, \quad e^\gamma = \frac{T_H}{T_C}. \quad (3.3)$$

In the small γ limit, to linear order in γ , this corresponds to a uniform temperature gradient across the length of the wire, which we take to be the x direction. We note that in a clean strictly one-dimensional wire, the temperature profile remains flat in the bulk, any changes occurring only at the edges [21,22]. However, measurements on thin nanowires show a temperature profile similar to above [23]. For these nanowires, the process to achieve a temperature gradient due to the phonon anharmonicity, phonon-impurity scattering, and/or electron-phonon interactions and for the particles to reach nonequilibrium steady states after the attachment of two leads with different temperature could be complicated. However, after the temperature gradient is formed, the perturbation brought by the temperature inhomogeneity contributes to the phonon Green's functions in first order, compared to the second-order contribution from the other scattering sources mentioned above. We therefore start from a free-phonon model and treat the temperature inhomogeneity as the dominant perturbation term. The second-order effects can be included in the future, adding to the contribution from the temperature inhomogeneity alone. In any case, an exact temperature profile is not important to illustrate the applicability of our framework; we therefore choose the simplest case, leaving a more accurate account of the temperature profile for future work. Figure 1 shows the system being considered, where $\Delta T = T_H - T_C$ is not necessarily small (for example, $T_C = 300$ K, $\Delta T = 30$ K), allowing us to develop a perturbative framework with $\gamma \sim \Delta T/T_C \ll 1$ ($\gamma = 0.1$ in the above example) being the expansion parameter.

In the continuum limit the quadratic part is

$$H^0 = \frac{1}{2} \int \vec{d}r \left[\dot{u}(\vec{r})\dot{u}(\vec{r}) + \frac{V}{2} u(\vec{r})\{u(\vec{r} - \vec{a}) + u(\vec{r} + \vec{a})\} \right], \quad (3.4)$$

where the overdot represents a time derivative. Here, $V = K/\rho a^3$, $u = \sqrt{\rho}U$, U being the displacement of the atoms from the equilibrium positions, and \vec{a} are the lattice vectors. In the simplest case, the perturbative pseudointeraction term generated by the temperature gradient along x is

$$H^{\text{pert}} = \frac{\gamma}{2L} \int \vec{d}r x \left[\dot{u}(\vec{r})\dot{u}(\vec{r}) + \frac{V}{2} u(\vec{r})\{u(\vec{r} - \vec{a}) + u(\vec{r} + \vec{a})\} \right]. \quad (3.5)$$

Note the explicit x -dependent terms reflecting the thermal inhomogeneity of the system, where we chose the midpoint of the wire as the origin $x = 0$. The most interesting part is the term with \vec{a} along x , and we will illustrate our method with a focus on this term. Note also that the interaction term is proportional to γ/L , not just γ .

We note that the linearized temperature profile $\beta(x) = \beta_0[1 + \gamma x/L]$ looks similar to the position-dependent field

$$\begin{aligned} & \delta^{(1)}D(x, x'; \vec{k}; t, t') \\ &= (-i) \frac{\gamma}{2L} \int dt_1 dq \delta(q) (e^{iqx} \{ (ix + \partial_q) [\omega_{\vec{k}}^2 D^0(\vec{k} + \vec{q}, t - t_1) D^0(\vec{k}, t_1 - t') - C^0(\vec{k} + \vec{q}, t - t_1) C^0(\vec{k}, t_1 - t')] \} \\ & \quad + e^{iqx'} \{ (ix' + \partial_q) [\omega_{\vec{k}}^2 D^0(\vec{k}, t - t_1) D^0(\vec{k} - \vec{q}, t_1 - t') - C^0(\vec{k}, t - t_1) C^0(\vec{k} - \vec{q}, t_1 - t')] \} \\ & \quad + aV \sin k_x a D^0(\vec{k}, t - t_1) D^0(\vec{k} - \vec{q}, t_1 - t')). \end{aligned} \quad (3.8)$$

After the q integral, and going to the frequency space, one can identify the retarded self-energy up to linear order in γ by comparing with the Dyson expansion ($D^R = D^{0R} \Sigma^R D^R$) as

$$\Sigma^R(x, x'; \vec{k}, \omega) = \frac{\gamma}{L} \left[\frac{x+x'}{2} \{ \omega_{\vec{k}}^2 - (\omega + i\delta)^2 \} - ia \frac{V}{2} \sin k_x a \right]. \quad (3.9)$$

The corresponding retarded phonon Green's function takes the form

$$\begin{aligned} D^R &= \frac{Z}{(\omega + i\delta)^2 - \omega_{\vec{k}}^2 + i \frac{\gamma a}{L} Z \frac{V}{2} \sin k_x a}, \\ Z &\equiv \frac{1}{1 + \frac{\gamma}{L} \frac{x+x'}{2}}. \end{aligned} \quad (3.10)$$

Note that the imaginary part in the denominator of D^R is proportional to $\gamma a/L$, where $\gamma \ll 1$ is already a small parameter. In addition, we assume $L \gg a$ so that the combination $\gamma a/L = \gamma/N \ll \gamma$ can be considered negligible in the large N limit. While the specific form of the imaginary part in (3.10) is model dependent, the prefactor $\gamma a/L$ (in the leading-order perturbation theory) ensures that the imaginary part remains negligible. Thus the final result for the Green's function, to

that particles couple to in the Luttinger formalism [24] with $\beta(r) = \beta_0[1 + \psi(r)]$ as discussed in Ref. [25]. Our framework, derived directly from the exact density matrix, clarifies the physical meaning of the fictitious ‘‘mechanical’’ field ψ , and with the explicit form of the field coming out of the temperature profile, one can conveniently go beyond the linear-response regime.

The phonon Green's function can be written as

$$D(1, 2) = -i \text{Tr}[\rho(t_0) \mathcal{T} u_{H_p}(1) u_{H_p}(2)]. \quad (3.6)$$

Here, we have used the phonon displacement fields instead of the raising and lowering operators since the interaction (3.5) has an explicit x dependence. In addition, it is useful to define a correlation function

$$C^0(1, 2) = \text{Tr}[\rho(t_0) \mathcal{T} u_{H^0}(1) \dot{u}_{H^0}(2)]. \quad (3.7)$$

The zeroth-order phonon Green's functions $D^0(\vec{p}, t - t')$ are well known and the correlation functions $C^0(\vec{p}, t - t')$ can be easily calculated. The first-order contribution to the phonon Green's function induced by the temperature gradient via the pseudointeraction term H^I , with \vec{a} along x , is then given by

first order in γ and in the large N limit, becomes

$$D^{R/A} = \frac{Z}{(\omega \pm i\delta)^2 - \omega_{\vec{k}}^2}. \quad (3.11)$$

It is interesting that the numerator Z defined in (3.10) mimics the quasiparticle weight factor in the standard equilibrium fermionic many-body quantum field theory. Note that at this level of approximation, the consistency relations between various self-energies (retarded, advanced, greater, and lesser) remain valid: $\Sigma^R = (\Sigma^A)^*$, $\Sigma^R - \Sigma^A = \Sigma^> - \Sigma^<$. We should emphasize that the consistency relations may not be automatically satisfied at each order within our perturbative formulation with the small parameter $\gamma \ll 1$ alone; the additional small parameter $a/L \ll 1$ (equivalently $N \gg 1$) in our current example is critical. On the other hand, these relations should be valid if an exact summation to all orders is available. The first-order corrections to the lesser and greater Green's functions then have the same common Z factor,

$$\begin{aligned} D^{\gtrless} &= Z D^{0\gtrless} = -iZ \frac{\pi}{\omega_{\vec{k}}} \\ & \quad \times [(N_{\vec{k}} + 1)\delta(\omega \mp \omega_{\vec{k}}) + N_{\vec{k}}\delta(\omega \pm \omega_{\vec{k}})], \end{aligned} \quad (3.12)$$

where $N_{\vec{k}}$ are the Bose distribution functions.

In particular, the phonon number density given by

$$\begin{aligned} n(\vec{r}) &= i G^<(\vec{r}' \rightarrow \vec{r}, t' \rightarrow t) \\ &= \int \frac{d\vec{k}}{(2\pi)^3} \frac{1}{1 + \gamma x/L} N_k, \end{aligned} \quad (3.13)$$

where G refers to the phonon Green's function involving the raising and lowering operators instead of the displacement fields. Equation (3.13) has the simple interpretation that the hotter parts ($x < 0$) have more phonons than the cooler parts ($x > 0$).

IV. THERMAL CONDUCTANCE

The usual definition of thermal conductivity assumes a linear-response regime with $\Delta T = (T_H - T_C) \rightarrow 0$. In a thermoelectric device we want ΔT to be large. The general frequency-dependent steady state thermal current across such a wire is given by [26,27]

$$\begin{aligned} J(\omega) &= \frac{\omega}{4} \int d\vec{r}_1 d\vec{r}_2 \{ [D_L^{(c)>}(\vec{r}_1, \vec{r}_2; \omega) \Sigma_L^<(\vec{r}_2, \vec{r}_1; \omega) \\ &\quad - D_L^{(c)<}(\vec{r}_1, \vec{r}_2; \omega) \Sigma_L^>(\vec{r}_2, \vec{r}_1; \omega)] - [L \rightarrow R] \}. \end{aligned} \quad (4.1)$$

Here, the superscript (c) on the phonon Green's function D refers to the central region (the wire), with subscripts L and R referring to the ends connecting the left and right leads, respectively. The subscripts L and R on the self-energy Σ refer to the left and right leads, respectively, and \vec{r}_1, \vec{r}_2 lie in the lead-central interface. For simplicity, we consider a quasi-one-dimensional wire with independent channels in the lead and compute the contribution from one channel. This simplifies the calculation by allowing us to ignore the spatial integrals. (A proper evaluation including all channels would change the prefactor to reflect the fraction of the incoming waves being transmitted/reflected across the two barriers that connect the leads with the wire but should be generally independent of γ .) The greater/lesser Green's functions in the central region are available from the previous section, Eq. (3.12), and the lead self-energy is defined to be proportional to lead's free Green's function, i.e.,

$$\begin{aligned} \Sigma_{L/R}^{\gtrless} \propto D_{L/R}^{0\gtrless} &= -i \frac{\pi}{\omega_k} [(N_k^{L/R} + 1) \delta(\omega \mp \omega_k) \\ &\quad + N_k^{L/R} \delta(\omega \pm \omega_k)]. \end{aligned} \quad (4.2)$$

Now writing the real space Green's functions in terms of their Fourier components and doing the k integral, the frequency-dependent current can be written as

$$\begin{aligned} J^\gamma(\omega) \propto \frac{n^2(\omega)}{\omega} \left[\frac{1}{1 - \frac{\gamma}{2}} N^L(\omega) - \frac{1}{1 + \frac{\gamma}{2}} N^R(\omega) \right. \\ \left. - \frac{\gamma}{1 - \frac{\gamma^2}{4}} N^0(\omega) \right] \Theta(\omega) + (\omega \rightarrow -\omega), \end{aligned} \quad (4.3)$$

where $n(\omega)$ is the phonon density of states, $N(\omega)$ is the Bose distribution function, and $\Theta(\omega)$ is the step function; the superscript 0 refers to the midpoint inverse temperature β_0 , while L and R refer to the temperatures at the left and right leads.

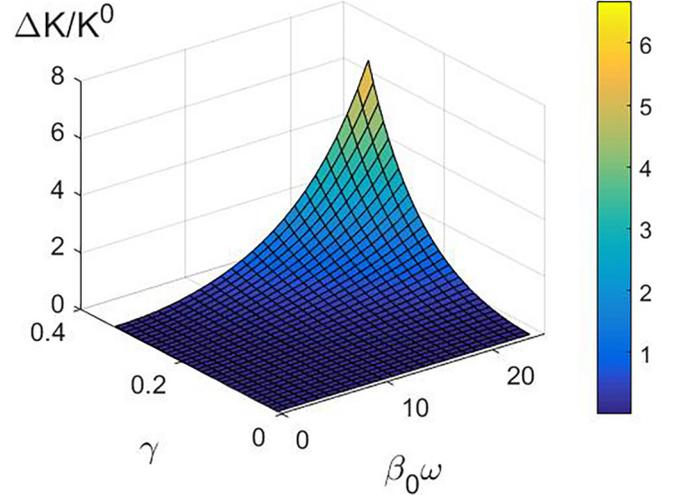


FIG. 2. The ratio $\Delta K/K^0 = [K^\gamma(\omega) - K^0(\omega)]/K^0(\omega)$ obtained from (4.4), plotted as functions of the parameters γ and $\beta_0\omega$.

The midpoint temperature β_0 appears in J^γ only in the combination $\beta_0\omega$ via the Bose distributions. The proportionality factor involves the coupling of the leads to the wire, which will depend on the system. To compare with experiments, a nonlinear thermal conductance has been proposed in Ref. [5], given by

$$\kappa_{nl} = \int K(\omega) d\omega, \quad K(\omega) \equiv \frac{J(\omega)}{\Delta T}. \quad (4.4)$$

In the following, we choose a constant phonon density of states to obtain the thermal current in (4.3) from acoustic phonons. Figure 2 shows the ratio

$$\frac{\Delta K}{K^0} = \frac{[K^\gamma(\omega) - K^0(\omega)]}{K^0(\omega)}, \quad K^0 = \lim_{\gamma \rightarrow 0} K^\gamma. \quad (4.5)$$

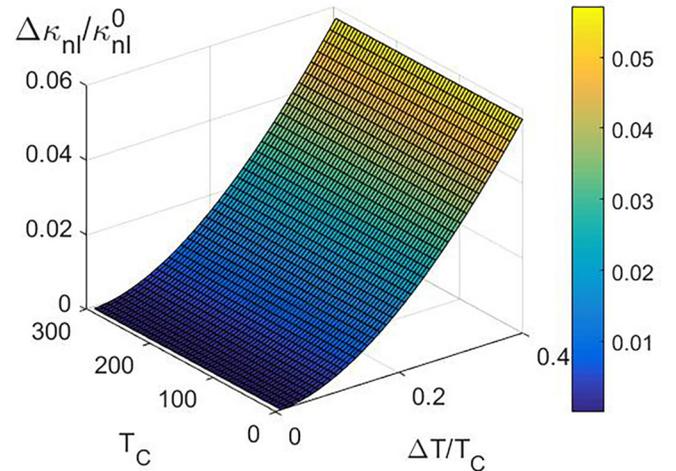


FIG. 3. The ratio $\Delta\kappa_{nl}/\kappa_{nl}^0 = [\kappa_{nl}^\gamma - \kappa_{nl}^0]/\kappa_{nl}^0$ obtained from (4.4), plotted as functions of $\Delta T/T_C$ and T_C .

Figure 3 shows the ratio

$$\frac{\Delta\kappa}{\kappa^0} = \frac{[\kappa_{\text{nl}}^\gamma - \kappa_{\text{nl}}^0]}{\kappa_{\text{nl}}^0}. \quad (4.6)$$

Note that in this case, contributions to κ_{nl} derive primarily from the $\omega \ll 1/\beta_0$ region of the current in Fig. 2. On the other hand, contributions from optical phonons with frequency $\omega \sim 20/\beta_0$, according to Fig. 2, would result in much larger changes in κ_{nl} . Note that thermal conductivity could be defined in a similar fashion, with an added length dependence. However, since we evaluate the ratio K/K^0 , the results will be valid for both conductance and conductivity. These predictions can be verified experimentally.

V. SUMMARY AND DISCUSSION

We have developed a perturbative framework within the standard nonequilibrium quantum field theory to study thermal current in a nanowire connected to hot and cold leads with arbitrary but fixed temperatures T_H and T_C , respectively. The framework allows us to consider the case $\Delta T = (T_H - T_C)$ not necessarily small, which results in a finite-temperature gradient across the wire and conventional field theory with a fixed temperature cannot be applied directly. The framework relies on starting from a temperature-dependent pseudo-Hamiltonian equivalent to an exact density matrix. While we choose to work with a uniform gradient of temperature in a long wire to show the need for and the effectiveness of our method, the framework should be adaptable to more complicated systems, including electrons, with different types of inhomogeneities in temperatures. Of course the method would be practically useful if a small parameter can be identified to

develop a perturbation theory, or if the pseudo-Hamiltonian is exactly soluble.

Based on this framework, we evaluate the frequency-dependent thermal current $J^\gamma(\omega)$ and the nonlinear thermal conductance $\kappa_{\text{nl}}^\gamma$ in a wire due to the temperature difference ΔT , to leading order in γ . In the absence of a theoretical framework to incorporate temperature inhomogeneities, experimental works so far have focused primarily on the linear-response regime, with $\Delta T \rightarrow 0$; we hope that our results would encourage experiments in the finite ΔT regime relevant for thermoelectric devices. If experiments involve larger γ , higher order in perturbation or a self-consistent calculation might be needed.

In real systems the wire would have other sources of scattering that might complicate the interpretation of the results. Although it is known that the effects of bulk disorder on thermal conductivity is negligible in the linear-response regime, the situation is different in the presence of surface disorder, which generates randomly positioned localized phonons [28–31]. It would therefore be important to include the surface disorder effects in this framework to study the effects of any coupling between the phonons generated by surface disorder and those arising from temperature inhomogeneities. In addition, the framework can be easily adapted to investigate the effects of finite ΔT on charge transport, needed to study the efficiency of thermoelectric devices.

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