# Mechanical relations between conductive and radiative heat transfer

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We present a general nonequilibrium Green's function formalism for modeling heat transfer in systems characterized by linear response that establishes the formal algebraic relationships between phonon and radiative conduction, and reveals how upper bounds for the former can also be applied to the latter. We also propose an extension of this formalism to treat systems susceptible to the interplay of conductive and radiative heat transfer, which becomes relevant in atomic systems and at nanometric and smaller separations where theoretical descriptions which treat each phenomenon separately may be insufficient. We illustrate the need for such coupled descriptions by providing predictions for a low-dimensional system of carbyne wires in which the total heat transfer can differ from the sum of its radiative and conductive contributions. Our framework has ramifications for understanding heat transfer between large bodies that may approach direct contact with each other or that may be coupled by atomic, molecular, or interfacial film junctions.

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Characterizing radiative and conductive heat transfer at the nanoscale is essential to understanding the operation of a wide variety of systems and technologies, including heat sinks, thermoelectric devices, thermal microscopy, thermal magnetic recording devices, coherent thermal sources, optoelectronic and optomechanical devices, and thermophotovoltaic devices [1–10]. Much progress has been made toward experimentally measuring heat conduction by phonons in molecular junctions and interfaces at contact [11–17], as well as radiative heat transfer between objects at separations $\gtrsim$10 nm [4,18–24]. Most commonly, conduction in the linear response regime is described atomistically using the nonequilibrium Green's function (NEGF) method [2,5–8,25–30], while radiative heat transfer is modeled through continuum fluctuational electrodynamics [4,9,31–34]. However, recent experiments [35–38] have yielded conflicting accounts of the nature of heat transfer in the extreme near field (ranging from subnanometric separations to $\lesssim$10 nm), raising questions about the interplay between conduction and radiation at such small separations. Simultaneously, recent theoretical works [8,39–45] have begun to shed light on the connections between the formalisms of conductive (whether electronic or phononic) and radiative heat transfer, but these have typically been subject to restrictions including neglect of electromagnetic retardation and consideration of translationally symmetric systems like planar sheets or slabs. In this paper we present a unified linear response formalism that can describe phonon conductive heat transfer (PCHT) and radiative heat transfer (RHT) for arbitrary geometries and separations. The approach puts descriptions of both effects on the same algebraic footing, which is useful for drawing mathematical and physical analogies. For illustration, we demonstrate that recent analytical upper limits on PCHT can be applied to RHT, and further show that our framework can be used to describe situations where both effects couple and contribute significantly to net heat transfer, of particular relevance to recent and ongoing experiments at the nanoscale [4,21,35–38].

Nanoscale PCHT has thus far been treated through atomistic theoretical frameworks primarily using one of two classes of methods. One approach is the so-called NEGF method [2,5–8,25–30], typically used to model heat transfer between two large or semi-infinite metallic or polar dielectric leads across a junction, taken to be either a single atom or molecule or a thin interfacial film; this method has not been applied so much to smaller material bodies exchanging heat. The NEGF method models each material body as being made of atoms, each of which corresponds to harmonic oscillator degrees of freedom along each Cartesian direction representing chemical bonds between neighboring atoms, whose strengths are typically computed via density functional theory. This harmonic model is a frequency domain method, and is valid at temperatures $\lesssim$500 K, when the spatial dimensions relevant to energy transport between the bodies under consideration are smaller than the phonon mean free path in the material, and when other tunable anharmonicities are negligible [2,8,29]. It is this NEGF method that we use to treat PCHT in this work, which is why we consistently use the term PCHT to specifically refer to *coherent* thermal phonon transport in the *linear* regime under the aforementioned conditions. Another typical approach for modeling PCHT is based on molecular dynamics [30,46–49], which is a time domain method that captures anharmonicity in short- and long-range interactions but frequently requires complicated empirical functional forms for interaction potentials.

 

RHT is typically treated using fluctuational electrodynamics, in which material bodies are modeled to have continuum susceptibilities that respond to EM fields propagating between them. Recent analytical and computational formulations include discrete dipolar and multipolar methods [50–54], scattering matrix methods [1,33,34,55–57], finite-difference time domain techniques [31,58,59], and surface or volume integral equation methods [9,32,60]. With the exception of finite-difference time domain methods, all of the other methods discussed are frequency domain methods, which require linear media; however, unlike the case of PCHT, the assumption of linear response is valid under a much broader range of scenarios (including temperature ranges) of relevance to RHT. These methods capture long-range EM effects, but their tendency to use semi-empirical rather than *ab initio* calculations makes them best suited for separations $\gtrsim$10 nm, and their typical neglect of nonlocality and boundary effects at the atomic scale can lead to unphysical predictions as the objects undergoing RHT approach contact. The fact that current experiments are beginning to probe smaller systems [35–37] suggests a need for better understanding of the connections between PCHT and RHT at nanoscale and smaller separations.

Our paper is organized as follows. In Sec. I we explain the general NEGF formalism for computing heat transfer in a system of massless bosonic excitations exhibiting linear response in different collections of bosonic degrees of freedom, which we generically call "components." We derive Landauer-like formulas for the spectrum of energy exchange between any two components, either coupled directly to one another or by a third, and then derive fully general Landauer bounds on heat transfer from them, decomposing the spectrum into transmission channels and bounding the transmission in each channel above by unity. We then explain the relationship between the general NEGF formalism and its application to PCHT and RHT, described in Secs. II and III, respectively. In Sec. IV we identify the relevant components and their couplings, and further clarify the analogies between PCHT and RHT, making it abundantly clear that RHT and PCHT are simply different manifestations of the same abstract principles of energy transport in linear systems. Beyond simply highlighting the abstract connections between the formalisms, in Sec. V we apply the general NEGF formalism to consider PCHT and RHT in a unified manner. We show that far from overcomplicating matters, such a unification is *necessary* in certain regimes. In particular, we consider a model system consisting of collinear atomically thin wires, and show that the resulting net heat transfer power does not simply follow from the sum of the individual radiative and conductive contributions, and may in fact fall below either or both of these contributions. Such an illustration is made possible by an extension of the retarded many-body framework of mesoscale fluctuational EM [61–63], which can account for atom-scale features of material response. Concluding remarks are given in Sec. VI.

## I. GENERAL LINEAR RESPONSE NEGF FORMALISM FOR HEAT TRANSFER

Consider a generic system exhibiting generalized displacements labeled $x(t)$, which may represent electronic wave functions, collective nuclear oscillations giving rise to phonons, EM fields, or other oscillatory phenomena, and respond linearly to generalized forces labeled $F(t)$. These degrees of freedom (DOFs) constitute collections which we generically call "components." Each component exhibits linear equations of motion representing its internal dynamics in isolation and in response to external forces, and each component may be linearly coupled to other components leading to energy transport among them. The following sections will make clear the identities of the components, couplings, generalized displacements, and generalized forces in different systems of interest, like PCHT or RHT; this section focuses on deriving relevant fully general formulas for energy transport among generic coupled components.

Generically, in the time domain, the power radiated or absorbed by a component may be written as $\dot{W} = \langle F(t)\frac{\partial x}{\partial t}\rangle$. Here $\langle\cdots\rangle$ denotes a time average in the steady state, which is equivalent to an ensemble average due to ergodicity. As we have specified that the internal dynamics and couplings are linear, we may equivalently work in the frequency domain, making it easier to apply the fluctuation-dissipation theorem [64] and thereby replace such ensemble averages with deterministic quantities representing the dissipation of the system.

In the frequency domain (where we will generally suppress dependence on angular frequency $\omega$ in the notation) we label these generalized displacements as $|x\rangle$ and the generalized forces as $|F\rangle$, as these quantities are vectors in a complex Hilbert space with the standard inner product. One of the operators relevant to this Hilbert space are the dynamical operator $\hat{Z}^{(0)}$, representing the dynamical equations of motion for each component in isolation, and can equivalently be seen as a generalized impedance or spring constant; its inverse $\hat{Y}^{(0)} = \hat{Z}^{(0)-1}$ represents a generalized admittance for the components in isolation, such that an external force $|F^{(0)}\rangle$ on the components *in isolation* produces a total displacement $|x\rangle = \hat{Y}^{(0)}|F^{(0)}\rangle$. However, energy exchange among components is only possible if couplings are present: as will become clearer later, these couplings may act directly between components, or may act through other components whose equations are eliminated, resulting in effective self-couplings for the remaining components. These couplings are generically represented by the linear operator $\Delta\hat{Z}$: the force $|F\rangle$ on other components due to a displacement $|x\rangle$ from equilibrium of a given component can be written as $|F\rangle = -\Delta\hat{Z}|x\rangle$.

We generally assume this system to be reciprocal, which physically implies an equivalence between emission of energy by sources and absorption of energy by receivers; mathematically, in this complex Hilbert space, $\hat{Z}^{(0)} = \hat{Z}^{(0)\top}$ and $\Delta\hat{Z} = \Delta\hat{Z}^{\top}$ hold, where $^{\top}$ denotes the transpose *without* conjugation and not the Hermitian adjoint $^{\dagger}$, and reciprocity of related operators follows from these relations. Additionally, causality, which physically implies response functions lag their source functions in the time domain, is mathematically expressed in the frequency domain as the condition that $\hat{Y}^{(0)}(-\omega^{\star}) = \hat{Y}^{(0)\star}(\omega)$ for any complex frequency $\omega$. Passivity, which physically implies that the system may conserve or dissipate energy (as in a lossy medium) but not amplify energy (as in a gain medium), is mathematically expressed as the condition that $\mathrm{asym}(Y^{(0)})$, which represents the dissipative contribution to the response of the uncoupled components, is Hermitian positive-definite (in the space of its own support) for any

real positive frequency $\omega$ [33]; we have defined $\text{asym}(\hat{A}) \equiv (\hat{A} - \hat{A}^\dagger)/(2i)$ for any operator $\hat{A}$.

We now turn to the equations of motion for this system in the presence of coupling between its different components. In particular, the total generalized displacement can be written as the sum of the initial displacement $|x^{(0)}\rangle$ and the response of the components $\hat{Y}^{(0)}$, in isolation, to the total generalized force $|F\rangle$; in order to avoid double-counting various contributions, the total generalized force simply arises from the total displacements through the couplings between different components $\Delta\hat{Z}$, as we assume that no other external forces contribute to the system dynamics, and these couplings are the only way for energy to be transmitted among different components. Mathematically this is written as

$$\begin{aligned} |x\rangle &= |x^{(0)}\rangle + \hat{Y}^{(0)}|F\rangle, \\ |F\rangle &= -\Delta\hat{Z}|x\rangle, \end{aligned} \tag{1}$$

and we formally solve this to yield

$$|x\rangle = \hat{Y}\hat{Z}^{(0)}|x^{(0)}\rangle, \tag{2}$$

where we define the total response $\hat{Y} = \hat{Z}^{-1}$ in terms of the total equations of motion (generalized impedance) $\hat{Z} = \hat{Z}^{(0)} + \Delta\hat{Z}$. We stress that the total response $\hat{Y}$ satisfies the same reciprocity, causality, and passivity properties as the decoupled response $\hat{Y}^{(0)}$.

At this point we specify that the system can be partitioned into $N$ components, with each component labeled $n$ specifying a certain set of DOFs; the operator $\hat{Z}^{(0)}$ (and also $\hat{Y}^{(0)}$ by extension) can be written as a block-diagonal matrix as it represents the equations of motion of each component in the absence of coupling between components, so if $\hat{P}_n$ is a projection into the subspace supported by the DOFs of component $n$, then each diagonal block of $\hat{Z}^{(0)}$ is $\hat{Z}^{(0)}_n = \hat{P}_n\hat{Z}^{(0)}\hat{P}_n$, and likewise each diagonal block of $\hat{Y}^{(0)}$ is $\hat{Y}^{(0)}_n = \hat{P}_n\hat{Y}^{(0)}\hat{P}_n$, with $\hat{Y}^{(0)}_n = \hat{Z}^{(0)-1}_n$. (We note that $\hat{Z}$ and $\hat{Y}$ will generally not be block-diagonal with respect to the different components.)

If each component $n$ is maintained independently at a corresponding temperature $T_n$ (uniformly for all of the DOFs constituting that component), then we may write the frequency-domain fluctuation-dissipation theorem as

$$\begin{aligned} &\hat{P}_m\langle|x^{(0)}(\omega)\rangle\langle x^{(0)}(\omega')|\rangle\hat{P}_n \\ &\quad = \frac{2\Pi(\omega, T_n)}{\omega}\,\text{asym}\left[\hat{Y}^{(0)}_n(\omega)\right]\delta_{mn} \times 2\pi\delta(\omega - \omega'), \end{aligned} \tag{3}$$

where $\langle\cdots\rangle$ represents the quantum statistical expectation value; our use of the Planck function $\Pi(\omega, T) = \frac{\hbar\omega}{2}\coth\left(\frac{\hbar\omega}{2k_B T}\right)$ implicitly assumes that all DOFs we consider, when quantized, obey Bose statistics with no chemical potential, which is appropriate for EM fields and for coupled mechanical oscillators under consideration. We also point out that the use of $\text{asym}(\hat{Y}^{(0)}_n)$, as opposed to $\text{asym}[(\hat{Z}^{(0)}_n + \Delta\hat{Z}_{nn})^{-1}]$ if $\hat{P}_n\Delta\hat{Z}\hat{P}_n = \Delta\hat{Z}_{nn} \neq 0$, is valid because the former includes dissipation only within component $n$, whereas the latter may implicitly include dissipation in other coupled components that have been eliminated. In order to compute the heat transfer from component $m$ to component $n$, we account only for fluctuations in component $m$, so that $|x^{(0)}\rangle = \hat{P}_m|x^{(0)}\rangle$ will hold, and compute the work done on component $n$ according to $|F\rangle = -\hat{P}_n\Delta\hat{Z}|x\rangle$ where $|x\rangle = \hat{Y}\hat{Z}^{(0)}\hat{P}_m|x^{(0)}\rangle$. We then write the absorbed power (energy transfer) as $\dot{W} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\langle\text{Tr}\,[-i\omega\hat{P}_n|x(\omega)\rangle\langle F(\omega')|\hat{P}_n]\,e^{-i(\omega-\omega')t}\rangle\,\frac{d\omega d\omega'}{(2\pi)^2}$. Algebraic manipulations involving the definitions of $|x\rangle$ and $|F\rangle$, along with the fluctuation-dissipation theorem in (3) applied to $|x^{(0)}\rangle$ and the causality properties of the relevant response quantities, yield

$$\begin{aligned} \dot{W} = \frac{2}{\pi}\int_0^\infty &\Pi(\omega, T_m) \\ &\times \text{Tr}[\hat{P}_m\,\text{asym}(\hat{Z}^{(0)\dagger})\hat{P}_m\hat{Y}^\dagger\,\text{asym}(\hat{P}_n\Delta\hat{Z})\hat{Y}\hat{P}_m]d\omega \end{aligned}$$

as the gross energy transfer from component $m$ at temperature $T_m$ to component $n$ among a collection of an arbitrary number of thermalized components. From this we define the NEGF energy transfer spectrum between components $m$ and $n$ as

$$\Phi^{(m)}_n = 4\,\text{Tr}[\hat{P}_m\,\text{asym}(\hat{Z}^{(0)\dagger})\hat{P}_m\hat{Y}^\dagger\,\text{asym}(\hat{P}_n\Delta\hat{Z})\hat{Y}\hat{P}_m] \tag{4}$$

independently of the temperature of each component, while the integrated net power transfer can be written as

$$\dot{W}_{m\to n} = \int_0^\infty [\Pi(\omega, T_m) - \Pi(\omega, T_n)]\Phi^{(m)}_n\,\frac{d\omega}{2\pi} \tag{5}$$

in terms of the component temperatures $T_m$ and $T_n$. Furthermore, the heat transfer coefficient (thermal conductance) between two components may be derived by replacing $\Pi(\omega, T_m) - \Pi(\omega, T_n)$ in the integrand with $\lim_{T_m\to T_n}\frac{\Pi(\omega,T_m)-\Pi(\omega,T_n)}{T_m-T_n} = \frac{\partial}{\partial T}\Pi(\omega, T)$. It is worth noting that reciprocity, which has not been exploited in these derivations thus far, is required to show that $\Phi^{(m)}_n = \Phi^{(n)}_m$ at each frequency.

The formula for $\Phi^{(m)}_n$ in (4) is valid for any number of components maintained at their own uniform temperatures, and constitutes a generalization of Landauer/Caroli formulas often used to describe PCHT and RHT [2,28,29,31,33,42,43,61,65,66]. The most fruitful analogies between RHT and PCHT can be extracted from consideration of heat transfer between two components, through direct contact or via contact with a third component. In what follows, we derive formulas for both situations: the first situation is most relevant to RHT between two bodies or PCHT combined with RHT between two bodies in direct contact, while the second situation is most relevant to PCHT combined with RHT between two bodies via a third intermediate body (typically a thin interface or a small atomic or molecular junction), though it can also be applied to formally deriving expressions for RHT between two bodies.

### A. Two components in direct contact

For two components, labeled 1 or 2, in direct contact with each other, we may write the operators $\hat{Z}^{(0)}$ and $\Delta\hat{Z}$ describing the equations of motion and couplings among these components may be written as $2 \times 2$ block matrices

$$\hat{Z}^{(0)} = \begin{bmatrix} \hat{Z}^{(0)}_1 & 0 \\ 0 & \hat{Z}^{(0)}_2 \end{bmatrix}, \tag{6}$$

$$\Delta\hat{Z} = \begin{bmatrix} \Delta\hat{Z}_{1,1} & \Delta\hat{Z}_{1,2} \\ \Delta\hat{Z}_{2,1} & \Delta\hat{Z}_{2,2} \end{bmatrix}, \tag{7}$$

which in turn implies that

$$\hat{Y}^{(0)} = \begin{bmatrix} \hat{Y}_1^{(0)} & 0 \\ 0 & \hat{Y}_2^{(0)} \end{bmatrix} \tag{8}$$

must also hold; the existence of nontrivial diagonal and off-diagonal blocks in $\Delta\hat{Z}$ typically arises from couplings to other components that are mathematically eliminated in favor of these two components. Evaluation of the energy transfer spectrum (4) requires inversion of these block matrices of operators, which is saved for Appendix A for the sake of brevity in this section. The result is written as

$$\begin{aligned} \Phi = 4\,\mathrm{Tr}\big[\,\mathrm{asym}\,\big(\hat{Z}_1^{(0)\dagger}\big)\big(\hat{Z}_1^{(0)\dagger} + \Delta\hat{Z}_{1,1}^{\dagger}\big)^{-1}\big[\hat{1} - \Delta\hat{Z}_{2,1}^{\dagger}\big(\hat{Z}_2^{(0)\dagger} + \Delta\hat{Z}_{2,2}^{\dagger}\big)^{-1}\Delta\hat{Z}_{1,2}^{\dagger}\big(\hat{Z}_1^{(0)\dagger} + \Delta\hat{Z}_{1,1}^{\dagger}\big)^{-1}\big]^{-1} \\ \times\ \Delta\hat{Z}_{2,1}^{\dagger}\big(\hat{Z}_2^{(0)\dagger} + \Delta\hat{Z}_{2,2}^{\dagger}\big)^{-1}\,\mathrm{asym}\,\big(\hat{Z}_2^{(0)\dagger}\big)\big(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\big)^{-1}\Delta\hat{Z}_{2,1} \\ \times\ \big[\hat{1} - \big(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\big)^{-1}\Delta\hat{Z}_{1,2}\big(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\big)^{-1}\Delta\hat{Z}_{2,1}\big]^{-1}\big(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\big)^{-1}\big] \end{aligned} \tag{9}$$

and as asym $(\hat{Z}_1^{(0)\dagger})$ and asym$(\hat{Z}_2^{(0)\dagger})$ are Hermitian positive-semidefinite operators with well-defined Hermitian square roots, then the energy transfer spectrum is nonnegative. Exploiting this further allows for factorizing $\mathrm{asym}(\hat{Z}_n^{(0)\dagger}) = [\mathrm{asym}(\hat{Z}_n^{(0)\dagger})^{1/2}]^2$ for each component $n \in \{1, 2\}$, and rearranging the trace allows for writing

$$\begin{aligned} \Phi(\omega) = 4\big\|\,\mathrm{asym}\,\big(\hat{Z}_2^{(0)\dagger}\big)^{1/2}\big(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\big)^{-1}\Delta\hat{Z}_{2,1}\big[\hat{1} - \big(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\big)^{-1}\Delta\hat{Z}_{1,2}\big(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\big)^{-1}\Delta\hat{Z}_{2,1}\big]^{-1} \\ \times\ \big(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\big)^{-1}\,\mathrm{asym}\,\big(\hat{Z}_1^{(0)\dagger}\big)^{1/2}\big\|_{\mathrm{F}}^2, \end{aligned} \tag{10}$$

where $\|\hat{A}\|_{\mathrm{F}} = \sqrt{\mathrm{Tr}[\hat{A}^{\dagger}\hat{A}]}$ is the Frobenius norm. This is the general NEGF formula for the energy transfer spectrum between two components in direct contact, in terms of their individual and mutual responses.

### B. Two components coupled via an intermediate component

We now consider the case of the two components, labeled 1 or 2, which are coupled only to a third intermediate component, labeled 3, but not directly to each other. Mathematically, this means the operators $\hat{Z}^{(0)}$ and $\Delta\hat{Z}$ describing the equations of motion and couplings among these components may be written as $3 \times 3$ block matrices

$$\hat{Z}^{(0)} = \begin{bmatrix} \hat{Z}_1^{(0)} & 0 & 0 \\ 0 & \hat{Z}_2^{(0)} & 0 \\ 0 & 0 & \hat{Z}_3^{(0)} \end{bmatrix}, \tag{11}$$

$$\Delta\hat{Z} = \begin{bmatrix} 0 & 0 & \Delta\hat{Z}_{1,3} \\ 0 & 0 & \Delta\hat{Z}_{2,3} \\ \Delta\hat{Z}_{3,1} & \Delta\hat{Z}_{3,2} & 0 \end{bmatrix}, \tag{12}$$

which in turn implies that

$$\hat{Y}^{(0)} = \begin{bmatrix} \hat{Y}_1^{(0)} & 0 & 0 \\ 0 & \hat{Y}_2^{(0)} & 0 \\ 0 & 0 & \hat{Y}_3^{(0)} \end{bmatrix} \tag{13}$$

must also hold, where the vanishing of the components $\Delta\hat{Z}_{1,2} = (\Delta\hat{Z}_{2,1})^{\top}$ follows from the assumption that components 1 and 2 have no direct coupling to each other; we also point out that compared to the general two-component formula, here we do not include couplings between a given component and itself (i.e., $\Delta\hat{Z}$ has vanishing diagonal blocks), as we assume that there are no other components which we have implicitly eliminated. Once again leaving the details to Appendix B, and again making use of the fact that asym$(\hat{Y}_1^{(0)})$ and asym$(\hat{Y}_2^{(0)})$ are Hermitian positive-semidefinite operators to factorize the trace expression, we write (4) as

$$\begin{aligned} \Phi(\omega) = 4\big\|\,\mathrm{asym}\,\big(\hat{Y}_2^{(0)}\big)^{1/2}\Delta\hat{Z}_{2,3} \\ \times\ \big(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,1}\hat{Y}_1^{(0)}\Delta\hat{Z}_{1,3} - \Delta\hat{Z}_{3,2}\hat{Y}_2^{(0)}\Delta\hat{Z}_{2,3}\big)^{-1} \\ \times\ \Delta\hat{Z}_{3,1}\,\mathrm{asym}\,\big(\hat{Y}_1^{(0)}\big)^{1/2}\big\|_{\mathrm{F}}^2 \end{aligned} \tag{14}$$

so that all operator products may be evaluated in the space of component 3. This is the general NEGF formula for the energy transfer spectrum between two components in contact only with a third in terms of their individual responses and mutual couplings.

In the previous subsection it was noted that for heat transfer between two components that are directly coupled, the couplings $\Delta\hat{Z}_{mn}$ for $m, n \in \{1, 2\}$ (particularly the diagonal blocks) often arise from mathematically eliminating another component to which these two components are coupled, even if those are the only couplings. At this point we rigorously prove this equivalence for the specific case where the two components are physically coupled only to a third component. We start by rewriting (2) in terms of the degrees of freedom of the three components and noting that $|x^{(0)}\rangle = \hat{P}_1|x^{(0)}\rangle$ can be used when considering energy transfer from component 1 to component 2. Explicitly, this means writing

$$\begin{bmatrix} \hat{Z}_1^{(0)} & 0 & \Delta\hat{Z}_{1,3} \\ 0 & \hat{Z}_2^{(0)} & \Delta\hat{Z}_{2,3} \\ \Delta\hat{Z}_{3,1} & \Delta\hat{Z}_{3,2} & \hat{Z}_3^{(0)} \end{bmatrix}\begin{bmatrix} |x_1\rangle \\ |x_2\rangle \\ |x_3\rangle \end{bmatrix} = \begin{bmatrix} \hat{Z}_1^{(0)}|x_1^{(0)}\rangle \\ 0 \\ 0 \end{bmatrix}$$

and then eliminating $|x_3\rangle = -\hat{Y}_3^{(0)}(\Delta\hat{Z}_{3,1}|x_1\rangle + \Delta\hat{Z}_{3,2}|x_2\rangle)$. This yields the simpler equation in terms of $2 \times 2$ block

matrices

$$\begin{bmatrix} \hat{Z}_1^{(0)} - \Delta\hat{Z}_{1,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,1} & -\Delta\hat{Z}_{1,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,2} \\ -\Delta\hat{Z}_{2,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,1} & \hat{Z}_2^{(0)} - \Delta\hat{Z}_{2,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,2} \end{bmatrix} \begin{bmatrix} |x_1\rangle \\ |x_2\rangle \end{bmatrix} = \begin{bmatrix} \hat{Z}_1^{(0)}|x_1^{(0)}\rangle \\ 0 \end{bmatrix},$$

where the replacements

$$\hat{Z}^{(0)} \to \begin{bmatrix} \hat{Z}_1^{(0)} & 0 \\ 0 & \hat{Z}_2^{(0)} \end{bmatrix},$$

$$\Delta\hat{Z} \to \begin{bmatrix} -\Delta\hat{Z}_{1,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,1} & -\Delta\hat{Z}_{1,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,2} \\ -\Delta\hat{Z}_{2,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,1} & -\Delta\hat{Z}_{2,3}\hat{Y}_3^{(0)}\Delta\hat{Z}_{3,2} \end{bmatrix}$$

may be made. Hence, the remainder of the derivation of the expression for heat transfer is the same, as (3) for component 1 and the expression $\dot{W} = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\langle \mathrm{Tr}[-i\omega\hat{P}_2|x(\omega)\rangle\langle F(\omega')|\hat{P}_2]e^{-i(\omega-\omega')t}\rangle\frac{d\omega d\omega'}{(2\pi)^2}$ for the power transfer are both unchanged, thereby proving the equivalence between the two expressions [Eqs. (10) and (14)] for the general NEGF energy transfer spectrum with these identifications in mind.

Writing the energy transfer spectrum as (14) cannot only clarify analogies between PCHT and RHT, but it also naturally leads to expressions for upper bounds on the spectrum. To derive such bounds, it will be helpful to define the operators $\hat{\Lambda}_n = \Delta\hat{Z}_{n,3}^{\dagger}\,\mathrm{asym}(\hat{Y}_n^{(0)})\Delta\hat{Z}_{n,3}$, being the dissipation of each component $n \in \{1, 2\}$ multiplied by the corresponding couplings to component 3, and the Green's function of component 3

$$\hat{Y}_3 \equiv \left(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,1}\hat{Y}_1^{(0)}\Delta\hat{Z}_{1,3} - \Delta\hat{Z}_{3,2}\hat{Y}_2^{(0)}\Delta\hat{Z}_{2,3}\right)^{-1}, \quad (15)$$

which is modified from its bare value $\hat{Y}_3^{(0)}$ due to couplings to components 1 and 2. Given this, we will show that the energy transfer spectrum can be written in the Landauer form [2,3,8,28] as $\Phi = \|\hat{t}\|_{\mathrm{F}}^2$ where $\hat{t} = 2\hat{\Lambda}_2^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2}$. The goal then will be to place bounds on the eigenvalues of $\hat{t}^{\dagger}\hat{t}$ at each $\omega$. The fact that $\hat{t}^{\dagger}\hat{t}$ is Hermitian positive-semidefinite makes clear that its eigenvalues, called the transmission eigenvalues (as $\hat{t}^{\dagger}\hat{t}$ is like a transmission intensity matrix), are all nonnegative, placing a lower bound on their values. The following will show how to derive upper bounds of unity on the transmission eigenvalues.

The derivations thus far have actually not made use of the reciprocity of the system, namely that $\hat{Z}_3^{(0)} = \hat{Z}_3^{(0)\top}$, $\hat{Z}_n^{(0)} = \hat{Z}_n^{(0)\top}$, and $\Delta\hat{Z}_{n,3} = \Delta\hat{Z}_{3,n}^{\top}$ for $n \in \{1, 2\}$, but these reciprocity relations are needed for the upper bounds on the transmission eigenvalues. Additionally, two further assumptions are needed, namely that $\mathrm{asym}(\hat{Z}_3^{(0)}) \to 0$, and that the block matrices $\Delta\hat{Z}_{n,3}$ for $n \in \{1, 2\}$ are purely real. These assumptions will later be justified for the particular cases of PCHT as well as RHT.

With this, it can be seen that $\mathrm{asym}(\hat{Y}_3) = \hat{Y}_3^{\dagger}\,\mathrm{asym}(\hat{Y}_3^{-1\dagger})\hat{Y}_3$. Expanding the middle term after exploiting $\mathrm{asym}(\hat{Z}_3^{(0)}) = 0$ gives $\mathrm{asym}(\hat{Y}_3^{-1}) = -\hat{\Lambda}_1 - \hat{\Lambda}_2$, as the real-valued and reciprocal nature of $\Delta\hat{Z}_{n,3}$ imply $\Delta\hat{Z}_{3,n}\,\mathrm{asym}(\hat{Y}_n^{(0)})\Delta\hat{Z}_{n,3} = \Delta\hat{Z}_{n,3}^{\dagger}\,\mathrm{asym}(\hat{Y}_n^{(0)})\Delta\hat{Z}_{n,3}$ for $n \in \{1, 2\}$. This means $\mathrm{asym}(\hat{Y}_3^{-1\dagger}) = -\,\mathrm{asym}(\hat{Y}_3^{-1}) = \hat{\Lambda}_1 + \hat{\Lambda}_2$. Therefore, $\mathrm{asym}(\hat{Y}_3) = \hat{Y}_3^{\dagger}(\hat{\Lambda}_1 + \hat{\Lambda}_2)\hat{Y}_3$. This expression may be rearranged as $\hat{Y}_3^{\dagger}\hat{\Lambda}_1\hat{Y}_3 + \hat{Y}_3^{\dagger}\hat{\Lambda}_2\hat{Y}_3 - \mathrm{asym}(\hat{Y}_3) = 0$, and as $\hat{\Lambda}_1$ is Hermitian positive-semidefinite, then $\hat{\Lambda}_1^{1/2}$ exists, so this expression may be multiplied on the left and right by $2\hat{\Lambda}_1^{1/2}$ to yield $\hat{t}^{\dagger}\hat{t} + 4\hat{\Lambda}_1^{1/2}\hat{Y}_3^{\dagger}\hat{\Lambda}_1\hat{Y}_3\hat{\Lambda}_1^{1/2} - \frac{2}{i}(\hat{\Lambda}_1^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2} - \hat{\Lambda}_1^{1/2}\hat{Y}_3^{\dagger}\hat{\Lambda}_1^{1/2}) = 0$, where as a reminder, $\hat{t} = 2\hat{\Lambda}_2^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2}$. Finally, adding the identity operator $\hat{1}$ to both sides yields $\hat{t}^{\dagger}\hat{t} + 4\hat{\Lambda}_1^{1/2}\hat{Y}_3^{\dagger}\hat{\Lambda}_1\hat{Y}_3\hat{\Lambda}_1^{1/2} - \frac{2}{i}(\hat{\Lambda}_1^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2} - \hat{\Lambda}_1^{1/2}\hat{Y}_3^{\dagger}\hat{\Lambda}_1^{1/2}) + \hat{1} = \hat{1}$. This expression can be rewritten as $\hat{t}^{\dagger}\hat{t} + (\hat{1} - \frac{2}{i}\hat{\Lambda}_1^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2})^{\dagger}(\hat{1} - \frac{2}{i}\hat{\Lambda}_1^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2}) = \hat{1}$, showing that $\hat{t}^{\dagger}\hat{t}$ is added another Hermitian positive-semidefinite operator to yield the identity. Therefore, the eigenvalues of $\hat{t}^{\dagger}\hat{t}$ can never exceed 1, matching the prior expressions [28,29,66,67]. Additionally, because the operator $\hat{1} - \frac{2}{i}\hat{\Lambda}_1^{1/2}\hat{Y}_3\hat{\Lambda}_1^{1/2}$ is not the zero operator, its rank must be at least 1, meaning at least one of its eigenvalues must be strictly positive; in turn, at least one of the eigenvalues of $\hat{t}^{\dagger}\hat{t}$ must be strictly less than 1. We stress that whenever heat transfer between two components that are directly coupled can be physically equated to heat transfer between the same two components with effective couplings only via a third (possibly aggregate) component, these transmission eigenvalue bounds must hold for that system. Additionally, we expect that even if $\Delta\hat{Z}$ were to have nonzero blocks other than $\Delta\hat{Z}_{n,3}$ (and their transposes) for components $n \in \{1, 2\}$, which could represent more general heat transfer between a pair of components among a collection of $N$ components (for any integer $N \geqslant 3$) by virtue of aggregating the other components into an overall third intermediate component, similar bounds should hold in general, though we do not prove that statement; put simply, Landauer bounds of unity should hold for each channel even between two components connected via a third where each of these components could in principle be connected to many other components in turn.

## II. APPLICATIONS TO PCHT

The general NEGF formalism and expression for the energy transfer spectrum (4) applies to PCHT among a collection of material bodies, modeled as effective harmonic oscillators connected to each other via harmonic short- or long-range couplings, each maintained at separate uniform temperatures . Prior works have typically focused on PCHT between two large bodies, typically leads acting as thermal reservoirs, exchanging heat via harmonic coupling through a third small body in between, typically a molecular junction or a thin interfacial film; computationally, this has the benefit of allowing most matrix evaluations to occur in the much smaller space of the intermediate body as opposed to the larger space of one of the leads. Given this, in what follows, we derive the equations of motion for collective atomic oscillations effecting phonons from the Lagrangian for three bodies, each comprising collections of coupled oscillators with masses $m_{\alpha a}$, displacements $x_{\alpha a i}$, and spring couplings $K_{\alpha a i,\beta b j}$ for body labels $\alpha, \beta \in \{1, 2, 3\}$, atomic labels $a, b, c$ within each body, and Cartesian indices $i, j, k \in \{x, y, z\}$, with sources only in body $\alpha = 1$ denoted $x_{1ai}^{(0)}$. Note for comparison

with previous work that bodies 1 and 2, representing infinite reservoirs (leads), are typically labeled L and R, while body 3, representing an compact intermediate (central) device, is typically labeled C. We emphasize that while our derivations focus on the particular case of two bodies connected to a third in order to make connections to past work clearer, the correspondence between abstract linear operators and specific quantities of interest to PCHT is easily generalized to PCHT among a collection of coupled bodies.

The Lagrangian for this system is written as

$$\begin{aligned}2L = &\sum_{a,i} m_{1a}\left(\dot{x}_{1ai} - \dot{x}^{(0)}_{1ai}\right)^2 \\ &- \sum_{a,i,a',i'} K_{1ai,1a'i'}\left(x_{1ai} - x^{(0)}_{1ai}\right)\left(x_{1a'i'} - x^{(0)}_{1a'i'}\right) \\ &- \sum_{a,i,c,k} (K_{1ai,3ck} + K_{3ck,1ai}) x_{1ai} x_{3ck} \\ &+ \sum_{b,j} m_{2b}\dot{x}^2_{2bj} - \sum_{b,j,b',j'} K_{2bj,2b'j'} x_{2bj} x_{2b'j'} \\ &- \sum_{b,j,c,k} (K_{2bj,3ck} + K_{3ck,2bj}) x_{2bj} x_{3ck} \\ &+ \sum_{c,k} m_{3c}\dot{x}^2_{3ck} - \sum_{c,k,c',k'} K_{3ck,3c'k'} x_{3ck} x_{3c'k'} \end{aligned} \tag{16}$$

and minimization of the action $S = \int_{-\infty}^{\infty} Ldt$ leads to the time domain classical equations of motion

$$\begin{aligned} &m_{1a}\ddot{x}_{1ai} + \sum_{a',i'} K_{1ai,1a'i'} x_{1a'i'} + \sum_{c,k} K_{1ai,3ck} x_{3ck} \\ &\quad = m_{1a}\ddot{x}^{(0)}_{1ai} + \sum_{a',i'} K_{1ai,1a'i'} x^{(0)}_{1a'i'}, \\ &m_{2b}\ddot{x}_{2bj} + \sum_{b',j'} K_{2bj,2b'j'} x_{2b'j'} + \sum_{c,k} K_{2bj,3ck} x_{3ck} = 0, \\ &m_{3c}\ddot{x}_{3ck} + \sum_{c',k'} K_{3ck,3c'k'} x_{3c'k'} + \sum_{a,i} K_{3ck,1ai} x_{1ai} \\ &\quad + \sum_{b,j} K_{3ck,2bj} x_{2bj} = 0 \end{aligned} \tag{17}$$

for these displacements. In the frequency domain these become

$$\begin{aligned} &-\omega^2 m_{1a} x_{1ai} + \sum_{a',i'} K_{1ai,1a'i'} x_{1a'i'} + \sum_{c,k} K_{1ai,3ck} x_{3ck} \\ &\quad = -\omega^2 m_{1a} x^{(0)}_{1ai} + \sum_{a',i'} K_{1ai,1a'i'} x^{(0)}_{1a'i'}, \\ &-\omega^2 m_{2b} x_{2bj} + \sum_{b',j'} K_{2bj,2b'j'} x_{2b'j'} + \sum_{c,k} K_{2bj,3ck} x_{3ck} = 0, \\ &-\omega^2 m_{3c} x_{3ck} + \sum_{c',k'} K_{3ck,3c'k'} x_{3c'k'} + \sum_{a,i} K_{3ck,1ai} x_{1ai} \\ &\quad + \sum_{b,j} K_{3ck,2bj} x_{2bj} = 0 \end{aligned} \tag{18}$$

and these equations can be collected into matrix form with vectors $x_\alpha$ and matrices $K_{\alpha\beta}$ and $M_\alpha$, upon which the identifications $\hat{Z}^{(0)}_\alpha \to K_{\alpha\alpha} - \omega^2 M_\alpha$ and $\Delta\hat{Z}_{\alpha\beta} \to (1 - \delta_{\alpha\beta})K_{\alpha\beta}$ can be made, where $K_{\alpha\beta} = K^\top_{\beta\alpha}$ are real valued, and $M_\alpha$ are real valued too; we note that the as the matrices $K$ encode spring constants which multiply *differences* in atomic positions (i.e., relative displacements) to yield forces, the diagonal blocks $K_{\alpha\alpha}$ entering $\hat{Z}^{(0)}_\alpha$ should actually include the effects of couplings to other bodies as are present in the off-diagonal blocks $K_{\alpha\beta}$ for all $\beta \neq \alpha$, so that all forces are balanced in the equations of motion. With these replacements, the energy transfer spectrum becomes

$$\begin{aligned} \Phi = 4\,\mathrm{Tr}[&K_{3,1}\,\mathrm{asym}[(K_{1,1} - \omega^2 M_1)^{-1}]K_{1,3}[K_{3,3} - \omega^2 M_3 \\ &- K_{3,1}(K_{1,1} - \omega^2 M_1)^{-1}K_{1,3} \\ &- K_{3,2}(K_{2,2} - \omega^2 M_2)^{-1}K_{2,3}]^{-1\dagger} \\ &\times K_{3,2}\,\mathrm{asym}[(K_{2,2} - \omega^2 M_2)^{-1}]K_{2,3}[K_{3,3} - \omega^2 M_3 \\ &- K_{3,1}(K_{1,1} - \omega^2 M_1)^{-1}K_{1,3} \\ &- K_{3,2}(K_{2,2} - \omega^2 M_2)^{-1}K_{2,3}]^{-1}], \end{aligned} \tag{19}$$

where the identifications

$$\hat{Y}^{(0)}_\alpha \to g_\alpha = (K_{\alpha\alpha} - \omega^2 M_\alpha)^{-1} \tag{20}$$

as the retarded Green's function of lead $\alpha \in \{1, 2\}$ (with $g^\dagger_\alpha$ being the advanced Green's function),

$$\hat{Y}_3 \to G = (K_{3,3} - \omega^2 M_3 - K_{3,1} g_1 K_{1,3} - K_{3,2} g_2 K_{2,3})^{-1} \tag{21}$$

as the retarded Green's function of the device including connections to the leads (with $G^\dagger$ being the advanced Green's function), and

$$\Lambda_\alpha = K_{3,\alpha}\,\mathrm{asym}(g_\alpha)K_{\alpha,3} \tag{22}$$

for $\alpha \in \{1, 2\}$ being the dissipation terms at the interface of the device with each lead can immediately be made. Thus, this general formalism does reproduce the standard Landauer formula [2,8,25,28]

$$\Phi(\omega) = 4\,\mathrm{Tr}[\Lambda_1 G^\dagger \Lambda_2 G] \tag{23}$$

for phonon heat transport between two leads across a device. Note that while $K_{\alpha\alpha}$ and $M_\alpha$ are real valued, $g_\alpha$ is complex valued because inversion of an infinite-dimensional matrix is made finite dimensional by considering propagation of phonons far from the device interface to be equivalent to energy loss (so $G$ is also complex valued in turn); alternatively, if the leads are large but finite, dissipation may be added heuristically by replacing, including in the definitions of $g_\alpha$, every instance of $-\omega^2 M_\alpha$ with $-i\omega B_\alpha - \omega^2 M_\alpha$ for $\alpha \in \{1, 2\}$ where the diagonal positive-definite matrices $B_\alpha$ represent appropriate dissipation coefficients for the oscillators. Additionally, the assumptions underlying the derivation of the upper bound on the transmission eigenvalues hold here, so those derivations remain valid in this context: all of the $K$ and $M$ matrices are real valued and reciprocal, and $\mathrm{asym}(K_{3,3} - \omega^2 M_3) = 0$ because the compact device will not have any channels for dissipation in the absence of coupling to reservoirs (leads). Thus, the general NEGF formalism for heat transfer in linear response systems can be exactly mapped to the specific NEGF formalism for linear PCHT.

Physically, the harmonic oscillators represent nuclei dressed by inner-shell electrons, and the couplings represent chemical bonds between these oscillators, typically computed via density functional theory and often anisotropic. We again stress that the correspondences $\hat{Z}^{(0)}_{\alpha} \to K_{\alpha\alpha} - \omega^2 M_{\alpha}$ and $\Delta\hat{Z}_{\alpha\beta} \to (1-\delta_{\alpha\beta})K_{\alpha\beta}$ for PCHT are generally applicable even beyond the specific case of two bodies coupled only to a third intermediate body, which allows more general scenarios for PCHT to be treated using (4); moreover, these derivations do not assume that the material bodies exhibit any particular geometry or spatial symmetry properties.

## III. APPLICATIONS TO RHT

The general NEGF formalism and expression for the energy transfer spectrum (4) also applies to RHT among a collection of linearly polarizable bodies that can radiate EM fields. Prior works have typically focused on RHT between two polarizable bodies, whether spatially compact or of infinite extent, in vacuum. The connection to the above general linear response formalism for heat transfer requires somewhat more of a conceptual leap compared to the connection for PCHT. In particular, components 1 and 2 are the polarizable material bodies in question, while component 3, rather than representing a material body, is actually the *vacuum EM field* pervading all of space. A Lagrangian for this system can easily be written for the case where the polarizable bodies are made of atomic harmonic oscillators, with equilibrium positions $\mathbf{r}_{\alpha a}$ for body $\alpha \in \{1,2\}$ and atom label $a$, and with charges $q_{\alpha a}$ that couple to EM fields; the sources are taken to be in body 1. That said, the results are generalizable to other linear media whose response functions are more complicated than those of harmonic oscillators, and to cases with more than two material bodies present; in particular, the use of partial bound charges associated with harmonic oscillators more accurately describes polar dielectric media compared to metals, but the results are generalizable to metals, semimetals, and other media with susceptibilities that may be nonlocal, inhomogeneous, or anisotropic.

The Lagrangian for this system is written as

$$\begin{aligned} 2L = &\sum_{a,i} m_{1a}\left(\dot{x}_{1ai} - \dot{x}^{(0)}_{1ai}\right)^2 \\ &- \sum_{a,i,a',i'} K_{1ai,1a'i'}\left(x_{1ai} - x^{(0)}_{1ai}\right)\left(x_{1a'i'} - x^{(0)}_{1a'i'}\right) \\ &+ 2\sum_{a,i} q_{1a}\dot{x}_{1ai}A_i(\mathbf{r}_{1a}) + \sum_{b,j} m_{2b}\dot{x}^2_{2bj} \\ &- \sum_{b,j,b',j'} K_{2bj,2b'j'}x_{2bj}x_{2b'j'} + 2\sum_{b,j} q_{2b}\dot{x}_{2bj}A_j(\mathbf{r}_{2b}) \\ &+ \int\left[\frac{1}{c^2}\left(\frac{\partial\mathbf{A}}{\partial t}\right)^2 - (\nabla\times\mathbf{A})^2\right]d^3x \end{aligned} \tag{24}$$

introducing the magnetic potential $\mathbf{A}$, working in the Weyl gauge (vanishing electric potential). Minimizing the action $S = \int_0^{\infty} L dt$ leads to the time domain classical equations of motion

$$\begin{aligned} &m_{1a}\ddot{x}_{1ai} + \sum_{a',i'} K_{1ai,1a'i'}x_{1a'i'} - q_{1a}E_i(\mathbf{r}_{1a}) \\ &\quad = m_{1a}\ddot{x}^{(0)}_{1ai} + \sum_{a',i'} K_{1ai,1a'i'}x^{(0)}_{1a'i'}, \\ &m_{2b}\ddot{x}_{2bj} + \sum_{b',j'} K_{2bj,2b'j'}x_{2b'j'} - q_{2b}E_j(\mathbf{r}_{2b}) = 0, \\ &\left(\nabla\times(\nabla\times) + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E} \\ &\quad = -\frac{1}{c^2}\left(\sum_a q_{1a}\ddot{\mathbf{x}}_{1a}\delta^3(\mathbf{x}-\mathbf{r}_{1a}) + \sum_b q_{2b}\ddot{\mathbf{x}}_{2b}\delta^3(\mathbf{x}-\mathbf{r}_{2b})\right) \end{aligned} \tag{25}$$

for the displacements $x_{1ai}$ and $x_{2bj}$ and electric field $\mathbf{E}(t,\mathbf{x}) = -\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t}$, where the magnetic contribution to the Lorentz force $\frac{q_{\alpha a}}{c}\dot{\mathbf{x}}_{\alpha a}\times\mathbf{B}$ is dropped for each atom as it is a nonlinear term that has negligible contribution for speeds much less than the speed of light $c$ (which is generally true for thermal fluctuations at reasonable temperatures). Although the third equation should initially be written in terms of $\mathbf{A}$, a partial time derivative is applied to both sides of the equation to simplify the equations in terms of $\mathbf{E}$. In the frequency domain, these equations of motion become

$$\begin{aligned} &-\omega^2 m_{1a}x_{1ai} + \sum_{a',i'} K_{1ai,1a'i'}x_{1a'i'} - q_{1a}E_i(\mathbf{r}_{1a}) \\ &\quad = -\omega^2 m_{1a}x^{(0)}_{1ai} + \sum_{a',i'} K_{1ai,1a'i'}x^{(0)}_{1a'i'}, \\ &\quad -\omega^2 m_{2b}x_{2bj} + \sum_{b',j'} K_{2bj,2b'j'}x_{2b'j'} - q_{2b}E_j(\mathbf{r}_{2b}) = 0, \\ &\left(\frac{c^2}{\omega^2}\nabla\times(\nabla\times) - 1\right)\mathbf{E} \\ &\quad = \left(\sum_a q_{1a}\mathbf{x}_{1a}\delta^3(\mathbf{x}-\mathbf{r}_{1a}) + \sum_b q_{2b}\mathbf{x}_{2b}\delta^3(\mathbf{x}-\mathbf{r}_{2b})\right) \end{aligned} \tag{26}$$

and these equations may again be collected into matrix form and identified with the generic linear response operators. For polarizable bodies $\alpha \in \{1,2\}$, the operators

$$\hat{Z}^{(0)}_{\alpha} \to K_{\alpha\alpha} - \omega^2 M_{\alpha} \tag{27}$$

are the equations of motion defining the response. Meanwhile, $\mathbf{E}$ is a field defined throughout all space, so matrix products correspond to convolution integrals in space: this means the operators

$$\hat{Z}^{(0)}_3 \to \frac{c^2}{\omega^2}\nabla\times(\nabla\times) - 1, \tag{28}$$

$$\hat{Y}^{(0)}_3 = \hat{Z}^{(0)-1}_3 \to \mathbb{G}^{\text{vac}} \tag{29}$$

correspond to the vacuum Maxwell partial differential operator and associated Green's function. Finally, in the first, second, and third equations, the coupling to the third component,

i.e., the vacuum EM field, corresponds to

$$\Delta\hat{Z}_{\alpha,3} \to -\sum_a q_{\alpha a}\delta_{ij}\delta^3(\mathbf{x}-\mathbf{r}_{\alpha a}), \tag{30}$$

which is the convolution operator representing the charge density of point dipoles constituting each polarizable body (with a sign flip due to the convention chosen for the general linear response formulas): these coupling operators are real-valued reciprocal operators, as evinced in the equations of motion. This also means that for $\alpha \in \{1,2\}$, the material response operators may be written in position space as

$$\begin{aligned}\Delta\hat{Z}_{3,\alpha}\hat{Y}^{(0)}_{\alpha}\Delta\hat{Z}_{\alpha,3} &\to \mathbb{V}_{\alpha ij}(\omega,\mathbf{x},\mathbf{x}') \\ &= \sum_{a,a'} q_{\alpha a}[(K_{\alpha\alpha}-\omega^2 M_{\alpha})^{-1}]_{ai,a'j}q_{\alpha a'} \\ &\quad\times \delta^3(\mathbf{x}-\mathbf{r}_{\alpha a})\delta^3(\mathbf{x}'-\mathbf{r}_{\alpha a'}),\end{aligned} \tag{31}$$

which is exactly the susceptibility $\mathbb{V}_\alpha$ of a collection of point dipolar harmonic oscillators, while

$$\hat{Y}_3 \to (\mathbb{G}^{\mathrm{vac}-1}-\mathbb{V}_1-\mathbb{V}_2)^{-1} = \mathbb{G} \tag{32}$$

is exactly the Maxwell Green's function in the presence of susceptibilities $\hat{\chi}_\alpha$. Thus, the heat transfer between the two polarizable bodies can be written as

$$\Phi(\omega) = 4\,\mathrm{Tr}[\mathrm{asym}(\mathbb{V}_1)\mathbb{G}^{\dagger}\,\mathrm{asym}(\mathbb{V}_2)\mathbb{G}] \tag{33}$$

which exactly matches the fluctuational EM expression [65]. Additionally, the assumptions underlying the derivation of the upper bound on the transmission eigenvalues hold here, so those derivations remain valid in this context: the coupling operators representing the negative charge densities and real valued and reciprocal, and $\mathrm{asym}(\mathbb{G}^{\mathrm{vac}-1}) = 0$ comes from the properties of Maxwell's equations, while the fact that $\mathrm{asym}(\mathbb{G}^{\mathrm{vac}})$ does not vanish due to free space supporting outward propagation of EM energy is irrelevant to those particular derivations. Thus, the general NEGF formalism for heat transfer in linear response systems can be exactly mapped to the specific fluctuational EM formalism for linear RHT.

Physically, the harmonic oscillators may represent valence electrons or nuclei dressed by inner-shell electrons, and the couplings, namely the effective charges, along with the effective masses and spring constants are again computed via density functional theory. We again stress that the correspondences $\Delta\hat{Z}_{3,\alpha}\hat{Y}^{(0)}_{\alpha}\Delta\hat{Z}_{\alpha,3} \to \mathbb{V}_{\alpha ij}(\omega,\mathbf{x},\mathbf{x}')$ for RHT are generally applicable even for more than two polarizable bodies coupled to the vacuum EM field, which allows more general scenarios for PCHT to be treated using (4) [9,61]. Furthermore, the derivation of Landauer-like formulas for RHT (33) is generally applicable for linear media even when the susceptibilities $\mathbb{V}_\alpha$ do not describe harmonic oscillator response functions; our use of harmonic oscillators was for convenience in writing a Lagrangian and explaining salient features through physical intuition. Finally, we emphasize that unlike previous work which has typically depended on high-symmetry geometries and the assumption of the EM near-field regime [39,40,42,43,45], these derivations are applicable to arbitrary geometries from the near- through far-field regimes.

## IV. COMPARISONS BETWEEN PCHT AND RHT

Before proceeding, it is useful to summarize the comparisons between PCHT and RHT specifically focusing on the case of two bodies interacting through a third component (either a third body for PCHT or the EM field for RHT), an analogy which is summarized Table I and illustrated schematically in Fig. 1. While the basic formalisms are essentially identical and both obey the same upper bounds, in what follows we emphasize a few of the distinctions.

The typical situation considered for PCHT involves two semi-infinite leads connected by a much smaller molecular junction or interfacial region. As a result, when mapping $\hat{Y}^{(0)}_{\alpha} \to g_\alpha$ for $\alpha \in \{1,2\}$, even though the microscopic oscillators have no dissipation so $\mathrm{asym}(g_\alpha^{-1}) \to 0$, the fact that the leads are semi-infinite and act as thermodynamic reservoirs means $\mathrm{asym}(g_\alpha) \neq 0$: this represents loss of energy through far-field propagation of phonons into the bulks of the leads. Meanwhile, when mapping $\hat{Y}^{(0)}_3 \to g_3$ for the junction or interfacial region, the compactness of that intermediate body precludes dissipation through far-field propagation of phonons, so not only is it true that $\mathrm{asym}(g_3^{-1}) \to 0$ but it is also true that $\mathrm{asym}(g_3) \to 0$. Moreover, the smallness of the intermediate body means that it is typically easier to evaluate the matrix products and inverses in the space of the intermediate body through (14). The situation is flipped for RHT, where typically energy exchange is considered between two compact bodies via EM fields that propagate through all of space. As a result, when mapping the response of lossless oscillators constituting each polarizable body in the mapping $\Delta\hat{Z}_{3,\alpha}\hat{Y}^{(0)}_{\alpha}\Delta\hat{Z}_{\alpha,3} \to \mathbb{V}_\alpha$ for compact bodies $\alpha \in \{1,2\}$, taking literally the lack of dissipation would strictly imply that $\mathrm{asym}(\mathbb{V}_\alpha) \to 0$, so heat transfer and other fluctuational EM phenomena would not exist. Realistically, these atomic oscillators are not perfectly lossless but are subject to losses through scattering and propagation of energy, which we do not consider here; this can be accounted for by properly including reservoir DOFs in the Lagrangian and performing

TABLE I. Comparison of components and relevant linear response quantities between PCHT and RHT within our NEGF heat-transfer formalism.

| Heat transfer mechanism | Phonons | Photons |
|---|---|---|
| Components 1, 2 | Infinite reservoirs (leads) | Polarizable bodies |
| Component 3 | Compact central device | Vacuum EM field (all space) |
| $\hat{Y}^{(0)}_{\alpha}$: $\alpha \in \{1,2\}$ | Uncoupled lead mechanical Green's function | Susceptibilities $\mathbb{V}_\alpha$ |
| $\Delta\hat{Z}_{\alpha,3}$: $\alpha \in \{1,2\}$ | Interface lead/device harmonic couplings | All atom charges |
| $\hat{Y}_3$ | Coupled device mechanical Green's function | Maxwell Green's function $(\mathbb{G}^{\mathrm{vac}-1}-\mathbb{V}_1-\mathbb{V}_2)^{-1}$ |

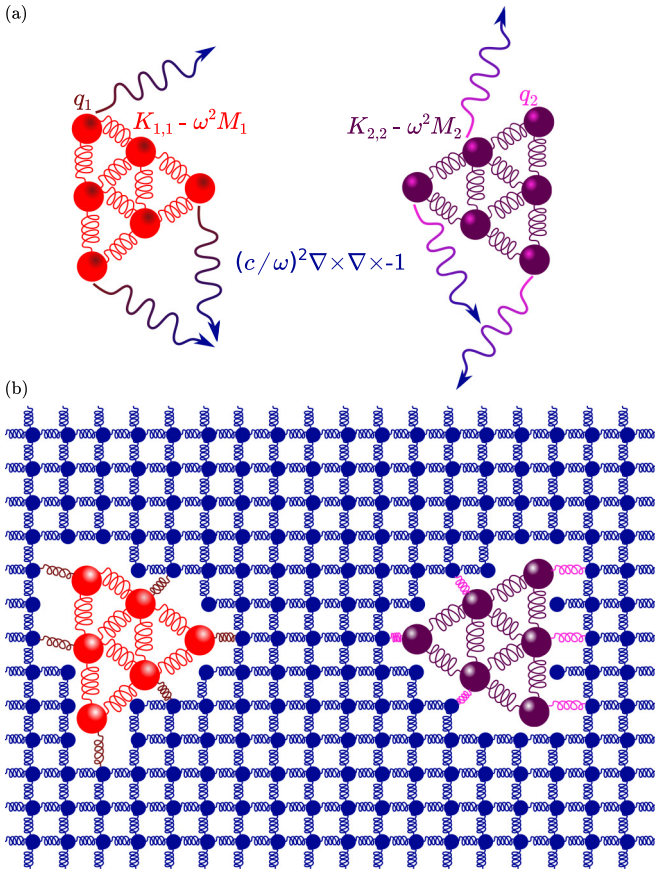


FIG. 1. Photon radiation. (a) Radiation of energy in free space between compact polarizable bodies. (b) Analogous situation for conduction: Compact phononic devices are coupled at each atom to an infinite lattice supporting propagation of phonons infinitely far away.

some renormalization like decimation as in the phonon case for a physically motivated reservoir, or more typically by phenomenologically adding an appropriate small imaginary part to some part of $\mathbb{V}_\alpha$.

Meanwhile, when mapping $\hat{Y}_3^{(0)} \to \mathbb{G}^{\text{vac}}$ through all of space, while it is true that $\text{asym}(\mathbb{G}^{\text{vac}-1}) \to 0$ allows the same Landauer bounds to hold for RHT as for PCHT, the ability of free space to support outward propagation of EM energy also means $\text{asym}(\mathbb{G}^{\text{vac}}) \neq 0$. Moreover, the fact that the polarizable bodies occupy compact regions in space (as opposed to all of space) means that it is typically easier to evaluate the matrix products and inverses in the spaces of the polarizable bodies through (10). In particular, by using the operator correspondences from the previous section and linking (14) to a special case of (10) as above, it can be shown that (10) exactly reproduces the T-operator formula for RHT [33]. Along these lines we finally note that in PCHT, the off-diagonal block of the Green's function of component 3 in isolation connecting the respective atoms coupled to each of the other components, which may be denoted $P_{3(2)}g_3P_{3(1)}$, has a size, and therefore a maximum rank, that scales as the surface areas of component 3 coupled with each of the other components. For the case of RHT, the analogous quantity is $\mathbb{P}_2\mathbb{G}^{\text{vac}}\mathbb{P}_1$, where $\mathbb{P}_\alpha$ is the projection operator onto the volume of body $\alpha$: this seems to contrast with the dependence on surface area for PCHT. However, the EM surface equivalence theorem [31,32,68–71] shows that the fields radiated by any volumetric polarization distribution to the exterior of some fictitious bounding surface can be exactly reproduced in that exterior region by an equivalent surface current distribution, which therefore suggests that the rank of $\mathbb{P}_2\mathbb{G}^{\text{vac}}\mathbb{P}_1$ actually scales with the *surface* of each body, thereby producing a similar result as for mechanical waves. The underlying physical reasons are a little different: the general boundary conditions of EM fields at material interfaces for radiation contrast with the specific form of coupling of nearest-neighbor atoms for phonon propagation. That said, the similarities can be intuitively understood as arising from the similar physics governing mechanical wave propagation through homogeneous media as EM wave propagation through vacuum or homogeneous media: the spring constant matrix $K$ governing mechanical wave propagation through a medium is essentially a discrete-space analog of the $\nabla\times(\nabla\times)$ operator governing EM wave propagation, and both of these operators are then equated to double time derivatives of the corresponding field quantities. Finally, we note that in the concluding remarks, we connect this paper to an accompanying paper [72] that leverages this generic NEGF formalism to generalize recent bounds on RHT [73,74] to include PCHT: we point out that these bounds rely heavily on the singular values of the off-diagonal blocks $\mathbb{P}_2\mathbb{G}^{\text{vac}}\mathbb{P}_1$ in the case of RHT, or $P_{3(2)}g_3P_{3(1)}$ in the case of PCHT.

## V. UNIFYING PCHT AND RHT

At nanometric and smaller separations, we expect that both PCHT and RHT could exhibit comparable contributions to overall heat transfer between two material bodies, whether through approach to direct contact or through contact with an intermediate junction [14,15,23,35–37]. Thus motivated, we use this section to present a method for unifying both forms of heat transfer in both of these scenarios. This method is based on the retarded many-body (RMB) framework of mesoscale fluctuational EM [61–63], allowing for accurate modeling of fluctuational EM phenomena, including RHT, in atom-scale systems.

Each body $\alpha \in \{1, 2, 3\}$ comprises $N_\alpha$ atoms labeled $a, b, c$. Each atom is centered at an equilibrium position $\mathbf{r}_{\alpha a}$ and has an effective nuclear oscillator of mass $m_{\text{I}\alpha a}$ which couples to other nuclear oscillators within the same body and which may couple to nuclear oscillators in other bodies at interfaces: these couplings are encoded in the matrices $K_{\text{I}\alpha\alpha}$ within the same body and $K_{\text{I}\alpha\beta}$ between different bodies, where the former has dimension $3N_\alpha \times 3N_\alpha$ while the latter has dimension $3N_\alpha \times 3N_\beta$. The effective nuclear oscillator in each atom is also coupled to an effective valence electronic oscillator of mass $m_{\text{e}\alpha a}$ through an isotropic spring constant $k_{\text{e}\alpha a}$. The valence electronic oscillators couple as point charges to the vacuum EM field via the charge $q_{\text{e}\alpha a}$; these electrons along with the inner electrons screen the nuclei, so we model the nuclear oscillators as having no direct coupling to the EM field. The displacements of the effective valence electronic oscillators are labeled $x_{\text{e}\alpha ai}$, while those of the nuclear oscillators are labeled $x_{\text{I}\alpha ai}$, for Cartesian direction $i$. We collect the

displacements into $3N_\alpha$-dimensional vectors $x_{e\alpha}$ and $x_{I\alpha}$, and the masses, charges, and valence electronic spring couplings into diagonal $3N_\alpha \times 3N_\alpha$ matrices $M_{e\alpha}$, $M_{I\alpha}$, $Q_{e\alpha}$, and $K_{e\alpha}$. Additionally, the electric field in vacuum must be evaluated at each equilibrium position $\mathbf{r}_{\alpha a}$ when entering the equations of motion for the effective valence electronic oscillators, so we collect the $N_\alpha$ Cartesian vectors $\mathbf{E}(\mathbf{r}_{\alpha a})$ into the $3N_\alpha$-dimensional vector $e_{e\alpha}$.

For two bodies coming into direct conductive contact (with no third intermediate material body present) and interacting via the vacuum EM field, we may use the above matrix notation to write the equations of motion as

$$\begin{aligned}
&(K_{e\alpha} - \omega^2 M_{e\alpha})x_{e\alpha} - K_{e\alpha}x_{I\alpha} - Q_{e\alpha}e_{e\alpha} \\
&\quad = [(K_{e\alpha} - \omega^2 M_{e\alpha})x_{e\alpha} - K_{e\alpha}x_{I\alpha}]\delta_{\alpha,1}, \\
&(K_{e\alpha} - \omega^2 M_{I\alpha})x_{I\alpha} + \sum_\beta K_{I\alpha\beta}x_{I\beta} - K_{e\alpha}x_{e\alpha} = 0, \\
&\left(\frac{c^2}{\omega^2}\nabla\times(\nabla\times) - 1\right)\mathbf{E} = \sum_{\alpha,a} q_{e\alpha a}\mathbf{x}_{e\alpha a}\delta^3(\mathbf{x} - \mathbf{r}_{\alpha a})
\end{aligned} \tag{34}$$

for each $\alpha, \beta \in \{1, 2\}$, $a \in \{1, \ldots, N_\alpha\}$, and $i \in \{x, y, z\}$, for sources in body 1. We may then formally solve the final equation and eliminate $e_{e\alpha}$ in favor of $x_{e\alpha}$ and $x_{I\alpha}$, yielding the equations of motion

$$\begin{aligned}
&(K_{e\alpha} - \omega^2 M_{e\alpha})x_{e\alpha} - K_{e\alpha}x_{I\alpha} - \sum_\beta Q_{e\alpha}G^{\text{vac}}_{\alpha\beta}Q_{e\beta}x_{e\beta} \\
&\quad = \left[(K_{e\alpha} - \omega^2 M_{e\alpha})x^{(0)}_{e\alpha} - K_{e\alpha}x^{(0)}_{I\alpha}\right]\delta_{\alpha,1}, \\
&(K_{e\alpha} - \omega^2 M_{I\alpha})x_{I\alpha} + \sum_\beta K_{I\alpha,\beta}x_{I\beta} - K_{e\alpha}x_{e\alpha} = 0,
\end{aligned} \tag{35}$$

where $G^{\text{vac}}_{\alpha\beta}$ is the $3N_\alpha \times 3N_\beta$ matrix whose elements are $G^{\text{vac}}_{ij}(\omega, \mathbf{r}_{\alpha a}, \mathbf{r}_{\beta b})$ for each pair of atomic coordinates. Hence, we identify the relevant operators as $2 \times 2$ block matrices

$$\begin{aligned}
\hat{Z}^{(0)}_\alpha &\to \begin{bmatrix} K_{e\alpha} - \omega^2 M_{e\alpha} & -K_{e\alpha} \\ -K_{e\alpha} & K_{e\alpha} + K_{I\alpha\alpha} - \omega^2 M_{I\alpha} \end{bmatrix}, \\
\Delta\hat{Z}_{\alpha\beta} &\to \begin{bmatrix} -Q_{e\alpha}G^{\text{vac}}_{\alpha\beta}Q_{e\beta} & 0 \\ 0 & K_{I\alpha\beta}(1 - \delta_{\alpha\beta}) \end{bmatrix},
\end{aligned} \tag{36}$$

where the top row and left column blocks represent the effective valence electronic DOFs, while the bottom row and right column blocks represent the effective nuclear degrees of freedom. Strictly speaking, the matrices $-\omega^2 M_{e\alpha}$ and $-\omega^2 M_{I\alpha}$ should respectively be replaced by $-i\omega B_{e\alpha} - \omega^2 M_{e\alpha}$ and $-i\omega B_{I\alpha} - \omega^2 M_{I\alpha}$ in order to account for nonzero dissipation, though the dissipation matrices $B_{e\alpha}$ and $B_{I\alpha}$ may be taken to be infinitesimal; also, once again, the diagonal blocks $K_{I\alpha\alpha}$ entering $\hat{Z}^{(0)}_\alpha$ should actually include the effects of couplings to nuclear oscillators in other bodies as are present in the off-diagonal blocks $K_{I\alpha\beta}$ for all $\beta \neq \alpha$. With details explained in [61,63], the RMB oscillator matrix parameters $Q_{e\alpha}$, $M_{e\alpha}$, $M_{I\alpha}$, $K_{e\alpha}$, and $K_{I\alpha\alpha}$ (the latter initially excluding couplings to nuclear oscillators in other bodies) along with the equilibrium atomic positions are all computed using density functional theory (DFT) for each body in isolation, while the matrices $B_{e\alpha}$ and $B_{I\alpha}$ are assigned phenomenological values. These $2 \times 2$ block matrices can then be used in place of $\hat{Z}^{(0)}_\alpha$ and $\Delta\hat{Z}_{\alpha\beta}$ in the formula for two components with general couplings (10) to find the combined heat transfer including PCHT and RHT: the couplings among valence electronic and nuclear DOFs through EM fields means that PCHT and RHT contributions are not separable, but in fact affect each other [8,42,43].

For two bodies whose nuclear coordinates are coupled only to a third intermediate body, which also has nuclear and valence electronic DOFs, in which all electronic coordinates are coupled to the EM field, the formalism is similar to above. In particular, the formulas in (34) still hold for all bodies $\alpha, \beta \in \{1, 2, 3\}$, although $K_{I1,3}$ and $K_{I2,3}$ and their transposes are the only nonzero off-diagonal blocks of $K_I$. With that caveat in mind, this further means that (35) and the correspondences in (36) holds as well for all bodies $\alpha, \beta \in \{1, 2, 3\}$. That said, the fact that $\Delta\hat{Z}_{\alpha\beta}$ has nonzero blocks for all $(\alpha, \beta)$ means that (14) cannot be used. Instead, the more general formula (4) for the energy transfer spectrum must be used, plugging the $2 \times 2$ block matrices in (36) into the overall $3 \times 3$ block matrices

$$\hat{Z}^{(0)} = \begin{bmatrix} \hat{Z}^{(0)}_1 & 0 & 0 \\ 0 & \hat{Z}^{(0)}_2 & 0 \\ 0 & 0 & \hat{Z}^{(0)}_3 \end{bmatrix}, \tag{37}$$

$$\Delta\hat{Z} = \begin{bmatrix} \Delta\hat{Z}_{1,1} & \Delta\hat{Z}_{1,2} & \Delta\hat{Z}_{1,3} \\ \Delta\hat{Z}_{2,1} & \Delta\hat{Z}_{2,2} & \Delta\hat{Z}_{2,3} \\ \Delta\hat{Z}_{3,1} & \Delta\hat{Z}_{3,2} & \Delta\hat{Z}_{3,3} \end{bmatrix} \tag{38}$$

to evaluate (4).

These formulas for the energy transfer spectrum and associated linear response operators are thus the application of the general NEGF formalism for combined PCHT and RHT. In contrast to the derivations of pure RHT which ultimately do not depend on the form of the susceptibilities $\mathbb{V}_\alpha$ as long as it is linear, these particular derivations do depend on the harmonicity of the material models, though they may be generalizable through a more complicated formalism. However, beyond that approximation as well as the assumptions regarding material dissipation, these formulas are independent of specific geometries and material properties, and can be evaluated in the EM near- or far-field regimes. Additionally, we point out that unlike previous works which have cast formulas for combined electronic conduction and RHT in a more complicated (Meir-Wingreen) form rather than the typical Landauer/Caroli form [42,43,45] as electrons and photons obey different quantum statistics, no such complication arises here because phonons and photons obey the same statistics.

We apply this unified formalism to an illustrative model of heat transfer between two collinear 250 atom-long atomically thin wires, taken to be made of carbon (i.e., carbyne wires), and particularly compute the heat transfer coefficient $\frac{\partial P}{\partial T}$ at room temperature ($T = 300$ K). Specifically, we compute the heat transfer coefficient $\frac{\partial P_{\text{both}}}{\partial T}$ by calculating the Landauer energy transfer spectrum $\Phi_{\text{both}}$ arising from plugging (36) as written into (4), $\frac{\partial P_{\text{rad}}}{\partial T}$ by computing $\Phi_{\text{rad}}$ arising from plugging (36) with $K_{I\alpha\beta} = 0$ for $\beta \neq \alpha$ (so $K_{I\alpha\alpha}$ refers only to the spring constant matrices among nuclei for each body in isolation) into (4), and $\frac{\partial P_{\text{cond}}}{\partial T}$ by computing $\Phi_{\text{cond}}$ arising from plugging (36) with $G^{\text{vac}} = 0$ for all pairs of electronic

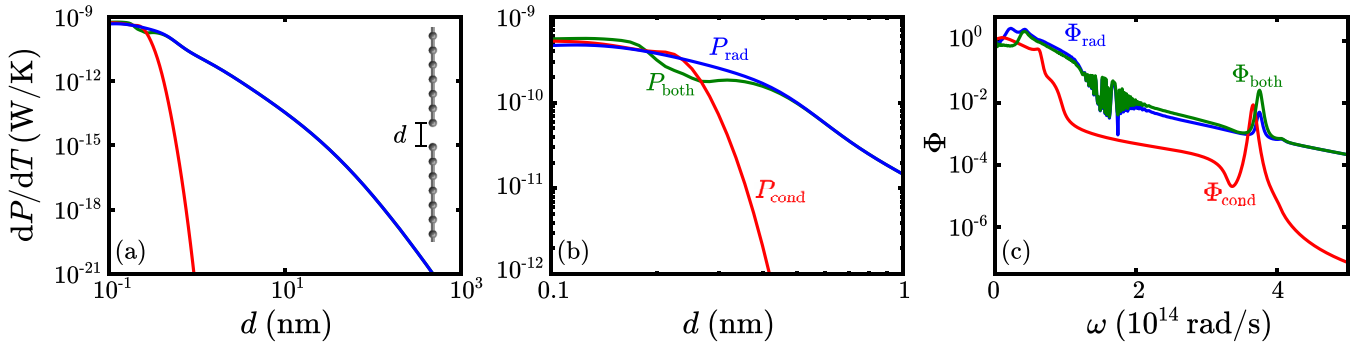


FIG. 2. Conduction and radiation between collinear wires. (a) Heat transfer coefficient $\frac{\partial P}{\partial T}$ at room temperature ($T = 300$ K) between two collinear 250 atom-long carbyne wires in vacuum, comparing the cases when heat transfer is due purely to radiation (blue) or conduction (red) versus both together (green). (b) Same as (a) zoomed in for $d \in [0.1\text{ nm}, 1\text{ nm}]$. (c) Landauer energy transfer spectrum $\Phi$ (independent of $T$) for $d = 0.281$ nm, clearly demonstrating the existence of nontrivial resonances.

oscillators into (4); in all cases, $\Phi$ refers to $\Phi_2^{(1)}$. Within each body, as described above, the charges, masses, and spring constants are all taken from DFT evaluated for each body in isolation, the matrix elements of the Maxwell Green's function $\mathbb{G}^{\text{vac}}$ are evaluated in a Gaussian basis to mitigate short-range EM divergences [61,62,75], and the dissipation matrices are chosen such that $B_e = \gamma_e M_e$ and $B_I = \gamma_I M_I$ hold with $\gamma_e = 10^{11}$ s$^{-1}$ and $\gamma_I = 10^{13}$ s$^{-1}$; the damping rates are chosen phenomenologically to be large enough to allow reasonably coarse frequency sampling, but small compared to the characteristic frequencies of the relevant polaritons. For computational simplicity, these properties are not recomputed as functions of the separation between the bodies, but while we expect such recomputation to yield significantly different results due to the greater probability of supporting longer-wavelength collective electronic and phononic waves when the wires are in proximity, such recomputation could in principle be performed consistently with this formalism. Likewise, for computational simplicity, the off-diagonal blocks of $K_I$ for each body (including both electronic and nuclear oscillator coordinates) have only the couplings between each end atom nearest to the other molecule be nonzero, and these are modeled via the Morse potential, but this could be further generalized in future work. The Morse potential spring constant for a bond of length $r$ compared to equilibrium length $a_0$ is computed as $k(r) = -\frac{1}{r-a_0}\frac{\partial U_{\text{Morse}}}{\partial r}$, where the potential energy $U_{\text{Morse}}(r) = U_{\text{min}}(1 - e^{-\sqrt{k_0/(2U_{\text{min}})}(r-a_0)})^2$ exhibits a harmonic well of depth $U_{\text{min}}$ and curvature defined by the equilibrium spring constant $k(a_0) = k_0$, all of which are empirical parameters, and exponentially decays as $r \gg a_0$.

As can be seen in Figs. 2(a) and 2(b), many interesting features arise from the coupling of conductive and radiative processes. The exponential decay of the Morse potential with distance means that for $d > 0.4$ nm, conduction ceases to have any meaningful effect on the heat transfer, and the total heat transfer aligns with that of pure radiation. However, for decreasing $d \leqslant 0.4$ nm, not only does conduction become more significant, but the total heat transfer including both radiative and conductive processes falls *below* the corresponding individual cases, and only rises above both for $d < 0.2$ nm before all three powers saturate. Therefore, this unified formalism is clearly necessary for subnanometric separations, as the total power including both PCHT and RHT is not simply the sum of the individual contributions (as has been found in related systems involving electronic conduction [42]), but behaves in a much more complicated way.

In Fig. 2(c) the Landauer energy transfer spectra $\Phi$ make clear that for small enough $d$ where conduction is nontrivial (plotted for $d = 0.281$ nm), the conduction spectrum only has nontrivial contributions at lower frequencies $\omega < 10^{14}$ rad/s. Meanwhile, the total spectrum rises above the radiation spectrum for larger $\omega$ but falls below for smaller $\omega$: the latter is more relevant given the exponential decay of $\frac{\partial \Pi(\omega,T)}{\partial T}$ with $\omega$, leading to $P_{\text{both}} < P_{\text{rad}}$ there. Ultimately, this occurs due to the confluence of EM screening as captured by the Gaussian basis functions along with shifts in the response due to conductive coupling between nuclei of the two different wires: not only does this shift the frequencies of resonances in the Landauer energy transfer spectra, but it can also suppress the resulting amplitudes. This therefore makes clear that the existence of situations where $P_{\text{both}}$ (or its derivative with respect to $T$) falls between or below $P_{\text{cond}}$ or $P_{\text{rad}}$ is not simply a fluke arising from a particular choice of $T$: $\Phi$ is independent of $T$, yet the spectrum $\Phi_{\text{both}}$, far from being a simple case of superimposing $\Phi_{\text{rad}}$ on $\Phi_{\text{cond}}$, shows a delicate interplay among radiative and conductive effects in creating new hybrid resonances. Our calculations are meant to be qualitatively illustrative of the complexities of heat transfer when both conduction and radiation contribute: they are not meant to be quantitatively predictive given the practical limitations in recomputing relevant oscillator parameters at each separation, but we stress that these limitations are not fundamental to the formalism we have presented.

## VI. CONCLUDING REMARKS

We have demonstrated a general NEGF formulation of heat transfer applicable to a wide variety of bosonic systems. This NEGF framework is general enough to explain the salient features of PCHT and RHT separately, show how upper bounds on PCHT can be generalized and then applied to RHT, and demonstrate how to unify PCHT and RHT in situations when both are strongly coupled and relevant. The latter is

particularly relevant at atomistic scales or separations, when continuum material models begin to fail and the net heat transfer is no longer simply the sum of individual radiative or phononic contributions. We stress that our approach is general enough to treat semiclassical heat transfer through other massless bosonic excitations, not just photons or phonons. Moreover, while our analysis of combined PCHT and RHT focused on effective valence electronic and nuclear response as being represented by coupled harmonic oscillators, more complicated linear response models could be considered as well, which we leave for future work. We expect this framework to pave the way for future works investigating the conjunction of PCHT and RHT in complex geometries, particularly at separations where each is relevant and where recent experiments have raised questions about where each form of heat transfer is dominant [35–37,41]. We further point out in Appendix C that this general formalism can, with appropriate changes, be applied to electron CHT (ECHT) by itself, but unifying ECHT with PCHT or RHT is theoretically much more challenging, so we leave that for future work.

In an accompanying paper [72] we generalize bounds previously derived for RHT [73,74] using the generic NEGF formalism for heat transfer in linear systems presented in this paper. We particularly apply such bounds to PCHT, showing that channel-based bounds on PCHT can be much tighter than the Landauer limits of unity [7,8,28].


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## APPENDIX A: BLOCK MATRIX INVERSION FOR TWO COMPONENTS COUPLED DIRECTLY

To derive (10) from (4), the two block matrices of interest are $\hat{Y}\hat{P}_1$ and $\mathrm{asym}(\hat{P}_2\Delta\hat{Z})$, of which the first requires inversion of a block matrix. In particular, we can immediately evaluate

$$\mathrm{asym}(\hat{P}_2\Delta\hat{Z}) = \begin{bmatrix} 0 & -\frac{1}{2i}\Delta\hat{Z}^{\star}_{1,2} \\ \frac{1}{2i}\Delta\hat{Z}_{2,1} & \mathrm{Im}(\Delta\hat{Z}_{2,2}) \end{bmatrix} \tag{A1}$$

in block form. Meanwhile, standard formulas for inversion of a block matrix yield

$$\hat{Y}\hat{P}_1 = \begin{bmatrix} \left[\hat{1} - \left(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\right)^{-1}\Delta\hat{Z}_{1,2}\left(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\right)^{-1}\Delta\hat{Z}_{2,1}\right]^{-1}\left(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\right)^{-1} \\ -\left(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\right)^{-1}\Delta\hat{Z}_{2,1}\left[\hat{1} - \left(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\right)^{-1}\Delta\hat{Z}_{1,2}\left(\hat{Z}_2^{(0)} + \Delta\hat{Z}_{2,2}\right)^{-1}\Delta\hat{Z}_{2,1}\right]^{-1}\left(\hat{Z}_1^{(0)} + \Delta\hat{Z}_{1,1}\right)^{-1} \end{bmatrix}, \tag{A2}$$

where multiplication on the right by $\hat{P}_1$ allows for picking out only the left column block. Putting everything together at this stage yields the result in the main text.

## APPENDIX B: BLOCK MATRIX INVERSION FOR TWO COMPONENTS COUPLED ONLY TO A THIRD

To derive (14) from (4), the two block matrices of interest are $\hat{Y}\hat{P}_1$ and $\mathrm{asym}(\hat{P}_2\Delta\hat{Z})$, of which the first requires inversion of a block matrix. To invert the $3\times 3$ block matrices, we exploit the fact that there are no couplings directly between components 1 and 2. This allows for defining subblocks such that

$$\hat{Z}^{(0)} = \begin{bmatrix} \hat{Z}_{\mathrm{A}}^{(0)} & 0 \\ 0 & \hat{Z}_3^{(0)} \end{bmatrix}, \tag{B1}$$

$$\Delta\hat{Z} = \begin{bmatrix} 0 & \Delta\hat{Z}_{\mathrm{A},3} \\ \Delta\hat{Z}_{3,\mathrm{A}} & 0 \end{bmatrix}, \tag{B2}$$

where aggregate operators for bodies 1 and 2 are defined as

$$\hat{Z}_{\mathrm{A}}^{(0)} = \begin{bmatrix} \hat{Z}_1^{(0)} & 0 \\ 0 & \hat{Z}_2^{(0)} \end{bmatrix}, \tag{B3}$$

$$\Delta\hat{Z}_{\mathrm{A},3} = \begin{bmatrix} \Delta\hat{Z}_{1,3} \\ \Delta\hat{Z}_{2,3} \end{bmatrix}, \tag{B4}$$

$$\Delta\hat{Z}_{3,\mathrm{A}} = \Delta\hat{Z}_{\mathrm{A},3}^{\top} \tag{B5}$$

for this system. Because components 1 and 2 lie in the top row and left column blocks of these new $2\times 2$ block matrices, and because $\hat{Z}^{(0)}$ is block-diagonal in this 2-by-2 aggregate block representation as well, then $\hat{Z}^{(0)}\hat{P}_1 = \hat{P}_1\hat{Z}^{(0)}\hat{P}_1$, so the energy transfer spectrum $\Phi = 4\mathrm{Tr}[\hat{P}_1\,\mathrm{asym}(\hat{Y}^{(0)})\hat{P}_1\hat{Z}^{(0)\dagger}\hat{Y}^{\dagger}\,\mathrm{asym}(\hat{P}_2\Delta\hat{Z})\hat{Y}\hat{Z}^{(0)}\hat{P}_1]$ can be computed by computing $\hat{Y}\hat{P}_1$, which is the left block column of $\hat{Y}$, and $\mathrm{asym}(\hat{P}_2\Delta\hat{Z}^{(0)\dagger})$. In particular, if

$$\hat{Y} = \begin{bmatrix} \hat{Z}_{\mathrm{A}}^{(0)} & \Delta\hat{Z}_{\mathrm{A},3} \\ \Delta\hat{Z}_{3,\mathrm{A}} & \hat{Z}_3^{(0)} \end{bmatrix}^{-1}, \tag{B6}$$

then

$$\hat{Y}\hat{P}_1 = \begin{bmatrix} \hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 + \hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\left(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\right)^{-1}\Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 \\ -\left(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\right)^{-1}\Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 \end{bmatrix}, \tag{B7}$$

where $\hat{Y}_{\mathrm{A}}^{(0)} = \hat{Z}_{\mathrm{A}}^{(0)-1}$, while

$$\mathrm{asym}(\hat{P}_2\Delta\hat{Z}) = \frac{1}{2i}\begin{bmatrix} 0 & \hat{P}_2\Delta\hat{Z}_{\mathrm{A},3} \\ -\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{P}_2 & 0 \end{bmatrix} \tag{B8}$$

is the expression in the block basis. Carrying out the operator product yields the complicated expression

$$\begin{aligned}
&\hat{P}_1\hat{Y}^{\dagger}\,\mathrm{asym}(\hat{P}_2\Delta\hat{Z})\hat{Y}\hat{P}_1 \\
&= \frac{1}{2i}\big(-\hat{P}_1\hat{Y}_{\mathrm{A}}^{(0)\dagger}\hat{P}_2\Delta\hat{Z}_{\mathrm{A},3}\big(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\big)^{-1}\Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 \\
&\quad - \hat{P}_1\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\big(\hat{Z}_3^{(0)\dagger} - \Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{3,\mathrm{A}}^{\dagger}\big)^{-1}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{Y}_{\mathrm{A}}^{(0)\dagger}\hat{P}_2\Delta\hat{Z}_{\mathrm{A},3}\big(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\big)^{-1}\Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 \\
&\quad + \hat{P}_1\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\big(\hat{Z}_3^{(0)\dagger} - \Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{3,\mathrm{A}}^{\dagger}\big)^{-1}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{P}_2\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\big(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\big)^{-1}\Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 \\
&\quad + \hat{P}_1\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\big(\hat{Z}_3^{(0)\dagger} - \Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{3,\mathrm{A}}^{\dagger}\big)^{-1}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{P}_2\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1\big),
\end{aligned} \tag{B9}$$

but this can be simplified as follows. The term $\hat{P}_2\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1 = 0$ (and the same is true of its Hermitian adjoint) because $\hat{Y}_{\mathrm{A}}^{(0)}$ is block-diagonal, with no correlations between objects 1 and 2. This therefore simplifies the expression above to

$$\hat{P}_1\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\big(\hat{Z}_3^{(0)\dagger} - \Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\hat{Y}_{\mathrm{A}}^{(0)\dagger}\Delta\hat{Z}_{3,\mathrm{A}}^{\dagger}\big)^{-1}\Delta\hat{Z}_{\mathrm{A},3}^{\dagger}\,\mathrm{asym}\left(\hat{P}_2\hat{Y}_{\mathrm{A}}^{(0)}\right)\Delta\hat{Z}_{\mathrm{A},3}\big(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3}\big)^{-1}\Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1$$

for which the block-diagonality of $\hat{Y}_{\mathrm{A}}^{(0)}$ once again allows for writing $\mathrm{asym}(\hat{P}_2\hat{Y}_{\mathrm{A}}^{(0)}) = \hat{P}_2\,\mathrm{asym}(\hat{Y}_2^{(0)})\hat{P}_2$. Additionally, performing the block vector-matrix-vector products within the inverses involving $\hat{Z}_3^{(0)}$ gives $\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,\mathrm{A}}\hat{Y}_{\mathrm{A}}^{(0)}\Delta\hat{Z}_{\mathrm{A},3} = \hat{Z}^{(0),3} - \Delta\hat{Z}_{3,1}\hat{Y}_1^{(0)}\Delta\hat{Z}_{1,3} - \Delta\hat{Z}_{3,2}\hat{Y}_2^{(0)}\Delta\hat{Z}_{2,3}$. The terms on the outside, namely $\hat{Y}_{\mathrm{A}}^{(0)}\hat{P}_1$ and its Hermitian adjoint, can be commuted to yield $\hat{P}_1\hat{Y}_1^{(0)}$ due to the block-diagonal structure, and in the trace expression this is then multiplied on the right by $\hat{Z}^{(0)}\hat{P}_1$, the result of which is simply $\hat{P}_1$ as $\hat{Y}_1^{(0)} = \hat{Z}_1^{(0)-1}$; this acts to the right of $\Delta\hat{Z}_{3,\mathrm{A}}$ to yield $\Delta\hat{Z}_{3,1}$. Putting this all together yields the result in the main text.

## APPENDIX C: ELECTRON CHT

In the main text we did not discuss electron CHT (ECHT) because electrons, being fermions, exhibit fundamentally different statistics from photons or phonons. Simultaneous treatment of ECHT with RHT or PCHT is therefore significantly more challenging than unifying RHT with PCHT. However, if ECHT is considered alone or in conjunction with heat transfer through other fermionic quasiparticles, the general formalism in this paper can, under similar approximations, be applied to such forms of heat transfer. We briefly discuss this for the case of ECHT alone.

Typically, mesoscale ECHT is described in terms of a tight-binding Hamiltonian [67], which can be expressed in terms of its blocks $\hat{H}_{mn}$ for $m, n \in \{1, 2, 3\}$; the diagonal blocks $\hat{H}_{nn}$ describe the tight-binding properties of each component $n$ in isolation, while the off-diagonal blocks $\hat{H}_{mn}$, for which it may be assumed that only $\hat{H}_{n,3} = (\hat{H}_{3,n})^{\dagger}$ are nonzero, describe the tight-binding couplings of the central component, labeled 3, to the other components, labeled 1 and 2. From this, the following correspondences may be observed; we note that in the context of ECHT, it is more common to use the energy $E = \hbar\omega$ instead of the angular frequency $\omega$. We identify $\hat{Y}_n^{(0)} \to \hat{g}_n = (\hat{H}_{nn} - E\,\hat{1}_n)^{-1}$ as the electronic Green's function of each component $n \in \{1, 2, 3\}$ in isolation. Furthermore, we identify $\Delta\hat{Z}_{3,n} \to \hat{H}_{3,n}$ for $n \in \{1, 2\}$; the basis of atomic sites is analogous to real space, and in the basis, it is typically true that the matrix elements of $\hat{H}_{3,n}$ are all real valued, so in this basis, the assumptions $\Delta\hat{Z}_{3,n} = (\Delta\hat{Z}_{3,n})^{\star} = (\Delta\hat{Z}_{n,3})^{\top}$ remain valid. Finally, when considering (14), the terms $\mathrm{asym}(\hat{Y}_n^{(0)}) \to \mathrm{asym}(\hat{g}_n)$ for $n \in \{1, 2\}$, which correspond to the electronic densities of states for each lead, do not vanish even though $\hat{H}_{nn}$ is Hermitian for each $n$ because the leads are large enough that irreversible outward propagation of electron wave functions must be considered, while the terms $\Delta\hat{Z}_{2,3}(\hat{Z}_3^{(0)} - \Delta\hat{Z}_{3,1}\hat{Y}_1^{(0)}\Delta\hat{Z}_{1,3} - \Delta\hat{Z}_{3,2}\hat{Y}_2^{(0)}\Delta\hat{Z}_{2,3})^{-1}\Delta\hat{Z}_{3,1} \to \hat{H}_{2,3}\hat{G}_3\hat{H}_{3,1}$, having defined the full electronic Green's function $\hat{G}_3 = (\hat{H}_{3,3} - E\,\hat{1}_3 - \hat{H}_{3,1}\hat{g}_1\hat{H}_{1,3} - \hat{H}_{3,2}\hat{g}_2\hat{H}_{2,3})^{-1}$ which is related to the density of states in the presence of the leads, correspond to the tight-binding couplings of the junction to the leads dressed by Fabry-Pérot-like multiple scattering terms within the junction accounting for the interfaces with the leads.

As a reminder, applying the equations expressing the total (frequency-integrated) heat transfer power $\dot{W}$ to ECHT requires replacing the Planck function $\Pi(\omega, T)$ with the Fermi-Dirac energy distribution function $(E - \mu)n_{\mathrm{FD}}(E, T) = (E - \mu)/\{\exp[(E - \mu)/(k_{\mathrm{B}}T)] + 1\}$ which depends on the chemical potential $\mu$. This can also be applied to the unification of ECHT with heat transfer from other fermionic particles or quasiparticles.

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