

Charge transfer electron-hole pair excitation and energy transport in hybrid electron-boson nano-junctions

Bing-Zhong Hu, Lei-Lei Nian, and Jing-Tao Lü*

*School of Physics and Wuhan National High Magnetic Field Center,
Huazhong University of Science and Technology, Wuhan 430074, P. R. China*

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We show that a current-carrying coherent electron conductor can be treated as bosonic energy baths involving different types of electron-hole pair excitation. Hybrid energy transport between the nonequilibrium electrons and bosons can be described by a Landauer-Büttiker formula at the lowest order in their coupling. This allows for simple, unified account of a variety of heat transport problems in hybrid electron-boson systems, including non-reciprocal heat transport, thermoelectrical current from a cold-spot and radiative cooling. Our theory paves the way of designing hybrid quantum devices for efficient energy control in the nanoscale.

Introduction.— Understanding nonequilibrium energy transport in nanoscale junctions is of crucial importance both for the fundamental development of quantum thermodynamics and for the practical application of nanoscale thermoelectric and optoelectronic devices. For phase coherent transport, the celebrated Landauer-Büttiker formalism has been successfully applied to study quasi-particle energy transport following different statistics, including electrons[1], photons[2–5], phonons[6–15] and magnons[16]. Wherein, the two baths connected to the system are assumed to be in thermal equilibrium with certain temperature, with the quasi-particle distribution function determined by its statistics, i.e., the Fermi-Dirac distribution for fermions, and the Bose-Einstein distribution for bosons. A difference in the distribution drives an energy current flow between the two thermal baths.

However, the Landauer-Büttiker approach fails to describe energy transport between quasi-particles following different statistics, ubiquitous in thermoelectric and optoelectronic processes of nano-junctions. Examples in molecular transport junctions include electroluminescence[17–20], Joule heating[21–26], current-induced cooling[24, 27–29], radiative cooling[30] and so on. Another difficulty arising in these processes is that the quasi-particles may be in nonequilibrium state due to driving from external bias.

In this work, we show that a voltage-biased coherent electron conductor can be described effectively by different types of bosonic electron-hole pair (EHP) excitation with possibly non-zero chemical potential. This is possible since energy transport between electrons and bosons is always accompanied by the generation or annihilation of different kinds of EHPs[31, 32]. As a result, to the second order in their coupling, energy transport between steady-state nonequilibrium electrons and bosons can be well described by a Landauer-Büttiker formula between bosonic baths with non-zero chemical potentials. Our theory thus generalizes the Landauer-Büttiker formalism to hybrid energy transport between possibly nonequilibrium baths. Furthermore, it provides a unified account of a variety of different thermal, thermoelectric and thermal-optical

processes.

Model.— We consider a model system schematically shown in Fig. 1 (a). The *system* composed of an independent set of bosonic degrees of freedom (DOF) taken as a set of harmonic oscillators. It couples to two kinds of baths. One is an equilibrium boson bath (ph-bath), modeled by an infinite number of harmonic oscillators. The other is an electron bath (e-bath), which itself includes a central part and two electrodes (L and R). The e-bath may be driven into a nonequilibrium steady state by a voltage bias applied between the two electrodes. Without loss of generality, we assume that the system couples only to the central region of the e-bath. Energy transport between the two baths takes place through their simultaneous coupling to the system.

We limit ourselves to non-interaction electrons and weak electron-boson interaction such that a lowest order expansion is valid[33]. Extension to interacting electrons is possible[34]. The electrons couple to the ‘displacement’ of the system harmonic oscillators

$$H_{es} = \sum_{i,j,k} M_{ij}^k c_i^\dagger c_j u_k. \quad (1)$$

Here, M_{ij}^k describes the coupling of the system mode k to the electronic transition between electron states i and j , and u_k is the ‘displacement’ operator of the system mode k . For phonons, it is the displacement, while for photons it is the vector potential. The system-ph-bath coupling is linear between harmonic oscillators and can be treated exactly.

Electron-hole pair excitation.— Our key observation is that the energy transport between the system and the electron bath can be modeled by different kinds of reactions between EHPs in the e-bath and the bosonic modes in the system. The creation and annihilation of the bosonic mode is always accompanied by the recombination and creation of EHPs. These processes can be expressed in the form of reactions

$$e_\alpha + h_\beta \rightleftharpoons b_n, \quad (2)$$

where e_α , h_β and b_n represent electron in electrode α , hole in electrode β and bosonic mode b_n in the system. Equivalently, we can write

$$e_\alpha \rightleftharpoons e_\beta + b_n, \quad (3)$$

representing inelastic electronic transition from electrode α to β , accompanied by emission of bosonic mode n (forward process). The backward direction corresponds to absorption process.

There are four types of EHPs which we label by the spatial location of the electron (α) and hole (β) states. They are schematically shown in Fig. 1 (c) and (d) for recombination and creation processes, respectively. They are denoted by EHP- i ($i = 1, 2, 3, 4$) and are further divided into two groups. The intra-electrode type includes 1/LL and 2/RR, and inter-electrode type includes 3/RL, 4/LR. Additional to energy transfer between e-bath and system, the generation and recombination of inter-electrode EHPs also involves charge transport across the system. We take the energy of mode $\varepsilon_n = \hbar\omega_n$ and that of the EHPs to be positive.

A generalized detailed balance relation applies to each of reactions

$$\frac{T_{\alpha \rightarrow \beta}}{T_{\alpha \leftarrow \beta}} = \exp[-\beta_B(\hbar\Omega - \mu_{\alpha\beta})]. \quad (4)$$

Here, $T_{\alpha \rightarrow \beta}$ and $T_{\alpha \leftarrow \beta}$ are the reaction rates for the forward (boson emission) and backward (boson absorption) processes in Eq. (2), respectively. They are obtained from the Fermi golden rule

$$T_{\alpha \rightarrow \beta} = \frac{2\pi}{\hbar} \sum_{i \in \alpha, f \in \beta} |M_{ij}^m|^2 \delta(\varepsilon_i - \varepsilon_f - \hbar\Omega) \times n_F(\varepsilon_i - \mu_\alpha)(1 - n_F(\varepsilon_f - \mu_\beta)). \quad (5)$$

Here, $n_{F/B}(\varepsilon, T) = [\exp(\beta_B \varepsilon) \pm 1]^{-1}$ the Fermi-Dirac/Bose-Einstein distributions, with $\beta_B = (k_B T)^{-1}$, $\mu_{\alpha\beta} = \mu_\alpha - \mu_\beta$, and $M_{ij}^m = \langle \psi_i(\varepsilon_i) | M | \psi_f(\varepsilon_f) \rangle$ is the transition matrix element from initial state in electrode α to final state in electrode β . The reverse rate $T_{\alpha \leftarrow \beta}$ can be written similarly. Thus, when reaching equilibrium with the EHP bath $\alpha\beta$, the bosonic mode follows a Bose-Einstein distribution at temperature T_e and chemical potential $\mu_{\alpha\beta}$. For intra-electrode processes, $\mu_{\alpha\beta} = 0$, we have the normal detailed balance relation, while for inter-electrode processes $\mu_{\alpha\beta}$ is determined by the applied voltage bias. Thus, the boson mode may acquire a non-zero chemical potential in nonequilibrium. This is consistent with the equilibrium condition for reaction 2.

Non-zero chemical potential of the EHPs indicates that energy and heat are not equivalent anymore for the inter-electrode EHPs. Taking the process 4 in Fig. 1(c) as an example, energy conservation requires $\varepsilon_\alpha = \varepsilon_\beta + \hbar\omega_n$. Due to the finite chemical potential, the heat flowing with the boson is $Q = \hbar\omega_n - \mu_{LR}$. Similarly, the heat flowing

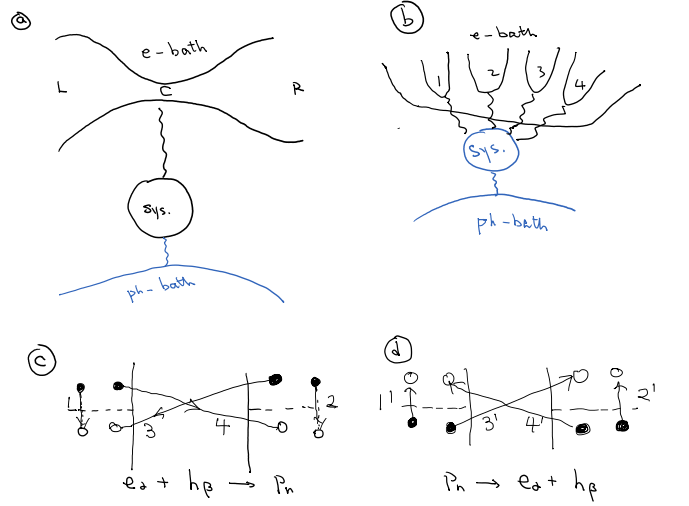


FIG. 1. (a) Schematics of the model we consider. The system consists a set of independent bosonic modes. It couples to an electron bath (e-bath), which is modeled as a conductor including a left (L) and a right (R) electrode, with temperature T_e and chemical potential μ_L and μ_R , respectively. The system further couples to an external thermal bath (ph-bath) at temperature T_{ph} . (b) The electron bath can be treated as four different kinds of electron-hole pair (EHP) baths (1-4), shown in (c). (c-d) Four kinds of EHP recombination (c) and excitation (d) processes. The EHPs are classified according to the spatial location of the electron (e_α) and the hole (h_β). (1,1') Both are at electrode L, (2,2') Both are at electrode R, (3,3') The electron at electrode R and a hole at electrode L. (4,4') The electron at electrode L and a hole at electrode R.

with bosons emitted by process 3 is $Q = \hbar\omega_n - \mu_{RL}$. For $T_e = T_{ph}$ and $\mu_{LR} > 0$, we arrive at a seemingly surprising result: the total energy $\hbar\omega_n$ emitted in process 4, normally termed Joule heat, actually include some chemical work. Even more counter-intuitively, when $\mu_{LR} > \hbar\omega_n$, Q becomes negative, meaning that the emitted boson carries negative entropy, which is a resource for work extraction.

Power spectrum and energy transport.— The key quantity to describe the EHP baths is the coupling-weighted power spectrum. It can be written as

$$\tilde{\Pi}_{mn}^{\alpha\beta}(\omega) = \left[n_B(\hbar\omega - \mu_{\alpha\beta}, T_e) + \frac{1}{2} \right] \Lambda_{mn}^{\alpha\beta}(\omega). \quad (6)$$

We have introduced the coupling-weighted EHP density of states (DOS)

$$\begin{aligned} \Lambda_{mn}^{\alpha\beta}(\omega) &= - \sum_{i \in \alpha, f \in \beta} M_{if}^m M_{fi}^n \delta(\varepsilon_i - \varepsilon_f - \hbar\omega) \\ &\times (n_F(\varepsilon_\alpha - \mu_\alpha, T_\alpha) - n_F(\varepsilon_\beta - \mu_\beta, T_\beta)) \\ &= - \int \frac{d\varepsilon_\alpha}{2\pi} \text{tr}[M^m A_\beta(\varepsilon - \hbar\omega) M^n A_\alpha(\varepsilon_\alpha)] \\ &\times (n_F(\varepsilon_\alpha - \mu_\alpha, T_\alpha) - n_F(\varepsilon_\beta - \mu_\beta, T_\beta)). \end{aligned} \quad (7)$$

Equation (6) follows the normal form of fluctuation-dissipation relation for an equilibrium boson bath, albeit with a possibly non-zero chemical potential $\mu_{\alpha\beta}$. The intra-electrode EHPs ($i=1,2$) are always in equilibrium with $\mu_{\alpha\alpha} = 0$ and temperature T_e . But the two inter-electrode EHPs ($i=3, 4$) have opposite chemical potential $\mu_{RL} = -\mu_{LR}$. They are non-zero when there is a voltage bias applied. To this end, we have shown that the *nonequilibrium* e-bath can be divided into four *equilibrium* EHP baths with different chemical potentials. This effective model is shown in Fig. 1 (b).

Within this effective EHP model, hybrid energy transport between the electrons and the system bosons can be treated as bosonic transport. To the lowest order approximation, we arrive at a Landauer-Büttiker formula for the energy transport from e-bath to the system as a summation of contributions from all the EHP baths

$$J = \sum_{\alpha,\beta} \int_0^{+\infty} \frac{d\omega}{2\pi} \hbar\omega \text{Tr}[\Lambda^{\alpha\beta}(\omega) \mathcal{A}_{ph}(\omega)] \times [n_B(\omega - \mu_{\alpha\beta}, T_e) - n_B(\omega, T_{ph})]. \quad (8)$$

Here, T_e and T_{ph} are the temperature of the e-bath and ph-bath, respectively. The trace Tr is over system DOF, with $\mathcal{A}_{ph} = D^r \Gamma_{ph} D^a$ the spectral function of the system due to coupling to the ph-bath. The summation over $\alpha\beta$ includes contributions from all the four types of EHPs. Each of them contributes to an energy transport channel.

In the following we show several applications of this central result. To be more specific, we consider a minimum model of the e-bath shown in Fig. 2. We have two electronic states 1 and 2 (on-site energies ε_1 and ε_2) couple to the electrodes L and R with coupling parameter γ_1 and γ_2 , respectively. Electron hopping between the two states is assisted by one bosonic mode, which at the same time couples to a ph-bath with coupling constant γ_{ph} .

Non-reciprocal heat transport.— Firstly, we consider the situation where the e-bath and ph-bath are in their own thermal equilibrium at two different temperature T_e and T_{ph} . This indicates that $\mu_L = \mu_R$ and $T_L = T_R = T_e$. If we ignore the energy dependence of A in Eq. (7), $\Lambda_{mn}(\omega) = \hbar\omega \text{tr}[M^m A M^n A]$ with $A = A_L + A_R$. Consequently, the transmission $\mathcal{T} = \text{Tr}[\Lambda \mathcal{A}_{ph}]$ does not depend on T_e . Equation (8) reduces to the Landauer formula for heat transport between two harmonic thermal baths. Thus, the EHPs behave as linear harmonic oscillator thermal baths.

On the other hand, if we consider the energy dependence of $A(\varepsilon)$, $\Gamma(\omega)$, \mathcal{T} will depend on T_e . Energy transport becomes nonlinear. In this case, non-reciprocal energy flow is possible, i.e., $J(\Delta T) \neq J(-\Delta T)$, with $\Delta T = T_e - T_{ph}$. We thus find a necessary condition for non-reciprocal energy transport in a hybrid electron-boson system: the electron DOS in the thermal window near the chemical potential has to be energy

dependent[35, 36]. For normal metal electrode, the energy scale of electrons is much larger than the thermal energy, leading to a flat DOS. The energy dependence of $A(\varepsilon)$ can be engineered by changing the electronic states of the central part. For example, discrete energy levels of a molecular junction or quantum dot can be used.

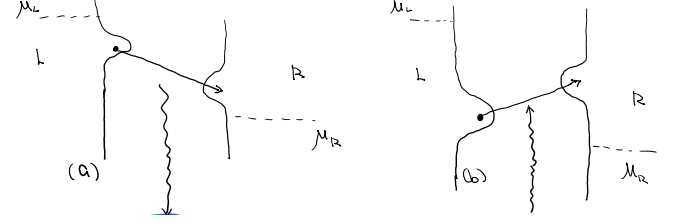


FIG. 2. Two limiting cases of electronic heating (a) and cooling (b). For the e-bath, we have a filled electronic level that couples to the left electrode with chemical potential μ_L , and an empty level that couples to the right electrode with chemical potential μ_R . Electron transport between the two states is mediated by the bosonic mode in the system. (a) Heating of bosonic mode due to resonant recombination of inter-electrode EHPs of type 4. (b) Cooling of bosonic mode due to resonant generation of EHPs of type 3.

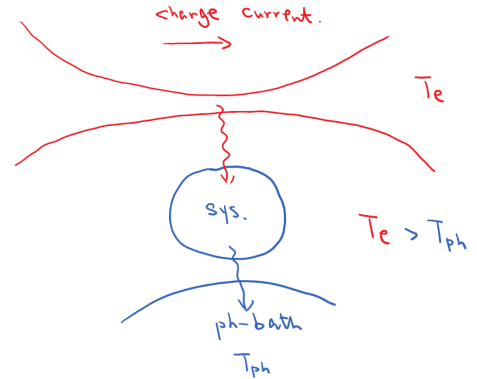


FIG. 3. Electrical current generation from a cold boson bath.

Electrical current from a cold-spot.— We can also study the thermoelectric transport of the temperature-biased electron-boson junction. When $T_{ph} \neq T_e$, in addition to the heat transport between system and e-bath, an electrical current may also be induced between the two electrodes[37, 38]. In our EHP picture, this is realized through coupling of the bosonic mode with two inter-electrode EHPs. Since they contribute to two electrical current with opposite directions, in order to get a non-zero electrical current, these two channels should not get canceled. Here, we consider the case where $T_{ph} < T_e$ and $\mu_L = \mu_R$. The temperature difference between e-bath and ph-bath generates a heat current flow from the e-bath to the ph-bath. At a result of the heat transport, electron transport between L and R electrode takes place.

Previously, electrical current generated from a phonon hot-spot ($T_{ph} > T_e$) has been considered[37]. The situation we consider here is somewhat counter-intuitive. Electricity is generated by cooling the ph-bath. This demonstrates the decoupling of heat and charge transport as an advantage of thermoelectricity in hybrid nano-junctions.

Electronic cooling of bosonic mode.— We now turn on the voltage bias in the e-bath. The applied voltage changes the initial and final electron states of the EHP excitation. Thus, the EHP DOS can be modified by voltage. More importantly, the inter-electrode EHPs acquire a non-zero chemical potential, given by $\pm\mu_{RL}$ respectively. We assume $eV = \mu_L - \mu_R > 0$ without loss of generality. The EHP-4 has a chemical potential of $-eV$, while EHP-3 gets a chemical potential with opposite value eV .

Change of the chemical potential breaks the equilibrium in the reaction, and drives the energy transport between e-bath and the system. Direction of energy flow depends on the relative magnitude of two fluxes. It can be engineered by tuning the electronic band structure, or more specifically, the transition probability of the two types of EHP excitation. Figure 2 shows two limiting cases. In Fig. 2 (a), process 4 is enhanced due to resonant inelastic electron tunneling when the separation between the two DOS peaks is resonant with energy of the bosonic mode. Consequently, energy flows from e-bath to the system. In Fig. 2 (b), the reverse of process 3 is resonantly enhanced, resulting in energy flow in the opposite direction. Electronic cooling becomes possible using this resonant enhancement. Very recently, experimental demonstration of near field radiative cooling using a reversely biased p - n junction has been demonstrated [30]. The experimental results can be understood using this simple model.

In summary, we have shown that a normal two-probe electron conductor can be effectively viewed as EHP baths with chemical potential determined by the applied voltage bias. This is made possible by introducing the inter-electrode charge transfer EHPs. Properties of the EHP baths can be engineered through tuning the parameters of the conductor and the external voltage bias. This bath engineering provides an efficient way of controlling hybrid energy and thermoelectric transport in electron-boson junctions.

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* jtlu@hust.edu.cn

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