Single Mode Heat Rectifier: Controlling Energy Flow Between Electronic Conductors

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We study heat transfer between conductors, mediated by the excitation of a monomodal harmonic oscillator. Using a simple model, we show that the onset of rectification in the system is directly related to the nonlinearity of the electron gas dispersion relation. When the metals have a strictly linear dispersion relation, a Landauer-type expression for the thermal current holds, symmetric with respect to the temperature difference. Rectification becomes prominent when deviations from linear dispersion exist, and the fermionic model cannot be mapped into a harmonic bosonized representation. The effects described here are relevant for understanding radiative heat transfer and vibrational energy flow in electrically insulating molecular junctions.

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Developing nanoscale devices with asymmetric conduction properties is a long-standing challenge for fundamental science, for resolving the microscopic mechanisms controlling transport, and for practical applications. In the past years, rectification in mesoscopic and molecular level systems have attracted much attention. Recent works demonstrated electrical rectification in mesoscopic semiconductor structures [1], one-dimensional (1D) systems of interacting electrons [2], and molecular level devices [3,4]. The growing interest in the thermal properties of nanoscale structures [5] turned rectification of phononic current into a topic of great interest, with implications on nanoscale machinery [6] and thermal computation [7]. The different setups demonstrating this effect [8–12] all rely on two basic assumptions: The device should be asymmetric with respect to the two terminals, and the system's normal modes should nonlinearly interact.

Radiative thermal conductance, where heat exchange between metals is mediated by the generation of photons, is a new topic of interest [13]. It was recently proved that at low temperatures photonic heat conduction is quantized [14], setting an upper bound on single-channel information flow [15]. From the practical aspect, at the nanoscale, radiative heat flow may compete with vibrational energy transfer; thus, these two processes must be considered for properly estimating the thermal conductance of molecular level systems [13].

In this Letter we investigate monomode mediated energy exchange between two metals and resolve the necessary conditions for manifesting thermal rectification. We show that when the metallic leads have a linear dispersion relation, or in other words, when the bosonization approach can be employed to yield a harmonic Hamiltonian, a Landauer-type expression for the energy current holds, symmetric with respect to the temperature difference. In contrast, for conductors with nonlinear dispersion relation, when the approximations involving the standard bosonization scheme break, the energy current can be rectified. Deviations from the Tomonaga-Luttinger bosons picture [16] thus eminently relate to the onset of rectification in the system.

The fermionic model consists of two metallic leads held at different temperatures, coupled by a single harmonic mode. The Hamiltonian includes three contributions $H_F = H_0 + H_e + V_F$,

$$H_0 = \omega_0 b_0^\dagger b_0, \qquad H_e = \sum_{\nu,k} \epsilon_k c_{\nu,k}^\dagger c_{\nu,k},$$
$$V_F = \sum_{\nu,k,k'} \alpha_{\nu,k;\nu,k'} c_{\nu,k}^\dagger c_{\nu,k'} f. \tag{1}$$

H_0 includes a single harmonic mode (subsystem) of frequency ω_0 and creation operator $b_0^\dagger$. It can be considered as a monomodal electromagnetic field, or, in a different context, it represents a local vibrational mode of an intermediate molecular unit. Henceforth we refer to it as a local mode. This mode interacts with two fermionic reservoirs (H_e), where $c_{\nu,k}^\dagger$ ($c_{\nu,k}$) creates (annihilates) an electron at the $\nu = L, R$ metal with momentum k, disregarding the spin degree of freedom. The oscillator metals interaction term V_F couples scattering processes within each metal to the subsystem degrees of freedom, where f is a local mode

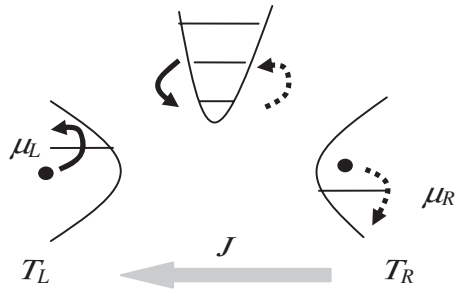

FIG. 1. A schematic representation of our model. While electron flow is blocked, even for $\mu_L > \mu_R$, heat current is flowing right to left ($T_R > T_L$) through an excitation of the local harmonic mode. The curved arrows represent energy transfer processes between the leads and the intermediate mode.

operator. For simplicity, we use $f = b_0^\dagger + b_0$ and assume that the coupling constants α_ν are energy independent and real numbers. Since V_F does not directly couple the reservoirs, the model (1) leaves out charge transfer processes, assuming the tunneling barrier is high and the constriction is long. However, energy can be transferred between the two metals mediated by the excitation of the local mode. For a schematic representation see Fig. 1.

While the aim of this Letter is fundamental, to understand the microscopic origin of asymmetric transport in simple fermionic Hamiltonians, the generic model studied here may be realized in different systems. (i) Radiative energy transfer, where the transfer of the lead's excess energy is mediated by the excitation of electromagnetic modes. Other mechanisms for energy transfer are neglected, e.g., thermionic emission, vibrational energy flow, and photon tunneling. (ii) Vibrational energy flow, where thermal energy is transferred between two metallic terminals through a vibrating link. Here the basic assumptions are that the molecular link electrically insulates and that vibrational energy cannot directly flow from the leads to the molecule due to a large mismatch of the metal-molecule phononic spectra.

For 1D noninteracting electrons with an unbounded strictly linear dispersion relation, $\epsilon_k = \epsilon_F + v_F(|k| - k_F)$, ϵ_F is the Fermi energy and v_F is the velocity at the Fermi energy, bosonization can be employed to yield an equivalent bosonic Hamiltonian, $H_B = H_0 + H_b + V_B$. Here H_0 is the same as above, while the fermionic degrees of freedom are written in terms of Tomonaga-Luttinger (TL) bosons [16], creation operator $b_{\nu,q}^\dagger$ ($\nu = L, R$),

$$\begin{aligned} H_b &= \sum_{\nu,q} \omega_q b_{\nu,q}^\dagger b_{\nu,q}; \quad \omega_q \propto q, \\ V_B &= \sum_{\nu,q} \kappa_{\nu,q}(b_{\nu,q}^\dagger + b_{\nu,q})(b_0^\dagger + b_0). \end{aligned} \tag{2}$$

The new coupling parameters $\kappa_{\nu,q}$ relate to the couplings α_ν [Eq. (1)] [17,18]. Assuming an Ohmic dissipation, the spectral function of the ν boson bath $g_\nu(\omega)$ can be written in terms of the fermionic parameters,

$$\begin{aligned} g_\nu(\omega) &\equiv 4\pi \sum_q \kappa_{\nu,q}^2 \delta(\omega - \omega_q) = 2\pi\omega\xi_\nu; \\ \xi_\nu &= \frac{1}{2}\left[\frac{2}{\pi}\mathrm{atan}(\pi\rho_\nu(\epsilon_F)\alpha_\nu)\right]^2, \end{aligned} \tag{3}$$

with $\rho_\nu(\epsilon_F)$ the density of states at the Fermi energy. The boson Hamiltonian H_B is fully harmonic, and thus can be written in terms of noninteracting collective modes. The heat current in this model can be exactly calculated to yield a Landauer-type expression [19],

$$J = \frac{2}{\pi}\int \mathcal{T}(\omega)[n_B^L(\omega) - n_B^R(\omega)]\omega d\omega. \tag{4}$$

Here $n_B^\nu(\omega) = [e^{\omega/T_\nu} - 1]^{-1}$ is the Bose-Einstein occupation factor with temperature T_ν. The L to R transmission coefficient is given by [19]

$$\mathcal{T}(\omega) = \frac{\omega^2\Gamma_B^L\Gamma_B^R}{[(\omega^2 - \omega_0^2)^2 + (\Gamma_B^L + \Gamma_B^R)^2\omega^2]},$$

with the relaxation rate $\Gamma_B^\nu = 2\pi\sum_q \kappa_{\nu,q}^2\delta(\omega - \omega_q)$. In the weak coupling limit, $\Gamma_B^\nu < \omega_0$, the transmission coefficient is sharply peaked around ω_0, and Eq. (4) reduces into a resonant energy transfer expression,

$$J = \omega_0 \frac{\Gamma_B^L\Gamma_B^R}{\Gamma_B^L + \Gamma_B^R}[n_B^L(\omega_0) - n_B^R(\omega_0)]. \tag{5}$$

Here Γ_B^ν is calculated at the (local oscillator) frequency ω_0. For weak coupling, $\pi\rho(\epsilon_F)\alpha < 1$, Eq. (3) yields $\xi = 2\rho(\epsilon_F)^2\alpha^2$, and the relaxation rate is given by

$$\Gamma_B^\nu = 2\pi\omega_0\alpha_\nu^2\rho_\nu^2(\epsilon_F). \tag{6}$$

Equations (4)–(6) describe the heat current of the model Hamiltonian (1) under the assumption that the two fermionic reservoirs have a strictly linear dispersion relation. It is clear that in this case thermal rectification cannot show up, since exchanging the temperatures of the leads simply reverses the sign of the heat current (4), not the absolute value [20]. We show next that when the bosonization method cannot be trivially employed, i.e., when the dispersion relation is not linear, the model (1) can bring in an interesting rectifying behavior.

Assuming a general dispersion relation, the dynamics of the model (1) is analyzed in the weak coupling system-bath limit. Going into the Markovian limit, the probabilities P_n to occupy the $|n\rangle$ state of the local oscillator satisfy the master equation

$$\dot{P}_n = \sum_m P_m k_{m\to n} - P_n \sum_m k_{n\to m}, \tag{7}$$

where the transition rate from the local oscillator state $|m\rangle$ to $|n\rangle$ is additive in the L and R reservoirs, $k_{n\to m} = k_{n\to m}^L + k_{n\to m}^R$, due to the linear form of the interaction [Eq. (1)] [10]. In steady state, the heat current across the system is given by (calculated, e.g., at the L side)

$$J = \sum_{m,n} E_{m,n} P_n k_{n\to m}^L, \tag{8}$$

with $E_{m,n} = E_m - E_n$. At the level of the golden rule formula, the transition rates are given by

$$\begin{aligned} k_{n\to m}^\nu &= 2\pi|f_{m,n}|^2 \sum_{k,k'} |\alpha_{\nu,k;\nu,k'}|^2 n_F^\nu(\epsilon_k)[1 - n_F^\nu(\epsilon_{k'})] \\ &\quad \times \delta(\epsilon_k - \epsilon_{k'} - E_{m,n}) \\ &= -2\pi|f_{m,n}|^2 n_B^\nu(E_{m,n}) \\ &\quad \times \int d\epsilon [n_F^\nu(\epsilon) - n_F^\nu(\epsilon - E_{m,n})]F_\nu(\epsilon). \end{aligned} \tag{9}$$

Focusing on the last equality, interestingly we see that the thermal properties of the reservoirs are concealed within both the Fermi-Dirac distribution function $n_F^\nu(\epsilon) = [e^{(\epsilon-\mu_\nu)/T_\nu} + 1]^{-1}$ and the Bose-Einstein occupation factor

$n_B^\nu(\epsilon) = [e^{\epsilon/T_\nu} - 1]^{-1}$. It is therefore clear that when the integral yields a temperature independent constant, the statistic of the reservoirs is fully bosonic. The other elements in (9) are the matrix elements of the system operator $f_{m,n} = \langle m|f|n\rangle$ and the dimensionless interaction term $F_\nu(\epsilon) = |\alpha_\nu|^2 \rho_\nu(\epsilon)\rho_\nu(\epsilon - E_{m,n})$.

We assume next that the density of states slowly varies in the energy window $E_{m,n}$. The interaction function is then expanded around the chemical potential [21],

$$F_\nu(\epsilon) \approx F_\nu(\mu_\nu) + \gamma_\nu \frac{|\epsilon| - \mu_\nu}{\mu_\nu}, \tag{10}$$

with γ_ν a dimensionless number of order unity. Using this form, the integration in Eq. (9) can be performed when the Fermi energies are much bigger than the conduction band edge, $\mu_\nu \gg E_c$ [21]. Making use of the following relationships, $\int_{-\infty}^{\infty} d\epsilon[n_F^\nu(\epsilon) - n_F^\nu(\epsilon - E_{m,n})] \approx -E_{m,n}$, $\int_{-\infty}^{\infty} |\epsilon| d\epsilon[n_F^\nu(\epsilon) - n_F^\nu(\epsilon - E_{m,n})] \approx -1.4E_{m,n}T_\nu$ $(T_\nu > |E_{m,n}|)$, we get

$$k^\nu_{n\to m} = 2\pi|f_{m,n}|^2 n_B^\nu(E_{m,n})E_{m,n}\left[F_\nu(\mu_\nu) + 1.4\gamma_\nu \frac{T_\nu}{\mu_\nu}\right]. \tag{11}$$

Note that $n_B(-E_{m,n}) = -[n_B(E_{m,n}) + 1]$; thus, the excitation and relaxation rates induced by the ν reservoir satisfy the detailed balance relation, $k^\nu_{n\to m}/k^\nu_{m\to n} = e^{-E_{m,n}/T_\nu}$. We consider next the limit of a constant density of states, taking $\gamma = 0$. Equation (11) then becomes

$$\begin{aligned} k^\nu_{n\to n-1} &= n\Gamma_F^\nu(\omega_0)[1 + n_B^\nu(\omega_0)]; \\ \Gamma_F^\nu(\omega_0) &= 2\pi F_\nu(\mu_\nu)\omega_0, \end{aligned} \tag{12}$$

where the bilinear interaction form was employed, $f = (b_0^\dagger + b_0)$, leading to nearest neighbor transitions only. We can also calculate the states' population in steady state by putting $\dot{P}_n = 0$ in Eq. (7) [10],

$$P_n = x^n(1 - x); \qquad x = \frac{\sum_\nu \Gamma_F^\nu(\omega_0) n_B^\nu(\omega_0)}{\sum_\nu \Gamma_F^\nu(\omega_0)[1 + n_B^\nu(\omega_0)]}. \tag{13}$$

Finally, the heat current for the $\gamma = 0$ case is calculated with the help of Eq. (8),

$$J = \omega_0 \frac{\Gamma_F^L \Gamma_F^R}{\Gamma_F^L + \Gamma_F^R}[n_B^L(\omega_0) - n_B^R(\omega_0)]. \tag{14}$$

Γ_F^ν is calculated at the frequency ω_0. In the linear dispersion limit we thus recover the resonant energy behavior (5) obtained with the equivalent boson Hamiltonian (2). Note that the rates calculated with the different methods, Γ_F^ν and Γ_B^ν, are equal; see Eqs. (6) and (12).

We evaluate next the current when $\gamma \neq 0$, i.e., for a model with an energy dependent density of states. For the linear coupling case, $f = b_0^\dagger + b_0$, Eq. (11) becomes $k^\nu_{n\to n-1} = n\Gamma_F^\nu(\omega_0)[1 + n_B^\nu(\omega_0)](1 + \lambda_\nu \frac{T_\nu}{\mu_\nu})$, where $\lambda_\nu = \frac{1.4\gamma_\nu}{F_\nu(\mu_\nu)}$. The steady state population is therefore given by Eq. (13) with $\Gamma_F^\nu \to \Gamma_F^\nu(1 + \lambda_\nu T_\nu/\mu_\nu)$. Next we calculate the energy current using Eq. (8). In the classical limit $(T_\nu > \omega_0)$ we obtain

$$J = \frac{\Gamma_F^L \Gamma_F^R (1 + \lambda_L \frac{T_L}{\mu_L})(1 + \lambda_R \frac{T_R}{\mu_R})}{\Gamma_F^L(1 + \lambda_L \frac{T_L}{\mu_L}) + \Gamma_F^R(1 + \lambda_R \frac{T_R}{\mu_R})}(T_L - T_R), \tag{15}$$

where the dimensionless parameter λ_ν effectively quantifies the deviation from the linear dispersion case.

Comparing Eq. (15) to the high temperature limit of Eq. (14) interestingly reflects the deviations from the TL boson picture in transport properties. Equation (15) has three important characteristics: First, the heat current obtained is a nonlinear function of the temperature difference. In fact, for $\Gamma_F^L \gg \Gamma_F^R$, $J \propto a_1\Delta T + \lambda a_2 \Delta T^2$. The constants $a_{1,2}$ depend on subsystem and bath parameters, $\Delta T = T_L - T_R$. Second, this expression manifests nontrivial controllability over the energy current by tuning the chemical potentials. Third, this result demonstrates thermal rectification, as the thermal current is different when switching the temperature bias, assuming some asymmetry is included, e.g., the chemical potentials, or the local mode-bath interactions are different at the two terminals. When the electronic properties of the reservoirs are equivalent, $\lambda \equiv \lambda_\nu$, $\mu_a \equiv \mu_\nu$, we find $\Delta J = J_{+\Delta T} + J_{-\Delta T} \propto -\lambda \frac{\Delta T^2}{\mu_a}(\Gamma_F^L - \Gamma_F^R)$, where $J_{\pm\Delta T} = J(T_L - T_R = \pm\Delta T)$. The proportionality factor is given by $G \approx \Gamma_F^L\Gamma_F^R/(\Gamma_F^L + \Gamma_F^R)^2$ for $\lambda T_\nu/\mu_a < 1$. Thus, a system made of a local harmonic mode coupled asymmetrically to two metals with energy dependent density of states rectifies heat. Rectification becomes more effective with increasing λ, i.e., when the density of states strongly varies with energy. In Fig. 2 we plot the absolute value of the ratio $J_{+\Delta T}/J_{-\Delta T}$, and find that it linearly departs from unity with increasing

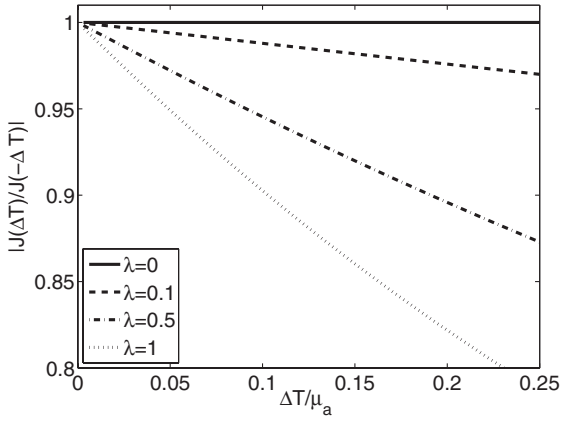

FIG. 2. Thermal rectification with increasing deviations from constant density of states. $\lambda = 0$ (full line); $\lambda = 0.1$ (dashed line); $\lambda = 0.5$ (dash-dotted line); $\lambda = 1$ (dotted line). $\Gamma_F^L = 0.5$, $\Gamma_F^R = 0.02$, $\mu_a = \mu_\nu = 1$; temperature of the cold bath is $T_{\text{low}} = 0.1$.

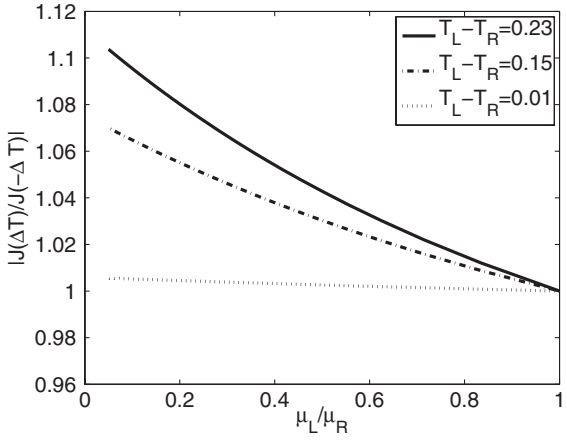

FIG. 3. Thermal rectification due to chemical potential difference. $\Delta T = 0.23$ (full line); $\Delta T = 0.15$ (dashed line); $\Delta T = 0.01$ (dotted line). $\Gamma_F^L = \Gamma_F^R = 0.1$, $\mu_L = 1$, $\lambda_L = \lambda_R = 1$.

temperature difference and λ, in agreement with the analytical expression ($T_a = \frac{T_L+T_R}{2}$),

$$|J_{+\Delta T}/J_{-\Delta T}| \approx 1 - \frac{\lambda}{\mu_a}\Delta T \frac{\Gamma_F^L - \Gamma_F^R}{(\Gamma_F^L + \Gamma_F^R)(1 + \frac{\lambda T_a}{\mu_a})}. \quad (16)$$

Figure 3 demonstrates the dependence of the rectifying behavior on the leads' chemical potentials for a system symmetrically coupled to the terminals using Eq. (15). We find that at a small temperature difference rectification is negligible, while for $\Delta T/\mu_L > 0.1$ asymmetry in the current can be large up to 10%. Note, however, that tunneling of electrons becomes a significant source of noise at large voltage bias and/or temperature differences.

The thermal current can be calculated for other models besides (10), expressing Eq. (9) as $k^{\nu}_{n\to m} = -2\pi|f_{m,n}|^2 n_B^{\nu}(E_{m,n})\alpha_{\nu}^2 g_{\nu}(T_{\nu})$, where $g_{\nu}(T_{\nu})$ is defined through this relation. Equation (16) then reduces to $\mathcal{R} \equiv |J_{+\Delta T}/J_{-\Delta T}| \approx g_R(T_R)/g_R(T_L)$ for the linear coupling model if $\alpha_L \gg \alpha_R$. We consider a superconducting R lead (L can be a normal metal), as it effectively suppresses normal electronic thermal conductance [5,14], and numerically evaluate $g_R(T)$ with the density of states $\rho_R(\epsilon) = (|\epsilon|/\sqrt{\epsilon^2 - \Delta^2})\Theta(|\epsilon| - \Delta)$. Using $\Delta = 0.2$ meV (aluminum), $\omega_0 = 0.4$ meV, and $T = 0.08$–0.2 meV, we get $g_R(T) \sim a_1 + a_2 T$ where $a_2 T \gg a_1$. The rectification ratio then becomes $\mathcal{R} \sim T_R/T_L$, which can be as large as 2.5 for the above parameters, potentially measurable using the setup of Ref. [22]. Similarly, a narrow band gap (0.1 eV) semiconducting lead yields $\mathcal{R} \sim 10$ for $\omega_0 = 0.1$ eV at the temperature range $T \sim 0.01$–0.5 eV.

Previous studies on phononic heat transfer in 1D chains demonstrated that anharmonic interactions are crucial for manifesting thermal rectification [8–11]. Our result complies with this observation: A fermionic reservoir with a nonlinear dispersion relation can be represented by a bath of bosons comprising nonlinear interactions [23]. The fermionic Hamiltonian (1) can thus be mapped into a fully bosonic model describing a harmonic link bilinearly connected to anharmonic thermal baths.

In summary, we present here a simple model of single-mode energy transfer between metals. The model can describe energy flow through vibrating link and radiative heat transfer. We resolve the microscopic requirements for manifesting rectification: For noninteracting electrons, rectification appears when the reservoirs' density of states or, analogously, system-bath couplings are energy dependent. Since dissipative reservoirs typically contain anharmonic interactions, finite rectification of the energy current between metals and dielectric surfaces is an inevitable effect.

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