

Sufficient Conditions for Thermal Rectification in Hybrid Quantum Structures

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We analytically identify sufficient conditions for manifesting thermal rectification in two-terminal junctions, including a subsystem connected to two reservoirs, within the quantum master equation formalism. We recognize two classes of rectifiers. In type A rectifiers, the reservoirs' energy structure is dissimilar. In type B rectifiers, the baths are identical but include particles whose statistics differ from that of the subsystem, to which they asymmetrically couple. Our study applies to various hybrid junctions including metals, dielectrics, and spins.

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Understanding heat transfer in hybrid structures is of fundamental and practical importance for controlling transport at the nanoscale and for realizing functional devices [1]. Among the systems that fall into this category are metal-molecule-metal junctions, the basic component of molecular electronic devices [2], and dielectric-molecule-dielectric systems, where vibrational energy flow activates reactivity and controls dynamics [3]. Single-mode *radiative* heat conduction between Ohmic metals was recently detected, showing that photonic thermal conductance is quantized [4]. Other hybrid systems with interesting thermal properties are electronic spin-nuclear spin interfaces [5], metal-molecule contacts with exciton-to-phonon energy transfer [6], and metal-superconductor junctions [7].

Thermal rectification, namely, an asymmetry of the heat current for forward and reversed temperature gradients, has recently attracted considerable theoretical [8–14] and experimental [15,16] attention. Most theoretical studies, confined to a specific realization, have observed this phenomenon in *phononic systems* using classical molecular dynamics simulations. It is still not clear what conditions the system should fulfill for showing the effect model independently and, in particular, what the roles are of the nonlinearity, asymmetry, and inhomogeneity of the structure [14]. In this Letter, we attempt a first step towards a general understanding of thermal rectification. We focus on a prototype quantum model, including a central quantized unit (subsystem) and two bulk objects (referred to as terminals/reservoirs/baths), and establish sufficient conditions for rectification. We identify two classes of rectifiers: (i) “type A,” where the reservoirs are dissimilar, i.e., of different mean energy, and (ii) “type B,” where the baths are identical, but their statistics differ from that of the subsystem, combined with unequal coupling strengths at the two ends, as explained below. These rectifiers could be realized in several subsystems (vibrational or radiation modes) and reservoirs (spin, metal, dielectrics); see Fig. 1.

Consider a one-dimensional hybrid structure where a central unit H_S interacts with two reservoirs H_ν^0 ($\nu = L, R$) of temperatures $T_\nu = \beta_\nu^{-1}$ via the coupling terms V_ν :

$$H = H_L^0 + H_R^0 + H_S + V_L + V_R. \quad (1)$$

Based on the continuity equation, the heat current from the left bath into the subsystem is given by $J = \frac{i}{2} \text{Tr}([H_L^0 - H_S, V_L]\rho)$; ($\hbar \equiv 1$) [17], where ρ is the total density matrix, and we trace over the subsystem and reservoirs degrees of freedom. The heat current is defined as positive when flowing left to right. In the steady state, the expectation value of the interaction is zero, $\text{Tr}(\frac{\partial V_L}{\partial t} \rho) = 0$, and the current at the L and R terminals is the same. We then obtain [17]

$$J = \text{Tr}[\hat{J}\rho]; \quad \hat{J} = \frac{i}{2}[V_L, H_S] + \frac{i}{2}[H_S, V_R]. \quad (2)$$

The subsystem Hamiltonian assumes a diagonal form, and we also consider separable couplings

$$H_S = \sum_n E_n |n\rangle\langle n|; \quad V_\nu = \lambda_\nu S B_\nu; \quad S = \sum_{n,m} S_{m,n} |m\rangle\langle n|. \quad (3)$$

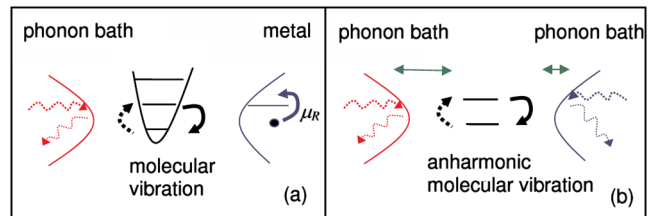


FIG. 1 (color online). Examples of hybrid rectifiers treated in this work. (a) “Type A”: Energy exchange between dissimilar terminals. The central object represents either a vibrational or a radiation mode. (b) “Type B”: Heat transfer between harmonic solids mediated by the excitation of an anharmonic (truncated) vibrational mode. Double-ended arrows represent strong (left) and weak (right) subsystem-bath couplings.

Here S is a subsystem operator, B_ν is an operator in terms of the ν bath degrees of freedom (DOF), and λ_ν is the subsystem-bath coupling strength, a real number. For simplicity, we set $S_{m,n} = S_{n,m}$. In what follows, we consider situations where B_L and B_R have the same dependence on the (respective) bath DOF, with different prefactors $\lambda_L \neq \lambda_R$. We refer to this scenario as “parametric asymmetry.” Note that if the commutator $[H_S, S] = 0$, the heat current trivially vanishes.

We begin and discuss the type A rectifier, constructed by adopting reservoirs with distinct properties, as we explain below. First we derive a general expression for the heat flux across the junction. The initial density matrix is assumed to be a tensor product of subsystem ρ_S and bath ρ_B factors, where $\rho_B = \rho_L(T_L) \otimes \rho_R(T_R)$ is a tensor product of the L and R baths’ terms with $\rho_\nu(T_\nu) = e^{-H_\nu^0/T_\nu} / \text{Tr}_\nu[e^{-H_\nu^0/T_\nu}]$. For convenience, we delete the direct reference to time. In terms of $T_a = T_L + T_R$ and $\Delta = T_L - T_R$, we can write the initial total density matrix as

$$\begin{aligned} \rho(T_a, \Delta) &= \rho_B \otimes \rho_S \\ &= \frac{1}{Z} e^{-[2T_a(H_L^0 + H_R^0) - 2\Delta(H_L^0 - H_R^0)]/(T_a^2 - \Delta^2)} \otimes \rho_S, \end{aligned} \quad (4)$$

where $Z = \text{Tr}[\rho_S \rho_B]$ is the partition function with the trace performed over the bath and system degrees of freedom. Furthermore, without loss of generality, the system density matrix is assumed to depend initially only on the average temperature $\rho_S \equiv \rho_S^{(0)}(T_a/2)$.

The expectation value of the energy current is given by evaluating $J(T_a, \Delta) = \text{Tr}[\hat{J}\rho(T_a, \Delta)]$, where \hat{J} is an Heisenberg representation operator $\hat{J} = e^{iHt}\hat{J}(0)e^{-iHt}$. Expanded in powers of Δ , we get

$$J(T_a, \Delta) = \frac{2\Delta}{T_a^2} \alpha_1(T_a) + \frac{(2\Delta)^2}{T_a^4} \alpha_2(T_a) + O(T_a, \Delta^3), \quad (5)$$

with the coefficients

$$\alpha_1(T_a) = \text{Tr}[\hat{J}(H_L^0 - H_R^0)\rho_T], \quad (6)$$

$$\alpha_2(T_a) = \frac{1}{2}\text{Tr}[\hat{J}(H_L^0 - H_R^0)^2\rho_T] - \alpha_1(T_a)\text{Tr}[(H_L^0 - H_R^0)\rho_T]. \quad (7)$$

Here $\rho_T = \frac{1}{Z_T} \rho_L(\frac{T_a}{2})\rho_R(\frac{T_a}{2})\rho_S^{(0)}(\frac{T_a}{2})$ is the density matrix at the average temperature $\frac{T_L + T_R}{2}$ with the partition function Z_{T_a} . Equation (7) was derived using the fact that $\text{Tr}[\hat{J}\rho_T]$ and all of its T derivatives are zero. Using the definition of the current operator (2), we obtain an explicit expression for α_1 :

$$\alpha_1(T_a) = i\text{Tr}\{[S, H_S](\lambda_L B_L H_L^0 + \lambda_R B_R H_R^0)\rho_T\}. \quad (8)$$

Since $[H_S, S] \neq 0$ [see discussion after Eq. (3)], the linear term in J is finite. We now examine the onset of thermal rectification, i.e., discuss the sufficient conditions for having $\alpha_2 \neq 0$. While there might be some special values of T_a where the two terms in (7) nullify, in general, since B_ν

and H_ν^0 are independent operators, the result is finite. Thus, in order to manifest rectification, it is enough to identify when one of the terms in (7) is nonzero. (i) The first expression is finite if $\text{Tr}[FH_L^0\rho_T] \neq \text{Tr}[FH_R^0\rho_T]$; $F = [S, H_S](\lambda_L B_L H_L^0 + \lambda_R B_R H_R^0)$. (ii) The second term is nonzero if $\text{Tr}[H_L^0\rho_T] \neq \text{Tr}[H_R^0\rho_T]$. Based on (ii), we deduce that rectification emerges when the reservoirs have different mean energy [18]:

$$\langle H_L^0 \rangle \equiv \text{Tr}_L[\rho_L(T)H_L^0] \neq \text{Tr}_R[\rho_R(T)H_R^0] \equiv \langle H_R^0 \rangle. \quad (9)$$

As an example, consider baths including a one-dimensional oscillator chain $H_\nu^0 = H_\nu^{\text{kin}} + H_\nu^{\text{pot}}$, where the kinetic energy H_ν^{kin} is quadratic in momentum, and the potential energy per particle is $C_n q^n$; $n \geq 2$, and q is the particle’s coordinate. In the classical limit using the equipartition relation, we obtain $\langle H_\nu^0 \rangle = T_\nu(\frac{1}{2} + \frac{1}{n})$. Thermal rectification thus emerges if the reservoirs have a nonidentical power n . Note that the separation to three segments (L , subsystem, and R) is often artificial: The junction could be practically made of a single structure with a varying potential energy, e.g., a nanotube whose composition gradually changes in space [15]. Finally, note that our discussion could be generalized to cases where $V_\nu = \lambda_\nu S_\nu B_\nu$; $S_L \neq S_R$; see Eq. (3).

We turn to the type B rectifier and show that for identical reservoirs rectification emerges when the subsystem and reservoirs have different statistics, in conjunction with some parametric asymmetry. It is easy to show that under (3) the steady-state current (2) becomes $J = i\sum_{n,m} E_{m,n} S_{m,n} \text{Tr}_B(\lambda_L B_L \rho_{m,n})$, where $E_{m,n} = E_m - E_n$ and Tr_B denotes the trace over the L and R reservoirs degrees of freedom. Employing the Liouville equation in the interaction picture, the elements of the total density matrix satisfy

$$\begin{aligned} \frac{d\rho_{m,n}}{dt} &= -i[V(t), \rho(0)]_{m,n} \\ &\quad - \int_0^t d\tau [V(t), [V(\tau), \rho(\tau)]]_{m,n}, \end{aligned} \quad (10)$$

where $V = V_L + V_R$ and $V(t)$ are interaction picture operators. Following standard weak coupling schemes [19], taking $\text{Tr}_B[V(t), \rho(0)] = 0$, and assuming the Markovian limit, the heat current becomes

$$J = \frac{1}{2} \sum_{n,m} E_{m,n} |S_{m,n}|^2 P_n (k_{n \rightarrow m}^L - k_{n \rightarrow m}^R), \quad (11)$$

where the transition rates are given by $k_{n \rightarrow m}^\nu = \lambda_\nu^2 \int_{-\infty}^{\infty} d\tau e^{iE_{n,m}\tau} \langle B_\nu(\tau) B_\nu(0) \rangle$, and the population $P_n = \text{Tr}_B(\rho_{n,n})$ satisfies the differential equation

$$\dot{P}_n = \sum_{\nu,m} |S_{m,n}|^2 P_m k_{m \rightarrow n}^\nu - P_n \sum_{\nu,m} |S_{m,n}|^2 k_{n \rightarrow m}^\nu. \quad (12)$$

In the steady state $\dot{P}_n = 0$, and we normalize the population to unity $\sum_n P_n = 1$. We consider next two representative models for the subsystem Hamiltonian and its

interaction with the baths. In the first model, the subsystem is a harmonic oscillator (HO) of frequency ω : $H_S = \sum_n n\omega |n\rangle\langle n|$. This can describe either a local radiation mode [4,20] or a vibrational mode of the trapped molecule. We also take $S = \sum_n \sqrt{n} |n\rangle\langle n-1| + \text{c.c.}$, motivated by the bilinear form $V_\nu \propto xB_\nu$, where x is a subsystem coordinate [10]. This implies that only transitions between nearest states are allowed:

$$k^\nu \equiv k_{n \rightarrow n-1}^\nu = \lambda_\nu^2 \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} \langle B_\nu(\tau) B_\nu(0) \rangle; \quad (13)$$

$$k_{n-1 \rightarrow n}^\nu = e^{-\beta_\nu \omega} k_{n \rightarrow n-1}^\nu.$$

We also introduce the short notation $k^\nu = \lambda_\nu^2 f_\nu(T_\nu)$, where f_ν , defined through (13), encompasses the effect of the bath operators. Solving (12) in the steady state using the rates (13), the heat current is calculated with (11):

$$J^{(\text{HO})} = - \frac{\omega [n_B^L(\omega) - n_B^R(\omega)]}{n_B^L(-\omega)/k^L + n_B^R(-\omega)/k^R}, \quad (14)$$

where $n_B^\nu(\omega) = [e^{\beta_\nu \omega} - 1]^{-1}$ is the Bose-Einstein distribution at $T_\nu = 1/\beta_\nu$, and $n_B^\nu(-\omega) = -[n_B^\nu(\omega) + 1]$.

Our second subsystem is a two-level system (TLS), where $H_S = \frac{\omega}{2} \sigma_z$ and $S = \sigma_x$. These terms may represent an electronic spin rotated by the environment [5] or an anharmonic molecular vibration [10]. Recalculating the long-time population (12), the heat flux reduces to

$$J^{(\text{TLS})} = \frac{\omega [n_S^L(\omega) - n_S^R(\omega)]}{n_S^L(-\omega)/k^L + n_S^R(-\omega)/k^R}, \quad (15)$$

with the rates (13) and the spin occupation factor $n_S^\nu(\omega) = [e^{\beta_\nu \omega} + 1]^{-1}$. We analyze next the structure of Eqs. (14) and (15). In each expression separately the currents for forward and reversed temperature gradients deviate in magnitude, if the denominators fulfill

$$\frac{n^L(-\omega)}{\lambda_L^2 f_L(T_L)} - \frac{n^L(-\omega)}{\lambda_R^2 f_R(T_L)} \neq \frac{n^R(-\omega)}{\lambda_L^2 f_L(T_R)} - \frac{n^R(-\omega)}{\lambda_R^2 f_R(T_R)}, \quad (16)$$

where n^ν are either the spin occupation factor or the Bose-Einstein distribution. Note that the two sides of the last equation depend on different temperatures; thus if $f_L(T) \neq f_R(T)$ resulting, e.g., from the use of dissimilar reservoirs, the system generally rectifies heat besides special points in the parameter space, depending on the details of the model. This case reduces to the type A rectifier discussed above. However, a more careful analysis of (16) reveals that rectification prevails even when $f_L(T) = f_R(T)$, as long as $\lambda_L \neq \lambda_R$ and the ratio $n^\nu(-\omega)/f_{L,R}(T_\nu)$ depends on the temperature T_ν . In other words, *the relaxation rates' temperature dependence should differ from the central unit particle statistics*. As we show next, the function $f_\nu(T)$ reflects the reservoirs statistics. We therefore classify type B rectifiers as junctions where the subsystem and bath differ in their statistics, and the identical reservoirs are asymmetrically coupled to the subsystem $\lambda_L \neq \lambda_R$. For example, a TLS asymmetrically coupled to two harmonic

baths rectifies heat [10]. We specify next the terminals and exemplify the two classes of thermal rectifiers in various structures.

Spin bath.—Assuming the environment includes a set of distinguishable noninteracting spin-1/2 particles ($p = 1, 2, \dots, P$), the Hamiltonian is given by summing all single particle contributions $H_\nu^0 = \sum_p h_{\nu,p}^0$ and $B_\nu = \sum_p b_{\nu,p}$. It can be shown that the relaxation rate (13) reduces to

$$k^\nu = Y_\nu(\omega) n_S^\nu(-\omega), \quad (17)$$

where $Y_\nu(\omega) = 2\pi\lambda_\nu^2 \sum_p |\langle 0|_p b_{\nu,p} |1\rangle_p|^2 \delta[\omega + \epsilon_p(0) - \epsilon_p(1)]$, with the p -particle eigenstates $|i\rangle_p$ and eigenvalues $\epsilon_p(i)$ ($i = 0, 1$), and $n_S^\nu(\omega) = [e^{\beta_\nu \omega} + 1]^{-1}$.

Solid/radiation field (harmonic bath).—The bath includes a set of independent harmonic oscillators, creation operator $a_{\nu,j}^\dagger$. System-bath interaction is further assumed to be bilinear: $H_\nu^0 = \sum_j \omega_j a_{\nu,j}^\dagger a_{\nu,j}$; $B_\nu = \sum_j (a_{\nu,j} + a_{\nu,j}^\dagger)$. This leads to the relaxation rate (13)

$$k^\nu = -\Gamma_B^\nu(\omega) n_B^\nu(-\omega), \quad (18)$$

where $\Gamma_B^\nu(\omega) = 2\pi\lambda_\nu^2 \sum_j \delta(\omega_j - \omega)$ is an effective system-bath coupling energy.

Metal.—Consider a metallic terminal, including a set of noninteracting spinless electrons, creation operator $c_{\nu,i}^\dagger$, where only scattering between electronic states within the same lead is allowed: $H_\nu^0 = \sum_i \epsilon_i c_{\nu,i}^\dagger c_{\nu,i}$; $B_\nu = \sum_{i,j} c_{\nu,i}^\dagger c_{\nu,j}$. The transition rate (13) can be written as

$$k^\nu = -2\pi\lambda_\nu^2 n_B^\nu(-\omega) \times \sum_{i,j} \delta(\epsilon_i - \epsilon_j + \omega) [n_F^\nu(\epsilon_i) - n_F^\nu(\epsilon_i + \omega)], \quad (19)$$

with the Fermi-Dirac function $n_F^\nu(\epsilon) = [e^{\beta_\nu(\epsilon - \mu_\nu)} + 1]^{-1}$ at the chemical potential μ_ν . One could also write [20]

$$k^\nu = -\Lambda^\nu(T_\nu, \omega) n_B^\nu(-\omega), \quad (20)$$

where $\Lambda^\nu(T_\nu, \omega) = 2\pi \int d\epsilon [n_F^\nu(\epsilon) - n_F^\nu(\epsilon + \omega)] F_\nu(\epsilon)$, with $F_\nu(\epsilon) = \lambda_\nu^2 \sum_{i,j} \delta(\epsilon - \epsilon_j + \omega) \delta(\epsilon_i - \epsilon)$. By assuming that the density of states slowly varies in the energy window ω , the F function could be expanded around μ . If the Fermi energy is much bigger than the conduction band edge, we obtain $\Lambda^\nu(T_\nu, \omega) \approx \Gamma_F^\nu(1 + \delta_\nu \frac{T_\nu}{\mu_\nu})$, where $\Gamma_F^\nu = 2\pi\omega F_\nu(\mu_\nu)$, and δ is a constant of order one, measuring the deviation from a flat band structure near μ_ν [20].

Consider, for example, a type A solid-HO-metal rectifier, representing an electronic to vibrational energy conversion device; see Fig. 1(a). This system might be realized by attaching an insulating molecule to a metal (STM tip), while the underneath surface is insulating. Setting the coupling strength at both contacts to be the same $\Gamma_B^L(\omega) = \Gamma_F^R(\omega)$, we can calculate the rectification ratio $\mathcal{R} \equiv | \frac{J(\Delta)}{J(-\Delta)} |$ using (14), (18), and (20)

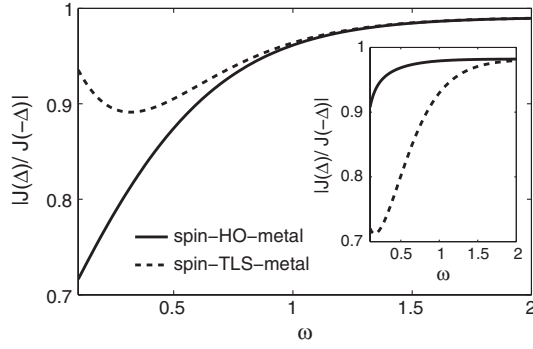


FIG. 2. Spin-HO-metal rectifier (solid line) and a spin-TLS-metal rectifier (dashed line). The rectification ratio is presented as a function of the subsystem spacing. $T_a = 0.5$, $\Delta = 0.1$, $\delta_R = 0.2$, and $\mu_R = 1$. Main plot: The subsystem is equally coupled to the two ends: $\Upsilon_L = \Gamma_F^R = 1$. Inset: The effect can be tuned by manipulating system-bath couplings: $\Upsilon_L = 1$, $\Gamma_F^R = 0.05$.

$$\mathcal{R} = \frac{(2 + \delta_R \frac{T_L}{\mu_R})(1 + \delta_R \frac{T_R}{\mu_R})}{(1 + \delta_R \frac{T_L}{\mu_R})(2 + \delta_R \frac{T_R}{\mu_R})} \sim 1 - \Delta \frac{\delta_R}{2\mu_R}. \quad (21)$$

Therefore, if the metal density of state varies with energy ($\delta \neq 0$), thermal rectification is presented [20]. Interestingly, we can show that in a solid-TLS-metal junction \mathcal{R} could be modulated to be greater or smaller than 1 by varying the gap ω . Thus, phonon-to-exciton heat conversion could be made effective, while the exciton-to-phonon route could be made ineffective, and vice versa. Figure 2 further displays the tunability of a spin-subsystem-metal junction. In the classical limit ($\omega < T_\nu$) rectification can be substantial, while in the quantum regime the effect is suppressed. Modifying the subsystem-metal coupling strength largely controls the rectification ratio (inset).

As a type B example, consider a solid-TLS-solid structure, modeling heat transfer between two macroscopic solids through the excitation of an anharmonic vibrational mode of an intermediated molecule [10]. In the classical limit [see (15) and (18)], we get $J(T_\omega, \Delta) \propto \frac{\Delta}{T_a}(1 - x \frac{\Delta}{T_a} + x^2 \frac{\Delta^2}{T_a^2} + \dots)$; $x = (\Gamma_B^L - \Gamma_B^R)/(\Gamma_B^L + \Gamma_B^R)$. This demonstrates that even Δ terms, i.e., thermal rectification, are directly linked to the parametric asymmetry.

Anharmonic interactions were, in particular, pointed out as responsible for thermal rectification. While it is obvious that harmonic systems do not rectify heat [21], not all anharmonic-asymmetric systems do bring in the effect. Consider a three-segment nonlinear oscillator chain where all units have identical potential energy, but the central part is parametrically-asymmetrically connected to the terminals. According to our analysis, this system, of homogeneous energy spectra, will not rectify heat.

In summary, while previous studies were focused on specific realizations, typically limited to the classical regime, with insights based on numerical simulations, we

have analytically deduced a sufficient condition for the onset of thermal rectification in generic hybrid structures within an open quantum system approach: (i) The reservoirs should be made dissimilar; e.g., the L terminal is harmonic, while the R side has an anharmonic potential energy function. (ii) The reservoirs could be taken equal, but their statistics should differ from that of the subsystem, combined with some parametric asymmetry; e.g., a solid-spin-solid junction rectifies heat when $\Gamma_B^L \neq \Gamma_B^R$.

Nonlinear energy transfer at the nanoscale is important for cooling electronic and mechanical devices and for controlling molecular reactivity. Our study applies to various interfaces: metals, insulators, and noninteracting spins. The central unit could represent a radiation/vibrational mode or an electronic excitation. Thermal rectification is thus an ubiquitous effect that could be observed in a variety of systems, phononic [15], electronic [16], and photonic [4,20].

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