Thermoodynamics of the polaron master equation at finite bias

Thilo Krause1,2,∗ Tobias Brandes1, Massimiliano Esposito3, and Gernot Schaller1,11

1 Institut für Theoretische Physik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany
2 Paul Drude Institut für Festkörperlektronik, Hausvogteiplatz 5-7, D-10117 Berlin, Germany
3 Complex Systems and Statistical Mechanics, University of Luxembourg, L-1511 Luxembourg, Luxembourg

We study coherent transport through a double quantum dot. Its two electronic leads induce electronic matter and energy transport and a phonon reservoir contributes further energy exchanges. By treating the system-lead couplings perturbatively, whereas the coupling to vibrations is treated non-perturbatively in a polaron-transformed frame, we derive a thermodynamic consistent low-dimensional master equation. When the number of phonon modes is finite, a Markovian description is only possible when these couple symmetrically to both quantum dots. For a continuum of phonon modes however, also asymmetric couplings can be described with a Markovian master equation. We compute the electronic current and dephasing rate. The electronic current enables transport spectroscopy of the phonon frequency and displays signatures of Franck-Condon blockade. For infinite external bias but finite tunneling bandwidths, we find oscillations in the current as a function of the internal bias due to the electron-phonon coupling. Furthermore, we derive the full fluctuation theorem and show its identity to the entropy production in the system.

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Electronic transport through quantum systems has been intensively studied both theoretically and experimentally over the last years. In part, this has been triggered by the fact that single molecules or quantum dot configurations are promising candidates for a variety of applications such as e.g. charge22 and spin55 qubits, or as single photon emitters, for example realized in semiconductor nanowires11,28. Single electron transistors offer a convenient tool to study the real-time dynamics of charges in quantum dots11. Furthermore, electronic currents through quantum systems can be used to probe quantum effects arising from the interaction with phonon modes17 which enables phonon spectroscopy8,26,45,56 as well as to understand vibrational induced decoherence4,29. Accompanying experimental results5,20,38,61, a great effort was done in theoretical works to interpret new effects19,52 and to encourage further studies24,26. The first approaches either studied the weak-coupling regime3,27,43 of electron-phonon interactions or considered configurations with a large applied voltage bias8,57. More recently, a strong focus has been put on arbitrarily strong electron-phonon coupling34,41,42,44,59 but these calculations were mainly done for infinite external bias configurations. Still, these studies reveal new interesting phenomena such as giant Fano factors31,33 and Franck-Condon blockade13,35,46,54. Consequently, it is natural to extend the knowledge and mathematical tools to the finite bias regime which was already accessed using numerical techniques1. However, we have observed that naive generalizations of well known infinite bias results to the finite bias regime may be thermodynamic inconsistent, which may e.g. manifest itself in non-vanishing currents in equilibrium setups. With renewed recent interest in generalizations of the phonon master equation to the finite bias limit40,58, it becomes important to present a thermodynamic analysis of the latter. Here, the fluctuation theorem12 offers a well known tool for proving thermodynamic consistency and for understanding the thermodynamic properties of a model because it directly confirms the second law of thermodynamics15,16,52. In particular, in this paper we present a double quantum dot model coupled to macroscopic electronic leads and a phonon reservoir.

The model is introduced in Sec. I. We put emphasis on the polaron transformation and its effect on the model Hamiltonian in terms of thermodynamic consistency. Staying in the polaron picture, we give a detailed description of the derivation of the quantum master equation in Sec. II and prove its thermodynamic consistency by deriving the fluctuation theorem in Sec. III. Finally, in Sec. IV, we analyze electronic current and dephasing rate for particular physical situations showing a Franck-Condon-like suppression in both quantities. We also investigate the possibility of phonon spectroscopy experiments. In addition, we discuss the performance of the model system as a thermoelectric generator converting a temperature gradient into useful power.

I. MODEL

A. Hamiltonian

We consider a system made of a double quantum dot in contact with multiple reservoirs \( \mathcal{H} = \mathcal{H}_S + \mathcal{H}_B + \mathcal{H}_{SB} \). The reservoirs \( \mathcal{H}_B = \mathcal{H}^{B}_R + \mathcal{H}^{B}_L \) and the system-bath coupling \( \mathcal{H}_{SB} = \mathcal{H}^{SB}_{el} + \mathcal{H}^{SB}_{ph} \) contain electronic and phonon contributions, respectively. The system Hamiltonian reads

\[
\mathcal{H}_S \equiv \epsilon_{\sigma} d_{\sigma}^\dagger d_{\sigma} + \epsilon_R d_R^\dagger d_R + T_{e}(d_{L} d_{R}^\dagger + d_{R} d_{L}^\dagger)
+ U d_{L}^\dagger d_{L} d_{R}^\dagger d_{R},
\]

where \( d_{\sigma}(d_{\sigma}^\dagger) \) annihilates (creates) an electron in dot \( \sigma \) with on-site energy \( \epsilon_{\sigma} \) (\( \sigma \in \{L, R\} \) throughout this pa-
unitary Lang-Firzov (polaron) transformation
phonon coupling on electronic transport we perform the
with the phononic absorption/emission amplitudes
occupation of the system induces vibrations in the phonon
phonon in mode \( q \) with phonon operator \( a_q \) annihilating (creates) electrons in mode \( k \) with energy \( \varepsilon_{k,\sigma} \). Note that we do not distinguish between the electronic spins, which implicitly assumes that e.g. the leads are completely polarized. Electronic transport through the system is enabled by the dot-lead interaction Hamiltonian
with the tunneling amplitudes \( t_{k,\sigma} \) (which we will treat perturbatively to second order later-on). Additionally, the system is coupled to a bosonic heat bath
with phonon absorption/emission amplitudes \( h_{q,\sigma} \) (which we will treat non-perturbatively later-on).

B. Polaron transformation

In order to investigate the impact of strong electron-phonon coupling on electronic transport we perform the unitary Lang-Firzov (polaron) transformation\(^6\), \( \hat{H} = U\hat{H}U^\dagger \), with the unitary operator \( U = e^{i\int_t^L d_t \alpha_L d_t^\dagger + d_t^\dagger \alpha_L} \). The anti-hermitian operator \( \hat{B}_\alpha \) is defined as
The details of the polaron transformation are shown in Appendix A. After the polaron transformation, the Hamiltonian admits a new decomposition into system, interaction, and reservoir contributions. It is important to note, however, that in general such decompositions are not unique: For example, for a system Hamiltonian \( \hat{H}_S \) and an interaction Hamiltonian of the general form

\( H_1 \to \sum_\alpha A_\alpha \otimes (B_\alpha - \kappa_\alpha \mathbb{1}) \) with numbers \( \kappa_\alpha \) leaves the total Hamiltonian invariant.

We remove this ambiguity by demanding that all thermal equilibrium expectation values of linear bath coupling operators should vanish. When the \( \langle B_\alpha \rangle \) do not vanish a priori, this requires to fix the numbers \( \kappa_\alpha \) as

\( \kappa_\alpha = \langle B_\alpha \rangle \). (7)

The total Hamiltonian can then be written as \( \hat{H} = \hat{H}_S + \hat{H}_B + \hat{H}_{SB} \) with the system contribution

\[ \hat{H}_S = \varepsilon_L d_L^\dagger d_L + \varepsilon_R d_R^\dagger d_R + U d_L^\dagger d_R d_R^\dagger d_L + (T_c e^{-2i\Phi} d_L^\dagger d_L^\dagger + T_c^* e^{+2i\Phi} d_R d_R) \] (8)

with renormalized local energy levels

\[ \varepsilon_\sigma \equiv \varepsilon_\sigma - \sum_q \frac{|h_{q,\sigma}|^2}{\omega_q} \] (9)

renormalized Coulomb repulsion

\[ \tilde{U} \equiv U - \sum_q \frac{h_{q,L}^2 h_{q,R} + h_{q,L} h_{q,R}^2}{\omega_q} \] (10)

and renormalized internal tunneling amplitude

\[ \tilde{T}_c \equiv T_c \kappa , \] (11)

where the complex-valued \( \kappa \) is defined by \( \kappa \equiv \langle e^{-\beta_{ph} B} \rangle \).

For a phonon reservoir in thermal equilibrium this yields \( \hat{H}_S = \hat{H}_{SB} \) (see Appendix B)

\[ \kappa = e^{-\sum_q \frac{|h_{q,L} - h_{q,R}|^2}{\omega_q^2} (\frac{1}{2} + n_{\omega_q})} e^{-i\Phi} \] (12)

containing the Bose-distribution \( n_B(\omega) = [e^{\beta_{ph} \omega} - 1]^{-1} \) with the inverse phonon bath temperature \( \beta_{ph} \). Here, the phase \( \Phi \) is defined via

\[ i\Phi \equiv \langle [B_L, B_R]/2 \rangle = \sum_q \frac{h_{q,L}^2 h_{q,R} - h_{q,L} h_{q,R}^2}{2\omega_q^2} \] (13)

We note that in the strong-coupling limit, attractive effective Coulomb interactions are in principle possible\(^2\).\(^3\). The bath Hamiltonian effectively retains its form

\[ \hat{H}_B = \sum_q \sum_{\sigma \in \{L,R\}} \varepsilon_{k,\sigma} c_{k,\sigma}^\dagger c_{k,\sigma} + \sum_q \omega_q a_q^\dagger a_q \] (14)

However, the interaction Hamiltonian \( \hat{H}_{SB} = \hat{H}_V + \hat{H}_T \) is made of two parts. The first describes electronic transitions between system and leads

\[ \hat{H}_V \equiv \sum_k (t_{k,L} d_L e^{-d_L^\dagger d_L i\Phi} e^{-B} c_{k,L}^\dagger + \text{h.c.}) \]

\[ + \sum_k (t_{k,R} d_R e^{+d_R^\dagger d_R i\Phi} e^{-B} c_{k,R}^\dagger + \text{h.c.}) , \] (15)

\( T_c \) is the internal electronic tunneling amplitude, and \( U \) is the Coulomb repulsion energy. The system is connected to two electronic leads left and right held at thermal equilibrium

\[ \mathcal{H}_{SB}^{cl} \equiv \sum_{k,\sigma \in \{L,R\}} (t_{k,\sigma} d_{\sigma} c_{k,\sigma}^\dagger + \text{h.c.}) , \] (3)

with the tunneling amplitudes \( t_{k,\sigma} \) (which we will treat perturbatively to second order later-on). Additionally, the system is coupled to a bosonic heat bath

\[ \mathcal{H}_{SB}^{ph} \equiv \sum_q \sum_{\sigma \in \{L,R\}} (h_{q,\sigma} a_q + \text{h.c.}) d_{\sigma}^\dagger d_{\sigma} , \] (5)

with the phononic absorption/emission amplitudes \( h_{q,\sigma} \) (which we will treat non-perturbatively later-on).
which are accompanied by multiple phonon emissions or absorptions. The second part describes transitions between left and right dots

\[ \mathcal{H}_T \equiv T e^{-2iF_dL_d^R t} (e^{-B_eL_e+B_R - \kappa}) + T e^{2iF_dL_d^R t} (e^{-B_eL_e-B_R - \kappa^*}) , \] (16)

which are also dressed by multiple phonon excitations, see Eq. (6).

The effect of the polaron transformation is visualized in Fig. 1. It is important to note that the coupling to the phonon modes is no longer linear in the annihilation and creation operators anymore, as can be seen by expanding the exponentials \( e^{\pm B_\alpha} \). Comparing the system Hamiltonians before and after the polaron transformation, we see that apart from the renormalized on-site energies and Coulomb repulsion the electron-phonon interaction also renormalizes the internal tunneling term. Consequently, the energy eigenbasis of \( \mathcal{H}_S \) is determined by the system-reservoir interaction strength in the original frame.

II. MASTER EQUATION IN THE STRONG ELECTRON-PHONON COUPLING LIMIT

The most general form of an interaction Hamiltonian is a decomposition into system (\( A_\alpha \)) and bath (\( B_\alpha \)) operators (here in the Schrödinger picture)

\[ \mathcal{H}_{SB} = \sum_\alpha A_\alpha \otimes B_\alpha . \] (17)

Such a tensor product decomposition is possible also for fermionic tunneling terms since one can map the fermionic operators to system and lead fermions via a Jordan-Wigner transform\(^{49}\). In standard derivations an often-used assumption is that the expectation values of all bath-coupling operators taken with respect to the equilibrium reservoir state \( \rho_B = e^{-\beta (H_B - \mu N_B)} / \text{Tr} \{ e^{-\beta (H_B - \mu N_B)} \} \) vanish, i.e., \( \langle B_\alpha \rangle = 0 \) for all \( \alpha \). It is crucial to note here that this assumption is fulfilled by fixing the shift \( \kappa \) to the value in Eq. (12).

Without the proper shift, the derivation would yield a Lindblad master equation which is thermodynamic inconsistent, and as an unphysical consequence, at zero bias configurations with equal temperatures the system does not thermalize and non-vanishing currents occur even at equilibrium. However, at infinite bias plus the wide-band limit one would also without shift reproduce well-known and tested results for the electronic current\(^{14,23,53}\) for vanishing electron-phonon coupling. This suggests that one has to be careful with generalizing infinite bias results to the finite bias regime.

We now proceed in the interaction picture (bold symbols), where system operators transform via \( A_\alpha(t) = e^{+iH_L t} A_\alpha e^{-iH_L t} \) and bath operators via \( B_\alpha(t) = e^{+iH_B t} B_\alpha e^{-iH_B t} \). Furthermore, the system operators can be decomposed as \( A_\alpha(t) = \sum_{ab} A_\alpha^{ab} \langle a | \langle b | e^{i(\epsilon_a - \epsilon_b)t} \rangle \rangle \),

\[ A_1 = d_L = (A_2)^\dagger, \ A_4 = d_R^\dagger = (A_4)^\dagger, \ A_5 = e^{-2iF_dL_d^R t} (A_6)^\dagger, \] (18)

FIG. 1: Sketch of the model before (top) and after (below) the polaron transformation. The double quantum dot system in serial configuration is coupled to electronic leads left and right each following Fermi-Dirac statistics with Fermi functions \( f_L \) and \( f_R \), respectively. If either temperatures or chemical potentials are chosen differently, a non-equilibrium situation is created which enables the exchange of matter and energy between those baths. The tunneling between system and leads is described by the tunneling rates \( \Gamma_L \) and \( \Gamma_R \). The quantum tunneling between left and right dot is modulated by the internal tunneling rate \( T_c \). Before the polaron transformation (with \( S = \sum_\sigma d_\sigma^\dagger B_\sigma \)) the phonon bath with Bose distribution \( \rho_B \) couples directly to the occupation of the quantum dots left and right. Due to the polaron transformation the coupling is shifted to the electronic jumps which now occur with multiple phonon emission or absorption processes. Another feature of the model in the polaron picture are the renormalized on-site energies and Coulomb repulsion which now depend on the phonon coupling strength so that the system energy eigenbasis \( \mathcal{H}_S \) is determined by the system-reservoir interaction strength in the original frame.
To make sure that we derive a Lindblad equation in integral form into the commutator yields with the von-Neumann equation (given in the interaction picture a continuum of phonon modes one may also obtain a bath correlation function. The expectation value of two bath operators defines the density matrix of the complete system obeys the master equation in the polaron frame, we will perform a derivation of a thermodynamic consistent low-dimensional master equation. In the following, we will present a derivation of a thermodynamic consistent low-dimensional master equation. In superoperator notation (i.e., arranging the density matrix elements in a vector containing first the populations and then the coherences), the time evolution of the system density matrix can be written as \( \dot{\rho}_S(t) = \mathcal{L}^{\text{pop}} \rho_S(t) + \mathcal{L}^{\text{ph}} \rho_S(t) \). The Lindblad Liouvillian \( \mathcal{L} \) is of dimension 16 × 16 and has in the system energy eigenbasis a block structure letting populations and coherences evolve independently. The block responsible for the populations \((4 \times 4)\) constitutes a rate equation, where the rate matrix is additively decomposable as \( \mathcal{L}^{\text{pop}} = \sum_{\sigma \in \{L,R\}} \mathcal{L}^{\text{pop}}_{\sigma} + \mathcal{L}^{\text{ph}} \). Here, \( \mathcal{L}^{\text{ph}} \) describes dressed electronic jumps between lead \( \sigma \) and system and \( \mathcal{L}^{\text{ph}} \) describes dressed electronic transitions within the system. We will denote the rates responsible for a transition from eigenstate \( j \) to eigenstate \( i \) by \( \Gamma_{ij}^\nu \), where \( \nu = L/R \) corresponds to transitions with an electronic jump over the left/right barrier whilst simultaneously emitting or absorbing phonons and \( \nu = \text{ph} \) correspond to internal electronic transitions between the polaron-transformed energy eigenstates. We label these as \( \{|0\}, \{-\}, \{+\}, \{|2\} \) with system eigenenergies

\[
\begin{align*}
\epsilon_0 \equiv 0, \\
\epsilon_- \equiv \frac{1}{2} \left( \epsilon_L + \epsilon_R - \sqrt{(\epsilon_L - \epsilon_R)^2 + 4|\bar{T}_c|^2} \right), \\
\epsilon_+ \equiv \frac{1}{2} \left( \epsilon_L + \epsilon_R + \sqrt{(\epsilon_L - \epsilon_R)^2 + 4|\bar{T}_c|^2} \right), \\
\epsilon_2 \equiv \epsilon_L + \epsilon_R + \bar{U}.
\end{align*}
\]

The Fourier transforms of the correlation functions

\[
\gamma_{\alpha\beta}(\omega) = \int dt e^{i\omega t} C_{\alpha\beta}(t)
\]
and matrix elements of the coupling operators enter the transition rates from energy eigenstate $b$ to eigenstate $a$ via

$$
\gamma_{ab,ab} = \sum_{\alpha\beta} \gamma_{\alpha\beta}(\varepsilon_b - \varepsilon_a) \langle a | A_{\beta} | b \rangle \langle a | A_{\alpha} | b \rangle^* .
$$

(26)

Explicitly, the non-vanishing rates $\Gamma_{ab}^\sigma$ from energy eigenstate $|b\rangle$ to energy eigenstate $|a\rangle$ associated with an electronic jumps to or from reservoir $\sigma \in \{L, R\}$ then become

$$
\begin{align*}
\Gamma_{L}^0 & = \gamma_{12}(\varepsilon_- - \varepsilon_0) \langle 0 | d^\dagger_L | - \rangle^2 , \\
\Gamma_{L}^{-0} & = \gamma_{12}(\varepsilon_0 - \varepsilon_-) \langle - | d^\dagger_L | 0 \rangle^2 , \\
\Gamma_{L}^{0+} & = \gamma_{12}(\varepsilon_+ - \varepsilon_0) \langle 0 | d^\dagger_L | + \rangle^2 , \\
\Gamma_{L}^{0-} & = \gamma_{12}(\varepsilon_0 - \varepsilon_+) \langle + | d^\dagger_L | 0 \rangle^2 , \\
\Gamma_{L}^{-2} & = \gamma_{12}(\varepsilon_2 - \varepsilon_-) \langle - | d^\dagger_L | 2 \rangle^2 , \\
\Gamma_{L}^{2-} & = \gamma_{12}(\varepsilon_- - \varepsilon_2) \langle 2 | d^\dagger_L | - \rangle^2 , \\
\Gamma_{L}^{2+} & = \gamma_{12}(\varepsilon_2 - \varepsilon_+) \langle + | d^\dagger_L | 2 \rangle^2 , \\
\Gamma_{R}^{0-} & = \gamma_{34}(\varepsilon_0 - \varepsilon_-) \langle - | d^\dagger_R | 0 \rangle^2 , \\
\Gamma_{R}^{0+} & = \gamma_{34}(\varepsilon_+ - \varepsilon_0) \langle 0 | d^\dagger_R | + \rangle^2 , \\
\Gamma_{R}^{0-} & = \gamma_{34}(\varepsilon_0 - \varepsilon_+) \langle + | d^\dagger_R | 0 \rangle^2 , \\
\Gamma_{R}^{-2} & = \gamma_{34}(\varepsilon_2 - \varepsilon_-) \langle - | d^\dagger_R | 2 \rangle^2 , \\
\Gamma_{R}^{2-} & = \gamma_{34}(\varepsilon_- - \varepsilon_2) \langle 2 | d^\dagger_R | - \rangle^2 , \\
\Gamma_{R}^{2+} & = \gamma_{34}(\varepsilon_2 - \varepsilon_+) \langle + | d^\dagger_R | 2 \rangle^2 , \\
\Gamma_{R}^{2+} & = \gamma_{34}(\varepsilon_+ - \varepsilon_2) \langle 2 | d^\dagger_R | + \rangle^2 .
\end{align*}
$$

(27)

We note that the matrix elements in the rates describing backward and forward processes triggered by the same reservoir are identical, such that local detailed balance is only induced by a corresponding Kubo-Martin-Schwinger (KMS)-type condition of the correlation functions. We discuss these in Sec. II A 1.

As a distinctive feature in comparison to a single quantum dot\textsuperscript{50}, one now obtains phonon-modified internal transitions, and the corresponding rates between energy eigenstates $|\rangle$ and $|+\rangle$ can be written as a quadratic form

$$
\begin{align*}
\Gamma_{\text{ph}}^{-+} & = (A_5^{-+}, (A_5^{-+})^*) \gamma(\varepsilon_+ - \varepsilon_-) \langle A_5^{-+} | A_5^{-+} \rangle^* , \\
\Gamma_{\text{ph}}^{++} & = (A_5^{++}, (A_5^{++})^*) \gamma(\varepsilon_+ - \varepsilon_-) \langle A_5^{++} | A_5^{++} \rangle^* .
\end{align*}
$$

(28)

with the matrix $\gamma(\omega)$ being given by

$$
\gamma(\omega) = \begin{pmatrix}
\gamma_{66}(\omega) & \gamma_{55}(\omega) \\
\gamma_{65}(\omega) & \gamma_{65}(\omega)
\end{pmatrix} .
$$

(29)

It can be shown that this matrix is hermitian and positive definite, such that we obtain true rates $\Gamma_{\text{ph}}^{++} \geq 0$ and $\Gamma_{\text{ph}}^{-+} \geq 0$. Furthermore, we note that since the correlation functions contained in the matrix (29) obey KMS relations of the form $\gamma_{\alpha\beta}(-\omega) = \gamma_{\beta\alpha}(+\omega)e^{-\beta_{\text{ph}}\omega}$ with inverse phonon reservoir temperature $\beta_{\text{ph}}$ (compare Sec. II A 2), this implies for the ratio of rates $\Gamma_{\text{ph}}^{++}/\Gamma_{\text{ph}}^{-+} = e^{-\beta_{\text{ph}}(\varepsilon_+ - \varepsilon_-)}$.

1. Lead-Phonon Correlation Function

From Eq. (20) it follows that the four non-vanishing contributions associated with electronic jumps into or out of the system can be written in a product form of electronic and phononic contributions\textsuperscript{50}

$$
C_{\alpha\beta}(\tau) = C_{\alpha\beta}^e(\tau)C_{\alpha\beta}^\text{ph}(\tau) ,
$$

(30)

with the electronic parts being given by

$$
\begin{align*}
C_{12}^e(\tau) & = \sum_k |t_{k\ell}|^2 f_L(\varepsilon_{k\ell})e^{i\varepsilon_{k\ell}\tau}, \\
C_{21}^e(\tau) & = \sum_k |t_{k\ell}|^2 [1 - f_L(\varepsilon_{k\ell})]e^{-i\varepsilon_{k\ell}\tau}, \\
C_{34}^e(\tau) & = \sum_k |t_{k\ell}|^2 f_R(\varepsilon_{k\ell})e^{i\varepsilon_{k\ell}\tau}, \\
C_{24}^e(\tau) & = \sum_k |t_{k\ell}|^2 [1 - f_R(\varepsilon_{k\ell})]e^{-i\varepsilon_{k\ell}\tau} .
\end{align*}
$$

(31)

Here, we have introduced the Fermi function $f_\sigma(\omega) \equiv [e^{\beta_\sigma(\omega - \mu_\sigma)} + 1]^{-1}$ of lead $\sigma$ with inverse temperature $\beta_\sigma$ and chemical potential $\mu_\sigma$. The tunneling amplitudes $t_{k\ell}$ lead to effective tunneling rates $\Gamma_\sigma(\omega) \equiv 2\pi \sum_k |t_{k\ell}|^2 \delta(\omega - \varepsilon_{k\ell})$, which can be used to convert the above summations into integrals. Later-on, we will parameterize them with a Lorentzian distribution\textsuperscript{62}

$$
\Gamma_\sigma(\omega) \equiv \frac{\Gamma_\sigma^0}{\omega^2 + \delta_\sigma^2} ,
$$

(32)

yielding a representation in terms of hypergeometric functions for $C_{\alpha\beta}^\text{ph}(\tau)$, which we omit here for brevity. For completeness we note that the separate Fourier transforms of the electronic parts $\gamma_{\alpha\beta}^e(\omega) = \int C_{\alpha\beta}^e(\tau)e^{i\omega\tau}d\tau$

$$
\begin{align*}
\gamma_{12}^e(\omega) & = \Gamma_L(\varepsilon_-)f_L(\varepsilon_+), \\
\gamma_{21}^e(\omega) & = \Gamma_L(\varepsilon_+)[1 - f_L(\varepsilon_-)], \\
\gamma_{34}^e(\omega) & = \Gamma_R(\varepsilon_-)f_R(\varepsilon_+), \\
\gamma_{43}^e(\omega) & = \Gamma_R(\varepsilon_+)[1 - f_R(\varepsilon_-)],
\end{align*}
$$

(33)

obey – since $f_\sigma(\omega) = e^{-\beta_\sigma(\omega - \mu_\sigma)}[1 - f_\sigma(\omega)]$ – the KMS-type relations

$$
\begin{align*}
\gamma_{12}^e(\omega) & = e^{-\beta_\text{ph}(\varepsilon_- - \mu_\sigma)}\gamma_{21}^e(\omega), \\
\gamma_{34}^e(\omega) & = e^{-\beta_\text{ph}(\varepsilon_+ - \mu_\sigma)}\gamma_{43}^e(\omega) .
\end{align*}
$$

(34)
The phonon contribution to the correlation function depends only on the terminal across which the electron jumps but not on the jump direction, i.e., we have $C_{12}^{\text{ph}}(\tau) = C_{21}^{\text{ph}}(\tau) = C_{L}^{\text{ph}}(\tau)$ and $C_{34}^{\text{ph}}(\tau) = C_{43}^{\text{ph}}(\tau) = C_{R}^{\text{ph}}(\tau)$. Using the Baker-Campbell-Hausdorff (BCH) formula, the phonon contribution explicitly computes to (see Appendix D1)

$$C_{\sigma}^{\text{ph}}(\tau) = e^{-K_{\sigma}(0)}K_{\sigma}(\tau),$$

with the abbreviation in the exponent

$$K_{\sigma}(\tau) = \sum_{q} \frac{|h_{q,\sigma}|^2}{\omega_q^2} \times$$

$$\times \left\{ n_{B}(\omega_q)e^{i\omega_q\tau} + [n_{B}(\omega_q) + 1]e^{-i\omega_q\tau} \right\} .$$

It is easy to show that $K_{\sigma}(\tau) = K_{\sigma}(-\tau - i\beta_{\text{ph}})$ holds, which transfers to the KMS condition for the phonon contribution to the correlation function

$$C_{\sigma}^{\text{ph}}(\tau) = C_{\sigma}^{\text{ph}}(-\tau - i\beta_{\text{ph}}).$$

The nature of the phonon contributions can now be quite distinct depending on whether one has a discrete (e.g. just a single mode) or continuous spectrum of phonon frequencies. In the continuum case, we can convert the sum in the exponent into an integral. Then, the phonon absorption emission amplitudes enter the corresponding rate as $J_{\sigma}(\omega) \equiv \sum_{q} |h_{q,\sigma}|^2 \delta(\omega - \omega_q)$, where $J_{L}(\omega)$ and $J_{R}(\omega)$ will be parametrized by a continuous function. For example, using the super-ohmic parameterization with exponential infrared cut-off at $\omega_{c}^2$ (we choose a super-ohmic representation to enable a Markovian description of the internal jumps in Sec. II A 2 too) and coupling strength $J_{\sigma}$, i.e.,

$$J_{\sigma}(\omega) \equiv J_{\sigma}\omega^{2}e^{-\frac{\omega^{2}}{\omega_{c}^{2}}},$$

we obtain for the integrals in the exponent

$$K_{\sigma}(\tau) = \int_{0}^{\infty} J_{\sigma}(\omega^{2}) \left\{ n_{B}(\omega)e^{i\omega\tau} + [n_{B}(\omega) + 1]e^{-i\omega\tau} \right\} d\omega$$

$$= \frac{2J_{\sigma}}{\beta^{2}} \Psi\left(1 + \frac{i\tau\omega_{c}^{2}}{\beta_{\text{ph}}\omega_{c}^{2}}\right) - \frac{J_{\sigma}(\omega_{c}^{2})^{2}}{(1 - i\tau\omega_{c}^{2})^{2}},$$

(39)

where $\Psi(x)$ denotes the derivative of the PolyGamma function $\Psi(x) = \Gamma'(x)/\Gamma(x)$. With the same super-ohmic spectral density, the renormalized on-site energies and Coulomb shift read explicitly

$$\xi_{\sigma} = \epsilon_{\sigma} - 2J_{\sigma}\omega_{c}^{2},$$

$$\bar{U} = U + \sum_{q} \frac{|h_{qL} - h_{qR}|^{2}}{\omega_{q}} - |h_{qL}|^{2} - |h_{qR}|^{2}$$

$$= U + 2J_{0}\omega_{c}^{2} - 2J_{L}(\omega_{c}^{2})^{3} - 2J_{R}(\omega_{c}^{2})^{3} .$$

We note here that since $K_{\sigma}(\tau)$ in Eq. (39) decays to zero for large $\tau$, the phonon correlation function $C_{\sigma}^{\text{ph}}(\tau)$ may remain finite for large $\tau$. Thanks to the influence of the electronic contributions the total correlation function will still decay, such that a Markovian approach is applicable. In this case we technically define separate Fourier transforms of the phonon contributions by

$$\gamma_{\sigma}^{\text{ph}}(\omega) = \int \left[ C_{\sigma}^{\text{ph}}(\tau) - C_{\sigma}^{\text{ph}}(\infty) \right] e^{i\omega\tau} d\tau$$

$$+ 2\pi C_{\sigma}^{\text{ph}}(\infty) \delta(\omega) .$$

(41)

Since the dressed correlation functions are given by products of electronic and phononic contributions in the time domain, the separate KMS relations (34) and (37) do not directly transfer in non-equilibrium setups. However, we can use our previous result (see appendix of Ref.50) that these correlation functions can be written conditioned upon the net number of emitted phonons into the different reservoir modes ($n_{q} < 0$ implies absorption from the phonon reservoir).

Formally, one has $\gamma_{\alpha\beta}(\omega) = \sum_{n} \gamma_{\alpha\beta,\mathbf{n}}(\omega)$, where the separate contributions are given by ($\Omega = (\omega_{1}, \ldots, \omega_{Q})$)

$$\gamma_{\alpha\beta,\mathbf{n}}(\omega) = \gamma_{\alpha\beta}^{d}(\omega - n \cdot \Omega) \prod_{q} e^{-\frac{|h_{q}|^{2}}{2}(1 + 2n_{q})} \times$$

$$\times \left(1 + \frac{n_{q}}{n_{B}}\right)^{n_{q}/2} \times$$

$$\times J_{n} \left(2\frac{|h_{q}|^{2}}{\omega_{q}^{2}}\sqrt{n_{q}^{0}(1 + n_{q}^{0})} \right) .$$

(42)

with $J_{n}(x) \equiv \sum_{k=0}^{\infty} \{(\omega^{2}/k!)\Gamma[k+n+1](x/2)^{2k+n}$

being the modified Bessel function of the first kind and $\Gamma[x] = \int_{0}^{\infty} e^{t-x} t^{k} dt$ being the Gamma-function. We note that when the electronic Fourier transforms are flat $\gamma_{\alpha\beta}^{d}(\omega - n \cdot \Omega) \rightarrow \gamma_{\alpha\beta}^{d}$, the normalization of the phonon contribution implies that the Fourier transform of the combined correlation function is also flat $\sum_{n} \gamma_{\alpha\beta,\mathbf{n}}(\omega) \rightarrow \gamma_{\alpha\beta}^{d}$. This implies that in the electronic wide-band ($\delta_{\sigma} \rightarrow \infty$) plus the infinite bias ($f_{L}(\omega) \rightarrow 1$ and $f_{R}(\omega) \rightarrow 0$) limits the phonons will have no effect on the dot-lead correlation functions.

Importantly, we note that even for different temperatures, these obey the KMS-type relation

$$\gamma_{12,+,\mathbf{n}_{L}}(-\omega) = e^{-\beta_{L}(\omega - n_{L} - \mathbf{n}_{L} \cdot \Omega)} e^{\beta_{\text{ph}}n_{L} \cdot \Omega} \times$$

$$\times \gamma_{21,-,\mathbf{n}_{L}}(\omega) ,$$

$$\gamma_{34,+,\mathbf{n}_{R}}(-\omega) = e^{-\beta_{R}(\omega - n_{R} - \mathbf{n}_{R} \cdot \Omega)} e^{\beta_{\text{ph}}n_{R} \cdot \Omega} \times$$

$$\times \gamma_{43,-,\mathbf{n}_{R}}(\omega) .$$

(43)

We see that the conventional KMS relation is reproduced when phonon and electronic temperatures are equal.
2. Interdot-Phonon Correlation Function

To evaluate the transitions between the states $|-\rangle \leftrightarrow |+\rangle$, we have to evaluate the correlation functions

\[
\begin{align*}
C_{55}(\tau) &= \langle e^{-B_L(\tau)} e^{+B_R(\tau)} e^{-B_L e^{+B_R}} \rangle - \kappa^2, \\
C_{60}(\tau) &= \langle e^{-B_R(\tau)} e^{+B_L(\tau)} e^{-B_R e^{+B_L}} \rangle - (\kappa^*)^2, \\
C_{66}(\tau) &= \langle e^{-B_L(\tau)} e^{+B_R(\tau)} e^{-B_R e^{+B_L}} \rangle - |\kappa|^2, \\
C_{65}(\tau) &= \langle e^{-B_R(\tau)} e^{+B_L(\tau)} e^{-B_L e^{+B_R}} \rangle - |\kappa|^2,
\end{align*}
\]

where we have used that $\kappa = \langle e^{-B_L e^{+B_R}} \rangle = \langle e^{-B_L(\tau)} e^{+B_R(\tau)} \rangle$ is inert with respect to transformations into the interaction picture. For the first bath correlation functions we obtain (see Appendix D 2)

\[
C_{55}(\tau) = \kappa^2 \left[ e^{-K(\tau)} - 1 \right],
\]

where – in analogy to Eq. (36) – we have

\[
K(\tau) = \sum_q \left( \frac{h_{q,L} - h_{q,R}}{\omega_q^2} \right) \times
\]

\[
\times \left\{ n_B(\omega_q) e^{i \omega_q \tau} + [n_B(\omega_q) + 1] e^{-i \omega_q \tau} \right\}.
\]

We note that for large times the correlation function vanishes for a continuum of phonon modes, facilitating a Markovian description. Two further correlation functions can be similarly evaluated

\[
C_{66}(\tau) = (\kappa^*)^2 \left[ e^{-K(\tau)} - 1 \right] = C_{55}(-\tau),
\]

where the latter equality can be easily seen by direct comparison. For the third correlation function we find

\[
C_{65}(\tau) = |\kappa|^2 \left[ e^{K(\tau)} - 1 \right].
\]

It can be easily seen that $C_{60}(t) \equiv C_{65}(t)$. Furthermore, we note that

\[
\kappa^2 = e^{-K(0)} e^{2\Phi}, \quad (\kappa^*)^2 = e^{-K(0)} e^{-2\Phi}, \\
|\kappa|^2 = e^{-K(0)}.
\]

From $K(-\tau) = K^*(+\tau)$ we conclude that the Fourier transform matrix of these correlation functions (29) is hermitian. It can be expressed by the two real-valued functions

\[
\gamma_\pm(\omega) = \int \left( e^{\pm K(\tau)} - 1 \right) e^{\pm i \omega \tau} d\tau
\]

and will be positive definite at frequency $\omega$ when $\gamma_-(\omega) < 0$ or, equivalently, when $\gamma_+^2(\omega) - \gamma_-^2(\omega) = [\gamma_+(\omega) - \gamma_-(\omega)][\gamma_+(\omega) + \gamma_-(\omega)] > 0$. The interdot phonon correlations obey KMS relations of the type (for $\alpha, \beta \in \{5, 6\}$)

\[
C_{\alpha\beta}(\tau) = C_{\beta\alpha}(-\tau - i\beta_{ph}),
\]

which follow from the definition of $K(\tau)$. For their Fourier transforms this implies $\gamma_{\alpha\beta}(-\omega) = \gamma_{\beta\alpha}(+\omega) e^{-i\beta_{ph}}$.

Finally, we note that this approach is valid for coupling to a continuum of phonon modes. A finite number of phonon modes would in general not lead to a decay of the inter-dot correlation functions $C_{55}(\tau)$, $C_{60}(\tau)$, $C_{65}(\tau)$, and $C_{66}(\tau)$, thus prohibiting a Markovian description. Furthermore, the electronic tunneling Hamiltonian $\mathcal{H}_V$ and the inter-dot tunneling Hamiltonian $\mathcal{H}_T$ must be small in the polaron frame. The first condition is consistent with a perturbative treatment of electron-lead tunneling amplitudes, whereas the second condition can be fulfilled by choosing either nearly symmetric electron-phonon couplings left and right, i.e. $h_{q,L} \approx h_q \approx h_{q,R}$ or by treating $T_e$ also perturbatively. If the electron-phonon coupling is exactly symmetric, also finite phonon modes can be treated with the approach.

3. Numerical phonon correlation function

In case of a continuous phonon spectrum, the Fourier transforms of the phonon correlation functions associated with external – compare Eq. (35) – and internal – compare Eqs. (45), (47), and (48) – electronic jumps cannot be obtained analytically in closed form. This complicates the calculation of the full transition rates whenever one is also interested in the heat exchanged with the phonon reservoir, as this requires evaluation of a convolution integral, where the phonon contribution to the integrand is itself a numerical Fourier integral. Here, we therefore aim to represent the Fourier-transform of the phonon contribution in a semi-exact fashion, respecting the thermodynamic KMS relations. For this, we note that the Gaussian

\[
\gamma_{ph}^{fit}(\omega) = a e^{-\left(\omega - \beta_{ph} \omega / 4\right)^2 / b}
\]

obeys for all fit parameters $a$ and $b$ and frequencies $\omega$ the KMS relation $\gamma_{ph}^{fit}(\omega) e^{\beta_{ph} \omega} = \gamma_{ph}(\omega)$, where $\beta_{ph}$ denotes the inverse phonon temperature. Naturally, by fitting the phonon correlation functions e.g. with multiple such Gaussian functions one would obtain a thermodynamic correct representation of the phonon correlation function. Here however, we are rather interested in thermodynamic principles and just use a single Gaussian function, where we fix the fit parameters by crudely matching $C_{ph}^{fit}(0)$ and $\int C_{ph}^{fit}(\tau) d\tau$ with the true values of the correlation function. We note that both $C_{ph}(0)$ and $\int C_{ph}(\tau) d\tau$ are always real-valued, such that the Fourier transform of the Gaussian approximation does not only obey the KMS condition but is also always positive.
III. SYMMETRIES IN THE FULL COUNTING STATISTICS

To deduce the counting statistics not only of electrons but also of the phonons, it would be necessary to identify the phonons emitted or absorbed with every electronic jump. However, here we are rather interested in the energy that by such processes is emitted into or absorbed from the phonon reservoir. For internal electronic transitions, the energy exchange follows directly from the change in the system state. In contrast, for transitions involving an electronic jump across the left or right terminal, one has to identify the separate phononic contributions to correctly partition the electronic and phononic contributions to the exchanged energy.

To identify a minimal set of transitions that has to be monitored for energy and particle exchange, we first consider the entropy production $\dot{S}_i$ in the system, which at steady state must be balanced by the entropy flow $\dot{S}_e$ from the electronic and phononic terminals

$$\dot{S}_i = -\dot{S}_e = -\sum_\nu \beta_\nu \dot{Q}_\nu$$

$$= -\beta_L (I_E^L - \mu_L I_M^L) - \beta_R (I_E^R - \mu_R I_M^R) - \beta_{ph} \dot{I}_{ph}^E,$$ (53)

where $I_E^L$, $I_M^L$, and $\dot{Q}_\nu$ denote the energy, matter, and heat currents from terminal $\nu$ into the system, respectively. Using the conservation laws for energy and matter

$$I_E^L + I_M^L + I_{ph}^L = 0, \quad I_E^R + I_M^R = 0,$$ (54)

we can eliminate two currents.

We choose to monitor the number of electrons entering the system from the left lead $I_M^{(L)}$, the energy that is transferred from the left lead into the system $I_E^{(L)}$, and the energy that is transferred from the phonon reservoir into the system $I_{ph}^{(L)}$. In terms of these quantities, the entropy production becomes

$$\dot{S}_i = (\beta_R - \beta_L) I_E^{(L)} + (\beta_L \mu_L - \beta_R \mu_R) I_M^{(L)} + (\beta_R - \beta_{ph}) I_{ph}^{(L)},$$ (55)

which is decomposable into affinities and fluxes. When we further assume that the electronic affinities of both leads are the same $\beta_L = \beta_R = \beta_0$, the entropy production can even be expressed with only two affinities and two fluxes

$$\dot{S}_i = \beta_0 (\mu_L - \mu_R) I_M^{(L)} + (\beta_0 - \beta_{ph}) I_{ph}^{(L)}.$$. (56)

Formally, the statistics of energy and matter transfers can be extracted by complementing the off-diagonal entries in the Liouvillian that describe the individual jump processes with counting fields. For the electronic hopping this is fairly standard and straightforward to do. It becomes a bit more involved however when one is interested in the statistics of energy exchanges: For the internal jumps the complete energy must have been exchanged with the phonon reservoir

$$\Gamma_{ph}^{\pm} \rightarrow \Gamma_{ph}^{\pm} e^{-i(\epsilon_+ - \epsilon_-)},$$

$$\Gamma_{ph}^{\pm} \rightarrow \Gamma_{ph}^{\pm} e^{+i(\epsilon_+ - \epsilon_-)}.$$ (57)

For the electronic jumps between system and both leads we however have to partition the emitted or absorbed energy into contributions from the electronic and phononic reservoirs, which first requires to decompose the transitions into different phonon contributions. Assuming for example a discrete phonon spectrum we have

$$\Gamma_{ij}^\sigma = \sum_n \Gamma_{ij}^n n,$$ (58)

where $\Gamma_{ij}^n$ describes a transition from energy eigenstate $j$ to $i$ together with the emission of $n$ phonons into the different phonon reservoir modes and an electronic jump to or from lead $\sigma \in \{L, R\}$. For a continuous phonon spectrum (which we will not discuss explicitly) we could use the convolution theorem to arrive at a similar decomposition

$$\Gamma_{ij}^\sigma = \int \Gamma_{ij}^\sigma (\omega) d\omega,$$ (59)

where $\Gamma_{ij}^\sigma (\omega)$ describes a transition from energy eigenstate $j$ to $i$ together with the emission of energy $\omega$ into the phonon reservoir and an electronic jump to or from lead $\sigma \in \{L, R\}$. This then implies the counting field replacements for the off-diagonal matrix elements in the Liouvillian

$$\Gamma_{ij,n}^L \rightarrow \Gamma_{ij,n}^L e^{i\int \chi (n_i - n_f) + \chi n_i - n_f n \Omega} e^{-i\phi n \Omega},$$

$$\Gamma_{ij,n}^R \rightarrow \Gamma_{ij,n}^R e^{-i\phi n \Omega},$$ (60)

where $n_i \in \{0, 1, 2\}$ denotes the number of electrons in energy eigenstate $i$. Thus, the Liouvillian is now dependent on the particle counting field $\chi$, the electronic energy counting field $\xi$, and the phonon energy counting field $\phi$, which we may for brevity combine in a vector $\chi = (\chi, \xi, \phi)$. The characteristic polynomial $D(\chi) = |L(\chi) - \lambda|$ of the now counting-field dependent Liouvillian formally equates to

$$D = [L_{11} - \lambda][L_{22} - \lambda][L_{33} - \lambda][L_{44} - \lambda]$$

$$- [L_{11} - \lambda][L_{22} - \lambda][L_{44} - \lambda]$$

$$- [L_{11} - \lambda][L_{33} - \lambda][L_{44} - \lambda]$$

$$- [L_{11} - \lambda][L_{22} - \lambda][L_{33} - \lambda]$$

$$- [L_{22} - \lambda][L_{33} - \lambda][L_{44} - \lambda]$$

$$- [L_{33} - \lambda][L_{44} - \lambda][L_{24} - \lambda]$$

$$+ [L_{11} - \lambda][L_{24} - \lambda][L_{33} - \lambda]$$

$$+ [L_{11} - \lambda][L_{34} - \lambda][L_{22} - \lambda]$$

$$+ [L_{11} - \lambda][L_{44} - \lambda][L_{22} - \lambda]$$

$$+ [L_{22} - \lambda][L_{34} - \lambda][L_{11} - \lambda]$$

$$+ [L_{22} - \lambda][L_{44} - \lambda][L_{11} - \lambda]$$

$$- [L_{11} - \lambda][L_{34} - \lambda][L_{44} - \lambda]$$

$$- [L_{11} - \lambda][L_{44} - \lambda][L_{24} - \lambda]$$

$$- [L_{22} - \lambda][L_{34} - \lambda][L_{44} - \lambda]$$

$$- [L_{22} - \lambda][L_{44} - \lambda][L_{11} - \lambda]$$

where it should be kept in mind that the counting fields only occur in the off-diagonal ($L_{i\neq j}$) contributions. With
the relations \((\sigma \in \{L, R\})\)

\[
\begin{align*}
\Gamma_{\sigma, n}^0 & \equiv e^{+\beta_{n}(\epsilon_{\sigma} - \epsilon_{\sigma})}, \\
\Gamma_{\sigma, n}^0 & \equiv e^{+\beta_{n}((\epsilon_{\sigma} + \epsilon_{\sigma}) + n \Omega)} e^{-\beta_{n} n \Omega}, \\
\Gamma_{\sigma, n}^0 & \equiv e^{+\beta_{n}((\epsilon_{\sigma} - \epsilon_{\sigma}) + n \Omega)} e^{-\beta_{n} n \Omega}, \\
\Gamma_{\sigma, n}^0 & \equiv e^{+\beta_{n}((\epsilon_{\sigma} - \epsilon_{\sigma}) + n \Omega)} e^{-\beta_{n} n \Omega}, \\
\Gamma_{\sigma, n}^0 & \equiv e^{+\beta_{n}((\epsilon_{\sigma} - \epsilon_{\sigma}) + n \Omega)} e^{-\beta_{n} n \Omega}, \tag{61}
\end{align*}
\]

one can show (compare Appendix E) that the characteristic polynomial stays invariant under the replacements

\[
-\chi \rightarrow +\chi + i(\beta_{RL} - \beta_{RR}R), \\
-\xi \rightarrow +\xi + i(\beta_{RR} - \beta_{RL}), \\
-\phi \rightarrow +\phi + i(\beta_{RR} - \beta_{ph}), \tag{62}
\]

where we recover the affinities in Eq. (55). This symmetry transfers to the long-term cumulant-generating function, and thus, the steady state fluctuation theorem for entropy production reads

\[
\lim_{t \rightarrow \infty} P_{n, \epsilon_{\sigma}, \epsilon_{ph}}(t) = e^{n\Delta}, \tag{63}
\]

with \(n \equiv (n_{L}, \epsilon_{L}, \epsilon_{ph})^T\) and \(\Delta \equiv (\beta_{RL} - \beta_{RR}R, \beta_{RR} - \beta_{RL}, \beta_{RR} - \beta_{ph})^T\). Due to the similar three-terminal setup, the same fluctuation theorem can be obtained for the single electron transistor.\(^{50}\)

### IV. RESULTS

The implications of the resulting master equation are of course manifold. Below, we present a selection of the most interesting phonon-induced features. For simplicity, we will discuss the case of symmetric couplings \(h_{qL} \equiv h_{qR}\) here.

#### A. Electronic current versus internal bias

We compute the electronic matter current for coupling to a single phonon mode at frequency \(\Omega\) and also for coupling to a continuum of phonons. Fig. 2 shows the electronic current at infinite external bias \((f_L \rightarrow 1, f_R \rightarrow 0)\) but finite bandwidths as a function of the internal bias \(\Delta \epsilon \equiv \epsilon_{L} - \epsilon_{R}\), which we define symmetrically with \(\epsilon_{L} \equiv +\Delta \epsilon / 2\) and \(\epsilon_{R} \equiv -\Delta \epsilon / 2\). The study of such currents is very common in theoretical\(^ {7,9,47,60}\) as well as experimental studies as they reveal many internal details of the transport setup. In Fig. 2, the black curve shows the pure electronic current without phonons \((h_{qL} = h_{qR} = 0)\) far away from the wide-band limit \((\delta_{L} / \Omega = \delta_{L} / \Omega = 0.1)\). Here, two electronic resonances at \(\pm(\epsilon_{L} - \epsilon_{R}) / 2\) become visible. The Lorentzian shape of the graph is characteristic for such models and stems from the matrix elements in front of the rates. For the colored curves we increase the electron-phonon coupling \((h_{qL} = h_{qR} = h_{q})\) at large phonon bath temperature \(\beta_{ph} \Omega = 0.1\) (due to the infinite-bias assumption the electronic temperature does not enter). Due to the coupling to a single phonon mode we see additional side peaks appearing at \(\Delta \epsilon = 2n \Omega\) with integer \(n\) (see solid red and blue curves), and these completely dominate the electronic peaks in the strong-coupling limit (solid blue). For smaller phonon bath temperatures, the resonances would be more pronounced for positive \(\Delta \epsilon\), since phonon emission into the bath is more likely (not shown). When we couple electronic transport to a continuum of phonon modes these detailed oscillations can not be resolved anymore (dashed curves in the background, see also the figure caption).
B. Current/Dephasing rate versus external bias

Typically, the current as a function of the external bias can be used to obtain internal system parameters via transport spectroscopy: Transition frequencies of the system entering the transport window will – at sufficiently small temperatures – induce steps in the current. In Fig. 3 we display the electronic matter current for different electron-phonon coupling strengths. Whereas – as a consequence of the phonon presence – the single-mode version (solid curves) displays now many additional plateaus that allow e.g. for spectroscopy of the phonon frequency, the continuous phonon versions (dashed and dotted) only display a suppression of the current for small bias. This phenomenon – termed Franck-Condon blockade\cite{32} – is also observed when the phonons are taken into account dynamically.

\[ \text{FIG. 3: Plot of the electronic current versus the external bias voltage for different electron-phonon coupling strengths.} \]

With increasing coupling strength, the steps corresponding to the bare electronic transitions (black curve) become supplemented by additional plateaus accounting for an increasing number of phonons involved in the transport process. The width of these smaller steps allows to determine the phonon frequency. Consistently, the continuum phonon reservoir (dashed curves in background) does not exhibit these smaller steps. Other parameters are chosen as: \( \Gamma / \Omega = 0.01, T / \Omega = 1.0, \beta L / \Omega = \beta R / \Omega = 0, \delta L / \Omega = \delta R / \Omega = 20.0 \). Continuum parameters were adjusted such that \( J_0^\infty J_\sigma (\omega) d\omega = |h|^2 / \Omega \) (dashed curves). Further approximating the continuum phonon correlation function with a single Gaussian as described in Sec. II A 3 yields for small bias quite analogous results (dotted curves).

Computing the dynamics of the coherences \( \langle - | \rho_0(t) | + \rangle = \langle + | \rho_0(t) | - \rangle^* \) yields a time evolution of the form \( \rho_{--}(t) = \gamma \rho_{--}(0) \). Looking at the absolute square of \( \rho_{--}(t) \) we find it decays exponentially with \( |\rho_{--}(t)|^2 = e^{-2\gamma t} |\rho_{--}(0)|^2 \), where this dephasing is induced by both electronic and phononic reservoirs.

The dephasing rate \( \mathcal{R} = 2\Phi / \Omega \) is a measure for the decay of the superposition of the states \( |-\rangle \) and \( |+\rangle \) to a classical mixture. When we neglect the asymmetry of the coupling \( h_{L,R} = h_{R,L} \), such that the internal transition rates in Eq. (29) vanish, we obtain for the dephasing rate

\[ \mathcal{R} = [\Gamma_{L}^{0-} + \Gamma_{R}^{0-} + \Gamma_{L}^{0+} + \Gamma_{R}^{0+} + \Gamma_{L}^{2+} + \Gamma_{R}^{2+} + \Gamma_{L}^{2-} + \Gamma_{R}^{2-}], \tag{64} \]

where we have used the abbreviations defined in Eq. (27). The phonon plateaus are also very well visible in the dephasing rate, see Fig. 4. Counter-intuitively, when we increase the electron-phonon coupling the dephasing rate first decreases before it increases again (compare orange curves in the inset). This suppression occurs in the current blockade regime. Interestingly, the dephasing rate becomes much smaller than the equilibrium dephasing rate observed without phonons. Thus, we find that while increasing the coupling strength to the phonon reservoir, the model effectively shows a decrease of the dephasing rate which is in stark contrast to general expectations. We attribute this behaviour to the conditioned state of the phonon reservoir.

\[ \text{FIG. 4: Dephasing rate } \mathcal{R} \text{ in units of } \Gamma_L = \Gamma_R = \Gamma \text{ versus the external bias voltage } V \text{ in units of } \Omega. \]
C. Thermoelectric Generator

Multi-terminal nanostructures may serve as nanomachines converting e.g. temperature gradients into electric power or conversely – using electric power to pump heat from a cold reservoir to a hot one. Here, we focus on the first case: A hot phonon bath and cold electronic reservoirs may induce an electronic current at vanishing bias – or even a current against a finite bias generating useful power. We note that whereas for a single-electron transistor (with its always-symmetric electron-phonon coupling) one would require non-flat electronic tunneling rates to see such an effect, this is different in the present model when we apply it to the case of a continuous phonon spectrum. To quantify the performance of such a device, it is instructive to relate the power output $P = -I_M V$ to the heat entering from the hot phonon reservoir $Q = P^R_{ph}$. Positivity of the entropy production then grants that the efficiency of this process

$$\eta = \frac{P}{Q} = \frac{I_M V}{P^R_{ph}} \leq 1 - \frac{T_{el}}{T_{ph}} = \eta_{Ca}$$

is upper-bounded by Carnot efficiency. In general however, the efficiency can be significantly smaller as is illustrated in Fig. 5. In fact, the inset shows that Carnot efficiency is not even reached at the new equilibrium, where the electronic matter current vanishes: Formally, this is due to the fact that – in contrast to previous weak-coupling models\(^\text{36,48}\) – the total entropy production does not vanish at this point, as expected for models not exhibiting the tight-coupling condition\(^\text{21}\).

V. SUMMARY

We have investigated coherent electronic transport strongly coupled to vibrations. The failure of the naive generalization of the infinite bias results to finite bias could be overcome by performing the secular-approximation in a new basis. The method presented here yields a low dimensional master equation in Lindblad form which accounts for thermodynamic consistency. Correspondingly, we could confirm the fluctuation theorem for entropy production analytically. Using the Full Counting Statistics we computed the electronic current versus internal and external bias. For the former we found oscillations in the current due to electronic-phonon coupling. The latter showed strong signatures of the Franck-Condon blockade which also comes along with a partial suppression of the dephasing rate in this regime. Our method can be generalized to more complex systems and, thus, allows applications in a variety of transport setups such as molecules. Furthermore, the analysis of the entropy production in the polaron master equation allows one to study the performance of thermoelectric generators in the strong-coupling regime.

FIG. 5: (Color Online) Plot of the matter and energy currents for a hot phonon and cold electronic reservoirs versus electronic bias voltage. In the lower right quadrant, the electronic matter current (bold black) runs against a potential gradient thereby generating power $P = -I_M V$. The first law manifests in the fact that all energy currents add up to zero. Parameters have been chosen such that the internal phonon-assisted transitions between eigenstates $\left| - \right>$ and $\left| + \right>$ dominate the phonon heat flow (solid blue versus dashed curves for external jumps). Relating the power output with the heat input from the phonon reservoir $Q = \left( P^R_{ph} + P^L_{ph} + P^M_{ph} \right)$ we see that the efficiency of this process (inset, for positive bias voltage only) remains significantly below Carnot efficiency. Other parameters: $\Gamma_L = \Gamma_R = \Gamma$, $J_L T_c^2 = J_R T_c^2 = 0.001$, $J_e T_c^2 = 1.0$, $w^L_\epsilon = w^R_\epsilon = w_\epsilon = 1.0 T_c$, $\epsilon_L = +0.5 T_c$, $\epsilon_R = -\epsilon_R$, $U = 5.0 T_c$, $\beta_L T_c = \beta_R T_c = 10.0$, $\beta_{ph} T_c = 1.0$.

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### Appendix A: Polaron transformation

We consider the polaron transformation

\[ U = e^{d_\sigma^\dagger d_\sigma + d_\sigma^\dagger d_\sigma^\dagger} \]

with the fermionic annihilation operators \( d_\sigma \) and the bosonic operators

\[ B_\sigma = \sum_q \left( \frac{\hbar q_\sigma}{\omega_q} a_q^\dagger - \frac{\hbar q_\sigma}{\omega_q} a_q \right) \]

with bosonic annihilation operators \( a_q \). To calculate the transformation rules, we recall the BCH relation

\[ e^{X}Ye^{-X} = \sum_{n=0}^{\infty} \frac{1}{n!}[X,Y]_n \]

with the short-hand notation \([X,Y]_n = [X,[X,Y]]_n\) and \([X,Y]_0 = Y\). We first note that the exponential in the polaron transformation can be written in a separated fashion

\[ U = e^{d_\sigma^\dagger d_\sigma B_\sigma + d_\sigma^\dagger d_\sigma^\dagger B_\sigma} e^{-d_\sigma^\dagger d_\sigma d_\sigma^\dagger d_\sigma} [B_\sigma,B_\sigma] / 2 \]

\[ U_{LR} = e^{d_\sigma^\dagger d_\sigma^\dagger d_\sigma^\dagger d_\sigma^\dagger} i\Phi \equiv [B_\sigma,B_\sigma] / 2 \]

\[ U_{LR}^\dagger U_{LR} \equiv U_{LR} \]

where it is easy to show that \( \Phi^* = \Phi \). Consequently, the adjoint operator is given by

\[ U_{LR}^\dagger = U_{LR}^\dagger U_{LR} \]

and we note that \([U_{LR},U_L] = [U_{LR},U_R] = 0\). Alternatively, we can also split the unitary transformation according to

\[ U = U_R U_L U_{LR}^\dagger U_{LR} \]

where again \([U_{LR},U_L] = [U_{LR},U_R] = 0\) holds.

#### 1. Left Mode Operators

We consider the action of the Polaron transformation on the left fermionic annihilation operator

\[ U d_L U^\dagger = U_L U_R U_L d_L U_{LR}^\dagger U_L^\dagger U_L U_{LR}^\dagger U_L^\dagger = U_L d_L e^{-d_\sigma^\dagger d_\sigma^\dagger} \]

\[ = U_L d_L e^{-\Phi} \]

\[ = d_L e^{-\Phi} \]

\[ = d_L e^{-d_\sigma^\dagger d_\sigma^\dagger} \Phi \]

The left fermionic creation operator then transforms according to

\[ U d_L^\dagger U^\dagger = d_L^\dagger e^{B_\sigma^\dagger} e^{d_\sigma} \Phi \]
2. Right Mode Operators

In a similar fashion, we evaluate the transformation of the right fermionic annihilation operator

$$U_d R U^\dagger = U_R U'_d U_L R U_R U_L U^\dagger = U_R U'_d U_L R U_R U^\dagger$$

and similarly for the creation operator

$$U_d^\dagger R U^\dagger = d_d e^{-\frac{i\Phi}{2}} e^{-\frac{d_L^\dagger d_L}{4}}.$$

and the adjoint operator becomes

$$U_d^\dagger R U^\dagger = d_d^\dagger e^{\frac{d_L^\dagger d_L}{4}} e^{\frac{i\Phi}{2}}.$$

and similarly for the electronic inter-dot tunneling Hamiltonian

$$H'_T = T_c d_L e^{-\frac{d_L^\dagger d_L}{2} + \frac{i\Phi}{2}} e^{-\frac{d_R^\dagger d_R}{2} - \frac{i\Phi}{2}} + \frac{T_c e^{-2i\Phi}}{2} d_L^\dagger e^{-B_e e^{i\Phi}} + \frac{T_c e^{2i\Phi}}{2} d_R^\dagger e^{-B_n e^{i\Phi}},$$

for the electronic lead tunneling Hamiltonian

$$H'_V = \sum_k \left( t_{kL} d_L e^{-\frac{d_L^\dagger d_L}{2} + \frac{i\Phi}{2}} e^{-B_n c_{kL}} + h.c. \right) + \sum_k \left( t_{kR} d_R e^{-\frac{d_R^\dagger d_R}{2} + \frac{i\Phi}{2}} e^{-B_n c_{kR}} + h.c. \right).$$

and for the electron-phonon interaction

$$H'_{e-\text{ph}} = \sum_q \left( h_{q\sigma} a_q + h^*_q a_q^\dagger \right) d_d^\dagger d_d.$$

3. Bosonic Operators

For the bosonic annihilation operator we obtain

$$U a_q U^\dagger = U_L a_q U_R U^\dagger = U_L U_R a_q U_R U_L^\dagger = a_q - \frac{h_{q}^* L}{\omega} d_d^\dagger d_d - \frac{h_{q}^* R}{\omega} d_d^\dagger d_d R.$$

and similarly for the creation operator

$$U a_q^\dagger U^\dagger = a_q^\dagger - \frac{h_{q} L^*}{\omega} d_d^\dagger d_d - \frac{h_{q} R^*}{\omega} d_d^\dagger d_d R.$$

4. Polaron transformation of the DQD Hamiltonian

The total Hamiltonian of the DQD is given by

$$\mathcal{H} = \sum_{k\sigma} \epsilon_{k\sigma} c_k^\dagger c_k + \sum_q \omega_q a_q \left( a_q + a^\dagger_q \right)$$

and

$$+ \epsilon_L d_d^\dagger d_d + \epsilon_R d_d^\dagger d_d R + U d_d^\dagger d_d R d_d^\dagger d_d R$$

$$+ T_c \left( d_d^\dagger d_d R + d_d^\dagger R d_d \right) + \sum_{k\sigma} \left( t_{kL} d_L^\dagger c_{k\sigma} + h.c. \right)$$

$$+ \sum_{q\sigma} \left( h_{q\sigma} a_q + h^*_{q\sigma} a_q^\dagger \right) d_d^\dagger d_d,$$

Applying the polaron transformation to the total Hamiltonian $\mathcal{H} = U^\dagger \mathcal{H} U^\dagger$ implies that some parts of the Hamiltonian will change. In particular, we have for the free bosonic Hamiltonian

$$H'_{ph} = \sum_q \omega_q \left( a_q + a^\dagger_q \right) \left( a_q + a^\dagger_q \right)$$

$$\times \left( a_q + a^\dagger_q \right) \left( a_q + a^\dagger_q \right) + \sum_{q\sigma} \left( h_{q\sigma} a_q + h^*_{q\sigma} a_q^\dagger \right) d_d^\dagger d_d.$$

with renormalized on-site energies (9) and the Coulomb interaction (10). When furthermore one demands that all expectation values of reservoir coupling operators should vanish (see below), one arrives at the splitting into system, reservoir, and interaction parts used in the paper.
Appendix B: Shift factor

We use that for a thermal state $\rho \propto e^{-\beta \sum \omega_\nu a_\nu^\dagger a_\nu}$, one has for all complex-valued numbers $a_\nu$

$$
\langle e^{-\sum (\omega_\nu a_\nu^\dagger a_\nu)} \rangle = e^{-\sum |\omega_\nu|^2 [n_\nu (\omega_\nu) + 1]/2}
$$

(B1)

with the Bose-distribution $n_\nu (\omega_\nu) = [e^{\beta \omega_\nu} - 1]^{-1}$. Applying that to the shift factor $\kappa$, for generality in the interaction picture, we obtain

$$
\kappa = \left\langle e^{-B_\nu (\tau) e^{-B_\nu (\tau)}^\dagger} \right\rangle = \left\langle e^{-B_\nu (\tau) + B_\nu (\tau)} \right\rangle e^{i\Phi} - B_\nu e^{i\Phi}
$$

(B2)

and see that $\kappa$ is independent of $\tau$. Therefore, we can already in the Schrödinger picture write the Hamiltonian in a way that is suitable for the derivation of a master equation with splitting into system, reservoir and interaction parts given by Eqns. (8), (14), and (15) and (16) in the paper, respectively.

Appendix C: Inverse polaron transform

To apply the inverse polaron transformation, it is useful to write it conditioned on the electronic occupation

$$
U = 1 + d_u^d u^r (e^{B_\nu} - 1) + d_u^d u^r (e^{B_\nu} - 1)
$$

$$
+ d_u^d u^r d_u^r d_r^r (e^{B_\nu} + e^{B_\nu} - 1)
$$

$$
= P_0 1 + P_L e^{B_\nu} + P_R e^{B_\nu} + P_{LR} e^{B_\nu}
$$

(C1)

with where the projectors $P_0 = (1 - d_u^d u^r)(1 - d_u^d u^r d_r^r)$, $P_2 = d_u^d u^r d_u^r d_r^r$, $P_L = d_u^d u^r (1 - d_u^d u^r d_r^r)$, and $P_R = (1 - d_u^d u^r d_u^r d_r^r) d_r^r d_r^r$ it becomes visible that depending on the system state in the localized basis - different unitary operations are applied on the reservoir. For the phonon reservoir state this implies

$$
U^\dagger \rho_B^{ph} U = P_0 \otimes \rho_B^{ph} + P_2 \otimes (e^{B_\nu} + e^{B_\nu} - 1)
$$

$$
+ P_L \otimes e^{B_\nu} \rho_B^{ph} e^{B_\nu} + P_R \otimes e^{B_\nu} \rho_B^{ph} e^{B_\nu}
$$

(C2)

Considering that these unitary operations displace the phonons

$$
e^{-B_\nu a_\nu^\dagger a_\nu e^{B_\nu}} = \left( a_\nu^\dagger + \frac{h_{q \nu}}{\omega_\nu} a_\nu + \frac{h_{q \nu}}{\omega_\nu} \right) \times
$$

$$
e^{-B_\nu - B_\nu a_\nu^\dagger a_\nu + B_\nu + B_\nu} = \left( a_\nu^\dagger + \frac{h_{q \nu}}{\omega_\nu} a_\nu + \frac{h_{q \nu}}{\omega_\nu} \right) \times
$$

$$
\left( a_\nu + \frac{h_{q \nu}}{\omega_\nu} a_\nu + \frac{h_{q \nu}}{\omega_\nu} \right)
$$

(C3)

the reservoir state becomes the displaced thermal state - conditioned on the electronic occupation of the system.

Specifically, when in the localized basis the system density matrix is written as

$$
\rho_{S} = \rho_0 P_0 + \rho_2 P_2 + \rho_L P_L + \rho_R P_R + \rho_{LR} P_{LR} + \rho_{RL} P_{RL}
$$

(C4)

with $P_{LR} = |L\rangle \langle R|$ and $P_{RL} = |R\rangle \langle L|$, it transforms according to

$$
U^\dagger \rho_{S} U = \rho_0 P_0 + \rho_2 P_2 + \rho_L P_L + \rho_R P_R + \rho_{LR} P_{LR} e^{-B_\nu e^{B_\nu}} + \rho_{RL} P_{RL} e^{-B_\nu e^{B_\nu}}
$$

(C5)

This implies that the total system-phonon density matrix in the original frame is given by

$$
\tilde{\rho} = U^\dagger \rho_{S} \otimes 1 U U^\dagger 1 \otimes \rho_B^{ph}
$$

(C6)

Appendix D: Bath correlation functions

1. Phonon BCF

We compute the expectation value of the phononic contribution in the Lead-Phonon bath correlation functions, cf. Sec. II A 1, given by

$$
C_{\sigma}^{\sigma} = \left\langle e^{-B_\nu (\tau) + B_\nu} \right\rangle - \left\langle e^{-B_\nu (\tau) + B_\nu} \right\rangle e^{B_\nu (\tau) + B_\nu} / 2
$$

$$
= \left\langle e^{-B_\nu (\tau) + B_\nu} \right\rangle \left(1 - e^{-i\sigma \epsilon} \right) \left(1 - e^{-i\sigma \epsilon} \right)
$$

$$
\times \left( e^{-\sum |\omega_\nu|^2 \sinh (\omega_\nu \tau)} \right)
$$

$$
= e^{-\sum |\omega_\nu|^2 \sinh (\omega_\nu \tau)} [n_\nu (\omega_\nu) + 1] \times
$$

$$
\times e^{-\sum |\omega_\nu|^2 \sinh (\omega_\nu \tau)}
$$

$$
= e^{-\sum |\omega_\nu|^2 \sinh (\omega_\nu \tau)} [n_\nu (\omega_\nu) + 1] \times
$$

$$
\times e^{-\sum |\omega_\nu|^2 \sinh (\omega_\nu \tau)}
$$

(D1)
And noting that it is invariant under the transformation $h_{q,\sigma} \rightarrow -h_{q,\sigma}$ we conclude

$$\begin{align*}
C_{12}(\tau) &= c_{12}(\tau) \sum_k |t_{k,L}|^2 f_L(\epsilon_{k,L}) e^{i \epsilon_{k,L} \tau}, \\
C_{21}(\tau) &= c_{12}(\tau) \sum_k |t_{k,L}|^2 [1 - f_L(\epsilon_{k,L})] e^{-i \epsilon_{k,L} \tau}, \\
C_{34}(\tau) &= c_{34}(\tau) \sum_k |t_{k,R}|^2 f_R(\epsilon_{k,R}) e^{i \epsilon_{k,R} \tau}, \\
C_{43}(\tau) &= c_{34}(\tau) \sum_k |t_{k,R}|^2 [1 - f_R(\epsilon_{k,R})] e^{-i \epsilon_{k,R} \tau}.
\end{align*}$$

2. Inter-dot BCF

We show explicitly that $C_{55}(\tau)$ is given by Eq. (45):

$$C_{55}(\tau) = \left\langle e^{\mathcal{B}_n(\tau) - \mathcal{B}_n(\tau) + \mathcal{B}_n - \mathcal{B}_n} \right\rangle e^{2i\Phi} \times \times e^{\epsilon_{\mathcal{B}_n(\tau) - \mathcal{B}_n(\tau) + \mathcal{B}_n - \mathcal{B}_n} / 2 - \kappa^2 = \left( e^{\sum_{q} \frac{\lambda_q}{2q}[(1 + e^{-i\omega q}) \sigma_q - \frac{\lambda_q}{2q}(1 + e^{-i\omega q}) \sigma_q] \right) e^{2i\Phi} \times \times e^{i \sum_{q} \frac{|\lambda_q|^2}{2q} \sin(\omega_q \tau) - \kappa^2 = e^{2i\Phi} \times \sum_{q} \frac{|\lambda_q|^2}{2q} [(1 + n_B(\omega_q)) e^{-i\omega q} + n_B(\omega_q) e^{i\omega q}] \\
\times e^{-e^{-\sum_{q} \frac{|\lambda_q|^2}{2q} [(1 + n_B(\omega_q)) e^{-i\omega q} + n_B(\omega_q) e^{i\omega q}] - 1]} = \kappa^2 - \sum_{q} \frac{|\lambda_q|^2}{2q} [(1 + n_B(\omega_q)) e^{-i\omega q} + n_B(\omega_q) e^{i\omega q}] - 1].
\end{align*}$$

The bath correction function $C_{60}(\tau)$ can be obtained via $C_{60}(\tau) = C_{55}(\tau)$. We show explicitly that $C_{60}(\tau)$ is given by Eq. (48):

$$C_{60}(\tau) = \left\langle e^{\mathcal{B}_n(\tau) - \mathcal{B}_n(\tau) + \mathcal{B}_n - \mathcal{B}_n} \right\rangle \times \times e^{\epsilon_{\mathcal{B}_n(\tau) - \mathcal{B}_n(\tau) + \mathcal{B}_n - \mathcal{B}_n} / 2 - \kappa^2 = \left( e^{\sum_{q} \frac{\lambda_q}{2q}[(1 + e^{-i\omega q}) \sigma_q - \frac{\lambda_q}{2q}(1 + e^{-i\omega q}) \sigma_q] \right) \times \times e^{-e^{-\sum_{q} \frac{|\lambda_q|^2}{2q} \sin(\omega_q \tau) - \kappa^2 = \kappa^2 - \sum_{q} \frac{|\lambda_q|^2}{2q} [(1 + n_B(\omega_q)) e^{-i\omega q} + n_B(\omega_q) e^{i\omega q}] - 1].
\end{align*}$$

Appendix E: Symmetries in the Characteristic polynomials

To show these symmetries, we show separate symmetries of the terms in the characteristic polynomial:

(D3) First, we note that trivially, the combination $\mathcal{L}_{32}$ does not depend on counting fields and is thus by construction inert to symmetry transformations of type (62).
such that e.g. products of the form $L_{12}L_{21}$ are invariant under the transformations (62), i.e., $\bar{L}_{12}\bar{L}_{21} = \bar{L}_{12}\bar{L}_{21}$.

Third, we consider combinations of three off-diagonal matrix elements by noting the additional symmetry

$$
L_{23}^- = e^{+\beta R(\epsilon_+ - \epsilon_-)} \bar{L}_{32}, \\
L_{32}^- = e^{-\beta R(\epsilon_+ - \epsilon_-)} \bar{L}_{23},
$$

which together with the symmetries in Eq. (E3) can be used to show that in the characteristic polynomial (60) the terms with three off-diagonal matrix elements are also inert under the transformations (62), i.e.,

$$
\bar{L}_{23}\bar{L}_{34}\bar{L}_{42} + \bar{L}_{24}\bar{L}_{43}\bar{L}_{32} = \bar{L}_{23}\bar{L}_{34}\bar{L}_{42} + \bar{L}_{24}\bar{L}_{43}\bar{L}_{32}, \\
\bar{L}_{12}\bar{L}_{23}\bar{L}_{31} + \bar{L}_{13}\bar{L}_{32}\bar{L}_{21} = \bar{L}_{12}\bar{L}_{23}\bar{L}_{31} + \bar{L}_{13}\bar{L}_{32}\bar{L}_{21}.
$$

Finally, we note that the terms $\bar{L}_{12}\bar{L}_{21}\bar{L}_{34}\bar{L}_{43}$ and $\bar{L}_{13}\bar{L}_{31}\bar{L}_{24}\bar{L}_{42}$ can be treated similarly to the terms with just two off-diagonal matrix elements, and that the last two terms in the characteristic polynomial (60) obey

$$
\bar{L}_{12}\bar{L}_{24}\bar{L}_{34}\bar{L}_{43} + \bar{L}_{13}\bar{L}_{34}\bar{L}_{42}\bar{L}_{21} = \bar{L}_{12}\bar{L}_{24}\bar{L}_{34}\bar{L}_{43} + \bar{L}_{13}\bar{L}_{34}\bar{L}_{42}\bar{L}_{21}
$$

(E6) which can be shown with Eqns. (E3).