

Open quantum systems – lecture 14

lecture
winter semester 2022/2023
Technische Universität Dresden

Dr. Gernot Schaller

January 25, 2023

Chapter 5

Inexact methods beyond weak coupling

The exact methods discussed in the previous chapter will fail for systems with interactions (e.g. quartic Hamiltonians in annihilation and creation operators): The Heisenberg equations of motions **will not close** and although the general thermodynamic statements remain true, we cannot calculate the discussed quantities like entropy production exactly. Likewise, the weak-coupling (BMS) master equation may fail if we consider e.g. strong-system reservoir interactions or non-Markovian reservoirs. To close the arising descriptive gap, some methods have been developed.

5.1 General polaron master equation

We consider an arbitrary system described by H_S (which may contain interactions) via a single hermitian coupling operator $S = S^\dagger$ to a bosonic reservoir

$$H = H_S + \sum_k \omega_k \left[b_k^\dagger + \frac{h_k}{\omega_k} S \right] \left[b_k + \frac{h_k^*}{\omega_k} S \right], \quad (5.1)$$

System Hamiltonian *System-Reservoir Operator*

with reservoir energies ω_k , bosonic annihilation operators b_k , and spontaneous emission amplitudes h_k . Importantly, we note that – if H_S has a lower spectral bound – the above Hamiltonian has a lower spectral bound for all values of the coupling strength h_k . Upon expansion and introducing the spectral function $\Gamma(\omega)$, this leads to a term

$$\Delta H_S = \sum_k \frac{|h_k|^2}{\omega_k} S^2 = \frac{1}{2\pi} \int_0^\infty \frac{\Gamma(\omega)}{\omega} d\omega S^2 \quad (5.2)$$

that renormalizes the system Hamiltonian and is typically neglected in weak-coupling treatments. When for example following the minimal coupling procedure, such a term will however arise generically. In our discussion here, we will keep it.

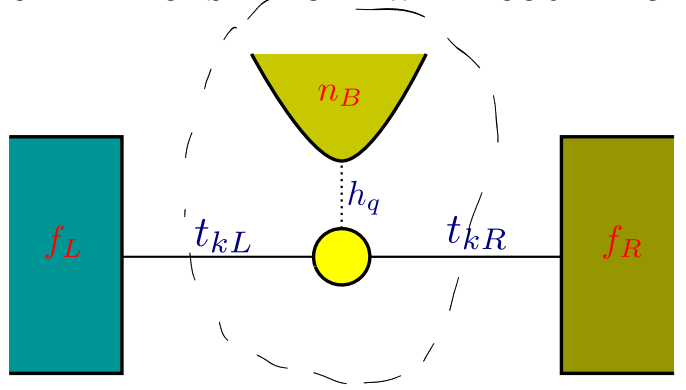
Now, the unitary **polaron transform**

$$U_P = \exp \left\{ S \sum_k \left(\frac{h_k^*}{\omega_k} b_k^\dagger - \frac{h_k}{\omega_k} b_k \right) \right\} \quad (5.3)$$

can be defined for arbitrary coupling operators S and has the following properties

$$\begin{aligned} U_P S U_P^\dagger &= S, \\ U_P b_k U_P^\dagger &= b_k - \frac{h_k^*}{\omega_k} S, \quad U_P b_k^\dagger U_P^\dagger = b_k^\dagger - \frac{h_k}{\omega_k} S, \end{aligned} \quad (5.4)$$

Figure 5.1: Sketch of a single-electron transistor that is capacitively coupled to a phonon reservoir. The interaction in the original Hamiltonian is of the pure dephasing type, i.e., to lowest order the SET energy will not be changed by the phonons. A conventional master equation treatment would yield no phonon effect on the SET dynamics.



where the first line follows trivially and the last two lines can be shown with the nested commutator expansion. In the polaron picture, the total Hamiltonian therefore becomes

$$H' = U_P H U_P^\dagger = U_P H_S U_P^\dagger + \sum_k \omega_k b_k^\dagger b_k. \quad (5.5)$$

e. Hall hopping

Now, if $[H_S, S] = 0$ as e.g. in the pure dephasing model, we would have $U_P H_S U_P^\dagger = H_S$ and the model was completely decoupled in the polaron frame. However, **realistic models will usually not do us this favor, and $U_P H_S U_P^\dagger$ will involve terms mediating the interaction between system and reservoirs.** The usual approach to derive a polaron master equation now defines a modified system Hamiltonian by the requirement that the first order expectation value of the interaction between system and reservoir should vanish in a thermal equilibrium state of the reservoir

$$H' = U_P H_S U_P^\dagger + \sum_k \omega_k b_k^\dagger b_k$$

$$= \underbrace{\text{Tr}_B \left\{ U_P H_S U_P^\dagger \frac{e^{-\sum_k \omega_k b_k^\dagger b_k}}{Z} \right\}}_{H'_S} + \underbrace{\left[U_P H_S U_P^\dagger - \text{Tr}_B \left\{ U_P H_S U_P^\dagger \frac{e^{-\sum_k \omega_k b_k^\dagger b_k}}{Z} \right\} \right]}_{H'_I} + \underbrace{\sum_k \omega_k b_k^\dagger b_k}_{H'_B}. \quad (5.6)$$

Handwritten blue wavy lines under H'_S and H'_B, and a green circle around H'_I.

The first term defines a renormalized system Hamiltonian, the second term denotes the system-reservoir interaction which by construction obeys $\text{Tr}_B \{ H'_I e^{-\beta H'_B} / Z \} = 0$, and the last term the reservoir contribution. In particular, we note that in this frame, in the strong coupling limit $\hbar \omega_k \rightarrow \infty$, none of these terms diverges. When deriving a master equation in this frame, instead of being perturbative in the $\hbar \omega_k$, we are rather perturbative in the term in square brackets, such that there exist regimes where the polaron master equation can be used to treat the strong-coupling limit.

5.2 Example: Phonon-coupled single electron transistor

As before, we consider a quantum dot model that is additionally coupled to phonons. To keep the analysis simple however, we follow Ref. [1] by considering an SET that is coupled to one, many, or even a continuum of phonon modes as depicted in Fig. 5.1.



5.2.1 Model

We write the full Hamiltonian as $H = H_{\text{SET}} + H_I + H_B^{\text{ph}}$, where the SET Hamiltonian is given by

$$H_{\text{SET}} = \underbrace{\epsilon d^\dagger d}_{\text{dot}} + \sum_{\nu \in \{L, R\}} \sum_k \underbrace{\left[\epsilon_{k\nu} c_{k\nu}^\dagger c_{k\nu} + t_{k\nu} d c_{k\nu}^\dagger + t_{k\nu}^* c_{k\nu} d^\dagger \right]}_{\substack{\text{Bader von SET} \\ \text{Tunnel}}} . \quad (5.7)$$

In addition, the central dot of the SET now interacts with a phonon reservoir

$$H_I = \underbrace{d^\dagger d}_{\text{S}} \otimes \sum_{q=1}^Q [h_q a_q + h_q^* a_q^\dagger] , \quad H_B^{\text{ph}} = \sum_{q=1}^Q \omega_q a_q^\dagger a_q \quad \leftarrow \text{Anzahl d. Phonon - modes} \quad (5.8)$$

$\Rightarrow [H_S, S] = 0$

containing Q phonon modes. Obviously, the interaction commutes with the central dot part of the SET Hamiltonian. Therefore, if one would conventionally derive a master equation for the population dynamics of the central quantum dot, the additional phonon bath would not affect the populations of the central dot at all – the interaction is of pure-dephasing type.

In general however, this cannot be true: The interaction does not commute with the total SET Hamiltonian, and therefore one must expect the phonons to have some effect. Indeed, extensive calculations with only a single phonon mode whose dynamics is completely taken into account have revealed a strong suppression of the electronic current when strongly-coupled phonons are present. This phenomenon has been termed Franck-Condon blockade [2].

To treat such cases within a master equation approach, we apply a transformation to the full Hamiltonian $H' = U H U^\dagger$ with the unitary operator

$$U = \exp \left\{ d^\dagger d \sum_q \left(\frac{h_q^*}{\omega_q} a_q^\dagger - \frac{h_q}{\omega_q} a_q \right) \right\} \equiv e^{d^\dagger d A} . \quad (5.9)$$

The above transformation is known as polaron or Lang-Firzov transformation [3, 4]. Obviously, the electronic leads are unaffected by the transformation, since $U c_{k\nu} U^\dagger = c_{k\nu}$, and also the central dot part is inert $U d^\dagger d U^\dagger = d^\dagger d$. There are multiple ways of proving the following relations

$$\begin{aligned} U d U^\dagger &= d e^{-A}, & U d^\dagger U^\dagger &= d^\dagger e^{+A}, \\ U a_q U^\dagger &= a_q - \frac{h_q^*}{\omega_q} d^\dagger d, & U a_q^\dagger U^\dagger &= a_q^\dagger - \frac{h_q}{\omega_q} d^\dagger d. \end{aligned} \quad (5.10)$$

These immediately also imply the relation

$$U a_q^\dagger a_q U^\dagger = a_q^\dagger a_q - \frac{d^\dagger d}{\omega_q} (h_q a_q + h_q^* a_q^\dagger) + \frac{|h_q|^2}{\omega_q^2} d^\dagger d . \quad (5.11)$$

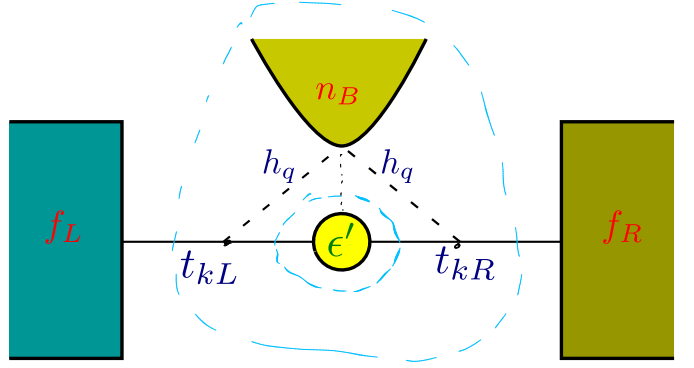
After the polaron transformation, the Hamiltonian therefore reads

$$\begin{aligned} H' &= \left(\epsilon - \sum_q \frac{|h_q|^2}{\omega_q} \right) d^\dagger d + \sum_{k\nu} \epsilon_{k\nu} c_{k\nu}^\dagger c_{k\nu} + \sum_q \omega_q a_q^\dagger a_q \\ &\quad + \sum_{k\nu} \left(t_{k\nu} d c_{k\nu}^\dagger e^{-A} + t_{k\nu}^* c_{k\nu} d^\dagger e^{+A} \right) , \end{aligned} \quad (5.12)$$

Resonanz
gedrehtes Tunnel

and thereby admits a new decomposition into system and bath Hamiltonians, see also Fig. 5.2. Most obvious, we observe a shift of the electronic level $\epsilon \rightarrow \epsilon' = \epsilon - \sum_q \frac{|h_q|^2}{\omega_q}$. Second, the electronic

Figure 5.2: After the polaron transformation, direct coupling between the central quantum dot and the phonons in Fig. 5.1 is transformed to the electronic tunnel couplings. The electron-phonon coupling may be treated non-perturbatively (dash-dotted lines) when the electronic tunnel couplings are treated perturbatively (dashed lines).



tunneling terms between central dot and the adjacent leads now become dressed by exponential operators

$$H'_I = \sum_{k\nu} \left[t_{k\nu} d c_{k\nu}^\dagger e^{-\sum_q \left(\frac{h_q^*}{\omega_q} a_q^\dagger - \frac{h_q}{\omega_q} a_q \right)} + t_{k\nu}^* c_{k\nu} d^\dagger e^{+\sum_q \left(\frac{h_q^*}{\omega_q} a_q^\dagger - \frac{h_q}{\omega_q} a_q \right)} \right], \quad (5.13)$$

=> <H'_I> = 0
nicht additiv in den Bädern

which demonstrates that every single electronic jump from the central dot to the leads may now trigger multiple phonon emissions or absorptions. This implies that a perturbative treatment in $t_{k\nu}$ still enables for a non-perturbative treatment of the phonon absorption and emission amplitudes h_q . Furthermore, this leads to the somewhat non-standard situation that already in the interaction Hamiltonian one has now operators from different reservoirs occurring in a product, which implies interesting properties for the correlation functions.

5.2.2 Reservoir equilibrium in the polaron picture

Before we proceed further by deriving a master equation in the displaced polaron frame, we remark that the solution from the displaced frame has to be transformed back to the original picture. A rate equation in the displaced frame implies that the full density matrix in the polaron frame is given by a product state of system and reservoir, where the phonon reservoir density matrix is given by the thermal equilibrium state $\rho'(t) = \rho'_S(t) \bar{\rho}_B^{(L)} \bar{\rho}_B^{(R)} \frac{e^{-\beta_{\text{ph}} H'_B}}{Z'_{\text{ph}}}$. The transformation back to the initial frame is given by the inverse polaron transformation

$$\begin{aligned} \rho(t) &= U^\dagger \rho'(t) U = U^\dagger \rho'_S(t) \bar{\rho}_B^{(L)} \bar{\rho}_B^{(R)} U U^\dagger \frac{e^{-\beta_{\text{ph}} H'_B}}{Z'_{\text{ph}}} U \\ &= U^\dagger \rho'_S(t) U \bar{\rho}_B^{(L)} \bar{\rho}_B^{(R)} \frac{e^{-\beta_{\text{ph}} U^\dagger H'_B U}}{Z'_{\text{ph}}}, \end{aligned} \quad (5.14)$$

where we have used that the polaron transformation (5.9) leaves the electronic reservoirs untouched. When the system density matrix does not exhibit coherences $\rho'_S(t) = P_E(t) d d^\dagger + P_F(t) d^\dagger d$, the unitary transformation will leave it untouched, such that only the reservoir part will be modified. With $H'_B = \sum_q \omega_q a_q^\dagger a_q$ we can with the inverse transformations of Eq. (5.10)

$$\begin{aligned} U^\dagger H'_B U &= \sum_q \omega_q a_q^\dagger a_q + d^\dagger d \otimes \sum_q (h_q a_q + h_q^* a_q^\dagger) + \sum_q \frac{|h_q|^2}{\omega_q} d^\dagger d \\ &= d^\dagger d \otimes \sum_q \left(\omega_q a_q^\dagger a_q + h_q a_q + h_q^* a_q^\dagger + \frac{|h_q|^2}{\omega_q} \mathbf{1} \right) + d d^\dagger \otimes \sum_q \omega_q a_q^\dagger a_q \end{aligned} \quad (5.15)$$

represent the operator in the exponential as a sum of commuting operators. Since for all operators $AB = BA = \mathbf{0}$ we have $e^{A+B} = e^A e^B$ we conclude

$$\begin{aligned} e^{-\beta_{\text{ph}} U^\dagger H'_B U} &= e^{-\beta_{\text{ph}} d^\dagger d \otimes \sum_q \omega_q (a_q^\dagger + h_q/\omega_q)(a_q + h_q^*/\omega_q)} e^{-\beta_{\text{ph}} d d^\dagger \otimes \sum_q \omega_q a_q^\dagger a_q} \\ &= \left[\mathbf{1} + d^\dagger d \left(e^{-\beta_{\text{ph}} \sum_q \omega_q (a_q^\dagger + h_q/\omega_q)(a_q + h_q^*/\omega_q)} - \mathbf{1} \right) \right] \left[\mathbf{1} + d d^\dagger \left(e^{-\beta_{\text{ph}} \sum_q \omega_q a_q^\dagger a_q} - \mathbf{1} \right) \right] \\ &= d^\dagger d e^{-\beta_{\text{ph}} \sum_q \omega_q (a_q^\dagger + h_q/\omega_q)(a_q + h_q^*/\omega_q)} + d d^\dagger e^{-\beta_{\text{ph}} \sum_q \omega_q a_q^\dagger a_q}. \end{aligned} \quad (5.16)$$

Comparing with the initial Hamiltonian, the phonon part of the first term in the last line is nothing but the thermal phonon state under the side constraint that the SET dot is filled. Formally, this can be seen by replacing $d^\dagger d \rightarrow 1$ in Eq. (5.8). Similarly, the other term is the thermalized phonon state when the SET dot is empty. Therefore, preparing the reservoir in a thermal state in the polaron-transformed frame implies that in the original frame, the reservoir state is conditioned on the state of the system. Inserting the assumption that there are no coherences in the system $\rho'_S(t) = P_E(t) d d^\dagger + P_F(t) d^\dagger d$, the full density matrix in the original frame becomes

$$\rho(t) = P_E(t) d d^\dagger \bar{\rho}_B^{(L)} \bar{\rho}_B^{(R)} \otimes \frac{e^{-\beta_{\text{ph}} \sum_q \omega_q a_q^\dagger a_q}}{Z'_{\text{ph}}} + P_F(t) d^\dagger d \bar{\rho}_B^{(L)} \bar{\rho}_B^{(R)} \otimes \frac{e^{-\beta_{\text{ph}} \sum_q \omega_q (a_q^\dagger + h_q/\omega_q)(a_q + h_q^*/\omega_q)}}{Z'_{\text{ph}}}. \quad (5.17)$$

Therefore, when the SET dot is occupied, the phonon state is given by a displaced thermal state, whereas when the SET dot is empty, it is just given by the thermal state corresponding to the original phonon Hamiltonian. The phonon dynamics thereby follows the system state immediately, which goes beyond the conventional Born approximation.

5.2.3 Polaron Rate Equation for discrete phonon modes

In the transformed frame, we do now proceed to derive a rate equation for the SET dot populations. We choose to count the phonons emitted into the phonon bath, to test the applicability of the counting field formalism. Here, we will use $N_{\text{ph}} = \sum_q a_q^\dagger a_q$ as the reservoir observable of interest. Identifying the bath coupling operators in the interaction Hamiltonian (5.13) as

$$B_{1\nu} = \sum_k t_{k\nu} c_{k\nu}^\dagger e^{-A}, \quad B_{2\nu} = \sum_k t_{k\nu}^* c_{k\nu} e^{+A} \quad (5.18)$$

electr. phonon

it becomes quite obvious that the reservoir correlation functions will now simultaneously contain contributions from electronic and phonon reservoirs. Recalling the definition of the generalized correlation function, we obtain a simple product form between electronic and phononic contributions

$$\begin{aligned} C_{12}^{\nu, \chi}(\tau) &= \langle \mathbf{B}_{1\nu}(\tau) B_{2\nu} \rangle = C_{12, \text{el}}^\nu(\tau) C_{12, \text{ph}}^\chi(\tau), \\ C_{21}^{\nu, \chi}(\tau) &= \langle \mathbf{B}_{2\nu}(\tau) B_{1\nu} \rangle = C_{21, \text{el}}^\nu(\tau) C_{21, \text{ph}}^\chi(\tau). \end{aligned} \quad (5.19)$$

factorized

Here, the electronic contributions are just the conventional ones known from the SET

$$\begin{aligned} C_{12, \text{el}}^\nu(\tau) &= \sum_k |t_{k\nu}|^2 f_\nu(\epsilon_{k\nu}) e^{+i\epsilon_{k\nu}\tau} = \frac{1}{2\pi} \int \Gamma_\nu(-\omega) f_\nu(-\omega) e^{-i\omega\tau} d\omega, \\ C_{21, \text{el}}^\nu(\tau) &= \sum_k |t_{k\nu}|^2 [1 - f_\nu(\epsilon_{k\nu})] e^{-i\epsilon_{k\nu}\tau} = \frac{1}{2\pi} \int \Gamma_\nu(\omega) [1 - f_\nu(\omega)] e^{-i\omega\tau} d\omega. \end{aligned} \quad (5.20)$$

In contrast, the phonon contributions are given by

$$C_{12,\text{ph}}^\chi(\tau) = \left\langle e^{-iN_{\text{ph}}\chi} e^{-\mathbf{A}(\tau)} e^{+iN_{\text{ph}}\chi} e^{+\mathbf{A}} \right\rangle, \quad C_{21,\text{ph}}^\chi(\tau) = \left\langle e^{-iN_{\text{ph}}\chi} e^{+\mathbf{A}(\tau)} e^{+iN_{\text{ph}}\chi} e^{-\mathbf{A}} \right\rangle, \quad (5.21)$$

with the phonon operator in the interaction picture

$$\mathbf{A}(\tau) = \sum_q \left(\frac{h_q^*}{\omega_q} a_q^\dagger e^{+i\omega_q\tau} - \frac{h_q}{\omega_q} a_q e^{-i\omega_q\tau} \right). \quad (5.22)$$

We note that by $h_q \rightarrow -h_q$, we transform $C_{12,\text{ph}}^\chi(\tau) \rightarrow C_{21,\text{ph}}^\chi(\tau)$, such that we actually only need to calculate one correlation function. To calculate phonon contribution to the correlation function, we can exploit that (with $\mathbf{A}^\chi(\tau) = e^{-iN_{\text{ph}}\chi} \mathbf{A}(\tau) e^{+iN_{\text{ph}}\chi}$)

$$[\mathbf{A}^\chi(\tau), A] = 2i \sum_q \frac{|h_q|^2}{\omega_q^2} \sin(\omega_q\tau - \chi) \quad (5.23)$$

is just a number, which implies – using the Baker-Campbell-Hausdorff relation

$$\begin{aligned} e^{-\mathbf{A}^\chi(\tau)} e^{+A} &= e^{A - \mathbf{A}^\chi(\tau) - 1/2[\mathbf{A}^\chi(\tau), A]} \\ &= e^{\sum_q \left(\frac{h_q^*}{\omega_q} a_q^\dagger (1 - e^{+i(\omega_q\tau - \chi)}) - \frac{h_q}{\omega_q} a_q (1 - e^{-i(\omega_q\tau - \chi)}) \right)} e^{-i \sum_q \frac{|h_q|^2}{\omega_q^2} \sin(\omega_q\tau - \chi)}. \end{aligned} \quad (5.24)$$

For a thermal reservoir, the phonon correlation function can be written as a product of single-mode

correlation functions $C_{12,\text{ph}}^\chi(\tau) = \prod_{q=1}^Q C_{12,\text{ph}}^{\chi,q}(\tau)$, where the single mode contributions read

$$\begin{aligned} C_{\text{ph}}^{\chi,q}(\tau) &= \left\langle e^{\frac{h_q^*}{\omega_q} a_q^\dagger (1 - e^{+i(\omega_q\tau - \chi)}) - \frac{h_q}{\omega_q} a_q (1 - e^{-i(\omega_q\tau - \chi)})} e^{-i \frac{|h_q|^2}{\omega_q^2} \sin(\omega_q\tau - \chi)} \right\rangle \\ &= \left\langle e^{\frac{h_q^*}{\omega_q} a_q^\dagger (1 - e^{+i(\omega_q\tau - \chi)})} e^{-\frac{h_q}{\omega_q} a_q (1 - e^{-i(\omega_q\tau - \chi)})} \right\rangle e^{-\frac{|h_q|^2}{\omega_q^2} (1 - e^{-i(\omega_q\tau - \chi)})}. \end{aligned} \quad (5.25)$$

By expanding the exponentials, we can evaluate the expectation value for thermal states, where the probability of having n quanta in the mode q is given by $P_n = (1 - e^{-\beta_{\text{ph}}\omega_q}) e^{-n\beta_{\text{ph}}\omega_q}$ as

$$\begin{aligned} \left\langle e^{\alpha_q^* a_q^\dagger} e^{-\alpha_q a_q} \right\rangle &= \sum_{n,m=0}^{\infty} \frac{(\alpha_q^*)^n}{n!} \frac{(-\alpha_q)^m}{m!} \sum_{\ell=0}^{\infty} P_\ell \langle \ell | (a_q^\dagger)^n (a_q)^m | \ell \rangle \\ &= \sum_{n=0}^{\infty} \frac{(-1)^n |\alpha_q|^{2n}}{(n!)^2} \sum_{\ell=0}^{\infty} P_\ell \langle \ell | (a_q^\dagger)^n (a_q)^n | \ell \rangle = \sum_{\ell=0}^{\infty} P_\ell \sum_{n=0}^{\ell} \frac{(-1)^n |\alpha_q|^{2n}}{(n!)^2} \frac{\ell!}{(\ell - n)!} \\ &= \sum_{\ell=0}^{\infty} P_\ell \mathcal{L}_\ell(|\alpha_q|^2) = e^{-|\alpha_q|^2 n_B^q} \end{aligned} \quad (5.26)$$

with the Bose distribution $n_B^q = [e^{\beta_{\text{ph}}\omega_q} - 1]^{-1}$ and Legendre polynomials, defined by the Rodriguez formula [5]

$$\mathcal{L}_n(x) = \frac{1}{2^n n!} \frac{d^n}{dx^n} [x^2 - 1]^n. \quad (5.27)$$

The single-mode contributions thus become with $\alpha_q = \frac{h_q}{\omega_q}(1 - e^{-i(\omega_q\tau - \chi)})$

$$C_{\text{ph}}^{\chi,q}(\tau) = \exp \left\{ \frac{|h_q|^2}{\omega_q^2} \left[e^{-i(\omega_q\tau - \chi)} (1 + n_B^q) + e^{+i(\omega_q\tau - \chi)} n_B^q - (1 + 2n_B^q) \right] \right\}, \quad (5.28)$$

such that finally, we obtain for the phonon correlation function

$$C_{12,\text{ph}}^{\chi}(\tau) = \exp \left\{ \sum_q \frac{|h_q|^2}{\omega_q^2} \left[e^{-i(\omega_q\tau - \chi)} (1 + n_B^q) + e^{+i(\omega_q\tau - \chi)} n_B^q - (1 + 2n_B^q) \right] \right\}. \quad (5.29)$$

The fact that the transformation $h_q \rightarrow -h_q$ leaves this result invariant implies that the phonon contribution is always the same in Eq. (5.19), such that we can drop the indices 12 and 21. Furthermore, we see that the phonon counting field occurs at the positions where one might have intuitively expected them. We note that the phonon correlation function obeys the KMS condition.

The observation that in the phonon correlation function (5.28) the terms proportional to $(1 + n_B^q)$ correspond to the emission of a phonon into the phonon reservoir and terms proportional to n_B^q alone are responsible for the absorption of a phonon from the reservoir enables one to derive the full phonon counting statistics from the model. Formally expanding the single mode correlation function into multiple emission (m') and absorption (m) events we would obtain a decomposition in the net number of phonon absorptions by the phonon bath $n = m' - m$, where $C_{\text{ph}}^{\chi,q}(\tau) = \sum_{n=-\infty}^{+\infty} C_{\text{ph}}^{q,n}(\tau) e^{in\chi}$, and $C_{\text{ph}}^{q,n}(\tau) = \frac{1}{2\pi} \int_{-\pi}^{+\pi} C_{\text{ph}}^{\chi,q}(\tau) e^{-in\chi} d\chi$ can be determined by the inverse Fourier transform. In particular, using that

$$C_{\text{ph}}^q(\tau) = e^{-\frac{|h_q|^2}{\omega_q^2}(1+2n_B^q)} \sum_{m,m'=0}^{\infty} \left(\frac{|h_q|^2}{\omega_q^2} \right)^{m+m'} \frac{(n_B^q)^m (1 + n_B^q)^{m'}}{m!m'!} e^{+i(m-m')\omega_q\tau} \quad (5.30)$$

one can show that by introducing the net number of phonon absorptions by the phonon bath $n = m' - m$, the correlation function can be represented as (below, we drop the counting field $\chi \rightarrow 0$, since we have an interpretation for each term)

$$C_{\text{ph}}^q(\tau) = \sum_{n=-\infty}^{+\infty} e^{-in\omega_q\tau} e^{-\frac{|h_q|^2}{\omega_q^2}(1+2n_B^q)} \left(\frac{1 + n_B^q}{n_B^q} \right)^{\frac{n}{2}} \mathcal{J}_n \left(2 \frac{|h_q|^2}{\omega_q^2} \sqrt{n_B^q(1 + n_B^q)} \right), \quad (5.31)$$

where $\mathcal{J}_n(x)$ denotes the modified Bessel function of the first kind [5] – defined by the solution of the differential equation $z^2 \mathcal{J}_n''(z) + z \mathcal{J}_n'(z) - (z^2 + n^2) \mathcal{J}_n(z) = 0$. Introducing for multiple modes the notation $\mathbf{n} = (n_1, \dots, n_Q)$, $\boldsymbol{\omega} = (\omega_1, \dots, \omega_Q)$, we therefore have for the full multi-mode phonon correlation function the representation

$$\begin{aligned} C_{\text{ph}}(\tau) &= \sum_{\mathbf{n}} e^{-i\mathbf{n} \cdot \boldsymbol{\omega} \tau} \prod_{q=1}^Q \left[e^{-\frac{|h_q|^2}{\omega_q^2}(1+2n_B^q)} \left(\frac{1 + n_B^q}{n_B^q} \right)^{\frac{n_q}{2}} \mathcal{J}_{n_q} \left(2 \frac{|h_q|^2}{\omega_q^2} \sqrt{n_B^q(1 + n_B^q)} \right) \right] \\ &= \sum_{\mathbf{n}} e^{-i\mathbf{n} \cdot \boldsymbol{\omega} \tau} C_{\text{ph}}^{\mathbf{n}}, \end{aligned} \quad (5.32)$$

where the simple exponential prefactor enables to calculate the Fourier transform of the full correlation function. In particular if only a single phonon mode is present, this enables a simple

calculation of the Fourier transform of the complete electron-phonon correlation function

$$\begin{aligned}\gamma_{12}^{\nu}(\omega) &= \sum_{\mathbf{n}_{\nu}} \gamma_{12,\text{el}}^{\nu}(\omega - \mathbf{n}_{\nu} \cdot \boldsymbol{\omega}) C_{\text{ph}}^{\mathbf{n}_{\nu}} = \sum_{\mathbf{n}_{\nu}} \gamma_{12,\mathbf{n}_{\nu}}^{\nu}(\omega), \\ \gamma_{21}^{\nu}(\omega) &= \sum_{\mathbf{n}_{\nu}} \gamma_{21,\text{el}}^{\nu}(\omega - \mathbf{n}_{\nu} \cdot \boldsymbol{\omega}) C_{\text{ph}}^{\mathbf{n}_{\nu}} = \sum_{\mathbf{n}_{\nu}} \gamma_{21,\mathbf{n}_{\nu}}^{\nu}(\omega).\end{aligned}\quad (5.33)$$

Here, the terms $\gamma_{12,\mathbf{n}_{\nu}}^{\nu}$ are interpreted as the emission of $\mathbf{n}_{\nu}$ phonons into the phonon reservoir whilst an electron jumps from lead ν onto the SET dot, whereas $\gamma_{21,\mathbf{n}_{\nu}}^{\nu}$ accounts for the emission of $\mathbf{n}_{\nu}$ when an electron is emitted to lead ν . Now, the bosonic KMS relation

$$C_{\text{ph}}^{-\mathbf{n}_{\nu}} = e^{-\beta_{\text{ph}} \mathbf{n}_{\nu} \cdot \boldsymbol{\omega}} C_{\text{ph}}^{+\mathbf{n}_{\nu}} \quad (5.34)$$

together with properties of the Fermi functions implies a KMS-type relation for the full correlation function

$$\gamma_{12,+\mathbf{n}_{\nu}}^{\nu}(-\omega) = e^{-\beta_{\nu}(\omega - \mu_{\nu} + \mathbf{n}_{\nu} \cdot \boldsymbol{\omega})} e^{+\beta_{\text{ph}} \mathbf{n}_{\nu} \cdot \boldsymbol{\omega}} \gamma_{21,-\mathbf{n}_{\nu}}^{\nu}(+\omega), \quad (5.35)$$

which now involves both the electronic and phononic temperatures. However, we note that when these temperatures are equal, the usual local detailed balance relations are reproduced. Deriving a secular-type rate equation for the dot occupation is now straightforward, the probabilities for finding the dot empty or filled are governed by the rate matrix

$$\mathcal{L} = \sum_{\nu \in \{L,R\}} \sum_{\mathbf{n}_{\nu}} \begin{pmatrix} -\gamma_{12,\mathbf{n}_{\nu}}^{\nu}(-\epsilon') & +\gamma_{21,-\mathbf{n}_{\nu}}^{\nu}(+\epsilon') \\ +\gamma_{12,\mathbf{n}_{\nu}}^{\nu}(-\epsilon') & -\gamma_{21,-\mathbf{n}_{\nu}}^{\nu}(+\epsilon') \end{pmatrix}, \quad (5.36)$$

where $\gamma_{12,\mathbf{n}_{\nu}}^{\nu}(-\epsilon')$ denotes the rate for an electron jumping onto the SET dot from lead ν whilst simultaneously emitting $\mathbf{n}_{\nu}$ phonons of the various modes into the phonon reservoir. Correspondingly, $\gamma_{21,-\mathbf{n}_{\nu}}^{\nu}(+\epsilon')$ denotes the rate for the inverse process. Having identified the rates for the various involved processes, we can proceed by introducing counting fields. For a three-terminal system with the phononic junction only allowing for energy exchange and with conservation laws on the total energy and particle number we can expect three counting fields to be sufficient for tracking the full entropy production. These can – for example – be the matter transfer from left to right and the energy emitted to the phonon bath counted separately for electronic jumps, such that we have the counting-field dependent version

$$\begin{aligned}\mathcal{L}(\chi, \xi_L, \xi_R) &= \begin{pmatrix} -\gamma_{12,\mathbf{n}_L}^L(-\epsilon') & +\gamma_{21,-\mathbf{n}_L}^L(+\epsilon') e^{-i\mathbf{n}_L \cdot \Omega \xi_L} \\ +\gamma_{12,\mathbf{n}_L}^L(-\epsilon') e^{+i\mathbf{n}_L \cdot \Omega \xi_L} & -\gamma_{21,-\mathbf{n}_L}^L(+\epsilon') \end{pmatrix} \\ &+ \begin{pmatrix} -\gamma_{12,\mathbf{n}_R}^R(-\epsilon') & +\gamma_{21,-\mathbf{n}_R}^R(+\epsilon') e^{+i\chi} e^{-i\mathbf{n}_R \cdot \Omega \xi_R} \\ +\gamma_{12,\mathbf{n}_R}^R(-\epsilon') e^{-i\chi} e^{+i\mathbf{n}_R \cdot \Omega \xi_R} & -\gamma_{21,-\mathbf{n}_R}^R(+\epsilon') \end{pmatrix},\end{aligned}\quad (5.37)$$

which enables one to reconstruct all energy and matter currents and thus the full entropy flow.

Here, we will first investigate the impact of the phonon presence on the electronic matter current. If one is only interested in the electronic current, we may set $\xi_L = \xi_R = 0$. The transition rates in the above Liouvillian become particularly simple in the case of a single phonon mode

$$\begin{aligned}\gamma_{12,+\mathbf{n}}^{\nu}(-\epsilon') &= \Gamma_{\nu}(\epsilon' + n\Omega) f_{\nu}(\epsilon' + n\Omega) e^{-\Lambda(1+2n_B)} \left(\frac{1+n_B}{n_B} \right)^{\frac{n}{2}} \mathcal{J}_n \left(2\Lambda \sqrt{n_B(1+n_B)} \right), \\ \gamma_{21,-\mathbf{n}}^{\nu}(+\epsilon') &= \Gamma_{\nu}(\epsilon' + n\Omega) [1 - f_{\nu}(\epsilon' + n\Omega)] e^{-\Lambda(1+2n_B)} \left(\frac{n_B}{1+n_B} \right)^{\frac{n}{2}} \mathcal{J}_n \left(2\Lambda \sqrt{n_B(1+n_B)} \right),\end{aligned}\quad (5.38)$$

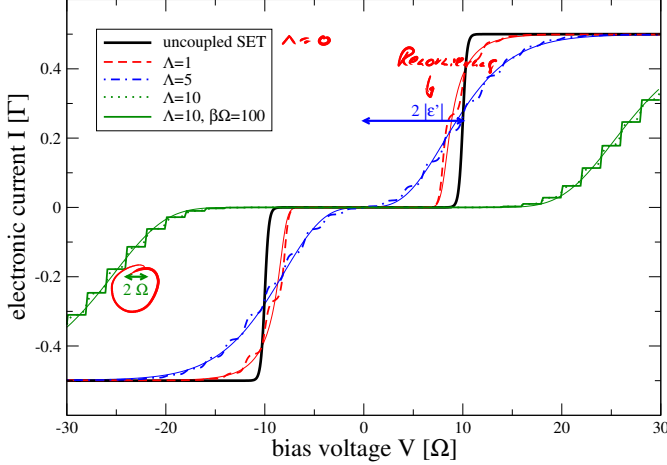


Figure 5.3: Electronic matter current versus bias voltage applied to the SET for vanishing (bold black) and increasing (dashed red, dash-dotted blue, and dotted green, respectively) coupling strengths $\Lambda = |h|^2/\Omega^2 = J_0$ to a single phonon mode of frequency Ω (bold curves) or to a continuum of phonon modes distributed according to an ohmic model (thin solid curves in background). The Franck-Condon blockade can within this model be understood in terms of a renormalization of the effective dot level $\epsilon' = \epsilon - \Lambda\Omega$, which – when $\Lambda\Omega \gg \epsilon$ will lead to current suppression. Furthermore, the steps in the electronic current observed for sufficiently low temperatures (solid green) admit for the transport spectroscopy of the phonon frequency Ω . In the multi-mode case (thin solid curves, for $\omega_c = \Omega$ and $J_0 = \Lambda$), current suppression due to the level renormalization is also observed but the steps in the current are no longer visible. Other parameters: $\Gamma_L = \Gamma_R = \Gamma$, $\beta_L = \beta_R = \beta_{ph} = \beta$, $\beta\Omega = 10$ (except the thin green curve), $\epsilon = 5\Omega$, $J_0 = \Lambda$, $\omega_c = \Omega$.

where $\Lambda = \frac{|h|^2}{\Omega^2}$ denotes the dimensionless coupling strength to the single phonon mode which is occupied according to $n_B = [e^{\beta_{ph}\Omega} - 1]^{-1}$. The resulting electronic matter current is depicted in Fig. 5.3.

Surprisingly, the simple 2×2 rate matrix predicts many signatures in the electronic current. For example, in the electronic matter current one can read off the renormalized dot level at sufficiently low electronic temperatures. In addition however, low temperatures also allow to determine the phonon frequency from the width of the multiple plateaus.

5.2.4 Thermodynamic interpretation

The present rate equation does not directly fit the standard thermodynamic discussion, since the contribution of the three reservoirs to the rates is not additive. Such non-additivity may lead to interesting properties: For example, it is possible to implement a QAR with three reservoirs and just a two-level system when the rates are non-additive [6]. Nevertheless, an interpretation in terms of stochastic thermodynamics is possible.

The strong modification of the electronic current is due to the fact that the phonons allow for processes that would normally be forbidden, see Fig. 5.4 In the trajectory in the figure, first an electron jumps in from the left lead to the initially empty SET whilst absorbing two phonons. The change of the system energy by $\Delta E = +\epsilon' = \Delta E_L + \Delta E_{ph}$ is supplied by both the left lead $\Delta E_L = \epsilon' - 2\Omega$ and the phonon bath $\Delta E_{ph} = +2\Omega$. In the second step, the electron leaves the dot towards the right lead whilst again absorbing three phonons. Again, the change of the system energy by $-\epsilon'$ is supplied by the right lead $\Delta E_R = -(\epsilon' + 3\Omega)$ and the phonon bath $\Delta E_{ph} = +3\Omega$. These energy and matter transfers can be used to construct the total heat exchanged between the

by

$$\begin{aligned} I_E^\nu &= \sum_n I_E^{(\nu,n)} = \sum_n [\gamma_{12,+n}(-\epsilon') \bar{P}_E - \gamma_{21,-n}(+\epsilon') \bar{P}_F] (\epsilon' + n\Omega), \\ I_E^{\text{ph}} &= \sum_n \left[I_E^{(n,L,\text{ph})} + I_E^{(n,R,\text{ph})} \right] = \sum_n \sum_\nu [\gamma_{21,-n}(+\epsilon') \bar{P}_F - \gamma_{12,+n}(-\epsilon') \bar{P}_R] n\Omega, \end{aligned} \quad (5.41)$$

whereas the total electronic matter current from lead ν is given by

$$I_M^\nu = \sum_n I_E^{(\nu,n)} = \sum_n [\gamma_{12,+n}(-\epsilon') \bar{P}_E - \gamma_{21,-n}(+\epsilon') \bar{P}_F]. \quad (5.42)$$

Similarly, the total entropy flow from the electronic leads is obtained by summing over all different n , and the total entropy flow from the phonon reservoirs is obtained by summing over the contributions from different n and different ν

$$\begin{aligned} \dot{S}_e^{(\nu)} &= \sum_n \dot{S}_{e,\text{el}}^{(\nu,n)} \\ \dot{S}_e^{\text{ph}} &= \sum_n \left(\dot{S}_{e,\text{ph}}^{(L,n)} + \dot{S}_{e,\text{ph}}^{(R,n)} \right). \end{aligned} \quad (5.43)$$

Altogether, the system obeys the laws of thermodynamics, which results in an overall positive entropy production. Consequently, we just note here that it is possible to verify a fluctuation theorem for entropy production, i.e., for $P_{n,e_{\text{ph}}^L,e_{\text{ph}}^R}(t)$ denoting the probability for trajectories with n electrons having traversed the SET from left to right and having emitted energy $e_{\text{ph}}^L = \mathbf{n}_L \cdot \boldsymbol{\omega}$ to the phonon reservoir during electronic jumps over the left and energy $e_{\text{ph}}^R = \mathbf{n}_R \cdot \boldsymbol{\omega}$ during jumps over the right barrier. In detail, it reads [1]

$$\lim_{t \rightarrow \infty} \frac{P_{+n,e_{\text{ph}}^L,e_{\text{ph}}^R}(t)}{P_{-n,-e_{\text{ph}}^L,-e_{\text{ph}}^R}(t)} = e^{[(\beta_R - \beta_L)\epsilon' + (\beta_L \mu_L - \beta_R \mu_R)]n + (\beta_{\text{ph}} - \beta_L)e_{\text{ph}}^L + (\beta_{\text{ph}} - \beta_R)e_{\text{ph}}^R}, \quad (5.44)$$

and it is straightforward to see that it reduces to the conventional fluctuation theorem when all temperatures are equal.

Disregarding the phonon counting statistics, we note that the system also obeys a fluctuation theorem involving the electronic transfer statistics only

$$\lim_{t \rightarrow \infty} \frac{P_{+n}(t)}{P_{-n}(t)} = e^{n\mathcal{A}_{\text{eff}}}, \quad (5.45)$$

where the effective affinity $\mathcal{A}_{\text{eff}}$ is however not related to the entropy production, it does, for example, depend on the details of the coupling.

5.2.5 Polaron Rate Equation for continuum phonon modes

Also for a continuum of phonon modes it is possible to obtain a master equation representation. Here, we directly represent the phonon correlation function (5.29), taking a counting field for the energy of the phonon reservoir into account. This then yields

$$C_{\text{ph}}^\xi(\tau) = \exp \left\{ \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} \left[e^{-i\omega(\tau-\xi)} (1 + n_B(\omega)) + e^{+i\omega(\tau-\xi)} n_B(\omega) - (1 + 2n_B(\omega)) \right] \right\}, \quad (5.46)$$

where we have introduced the spectral density $J(\omega) = \sum_q |h_q|^2 \delta(\omega - \omega_q)$, and ξ is a counting field responsible for the energy of the phonon reservoir. When we choose the common ohmic parametrization $J(\omega) = J_0 \omega e^{-\omega/\omega_c}$ with dimensionless coupling strength J_0 and cutoff frequency ω_c , the integral can be solved exactly. Writing the Bose-Einstein distributions as a geometric series and resumming all separate integral contribution, we finally obtain for the phonon correlation function

$$C_{\text{ph}}^\xi(\tau) = \left[\frac{\Gamma\left(\frac{1+\beta_{\text{ph}}\omega_c+i(\tau-\xi)\omega_c}{\beta_{\text{ph}}\omega_c}\right) \Gamma\left(\frac{1+\beta_{\text{ph}}\omega_c-i(\tau-\xi)\omega_c}{\beta_{\text{ph}}\omega_c}\right)}{\Gamma^2\left(\frac{1+\beta_{\text{ph}}\omega_c}{\beta_{\text{ph}}\omega_c}\right) (1+i(\tau-\xi)\omega_c)} \right]^{J_0}, \quad (5.47)$$

where $\Gamma(x) = \int_0^\infty t^{x-1} e^{-t} dt$ denotes the Γ -function. The observation that $C_{\text{ph}}^\xi(\tau) \stackrel{\text{KAS}}{=} C_{\text{ph}}(\tau - \xi)$ (generally true for energy counting and an initial state that is diagonal in the energy eigenbasis) leads to the relation

$$\gamma_{\text{ph}}^\xi(\omega) = e^{+i\omega\xi} \gamma_{\text{ph}}(\omega). \quad (5.48)$$

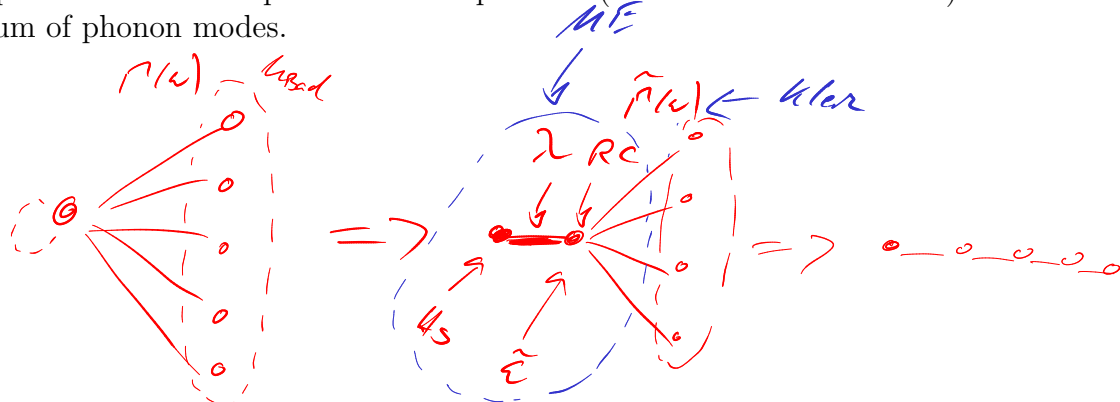
We note from Eq. (5.46) that for particular parametrizations of the spectral coupling density one can expect that for large times the phonon correlation functions may remain finite $\lim_{t \rightarrow \infty} C_{\text{ph}}(\tau) \neq 0$. However, the total correlation function is given by a product of electronic (which decay) and phonon correlation functions. Its Fourier transform (that enters the rates) can be calculated numerically from a convolution integral

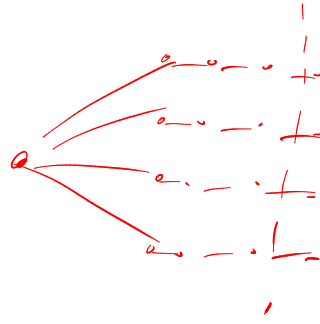
$$\begin{aligned} \gamma_{12}^{\nu, \xi_\nu}(-\epsilon') &= \frac{1}{2\pi} \int d\omega \Gamma_\nu(-\omega) f_\nu(-\omega) \gamma_{\text{ph}}(-\epsilon' - \omega) e^{-i(\epsilon' + \omega)\xi_\nu}, \\ \gamma_{21}^{\nu, \xi_\nu}(+\epsilon') &= \frac{1}{2\pi} \int d\omega \Gamma_\nu(+\omega) [1 - f_\nu(+\omega)] \gamma_{\text{ph}}(+\epsilon' - \omega) e^{+i(\epsilon' - \omega)\xi_\nu}, \end{aligned} \quad (5.49)$$

and enters in this case a rate matrix of the form

$$\mathcal{L}(\chi, \xi_L, \xi_R) = \begin{pmatrix} -\gamma_{12}^L(-\epsilon') & +\gamma_{21}^{L, \xi_L}(+\epsilon') \\ +\gamma_{12}^{L, \xi_L}(-\epsilon') & -\gamma_{21}^L(+\epsilon') \end{pmatrix} + \begin{pmatrix} -\gamma_{12}^R(-\epsilon') & +\gamma_{21}^{R, \xi_R}(+\epsilon') e^{+i\chi} \\ +\gamma_{12}^{R, \xi_R}(-\epsilon') e^{-i\chi} & -\gamma_{21}^R(+\epsilon') \end{pmatrix}, \quad (5.50)$$

from which the electronic matter current can be directly deduced. With the choices $J_0 = \frac{|h|^2}{\Omega^2}$ and $\omega_c = \Omega$ the electronic current is for high temperatures quite similar as if one would have only a single phonon mode. Also the symmetries are similar to that of Eq. (5.37), and a similar fluctuation theorem arises from that. The crucial difference however is that at low temperatures, the phonon plateaus are no longer visible – compare the thin solid versus the bold curves in Fig. 5.3. Since for the continuum model many different phonon frequencies contribute, this is expected. Interestingly however, the current suppression due to the presence of the phonons (Franck-Condon blockade) is also visible for a continuum of phonon modes.





Bibliography

- [1] G. Schaller, T. Krause, T. Brandes, and M. Esposito. Single-electron transistor strongly coupled to vibrations: counting statistics and fluctuation theorem. *New Journal of Physics*, 15:033032, 2013.
- [2] J. Koch and F. von Oppen. Franck-condon blockade and giant Fano factors in transport through single molecules. *Physical Review Letters*, 94:206804, 2005.
- [3] G. D. Mahan. *Many-Particle Physics*. Springer Netherlands, 2000.
- [4] T. Brandes. Coherent and collective quantum optical effects in mesoscopic systems. *Physics Reports*, 408:315–474, 2005.
- [5] Milton Abramowitz and Irene A. Stegun, editors. *Handbook of Mathematical Functions*. National Bureau of Standards, 1970.
- [6] Anqi Mu, Bijay Kumar Agarwalla, Gernot Schaller, and Dvira Segal. Qubit absorption refrigerator at strong coupling. *New Journal of Physics*, 19:123034, 2017.