Weak coupling approximations in non-Markovian Transport

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We study the transport properties of the Fano-Anderson model with non-Markovian effects, which are introduced by making one tunneling rate energy-dependent. We show that the non-Markovian master equation may fail if these effects are strong. We evaluate the stationary current, the zero frequency current noise and the occupation dynamics of the resonant level by means of a quantum master equation approximation within different approximation schemes and compare the results to the exact solution obtained by scattering theory and Green’s functions.

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I. INTRODUCTION

The ongoing progress in measuring tiny fluctuations of charge currents through nanoscale conductors has led to an increased theoretical interest in non-Markovian effects revealed in such experiments. Of particular interest are the non-Markovian dynamics induced by the coupling to fermionic and/or bosonic environments and their influence on steady-state transport observables such as the cumulants of the stochastic charge transfer process. Our present work focuses on such effects due to the coupling to electronic reservoirs.

There exist various techniques for describing open quantum systems coupled to fermionic reservoirs, e.g., scattering theory and Green’s functions, or quantum master equations starting from the von-Neumann equation for the total density operator, or the Wigner-Boltzmann approach. Master equations are widely considered with a conductor-lead coupling in Born-Markov approximation (see e.g. ), which is only strictly valid for weak coupling and constant contact density of states in the energy range of interest. Consequently, to explore effects beyond the common Born-Markov-approximation, one would study higher-order perturbation theory in the contact coupling (e.g. ), and/or allow for energy-dependent tunneling rates, i.e., go beyond the wide-band approximation. In this work we choose the latter whilst analyzing perturbative approaches in lowest-order tunnel coupling only. In particular, we address the question of in which limits it is possible to describe non-Markovian physics with master equations.

A conceptionally simple model for this purpose consists of a single resonant level (e.g. the ground state of a quantum dot or a molecule) weakly coupled to two electronic leads in equilibrium and a Lorentzian-shaped density of states for one of the leads. This model is equivalent to two serially coupled quantum dots and can be interpreted as a quantum dot coupled to a reservoir with finite electron relaxation time.

To enable an exact solution, and for the sake of simplicity, we neglect Coulomb interaction and, consequently, prominent effects like Coulomb blockade or Kondo correlations. This, of course, constrains the use of our results for quantitative understandings of transport experiments. Our aim, however, is a comparison of different approximation schemes for master equations in an electronic transport problem that has an exact solution.

We will show that the non-Markovian master equation (NMME) in the wide-band limit produces reasonable results for the current and noise. However, reducing the band-width yields qualitative and quantitative deviations — even the emergence of unphysical results, like negative Fano factors. The exact time evolution of the resonant level occupation can only be obtained by the NMME in the wide-band limit where even a Markovian master equation covers the exact dynamics. In the short-time limit, the NMME result well approximates the exact evolution regardless of the bandwidth. However, for very small bandwidths, the NMME generates negative, unphysical probabilities. We demonstrate how this can be avoided by a dynamical coarse graining method.

The paper is organized as follows: In Secs. II.A and II.B. we introduce the model and provide the known exact solution obtained by scattering theory and Green’s functions. In Sec. II.C. the equivalence to the double-dot model is discussed. In Sec. III.A we introduce the non-Markovian master equation and in Sec. III.B. the dynamical-coarse-graining approach. In Sec. IV the steady-state current and the Fano factor are compared to the exact and NMME solution. Finally, the occupation dynamics of the resonant level is discussed.

II. MODEL

A. Hamiltonian

We start from the well-known single resonant level model that is described by a Hamiltonian where tunneling between two leads can be realized via a localized quantum dot state,

\[ H = \epsilon_d d_\dagger d + \sum_{k,a} \epsilon_{ka} c_{ka}^\dagger c_{ka} + \sum_{k,a} (t_{ka} d_\dagger c_{ka} + \text{h.c.}) \, . \]  (1)

The fermionic operators annihilate/create an electron on the dot and annihilate/create an elec-
electron in lead $a \in \{ R, L \}$ with momentum $k$. We choose a Lorentzian-shaped right tunneling rate, like in\cite{25,26}:

$$
\Gamma_R(\omega) = \frac{\Gamma_{R,0}^2}{\pi} \left( \frac{\delta_R}{(\omega - \epsilon_R)^2 + \delta_R^2} \right),
$$

and a flat left tunneling rate $\Gamma_L$, both related to the microscopic parameters via $\Gamma_a(\omega) = 2\pi \sum_k |t_{ka}|^2 \delta(\omega - \epsilon_{ka})$. A sketch of the model is given in Fig. 1. We work in the infinite bias limit, where we couple the left and right lead to two particle reservoirs in a way that the left lead is completely occupied and the right lead is completely empty, i.e., $\mu_L \to \infty$ and $\mu_R \to -\infty$ such that the left Fermi function takes the value 1 and the right one takes the value 0.

The retarded and advanced self energies of our model are evaluated exactly\cite{14,32,33,34} as

$$
\Sigma^{R/A}(\omega) = \sum_{k,a} \frac{|t_{ka}|^2}{\omega - \epsilon_{ka} \pm \delta}, = \Lambda(\omega) \mp i \frac{\Gamma(\omega)}{2}.
$$

The imaginary part $\Gamma(\omega) = \Gamma_L(\omega) + \Gamma_R(\omega)$ is connected to the inverse lifetime, whereas the real part

$$
\Lambda(\omega) = \frac{1}{2\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\Gamma(\omega')}{\omega - \omega'},
$$

(where $\mathcal{P}$ denotes the principal value) induces a level shift which is fully determined by the tunneling rate (Kramers-Kronig relation). It will turn out that all quantities concerning transport statistics and the occupation of the quantum dot can be expressed in terms of $\Gamma_a(\omega)$ and $\epsilon_d$ only.

### B. Exact solution

To evaluate transport quantities we need the transmission coefficient\cite{35},

$$
T(\omega) = \frac{\Gamma_L(\omega)\Gamma_R(\omega)}{|\omega - \epsilon_d - \Sigma_R(\omega)|^2},
$$

which is related to the dot spectral function $A(\omega) := i \left[ GR(\omega) - G^A(\omega) \right]$ via $T(\omega) = \frac{\Gamma_L(\omega)\Gamma_R(\omega)}{1 + i\delta(\omega)} A(\omega)$. Scattering theory then yields the current $I$ and the zero-frequency noise $S$ which at infinite bias are

$$
I = e \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} T(\omega),
$$

$$
S = e^2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} T(\omega) \left[ 1 - T(\omega) \right],
$$

where $e$ is the elementary charge (chosen negative). The time-dependent occupation probability $n_d(t)$ can be expressed with Green’s functions\cite{36,37}. We use that at infinite bias the left Fermi function is unity and the right is zero. For simplicity we assume $n_d(0) = 0$ and thus

$$
n_d(t) = -i \int \sum_k G_{d,ka}(t) i G^A_{d,kL}(0) = \sum_k \left[ G_{d,ka}(t) \right]^*,
$$

where we have used $G^A_{ka,\pm}(t) = \left[ G_{d,ka}(t) \right]^*$.

The required Green’s functions are obtained using equations of motion\cite{14,32,33,34} and read

$$
G^R_{d,kL}(t) = t_{kL} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{1}{\omega - \epsilon_{kL} + i0^+} e^{-i\omega t},
$$

which, upon inserting $1 = \int_{-\infty}^{\infty} \delta(\omega - \epsilon_{kL}) d\omega$, yields the explicit result

$$
n_d(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_L \times \
\left[ \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{1}{\omega' - \omega + i0^+} e^{-i\omega' t} \right]^2.
$$

This can alternatively be obtained from a direct calculation without Green’s functions\cite{38}.

For the Lorentzian-shaped right tunneling rate $\frac{\delta_R}{2}$, the Kramers-Kronig-relation\cite{11} yields the level-shift function

$$
\Lambda(\omega) = \frac{\Gamma_{R,0}^2}{2\pi} \left( \frac{\omega - \epsilon_R}{(\omega - \epsilon_R)^2 + \delta_R^2} \right),
$$

and the self energy\cite{35} can be simplified to

$$
\Sigma^{R/A}(\omega) = \mp i \frac{\Gamma_L}{2} + \frac{\Gamma_{R,0}^2}{2\pi} \frac{1}{\omega - \epsilon_R \pm i\delta_R}.
$$

With Lorentzian shaped tunneling rates we can analytically integrate the expressions for current and noise,
and with the abbreviations \( \tilde{\Gamma} := \Gamma_L + 2\delta_R \) and \( \epsilon := \epsilon_d - \epsilon_R \), we obtain

\[
I = e^{2\Gamma_L} \frac{2\Gamma_L \delta_R \Gamma_{R,0}/\pi}{(\Gamma_L \delta_R + \Gamma_{R,0}/\pi)^2 + 4e^2 \Gamma_L \delta_R},
\]

\[
S = I e \left( 1 - \frac{2\Gamma_L \delta_R \Gamma_{R,0}/\pi}{\pi \Gamma R} \times \frac{4e^2 (\Gamma_L^2 + 8\delta_R^2) + \Gamma^4 (2\Gamma_L \delta_R + 2\Gamma_{R,0}/\pi)^2}{(\Gamma_L \delta_R + \Gamma_{R,0}/\pi)^2 + 4e^2 \Gamma_L \delta_R} \right).
\]

(12)

C. Comparison with double dot model

Our model has an exact correspondence with an effective wide band two level Fano-Anderson model\(^{25}\) with Hamiltonian

\[
\hat{H} = \epsilon_L d_L^\dagger d_L + \epsilon_R d_R^\dagger d_R + \hat{T}_C d_L^\dagger d_R + \hat{T}_C^\dagger d_R^\dagger d_L + \sum_{k,a} \epsilon_{ka} c_{ka}^\dagger c_{ka} + \sum_{k,a} (\epsilon_{ka} c_{ka}^\dagger c_{ka} + \text{h.c.})
\]

(13)

with a left and a right dot state, \( d_L \) and \( d_R \), and a coupling \( \hat{T}_C \) between them. Each lead couples only to its adjacent dot. The transmission probability is\(^{39}\)

\[
\tilde{T}(\omega) = \frac{\Gamma_L \Gamma_R |\tilde{T}_C|^2}{|\omega - \epsilon_L + i\Gamma_L/2|(|\omega - \epsilon_R + i\Gamma_R/2| - |\tilde{T}_C|^2)}.
\]

Comparison to Eq. \( [5] \), with appropriate \( \Gamma_R(\omega) \) and \( \Lambda(\omega) \), reveals the exact mapping \( \tilde{T}(\omega) = T(\omega) \) by the correspondence

\[
\begin{align*}
\Gamma_L & \leftrightarrow \Gamma_L, \\
|\tilde{T}_C|^2 & \leftrightarrow \Gamma_{R,0}/(2\pi), \\
\Gamma_R & \leftrightarrow 2\delta_R, \\
\epsilon_L & \leftrightarrow \epsilon_d, \\
\epsilon_R & \leftrightarrow \epsilon_R.
\end{align*}
\]

(15)

Since the cumulant generating function can be expressed solely in terms of the transmission probabilities\(^{40,41}\), not only the first two cumulants\(^{25}\), but all current cumulants, of the two models coincide.

When we plot current and Fano factor \( F = S/(Ie) \) as a function of the detuning \( \epsilon_d - \epsilon_R \) in Fig. \( [2] \) we find the typical structures of the corresponding quantities for the non-interacting double quantum dot\(^{42,43}\). The current exhibits a maximum when the detuning vanishes. For the noise we find a minimum in resonance for broad tunneling rates, when \( \delta_R \) becomes sufficiently small a new maximum in the Fano factor at resonance grows out of the minimum.

III. NON-MARKOVIAN DYNAMICS

A. Non-Markovian Master Equation (NMME)

To treat non-Markovian effects in a transport master equation framework one writes the master equation as an integro-differential equation for the \( n \)-resolved reduced density matrix

\[
\dot{\rho}_n(t) = \sum_{n'} \int_0^t \mathcal{W}_{n-n'}(t-t')\rho_{n'}(t')dt',
\]

(16)

where \( n \) denotes the number of charges that have crossed the considered system. In Appendix \( [A] \) we show how our system can be described with such an equation using the Born approximation. All \( \mathcal{W}_n(t) \) vanish except when \( n = 0 \) or \( n = 1 \), where we find

\[
\begin{align*}
\mathcal{W}_0(t) &= \begin{pmatrix} -\gamma_L(t) & 0 \\ \gamma_L(t) & -\gamma_R(t) \end{pmatrix}, \\
\mathcal{W}_1(t) &= \begin{pmatrix} 0 & \gamma_R(t) \\ 0 & 0 \end{pmatrix}
\end{align*}
\]

(17)

with

\[
\begin{align*}
\gamma_R(t) &= \frac{2}{2\pi} \int \frac{d\omega}{\pi} \Gamma_R(\omega) \cos([\omega - \epsilon_d]t) \\
\gamma_L(t) &= \frac{2}{2\pi} \int \frac{d\omega}{\pi} \Gamma_L \cos([\omega - \epsilon_d]t)
\end{align*}
\]

(18)

We proceed with performing a Fourier summation and a Laplace transform via

\[
\mathcal{W}(\chi, z) = \sum_n e^{inz} \int_0^\infty e^{-zt} \mathcal{W}_n(t)dt = \mathcal{W}_0(z) + \mathcal{W}_1(z)e^{i\chi}
\]

(19)

where

\[
\begin{align*}
\hat{\gamma}_R(z) &= \frac{\Gamma_{R,0}}{\pi} \frac{z + \delta_R}{(z + \delta_R)^2 + (\epsilon_R - \epsilon_d)^2}, \\
\hat{\gamma}_L(z) &= \Gamma_L.
\end{align*}
\]

(20)

We will use the abbreviation \( \hat{\gamma}(z) = \hat{\gamma}_L(z) + \hat{\gamma}_R(z) \). In order to avoid the tedious inverse Laplace transform we can use a recently developed elegant method to evaluate current and noise\(^{11,44}\). As it has only applied a few times until now\(^{11,44,45,46}\), we show the explicit calculation for our model in appendix \( [3] \) and obtain the following...
for the right tunneling rate \( \Gamma_R(\omega) \) as a function of the energy \( \omega \). We further show how the current \( I \) and Fano factor \( F \) behave as a function of the detuning \( \epsilon_d - \epsilon_R \). The bandwidth \( \delta_R \) takes the three values shown on the right. For all curves we have \( \Gamma_L = \Gamma_R(0) \). Results from exact Green’s functions.

formulæ:

\[
I = \frac{e^{-\frac{\hat{\gamma}_L(0)\hat{\gamma}_R(0)}{\hat{\gamma}(0)}}}{\hat{\gamma}(0)},
\]

\[
S = \frac{\exp \left\{ \frac{\hat{\gamma}_R(0)}{\hat{\gamma}(0)} \right\} [1 + 2\hat{\gamma}_L(0)]}{\hat{\gamma}(0)}\frac{\exp \left\{ \frac{\hat{\gamma}_L(0)}{\hat{\gamma}(0)} \right\} [1 + 2\hat{\gamma}_R(0)]}{\hat{\gamma}(0)}.
\]

We easily obtain the occupation of the dot by evaluating the density matrix in Laplace space, \( \hat{\rho}(z) = [z - \hat{W}(z)]^{-1} \hat{\rho}(t = 0) \). To obtain the time-resolved dynamics, one has to perform the inverse Laplace transform (Bromwich integral) by collecting all the corresponding residues.

Markovian master equation (MME) — A Markovian master equation follows from \[ \text{(10)} \] by the integration \( \mathcal{L}_n = \int_0^\infty \mathcal{W}_n(t)dt = \mathcal{W}_n(z = 0) \) and leads to

\[
\mathcal{L}_0 = \begin{pmatrix} -\Gamma_L & 0 \\ \Gamma_L & -\Gamma_R(\epsilon_d) \end{pmatrix}, \quad \mathcal{L}_1 = \begin{pmatrix} 0 & \Gamma_R(\epsilon_d) \\ \Gamma_L & 0 \end{pmatrix}.
\]

When we use this to evaluate noise we end up with the same result as Eq. \[ \text{(21)} \], but without the derivatives \( \hat{\gamma}_L(0) \) and \( \hat{\gamma}_R(0) \). For the equilibrium density matrix we find \( \rho_{00}(t = \infty) = \Gamma_R(\epsilon_d)/(\Gamma_L + \Gamma_R(\epsilon_d)) \), which is identical with the NMME result. The advantage of the additional Markovian approximation is that the positivity of the density matrix will be conserved as one obtains a Lindblad type master equation. \[ \text{(22)} \]

the disadvantage is that some information about the shape of the tunneling rates is lost.

B. Dynamical Coarse Graining (DCG)

A second approach to quantum transport is the recently developed dynamical coarse graining method.\[ \text{(15)} \] The coarse graining method is also a second order weak coupling approximation although it can be extended to higher orders. Instead of solving a single master equation, it solves a continuous set \( \dot{\rho}_\tau(t) = \mathcal{L}_\tau \rho_\tau(t) \) and then interpolates through the solutions \( \rho_\tau(t) = e^{\mathcal{L}_\tau \cdot t} \rho_0 \) at \( t = \tau \). The coarse grained Liouvillian can be derived by matching the second order expansion of the formal solution in the interaction picture \( \chi(t) = U(t) \chi(0) U^\dagger(t) \) to the second order expansion of \( \rho_\tau(t) = e^{\mathcal{L}_\tau \cdot t} \rho_0 \) at time \( t = \tau \). For our specific model, following reference we obtain

\[
\mathcal{L}_\tau = \int_{-\infty}^{\infty} d\omega \frac{\sin(\omega - \epsilon_d)}{2\pi} \left( \frac{-\Gamma_L(\omega) + \Gamma_R(\omega)}{\Gamma_L(\omega) - \Gamma_R(\omega)} \right),
\]

where \( \sin x = \frac{\sin x}{x} \). The Coarse Graining method combines the two advantages of the Born and the Born-Markov approximation: For finite times it is sensitive to the shape of the tunneling rates, and at the same time it preserves positivity since the \( \mathcal{L} \) are of Lindblad form. Due to the identity \[ \lim_{\tau \to -\infty} \tau \cdot \sin \left[ \frac{(\omega - \epsilon_d)}{2} \right] = 2\pi \delta(\omega - \epsilon_d) \] we see that for large times the coarse graining method yields the same steady state density matrix as the two other approximations. Also for current and Fano factor we have reproduced the results of the MME. In contrast to fixed graining-time derivations of master equation, DCG dynamically adapts the coarse-graining time with the physical time, which in the long-time limit yields the Born Markov secular approximation. Both approaches yield completely positive maps but may lead to different stationary states.

IV. DISCUSSION

A. Current

To get a first idea of the difference between the exact solution and the NMME it is instructive to take a look at the stationary current \( I \). The stationary current is not sensitive to non-Markovian effects, since it only depends on the steady state occupation, and thus all three
We show results for the Fano factor \( \epsilon_d - \epsilon_R \) indicated in each plot.

**B. Fano factor**

In Fig. 4 we show results for the Fano factor \( F = S/(Ie) \) (exact, with NMME and with MME/DCG). When the condition

\[
\frac{\delta R}{\Gamma_{R,0}} \gg 1
\]  

is fulfilled, all formalisms agree. Furthermore, for constant tunneling rates, the NMME yields the Markovian result \( F = \frac{\Gamma_{L,0}^2 + \Gamma_{R,0}^2 (\epsilon_d - \epsilon_R)}{(\Gamma_{L,0} + \Gamma_{R,0})^2} \).

The exact Fano factor shows one minimum for small detuning and two minima for large detuning, which originates from the double peak structure of the spectral function shown in Fig. 3. However, there is no simple quantitative connection between the locations of the extrema in the Fano factor and in the spectral function. This is similar to the Fano factor as function of the detuning in Fig. 2 where for small width \( \delta R \) a second minimum appears, that is again not connected to the spectral function’s properties in a simple way.

The NMME reproduces one of the two minima, but not both, and it produces super-Poissonian noise, where it should not appear. If the detuning \( \epsilon_d - \epsilon_R \) becomes very small, the NMME even overestimates the minimum so strongly that it yields an unphysical negative Fano factor.

The MME is not everywhere close to the exact result, but it is, on average, closer than the NMME result, and per construction yields physical results (Lindblad form). For the DCG method we have found that it yields the same result as the MME.
we find that at sufficiently wide

\[ \delta_R \geq \Gamma_{R,0} \]

all three approximations meet the exact result very well.

For smaller \( \delta_R \) one recognizes that in the stationary limit, the three approximations coincide with each other but not with the exact solution. The most prominent feature of the exact occupation \( n_d(t) \) are oscillations as a function of time \( t \). The NMME approximation captures the oscillations as non-Markovian features of the reservoirs, as it should, but it strongly overestimates them. If the steady state is sufficiently close to zero, the NMME can lead to negative occupation probabilities, like in the third plot.

In order to better understand the exact result, we explicitly perform the two integrations in Eq. (25), which leads to

\[
n_d(t) = \\
\delta_R (\Gamma^2 + 4 \epsilon^2) + \Gamma \frac{\Gamma_{R,0}^2}{\pi} + 2 \Gamma L \ell \left( \frac{\omega_1 - \omega_2}{\omega_1 - \omega_2} \right) \epsilon (\omega_0 - \Gamma/2) t, \tag{25}
\]

with the abbreviations \( \bar{\Gamma} \) := \( \Gamma_{L} + 2 \delta_R \) and \( \epsilon := \epsilon_d - \epsilon_R \).

The frequencies \( \omega_1 \) and \( \omega_2 \) are the poles of the spectral function \( A(\omega) \) with negative imaginary part, and \( \omega_1^* \) and \( \omega_2^* \) are its other two poles. The first part of \( n(t) \) in Eq. (25) corresponds to the steady state. The next two lines describe how the system performs exponential decay towards this steady state, because the exponents are real and negative. The last line of Eq. (25) is responsible for the oscillations, because its exponent contains the imaginary part \( i \omega_0 t \), which is given by

\[
\omega_0 = \text{Re} \left\{ \left[ (\epsilon_d + \epsilon_R - i \bar{\Gamma}/2)^2 \right. \right.
\]

\[
\left. -4(\epsilon_R - i \delta_R)(\epsilon_d - i \Gamma L/2) + 4 \frac{\Gamma_{R,0}^2}{2\pi} \right]^{1/2} \right\}. \tag{26}
\]

If we switch off the coupling to the left side and the width on the right, i.e., \( \Gamma_{L} \to 0 \) and \( \delta_R \to 0 \), we recover the frequency:

\[
\omega_0 = \sqrt{\epsilon^2 + 4 \frac{\Gamma_{R,0}^2}{2\pi}}
\]

doing coherent oscillations in an isolated two-level system (remember the mapping to the double dot model in Eq. (14)). In Fig. 6 one recognizes that in the absence of the detuning \( \epsilon_d \to \epsilon_R \), the frequency \( \omega_0 \) completely vanishes with a nonanalyticity where \( \delta_R/\Gamma_{R,0} \) is of the order of 1. This nonanalyticity appears, when the radicand in Eq. (26) changes sign (at \( \epsilon_d = \epsilon_R \) it is purely real). When detuning is present, the frequency \( \omega_0 \) does not completely vanish for \( \delta_R \to \infty \), but the oscillations of \( n_d(t) \) are damped with the rate \( \Gamma/2 = \delta_R + \Gamma L/2 \), such that in the Markovian limit, \( \delta_R \to \infty \), no oscillations survive.

C. Time resolved occupation probabilities

One key advantage of the DCG approach results from its ability to preserve positivity and at the same time to be more sensitive to the shape of the tunneling rates than the MME\textsuperscript{48}. We therefore evaluate the time dependent occupation probability of the localized level with four different methods: exact, with the NMME, with the MME, and with DCG. In Fig. 5 we find that at sufficiently wide bands, i.e., \( \delta_R \geq \Gamma_{R,0} \), all three approximations meet the exact result very well.

FIG. 5: Time dependent occupation probability \( n_d(t) \), exact solution and solution in the three presented approximations. Parameters are \( \epsilon_d = \Gamma_{R,0} \) and \( \epsilon_d - \epsilon_R = \Gamma_L = 0.1 \Gamma_{R,0} \). The width \( \delta_R \) takes the values denoted in each plot.

FIG. 6: Frequency \( \omega_0 \) as a function of the detuning \( \omega_d - \omega_R \) for the parameters \( \Gamma_L = 0.1 \Gamma_{R,0} \) and \( \epsilon_d = \Gamma_{R,0} \).
V. CONCLUSIONS

The main message of this paper is that one must be careful with non-Markovian master equations (NMMEs). We know that what we have here called “Markovian master equations” (MMEs) may lead to incorrect results when non-Markovian effects are strong. However, we have found that in the non-Markovian regime NMMEs can be worse than MMEs. On the one hand, NMMEs can give quantitative errors such as occur in Fig. 4. On the other, the positivity of the density matrix is not in general conserved, which can lead to unphysical results like a negative Fano factor and occupation probability, as in Fig. 4 and 5. We emphasize that the failure of the NMME does not result from the new evaluation techniques we have used, but instead from the way the Born approximation is performed in the derivation of Eq. 4.

We do not want to discourage using of NMMEs in general, but each time they are applied one should reason carefully why one expects them to yield better results than MMEs.

We have tried the dynamical coarse graining (DCG) method as an alternative approach that is more sophisticated than the MME, but still in Lindblad form. For the time-independent quantities I, F and n(t → ∞) we have found no improvement in comparison to the MME. For the time-dependent occupation probability however, DCG does yield better results, at least for small times. We expect a similar improvement for the frequency-dependent Fano factor.

For the model considered here, the condition $\frac{\Gamma_n}{\delta n} \ll 1$ must be fulfilled to ensure a good quality of the three presented weak coupling approximations. It is certainly interesting to explore higher-order corrections to the presented perturbative approximations. However, such extensions should be treated with caution, since nice features such as positivity may be lost when higher orders are included.

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APPENDIX A: DERIVATION OF THE KERNEL

We decompose our Hamiltonian into the free part $H_0 = \epsilon_d d^+ d + \sum_{k,a} \epsilon_{ka} c_k^+ c_{ka}$ and the coupling $V = \sum_{k,a}(t_{ka} d^+ c_{ka} + t_{ka}^* c_{ka}^+ d)$. This enables us to define the density matrix in the interaction picture, $\tilde{\rho}(t)$. It obeys the Liouville-von-Neumann equation

$$\dot{\tilde{\rho}}(t) = -i[\tilde{V}(t),\tilde{\rho}(t)],$$

where $\tilde{V}(t) = e^{iH_0 t} V e^{-iH_0 t}$ is the interaction picture version of the coupling. A tilde will mark the interaction picture in all following text. By iterating Eq. (A1) twice, we obtain

$$\dot{\tilde{\rho}}(t) = -i[\tilde{V}(t),\tilde{\rho}(t)] - \int_0^t dt'[\tilde{V}(t'),[\tilde{V}(t'),\tilde{\rho}(t')]].$$

At this point we perform the second-order weak coupling approximation by replacing the full density matrix $\tilde{\rho}(t')$ by a tensor product of the reduced density matrix $\tilde{\rho}(t)$ and the bath density matrix $R_0$, which we assume to be constant in time (Born approximation). The partial trace over the first commutator in equation (A2) vanishes. To proceed we resolve the coupling into system operators $S_i$ and bath operators $B_j$ such that

$$\tilde{V}(t) = \tilde{S}_1(t) \tilde{B}_1(t) + \tilde{B}_2(t) \tilde{S}_2(t).$$

where

$$\tilde{S}_1(t) = \tilde{d}^+(t), \quad \tilde{B}_1(t) = \sum_{k,a} t_{ka} \tilde{c}_{ka}(t),$$

$$\tilde{S}_2(t) = \tilde{d}(t), \quad \tilde{B}_2(t) = \sum_{k,a} t_{ka}^* \tilde{c}_{ka}^+(t).$$

In the calculation that follows we find that the off-diagonal elements of $\tilde{\rho}(t)$ decouple from the diagonal ones. With this knowledge we can choose the off-diagonals to be zero and neglect them in the density matrix, which we can therefore consider as a vector with two entries:

$$\tilde{\rho}(t) = \tilde{\rho}_{00}(t)d^+d + \tilde{\rho}_{11}(t)d^+d = \left(\begin{array}{c} \tilde{\rho}_{00}(t) \\ \tilde{\rho}_{11}(t) \end{array}\right).$$

To evaluate the double commutator in equation (A2) is lengthy but straightforward and yields

$$\dot{\tilde{\rho}}(t) = \int_0^t dt' W(t-t') \tilde{\rho}(t')$$

with

$$W(t) = \left(\begin{array}{cc} -\gamma_L(t) & \gamma_R(t) \\ \gamma_L(t) & -\gamma_R(t) \end{array}\right).$$

For the entries of $W(t)$ we need to explicitly perform traces over the reservoirs:

$$\gamma_L(t) = e^{i\epsilon_d t} \text{Tr}\{\tilde{B}_2(0) \tilde{B}_1(t) R_0\} + e^{-i\epsilon_d t} \text{Tr}\{\tilde{B}_2(t) \tilde{B}_1(0) R_0\}.$$  

With infinite bias, i.e., $R_0 = \sum_{k} \frac{1}{\gamma_L} c_k L c_k L$ this becomes

$$\gamma_L(t) = \sum_{k} |t_{kl}|^2 \cdot 2 \cos(|\epsilon_L - \epsilon_d| t)$$

$$= 2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_L(\omega) \cos[(\omega - \epsilon_d) t].$$

In the same manner we get

$$\gamma_R(t) = 2 \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Gamma_R(\omega) \cos[(\omega - \epsilon_d) t].$$

Physically, the upper right matrix element of $W(t)$ describes a jump from the dot to the right lead. Thus we can say that it increases the number of passed electrons by one, while all other matrix elements leave it unchanged. Thus we distinguish

$$W_0(t) = \left(\begin{array}{cc} -\gamma_L(t) & 0 \\ \gamma_L(t) & -\gamma_R(t) \end{array}\right),$$

$$W_1(t) = \left(\begin{array}{cc} 0 & \gamma_R(t) \\ \gamma_L(t) & 0 \end{array}\right).$$

APPENDIX B: EVALUATION OF CURRENT AND NOISE

We use a bra-ket like notation where the equilibrium state is represented by the ket

$$|0\rangle = \lim_{t \to \infty} \rho(t) = \frac{1}{\gamma(0)} \left(\begin{array}{c} \tilde{\gamma}_R(0) \\ \tilde{\gamma}_L(0) \end{array}\right).$$
that we obtain by setting $\dot{\rho} = 0$ in Eq. \[16\]. We define the bra

$$\langle \tilde{0} | = (1, 1),$$

(B2)

and construct the projector

$$\mathcal{P} = \mathcal{P}^2 = |0\rangle \langle \tilde{0}| = \frac{1}{\gamma(0)} \begin{pmatrix} \gamma_R(0) & \gamma_R(0) \\ \gamma_L(0) & -\gamma_L(0) \end{pmatrix}.$$  \hspace{1cm} (B3)

and the projector $\mathcal{Q} = \mathcal{Q}^2 = \mathbb{1} - \mathcal{P}$. The resolvent of the Laplace-transformed kernel from Eq. \[19\] is $\mathcal{R}(\Lambda, \chi, z) := \mathcal{Q}[\mathcal{W}(\chi, z) - \Lambda \cdot \mathbb{1}]^{-1}\mathcal{Q}$. We only need its value at zero, $\mathcal{R} := \mathcal{R}(0, 0, 0)$, which is

$$\mathcal{R} = \frac{1}{(\gamma_L(0) + \gamma_R(0))^2} \begin{pmatrix} -\gamma_L(0) & \gamma_R(0) \\ \gamma_L(0) & -\gamma_R(0) \end{pmatrix}.$$  \hspace{1cm} (B4)

We introduce the coefficients of the kernel’s Taylor series via

$$\mathcal{W}(\chi, z) = \mathcal{W} + \mathcal{W}' \chi + \mathcal{W}' z + \frac{1}{2}(\mathcal{W}'' \chi^2 + 2\mathcal{W}' \chi z + \mathcal{W}' z^2) + \cdots$$

(B5)

The expressions for current and the zero frequency noise\[11,14] are now obtained as

$$I = e \langle \langle \tilde{0} | \mathcal{W}' | 0 \rangle \langle 0 | \mathcal{W}' | \tilde{0} \rangle \rangle / i,$$

$$S = e^2 \left[ \langle \langle \tilde{0} | \mathcal{W}'' | 0 \rangle \langle 0 | \mathcal{W}' \mathcal{R} | \mathcal{W}' | 0 \rangle \rangle - 2 \langle \langle \tilde{0} | \mathcal{W}' \mathcal{R} | \mathcal{W}' | 0 \rangle \rangle \right] / i^2$$

$$- 2ie \left[ \langle \langle \tilde{0} | \mathcal{W}' | 0 \rangle \langle 0 | \mathcal{W}' \mathcal{R} | \mathcal{W}' | 0 \rangle \rangle - \langle \langle \tilde{0} | \mathcal{W}' \mathcal{R} | \mathcal{W}' | 0 \rangle \rangle \right].$$  \hspace{1cm} (B6)

Performing the derivatives and matrix multiplications, leads to the results of Eq. \[21\].