The Accuracy of Perturbative Master Equations

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We consider open quantum systems with dynamics described by a stationary master equations that has a perturbative expansion in the system-environment interaction. We show that, contrary to intuition, late-time solutions of order-$2n$ accuracy require an order-$(2n+2)$ master equation. We give two examples of such inaccuracies in the solutions to an order-$2n$ master equation, order-$2n$ inaccuracies in the steady state of the system and order-$2n$ positivity violations, and we show how these arise in a specific example for which exact solutions are available. This result has a wide-ranging impact on the validity of coupling (or friction) sensitive results derived from second-order convolutionless, Nakajima-Zwanzig, Redfield, and Born-Markov master equations.

INTRODUCTION

An open quantum system is a quantum system that interacts with some environment whose degrees of freedom have been coarse grained over (i.e., traced out), and its dynamics are described by a master equation governing the reduced density matrix $\boldsymbol{\rho}$. Exact master equations for the stochastic dynamics of open quantum systems are, in general, beyond the reach of the simple theorist. However, perturbative master equations (in the system-environment interaction) can be derived in a variety of different ways [1–3] and find application in many branches of physics and chemistry [4–7]. In the time-local representation (also called the convolutionless or Markovian representation), the dynamics of the reduced density matrix of the system can be expressed with a quantum Liouville equation

$$\frac{d}{dt}\boldsymbol{\rho}(t) = \boldsymbol{\mathcal{L}}(t)\,\boldsymbol{\rho}(t)\,, \tag{1}$$

where, despite the apparent time-local form, non-Markovian behavior may be encapsulated in the time dependence of the Liouvillian $\boldsymbol{\mathcal{L}}(t)$. As a perturbative approximation, $\boldsymbol{\mathcal{L}}(t)$ is expanded powers in the system-environment interaction c and truncated to some order.

We will consider a stationary perturbative master equation, where the Liouvillian $\boldsymbol{\mathcal{L}}(t)$ is time independent. We will assume that the perturbative expansion of $\boldsymbol{\mathcal{L}}$ is in even powers of the coupling, because as we explain in the next section this can naturally arise from a microscopic derivation of the open-system dynamics. The expansion of $\boldsymbol{\mathcal{L}}$ will then take the form

$$\boldsymbol{\mathcal{L}} = \sum_{n=0}^{\infty} \boldsymbol{\mathcal{L}}_{[2n]}\,, \tag{2}$$

$$\boldsymbol{\mathcal{L}}_{[0]}\,\boldsymbol{\rho} = \left[-\imath\,\mathbf{H}, \boldsymbol{\rho}\right]\,, \tag{3}$$

where $\boldsymbol{\mathcal{L}}_{[2n]} = \mathcal{O}(c^{2n})$ and to zeroth-order the system is driven in a unitary manner by its Hamiltonian $\mathbf{H}$.

The most well-known perturbative master equation is the second-order master equation, as it can be equivalent to the Redfield and Born-Markov master equations. This is partly due to the fact that in the Markovian limit, the second-order master equation is exact. But equivalence with the previous approximate master equations does not carry to fourth-order and there perturbation theory is strictly superior. One might easily assume that solving the second-order master equation defined by the Liouvillian $\boldsymbol{\mathcal{L}}_{[0]} + \boldsymbol{\mathcal{L}}_{[2]}$ would yield a solution that would match the exact solution to the exact master equation up to second order, having error terms of order $\mathcal{O}(c^4)$; however we will show that in general they will differ by second-order terms, so that one can only say they are in perturbative agreement at zeroth order.

One very significant implication of these facts is for the positivity. Not being exact, nor generally of Lindblad form [8, 9], exact complete positivity is not guaranteed for solutions to a perturbative master equation. Solutions can and should be completely positive to the relevant perturbative order, and as we show in this work that order is not what one might naively expect. Solutions to the second-order master equation can in general violate positivity by an amount that is $\mathcal{O}(c^2)$. We show that to find solutions good to second-order, canonical perturbation theory generally demands the fourth-order Liouvillian.

MASTER EQUATIONS FROM A MICROSCOPIC MODEL

In a system derived from a microscopic model, the coarse-grained environment can act as a source of noise, dissipation, and decoherence; thus its influence provides a model of dissipative quantum mechanics more general than Markovian (white-noise) models which can be constructed more phenomenologically. Such microscopic models can still lead to a stationary Liouvillian of the type we specified in Eq. (2).

Given a stationary system Hamiltonian and stationary bath correlations, Gaussian noise distributionals (e.g. noise generated via linear coupling to an environment of harmonic oscillators) *may* allow the master equation to have a stationary late-time limit [10]

$$\boldsymbol{\mathcal{L}}(\infty) = \lim_{t\to\infty} \boldsymbol{\mathcal{L}}(t) \,, \tag{4}$$

so that the late-time and weak-coupling limits commute; otherwise perturbation theory cannot be used for long durations of time. Gaussian noise processes are categorized by their second-order noise correlation, and whether or not the master equation will have a stationary limit is dependent upon how localized this noise correlation is. Well-localized noise correlations (e.g. Gaussian or exponential) can lead to a very well-behaved master equation, whereas long-ranged noise correlations (e.g. Cauchy) can produce a more pathological master equation which cannot be accurately analyzed in a perturbative fashion. Exact examples of this phenomena are given in Ref. [11] in the context of quantum Brownian motion with Ohmic and sub-Ohmic couplings. Moreover, the exact solutions $\boldsymbol{\rho}(t)$ can be very well-behaved even if $\boldsymbol{\mathcal{L}}(t)$ is not. Markovian representations (and, more generally, effective equations of motion) are not always suitable, particularly for highly non-Markovian dynamics.

ACCURACY OF SOLUTIONS

It is clear that if Eqs. (1) and (2) are well defined then for sufficiently short times an order-$2n$ master equation (in which the sum in Eq. (2) only includes terms up to order $2n$) can produce a solution that is also accurate to order $2n$. What we find is that for a stationary master equation, or a master equation that is suitably convergent at late times as discussed in the previous section, a solution to the order-$2n$ master equation is only accurate to order $2n-2$ at later times. The reason is an ultimately mundane but slightly subtle result of degenerate perturbation theory.

Assuming we have the perturbative expansion of a stationary master equation (i.e., an expansion of $\boldsymbol{\mathcal{L}}$), we then seek perturbative solutions obtained by applying canonical perturbation theory of the eigenvalue problem

$$\boldsymbol{\mathcal{L}}\, \mathbf{o} = f\, \mathbf{o} \,, \tag{5}$$

given that we already know the zeroth-order solutions

$$\boldsymbol{\mathcal{L}}_{[0]}\, |\omega_i\rangle\langle\omega_j| = -\imath\, \omega_{ij}\, |\omega_i\rangle\langle\omega_j| \;, \tag{6}$$

where $\mathbf{H}\,|\omega\rangle = \omega\,|\omega\rangle$ and $\omega_{ij} = \omega_i - \omega_j$ denote the (free) energy basis of the system. In the appropriate regime of validity, exact solutions to the perturbative master equation should agree with the perturbative solutions to the exact master equation up to the appropriate order.

Before determining what the appropriate level of accuracy is, we will first demonstrate that there is issue with the naive expectation of order-$2n$ accuracy. This argument is a generalization of one found in Ref. [12], where the discrepancy was noticed for the second-order equilibrium state. Let $\mathbf{o}_{(2n)}$ be any eigenoperator of the Liouvillian which is zeroth-order stationary (diagonal) and supposedly accurate to order $2n$.

$$\boldsymbol{\mathcal{L}}\{\mathbf{o}_{(2n)}\} = f_{(2n)}\, \mathbf{o}_{(2n)} + \mathcal{O}(c^{2n+2}) \,, \tag{7}$$

where $f_{(2n)}$ is its associated eigenvalue and is accurate to order $2n$ and vanishing at zeroth-order. Then one may add any order-$2n$ operator $\boldsymbol{\delta o}_{[2n]}$ that is zeroth-order-stationary (diagonal) and still have

$$\boldsymbol{\mathcal{L}}\{\mathbf{o}_{(2n)} + \boldsymbol{\delta o}_{[2n]}\} = f_{(2n)}\, \{\mathbf{o}_{(2n)} + \boldsymbol{\delta o}_{[2n]}\} + \mathcal{O}(c^{2n+2}) \tag{8}$$

which demonstrates that there is an order $2n$ ambiguity in the diagonal entries if one only compares terms up to order $2n$.

Now we will proceed to our main proof where we show how this issue arises, that this is the full extent of the problem, and precisely how it can be remedied. Note that perturbation theory with master equations is always degenerate perturbation theory as $\omega_{ii} = \omega_{jj} = 0$ trivially. For simplicity let us assume no other degeneracy in the spectrum of the free Liouvillian (though the possibility of extra degeneracy or near degeneracy arising from resonance can be suitably dealt with). This degenerate subspace corresponds to the space of operators that are diagonal in the energy basis of the free system.

Perturbation theory tells us that the second-order corrections to all eigenvalues and eigenoperators of $\boldsymbol{\mathcal{L}}$ outside the degenerate subspace (off-diagonal operators) can be computed using only the second-order master equation:

$$f_{ij}^{[2]} = \langle\omega_i|\, \boldsymbol{\mathcal{L}}_{[2]}\{|\omega_i\rangle\langle\omega_j|\}\, |\omega_j\rangle \;, \tag{9}$$

$$\langle\omega_{i'}|\, \mathbf{o}_{ij}^{[2]}\, |\omega_{j'}\rangle = \frac{\langle\omega_{i'}|\, \boldsymbol{\mathcal{L}}_{[2]}\{|\omega_i\rangle\langle\omega_j|\}\, |\omega_{j'}\rangle}{-\imath(\omega_{ij} - \omega_{i'j'})} \,. \tag{10}$$

As usual in degenerate perturbation theory, to compute the corrections to eigenoperators from the degenerate subspace, which all satisfy $\boldsymbol{\mathcal{L}}_{[0]}\, \mathbf{o}^{[0]} = \mathbf{0}$, we must first find diagonalize $\boldsymbol{\mathcal{L}}$ on the subspace to find the basis which is compatible with the branching under perturbation. The associated characteristic equation is

$$\mathbf{W}\, \vec{o} = f\, \vec{o} \,, \tag{11}$$

$$\vec{o}_i \equiv \langle\omega_i|\, \mathbf{o}\, |\omega_i\rangle \;, \tag{12}$$

where $\mathbf{W}$ defines the Pauli master equation

$$\langle\omega_i|\, \mathbf{W}\, |\omega_j\rangle = \langle\omega_i|\, \boldsymbol{\mathcal{L}}\{|\omega_j\rangle\langle\omega_j|\}\, |\omega_i\rangle \;. \tag{13}$$

Therefore Eq. (11) must be solved for with $\mathbf{W}_{[2]}$ exactly, and then the further effects of $\mathbf{W}_{[4]}$, $\mathbf{W}_{[6]}$, etc., can be incorporated via canonical perturbation theory. [Note that this is slightly more complicated than the usual canonical perturbation in the Schrödinger equation where one knows the Hamiltonian perturbation exactly.] The eigenvalues obtained in diagonalizing $\mathbf{W}_{[2]}$ give the second-order corrections $f^{[2]}$ to the eigenvalues of $\boldsymbol{\mathcal{L}}$ and the correct zeroth-order eigenoperators $\mathbf{o}^{[0]}$ for the degenerate subspace. Degenerate perturbation theory tells us that in order to calculate each $\vec{\mathbf{o}}_i^{[2]}$ for the degenerate subspace one actually requires $\mathbf{W}_{[4]}$ from the fourth-order master equation; it will contribute the second-order correction

$$\sum_{j\neq i} \frac{\left(\vec{\mathbf{o}}_j^{[0]}\right)^\star \mathbf{W}_{[4]} \left(\vec{\mathbf{o}}_i^{[0]}\right)}{f_i^{[2]} - f_j^{[2]}} \vec{\mathbf{o}}_j^{[0]} \,, \tag{14}$$

where $\vec{\mathbf{o}}_i^\star$ is the left eigen-vector of $\mathbf{W}$ such that $\vec{\mathbf{o}}_i^\star \mathbf{W} = \vec{\mathbf{o}}_i^\star f_i$ and $\vec{\mathbf{o}}_j^\star \vec{\mathbf{o}}_i = \delta_{ij}$. Such corrections would be fourth order in a non-degenerate problem, but because the free Liouvillian is always degenerate, they become second order as the relevant lowest-order nonvanishing eigenvalue splitting is always second order here. Without this information from the fourth-order master equation, one cannot generate the complete second-order solution.

Finally note that this requirement must extend even to exact solutions of the perturbative master equation. A perturbative solution to the second-order master equation will be equivalent to solving the full master equation perturbatively and then artificially setting $\boldsymbol{\mathcal{L}}_{[4]}$ and all higher-order contributions to the Liouvillian to vanish. From this we know that these two solutions must differ by a term that is $\mathcal{O}(c^2)$. Since the exact solutions to the perturbative and full master equations each differ from the corresponding perturbative solutions by terms of $\mathcal{O}(c^4)$, we can conclude from our analysis that even the exact solution to the second-order master equation differs from the exact solution to full master equation by a term of $\mathcal{O}(c^2)$. In next section we use the example of quantum Brownian motion, where an exact solution is available, to show that the second-order corrections arising from the fourth-order Liouvillian are indeed present.

More generally, while the short-time accuracy of an order-$2n$ master equation can also be order $2n$, the long-time accuracy can only be order $2n-2$. To obtain order-$2n$ solutions one requires not only the order-$2n$ master equation but in addition the order-$(2n+2)$ Pauli master equation. In particular, the second-order master equation after taking the *rotating-wave approximation* [6] will contain just enough terms to generate solutions which are accurate to zeroth-order [13]. The full second-order master equation improves upon this but not enough to generate the full second-order solutions.

Among the information missing due to the second-order errors of the solution to the second-order master equation are important contributions to the asymptotic state of the system. When coupled to a thermal reservoir the system must asymptote to $\boldsymbol{\rho} \propto e^{-\beta\,\mathbf{H}}$ for vanishing system-environment coupling. One often desires to find the additional environmentally induced system-system correlations (and possibly entanglement) provided by perturbative corrections, but these will not be given correctly by directly finding the steady state of the second-order master equation. However, at least for zero-temperature noise, it is still possible to easily construct via other methods the order-$2n$ corrections using only order-$2n$ master equation coefficients [10, 14].

Another important characteristic that is mangled by the second-order master equation is positivity, as was mentioned in the introduction. The second-order inaccuracies that arise from using the second-order master equation imply that the diagonal elements of the density matrix in the (free) energy basis are off by second-order terms. This can lead to second-order violations of positivity.

EXAMPLE: QBM

As an example of an exactly solvable open system, let us consider the master equation of an oscillator bilinearly coupled to an environment of oscillators initially in a thermal state [15]:

$$\frac{d}{dt}\boldsymbol{\rho} = [-\imath\,\mathbf{H}_\mathrm{R}, \boldsymbol{\rho}] - \imath\,\Gamma\,[\mathbf{x}, \{\mathbf{p}, \boldsymbol{\rho}\}] - M D_{pp}\,[\mathbf{x}, [\mathbf{x}, \boldsymbol{\rho}]] - D_{xp}\,[\mathbf{x}, [\mathbf{p}, \boldsymbol{\rho}]] \,, \tag{15}$$

where $\mathbf{H}_\mathrm{R}$ is the system Hamiltonian but with renormalized frequency Ω_R, Γ is the dissipation coefficient, D_{pp} and D_{xp} are the regular and anomalous diffusion coefficients. In Ref. [11] exact solutions are given. In the stationary limit, the system relaxes into a Gaussian state with phase-space covariance

$$\boldsymbol{\sigma}_T = \begin{bmatrix} \frac{1}{M\Omega_\mathrm{R}^2}\left(\frac{1}{2\Gamma}D_{pp} - D_{xp}\right) & 0 \\ 0 & \frac{M}{2\Gamma}D_{pp} \end{bmatrix} . \tag{16}$$

One can see that for a second-order master equation the contribution from the regular diffusion D_{pp}/Γ starts at zeroth order while the contribution from anomalous diffusion D_{xp} starts at second order. The full second-order contribution from the regular diffusion requires the fourth-order coefficients.

It so happens that the regular diffusion coefficient is well behaved at second order and high-frequency sensitive (in terms of the largest bath modes, e.g. the cutoff) at fourth order. The anomalous diffusion coefficient is proportionally high-frequency sensitive at second order and so any exact solution should have a position uncertainty which is insensitive to high frequencies. Therefore solutions to the second-order master equation can produce a position uncertainty with (negative) high-frequency sensitivity at second order. Such a solution could potentially violate the Heisenberg uncertainty principle and even classical probability if the cutoff is large enough.

DISCUSSION

We have shown that even when provided with a stationary master equation describing dynamics that are amenable to perturbative solution, the solutions to an order-$2n$ perturbative master equation are, in general, only accurate to order-$(2n-2)$, a step down from that of the master equation itself. This has a wide-range of implications upon the common use of second-order master equations and related master equations derived from second-order dynamics: the Redfield, Born-Markov, and many Lindblad equations. Moreover, not even a nonlocal representation, such as with the Nakajima-Zwanzig master equation can avoid this effect, as a thorough analysis of time-local and nonlocal dynamics shows their asymptotics to be perturbatively the same [10].

To be more specific, the second-order master equation can provide all second-order timescales and off-diagonal density matrix elements (in the free energy basis). However it can only provide the diagonal matrix elements with zeroth-order accuracy, and the missing information is the most relevant to positivity in the low-temperature regime. Therefore the second-order master equation can produce second-order positivity violations, whereas the full second-order solutions are positive to second-order. Likewise, the steady state of the second-order master equation may only agree with the steady state of the full master equation to zeroth order.

There are three mathematical limits in which the second-order master equation will give solutions accurate to second order: The first is early times, where t is small compared to any of the second-order damping time scales. The second is the Markovian limit, because in this limit the second-order master equation is exact. The third is the limit employed by Davies [16] where one rewrites the master equation in terms of the rescaled time parameter $\tau = c^2 t$ and then takes the limit $c \to 0$ (for $\tau \neq 0$ this effectively amounts to taking a simultaneous $t \to \infty$ limit). In this limit all corrections to the eigenoperators of the Liouvillian vanish, and the only effect of the environment is to introduce damping rates through corrections to the eigenvalues, which are correctly captured by the perturbative master equation. Thus, the inaccuracies of second-order master equation we have addressed may be sufficiently suppressed even at late times if a physical system is sufficiently close to being described by one of these limits. Therefore our results should be most heeded in the non-Markovian regime of low temperature or long-ranged correlations and with non-vanishing dissipation rates.

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