Canonically Consistent Quantum Master Equation

Tobias Becker,[1,*] Alexander Schnell,[1,†,§] and Juzar Thingna[2,3,‡,§]
[1]*Institut für Theoretische Physik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany*
[2]*Department of Physics and Applied Physics, University of Massachusetts, Lowell, Massachusetts 01854, USA*
[3]*Center for Theoretical Physics of Complex Systems, Institute for Basic Science (IBS), Daejeon 34126, Republic of Korea*

(Received 3 June 2022; revised 1 August 2022; accepted 11 October 2022; published 8 November 2022)

We put forth a new class of quantum master equations that correctly reproduce the asymptotic state of an open quantum system beyond the infinitesimally weak system-bath coupling limit. Our method is based on incorporating the knowledge of the reduced steady state into its dynamics. The correction not only steers the reduced system toward a correct steady state but also improves the accuracy of the dynamics, thereby refining the archetypal Born-Markov weak-coupling second-order master equations. In case of equilibrium, we use the exact mean-force Gibbs state to correct the Redfield quantum master equation. By benchmarking it with the exact solution of the damped harmonic oscillator, we show that our method also helps correct the long-standing issue of positivity violation, albeit without complete positivity. Our method of a canonically consistent quantum master equation opens a new perspective in the theory of open quantum systems leading to a reduced density matrix accurate beyond the commonly used Redfield and Lindblad equations, while retaining the same conceptual and numerical complexity.

DOI: 10.1103/PhysRevLett.129.200403

Introduction.—The central problem of the theory of open quantum systems is to describe the dynamics of a quantum system in contact with a reservoir [1–3]. Most applications describe the dynamics using a weak-coupling quantum master equation (QME) such as Lindblad [1,4,5] or Redfield [1,3,6] that are valid under a stringent set of assumptions. Going beyond this standard weak-coupling approach is tedious and despite formal equations available for the past 45 years [7–10], there exist only a handful of model-independent practical methods that go beyond weak coupling [11–28]. However, these approaches are generally complex and not easily implementable especially when dealing with many-body quantum systems.

Hence, most studies are restricted to equations that are second order in the system-bath coupling. While being simple, such second-order equations are not devoid of issues. For example, the least approximative Redfield equation can lead to unphysical negative populations [26,29–33]. The quantum optical master equation (also known as the Lindblad equation) gives an equilibrium reduced density matrix that for a wide class of models is independent of the system-bath coupling strength [34] contrary to the notion of the Hamiltonian of mean force [35,36]. Moreover, any second-order master equation has issues with its accuracy such that the inaccuracies develop over time leading to a wrong steady state [37,38].

In this Letter, we use the asymptotic state of the system to develop a QME that goes beyond these common weak coupling approximations. A similar approach was highly successful to improve on the conventional classical Fokker-Planck equation [39], but its quantum counterpart is still missing. We address this gap and develop a fully quantum formulation that uses the asymptotic state to correctly steer the transient dynamics. In case of a quantum system connected to a single reservoir, we use the generalized canonical distribution also known as the mean-force Gibbs state [40] [see Eq. (5)] to correct for the transient dynamics and corroborate our findings with the exactly solvable quantum dissipative harmonic oscillator. The generalized canonical distribution incorporates effects of finite system-reservoir coupling giving a solution that also *correctly* captures finite-coupling effects. This *canonically consistent quantum master equation* (CCQME) is as universal and versatile as standard weak-coupling QMEs, making it applicable to a wide range of scenarios ranging from quantum optics, chemical physics, statistical physics, and more recently quantum information and -thermodynamics.

A key feature of our approach is its simplicity. Even though we obtain solutions that are accurate beyond weak coupling we do not require any additional information than what is needed in a weak-coupling Redfield equation. In other words, we do not require cumbersome fourth-order tensors involving multidimensional integrals [16] or elaborate numerical methods [11,13,17,18,41] that restrict treatable Hilbert space dimensions. We even find accurate agreement with the numerically exact hierarchy equation of motion approach and demonstrate our methods applicability for an interacting many-body open quantum system [42]. Moreover, using recent advances in evaluating the asymptotic state of nonequilibrium quantum systems driven by multiple reservoirs [43] or an external drive [44,45], our approach is easily extendable to study the dynamics of

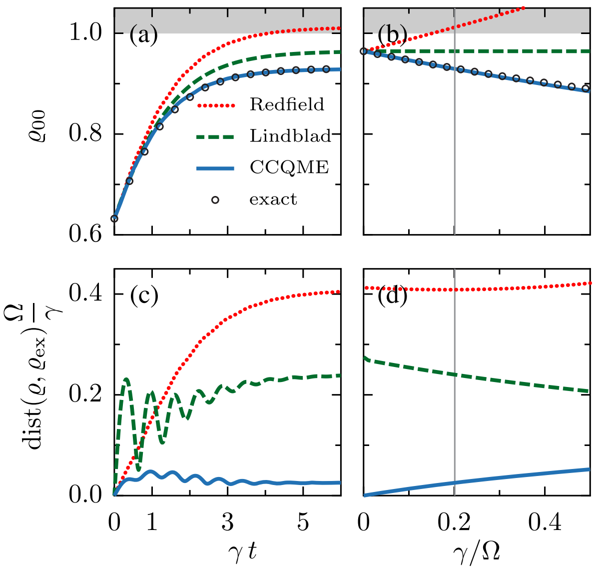

FIG. 1. Comparison between the Redfield (dotted red), Lindblad (dashed green), CCQME (solid blue), and the nonperturbative exact solution (open circles) for a harmonic oscillator system with frequency Ω coupled to a thermal bath with strength γ. The ground-state population dynamics are shown in (a), while (b) depicts the steady-state populations. The gray regions in (a) and (b) indicate unphysical regimes where the ground-state populations exceed 1. Deviation of the density matrix to the exact result characterized by the trace distance $\mathrm{dist}(\varrho, \varrho_{\mathrm{ex}})$ is shown for the dynamics in (c) and steady state in (d). The system is initiated in $\varrho(0) = \exp[-H_S/\Omega]/Z_S$, and we truncate to the $N = 60$ lowest levels. The bath temperature is $T/\Omega = 0.3$ and has an Ohmic Lorentz-Drude spectral density, $J(\omega) = \gamma\omega/(1 + \omega^2/\omega_D^2)$ with cutoff $\omega_D/\Omega = 5$ and strength $\gamma/\Omega = 0.2$ [(a) and (c)] marked by a gray vertical line in (b) and (d).

externally or boundary driven quantum systems which we show in the Supplemental Material [42] for the damped harmonic oscillator driven by two baths.

Preliminaries.—We consider an autonomous system of interest H_S coupled to a thermal bath H_B such that the composite Hamiltonian reads as $H_{\mathrm{tot}} = H_0 + H_{\mathrm{int}}$ with $H_0 = H_S + H_B$. The system couples to the reservoir via a general interaction $H_{\mathrm{int}} = S \otimes B$ with S (B) being any Hermitian operator acting in the system (bath) Hilbert space. This is assumed to keep notation light; the generalization of our results to interactions that are a sum of such direct products is straightforward. Throughout this Letter we work in units where $\hbar = k_B = 1$. The composite system evolves unitarily, and we are interested in the dynamics of the reduced density matrix $\varrho(t) = \mathrm{tr}_B(\varrho_{\mathrm{tot}}(t)) \equiv \Lambda_t[\varrho(0)]$. Under weak-coupling and Born approximations using decoupled initial conditions the dynamical map is given by [1] (see also the Supplemental Material [42])

$$\Lambda_t[\cdot] \simeq \Lambda_t^0\left[\mathbb{I}[\cdot] + \int_0^t d\tau \Lambda_{-\tau}^0[\mathcal{R}_\tau[\Lambda_\tau^0[\cdot]]]\right]. \quad (1)$$

Above, $\Lambda_t^0[\cdot] = \exp[-iH_S t] \cdot \exp[iH_S t]$ is the noninteracting evolution superoperator, $\mathbb{I}[X] = X$ the identity superoperator, and $\mathcal{R}_t$ the time-dependent Redfield superoperator [6] given by $\mathcal{R}_t[\cdot] = \int_0^t d\tau([\tilde{S}(-\tau)\cdot, S]C(\tau) + \mathrm{H.c.})$, where $C(t) = \mathrm{Tr}_B(\exp[iH_B t]B\exp[-iH_B t]B\rho_B)$ is the two-point bath correlator with $\rho_B = \exp[-\beta H_B]/Z_B$ being the initial density matrix of the bath and $\tilde{S}(t) = \Lambda_{-t}^0[S]$. To obtain Eq. (1) one uses the Dyson expansion for the unitary time-evolution operator of the composite system to obtain a perturbative series in H_{int}. Any truncation of this series leads to divergences with respect to time such that at second order the map Λ_t diverges linearly in time [15,38] (see also the Supplemental Material [42]).

Alternatively, in the standard open quantum systems framework one avoids the map and its divergence by taking the time derivative of Eq. (1) to obtain a first-order differential equation,

$$\partial_t\varrho(t) = -i[H_S, \varrho(t)] + \mathcal{R}_t[\Lambda_t^0[\varrho(0)]], \quad (2)$$

which precedes the Redfield equation since the dissipator $\mathcal{R}_t$ acts on the initial state $\varrho(0)$. To second order (Born approximation) in H_{int}, one often replaces

$$\Lambda_t^0[\varrho(0)] \approx \varrho(t). \quad (3)$$

As we see from Eq. (1), the above substitution results in errors at a higher order in system-bath coupling leaving the differential equation correct up to second order, as desired. The resulting QME is known as the time-dependent Redfield equation [6] that violates complete positivity (CP) and hence is less preferred over the (secular) Lindblad equation [1,4,5,42]. Despite the lack of CP, a stringent restriction for physical maps [31,46], the Redfield equation is able to capture finite system-bath coupling effects for which the Lindblad equation is insensitive [26,47]. Thus, it is only recently that the Redfield equation has gained popularity as a tool to incorporate finite system-bath coupling effects [34,43,44,48–50]. In Fig. 1 we demonstrate this for a dissipative harmonic oscillator, described in detail later in the Letter. The Redfield equation violates positivity in Figs. 1(a) and 1(b), and errors build up over time such that the steady state shows finite errors in second order in H_{int} [see Fig. 1(d)]. To resolve these issues, we propose below a scheme that uses the mean-force Gibbs state from equilibrium statistical mechanics [35,36,40] to correct the Redfield equation, specifically improving on the approximation in Eq. (3). Our approach, despite not being CP, avoids unphysical negative populations for finite coupling strengths (see Fig. 1) and ensures that the equilibrium state is mean-force Gibbs [35,36,40] up to second order in system-bath coupling.

Canonically consistent quantum master equation (CCQME).—To start with, we assume Q_t to be the superoperator that generates the second-order contributions for the time evolution

$$\varrho(t) \simeq (\mathbb{I} + Q_t)[\Lambda_t^0[\varrho(0)]]. \tag{4}$$

To find the Markovian master equation generated by this dynamical map we take the formal inverse $(\mathbb{I} + Q_t)^{-1} \simeq (\mathbb{I} - Q_t)$ and use this relation to replace the freely evolving state on the right hand side of Eq. (2). We obtain the QME $\partial_t\varrho(t) = -i[H_S, \varrho(t)] + \mathcal{R}_t[(\mathbb{I} - Q_t)[\varrho(t)]]$, which involves a fourth-order correction to the Redfield equation.

Instead of following pathological perturbative expansions for the dynamical map that involve divergences in each order separately, we make a proposal that can be viewed to combine notions of quantum mechanics and statistical mechanics in a holistic way. While quantum mechanics is a dynamical theory for the microscopic degrees of freedom, statistical mechanics describes systems in equilibrium by very few parameters only. Profound mechanisms have been developed to connect both fields, such as eigenstate thermalization hypothesis [51], canonical- [52], and dynamical [53] typicality. For finite coupling, we assume that once the system is coupled to the reservoir the system eventually relaxes to the mean force Gibbs distribution

$$\lim_{t\to\infty}\varrho(t) = \frac{\mathrm{Tr}_B(e^{-\beta H_{\mathrm{tot}}})}{Z_{\mathrm{tot}}} \simeq (\mathbb{I} + \bar{Q})[\varrho_G], \tag{5}$$

with $\varrho_G = \exp[-\beta H_S]/Z_S$ being the canonical Gibbs state obtained in the infinitesimally weak-coupling long-time limit of $\varrho(t)$. In particular, for finite coupling, the reduced state of the system deviates from the canonical Gibbs distribution, and the lowest-order correction $\bar{Q}[\varrho_G]$ is second order in system-bath coupling.

Based on the similarities between the dynamical map, Eq. (4), and the equilibrium state, Eq. (5), it becomes evident that Q_∞ can be replaced with $\bar{Q}$. Note here that the superoperators Q_∞ and $\bar{Q}$ are not strictly equivalent. The difference between them stems from the order in which the long-time and weak-coupling limits are performed. In equilibrium statistical mechanics, which leads to the mean-force Gibbs state, we take the infinite-time limit first, followed by the weak-coupling limit to obtain a nondivergent $\bar{Q}$, whereas within the quantum framework we take the weak-coupling limit first and the infinite-time limit later to obtain Q_∞ that is divergent.

In problems such as those tackled in this Letter, a sensible (convergent) answer is given when the order of the limits is dictated by statistical mechanics rather than quantum dynamics. Therefore, we replace Q_t with its long-time version given by statistical mechanics, $\bar{Q}$. This gives our main result, the CCQME

$$\partial_t\varrho(t) = -i[H_S, \varrho(t)] + \mathcal{R}_t[(\mathbb{I} - \bar{Q})[\varrho(t)]]. \tag{6}$$

It goes beyond standard second-order treatments and is consistent with statistical mechanics. Note the following subtlety: Since Eq. (5) only fixes the action of the superoperator $\bar{Q}$ on the Gibbs state, the action of $\bar{Q}$ on all other states is in principle undetermined. Different CCQMEs are, thus, possible that are expected to perform equally well for the steady state; however the transient dynamics depends on the particular choice.

By construction it is straightforward to prove [using Eq. (5) in Eq. (6)] that the steady-state solution of the CCQME is given by the mean-force Gibbs state,

$$0 = -i\Delta[\bar{Q}[\varrho_G]] + \mathcal{R}_\infty\Big[\underbrace{(\mathbb{I} - \bar{Q})[(\mathbb{I} + \bar{Q})}_{\simeq\mathbb{I}}[\varrho_G]]\Big], \tag{7}$$

with $\Delta[\cdot] = [H_S, \cdot]$, and terms in the underbrace are approximated to the identity superoperator by ignoring sixth-order system-bath coupling strength contributions. The above equation yields the necessary condition $i\Delta[\bar{Q}[\cdot]] = \mathcal{R}_\infty[\cdot]$, which we later show to be valid. The CCQME does not need to be completely positive but relaxes toward the positive mean-force Gibbs state. Furthermore, we observe for the dissipative harmonic oscillator (see below) that the CCQME violates the positivity of the density matrix when the strength γ is greater than the frequency Ω of the central oscillator, improving the validity regime as compared to weak-coupling approaches [26,54] (see the Supplemental Material for more details [42]).

Using canonical perturbation theory [34], we show that one choice for the superoperator $\bar{Q}$ can be expressed as (see the Supplemental Material [42])

$$\bar{Q}[\varrho] = \mathcal{P}\frac{1}{i\Delta}\{\mathcal{R}_\infty[\varrho]\} + \mathcal{P}^c\left[\sum_{nl}\left[\mathcal{L}(L_{nl})[\varrho] + |S_{nl}|^2 W''_{ln}\frac{\partial\varrho}{\partial E_n}\right]\right]. \tag{8}$$

Above $\mathcal{P} = \sum_{n\neq m}\mathcal{P}_{nm}$ is the projector into the coherent subspace with $\mathcal{P}_{nm} = |n\rangle\langle n|\cdot|m\rangle\langle m|$. Moreover, $|n\rangle$ (E_n) are the eigenstates (nondegenerate eigenenergies) of H_S so that the superoperator Δ is invertible in this subspace, and $\mathcal{P}^c = \sum_n \mathcal{P}_{nn}$ is the projector into the complimentary subspace of $\mathcal{P}$. Acting with the superoperator $i\Delta$ on Eq. (8), the second term vanishes due to the commutator in the superoperator Δ, and the first term yields $i\Delta[\bar{Q}[\cdot]] = \mathcal{R}_\infty[\cdot]$, which satisfies the necessary condition for the steady-state equation above. In the complementary subspace for which $\mathcal{P}^c$ projects to the populations we find a Lindblad contribution $\mathcal{L}(L) = L\cdot L^\dagger - \frac{1}{2}\{L^\dagger L, \cdot\}$

with jump operators $L_{nl} = \sqrt{|S_{nl}|^2 V''_{nl}} |n\rangle\langle l|$, with V''_{nl} being the imaginary part of $V(E_n - E_l) = V'_{nl} + iV''_{nl}$. Here $V(E) = \partial W(E)/\partial E$ is defined using the Fourier-Laplace transform of the bath correlator $W(E) = \int_0^\infty dt\, C(t) \exp[-iEt]$ used in the Redfield equation. The function $W(E_n - E_l) = W'_{nl} + iW''_{nl}$ contains real and imaginary parts typically referred to as rates and lamb shifts, respectively. On the other hand the last term in Eq. (8) can be obtained as [42]

$$\frac{\partial \varrho}{\partial E_n} = \frac{\sum_{l\neq n} |S_{nl}|^2 [V'_{nl} p_l + V'_{ln} p_n]}{\sum_{l\neq n} |S_{ln}|^2 W'_{ln}} |n\rangle\langle n|, \qquad (9)$$

where $p_n = \langle n|\varrho|n\rangle$ are the populations. For the canonical Gibbs state this term simplifies to $\partial \varrho_G/\partial E_n = -\beta \mathcal{P}_{nn}[\varrho_G]$.

Benchmark with exact dynamics.—We corroborate our method with the exactly solvable dynamics for the damped harmonic oscillator whose total system-bath Hamiltonian [55] reads as $H_{\mathrm{tot}} = H_{\mathrm{S}} + \sum_k^\infty [p_k^2/(2m_k) + m_k\omega_k^2 (q_k - c_k m_k^{-1} \omega_k^{-2} q)^2/2]$. In units where the particle mass is set to 1 we have $H_{\mathrm{S}} = p^2/2 + \Omega^2 q^2/2$. For a bath in thermal equilibrium that is factorized initially from the state of the oscillator one obtains the exact (nonperturbative in γ) QME [56–60],

$$\partial_t \varrho_{\mathrm{ex}}(t) = -\frac{i}{2}[p^2 + \gamma_q(t) q^2, \varrho_{\mathrm{ex}}(t)] - D_p(t)\{q, [q, \varrho_{\mathrm{ex}}(t)]\} - \frac{i}{2}\gamma_p(t)[q, \{p, \varrho_{\mathrm{ex}}(t)\}] + D_q(t)\{q, [p, \varrho_{\mathrm{ex}}(t)]\}.$$

Here the damping coefficient $\gamma_p(t)$ is derived from the system correlation, and the diffusive coefficients $D_q(t)$ and $D_p(t)$ depend on the bath correlation. A detailed discussion of the parameters can be found in Refs. [42,56–58].

In Fig. 1 we benchmark the dynamics [(a) and (c)] and the steady state [(b) and (d)] obtained via the Redfield equation, the Lindblad equation, and the CCQME with the exact solution. For all simulations we assume that the bath correlations decay faster than the timescale of the dynamics leading to an autonomous generator (see Ref. [42] for the spin-boson model solved with the full time-dependent generator). For strong coupling $\gamma/\Omega \simeq 0.2$ and low bath temperature $T/\Omega = 0.3$ we observe a breakdown of the Redfield theory as the ground-state population exceeds 1 in Fig. 1(a). We quantify the deviation from the exact result, i.e., error, with the trace distance $\mathrm{dist}(\varrho, \varrho_{\mathrm{ex}}) = (1/2)\mathrm{Tr}\{\sqrt{[\varrho(t) - \varrho_{\mathrm{ex}}(t)]^2}\}$, a metric bounded by 1. At small temperatures the trace distance is nearly equivalent to the percentage error in the ground-state population, i.e., the maximum trace distance of 0.08 (for $\gamma = 0.2\ \Omega$) for the Redfield case in Fig. 1(c) represents a maximum error of ≈8%. The Lindblad equation shows large oscillations in the trace distance (≈5%) but does not suffer from unphysical solutions. The CCQME reproduces the ground-state population for all times in Fig. 1(a) and shows small deviations ($< 1\% \ \forall \ t$) from the exact solution in Fig. 1(c) improving upon the Redfield and Lindblad solutions in the strong-coupling and low-temperature regime.

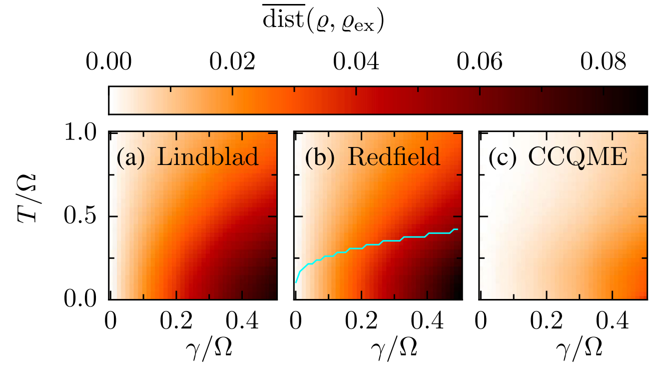

FIG. 2. Time-averaged trace distance $[\overline{\mathrm{dist}}(\varrho, \varrho_{\mathrm{ex}}) = \tau_R^{-1} \int_0^{\tau_R} dt\, \mathrm{dist}(\varrho, \varrho_{\mathrm{ex}})]$ over relaxation time $\tau_R = 2/\gamma$ as a function of temperature and coupling strength for the Lindblad equation (a), Redfield equation (b), and CCQME (c). The Redfield steady-state solution violates positivity in the parameter regime below the solid teal line. Other parameters are the same as Fig. 1.

In equilibrium, finite coupling leads to an effective higher temperature such that the ground-state population decreases as a function of the coupling strength. In Fig. 1(b) the CCQME accurately reproduces this trend and also perfectly matches with the exact curve in the strong coupling regime. On the one hand, the Lindblad solution remains independent of the system-bath coupling γ. On the other hand, the Redfield not only gives an incorrect ground-state population but also erroneously predicts that as the system-bath coupling strength increases the system cools down. At moderate values of the coupling, the ground-state population $\varrho_{00} > 1$ implying that the excited states have unphysical negative populations. Since both the exact steady-state solution and the approximate ones from QMEs contain all powers of γ, we would like to quantify how much the QMEs deviate from the exact solution at second order. This deviation in the solution can be quantified by dividing the trace distance by the coupling strength and numerically approaching the limit of $\gamma/\Omega \to 0$ [see Fig. 1(d)]. If the QME and the exact solutions match at second-order $O(\gamma)$, this measure approaches zero as $\gamma/\Omega \to 0$ as seen for the CCQME [solid blue line in Fig. 1(b)]. The other QMEs give a finite deviation indicating discrepancies at $O(\gamma)$.

To get a complete overview of the deviations even in the dynamics for the entire parameter space, in Fig. 2 we calculate the time-averaged trace distance to the exact result for the entire relaxation process as a function temperature and coupling strength initiating the system in an out-of-equilibrium initial state. For strong coupling the Lindblad and Redfield equations are only valid for high bath temperatures, i.e., deep classical limit. Contrastingly, the CCQME leads to a more accurate result in the full parameter regime. Importantly, the CCQME solution gives positive density matrices in the entire parameter range in sharp contrast to the Redfield that fails at low temperatures [below solid teal line in Fig. 2(b)].

Quantum transport.—The CCQME can also be implemented for transport setups [61–65] wherein the system is driven by several independent baths $H_B = \sum_i H_B^i$ that couple via the interaction Hamiltonian $H_{\mathrm{int}} = \sum_i S^i \otimes B^i$. At second order of the interaction, the Redfield and Lindblad equation is obtained by adding the dissipative superoperators for the individual baths. However beyond second order cross correlations build up [14,16], which occur naturally in the boundary driven CCQME

$$\partial_t \varrho(t) = -i[H_S, \varrho(t)] + \sum_i \mathcal{R}_t^i \left[\left(\mathbb{I} - \sum_j \bar{Q}^j \right) [\varrho(t)] \right], \quad (10)$$

by the products of superoperators for different baths. We elucidate this idea further by studying a boundary driven harmonic oscillator and corroborating the CCQME with exact results in the Supplemental Material [42].

Summary.–In this Letter, we proposed a QME that corrects the standard Born-Markov equations (Redfield or Lindblad) incorporating effects of higher order system-bath coupling. The CCQME draws inspiration from the statistical mean-force Gibbs state and correctly steers the dynamics of a quantum system coupled with finite strength to a reservoir. By construction it yields the exact equilibrium state and significantly improves the dynamics as compared to the Redfield or Lindblad equation. Here we focus on a consistent second-order theory that should be generalizable to higher order [42]. Despite not being completely positive our approach does not suffer from negative solutions. The CCQME is not only accurate but also easy to implement since it requires no additional information as compared to the Redfield equation. Moreover it is model independent and could shed light onto finding lowest order effects due to the presence of system-bath coupling in the emerging field of strong-coupling quantum thermodynamics [35,36,66–68] or in dissipative quantum many-body systems as demonstrated in the Supplemental Material using an Ising chain [42]. The CCQME could also be extended to study transport through systems strongly connected to multiple reservoirs aiding the field of strong-coupling quantum transport [24,69–71].

We thank Peter Hänggi, Peter Talkner, Walter Strunz, André Eckardt, and Gernot Schaller for helpful discussions. This research was supported by the Institute for Basic Science in South Korea (IBS-R024-Y2) and by the German Research Foundation (DFG) via the Collaborative Research Center SFB910 under Project No. 163436311.

[*]tobias.becker@tu-berlin.de
[†]schnell@tu-berlin.de
[‡]juzar_thingna@uml.edu
[§]A. S. and J. T. contributed equally to this work.

[1] H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Oxford University Press, New York, 2007).
[2] U. Weiss, *Quantum Dissipative Systems* (World Scientific, Singapore, 2012), 10.1142/8334.
[3] H. J. Carmichael, *Statistical Methods in Quantum Optics 1* (Springer-Verlag, Berlin, 1999), 10.1007/978-3-662-03875-8.
[4] G. Lindblad, On the generators of quantum dynamical semigroups, Commun. Math. Phys. **48**, 119 (1976).
[5] V. Gorini, A. Kossakowski, and E. C. G. Sudarshan, Completely positive dynamical semigroups of n-level systems, J. Math. Phys. (N.Y.) **17**, 821 (1976).
[6] A. G. Redfield, The theory of relaxation processes, Adv. Magn. Reson. **1**, 1 (1965).
[7] S. Nakajima, On quantum theory of transport phenomena: Steady diffusion, Prog. Theor. Phys. **20**, 948 (1958).
[8] R. Zwanzig, Ensemble method in the theory of irreversibility, J. Chem. Phys. **33**, 1338 (1960).
[9] A. Fulinski and W. Kramarczyk, On the exact master equations, Physica (Utrecht) **39**, 575 (1968).
[10] F. Shibata, Y. Takahashi, and N. Hashitsume, A generalized stochastic liouville equation. non-markovian versus memoryless master equations, J. Stat. Phys. **17**, 171 (1977).
[11] Y. Tanimura and R. Kubo, Time evolution of a quantum system in contact with a nearly Gaussian-Markoffian noise bath, J. Phys. Soc. Jpn. **58**, 101 (1989).
[12] B. B. Laird, J. Budimir, and J. L. Skinner, Quantum-mechanical derivation of the Bloch equations: Beyond the weak-coupling limit, J. Chem. Phys. **94**, 4391 (1991).
[13] N. Makri and D. E. Makarov, Tensor propagator for iterative quantum time evolution of reduced density matrices. I. Theory, J. Chem. Phys. **102**, 4600 (1995).
[14] S. Jang, J. Cao, and R. J. Silbey, Fourth-order quantum master equation and its Markovian bath limit, J. Chem. Phys. **116**, 2705 (2002).
[15] C. Timm, Time-convolutionless master equation for quantum dots: Perturbative expansion to arbitrary order, Phys. Rev. B **83**, 115416 (2011).
[16] J. Thingna, H. Zhou, and J.-S. Wang, Improved Dyson series expansion for steady-state quantum transport beyond the weak coupling limit: Divergences and resolution, J. Chem. Phys. **141**, 194101 (2014).
[17] J. Iles-Smith, N. Lambert, and A. Nazir, Environmental dynamics, correlations, and the emergence of noncanonical equilibrium states in open quantum systems, Phys. Rev. A **90**, 032114 (2014).
[18] J. Cerrillo and J. Cao, Non-Markovian Dynamical Maps: Numerical Processing of Open Quantum Trajectories, Phys. Rev. Lett. **112**, 110401 (2014).
[19] S. Alipour, A. T. Rezakhani, A. P. Babu, K. Mølmer, M. Möttönen, and T. Ala-Nissila, Correlation-Picture Approach

to Open-Quantum-System Dynamics, Phys. Rev. X **10**, 041024 (2020).

[20] A. Purkayastha, G. Guarnieri, M. T. Mitchison, R. Filip, and J. Goold, Tunable phonon-induced steady-state coherence in a double-quantum-dot charge qubit, npj Quantum Inf. **6**, 27 (2020).

[21] A. Imamoḡlu, Stochastic wave-function approach to non-Markovian systems, Phys. Rev. A **50**, 3650 (1994).

[22] B. M. Garraway, Nonperturbative decay of an atomic system in a cavity, Phys. Rev. A **55**, 2290 (1997).

[23] L. Mazzola, S. Maniscalco, J. Piilo, K.-A. Suominen, and B. M. Garraway, Sudden death and sudden birth of entanglement in common structured reservoirs, Phys. Rev. A **79**, 042302 (2009).

[24] N. Anto-Sztrikacs and D. Segal, Strong coupling effects in quantum thermal transport with the reaction coordinate method, New J. Phys. **23**, 063036 (2021).

[25] A. Nazir and G. Schaller, The reaction coordinate mapping in quantum thermodynamics, in *Thermodynamics in the Quantum Regime: Fundamental Aspects and New Directions*, edited by F. Binder, L. A. Correa, C. Gogolin, J. Anders, and G. Adesso (Springer International Publishing, Cham, 2018), pp. 551–577.

[26] R. Hartmann and W. T. Strunz, Accuracy assessment of perturbative master equations: Embracing nonpositivity, Phys. Rev. A **101**, 012103 (2020).

[27] A. Trushechkin, Quantum master equations and steady states for the ultrastrong-coupling limit and the strong-decoherence limit, arXiv:2109.01888.

[28] F. Cerisola, M. Berritta, S. Scali, S. A. R. Horsley, J. D. Cresser, and J. Anders, Quantum-classical correspondence in spin-boson equilibrium states at arbitrary coupling, arXiv:2204.10874.

[29] V. Romero-Rochin and I. Oppenheim, Relaxation properties of two-level systems in condensed phases, Physica (Amsterdam) **155A**, 52 (1989).

[30] A. Suárez, R. Silbey, and I. Oppenheim, Memory effects in the relaxation of quantum open systems, J. Chem. Phys. **97**, 5101 (1992).

[31] P. Pechukas, Reduced Dynamics Need Not Be Completely Positive, Phys. Rev. Lett. **73**, 1060 (1994).

[32] E. Geva, E. Rosenman, and D. Tannor, On the second-order corrections to the quantum canonical equilibrium density matrix, J. Chem. Phys. **113**, 1380 (2000).

[33] A. Montoya-Castillo, T. C. Berkelbach, and D. R. Reichman, Extending the applicability of Redfield theories into highly non-Markovian regimes, J. Chem. Phys. **143**, 194108 (2015).

[34] J. Thingna, J.-S. Wang, and P. Hänggi, Generalized Gibbs state with modified Redfield solution: Exact agreement up to second order, J. Chem. Phys. **136**, 194110 (2012).

[35] C. Jarzynski, Stochastic and Macroscopic Thermodynamics of Strongly Coupled Systems, Phys. Rev. X **7**, 011008 (2017).

[36] P. Talkner and P. Hänggi, Colloquium: Statistical mechanics and thermodynamics at strong coupling: Quantum and classical, Rev. Mod. Phys. **92**, 041002 (2020).

[37] T. Mori and S. Miyashita, Dynamics of the density matrix in contact with a thermal bath and the quantum master equation, J. Phys. Soc. Jpn. **77**, 124005 (2008).

[38] C. H. Fleming and N. I. Cummings, Accuracy of perturbative master equations, Phys. Rev. E **83**, 031117 (2011).

[39] P. Hänggi, H. Grabert, P. Talkner, and H. Thomas, Bistable systems: Master equation versus Fokker-Planck modeling, Phys. Rev. A **29**, 371 (1984).

[40] A. S. Trushechkin, M. Merkli, J. D. Cresser, and J. Anders, Open quantum system dynamics and the mean force Gibbs state, AVS Quantum Sci. **4**, 012301 (2022).

[41] A. Fruchtman, N. Lambert, and E. M. Gauger, When do perturbative approaches accurately capture the dynamics of complex quantum systems?, Sci. Rep. **6**, 28204 (2016).

[42] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.129.200403 for (**A**) Dyson series expansion of the dynamical map, (**B**) Secular Lindblad master equation, (**C**) Canonical perturbation theory for the mean-force Gibbs state, (**D**) Exact master equation for damped harmonic oscillator, (**E**) CCQME regime of validity, (**F**) CCQME for a system connected to multiple baths, (**G**) Spin-boson model, and (**H**) Ising chain.

[43] J. Thingna, J.-S. Wang, and P. Hänggi, Reduced density matrix for nonequilibrium steady states: A modified redfield solution approach, Phys. Rev. E **88**, 052127 (2013).

[44] T. Shirai, J. Thingna, T. Mori, S. Denisov, P. Hänggi, and S. Miyashita, Effective Floquet-Gibbs states for dissipative quantum systems, New J. Phys. **18**, 053008 (2016).

[45] G. Engelhardt, G. Platero, and J. Cao, Discontinuities in Driven Spin-Boson Systems due to Coherent Destruction of Tunneling: Breakdown of the Floquet-Gibbs Distribution, Phys. Rev. Lett. **123**, 120602 (2019).

[46] A. Shaji and E. C. G. Sudarshan, Who's afraid of not completely positive maps?, Phys. Lett. A **341**, 48 (2005).

[47] X. Xu, J. Thingna, C. Guo, and D. Poletti, Many-body open quantum systems beyond Lindblad master equations, Phys. Rev. A **99**, 012106 (2019).

[48] D. Tupkary, A. Dhar, M. Kulkarni, and A. Purkayastha, Fundamental limitations in Lindblad descriptions of systems weakly coupled to baths, Phys. Rev. A **105**, 032208 (2022).

[49] M. Łobejko, M. Winczewski, G. Suárez, R. Alicki, and M. Horodecki, Towards reconciliation of completely positive open system dynamics with the equilibration postulate, arXiv:2204.00643.

[50] G. Timofeev and A. Trushechkin, Hamiltonian of mean force in the weak-coupling and high-temperature approximations and refined quantum master equations, arXiv:2204.00599.

[51] J. M. Deutsch, Eigenstate thermalization hypothesis, Rep. Prog. Phys. **81**, 082001 (2018).

[52] S. Goldstein, J. L. Lebowitz, R. Tumulka, and N. Zanghì, Canonical Typicality, Phys. Rev. Lett. **96**, 050403 (2006).

[53] C. Bartsch and J. Gemmer, Dynamical Typicality of Quantum Expectation Values, Phys. Rev. Lett. **102**, 110403 (2009).

[54] G. Homa, A. Csordás, M. A. Csirik, and J. Z. Bernád, Range of applicability of the Hu-Paz-Zhang master equation, Phys. Rev. A **102**, 022206 (2020).

[55] A. O. Caldeira and A. J. Leggett, Quantum tunnelling in a dissipative system, Ann. Phys. (N.Y.) **149**, 374 (1983).

[56] F. Haake and R. Reibold, Strong damping and low-temperature anomalies for the harmonic oscillator, Phys. Rev. A **32**, 2462 (1985).
[57] B. L. Hu, J. P. Paz, and Y. Zhang, Quantum brownian motion in a general environment: Exact master equation with nonlocal dissipation and colored noise, Phys. Rev. D **45**, 2843 (1992).
[58] R. Karrlein and H. Grabert, Exact time evolution and master equations for the damped harmonic oscillator, Phys. Rev. E **55**, 153 (1997).
[59] J. P. Paz, Physical origins of time asymmetry, in *Decoherence in Quantum Brownian Motion* (Cambridge University Press, Cambridge, England, 1994), Vol. 1, pp. 213–220.
[60] S. Maniscalco, F. Intravaia, J. Piilo, and A. Messina, Misbeliefs and misunderstandings about the non-markovian dynamics of a damped harmonic oscillator, J. Opt. B **6**, S98 (2004).
[61] N. Li, J. Ren, L. Wang, G. Zhang, P. Hänggi, and B. Li, Colloquium: Phononics: Manipulating heat flow with electronic analogs and beyond, Rev. Mod. Phys. **84**, 1045 (2012).
[62] M. Thoss and F. Evers, Perspective: Theory of quantum transport in molecular junctions, J. Chem. Phys. **148**, 030901 (2018).
[63] I. Knezevic and B. Novakovic, Time-dependent transport in open systems based on quantum master equations, J. Comput. Electron. **12**, 363 (2013).
[64] J. E. Elenewski, D. Gruss, and M. Zwolak, Communication: Master equations for electron transport: The limits of the markovian limit, J. Chem. Phys. **147**, 151101 (2017).
[65] D. K. Ferry, J. Weinbub, M. Nedjalkov, and S. Selberherr, A review of quantum transport in field-effect transistors, Semicond. Sci. Technol. **37**, 043001 (2022).
[66] M. Esposito, M. A. Ochoa, and M. Galperin, Nature of heat in strongly coupled open quantum systems, Phys. Rev. B **92**, 235440 (2015).
[67] P. Strasberg and M. Esposito, Stochastic thermodynamics in the strong coupling regime: An unambiguous approach based on coarse graining, Phys. Rev. E **95**, 062101 (2017).
[68] D. Newman, F. Mintert, and A. Nazir, Performance of a quantum heat engine at strong reservoir coupling, Phys. Rev. E **95**, 032139 (2017).
[69] J.-S. Wang, B. K. Agarwalla, H. Li, and J. Thingna, Nonequilibrium Green's function method for quantum thermal transport, Front. Phys. **9**, 673 (2014).
[70] N. Bergmann and M. Galperin, A Green's function perspective on the nonequilibrium thermodynamics of open quantum systems strongly coupled to baths, Eur. Phys. J. Special Topics **230**, 859 (2021).
[71] G. T. Landi, D. Poletti, and G. Schaller, Non-equilibrium boundary driven quantum systems: Models, methods and properties, arXiv:2104.14350.