Quantum master equation with dissipators regularized by thermal fluctuations

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We report an alternate formulation of the quantum master equation (QME) to describe the dynamics of a quantum system weakly coupled to a heat bath, in the presence of weak external driving. A key feature of this approach is the introduction of an explicit Hamiltonian to model the thermal fluctuations in the heat bath. We show that the resulting time coarse-grained dynamical equation for the quantum system has dissipators with a natural regulator, which emerges from an ensemble average over the fluctuations. Importantly, such regularized dissipators arise from the second-order contributions of both the external drive as well as the system-environment coupling. We show that the second-order drive terms, regularized to time-scales set by the fluctuations, result in dynamic drive-induced frequency shifts as well as drive-dependent relaxation phenomena. Considering the specific case of an ensemble of two-level systems, subjected to a linearly polarized external drive, we derive the modified Bloch Equations with such drive-dependent shift and damping terms. The resulting drive-induced frequency shifts converge to the known forms of dynamic frequency shifts, such as the Bloch-Siegert shift or the dynamic Stark shift, in appropriate limits. The Kramers-Kronig pairs of these frequency shifts - manifest as drive-dependent damping terms in the modified Bloch Equations and help explain the Redfield limit of free-induction-decay (FID) rates as well as the non-Bloch decay of Rabi oscillations in isotropic medium. Our method predicts correct orders of magnitudes of non-Bloch decay rates in isotropic medium as well as their observed temperature dependence. The QME reported here, correctly describes all known aspects of the driven-dissipative dynamics up to second-order of an open quantum system with appropriate thermal signatures and as such is expected to provide deeper insights into the study of quantum information processing on real systems.

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I. INTRODUCTION

Driven-dissipative systems form the quintessential model for a vast majority of physical phenomena. For an ensemble of quantum systems in particular, the dissipation and other nonequilibrium behavior are explained by quantum master equations (QMEs), where the systems in question are coupled to a heat bath [1–3]. A vast literature exists on the different formulations of the QME explaining different aspects of driven-dissipative dynamics that are of experimental relevance [4]. QMEs, which are common in magnetic resonance spectroscopy, involve a second-order perturbative expansion of the system-bath coupling while the external drive to the system is usually treated in the linear regime [1,5]. A parallel approach assumes that the coupling and the drive induce independent rates of variation in the system resulting in a unitary response to the drive while retaining the second-order system-bath response in the form of a Lindblad dissipator [2,3]. The Floquet-Markov QMEs, introduced by Hänggi and others, provide a third approach, which becomes important especially in the strong-driving regime [6–8]. In spite of their success, the known forms of QMEs do not adequately explain many subtle features of driven-dissipative dynamics that are observed in experiments. The existence of drive-strength-dependent relaxation rates, first proposed in a seminal work by Redfield, provides for one such example [9,10]. Following DeVoe and Brewer's confirmation of the Redfield limit of relaxation rates for optical transitions in solids, Lendi proposed a phenomenological extension of the QME to explain the same for a specific choice of the relaxation parameters [10–12]. Although the drive dependence of relaxation rates becomes more pronounced at stronger field strengths, Lendi has pointed out how the driven-dissipative dynamics in magnetic resonance systems can deviate from the predictions of Bloch equations even for small drive strengths [12]. Boscaino *et al.* have also reported the dependence of two-photon free-induction-decay (FID) rates on the drive amplitude in electron-spin resonance experiments as well as non-Bloch decay of transient nutations in spin-$1/2$ systems [13–16]. Shakhmuratov *et al.* first provided a theoretical framework to explain both these phenomena in appropriate limits by introducing drive-induced fluctuating complex fields in the system description [17]. But their approach was unable to explain the concentration dependence of the non-Bloch regime, demonstrated by Agnello *et al.* [16,17]. More recent investigations by Nellutla *et al.* and Bertaina *et al.* have shown a nonlinear dependence of the decay rate of Rabi oscillations on the drive amplitude [18,19]. McCutcheon *et al.* have shown that after an initial rotating-wave approximation (RWA) on the external drive, a variational polaron transformation of the relevant Hamiltonians can be used to derive a QME [4,20]. Their approach predicts frequency renormalization as well as damping terms with quadratic dependence on the drive strength. Such effects have previously been exemplified for the exciton Rabi oscillations in quantum dots [21,22]. The drive

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dependence of the decay rates of exciton Rabi oscillations in two-level semiconductor systems has also been explained by using a dressed density-matrix master equation in the RWA [23]. We have shown elsewhere that the experimentally measured decay rates of Rabi oscillations in an ensemble of spin-$1/2$ nuclei in an isotropic environment show a quadratic dependence on the drive amplitude [24].

On the other hand, a well-known phenomenon in all resonant spectroscopic problems is the existence of dynamic frequency shifts proportional to the square of the drive amplitude, which is not well explained by QMEs. The earliest example of such an effect pertains to the Bloch-Siegert shift, which has traditionally been derived for isolated quantum systems, i.e., a microcanonical setup, where there is no external source of dissipation [25–30]. Recently, the scientific community has seen huge interest in deriving these effects in a dissipative environment, resulting in a plethora of alternate approaches [31–33]. Of these, the counter-rotating hybridized rotating-wave (CHRW) method of Yan *et al.*, which results in a Floquet-Markov-type master equation, as well as the Keldysh perturbative approach presented by Stace and Müller, predict signatures of the Bloch-Siegert effect in the steady state [31,33]. The Krylov-Bogoliubov-Mitropolsky (KBM) method in Saiko *et al.*'s work predicts Bloch-Siegert-type frequency shifts only for a longitudinal external drive, from a master equation where the dissipator is a phenomenological construct [32].

It is evident that the drive-dependent relaxation rates in isotropic environments can only arise from higher-order effects of the external drive, since a unitary or even a first-order term cannot predict a modification of the dissipators. The Floquet-Markov approaches might serve as a probable method, especially for very strongly driven systems, but explicit evaluation of the modified dissipators for comparison with experimental results can be a formidable task [6–8]. We note that the path-integral methods [34] to study driven-dissipative dynamics usually involve efficient numerical approaches, such as the quasiadiabatic propagator path integral (QUAPI) algorithm and the path integral Monte Carlo scheme (PIMC) [35–38]. In all such efforts, the influence functional proposed by Feynman and Vernon is evaluated over a large set of stochastic trajectories [34]. Such numerical routes find higher-order contributions of drive but provide no direct construction of a QME.

Also, the form of the dynamical frequency shifts implies the existence of nonlinear drive-dependent terms that are independent of the system-bath coupling. But as noted before, the signature of such frequency shifts is usually derived from a QME in the steady-state limit, as in Müller *et al.*'s work in which the second-order terms of the drive do not have a natural regulator, i.e., a high-frequency cutoff [25,33]. It has been pointed out by Stace *et al.* that, in order to study the transient dynamics of a driven-dissipative system, one has to consider all the poles with negative real values of the higher-order terms in the Laplace domain [39]. The question naturally arises as to how such poles are defined for second-order drive contributions when no Drude-like high-frequency cutoff can be defined for these terms. At this point, it is interesting to note that Karplus and Schwinger's theory of saturation and power broadening in microwave spectroscopy introduces a density matrix that captures the effects of random collisions [40]. Also, the commonly adopted theories explain the drive dependence of relaxation rates, either by the introduction of fluctuations in the energy levels of each member of a quantum ensemble induced by their local environments, or the assumption of a drive-induced additional noise term [17,41–44]. Clearly, if the fluctuations are introduced in the local environments constituting a thermal bath, which is not unphysical, the bath density matrix can capture the correlation of such fluctuations in all the second-order terms. As such, the second-order drive contributions will be naturally regularized by the time scales of the fluctuations, and one can, in principle, calculate their transient effects. The presence of these regulators implies that such terms will have both real and imaginary contributions. While the real parts make the relaxation rates drive-dependent, corresponding imaginary parts provide the dynamic frequency shifts.

In this work, we report an alternate formulation of the QME for weakly driven quantum systems, where drive-induced dynamic frequency shifts result from the dispersive part of a second-order complex term, while the corresponding absorptive part renders the relaxation rates drive-dependent. Our approach involves the assumption of an explicit Hamiltonian to model the thermal fluctuations in a heat bath, since it is expected that a thermal bath would undergo fluctuations irrespective of the presence of a coupled quantum system. We note that the thermal fluctuations originating from collisional processes are essentially smooth functions of time in a very fine-grained scale. But as the system dynamics is expected to be much slower, such fluctuations are modeled as δ-correlated Gaussian processes in the timescales of system dynamics. As such, we use a normal Schrödinger evolution due to the time-dependent fluctuation Hamiltonian as well, while using its statistical properties as an asymptotic expression, only for performing ensemble averages. The use of an explicit noise (often Gaussian) in the Hamiltonian is common in the approaches based on the stochastic Schrödinger equation (SSE), however the subsequent analysis differs considerably from the route that we intend to take [4]. Our approach is more similar to Langevin dynamics, where we can write the ordinary dynamical equations even with a fluctuation Hamiltonian, since the fluctuations are not truly stochastic at all timescales. We use the method of coarse-graining in time and finite propagation under all relevant Hamiltonians so as to realize the fact that in the timescales of the dynamics of the quantum system, many instances of the fluctuations take place. Under these assumptions, both the weak drive and the coupling can be treated perturbatively. The leading second-order terms thus obtained—after ensemble averaging—take the form of dissipators regularized by the fluctuations. We show that for a two-level system (TLS) subjected to a linearly polarized transverse drive, the dispersive (imaginary) parts of the second-order drive susceptibilities yield the well-known second-order frequency shifts in appropriate limits, while their Kramers-Kronig pairs lead to drive-dependent relaxation phenomena. Unlike the commonly adopted approaches, our method does not require an RWA to obtain drive-dependent frequency shifts, and as such it includes the frequency renormalization due to counter-rotating terms of the drive as well along with their absorptive counterparts [4,20–22,35–38]. Later, we discuss the merits of our results in the context of various experimental data reported earlier and cited in the previous paragraphs.

II. DESCRIPTION OF THE PROBLEM

We consider a driven quantum system, which interacts with its local environment through a finite set of degrees of freedom. The drive and the interaction Hamiltonians are assumed to be weak, i.e., the strengths of these Hamiltonians are much less than the magnitude of the bare Hamiltonian of this quantum system. As such, the dynamics induced by the drive and the coupling remains within the perturbative regime. An ensemble of such quantum systems constitutes our system of interest and will be simply referred to as "system" in the rest of the paper. The collection of the corresponding local environments constitutes a thermal bath whose various time correlations are governed by the nature and the extent of the thermal fluctuations. Thus, a single representative quantum system of this collection, along with its local environment, evolves under the following Hamiltonian (in frequency units):

$$\mathcal{H}(t) = \mathcal{H}_{\rm S}^{\circ} + \mathcal{H}_{\rm L}^{\circ} + \mathcal{H}_{\rm SL} + \mathcal{H}_{\rm S}(t) + \mathcal{H}_{\rm L}(t), \tag{1}$$

where $\mathcal{H}_{\rm S}^{\circ}$ and $\mathcal{H}_{\rm L}^{\circ}$ represent the static Hamiltonians of the quantum system and its local-environment, respectively, which are weakly coupled by the term $\mathcal{H}_{\rm SL}$. $\mathcal{H}_{\rm S}(t)$ denotes the external drive to the quantum system. It is assumed that the bath is in thermal equilibrium at an inverse temperature β. The local environment experiences equilibrium fluctuations $\mathcal{H}_{\rm L}(t)$, which solely act on the bath degrees of freedom. $\mathcal{H}_{\rm L}(t)$ may assume different values for different ensemble members while respecting the constraints imposed by the requirement of sustained thermal equilibrium. Apart from $\mathcal{H}_{\rm L}(t)$, the other terms in the Hamiltonian are identical for all ensemble members and hence $\mathcal{H}_{\rm L}^{\circ}$ determines the equilibrium density matrix of the bath. Since the fluctuations do not drive the bath away from equilibrium, we choose $\mathcal{H}_{\rm L}(t)$ to be diagonal in the eigenbasis $\{|\phi_j\rangle\}$ of $\mathcal{H}_{\rm L}^{\circ}$, represented by $\mathcal{H}_{\rm L}(t) = \sum_j f_j(t)|\phi_j\rangle\langle\phi_j|$. $f_j(t)$'s are modeled as independent, Gaussian, δ-correlated stochastic variables with zero mean and standard deviation κ. We emphasize that the statistical properties of $f_j(t)$'s are asymptotic limits used to model the nature of thermal fluctuations. In the timescale of bath dynamics, they are smooth functions of time, while their two-point correlations are extremely short-lived.

Starting with this description, we perform all subsequent calculations in the interaction representation of $\mathcal{H}_{\rm S}^{\circ} + \mathcal{H}_{\rm L}^{\circ}$, and all Hamiltonians in this representation are denoted by H with relevant subscripts. Since $\mathcal{H}_{\rm L}(t)$ commutes with $\mathcal{H}_{\rm L}^{\circ}$ at all times, the form of the local fluctuations remains unchanged in the interaction representation. In the following sections, we derive a master equation for the system described above and explicitly show the relevant dynamical equations in the context of a TLS.

III. MASTER EQUATION WITH FINITE PROPAGATION FOR FLUCTUATIONS

We seek to derive a quantum master equation (QME), which captures the dynamics of the system described in the previous section. Since our problem concerns a single Hilbert space, a part of which undergoes rapid fluctuations, we follow the standard practice of (i) propagating for a large enough time Δt (> 0) over which fluctuations can be adequately averaged out, yet in the same interval $H_{\rm S}$ and $H_{\rm SL}$ should remain linearizable as usual, (ii) taking the ensemble average and a partial trace over the bath variables, and (iii) finally dividing both sides of the equation by Δt and replacing the resulting coarse-grained time derivative by an ordinary one to get the final QME [2]. The step (i) requires that the system and the fluctuations have widely separated timescales, i.e., $\tau_c \ll \tau_s$, where τ_c is the time during which the bath correlations are significant and τ_s denotes the timescale of system dynamics (determined by the magnitudes of $H_{\rm S}$ and $H_{\rm SL}$), such that we can find a Δt that obeys $\tau_c \ll \Delta t \ll \tau_s$.

We begin from the von Neumann–Liouville equation for a single member of the system along with its local environment, whose density matrix is denoted by $\widetilde{\rho}(t)$,

$$\frac{d}{dt}\widetilde{\rho}(t) = -i[H(t),\widetilde{\rho}(t)], \tag{2}$$

where $H(t) = H_{\rm S}(t) + H_{\rm SL}(t) + H_{\rm L}(t)$. The formal solution of the above equation for a finite-time interval t to $t + \Delta t$ is given by

$$\widetilde{\rho}(t+\Delta t) = \widetilde{\rho}(t) - i\int_t^{t+\Delta t} dt_1[H(t_1),\widetilde{\rho}(t_1)]. \tag{3}$$

For the dynamics of the system part, we obtain from the above, by taking partial trace over the environment degrees of freedom,

$$\begin{aligned}\widetilde{\rho}_{\rm S}(t+\Delta t) &= {\rm Tr}_{\rm L}\{\widetilde{\rho}(t+\Delta t)\} = {\rm Tr}_{\rm L}\{\widetilde{\rho}(t)\} - i\int_t^{t+\Delta t} dt_1 {\rm Tr}_{\rm L}[H_{\rm eff}(t_1) + H_{\rm L}(t_1), U(t_1)\widetilde{\rho}(t)U^\dagger(t_1)]\\ &= \widetilde{\rho}_{\rm S}(t) - i\int_t^{t+\Delta t} dt_1 {\rm Tr}_{\rm L}[H_{\rm eff}(t_1), U(t_1)\widetilde{\rho}(t)U^\dagger(t_1)],\end{aligned} \tag{4}$$

where $H_{\rm eff}(t) = H_{\rm S}(t) + H_{\rm SL}(t)$, $U(t_1) = U(t_1,t) = T\exp[-i\int_t^{t_1} dt_2 H(t_2)]$, and T is the Dyson time-ordering operator. In the above, the commutator involving $H_{\rm L}(t_1)$ vanishes due to the partial trace and $\widetilde{\rho}_{\rm S}(t)$ denotes the density matrix of the particular member of the system ensemble.

To obtain a master equation for the system, we perform a finite-time propagation of the right-hand side of (4) by keeping terms only up to the leading second order in $H_{\rm eff}$ while retaining all orders of $H_{\rm L}$. We emphasize that this construction is at the immediate next level of approximations as that of Bloch and Wangsness (barring the fluctuations), where only the leading linear order of $H_{\rm S}$ was retained while having quadratic orders in $H_{\rm SL}$ [1]. Since we intend to capture the dynamics of the system part while the local environment undergoes a large number of fluctuation instances, a form of the propagator U is required that captures the finite propagation due to $H_{\rm L}$ while only retaining the leading-order linear terms in $H_{\rm eff}$, in order to capture the overall second-order effect of the latter. Such a form of the propagator is readily available from the Neumann series as (Appendix A)

$$U(t_1) \approx U_L(t_1) - i\int_t^{t_1} dt_2 H_{\rm eff}(t_2) U_L(t_2), \quad t \leqslant t_1 \leqslant t + \Delta t \tag{5}$$

with $U_L(t_1) = U_L(t_1,t) = T\exp[-i\int_t^{t_1} dt_2 H_L(t_2)]$. We note that the above truncated form of U is strictly applicable only (i) if at least a part of $H_{\rm eff}(t)$ does not commute with $H_L(t)$ (i.e., $H_{\rm SL} \neq 0$) and (ii) $H_{\rm eff}(t)$ has a timescale much slower than the timescale of the fluctuations. In the case in which $H_{\rm SL} = 0$, we have $U(t_1) = U_S(t_1)U_L(t_1)$ with $U_S(t_1) = \exp[-i\int_t^{t_1} dt_2 H_S(t_2)]$, which results in pure unitary evolution of the spin systems under the external drive in the form of a Dyson series. Therefore, setting the coupling to the local environment to zero results in a system dynamics that is completely decoupled from the bath dynamics. Our use of Eq. (5) requires that $H_{\rm SL} \neq 0$ in the subsequent calculations.

We substitute Eq. (5) in Eq. (4) to obtain

$$\begin{aligned}
&\widetilde{\rho}_S(t+\Delta t) \\
&= \widetilde{\rho}_S(t) - i\int_t^{t+\Delta t} dt_1 \mathrm{Tr}_L[H_{\rm eff}(t_1), U_L(t_1)\widetilde{\rho}(t)U_L^\dagger(t_1)] \\
&\quad - \int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2 \mathrm{Tr}_L[H_{\rm eff}(t_1), H_{\rm eff}(t_2)\, U_L(t_2)\widetilde{\rho}(t)U_L^\dagger(t_1) \\
&\quad - U_L(t_1)\widetilde{\rho}(t)U_L^\dagger(t_2)\, H_{\rm eff}(t_2)] + O\big[H_{\rm eff}^3\big].
\end{aligned} \tag{6}$$

We note that the above form is exact up to the leading second order in $H_{\rm eff}$ and yet captures evolution solely under H_L up to, in principle, infinite orders through the U_L terms.

Next, we perform ensemble averaging of both sides of Eq. (6) and neglect the third- and higher-order contributions of $H_{\rm eff}$. Assuming that at the beginning of the coarse-graining interval the density matrix for the full system and bath can be factorized into that of the system and the bath with the latter at thermal equilibrium at an inverse temperature β, we obtain

$$\overline{U_L(t_1)\widetilde{\rho}(t)U_L^\dagger(t_2)} = \rho_S(t) \otimes \rho_L^{\rm eq} \exp\left(-\frac{1}{2}\kappa^2|t_1 - t_2|\right), \tag{7}$$

where $\rho_S(t)$ denotes the density matrix of the system whereas $\rho_L^{\rm eq}$ denotes the equilibrium density matrix of the bath in the interaction representation (Appendix B). The overline in Eq. (7) denotes ensemble averaging. The above expression is reminiscent of Karplus and Schwinger's construction of the density matrix of an ensemble of colliding molecules [40]. Using the above result, we find that the integrands in the second-order terms of the coarse-grained equation (6) take the form of a double commutator decaying within the timescale of $2/\kappa^2$. Thus $2/\kappa^2$ forms the upper bound of the timescales during which the bath correlations are significant and as such we replace it by τ_c. We thus have an equation of the form

$$\rho_S(t+\Delta t) - \rho_S(t) = -i\int_t^{t+\Delta t} dt_1 \mathrm{Tr}_L\big[H_{\rm eff}(t_1), \rho_S(t)\otimes\rho_L^{\rm eq}\big] - \int_t^{t+\Delta t} dt_1 \int_t^{t_1} dt_2 \mathrm{Tr}_L\big[H_{\rm eff}(t_1), \big[H_{\rm eff}(t_2), \rho_S(t)\otimes\rho_L^{\rm eq}\big]\big] e^{-|t_1-t_2|/\tau_c}. \tag{8}$$

Next, following the prescription of Cohen-Tannoudji *et al.*, we divide both sides of the resulting equation by Δt and approximate the coarse-grained time derivative ($\frac{\Delta\rho_S(t)}{\Delta t} = \frac{1}{\Delta t}[\rho_S(t+\Delta t) - \rho_S(t)]$) thus obtained on the left-hand side by an ordinary time derivative [2]. The resulting equation is time-local since the right-hand side depends only on the present state $\rho_S(t)$ as in Cohen-Tannoudji *et al.*'s description [2]. With $\mathrm{Tr}_L[H_{\rm SL}\rho_L^{\rm eq}] = 0$ as described in Schaller and Brandes' work as well as in Alicki *et al.*'s analysis, the coarse-grained equation can be expressed in the Lindblad-Gorini-Kossakowski-Sudarshan form for any $\Delta t \geqslant 0$ after transforming back to the Schrödinger picture [4,45,46]. The exponential factor, $\exp(-|t_1 - t_2|/\tau_c)$, can be absorbed in the stationary two-point correlation of bath operators obtained after the partial tracing, in the second-order terms involving only $H_{\rm SL}$. In the second-order drive terms, the exponential factor alone plays the role of a two-point correlation. The Kossakowski matrix, involving Fourier transforms of two-point correlation functions, is thus positive-semidefinite and as such generates a trace and positivity-preserving dynamical map resulting in a Markovian master equation [4,45–47].

Subsequently, we take the limit $\Delta t/\tau_c \to \infty$ to arrive at the following master equation:

$$\frac{d}{dt}\rho_S(t) = -i\,\mathrm{Tr}_L\big[H_{\rm eff}(t), \rho_S(t)\otimes\rho_L^{\rm eq}\big]^{\rm sec} - \int_0^\infty d\tau\, \mathrm{Tr}_L\big[H_{\rm eff}(t), \big[H_{\rm eff}(t-\tau), \rho_S(t)\otimes\rho_L^{\rm eq}\big]\big]^{\rm sec} e^{-|\tau|/\tau_c}, \tag{9}$$

where the superscript "sec" denotes that only the secular contributions are retained (ensured by the coarse graining) [2]. Unlike the usual forms of the master equation found in the literature, Eq. (9) has a finite, time-nonlocal, second-order contribution of the external drive to the system [1–3,5]. Equation (9) yields Lorentzian spectral density functions due to the presence of the exponential decay term and predicts the relaxation behavior along with the first-order nutation of the driven-dissipative system as in other forms of the QMEs [1–3,5].

IV. APPLICATION TO A TLS

We now use the master equation (9) to describe the dynamics of a driven-dissipative TLS. Spin-$\frac{1}{2}$ systems provide the most common example of TLS, and as such we define our Hamiltonians to describe such an ensemble. We assume $\mathcal{H}_S^\circ = \omega_\circ I_z$ to be the bare spin Hamiltonian and $\mathcal{H}_S(t) = \widetilde{\omega}_1\cos(\omega t)I_x = 2\omega_1\cos(\omega t)I_x$ to be the drive Hamiltonian. Here $I_\alpha, \alpha \in \{x,y,z\}$, denotes the components of the spin-angular-momentum operator, and they are given by the Pauli matrices as $I_\alpha = \frac{1}{2}\sigma_\alpha$. Also, $\omega_\circ$ denotes the Larmor frequency of the spins and $\widetilde{\omega}_1$ denotes the full strength of the chosen linearly polarized drive. The choice of linear polarization is deliberate since we intend to capture the effects of the counter-rotating terms in the second order, if any. We further assume that the external drive is nearly resonant, i.e., $\omega = \omega_\circ + \Delta\omega$, where $\Delta\omega/\omega_\circ \to 0$. This implies that the heterodyne detection followed by low-pass filtering common in spectroscopic measurements is equivalent to measurements made in a corotating frame of frequency ω [48]. As such, the interaction representations of the relevant corotating spin-1/2 observables are given by $F^{\mathrm{R}}_\alpha(t) = e^{-i\Delta\omega t I_z} I_\alpha e^{i\Delta\omega t I_z}$, where $\alpha \in \{x,y,z\}$. The dynamics of the expectation value of $F^{\mathrm{R}}_\alpha(t)$, denoted by $M_\alpha(t) = \mathrm{Tr}_{\mathrm{S}}[F^{\mathrm{R}}_\alpha(t)\rho_{\mathrm{S}}(t)]$, is given by

$$\frac{d}{dt}M_\alpha(t) = \mathrm{Tr}_{\mathrm{S}}\left[\left\{\frac{d}{dt}F^{\mathrm{R}}_\alpha(t)\right\}\rho_{\mathrm{S}}(t)\right] + \mathrm{Tr}_{\mathrm{S}}\left[F^{\mathrm{R}}_\alpha(t)\left\{\frac{d}{dt}\rho_{\mathrm{S}}(t)\right\}\right], \tag{10}$$

where Tr_{S} denotes the trace over the system degrees of freedom. We use our QME (9) on the right-hand side of the above equation to obtain the dynamical equations for $M_\alpha(t)$. The relevant timescales for the QME are $\tau_{\mathrm{c}} \ll \Delta t \ll \omega_1^{-1}, \omega_{\mathrm{SL}}^{-1}$ and $\omega_\circ^{-1} \leqslant \Delta t$, where ω_{SL} denotes the strength of the coupling [2].

The external drive, in the interaction representation, is $H_{\mathrm{S}}(t) = \omega_1[F^{\mathrm{C}}_{\mathrm{x}}(t) + F^{\mathrm{R}}_{\mathrm{x}}(t)]$, where the counter-rotating component of the drive field is $F^{\mathrm{C}}_{\mathrm{x}}(t) = e^{i\Omega t I_z} I_x e^{-i\Omega t I_z}$ with $\Omega = \omega + \omega_\circ$. The dynamical equations for $M_\alpha(t)$ can now be obtained directly from Eqs. (10) and (9) using the observables defined above. The near-resonance condition demands that in the secular limit, only the terms in $H_{\mathrm{S}}(t)$ with frequency $\Delta\omega$ (resonant or corotating terms), i.e., terms in $F^{\mathrm{R}}_{\mathrm{x}}(t)$, contribute in the first order of Eq. (9). In the following, we assume that the heat bath is isotropic, i.e., $\mathrm{Tr}_{\mathrm{L}}[H_{\mathrm{SL}}(t)\rho_{\mathrm{S}}(t) \otimes \rho^{\mathrm{eq}}_{\mathrm{L}}] = 0$, which in turn ensures that the cross terms between the drive and the coupling, in Eq. (9), vanish identically in the second order [1–3]. Thus $H_{\mathrm{SL}}(t)$ has no first-order contribution in Eq. (9) and its second-order contribution leads to the relaxation times T_1 and T_2 (longitudinal and transverse relaxation times, respectively) as well as the equilibrium value $M_\circ$, exactly in the same way as in Wangsness and Bloch's work [1]. The nonisotropic situation is beyond the scope of this work and will be discussed elsewhere.

On the other hand, the second-order secular drive terms have contributions from both the resonant $[F^{\mathrm{R}}_{\mathrm{x}}(t)]$ as well as the nonresonant $[F^{\mathrm{C}}_{\mathrm{x}}(t)]$ parts, resulting in complex susceptibilities proportional to ω_1^2. The secular integration in Eq. (8), i.e., integration over t_1, makes the cross terms between $F^{\mathrm{R}}_{\mathrm{x}}(t)$ and $F^{\mathrm{C}}_{\mathrm{x}}(t)$ as well as the nonsecular self-terms of $F^{\mathrm{C}}_{\mathrm{x}}(t)$ negligibly small in the second order [2]. Thus the master equation (9) retains only the secular self-terms of $F^{\mathrm{C}}_{\mathrm{x}}(t)$ in the second order of drive perturbation while retaining all possible self-terms from $F^{\mathrm{R}}_{\mathrm{x}}(t)$, which is manifestly secular. The absorptive and dispersive components of the second-order drive susceptibilities thus obtained involve Lorentzian spectral-density functions centered at $\Delta\omega$ and Ω and result in additional damping and shift terms in the dynamical equations.

Neglecting Lamb-Shift contributions from $H_{\mathrm{SL}}(t)$, we then arrive at the following form of the Bloch equations:

$$\begin{aligned}
\frac{d}{dt}M_{\mathrm{x}}(t) &= \Delta\omega_x M_{\mathrm{y}}(t) - \Gamma_x M_{\mathrm{x}}(t),\\
\frac{d}{dt}M_{\mathrm{y}}(t) &= -\Delta\omega_y M_{\mathrm{x}}(t) - \omega_1 M_{\mathrm{z}}(t) - \Gamma_y M_{\mathrm{y}}(t),\\
\frac{d}{dt}M_{\mathrm{z}}(t) &= \omega_1 M_{\mathrm{y}}(t) - \Gamma_z M_{\mathrm{z}}(t) + \frac{1}{T_1}M_\circ,
\end{aligned}\tag{11}$$

where the decay rates are $\Gamma_x = \frac{1}{T_2} + \eta_x$, $\Gamma_y = \frac{1}{T_2} + \eta_y$, and $\Gamma_z = \frac{1}{T_1} + \eta_z$, with

$$\begin{aligned}
\eta_x &= \omega_1^2 \frac{1}{2}\left[\frac{\tau_c}{1+\Omega^2\tau_c^2}\right],\\
\eta_y &= \omega_1^2\left[\frac{1}{2}\left(\frac{\tau_c}{1+\Omega^2\tau_c^2}\right) + \frac{\tau_c}{1+\Delta\omega^2\tau_c^2}\right],\\
\eta_z &= \omega_1^2\left[\frac{\tau_c}{1+\Omega^2\tau_c^2} + \frac{\tau_c}{1+\Delta\omega^2\tau_c^2}\right].
\end{aligned}\tag{12}$$

The frequency shifts are given by $\Delta\omega_x = \Delta\omega - \delta\omega_{\mathrm{C}}$ and $\Delta\omega_y = \Delta\omega - \delta\omega_{\mathrm{C}} + \delta\omega_{\mathrm{R}}$, where

$$\delta\omega_{\mathrm{C}} = \frac{1}{2}\left(\frac{\omega_1^2\Omega\tau_c^2}{1+\Omega^2\tau_c^2}\right) \tag{13}$$

is the frequency shift originating from the counter-rotating term $F^{\mathrm{C}}_{\mathrm{x}}(t)$, while

$$\delta\omega_{\mathrm{R}} = \frac{\omega_1^2\Delta\omega\tau_c^2}{1+\Delta\omega^2\tau_c^2} \tag{14}$$

is the same from the resonant term $F^{\mathrm{R}}_{\mathrm{x}}(t)$. The additional drive-dependent frequency shifts and the damping coefficients thus arrived at are Kramers-Kronig pairs obtained from the second-order drive susceptibilities.

V. COMPARISON WITH OTHER THEORETICAL AND EXPERIMENTAL RESULTS

In the following, we describe the implications of the second-order drive terms in the dynamics of a driven-dissipative TLS at various limits.

A. Bloch-Siegert shift

We note that in the limit $\Omega\tau_c > 1$, a condition often met in solid-state magnetic resonance spectroscopy because of slow fluctuations, $\delta\omega_{\mathrm{C}}$ converges to $\omega_1^2/2\Omega$, which we identify with the familiar Bloch-Siegert shift. For a more explicit comparison with Bloch and Siegert's original expression, the condition $\Delta\omega = 0$ introduces a shift in the resonance field given by

$$\delta\omega_{\mathrm{C}} = \frac{\omega_1^2}{4\,\omega_\circ} = \frac{\widetilde{\omega}_1^2}{16\,\omega_\circ} \tag{15}$$

in this limit [25]. Also, $\Delta\omega = 0$ implies $\delta\omega_{\mathrm{R}} = 0$ and the only frequency shift term arises from the counter-rotating component of the external drive, i.e., $\delta\omega_{\mathrm{C}}$. We note that the above expression for the limiting value of $\delta\omega_{\mathrm{C}}$ [Eq. (15)] matches with the known form of the Bloch-Siegert shift, in an isolated quantum system, obtained from a variety of different approaches, including Floquet dynamics, Magnus expansion, Fer expansion, CHRW, as well as Bloch and Siegert's original exposition [25–28,31]. However, the general form of the Bloch-Siegert shift, $\delta\omega_{\mathrm{C}}$, which depends on τ_c [Eq. (13)], has not been reported before. As such, it is expected that $\delta\omega_{\mathrm{C}}$ would depend on temperature unlike the claims from other theoretical treatments.

B. Frequency shift at large detuning

For a largely detuned, circularly polarized external drive of the form $H_{\mathrm{S}}(t) = \omega_1 F_{\mathrm{x}}^{\mathrm{R}}(t)$, with $\Delta\omega\Delta t \geqslant 1$, the secular approximation retains only the cross-commutators between I_+ and I_- in the second-order drive terms [2]. In this limit, the frequency shift $\delta\omega_{\mathrm{R}}$ is given by

$$\delta\omega_{\mathrm{R}} = \frac{1}{2}\left(\frac{\omega_1^2\Delta\omega\tau_c^2}{1+\Delta\omega^2\tau_c^2}\right). \tag{16}$$

Again, for $\Delta\omega\tau_c > 1$, $\delta\omega_{\mathrm{R}}$ approaches $\omega_1^2/2\Delta\omega$ as reported in Ref. [49]. It is known from the very early experiments on optical spectroscopy that two-photon "light shifts" have contributions from both the counter-rotating (Ω) as well as corotating ($\Delta\omega$) parts of an off-resonant excitation, consistent with a second-order perturbation in a Floquet computational basis [27,50–52]. As in the previous section, the generalized τ_c-dependent form of this shift [Eq. (14)] has not been reported before. The τ_c dependence of $\delta\omega_{\mathrm{R}}$ implies its temperature dependence. Moreover, unlike the result of an ordinary perturbation in the Floquet basis, the generalized form of $\delta\omega_{\mathrm{R}}$ vanishes in the on-resonance condition ($\Delta\omega \to 0$) without incurring any divergences [50,52]. Thus the only drive-dependent frequency shift in the case of on-resonance excitation arises from the counter-rotating component $F_{\mathrm{x}}^{\mathrm{C}}(t)$, as expected [25–27].

C. Redfield limit of the FID rate

Equations (11) are of the same form as the modified Bloch equations of Shakhmuratov *et al.* [Eq. (11) of Ref. [17]]. Hence the FID rate, Γ_{FID}, is given by the same expression as in their work [17], i.e.,

$$\Gamma_{\mathrm{FID}} = \frac{1}{T_2} + \sqrt{\Gamma_x\Gamma_y + \omega_1^2\frac{\Gamma_x}{\Gamma_z}}. \tag{17}$$

When the external drive is such that $\{\eta_x,\eta_y,\eta_z\} \gg \{1/T_1,1/T_2\}$, the decay rates are dominated by the second-order drive contributions and we have $\Gamma_x \approx \eta_x$, $\Gamma_y \approx \eta_y$, and $\Gamma_z \approx \eta_z$. Now if $\omega_1^2\tau_c < 1$, we can approximate Γ_{FID} as

$$\Gamma_{\mathrm{FID}} = \frac{1}{T_2} + \omega_1\sqrt{\frac{\eta_x}{\eta_z}}. \tag{18}$$

For $\{\Omega\tau_c,\omega\tau_c\} < 1$, the FID rate can be further approximated as

$$\Gamma_{\mathrm{FID}} = \frac{1}{T_2} + \frac{\omega_1}{2}, \tag{19}$$

which illustrates the Redfield limit [9,11,17].

D. Non-Bloch decay of Rabi oscillations

From Eqs. (11) we find that the decay rate of Rabi oscillations or transient nutations (TN), Γ_{TN}, is given by

$$\Gamma_{\mathrm{TN}} = \tfrac{1}{2}(\Gamma_z + \Gamma_y), \tag{20}$$

which is manifestly quadratic in the drive strength ω_1 [17]. Although this result explains the increase in decay rates of Rabi oscillations with drive strength, it does not corroborate their linear dependence, observed in dipolar solids [15–17]. To this end, we note that the observed linear dependence of Rabi decay rates in solids originates from signal averaging in dipolar-coupled spin networks as shown by De Raedt *et al.* and Baibekov [53,54]. More recent measurements by Nellutla *et al.* as well as Bertaina *et al.* report a nonlinear dependence of the decay rates of Rabi oscillations, which may arise from crystal imperfections in solids or a second-order drive term as in our case [18,19,54].

Interestingly, Ramsay *et al.*'s theoretical as well as experimental investigations on excitonic Rabi oscillations in the weak-coupling limit as well as the variational polaron method of McCutcheon *et al.* do predict frequency renormalization and damping terms quadratic in the Rabi frequency [20–22]. The limitation of these and related approaches lies in the fact that the methods rely on an *a priori* RWA. Hence, these approaches cannot, in principle, predict Bloch-Siegert-like frequency shifts, which exclusively originate from the counter-rotating components of an external drive, as the RWA removes all counter-rotating terms from the Hamiltonian. A more physical approach is to use a secular approximation using the time-coarse graining of the dynamical equations, which we have adopted in our derivation [2,4]. Also, the Bloch-Siegert-type frequency shifts are independent of the system-bath coupling-strength and can be derived even for an isolated quantum system as mentioned before. On the contrary, the dynamic frequency shifts obtained from the above methods [20–22] explicitly depend on the system-bath coupling.

Recent investigations on the Rabi oscillations of an ensemble of nuclear spins in an isotropic liquid using nuclear magnetic resonance (NMR) spectroscopy show that the decay rates of Rabi oscillations have a quadratic dependence on the Rabi frequency—the decay law being given by $\Gamma_{\mathrm{TN}} = \lambda_0 + \lambda_1\omega_1^2$ [24]. This quadratic form is corroborated by an estimation of Γ_{TN} from our modified Bloch equations (11) using Eq. (20). The most interesting feature of the experimental data is that the measured order of magnitude of λ_1 matches with the order of magnitude of rotational correlation times of molecules in liquids (picoseconds) [24,55–58]. Since τ_c is a timescale during which the bath correlations are significant in our model, we can expect that in an isotropic liquid at a finite temperature, τ_c is of the order of the rotational correlation times, i.e., ~picoseconds. For $\Omega \sim 10^9$ Hz and $\Delta\omega \to 0$ Hz as in this experiment, the Lorentzian damping rates η_x, η_y, and η_z can be approximated by terms proportional to $\omega_1^2\tau_c$ in exact agreement with the orders of magnitudes of non-Bloch decay rates measured experimentally [24]. It is worthwhile to mention here that the non-Bloch decay rates in the works of Ramsay *et al.*, McCutcheon *et al.*, and Mogilevtsev *et al.* are proportional to the square of the coupling strength ω_{SL} [20–23]. One can get an idea about the magnitude of the coupling strength from a measurement of the T_1 and T_2 relaxation times defined in the previous section. In the experiment on the non-Bloch decay or Rabi oscillations reported by the authors, these relaxation times were ~ 1 s [24]. As such, it is evident that the order of magnitude of the non-Bloch decay rates measured therein cannot be explained by terms proportional to $\omega_1^2\omega_{\mathrm{SL}}^2$. Moreover, Ramsay *et al.*'s theoretical exposition introduces the drive-independent relaxation rates phenomenologically, whereas our method explains both the drive-dependent and drive-independent contributions to the relaxation rates as discussed in the previous section [21].

Thus the QME derived here remains the only approach that can correctly describe the drive-dependent non-Bloch decay in nuclear spin ensembles in isotropic liquids with correct orders of magnitude. Also, since the rotational correlation times of molecules in liquids decrease with increase in temperature, it is expected that the non-Bloch regime will become less prominent with an increase in temperature in these systems. This is also in agreement with the experimental results obtained by the authors [24].

We note that the Floquet Markov master equation derived by Yan *et al.* for a driven-dissipative TLS using the CHRW method does give signatures of the Bloch-Siegert shift as well as drive-dependent line-broadening [31]. But a closed-form expression for these quantities has not been obtained for this system [31]. Also the drive-dependent damping rates that can be obtained from Floquet Markov master equations will depend on ω_{SL} and as such cannot explain the existence of terms of the form $\omega_1^2\tau_c$ described above.

VI. DISCUSSIONS

The second-order effects of the irradiation appear as shift and damping terms with magnitudes proportional to the square of the drive strength. As such, these terms remain in the equation of motion even when $H_{\mathrm{SL}} = 0$, an apparently paradoxical result. We have laid down the premise that, for this derivation, from Eq. (6) and beyond, $H_{\mathrm{SL}} \neq 0$. However, as discussed below, we can still resolve the paradox by carefully checking the other limits, whose values we have assumed to be based on the magnitude of H_{SL}, i.e., ω_{SL}. At $\omega_{\mathrm{SL}} = 0$, the Hilbert space relevant to the problem would be a direct product of two disjoint Hilbert spaces, and complete unitary dynamics is expected as discussed before. To this end, we note that our treatment begins with a choice of Δt over which many instances of the fluctuation have been assumed to take place. After an ensemble average over the fluctuations and a partial trace over the lattice variables, we obtain the final equation by approximating the coarse-grained derivative over Δt by an ordinary time derivative. Such an assumption is meaningful only when $\omega_{\mathrm{SL}} \neq 0$, i.e., when the system and bath are parts of a common Hilbert space, and as such a wide timescale separation exists in the problem. Therefore, the choice of setting $H_{\mathrm{SL}} = 0$ (i.e., $\omega_{\mathrm{SL}} = 0$) would naturally be accomplished provided one selects $\Delta t \to 0$ as well. In fact, analogous treatments often scale Δt with ω_{SL} to unambiguously indicate that Δt and ω_{SL} are not two independent parameters [59,60]. It is obvious that instead of setting $\Delta t/\tau_c \to \infty$ if we take the limit $\Delta t \to 0$, after taking partial trace over the lattice and dividing both sides of Eq. (8) by Δt, we immediately recover the pure unitary dynamics due to the irradiation, since all second-order terms vanish in this limit.

It is important to delve deeper into the origin of the exponential decay factor, $\exp(-|\tau|/\tau_c)$, which regularizes all the second-order terms of Eq. (9). The generic state of a particular member of our system coupled with its local environment, $|\psi(t)\rangle$, can be expanded in the product basis as $|\psi(t)\rangle = \sum_{j,k} c_{jk}(t)|\chi_j\rangle \otimes |\phi_k\rangle$, where $\{|\chi_j\rangle\}$ are the eigenstates of $\mathcal{H}_{\mathrm{S}}^{\circ}$. Thus $U_{\mathrm{L}}(t_1)$ acting on $|\psi(t)\rangle$ introduces random phases into the state function as $U_{\mathrm{L}}(t_1)|\psi(t)\rangle = \sum_{j,k} c_{jk}(t) \exp\{-i\int_t^{t_1} dt_2 f_k(t_2)\}|\chi_j\rangle \otimes |\phi_k\rangle$. In all the second-order terms of Eq. (6), U_{L}'s appear with time instances inherited from the Hamiltonian H_{eff}. In these terms, the external drive acts at time instants t_1 and t_2 preserving secularity while the state functions pick up random phases from the fluctuations through $U_{\mathrm{L}}(t_1)U_{\mathrm{L}}^{\dagger}(t_2)$. Therefore, although the drive $H_{\mathrm{S}}(t)$ commutes with the fluctuation $H_{\mathrm{L}}(t)$, the random phases thus picked up by the state functions over the coarse-grained time interval $|t_1 - t_2|$ give rise to a decay after ensemble averaging.

As illustrated in the preceding section, our QME (9) provides a single approach to describe almost all features of driven-dissipative quantum dynamics up to the second order in the drive strength. The simplicity of the prescribed method further makes it more usable in predicting results of realistic experiments. For example, the method outlined here does not require complicated frame transformations as in CHRW, variational-polaron, and other related methods [20–22,31]. Also, RWA-type approximations required in these methods are not necessary in our approach [20–22,31]. On the other hand, Floquet-based methods for describing driven-dissipative quantum dynamics do not provide a closed-form expression for drive-dependent decay rates, which matches with the reported experimental results [6,31]. We note that for a drive having multiple frequency modes, our approach can directly be applied, whereas the methods mentioned above may turn out to be unsuitable. Calculations based on the Floquet theoretic approaches will be formidable while the methods using frame transformations would become impractical due to the requirement of a cascade of such transformations.

The form of the fluctuations chosen to describe our system ensures that the bath spectral densities have a Lorentzian form as is expected in gaseous or liquid environments, where fluctuations in the molecular degrees of freedom can be considered to be classical [45]. To obtain other forms of the spectral densities, different models for the fluctuations may be considered.

VII. CONCLUSION

The QME (9) provides a possible way to explain the second-order drive-dependent phenomena observed in open quantum systems. The key step of this construction involves the regularization of the second-order dissipators originating from the coupling as well as the drive by the environmental fluctuations. The real and imaginary parts of the second-order drive contributions thus obtained explain the drive dependence of relaxation rates and frequency shifts, respectively. Our method has several advantages over the conventional approaches used to describe higher-order effects of a resonant drive. First of all, our derivation does not require an *a priori* assumption of RWA and hence it can capture the contributions of the counter-rotating part of an external drive in the form of the Bloch-Siegert shift and its corresponding absorptive part. Unlike a perturbative treatment of isolated systems in the Floquet basis, the second-order frequency shift obtained from the corotating component of an external drive in our method does not diverge in the on-resonance condition [50,52]. On the contrary, it becomes vanishingly small, as expected from other related theories [25–27]. Since the Bloch-Siegert shift has traditionally been derived from a microcanonical perspective or in the regime of a dynamical steady state, its absorptive Kramers-Kronig pair has not been reported before. Secondly, our proposed method does not depend on any specific form of the bath Hamiltonian and system-bath coupling, which calls for a wider range of applicability. The only model that has been assumed pertains to the form of thermal fluctuations, which, as mentioned before, are asymptotic properties expressing our ignorance of the full many-particle dynamics. As thermal fluctuations depend on the temperature of the bath, our approach provides a probable indication on the temperature dependence of these second-order drive terms.

Also, our method accurately describes the non-Bloch decay of Rabi oscillations in liquid NMR with correct orders of magnitude and temperature dependence. Decay rates proportional to $\omega_1^2\tau_c$, as obtained in this system, cannot be obtained with correct magnitudes from any other method describing driven-dissipative dynamics. Another major advantage of our approach is its inherent simplicity, which makes it more suitable to apply in cases in which the quantum system is subjected to a multifrequency drive. While the derived QME (9) is general, the assumption of an isotropic heat bath implies that the dynamical equations (11) derived for a TLS using (9) are directly applicable to liquids and gases. This method can easily be extended to an anisotropic medium by suitably modifying or adding appropriate Hamiltonians for such systems. We also note that the choice of δ-correlated Gaussian fluctuations leads to the regularization of the dissipators in our work, but other noise models as well as fluctuation Hamiltonians may also serve the purpose. Since no realistic quantum system can truly be isolated from the environment, faster quantum manipulation of qubits with stronger drives results in larger values of drive-dependent damping rates and frequency shifts. In view of this, we surmise that the knowledge of drive-dependent second-order terms would thus provide insights for designing more efficient qubit manipulation protocols.

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APPENDIX A: CONSTRUCTION OF THE FINITE PROPAGATOR

Following the description of the system and the bath laid out in the body of the paper and the notations introduced therein, we intend to construct a finite propagator $U(t_1)$, valid for the coarse-grained time interval, $t_1 \in [t,t+\Delta t]$. The coarse-graining time Δt is such that the contribution of H_{eff} in $U(t_1)$ can be linearized, and only the leading first-order terms are retained. On the contrary, many instances of the fluctuation take place within Δt, and as such we retain all possible higher-order terms of H_{L}. The explicit construction of the finite-time propagator begins from the Schrödinger equation:

$$\frac{d}{dt}U(t) = -i\,H(t)\,U(t) \tag{A1}$$

and its formal solution in the domain $[t,t_1]$ (with $t_1 > t$):

$$\begin{aligned} U(t_1) &= \mathbb{1} - i\int_t^{t_1} dt_2\, H(t_2)\,U(t_2) \\ &= \mathbb{1} - i\int_t^{t_1} dt_2\, H_{\text{eff}}(t_2)\,U(t_2) - i\int_t^{t_1} dt_2\, H_{\text{L}}(t_2)\,U(t_2). \end{aligned} \tag{A2}$$

Since $t_1 \in [t,t+\Delta t]$ by assumption, the interval $(t_1 - t) \ll \tau_s$ and as such further propagation due to H_{eff} can be neglected on the right-hand side of Eq. (A2). Thus collecting all the remaining terms on the right-hand side of Eq. (A2), we get a finite propagator with a leading linear order term in H_{eff} of the form

$$\begin{aligned} U(t_1) \approx \mathbb{1} &- i\int_t^{t_1} dt_2\, H_{\text{eff}}(t_2)\,U_{\text{L}}(t_2) \\ &- i\int_t^{t_1} dt_2\, H_{\text{L}}(t_2)\,U_{\text{L}}(t_2). \end{aligned} \tag{A3}$$

Combining the last term on the right-hand side with the identity, we can rewrite the above equation as

$$U(t_1) \approx U_{\text{L}}(t_1) - i\int_t^{t_1} dt_2\, H_{\text{eff}}(t_2)\,U_{\text{L}}(t_2). \tag{A4}$$

APPENDIX B: EMERGENCE OF THE REGULATOR FROM THERMAL FLUCTUATIONS

Following the usual practice, we too assume that at the beginning of the coarse-graining interval the full density matrix has the factorized form

$$\rho(t) = \rho_{\text{S}}(t) \otimes \rho_{\text{L}}^{\text{eq}}, \tag{B1}$$

where $\rho(t) = \overline{\widetilde{\rho}(t)}$ and $\rho_{\text{L}}^{\text{eq}} = \exp(-\beta\mathcal{H}_{\text{L}}^{\circ})/\mathcal{Z}_{\text{L}}$ denotes the equilibrium density matrix of the bath, $\mathcal{Z}_{\text{L}}$ being the partition function [1–3].

We thus have

$$\begin{aligned} &\overline{U_{\text{L}}(t_1)\widetilde{\rho}(t)U_{\text{L}}^{\dagger}(t_2)} \\ &= \rho_{\text{S}}(t) \otimes \sum_j \frac{e^{-\beta\omega_j}}{\mathcal{Z}_{\text{L}}}|\phi_j\rangle\langle\phi_j| \\ &\quad \times \overline{\exp\left\{-i\int_t^{t_1} dt_3\, f_j(t_3) + i\int_t^{t_2} dt_4\, f_j(t_4)\right\}}. \end{aligned} \tag{B2}$$

In the above expression, ω_j denotes the eigenvalue of $\mathcal{H}_{\text{L}}^{\circ}$ corresponding to $|\phi_j\rangle$, and we have made use of the fact that $H_{\text{L}}(t_1)$ and thus the propagators $U_{\text{L}}(t_1)$ are independent of the initial distribution of the lattice states $\forall\, t_1 \geqslant t$.

Thus we obtain

$$\overline{U_{\mathrm{L}}(t_1)\widetilde{\rho}(t)U_{\mathrm{L}}^{\dagger}(t_2)} = \rho_{\mathrm{S}}(t)\otimes\rho_{\mathrm{L}}^{\mathrm{eq}}\exp\left(-\frac{1}{2}\kappa^2|t_1-t_2|\right). \tag{B3}$$

In deriving the above, we have used the cumulant expansion for Gaussian stochastic processes with usual δ correlation in time, for which only the terms up to the second cumulant survive. A further assumption of zero mean (as in our model) leaves only the exponentially decaying factor, $\exp(-\frac{1}{2}\kappa^2|t_1-t_2|)$. We note that as $2/\kappa^2$ becomes small, the exponential regulator vanishes faster with increasing $|t_1-t_2|$—the limiting case being a memoryless bath.

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