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Non-perturbative environmental influence on dephasing

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Environmental noise leads to dephasing and relaxation in a quantum system. Often, a rigorous treatment of multiple noise sources within a system-bath approach is not possible. We discuss the influence of environmental fluctuations on a quantum system whose dynamics is dephasing already due to a phenomenologically treated additional noise source. For this situation, we develop a path integral approach, which allows to treat the system-environment coupling numerically exact, and additionally we extend standard perturbative approaches. We observe strong deviations between the numerical exact and the perturbative results even for weak system-bath coupling. This shows that standard perturbative approaches fail for additional, even weak, system-bath couplings if the system dynamics is already dissipative.

Open quantum dynamics is a very successful approach to describe and treat dissipative effects like relaxation, decoherence and dephasing in quantum systems [1–3]. Dissipation results therein by coupling the quantum system of interest to an environment. The latter is typically described by a set of harmonic oscillators bilinear coupled to the system. The according system-bath model can then be treated either perturbatively or by numerical exact methods. This allows successful treatment of problems like energy transfer in photosynthetic complexes [4, 5], fluorescence properties of optical quantum dots [6] and dephasing in various qubit realizations [7], for example, in two-electron charge qubits [8].

The quantum systems of interest are typically subject to various noise sources. Charge and flux qubits, for example, experience noise due to phonons, voltage fluctuations in the various gates, charged defects and currents through nearby quantum point contacts [7, 9]. Chromophores in photosynthetic complexes are disturbed by strong environmental fluctuations due to intra- and intermolecular vibrations of the photoactive complexes, vibrations of embedding proteins, solvent fluctuations and the charge separation in the reaction center [5]. Usually, the various noise sources are described by one effective bath if they all couple identically to the quantum system. Alternatively, one focusses on the main noise source and treats the others phenomenologically. Pure dephasing effects in flux qubits due to defects are phenomenologically treated by introducing dephasing rates. In energy transfer in photosynthetic complexes the reaction center is often included as energy sink described by phenomenological Lindblad rates [10].

A system-bath approach treats the environmental influence on a quantum system with Hamiltonian dynamics. In contrast, the dynamic of problems, which include a phenomenological sink or dephasing rate, is Liouvillian, i.e. is determined by a Liouville von Neumann equation. Environmental noise which additionally acts on such a dissipative quantum system can then not be treated within a standard system-bath approach. Approximately, one might determine the influence of each noise source independently. Thereby, all cross correla-

tions between the different fluctuations are neglected.

We extend the numerical exact quasi-adiabatic path integral approach [11–13] and a perturbative approach [14, 15] to allow for Liouvillian system dynamics. We then discuss a quantum two-level system (TLS) which dephases via a phenomenological dephasing rate γ_D and is subject to environmental fluctuations which we treat with the extended system-bath approach. We observe that the dephasing of the TLS due to γ_D strongly suppresses the dissipative influence of the environmental fluctuations. Surprisingly, the perturbative results differ quantitatively and qualitatively strongly from numerical exact results even at weak system-bath coupling. Thereby, the dephasing due to γ_D is treated exactly in both cases. Accordingly, perturbative treatment of system-bath coupling for a dephasing quantum two-level system fails.

We study a quantum two level system (TLS) with dipolar coupling Δ leading to a system Hamiltonian

$$H_S = \frac{\Delta}{2} \sigma_x. \quad (1)$$

The TLS is disturbed by two independent fluctuation sources of which we model one as a harmonic bath, i.e. $H_{SB,z}$, leading to a Hamiltonian $H = H_S + H_{SB,z}$ with

$$H_{SB,z} = \sum_{k=1}^M \frac{p_k^2}{2m_k} + \frac{1}{2} m_k \omega_k^2 \left(q_k - \frac{\lambda_k \hat{\sigma}_z}{m_k \omega_k^2} \right)^2 \quad (2)$$

and $[q_k, p_k] = i\hbar \delta_{k,k'}$. Herein, the q_k and p_k are the position and momentum of mode k with frequency ω_k coupled via λ_k to the system. Explicitly, the system-bath coupling terms are $H_{I,z} = -\hat{\sigma}_z \sum_{k=1}^M \lambda_k q_k$ and $H_{SB,z} = H_{I,z} + H_{B,z}$. The (longitudinal) fluctuations of the energy difference between the eigenstates to σ_z , induced by $H_{SB,z}$, result in energy exchange between system and bath and thus relaxation and dephasing. For simplicity, we employ an Ohmic spectral function for the bath, i.e.

$$G_z(\omega) = \sum_{k=1}^M \frac{\lambda_k^2}{2m_k \omega_k} \delta(\omega - \omega_k) = \gamma_z \omega \frac{\omega_c^2}{\omega_c^2 + \omega^2} \quad (3)$$

with coupling strength γ_z and cut-off frequency ω_c .

The second fluctuation source is assumed to cause pure dephasing. Most often pure dephasing noise sources are difficult to characterize in detail and a full description is missing. At temperatures $k_B T \simeq \Delta$, typically pure dephasing can effectively be treated by incorporating into the von-Neumann equation explicit dephasing terms [6, 17–21]. Thus, the dynamics of the TLS is determined by the von-Neumann equation

$$\partial_t W = -\frac{i}{\hbar}[H_S + H_{SB,z}, W] - \Gamma_D W = \mathcal{L}_e W \quad (4)$$

for the statistical operator $W(t)$ with $\Gamma_D = \text{diag}(0, 0, \gamma_D, \gamma_D)$ in a basis $\{\mathbb{1}, \sigma_x, \sigma_y, \sigma_z\}$ and the dephasing rate γ_D . The right hand side of (4) defines the Liouvillian $\mathcal{L}_e = -\frac{i}{\hbar}[H_S + H_{SB,z}, \cdot] - \Gamma_D$. We aim now at a system-bath approach which allows a numerical exact treatment of the $H_{SB,z}$ noise in eq. (4).

Introducing the time evolution super-operator $\mathcal{U}(t, t_0)$ via $W(t) = \mathcal{U}(t, t_0)W(t_0)$ and $\mathcal{U}(t, t_0) = \exp(\mathcal{L}_e(t - t_0))$ allows for an alternative description of the dynamics in terms of a Dyson equation

$$\begin{aligned} \mathcal{U}(t, t_0) &= \mathcal{U}_0(t, t_0) + \int_{t_0}^t ds \mathcal{U}_0(t, s) \mathcal{L}_I \mathcal{U}_0(s, t_0) \\ &+ \int_{t_0}^t ds \int_{t_0}^s ds' \mathcal{U}_0(t, s) \mathcal{L}_I \mathcal{U}_0(s, s') \mathcal{L}_I \mathcal{U}(s', t_0) \end{aligned} \quad (5)$$

which facilitates perturbative approaches. The bare evolution is $\mathcal{U}_0(t, t_0) = \exp(\mathcal{L}_0(t - t_0))$ with $\mathcal{L}_0 = -\frac{i}{\hbar}[H_S + H_{B,z}, \cdot] - \Gamma_D$ and the system-bath coupling leads to $\mathcal{L}_I = -\frac{i}{\hbar}[H_{I,z}, \cdot]$. Typically, a factorized initial state $W(0) = \rho_S(0) \otimes \rho_{Bz,eq}$ with the bath in thermal equilibrium, i.e. $\rho_{Bz,eq} = e^{-\beta H_{SB,z}} / \text{Tr}\{e^{-\beta H_{SB,z}}\}$ is assumed. The effective system dynamics under the influence of the environment is obtained by integrating out the bath degrees of freedom leading to $\rho_{\text{eff}}(t) = \text{Tr}_B\{W(t)\}$ or, alternatively, the effective time evolution super-operator $\mathcal{U}_{\text{eff}}(t, t_0) = \langle \mathcal{U}(t, t_0) \rangle_B = \text{Tr}_B\{\mathcal{U}(t, t_0) \rho_{Bz,eq}\}$.

In case of purely Hamiltonian dynamics, i.e. for vanishing dephasing γ_D , resummed perturbative treatments leading to Redfield-type master equations are efficient for small system-bath coupling strength, i.e. $\gamma_z \ll 1$, to describe the system dynamics and the dissipative bath influence [16]. One such approach is RESPET [14, 15] which derives the effective time evolution super-operator by integrating out the bath degrees of freedom in equation (5). For Liouvillian system dynamics, i.e. finite γ_D , such a resummed perturbative treatment is formally leading to

$$\begin{aligned} \mathcal{U}_{\text{eff}}(t, t_0) &= \mathcal{U}_S(t, t_0) \\ &+ \int_{t_0}^t ds \int_{t_0}^s ds' \mathcal{U}_S(t, s) \mathcal{M}(s, s') \mathcal{U}_{\text{eff}}(s', t_0). \end{aligned} \quad (6)$$

Therein, $\mathcal{U}_S(t, t_0) = \exp(\mathcal{L}_S(t - t_0))$ with $\mathcal{L}_S = -\frac{i}{\hbar}[H_S, \cdot] - \Gamma_D$. The memory kernel $\mathcal{M}(s, s')$ is in lowest order perturbative treatment given as $\mathcal{M}(s, s') = \langle \mathcal{L}_I \mathcal{U}_0(s, s') \mathcal{L}_I \rangle_B$.

As dynamic observable we discuss $P_z(t) = \langle \sigma_z \rangle(t)$ whose derivation is now straight forward. We obtain for $P_z(t) \simeq \cos \Delta t e^{-\Gamma_p t}$ with the rate

$$\begin{aligned} \Gamma_p(\gamma_D) &= \gamma_D - \int_{-\infty}^{\infty} d\omega G(\omega) \coth\left(\beta \frac{\omega}{2}\right) \frac{\gamma_D}{(\Delta - \omega)^2 + \gamma_D^2} \\ &+ 2\pi \text{Re} \left\{ G(\Delta - i\gamma_D) \coth\left(\beta \frac{\Delta - i\gamma_D}{2}\right) \right\} \end{aligned} \quad (7)$$

for initial system state $\rho_S(0) = \frac{1}{2}(\mathbb{1} + \sigma_z)$. Note that additional small non-Markovian contributions to $P_z(t)$ are neglected since they do not exhibit damped oscillations and, thus, will not contribute to an observed damping rate.

Little is known about the reliability of such a perturbative approach to treat a system-bath coupling when the system dynamics is Liouvillian instead of Hamiltonian, i.e. for finite γ_D . In order to test it, we must devise a numerical exact approach to treat the dynamics, specifically the $H_{SB,z}$ noise in Eq.(4). The regular QUAPI approach [11, 12] employs a time discretization to split the quantum mechanical time evolution operator $U(t) = \exp(-iHt)$ which combined with a symmetric Trotter splitting leads to a description in terms of a path integral where the bath effects enter via a Feynman-Vernon influence functional. The effective dynamics of Eq. (4) can, however, not be described in terms of a quantum mechanical time evolution operator. Instead we must employ a time-evolution super operator

$$\mathcal{U}_e(t) = e^{\mathcal{L}_e t} \text{ and } W(t) = \mathcal{U}_e(t)W(0). \quad (8)$$

Thereby, $\mathcal{L}_e = \mathcal{L}_S + \mathcal{L}_{SB,z}$ and $\mathcal{L}_{SB,z} = -(i/\hbar)[H_{SB,z}, \cdot]$ and $\mathcal{L}_S = -(i/\hbar)[H_S, \cdot] - \Gamma_D$. To proceed we discretize time in N steps, i.e. $t = N\Delta t$, and $\mathcal{U}_e = \prod_{j=1}^N \mathcal{U}_{e,j}$ with $\mathcal{U}_{e,j} = e^{\mathcal{L}_e \Delta t}$. Inserting furthermore $\mathbb{1}$ -super-operators, i.e.

$$\mathbb{1} = \int_{-\infty}^{\infty} d\sigma^+ \int_{-\infty}^{\infty} d\sigma^- \int_{-\infty}^{\infty} d\mathbf{q}^+ \int_{-\infty}^{\infty} d\mathbf{q}^- |\sigma^\pm, \mathbf{q}^\pm\rangle \langle \sigma^\pm, \mathbf{q}^\pm|.$$

in between all time steps leads to a discretized path sum. Herein the super-states $|\sigma^\pm, \mathbf{q}^\pm\rangle = |\sigma^\pm, \mathbf{q}^\pm\rangle \langle \sigma^\mp, \mathbf{q}^\mp|$ and the scalar product is defined as $(A|B) = \text{Tr}\{A^\dagger \cdot B\}$ for operators A and B acting on the Hilbert space.

To proceed we need the elements of the time-evolution super operator

$$\begin{aligned} (\sigma_j^\pm, \mathbf{q}_j^\pm | e^{\mathcal{L}_e \Delta t} | \sigma_{j-1}^\pm, \mathbf{q}_{j-1}^\pm \rangle = \\ \text{Tr} \{ |\sigma_j^-, \mathbf{q}_j^- \rangle \langle \sigma_j^+, \mathbf{q}_j^+ | e^{\mathcal{L}_e \Delta t} [|\sigma_{j-1}^+, \mathbf{q}_{j-1}^+ \rangle \langle \sigma_{j-1}^-, \mathbf{q}_{j-1}^- |] \} \end{aligned} \quad (9)$$

where we inserted the $[\cdot]$ -brackets in order to highlight on which operator the time evolution super-operator acts. Employing the symmetric Trotter splitting to split the time-evolution super operator leads to

$$\mathcal{U}_j \simeq e^{\mathcal{L}_{SB,z} \Delta t / 2} e^{\mathcal{L}_S \Delta t} e^{\mathcal{L}_{SB,z} \Delta t / 2} + O(\Delta t^3).$$

With $e^{\mathcal{L}_{SB}\Delta t/2}[A] = e^{-iH_{SB}\Delta t/2\hbar} A e^{iH_{SB}\Delta t/2\hbar}$ we obtain

$$\begin{aligned} \langle \sigma_j^\pm, \mathbf{q}_j^\pm | e^{\mathcal{L}_e \Delta t} | \sigma_{j-1}^\pm, \mathbf{q}_{j-1}^\pm \rangle &= \langle \sigma_j^\pm | e^{\mathcal{L}_S \Delta t} | \sigma_{j-1}^\pm \rangle \cdot \\ \cdot \langle \mathbf{q}_j^\pm | e^{-iH_{SB}(\sigma_j^\pm)\Delta t/2\hbar} e^{-iH_{SB}(\sigma_{j-1}^\pm)\Delta t/2\hbar} | \mathbf{q}_{j-1}^\pm \rangle \\ \cdot \langle \mathbf{q}_{j-1}^- | e^{iH_{SB}(\sigma_{j-1}^-)\Delta t/2\hbar} e^{iH_{SB}(\sigma_j^-)\Delta t/2\hbar} | \mathbf{q}_j^- \rangle. \end{aligned} \quad (10)$$

Thus, as in the regular QUAPI scheme the system and the bath dynamics are separated on a single time slice and the bath influences are summed up into an influence functional. Assuming a factorized initial condition, we obtain for the components of the effective statistical operator of the system

$$\begin{aligned} \langle \sigma_N^\pm | \rho_{\text{eff}}(t) | \sigma_N^\mp \rangle &= \prod_{j=0}^{N-1} \int_{-\infty}^{\infty} d\sigma_j^+ \int_{-\infty}^{\infty} d\sigma_j^- \langle \sigma_{j+1}^\pm | e^{\mathcal{L}_S \Delta t} | \sigma_j^\pm \rangle \\ \cdot \langle \sigma_0^\pm | \rho(0) | \sigma_0^\mp \rangle \cdot I^{(N)}(\sigma_0^\pm, \sigma_1^\pm, \dots, \sigma_N^\pm). \end{aligned} \quad (11)$$

with

$$\langle \sigma_{j+1}^\pm | e^{\mathcal{L}_S \Delta t} | \sigma_j^\pm \rangle = \text{Tr} \{ | \sigma_{j+1}^- \rangle \langle \sigma_{j+1}^+ | e^{\mathcal{L}_S \Delta t} | \sigma_j^+ \rangle \langle \sigma_j^- | \} \quad (12)$$

In the iterative scheme of Makri and Makarov [11, 12] the Hamiltonian system dynamics enters only in the combination $\langle \sigma_{j+1}^+ | e^{-iH_S \Delta t/\hbar} | \sigma_j^+ \rangle \cdot \langle \sigma_j^- | e^{iH_S \Delta t/\hbar} | \sigma_{j+1}^- \rangle$ which equals exactly $\langle \sigma_j^\pm | e^{\mathcal{L}_S \Delta t} | \sigma_{j-1}^\pm \rangle$ for the special case of Hamiltonian system dynamics, i.e. $\mathcal{L}_S = -(i/\hbar)[H_S, \cdot]$. Thus, for our more general case of Liouvillian system dynamics, i.e. $\mathcal{L}_S \neq -(i/\hbar)[H_S, \cdot]$ we can still construct an iterative scheme when restricting to a finite memory time following the procedure as outlined by Makri and Makarov [11, 12]. Thus, (11) readily allows to extend the quasi-adiabatic path integral approach to treat numerical exactly the influence of environmental fluctuations on any system dynamics which can be cast into a Liouvillian equation (4). This is our first result.

Equipped with two methods we study the influence of environmental fluctuations on an already dephasing quantum system. We determine the decoherence rate Γ of a symmetric quantum two-level system (1) by fitting $f(t) = \cos \Delta t e^{-\Gamma t}$ to the numerical results for the $\hat{\sigma}_z$ expectation value employing our hybrid-QUAPI scheme. The inset of Fig.1 shows the decoherence rate Γ versus the dephasing rate γ_D . We use $k_B T = \Delta$ and $\omega_c = 5\Delta$ and find that Γ increases monotonically with γ_D for all studied system-bath couplings γ_z . In order to separate out the contribution from the system-bath coupling Fig.1 plots the difference of decoherence rate and γ_D normalized by the RESPET result $\Gamma_p(\gamma_D = 0)$ for vanishing γ_D . For small γ_z and γ_D we observe that the decoherence rate is simply the sum of the dephasing rate γ_D and the weak coupling result for the longitudinal fluctuations. With increasing system-bath coupling γ_z the ratio of $(\Gamma - \gamma_D)/\Gamma_p(\gamma_D = 0)$ decreases as higher order effects set in. Surprisingly, for all system-bath couplings the ratio decreases also for increasing γ_D . Thus, increasing a phenomenological dephasing suppresses the decohering effect of longitudinal fluctuations. In contrast, the relaxation rate (determined by fitting an exponential decay to

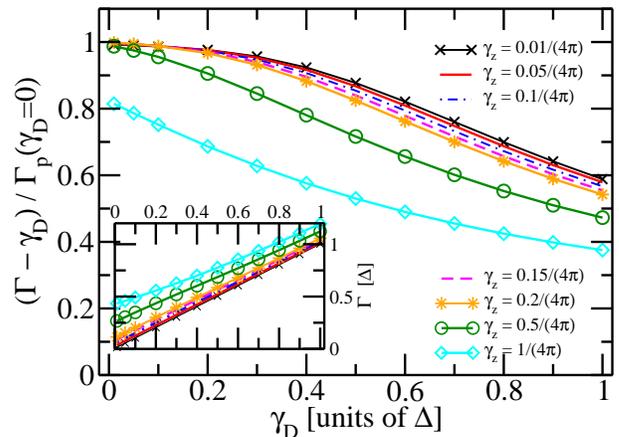


FIG. 1. The difference of decoherence rate and γ_D normalized by $\Gamma_p(\gamma_D = 0)$ is plotted versus dephasing rate γ_D for various system-bath couplings γ_z for $k_B T = \Delta$ and $\omega_{c,z} = 5\Delta$. The inset plots the decoherence rate directly.

$\langle \hat{\sigma}_x \rangle(t)$ (data not shown) is constant within the numerical accuracy of the hybrid-QUAPI scheme.

The observed suppression of decoherence is studied in more detail in Fig.2. The ratio $(\Gamma - \gamma_D)/\Gamma_p(\gamma_D = 0)$ is plotted versus γ_D for a rather strong system-bath coupling $\gamma_z = 0.2/(4\pi)$ at five temperatures, i.e. $k_B T = 0.01\Delta$ (red crosses), 0.05Δ (green squares), 0.2Δ (blue squares), 0.5Δ (magenta squares) and Δ (orange squares). Data for the two temperatures $k_B T = 0.01\Delta$ and 0.05Δ coincide which, thus, represents the low temperature limit. Additionally, data is shown for a weak system-bath coupling $\gamma_z = 0.01/(4\pi)$ at three temperatures, i.e. $k_B T = 0.2\Delta$ (blue circles), 0.5Δ (magenta circles) and Δ (orange circles). Perturbative results following (7) are given as lines. Since we restricted the perturbative calculation to lowest order the ratio $(\Gamma - \gamma_D)/\Gamma_p(\gamma_D = 0)$ (with Γ determined by RESPET) does not depend on the system bath coupling γ_z . The QUAPI data shows small deviations between the $\gamma_z = 0.01/(4\pi)$ and $\gamma_z = 0.2/(4\pi)$ for temperatures $k_B T = 0.2\Delta$ and Δ but not for $k_B T = 0.5\Delta$ (within the accuracy of the data). This lack of γ_z dependence points towards a lowest order effect in the system-bath coupling as determined by the extended RESPET. Surprisingly, however, QUAPI results differ substantially from the RESPET results except for very small dephasing γ_D . Furthermore, RESPET shows with increasing γ_D at first a suppression of decoherence and then an increase. The minimum shifts towards larger γ_D with increasing temperature and, thus, is only visible for $k_B T = 0.2\Delta$ in Fig. 2. In contrast, the correct behaviour as determined by QUAPI shows at first a decrease of decoherence which seems to level off for larger γ_D .

To investigate this further Fig.3 (upper graph) plots the data for $\gamma_z = 0.01/(4\pi)$ and $k_B T = 0.2\Delta$ and $k_B T = 0.5\Delta$ for an extended range of γ_D . Therein, the γ_D dependence is very weak for $\gamma_D \gtrsim 2\Delta$. The

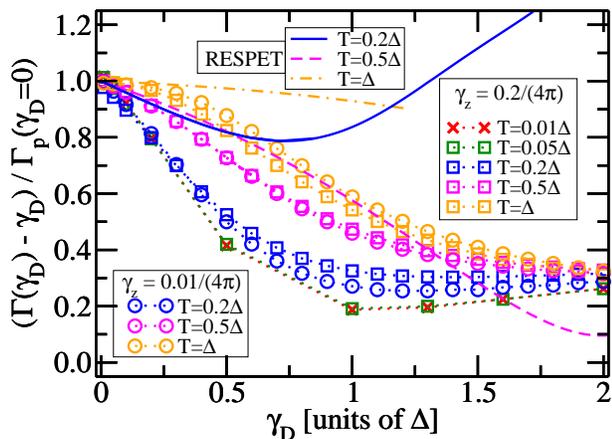


FIG. 2. The difference of decoherence rate and γ_D normalized by $\Gamma_p(\gamma_D = 0)$ is plotted versus dephasing rate γ_D for five temperatures and two system-bath couplings and compared to the RESPET result (7).

$k_B T = 0.2\Delta$ data exhibits a shallow minimum but the $k_B T = 0.5\Delta$ simply levels off.

In total, we find that a perturbative approach, which is standard to treat weak system-bath coupling successfully, fails when the system dynamics is not Hamiltonian but follows an Liouvillian dynamics. In detail, we studied a quantum two-level system with phenomenological dephasing. One might argue heuristically that a large dephasing rate γ_D is the result of strong environmental noise and further that such a strongly coupled environment even invalidates a perturbative treatment of an additional independent noise source even when this noise is weak, i.e. its system-bath coupling is small, i.e. $\gamma_z \ll 1$. Then, discrepancy between QUAPI and RESPET should occur only for large γ_D . Fig. 3 (lower graph) plots the data for $\gamma_z = 0.01/(4\pi)$ and $k_B T = 0.2\Delta$, 0.5Δ and Δ with a focus on small dephasing rate γ_D . As expected RESPET and QUAPI results agree for vanishing phenomenological dephasing. Sizeable differences, however, already occur for $\gamma_D \gtrsim 0.1\Delta$ with stronger deviations at lower temperatures.

We have developed an effective treatment to determine the non-equilibrium dynamics of a dephasing quantum system subjected to additional environmental fluctuations. The dynamics of the dephasing quantum system is described by an phenomenological Liouville von Neumann equation and the coupling to the additional environment is treated within a system bath approach. To treat this system-bath coupling we have extended the quasi-adiabatic path integral scheme to allow the (numerical) exact treatment of a system-bath problem when

the system dynamics is determined by a Liouville von Neumann equation rather than a Hamiltonian. We have then studied the dynamics of the dephasing quantum two-level system.

We observe a suppression of the contribution from the environment to the decoherence rate with increasing de-

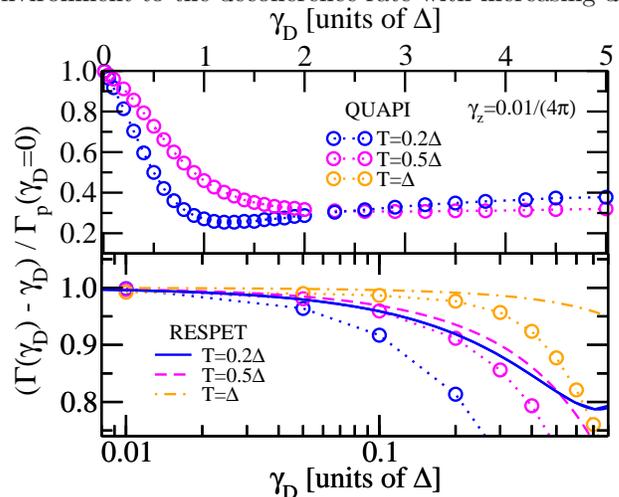


FIG. 3. The difference of decoherence rate and γ_D normalized by $\Gamma_p(\gamma_D = 0)$ is plotted versus dephasing rate γ_D for two temperatures and an extend γ_D range (upper graph) and for three temperatures with a focus on small γ_D (lower graph).

phasing. Thus, dephasing suppresses the effects of additional environmental fluctuations. We then additionally determined the dynamics treating the system-bath coupling perturbatively. Surprisingly, we find strong quantitative and qualitative deviations between the perturbative and the (numerically) exact results for the environmental influences even for system-bath couplings which are normally considered to justify a perturbative treatment. This shows that the interplay of dephasing and additional environmental noise gives rise to peculiar non-perturbative effects.

These results are important to evaluate the dynamics, for example, of qubits, of photosynthetic complexes and also of quantum transport experiments. In all these case multiple noise sources influence the quantum system of interest. In charge and also flux qubits pure dephasing noise is notoriously difficult to characterize and, thus, typically treated phenomenologically whereas the other noise sources are most often perturbatively treated. Our results show that these perturbative evaluations should be used with utmost care.

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