Pure Dephasing of Light-Matter Systems in the Ultrastrong and **Deep-Strong Coupling Regimes**

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Pure dephasing originates from the nondissipative information exchange between quantum systems and environments, and plays a key role in both spectroscopy and quantum information technology. Often pure dephasing constitutes the main mechanism of decay of quantum correlations. Here we investigate how pure dephasing of one of the components of a hybrid quantum system affects the dephasing rate of the system transitions. We find that, in turn, the interaction, in the case of a light-matter system, can significantly affect the form of the stochastic perturbation describing the dephasing of a subsystem, depending on the adopted gauge. Neglecting this issue can lead to wrong and unphysical results when the interaction becomes comparable to the bare resonance frequencies of subsystems, which correspond to the ultrastrong and deepstrong coupling regimes. We present results for two prototypical models of cavity quantum electrodynamics: the quantum Rabi and the Hopfield model.

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Introduction.-In reality, there is no perfectly isolated quantum system. For example, the coupling of a radiating atom with the infinitely many modes of a free electromagnetic field results in decoherence and spontaneous emission. Such interaction determines an energy relaxation time T_1 associated to a given optical transition. If the population of an excited state decays, so does the polarization too, which results in decoherence. In the presence of only energy relaxation mechanisms, such transverse relaxation time is $T_2 = 2T_1$ [1,2]. However, quantum systems, displaying optical transitions, do not only interact with an electromagnetic field, but can be affected by additional dephasing mechanisms inducing the decay of the dipole coherence without changing the populations of the systems. These pure dephasing effects can originate from fluctuations in the environmental fields affecting the phases of the emitter wave functions; see, e.g., Refs. [3-8]. In general, the phase (transverse) relaxation time is most often shorter than twice the energy relaxation time: $T_2 \leq 2T_1$. In optical spectroscopy, the full width at half maximum (FWHM) of homogeneous broadening corresponds to $2/T_2$.

It is well known that decoherence tends to destroy quantum coherence and quantum correlations [9,10]. It is known that this mechanism becomes faster with the increase of the size of a quantum system [11]. This explains the absence of quantum superpositions in the macroscopic world [12]. Decoherence can, thus, strongly affect and limit quantum information processing (QIP) [13,14]. Depending on the specific environment, mechanisms to protect qubits from dephasing have been proposed (see, e.g., Refs. [14–18]).

Devices for QIP, secure communication, and highprecision sensing were implemented combining different systems ranging from photons, atoms, and spins to mesoscopic superconducting and nanomechanical structures. Complementary functionalities of these hybrid quantum systems can be essential for the development of new quantum technologies [19-21]. Understanding how decoherence of one or more subsystems can affect the performance of the whole system is an interesting problem, relevant for improving the performance of quantum devices [22,23].



FIG. 1. Pictorial representation of a two-level system interacting with a single-mode cavity field, when both subsystems are affected by pure dephasing.

Cavity [10] and circuit [24,25] quantum electrodynamics (QED) systems are among the most studied hybrid quantum systems. They are playing a key role in quantum optics and in the development of new quantum technologies [26–29]. Pure dephasing can significantly affect the performance of these systems, not necessarily in a negative way. For example, it has been shown that pure dephasing is a promising resource for solid-state emitters, since it can improve the performance of nanophotonic devices, such as single-photon sources and nanolasers [30].

Decoherence effects in hybrid quantum systems are often introduced by using the standard quantum optics master equation, where the coupling of a multicomponent system with the environment is introduced by neglecting the interaction between the subsystems. When such interaction is not negligible compared with the bare transition frequencies of the components, as in the light-matter ultrastrong coupling (USC) or deep-strong coupling (DSC) regimes [31,32], this approximation can give rise to unphysical results. These regimes can give rise to new physical effects and applications (see, e.g., [33–39]), and they also challenge our understanding of fundamental aspects of cavity QED, like a proper definition of subsystems, their quantum measurements, and the structure of the light-matter ground states, leading also to gauge ambiguities [40–46,48].

A master equation method for cavity QED systems, describing both losses and pure dephasing, and taking into account light-matter interaction, has been proposed in Refs. [22,23]. However, these models, as well as previous ones, consider perturbation Hamiltonians for pure dephasing which are not affected by light-matter interaction. Here we show that the interaction between light and matter can significantly affect the form of a stochastic perturbation describing the dephasing of one of the components. We find that neglecting this issue can lead to wrong and unphysical results in both USC and DSC regimes. We present results for two prototypical models of cavity QED: the quantum Rabi model (QRM) and the Hopfield model. However, the approach here considered can also be applied to describe more complex light-matter systems.

Quantum Rabi model.—Pure dephasing effects on both the qubit and electromagnetic field can be described by introducing two zero-mean stochastic functions $f_c(t)$, $f_q(t)$ modulating their resonance frequency (Fig. 1). The perturbation Hamiltonian can be written as

$$\hat{\mathcal{V}}_{dep} = f_c(t)\hat{a}^{\dagger}\hat{a} + f_q(t)\hat{\sigma}_z.$$
 (1)

By expanding $\hat{\mathcal{V}}_{dep}$ in the basis of the eigenstates of the total system Hamiltonian, a master equation describing the effects of qubit dephasing on the system dynamics can be obtained [22]. For the sake of simplicity, we consider stochastic functions with a low-frequency spectral density (with respect to the relevant transition frequencies of the

system). The resulting master equation can be written as $(\hbar = 1)$ [47]

$$\frac{d}{dt}\hat{\rho}(t) = -i[\hat{H}_s,\hat{\rho}] + \frac{\gamma_{\phi}^{(q)}}{2}\mathcal{D}[\hat{\Phi}]\hat{\rho} + \frac{\gamma_{\phi}^{(c)}}{2}\mathcal{D}[\hat{\Xi}]\hat{\rho}, \quad (2)$$

where \hat{H}_s is the Hamiltonian of the total system and

$$\mathcal{D}[\hat{O}]\hat{\rho} = \frac{1}{2} (2\hat{O}\,\hat{\rho}\,\hat{O}^{\dagger} - \hat{\rho}\hat{O}^{\dagger}\hat{O} - O^{\dagger}\hat{O}\,\hat{\rho}) \qquad (3)$$

is the Lindbladian superoperator, while $\hat{\Phi} = \sum_{j} \sigma_{z}^{jj} |j\rangle \langle j|$ and $\hat{\Xi} = \sum_{j} \langle j | \hat{a}^{\dagger} \hat{a} | j \rangle |j\rangle \langle j|$, with $|j\rangle$ being the eigenstates of \hat{H}_{s} , and $\sigma_{z}^{jj} = \langle j | \hat{\sigma}_{z} | j \rangle$. The bare dephasing rates $\gamma_{\phi}^{x} = 2S_{f}(0)$ are determined by the low-frequency spectral densities $S_{f}^{(x)}(\omega)$ of $f_{x}(t)$, with x = q, *c*. Additional dephasing terms can appear, when the spectral density functions $S_{f}(\omega)$ are not negligible at the transition frequencies of the system (see the Supplemental Material [49]).

We apply the above procedure to the simplest model of cavity QED, i.e., the QRM. Its Hamiltonian in the dipole gauge can be written as $\hat{\mathcal{H}}_D = \hat{\mathcal{H}}_{ph} + \hat{\mathcal{H}}_q + \hat{\mathcal{V}}_D$, where $\hat{\mathcal{H}}_q = \omega_q \hat{\sigma}_z/2$, and the free field Hamiltonian is $\hat{\mathcal{H}}_{\rm ph} =$ $\omega_c \hat{a}^{\dagger} \hat{a}$, where $\hat{\sigma}_i (j = x, y, z)$ are the Pauli operators, and \hat{a} and \hat{a}^{\dagger} are the photon destruction and creation operators. Neglecting the constant term $\eta^2 \omega_c$, the interaction term can be written as $\hat{\mathcal{V}}_D = -i\eta\omega_c(\hat{a} - \hat{a}^{\dagger})\hat{\sigma}_x$, where η is the normalized qubit-cavity coupling strength. It has been shown that the standard quantum Rabi Hamiltonian in the Coulomb gauge violates gauge invariance [40]. The correct Coulomb-gauge quantum Rabi Hamiltonian [41] can be obtained by writing the sum of the free field and matter Hamiltonians and then by applying a suitable unitary transformation (generalized minimal coupling) to the free matter Hamiltonian [41]: $\hat{\mathcal{H}}_C = \hat{\mathcal{H}}_{ph} + \hat{\mathcal{U}}\hat{\mathcal{H}}_q\hat{\mathcal{U}}^{\dagger}$, where $\hat{\mathcal{U}} = \exp[i\hat{\mathcal{A}}\hat{\sigma}_{x}]$, with $\hat{\mathcal{A}} = \eta(\hat{a} + \hat{a}^{\dagger})$. We obtain

$$\hat{\mathcal{H}}_{C} = \hat{\mathcal{H}}_{\rm ph} + \frac{\omega_{q}}{2} [\hat{\sigma}_{z} \cos(2\hat{\mathcal{A}}) + \hat{\sigma}_{y} \sin(2\hat{\mathcal{A}})].$$
(4)

The dipole and Coulomb gauge Hamiltonians are related by the unitary gauge transformation $\hat{\mathcal{H}}_D = \hat{\mathcal{U}}^{\dagger} \hat{\mathcal{H}}_C \hat{\mathcal{U}}$; thus the dipole gauge Hamiltonian can also be obtained by applying a generalized minimal coupling replacement to the free field Hamiltonian: $\hat{\mathcal{H}}_D = \hat{\mathcal{U}}^{\dagger} \hat{\mathcal{H}}_{ph} \hat{\mathcal{U}} + \hat{\mathcal{H}}_a$.

Following the standard approach, pure dephasing effects can be directly introduced by using Eqs. (1) and (2), which provides gauge invariant expectation values, as can be easily shown [50]. However, this is not sufficient to ensure that the obtained results are physically correct. However, we will show below that this naive approach can provide incorrect and/or gauge dependent results, especially when the light-matter interaction strength is very strong. Actually, light-matter interaction can modify the form of quantum operators describing physical observables, and these changes are usually gauge dependent [51]. For example, in the Coulomb gauge the form of the physical momentum of the particle is affected by light-matter interaction, while in the dipole gauge it is interaction independent. On the contrary, the dipole gauge affects the definition of the field momentum. As a consequence, in this gauge, the canonical momentum is no more proportional to the electric field operator. We may thus expect that the form of operators describing pure dephasing shall be modified by light-matter interaction too. In order to obtain correct descriptions of pure dephasing effects, as well as gauge-invariant results, in the presence of light-matter interactions one has to apply the generalized minimal coupling replacements considered above to pure dephasing perturbations in Eq. (1) too. In the Coulomb and dipole gauge, respectively, we obtain

$$\hat{\mathcal{V}}_{\phi}^{C} = f_{q}(t)\hat{\sigma}_{z}^{C} + f_{c}(t)\hat{a}^{\dagger}\hat{a}, \qquad (5)$$

$$\hat{\mathcal{V}}^{D}_{\phi} = f_q(t)\hat{\sigma}_z + f_c(t)\hat{a}^{\dagger}_D\hat{a}_D, \qquad (6)$$

where $\hat{\sigma}_z^C = \hat{\mathcal{U}}\hat{\sigma}_z\hat{\mathcal{U}}^{\dagger}$ and $\hat{a}_D = \hat{\mathcal{U}}^{\dagger}\hat{a}\hat{\mathcal{U}} = \hat{a} + i\eta\hat{\sigma}_x$ are atomic and field operators modified by the light-matter interaction in the Coulomb and dipole gauge, respectively.

In the following, we label the QRM states by generalizing the notation of the Jaynes-Cummings (JC) model. In particular, $|\tilde{0}\rangle$ denotes the ground state, and $|\tilde{n}_{\pm}\rangle$ the states that tend to the JC states $|n_{\pm}\rangle$, when the coupling vanishes. Moreover, we use not-primed (primed) states to indicate the Coulomb (dipole) gauge states. As an example we analyze pure dephasing effects on the two lowest transitions in the QRM: $\alpha_{\pm} \equiv (\tilde{1}_{\pm}, \tilde{0})$, and considering only qubit pure dephasing [$f_c(t) = 0$]. In the interaction picture, from Eq. (2), we obtain [49]

$$\dot{\tilde{\rho}}_{\alpha'_{\pm}}(t) = -(\gamma_{\phi}^{\alpha'_{\pm}}/2)\tilde{\rho}_{\alpha'_{\pm}}(t),$$
(7)

with

$$\gamma_{\phi}^{\alpha'_{\pm}} = \frac{\gamma_{\phi}^{(q)}}{2} \left| \sigma_{z}^{\tilde{1}'_{\pm},\tilde{1}'_{\pm}} - \sigma_{z}^{\tilde{0}',\tilde{0}'} \right|^{2}.$$
 (8)

We observe that the obtained dephasing rates are gauge invariant $(\gamma_{\phi}^{\alpha'_{\pm}} = \gamma_{\phi}^{\alpha_{\pm}})$, because the expectation values are unitary invariant, when transforming both operator and states: $\gamma_{\phi}^{\alpha_{\pm}} = \gamma_{\phi}^{(q)} |\sigma_z^{C,\tilde{1}_{\pm},\tilde{1}_{\pm}} - \sigma_z^{C,\tilde{0},\tilde{0}}|^2/2$. Figure 2(a) displays the normalized pure dephasing rate $\gamma_{\tilde{1}'_{\pm},\tilde{0}}/\gamma_{\phi}^0$ for the two lowest energy transitions, considering a small qubit-cavity detuning $\delta = 3 \times 10^{-3}$ and in the case of only qubit pure dephasing. In the limit of negligible coupling strength, where $|\tilde{1}'_{+}\rangle \rightarrow |e, 0\rangle$ and $|\tilde{1}'_{-}\rangle \rightarrow |g, 1\rangle$, the standard results



FIG. 2. Quantum Rabi model. Normalized pure dephasing rate for the two lowest energy transitions, for a small qubit-cavity detuning $\delta = 3 \times 10^{-3}$ and considering only the qubit pure dephasing. (a) Correct gauge-invariant versus (b) wrong Coulomb gauge results.

are recovered, and only $(\tilde{1}'_{-}, \tilde{0}')$ is affected by the qubit pure dephasing. When the coupling becomes comparable to the detuning, as expected, pure dephasing is shared among the two transitions, since the energy eigenstates $|\tilde{1}'_{+}\rangle$ tend to become an equally weighted superposition of $|e, 0\rangle$ and $|g,1\rangle$. For the normalized coupling strengths $\eta > 0.1$ (the USC regime), pure dephasing becomes less effective for the transition $(\tilde{1}'_{-}, \tilde{0})$, until at stronger couplings (the DSC regime), both the transitions tend to become dephasing free. This behavior reflects the fact that, when the coupling rate is larger than the bare qubit frequency, a fluctuation at the qubit resonance frequency can have a very low impact on the dressed-state energies. On the contrary, Fig. 2(b) shows a wrong large pure dephasing rate for the lowest energy transition. Analogous calculations can be carried out for the case of cavity pure dephasing [49].

Hopfield model.—A similar analysis can be carried out for polaritons. We consider the simplest version of the Hopfield model [52], describing the interaction of a singlemode electromagnetic resonator with a bosonic matter field (with the bosonic annihilation \hat{b} and creation \hat{b}^{\dagger} operators) modeling some kind of collective matter excitations. The system Hamiltonian in the dipole gauge reads as

$$\hat{H}_{\rm D} = \hat{H}_0 + i\lambda\omega_c(\hat{a}^{\dagger} - \hat{a})(\hat{b} + \hat{b}^{\dagger}) + \omega_c\lambda^2(\hat{b} + \hat{b}^{\dagger})^2, \quad (9)$$

where $\hat{H}_0 = \omega_c \hat{a}^{\dagger} \hat{a} + \omega_x \hat{b}^{\dagger} \hat{b}$, and λ is the normalized coupling strength. An equivalent model can be obtained in the Coulomb gauge [53]:

$$\hat{H}_{C} = \hat{H}_{0} - i\omega_{x}\lambda(\hat{b}^{\dagger} - \hat{b})(\hat{a}^{\dagger} + \hat{a}) + \mathcal{D}(\hat{a}^{\dagger} + \hat{a})^{2}, \qquad (10)$$

where $\mathcal{D} = \omega_x \lambda^2$. These two Hamiltonians can be directly obtained by generalized minimal coupling replacements: $H_C = \omega_c \hat{a}^{\dagger} \hat{a} + \omega_x \hat{U} \hat{b}^{\dagger} \hat{b} \hat{U}^{\dagger}$ and $H_D = \omega_c \hat{U}^{\dagger} \hat{a}^{\dagger} \hat{a} \hat{U} + \omega_x \hat{b}^{\dagger} \hat{b}$, where $\hat{U} = \exp[i\lambda(\hat{a} + \hat{a}^{\dagger})(\hat{b} + \hat{b}^{\dagger})]$. As is well known, the interaction gives rise to polaritonic resonances, which results from the mixing of the two bosonic modes. It is possible to diagonalize the system expressing the photon and exciton operators in terms of polaritonic (bosonic) operators [52]. For $\mu = 1$, 2 (lower and upper polariton, respectively), we have

$$\hat{y} = \sum_{\mu=1}^{2} \left(U_{y}^{\mu} \hat{P}_{\mu} - V_{y}^{\mu} \hat{P}_{\mu}^{\dagger} \right), \qquad (\hat{y} = \hat{a}, \hat{b}).$$
(11)

The diagonalization procedure determines both polariton eigenfrequencies Ω_{μ} , which are gauge invariant, and the Hopfield coefficients, which are gauge dependent. As a consequence, also the polariton operators are gauge dependent. We use primed operators and coefficients for the dipole gauge.

By neglecting issues related to the light-matter interaction, dephasing effects can be modeled by introducing the perturbation Hamiltonian

$$\hat{V}_{dep}(t) = f_c(t)\hat{a}^{\dagger}\hat{a} + f_x(t)\hat{b}^{\dagger}\hat{b}, \qquad (12)$$

describing the stochastic fluctuation of the resonance frequencies of the components. Following the reasoning of the previous section, when including the light-matter interaction, it turns out that Eq. (12) is incorrect, and its corrected form is gauge dependent:

$$\hat{V}_{dep}^{D}(t) = f_{c}(t)\hat{a}_{D}^{\dagger}\hat{a}_{D} + f_{x}(t)\hat{b}^{\dagger}\hat{b}, \qquad (13)$$

$$\hat{V}_{dep}^{C}(t) = f_{c}(t)\hat{a}^{\dagger}\hat{a} + f_{x}(t)\hat{b}_{C}^{\dagger}\hat{b}_{C}, \qquad (14)$$

where $\hat{a}_D = \hat{T} \hat{a} \hat{T}^{\dagger} = \hat{a} + i\lambda(\hat{b} + \hat{b}^{\dagger})$ and $\hat{b}_C = \hat{T}^{\dagger}\hat{b} \hat{T} = \hat{b} - i\lambda(\hat{a} + \hat{a}^{\dagger})$. Notice that here \hat{a}_D (\hat{b}_C) is the *physical* photonic (excitonic) annihilation operator in the dipole (Coulomb) gauge. By *physical*, we mean the operators that describe the annihilation of the physical quanta of the fields [53]. The polariton pure dephasing rates can be obtained by expanding Eqs. (13) and (14) in terms of the polariton operators, and then applying the standard master equation method to obtain the Lindbladian terms, in analogy with the results of the previous section [49]. From the obtained master equation, the equations of motion for the mean values of the polariton operators are $\partial_t \langle \hat{P}_\mu \rangle = (-i\Omega_\mu - \gamma_{d\nu}^\mu/2) \langle \hat{P}_\mu \rangle$, where

$$\gamma^{\mu}_{\phi} = \gamma^{0}_{c} (|U^{\mu}_{a}|^{2} + |V^{\mu}_{a}|^{2}) + \gamma^{0}_{x} (|U^{\mu\prime}_{b}|^{2} + |V^{\mu\prime}_{b}|^{2}).$$
(15)

This result can be very different from what could be obtained starting from Eq. (12) and ignoring the modifications in the perturbation Hamiltonian induced by the light-matter interaction. Figure 3(a) shows the normalized pure dephasing rates for the two polariton modes $(\gamma_{\phi}^{\mu}/\gamma_{x}^{0})$, for the case of the zero photonic noise $(\gamma_{c}^{0} = 0)$, and



FIG. 3. Hopfield model. Normalized pure dephasing rate of the lower and upper polaritons, originating from exciton dephasing, versus the normalized coupling strength, obtained for different exciton-cavity detunings, and considering only the matter pure dephasing.

considering three different values of the exciton-cavity detuning δ . We observe that, at large coupling rates, independently of the detuning, the lower polariton dephasing rate tends to zero. This effect is a direct consequence of the fact that the lower polariton resonance frequency tends rapidly to zero for $\lambda \to \infty$ [see Fig. 4(c)], independently of the detuning. This implies that any small fluctuation of the resonance frequencies of the components does not induce fluctuations and, hence, dephasing in the polariton mode. For comparison, Figs. 4(a)–4(b) display the wrong result $\gamma_{\phi}^{\mu}/\gamma_{x}^{0} = |U_{b}^{\mu}|^{2} + |V_{b}^{\mu}|^{2}$, obtained by neglecting the changes



FIG. 4. Hopfield model. Wrong (see text) normalized pure dephasing rates of the lower and upper polaritons, originating from exciton dephasing, versus the normalized coupling strength, obtained for two different exciton-cavity detunings, and considering only the matter pure dephasing (a),(b). (c) Frequencies of the two polariton modes for a qubit-cavity detuning $\delta/\omega_c = 3 \times 10^{-3}$.

of the form of subsystems-observables, which can be induced by the interaction, as calculated for two different detunings. Evident differences emerge when entering the USC regime with $\lambda \sim 0.1$. Moreover, at larger coupling rates, in the DSC regime, the behavior of the lower and upper polaritons is clearly inverted.

Conclusions.-We have shown how to calculate correctly the pure dephasing rate in cavity QED systems, considering two prototypical models: the QRM and the Hopfield model. In the latter model, we found that pure dephasing effects in the lower polariton branch tend to be reduced in the USC regime, and tend to get suppressed increasing further the coupling [see Fig. 3(a)]. On the contrary, the influence of pure dephasing increases at increasing coupling strengths for upper polaritons. We hope that these results will stimulate experimental tests for various polariton systems, where these interaction regimes have been observed [31]. In a number of experiments, it was observed that the upper polariton clearly displays a larger line broadening with respect to the lower one [54–57] in agreement with the results presented here. However, since in these systems different broadening mechanisms enter into play, further investigations are required. The approach shown here can be applied to more complex light-matter systems and/or to full quantum models of pure dephasing [22,58]. The general lesson is that when the light-matter interaction rate becomes comparable to the bare resonance frequencies of the relevant bare transitions of the system components, the generalized minimal coupling replacements introducing the light-matter interaction have to be also applied to any perturbation affecting the matter or light subsystems.

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derivation of pure dephasing terms on both quantum Rabi and Hopfield models. Analytical derivation of dephasing rates transitions.

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