Memory effects in spontaneous emission processes

Arne L. Grimsmo,^{1,2,*} Asle H. Vaskinn,^{1,†} Per K. Rekdal,^{3,‡} and Bo-Sture K. Skagerstam^{1,4,§}

¹Department of Physics, The Norwegian University of Science and Technology, N-7491 Trondheim, Norway

²Department of Physics, The University of Auckland, Private Bag 92019, Auckland, New Zealand

³Molde University College, P.O. Box 2110, N-6402 Molde, Norway

⁴Centre for Advanced Study (CAS), Drammensveien 78, N-0271 Oslo, Norway

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We consider a quantum-mechanical analysis of spontaneous emission in terms of an effective two-level system with a vacuum decay rate Γ_0 and transition angular frequency ω_A . Our analysis is in principle exact, even though presented as a numerical solution of the time evolution including memory effects. The results so obtained are confronted with previous discussions in the literature. In terms of the *dimensionless* lifetime $\tau = t\Gamma_0$ of spontaneous emission, we obtain deviations from exponential decay of the form $O(1/\tau)$ for the decay amplitude as well as the previously obtained asymptotic behaviors of the form $O(1/\tau^2)$ or $O(1/\tau \ln^2 \tau)$ for $\tau \gg 1$. The actual asymptotic behavior depends on the adopted regularization procedure as well as on the physical parameters at hand. We show that for any reasonable range of τ and for a sufficiently large value of the required angular frequency cutoff ω_c of the electromagnetic fluctuations, i.e., $\omega_c \gg \omega_A$, one obtains either a $O(1/\tau^2)$ or a $O(1/\tau^2)$ dependence. In the presence of physical boundaries, which can change the decay rate with many orders of magnitude, the conclusions remains the same after a suitable rescaling of parameters.

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I. INTRODUCTION

The development concerning the manipulation of single atoms and their interaction with the electromagnetic field has reached an impressive state of art in recent years (see, e.g., Refs. [1-4]). Experimental studies have, e.g., shown that artificial atoms can lead to a Lamb shift of the order of a few percent of the typical emission line [5]. Rather old issues concerning the necessary deviations from exponential decay (see, e.g., Refs. [6-22]) may therefore be confronted with our current theoretical and experimental understanding of decaying quantum systems. Concerning experimental studies of deviations from the conventional exponential decay we notice in particular the study of decaying τ leptons [23], the observed deviations in quantum-mechanical tunneling processes [24], and the power-law behavior of the decay at times scales larger than twenty lifetimes in dissolved organic materials [25]. In nuclear physics the decay of thorium has also been suggested as a potential target for large-time deviations [26].

In the context of elementary-particle physics gaugeinvariant definitions of observable quantities are of central importance and the definition of the decay width of an unstable particle is highly nontrivial (see, e.g., [27]) in this respect. In studies of a possible proton decay in nature the time dependence of decaying systems may also play an important role [28]. Recently, it has also been speculated that a nonexponential decay of slowly decaying ¹⁴C nuclei may be of practical importance in radioisotope calibration methods [29].

In the presence of material bodies with nontrivial dispersive properties, it has been observed that the decay time of atoms can be changed by many orders of magnitude (see, e.g., Refs. [30-34]), which also has been observed in the laboratory [35-40] in the context of atomic chips. A natural question to be considered could then be to what extent deviations from an exponential decay can be observed. This issue has been addressed in great detail in the literature in terms of a model of a two-level system interacting with a continuous number of degrees of freedom of the electromagnetic field [41-45]. Such a model appears to be of relevance in the analysis of the stability properties of atomic chips when applied to hyperfine transitions and, in addition, taking dissipative effects into account [30-34]. A related and exactly solvable model of a two-level system, which also leads to deviations from exponential decay, has been discussed in the context of a model of qubit dephasing (see, e.g., [46-48]).

Here we will investigate possible deviations from exponential decay for, in particular, small and large τ , following the work of Refs. [41,43,44]. The role of the physical parameters at hand will be made explicit and transparent. Analytical as well as numerical methods will be employed and will be shown to lead to a consistent picture of the small- and largetime deviations from a constant decay rate. The analytical properties of the Laplace-transformed decay amplitude plays an essential role in the analysis. It is, of course, well known that, under various assumptions, one obtains in general a decay probability which is larger than the conventional exponential decay at sufficiently small decay times τ (for a review see, e.g., [16]) as well as a typical power-law behavior in $1/\tau$ for sufficiently large decay times τ (see, e.g., Refs. [15,49]). In exactly solvable models one finds, e.g., a $1/\tau^2$ (see, e.g., Refs. [14,18]) or a $1/\tau^{3/2}$ (see, e.g., Refs. [7,19,21]) asymptotic behavior in the decay amplitude of an unstable system. In the model we consider, we obtain $O(1/\tau)$ for the decay amplitude as well as the previously obtained asymptotic behaviors of the form $O(1/\tau^2)$ or $O(1/\tau \ln^2 \tau)$ for $\tau \gg 1$ [41,43,44].

^{*}arne.grimsmo@ntnu.no

[†]asle.vaskinn@ntnu.no

[‡]per.k.rekdal@himolde.no

[§]bo-sture.skagerstam@ntnu.no

The paper is organized as follows. In the next section we outline the theoretical framework and explicitly verify unitarity of the two-level system. A regularized version of the integral equation for the decay amplitude is derived in Sec. III A and exact short-time and long-time expansions are obtained in Sec. III B. Laplace techniques are employed in Secs. III C and III D in order to compare with previous asymptotic expansion results as given in the literature and the important role of the regularization procedure made use of is emphasized. Final remarks are given in Sec. IV and various technical details of some of the calculations are, for the readers convenience, summarized in Appendixes A, B, and C.

II. GENERAL THEORY

Let us consider a neutral atom at a fixed position \mathbf{r}_A . In order to be specific, we will explicitly consider hyper-fine interactions but the results obtained can easily be rephrased in terms of electric-dipole interactions. The magnetic moment of the atom interacts with the quantized magnetic field via a conventional Zeeman coupling. The total, unrenormalized, Hamiltonian has then the standard form

$$H = \sum_{\alpha} \hbar \omega_{\alpha} |\alpha\rangle \langle \alpha| + \int d^3 r \int_0^\infty d\omega \hbar \omega \, \hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}(\mathbf{r}, \omega) + H', \qquad (1)$$

where the effective interaction part is

$$H' = -\sum_{\alpha} \sum_{\beta} |\alpha\rangle \langle \beta | \boldsymbol{\mu}_{\alpha\beta} \cdot \mathbf{B}(\mathbf{r}_A).$$
(2)

The Hamiltonian *H* can be regarded as a low-energy effective description of a more fundamental and renormalizable theory of electromagnetic processes, i.e., quantum electrodynamics. Here $\hat{\mathbf{f}}(\mathbf{r},\omega)$ is an annihilation operator for the quantized magnetic field, $|\alpha\rangle$ denotes the atomic state, and E_{α} is the corresponding energy. We assume nondegenerate states, i.e., $E_{\alpha} \neq E_{\beta}$ for $\alpha \neq \beta$. The magnetic moment of the atom is $\boldsymbol{\mu}_{\alpha\beta} = \langle \alpha | \hat{\boldsymbol{\mu}} | \beta \rangle$, where $\hat{\boldsymbol{\mu}}$ is the magnetic-moment operator. The magnetic moment will typically be of the form $\boldsymbol{\mu}_{\alpha\beta} = g_S \langle \alpha | \mathbf{S}/\hbar | \beta \rangle e \hbar/2m_e$, where \mathbf{S}/\hbar denotes a dimensionless spin operator and g_S is an appropriate gyromagnetic factor. The quantized magnetic field $\mathbf{B}(\mathbf{r}) = \mathbf{B}^{(+)}(\mathbf{r}) + \mathbf{B}^{(-)}(\mathbf{r})$ is expressed in terms of $\mathbf{B}^{(+)}(\mathbf{r}) = \nabla \times \mathbf{A}^{(+)}(\mathbf{r})$, where $\mathbf{B}^{(-)}(\mathbf{r}) = [\mathbf{B}^{(+)}(\mathbf{r})]^{\dagger}$, and where the vector potential is given by

$$\mathbf{A}^{(+)}(\mathbf{r}) = \mu_0 \int_0^\infty d\omega' \int d^3 r' \,\omega' \sqrt{\frac{\hbar\epsilon_0}{\pi}} \epsilon_I(\mathbf{r}',\omega') \\ \times \mathbf{G}(\mathbf{r},\mathbf{r}',\omega') \cdot \hat{\mathbf{f}}(\mathbf{r}',\omega').$$
(3)

Here the imaginary part of the complex permittivity is $\epsilon_I(\mathbf{r},\omega)$ and obeys the Kramer-Kronig dispersion relations. The dyadic Green tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ is the unique solution to the Helmholtz equation

$$\vec{\nabla} \times \vec{\nabla} \times \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \epsilon(\mathbf{r}, \omega) \boldsymbol{G}(\mathbf{r}, \mathbf{r}', \omega)$$
$$= \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}, \tag{4}$$

where the arrow in $\overrightarrow{\nabla}$ denotes a derivation with respect to the first argument in the dyadic Greens function. Since

the Helmholtz equation is a linear differential equation, the associated Green's tensor can be written as a sum according to

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = \mathbf{G}^{\mathbf{0}}(\mathbf{r},\mathbf{r}',\omega) + \mathbf{G}^{\mathbf{S}}(\mathbf{r},\mathbf{r}',\omega), \tag{5}$$

where $G^0(\mathbf{r}, \mathbf{r}', \omega)$ represents the contribution of the direct waves from a pointlike radiation source in an unbounded medium, which is vacuum in our case, and $G^{S}(\mathbf{r}, \mathbf{r}', \omega)$ describes the scattering contribution of multiple reflection waves from the body under consideration. The presence of the vacuum part $G^0(\mathbf{r}, \mathbf{r}', \omega)$ in Eq. (5) will in general give rise to divergences. A regularization prescription is therefore required. For electric-dipole transitions it is well known that it is sufficient to subtract an energy shift, corresponding to the introduction of a renormalized mass, which leaves us with a logarithmic dependence of a cutoff frequency. For magnetic transitions this subtraction procedure is, however, not sufficient as we will discuss in more detail below. Our strategy is to allow for a sufficient number of subtractions to generate a logarithmic cutoff dependence also for magnetic transitions.

For reason of simplicity, we now limit our attention to a two-level atom approximation, i.e., an atom with an excited state and a ground state with the angular frequency transition $\omega_A \equiv (E_e - E_g)/\hbar > 0.$

We consider the Hamiltonian in Eq. (1) and apply the wellknown Weisskopf-Wigner theory for the transitions $e \rightarrow g$. The solution to the time-dependent Schrödinger equation in the rotating-wave approximation (RWA), i.e., applying $H' \approx H_{\text{RWA}}$, where

$$H_{\text{RWA}} = -|e\rangle\langle g|\boldsymbol{\mu}_{eg} \cdot \mathbf{B}^{(+)}(\mathbf{r}_A) + \text{H.c.}, \tag{6}$$

is then $(\omega_g \equiv E_g/\hbar \text{ and } \omega_e \equiv E_e/\hbar)$

$$\begin{split} |\psi(t)\rangle &= c_e(t)e^{-i\omega_e t}|e\rangle \otimes |0\rangle + \int d^3 r \int_0^\infty d\omega \sum_{m=1}^3 \\ &\times c_g(\mathbf{r},\omega,m|t)e^{-i(\omega+\omega_g)t}|g\rangle \otimes |\mathbf{r},\omega,m\rangle, \end{split}$$
(7)

as we have ignored any higher-order photon state than the zero-photon and one-photon state.

Here $|e\rangle$ and $|g\rangle$ are the excited state and ground state for the atom, respectively. Furthermore, $|0\rangle$ denotes the vacuum of the electromagnetic field and $|\mathbf{r}, \omega, m\rangle = \hat{f}_m^{\dagger}(\mathbf{r}, \omega) |0\rangle$ is a one-photon state. We will restrict ourselves to the initial conditions $c_e(0) = 1$ and $c_g(\mathbf{r}, \omega, m|t = 0) = 0$. More general initial conditions will require a more cumbersome analysis and will not be discussed in the present paper. The probability amplitudes $c_e(t)$ and $c_g(\mathbf{r}, \omega, m|t)$ must now obey the unitarity condition

$$|c_e(t)|^2 + \int d^3r \int_0^\infty d\omega \sum_{m=1}^3 |c_g(\mathbf{r},\omega,m|t)|^2 = 1.$$
(8)

The probability amplitude for the excited atomic state $c_e(t)$ is then determined by (see, e.g., Refs. [50,51])

$$\frac{dc_e(t)}{dt} = \int_0^t dt' K(t-t') c_e(t'),$$
(9)

where the unregularized kernel K(t) is

$$K(t) = -\frac{1}{2\pi} \int_0^\infty d\omega \, e^{-i(\omega - \omega_A)t} \Gamma(\mathbf{r}_A, \omega).$$
(10)

Here we have made use of the conventional spin-flip decay rate for spontaneous emission as given by

$$\Gamma(\mathbf{r},\omega) = \frac{2\mu_0}{\hbar} \boldsymbol{\mu}_{eg} \cdot \operatorname{Im}[\overrightarrow{\nabla} \times \mathbf{G}(\mathbf{r},\mathbf{r},\omega) \times \overleftarrow{\nabla}] \cdot \boldsymbol{\mu}_{ge}, \quad (11)$$

where the arrow in $\overleftarrow{\nabla}$ now denotes a derivation with respect to the second argument in the dyadic Greens function. Below we will also make use of the expression

$$c_{g}(\mathbf{r},\omega,m|t) = \frac{i}{c}\omega\sqrt{\frac{\mu_{0}\varepsilon_{I}(\mathbf{r},\omega)}{\pi\hbar}} \{\boldsymbol{\mu}_{ge}^{*} \cdot [\overrightarrow{\nabla} \times \mathbf{G}^{*}(\mathbf{r}_{A},\mathbf{r},\omega)]\}_{m} \times I(\omega-\omega_{A},t),$$
(12)

where we have defined the integral

$$I(\omega,t) = \int_0^t dt' e^{i\omega t'} c_e(t').$$
(13)

The useful identity

$$\sum_{m=1}^{3} \int d^{3}r \frac{\omega^{2}}{c^{2}} \varepsilon_{I}(\mathbf{r},\omega) G_{jm}(\mathbf{r}',\mathbf{r},\omega) G_{lm}(\mathbf{r}'',\mathbf{r},\omega)$$
$$= \operatorname{Im} G_{jl}(\mathbf{r}',\mathbf{r}'',\omega), \qquad (14)$$

and Eq. (12) now lead to

$$\int d^3r \int_0^\infty d\omega \sum_{m=1}^3 |c_g(\mathbf{r},\omega,m|t)|^2$$
$$= \frac{1}{2\pi} \int_0^\infty d\omega \,\Gamma(\mathbf{r}_{\mathbf{A}},\omega) |I(\omega-\omega_A,t)|^2.$$
(15)

It now follows from Eqs. (9) and (15) that

$$\frac{d}{dt}\left(|c_e(t)|^2 + \int d^3r \int_0^\infty d\omega \sum_{m=1}^3 |c_g(\mathbf{r},\omega,m|t)|^2\right) = 0,$$
(16)

i.e., the unitarity condition Eq. (8) is fulfilled for all times as it should be. We notice, what may appear to be a trivial fact, that general time-dependent phase redefinitions of the amplitudes will not change the unitary condition Eq. (8). This circumstance will, nevertheless, be useful below in order to circumvent divergent vacuum fluctuations of the theory.

Equation (9), a well-known Volterra integral equation of the second kind, may be integrated with respect to time. The result is then

$$c_e(t) = 1 + \int_0^t dt' \kappa(t - t') c_e(t'), \qquad (17)$$

where

$$\kappa(t) \equiv \int_0^t dt' \, K(t') \tag{18}$$

is the time-integrated kernel, i.e.,

$$\kappa(t) = \frac{1}{2\pi} \int_0^\infty d\omega \frac{e^{-i(\omega-\omega_A)t} - 1}{i(\omega-\omega_A)} \Gamma(\mathbf{r}_A, \omega).$$
(19)

In passing, we mention that a rate similar to Eq. (11) may be derived for electric-dipole transitions (see, e.g., Refs. [50–53]).

In that case, the spontaneous decay rate $\Gamma(\mathbf{r},\omega)$ should be replaced by

$$\Gamma_E(\mathbf{r},\omega) = \frac{2\omega_A^2}{\hbar\epsilon_0 c^2} \boldsymbol{d}_{eg} \cdot \operatorname{Im}[\mathbf{G}(\mathbf{r},\mathbf{r},\omega)] \cdot \boldsymbol{d}_{ge}, \qquad (20)$$

where d_{eg} is the electric-dipole moment for the transition $e \rightarrow g$. By comparing Eqs. (11) and (20) it is now straightforward to translate between magnetic-dipole and electric-dipole transitions which will be made use of below. When referring to electric-dipole transitions we therefore assume that one makes use of the appropriate decay rate, i.e., Eq. (20).

III. Vacuum

A. Exact

The dyadic Green tensor for vacuum, $\mathbf{G}^{0}(\mathbf{r},\mathbf{r},\omega)$, is given by a well-known expression (see, e.g., Refs. [50,51])

$$\operatorname{Im}[\overrightarrow{\nabla} \times \mathbf{G}^{0}(\mathbf{r},\mathbf{r},\omega) \times \overleftarrow{\nabla}] = \frac{\omega^{3}}{6\pi c^{3}} \begin{bmatrix} 1 & 0 & 0\\ 0 & 1 & 0\\ 0 & 0 & 1 \end{bmatrix}, \quad (21)$$

independent of the position \mathbf{r} as it should be due to the translational invariance of the vacuum quantum state.

In this case the kernel in Eq. (19) reduces to the vacuum kernel $\kappa_n(t)$ and reads

$$\kappa_n(t) = \frac{\Gamma_0}{2\pi} \int_0^{\omega_c} d\omega \left(\frac{\omega}{\omega_A}\right)^n \frac{e^{-i(\omega-\omega_A)t} - 1}{i(\omega-\omega_A)}, \qquad (22)$$

with n = 3, and where we have introduced a cutoff frequency ω_c in order to make the integral finite. The decay rate of a magnetic spin-flip transition for a two-level atom with no angular momentum and a negligible nuclear moment in free space is then (see, e.g., Ref. [31])

$$\Gamma_0 = \bar{\Gamma}_0 S^2, \tag{23}$$

with

$$\bar{\Gamma}_0 = \mu_0 \frac{(\mu_B g_S)^2}{3\pi\hbar} k_A^3,$$
(24)

and $k_A \equiv \omega_A/c$ is the wave number in vacuum. Here we have introduced the dimensionless spin factor $S^2 \equiv S_x^2 + S_y^2 + S_z^2$, where $S_j \equiv \langle g | \hat{S}_j / \hbar | e \rangle$ is the dimensionless matrix element component of the electron-spin operators \hat{S}_j corresponding to the transition $|e\rangle \rightarrow |g\rangle$, with j = x, y, z. Furthermore, $\mu_B = e\hbar/2m_e$ is the conventional Bohr magneton and $g_S \approx 2$ is the electron-spin g_S factor.

Clearly, the time-dependent kernel in Eq. (22) is divergent as $\omega_c \rightarrow \infty$ and it is not entirely clear to us how to make sense of this kernel for all times *t*. Since, as we have mentioned above, magnetic-dipole transitions are analogues to electric-dipole transitions, we therefore appeal to Bethe's mass-renormalization [54] procedure as far as dealing with divergences are concerned. At large *t* we recall that one then, e.g., can make use of the distributional identity

$$\lim_{t \to \infty} \frac{e^{-i(\omega - \omega_A)t} - 1}{\omega - \omega_A} = P\left(\frac{1}{\omega_A - \omega}\right) - i\pi\delta(\omega - \omega_A),$$
(25)

where one from the principal part $P(1/(\omega_A - \omega))$ then adds a term $P(1/\omega)$ which, when summed over all possible final states, corresponds to Bethe's introduction of a renormalized mass. In the electric-dipole case, the finite-time kernel $\kappa_n(t)$ will then be regularized in such a manner that, for large *t*, energy shifts are reduced to at most a logarithmic dependence of the cutoff frequency ω_c . As we will see below in Sec. III D this regularization can be carried out in terms of frequency shift and a conventional mass renormalization for all times with a mass counterterm.

In the present case of magnetic transitions, which corresponds to an interaction which is not directly renormalizable in the same fashion as for electric-dipole transitions (see, e.g., Ref. [55]) we, nevertheless, proceed in a manner which treats these different transitions in an equal manner as far as divergences are concerned. We therefore subtract the secondorder expansion of the denominator in Eq. (19) for $\omega \gg \omega_A$ in such a way that at most a logarithmic dependence of the cutoff frequency ω_c remains, i.e.,

$$\frac{1}{\omega - \omega_A} \to \frac{1}{\omega - \omega_A} - \left(\frac{1}{\omega} + \frac{\omega_A}{\omega^2} + \frac{\omega_A^2}{\omega^3}\right), \quad (26)$$

which leaves us with the following regularized version of the kernel Eq. (22):

$$\kappa_0^R(t) = \frac{\Gamma_0}{2\pi} \int_0^{\omega_c} d\omega \frac{e^{-i(\omega-\omega_A)t} - 1}{i(\omega-\omega_A)}.$$
 (27)

The kernel $\kappa_0^R(t)$ is therefore obtained from $\kappa_0(t)$ in Eq. (22) using the rule $\Gamma(\mathbf{r},\omega) \to \Gamma(\mathbf{r},\omega_A)$. With regard to the amplitude $c_g(\mathbf{r},\omega,m|t)$ we observe that unitarity prevails if we in Eq. (15) also make use of the same prescription. The fact that unitarity is preserved is our primary motivation for the introduction of the regularization procedure above. Below we will, however, investigate the effect of keeping a dependence ω/ω_A instead of $(\omega/\omega_A)^3$ in the kernel Eq. (22). This would be in line with the standard electric-dipole transition considerations as used in, e.g., Refs. [41–45]. As we will verify below, the choice of regularization procedure will affect the large-time behavior of the decaying system.

In Eq. (27) we make use of a cutoff frequency ω_c in order to make the frequency integral finite. Since our calculation is nonrelativistic we can, e.g., identify this cutoff with $m_e c^2/\hbar$, but one may also regard this as a free parameter when one, e.g., considers artificial atoms (Ref. [5] and references cited therein). We therefore introduce a dimensionless parameter cutoff parameter Λ as defined by

$$\Lambda \equiv \frac{\omega_c}{\omega_A}.$$
 (28)

The subtraction procedure of $\kappa_0(t)$ above enables us to extract the leading logarithmic dependence of the cutoff parameter Λ , and, in the end, we have therefore replaced the vacuum kernel $\kappa_0(t)$ with the following regularized kernel $\kappa_0^R(t)$:

$$\kappa_0^R(t) = -\frac{\Gamma_0}{2\pi} \int_{-\omega_A}^{(\Lambda-1)\omega_A} dx \frac{\sin(xt)}{x} - i\frac{\Gamma_0}{2\pi} \int_{\omega_A}^{(\Lambda-1)\omega_A} dx \frac{\cos(xt)}{x} + i\frac{\Gamma_0}{2\pi} \ln(\Lambda-1).$$
(29)

The imaginary logarithmic term in this equation corresponds to an induced Lamb shift due to vacuum fluctuations since it can be removed by introducing an angular frequency shift, i.e.,

$$\tilde{\omega}_A \equiv \omega_A - \frac{\Gamma_0}{2\pi} \ln(\Lambda - 1), \qquad (30)$$

and applying the transformation

$$c_e(t) \to \tilde{c}_e(t) \equiv c_e(t) \exp\left(-i\frac{\Gamma_0 t}{2\pi}\ln(\Lambda - 1)\right),$$
 (31)

as can be seen by making use of Eq. (9). We also find it convenient to define the kernel

$$\tilde{\kappa}_{0}^{R}(t) = -\frac{\Gamma_{0}}{2\pi} \left\{ \operatorname{Si}[(\tilde{\Lambda} - 1)\tilde{\omega}_{A}t] + \operatorname{Si}[\tilde{\omega}_{A}t] \right\} - i\frac{\Gamma_{0}}{2\pi} \left\{ \operatorname{Ci}[(\tilde{\Lambda} - 1)\tilde{\omega}_{A}t] - \operatorname{Ci}[\tilde{\omega}_{A}t] \right\}, \quad (32)$$

where Ci(x) and Si(x) are the standard cosine and sine integral, respectively (see, e.g., Ref. [56]) and $\tilde{\Lambda} \equiv \Lambda \omega_A / \tilde{\omega}_A \simeq \Lambda$ in view of the fact that Λ is considered to be a large cutoff parameter. The probability amplitude $\tilde{c}_e(t)$ is now a solution to the integral equation Eq. (17) provided the kernel $\kappa_0^R(t)$ is replaced by $\tilde{\kappa}_0^R(t)$, i.e., we consider

$$\tilde{c}_{e}(t) = 1 + \int_{0}^{t} dt' \tilde{\kappa}_{0}^{R}(t - t') \tilde{c}_{e}(t').$$
(33)

The solutions of the corresponding integral equation cannot be obtained in closed form and therefore we will resort to a numerical treatment. The numerical results as presented in the present paper involve algorithms with arbitrary numerical precision.

B. Approximations

It is convenient to define a dimensionless time parameter $\tau \equiv \Gamma_0 t$ as well as the parameters $b_A \equiv \omega_A / \Gamma_0$ and $\tilde{b}_A \equiv \tilde{\omega}_A / \Gamma_0$, i.e.,

$$\tilde{b}_A = b_A - \frac{\ln(\Lambda - 1)}{2\pi}.$$
(34)

Let us now consider small-time scales such that $(\tilde{\Lambda} - 1)\tilde{b}_A \tau \ll 1$ and $\tilde{b}_A \tau \ll 1$. Such small times are basically of academic interest as $t \ll \hbar/m_e c^2 = 1.29 \times 10^{-21}$ s unless, as suggested above, a different scale can be provided by the use of, e.g., artificial atoms. Virtual particles may then be created and our theory is not strictly valid. The leading series expansion of the kernel $\tilde{\kappa}_0^R(t)$, as given by Eq. (32), for such small-time parameters τ is

$$\tilde{\kappa}_0^R(\tau) \approx -\frac{\Gamma_0}{2\pi} \Lambda \tilde{b}_A \tau - i \frac{\Gamma_0}{2\pi} \ln(\Lambda - 1), \qquad (35)$$

using $\tilde{\Lambda} \approx \Lambda$. Apart from a sign, the imaginary part of this kernel is the same as the last imaginary part of Eq. (29). It is therefore convenient to invert the transformation Eq. (31) and consider the probability amplitude $c_e(\tau)$.

Substituting the kernel $\kappa_0^R(\tau)$ into Eq. (17), we obtain

$$c_e(\tau) \approx 1 - \frac{\Lambda}{4\pi b_A} (b_A \tau)^2, \qquad (36)$$

using the approximation $c_e(t') \approx 1$ in Eq. (17) since time parameter τ is small. We observe that in Eq. (36) the parameter b_A enters and not \tilde{b}_A . In passing, we also mention that an expansion for small times is also given in Ref. [43] [their Eq. (3.21)] in the case of electric-dipole transitions, with the result

$$c_e(\tau) \approx 1 - \frac{\Lambda^2}{8\pi} (b_A \tau)^2. \tag{37}$$

The difference between Eqs. (36) and (37), in which case, as mentioned above, the factor $(\omega/\omega_A)^3$ in Eq. (22) is replaced by ω/ω_A , illustrates the relevance of the subtraction procedure. Below we will see that this effect is even more pronounced at large vales of τ . The discrepancy between these last two expressions is easily explained by the fact that different regularization procedures have been applied.

Let us still consider small times $\tilde{b}_A \tau \ll 1$ but such that $(\tilde{\Lambda} - 1)\tilde{b}_A \tau \gg 1$. Such time scales are of relevance when one, e.g., studies the Zeno effect for quantum systems (see, e.g., Refs. [22,57]). For such intermediate times, the kernel in Eq. (32) is reduced to

$$\tilde{\kappa}_0^R(\tau) \approx -\frac{\Gamma_0}{4} + i \frac{\Gamma_0}{2\pi} [\gamma_E + \ln(\tilde{b}_A \tau)], \qquad (38)$$

where $\gamma_E \approx 0.577\,216$ is Euler's constant. Substituting Eq. (38) into Eq. (33), expressed in terms of $\tilde{c}_e(\tau)$ and using $\tilde{c}_e(\tau') \approx 1$, we may then carry out the time integration, with the result

$$\tilde{c}_e(\tau) \approx 1 - \frac{\tau}{4} + i \frac{\tau}{2\pi} [\gamma_E + \ln(\tilde{b}_A \tau) - 1].$$
(39)

The corresponding probability $|\tilde{c}_e(\tau)|^2 = |c_e(\tau)|^2$ is illustrated in Figs. 1 and 2 (dash-dotted line) together with the exact numerical solution, i.e., the numerical solution of Eq. (33) with the exact kernel Eq. (32) (solid line).



FIG. 1. Probability $|c_e(\tau)|^2 = |\tilde{c}_e(\tau)|^2$ as a function of $\tau \equiv \Gamma_0 t$. The solid curve corresponds to the exact numerical solution, i.e., solution of Eq. (33) with the kernel $\tilde{\kappa}_0^R(t)$ as given by Eq. (32) in the main text for a two-level system in vacuum. The upper (dash-dotted) curve corresponds to the small-time, $\tilde{b}_A \tau \ll 1$, expansion Eq. (39). The lower (dotted) curve corresponds to the exponential decay $\exp(-\tau)$ with expected deviations at small τ . The values of the relevant parameters are $\tilde{b}_A \equiv \tilde{\omega}_A / \Gamma_0 = 10$ and $\Lambda = 1000$.



FIG. 2. Probability $|c_e(\tau)|^2 = |\tilde{c}_e(\tau)|^2$ as a function of $\tau \equiv \Gamma_0 t$ as in Fig. 1, but for a smaller τ interval.

Finally, for large times as compared to the shifted atomic frequency transition, i.e., $\tilde{b}_A \tau \gg 1$, and also $(\tilde{\Lambda} - 1)\tilde{b}_A \tau \gg 1$, the kernel Eq. (32) is, to first leading order,

$$\tilde{\kappa}_0^R(\tau) \approx -\frac{\Gamma_0}{2} + \frac{\Gamma_0}{2\pi} \frac{e^{ib_A \tau}}{\tilde{b}_A \tau}.$$
(40)

One may then show that (see next section and Appendix B)

$$\tilde{c}_e(\tau) \approx e^{-\tau/2} + \frac{e^{i\tilde{b}_A\tau}}{2\pi i\tau [\tilde{b}_A - \ln(\tilde{b}_A\tau)/2\pi]^2}, \qquad (41)$$

provided \tilde{b}_A is sufficiently large, and where we have made use of $\tilde{\Lambda} \approx \Lambda$. This solution includes the well-known exponential decay as well as a correction term which dominates $|\tilde{c}_e(\tau)|^2$



FIG. 3. Probability $|c_e(\tau)|^2 = |\tilde{c}_e(\tau)|^2$ as a function of $\tau \equiv \Gamma_0 t$. The solid curve corresponds to the exact numerical solution, i.e., solution of Eq. (33) with the kernel $\tilde{\kappa}_0^R(t)$ as in Eq. (32) for a two-level system in vacuum. The almost overlapping dotted curve corresponds to Eq. (41). The values of the relevant parameters are $\tilde{b}_A \equiv \tilde{\omega}_A / \Gamma_0 = 10$ and $\Lambda = 1000$. The inset shows a limited part of the larger figure and illustrates the accuracy of the asymptotic form Eq. (41).

for large values of τ . Eq. (41) is plotted in Fig. 3 (dotted line) together with the exact numerical solution (solid line).

The last term in Eq. (41) will dominate over the exponential for times $\tau \gtrsim \tau^*$, where τ^* is determined by the transcendental equation

$$e^{-\tau^*/2} \approx rac{1}{2\pi \, \tau^* [\tilde{b}_A - \ln(\tilde{b}_A \tau^*)/2\pi]^2},$$
 (42)

valid for a sufficiently large value of \tilde{b}_A . For $\tilde{b}_A = 10$ and $\Lambda = 1000$ as in Fig. 3, the solution of this equation is, e.g., $\tau^* \approx 18.5$. The $\tau \ln^2(\tilde{b}_A \tau)$ dependence in Eqs. (41) and (42) becomes more important than the τ dependence only for very large-time scales $\tau \gtrsim \tau_{\ln}$, where $\tau_{\ln} = \exp(2\pi \tilde{b}_A)/\tilde{b}_A$, in which case $|\tilde{c}_e(\tau)|^2$ becomes exponentially close to zero with an increasing value of \tilde{b}_A .

C. Asymptotic expansion

Laplace transform techniques have been used to investigate possible deviations from exponential decay as in Refs. [41,43,44]. As shown in particular by Seke and Herfort [43,44], careful considerations of the analytical properties of the decay amplitude are required in order to extract the asymptotic behavior of $c_e(t)$. The result of Refs. [41,43,44] may seem to be partly contradictory. It is therefore of some interest to investigate in detail in what manner the asymptotic behavior according to Eq. (41) is obtained by making use of Laplace transform techniques. As we will see, it then becomes apparent that the various asymptotic forms of the decay amplitude are all valid but depend on the time scale and other physical parameters at hand.

In order to obtain the corresponding asymptotic expansion, as τ becomes large, we therefore find it convenient to consider the Laplace transform of the differential equation for the rescaled amplitude $\tilde{c}_e(t)$ as defined in Eq. (31), i.e.,

$$\frac{d\tilde{c}_e(t)}{dt} = \int_0^t dt' \tilde{K}_0^R(t-t') \tilde{c}_e(t') - i \frac{\Gamma_0}{2\pi} \ln(\Lambda-1) \tilde{c}_e(t),$$
(43)

where the regularized kernel $K_n^R(t)$ is

$$\tilde{K}_{n}^{R}(t) = -\frac{\Gamma_{0}}{2\pi} \int_{0}^{\omega_{c}} d\omega \left(\frac{\omega}{\omega_{A}}\right)^{n} e^{-i(\omega-\tilde{\omega}_{A})t}, \qquad (44)$$

with, for the moment, n = 0. The Laplace transform $\tilde{c}_e(s)$ is then given by

$$\tilde{c}_e(s) = \frac{1}{s - \tilde{K}_0^R(s) + i\Gamma_0 \ln(\Lambda - 1)/2\pi},$$
(45)

where we have identified

$$\tilde{K}_{0}^{R}(s) = i \frac{\Gamma_{0}}{2\pi} \int_{0}^{\omega_{c}} d\omega \frac{1}{\omega - \tilde{\omega}_{A} - is}$$
$$= i \frac{\Gamma_{0}}{2\pi} \bigg[\ln \bigg(1 - \frac{\omega_{c}}{is + \tilde{\omega}_{A}} \bigg) \bigg], \qquad (46)$$

in terms of the principal branch of the natural logarithm In function. The inverse Laplace transform in terms of the Bromwich integral and the dimensionless time parameter τ can then formally, after a suitable Wick ($u = \tilde{b}_A + is/\Gamma_0$) rotation, be written in the form

$$\tilde{c}_{e}(\tau) = \frac{e^{i\tilde{b}_{A}\tau}}{2\pi i} \int_{\infty+i\gamma}^{-\infty+i\gamma} du \, e^{-iu\tau} \times \left[u - b_{A} + \frac{1}{2\pi} \ln\left(1 - \frac{\Lambda_{c}}{u}\right) \right]^{-1}, \quad (47)$$

where we have defined

$$\Lambda_c \equiv \frac{\omega_c}{\Gamma_0},\tag{48}$$

and where the positive real number γ is chosen in such a way that the possible singularities of the integrand are above the integration contour.

We now follow the methods of, in particular, Refs. [43,44], where more details can be found, and use Eq. (47) in order to extract the large τ , i.e., $u \rightarrow 0$, asymptotic expansion for $\tilde{c}_e(\tau)$. As in Refs. [43,44], we identify the functions

$$M_0(u) \equiv u - b_A + \frac{1}{2\pi} [\log(u - \Lambda_c) - \log(u)]$$
 (49)

and

$$M_1(u) \equiv M_0(u) + i, \tag{50}$$

where the log function now, and in the rest of the paper, stands for the multivalued natural logarithm function. The appropriate Riemann surface for the function $\log(u - \Lambda_c) - \log(u)$ has been obtained in Ref. [43]. Here one joins a second Riemann sheet at the branch cut along the real *u* axis from u = 0 to $u = \Lambda_c$.

The integration contour in Eq. (47) can then be deformed into two curves of integration that run parallel to the Im(u) axis. One of these curves runs from $-i\infty + \Lambda_c + \epsilon$, above and around the branch point $u = \Lambda_c$ on the first Riemann sheet, and then towards $-i\infty + \Lambda_c - \epsilon$ on the second Riemann sheet, where ϵ is a small and positive real number. Similarly, the other curve of integration runs from $-i\infty + \epsilon$ on the second Riemann sheet, above and around the branch point u = 0 on the first Riemann sheet, and then towards $-i\infty - \epsilon$ on the first Riemann sheet.

The function $M_1(u)$ is obtained from $M_0(u)$ when passing to the second sheet of the Riemann surface construction. It can be shown that each of the functions $M_0(u)$ and $M_1(u)$ have no poles and only one zero on each of the Riemann sheets considered [43,44]. On the first Riemann sheet, where the original functions in Eq. (47) are defined, there is, in addition, a zero of $M_0(u)$ along the real axis at $u_0 \approx \Lambda_c(1 + \exp[-2\pi \Lambda_c])$. The corresponding residue is exponentially suppressed, i.e., $Z_0 \equiv 1/(dM_0(u)/du)_{u=u_0} \approx 2\pi \Lambda_c \exp(-2\pi \Lambda_c)$, apart from a phase factor, and the corresponding pole contribution has therefore been neglected.

In the deformation of the integration contour in Eq. (47), and on the second Riemann sheet, one now encounters the conventional Wigner-Weisskopf–like pole contribution at $u = u_1 \approx \tilde{b}_A - i/2$, provided $\tilde{b}_A \gg 1$ due to Eq. (50). Since $dM_1(u)/du = 1 + \Lambda_c/2\pi u(u - \Lambda_c) \approx 1 - 1/\tilde{b}_A \approx 1$ for $u = u_1$ provided $\tilde{b}_A \gg 1$, this pole leads to a residue $Z_1 \equiv 1/(dM_1(u)/du)_{u=u_1} \approx 1$. This Wigner-Weisskopf–like pole contribution therefore contributes to the amplitude $\tilde{c}_e(\tau)$ with $\exp(-\tau/2)$, i.e., the expected and conventional amplitude describing exponential decay. Including the two integrals emerging from the branch points mentioned above we then, finally, obtain that the result

$$\tilde{c}_{e}(\tau) = e^{-\tau/2} - \frac{e^{i\tilde{b}_{A}\tau}}{2\pi\tau}I_{1} - \frac{e^{i\tilde{b}_{A}(1-\tilde{\Lambda})\tau}}{2\pi\tau}I_{2},$$
(51)

where we have defined the integrals

$$I_1 = \int_0^\infty ds \, e^{-s} \left[\frac{1}{M_0(-is/\tau)} - \frac{1}{M_1(-is/\tau)} \right] \quad (52)$$

and

$$I_{2} = \int_{0}^{\infty} ds \, e^{-s} \left[\frac{1}{M_{1}(\Lambda_{c} - is/\tau)} - \frac{1}{M_{0}(\Lambda_{c} - is/\tau)} \right],$$
(53)

similar to the results of Refs. [43,44]. In Appendix B we now show that, as $\tau \gg 1$,

$$I_1 \approx \frac{\iota}{[\tilde{b}_A - \ln(\tilde{b}_A \tau)/2\pi]^2},\tag{54}$$

provided $|b_A - \ln(\Lambda_c \tau)/2\pi| \approx |\tilde{b}_A - \ln(\tilde{b}_A \tau)/2\pi| \gg 1$. In the same manner it also follows that

$$I_2 \approx \frac{\iota}{[\Lambda_c - b_A - \ln(\Lambda_c \tau)/2\pi]^2},$$
(55)

provided $|\Lambda_c - b_A - \ln(\Lambda_c \tau)/2\pi| \gg 1$. As long as $\Lambda_c \gg b_A$ it is now clear that I_1 will dominate over I_2 . Due to Eq. (54) we now recover the result as given in Eq. (41). It now also follows that $I_1 \approx 1/\tilde{b}_A^2$ unless τ is exponentially large, i.e., $\tau \gg e^{2\pi \tilde{b}_A}/\tilde{b}_A$, in which case the decay amplitude $\tilde{c}_e(\tau)$ is exponentially small.

When compared to the analysis of Refs. [43,44] we remark that our approach to the asymptotic behavior of integrals like Eqs. (52) and (B1) has a much broader range of applicability. In particular, we notice that, as long as $\tau \ll e^{2\pi \tilde{b}_A}/\tilde{b}_A$), the I_1 integral leads to a $O(1/\tau)$ behavior of the decay amplitude $\tilde{c}_e(\tau)$. It is only in the opposite limit that the proposed $O(1/\tau \ln^2 \tau)$ of Refs. [43,44] emerges, i.e., for $\tau \gg e^{2\pi \tilde{b}_A}/\tilde{b}_A$. In this case the decay amplitude, however, becomes exponentially small for \tilde{b}_A sufficiently large. We also find it important to express the asymptotic behavior in terms of dimensionless quantities like τ and not in terms of the time variable t with a physical dimension as in Refs. [43,44].

D. Comparison with Refs. [41,43,44]

As alluded to above, in many cases of the study of deviations from the exponential decay of atomic spontaneous emission processes, various approximations have been applied in order to obtain explicit and analytical expressions as, e.g., in Refs. [41,43,44] and references cited therein. In these references one considers electric-dipole transitions and, in the integral equation Eq. (17) for the vacuum case, the kernel $\kappa(\tau)$ should therefore be replaced by kernel $\kappa_1^R(\tau) \equiv \kappa_1(\tau)$ as defined by Eq. (22), i.e., with a factor ω/ω_A in the integrand rather than $(\omega/\omega_A)^3$. Contrary to the regularization procedure employed in Sec. III, this factor ω/ω_A is, however, kept untouched and a suitable subtraction is carried out at a later stage in the analysis. As we now will verify, the actual subtraction procedure in order to make the corresponding kernel finite will effect the large-time deviation from exponential decay. If the

cutoff frequency ω_c again is regarded to be a finite physical quantity, we notice that the regularized kernel $\kappa_1^R(\tau)$ may be written in terms of $\kappa_0^R(\tau)$ as given by Eq. (27):

$$\kappa_1^R(\tau) \equiv \kappa_0^R(\tau) + \frac{\Gamma_0}{2\pi b_A \tau} (e^{-i(\Lambda - 1)b_A \tau} - e^{ib_A \tau}) + i \frac{\Gamma_0}{2\pi} \Lambda.$$
(56)

Due to this identity we realize that the kernel $\kappa_1^R(\tau)$ is linear in the dimensionless cutoff frequency rather than the logarithmic dependence of $\kappa_0^R(\tau)$ as in Eq. (29). We observe that the linear Λ term may now be absorbed in an energy shift such that the kernel $\kappa_1^R(\tau)$ is replaced according to

$$\kappa_1^R(\tau) \to \kappa_1^R(\tau)$$

$$\equiv \kappa_0^R(\tau) + \frac{\Gamma_0}{2\pi b'_A \tau} (e^{-i(\Lambda - 1)b'_A \tau} - e^{ib'_A \tau}), \quad (57)$$

where $b'_A \equiv \omega'_A / \Gamma_0$, and

$$\omega'_A \equiv \omega_A - \frac{\Gamma_0}{2\pi}\Lambda, \qquad (58)$$

and where also the ω_A dependence in $\kappa_0^R(\tau)$ is replaced by ω'_A . Such a frequency shift preserves unitarity. As in Refs. [41,43,44], the linear Λ dependence can therefore be removed altogether by applying Bethe's mass renormalization [54], i.e., by including a suitable mass counterterm in the Hamiltonian. After this removal of the linear Λ dependence and by also applying the transformation in Eq. (31), the kernel $\kappa_1^R(\tau)$ is transformed into the kernel $\tilde{\kappa}_1^R(\tau)$ according to

$$\kappa_1^R(\tau) \to \tilde{\kappa}_1^R(\tau)$$

$$\equiv \tilde{\kappa}_0^R(\tau) + \frac{\Gamma_0}{2\pi \tilde{b}_A \tau} (e^{-i(\tilde{\Lambda}-1)\tilde{b}_A \tau} - e^{i\tilde{b}_A \tau}).$$
(59)

The asymptotic form of the kernel $\tilde{\kappa}_1^R(\tau)$ is now such that the subleading contribution form the asymptotic form of the kernel $\tilde{\kappa}_0^R(\tau)$, as given by Eq. (40), is canceled and therefore

$$\tilde{\kappa}_1^R(\tau) \approx -\frac{\Gamma_0}{2} + \frac{\Gamma_0}{2\pi} \frac{e^{-i(\tilde{\Lambda}-1)\tilde{b}_A \tau}}{\tilde{b}_A \tau},\tag{60}$$

for sufficiently large τ . Hence the methods of Appendix A would then lead to

$$\tilde{c}_e(\tau) \approx e^{-\tau/2} - \frac{e^{-i(\bar{\Lambda}-1)\bar{b}_A \tau}}{2\pi i \tau \tilde{\Lambda} \tilde{b}_A^2},\tag{61}$$

apart from possible $1/\tau^2$ corrections provided \tilde{b}_A is sufficiently large.

As in Sec. IIIC, a more precise asymptotic expansion as τ becomes large, and which also takes the Lamb shift Eq. (30) more carefully into account, can be obtained by a consideration of a Bromwich integral. The Laplace transform of the differential equation for the rescaled amplitude $\tilde{c}_e(t)$ reads

$$\frac{d\tilde{c}_e(t)}{dt} = \int_0^t dt' \tilde{K}_1^R(t-t') \tilde{c}_e(t') -i \frac{\Gamma_0}{2\pi} \ln(\Lambda-1) \tilde{c}_e(t) - i \frac{\delta m c^2}{\hbar} \tilde{c}_e(t), \quad (62)$$

where the regularized kernel $\tilde{K}_{1}^{R}(t)$ is

$$\tilde{K}_{1}^{R}(t) = -\frac{\Gamma_{0}}{2\pi} \int_{0}^{\omega_{c}} d\omega \frac{\omega}{\omega_{A}} e^{-i(\omega - \tilde{\omega}_{A})t}, \qquad (63)$$

and where the unitarity preserving transformation as defined by Eq. (31) has been applied. In Eq. (62) we have also included the contribution due to the additional mass counterterm $\delta m c^2/\hbar$ in the Hamiltonian which has the role of canceling the linear Λ dependence in the kernel Eq. (63). After a tedious but straightforward analysis, following the same Laplace-transformation techniques as in Refs. [43,44], the large τ expansion of $\tilde{c}_e(\tau)$ can now be obtained. Similar to the definitions of $M_0(u)$ and $M_1(u)$ in Sec. III C, we therefore introduce the functions

$$N_0(u) \equiv u - b_A + \frac{u}{2\pi b_A} [\log(u - \Lambda_c) - \log u] \quad (64)$$

and

$$N_1(u) \equiv N_0(u) + i\frac{u}{b_A},\tag{65}$$

where the linear Λ dependence in Eq. (62) now has explicitly been removed by the mass-renormalization procedure. The solution to Eq. (62) can then be written in the form

$$\tilde{c}_e(\tau) = \frac{e^{ib_A \tau}}{2\pi i} \int_{\infty+i\gamma}^{-\infty+i\gamma} du \, e^{-iu\tau} \frac{1}{N_0(u)}.$$
(66)

By deforming the integration contour in Eq. (66) in the same manner as in the derivation of Eq. (51), one then obtains the result

$$\tilde{c}_{e}(\tau) = e^{-\tau/2} - \frac{e^{i\tilde{b}_{A}\tau}}{2\pi\tau} J_{1} - \frac{e^{i\tilde{b}_{A}(1-\tilde{\Lambda})\tau}}{2\pi\tau} J_{2},$$
(67)

where we have defined the integrals

$$J_1 = \int_0^\infty ds \, e^{-s} \left(\frac{1}{N_0(-is/\tau)} - \frac{1}{N_1(-is/\tau)} \right) \tag{68}$$

and

$$J_2 = \int_0^\infty ds \, e^{-s} \left(\frac{1}{N_1(\Lambda_c - is/\tau)} - \frac{1}{N_0(\Lambda_c - is/\tau)} \right).$$
(69)

We have therefore actually reproduced the results of Refs. [43, 44], but now expressed in terms of dimensionless parameters.

As expanded upon in Appendix C, the integrals J_1 and J_2 can now be analyzed in a manner similar to the integrals I_1 and I_1 of Sec. III C. If $\tau \gg 1$ and $\Lambda_c \gg 1$ and, in addition, $b_A \tau \gtrsim 1 + \ln(\Lambda_c)/2\pi b_A$, we then obtain

$$J_1 \approx \frac{1}{b_A^3 \tau}.$$
 (70)

For the integral J_2 it also follows that, for $\tau \gg 1$,

$$J_2 \approx \frac{1}{i\Lambda[b_A - \ln(\Lambda_c \tau)/2\pi]^2},\tag{71}$$

provided $|b_A - \ln(\Lambda_c \tau)/2\pi| \gg 1$. The insertion of these asymptotic expansions into Eq. (67) therefore leads to

$$\tilde{c}_{e}(\tau) \approx e^{-\tau/2} - e^{i\tilde{b}_{A}\tau} \frac{1}{2\pi b_{A}^{3}} \frac{1}{\tau^{2}} - \frac{e^{-i(\tilde{\Lambda}-1)\tilde{b}_{A}\tau}}{2\pi i\Lambda} \frac{1}{\tau} \frac{1}{[b_{A} - \ln(\Lambda_{c}\tau)/2\pi]^{2}}.$$
 (72)

The last term in Eq. (72) with the $[b_A - \ln(\Lambda_c \tau)/2\pi]$ dependence has been obtained by using the technique of Appendix B in order to see how the claimed $\tau \ln^2 \tau$ dependence of Refs. [43,44] actually emerges. Here we again stress that our results are expressed in terms of dimensionless quantities in contrast to the results of Refs. [43,44].

At this point we now quote the following explicit, but approximative, expression for the probability amplitude $c_e(\tau)$ result of Knight and Milonni in Ref. [41]:

$$c_{e}(\tau) = e^{-\tau/2 + i\delta b\tau} - \frac{e^{ib_{A}\tau}}{2\pi i}$$

$$\times \int_{0}^{\infty} dx \, e^{-\Lambda x b_{A}\tau} \left(\frac{1}{x - \frac{i}{\Lambda} + i\frac{x}{2b_{A}} - \frac{x}{2\pi b_{A}} \left[\ln x - i\frac{\pi}{2}\right]} - \frac{1}{x - \frac{i}{\Lambda} - \frac{x}{2\pi b_{A}} \left[\ln x + i\frac{\pi}{2}\right]}\right), \tag{73}$$

where $\delta b = \delta \omega / \Gamma_0$, and

$$\delta\omega = \frac{\Gamma_0}{2\pi}\ln(\Lambda - 1) \tag{74}$$

is the angular frequency shift of the excited state $|e\rangle$. This expression, which we have verified [58], is obtained by deforming integration contours of the Bromwich integral by only considering the analytical properties of the natural logarithm In function and not taking into account the additional analytical conditions imposed by the appearance of the functions $N_0(u)$ and $N_1(u)$ as discussed above. For large times, i.e., $b_A \tau \gg 1$, this expression is reduced to [41,58]

$$c_e(\tau) \approx e^{-\tau/2 + i\delta b\tau} - \frac{1}{2\pi b_A} \frac{1}{(b_A \tau)^2} e^{ib_A \tau}.$$
 (75)

We notice that up to a phase redefinition, this equation and Eq. (72) only differ by that last Seke-Herfort term of Eq. (72). It is only in the limit $\Lambda \to \infty$ that Eq. (72) leads to $1/\tau^2$ asymptotic behavior of Knight and Milonni. It is now also clear that it is only for time scales $\tau \gtrsim e^{2\pi b_A}/\Lambda_c$ that the $1/\tau \ln^2 \tau$ asymptotic behavior of Seke-Herfort emerges. Such time scales are typically exponentially large leading to an exceedingly small decay amplitude $c_e(\tau)$ unless Λ_c is very large. But as we have seen above, in such a case the $1/\tau^2$ asymptotic behavior of Knight and Milonni begins to dominate. We also realize that Eq. (72) leads to a dominant $1/\tau$ behavior for $\tau \lesssim e^{2\pi b_A}/\Lambda_c$ unless Λ_c again is not arbitrarily large.

By a comparison of Eqs. (41) and (72) we also realize, due to the presence of the factor $1/\Lambda$ in the Seke-Herfort term in Eq. (72), that the regularization procedure actually affects the large-time behavior of the decay amplitude $c_e(\tau)$.

We illustrate some of these features in Figs. 4 and 5. For comparison, Eqs. (73) and (75) are plotted in Fig. 4 together with the exact numerical solution. In Fig. 5 we have plotted the solution of Eq. (33) with the kernel $\tilde{\kappa}_1^R(\tau)$ of Eq. (59) together with Eqs. (73) and (75). As long as the Seke-Herfort term in Eq. (72) dominates for sufficiently large values of τ , this contribution will dominate the conventional exponential decay term if $\tau \gtrsim \tau_*$, where τ_* is determined by the transcendental



FIG. 4. Probability $|c_e(\tau)|^2 = |\tilde{c}_e(\tau)|^2$ as a function of $\tau \equiv \Gamma_0 t$. The upper curve corresponds to the exact numerical solution, i.e., solution of Eq. (33) with the kernel $\tilde{\kappa}_0^R(t)$ as in Eq. (32) for a two-level system in vacuum which, within the numerical accuracy of the figure, overlaps with the asymptotic formula Eq. (41). The lower curve corresponds to the main result in Ref. [41], i.e., Eq. (73). The asymptotic form according to Eq. (75) is in excellent agreement with the numerical evaluation of Eq. (73). The values of the relevant parameters are $\tilde{b}_A = 10$ and $\Lambda = 1000$. As a reference, the straight line in the figure corresponds to a pure exponential decay, i.e., $|c_e(\tau)|^2 = e^{-\tau}$.

equation

$$e^{-\tau_*/2} \approx \frac{1}{2\pi \Lambda \tau_* \left[b_A - \ln(\Lambda_c \tau_*)/2\pi \right]^2},$$
 (76)



FIG. 5. Probability $|c_e(\tau)|^2 = |\tilde{c}_e(\tau)|^2$ as a function of $\tau \equiv \Gamma_0 t$. The upper curve corresponds to the exact numerical solution of the Volterra equation as considered in Refs. [43,44], i.e., of Eq. (33) with the kernel $\tilde{\kappa}_1^R(\tau)$ of Eq. (59) for a two-level system in vacuum. For large values of τ the solution is dominated by the $1/\tau$ asymptotic behavior according to Eq. (72). The lower curve corresponds to Eqs. (73) and (75), which overlaps within the numerical accuracy of the figure, with a characteristic $1/\tau^2$ asymptotic behavior for $c_e(\tau)$. The values of the relevant parameters are $\tilde{b}_A = 1000$ and $\Lambda = 1000$.

With the choice of parameters as in Fig. 5 and $\tau \gtrsim \tau_* \approx 53$, one finds that Eq. (72) in this case leads to $1/\tau$ scaling rather than the $1/\tau^2$ asymptotic scaling for the decay amplitude $c_e(\tau)$ according to Eq. (73).

IV. FINAL REMARKS

We have seen that the procedure of regularization of the decay amplitude $c_e(\tau)$ actually influences the deviation from exponential decay at large values of the time parameter τ . It would, of course, be of interest if one, e.g., in terms of artificial atoms could experimentally investigate deviations from exponential decay at large times and thereby infer a possible presence of the Seke-Herfort term of Eq. (72) or the related $1/\tau$ behavior obtained in Eq. (41). As alluded to in the introductory remarks and in the case of atom chips, the presence of material bodies can drastically change the characteristic lifetime (see, e.g., Refs. [30-34]) and one could then perhaps expect to encounter deviations from exponential decay. With the parameters of Refs. [31,33,34], i.e., $S^2 = 1/8$, $g_S \approx 2$, and $\omega_A/2\pi = 560$ kHz, we have that $b_A \approx 3 \times 10^{32}$. Using the cutoff Λ according to Eq. (28), i.e., $\Lambda \approx 2 \times 10^{14}$, one finds that $b_A \approx \tilde{b}_A$ and that $\tau^* \approx 315$ and $\tau_* \approx 380$ according to Eqs. (42) and (76), respectively. Since our analysis involves the dimensionless parameter τ , this means that even though the effective decay rate Γ can be changed by, say, twenty orders of magnitude (see, e.g., Refs. [30-34]), one still would have to consider several hundred decay times in order to see deviations from exponential decay. This means that for atom chips the conventional exponential decay description applies with an exceedingly high accuracy. As we, however, have remarked in the Introduction, experimental studies have, e.g., shown that artificial atoms can lead to a Lamb shift of the order of a few percent of the typical emission line [5]. We now estimate the corresponding relevant parameters according to Ref. [5]. The natural frequency is $\omega_A/2\pi \approx O(\text{GHz})$ and the effective rate Γ_0 is estimated from the Lorentzian fit to the experimental data to be $\Gamma_0 \approx 0.5 \times 10^{-2} \omega_A/2\pi$. With a Lamb shift of the order of one percent of ω_A , we then have that $\Gamma_0 \ln(\Lambda - 1)/2\pi \approx 10^{-2} \omega_A$, i.e., the effective cutoff Λ is then given by $\Lambda \approx e^{8\pi^2}$. One then finds somewhat more optimistic numbers $\tau^* \approx 40$ and $\tau_* \approx 200$ according to Eqs. (42) and (76), respectively, which may open the door for observing deviations from exponential decay in a system composed of artificial atoms. Finally, we also notice the resemblance of the semilogarithmic plots for $|c_e(\tau)|^2$ as in Figs. 3 and 4 in terms of the natural parameters τ and $\Delta Et = b_E \tau$, with $b_E = \Delta E / \Gamma_0$ for any typical intrinsic energy scale ΔE , and similar results obtained in Refs. [7,15] despite the fact that they refer to different physical systems.

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APPENDIX A: LARGE-TIME EXPANSION

In Sec. III B, the kernel $\tilde{\kappa}_0^R(\tau)$ was considered for three different regimes: small, intermediate, and large times τ for a given \tilde{b}_A and given Λ . The integral in the Volterra Eq. (33) may then trivially be split into these three intervals, where one interval relieves the other. For large times $\tau \gg 1$ and, in addition $\tilde{b}_A \gg 1$, two of these integrals are negligible. The Volterra equation in Eq. (33) is then left with an integral corresponding to the kernel in Eq. (40). This integral may be computed analytically:

$$\tilde{c}_{e}(\tau) - 1 \approx \int_{0}^{\tau - 1/\tilde{b}_{A}} dx \left\{ -\frac{1}{2} + \frac{1}{2\pi} \frac{e^{i\tilde{b}_{A}(\tau - x)}}{\tilde{b}_{A}(\tau - x)} \right\} e^{-x/2}$$

$$= -1 + e^{-\tau/2 + 1/2\tilde{b}_{A}} - \frac{1}{2\pi\tilde{b}_{A}} E_{1}[-(1/2 + i\tilde{b}_{A})\tau] e^{-\tau/2}$$

$$+ \frac{1}{2\pi\tilde{b}_{A}} E_{1}[-(1/2\tilde{b}_{A} + i)] e^{-\tau/2}, \quad (A1)$$

where $E_1(z)$ is the exponential integral for a complex argument z. As $\tau \gg 1$ and $\tilde{b}_A \gg 1$, the latter expression may now be simplified by making use of the leading asymptotic expansion of the E_1 functions in Eq. (A1) for large values of the parameter τ :

$$\tilde{c}_e(\tau) \approx e^{-\tau/2} + \frac{e^{i\tilde{b}_A \tau}}{2\pi i \tilde{b}_A^2 \tau} \frac{1}{1 - i/2\tilde{b}_A},$$
 (A2)

i.e., Eq. (41) if $\tilde{b}_A \gg 1$. As alluded to in the main text, Eq. (A2) remains valid for large values of τ provided $\tilde{b}_A \gg \ln(\tilde{b}_A \tau)/2\pi$, a condition that emerges from further iterations of the Volterra equation in Eq. (33). As discussed in Appendix B, a more precise asymptotic form than the one given by Eq. (A2) can be obtained by making use of Laplace transform techniques.

APPENDIX B: ASYMPTOTIC EXPANSION OF I1 AND I2

Here we first consider the integral I_1 as defined in Eq. (52) in Sec. III C, which we rewrite in the following convenient form:

$$I_{1} = \int_{0}^{\infty} ds \, e^{-s} \left[\frac{1}{-is/\tau - b_{A} + \ln(1 + \Lambda_{c}\tau/is)/2\pi} - \frac{1}{-is/\tau - b_{A} + \ln(1 + \Lambda_{c}\tau/is)/2\pi + i} \right], \quad (B1)$$

i.e., as τ becomes large,

$$I_{1} \approx -\int_{0}^{\infty} ds \, e^{-s} \left[\frac{1}{a + \ln s/2\pi + i/4} - \frac{1}{a + \ln s/2\pi - 3i/4} \right], \tag{B2}$$

where we have defined

$$a \equiv b_A - \frac{\ln(\Lambda_c \tau)}{2\pi} \approx \tilde{b}_A - \frac{\ln(\tilde{b}_A \tau)}{2\pi}, \qquad (B3)$$

if $\Lambda \approx \tilde{\Lambda}$. One now finds, by a combination of analytical and numerical methods, that the real part of I_1 can be neglected, provided *a* is big enough, i.e., $|a| \gtrsim 10$, and hence $I_1 \approx i/a^2$. By making use of the same reasoning as above one can now verify that $I_2 \approx i/\tilde{a}^2$, where now $\tilde{a} \equiv \Lambda_c - b_A - \ln(\Lambda_c \tau)/2\pi$ with $\Lambda_c \equiv \omega_c/\Gamma_0$. Therefore, the contribution due to I_2 can be neglected in comparison with I_1 , for any reasonable range of τ , provided that $\Lambda_c \gg \tilde{b}_A$.

APPENDIX C: ASYMPTOTIC EXPANSION OF J_1 AND J_2

Here we first consider the integral J_1 as defined in Eq. (68) in Sec. III D for large values of τ where we, due to the presence of the factor e^{-s} in the integrand of J_1 , make use of the approximations

$$N_0(-is/\tau) \approx -b_A \left(1+i\frac{s}{b_A\tau}\delta\right),$$
 (C1)

with $\delta \equiv 1 + \ln(\Lambda_c)/2\pi b_A$, as well as

$$N_1(-is/\tau) \approx -b_A \left(1+i\frac{s}{b_A\tau}\delta\right) + \frac{s}{b_A\tau},$$
 (C2)

as τ becomes large. A series expansion in $s/b_A \tau$ then leads to the convergent integral

$$J_1 \approx \frac{1}{b_A^3 \tau} \int_0^\infty ds \, e^{-s} \frac{s}{(1+is\delta/b_A \tau)^2}.$$
 (C3)

The effective expansion time parameter is then actually $b_A \tau$ instead of τ . We therefore find that $J_1 \approx 1/b_A^3 \tau$ for sufficiently large values of the parameter $b_A \tau$, i.e., $b_A \tau \gtrsim 1 + \ln(\Lambda_c)/2\pi b_A$. In the opposite limit J_1 will be suppressed by factors like $\ln(\delta/b_A \tau)/(\delta/b_A \tau)^2$ in addition to the $1/b_A^3 \tau$ behavior. More care has to be taken when considering the integral J_2 . For large values of τ , we then make use of the expansions

$$N_0(\Lambda_c - is/\tau) \approx \Lambda\left(a - \frac{1}{4}i + \frac{\ln s}{2\pi}\right),$$
 (C4)

with a as in Eq. (B3), as well as

$$N_1(-is/\tau) \approx \Lambda\left(a + \frac{3}{4}i + \frac{\ln s}{2\pi}\right).$$
 (C5)

Here we have made use of the simple fact that $\Lambda_c = \Lambda b_A$. As long as |a| is big enough it can now be argued that we can neglect the $\ln s/2\pi$ term in the N_0 and N_1 functions above. This is so since for large $s \gtrsim e^{2\pi |a|}$ the integrand of J_2 is exponentially suppressed, while for sufficiently small $s \lesssim e^{-2\pi |a|}$ there will only be an exponentially small range of integration. Proceeding in this manner and by series expansions in -i/4a and 3i/4a, we then find that the dominant contribution to J_2 is imaginary and that $J_2 \approx 1/i\tilde{\Lambda}a^2$. By numerical methods we find that this is indeed an excellent approximation if $|a| \gtrsim 10$ when evaluating $|c_e(t)|^2$.

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