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Comparison of density matrix and state vector approaches to dissipative evolution of hyperfine levels coupled to optical and radio-frequency fields in the two-level approximation

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We consider a three-level atom interacting with two optical and one microwave fields in the adiabatic approximation resulting in a simplified description in the terms of a two-level system. Working within the reduced density operator framework, we assume this two-level system to be affected by two types of environment, described by some ad hoc non-Hermitian Hamiltonian and Gorini-Kossakowski-Sudarshan-Lindblad's models. We compare the three types of dissipative evolution which can occur: driven by equations for a normalized density matrix, a non-normalized density matrix, and a normalized state vector. Using the latter type, we derive an effective Hamiltonian, which encodes information about not only the Hamiltonian part of an original master equation but also its non-Hamiltonian (Liouvillian) part. The Hamiltonian turns out to be dependent on the wavefunction itself: the effects of above-mentioned environments induce, respectively, cubic and quintic nonlinear terms. For evolutions driven by density operators, we study various indicators of quantum purity. It is shown that if a trace of density operator is not conserved then conventional von Neumann entropy can no longer be used as a purity indicator; therefore we introduce the purity-normalized definition of quantum statistical entropy.

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1. INTRODUCTION

In laser-excited Raman interaction experiments, atoms are pumped into a dressed state, which is a linear combination of hyperfine levels which have a separation of microwave radio frequency (rf) [1]. A microwave rf field can also be applied to excite the optical Raman trapped state.

A simple model of this process is based on a three-level Λ-type configuration with two optical fields and a microwave field [2, 3]. In this scheme, one assumes that the lowest hyperfine state $|1\rangle$ with an optical frequency is long-lived, the other hyperfine state $|3\rangle$ with a microwave frequency is also long-lived, whereas the excited state $|2\rangle$ coupled to an optical frequency field is short-lived [4].

Because of the short life of the excited state, one can consider its adiabatic elimination and arrive at the model of an atom with two hyperfine energy levels dressed by the rf field.

These and similar three- and two-level (TLS) configurations demonstrate a large number of physical effects related to quantum coherence. One of them is the rf-assisted electromagnetically induced transparency (EIT) phenomenon, where a microwave field affects the coherence of two ground states thus causing a change in the maximum transmission of a probe field [5, 6]. Another example is the stimulated Raman adiabatic passage (STIRAP) process, which is a method of state-to-state coherent control by using partially overlapping pulses to produce complete population transfer between two quantum states [7–9]. Yet another example are semiconductor quantum dots in electromagnetic cavity, which are important in quantum computer implementations [10–12].

In this paper, we consider a Λ-type three-level configuration and consider its adiabatic approximation, referred to as Λ-TLS model. In Sec. 2 we describe the model, both in its original and approximated TLS description. In sections 3 and 4 we formulate and apply to this model the various generalizations of the von Neumann and Schrödinger equations for open systems, assuming some *ad hoc* types of environment effects. We derive observables and various indicators for three types of dissipative evolution assuming the same initial conditions and values of parameters. Results are discussed and conclusions are drawn in Sec. 5.

2. THE MODEL

Let us consider a Λ-type three-level configuration where two hyperfine states $|1\rangle$ and $|3\rangle$ are coupled by a radio-frequency field, whereas two optical fields couple them to the state $|2\rangle$, which is the excited state of the system [3–6, 13]. The original Hamiltonian of the configuration can be written in the rotating frame of the optical fields, then it gets simplified by adding a Hermitian adjoint part, which eventually results in the following

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model:

$$\hat{H}^{(3)} = \hbar\Delta_1|1\rangle\langle 1| + \hbar\Delta_2|3\rangle\langle 3| - \hbar\omega_1(|1\rangle\langle 2| + |2\rangle\langle 1|) - \hbar\omega_2(|3\rangle\langle 2| + |2\rangle\langle 3|) - \hbar\omega_{\rm rf}\left(e^{i\phi}|1\rangle\langle 3| + e^{-i\phi}|3\rangle\langle 1|\right) - \hbar\Delta_+\hat{I}, \qquad (1)$$

where ω_1 and ω_2 are Rabi frequencies of the optical fields, $\omega_{\rm rf}$ and ϕ are, respectively, the amplitude and phase of microwave field coupling, Δ_1 and Δ_2 are optical detunings, $\Delta_+ = (\Delta_1 + \Delta_2)/2$, and $\hat{I}$ denotes the unit operator of an appropriate rank throughout the paper.

From the corresponding Schrödinger equation,

$$i\hbar\,\partial_t\,|\psi\rangle = \hat{H}^{(3)}|\psi\rangle, \qquad (2)$$

one derives differential equations for wavefunctions in a $|a\rangle$'s representation:

$$i\partial_t \begin{pmatrix}\psi_1\\ \psi_2\\ \psi_3\end{pmatrix} = \begin{pmatrix}\Delta_1 - \Delta_+ & -\omega_1 & -\omega_{\rm rf}\,e^{i\phi}\\ -\omega_1 & -\Delta_+ & -\omega_2\\ -\omega_{\rm rf}\,e^{-i\phi} & -\omega_2 & \Delta_2 - \Delta_+\end{pmatrix}\begin{pmatrix}\psi_1\\ \psi_2\\ \psi_3\end{pmatrix}, \qquad (3)$$

where $\psi_a(t) = \langle a|\psi(t)\rangle$, $a = 1, 2, 3$.

Furthermore, if the excited state's wavefunction slowly varies in time, then one can impose the adiabatic approximation $\partial_t\psi_2 = 0$. From Eqs. (3) we then obtain

$$\psi_2 = -(\omega_1\psi_1 + \omega_2\psi_3)/\Delta_+, \qquad (4)$$

which allows us to reduce our model to the two-level system

$$i\hbar\partial_t \begin{pmatrix}\psi_1\\ \psi_3\end{pmatrix} = \hat{H}^{(2)}\begin{pmatrix}\psi_1\\ \psi_3\end{pmatrix}, \qquad (5)$$

with the Hamiltonian

$$\hat{H}^{(2)} = \hbar\begin{pmatrix}\Delta_1 + \frac{\omega_1^2}{\Delta_+} & \frac{\omega_1\omega_2}{\Delta_+} - \omega_{\rm rf}\,e^{i\phi}\\ \frac{\omega_1\omega_2}{\Delta_+} - \omega_{\rm rf}\,e^{-i\phi} & \Delta_2 + \frac{\omega_2^2}{\Delta_+}\end{pmatrix} = \hbar\omega_+\begin{pmatrix}0 & 1 - \Omega\,e^{i\phi}\\ 1 - \Omega\,e^{-i\phi} & \chi\end{pmatrix}, \qquad (6)$$

where we denoted $\omega_+ = \omega_1\omega_2/\Delta_+$, $\Omega = \omega_{\rm rf}/\omega_+$, and $\chi = \omega_2/\omega_1 - \omega_1/\omega_2$ is the measure of difference between optical frequencies. The second matrix in this Eq. (6) is obtained by subtracting a constant term which does not affect energy eigenvalues. This Hamiltonian will be referred to as Λ-TLS model in what follows.

To summarize this section, equations (5) and (6) define the approximate model as a two-level atom affected by three electromagnetic fields, but without other environment effects. The latter will be introduced in the next sections using the quantum-statistical approach.

3. GENERAL EQUATIONS

The quantum-statistical approach adopted in this paper is a framework for formulating models dealing exclusively with the degrees of freedom of a subsystem, whilst taking into consideration the effect of the environment by the induced terms in the subsystem's evolution equations. This formalism is particularly useful if one does not have full information about the environment and does not want to assume it *a priori*, but rather attempts to find the induced terms which provide the best agreement with experimental data.

We thus regard the reduced density operator of a subsystem as a primary operator for dynamical and statistical descriptions. If the density operator is normalized,

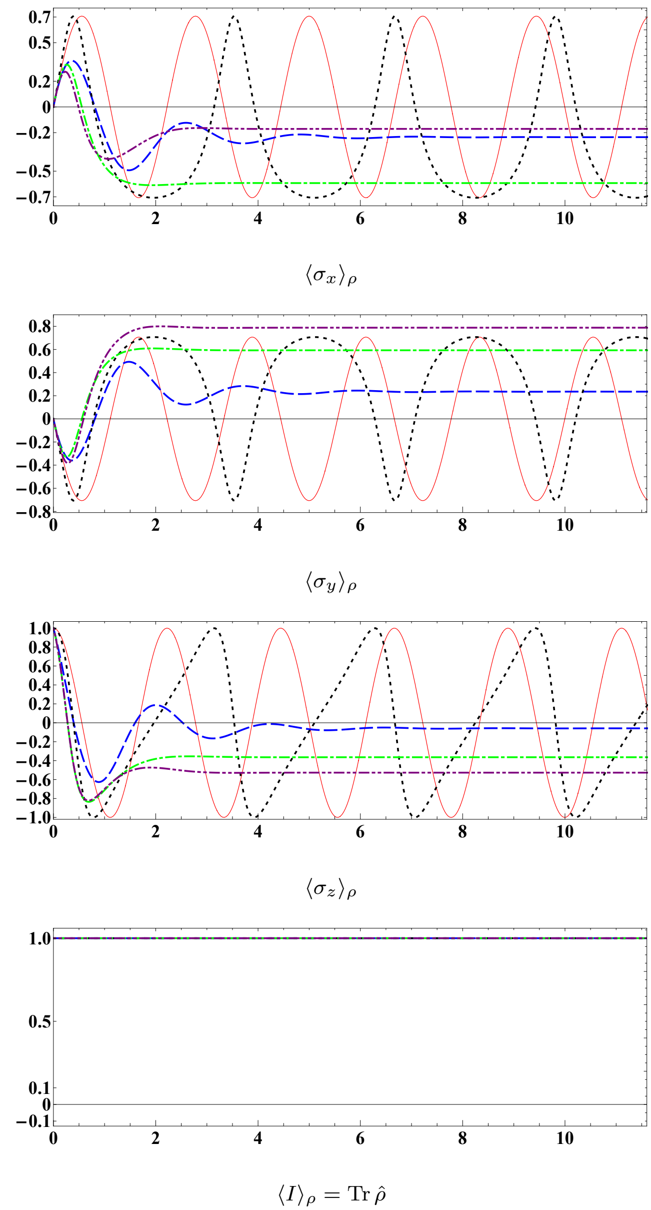

FIG. 1: Spin averages and normalized density operator's trace as functions of $\omega_+ t$, at the following values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$, respectively: 1, $\pi/2$, 0, 0, 0 (solid red line), 1, $\pi/2$, 0, 0, 1 (dashed blue line), 1, $\pi/2$, 0, 1, 0 (dotted black line), 1, $\pi/2$, 0, 1, 1 (dash-dotted green line), 1/2, $\pi/2$, 1, 1, 1 (dash-double-dotted purple line).

then we denote it by $\hat{\rho}$, otherwise by $\hat{\varrho}$. The relation between these notions is straightforward: $\hat{\rho} = \hat{\varrho}/\text{Tr}\,\hat{\varrho}$. Correspondingly, definitions for statistical mean values are $\langle \hat{O} \rangle_\rho \equiv \text{Tr}\,(\hat{\rho}\,\hat{O})$, $\langle \hat{O} \rangle_\varrho \equiv \text{Tr}\,(\hat{\varrho}\,\hat{O}) = \langle \hat{O} \rangle_\rho \,\text{Tr}\,\hat{\varrho}$, where subscripts indicate which density operator is used for deriving.

We assume that the effect of the environment upon a (sub)system can be introduced in two different (not mutually exclusive) ways [14].

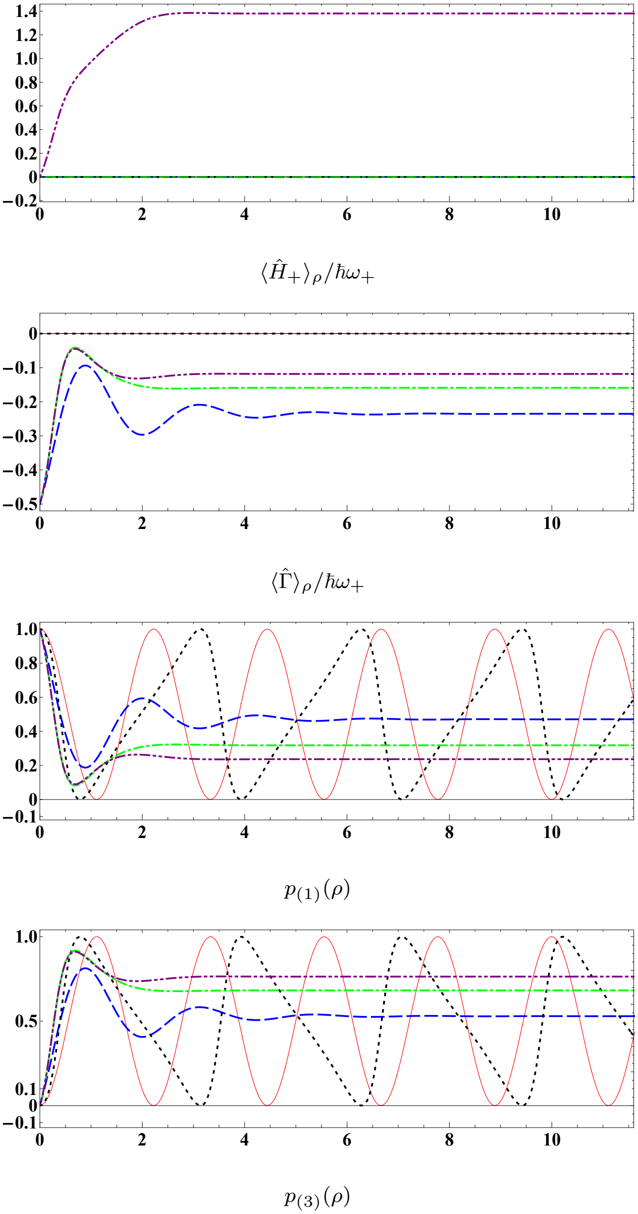

FIG. 2: Mean values of energy, decay operator, and populations of levels 1 and 3, under normalized density operator evolution, as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1. Plot of the population difference $p_{(1)}(\rho) - p_{(3)}(\rho) = \langle \sigma_z \rangle_\rho$ can be found in the previous figure.

- The first type of dissipation is described by using the Markovian approximation and quantum Markov semigroup. According to the Gorini-Kossakowski-Sudarshan-Lindblad (GKSL), or simply Lindblad, approach [15–18], it can be introduced by adding the non-Hamiltonian (Liouvillian) term to the von Neumann evolution equation for a density operator $\hat{\vartheta}$. This term, often referred to as the Lindblad's *dissipator*, reads:

$$\sum_k \gamma_k \left(\hat{L}_k \hat{\vartheta} \hat{L}_k^\dagger - \frac{1}{2} \{ \hat{\vartheta},\, \hat{L}_k^\dagger \hat{L}_k \} \right) = \frac{1}{2} \sum_k \gamma_k \left([\hat{L}_k,\, \hat{\vartheta} \hat{L}_k^\dagger] + [\hat{L}_k \hat{\vartheta},\, \hat{L}_k^\dagger] \right), \qquad (7)$$

where $\hat{L}_k$ are called the collapse or jump operators, γ's are non-negative couplings, square and curly brackets denote, respectively, commutator and anticommutator, the summation index runs $k = 1, \dots, N^2 - 1$, where N being the dimension of a system. Notice that the GKSL term is traceless

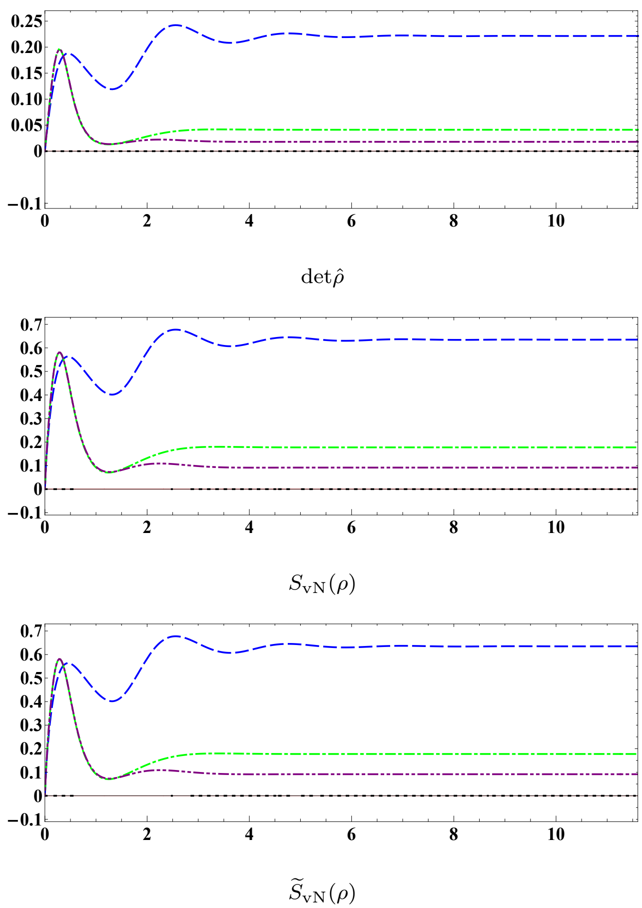

FIG. 3: Purity measures and entropy functions under normalized density operator evolution as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1.

and linear with respect to the density matrix.

- To introduce the dissipation effects of the second type, we assume that the environment can induce not only Hermitian but also anti-Hermitian corrections to the subsystem's Hamiltonian [19, 20], such that the resulting Hamiltonian operator becomes non-Hermitian:

$$\hat{\mathcal{H}} = \hat{H}_+ + \hat{H}_-, \quad \hat{H}_\pm \equiv \tfrac{1}{2}(\hat{\mathcal{H}} \pm \hat{\mathcal{H}}^\dagger) = \pm \hat{H}_\pm^\dagger, \tag{8}$$

where $\hat{H}_\pm$ are self- and skew-adjoint components; it is also convenient to introduce the self-adjoint operator $\hat{\Gamma} \equiv i\hat{H}_-$. Nowadays non-Hermitian Hamiltonians (NH) can be found in nearly all branches of quantum physics, to mention only a very recent literature [21–38]. Note that the dissipation models described by NH Hamiltonians are not necessarily related to the Markovian approximation or to quantum jumps *per se*, instead they must be regarded as a general framework. Below we will show that these models induce Liouvillian terms in a generalized von Neumann equation, which are nonlinear with respect to a density matrix and in general nonlocal, unlike the Markovian case (7).

As mentioned above and elsewhere, GKSL approach crucially relies on the Markovian approximation, whereas the NH Hamiltonian formalism does not. One the other hand, one cannot guarantee that all realistic quantum systems are predominantly Markovian. Therefore, a consistent way would be to begin with a maximally general framework available, such as a combination of GKSL and NH terms, construct a working model and compare its predicted curves for observables with actual measurements. This workflow would provide empirical bounds for the model's parameters, confirm or rule out some of the terms.

Furthermore, in the work [39] it was established that in the presence of the above-mentioned types of dissipative effects, a general quantum system can be described in at least five different ways: two ways if one takes into account both mixed and pure states and works in terms of density matrices, and three ways if one restricts oneself to pure states and deals with state vectors only. Another key difference between these types is the norm's behaviour of a density matrix or state vector during time evolution: the norm is conserved for two types, whereas for other three it is not.

However, in the case of two-level systems, pure states can be shown to have a unit norm at all times, see Eqs. (A.13) and (A.14), which rules out all state vector evolution scenarios with a non-conserved norm. Keeping in mind the Λ-TLS applications, we will omit those scenarios in what follows. The remaining evolution equations to be considered, two for density operators and one for a state vector, are:

i. *Normalized density operator evolution.* The equation reads

$$\begin{aligned}\frac{d}{dt}\hat{\rho} &= -\frac{i}{\hbar}\left(\hat{\mathcal{H}}\hat{\rho} - \hat{\rho}\,\hat{\mathcal{H}}^\dagger\right) + \frac{i}{\hbar}\langle \hat{\mathcal{H}} - \hat{\mathcal{H}}^\dagger\rangle_\rho\, \hat{\rho} + \sum_k \gamma_k \left(\hat{L}_k \hat{\rho} \hat{L}_k^\dagger - \frac{1}{2}\{\hat{\rho},\, \hat{L}_k^\dagger \hat{L}_k\}\right) \\ &= -\frac{i}{\hbar}\left[\hat{H}_+, \hat{\rho}\right] - \frac{1}{\hbar}\left\{\hat{\Gamma}, \hat{\rho}\right\} + \frac{2}{\hbar}\langle\hat{\Gamma}\rangle_\rho\, \hat{\rho} + \sum_k \gamma_k \left(\hat{L}_k \hat{\rho} \hat{L}_k^\dagger - \frac{1}{2}\{\hat{\rho},\, \hat{L}_k^\dagger \hat{L}_k\}\right),\end{aligned} \tag{9}$$

where $\langle\hat{\Gamma}\rangle_\rho = \mathrm{tr}(\hat{\rho}\,\hat{\Gamma})$ according to our notations. One can check that the trace of the density operator $\hat{\rho}$ is indeed conserved during evolution.

ii. *Non-normalized density operator evolution.* The evolution equation reads:

$$\frac{d}{dt}\hat{\varrho} = -\frac{i}{\hbar}\left(\hat{\mathcal{H}}\hat{\varrho} - \hat{\varrho}\,\hat{\mathcal{H}}^\dagger\right) + \sum_k \gamma_k \left(\hat{L}_k \hat{\varrho} \hat{L}_k^\dagger - \frac{1}{2}\{\hat{\varrho},\, \hat{L}_k^\dagger \hat{L}_k\}\right) = -\frac{i}{\hbar}\left[\hat{H}_+, \hat{\varrho}\right] - \frac{1}{\hbar}\left\{\hat{\Gamma}, \hat{\varrho}\right\} + \sum_k \gamma_k \left(\hat{L}_k \hat{\varrho} \hat{L}_k^\dagger - \frac{1}{2}\{\hat{\varrho},\, \hat{L}_k^\dagger \hat{L}_k\}\right), \tag{10}$$

which is different from Eq. (9) by the absent compensating term proportional to $\langle\hat{\Gamma}\rangle$. Without this term, the trace of $\hat{\varrho}$ is not conserved during evolution, which leads to a non-invariant probability space. As a result, non-normalized density operators describe open systems which irreversibly decay or diverge. Some illustrative comparative examples between non-normalized and normalized density operators can be found in Ref. [40].

iii. *Normalized state vector evolution.* It is described by the generalization of the Schrödinger equation, which takes into account both non-Hermitian Hamiltonian corrections and GKSL-type quantum jumps:

$$i\hbar\,\partial_t\,|\Psi\rangle = \hat{\mathcal{H}}_{\text{eff}}\,|\Psi\rangle, \tag{11}$$

where $|\Psi\rangle$ is the normalized state vector and the effective Hamiltonian reads:

$$\hat{\mathcal{H}}_{\text{eff}} \equiv \hat{\mathcal{H}} - \frac{1}{2}\langle \hat{\mathcal{H}} - \hat{\mathcal{H}}^\dagger \rangle_\Psi \hat{I} + \frac{i}{2}\hbar \sum_k \gamma_k \left[\left(\langle \hat{L}_k^\dagger \rangle_\Psi - \hat{L}_k^\dagger \right) \hat{L}_k - \langle \hat{L}_k^\dagger \rangle_\Psi \langle \hat{L}_k \rangle_\Psi + \langle \hat{L}_k^\dagger \hat{L}_k \rangle_\Psi \right] = \hat{H}_{\text{eff}} - i\,\hat{\Gamma}_{\text{eff}}, \quad (12)$$

$$\hat{H}_{\text{eff}} \equiv \frac{1}{2}\left(\hat{\mathcal{H}}_{\text{eff}} + \hat{\mathcal{H}}_{\text{eff}}^\dagger \right) = \hat{H}_+ + \frac{i}{4}\hbar \sum_k \gamma_k \left(\langle \hat{L}_k^\dagger \rangle_\Psi \hat{L}_k - \hat{L}_k^\dagger \langle \hat{L}_k \rangle_\Psi \right),$$

$$\hat{\Gamma}_{\text{eff}} \equiv \frac{i}{2}\left(\hat{\mathcal{H}}_{\text{eff}} - \hat{\mathcal{H}}_{\text{eff}}^\dagger \right) = \hat{\Gamma} - \langle \hat{\Gamma} \rangle_\Psi \hat{I} - \frac{1}{4}\hbar \sum_k \gamma_k \left[\langle \hat{L}_k^\dagger \rangle_\Psi \hat{L}_k + \hat{L}_k^\dagger \langle \hat{L}_k \rangle_\Psi - 2\hat{L}_k^\dagger \hat{L}_k - 2\left(\langle \hat{L}_k^\dagger \rangle_\Psi \langle \hat{L}_k \rangle_\Psi - \langle \hat{L}_k^\dagger \hat{L}_k \rangle_\Psi \right) \hat{I} \right],$$

where $\hat{H}_{\text{eff}}$ and $\hat{\Gamma}_{\text{eff}}$ are self-adjoint operators, and $\langle\ .\ \rangle_\Psi = \langle \Psi|\ .\ |\Psi\rangle$ is a notation for wave-mechanical mean values. This state vector is related to the density matrices via the procedures of pure-state reduction and normalization: $|\Psi\rangle = \langle \mho|\mho\rangle^{-1/2}\,|\mho\rangle = \langle \Phi|\Phi\rangle^{-1/2}\,|\Phi\rangle$, where $|\mho\rangle$ and $|\Phi\rangle$ are special cases of, respectively, non-normalized and normalized density matrices: $\hat{\varrho} \to |\mho\rangle\langle\mho|$ and $\hat{\rho} \to |\Phi\rangle\langle\Phi|$, see Ref. [39] for further details. One can directly check from Eq. (11) that the norm of $|\Psi\rangle$ is indeed conserved at all times

$$\partial_t \langle \Psi|\Psi\rangle = 0,\ \langle \Psi|\Psi\rangle = 1, \quad (13)$$

and also that the gain and loss are balanced, on average, during this type of evolution:

$$\langle \hat{\mathcal{H}}_{\text{eff}} \rangle_\Psi = \langle \hat{H}_+ \rangle_\Psi, \quad \langle \hat{\Gamma}_{\text{eff}} \rangle_\Psi = 0, \quad (14)$$

which is expected if a state vector's norm is conserved.

Evolution types (i-iii) can be found in the same system, though in different situations. Equation (9) is useful when one needs to know the evolution of both pure and mixed states of a system if the probability space of the latter is expected to be invariant. Equation (10) has a similar application, except that it is used when the system is known to experience an unbalanced gain or loss of particles, thus leading to its irreversible decay or blow-up.

Finally, equation (11) is useful if one needs to neglect statistical effects and focus entirely on quantum-mechanical evolution with conserved purity and invariant probability space. This equation is also useful to derive an effective Hamiltonian, as defined by the formula (12), which contains information, or some portion thereof, about not only Hamiltonian but also Liouvillian terms, originally stored in Eq. (9).

4. Λ-TLS WITH DISSIPATION

In this section we introduce environment effects to the model (6), and, using the approaches from the previous section, we describe three types of dissipative evolution which can occur. We therefore assign the following Hamiltonian operator

$$\hat{H}_+ = \hat{H}^{(2)} = \hbar\omega_+ \begin{pmatrix} 0 & 1 - \Omega\, \mathrm{e}^{i\phi} \\ 1 - \Omega\, \mathrm{e}^{-i\phi} & \chi \end{pmatrix} \quad (15)$$
$$= \hbar\omega_+ \left[(1 - \Omega\cos\phi)\hat{\sigma}_x + \Omega \sin\phi\, \hat{\sigma}_y - \frac{\chi}{2}\left(\hat{\sigma}_z - \hat{I} \right) \right],$$

to be used in all the evolution equations to be considered in what follows.

This Hamiltonian describes a two-level atom interacting with three electromagnetic fields, but absent other kinds of environment effects. The latter can be added to our model in two different ways: by adding a skew-adjoint term to the original Hamiltonian:

$$\hat{\mathcal{H}} = \hat{H}^{(2)} - i\hat{\Gamma}, \quad \hat{\Gamma} = \hbar\gamma_0 \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} = \hbar\gamma_0 \hat{\sigma}_z, \quad (16)$$

and assuming that the system can also undergo GKSL-type quantum jumps, driven by the following operator:

$$\hat{L}_k = \begin{cases} \hat{\sigma}_-, & k = 1, \\ 0, & k > 1, \end{cases} \qquad \gamma_k = \begin{cases} \gamma_1, & k = 1, \\ 0, & k > 1, \end{cases} \quad (17)$$

where we use notations from the Appendix. For the calculations that follow, we introduce the rescaled decay coefficients $\tilde{\gamma}_0 = \gamma_0/\omega_+$ and $\tilde{\gamma}_1 = \gamma_1/\omega_+$, and the shorthand notation $\tilde{\gamma}_c = \tilde{\gamma}_1 + 2\tilde{\gamma}_0 = (\gamma_1 + 2\gamma_0)/\omega_+$.

Note that both models (16) and (17) are not the most general ones, but they are relatively simple for the comparative purposes of this paper. Further discussion will be given in the concluding section.

For the sake of comparison, we assume common sets of values for model parameters $\{\Omega,\ \phi,\ \chi,\ \tilde{\gamma}_0,\ \tilde{\gamma}_1\}$ to be used in computations of evolution equations: $\{1,\ \pi/2,\ 0,\ 0,\ 0\}$, $\{1,\ \pi/2,\ 0,\ 0,\ 1\}$, $\{1,\ \pi/2,\ 0,\ 1,\ 0\}$, $\{1,\ \pi/2,\ 0,\ 1,\ 1\}$, and $\{1/2,\ \pi/2,\ 1,\ 1,\ 1\}$. Their choice will be explained in the concluding section.

Finally, we assume a common initial condition:

$$\hat{\rho}(0) = \hat{\varrho}(0) = |\Psi(0)\rangle\langle\Psi(0)| = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} = \frac{1}{2}\Big(\hat{\sigma}_z + \hat{I}\Big), \quad (18)$$

which is the pure state corresponding to the ground state of the Λ-TLS system. Here the timestamp $t = 0$ marks the moment of time when the system starts to evolve.

4.1. Evolution of normalized density matrix

Here we consider the evolution of the system (16)-(17) from the initial state (18), which is described by the master equation (9). The matrix $\hat{\rho}$ can be represented in terms of spin averages

$$\hat{\rho} = \frac{1}{2}\begin{pmatrix} \mathrm{Tr}\,\hat{\rho} + \langle\sigma_z\rangle_\rho & \langle\sigma_x\rangle_\rho - i\langle\sigma_y\rangle_\rho \\ \langle\sigma_x\rangle_\rho + i\langle\sigma_y\rangle_\rho & \mathrm{Tr}\,\hat{\rho} - \langle\sigma_z\rangle_\rho \end{pmatrix}, \quad (19)$$

according to Eq. (A.5). In this formula we keep $\mathrm{Tr}\,\hat{\rho}$ as an unknown although we know that

$$\mathrm{Tr}\,\hat{\rho}(t) = 1, \quad (20)$$

because we want to obtain this value dynamically, as a solution of evolution equations.

For the ansatz (19), equations (9), (16)-(17) yield a set of differential equations for four unknown functions

$$\omega_+^{-1}\frac{d}{dt}\begin{pmatrix} \langle\sigma_x\rangle_\rho \\ \langle\sigma_y\rangle_\rho \\ \langle\sigma_z\rangle_\rho \\ \mathrm{Tr}\,\hat{\rho} \end{pmatrix} = \begin{pmatrix} 2\tilde{\gamma}_0\langle\sigma_z\rangle_\rho - \tilde{\gamma}_1/2 & \chi & 2\Omega\sin\phi & 0 \\ -\chi & 2\tilde{\gamma}_0\langle\sigma_z\rangle_\rho - \tilde{\gamma}_1/2 & -2(1-\Omega\cos\phi) & 0 \\ -2\Omega\sin\phi & 2(1-\Omega\cos\phi) & 2\tilde{\gamma}_0\langle\sigma_z\rangle_\rho - \tilde{\gamma}_1 & -\tilde{\gamma}_c \\ 0 & 0 & -2\tilde{\gamma}_0 & 2\tilde{\gamma}_0\langle\sigma_z\rangle_\rho \end{pmatrix}\begin{pmatrix} \langle\sigma_x\rangle_\rho \\ \langle\sigma_y\rangle_\rho \\ \langle\sigma_z\rangle_\rho \\ \mathrm{Tr}\,\hat{\rho} \end{pmatrix}, \quad (21)$$

with the initial conditions

$$\langle\sigma_x\rangle_\rho|_{t=0} = \langle\sigma_y\rangle_\rho|_{t=0} = 0, \quad \langle\sigma_z\rangle_\rho|_{t=0} = \mathrm{Tr}\,\hat{\rho}(0) = 1, \quad (22)$$

which can be deduced from Eqs. (18) and (19). One can check that the normalization condition (20) is indeed a solution of Eqs. (21), therefore the corresponding equation can be eliminated from the system when doing analytical computations (for numerical computations such elimination is not necessary).

Notice that Eqs. (21) are nonlinear differential equations, due to quadratic terms $\langle\sigma_i\rangle_\rho\langle\sigma_z\rangle_\rho$ which occur. This, however does not cause technical complications for analytical computations, because the equations can always be transformed to linear differential equations for functions $f_i(t) = \langle\sigma_i\rangle_\rho\,\mathrm{Tr}\,\hat{\rho}$ $(i = x, y, z)$, solved, then back-transformed.

Furthermore, solutions of the system (21), (22) are plotted in Fig. 1 for the various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, and $\tilde{\gamma}_1$ of the model, whereas the parameter ω_+ defines the time scale.

Using these solutions, we can compute further physical observables, such as the mean value of the energy defined as the self-adjoint Hamiltonian part (16):

$$\langle\hat{H}_+\rangle_\rho = \hbar\omega_+\left[(1-\Omega\cos\phi)\langle\sigma_x\rangle_\rho + \Omega\sin\phi\,\langle\sigma_y\rangle_\rho - \frac{1}{2}\chi(\langle\sigma_z\rangle_\rho - \mathrm{Tr}\,\hat{\rho})\right], \quad (23)$$

mean value of the decay operator (16):

$$\langle\hat{\Gamma}\rangle_\rho = \hbar\gamma_0\langle\sigma_z\rangle_\rho, \quad (24)$$

and populations of levels $|1\rangle$ and $|3\rangle$:

$$p_{(3)}(\rho) = \mathrm{Tr}\,\hat{\rho} - p_{(1)}(\rho) = \frac{1}{2}(\mathrm{Tr}\,\hat{\rho} - \langle\sigma_z\rangle_\rho), \quad (25)$$

according to formulae (A.2) and (A.3). Their evolution plots can be found in Fig. 2.

Last but not least, it is useful to know the behaviour of purity measure values, such as the determinant of density matrix:

$$\det\hat{\rho} = \frac{1}{4}\left[(\mathrm{Tr}\,\hat{\rho})^2 - \langle\sigma\rangle_\rho^2\right] = \frac{1}{4}\left(1 - \langle\sigma\rangle_\rho^2\right), \quad (26)$$

according to Eqs. (A.8) and (20), and entropy functions

$$S_{\mathrm{vN}}(\rho) = \widetilde{S}_{\mathrm{vN}}(\rho) = -\frac{1}{2}[(1+\langle\sigma\rangle_\rho)\ln(1+\langle\sigma\rangle_\rho) + (1-\langle\sigma\rangle_\rho)\ln(1-\langle\sigma\rangle_\rho)] + \ln 2, \quad (27)$$

according to Eqs. (A.18), (A.22) and (20). Their evolution plots are given in Fig. 3. Notice that the normalization (20) is the reason why the von Neumann entropy coincides with the purity-normalized von Neumann entropy in this case.

Further discussion of these results is postponed until section 5.

4.2. Evolution of non-normalized density matrix

Let us consider the evolution of the same system (16)-(17) from the same initial state (18), which is described by the master equation (10). The matrix $\hat{\varrho}$ can be represented in terms of spin averages

$$\hat{\varrho} = \frac{1}{2} \begin{pmatrix} \text{Tr}\,\hat{\varrho} + \langle\sigma_z\rangle_\varrho & \langle\sigma_x\rangle_\varrho - i\langle\sigma_y\rangle_\varrho \\ \langle\sigma_x\rangle_\varrho + i\langle\sigma_y\rangle_\varrho & \text{Tr}\,\hat{\varrho} - \langle\sigma_z\rangle_\varrho \end{pmatrix}, \tag{28}$$

according to Eq. (A.5).

For the representation (28), equations (10), (16)-(17) yield a set of linear differential equations for four unknown functions

$$\omega_+^{-1} \frac{d}{dt} \begin{pmatrix} \langle\sigma_x\rangle_\varrho \\ \langle\sigma_y\rangle_\varrho \\ \langle\sigma_z\rangle_\varrho \\ \text{Tr}\,\hat{\varrho} \end{pmatrix} = \begin{pmatrix} -\tilde{\gamma}_1/2 & \chi & 2\Omega\sin\phi & 0 \\ -\chi & -\tilde{\gamma}_1/2 & -2(1-\Omega\cos\phi) & 0 \\ -2\Omega\sin\phi & 2(1-\Omega\cos\phi) & -\tilde{\gamma}_1 & -\tilde{\gamma}_c \\ 0 & 0 & -2\tilde{\gamma}_0 & 0 \end{pmatrix} \begin{pmatrix} \langle\sigma_x\rangle_\varrho \\ \langle\sigma_y\rangle_\varrho \\ \langle\sigma_z\rangle_\varrho \\ \text{Tr}\,\hat{\varrho} \end{pmatrix}, \tag{29}$$

with the initial conditions

$$\langle\sigma_x\rangle_\varrho|_{t=0} = \langle\sigma_y\rangle_\varrho|_{t=0} = 0, \quad \langle\sigma_z\rangle_\varrho|_{t=0} = \text{Tr}\,\hat{\varrho}(0) = 1, \tag{30}$$

which can be deduced from Eqs. (18) and (28).

Solutions of the system (29), (30) are plotted in Fig. 4 for various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, and $\tilde{\gamma}_1$ of the model, whereas the parameter ω_+ defines the time scale.

Using these solutions, we can compute other physical observables such as the mean value of energy defined as the self-adjoint Hamiltonian part (16):

$$\langle\hat{H}_+\rangle_\varrho = \hbar\omega_+ \left[(1-\Omega\cos\phi)\langle\sigma_x\rangle_\varrho + \Omega\sin\phi\,\langle\sigma_y\rangle_\varrho - \frac{1}{2}\chi(\langle\sigma_z\rangle_\varrho - \text{Tr}\,\hat{\varrho}) \right], \tag{31}$$

mean value of the decay operator (16):

$$\langle\hat{\Gamma}\rangle_\varrho = \hbar\gamma_0\langle\sigma_z\rangle_\varrho, \tag{32}$$

and populations of levels $|1\rangle$ and $|3\rangle$:

$$p_{(3)}(\varrho) = \text{Tr}\,\hat{\varrho} - p_{(1)}(\varrho) = \frac{1}{2}(\text{Tr}\,\hat{\varrho} - \langle\sigma_z\rangle_\varrho), \tag{33}$$

according to formulae (A.2) and (A.3). Their evolution profiles are given in Fig. 5.

Last but not least, it is useful to know the behaviour of purity measure values, such as the determinant of density matrix:

$$\det\hat{\varrho} = \frac{1}{4}\left[(\text{Tr}\,\hat{\varrho})^2 - \langle\sigma\rangle_\varrho^2\right], \tag{34}$$

defined according to Eq. (A.8); and entropy functions

$$S_{\text{vN}}(\varrho) = -\frac{1}{2}[(\text{Tr}\,\hat{\varrho} + \langle\sigma\rangle_\varrho)\ln(\text{Tr}\,\hat{\varrho} + \langle\sigma\rangle_\varrho) + (\text{Tr}\,\hat{\varrho} - \langle\sigma\rangle_\varrho)\ln(\text{Tr}\,\hat{\varrho} - \langle\sigma\rangle_\varrho)] + \ln 2\ \text{Tr}\,\hat{\varrho}, \tag{35}$$

$$\widetilde{S}_{\text{vN}}(\varrho) = S_{\text{vN}}(\varrho) + \text{Tr}\hat{\varrho}\ \ln(\text{Tr}\hat{\varrho}), \tag{36}$$

according to Eqs. (A.18) and (A.22). Their evolution plots are given in Fig. 6.

Notice from this figure that the canonical von Neumann entropy takes a zero value not only at $t = 0$, because the system's state is assumed to be initially pure, but sometimes also later, when the system is not in a pure state, cf. the panel for $\det\hat{\varrho}$. This illustrates what we established in the Appendix: if a trace of density matrix is not conserved then the vanishing $S_{\rm vN}$ is no longer a sufficient condition for a state to be pure. Instead, the necessary and sufficient condition for the latter is the vanishing purity-normalized entropy $\widetilde{S}_{\rm vN}$. Figure 6 shows that the behaviour of $\widetilde{S}_{\rm vN}$ does correlate with another purity measure, $\det\hat{\varrho}$.

More detailed discussion of these results can be found in section 5.

4.3. Evolution of normalized state vector

Let us consider now the evolution of the same system (16)-(17) from the same initial state (18), which is described by the generalized Schrödinger equation (11). Assuming the representation

$$|\Psi\rangle \mapsto \begin{pmatrix}\Psi_1\\ \Psi_3\end{pmatrix}, \quad \langle\Psi|\Psi\rangle = |\Psi_1|^2 + |\Psi_3|^2 = 1, \tag{37}$$

we obtain from Eqs. (11)-(17) the following evolution equation:

$$i\hbar\,\partial_t \begin{pmatrix}\Psi_1\\ \Psi_3\end{pmatrix} = \hat{\mathcal{H}}_{\rm eff}^{(2)} \begin{pmatrix}\Psi_1\\ \Psi_3\end{pmatrix}, \tag{38}$$

where the effective Hamiltonian and decay operator are, respectively:

$$\hat{\mathcal{H}}_{\rm eff}^{(2)} = \hat{H}^{(2)} - i\hat{\Gamma}_{\rm eff}, \tag{39}$$

$$\begin{aligned}\hat{\Gamma}_{\rm eff} &= \frac{\hbar\omega_+\tilde{\gamma}_1}{2}\begin{pmatrix}\left(1-|\Psi_1|^2\right)\left(1+|\Psi_1|^2+4\tilde{\gamma}_0/\tilde{\gamma}_1\right) & -\frac{1}{2}\Psi_1\Psi_3^*\\ -\frac{1}{2}\Psi_1^*\Psi_3 & -|\Psi_1|^2\left(|\Psi_1|^2+4\tilde{\gamma}_0/\tilde{\gamma}_1\right)\end{pmatrix}\\ &= \frac{\hbar\omega_+\tilde{\gamma}_1}{4}\left[f_c(\Psi)\hat{I} - f_+(\Psi)\hat{\sigma}_x - i f_-(\Psi)\hat{\sigma}_y + (1+4\tilde{\gamma}_0/\tilde{\gamma}_1)\,\hat{\sigma}_z\right],\end{aligned} \tag{40}$$

where we denoted $f_c(\Psi) = 1-2|\Psi_1|^4+4\tilde{\gamma}_0\left(1-2|\Psi_1|^2\right)/\tilde{\gamma}_1$ and $f_\pm(\Psi) = (\Psi_1\Psi_3^*\pm\Psi_1^*\Psi_3)/2$, and used the normalization (37), where deemed helpful. Notice that the effective Hamiltonian (39) depends on wavefunctions, therefore our Schrödinger equation is essentially nonlinear with respect to the wavefunction.

For solving, we must supplement these equations with the initial conditions:

$$|\Psi_1(0)| = 1,\ \ \Psi_3(0) = 0, \tag{41}$$

which originated from Eq. (18).

Equations (37)-(41) can be alternatively rewritten in terms of spin averages, by using the formulae

$$\begin{aligned}|\Psi_1|^2 &= \frac{1}{2}(\langle\sigma\rangle_\Psi + \langle\sigma_z\rangle_\Psi) = \frac{1}{2}(\langle\Psi|\Psi\rangle + \langle\sigma_z\rangle_\Psi),\\ |\Psi_3|^2 &= \frac{1}{2}(\langle\sigma\rangle_\Psi - \langle\sigma_z\rangle_\Psi) = \frac{1}{2}(\langle\Psi|\Psi\rangle - \langle\sigma_z\rangle_\Psi),\\ \frac{\Psi_3}{\Psi_1} &= \frac{\langle\sigma_x\rangle_\Psi + i\langle\sigma_y\rangle_\Psi}{\langle\sigma\rangle_\Psi + \langle\sigma_z\rangle_\Psi} = \frac{\langle\sigma_x\rangle_\Psi + i\langle\sigma_y\rangle_\Psi}{\langle\Psi|\Psi\rangle + \langle\sigma_z\rangle_\Psi},\end{aligned} \tag{42}$$

where

$$\langle\sigma\rangle_\Psi \equiv \left[\langle\sigma_x\rangle_\Psi^2 + \langle\sigma_y\rangle_\Psi^2 + \langle\sigma_z\rangle_\Psi^2\right]^{1/2} = \langle\Psi|\Psi\rangle = 1, \tag{43}$$

and we used Eqs. (A.12) and (A.16) from the Appendix.

Then Eqs. (37)-(41) transform into a set of differential equations for three spin averages as functions of time

$$\omega_+^{-1}\frac{d}{dt}\begin{pmatrix}\langle\sigma_x\rangle_\Psi\\ \langle\sigma_y\rangle_\Psi\\ \langle\sigma_z\rangle_\Psi\end{pmatrix} = \begin{pmatrix}-f(\sigma_z)\,\langle\sigma_z\rangle_\Psi & \chi & 2\Omega\sin\phi\\ -\chi & -f(\sigma_z)\,\langle\sigma_z\rangle_\Psi & -2(1-\Omega\cos\phi)\\ -2\Omega\sin\phi & 2(1-\Omega\cos\phi) & f(\sigma_z)\,(\langle\sigma_z\rangle_\Psi^2-1)\end{pmatrix}\begin{pmatrix}\langle\sigma_x\rangle_\Psi\\ \langle\sigma_y\rangle_\Psi\\ \langle\sigma_z\rangle_\Psi\end{pmatrix}, \tag{44}$$

where $f(\sigma_z) = \tilde{\gamma}_1(1-\langle\sigma_z\rangle_\Psi - 4\tilde{\gamma}_c/\tilde{\gamma}_1)/4 = -\tilde{\gamma}_1(\langle\sigma_z\rangle_\Psi + 3 + 8\tilde{\gamma}_0/\tilde{\gamma}_1)/4$, supplemented with the initial conditions

$$\langle\sigma_x\rangle_\Psi|_{t=0} = \langle\sigma_y\rangle_\Psi|_{t=0} = 0, \quad \langle\sigma_z\rangle_\Psi|_{t=0} = 1, \tag{45}$$

and the Bloch sphere constraint (43).

Solutions of the system (44), (45) are plotted in Fig. 7 for the various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, and $\tilde{\gamma}_1$ of the model, whereas the parameter ω_+ defines the time scale.

Using these solutions, we can compute other physical observables such as the mean value of energy defined as a self-adjoint part of the effective Hamiltonian (39)

$$\langle \hat{H}_+ \rangle_\Psi = \langle \hat{H}^{(2)} \rangle_\Psi = \hbar\omega_+ \left[(1 - \Omega\cos\phi) \langle \sigma_x \rangle_\Psi + \Omega \sin\phi \, \langle \sigma_y \rangle_\Psi - \frac{1}{2}\chi \left(\langle \sigma_z \rangle_\Psi - 1 \right) \right], \tag{46}$$

a mean value of the decay operator (cf. Eq. (14)):

$$\langle \hat{\Gamma}_{\text{eff}} \rangle_\Psi = \frac{\hbar\omega_+ \tilde{\gamma}_1}{2} |\Psi_1|^2 \left(1 - |\Psi_1|^2 - |\Psi_3|^2 \right) \left(1 + |\Psi_1|^2 + 4\tilde{\gamma}_0/\tilde{\gamma}_1 \right) = 0, \tag{47}$$

and populations of levels $|1\rangle$ and $|3\rangle$:

$$p_{(3)}(\Psi) = 1 - p_{(1)}(\Psi) = \frac{1}{2}(1 - \langle \sigma_z \rangle_\Psi), \tag{48}$$

according to formulae (A.2) and (A.3). Their evolution plots can be found in Fig. 8.

Further discussion of these results is given in the next section.

5. DISCUSSION AND CONCLUSION

In this paper, we considered an adiabatic approximation of the Λ configuration of a three-level atom in the presence of two optical and one microwave fields, which led us to a simplified description in terms of a two-level system.

We then imposed the presence of two types of dissipative environments, described by TLS models of the non-Hermitian and GKSL type defined through, respectively, Eqs. (16) and (17). They are not the most general ones: for realistic applications, they must be substantially expanded, especially the non-Hermitian part. In fact, the chosen NH dissipative coupling is too "weak" for modeling damping and decay processes, as will be discussed below. However, these environments are popular and simple enough for the comparative purposes of this paper.

Using the same (sub)system Hamiltonian, environment effects model and initial conditions, we compared three types of dissipative evolution which can occur: described by a normalized density matrix, a non-normalized density matrix, and a normalized state vector. Types of state vector evolution with non-conserved norm turned out to be unphysical in the TLS case, see Appendix for details, and thus were excluded in this report.

Evolution equations for the density matrices are in fact quantum master equations which are generalizations of the von Neumann equations for open systems, while the evolution equation for the normalized state vector is the generalized Schrödinger equation. Although the latter technically is simply a restriction to the evolution of pure states, it still has a useful purpose: it allows us to derive an effective Hamiltonian, which encodes information not only about the Hamiltonian part of a master equation but also about its non-Hamiltonian (Liouvillian) part. This effective Hamiltonian turns out to be not an ordinary operator, because it depends on mean values which contain a wavefunction, hence on a wavefunction too. For example, in our TLS model, NH and GKSL environments induce, respectively, cubic and quintic nonlinear terms with respect to a wavefunction, see Eq. (40).

For computations of observables and other averages we chose the following five sets of values for five parameters $\{\Omega,\ \phi,\ \chi,\ \tilde{\gamma}_0,\ \tilde{\gamma}_1\}$ of the model: $\{1,\ \pi/2,\ 0,\ 0,\ 0\}$ (labelled in plots by solid lines), $\{1,\ \pi/2,\ 0,\ 0,\ 1\}$ (dashed lines), $\{1,\ \pi/2,\ 0,\ 1,\ 0\}$ (dotted), $\{1,\ \pi/2,\ 0,\ 1,\ 1\}$ (dash-dotted), and $\{1/2,\ \pi/2,\ 1,\ 1,\ 1\}$ (dash-double-dotted).

The first set of values, where $\tilde{\gamma}_0 = \tilde{\gamma}_1 = 0$, refers to the case when all external environment effects are switched off. The second set of parameters refers to the case when NH dissipation is off but GKSL is on ($\tilde{\gamma}_0 = 0$, $\tilde{\gamma}_1 \neq 0$), third set is other way around ($\tilde{\gamma}_0 \neq 0$, $\tilde{\gamma}_1 = 0$), and fourth set is when both environment effects (16) and (17) are on ($\tilde{\gamma}_0 \neq 0$, $\tilde{\gamma}_1 \neq 0$).

The fifth set is similar to the fourth one except that $\chi \neq 0$, i.e., there is an asymmetry between optical fields, unlike the other sets, for which $\chi = 0$. This set illustrates that the asymmetry between optical field frequencies causes the average energy of the system to differ from zero, cf. top panels in Figs. 2, 5 and 8.

The comparison of the second (GKSL-dominated) and third (NH-dominated) sets reveals that the environment effect (17) causes the amplitude damping of dissipation-free system's oscillations, while the environment (16) only distorts them – changes their shape, frequency or amplitude. This is because the chosen model (16) is not entirely suitable for describing evolutions with damping. The latter are better described by a non-normalized density matrix and decay operator given by, for example, $\hat{\Gamma} \propto \left(\begin{smallmatrix} 1 & 0 \\ 0 & 1 \end{smallmatrix}\right)$; note that decay operators proportional to the unit matrix do not contribute to evolutions with a normalized density matrix or a state vector.

Last but not least, important information was received about the behaviour of the quantum von Neumann entropy. It was shown that for evolution with a non-normalized density operator, the standard definition of von Neumann entropy can no longer be used as a purity indicator – because its zeros become a necessary but not sufficient condition for a state to be pure. Instead we introduced a purity-normalized definition (both definitions coincide when a trace of density operator equals one).

We studied the behaviour of both entropies in our Λ-TLS model with postulated environment effects and values of parameters. For normalized density matrix evolution the von Neumann entropy asymptotically approaches a constant value, see Fig. 3, whereas for non-normalized density matrix evolution the von Neumann entropies can oscillate, asymptotically diverge or approach a constant, see Fig. 6.

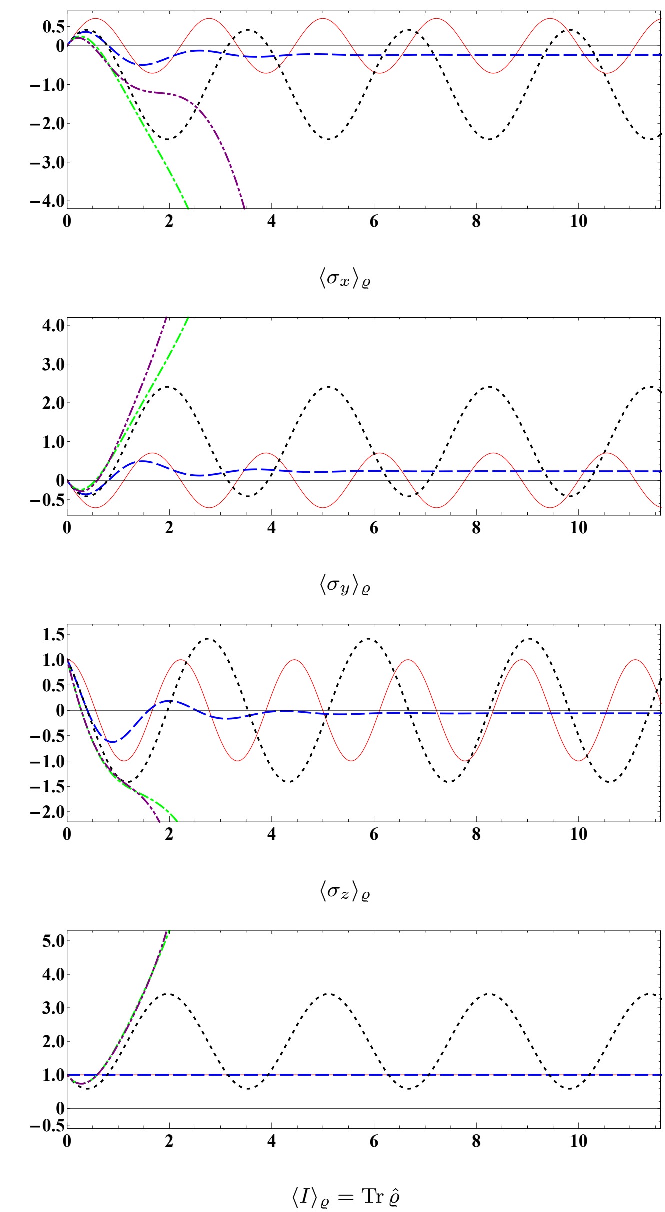

FIG. 4: Spin averages and non-normalized density operator's trace as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1.

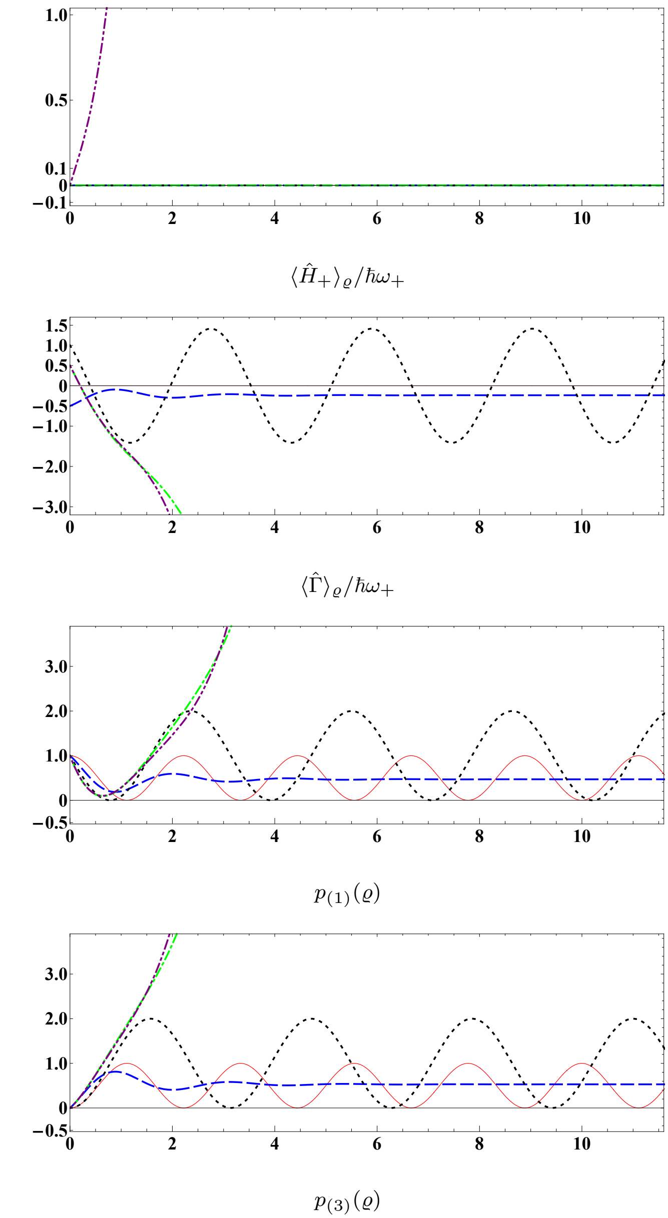

FIG. 5: Mean values of energy, decay operator, and populations of levels 1 and 3, under non-normalized density operator evolution, as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1. Plot of the population difference $p_{(1)}(\varrho) - p_{(3)}(\varrho) = \langle\sigma_z\rangle_\varrho$ can be found in the previous figure.

Acknowledgments

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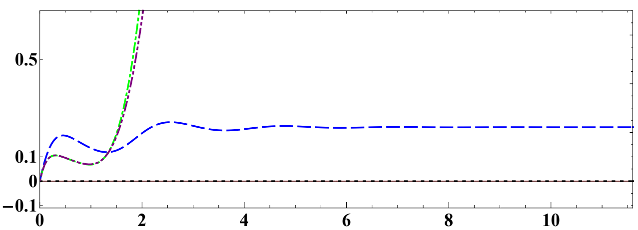
APPENDIX: GENERAL TWO-LEVEL SYSTEMS

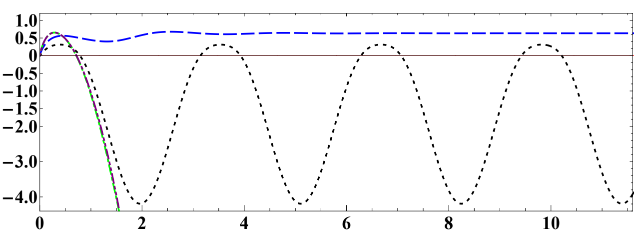
Here we note notations and useful formulae regarding quantum two-level systems described by a general density matrix (whose trace is not necessarily constant or one).

The Hilbert space of any quantum two-level system is formed by two states, the ground state $|g\rangle$ and the excited state $|e\rangle$. An arbitrary quantum state of such system can be written in the orthogonal basis of the Pauli matrices $\hat{\sigma}_x = |e\rangle\langle g| + |g\rangle\langle e| = \left(\begin{smallmatrix} 0 & 1 \\ 1 & 0 \end{smallmatrix}\right)$, $\hat{\sigma}_y = i\left(|e\rangle\langle g| - |g\rangle\langle e|\right) = \left(\begin{smallmatrix} 0 & -i \\ i & 0 \end{smallmatrix}\right)$, $\hat{\sigma}_z = |e\rangle\langle e| - |g\rangle\langle g| = \left(\begin{smallmatrix} 1 & 0 \\ 0 & -1 \end{smallmatrix}\right)$, and the unit matrix $\hat{I} = |e\rangle\langle e| + |g\rangle\langle g| = \left(\begin{smallmatrix} 1 & 0 \\ 0 & 1 \end{smallmatrix}\right)$.

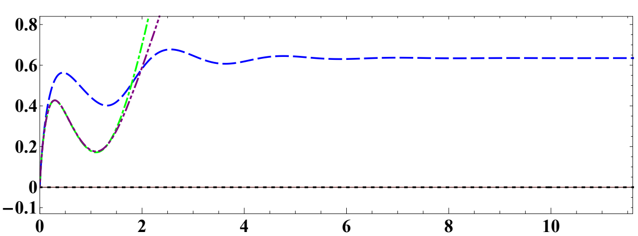
The trace and determinant of each of the Pauli matrices are 0 and -1, respectively, and their eigenvalues are 1 and -1. The linear combinations of the Pauli matrices $\hat{\sigma}_+ = \frac{1}{2}\left(\hat{\sigma}_x + i\hat{\sigma}_y\right) = |e\rangle\langle g| = \left(\begin{smallmatrix} 0 & 1 \\ 0 & 0 \end{smallmatrix}\right)$ and $\hat{\sigma}_- = \hat{\sigma}_+^\dagger = \frac{1}{2}\left(\hat{\sigma}_x - i\hat{\sigma}_y\right) = |g\rangle\langle e| = \left(\begin{smallmatrix} 0 & 0 \\ 1 & 0 \end{smallmatrix}\right)$ are often



$\det\hat{\varrho}$

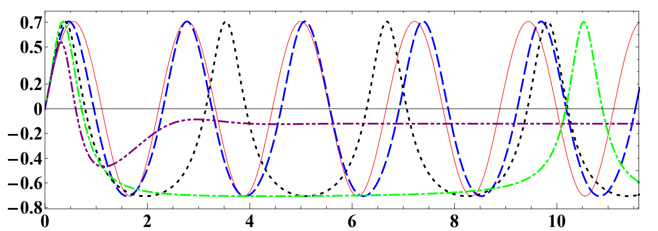


$S_{\mathrm{vN}}(\varrho)$

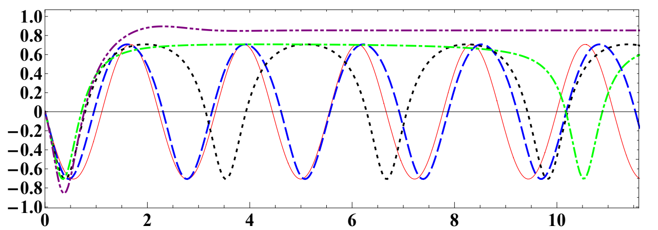


$\widetilde{S}_{\mathrm{vN}}(\varrho)$

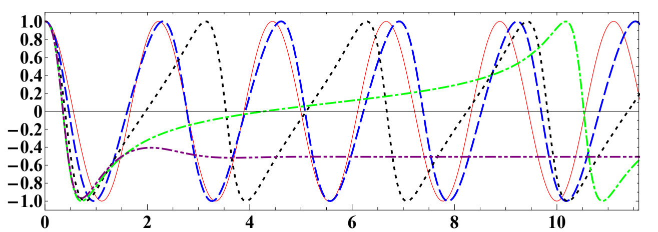
FIG. 6: Purity measures and entropy functions under non-normalized density operator evolution as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1.



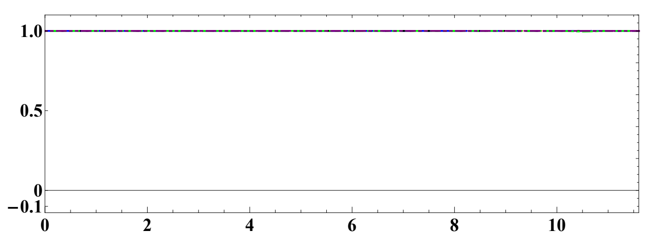
$\langle \sigma_x \rangle_\Psi$



$\langle \sigma_y \rangle_\Psi$



$\langle \sigma_z \rangle_\Psi$



$\langle I \rangle_\Psi = \langle \Psi | \Psi \rangle$

FIG. 7: Spin averages and wavefunction's norm under pure-state evolution as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1.

used too. The matrices obey the properties: $[\hat{\sigma}_i, \hat{\sigma}_j] = 2i \sum_{k=1}^{3} \epsilon_{ijk} \hat{\sigma}_k$, $\{\hat{\sigma}_i, \hat{\sigma}_j\} = 2\delta_{ij}\hat{I}$, $\hat{\sigma}_+^2 = \hat{\sigma}_-^2 = 0$, $\hat{\sigma}_z^2 = 1$, $\hat{\sigma}_\pm \hat{\sigma}_\mp = \frac{1}{2}(\hat{I} \pm \hat{\sigma}_z)$, $\hat{\sigma}_z \hat{\sigma}_\pm = -\hat{\sigma}_\pm \hat{\sigma}_z = \pm \hat{\sigma}_\pm$, where indices 1, 2 and 3 correspond to x, y and z, respectively.

Since the Pauli matrices, together with the unit matrix, form a complete set in two-dimensional Hilbert space, an arbitrary 2×2 matrix $\hat{G}$ can be written in

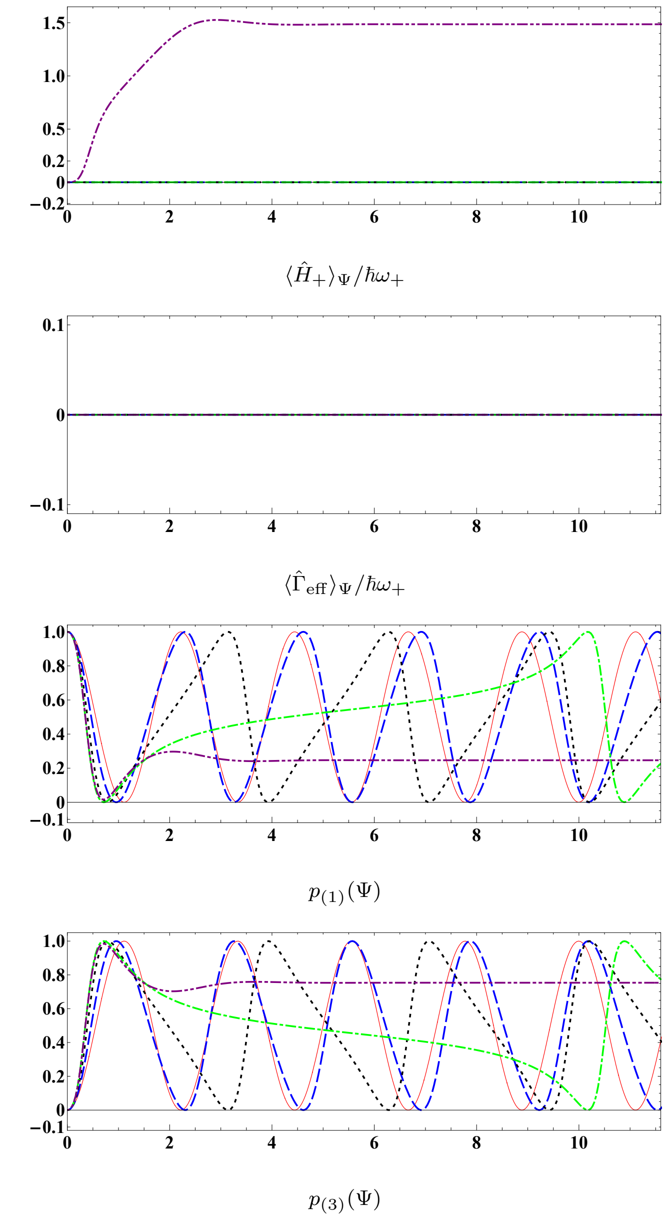


FIG. 8: Mean values of energy, decay operator, and populations of levels 1 and 3, under pure-state evolution, as functions of $\omega_+ t$, at various values of parameters Ω, ϕ, χ, $\tilde{\gamma}_0$, $\tilde{\gamma}_1$. Parameter values and legend are the same as in Fig. 1. Plot of the population difference $p_{(1)}(\Psi) - p_{(3)}(\Psi) = \langle \sigma_z \rangle_\Psi$ is given in the previous figure.

their basis in the following form

$$\begin{aligned} \hat{G} &= \frac{1}{2}\left(\mathrm{Tr}\, \hat{G}\ \hat{I} + \sum_{i=1}^{3} \langle \sigma_i \rangle_G \, \hat{\sigma}_i \right) \\ &= \frac{1}{2}\left(\mathrm{Tr}\, \hat{G}\ \hat{I} + \langle \sigma_z \rangle_G \, \hat{\sigma}_z \right) + \langle \sigma_- \rangle_G \, \hat{\sigma}_+ + \langle \sigma_+ \rangle_G \, \hat{\sigma}_- \\ &= \begin{pmatrix} \frac{1}{2}\left(\mathrm{Tr}\, \hat{G} + \langle \sigma_z \rangle_G \right) & \langle \sigma_- \rangle_G \\ \langle \sigma_+ \rangle_G & \frac{1}{2}\left(\mathrm{Tr}\, \hat{G} - \langle \sigma_z \rangle_G \right) \end{pmatrix}, \end{aligned} \tag{A.1}$$

where $\langle \sigma_i \rangle_G = \mathrm{Tr}(\hat{\sigma}_i \hat{G})$; keeping in mind the index notations above.

Density operator. Let us consider an arbitrary density matrix $\hat{\vartheta}$ whose trace $\mathrm{Tr}\, \hat{\vartheta}$ is not necessarily one, or even constant. Using it, one can define the primary observables as statistical averages: population of the excited and ground states, respectively:

$$p_e(\vartheta) = \vartheta_{ee} = \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta} - \langle \sigma_z \rangle_\vartheta \right), \tag{A.2}$$

$$p_g(\vartheta) = \vartheta_{gg} = \mathrm{Tr}\, \hat{\vartheta} - p_e = \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta} + \langle \sigma_z \rangle_\vartheta \right), \tag{A.3}$$

and coherence

$$\langle \sigma_+ \rangle_\vartheta = \vartheta_{ge} = \mathrm{Tr}\,(\hat{\vartheta} \hat{\sigma}_+), \tag{A.4}$$

where the indices e and g label the corresponding components of the matrix $\hat{\vartheta}$.

In terms of spin averages, a density matrix can be written as

$$\begin{aligned} \hat{\vartheta} &= \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta}\ \hat{I} + \sum_{i=1}^{3} \langle \sigma_i \rangle_\vartheta \, \hat{\sigma}_i \right) \\ &= \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta}\ \hat{I} + \langle \sigma_z \rangle_\vartheta \, \hat{\sigma}_z \right) + \langle \sigma_- \rangle_\vartheta \, \hat{\sigma}_+ + \langle \sigma_+ \rangle_\vartheta \, \hat{\sigma}_- \\ &= \begin{pmatrix} \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta} + \langle \sigma_z \rangle_\vartheta \right) & \langle \sigma_- \rangle_\vartheta \\ \langle \sigma_+ \rangle_\vartheta & \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta} - \langle \sigma_z \rangle_\vartheta \right) \end{pmatrix}, \end{aligned} \tag{A.5}$$

where we used the formula (A.1). Both eigenvalues of this matrix can be easily computed

$$\Lambda_\pm(\vartheta) = \frac{1}{2}\left(\mathrm{Tr}\, \hat{\vartheta} \pm \langle \sigma \rangle_\vartheta \right), \tag{A.6}$$

where $\langle \sigma \rangle_\vartheta \equiv \sqrt{\langle \sigma_x \rangle_\vartheta^2 + \langle \sigma_y \rangle_\vartheta^2 + \langle \sigma_z \rangle_\vartheta^2}$. By using the inequality below, it can be shown that these eigenvalues are non-negative if $\mathrm{Tr}\, \hat{\vartheta}$ is itself non-negative. This indicates that the matrix $\hat{\vartheta}$ is positive-semidefinite, as required by its statistical interpretation.

It is also useful to compute the eigenvectors of the matrix $\hat{\vartheta}$ and present it in the form

$$\hat{\vartheta} = \hat{Q}(\vartheta)\, \hat{\Lambda}(\vartheta)\, \hat{Q}(\vartheta)^{-1}, \tag{A.7}$$

with

$$\hat{\Lambda}(\vartheta) = \begin{pmatrix} \Lambda_-(\vartheta) & 0 \\ 0 & \Lambda_+(\vartheta) \end{pmatrix} = \frac{1}{2}\begin{pmatrix} \mathrm{Tr}\,\hat{\vartheta} - \langle\sigma\rangle_\vartheta & 0 \\ 0 & \mathrm{Tr}\,\hat{\vartheta} + \langle\sigma\rangle_\vartheta \end{pmatrix},$$

$$\hat{Q}(\vartheta) = \begin{pmatrix} \frac{2\Lambda_-(\vartheta)+\langle\sigma_z\rangle_\vartheta - \mathrm{Tr}\,\hat{\vartheta}}{\langle\sigma_x\rangle_\vartheta + i\langle\sigma_y\rangle_\vartheta} & \frac{2\Lambda_+(\vartheta)+\langle\sigma_z\rangle_\vartheta - \mathrm{Tr}\,\hat{\vartheta}}{\langle\sigma_x\rangle_\vartheta + i\langle\sigma_y\rangle_\vartheta} \\ 1 & 1 \end{pmatrix} = \begin{pmatrix} \frac{\langle\sigma_z\rangle_\vartheta - \langle\sigma\rangle_\vartheta}{\langle\sigma_x\rangle_\vartheta + i\langle\sigma_y\rangle_\vartheta} & \frac{\langle\sigma_z\rangle_\vartheta + \langle\sigma\rangle_\vartheta}{\langle\sigma_x\rangle_\vartheta + i\langle\sigma_y\rangle_\vartheta} \\ 1 & 1 \end{pmatrix},$$

where $\hat{Q}(\hat{\vartheta})$ is the matrix composed of the $\hat{\vartheta}$'s eigenvectors arranged in columns.

One can check that during the evolution the spin averages obey the following identity

$$\langle\sigma\rangle_\vartheta^2 \equiv \langle\sigma_x\rangle_\vartheta^2 + \langle\sigma_y\rangle_\vartheta^2 + \langle\sigma_z\rangle_\vartheta^2 = (\mathrm{Tr}\,\hat{\vartheta})^2 - 4\Lambda_+(\vartheta)\,\Lambda_-(\vartheta) = (\mathrm{Tr}\,\hat{\vartheta})^2 - 4\det\hat{\vartheta}, \tag{A.8}$$

from which one can deduce the inequality

$$\langle\sigma\rangle_\vartheta^2 \leqslant (\mathrm{Tr}\,\hat{\vartheta})^2, \tag{A.9}$$

which can be used above to demonstrate the positive-semidefinitess of the matrix $\hat{\vartheta}$.

Pure states. Inequality (A.9) is saturated for the states for which $\det\hat{\vartheta}$ vanishes:

$$\det\hat{\vartheta}_p = 0, \tag{A.10}$$

which are the pure states, i.e., the idempotent states described by a ray in Hilbert space. In other words, their density matrix is a projector and it can be written as a direct product of a two-vector and its Hermitian conjugate:

$$\hat{\vartheta}_p = \hat{\vartheta}_p^2, \quad \hat{\vartheta}_p \mapsto |\Theta\rangle\langle\Theta|, \tag{A.11}$$

where Θ is a corresponding wavefunction. Therefore, spin averages for a pure state lie on the Bloch sphere

$$\langle\sigma\rangle_\Theta^2 \equiv \langle\sigma_x\rangle_\Theta^2 + \langle\sigma_y\rangle_\Theta^2 + \langle\sigma_z\rangle_\Theta^2 = (\mathrm{Tr}\,\hat{\vartheta}_p)^2 = \langle\Theta|\Theta\rangle^2, \tag{A.12}$$

where $\langle\Theta|\Theta\rangle$ is the inner product of the two-vector $|\Theta\rangle$ and its Hermitian conjugate.

Furthermore, a general self-adjoint 2×2 matrix, which satisfies the identity (A.11) and is neither the null nor the unit matrix (those two are not suitable as density matrices because of the corresponding spin averages turning zeros), can be written in a two-parametric form:

$$\hat{\vartheta}_p(c_1,c_2) = \frac{1}{2}\left(\hat{I} + \sqrt{1-4(c_1^2+c_2^2)}\,\hat{\sigma}_z\right) + c_1\,\hat{\sigma}_x + c_2\,\hat{\sigma}_y = \begin{pmatrix} \frac{1}{2}\left[1+\sqrt{1-4(c_1^2+c_2^2)}\right] & c_1 - i\,c_2 \\ c_1 + i\,c_2 & \frac{1}{2}\left[1-\sqrt{1-4(c_1^2+c_2^2)}\right] \end{pmatrix}, \tag{A.13}$$

where parameters c_1 and c_2 are arbitrary real numbers representing, respectively, the real and imaginary parts of the coherence. The trace and determinant of the matrix (A.13) are:

$$\mathrm{Tr}\,\hat{\vartheta}_p = \langle\Theta|\Theta\rangle = 1, \ \det\hat{\vartheta}_p = 0, \tag{A.14}$$

and the corresponding spin averages are:

$$\langle\sigma_x\rangle_p = 2c_1, \ \langle\sigma_y\rangle_p = 2c_2, \ \langle\sigma_z\rangle_p = \sqrt{1-4(c_1^2+c_2^2)}, \tag{A.15}$$

where $\langle\sigma_i\rangle_p \equiv \mathrm{Tr}(\hat{\vartheta}_p\sigma_i) = \langle\Theta|\sigma_i|\Theta\rangle = \langle\sigma_i\rangle_\Theta$.

Finally, let us note the representation of an arbitrary TLS wave function, $|\Theta\rangle \mapsto \begin{pmatrix}\Theta_g \\ \Theta_e\end{pmatrix}$, in terms of spin averages:

$$\begin{aligned} |\Theta_g|^2 &= \frac{1}{2}(\langle\sigma\rangle_\Theta + \langle\sigma_z\rangle_\Theta) = \frac{1}{2}(1+\langle\sigma_z\rangle_\Theta), \\ |\Theta_e|^2 &= \frac{1}{2}(\langle\sigma\rangle_\Theta - \langle\sigma_z\rangle_\Theta) = \frac{1}{2}(1-\langle\sigma_z\rangle_\Theta), \\ \frac{\Theta_e}{\Theta_g} &= \frac{\langle\sigma_x\rangle_\Theta + i\langle\sigma_y\rangle_\Theta}{\langle\sigma\rangle_\Theta + \langle\sigma_z\rangle_\Theta} = \frac{\langle\sigma_x\rangle_\Theta + i\langle\sigma_y\rangle_\Theta}{1+\langle\sigma_z\rangle_\Theta}, \end{aligned} \tag{A.16}$$

where we used formula (A.12). These relations are useful when rewriting any TLS Schrödinger equations in terms of spin averages.

Entropy. In quantum statistical mechanics, the von Neumann entropy is used most often because it is an obvious extension of the concept of the Gibbs entropy from classical statistical mechanics. For an arbitrary density matrix ϑ, this entropy is defined as

$$S_{\rm vN}(\vartheta) \equiv -\langle \ln \hat{\vartheta} \rangle_\vartheta = -{\rm Tr}(\hat{\vartheta} \ln \hat{\vartheta}), \tag{A.17}$$

assuming the units $k_B = 1$, where k_B being the Boltzmann constant. In the TLS case, this definition reads

$$\begin{aligned} S_{\rm vN}(\vartheta) &= -\Lambda_+(\vartheta) \ln [\Lambda_+(\vartheta)] - \Lambda_-(\vartheta) \ln [\Lambda_-(\vartheta)] \\ &= -\frac{1}{2}\Big[\Big({\rm Tr}\,\hat{\vartheta} + \langle\sigma\rangle_\vartheta\Big) \ln\Big({\rm Tr}\,\hat{\vartheta} + \langle\sigma\rangle_\vartheta\Big) + \Big({\rm Tr}\,\hat{\vartheta} - \langle\sigma\rangle_\vartheta\Big) \ln\Big({\rm Tr}\,\hat{\vartheta} - \langle\sigma\rangle_\vartheta\Big)\Big] + \ln 2 \;{\rm Tr}\,\hat{\vartheta}, \end{aligned} \tag{A.18}$$

if one uses the decomposition (A.7), identity $\ln \hat{\vartheta} = \hat{Q}(\vartheta) \ln [\hat{\Lambda}(\vartheta)]\, \hat{Q}(\vartheta)^{-1}$ and trace's properties.

In "conventional" statistical mechanics (where a density operator has a unit trace at all times), von Neumann entropy is often regarded as a measure of purity, because it is zero for a pure state and maximal for a maximally mixed state.

What happens if the trace of a density operator is not conserved to be one? From Eqs. (A.6), (A.8) and (A.14) we obtain:

$$\Lambda_+(\vartheta_p) = {\rm Tr}\,\hat{\vartheta}_p = 1, \;\; \Lambda_-(\vartheta_p) = 0, \tag{A.19}$$

therefore, Eq. (A.18) yields

$$S_{\rm vN}(\vartheta_p) = -{\rm Tr}\,\hat{\vartheta}_p \;\ln\left({\rm Tr}\,\hat{\vartheta}_p\right) = 0. \tag{A.20}$$

However, is the opposite true, i.e., does the vanishing $S_{\rm vN}$ mean that the corresponding state is pure?

Let us consider the state $\hat{\vartheta}_0$ for which the von Neumann entropy vanishes. According to Eq. (A.18), we obtain the equivalence

$$S_{\rm vN}(\vartheta_0) = 0 \;\Leftrightarrow\; \Lambda_+(\vartheta_0)^{\Lambda_+(\vartheta_0)} \Lambda_-(\vartheta_0)^{\Lambda_-(\vartheta_0)} = 1, \tag{A.21}$$

which can be regarded as an algebraic equation for $\hat{\vartheta}_0$. This equation has one obvious solution $\hat{\vartheta}_0 = \hat{\vartheta}_p$, due to Eq. (A.19), thus confirming the property (A.20), but it also has other solutions if ${\rm Tr}\,\hat{\vartheta}_0 \neq 1$. Therefore, vanishing $S_{\rm vN}$ is a necessary but not sufficient condition for a state to be pure.

In this situation, it is convenient to introduce purity-normalized von Neumann entropy

$$\begin{aligned} \widetilde{S}_{\rm vN}(\vartheta) &\equiv -\langle \ln(\hat{\vartheta}/{\rm Tr}\hat{\vartheta}) \rangle_\vartheta = -{\rm Tr}\Big[\hat{\vartheta} \,\ln(\hat{\vartheta}/{\rm Tr}\hat{\vartheta})\Big] \\ &= S_{\rm vN}(\vartheta) + {\rm Tr}\hat{\vartheta}\;\ln {\rm Tr}\hat{\vartheta}, \end{aligned} \tag{A.22}$$

which is similar to the entropy functions discussed in Refs. [41, 42]. It is straightforward to show that the condition of this entropy to take a zero value is equivalent to the algebraic equation:

$$\widetilde{S}_{\rm vN}(\vartheta_0) = 0 \;\Leftrightarrow\; K^K - (K+1)^{K+1} = 0, \tag{A.23}$$

where $K = \Lambda_-(\vartheta_0)/\Lambda_+(\vartheta_0)$ is a ratio of $\hat{\vartheta}_0$'s eigenvalues.

The ratio K must always be finite and non-negative, according to Eqs. (A.6) and (A.9). On the other hand, the equation for K has one, and only one, root in the field of non-negative numbers: $K = 0$, or $\hat{\vartheta}_0 = \hat{\vartheta}_p$. Therefore, vanishing $\widetilde{S}_{\rm vN}$ is both a necessary and sufficient condition for a state to be pure.

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