



Permutational symmetry for identical multilevel systems: A second-quantized approach

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We develop a framework that provides a straightforward approach to fully exploit the permutational symmetry of identical multilevel systems. By taking into account the permutational symmetry, we outline a simple scheme that allows us to map the dynamics of N identical d -level systems to the dynamics of d bosonic modes with N particles, achieving an exponential reduction on the dimensionality of the problem in a simple and straightforward way. In particular, we consider the Lindblad dynamics of several identical multilevel systems interacting with a common subsystem under the action of collective dissipation terms.

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I. INTRODUCTION

When dealing with a collection of N d -level systems, a well-known problem is the so-called curse of dimensionality, i.e., the fact that the dimension of the Hilbert space scales exponentially as d^N . However, in many different physical phenomena, such as lasing [1,2], phase transitions [3–5], superradiance [6,7], strong coupling with organic molecules [8,9] and microwave photonics [10], the theoretical modeling usually assumes that the emitters are identical. In these situations, permutational symmetry of the N d -level systems can be used to greatly reduce the complexity of the problem. This was addressed in the works of Gegg *et al.* [11–13], Shammah *et al.* [14], and Kirton and Keeling [15,16]. In these works, the dynamics of an open quantum system composed of several identical emitters interacting with a common subsystem under the action of individual, but identical, collapse operators is considered. By exploiting the permutational symmetry of the density matrix in the symmetrized Liouville space, a huge reduction in the complexity of the problem is achieved, allowing calculations for larger numbers of emitters than possible otherwise. These efforts were conducted for an ensemble of multilevel systems [11–13] and specialized for the case of two-level systems [14–16]. In the absence of individual dephasing operators and for appropriate initial states, one can further restrict the Hilbert space to the totally symmetric subspace [11,13]. In the case of two-level systems, the construction of the totally symmetric subspace can be achieved by using the Dicke basis, restricting the Hilbert space to the highest superspin subspace [6,7].

In this paper, we notice that working in the totally symmetric subspace is completely equivalent to restricting the possible states to bosonic many-body states. Therefore, by applying the rules of second quantization for bosons, we achieve

the reduction to the totally symmetric subspace in a simple and straightforward way, mapping the dynamics of N identical d -level systems to the dynamics of d bosonic modes with N particles.

II. THEORY

We start by considering the dynamics of N identical d -level systems interacting with a common subsystem and under the action of collective dissipation terms, described by the Lindblad master equation,

$$\dot{\rho} = -i[H, \rho] + \sum_i \mathcal{L}_{C_i}[\rho], \quad (1)$$

where H is the Hamiltonian, possibly time dependent, and $\mathcal{L}_{C_i}[\rho] = C_i \rho C_i^\dagger - \frac{1}{2}(C_i^\dagger C_i \rho + \rho C_i^\dagger C_i)$ is the Lindblad dissipator for the collapse operator C_i . In the following, we assume the Hamiltonian, H , to be invariant under any permutation of the d -level systems. We also restrict the collapse operators, C_i , to collective operators that are also invariant under any permutation of the d -level systems. For the case where the collapse operators may act locally on each d -level system, we must construct and work on the symmetrized Liouville space and this was taken into account in Refs. [11–16]. In this paper, we restrict to the case where both the Hamiltonian and the collapse operators are invariant under any permutation of the emitters.

We may define the symmetrization operator

$$S = \frac{1}{N!} \sum_{\pi} P_{\pi} \quad (2)$$

where P_{π} is a permutation operator and π runs over all possible permutations of the d -level systems. A permutationally invariant operator, O , then satisfies $[S, O] = 0$. We also assume that the initial state, $\rho_0 = \sum_i p_i |\psi_i\rangle\langle\psi_i|$, is a totally symmetric state, i.e., $S|\psi_i\rangle = |\psi_i\rangle$ for all $|\psi_i\rangle$. Since both the Hamiltonian, H , and the collapse operators, C_i , are permutationally invariant operators, when solving the Lindblad

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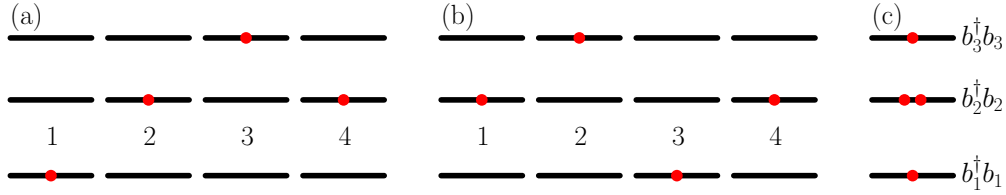


FIG. 1. (a), (b) Schematic representation of two product states of four three-level systems: (a) $|1\rangle_1|2\rangle_2|3\rangle_3|2\rangle_4$ and (b) $|2\rangle_1|3\rangle_2|1\rangle_3|2\rangle_4$. If we restrict dynamics to the totally symmetric subspace, these two states will be always in a symmetric superposition that can be correctly taken into account by using many-body bosonic states: (c) $\frac{1}{\sqrt{2!}}b_3^\dagger b_2^\dagger b_1^\dagger|\text{vac}\rangle$.

dynamics, the density matrix will always remain in the totally symmetric subspace. This may be used to substantially reduce the dimensionality of the problem. As already noticed in Refs. [11,13], this reduction of dimensionality is even larger than the one obtained by working in the symmetrized Liouville space. We must stress that the difference in the restrictions imposed in this paper and the symmetrized Liouville space approach is that here each collapse operator C_i must be permutationally invariant, whereas in Refs. [11–16] only the sum of all collapse operators, $\sum_i C_i$, must be permutationally invariant.

We can write any permutationally invariant M -body operator acting solely on the emitters as

$$O_{\text{em}}^M = \frac{1}{M!} \sum_{i_1 \dots i_M}' \sum_{\substack{\alpha_1 \dots \alpha_M \\ \beta_1 \dots \beta_M}}^d V_{\alpha_1 \dots \alpha_M}^{\beta_1 \dots \beta_M} \sigma_{\beta_1, \alpha_1}^{i_1} \dots \sigma_{\beta_M, \alpha_M}^{i_M} \quad (3)$$

where $\sigma_{\beta, \alpha}^j = |\beta\rangle_j \langle \alpha|_j$ is an operator acting on emitter j , and the primed sum indicates that all indices i_1, \dots, i_M have to be distinct. Using this definition, any permutationally invariant operator acting on the emitters and containing up to M -body terms can be written as

$$O_{\text{em}} = \sum_{J=1}^M O_{\text{em}}^J. \quad (4)$$

In general, a permutationally invariant operator that may act on the emitters and on a common subsystem can be written as the sum of three terms:

$$O = O_{\text{em}} + O_{\text{em-sub}} + O_{\text{sub}}, \quad (5)$$

where O_{em} (O_{sub}) is an operator acting only on the emitters (subsystem). The interaction term, $O_{\text{em-sub}}$, can be written as $O_{\text{em-sub}} = \sum_q A_q B_q$, where A_q acts solely on the emitters and must have the form of Eq. (4) and B_q is an operator acting on the common subsystem.

At this point, one may realize that if the state of the system is restricted to the totally symmetric Hilbert space one can take advantage of all the formalisms of second quantization for bosons, for which the many-body states are automatically restricted to the totally symmetric Hilbert space. This can be done by applying the rules of second quantization [17] and mapping all the operators to a second-quantized form. For instance, Eq. (3) becomes

$$O_{\text{em}}^M = \frac{1}{M!} \sum_{\substack{\alpha_1 \dots \alpha_M \\ \beta_1 \dots \beta_M}}^d V_{\alpha_1 \dots \alpha_M}^{\beta_1 \dots \beta_M} b_{\beta_1}^\dagger \dots b_{\beta_M}^\dagger b_{\alpha_1} \dots b_{\alpha_M} \quad (6)$$

where b_α^\dagger and b_α are the bosonic creation and annihilation operators of an emitter in state α . The recipe then simply consists in applying these rules to all the relevant operators, i.e., the Hamiltonian, the collapse operators, and all the desired observables, giving a Hamiltonian that can be easily implemented with standard quantum optics packages such as QUTIP [18]. For the typical cases where the number of emitters N is fixed, one can restrict the Hilbert space to the N -particle subspace, for which $\langle \sum_{\alpha=1}^d b_\alpha^\dagger b_\alpha \rangle = N$. Similarly, for the initial state, $\rho_0 = \sum_i p_i |\psi_i\rangle \langle \psi_i|$, each $|\psi_i\rangle$ has to be mapped to its second-quantized version, i.e., expressed in the Fock space.

This approach thus solves the dynamics of N permutationally invariant d -level systems by treating it as the dynamics of N bosons in a system with d modes, see Fig. 1 for a schematic picture. It has the usual advantages of a second-quantized formulation. In particular, it is not necessary to explicitly construct a totally symmetric subspace, and the correct symmetry enhancement factors are automatically encoded within and obtained from the bosonic operator algebra.

III. RESULTS

The study of few-level emitters interacting with light is at the core of our understanding of light-matter interaction. In principle, the problem of light-matter interaction can be fully understood within the laws of quantum electrodynamics (QED) [19]. However, for practical applications in fields such as cavity QED, quantum optics, quantum nanophotonics, and quantum plasmonics, a very common assumption is that matter degrees of freedom can be described using only a few levels and that the interaction with light is dominated by a single mode of the electromagnetic field. In the case of two-level systems, this leads to the well-known Rabi [20], Dicke [6], Jaynes-Cummings [21], and Tavis-Cummings [22] models, which differ in the number of emitters and the use of the rotating wave approximation. Nevertheless, even when dealing with an ensemble of few-level systems, the exponential scaling of the Hilbert-space dimension quickly makes the problem intractable and symmetry considerations must be taken into account to reduce the dimension of the problem. In the context of the interaction of an ensemble of identical two-level systems with a cavity mode, superradiance can be observed. To study superradiance, it is useful to rewrite the Hamiltonian using spin operators and use the so-called Dicke basis [7]. In the following, we demonstrate our approach for several examples within this context.

A. Tavis-Cummings model

As a first example to illustrate our approach, we will apply it to the Tavis-Cummings model [22]. For this relatively simple example, we show explicitly that the state space and matrix elements within the second quantized picture are the same as in conventional approaches. The Tavis-Cummings Hamiltonian is given by

$$H_{\text{TC}} = \sum_{j=1}^N \frac{\omega_0}{2} (\sigma_{e,e}^j - \sigma_{g,g}^j) + \omega_c a_c^\dagger a_c + g \sum_{j=1}^N (a_c \sigma_{e,g}^j + a_c^\dagger \sigma_{g,e}^j) \quad (7)$$

where g (e) stands for the ground (excited) state, N is the number of two-level systems, ω_0 (ω_c) is the two-level system (cavity) energy, and a_c is the bosonic annihilation operator for the cavity. A standard approach is to rewrite the above Hamiltonian using spin operators and work in the Dicke basis [7], where $S_+ = \sum_{j=1}^N \sigma_{e,g}^j$, $S_- = (S_+)^\dagger$ and $S_z = \sum_{j=1}^N \frac{1}{2} (\sigma_{e,e}^j - \sigma_{g,g}^j)$. The Hamiltonian can then be written as

$$H_{\text{TC}} = \omega_0 S_z + \omega_c a_c^\dagger a_c + g(a_c S_+ + a_c^\dagger S_-), \quad (8)$$

and the emitter states are $|s, m\rangle$, where s and m are the quantum numbers associated to S^2 and S_z . The totally symmetric subspace is then the highest spin subspace, where $s = N/2$.

If we instead apply our approach and second quantize Eq. (7), we obtain

$$H_{\text{TC}} = \frac{\omega_0}{2} (b_e^\dagger b_e - b_g^\dagger b_g) + \omega_c a_c^\dagger a_c + g(a_c b_e^\dagger b_g + a_c^\dagger b_g^\dagger b_e). \quad (9)$$

In order to see that both approaches are completely equivalent in the totally symmetric subspace ($s = N/2$), we examine the matrix elements of S_- . The action of S_- on a Dicke state is

$$S_- |s, m\rangle = \sqrt{s(s+1) - m(m-1)} |s, m-1\rangle. \quad (10)$$

Here, $m = N_{\text{exc}} - N/2$ is directly related to the number of excited emitters, N_{exc} .

For the second-quantized version, S_- maps to $b_g^\dagger b_e$, which acts on the Fock states $|n_g, n_e\rangle$, where $n_g = N - N_{\text{exc}}$ and $n_e = N_{\text{exc}}$, as

$$b_g^\dagger b_e |n_g, n_e\rangle = \sqrt{(n_g+1)n_e} |n_g+1, n_e-1\rangle. \quad (11)$$

The Dicke state $|s = N/2, m = N_{\text{exc}} - N/2\rangle$ is equal to the Fock state $|N - N_{\text{exc}}, N_{\text{exc}}\rangle$, and comparing Eqs. (10) and (11) shows that the matrix elements are indeed equal. The equality can also easily be checked for $S_z \equiv \frac{1}{2} (b_e^\dagger b_e - b_g^\dagger b_g)$. Therefore, within the totally symmetric subspace, it is completely equivalent to work with either of the two Hamiltonians, Eq. (7) or Eq. (9).

B. Holstein-Tavis-Cummings model

The field of molecular polaritonics and polaritonic chemistry [23–28] studies how to manipulate and use the changes in electronic and vibrational structure and dynamics of molecules under strong coupling with confined modes of light. Since molecules are complex systems with significant internal structure due to rovibrational (nuclear) motion,

describing them as two-level systems is often not a good approximation. At the same time, the influence of individual collapse operators acting on each molecule can often be neglected. On the one hand, their individual radiative decay (on scales of nanoseconds) is often much slower than the dynamics of interest. On the other hand, the influence of the vibrational modes that is sometimes included through a pure-dephasing Lindblad term (which has to be replaced by a more careful treatment under strong light-matter coupling to prevent unphysical effects [29]) can be much better described by treating some vibrational modes (or superpositions of them corresponding to so-called reaction coordinates) explicitly, which allows neglecting the other ones at reasonably short timescales [30,31]. These considerations apply especially for organic molecules interacting with a plasmonic nanocavity [32,33], since their ultrafast loss is typically the dominant decay channel in the system. Explicit inclusion of nuclear degrees of freedom also allows us to represent many effects that cannot be understood within a two-level system description [8,34]. A workhorse in this field is the so-called Holstein-Tavis-Cummings model [8], in which the molecule is approximated using the Holstein model, i.e., two displaced harmonic oscillators for the electronic ground and excited states. Therefore, when dealing with molecular polaritonics, it is common to face situations where one needs to solve the dynamics of identical multilevel systems without any individual collapse operator.

The Holstein-Tavis-Cummings Hamiltonian can be written as

$$H_{\text{HTC}} = \omega_c a_c^\dagger a_c + \sum_{i=1}^{N_{\text{mol}}} H_{\text{mol}}^{(i)} + \sum_{i=1}^{N_{\text{mol}}} H_{\text{cav-mol}}^{(i)}, \quad (12)$$

$$H_{\text{mol}}^{(i)} = \omega_e \sigma_i^+ \sigma_i^- + \omega_v c_i^\dagger c_i - \lambda_v \sigma_i^+ \sigma_i^- (c_i^\dagger + c_i) \quad (13)$$

$$H_{\text{cav-mol}}^{(i)} = g(\sigma_i^+ a_c + a_c^\dagger \sigma_i^-), \quad (14)$$

where σ_i^+ (σ_i^-) is the raising (lowering) operator for the electronic state in molecule i with excitation energy ω_e , whereas c_i is the annihilation operator for the vibrational mode in molecule i , with frequency ω_v and exciton-phonon coupling strength λ_v . The cavity is described through the photon annihilation (creation) operators a_c (a_c^\dagger), with photon energy ω_c . In addition to the coherent dynamics described by the Hamiltonian, the cavity mode decays with rate γ_c , described by a standard Lindblad decay operator $C = \sqrt{\gamma_c} a_c$.

The Holstein-Tavis-Cummings Hamiltonian can be rewritten in terms of the eigenstates of the single-molecule Hamiltonian,

$$H_{\text{mol}}^{(i)} = \sum_{s=g,e} \sum_v \omega_{s,v} |s, v\rangle_i \langle s, v|_i, \quad (15)$$

which are labeled as $|g, v\rangle_i$ and $|e, v\rangle_i$ for vibrational sublevel v in the electronic ground and excited state, respectively. Their corresponding energies are $\omega_{g,v} = \omega_v v$ and $\omega_{e,v} = \omega_e + \omega_v v - \lambda_v^2 / \omega_v$. In this basis, the light-matter interaction operator is given by

$$H_{\text{cav-mol}}^{(i)} = g \sum_{v,v'} (a_c F_{vv'} |e, v\rangle_i \langle g, v'|_i + \text{H.c.}), \quad (16)$$

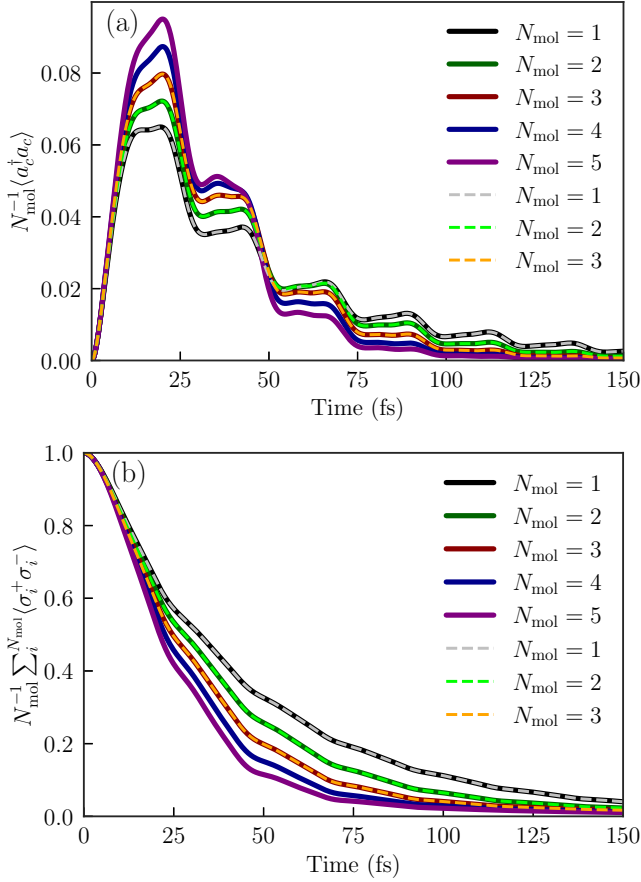


FIG. 2. (a) Normalized cavity population $\langle a_c^\dagger a_c \rangle N_{\text{mol}}^{-1}$ in the Holstein-Tavis-Cummings model, for $N_{\text{mol}} = 1, 2, 3, 4, 5$ calculated both within the second-quantized approach (full lines) and without exploiting permutational symmetry (dashed lines). See main text for parameters. (b) The same for the electronic excited-state population, $\sum_i \langle \sigma_i^+ \sigma_i^- \rangle N_{\text{mol}}^{-1}$.

where $F_{v,v'} = \langle e, v | \sigma_i^+ | g, v' \rangle$ is a vibrational overlap integral or Franck-Condon factor and can be analytically obtained.

Since both the Hamiltonian and collapse operators are permutationally invariant, we can map Eq. (12) to its second-quantized form as long as the initial state is fully symmetric. This gives

$$H_{\text{HTC}} = \omega_c a_c^\dagger a_c + \sum_{s=g,e} \sum_v \omega_{s,v} b_{s,v}^\dagger b_{s,v} + g \sum_{v,v'} (a_c F_{v,v'} b_{e,v}^\dagger b_{g,v'} + \text{H.c.}). \quad (17)$$

In the following, we choose parameter values typical for organic molecules such as anthracene coupled to nanoplasmonic cavities [30], with $\omega_e = 3.5$ eV, $\omega_v = 0.182$ eV, $\lambda_v = 0.096$ eV, $\gamma_c = 0.2$ eV, $g = 0.035$ eV. We set the cavity photon energy to be on resonance with the emission peak of the molecule, $\omega_c = \omega_e - 2\lambda_v^2/\omega_v$. The initial state is chosen to be the fully inverted state, i.e., the state where all molecules are instantaneously excited to the electronic excited state by a vertical Franck-Condon transition.

In Figs. 2(a) and 2(b), we show the numerical results for the dynamics for the cavity and excited-state population,

respectively. In the basis truncation for the single-molecule Hilbert space, we include the six lowest vibrational states for the ground state and the four lowest vibrational states for the electronic excited state, which gives converged results. Within the second-quantized approach, we show results up to $N_{\text{mol}} = 5$, while without resorting to any permutational symmetry we show results up to $N_{\text{mol}} = 3$.

The exciton population in Fig. 2(b) displays a clear enhancement of the spontaneous emission due to Dicke superradiance [6] as the number of emitters is increased. Furthermore, a modulation of the decay rate with a period of about 22 fs can be observed. This modulation is more clearly visible in the cavity population [see Fig. 2(a)] and is a signature of the vibrational motion [30], which has a period of $T_v = 2\pi/\omega_v = 22.7$ fs.

As expected, both approaches are completely equivalent. However, while the Hilbert space for the brute-force approach reaches size $N_{\text{Hilb}} = 4000$ for three molecules, it only has size $N_{\text{Hilb}} = 220$ within the second-quantized approach exploiting the permutational symmetry. For five molecules, this advantage improves to $N_{\text{Hilb}} = 12012$ versus $N_{\text{Hilb}} = 600000$. Here, it should be noted that the size of the density matrix that is propagated in the Lindblad master equation is $N_{\text{Hilb}} \times N_{\text{Hilb}}$, while the Liouvillian superoperator describing this evolution can be formally treated as a $N_{\text{Hilb}}^2 \times N_{\text{Hilb}}^2$ matrix.

C. Three-level systems

To give another numerical example to illustrate this mapping, we formulate a simple model Hamiltonian of N d -level systems, where the levels of each emitter are equally separated in energy by ω_e , coupled to a cavity mode with frequency $\omega_c = \omega_e = 1$ eV. The transition operator of each emitter is defined as $\mu_i = \mu_i^+ + \mu_i^-$, where $\mu_i^+ = \sum_{v=1}^{d-1} \sigma_{v,v+1}^i$ and $\mu_i^- = (\mu_i^+)^{\dagger}$ and each emitter is coupled to the cavity mode by a coupling strength of $g = \frac{0.15}{\sqrt{N}}$ eV in the rotating wave approximation. We also include an all-to-all dipole-dipole interaction term, $H_{d-d} = D \sum_{k,j \neq k} \mu_k \mu_j$, where $D = 0.1$ eV. The Hamiltonian is then given by

$$H = \sum_{i=1}^N \sum_{v=1}^d \omega_v \sigma_{v,v}^i + \omega_c a_c^\dagger a_c + H_{d-d} + g \sum_{i=1}^N (\mu_i^- a_c^\dagger + \text{H.c.}), \quad (18)$$

where $\omega_v = v\omega_e$. The system is under the action of the incoherent decay of the cavity, $C_{\text{cav}} = \sqrt{\gamma_c} a_c$ where $\gamma_c = 0.15$ eV, as well as collective spontaneous emission, $C_v = \sqrt{\Gamma_\downarrow} \sum_{i=1}^N \sigma_{v,v+1}^i$, where v runs from 1 to $d-1$ and $\Gamma_\downarrow = 0.05$ eV. The initial state is chosen to be the fully inverted state, $|\psi_0\rangle = \prod_{j=1}^N |d\rangle_j \otimes |\chi\rangle_{\text{cav}}$, where all emitters are in the most excited state and the cavity is in the vacuum state.

The above Hamiltonian and collapse operators are clearly invariant under any permutation of the emitters. Also, the initial state belongs to the totally symmetric subspace. Therefore, we can again proceed with the mapping by second quantizing all relevant operators. In particular, the Hamiltonian can be

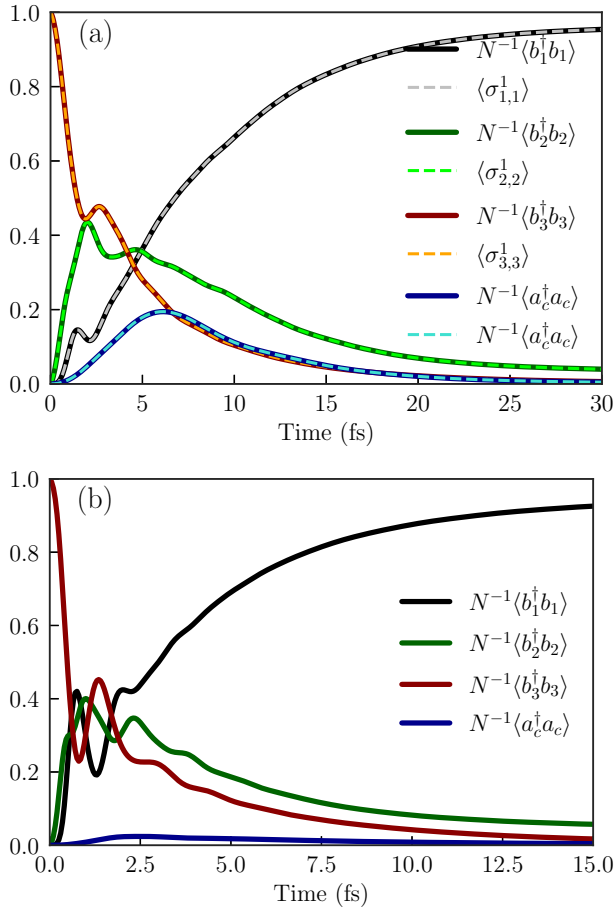


FIG. 3. (a) Dynamics for $N = 5$ three-level emitters calculated within the second-quantized approach (full lines) and without using the permutational symmetry (dashed lines). See main text for parameters. (b) The same for $N = 17$ emitters, which is only possible with reasonable effort when using the second-quantized approach.

written as

$$H = \sum_{v=1}^d \omega_v b_v^\dagger b_v + \omega_c a_c^\dagger a_c + H_{d-d} + g \sum_{i=1}^N \sum_{v=1}^d (b_v^\dagger b_{v+1} a_c^\dagger + \text{H.c.}), \quad (19)$$

where

$$H_{d-d} = D \sum_{v=1}^{d-1} \sum_{\mu=1}^{d-1} (b_v^\dagger b_\mu^\dagger b_{v+1} b_{\mu+1} + b_{v+1}^\dagger b_{\mu+1}^\dagger b_v b_\mu + b_{v+1}^\dagger b_\mu^\dagger b_v b_{\mu+1} + b_v^\dagger b_{\mu+1}^\dagger b_{v+1} b_\mu). \quad (20)$$

Note that this operator is expressed using normal ordering. The $d - 1$ collective spontaneous emission collapse operators can be rewritten as $C_v = \sqrt{\Gamma} \downarrow b_v^\dagger b_{v+1}$. Finally, the initial state is just $|\psi_0\rangle = \frac{1}{\sqrt{N!}} (b_d^\dagger)^N |\text{vac}\rangle_{\text{em}} |\text{vac}\rangle_{\text{cav}}$.

In Figs. 3 and 4, we show the results of the dynamics for the case of three-level systems, i.e., $d = 3$. The total population of the different levels, $\langle \sum_{i=1}^N \sigma_{i,i}^1 \rangle$, can be mapped in the second-quantized approach to $\langle b_i^\dagger b_i \rangle$. Figure 3(a) shows the results for

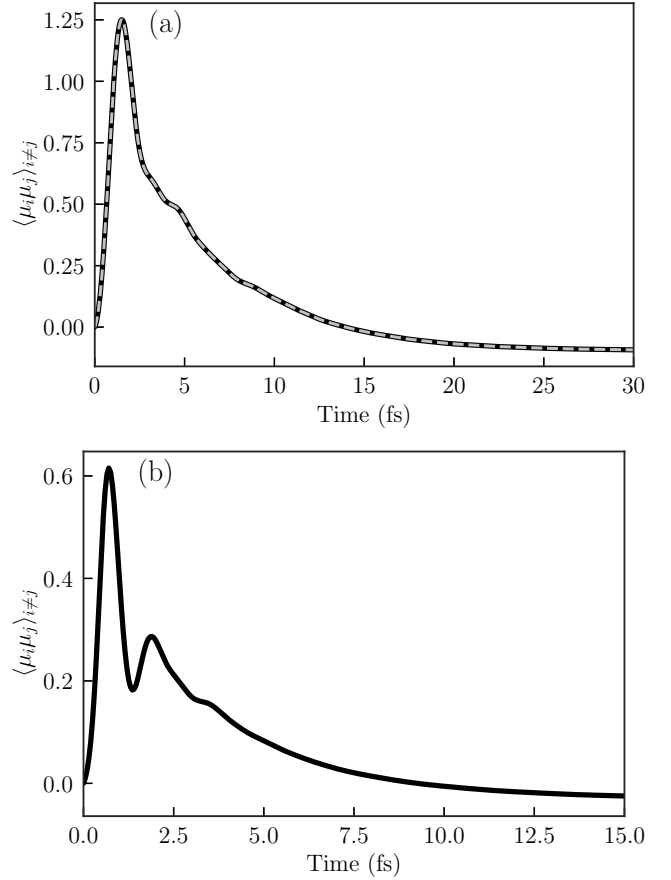


FIG. 4. (a) Expectation value of the dipole-dipole interaction term, $\langle \mu_i \mu_j \rangle$ for $i \neq j$ for the simulation shown in Fig. 3(a) (with $N = 5$), within the second-quantized approach (full lines) and without using the permutational symmetry (dashed lines). (b) The same for the simulation in Fig. 3(b) with $N = 17$ emitters, only using the second-quantized approach.

$N = 5$. In Fig. 3(b), we show the results for $N = 17$ emitters. In Fig. 4, we show the time-dependent expectation value of the dipole-dipole interaction term, $\langle \mu_i \mu_j \rangle$ for $i \neq j$.

Again, as expected, the second-quantized approach is completely equivalent to the direct solution. In this case, the brute-force approach is numerically intractable, as the number of entries in the density matrix is $d^{2N} N_c^2$, where N_c is the dimension of the cavity Hilbert space. When working only with the totally symmetric subspace, the number of entries in the density matrix is reduced to

$$\left(\frac{(N + d - 1)!}{N!(d - 1)!} \right)^2 N_c^2, \quad (21)$$

greatly reducing the size of the dynamical object. When comparing with the approach that uses the symmetrized Liouville space, where the number of entries in the density matrix is $\frac{(N+d^2-1)!}{N!(d^2-1)!} N_c^2$ [11,13], we also get a substantial reduction. As an example, for $d = 3$ and $N = 17$, we have a reduction of the number of entries in the density matrix by a factor of 37.

Since we start in the fully inverted state, the dipole-dipole interaction, H_{d-d} , starts to transfer population from the highest

excited emitter state to the intermediate excited state and to a smaller extent to the emitter ground state. After this first moment, the cavity starts to become populated and, due to its decay, drives the system to its overall ground state (see Fig. 3). It is important to notice that due to the dipole-dipole interaction, H_{d-d} , the ground state of the system is not the state where all emitters are in their bare ground state. Consequently, there is a residual population of the intermediate excited state for long times (see Fig. 3). This is also the reason why the dipole-dipole interaction goes to negative values for long times (see Fig. 4).

D. N -excitation subspace

When working with the dynamics of emitters coupled to cavity modes, there are situations in which we are not interested in working with the full excitation subspace. Indeed, in many common cases, restricting to the first or second excitation subspace is enough [9,34]. Implementing such a restriction within the current approach is rather simple, as one only needs to define an operator that determines the number of excitations in terms of creation and annihilation operators of the emitter levels. As an example, if one is working with the Holstein-Tavis-Cummings model, where each emitter is described as having two electronic states, ground and excited, with one vibrational mode, one could define a subspace where restrictions are imposed on either the electronic or nuclear excitations, or both.

As a concrete example, we discuss vibrational strong coupling for the case where a single (approximately harmonic) vibrational mode per molecule is in resonance with a cavity mode. The simplest Hamiltonian to model collective vibrational strong coupling is [29]

$$H = \omega_c a_c^\dagger a_c + \sum_{i=1}^{N_{\text{mol}}} \omega_v c_i^\dagger c_i + \sum_{i=1}^{N_{\text{mol}}} g(a_c^\dagger c_i + \text{H.c.}), \quad (22)$$

where a_c is the annihilation operator for the cavity mode with frequency ω_c , and c_i is the annihilation operator of the optically active vibrational mode of molecule i , characterized by its frequency ω_v . N_{mol} is the number of molecules, and the cavity-phonon interaction is given by g . Rewriting the vibrational operators using the eigenstates of the harmonic oscillator, $c_i^\dagger = \sum_{n=0}^{\infty} \sqrt{n+1} |n+1\rangle_i \langle n|_i$, Eq. (22) can be written as

$$H = \omega_c a_c^\dagger a_c + \sum_{i=1}^{N_{\text{mol}}} \sum_{n=0}^{\infty} n \omega_v |n\rangle_i \langle n|_i + \sum_{i=1}^{N_{\text{mol}}} \sum_{n=0}^{\infty} g(a_c \sqrt{n+1} |n+1\rangle_i \langle n|_i + \text{H.c.}). \quad (23)$$

This Hamiltonian is permutationally invariant under the exchange of any two molecules. If the initial state is in the totally

symmetric subspace, we can map the Hamiltonian to

$$H = \omega_c a_c^\dagger a_c + \sum_{n=0}^{\infty} n \omega_v b_n^\dagger b_n + \sum_{n=0}^{\infty} g(a_c \sqrt{n+1} b_{n+1}^\dagger b_n + \text{H.c.}), \quad (24)$$

where b_n is the bosonic annihilation operator for the state $|n\rangle$ and the states of interest are restricted to the subspace where $\langle \sum_{n=0}^{\infty} b_n^\dagger b_n \rangle = N_{\text{mol}}$. For regimes in which $\omega_v \approx \omega_c$, it is reasonable to work in the N_{exc} -excitation subspace [35]. In this formalism, this additional restriction can be simply formulated as $\langle \sum_{n=0}^{\infty} n b_n^\dagger b_n + a_c^\dagger a_c \rangle = N_{\text{exc}}$.

IV. CONCLUSION

To conclude, we have proposed a scheme to fully exploit the permutational symmetry of identical but arbitrary emitters when only collective dissipation operators are considered. This scheme relies on the fact that the totally symmetric subspace is equivalent to a bosonic many-body state. After mapping all relevant operators to a second-quantized picture using a simple procedure, the explicit construction of the totally symmetric subspace from direct state products is not required anymore. This approach thus provides a straightforward and easily implemented way to treat such systems while fully exploiting their permutational symmetry to significantly reduce the size of the Hilbert space. We discuss several examples, such as the Tavis-Cummings model, the Holstein-Tavis-Cummings model, and a model Hamiltonian where two-body operators are taken into account, and explicitly demonstrate the equivalence of the second-quantized approach to direct solution.

We expect that this paper will be helpful for simulations that can fully exploit the permutational symmetry of emitters in totally symmetric cases in a very simple way. This can be especially useful for situations where each emitter must be considered as having an internal structure that goes beyond the two-level approximation, such as necessary in the field of molecular polaritonics. For such systems, the current approach can provide a significant reduction of the numerical complexity for very little effort.

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