Quantum Rabi Model for N-State Atoms

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A tractable N-state Rabi Hamiltonian is introduced by extending the parity symmetry of the two-state model. The single-mode case provides a few-parameter description of a novel class of periodic systems, predicting that the ground state of certain four-state atom-cavity systems will undergo parity change at strong-coupling. A group-theoretical treatment provides physical insight into dynamics and a modified rotating wave approximation obtains accurate analytical energies. The dissipative case can be applied to study excitation energy transfer in molecular rings or chains.

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Interactions between spin systems and harmonic oscillators (boson modes) have been studied for over 70 years [1–3]. One of the most well known, the quantum Rabi model [1], is a Hamiltonian describing interactions between a two-level system and a cavity mode. The model has also formed a basis of understanding for exciton-phonon interactions [4] and, along with its multimode extension, has numerous established applications in chemistry and physics (see [5–7] and references therein). The Jaynes-Cummings (J-C) [3] model is obtained by taking the Rabi model in the rotating wave approximation (RWA), where the "counterrotating" terms are ignored (see, e.g., [8]). While the J-C model is sufficient to study small atom-field coupling, the RWA breaks down as the coupling approaches the mode frequency [9,10] and the full Rabi model is needed. Experimental techniques have accessed these strong-coupling regimes [11] and there is much ongoing interest in future experimental realizations in both cavity [12] and circuit [13] QED.

Many-site spin-boson interaction, e.g., multistate atom-cavity interaction [14] or excitation energy transfer in multichromophoric systems [15], continues to be a subject of significant interest, dictating a need for extensions of the two-state model. Extensions of the J-C model have been studied extensively [16–19], but are not applicable in the strong-coupling regime. Exciton-phonon generalizations which extend the parity symmetry of the Rabi model [20] are neither tractable nor applicable to atom-cavity systems. Most importantly, the Rabi model is the single-mode version of a dissipative (infinite-mode) spin-boson model [21], signifying that light-matter interaction is a simplified manifestation of a more fundamental interaction between a two-state system and a dissipative environment. Previous dissipative [22–25] generalizations have neither extended the symmetry nor preserved this correspondence. Motivated by these properties, this Letter presents a symmetry-preserving N-state extension of the Rabi model. The extension includes counterrotating terms in a rigorous, intuitive, and mathematically manageable way, using a minimal number of parameters and paving the way for applications to multilevel atom-cavity experiments at both weak and strong coupling. A group-theoretical approach [2] provides numerical advantages and physical insight into dynamics of the model. The symmetric generalized RWA [7] is applied to obtain accurate analytical energies and eigenstates valid for strong coupling. The above procedures are significantly simplified via the generalized spin matrices [26], providing a useful tool for the treatment of finite quantum systems. The corresponding infinite-mode extension can in turn be applied to periodic dissipative N-state systems.

This work will present the N-state Rabi model's physical motivation in two different representations, discussing the $N = 3$, 4 cases from the viewpoint of atom-cavity physics. A discussion of the conserved quantum numbers, a third symmetry-adapted representation, and dynamical properties of the single-mode case follows. The remaining space is devoted to a brief description of the dissipative case. For reference, the three representations discussed here are graphically outlined in Fig. 1 for $N = 2$, 3, 4.

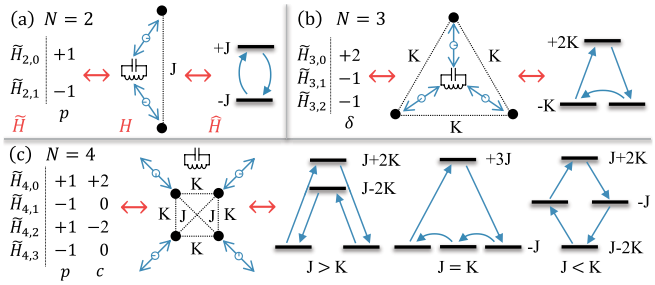

FIG. 1 (color online). (a) The partially diagonalized (left panel), position (center), and energy (right) representations form three equivalent interpretations of the two-state Rabi model. (b) and (c) analogously depict the N-state Rabi model from Eq. (6) with $N = 3$, 4, respectively. The parameters J, K form the energies of the system, $\{p, \delta, c\}$ are each system's conserved quantum numbers, and $\tilde{H}_{N,n}$ are boson chains (defined in text).

Position representation.—Consider the Hamiltonian $H = H_{\rm sys} + H_{\rm field} + H_{\rm int}$ describing an N-state system interacting with electric and magnetic fields ($\mathbf{E}$ and $\mathbf{B}$) of a cavity mode of frequency ω and wave number $\mathcal{K}$. The Hamiltonian will be formulated in the *position* representation, where the interaction $H_{\rm int}$ is on the (spin) diagonal. This differs from the traditional introduction of the Rabi model in the *energy* representation, where the interaction is off-diagonal (p. 194 of [8]). While the two formulations are equivalent for the two-state case, this version provides a natural symmetry-based extension. The system is defined by

$$H_{\rm sys} = \sum_k \sum_{n=0}^{N-1} J_k(|n\rangle\langle n+k| + |n+k\rangle\langle n|), \tag{1}$$

where $\{|n\rangle\}_{n=0}^{N-1}$ form a complete set of position eigenstates and $k \neq 0$ sums over all sites. Transforming H into the energy representation, i.e., diagonalizing $H_{\rm sys}$, obtains an N-state "atom" with energies determined by the parameters J_k. The interaction is

$$H_{\rm int} = -\mathbf{d}_E \cdot \mathbf{E} - \mathbf{d}_B \cdot \mathbf{B}, \tag{2}$$

where $\mathbf{d}_{E(B)}$ is the electric (magnetic) dipole moment operator. The cavity mode can be quantized [8] with $\mathbf{E} \propto (a + a^\dagger)\sin(\mathcal{K}z)$, $\mathbf{B} \propto i(a^\dagger - a)\cos(\mathcal{K}z)$, and $H_{\rm field} = \omega a^\dagger a$ (with $a^\dagger$ and a denoting creation and annihilation operators of the mode). Switching sin↔ cos by introducing $b = ae^{-i\pi/2}$, discretizing the z axis over the N position states of the atom ($\mathcal{K}z|n\rangle = \frac{2\pi n}{N}|n\rangle$), and relegating the coupling strengths to a parameter λ obtains the N-state Rabi Hamiltonian [27]

$$H = \omega b^\dagger b + \lambda \sum_{n=0}^{N-1} (be^{i2\pi n/N} + b^\dagger e^{-i2\pi n/N})|n\rangle\langle n| + \sum_k \sum_{n=0}^{N-1} J_k(|n\rangle\langle n+k| + |n+k\rangle\langle n|). \tag{3}$$

For the two-state case ($N = 2$), this simplifies to the original Rabi model

$$H_2 = \omega b^\dagger b + \lambda(b + b^\dagger)\sigma_z + J\sigma_x, \tag{4}$$

where $J \equiv J_{N/2}$ and $\sigma_{x,z}$ are the usual Pauli matrices. One can also interpret H as a normal mode smeared over a tunneling N-site system (discussed later). As a result, this extension maintains the correspondence between atom-field interaction and a more general spin-boson model.

It will now be shown that reexpressing the model in terms of the generalized spin matrices, the unitary generalization of the Pauli matrices [26], will reduce mathematical complexity while increasing physical understanding. Suppressing dependence on N, generalized spin matrices for $0 \leq j, k < N$ are defined (modulo N) as

$$S_{j,k} = \sum_{n=0}^{N-1} e^{i2\pi nj/N}|n\rangle\langle n+k| = (S_{1,0})^j (S_{0,1})^k. \tag{5}$$

With the details relegated to [28], the reader need only keep in mind the function of the two indices: j determines the phase at each entry n while k determines the entry's location. The matrices $S_{1,0}$ and $S_{1,0}^\dagger$ concisely express $H_{\rm int}$ while $S_{0,k} + S_{0,k}^\dagger$ describes the neighbor couplings of $H_{\rm sys}$. For $0 < k \leq \kappa \equiv \lfloor \frac{1}{2}(N-1) \rfloor$ (with $\lfloor N \rfloor$ the floor function), Eq. (3) is thus reexpressed as

$$H = \omega b^\dagger b + \lambda(bS_{1,0} + b^\dagger S_{1,0}^\dagger) + JS_{0,N/2} + \sum_{k=1}^{\kappa} J_k(S_{0,k} + S_{0,k}^\dagger). \tag{6}$$

Energy representation.—One can now transform H into the energy representation $\hat{H} = V^\dagger H V$ using the unitary transformation

$$V = \frac{1}{\sqrt{N}} \sum_{k=0}^{N-1} e^{i2\pi k^2/N} S_{k,k} S_{1,0}, \tag{7}$$

linking the above formulation with the well-established picture of dipole transitions in N-state systems [8]. The transformed Hamiltonian

$$\hat{H} = \omega b^\dagger b + \lambda(bS_{2,1}e^{i4\pi/N} + b^\dagger S_{2,1}^\dagger e^{-i4\pi/N}) + \hat{H}_{\rm sys} \tag{8}$$

models an N-state atom coupled to a field mode (with the option for more modes [29]). The symmetry of the coupling determines which states are coupled by the mode and the state energies are determined by $\hat{H}_{\rm sys}$. For $N = 2$, Eq. (8) reduces to $\hat{H}_2$, the Rabi Hamiltonian in the energy representation [$\sigma_x \leftrightarrow \sigma_z$ in Eq. (4)]. The three- and four-state cases are reviewed below.

$N = 3$: Setting $J_1 \equiv K$ in Eq. (6), the three-state case in the energy representation is

$$\hat{H}_3 = \begin{pmatrix} \omega b^\dagger b + 2K & \lambda b e^{-i2\pi/3} & \lambda b^\dagger \\ \lambda b^\dagger e^{i2\pi/3} & \omega b^\dagger b - K & \lambda b e^{i2\pi/3} \\ \lambda b & \lambda b^\dagger e^{-i2\pi/3} & \omega b^\dagger b - K \end{pmatrix}. \tag{9}$$

The above is a three-level atom with an initially degenerate ground state and energy separation $3K$ coupled to one cavity mode [29]. The states are thus arranged in a Λ configuration (with inversion of K obtaining a V configuration), similar to well-studied Λ-systems [16]. However, dipole transitions occur between all three levels while extending the parity symmetry and maintaining the relative simplicity of the two-state Rabi model. The bottom left entry in Eq. (9) describes the process in which the atom makes a transition from the upper to one of the lower levels and a photon is annihilated [8]. The RWA (with respect to $\omega b^\dagger b + \hat{H}_{\rm sys}$) removes this transition, relating Eq. (9) to

well-established extensions of the J-C model [16]. The coupling between the ground states represents an ac-Stark shift (similar to the J-C model in the dispersive regime [30], relevant to nondemolition measurements), which interestingly remains relevant after the RWA.

$N = 4$: For the four-state case, $\hat{H}_{\rm sys} = \mathrm{diag}\{J + 2K, -J, J - 2K, -J\}$. Depending on the relation between $J > 0$ and K [Fig. 1(c)], one can obtain either a double-Λ, tripod, or $\diamond$ four-state configuration [19,31]. Inversion of J obtains inverted tripod and double-Λ configurations; inversion of K leaves the system invariant just like inversion of J for $N = 2$. The cavity frequency ω can be tuned to the three possible transition frequencies of the atom, producing a four-parameter model for treating single- and (in the $\diamond$ case) multilevel transitions in several related systems. Additionally, H_4 can be separated into two effective two-state systems as $K \to 0$. One striking feature is that the ground state can change for increasing values of λ, a property not seen at $N < 4$. Shown in Fig. 2(b) for a particular $\diamond$-configuration, the original ground state at small coupling (blue [dark gray]) is surpassed by the unperturbed first excited state (green [medium gray]) as the coupling increases.

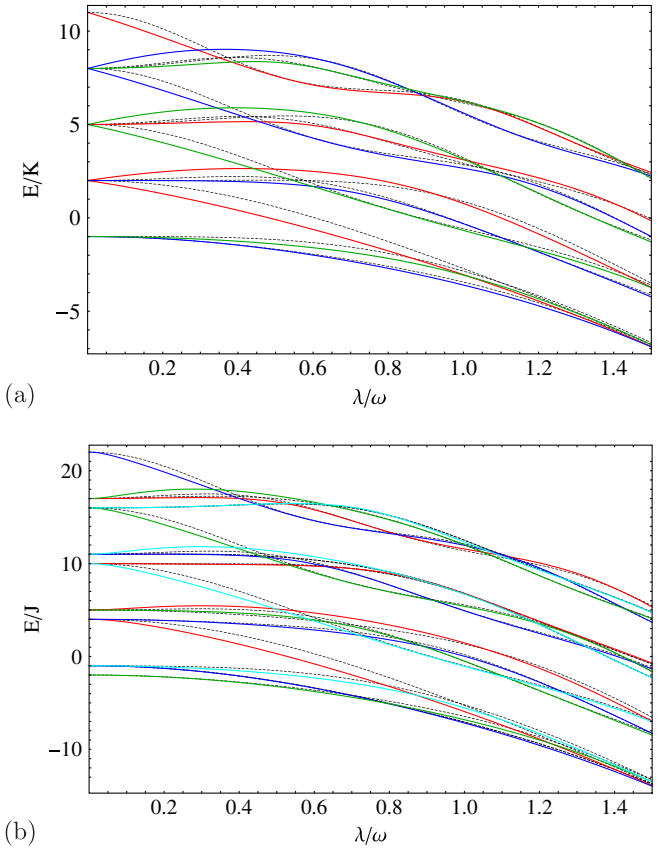

FIG. 2 (color online). Correlation diagrams of energy vs coupling λ for (a) H_3, a resonant Λ-configuration with $K = \omega/3$, and (b) H_4, a $\diamond$-configuration with $K = \omega/4$ and $J = \omega/6$ ($\omega = 1$). The numerical energies belong to chains $\tilde{H}_{N,n}$ (for $n < N = 3, 4$) represented as red (medium-dark gray), blue (dark gray), green (medium gray), and cyan (light gray), in that order, while approximate S-GRWA energies are dashed. The model predicts that the $\diamond$-configuration may have a different ground state at $\lambda/\omega \approx 1$ than at weak coupling.

Conserved quantum numbers.—The generalized spin matrices allow one to easily construct the complete set of conserved quantum numbers for H, providing insight into dynamics [9] and integrability [6]. It can be shown that the Hamiltonian (6) possesses an N-fold rotational symmetry and commutes with the rotations $\{\mathcal{R}_n S_{0,n}\}_{n=0}^{N-1}$, where the operator $\mathcal{R}_n \equiv \exp(i\frac{2\pi}{N} n b^\dagger b)$ and the parity operator $\mathcal{R} \equiv \mathcal{R}_{N/2}$ is present for even N. These can be compiled into the general N-state commuting operator

$$\mathbf{N} = J\mathcal{R}S_{0,N/2} + \sum_{k=1}^{\kappa} J_k(\mathcal{R}_k S_{0,k} + \mathcal{R}_k^\dagger S_{0,k}^\dagger), \qquad (10)$$

consisting of the family of κ commuting Hermitian operators multiplied by site couplings J_k (with the additional parity operator for even N). For the original $N = 2$ case, $\mathbf{N}$ reduces to J multiplied by the well-known spin-boson parity $\sigma_x\mathcal{R}$ [6,9]. This result shows that these N-state atom-cavity systems not only preserve parity for any even N, but are classified by other quantum numbers for $N > 2$. For example, the three-state case contains a conserved quantum number $\delta = 2, -1$ while the four-state system has two: parity $p = \pm 1$ and "cascade" number $c = 0, \pm 2$ (see Fig. 1(c) and [28]).

Analytical insight.—The rotational symmetry of H allows decomposition into N infinite-dimensional subspaces (*boson chains*, denoted as $\tilde{H}_{N,n}$) via a group-theoretic transformation U [28]. In this partially diagonalized representation, the Hamiltonian $\tilde{H} = U^\dagger H U$ is diagonal in the spin subspace with $\langle n'|\tilde{H}|n\rangle = \delta_{n',n}\tilde{H}_{N,n}$. These chains are isomorphic to H and provide significant numerical advantages [7]. For the two-site case, $\tilde{\mathbf{N}} \to J\sigma_z$, resulting in parity chains [9], shown in the left panel of Fig. 1(a). The chains and their respective quantum numbers for the three- and four-state cases are depicted in Figs. 1(b) and 1(c), respectively. The numerical energies for H_3 and H_4 are plotted in Figs. 2(a) and 2(b), respectively, and each chain is labeled by a color. The spectrum of H demonstrates the familiar braidlike crossing pattern of the two-level Rabi Hamiltonian with the addition of more braids.

The group-theoretical approach is also useful for extending analytical approximations, such as the symmetric generalized RWA (S-GRWA [7], applied in [28]). The S-GRWA energies (dashed in Fig. 2) are most accurate in the deep-strong coupling regime ($\lambda \gtrsim \omega$), where the symmetry and chain structure dominate the spectrum [9]. While the S-GRWA also fares well at $\lambda \ll \omega$, the symmetry is not terribly relevant in that region and the original RWA may be applied without loss of accuracy. In the weak coupling regime, it is anticipated that the dynamics will exhibit collapse and revival phenomena reminiscent of the

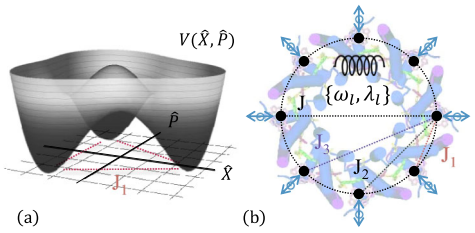

FIG. 3 (color online). In a manner analogous to [21], (a) is a pictorial representation of a symmetric three-well potential in the three-state limit. (b) is a visualization of the B800 ring in LH-II [35], representing a specific application of the N-state spin boson to modeling multichromophoric energy transfer in periodic systems.

original J-C model [8], but this time between multiple atomic states. The notably different behavior of H in the strong-coupling regime will likely be an extension of that described in [9] and will be chain dependent. Both of these regimes (as well as transitions between them) reveal opportunities for interesting realizations of both well-known and newly discovered phenomena of the original two-level case.

Extensions.—Having examined the single-mode case, the dissipative version is now defined. As an extension of Leggett *et al.* [21], consider a continuous N-well system with symmetric potential $V(\hat{X}, \hat{P})$ where the dynamics is restricted to the N-dimensional subspace of the well ground states. One then obtains H_{sys} by introducing tunneling matrix elements J_k between the wells [see Fig. 3(a) for $N = 3$]. With the dissipative environment approximated by a continuum of modes $\{\hat{q}_l, \hat{p}_l\}$, the N-state spin-boson Hamiltonian is simply Eq. (6) with $\{b, \omega, \lambda\} \rightarrow \{b_l, \omega_l, \lambda_l\}$. The interaction term satisfies the criteria of [32] and simplifies to the degenerate two-site spin-boson model at $N = 2$. Other continuum normal modes can be added in a similar fashion [29].

Since the N-state model preserves rotational symmetry, the infinite-mode H is an effective model for the single excitation manifold of a molecular ring or periodic chain interacting with a normal mode of a collective uncorrelated vibrational bath [23]. This model specifically includes the geometrical structure of the system, an important property in excitation energy transfer [33]. Couplings J_k between all sites in the ring are included, allowing one to model systems with interactions other than the nearest neighbor. This version can model photoexcitation dynamics of molecular trimers [24,34] and larger rings. A specific example is the 8-9 member B800 ring of photosynthetic LH-II [35], illustrated in Fig. 3(b). Recently developed methods [15] for spin-boson dynamics can readily be applied to reveal similar insight into many-site systems as previous approaches [21,36] have revealed in the simplest two-site case.

As a final note, instead of extending the number of modes (or even reservoirs [37]), the N-state Rabi model can be extended to many N-state systems. This approach would be similar to previous extensions of the two-state case [38], but would include odd N, potentially revealing phase transitions and other interesting physics.

Summary.—This work introduces an extension of the two-state Rabi model [1] to describe dynamics of a more general N-state periodic system. The symmetry of the system is utilized in a group-theoretical approach, revealing insight into its energies and conserved quantities while also simplifying numerical analysis. A class of matrices [26] provides an efficient method for obtaining the above results. Finally, the proposed infinite-mode extension generalizes the two-site spin-boson model [21] to dissipative periodic N-site systems.

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[28] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.108.180401 for details on the generalized spin matrices, the Fulton-Gouterman [2] transformation that partially diagonalizes H, and the S-GRWA.

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