Classification of Dark States in Multi-level Dissipative Systems

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Dark states are eigenstates or steady-states of a system that are decoupled from the radiation. Their use, along with associated techniques such as Stimulated Raman Adiabatic Passage, has extended from atomic physics where it is an essential cooling mechanism, to more recent versions in condensed phase where it can increase the coherence times of qubits. These states are often discussed in the context of unitary evolution and found with elegant methods exploiting symmetries, or via the Bruce-Shore transformation. However, the link with dissipative systems is not always transparent, and distinctions between classes of CPT are not always clear. We present a detailed overview of the arguments to find stationary dark states in dissipative systems, and examine their dependence on the Hamiltonian parameters, their multiplicity and purity. We find a class of dark states that depends not only on the detunings of the lasers but also on their relative intensities. We illustrate the criteria with the more complex physical system of the hyperfine transitions of $^{87}$Rb and show how a knowledge of the dark state manifold can inform the preparation of pure states.

I. INTRODUCTION

Coherent population trapping (CPT) and transfer in few-level systems consists of the preparation of pure states or the coherent transfer among them by the use of control fields and auxiliary excited levels [1,3]. This arrangement is immune to certain damping processes. The main mechanisms, CPT and Stimulated Raman Adiabatic Passage (STIRAP) have been most studied in the $\Lambda$ three-level system consisting of two-ground states and one excited state where the ground-excited transitions can be independently addressed. There are two viewpoints of CPT depending on whether one includes dissipation or not. In a Hamiltonian system with unitary evolution, asymmetric linear combinations of the ground states decouple from a radiation field provided some conditions are met for the field frequencies. In a dissipative system, this asymmetric combination becomes the stationary state regardless of initial conditions. Originating in atomic and molecular physics [4,5], it can be used for atomic cooling, metrology [6,8], testing QED, and has been extended to state initialization in quantum information [9,10] including qubit gates, and solid-state systems such as spin systems in nitrogen vacancies [11], superconducting circuits [12], semiconductor heterostructures [13,16] where coherence lifetimes have been successfully extended by orders of magnitude [17,18]. Some recent proposals suggest its use in plasmonic systems [19]. Multiple level systems are also quite common in atomic and solid-state devices [3,10,17,18,20,22]. In order to find the solutions to dark states various approaches have been followed. An analysis based on symmetries and constants of the motion has permitted the generalization to multilevel systems as long as the system preserves some symmetries notably with respect to the decouplings [23,24]. The Morris-Shore transformation separates a set of $N_g$ degenerate ground states and $N_e$ degenerate excited states into pairs of bright states, dark states and spectator states [25,27], recently extended to the case of small detunings [28]. The transformation relies on having a number of ground states in excess in order to result in dark states. There are other instances, however, where certain states might decouple from the radiation that fall outside of these considered cases (for example for equal number of ground and excited states).

In this work, we give an overview for the conditions to find dark states in dissipative systems in Lindblad form, and systematically study the multiplicity of dark stationary states, their purity and their dependence on laser field detunings and Rabi frequencies.

II. GENERAL CONSIDERATIONS

A. System and definitions

We consider an $N$-level systems which can be divided into $N_g$ ground states $|g_i\rangle$ with energy $E_{g_i}^g$ $(i = 1, 2, \cdots, N_g)$, and $N_e = N - N_g$ excited states $|e_j\rangle$ with energy $E_{e_j}^e$ $(j = 1, 2, \cdots, N_g)$, that decay to the ground states via spontaneous emission or coupling to a bath, at rate $\gamma_{ij}$, (see Figure 1). The Hamiltonian of the system

\begin{equation}
\hat{H} = \sum_{i=1}^{N} \omega_{g_i} |g_i\rangle \langle g_i| + \sum_{j=1}^{N_g} \omega_{e_j} |e_j\rangle \langle e_j| - \sum_{i,j=1}^{N_g} \gamma_{ij} |g_i\rangle \langle e_j|.
\end{equation}

Here $\omega_{g_i}$ and $\omega_{e_j}$ are the transition frequencies between ground and excited states, respectively.
Dissipation can be written as:

\[ \text{ground manifold} \rightarrow \text{excited manifold} \]

\[ \text{Dissipation} \]

\[ \text{Radiative coupling} \]

The excited \( \sigma \) of the operators can now be time independent and can be written in terms of the energy and frequency are equivalent. The Hamiltonian \( \text{H} \) is now time independent and can be written in terms of the operators \( \sigma^\dagger_{ij} = |g_i\rangle\langle g_i| - |e_j\rangle\langle e_j| \) (see appendix A) as:

\[ H = \sum_{i=1}^{N_g} \sum_{j=1}^{N_e} \left[ \Delta_{ij} \sigma^\dagger_{ij} + (V_{ij}|g_i\rangle\langle e_j| + \text{H.C.}) \right]. \]

where \( V_{ij} = F_{ij} e^{i\phi_{ij}} \) are the complex Rabi frequencies and

\[ \Delta_{ij} = E_j^{(e)} - E_i^{(g)} - \omega_{ij} \]

is the detuning between the atomic transition energy \( E_j^{(e)} - E_i^{(g)} \) and the laser frequency \( \omega_{ij} \). \( \gamma_{ij} D_{ij}(\rho) \) describes the decay of excited state \( |e_j\rangle \) to ground state \( |g_i\rangle \). According to the Lindblad equation [29, 30]:

\[ D_{ij}(\rho) = -\frac{1}{2} \left\{ \sigma^\dagger_{ij} \sigma_{ij}, \rho \right\} + \sigma_{ij} \rho \sigma^\dagger_{ij} \]

where \( \{A, B\} \) is the anti-commutator of \( A \) and \( B \) and \( \sigma_{ij} = |g_i\rangle\langle e_j| \).

We show in appendix A that in the RWA approximation this time-independent formulation is possible if and only if there is at most one laser coupling associated to each transition \( g_i \leftrightarrow e_j \), as we have mentioned above, but also if to each such coupled transition, we can assign two real numbers \( \epsilon_i, \epsilon_j \) such that:

\[ \epsilon_i - \epsilon_j = \omega_{ij}. \]

This condition cannot always be met for arbitrary frequencies and can impose a relation between the laser frequencies.

There are also important cases of interest where control fields are added within the ground or excited manifolds. These can be resolved by block-diagonalizing them and redefining the ground \( |g_i\rangle \) or excited \( |e_j\rangle \) states so that the problem is brought back to the desired form. Some of these schemes have been shown to be desirable for example in the acceleration of cooling of trapped ions [31, 32].

**B. General conditions for the existence of dark states**

We say that a stationary state is dark when it involves only the ground-state manifold (there is no population in the excited states). In general the steady-state has non-zero density on both the ground state \( \{|g_i\rangle\} \) and excited state \( \{|e_j\rangle\} \) manifolds, and only fulfills the conditions of dark states for specific values of the parameters. The problem of tuning to dark states consists in finding the set of parameters, that is the laser intensities and frequencies, for which the steady state belongs to the ground state manifold \( \{|g_i\rangle\} \).

It is convenient to rewrite the Lindblad operator as a non-hermitian Hamiltonian part \( L_\hat{H} \) and a quantum jump operator \( J \), as has been written often for example in the study of blinking or quantum trajectory theory [33]:

\[ L\rho = L_\hat{H}\rho + J(\rho) \]

where \( L_\hat{H}\rho = -i(\hat{H} \rho - \rho \hat{H}^\dagger) \) with \( \hat{H} = H - i\Gamma \) with \( \Gamma = \sum_{ij} \gamma_{ij} \sigma^\dagger_{ij} \sigma_{ij} \) and \( J(\rho) = \sum_{ij} \gamma_{ij} \sigma_{ij} \rho \sigma^\dagger_{ij} \).

Our general method to obtain the conditions for dark states relies on the following theorem:

**Theorem 1.** The \( N \)-level system whose evolution is governed by the Lindblad operator \( L \) given by Eq. (2) has a dark state \( \rho_d \) if and only if \( L_\hat{H}\rho_d = 0 \).
In other words, a dark state is an eigenstate of \( \rho \) with a zero eigenvalue. The proof is given on appendix \([3]\) we just give the heuristic of the proof here. It is based on two observations.

1. In general the eigenvalues of \( \rho \) have a real part which is strictly negative, which is related to the fact that the eigenvalues of \( H \) have a strictly negative imaginary part due to the total decay rate \( \sum \gamma_{ij} \) of excited states \( \{|e_j\}\). The only way to have a zero eigenvalue is such that the corresponding eigenstate \( \rho d \) has no component on these decaying excited states \( \{|e_j\}\).

2. If \( \rho \), then also \( \rho d = 0 \) as the jump operator gives zero, \( J(\rho d) = 0 \), on any state in the subspace spanned by \( \{|g_i\}\).

In that way, when \( \rho = 0 \), we ensure that the corresponding eigenvector is a steady state of \( L \) with no component in the excited states. Hence, it is a dark state. The reverse is also true, all dark state \( \rho d \) fulfill \( \rho \rho d = 0 \).

As dark states belong to the subspace spanned by the ground states only, it is thus convenient to define the projection operator on this subspace \( P = \sum_{i=1}^{N_{g}}|g_i\rangle\langle g_i| \) and its orthogonal complement \( Q = \mathbb{1} - P = \sum_{j=1}^{N_{s}}|e_j\rangle\langle e_j| \). Where \( \mathbb{1} \) is the identity operator in the Hilbert space \( \mathcal{H} \) spanned by the \( N \) states. We also define super-projectors \( \mathcal{P} \) and \( \mathcal{Q} = \mathbb{1} - \mathcal{P} \), where here \( \mathcal{P} \) means the identity operator on the Liouville space of linear operators on \( \mathcal{H} \). These super-projectors acts as superoperator in the following way: \( \mathcal{P} \rho = \rho \mathcal{P} \) and \( \mathcal{Q} \rho = \rho \mathcal{Q} + \mathcal{Q} \rho \mathcal{P} + \mathcal{Q} \rho Q \).

By inserting the identity \( \mathbb{1} \), between \( \rho = 0 \), and projecting the resulting equation with the two super-projectors, we obtain the following two equations:

\[
\mathcal{P} \rho L_{\mathcal{H}} \mathcal{P} \rho = \mathcal{P} L_{\mathcal{H}} \mathcal{Q} \rho = 0 \\
\mathcal{Q} \rho L_{\mathcal{H}} \mathcal{P} \rho = \mathcal{Q} L_{\mathcal{H}} \mathcal{Q} \rho = 0. \tag{8}
\]

As dark states belong entirely to the ground state manifold, we can enforce the condition \( \mathcal{P} \rho = \rho \) and thus \( \mathcal{Q} \rho = 0 \). In appendix \([3]\) we show that using these conditions, Eqs. \([8]\) become:

\[
[\text{PHP}, \rho] = 0 \tag{9}
\]

\[
Q \rho P = 0. \tag{10}
\]

From the first equation, Eq. \([9]\), we infer that there exists a common orthonormal basis of \( \text{PHP} \) in which the matrix representation of \( \rho \) and \( \text{PHP} \) is diagonal. But \( \text{PHP} \) is diagonal in the \( \{\{g_i\}\} \) basis,

\[
\text{PHP} = \sum_{i=1}^{N_{g}} \mathcal{E}_{i} |g_i\rangle\langle g_i| \tag{11}
\]

where we have defined (see Eq. \([3]\) and Eq. \([4]\))

\[
\mathcal{E}_{i} = \sum_{j} \Delta_{ij}. \tag{12}
\]

Therefore, if the \( \text{PHP} \) spectrum is non degenerate then the only solutions to Eq. \([12]\) are matrices \( \rho \) which are also diagonal in this basis. But this is a trivial solution and in this case Eq. \([10]\) implies that \( V_{ij} = 0 \). Hence, non trivial solutions can arise if and only if \( \text{PHP} \) has a degenerate spectrum. This degeneracy condition translates to a constraint on the laser frequencies. For instance, the requirement that two \( \text{PHP} \) eigenvalues are equal, \( \mathcal{E}_{1} = \mathcal{E}_{2} \), consists in a relation between the detunings \( \sum \Delta_{ij} - \Delta_{i'j} \) = 0 (by Eq. \([12]\)), which translates into a relation between laser frequencies (see Eq. \([4]\)).

Let us denote \( \mathcal{P}_{s} \), the orthogonal projectors on the eigen-subspace of dimension \( d_{s} \), associated to the \( d_{s} - 1 \) times degenerate eigenvalue \( \mathcal{E}_{s} \). We have \( \sum_{s} \mathcal{P}_{s} = \mathbb{1} \), \( \mathcal{P}_{s} \mathcal{P}_{s'} = \delta_{ss'} \mathcal{P}_{s} \), and \( \sum_{s} d_{s} = N_{g} \). Then \( \text{PHP} \) can be written as

\[
\text{PHP} = \sum_{s} \mathcal{E}_{s} \mathcal{P}_{s} \tag{13}
\]

and the dark states \( \rho \) must have a block diagonal form:

\[
\rho = \sum_{\{s; d_{s} > 1\}} \mathcal{P}_{s} \rho \mathcal{P}_{s} = \sum_{\{s; d_{s} > 1\}} \mathcal{P}_{s} \rho \mathcal{P}_{s}, \tag{14}
\]

where \( \rho_{s} = \mathcal{P}_{s} \rho \mathcal{P}_{s}/\text{Tr}[\mathcal{P}_{s} \rho \mathcal{P}_{s}] \) is a \( d_{s} \times d_{s} \) normalized density matrix, and \( p_{s} = \text{Tr}[\mathcal{P}_{s} \rho \mathcal{P}_{s}] \).

The second constraint given by Eq. \([10]\) can now be written as:

\[
\forall s; \quad d_{s} > 1 \implies \text{QHP}_{s} \rho_{s} = 0. \tag{15}
\]

We deduce that \( \rho_{s} \) can in principle be any positive, Hermitian, with trace one linear operator defined on \( \text{ker}[\text{QHP}_{s}] \), the kernel of \( \text{QHP}_{s} \subset \text{P}_{s} \mathcal{H} \).

To conclude this section, we summarize: to have a dark state, \( \text{PHP} \) must have a degenerate spectrum, this puts a constraint on the laser frequencies. The dark state is a statistical superposition of states \( \rho_{s} \), defined on \( \text{ker}[\text{QHP}_{s}] \) where \( \mathcal{P}_{s} \) are the orthogonal projectors on the eigen-subspaces of \( \text{PHP} \), corresponding to degenerate eigenvalues of \( \text{PHP} \). Depending on the dimension of \( \text{ker}[\text{QHP}_{s}] \) this last condition may or may not impose a condition on the Rabi frequencies \( V_{ij} \), this is the subject of the next section.

### C. Dimension, unicity and purity

In addition to the constraint on laser frequencies, the condition that \( \rho_{s} \) must be defined on \( \text{ker}[\text{QHP}_{s}] \) can be satisfied if and only if the \( \text{ker}[\text{QHP}_{s}] \) is not empty, \( \dim[\text{ker}[\text{QHP}_{s}]] \geq 1 \). By the rank-nullity theorem, \( \dim[\text{ker}[\text{QHP}_{s}]] + \text{rank}[\text{QHP}_{s}] = \text{rank}[\mathcal{P}_{s}] = d_{s} \). Hence, a dark state can exist if and only if for at least one of the eigen-subspace \( \mathcal{P}_{s} \mathcal{H} \) of dimension \( d_{s} \),

\[
\text{rank}[\text{QHP}_{s}] \leq d_{s} - 1. \tag{16}
\]

As \( \text{rank}[\text{QHP}_{s}] \leq \text{rank}[Q] = N_{e} \), then two different cases are in order:
• case 1 \( N_e \leq d_s - 1 \). In this case, the Eq. (16) is always fulfilled, regardless of the values taken by the Rabi frequencies \( V_{ij} \). The constraint on the laser frequencies \( \omega_{ij} \), giving the degeneracy of PHP and determining the dimension \( d_s \) of the eigen-subspace is necessary and sufficient for the existence of a dark state. We have supposed that the constraint imposed by the RWA approximation (Eq. (6)) have already been fulfilled. The \( \Lambda \), the M systems and the so-called “fan” \cite{27} or “multipoled” systems \cite{21}, belong to this case. They will be discussed in more detail in the next section.

• case 2 \( N_e > d_s \). In this case, rank [QHP] \( \leq N_e - 1 \). Lowering the rank of QHP to a lower value than rank \( Q \) can not be obtained for all value of the Rabi frequencies. In other words, the \( V_{ij} \) must satisfy some relations such that the kernel QHP be non-empty. Therefore, in this case, the existence of a dark requires that in addition to the laser frequencies \( \omega_{ij} \), the Rabi frequencies \( V_{ij} \) must fulfill some constraints. The specific case \( N_g = N_e = 2 \) which belongs to this case, will be discussed with more details in the next section.

We see that in general, when \( \text{dim} \{ \ker \left[ QHP \right] \} > 1 \), or when there is more than one eigenvalues of PHP which is degenerate, then multiple dark states may exist. More specifically, the stationary dark state can be represented by any density operator \( \rho \) defined on \( \bigoplus_{\{s,d_i \geq 1\}} \ker \left[ QHP_s \right] \) (see Eq. (14) and Eq. (15)). That is, if \( N_s = \text{dim} \{ \ker \left[ QHP_s \right] \} \), then a stationary dark state can be represented by any block-diagonal \( M \times M \) density matrix, where \( M = \sum_{\{s,d_i \geq 1\}} N_s \), where the sum runs over all degenerate eigen-subspaces \( \ker \left[ QHP \right] \) of \( H \), and each block is an \( N_s \times N_s \) positive, Hermitian matrix. Therefore, the stationary dark states which is reached asymptotically in time will depend on the initial state. The dark state will be unique if and only if there is only one eigenvalue \( \mathcal{E}_s \) which is degenerate, and dim \{ker \left[ QHP_s \right] \} = 1. We note that in this case the dark state is a pure state. We conclude that, for dark states, unicity implies purity. Hence, if there is a mixed dark state then it is not unique. Indeed, suppose that the dark state is a mixed state \( \rho = p_1 |\psi_1 \rangle \langle \psi_1| + p_2 |\psi_2 \rangle \langle \psi_2| \), where \( p_1 \) and \( p_2 \) are the eigenvalues of \( \rho \) and \( |\psi_1 \rangle, |\psi_2 \rangle \) are the two corresponding orthonormal eigenvectors. Because \( \rho \) is a dark state, its two eigenstates must belong to \{ker \left[ PHP \right] \}. But then any linear combination or any statistical superposition of two states will fulfill the dark state condition Eq. (10). Then, there is not a unique dark state.

A simple way to achieve a unique dark state is to tune the frequencies such that the dimension of the unique degenerate subspace \( d_s \) fulfills the equality \( N_e = d_s - 1 \). As we are in the case 1 (\( N_e \leq d_s - 1 \)), there is a dark state regardless of the values taken by the \( V_{ij} \), but in addition, \text{dim} \{ \ker \left[ QHP_s \right] \} = d_s - \text{rank} \left[ PHP \right] = d_s - N_e = 1. \text{rank} \left[ PHP \right] = N_e = d_s - 1 \) because we suppose that each considered excited state \( |e_j \rangle \) is coupled to at least one ground state \( |g_i \rangle \) by a Rabi frequency \( V_{ij} \).

This is why such M systems \cite{27} have attracted attention as a generalisation of the very well known \( \Lambda \) systems where \( d_s = N_g = 2 \) and \( N_e = 1 \). Specific example illustrating these general considerations will be given in the next section.

III. EXAMPLES

In this section we illustrate the preceding discussion with four examples that illustrate the different cases from section II: a case with a unique stationary dark state, a case with a dark stationary subspace, and a case that is overspecified and whose dark state depends on the Rabi frequencies. The fourth example is the more complex system of the 11 hyperfine levels of \( ^{87}\text{Rb} \). For each case we will review the necessary condition to have a degenerate subspace and a non-zero kernel for QHP, and the resulting dark state subspaces.

A. Example 1: unique stationary state for zigzag systems \( (N_g = N_e + 1) \)

We refer to zigzag or M systems as those where the connectivity given by the laser fields follows the pattern ground-(excited-ground)\( n \) (Figure 2a). We consider in particular the M system with \( N_g = 3 \) and \( N_e = 2 \) which has been discussed elsewhere \cite{22}. It is a system where \( N_e = N_g + 1 \) so that \text{dim} \{ ker \left[ QHP \right] \} = 1 and \text{dim} \{ ker \left[ L \right] \} = 1, \) provided that all detunings are equal. Figure 2a shows the evolution of the populations with initial conditions \( \rho(t = 0) = |e_1 \rangle \langle e_1| \) in the site basis \{\( |g_i \rangle; i = 1, 2, 3 \}\} and \{\( |e_j \rangle; j = 1, 2 \}\} and in the eigenstate of \( H \) basis \{\( \phi_n \); \( n = 1, \cdots, 5 \)\}. As expected, the system evolves towards a pure dark state which here is \( |\phi_5 \rangle \).

Variations on the zigzag systems can be obtained by introducing additional coupling between ground and excited states. These modify the configuration of the stationary state over the ground state sites, but do not change its existence, uniqueness or purity. Because these additional couplings create connectivity loops, the frequency of these additional laser fields cannot be independently chosen if we want to satisfy the RWA (Figure 2b and Appendix A). In the particular case we are considering (see figure 2b), the additional Rabi frequency \( V_{31} \) must correspond to a laser frequency \( \omega_{31} \) fulfilling: \( \omega_{31} = \omega_{21} + \omega_{32} - \omega_{22} \). The maximum number of couplings is \( N_g N_e \), one for each couple \( (g_i, e_j) \). Other control fields that fall beyond the scope of this article, are worth mentioning. For example, Cerillo et al. propose the addition of a control field between the ground states of a \( \Lambda \) system as a means for accelerating the cooling rate \cite{31} \cite{32}. While the ground state can be prediagonalized and the magnitude of the ground-excited couplings redefined (thus leading to the starting point of this article),
this has as a consequence to mix the restriction on detunings with restrictions on Rabi frequencies, resulting in dark states that depend on the intensity of the laser field as well. This point will be retaken in Example 3, where other examples of intensity dependent dark states are illustrated.

![Diagram of the system](image)

FIG. 2: Populations of the $M$ system as a function of time in the a) Diagram of the $M$ system connectivity and probability amplitudes of the pure, dark, stationary state (red is positive probability amplitude, blue is negative). The dynamics of the system is plotted in the site basis and in the eigenstate basis. The system evolves towards a pure state occupying only the ground states. Parameters are $V_{21} = 0.56, V_{22} = 0.23, V_{32} = 0.45$, $\gamma_{11} = 0.04, \gamma_{12} = 0.01, \gamma_{13} = 0.09, \gamma_{21} = 0.14, \gamma_{22} = 0.02, \gamma_{23} = 0.04$. b) Same as a) in the presence of the coupling $V_{31} = 0.57$. In both cases the dark state is $\phi_5$ (all values in units of $V_{11}$).

### B. Example 2: dark stationary subspace for fan systems ($N_g \geq 2, N_e = 1$)

Fan or multipod systems consist of $N_g$ ground states and a single excited state (the $\Lambda$ configuration is also an instance of a fan system although it has a unique stationary state). In general the stationary states of fan systems can be arranged in a number of degenerate subspaces, generated by pure states $|\phi_n\rangle = \sum_{i=1}^{d_z} c_{ni} |g_i\rangle$ where from Eq. (10), the coefficients $c_{ni}$ must fulfill:

$$\sum_{i=1}^{d_z} V_{i1} c_{ni} = 0$$

(17)

where $V_{i1}$ are the Rabi frequencies of the laser coupling ground state $|g_i\rangle$ and the unique excited state $|e_1\rangle$. Fan systems have stationary states of high multiplicity: for each subspace of dimension $d_z$ the kernel of the Liouvillian will have a dimension $(d_z - 1)^2$ as well as $(d_z - 1)^2$ conserved quantities (obtained as the left eigenvalues of the Liouvillian $[34,35]$).

We specifically consider the case $N_g = 4$. We begin with all four ground states forming a degenerate manifold and detune them one by one to assess its effect on the steady-state - whether it remains dark, pure and how its multiplicity changes. We choose equal couplings to the excited state for ease of visualization, and unequal relaxations back to the ground state (Fig. 3.b). In both cases the dark state is $\phi_5$ (Tr($\rho^2$) = 0.38) determined by the conserved quantities that depend on the relaxation rates. Thus, although the condition to have the dark states depends exclusively on the Rabi frequencies and frequency detunings, the final reached state is a density matrix which depends also on the relaxation rates and on the initial state.

b) One detuned ground state ($d_1 = 3$). The system qualitatively similar to case a) has dim [ker $[QHP]$] = 3 and dim [ker $[L]$] = 9 (Fig. 3.d). A system initially in the excited state will evolve towards a mixed $3 \times 3$ density matrix (Tr($\rho^2$) = 0.38) determined by the conserved quantities which depend on the detunings.

c) Pairwise degenerate states ($d_1 = 2$ and $d_2 = 2$). Detuning a second ground state to the same value as the one in case b), results in pairwise degenerate levels (Fig. 3.c). We must separately consider the degenerate subspaces, so we have dim [ker $[QHP_1]$] = 1 and dim [ker $[QHP_2]$] = 1, and dim(ker($L$)) = $1^2 + 1^2 = 2$. The evolution converges to the mixture of two pure states $p_1 |\phi_1\rangle + p_2 |\phi_2\rangle$, where each $|\phi_n\rangle$ ($n = 1, 2$) is the stationary state of a $\Lambda$ system. The weights $p_n$ ($n = 1, 2$) depend on the dissipation rates. These are larger for the second manifold ($|g_3\rangle, |g_4\rangle$) than for the first one ($|g_1\rangle, |g_2\rangle$) and so the second manifold is more heavily populated.

d) Minimum degenerate manifold ($d_1 = 2$). The final detuning scheme keeps only a degenerate pair of levels (Fig. 3.d). Then, dim(ker($QHP_1$)) = 1 and dim(ker($L$)) = 1. Because the dark state is unique, it is also pure (see above). By properly choosing the detunings an experimentalist can localize in energy or space (if each energy level is spatially separated) via pumping.

We also note that because the excited states relax to the ground state, any dark state will accumulate all population and become the steady-state of the system. This is why in example d) the detuning of the other ground states does not render the state bright.
C. Example 3: A Rabi frequency conditionned dark state: Pairs of two-level systems \((N_b \leq N_e)\)

We consider a system with two levels in the excited state and two levels in the ground state (Figure 3). As \(d_s = 2\), we have \(\dim \ker [QHP] = 2 - \text{rank} [QHP]\). The generic case corresponds to \(\text{rank} [QHP] = \text{rank} [Q] = 2\) and in this case the kernel is empty - no dark state can exist in general, in contrast to the previous two examples. We will see that a dark state may exist but not for all values of the Rabi frequencies \(V_{ij}\). Furthermore, the existence of a loop in the connectivity constrains the laser frequencies to fulfill the relation: \(\omega_{21} = \omega_{11} + \omega_{22} - \omega_{12}\), such that the RWA results in a time independent Hamiltonian (see Appendix A).

The Hamiltonian is (after a convenient referencing of
the zero point energy):

$$H = \begin{bmatrix} 0 & 0 & V_{11} & V_{12} \\ 0 & \mathcal{E}_2 & V_{21} & V_{22} \\ V_{11}^* & V_{21}^* & \mathcal{E}_3 & 0 \\ V_{12}^* & V_{22}^* & 0 & \mathcal{E}_4 \end{bmatrix}$$  \hspace{1cm} (18)

The degeneracy condition implied by Eq. (9) requires that $\mathcal{E}_2 = 0$. The constraint given by Eq. (15) results in a relation between Rabi frequencies:

$$V_{11}V_{22} = V_{12}V_{21}. \hspace{1cm} (19)$$

Figures 5a and 5b show the dynamics for a $\Lambda$ system and a $N_g = 2, N_e = 2$ system, respectively, to compare the effect of an additional excited state tuned to the dark state condition. Although the dynamical evolution differs slightly, the stationary state is identical. Increasing one of the Rabi frequencies gets the system out of the dark state condition onto a mixed stationary state where all four states are occupied, as shown in Figure 5c.

We can understand the setup and restrictions on the Rabi Frequencies by viewing the system as a pair of $\Lambda$ geometries on the same ground states. Because only one excited level is enough to fully specify the dark state condition (in general to fully specify a unique dark state one needs at least $N_g - 1$ excited states), the second $\Lambda$ system must be adapted to the ground state population specified by the first $\Lambda$ system. This can be achieved by tuning the Rabi frequencies.

The map of excited state population vs. Rabi frequency and detuning (see Fig. 4) reveals a dark state in frequency detuning as well as in Rabi frequency. The asymmetry in the map between positive and negative values of the Rabi frequency illustrates the phase sensitivity of the dark state.

Given a complex coupling $V_{ij} = F_{ij} e^{i\phi_{ij}}$, the requirement (19) separates the constraint on the magnitudes of the fields $|F_{11}| / |F_{22}| = |F_{12}| / |F_{21}|$ and on the relative phases $\phi_{11} + \phi_{22} = \phi_{12} + \phi_{21}$. Such a dependence of the dark state on the Rabi frequencies could result in new metrology tools.

D. Hyperfine splittings of $^{87}$Rb atoms

The characteristics of dark states outlined here allow a shortcut to establish their presence in more complex systems. As an example we take the study of CPT in multi-Zeeman-sublevel $^{87}$Rb atoms, as considered by Ling et. al in Ref. [36]. In their publication, Ling et. al have investigated the possibility of obtaining CPT using only two co-propagating linearly polarized lasers addressing the two transitions that connect the higher energy level $P_{1/2}F' = 1$ to the two lower levels $S_{1/2}, F = 2$ and $S_{1/2}, F = 1$, respectively. The main point is that each of these 3 levels has a hyperfine sublevel structure. Indeed, the $S_{1/2}, F = 2$ is composed by 5 degenerated sublevels ($m_F = -2, -1, \cdots, 2$), and both $P_{1/2}F' = 1$ and $S_{1/2}, F = 1$ are composed by 3 degenerated sublevels each ($m_F = -1, 0, 1$). Taking the quantization axis as the propagation direction, results in $\sigma^+, \sigma^-$ transitions with equal strength, as both lasers are linearly polarized [36].

The energy levels as well as all possible transitions ($\sigma^+$, $\sigma^-$ and $s$) are shown in Figure 6 (boxed scheme). We will expand the scope of possible transitions beyond those considered by Lin et al. by introducing additional lasers that can induce $\Delta m_F = 0$ and $\Delta m_F = \pm 1$ transitions. The selected schemes are shown in Figure 6. We apply to these cases the criteria developed in this article to obtain the number, dimension and purity of dark states without the need for expensive numerical simulations.

As a first step we identify the number of degenerate subspaces that can independently sustain dark states (see Eq. (13)). These degenerate subspaces can occur on account of different detunings (see e.g. Figure 4), or because the connectivity results in two decoupled (ground-excited) systems such as in scheme 1, 3, 6 and 7 of Figure 6. For simplicity we assume in our analysis that all detunings are equal. The next step is to calculate the kernel of $QHP_s$ (see Eq. (15)). The dimension of the resulting dark state manifold is $\dim[\ker(QHP_s)] = d_s - \dim[\text{rank}(QHP_s)]$. For the connectivities considered, if $N_g > N_e + 1$, there will always be a dark state for any field intensity, provided that the detuning condition is met (we note that $N_g$ and $N_e$ are only defined with respect to coupled states so that, for instance, $N_g = 5$ in Scheme 1 of figure 6 and the sublevels of the $F = 1$ manifold does not contribute to $N_g$). These are the cases of all schemes except 5. In this case, we need that $\dim[\text{rank}(QHP_s)] < N_e$, which can be obtained in the case by adjusting the intensities. The conditions are $-V_{11}V_{23} = V_{12}V_{21}V_{33}$, where we have used the nomenclature of this paper where the first index la-
We have presented an overview of dark states conditions on dissipative systems classified as a function of the number of ground and excited states. The condition can be reduced to a condition on the Hamiltonian part of the evolution. The number of excited and ground states naturally separates the systems into a case where CPT depends only on the detunings, and one where CPT appears only once conditions on the detunings and Rabi frequencies are met. When the kernel has multiplicities higher than one, the stationary states are mixed and the term coherent population trapping becomes less apt. Conserved quantities determine the final state and these depend on the dissipative rates of the system.

IV. CONCLUSIONS

We use throughout the article the rotating wave approximation (RWA) to turn the time-dependent Hamiltonian into a time-independent Hamiltonian. For N-level systems with arbitrary connectivity this may impose important constraints on the wavelengths or detunings that can be used.

Let start from the original time-dependent Schrödinger equation for the unitary evolution operator generated by the time dependent Hamiltonian $H(t)$, $iU(t) = H(t)U(t)$, where $H(t)$ can be written as:

$$H(t) = \sum_{i=1}^{N} E_i |i⟩⟨i| + \sum_{i>j=1}^{N} F_{ij}(t) (|i⟩⟨j| + |j⟩⟨i|)$$ (A1)

and $F_{ij}(t) = 2F_{ij} \cos(\omega_{ij} t + \phi_{ij})$, where the $F_{ij}$, $\omega_{ij}$ and $\phi_{ij}$ are time-independent constants.

We look for a diagonal operator $H_0 = \sum_{i=1}^{N} \epsilon_i |i⟩⟨i|$, such that after applying the unitary operator $U_0 = e^{-iH_0t}$, the original time dependent Schrödinger equation becomes time-independent within a good approximation.

Specifically, let write $U(t) = U_0(t)U_1(t)$, then $U_1(t)$ fulfills the Schrödinger $i\dot{U}_1 = H_1(t)U_1(t)$, where $H_1 =$
TABLE I: Characteristics of the dark states corresponding to the systems illustrated in Figure 6. The first 3 columns indicate the polarization of the laser inducing the \( F=2 \leftrightarrow F'=1 \) transition and the next 3 columns are for the laser polarization coupling the states corresponding to the \( F=1 \leftrightarrow F'=1 \) transition. In some case 2 manifolds of dark states can take place: columns \( d_1 \) and \( d_2 \). In each \( d_1 \) and \( d_2 \) column, the atomic states involved, the dimension, purity and the number of cycles of the dark states are indicated. Scheme 5 is marked as a dark state, although the conditions imposed on the intensities cannot be fulfilled with the radiative transitions’ degrees of freedom considered here (see text for details).

$$\sum_{i=1}^{N} (E_i - \epsilon_i) |i⟩⟨i| + V(t)$$

where \( V(t) \) is given by:

$$V(t) = \sum_{i>j=1}^{N} F_{ij}(t) \left( e^{i(\epsilon_i - \epsilon_j)\Delta t} |i⟩⟨j| + H.C \right), \tag{A2}$$

where H.C means the hermitian conjugate of the preceding term.

Using the explicit expression of \( F_{ij}(t) \) as \( F_{ij}(t) = (V_{ij} e^{i\omega_{ij}t} + V_{ji}^* e^{-i\omega_{ij}t}) \), where \( V_{ij} = F_{ij} e^{i\delta_{ij}} \), we obtain:

$$V(t) = \sum_{i>j=1}^{N} \left( V_{ij} e^{i(\epsilon_i - \epsilon_j - \omega_{ij})\Delta t} |i⟩⟨j| + H.C \right) + \left( V_{ij} e^{i(\epsilon_i - \epsilon_j + \omega_{ij})\Delta t} |i⟩⟨j| + H.C \right) \tag{A3}$$

The RWA consists in choosing a set of energies \( \epsilon_i \) such that

$$\epsilon_i - \epsilon_j = \omega_{ij} \tag{A4}$$

and such that the terms oscillating at the frequencies \( \epsilon_i - \epsilon_j + \omega_{ij} = 2\omega_{ij} \), corresponding to the third line of Eq. (A3), can be safely neglected.

Within this approximation, \( U_I(t) \) fulfills a time-independent Schrödinger equation, \( \dot{U}_I \simeq H_I U_I(t) \), where \( H_I \) is time independent and given by:

$$H_I = \sum_{i=1}^{N} (E_i - \epsilon_i) |i⟩⟨i| + \sum_{i>j=1}^{N} (V_{ij} |i⟩⟨j| + H.C). \tag{A5}$$

It is convenient to introduce the operators \( \sigma_{ij}^z = |i⟩⟨j| - |j⟩⟨i| \) and take advantage of the condition Eq. (A4), in order to rewrite \( H_I \) as:

$$H_I = \sum_{i>j=1}^{N} \left[ \Delta_{ij} \sigma_{ij}^z + (V_{ij} |i⟩⟨j| + H.C) \right]. \tag{A6}$$

where we have defined the detunings \( \Delta_{ij} = E_i - E_j - \omega_{ij} \) and where we have ignored a term proportional to the identity which corresponds to an arbitrary energy origin.

The unitary transform \( U_0 \) does not affect the dissipative part of the Lindblad operator (Eq. (2)). Indeed, \( \rho_t = U_0^\dagger \rho_0 U_0 \) fulfill the Lindblad equation \( \dot{\rho}_t = -i[H_I, \rho_t] + \sum_{ij} \gamma_{ij} D_{ij}(\rho) U_0 D_{ij}^\dagger(\rho) U_0^\dagger \tag{A5} \). Therefore within the RWA approximation, the Lindblad operator is time independent.

We note that this is possible only if we can find a solution of Eq. (A4), giving the \( N \) unknowns \( \epsilon_i \), as a function of the laser frequencies \( \omega_{ij} \). We infer that in general the number of equation must be less than \( N \). But this constraint is not sufficient. Even when the number of equations is less than \( N \), additional constraints on the laser frequencies \( \omega_{ij} \) may be imposed to obtain a solution of Eq. (A4). This is always the case when there is a closed cycle in the set of couple \( (i,j) \). For example, consider a \( N \)-level system with \( N \geq 3 \), where the transitions \( (i,j) = (1,2), (2,3) \) and \( (1,3) \) are driven by 3 different laser fields. Then, summing Eq. (A4), over \( i \) and \( j \), with \( i > j \), gives \( \omega_{12} + \omega_{23} + \omega_{13} = 0 \).

**Appendix B: Proof of theorem 1**

**Proof.** If \( \det[L_R] = 0 \), then \( L_R \) has a right eigenvector \( \rho_d \) which fulfills \( \dot{\rho}_d = -i[\Gamma, \rho_d] \). \( \rho_d \) is a steady state of the dynamics generated by \( L_R \). We separate explicitly the Hermitian and non-Hermitian part of \( \dot{H} \) and this condition becomes:

$$[H, \rho_d] - i[\Gamma, \rho_d] = 0 \tag{B1}$$
where \( \Gamma \) is given by \( \Gamma = \sum_{ij} \gamma_{ij} \sigma_{ij} \sigma_{ij}^\dagger \) with \( \sigma_{ij} = |g_i\rangle\langle e_j| \)

or

\[
\Gamma = \sum_{j=1}^{N_e} \Gamma_j |e_j\rangle\langle e_j|
\]  

\[\text{(B2)}\]

with \( \Gamma_j = \sum_{i=1}^M \gamma_{ij} > 0 \) is the total decay rate of the excited state \( |j\rangle \). Hence, \( \Gamma \) is a completely positive diagonal matrix and the ground states manifold \( \{ |g_i\rangle \} \) is a subspace of its kernel, \( \Gamma |g_i\rangle = 0; \forall i = 1, 2, \ldots, N_e \).

Taking the trace of Eq. (B1), gives us \( \text{Tr} \Gamma \rho_d = 0 \), as the trace of the commutator gives zeros. Using Eq. (B2), this last condition can be written as:

\[
\sum_{j=1}^{N_e} \Gamma_j \langle e_j | \rho_d |e_j\rangle = 0. \quad \text{(B3)}
\]

But each term of this sum is positive, therefore each term must be zero, thus \( \langle e_j | \rho_d |e_j\rangle = 0; \forall i = 1, 2, \ldots, N_e \). We conclude that \( \rho_d \) has no population in the excited state manifold, therefore it can neither have coherence involving excited states, \( \langle e_j | \rho_d |g_i\rangle = 0 \). We conclude that \( \rho_d \) lies in the ground state manifold \( \{ |g_i\rangle \} \).

Now we consider the original Lindblad operator \( L \) which includes the quantum jump operator \( \sum_{ij} \gamma_{ij} \sigma_{ij} \rho_d \sigma_{ij}^\dagger \), written explicitly as:

\[
J(\rho_d) = \sum_{ij} |g_i\rangle\langle e_j| \langle e_j| \rho_d |g_i\rangle = 0
\]

\[\text{(B4)}\]

where we have used \( \langle e_j | \rho_d |e_j\rangle = 0 \). Therefore \( L \rho_d = 0 \).

Finally \( \rho_d \) is a steady state of \( L \) with no component in the excited state manifold, hence it is a dark state.

We have thus proved that if \( L \rho = 0 \) then \( \rho \) is a dark state. The converse, is obviously true.

**Appendix C: From super-projector to projector**

Starting from equations

\[
\mathcal{P} L_{\tilde{H}} P \rho + \mathcal{P} L_{\tilde{H}} Q \rho = 0
\]

\[
QL_{\tilde{H}} P \rho + Q L_{\tilde{H}} Q \rho = 0.
\]

\[\text{(C1)}\]

we would like to prove their equivalence with the following equations:

\[
[PHP, \rho] = 0
\]

\[
QHP \rho = 0.
\]

\[\text{(C2)}\]

\[\text{(C3)}\]

We first enforce \( \rho = P \rho \) and \( Q \rho = 0 \). Then:

\[
\mathcal{P} L_{\tilde{H}} P \rho = 0
\]

\[
QL_{\tilde{H}} P \rho = 0.
\]

\[\text{(C4)}\]

It is convenient to introduce the column form \( \rho_s \) of \( \rho \), which convert an \( n \times n \) matrix \( \rho \), to a \( N^2 \times \) column vector \( \rho_s \). In this transformation, the operation \( AP \sigma B^\dagger \) is mapped to \( B \otimes AP \sigma \), where \( B \) denote the conjugate of \( B \).

The effect of this mapping on the super-projector is as follows:

\[
\mathcal{P} \rho \rightarrow P \otimes P \rho_s
\]

\[
Q \rho \rightarrow (P \otimes Q + Q \otimes P + Q \otimes Q) \rho_s.
\]

\[\text{(C5)}\]

The superoperator \( L_{\tilde{H}} \) is mapped as:

\[
L_{\tilde{H}} \rho \rightarrow -i \left( \mathbb{1} \otimes \tilde{H} - \tilde{H}^\dagger \otimes \mathbb{1} \right) \rho_s
\]

\[\text{(C6)}\]

where \( \tilde{H}^\dagger \) is the transposed of \( \tilde{H} \). Using Eqs. (C5) and Eq. (C6), we can map Eqs. (C4) as:

\[
(P \otimes PHP - PHP \otimes P) \rho_s = 0
\]

\[
\left( P \otimes QHP - Q \tilde{H}^\dagger P \otimes P \right) \rho_s = 0,
\]

\[\text{(C7)}\]

\[\text{(C8)}\]

where we have used that \( PHP = P \tilde{H}^\dagger P = PHP \) and \( Q HP = QHP \). By reversing the mapping, the first equation (Eq. (C7)) gives

\[
[PHP, P \rho P] = 0
\]

\[\text{(C9)}\]

which is Eq. (9) of the main text.

For the second equation (Eq. (C8)) we use that the dark state fulfills \( \rho = P \rho P \). Therefore, the second term \( Q \tilde{H}^\dagger P \otimes P \rho_s = 0 \). Indeed, by reversing the mapping, it can be written as \( P \rho PQ \tilde{H}^\dagger P = 0 \). Hence, Eq. (C8) is equivalent to \( P \otimes QHP \rho_s = 0 \), which, by reversing the mapping, gives:

\[
QHP \rho P = 0,
\]

\[\text{(C10)}\]

which is Eq. (10) of the main text.

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FIG. 6: Possible connectivities for the hyperfine levels of $^{87}$Rb atoms. In the case of multiple manifold of dark states these are labeled by different colors (Blue (full) corresponds to $d_1$ and red (dashed) corresponds to $d_2$ of Table I). The case investigated in Ref. [36] corresponds to case 3.