Hamiltonian design to prepare arbitrary states of four-level systems

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We propose a method to manipulate four-level systems with specific coupling configurations by means of time-dependent couplings and constant energy shifts (detunings in quantum-optical realizations). We inversely engineer the Hamiltonian, in ladder, tripod, or diamond configurations, to prepare arbitrary states using the geometry of four-dimensional rotations to set the state populations; specifically, we use Cayley's factorization of a general rotation into right- and left-isoclinic rotations.

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I. INTRODUCTION

The coherent state manipulation and control of multiple-level quantum systems plays a significant role in atomic, molecular, and optical physics, with applications in existing or developing quantum technologies and quantum information processing [1]. Slow adiabatic protocols may be used for some transitions but they require long times, and detrimental effects of noise and perturbations accumulate. This has motivated the development of a set of techniques denominated "shortcuts to adiabaticity" (STA) to speed up the processes, which include counterdiabatic driving [2,3], inverse engineering based on invariants [4], Lie algebraic methods [5–8], fast quasi-adiabatic approaches [9], or fast-forward approaches [10–12].

Some of these methods require us to add terms in the Hamiltonian which are not easy or possible to implement in practice [4,13–15]. This problem has been addressed in specific systems by optimizing physically available terms [15], or by unitary transformations making use of the Lie algebraic structure of the dynamics [14,16–19]. However, generic solutions are not known and, as the system complexity and number of generators increase, the Lie algebraic methods may become numerically unstable or cumbersome to apply. These difficulties were already noticed in three-level or four-level systems where only certain couplings are allowed by symmetry, so alternative or complementary approaches are currently being explored that restrict the shortcuts to a set of physically allowed couplings [20].

The scope of STA methods has, in practice, outranged the original aim in many applications, so they can be applied to drive general transitions, regardless of whether the initial and final states can be adiabatically connected: for example, transitions where the initial state is an eigenstate of the initial Hamiltonian whereas the final state is not an eigenstate of the final Hamiltonian. This broader perspective merges with "inverse engineering" methods of the Hamiltonian to achieve unitary transformations or arbitrary transitions [21–27], and is the one adopted in this paper. In other words, even if we were initially motivated to solve the realizability of STA approaches, in fact, we shall address general four-level transitions for specific coupling configurations. In a similar vein, in Ref. [20], the authors proposed a scheme to control three-level system dynamics by separating the evolution into population changes, which may be parameterized using Rodrigues' rotation formula, and phase changes. This separation was used to inversely construct the Hamiltonian of the three-level system so as to drive a given transition with allowed couplings and vanishing forbidden couplings. Our goal here is to explore the extension of this concept to four-level systems. Certain couplings should not appear in the final Hamiltonian to implement specific four-level configurations such as a "diamond," a "tripod," or a "ladder." The population dynamics are now represented by rotations in a four-dimensional (4D) space, which are considerably more complex and less intuitive than in three dimensions. We have found a description of the rotation in terms of isoclinic matrices and quaternions, making use of Cayley's factorization, more convenient to perform the inversion than a generalized Rodrigues' formula, see Sec. II. In Sec. III, we find the Hamiltonian for the different configurations and provide examples. The appendices address technical points: long formulas in Appendix A, a short account of quaternions for 4D rotations in Appendix B, and details of quantum optical realizations in Appendix C.

Four-level systems are widely found and used in different contexts such as atomic physics, optical lattices [28–30], or waveguides [31–33], with applications such as electromagnetically induced transparency (EIT) [28,34,35], electromagnetically induced absorption [29], or beam splitting [31,32]. Most of the results in this paper are set in an abstract way, without specifying necessarily the physical system, but the notation is chosen as in a quantum-optical realization where atomic internal levels are coupled by laser fields, consistent with Rabi frequencies or detunings as matrix elements of the Hamiltonian. An explicit connection for the diamond configuration is worked out in Appendix C.

II. 4D ROTATIONS

Consider a four-level system in the state $|\psi(t)\rangle = c_1(t)|1\rangle + c_2(t)e^{i\varphi_2(t)}|2\rangle + c_3(t)e^{i\varphi_3(t)}|3\rangle + c_4(t)e^{i\varphi_4(t)}|4\rangle$, where $c_n(t)$ are real probability amplitudes of bare states $|n\rangle$ satisfiying the normalization $c_1^2(t) + c_2^2(t) + c_3^2(t) + c_4^2(t) = 1$, and the $\varphi_n(t)$ are relative phases. Following Ref. [20], we separate phase and amplitude information by writing $|\psi(t)\rangle = K(t)|\psi_r(t)\rangle$, where $K(t) = |1\rangle\langle 1| + e^{i\varphi_2(t)}|2\rangle\langle 2| + e^{i\varphi_3(t)}|3\rangle\langle 3| + e^{i\varphi_4(t)}|4\rangle\langle 4|$ and $|\psi_r(t)\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle + c_3(t)|3\rangle + c_4(t)|4\rangle$. $K(t)$ is a unitary transformation that contains the phases and $|\psi_r(t)\rangle$ represents a 4D vector on the surface of a 4D sphere. The state $|\psi(t)\rangle$ and the associated $|\psi_r(t)\rangle$ evolve via time-evolution operators $U(t)$ and $U_r(t)$ related by $U_r(t) = K^\dagger(t)U(t)K(0)$,

$$|\psi(t)\rangle = U(t)|\psi(0)\rangle, \tag{1}$$

$$|\psi_r(t)\rangle = U_r(t)|\psi_r(0)\rangle, \tag{2}$$

where we set initial time as 0. $U_r(t)$ represents a 4D rotation displacing points on the surface of the 4D sphere. In the 4D real space, we define the rotation Hamiltonian as

$$H_r(t) = i\hbar\dot{U}_r(t)U_r^\dagger(t), \tag{3}$$

such that $i\hbar\dot{U}_r(t) = H_r(t)U_r(t)$, whereas the total Hamiltonian is

$$\begin{aligned} H(t) &= i\hbar\dot{U}(t)U^\dagger(t) \\ &= i\hbar\dot{K}(t)K^\dagger(t) + K(t)H_r(t)K^\dagger(t). \end{aligned} \tag{4}$$

A. Rotations in $\mathbb{E}^4$

In 4D Euclidean space $\mathbb{E}^4$, a 4D rotation with center O can be expressed by a rotation matrix [36–38]

$$\begin{pmatrix} \cos\alpha & -\sin\alpha & 0 & 0 \\ \sin\alpha & \cos\alpha & 0 & 0 \\ 0 & 0 & \cos\beta & -\sin\beta \\ 0 & 0 & \sin\beta & \cos\beta \end{pmatrix} \tag{5}$$

in some appropriate orthogonal coordinates $\tilde{w}\tilde{x}\tilde{y}\tilde{z}$. Instead of having an axis of rotation as in 3D, 4D rotations are defined by a pair of completely orthogonal planes of rotation ($\tilde{w}$-$\tilde{x}$ and $\tilde{y}$-$\tilde{z}$ in the example), α and β are the angles of rotation with respect to the origin of any point on the $\tilde{w}$-$\tilde{x}$ and $\tilde{y}$-$\tilde{z}$ planes, respectively. More details can be found, e.g., in Refs. [36–38].

We may classify the rotations based on the α and β angles. If $\alpha \neq \beta \neq 0$, the rotation is a *double rotation*. There are two completely orthogonal (invariant) planes of rotation, with just the point O in common. Points in the first plane rotate through α with respect to the origin, and in the second plane rotate through β. For a general double rotation, the planes of rotation and angles are unique. Points which are not in the two planes rotate with respect to the origin through an angle between α and β.

If either of α or β are zero, the rotation is a *simple rotation* about the rotation center O. There is a fixed plane whose points do not change, whereas half lines from O orthogonal to this plane are displaced through the nonzero angle (α or β).

If $\alpha = \pm\beta$, the rotation is *isoclinic* and all nonzero points are rotated through the same angle. Then there are infinitely many pairs of orthogonal planes that can be treated as planes of rotation [36]. An isoclinic rotation can be left or right isoclinic (depending on whether $\alpha = \beta$ or $\alpha = -\beta$) [39]. According to Cayley's factorization [40,41], any 4D rotation matrix can be decomposed into the product of a right- and a left-isoclinic matrix. This decomposition is also conveniently expressed in terms of quaternions, as discussed in the following subsection.

B. Isoclinic rotations and quaternions

In 4D Euclidean space, an arbitrary point C can be represented as a column vector $(w,x,y,z)^T$ or as $C = w + xi + yj + zk$ [42,43]. If $|C|^2 = w^2 + x^2 + y^2 + z^2 = 1$ we call it unit quaternion. A general 4D rotation takes C to C', according to

$$C' = qCp, \tag{6}$$

where $q = q_w + q_x i + q_y j + q_z k$ and $p = p_w + p_x i + p_y j + p_z k$ are two unit quaternions. See Appendix A for a minimal introduction to quaternion algebra. In more common matrix language, the rotation reads

$$C' = M_L M_R C, \tag{7}$$

$$\begin{aligned} \begin{pmatrix} w' \\ x' \\ y' \\ z' \end{pmatrix} &= \begin{pmatrix} q_w & -q_x & -q_y & -q_z \\ q_x & q_w & -q_z & q_y \\ q_y & q_z & q_w & -q_x \\ q_z & -q_y & q_x & q_w \end{pmatrix} \\ &\quad \times \begin{pmatrix} p_w & -p_x & -p_y & -p_z \\ p_x & p_w & p_z & -p_y \\ p_y & -p_z & p_w & p_x \\ p_z & p_y & -p_x & p_w \end{pmatrix} \begin{pmatrix} w \\ x \\ y \\ z \end{pmatrix}, \end{aligned} \tag{8}$$

a formula due to Van Elfrinkhof [39,40]. M_L and M_R are isoclinic matrices [42,43], so $R = M_L M_R = M_R M_L$ is a 4D rotation matrix without loss of generality. Furthermore, $R^\dagger R = RR^\dagger = I$ due to $|q|^2 = q_w^2 + q_x^2 + q_y^2 + q_z^2 = 1$ and $|p|^2 = p_w^2 + p_x^2 + p_y^2 + p_z^2 = 1$. A summary of further relations between quaternions and 4D rotations, such as the relation between the isoclinic matrices and the orthogonal rotation planes and corresponding rotation angles, may be found in Appendix A.

III. HAMILTONIAN INVERSE ENGINEERING

In this section, we will make use of the rotation formula (8) to engineer the Hamiltonian and dynamics to drive a four-level system from an initial state to a final state. We substitute $U_r(t) = R(t)$ in Eq. (3), where the quaternion components are generally time dependent. The corresponding rotation Hamiltonian has the following structure:

$$\begin{aligned} H_r(t) &= i\hbar\dot{U}_r(t)U_r^\dagger(t) \\ &= i\hbar\begin{pmatrix} 0 & \Omega_{12}(t) & \Omega_{13}(t) & \Omega_{14}(t) \\ -\Omega_{12}(t) & 0 & \Omega_{23}(t) & \Omega_{24}(t) \\ -\Omega_{13}(t) & -\Omega_{23}(t) & 0 & \Omega_{34}(t) \\ -\Omega_{14}(t) & -\Omega_{24}(t) & -\Omega_{34}(t) & 0 \end{pmatrix}, \end{aligned} \tag{9}$$

where the real elements $\Omega_{nm}(t)$ are functions of the unit quaternion components (the explicit expression is given in Appendix B).

Taking the relative phases into account, the total Hamiltonian (4) is

$$\begin{aligned} H(t) &= i\hbar\dot{U}(t)U^\dagger(t) \\ &= i\hbar\dot{K}(t)K^\dagger(t) + K(t)H_r(t)K^\dagger(t) \\ &= \hbar[-\dot{\varphi}_2(t)|2\rangle\langle 2| - \dot{\varphi}_3(t)|3\rangle\langle 3| - \dot{\varphi}_4(t)|4\rangle\langle 4| \\ &\quad + i(e^{-i\varphi_2(t)}\Omega_{12}(t)|1\rangle\langle 2| + e^{-i\varphi_3(t)}\Omega_{13}(t)|1\rangle\langle 3| \end{aligned}$$

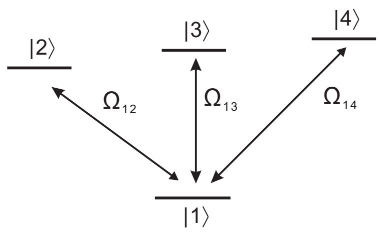

FIG. 1. Energy-level scheme for the inverse-tripod configuration with three nonzero couplings Ω_{12}, Ω_{13}, and Ω_{14}.

$$
\begin{aligned}
&+ e^{-i\varphi_4(t)}\Omega_{14}(t)|1\rangle\langle 4| + e^{i[\varphi_2(t)-\varphi_3(t)]}\Omega_{23}(t)|2\rangle\langle 3| \\
&+ e^{i[\varphi_2(t)-\varphi_4(t)]}\Omega_{24}(t)|2\rangle\langle 4| \\
&+ e^{i[\varphi_3(t)-\varphi_4(t)]}\Omega_{34}(t)|3\rangle\langle 4|)] + \text{H.c.}. \qquad (10)
\end{aligned}
$$

The physical interpretation of this Hamiltonian depends on the system considered. In quantum optics, this is to be interpreted as an interaction picture Hamiltonian where the diagonal terms are not energies of the bare levels, as depicted, e.g., in Fig. 1, but detunings, see Appendix C.

It proves useful to parametrize the quaternion components in terms of generalized spherical angles [44,45],

$$
\begin{aligned}
q_w &= \cos\gamma_1, \\
q_x &= \sin\gamma_1 \cos\theta_1, \\
q_y &= \sin\gamma_1 \sin\theta_1 \cos\phi_1, \\
q_z &= \sin\gamma_1 \sin\theta_1 \sin\phi_1, \\
p_w &= \cos\gamma_2, \\
p_x &= \sin\gamma_2 \cos\theta_2, \\
p_y &= \sin\gamma_2 \sin\theta_2 \cos\phi_2, \\
p_z &= \sin\gamma_2 \sin\theta_2 \sin\phi_2, \qquad (11)
\end{aligned}
$$

where $0 \leqslant \phi_{1,2} \leqslant 2\pi$, $0 \leqslant \theta_{1,2},\gamma_{1,2} \leqslant \pi$, and all angles may be time dependent. The explicit expression of the Hamiltonian (9) in terms of these angles is in Appendix B. We denote the initial ($t = 0$) and final states ($t = T$) as

$$
\begin{aligned}
|\psi(0)\rangle &= a_1|1\rangle + \sum_{k=2}^{4} a_k e^{i\epsilon_k}|k\rangle, \\
|\psi(T)\rangle &= b_1|1\rangle + \sum_{k=2}^{4} b_k e^{i\epsilon'_k}|k\rangle, \qquad (12)
\end{aligned}
$$

so that on the 4D sphere, $|\psi_r(0)\rangle = a_1|1\rangle + a_2|2\rangle + a_3|3\rangle + a_4|4\rangle$ $(a_1^2 + a_2^2 + a_3^2 + a_4^2 = 1)$ and $|\psi_r(T)\rangle = b_1|1\rangle + b_2|2\rangle + b_3|3\rangle + b_4|4\rangle$ $(b_1^2 + b_2^2 + b_3^2 + b_4^2 = 1)$. Since $|\psi_r(T)\rangle = U_r(T)|\psi_r(0)\rangle$, we have four equations:

$$
\begin{pmatrix} b_1 \\ b_2 \\ b_3 \\ b_4 \end{pmatrix} = U_r(T) \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix}. \qquad (13)
$$

If the angles at time T and the initial a_j components are fixed, these equations specify the final coefficients b_j. Alternatively, if both initial and final coefficients are given, we have four equations and six variables to play with. The additional freedom may be used to cancel certain terms in the Hamiltonian as demonstrated below.

A. The inverse tripod configuration

As a first four-level system, we consider the "inverse tripod" configuration in Fig. 1. The three excited states ($|2\rangle$, $|3\rangle$, and $|4\rangle$) are coupled to the ground state $|1\rangle$ by three couplings Ω_{12}, Ω_{13}, and Ω_{14}, respectively [34,46,47]. In this configuration, the transitions $|2\rangle \leftrightarrow |3\rangle$, $|2\rangle \leftrightarrow |4\rangle$, and $|3\rangle \leftrightarrow |4\rangle$ are not allowed, so we want to cancel these couplings in the Hamiltonian (9). One possible choice to set $\Omega_{23}(t) = \Omega_{34}(t) = \Omega_{24}(t) = 0$ is

$$
\begin{aligned}
\phi_2 &= \phi_1 = \phi, \\
\theta_2 &= \theta_1 = \theta, \\
\gamma_2 &= \gamma_1 = \gamma(t), \qquad (14)
\end{aligned}
$$

see Eq. (B1), where ϕ and θ are constants and $\gamma(t)$ may generally depend on time. The angles are equal for both isoclinic matrices, so $U_r(t)$ becomes, geometrically, a simple rotation (see Appendix A), and the rotation Hamiltonian reduces to

$$
\begin{aligned}
H_r(t) = &-2i\hbar\{[\dot\gamma(t)\cos\theta]|1\rangle\langle 2| + [\dot\gamma(t)\cos\phi\sin\theta]|1\rangle\langle 3| \\
&+ [\dot\gamma(t)\sin\phi\sin\theta]|1\rangle\langle 4|\} + \text{H.c.}. \qquad (15)
\end{aligned}
$$

For this particular case, the couplings

$$
\begin{aligned}
\Omega_{12}(t) &= 2\dot\gamma(t)\cos\theta, \\
\Omega_{13}(t) &= 2\dot\gamma(t)\sin\theta\cos\phi, \\
\Omega_{14}(t) &= 2\dot\gamma(t)\sin\theta\sin\phi, \qquad (16)
\end{aligned}
$$

take the form of cartesian coordinates of a point on a sphere in terms of spherical coordinates. Starting from the ground state $|1\rangle$ we have freedom to achieve any final state. Setting $a_1 = 1$, $a_2 = a_3 = a_4 = 0$, and from Eq. (13) we get

$$
\begin{aligned}
b_1 &= A, \quad b_2 = BC, \\
b_3 &= BDE, \quad b_4 = BDF, \qquad (17)
\end{aligned}
$$

with

$$
\begin{aligned}
A &= \cos[2\gamma(T)], \quad B = \sin[2\gamma(T)], \\
C &= \cos\theta, \quad D = \sin\theta, \\
E &= \cos\phi, \quad F = \sin\phi, \qquad (18)
\end{aligned}
$$

obeying the conditions $A^2 + B^2 = 1$, $C^2 + D^2 = 1$, and $E^2 + F^2 = 1$. The system in Eq. (17) with the above conditions has solution

$$
\begin{aligned}
A &= b_1, \quad B = \sqrt{b_2^2 + b_3^2 + b_4^2}, \\
C &= \frac{b_2}{\sqrt{b_2^2 + b_3^2 + b_4^2}}, \quad D = \frac{\sqrt{b_3^2 + b_4^2}}{\sqrt{b_2^2 + b_3^2 + b_4^2}}, \\
E &= \frac{b_3}{\sqrt{b_3^2 + b_4^2}}, \quad F = \frac{b_4}{\sqrt{b_3^2 + b_4^2}}, \qquad (19)
\end{aligned}
$$

where we take positive square roots, so it is possible to drive population transfers between the ground state and any final state. To exemplify the method, let us implement the rotation

$|1\rangle \to |\psi_r(T)\rangle = \frac{1}{\sqrt{3}}(|2\rangle + |3\rangle + |4\rangle)$. Substituting $b_1 = 0$, $b_2 = 1/\sqrt{3}$, $b_3 = 1/\sqrt{3}$, and $b_4 = 1/\sqrt{3}$ in Eq. (19) and using Eq. (18) we get four equations for $\gamma(T)$, θ, and ϕ with solutions

$$\gamma(T) = \frac{\pi}{4}, \quad \theta = \arctan\sqrt{2}, \quad \phi = \frac{\pi}{4}. \tag{20}$$

We use an Ansatz for $\gamma(t)$ consistent with $\gamma(T)$, $\gamma(t) = \frac{\pi}{8}[1 - \cos(\frac{\pi t}{T})]$, to determine the time dependence of the Hamiltonian according to Eq. (16). Notice that this is just a simple choice; we could use different functions, e.g., to optimize some physically relevant variable or improve robustness.

Now let us discuss the phases. Consistent with specific initial and final phases, we use simple linear interpolation Ansätze,

$$\varphi_k(t) = \epsilon_k + \Delta_k t, \tag{21}$$

where

$$\Delta_k = (\epsilon_k' - \epsilon_k)/T, \quad (k = 2,3,4) \tag{22}$$

may be interpreted as constant detunings in a quantum-optical realization, see Appendix C. Substituting them in Eq. (10), the total Hamiltonian is

$$\begin{aligned} H(t) = -\hbar\Bigg\{\sum_{k=2}^{4}\Delta_k|k\rangle\langle k| + i[e^{-i(\epsilon_2+\Delta_2 t)}\Omega_{12}(t)|1\rangle\langle 2| \\ + e^{-i(\epsilon_3+\Delta_3 t)}\Omega_{13}(t)|1\rangle\langle 3| \\ + e^{-i(\epsilon_4+\Delta_4 t)}\Omega_{14}(t)|1\rangle\langle 4|]\Bigg\} + \text{H.c.}. \end{aligned} \tag{23}$$

As an example, let us choose as initial and final phases $\epsilon_k = 0$, $\epsilon_k' = \frac{\pi}{3}$, $k = 2,3,4$, and apply Eq. (21) for $\varphi_k(t)$. Figure 2(a) shows the common smooth amplitude of the couplings, and Fig. 2(b) demonstrates the perfect population transfer.

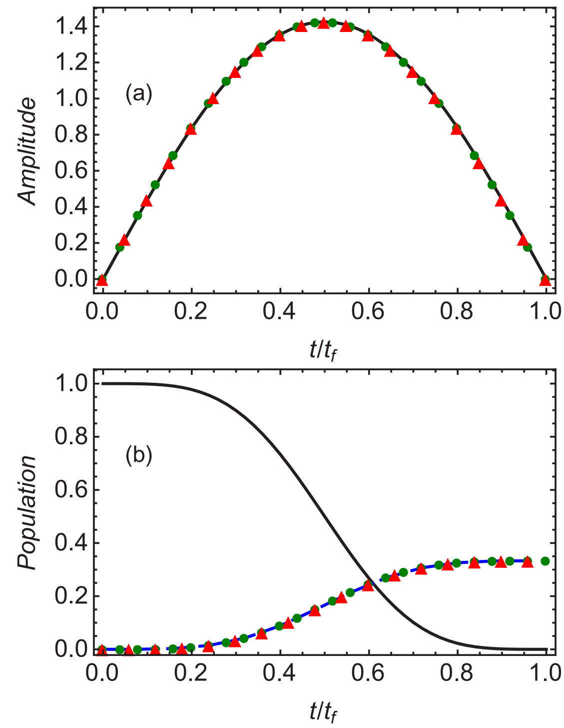

FIG. 2. (a) Overlapping couplings $\Omega_{12}(t)$ (solid black line), $\Omega_{13}(t)$ (green dots), and $\Omega_{14}(t)$ (red triangles). (b) Populations of $|1\rangle$ (solid black line), $|2\rangle$ (long-dashed blue line), $|3\rangle$ (green dots), and $|4\rangle$ (red triangles). Parameters: $\phi = \frac{\pi}{4}$, $\theta = \arctan\sqrt{2}$, $\epsilon_k = 0$, and $\epsilon_k' = \pi/3$, for $k = 2,3,4$.

B. The diamond configuration

Now we will focus on the diamond configuration shown in Fig. 3. In this configuration, one ground state $|1\rangle$ is coupled in a V-type structure to two intermediate states $|2\rangle$, $|3\rangle$, which are themselves coupled to a common excited state $|4\rangle$ in a Λ-type structure (see examples in atomic systems in Refs. [35,48,49] and in optical lattices in Ref. [30]). Figure 3 shows that the transitions $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ are not allowed so, they must be canceled in the Hamiltonian (9). To remove the unwanted terms, we proceed similarly as in the inverse tripod, taking now

$$\begin{aligned} \phi_1 &= \phi_2 = 0, \\ \dot\theta_1 &= \dot\theta_2 = \dot\phi_1 = \dot\phi_2 = 0, \end{aligned} \tag{24}$$

to achieve $\Omega_{14}(t) = \Omega_{23}(t) = 0$, which gives for the other couplings

$$\begin{aligned} \Omega_{12}(t) &= -[\dot\gamma_1(t)\cos\theta_1 + \dot\gamma_2(t)\cos\theta_2], \\ \Omega_{13}(t) &= -[\dot\gamma_1(t)\sin\theta_1 + \dot\gamma_2(t)\sin\theta_2], \\ \Omega_{24}(t) &= \dot\gamma_1(t)\sin\theta_1 - \dot\gamma_2(t)\sin\theta_2, \\ \Omega_{34}(t) &= -[\dot\gamma_1(t)\cos\theta_1 - \dot\gamma_2(t)\cos\theta_2]. \end{aligned} \tag{25}$$

The evolution operator $U_r(t)$ simplifies, and the rotating Hamiltonian becomes

$$\begin{aligned} H_r(t) = -\hbar\{&[\dot\gamma_1(t)\cos\theta_1 + \dot\gamma_2(t)\cos\theta_2]|1\rangle\langle 2| \\ &+ [\dot\gamma_1(t)\sin\theta_1 + \dot\gamma_2(t)\sin\theta_2]|1\rangle\langle 3| \\ &+ [-\dot\gamma_1(t)\sin\theta_1 + \dot\gamma_2(t)\sin\theta_2]|2\rangle\langle 4| \\ &+ [\dot\gamma_1(t)\cos\theta_1 - \dot\gamma_2(t)\cos\theta_2]|3\rangle\langle 4|\} + \text{H.c.}. \end{aligned} \tag{26}$$

To design the Hamiltonian for a rotation from $|\psi_r(0)\rangle = |1\rangle$, we set $a_1 = 1$, $a_2 = a_3 = a_4 = 0$ and substitute the unitary

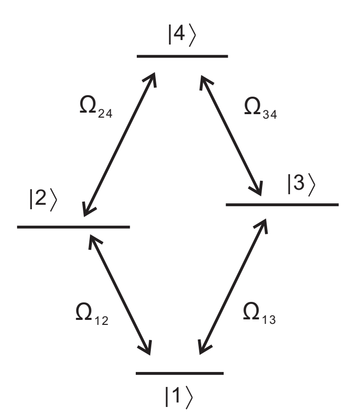

FIG. 3. Energy-level scheme for the diamond-type configuration with four couplings Ω_{12}, Ω_{13}, Ω_{24}, and Ω_{34}.

evolution matrix in Eq. (13),

$$b_1 = AB - CD(EF + GH), \quad b_2 = CBE + ADF,$$
$$b_3 = CBG + ADH, \quad b_4 = CD(HE - GF), \tag{27}$$

where

$$\begin{aligned} A &= \cos\gamma_1(T), \quad B = \cos\gamma_2(T),\\ C &= \sin\gamma_1(T), \quad D = \sin\gamma_2(T),\\ E &= \cos\theta_1, \quad F = \cos\theta_2,\\ G &= \sin\theta_1, \quad H = \sin\theta_2, \end{aligned} \tag{28}$$

so that $A^2 + C^2 = 1$, $B^2 + D^2 = 1$, $E^2 + G^2 = 1$, and $F^2 + H^2$. The solution in terms of the final state coefficients is

$$A = \frac{b_3E - b_2G}{\sqrt{b_4^2 + (b_3E - b_2G)^2}}, \quad B = \frac{[(b_1b_3 + b_2b_4)E + (b_3b_4 - b_1b_2)G]\sqrt{b_4^2 + (b_3E - b_2G)^2}}{(b_3^2 + b_4^2)E^2 - 2b_2b_3EG + (b_2^2 + b_4^2)G^2},$$

$$C = \frac{b_4}{\sqrt{b_4^2 + (b_3E - b_2G)^2}}, \quad D = \sqrt{1 - \frac{[(b_1b_3 + b_2b_4)E + (b_3b_4 - b_1b_2)G]^2[b_4^2 + (b_3E - b_2G)^2]}{[(b_3^2 + b_4^2)E^2 - 2b_2b_3EG + (b_2^2 + b_4^2)G^2]^2}},$$

$$F = -\frac{[(b_4b_1 - b_2b_3)E + (b_2^2 + b_4^2)G]\sqrt{b_4^2 + (b_3E - b_2G)^2}}{[(b_3^2 + b_4^2)E^2 - 2b_2b_3EG + (b_2^2 + b_4^2)G^2]D},$$

$$H = \frac{[(b_3^2 + b_4^2)E - (b_2b_3 + b_1b_4)G]\sqrt{b_4^2 + (b_3E - b_2G)^2}}{[(b_3^2 + b_4^2)E^2 - 2b_2b_3EG + (b_2^2 + b_4^2)G^2]D}, \tag{29}$$

so there is freedom to fix the value of the angle θ_1, see Eq. (28). The other angles, $\gamma_{1,2}(T)$ and θ_2, are found from Eq. (28). As an example, we study the population transfer from $|1\rangle$ to the final states $|\psi(T)\rangle = \frac{1}{\sqrt{2}}(|2\rangle \pm i|3\rangle)$. Substituting $b_1 = 0$, $b_2 = 1/\sqrt{2}$, $b_3 = 1/\sqrt{2}$, and $b_4 = 0$ in Eq. (29), choosing $\theta_1 = \pi/2$ and using Eq. (28), we find for the angles the values

$$\gamma_1(T) = \pi, \quad \gamma_2(T) = \frac{\pi}{2}, \quad \theta_2 = -\frac{3\pi}{4}. \tag{30}$$

For $\gamma_1(t)$ and $\gamma_2(t)$, we pick out smooth functions consistent with the values at T,

$$\begin{aligned} \gamma_1(t) &= \frac{\pi}{2}\left[1 - \cos\left(\frac{\pi t}{T}\right)\right],\\ \gamma_2(t) &= \frac{\pi}{4}\left[1 - \cos\left(\frac{\pi t}{T}\right)\right]. \end{aligned} \tag{31}$$

To find the full Hamiltonian we use Eqs. (10) and (21), where the Δ_k are chosen to satisfy the boundary conditions of the example,

$$\begin{aligned} &\epsilon_k = 0,\\ &\epsilon_2' = 0, \quad \epsilon_3' = \pm\pi/2, \quad \epsilon_4' = 0. \end{aligned} \tag{32}$$

The results are shown in Fig. 4. Figure 4(b) shows the perfect population transfer.

C. The *N*-type configuration

The last four-level structure we study is the N-type level scheme [28], with three nonzero couplings Ω_{12}, Ω_{23}, and Ω_{34}, see Fig. 5 (the ladder configuration is equivalent from the point of view of the present inverse method and would be treated similarly.). This configuration is applied, for example, to realize the phenomenon of EIT and population transfers in optical lattice systems [28,29,50]. To eliminate the unwanted terms, i.e., to have $\Omega_{13}(t) = \Omega_{14}(t) = \Omega_{24}(t) = 0$ in Eq. (9), one possible solution is

$$\dot\phi_1 = \dot\phi_2 = \dot\theta_1 = \dot\theta_2 = 0, \tag{33}$$

$$\phi_1 = \phi_2 = \frac{\pi}{2}, \tag{34}$$

$$\dot\gamma_1 = -\frac{\sin\theta_2}{\sin\theta_1}\dot\gamma_2. \tag{35}$$

The Hamiltonian $H_r(t)$ becomes

$$\begin{aligned} H_r(t) = i\hbar[&(\cot\theta_1\sin\theta_2 - \cos\theta_2)\dot\gamma_2(t)|1\rangle\langle 2|\\ &+ 2\sin\theta_2\dot\gamma_2(t)|2\rangle\langle 3|\\ &+ (\cot\theta_1\sin\theta_2 + \cos\theta_2)\dot\gamma_2(t)|3\rangle\langle 4|] + \text{H.c.}, \end{aligned} \tag{36}$$

and the couplings are

$$\begin{aligned} \Omega_{12}(t) &= \dot\gamma_2(t)(\sin\theta_2\cot\theta_1 - \cos\theta_2),\\ \Omega_{23}(t) &= 2\dot\gamma_2(t)\sin\theta_2,\\ \Omega_{34}(t) &= \dot\gamma_2(t)(\sin\theta_2\cot\theta_1 + \cos\theta_2). \end{aligned} \tag{37}$$

Unlike the previous cases, we do not find an analytical expression for the general solution of $U_r(T)$ in Eq. (13) for the initial

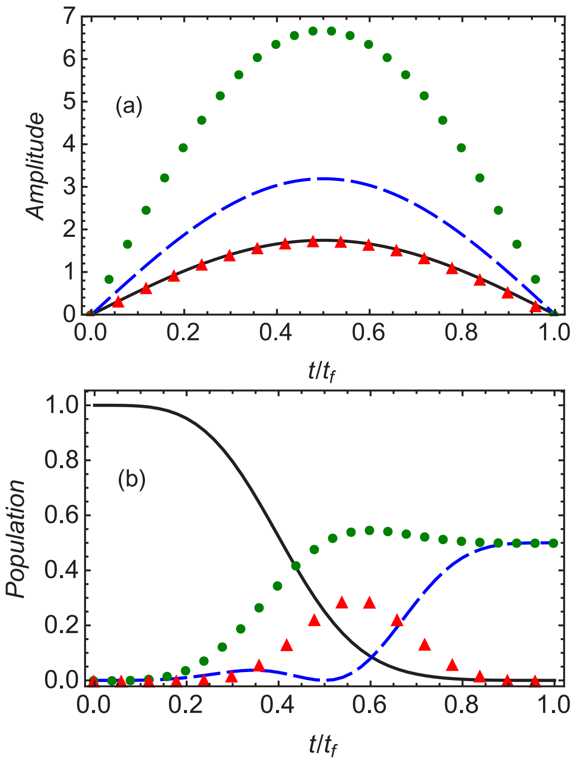

FIG. 4. (a) Couplings $\Omega_{12}(t)$ (solid black line), $\Omega_{13}(t)$ (long-dashed blue line), $\Omega_{24}(t)$ (green dots), and $\Omega_{34}(t)$ (red triangles), $\Omega_{12}(t) = \Omega_{34}(t)$. (b) Populations of $|1\rangle$ (solid black line), $|2\rangle$ (long-dashed blue line), $|3\rangle$ (green dots), and $|4\rangle$ (red triangles). The parameters are $\phi_1 = \phi_2 = 0, \theta_1 = \frac{\pi}{2}, \theta_2 = -\frac{3\pi}{4}, \epsilon_k = 0, \epsilon_2' = \epsilon_4' = 0$, and $\epsilon_3' = \pm\pi/2$.

state $|\psi_r(0)\rangle = |1\rangle$. However, for a given final state, the system can be solved to get the needed angles. As an example, let us engineer the interaction for a rotation from $|\psi_r(0)\rangle = |1\rangle$ to $|\psi_r(T)\rangle = |4\rangle$. From Eq. (13) and $U_r(T)$, we get four equations for $\gamma_1(T)$, $\gamma_2(T)$, [note that $\gamma_1 = -\frac{\sin\theta_2}{\sin\theta_1}\gamma_2 + c$, see Eq. (35)], θ_1, and θ_2 with solutions $\theta_1 = \pi/6, \theta_2 = \pi/2, \gamma_2(T) = -\pi/2$, $\gamma_1(T) = \pi$. We choose again $\gamma_2(t) = \frac{\pi}{4}[\cos(\frac{\pi t}{T}) - 1]$ as a smooth Ansatz, so $H_r(t)$ takes the form

$$H_r(t) = i\hbar[\Omega_{12}(t)|1\rangle\langle 2| + \Omega_{23}(t)|2\rangle\langle 3| + \Omega_{34}(t)|3\rangle\langle 4|] + \text{H.c.}, \tag{38}$$

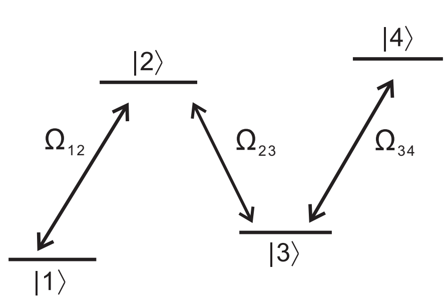

FIG. 5. Energy-level scheme for the four-level N configuration. There are three allowed couplings, Ω_{12}, Ω_{23}, and Ω_{34}.

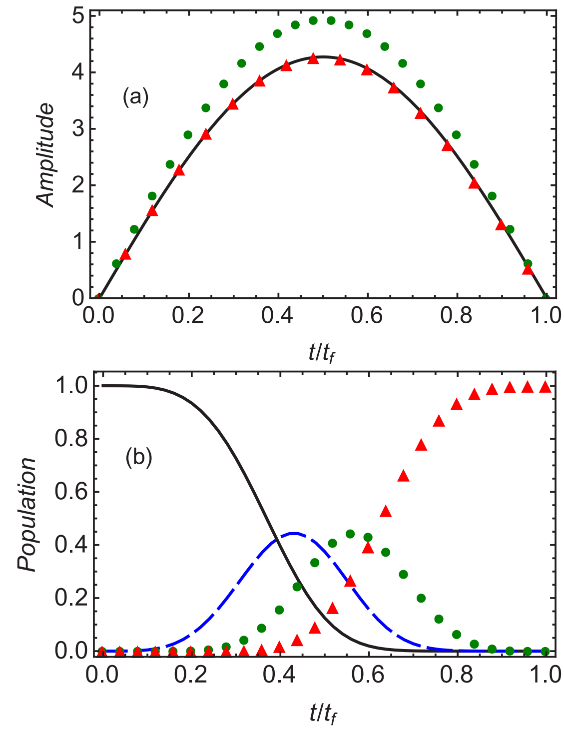

FIG. 6. (a) Couplings $\Omega_{12}(t)$ (solid black line), $\Omega_{23}(t)$ (green dots), and $\Omega_{34}(t)$ (red triangles). (b) Populations of $|1\rangle$ (solid black line), $|2\rangle$ (long-dashed blue line), $|3\rangle$ (green dots), and $|4\rangle$ (red triangles). The parameters are $\theta_1 = \pi/6$, $\theta_2 = \pi/2$, $\gamma_1(T) = \pi$, $\gamma_2(T) = -\pi/2$, $\epsilon_k = 0$, $\epsilon_2' = 0$, $\epsilon_3' = 0$, $\epsilon_4' = \pi/6$.

where

$$\Omega_{12}(t) = \Omega_{34}(t) = -\frac{\sqrt{3}\pi^2}{4T}\sin\left(\frac{\pi t}{T}\right),$$
$$\Omega_{23}(t) = -\frac{\pi^2}{2T}\sin\left(\frac{\pi t}{T}\right). \tag{39}$$

As for the phases, we may use the simple linear interpolation (21). For an example with boundary conditions

$$\epsilon_k = 0, \quad \epsilon_2' = \epsilon_3' = 0, \quad \epsilon_4' = \pi/6, \tag{40}$$

Fig. 6 shows the couplings (a) and population transfer (b) from state $|1\rangle$ to the desired state $e^{i\epsilon_4'}|4\rangle$.

IV. DISCUSSION

We have set a method to design four-level Hamiltonians so as to drive, in principle in an arbitrary time, specific transitions for different, preselected configurations of the couplings. For arbitrary final states, the method requires full control of the real and imaginary parts of the couplings, and of constant energy shifts. The possibility to realize this type of control will depend on the specific system and physical realization of the Hamiltonian (10). In an atomic system subjected to optical laser fields, this is an interaction picture Hamiltonian after applying the rotating wave approximation (RWA), see Appendix C, where the diagonal terms can be interpreted as detunings, and the nondiagonal terms as complex Rabi frequencies. Independent control may be required of the real and imaginary parts of the Rabi frequencies for final states with nonzero phases.

In all configurations studied, our method provides the Hamiltonian to transfer the ground state $|1\rangle$ to any other state "in one step," as represented in Eq. (1). How about other transitions? In principle, *any* transition is achievable in two steps by using $|1\rangle$ as an intermediate trampoline, jumping first from an arbitrary initial state to $|1\rangle$ with the inverse operation, and then from $|1\rangle$ to an arbitrary final state. Setting the domain of transitions where one stroke is enough is an interesting open question. Note that our emphasis here is in transitions among four-level system states, which differs in aim and scope from the work in Refs. [22,23] applied to the tripod. We consider different coupling configurations, while Refs. [22,23] concentrate on the tripod or, more generally, N-pod configurations. Specifically for the tripod, state $|1\rangle$ is regarded in Refs. [22,23] as an ancillary state to achieve arbitrary unitary matrices or transitions in the qutrit space spanned by $\{|2\rangle,|3\rangle,|4\rangle\}$. Four-level state engineering may be handled as in Refs. [22,23] with a four-pod scheme, i.e., with five levels, where four of them are coupled to the ancillary level.

We intend to apply these results in different scenarios—for example, to manipulate the spin state in quantum dots with spin-orbit coupling and electric-field control [51]. Broad fields for applications are multivalued quantum logic, e.g., qudit-based quantum information and quantum-gates engineering [26]. The N-type or diamond linkages, in particular, may be used to implement quantum phase-gate operations [35,52].

As for generalizations, the geometry of rotations in higher dimensions has been much less studied than that in 3D or 4D, but there are different approaches available [53,54] that could be used to generalize the current scheme to systems with more levels.

ACKNOWLEDGMENTS

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APPENDIX A: QUATERNIONS AND 4D ROTATIONS

A quaternion $\mathbf{q}$ can be defined as the sum of a scalar q_w and a vector $\vec{q}$, namely [55]

$$\mathbf{q} = q_w + \vec{q} = q_w + q_x\boldsymbol{i} + q_y\boldsymbol{j} + q_z\boldsymbol{k}. \tag{A1}$$

The rule of product of two quaternions is defined by

$$\boldsymbol{i}^2 = \boldsymbol{j}^2 = \boldsymbol{k}^2 = \boldsymbol{i}\boldsymbol{j}\boldsymbol{k} = -1. \tag{A2}$$

If $|\mathbf{q}|^2 = 1$, namely, $q_w^2 + q_x^2 + q_y^2 + q_z^2 = 1$, $\mathbf{q}$ is a unit quaternion and $\mathbf{q}^{-1} = \bar{q}$. If $\mathbf{u} = \vec{u}$ and $|\mathbf{u}|^2 = 1$, $\mathbf{u}$ is a pure unit quaternion, and every pure unit quaternion is a square root of -1. A unit quaternion can be expressed in terms of a real number γ and a pure unit quaternion $\mathbf{u}$ as

$$\mathbf{q} = e^{\mathbf{u}\gamma} = \cos\gamma + \mathbf{u}\sin\gamma. \tag{A3}$$

Consider two arbitrary unit quaternions $\mathbf{p}$ and $\mathbf{q}$. We may choose proper pure unit quaternions $\mathbf{u}$ and $\mathbf{v}$ with corresponding real numbers γ_1 and γ_2, so that $\mathbf{p} = e^{\mathbf{u}\gamma_1}$ and $\mathbf{q} = e^{\mathbf{v}\gamma_2}$. As noted in Sec. II A, an arbitrary rotation R in $\mathbb{E}^4$ of a four-vector C can be represented by the product qCp, associated with left and right isoclinic rotations with rotation angles γ_1 and γ_2. R also corresponds to a product of rotations in two mutually orthogonal planes [39,40,42,43]. If $\mathbf{u} \neq \pm\mathbf{v}$, R rotates the plane spanned by $\mathbf{u}+\mathbf{v}$ and $\mathbf{uv}-1$ through the angle $|\gamma_1+\gamma_2|$, and the plane spanned by $\mathbf{v}-\mathbf{u}$ and $\mathbf{uv}+1$ through the angle $|\gamma_1-\gamma_2|$, respectively [55]. If $\mathbf{u} = \pm\mathbf{v}$, the planes are spanned by 1 and $\mathbf{u}$ and its orthogonal complement, and the rotation angles are as well $|\gamma_1+\gamma_2|$ and $|\gamma_1-\gamma_2|$ [55].

APPENDIX B: HAMILTONIAN AND EVOLUTION

Using Eqs. (8), (9), and (11), the parameterized rotation Hamiltonian is given by

$$\begin{aligned}
H_r(t) &= i\hbar\dot{U}_r(t)U_r^\dagger(t) \\
&= i\hbar\{[\sin\gamma_1\sin\theta_1(\dot{\theta}_1\cos\gamma_1 - \dot{\phi}_1\sin\gamma_1\sin\theta_1) + \sin\gamma_2\sin\theta_2(\dot{\theta}_2\cos\gamma_2 + \dot{\phi}_2\sin\gamma_2\sin\theta_2) - \dot{\gamma}_1\cos\theta_1 - \dot{\gamma}_2\cos\theta_2]|1\rangle\langle 2| \\
&\quad + [\dot{\theta}_1\sin\gamma_1(\sin\gamma_1\sin\phi_1 - \cos\gamma_1\cos\theta_1\cos\phi_1) - \dot{\theta}_2\sin\gamma_2(\sin\gamma_2\sin\phi_2 + \cos\gamma_2\cos\theta_2\cos\phi_2) - \dot{\gamma}_1\sin\theta_1\cos\phi_1 \\
&\quad - \dot{\gamma}_2\sin\theta_2\cos\phi_2 + \dot{\phi}_1\sin\gamma_1\sin\theta_1(\cos\gamma_1\sin\phi_1 + \sin\gamma_1\cos\theta_1\cos\phi_1) + \dot{\phi}_2\sin\gamma_2\sin\theta_2(\cos\gamma_2\sin\phi_2 \\
&\quad - \sin\gamma_2\cos\theta_2\cos\phi_2)]|1\rangle\langle 3| + [-\dot{\theta}_1\sin\gamma_1(\sin\gamma_1\cos\phi_1 + \cos\gamma_1\cos\theta_1\sin\phi_1) + \dot{\theta}_2\sin\gamma_2(\sin\gamma_2\cos\phi_2 \\
&\quad - \cos\gamma_2\cos\theta_2\sin\phi_2) - \dot{\gamma}_1\sin\theta_1\sin\phi_1 - \dot{\gamma}_2\sin\theta_2\sin\phi_2 - \dot{\phi}_1\sin\gamma_1\sin\theta_1(\cos\gamma_1\cos\phi_1 - \sin\gamma_1\cos\theta_1\sin\phi_1) \\
&\quad - \dot{\phi}_2\sin\gamma_2\sin\theta_2(\cos\gamma_2\cos\phi_2 + \sin\gamma_2\cos\theta_2\sin\phi_2)]|1\rangle\langle 4| + [-\dot{\theta}_1\sin\gamma_1(\sin\gamma_1\cos\phi_1 \\
&\quad + \cos\gamma_1\cos\theta_1\sin\phi_1) - \dot{\theta}_2\sin\gamma_2(\sin\gamma_2\cos\phi_2 - \cos\gamma_2\cos\theta_2\sin\phi_2) - \dot{\gamma}_1\sin\theta_1\sin\phi_1 \\
&\quad + \dot{\gamma}_2\sin\theta_2\sin\phi_2 - \dot{\phi}_1\sin\gamma_1\sin\theta_1(\cos\gamma_1\cos\phi_1 - \sin\gamma_1\cos\theta_1\sin\phi_1) + \dot{\phi}_2\sin\gamma_2\sin\theta_2(\cos\gamma_2\cos\phi_2 \\
&\quad + \sin\gamma_2\cos\theta_2\sin\phi_2)]|2\rangle\langle 3| + [-\dot{\theta}_1\sin\gamma_1(\sin\gamma_1\sin\phi_1 - \cos\gamma_1\cos\theta_1\cos\phi_1) - \dot{\theta}_2\sin\gamma_2(\sin\gamma_2\sin\phi_2
\end{aligned}$$

$$+\cos\gamma_2\cos\theta_2\cos\phi_2)+\dot{\gamma}_1\sin\theta_1\cos\phi_1-\dot{\gamma}_2\sin\theta_2\cos\phi_2-\dot{\phi}_1\sin\gamma_1\sin\theta_1(\cos\gamma_1\sin\phi_1+\sin\gamma_1\cos\theta_1\cos\phi_1)$$
$$+\dot{\phi}_2\sin\gamma_2\sin\theta_2(\cos\gamma_2\sin\phi_2-\sin\gamma_2\cos\theta_2\cos\phi_2)]|2\rangle\langle 4|+[\sin\gamma_1\sin\theta_1(\dot{\theta}_1\cos\gamma_1-\dot{\phi}_1\sin\gamma_1\sin\theta_1)$$
$$-\sin\gamma_2\sin\theta_2(\dot{\theta}_2\cos\gamma_2+\dot{\phi}_2\sin\gamma_2\sin\theta_2)-\dot{\gamma}_1\cos\theta_1+\dot{\gamma}_2\cos\theta_2]|3\rangle\langle 4|\}+\text{H.c.}. \tag{B1}$$

APPENDIX C: CONNECTION WITH QUANTUM OPTICS (DIAMOND CONFIGURATION)

To relate the Hamiltonian of the inverse engineering approach, Eq. (10), to an interaction picture Hamiltonian for a four-level atom illuminated by laser fields, we assume a semiclassical description of the interaction of the atom with coupling laser fields. Neglecting atomic motion, the Hamiltonian in the Schrödinger picture for the diamond configuration and fields composed by combinations of out-of-phase quadrature components is

$$\begin{aligned}H(t)=\hbar\Big\{&\tilde{\Omega}_{12}(t)[|1\rangle\langle 2|+|2\rangle\langle 1|]\cos(\omega_{12}t+\phi_{12})-\tilde{\Omega}'_{12}(t)[|1\rangle\langle 2|+|2\rangle\langle 1|]\sin(\omega_{12}t+\phi_{12})\\&+\tilde{\Omega}_{13}(t)[|1\rangle\langle 3|+|3\rangle\langle 1|]\cos(\omega_{13}t+\phi_{13})-\tilde{\Omega}'_{13}(t)[|1\rangle\langle 3|+|3\rangle\langle 1|]\sin(\omega_{13}t+\phi_{13})\\&+\tilde{\Omega}_{24}(t)[|2\rangle\langle 4|+|4\rangle\langle 2|]\cos(\omega_{24}t+\phi_{24})-\tilde{\Omega}'_{24}(t)[|2\rangle\langle 4|+|4\rangle\langle 2|]\sin(\omega_{24}t+\phi_{24})\\&+\tilde{\Omega}_{34}(t)[|3\rangle\langle 4|+|4\rangle\langle 3|]\cos(\omega_{34}t+\phi_{34})-\tilde{\Omega}'_{34}(t)[|3\rangle\langle 4|+|4\rangle\langle 3|]\sin(\omega_{34}t+\phi_{34})+\sum_{i=2}^{4}\omega_i|i\rangle\langle i|\Big\},\end{aligned} \tag{C1}$$

where we use the vector basis $|1\rangle=\begin{pmatrix}1\\0\\0\\0\end{pmatrix},|2\rangle=\begin{pmatrix}0\\1\\0\\0\end{pmatrix},|3\rangle=\begin{pmatrix}0\\0\\1\\0\end{pmatrix},|4\rangle=\begin{pmatrix}0\\0\\0\\1\end{pmatrix}$. $\tilde{\Omega}_{ij}(t),\tilde{\Omega}'_{ij}(t)$ are the atom-field coupling strengths (Rabi frequencies), assumed real for simplicity, and ϕ_{ij} the phases of the coherent driving fields. The atomic levels $|i\rangle$ have energies $\hbar\omega_i$ and the fields have angular frequencies ω_{ij}. We choose the energy zero to match that of level $|1\rangle$ ($\omega_1=0$).

To transform the system into a laser-adapted interaction picture (rotating frame), we define the unitary operator

$$U_0(t)=\begin{pmatrix}1&0&0&0\\0&e^{i(\omega_{12}t+\phi_{12})}&0&0\\0&0&e^{i(\omega_{13}t+\phi_{13})}&0\\0&0&0&e^{i[(\omega_{12}+\omega_{24})t+\phi_{12}+\phi_{24}]}\end{pmatrix}. \tag{C2}$$

Using

$$H_I(t)=U_0(t)H(t)U_0^\dagger(t)+i\hbar\dot{U}_0(t)U_0^\dagger(t), \tag{C3}$$

and imposing the four-photon resonance condition [49,56,57]

$$\omega_{13}+\omega_{34}=\omega_{12}+\omega_{24}, \tag{C4}$$

the Hamiltonian in the interacting picture is

$$\begin{aligned}H_I(t)=\frac{\hbar}{2}\{&2(\omega_2-\omega_{12})|2\rangle\langle 2|+2(\omega_3-\omega_{13})|3\rangle\langle 3|+2(\omega_4-\omega_{12}-\omega_{24})|4\rangle\langle 4|\\&+\tilde{\Omega}_{12}(t)[(1+e^{-2i(\omega_{12}t+\phi_{12})})|1\rangle\langle 2|+(1+e^{2i(\omega_{12}t+\phi_{12})})|2\rangle\langle 1|]\\&+i\tilde{\Omega}'_{12}(t)[(1-e^{-2i(\omega_{12}t+\phi_{12})})|1\rangle\langle 2|-(1-e^{2i(\omega_{12}t+\phi_{12})})|2\rangle\langle 1|]\\&+\tilde{\Omega}_{13}(t)[(1+e^{-2i(\omega_{13}t+\phi_{13})})|1\rangle\langle 3|+(1+e^{2i(\omega_{13}t+\phi_{13})})|3\rangle\langle 1|]\\&+i\tilde{\Omega}'_{13}(t)[(1-e^{-2i(\omega_{13}t+\phi_{13})})|1\rangle\langle 3|-(1-e^{2i(\omega_{13}t+\phi_{13})})|3\rangle\langle 1|]\\&+\tilde{\Omega}_{24}(t)[(1+e^{-2i(\omega_{24}t+\phi_{24})})|2\rangle\langle 4|+(1+e^{2i(\omega_{24}t+\phi_{24})})|4\rangle\langle 2|]\\&+i\tilde{\Omega}'_{24}(t)[(1-e^{-2i(\omega_{24}t+\phi_{24})})|2\rangle\langle 4|-(1-e^{2i(\omega_{24}t+\phi_{24})})|4\rangle\langle 2|]\\&+\tilde{\Omega}_{34}(t)[(1+e^{-2i(\omega_{34}t+\phi_{34})})e^{-i\Phi}|3\rangle\langle 4|+(1+e^{2i(\omega_{34}t+\phi_{34})})e^{i\Phi}|4\rangle\langle 3|]\\&+i\tilde{\Omega}'_{34}(t)[(1-e^{-2i(\omega_{34}t+\phi_{34})})e^{-i\Phi}|3\rangle\langle 4|-(1-e^{2i(\omega_{34}t+\phi_{34})})e^{i\Phi}|4\rangle\langle 3|]\},\end{aligned} \tag{C5}$$

where

$$\Phi=\phi_{12}-\phi_{13}+\phi_{24}-\phi_{34}. \tag{C6}$$

Applying now a RWA to get rid of the counter-rotating terms we end up with

$$H_{I,RWA}(t) = \frac{\hbar}{2}\begin{pmatrix} 0 & \tilde{\Omega}_{12}(t)+i\tilde{\Omega}'_{12}(t) & \tilde{\Omega}_{13}(t)+i\tilde{\Omega}'_{13}(t) & 0 \\ \tilde{\Omega}_{12}(t)-i\tilde{\Omega}'_{12}(t) & \tilde{\Delta}_2 & 0 & \tilde{\Omega}_{24}(t)+i\tilde{\Omega}'_{24}(t) \\ \tilde{\Omega}_{13}(t)-i\tilde{\Omega}'_{13}(t) & 0 & \tilde{\Delta}_3 & (\tilde{\Omega}_{34}(t)+i\tilde{\Omega}'_{34}(t))e^{-i\Phi} \\ 0 & \tilde{\Omega}_{24}(t)-i\tilde{\Omega}'_{24}(t) & (\tilde{\Omega}_{34}(t)-i\tilde{\Omega}'_{34}(t))e^{i\Phi} & \tilde{\Delta}_4 \end{pmatrix}, \tag{C7}$$

where $\tilde{\Delta}_i$ $(i = 2,3,4)$ are the detunings defined as

$$\begin{aligned} \tilde{\Delta}_2 &= 2(\omega_2 - \omega_{12}), \\ \tilde{\Delta}_3 &= 2(\omega_3 - \omega_{13}), \\ \tilde{\Delta}_4 &= 2(\omega_4 - \omega_{12} - \omega_{24}). \end{aligned} \tag{C8}$$

Assuming that the phases of the coherent driving fields can be manipulated to satisfy

$$\phi_{12} - \phi_{13} + \phi_{24} - \phi_{34} = 0, \tag{C9}$$

the Hamiltonian in Eq. (C7) has the structure of the one in Eq. (10).

Notice that, the four-photon resonance condition (C4) is key to find a simple Hamiltonian structure in terms of the Rabi frequencies for closed-loop configurations. Equating the diagonal terms, $-\Delta_i = \tilde{\Delta}_i/2$, the laser (angular) frequencies are

$$\begin{aligned} \omega_{12} &= \omega_2 - \frac{\epsilon'_2 - \epsilon_2}{2T}, \\ \omega_{13} &= \omega_3 - \frac{\epsilon'_3 - \epsilon_3}{2T}, \\ \omega_{24} &= \omega_4 - \omega_2 + \frac{\epsilon'_2 - \epsilon_2}{2T} - \frac{\epsilon'_4 - \epsilon_4}{2T}, \end{aligned} \tag{C10}$$

and, to satisfy the four-photon resonance condition,

$$\omega_{34} = \omega_4 - \omega_3 - \frac{\epsilon'_4 - \epsilon_4}{2T} + \frac{\epsilon'_3 - \epsilon_3}{2T}. \tag{C11}$$

Comparing the nondiagonal terms in Eqs. (C7) and (10), we find the form of the Rabi frequencies,

$$\tilde{\Omega}_{jk} = 2e^{i(\phi_j - \phi_k)t}\Omega_{jk}, \tag{C12}$$

with $\phi_1 = 0$, ϕ_k $(k = 2,3,4)$ given by Eqs. (21) and (22), and $\tilde{\Omega}_{jk} = \tilde{\Omega}_{jk} + i\tilde{\Omega}'_{jk}$.

For other configurations that do not form a closed loop, similar steps may be followed, but the four-photon resonance condition is not imposed.

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