Hamiltonian design to prepare arbitrary states of four-level systems

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We propose a method to manipulate, possibly faster than adiabatically, four-level systems with 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.

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 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” to speed up the processes, which include counter-diabatic driving [2, 3], inverse engineering based on invariants [4], Lie algebraic methods [5–6], fast quasi-adiabatic approaches [7], or fast-forward approaches [8–10]. Some of these methods require to add terms in the Hamiltonian which are not easy or possible to implement in practice [4, 11, 12]. This problem has been addressed in specific systems by optimizing physically available terms [12], or by unitary transformations making use of the Lie algebraic structure of the dynamics [13, 14, 15]. 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 may be already noticed in three-level or four-level systems, so alternative or complementary approaches are currently being explored.

In Ref. [18] 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 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 4-level configurations such as a “diamond”, a “tripod”, or a “ladder”. The population dynamics is now represented by rotations in a four dimensional 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 formulae 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 [19–21], or waveguides [22–24], with applications such as electromagnetically induced transparency (EIT) [18, 25, 26], electromagnetically induced absorption [20], or beam splitting [22, 23]. 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 \(|\psi(t)\rangle = c_1(t)|1\rangle + c_2(t)e^{i\phi_2(t)}|2\rangle + c_3(t)e^{i\phi_3(t)}|3\rangle + c_4(t)e^{i\phi_4(t)}|4\rangle\), where \(c_n(t)\) are real probability amplitudes of bare states \(|n\rangle\) satisfying the normalization \(c_1^2(t) + c_2^2(t) + c_3^2(t) + c_4^2(t) = 1\), and the \(\phi_n(t)\) are relative phases. Following [18], we separate phase and amplitude information by writing \(|\psi(t)\rangle = K(t)|\psi_r(t)\rangle\), where \(K(t) = |1\rangle\langle 1| + e^{i\phi_2(t)}|2\rangle\langle 2| + e^{i\phi_3(t)}|3\rangle\langle 3| + e^{i\phi_4(t)}|4\rangle\langle 4|\) 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 4-dimensional (4D) vector on the surface of a 4D sphere. The states \(|\psi(t)\rangle\) and \(|\psi_r(t)\rangle\) evolve via time-evolution operators \(U(t)\) and \(U_r(t)\) related by \(U_r(t) = K(t)^{-1}U(t)K(0)\),

\[
|\psi(t)\rangle = U(t)|\psi(0)\rangle,
|\psi_r(t)\rangle = U_r(t)|\psi_r(0)\rangle,
\]

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 four-dimension real space, we define the rotation Hamiltonian as

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

$$
H(t) = i\hbar \dot{U}_r(t)U_r^\dagger(t) = i\hbar \dot{K}(t)K^\dagger(t) + K(t)H_r(t)K^\dagger(t). \quad (3)
$$

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

In four dimensional Euclidean space $\mathbb{E}^4$, a 4D rotation with centre $O$ can be expressed by a rotation matrix $[27-29]\:

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

(4)

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 ($\tilde{w}\tilde{z}$ and $\tilde{y}\tilde{z}$ in the example), $\alpha$ and $\beta$ are the angles of rotation with respect to the origin of any point on the $\tilde{w}\tilde{z}$ and $\tilde{y}\tilde{z}$ planes, respectively. More details can be found e.g. in [27-29].

We may classify the rotations based on the $\alpha$ and $\beta$ 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 $\alpha$ with respect to the origin, and in the second plane rotate through $\beta$. 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 $\alpha$ and $\beta$.

If either of $\alpha$ or $\beta$ 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 non-zero angle ($\alpha$ or $\beta$).

If $\alpha = \pm \beta$ the rotation is isoclinic and all non-zero points are rotated through the same angle. Then there are infinitely many pairs of orthogonal planes that can be treated as planes of rotation [30]. An isoclinic rotation can be left- or right-isoclinic (depending on whether $\alpha=\beta$ or $\alpha=-\beta$) [31]. According to Cayley’s factorization [31, 32], 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 $C = w + xi + yj + zk$ [33, 34]. 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

$$
C' = qCp,
$$

where $q = q_w + q_xi + q_yj + q_zk$ and $p = p_w + p_xi + p_yj + p_zk$ are two unit quaternions. See the Appendix A for a minimal introduction to quaternion algebra. In more common matrix language, the rotation reads

$$
C' = M_L M_R C,
$$

(6)

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

(7)

III. HAMILTONIAN INVERSE ENGINEERING

In this section, we will make use of the rotation formula (7) 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. (2), where the quaternion components are generally time dependent. The corresponding rotation Hamiltonian has the follow-
where $0 \leq \phi_{1,2} \leq 2\pi$, $0 \leq \theta_{1,2}, \gamma_{1,2} \leq \pi$, and all angles may be time dependent. The explicit expression of the Hamiltonian $H$ in terms of these angles is in Appendix 3.

We denote the initial and final states, at time $t = T$, as $|\psi_r(0)\rangle = a_1|1\rangle + a_2|2\rangle + a_3|3\rangle + a_4|4\rangle$ ($a_1^2 + a_2^2 + a_3^2 + a_4^2 = 1$) and $|\psi_r(T)\rangle = b_1|1\rangle + b_2|2\rangle + b_3|3\rangle + b_4|4\rangle$ ($b_1^2 + b_2^2 + b_3^2 + b_4^2 = 1$), with phases $\varphi_k(0) = \epsilon_k$ and $\varphi_k(T) = \epsilon'_k$ ($k = 2, 3, 4$). Since $|\psi_r(T)\rangle = U_r(T)|\psi_r(0)\rangle$, we have four equations

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

(11)

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 $\Omega_{12}$, $\Omega_{13}$, and $\Omega_{14}$, respectively. 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 $H$. One possible choice to set $\Omega_{23}(t) = \Omega_{34}(t) = \Omega_{24}(t) = 0$ is

$$
\begin{align*}
    q_u &= \cos \gamma_1, \\
    q_x &= \sin \gamma_1 \cos \theta_1, \\
    q_y &= \sin \gamma_1 \sin \theta_1 \cos \phi_1, \\
    q_z &= \sin \gamma_1 \sin \theta_1 \sin \phi_1, \\
    p_u &= \cos \gamma_2, \\
    p_x &= \sin \gamma_2 \cos \theta_2, \\
    p_y &= \sin \gamma_2 \sin \theta_2 \cos \phi_2, \\
    p_z &= \sin \gamma_2 \sin \theta_2 \sin \phi_2,
\end{align*}
$$

(10)

see Eq. 10, where $\phi$ and $\theta$ are constants and $\gamma(t)$ may generally depend on time. The angles are equal for both isoclinic matrices, so the evolution operator

![Energy level scheme for the inverse-tripod configuration with three non-zero couplings $\Omega_{12}$, $\Omega_{13}$ and $\Omega_{14}$](image)
is a simple rotation (see a geometrical explanation in Appendix A), and the rotation Hamiltonian reduces to

$$ H_r(t) = -2i\hbar \left\{ [\gamma(t) \cos \theta]|1\rangle \langle 2 | + [\gamma(t) \cos \phi \sin \theta]|1\rangle \langle 3 | + [\gamma(t) \sin \phi \sin \theta]|1\rangle \langle 4 | \right\} + H.c.$$  

(14)

For this particular case, the couplings

$$\Omega_{12}(t) = \dot{\gamma}(t) \cos \theta, \quad \Omega_{13}(t) = \dot{\gamma}(t) \sin \theta \cos \phi, \quad \Omega_{14}(t) = \dot{\gamma}(t) \sin \theta \sin \phi,$$  

(15)

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 substituting Eq. \ref{eq:13} in Eq. \ref{eq:11} we get

$$b_1 = \cos [2\gamma(T)], \quad b_2 = \sin [2\gamma(T)] \cos \theta, \quad b_3 = \sin [2\gamma(T)] \sin \theta \cos \phi, \quad b_4 = \sin [2\gamma(T)] \sin \theta \sin \phi,$$  

(16)

which we rewrite as

$$b_1 = A, \quad b_2 = B, \quad b_3 = BDE, \quad b_4 = BDF,$$  

(17)

with

$$A = \cos [2\gamma(T)], \quad B = \sin [2\gamma(T)], \quad C = \cos \theta, \quad D = \sin \theta, \quad E = \cos \phi, \quad F = \sin \phi,$$  

(18)

obeying the conditions $A^2 + B^2 = 1, C^2 + D^2 = 1$ and $E^2 + F^2 = 1$. The system in Eq. \ref{eq:17} with the above conditions has solution

$$A = b_1, \quad B = \sqrt{b_2^2 + b_3^2 + b_4^2}, \quad C = \frac{b_2}{\sqrt{b_2^2 + b_3^2 + b_4^2}}, \quad D = \frac{b_3}{\sqrt{b_2^2 + b_3^2 + b_4^2}}, \quad E = \frac{b_4}{\sqrt{b_2^2 + b_3^2 + b_4^2}},$$  

(19)

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 transition $|1\rangle \rightarrow \sum_{k=2}^4 \sqrt{\frac{1}{4}} (|2\rangle + |3\rangle + |4\rangle)$ in Appendix C. Substituting $b_1 = 0, b_2 = 1/\sqrt{3}, b_3 = 1/\sqrt{3}$ and $b_4 = 1/\sqrt{3}$ in Eq. \ref{eq:19} and using Eq. \ref{eq:13} we get four equations for $\gamma(T), \theta$ and $\phi$ with solutions

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

(20)

We now use an ansatz for $\gamma(t)$ consistent with $\gamma(T)$, $\gamma(t) = \frac{\pi}{4} - \frac{\pi}{4} [1 - \cos (\frac{\pi}{4})]$. It will determine the time-dependence of the Hamiltonian by Eq. \ref{eq:15}. Notice that this is just a simple choice, we could use different functions, e.g. to optimize some physically relevant variable or improve robustness.

For the phases we use simple linear interpolation ansatzes,

$$\varphi_k(t) = \epsilon_k + \Delta_k t,$$  

(21)

where

$$\Delta_k = (\epsilon'_k - \epsilon_k)/T, \quad (k = 2, 3, 4)$$  

(22)

may be interpreted as constant detunings in a quantum-optical realization, see Appendix C. Substituting them in Eq. \ref{eq:9}, the total Hamiltonian is

$$H(t) = -\hbar \left\{ \sum_{k=2}^4 \Delta_k |k\rangle \langle k | + i[\epsilon_k |e^{-i(t_2 + \Delta_k t)}\Omega_{12}(t)|1\rangle \langle 2 | + e^{-i(t_3 + \Delta_k t)}\Omega_{13}(t)|1\rangle \langle 3 | + e^{-i(t_4 + \Delta_k t)}\Omega_{14}(t)|1\rangle \langle 4 | + e^{i(t_2 - \epsilon_2) + (\Delta_2 - \Delta_3)t}\Omega_{23}(t)|2\rangle \langle 3 | + e^{i(t_2 - \epsilon_4) + (\Delta_2 - \Delta_4)t}\Omega_{24}(t)|2\rangle \langle 4 | + e^{i(t_3 - \epsilon_3) + (\Delta_3 - \Delta_4)t}\Omega_{34}(t)|3\rangle \langle 4 | \right\} + H.c.$$  

(23)

As an example, let us choose the following boundary conditions,

$$\epsilon_k = 0, \quad \epsilon'_k = \frac{\pi}{3}.$$  

(24)
FIG. 2: (Color online) (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 $k = 2, 3, 4$.

The evolution operator becomes

\[
U_r(t) = [\cos \gamma_1(t) \cos \gamma_2(t) - \cos (\theta_1 - \theta_2) \sin \gamma_1(t) \sin \gamma_2(t)] |1\rangle \langle 1| - [\sin \gamma_1(t) \cos \gamma_2(t) \cos \theta_1 + \cos \gamma_1(t) \sin \gamma_2(t) \cos \theta_2] |1\rangle \langle 2|
\]

\[
- [\sin \gamma_1(t) \cos \gamma_2(t) \sin \theta_1 + \cos \gamma_1(t) \sin \gamma_2(t) \sin \theta_2] |1\rangle \langle 3| - [\sin \gamma_1(t) \sin \gamma_2(t) \sin (\theta_1 - \theta_2)] |1\rangle \langle 4|
\]

\[
+ [\sin \gamma_1(t) \cos \gamma_2(t) \cos \theta_1 + \cos \gamma_1(t) \sin \gamma_2(t) \sin \theta_2] |2\rangle \langle 1| + [\cos \gamma_1(t) \cos \gamma_2(t) - \cos (\theta_1 + \theta_2) \sin \gamma_1(t) \sin \gamma_2(t)] |2\rangle \langle 2|
\]

\[
- [\sin \gamma_1(t) \sin \gamma_2(t) \sin (\theta_1 + \theta_2)] |2\rangle \langle 3| + [\sin \gamma_1(t) \cos \gamma_2(t) \sin \theta_1 - \cos \gamma_1(t) \sin \gamma_2(t) \sin \theta_2] |2\rangle \langle 4|
\]

\[
+ [\sin \gamma_1(t) \cos \gamma_2(t) \sin \theta_1 + \cos \gamma_1(t) \sin \gamma_2(t) \sin \theta_2] |3\rangle \langle 1| - [\sin \gamma_1 \sin \gamma_2 \sin (\theta_1 + \theta_2)] |3\rangle \langle 2|
\]

\[
+ [\cos \gamma_1(t) \cos \gamma_2(t) + \cos (\theta_1 + \theta_2) \sin \gamma_1(t) \sin \gamma_2(t)] |3\rangle \langle 3| - [\sin \gamma_1(t) \cos \gamma_2(t) \cos \theta_1 - \cos \gamma_1(t) \sin \gamma_2(t) \cos \theta_2] |3\rangle \langle 4|
\]

\[
- [\sin \gamma_1(t) \sin \gamma_2(t) \sin (\theta_1 - \theta_2)] |4\rangle \langle 1| - [\sin \gamma_1(t) \cos \gamma_2(t) \sin \theta_1 - \cos \gamma_1(t) \sin \gamma_2(t) \sin \theta_2] |4\rangle \langle 2|
\]

\[
+ [\sin \gamma_1(t) \cos \gamma_2(t) \cos \theta_1 - \cos \gamma_1(t) \sin \gamma_2(t) \cos \theta_2] |4\rangle \langle 3| + [\cos \gamma_1(t) \cos \gamma_2(t) + \cos (\theta_1 - \theta_2) \sin \gamma_1(t) \sin \gamma_2(t)] |4\rangle \langle 4|
\]

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 $\lambda$-type structure (see examples in atomic systems in Refs. [26, 39, 40] and in optical lattices in [21]). Figure 3 shows that the transitions $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ are not allowed so, they must be cancelled in the Hamiltonian $\tilde{H}$. To remove the unwanted terms we proceed similarly as in the inverse tripod, taking now

\[
\phi_1 = \varphi_2 = 0, \quad \theta_1 = \theta_2 = \phi_1 = \phi_2 = 0,
\]

(25)

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

\[
\Omega_{12}(t) = - [\dot{\gamma}_1(t) \cos \theta_1 + \dot{\gamma}_2(t) \cos \theta_2],
\]

\[
\Omega_{13}(t) = - [\dot{\gamma}_1(t) \sin \theta_1 + \dot{\gamma}_2(t) \sin \theta_2],
\]

\[
\Omega_{24}(t) = \dot{\gamma}_1(t) \sin \theta_1 - \dot{\gamma}_2(t) \sin \theta_2,
\]

\[
\Omega_{34}(t) = - [\dot{\gamma}_1(t) \cos \theta_1 - \dot{\gamma}_2(t) \cos \theta_2].
\]

(26)

The evolution operator becomes

\[
U_r(t) = \ldots
\]

(27)
and the rotating Hamiltonian is

\[
H_r(t) = -i\hbar \left\{ [\gamma_1(t) \cos \theta_1 + \gamma_2(t) \cos \theta_2] |1\rangle \langle 2 | \\
+ [\gamma_1(t) \sin \theta_1 + \gamma_2(t) \sin \theta_2] |1\rangle \langle 3 | \\
+ [-\gamma_1(t) \sin \theta_1 + \gamma_2(t) \sin \theta_2] |2\rangle \langle 4 | \\
+ [\gamma_1(t) \cos \theta_1 - \gamma_2(t) \cos \theta_2] |3\rangle \langle 4 | \right\} + H.c. \\
(28)
\]

To design the Hamiltonian for a transition from \( |\psi_r(0)\rangle = |1\rangle \), we set \( a_1 = 1 \), \( a_2 = a_3 = a_4 = 0 \) and substitute Eq. \( 27 \) in Eq. \( 11 \).

\[
b_1 = \cos \gamma_1(T) \cos \gamma_2(T) - \cos (\theta_1 - \theta_2) \sin \gamma_1(T) \sin \gamma_2(T), \\
b_2 = \sin \gamma_1(T) \cos \gamma_2(T) \cos \theta_1 + \cos \gamma_1(T) \sin \gamma_2(T) \cos \theta_2, \\
b_3 = \sin \gamma_1(T) \cos \gamma_2(T) \sin \theta_1 + \cos \gamma_1(T) \sin \gamma_2(T) \sin \theta_2, \\
b_4 = -\sin \gamma_1(T) \sin \gamma_2(T) \sin (\theta_1 - \theta_2). \\
(29)
\]

Using the change of variables

\[
A = \cos \gamma_1(T), \quad B = \cos \gamma_2(T), \\
C = \sin \gamma_1(T), \quad D = \sin \gamma_2(T), \\
E = \cos \theta_1, \quad F = \cos \theta_2, \\
G = \sin \theta_1, \quad H = \sin \theta_2, \\
(30)
\]

the equations in \( 29 \) become

\[
b_1 = AB - CD(\theta E + GH), \\
b_2 = CBE + ADF, \\
b_3 = CBG + ADH, \\
b_4 = CD(\theta E - GF), \\
(31)
\]

where \( A^2 + C^2 = 1, B^2 + D^2 = 1, E^2 + G^2 = 1 \) and \( F^2 + H^2 \). The solution in terms of the final state coefficients is

\[
A = \frac{b_1 E - b_2 G}{\sqrt{b_1^2 + (b_3 E - b_2 G)^2}}, \\
B = \frac{[(b_1 b_3 + b_2 b_4) E + (b_3 b_4 - b_1 b_2) G] \sqrt{b_1^2 + (b_3 E - b_2 G)^2}}{\sqrt{b_3^2 + b_4^2) E^2 - 2b_2 b_3 E G + (b_2^2 + b_4^2) G^2}}, \\
C = \frac{b_4}{\sqrt{b_1^2 + (b_3 E - b_2 G)^2}}, \\
D = \sqrt{1 - \frac{(b_1 b_4 + b_2 b_3) E + (b_3 b_4 - b_1 b_2) G)^2 [b_1^2 + (b_3 E - b_2 G)^2]}{[b_3^2 + b_4^2) E^2 - 2b_2 b_3 E G + (b_2^2 + b_4^2) G^2]^2}}, \\
F = -\frac{[(b_1 b_1 - b_2 b_3) E + (b_3 b_4 - b_1 b_2) G] \sqrt{b_1^2 + (b_3 E - b_2 G)^2}}{\sqrt{b_3^2 + b_4^2) E^2 - 2b_2 b_3 E G + (b_2^2 + b_4^2) G^2 D}}, \\
H = \frac{[(b_1^2 + b_4^2) E - (b_2 b_3 + b_1 b_4) G] \sqrt{b_1^2 + (b_3 E - b_2 G)^2}}{[b_3^2 + b_4^2) E^2 - 2b_2 b_3 E G + (b_2^2 + b_4^2) G^2 D}}, \\
(32)
\]

where \( E \) and \( G \) must obey \( E^2 + G^2 = 1 \), so there is freedom to fix the value of the angle \( \theta_1 \), see Eq. \( 30 \). The other angles, \( \gamma_{1,2}(T) \) and \( \theta_2 \), are found from Eq. \( 30 \). As an example, we study the population transfer from \( |1\rangle \) to the final state \( |\psi(T)\rangle = \frac{1}{\sqrt{2}} (|2\rangle \pm |3\rangle) \). Substituting \( b_1 = 0, b_2 = 1/\sqrt{2}, b_3 = 1/\sqrt{2} \) and \( b_4 = 0 \) in Eq. \( 32 \), choosing \( \theta_1 = \pi/2 \) and using Eq. \( 30 \) we find for the angles the values

\[
\gamma_1(T) = \pi, \quad \gamma_2(T) = \frac{\pi}{2}, \quad \theta_2 = -\frac{3\pi}{4}. \\
(33)
\]

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

\[
\gamma_1(t) = \frac{\pi}{2} \left[ 1 - \cos \left( \frac{\pi t}{T} \right) \right], \\
\gamma_2(t) = \frac{\pi}{4} \left[ 1 - \cos \left( \frac{\pi t}{T} \right) \right]. \\
(34)
\]

To find the full Hamiltonian we use Eq. \( 3 \) with \( \varphi_k(t) = \epsilon_k + \Delta_k t, \quad k = 2, 3, 4 \), where the \( \Delta_k \) are chosen to satisfy the boundary conditions of the example,

\[
\epsilon_k = 0, \quad \epsilon'_2 = 0, \epsilon_3 = \pm \pi/2, \epsilon'_4 = 0. \\
(35)
\]
and \( \Omega \) to eliminate the unwanted terms, i.e., to have \( \Omega_{34} \) transfers in optical lattice systems \([19, 20, 41]\). To eliminate the unwanted terms, i.e., to have \( \Omega_{34} \) transfers in optical lattice systems \([19, 20, 41]\).

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 \([19]\), with three non-zero couplings \( \Omega_{12}, \Omega_{23}, \) and \( \Omega_{34} \), see Fig. 5 (A ladder configuration would be treated similarly). This configuration is applied, for example, to realize the phenomenon of EIT and population transfers in optical lattice systems \([19, 20, 41]\). To eliminate the unwanted terms, i.e., to have \( \Omega_{13} \) and \( \Omega_{24} \) transfers in optical lattice systems \([19, 20, 41]\).

\[
\Omega_{12}(t) = 0 \quad \text{in Eq. (38)} \quad \text{one possible solution is}
\]

\[
\begin{align*}
\dot{\phi}_1 &= \dot{\phi}_2 = \dot{\theta}_1 = \dot{\theta}_2 = 0, \\
\phi_1 &= \phi_2 = \frac{\pi}{2}, \\
\dot{\gamma}_1 &= -\frac{\sin \theta_2}{\sin \theta_1} \dot{\gamma}_2.
\end{align*}
\]

The Hamiltonian \( H_r(t) \) becomes

\[
H_r(t) = i \left[ (\cot \theta_1 \sin \theta_2 - \cos \theta_2) \dot{\gamma}_2(t) |1 \rangle \langle 2 | + 2 \sin \theta_2 \dot{\gamma}_2(t) |2 \rangle \langle 3 | + (\cot \theta_1 \sin \theta_2 + \cos \theta_2) \dot{\gamma}_2(t) |3 \rangle \langle 4 | + H.c. \right]
\]

and the couplings are

\[
\begin{align*}
\Omega_{12}(t) &= \dot{\gamma}_2(t) (\sin \theta_2 \cot \theta_1 - \cos \theta_2), \\
\Omega_{23}(t) &= 2 \dot{\gamma}_2(t) \sin \theta_2, \\
\Omega_{34}(t) &= \dot{\gamma}_2(t) (\sin \theta_2 \cot \theta_1 + \cos \theta_2).
\end{align*}
\]

Unlike the previous cases, we do not find an analytical expression for the general solution of \( U_r(T) \) in Eq. (11) for the initial state \( |\psi(0)\rangle = |1\rangle \). However, for a given final state the system can be solved to get the needed angles. As an example, let us engineer the interaction to go from \( |\psi(0)\rangle = |1\rangle \) to \( |\psi(T)\rangle = |4\rangle \). From Eq. (11) and Eq. (32), we get four equations for \( \gamma_1(T), \gamma_2(T), \)

**FIG. 6: (Color online) (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 \( \theta_1 = \pi/6, \theta_2 = \pi/2, \gamma_1(T) = \pi, \gamma_2(T) = -\pi/2, \epsilon_2 = 0, \epsilon_3 = 0, \epsilon_4 = 0, \epsilon^\prime_2 = 0, \epsilon^\prime_3 = 0, \epsilon^\prime_4 = \pi/6. \)**
(b) from state Fig. 6 shows the couplings (a) and population transfer systems with more levels. That could be used to generalize the current scheme to or 4D, but there are different approaches available [43, 44] for higher dimensions has been much less studied than in 3D.

For example, to manipulate the spin state in quantum non-zero phases. For an example with boundary conditions.

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

\[ \varepsilon_k = 0, \quad \varepsilon'_2 = \varepsilon'_3 = 0, \quad \varepsilon'_4 = \pi/6. \]  

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

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 level of control will depend on the specific system and physical realization of the Hamiltonian [9].

In an atomic system subjected to optical laser fields, this is an interaction picture Hamiltonian after applying the rotating wave approximation, see Appendix C, where the diagonal terms can be interpreted as detunings, and the non-diagonal terms as complex Rabi frequencies. Independent control may be required of the real and imaginary parts of the Rabi frequencies for final states with non-zero phases.

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 [12]. As for generalizations, the geometry of rotations in higher dimensions has been much less studied that in 3D or 4D, but there are different approaches available [43, 44] that could be used to generalize the current scheme to systems with more levels.

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Appendix A: Quaternions and 4D rotations

A quaternion \( q \) can be defined as the sum of a scalar \( q_w \) and a vector \( \bar{q} \); namely [45]

\[ q = q_w + \bar{q} = q_w + q_xi + q_yj + q_zk. \]  

(A1)

The rule of product of two quaternions is defined by

\[ i^2 = j^2 = k^2 = ijk = -1. \]  

(A2)

If \( |q|^2 = 1 \), namely, \( q_w^2 + q_x^2 + q_y^2 + q_z^2 = 1 \), \( q \) is a unit quaternion and \( q^{-1} = \bar{q} \). If \( u = \bar{u} \) and \( |u|^2 = 1 \), \( u \) 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 \( \gamma \) and a pure unit quaternion \( u \) as

\[ q = e^{u\gamma} = \cos \gamma + u \sin \gamma. \]  

(A3)

Consider two arbitrary unit quaternions \( p \) and \( q \). We may choose proper pure unit quaternions \( u \) and \( v \) with corresponding real numbers \( \gamma_1 \) and \( \gamma_2 \), so that \( p = e^{u\gamma_1} \) and \( q = e^{v\gamma_2} \). As noted in Sec. II A an arbitrary rotation \( R \) in \( \mathbb{E}^4 \) of a 4-vector \( C \) can be represented by the product \( qCp \), associated with left and right isoclinic rotations with rotation angles \( \gamma_1 \) and \( \gamma_2 \). \( R \) also corresponds to a product of rotations in two mutually orthogonal planes [30, 31, 33, 34]. If \( u \neq \pm v \), \( R \) rotates the plane spanned by \( u + v \) and \( uv - 1 \) through the angle \( |\gamma_1 + \gamma_2| \), and the plane spanned by \( v - u \) and \( uv + 1 \) through the angle \( |\gamma_1 - \gamma_2| \), respectively [45]. If \( u = \pm v \), the planes are spanned by \( 1 \) and \( u \) and its orthogonal complement, and the rotation angles are as well \( |\gamma_1 + \gamma_2| \) and \( |\gamma_1 - \gamma_2| \) [45].

Appendix B: Hamiltonian and evolution

Using Eqs. (18,19), the parameterized Hamiltonian is given by
\[ H_r(t) = i\hbar \dot{U}_r(t)U_r^\dagger(t) \]
\[ = i\hbar \left[ \sin \gamma_1 \sin \theta_1 (\dot{\phi}_1 \sin \gamma_1 \sin \theta_1) + \sin \gamma_2 \sin \theta_2 (\dot{\phi}_2 \sin \gamma_2 \sin \theta_2) - \dot{\gamma}_1 \cos \theta_1 - \dot{\gamma}_2 \cos \theta_2 \right] |1\rangle \langle 2 | \quad (1) \]
\[ + i\hbar \left[ -\dot{\gamma}_1 \sin \gamma_1 (\dot{\phi}_1 \cos \gamma_1) - \dot{\gamma}_2 \sin \gamma_2 (\dot{\phi}_2 \cos \gamma_2) - \dot{\gamma}_1 \sin \theta_1 \cos \phi_1 \right. \]
\[ - \dot{\gamma}_2 \sin \theta_2 \cos \phi_2 + \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\cos \gamma_1 \sin \phi_1 + \sin \gamma_1 \cos \theta_1 \cos \phi_1) + \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\cos \gamma_2 \sin \phi_2 - \sin \gamma_2 \cos \theta_2 \cos \phi_2) - \dot{\gamma}_1 \sin \theta_1 \cos \phi_1 \]
\[ - i\hbar \left[ -\dot{\theta}_1 \sin \gamma_1 (\dot{\phi}_1 \cos \gamma_1) + \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\cos \gamma_1 \cos \phi_1 - \sin \gamma_1 \cos \theta_1 \sin \phi_1) - \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\cos \gamma_2 \cos \phi_2 + \sin \gamma_2 \cos \theta_2 \cos \phi_2) - \dot{\gamma}_1 \sin \theta_1 \sin \phi_1 \right. \]
\[ + \dot{\gamma}_2 \sin \theta_2 \sin \phi_2 - \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\cos \gamma_1 \cos \phi_1 - \sin \gamma_1 \cos \theta_1 \sin \phi_1) + \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\cos \gamma_2 \cos \phi_2 + \sin \gamma_2 \cos \theta_2 \cos \phi_2) \right] |1\rangle \langle 2 | \quad (2) \]
\[ + i\hbar \left[ -\dot{\theta}_1 \sin \gamma_1 (\sin \gamma_1 \sin \phi_1 - \cos \gamma_1 \cos \theta_1 \sin \phi_1) - \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\sin \gamma_1 \cos \phi_1 - \cos \gamma_1 \cos \theta_1 \cos \phi_1) + \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\sin \gamma_2 \cos \phi_2 - \cos \gamma_2 \sin \theta_2 \cos \phi_2) - \dot{\gamma}_1 \sin \theta_1 \sin \phi_1 \right. \]
\[ - \dot{\gamma}_2 \sin \theta_2 \cos \phi_2 + \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\cos \gamma_1 \cos \phi_1 - \sin \gamma_1 \cos \theta_1 \sin \phi_1) + \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\cos \gamma_2 \cos \phi_2 + \sin \gamma_2 \cos \theta_2 \cos \phi_2) - \dot{\gamma}_1 \sin \theta_1 \sin \phi_1 \right] \right] |1\rangle \langle 2 | \quad (3) \]
\[ + i\hbar \left[ -\dot{\theta}_1 \sin \gamma_1 (\sin \gamma_1 \sin \phi_1 - \cos \gamma_1 \cos \theta_1 \sin \phi_1) - \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\cos \gamma_1 \cos \phi_1 - \sin \gamma_1 \cos \theta_1 \cos \phi_1) + \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\cos \gamma_2 \cos \phi_2 + \sin \gamma_2 \cos \theta_2 \cos \phi_2) - \dot{\gamma}_1 \sin \theta_1 \sin \phi_1 \right. \]
\[ - \dot{\gamma}_2 \sin \theta_2 \cos \phi_2 + \dot{\phi}_1 \sin \gamma_1 \sin \theta_1 (\cos \gamma_1 \cos \phi_1 - \sin \gamma_1 \cos \theta_1 \sin \phi_1) + \dot{\phi}_2 \sin \gamma_2 \sin \theta_2 (\cos \gamma_2 \cos \phi_2 + \sin \gamma_2 \cos \theta_2 \cos \phi_2) - \dot{\gamma}_1 \sin \theta_1 \sin \phi_1 \right] \right] |1\rangle \langle 2 | \quad (4) \]
\[ = i\hbar \left[ \sin \gamma_1 \sin \theta_1 (\dot{\phi}_1 \cos \gamma_1 - \dot{\phi}_1 \sin \gamma_1 \sin \theta_1) - \sin \gamma_2 \sin \theta_2 (\dot{\phi}_2 \cos \gamma_2 - \dot{\phi}_2 \sin \gamma_2 \sin \theta_2) - \dot{\gamma}_1 \cos \theta_1 - \dot{\gamma}_2 \cos \theta_2 \right] |3\rangle \langle 4 | \quad (B1) \]

The time-dependent evolution operator parameterized by the generalized spherical angles in Eq. (10) is

\[ U_r(t) = \langle \cos \gamma_1 \cos \phi_1 - \sin \gamma_1 \sin \gamma_2 \sin \theta_1 \sin \theta_2 \sin \phi_1 + \cos \gamma_1 \sin \gamma_2 \sin \theta_1 \sin \theta_2 \sin \phi_2 \rangle \] 1 \rangle \langle 1 \rangle,
\[ \langle \sin \gamma_2 \sin \theta_1 \sin \theta_2 \sin \phi_1 + \cos \gamma_1 \sin \gamma_2 \sin \theta_1 \sin \theta_2 \sin \phi_2 \rangle \] 2 \rangle \langle 1 \rangle,
\[ \langle \sin \gamma_2 \sin \gamma_1 (\sin \theta_1 \sin \theta_2 \sin \phi_1 - \cos \gamma_1 \sin \theta_2 \sin \phi_2) + \sin \theta_2 \sin \gamma_1 \cos \gamma_2 \sin \phi_2 + \sin \theta_2 \sin \gamma_1 \cos \gamma_2 \cos \phi_2 \rangle \] 3 \rangle \langle 1 \rangle,
\[ \langle \sin \gamma_2 \sin \gamma_1 (\sin \theta_1 \sin \theta_2 \sin \phi_1 - \cos \gamma_1 \sin \theta_2 \sin \phi_2) + \sin \theta_2 \sin \gamma_1 \cos \gamma_2 \cos \phi_2 \rangle \] 4 \rangle \langle 1 \rangle,
\[ \langle -\sin \gamma_2 \sin \gamma_1 \sin \phi_1 - \cos \gamma_1 \sin \gamma_2 \sin \phi_2 \rangle \] 1 \rangle \langle 2 \rangle,
\[ \langle \sin \gamma_1 \sin \gamma_2 \sin \theta_2 \sin \phi_1 - \cos \gamma_1 \sin \gamma_2 \sin \theta_2 \sin \phi_2 \rangle \] 2 \rangle \langle 2 \rangle,
\[ \langle \sin \theta_1 \sin \gamma_1 \cos \gamma_2 \sin \phi_1 - \sin \gamma_2 \sin \gamma_1 \cos \gamma_2 \sin \phi_2 \rangle \] 1 \rangle \langle 3 \rangle,
\[ \langle \sin \theta_2 \sin \gamma_1 \cos \gamma_2 \sin \phi_2 - \sin \gamma_2 \sin \gamma_1 \cos \gamma_2 \sin \phi_1 \rangle \] 2 \rangle \langle 3 \rangle,
\[ \langle \sin \gamma_1 \sin \gamma_2 \cos \phi_1 - \sin \gamma_2 \sin \gamma_1 \cos \phi_2 \rangle \] 3 \rangle \langle 3 \rangle,
\[ \langle \cos \theta_1 \sin \gamma_1 \cos \gamma_2 - \sin \gamma_2 \sin \theta_1 \sin \theta_2 \sin \phi_1 + \cos \theta_2 \sin \gamma_1 \rangle \] 4 \rangle \langle 3 \rangle,
\[ \langle \sin \theta_1 \sin \gamma_1 (-\cos \gamma_2) \sin \phi_1 - \sin \gamma_2 \sin \gamma_1 \sin \gamma_2 \sin \phi_2 \rangle \] 4 \rangle \langle 4 \rangle,
\[ \langle \sin \theta_1 \sin \gamma_1 \cos \gamma_2 \sin \phi_1 - \sin \gamma_2 \sin \gamma_1 \cos \gamma_2 \sin \phi_2 \rangle \] 3 \rangle \langle 3 \rangle,
\[ \langle \cos \theta_2 \cos \gamma_1 \cos \phi_2 - \sin \gamma_2 \sin \gamma_1 \sin \gamma_2 \sin \phi_1 - \cos \theta_2 \sin \gamma_1 \rangle \] 2 \rangle \langle 4 \rangle,
\[ \langle \cos \theta_2 \cos \gamma_1 \cos \phi_2 - \sin \gamma_2 \sin \gamma_1 \sin \gamma_2 \sin \phi_1 - \cos \theta_2 \sin \gamma_1 \rangle \] 2 \rangle \langle 4 \rangle,
\[ \langle \sin \gamma_1 \sin \gamma_2 \sin \phi_1 + \cos \gamma_1 \cos \phi_2 \rangle \] 4 \rangle \langle 4 \rangle.

(B2)

Appendix C: Connection with quantum optics (diamond configuration)

To relate the Hamiltonian of the inverse engineering approach, Eq. (9), 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
\[
H(t) = \hbar \left\{ \tilde{\Omega}_{12}(t) [\ket{1}ra{2} + \ket{2}ra{1}] \cos (\omega_{12} t + \phi_{12}) - \tilde{\Omega}'_{12}(t) [\ket{1}ra{2} + \ket{2}ra{1}] \sin (\omega_{12} t + \phi_{12}) + \hbar \right. \\
+ \tilde{\Omega}_{13}(t) [\ket{1}ra{3} + \ket{3}ra{1}] \cos (\omega_{13} t + \phi_{13}) - \tilde{\Omega}'_{13}(t) [\ket{1}ra{3} + \ket{3}ra{1}] \sin (\omega_{13} t + \phi_{13}) \\
+ \tilde{\Omega}_{24}(t) [\ket{2}ra{4} + \ket{4}ra{2}] \cos (\omega_{24} t + \phi_{24}) - \tilde{\Omega}'_{24}(t) [\ket{2}ra{4} + \ket{4}ra{2}] \sin (\omega_{24} t + \phi_{24}) \\
+ \tilde{\Omega}_{34}(t) [\ket{3}ra{4} + \ket{4}\bra{3}] \cos (\omega_{34} t + \phi_{34}) - \tilde{\Omega}'_{34}(t) [\ket{3}\bra{4} + \ket{4}\bra{3}] \sin (\omega_{34} t + \phi_{34}) \\
+ \sum_{i=2}^{4} \omega_{i} |i\rangle\langle i| \right\},
\]  

where we use the vector basis \(|1\rangle = \left( \begin{array}{c} 1 \\ 0 \\ 0 \\ 0 \end{array} \right)\), \(|2\rangle = \left( \begin{array}{c} 0 \\ 1 \\ 0 \\ 0 \end{array} \right)\), \(|3\rangle = \left( \begin{array}{c} 0 \\ 0 \\ 1 \\ 0 \end{array} \right)\), \(|4\rangle = \left( \begin{array}{c} 0 \\ 0 \\ 0 \\ 1 \end{array} \right)\). \(\tilde{\Omega}_{ij}(t), \tilde{\Omega}'_{ij}(t)\) are the atom-field coupling strengths (Rabi frequencies), assumed real for simplicity, and \(\phi_{ij}\) the phases of the coherent driving fields. The atomic levels \(|i\rangle\) have energies \(\hbar \omega_{i}\) and the fields have angular frequencies \(\omega_{ij}\). 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

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

Using

\[
H_{I}(t) = U_{0}(t)H(t)U_{0}^{\dagger}(t) + i\hbar U_{0}(t)U_{0}^{\dagger}(t),
\]
and imposing the four-photon resonance condition [48]– the Hamiltonian in the interacting picture is

\[
H_{I}(t) = \hbar \left\{ 2(\omega_{2} - \omega_{12})|2\rangle\langle 2| + 2(\omega_{3} - \omega_{13})|3\rangle\langle 3| + 2(\omega_{4} - \omega_{12} - \omega_{24})|4\rangle\langle 4| \right. \\
+ \tilde{\Omega}_{12}(t) \left( 1 + e^{-2i(\omega_{12} t + \phi_{12})} \right) |1\rangle\langle 2| + \left( 1 + e^{2i(\omega_{12} t + \phi_{12})} \right) |2\rangle\langle 1| \\
+ i\tilde{\Omega}_{12}(t) \left( 1 - e^{-2i(\omega_{12} t + \phi_{12})} \right) |1\rangle\langle 2| - \left( 1 - e^{2i(\omega_{12} t + \phi_{12})} \right) |2\rangle\langle 1| \\
+ \tilde{\Omega}_{13}(t) \left( 1 + e^{-2i(\omega_{13} t + \phi_{13})} \right) |1\rangle\langle 3| + \left( 1 + e^{2i(\omega_{13} t + \phi_{13})} \right) |3\rangle\langle 1| \\
+ i\tilde{\Omega}_{13}(t) \left( 1 - e^{-2i(\omega_{13} t + \phi_{13})} \right) |1\rangle\langle 3| - \left( 1 - e^{2i(\omega_{13} t + \phi_{13})} \right) |3\rangle\langle 1| \\
+ \tilde{\Omega}_{24}(t) \left( 1 + e^{-2i(\omega_{24} t + \phi_{24})} \right) |2\rangle\langle 4| + \left( 1 + e^{2i(\omega_{24} t + \phi_{24})} \right) |4\rangle\langle 2| \\
+ i\tilde{\Omega}_{24}(t) \left( 1 - e^{-2i(\omega_{24} t + \phi_{24})} \right) |2\rangle\langle 4| - \left( 1 - e^{2i(\omega_{24} t + \phi_{24})} \right) |4\rangle\langle 2| \\
+ \tilde{\Omega}_{34}(t) \left( 1 + e^{-2i(\omega_{34} t + \phi_{34})} \right)e^{-i\Phi} |3\rangle\langle 4| + \left( 1 + e^{2i(\omega_{34} t + \phi_{34})} \right)e^{i\Phi} |4\rangle\langle 3| \\
+ i\tilde{\Omega}_{34}(t) \left( 1 - e^{-2i(\omega_{34} t + \phi_{34})} \right)e^{-i\Phi} |3\rangle\langle 4| - \left( 1 - e^{2i(\omega_{34} t + \phi_{34})} \right)e^{i\Phi} |4\rangle\langle 3| \right\}.
\]

where

\[\Phi = \phi_{12} - \phi_{13} + \phi_{24} - \phi_{34}.\]  

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

\[ H_{1, RW A}(t) = \frac{\hbar}{2} \left( \begin{array}{ccc}
0 & \tilde{\Omega}_{12}(t) + i\tilde{\Omega}'_{12}(t) & \tilde{\Omega}_{13}(t) + i\tilde{\Omega}'_{13}(t) & \tilde{\Omega}_{14}(t) + i\tilde{\Omega}'_{14}(t) \\
\bar{\Delta}_2 & 0 & \tilde{\Delta}_3 \\
0 & \tilde{\Delta}_3 & 0 \\
\tilde{\Omega}_{24}(t) & -i\tilde{\Omega}'_{24}(t) & (\tilde{\Omega}_{24}(t) + i\tilde{\Omega}'_{24}(t))e^{i\phi} & \tilde{\Delta}_4
\end{array} \right), \]  

(C7)

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

\[
\tilde{\Delta}_2 = 2(\omega_2 - \omega_{12}), \\
\tilde{\Delta}_3 = 2(\omega_3 - \omega_{13}), \\
\tilde{\Delta}_4 = 2(\omega_4 - \omega_{12} - \omega_{24}).
\]

(C8)

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

\[
\phi_{12} - \phi_{13} + \phi_{24} - \phi_{34} = 0,
\]

(C9)

the Hamiltonian in Eq. (C7) has the structure of the one in Eq. (4).

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, \(-\Delta_i = \Delta_i/2\), the laser (angular) frequencies are

\[
\omega_{12} = \omega_2 - \frac{\epsilon_2' - \epsilon_2}{2T},
\]

\[
\omega_{13} = \omega_3 - \frac{\epsilon_3' - \epsilon_3}{2T},
\]

\[
\omega_{24} = \omega_4 - \omega_2 + \frac{\epsilon_4' - \epsilon_2}{2T} - \frac{\epsilon_4' - \epsilon_4}{2T},
\]

(C10)

and, to satisfy the four-photon resonance condition,

\[
\omega_{34} = \omega_4 - \omega_3 - \frac{\epsilon_4' - \epsilon_4}{2T} + \frac{\epsilon_3' - \epsilon_3}{2T}.
\]

(C11)

Comparing the non-diagonal terms in Eqs. (C7) and (9) we find the form of the Rabi frequencies,

\[
\tilde{\Omega}_{jk} = 2e^{i(\phi_j - \phi_k)} \tilde{\Omega}_{jk},
\]

(C12)

with \(\phi_k = 0, \ \phi_k (k = 2, 3, 4)\) given by Eqs. (21,22), 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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