A New Control Scheme of Multilevel Quantum System Based on Effective Decomposition by Intense CW Lasers†

M. Sugawara,* M. Tamaki, and S. Yabushita

Department of Fundamental Science and Technology, Graduate School of Science and Technology, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223-8522, Japan

Received: April 28, 2007; In Final Form: July 10, 2007

We propose a new scheme for quantum dynamics control of multilevel system using intense lasers. To do so, we apply intense CW lasers to create a strongly coupled subsystem with which one can make the complementary space effectively isolated, and we apply the established control schemes to the isolated subsystem. We have also obtained an effective Hamiltonian for the target subsystem with the help of the second-order perturbation theory. Numerical demonstrations on model systems show that the present decomposition scheme effectively works for population dynamics control. It is also found that relaxation processes can be suppressed under the proposed scheme.

1. Introduction

Quantum dynamics control using lasers has recently gathered great interest as it covers wide-ranging control targets from molecular motions, electronic transitions to quantum device control, and so on.[1−34] One of the most basic quantum control scheme is the one focusing on quantum level population dynamics. For example, a π-pulse scheme works efficiently on 100% population inversion in a simple two-level system,[35,36] while stimulated Raman adiabatic passage (STIRAP) has been successfully applied to the population transfer in the Λ-type three-level system.[16−18] There are also quantum control schemes proposed for specific multilevel systems, which consist of more than three levels.[19−28] These schemes depend on the system Hamiltonian or the analytical expressions of eigenvalues and eigenvectors as functions of laser parameters. However, it is often difficult to obtain analytical solutions for general multilevel systems consisting of more than four levels. On the other hand, there is an optimal control theory (OCT) that can be applied to arbitrary control problems including multilevel quantum system dynamics.[29−34] However, control fields designed by the OCT tend to show very complicated time dependence, which makes it difficult to realize experimentally. This happens because we need to consider all the levels with equal importance in the global laser optimization. It would make the laser designing process simple and easy if one could pick up several levels of interest and confine the population dynamics within those levels. In this study, we aim to propose a general scheme for controlling multilevel systems from such a point of view. First, we focus on effectively decomposing a multilevel quantum system into compact subsystems, which consist of a small number of levels, by irradiating intense CW laser fields. Next, we apply well-established control schemes onto the isolated subsystem. We also show that the decomposition scheme can be utilized for the population dynamics control of several variations of multilevel systems by numerical demonstrations.

† Part of the "Sheng Hsien Lin Festschrift".

* Corresponding author. E-mail: michi@chem.keio.ac.jp.

2. Theoretical

We consider a general multilevel system as shown in Figure 1. We split the total system in two subspaces, which we call A- and B-spaces, consisting of N states $\{|A_1\rangle,|A_2\rangle,....,|A_N\rangle\}$ and M states $\{|B_1\rangle,|B_2\rangle,....,|B_M\rangle\}$, respectively. We define the A-space so that there exist no direct optical transitions allowed between the states. On the other hand, the B-space consists of the states that can be strongly coupled with each other through optical interaction Ω. Hereafter, we consider the case in which optical interactions between A and B-spaces (V_1,V_2, ...) are weak enough to be treated by the perturbation theory. We introduce common detuning parameter Δ for the transitions between $|A_i\rangle$ and $|B_j\rangle$. Our main objective is to effectively decompose the total system into A- and B-spaces and create an isolated subsystem in the A-space in which we introduce indirect optical transitions for quantum dynamics control.

To treat laser fields quantum mechanically, we adopt the dressed state picture.[37] The dressed Hamiltonian matrix for the multilevel system in Figure 1 is given as

$$\mathbf{H} = \begin{pmatrix} \mathbf{H}_A & \mathbf{V}_{AB} \\ \mathbf{V}_{BA} & \mathbf{H}_B \end{pmatrix} \tag{1}$$

Here, $\mathbf{H}_A$ and $\mathbf{H}_B$ are $N \times N$ and $M \times M$ square Hamiltonian matrices for A- and B-spaces and $\mathbf{V}_{AB}(= \mathbf{V}_{BA}^T)$ is a $M \times N$ ($N \times M$) matrix. Because there are no direct optical interactions within the A-space, $\mathbf{H}_A$ is a diagonal matrix given as

$$\mathbf{H}_A = \begin{pmatrix} \Delta & & 0 \\ & \Delta & \\ 0 & & \ddots & \\ & & & \Delta \end{pmatrix} \equiv \Delta \mathbf{I}_A \tag{2}$$

where $\mathbf{I}_A$ is the $N \times N$ unit matrix. The B-space Hamiltonian, $\mathbf{H}_B$, is given as

$$\mathbf{H}_B = \begin{pmatrix} -i\Gamma_1 & \Omega_{12} & \cdots & \Omega_{1M} \\ \Omega_{21} & -i\Gamma_2 & & \Omega_{2M} \\ \vdots & & \ddots & \vdots \\ \Omega_{M1} & \Omega_{M2} & \cdots & -i\Gamma_M \end{pmatrix} \tag{3}$$

10.1021/jp073268m CCC: $37.00
Published on Web 08/25/2007

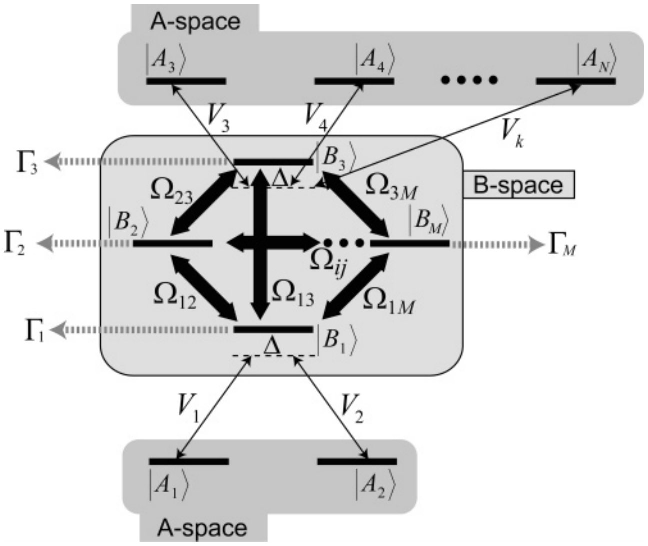

Figure 1. Schematic diagram of a general multilevel quantum system. There are no direct optical transition path between the states $\{|A_i\rangle\}$ in A-space, while states in B-space $\{|B_j\rangle\}$ can be closely coupled with each other via strong optical interactions Ω_{ij}. $\{|A_i\rangle\}$ and $\{|B_j\rangle\}$ are coupled with each other through the weak interactions, V_k.

where Ω_{ij} denotes the optical interaction between the states within the B-space and Γ_i is the population decay constant due to the relaxation process associated to the state $|B_i\rangle$.

Now, we consider the Schrödinger equation in a matrix-vector representation

$$\mathbf{H}\cdot\mathbf{c}_i = \varepsilon_i \mathbf{c}_i \qquad (i = 1, ..., N + M) \tag{4}$$

where ϵ_i and $\mathbf{c}_i$ are the ith eigenvalue and its corresponding eigenvector. One can rewrite eq 4 in a matrix form as

$$\mathbf{H}\cdot\mathbf{U} = \mathbf{U}\cdot\mathbf{E} \tag{5}$$

where

$$\mathbf{E} \equiv \begin{pmatrix} \varepsilon_1 & & 0 \\ & \varepsilon_2 & \\ 0 & & \ddots \\ & & & \varepsilon_{N+M} \end{pmatrix}, \mathbf{U} \equiv (\mathbf{c}_1, \mathbf{c}_2, \cdots, \mathbf{c}_{N+M}) \tag{6}$$

Here, we redefine $\mathbf{E}$ and $\mathbf{U}$ using minor matrices as

$$\mathbf{E} = \begin{pmatrix} \mathbf{E}_A & \mathbf{0} \\ \mathbf{0} & \mathbf{E}_B \end{pmatrix}, \mathbf{U} = \begin{pmatrix} \mathbf{U}_{AA} & \mathbf{U}_{AB} \\ \mathbf{U}_{BA} & \mathbf{U}_{BB} \end{pmatrix} \tag{7}$$

where $\mathbf{E}_A$ and $\mathbf{E}_B$ are $N \times N$ and $M \times M$ diagonal matrices given as

$$\mathbf{E}_A = \begin{pmatrix} \varepsilon_1 & 0 & 0 \\ 0 & \ddots & 0 \\ 0 & 0 & \varepsilon_N \end{pmatrix}, \mathbf{E}_B = \begin{pmatrix} \varepsilon_{N+1} & 0 & 0 \\ 0 & \ddots & 0 \\ 0 & 0 & \varepsilon_{N+M} \end{pmatrix} \tag{8}$$

while $\mathbf{U}_{AA}$ and $\mathbf{U}_{BB}$ are $N \times N$ and $M \times M$ matrices, respectively. Note that $\mathbf{U}_{BA}$ ($\mathbf{U}_{AB}$) denotes a mixing between A- and B-spaces through the weak optical interactions (V_1,V_2, ...). To apply the perturbation theory, we write the total Hamiltonian as

$$\mathbf{H} = \mathbf{H}_0 + \lambda\mathbf{V} \tag{9}$$

where

$$\mathbf{H}_0 = \begin{pmatrix} \Delta\mathbf{I}_A & \mathbf{0} \\ \mathbf{0} & \mathbf{H}_B \end{pmatrix}, \mathbf{V} = \begin{pmatrix} \mathbf{0} & \mathbf{V}_{AB} \\ \mathbf{V}_{BA} & \mathbf{0} \end{pmatrix} \tag{10}$$

Here, λ is a dimensionless parameter introduced to clarify the perturbation order. We expand $\mathbf{E}$ and $\mathbf{U}$ in eq 5 as

$$\mathbf{E} = \mathbf{E}^{(0)} + \lambda\mathbf{E}^{(1)} + \lambda^2\mathbf{E}^{(2)} + \cdots = \sum_{n=0} \lambda^n \mathbf{E}^{(n)} \tag{11}$$

$$\mathbf{U} = \mathbf{U}^{(0)} + \lambda\mathbf{U}^{(1)} + \lambda^2\mathbf{U}^{(2)} + \cdots = \sum_{n=0} \lambda^n \mathbf{U}^{(n)} \tag{12}$$

We define $\mathbf{E}^{(n)}$ and $\mathbf{U}^{(n)}$ as

$$\mathbf{E}^{(n)} = \begin{pmatrix} \mathbf{E}_A^{(n)} & \mathbf{0} \\ \mathbf{0} & \mathbf{E}_B^{(n)} \end{pmatrix}, \mathbf{U}^{(n)} = \begin{pmatrix} \mathbf{U}_{AA}^{(n)} & \mathbf{U}_{AB}^{(n)} \\ \mathbf{U}_{BA}^{(n)} & \mathbf{U}_{BB}^{(n)} \end{pmatrix} \tag{13}$$

Each component is expanded accordingly as

$$\mathbf{E}_I = \mathbf{E}_I^{(0)} + \lambda\mathbf{E}_I^{(1)} + \lambda^2\mathbf{E}_I^{(2)} + \cdots = \sum_{n=0} \lambda^n \mathbf{E}_I^{(n)} \tag{14}$$

$$\mathbf{U}_{IJ} = \mathbf{U}_{IJ}^{(0)} + \lambda\mathbf{U}_{IJ}^{(1)} + \lambda^2\mathbf{U}_{IJ}^{(2)} + \cdots = \sum_{n=0} \lambda^n \mathbf{U}_{IJ}^{(n)} \tag{15}$$

where $I,J = \{A,B\}$. Inserting eqs 11 and 12 into eq 5 and comparing both sides of the equation up to the second order of λ gives following formula:

$$\lambda^0\text{:}\ \mathbf{H}_0\cdot\mathbf{U}^{(0)} = \mathbf{U}^{(0)}\cdot\mathbf{E}^{(0)} \tag{16}$$

$$\lambda^1\text{:}\ \mathbf{H}_0\cdot\mathbf{U}^{(1)} - \mathbf{U}^{(1)}\cdot\mathbf{E}^{(0)} + \mathbf{V}\cdot\mathbf{U}^{(0)} = \mathbf{U}^{(0)}\cdot\mathbf{E}^{(1)} \tag{17}$$

$$\lambda^2\text{:}\ \mathbf{H}_0\cdot\mathbf{U}^{(2)} - \mathbf{U}^{(2)}\cdot\mathbf{E}^{(0)} - \mathbf{U}^{(1)}\cdot\mathbf{E}^{(1)} + \mathbf{V}\cdot\mathbf{U}^{(1)} = \mathbf{U}^{(0)}\cdot\mathbf{E}^{(2)} \tag{18}$$

First, we consider eq 16. Inserting eqs 10 and 13 into eq 16 gives

$$\Delta\mathbf{I}_A\cdot\mathbf{U}_{AA}^{(0)} = \mathbf{U}_{AA}^{(0)}\cdot\mathbf{E}_A^{(0)} \tag{19}$$

$$\mathbf{H}_B\cdot\mathbf{U}_{BB}^{(0)} = \mathbf{U}_{BB}^{(0)}\cdot\mathbf{E}_B^{(0)} \tag{20}$$

Notice that $\mathbf{U}_{AA}^{(0)}$ cannot be determined uniquely by eq 19. Because $\mathbf{I}_A$ is a unit matrix, arbitrary vector in the A-space $\mathbf{c}_A$ can satisfy $\mathbf{H}_A\cdot\mathbf{c}_A = \Delta\mathbf{c}_A$, thus we have $\mathbf{E}_A^{(0)} = \Delta\mathbf{I}_A$. On the other hand, one can obtain the zeroth-order states, $\mathbf{U}_{BB}^{(0)}$, and corresponding eigenvalues, $\mathbf{E}_B^{(0)}$, by solving eq 20.

Next, we consider the first-order corrections onto eigenvalues, $\mathbf{E}_A^{(1)}$ and $\mathbf{E}_B^{(1)}$. One can rewrite eq 17 explicitly in a matrix representation as

$$\begin{pmatrix} \Delta\mathbf{I}_A\cdot\mathbf{U}_{AA}^{(1)} - \mathbf{U}_{AA}^{(1)}\cdot\mathbf{E}_A^{(0)} & \Delta\mathbf{I}_A\cdot\mathbf{U}_{AB}^{(1)} - \mathbf{U}_{AB}^{(1)}\cdot\mathbf{E}_B^{(0)} + \mathbf{V}_{AB}\cdot\mathbf{U}_{BB}^{(0)} \\ \mathbf{H}_B\cdot\mathbf{U}_{BA}^{(1)} - \mathbf{U}_{BA}^{(1)}\cdot\mathbf{E}_A^{(0)} + \mathbf{V}_{BA}\cdot\mathbf{U}_{AA}^{(0)} & \mathbf{H}_B\cdot\mathbf{U}_{BB}^{(1)} - \mathbf{U}_{BB}^{(1)}\cdot\mathbf{E}_B^{(0)} \end{pmatrix} = \begin{pmatrix} \mathbf{U}_{AA}^{(0)}\cdot\mathbf{E}_A^{(1)} & \mathbf{0} \\ \mathbf{0} & \mathbf{U}_{BB}^{(0)}\cdot\mathbf{E}_B^{(1)} \end{pmatrix} \tag{21}$$

Comparing both sides of eq 21 together with the relations, $\mathbf{E}_A^{(0)} = \Delta\mathbf{I}_A$, $\mathbf{U}_{BA}^{(1)}\cdot\mathbf{E}_A^{(0)} = \mathbf{U}_{BA}^{(1)}\cdot(\Delta\mathbf{I}_A) = (\Delta\mathbf{I}_B)\cdot\mathbf{U}_{BA}^{(1)}$, $(\Delta\mathbf{I}_A)\cdot\mathbf{U}_{AB}^{(1)} = \mathbf{U}_{AB}^{(1)}\cdot(\Delta\mathbf{I}_B)$, gives the following:

$$\mathbf{U}_{AA}^{(0)}\cdot\mathbf{E}_{A}^{(1)} = \mathbf{0} \tag{22}$$

$$\mathbf{U}_{AA}^{(0)}\cdot(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)}) + \mathbf{V}_{AB}\cdot\mathbf{U}_{BB}^{(0)} = \mathbf{0} \tag{23}$$

$$(\mathbf{H}_B - \Delta\mathbf{I}_B)\cdot\mathbf{U}_{BA}^{(1)} + \mathbf{V}_{BA}\cdot\mathbf{U}_{AA}^{(0)} = \mathbf{0} \tag{24}$$

$$\mathbf{H}^{B}\cdot\mathbf{U}_{BB}^{(1)} - \mathbf{U}_{\mathrm{BB}}^{(1)}\cdot\mathbf{E}_B^{(0)} = \mathbf{U}_{BB}^{(0)}\cdot\mathbf{E}_B^{(1)} \tag{25}$$

From eq 22 and $\mathbf{U}_{AA}^{(0)} \neq \mathbf{0}$, one obtains $\mathbf{E}_A^{(1)} = \mathbf{0}$, which indicates that there are no first-order energy corrections in the A-space. The first-order mixing between A and B-spaces, $\mathbf{U}_{AB}^{(1)}$ and $\mathbf{U}_{BA}^{(1)}$, are given from eqs 23 and 24 as

$$\mathbf{U}_{AB}^{(1)} = -\mathbf{V}_{AB}\cdot\mathbf{U}_{BB}^{(0)}\cdot(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)})^{-1} \tag{26}$$

$$\mathbf{U}_{BA}^{(1)} = (\Delta\mathbf{I}_B - \mathbf{H}_B)^{-1}\cdot\mathbf{V}_{BA}\cdot\mathbf{U}_{AA}^{(0)} = \mathbf{U}_{BB}^{(0)}\cdot(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)})^{-1}\cdot\mathbf{U}_{BB}^{(0)-1}\cdot\mathbf{V}_{BA}\cdot\mathbf{U}_{AA}^{(0)} \tag{27}$$

We use $\mathbf{E}_B^{(0)} = \mathbf{U}_{BB}^{(0)-1}\cdot\mathbf{H}_B\cdot\mathbf{U}_{BB}^{(0)}$ for deriving the last expression of eq 27. Note that the first-order mixing is dependent on $(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)})^{-1}$. Because both $\mathbf{E}_B^{(0)}$ and $\Delta\mathbf{I}_B$ are diagonal matrices, $(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)})^{-1}$ is also given as a diagonal matrix with the diagonal elements defined as $[(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)})^{-1}]_{ii} = 1/\{\Delta - (\mathbf{E}_B^{(0)})_{ii}\}$. Thus, the mixing becomes negligibly small as all the differences between $(\mathbf{E}_B^{(0)})_{ii}$ and Δ become large. Notice that this condition can be achieved by introducing large Ω_{if} in $\mathbf{H}_B$, or applying intense lasers to the B-space. Thus, one can effectively decompose the total multilevel system into effectively isolated A and B spaces with the condition, $\Omega_{if} \gg V_k$. One should note that the complicated B-space consisting of three or more levels could make it difficult to achieve the clean decomposition. This is because there could be the zeroth-order eigenstates in the B-space with the eigenvalue close to Δ, which causes significant mixing with the A-space.

Next, we consider the second-order corrections. Comparing each element of both sides of eq 18 together with $\Delta\mathbf{I}_A = \mathbf{E}_A^{(0)}$, $\mathbf{E}_A^{(1)} = \mathbf{0}$, one obtains the following:

$$\mathbf{V}_{AB}\cdot\mathbf{U}_{BA}^{(1)} = \mathbf{U}_{AA}^{(0)}\cdot\mathbf{E}_A^{(2)} \tag{28}$$

$$\mathbf{U}_{AB}^{(2)}\cdot(\Delta\mathbf{I}_B - \mathbf{E}_B^{(0)}) - \mathbf{U}_{AB}^{(1)}\cdot\mathbf{E}_B^{(1)} + \mathbf{V}_{AB}\cdot\mathbf{U}_{BB}^{(1)} = \mathbf{0} \tag{29}$$

$$(\mathbf{H}_B - \Delta\mathbf{I}_B)\cdot\mathbf{U}_{BA}^{(2)} + \mathbf{V}_{BA}\cdot\mathbf{U}_{AA}^{(1)} = \mathbf{0} \tag{30}$$

$$\mathbf{H}_B\cdot\mathbf{U}_{BB}^{(2)} - \mathbf{U}_{BB}^{(2)}\cdot\mathbf{E}_B^{(0)} - \mathbf{U}_{BB}^{(1)}\cdot\mathbf{E}_B^{(1)} + \mathbf{V}_{BA}\cdot\mathbf{U}_{AB}^{(1)} = \mathbf{0} \tag{31}$$

Because we are interested in the dynamics control of the A-space, we focus on the second-order correction $\mathbf{E}_A^{(2)}$. Inserting eq 27 into eq 28 gives

$$\mathbf{V}_{BA}\cdot\mathbf{U}_{BB}^{(2)} - \mathbf{U}_{BB}^{(2)}\cdot\mathbf{E}_B^{(0)} - \mathbf{U}_{BB}^{(1)}\cdot\mathbf{E}_B^{(1)} + \mathbf{V}_{BA}\cdot\mathbf{U}_{AB}^{(1)} = \mathbf{0} \tag{32}$$

Solving this eigenvalue problem gives the second-order correction, $\mathbf{E}_A^{(2)}$, together with the zeroth-order states in the A-space, $\mathbf{U}_{AA}^{(0)}$. Here, we introduce the effective Hamiltonian for the A-space defined as

$$\mathbf{H}^{(\mathrm{eff})} = \Delta\mathbf{I}_A + \mathbf{V}_{AB}\cdot(\Delta\mathbf{I}_B - \mathbf{H}_B)^{-1}\cdot\mathbf{V}_{BA} \tag{33}$$

It is readily seen that $\mathbf{U}_{AA}^{(0)}$ satisfies the effective Schrödinger equation with respect to the A-space,

$$\mathbf{H}^{(\mathrm{eff})}\cdot\mathbf{U}_{AA}^{(0)} = \mathbf{U}_{AA}^{(0)}\cdot\mathbf{E}^{(\mathrm{eff})} \tag{34}$$

where $\mathbf{E}^{(\mathrm{eff})}$ is a diagonal matrix defined as $(\mathbf{E}^{(\mathrm{eff})})_{ii} = \epsilon_i^{(\mathrm{eff})} = \Delta + \epsilon_i^{(2)}$ with $\epsilon_i^{(2)} = (\mathbf{E}_A^{(2)})_{ii}$.

Here, we discuss the relation between the present method and the reduction scheme utilizing the projection operator method.[3,4] The reduction scheme gives the formal operator $\tilde{M}$ that exactly describes the subspace dynamics of interest. First, one should note that the effective Hamiltonian, eq 33, is equivalent to the approximate $\tilde{M}$ given in ref 3. In the derivation, the frequency variable ω is replaced by the typical frequency, ω_0, of the subspace, which directly corresponds to the system setting that we take the common detuning parameter Δ in the A-space. The difference between this work and ref 3 is in the definition of subspaces. In ref 3, the reduction scheme is used to obtain the correction dynamics onto the strongly coupled subspace, which arises from the weak interspace couplings. On the contrary, in this study, we intend to produce the optically driven dynamics in the A-space in which there originally exist no direct optical transitions. In addition to that, one of the advantages of our formulation is that one can obtain higher-order corrections by continuing the expansion of eqs 11 and 12 if needed, while it is rather obscure to upgrade the approximation of $\tilde{M}$.

Here we emphasize again that the common detuning condition in the A-space, $\mathbf{H}_A = \Delta\mathbf{I}_A$, plays an important role in introducing the optical transitions in the A-space. Suppose we take different detuning for each transition within the A-space, $\mathbf{U}_{AA}^{(0)}$, is determined as the unit matrix, which fails to derive the effective Hamiltonian eq 33.

Finally, we briefly state how to obtain the dynamics in the isolated A-space using $\mathbf{H}^{(\mathrm{eff})}$. The time evolution of an arbitrary initial vector $\mathbf{d}(0)$ in the A-space under the decomposition condition is given as

$$\mathbf{d}(t) = \exp[-i\mathbf{H}^{(\mathrm{eff})}t/\hbar]\cdot\mathbf{d}(0) = \mathbf{U}_{AA}^{(0)}\cdot\exp[-i\mathbf{E}^{(\mathrm{eff})}t/\hbar]\cdot\mathbf{U}_{AA}^{(0)-1}\cdot\mathbf{d}(0) \tag{35}$$

Note that eq 34 is used in deriving the final expression. More specifically, the time-evolution of the ith component of the state vector, $d_i(t) \equiv (\mathbf{d}(t))_i$, is given as

$$d_i(t) = \sum_{jl} u_{if} \exp[-i\epsilon_j^{(\mathrm{eff})}t/\hbar]u_{jl}^{-1}d_l(0) \tag{36}$$

where $u_{if} \equiv (\mathbf{U}_{AA}^{(0)})_{ij}$ and $u_{jl}^{-1} \equiv (\mathbf{U}_{AA}^{(0)-1})_{ij}$. Thus, the population dynamics of the ith state in the A-space is given as

$$|d_i(t)|^2 = \left(\sum_{km} u_{ik} \exp[-i\epsilon_k^{(\mathrm{eff})}t/\hbar]u_{km}^{-1}d_m(0)\right)^* \left(\sum_{jl} u_{ij} \exp[-i\epsilon_j^{(\mathrm{eff})}t/\hbar]u_{jl}^{-1}d_l\right) = \sum_{kmjl} u_{ik}^* u_{ij}(u_{km}^{-1})^* u_{jl}^{-1} \exp[-i(\epsilon_j^{(\mathrm{eff})} - \epsilon_k^{(\mathrm{eff})})t/\hbar]d_m^*(0)d_l(0) \tag{37}$$

From eq 37, one can see that the energy difference, $\epsilon_j^{(\mathrm{eff})} - \epsilon_k^{(\mathrm{eff})}$, plays an important role in the population dynamics, and there is no dynamics invoked if $\epsilon_j^{(\mathrm{eff})} - \epsilon_k^{(\mathrm{eff})} = 0$ for all the combinations of j and k. Note that the energy differences originates from the second-order corrections, i.e., $\epsilon_j^{(\mathrm{eff})} - \epsilon_k^{(\mathrm{eff})} = \epsilon_j^{(2)} - \epsilon_k^{(2)}$. Thus, we need to take into account $\mathbf{E}_A^{(2)}$ in order to consider the population dynamics due to the indirect optical transitions in the A-space.

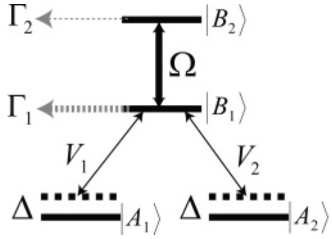

Figure 2. Schematic diagram of the branch-type four-level system. A-space consists of $|A_1\rangle$ and $|A_2\rangle$, while B-space consists of $|B_1\rangle$ and $|B_2\rangle$. There is a weak optical interaction V_1 (V_2) between $|A_1\rangle$ ($|A_2\rangle$) and $|B_1\rangle$, while $|B_1\rangle$ and $|B_2\rangle$ are strongly coupled through interaction Ω. Γ_1 (Γ_2) denotes a population decay constant associated with $|B_1\rangle$ ($|B_2\rangle$), and Δ is a detuning parameter that is commonly taken for the transitions, $|A_1\rangle \leftrightarrow |B_1\rangle$ and $|A_2\rangle \leftrightarrow |B_1\rangle$.

3. Applications to Multilevel Model Systems

Now, we apply the present decomposition scheme to various multilevel model systems. We first consider the branch-type four-level system shown in Figure 2. The A-space consists of $|A_1\rangle$ and $|A_2\rangle$, and there is no direct optical interaction between them, while $|B_1\rangle$ and $|B_2\rangle$ in the B-space can be strongly coupled through the interaction Ω. Note that only the $|B_1\rangle$ state is optically accessible from the two states in the A-space via interactions V_1 and V_2. The total Hamiltonian is given as

$$\mathbf{H}_{B4} = \begin{pmatrix} \mathbf{H}_A & \mathbf{V}_{AB} \\ \mathbf{V}_{BA} & \mathbf{H}_B \end{pmatrix} \tag{38}$$

where minor matrices are defined as

$$\mathbf{H}_A = \Delta \mathbf{I}_A = \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix}, \mathbf{H}_B = \begin{pmatrix} -i\Gamma_1 & \Omega \\ \Omega & -i\Gamma_2 \end{pmatrix} \tag{39}$$

$$\mathbf{V}_{AB} = \begin{pmatrix} V_1 & 0 \\ V_2 & 0 \end{pmatrix}, \mathbf{V}_{BA} = \begin{pmatrix} V_1 & V_2 \\ 0 & 0 \end{pmatrix} \tag{40}$$

Here, the basis set in the dressed state picture $\{|\tilde{A}_1\rangle,|\tilde{A}_2\rangle|\tilde{B}_1\rangle,|\tilde{B}_1\rangle\}$ is defined as

$$\begin{aligned} |\tilde{A}_1\rangle &\equiv |A_1\rangle \otimes |n+1\rangle|w+1\rangle|m\rangle \\ |\tilde{A}_2\rangle &\equiv |A_2\rangle \otimes |n\rangle|w+1\rangle|m+1\rangle \\ |\tilde{B}_1\rangle &\equiv |B_1\rangle \otimes |n\rangle|w+1\rangle|m\rangle \\ |\tilde{B}_2\rangle &\equiv |B_2\rangle \otimes |n\rangle|w\rangle|m+1\rangle \end{aligned} \tag{41}$$

where $|n\rangle$, $|w\rangle$, and $|m\rangle$ denote the number states of the laser fields relevant to the optical interactions V_1, Ω, and V_2, respectively. The first-order mixing of the B-space components into the A-space, $\mathbf{U}_{BA}^{(1)}$, is given from eq 27 as

$$\mathbf{U}_{BA}^{(1)} = \frac{\sqrt{{V_1}^2 + {V_2}^2}}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2}\begin{pmatrix} 0 & \Delta + i\Gamma_2 \\ 0 & \Omega \end{pmatrix} \tag{42}$$

From eq 42, one can see the decomposition condition, $\Omega \gg V_1,V_2$, makes $\mathbf{U}_{BA}^{(1)}$ negligibly small. We obtain the effective Hamiltonian $\mathbf{H}_{B4}^{(\text{eff})}$ from eq 33 as

$$\mathbf{H}_{B4}^{(\text{eff})} = \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix} + \frac{\Delta + i\Gamma_2}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2}\begin{pmatrix} {V_1}^2 & V_1V_2 \\ V_1V_2 & {V_2}^2 \end{pmatrix} \tag{43}$$

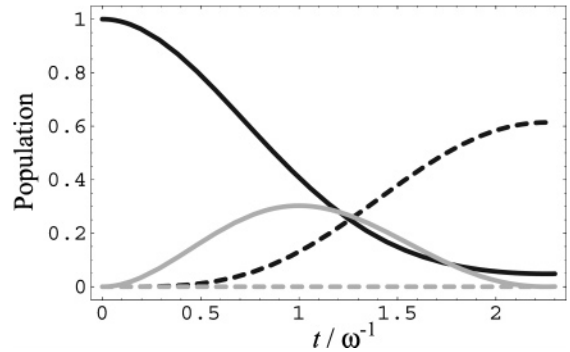

Figure 3. Population dynamics of the branch-type four-level system with $\Gamma_1 = 0.5$, $\Gamma_2 = 0$ under the laser condition, $V_1 = V_2 = 1$, $\Omega = 0$, $\Delta = 0$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

One obtains $\mathbf{U}_{AA}^{(0)}$ and its eigenvalues $\mathbf{E}^{(\text{eff})}$ by diagonalizing $\mathbf{H}_{B4}^{(\text{eff})}$ as

$$\mathbf{U}_{AA}^{(0)} = \frac{1}{\sqrt{{V_1}^2 + {V_2}^2}}\begin{pmatrix} -V_2 & V_1 \\ V_1 & V_2 \end{pmatrix} \tag{44}$$

and

$$\mathbf{E}^{(\text{eff})} = \mathbf{E}_A^{(0)} + \mathbf{E}_A^{(2)} = \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix} + \frac{\Delta + i\Gamma_2}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2}\begin{pmatrix} 0 & 0 \\ 0 & {V_1}^2 + {V_1}^2 \end{pmatrix} \tag{45}$$

Together with the condition $\Omega \gg \Delta,\Gamma_1,\Gamma_2$, $\mathbf{E}^{(\text{eff})}$ can be approximated to

$$\mathbf{E}^{(\text{eff})} \approx \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix} - \frac{\Delta + i\Gamma_2}{\Omega^2}\begin{pmatrix} 0 & 0 \\ 0 & {V_1}^2 + {V_2}^2 \end{pmatrix} = \begin{pmatrix} \varepsilon_1^{(\text{eff})} & 0 \\ 0 & \varepsilon_2^{(\text{eff})} \end{pmatrix} \tag{46}$$

The expression of eq 46 is highly suggestive. The coherent dynamics within the isolated A-space is invoked by the differences in the real part of the eigenvalues, $\text{Re}[\epsilon_1^{(\text{eff})} - \epsilon_2^{(\text{eff})}]$, as shown in eq 37. Note that this quantity is given as $\Delta({V_1}^2 + {V_2}^2)/\Omega^2$ in the present case. Because it is proportional to the detuning parameter Δ, the condition $\Delta \neq 0$ is required for introducing optically driven dynamics in the A-space. It is intriguing feature that the time scale of the invoked population dynamics can be controllable by changing the detuning parameter Δ. Note also that Γ_1 disappears in eq 46, which implies that one can suppress the relaxation process due to Γ_1 by introducing large Ω.

Now, we will see how we utilize the effective decomposition for control problems. Here, we aim at 100% population transfer from the initial state $|A_1\rangle$ to the target state $|A_2\rangle$. Because direct optical transition between $|A_1\rangle$ and $|A_2\rangle$ is prohibited, we need to utilize the intermediate state $|B_1\rangle$ that optically connects those two states. However, it suffers from an associated relaxation process characterized by the population decay constant, from which we take $\Gamma_1 = 0.5$. Here, all the parameter values for Ω,V_1, V_2, Δ, Γ_1, and Γ_2 are measured in the unit of $\hbar\omega$, where ω is the transition frequency of $|A_1\rangle \leftrightarrow |B_1\rangle$. Primitive laser settings, in which only $|B_1\rangle$ is used as an intermediate state, do not efficiently work because the population considerably escapes from the system. Shown in Figure 3 is the population dynamics under such a primitive laser condition, i.e., $V_1 = V_2 = 1$, $\Omega =$

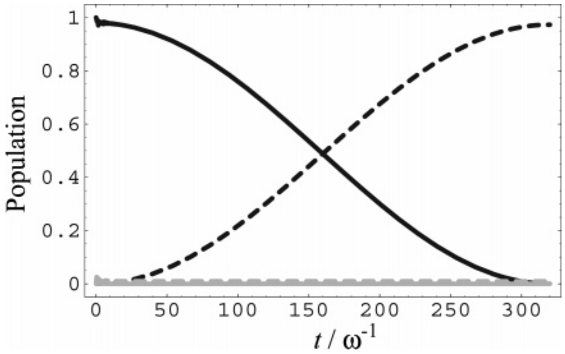

Figure 4. Population dynamics of the branch-type four-level system with $\Gamma_1 = 0.5$, $\Gamma_2 = 0$ under the laser condition, $V_1 = V_2 = 1$, $\Omega = 10$, $\Delta = 0.5$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

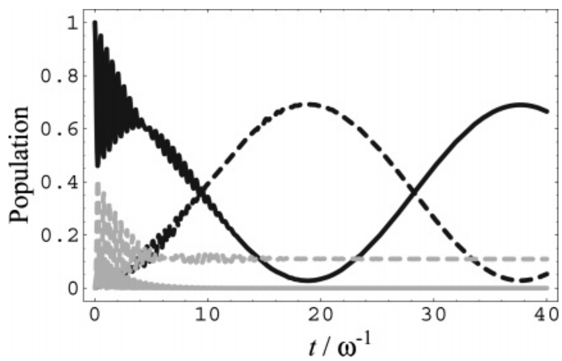

Figure 5. Population dynamics of the branch-type four-level system with $\Gamma_1 = 0.5$, $\Gamma_2 = 0$ under the laser condition, $V_1 = V_2 = 5$, $\Omega = 10$, $\Delta = 0.5$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

0, $\Delta = 0$. The final yield of the target state is only 60% because 33.8% of the population is lost through the relaxation process from $|B_1\rangle$. Now, we introduce the second intermediate state $|B_2\rangle$ with $\Gamma_2 = 0$. We consider utilizing $|B_2\rangle$ together with the decomposition condition $\Omega \gg V_1,V_2,\Delta,\Gamma_1,\Gamma_2$ in order to suppress the relaxation from $|B_1\rangle$. Shown in Figure 4 is the population dynamics under the laser parameters, $V_1 = V_2 = 1$, $\Omega = 10$, $\Delta = 0.5$. It is clearly seen that the population dynamics occurs in the A-space, without populating $|B_1\rangle$ or $|B_2\rangle$, which indicates that the A-space is effectively isolated as a weakly interacting two-level system. Its oscillatory population dynamics between $|A_1\rangle$ and $|A_1\rangle$ is characterized by well-known Rabi oscillation and its period T_R is given as $2\pi\Delta(V_1^2 + V_2^2)/\Omega^2$. Significant amount of the initial population can be transferred to $|A_1\rangle$ by terminating the laser at $t = T_R/2$, which corresponds to the π-pulse control scheme. It is shown in Figure 4 that 97.2% of the population is transferred to the target state $|A_1\rangle$ at $t = 314\omega^{-1}$. The total population loss is less than 2%. Thus, the population loss due to the relaxation process of $|B_1\rangle$ is drastically suppressed by applying the intense CW laser between $|B_1\rangle$ and $|B_1\rangle$. For comparison, we show the results for the parameter set that violates the condition satisfied in Figure 2. Shown in Figure 5 is the population dynamics with $V_1 = V_2 = 5$, $\Omega = 10$, $\Delta = 0.5$, in which the decomposition condition $\Omega \gg V_1,V_2$ is not satisfied. It is seen that the effective decomposition breaks down and the initial population on $|A_1\rangle$ escapes to the B-space. The population disappears through the dissipative state $|B_1\rangle$, while $|B_2\rangle$ is constantly populated. Shown in Figure 6 is the population dynamics under the condition, $V_1 = V_2 = 5$, $\Omega = 10$, $\Delta = 5$. The population dynamics occurs only in the A-space, which implies that the effective decomposition is well achieved with

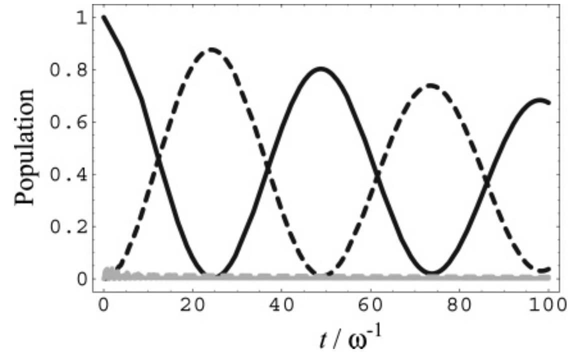

Figure 6. Population dynamics of the branch-type four-level system with $\Gamma_1 = 0.5$, $\Gamma_2 = 0$ under the laser condition, $V_1 = V_2 = 1$, $\Omega = 10$, $\Delta = 5$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

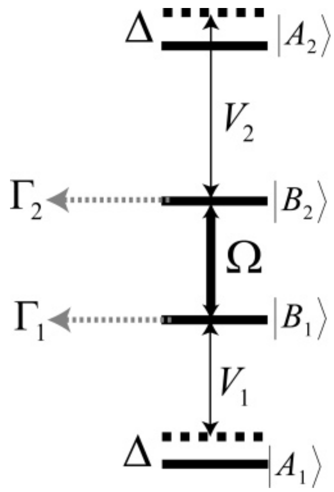

Figure 7. Schematic diagram of the ladder-type four-level system. A-space consists of $|A_1\rangle$ and $|A_2\rangle$, while B-space consists of $|B_1\rangle$ and $|B_2\rangle$. There are weak optical interactions V_1 and V_2 corresponding to the transitions $|A_1\rangle \leftrightarrow |B_1\rangle$ and $|A_2\rangle \leftrightarrow |B_2\rangle$, respectively, while $|B_1\rangle$ and $|B_2\rangle$ are strongly coupled through interaction Ω. Γ_1 (Γ_2) denotes a population decay constant associated with $|B_1\rangle$ ($|B_2\rangle$), and Δ is a detuning parameter that is commonly taken for the transitions $|A_1\rangle \leftrightarrow |B_1\rangle$ and $|A_2\rangle \leftrightarrow |B_2\rangle$.

the condition $\Omega \gg V_1,V_2$. However, the total population does not conserve because the suppression of the relaxation from $|B_1\rangle$ is insufficient. This is because Γ_1 in the eq 45 cannot be neglected because of the relatively large Δ.

Next, we consider the ladder-type four-level system shown in Figure 7. The initial state is taken to be $|A_1\rangle$, while we aim at 100% population transfer onto $|A_2\rangle$ via intermediate states $|B_1\rangle$ and $|B_2\rangle$, each of which possesses relaxation paths characterized by Γ_1 and Γ_2, respectively. The total Hamiltonian is given as

$$\mathbf{H}_{L4} = \begin{pmatrix} \mathbf{H}_A & \mathbf{V}_{AB} \\ \mathbf{V}_{BA} & \mathbf{H}_B \end{pmatrix} \quad (47)$$

where

$$\mathbf{H}_A = \Delta\mathbf{I}_A = \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix}, \mathbf{H}_B = \begin{pmatrix} -i\Gamma_1 & \Omega \\ \Omega & -i\Gamma_2 \end{pmatrix} \quad (48)$$

and

$$\mathbf{V}_{BA} = \begin{pmatrix} V_1 & 0 \\ 0 & V_2 \end{pmatrix}, \mathbf{V}_{AB} = \begin{pmatrix} V_1 & 0 \\ 0 & V_2 \end{pmatrix} \quad (49)$$

The first-order mixing is given from eq 27 as

$$\mathbf{U}_{BA}^{(1)} = \frac{V_1}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2} \begin{pmatrix} \frac{(\Delta + i\Gamma_2)M_{(+)} + 2V_2^{\,2}\Omega^2}{N_1} & \frac{(\Delta + i\Gamma_2)M_{(-)} + 2V_2^{\,2}\Omega^2}{N_2} \\ \frac{\{M_{(+)} + 2(\Delta + i\Gamma_2)V_2^{\,2}\}\Omega}{N_1} & \frac{\{M_{(-)} + 2(\Delta + i\Gamma_2)V_2^{\,2}\}\Omega}{N_2} \end{pmatrix} \tag{50}$$

where N_1 and N_2 are normalization constants for the zeroth-order basis set $\mathbf{U}_{AA}^{(0)}$ defined as

$$N_1 = \sqrt{M_{(+)}^{\,2} + 4V_1^{\,2}V_2^{\,2}\Omega^2}$$
$$N_2 = \sqrt{M_{(-)}^{\,2} + 4V_1^{\,2}V_2^{\,2}\Omega^2} \tag{51}$$

with

$$M_{(+)} \equiv L_{(-)} + \sqrt{L_{(-)}^{\,2} + 4V_1^{\,2}V_2^{\,2}\Omega^2}$$
$$M_{(-)} \equiv L_{(-)} - \sqrt{L_{(-)}^{\,2} + 4V_1^{\,2}V_2^{\,2}\Omega^2} \tag{52}$$

$$L_{(-)} = (\Delta + i\Gamma_2)V_1^{\,2} - (\Delta + i\Gamma_1)V_2^{\,2}$$
$$L_{(+)} = (\Delta + i\Gamma_2)V_1^{\,2} + (\Delta + i\Gamma_1)V_2^{\,2} \tag{53}$$

It is difficult to confirm the decomposition condition directly from eq 50 because of the complicated dependency of $L_{(\pm)}$, $M_{(\pm)}$, N_1, and N_2 on the laser parameters, V_1, V_2, Δ, and Ω. Therefore, we restrict ourselves to the condition, $\Omega \gg \Delta,\Gamma_1,\Gamma_2$, which leads to

$$\mathbf{U}_{BA}^{(1)} \cong -\frac{1}{\sqrt{2}\Omega}\begin{pmatrix} V_2 & V_2 \\ V_1 & -V_1 \end{pmatrix} \tag{54}$$

Here, we use $M_{(\pm)} \cong \pm 2V_1V_2\Omega$, $N_1 = N_2 \cong 2\sqrt{2}V_1V_2\Omega$ when $\Omega \gg \Delta,\Gamma_1,\Gamma_2$. Note that eq 54 indicates that the first-order mixing becomes negligibly small under the condition, $\Omega \gg V_1$, V_2. Thus, the combined laser condition $\Omega \gg V_1,V_2,\Delta,\Gamma_1,\Gamma_2$ makes it possible to effectively decompose the ladder-type four-level system.

The effective Hamiltonian for the isolated A-space is given as

$$\mathbf{H}_{L4}^{(\mathrm{eff})} = \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix} + \frac{1}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2}\begin{pmatrix} (\Delta + i\Gamma_2)V_1^{\,2} & V_1V_2\Omega \\ V_1V_2\Omega & (\Delta + i\Gamma_1)V_2^{\,2} \end{pmatrix} \tag{55}$$

By diagonalizing eq 55, one obtains eigenvalues

$$\mathbf{E}_A = \mathbf{E}_A^{(0)} + \mathbf{E}_A^{(2)} = \begin{pmatrix} \Delta & 0 \\ 0 & \Delta \end{pmatrix} + \frac{1}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2}\begin{pmatrix} \mathrm{M}_{(+)}/2 & 0 \\ 0 & \mathrm{M}_{(-)}/2 \end{pmatrix} \tag{56}$$

and

$$\mathbf{U}_{AA}^{(0)} = \begin{pmatrix} M_{(+)}/N_1 & M_{(-)}/N_2 \\ 2V_1V_2\Omega/N_1 & 2V_1V_2\Omega/N_2 \end{pmatrix} \tag{57}$$

In contrast to the case of the branch-type four-level system, off-diagonal elements of the effective Hamiltonian $\mathbf{H}_{L4}^{(\mathrm{eff})}$ is not proportional to Δ but to Ω. It should be also noted that the imaginary part of $\mathbf{E}_A^{(2)}$ can be approximated to $\mathrm{Im}[L_{(-)}] = \Gamma_2 V_1^2 - \Gamma_1 V_2^2$ under the condition $\Omega \gg \Delta$. Thus, one can minimize the overall relaxation process by taking V_1,V_2, which satisfies the condition $V_1^2/V_2^2 = \Gamma_1/\Gamma_2$ together with large Ω.

Shown in Figure 8 are the results of numerical calculations for the case $\Gamma_1 \ll \Gamma_2$. The population decay constants are taken to be $\Gamma_1 = 0.02$ and $\Gamma_2 = 0.32$, respectively. We aim at 100% population transfer from $|A_1\rangle$ to $|A_2\rangle$ avoiding the population loss as far as possible. Shown in Figure 8 is the population dynamics with a primitive laser setting, $V_1 = V_2 = \Omega = 1$, $\Delta = 0$. Because the decomposition condition $\Omega \gg V_1,V_2$ is not satisfied, the population oscillates between the A- and B-spaces and the population loss from the B-space is significant. The remaining population at $t = 10\omega^{-1}$ is only 30.9%. Next, we apply the decomposition condition, $\Omega \gg V_1,V_2,\Delta,\Gamma_1,\Gamma_2$. Shown in Figure 9 is the population dynamics under the laser condition, $V_1 = 0.1$, $V_2 = 0.4$, $\Omega = 10$, $\Delta = 0$. It is seen that the population dynamics occurs only in the A-space without populating $|B_1\rangle$, $|B_2\rangle$, which denotes that the decomposition is neatly achieved. The Rabi oscillation between $|A_1\rangle$ and $|A_1\rangle$ is observed, and 97.4% of the population is transferred onto the target state $|A_2\rangle$ at $t = T_R/2 = 393\omega^{-1}$. Note that we take the parameter values of V_1 and V_2, satisfying the condition $V_2^2/V_1^2 = \Gamma_2/\Gamma_1 = 16$ in order to minimize the population loss due to the relaxation processes. On the other hand, shown in Figure 10 is the population dynamics with the same laser condition as those in Figure 9 except V_1 and V_2, whose values are exchanged, i.e., $V_1 = 0.4$, $V_2 = 0.1$. It is seen that about 20% of the total population is lost although the decomposition is well achieved.

Finally, we consider the branch-type five-level system, which is shown in Figure 11. The total Hamiltonian is given as

$$\mathbf{H}_{5B} = \begin{pmatrix} \mathbf{H}_A & \mathbf{V}_{AB} \\ \mathbf{V}_{BA} & \mathbf{H}_B \end{pmatrix} \tag{58}$$

where

$$\mathbf{H}_A = \mathbf{I}_A = \begin{pmatrix} \Delta & 0 & 0 \\ 0 & \Delta & 0 \\ 0 & 0 & \Delta \end{pmatrix}, \mathbf{H}_B = \begin{pmatrix} -i\Gamma_1 & \Omega \\ \Omega & -i\Gamma_2 \end{pmatrix} \tag{59}$$

with

$$\mathbf{V}_{BA} = \begin{pmatrix} V_1 & 0 & V_3 \\ 0 & V_2 & 0 \end{pmatrix}, \mathbf{V}_{AB} = \begin{pmatrix} V_1 & 0 \\ 0 & V_2 \\ V_3 & 0 \end{pmatrix} \tag{60}$$

The first-order mixing is obtained from eq 27 as

$$\mathbf{U}_{BA}^{(1)} = \frac{V_2}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2}\begin{pmatrix} 0 & \frac{\{-M_{(-)} + 2(\Delta + i\Gamma_2)V_{13}^{\,2}\}\Omega}{N_2} & \frac{\{-M_{(+)} + 2(\Delta + i\Gamma_2)V_{13}^{\,2}\}\Omega}{N_3} \\ 0 & \frac{-(\Delta + i\Gamma_1)M_{(-)} + 2V_{13}^{\,2}\Omega^2}{N_2} & \frac{-(\Delta + i\Gamma_1)M_{(+)} + 2V_{13}^{\,2}\Omega^2}{N_3} \end{pmatrix} \tag{61}$$

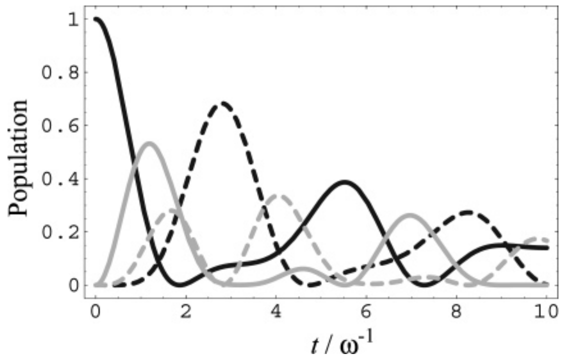

Figure 8. Population dynamics of the ladder-type four-level system with $\Gamma_1 = 0.02$, $\Gamma_2 = 0.32$ under the laser condition, $V_1 = V_2 = \Omega = 1$, $\Delta = 0$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

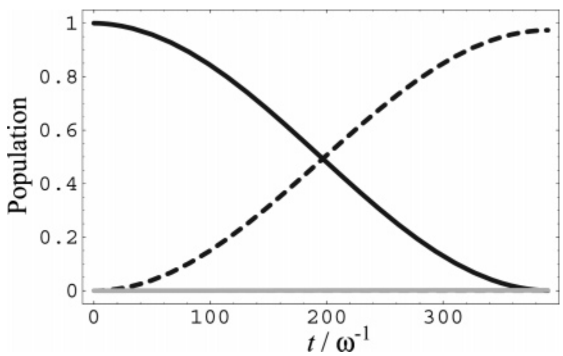

Figure 9. Population dynamics of the ladder-type four-level system with $\Gamma_1 = 0.02$, $\Gamma_2 = 0.32$ under the laser condition, $V_1 = 0.1$, $V_2 = 0.4$, $\Omega = 10$, $\Delta = 0$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

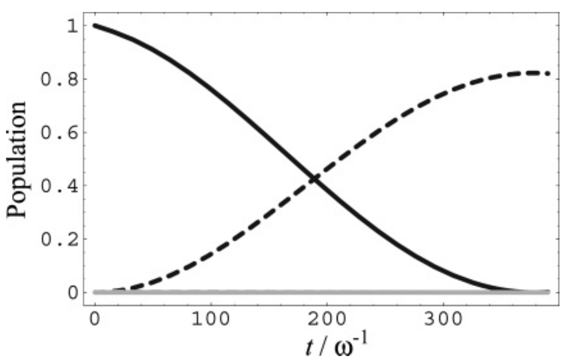

Figure 10. Population dynamics of the ladder-type four-level system with $\Gamma_1 = 0.02$, $\Gamma_2 = 0.32$ under the laser condition, $V_1 = 0.4$, $V_2 = 0.1$, $\Omega = 10$, $\Delta = 0$. Solid, broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

where

$$M_{(+)} = L_{(-)} + \sqrt{{L_{(-)}}^2 + 4{V_{13}}^2{V_2}^2\Omega^2}$$
$$M_{(-)} = L_{(-)} - \sqrt{{L_{(-)}}^2 + 4{V_{13}}^2{V_2}^2\Omega^2} \qquad (62)$$

with

$$L_{(-)} = (\Delta + i\Gamma_2){V_{13}}^2 - (\Delta + i\Gamma_1){V_2}^2$$
$$L_{(+)} = (\Delta + i\Gamma_2){V_{13}}^2 - (\Delta + i\Gamma_1){V_2}^2 \qquad (63)$$
$${V_{13}}^2 = {V_1}^2 + {V_3}^2$$

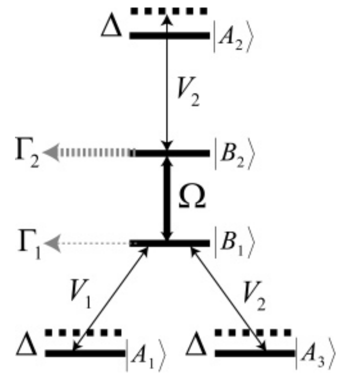

Figure 11. Schematic diagram of the branch-type five-level system. A-space consists of $|A_i\rangle$ ($i = 1, 2, 3$), and B-space consists of $|B_i\rangle$ ($i = 1, 2$). There are weak optical interactions V_1, V_2, and V_3, corresponding to the transitions $|A_1\rangle \leftrightarrow |B_1\rangle$, $|A_2\rangle \leftrightarrow |B_2\rangle$, and $|A_3\rangle \leftrightarrow |B_1\rangle$, respectively, while $|B_1\rangle$ and $|B_2\rangle$ are strongly coupled through interaction Ω. Γ_1 (Γ_2) denotes a population decay constant associated with $|B_1\rangle$ ($|B_2\rangle$), and Δ is a detuning parameter that is commonly taken for the transitions $|A_1\rangle \leftrightarrow |B_1\rangle$, $|A_2\rangle \leftrightarrow |B_2\rangle$, and $|A_3\rangle \leftrightarrow |B_1\rangle$.

Here, N_1, N_2, and N_3 are normalization constants for $\mathbf{U}_{AA}^{(0)}$, which are defined as

$$N_1 = V_{13}$$
$$N_2 = \sqrt{{M_{(-)}}^2 + 4{V_{13}}^2{V_2}^2\Omega^2} \qquad (64)$$
$$N_3 = \sqrt{{M_{(+)}}^2 + 4{V_{13}}^2{V_2}^2\Omega^2}$$

Under the condition $\Omega \gg \Delta,\Gamma_1,\Gamma_2$, we find

$$\mathbf{U}_{BA}^{(1)} \cong -\frac{1}{\sqrt{2}\Omega}\begin{pmatrix} 0 & V_2 & -V_2 \\ 0 & V_{13} & V_{13} \end{pmatrix} \qquad (65)$$

Here we use the approximations, $M_{(\pm)} \cong \pm 2V_{13}V_2\Omega^2$, $N_2 = N_3 \cong 2\sqrt{2}V_{13}V_2\Omega$. Thus, the laser condition, $\Omega \gg V_1,V_2,V_3,\Delta,\Gamma_1,\Gamma_2$, makes it possible to neglect the first-order mixing or achieve the decomposition. Under such a condition, one obtains the effective Hamiltonian for the isolated A-space as

$$\mathbf{H}_{5B}^{(\mathrm{eff})} = \begin{pmatrix} \Delta & 0 & 0 \\ 0 & \Delta & 0 \\ 0 & 0 & \Delta \end{pmatrix} + \frac{1}{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2} \begin{pmatrix} (\Delta + i\Gamma_2){V_1}^2 & V_1V_2\Omega & (\Delta + i\Gamma_2)V_1V_3 \\ V_1V_2\Omega & (\Delta + i\Gamma_1){V_2}^2 & V_2V_3\Omega \\ (\Delta + i\Gamma_2)V_1V_3 & V_2V_3\Omega & (\Delta + i\Gamma_2){V_3}^2 \end{pmatrix} \qquad (66)$$

Diagonalization of $\mathbf{H}_{5B}^{(\mathrm{eff})}$ gives

$$\mathbf{E}_A^{(0)} + \mathbf{E}_A^{(2)} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & \Delta & 0 \\ 0 & 0 & \Delta \end{pmatrix} + \frac{1}{2\{(\Delta + i\Gamma_1)(\Delta + i\Gamma_2) - \Omega^2\}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & L_{(+)} + \sqrt{{L_{(-)}}^2 + 4{V_{13}}^2\Omega^2} & 0 \\ 0 & 0 & L_{(+)} - \sqrt{{L_{(-)}}^2 + 4{V_{13}}^2\Omega^2} \end{pmatrix} \qquad (67)$$

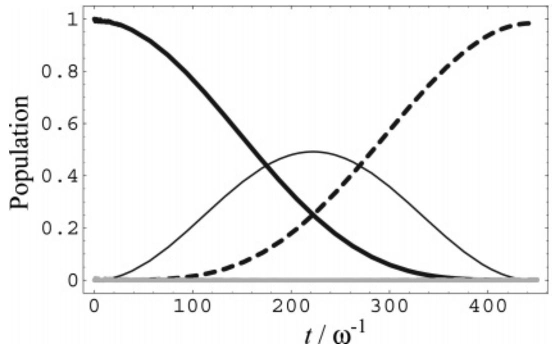

Figure 12. Population dynamics of the branch-type five-level system with $\Gamma_1 = 0.1$, $\Gamma_2 = 0.01$ under the laser condition, $V_1 = V_3 = 1$, $V_2 = 0.1$, $\Omega = 20$, $\Delta = 0$. Thick solid, thin solid, thick broken, gray solid, and gray broken lines denote the population dynamics of $|A_1\rangle$, $|A_2\rangle$, $|A_3\rangle$, $|B_1\rangle$, and $|B_2\rangle$, respectively.

together with the corresponding eigenvectors or zeroth-order states

$$\mathbf{U}_{AA}^{(0)} = \begin{pmatrix} -V_3/N_1 & 2V_1V_2\Omega/N_2 & 2V_1V_2\Omega/N_3 \\ 0 & -M_{(-)}/N_2 & -M_{(+)}/N_3 \\ V_1/N_1 & 2V_2V_3\Omega/N_2 & 2V_2V_3\Omega/N_3 \end{pmatrix} \quad (68)$$

The effective Hamiltonian eq 66 implies that one can create a Λ-type three-level system in the isolated A-space with the condition $\Delta = 0$, $\Gamma_2 \cong 0$. As an example of the population dynamics control, we consider a particular case with $\Gamma_1 = 0.1$, $\Gamma_2 = 0.01$. Our control objective is the population transfer from $|A_1\rangle$ to $|A_3\rangle$ via the intermediate state $|A_2\rangle$. From the diagonal elements of eq 66, it is readily expected we should take small V_2 to avoid the relaxation process Γ_1, which is ten times larger than Γ_2. Shown in Figure 12 is the population dynamics under the parameter setting, $V_1 = V_3 = 1$, $V_2 = 0.1$, $\Omega = 20$, $\Delta = 0$. The initial population on $|A_1\rangle$ is transferred to the target state $|A_3\rangle$ via the intermediate state $|A_2\rangle$ with the final yield, 98.3%. Note also that the relaxation process is well suppressed, as in the case of the branch-type four-level system, and the overall population loss is less than 2%.

Finally, we discuss possible applications of the present scheme onto the realistic molecular systems. One of the typical examples is the isomerization reaction control by laser fields.[6,38] In such a system, the potential surface/curve has multiple minima corresponding to isomers. Because there are significant potential barriers between those isomer states, the corresponding wave functions, which are localized in the potential minima, possess very little overlap to each other. Thus, one cannot expect reaction control using direct optical transitions. To avoid such difficulties, one resorts to using intermediate excited states that are optically accessible from both initial and target isomer states. However, those excited states are likely to be dissipative. In such a case, we can utilize the present control scheme to promote the desired isomerization avoiding the population loss via dissipation.

Notice that one can extract four- or five-level systems from the manifold levels of the realistic molecular system by irradiating lasers with resonant/near-resonant frequency and intensities corresponding to the condition $\Omega > V$. Note also that the peculiar condition for the A-space, i.e., no direct optical transitions, can be achieved readily by not applying the lasers to the transitions in the A-space, even though they are optically allowed.

4. Summary and Conclusion

In this study, we have proposed a general scheme for controlling quantum dynamics of a general multilevel system using CW lasers. The scheme is based on the following procedure: (1) decompose a total system into simple subsystems using intense CW lasers, and (2) apply well-established laser control schemes onto the effectively isolated subsystem. We have also derived the effective Hamiltonian that describes the dynamics within the isolated subsystem based on the second-order perturbation. The scheme has been successfully applied to four- and five-level systems and found to be useful for suppression of the relaxation process as well. One of the advantages of the present control scheme is that one can avoid a fine phase control between the intense lasers utilized for the effective decomposition and the weak lasers. It should be also noted that one does not have to resort to a generic laser optimization scheme, which requires relatively massive computations and tends to produce complicated control fields.

Acknowledgment. This research was supported, in part, by the Japan Society for the Promotion of Science (JSPS), Grant-in-Aid for Scientific Research(C), KAKENHI(17550019), and Keio Gijuku Fukuzawa Memorial Fund for the Advancement of Education and Research.

References and Notes

(1) Tannor, D. J.; Rice, S. A. *J. Chem. Phys.* **1985**, *83*, 5013.
(2) Tannor, D. J.; Rice, S. A. *Adv. Chem. Ser.* **1988**, *70*, 441.
(3) Tang, H.; Kosloff, R.; Rice, S. A. *J. Chem. Phys.* **1996**, *104*, 5457.
(4) Shore, B. W. *The Theory of Coherent Atomic Excitation*; Wiley: New York, 1975; Vol. 2.
(5) Kosloff, R.; Rice, S. A.; Gaspard, P.; Tersigni, S.; Tannor, D. J. *Chem. Phys.* **1989**, *139*, 201.
(6) Sugawara, M.; Fujimura, Y. *J. Chem. Phys.* **1994**, *100*, 5646.
(7) Chelkowski, S.; Bandrauk, A. D. *Chem. Phys. Lett.* **1995**, *233*, 185.
(8) Sugawara, M.; Yoshizawa, S.; Yabushita, S. *Chem. Phys. Lett.* **2001**, *350*, 253.
(9) Sugawara, M. *Chem. Phys. Lett.* **2002**, *358*, 290.
(10) Sugawara, M. *J. Chem. Phys.* **2003**, *118*, 6784.
(11) Yamaki, M.; Hoki, K.; Ohtsuki, Y.; Kono, H.; Fujimura, Y. *J. Am. Chem. Soc.* **2005**, *127*, 7300.
(12) Ohtsuki, Y. *Chem. Phys. Lett.* **2005**, *404*, 126.
(13) Abe, M.; Ohtsuki, Y.; Fujimura, Y.; Domcke, W. *J. Chem. Phys.* **2005**, *123*, 144508.
(14) Teranishi, Y.; Ohtsuki, Y.; Hosaka, K.; Chiba, H.; Katsuki, H.; Ohmori, K. *J. Chem. Phys.* **2006**, *124*, 114110.
(15) Sugawara, M. *J. Chem. Phys.* **2005**, *123*, 204115.
(16) Kuhn, A.; Coulston, G. W.; He, G. Z.; Schiemann, S.; Bergmann, K. *J. Chem. Phys.* **1992**, *96*, 4215.
(17) Gaubatz, U.; Rudecki, P.; Schiemann, S.; Bergmann, K. *J. Chem. Phys.* **1990**, *92*, 5363.
(18) Nishikawa, K.; Yamaguchi, T.; Sakamoto, I.; Nishi, K.; Ohta, Y.; Nagao, H. *Synth. Met.* **2003**, *137*, 1437.
(19) Ohta, Y.; Yoshimoto, T.; Nagao, H.; Nishikawa, K. *Int. J. Quantum. Chem.* **2000**, *80*, 1068.
(20) Malinovsky, V. S.; Meiyer, C.; Tannor, D. J. *Chem. Phys.* **1997**, *221*, 67.
(21) Kobrak, M. N.; Rice, S. A. *Phys. Rev. A* **1998**, *57*, 2885.
(22) Kis, Z.; Karpati, A.; Shore, B. W.; Vitanov, N. V. *Phy. Rev. A* **2004**, *70*, 053405.
(23) Niu, Y.; Gong, S. *J. Phys. Soc. Jpn.* **2004**, *73*, 2131.
(24) Gong, J.; Rice, S. A. *J. Chem. Phys.* **2004**, *120*, 3777.
(25) Gong, J.; Rice, S. A. *J. Chem. Phys.* **2004**, *120*, 5117.
(26) Gong, J.; Rice, S. A. *J. Chem. Phys.* **2004**, *120*, 9984.
(27) Gong, J.; Rice, S. A. *J. Chem. Phys.* **2004**, *121*, 1364.
(28) Sugawara, M. *Chem. Phys. Lett.* **2006**, *428*, 457.
(29) Shi, S. H.; Rabitz, H. *J. Chem. Phys.* **1990**, *92*, 364.
(30) Rabitz, H.; de Vivie-Riedle, R.; Motzkus, M.; Kompa, K. *Science*. **2000**, *288*, 824.
(31) Rabitz, H.; Shi, S. *J. Chem. Phys.* **1990**, *92*, 364.
(32) Gross, P.; Neuhauser, D.; Rabitz, H. *J. Chem. Phys.* **1993**, *98*, 4557.
(33) Ohtsuki, Y.; Turinici, G.; Rabitz, H. *J. Chem. Phys.* **2004**, *120*, 5509.
(34) Ohtsuki, Y.; Sugawara, M.; Kono, H.; Fujimura, Y. *Bull. Chem. Soc. Jpn.* **2001**, *74*, 1167.
(35) Lamb, G. L., Jr. *Rev. Mod. Phys.* **1971**, *43*, 99.
(36) Allen, J.; Eberly, J. H. *Optical Resonance and Two Level Atoms*,; Wiley: New York, 1975.
(37) Light, P. L.; Milonni, P. W. *Phys. Rep.* **1980**, *66*, 21.
(38) Carreira, L. A.; Lord, R. C. *J. Chem. Phys.* **1969**, *51*, *2735*.