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Hamiltonian Reduction of Quantum Systems Controlled by Pulses

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We explores Hamiltonian reduction in pulse-controlled finite-dimensional quantum systems with near-degenerate eigenstates. A quantum system with a non-degenerate ground state and several near-degenerate excited states is controlled by a short pulse, and the objective is to maximize the collective population on all excited states when we treat all of them as one level. Two cases of the systems are shown to be equivalent to effective two-level systems. When the pulse is weak, simple relations between the original systems and the reduced systems are obtained. When the pulse is strong, these relations are still available for pulses with only one frequency under the first-order approximation.

Key words: Quantum control, Population transfer, Laser pulse, Dimension reduction

I. INTRODUCTION

Quantum theory has established the foundation for understanding and explaining phenomena at atomic and sub-atomic scales, and promoted the development of laser, semi-conductor, nuclear power, and so on. During its development, quantum theory is fusing with other branches of science. Quantum control is a new area of science originated from the combination of quantum and control theories. The applications of quantum control in chemical reactions have already produced exciting achievements [1, 2] and the invention of laser further accelerated the development of quantum control techniques. In particular, laser coherence controlling manipulates the quantum state by quantum coherent effects. Many efficient schemes for selective population transfer have been developed, such as optimal control [3], coherent control [4], stimulated Raman adiabatic passage (STIRAP) control [5, 6], *etc.* A number of control experiments, including on complex systems [1, 2, 7], employ closed-loop optimal control [8]. The essential idea of population transfer is to begin with an atom or molecule in which the internal structure is in a specified discrete quantum state initially and then, by exposing this system to a controlled pulse of radiation, force the internal structure into a desired target state [9]. Shore has given a detailed review about coherent manipulations of atoms using laser [10].

Recent studies have investigated the effects of field noise and decoherence, and shown that controlled quantum dynamics can survive from intensive field noise and decoherence even cooperate with them under special circumstances [11, 12]. The foundation of cooperating between control field and field noise or decoherence was explained perfectly by the perturbation theory of quantum dynamics [12, 13]. The solutions for perfect population transfer in non-degenerate multi-level quantum systems have been studied [14]. In these theoretical models, there is an assumption that each component of the contorl pulse is only resonant with one transition of the system. However, in many cases of laser exciting atoms or molecules [15], spectrum width of the laser is not sharp enough to identify near-degenerate levels.

In this work, we explore Hamiltonian reduction in pulse-controlled finite-dimensional quantum systems with near-degenerate eigenstates. It is proved that a multi-level system with a non-degenerate ground state and several near-degenerate excited states controlled by a short pulse can be reduced to a two-level system in the case of both weak and strong fields. Quantum dissipation is not considered in this work, because the driving pulses are assumed to be short. The effect of dissipation in finite-dimensional systems has been considered carefully [16, 17].

The general model of Hamiltonian reduction in pulse-controlled quantum systems and the N-level system reducing to a two-level system with the weak control pulse are given. We also analyze the case of strong control pulses with only one frequency, and show how the system reduces to a two-level system even for the strong control pulses.

II. MODEL

We consider an N-level quantum system controlled by short pulse. The system consists of a ground state $|0\rangle$, and $N-1$ near degenerate excited states $|k\rangle$, k=1,

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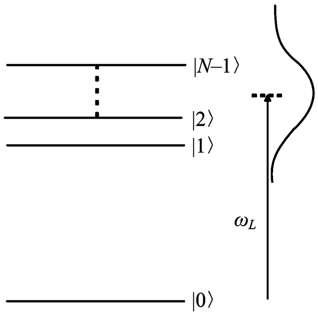

FIG. 1 N-level quantum system with a ground state and $N-1$ near-degenerate excited states. The system is driven by a short pulse with bandwidth much larger than that of the excited states.

$\cdots$, $N-1$, as shown in Fig.1.

The system Hamiltonian H contains two parts, *i.e.* H_0, the field-free Hamiltonian, and $-\mu E(t)$, the interaction with external field. Here μ is the dipole operator which couples the field-free Hamiltonian with laser pulse $E(t)$. We write the Hamiltonian in Schrödinger picture as follows:

$$H = H_0 - \mu E(t) \tag{1}$$

$$H_0 = \sum_{k=1}^{N-1} (\omega_L + \delta_k) |k\rangle \langle k| \tag{2}$$

$$\mu = \sum_{k=1}^{N-1} \mu_k |k\rangle \langle 0| + \text{h.c.} \tag{3}$$

here, h.c. indicates Hermite conjugate and $\omega_L + \delta_k$ represents energy of the kth excited state with detuning $\delta_k \ll \omega_L$. The controlling field $E(t)$ has the form [18],

$$E(t) = 2s(t) \sum_{j=1}^{n} A_j \cos\left[(\omega_L + \eta_j)t + \varphi_j\right] \tag{4}$$

where $s(t)$ is the envelope of the pulse, $\omega_L + \eta_j$ is the frequency of the pulse with $\eta_j \ll \omega_L$, A_j is the amplitude and φ_j is the phase. The system is initially on the ground state at time t_0,

$$\Psi_S(t_0) = |0\rangle \tag{5}$$

The control objective p_e is the population on all excited states at final time t_f,

$$p_e = \sum_{k=1}^{N-1} |\langle k|\Psi_S(t_f)\rangle|^2 \tag{6}$$

where $\Psi_S(t)$ is the wavepacket of the system whose evolution is governed by Schrödinge equation,

$$i\frac{\mathrm{d}\psi_S(t)}{\mathrm{d}t} = [H_0 - \mu E(t)]\,\psi_S(t) \tag{7}$$

We will show that this multi-level system behaves dynamically as a two-level system,

$$H_e = (\omega_L + \delta_e) |1\rangle \langle 1| - \mu_e(|1\rangle \langle 0| + |0\rangle \langle 1|)E(t) \tag{8}$$

when the pulse is short enough to make its bandwidth Δ much larger than detuning Δ_k. It is shown that the Hamiltonian of the effective two-level system is closely related to that of the N-level system. Analytical relations between original system and reduced system are obtained in two cases.

III. WEAK FIELD WITH MULTI-FREQUENCIES

If the field is weak, then the perturbation approximation is valid for further discussion. Eliminating H_0 in Eq.(7) through substitution,

$$\psi(t) = \exp(iH_0t)\psi_S(t) \tag{9}$$

we obtain the dynamical equation in interaction picture,

$$i\frac{\mathrm{d}\psi(t)}{\mathrm{d}t} = -V_I(t)\psi(t) \tag{10}$$

$$\begin{aligned} V_I(t) &= \exp(iH_0t)\mu E(t)\exp(-iH_0t) \\ &= E(t)\left\{\sum_{k=1}^{N-1} \mu_k |k\rangle \langle 0| \cdot \right. \\ &\qquad \left. \exp[i(\omega_L + \delta_k)t] + \text{h.c.}\right\} \end{aligned} \tag{11}$$

Perturbative solution of Eq.(10) is

$$\begin{aligned} \psi(t_f) &= \exp_+\left[i\int_{t_0}^{t_f} V_I(t)\,\mathrm{d}t\right]\psi(t_0) \\ &\approx \left[1 + i\int_{t_0}^{t_f} V_I(t)\mathrm{d}t\right]\psi(t_0) \end{aligned} \tag{12}$$

here, $\exp_+$ is the time-ordered exponential [19]. Hence, the population on the kth excited state is

$$\begin{aligned} p_k &\approx \left|\int_{t_0}^{t_f} \langle k|V_I(t)|0\rangle\,\mathrm{d}t\right|^2 \\ &= \left|\mu_k \int_{t_0}^{t_f} E(t)\exp[i(\omega_L + \delta_k)t]\mathrm{d}t\right|^2 \\ &\approx {\mu_k}^2 |\widetilde{E}(\omega_L + \delta_k)|^2 \end{aligned} \tag{13}$$

here, we assume the control field is almost zero when $t > t_f$ and $t < t_0$. $\widetilde{E}(\omega)$ is the Fourier component of the control field, *i.e.*

$$\widetilde{E}(\omega) = \int_{-\infty}^{\infty} E(t)\exp(i\omega t)\mathrm{d}t \tag{14}$$

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Collective population on all excited states is

$$\begin{aligned} p_e &= \sum_{k=1}^{N-1} p_k \\ &\approx \sum_{k=1}^{N-1} {\mu_k}^2 \left|\widetilde{E}(\omega_L+\delta_k)\right|^2 \\ &\approx \sum_{k=1}^{N-1} {\mu_k}^2 \{\left|\widetilde{E}(\omega_L)\right|^2 + 2\delta_k \mathrm{Re}[\widetilde{E}^*(\omega_L)\widetilde{E}'(\omega_L)]\} \end{aligned} \quad (15)$$

We only keep the first-order Taylor expansion with respect to detuning δ_k in Eq.(15), since δ_k is much smaller than bandwidth of the pulse. It is easy to compare it with the population on the excited state of a effective two-level system,

$$\begin{aligned} p'_e &\approx {\mu_e}^2 \left|\widetilde{E}\left(\omega_L+\delta_e\right)\right|^2 \\ &\approx {\mu_e}^2 \{\left|\widetilde{E}(\omega_L)\right|^2 + 2\delta_e \mathrm{Re}[\widetilde{E}^*(\omega_L)\widetilde{E}'(\omega_L)]\} \end{aligned} \quad (16)$$

Hence the system can be reduced to a two-level one with the parameters

$${\mu_e}^2 = \sum_{k=1}^{N-1} {\mu_k}^2 \quad (17)$$

$$\delta_e = \frac{\sum_{k=1}^{N-1} {\mu_k}^2 \delta_k}{\sum_{k=1}^{N-1} {\mu_k}^2} \quad (18)$$

here we have assumed $\mathrm{Re}[\widetilde{E}^*(\omega_L)\widetilde{E}'(\omega_L)]$ is not zero, otherwise, the relation between detunings should be

$${\delta_e}^2 = \frac{\sum_{k=1}^{N-1} {\mu_k}^2 {\delta_k}^2}{\sum_{k=1}^{N-1} {\mu_k}^2} \quad (19)$$

or even higher order. Hence, the multi-level system in Fig.1 reduces to a two-level energy system when the control pulse is short and weak.

IV. STRONG FIELD WITH ONLY ONE FREQUENCY

If the control pulse is strong, it is not easy to get simple analytical relations between the original system and the reduction system, and numerical simulation is a better choice, because the perturbation approximation is invalid. However, if the field has only one frequency, it is still possible to obtain analytical relations as shown below. In this case, dynamics of the system is still given by Eq.(7), but with the pulse $E(t)$ being a simple form:

$$E(t) = 2s(t)A\cos(\omega_L t+\varphi) \quad (20)$$

where ω_L is the frequency of the pulse and $s(t)$ is the pulse envelope function. Considering the interaction of this laser pulse with the N-level system in Fig.1, we can express Eq.(7) as,

$$i\frac{\mathrm{d}\psi_S(t)}{\mathrm{d}t} = [H_L - V(t)]\,\psi_S(t) \quad (21)$$

$$H_L = \omega_L \sum_{k=1}^{N-1} |k\rangle\langle k| \quad (22)$$

$$V(t) = \mu E(t) + H_L - H_0 \quad (23)$$

Similarly as mentioned above, the substitution

$$\psi_1(t) = \exp[i(H_L t - \varphi|0\rangle\langle 0|)]\psi_S(t) \quad (24)$$

eliminates H_L and produces the dynamical equation in interaction picture:

$$i\frac{\mathrm{d}\psi_1(t)}{\mathrm{d}t} = -V_I(t)\psi_1(t) \quad (25)$$

$$\begin{aligned} V_I(t) &= \exp[i(H_L t - \varphi|0\rangle\langle 0|)]V(t)\cdot \\ &\quad \exp[-i(H_L t - \varphi|0\rangle\langle 0|)] \\ &= 2s(t)A\cos(\omega_L t+\varphi)\Big\{\exp[i(\omega_L t+\varphi)]\cdot \\ &\quad \sum_{k=1}^{N-1} \mu_k|k\rangle\langle 0| + \mathrm{h.c.}\Big\} + H_L - H_0 \end{aligned} \quad (26)$$

If the external field is not very strong, then we can use rotating wave approximation (RWA) [20, 21] to omit all high-frequency terms

$$\begin{aligned} &2\cos(\omega_L t+\varphi)\exp[i(\omega_L t+\varphi)] = \\ &\quad = 1-\exp[2i(\omega_L t+\varphi)] \approx 1 \end{aligned} \quad (27)$$

and obtain $V_I(t)$ as

$$V_I(t) \approx s(t)H_1 - V_1 \quad (28)$$

$$H_1 = \sum_{k=1}^{N-1} \Omega_k\left(|0\rangle\langle k| + |k\rangle\langle 0|\right) \quad (29)$$

$$\begin{aligned} V_1 &= H_0 - H_L \\ &= \sum_{k=1}^{N-1} \delta_k |k\rangle\langle k| \end{aligned} \quad (30)$$

where Ω_k is the kth Rabi frequency defined as

$$\Omega_k = \mu_k A \quad (31)$$

We have assumed that all detunings of the system $\{\delta_k\}$ are much less than Δ, which is the bandwidth of the control pulse. Here, it is possible to use another type of perturbation approximation. At first, we can separate H_I into two parts: the main Hamiltonian H_1 and perturbative Hamiltonian V_1. The substitution

$$\psi_2(t) = \exp[-iH_1\widehat{s}(t,t_0)]\psi_1(t) \quad (32)$$

$$\widehat{s}(t_2,t_1) = \int_{t_1}^{t_2} s(\tau)\mathrm{d}\tau \quad (33)$$

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yields the dynamical equation in interaction picture:

$$i\frac{\mathrm{d}\psi_2(t)}{\mathrm{d}t} = V_2(t)\psi_2(t) \quad (34)$$

$$V_2(t) = \exp[-iH_1\widehat{s}(t,t_0)]V_1\exp[iH_1\widehat{s}(t,t_0)] \quad (35)$$

V_1 and $V_2(t)$ can be considered to be small because δ_k is much smaller than $t_f-t_0\approx\Delta$. Perturbative treatment produces the solution

$$\psi_2(T_f) = \exp_+\left[-i\int_{t_0}^{t_f}V_2(t)\mathrm{d}t\right]\psi_2(t_0) \approx \left[1-i\int_{t_0}^{t_f}V_2(t)\mathrm{d}t\right]\psi_2(t_0) \quad (36)$$

Hence, population on the kth excited level is

$$p_k \approx \left|\langle k|\exp[iH_1\widehat{s}(t_f,t_0)]\left[1-i\int_{t_0}^{t_f}V_2(t)\mathrm{d}t\right]|0\rangle\right|^2 = \left|\langle k|\exp[iH_1\widehat{s}(t_f,t_0)] - \int_{t_0}^{t_f}\exp[iH_1\widehat{s}(t_f,t)]\cdot V_1\exp[iH_1\widehat{s}(t,t_0)]\,|0\rangle\,\mathrm{d}t\right|^2 \quad (37)$$

For a rectangle pulse

$$s(t) = \begin{cases}1, & 0\le t\le t_f\\ 0, & \text{else}\end{cases} \quad (38)$$

where we set t_0=0, it is possible to get an analytical form of p_k. The key to deriving p_k lies in the calculation of the exponential of H_1. Since H_1 has a simple form, we can verify that it has only two non-zero eigenvalues, *i.e.* Ω_e and $-\Omega_e$, and the corresponding eigenvectors are

$$|v_1\rangle = \frac{\Omega_e}{\sqrt{2}},\frac{\Omega_1}{\sqrt{2}},\cdots,\frac{\Omega_{N-1}}{\sqrt{2}} \quad (39)$$

$$|v_2\rangle = \frac{\Omega_e}{\sqrt{2}},-\frac{\Omega_1}{\sqrt{2}},\cdots,-\frac{\Omega_{N-1}}{\sqrt{2}} \quad (40)$$

in which

$${\Omega_e}^2 = \sum_{k=1}^{N-1}{\Omega_k}^2 \quad (41)$$

Assume that H_1 will be diagonalized by an orthogonal matrix O, *i.e.* $\mathrm{O}^{\mathrm{T}}H_1\mathrm{O}=\Gamma$ while $\Gamma=\Omega_e|0\rangle\langle 0|-\Omega_e|1\rangle\langle 1|$, and further decompose O as $A+S$ in which $A=(|v_1\rangle|v_2\rangle|0\rangle\cdots|0\rangle)$ while $|0\rangle$ is the null vector. Utilizing the orthogonality of O, we have $A\cdot A^{\mathrm{T}}+S\cdot S^{\mathrm{T}}=\mathrm{II}$, and then obtain $\exp[H_1]$ as follows:

$$\exp[H_1] = A\exp[\Gamma]A^{\mathrm{T}}+\mathrm{II}-AA^{\mathrm{T}} \quad (42)$$

This formula allows us to calculate the exponential of H_1 and further get a simple analytical form of the population of each excited level:

$$p_k \approx \left|i\frac{\Omega_k}{\Omega_e}\sin(\Omega_e t_f) + \frac{\Omega_k}{{\Omega_e}^2}\delta_k\cdot 2\sin^2\left(\frac{1}{2}\Omega_e t_f\right) + \frac{\Omega_k}{{\Omega_e}^4}\sum_{j=1}^{N-1}\delta_j{\Omega_j}^2\cdot\left[\frac{1}{2}\Omega_e t_f\sin(\Omega_e t_f) - 2\sin^2\left(\frac{1}{2}\Omega_e t_f\right)\right]\right|^2 \quad (43)$$

Then the population on all excited states is

$$p_e = \sum_{k=1}^{N-1}p_k \approx \sin^2(\Omega_e t_f) + \frac{1}{{\Omega_e}^4}\sum_{k=1}^{N-1}\Omega_k^2\cdot\left\{\delta_k 2\sin^2\left(\frac{1}{2}\Omega_e t_f\right) + \frac{1}{{\Omega_e}^2}\sum_{j=1}^{N-1}\delta_j{\Omega_j}^2\cdot\left[\frac{1}{2}\Omega_e t_f\sin(\Omega_e t_f) - 2\sin^2\left(\frac{1}{2}\Omega_e t_f\right)\right]\right\}^2 \quad (44)$$

Corresponding population on the excited state of the effective two-level system is

$$p'_e = \left|i\sin(\mu_e A t_f) + \frac{1}{2}\delta_e t_f\sin(\mu_e A t_f)\right|^2 \quad (45)$$

Hence the parameters for the effective two-level system are

$${\mu_e}^2 = \sum_{k=1}^{N-1}{\mu_k}^2 \quad (46)$$

$${\delta_e}^2 = \frac{1}{{\mu_e}^4A^2{t_f}^2}\sum_{k=1}^{N-1}{\mu_k}^2\left\{2\delta_k\mathrm{tg}\left(\frac{1}{2}\mu_e A t_f\right) + \frac{1}{{\mu_e}^2}\sum_{j=1}^{N-1}\delta_j{\mu_j}^2\left[\mu_e A t_f - 2\mathrm{tg}\left(\frac{1}{2}\mu_e A t_f\right)\right]\right\}^2 \quad (47)$$

It is interesting to see that the effective δ_e depends on the amplitude of the control pulse A, and when A approaches zero,

$${\delta_e}^2 \to \frac{\sum_{k=1}^{N-1}{\mu_k}^2{\delta_k}^2}{\sum_{k=1}^{N-1}{\mu_k}^2} \quad (48)$$

The relation goes back to Eq.(19). Hence the multi-level system in Fig.1 reduces to a two-level energy system for the strong control pulse.

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V. CONCLUSION AND DISCUSSION

Interaction between laser and matter is an important. It reflects the optical properties of matter and can be employed to achive through controlling amplitudes and phases of the laser field. In this work, we conduct intensive research into the degeneracy of energy level of multi-level systems under different control lasers. The interaction of multi-level systems and different laser fields is systematically studied. Quantum systems with a ground state and several near-degenerate excited states are controlled by short pulses. The system is shown to behave like a two-level system with simple effective parameters when the objective is to control collective population on all excited states. We point out that the dimension of a multi-level system can be reduced under certain conditions. In order to get the precise expressions of the approximate pulse-driven two-level system from the original multi-level system, we consider the effect of the near-degenerate eigenstates and the strength of the laser, and treat the system under rotating wave approximation and perturbative approximation. In the cases of both weak and strong control pulses, we obtain the analytic expressions of equivalent parameters of the effective two-level system. The results can be applied to general multi-level system. The conclusions could be utilized to identify the effective dimension of a quantum multi-level system controlled by laser, and could be used to simplify the design of the control strategies.

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[1] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, and G. Gerber, Science **282**, 919 (1998).
[2] C. Daniel, J. Full, L. González, C. Lupulescu, J. Manz, A. Merli, Š. Vajda, and L. Wöste, Science **299**, 536 (2003).
[3] A. P. Peirce, M. A. Dahleh, and H. Rabitz, Phys. Rev. A **37**, 4950 (1988).
[4] A. Assion, T. Baumert, J. Helbing, V. Seyfried, and G. Gerber, Chem. Phys. Lett. **259**, 488 (1996).
[5] U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann, J. Chem. Phys. **92**, 5363 (1990).
[6] M. N. Kobrak and S. A. Rice, Phys. Rev. A **57**, 2885 (1998).
[7] J. Petersen, R. Mitrić, V. B. Koutecký, Jean-Pierre Wolf, J. Roslund, and H. Rabitz, Phys. Rev. Lett. **105**, 073003 (2010).
[8] R. S. Judson and H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992).
[9] N. V. Vitanov, T. Halfmann, B. W. Shore, and K. Bergmann, Annu. Rev. Phys. Chem. **52**, 763 (2001).
[10] B. W. Shore, Acta Phys. Slovaca **58**, 243 (2008).
[11] F. Shuang and H. Rabitz, J. Chem. Phys. **121**, 9270 (2004).
[12] F. Shuang and H. Rabitz, J. Chem. Phys. **124**, 154105 (2006).
[13] F. Shuang, H. Rabitz, and M. Dykman, Phys. Rev. E **75**, 021103 (2007).
[14] X. Chen, Y. X. Wang, Y. J. Ge, J. H. Shi, H. Rabitz, and F. Shuang, J. Theor. Comp. Chem. **9**, 847 (2010).
[15] E. S. Kyoseva and N. V. Vitanov, Phys. Rev. A **73**, 023420 (2006).
[16] R. Xu, Y. J. Yan, Y. Ohtsuki, Y. Fujimura, and H. Rabitz, J. Chem. Phys. **120**, 6600 (2004).
[17] R. X. Xu, B. L. Tian, J. Xu, and Y. J. Yan, J. Chem. Phys. **130**, 074107 (2009).
[18] A. Lindinger, S. M. Weber, C. Lupulescu, F. Vetter, M. Plewicki, A. Merli, L. Woste, A. F. Bartelt, and H. Rabitz, Phys. Rev. A **71**, 013419 (2005).
[19] P. Grigolini and F. Marchesoni, Adv. Chem. Phys. **62**, 29 (1985).
[20] I. I. Rabi, N. F. Ramsey, and J. Schwinger, Rev. Mod. Phys. **26**, 167 (1954).
[21] B. W. Shore, *The Theory of Coherent Atomic Excitation*, Vol. 2, New York: Wiley, (1990).

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