Geometric control for atomic systems

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Abstract

The problem of explicit generation of unitary operators for atomic systems with degenerate energy levels is considered. The Lie algebra structure is used to derive constructive control schemes for the creation of arbitrary superposition states and selective population interchanges for a transition between two three-fold degenerate energy levels.

1 Introduction

The problem of explicit generation of arbitrary unitary operators for quantum systems using the system's Lie algebra structure and geometric control has been the subject of several recent papers on quantum control [1, 2, 3, 4, 5, 6, 7, 8]. It has been studied especially for coupled spin systems [1, 2] and generic N-level systems with non-degenerate energy levels and transition frequencies [6]. The results have also been applied to related problems such as state preparation and optimization of observables for quantum systems [8].

In this paper we study the problem of constructive control for atomic systems. Unlike molecular systems, where the density of the ro-vibrational states poses a serious challenge for geometric control, atomic systems are generally rather good candidates for this technique since there are no ro-vibrational states to worry about, and the energy levels for electronic transitions are sufficiently separated in most cases, to allow frequency-selective excitation of individual transitions while neglecting the effect of the field on the other (off-resonant) transitions.

Unfortunately, the results on constructive control of generic N-level quantum systems cited above are often not directly applicable to electronic transitions in atomic systems since most atomic energy levels are degenerate. Strictly speaking, the precise energy level structure of a real atom depends on various factors such as the spin of the atomic nucleus, electron spin, and spin–orbit interactions for the particular species of atom under consideration. For many purposes, however, it suffices to account for the basic degeneracy of the atomic energy levels without considering their fine or hyperfine structure. We shall therefore restrict ourselves in the following to such models.

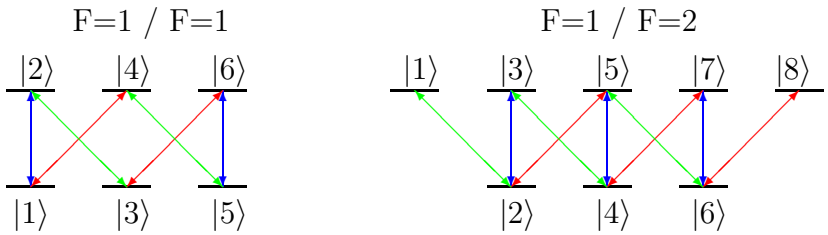

Figure 1: Transition diagram for $F = 1$ to $F' = 1$ transition (left) and $F = 1$ to $F' = 2$ transition (right). Blue indicates coupling by a linearly polarized field, while green / red indicate coupling by a left / right circularly polarized field, respectively.

2 Basic facts from atomic quantum theory

If the fine structure is neglected, the degeneracy of an atomic energy level is $2F+1$, where F is a quantum number that can take positive integer and half integer values. Since transitions between levels with F-values differing by more than one are usually prohibited by atomic selection rules, we only have to consider transitions with $\Delta F = 0$ or $\Delta F = \pm 1$. The sublevels of a degenerate atomic energy level are usually distinguished by a quantum number m, which can take values from $-F$ to F. For integer values of F, there are $2F + 1$ sublevels and m can take integer values from $-F$ to F. For half integer values of F there are $2F$ sublevels and m can take half integer values ranging from $-F$ to F.

The interaction of the system with an external field of the appropriate frequency depends on the polarization of the field. A linearly polarized field generally couples only sublevels with the same quantum number m, while a left or right circularly polarized field will only couple transitions between sublevels with $\Delta m = \pm 1$, respectively. Transitions between sublevels with $|\Delta m| > 1$ are prohibited, as are $m = 0$ to $m = 0$ transitions if both energy levels have the same F value (and $F > 0$). The coupling diagrams for a transition $F = 1$ to $F' = 1$ and a transition $F = 1$ to $F' = 2$ are shown in figure 1.

3 Implications for control of atomic transitions

Given the basic constraints mentioned the previous section, we see immediately that the number of transitions that are simultaneously excited by an external control field, whether linearly, left or right circularly polarized, is one only for transitions from $F = 0$ to $F' = 0$ and $F = 0$ to $F' = 1$. In all other cases, a control field simultaneously couples multiple sublevels. For instance, for $F = 1$ to $F' = 1$ transitions we always simultaneously couple two sublevels; for $F = 1$ to $F' = 2$ transitions, three sublevels are simultaneously coupled; for $F = 2$ to $F' = 2$, there are four, etc. The degree of controllability of these transitions was studied in [9], where it was shown that transitions with $\Delta F = \pm 1$ are always mixed-state controllable, while transitions with $\Delta F = 0$ (except for transitions $F = 0$ to $F' = 0$) are only pure-state controllable in general. We shall now improve these results by using the basic structure of

the Lie algebra to derive constructive control schemes.

A transition between two $F = 0$ levels corresponds to the trivial case of a non-degenerate two-level system, which has been extensively studied [3, 4]. The constructive control techniques presented in [6, 8] are also directly applicable to $F = 0$ to $F' = 1$ transitions since, despite the degeneracy of the $F = 1$ level, we can selectively address each of the three possible transitions by choosing either linearly, left or right circularly polarized fields. For all other cases, however, the situation is quite different. Due to space constraints we shall only consider the case $F = 1$ to $F' = 1$. However, although each case is slightly different, the basic techniques apply in all cases.

Any external field driving a transition between two three-fold degenerate energy levels will simultaneously couple two sublevels, no matter how the polarization of the field is chosen. See figure 1. Moreover, if we assume that the transition probabilities for allowed transitions between all sublevels are equal, for instance, then the system is *not* mixed-state or operator controllable since the dynamical Lie algebra generated is $sp(3)$ and the dynamical Lie group of the system contains only unitary operators that satisfy $\hat{U}^T \hat{J} \hat{U} = \hat{J}$, where $\hat{J}$ is an anti-diagonal matrix whose non-zero elements are $\{+1, -1, +1, -1, +1, -1\}$. Thus, not every unitary operator can be dynamically generated, including especially the permutation matrices that correspond to selective excitation of a single sublevel [9]. However, the system is at least pure-state controllable in all cases. Thus, it is possible, for instance, to create arbitrary superposition states from a pure initial state, and certain unitary operations for mixed states can also be implemented. We shall now study the evolution of the control system to derive constructive control schemes for these applications.

4 Evolution of the control system

Using a rotating frame and the rotating wave approximation, the dynamical equation for the time evolution operator $\hat{U}(t, t_0)$ of the system subject to a control field of the form $f(t) = 2A(t)\cos(\omega_0 t + \phi)$, which is resonant with the transition frequency $\omega_0 = (E_2 - E_1)/\hbar$, is (see [8], appendix A)

$$\dot{U}_I(t) = A(t)\mathrm{i}\hat{H}_I \hat{U}_I, \tag{4.1}$$

where we have chosen the time units $[t] = \omega_0^{-1}$ and field units $[A] = \hbar\omega_0 p_{12}^{-1}$, p_{12} being the dipole moment of the transition. Note that the amplitude of the field $A(t)$ must be slowly varying compared to ω_0^{-1} for this approximation to be valid. $\hat{U}(t, t_0)$ determines the time evolution of a pure state $|\Psi(t_0)\rangle$ via $|\Psi(t)\rangle = \hat{U}(t, t_0)|\Psi(t_0)\rangle$ and the evolution of a density matrix via $\hat{\rho}(t) = \hat{U}(t, t_0)\hat{\rho}_0\hat{U}(t, t_0)^\dagger$.

The interaction Hamiltonian $\hat{H}_I$ in equation (4.1) depends on the polarization of the field. In the following, we use $\hat{H}_1$ for a linearly polarized field, and $\hat{H}_2$ / $\hat{H}_3$ for a left / right circularly polarized field, respectively. If the sublevels are labelled as in figure 1 (left) then

we have concretely

$$\begin{aligned} \mathrm{i}\hat{H}_1 &= \sin(\phi)d_1(\hat{x}_{1,2}-\hat{x}_{5,6})-\cos(\phi)d_1(\hat{y}_{1,2}-\hat{y}_{5,6}) \\ \mathrm{i}\hat{H}_2 &= \sin(\phi)d_2(\hat{x}_{1,4}+\hat{x}_{3,6})-\cos(\phi)d_2(\hat{y}_{1,4}+\hat{y}_{3,6}) \\ \mathrm{i}\hat{H}_3 &= -\sin(\phi)d_2(\hat{y}_{2,3}+\hat{y}_{4,5})+\cos(\phi)d_2(\hat{y}_{2,3}+\hat{y}_{4,5}) \end{aligned} \tag{4.2}$$

where we set $\hat{x}_{m,n}=\hat{e}_{m,n}-\hat{e}_{n,m}$, $\hat{y}_{m,n}=\mathrm{i}(\hat{e}_{m,n}+\hat{e}_{n,m})$, and $\hat{e}_{m,n}$ is a square matrix of dimension N (here $N=6$) whose elements are zero except for the mth row and nth column entry, which is one. The coefficients d_1 and d_2 are relative transition strengths, which are determined by the Clebsh-Gordon coefficients [10].

We shall follow convention and indicate a linearly, left circularly and right circularly polarized field by σ, π^- and π^+, respectively. If we apply a control field with one of the polarizations above from time $t=t_0$ to $t=t_1$ then integrating the equation of motion for the evolution operator $\hat{U}_I$ leads to $\hat{U}_I(t_1,t_0)=\hat{U}(C,\phi,*)$ where ϕ is the initial pulse phase and $C=d_*\int_{t_0}^{t_1}A(t)\,dt$ is half the effective pulse area. Concretely, we have $d_*=d_1$ for linearly polarized light, and $d_*=d_2$ for circularly polarized light, as well as:

$$\begin{aligned} \hat{U}_I(C,\phi,\sigma) &= \cos(C)(\hat{z}_{1,2}+\hat{z}_{5,6})+\hat{z}_{3,4} \\ &\quad -\mathrm{i}e^{\mathrm{i}\phi}\sin(C)(\hat{e}_{1,2}-\hat{e}_{5,6})-\mathrm{i}e^{-\mathrm{i}\phi}\sin(C)(\hat{e}_{2,1}-\hat{e}_{6,5}) \end{aligned} \tag{4.3}$$

$$\begin{aligned} \hat{U}_I(C,\phi,\pi^+) &= \cos(C)(\hat{z}_{1,4}+\hat{z}_{3,6})+\hat{z}_{2,5} \\ &\quad -\mathrm{i}e^{\mathrm{i}\phi}\sin(C)(\hat{e}_{1,4}+\hat{e}_{3,6})-\mathrm{i}e^{-\mathrm{i}\phi}\sin(C)(\hat{e}_{4,1}+\hat{e}_{6,3}) \end{aligned} \tag{4.4}$$

$$\begin{aligned} \hat{U}_I(C,\phi,\pi^-) &= \cos(C)(\hat{z}_{2,3}+\hat{z}_{4,5})+\hat{z}_{1,6} \\ &\quad +\mathrm{i}e^{\mathrm{i}\phi}\sin(C)(\hat{e}_{2,3}+\hat{e}_{4,5})+\mathrm{i}e^{-\mathrm{i}\phi}\sin(C)(\hat{e}_{3,2}+\hat{e}_{5,4}) \end{aligned} \tag{4.5}$$

depending on the polarization of the field. For convenience, we have set $\hat{z}_{m,n}=\hat{e}_{m,m}+\hat{e}_{n,n}$.

5 Control scheme for selective population exchanges

Choosing $C=\frac{\pi}{2}$, i.e., control pulses with effective pulse area π leads to

$$\hat{U}_I(\tfrac{\pi}{2},\phi,\sigma) = \hat{z}_{34}-\mathrm{i}e^{\mathrm{i}\phi}(\hat{e}_{1,2}-\hat{e}_{5,6})-\mathrm{i}e^{-\mathrm{i}\phi}(\hat{e}_{2,1}-\hat{e}_{6,5}) \tag{5.6}$$

$$\hat{U}_I(\tfrac{\pi}{2},\phi,\pi^+) = \hat{z}_{2,5}-\mathrm{i}e^{\mathrm{i}\phi}(\hat{e}_{1,4}+\hat{e}_{3,6})-\mathrm{i}e^{-\mathrm{i}\phi}(\hat{e}_{4,1}+\hat{e}_{6,3}) \tag{5.7}$$

$$\hat{U}_I(\tfrac{\pi}{2},\phi,\pi^-) = \hat{z}_{1,6}+\mathrm{i}e^{\mathrm{i}\phi}(\hat{e}_{2,3}+\hat{e}_{4,5})+\mathrm{i}e^{-\mathrm{i}\phi}(\hat{e}_{2,3}+\hat{e}_{4,5}) \tag{5.8}$$

These operators correspond to simultaneous permutations of the populations of levels $|1\rangle$, $|2\rangle$ and $|5\rangle$, $|6\rangle$; $|1\rangle$, $|4\rangle$ and $|3\rangle$, $|6\rangle$; and $|2\rangle$, $|3\rangle$ and $|4\rangle$, $|5\rangle$, respectively.

Assume the initial populations of the lower levels are w_1, w_3 and w_5, respectively, and the populations of the upper levels are zero. Although we know that selective excitation of a sublevel is not possible, we see immediately that we can interchange the populations of any two of the lower sublevels. Let $\hat{P}_1=\hat{U}_I(\frac{\pi}{2},\phi_1,\sigma)$, $\hat{P}_2=\hat{U}_I(\frac{\pi}{2},\phi_2,\pi^+)$ and $\hat{P}_3=\hat{U}_I(\frac{\pi}{2},\phi_3,\pi^-)$. It is easy to check that we have for any choice of the initial pulse phases ϕ_n $(n=1,2,3)$

$$(\hat{P}_1\hat{P}_3\hat{P}_1)\mathrm{diag}(w_1,0,w_3,0,w_5,0)(\hat{P}_1\hat{P}_3\hat{P}_1)^\dagger=\mathrm{diag}(w_3,0,w_1,0,w_5,0), \tag{5.9}$$

i.e., a sequence of three effective π pulses with polarization σ, π^- and σ, respectively, interchanges the populations of the states $|1\rangle$ and $|3\rangle$. Similary, it can be shown that a sequence of three π pulses with polarization σ, π^+ and σ, respectively, interchanges the populations of the states $|3\rangle$ and $|5\rangle$, and a sequence of three π pulses with polarization π^+, π^- and π^+, respectively, interchanges the populations of the states $|1\rangle$ and $|5\rangle$.

6 Creation of arbitrary superposition states

While being able to interchange sublevel populations is interesting, we can do much better than that. Assume that we have prepared the system initially in the pure state $|1\rangle$. Since the system is pure-state controllable, we can prepare any superposition state starting with $|1\rangle$. We shall now a give a general control scheme that allows us to prepare the system in any coherent superposition of the electronic ground states $|1\rangle$, $|3\rangle$ and $|5\rangle$, i.e., the lower sublevels. Assume that we wish to create the superposition state

$$|\Psi\rangle = c_1|1\rangle + c_2|3\rangle + c_3|5\rangle, \qquad |c_1|^2 + |c_2|^2 + |c_3|^2 = 1. \tag{6.10}$$

Note that the coefficients c_n are complex, i.e., $c_n = |c_n|e^{i\theta_n}$ but we may assume $\theta_1 = 0$. Setting

$$C_1 = \arcsin(|c_1|), \quad C_2 = \arcsin(|c_2|/\sqrt{|c_1|^2 + |c_2|^2}), \quad C_3 = \frac{\pi}{2} \tag{6.11}$$

leads to

$$\begin{aligned}
\hat{U}_1 = \hat{U}_I(C_1, \phi_1, \pi^+) &= \cos(C_1)(\hat{z}_{1,4} + \hat{z}_{3,6}) + \hat{z}_{2,5} \qquad (6.12)\\
&\quad -\mathrm{i}e^{\mathrm{i}\phi_1}\sin(C_1)(\hat{e}_{1,4} + \hat{e}_{3,6}) - \mathrm{i}e^{-\mathrm{i}\phi_1}\sin(C_1)(\hat{e}_{4,1} + \hat{e}_{6,3})\\
\hat{U}_2 = \hat{U}_I(C_2, \phi_2, \sigma) &= \cos(C_2)(\hat{z}_{1,2} - \hat{z}_{5,6}) + \hat{z}_{34} \qquad (6.13)\\
&\quad -\mathrm{i}e^{\mathrm{i}\phi_2}\sin(C_2)(\hat{e}_{1,2} - \hat{e}_{5,6}) - \mathrm{i}e^{-\mathrm{i}\phi_2}\sin(C_2)(\hat{e}_{2,1} - \hat{e}_{6,5})\\
\hat{U}_3 = \hat{U}_I(C_3, \phi_3, \pi^-) &= \hat{z}_{1,6} + \mathrm{i}e^{\mathrm{i}\phi_3}(\hat{e}_{2,3} + \hat{e}_{4,5}) + \mathrm{i}e^{-\mathrm{i}\phi_3}(\hat{e}_{3,2} + \hat{e}_{5,4}) \qquad (6.14)
\end{aligned}$$

with $\cos(C_1) = \sqrt{|c_1|^2 + |c_2|^2}$, $\sin(C_1) = |c_3|$, $\cos(C_2) = |c_1|/\sqrt{|c_1|^2 + |c_2|^2}$ and $\sin(C_2) = |c_2|/\sqrt{|c_1|^2 + |c_2|^2}$. Furthermore, if we choose $\phi_1 = -\theta_2$, $\phi_2 = -\theta_1$ and $\phi_3 = 0$ then we have

$$(\hat{U}_3\hat{U}_2\hat{U}_1)\hat{\rho}_0(\hat{U}_3\hat{U}_2\hat{U}_1)^\dagger = \hat{\rho}_1 \tag{6.15}$$

with $\hat{\rho}_0 = |1\rangle\langle 1| = \mathrm{diag}(1,0,0,0,0,0)$ and $\hat{\rho}_1 = |\Psi\rangle\langle\Psi|$. Thus, by a applying a sequence of three pulses with effective pulse areas $2C_1$, $2C_2$ and $2C_3$, initial phases ϕ_1, ϕ_2 and ϕ_3, and polarizations π^+, σ and π^-, respectively, we can create the desired superposition state.

Note that there is a slight problem with the choice of the initial pulse phases. The decomposition above suggests that the phase of the first pulse should be $-\theta_2$. However, since we have used a rotating frame and the phase of the coherences $\rho_{1,4}$ and $\rho_{4,1}$ evolves freely during the application of the second pulse, the phase of the first pulse should really be chosen such that $\phi_1 + \omega_0\Delta T_2 = -\theta_2$ modulo 2π, where ΔT_2 is the length of the second pulse, to ensure that the final superposition state has the correct phase correlation.

7 Conclusion

We have shown how the Lie algebra structure of the control system can be used to derive constructive control schemes for atomic systems with degenerate energy levels. Concretely, we have considered the case of a transition between two three-fold degenerate energy levels, and presented control schemes for selective population interchanges and the creation of arbitrary superposition states. Future work will address transitions with higher degeneracy, and the derivation of constructive control schemes for other control problems of interest.

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