Geometric control for atomic systems

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Introduction

- Topic: Explicit generation of unitary operators for atomic systems using the Lie algebra structure and geometric control.

- Geometric control techniques have been applied successfully to, e.g., coupled spin systems and generic N-level systems with non-degenerate energy levels and transition frequencies.

- Atomic systems are good candidates for geometric control

  - electronic energy levels usually well separated
  - no ro-vibrational states to worry about

  but prior results are not directly applicable since

  - energy levels usually degenerate
  - selective excitation of a single transition often not possible.

  Thus, a generalization of prior techniques is necessary.

Basic facts from atomic quantum theory

If fine / hyperfine structure is negligible:

- The degeneracy of an atomic energy level is $2F + 1$, where F is a quantum number which can take positive integer or half integer values.
- Sublevels of a degenerate energy level are distinguished by a quantum number m, which can take integer or half integer values ranging from $-F$ to F.

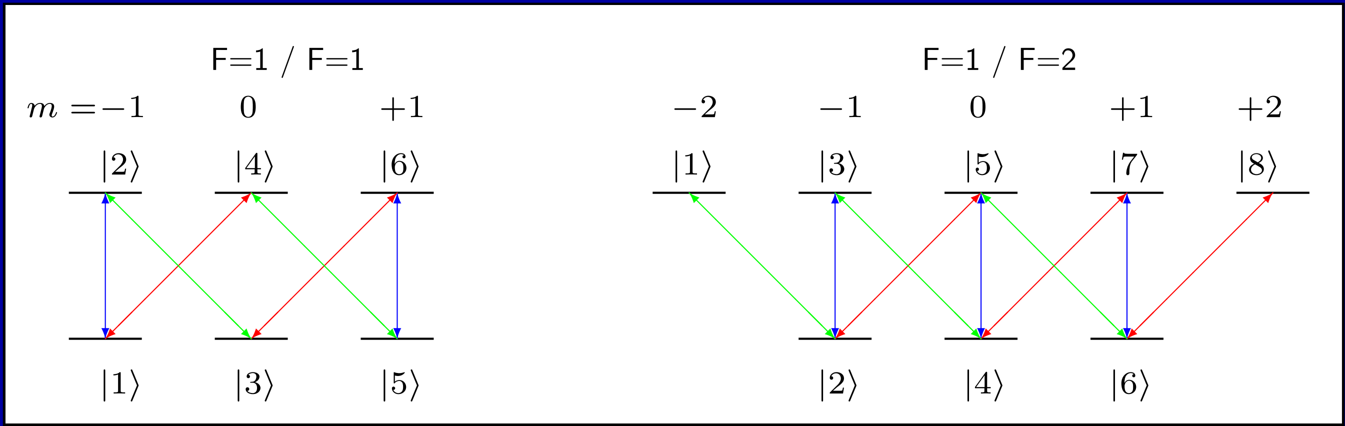
Atomic selection rules:

- Transitions between levels with $|\Delta F| > 1$ prohibited.
- Transitions between sublevels with $|\Delta m| > 1$ prohibited.
- Transition $m = 0$ to $m = 0$ usually prohibited if $\Delta F = 0$.

Interaction of the system with external field of the appropriate frequency depends on polarization of the field.

Some transition diagrams

Transition diagram for $F = 1$ to $F' = 1$ transition (left) and $F = 1$ to $F' = 2$ transition (right):



- A linearly polarized field couples only sublevels with the same quantum number m — blue arrows.
- A left/right circularly polarized field couples only transitions between sublevels with $\Delta m = \pm 1$, respectively — green/red arrows.
- Transitions with same color are simultaneously coupled.

Implications for control of atomic systems

Theory of atomic excitation shows that:

- Selective excitation of a single transition between sublevels by a suitably polarized field is possible only for $F = 0$ / $F' = 0$, and $F = 0$ / $F' = 1$ transitions.
- In all other cases, any control field simultaneously couples multiple sublevels, e.g., for $F = 1$ to $F' = 1$ transitions we always simultaneously excite two sublevels; for $F = 1$ to $F' = 2$ transitions, three, etc.

Degree of controllability for integer values of F:

- Transitions with $\Delta F = \pm 1$ are always mixed-state controllable — Lie algebra type $su(4F + 4)$ or $su(4F)$
- Transitions with $\Delta F = 0$ are only pure-state controllable (if $F > 0$) — Lie algebra type $sp(2F + 1)$.

Constructive Control Schemes

- The case $F = 0$ / $F' = 0$ corresponds to the trivial case of a non-degenerate two-level system, which has been extensively studied.

- The constructive control techniques for generic N-level systems are also applicable to $F = 0$ / $F' = 1$ transitions since, despite the degeneracy of the $F = 1$ level, we can selectively address each possible transition by choosing the polarization of the field.

- For all other cases, however, selective excitation of a single transition between sublevels is not possible, and geometric control techniques for generic N-level systems are thus not directly applicable.

AT CLOSER LOOK AT THE CASE $F=1$ / $F'=1$

- An external field driving a transition between two three-fold degenerate energy levels simultaneously couples two sublevels, no matter which field polarization we choose.

- 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.

- However, it is possible, e.g., to create arbitrary superposition states from a pure initial state, and certain unitary operations for mixed states can also be implemented.

Evolution of the system subject to control

- The time evolution of a pure state $|\Psi(t_0)\rangle$ or a mixed state $\hat{\rho}(t_0)$ is determined by the time evolution operator $\hat{U}(t,t_0)$ via $|\Psi(t)\rangle = \hat{U}(t,t_0)|\Psi(t_0)\rangle$ and $\hat{\rho}(t) = \hat{U}(t,t_0)\hat{\rho}_0\hat{U}(t,t_0)^\dagger$.

- $\hat{U}(t,t_0) = \hat{U}_0(t,t_0)\hat{U}_I(t,t_0)$ where $\hat{U}_0(t,t_0)$ is the free evolution operator and $\hat{U}_I(t,t_0)$ is the dynamical influence of the interaction with the control field.

- If the system is subject to a control field of the form $f(t) = 2A(t)\cos(\omega_0 t + \phi)$ with $\omega_0 = (E_2 - E_1)/\hbar$, and $A(t)$ is slowly varying compared to ω_0, then the RWA leads to

$$\boxed{\dot{U}_I(t,t_0) = A(t)i\hat{H}_I\hat{U}_I(t,t_0)}$$

- The units for time and field strength are ω_0^{-1} and $\hbar\omega_0 p_{12}^{-1}$, respectively, where p_{12} is the dipole moment of the transition.

Interaction Hamiltonian for different field polarizations

The interaction Hamiltonian $\hat{H}_I$ depends on the polarization of the field. If the sublevels are labelled as in the coupling diagram, we have

- For a linearly polarized field
  $i\hat{H}_1 = d\sin(\phi)(-\hat{x}_{1,2} + \hat{x}_{5,6}) - d\cos(\phi)(-\hat{y}_{1,2} + \hat{y}_{5,6})$
- For a right circularly polarized field
  $i\hat{H}_2 = -d\sin(\phi)(\hat{x}_{1,4} + \hat{x}_{3,6}) + d\cos(\phi)(\hat{y}_{1,4} + \hat{y}_{3,6})$
- For a left circularly polarized field
  $i\hat{H}_3 = d\sin(\phi)(\hat{y}_{2,3} + \hat{y}_{4,5}) - d\cos(\phi)(\hat{y}_{2,3} + \hat{y}_{4,5})$

where $d = \frac{1}{\sqrt{2}}$ and we set $\hat{x}_{m,n} = \hat{e}_{m,n} - \hat{e}_{n,m}$, $\hat{y}_{m,n} = i(\hat{e}_{m,n} + \hat{e}_{n,m})$, 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.

Generators of the dynamical evolution

- Following convention, we shall indicate linear, left and right circular polarization by σ, π^- and π^+, respectively.

- If we apply a control field with one of these polarizations from time $t = t_0$ to $t = t_1$, 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 $C = \frac{1}{\sqrt{2}} \int_{t_0}^{t_1} A(t)\, dt$ is half the effective pulse area.

- Concretely, we have $(\hat{z}_{m,n} = \hat{e}_{m,m} + \hat{e}_{n,n})$

$$
\begin{aligned}
\hat{U}_I(C, \phi, \sigma) &= \cos(C)(\hat{z}_{1,2} + \hat{z}_{5,6}) + \hat{z}_{3,4} + i \sin(C) \times \\
&\quad \left[e^{i\phi}(\hat{e}_{1,2} - \hat{e}_{5,6}) + e^{-i\phi}(\hat{e}_{2,1} - \hat{e}_{6,5})\right] \\
\hat{U}_I(C, \phi, \pi^+) &= \cos(C)(\hat{z}_{1,4} + \hat{z}_{3,6}) + \hat{z}_{2,5} + i \sin(C) \times \\
&\quad \left[e^{i\phi}(\hat{e}_{1,4} + \hat{e}_{3,6}) + e^{-i\phi}(\hat{e}_{4,1} + \hat{e}_{6,3})\right] \\
\hat{U}_I(C, \phi, \pi^-) &= \cos(C)(\hat{z}_{2,3} + \hat{z}_{4,5}) + \hat{z}_{1,6} - i \sin(C) \times \\
&\quad \left[e^{i\phi}(\hat{e}_{2,3} + \hat{e}_{4,5}) e^{-i\phi}(\hat{e}_{3,2} + \hat{e}_{5,4})\right]
\end{aligned}
$$

Selective population exchanges

For control pulses with effective pulse area π ($C = \frac{\pi}{2}$) we have

$$\begin{aligned}
\hat{P}_1 &:= \hat{U}_I(\tfrac{\pi}{2}, \phi, \sigma) &= \hat{z}_{34} + ie^{i\phi}(\hat{e}_{1,2} - \hat{e}_{5,6}) + ie^{-i\phi}(\hat{e}_{2,1} - \hat{e}_{6,5}) \\
\hat{P}_2 &:= \hat{U}_I(\tfrac{\pi}{2}, \phi, \pi^+) &= \hat{z}_{2,5} + ie^{i\phi}(\hat{e}_{1,4} + \hat{e}_{3,6}) + ie^{-i\phi}(\hat{e}_{4,1} + \hat{e}_{6,3}) \\
\hat{P}_3 &:= \hat{U}_I(\tfrac{\pi}{2}, \phi, \pi^-) &= \hat{z}_{1,6} - ie^{i\phi}(\hat{e}_{2,3} + \hat{e}_{4,5}) - ie^{-i\phi}(\hat{e}_{2,3} + \hat{e}_{4,5})
\end{aligned}$$

The operators $\hat{P}_k$ 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.

Although selective excitation of any single sublevel is not possible, other selective population exchanges are possible.

Example: Selective population exchanges

If 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, then we can interchange the populations of any two of the lower sublevels:

- A sequence of three π pulses with polarization σ, π^- and σ, respectively, interchanges the populations of the states $|1\rangle$ and $|3\rangle$:

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

  (for any choice of the initial pulse phases ϕ_n).

- A sequence of three π pulses with polarization σ, π^+ and σ, respectively, interchanges the populations of the states $|3\rangle$ and $|5\rangle$;

- A sequence of three π pulses with polarization π^+, π^- and π^+, respectively, interchanges the populations of the states $|1\rangle$ and $|5\rangle$.

Creation of arbitrary superposition states

- Assume we have prepared the system initially in the pure state $|1\rangle$.
- Since the system is pure-state controllable, we can create any superposition state starting with $|1\rangle$.
- We shall now show how to create any coherent superposition $|\Psi\rangle = c_1|1\rangle + c_2|3\rangle + c_3|5\rangle$ of the lower sublevels $|1\rangle$, $|3\rangle$ and $|5\rangle$, i.e., the electronic ground states.
- The coefficients c_n are complex, i.e., $c_n = |c_n|e^{i\theta_n}$ but we may assume $\theta_1 = 0$ and $|c_1|^2 + |c_2|^2 + |c_3|^2 = 1$ for a normalized state.

Creation of arbitrary superposition states

- Choosing the pulse area constants $C_1 = \arcsin(|c_1|)$, $C_2 = \arcsin(|c_2|/\sqrt{|c_1|^2+|c_2|^2})$ and $C_3 = \frac{\pi}{2}$ leads to

$$\begin{aligned}
\hat{U}_1 = \hat{U}_I(C_1,\phi_1,\pi^+) &= \cos(C_1)(\hat{z}_{1,4}+\hat{z}_{3,6})+\hat{z}_{2,5}+i\sin(C_1)\times \\
&\quad \left[e^{i\phi_1}(\hat{e}_{1,4}+\hat{e}_{3,6})+e^{-i\phi_1}(\hat{e}_{4,1}+\hat{e}_{6,3})\right] \\
\hat{U}_2 = \hat{U}_I(C_2,\phi_2,\sigma) &= \cos(C_2)(\hat{z}_{1,2}-\hat{z}_{5,6})+\hat{z}_{34}+i\sin(C_1)\times \\
&\quad \left[e^{i\phi_2}(\hat{e}_{1,2}-\hat{e}_{5,6})+e^{-i\phi_2}(\hat{e}_{2,1}-\hat{e}_{6,5})\right] \\
\hat{U}_3 = \hat{U}_I(C_3,\phi_3,\pi^-) &= \hat{z}_{1,6}-ie^{i\phi_3}(\hat{e}_{2,3}+\hat{e}_{4,5})-ie^{-i\phi_3}(\hat{e}_{3,2}+\hat{e}_{5,4})
\end{aligned}$$

  where $\cos(C_1) = \sqrt{|c_1|^2+|c_2|^2}$, $\sin(C_1) = |c_3|$, and $\cos(C_2) = |c_1|/\sqrt{|c_1|^2+|c_2|^2}$, $\sin(C_2) = |c_2|/\sqrt{|c_1|^2+|c_2|^2}$.

- Choosing furthermore $\phi_1 = -\theta_3$, $\phi_2 = -\theta_2$ and $\phi_3 = 0$ gives

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

  for the initial state $\hat{\rho}_0 = |1\rangle\langle 1| = \mathrm{diag}(1,0,0,0,0,0)$ and the target state $\hat{\rho}_1 = |\Psi\rangle\langle\Psi|$.

Creation of arbitrary superposition states

- Thus, the decomposition suggests that applying a sequence of three pulses with effective pulse areas

$$2\arcsin(|c_1|),\ 2\arcsin(|c_2|/\sqrt{|c_1|^2+|c_2|^2}) \text{ and } \pi,$$

  initial phases $-\theta_3$, $-\theta_2$ and 0, and polarizations π^+, σ and π^-, respectively, creates the desired superposition state.

- 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

$$\phi_1 + \omega_0 \Delta T_2 = -\theta_3 \text{ modulo } 2\pi,$$

  where ΔT_2 is the length of the second pulse, to ensure that the final superposition state has the correct phase correlation.

Conclusion

Summary:

- We have shown how to use atomic selection rules to determine the generators of the dynamical Lie algebra for atomic systems with degenerate energy levels, and how to use the dynamical generators to derive constructive control schemes for such systems.

- As an example we considered the case of a transition between two three-fold degenerate energy levels, and presented control schemes for selective population exchanges and the creation of arbitrary superpositions of the ground state levels.

Future work:

- Constructive control schemes for transitions with higher degeneracy

- Derivation of constructive control schemes for arbitrary unitary operators in dynamical Lie group.