Multi-polarization control of rotational motion through dipole coupling

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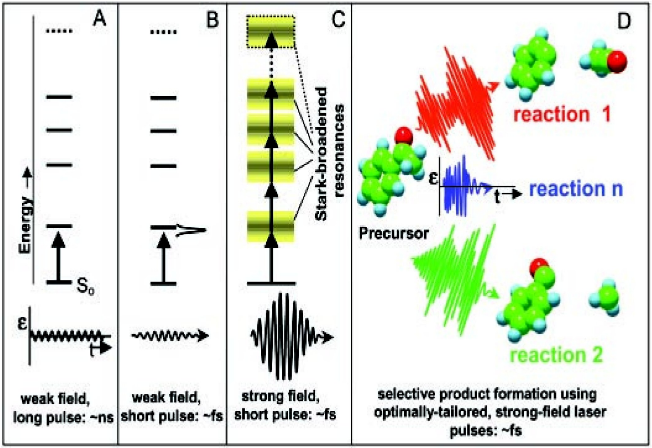
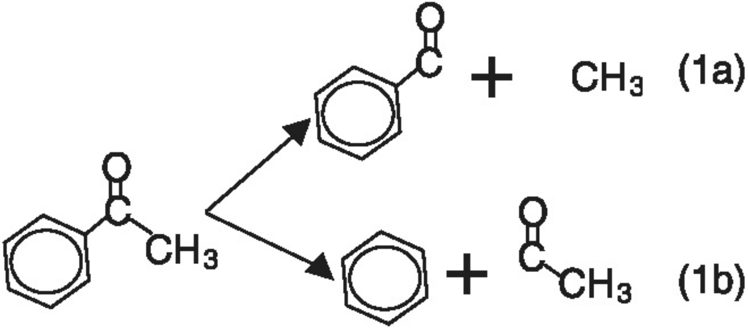
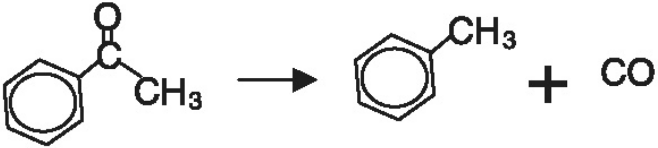


Figure: R. J. Levis, G.M. Menkir, and H. Rabitz. *Science*, 292:709–713, 2001



Scheme 1.

Figure: SELECTIVE dissociation of chemical bonds (laser induced).
Other examples: CF_3 or CH_3 from CH_3COCF_3 ...
(R. J. Levis, G.M. Menkir, and H. Rabitz. *Science*, 292:709–713, 2001).



Scheme 2.

Figure: Selective dissociation AND CREATION of chemical bonds (laser induced).
Other examples: CF_3 or CH_3 from CH_3COCF_3 ...
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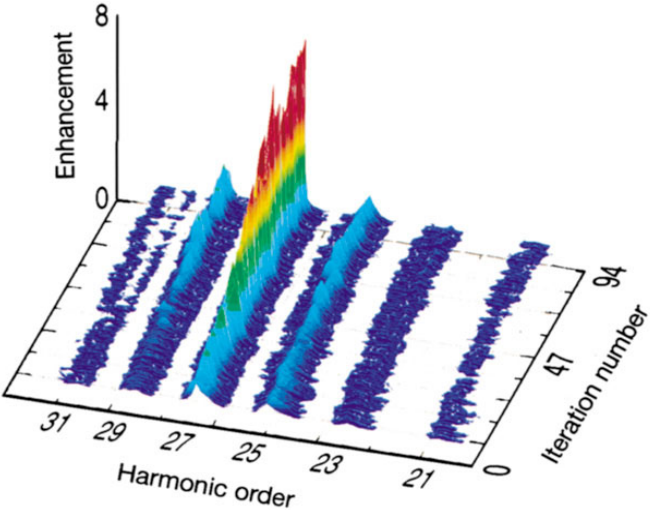


Figure: Experimental High Harmonic Generation (argon gas) obtain high frequency lasers from lower frequencies input pulses $\omega \rightarrow n\omega$ (electron ionization that come back to the nuclear core) (R. Bartels et al. Nature, 406, 164, 2000).

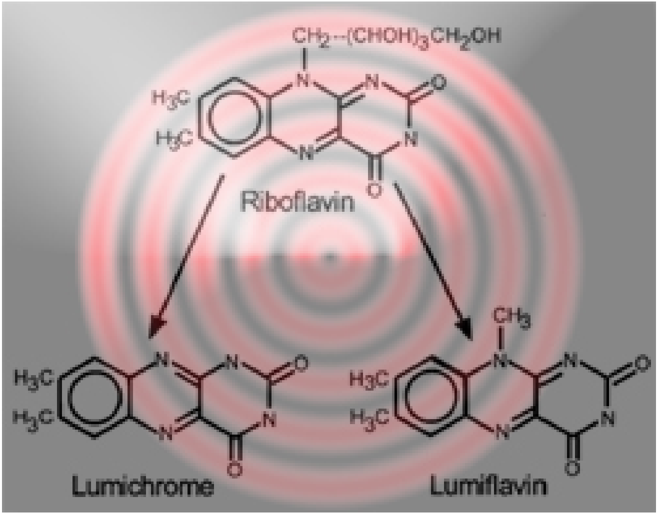


Figure: Studying the excited states of proteins. F. Courvoisier et al., App.Phys.Lett.

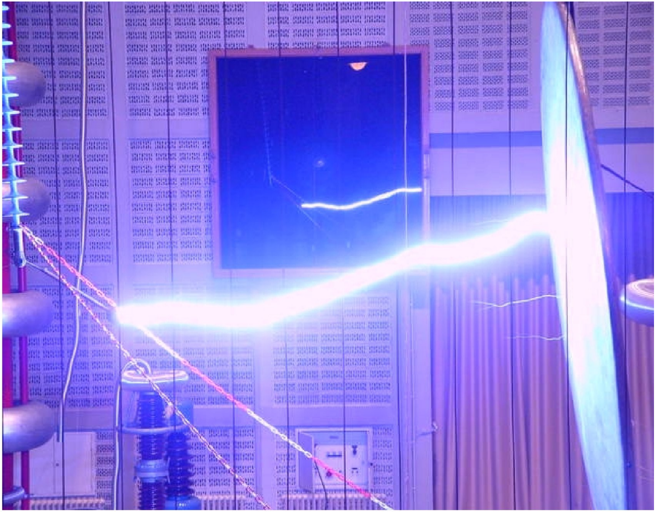


Figure: thunder control : experimental setting ; J. Kasparian Science, 301, 61 – 64 team of J.P.Wolf @ Lyon / Geneve , ...

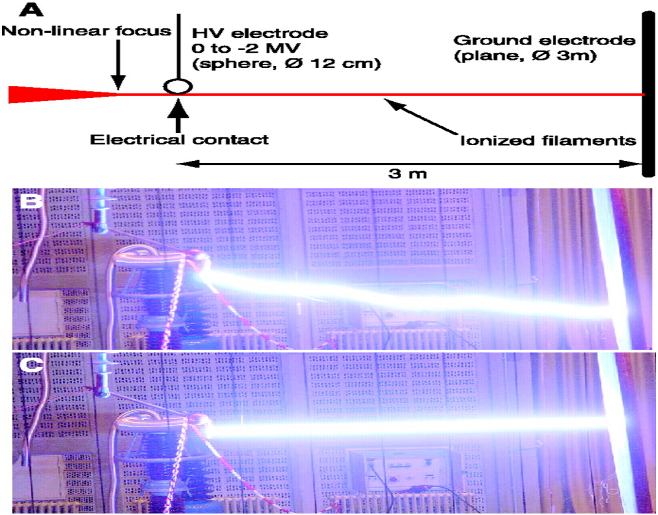


Figure: thunder control : (B) random discharges ; (C) guided by a laser filament ; J. Kasparian Science, 301, 61 – 64 team of J.P.Wolf @ Lyon / Geneve , ...

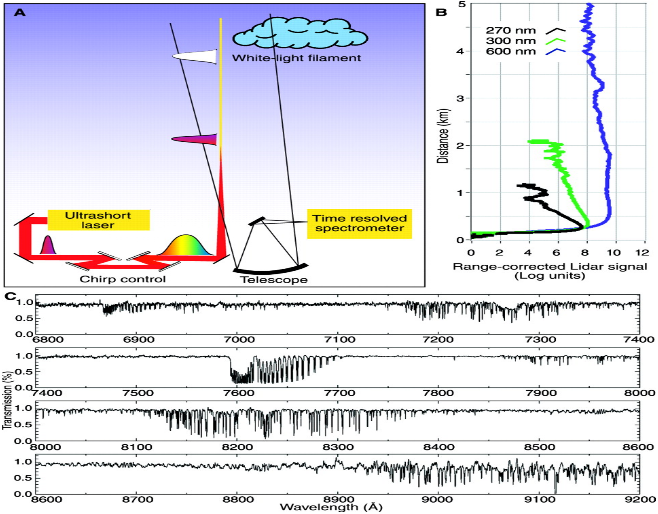


Figure: LIDAR = atmosphere detection; the pulse is tailored for an optimal reconstruction at the target : 20km = OK ! ; J. Kasparian Science, 301, 61 – 64

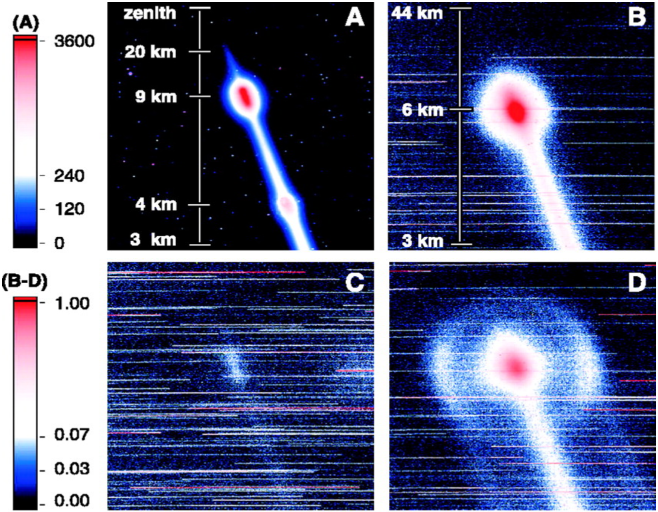


Figure: Creation of a white light of high intensity and spectral width ; J. Kasparian Science, 301, 61 – 64

Other applications

- EMERGENT technology
- creation of particular molecular states
- long term: logical gates for quantum computers
- fast "switch" in semiconductors
- ...

Single quantum system, bilinear control

Time dependent Schrödinger equation

$$\begin{cases} i\frac{\partial}{\partial t}\Psi(x,t) = H_0\Psi(x,t) \\ \Psi(x,t=0) = \Psi_0(x). \end{cases} \tag{1}$$

Add external BILINEAR interaction (e.g. laser)

$$\begin{cases} i\frac{\partial}{\partial t}\Psi(x,t) = (H_0 - \epsilon(t)\mu(x))\Psi(x,t) \\ \Psi(x,t=0) = \Psi_0(x) \end{cases} \tag{2}$$

Ex.: $H_0 = -\Delta + V(x)$, unbounded domain
Evolution on the unit sphere: $\|\Psi(t)\|_{L^2} = 1, \ \forall t \geq 0.$

Controllability

A system is controllable if for two arbitrary points Ψ_1 and Ψ_2 on the unit sphere (or other ensemble of admissible states) it can be steered from Ψ_1 to Ψ_2 with an admissible control.

Norm conservation : controllability is equivalent, up to a phase, to say that the projection to a target is $= 1$.

Galerkin discretization of the Time Dependent Schrödinger equation

$$i\frac{\partial}{\partial t}\Psi(x,t) = (H_0 - \epsilon(t)\mu)\Psi(x,t)$$

- basis functions $\{\psi_i; i = 1, ..., N\}$, e.g. the eigenfunctions of the H_0: $\psi_k = e_k\psi_k$

- wavefunction written as $\Psi = \sum_{k=1}^{N} c_k\psi_k$

- We will still denote by H_0 and μ the matrices $(N \times N)$ associated to the operators H_0 and μ : $H_{0\,kl} = \langle\psi_k|H_0|\psi_l\rangle$, $\mu_{kl} = \langle\psi_k|\mu|\psi_l\rangle$,

Lie algebra approaches

To assess controllability of

$$i\frac{\partial}{\partial t}\Psi(x,t) = (H_0 - \epsilon(t)\mu)\Psi(x,t)$$

construct the "dynamic" Lie algebra $L = Lie(-iH_0, -i\mu)$:

$$\begin{cases} \forall M_1, M_2 \in L,\ \forall \alpha, \beta \in \boldsymbol{R} : \alpha M_1 + \beta M_2 \in L \\ \forall M_1, M_2 \in L, [M_1, M_2] = M_1 M_2 - M_2 M_1 \in L \end{cases}$$

Theorem If the group e^L is compact any $e^M \psi_0$, $M \in L$ can be attained.

"Proof" $M = -iAt$: trivial by free evolution

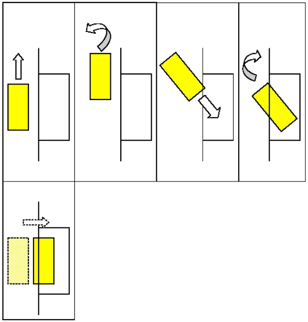
Trotter formula:

$$e^{i(AB-BA)} = \lim_{n\to\infty}\left[e^{-iB/\sqrt{n}}e^{-iA/\sqrt{n}}e^{iB/\sqrt{n}}e^{iA/\sqrt{n}}\right]^n$$

Operator synthesis (“lateral parking”)

$$\text{Trotter formula: } e^{i[A,B]} = \lim_{n\to\infty}\left[e^{-iB/\sqrt{n}}e^{-iA/\sqrt{n}}e^{iB/\sqrt{n}}e^{iA/\sqrt{n}}\right]^n$$

$e^{\pm iA}$ = advance/reverse ; $e^{\pm iB}$ = turn left/right



Corollary. If $L = u(N)$ or $L = su(N)$ (the (null-traced) skew-hermitian matrices) then the system is controllable.

"Proof" For any Ψ_0, Ψ_T there exists a "rotation" U in $U(N) = e^{u(N)}$ (or in $SU(N) = e^{su(N)}$) such that $\Psi_T = U\Psi_0$.

• (Albertini & D'Alessandro 2001) Controllability also true for L isomorphic to $sp(N/2)$ (unicity).

$sp(N/2) = \{M : M^* + M = 0, M^t J + JM = 0\}$ where J is a matrix unitary equivalent to $\begin{pmatrix} 0 & I_{N/2} \\ -I_{N/2} & 0 \end{pmatrix}$ and $I_{N/2}$ is the identity matrix of dimension $N/2$

Outline

Physical picture

• linear rigid molecule, Hamiltonian $H = B\hat{J}^2$, $B =$ rotational constant, $\hat{J} =$ angular momentum operator.
• control= electric field $\overrightarrow{\epsilon(t)}$ by the dipole operator $\overrightarrow{d}$. Field $\overrightarrow{\epsilon(t)}$ is multi-polarized i.e. x, y, z components tuned independently

Time dependent Schrödinger equation ($\theta, \phi =$ polar coordinates):

$$i\hbar \frac{\partial}{\partial t}|\psi(\theta,\phi,t)\rangle = (B\hat{J}^2 - \overrightarrow{\epsilon(t)} \cdot \overrightarrow{d})|\psi(\theta,\phi,t)\rangle \tag{3}$$
$$|\psi(0)\rangle = |\psi_0\rangle, \tag{4}$$

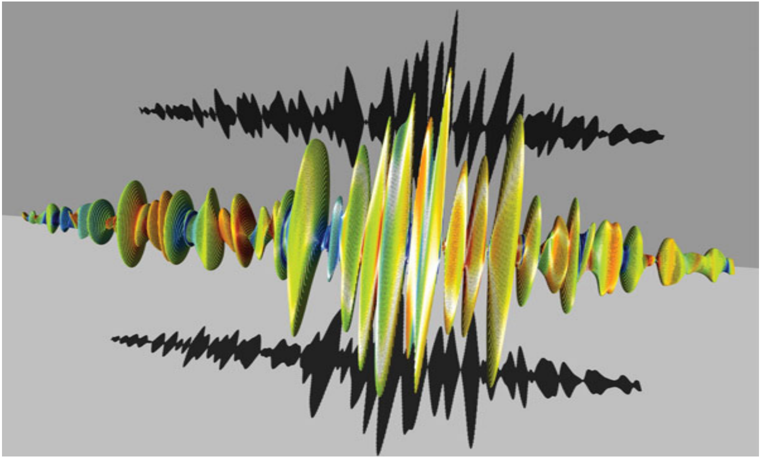


Figure: Elliptically polarized laser and its two components (from T. Brixner & G. Gerber, Opt. Lett 26(557) 2001.). See also Hertz et al. PRA 76(043423) 2007 for control of rotation.

Discretization

Eigenbasis decomposition of $B\hat{J}^2$ with spherical harmonics ($J \geq 0$ and $-J \leq m \leq J$):

$B\hat{J}^2|Y_J^m\rangle = E_J|Y_J^m\rangle$,

$E_J = BJ(J+1)$. highly degenerate !

Note $E_{J+1} - E_J = 2B(J+1)$, we truncate : $J \leq J_{max}$.

Refs: G.T. H. Rabitz : J Phys A (to appear), preprint http://hal.archives-ouvertes.fr/hal-00450794/en/

Dipole interaction

Dipole in space fixed cartesian coordinates
$\overrightarrow{\epsilon(t)} \cdot \overrightarrow{d} = \epsilon_x(t)x + \epsilon_y(t)y + \epsilon_z(t)z$ $\overrightarrow{x}$, $\overrightarrow{y}$ and $\overrightarrow{z}$,
components $\epsilon_x(t), \epsilon_y(t), \epsilon_z(t) =$ independent.

Using as basis the $J = 1$ spherical harmonics

$$Y_1^{\pm 1} = \frac{\mp 1}{2}\sqrt{\frac{3}{2\pi}}\frac{x \pm iy}{r}, \quad Y_1^0 = \frac{1}{2}\sqrt{\frac{3}{\pi}}\frac{z}{r}, \tag{5}$$

We obtain $\overrightarrow{\epsilon(t)} \cdot \overrightarrow{d} = \epsilon_0(t)d_{10}Y_1^0 + \epsilon_{+1}(t)d_{11}Y_1^1 + \epsilon_{-1}(t)d_{1-1}Y_1^{-1}$.
After rescaling

$$\overrightarrow{\epsilon(t)} \cdot \overrightarrow{d} = \epsilon_0(t)Y_1^0 + \epsilon_{+1}(t)Y_1^1 + \epsilon_{-1}(t)Y_1^{-1}. \tag{6}$$

Discretization

D_k = matrix of Y_1^k $(k = -1, 0, 1)$. Entries:

$$(D_k)_{(Jm),(J'm')} = \langle Y_J^m | Y_1^k | Y_{J'}^{m'} \rangle = \int (Y_J^m)^*(\theta,\phi) Y_1^k(\theta,\phi) Y_{J'}^{m'}(\theta,\phi) \sin(\theta) d\theta d$$

$$= \sqrt{\frac{3(2J+1)(2J'+1)}{4\pi}} \begin{pmatrix} J & 1 & J' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J & 1 & J' \\ m & k & m' \end{pmatrix}.$$

$\begin{pmatrix} J & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}$ and $\begin{pmatrix} J & 1 & J' \\ m & k & m' \end{pmatrix}$ = Wigner 3J-symbols

Entries are zero except when $m + k + m' = 0$ and $|J - J'| = 1$.

Discrete TDSE

$\Psi(t)$= coefficients of $\psi(\theta, \phi, t)$ with respect to the spherical harmonic basis

$$\begin{cases} i\frac{\partial}{\partial t}\Psi(t) = (E - \epsilon_0(t)D_0 + \epsilon_{-1}(t)D_{-1} + \epsilon_1(t)D_1)\Psi(t) \\ \Psi(t=0) = \Psi_0. \end{cases} \tag{8}$$

E = diagonal matrix with entries E_J for all Jm, $-J \leq m \leq J$, $J \leq J_{max}$.

Coupling structure

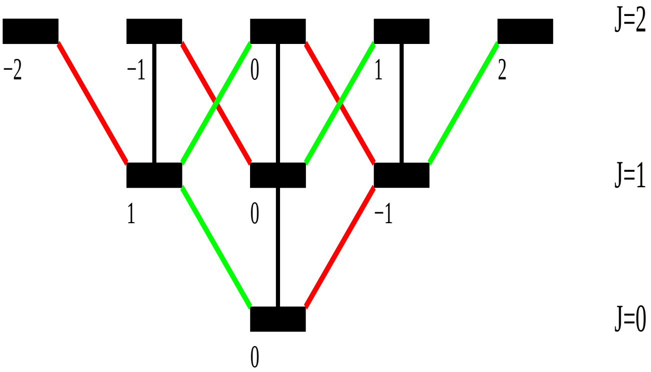


Figure: The three matrices D_k, $k = -1, 0, 1$ coupling the eigenstates are each represented by a different color (green, black, red) for $J_{max} = 2$. On the J-th line from bottom, the states are from left to right in order $|Y_J^{m=-J}\rangle, \ldots, |Y_J^{m=J}\rangle$ for even values of J and $|Y_J^{m=J}\rangle, \ldots, |Y_J^{m=-J}\rangle$ for odd values of J. The m quantum number labelings are indicated in the figure.

Outline

Controllability with 3 fields

Theorem (GT, H.Rabitz '10)

Let $J_{max} \geq 1$ *and denote* $N = (J_{max}+1)^2$. *Let* E, D_k, $k = -1, 0, 1$ *be* $N \times N$ *matrices indexed by* Jm *with* $J = 0, ..., J_{max}$, $|m| \leq J$ *where:*

$$E_{Jm;J'm'} = \delta_{JJ'}\delta_{mm'}E_J \tag{9}$$

$$(D_0)_{Jm,J'm'} \neq 0 \Leftrightarrow |J-J'| = 1,\ m+m' = 0 \tag{10}$$

$$(D_1)_{Jm,J'm'} \neq 0 \Leftrightarrow |J-J'| = 1,\ m+m'+1 = 0 \tag{11}$$

$$(D_{-1})_{Jm,J'm'} \neq 0 \Leftrightarrow |J-J'| = 1,\ m+m'-1 = 0. \tag{12}$$

and recall that

$$E_J = J(J+1). \tag{13}$$

Then the system described by E, D_{-1}, D_0, D_1 *is controllable.*

Controllability with 3 fields

Proof Idea: construct the Lie algebra spanned by iE, iD_k; begin by first iterating the commutators, obtain generators for any transition (degenerate); then combine the results using the coupling structure.

Controllability with 3 fields

Theorem (GT, H.R '10)

Consider a finite dimensional system expressed in an eigenbasis of its internal Hamiltonian E with eigenstates indexed $a = (Jm)$ *with* $J = 0, \ldots, J_{max}$, $m = 1, \ldots, m_J^{max}$, $m_0^{max} = 1$, *and such that*

$$E_{Jm;J'm'} = \delta_{JJ'}\delta_{mm'}E_J, \quad E_{J+1} - E_J \neq E_{J'+1} - E_{J'}, \forall J \neq J'. \quad (14)$$

Consider K coupling matrices D_k, $k = 1, \ldots, K$ *such that*

$$(D_k)_{(Jm),(J'm')} \neq 0 \Rightarrow |J - J'| = 1 \quad (15)$$

$$(D_k)_{(Jm),(J'm')} \neq 0, (D_k)_{(Jm),(J''m'')} \neq 0, \ J \leq J' \leq J'' \Rightarrow J' = J'', m' = m'' \quad (16)$$

If the graph of the system is connected then the system is controllable.

Controllability with 3 fields

Remark

The results can be extended to the case of a symmetric top molecule; the energy levels are described by three quantum numbers E_{JKm} *with* $|m| \leq J$, $|K| \leq J$ *and*

$$E_{JKm} = C_1 J(J+1) + C_2 K^2, \qquad (17)$$

(for some constants C_1 *and* C_2*); if the initial state is in the ground state, or any other state with* $K = 0$ *the coupling operators have the same structure as in Thm. 3.1 and thus any linear combination of eigenstates with quantum numbers* $J, K = 0, m$ *can be reached (same result directly applies).*

Outline

Controllability with fixed linear combination

What if $\epsilon_k(t)$, $k = -1, 0, 1$ are not chosen independently but with a locked linear dependence through coefficients α_k :
$\overrightarrow{\epsilon(t)} \cdot \overrightarrow{d} = \epsilon(t)\{\alpha_{-1} Y_1^{-1} + \alpha_0 Y_1^0 + \alpha_1 Y_1^1\}$.
There exist non-controllable cases for **any** given linear combination:

$$E = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & 2 \end{pmatrix}, \ \overrightarrow{e(t)} \cdot \overrightarrow{d} = \epsilon(t)\mu, \ \mu = \begin{pmatrix} 0 & \alpha_{-1} & \alpha_0 & \alpha_1 \\ \alpha_{-1} & 0 & 0 & 0 \\ \alpha_0 & 0 & 0 & 0 \\ \alpha_1 & 0 & 0 & 0 \end{pmatrix}$$

This system is such that for all α_k ($k = -1, 0, 1$) the Lie algebra generated by iE and $i\mu$ is $u(2)$, thus the system is not controllable with one laser field (but ok with 3).

Controllability with fixed linear combination

Theorem

Let $A, B_1, \ldots, B_K$ be elements of a finite dimensional Lie algebra L. For $\alpha = (\alpha_1, \ldots, \alpha_K) \in \mathbb{R}^K$ we denote L_α as the Lie algebra generated by A and $B_\alpha = \sum_{k=1}^{K} \alpha_k B_k$. Define the maximal dimension of L_α

$$d^1_{A,B_1,\ldots,B_K} = \max_{\alpha \in \mathbb{R}^K} dim_{\mathbb{R}}(L_\alpha). \tag{19}$$

Then with probability one with respect to α, $dim(L_\alpha) = d^1_{A,B_1,\ldots,B_K}$.

Remark

The dimension $d^1_{A,B_1,\ldots,B_K}$ is specific to the choice of coupling operators B_k (easily computed).

Controllability with fixed linear combination

Proof. List of all possible iterative commutators constructed from A and B_α:

$$\mathcal{C}^\alpha = \{\zeta_1^\alpha = A, \zeta_2^\alpha = B, \zeta_3^\alpha = [A, B_\alpha], \zeta_4^\alpha = [B_\alpha, A], \zeta_5^\alpha = [A, [A, B_\alpha]], ...\}. \tag{20}$$

Note : $\zeta_{i_1}^\alpha, ..., \zeta_{i_r}^\alpha$= linearly independent $\iff$ Gram determinant is non-null (analytic criterion of α);
One of the following alternatives is true:
- either this function is identically null for all α (which is the case e.g., for $\{\zeta_3^\alpha, \zeta_4^\alpha\}$)
- or it is non-null everywhere with the possible exception of a zero measure set.

Let $\mathcal{F}$ dense in $\mathbb{R}^K$ such that if $\zeta_{i_1}^\alpha, ..., \zeta_{i_r}^\alpha$ are linearly independent for one value of $\alpha \in \mathbb{R}^K$ then they are linearly independent for all $\alpha' \in \mathcal{F}$.

Controllability with fixed linear combination

Denote by $\alpha^\star$ some value such that $dim_{\mathbb{R}}(L_{\alpha^\star}) = d^1_{A,B_1,...,B_K}$; then there exists a set such that $\{\zeta^{\alpha^\star}_{i_1}, ..., \zeta^{\alpha^\star}_{i_{d^1_{A,B_1,...,B_K}}}\}$ are linearly independent; thus $\{\zeta^{\alpha^\star}_{i_1}, ..., \zeta^{\alpha^\star}_{i_{d^1_M}}\}$ linearly independent for any $\alpha \in \mathcal{F}$; thus $dim_{\mathbb{R}}(L_\alpha) \geq d^1_{A,B_1,...,B_K}$ for all $\alpha \in \mathcal{F}$, q.e.d. (maximality of $d^1_{A,B_1,...,B_K}$).

Remark

In numerical tests the Lie algebra generated by iE and $iD_\alpha = i\sum_{k=-1}^{1} \alpha_k D_k$. always had dimension $(N-2)^2$; can this be proved ???

Open question: the algebras for $\alpha \in \mathbb{R}^K$ are isomorphic to subalgebras of the Lie algebra with maximal dimension ?

Outline

- Background on controllability criteria

1. Control of rotational motion
   - Physical picture
2. Controllability assessment with three independently polarized field components
3. Controllability for a locked combination of lasers
4. Controllability with two lasers
   - Field shaped in the $\vec{z}$ and $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ directions
   - Field shaped in the $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ and $\frac{\vec{x}-i\vec{y}}{\sqrt{2}}$ directions

Controllability with two lasers

Theorem

Let $A, B_1, \ldots, B_K$ be elements of a finite dimensional Lie algebra L. We denote for $\alpha = (\alpha_1, \ldots, \alpha_K) \in \mathbb{R}^K$ and $\beta = (\beta_1, \ldots, \beta_K) \in \mathbb{R}^K$ by $L_{\alpha,\beta}$ the Lie algebra generated by A, $B_\alpha = \sum_{k=1}^K \alpha_k B_k$ and $B_\beta = \sum_{k=1}^K \beta_k B_k$.
Define the maximal dimension of L_α

$$d^2_{A,B_1,\ldots,B_K} = \max_{\alpha \in \mathbb{R}^K} dim_{\mathbb{R}}(L_{\alpha,\beta}). \tag{21}$$

Then with probability one with respect to α, β, $dim(L_{\alpha,\beta}) = d^2_M$.

Field shaped in the $\vec{z}$ and $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ directions

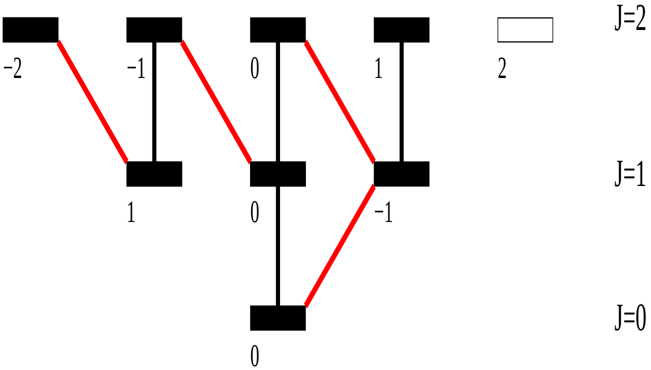


Figure: Field shaped in the $\vec{z}$ and $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ directions, same conventions apply. The $\epsilon_{-1} = 0$, the coupling realized by the operator D_{-1} disappears and the state $|Y_{J_{max}}^{m=J_{max}}\rangle$ is not connected with the others. In particular the population in state $|Y_{J_{max}}^{m=J_{max}}\rangle$ cannot be changed by the two lasers and thus will be a conserved quantity.

Field shaped in the $\vec{z}$ and $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ directions

Theorem

Consider the model of Thm.3.1 with $\epsilon_{-1} = 0$. Let $|\psi_I\rangle$ and $|\psi_F\rangle$ be two states that have the same population in $|Y_{J_{max}}^{m=J_{max}}\rangle$ i.e., $|\langle\psi_I, Y_{J_{max}}^{m=J_{max}}\rangle|^2 = |\langle\psi_F, Y_{J_{max}}^{m=J_{max}}\rangle|^2$. Then $|\psi_F\rangle$ can be reached from $|\psi_I\rangle$ with controls $\epsilon_0(t)$ and $\epsilon_1(t)$.

Similar analysis applies for $\vec{z}$ and $\frac{\vec{x}-i\vec{y}}{\sqrt{2}}$ directions; the population of $|Y_{J_{max}}^{m=-J_{max}}\rangle$ is conserved and the compatibility relation reads:

$$|\langle\psi_I, Y_{J_{max}}^{m=-J_{max}}\rangle|^2 = |\langle\psi_F, Y_{J_{max}}^{m=-J_{max}}\rangle|^2. \tag{22}$$

Field shaped in the $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ and $\frac{\vec{x}-i\vec{y}}{\sqrt{2}}$ directions

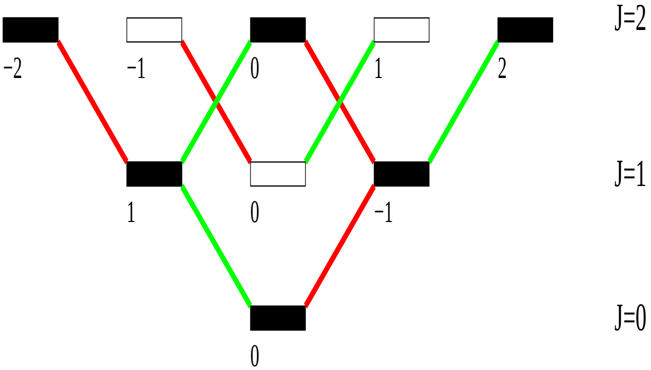


Figure: Field shaped in the $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ and $\frac{\vec{x}-i\vec{y}}{\sqrt{2}}$ directions. Two connectivity sets appear: $X_1 = \{|Y_0^0\rangle, |Y_1^{\pm 1}\rangle, |Y_2^{\pm 2}\rangle, |Y_2^0\rangle, \ldots\}$ connected with $|Y_0^0\rangle$ (filled black rectangles) and $X_2 = \{|Y_1^0\rangle, |Y_2^{\pm 1}\rangle, |Y_3^{\pm 2}\rangle, |Y_3^0\rangle, \ldots\}$. connected with $|Y_1^0\rangle$ (empty rectangles).

Field shaped in the $\frac{\vec{x}+i\vec{y}}{\sqrt{2}}$ and $\frac{\vec{x}-i\vec{y}}{\sqrt{2}}$ directions

Conservation law

$$\sum_{|Y_J^m\rangle \in X_1} |\langle \psi_I, Y_J^m \rangle|^2 = \sum_{|Y_J^m\rangle \in X_1} |\langle \psi_F, Y_J^m \rangle|^2. \tag{23}$$

Theorem

Consider the model of the Thm.3.1 with $\epsilon_{-1} = 0$. Let $|\psi_I\rangle$ and $|\psi_F\rangle$ be two states compatible in the sense of Eqn. (23). Then $|\psi_F\rangle$ can be reached from $|\psi_I\rangle$ with controls $\epsilon_{-1}(t)$ and $\epsilon_1(t)$.

Proof Construct Lie algebra for each laser then use the controllability criterion for independent systems.