Quantum control of Rydberg atoms using impulsive control fields

[3D Schroedinger equation with Coulomb potential controlled by (time) delta function controls]

Chitra Rangan

Department of Physics, University of Windsor

Windsor, Ontario, Canada

Windsor, Ontario
Canada
- The rose city

Framework

- System: Rydberg wave packet (RWP):

  Superposition of energy eigenstates exhibiting Keplerian motion about a nucleus (-1/r potential). Dynamical time (drift/field-free evolution) ~10ps.

- Control: Terahertz half-cycle pulse (HCP):

  Approximately an impulse. Controls (~1ps) are much faster than the drift.

  D. You, R. R. Jones, D.R. Dykaar, and P.H. Bucksbaum, Optics Letters 18, 290 (1993).

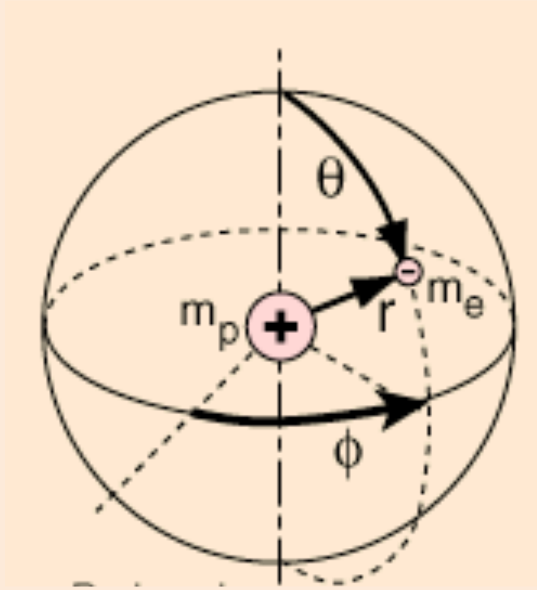
- Decoherence:

  Negligible sources of decoherence. Coherence time ~6ns. Coherent evolution.

Outline

- Introduction to RWPs & HCPs
- Case 1: Control with single HCP
  - Optimal control
- If time permits… Control with two kicks
- Case 2a: Control can be treated semiclassically
  - New method for determining the width of an RWP
- Case 2b: Control can only be treated quantum mechanically
  - Removing & inserting coherences from a subspace

3D Coulomb problem



$$i\dot{\psi} = -\frac{\nabla^2\psi}{2} - \frac{1}{r}\psi$$

- Spherical coordinates
- Separation of variables

$$\Psi(r,\theta,\phi) = R(r)P(\theta)F(\phi)$$

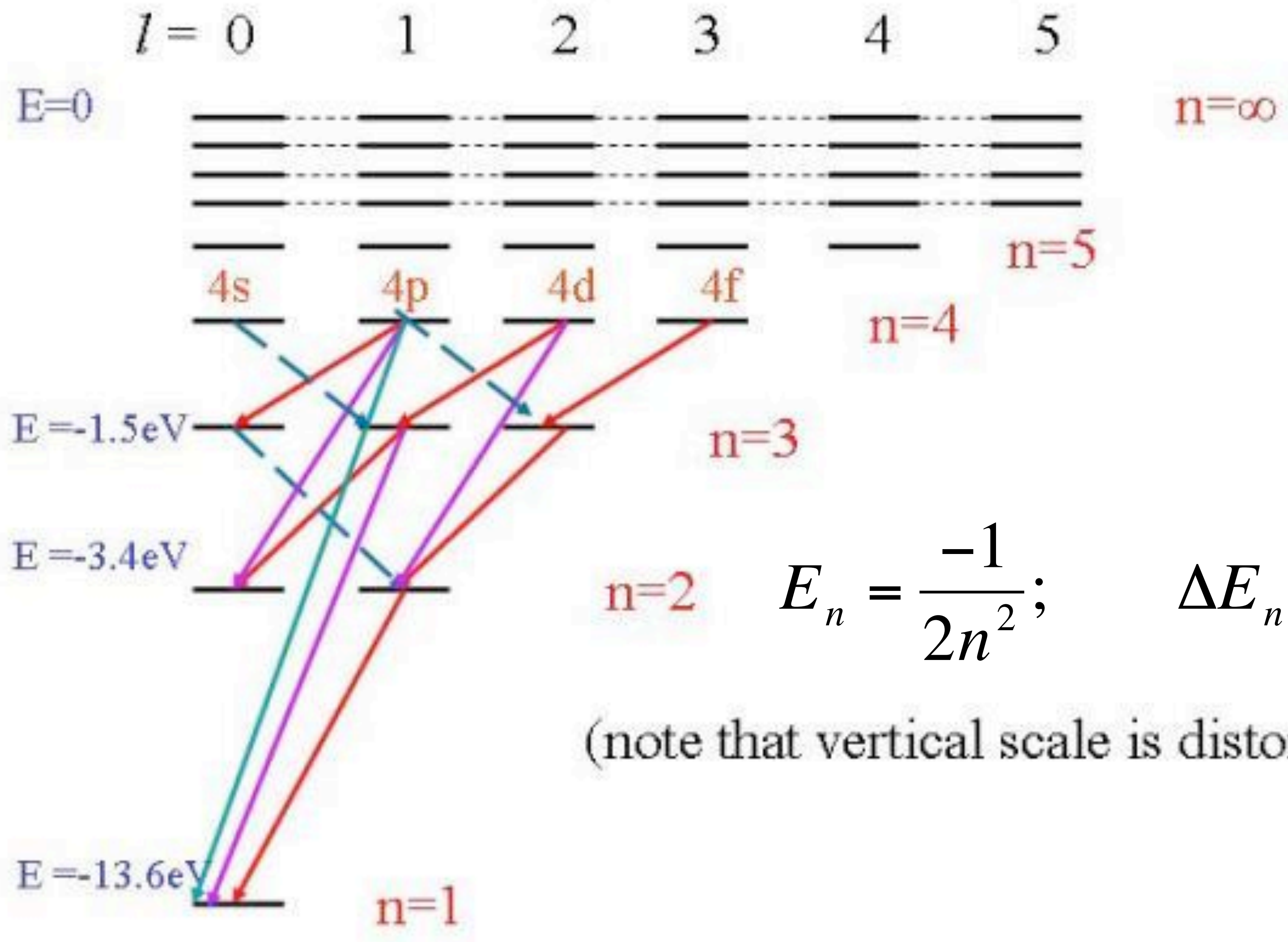
- Energy eigenstates:

$$\psi_{nlm}(r,\theta,\phi) = R_{nl}(r)P_l(\theta)F_m(\phi)$$

~ Laguerre polynomials **Spherical harmonics Y_{lm}**

$$H\psi_{nlm}(r,\theta,\phi) = E_n\psi_{nlm}(r,\theta,\phi) = \frac{1}{2n^2}\psi_{nlm}(r,\theta,\phi)$$

Energy level diagram



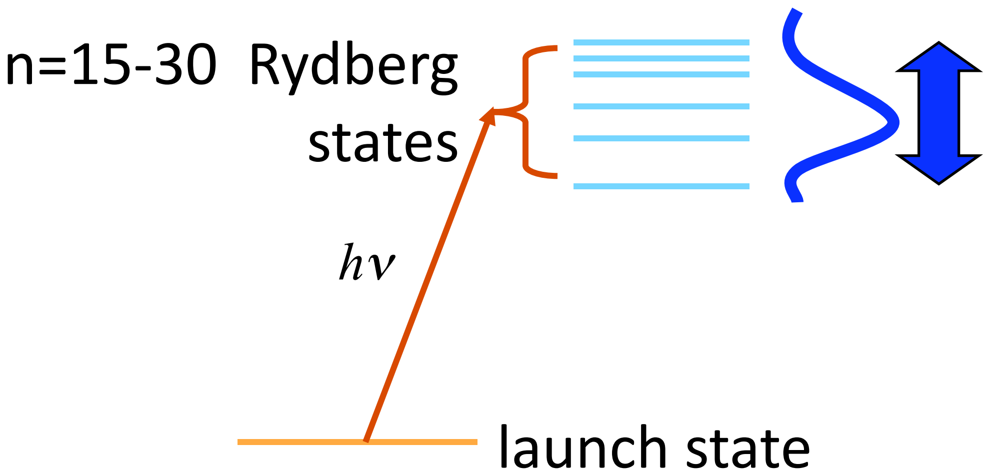
$$E_n = \frac{-1}{2n^2}; \qquad \Delta E_n = \frac{1}{n^3}$$

(note that vertical scale is distorted)

Rydberg Atom Wave Packet

The state vector is a coherent superposition of highly excited states of a one electron atom – a Rydberg wave packet

$$|\psi\rangle = \sum_n c_n \, |\phi_n\rangle$$



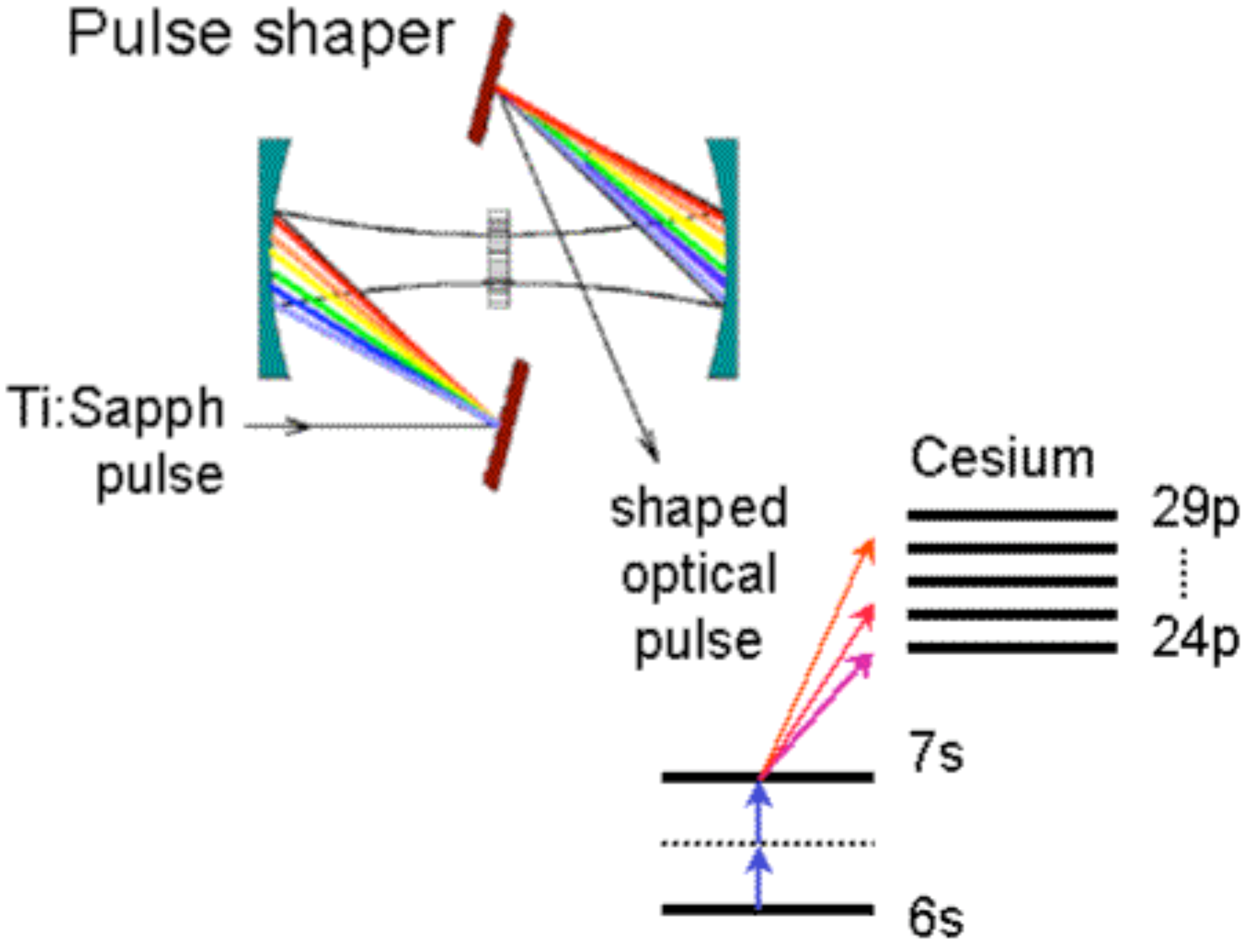
The energy level spacings are ~ THz

Dynamical time scales ~ 10 ps

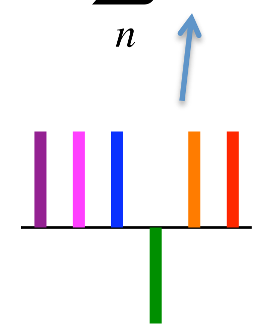
$$|\psi(t)\rangle = \sum_n c_n e^{-iE_n t} \, |\phi_n\rangle$$

Typical states: n~25 states of an alkali atom

Sculpting a Rydberg wave packet

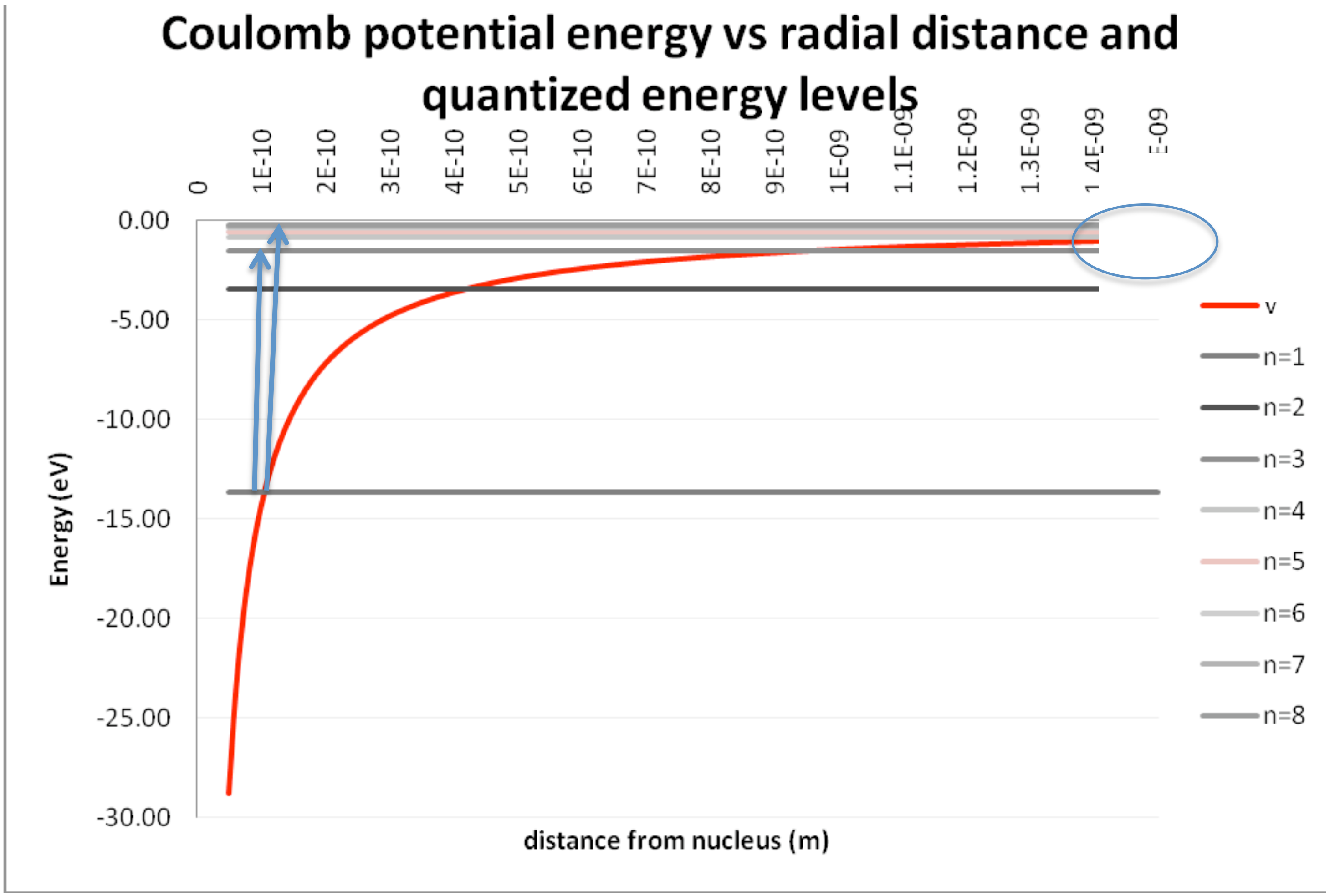


$$|\psi\rangle = \sum_n c_n \, |\phi_n\rangle$$



T. C. Weinacht, J. Ahn, and P. H. Bucksbaum, *Phys. Rev. Lett. 80, 5508 (1998).*

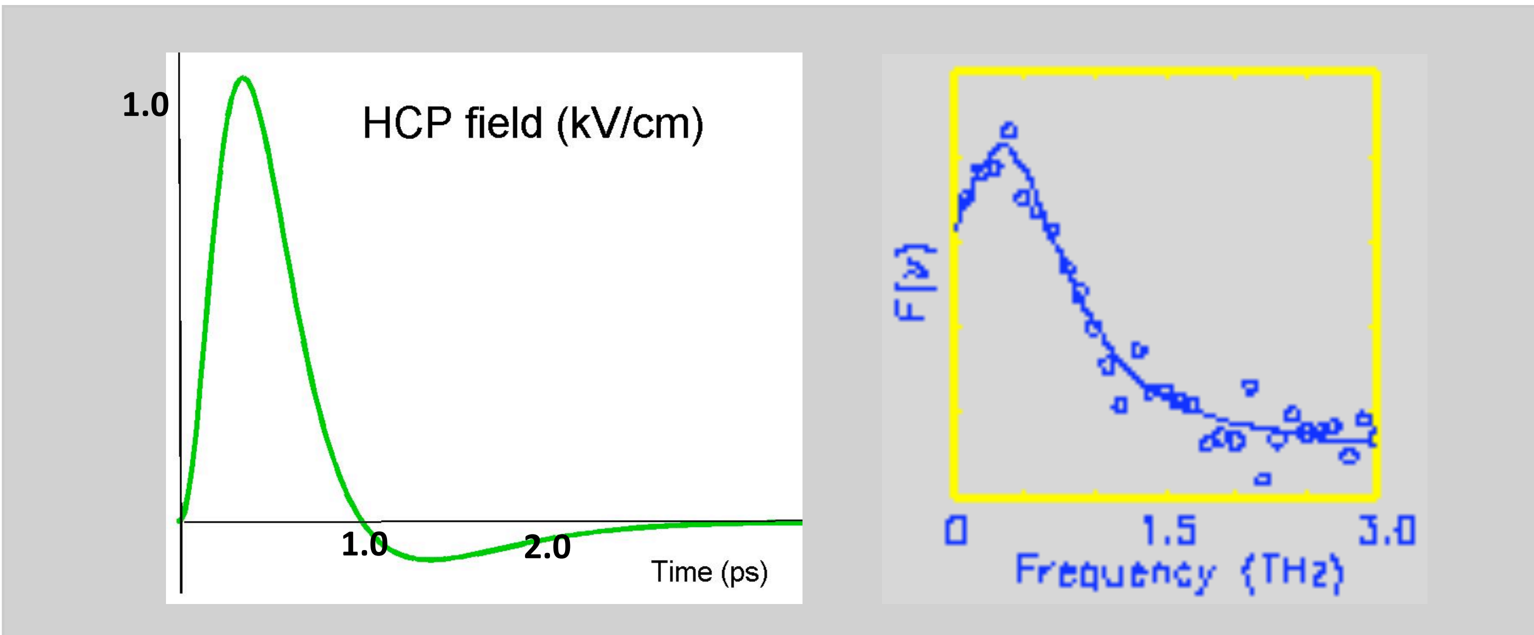
Bound and continuum wave packets



$$|\psi(t)\rangle = \sum_n c_n e^{-iE_n t/\hbar} |\phi_n\rangle \qquad |\psi(t)\rangle = \int dE c(E) e^{-iEt/\hbar} |\phi(E)\rangle$$

Control: THz Half-cycle pulse

- Unipolar electromagnetic field
- FWHM ~0.5ps



- Short unipolar lobe, then long negative tail

Impulse approximation

- When $T_{HCP} << T_{dynamical}$
- HCP ~ 'impulse' in direction of polarization
- Atoms do not feel effect of negative tail
- Can transfer momentum to a free electron

Control equation

$$i\dot{\psi}(\vec{r},t) = -\frac{\nabla^2\psi(\vec{r},t)}{2} - \frac{1}{r}\psi(\vec{r},t)$$
$$+\left(Q_1\delta(t_1)\vec{r}\cdot\hat{n}_1 + Q_2\delta(t_2)\vec{r}\cdot\hat{n}_2\right)\psi(\vec{r},t)$$

- We assume the first kick Q_1 is along the quantization axis (z-axis) – 2D problem
- The second kick can either be along the (z-axis) – still a 2D problem; or make some angle to the z-axis – 3D problem
- Comparisons to experiments in alkali atoms, potential slightly different from Coulomb

Numerical approaches

- Truncated basis of spherical harmonics

1. Essential states basis – energy truncation useful for studying bound state-to-state problems
   - If impulse is in the z-direction
   - Numerical diagonalization of impulse operator

$$
\begin{aligned}
|\psi^+\rangle &= e^{iQr\cos\theta}|\psi^-\rangle \\
&= S e^{iQr\Lambda} S^\dagger |\psi^-\rangle
\end{aligned}
$$

Rangan and Murray, Phys. Rev. A ***72****,* 053409 (2005)

Kicks in arbitrary directions

- Perpendicular kicks, impulse delivered along the arbitrary axis
- Rotate the desired axis onto the z-axis, kick along the new z, and then rotate back using D – matrices

$$R(\alpha, \beta, \gamma) Y_l^m(\theta, \phi) = \sum_{m'} D^l_{m,m'}(R) \hat{Y}_l^{m'}(\theta, \phi)$$

Numerical approaches

2. Representation using truncated radial grid and spherical harmonics (r-l basis)

$$|\psi\rangle = \sum_{l,m} \int_0^\infty \rho_{l,m}(r) |r; l, m\rangle dr$$

– Free evolution by implicit propagator (for 2D)

$$|\psi(t+\Delta t)\rangle \approx \left(1 + \frac{i\Delta t}{2} H\right)^{-1} \left(1 - \frac{i\Delta t}{2} H\right) |\psi(t)\rangle + O(\Delta t^3)$$

– Inversion of a penta-diagonal matrix for each value of l

– Time step limited by strength of kick

Computational Physics, Koonin eq. (7.30)

Numerical approaches

3. Represent radial wave function via collocation using (symmetry suited) Laguerre functions
   – Boyd, Rangan and Bucksbaum, Journal of Computational Physics, **188**, 56 (2003)

- Propagate using Chebychev propagator
  – H. Tal-Ezer and R. Kosloff, J. Chem. Phys. **81**, 3967 (1984)
- Fast, but not so good for studying Coulomb problem

Calculate spectrum of final state

- Energy spectrum by window method:

$$\langle E|\psi\rangle \approx \frac{\pi}{\sqrt{2}}\langle\psi|\frac{\gamma^3}{(H-E)^4+\gamma^4}|\psi\rangle$$

*Schafer and Kulander, Phys. Rev. A **42**, 5794 (1990)*

- Final state has both bound (discrete) and unbound (continuum) components

Coherent interactions of HCPs with RWPs

Q	
0.0014a.u.	Information storage & retrieval
0.0023a.u.	Synthesizing eigenstates
0.0046a.u.	Quantum search algorithm
0.01a.u.	Detecting angular momentum
0.02a.u.	HCP assisted recombination
Higher	Stabilization, imaging, chaos, …
Need all Q	Impulsive momentum retrieval
	Ionizing away from the atomic core

Atomic units: $e = m_e = \hbar = 1$

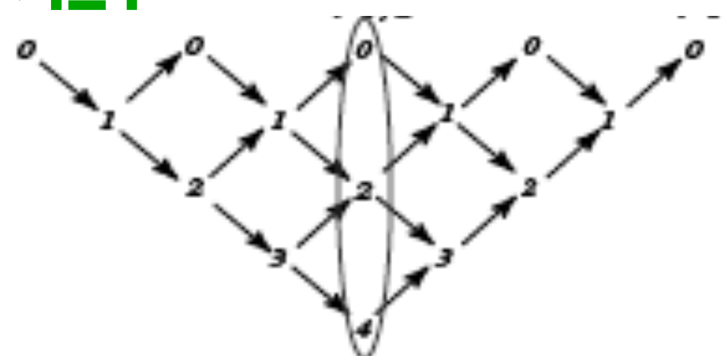
Control mechanism - quantum

Impulsive interaction

Propagator $U = e^{iQz}$

$$|\Psi_{final}\rangle = e^{iQz}\,|\Psi_{initial}\rangle$$

- To first order, HCP couples $l \rightarrow l\pm1$
- As Q increases:
  - $\langle l\rangle$ increases
  - $\max(\Sigma_n|\langle nl|\Psi\rangle|^2)$ goes towards higher 'l'



Control mechanism - classical

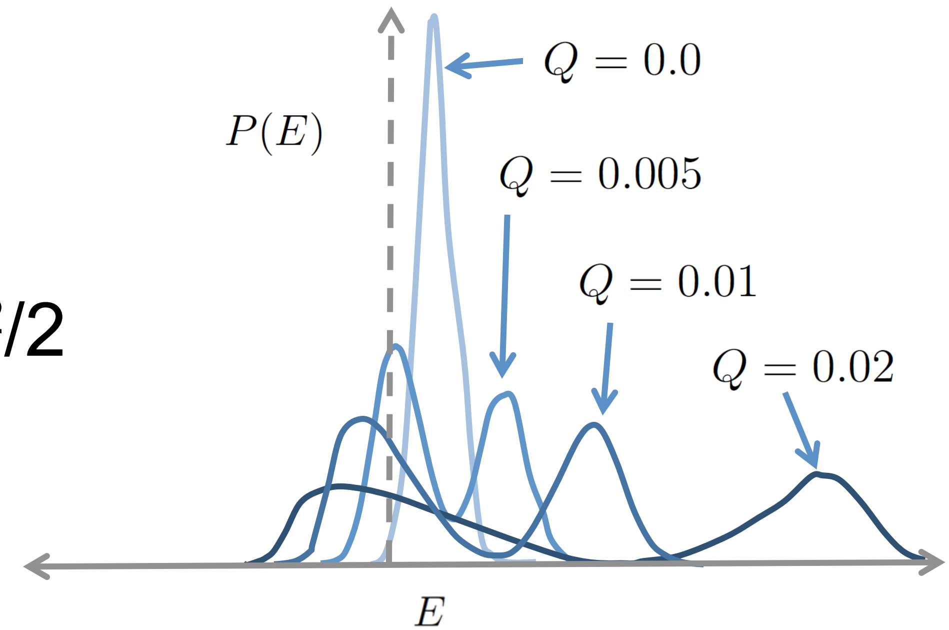
Impulsive interaction

HCP boosts the momentum of the electron

$\mathbf{p}_f = \mathbf{p}_i + \mathbf{Q}$

$E_f = \mathbf{p}_f^2/2$

$E_f = E_i + \mathbf{p}_i \cdot \mathbf{Q} + Q^2/2$



HCP also provides a torque to the bound electron increasing its angular momentum

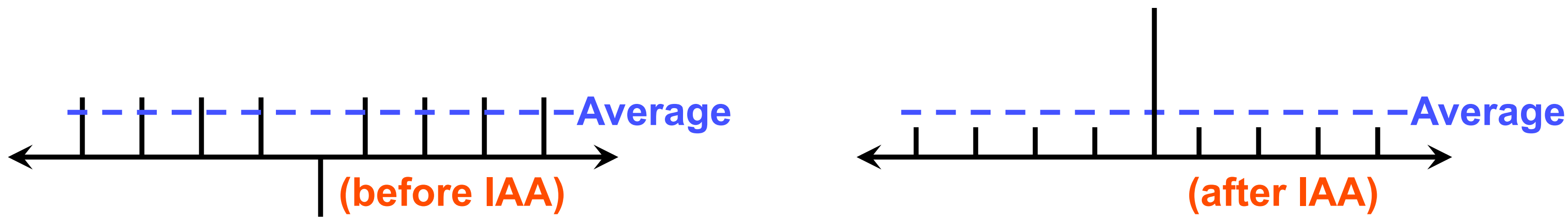
Case I. Performing a quantum algorithm in a Rydberg atom using an HCP

Single kick

- Collaborators: Phil Bucksbaum and group: Jae Ahn, Joel Murray, Haidan Wan, Santosh Pisharody, James White

Inversion-about-average operation

Grover, Phys. Rev. Lett. **79**, 325 (1998)



Conversion of phase information to amplitude information

$$\begin{pmatrix} -1+2/N & 2/N & 2/N & \cdots & 2/N \\ 2/N & -1+2/N & 2/N & \cdots & 2/N \\ 2/N & 2/N & -1+2/N & \cdots & 2/N \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 2/N & 2/N & 2/N & \cdots & -1+2/N \end{pmatrix} \begin{pmatrix} 1 \\ -1 \\ 1 \\ \vdots \\ 1 \end{pmatrix} = \begin{pmatrix} 1-4/N \\ 3-4/N \\ 1-4/N \\ \vdots \\ 1-4/N \end{pmatrix}$$

Ψ_i Ψ_T

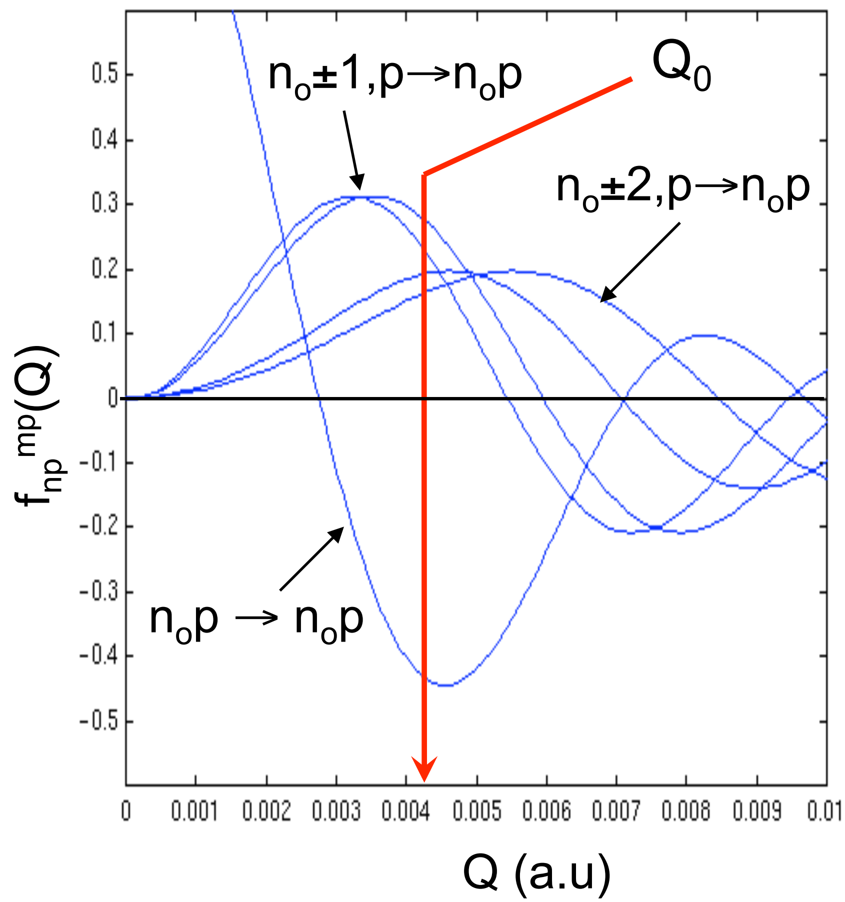
Control trick – know the atom

Impulsive interaction

$$|\Psi_{final}\rangle = e^{iQz}\,|\Psi_{initial}\rangle$$

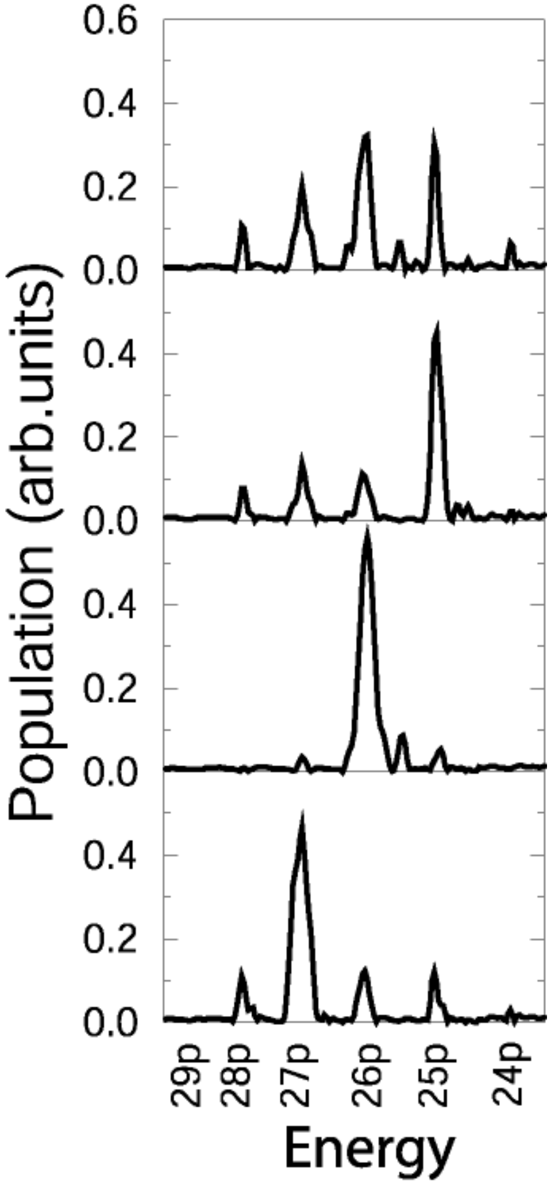
Matrix element

$$f_{nl}^{ml'}(Q_0) = \langle m,l'|\, e^{iQz}\,|n,l\rangle$$



Arrange wave packet superposition to obtain desired outcome

HCP can perform a quantum search



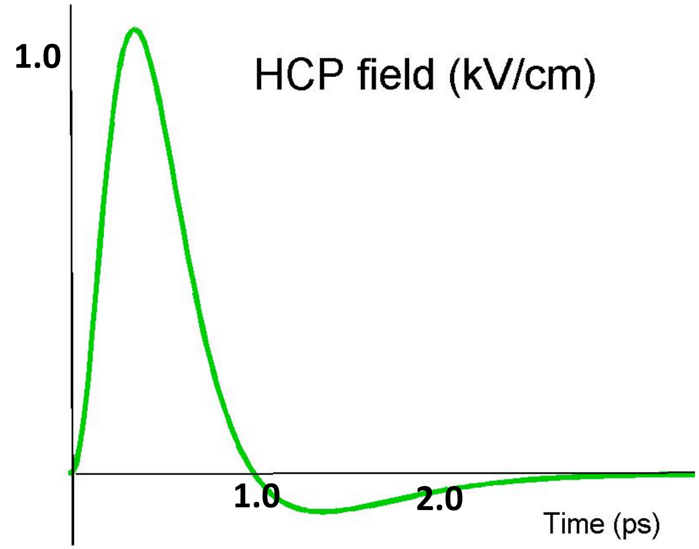
Before HCP

t=2.1ps
(000010)

t =4.2ps
(000100)

t =4.7ps
(001000)

Phys. Rev. Lett. 86, 1179 (2001)



Both impulse model and realistic model of the HCP give excellent agreement with the experiment.

Phys. Rev. A, 66, 22312 (2002)

Optimal Control

Shi & Rabitz (1988, 1990), Kosloff et al (1989), …

Find the control field E(t), $0 \leq t \leq T$

Initial state: $|\Psi(t=0)\rangle = |24p\rangle + |25p\rangle - |26p\rangle + |27p\rangle + |28p\rangle + |29p\rangle$

Target functional: $|\langle 26p|\Psi(T)\rangle|^2$ — maximize

Cost functional: $\int_0^T I(t)|E(t)|^2 dt$ — minimize (penalty parameter)

Constraint: Schrödinger's equation $|\dot{\Psi}(t)\rangle + \iota H(t,E(t))|\Psi(t)\rangle = 0 + \text{c.c.}$

Introduce Lagrange multiplier: $|\lambda(t)\rangle$

Maximize unconstrained functional:

$$J = |\langle 26p|\Psi(T)\rangle|^2 - \int_0^T I(t)|E(t)|^2 dt - 2\,\mathrm{Re}\int_0^T dt\left(\langle\lambda(t)|\dot{\Psi}(t)\rangle + \iota H(t,E(t))|\Psi(t)\rangle\right)$$

Use Krotov's algorithm with Tannor's update rule

Rabitz (1991); Krotov (1983, 1987); Rabitz (1998); Tannor (1992)

Krotov method: J= terminal part + non-terminal part

$$= G(t=0, t=T) \quad + \int_0^T dt\, R(t)$$

The optimal E(t) maximizes both parts.

Using modified objective:

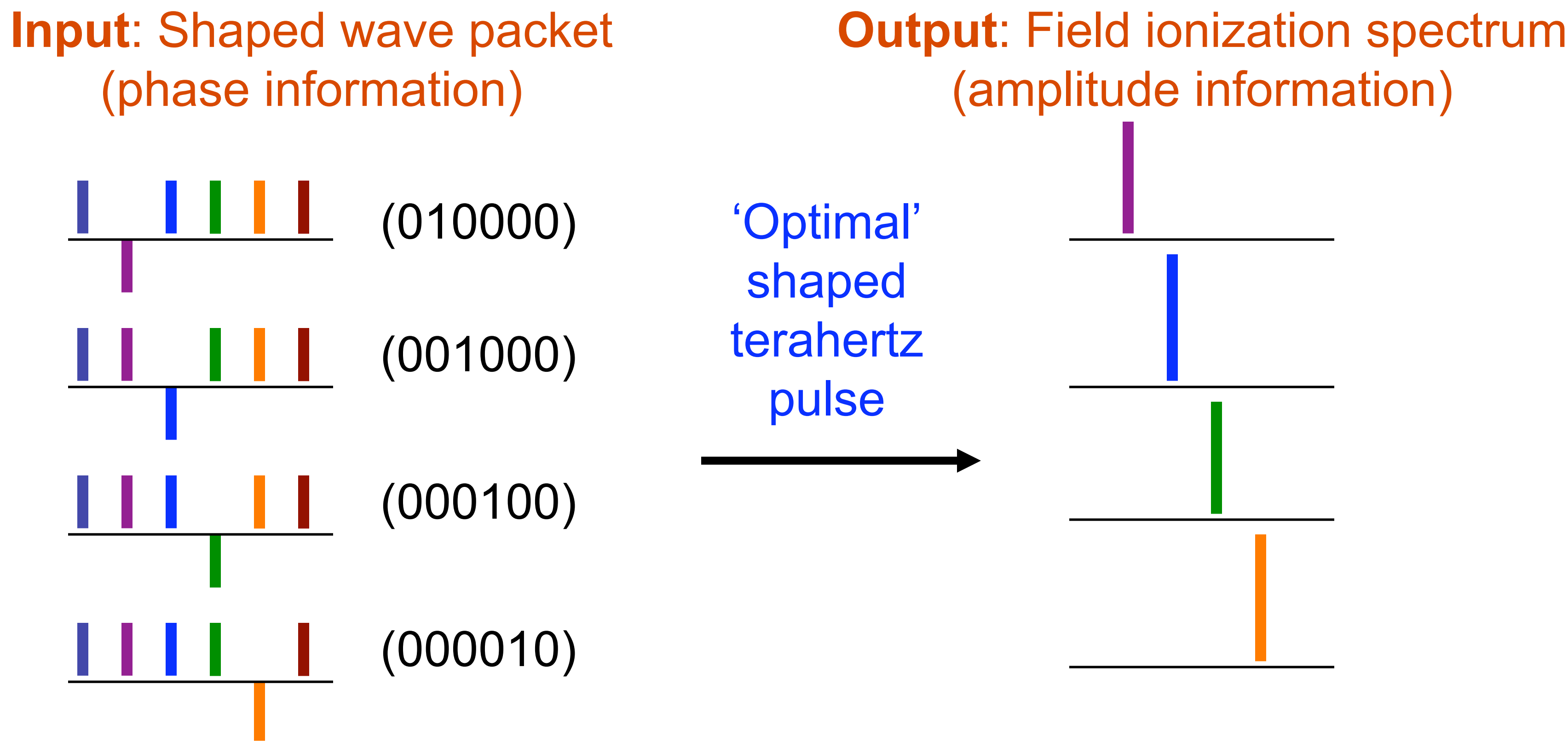
Change in the field at the k+1th iteration:

$$\Delta E(t) = \frac{-\iota}{l(t)} \langle \lambda^k(t) \,|\, z \,|\, \Psi^{k+1}(t) \rangle$$

- $T \approx 8$ps, nt = 1000
- Wave packet propagation: split-operator method
- Essential basis of 187 states: $21 \leq n \leq 31$, $l < 17$
- Basis states calculated by a grid-based pseudopotential method

But we actually want to …

Design a shaped broadband terahertz pulse than can optimize the performance of the search algorithm



Independent subspace model

Shape of terahertz pulse is calculated by introducing a modified target functional

$|\psi_i> = |010000> + |001000> + |000100> + |000010>$

$(|001000> = |24p> + |25p> - |26p> + |27p> + |28p> + |29p>)$

Target = $|<25p|\psi(T)>|^2 + |<26p|\psi(T)>|^2 + |<27p|\psi(T)>|^2 + |<28p|\psi(T)>|^2$

→ maximize

Each ‘subspace’ evolves independently under the influence of the same terahertz field.

Change in control field at k+1th iteration:

$$\Delta E(t) = \frac{-i}{I(t)} \sum_{i=1}^{N} \langle \lambda^{k}{}_{(i)}(t) \,|\, z \,|\, \Psi^{k+1}{}_{(i)}(t) \rangle$$

Again use Krotov's algorithm with Tannor's update rule

Using modified objective:

Change in the field at the k+1th iteration:

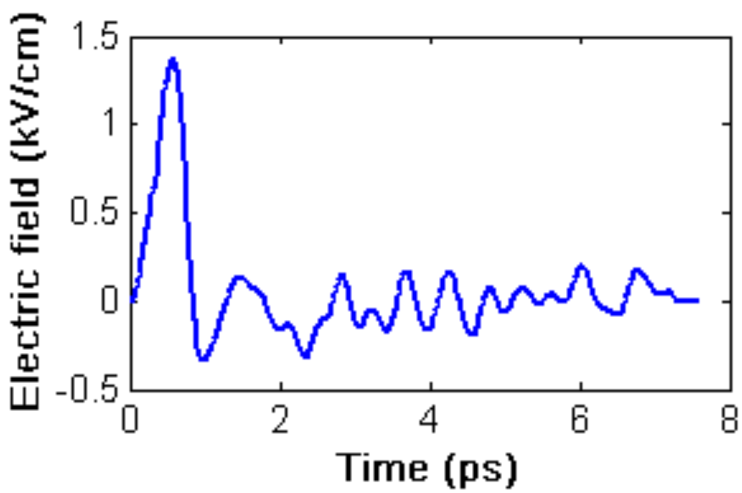
$$\Delta E(t) = \frac{-\iota}{I(t)} \sum_{i=1}^{N} \langle \lambda^{k}{}_{(i)}(t) | z | \Psi^{k+1}{}_{(i)}(t) \rangle$$

Gives optimal pulse for inversion about mean algorithm for all initial states within a subspace

Also called: state-independent control, multi-operator control, optimizing a unitary transformation, optimizing a quantum operator, W-problem, …

Phys. Rev. A, **64**, 33417 (2001)

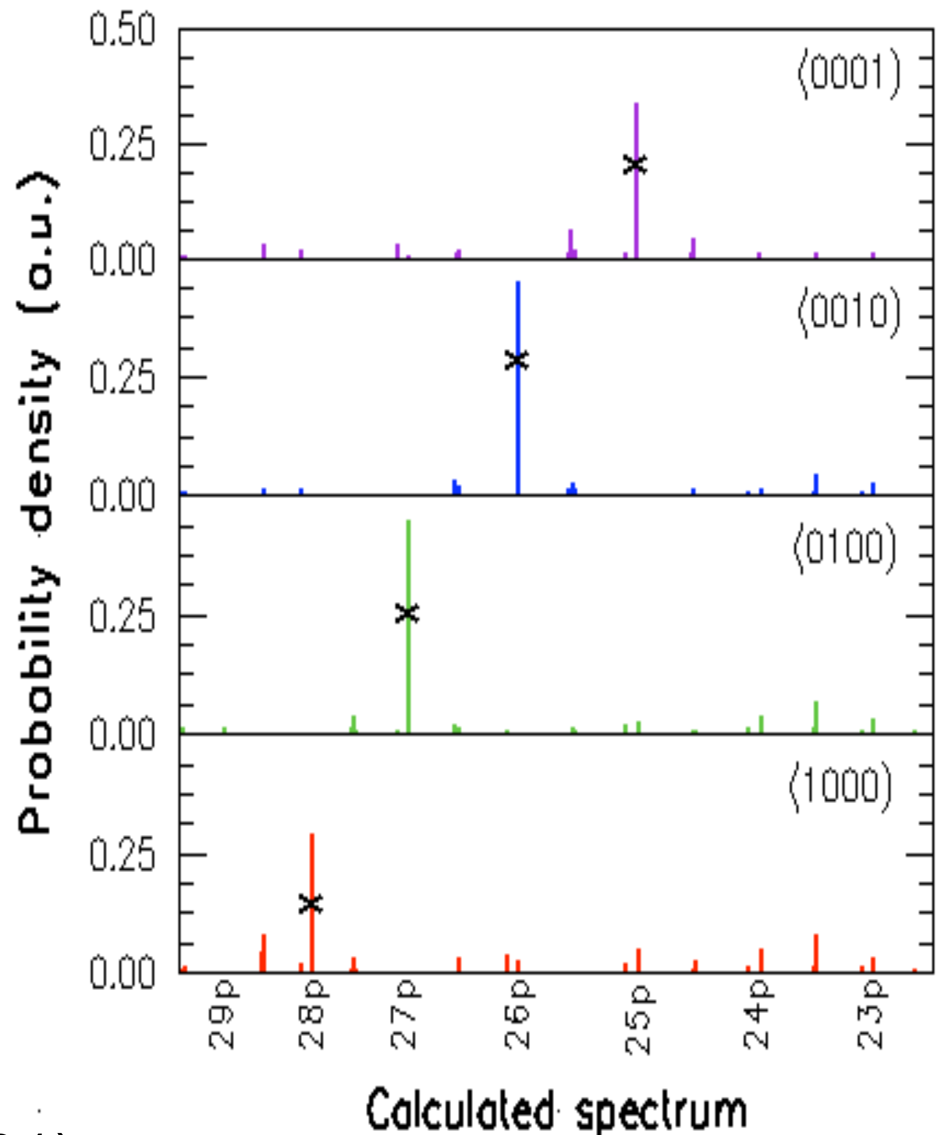
Optimal Pulse for Phase Retrieval



Database:
|24p⟩ to |29p⟩ of Cs

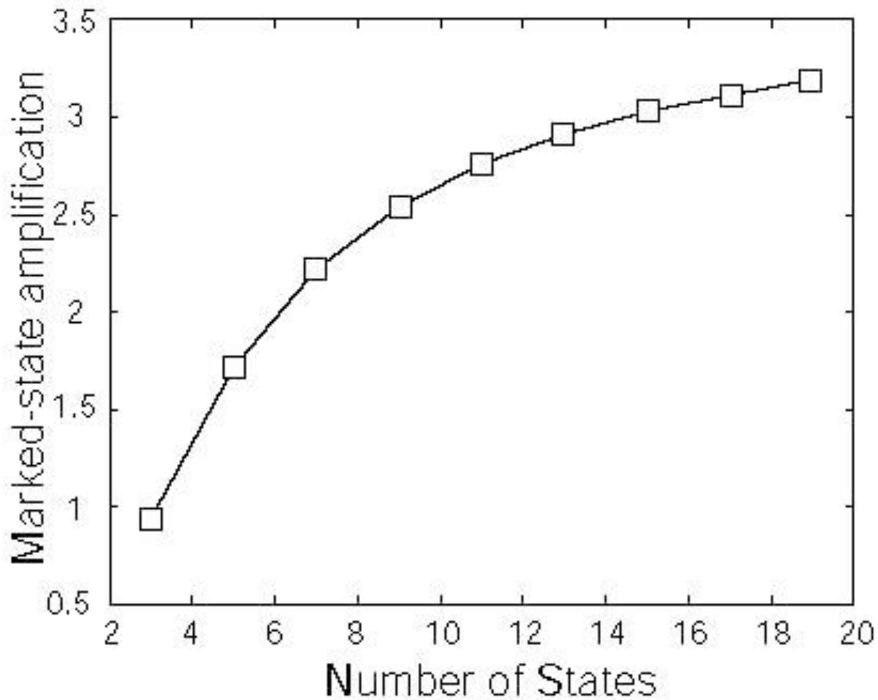
Guess pulse:
Half-cycle pulse

Optimal pulse:
Shaped THz pulse



Phys. Rev. A, **64**, 33417 (2001)

Scaling of the implementation



With increasing database size, the amplification of the marked bit tends to a value of 4.

localized wave packet

delocalized eigenstate

$$|\Psi_i\rangle = |24p\rangle + |25p\rangle - |26p\rangle + |27p\rangle + |28p\rangle + |29p\rangle = (|24p\rangle + \cdots + |29p\rangle) - 2|26p\rangle$$

A half-cycle pulse destroys the localized wave packet while leaving the extended eigenstate untouched.

$$|\Psi_f\rangle \approx -2|26p\rangle; \qquad |\langle 26p|\Psi_f\rangle|^2 / |\langle 26p|\Psi_i\rangle|^2 \approx 4$$

Example IIa. Two kicks

Kick strengths are low so that the problem is completely bound, i.e, no ionization

Hiding and retrieving coherence from within a subspace

Collaborators:

Phil Bucksbaum's group

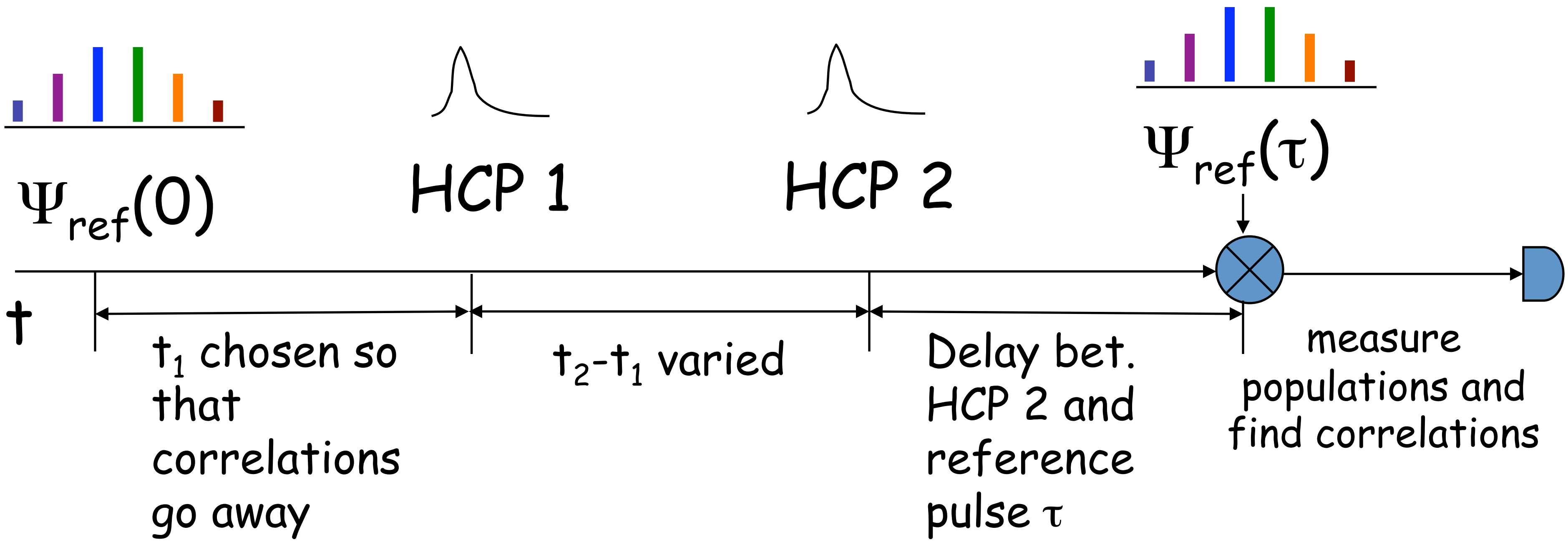
(when at the University of Michigan)

Joel Murray

Santosh Pisharody

Haidan Wen

Schematic



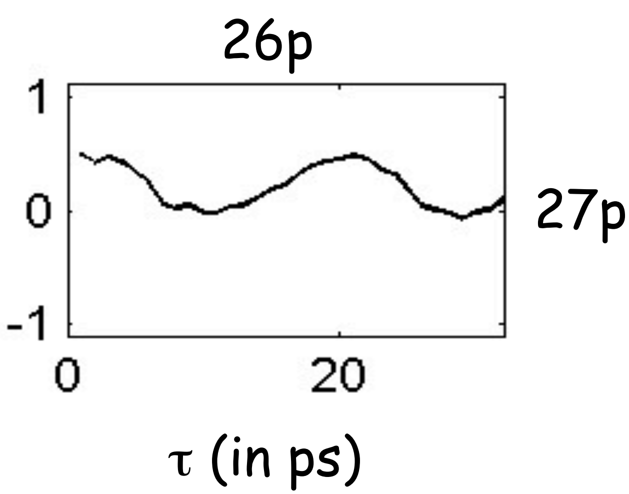
Finite subsystem: n=26-31, p-states |k⟩ of cesium.

Measure: Correlations between state populations after time delay

$$r_{jk}^{meas} = \sqrt{\left(1 - \frac{\sigma_{N_j}^2}{\sigma_{j_{meas}}^2}\right)\left(1 - \frac{\sigma_{N_k}^2}{\sigma_{k_{meas}}^2}\right)} \times$$

$$\cos\left(\left(\phi_{j1} - \phi_{k1}\right) - \left(\phi_{j2} - \phi_{k2}\right) - \left(\omega_j - \omega_k\right)\tau\right)$$

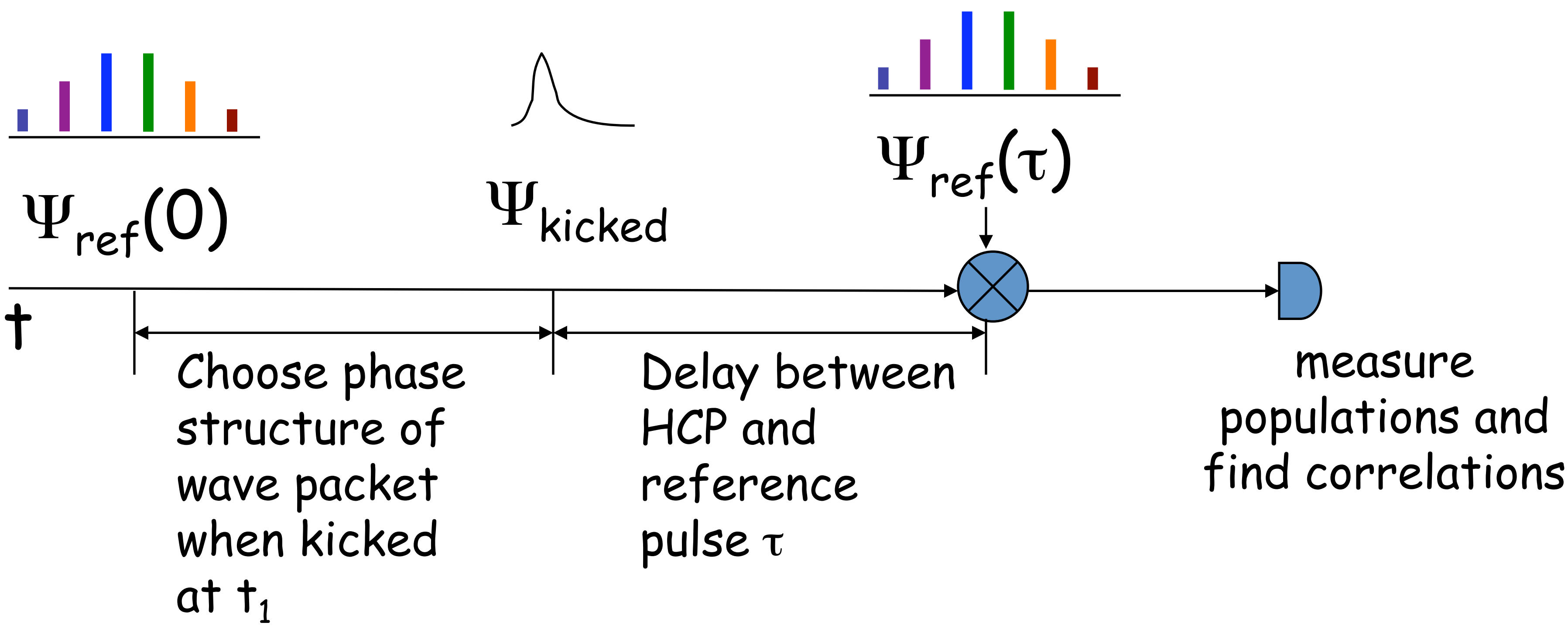
$$= |r_{jk}^{meas}| \cos(\Phi_{jk} - \omega_{jk}\tau).$$



Amplitude of correlation is a measure of the 'recoverability' of stored phase coherence.

Storage/hiding of phase coherence

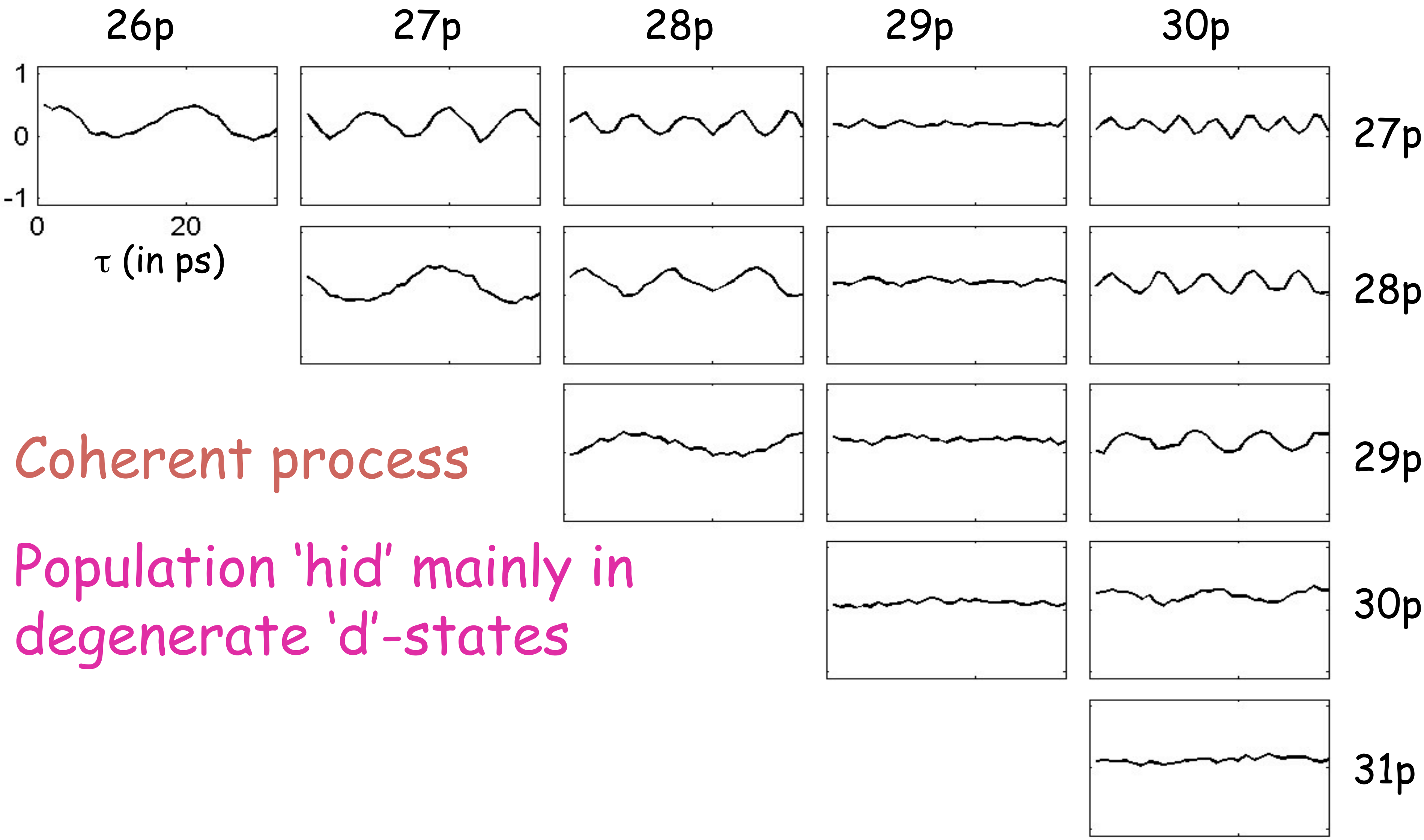
Murray *et al.*, Phys. Rev. A 71, 023408 (2005).



Correlations provide information regarding the phase coherence between the various eigenstates.

Amplitude of correlation is a measure of the 'recoverability' of stored coherence.

After first kick: Correlations go away at specific t_1 for all τ : Phys. Rev. A 71, 023408 (2005)

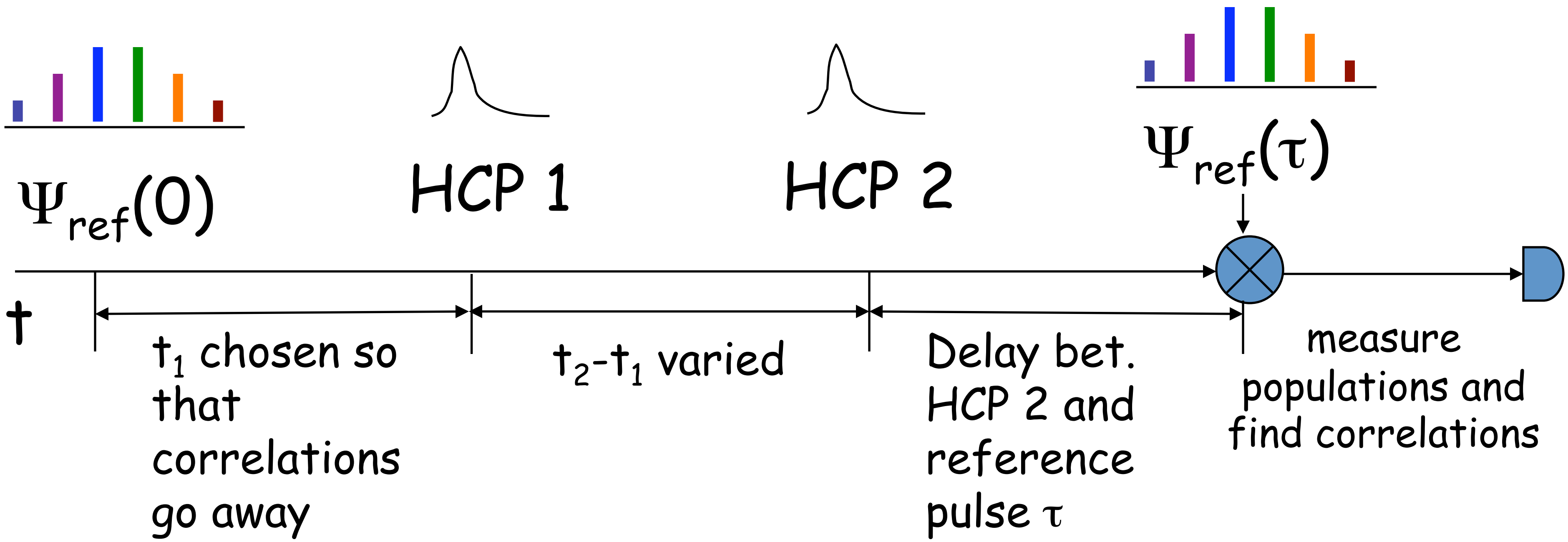


Coherent process

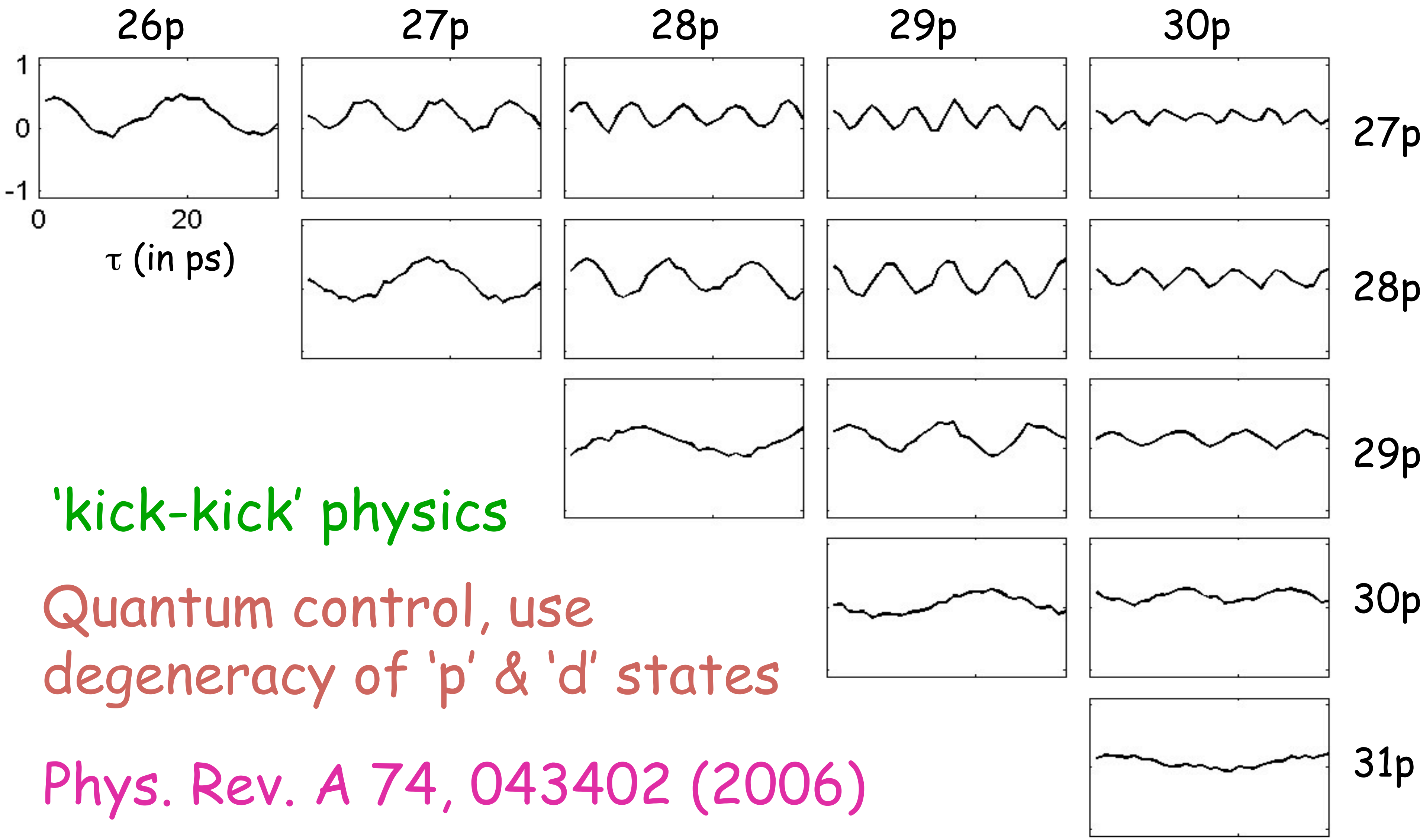
Population 'hid' mainly in degenerate 'd'-states

Recovering stored coherence

Use second HCP



After second kick at t_2: Correlations come back for all t_2-t_1



'kick-kick' physics

Quantum control, use degeneracy of 'p' & 'd' states

Phys. Rev. A 74, 043402 (2006)

Case IIb. Kick-kick

Kick strengths are high enough that continuum states are involved

Collaborator:

Jeffrey Rau (now doing a PhD at U. Toronto)

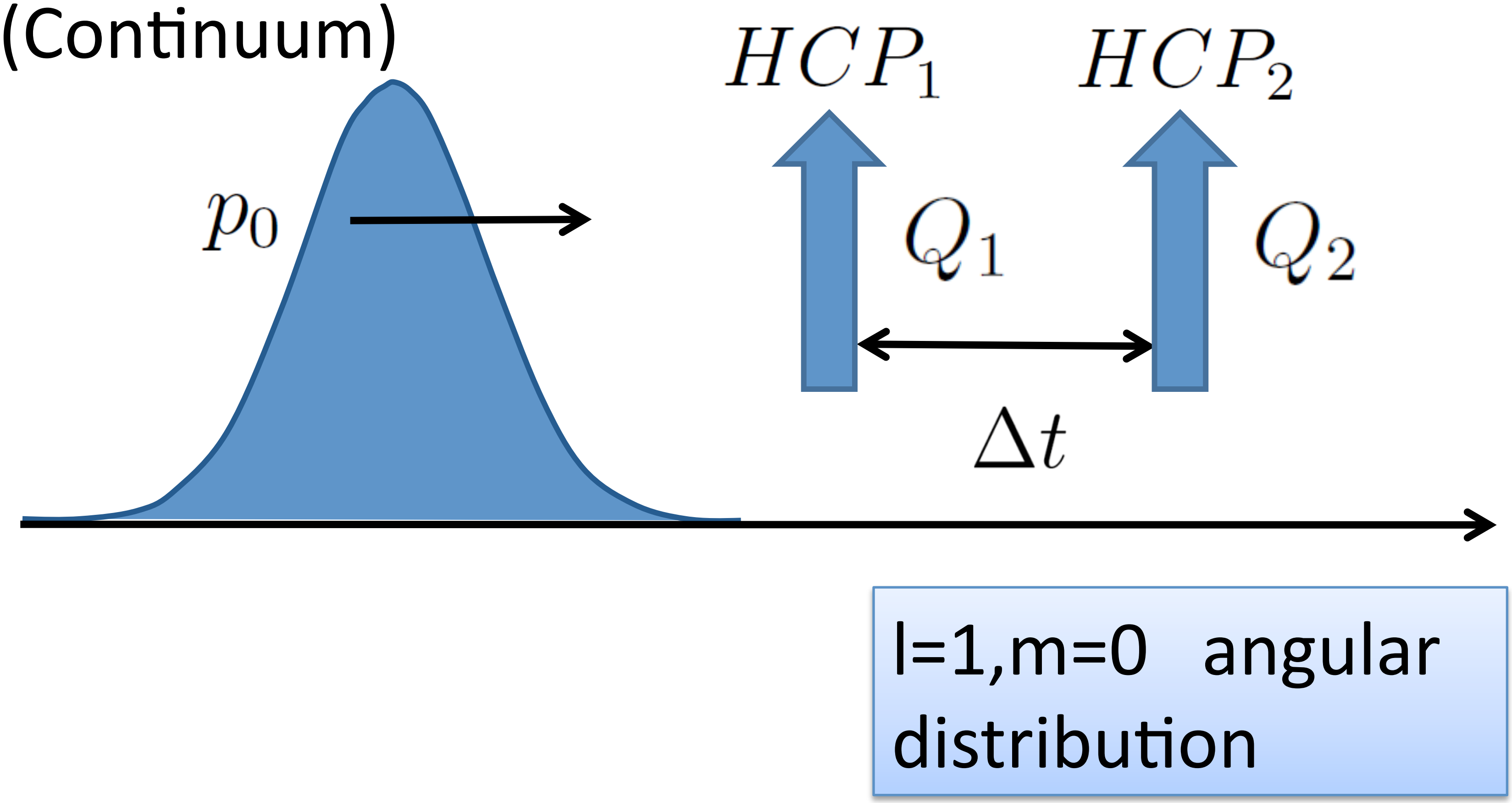
- Motivation: ‘kick-kick’ experiments of

  Ziebel and Jones, *Phys Rev A **68**, 023410 (2003)*

Schematic

Radial wave packet created by laser excitation, defines the z-axis

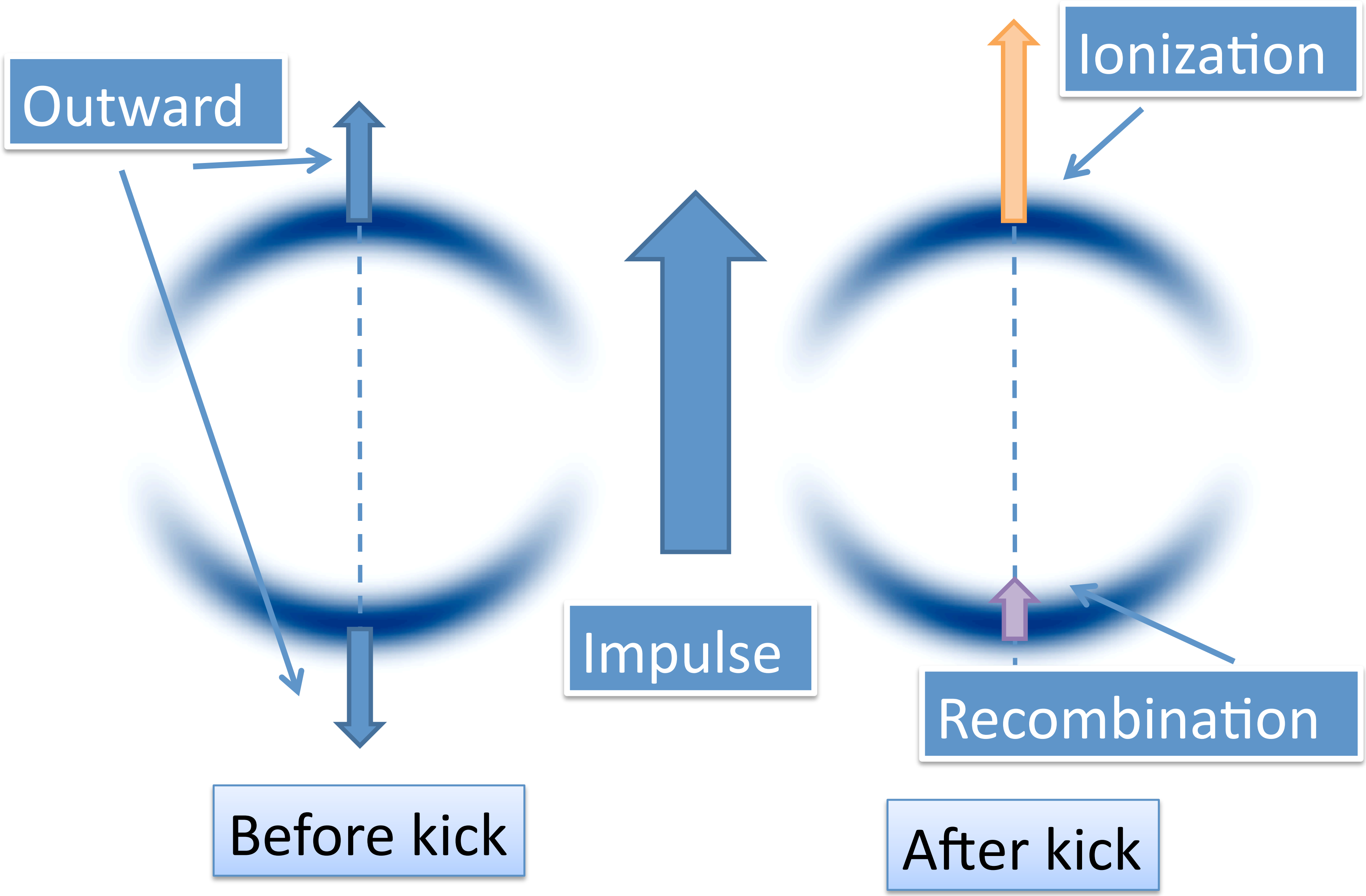
Radial wave packet (Continuum)



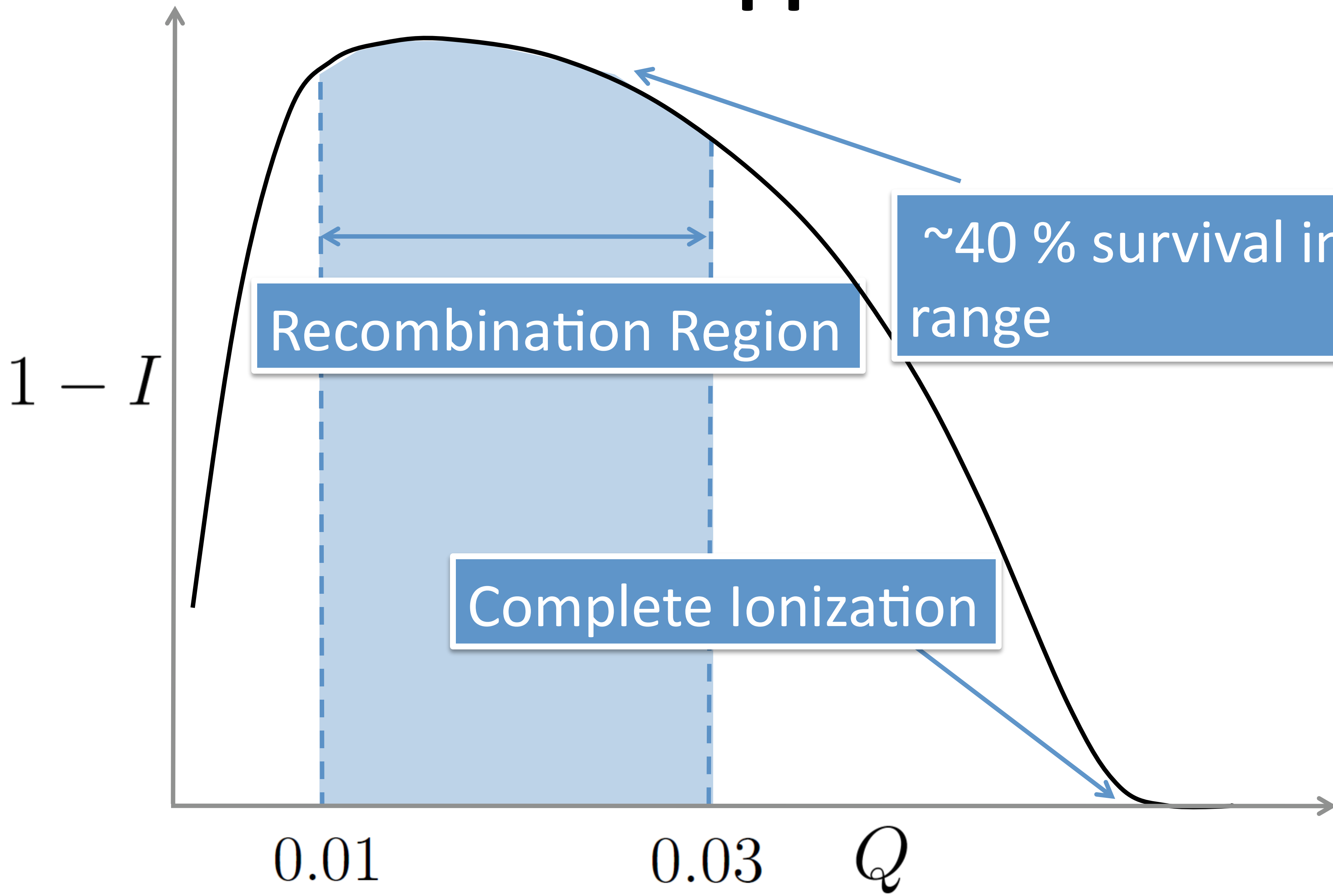
Measure/calculate

- Ionized fraction (integral of all the positive energy components)
- Recombination fraction (1-Ionized fraction) a.k.a. survival probability

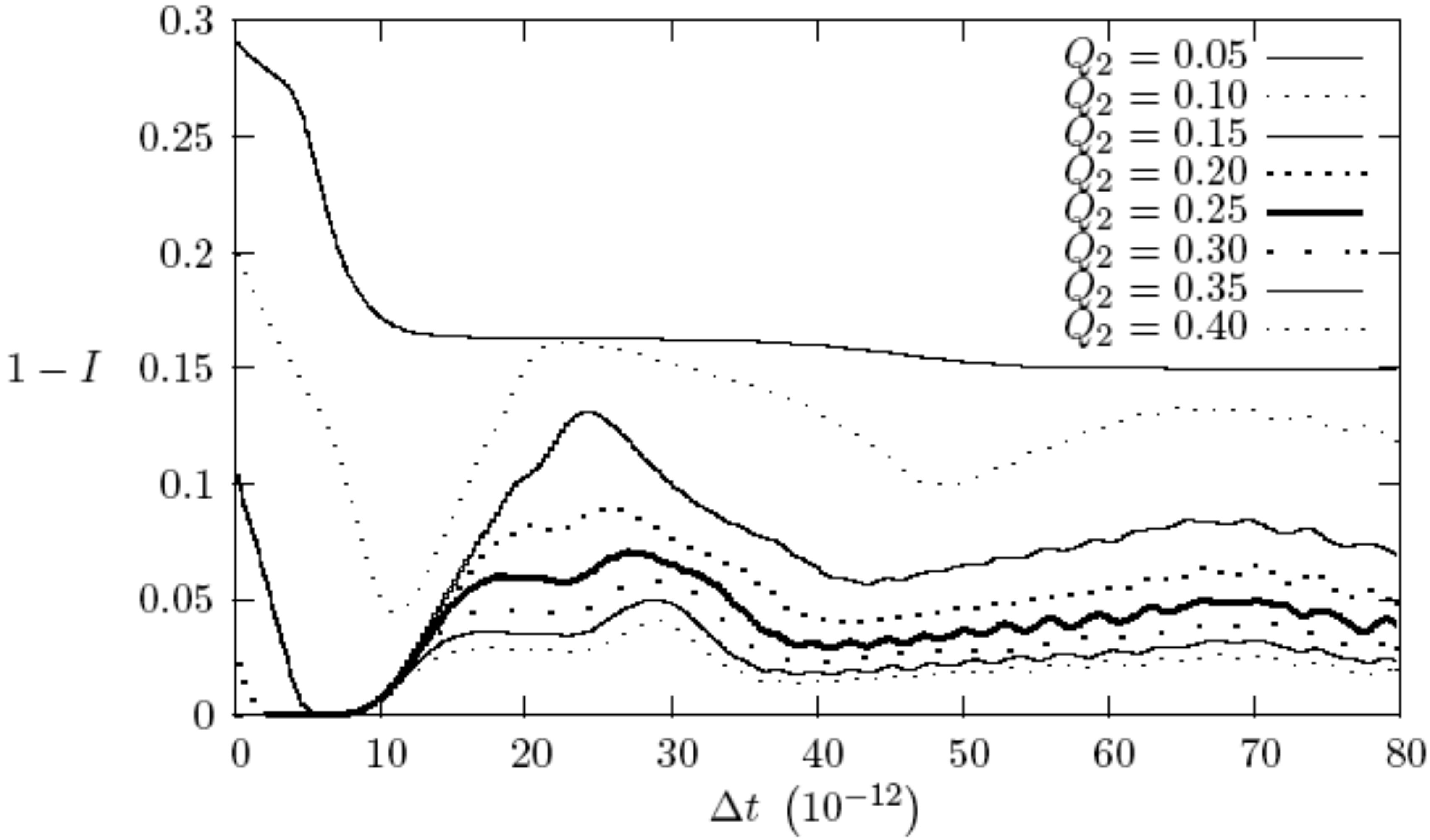
Recombination after first z-kick



Recombination: suppress ionization

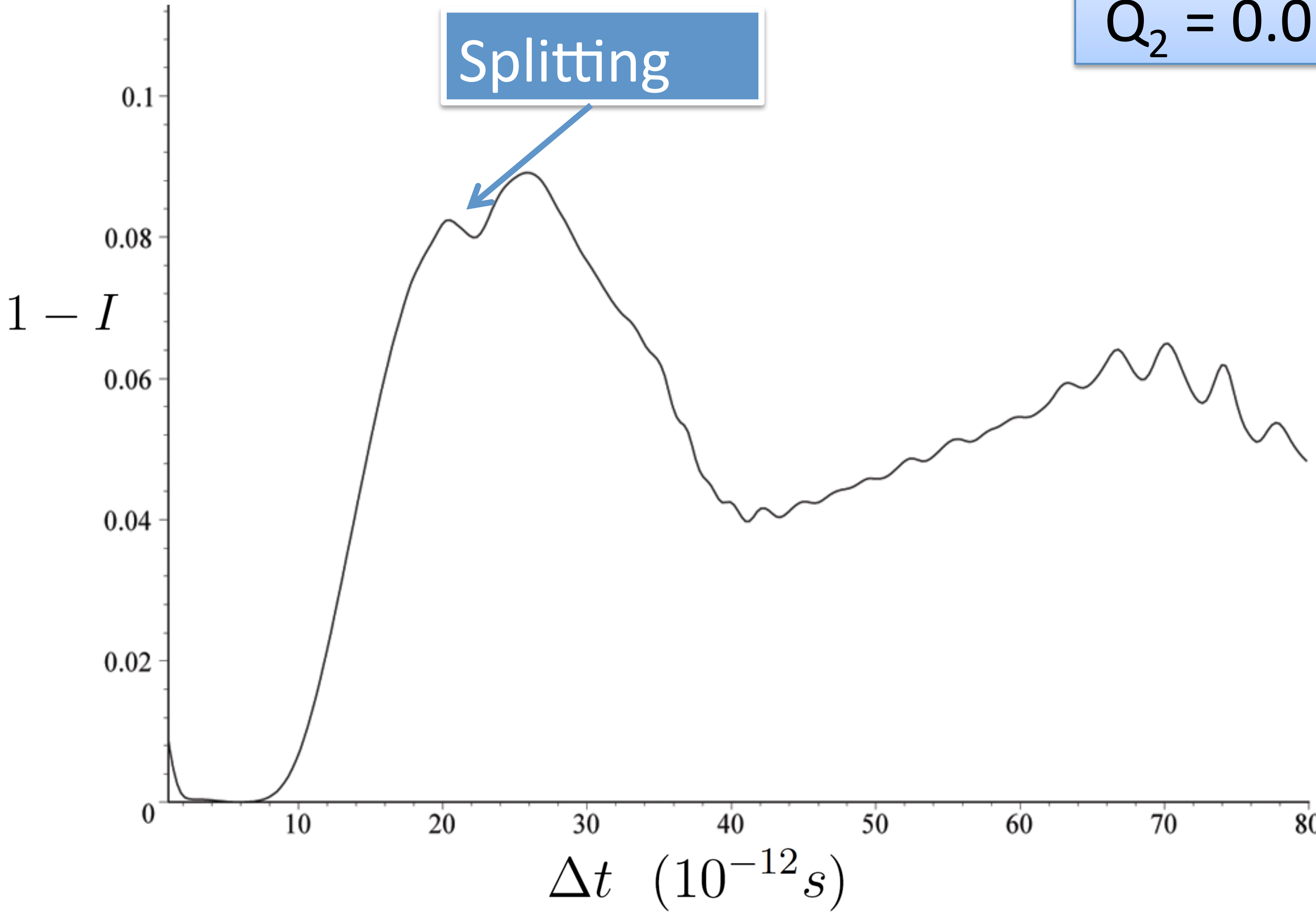


Q_1 = 0.02 is **fixed**, Q_2 is *varied*

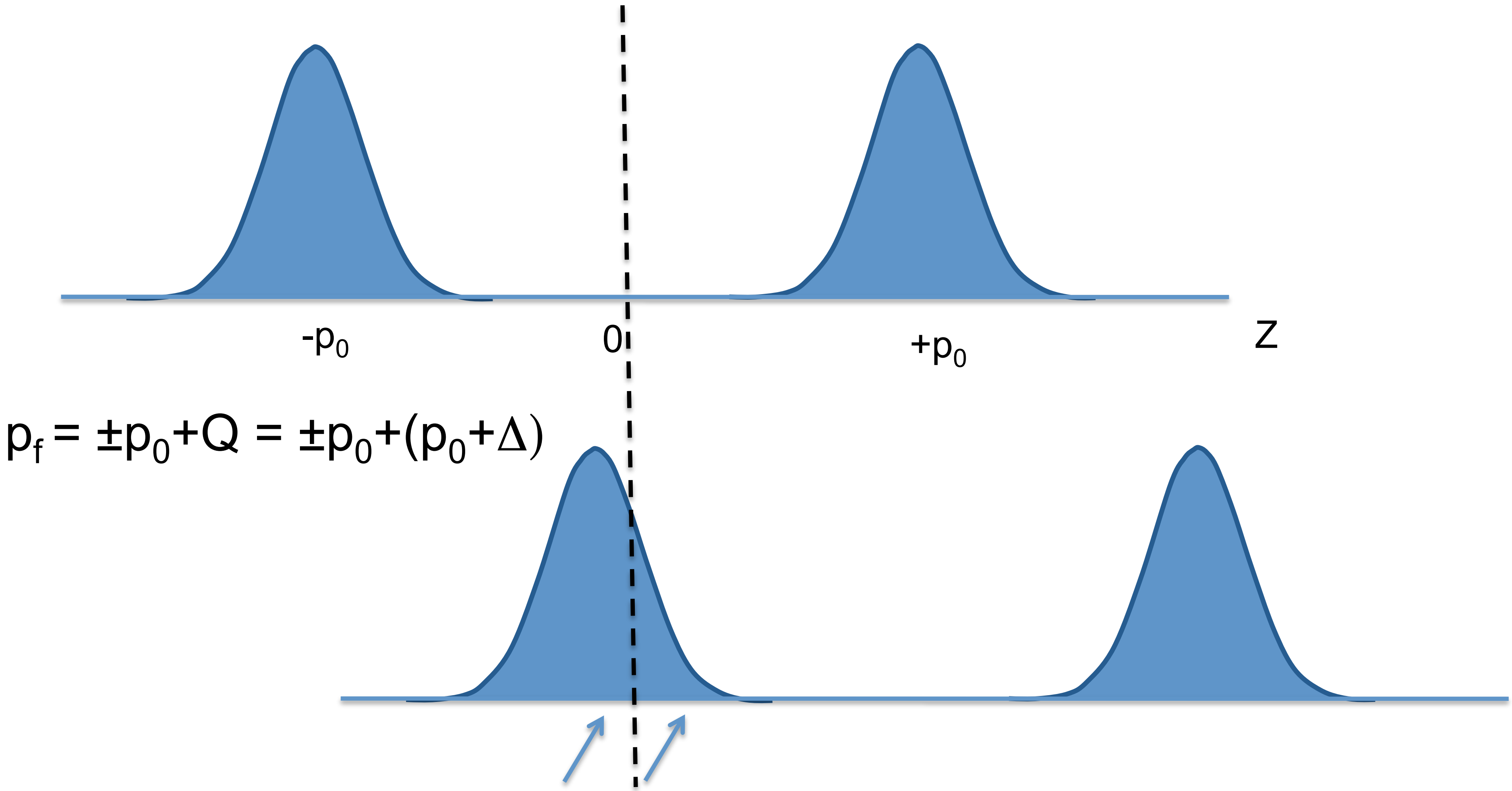


Kicks ~ initial momentum

Q_1 is 0.02
Q_2 = 0.02



- kick strength near the initial radial momentum



Produces two peaks in the survival curve

Summary

- Impulse operator can perform inversion about average in Rydberg atom: Phys. Rev. Lett. 86, 1179 (2001)
- Augmented optimal control to optimize a quantum algorithm: Phys. Rev. A, 64, 33417 (2001)

Kick-kick control:

- Out-of-subspace trajectory control – information hiding and retrieval, protecting coherence: Phys. Rev. A, 74, 43402 (2006).
- QIP with angular momentum states: Phys. Rev. A 72, 053409 (2005); Phys. Rev. A 68, 53405 (2003).
- HCP assisted ionization and recombination – J.G. Rau & C. Rangan (unpublished).

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Research group:

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Mr. Amin Torabi (MSc)

Mr. John Donohue (UG)

Mr. Mustafa Sheikh (UG)

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patient centred excellence

OAML

MITACS

Ontario Centres of Excellence

UNIVERSITY OF WINDSOR

Ontario

Canada Foundation for Innovation
Fondation canadienne pour l'innovation

NSERC CRSNG

ICIP CIPI

Trapped-ion control work

- Finite (approximate) controllability of trapped-ion quantum states (even beyond the Lamb-Dicke limit) IEEE Trans. Aut. Control, v. 55, pp.1797-1805 (2010).
- Only eigenstate controllability is possible in spin-half coupled to two harmonic oscillators (cannot use Law-Eberly schemes for gates) Quantum Information Processing, v. 7, pp. 33-42 (2008).
- Bichromatic control by truncating the Hilbert space: Phys. Rev. Lett., 92, 113004 (2004).
- Spin-half coupled to finite harmonic oscillator is controllable; quantum transfer graphs: J. Math. Phys., v. 46, art. no. 32106 (2005).
- If an n-qubit system has a symmetric distribution of field-free eigenenergies, the system can be controlled by only $2^n(2^n+1)$ elements of the $sp(2^n)$ algebra: Phys. Rev. A, 76, 33401 (2007).