Quantum Control of Atomic and Molecular Processes by Shaped Laser Field

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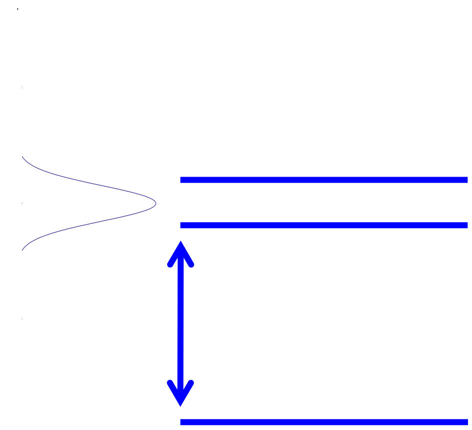
AMO Physics Seminar @ NTHU (Tainan)

Outline

Quantum Control

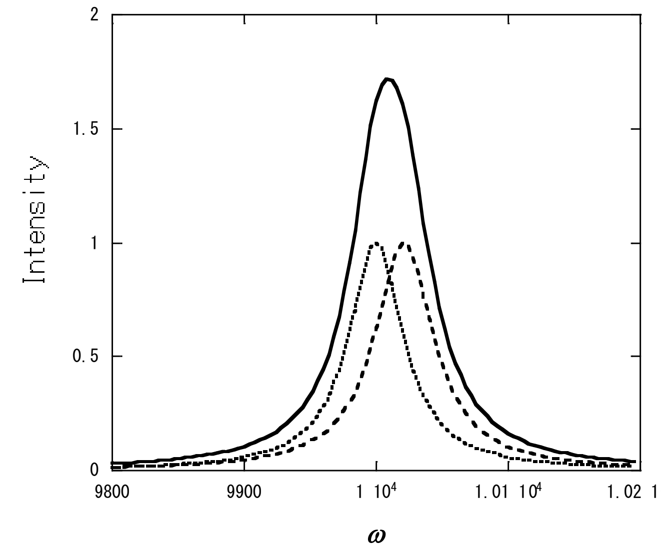
- Ultrafast Selective Excitation

  A pair of weak pulses
  Quadratic Chirping

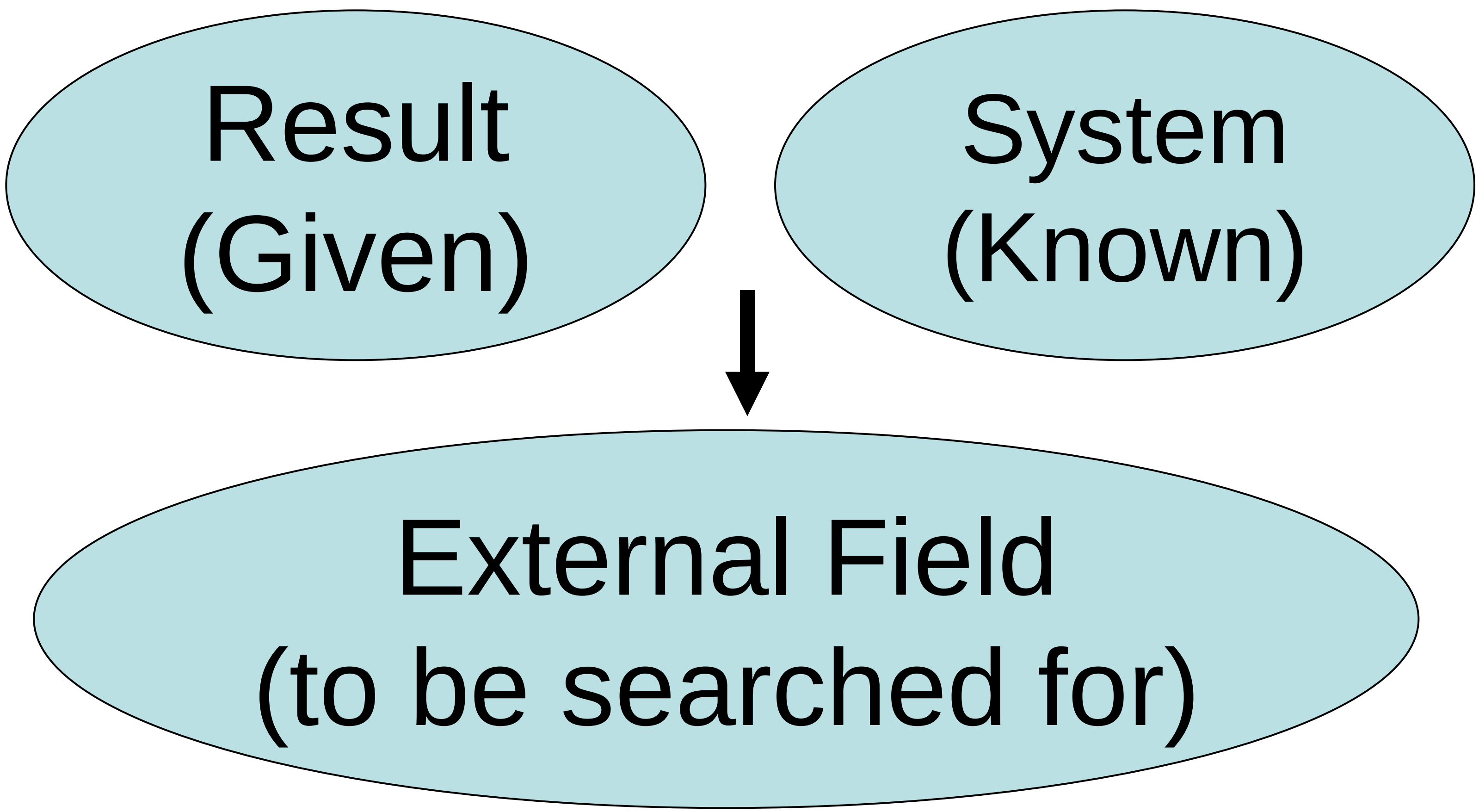


- Quantum Control Spectroscopy
  (overlapping by natural width)

- Molecular computer
  (Classical computer)



Quantum Control



Why Laser?

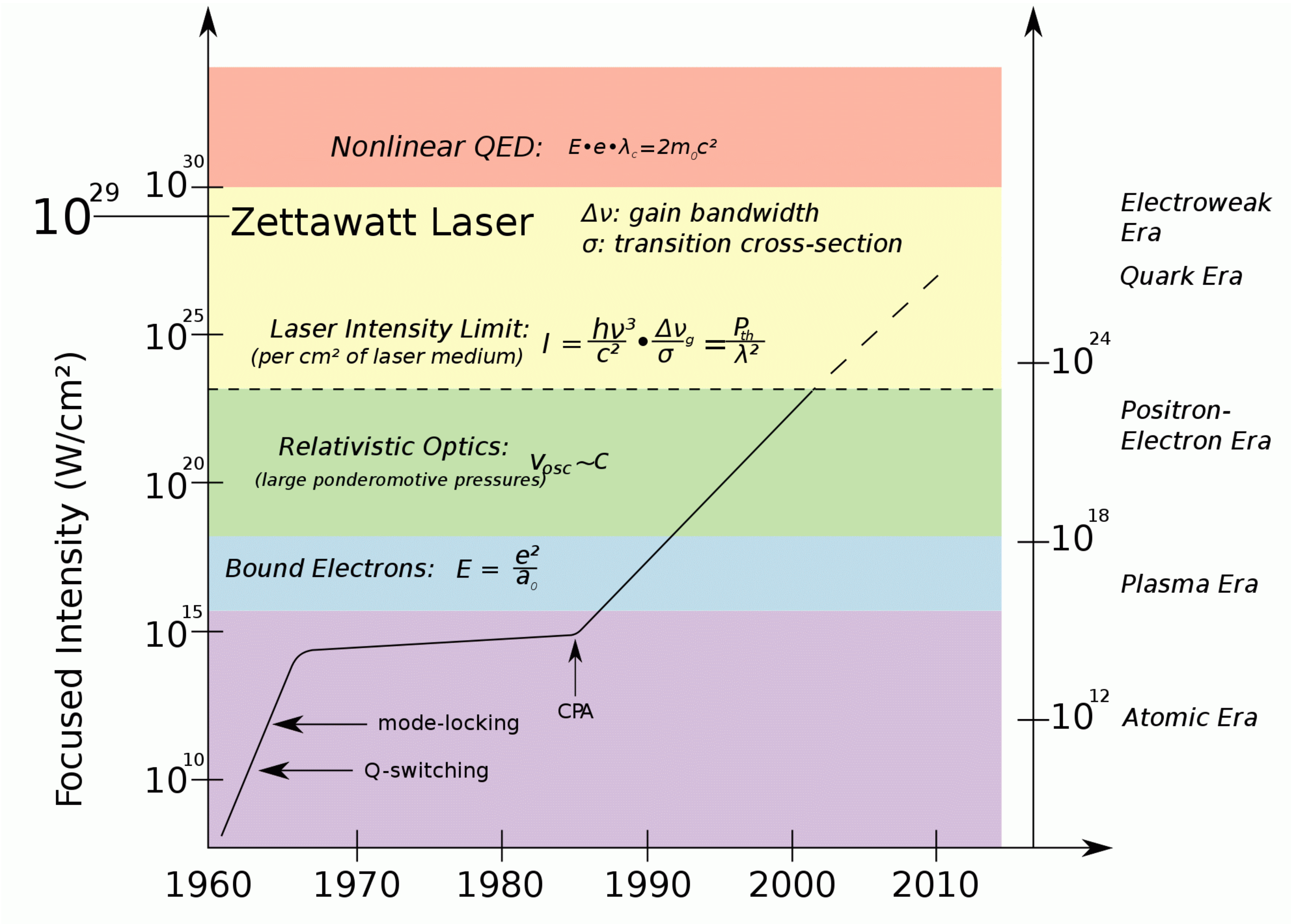
Coherence

High Intensity

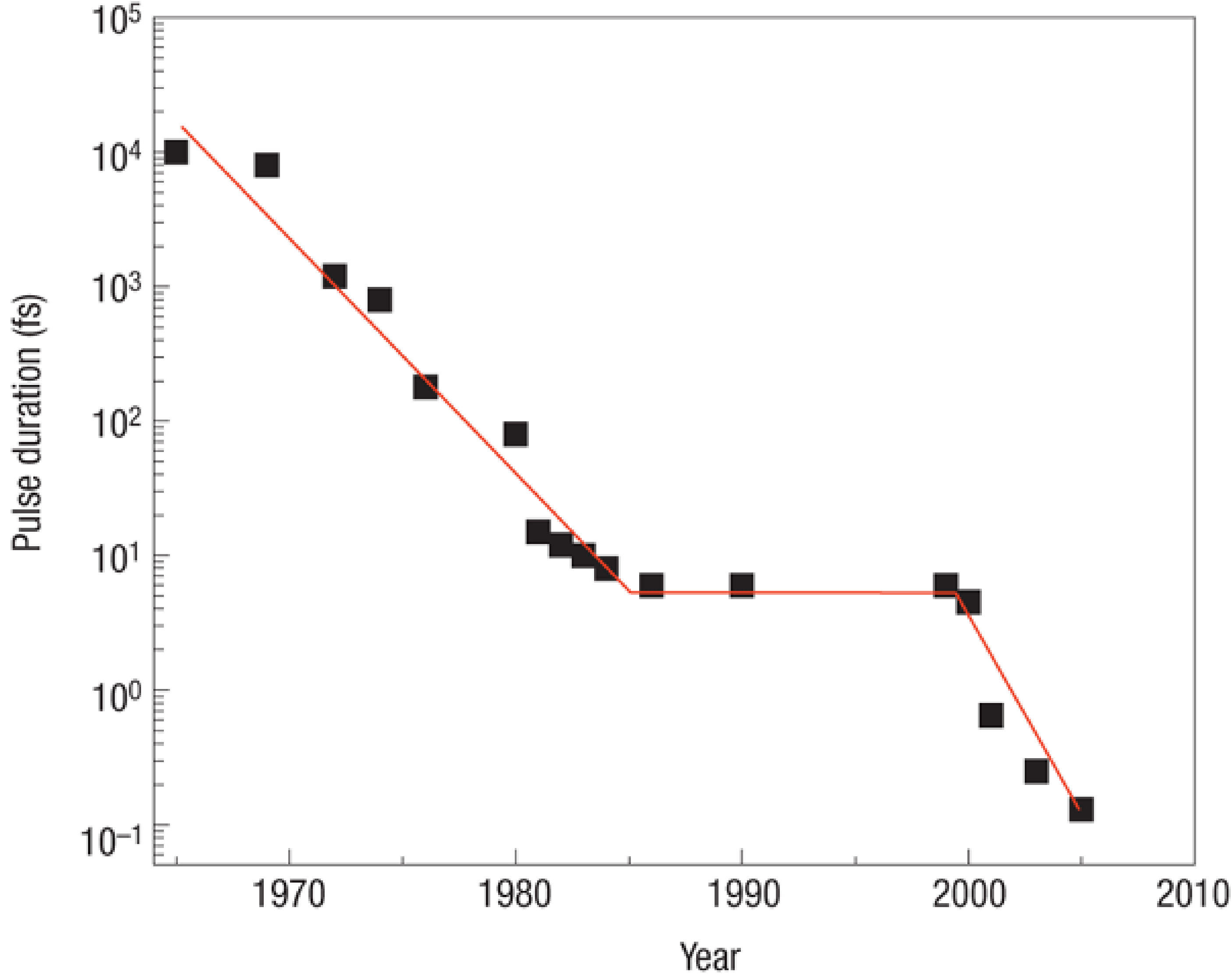
Short Pulse (Broad Band)

Pulse Shaping

History of Laser Intensity

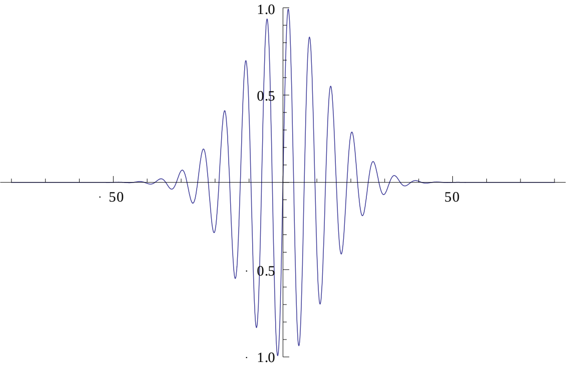


History of Laser Pulse Duration

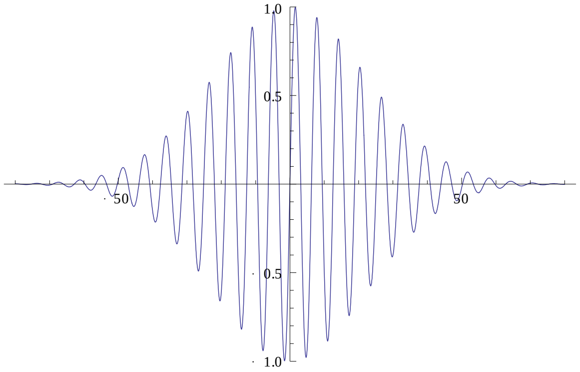


Laser Pulse

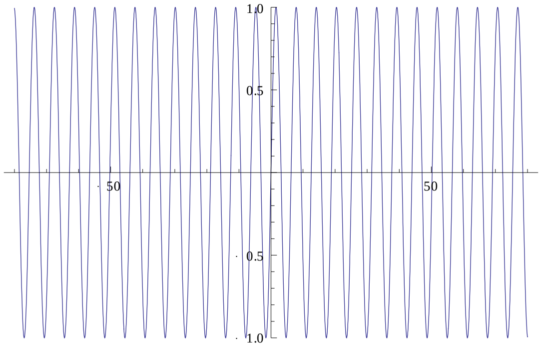
Short-Pulsed Laser



Long-Pulsed Laser

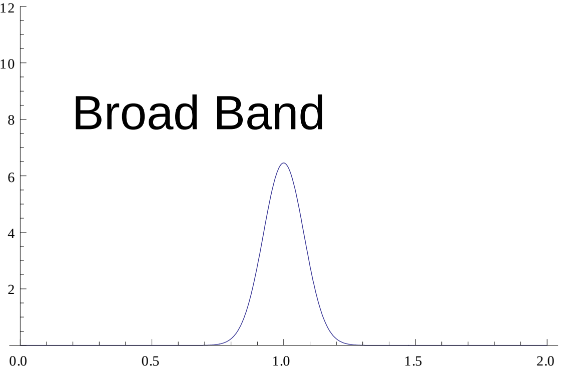


CW Laser

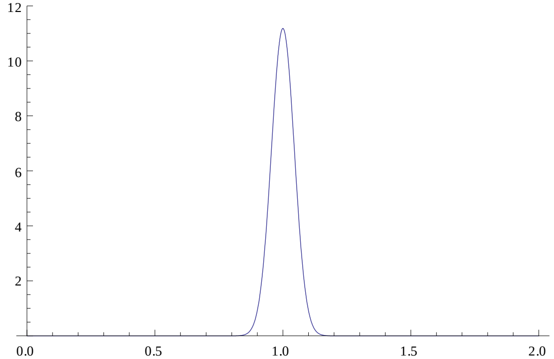


Time Domain

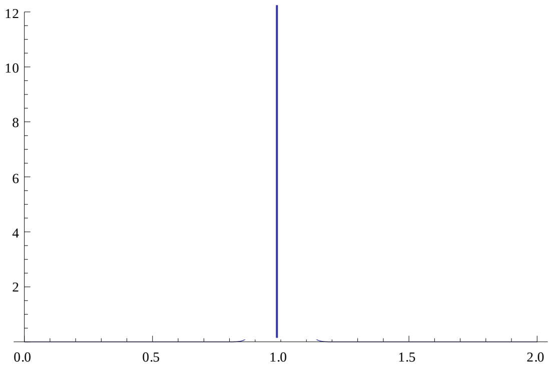
Broad Band



Narrow Band

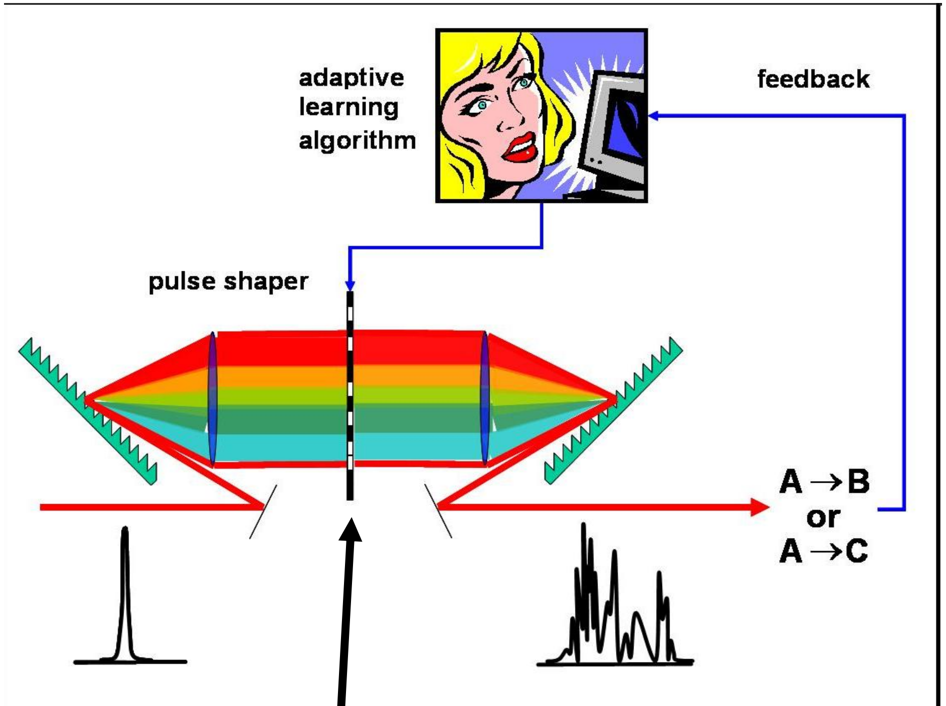


Monochromatic



Frequency Domain

Pulse Shaper



Fourier Expansion

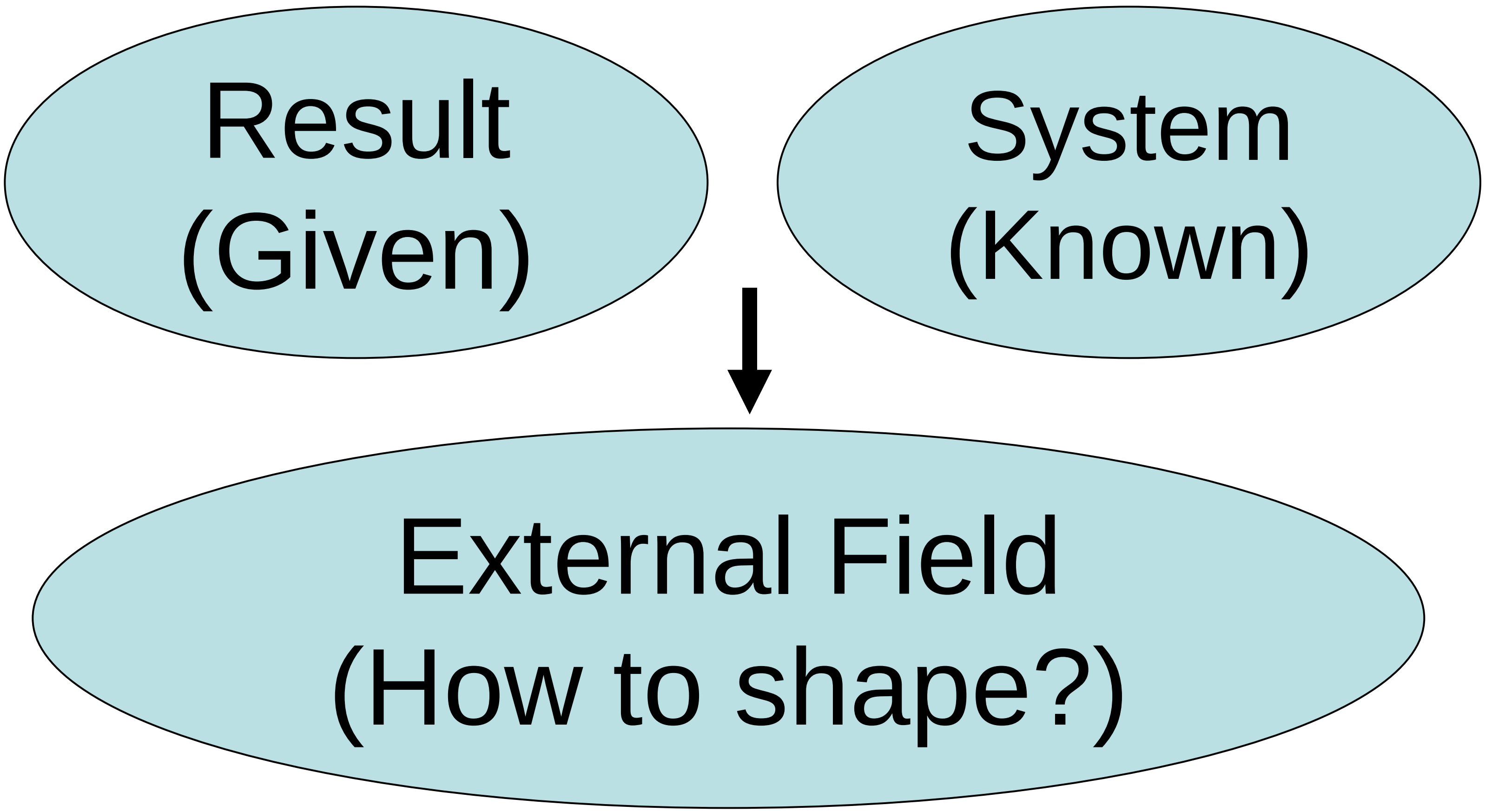
$$f(t) = \sum_j c_j e^{i(j\omega t)}$$

Control of the Fourier coefficients

LCD
(Transmittance & Refractive indexes are controlled.)

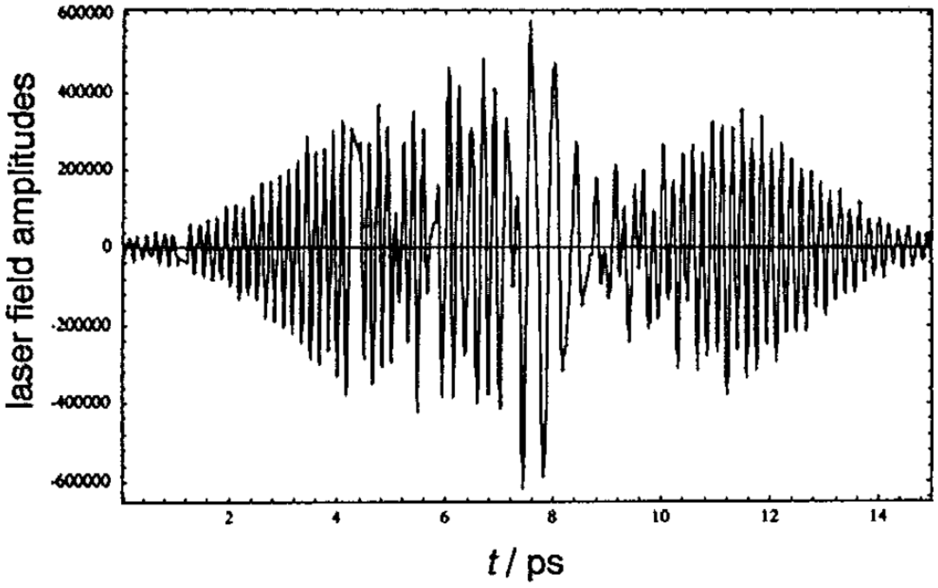
What is the optimal shape?

Quantum Control



Numerical optimization of the laser field for isomarization trimethylenimine

M. Sugawara and Y. Fujimura J. Chem. Phys. 100 5646 (1994)



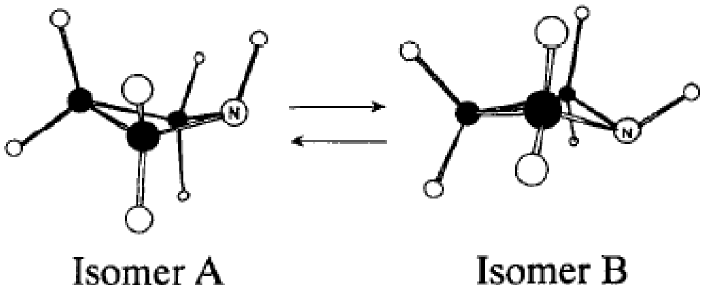


FIG. 1. Two different conformations of trimethylenimine.

Complicated Wave Form

Why this shape?
Any simpler?

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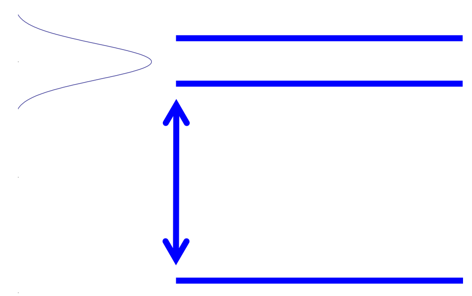

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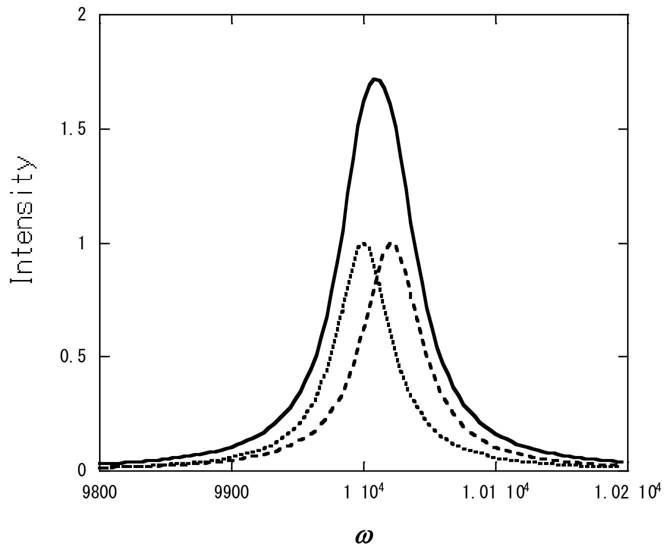


Schrodinger Equation

Time Dependent Schrodinger Equation

$$i\hbar \frac{d}{dt}\Psi\left(r,t\right) = H\left(r,t\right)\Psi\left(r,t\right)$$

If the Hamiltonian is time-independent

Time dependent part

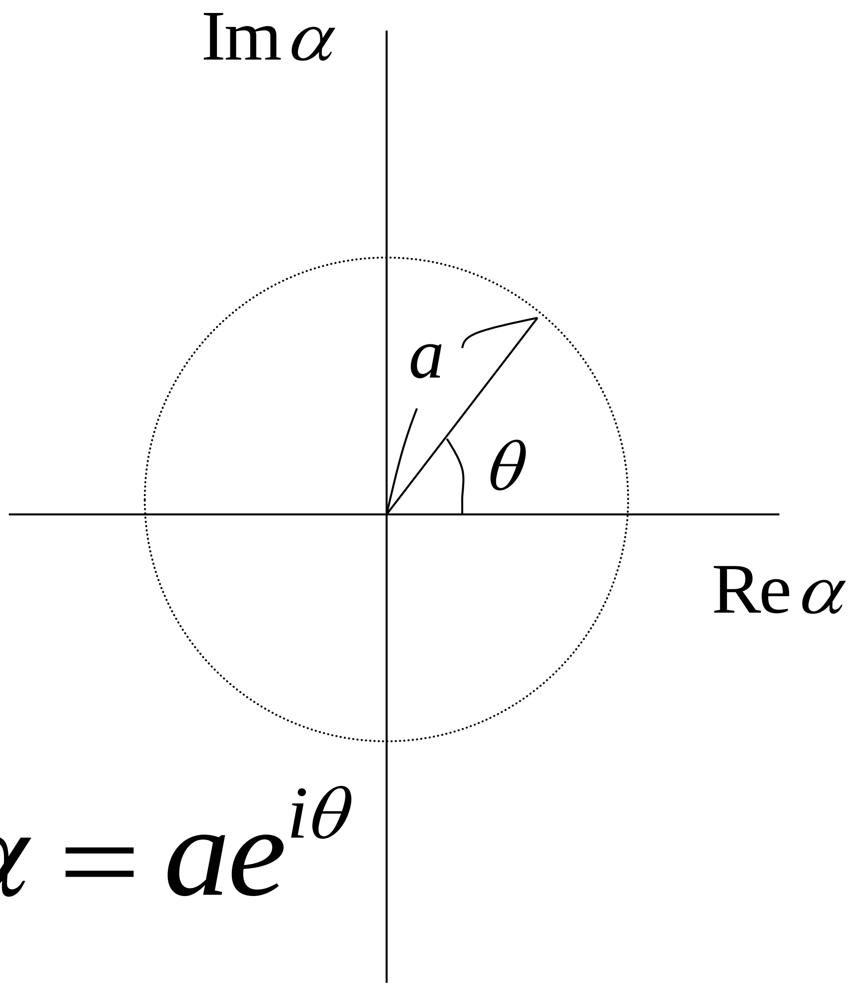
$$\frac{d}{dt}H\left(r,t\right) = 0, \quad \Psi\left(r,t\right) = e^{-iEt/\hbar}\Phi\left(r\right)$$

Time independent part
(Eigen Function)

Stationary Schrodinger Equation

$$\left[-\frac{\hbar^2}{2m}\frac{d^2}{dr^2} + V\left(r\right)\right]\Phi\left(r\right) = E\Phi\left(r\right)$$

Exponential Function

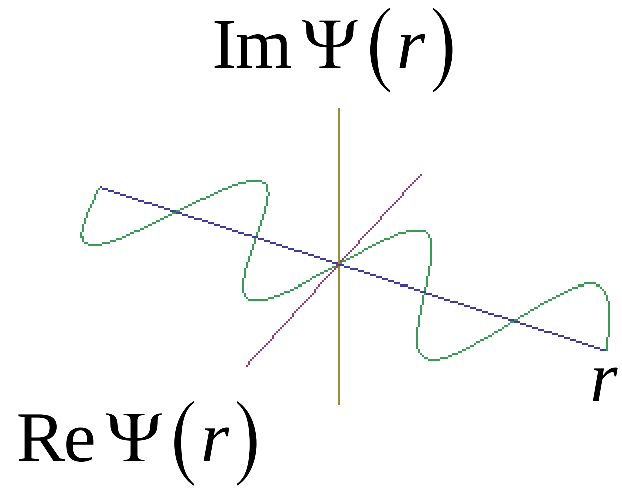


$$\alpha = ae^{i\theta}$$

$$\Psi(r,t) = e^{-iEt/\hbar}\Phi(r)$$

$$\Phi(r) = \sin(r)$$

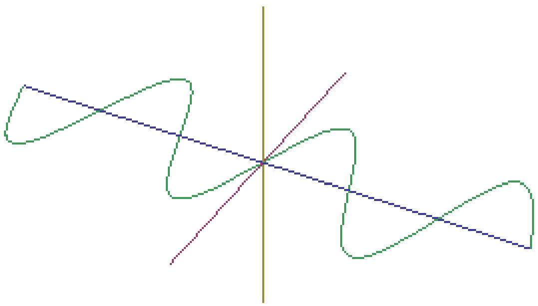
Time evolution of an eigen function

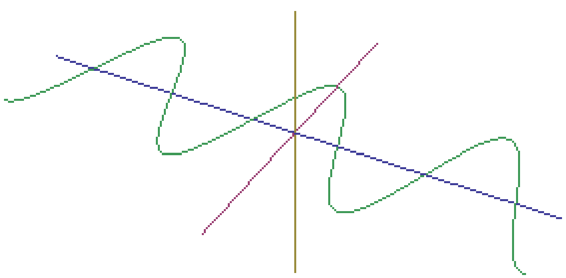


The shape does not change

A Superposition of Eigen States

$$\Psi_1(r) = e^{-iE_1 t/\hbar}\sin(r)$$

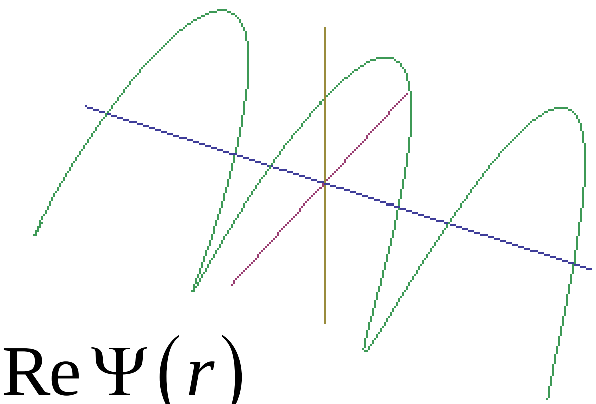




$$\Psi_2(r) = e^{-iE_2 t/\hbar}\cos(r)$$

$$\Psi_1(r) + \Psi_2(r)$$
$$= e^{-iE_1 t/\hbar}\left(\sin(r) + e^{-i(E_2 - E_1)t/\hbar}\cos(r)\right)$$

$\mathrm{Im}\,\Psi(r)$



r

$\mathrm{Re}\,\Psi(r)$

Time dependent shape

Period $T = \dfrac{2\pi\hbar}{E_2 - E_1}$

Time-Dependent Problems

$$H(r,t) = H_0(r) + H'(r,t)$$

Weak Field Limit

Time Dependent Perturbation

Time-Dependent Perturbation Theory

The coupled Schrodinger Equations

$$\Psi(t) = \sum_j c_j(t)\psi_j(t)$$

The Initial Conditions

$$c_j(-\infty) = \begin{cases} 1 & (j = 0) \\ 0 & (j \neq 0) \end{cases}$$

Perturbative Transition Amplitude

$$c_j(t) = \begin{cases} 1 & (j = 0) \\ \int_{-\infty}^{t} \langle \psi_j | H'(\tau) | \psi_0 \rangle d\tau & (j \neq 0) \end{cases}$$

Perturbation Theory for Laser Dynamics

Dipole approximation for light-matter interaction

$$H'(r,t) = -\vec{\mu}\cdot\vec{F}(t)$$

Matrix Elements

$$\langle\psi_0|H'(\tau)|\psi_j\rangle = \exp\left[\frac{i}{\hbar}\left(E_j - E_0\right)t\right]\vec{\mu}_{0j}\cdot\vec{F}(t)$$

$$\vec{\mu}_{0j} = \langle\varphi_0|\vec{\mu}|\varphi_j\rangle \quad \left(\text{here } \psi_j = e^{iE_j t/\hbar}\varphi_j\right)$$

Transition amplitude

$$c_j(\infty) = \int_{-\infty}^{\infty}\langle\psi_0|H'(\tau)|\psi_j\rangle d\tau = \vec{\mu}_{0j}\cdot\vec{F}(\Delta E)$$

Laser Field in the Energy Domain

$$\vec{F}(\Delta E) = \int_{-\infty}^{\infty} dt \exp\left[\frac{i}{\hbar}(\Delta E)t\right]\vec{F}(t)$$

Linearity of the Perturbation Theory

Additivity

$$H = H_0 + H_1(t) + H_2(t)$$

$$c_j(t) = \int_{-\infty}^{t} \langle \phi_0 | H_a(\tau) + H_b(\tau) | \phi_j \rangle d\tau$$

$$= \int_{-\infty}^{t} \langle \phi_0 | H_a(\tau) | \phi_j \rangle d\tau + \int_{-\infty}^{t} \langle \phi_0 | H_a(\tau) | \phi_j \rangle d\tau$$

Homogeneity

$$H = H_0 + \alpha H'(t)$$

$$c_j(t) = \int_{-\infty}^{t} \langle \phi_0 | \alpha H'(\tau) | \phi_j \rangle d\tau$$

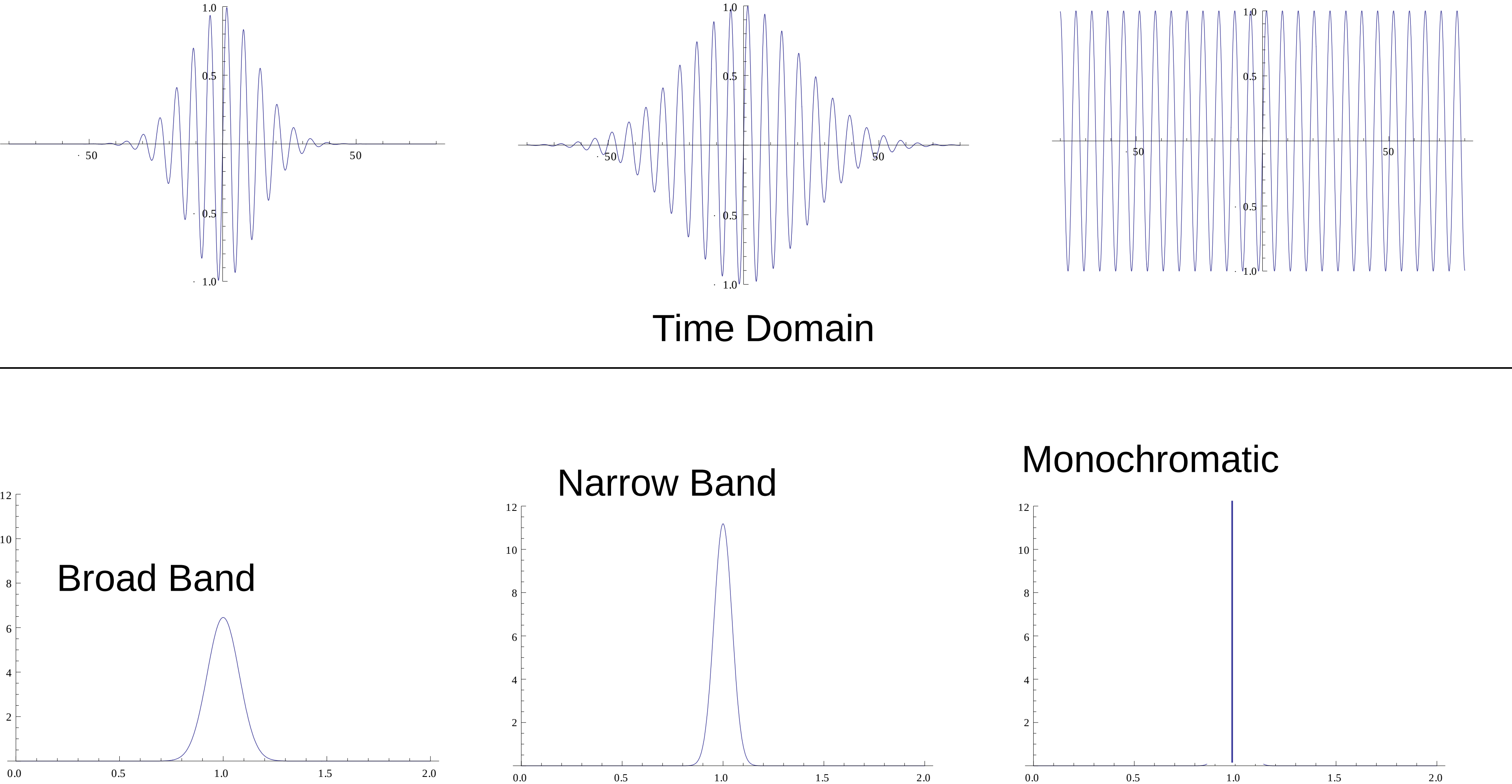
$$= \alpha \int_{-\infty}^{t} \langle \phi_0 | H'(\tau) | \phi_j \rangle d\tau$$

Laser Pulse

Short-Pulsed Laser

Long-Pulsed Laser

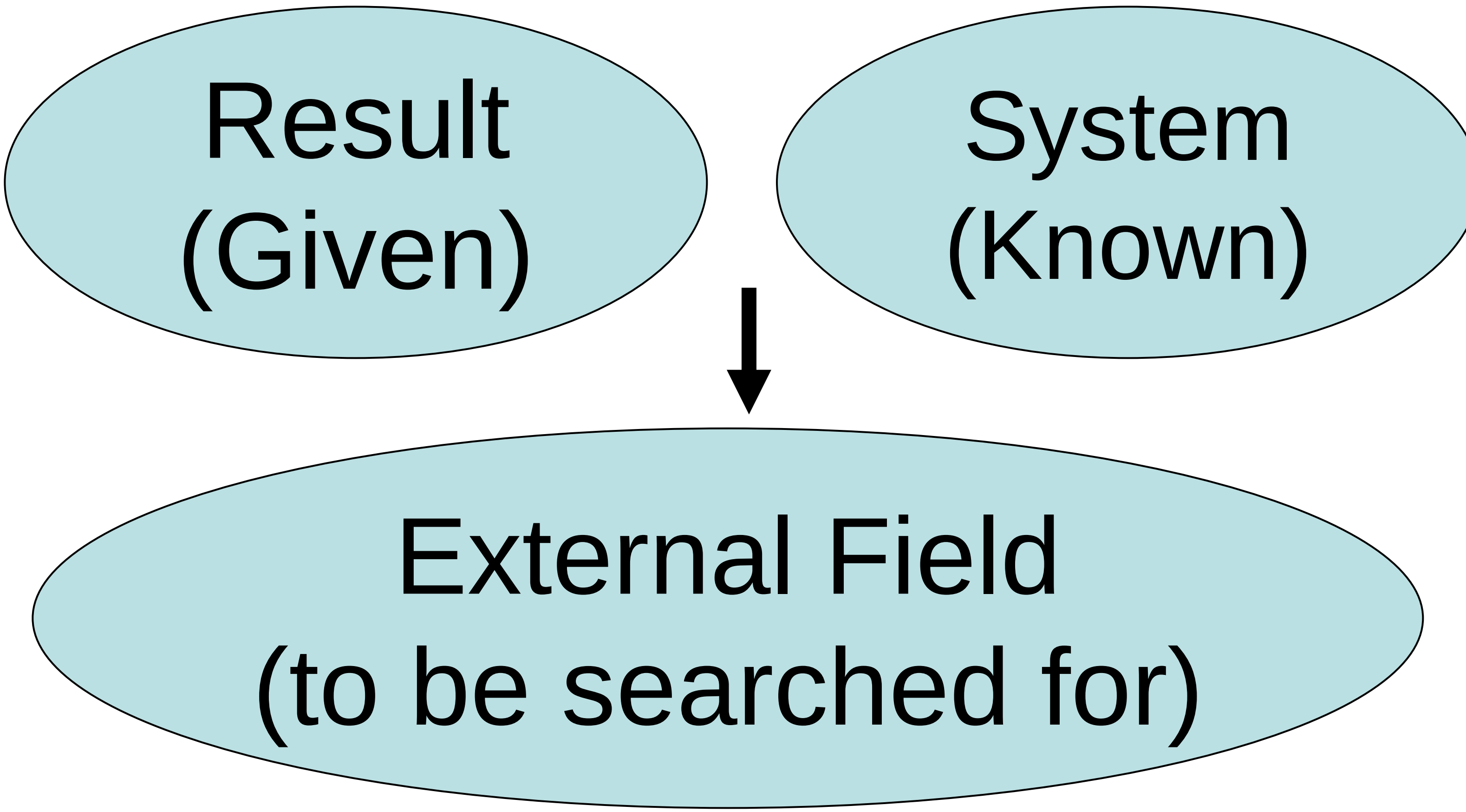
CW Laser



Frequency Domain

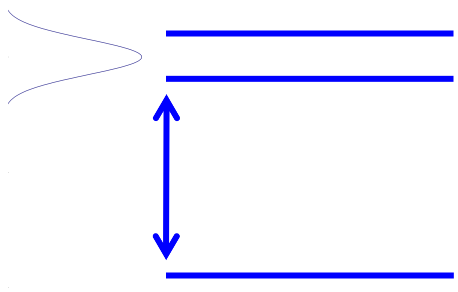
Laser Control of Atomic and Molecular Processes

Quantum Control



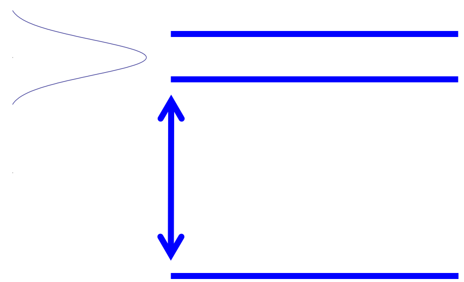
Inverse problem

Ultrafast Selective Excitation



Ultrafast Selective Excitation

- Intense & Ultrafast Laser
  → fast transition
- Nonlinear Process & Broad Bandwidth
  → undesired transition
- Selective Excitation
  → Excite to a desired state
  → Suppressing undesired transition



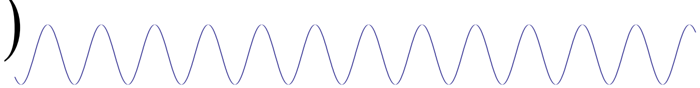
- How much fast selection is possible?

Ramsey Fringe
(Perturbative Double Pulses)

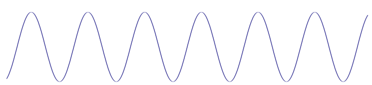
$$\vec{F}(t) = \exp\left[-\frac{t^2}{\Delta T^2}\right]\exp[i\omega t] + \exp\left[-\frac{(t-\tau)^2}{\Delta T^2}\right]\exp\left[i\{\omega(t-\tau)-\delta\}\right]$$

Phase Difference

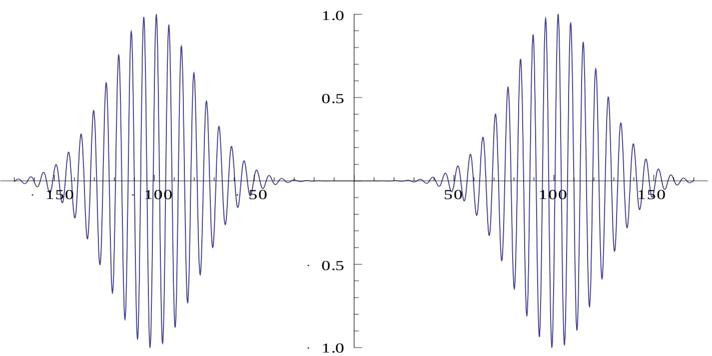
$c_j^1(t)$



Interference



$c_j^1(t)\exp\left[-i(\omega_{01}\tau+\delta)\right]$



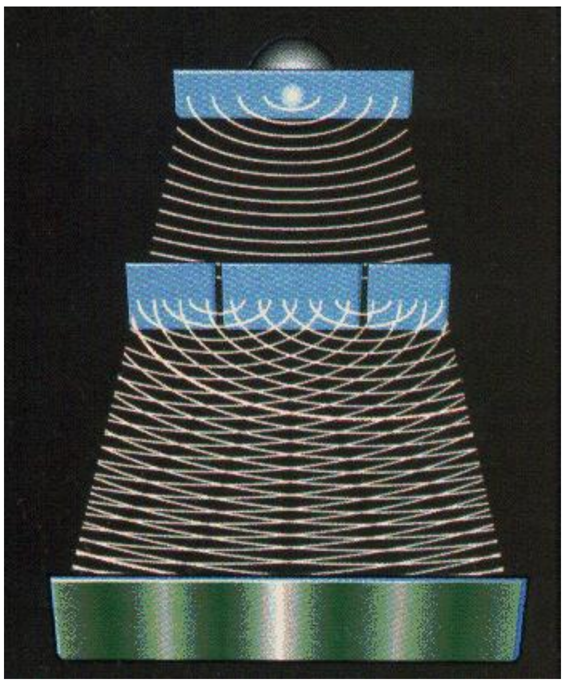
τ : Time delay

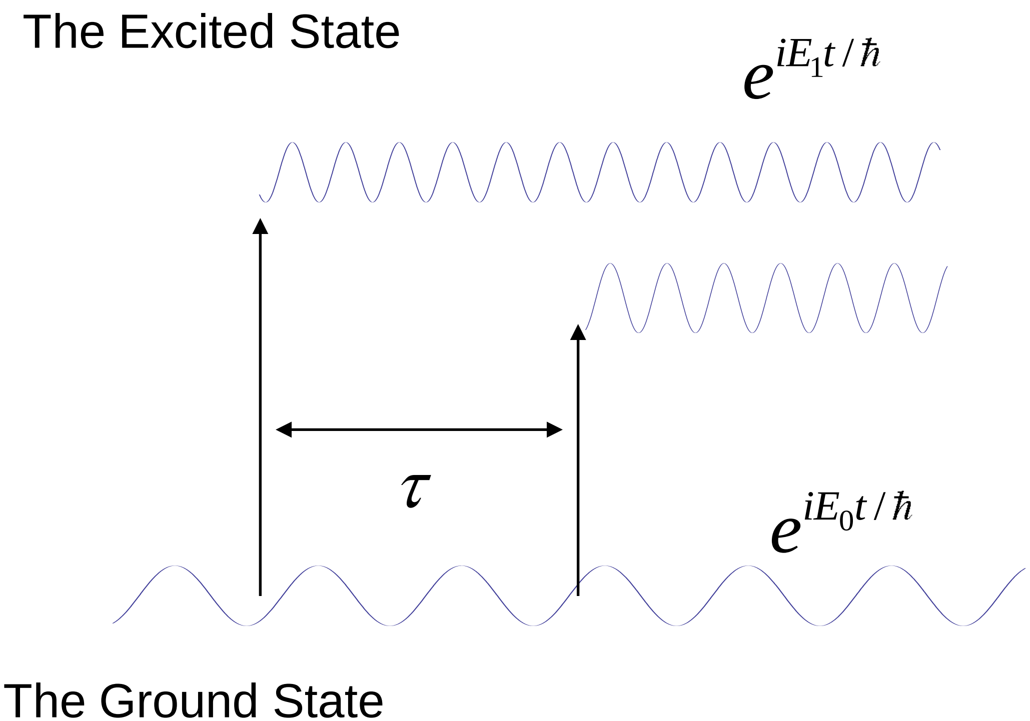
Transition Amplitude

$$c_j(t) = c_j^1(t) + c_j^1(t)\exp\left[-i(\omega_{01}\tau+\delta)\right]$$

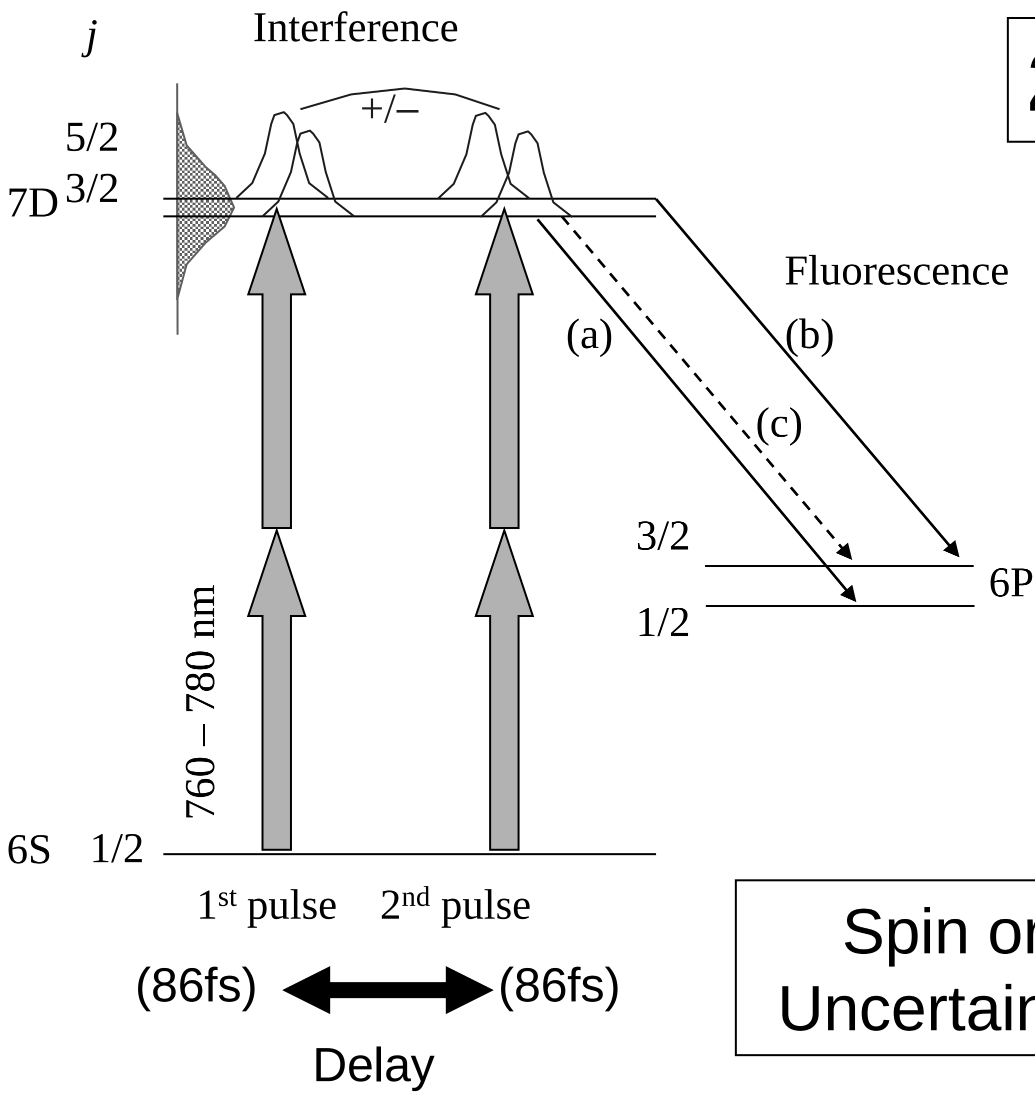
Young's Interference Experiment

Interpretation of $c_j\left(t\right) = c_j^1\left(t\right) + c_j^1\left(t\right)\exp\left[-i\left(\omega_{01}\tau + \delta\right)\right]$





Selective Excitation of Cs atom
（Separation of Fine Structure）



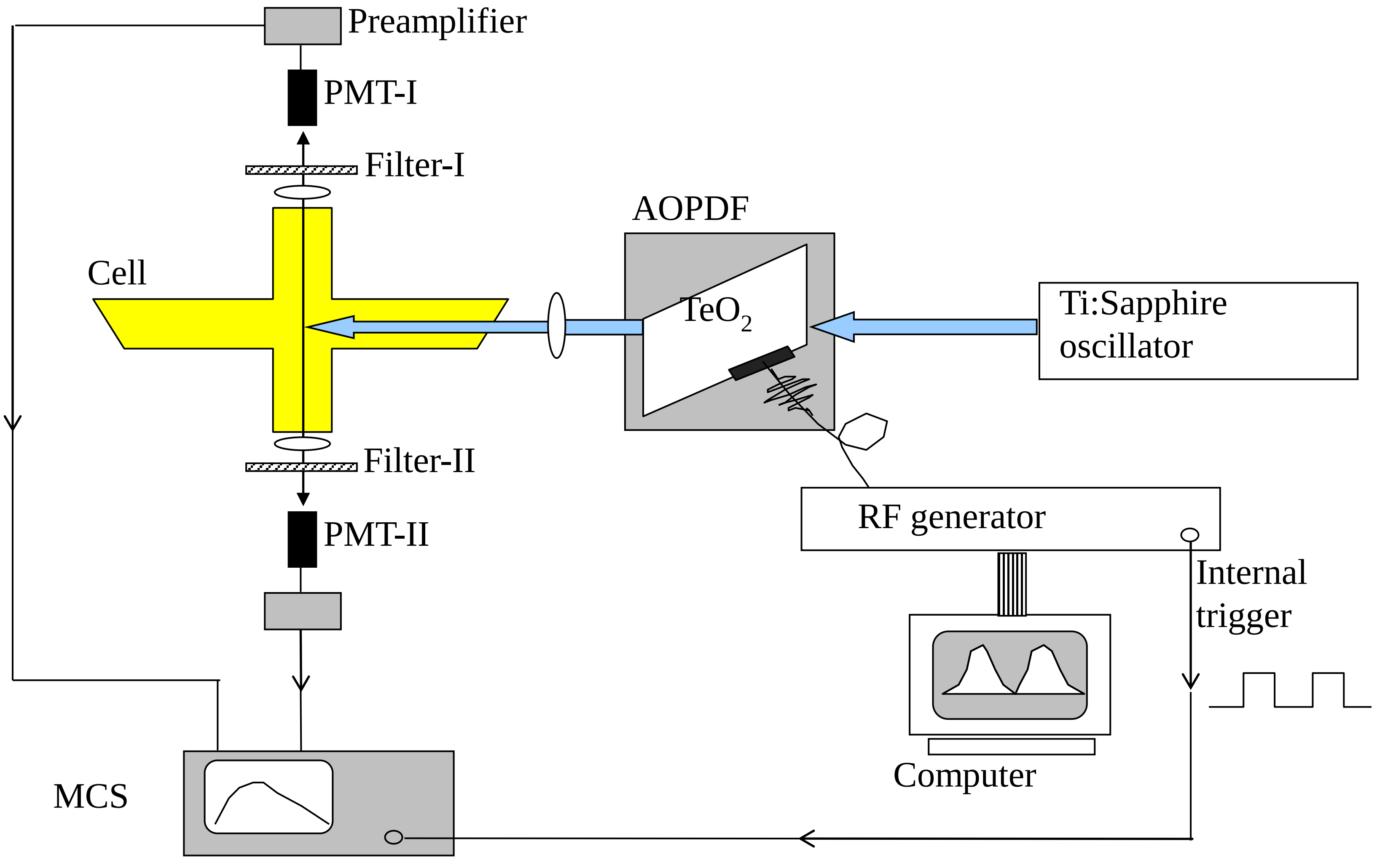
2 pulse interference

- Parameters
  - Time delay
  - phase difference
- Interference
- Suppression of a specific transition

Spin orbit splitting $\Delta E = 21\mathrm{cm}^{-1}$

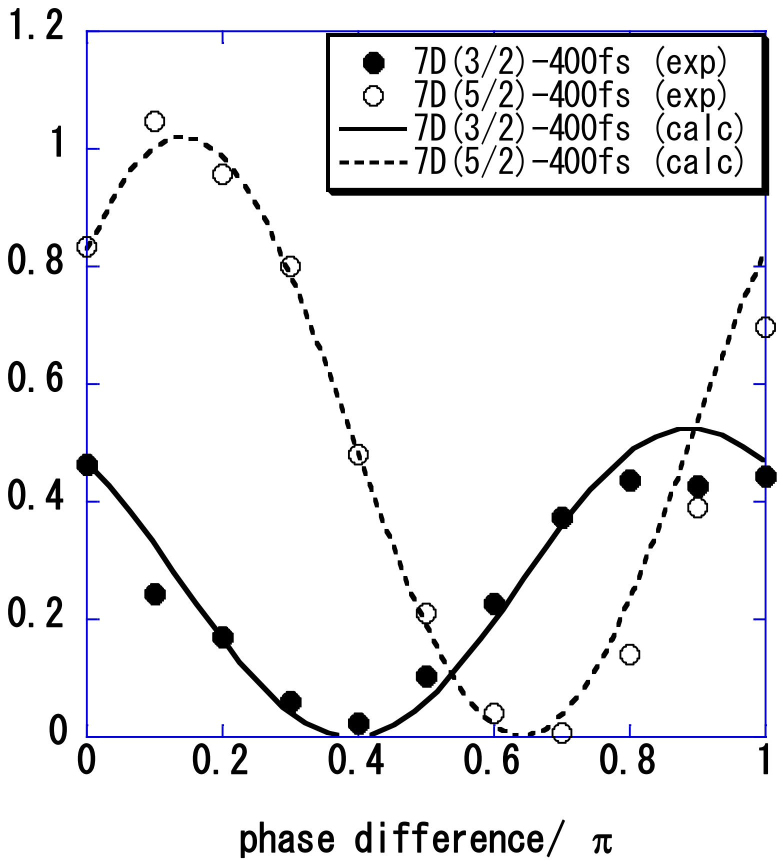
Uncertainty limit $\Delta t = 1/\Delta E = 800\mathrm{fs}$

Experimental Facility

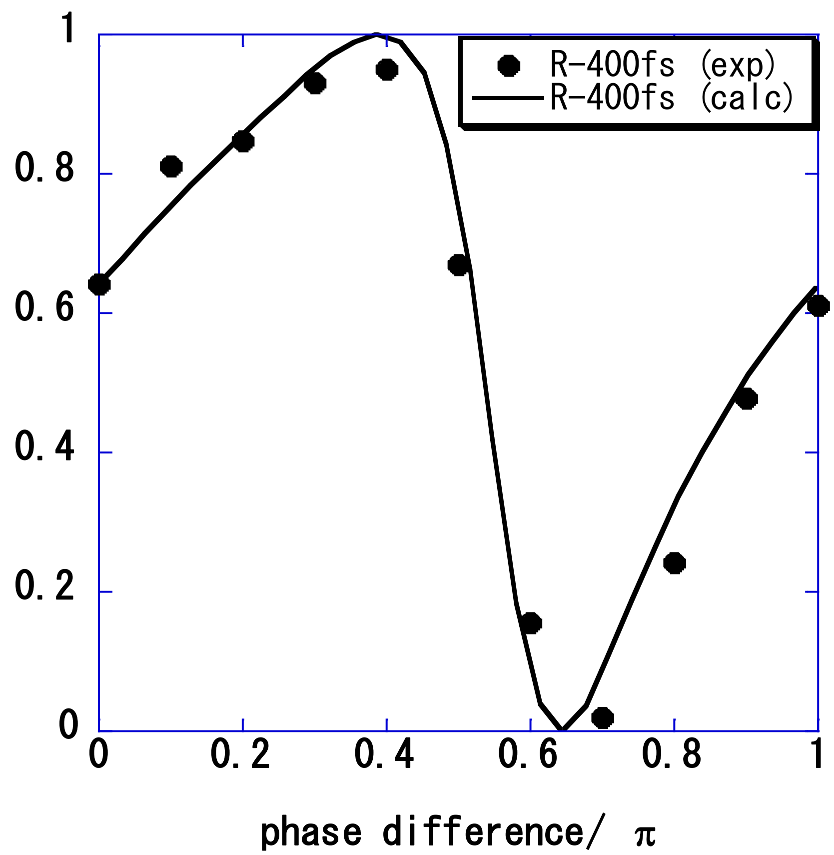


Delay:　400ｆｓ
（Experiment and Theory）

Normalized transition probability

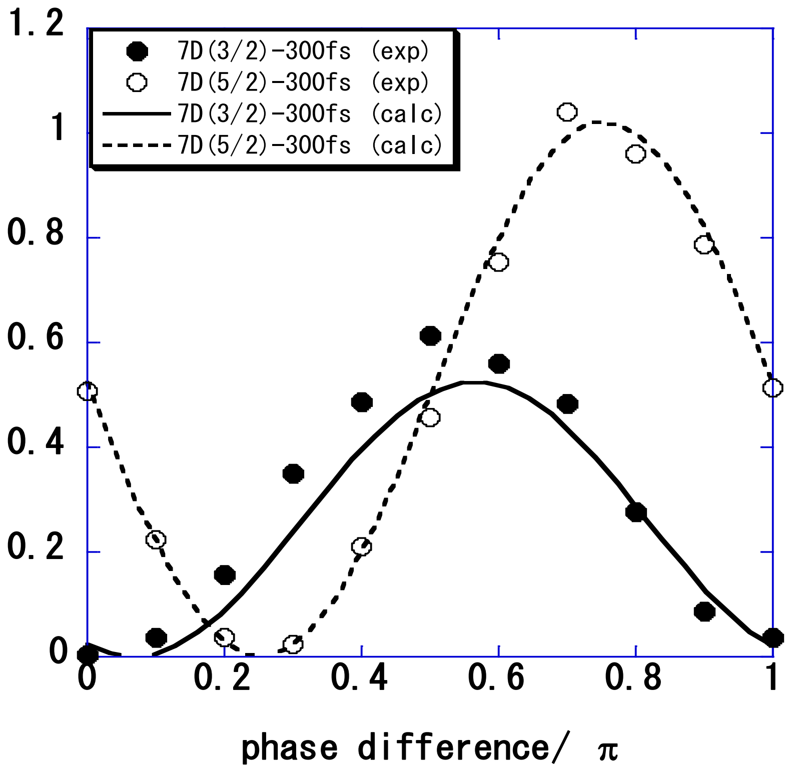


Branching ratio

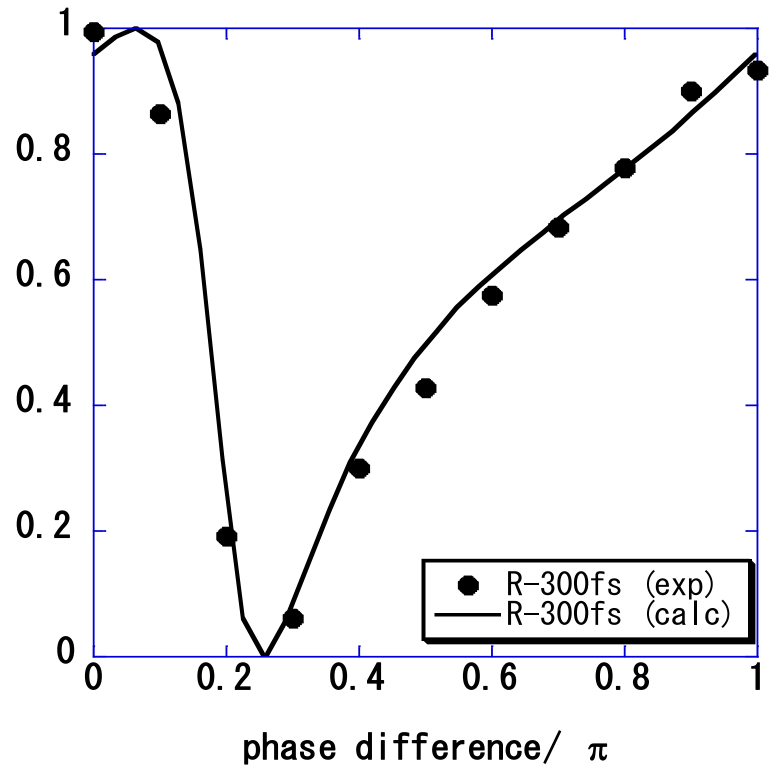


Delay　300fs
（Exp. & Theory）

Normalized transition probability

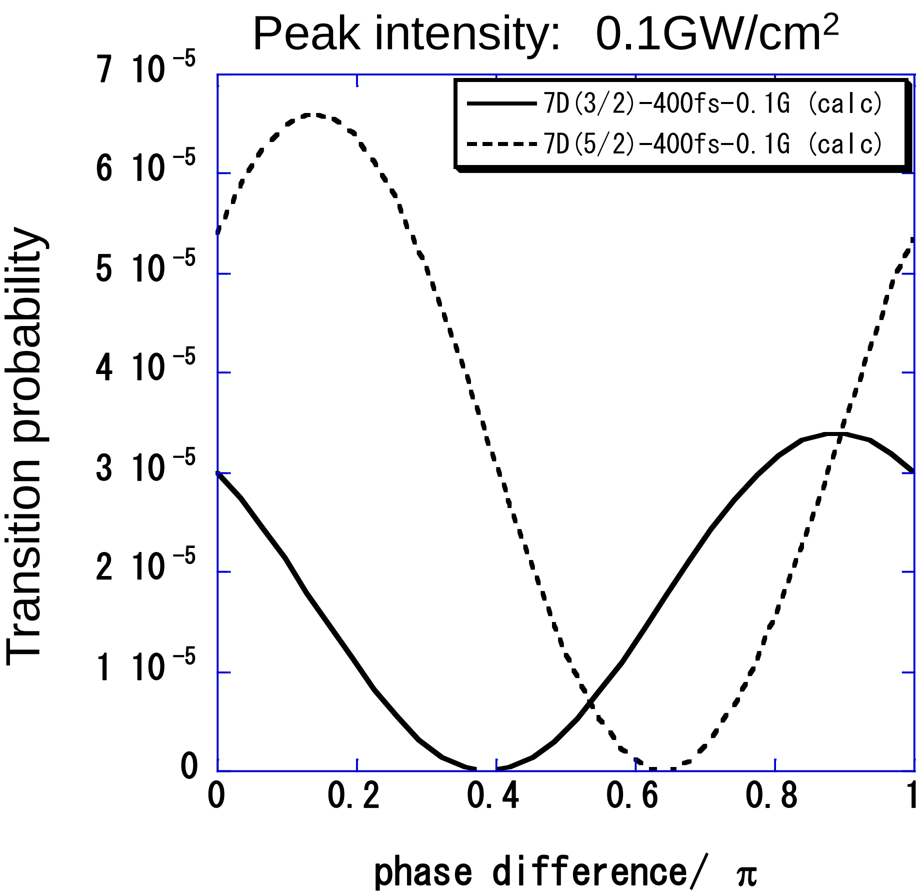


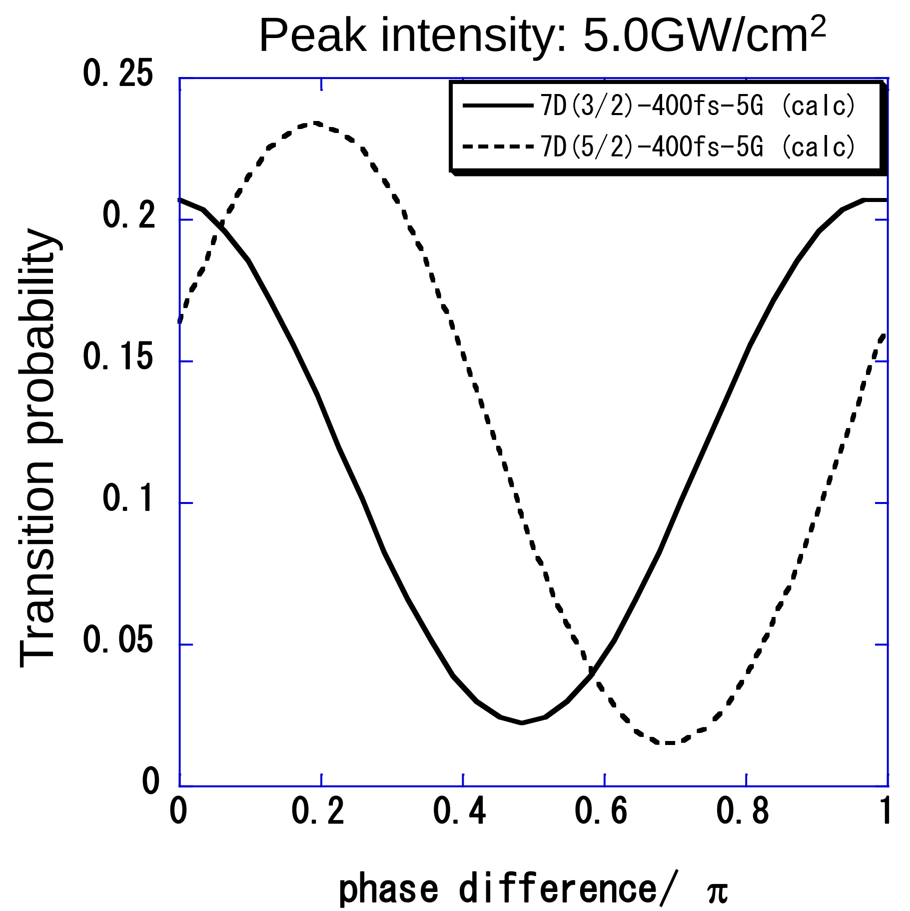
Branching ratio



Selection is possible even when $t < \Delta t = 1/\Delta E = 800\text{fs}$

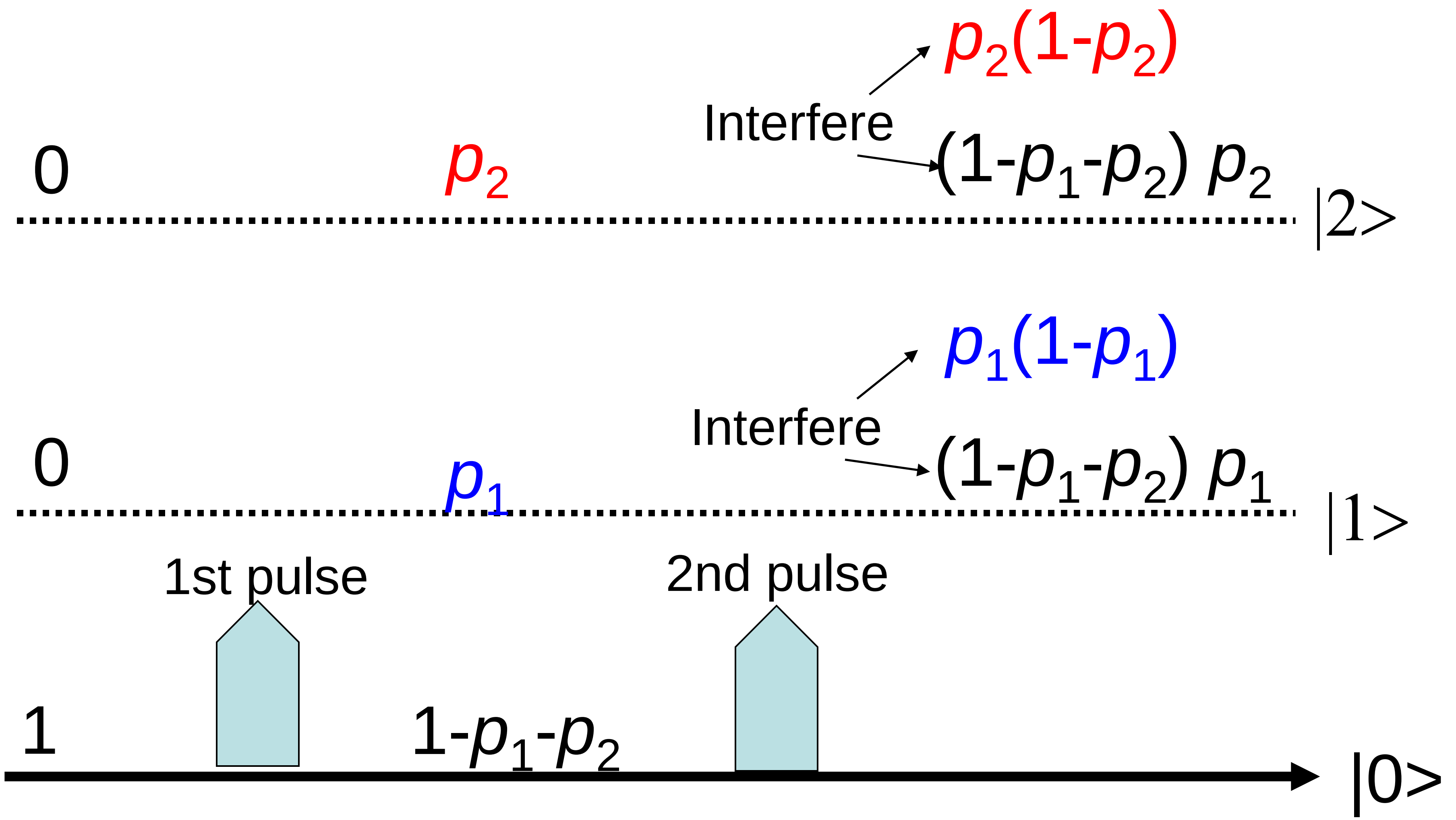
Breakdown of the Selectivity
(Theoretical simulation)





Large transition probability → bad selectivity（nonlinear effect）

Breakdown of the selectivity



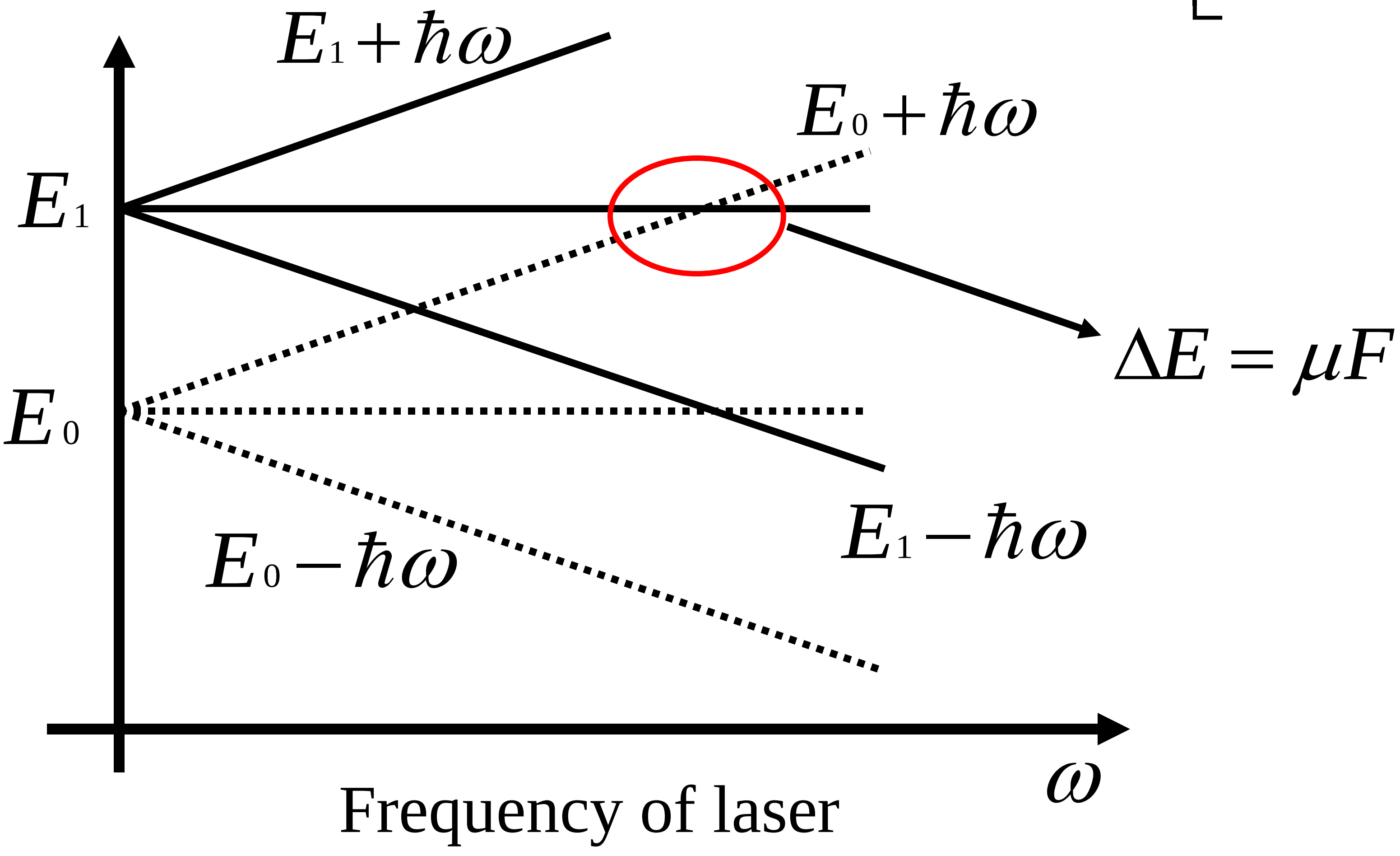
Selection → $p_1, p_2 << 1$ （Linearity）

Non-Perturbative Selective Excitation

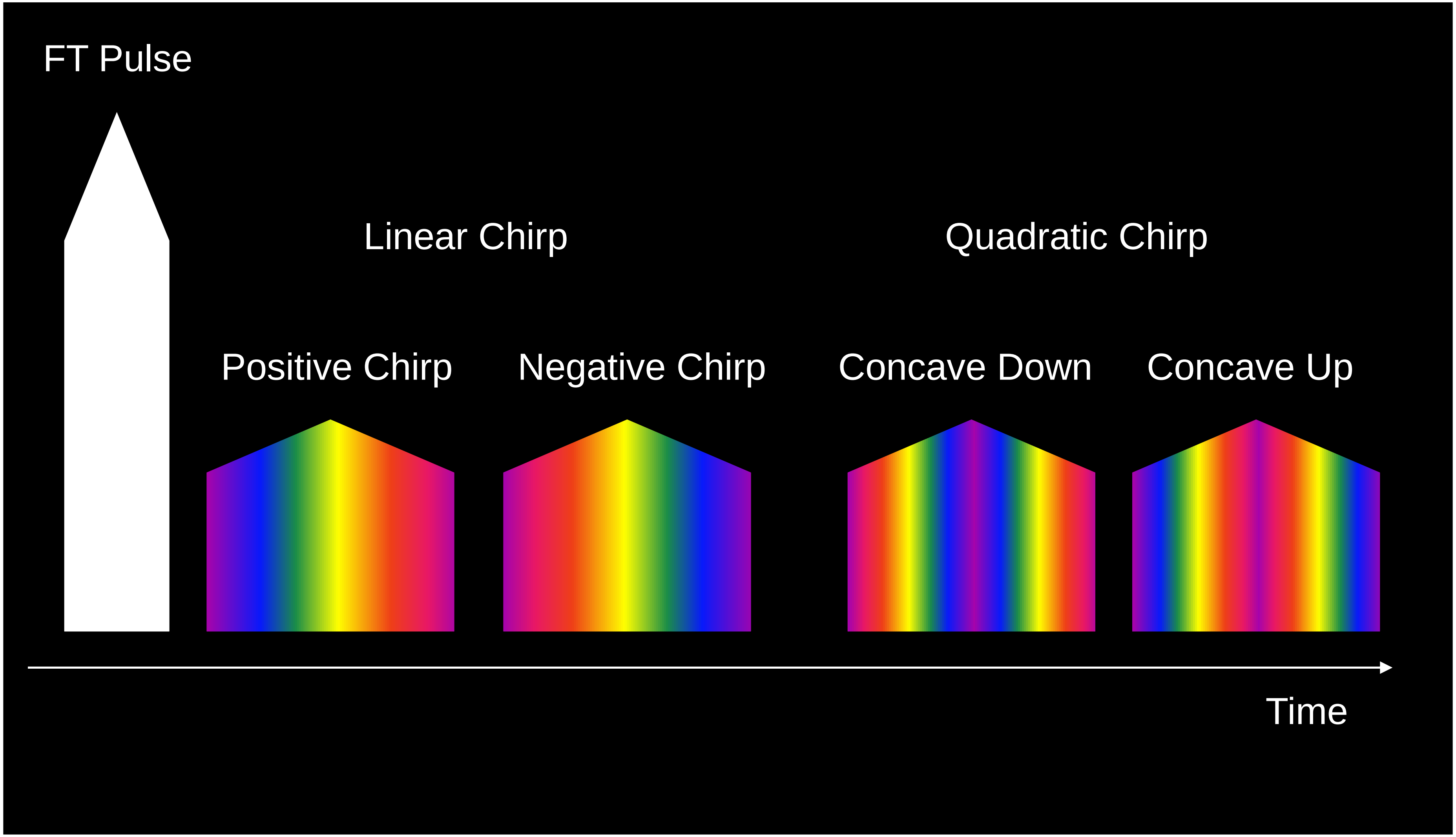
Quadratic Chirping

Quasi-state diagram

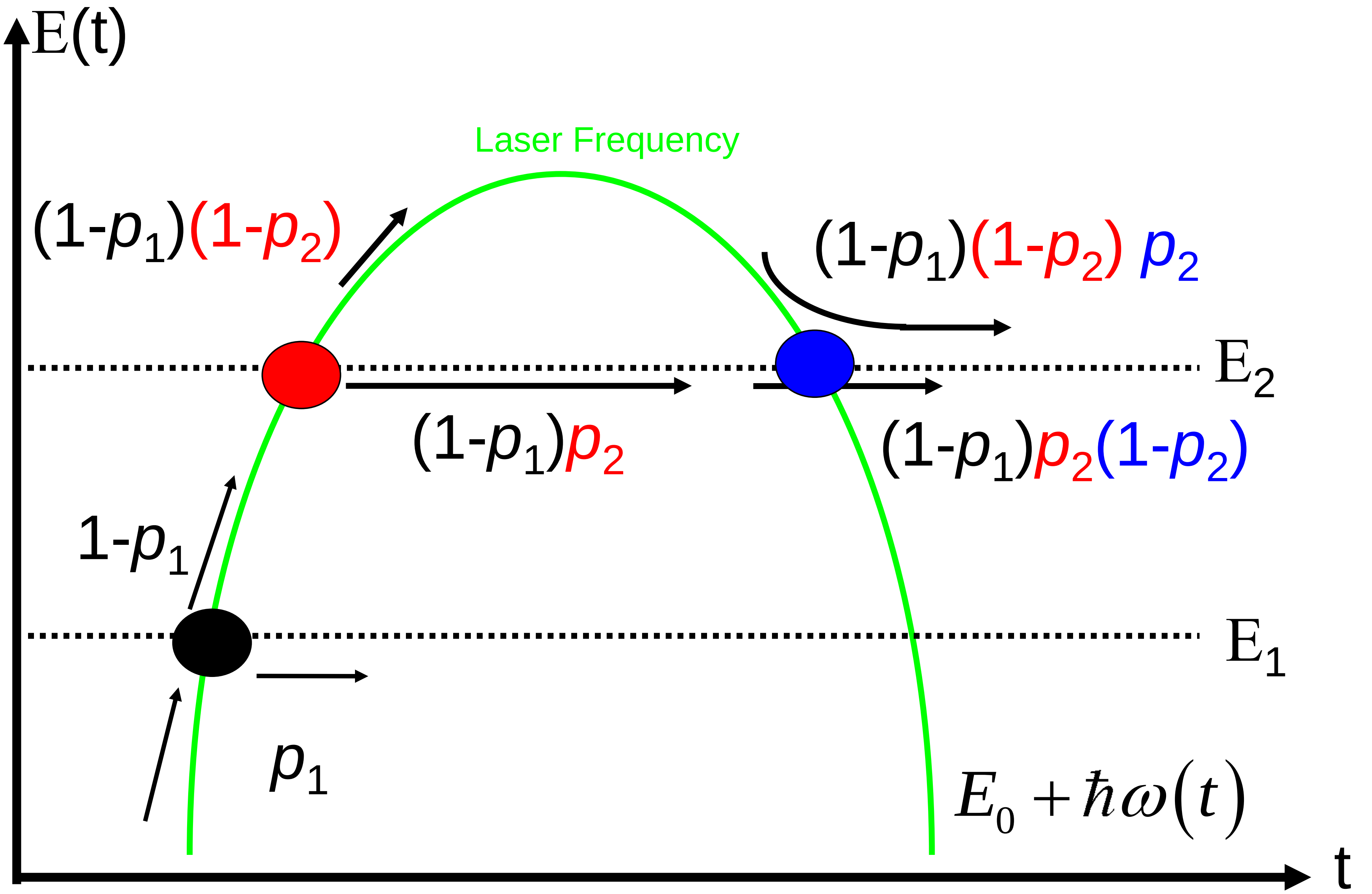
$$H' = \mu F(t)\cos\left[\int^{t}\omega(t')\,dt'\right]$$



Chirping
(time dependent frequency)

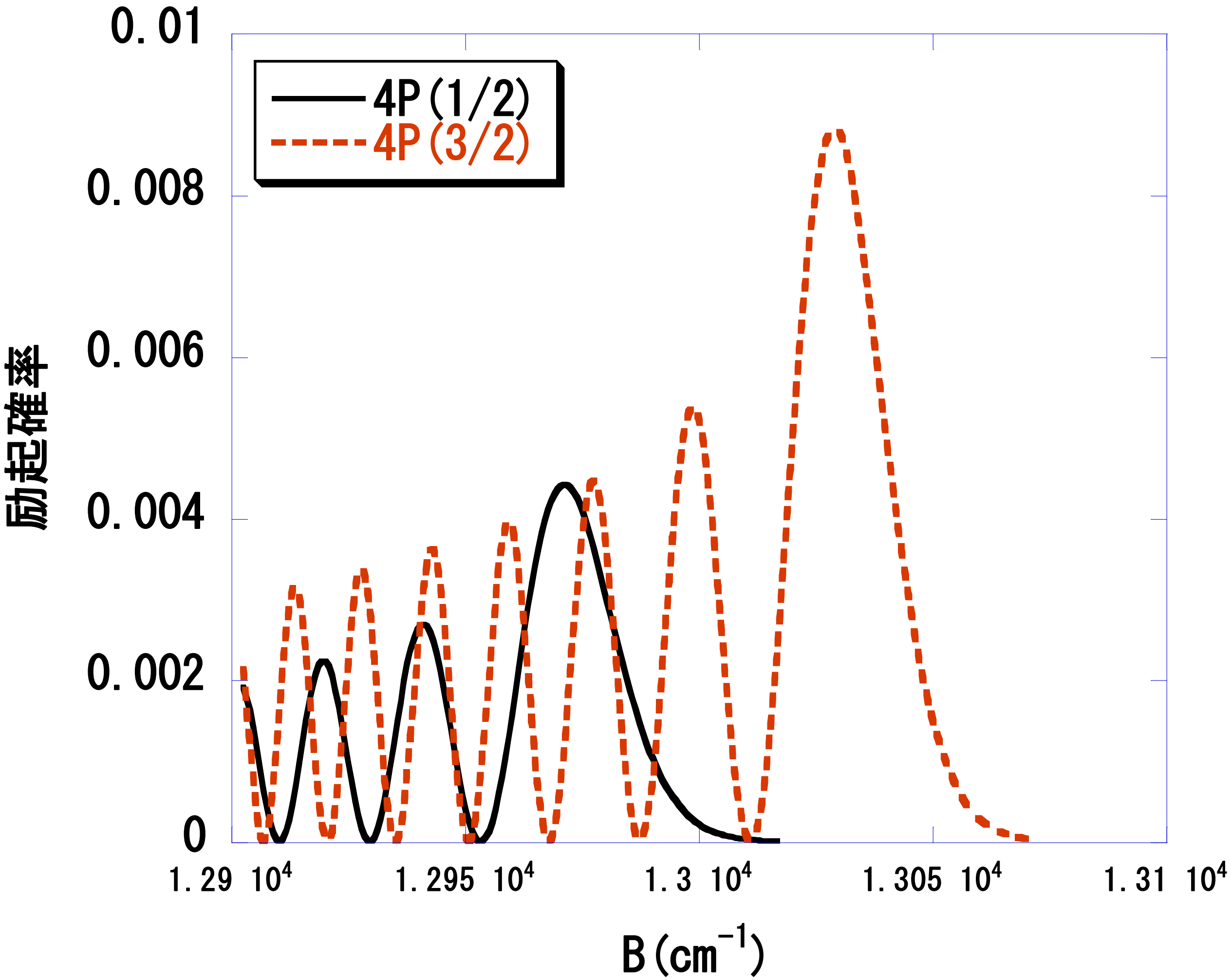


Selective Excitation by Quadratic chirping

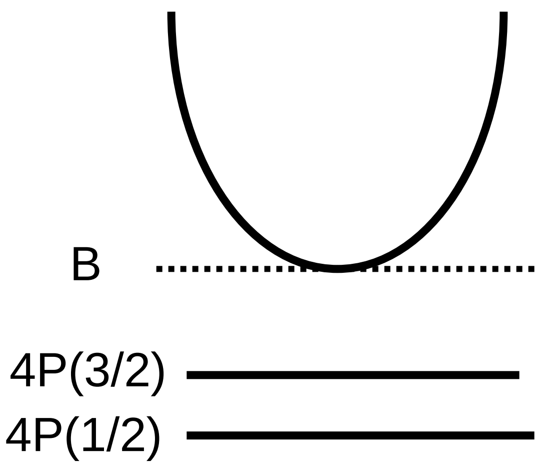


Selective Excitaion of K atom by Quadratic chirping
(Simulation)

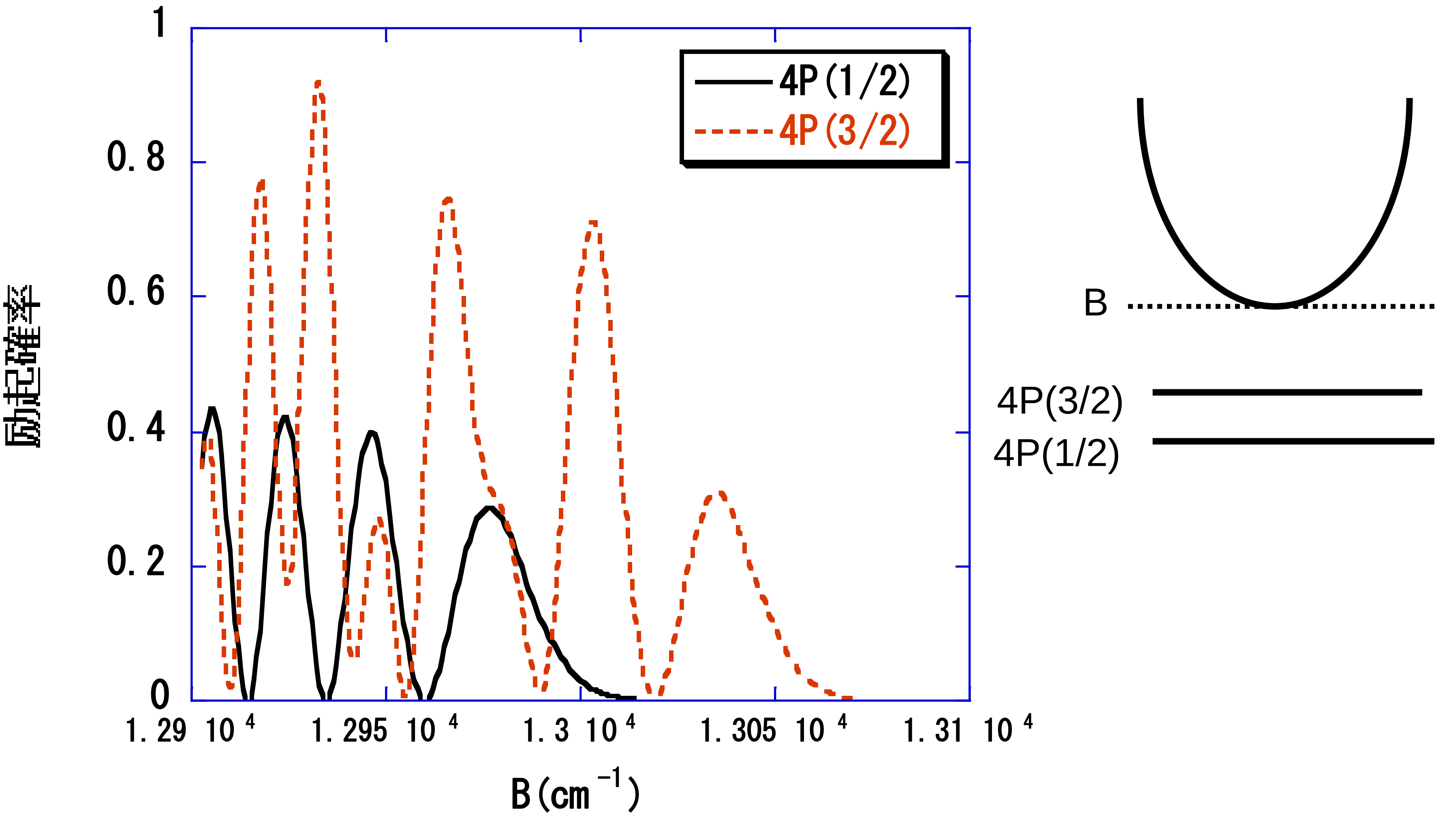
Perturbative region (1 MW/cm^2)



Small Probability
↓
Both selective



High Intensity (0.125 GW/cm²)



Upper level (Red)

↓

Incomplete destruction

Lower level (Black)

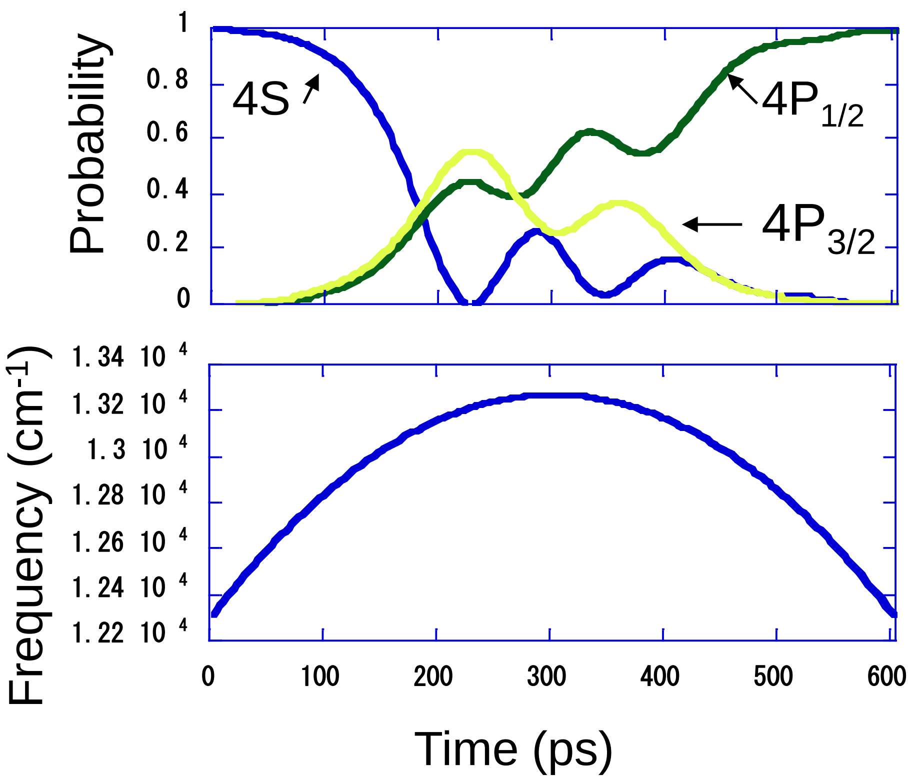
↓

Complete destruction

Complete selective excitation of K atom

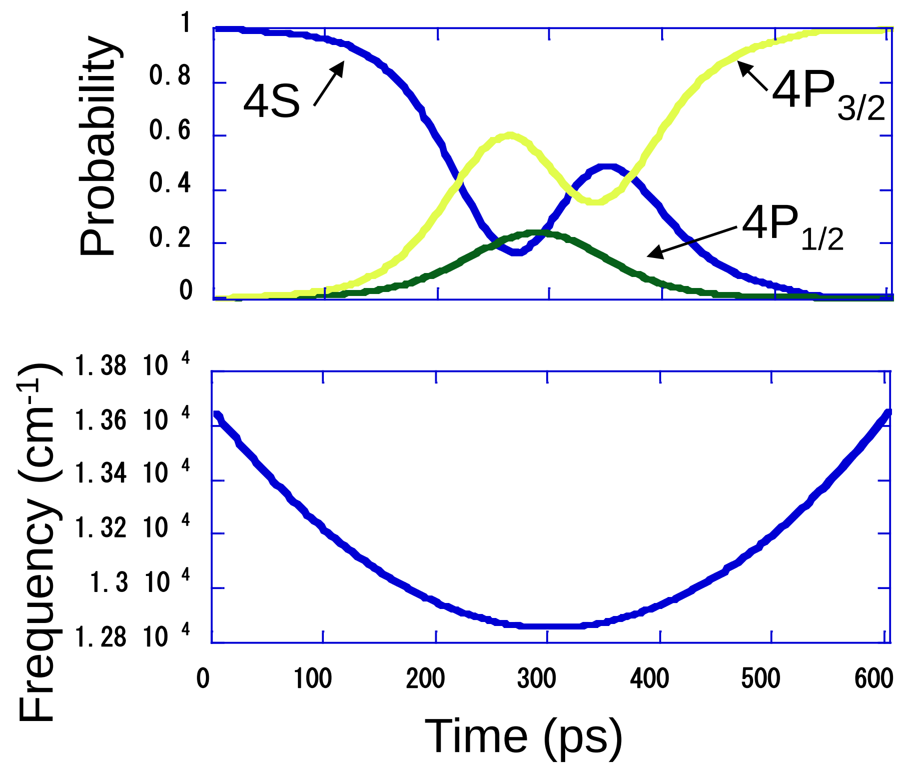
$4S \rightarrow 4P_{1/2}$ Excitation

Intensity 0.36 GW/cm2
Bandwidth 973 cm-1



$4S \rightarrow 4P_{3/2}$ Excitation

Intensity 0.125 GW/cm2
Bandwidth 803 cm-1



Complete & Selective $\Rightarrow$ Transition time ~ $1/\Delta E$

Selective Excitation

- Selection utilizing interference

- Two Pulse Sequence (Ramsey)

  Perturbative (Small Probability)

  Can be faster than the uncertainty limit

- Quadratic Chirping

  Non-perturbative (Large Probability)

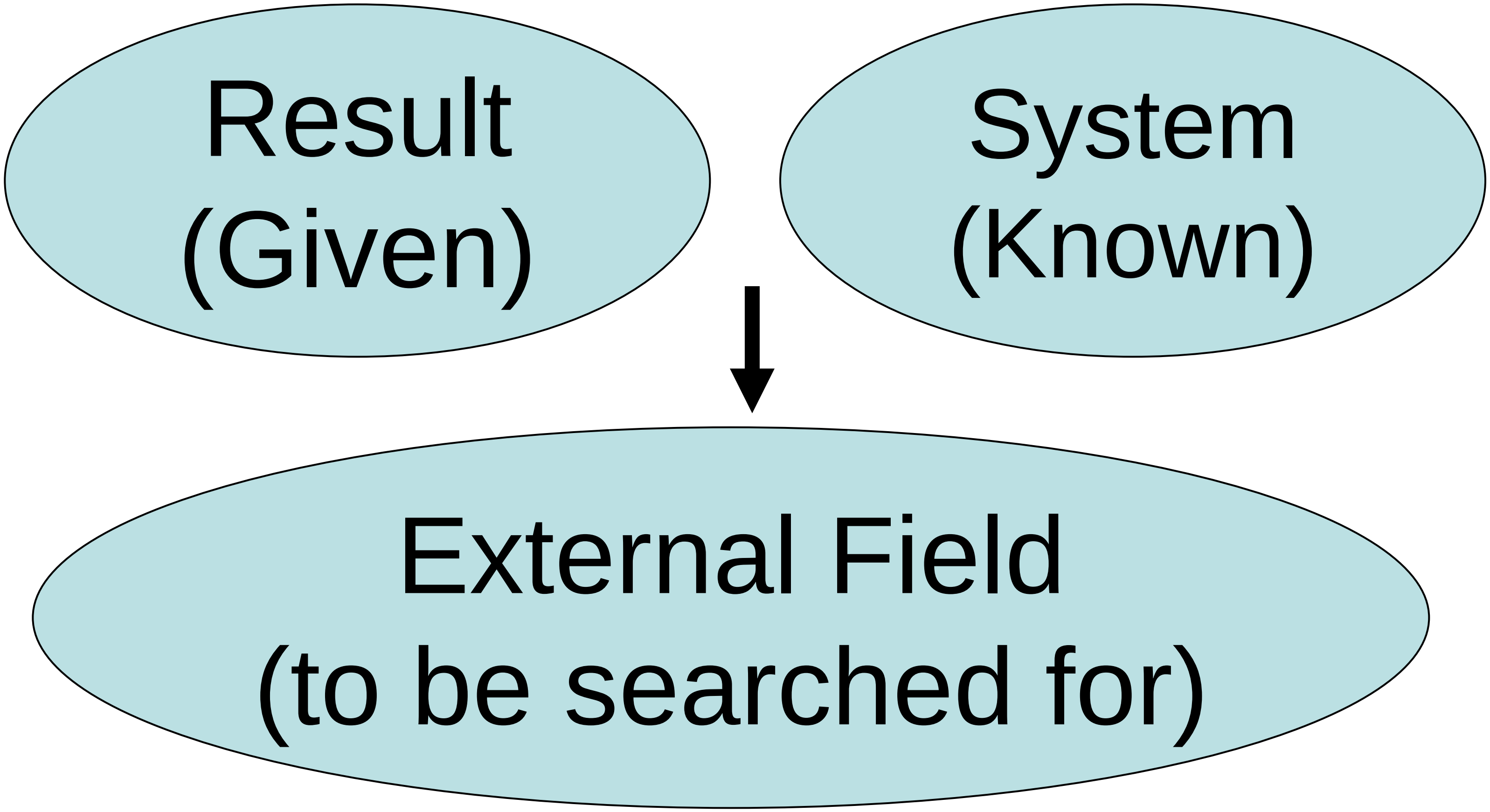
  Complete & Selective Excitation

  (As fast as the uncertainty limit)

Spectroscopy Utilizing Quantum Control

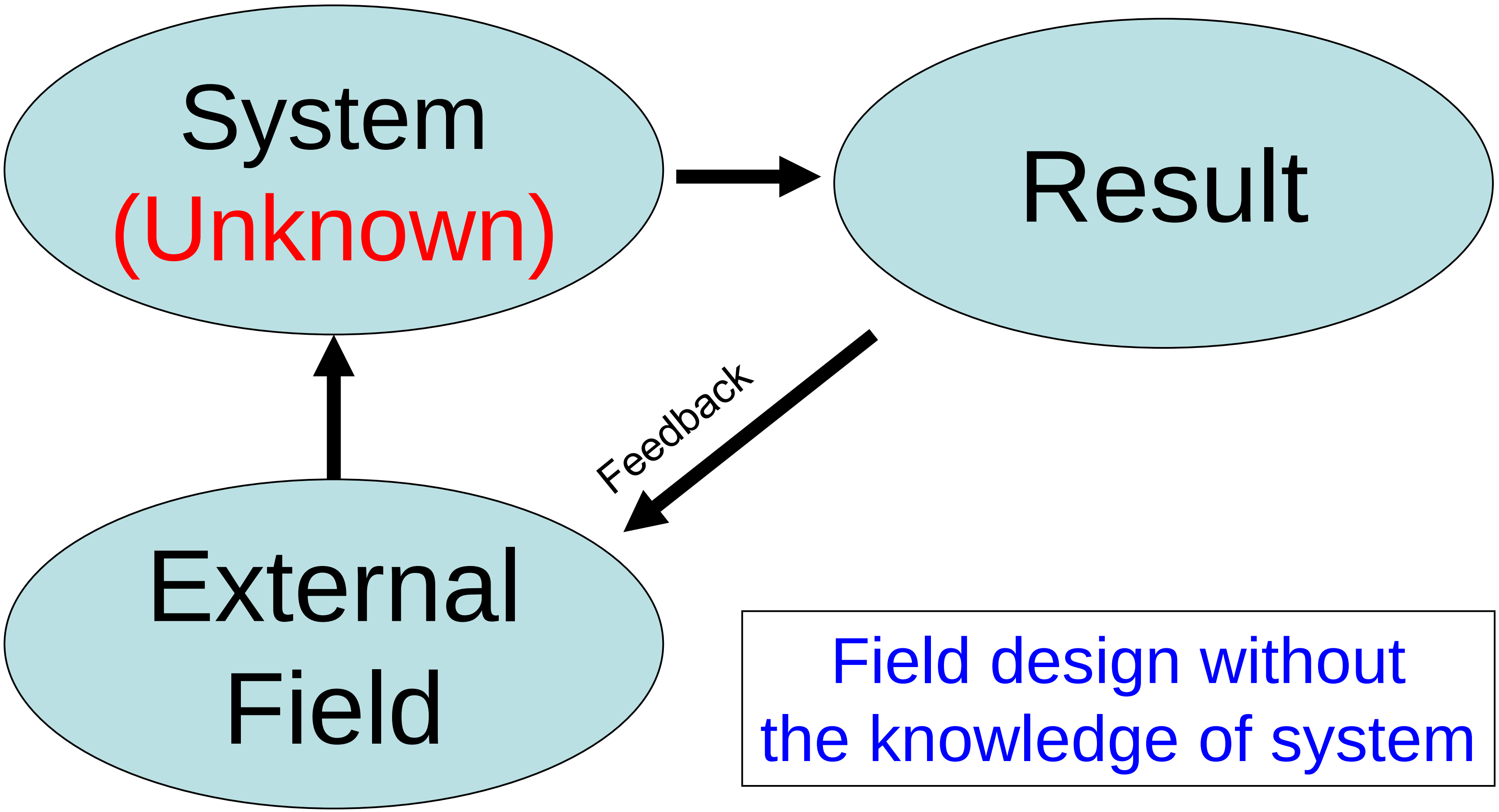
Spectroscopy for short-lived resonance states

Quantum Control



Inverse problem

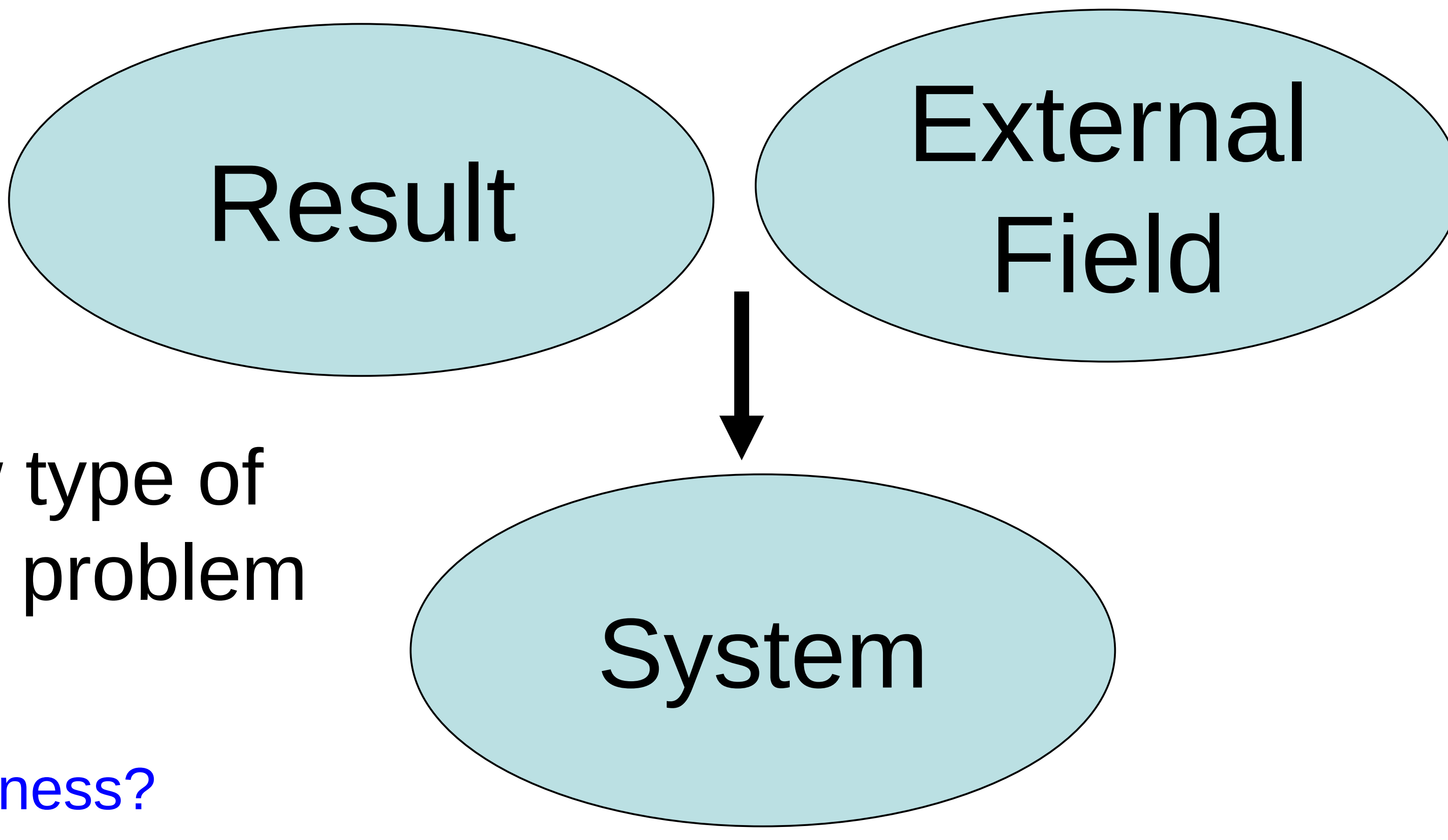
Feedback quantum control
(Experiment)



Field design without
the knowledge of system

Feedback spectroscopy

System information is obtained from the optimal external field



A new type of inverse problem

Uniqueness?

Feedback Spectroscopy

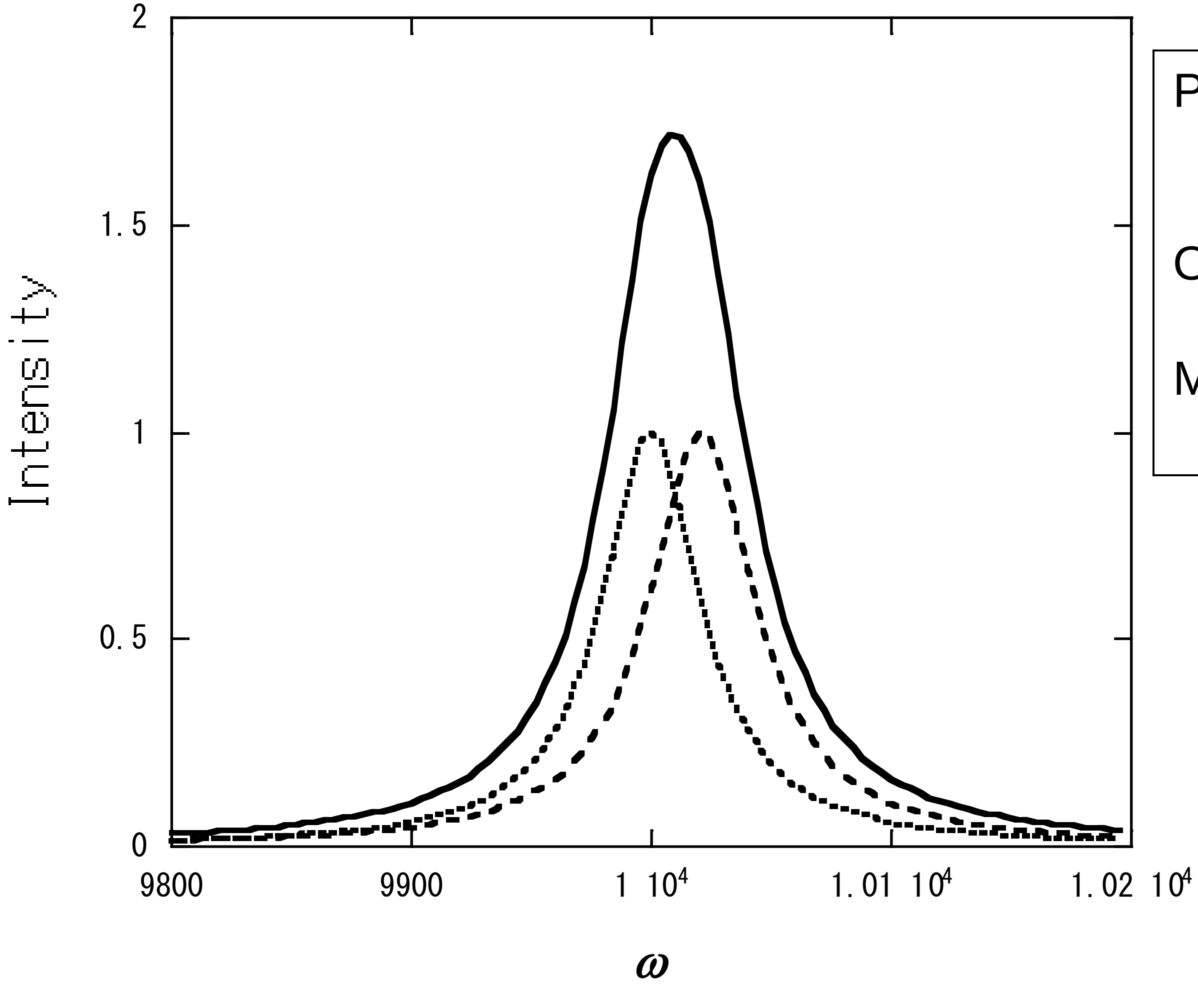
Benefits

- Utilizing complicated laser pulse
- Creation of special quantum states

Uniqueness is required!!

(Proper settings of the problem & constrain)

State Selective Spectroscopy for short lived resonance states



Peaks having the natural width
(dotted & broken lines)

Overlapping resonance

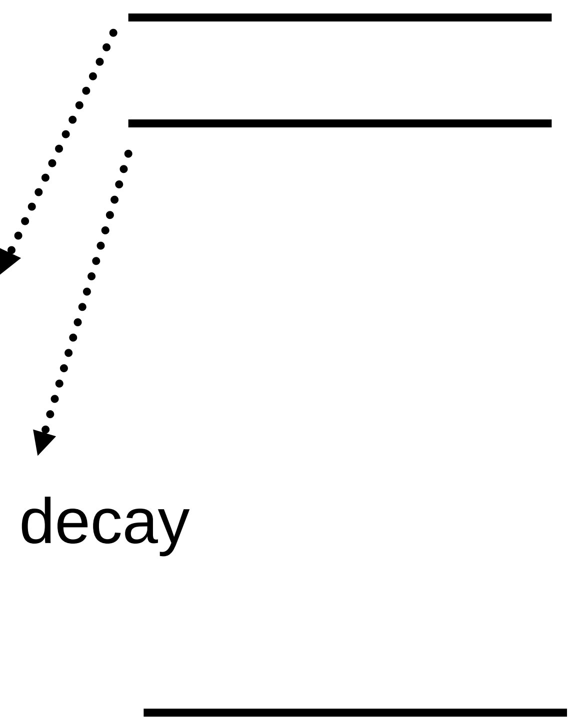
Mixture of the signals
(Solid line)

State selected signal
-> Possible?

State selective excitation

Excited states having decaying process

$$E = \varepsilon + \frac{\Gamma}{2} i$$



Decay process

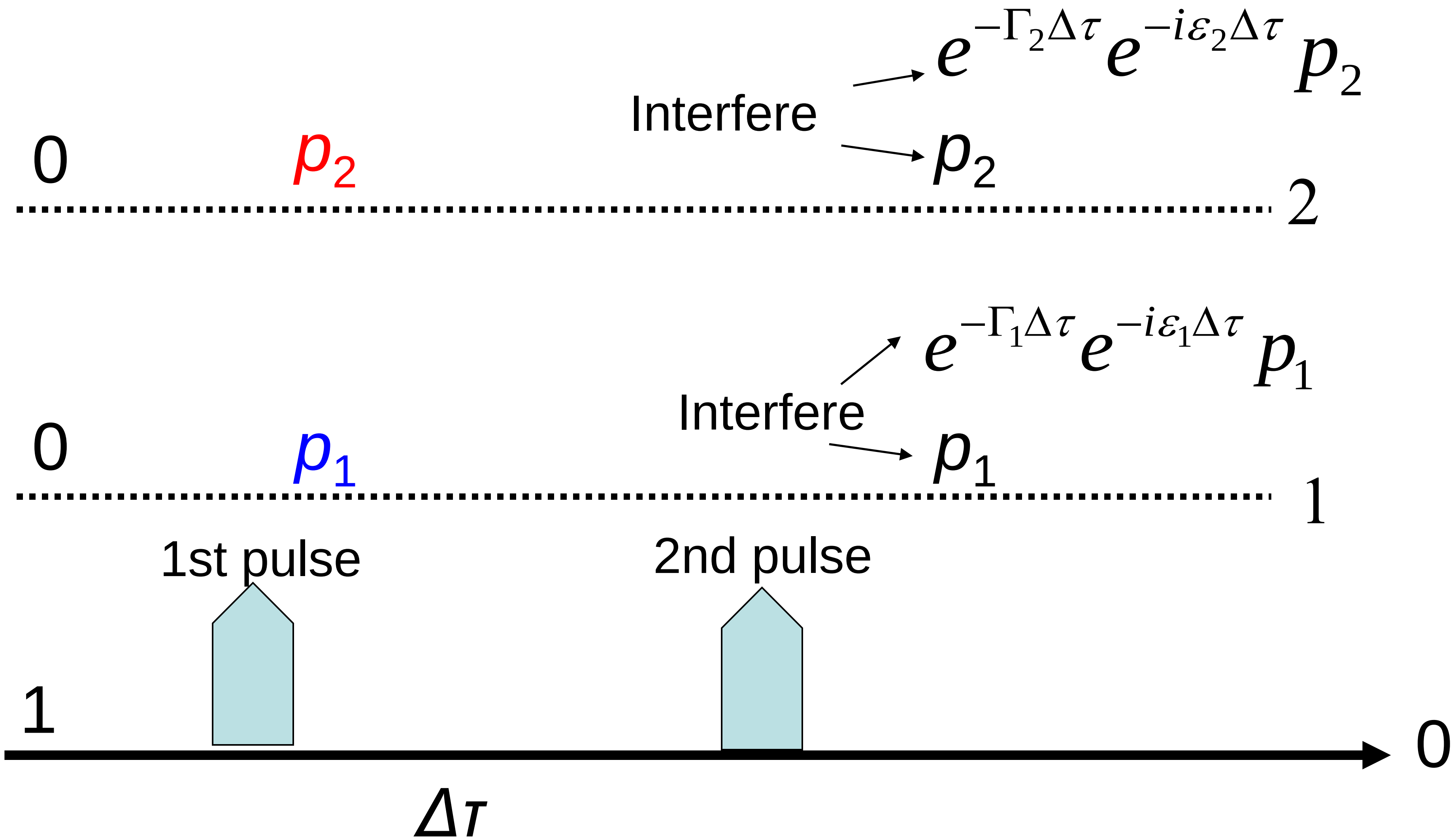
- Finite Lifetime
- Energy width (Natural width)

$$\exp(iEt) = \exp\left(i\varepsilon t - \frac{\Gamma}{2} t \right)$$

$$= \exp(-i\varepsilon t)\exp\left(-\frac{\Gamma}{2} t \right)$$

Selective excitation to decaying state

Breakdown of selectivity due to the decaying process



Incomplete interference due to the decaying process

How to achieve the selection

- Modify the intensity of the 2nd pulse

$$r = {I_2}/{I_1} = e^{-\Gamma\Delta\tau}$$

Reduce the intensity
（condition for the intensity ratio）

$$(E-\omega)\Delta\tau = \delta + (2n+1)\pi$$

Destructive intereference
（condition for the phase）

Selection is possible even for the decaying states

Intensity ratio $\rightarrow$ Lifetime （Width）
Phase difference & Delay $\rightarrow$ Energy（Position）

Feedback spectroscopy?

System information is obtained from the optimal external field

Result (selection)

External Field

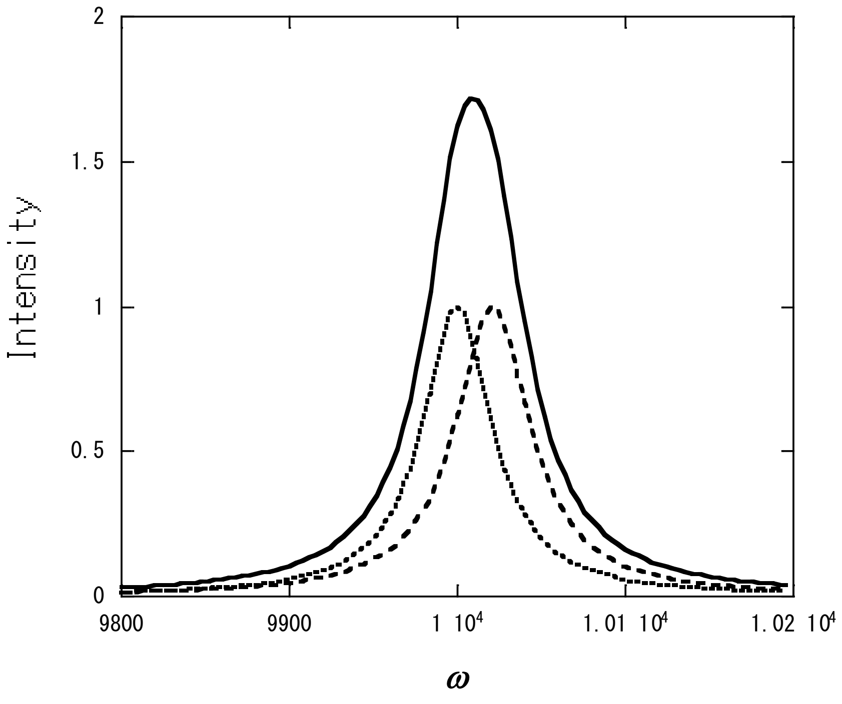
System (Position & with)

A new type of inverse problem

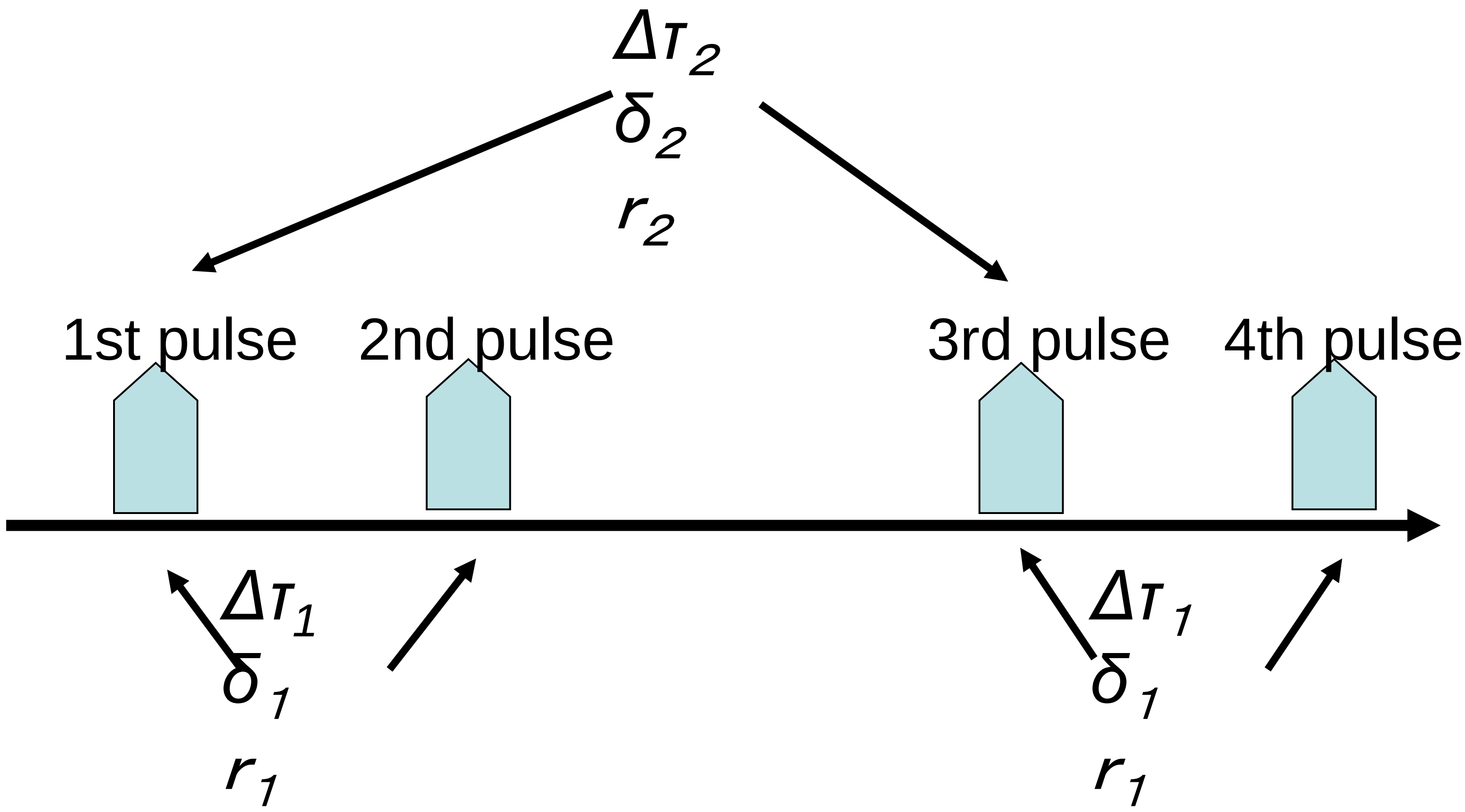
Uniqueness?

Problem

It is impossible to measure the selection ratio!



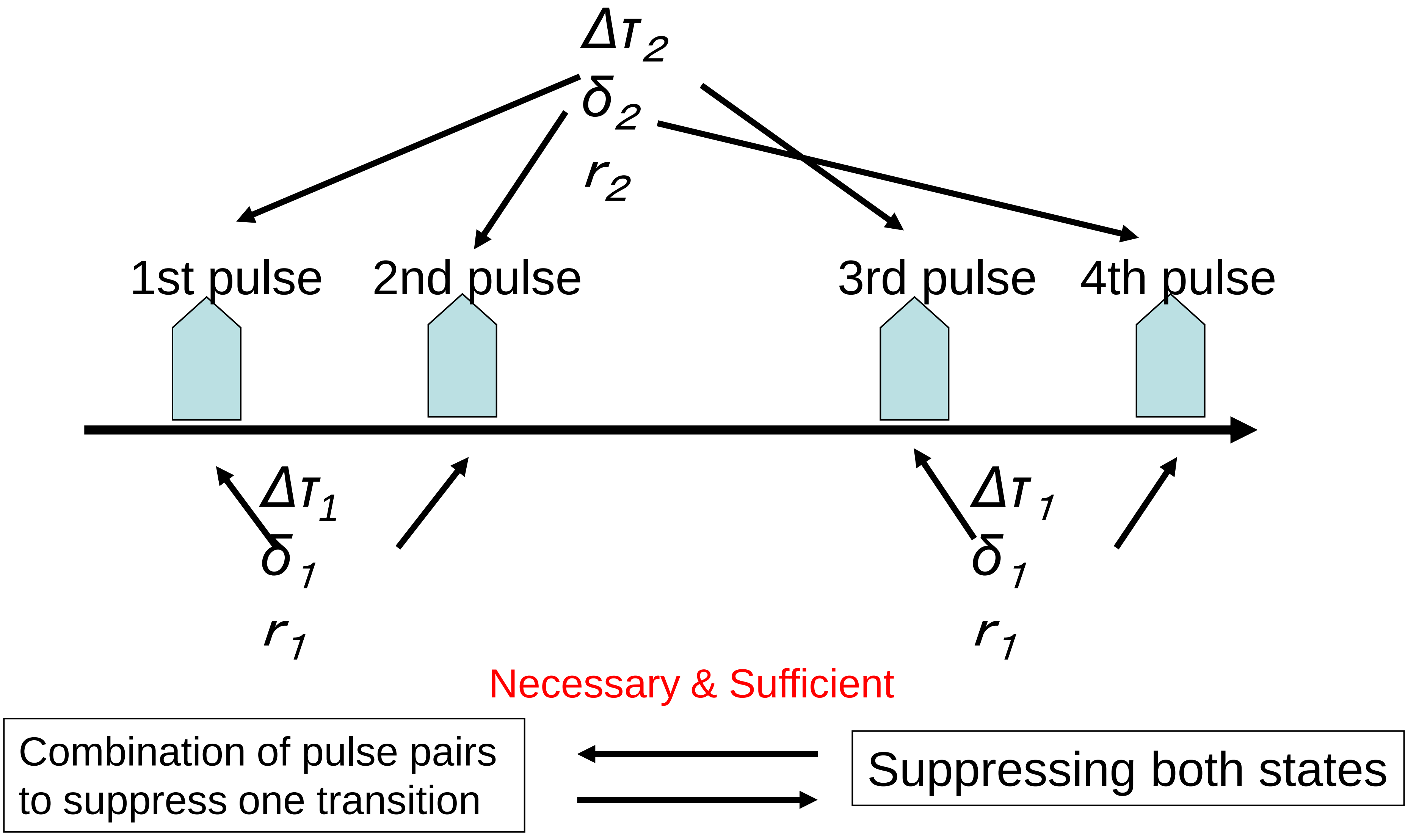
4 pulse irradiation
(Suppressing both two states)



Necessary & Sufficient

Combination of pulse pairs to suppress one transition	$\Longleftrightarrow$	Suppressing both states

4 pulse irradiation
(Suppressing both two states)



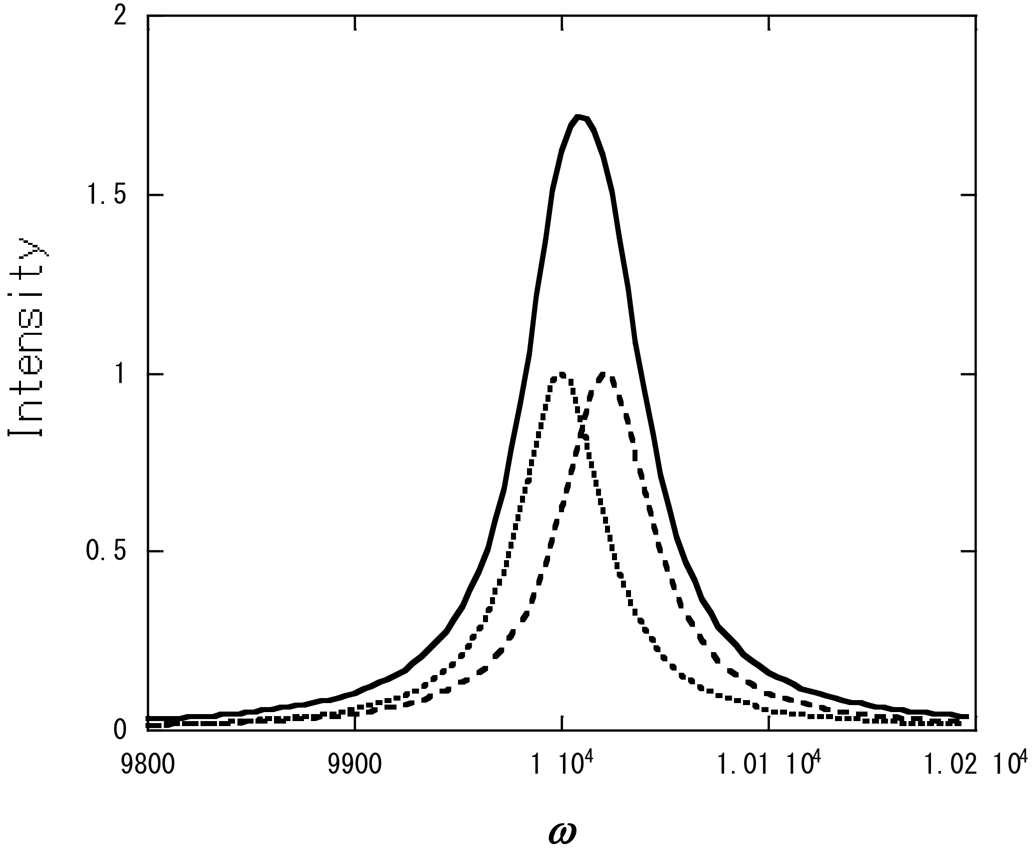
New Spectroscopy

- Irradiating a train of 4 pulses
- Searching for a condition to achieve zero total excitation probability

- 1st + 3rd pair -> selecting one transition
- 2nd + 4th pair -> selecting the other

- positions and widths of both states
- Enabling state selective pump probe

Model



$$E_1 = 10000 - 25i \ \ [\mathrm{cm}^{-1}]$$

$$E_2 = 10021 - 27i \ \ [\mathrm{cm}^{-1}]$$

$$\Delta\tau_1 = 300 \text{ fs}$$

$$\Delta\tau_2 = 330 \text{ fs}$$

Feedback Scheme

- Parameters $\longrightarrow$ $r_1, r_2, \delta_1, \delta_2$

Phase differences

Intensity ratio

$$a = (\delta_1 + \delta_2)/2$$

$$b = (\delta_1 - \delta_2)/2$$

$$c = (r_1 + r_2)/2$$

$$d = (r_1 - r_2)/2$$

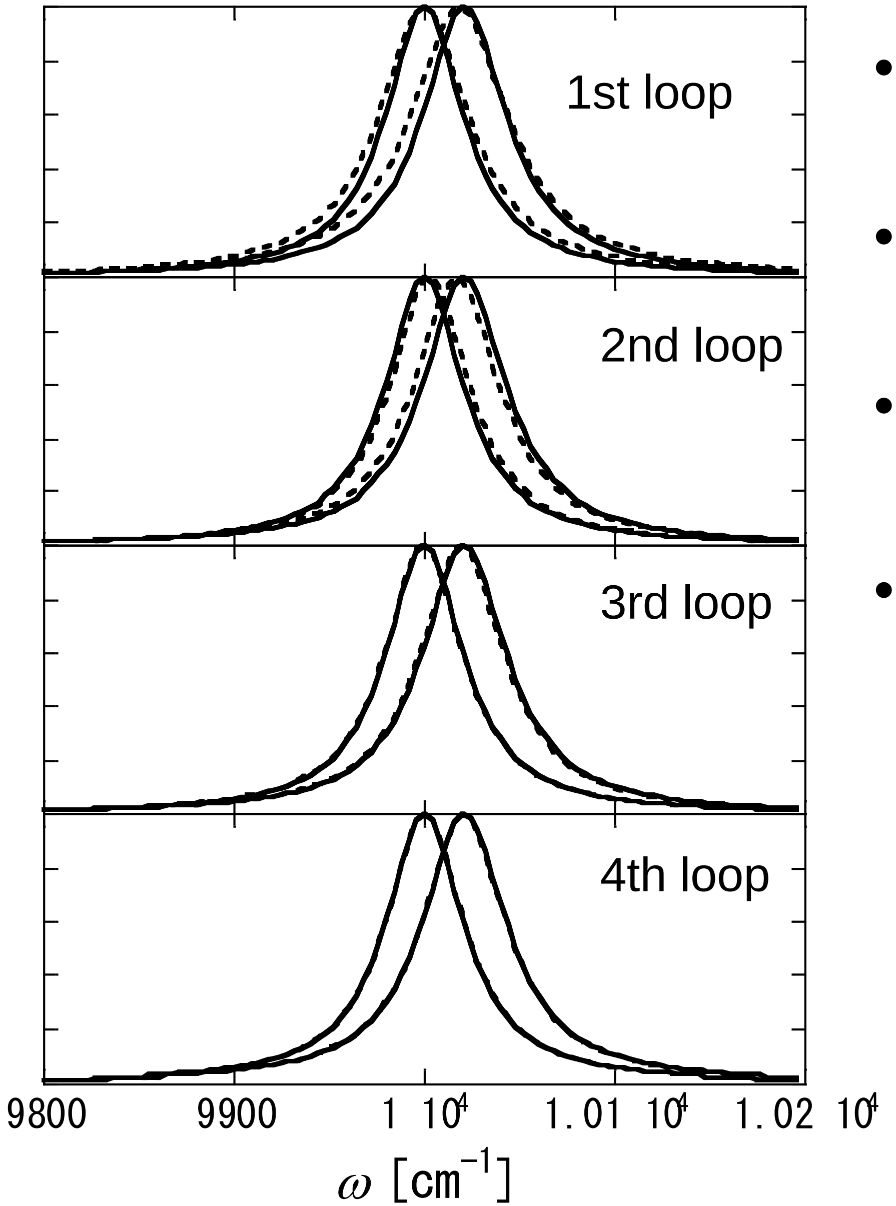
Successive optimization of single parameter from a, b, c, d, a, b,,,

(to obtain the minima of total excitation)

Spectroscopic data and the selection ratio obtained after *n*th optimization

# of loop	$\mathrm{Re}(E_1)$	$\mathrm{Im}(E_1)$	$\mathrm{Re}(E_2)$	$\mathrm{Im}(E_2)$	P_1/P_2	P_2/P_1
1	9999.5	28.8385	10018.1	31.1767	0.102	0.078
2	10002.7	25.3285	10016.7	27.4977	0.0565	0.0325
3	9999.5	25.0348	10019.6	26.9731	0.00238	0.00315
4	**10000.1**	**25.0348**	**10020.7**	**26.9731**	**0.000471**	**0.000301**
Exact	**10000**	**25**	**10021**	**27**	**0**	**0**

Results

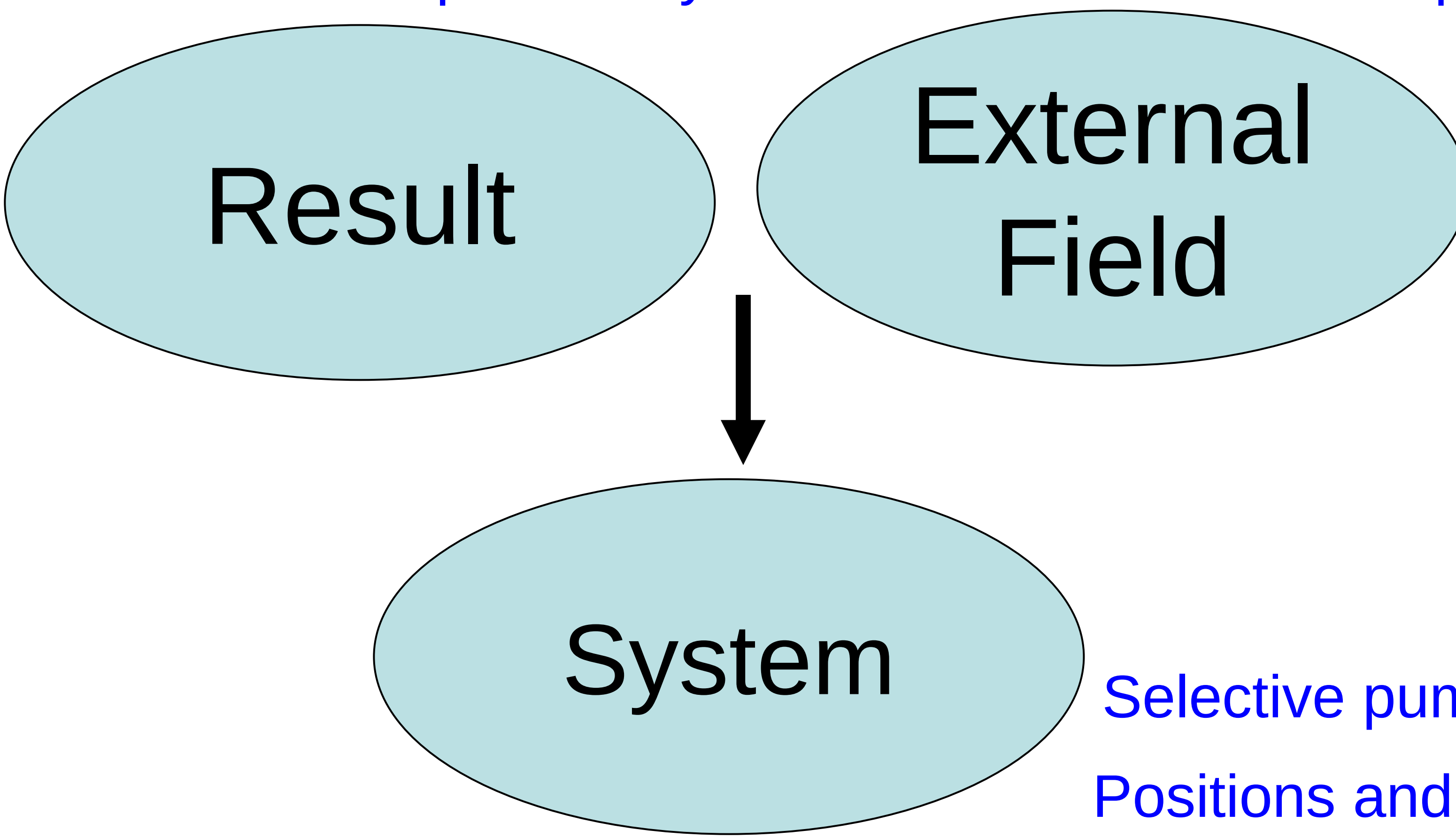


- State selective spectra
- Rapid convergence
- State selective pumping
- Powerful method for the study of ultrafast phenomenon

Feedback spectroscopy

Zero total excitation probability

Pulse train of 4 pulses



Selective pumping

Positions and widths

Quantum Control Spectroscopy

- Feedback scheme provides the optimal conditions for a pulse train of four pulses to achieve zero total excitation
- Parameters of the optimal pulse train give the positions and widths
- Selective pumping pulse pair can also be obtained (state selective time resolved spectra)
- Can be extended to N level system
- Applicable to autoionization (Auger) and predissociation

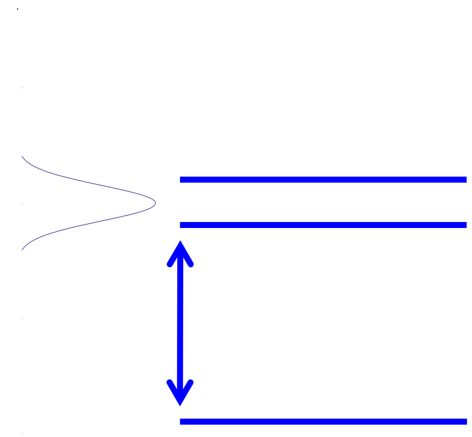
Summary

Quantum Control

- Ultrafast Selective Excitation

  A pair of weak pulses

  Quadratic Chirping

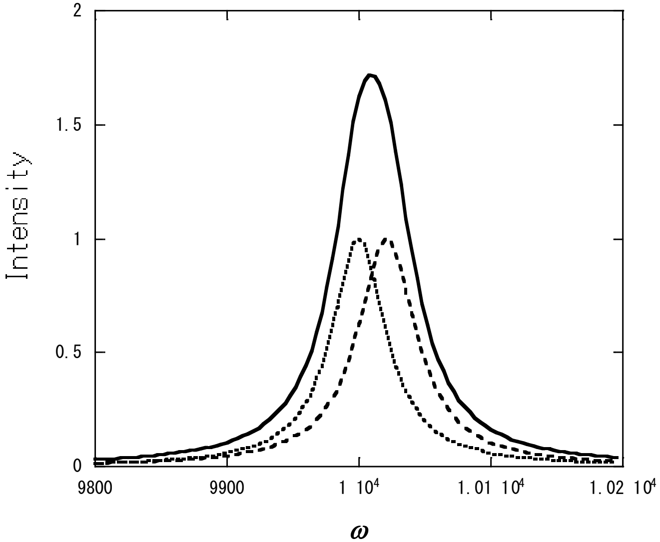


- Quantum Control Spectroscopy

  (overlapping by natural width)

- Molecular computer

  (Classical computer)



RESEARCH HIGHLIGHTS

Nature 465 (2010)

QUANTUM INFORMATION

Leak-proof chips

Phys. Rev. Lett. doi:10.1103/PhysRevLett.104.180501 (2010)

Semiconducting chips are fast approaching classical limits. Electrical circuits have become atomically thin, causing errors in their logic gates when current leaks out. However, quantum manipulations of atoms and small molecules offer a way around these limits.

Kenji Ohmori at the Institute for Molecular Science in Okazaki, Japan, and his colleagues describe a new logic component that could be used in quantum-information science. It is an ultra-fast Fourier transform, a standard mathematical tool used in electronic signal processing to convert signals from one function to another.

The team excited an iodine molecule such that its quantum vibrations executed Fourier transforms in just 145 femtoseconds — several orders of magnitude faster than is possible in today's computer chips. The technique shows another way in which a quantum computer could, in theory, be both faster and more accurate than a classical computer. **E.H.**

Quantum control and Classical computation

Selected for a Viewpoint in *Physics*

PRL **104,** 180501 (2010) PHYSICAL REVIEW LETTERS week ending 7 MAY 2010

Ultrafast Fourier Transform with a Femtosecond-Laser-Driven Molecule

Kouichi Hosaka,[1,2] Hiroyuki Shimada,[1,2] Hisashi Chiba,[1,2] Hiroyuki Katsuki,[1,2,3] Yoshiaki Teranishi,[2,4] Yukiyoshi Ohtsuki,[2,4] and Kenji Ohmori[1,2,3,*]

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(Received 7 January 2010; published 3 May 2010)

Wave functions of electrically neutral systems can be used as information carriers to replace real charges in the present Si-based circuit, whose further integration will result in a possible disaster where current leakage is unavoidable with insulators thinned to atomic levels. We have experimentally demonstrated a new logic gate based on the temporal evolution of a wave function. An optically tailored vibrational wave packet in the iodine molecule implements four- and eight-element discrete Fourier transform with arbitrary real and imaginary inputs. The evolution time is 145 fs, which is shorter than the typical clock period of the current fastest Si-based computers by 3 orders of magnitudes.

DOI: 10.1103/PhysRevLett.104.180501 PACS numbers: 03.67.Lx, 33.80.−b, 42.50.Dv, 82.53.Kp

Phys. Rev. Lett. 104 180501 (2010)

Ultrafast Fourier Transformation with Molecule & Pulsed Laser

Phys. Rev. Lett. 104 180501 (2010)