Quantum Control of Atomic and Molecular Processes by Shaped Laser Field

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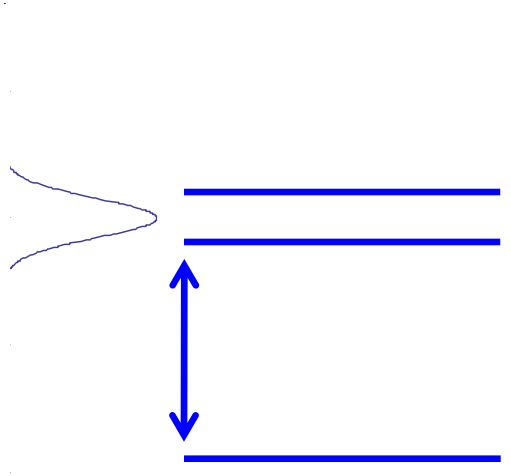
Outline

- Introduction to Quantum Mechanics

  Schrodinger Equation
  Stationary Problem
  Time-Dependent Problem



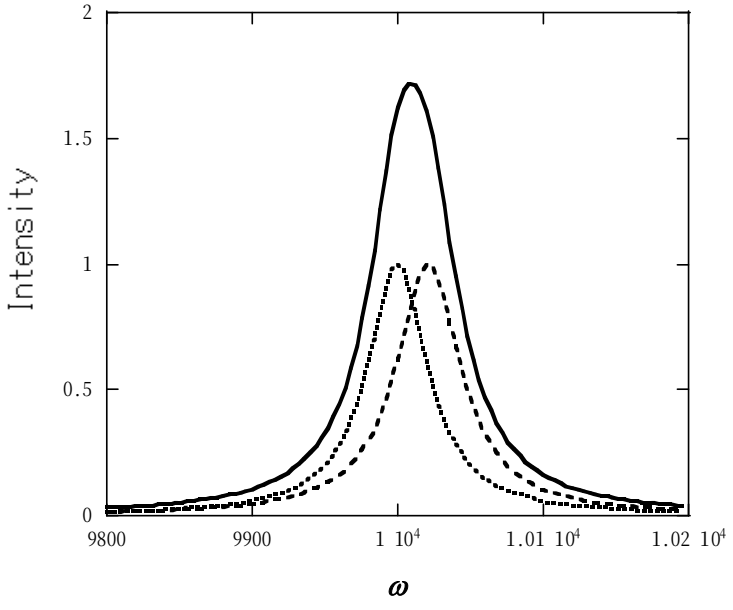
Quantum Control

- Ultrafast Selective Excitation

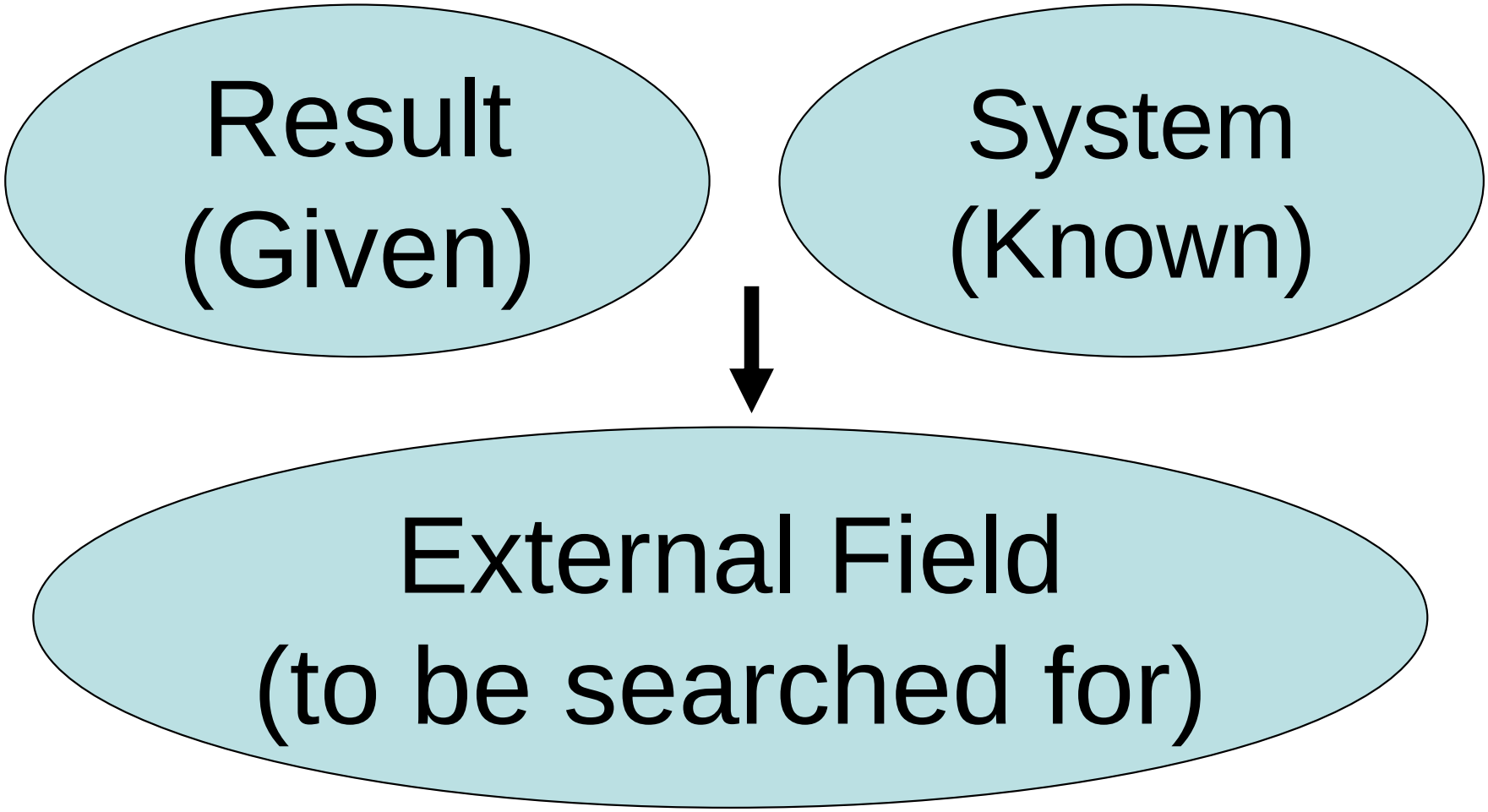
  A pair of weak pulses
  Quadratic Chirping

- Quantum Control Spectroscopy
  (overlapping by natural width)

- Molecular computer
  (Classical computer)



Quantum Control



Why Laser?

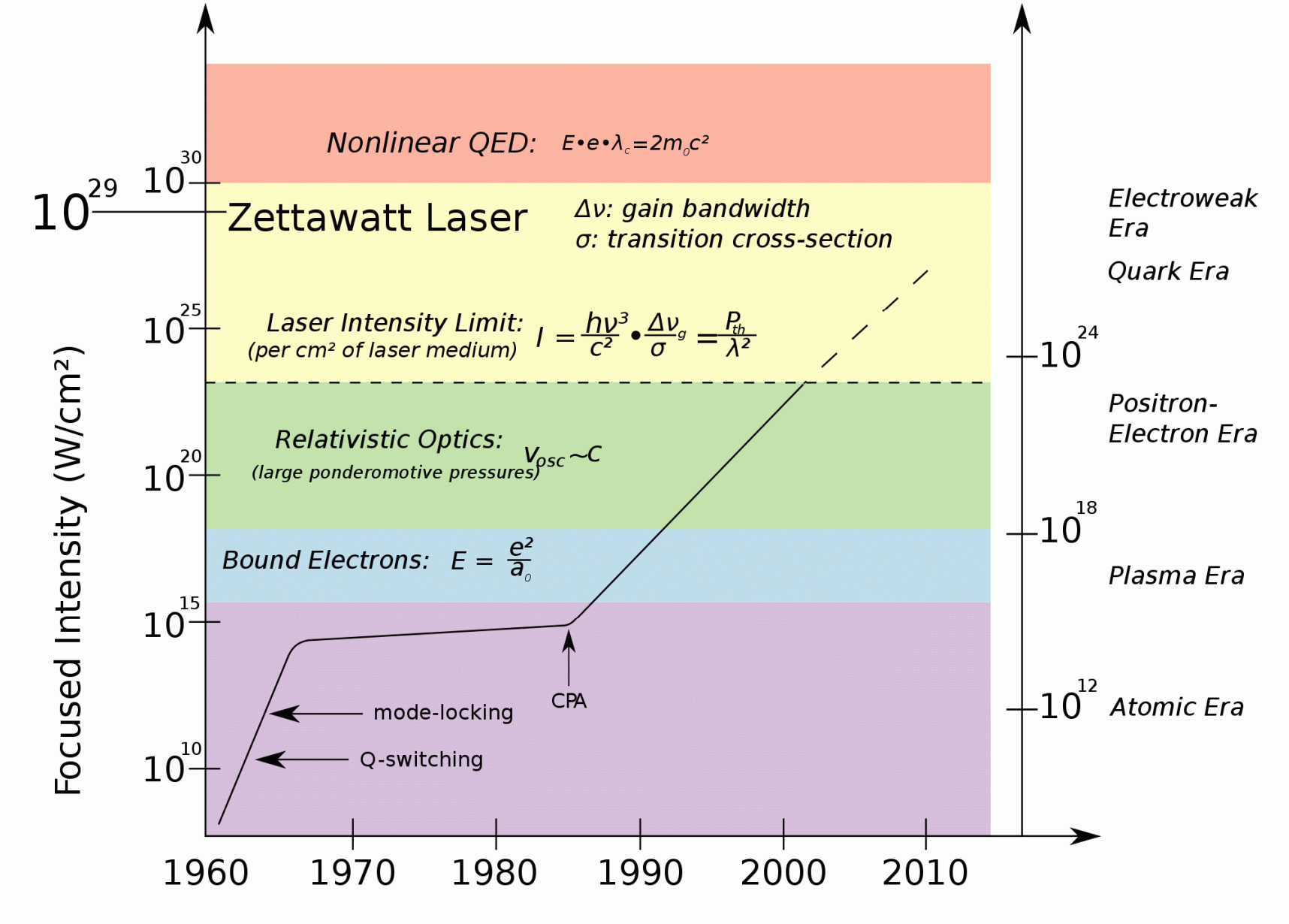
Coherence

High Intensity

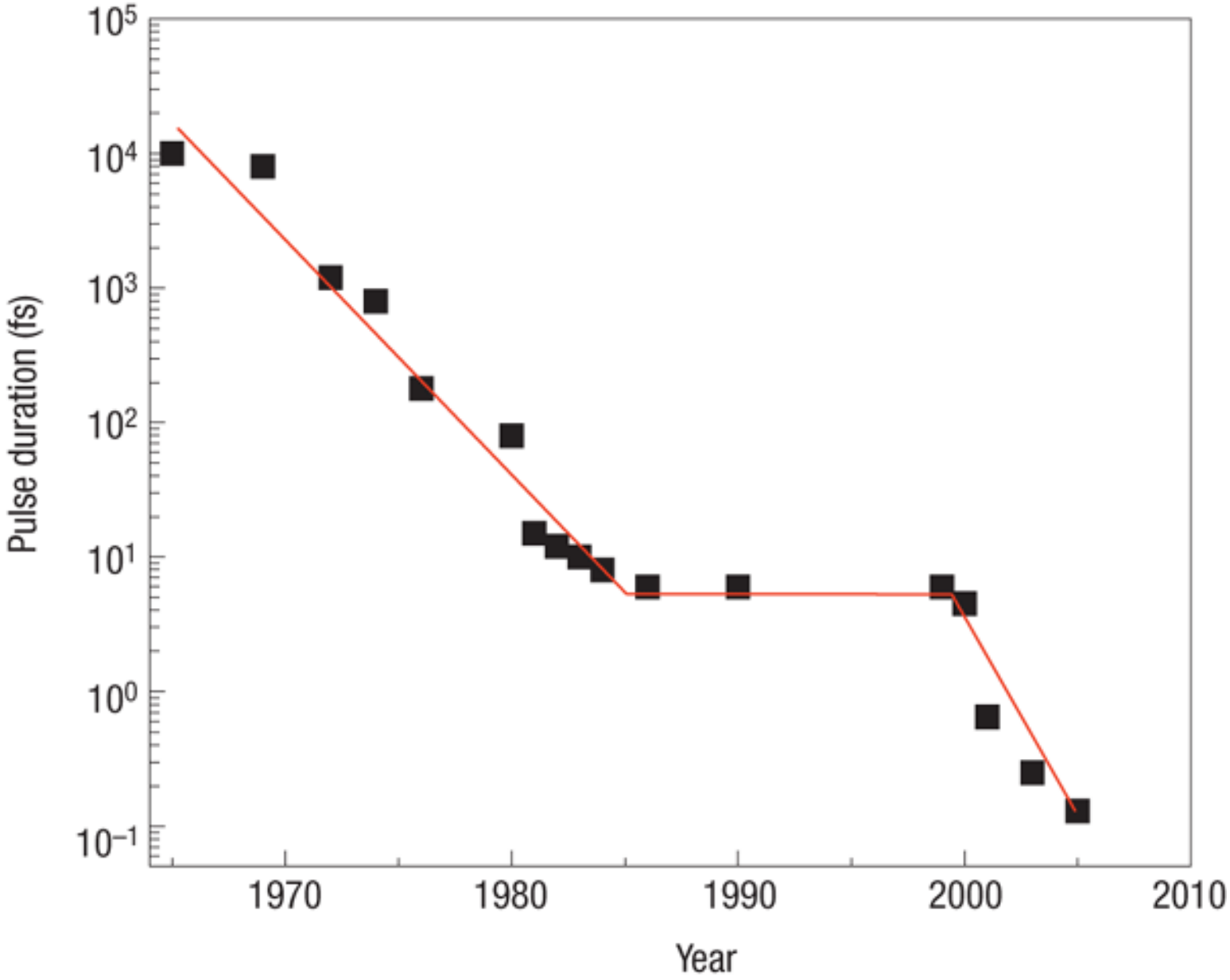
Short Pulse (Broad Band)

Pulse Shaping

History of Laser Intensity

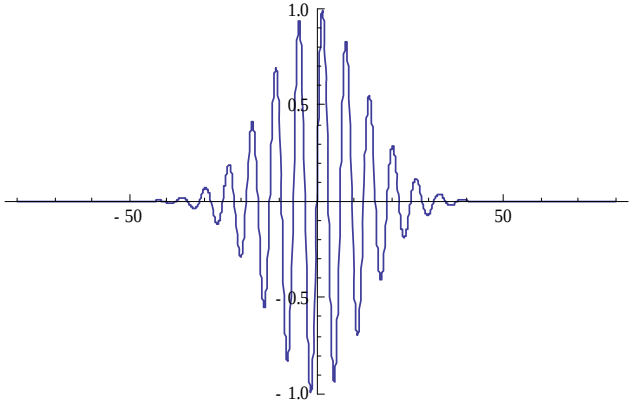


History of Laser Pulse Duration

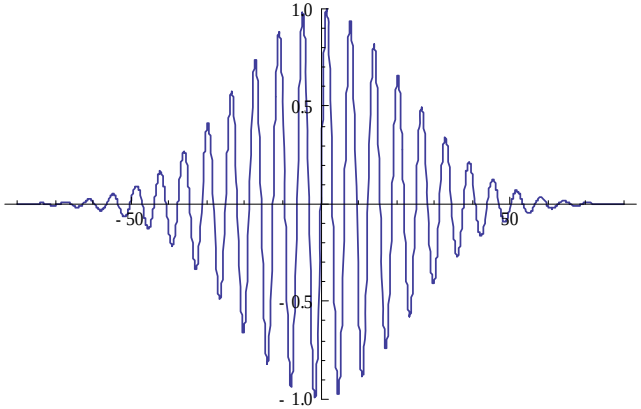


Laser Pulse

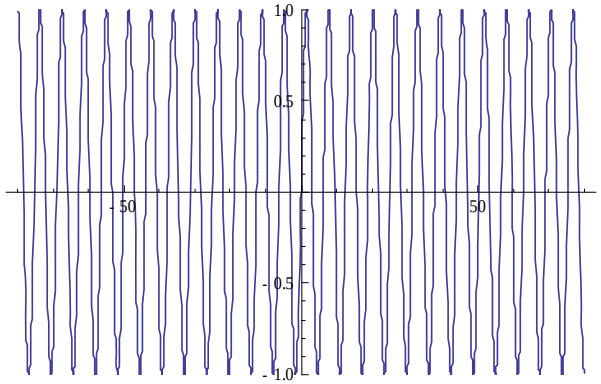
Short-Pulsed Laser



Long-Pulsed Laser

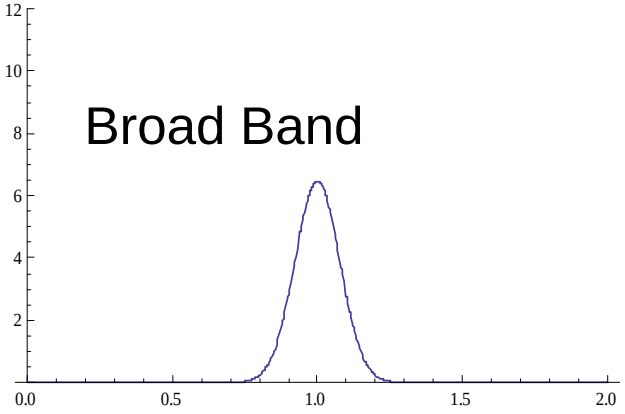


CW Laser

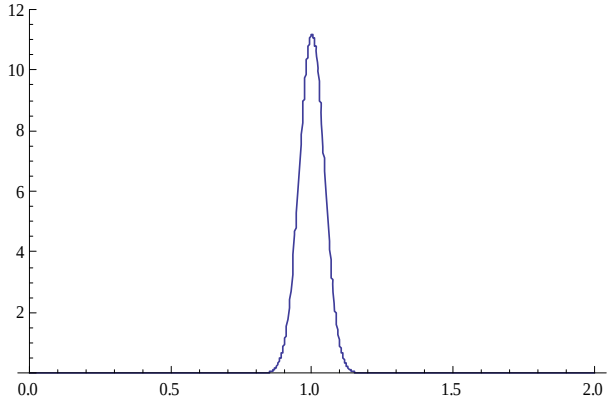


Time Domain

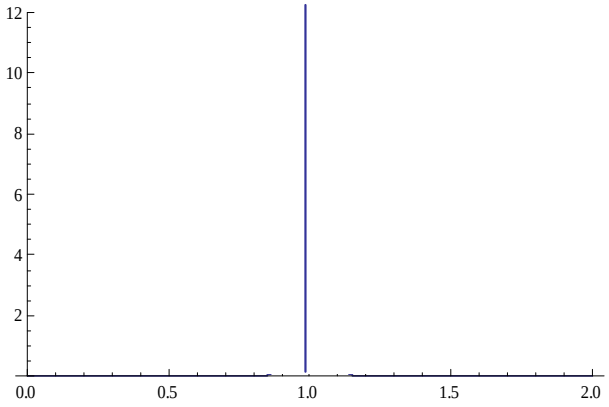
Broad Band



Narrow Band

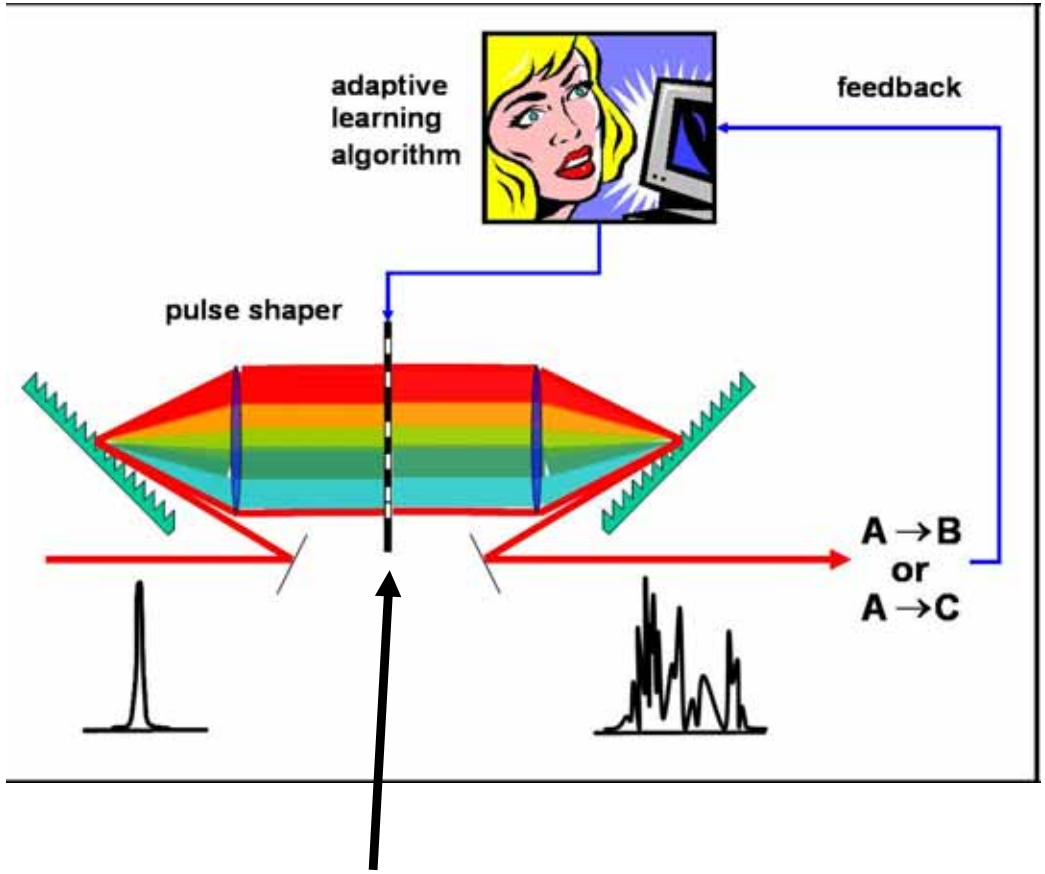


Monochromatic



Frequency Domain

Pulse Shaper



LCD
(Transmittance & Refractive indexes are controlled.)

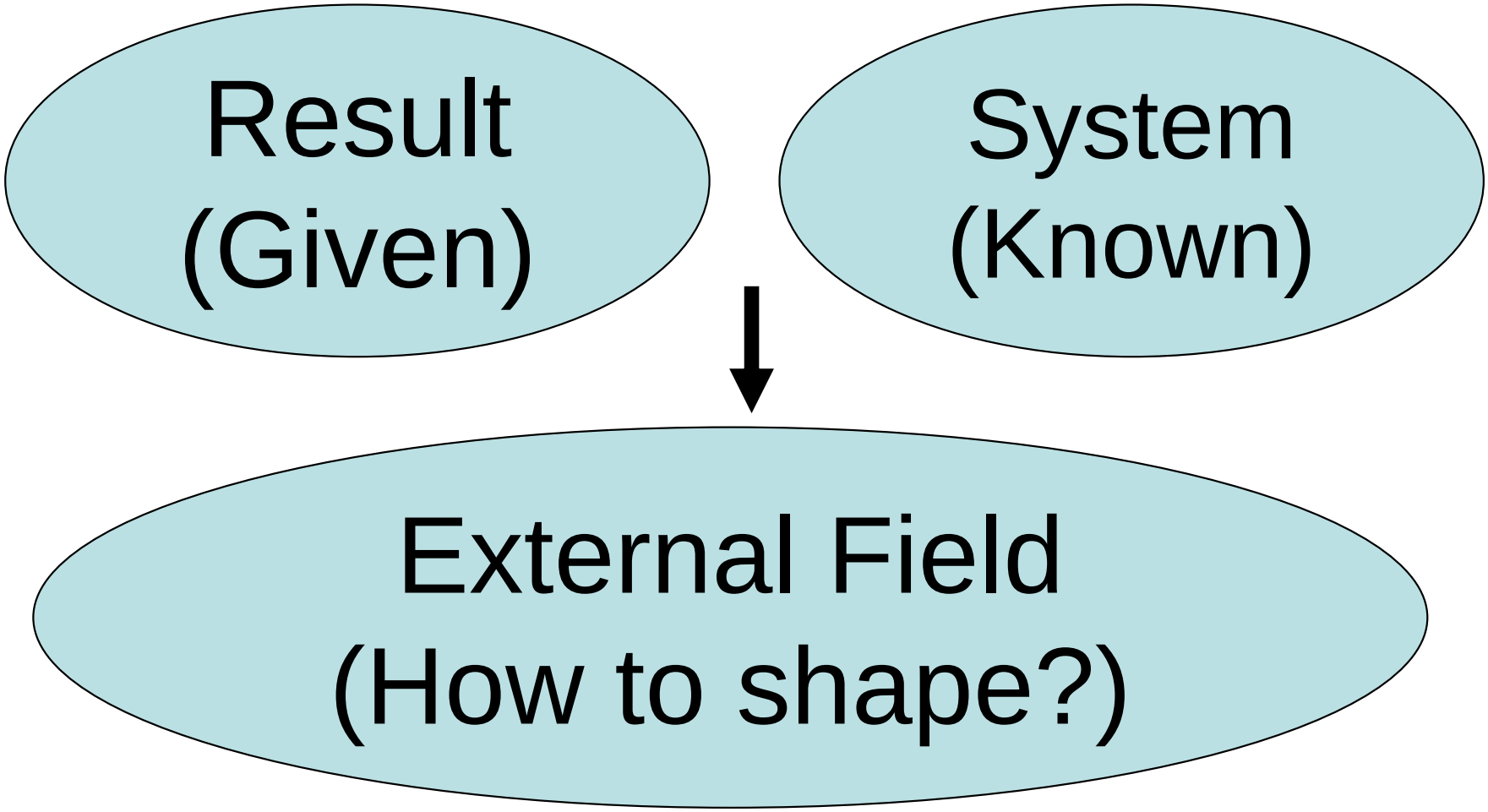
Fourier Expansion

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

Control of the Fourier coefficients

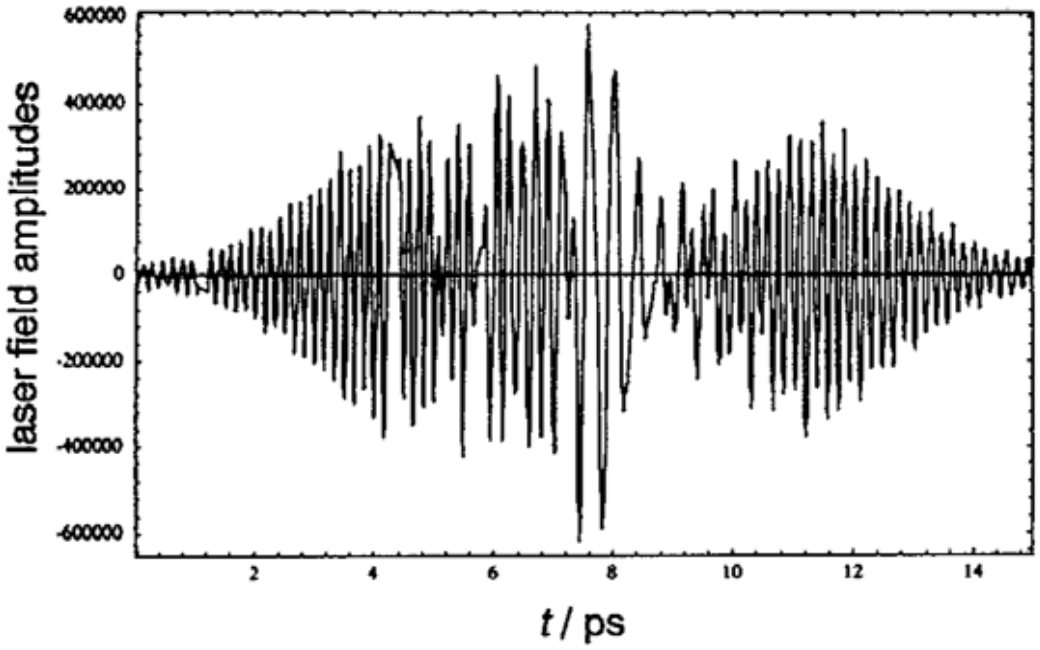
What is the optimal shape?

Quantum Control



Numerical optimization of the laser field for isomarization trimethylenimine

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



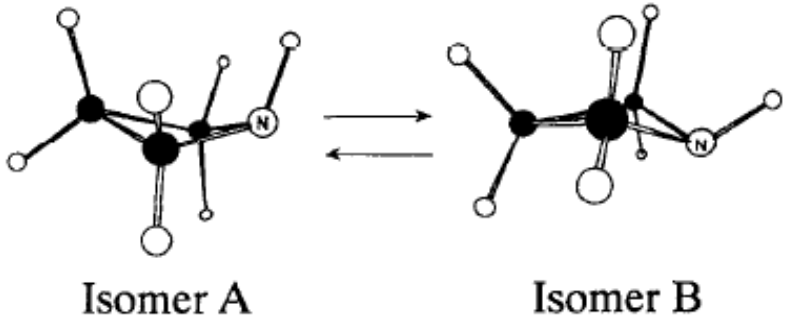


FIG. 1. Two different conformations of trimethylenimine.

Complicated Wave Form

Why this shape?
Any simpler?

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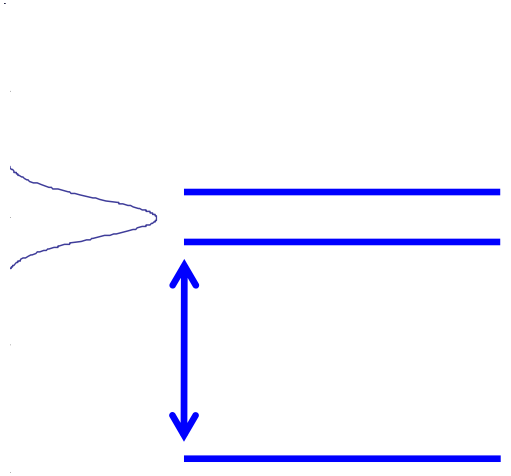
  Schrodinger Equation
  Stationary Problem
  Time-Dependent Problem



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  (overlapping by natural width)

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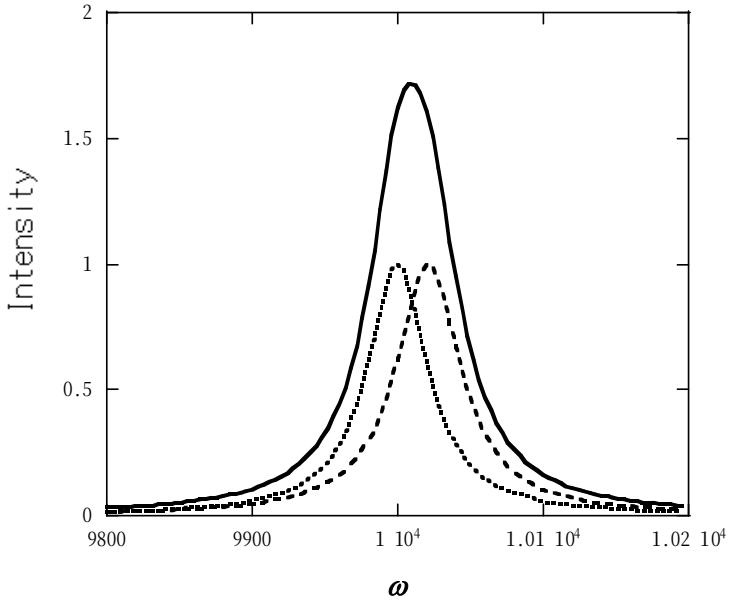

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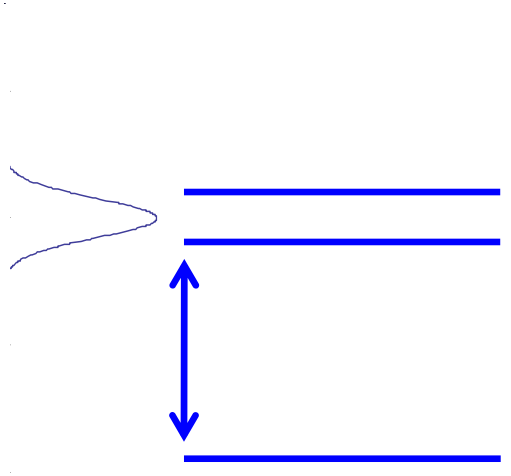

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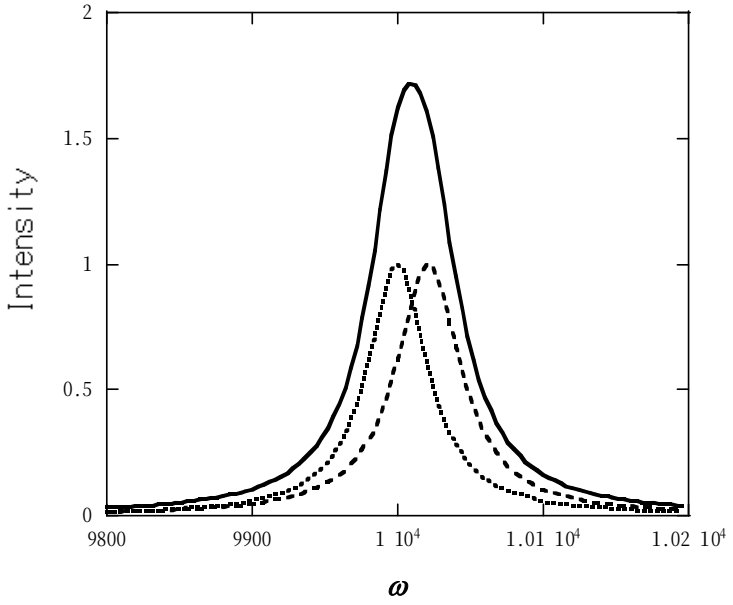


- Quantum Control Spectroscopy
  (overlapping by natural width)

- Molecular computer
  (Classical computer)

Quick Review on Quantum Mechanics

- Stationary problem

  Semi-classical picture, Eigenstates,
  Time propagation, Uncertainty Principle,,,

- Dynamical Problem

  Perturbation, Dressed State, Rabi oscillation,
  Adiabaticity, Nonadiabatic transition,,,,

Schrodinger Equation

Time Dependent Schrodinger Equation

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

If the Hamiltonian is time-independent

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

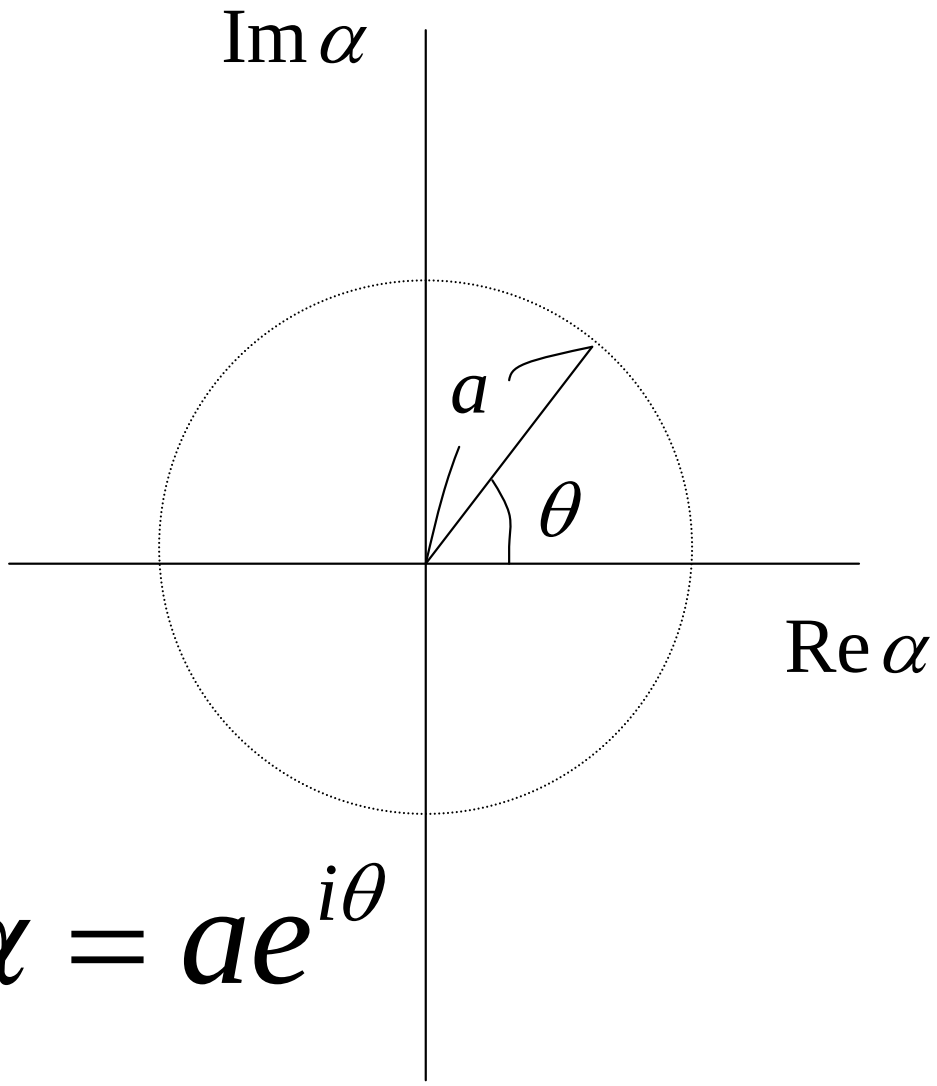
Time dependent part

Time independent part
(Eigen Function)

Stationary Schrodinger Equation

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

Exponential Function

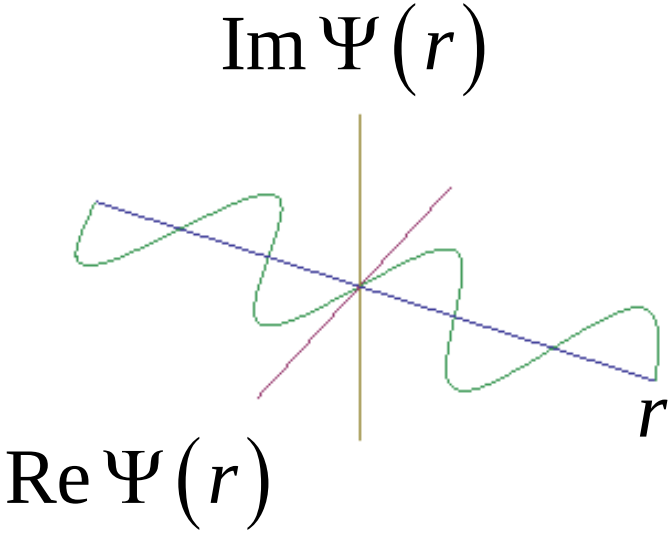


$$\alpha = ae^{i\theta}$$

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

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

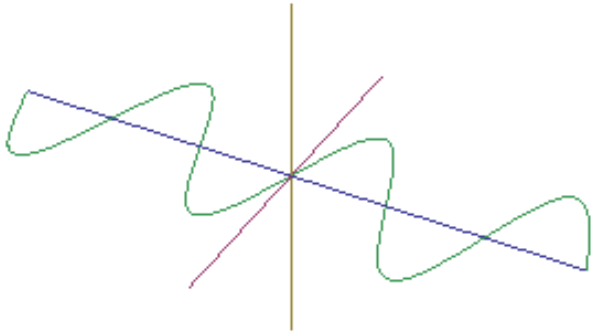
Time evolution of an eigen function

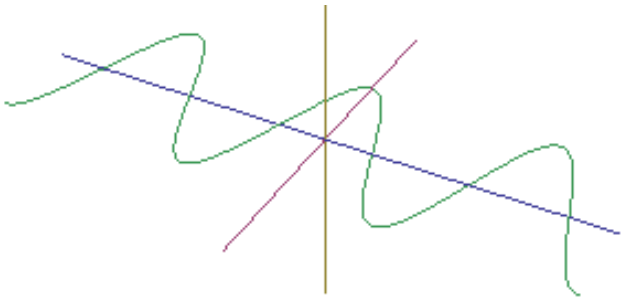


The shape does not change

A Superposition of Eigen States

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



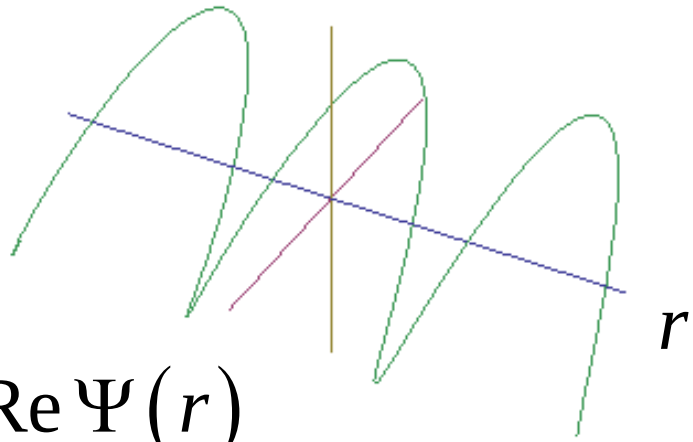


$$\Psi_2(r) = e^{-iE_2 t/\hbar} \cos(r)$$

$$\Psi_1(r) + \Psi_2(r)$$

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

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



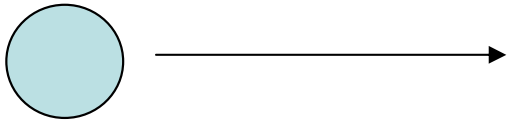
$\mathrm{Re}\,\Psi(r)$

Time dependent shape

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

Semiclassical Picture

Classical Particle



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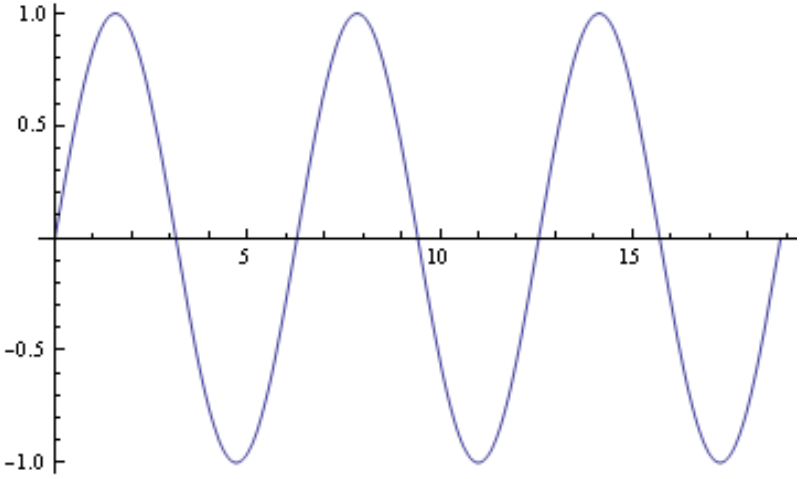
Property of wave



Amplitude

$$\propto \frac{1}{\sqrt{p(r)}} \propto \frac{1}{\sqrt{v(r)}}$$

Classical Probability

Wave Length

$$\propto \frac{2\pi\hbar}{p(r)}$$

De Broglie

Wave Function of a Free Particle

Time Independent Schrodinger Equation

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

$$V(r)=V_0 \text{ (Constant)}$$

Plane wave

$$\Phi(r)=e^{\pm ipr/\hbar}$$

$$p=\sqrt{2m(E-V_0)}$$

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

Plane Wave and WKB

Constant Potential

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

r-dependent Potential

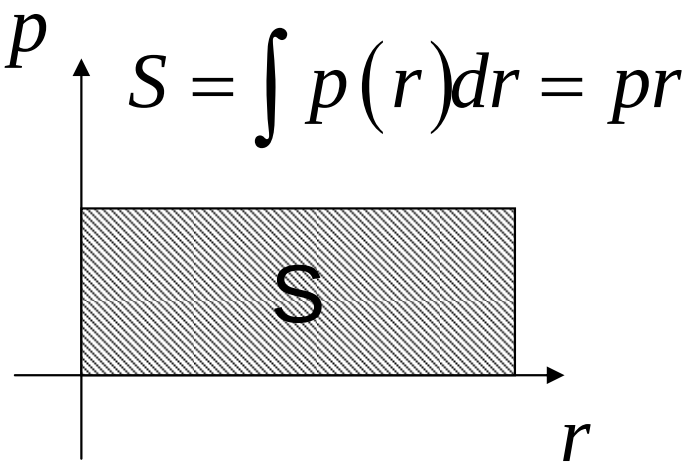
$$\left[-\frac{\hbar^2}{2m}\frac{d^2}{dr^2}+V(r)\right]\Phi(r)=E\Phi(r)$$

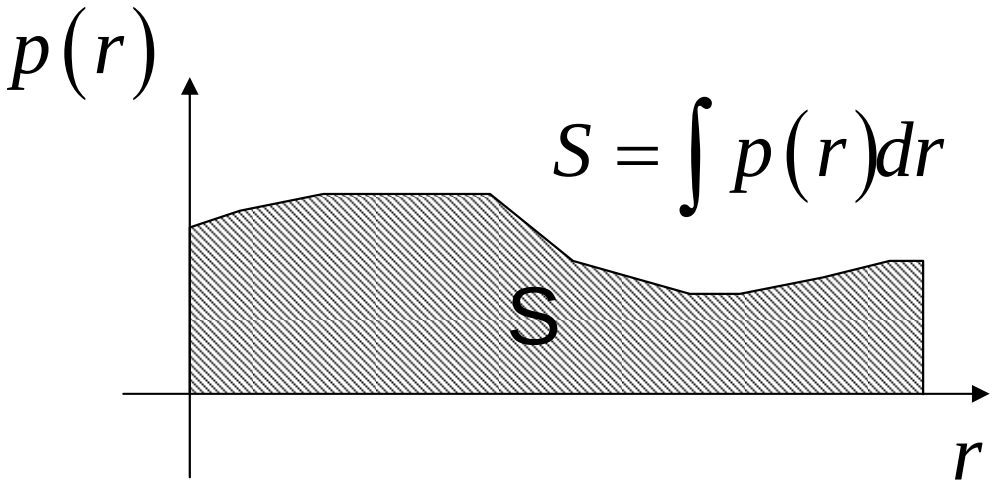
Plane wave

$$\Phi(r)=A\exp\left[\pm\frac{i}{\hbar}pr\right]$$

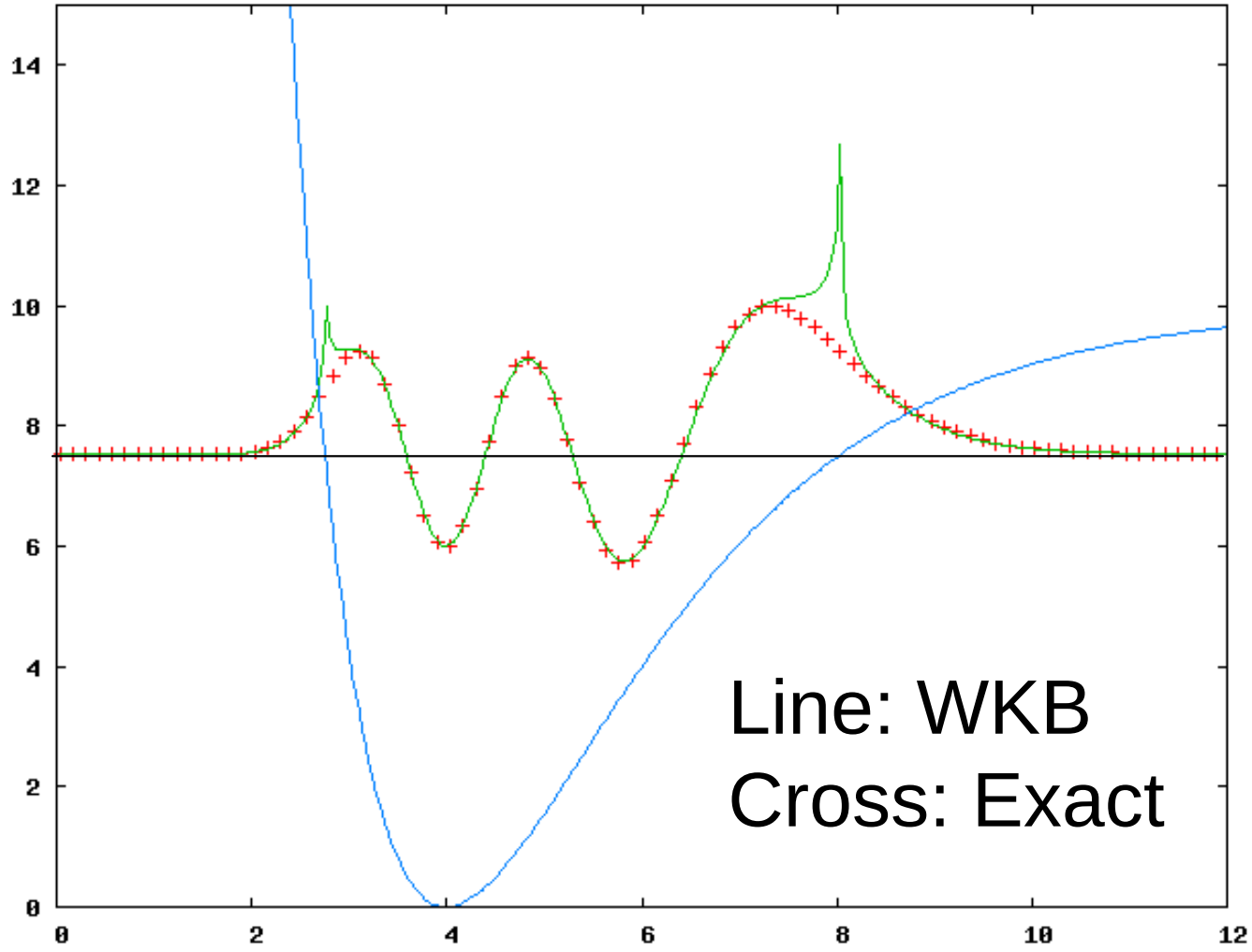
WKB Approximation

$$\Phi(r)=\frac{A}{\sqrt{p(r)}}\exp\left[\pm\frac{i}{\hbar}\int p(r)dr\right]$$

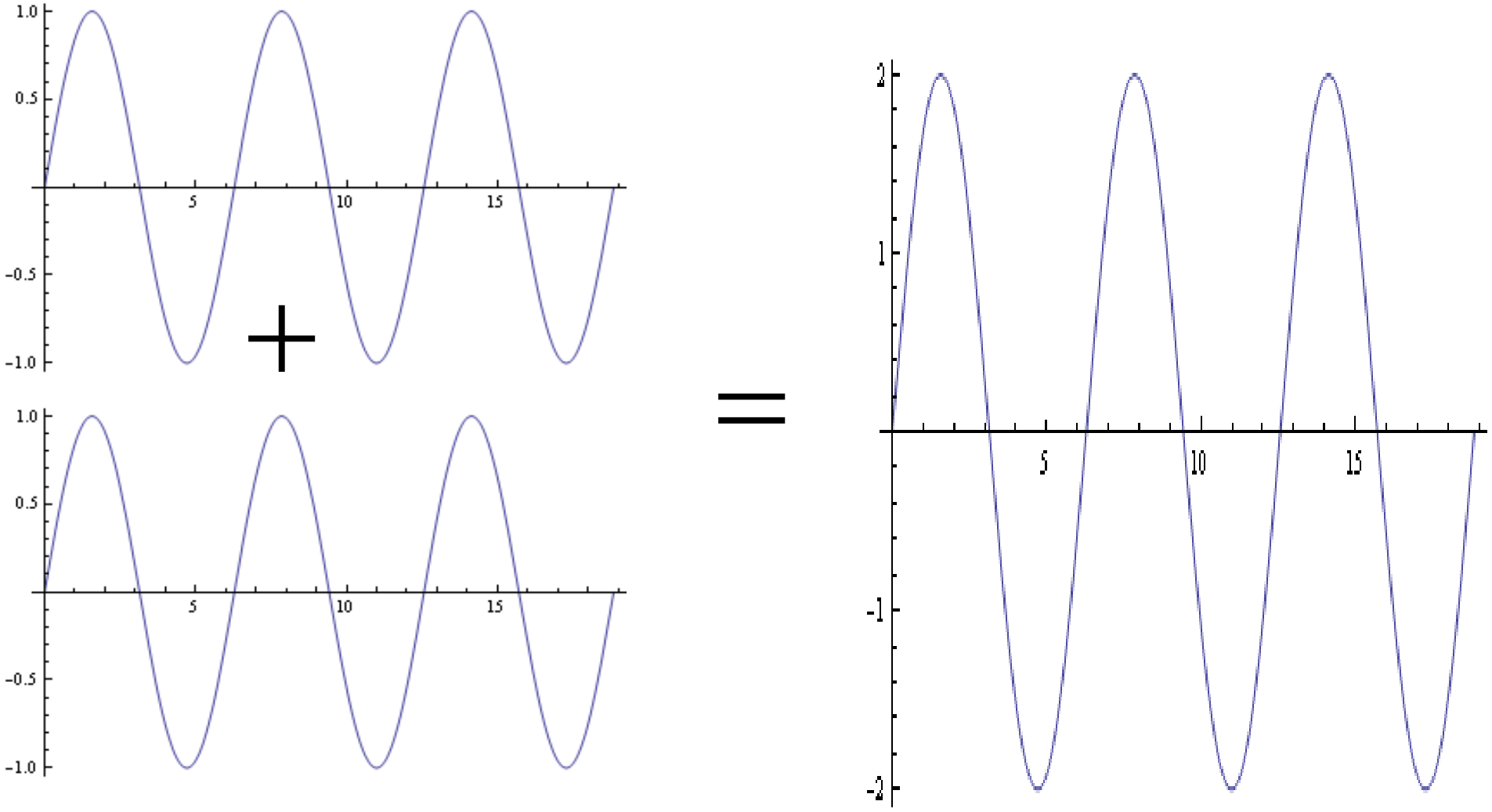




WKB and Exact Wave functions

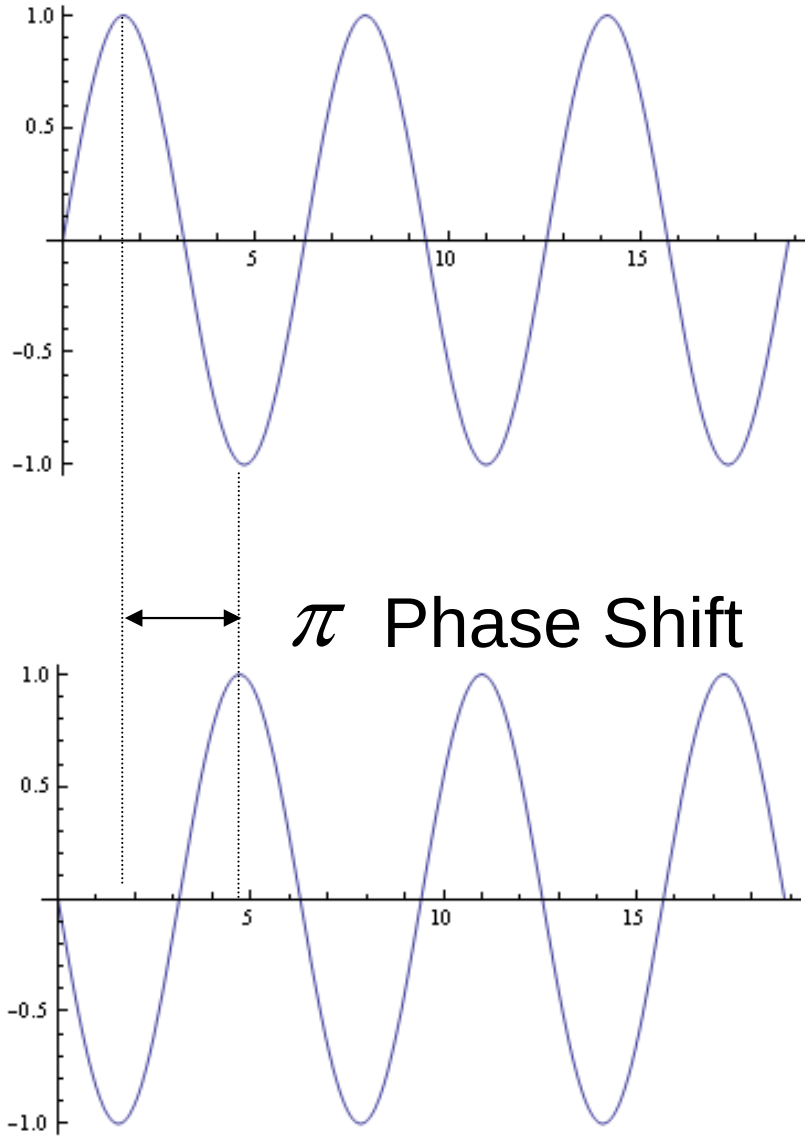


Constructive Interference



Zero Shift

Destructive Interference



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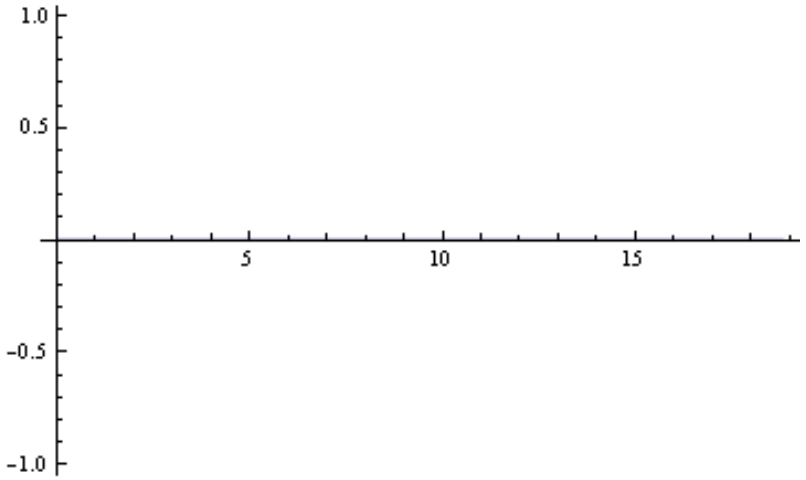
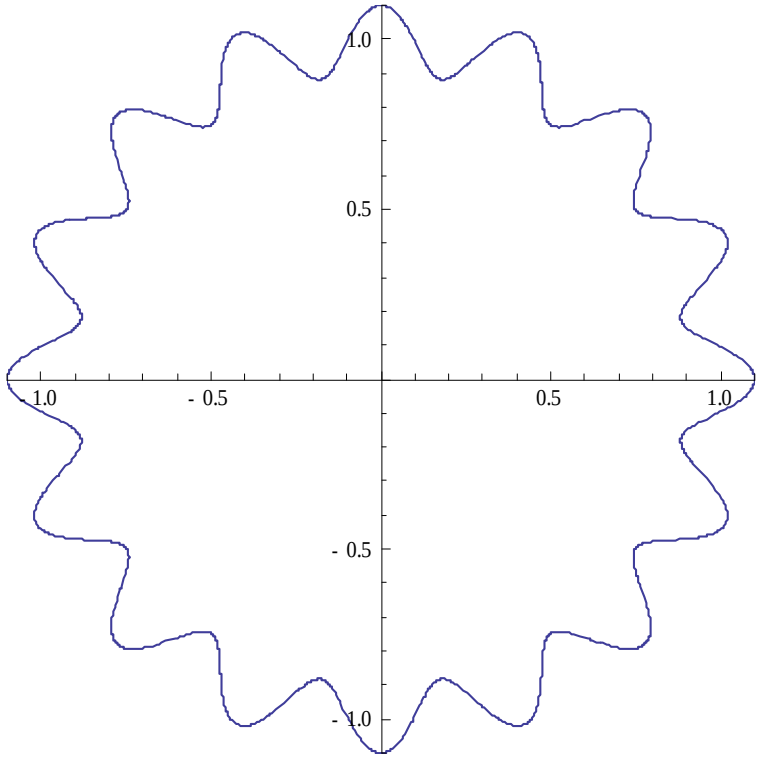
Vanish!

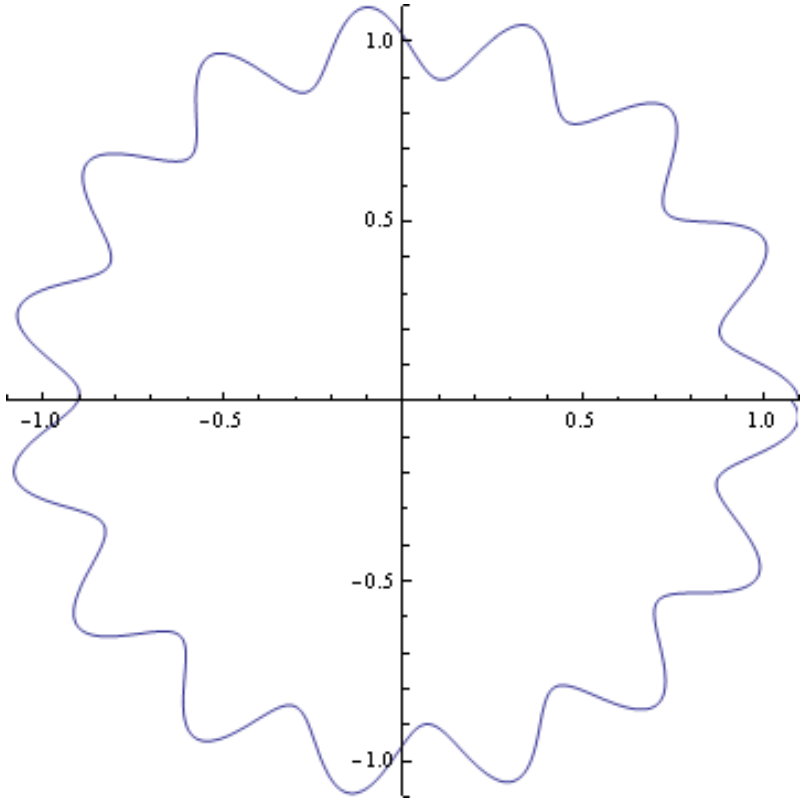


Bohr's Quantization Condition

Eigen State $\frac{1}{\hbar} pr = n$



Not satisfying the condition
(The First Cycle)



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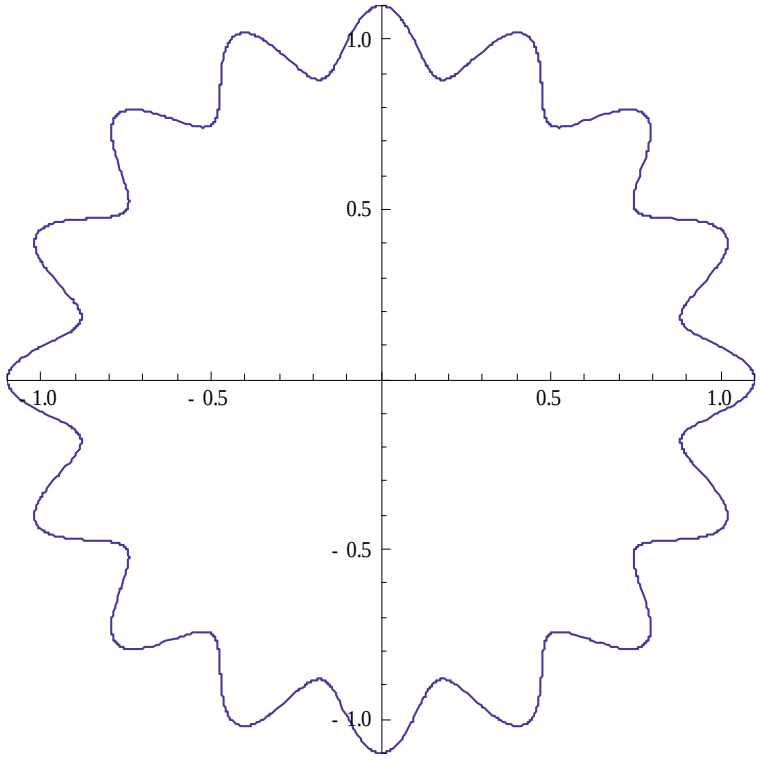
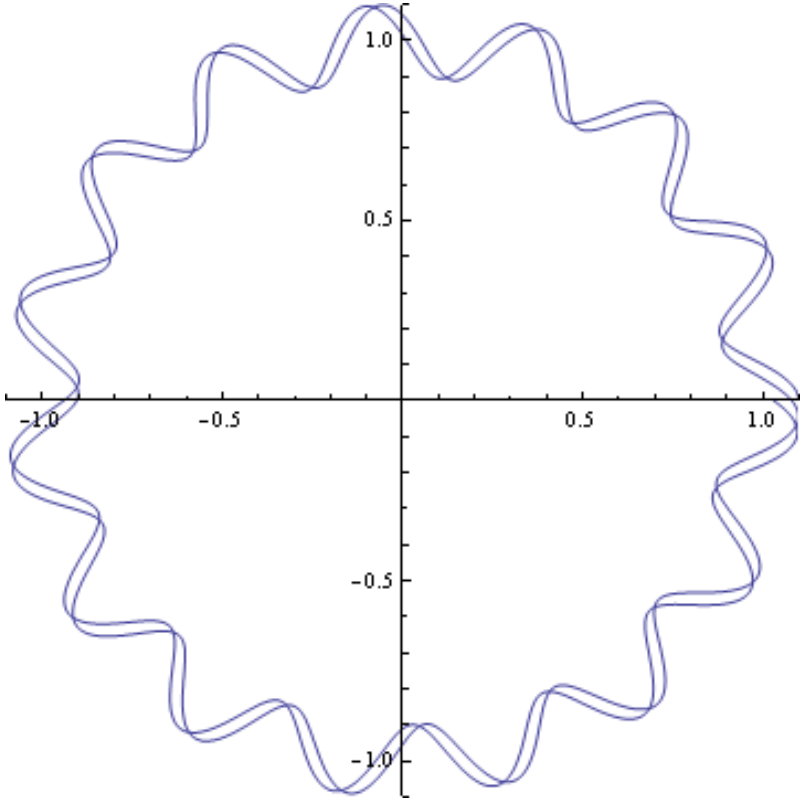


Not satisfying the condition
(After two cycles)



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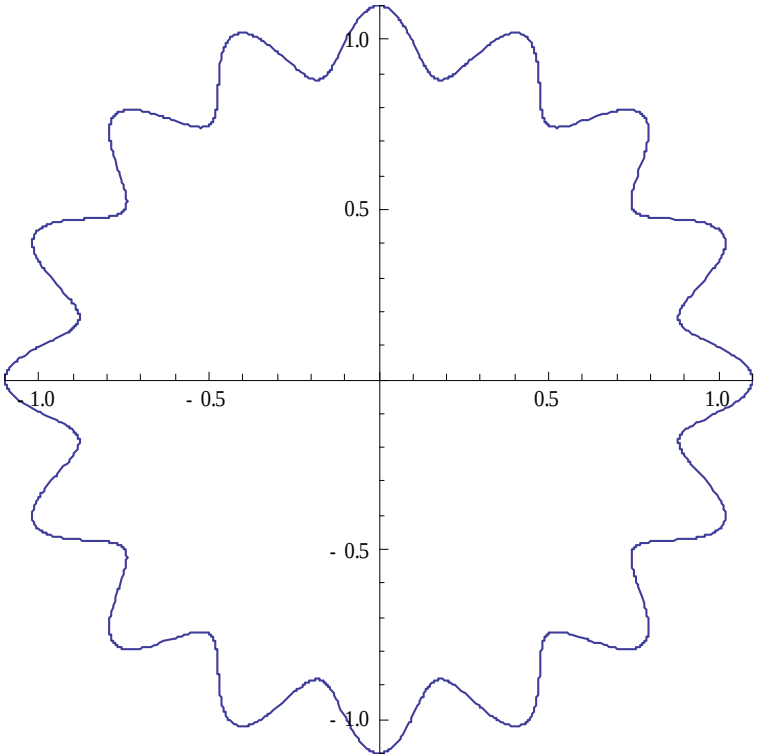
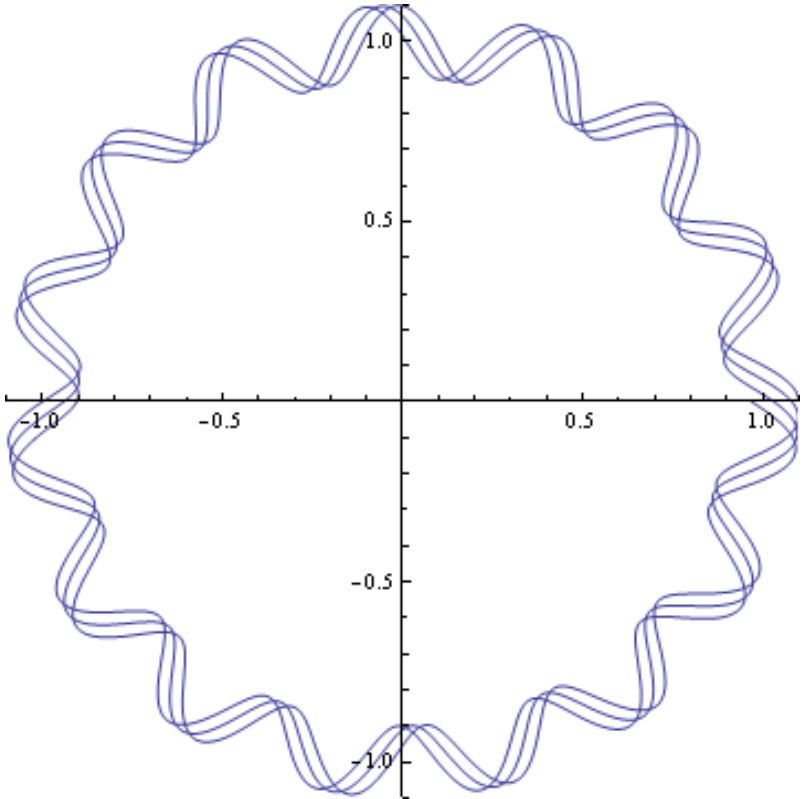


Not satisfying the condition
(After three cycles)



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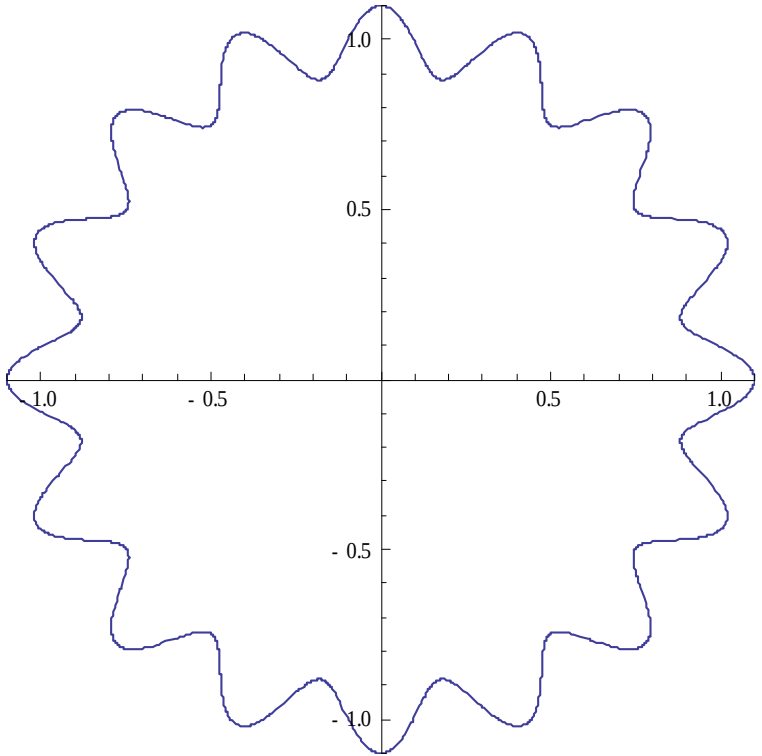
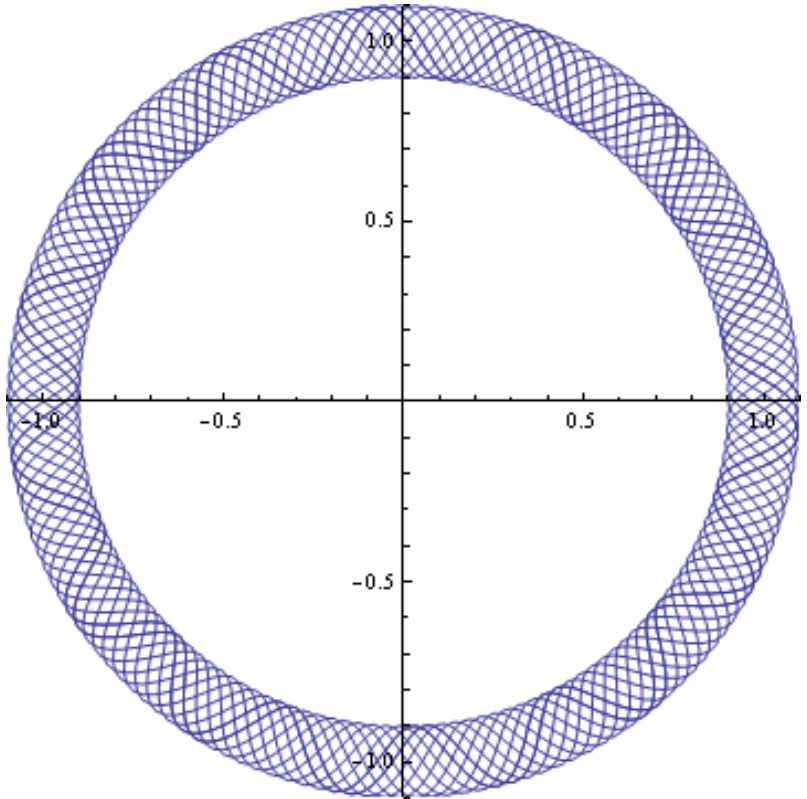
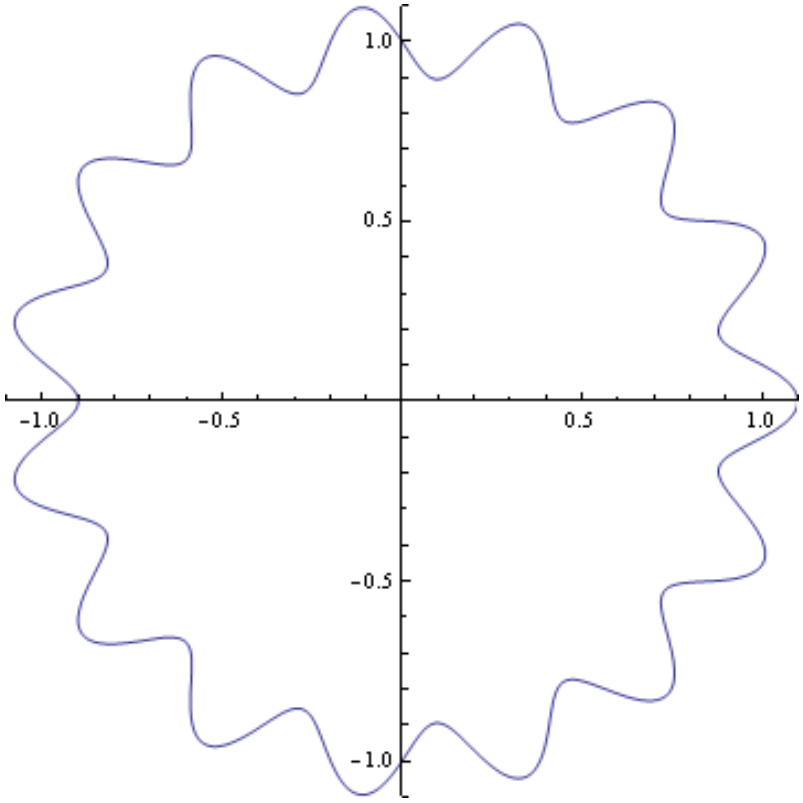


Not satisfying the condition
(After Many cycles)



Time Dependence

Eigen State



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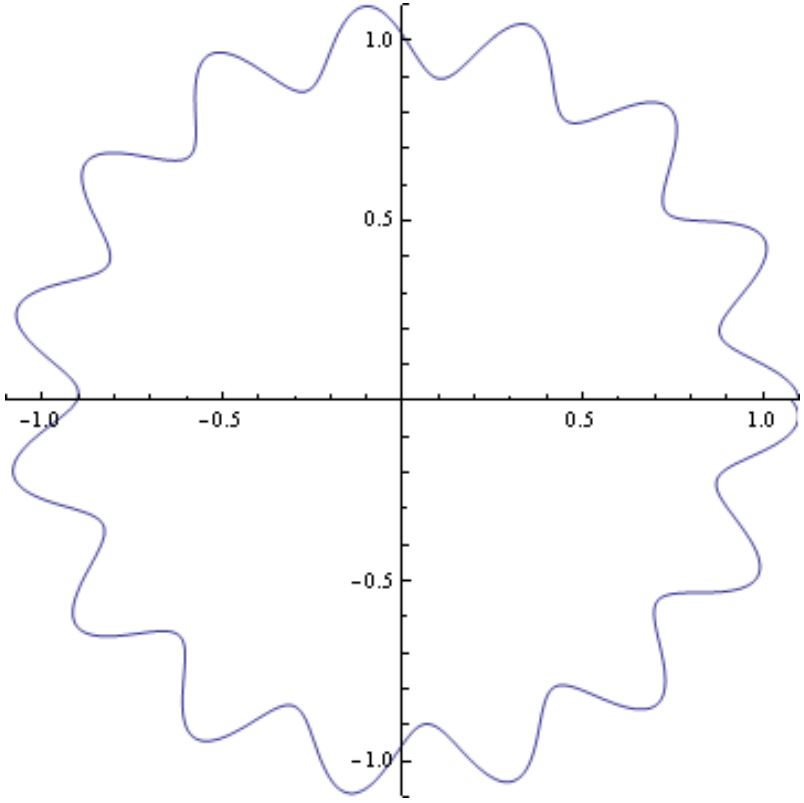
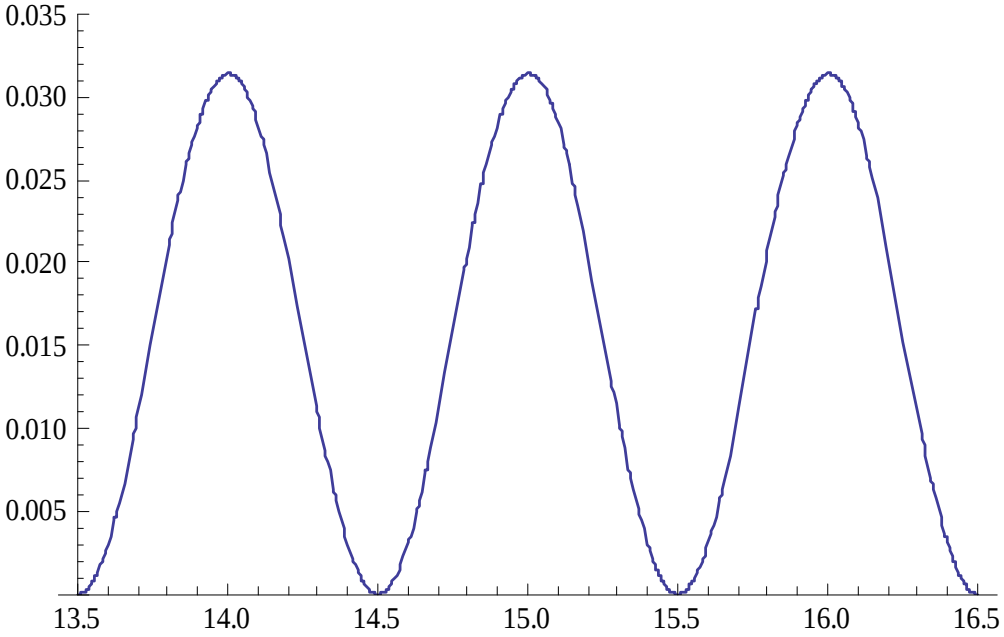


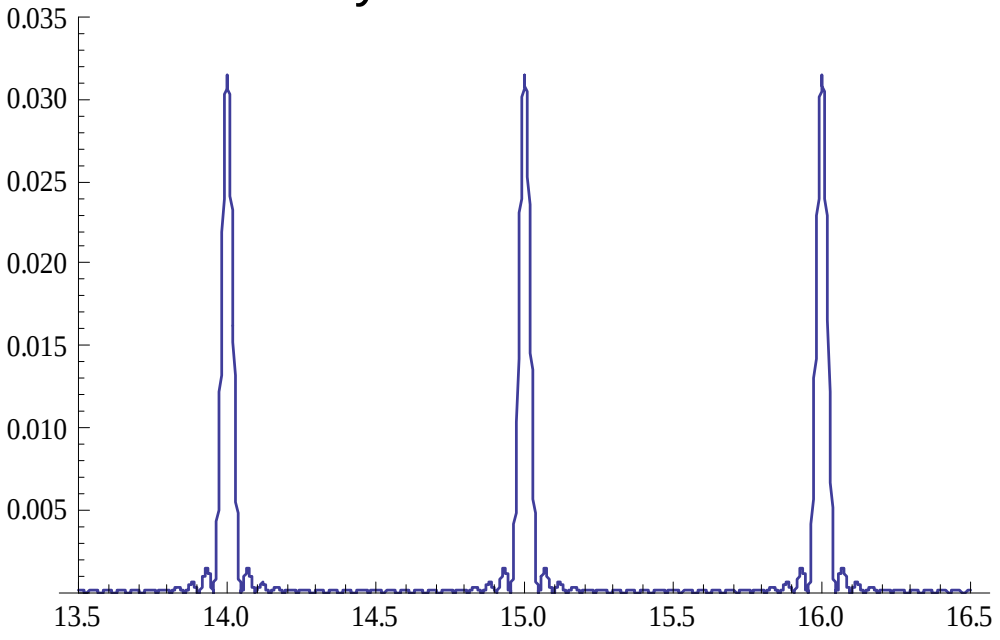
How can a non-stationary state survive?

After 2 cycles



$$\frac{1}{\pi\hbar}\,pr$$

After 45 cycles



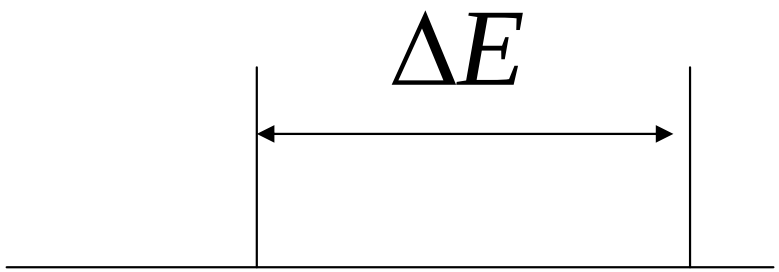
$$\frac{1}{\pi\hbar}\,pr$$

Stick Spectra can be observed only after an infinitely long time

Uncertainty Principle

Uncertainty Principle

$$\Delta E \Delta t \geq 2\pi\hbar$$



In order to distinguish two energy levels

$$\Delta\tau \geq \frac{2\pi\hbar}{\Delta E} \approx T$$

Observation time

Uncertainty limit

Classical period of motion

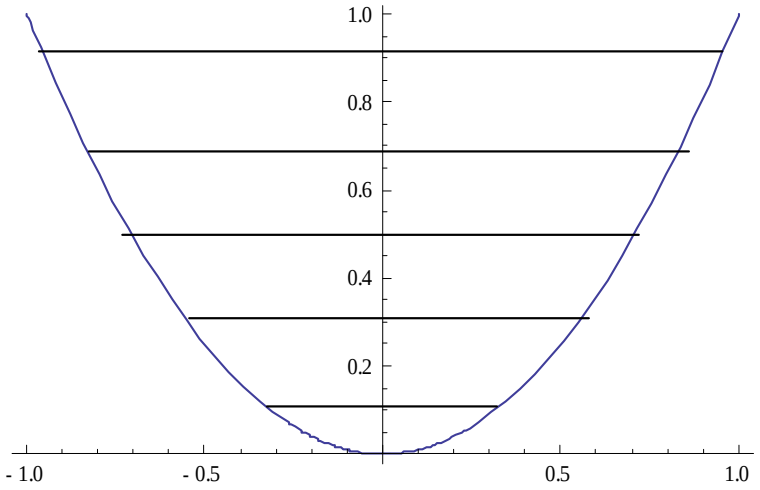
Classical period of motion

$$T \approx \frac{2\pi\hbar}{\Delta E}$$

Harmonic Oscillator

Energy Levels

$$E_n = \hbar\omega\left(n + \frac{1}{2}\right)$$



Energy Spacing

$$\Delta E = E_{n+1} - E_n = \hbar\omega$$

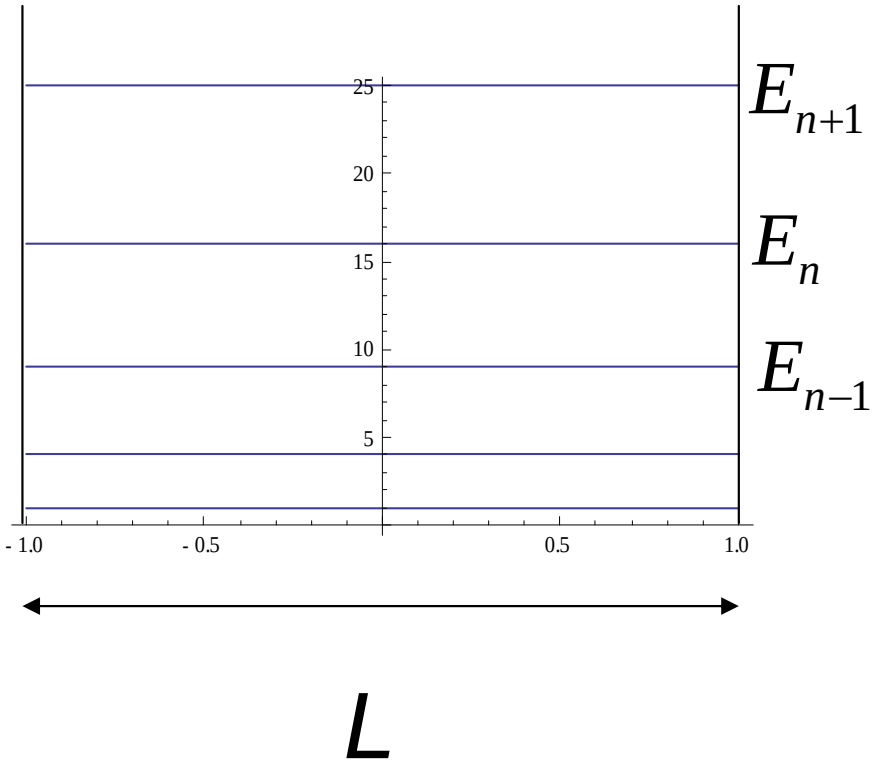
Classical Period of motion

$$T = \frac{2\pi}{\omega} \quad \Rightarrow \quad T = \frac{2\pi\hbar}{\Delta E}$$

Particle in a Box

Energy Levels

$$E_n = \frac{\hbar^2 n^2 \pi^2}{2mL^2}$$



Energy Spacing

$$\Delta E_n = \frac{\left(E_{n+1} - E_n\right) - \left(E_n - E_{n-1}\right)}{2}$$

$$= \frac{\hbar^2 \pi^2}{mL^2} n$$

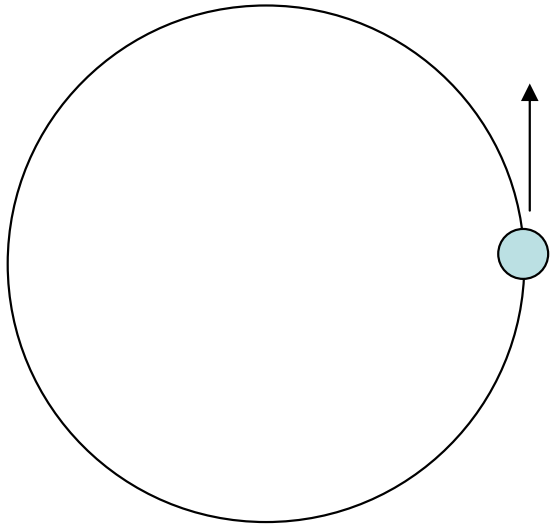
Period of motion

$$T_n = \frac{2L}{v} = \frac{2L}{\sqrt{2E_n / m}} = \frac{2mL^2}{n\pi\hbar} \quad \Rightarrow \quad T_n = \frac{2\pi\hbar}{\Delta E_n}$$

Rotational Motion

A Particle having angular momentum J

$$J = mrv = mr^2\dot{\theta}$$



Period of motion

$$T_J = \frac{2\pi}{\dot{\theta}} = \frac{2\pi mr^2}{J}$$

Energy Spacing obtained from the classical period

$$\Delta E_J = \frac{2\pi\hbar}{T_J} = \frac{J\hbar}{mr^2}$$

Energy Spacing

$$\Delta E_j = \frac{\left(E_{j+1} - E_j\right) - \left(E_j - E_{j-1}\right)}{2} = \frac{\hbar}{mr^2}\left(J + \frac{1}{2}\right)$$

Time-independent Hamiltonian

Amplitude: $\propto \frac{1}{\sqrt{p(r)}} \propto \frac{1}{\sqrt{v(r)}}$

Wavelength: $\propto \frac{2\pi\hbar}{p(r)}$

Interference: Uncertainty principle

Energy Spacing: $\Delta E = \frac{2\pi\hbar}{T_{classical}}$

Eigenstate: Time independent shape

Superposition of two Eigenstates:
time-dependent shape with period $\Delta E = \frac{2\pi\hbar}{T}$

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

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

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

Transition amplitude

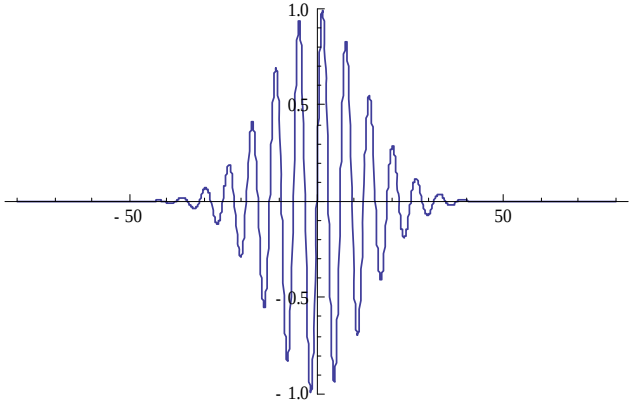
$$c_j(\infty) = \int_{-\infty}^{\infty} \langle \psi_0 | H'(\tau) | \psi_j \rangle d\tau = \vec{\mu}_{0j}\cdot\vec{F}(\Delta E)$$

Laser Field in the Energy Domain

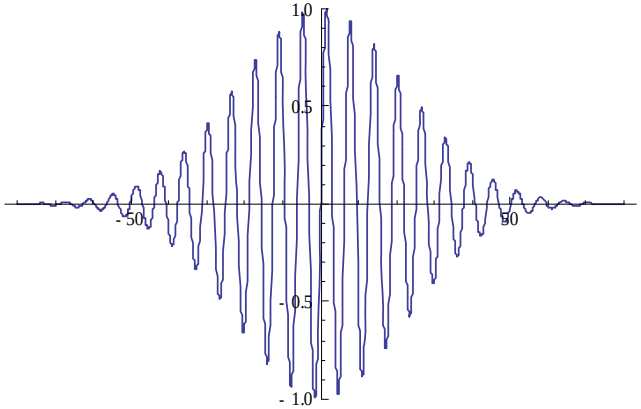
$$\vec{F}(\Delta E) = \int_{-\infty}^{\infty} dt \exp\left[\frac{i}{\hbar}(\Delta E)t\right]\vec{F}(t)$$

Laser Pulse

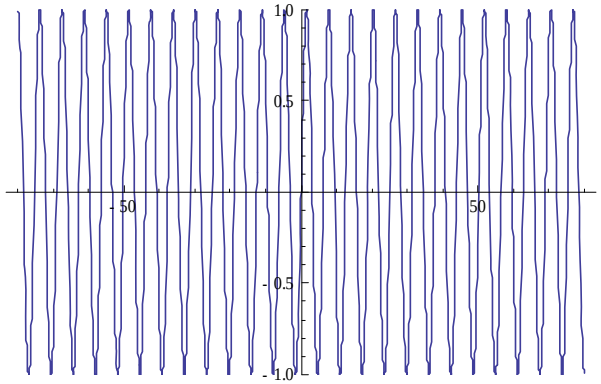
Short-Pulsed Laser



Long-Pulsed Laser

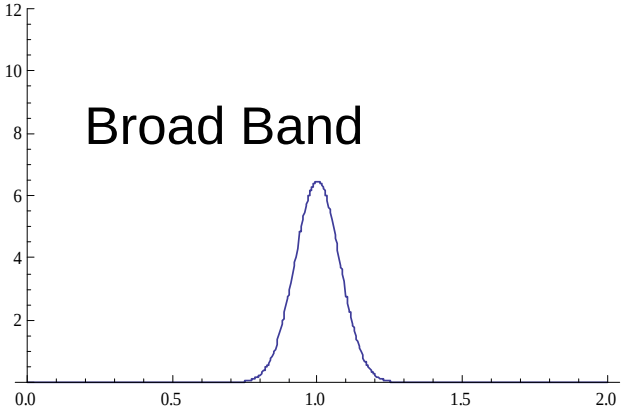


CW Laser

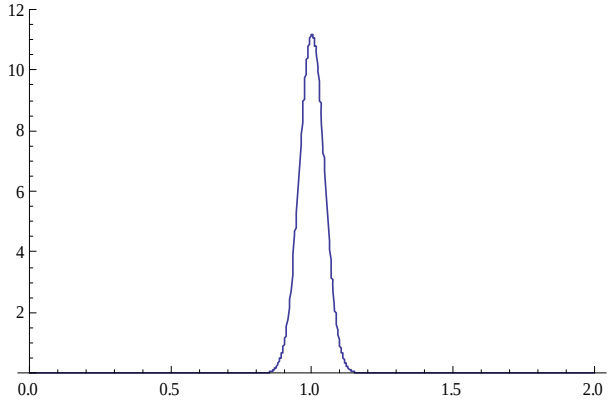


Time Domain

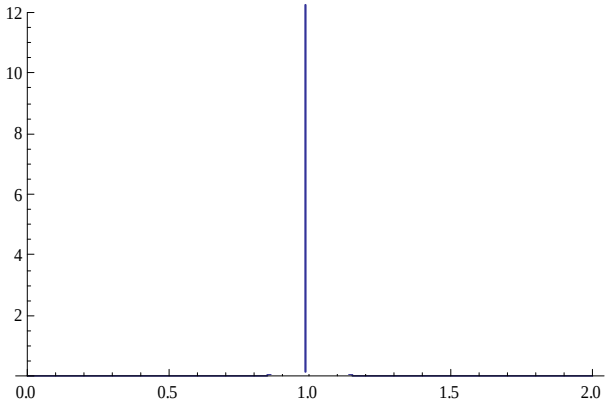
Broad Band



Narrow Band



Monochromatic



Frequency Domain

Linearity of the Perturbation Theory

Additivity

$$H = H_0 + H_a(t) + H_b(t)$$

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

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

Homogeneity

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

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

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

Time-Periodic Interaction

Floquet Theory
Dressed State Picture

Floquet Theory

(Exact Treatment for periodic Hamiltonian)

Schrodinger Equation

$$i\hbar \frac{d}{dt}\Psi(t) = H(t)\Psi(t)$$

Time periodic Hamiltonian

$$H(t+T) = H(t)$$

Wavefunction (the Floquet theorem)

$$\psi_j(t) = \exp\left(-i\frac{\varepsilon_j}{\hbar}t\right)\phi_j(\tau)$$

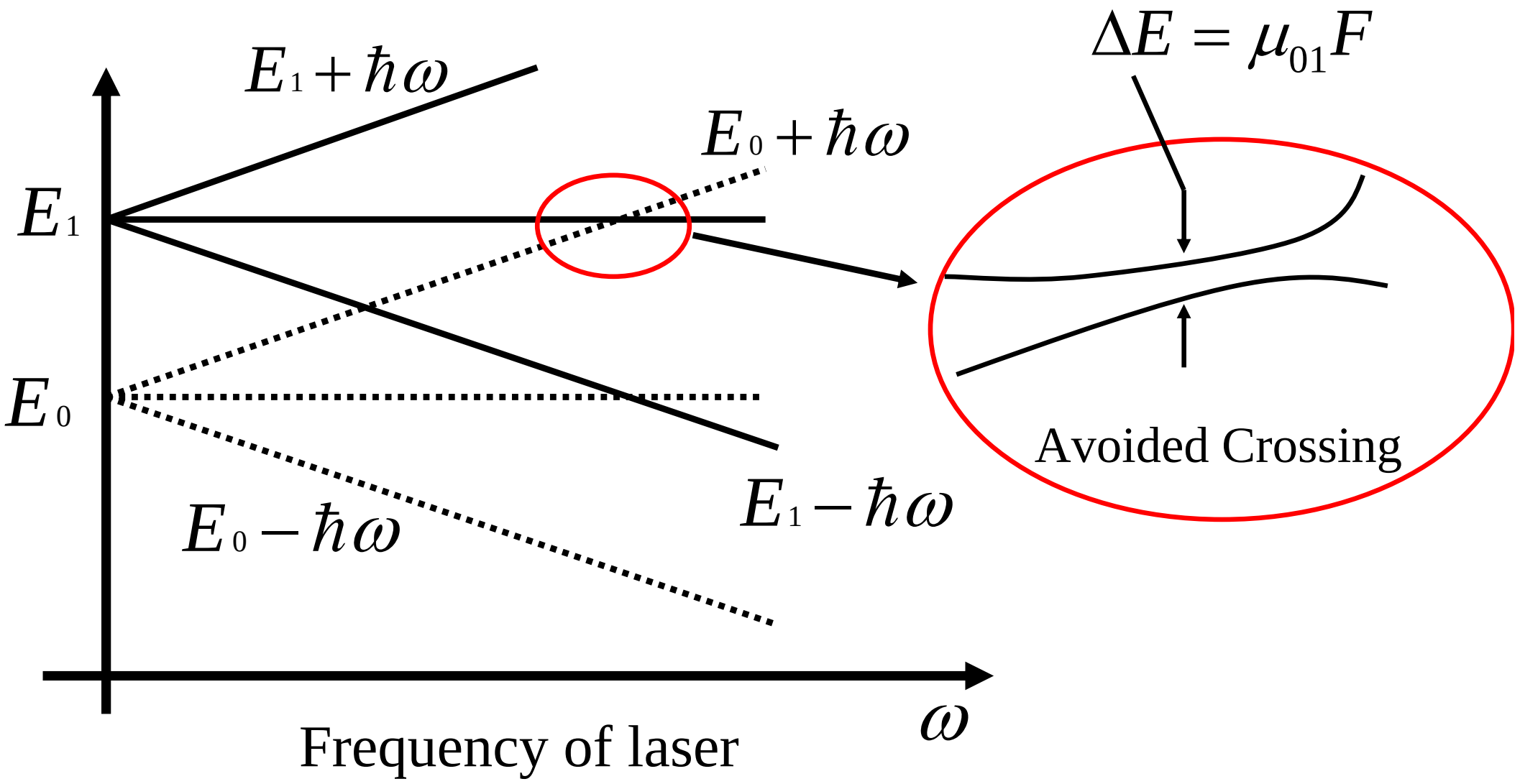
ε_j: Quasi-Energy

$$\phi_j(\tau+T) = \phi_j(\tau)$$

$\phi_j(\mathrm{t})$: Quasi-State

Atom + Laser

Quasi-state: (atom + photon + atom-photon interaction)



Stationary VS Floquet

Time-independent Potential

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

Eigenstate basis

$$\Psi_j\left(r,t\right)=e^{-iE_jt/\hbar}\Phi_j\left(r\right)$$

time-periodic Potential

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

Quasi-state basis

$$\psi_j\left(r,t\right)=e^{-i\varepsilon_jt/\hbar}\phi_j\left(r,\tau\right)$$

$$\left(t=nT+\tau\right)$$

Initial state

$$\Psi\left(r,0\right)=\Phi_{j=0}\left(r\right)$$

Initial state

$$\psi\left(r,0\right)=\sum_j c_j\phi_j\left(r,0\right)$$

Two-state case

Initial state

$$\psi(r,0) = \sum_{j=1}^{2} c_j \phi_j(r,0)$$

$$\psi(r,t) = c_1 \phi_1(r,\tau) e^{-i\varepsilon_1 t/\hbar} + c_2 \phi_2(r,\tau) e^{-i\varepsilon_2 t/\hbar}$$

Superposition state → Periodic motion of wavefunction

Rabi-frequency

$$\Omega = \frac{1}{\hbar}(\varepsilon_2 - \varepsilon_1)$$

Rotating-wave Approximation

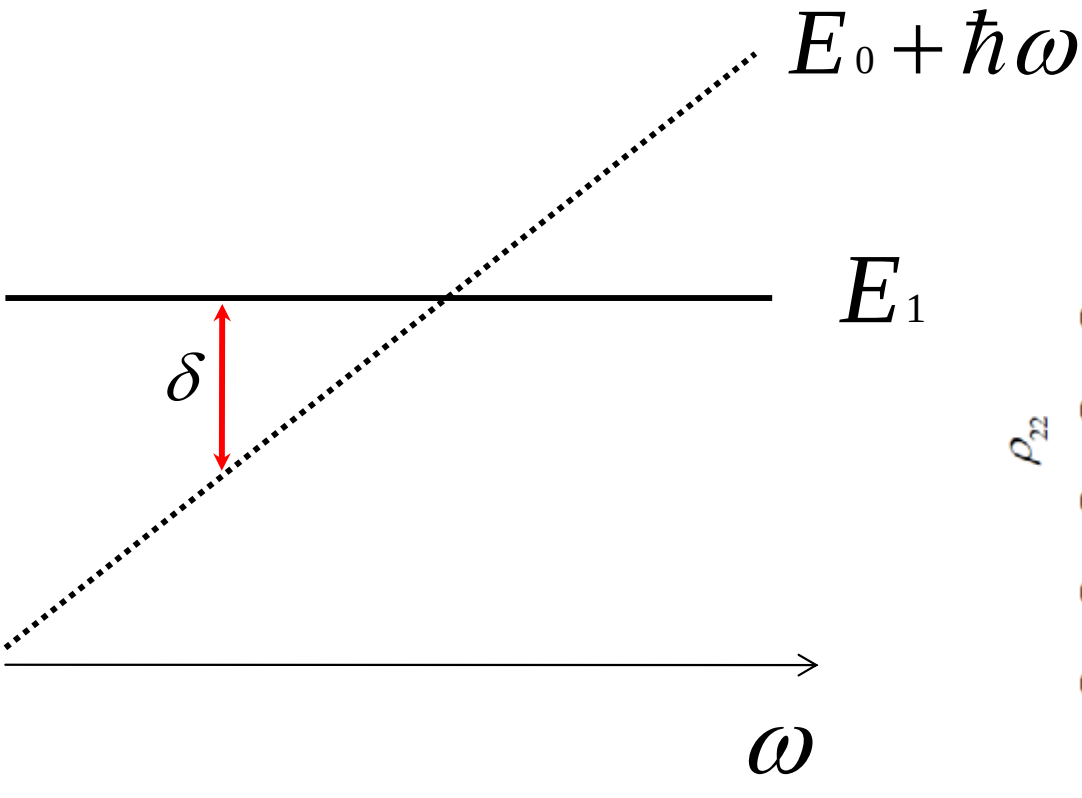
$\left(\text{Consider only } E_0 + \hbar\omega,\ E_1,\ \text{and } \mu F\right)$

Excitation Probability

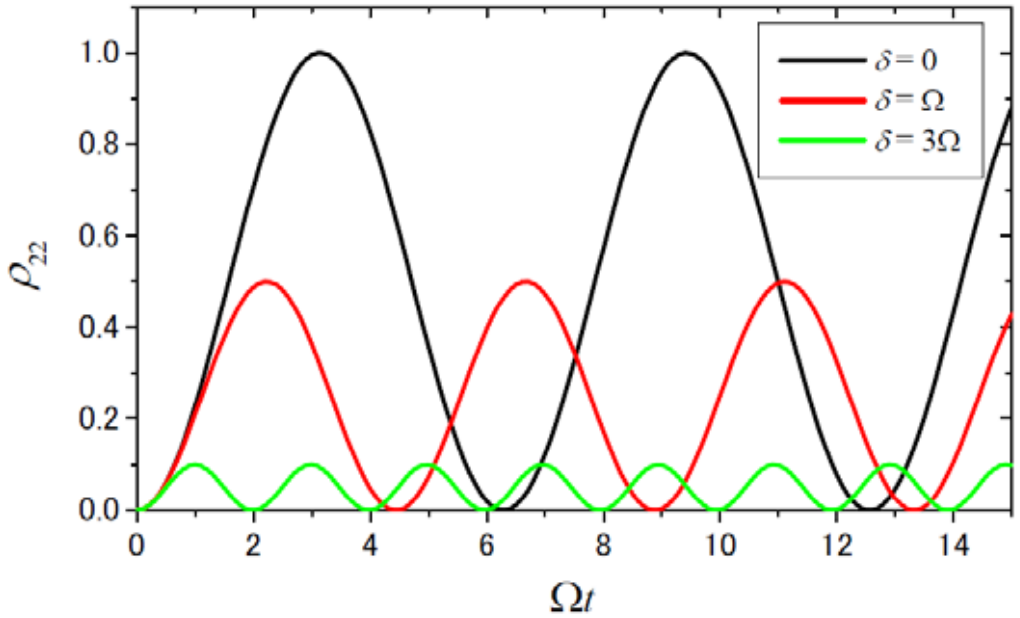
$$P_{12}(t) = \frac{(\mu F)^2}{\delta^2 + (\mu F)^2} \sin^2 \frac{\Omega}{2} t$$

Rabi Frequency

$$\Omega = \frac{1}{\hbar}(\varepsilon_2 - \varepsilon_1) = \frac{1}{\hbar}\sqrt{\delta^2 + (\mu F)^2}$$



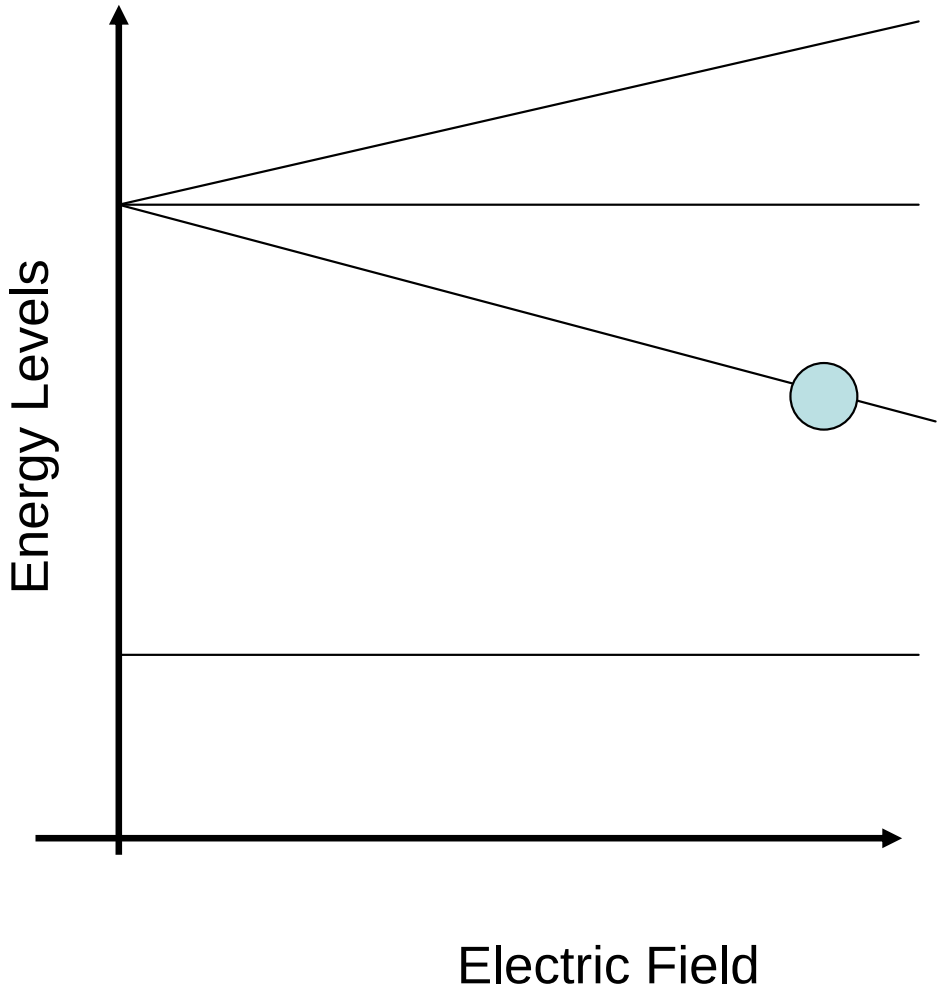
Excitation Probability



Slow Limit

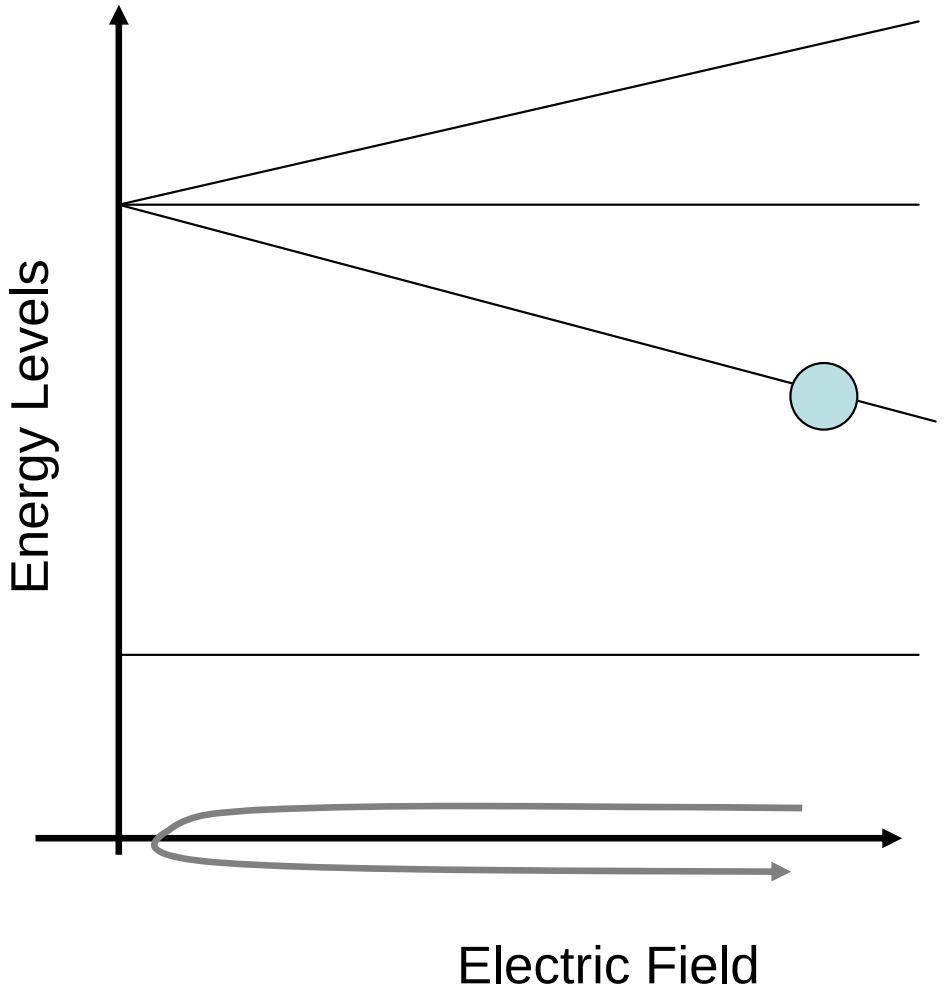
Adiabatic Approximation

Adiabatic Approximation
Example: Stark Effect



$$H(r, f)\varphi_j^{(a)}(r, f) = E_j(f)\varphi_j^{(a)}(r, f)$$

Adiabatic Approximation
Example: Stark Effect



$$\phi_j^{(a)}(r,t) = \exp\left[-i\int_0^t \frac{E_j(\tau)}{\hbar} d\tau\right]\varphi_j^{(a)}(r,t)$$

$$H(r,t)\varphi_j^{(a)}(r,t) = E_j(t)\varphi_j^{(a)}(r,t)$$

$(t$ is assumed constant$)$

Nonadiabatic Transition
Transition due to breakdown of the adiabatic approximation

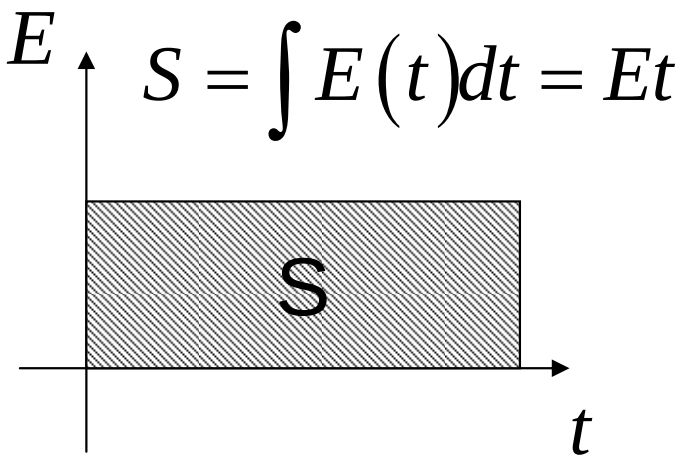
Stationary and Adiabatic

Time-independent Potential

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

Eigenstate

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

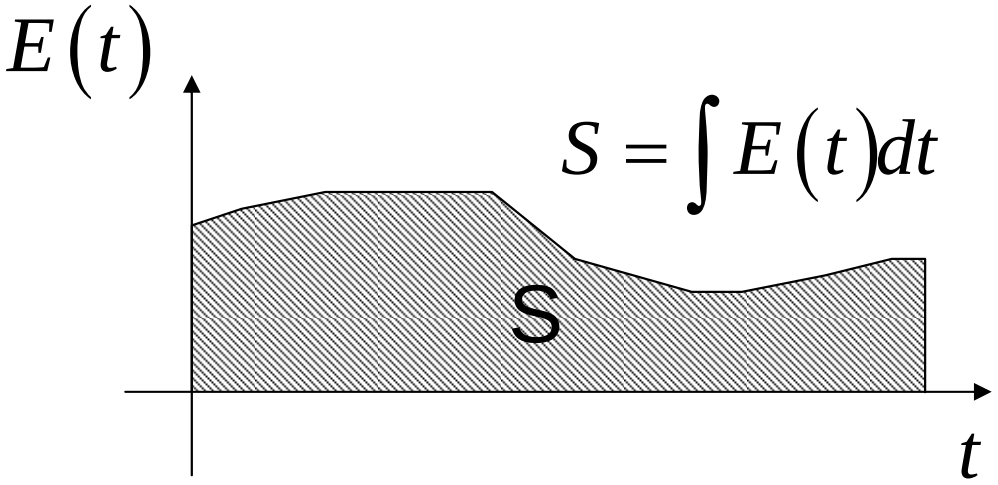


time-dependent Potential

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

Adiabatic

$$\Psi(r,t)=\exp\left[-i\int_0^t\frac{E(\tau)}{\hbar}d\tau\right]\Phi(r,t)$$



WKB and Adiabatic

r-dependent Potential

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

time-dependent Potential

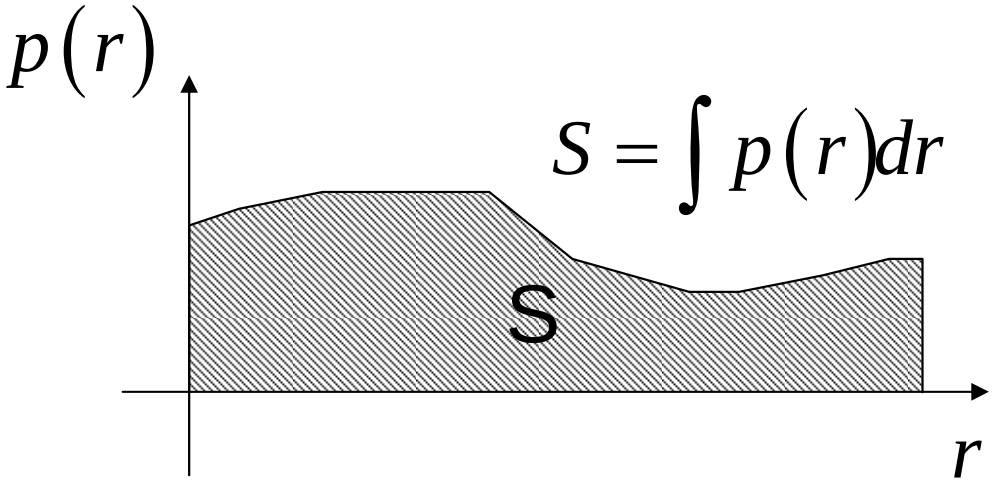
$$i\hbar\frac{d}{dt}\Psi(r,t)=H(r,t)\Psi(r,t)$$

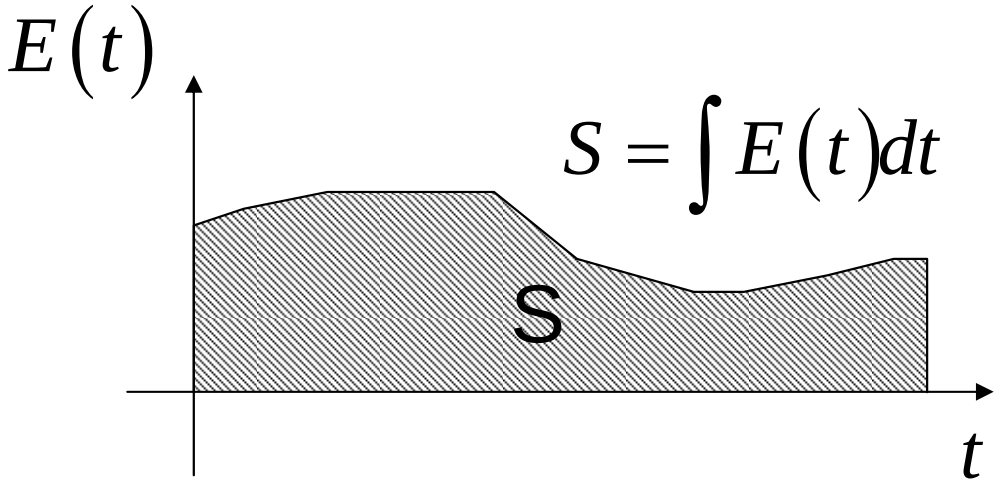
WKB

$$\Phi(r)=\frac{A}{\sqrt{p(r)}}\exp\left[\pm\frac{i}{\hbar}\int p(r)dr\right]$$

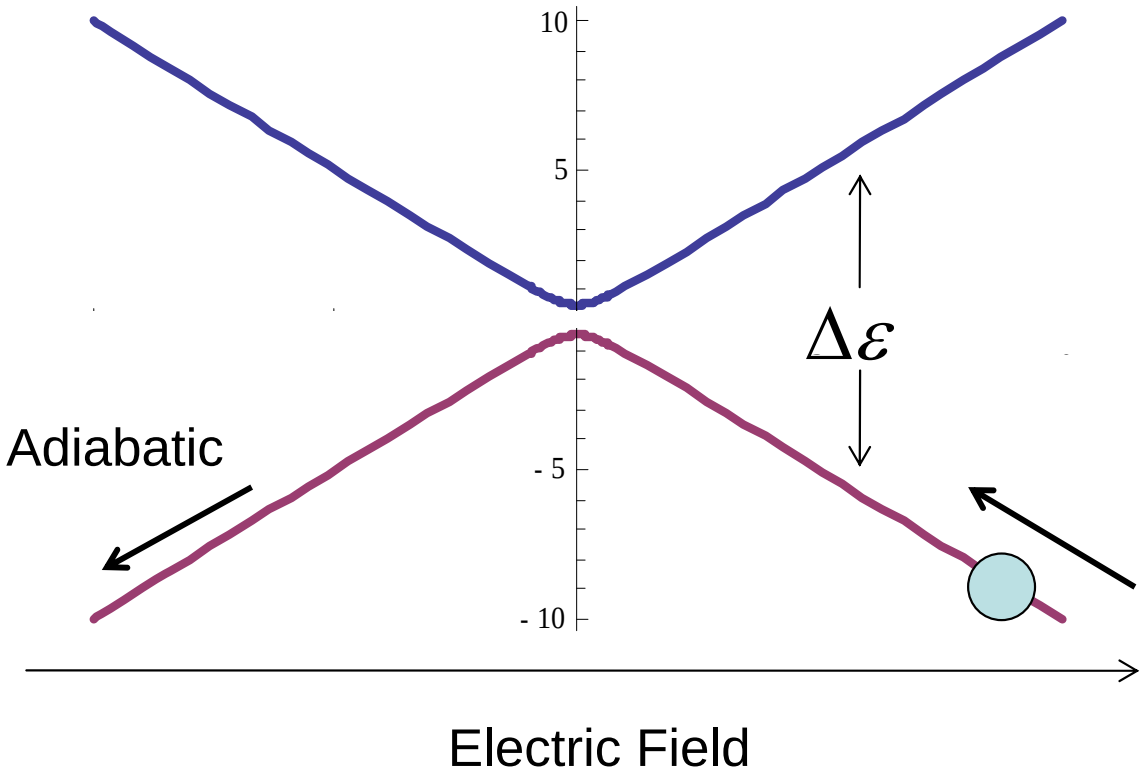
Adiabatic

$$\Psi(r,t)=\exp\left[-i\int_0^t\frac{E(\tau)}{\hbar}d\tau\right]\Phi(r,t)$$





Adiabatic? Nonadiabatic?



Rapid sweeping of the field

↓

Short time stay at a point

↓

Large energy uncertainty

↓

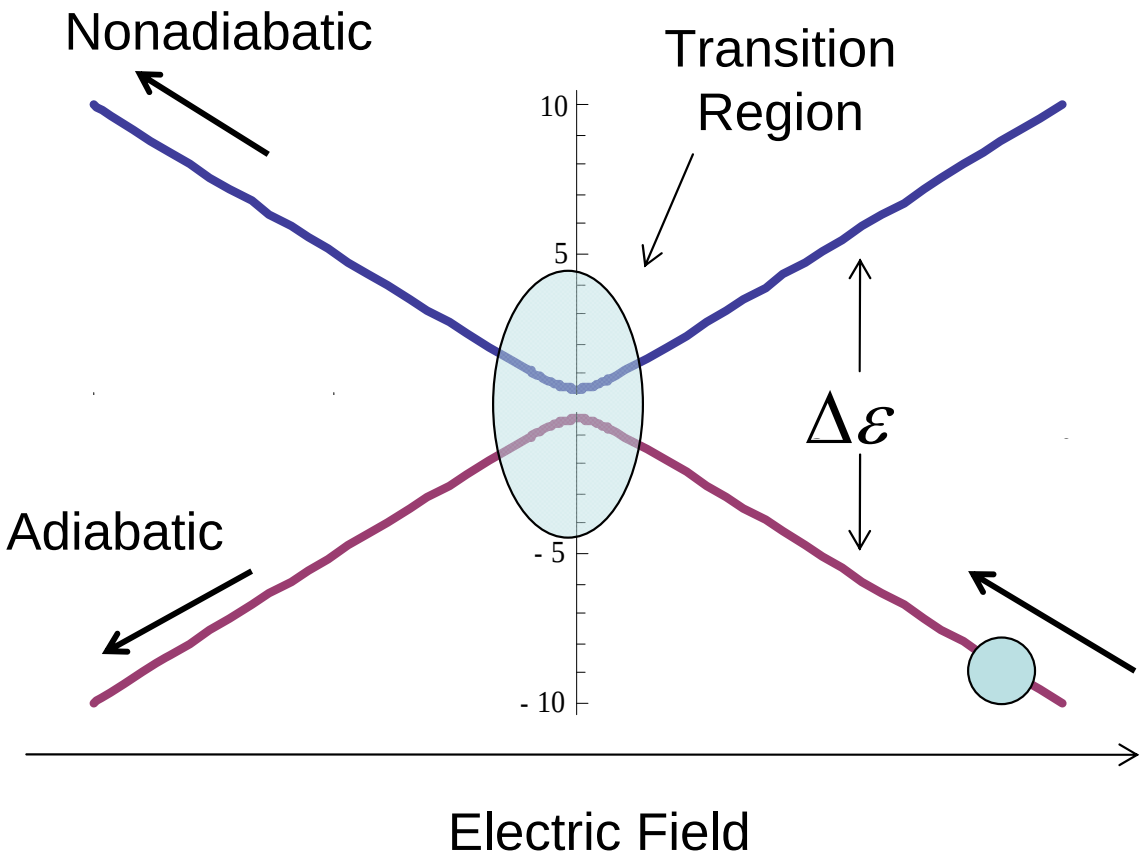
$\Delta E \geq \Delta\varepsilon$

↓

Nonadiabatic Transition

Fast sweep and small energy spacing -> nonadiabatic

Adiabatic? Nonadiabatic?



Rapid sweeping of the field

↓

Short time stay at a point

↓

Large energy uncertainty

↓

$\Delta E \geq \Delta\varepsilon$

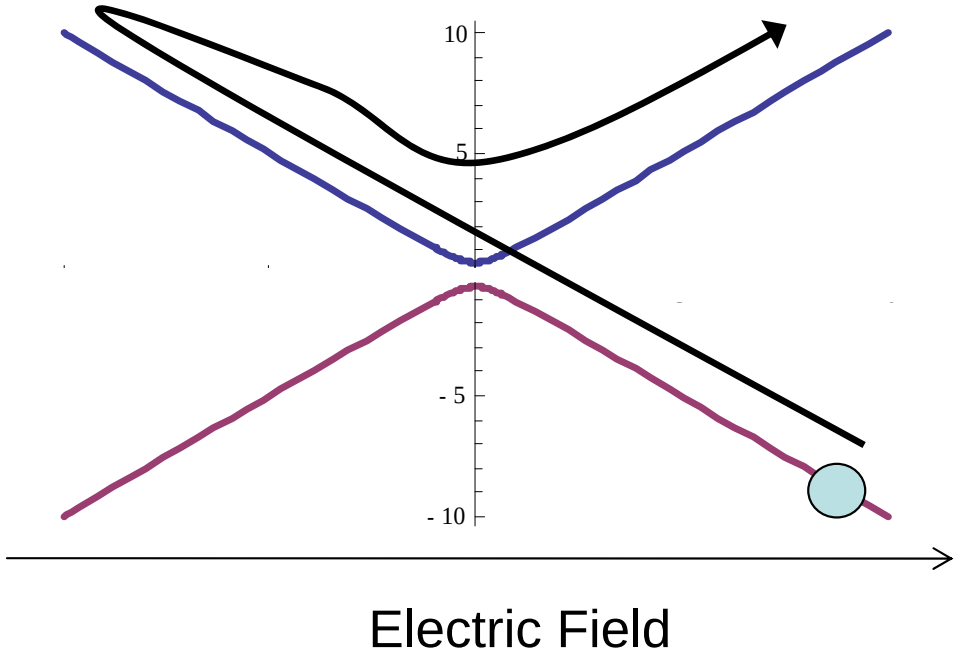
↓

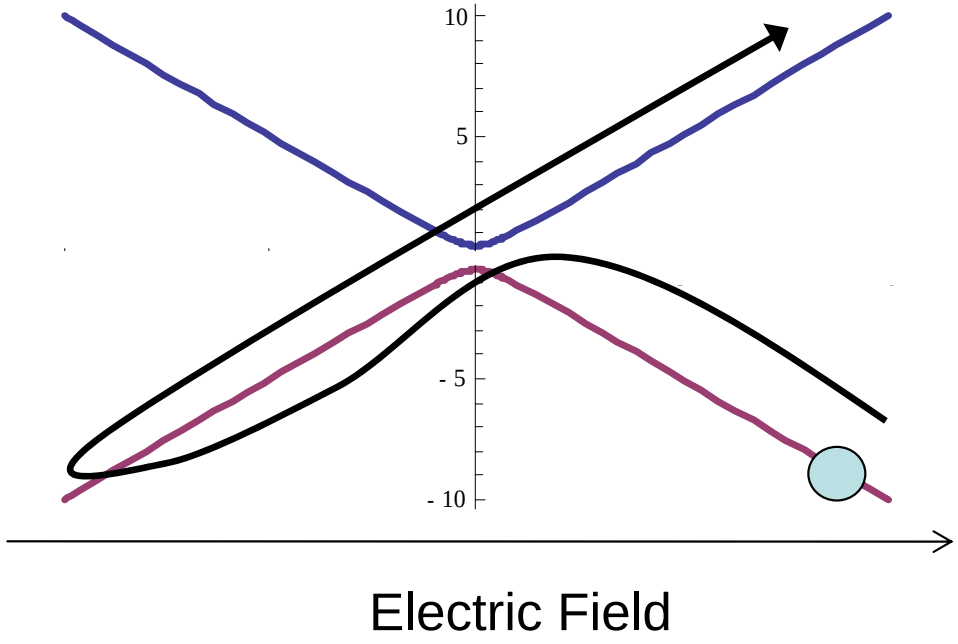
Nonadiabatic Transition

Fast sweep and small energy spacing -> nonadiabatic

Excitation after one period of oscillation

$$S = \int E(t)dt$$

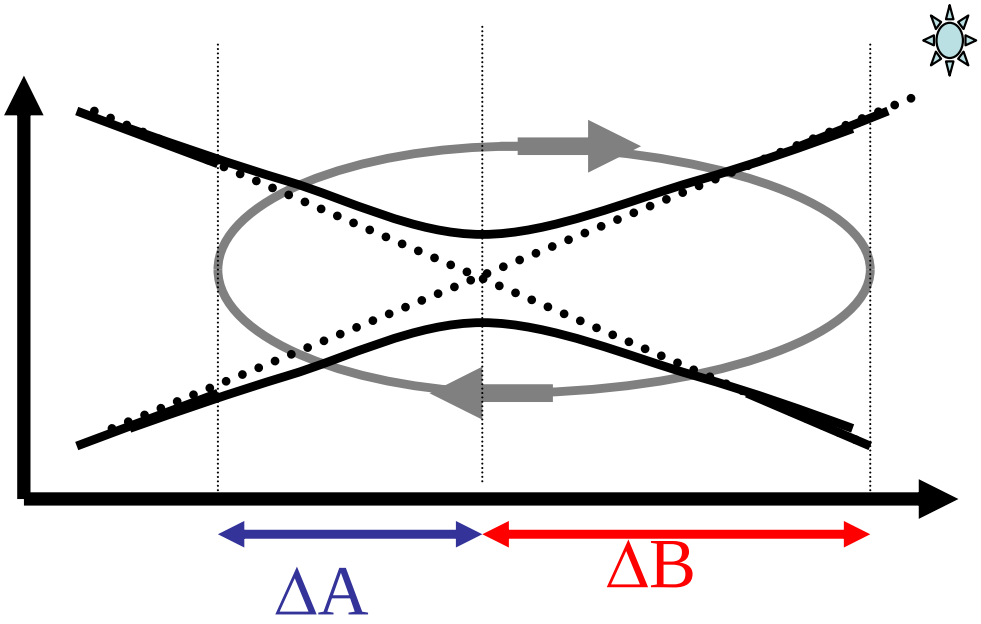




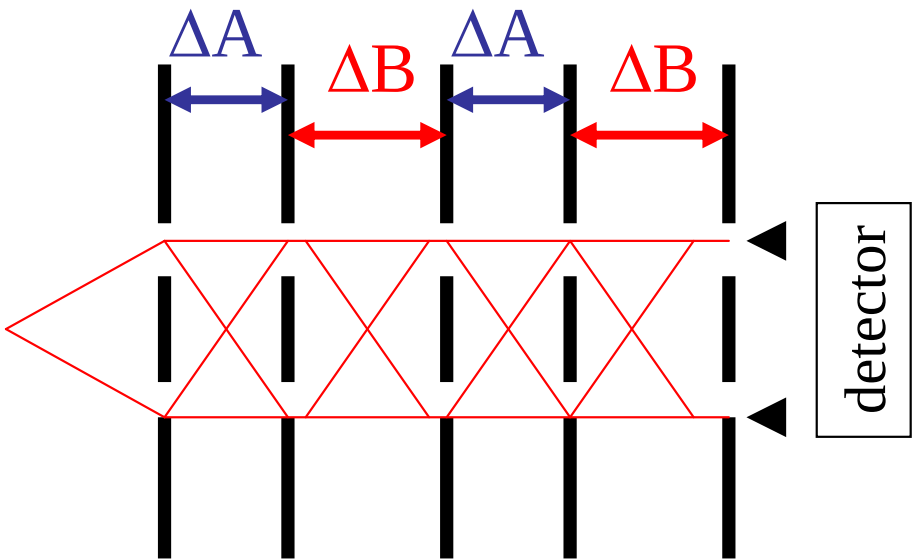
Two paths interference

Control of nonadiabatic transition

Teranishi and Nakamura, Phys. Rev. Lett. 81, 2032

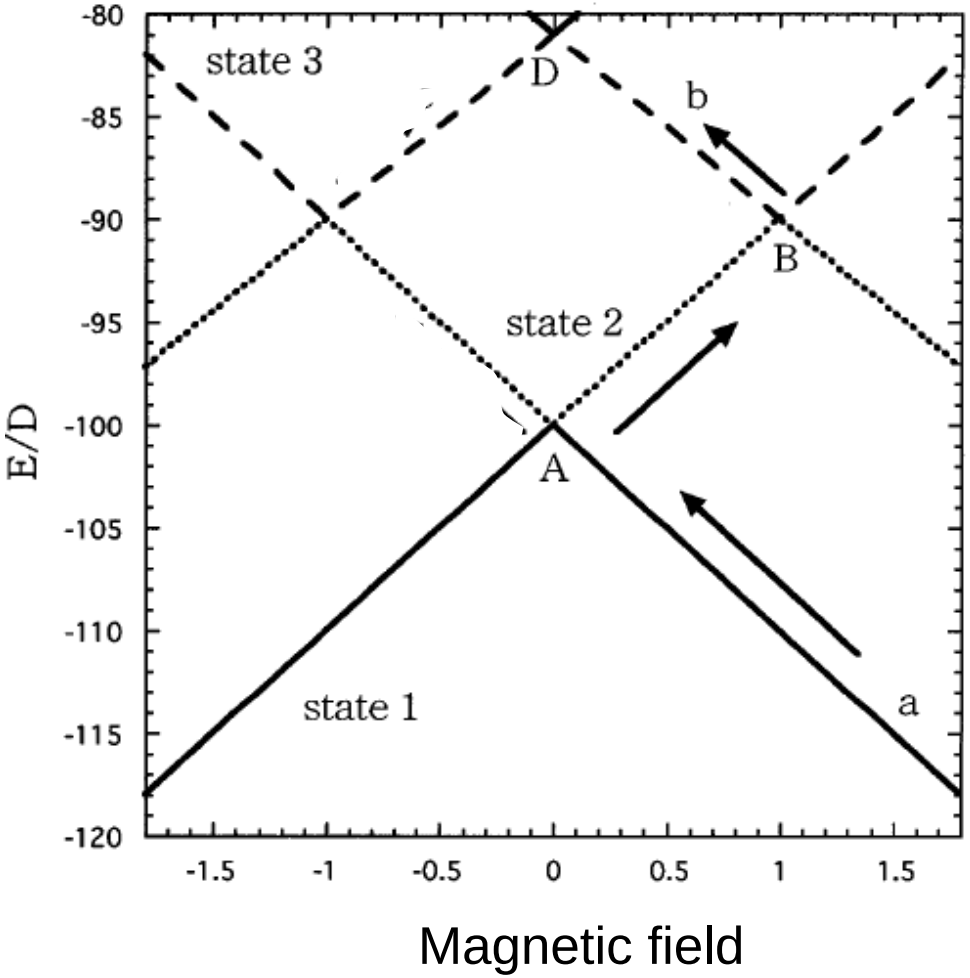


- Periodic sweep of adiabatic parameter
  - Bifurcation at the crossing
  - Phase can be controlled by ΔA, ΔB
  - Interference effects

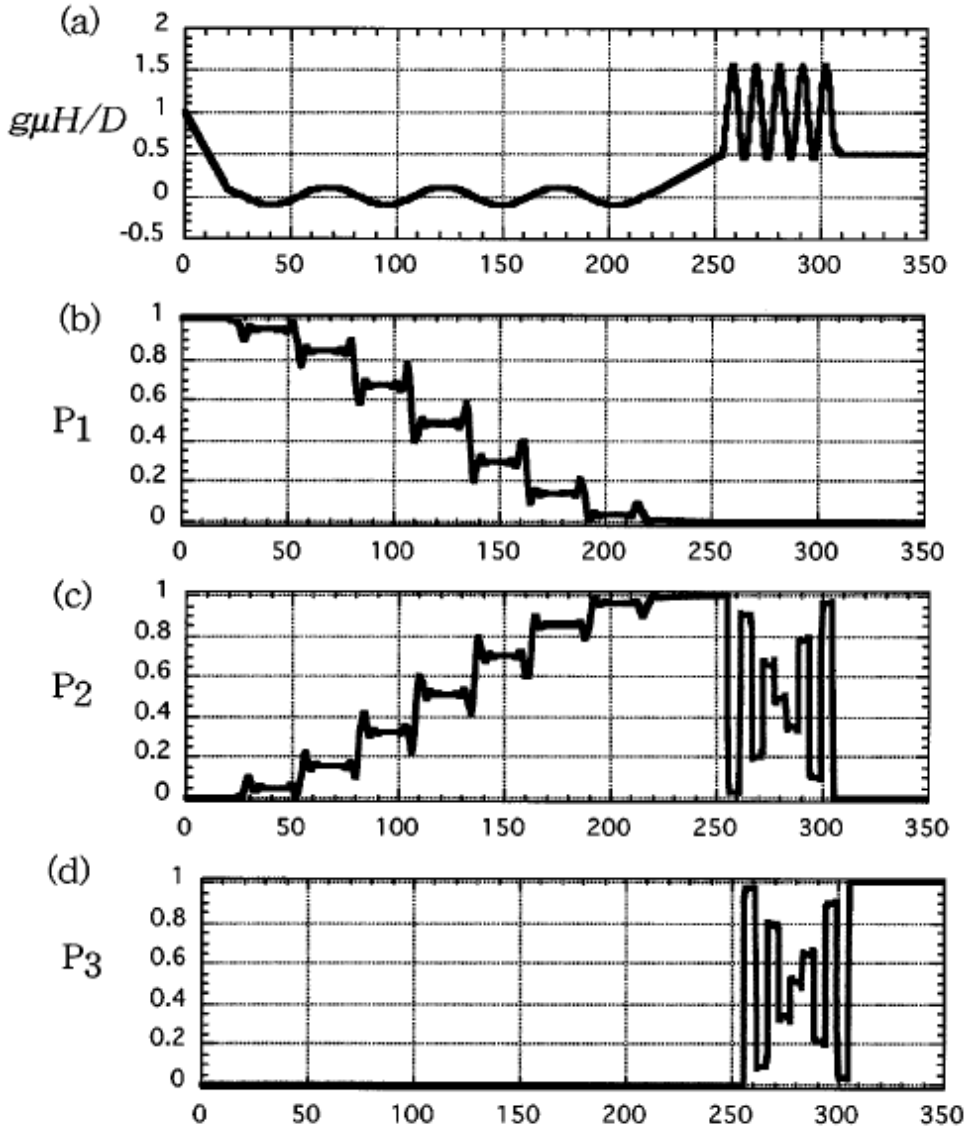


- Multiple double slits
  - Bifurcation at slits
  - Interference can be controlled by ΔA, ΔB

Example of control

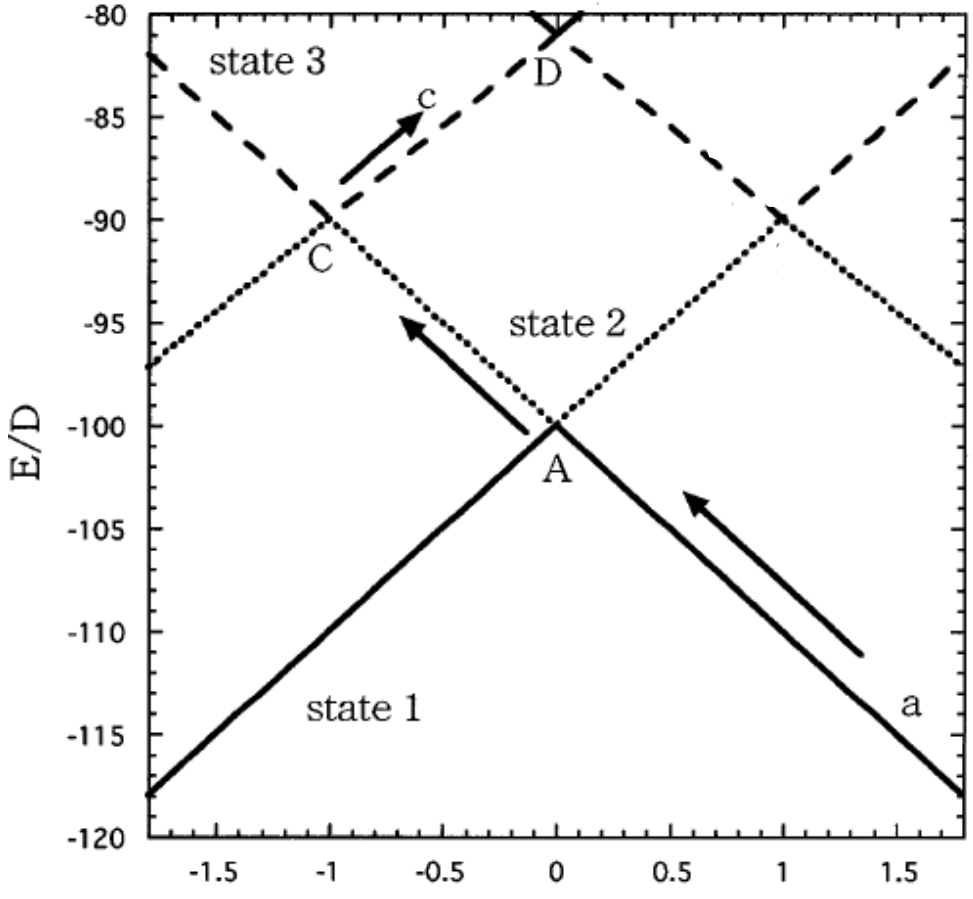


Magnetic field

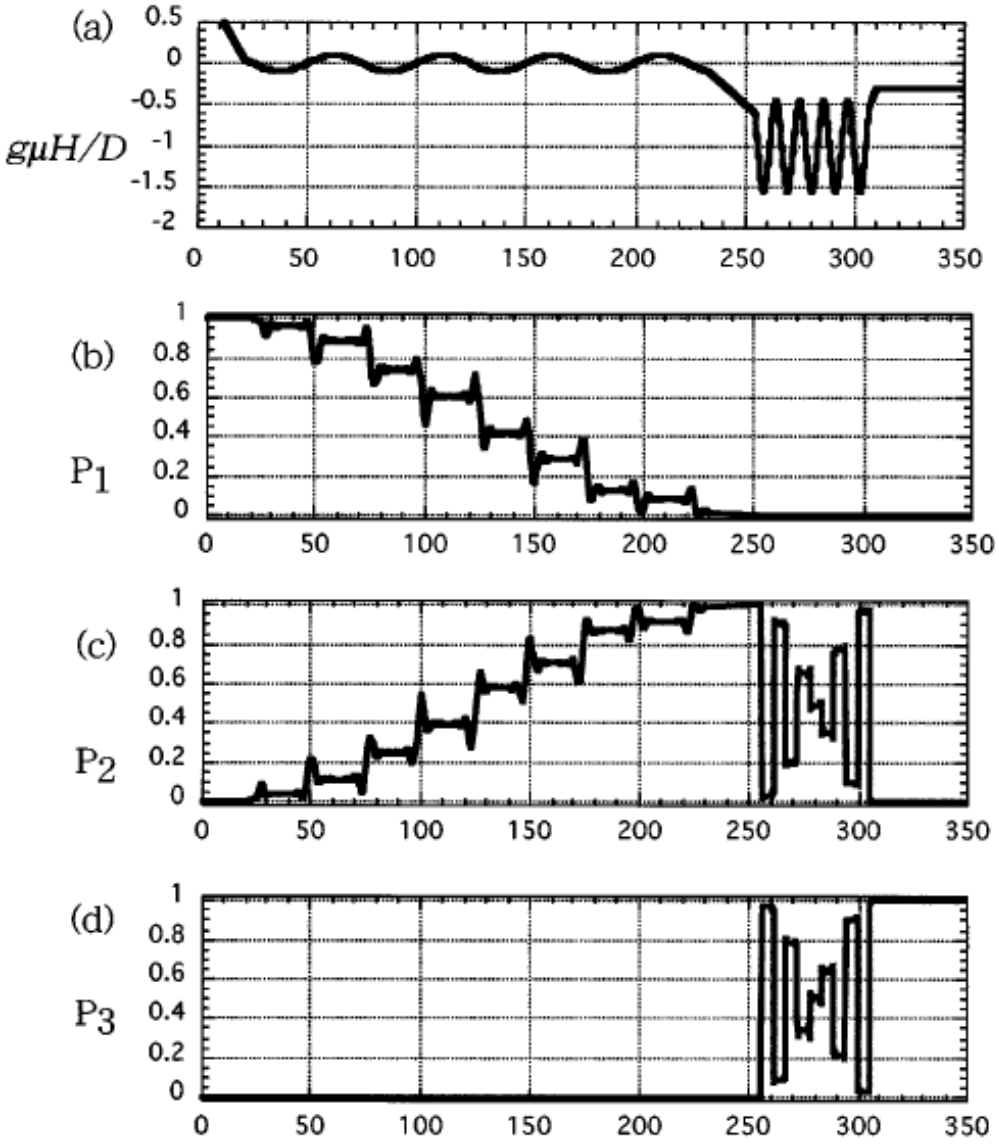


Time

Example of control



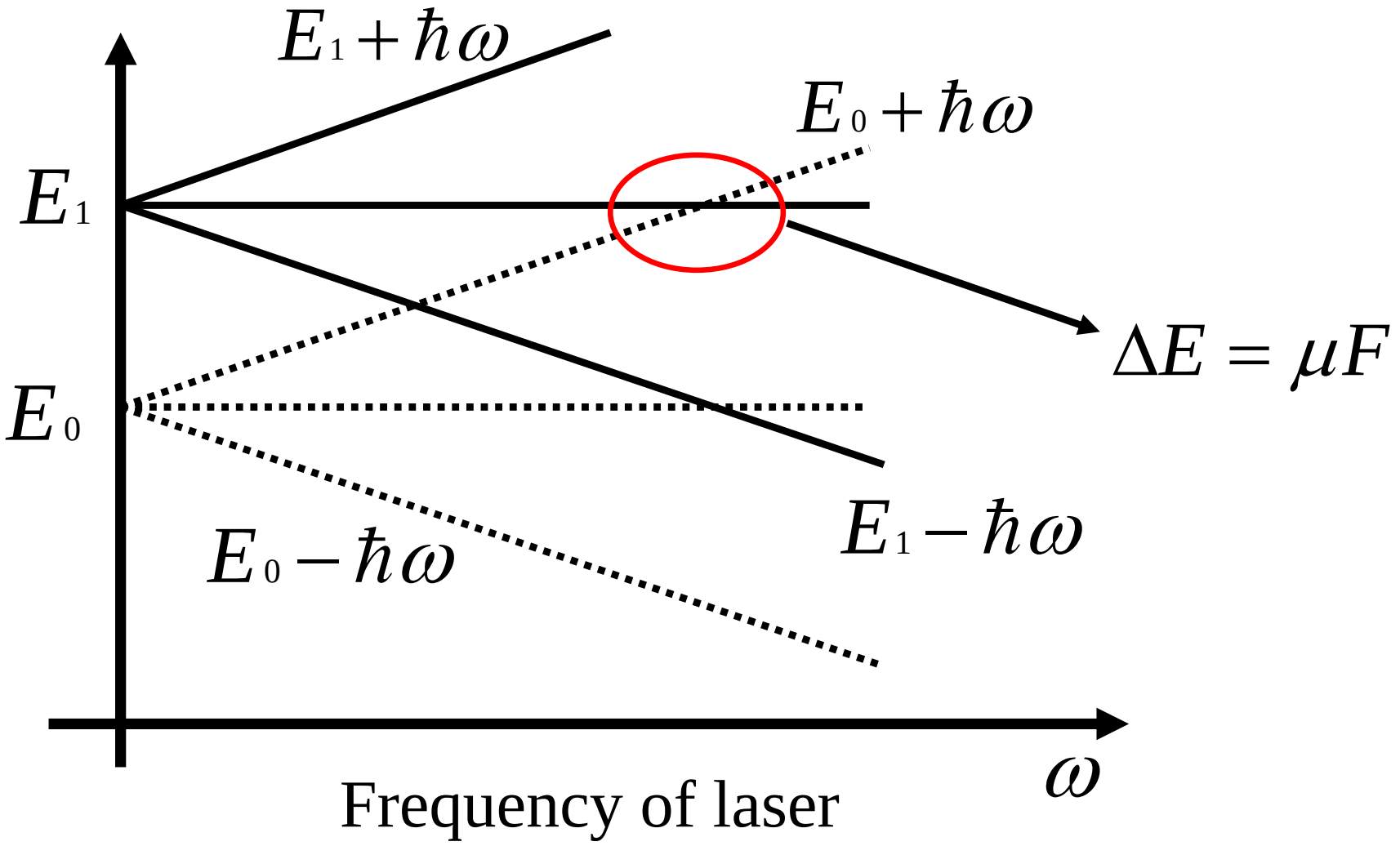
Magnetic field



Time

Laser Control with shaped pulse

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



Time-dependent problem

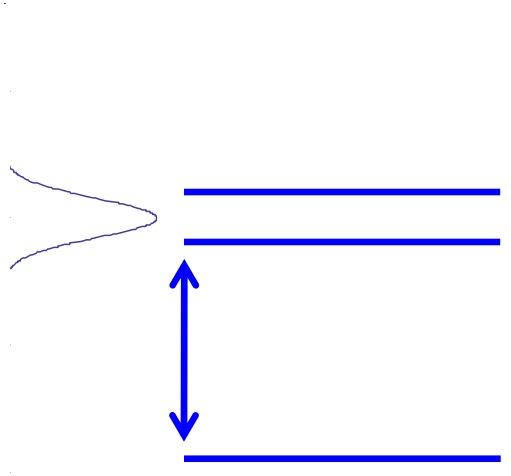
- Perturbation (Weak field limit)
  Transition amplitude → Fourier Component
  Linearity (Additivity and Homegeneity)

- Floquet (Periodic)
  Eigenstate →Superposition of quasi-states

- Adiabatic (Slow case) and Nonadibatic
  Periodic sweep of adiabatic parameter
  → Control

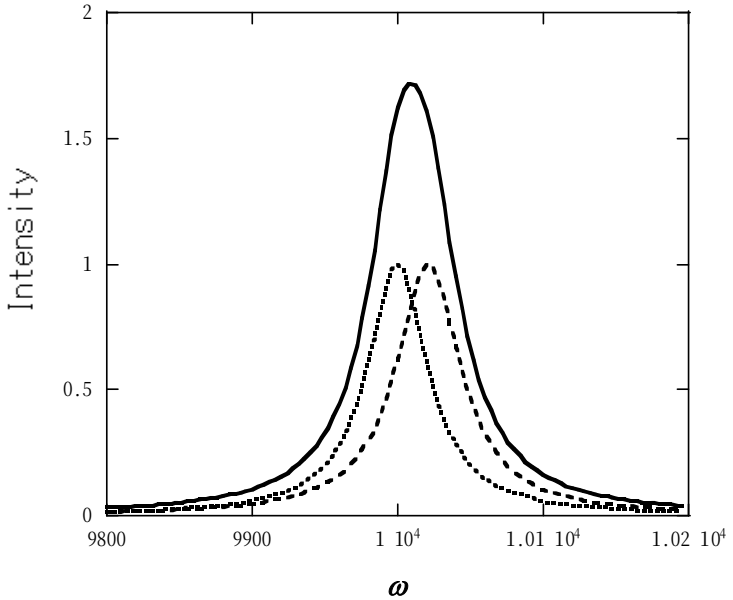
Outline

- Introduction to Quantum Mechanics
  - Schrodinger Equation
  - Stationary Problem
  - Time-Dependent Problem



Quantum Control

- Ultrafast Selective Excitation
  - A pair of weak pulses
  - Quadratic Chirping

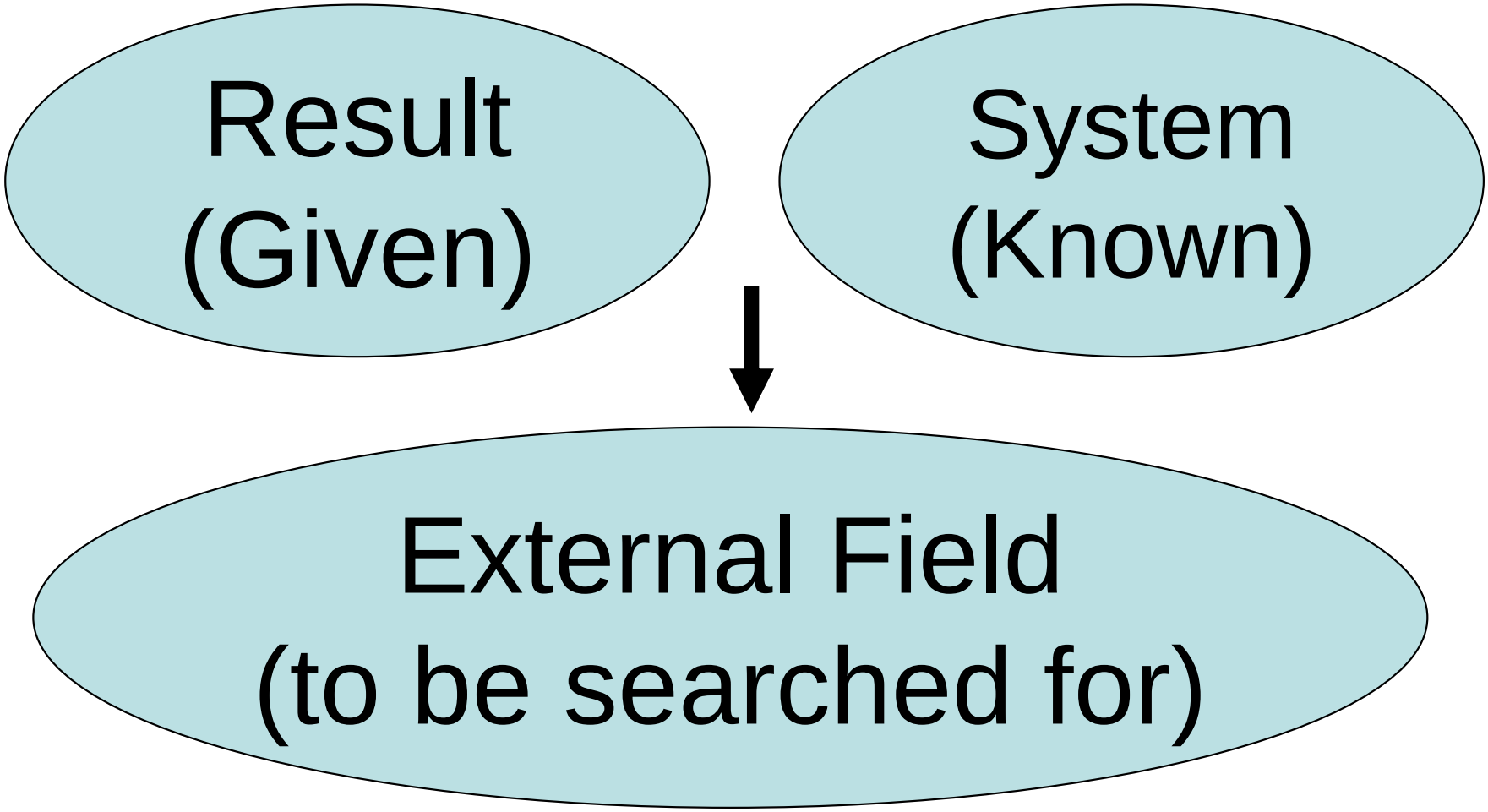


- Quantum Control Spectroscopy
  (overlapping by natural width)

- Molecular computer
  (Classical computer)

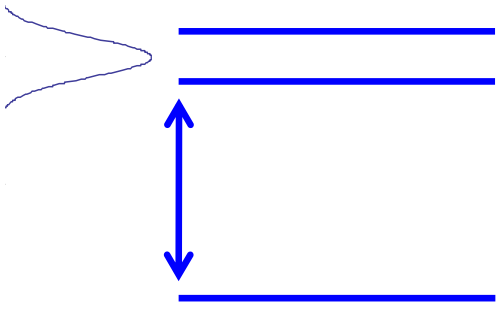
Laser Control of Atomic and Molecular Processes

Quantum Control



Inverse problem

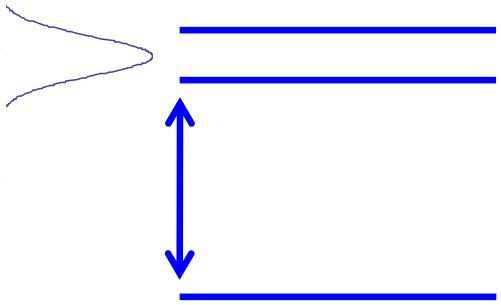
Ultrafast Selective Excitation



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)

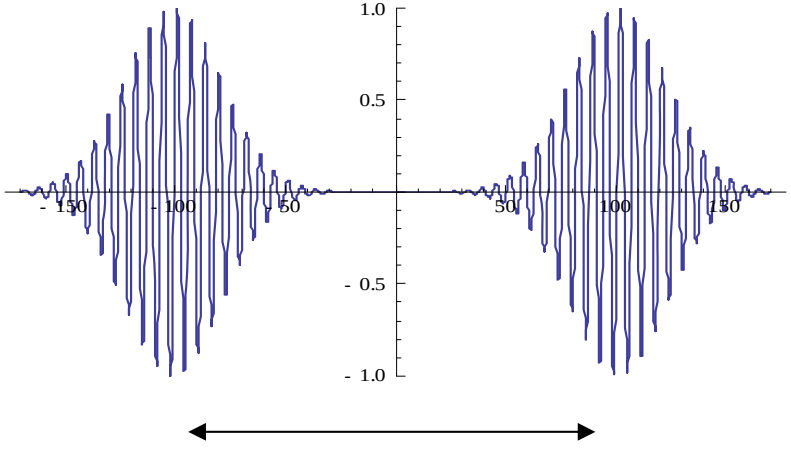
$$\vec{F}(t) = \exp\left[-\frac{t^2}{\Delta T^2}\right]\exp[i\omega t] + \exp\left[-\frac{(t-\tau)^2}{\Delta T^2}\right]\exp\left[i\{\omega(t-\tau)-\delta\}\right]$$

Phase Difference

$c_j^1(t)$

Interference

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



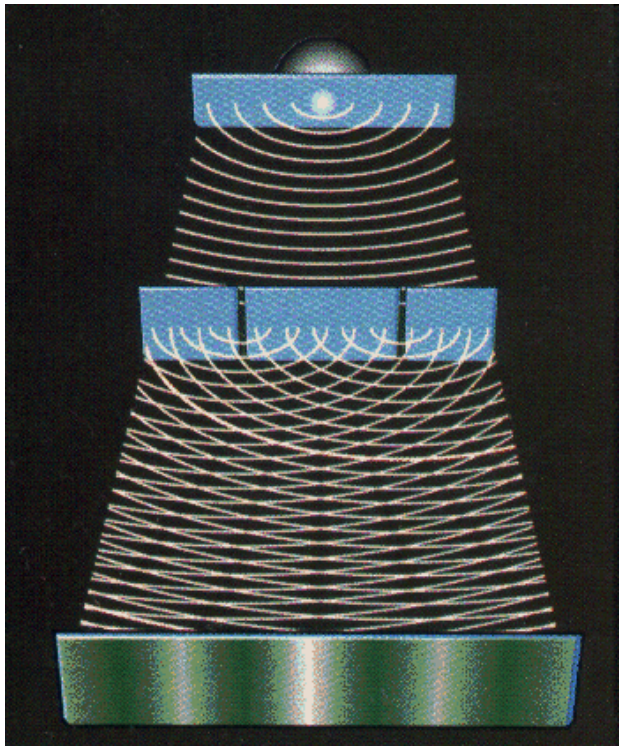
$|\tau$: Time delay $|$

Transition Amplitude

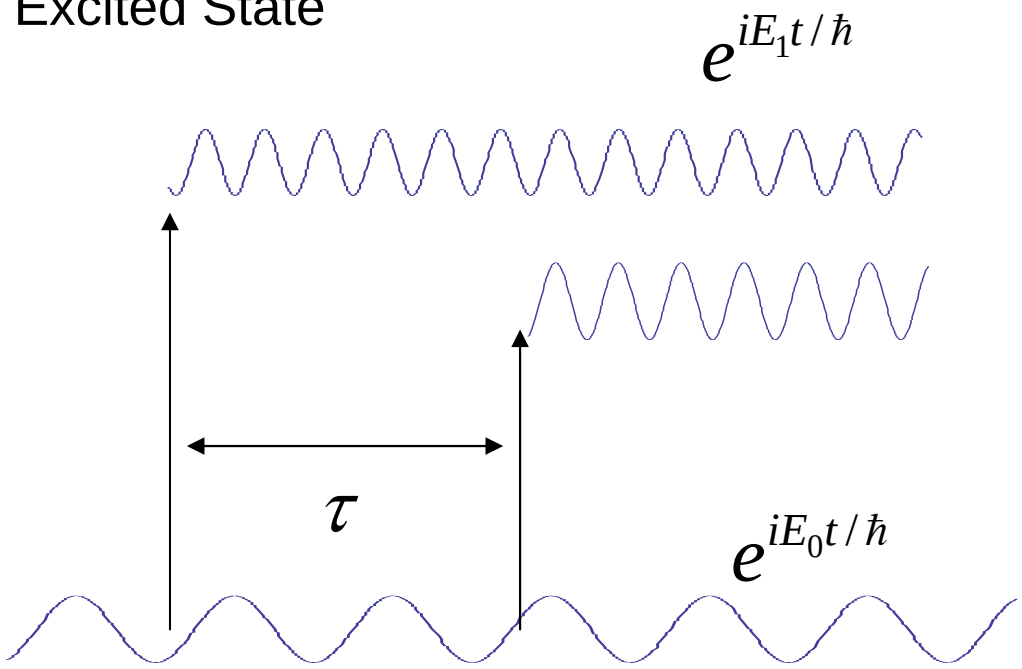
$$c_j(t) = c_j^1(t) + c_j^1(t)\exp\left[-i(\omega_{01}\tau+\delta)\right]$$

Young's Interference Experiment

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

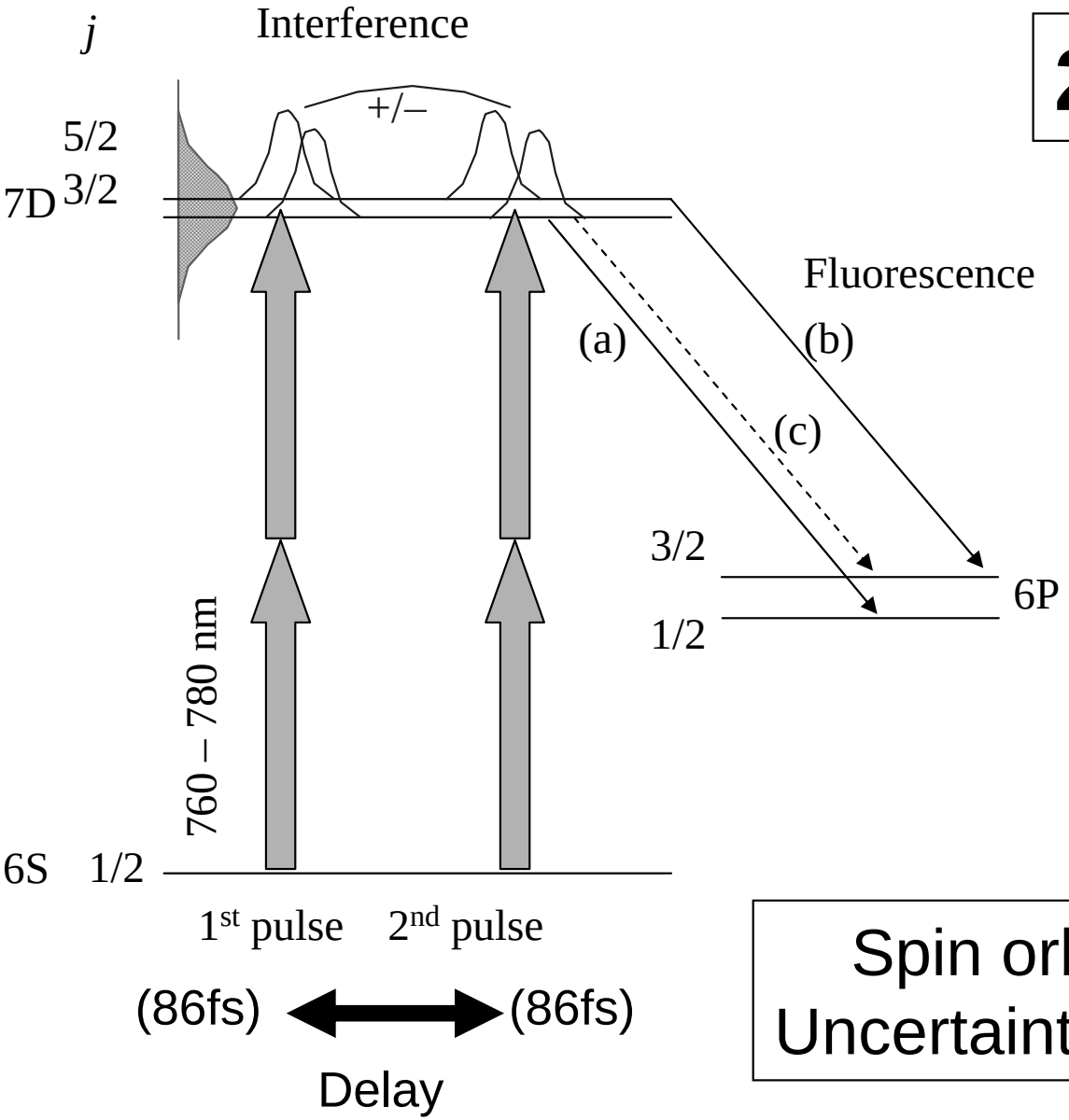


The Excited State



The Ground State

Selective Excitation of Cs atom
（Separation of Fine Structure）



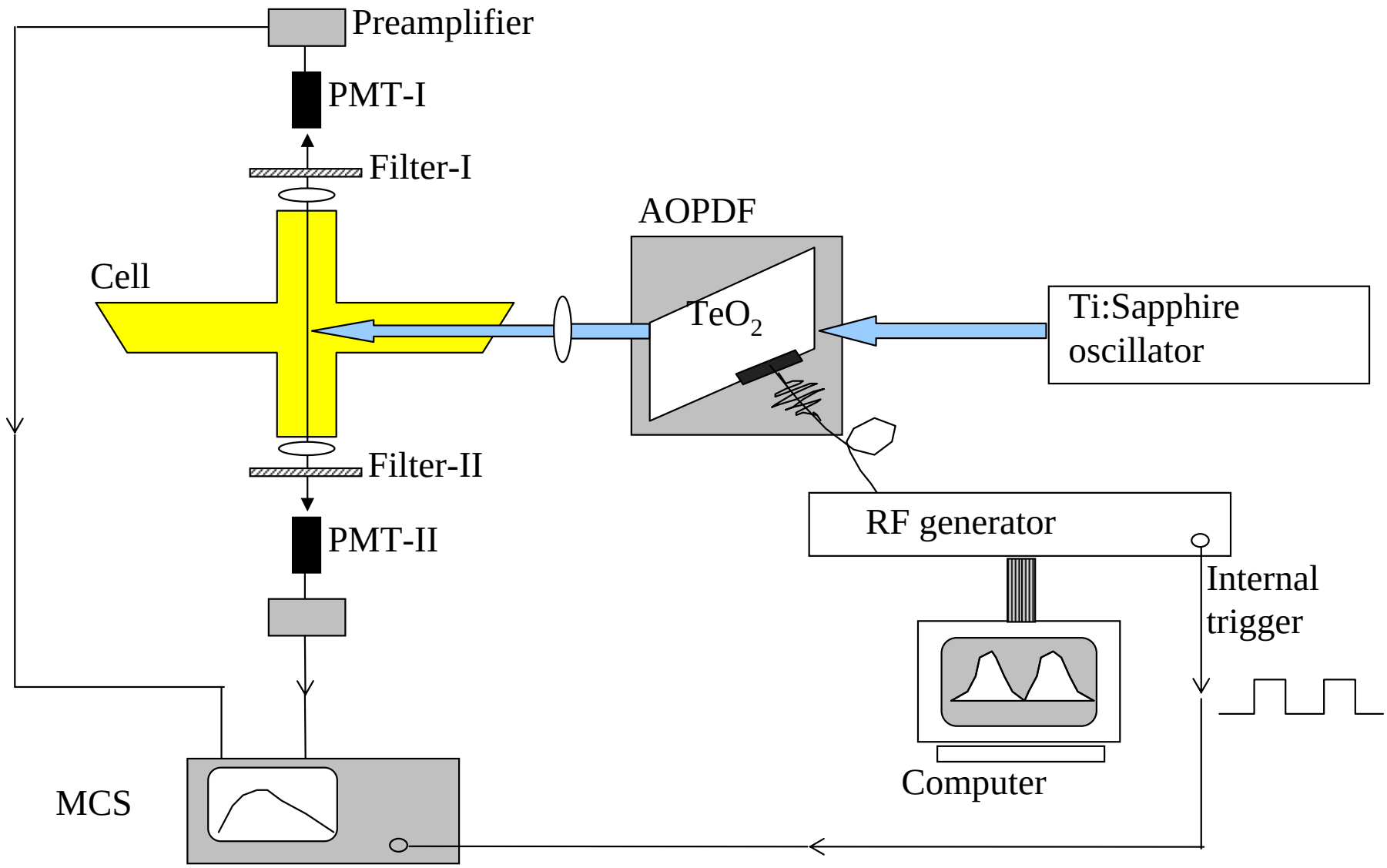
2 pulse interference

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

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

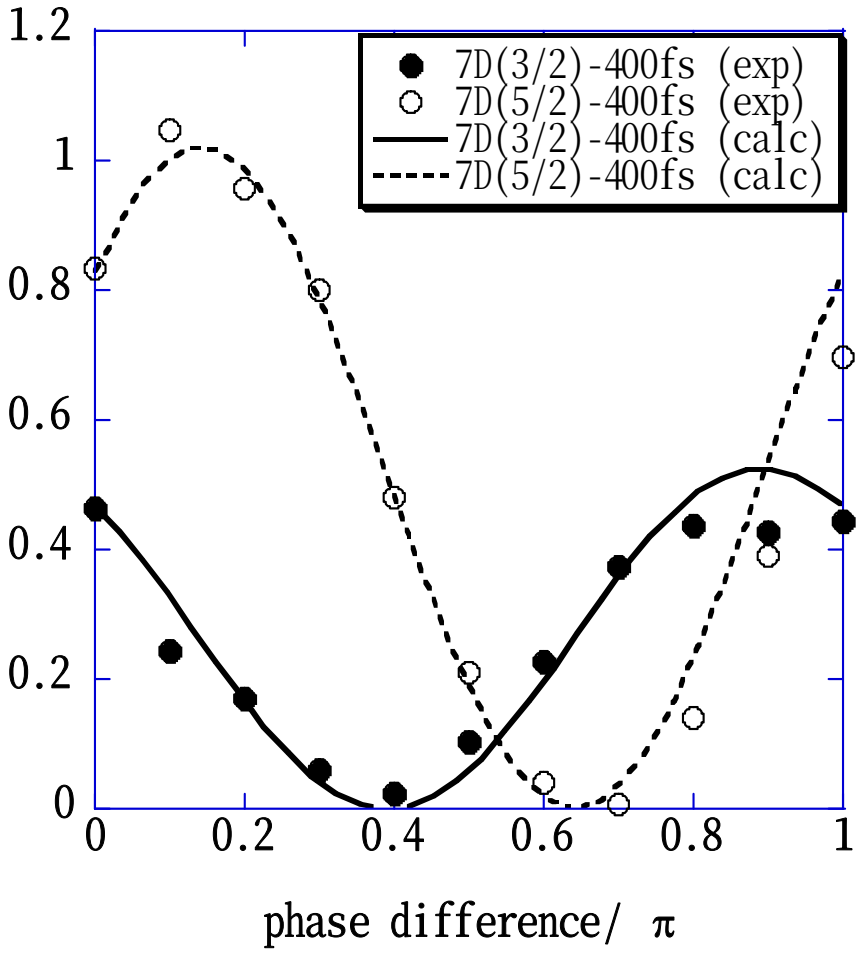
Uncertainty limit　$\Delta t = 1/\Delta E = 800\mathrm{fs}$

Experimental Facility

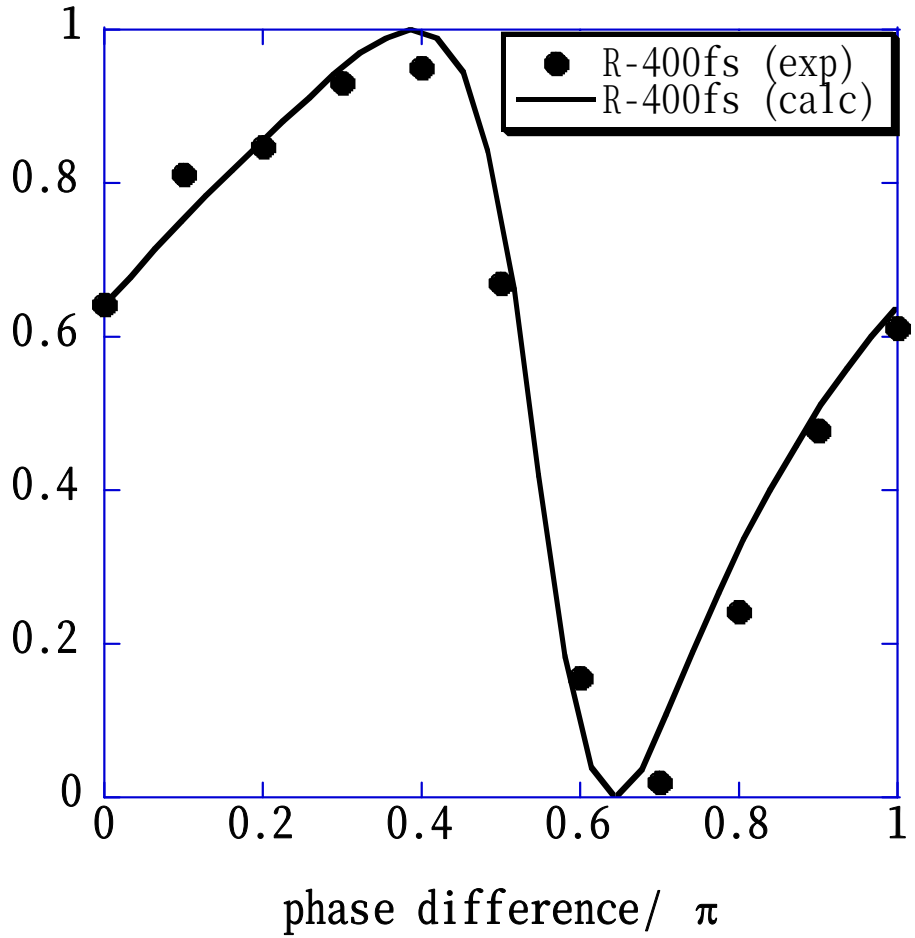


Delay: 400 f s
（Experiment and Theory）

Normalized transition probability

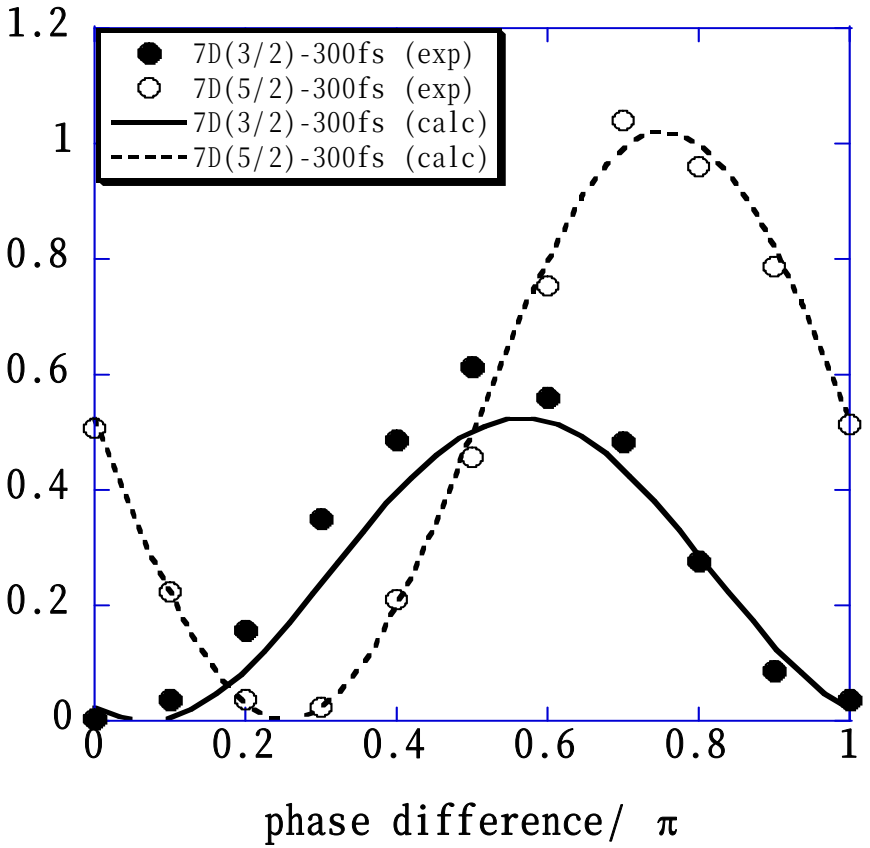


Branching ratio

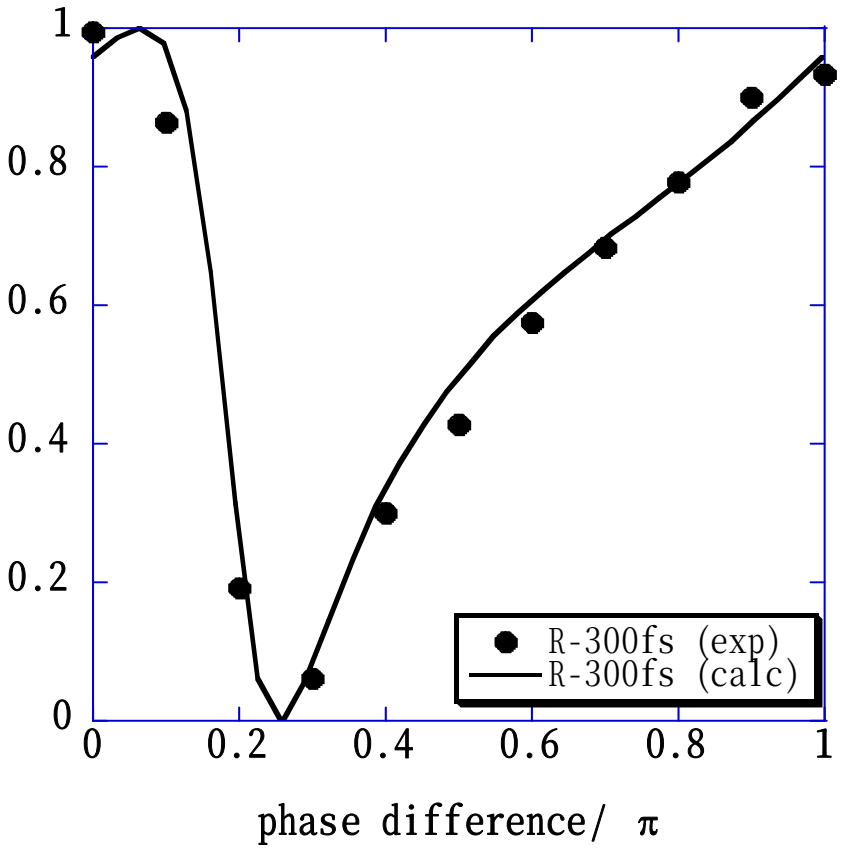


Delay 300fs
（Exp. & Theory）

Normalized transition probability

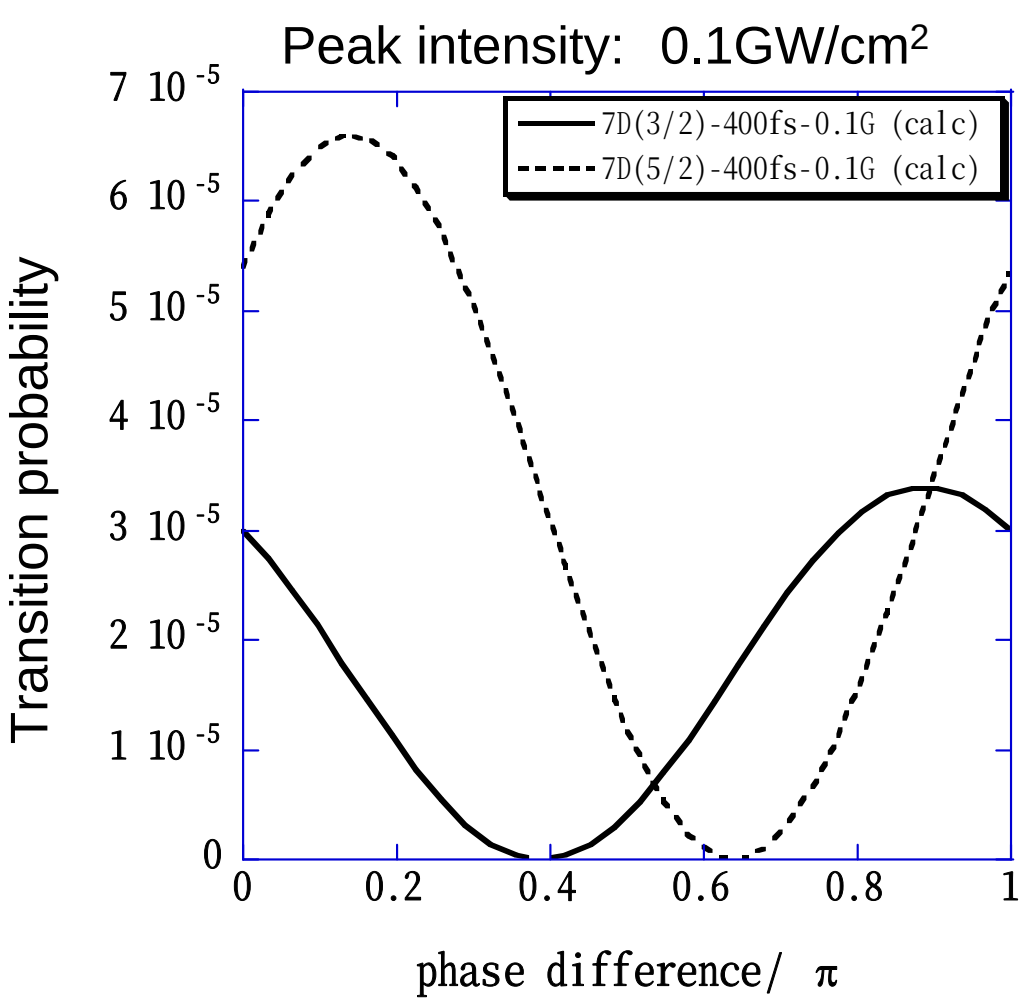


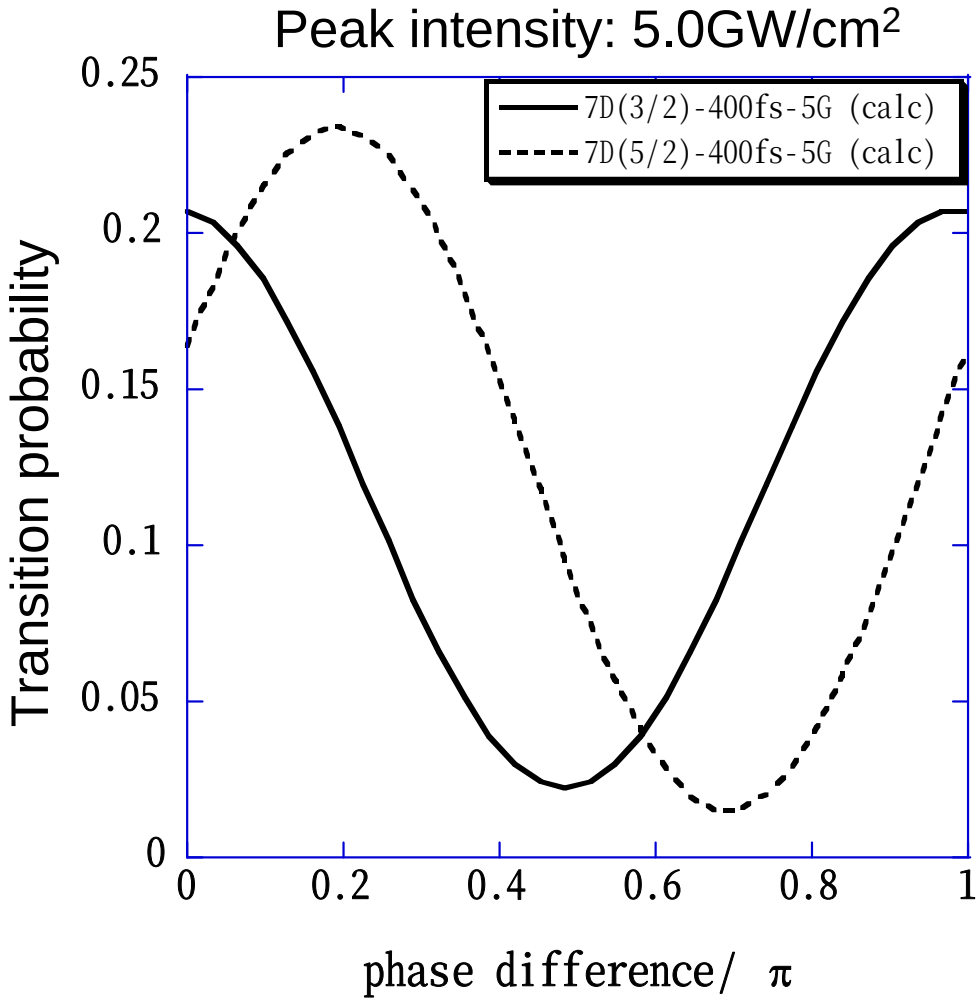
Branching ratio



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

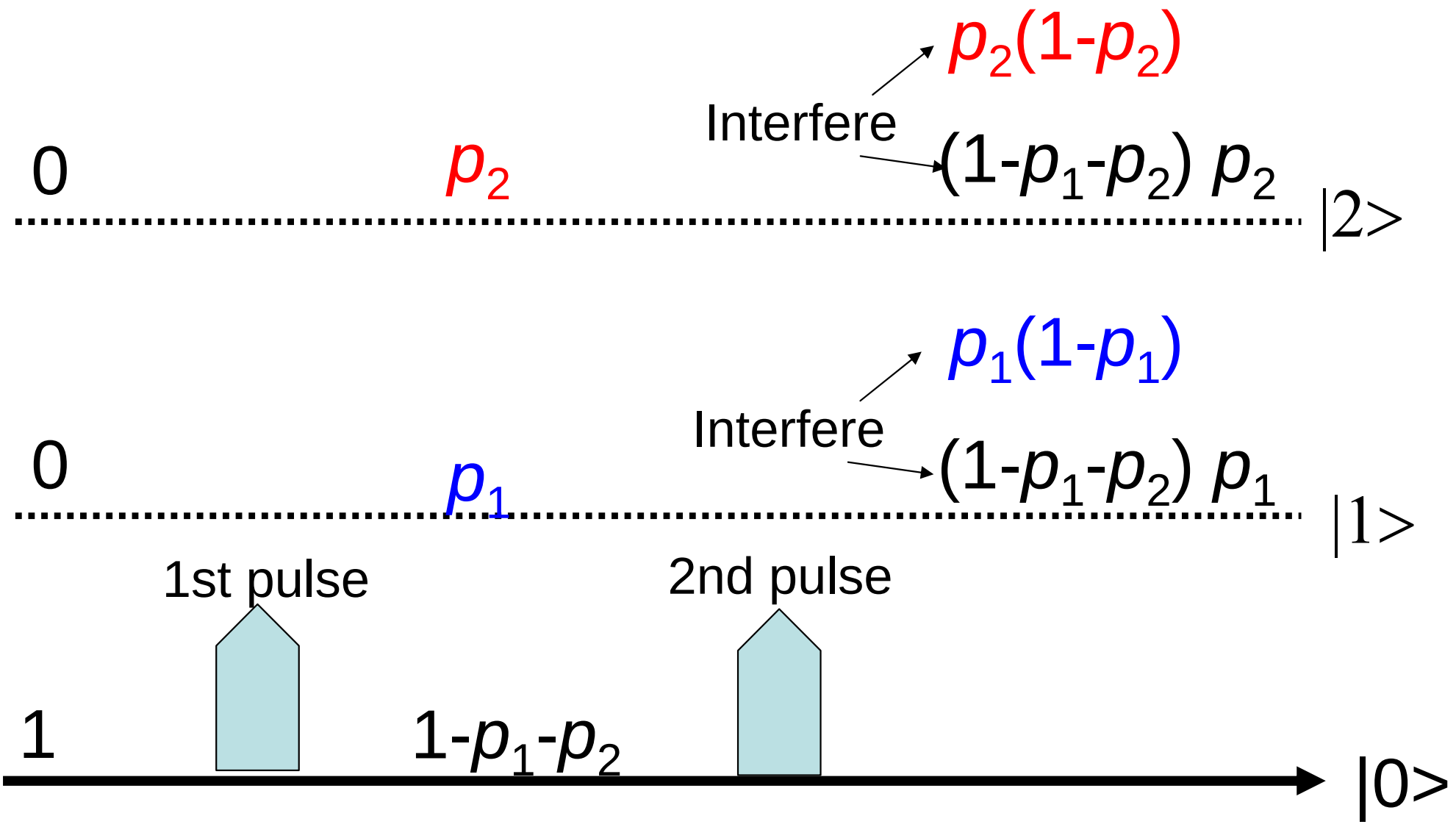
Breakdown of the Selectivity
(Theoretical simulation)





Large transition probability → bad selectivity (nonlinear effect)

Breakdown of the selectivity



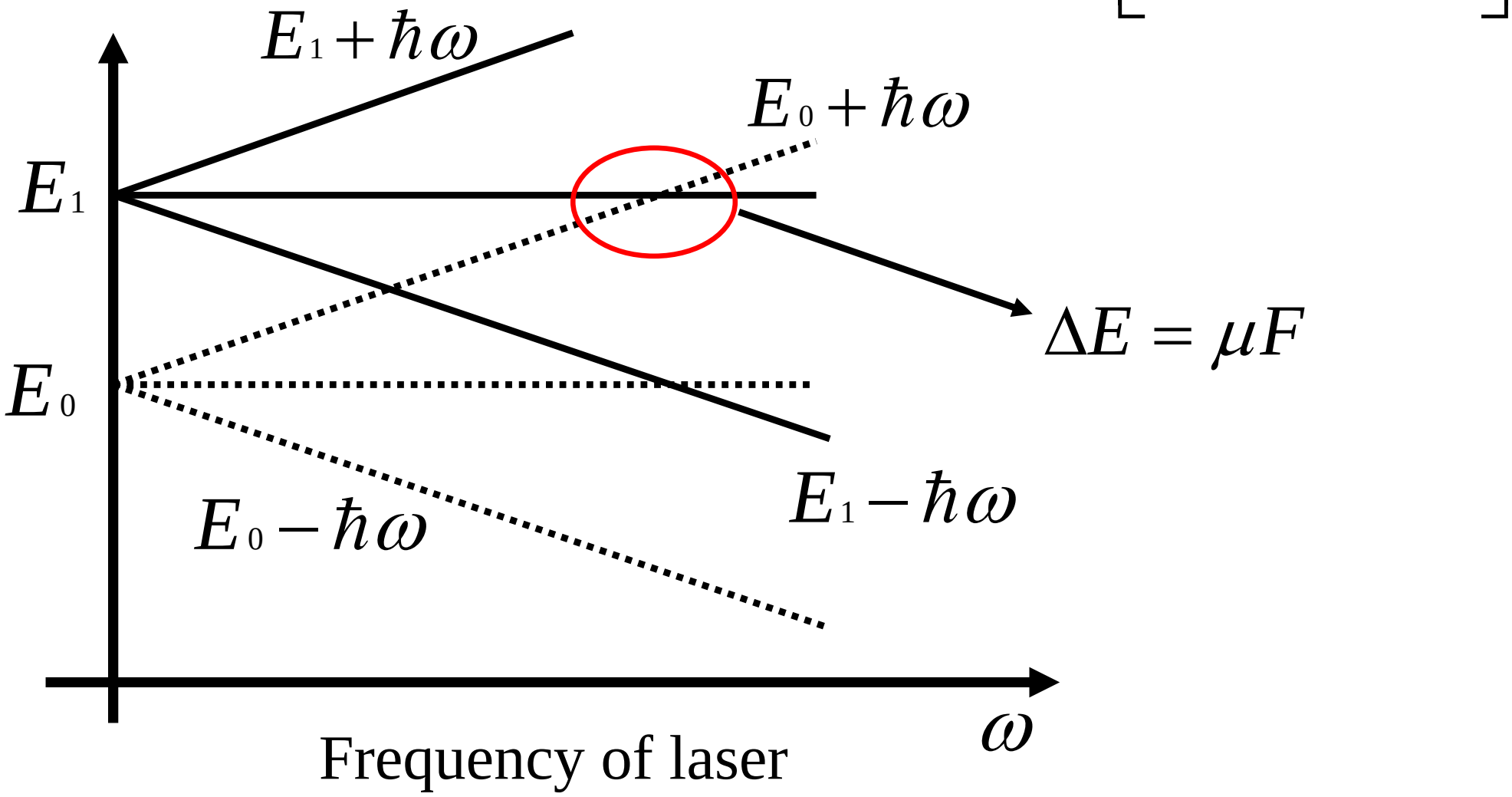
Selection → $p_1, p_2 \ll 1$ (Linearity)

Non-Perturbative Selective Excitation

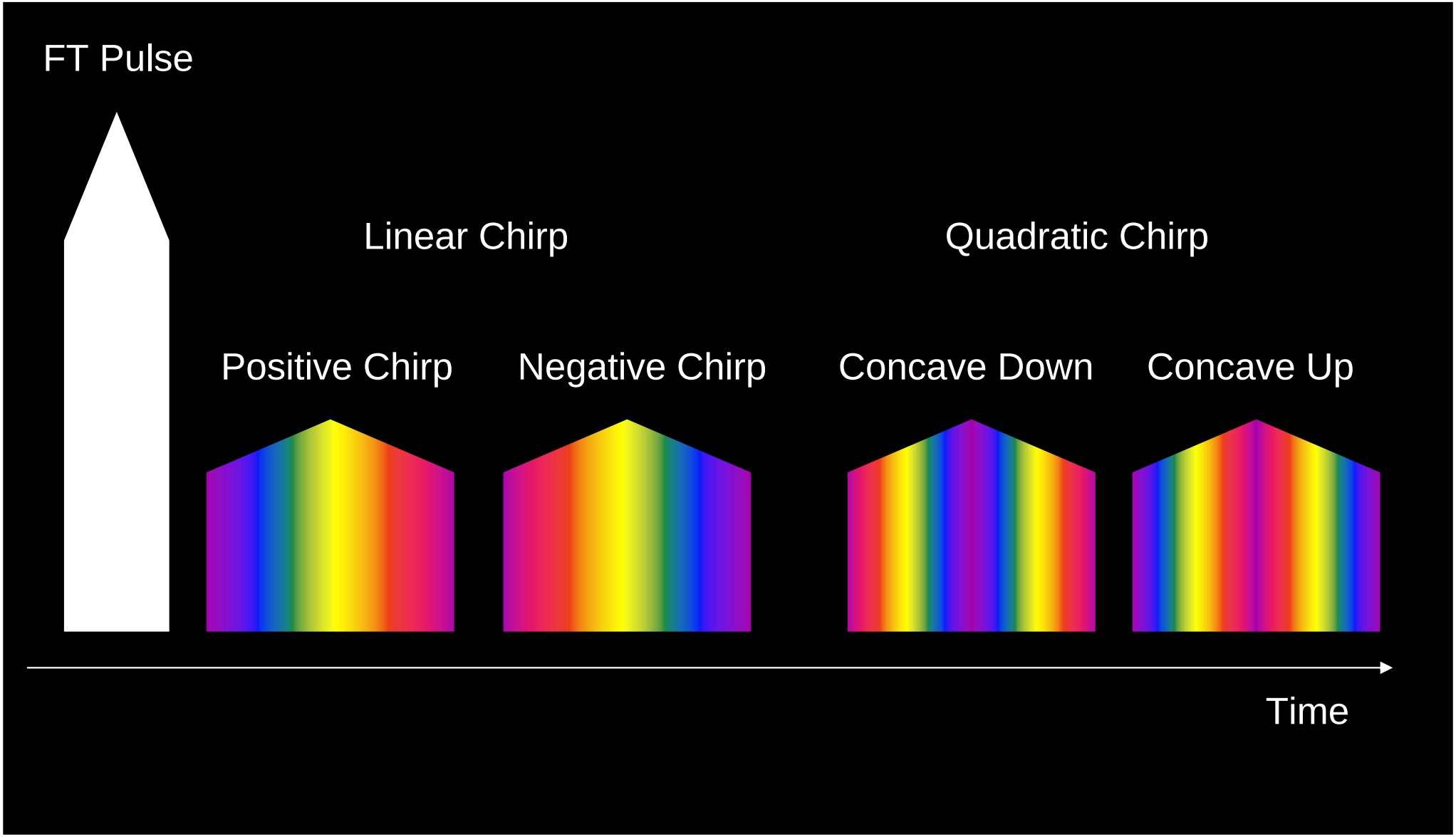
Quadratic Chirping

Quasi-state diagram

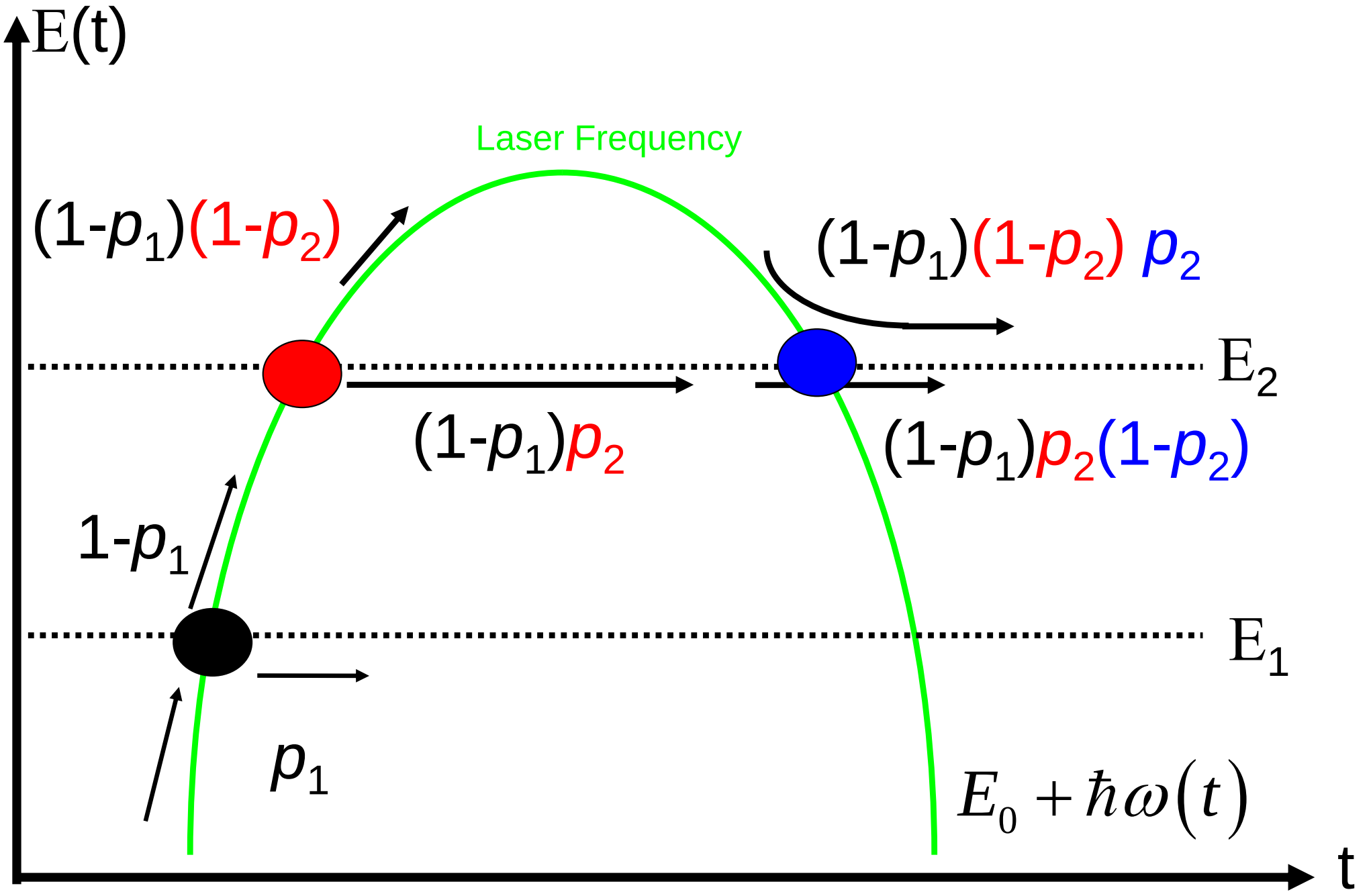
$$H' = \mu F(t)\cos\left[\int^{t} \omega(t')\,dt'\right]$$



Chirping
(time dependent frequency)

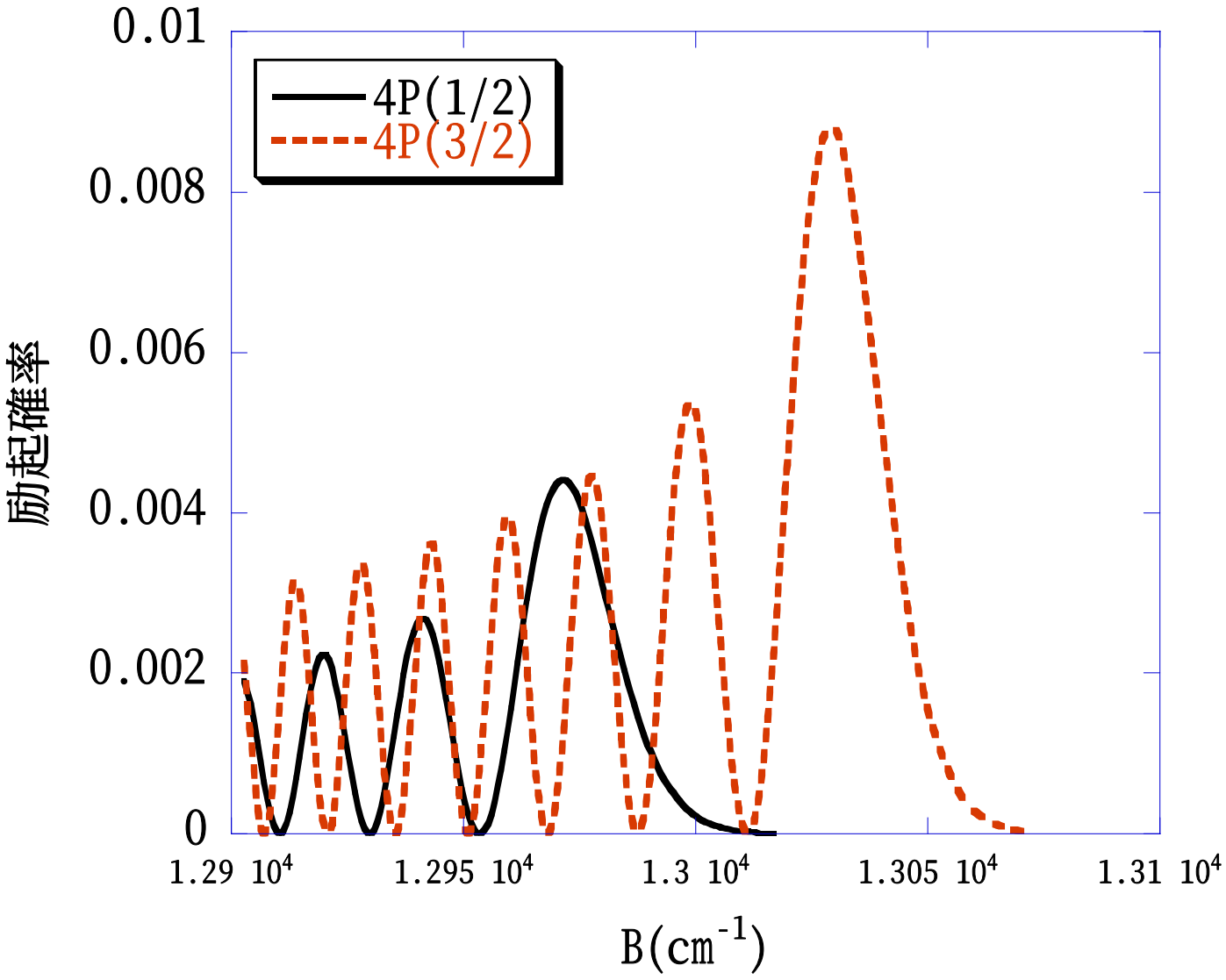


Selective Excitation by Quadratic chirping



Selective Excitaion of K atom by Quadratic chirping
(Simulation)

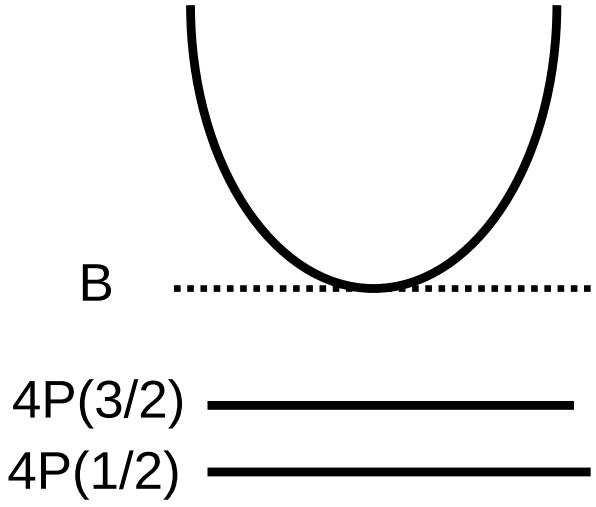
Perturbative region (1 MW/cm^2)



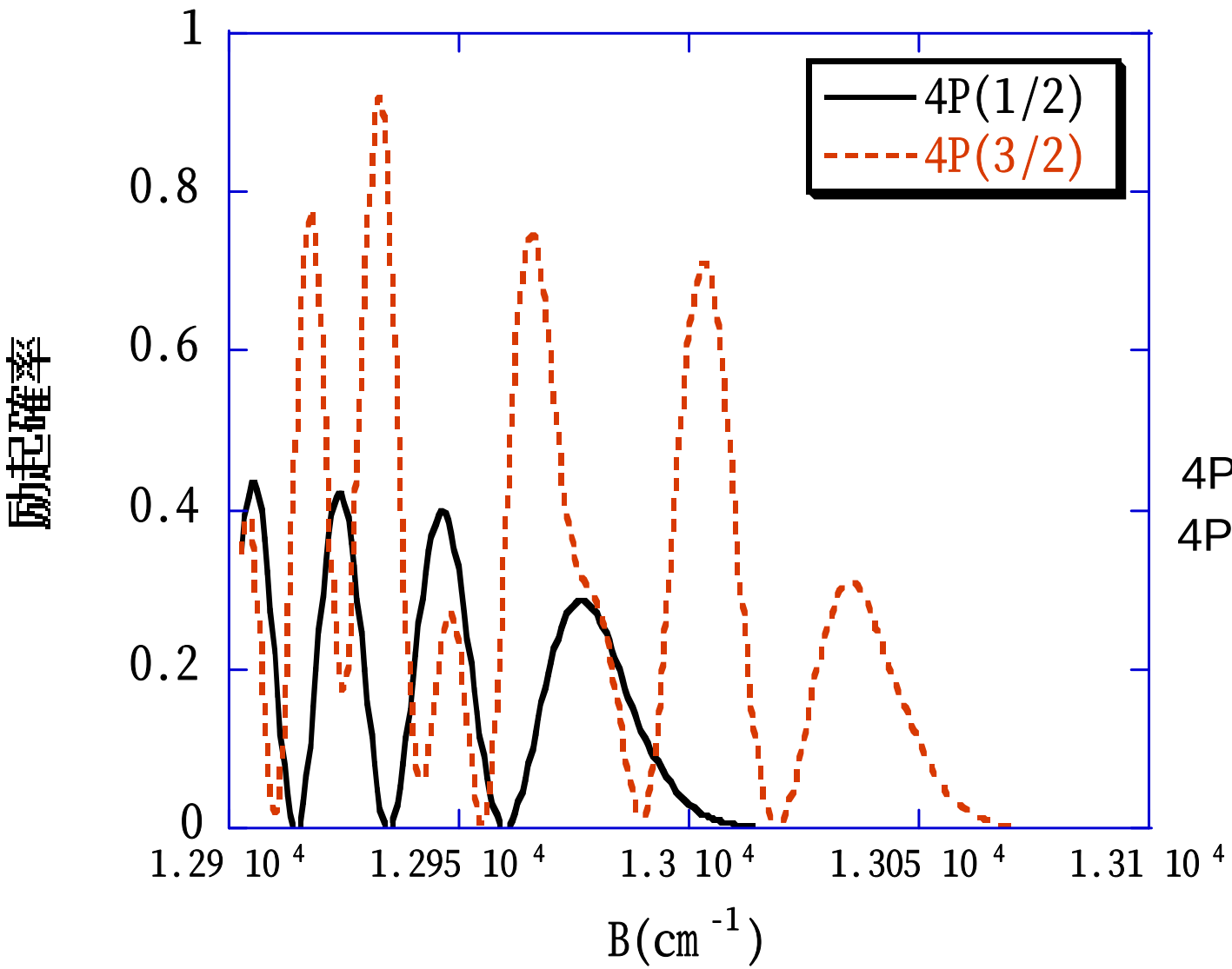
Small Probability

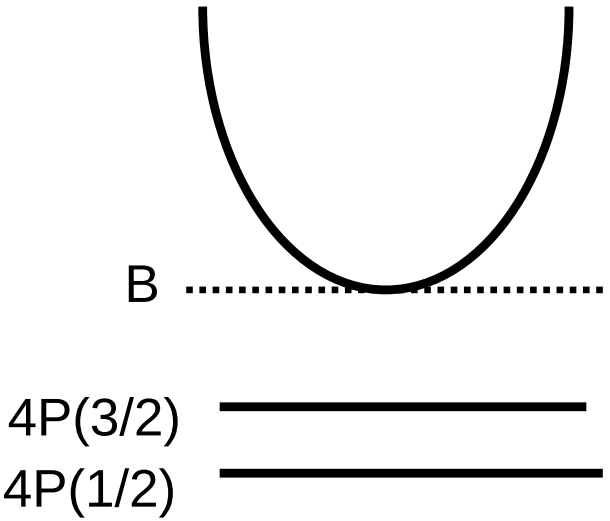
↓

Both selective



High Intensity (0.125 GW/cm^2)





Upper level (Red)

↓

Incomplete destruction

Lower level (Black)

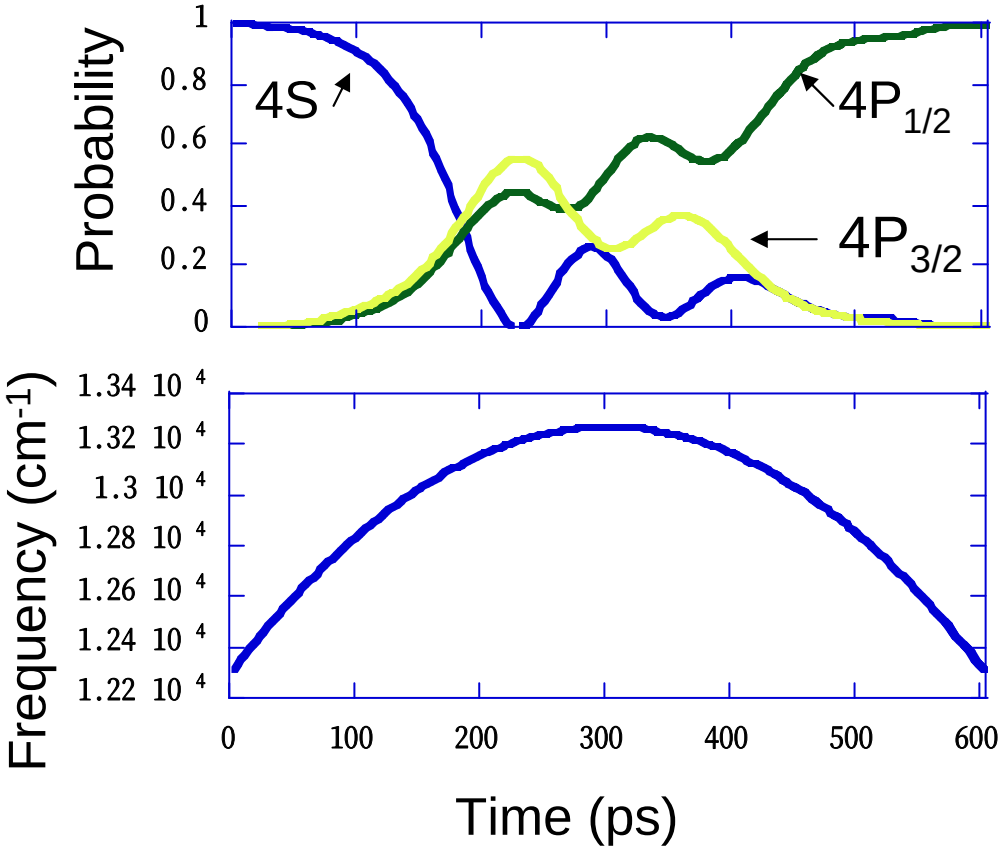
↓

Complete destruction

Complete selective excitation of K atom

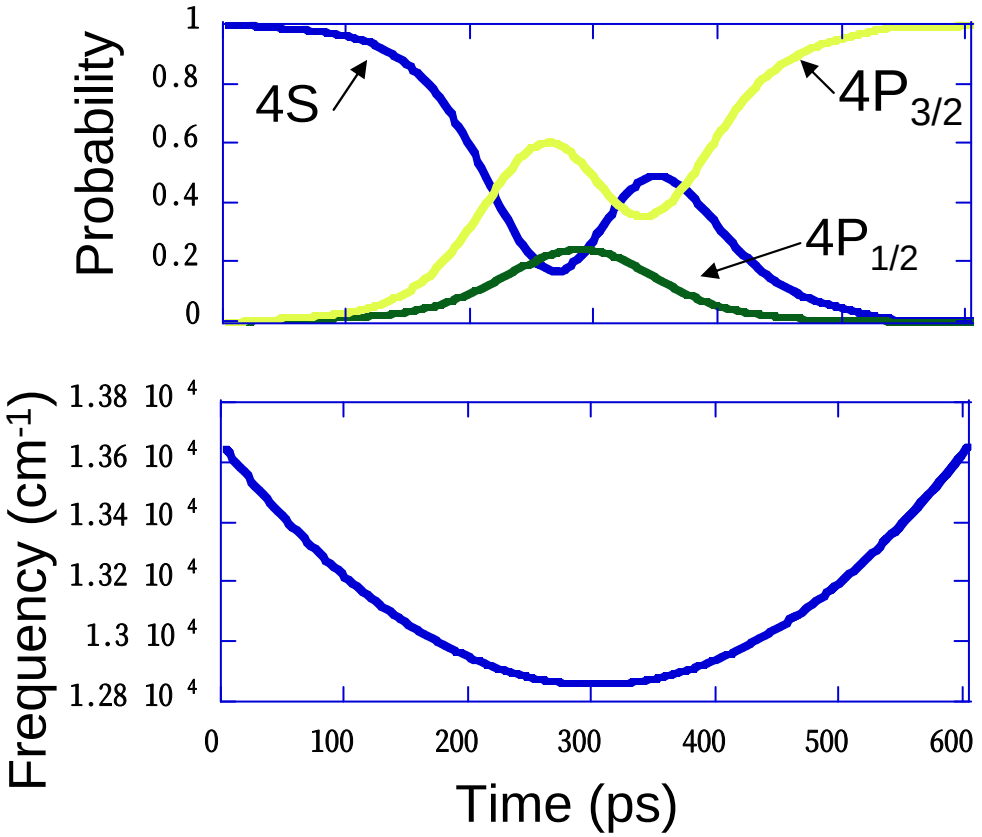
4S → $4P_{1/2}$ Excitation

Intensity 0.36 GW/cm2
Bandwidth 973 cm-1



4S → $4P_{3/2}$ Excitation

Intensity 0.125 GW/cm2
Bandwidth 803 cm-1



Complete & Selective ⇒ 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)

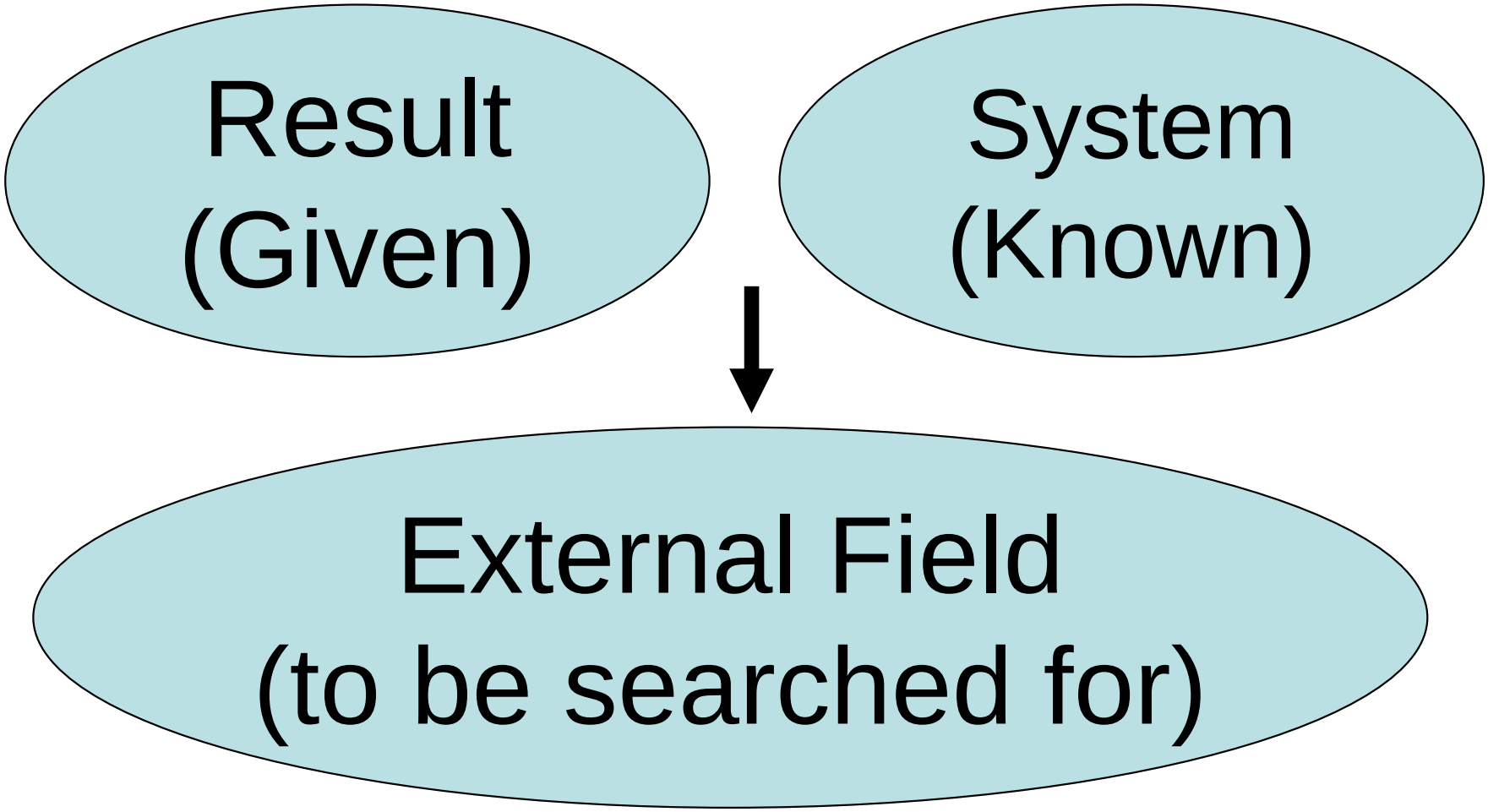
  Complete & Selective Excitation

  (As fast as the uncertainty limit)

Spectroscopy Utilizing Quantum Control

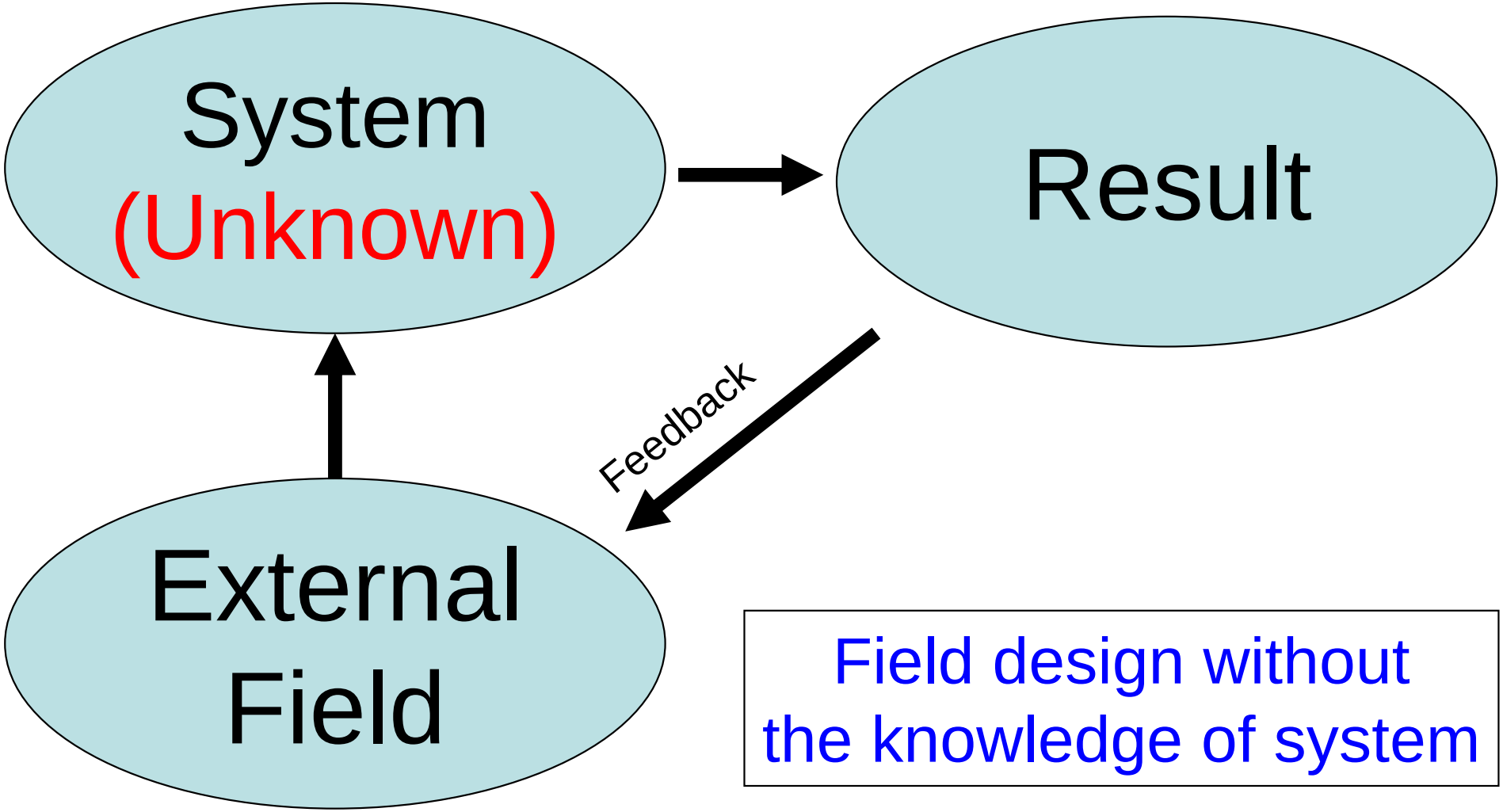
Spectroscopy for short-lived resonance states

Quantum Control



Inverse problem

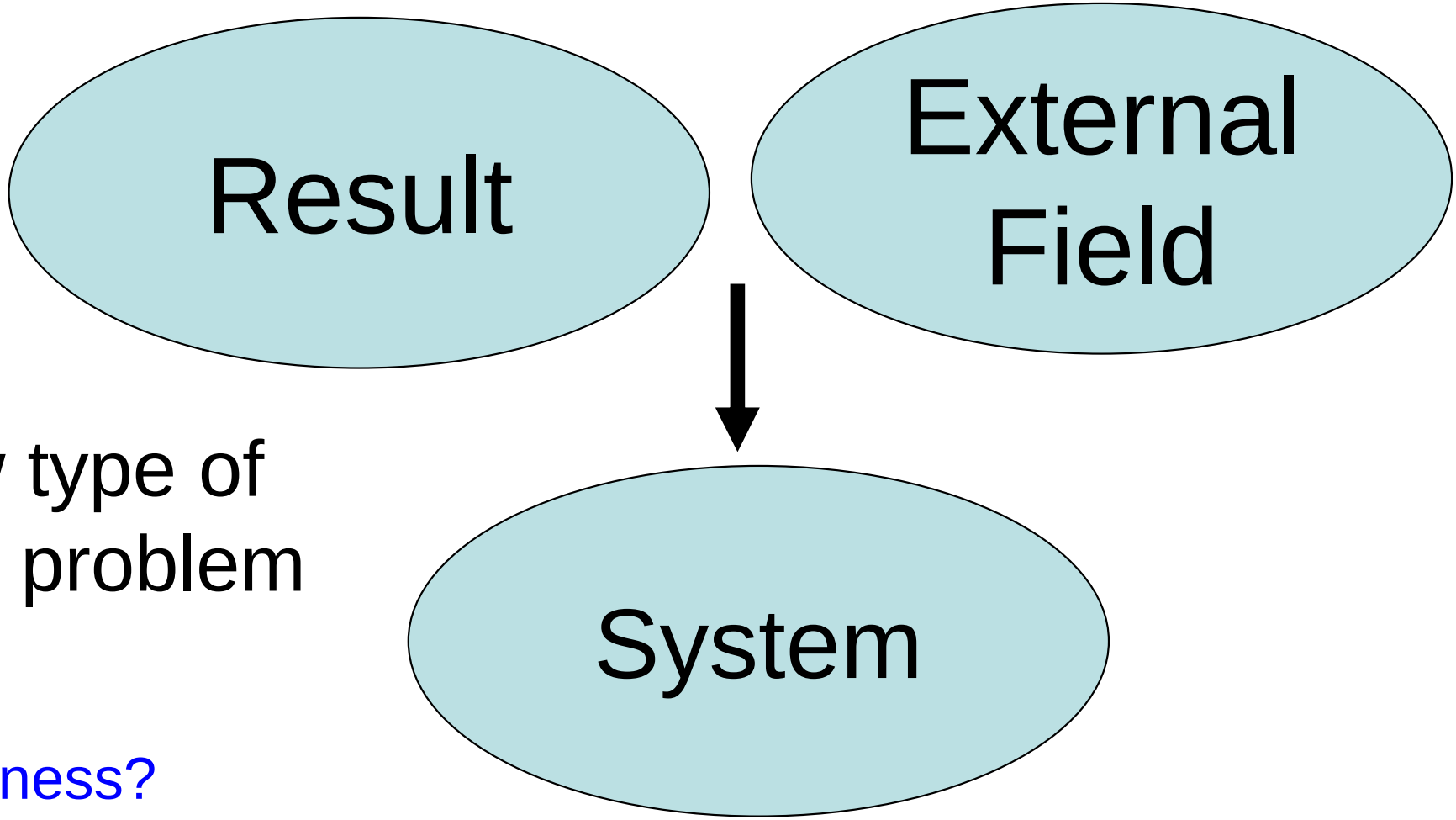
Feedback quantum control (Experiment)



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

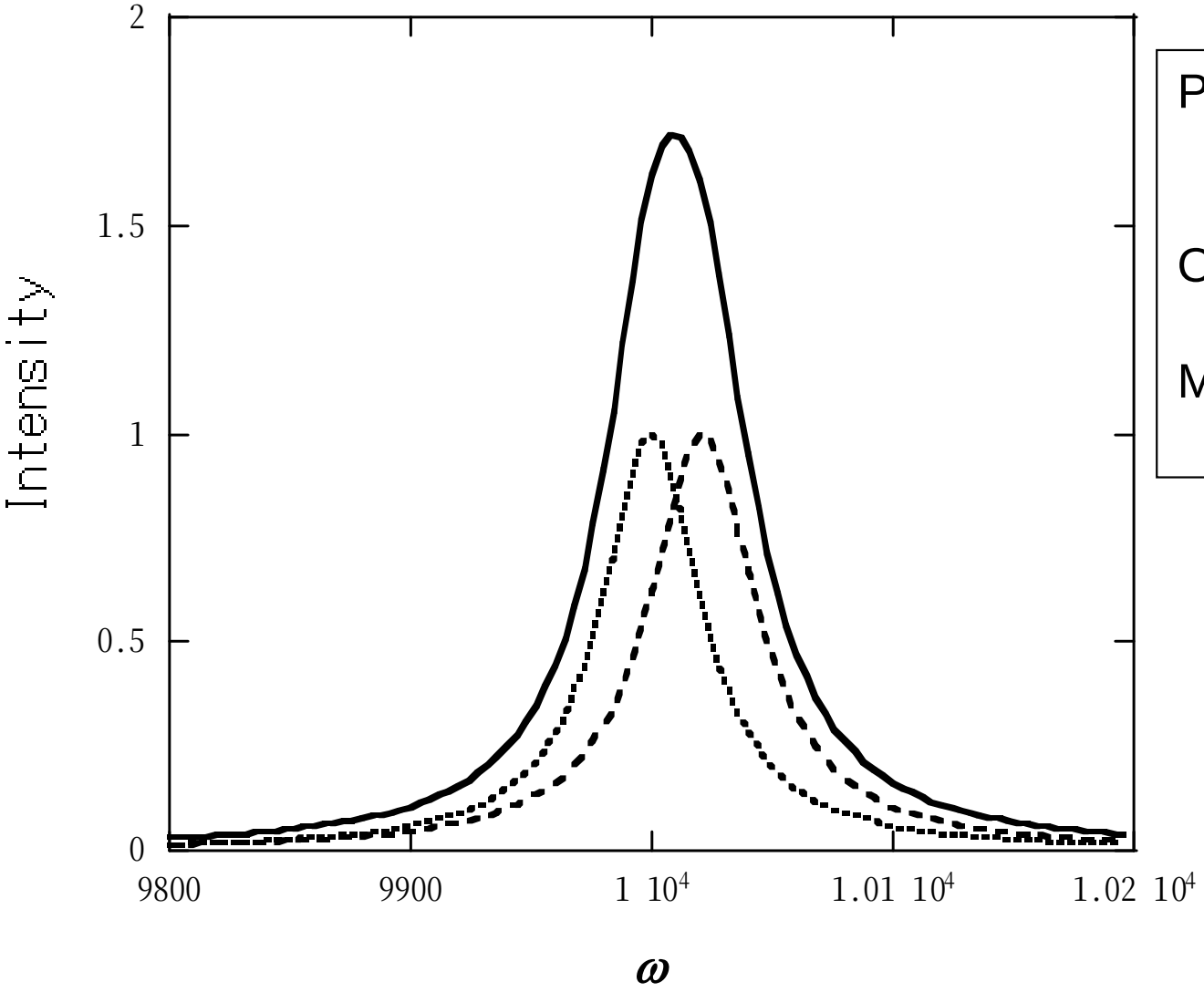
Benefits

- 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

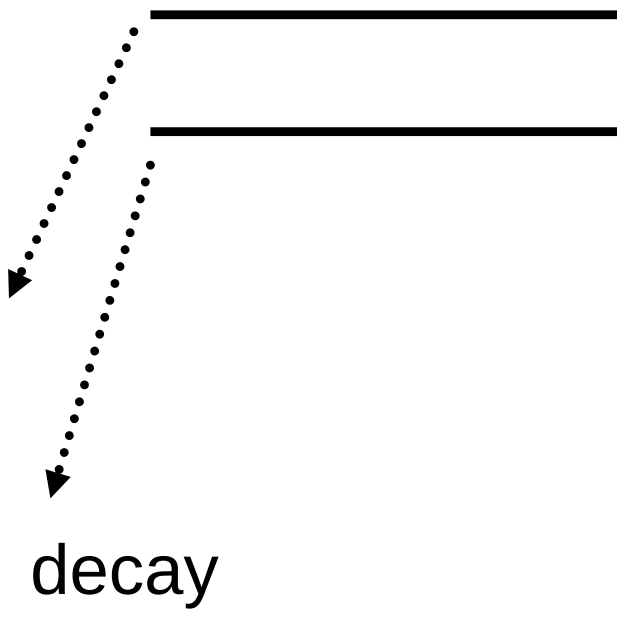
Mixture of the signals
(Solid line)

State selected signal
-> Possible?

State selective excitation

Excited states having decaying process

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



Decay process

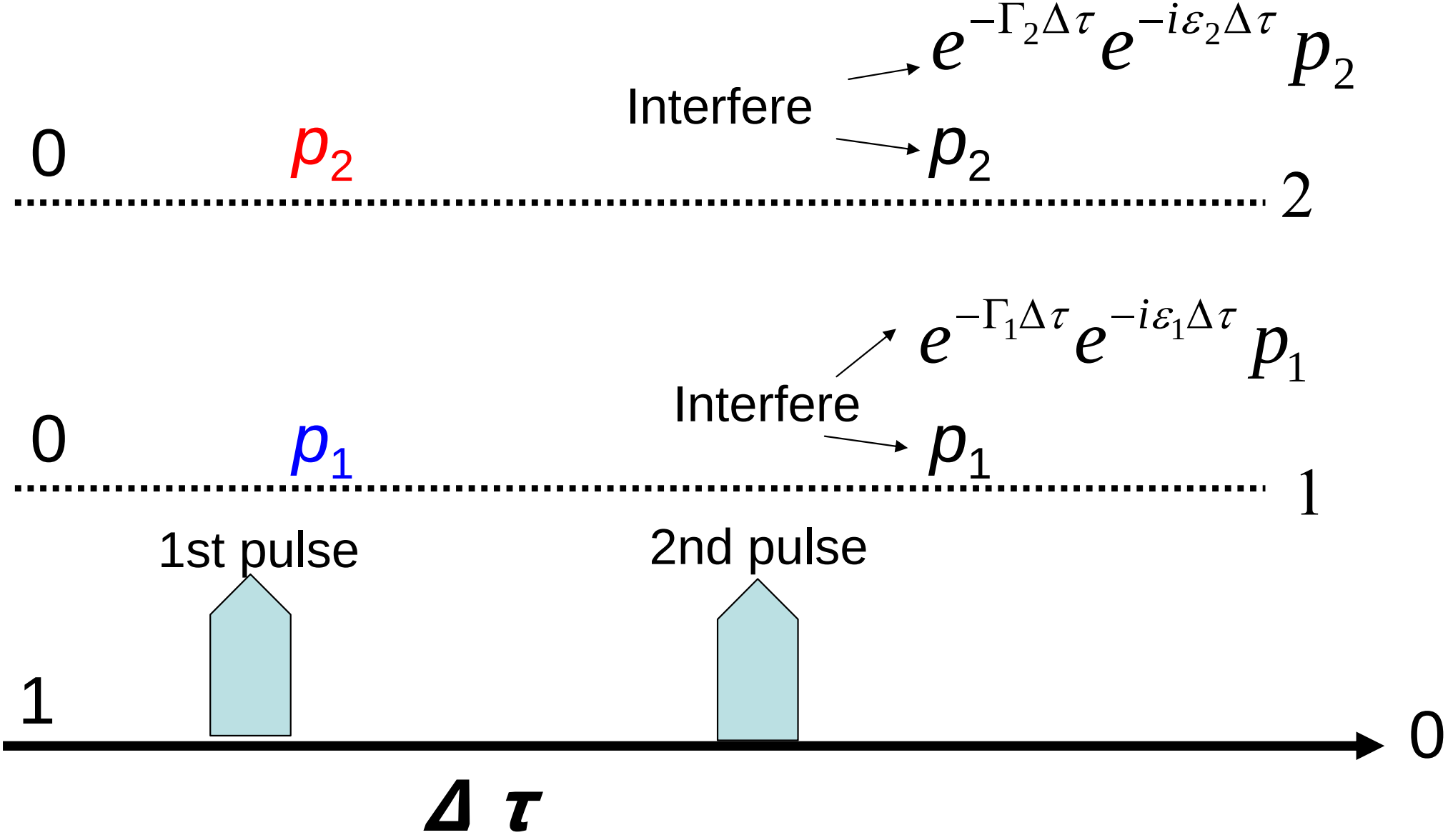
- Finite Lifetime
- Energy width (Natural width)

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

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

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)

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

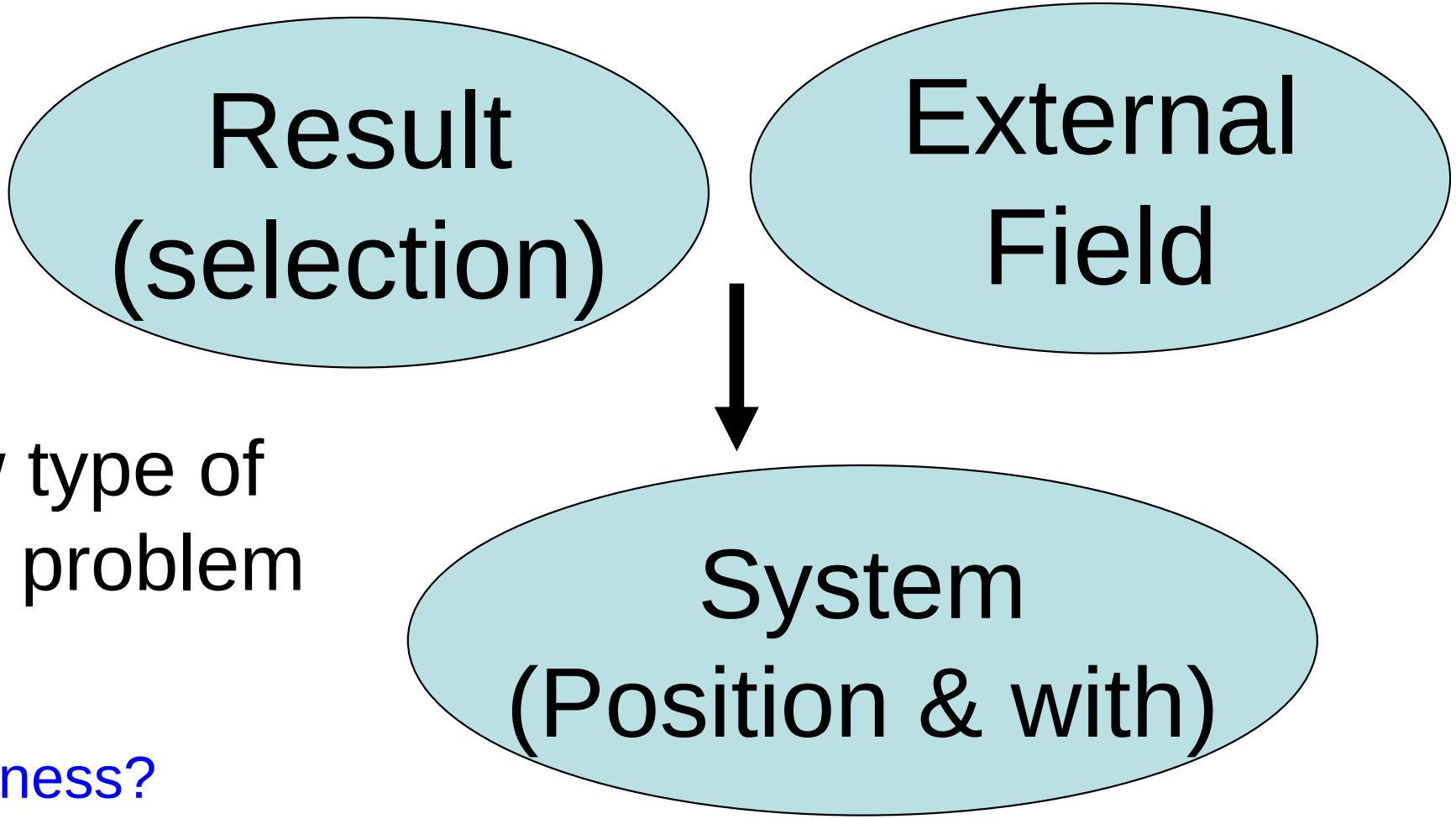
Destructive intereference
(condition for the phase)

Selection is possible even for the decaying states

Intensity ratio → Lifetime (Width)
Phase difference & Delay → Energy (Position)

Feedback spectroscopy?

System information is obtained from the optimal external field

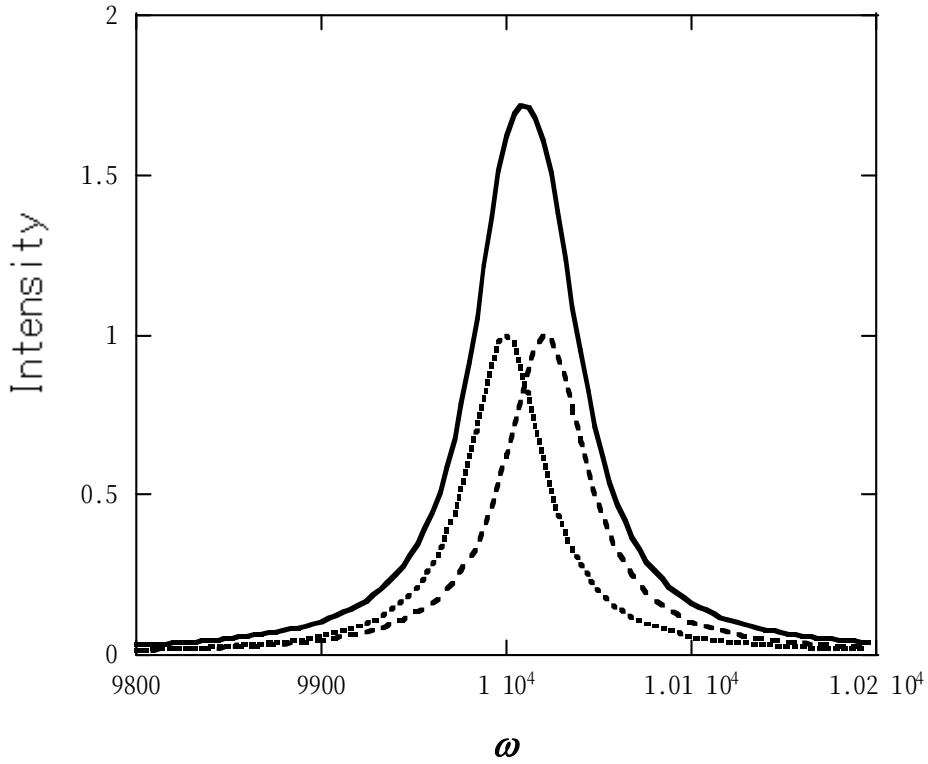


A new type of inverse problem

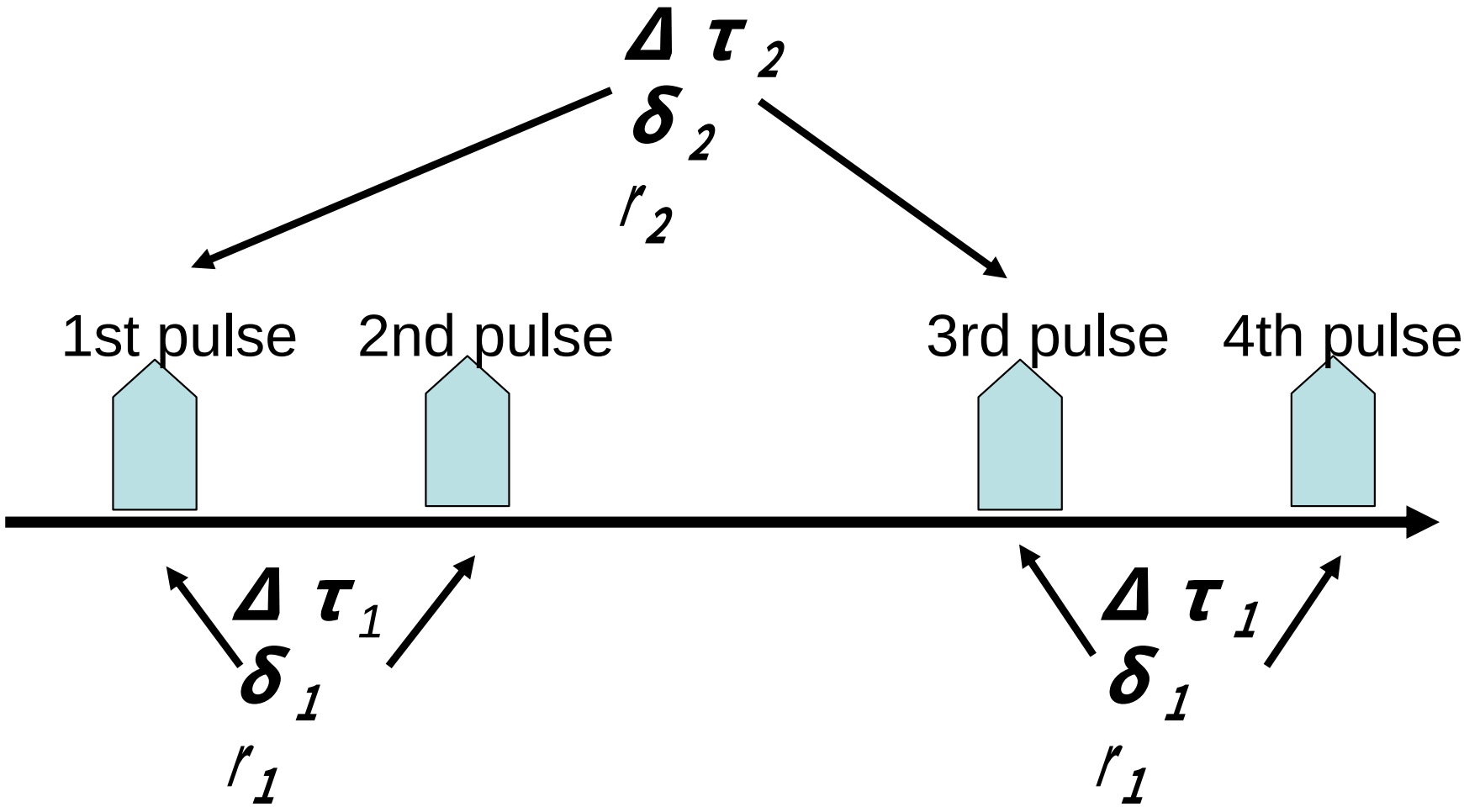
Uniqueness?

Problem

It is impossible to measure the selection ratio!



4 pulse irradiation
(Suppressing both two states)



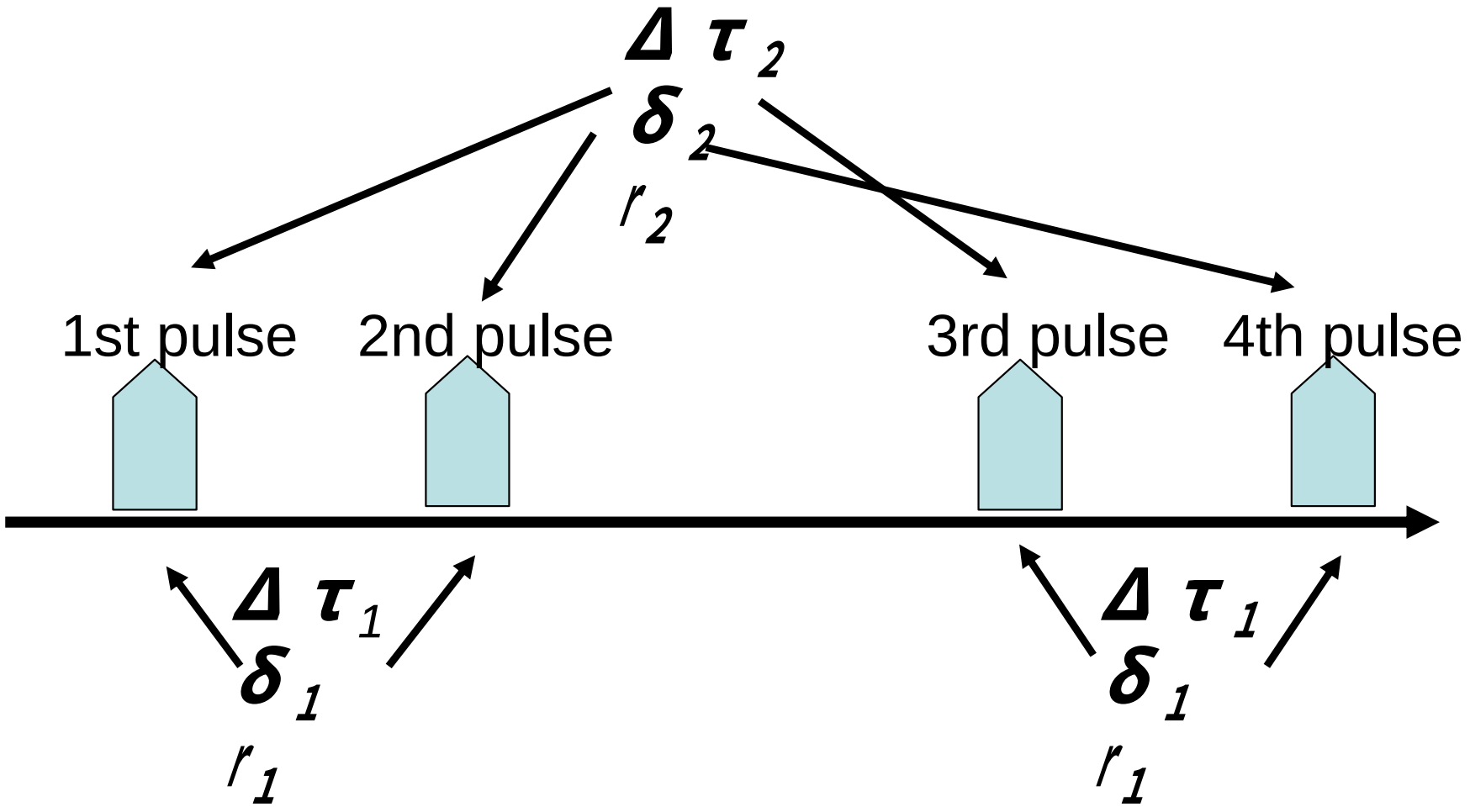
Necessary & Sufficient

Combination of pulse pairs to suppress one transition

⟷

Suppressing both states

4 pulse irradiation
(Suppressing both two states)



Necessary & Sufficient

Combination of pulse pairs to suppress one transition

⟷

Suppressing both states

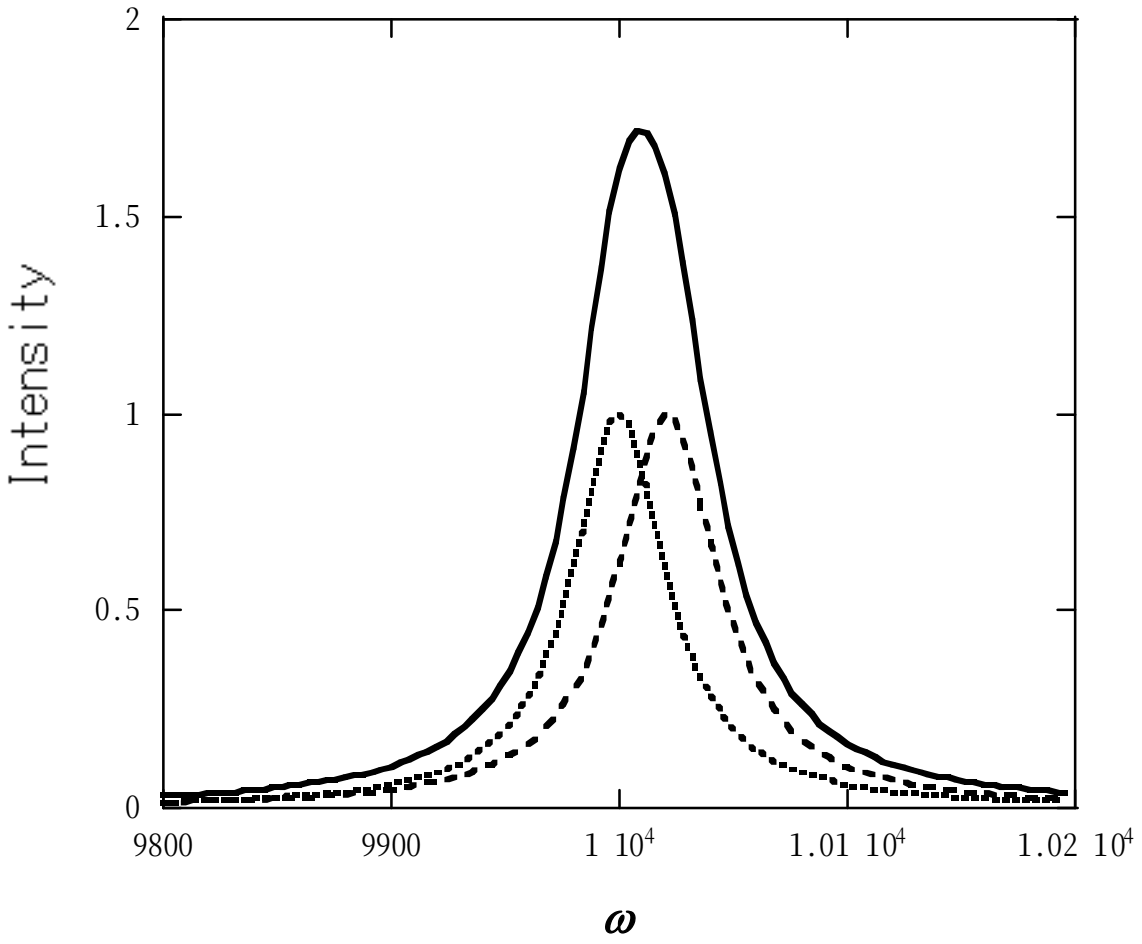
New Spectroscopy

- 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 $\rightarrow$ $r_1, r_2, \delta_1, \delta_2$

Phase differences

Intensity ratio

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

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

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

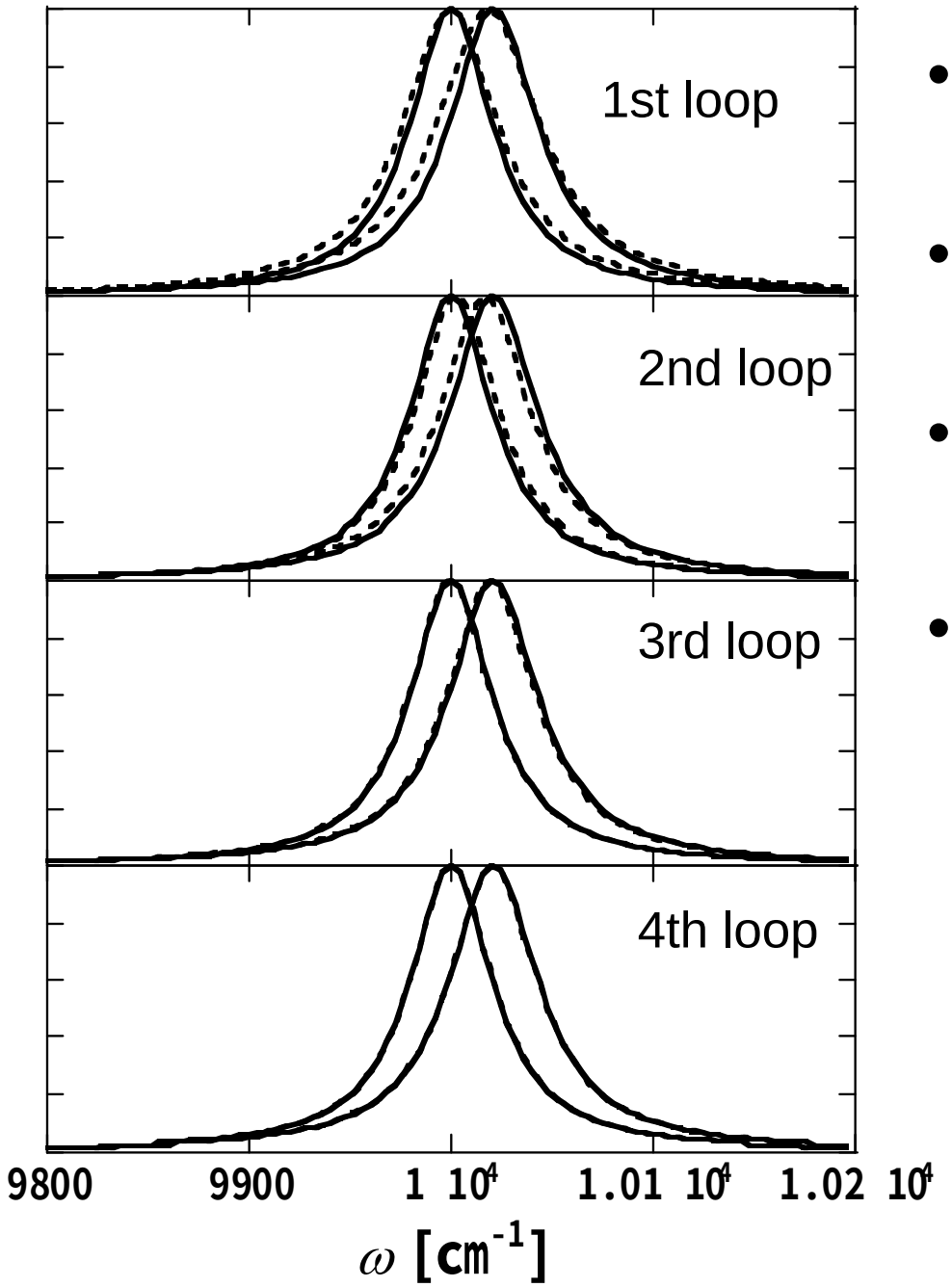
$$d = (r_1 - r_2)/2$$

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

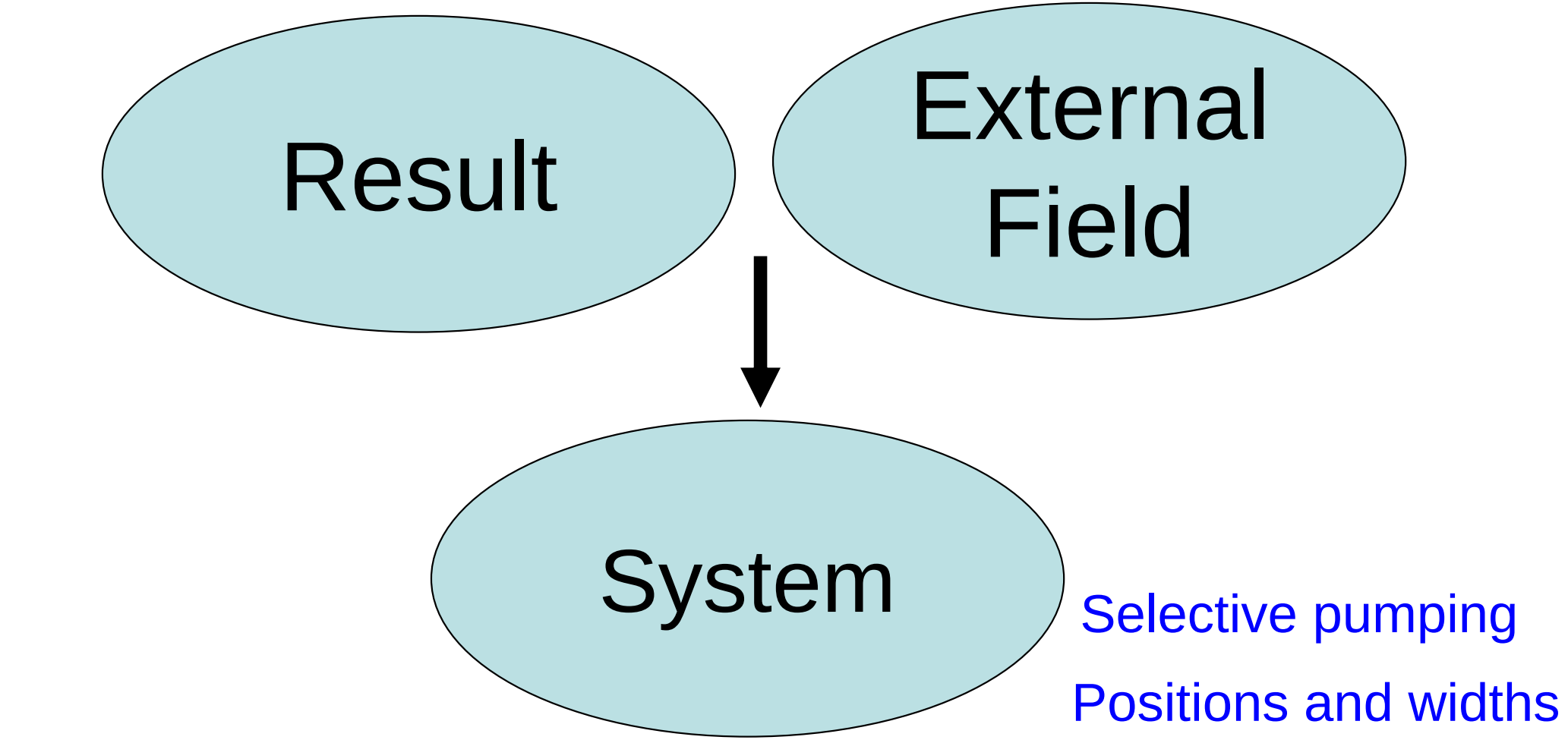
# of loop	Re(E_1)	Im(E_1)	Re(E_2)	Im(E_2)	P_1/P_2	P_2/P_1
1	9999.5	28.8385	10018.1	31.1767	0.102	0.078
2	10002.7	25.3285	10016.7	27.4977	0.0565	0.0325
3	9999.5	25.0348	10019.6	26.9731	0.00238	0.00315
4	**10000.1**	**25.0348**	**10020.7**	**26.9731**	**0.000471**	**0.000301**
Exact	**10000**	**25**	**10021**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

Results



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

Feedback spectroscopy



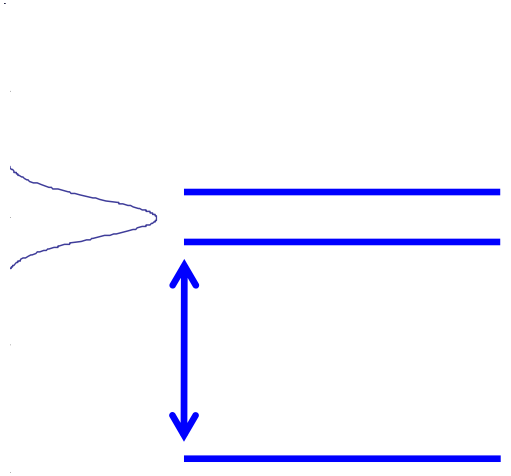
Quantum Control Spectroscopy

- 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

- Introduction to Quantum Mechanics

  Schrodinger Equation
  Stationary Problem
  Time-Dependent Problem



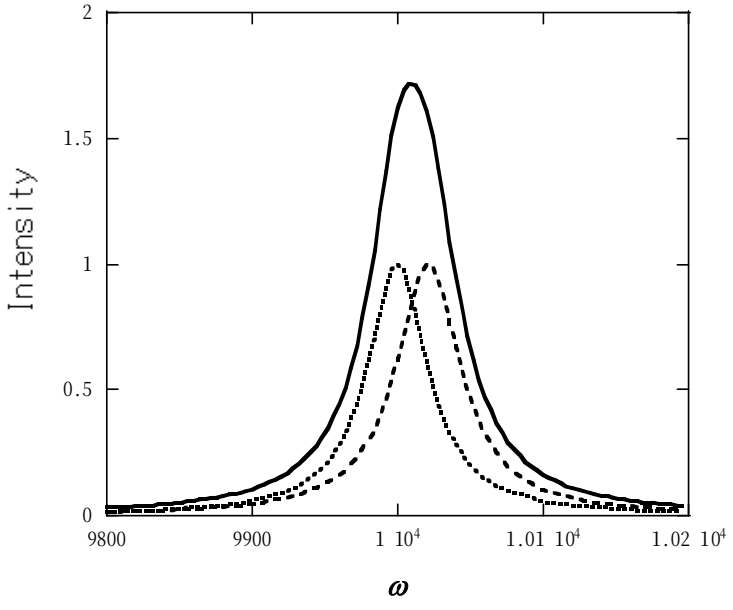
Quantum Control

- Ultrafast Selective Excitation

  A pair of weak pulses
  Quadratic Chirping

- Quantum Control Spectroscopy
  (overlapping by natural width)

- Molecular computer
  (Classical computer)



nature Vol 465|13 May 2010

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)