
Time-Dependent Schrödinger Equation

- Classical electron in a laser field
- Time-dependent Schrödinger equation: the basics
- Numerical approaches to TDSE
- Strong field S-matrix and the Strong field approximation

I will use atomic units, where $m=e=\hbar=1$

Classical electron in a laser field

$$\mathbf{E}(t) = E \cos \omega t$$


$\frac{d\mathbf{v}}{dt} = -\mathbf{E}(t)$

Define $\mathbf{A}(t)$: $\mathbf{E}(t) = -\partial\mathbf{A}/\partial t = -d\mathbf{A}/dt$ — $\mathbf{A}(t)$ does not depend on x (the dipole approximation)

$$\frac{d\mathbf{v}}{dt} = -\mathbf{E}(t) = d\mathbf{A}/dt \quad d(\mathbf{v} - \mathbf{A})/dt = 0$$

Then

$$\mathbf{v}(t) - \mathbf{A}(t) = \mathbf{P} = \text{const} \quad \text{and} \quad \mathbf{v}(t) = \mathbf{P} + \mathbf{A}(t)$$

\mathbf{P} is canonical momentum, different from the kinetic momentum $\mathbf{p} = m\mathbf{v} = \mathbf{v}$

Classical electron in a laser field

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$\frac{d\mathbf{v}}{dt} = -\mathbf{E}(t)$

$$\mathbf{v}(t) - \mathbf{A}(t) = \mathbf{P} = \text{const} \quad \text{and} \quad \mathbf{v}(t) = \mathbf{P} + \mathbf{A}(t)$$

\mathbf{P} is canonical momentum, different from the kinetic momentum $\mathbf{p} = m\mathbf{v} = \mathbf{v}$

$$H = [\mathbf{P} + \mathbf{A}(t)]^2 / 2$$

yields correct equations:

$$\mathbf{v} = \partial H / \partial \mathbf{P} = \mathbf{P} + \mathbf{A}(t)$$

$$d\mathbf{P}/dt = -\partial H / \partial \mathbf{x} = 0$$

Classical electron in a laser field

$$\mathbf{E}(t) = \mathbf{E} \cos \omega t$$


$\frac{d\mathbf{v}}{dt} = -\mathbf{E}(t)$

$H = [\mathbf{P} + \mathbf{A}(t)]^2 / 2$ yields correct Newton equations.

But:

$$H = \mathbf{p}^2 / 2 + \mathbf{x} \cdot \mathbf{E}$$

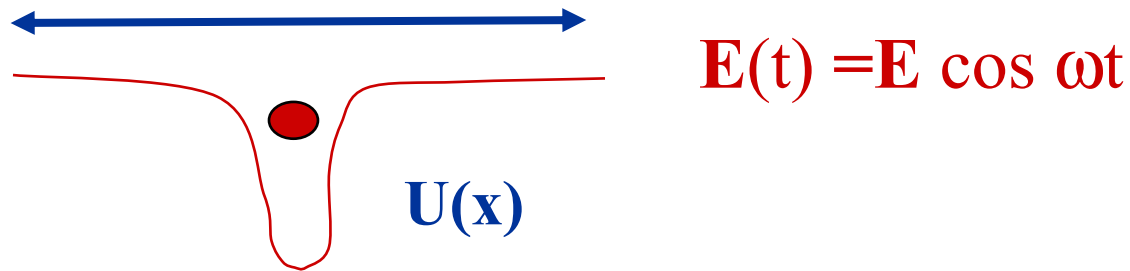
also yields the Newton equation:

$$\begin{aligned} \mathbf{v} = d\mathbf{x}/dt &= \partial \mathbf{H} / \partial \mathbf{p} = \mathbf{p} \\ d\mathbf{p}/dt &= -\partial \mathbf{H} / \partial \mathbf{x} = -\mathbf{E}(t) \end{aligned} \quad \Rightarrow \quad \frac{d\mathbf{v}}{dt} = -\mathbf{E}(t)$$

You can change H without changing the Newton eqs. of motion.

Such modifications are called ‘Gauge transformations’

The two gauges



$$H_{pA} = \frac{1}{2} [\mathbf{P} + \mathbf{A}(t)]^2 + U(\mathbf{x})$$

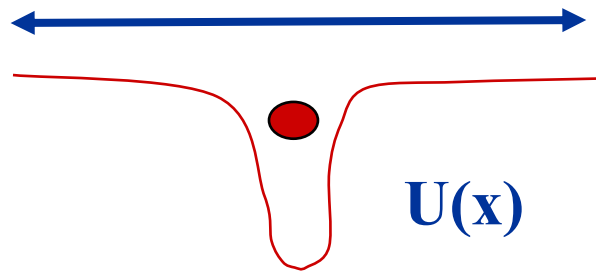
$$H_{dE} = \frac{\mathbf{p}^2}{2} + U(\mathbf{x}) + \mathbf{xE}(t)$$

Both \mathbf{P} and \mathbf{p} are momenta, but they have completely different physical meaning

\mathbf{xE} is the interaction of the electron with the field in a.u.:

$$H_{dE} = \frac{\mathbf{p}^2}{2} + U(\mathbf{x}) + \mathbf{xE}(t) = \frac{\mathbf{p}^2}{2} + U(\mathbf{x}) - \mathbf{exE}(t) = \frac{\mathbf{p}^2}{2} + U(\mathbf{x}) - \mathbf{dE}(t)$$

The TDSE and the two gauges



$$\mathbf{E}(t) = \mathbf{E} \cos \omega t$$

$$i\partial_t \Psi = H\Psi$$

$$\hat{H}_{pA} = \frac{1}{2} [\hat{P} + A(t)]^2 + U(x)$$

$$\hat{H}_{dE} = \frac{\hat{p}^2}{2} + U(x) - \hat{d}E(t)$$

$$\Psi_{dE}(t) = e^{iA(t)x} \Psi_{pA}(t)$$

Both \mathbf{P} and \mathbf{p} are the same momentum operator $-i \partial/\partial \mathbf{x}$, but the corresponding numbers have completely different physical meaning.

All observables are the same in both gauges if calculations are exact.

Interpretations and predictions based on approximations often run into problems if the difference between \mathbf{P} and \mathbf{p} is not properly recognized.

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [\hat{H}_0 + \hat{V}(t)] \Psi$$

H_0 is field-free Hamiltonian, $V(t) = -\mathbf{d} \cdot \mathbf{E}$ is interaction with the field.

- Approaches could be very different if continuum (ionization) is important / not important.

- Consider bound motion first

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [\hat{H}_0 + \hat{V}(t)] \Psi \qquad \Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

ϕ_n are eigenstates of H_0 with energies E_n .

They form complete basis set, hence Ψ can be written as above.

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [\hat{H}_0 + \hat{V}(t)] \Psi \qquad \Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

Substitute into TDSE:

$$i \sum_n \dot{a}_n(t) \phi_n(x) = \sum_n a_n(t) E_n \phi_n(x) + \hat{V} \sum_n a_n(t) \phi_n(x)$$

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [H_0 + V(t)] \Psi$$

$$\Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

Introduce the Dirac notations

$$i \sum_n \dot{a}_n(t) |\phi_n\rangle = \sum_n a_n(t) E_n |\phi_n\rangle + \hat{V} \sum_n a_n(t) |\phi_n\rangle$$

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [H_0 + V(t)] \Psi \qquad \Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

Multiply with $\langle \phi_k |$ both sides and use orthogonality $\langle \phi_k | \phi_n \rangle = \delta_{kn}$:

$$\langle \phi_k | * i \sum_n \dot{a}_n(t) | \phi_n \rangle = \sum_n a_n(t) E_n | \phi_n \rangle + \hat{V} \sum_n a_n(t) | \phi_n \rangle$$

$$i \dot{a}_k(t) = E_k a_k(t) + \sum_n V_{kn}(t) a_n(t)$$

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [H_0 + V(t)] \Psi$$

$$\Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

$$i \dot{a}_k(t) = E_k a_k(t) + \sum_n V_{kn}(t) a_n(t)$$

Huge simplification: set of simple 1-st order DE, coupled to each other.
Works for complex multi-electron dynamics if you know E_k and V_{kn}

All complexity is hidden in energies E_k and matrix elements $\langle \phi_k | V | \phi_n \rangle =$
 $V_{kn} = -d_{kn} E(t)$ *need a friend who is a good quantum chemist*

TDSE in laser fields

$$i \frac{\partial \Psi}{\partial t} = [H_0 + V(t)] \Psi$$

$$\Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

$$i \dot{a}_k(t) = E_k a_k(t) + \sum_n V_{kn}(t) a_n(t)$$

All complexity is hidden in energies E_k and matrix elements $\langle \phi_k | V | \phi_n \rangle = V_{kn} = -d_{kn} E(t)$

Need a friend who is a good quantum chemist



Serguei Patchkovskii,
NRC, Ottawa

TDSE in laser fields

$$\Psi(x, t) = \sum_n a_n(t) \phi_n(x)$$

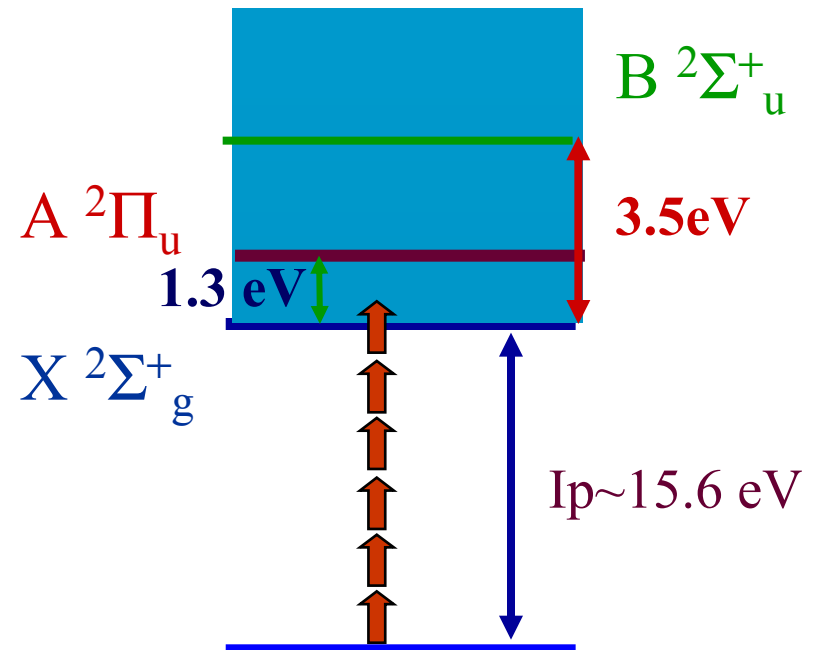
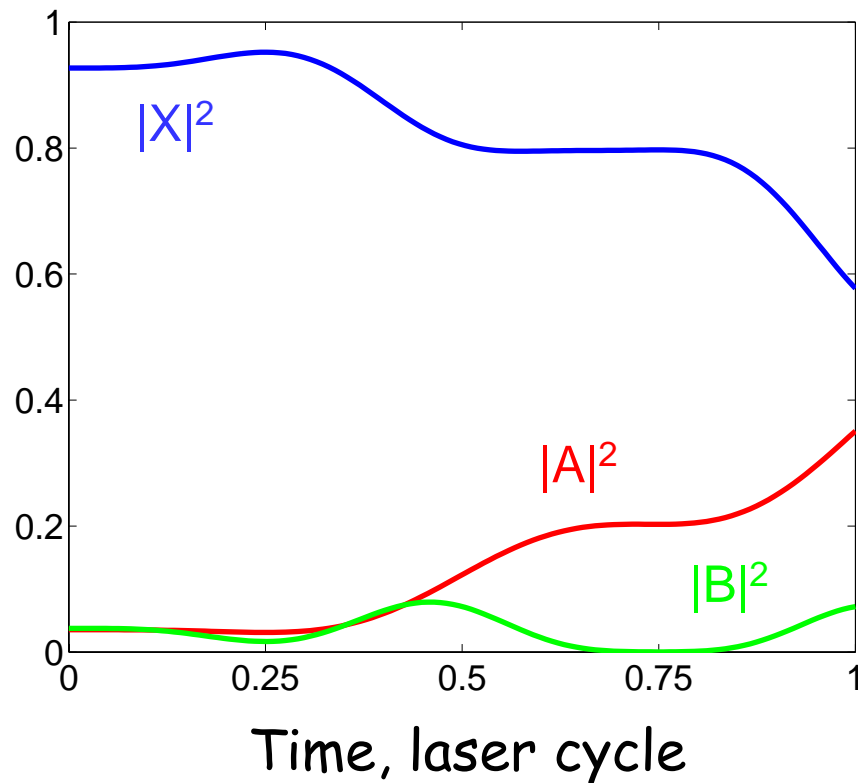
$$i\dot{a}_k(t) = E_k a_k(t) + \sum_n V_{kn}(t) a_n(t)$$

$$i\dot{A} = B(t)A \quad A = \begin{bmatrix} a_1 \\ a_2 \\ a_3 \end{bmatrix} \quad B = \begin{bmatrix} E_1 & V_{12} & V_{13} \\ V_{21} & E_2 & V_{23} \\ V_{31} & V_{32} & E_3 \end{bmatrix}$$

Numerical approach: Runge - Kutta

Example: attosecond dynamics in N_2^+

Populations of ionic states



Initial condition: population of the polarized ground state of N_2^+ upon ionization, $I=10^{14}W/cm^2$

Note to remember

$$i \frac{\partial \Psi}{\partial t} = [H_0 + V(t)] \Psi \quad \Psi(x, t) = \sum_n a_n(t) e^{-iE_n t} \phi_n(x)$$

$$i \dot{a}_k(t) = \sum_n V_{kn}(t) e^{i(E_k - E_n)t} a_n(t)$$

This is called ‘The Interaction Picture’

Pay attention to how the wave-function is defined/written

TDSE in integral form

$$i\partial_t \Psi = H\Psi \Rightarrow$$

$$\Rightarrow \Psi(t) = e^{-i\int_0^t \hat{H}(\tau) d\tau} \Psi(0)$$

The exponent in front is a **propagator**.

The integral form is no better than the differential form, unless you somehow happened to know the propagator

Dealing with the propagator: the split-step method

$$i\partial_t \Psi = H\Psi \Rightarrow$$

$$\Rightarrow \Psi(t) = e^{-i\int_0^t \hat{H}(\tau) d\tau} \Psi(0)$$

How does one evaluate the exponential operator?

The first thing to do – break the propagation it into small time steps

Do $n=1,100\dots 0$

$$\Psi(t_{n-1} + \Delta t) = e^{-i\hat{H}(t_{n-1} + \Delta t/2)\Delta t} \Psi(t_{n-1})$$

End Do

The split-step method

Do n=1,100...0

$$\Psi(t_{n-1} + \Delta t) = e^{-i\hat{H}(t_{n-1} + \Delta t/2)\Delta t} \Psi(t_{n-1})$$

End Do

Still need to find exponential operator over a short time-interval

$$e^{-i\hat{H}(t_{n-1} + \Delta t/2)\Delta t} = e^{-i[\hat{H}_0 + \hat{x}E(t_{n-1} + \Delta t/2)]\Delta t} = ?$$

The problem is the kinetic energy - it does not commute with xE or $U(x)$

$$\hat{H}(t) = -\frac{1}{2} \frac{d^2}{dx^2} + U(x) + xE(t) = \hat{K} + U(x, t)$$

$$e^{-i(\hat{K} + U)\Delta t} \neq e^{-i\hat{K}\Delta t} e^{-iU\Delta t}$$

The split-step method: the good news

For small time steps corrections due to non-zero commutators are small!

$$e^{-i(\hat{K}+U)\Delta t} \cong e^{-iU\Delta t/2} e^{-i\hat{K}\Delta t} e^{-iU\Delta t/2} + O(\Delta t^3)$$

$$e^{-i(\hat{K}+U)\Delta t} \cong e^{-i\hat{K}\Delta t/2} e^{-iU\Delta t} e^{-i\hat{K}\Delta t/2} + O(\Delta t^3)$$

To deal with

$$e^{-i\frac{1}{2}\hat{p}^2\Delta t}\Psi(x,t)$$

remember that it is just a multiplication in p-space

The split-step method

Do n=1,100...0

Multiplication:

$$\Phi(x, t) = e^{-iU(x,t)\Delta t/2} \Psi(x, t)$$

Fourier transform:

$$\tilde{\Phi}(p, t) = \text{FFT}[\Phi(x, t)]$$

Multiplication:

$$\tilde{\Phi}(p, t + \Delta t) = e^{-i\frac{1}{2}p^2\Delta t} \tilde{\Phi}(p, t)$$

Inverse Fourier transform:

$$\Phi(x, t + \Delta t) = \text{FFT}^{-1}[\tilde{\Phi}(p, t + \Delta t)]$$

Multiplication:

$$\Psi(x, t + \Delta t) = e^{-iU(x,t)\Delta t/2} \Phi(x, t + \Delta t)$$

End Do

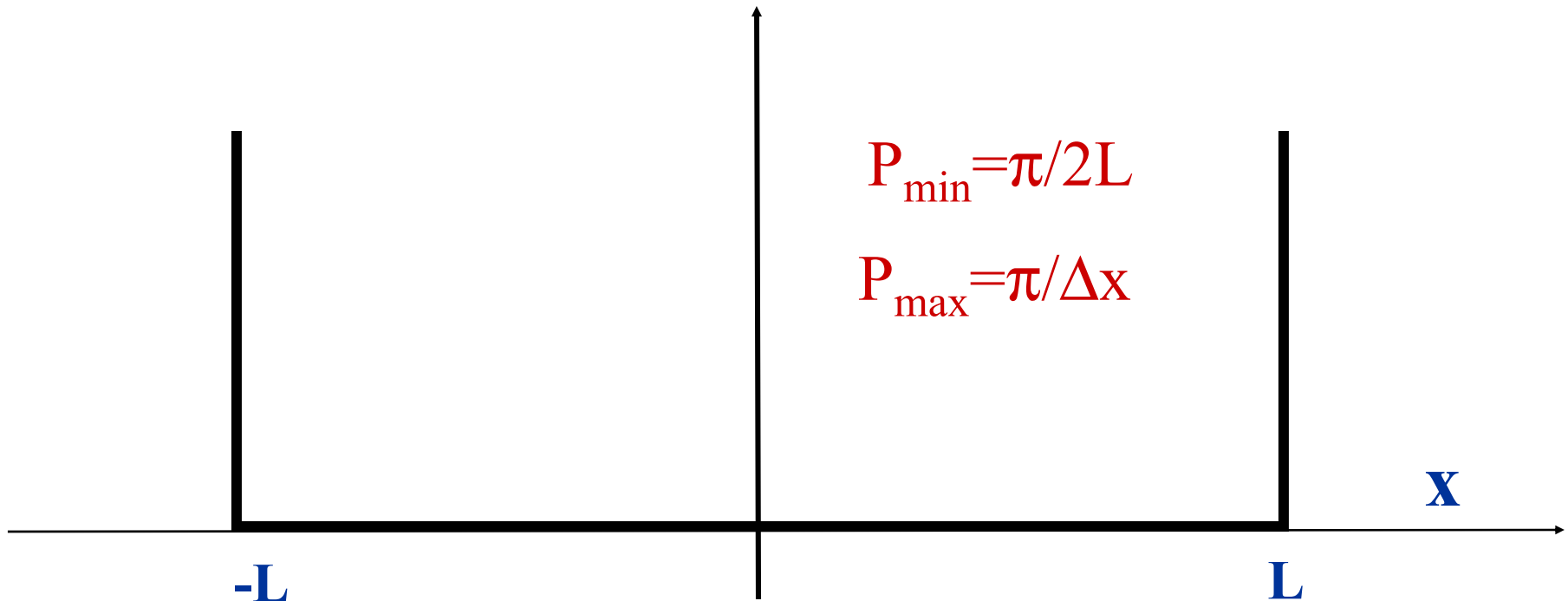
Also works for bound dynamics, but using basis is much better

The split-step method: cautionary notes

Note # 1:

For a box in coordinate space between $-L \dots +L$ and grid step Δx

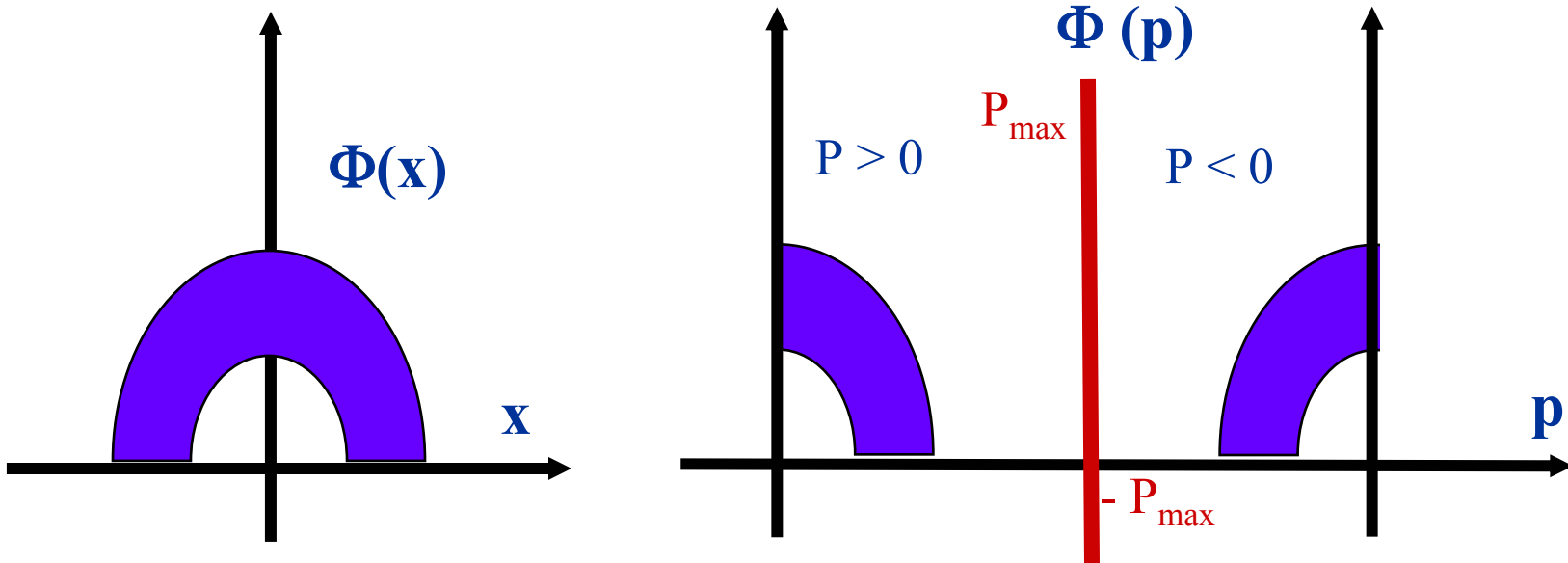
- Smallest momentum $P_{\min} = \pi/2L$
- Largest momentum $P_{\max} = \pi/\Delta x$



The split-step method: cautionary notes

Note # 2:

- FFT often screws up the array as shown in the figure below
- Careful when assigning the elements of the array $\Phi(p)$ to correct momenta:
take a Gaussian in x , FFT it, look at the result!

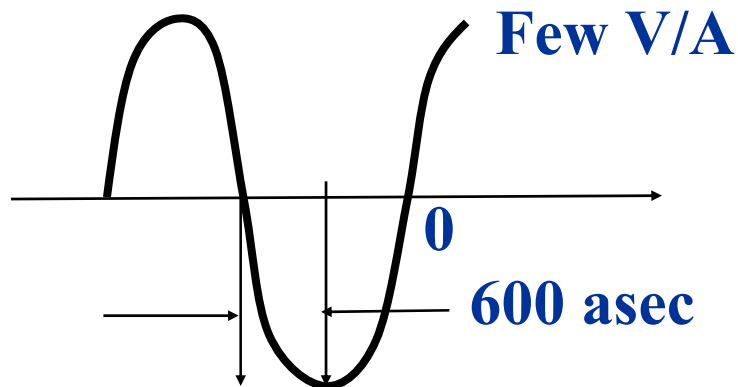


TDSE in strong fields: why is it hard?

1. Velocity could be very high: $v \sim E/\omega$, could go up to a few E/ω

Take intensity $I \sim 10^{14} \text{W/cm}^2$, ω for 800nm, and you get $mv^2/2 \sim 100 \text{ eV}$

2. In <1 fsec field changes from zero to very strong: a lot can happen in 10 asec



3. Electron excursion is large:
oscillation amplitude $E/\omega^2 \sim 20-30 \text{ au}$

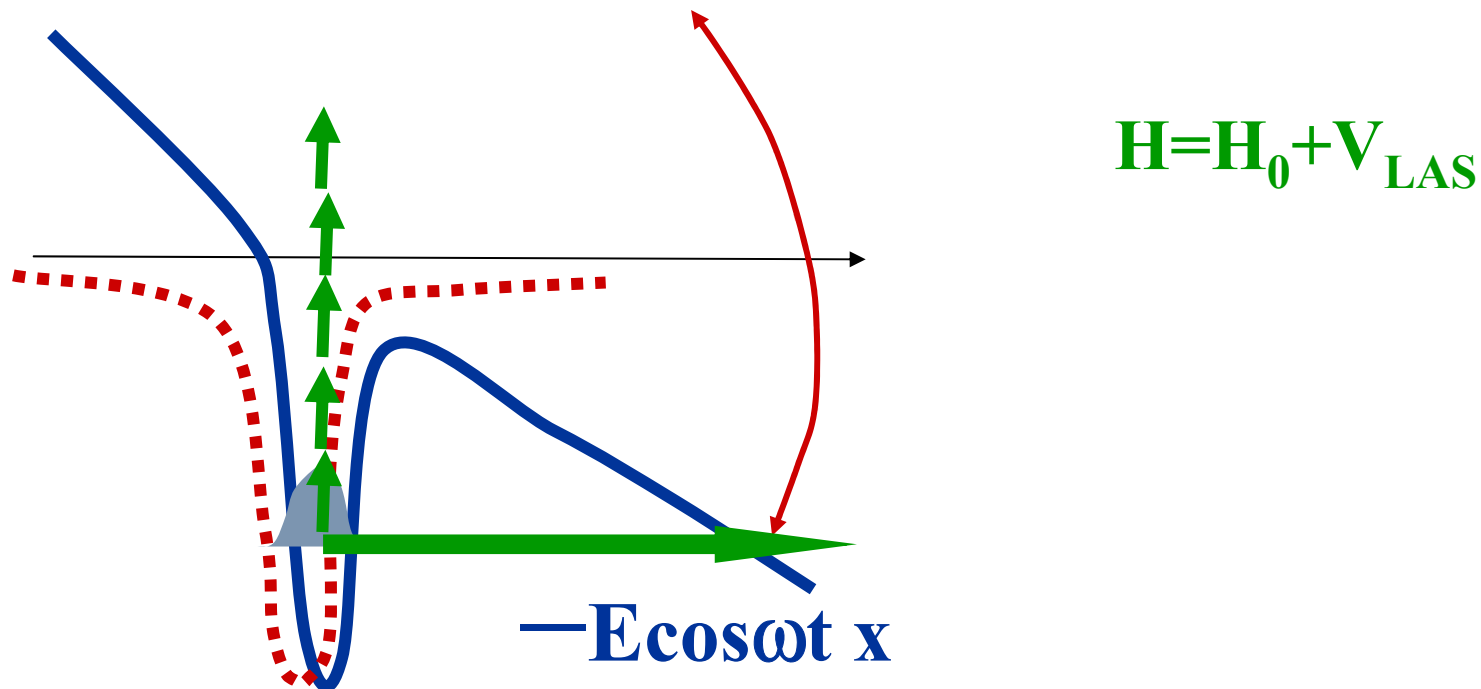
If we want electron spectra, we have to keep fast electrons for long times – very large distances needed (e.g. 500 au)

Need: very high time resolution (small Δt), very large distances L , very high energies hence very small Δx

Strong-field S-matrix approach

- S-matrix approach to strong fields
- Strong field approximation
- Strong field ionization
- MPI vs Tunneling – a peaceful coexistence
- Electron after tunneling: how does it look?
- Resonances in strong-field ionization

Strong-field ionization



What is the dynamics of ionization?

When does it go vertical, and when horizontal?

How does the electron wavepacket look like?

TDSE in integral form

$$i\partial_t \Psi = H\Psi \Rightarrow$$

$$\Rightarrow \Psi(t) = e^{-i\int_0^t \hat{H}(\tau) d\tau} \Psi(0)$$

The exponent in front is a **propagator**.

The integral form is no better than the differential form, unless you somehow happened to know the propagator

TDSE in integral form II: S-matrix expressions

$$\mathbf{H}=\mathbf{H}_0+\mathbf{V},$$

$$\Psi(\mathbf{t})=\Psi_0(\mathbf{t})+\Delta\Psi$$

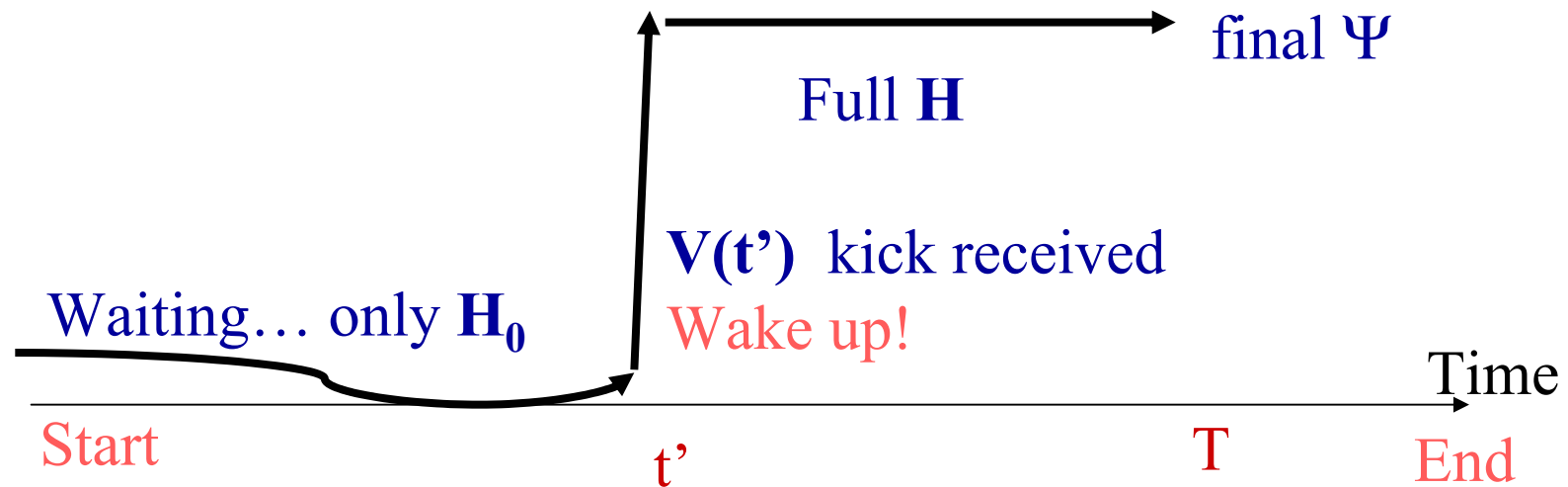
$$\Delta\Psi(\mathbf{T}) = -i \int_0^{\mathbf{T}} dt' e^{-i \int_{t'}^{\mathbf{T}} dt'' H(t'')} \mathbf{V}(t') e^{-i \int_0^{t'} dt'' H_0(t'')} \Psi_0(t=0)$$

- This looks even worse than before. However, this form is well suited for various approximations

(One example is the standard **perturbation theory**: replace \mathbf{H} with \mathbf{H}_0)

Physical picture of general S-matrix expressions

$$\Delta\Psi(T) = -i \int_0^T dt' e^{-i \int_{t'}^T dt'' H(t'')} V(t') e^{-i \int_0^{t'} dt'' H_0(t'')} \Psi_0(t=0)$$



S-matrix amplitudes

Let the initial state be $\Psi(t=0)=\Psi_0=\Psi_g$

Exact:

$$a_p(T) = -i \int_0^T dt' \langle \mathbf{p} | e^{-i \int_0^{t'} dt'' H(t'')} V(t') e^{-i \int_0^{t'} dt'' H_0(t'')} | \Psi_0 \rangle$$

Outline

S-matrix approach to strong fields

-Strong field approximation

-Strong field ionization

-MPI vs Tunneling – a peaceful coexistence

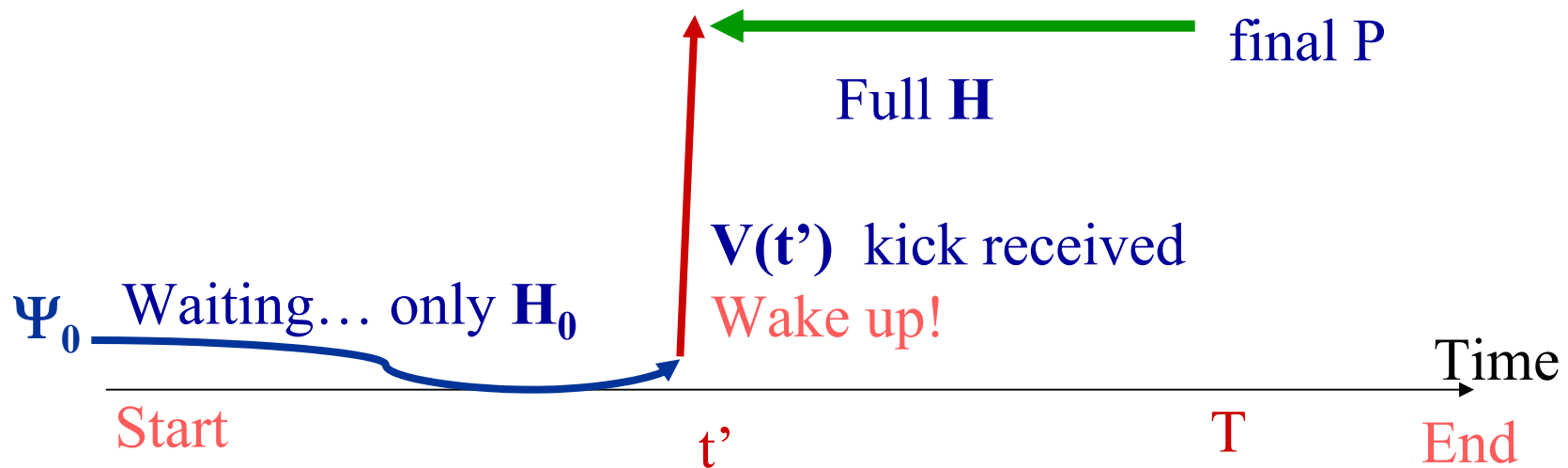
- Electron after tunneling: how does it look?

-Resonances in strong-field ionization

The SFA and the Volkov propagator

Exact:

$$a_p(T) = -i \int dt' \langle \mathbf{p} | e^{-i \int_{t'}^T dt'' H(t'')} V(t') e^{-i \int dt'' H_0(t'')} | \Psi_0 \rangle$$



The SFA and the Volkov propagator

Exact:

$$a_p(T) = -i \int^T dt' \left\langle \mathbf{p} \left| e^{-i \int_{t'}^T dt'' H(t'')} \right. V(t') e^{-i \int_{t'}^{t'} dt'' H_0(t'')} \right| \Psi_0 \rangle$$

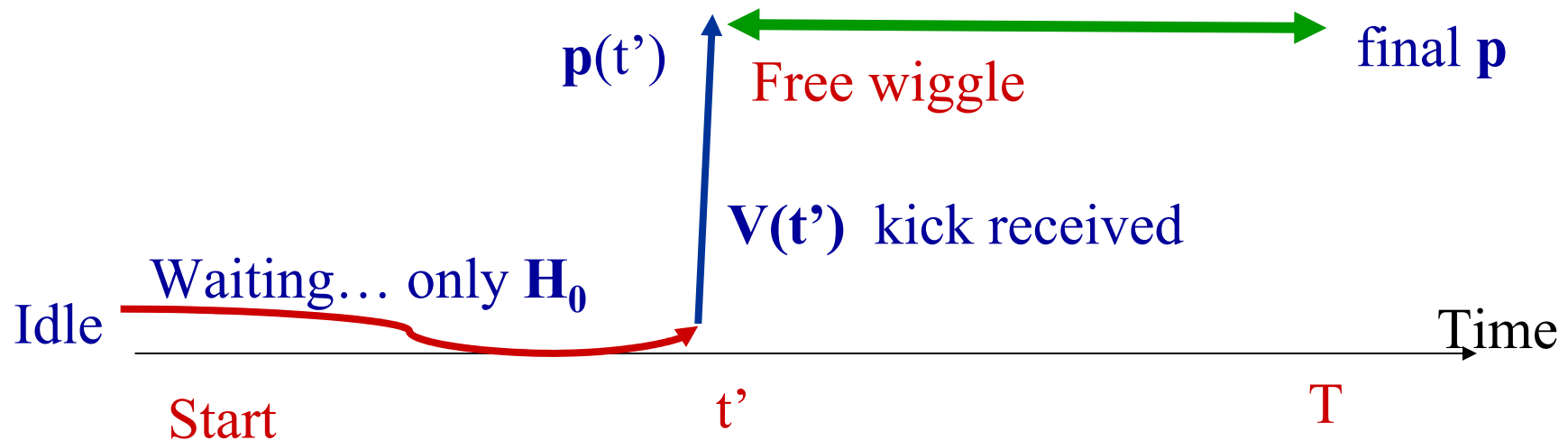
Strong Field Approximation :

replace $\left\langle \mathbf{p} \left| e^{-i \int_{t'}^T dt'' H(t'')} \right.$ with $\left\langle \mathbf{p} \left| e^{-i \int_{t'}^T dt'' H_{LAS}(t'')} \right.$

The physics of SFA

Strong Field Approximation :

replace $\langle \mathbf{p} | e^{-i \int_{t'}^T dt'' H(t'')}$ with $\langle \mathbf{p} | e^{-i \int_{t'}^T dt'' H_{LAS}(t'')}$



Volkov propagator, Length gauge

$$\hat{H}_{LAS,dE} = \frac{\hat{p}^2}{2} - \hat{d}E(t) = \frac{\hat{p}^2}{2} + xE(t)$$

Free wiggle, length gauge

$$\mathbf{p}(t') = \mathbf{p}(T) - \mathbf{A}(T) + \mathbf{A}(t')$$

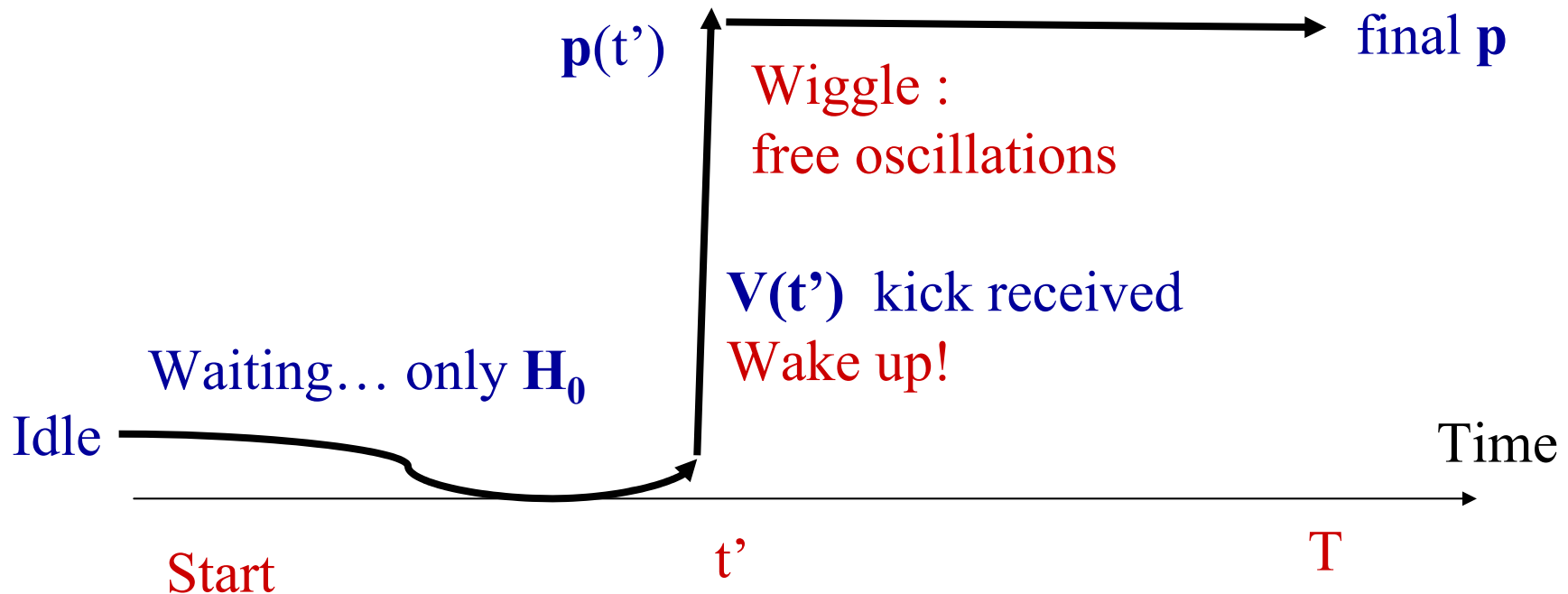
then

$$\langle \mathbf{p} | e^{-i \int_{t'}^T dt'' H_{LAS}(t'')} = e^{-i \int_{t'}^T dt'' [\mathbf{p} - \mathbf{A}(T) + \mathbf{A}(t'')]^2} \langle \mathbf{p} - \mathbf{A}(T) + \mathbf{A}(t') |$$

SFA, length gauge: Equations and physics

$$\mathbf{a}_p(\mathbf{T}) = -i \int dt' e^{-i \frac{1}{2} \int_{t'}^{\mathbf{T}} dt'' [\mathbf{p} - \mathbf{A}(\mathbf{T}) + \mathbf{A}(t'')]^2 + i I_p t'} \mathbf{E} \cos \omega t' \langle \mathbf{p}(t') | \mathbf{x} | \Psi_g \rangle$$

$$\mathbf{p}(t') = \mathbf{p} - \mathbf{A}(\mathbf{T}) + \mathbf{A}(t')$$



Volkov propagator, velocity gauge

$$\hat{H}_{LAS,pA} = \frac{1}{2}[\hat{P} + A(t)]^2$$

Free wiggler, velocity gauge

$$\mathbf{P}(t') = \mathbf{P}(T), \quad \mathbf{v}(t') = \mathbf{P} + \mathbf{A}(t')$$

then

$$\langle \mathbf{p} | e^{-i \int_{t'}^T dt'' H_{LAS}(t'')} \rangle = e^{-i \frac{1}{2} \int_{t'}^T [P + A(t'')]^2 dt''} \langle P |$$

SFA: velocity gauge vs length gauge

$$a_P(T) = i \int dt' e^{-i \frac{1}{2} \int_{t'}^T [P + A(t'')]^2 dt'' + i I_p t'} \frac{E}{\omega} \sin \omega t' \langle P | \hat{p} | \Psi_g \rangle$$

Compare this with length gauge:

$$\mathbf{a}_p(T) = -i \int dt' e^{-i \frac{1}{2} \int_{t'}^T dt'' [\mathbf{p} - \mathbf{A}(T) + \mathbf{A}(t'')]^2 + i I_p t'} \mathbf{E} \cos \omega t' \langle \mathbf{p}(t') | \mathbf{x} | \Psi_g \rangle$$

In length gauge, to create \mathbf{p} at T , we start with $\mathbf{p}(t') = \mathbf{p} - \mathbf{A}(T) + \mathbf{A}(t')$

In velocity gauge, we take P from the very beginning

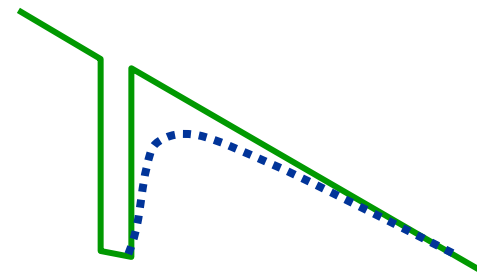
What are the main problems with SFA?

Starting from the ground state, we have

$$\mathbf{a}_p(\mathbf{T}) = -i \int dt' e^{-i \frac{1}{2} \int_{t'}^{\mathbf{T}} dt'' [\mathbf{p} - \mathbf{A}(\mathbf{T}) + \mathbf{A}(t'')]^2 + i I_p t'} \mathbf{E} \cos \omega t' \langle \mathbf{p}(t') | \mathbf{x} | \Psi_{\text{gs}} \rangle$$

Main problems:

- No laser induced dynamics inside the potential well – only field-free
- No AC-Stark shift, no resonances due to bound states
- Incorrect shape of the potential barrier
- Not gauge-invariant



SFA remarks

The SFA is full of drawbacks and "wrongs" that fly right into the face of any rigorous quantum theory

But it works for getting the basic physics right - and that's what counts in the end

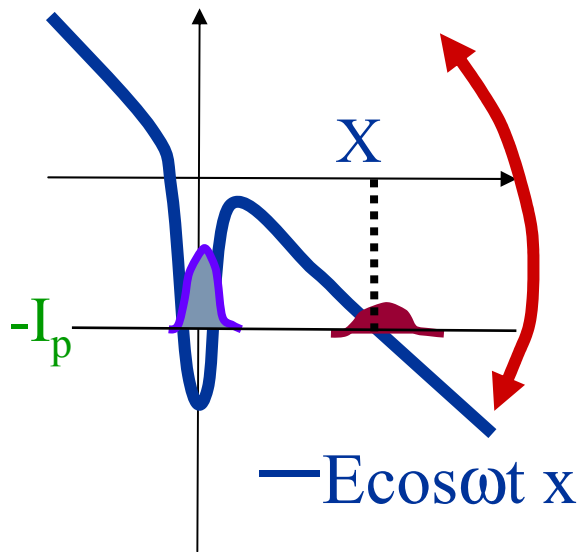
However, SFA has to be accompanied by physical understanding of what each approximation means.

Straightforward –**Blind** - application of SFA-type approaches without reference to physical intuition may lead to **complete nonsense** in the end

Outline

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- Two gauges: length and velocity
- S-matrix approach to strong fields
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- Strong field ionization**
- Electron after tunneling: how does it look?
- MPI vs Tunneling – a peaceful coexistence
- Resonances in strong-field ionization

Strong-field ionization, SFA



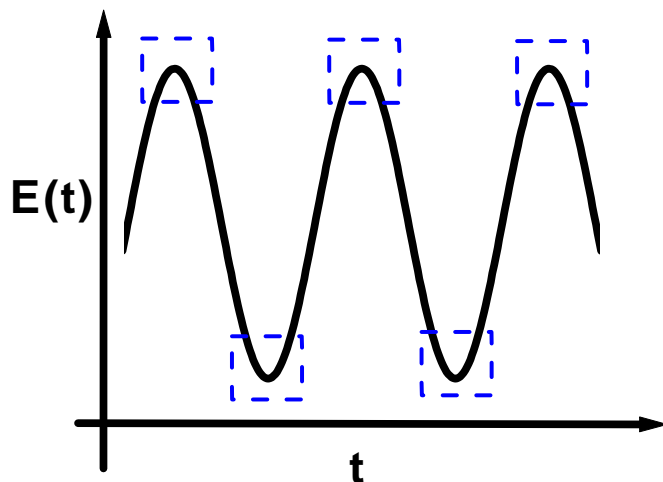
How static is the barrier? – Tunneling time
Keldysh, Buttiker & Landauer

$$X \approx I_p/E \quad , \quad v \approx \sqrt{2I_p}/2$$

$$\tau = X/v = \sqrt{2I_p}/E, \quad \gamma = \omega\tau = \omega\sqrt{2I_p}/E$$

Static for $\gamma < 1$, oscillating for $\gamma > 1$ –

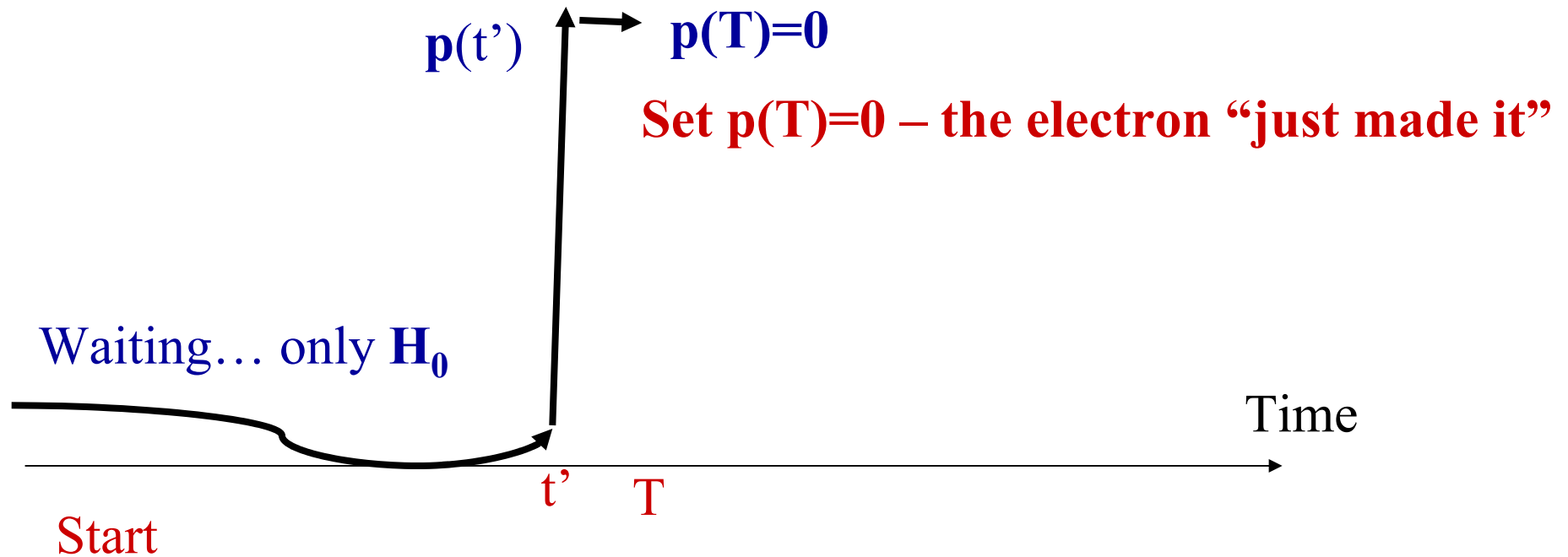
but $\gamma > 1$ does not mean that tunneling is not there! It means that tunneling is non-adiabatic



Electron shows up near the peaks not only for $\gamma < 1$ but also for $\gamma > 1$

Ionization - a **simple** calculation

Work in length gauge



Use t' closest to T : the electron “just made it”

Ionization – a simple calculation

$$a_p(T) \sim \int dt' e^{-i\frac{1}{2}\int_{t'}^T dt'' [p-A(T)+A(t'')]^2 + iI_p t'} E \cos \omega t' z_{p'g}$$

Stationary points in t' : [derivative of phase w.r. to t'] = 0

$$\frac{[p - A(T) + A(t')]^2}{2} + I_p = 0 \quad \text{NB: } t' \text{ is complex!}$$

- Set $p=0$ – the electron “just made it”
- Look at peaks of $E \cos \omega T$: Set $\cos \omega T = 1$ and $\sin \omega T = 0$, $A(T) = 0$
– get ionization where it matters most

$$\frac{[A(t')]^2}{2} + I_p = 0 \quad \text{Find } t' \text{ closest to } T: \text{Re}(t') = T -$$

the electron “just made it”

Saddle points for ionization

$$\frac{[E/\omega]^2}{2} \sin^2 \omega t' + I_p = 0$$

$t' = i\tau$ is always complex!!

$$\frac{[E/\omega]^2}{2} \operatorname{sh}^2 \omega \tau - I_p = 0 \quad \frac{I_p}{[E/\omega]^2/2} = \gamma^2 \quad \operatorname{sh}^2 \omega \tau - \gamma^2 = 0$$

For $\gamma \ll 1$

$\operatorname{sh} \omega \tau \sim \omega \tau$

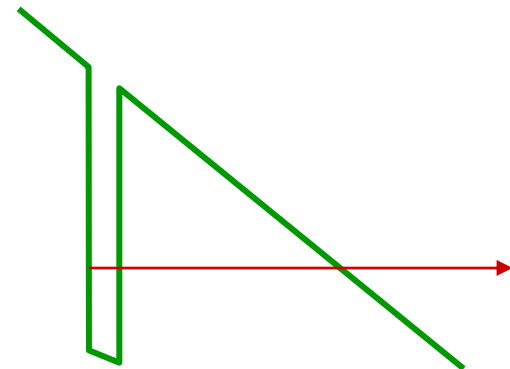
$$\omega \tau = \gamma, \quad \tau = \gamma/\omega = \sqrt{2I_p}/E$$

For $\gamma \gg 1$

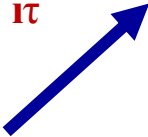
$\operatorname{sh} \omega \tau \sim \exp[\omega \tau]/2$

$$\omega \tau = \ln 2\gamma,$$

τ – the time spent in classically forbidden region – changes with γ



Using the saddle point

$$\mathbf{a}_{\text{ion}} \sim \int dt' e^{-i \frac{1}{2} \int_{t'}^T dt'' [A(t'')]^2 + i I_p t'} \sim e^{-i \frac{1}{2} \int_{it}^0 dt'' [A(t'')]^2 + i I_p (it)}$$


This is the action in classically forbidden region.

Careful to pick $i\tau$ or $-i\tau$

For small $\gamma < 1$ we get $\exp[-(2I_p)^{3/2}/3E]$ – DC tunneling exponent

For large $\gamma \gg 1$ we get $\exp[-(2I_p/\omega) \ln \gamma] = [1/\gamma^2]^{I_p/\omega} \sim [E^2]^{I_p/\omega}$

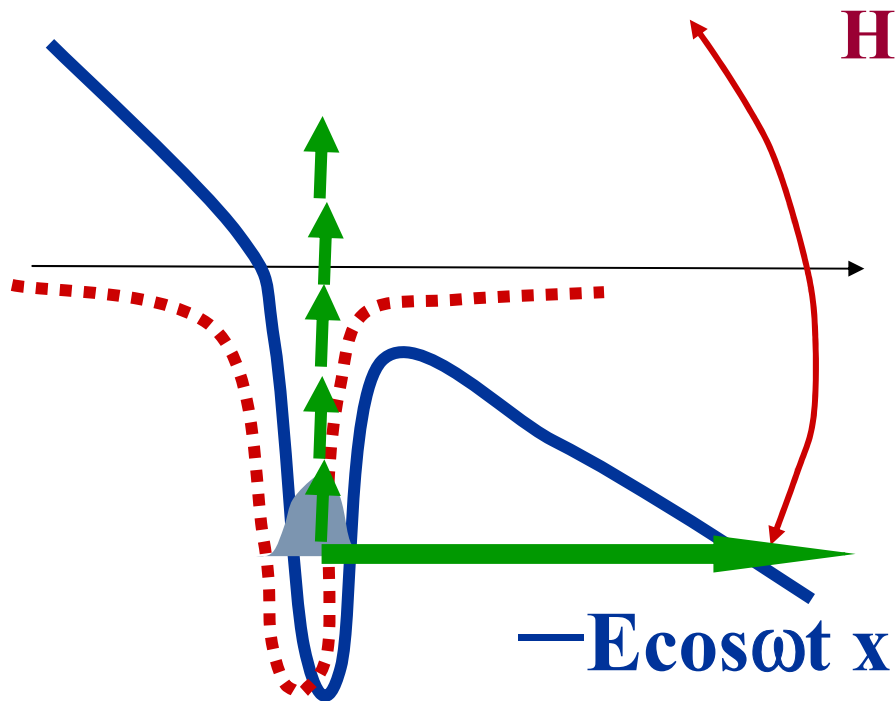
–MPI power law

BUT: Always have imaginary time and classically forbidden region

Outline

- Classical electron in a laser field
- Two gauges: length and velocity
- S-matrix approach to strong fields
- Strong field approximation
- Strong field ionization
- MPI vs Tunneling – a peaceful coexistence**
- Electron after tunneling: how does it look?
- Resonances in strong-field ionization

Tunneling vs Multiphoton ionization



How do the two channels co-exist?

Ionization includes:

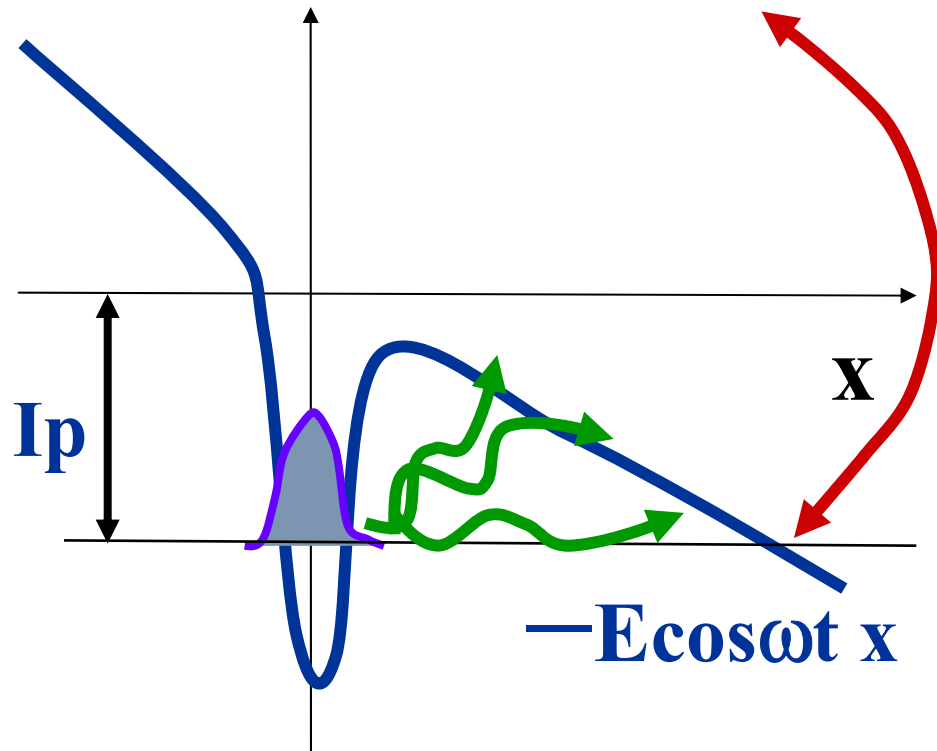
1. Dynamics under the barrier
2. Dynamics inside the well

NB: Keldysh-like theories ignore dynamics inside the well.

MPI in Keldysh-like theories is only via classically forbidden region
Remember – we always had a saddle point with imaginary time $t' = i\tau$!

Non-adiabatic tunneling

$\gamma = \omega\tau < 1$ –adiabatic tunneling. What if $\gamma = \omega\tau \sim$ or > 1 ?



$\gamma = \omega\tau$ refers only to motion under the barrier.

It says nothing about dynamics inside the well.

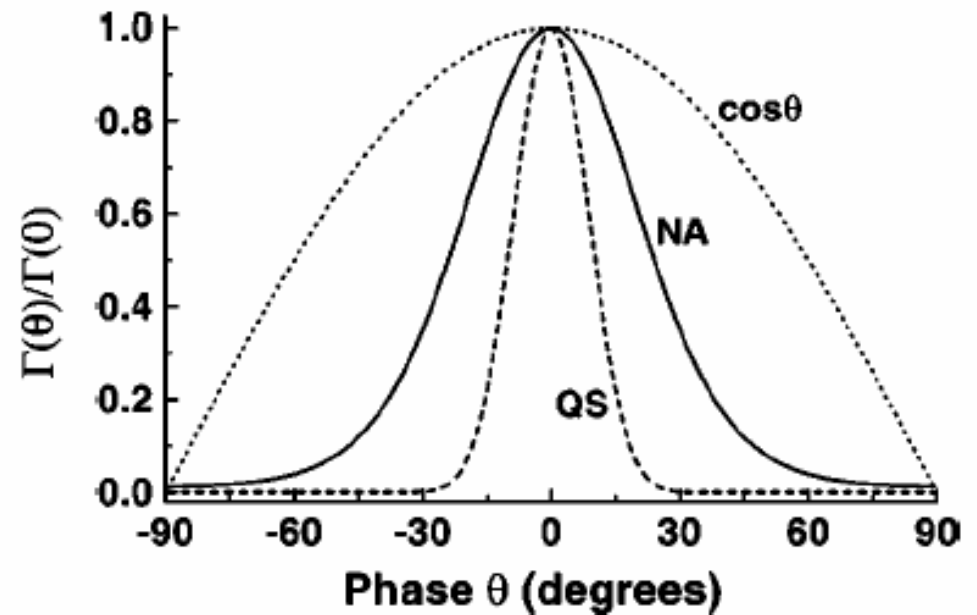
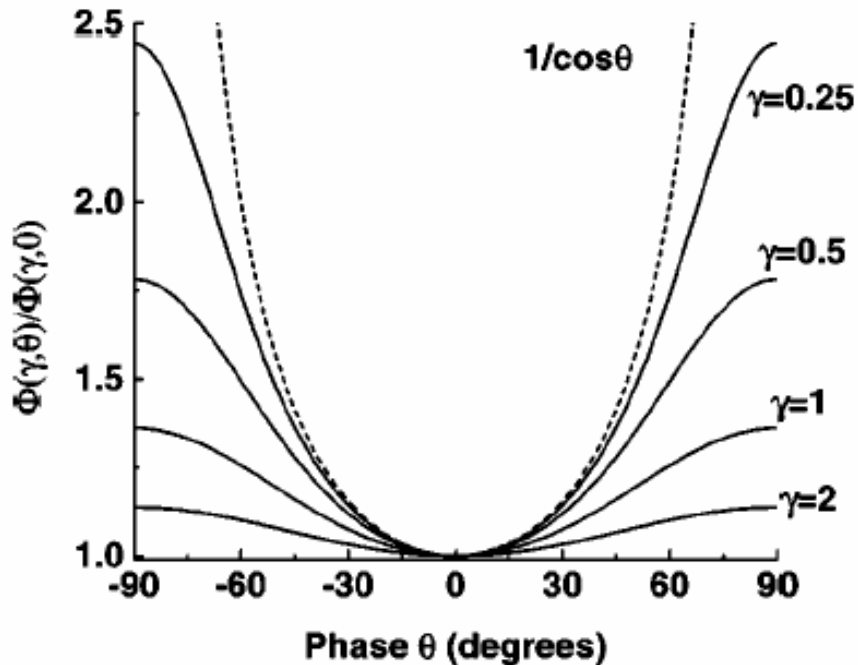
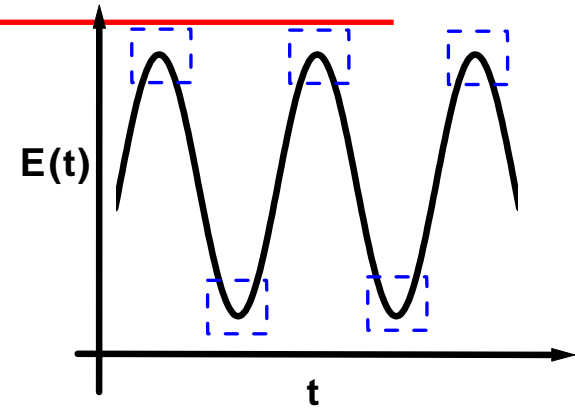
$\gamma < 1$ means that barrier is stationary during tunneling

$\gamma > 1$ means barrier is moving (heating in forbidden region)

MPI in Keldysh-like theories is always via classically forbidden region (i.e. **always involves tunneling**)

Non-adiabatic tunneling

$$\Gamma(\omega t) \sim \exp\left[-\frac{E^2}{\omega^3} \Phi(\gamma, \omega t)\right]$$

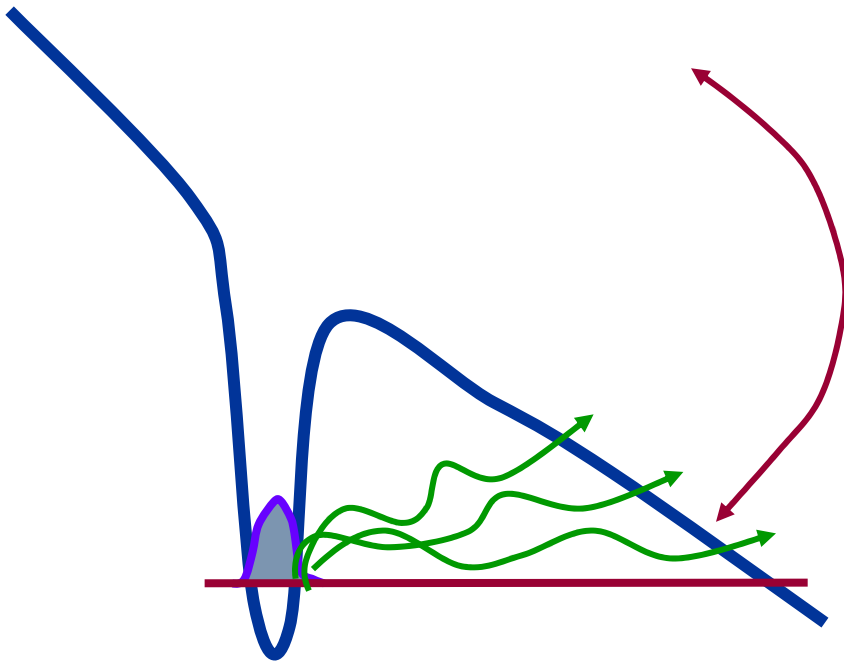


He, 780 nm, $5e13$ W/cm² $\gamma \sim 2$

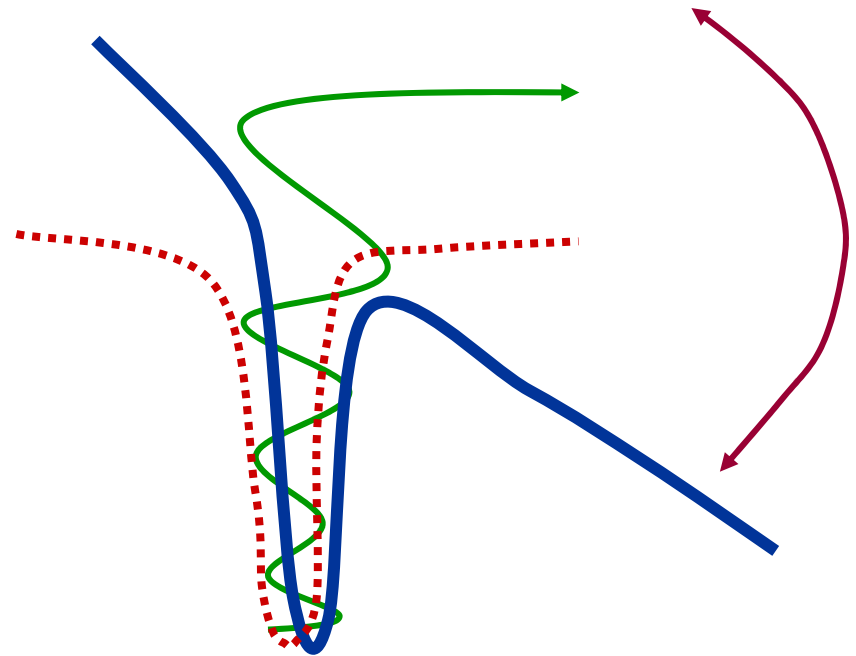
Step-wise ionization near peaks of the instantaneous field persists for $\gamma > 1$

Non-adiabatic tunneling and MPI

Present in Keldysh-like theories



Absent in Keldysh-like theories



For IR light $\omega_L \ll \omega_0$, dynamics inside the well is adiabatic:

Right channel is negligible for IR fields

Single active electron works for the first ionization step

because doubly excited states are too high, 2e tunneling is too hard

Remarks on tunneling and MPI

$\gamma = \omega\tau$ refers only to the motion under the barrier. It says nothing about the dynamics inside the well.

$\gamma < 1$ means that barrier is stationary during tunneling

$\gamma > 1$ means barrier is moving, heating in the forbidden region

$\gamma > 1$ **does NOT** mean that there is no tunneling

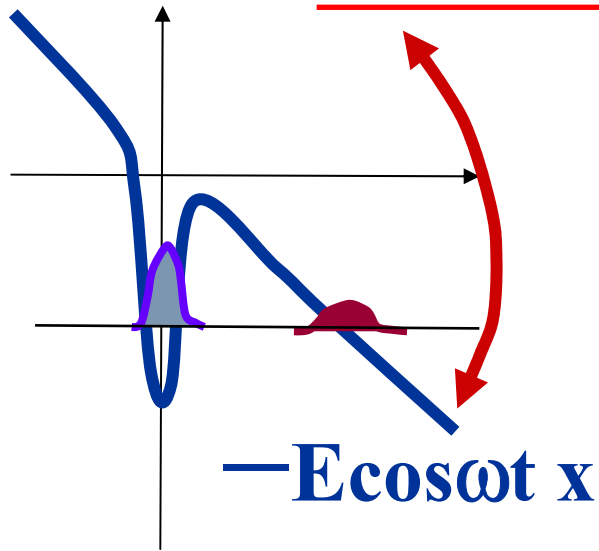
MPI in Keldysh-like theories is always via classically forbidden region - **always involves tunneling (Non-adiabatic tunneling)**

Step-wise ionization near peaks of the instantaneous field persists for $\gamma > 1$

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The wavepacket shape, $\gamma < 1$



$$W(v_{\parallel}, v_{\perp}) \sim \exp \left[-\frac{2(2I_p + v_{\perp}^2)^{3/2}}{3E \cos \omega t(v_{\parallel})} \right]$$

$$v_{\parallel} = \frac{E}{\omega} \sin \omega t(v_{\parallel}) = \sqrt{4Up} \sin \omega t(v_{\parallel})$$

$$W(v_{\parallel}, v_{\perp}) \sim W(0,0) e^{-\frac{(2I_p)^{3/2}}{3E} \frac{v_{\parallel}^2}{4Up}} e^{-v_{\perp}^2 \tau}$$

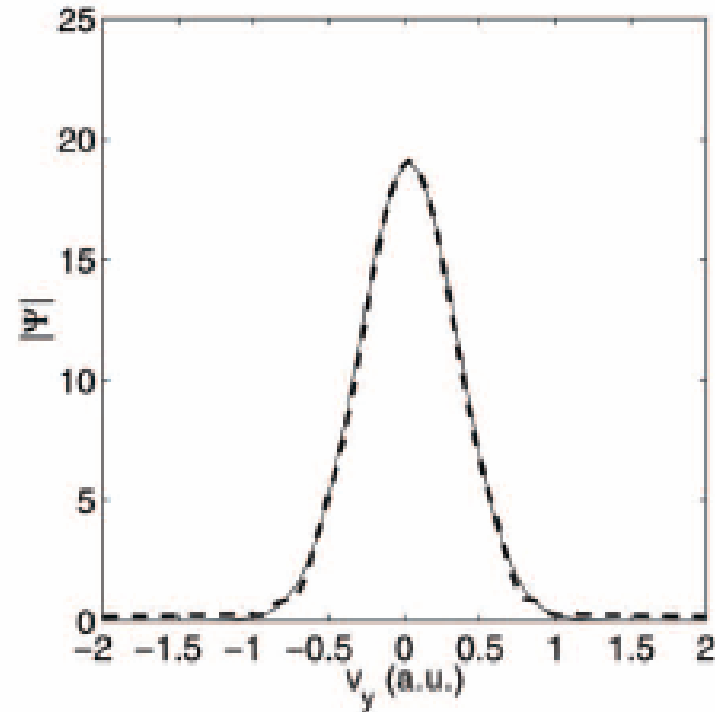
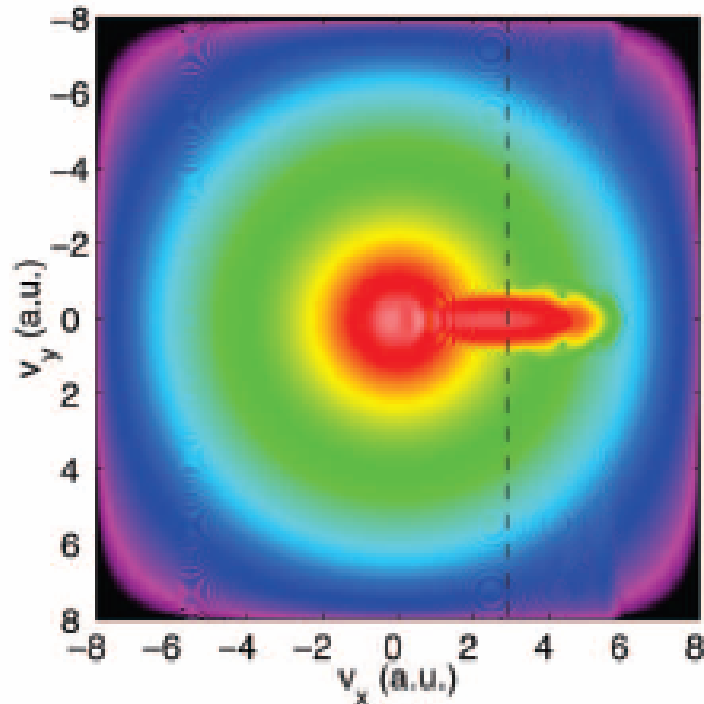
$$\omega \tau = \gamma, \quad \tau = \gamma / \omega = \sqrt{2I_p} / E$$

Distribution of v_{\parallel} (along field) is much longer than perpendicular

The wavepacket shape

Anatomy of strong field ionization

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$I=7.e14\text{W/cm}^2$ $\lambda=800\text{ nm}$

$I_p \sim 18\text{ eV}$

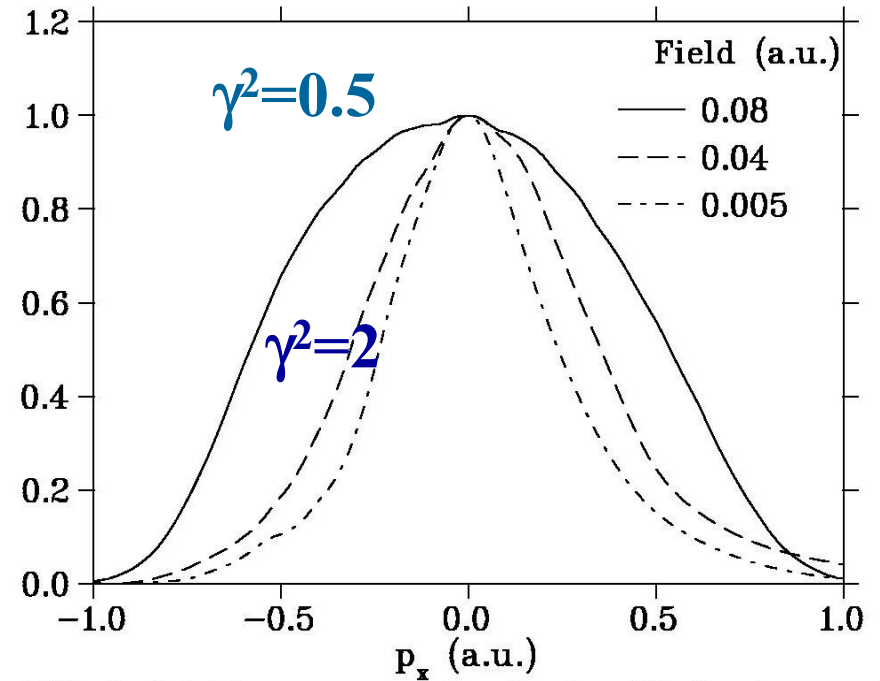
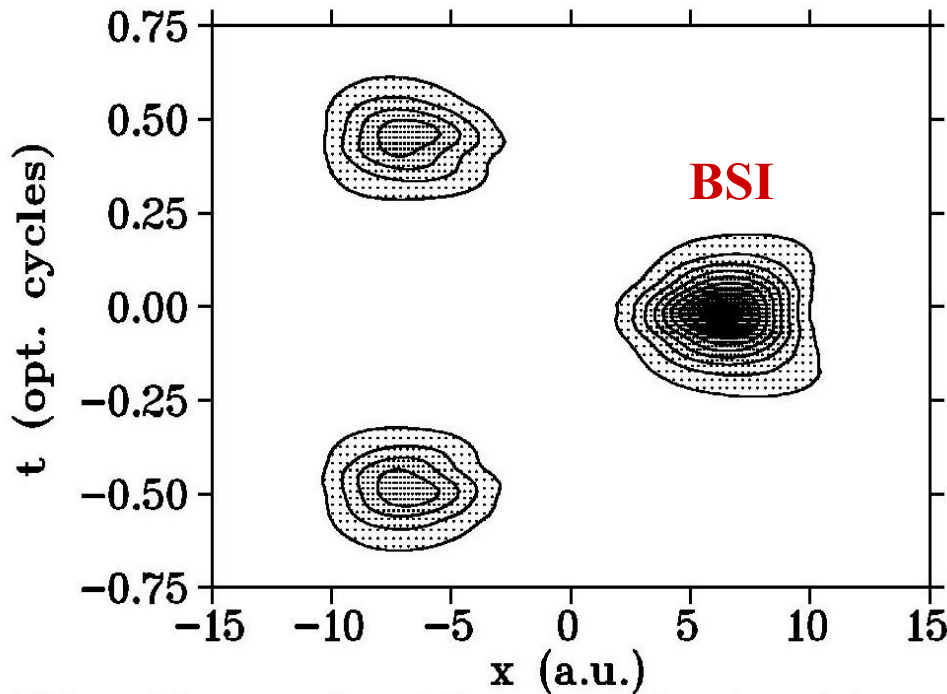
Wavepacket after 1 half-cycle

Parallel distribution is much longer than perpendicular

$$V_A(x, y) = -\frac{1}{\sqrt{x^2 + y^2 + 0.25}}$$

Strong-field ionization around $\gamma \sim 1$

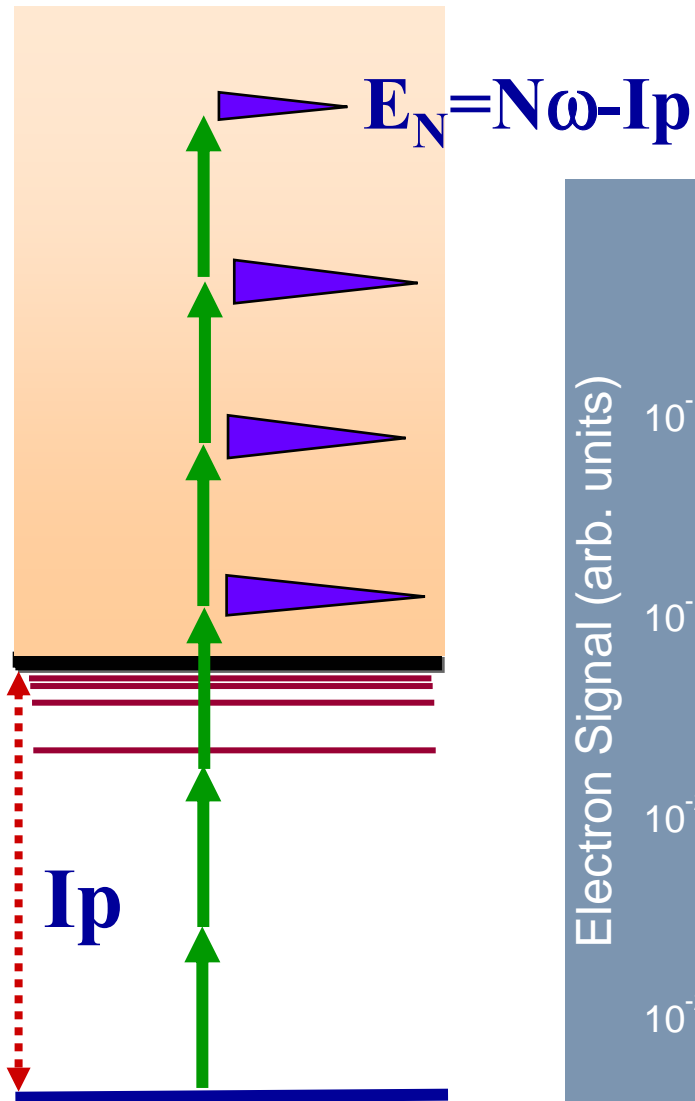
From A. Scrinzi: H, 800nm, 5fs, 2×10^{14} W/cm²



Barrier suppression at 1.4×10^{14} W/cm²

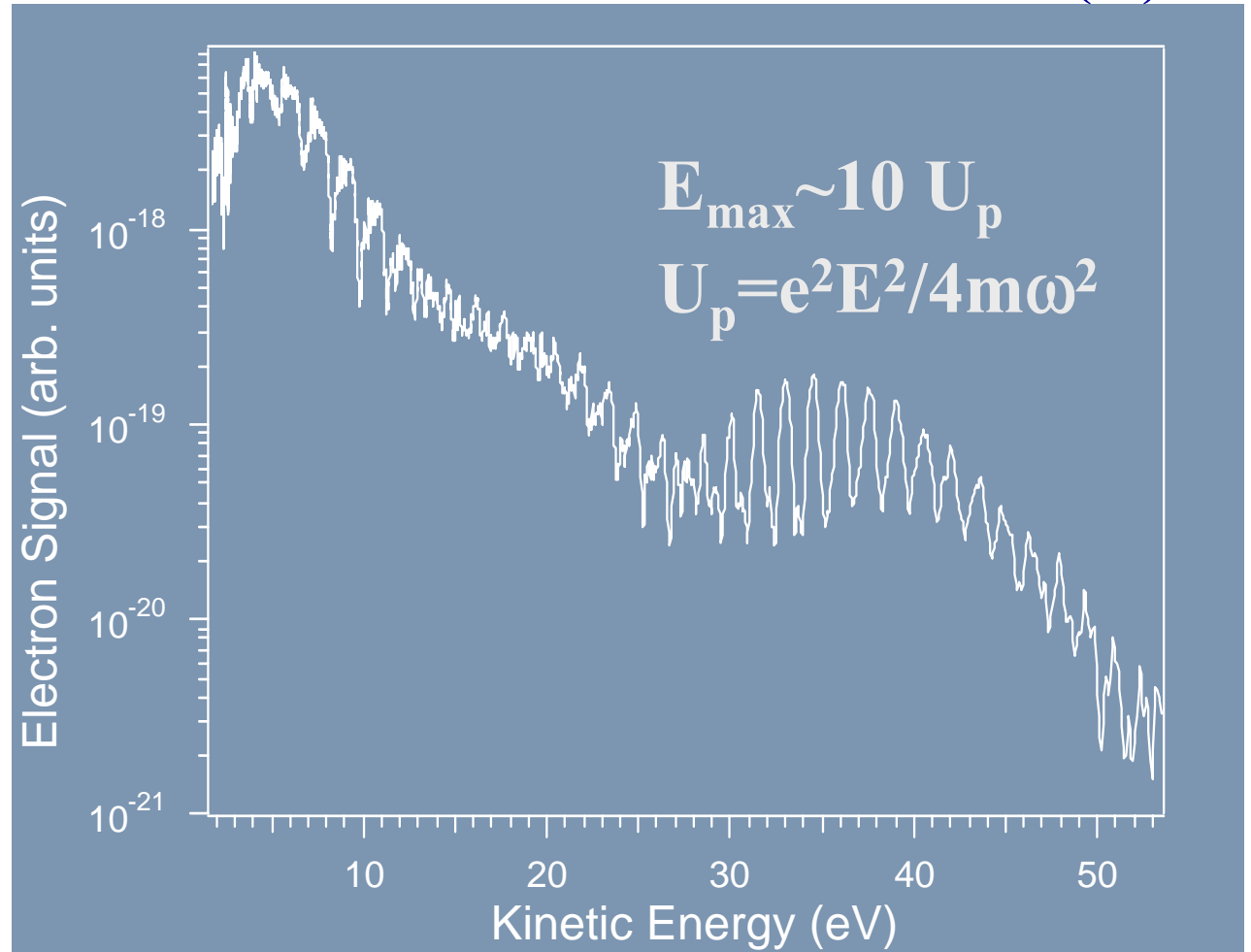
Even around $\gamma \sim 1$ electron shows up near the peaks of the field
The wavepacket's shape is as expected

Above Threshold Ionization

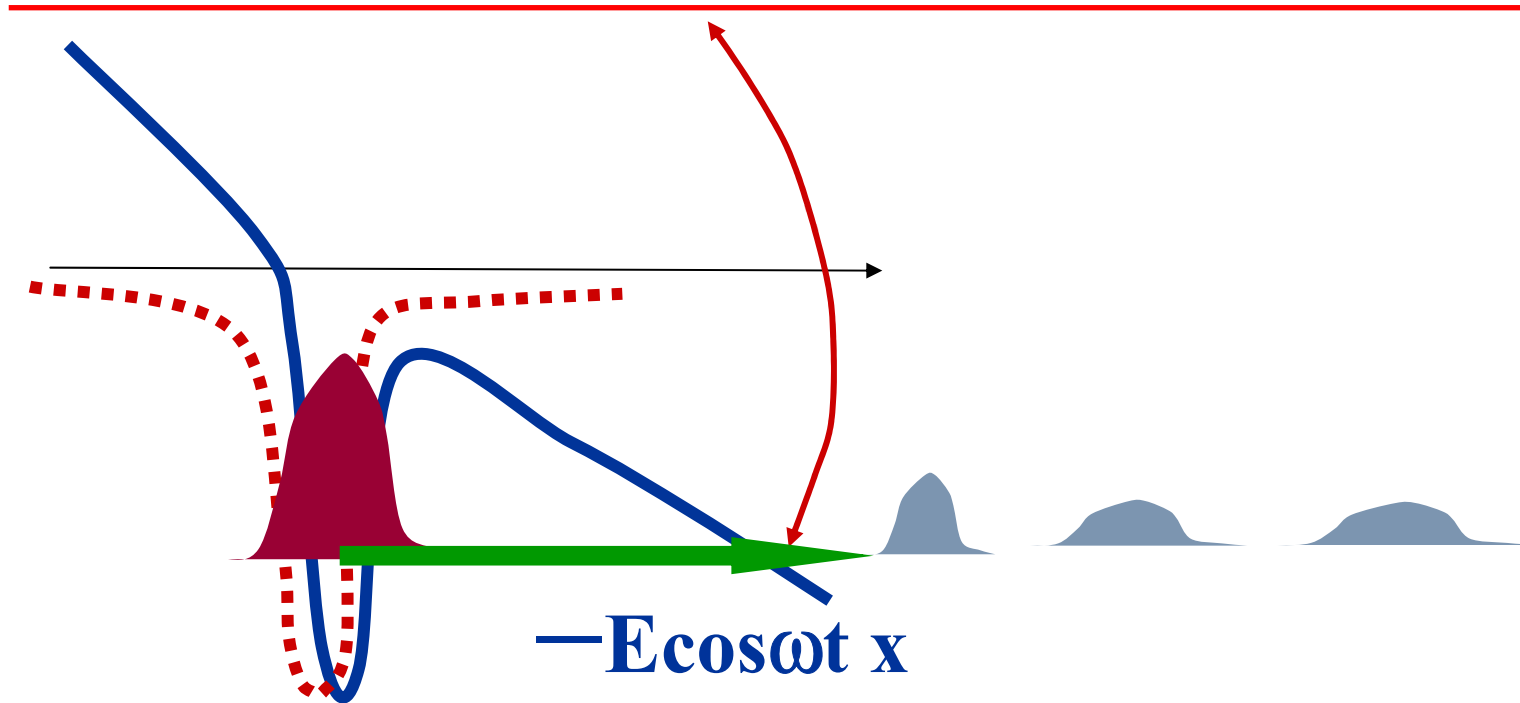


Argon, 120 fs Ti:S, $7 \cdot 10^{13} \text{ W/cm}^2$

L. van Woerkom's website & M. Nandor et al
PRA (99)



ATI structure in tunneling regime



Periodicity in ionization leads to periodic bursts of electrons
-- discrete structures in energy domain

Tunneling does not mean that ATI spectrum loses its discrete structure

Outline

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- Electron after tunneling: how does it look?
- **Multiphoton resonances in strong-field ionization**

Tunneling into Rydberg states

$$\Delta a_{fi}(T) \sim e^{-i \int_{t_0}^T dt'' [E_f(t'') - E_i(t'')]}$$

Initial state – ground state: $E_i = -I_p$

Final state – Rydberg state $E_f(t) = A^2(t)/2 + E_f = A^2(t)/2$

Looks just like tunneling into $p=0$. How does it happen?

-Coulomb potential traps “tunneled” electrons

$$V_{\text{drift}} = (E/\omega)\sin\omega t - \Delta V_{\text{Coul}}$$

Tunneling into Rydberg states

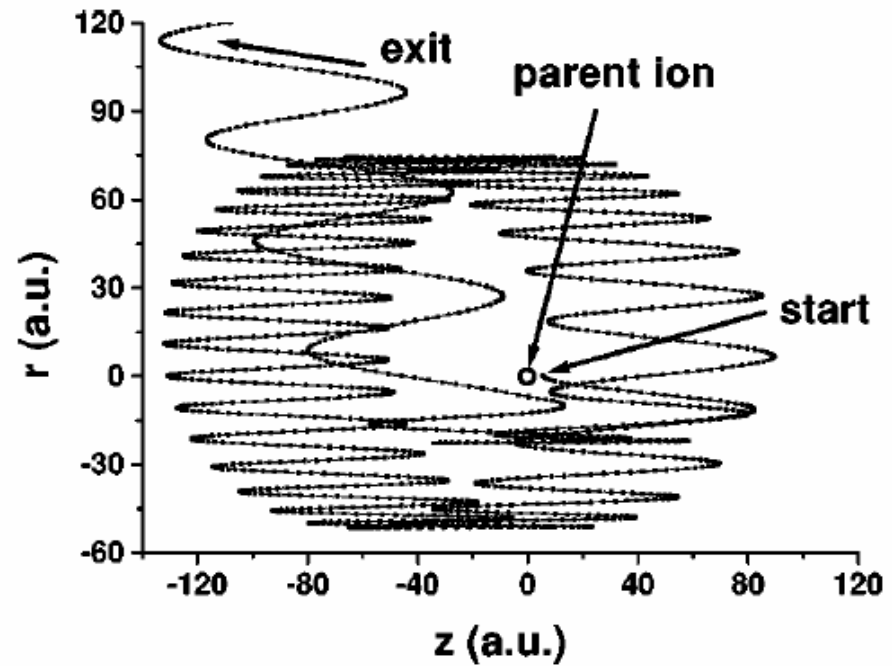
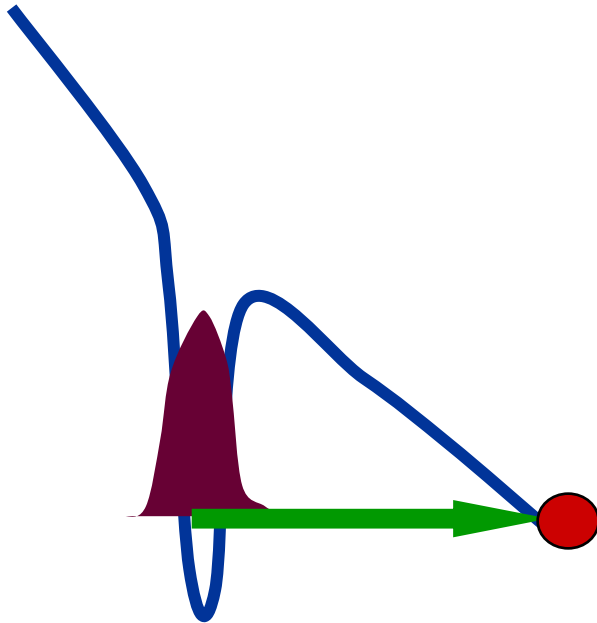
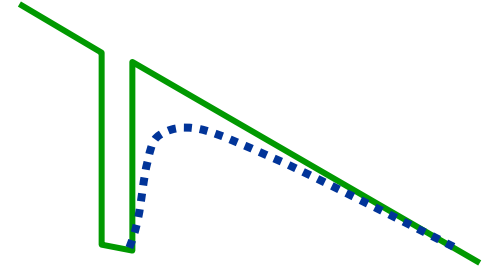


FIG. 11. Long-term trapping of an active electron into a Rydberg orbit after tunneling.

Over many periods, interference leads to resonances

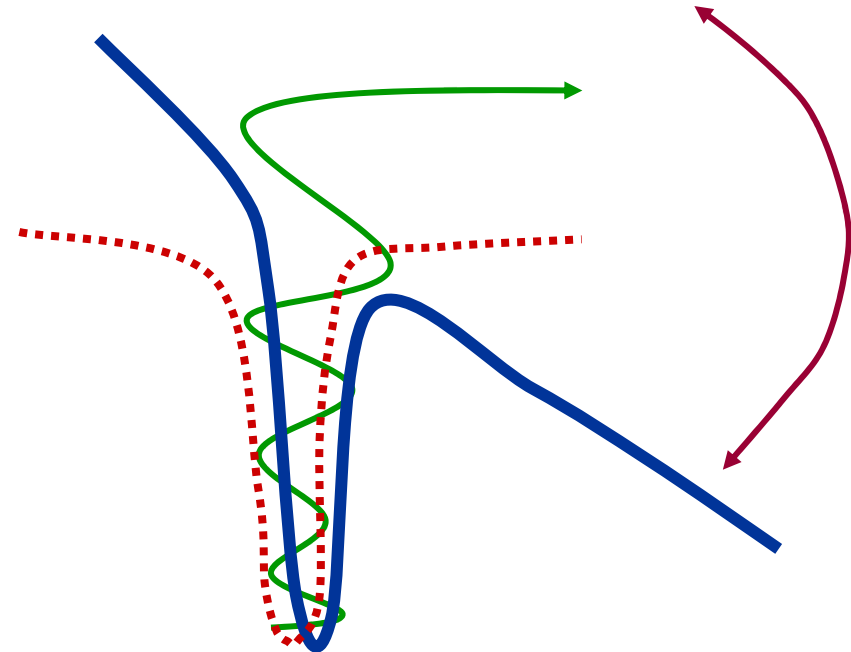
What is missing in SFA model of ionization ?

1. Coulomb effects: modify shape of the barrier



2. Stark shift and polarization

3. Classical-like heating inside the well





Adiabatic States & Non-adiabatic transitions

The Floquet states

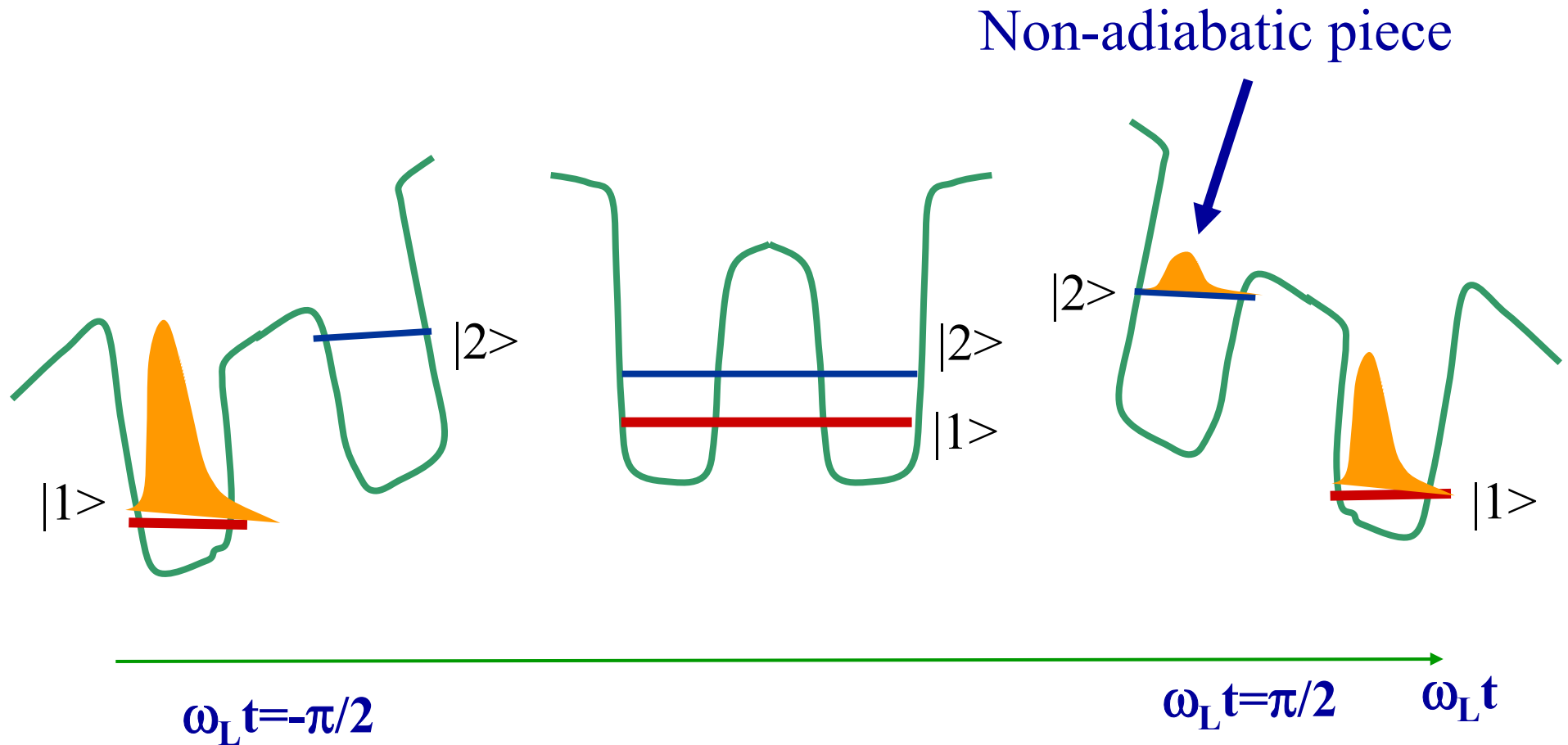
The Strong Field Approximation

The Dykhne approach. Adiabatic states and NA transitions

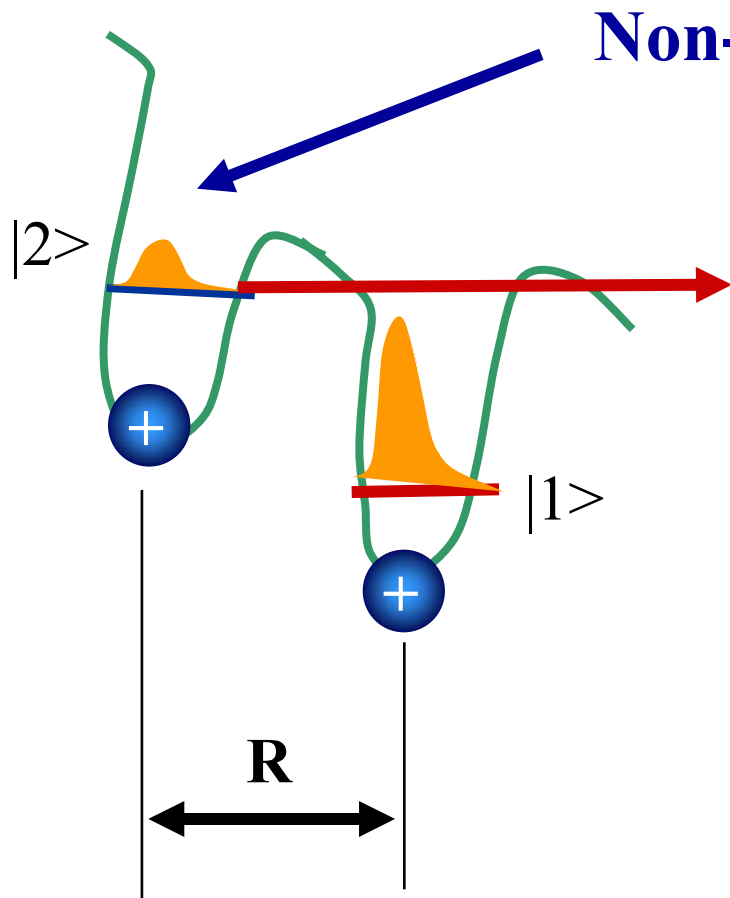
Double-well potential and enhanced ionization

Double-well potential

A model of a two-level system



Enhanced Ionization



Tunneling

$$A_{\text{ENH.ION}} \sim A_{\text{NA LOC}} A_{\text{TUNN}}$$

- EI is possible in neutral molecules too – all you need is localize electron in an up-hill state (charge transfer)
- EI is a clear example of dynamics inside the potential well

What are the main problems with SFA?

Starting from the ground state: the first part of the propagation is

$$\langle \mathbf{p}(t') | V(t') e^{-i \int_0^{t'} dt'' H_0(t'')} | \Psi_0 \rangle = e^{i \mathbf{I}_p t'} E \cos \omega t' \langle \mathbf{p}(t') | \mathbf{x} | \Psi_g \rangle$$

Then the full expression is

$$\mathbf{a}_p(\mathbf{T}) = -i \int d\mathbf{t}' e^{-i \frac{1}{2} \int_{t'}^T dt'' [p - A(\mathbf{T}) + A(\mathbf{t}'')]^2 + i \mathbf{I}_p t'} E \cos \omega t' \langle \mathbf{p}(t') | \mathbf{x} | \Psi_g \rangle$$

The integrand is very fast oscillating - look for stationary phase points

$$\int f(t') e^{iS(t')} dt' \sim f(t_0) e^{iS(t_0)} \Delta t_0 \quad \text{where } \partial S(t'=t_0) / \partial t' = 0$$

What are the main problems with SFA?

Starting from the ground state

$$\langle p(t') | V(t') e^{-i \int_0^{t'} dt'' H_0(t'')} | \Psi_0 \rangle = e^{i I_p t'} E \cos \omega t' \langle p(t') | x | \Psi_g \rangle$$

The full expression is

$$a_p(T) = -i \int dt' e^{-i \frac{1}{2} \int_{t'}^T dt'' [p - A(T) + A(t'')]^2 + i I_p t'} E \cos \omega t' \langle p(t') | x | \Psi_g \rangle$$

Main problems:

- No laser induced dynamics inside the potential well – only field-free
- No AC-Stark shift, no resonances
- Incorrect shape of the potential barrier
- Not gauge-invariant
- Could be sensitive to where the origin is

Saddle points for ionization

$$\frac{[E/\omega]^2}{2} \sin^2 \omega t' + I_p = 0$$

$t' = i\tau$ is always complex!!

$$\frac{[E/\omega]^2}{2} \operatorname{sh}^2 \omega \tau - I_p = 0 \quad \frac{I_p}{[E/\omega]^2/2} = \gamma^2 \quad \operatorname{sh}^2 \omega \tau - \gamma^2 = 0$$

For $\gamma \ll 1$

$\operatorname{sh} \omega \tau \sim \omega \tau$

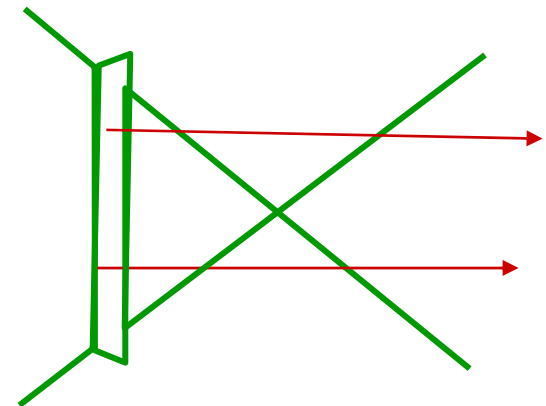
$$\omega \tau = \gamma, \quad \tau = \gamma/\omega = \sqrt{2I_p}/E$$

For $\gamma \gg 1$

$\operatorname{sh} \omega \tau \sim \exp[\omega \tau]/2$

$$\omega \tau = \ln 2\gamma,$$

τ – the time spent in classically forbidden region – changes with γ



Adiabatic States

The Schroedinger equation is

$$i\partial_t \Psi = [H_0 + V(t)]\Psi$$

Suppose $V(t)$ is very slow. Adiabatic states Φ_n are given by:

$$[H_0 + V(t)]\Phi_n(t) = E_n(t)\Phi_n(t)$$

Adiabatic approximation: system starting in $|n\rangle$ evolves as

$$\Psi(T) = e^{-i \int dt E_n(t)} \Phi_n(T)$$

Non-adiabatic transitions

Adiabatic state

$$\Psi(T) = e^{-i \int^T dt E_n(t)} \Phi_n(T)$$

is NOT a solution of

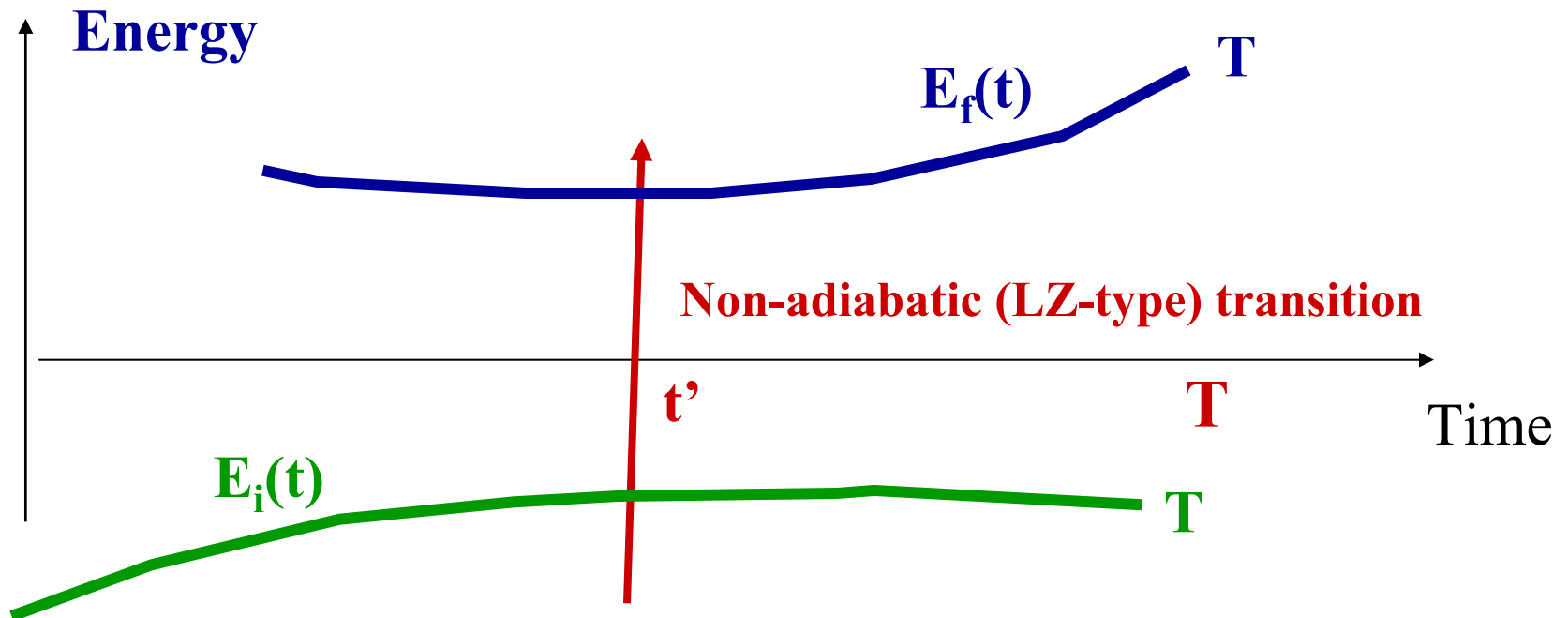
$$i\partial_t \Psi = [H_0 + V(t)]\Psi$$

because of

$$\partial_t \Phi_n(t) \neq 0$$

Non-adiabatic transitions

$$a_{fi}(T) \sim \int dt' e^{-i \int_{t'}^T dt'' E_f(t'')} \langle \Phi_f | \partial_t \Phi_i \rangle (t') e^{-i \int_{t'}^{t'} dt'' E_i(t'')}$$



Very similar to S-matrix, but the laser field affects both states!

The Dykhne result

$$\mathbf{a}_{fi}(\mathbf{T}) \sim \int dt' e^{-i \int_{t'}^{\mathbf{T}} dt'' E_f(t'')} \langle \Phi_f | \partial_t \Phi_i \rangle e^{-i \int_{t_0}^{t'} dt'' E_i(t'')}$$

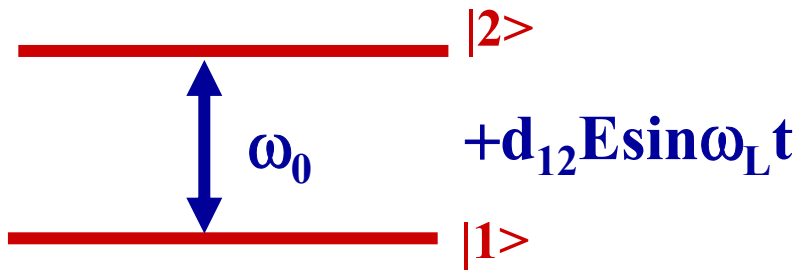
Use stationary phase method: *derivative (phase) w.r.t. $t'=0$!*

$$E_f(t') - E_i(t') = 0 \rightarrow t_0$$

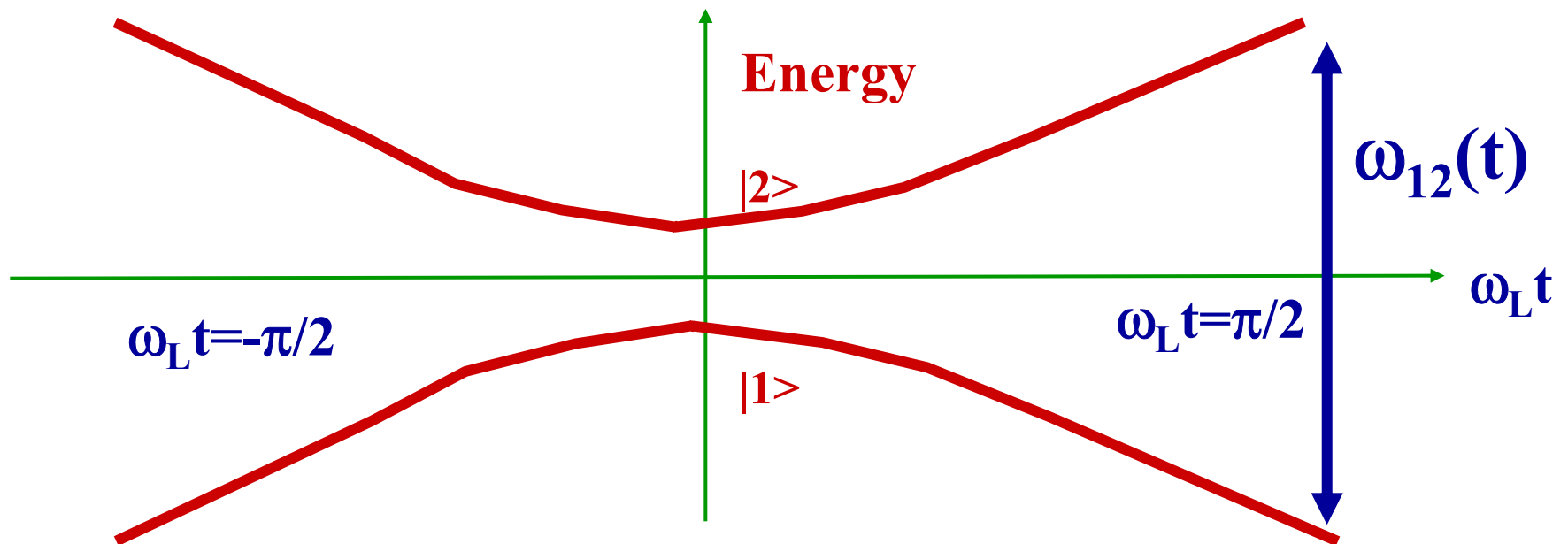
$$\Delta \mathbf{a}_{fi}(\mathbf{T}) \sim e^{-i \int_{t_0}^{\mathbf{T}} dt'' [E_f(t'') - E_i(t'')]}$$

- the Dykhne result – just like in tunneling

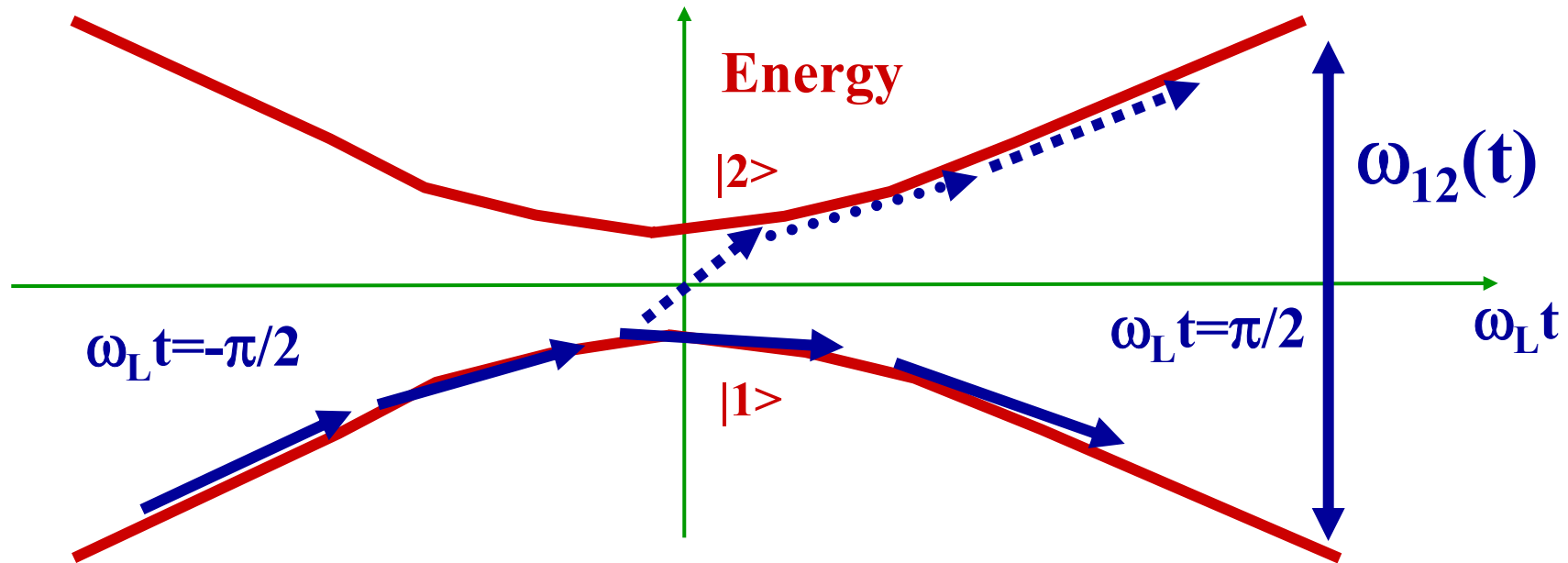
The two-level system



Adiabatic approximation:
treat $\omega_L t$ as a parameter



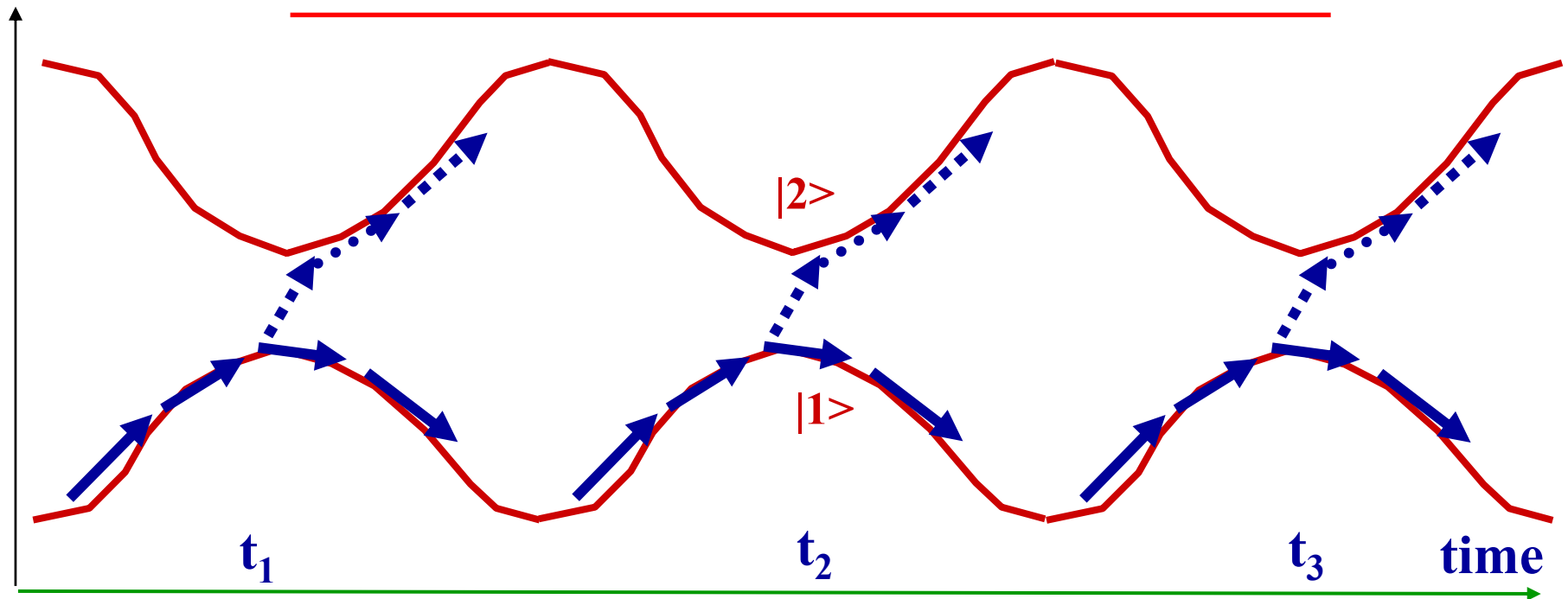
Non-adiabatic transitions



$$\omega_{12}(t) = \sqrt{\omega_0^2 + (2d_{12}E \sin \omega t)^2} = 0$$

Again complex time: $t=i\tau$ $\omega_0^2 - (2d_{12}E \operatorname{sh} \omega \tau)^2 = 0$

Sub-cycle NA transitions & Resonances



Multiphoton resonance means constructive interference of non-adiabatic transitions at $t_1, t_2, t_3 \dots$

$$\int_{t_1}^{t_2} dt [E_2(t) - E_1(t)] = (2n+1)\pi$$