MAX BORN-INSTITUTE

Electron recollision and High Harmonic Generation

Tutorial

Attofel summer school 3 May 2011, Crete

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HHG: How to generate them fast?

- S-matrix expression for HHG dipole (one electron)
- Stationary phase method and factorization of the HHG dipole (ionization, propagation, recombination)

- Stationary phase equations for HHG:

  The Lewenstein model

- The classical 3-step photoelectron model: where it goes wrong and how it can be improved

- HHG dipole for many electrons, including laser-induced dynamics in the ionic core between ionization and recombination

HHG: the non-linear optics perspective

HHG is frequency up-conversion. It results from macroscopic response of the medium:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2}$$

The response is described by polarization **P(t,z)** induced in the medium:

- response of atoms/molecules
- response of free electrons
- guiding medium (e.g. hollow core fiber)
- etc

This lecture is about P(t) from individual atoms/molecules

HHG: the non-linear optics perspective

This lecture is about P(t) from individual atoms/molecules

$$P(t) = nD(t)$$

n- number density

$$D(t) = \left\langle \Psi(\vec{r},t) \middle| \hat{d} \middle| \Psi(\vec{r},t) \right\rangle$$

$$i\frac{\partial \Psi(\vec{r},t)}{\partial t} = H(t)\Psi(\vec{r},t)$$

The key is to find the wavefunction

The S-matrix expressions (one electron)

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t}=\hat{H}(t)\Psi(\vec{r},t)$$

$$\hat{H}(t)=\hat{H}_0+\hat{V}_L(t)$$

The hard part

Easy part

Exact:

$$\Psi(\vec{r},t)=e^{-i\hat{H}_0(t-t_0)}\Psi_g(\vec{r})-i\int_{t_0}^{t}dt'\,e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau}V_L(t')e^{-i\hat{H}_0(t'-t_0)}\Psi_g(\vec{r})$$

The S-matrix expressions (one electron)

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t}=\hat{H}(t)\Psi(\vec{r},t)$$

$$\hat{H}(t)=\hat{H}_0+\hat{V}_L(t)$$

The SFA:

Neglect the Coulomb potential

$$\Psi(\vec{r},t)=\underbrace{e^{i\hat{H}_0(t-t_0)}\Psi_g(\vec{r})}_{\text{Bound part}}-\underbrace{i\int_{t_0}^{t}dt'\,\boxed{e^{-i\int_{t'}^{t}\hat{H}_{LAS}(\tau)d\tau}}\,V_L(t')e^{-i\hat{H}_0(t'-t_0)}\Psi_g(\vec{r})}_{\text{Continuum part}}$$

S-matrix expression for HHG dipole (one electron)

$$D(t) = \left\langle \Psi(\vec{r},t) \middle| \hat{d} \middle| \Psi(\vec{r},t) \right\rangle$$

$$D(t) \approx -i \underbrace{\left\langle \Psi_g(\vec{r}) e^{i\hat{H}_0(t-t_0)} \right|}_{\text{Bound part}} \underbrace{\hat{d} \int_{t_0}^{t} dt' e^{-i\int_{t'}^{t} \hat{H}_{LAS}(\tau)d\tau} V_L(t') e^{-i\hat{H}_0(t'-t_0)} \left| \Psi_g(\vec{r}) \right\rangle}_{\text{Continuum part}} + c.c.$$

S-matrix expression for HHG dipole (one electron)

$$D(t) = \langle \Psi(\vec{r},t) | \hat{d} | \Psi(\vec{r},t) \rangle$$

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$$1 = \int_{-\infty}^{+\infty} d\vec{k} \left| \vec{k} + \vec{A}(t') \right\rangle \left\langle \vec{k} + \vec{A}(t') \right|$$

Volkov functions, the length gauge:

$$e^{-i\int_{t'}^{t} \hat{H}_{LAS}(\tau)d\tau} \left| \vec{k} + \vec{A}(t') \right\rangle = e^{-i\frac{1}{2}\int_{t'}^{t} [k+A(\tau)]^2 d\tau} \left| \vec{k} + \vec{A}(t) \right\rangle$$

S-matrix expression for HHG dipole (one electron)

$$D(t) = -i\int_{-\infty}^{+\infty} d\vec{k}\int_{t_0}^{t} dt'\, d^*\left(\vec{k}+\vec{A}(t)\right)e^{-iS(t,t',k)}F_L(t')d\left(\vec{k}+\vec{A}(t')\right)+c.c.$$

recombination

Ionization? –Not yet!

Phase (action)

$$S(t,t',k) = \frac{1}{2}\int_{t'}^{t}\left(\vec{k}+\vec{A}(\tau)\right)^2 d\tau + I_p\left(t-t'\right)$$

Lewenstein et al, 1994

S-matrix expression for HHG dipole (one electron)

$$D(t) = -i \int_{-\infty}^{+\infty} d\vec{k} \int_{t_0}^{t} dt' \, d^*\left(\vec{k}+\vec{A}(t)\right) e^{-iS(t,t',k)} F_L(t') d\left(\vec{k}+\vec{A}(t')\right) + c.c.$$

S (t,t′,k) is large, the integrand is a highly oscillating function

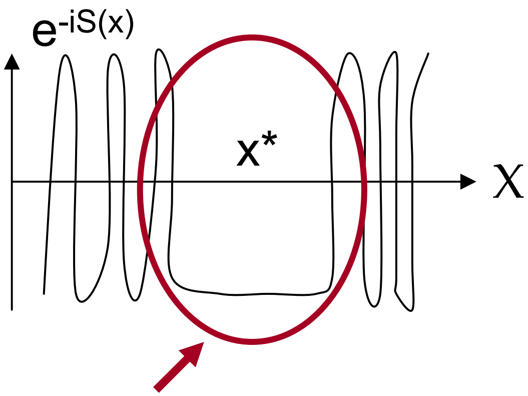
Evaluate D(t)?

- Numerically – be careful to take care of highly oscillating integrands
- Analytically – use highly oscillating integrands to your advantage

  -Analytic approach supplies:

  - “time-energy mapping”, important for attosecond imaging
  - approximate picture of HHG as a 3-step process involving ionization, propagation, recombination
  - <u>extension beyond SFA and single electron!</u>

Stationary phase method



Stationary phase region $1/\sqrt{S''}$

$$\int_a^b dx\, f(x) e^{-i\lambda S(x)} \qquad \lambda >> 1$$

The integral is accumulated in points x*, where phase is stationary:

$$S'(x^*) = 0$$

Idea:

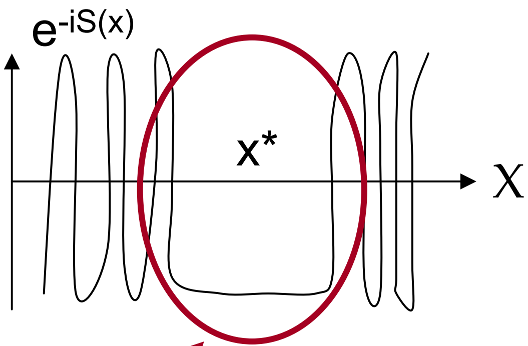
$$S(x) = S(x^*) + S'(x^*)(x - x^*) + S''(x^*)\frac{(x - x^*)^2}{2}$$

$$\int_a^b dx\, f(x) e^{-i\lambda S(x)} \approx f(x^*) e^{-i\lambda S(x^*)} \int_{-\infty}^{+\infty} dx\, e^{-i\lambda S''(x^*)\frac{(x - x^*)^2}{2}}$$

Can be evaluated analytically

Stationary phase method

(see Mathews, Walker, Mathematical Physics)



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$$\int_a^b dx\, f(x) e^{-i\lambda S(x)} = \sqrt{\frac{2\pi}{\lambda |S''(x^*)|}}\left[f(x^*) + O\left(\lambda^{-1}\right)\right] e^{-i\lambda S(x^*) + \frac{i\pi}{4} sign(S''(x^*))}$$

$$\int_\gamma dx\, f(z) e^{-i\lambda S(z)}$$

For contour integrals in complex plane a similar idea leads to the saddle point method

Saddle point method for HHG dipole

Harmonic spectrum: $$D(N\omega) \propto \int_{-\infty}^{+\infty} dt \int_{t_0}^{t} dt' \int_{-\infty}^{+\infty} d\vec{k}\, e^{-iS(t,t',k)} e^{iN\omega t}$$

Phase=S(t,t',k)-Nω must be stationary wrt t,t',k

$$S(t,t',k) = \frac{1}{2}\int_{t'}^{t} \left(\vec{k} + \vec{A}(\tau)\right)^2 d\tau + I_p\left(t - t'\right)$$

1) $\dfrac{\partial S}{\partial t'} = 0$

2) $\dfrac{\partial S}{\partial k_{\parallel}} = 0 \quad \dfrac{\partial S}{\partial k_{\perp}} = 0$

3) $\dfrac{\partial S}{\partial t} = N\omega$

Ionization time t_i

Canonical momentum k_s

Recombination time t_r

If we know t_i, t_r, k_s, we know D(Nω)

Lewenstein et al, 1994

Results of saddle point integration

Now we need to find k_s, t_r, t_i

$$\frac{\partial S}{\partial t'} = \frac{1}{2}\left(\vec{k}_s + \vec{A}(t_i)\right)^2 + I_p = 0$$

ionization

$$\frac{\partial S}{\partial k_{\parallel}} = \int_{t_i}^{t_r} \left(k_{\parallel} + A(\tau)\right) d\tau = 0$$

$$\frac{\partial S}{\partial k_{\perp}} = k_{\perp}(t_r - t_i) = 0 \qquad k_{\perp} = 0$$

return

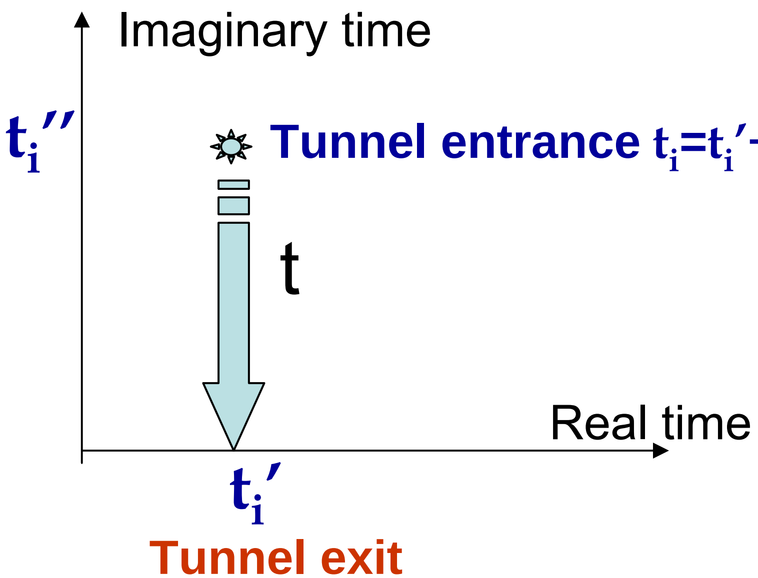
$$\frac{\partial S}{\partial t} = \frac{1}{2}\left(\vec{k}_s + \vec{A}(t_r)\right)^2 + I_p = N\omega$$

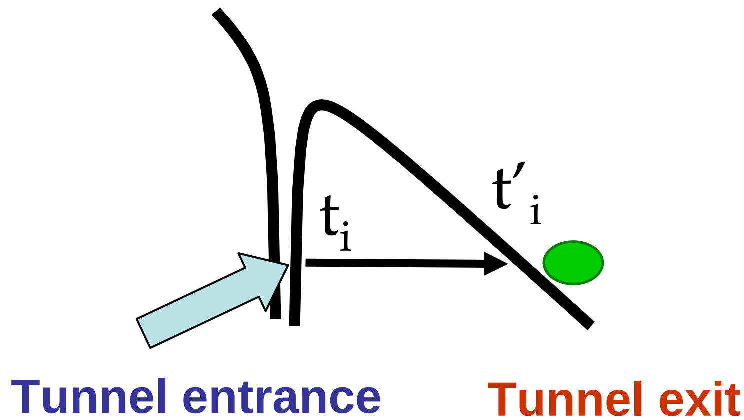
recombination

Complex ionization time

1) $$\frac{1}{2}\left(k_s + A(t_i)\right)^2 + I_p = 0$$

Ionization time t_i is complex $t_i = t_i' + i\, t_i''$





$$z_{ex} = i\int_{t_i''}^{0} d\tau\left(k_s + A(t_i' + i\tau)\right) = z'_{ex} + iz''_{ex}$$

The coordinate at the exit is complex

Recombination time and canonical momentum

2) Return

$$\int_{t_i}^{t_r} \left(k_s + A(\tau)\right) d\tau = 0$$

Displacement between entering the barrier (start of ionization) and recombination should be zero

Imaginary displacement "under the barrier" must be compensated: **t_r is complex**

3) Energy conservation

$$\frac{1}{2}\left(k_s + A(t_r)\right)^2 + I_p = N\omega$$

Energy conservation dictates that electron velocity at the time of recombination is real

Since recombination time t_r is complex, **canonical momentum k_s is also complex**

In general, k_s, t_r, t_i are all complex. Only the observable – the photon energy $N\omega$ – is real

Quantum orbits (Salieres et al, 2000)

How to solve 3 saddle point equations? Total 6 unknowns: $t_i', t_i'', t_r', t_r'', k_s', k_s''$

$$\frac{1}{2}\left(\vec{k}_s + \vec{A}(t_i)\right)^2 + I_p = 0 \qquad \int_{t_i}^{t_r}\left(k_s + A(\tau)\right)d\tau = 0 \qquad \frac{1}{2}\left(k_s + A(t_r)\right)^2 + I_p = N\omega$$

Total 6 equations: for real and imaginary parts.

Goal: Set N $\rightarrow$ Find $t_i', t_i'', t_r', t_r'', k_s', k_s''$

All 6 eqs. do not have analytical solutions.

Step 1: express everything via return time (imaginary and real), using 4 eqs.
Step 2: solve the remaining 2 equations together

Solving the saddle point equations for Nω>Ip

Specify the field: $F(t) = F_0 \cos(\omega t)$ $\quad A(t) = -\dfrac{F_0}{\omega}\sin\omega t = -v_0 \sin\omega t$

Dimensionless variables:

$$k_1 = \frac{k'_s}{v_0} \qquad k_2 = \frac{k_s''}{v_0} \qquad \gamma^2 = \frac{2I_p\omega^2}{F^2} = \frac{I_p}{2U_p}$$

$$\varphi_r = \omega t_r \qquad \varphi_i = \omega t_i \qquad \gamma_N^2 = \frac{N\omega - I_p}{2U_p}$$

Step 1 a.

$$\left.\frac{1}{2}\left(\vec{k}_s + \vec{A}(t_r)\right)^2 = N\omega - I_p \right\}$$

Real part

Imaginary part

$$k_1 = \cosh\varphi''_r \sin\varphi'_r + \gamma_N$$

$$k_2 = \sinh\varphi''_r \cos\varphi'_r$$

For each N we have expressed k_2 and k_1 via φ_r' , φ_r''

Solutions of the saddle point equations

Step 1 b.

$$\frac{1}{2}\left(k_s + A(t_i)\right)^2 + I_p = 0$$

Real part

$$\sin\varphi'_i \cosh\varphi''_i = k_1$$

Imaginary part

$$\sinh\varphi''_i \cos\varphi'_i = \gamma + k_2$$

$$\varphi'_i = \arcsin\left(\sqrt{(P+D)/2}\right) \qquad P = k_1^{\,2} + \tilde{\gamma}^2 + 1 \qquad \tilde{\gamma} = \gamma + k_2$$

$$\varphi''_i = \operatorname{arccosh}\left(\sqrt{(P-D)/2}\right) \qquad D = \sqrt{P^2 - 4k_1^{\,2}}$$

For each N we have expressed k_2 and k_1 via φ_r' , φ_r'' and we have expressed φ_i' , φ_i'' via k_1, k_2

Hence φ_i' , φ_i'' and k_1, k_2 are all expressed via φ_r' , φ_r''

Now we can use the remaining equations to find φ_r' , φ_r''

Solutions of the saddle point equations

Step 2: Now use the last equation:

$$\int_{t_i}^{t_r} \left(k_s + A(\tau)\right) d\tau = 0$$

Real part $\quad F_1 = k_1\left(\varphi'_r - \varphi'_i\right) - k_2\left(\varphi''_r - \varphi''_i\right) - \cos\varphi'_i \cosh\varphi''_i + \cosh\varphi''_r \cos\varphi'_r = 0$

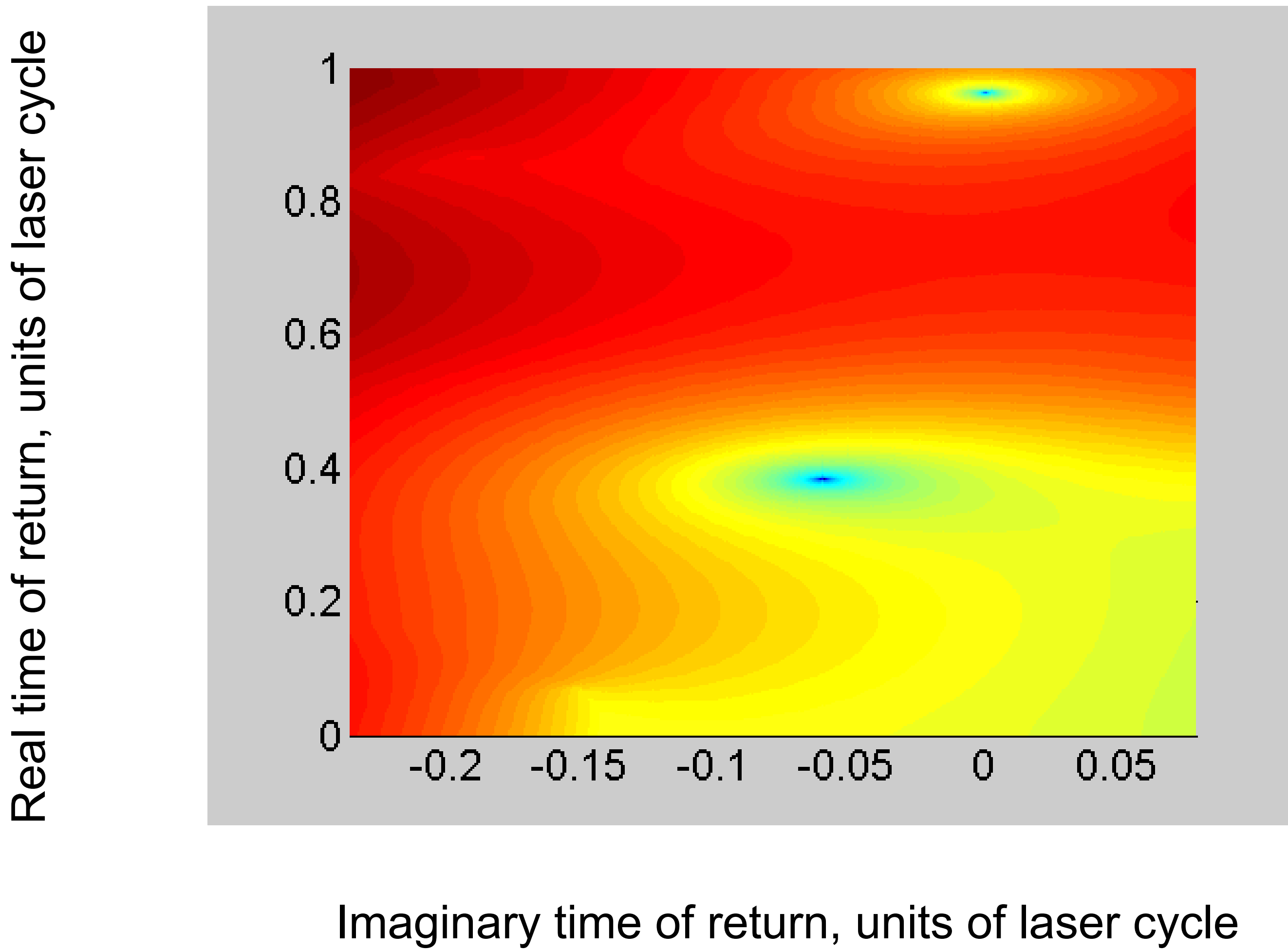
Im part $\quad F_2 = k_1\left(\varphi''_r - \varphi''_i\right) + k_2\left(\varphi'_r - \varphi'_i\right) + \sin\varphi'_i \sinh\varphi''_i - \sinh\varphi''_r \sin\varphi'_r = 0$

$$\left[F_1\left(N, \varphi'_r, \varphi''_r\right)\right]^2 + \left[F_2\left(N, \varphi'_r, \varphi''_r\right)\right]^2 = 0$$

Set grid of φ_r' and φ_r'' and numerically find minimum of this surface for each N

Solutions

Harmonic 11

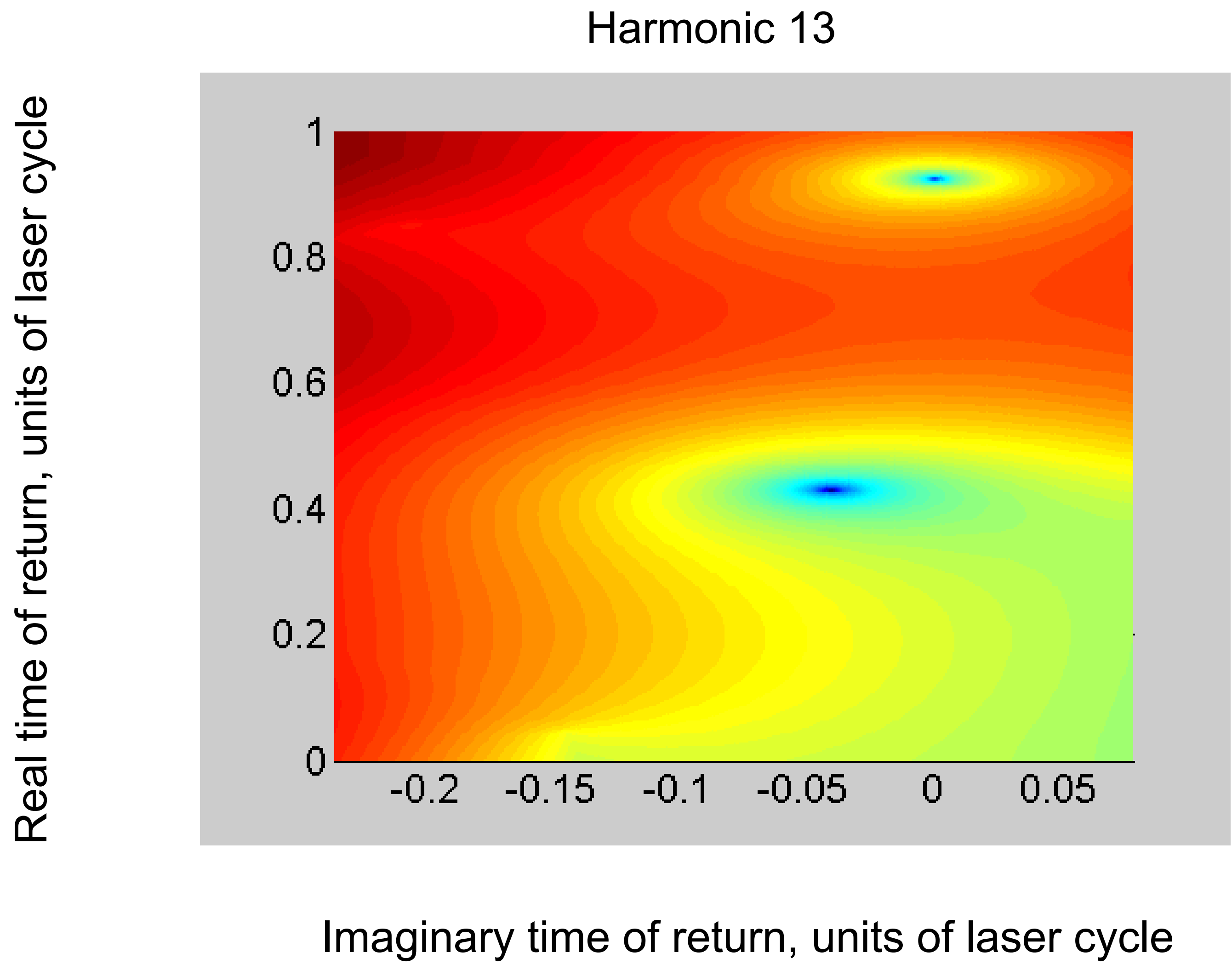


I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions



I_p=15.6 eV

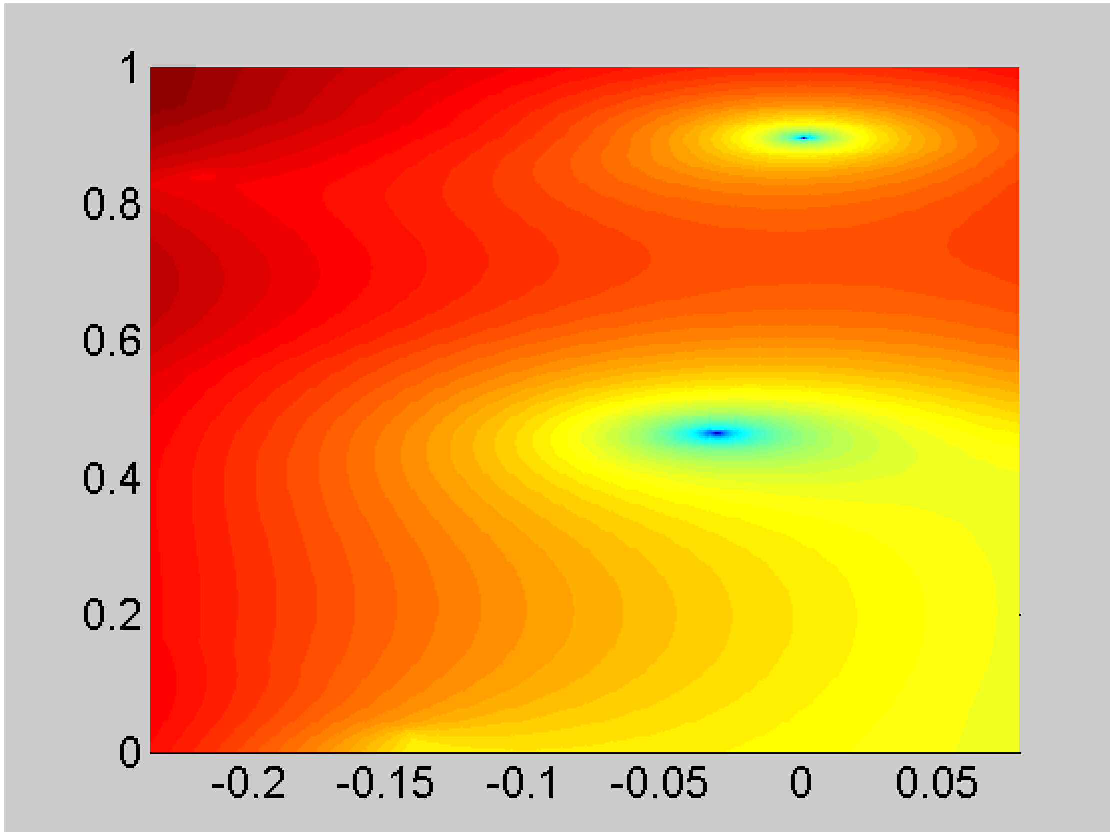
I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions

Harmonic 15

Real time of return, units of laser cycle



I_p=15.6 eV

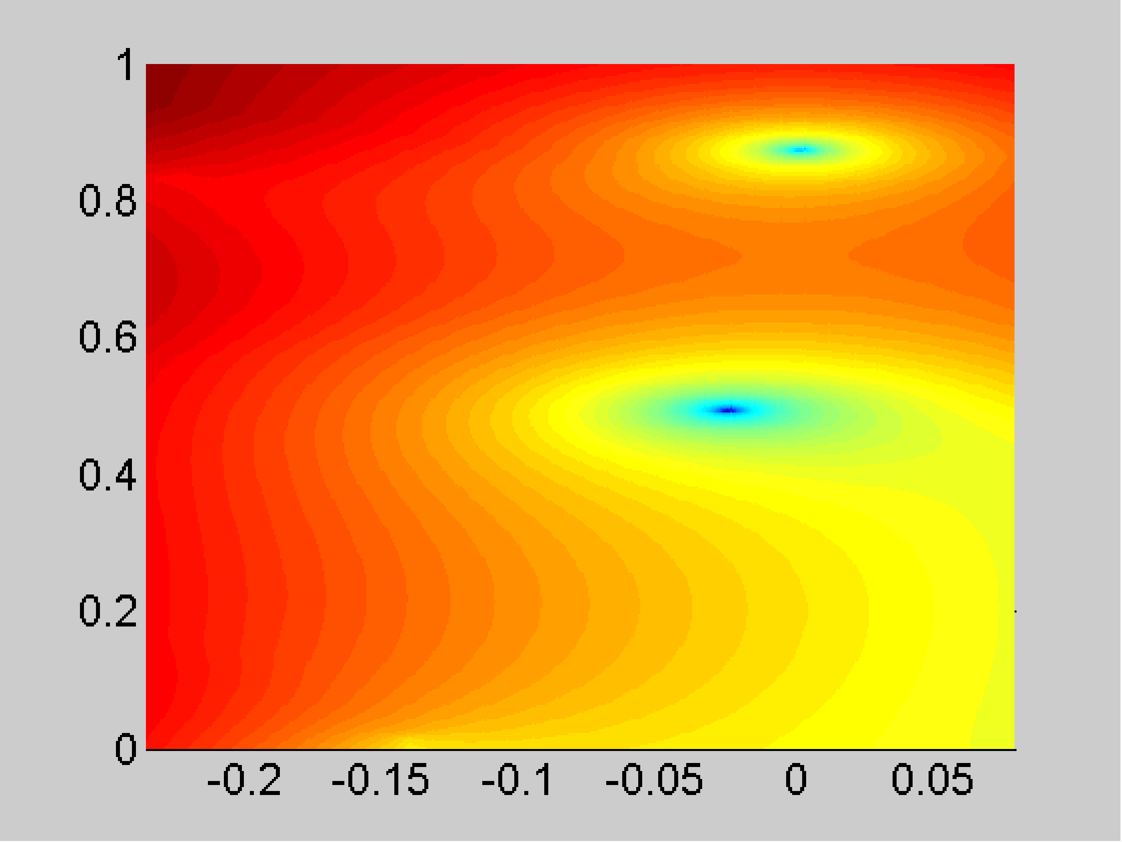
I=1.3 10^{14}W/cm^2

ω=1.5 eV

Imaginary time of return, units of laser cycle

Solutions

Harmonic 17



Real time of return, units of laser cycle

Imaginary time of return, units of laser cycle

I_p=15.6 eV

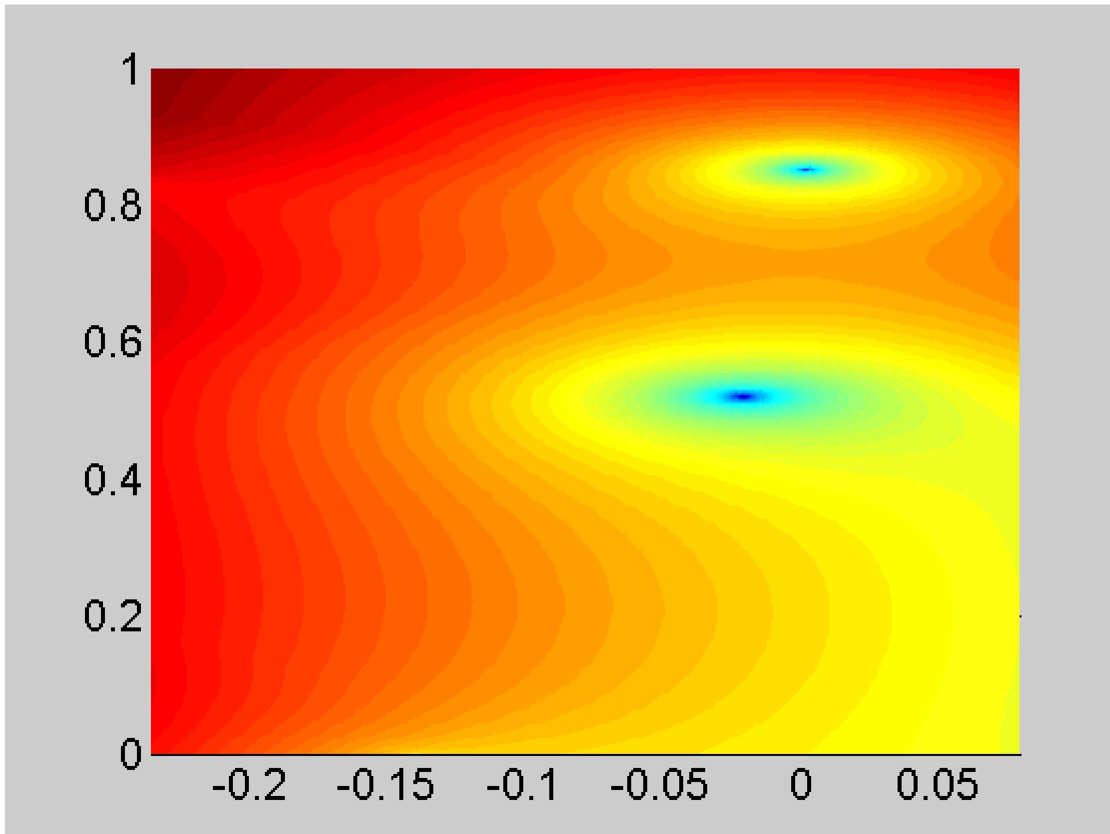
I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions

Harmonic 19

Real time of return, units of laser cycle



I_p=15.6 eV

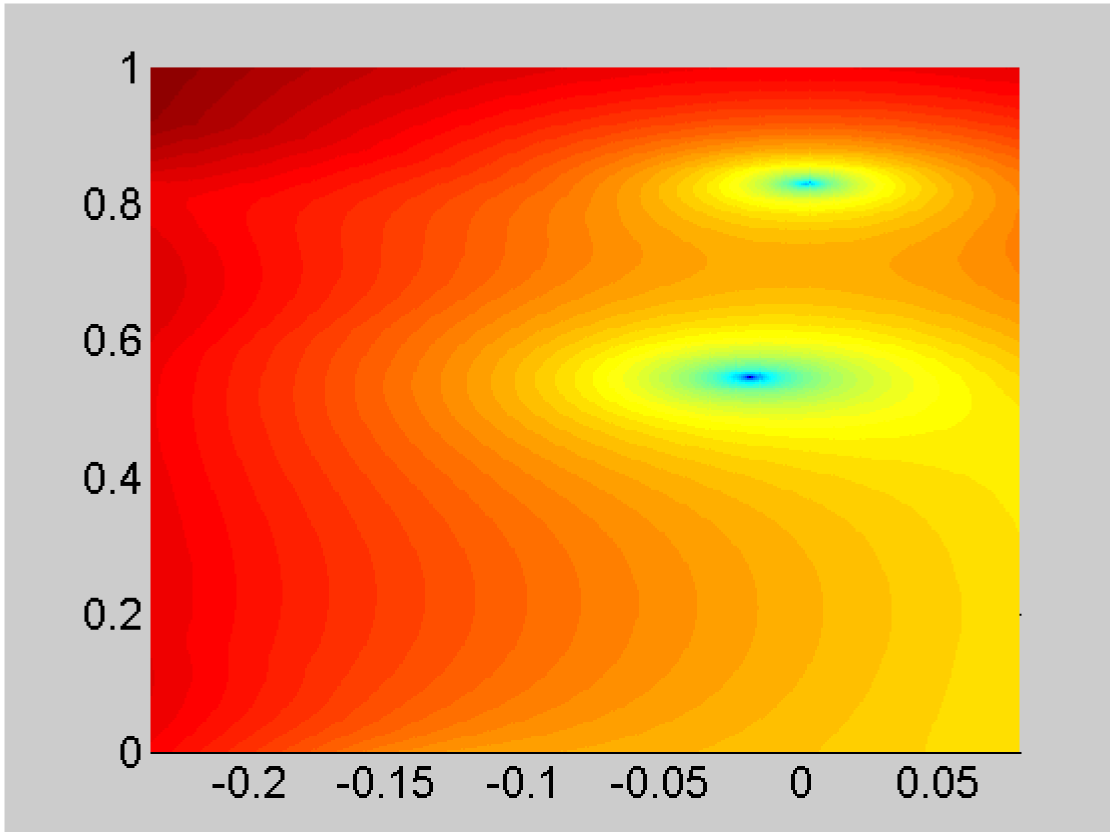
I=1.3 10^{14}W/cm^2

ω=1.5 eV

Imaginary time of return, units of laser cycle

Solutions

Harmonic 21



Real time of return, units of laser cycle

Imaginary time of return, units of laser cycle

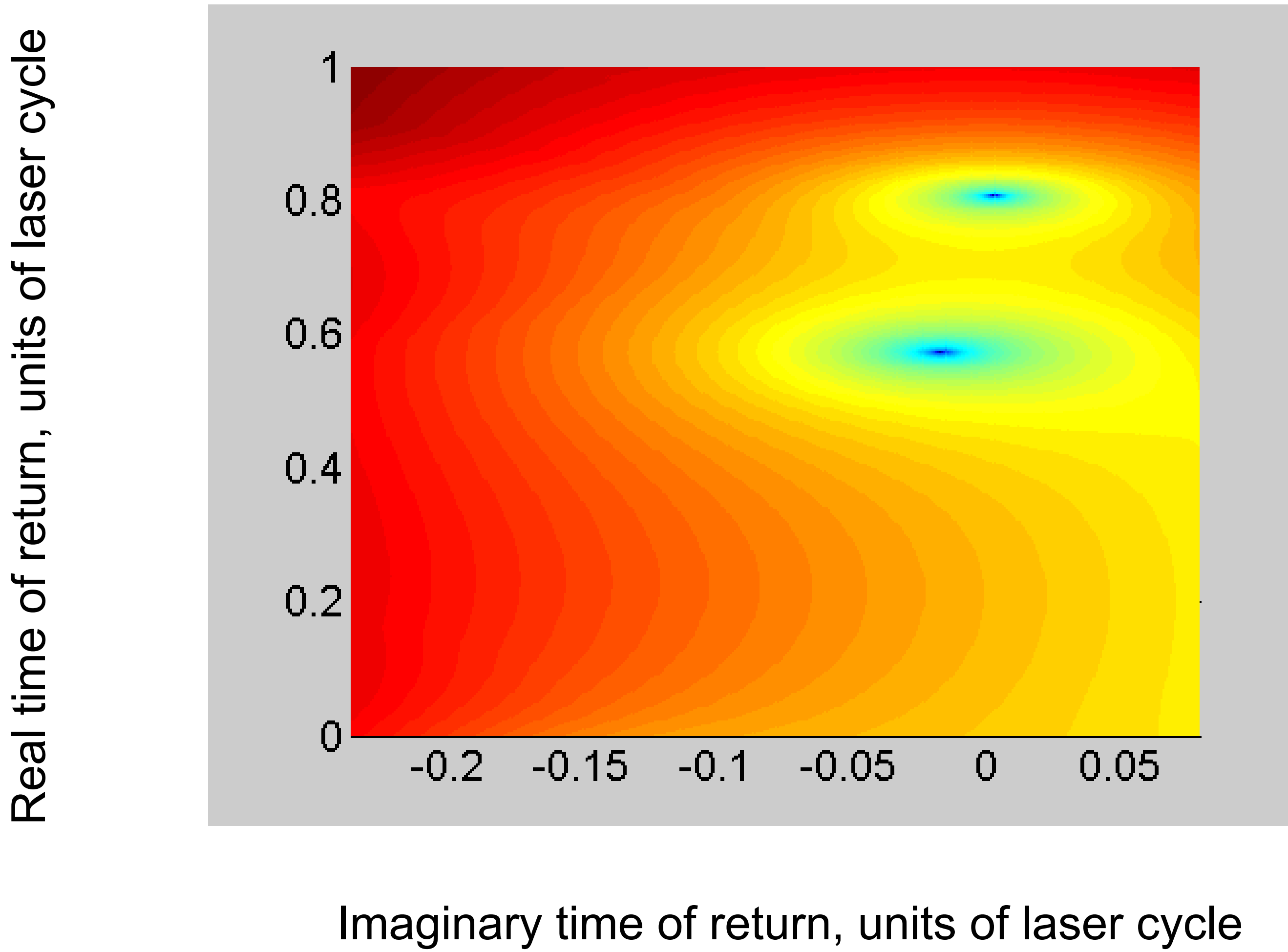
I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions

Harmonic 23



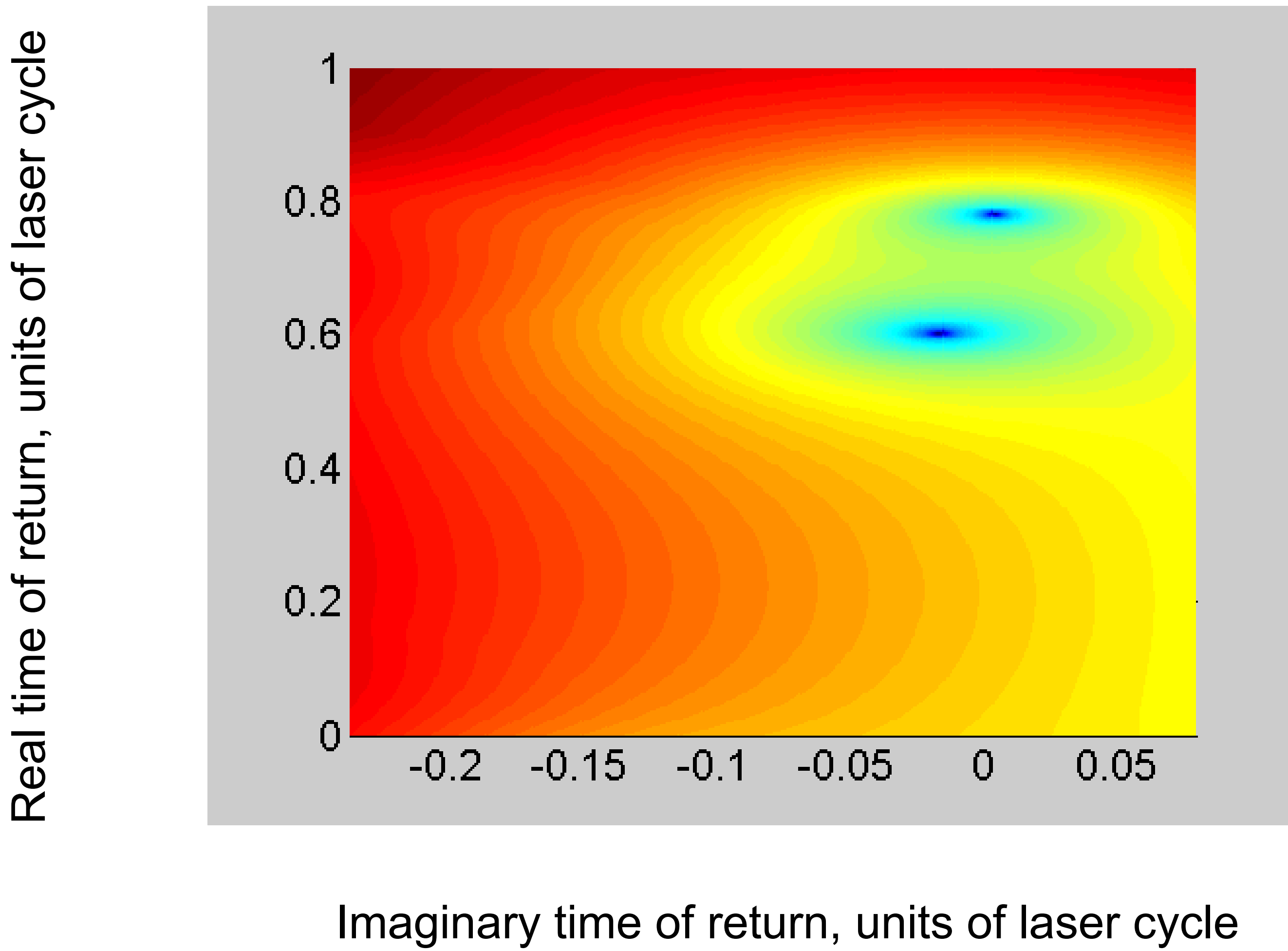
I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions

Harmonic 25



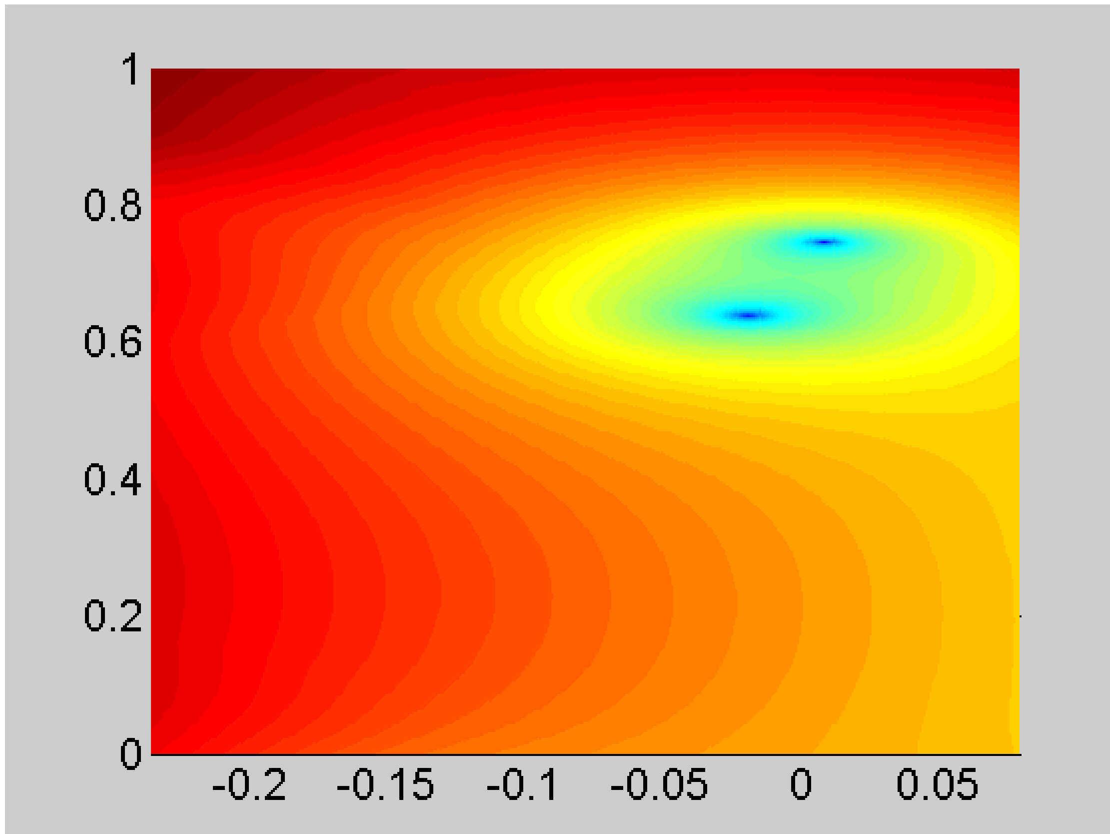
I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions

Harmonic 27



Real time of return, units of laser cycle

Imaginary time of return, units of laser cycle

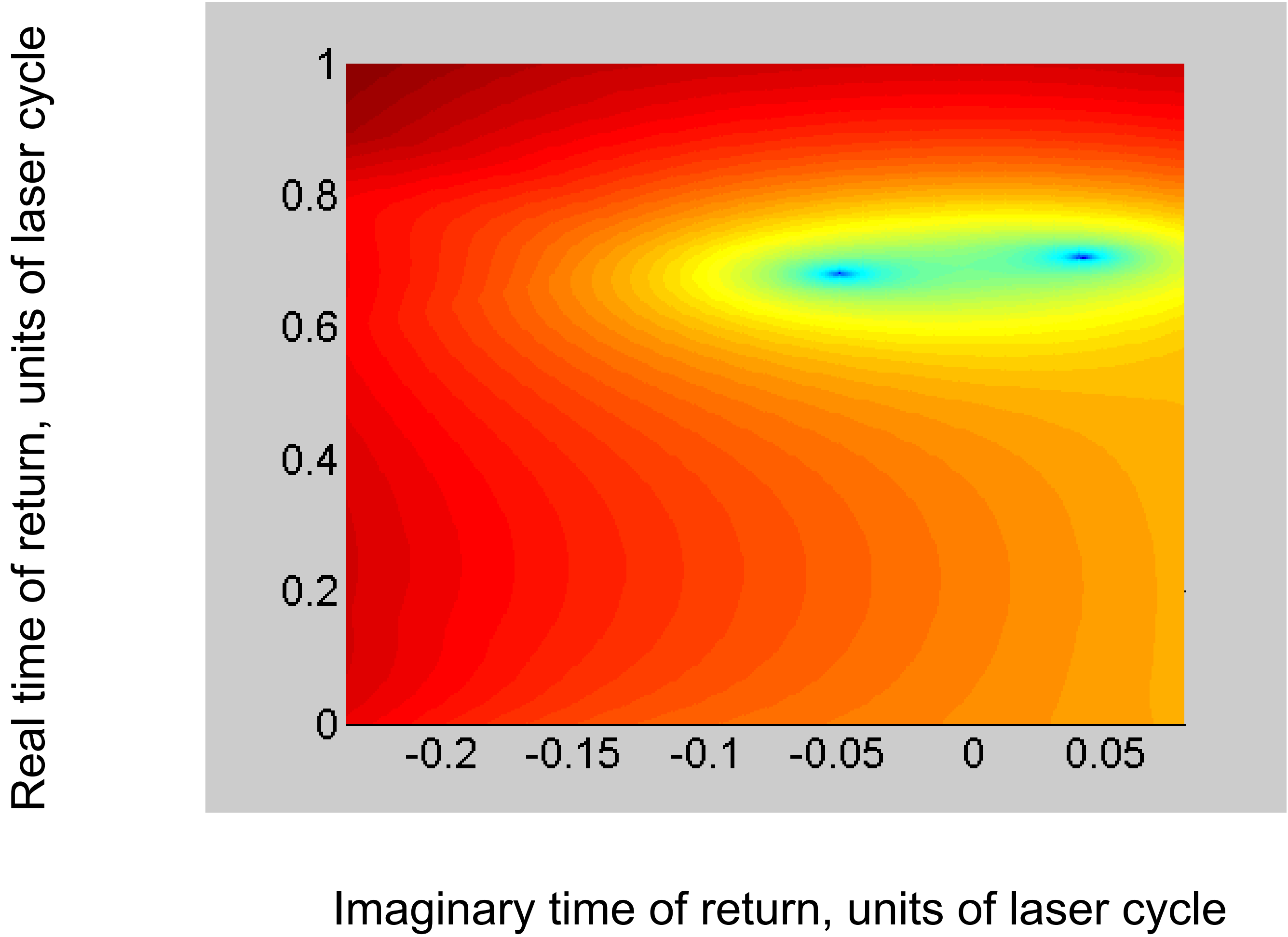
I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions

Harmonic 29

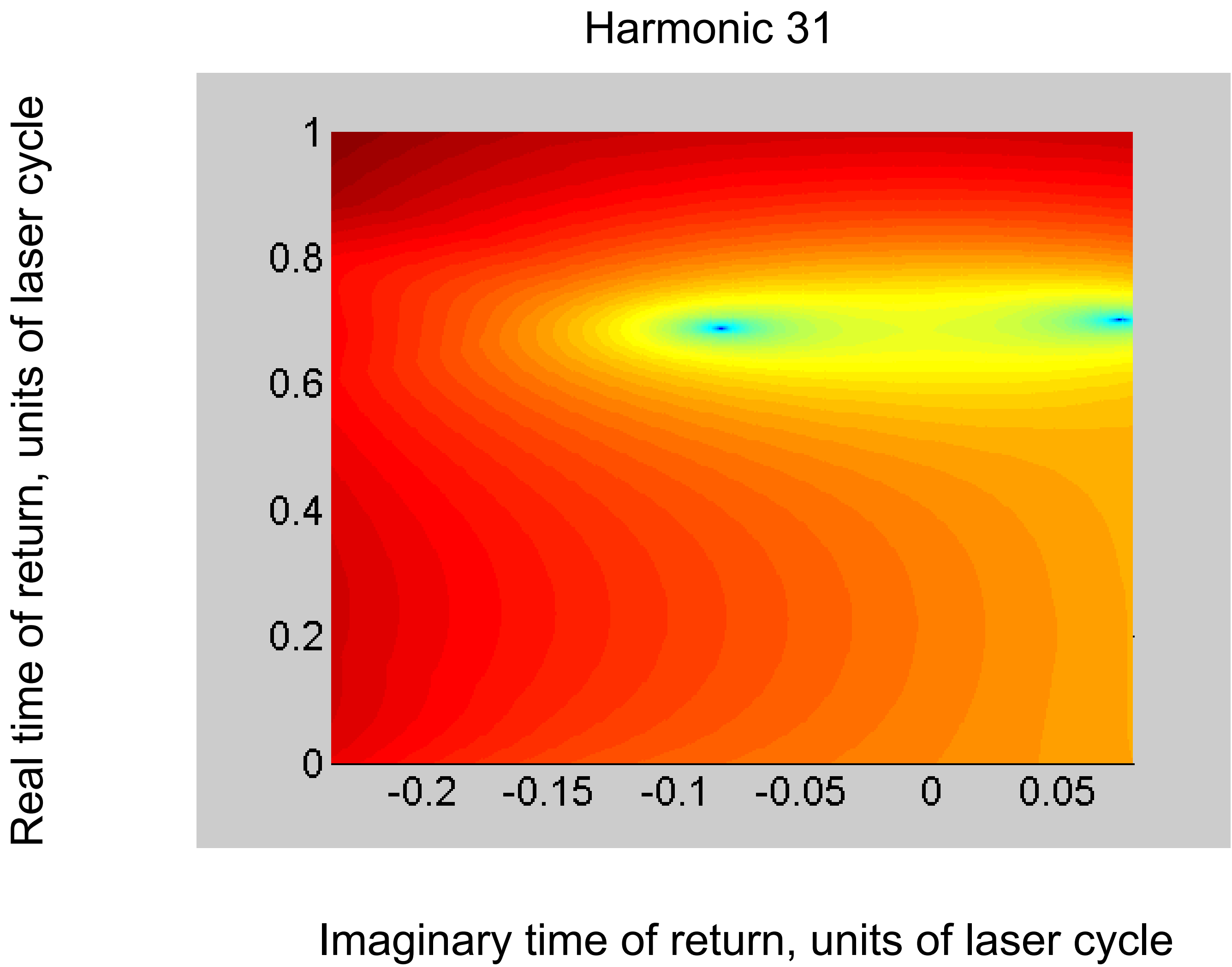


I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Solutions



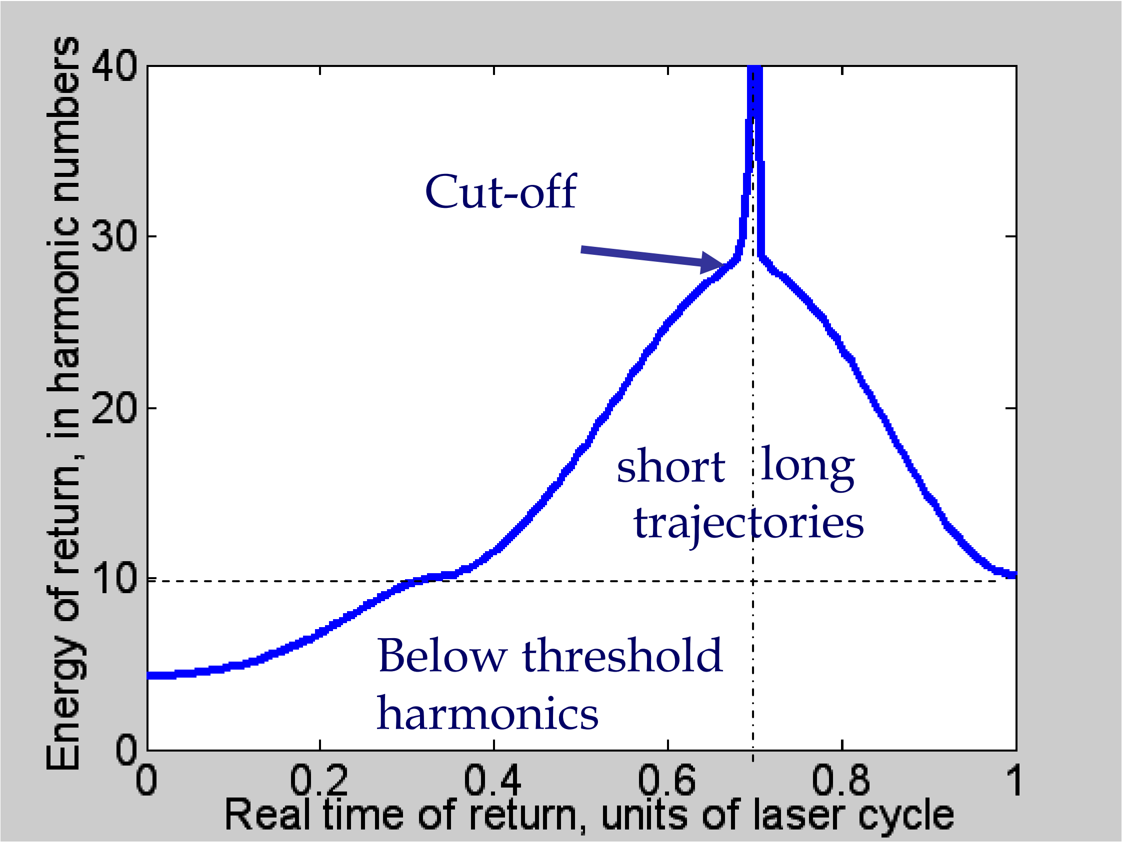
I_p=15.6 eV

I=1.3 10^{14}W/cm^2

ω=1.5 eV

Energy of return

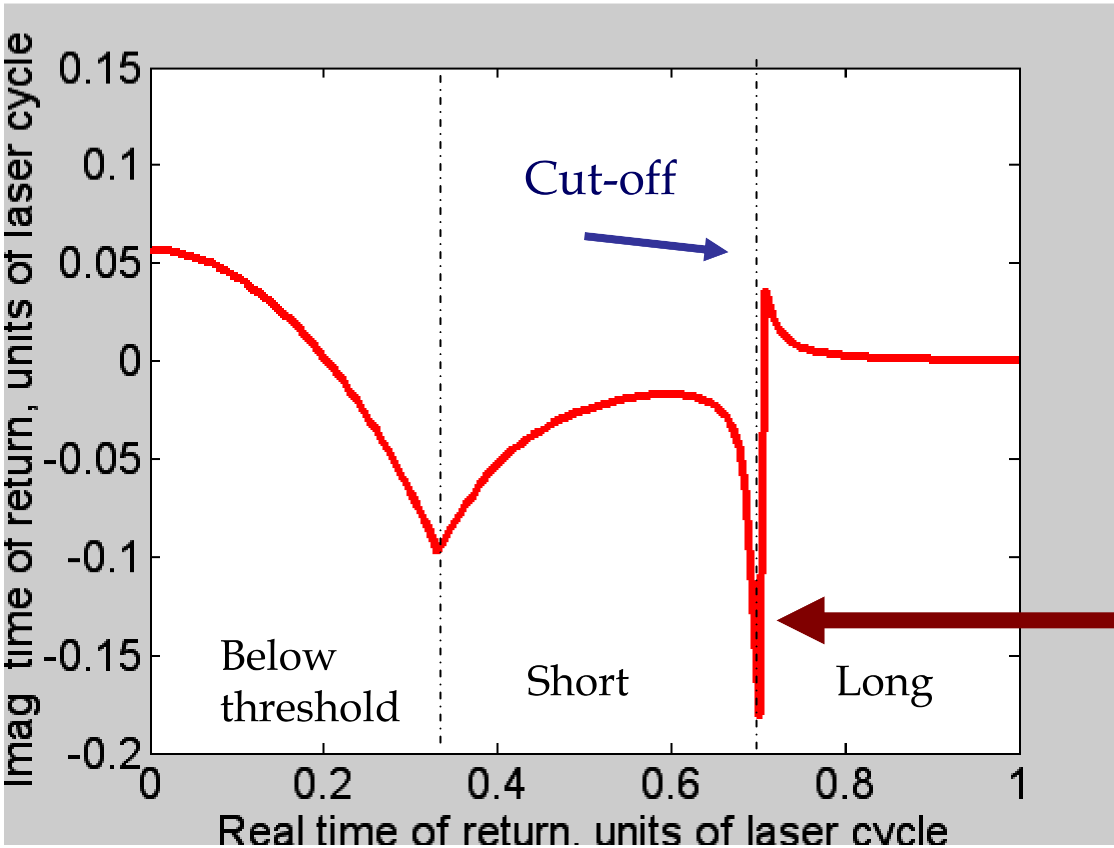
Short and long trajectories: two different saddle point solutions for the same Energy of return (Harmonic number)



Saddle point method breaks down near the cut-off: 2 saddle points merge ($S_{tt}''=0$)

Imaginary time of return

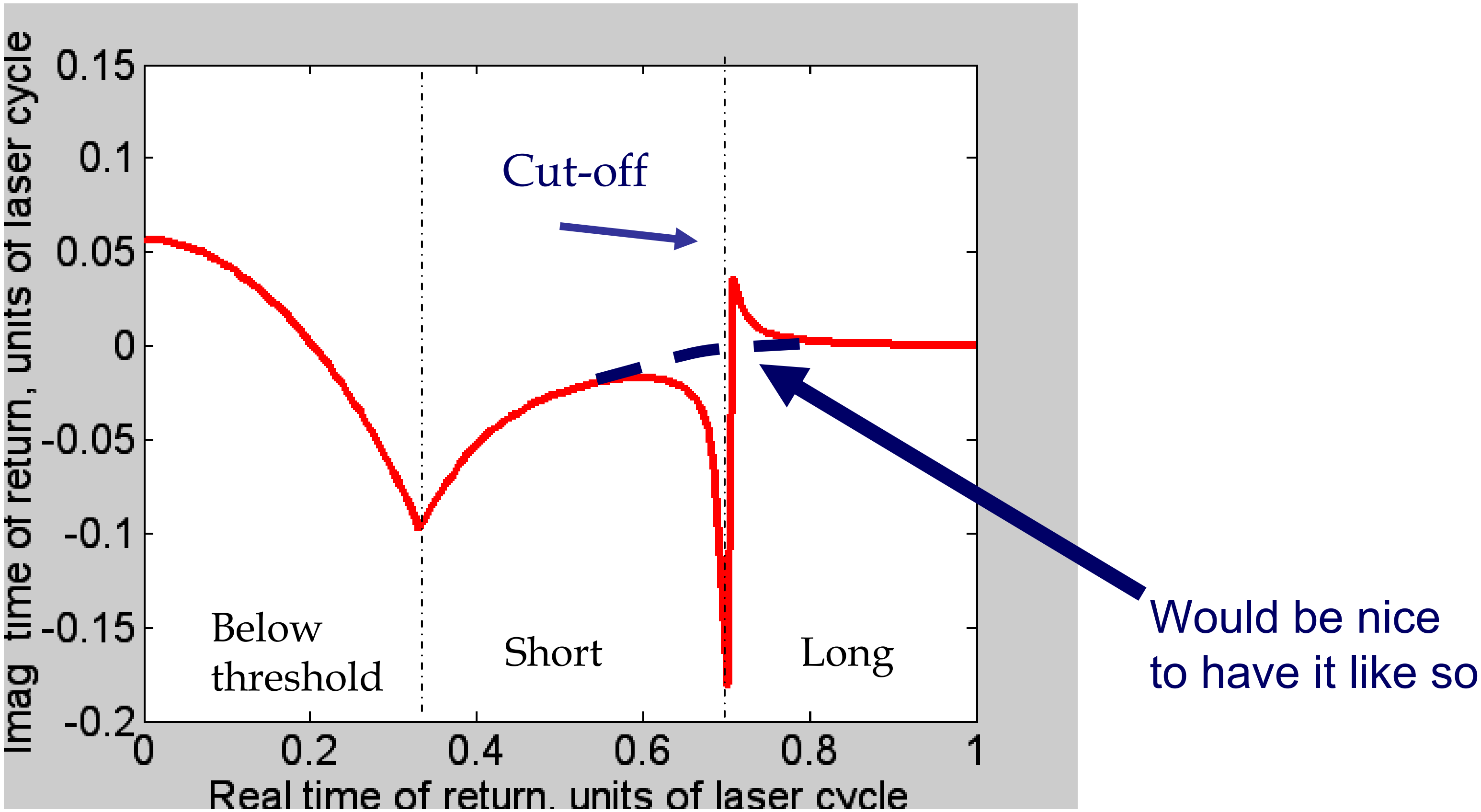
Imaginary& real time of return define integration contour in complex plane: only along this contour the energy of return is real.



Difficult part is here: divergence

Imaginary time of return

Imaginary& real time of return define integration contour in complex plane: only along this contour the energy of return is real.



3 May 2011

Treating the cut-off region

Step1:

Find $t_r = t_0$ (and $t_i = t_{i0}$, $k_s = k_{s0}$), such that $S_{tt}''(t_{r0}, t_{i0}, k_{s0}) = 0$, i.e. $dE_{ret}/dt = 0$

(Pick the cut-off (real) return time for t_0)

Step2:

Expand the action $S(t_r, t_i, k_s)$ around t_0 (the uniform approximation)

$$S(t, t_{i0}, k_{i0}) = S(t_0, t_{i0}, k_{i0}) + S_t'(t_0, t_{i0}, k_{i0})(t - t_0) + S_{ttt}'''(t_0, t_{i0}, k_{i0}) \frac{(t - t_0)^3}{6}$$

Step3:

The resulting integral for dipole:

$$D(N\omega) \propto \int_{-\infty}^{+\infty} dt\, e^{-iS(t, t_{i0}, k_{s0})} e^{iN\omega t} + c.c.$$

can be calculated analytically using Airy function:

$$\int_{-\infty}^{+\infty} \cos(at^3 \pm xt) \equiv \frac{\pi}{(3a)^{1/3}} Ai\left[\pm \frac{x}{(3a)^{1/3}} \right]$$

Treating the cut-off region

Introduce the cut-off harmonic number N_0 and "the distance from cut-off" $\Delta N = N - N_0$:

$$S_t^{'}(t_0) = E_{ret}(t_0) + I_p \equiv N_0\omega$$

(N_0 does not have to be integer)

The dipole near cut-off is expressed via Airy function :

$$D(N\omega) \propto \int_{-\infty}^{+\infty} dt\, e^{-iS(t,t_{i0},k_{s0})} e^{iN\omega t} + c.c. = \int_{-\infty}^{+\infty} \cos\left(\frac{\chi}{6}\xi^3 + \Delta N\omega\xi\right) d\xi$$

$$D(N\omega) \propto \frac{2\pi}{[\chi/2]^{1/3}} Ai\left[\frac{\Delta N\omega}{(\chi/2)^{1/3}}\right]$$

$\Delta N<0$ before cut-off: $Ai \sim \cos[-(\Delta N\omega)^{3/2}(8/9\chi)^{1/2}]$

(oscillations are due to interference of short and long)

$\Delta N>0$ after cut-off: $Ai \sim \exp[-(\Delta N\omega)^{3/2}(8/9\chi)^{1/2}]$

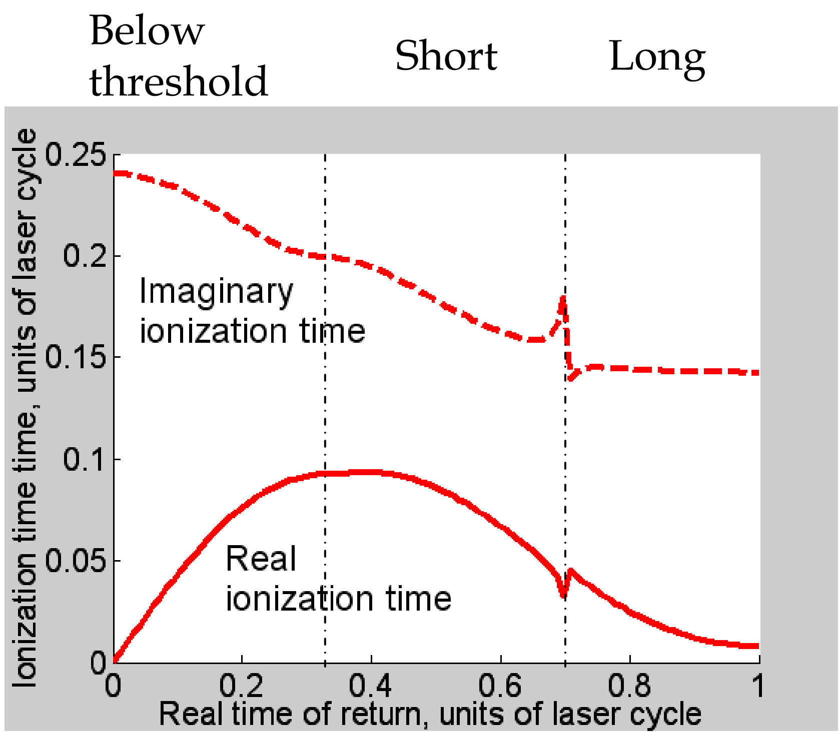
(exponential decrease of HH spectrum after cut-off)

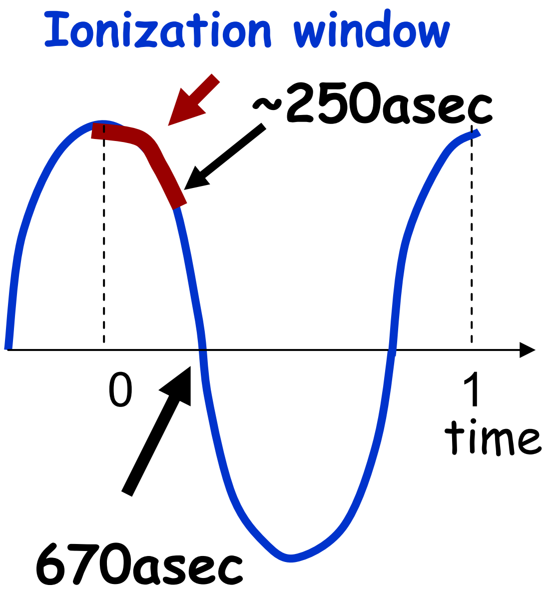
$$\chi = -S_{ttt}^{'''}(t_0)$$

$$\chi \cong v(t_0)F_0\omega \qquad F_t^{'}(t_0) \cong F_0\,\omega$$

$$F(t_0) \cong 0$$

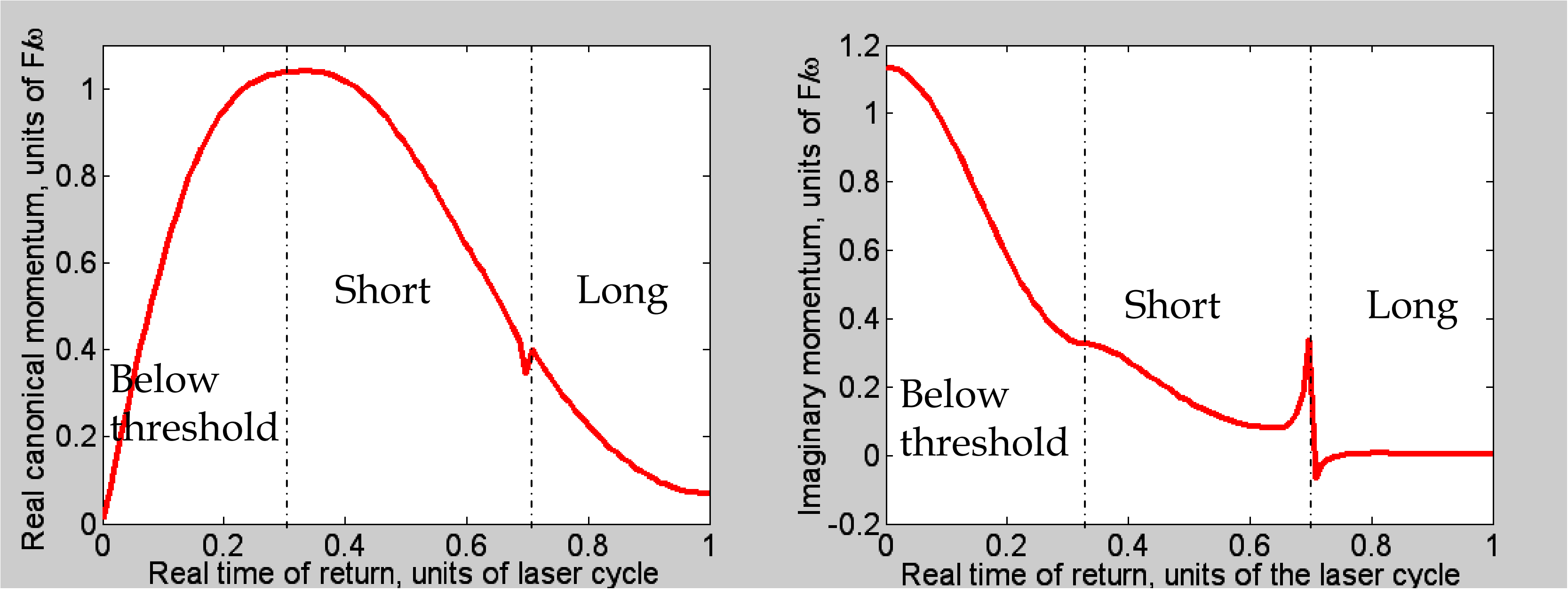
Ionization times: sub-cycle dynamics of ionization





Imaginary ionization time defines ionization probability:
short trajectories are suppressed
Real ionization time defines "duration of ionization window"

Canonical momenta



Long trajectories: imaginary canonical momentum is very small
Short trajectories: substantial imaginary canonical momentum
Photoelectrons: registered at the detector - canonical momentum is real

Classical 3-step model: photoelectron perspective

Define "classical ionization time" – "time of birth", when electron velocity is zero.

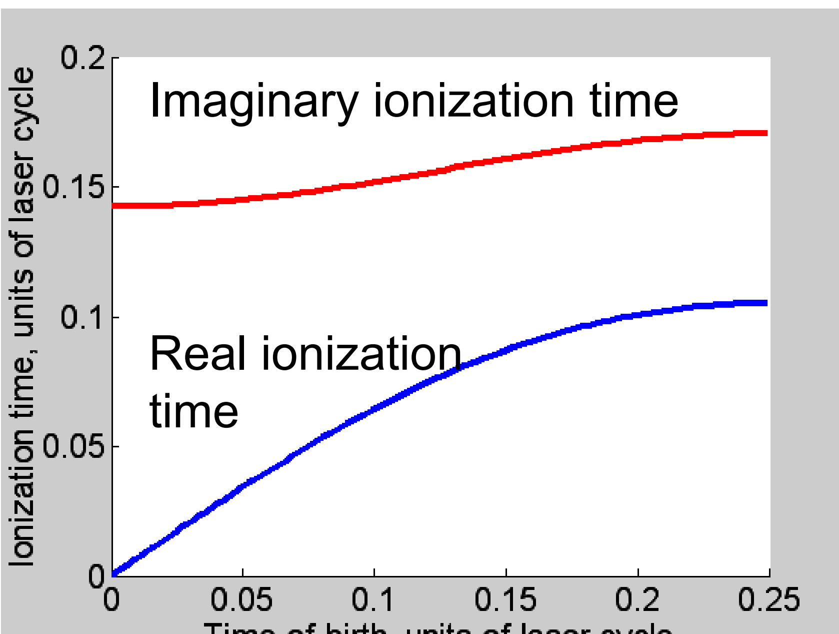
$v(t_B)=k+A(t_B)=0$

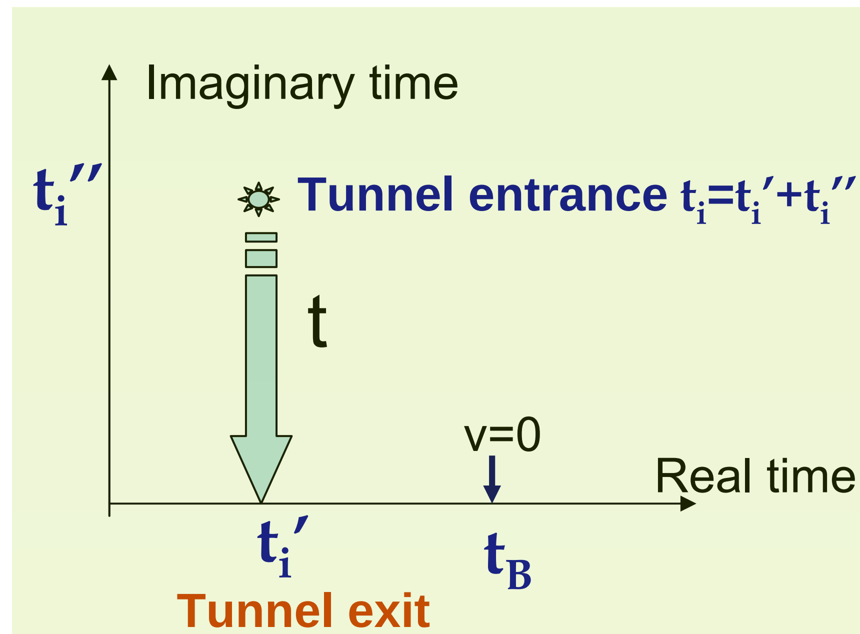
$$\frac{1}{2}\left(A(t_i)-A(t_B)\right)^2+I_p=0$$

$v(t_i)=A(t_i)-A(t_B)=-i\,\gamma$

$k=-A(t_B)$

Different from "quantum ionization times", since k is forced to be real.

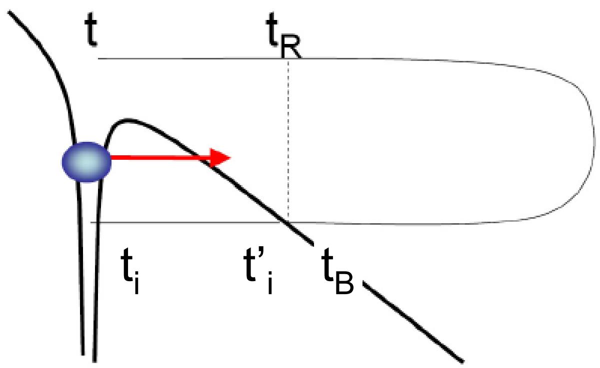




Classical 3-step model: photoelectron perspective

Define classical return time: $$\int_{t_B}^{t_R} \left(A(\tau) - A(t_B)\right) d\tau = 0$$

Electron returns to the point (coordinate), where it had zero velocity



$t_B > t_i'$, $v(t_B)=0$

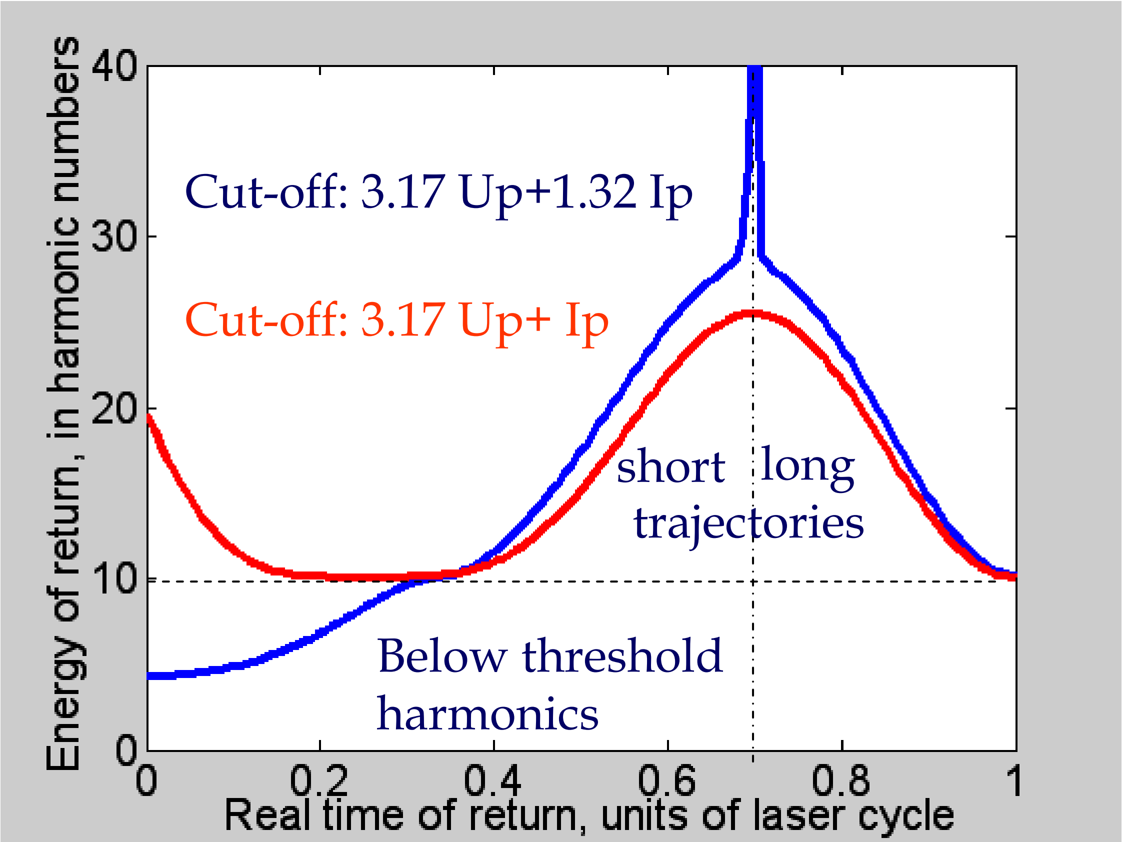
$v(t_i')=k+A(t_i')<0$

The electron is not born with v=0.

Classical return energy: $E=(A(t_R)-A(t_B))^2/2$

Classical energy of return

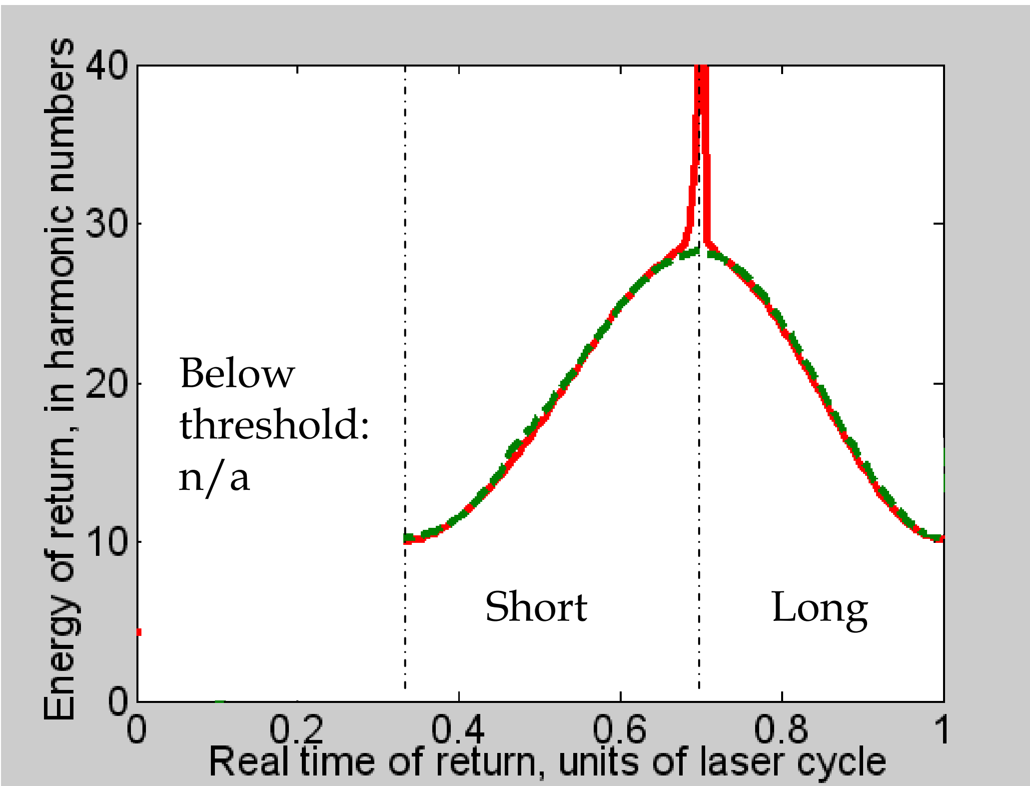
Classical cut-off corresponds to lower energies: electron does not return to the core



Can we improve these results? Let's let the photo-electrons return to the core.

Photoelectrons vs Lewenstein model

Energy at closest approach: not all electrons can return exactly to the core since we limited $k=-A(t_B)$



Lewenstein model

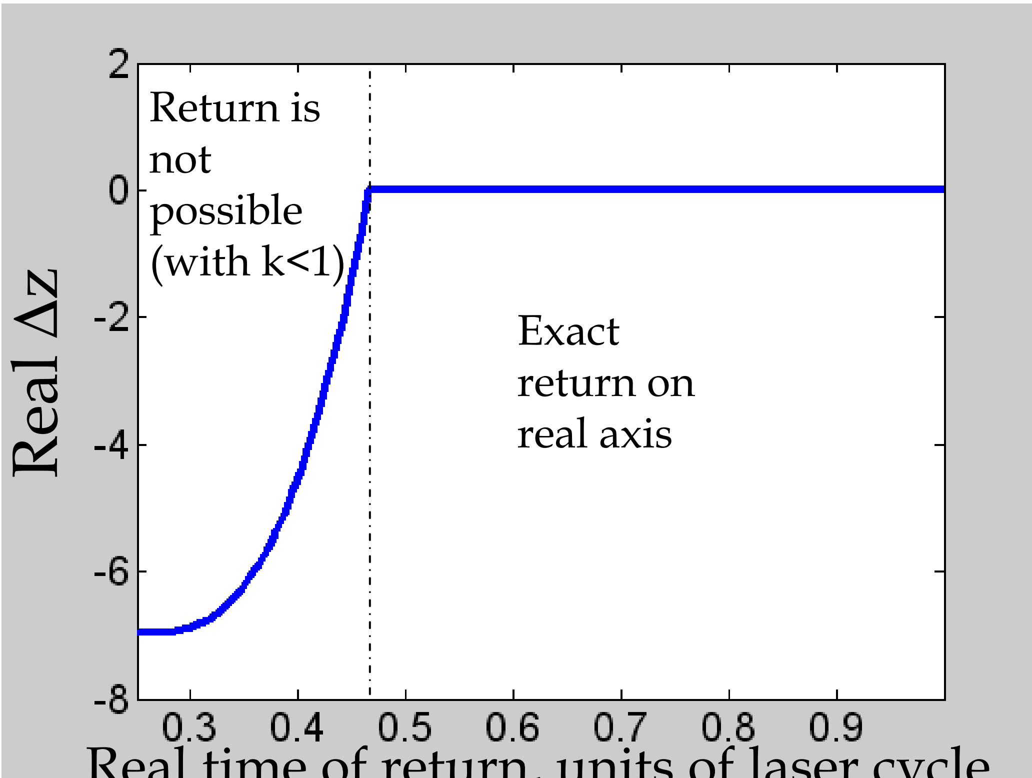
Improved 3 –step model (k<1) + relaxed return condition:

- Neglect Imag $\Delta z = 0$
- Minimize Real Δz

The strict requirement of perfect return is an artefact of neglecting the size of the ground state. Relaxing this requirement seems quite reasonable!

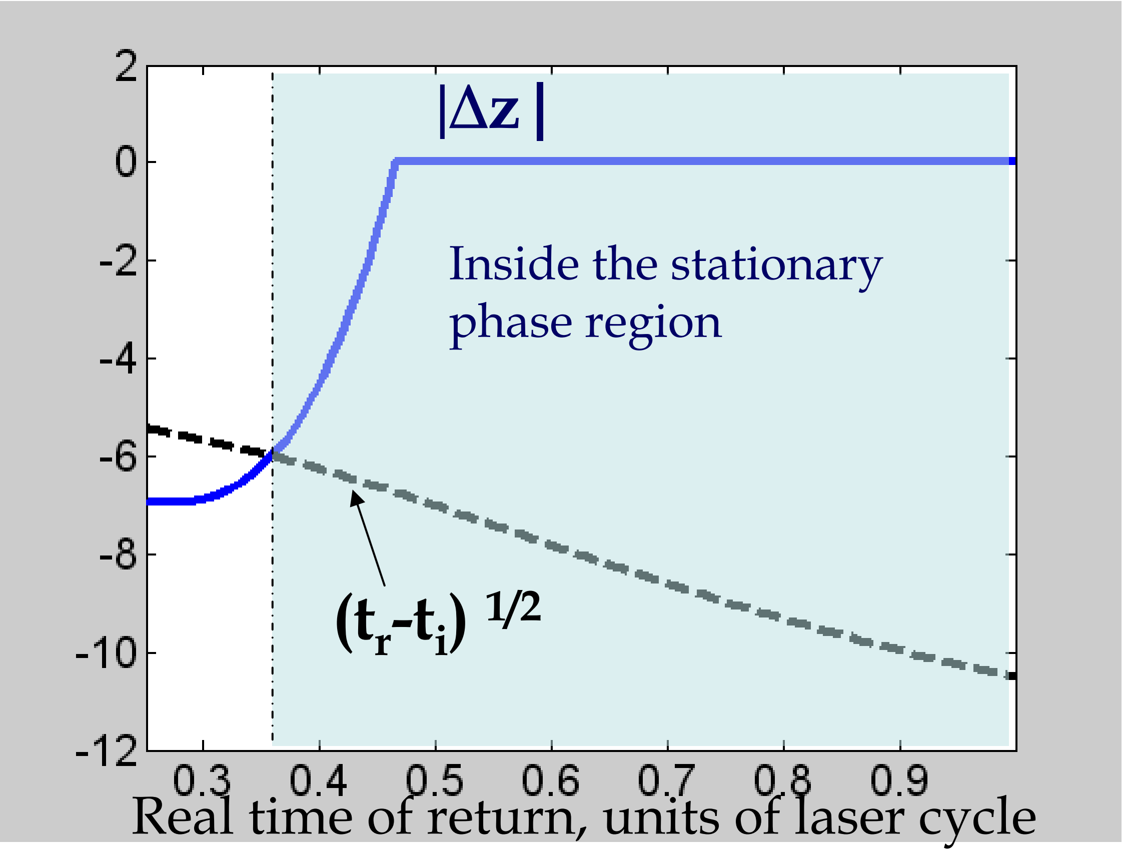
Photoelectrons: return coordinate

Not all electrons can return exactly to the core since we limited $k=-A(t_B)$



Photoelectrons: saddle point region

Real k should be within the saddle point region of the exact complex saddle point. This region is ~ $|S''_{kk}|^{-1/2}$



$|\Delta z| < (t_r - t_i)^{1/2}$

$|\Delta k| < |S''_{kk}|^{-1/2}$

$|\Delta k| < |t_r - t_i|^{1/2}$

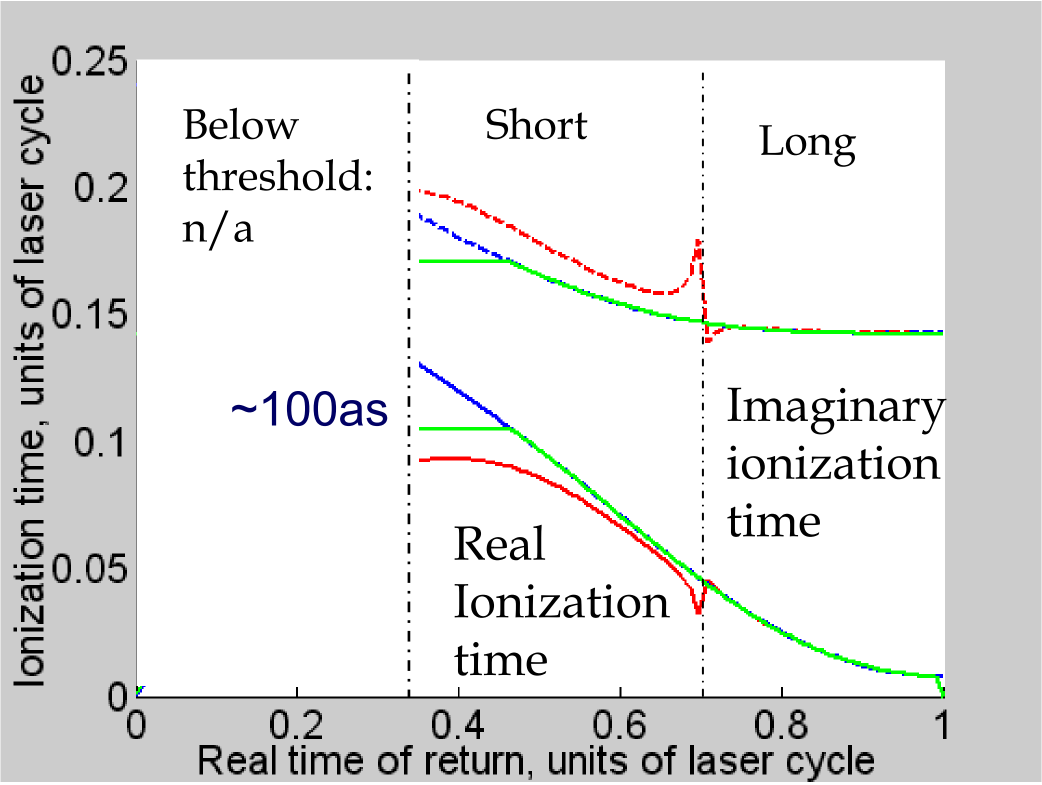
$|\Delta k| = \Delta z/(t_r - t_i)$

$|\Delta z| < (t_r - t_i)^{1/2}$

$|\Delta k|$ - difference between the exact k_s and photolelctron k_s
$|\Delta z|$ - distance of the closest approach to the core

Photoelectrons vs Lewenstein model

Ionization time



Lewenstein model

Photoelectrons, k is not restricted (k can be >1)

Improved 3 –step model + relaxed return condition

Long trajectories: good agreement due to small imaginary displacement

Factorization of the dipole

Dipole = product of 3 amplitudes: ionization, propagation, recombination

$$D(N\omega) = a_{rec}(k_s;t_R)\,a_{prop}(k_s;t_R,t_i)\,a_{ion}(k_s;t_i)$$

This factorization is rigorous within the photo-electron picture, i.e. if the imaginary part of the canonical momentum is negligible

The next step is to take each amplitude separately and improve it beyond the SFA and the simple model of an ion without internal states.

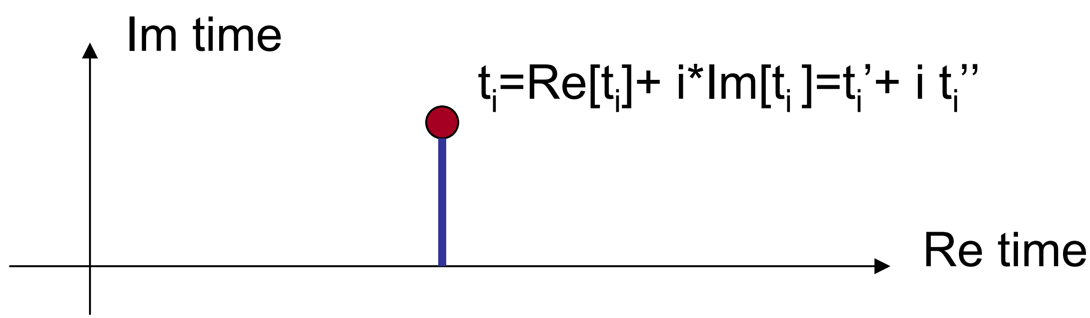
Results of saddle point integration

Dipole = product of 3 amplitudes: ionization, propagation, recombination

$$D(N\omega) = a_{rec}(k_s;t_R)\, a_{prop}(k_s;t_R,t_i)\, a_{ion}(k_s;t_i)$$

The amplitudes are (within saddle point): 1. Ionization

$$a_{ion}(k_s;t) \sim d[k_s + A(t_i)]\, e^{-i\frac{1}{2}\int_{t_i}^{t_i'}\left(\vec{k}_s+\vec{A}(\tau)\right)^2 d\tau - iI_p(t'_i - t_i)}\, \frac{\sqrt{\pi}}{\sqrt{S''_{t_i,t_i}}}$$



Results of saddle point integration

Dipole = product of 3 amplitudes: ionization, propagation, recombination

$$D(N\omega) = a_{rec}(k_s;t_R)\, a_{prop}(k_s;t_R,t_i)\, a_{ion}(k_s;t_i)$$

The amplitudes are (within saddle point):

Propagation: wavepacket spreading and phase accumulation

$$a_{prop}(k_s;t_R;t'_i) \sim e^{-i\frac{1}{2}\int_{t'_i}^{t_R}\left(\vec{k}_s+\vec{A}(\tau)\right)^2 d\tau - iI_p(t_R-t'_i)} \frac{\sqrt{\pi^3}}{[t_R-t_i]^{3/2}}$$

Results of saddle point integration

Dipole = product of 3 amplitudes: ionization, propagation, recombination

$$D(N\omega) = a_{rec}(k_s; t_R)\, a_{prop}(k_s; t_R, t'_i)\, a_{ion}(k_s; t_i)$$

The amplitudes are (within saddle point):

Recombination: proportional to the recombination dipole

$$a_{rec}(k_s; t_R) \sim d^*(k_s + A(t_R)) \frac{\sqrt{\pi}}{\sqrt{S''_{t_R t_R}}}$$

Ionization amplitude

$$a_{ion}(k_s;t) = d[k_s + A(t_i)]\, e^{-i\frac{1}{2}\int_{t_i}^{t_i'}\left(\vec{k}_s+\vec{A}(\tau)\right)^2 d\tau - iI_p(t'_i - t_i)}\, \frac{\sqrt{\pi}}{\sqrt{S''_{t_i,t_i}}}$$

This expression came from applying the saddle point approximation to

$$a(k_s,t) = -i\int_{t_0}^{t} dt'\, e^{-iS(t,t',k)+I_p t'} F_L(t')\, d\left(\vec{k}_s + \vec{A}(t')\right)$$

This integral has been extensively studied by Keldysh, PPT (Popov, Perelomov, Teren'tev), and others. The SFA result can be significantly improved!

Ionization amplitude

The recipe for atoms: (PPT, Keldysh, Yudin-Ivanov, Becker-Faisal, Popruzhenko-Bauer)

Sub-cycle rates

$$a_{ion}(k_s,t_i) = R_{l,m}(I_p,F)\boxed{e^{-\mathrm{SFAexponent}(k_s,t_i)}}$$

-Calculate exponential dependence with SFA

- Add Coulomb correction R_{lm} to account for the core.

- The Coulomb correction depends on l, m

- With reasonable accuracy and for linearly polarized or low-frequency fields the Coulomb correction can be taken from static tunneling

The recipe for molecules: Take the Coulomb correction from static tunneling rates (MO-ADK (Tong&Lin), recently Murray & Ivanov)

The Multi-Channel Case

Exact 'continuum' part: electron+ion

$$\Psi_c(t) = -i\int_{t_0}^{t} dt' e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau} V_L(t') e^{-i\hat{H}_0(t'-t_0)}\Psi_g$$

$$1 = \sum_n \int_{-\infty}^{+\infty} d\vec{k}\left|\vec{k}+\vec{A}(t')\right\rangle\left|n\right\rangle\left\langle n\right|\left\langle\vec{k}+\vec{A}(t')\right|$$

Polarized Core states

Continuum states

$$\Psi_c(t) = -i\sum_n \int_{-\infty}^{+\infty} d\vec{k}\int_{t_0}^{t} dt' e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau}\left|\vec{k}(t')\right\rangle\left|n\right\rangle\left\langle n\right|\left\langle\vec{k}(t')\right|V_L(t')e^{-i\hat{H}_0(t'-t_0)}\left|\Psi_g\right\rangle$$

Dyson orbital: $\Psi_D^n \propto \left\langle n \middle| \Psi_g \right\rangle$

$$\left\langle\vec{k}(t')\right|V_L(t')e^{-i\hat{H}_0(t'-t_0)}\left|\Psi_D^n\right\rangle$$

The Multi-Channel Case

Exact 'continuum' part: electron+ion

$$\Psi_c(t) = -i\int_{t_0}^{t} dt'\, e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau}\, V_L(t')\, e^{-i\hat{H}_0(t'-t_0)}\,\Psi_g$$

$$1 = \sum_n \int_{-\infty}^{+\infty} d\vec{k} \left|\vec{k}+\vec{A}(t')\right\rangle |n\rangle\langle n| \left\langle \vec{k}+\vec{A}(t')\right|$$

Polarized Core states

Continuum states

$$\Psi_c(t) = -i\sum_n \int_{-\infty}^{+\infty} d\vec{k} \int_{t_0}^{t} dt'\, e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau} \left|\vec{k}(t')\right\rangle |n\rangle \left\langle \vec{k}(t')\right| V_L(t')\, e^{-i\hat{H}_0(t'-t_0)} \left|\Psi_D^n\right\rangle$$

Neglect e-e correlation after ionization: the continuum electron moves in a self-consistent field of the core

The Multi-Channel Case

Exact 'continuum' part: electron+ion

$$\Psi_c(t) = -i\sum_n \int_{-\infty}^{+\infty} d\vec{k} \int_{t_0}^{t} dt' e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau} \left|\vec{k}(t')\right\rangle \left|n\right\rangle \left\langle \vec{k}(t')\right| \left\langle n\right| V_L(t') e^{-i\hat{H}_0(t'-t_0)} \left|\Psi_D^n\right\rangle$$

Neglect e-e correlation after ionization: the continuum electron moves in a self-consistent field of the core

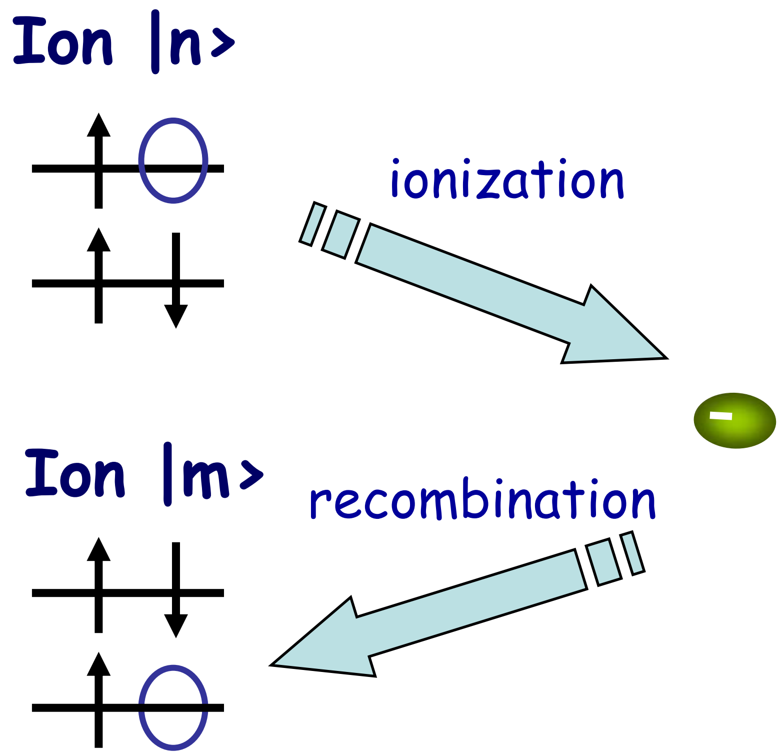
$$\Psi_c(t) = -i\sum_n \int_{-\infty}^{+\infty} d\vec{k} \int_{t_0}^{t} dt' \underbrace{e^{-i\int_{t'}^{t}\hat{H}_c(\tau)d\tau} \left|\vec{k}(t')\right\rangle}_{\text{continuum}} \underbrace{e^{-i\int_{t'}^{t}\hat{H}_i(\tau)d\tau} \left|n\right\rangle}_{\text{ion}} \left\langle \vec{k}(t')\right| \left\langle n\right| V_L(t') e^{-i\hat{H}_0(t'-t_0)} \left|\Psi_D^n\right\rangle$$

Evolution in the continuum – like before

Evolution in the ion – start in |n> at t', end in |m> at t, amplitude $a_{mn}(t,t')$

The Multi-Channel Case

$$\Psi_c(t) = -i\sum_n \int_{-\infty}^{+\infty} d\vec{k} \int_{t_0}^{t} dt'\, \underbrace{e^{-i\int_{t'}^{t}\hat{H}_c(\tau)d\tau} \left|\vec{k}(t')\right\rangle}_{\text{continuum}} \underbrace{e^{-i\int_{t'}^{t}\hat{H}_i(\tau)d\tau} \left|n\right\rangle}_{\text{ion}} \left\langle\vec{k}(t')\right| V_L(t') e^{-i\hat{H}_0(t'-t_0)} \left|\Psi_D^n\right\rangle$$



The harmonic dipole in the multi-channel case

The harmonic dipole is a sum over all ionic states at t_{ion} and t_{rec}

$$D(N\omega) = \sum_{n,m} a_{rec,m}(k_s;t_R)\, a_{prop,mn}(k_s;t_R,t_i)\, a_{ion,n}(k_s;t_i)$$

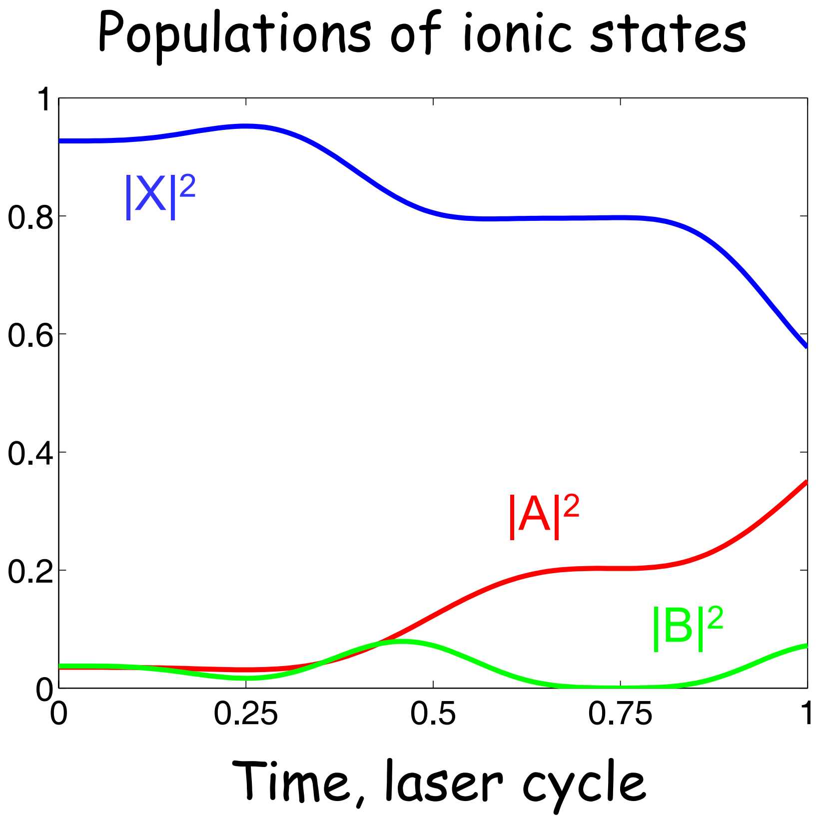
The key change is in the propagation amplitude— it includes transitions between the initial (n) and the final (m) states of the ionic core:

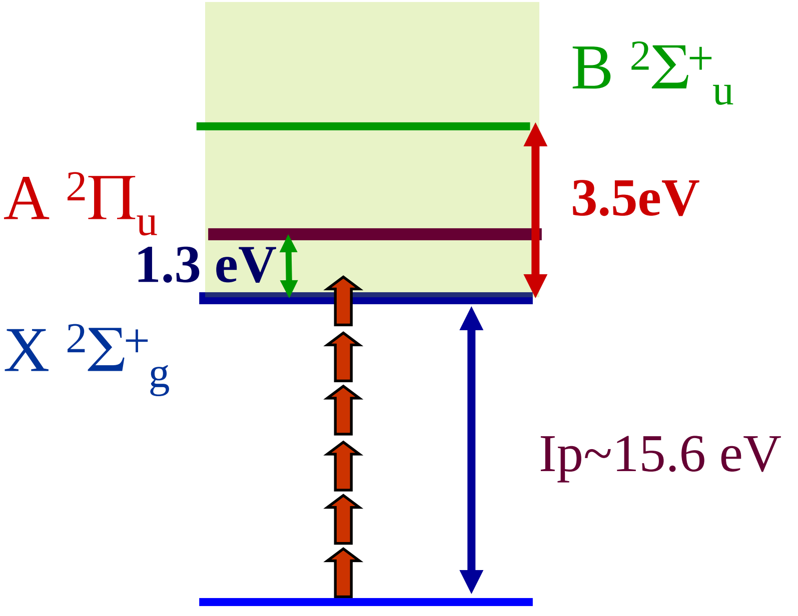
$$a_{prop,mn}(k_s;t_R;t'_i) \sim a_{core,mn}(t_R,t'_i)\, e^{-i\frac{1}{2}\int_{t'_i}^{t_R}\left(\vec{k}_s+\vec{A}(\tau)\right)^2 d\tau - iI_{p,n}(t_R-t'_i)} \frac{\sqrt{\pi^3}}{[t_R-t_i]^{3/2}}$$

Recombination –the electron recombines with the ion in the state m:

$$a_{rec,m}(k_s;t_R) \sim d_m{}^*(k_s + A(t_R)) \sqrt{\pi} \Big/ \sqrt{S''_{t_R t_R}}$$

Example of core dynamics in the multi-channel case





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

Find dipole couplings between the states A,B,X.

Solve 3-level system numerically