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IMPRS UFAST

Course 2: Basic Technologies

Part IV: High Harmonic Generation

The route to sub-fs physics

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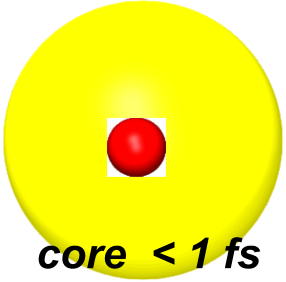
Outline

- Attosecond Physics
- Strong field ioniation (Keldysh / ADK)
- HHG
  - Wavelength scaling
  - Conversion efficiencies
- Techniques
  - Ultrashort pulses
  - CEP
  - PG
  - DOG
- X-ray optics in pump-probe setups

timestructure of atomic processes

time ↔ energy : $\Delta t \sim h/\Delta E$



valence n = 1 ~ 1 fs

object of study:
dynamics
of inner-shell processes

Rydberg n > 1 fs ... µs

but visible light: T ~ 2 fs

requires: attosecond XUV-pulse

Strong field ionization

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Ionization in strong fields

Let us assume the case of the hydrogen atom:

The strength of the Coulomb potential experienced by an electron in the first Bohr orbit of atomic hydrogen is defined by

$$\mathcal{E}_a = \frac{e}{(4\pi\varepsilon_0)a_0^2} = 5.14221 \times 10^9 \mathrm{V/cm}$$

and we determin the field strength of a laser electric field from the intensity with

$$I = \frac{1}{2}\varepsilon_0 c E^2$$

thus the Coulomb pontential of the hydrogen atom corresponds to a laser electric field intensity of $\boldsymbol{\sim 3.51 \cdot 10^{16}\ W/cm^2}$

Interaction with a time varying electric field

The perturbation of a time varying electric field of the typical strength of a laser interacting with an atom cannot be treated in terms of perturbation theory anymore.

- the perturbation of the Coulomb potential is not weak!
- the strong time varying potential competes with the Coulomb binding potential

The dynamics of the ionization process of a bound electron inside the potential is strongly determined by the instantaneous strength of the applied electric field.

The strength of the electric field can change by orders of magnitude within a cycle of the laser electric field

The Keldysh approach (1965)

The Keldysh approach (1965)

If we assume a classical partical with binding energy I_p and a field strength that sufficiently supresses the coulomb potential, we can define a „barrier thickness“

$$l = \frac{I_p}{e\,\mathcal{E}_0}$$

and a corresponding „tunneling time“ the electron needs to penetrate through the rectangular barrier

$$\tau = \frac{\sqrt{2mI_p}}{e\,\mathcal{E}_0}$$

Keldysh parameter

Multiplying with the frequency of the laser field leads to the Keldysh parameter

$$\gamma = \tau\omega_L = \omega_L \frac{\sqrt{2mI_p}}{e\mathcal{E}_l} = \sqrt{\frac{I_P}{2U_P}}$$

The Keldysh parameter γ distinguishes between the ionization processes in the limiting cases $\gamma \gg 1$ for multi photon ionization (MPI) and $\gamma \ll 1$ for tunneling ionization (TI)

Consequences for ultrashort laser pulses

- The value of τ is determined by the frequency of the laser field. Regarding high frequencies for the applied field there should appear a frequency dependent tunneling probability.
- The ponderomotive potential $U_p(t)$ defines a time varying relation for either the MPI or TI regime to dominate the ionization process.

γ should be interpreted in terms of the dominance of one process in respect to the other.

Towards High Harmonic Generation

The Keldysh interpretation allows calculation of ionization probabilities of atomic bound states in strong laser fields including excitations and resonances and even the dependence on the time evolution of the laser field in the *Quasi-Static-Approximation (QSA)*

$$W_K = \frac{\sqrt{6\,\pi}}{4}\frac{I_p}{\hbar}\sqrt{1-\frac{e\mathcal{E}_0\hbar}{m^{1/2}I_p^{3/2}}} \times \exp\left[-\frac{4}{3}\frac{\sqrt{2\,m}\,I_p^{3/2}}{e\hbar\mathcal{E}_0}\left(1-\frac{m\,\omega^2\,I_p}{5\,e^2\mathcal{E}_0^2}\right)\right]$$

<u>Limitations</u>

- low-frequency approximation for the applied electric field
- the final state of the electron is a free electron oscillating in the laser field, which is known as a final nonperturbative Volkov state
- does not include any kind of species dependence in the ionization rate calculation

ADK Theory (Ammosov, Delone and Krainov)

Description of the ionization of complex atoms and atomic ions in arbitrary states

The ionization rate equation in atomic units is given by

$$W_{m\,ADK} = |C_{n^*l^*}|^2 \quad f_{lm} \quad I_p\sqrt{\frac{6}{\pi}}\left(\frac{2(2I_p)^{3/2}}{\mathcal{E}}\right)^{2n^*-|m|-3/2} \times \quad \exp\left(-\frac{2(2I_p)^{3/2}}{3\mathcal{E}}\right)$$

with

$$f_{lm} = \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!}$$

$$|C_{n^*l^*}|^2 = \frac{2^{2n^*}}{n^*\Gamma(n^*+l^*+1)\Gamma(n^*-l^*)}$$

Ionization Probability

$$W_{m\,ADK} = |C_{n^*l^*}|^2 \quad f_{lm} \quad I_p\sqrt{\frac{6}{\pi}}\left(\frac{2(2I_p)^{3/2}}{\mathcal{E}}\right)^{2n^*-|m|-3/2}$$
$$\times \quad \exp\left(-\frac{2(2I_p)^{3/2}}{3\mathcal{E}}\right)$$

n^* effective principal quantum number
m magnetic quantum number
l the angular momentum
I_p is the atomic ionization potential
$\mathcal{E}$ the electric field strength of the laser.

For ionization rate calculation the ground state values for n* and l are mainly used, which leads to $l^* = n^* - 1$.

Ionization rate equation

Averaging over all magnetic quantum numbers gives the complete ionization rate

$$W_{ADK} = \frac{1}{2l+1} \sum_{m=-1}^{l} W_{m\,ADK}$$

for a gaussian laser pulse

$$\mathcal{E}(t) = \mathcal{E}_0 \exp\left(-\frac{2\ln 2\, t^2}{\tau_{in}}\right) cos(\omega_0\, t)$$

the total number of ions created within a subfraction of the laser pulse starting to act on the atom at time t'

$$\widetilde{W}_{frac} = \int_{t}^{+\infty} W_{ADK}(\mathcal{E}(t'))dt'$$

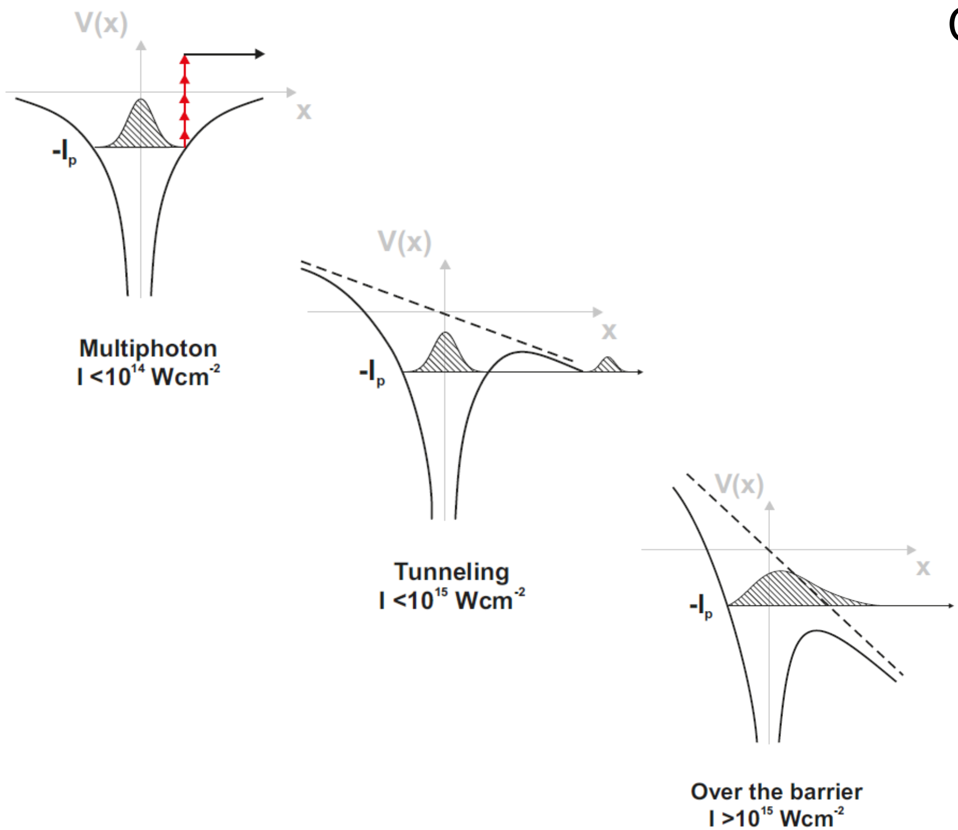
Ionization rate in the quasistatic limit

$$\Gamma_{QS}(t) = 4\frac{(2Ip)^{5/2}}{\mathcal{E}(t)} \times \exp\left[-\frac{2(2Ip)^{3/2}}{3\mathcal{E}(t)}\right]$$

a critical laser intensity I_c can be defined describing the intensity at which the Coulomb barrier starts to be suppressed

$$\begin{aligned} I_c[W/cm^2] &= \frac{\pi^2 c\epsilon^3 I_p^4}{2Z^2 e^6} \\ &= 4 \cdot 10^9 I_p^4[eV] Z^2 \end{aligned}$$

Laser driven ionization



Coulomb barrier supression intensity

$$I_c[W/cm^2] = \frac{\pi^2 c\epsilon^3 I_p^4}{2Z^2e^6} = 4\cdot 10^9 I_p^4[eV]Z^2$$

Keldysh parameter

$$\gamma = \tau\omega_L = \omega_L\frac{\sqrt{2mI_p}}{e\mathcal{E}_l} = \sqrt{\frac{I_P}{2U_P}}$$

$\gamma \gg 1$ for *multi photon ionization (MPI)*

$\gamma \ll 1$ for *tunneling ionization (TI)*

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High Harmonic Generation

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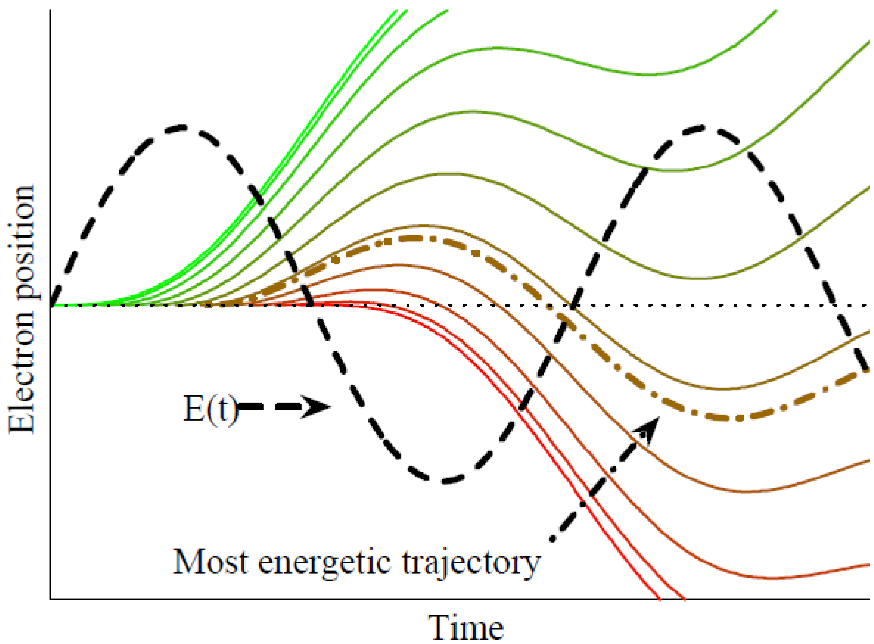
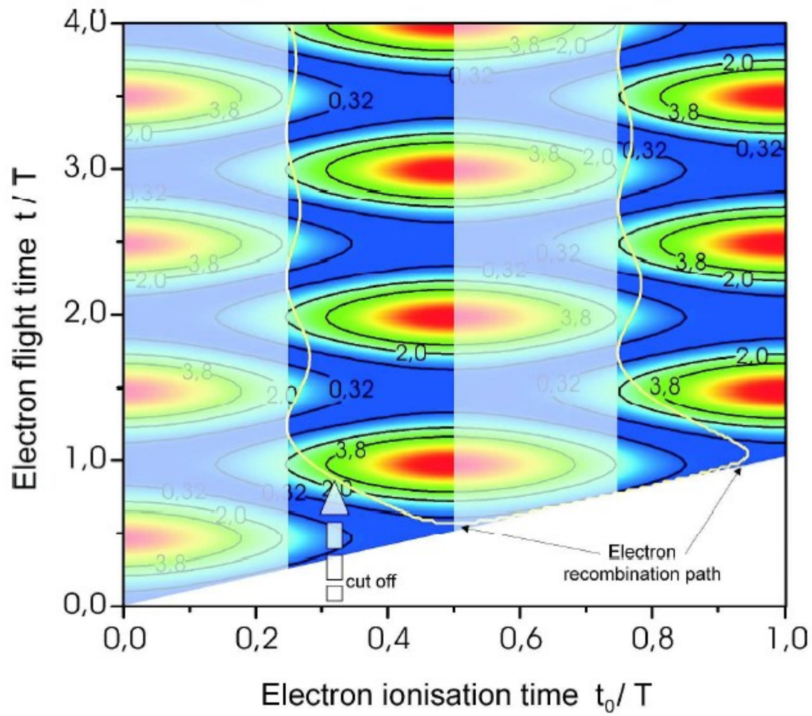
propagation of a free electron in a laserelectric field

$$
\begin{aligned}
\ddot{x}(t) &= E_0 \cos \omega t \\
\dot{x}(t) &= \frac{E_0}{\omega} \sin \omega t - \frac{E_0}{\omega} \sin \omega t_0 \\
x(t) &= -\frac{E_0}{\omega^2} \cos \omega t - (t - t_0) \frac{E_0}{\omega} \sin \omega t_0 + \frac{E_0}{\omega^2} \cos \omega t_0,
\end{aligned}
$$

t_0 time at which the electron was released from the atom assumed that the electron is released with zero initial velocity.





The position of the electron as function of time for different “ionization times" t_0. The “most energetic trajectory” refers to the solution where the electron encounters the nucleus with the maximal kinetic energy

three step model / Lewenstein 1994

$$i|\Psi(x,t)\rangle = \left[-\frac{1}{2}\nabla^2 + V(x) - E\cos(t)x\right]|\Psi(x,t)\rangle$$

The basic assumptions for this formulation of HHG are:

- the contribution to the evolution of the system of all bound states except the ground state $|0\rangle$ can be neglected
- the depletion of the ground state can be neglected since $U_p < U_{sat}$, U_{sat} being the saturation energy that completely ionizes the atom in one optical period
- in the continuum the electron can be treated as a free particle moving in the electric field with no effect of $V(x)$

The cut-off law within the Lewenstein model

As a result the maximum energy that can be extrated from an electron in the ponderomotive potential of the laser electric field is given by

$$E_{hh}^{cutoff} = I_p + 3.17 \cdot U_p$$

As a result of the more accurate quantum mechanical calculations the *cutoff law* has been corrected giving

$$E_{hh}^{cutoff} = 3.17\, U_p + I_p \cdot F(I_p/U_p)$$

For $I_p \ll U_p \Longrightarrow F(I_p/U_p) \simeq 1.32.$

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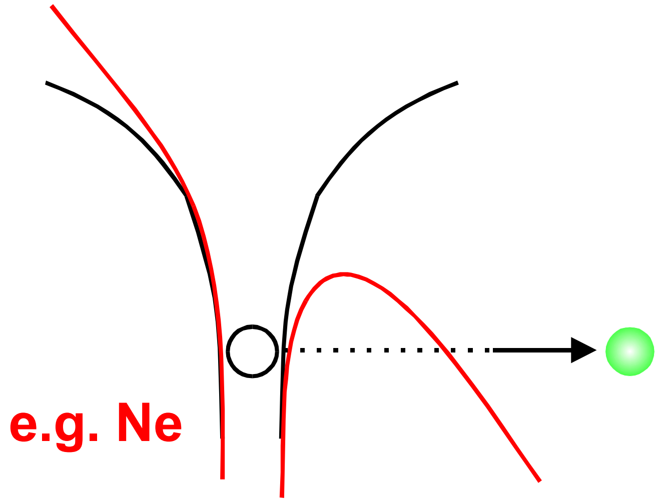
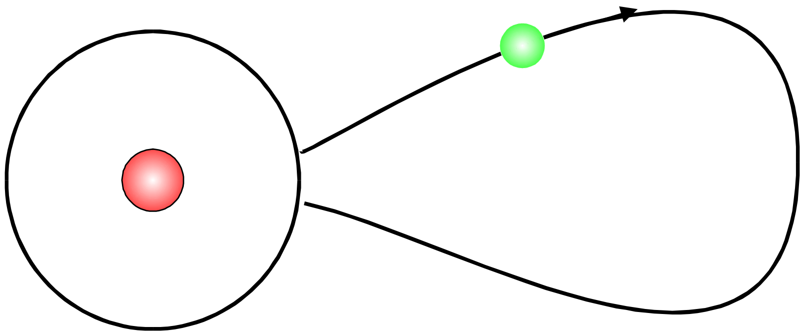
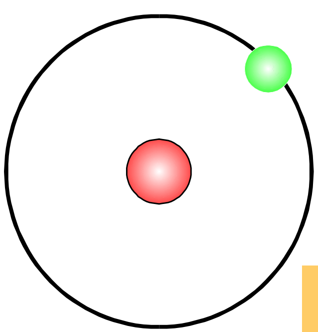


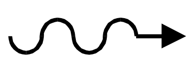
1. Tunnel-Ionisation at > 10^{14} W/cm^2

2. Electron acceleration in laser electric field

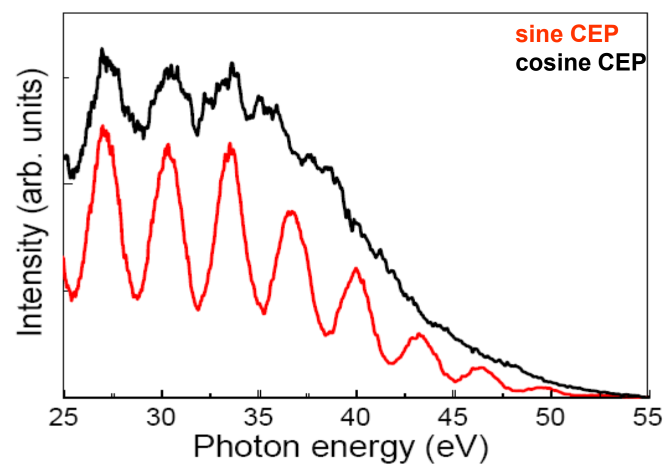


3. Return to parent ion and recombination





$h\nu_X = n * h\nu_{Laser}$ *(n uneven)*



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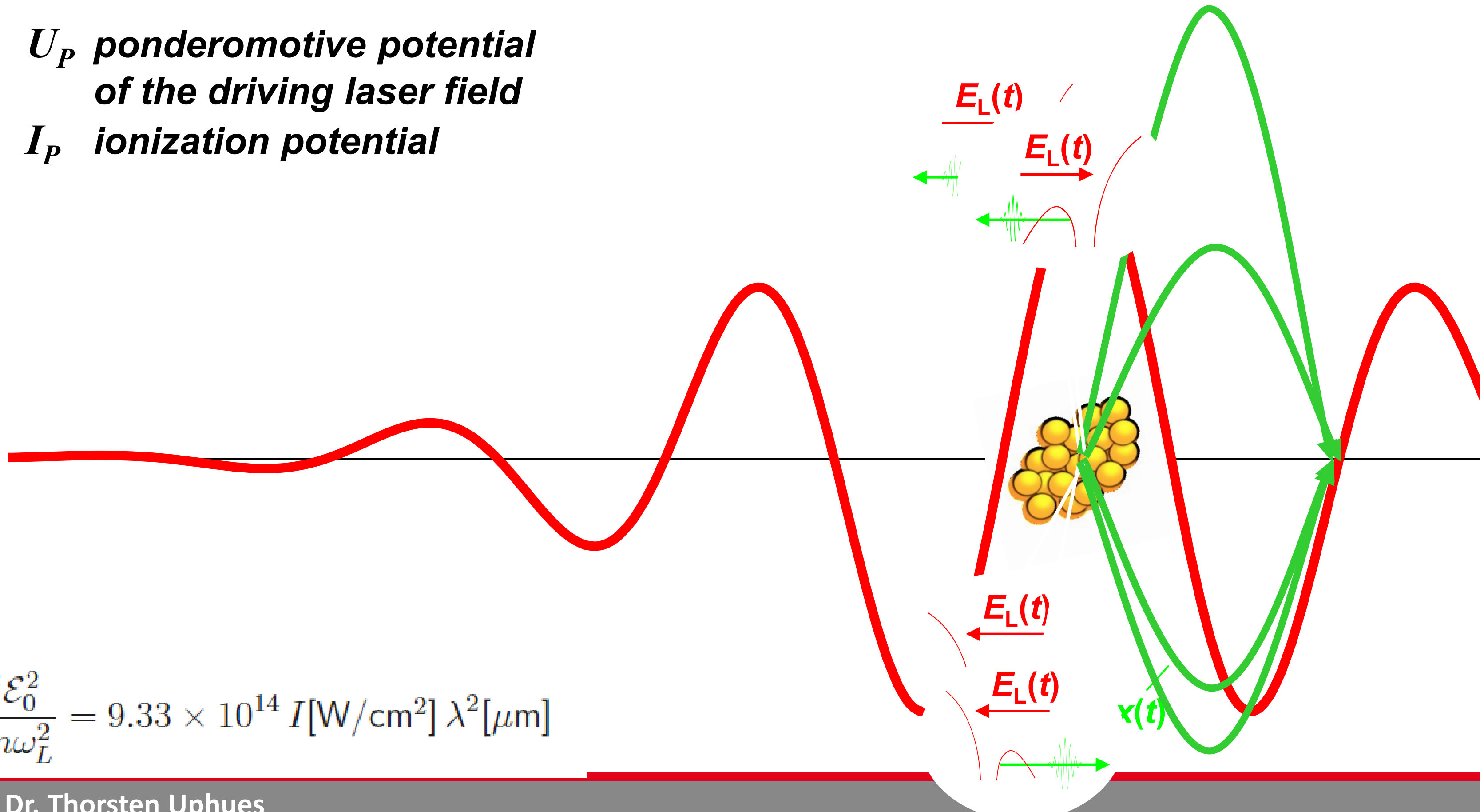
Maximum Energy Gain

$$E_{hh}^{cutoff} = 3.17\, U_p + I_p \cdot F(I_p/U_p)$$

$$I_p \ll U_p \Longrightarrow F(I_p/U_p) \simeq 1.32$$

U_P ***ponderomotive potential of the driving laser field***

I_P ***ionization potential***



$$U_p[\mathrm{eV}] = \frac{e^2 \mathcal{E}_0^2}{4\, m \omega_L^2} = 9.33 \times 10^{14}\, I[\mathrm{W/cm^2}]\, \lambda^2[\mu\mathrm{m}]$$

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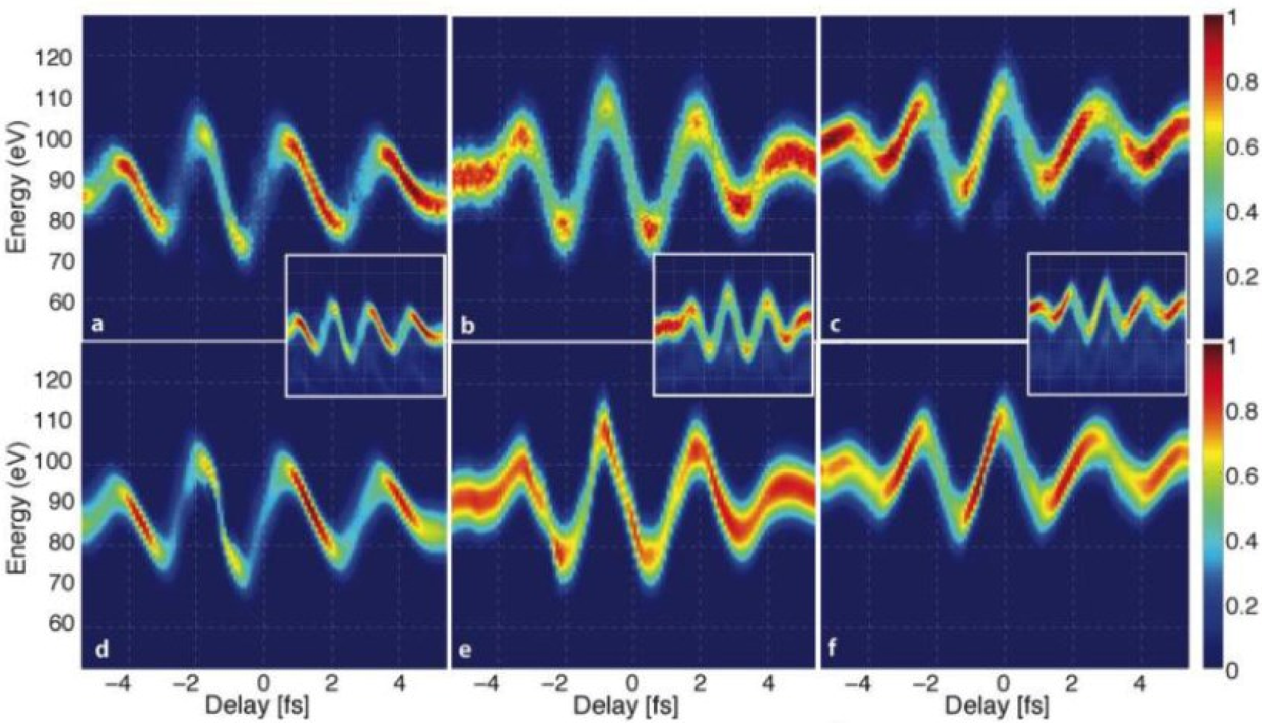
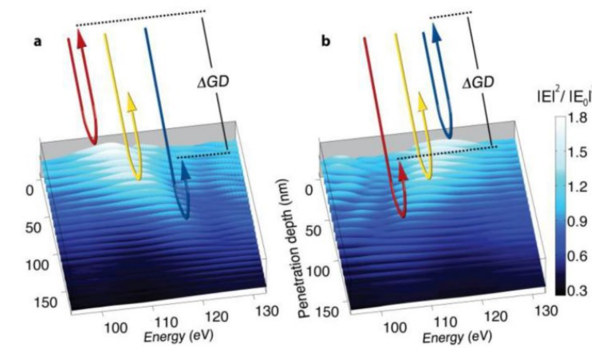
harmonics are chirped!





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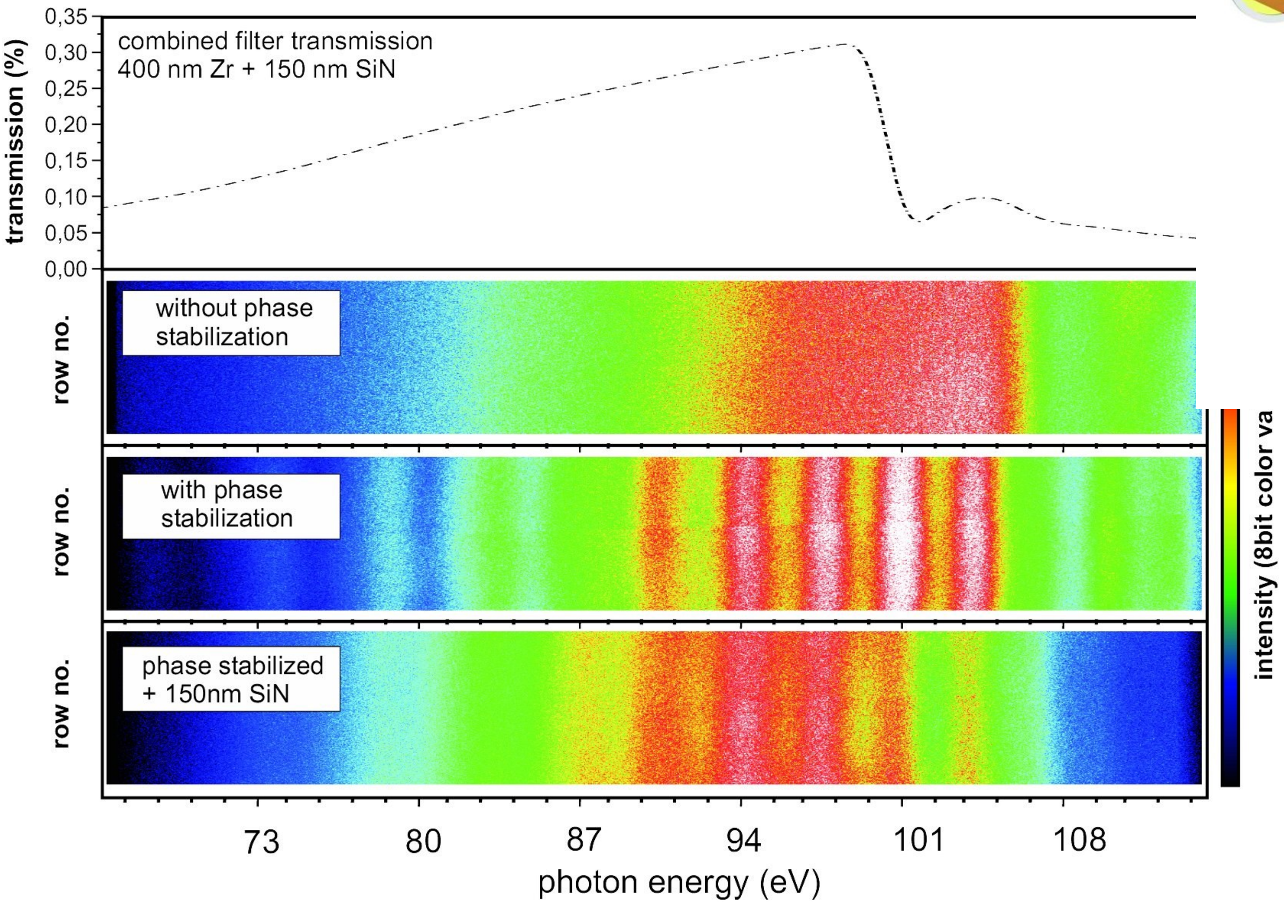
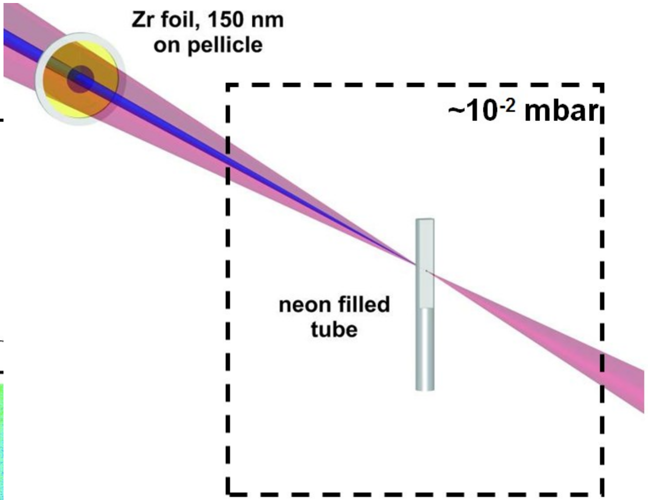
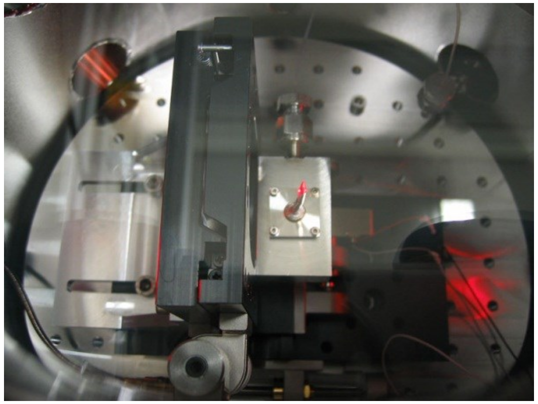
HHG Generation







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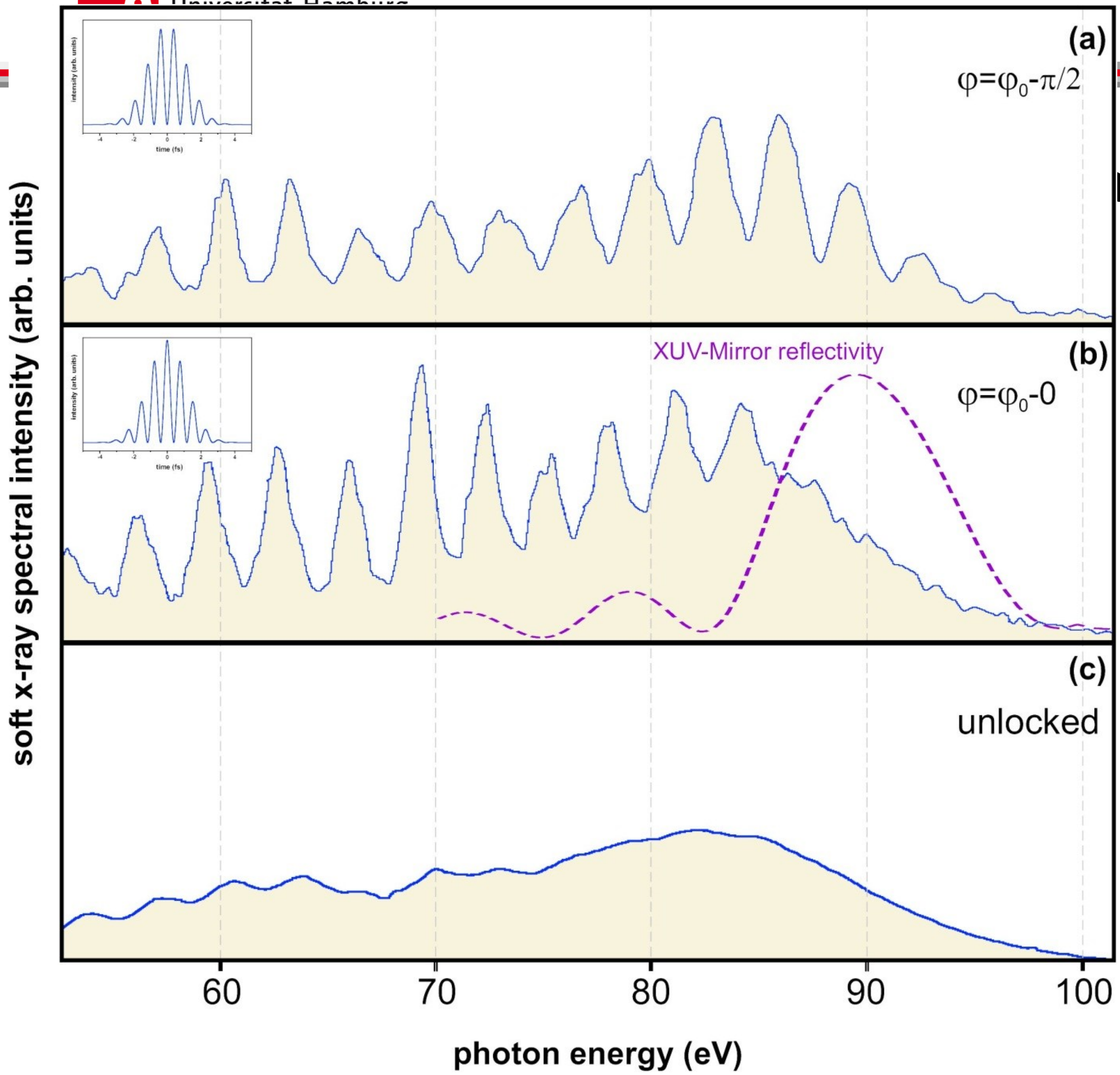

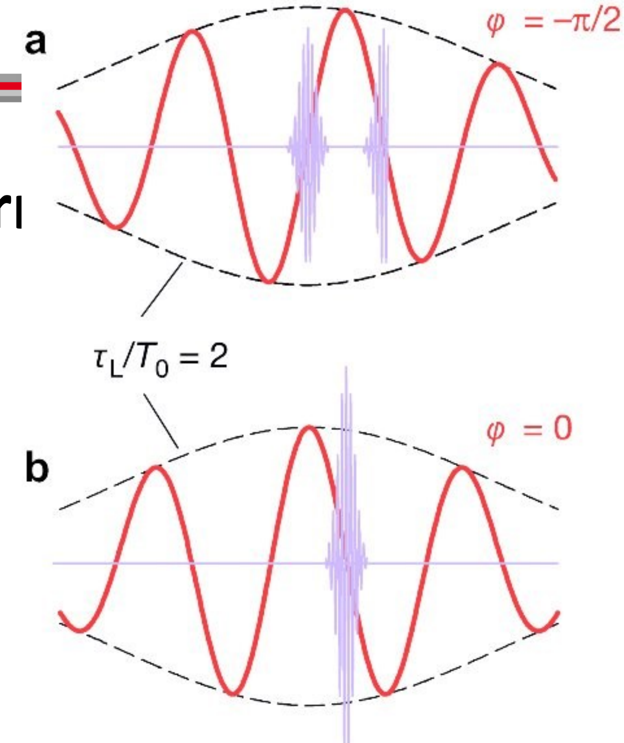

Careful adjustment of the Carrier Envelope Phase (CEP) necessary to generate isolated attosecond pulses

conversion efficiency

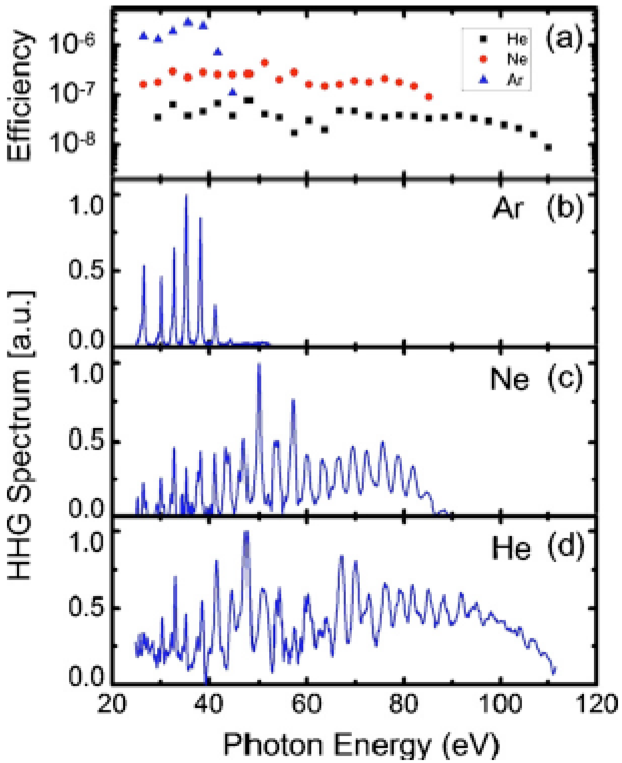

FIG. 2. (Color online) HHG driven by 800 nm, 35 fs driver pulses: (a) conversion efficiencies for Ar (50 mbar, 0.6 mJ), Ne (300 mbar, 2.0 mJ), and He (2.0 bar, 2.0 mJ) using a nozzle positioned 1 cm before the focus for all the gases; (b)-(d) HHG spectra, respectively, for Ar, Ne, and He.

$$\eta = 0.0236 \frac{\sqrt{2I_p}\,\omega_0^5 |a_{\mathrm{rec}}|^2 |g(\Delta k, L)|^2}{E_0^{16/3}\Omega_{\mathrm{cutoff}}^2 \sigma^2(\Omega_{\mathrm{cutoff}})} \frac{1-\beta^{4(N-1)}}{(1-\beta^4)N} \times |1+\beta|^2 \kappa_0 w[E(tb_{\mathrm{cutoff}})],$$

where

$$g(\Delta k, L) = [e^{i(\Delta k \cdot L)} - e^{-L/(2 \cdot L_{\mathrm{abs}})}]/[1 + 2i(\Delta k \cdot L_{\mathrm{abs}})]$$

phase-mismatch form factor with a phase mismatch of Δk.

Falcão-Filho, (2010). *Appl. Phys. Lett. 97*(6), 061107

N	is the number of cycles of the driver pulse
$\beta = \|a(\pi/\omega)\|^2$	denoting the probability to find the atom in the ground state

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CEP Stabilization

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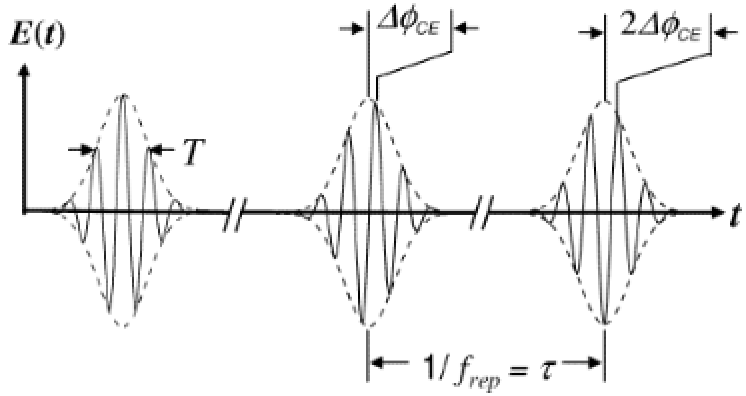
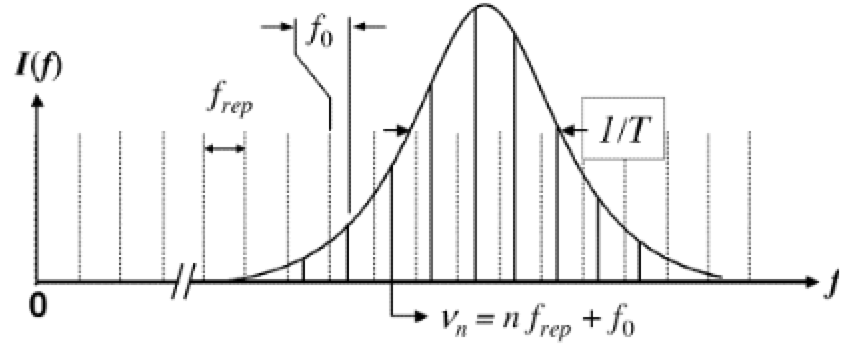
CE Phase

a) Time domain



IEEE J. Sel. Top. Quant. Electron. 9,1002 (2003)

(b) Frequency domain

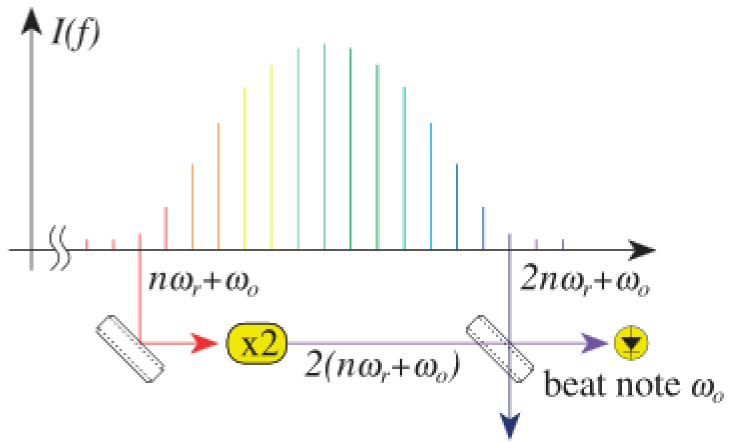


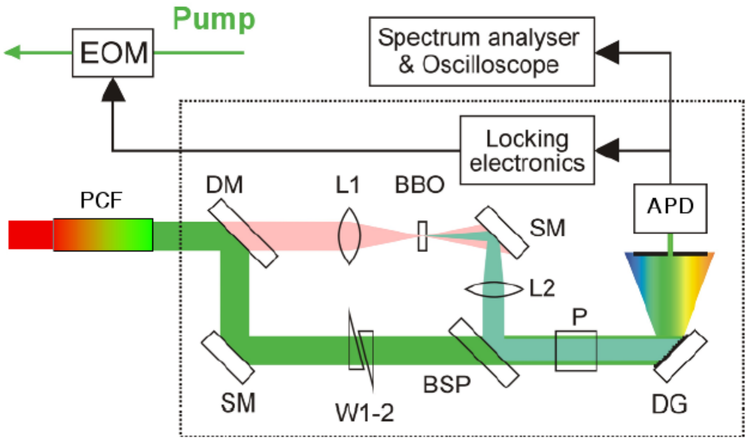
$$f_0 = f_{rep} \cdot \Delta\varphi_{CE} / 2\pi$$

- Oscillator output
  - Comb = modes
  - f_0 = comb frequency offset
  - $\Delta\varphi_{CE}$ = CE Offset (CEO)

Frequency domain representation

stabilization of the CEP phase



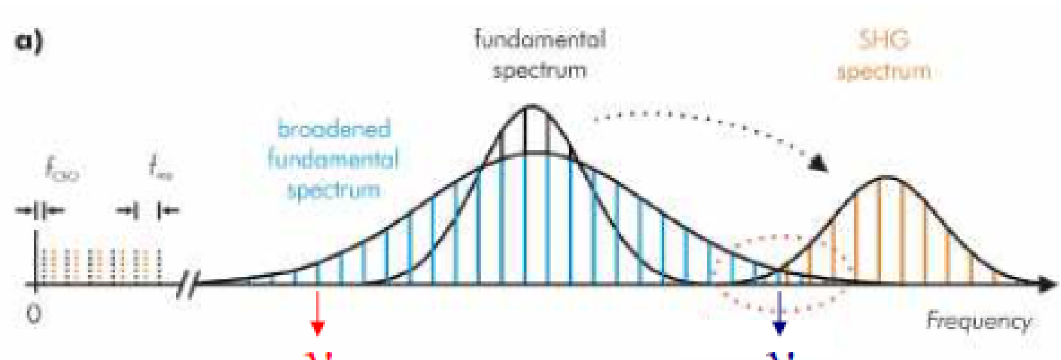


$$E_{\text{fund}}(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\sqrt{I_{\text{fund}}(\omega)}\exp[i(\varphi_{\text{fund}}(\omega) - \omega t + \phi)]\mathrm{d}\omega + cc$$

$$E_{\text{SH}}(t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\sqrt{I_{\text{SH}}(\omega)}\exp[i(\varphi_{\text{SH}}(\omega) - \omega(t+\tau) + 2\phi)]\mathrm{d}\omega + cc$$

$$\begin{aligned} S(\omega) &= I_{\text{fund}}(\omega) + I_{\text{SH}}(\omega) \\ &\quad +2\sqrt{I_{\text{fund}}(\omega) I_{\text{SH}}(\omega)}\cos(\varphi_{\text{SH}}(\omega) - \varphi_{\text{fund}}(\omega) + \omega\tau + \phi) \end{aligned}$$

- f-2f interferometer
  - Spectral broadening
    - ν_n and ν_{2n}
    - CEP preserving?



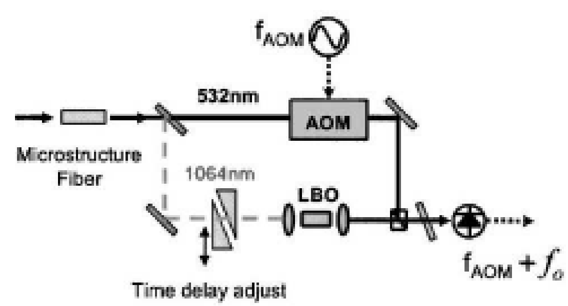
  - Frequency doubling of "red" part

    $$\nu_n = n.f_{rep} + f_0 \longrightarrow 2\nu_n = 2(n.f_{rep} + f_0)$$

  - Beat note with "blue" part

    $$2\nu_n - \nu_{2n} = 2(n.f_{rep} + f_0) - (2n.f_{rep} + f_0) = f_0$$

    - Mach-Zehnder interferometer
    - Single point detection



J. Opt. Soc. Am. B 21, 1098 (2004)

  - Self referencing: loss of absolute phase value

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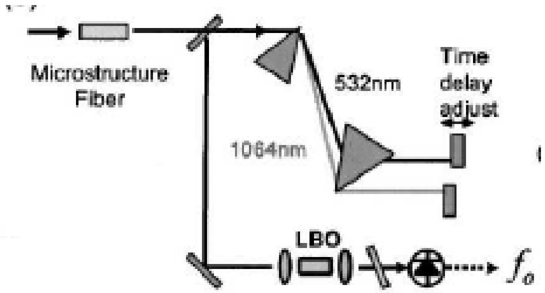
- Broadening in PCF
  - AM/PM in fiber Opt. Lett. 27, 445 (2002)
  - Octave spanning oscillators
- MZ stability
  - Michelson Opt. Lett. 31, 1011 (2006)
    - Still bulky
  - Common path interferometer J. Opt. Soc. Am. B 21, 1098 (2004)
    - Small group delay compensation range
  - Wollaston prisms Opt. Lett. 35, 1209 (2010)
  - Stabilization Opt. Express 14, 9758 (2006)
- 2 steps in 1
  - Broadening + SHG in ZnO layers Opt. Lett. 27, 2127 (2002)
  - Broadening + DFG in PPLN Opt. Lett. 30, 332 (2005)
    - No fiber, no interferometer
    - Useful spectrum
    - <7fs pulses



J. Opt. Soc. Am. B 21, 1098 (2004)

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alternative measurement method: stereo ATI

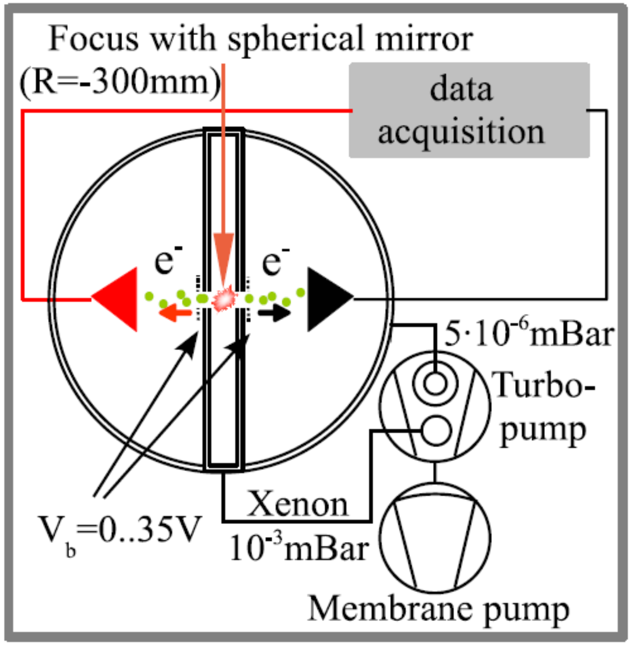
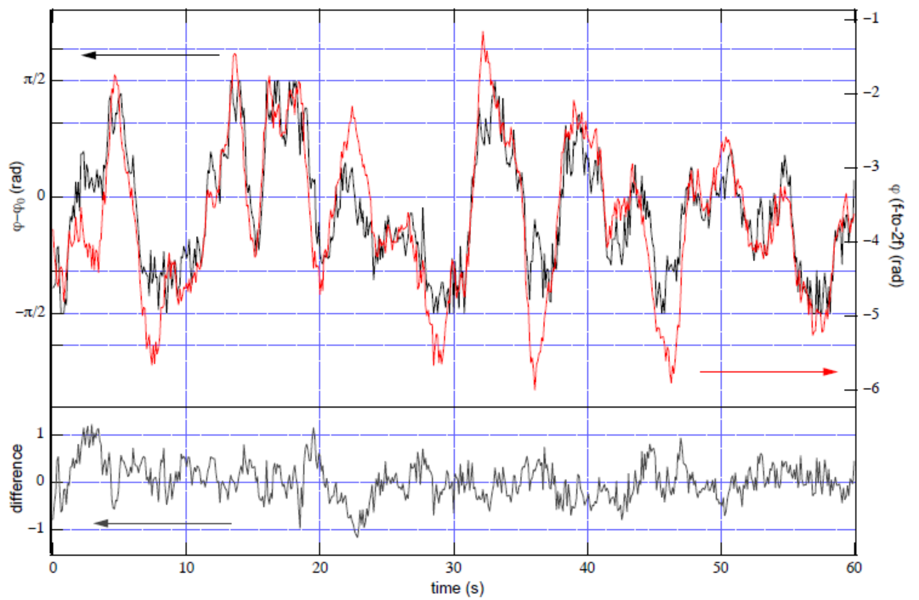


Figure 2.8: Schematic of the mini stereo ATI apparatus.

$$\phi = \arcsin\left(A\,\frac{\mathrm{N_{left}} - \mathrm{N_{right}}}{\mathrm{N_{left}} + \mathrm{N_{right}}}\right) + \phi_0$$



red: f-to-2f black: Stereo ATI

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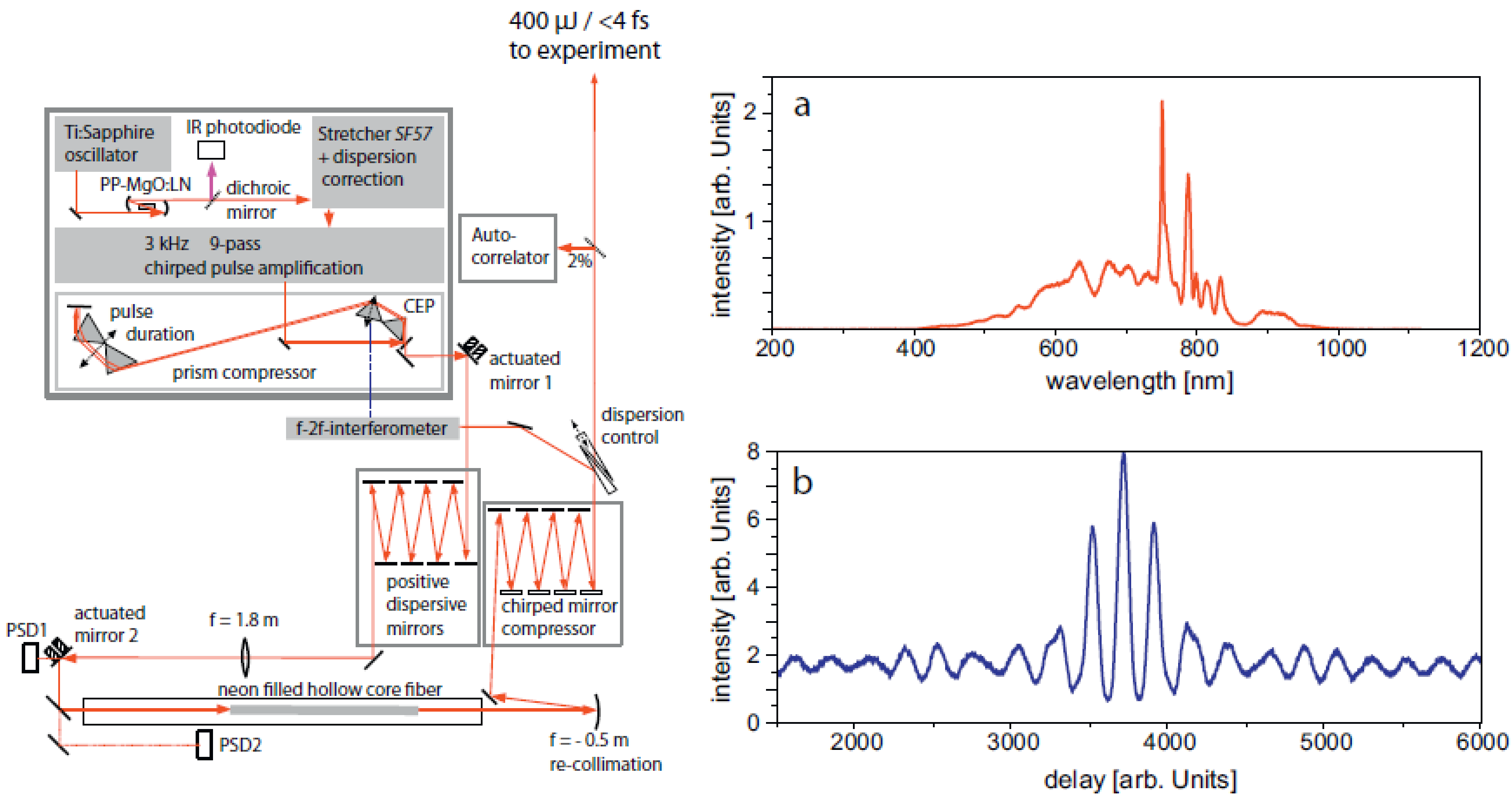
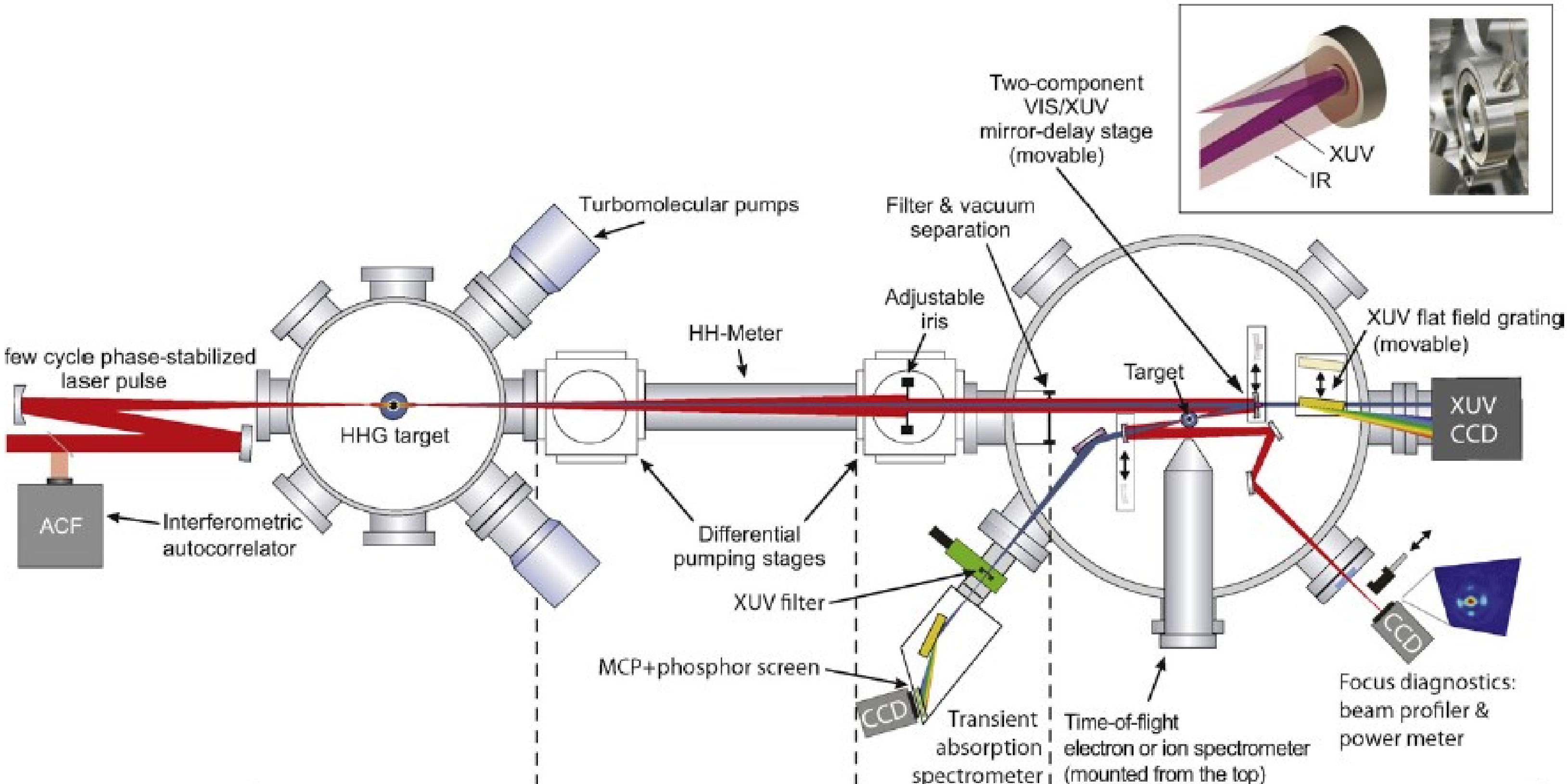
typical laser systems for sub-fs spectroscopy





Background pressure: 5×10^{-7} mbar	Background pressure: $<1\times10^{-6}$ mbar	Background press.: $<5\times10^{-8}$ mbar	Background pressure: $<1\times10^{-9}$ mbar
Operating pressure: $<3\times10^{-2}$ mbar HHG target pressure: ~350 mbar	Operating pressure: $<4\times10^{-4}$ mbar	Operating pressure: $<1\times10^{-6}$ mbar	Operating pressure: $<1\times10^{-5}$ mbar (electron and ion spectroscopy) 1×10^{-3} mbar (XUV absorption spectroscopy)

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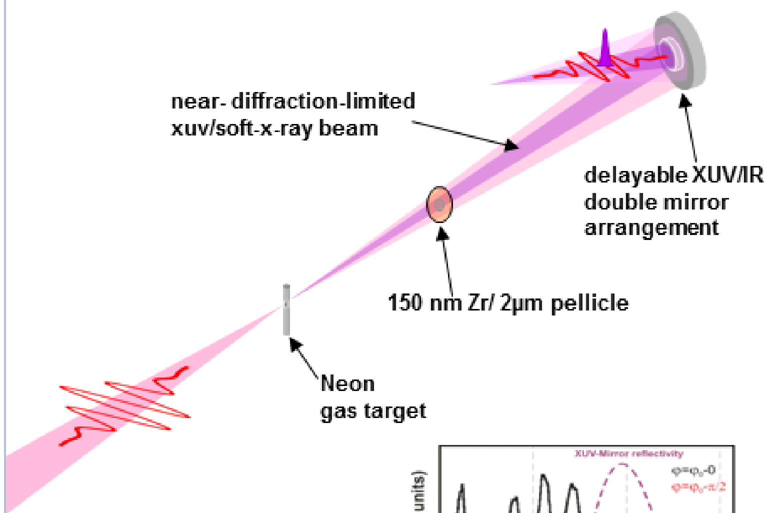
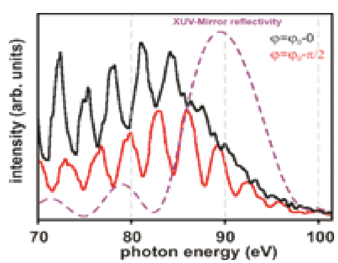
bandwidth filter for cut-off hamonics



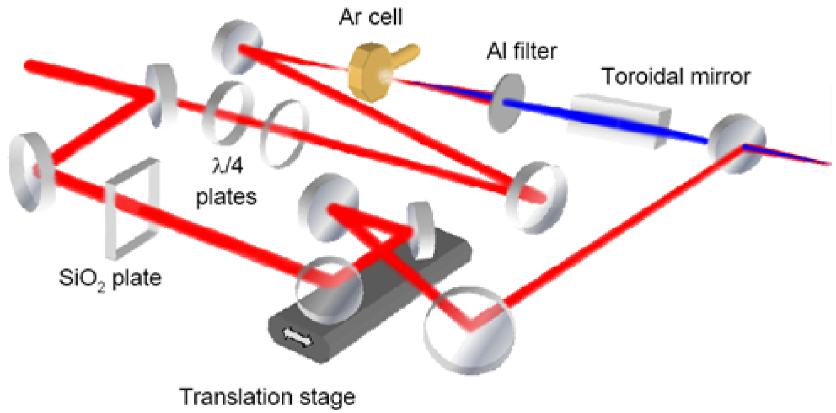
pulse duration τ <5 fs
pulse energy ~0.5 mJ
phase stabilized



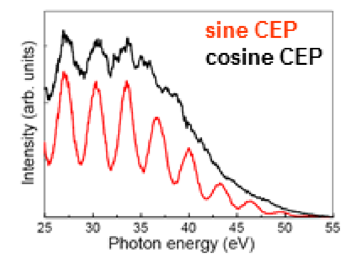
- Co-Linear propagation
- high photon energy
- virtualy jitter-free setup
- XUV bandwidth determined by pulse duration
- cut-off very sensitive on pulse fluctuations
- minimum pulse duration determined by mirror reflectivity

MPQ

Polarization Gating



pulse duration τ = 5 fs;
phase stabilized
Gate width δ= 6.2 fs



- interferometric setup
- high intensity losses due to the technique
- low photon energy
- XUV bandwidth determined by gate width
- pulse duration only limited by low pass filter
- sensitive on mechanical vibrations

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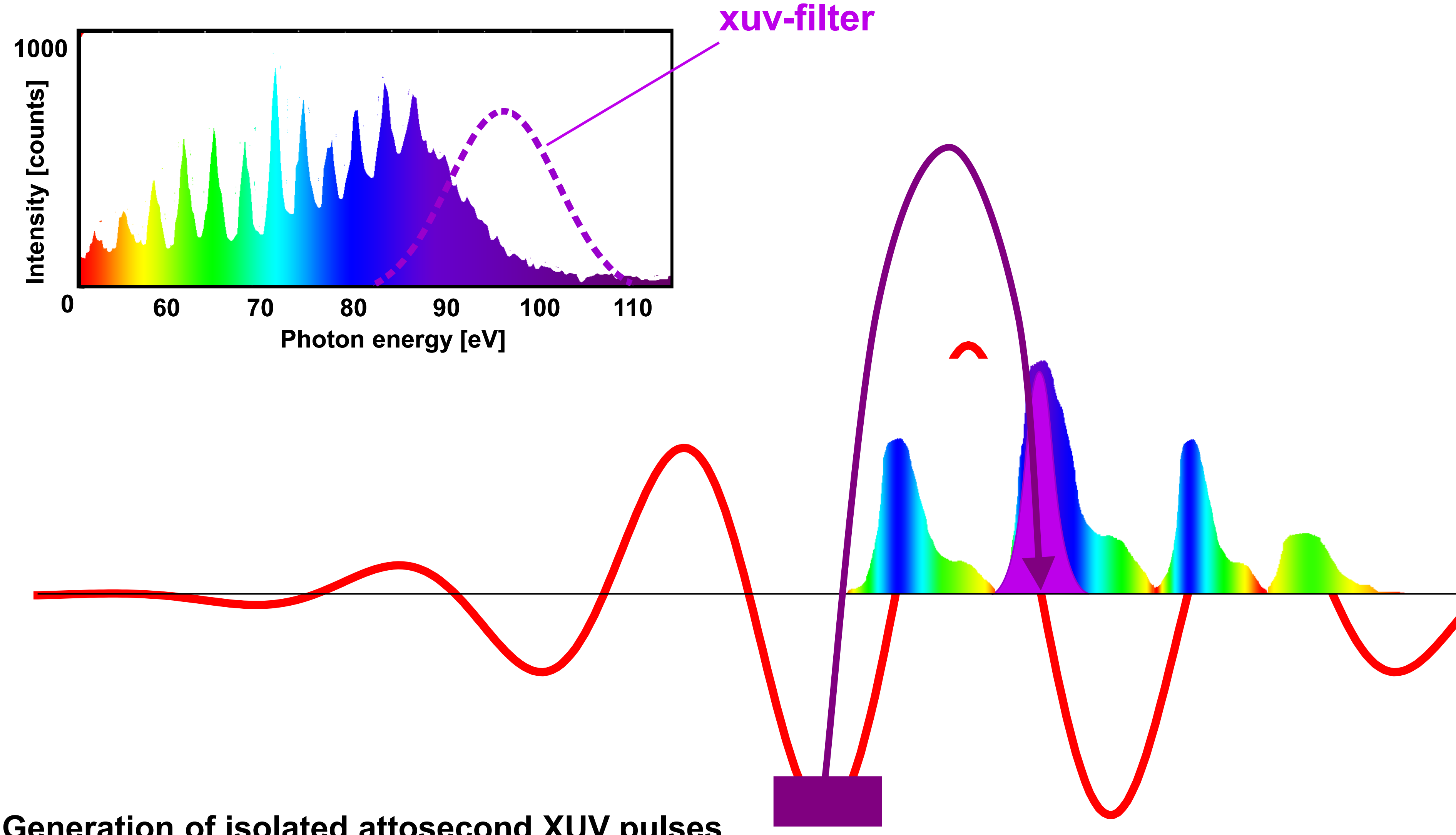

Generation of isolated attosecond XUV pulses

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Multilayer mirror – principle of constructive interference

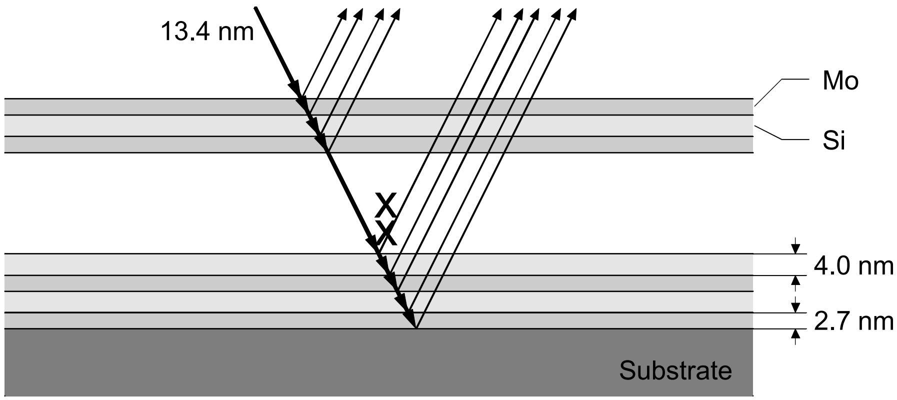
Bragg:

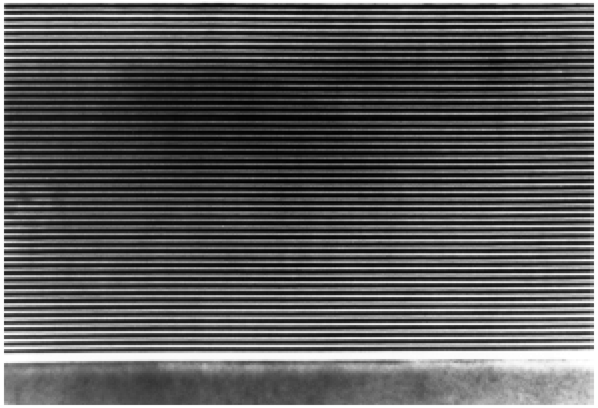
$\lambda = 2d \cos \alpha$

$d \sim 1/\cos \alpha$

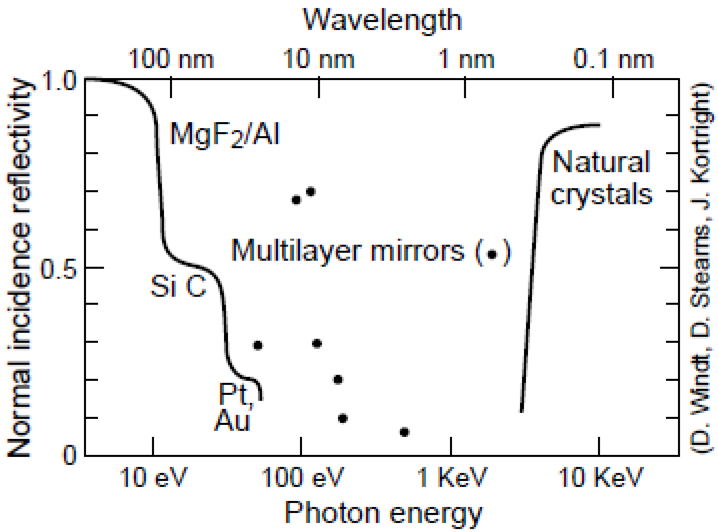
d … optical thickness of period

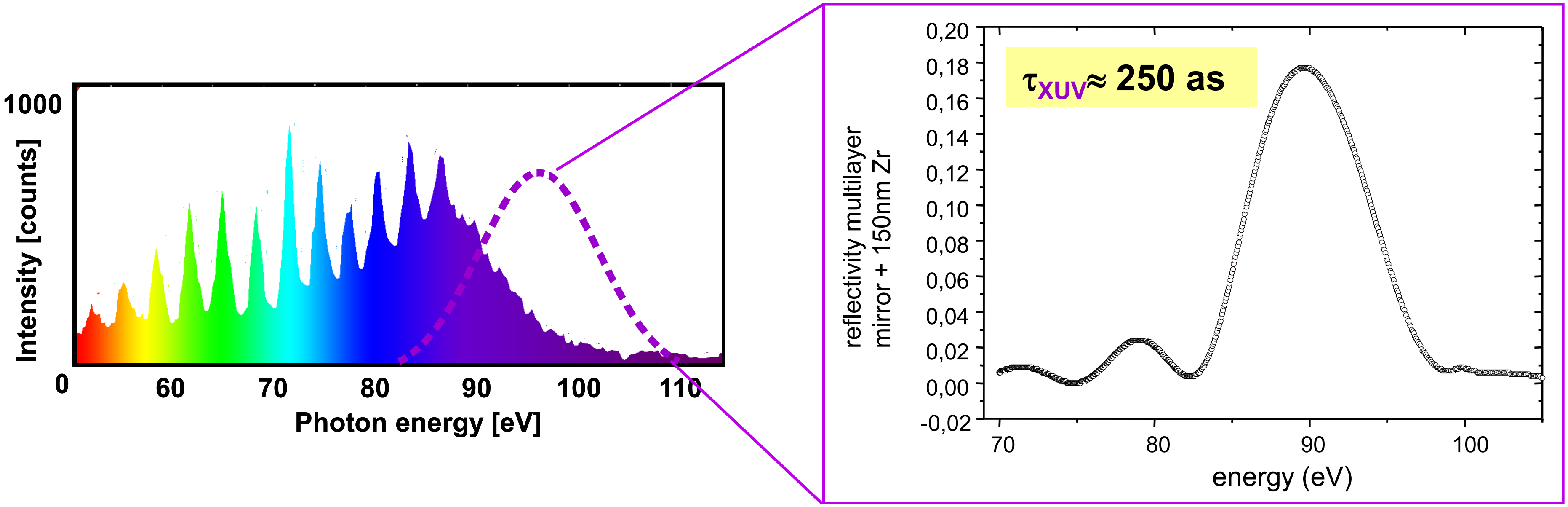
α …angle of incidence

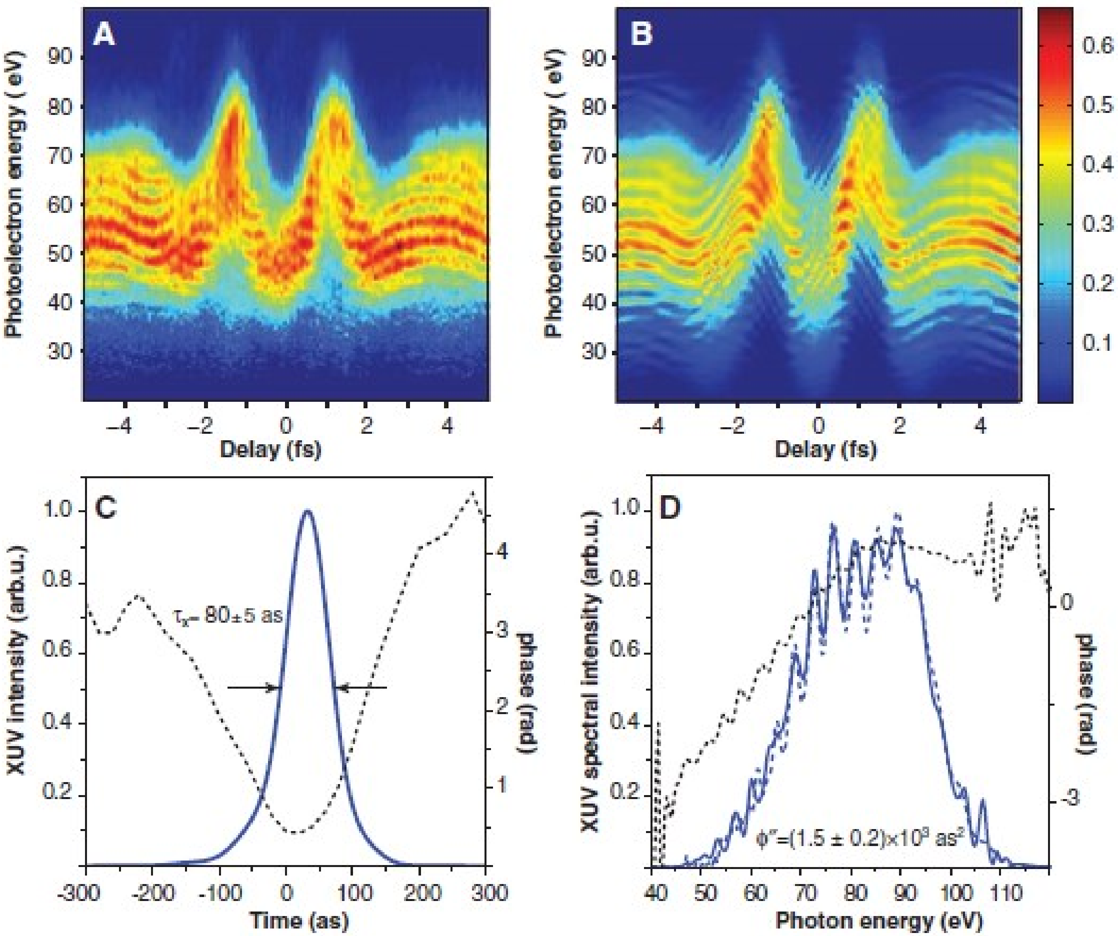




(W/C, T. Nguyen)




1000
Intensity [counts]
0
60
70
80
90
100
110
Photon energy [eV]
0,20
0,18
0,16
0,14
0,12
0,10
0,08
0,06
0,04
0,02
0,00
-0,02
reflectivity multilayer
mirror + 150nm Zr
70
80
90
100
energy (eV)
$\tau_{XUV} \approx$ 250 as



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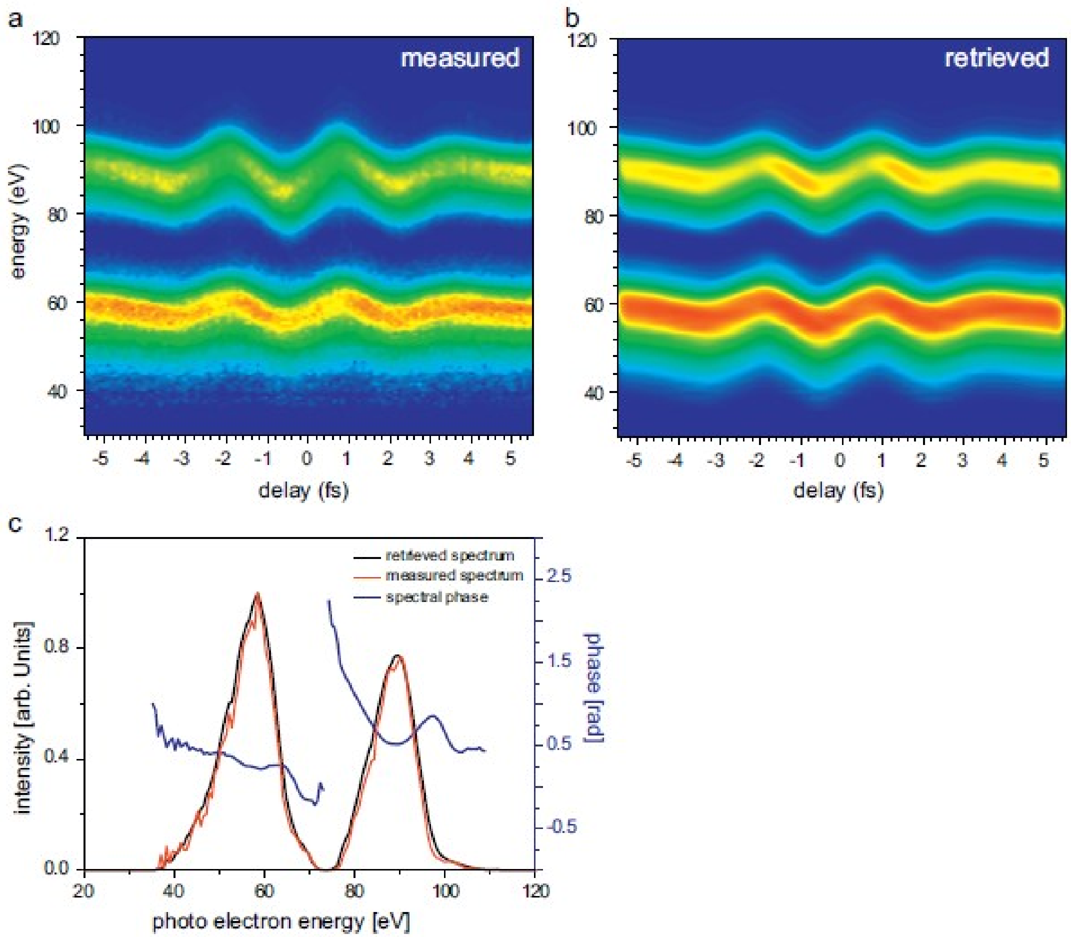

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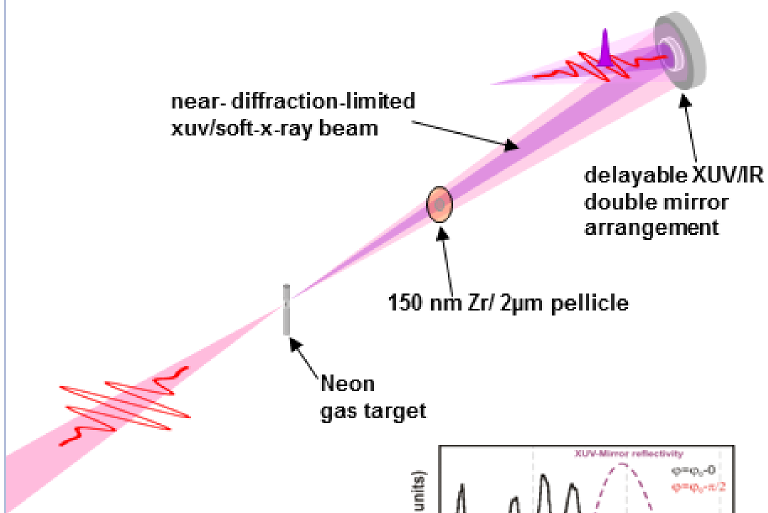
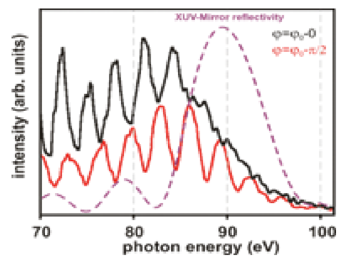
bandwidth filter for cut-off hamonics



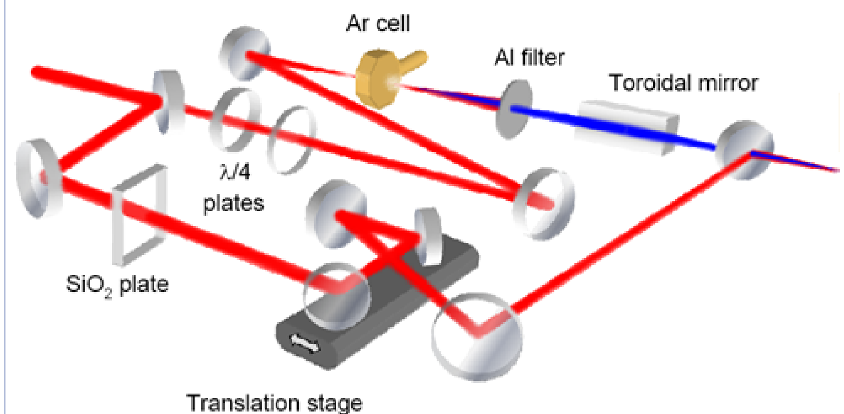
pulse duration τ <5 fs
pulse energy ~0.5 mJ
phase stabilized



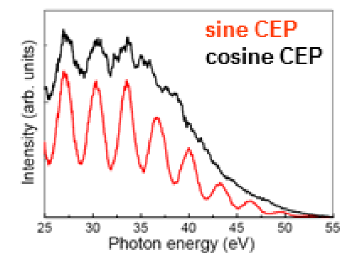
- Co-Linear propagation
- high photon energy
- virtualy jitter-free setup
- XUV bandwidth determined by pulse duration
- cut-off very sensitive on pulse fluctuations
- minimum pulse duration determined by mirror reflectivity

MPQ

Polarization Gating



pulse duration τ = 5 fs;
phase stabilized
Gate width δ= 6.2 fs



- interferometric setup
- high intensity losses due to the technique
- low photon energy
- XUV bandwidth determined by gate width
- pulse duration only limited by low pass filter
- sensitive on mechanical vibrations

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Generation of isolated Attosecond Pulses by „Polarization Gating“

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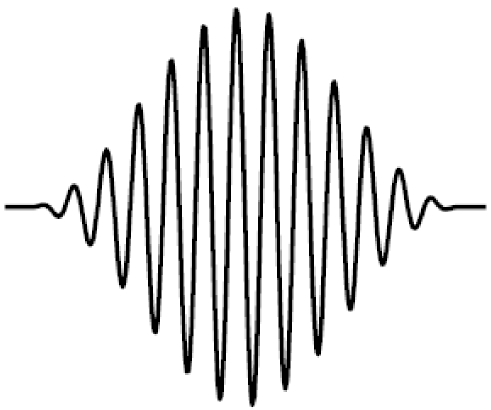
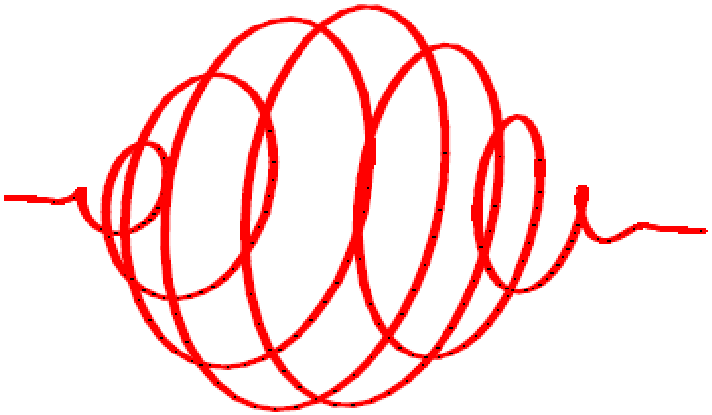
linear polarisation



electron returns to the parent ion:

emission of high harmonic radiation possible

circular polarisation



electron does *not* return to the parent ion:

emission of high harmonic radiation hardly suppressed

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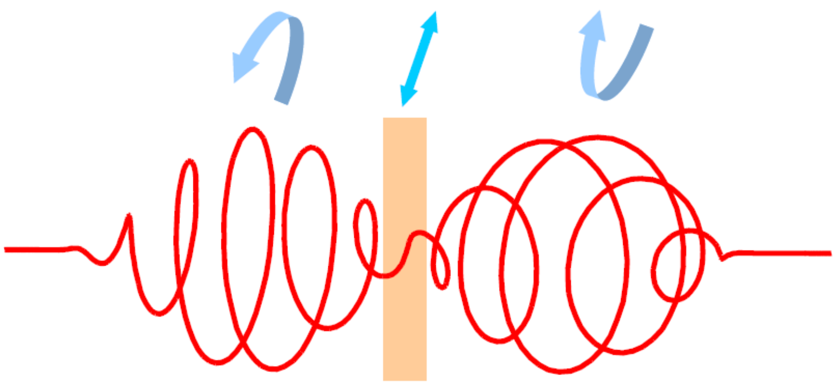
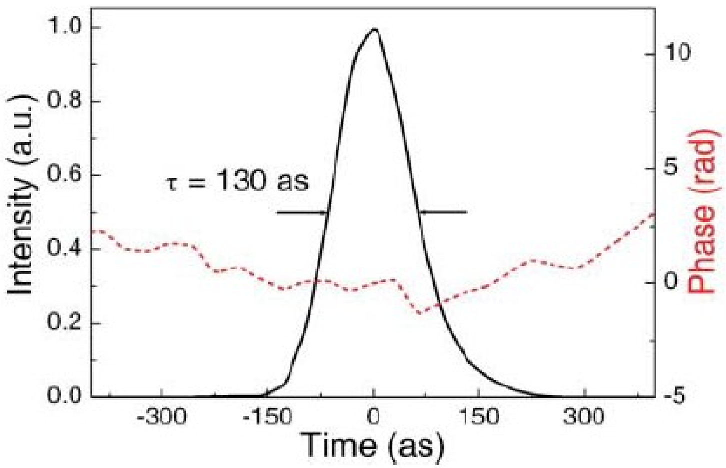
time-dependent polarisation gate



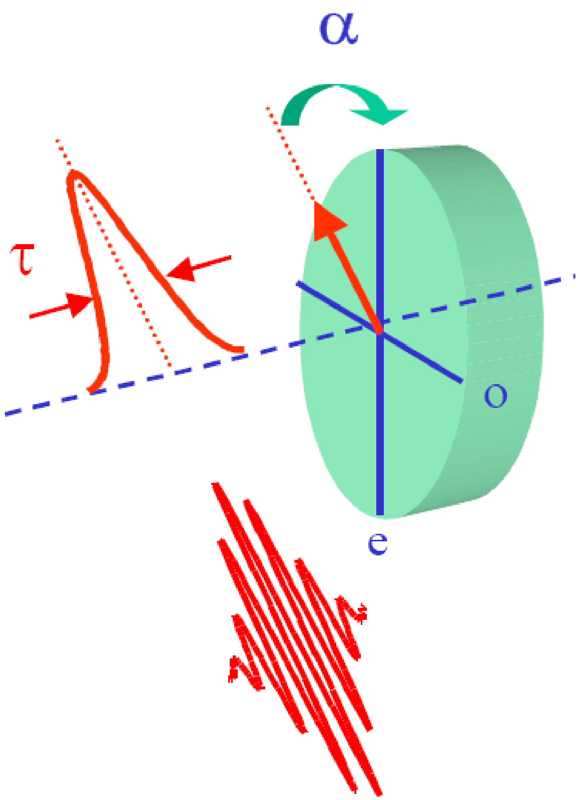


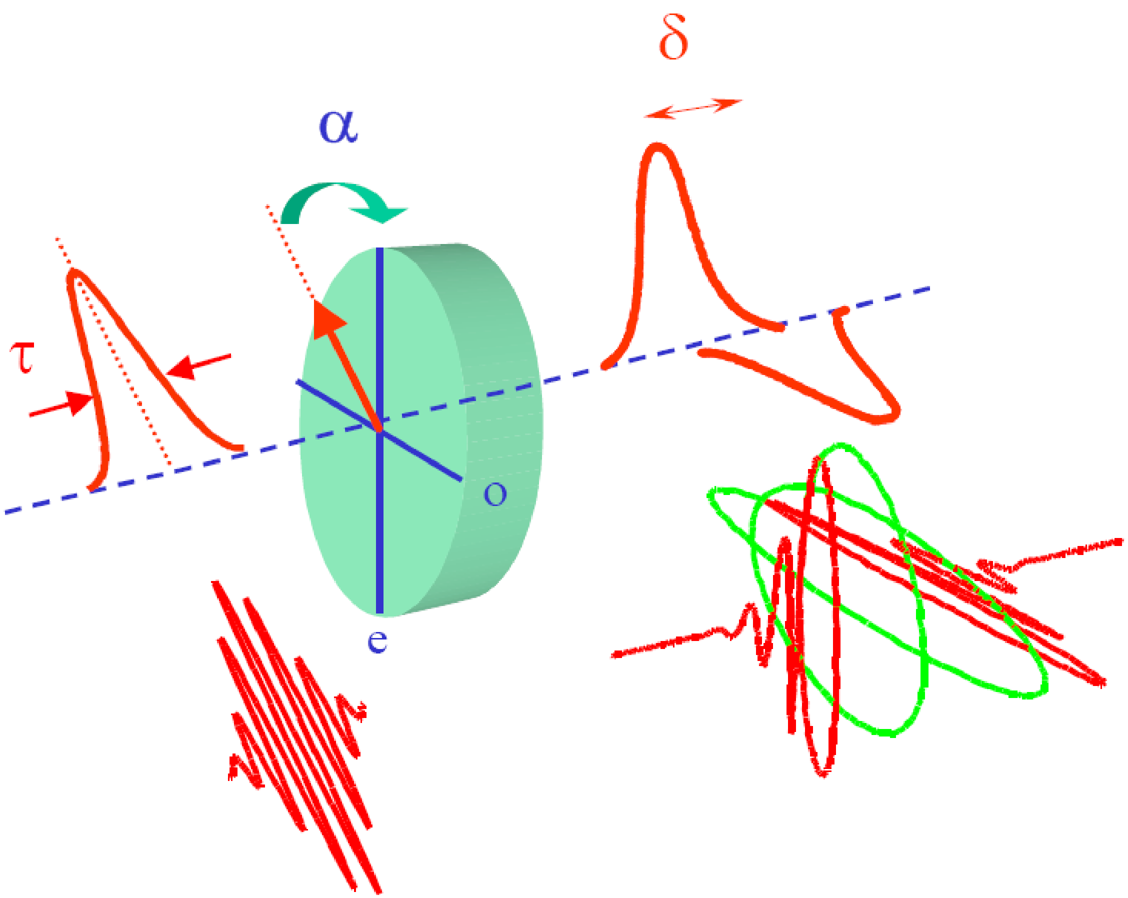
Generation of XUV continuum with PG applying:

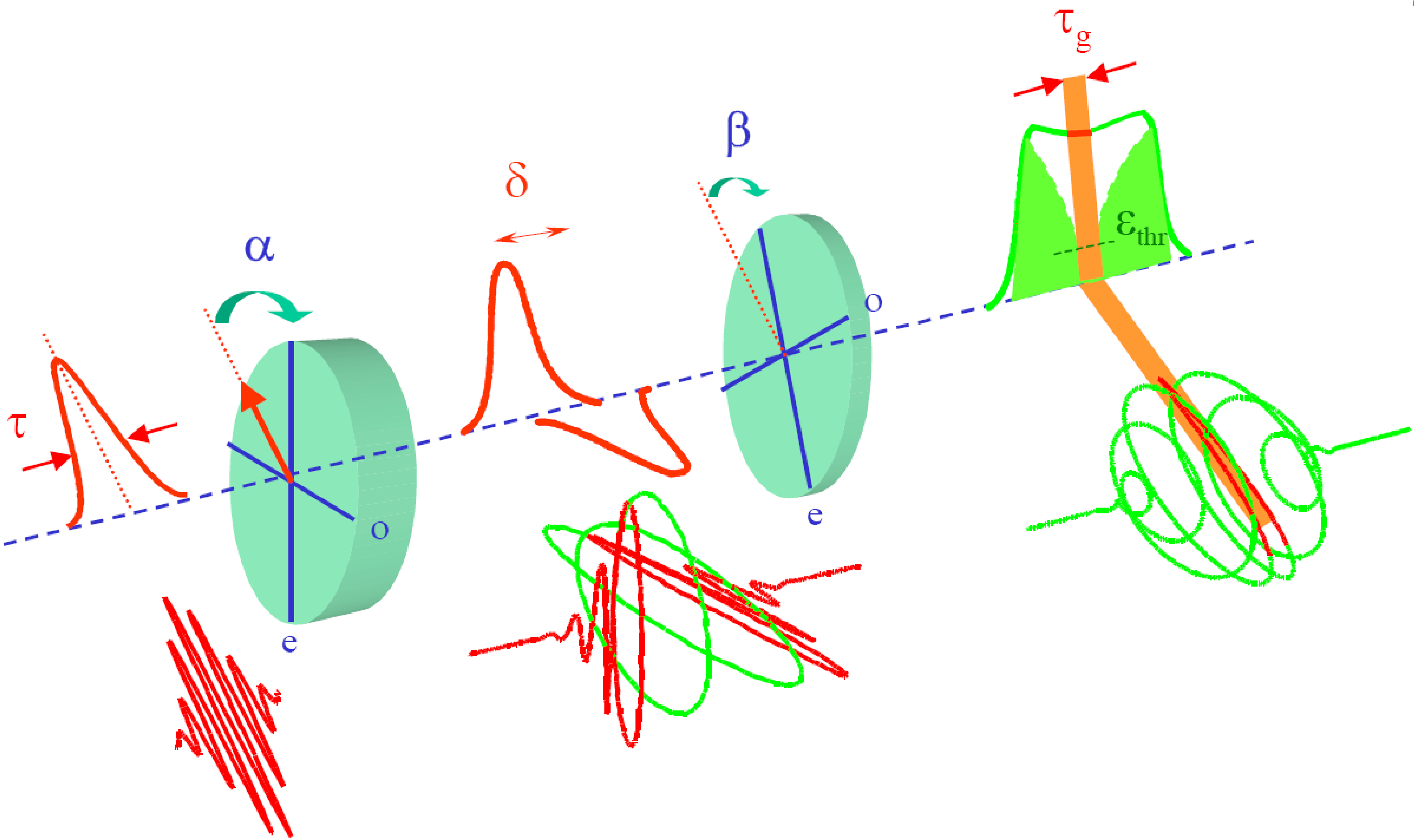
G. Sansone et al., Science 314,443 (2006)

- few-cycle pulses
- Carrier Envelope Phase stabilization
- dispersion control

P. Corkum *et al.*,Opt. Lett. **19,**1870 (1994)
O. Tcherbakoff*et al.*,Phys. Rev. A **68,**043804 (2003)


α
τ
o
e


δ
α
τ
o
e



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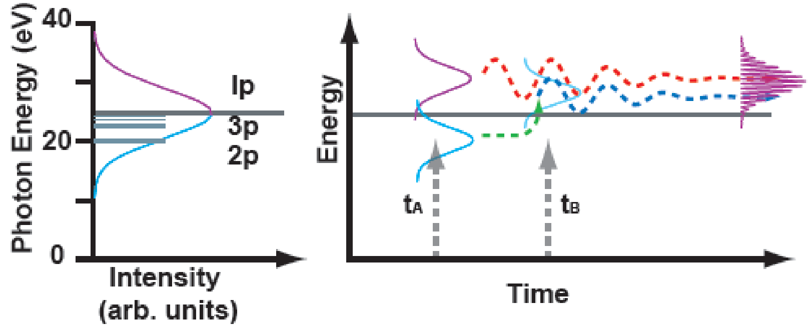

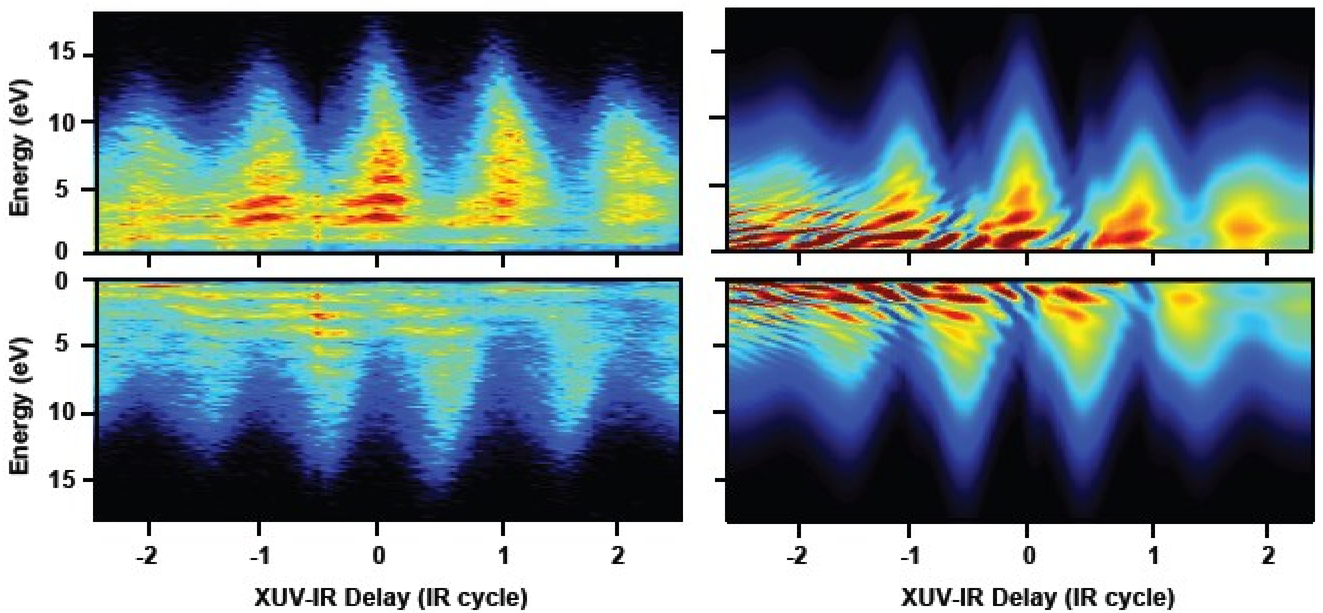

Fig. 2. experimental (left) and TDSE (right) results

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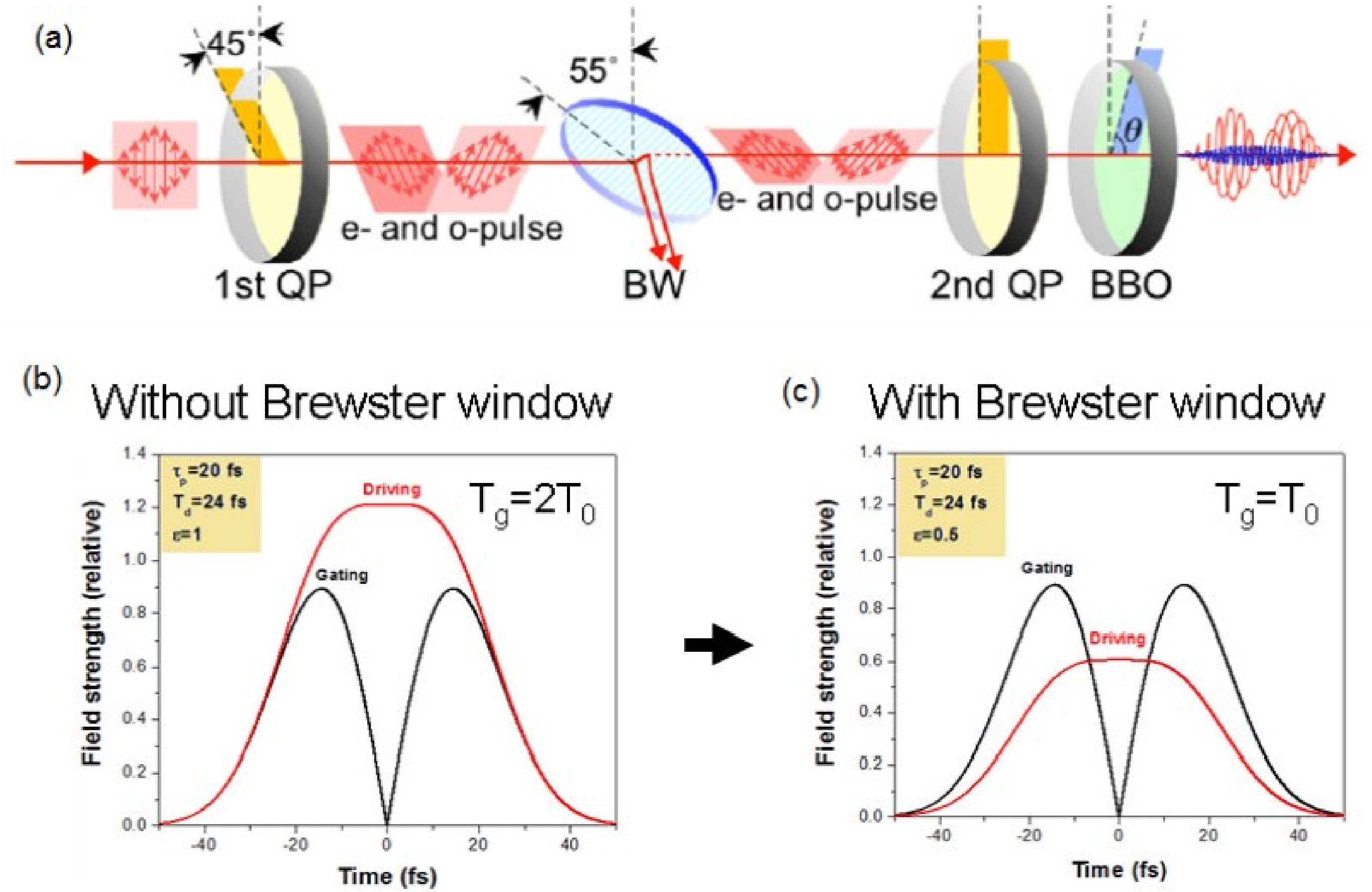
generalized double optical gating



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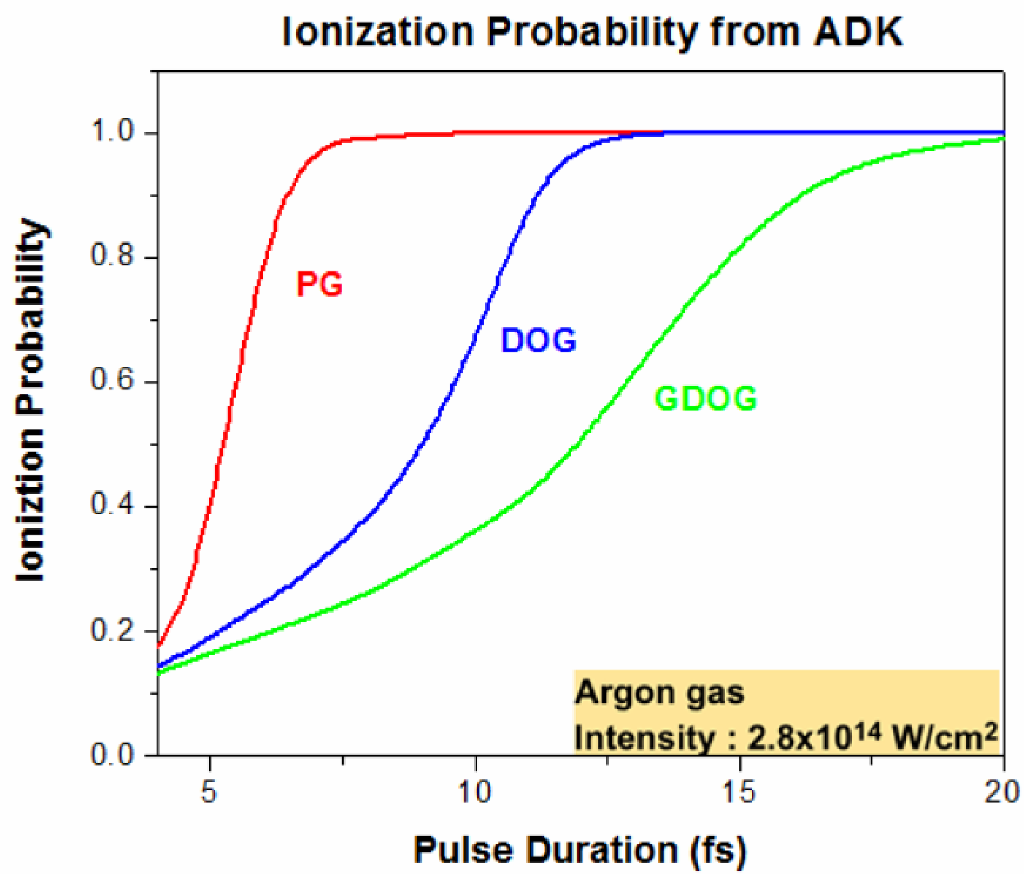
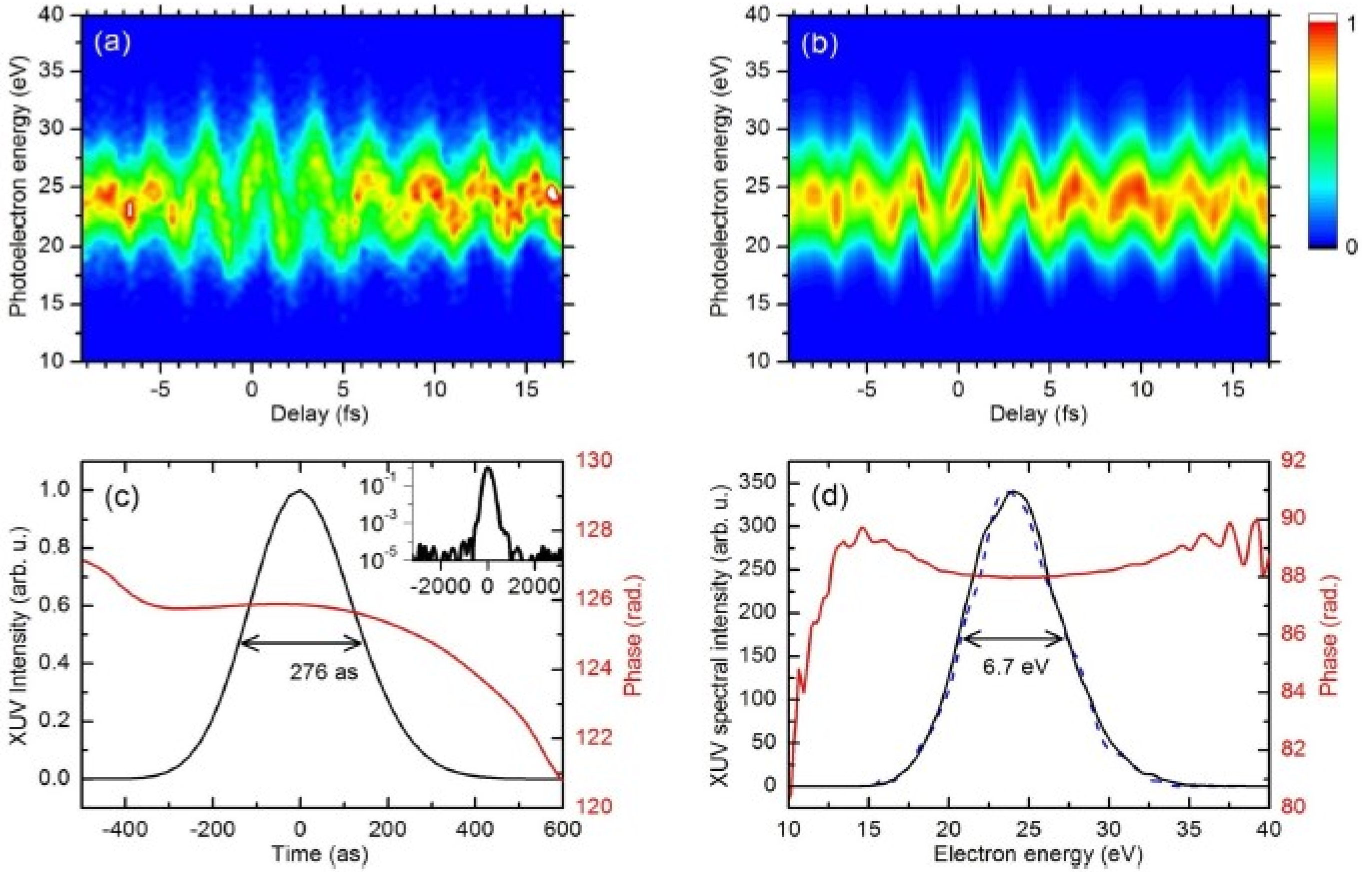
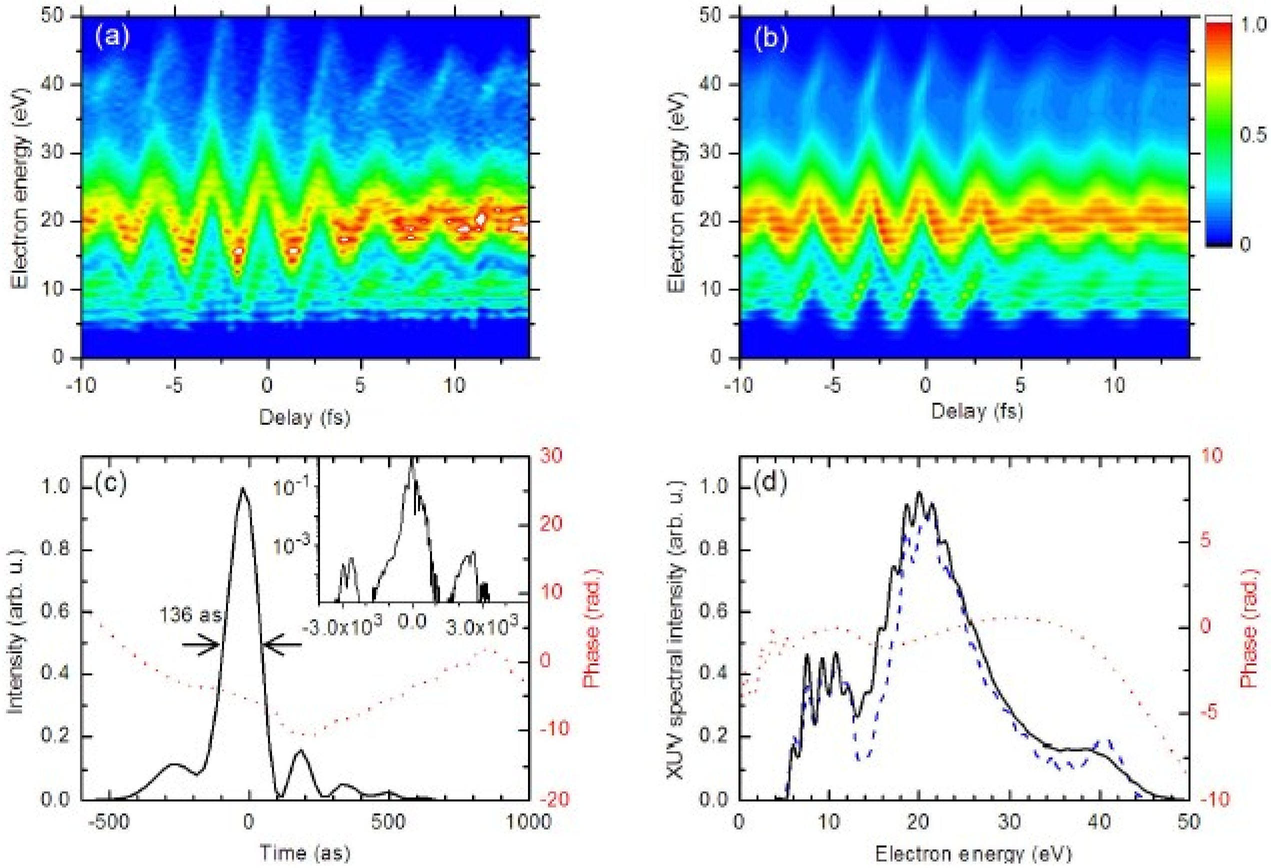

Ionization Probability from ADK
Ioniztion Probability
0.0
0.2
0.4
0.6
0.8
1.0
5
10
15
20
Pulse Duration (fs)
PG
DOG
GDOG
Argon gas
Intensity : 2.8x10^14 W/cm^2


(a)
(b)
(c)
(d)
Photoelectron energy (eV)
Delay (fs)
XUV Intensity (arb. u.)
Time (as)
276 as
Phase (rad.)
XUV spectral intensity (arb. u.)
Electron energy (eV)
6.7 eV


(a)
(b)
Electron energy (eV)
Delay (fs)
1.0
0.5
0
(c)
136 as
Intensity (arb. u.)
Phase (rad.)
Time (as)
-3.0x10^3
0.0
3.0x10^3
(d)
XUV spectral intensity (arb. u.)
Electron energy (eV)

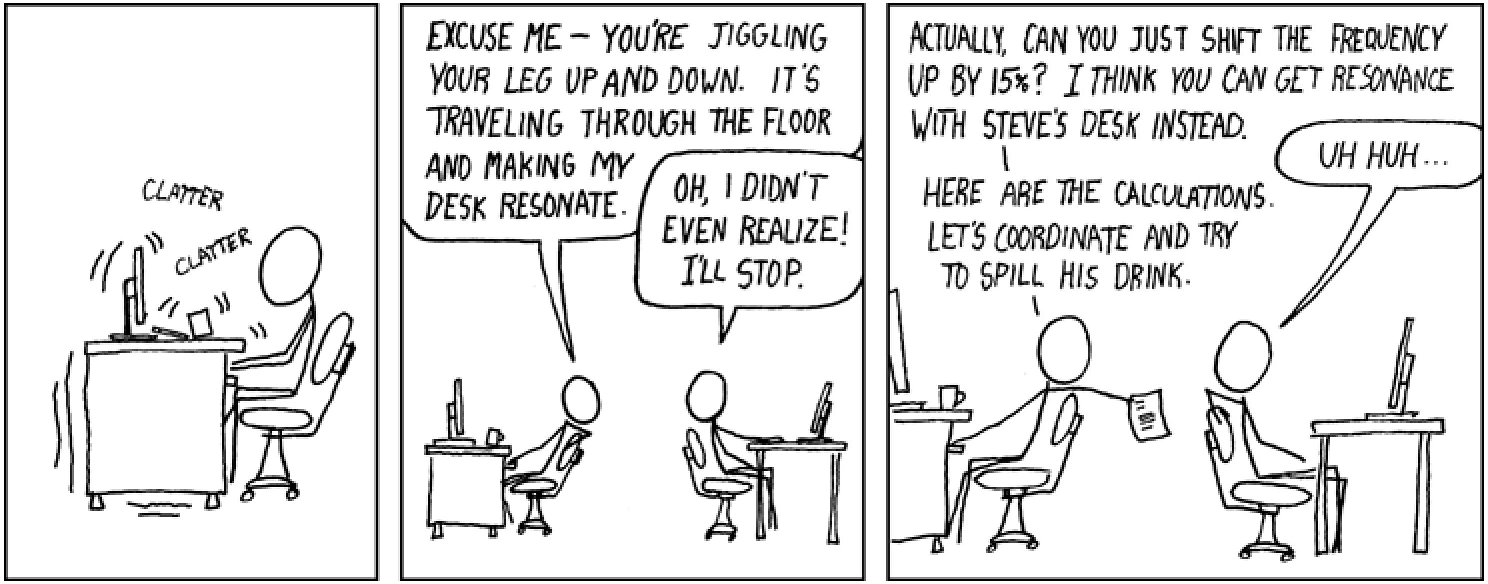
Comparison of different methods

Table 1 | Relevant parameters of current attosecond sources based on HHG in gases.

Method	Gas	$\hbar\omega$ (eV)	τ (as)	*E*	Type	Efficiency	Reference
AG	Neon	80	80	0.5 nJ	IAP	1.7×10^{-6}	22
PG	Argon	35	130	70 pJ	IAP	3.5×10^{-7}	26,27
PG	Neon	60	—	1 pJ	IAP	5×10^{-9}	26
DOG	Argon	38	—	6.5 nJ	IAP	6×10^{-6}	31
DOG	Neon	45	107	170 pJ	IAP	1.5×10^{-7}	31,85
IG	Xenon	27	155	9 nJ	IAP	2.6×10^{-5}	50
IPG	Xenon	50	<1,000	20 nJ	IAP	2×10^{-5}	36
LF	Xenon	20	320	10 μJ	APT	7×10^{-4}	16

$\hbar\omega$ is the central energy of the XUV spectrum, τ is the measured pulse duration and *E* is the pulse energy at the source (in the case of isolated pulses, *E* corresponds to the XUV energy in the spectral region characterized by a continuous profile). The efficiency is defined as the ratio between the XUV pulse energy and the driving laser pulse energy. AG, amplitude gating; PG, polarization gating; DOG, double-optical gating; IG, ionization gating; IPG, interferometric polarization gating; LF, loose-focusing; IAP, isolated attosecond pulses; and APT, attosecond pulse trains.

Thank you!


CLATTER
CLATTER
EXCUSE ME – YOU'RE JIGGLING YOUR LEG UP AND DOWN. IT'S TRAVELING THROUGH THE FLOOR AND MAKING MY DESK RESONATE.
OH, I DIDN'T EVEN REALIZE! I'LL STOP.
ACTUALLY, CAN YOU JUST SHIFT THE FREQUENCY UP BY 15%? I THINK YOU CAN GET RESONANCE WITH STEVE'S DESK INSTEAD.
HERE ARE THE CALCULATIONS. LET'S COORDINATE AND TRY TO SPILL HIS DRINK.
UH HUH...