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Advanced School on Synchrotron and Free Electron Laser Sources and their Multidisciplinary Applications

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Ultrafast VUV and soft X-ray pulses (production by harmonic generation and features)

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OUTLINE

Introduction

Some basics

HG in crystals : where do we stop

Low-order HG in Gases

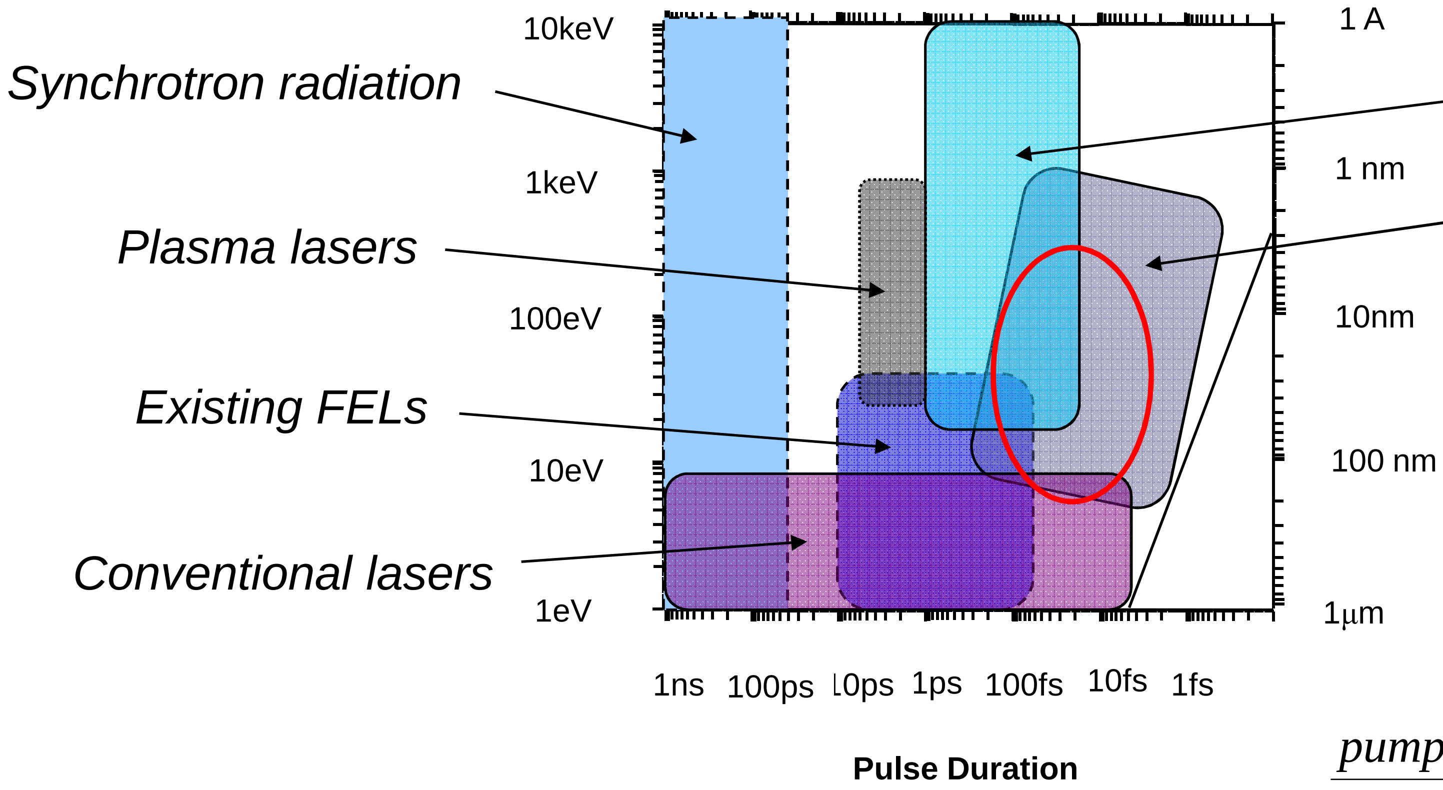
High order HG in Gases (HHG)

Trends in HHG

Features of the HHG light

INTRODUCTION

Main sources of laser-like beams/pulses in the 180-3 nm range



$$\frac{pump-power}{m^3} \sim \frac{10^{-20}}{\lambda^4 L}\sqrt{\frac{T}{Mn}}$$

Harmonic generation

Regimes:

1. Peak power density from 10^3 to 10^{13} W/cm^2 - **weak perturbation limit**

Nonlinear polarisation :

$$\vec{P} = \varepsilon_0 (\chi^{(1)} \vec{E} + \chi^{(2)} \vec{E}^2 + \chi^{(3)} \vec{E}^3 + \ldots)$$

Phase matching:

$n\mathbf{k}_{\omega} = \mathbf{k}_{n\omega}$

In NL crystals :

- PM by birefringence
- $\chi^{(2)} \sim 10^{-12}$ m/V

efficienciy of SHG (2ω)~50% ,THG(3ω)~20%,… FHG(5ω)~1%

Limits:

- transparency region : 6.9 eV BBO, ~8eV KBBF , ~10eV SBBO
- No phase matching for SHG , mixing with longer wavelengths

Shortest wavelengths: 157 nm , FHG of Ti: sapphire in KBBF (1)
130 nm , SHG (not in PM!) in SBBO

Low-order HG in gases

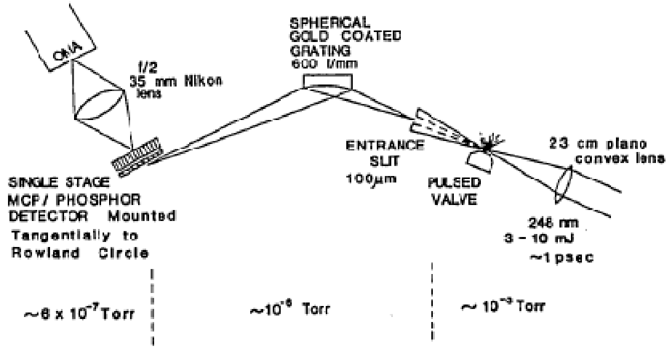
Features:
-Odd harmonics only
-Transmission in the VUV-EUV
-Low nonlinear susceptibility

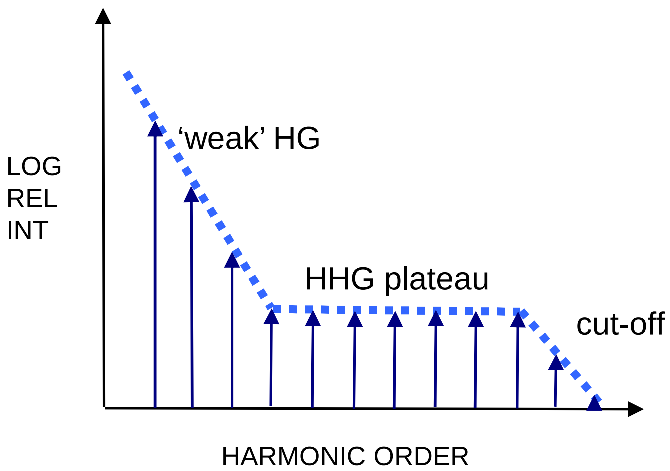
$$\eta = \frac{P_3}{P_1} = \frac{3\pi^2}{\varepsilon_0^2 c^2 \lambda_1^4} N^2 [\chi_{\lambda_3}^{(3)}]^2 P_1^2 |\Phi|^2$$ (in SI units)

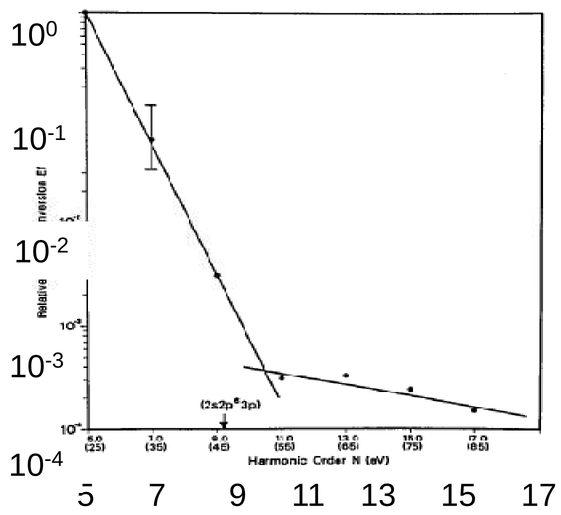
- Regions where Δk~0 (or small enough) can be found by adjusting gas mixture and focusing conditions
- Feasible for THG in the EUV starting with laser wavelength in the deep UV Excimer or Third harmonic YAG : 3% efficiency in generating 118. 2 nm (10.5 eV) in gas cell with mixture Xe:Ar;
Tuneable source (Nd:YAG harmonic+dye) for tuneable THG in gas jets
-> 10^{-4} efficiency in generating 70- 100 nm (~18 – 10 eV)
->Could be used down to 63 nm (~20 eV) by use of Ti:sapphire harmonics in BBO as a source to generate ~1 μJ per pulse (2 to 5×10^{11} ph/pulse)

HHG in gases

- At very high intensities (>10^{14} W/cm^2)
- the weak perturbation limit breaks ->plateau in the harmonic conversion efficiency







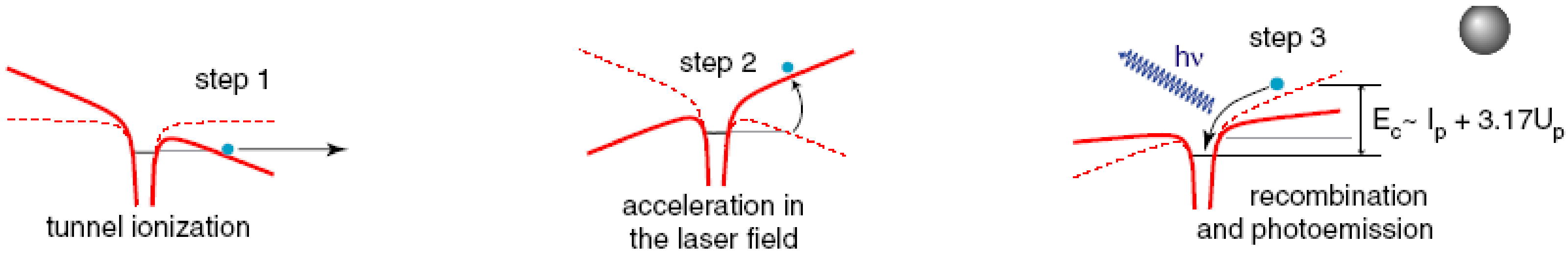
From McFerson et al, JOSA B 4 (1987), 595

HHG in gases

Theoretical Description:

-Semi-classical: The three-step model of HHG

Corkum Phys.Rev.Lett.**71** (1994); Kulander et al Proc.SILAP III, ed. B.Piraux (Plenum), 95-110.



->Correctly predicts most of the observed features

Cutoff: $h\nu_{max} = I_p + 3.2U_p$, where $U_p = e^2E^2/(4m\omega^2) = 9.33\times10^{-14}\ I_f\lambda_f^2$

eV W/cm^2 μm

-Fully quantum-mechanical treatment

Lewenstein et al, Phys.Rev.A 49 (1994), 2117

$$\Phi_q = q\omega t_f - S(p_{st}, t_i, t_f)$$

HHG in gases

Directions for parameter improvement

A. Phase-matching
   1. HG in waveguide (hollow fibre)
   2. Corrugated waveguide (QPM)
   3. Multiple gas jets (QPM)
   4. Counter-propagating beam (QPM)
   5. Non-adiabatic self-phase matching (HHG with very short pulses)
   6. Use of gas mixtures

B. Long wavelength excitation for increasing cut-off (pump source development)

C. Selective harmonic enhancement:
- HHG with temporally shaped pulses
- Bi-harmonic fields

D. Wavelength tuning:
- OPA pump
- Use of ‘blue’ shift with changing gas pressure

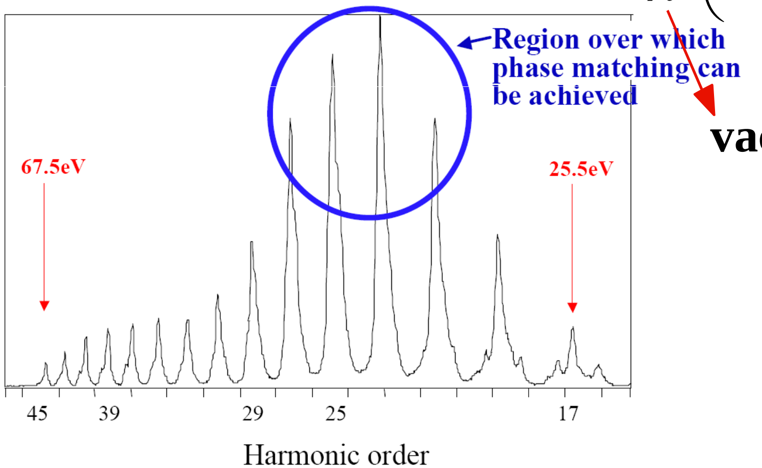
HHG in gases

A1. Phase matching in guided wave HHG

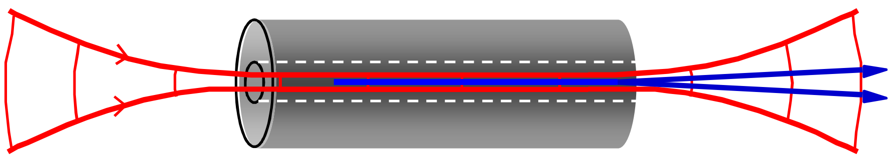
Phase mismatch $$\Delta k = qk_f - k_q = \Delta k_{gas} + \Delta k_{wg} + \Delta k_{plasma}$$

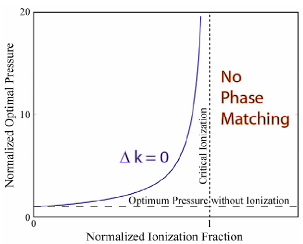
Fundamental $$k = \frac{2\pi}{\lambda}\left(1 + N_a\delta(\lambda) - \frac{1}{2}\left[\frac{U_{nm}\lambda}{2\pi a}\right]^2 - \frac{1}{2}\frac{N_e r_e \lambda^2}{\pi}\right)$$

vacuum **gas** **waveguide** **ionization**



Hollow fibre

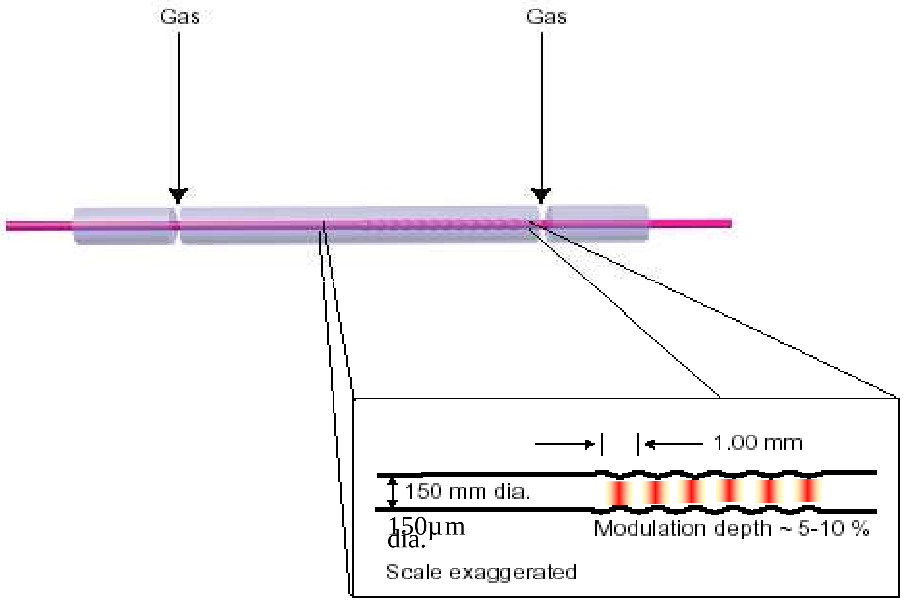


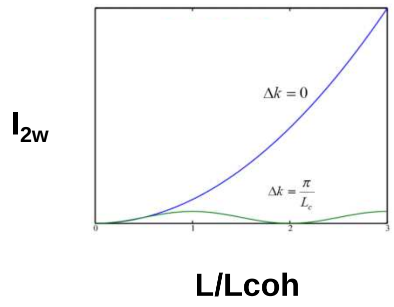


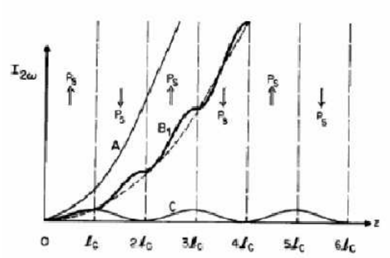
C. Durfee et al., Opt Lett 22, 1565 (1997)

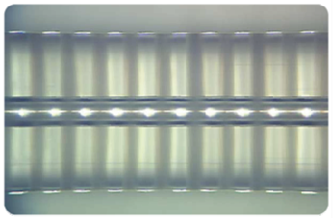
HHG in gases

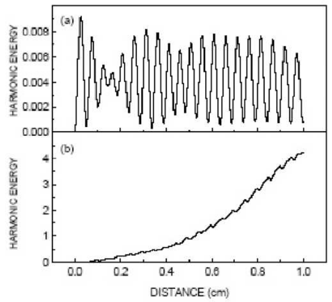
A.3. Quasi-Phase Matching (QPM) in waveguide HHG







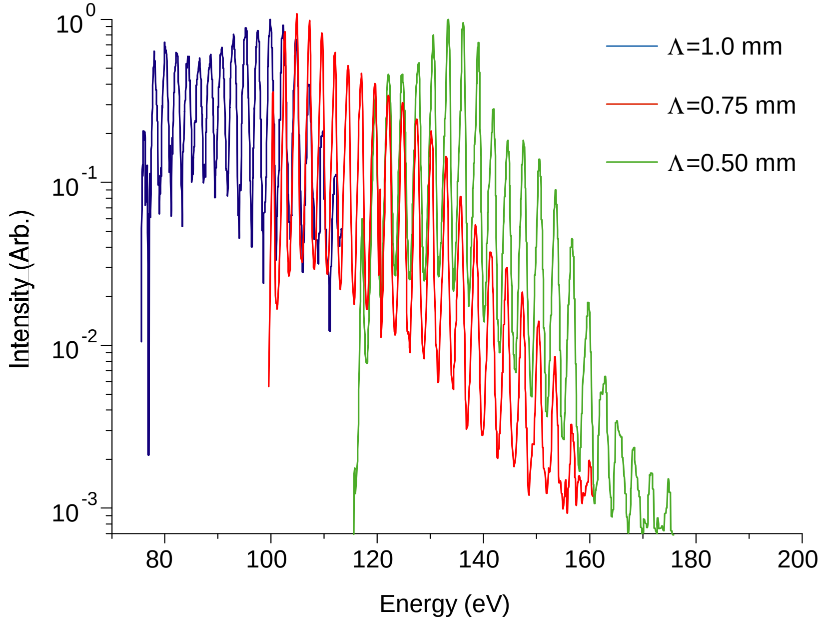




A. Paul et al, *Nature* **421** (2003), 51

I.P.Christov et al, *OptExpress* **7** (2000), 362

HHG in gases



Cut-off shift to higher energy by QPM in He, driving pulse 25 ps, Peak intensity ~5x10^{14} W/cm^2,

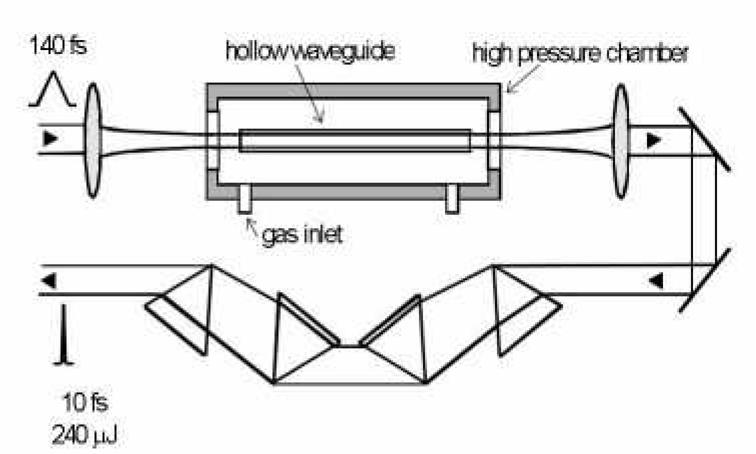
A. Paul et al, *Nature* **421** (2003) 51

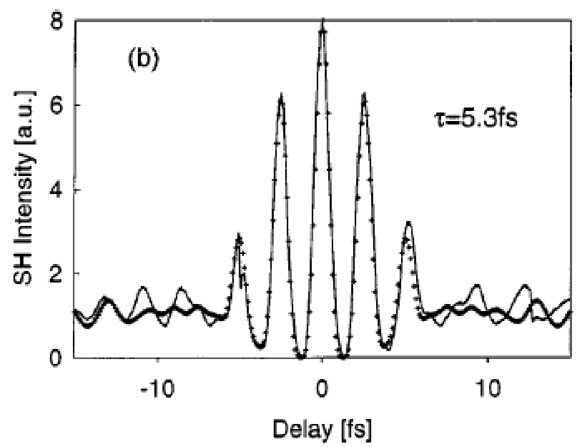
HHG in gases

A.4. Use of very short pulses: non-adiabatic regime

Few-cycle fundamental pulse (~5 fs at 800 nm)

(approach for generation proposed at Politecnico di Milano and Viena, demonstrated 5 fs , 70 µJ)





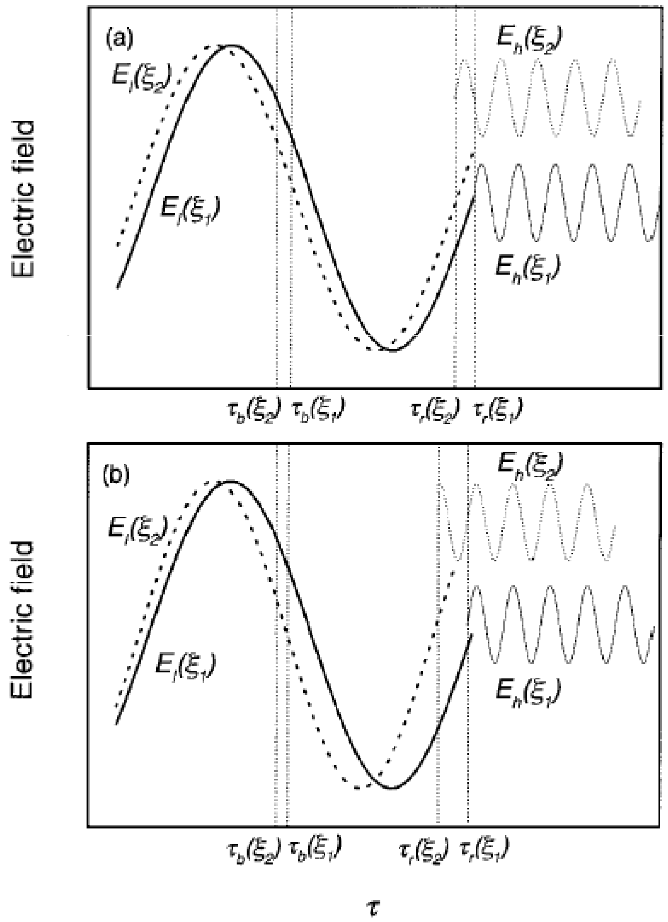
M. Nisoli et al, **Opt. Lett. 22**, 522 (1997)

HHG in gases

A.4. Non-adiabatic self-phase matching (NSPM)

Very short pulses focused to 0.2-1x10^{16} W/cm^{2}
Proposal and numerical simulations: Tempea et al, *Phys Rev Lett* **84** (2000)4329)
Experimental results : E.Seres et al, *Phys Rev Lett* **92 (2004), 163002**

Laser source: 5 fs, 300 μJ , focused to 30-40 μm
Medium: thin jet (0.5 mm), 0.5 bar



HHG in gases

B. Increase wavelength of excitation

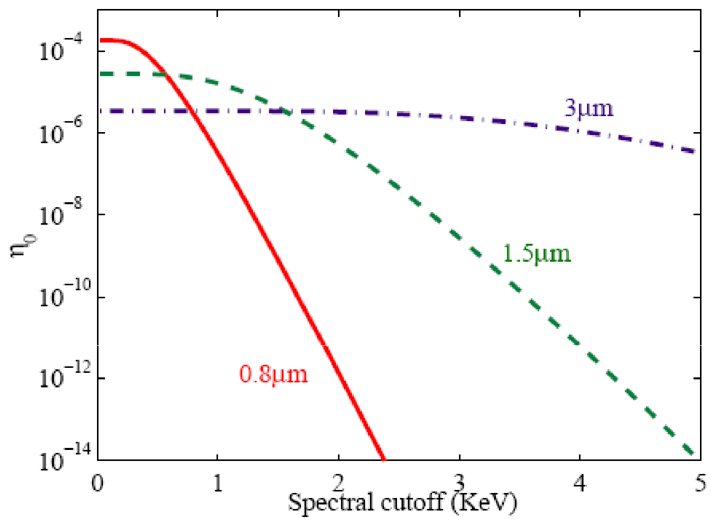
– cutoff increase expected

$h\nu_{max} \sim I_p + I_L\lambda^2$

Needs development of high

Energy parametric amplifier systems in the 2 μm

region

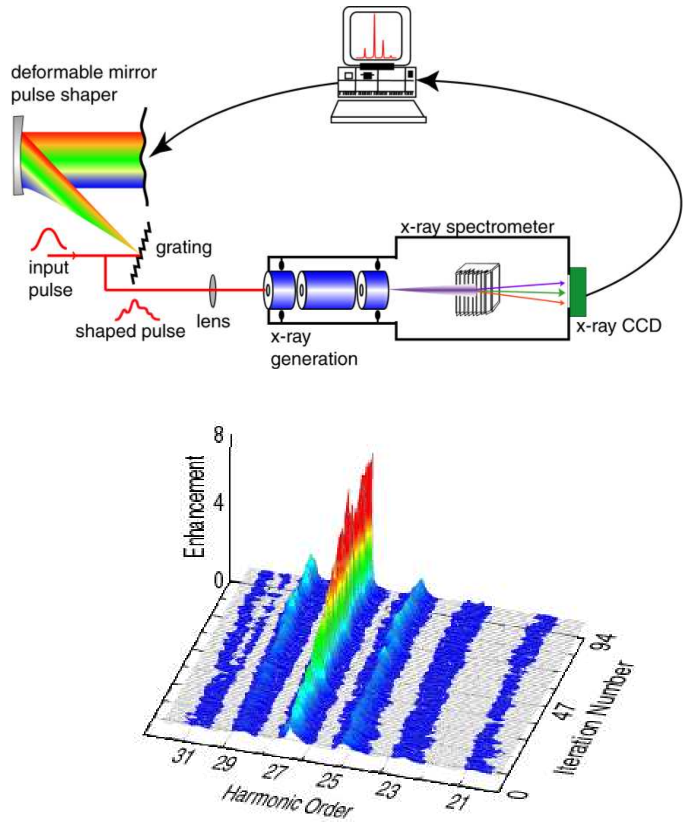


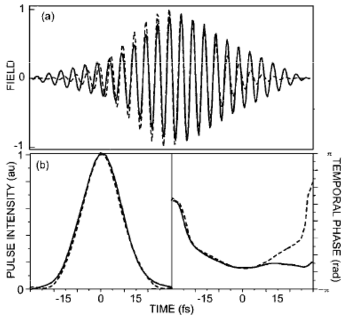
A. Gordon et al, Opt. Express **13**, 2941-2947 (2005)

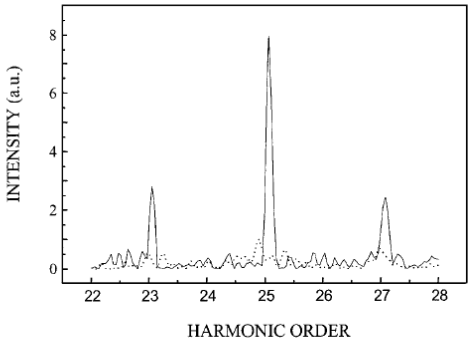
HHG in gases

Temporal pulse shaping

Optimization of harmonic yield for a given harmonic order : very small changes in the driving pulse temporal phase induce substantial enhancement







Theory : Christov et al, PRL 86 (2001), 5458

Bartels et al, Nature **406** (2000) 164

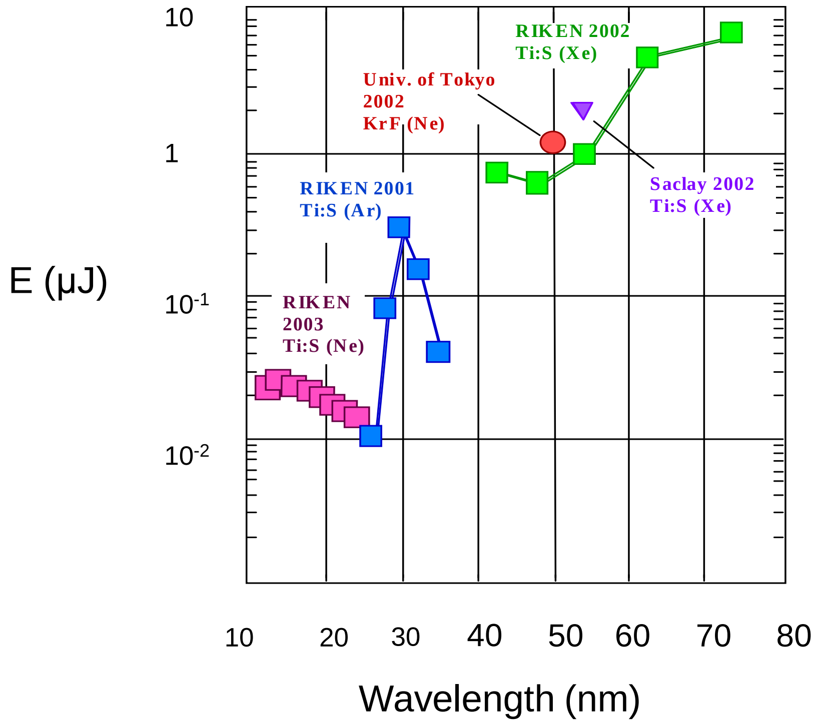
HHG in gases

PARAMETERS

Pulse energy

- Above 1μJ from 40 to 80 nm
-10-100 nJ in the 10-40 nm range

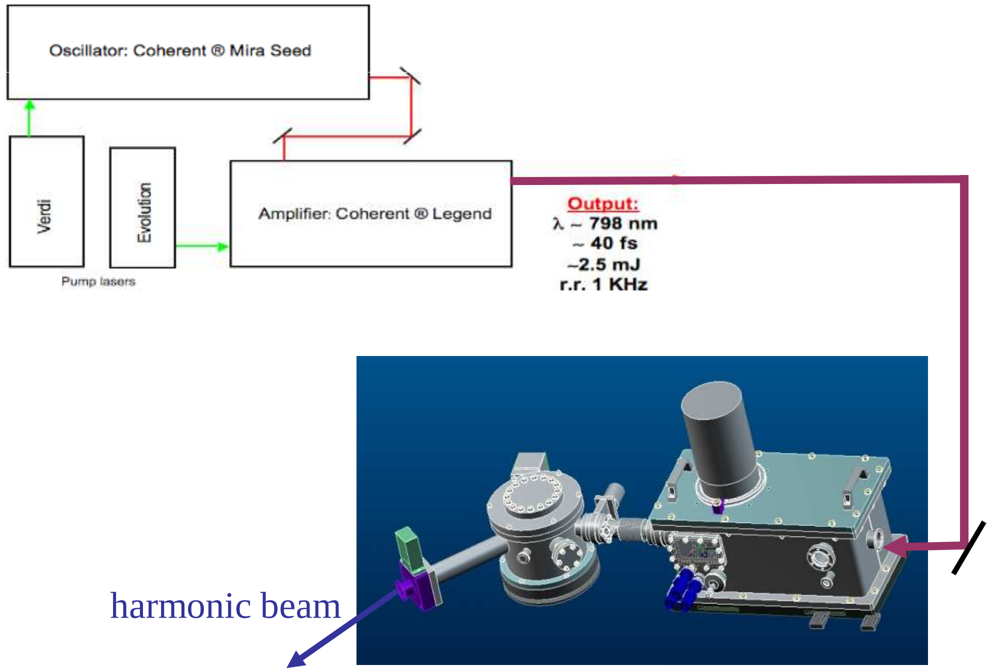
Laser source: Ti:saphire at 800 nm
For low rep rate high-energy regime:
E_p~30-50 mJ, t_p ~30-50 fs , rep rate ~ 10 Hz
Geometry: loose focusing (f_l ~2-5 m)
For low-medium energy HHG:
E_p~2-3 mJ, t_p<30 fs , ~1KHz , sharper focusing f_l<1m

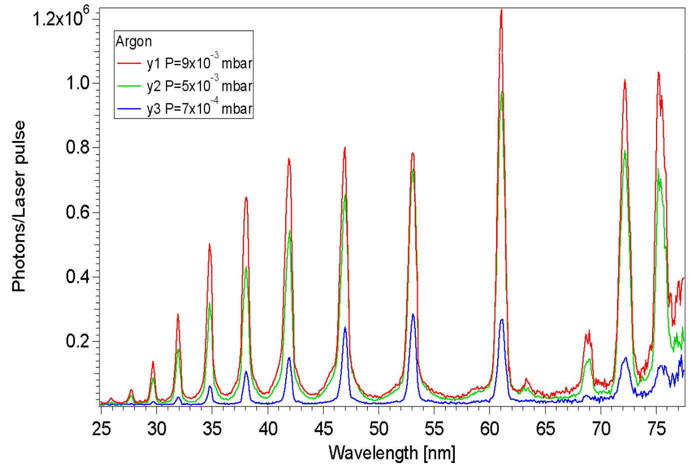


HHG setup at Elettra

LASER SOURCE PARAMETERS:
Ti:Sapphire laser system
• Coherent® Legend® amplifier
$\lambda_0 \approx 798$ nm
Pulse energy ≈ 2.1 mJ
$\tau \approx 50$ fs
Upgrade in progress: pulse compression in hollow fibre
Amplifier Repetition Rate:1 KHz
• Beam spot size after 250 mm focusing lens ≈ 40 μm

MONOCHROMATOR SPECIFICATIONS:
• Grating:
576 grooves/mm
1000 mm radius
blazed profile
gold-coated
• Source-grating distance: 450 mm
• Grating-slit distance: 350 mm
• Subtended angle: 135°
• Al filter thickness: 200 nm





Slide by B.Ressel ,
group of F.Parmigiani at Elettra

HHG in gases

PARAMETERS

Max photon energy

10^2-10^3 ph/s within 10% bw at **1.3 keV**

(J.Seres et al, Nature 433 (2005), 596);

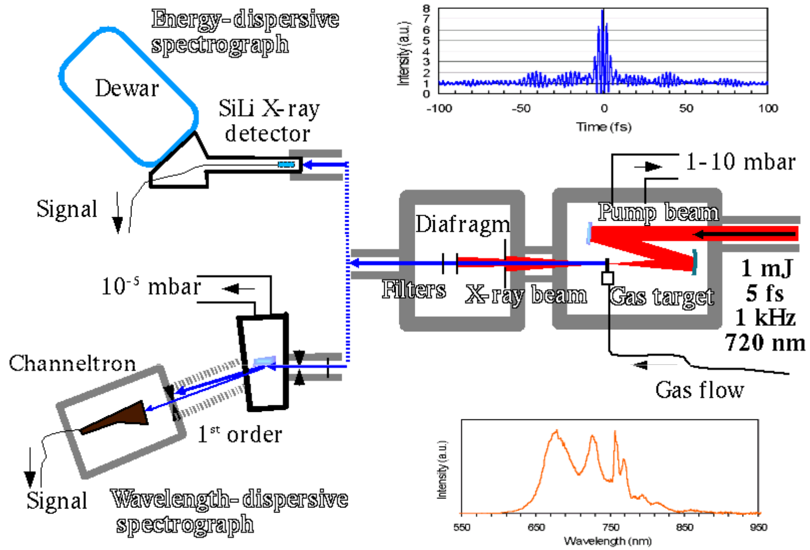
$2x10^5$ ph/s at **700 eV**

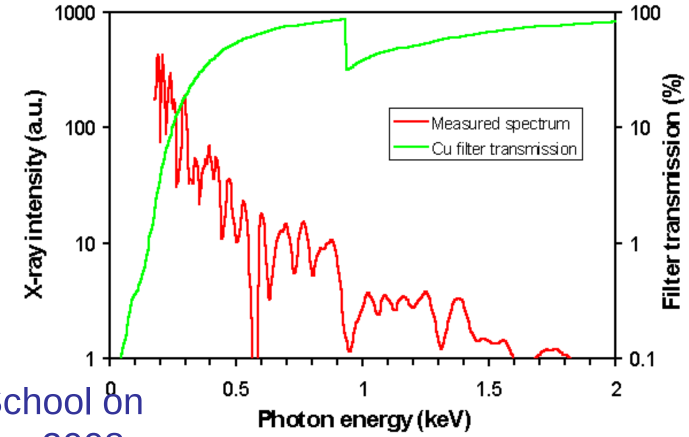
$2x10^7$ ph/s at **280 eV**

$1x10^8$ ph/s at **200 eV**

$5x10^8$ ph/s at **100 eV**

(E.Seres et al**,** *PRL* **92 (2004), 163002)**





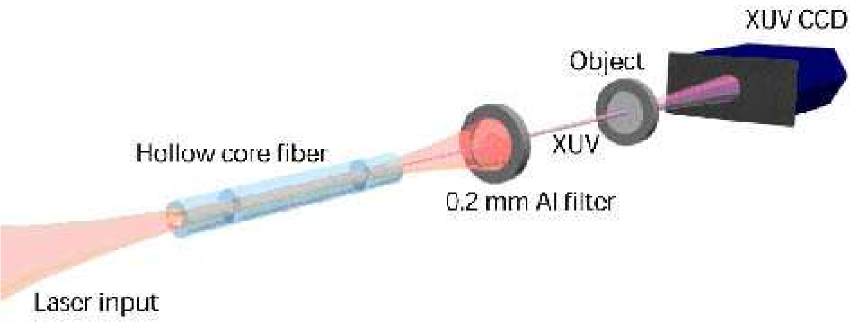
HHG in gases

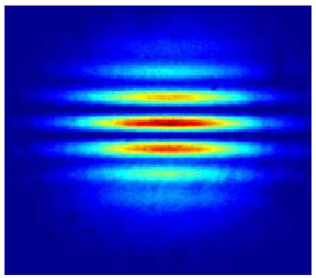
Parameters

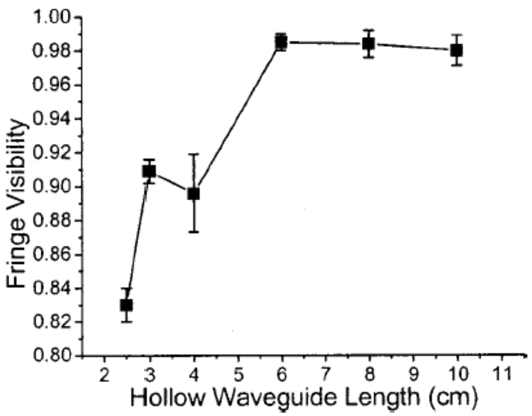
Spatial coherence – depends strongly on the generation scheme

Nearly 100% fringe visibility with 3 to 5 segment hollow-fibre filled with Ar, 40 Torr, harmonic order 23-39 (36-45 eV)

A.Libertun et al, Appl Phys Lett **84** (2004), 3903







Setup and Young fringes produced at 13 nm by the Kapteyn-Murnane group

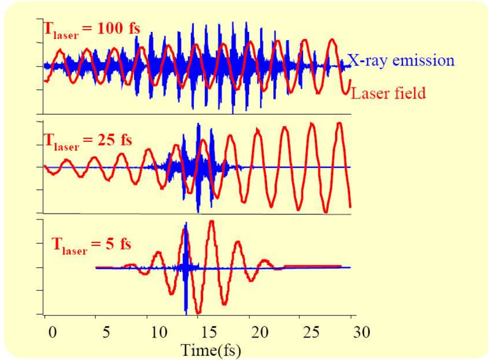
HHG in gases

Parameters

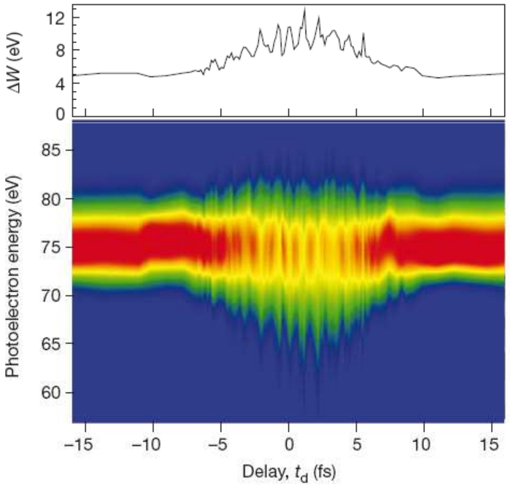
Temporal properties and attosecond pulse generation

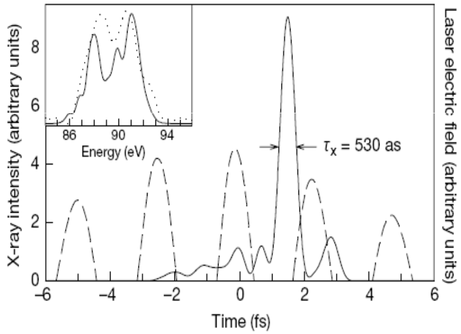
Two approaches:

1. Isolation of single as peak by using a few-cycle IR pulse and selecting the cutoff of HHG in a thin jet



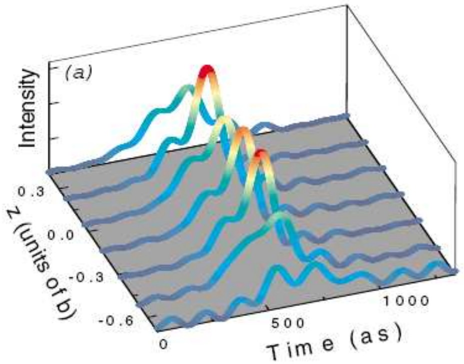
Christov et al, Phys.Rev.Lett **78 (1997)1251**





Hentschel et al, Nature **414 (2001) 504**

2. Generation of atosecond pulse train , if the harmonics in the plateau are phase locked . IR source: 45 fs, 40 mJ; gas jet: 1 mm Ne



M.B.Danailov, ICTP School on Synchrotron Radiation 2008

Mariesse et al, Phys Rev Lett **93** (2004), 163901