



**The Abdus Salam
International Centre for Theoretical Physics**



1936-16

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

7 - 25 April 2008

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

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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

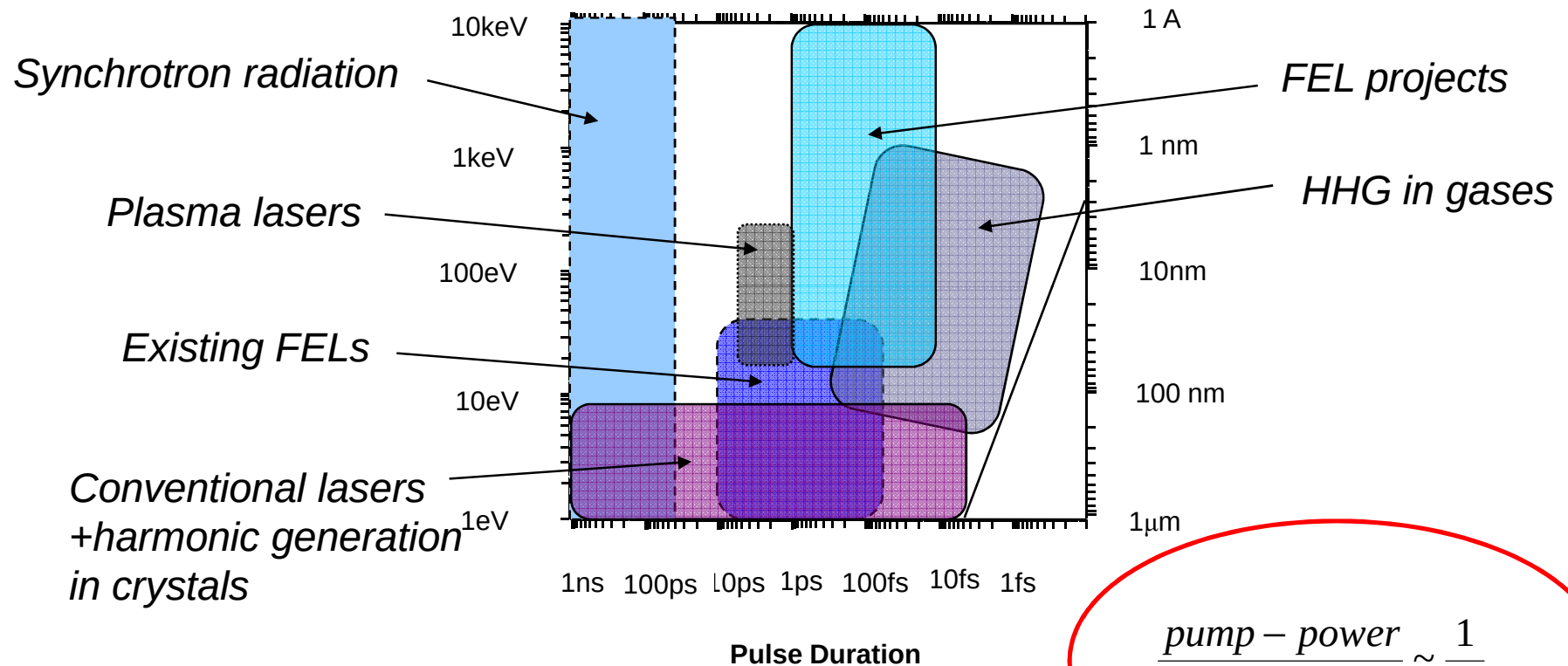
High order HG in Gases (HHG)

Trends in HHG

Features of the HHG light

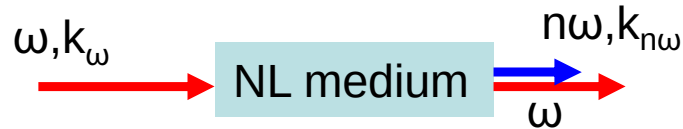
INTRODUCTION

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



$$\frac{\text{pump} - \text{power}}{m^3} \sim \frac{1}{\lambda^4}$$

Harmonic generation



Regimes:

1. Peak power density from 10^3 to 10^{13} W/cm² - **weak perturbation limit**

Nonlinear polarisation :

$$\vec{P} = \epsilon_0 (\chi^{(1)} \vec{E} + \chi^{(2)} \vec{E}^2 + \chi^{(3)} \vec{E}^3 + \dots)$$

Phase matching:

$$n k_\omega = k_{n\omega}$$

In NL crystals :

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

efficiency 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
130 nm , SHG (not in PM!) in SBBO

Low-order HG in gases

Features:

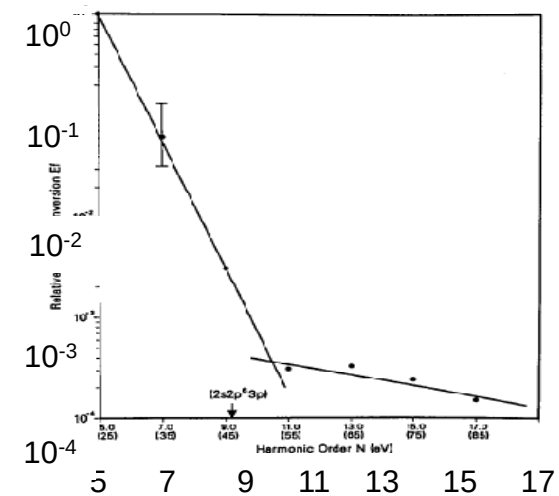
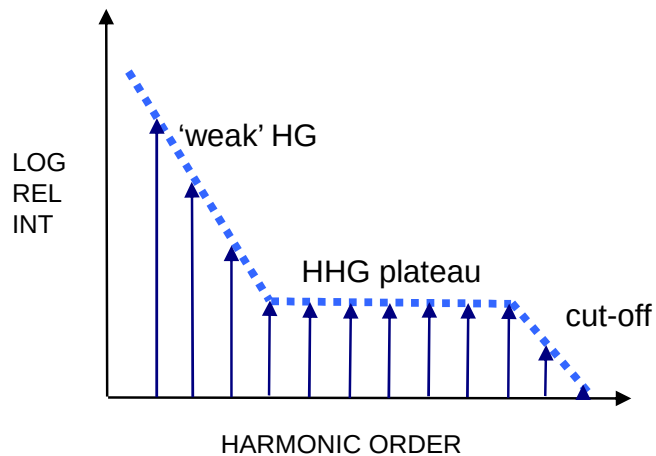
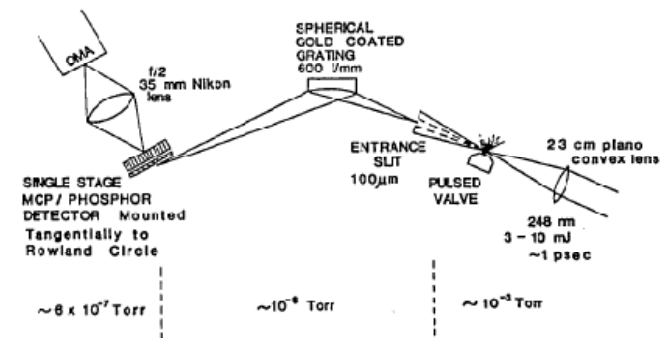
- Transmission in the VUV-EUV
- Odd harmonics only
- Low nonlinear susceptibility

$$\eta = \frac{P_3}{P_1} = \frac{3\pi^2}{\epsilon_0^2 c^2 \lambda_1^4} N^2 [\chi_{\lambda_3}^{(3)}]^2 P_1^2 |\Phi|^2 \quad (\text{in SI units})$$

- Regions where $\Delta k \sim 0$ (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/cm²)
the weak perturbation limit breaks -> plateau in
the harmonic conversion efficiency



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

HHG in gases

Directions for parameter improvement

- A. Phase-matching: standard scheme-> focusing in a gas-cell/jet and pressure optimization
 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
 - Selection of a bandwidth from a continuum

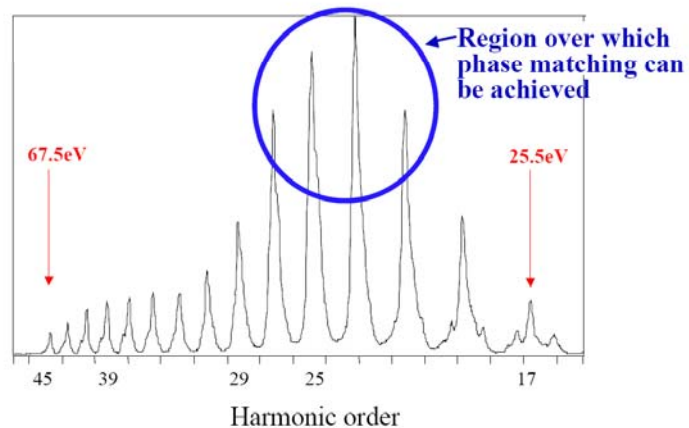
HHG in gases

A1. Phase matching in guided wave HHG

Phase mismatch

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

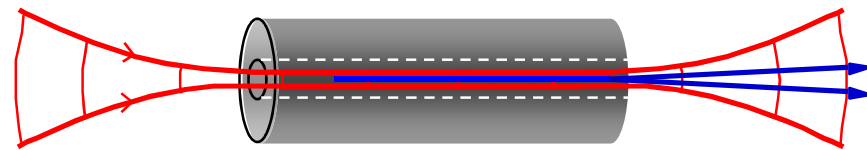
Fundamental



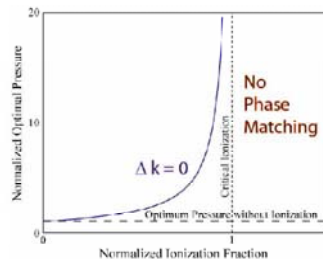
$$k = \frac{2\pi}{\lambda} \left(1 + N_a \delta(\lambda) - \frac{1}{2} \left[\frac{U_{nm} \lambda}{2\pi a} \right]^2 - \frac{1}{2} \frac{N_e r_e \lambda^2}{\pi} \right)$$

vacuum
gas
waveguide
ionization

Hollow fibre



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

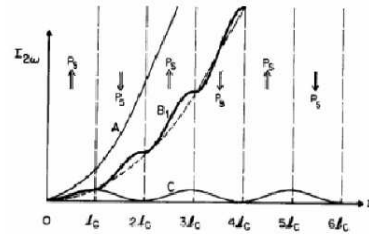
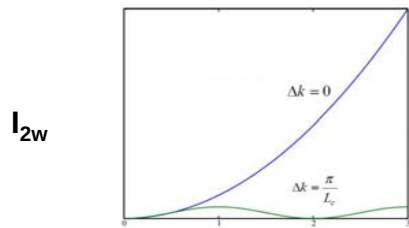


Critical ionization: 0.5%, 1%, 5%
For He, Ne and Ar

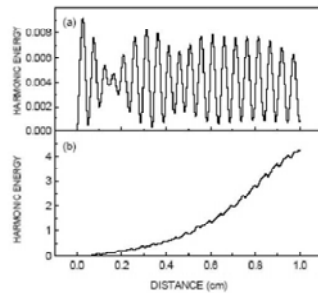
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Synchrotron Radiation 2008

HHG in gases

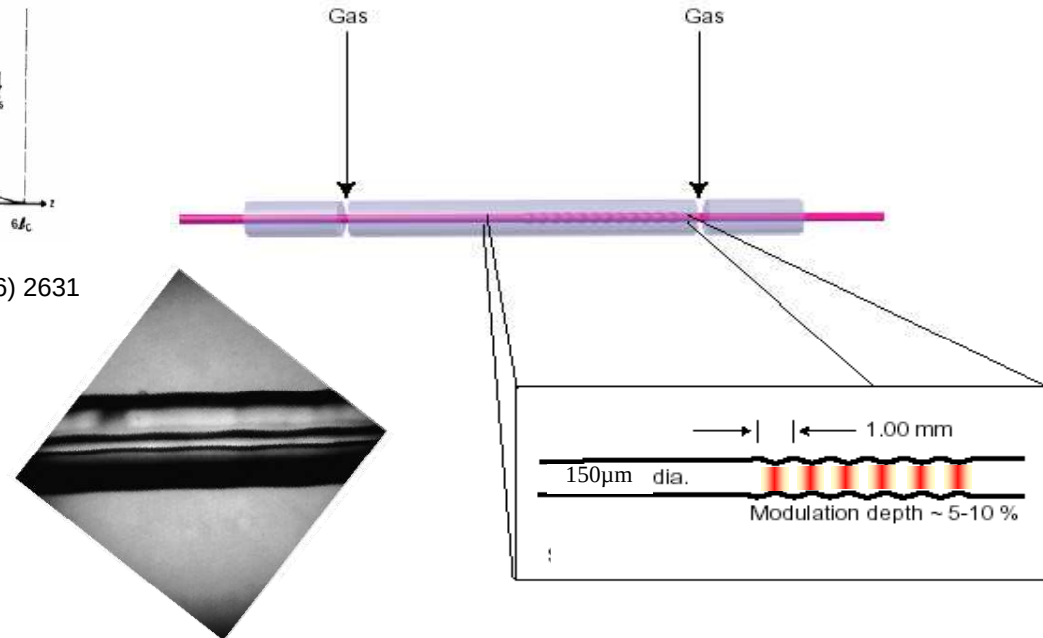
A.3. Quasi-Phase Matching (QPM) in waveguide HHG



Fejer et al, JQE **28** (1996) 2631



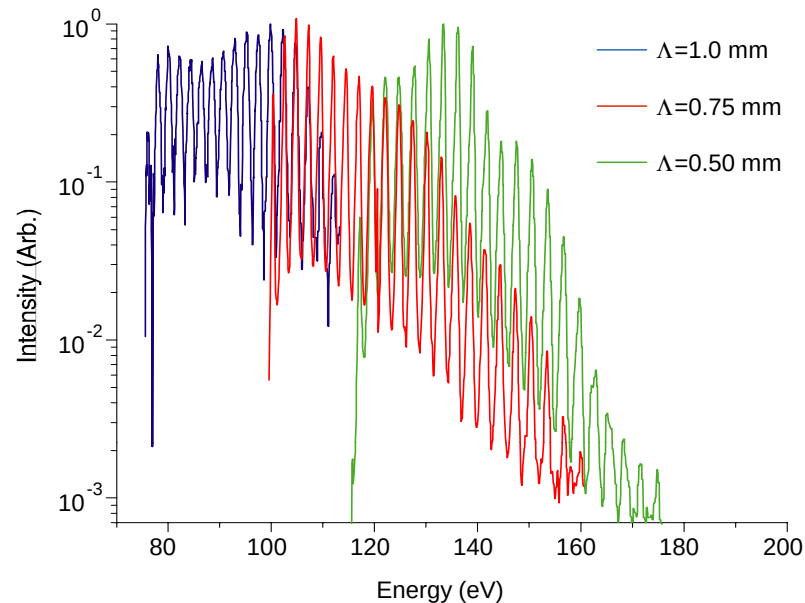
I.P.Christov et al, *OptExpress* **7**(2000), 362



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

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HHG in gases

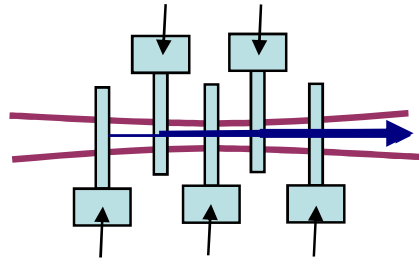


Cut-off shift to higher energy by QPM in He, driving pulse 25 ps,
Peak intensity $\sim 5 \times 10^{14}$ W/cm²,

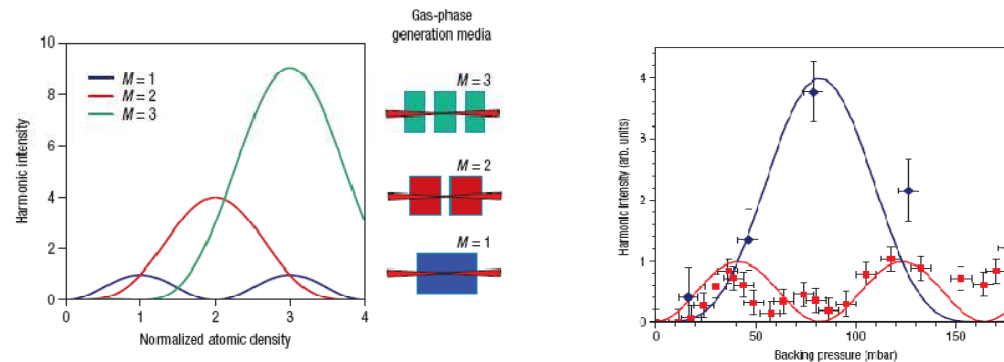
A. Paul et al, *Nature* **421** (2003) 51

HHG in gases

A.3. QPM using multiple gas jets

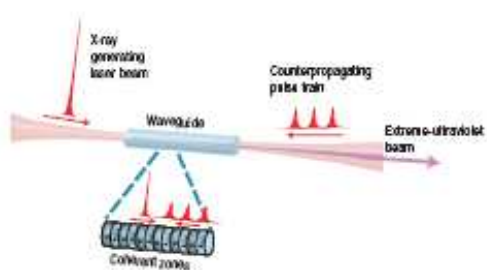


J.Seres et al, *Nature Phys* **3** (2007), 878

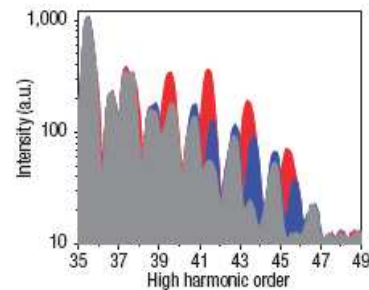


Theoretical prediction for QPM with 1-3 jets (left) and experimental result (right) for a single and two jets at optimised distance

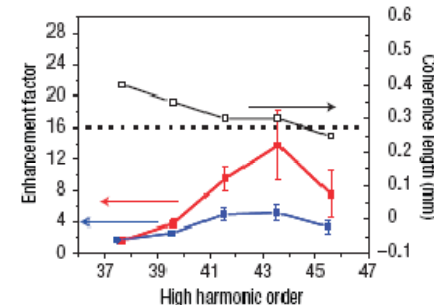
A.4. QPM using counter-propagating pulse/beam



X.Zhang et al, *Nature Phys* **3** (2007), 274



HHG with no (grey), one (blue) and two (red) c-prop pulses

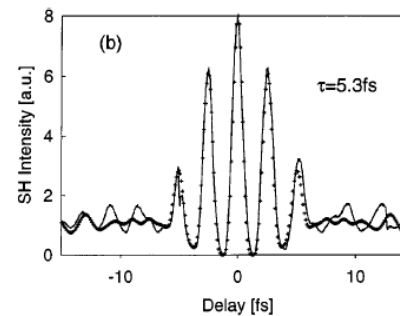
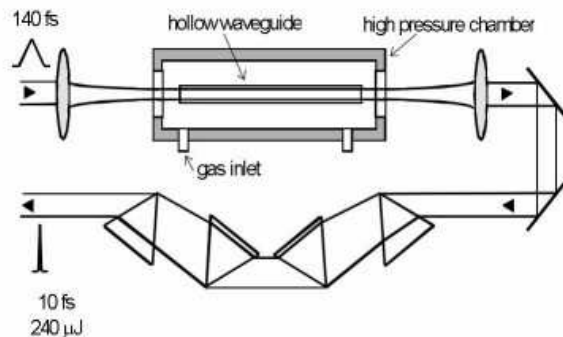


HHG enhancement with one (blue) and two (red) c-prop pulses

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HHG in gases

A.4. Use of very short pulses: non-adiabatic regime, non-adiabatic self-phase matching (NSPM)



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

Approach for generation of few-cycle fundamental pulse (~ 5 fs at 800 nm) proposed at Politecnico di Milano and Viena

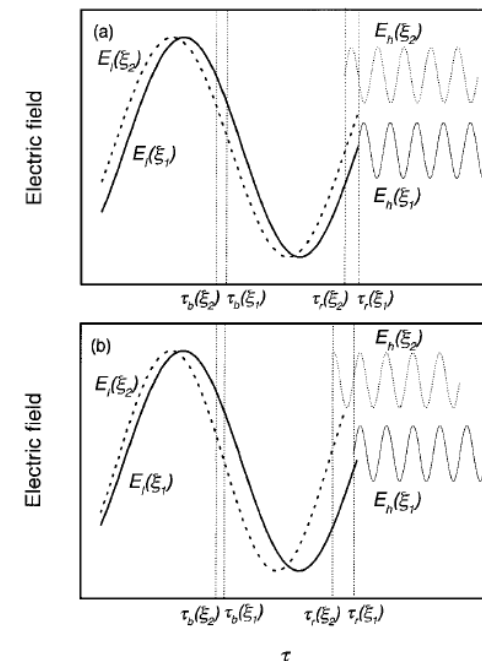
NSPM: very short pulses focused to $0.2\text{-}1 \times 10^{16} \text{ W/cm}^2$;

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 μ J , focused to 30-40 μ m

Medium: thin jet (0.5 mm), 0.5 bar



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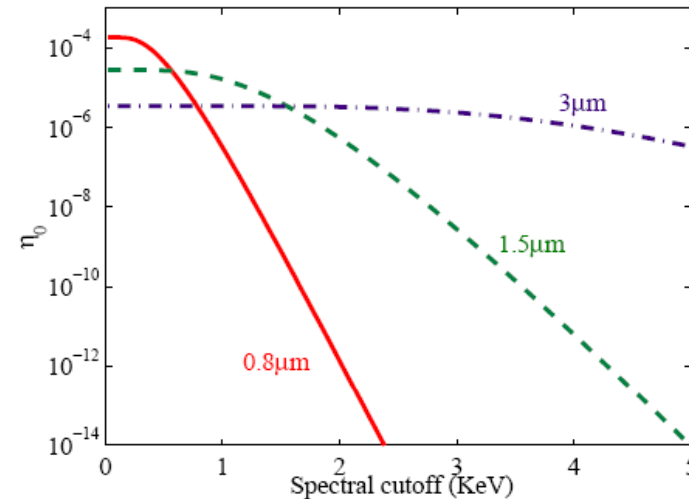
HHG in gases

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
 μm region

Problem: loss of efficiency !

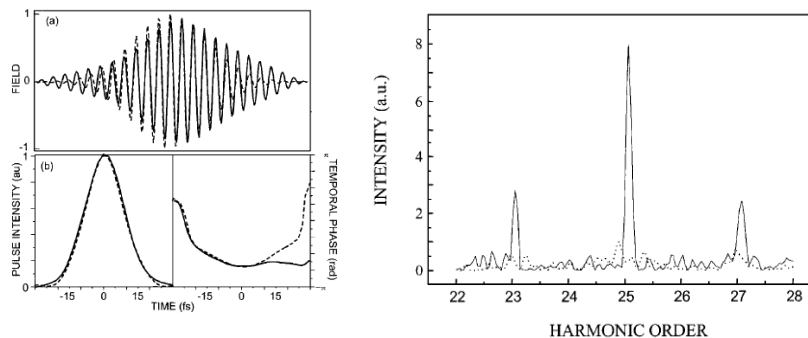


A. Gordon et al, Opt. Express **13**, 2941-2947 (2005)

HHG in gases

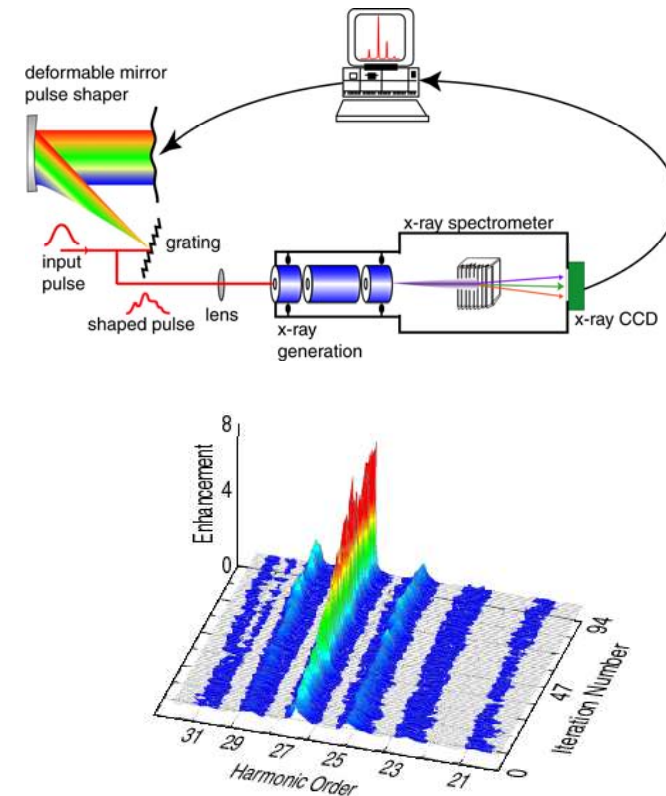
Temporal pulse shaping

a. 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

b. Quantum path control using CEO phase control *Phys.Rev.A* **73** (2006), 053408



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

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HHG in gases

PARAMETERS

Pulse energy

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

Laser source: Ti:sapphire at 800 nm

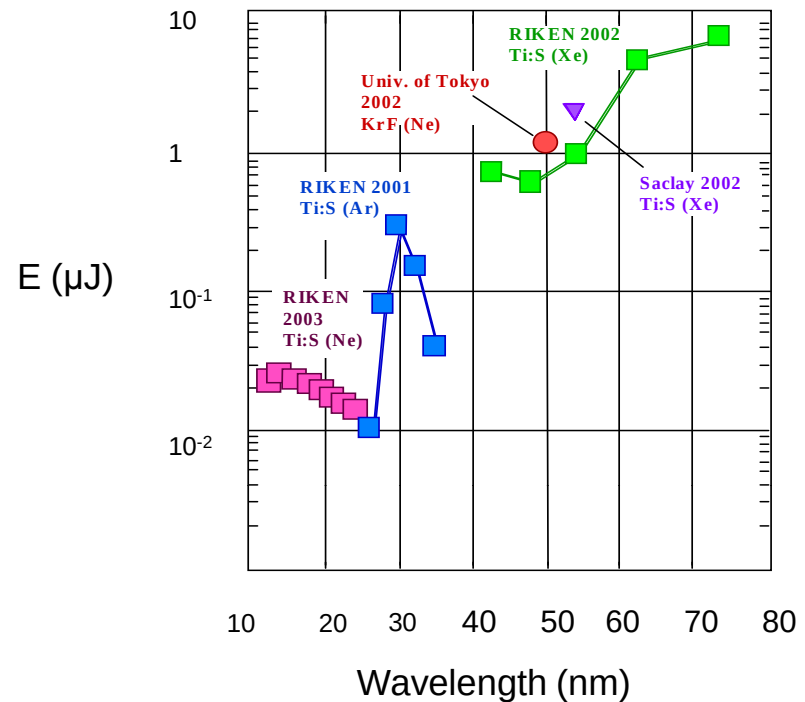
For low rep rate high-energy regime:

$E_p \sim 30\text{-}50\text{ mJ}$, $t_p \sim 30\text{-}50\text{ fs}$, rep rate $\sim 10\text{ Hz}$

Geometry: loose focusing ($f_l \sim 2\text{-}5\text{ m}$)

For low-medium energy HHG:

$E_p \sim 2\text{-}3\text{ mJ}$, $t_p < 30\text{ fs}$, $\sim 1\text{ KHz}$, sharper focusing $f_l < 1\text{ m}$



HHG setup at Elettra

LASER SOURCE PARAMETERS:

Ti:Sapphire laser system

- Coherent® Legend® amplifier

$\lambda_0 \approx 798 \text{ nm}$

Pulse energy $\approx 2.1 \text{ mJ}$

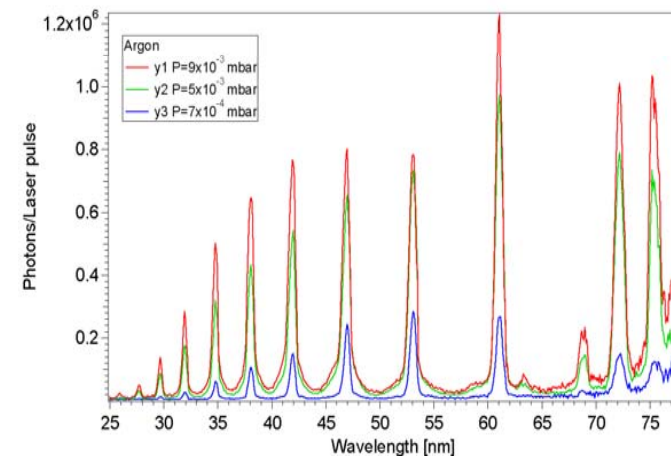
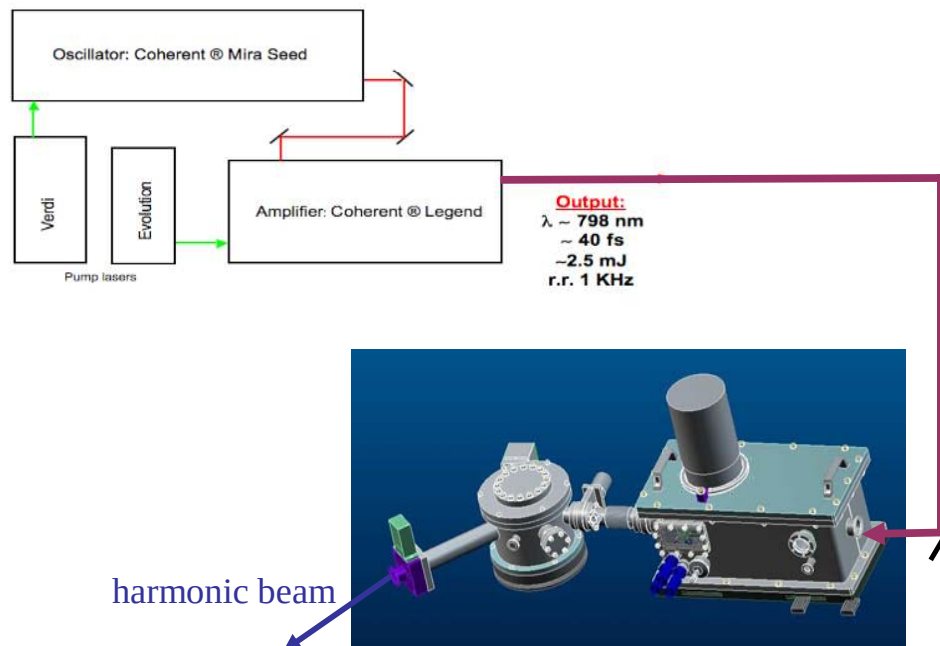
$\tau \approx 50 \text{ fs}$

Upgrade in progress: pulse compression in hollow fibre

Amplifier Repetition Rate: 1 KHz

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*

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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);

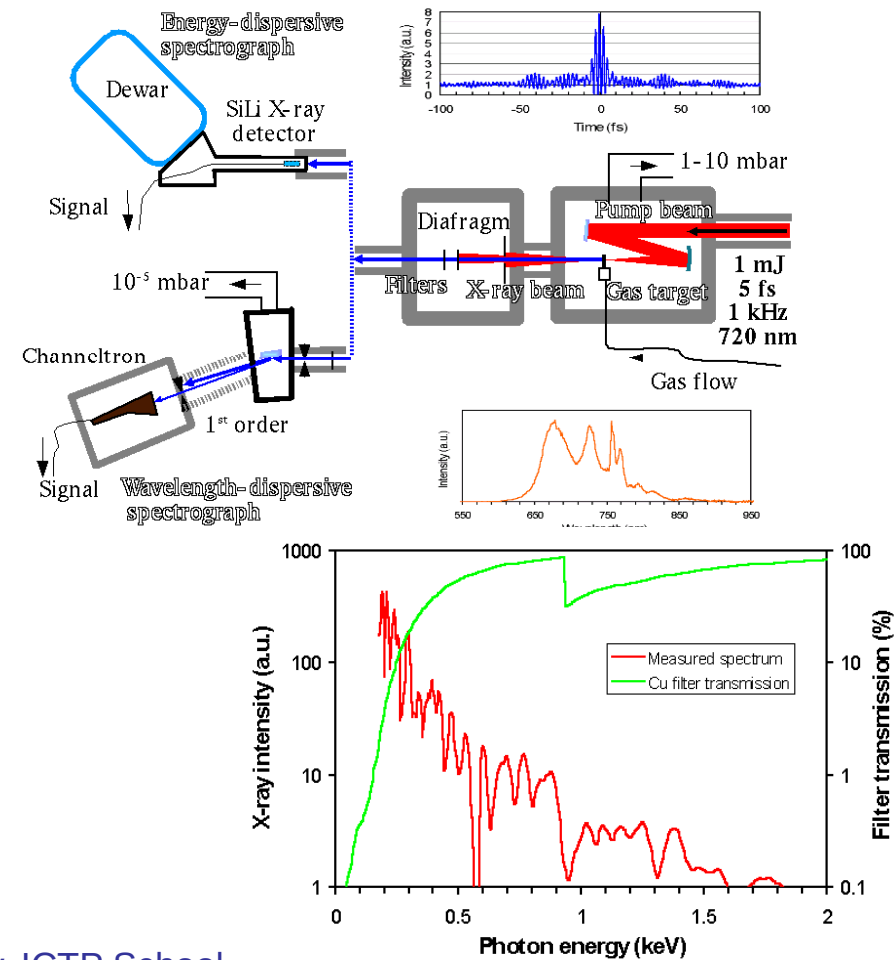
2×10^5 ph/s at **700 eV**

2×10^7 ph/s at **280 eV**

1×10^8 ph/s at **200 eV**

5×10^8 ph/s at **100 eV**

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



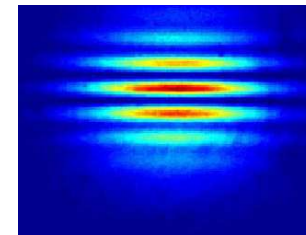
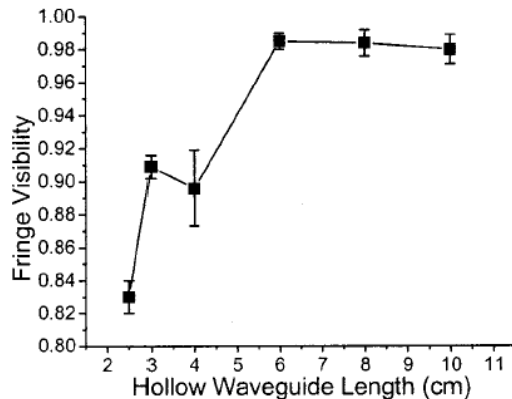
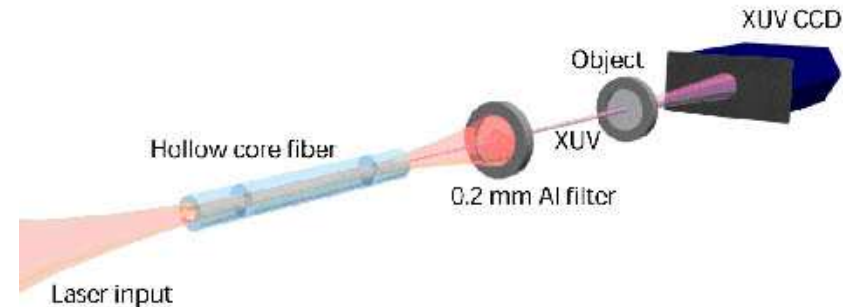
HHG in gases

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 (drawing from H.Kapteyn's home page)

HHG in gases

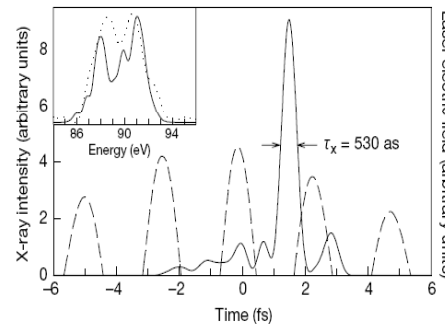
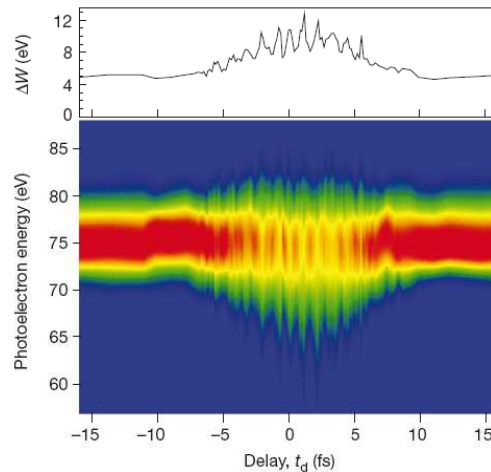
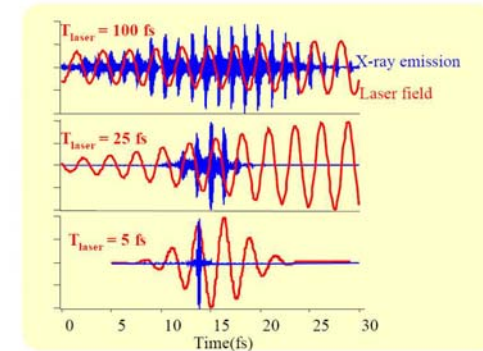
Parameters

Temporal properties: burst of sub-fs spikes separated by half cycle of the fundamental; can be isolated by a monochromator

Attosecond pulse generation

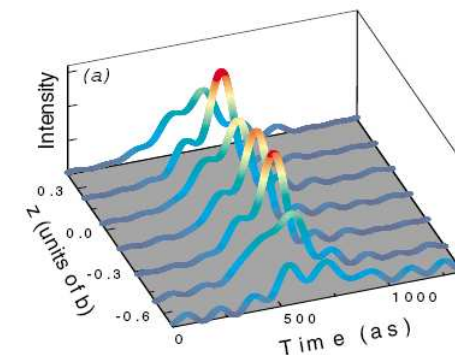
Two approaches:

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



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

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



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

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