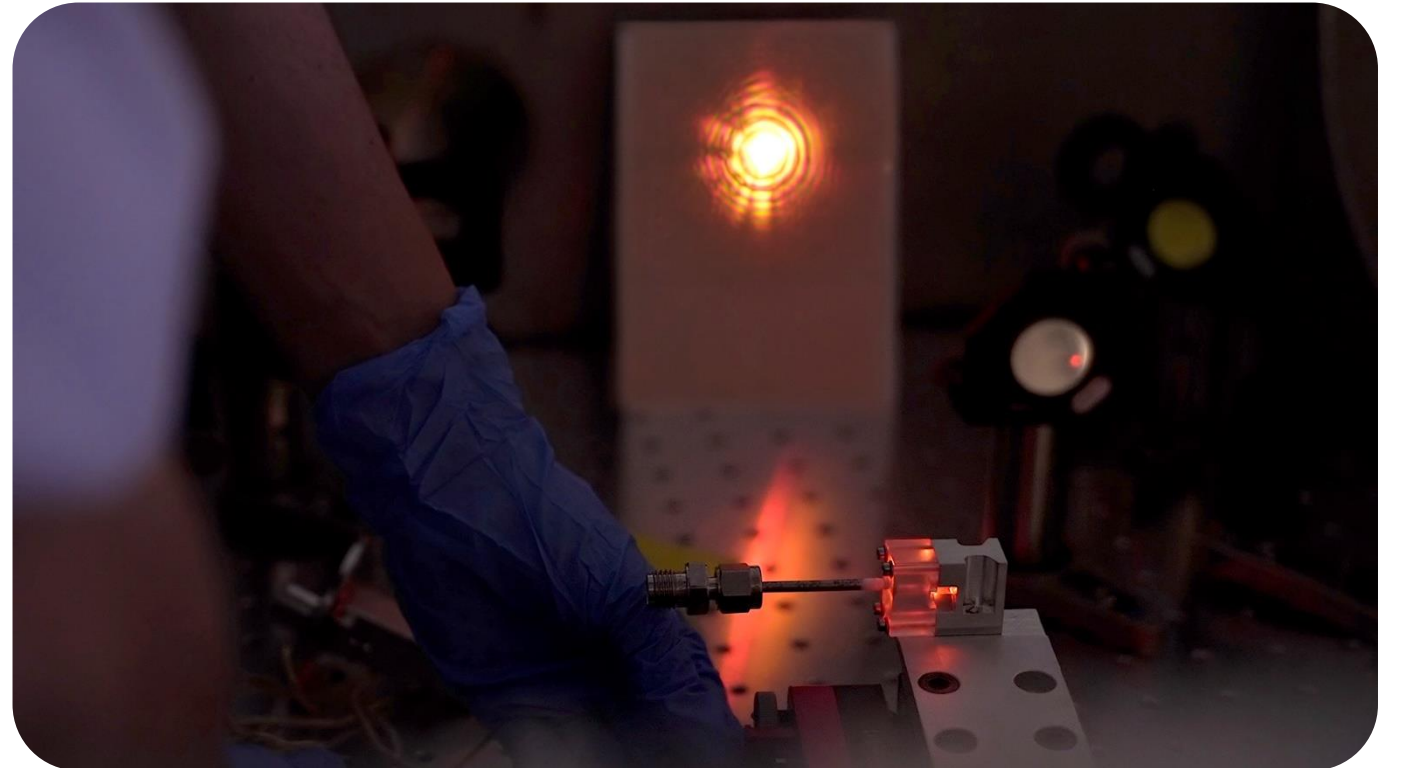


Development of table-top XUV and soft-X sources based on microfluidic devices

Anna Gabriella Ciriolo



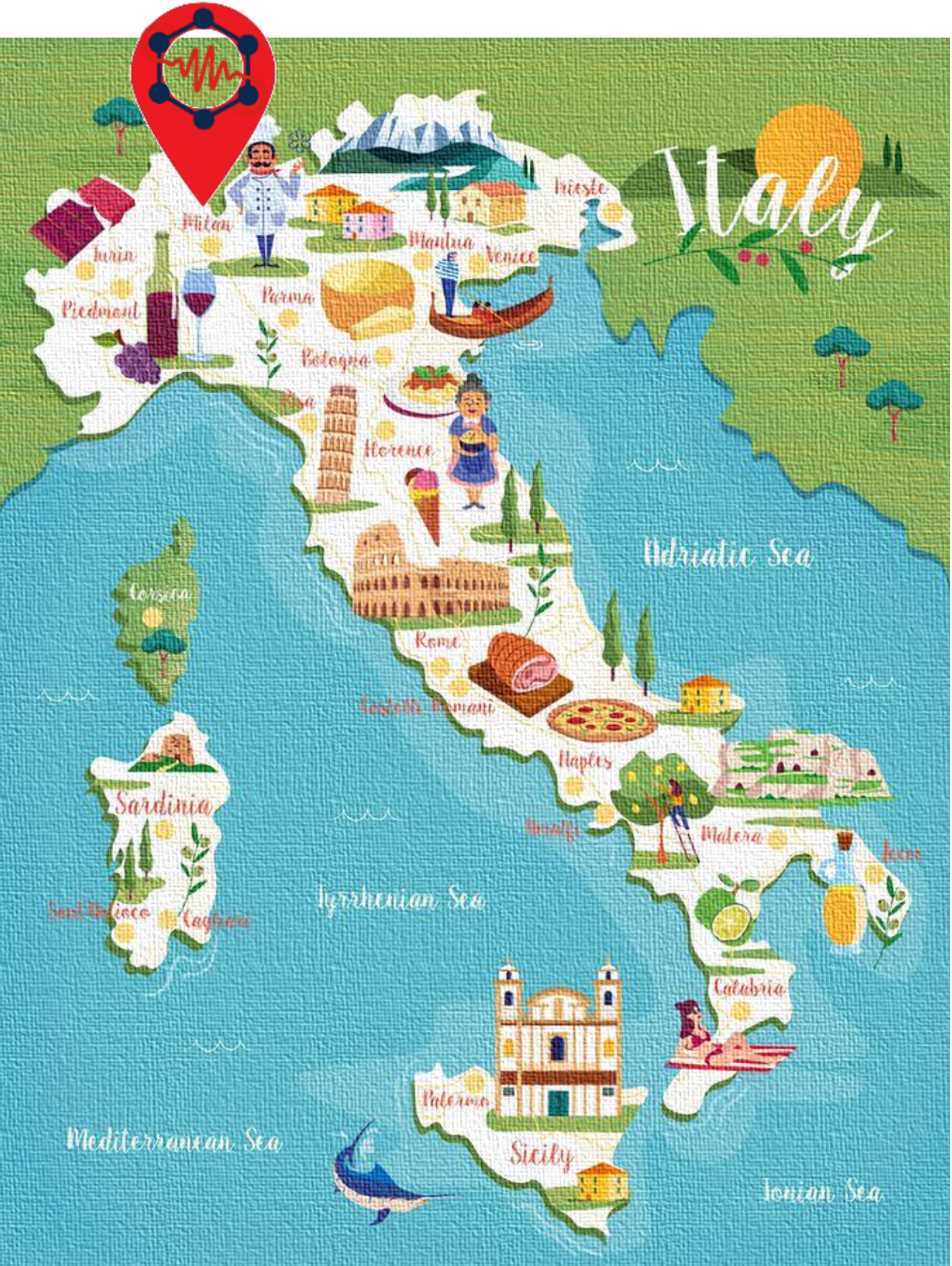


The **U**ltrafast **DYN**amics **I**n matter team

ultrafast spectroscopy from THz to X-rays

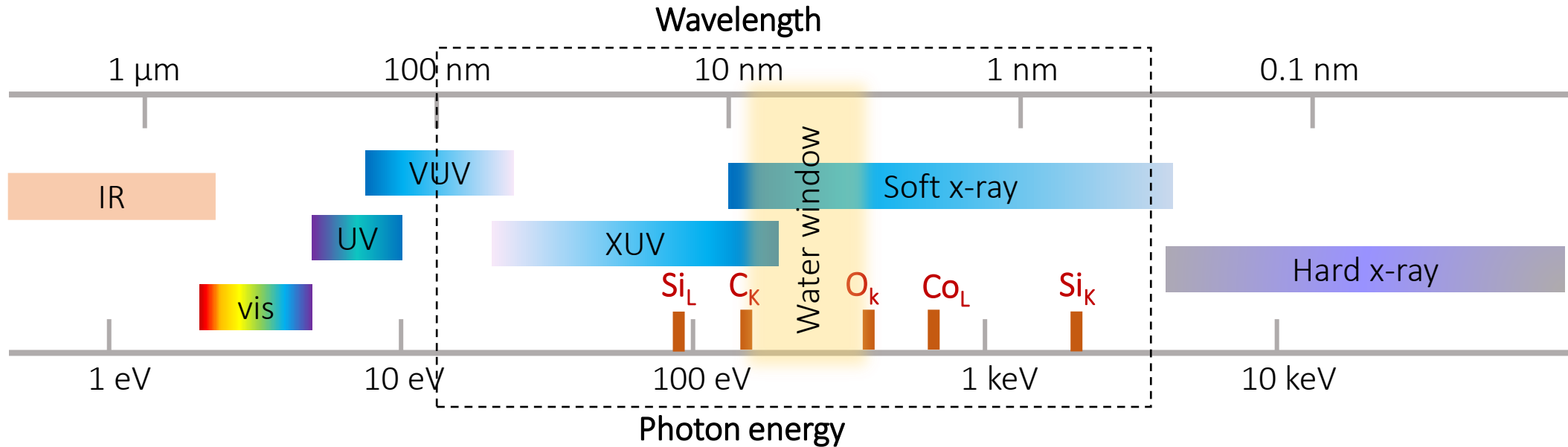


UDynI team IN A NUTSHELL



Milan,
Politechnic University

eXtreme UltraViolet (XUV) and Soft X-ray sources



K edges

B @ 188 eV
C @ 284 eV
O @ 543 eV

L edges

Al @ 72 eV
Si @ 99 eV
P @ 135 eV
S @ 163 eV

M edges

Ga @ 19 eV
Ge @ 30 eV
As @ 42 eV
Ti @ 33 eV
Br @ 50 eV
Fe @ 52 eV
Co @ 59 eV
Ni @ 67 eV

N edges

I @ 70 eV
Pb @ 150 eV

Edges identify the Binding Energy (BE) of a core shell

K edge → BE of n=1 shell

L edge → BE of n=2 shell

M edge → BE of n=3 shell

N edge → BE of n=4 shell

Semiconductors

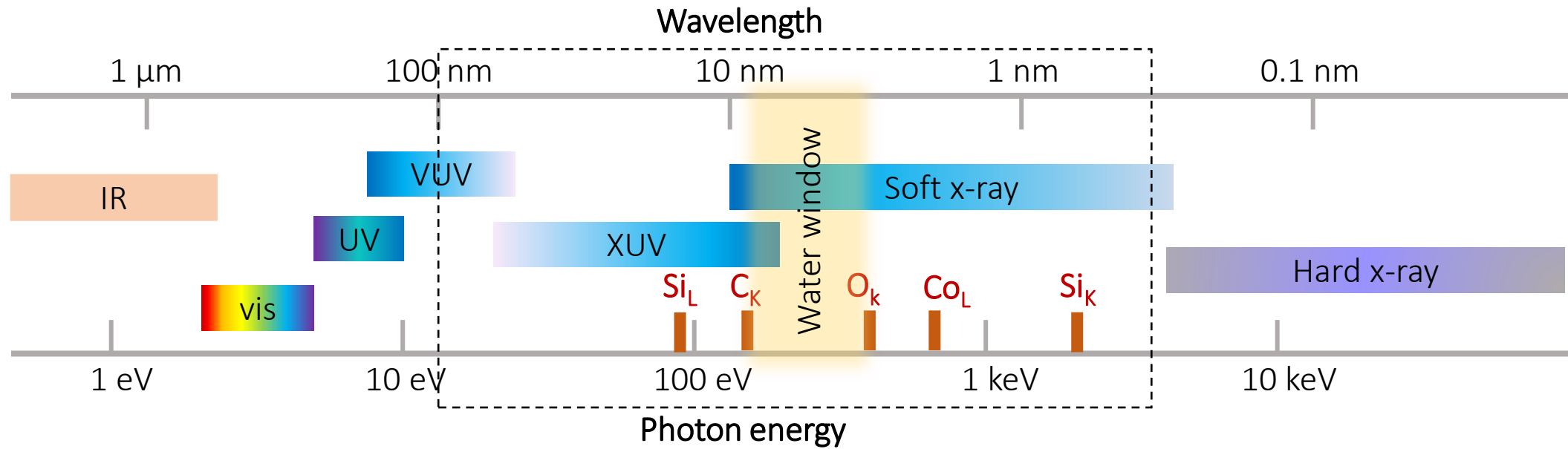
Metals

Halogen

Bio-relevant

The energies of absorption edges in X-ray absorption spectra reveal the identity of the corresponding absorbing elements.

eXtreme UltraViolet (XUV) and Soft X-ray sources



Free electron lasers

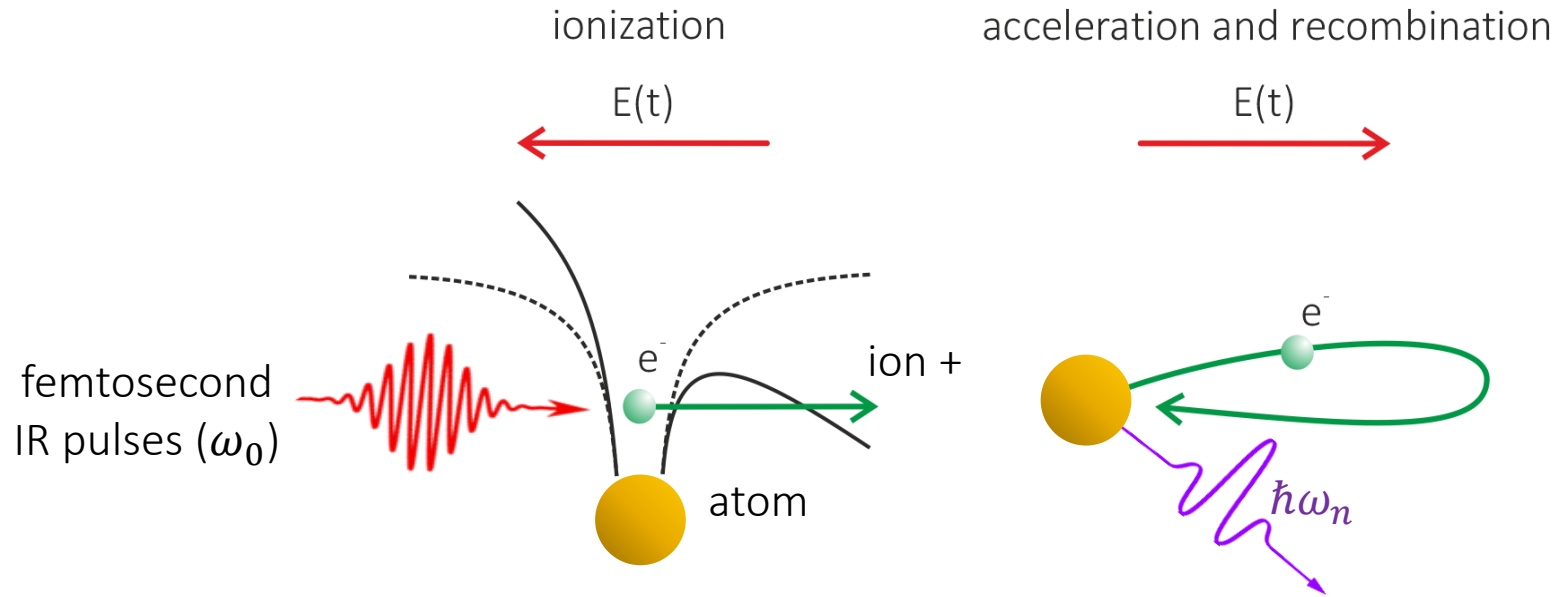
- high intensity (10^{20} W/cm²)
- spectral purity
- polarization control



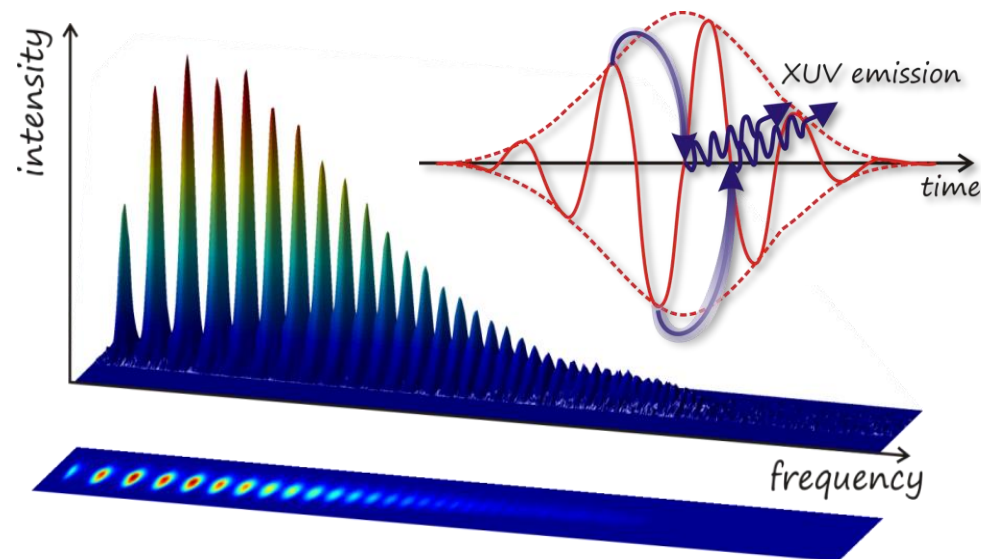
Table-top sources (high-order harmonic generation)

- Price & Accessibility
- Temporal resolution (50 as)
- Flexibility

Table-top sources: High-order Harmonic Generation (HHG)



- I. **Ionization** by tunnel ionization
- II. **Propagation**: the free electron is accelerated by the laser field
- III. **Recombination**: emission of a high-energy photon



Broadband spectrum, $\hbar\omega_{max} = I_p + 3.17 U_p$ ($U_p \sim E^2 \lambda_0^2$)

Generation of **attosecond pulses** (as, $1 \text{ as} = 10^{-18} \text{ s}$)

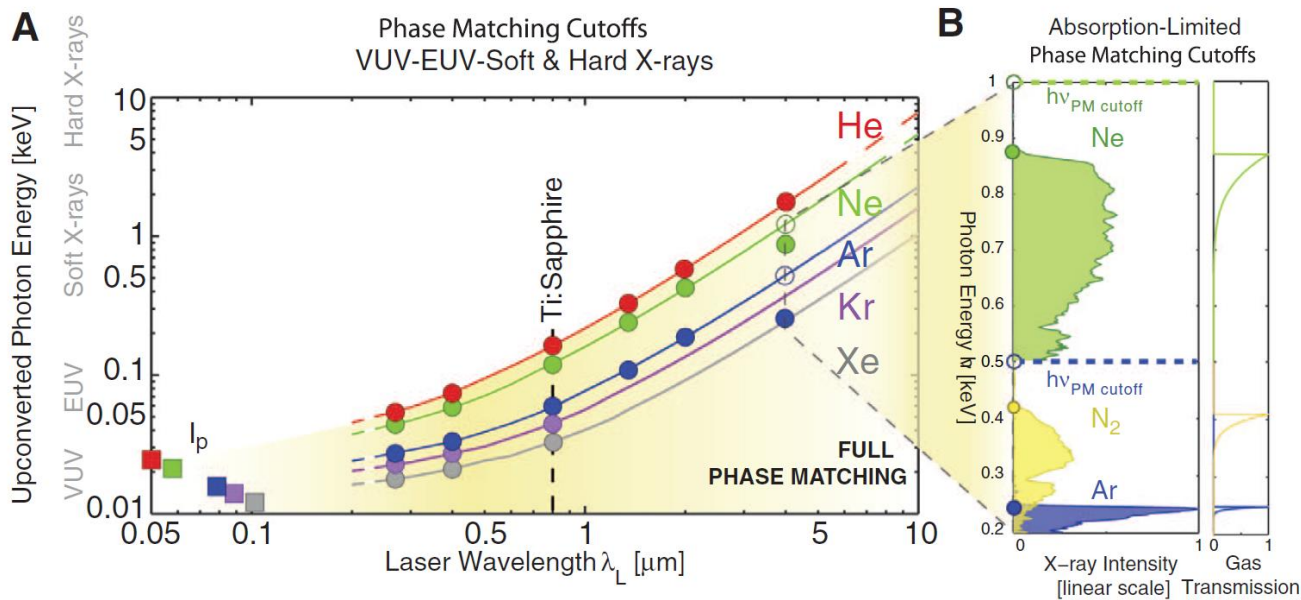
low conversion efficiency ($\approx 10^{-6} \div 10^{-9}$ with 800-nm driver)

How to increase the HHG cutoff

- Increase I_p
- Increase the driving intensity
- Increase the driving wavelength

$$\hbar\omega_{max} = I_p + 3.17 U_p$$

$$U_p \propto E^2 \lambda_0^2$$

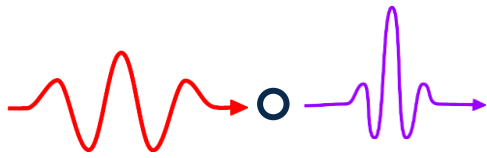


T. Popmintchev et al. *Science* 336.6086 (2012): 1287-1291.

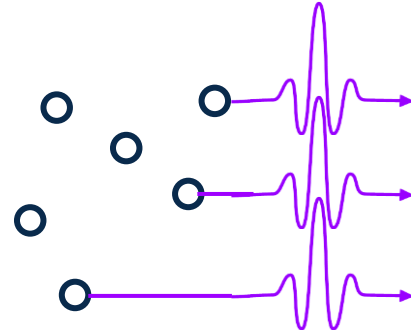
Physical constraints

- Conversion efficiency (CE) scales as $\sim\lambda^{-6}$
(CE @ 1.6 μm is 2^{-6} lower than @ 800nm)
- Collective effects: phase matching and absorption

Phase matching in HHG



single atom



In HHG $\varphi_{driving\ pol} - \varphi_{harmonic} = 0$

- gas medium
- driving laser wavelength/intensity
- geometry of the interaction

q-th harmonic order $k_{driving} - q \cdot k_q = \Delta k_q(x) = \Delta k_q^{disp} + \Delta k_q^{dipole} + \Delta k_q^{geom}$

$\Delta k_{Dispersion}$ —due to the optical dispersion of the generating medium (neutral atoms and free electrons),

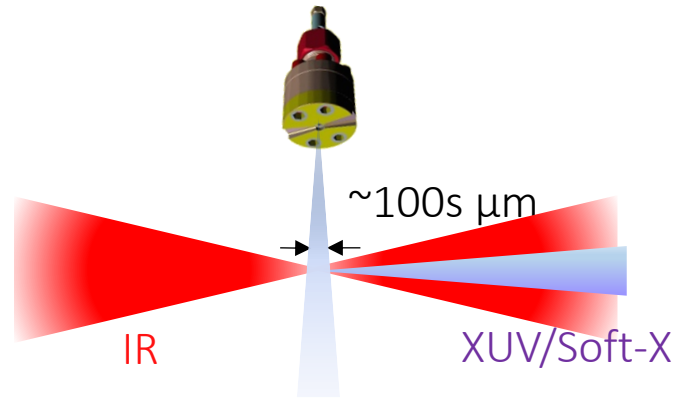
Δk_{Dipole} —due to the intensity dependent dipole phase,

Δk_{geom} —the geometrical wave vector mismatch caused by focusing/waveguide

$$\frac{dE_q}{dx} = \underbrace{b(x)}_{\text{source}} e^{i \int_{-x_0}^x \underbrace{\Delta k_q(x') dx'}_{\text{phase-mismatch}}} - \underbrace{\alpha_q(x)}_{\text{absorption}} E_q$$

Generation strategies

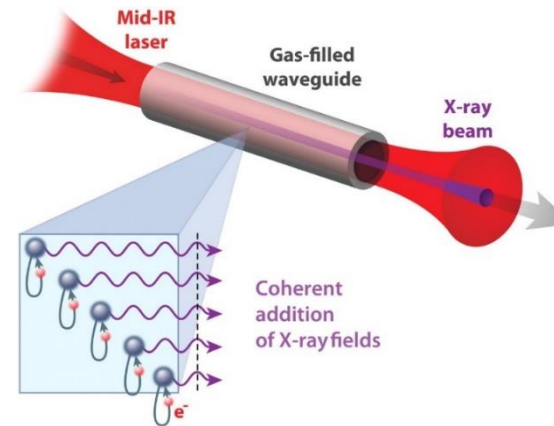
Free gas expansion



- X** Short interaction volume (~100s μm)
- ✓** Low absorption

P. Salieres et al., Phys. Rev. Lett. **74**, 3776 (1995)

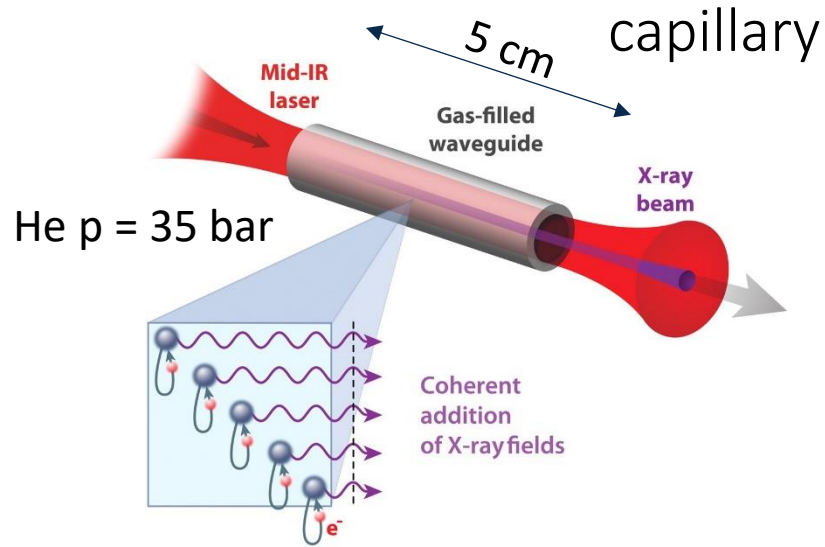
Confined gas media (capillary, gas cell)



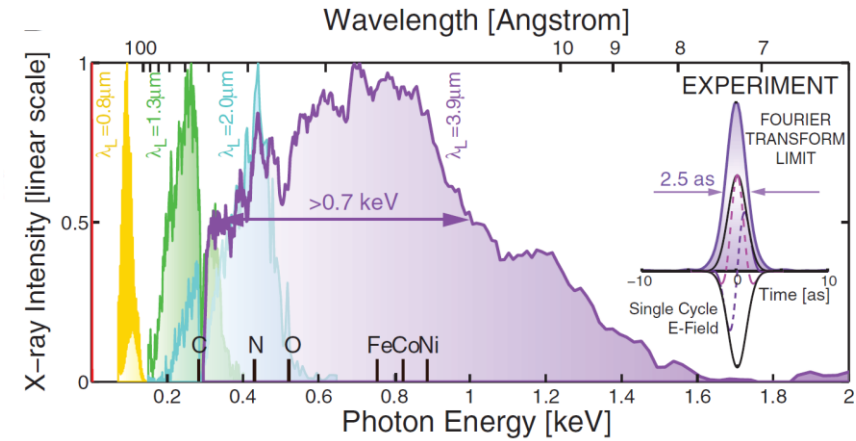
- ✓** Long interaction volume (~10s cm)
- ✓** High-density gas media (up to 100s bar)
- X** Absorption

C. G. Durfee III et al., PRL **83**, 2187 (1999)
T. Popmintchev et al., PNAS **106**, 10516 (2009)

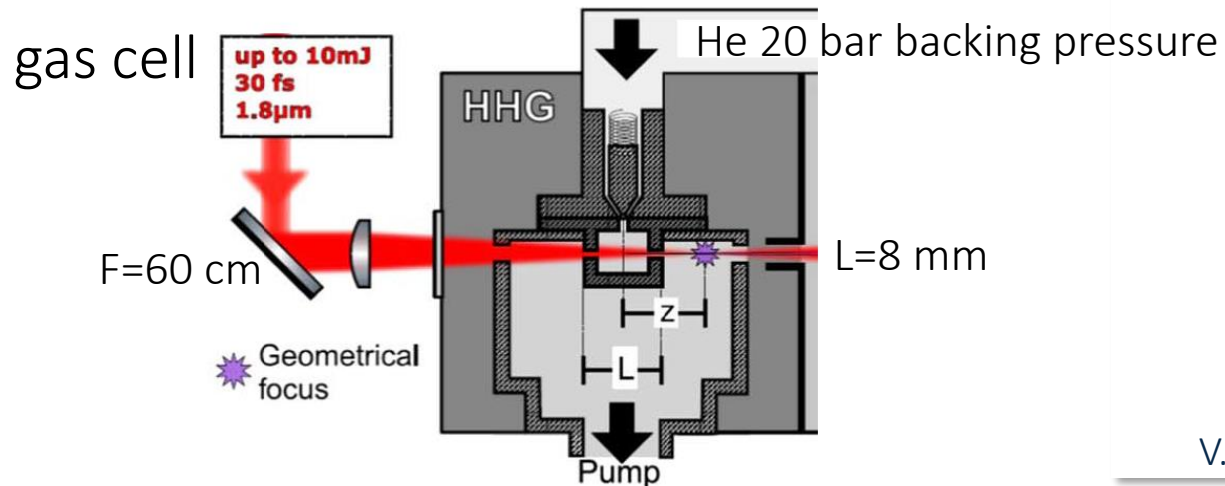
State of the art in table-top HHG



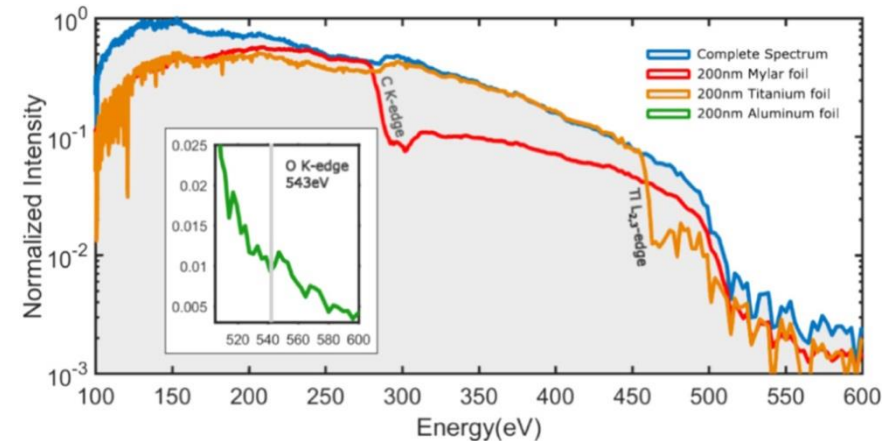
HHG up to 1 keV driven by mid-IR pulses at 3.9 μm



T. Popmintchev et al. *Science* 336.6086 (2012): 1287-1291.

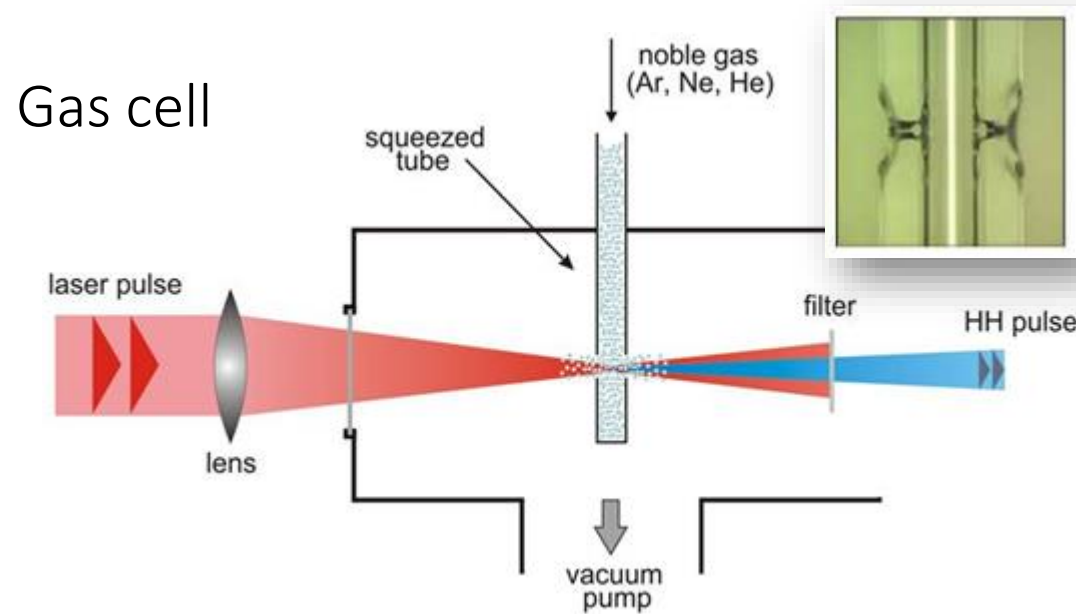


Water-window HHG sources driven by near-IR pulses

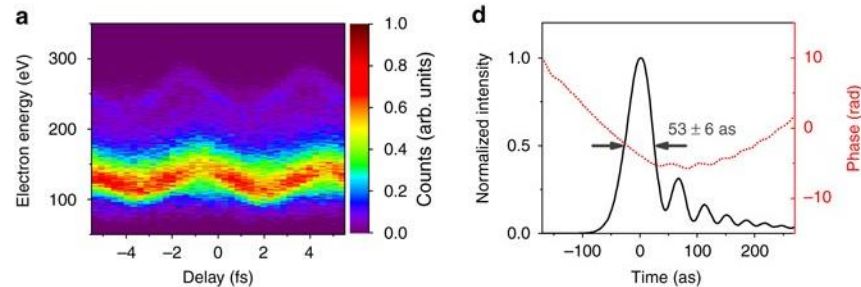


V. Cardin et al., *J. Phys. B: At. Mol. Opt. Phys.* 51 (2018) 174004

State of the art in table-top HHG and attosecond sources

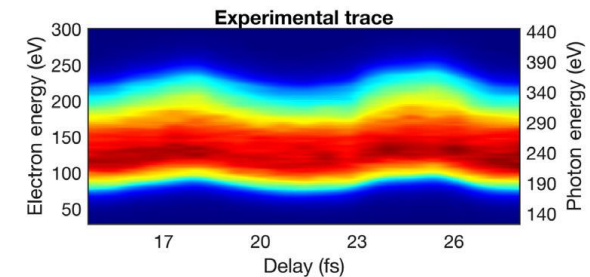


53 as X-ray pulses up to the C_k by 1.8 μm driver
1.5 mm-long cell, 1 bar neon gas



J. Li et al. *Nature communications* 8.1 (2017): 186.

322 as pulses in the water window by 1.8 μm driver
500 μm -long cell, 3 bar neon gas

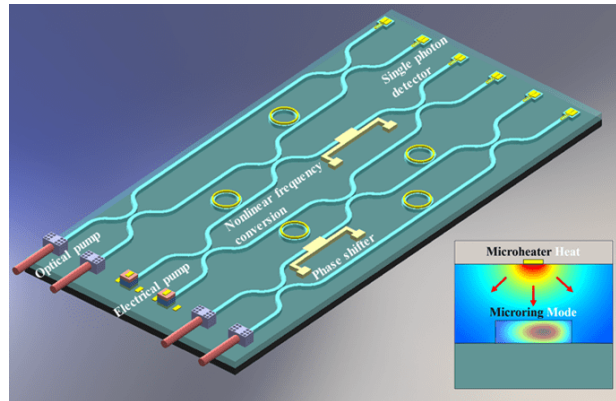


S. L. Cousin et al., *Phys. Rev. X* 7, 041030 (2017)

A novel approach: microfluidic devices for HHG and attosecond science

Inspired to integrated photonics

Vis-IR: Dielectric waveguides

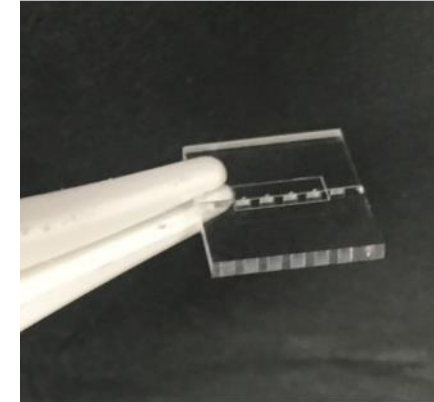


Absorption hinders bulk
Photonics in the XUV-SXR



Replacing dielectric with
hollow waveguides

XUV-X: hollow waveguides



The potentials of microfluidics applied to XUV – soft X:

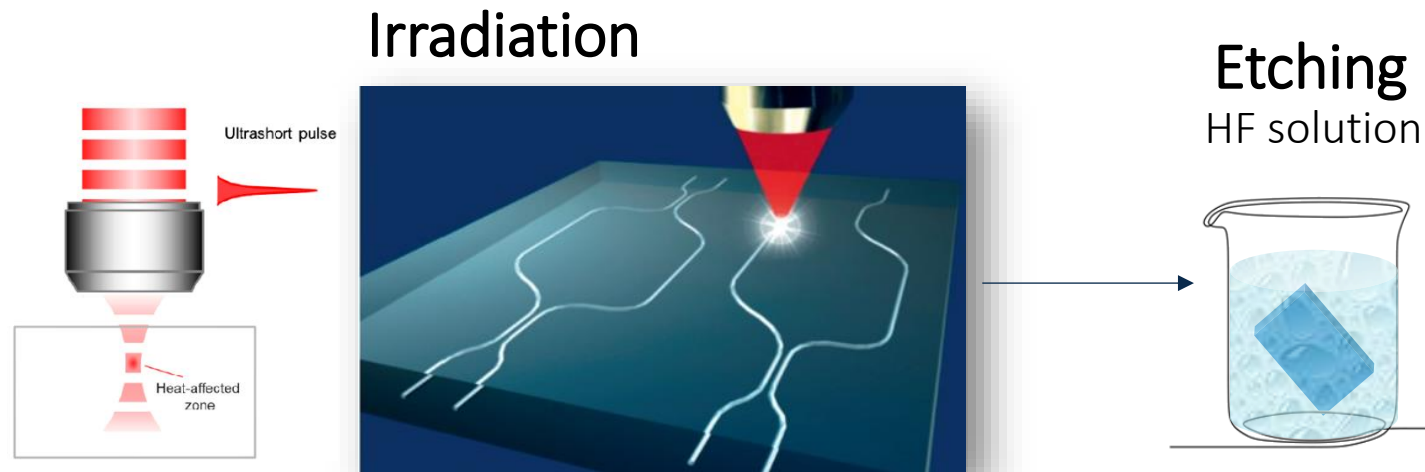
- Efficiency of HHG in capillaries/waveguides
- Propagation the fundamental and XUV/X-rays beams in a waveguide regime
- Micro-manipulation of gas (or liquid samples)
- Miniaturization

The technology

In collaboration with
The Femtosecond Laser Micromachining group
Dr. R. Martínez-Vázquez



Femtosecond Laser Irradiation followed by Chemical Etching (FLICE)

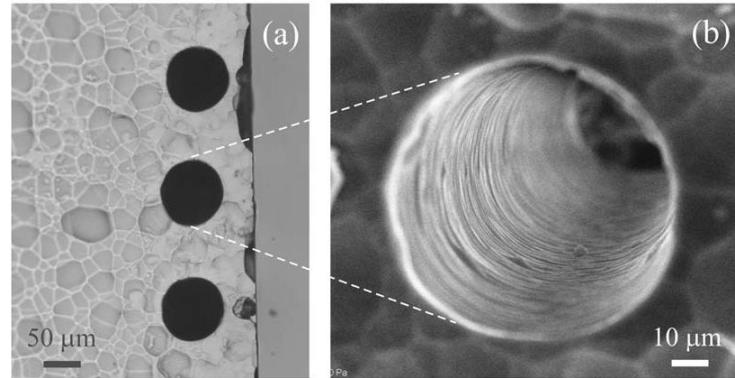


Etching rate ~ 20 times faster in the irradiated regions



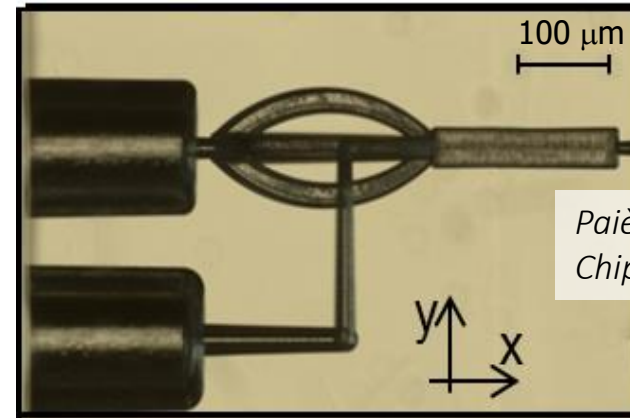
FLICE capability

Maselli et al., *Appl. Phys. Lett.*, 2006



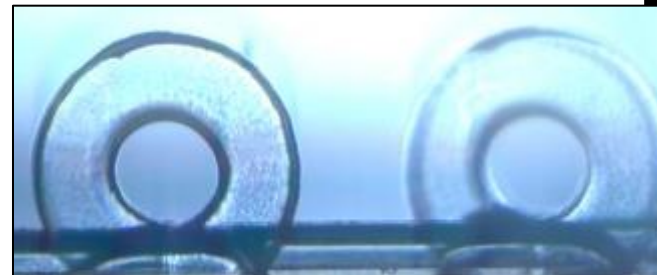
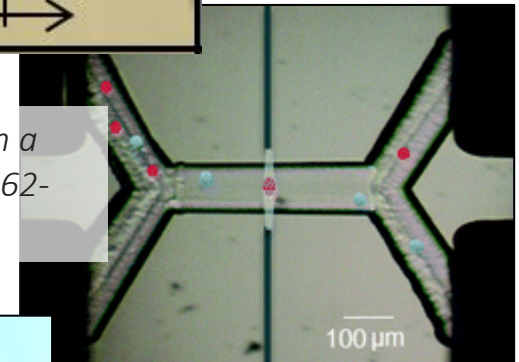
- ✓ 3D hollow structures
- ✓ high accuracy ($<1 \mu\text{m}$)

Lab-On-a Chip (LOC)
integrates multiple laboratory
functions on a single, handheld chip.

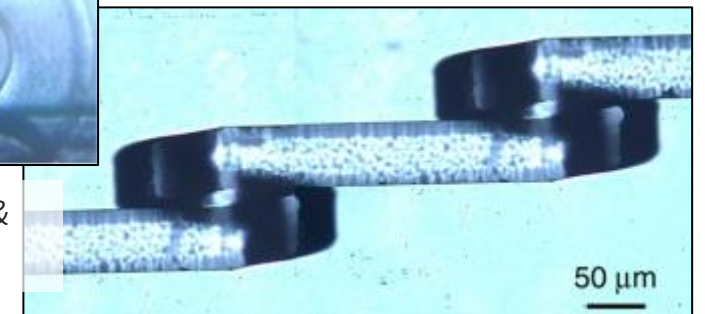


Paiè Petra, et al., *Lab on a Chip* **14** 1826-1833 (2014).

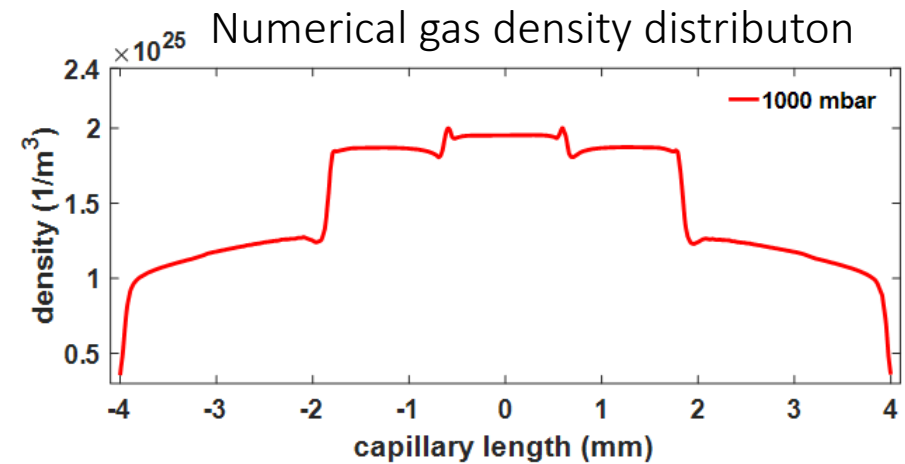
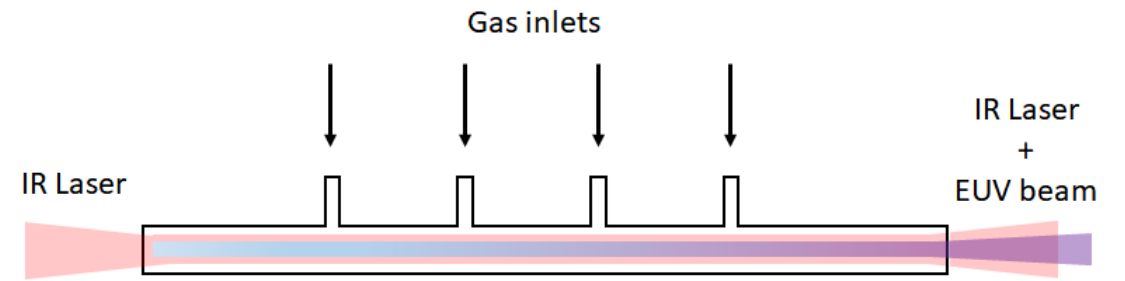
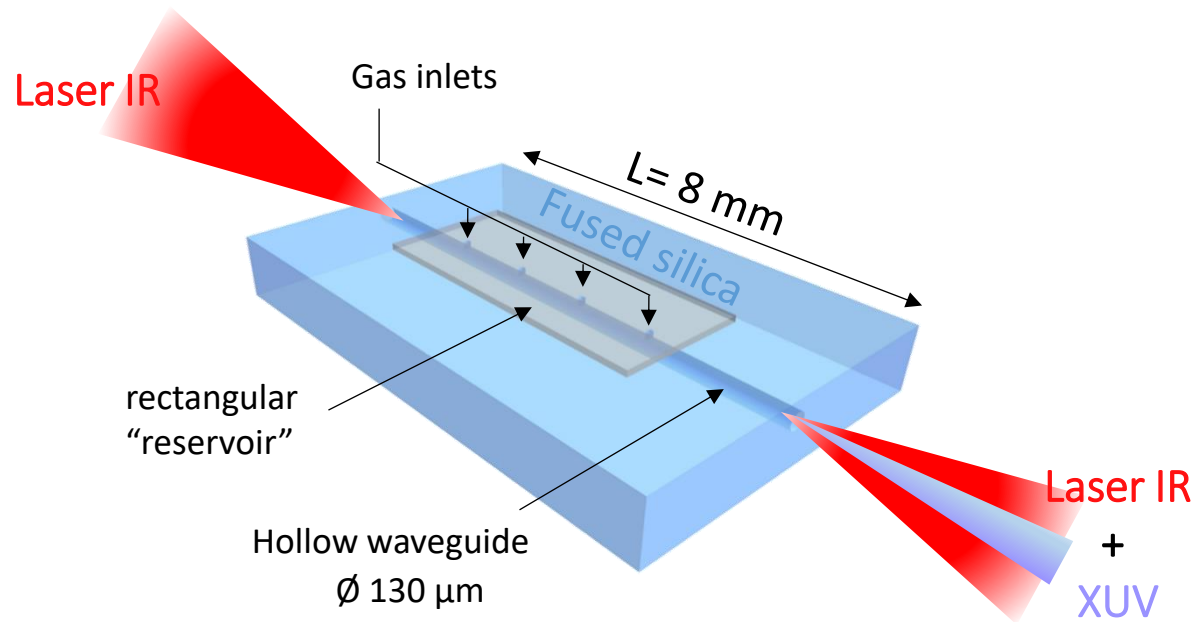
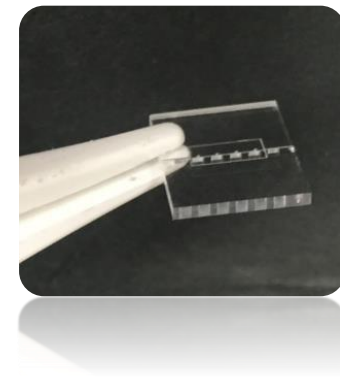
Yang, T., et al, *Lab on a Chip* **15.5** (2015): 1262-1266.



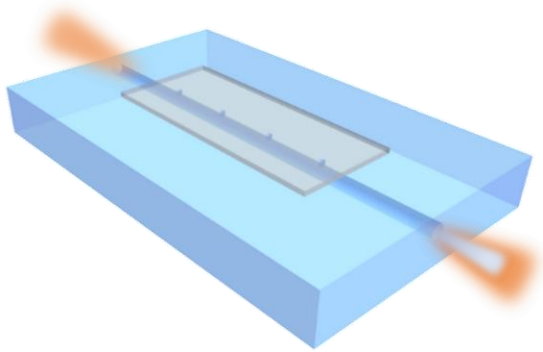
Paiè, Petra, et al. *Microsystems & nanoengineering* **3.1** (2017): 1-8.



A microfluidic device for HHG



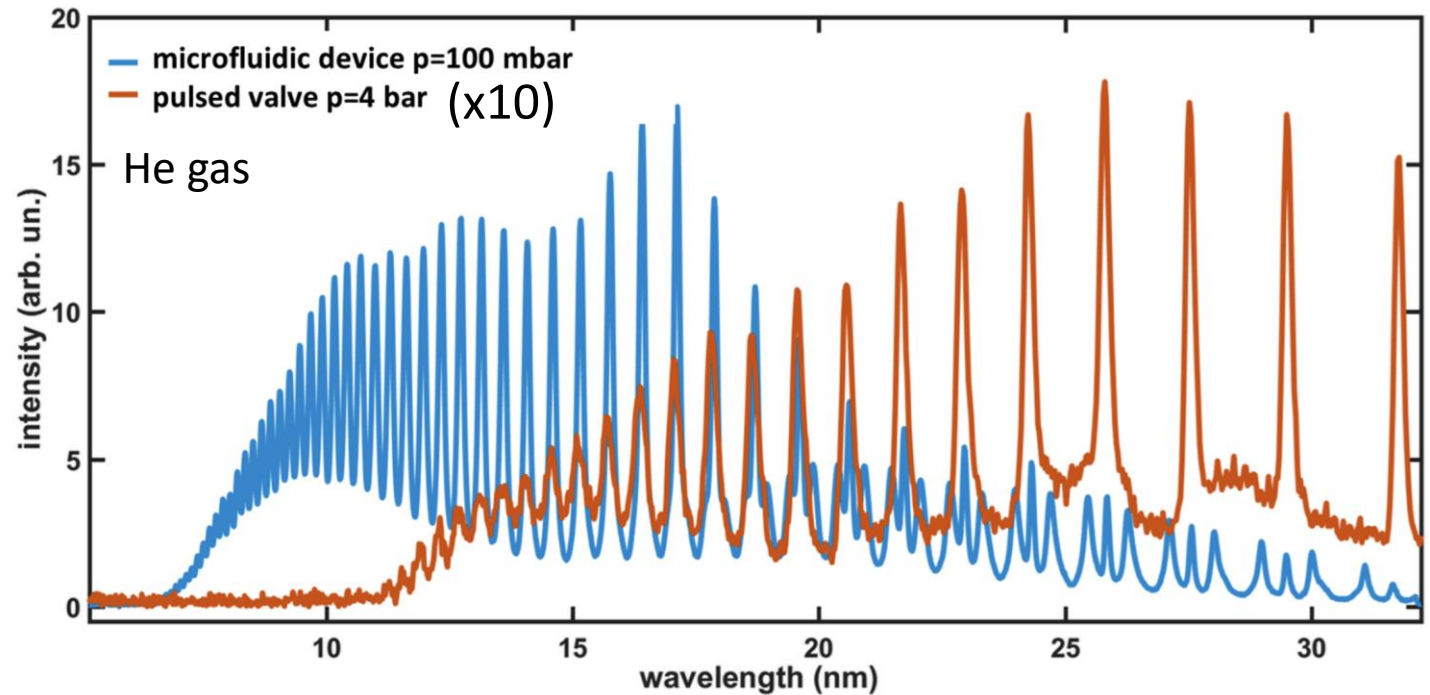
HHG spectra from the microfluidic cell



Amplified Ti:Sa laser system @ 800nm
(Amplitude, Aurora laser system)

- Repetition rate 1 kHz
- Pulse energy 450 μ J
- Pulse duration 30 fs

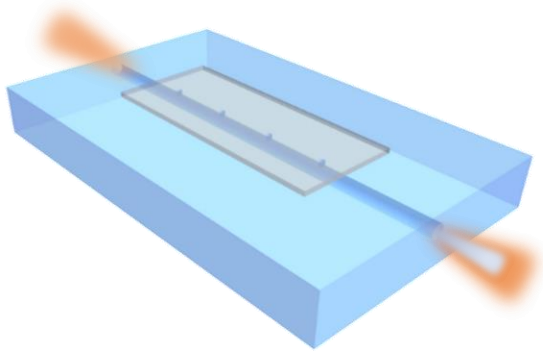
Microfluidic device vs gas valve



A. G. Ciriolo et al., *Journal of Physics: Photonics* 2 024005 (2020)

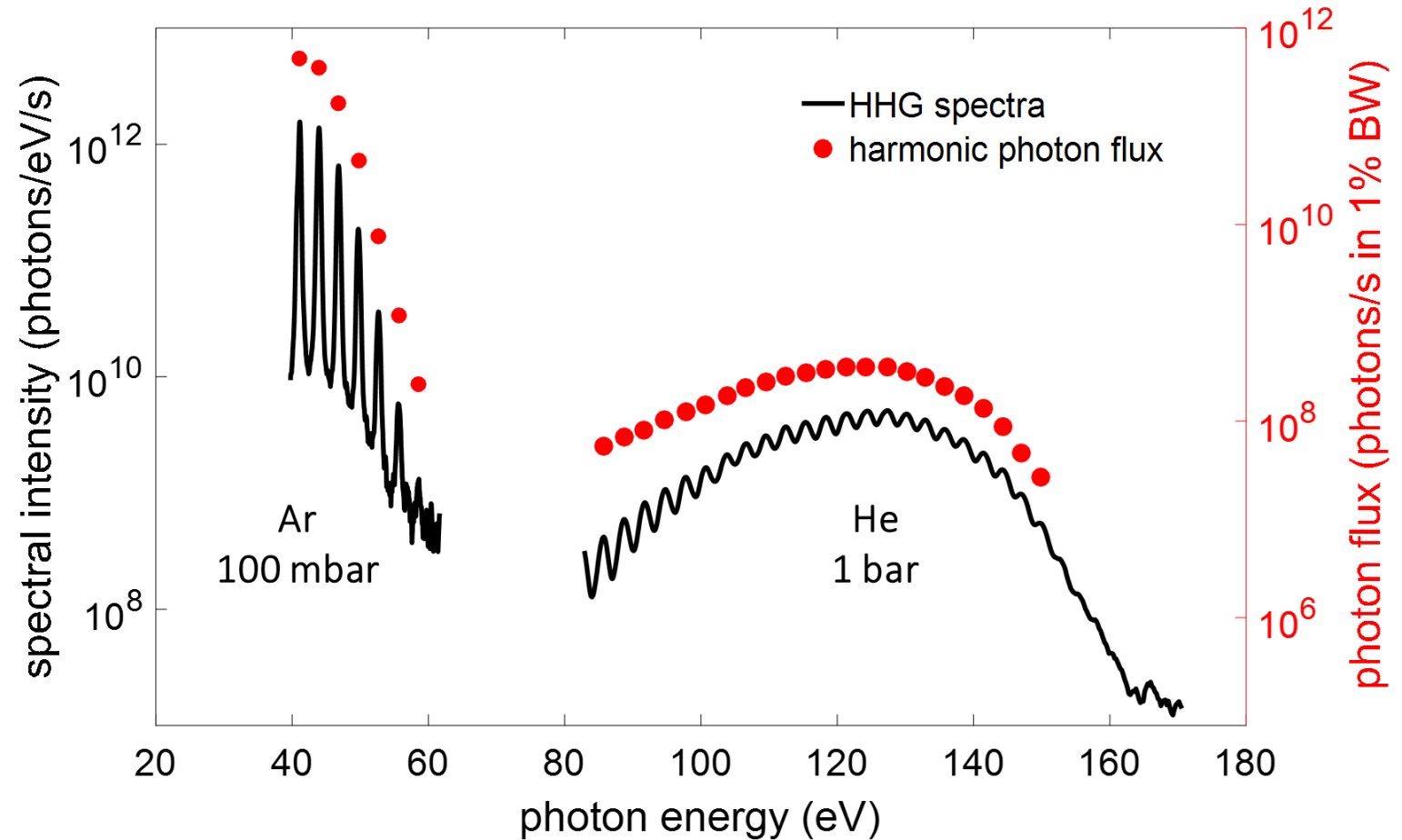
A. G. Ciriolo, et al., *APL Photonics* 7.11 (2022).

HHG spectra from the microfluidic cell



Amplified Ti:Sa laser system @ 800nm
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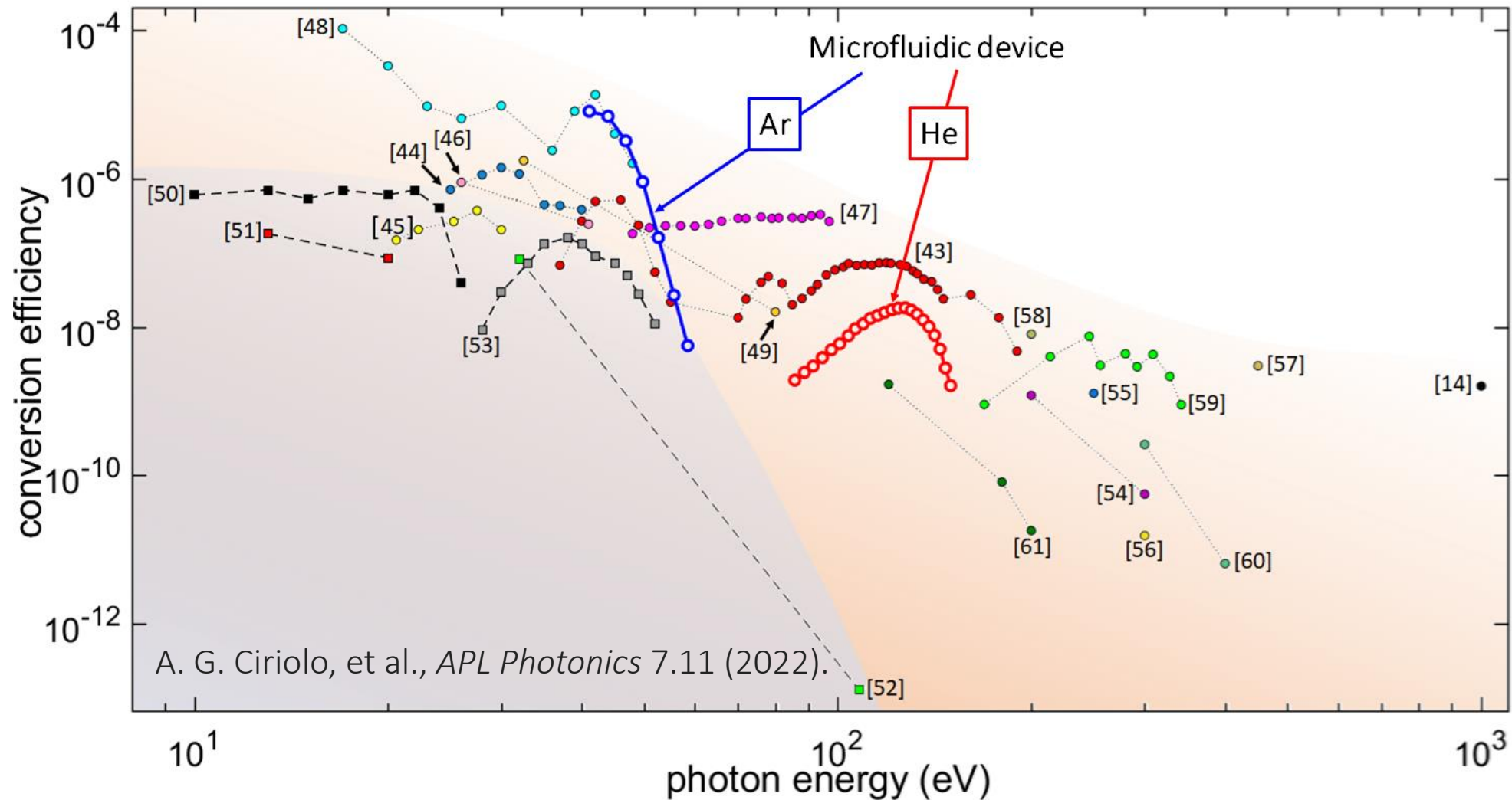
- Repetition rate 1 kHz
- Pulse energy 450 μ J
- Pulse duration 30 fs



A. G. Ciriolo et al., *Journal of Physics: Photonics* 2 024005 (2020)

A. G. Ciriolo, et al., *APL Photonics* 7.11 (2022).

Performances of the microfluidic source



[48] E. J. Takahashi, et al., *IEEE JSTQE* 10, 1315 (2004) → gas cell

[43] C. Ding, *Optics express*, vol. 22, pp. 6194-6202 (2014) → capillary

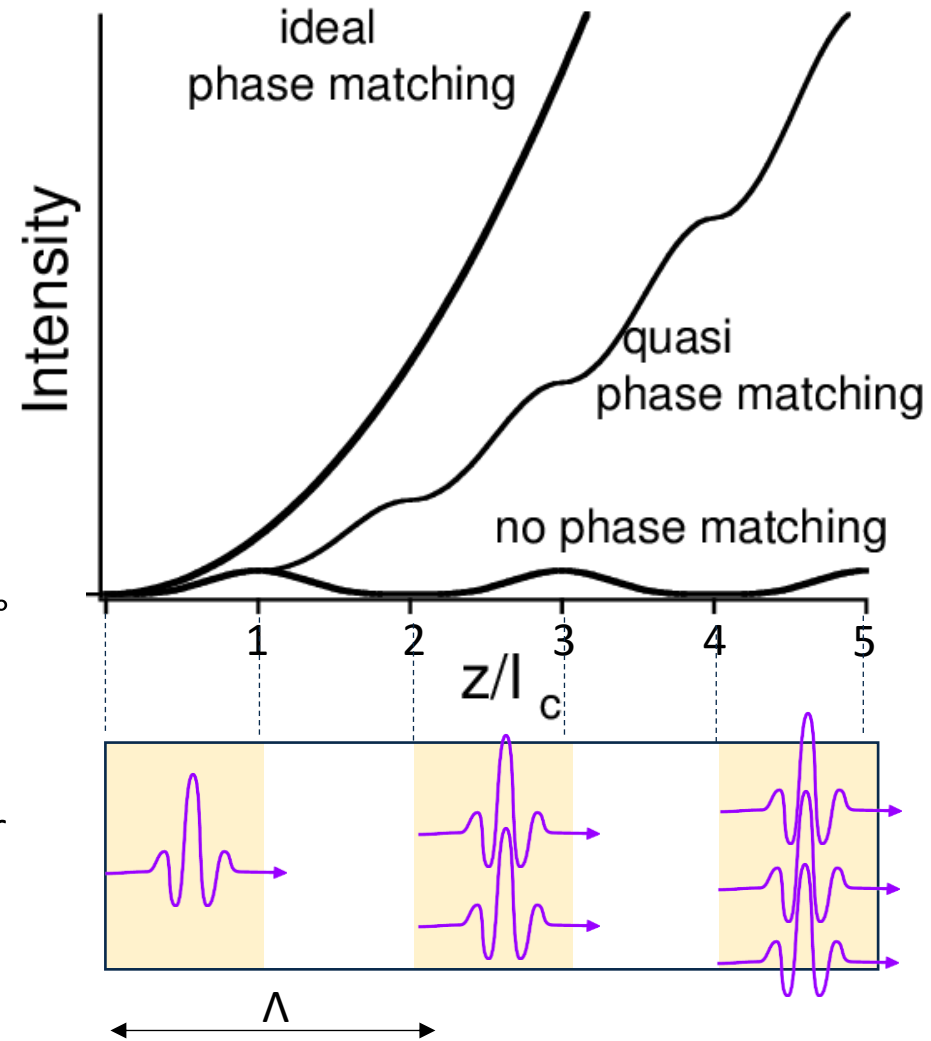
Quasi Phase Matching (QPM)

Coherence length

$$l_c = \frac{\pi}{\Delta k}$$

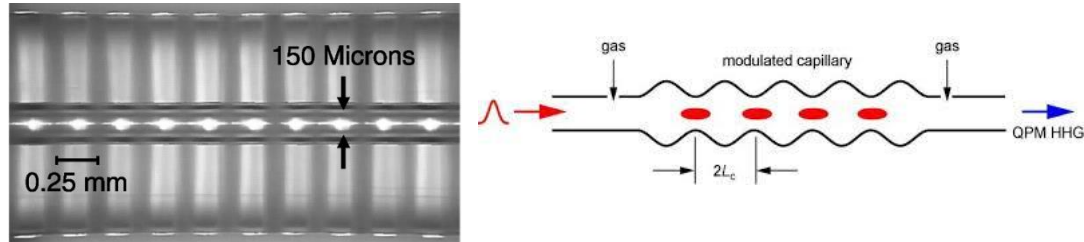
Distance at which the phase difference of two emitters is 180°

spatial modulation of the nonlinear interaction at a regular interval $\Lambda = 2l_c$



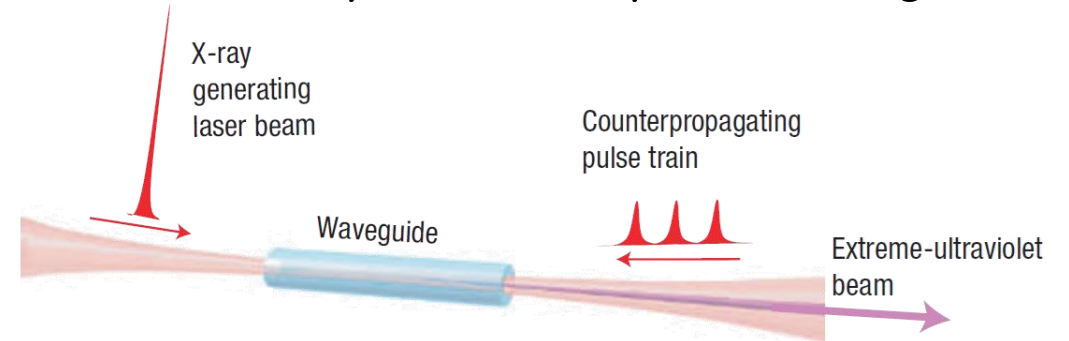
QPM strategies in HHG

Intensity modulation by waveguide diameter modulation



Gibson, Emily A., et al. *Science* 302.5642 (2003): 95-98.

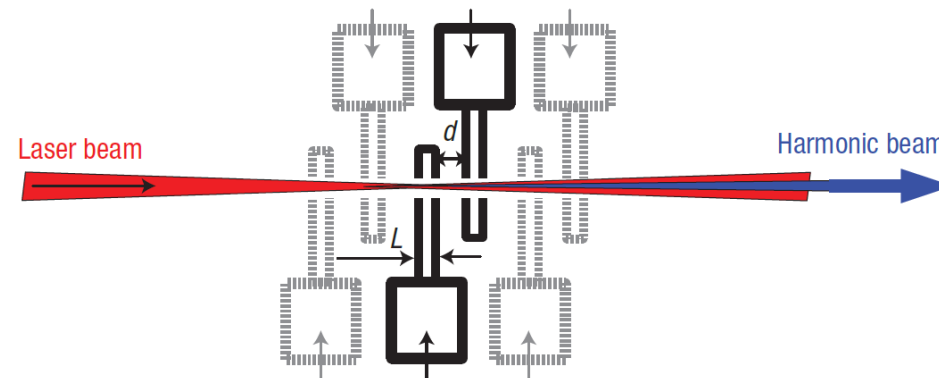
Intensity modulation by mode beating



Zhang, Xiaoshi, et al. *Nature Physics* 3.4 (2007): 270-275.

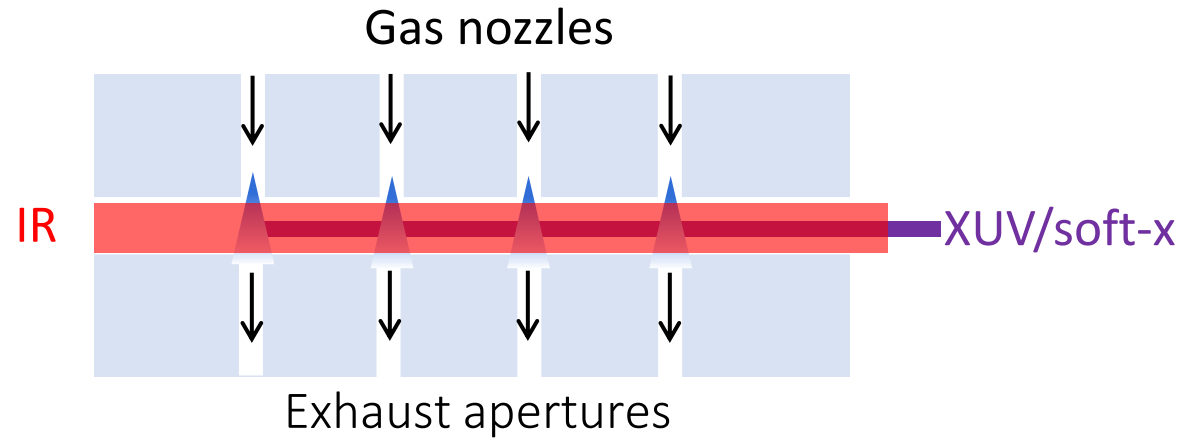
Zepf, Matthew, et al. *PRL* 99.14 (2007): 143901.

Gas density modulation by multiple gas-jets

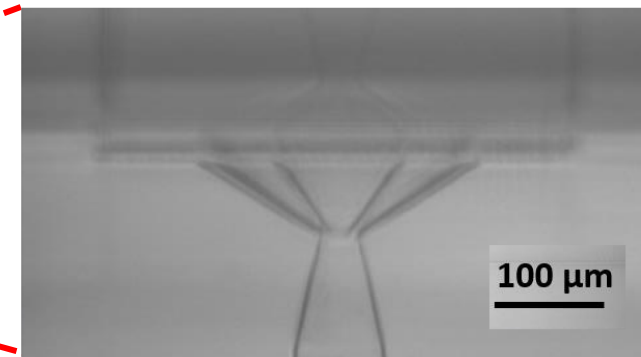
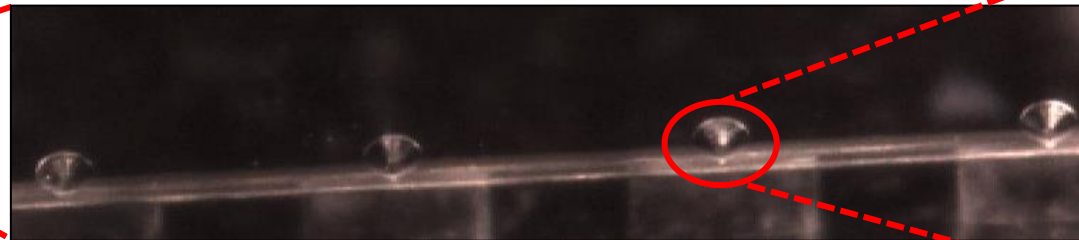
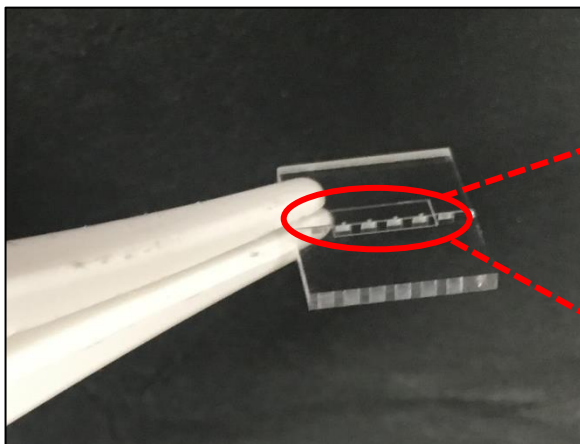


Seres, Jozsef, et al. *Nature Physics* 3.12 (2007): 878-883.

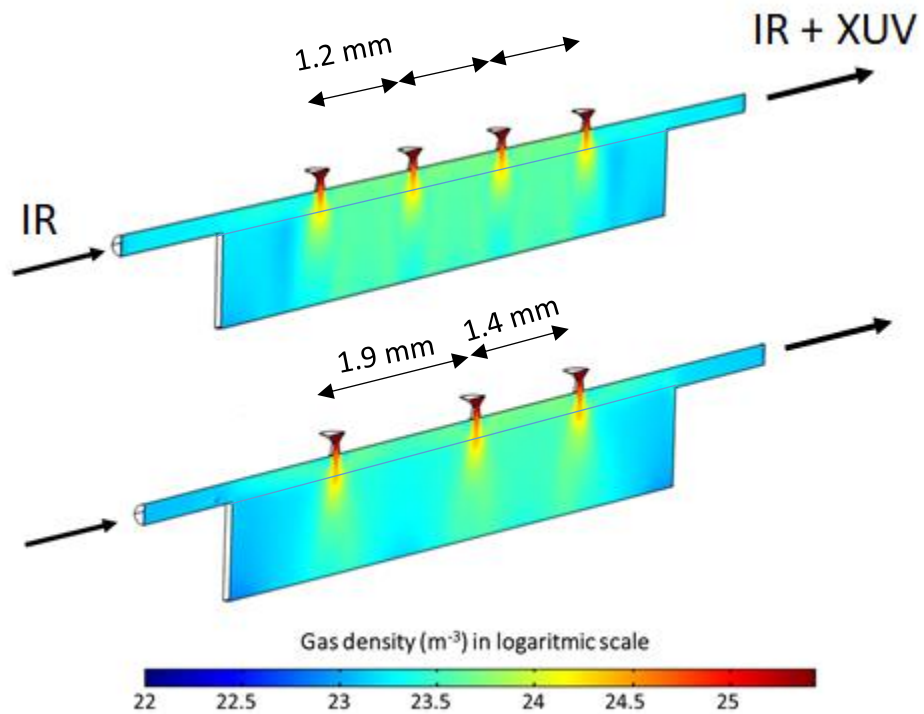
QPM in an integrated array of gas-jets



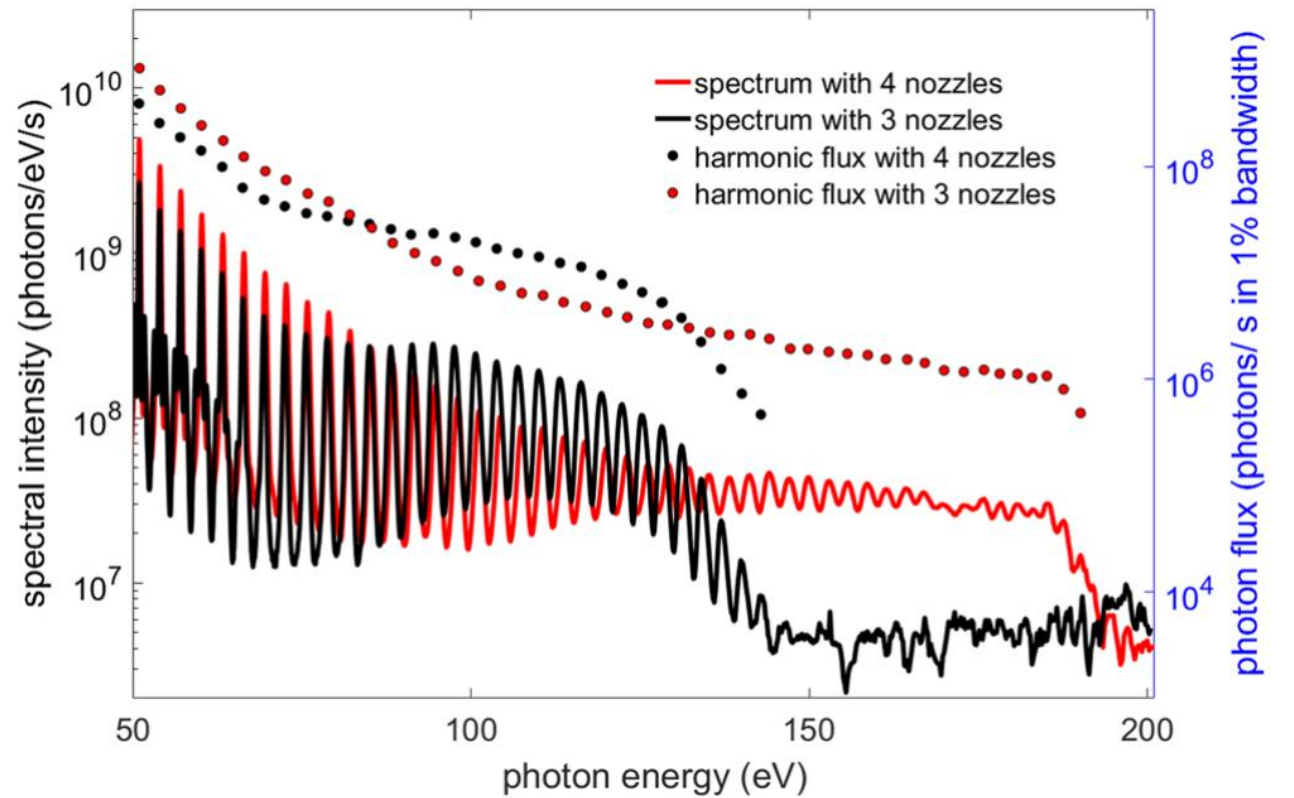
Array of **De Laval micro-nozzles** for confined supersonic gas expansion



HHG in multi-nozzle devices



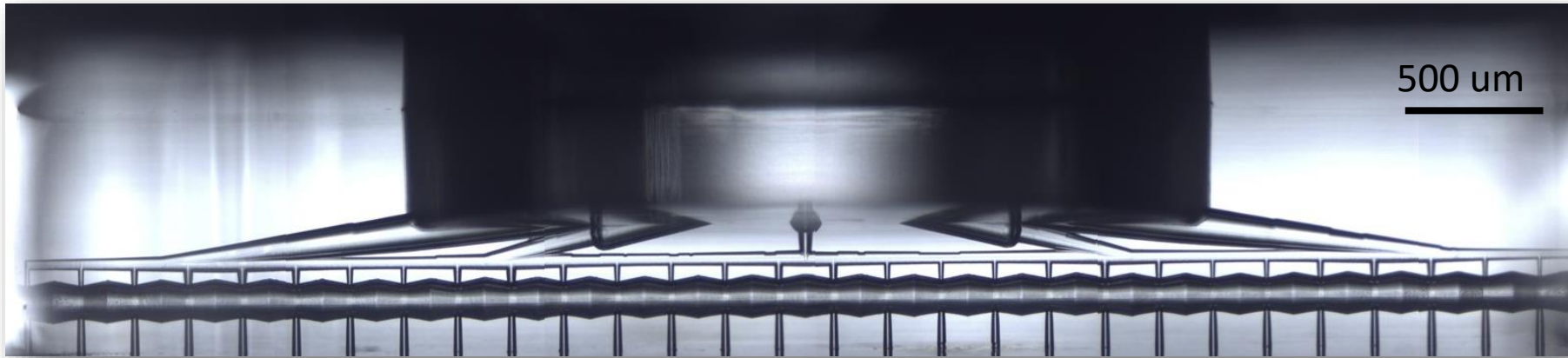
Helium gas



A. G. Ciriolo, et al., *APL Photonics* 7.11 (2022).

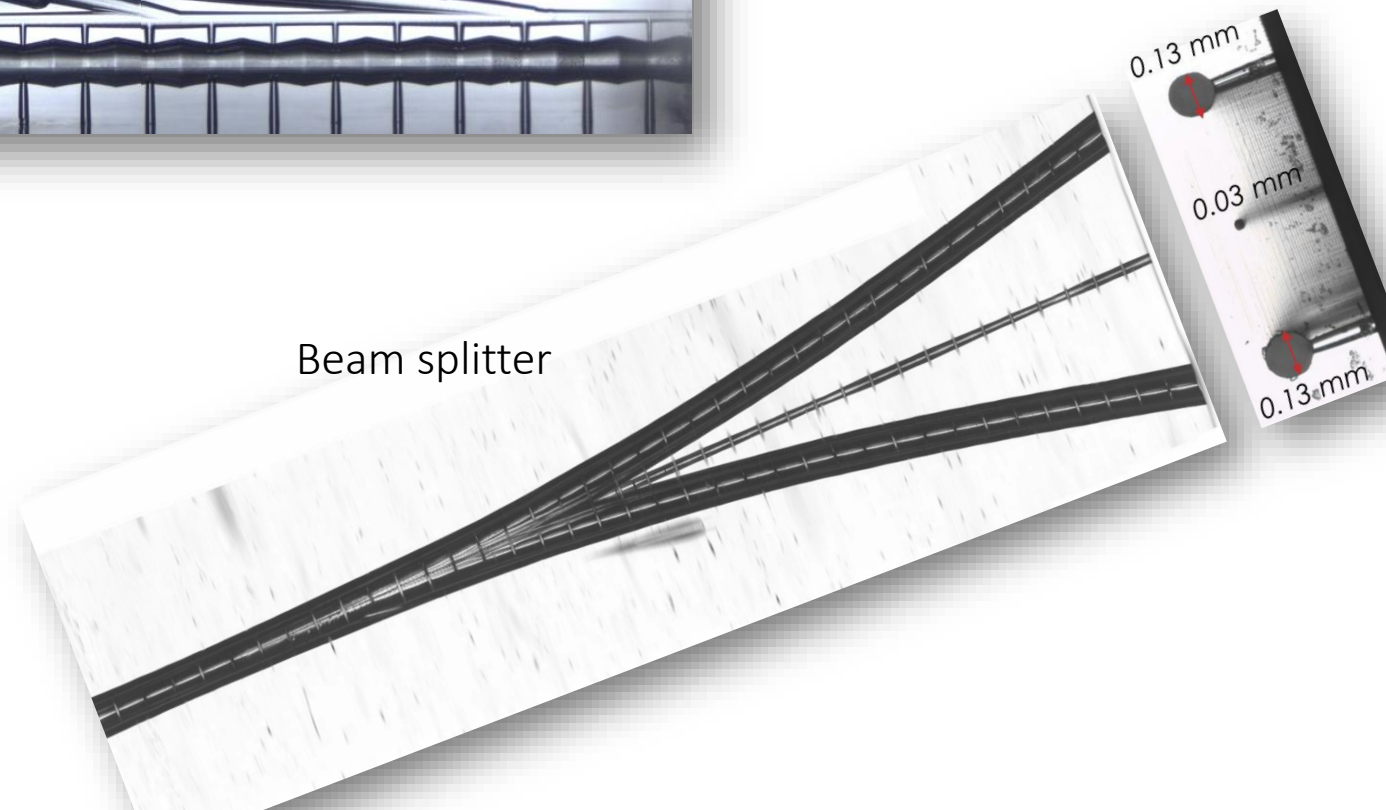
Toward more complex devices

30 nozzles - periodic modulation of the diameter (from 100 to 130 μm)



Ciriolo, A. G., et al. *Micromachines* 11.2 (2020): 165.

Lab-On-a-Chip
or XUV/Soft-X spectroscopy
and Atto Science



Towards LOC for ultrafast XUV science: IR rejection

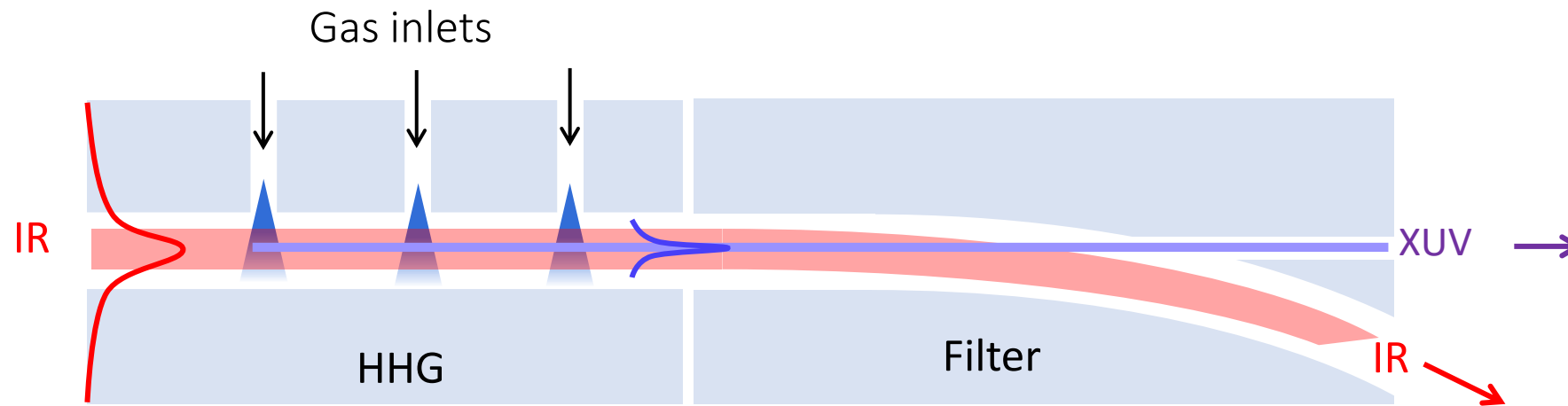
Metallic thin filters

IR attenuation $I/I_0 \approx 10^{-6} \div 10^{-7}$
Expensive and prone to damage

Al filter: 20-70 eV
Zr filter: 60-200 eV



Integrated beam splitter



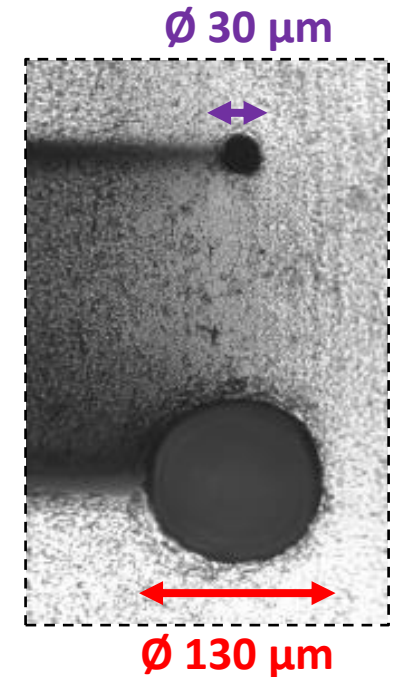
XUV beam spatial property

Confined in the center of the waveguide
 $r_{xuv} \ll a$ (with a waveguide radius)

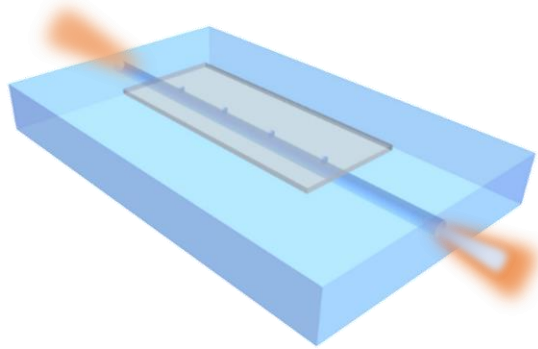
IR attenuation law

$$\alpha_{nm} = \frac{\lambda^2}{a^3} U_{nm}(\nu)$$

Output section



Structured light in a hollow waveguide



Hybrid modes (EH)

Non-zero electric and magnetic fields in the direction of propagation.

Transverse electric (TE) modes

No electric field in the direction of propagation.

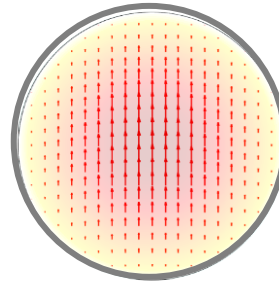
Transverse magnetic (TM) modes

No magnetic field in the direction of propagation.

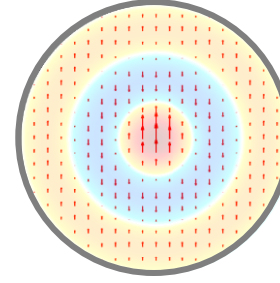
Vector modes/ beams: a beam where the **polarization state varies spatially** across the beam profile

modes

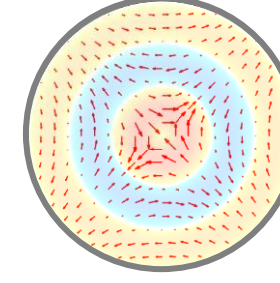
EH_{11}



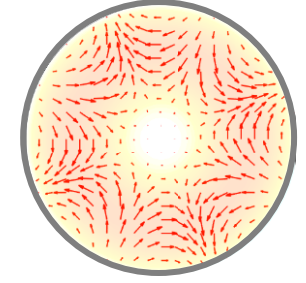
EH_{13}



EH_{23}



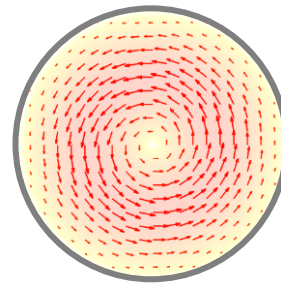
EH_{41}



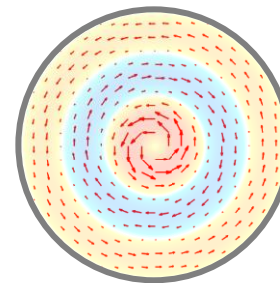
$$E_{n,m}(\theta) = J_{n-1}(u_{n,m}r/a) \cos n\theta$$

$$E_{n,m}(r) = J_{n-1}(u_{n,m}r/a) \sin n\theta$$

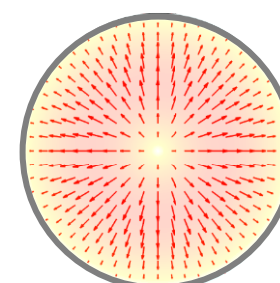
TE_{01}



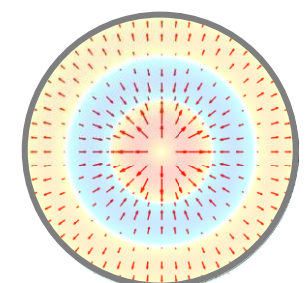
TE_{03}



TM_{01}



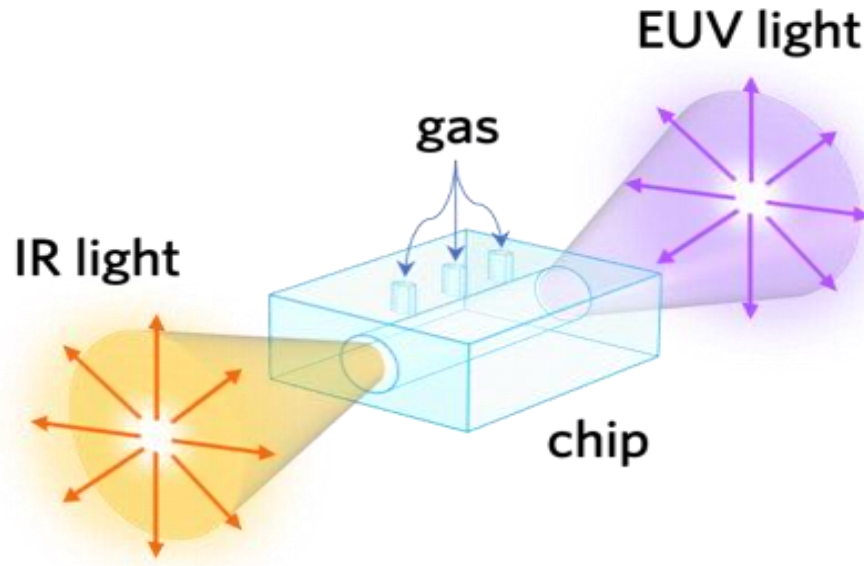
TM_{03}



$$E_{0,m}(\theta) = J_1(u_{0,m}r/a)$$

$$E_{0,m}(r) = J_1(u_{0,m}r/a)$$

HHG driven by vectorial fields



HHG locally preserves the polarization orientation of the driver:
generation of XUV vector beam of TM_{01} and TE_{01} symmetry

Laser
polarization



Vortex waveplate
(VWP)

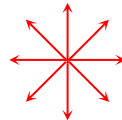


Liquid crystal

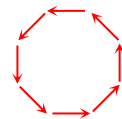


Input
polarization

RAD

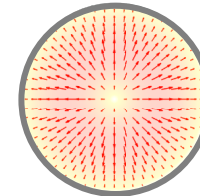


AZI

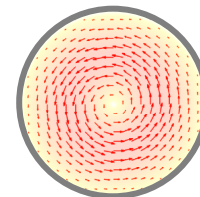


Excited mode

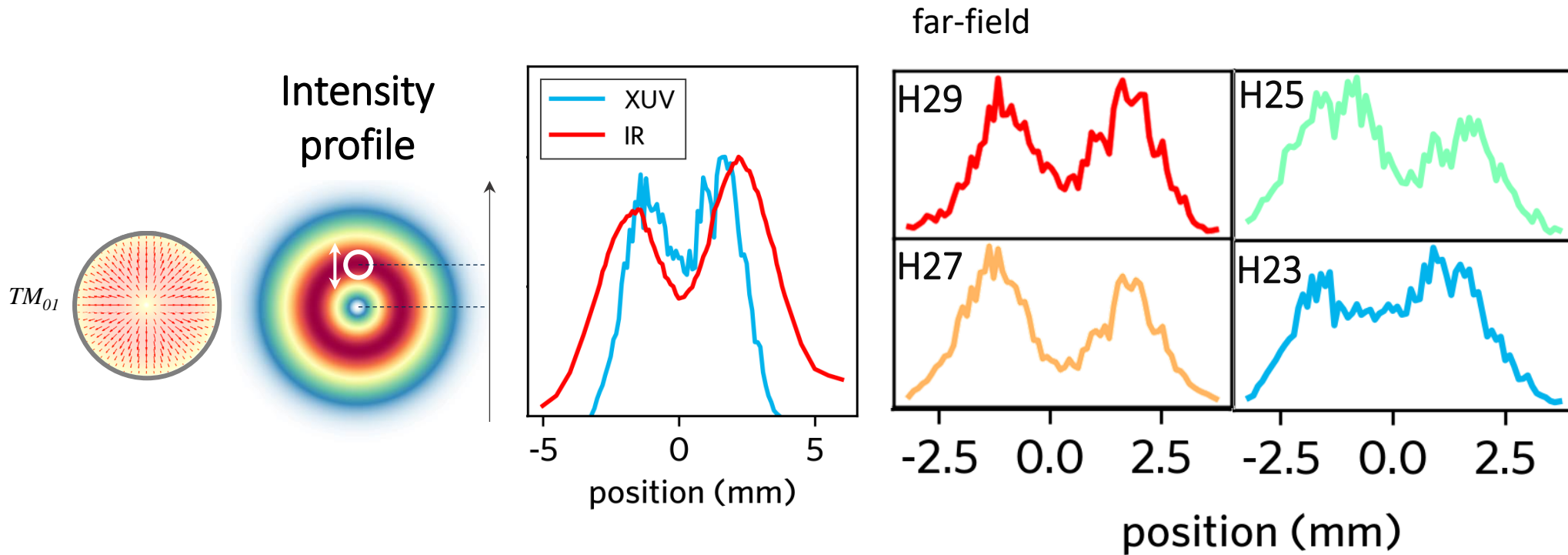
TM_{01}



TE_{01}

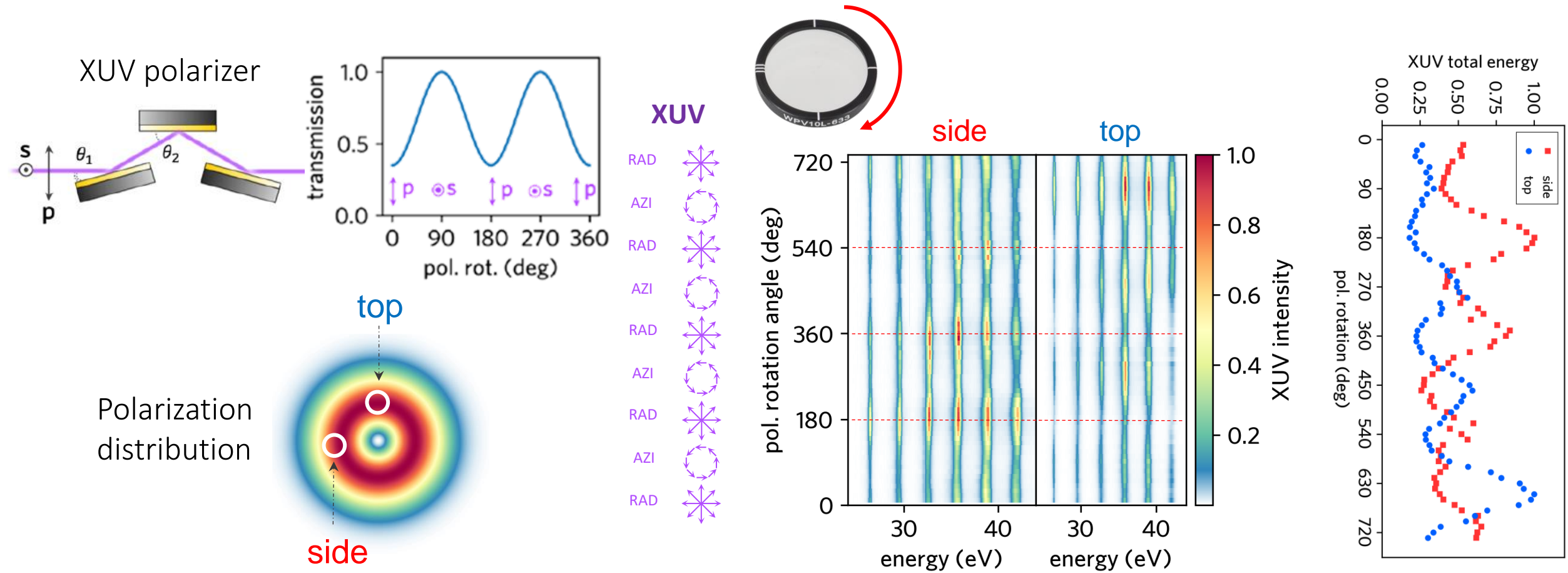


XUV Vector Beam: intensity profile



Singularity in the center of the intensity profile

XUV vector beam: polarization

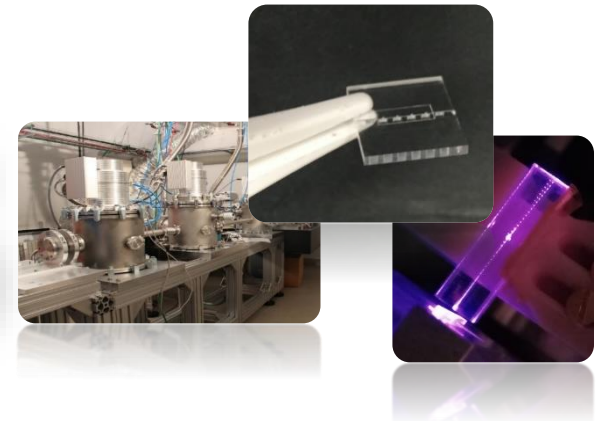


In collaboration with Dr. R. Piccoli
(Università Cà Foscari, Venezia)

R. Piccoli et al., <http://arxiv.org/abs/2403.11006>.

Conclusion

- Prototyping of **hollow waveguide-based circuits** for efficient HHG
- A compact table-top source of coherent **XUV vector beams**
- Scaling to XUV generation in the **water window** (280-500 eV) by using near/mid-IR drivers
- More complex systems: X-rays LOC





acknowledgments



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H2020-MSCA-ITN-2019 GA 860553



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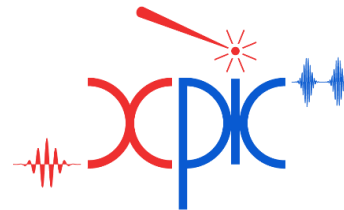
ERC-2018-PoC GA 813103



CNR laboratorio congiunto



MIUR ELI ESFRI ROADMAP



H2020-FETOPEN-2020 GA 964588

CONQUEST

MIUR PRIN 2020JZ5N9M

Thank you for your attention!