EUV “Photonics” of High-Harmonic Generation and Applications

Emily Gibson, Ariel Paul, Time Lei, Xiaoshi Zhang, Ariel Libertun, Daisy Raymondson, Margaret Murnane, and Henry C. Kapteyn

JILA, University of Colorado and NIST

Ivan Christov

Sofia University

David Attwood, Yanwei Liu

Center for X-Ray Optics,Lawrence Berkeley Labs.

NIST-CU

NIST-CU

Colorado State University

EUV

An NSF Engineering Research Center

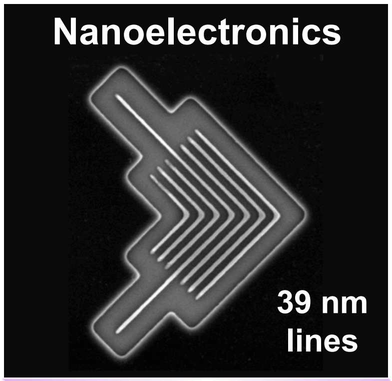
The Kapteyn-Murnane Group

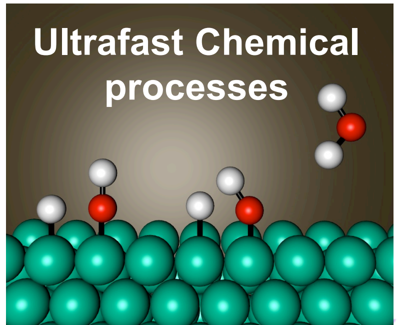
NSF Engineering Research Center on Extreme Ultraviolet Science and Technology

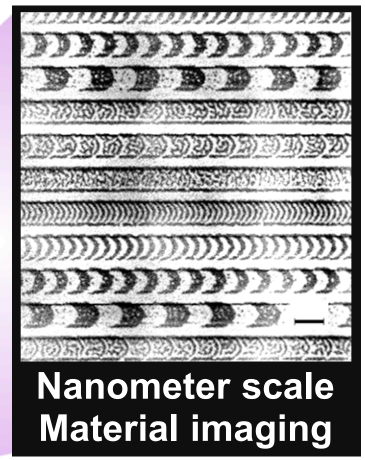
EUV ERC

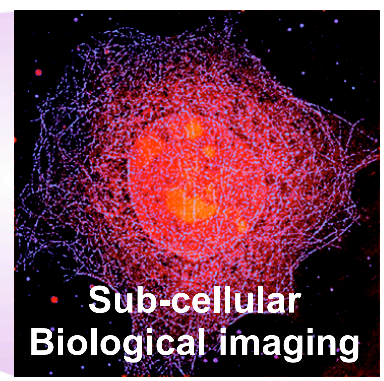
NSF ERC for Extreme Ultraviolet Science & Technology

Vision: make EUV light technology widely accessible to solve challenging scientific and technological problems.

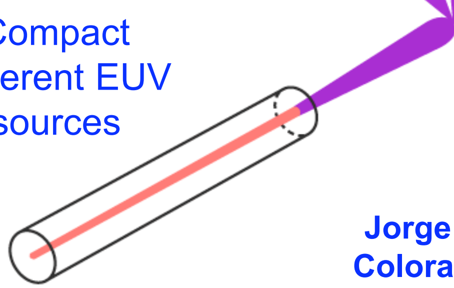








Compact coherent EUV sources



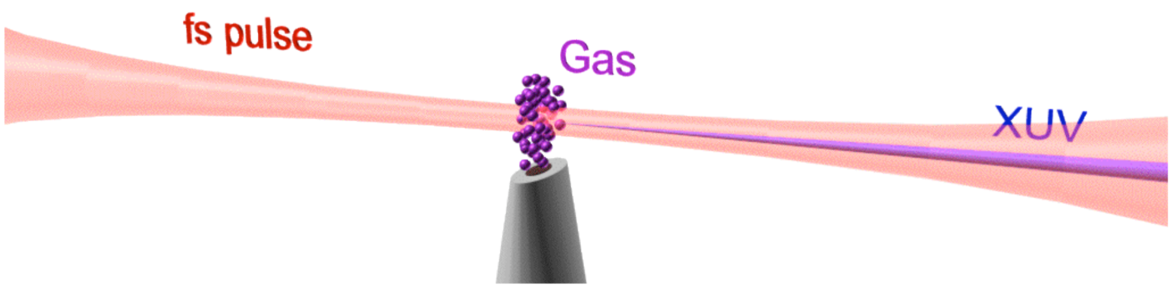
Jorge J. Rocca, Director
Colorado State University
Margaret M. Murnane, Deputy Director
University of Colorado, Boulder

Outline

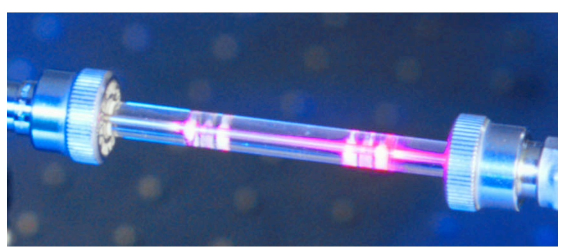
- Coherent short wavelength science - "laser-like" beams in EUV with femto-to-atto time resolution
- Extend wavelength range and efficiency using "photonics"
- Applications
  - Plasma dynamics studies
  - *Ultrafast, high frequency, photoacoustic response of materials*
  - Ultrafast photoemission spectroscopy to observe molecular motion on surfaces

Extreme nonlinear optics: high harmonics (HHG)

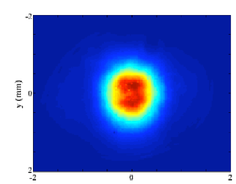
- Coherent EUV light is generated by ionizing a gas with a fs laser
- Broad range of harmonics generated 4.5 up to 550 eV
- "Laser-like" coherent beams in EUV
  - R. Bartels et al, Science 297, 376 (2002), Nature 406, 164 (2000)



Gas jet

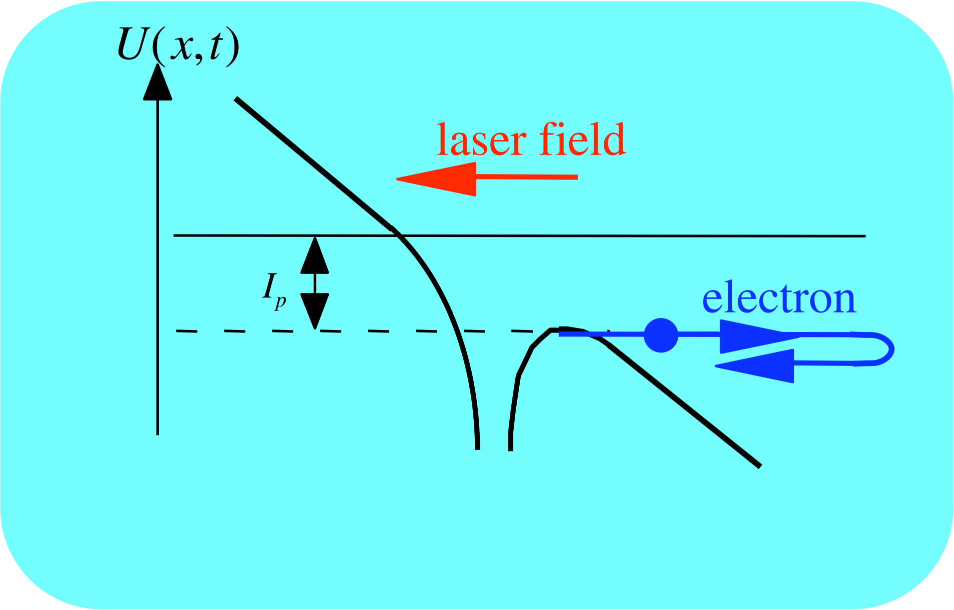


Fiber

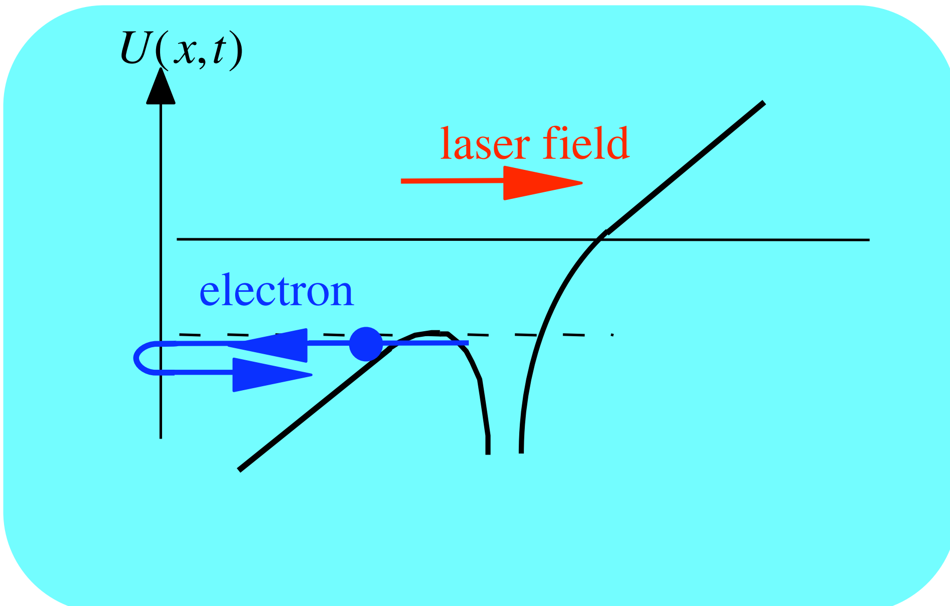


EUV beam

X-rays are generated by atoms being ripped apart



X-rays are generated by atoms being ripped apart

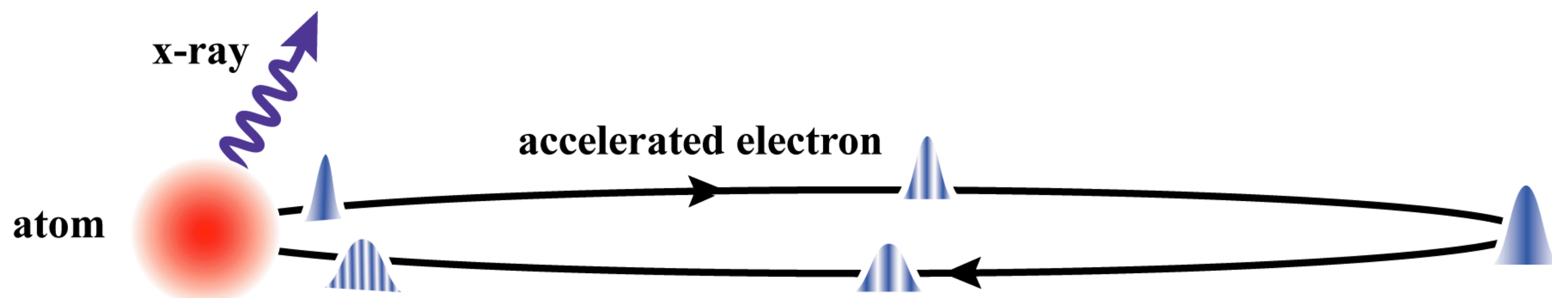


$$h\upsilon_{cutoff} = I_p + 3.2U_p$$

ionization potential of atom

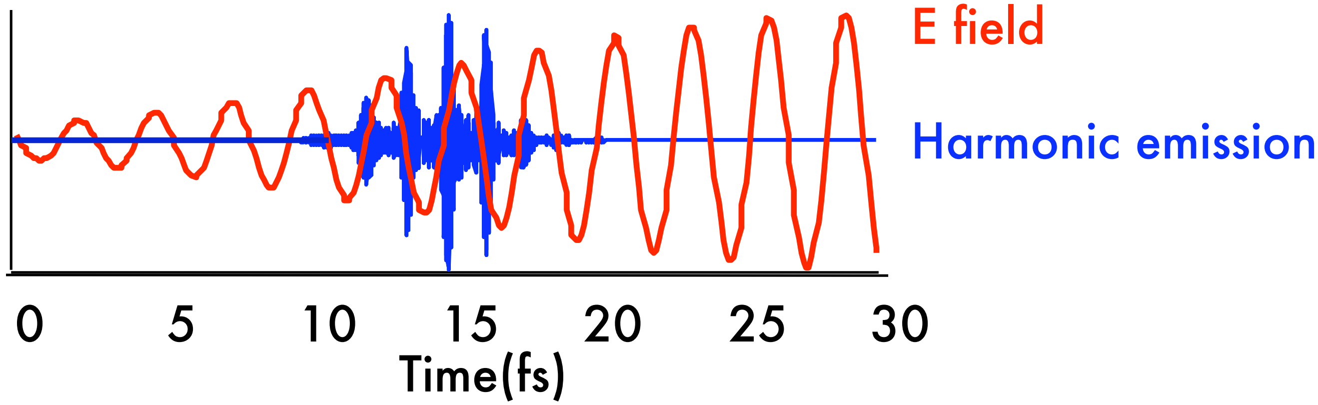
$U_p \approx I_L \lambda^2$ quiver energy of e^-

$$\varphi_{x\text{-ray}} \approx \varphi_{Laser} \text{ and } I_{Laser}$$



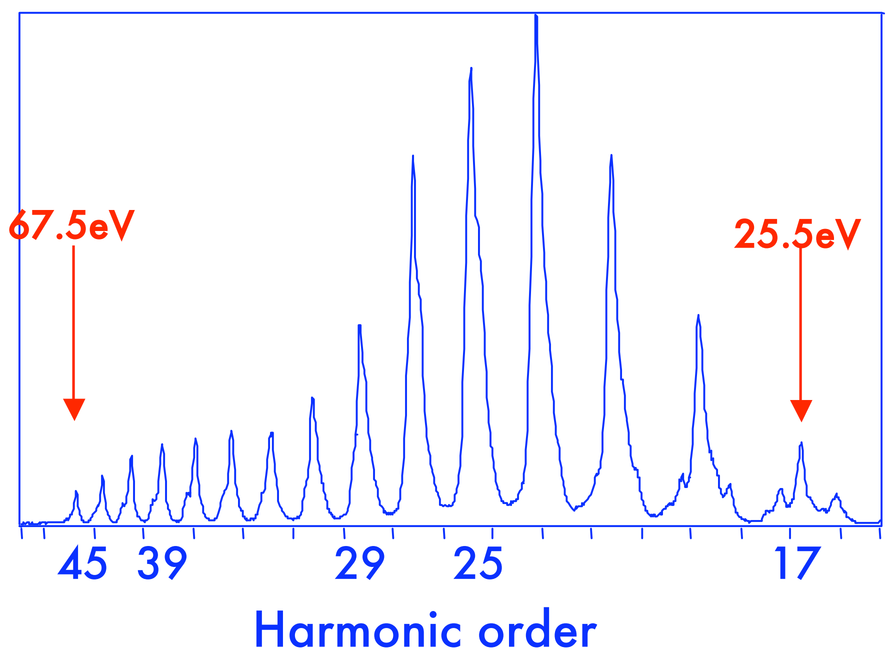
HHG emission in time and frequency:

time



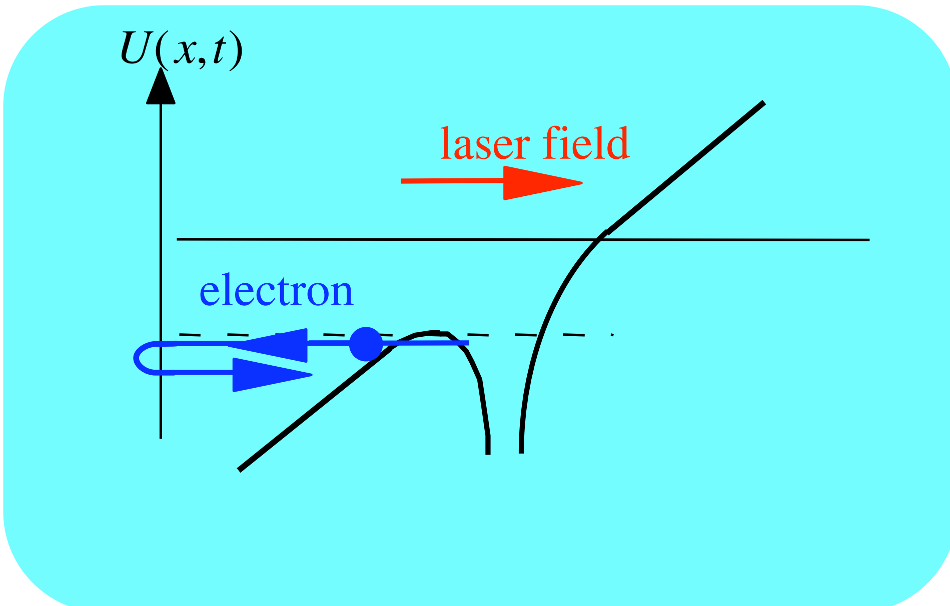
I. Christov et al, PRL 78, 1251, (1997)

frequency



J. Zhou et al, PRL 76(5), 752-755 (1996)

X-rays are generated by atoms being ripped apart

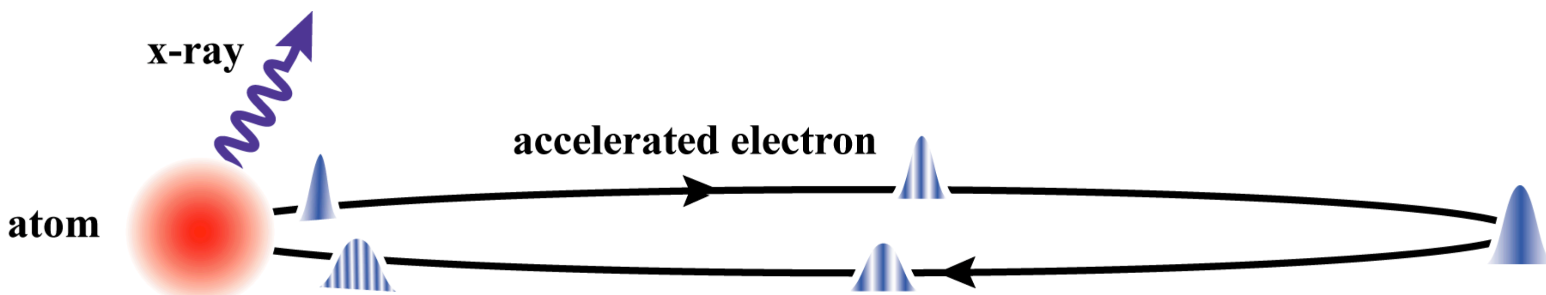


$$h\upsilon_{cutoff} = I_p + 3.2U_p$$

ionization potential of atom

$U_p \approx I_L \lambda^2$
quiver energy of e^-

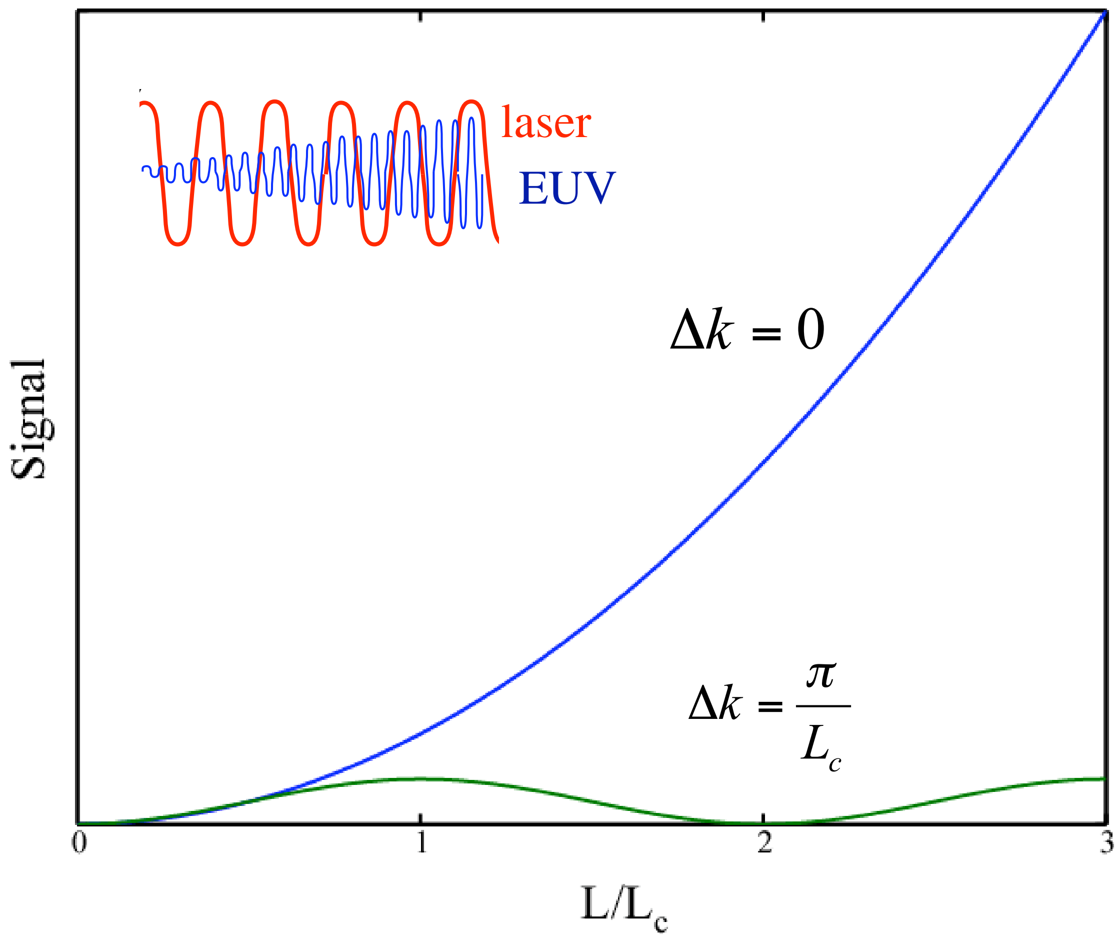
$\varphi_{x\text{-}ray} \approx \varphi_{Laser}$ and I_{Laser}



No limit in theory to harmonic photon energy - BUT two practical limits until recently-- 1) Phase matching; 2) No emission from ions

Need for phase matching

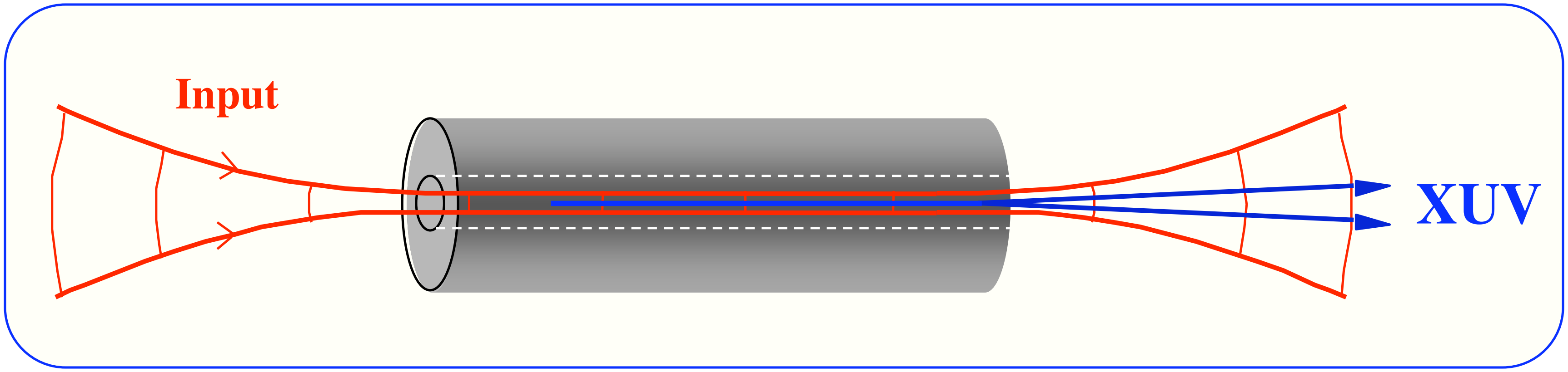
$$\overline{E}(\overline{r},t) \sim e^{-i(\omega_i t - k_i z)} \quad , \quad I_{HHG} \propto \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2} \quad where \;\; \Delta k = k_{hhg} - q k_{vis}$$



- **Use phase matching (Δk=0) for efficient EUV generation**
- ***If Δk ≠ 0, adjust or restrict emission from regions that are out of phase***
- **Coherence lengths for HHG in the presence of high levels of ionization are μm - mm**

Phase-matched frequency conversion in waveguides:

C. Durfee et al., Optics Letters 22, 1565 (1997)
A. Rundquist, et al, Science, vol. 280, pp. 1412-1415 (1998)

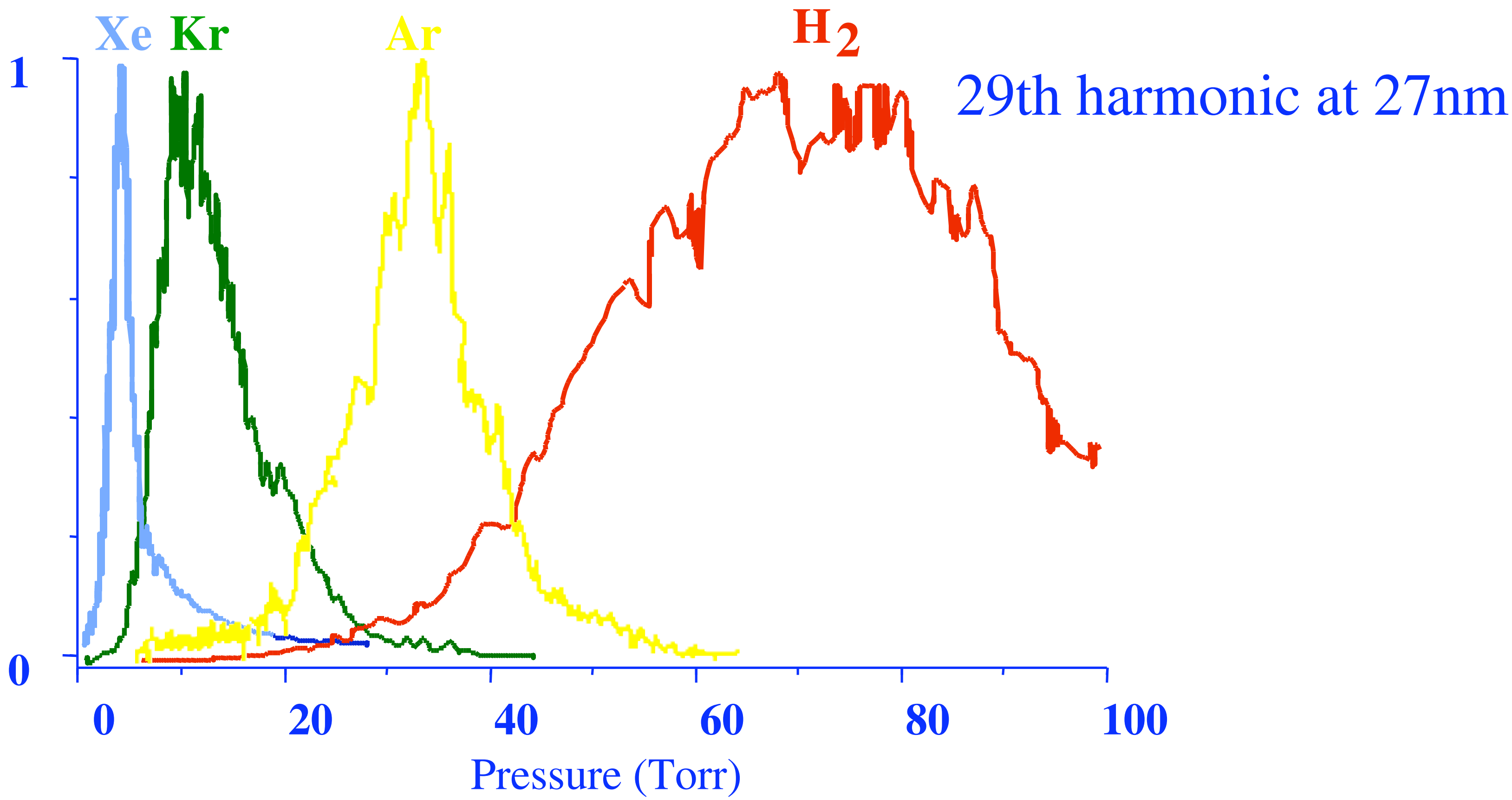


- Waveguide creates plane-wave geometry
- Waveguide can control the phase velocity ($v_p = \omega/k$)

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

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

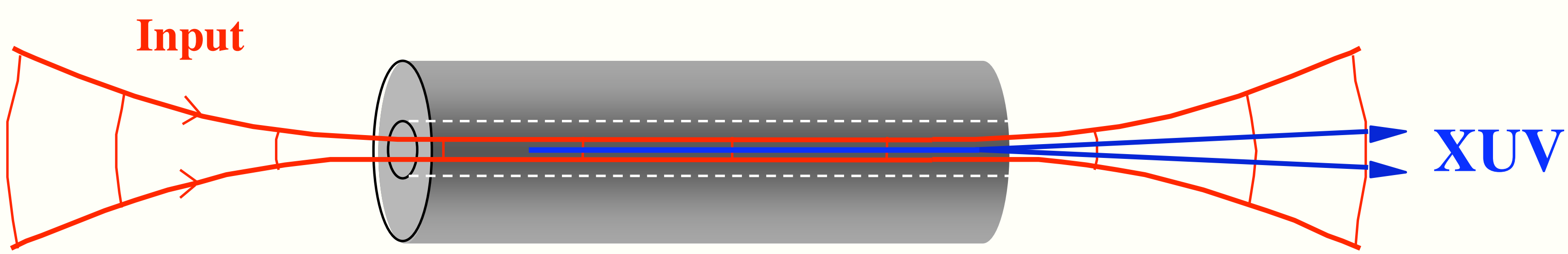
Pressure-tuned phase-matching of soft x-rays:



- Phase-matched length in fiber: 1-3 cm
- Output enhanced by 10^2-10^3
- >10^{12} Photons/sec @ 50 eV

Problem: HHG generated during ionization:

C. Durfee et al., Optics Letters 22, 1565 (1997)
A. Rundquist, et al, Science, vol. 280, pp. 1412-1415 (1998)



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

vacuum gas waveguide ionization

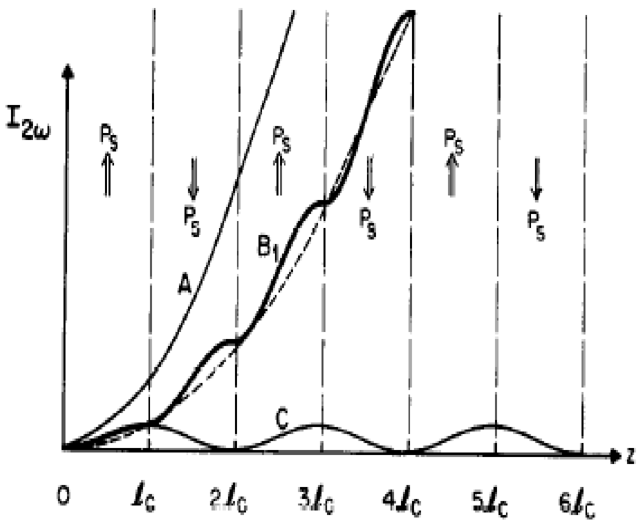
Higher harmonics are generated at higher laser intensities and higher levels of ionization => impossible to WG phase-match above ≈80eV or ionization > 5%

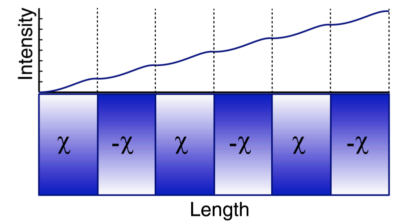
Quasi-Phase-Matching can adjust for phase mis-match:

- Traditional Quasi-Phase-Matching, $\Delta k = K_m = \dfrac{2\pi m}{\Lambda}$

Λ= Periodicity of nonlinear medium

$$m = 1$$





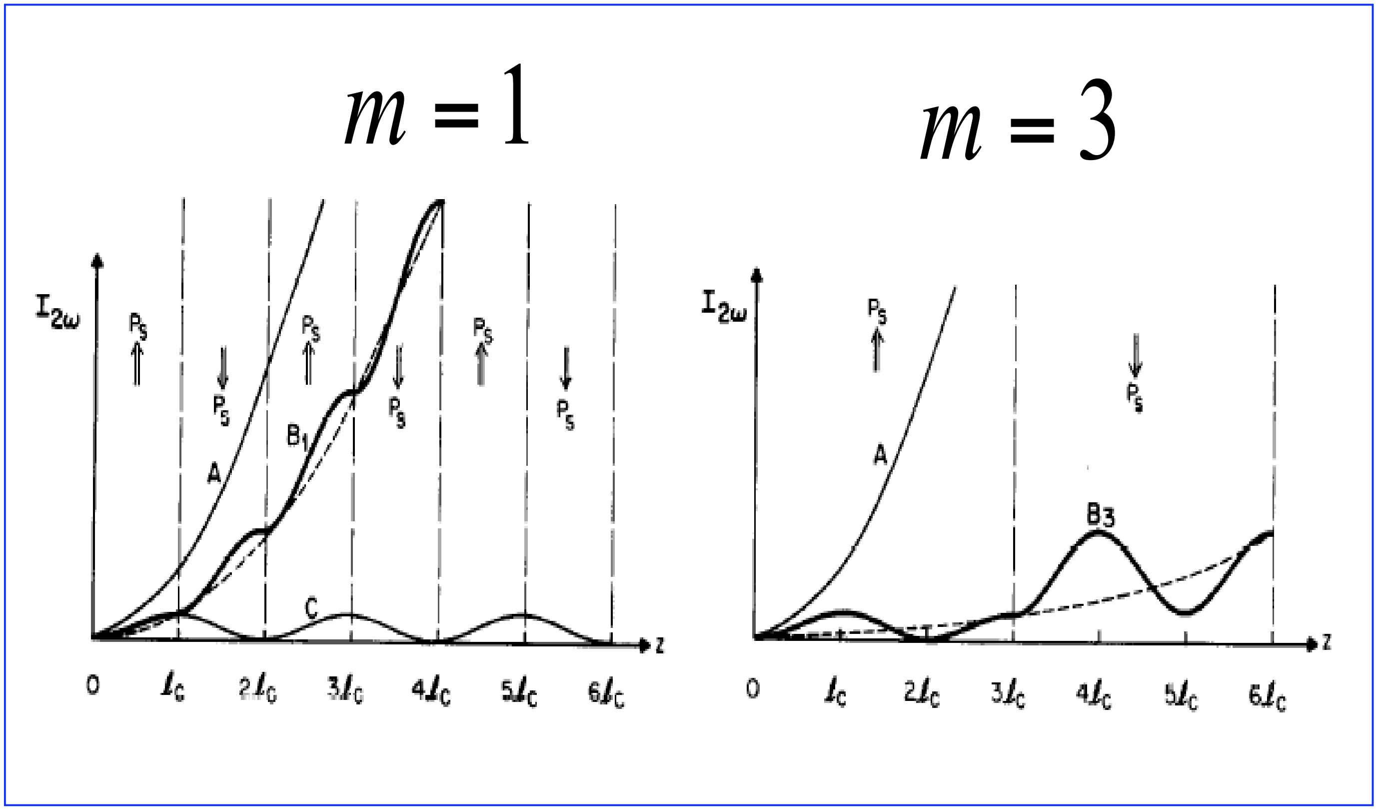
Periodically poled materials

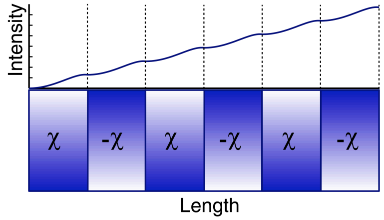
*Armstrong et al., PRA **127**, 1918 (1962); Fejer et al.,JQE **28**, 2631 (1992); Opt. Lett. **22**,1834 (1997)*

Quasi-Phase-Matching can adjust for phase mis-match:

- Traditional Quasi-Phase-Matching, $\Delta k = K_m = \dfrac{2\pi m}{\Lambda}$

Λ= Periodicity of nonlinear medium

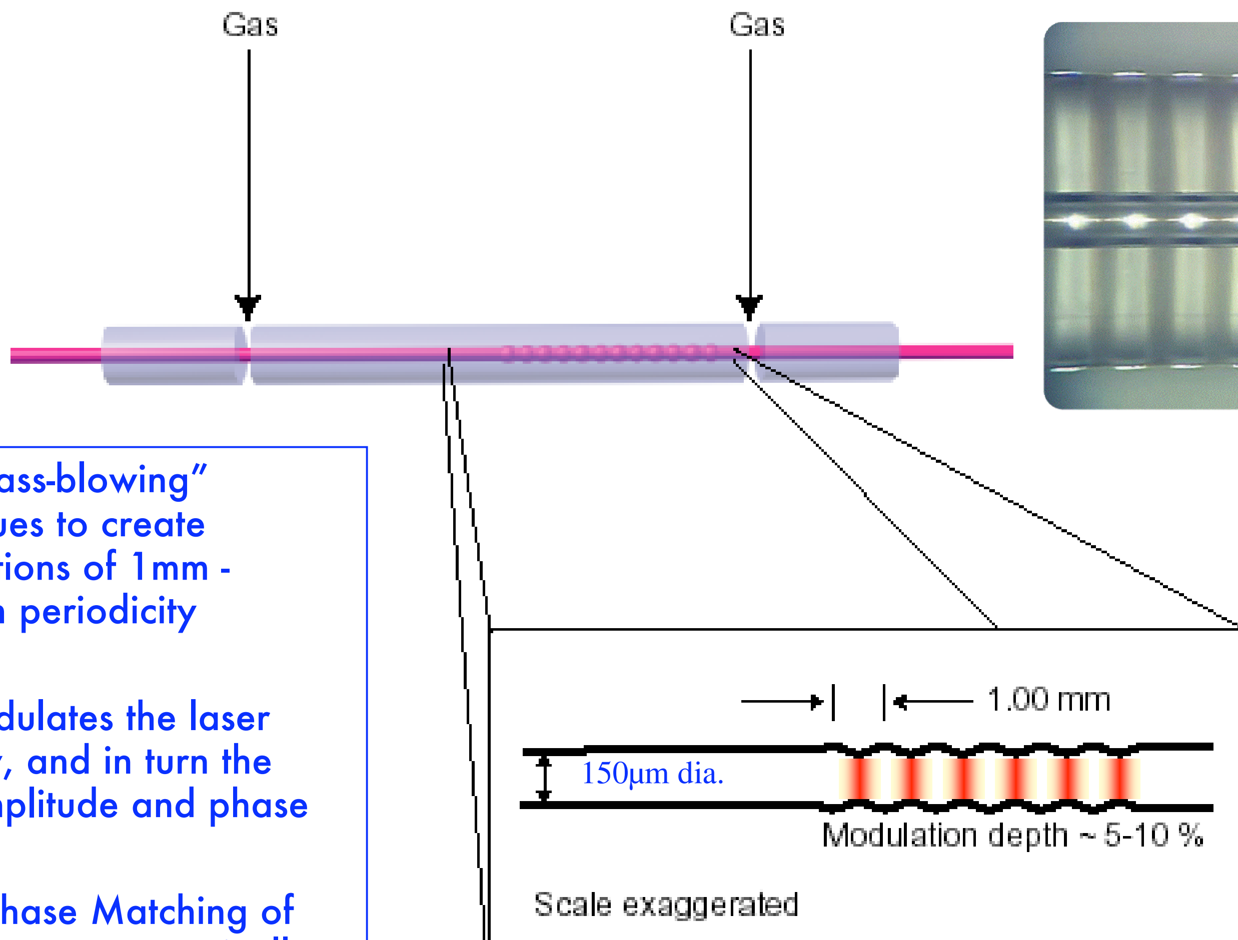


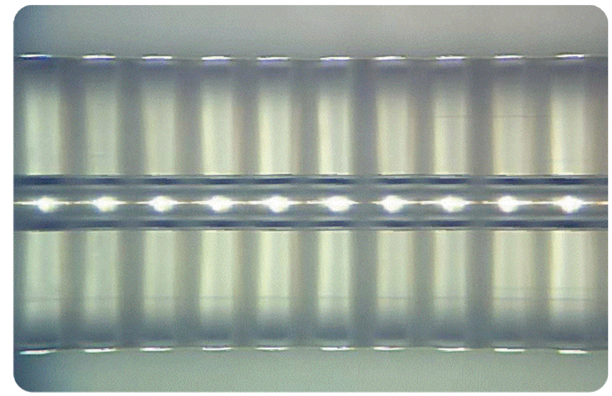


Periodically poled materials

*Armstrong et al., PRA **127**, 1918 (1962); Fejer et al.,JQE **28**, 2631 (1992); Opt. Lett. **22**,1834 (1997)*

EUV Photonics: modulated fibers

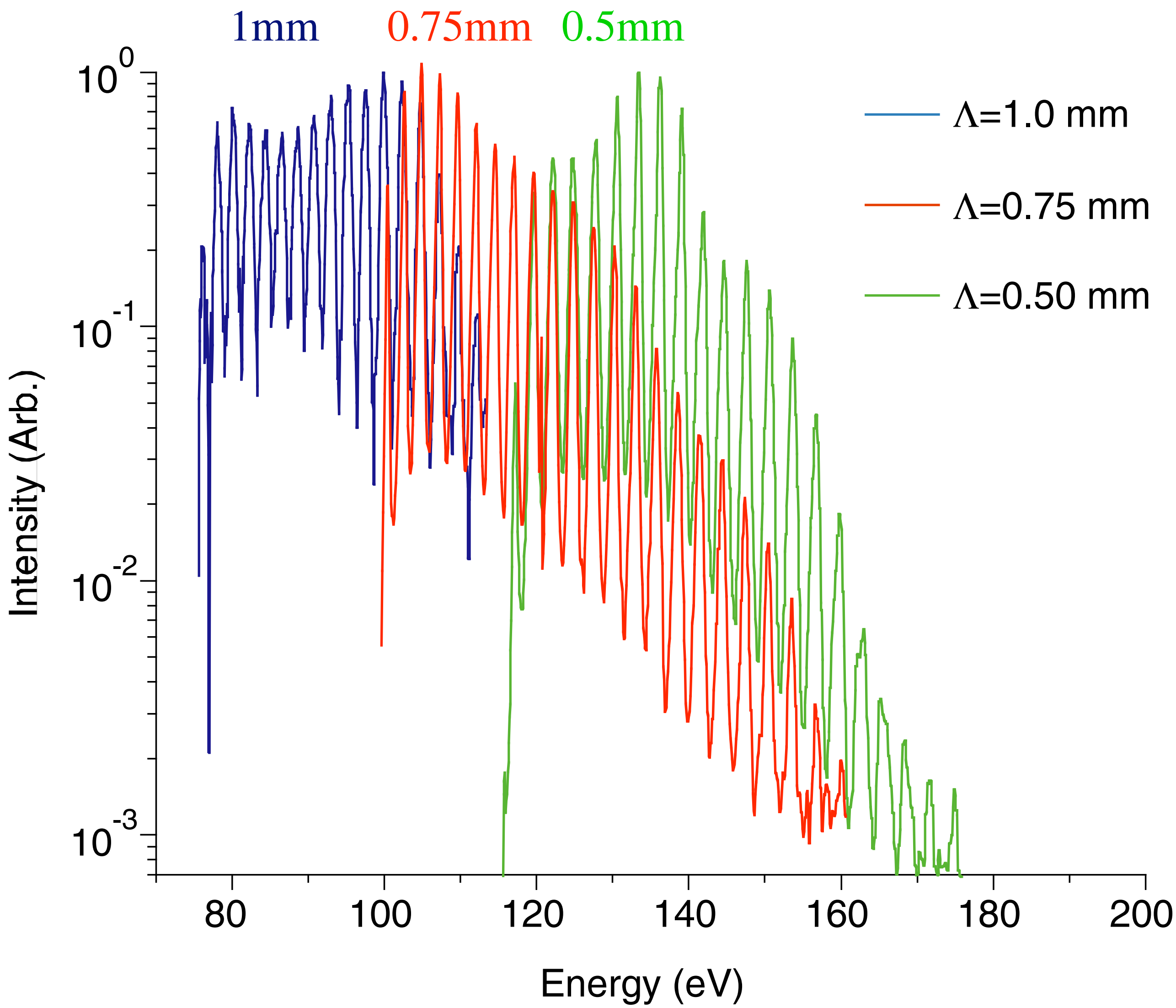




- Use "glass-blowing" techniques to create modulations of 1mm - 0.25mm periodicity
- This modulates the laser intensity, and in turn the EUV amplitude and phase
- Quasi Phase Matching of HHG occurs automatically by suppressing emission from regions that are out of phase

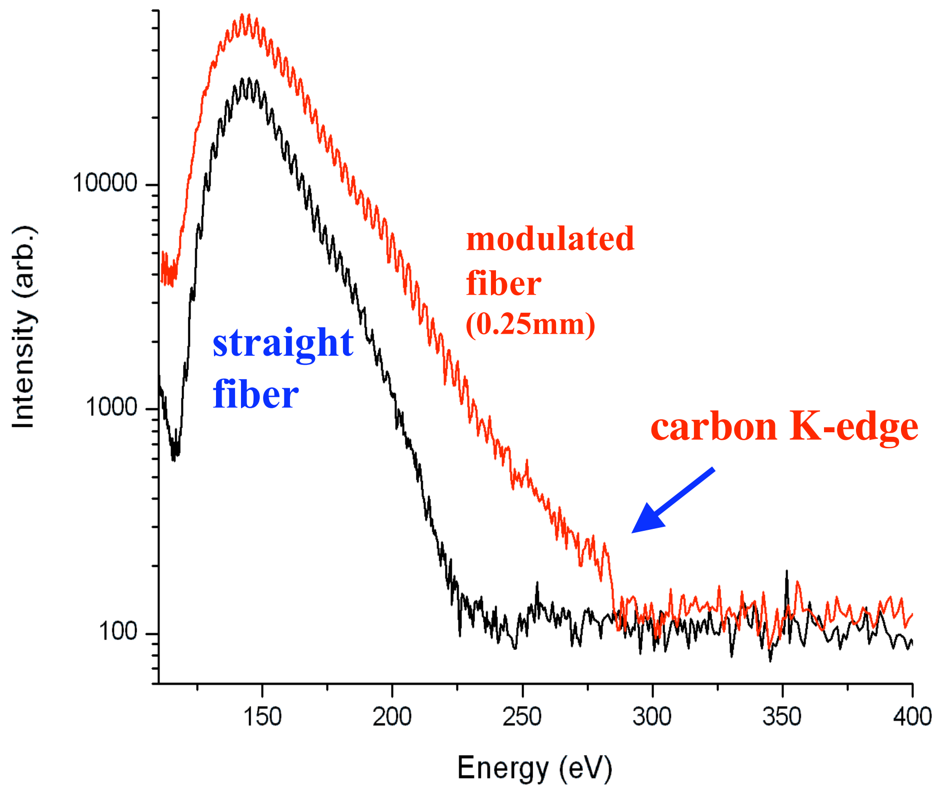
– A. Paul et al, *Nature* **421**, 51-54 (2003)

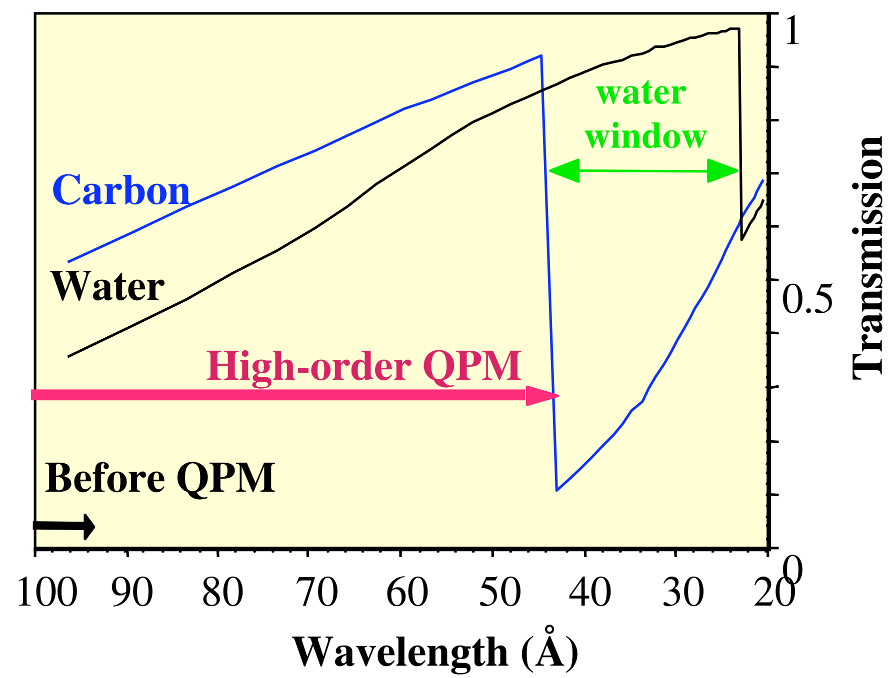
Shorter modulation periods give 100eV higher energy!



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

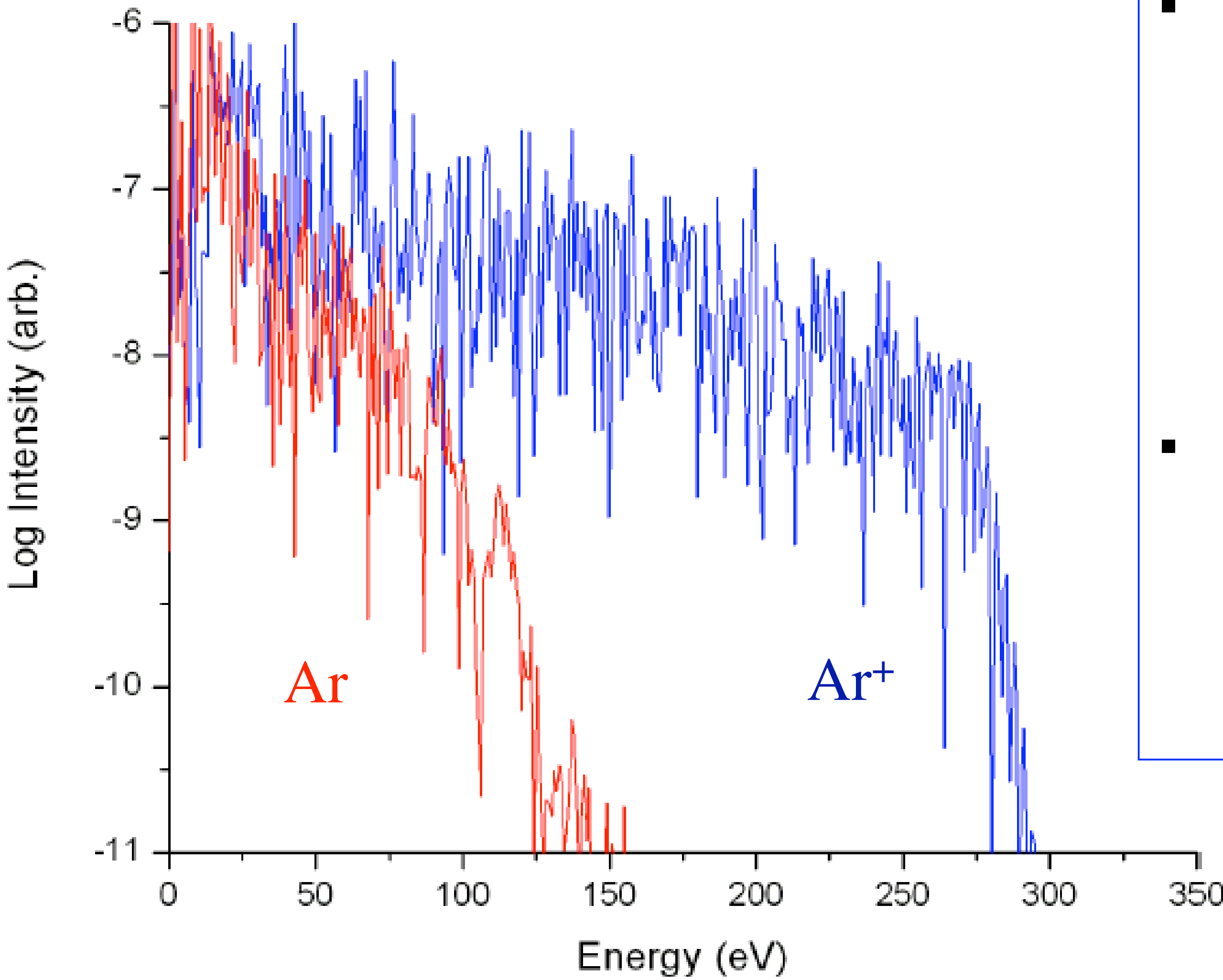
Quasi phase matched HHG in a fully-ionized plasma





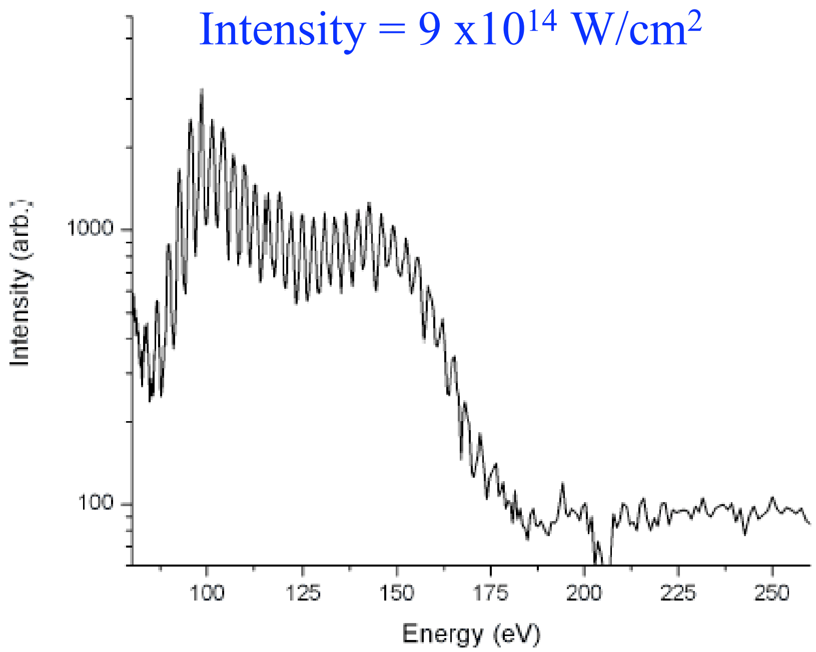
- Can create partial phase matching even when *fully* ionized (m=5 QPM)
  - E. Gibson et al., *Science* **302**, 95 (2003)
- Possibility to increase flux by orders of magnitude
- Photon energies up to keV using only modest (10x) increase in intensity

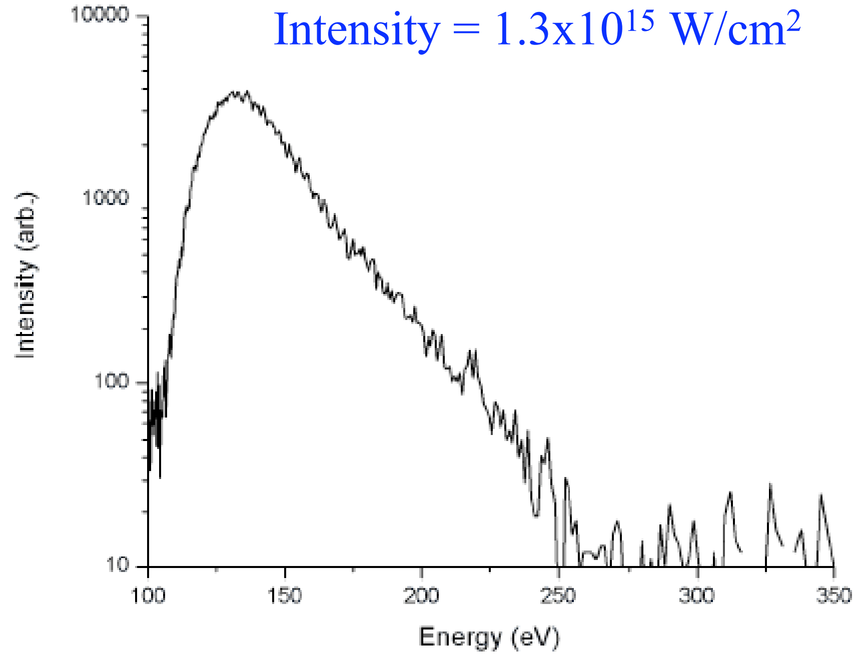
Can harmonic emission from ions extend photon energy?



- Quantum calculations (Christov) show that the HHG emission from Ar ions is as bright as that from atoms, and extends to higher energies
- However, observation of HHG from ions difficult– highest Ar harmonics observed <100eV

Waveguide counteracts plasma defocusing:

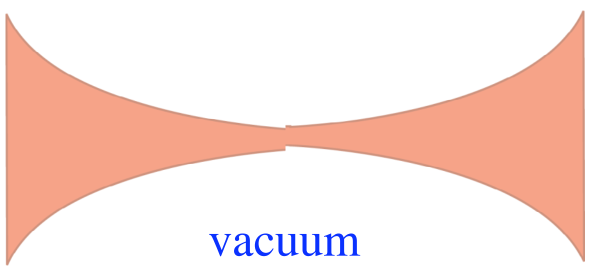


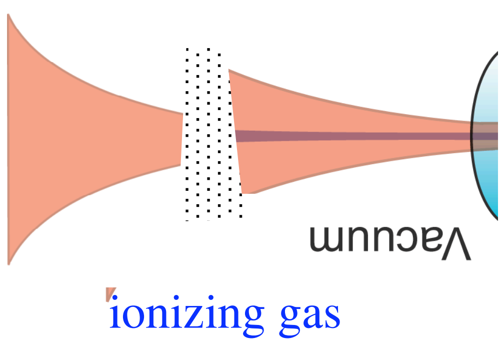


- At intensities of > 10^{15} Wcm^{-2}, extend HHG to 250eV in Ar
  - E. Gibson et al, PRL 92, 033001 (2004)
- HHG from ionization of Ar^{+} –> Ar^{++}
- Represents extension of cutoff by 100 harmonics– 150eV!

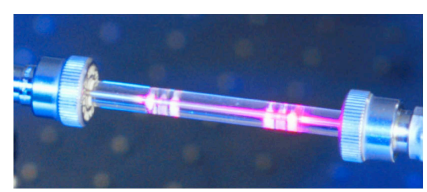
Why was it so difficult to observe HHG from ions?

Plasma-induced defocusing

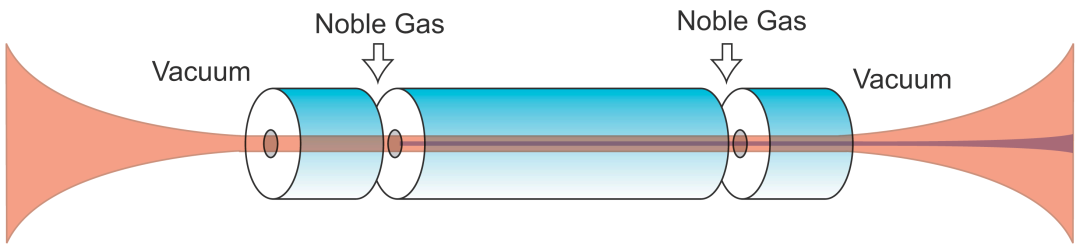


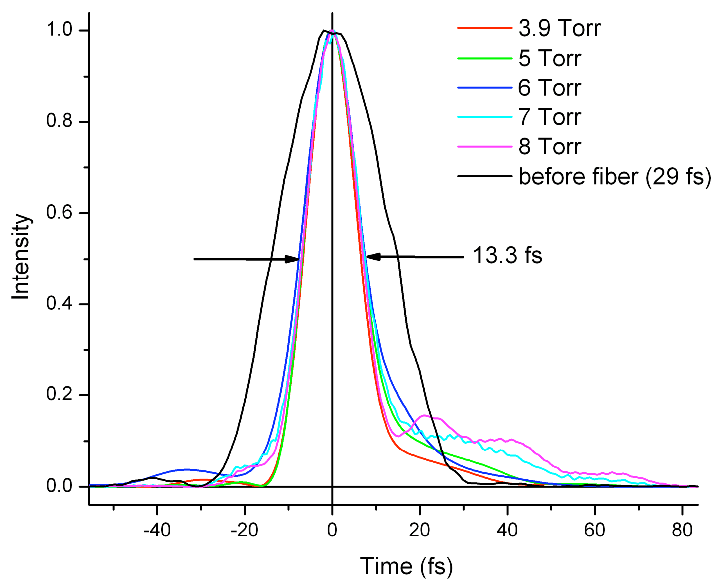


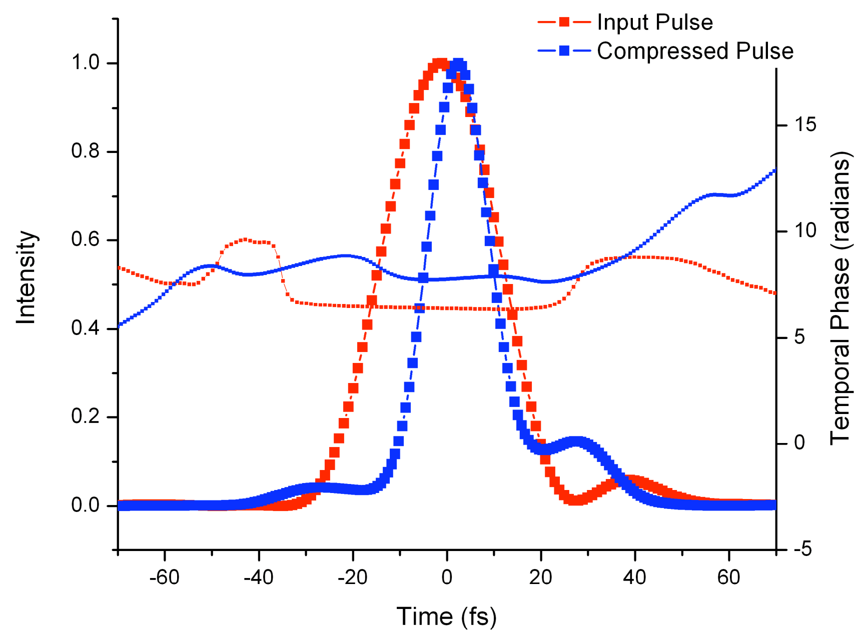
- For 7 Torr, 100% ionized gas, the defocusing length is 2.8mm, compared with confocal parameter of ≈ 1cm
- In a gas jet – limit to the intensity and ionization before plasma breaks up the pulse
  - Wahlström PRA 51, 585 (1995)
- Hollow waveguides guide the laser even with a plasma and maintains high intensity and good mode
- Emission from ions allows extension of harmonics to very high energies



New mechanism for pulse self-compression

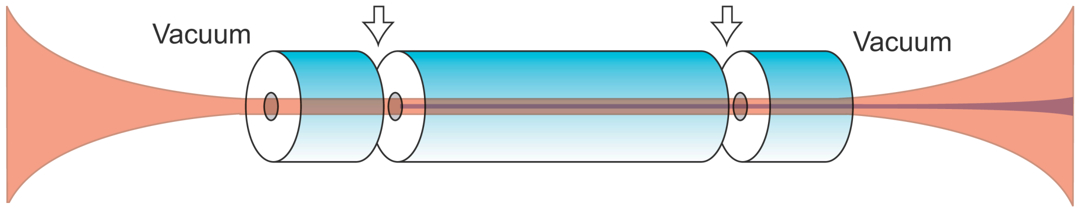


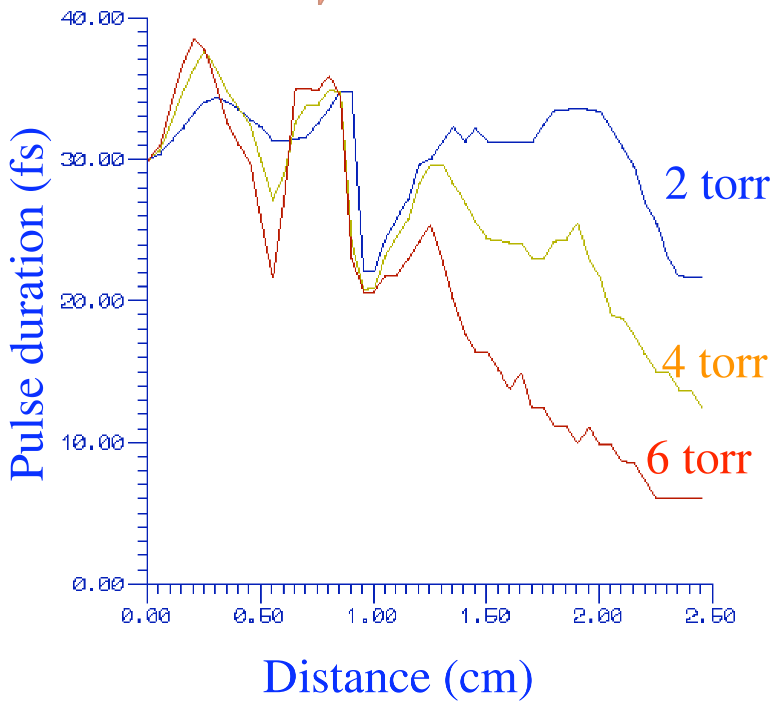


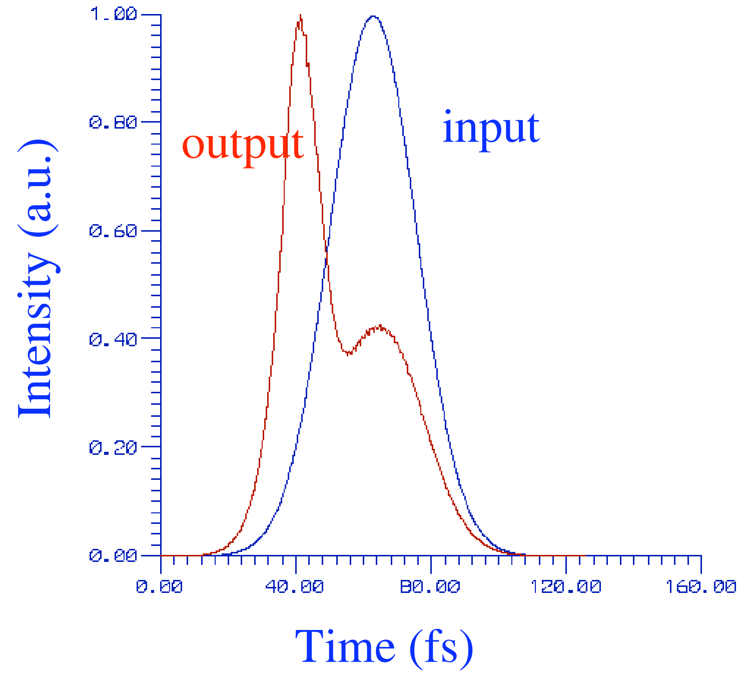


- Pulse emerges from fiber significantly shorter in time (13fs) compared with input (29fs) WITHOUT NEED FOR DISPERSIVE PULSE COMPRESSION

Theory of plasma-waveguide self compression

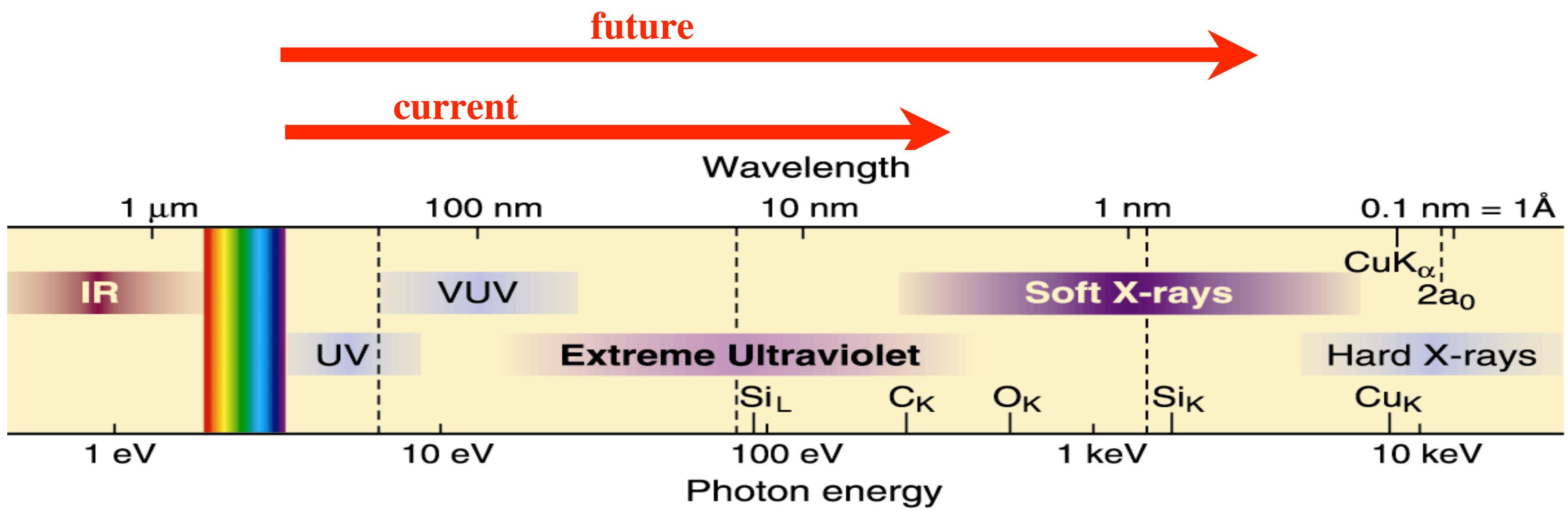






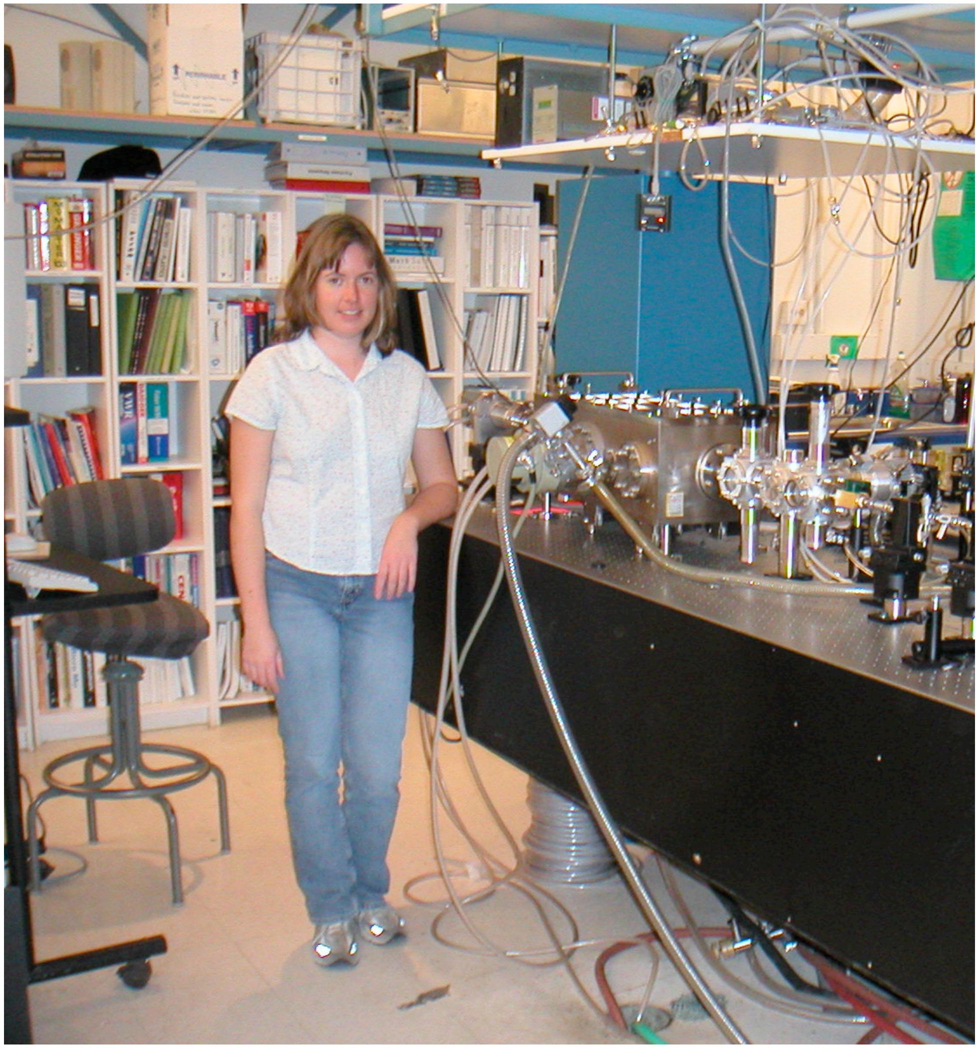
- Theory shows that the temporal compression is due to a spatio-temporal reshaping effect as the pulse refracts from the plasma and is guided in the waveguide.
- Effect much weaker in free-focus jet

Potential range of compact coherent x-ray sources

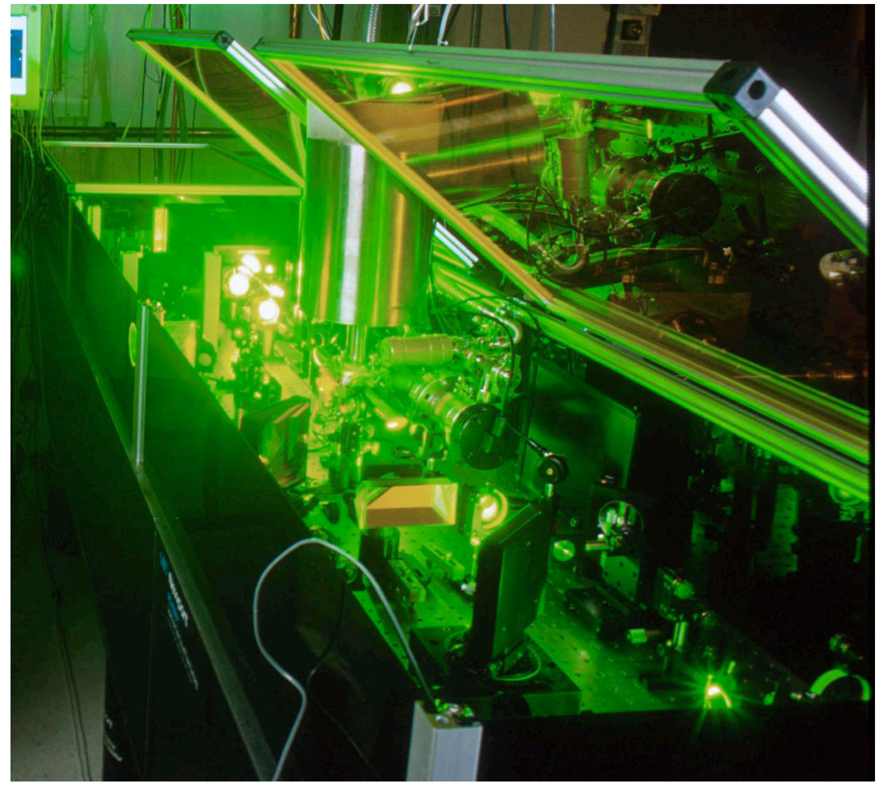


- Photon energy scales linearly with the laser intensity
- 10^{16} Wcm^{-2}, should generate multi-keV harmonics from ions
- Need QPM techniques to enhance these harmonics
- Guided geometry will still work

Experimental set-up

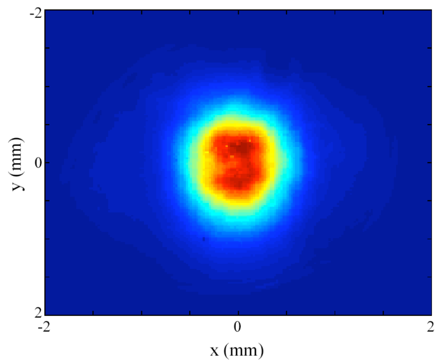


Emily Gibson

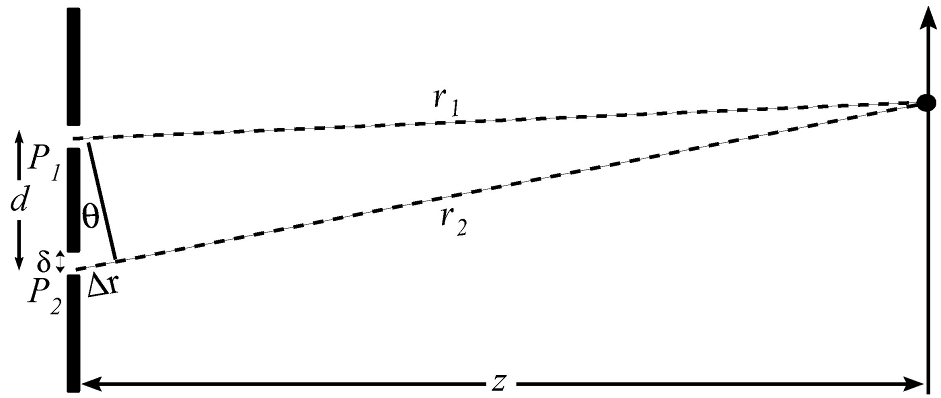


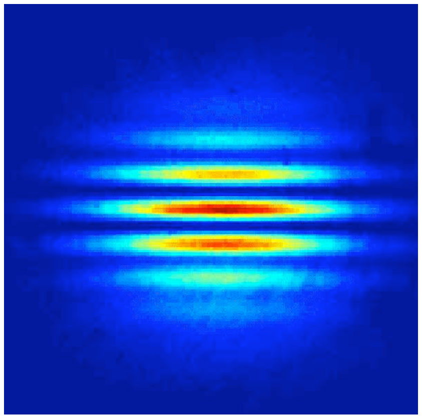
Single-stage, cryo-cooled, 7W multi-kHz laser

Coherent imaging using high-harmonic radiation



EUV beam





EUV interference pattern

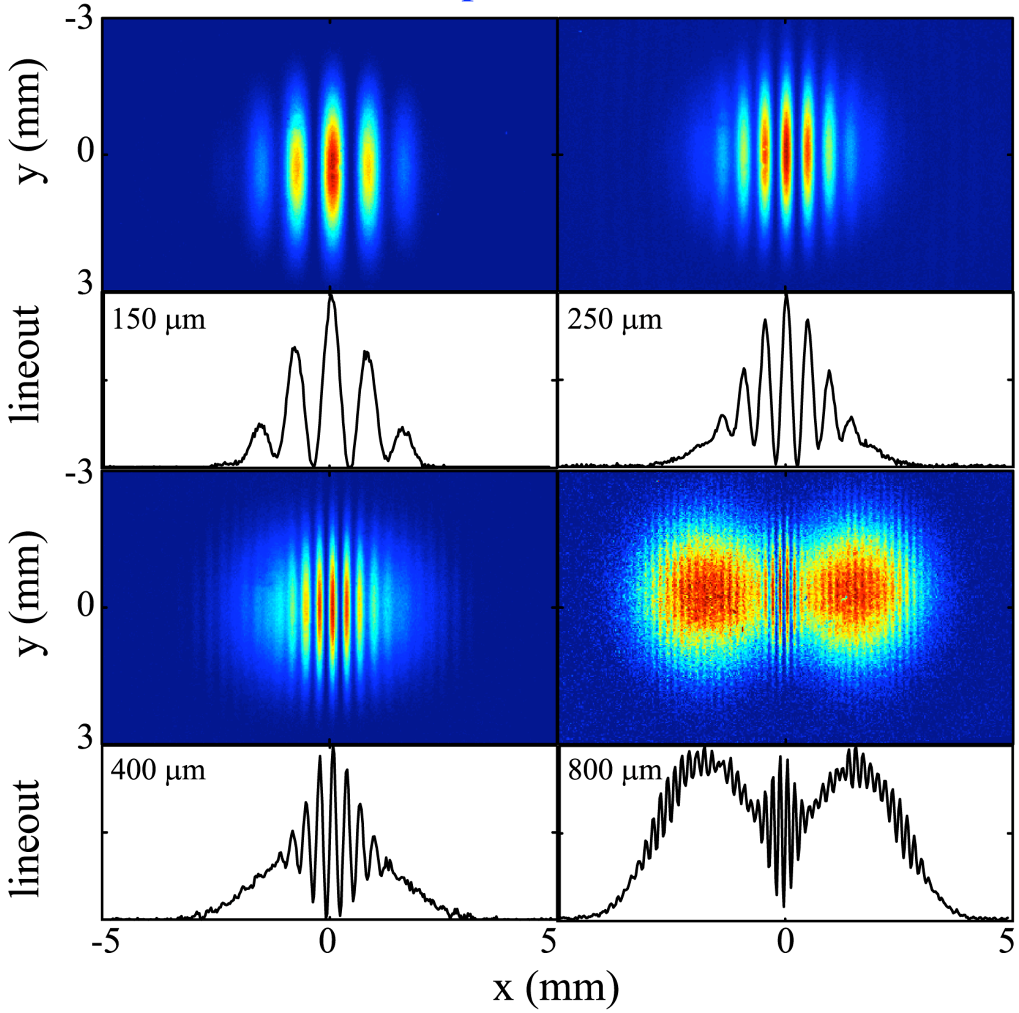
- Repeat 200 year old experiment – Young's Double Slit
  - Young, Philos. Trans. R. Soc. XCII 12, 387 (1802).
  - E. Wolf et al., JOSA 46, 895 (1957; Opt. Lett. 6, 168 (1981).

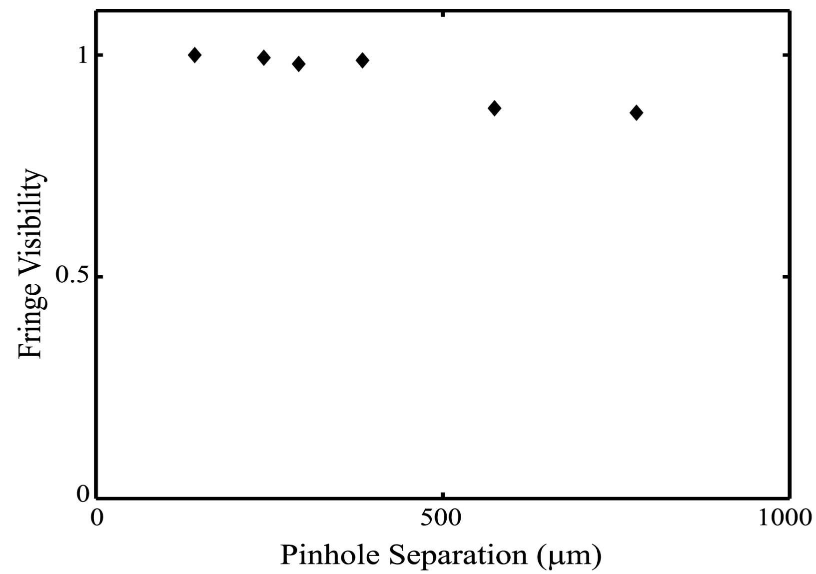
NIST-CU

Nearly perfect spatial coherence in the EUV

EUV
An NSF Engineering Research Center

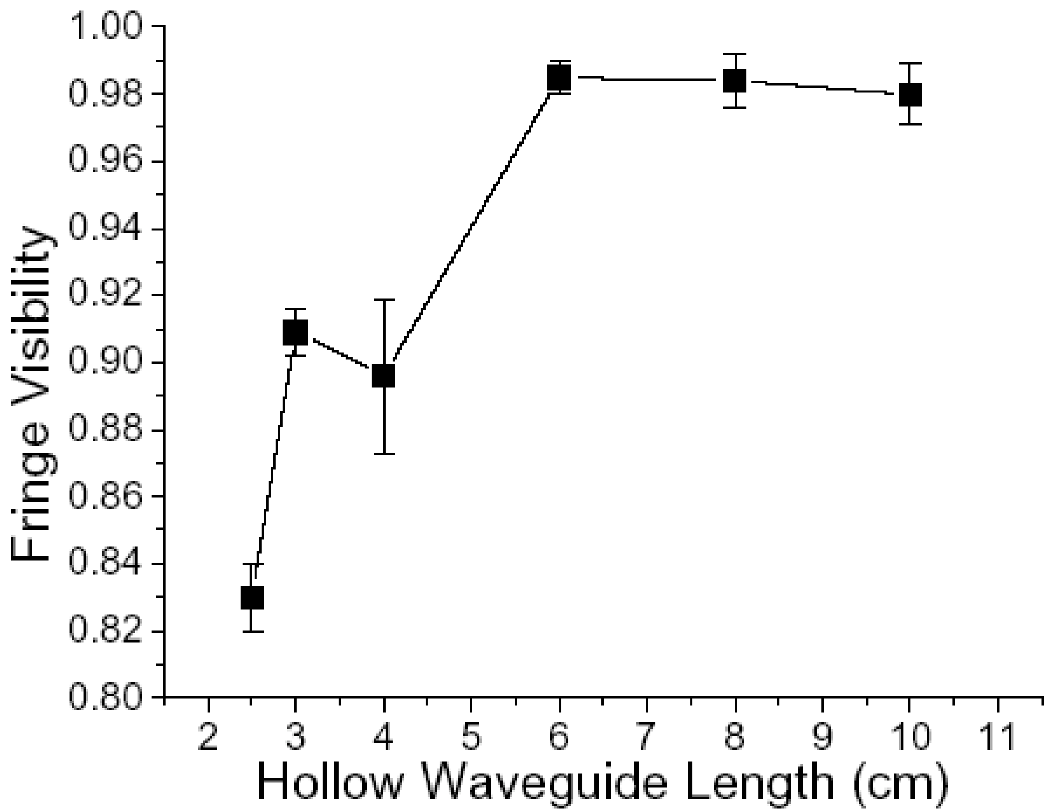
Double-slit diffraction patterns from 1mm EUV beam

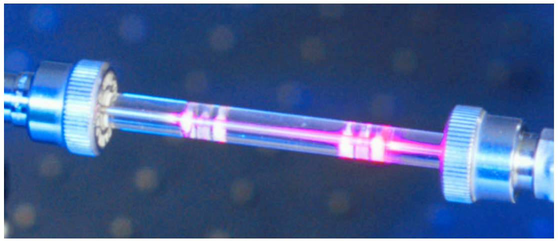




- R. Bartels et al, Science 297, 376 (2002)
- R. Bartels et al, Opt. Lett. 27, 707 (2002)

"Laser-like" beams only for long lengths



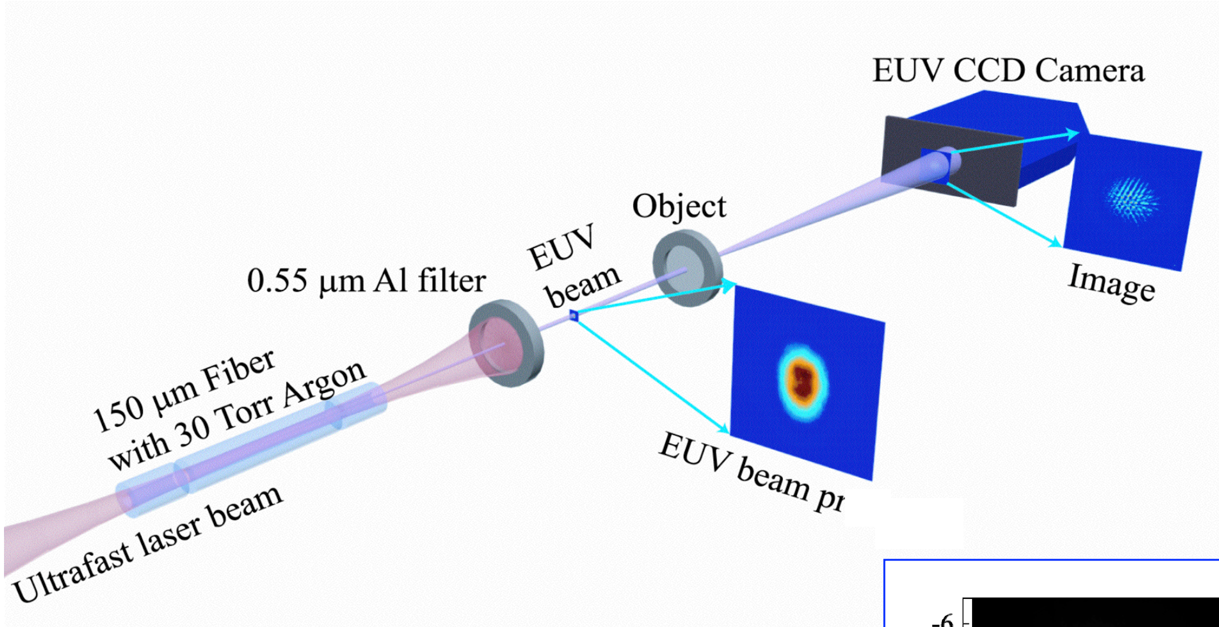


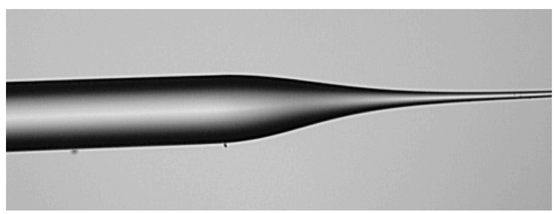
Fiber length

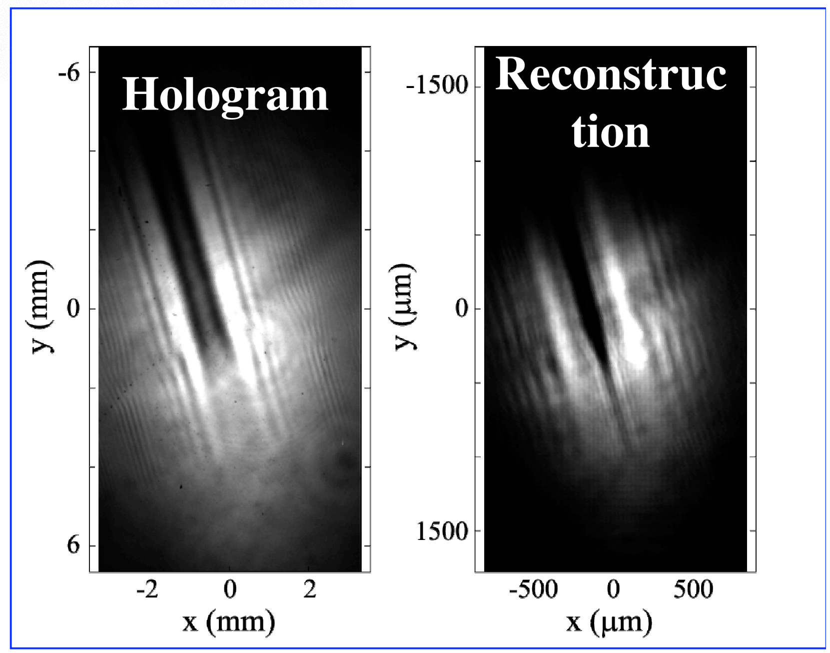
- A. Libertun et al, Applied Physics Letters, TBP (2004)

- "Laser-like" beams only for long interaction lengths in fibers
  - Long lengths are needed to form a guided mode with a flat phase
- Spatial coherence much lower (≈ 50%) from gas jets
- Rich physics in guided beam propagation in plasmas

Simple EUV Gabor holography of static objects

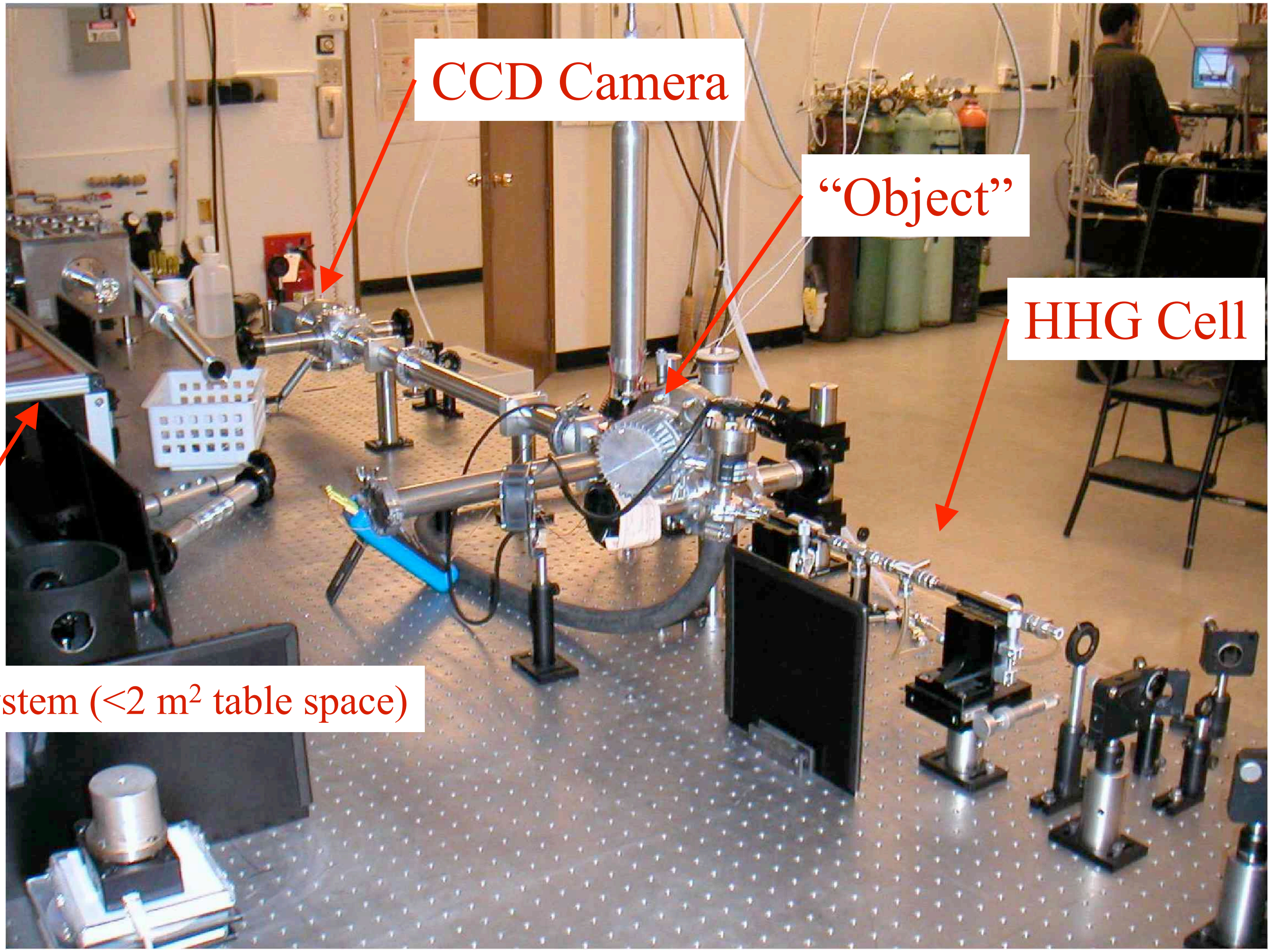




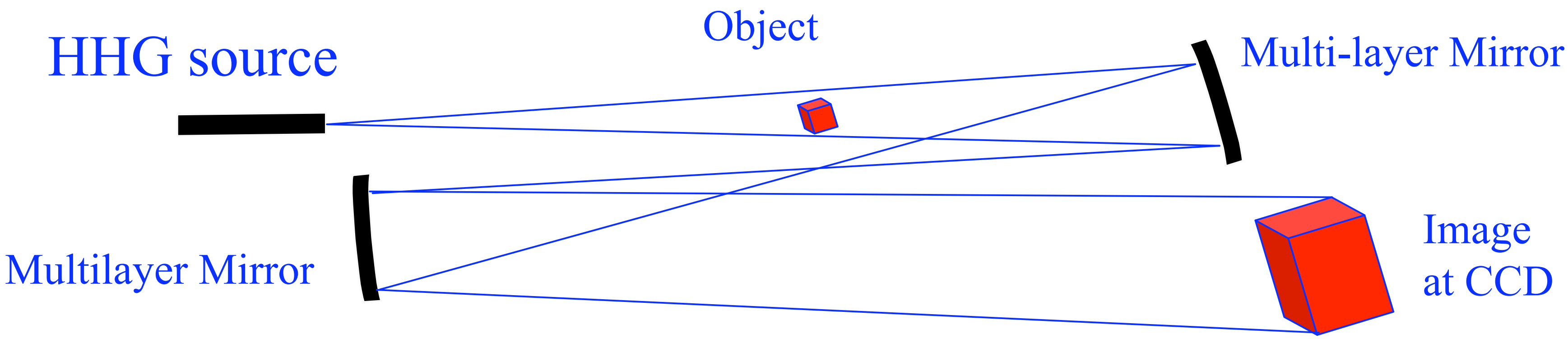


- Bartels et al, Science 297, 376 (2002)
- Bartels et al, Opt. Lett. 27, 707 (2002)

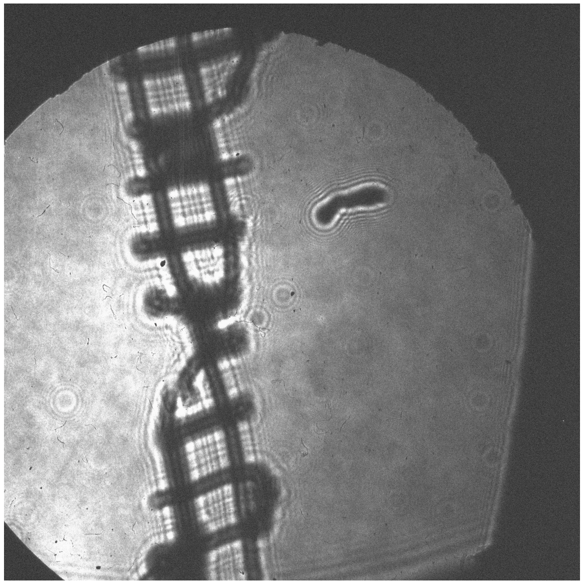
Experimental EUV beamline:



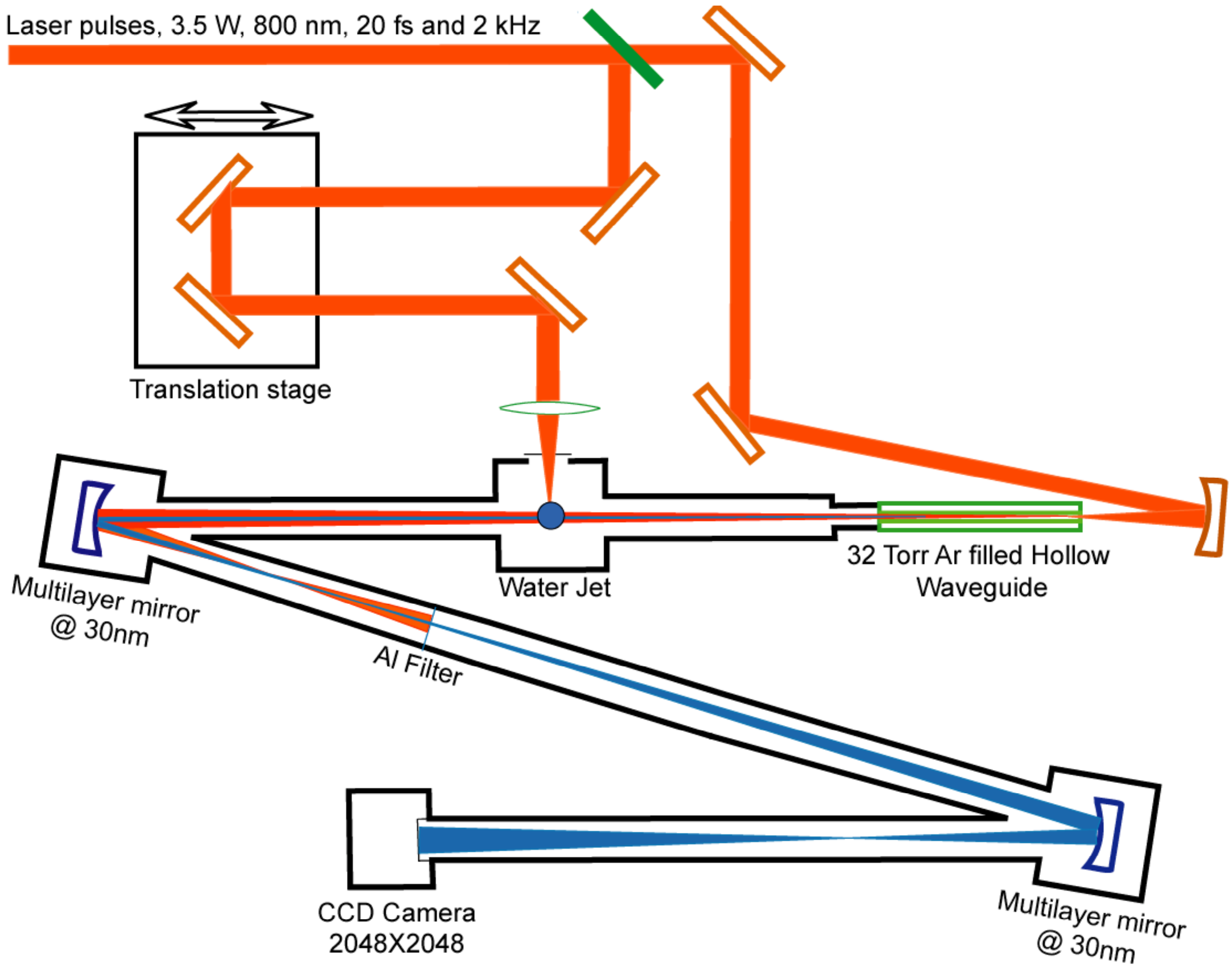
Large working-distance magnification system



- 20x magnification
- Resolution ~1 micron
- Image out-of-focus to show fringes

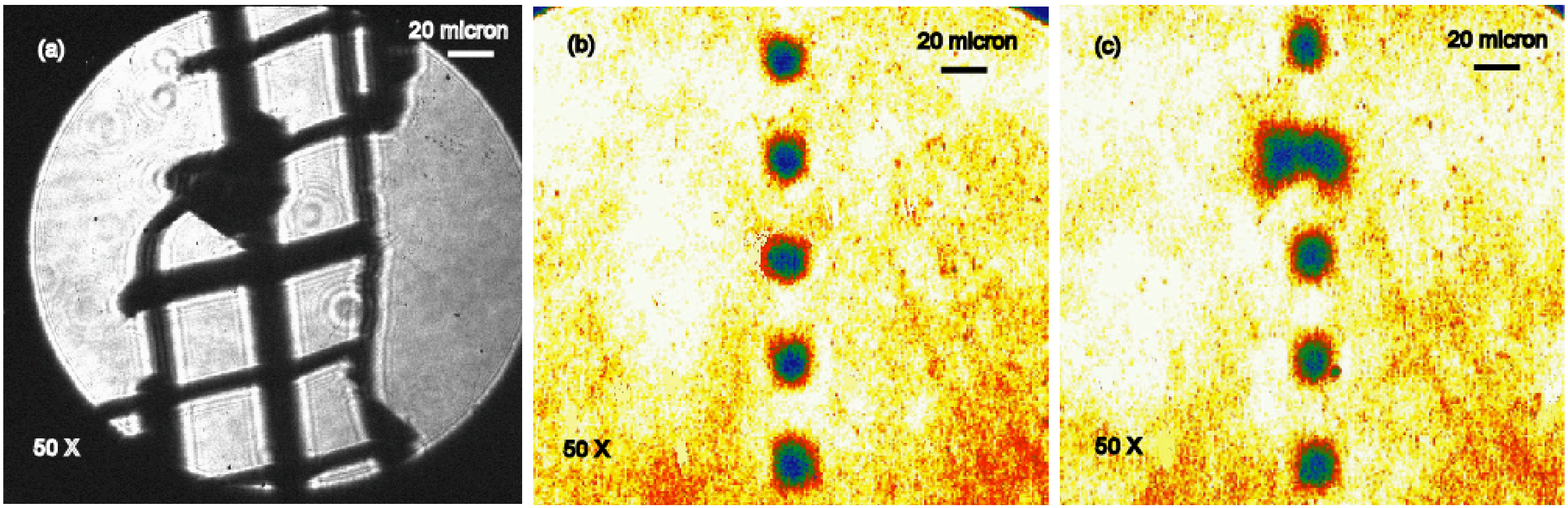


Time-resolved stroboscopic imaging



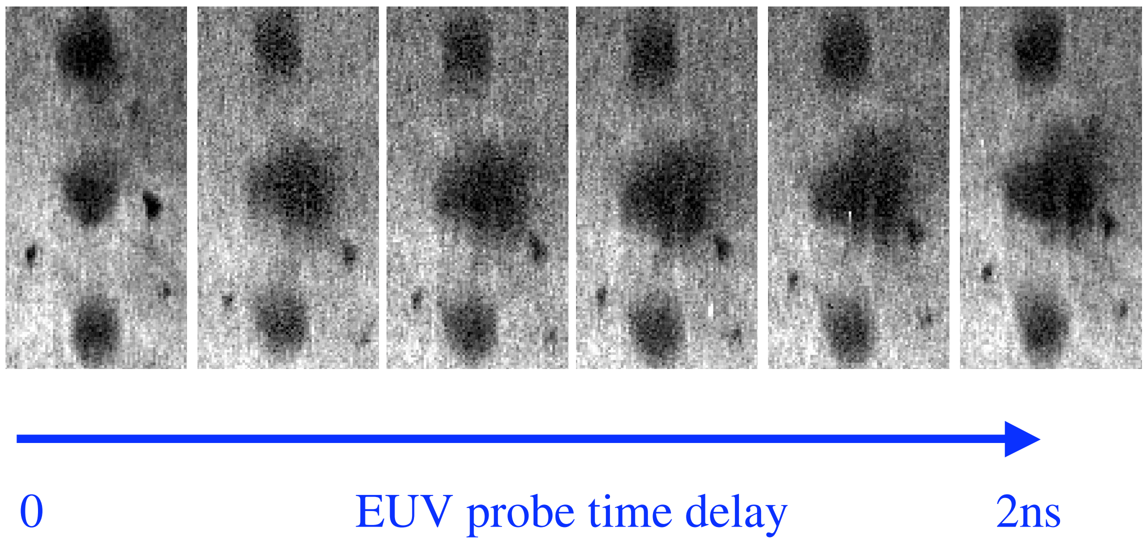
Dynamic Imaging

- 50x magnification
- 3 minute exposure @ 2 kHz



Static mesh | **Static droplet** | **"Perturbed" droplet**

Time-resolved dynamics imaged using HHG:

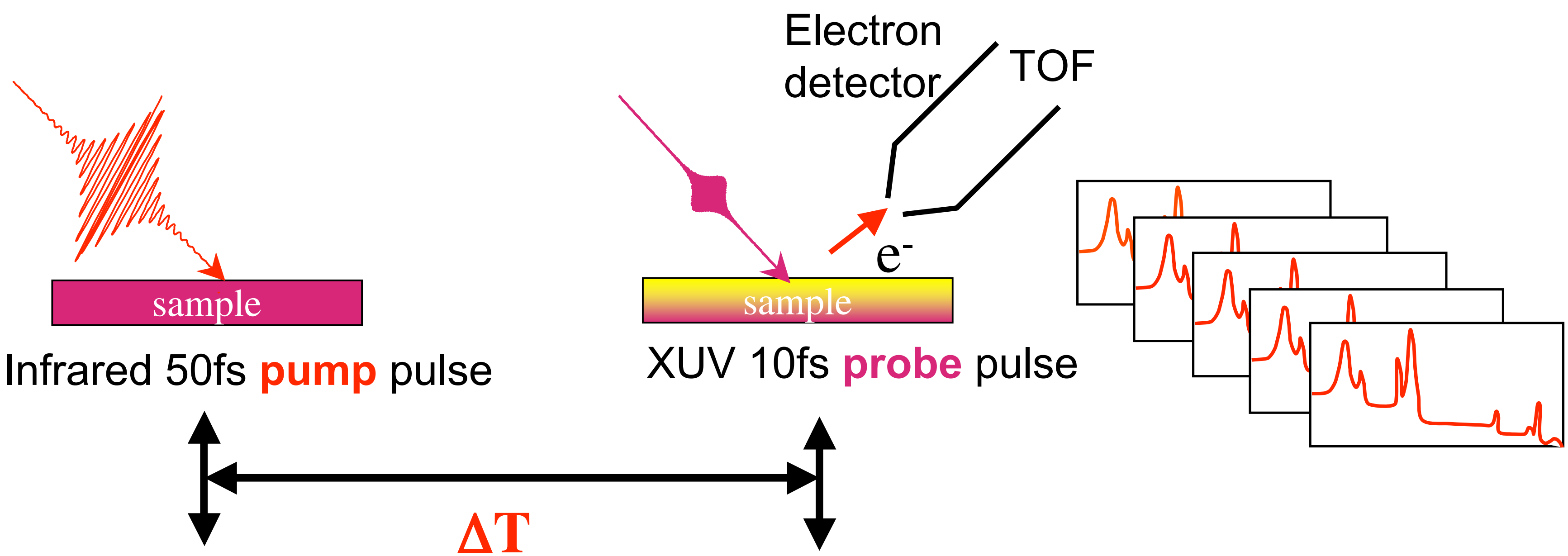


Scientific Possibilities

- Detailed data on light coupling into high-density plasmas
- "Warm dense matter" studies
- Shocks
  - Expansion into a background gas
- Underdense plasma beam propagation
  - Vaporizing prepulse
- Colliding plasmas
  - Illuminate two drops
- Droplet source of interest for EUV lithography LPP sources
- Generation of keV harmonics to study thicker samples

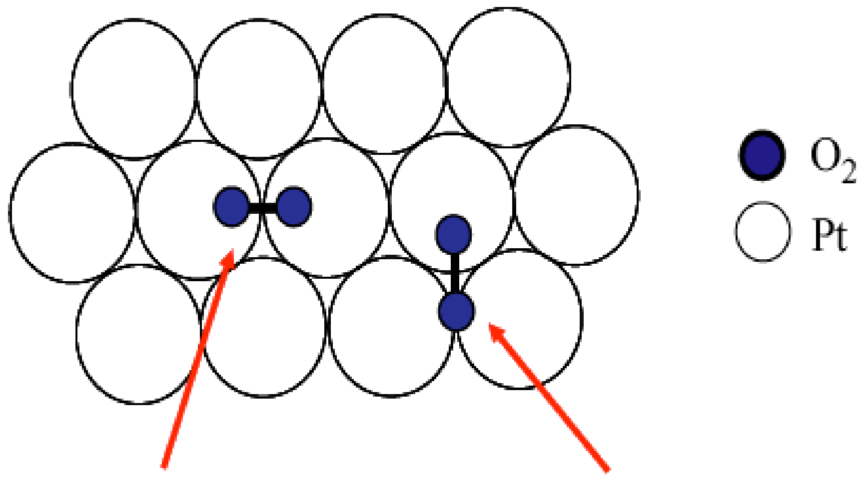
Using fast EUV light pulses to observe surface reactions

Time-Resolved Photoemission



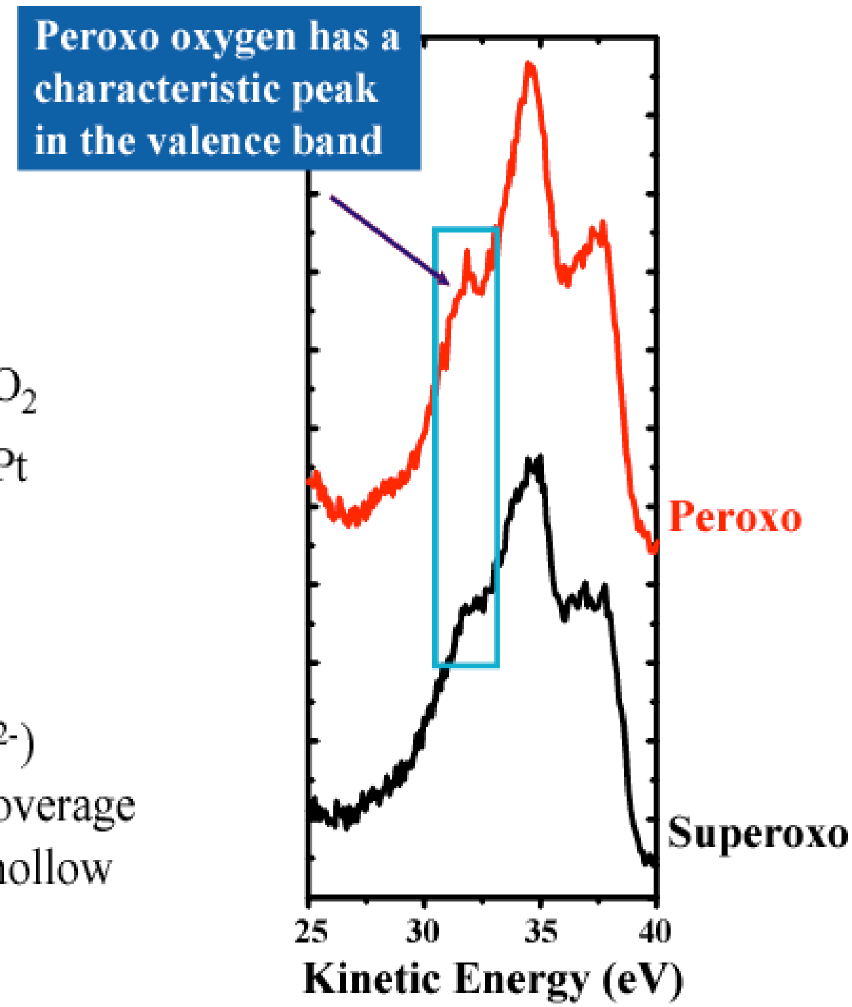
Use ultrafast EUV pulses to study molecular dynamics

O_2 adsorbed on Pt(111) at –197°C,



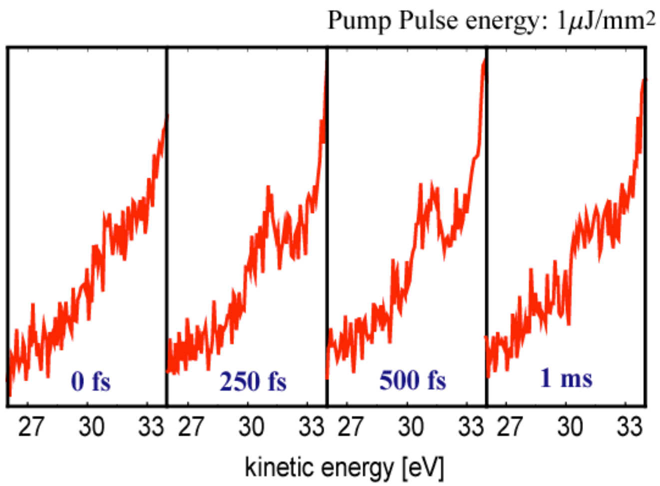
Superoxo-like (O_2^-)
dominates at high O_2 coverage
resides in bridge site

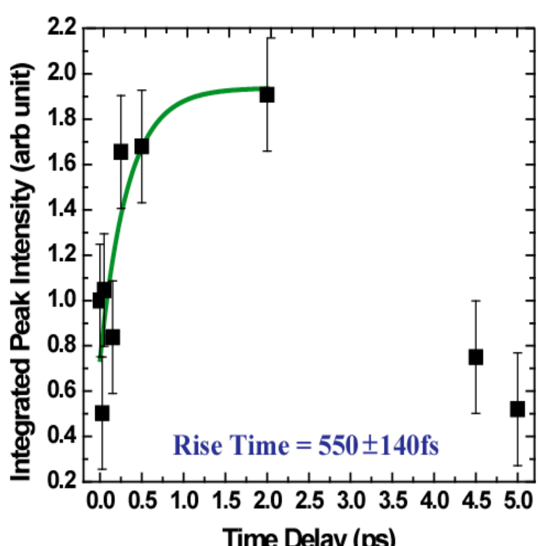
Peroxo-like (O_2^{2-})
dominates at low O_2 coverage
resides in three fold hollow

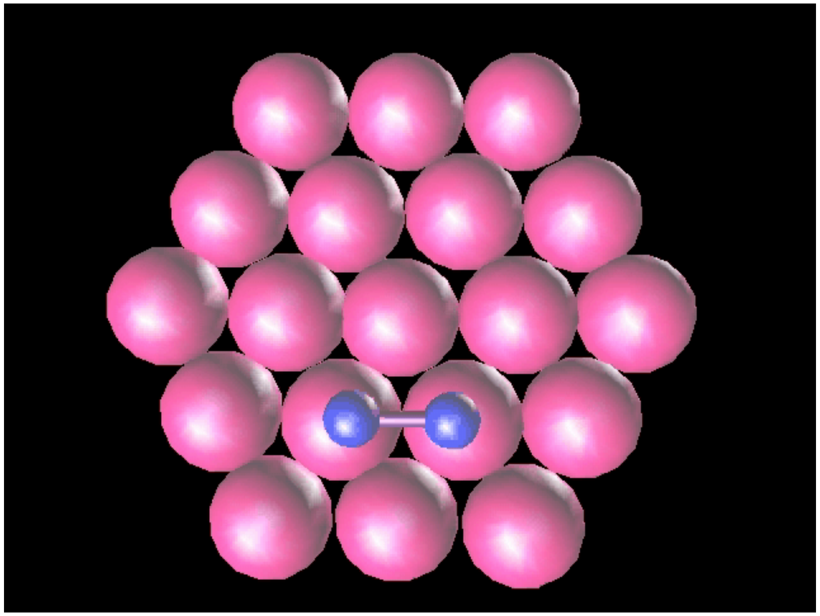


Ho, Mazur, Haffner

Time-resolved photoemission: O_2 rotation on Pt



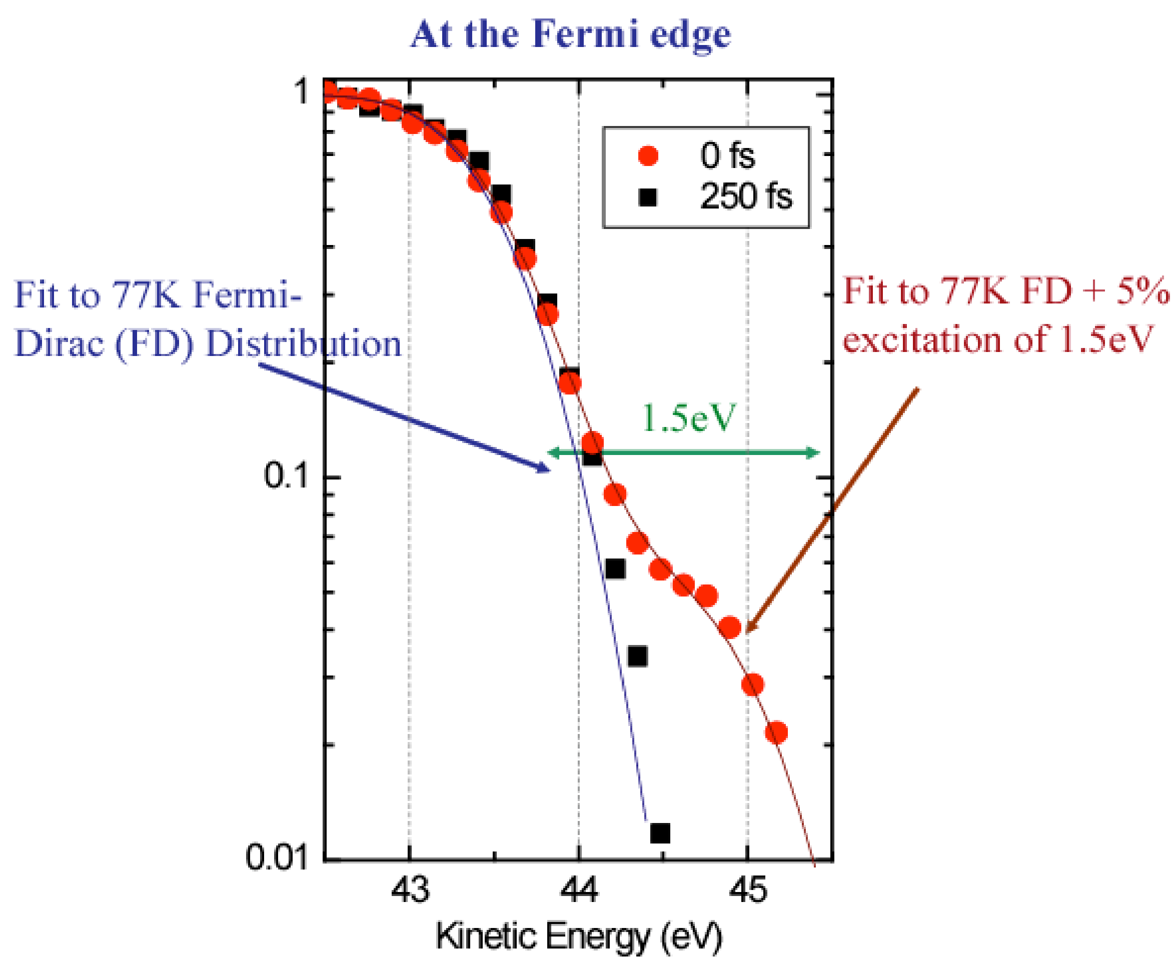




PRL 87, 25501 (2001)

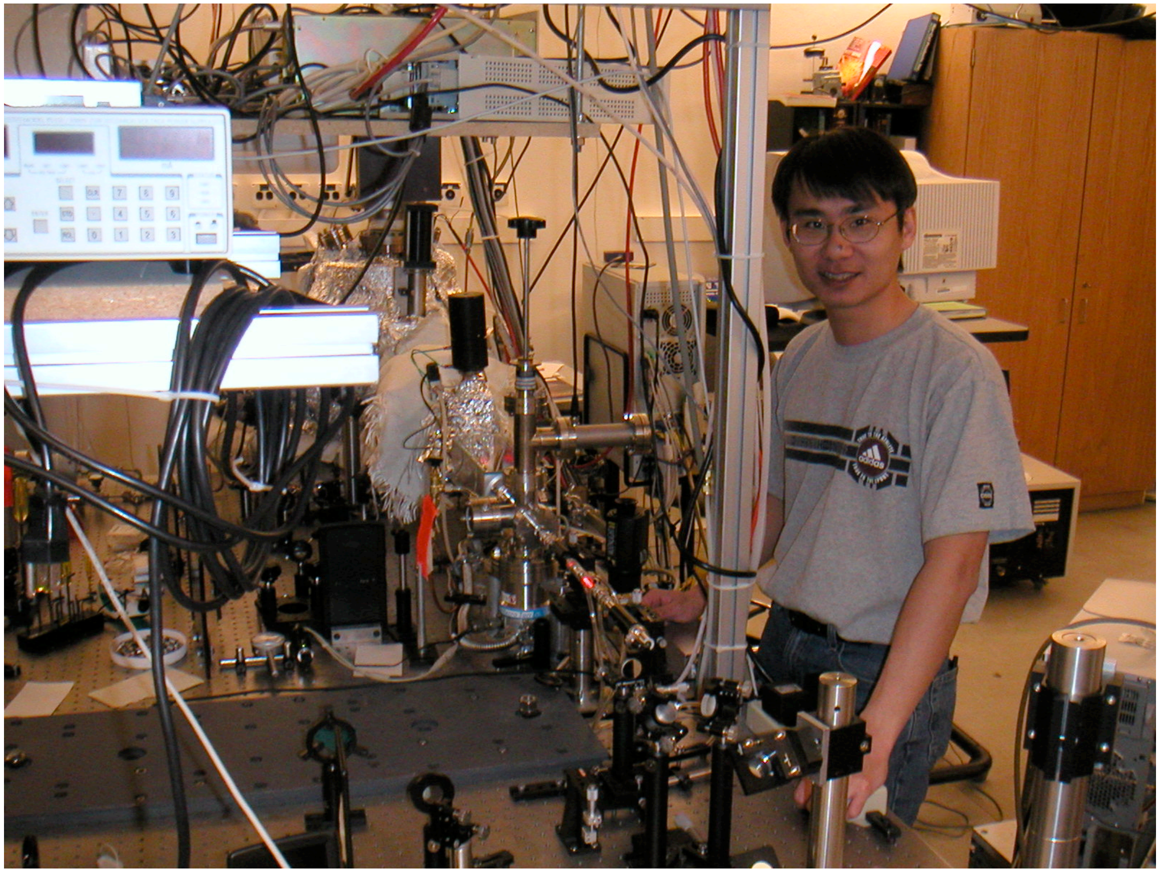
Hot electron mediated surface charge transfer process on 100-500fs timescales

Electrons are heated rapidly and cool rapidly within 250fs



Experimental Setup

EUV
An NSF Engineering Research Center



Tim Lei

Conclusions

- Ultrafast EUV "Photonics" makes possible versatile new light sources -
  - Laser-like beams at photon energies up to ≈ keV
  - Unprecedented time resolution, coherence, size
- Basic feasibility of stroboscopic imaging of small-scale plasmas demonstrated
- Novel linear and nonlinear spectroscopies using EUV light
  - Site-specific information about atoms and molecules
  - Spectroscopy with simultaneously high spatial and temporal resolution

- Gratefully acknowledge funding from
  - NSF
  - DOE BES
  - NNSA