Extreme Ultraviolet Frequency Combs: Principles and Applications: *Part 1*

Thomas K. Allison
Stony Brook University

ICTP Winter College on Optics
Feb. 18, 2016

Long Island

Stony Brook University

BROOKHAVEN
NATIONAL LABORATORY

Four new AMO/Chemical Physics Faculty!

- Frequency combs
- Molecular dynamics
- Strong Fields/Control
- Quantum optics
- Laser cooling
-BEC/Many body QM
-Super-resolution microscopy
-Cold molecular ions

More information at:
http://allisongroup.physics.sunysb.edu

Outline

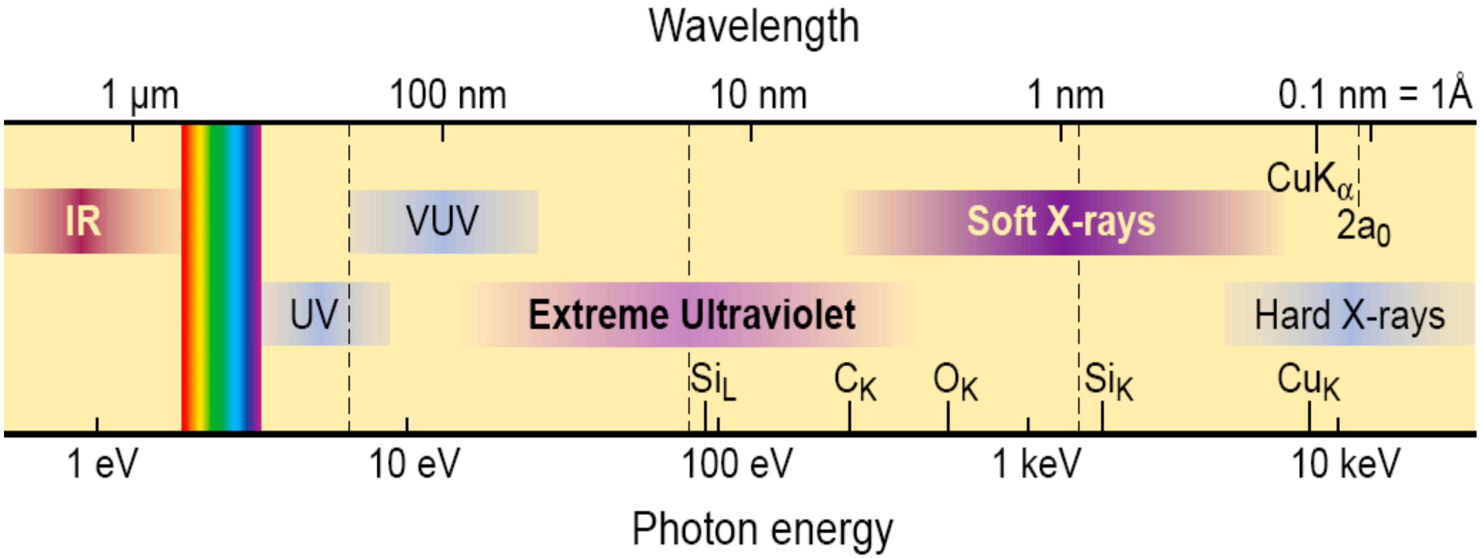
Part 1

- Why make an extreme ultraviolet frequency comb?
- Why is this hard?
- Basic physics of high-order harmonic generation.

Part 2

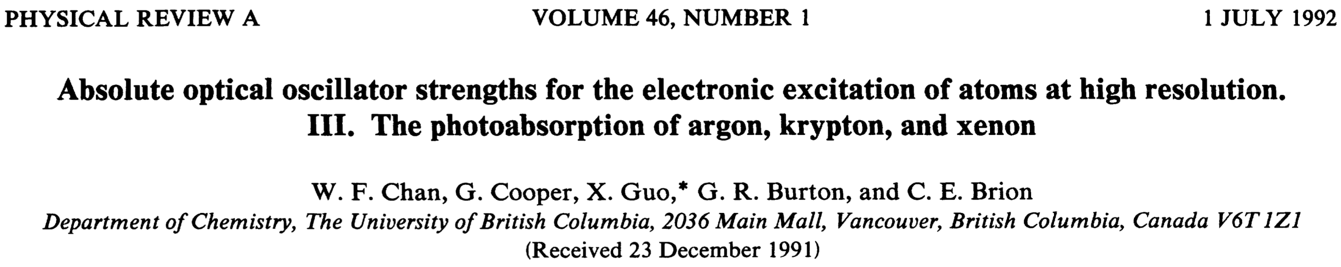
- High resolution spectroscopy with short pulses.
- The "Ramsey-Comb" Approach
- Cavity-Enhanced high-order harmonic generation
- Important results and future developments

The Extreme Ultraviolet (XUV)



- Atoms and molecules contain the bulk of their oscillator strength in the XUV
- All materials are strongly absorbing ⇒ Optics are challenging.
- But… a lot of rich physics in this regime where the light-matter interaction is strong.

XUV Absorption

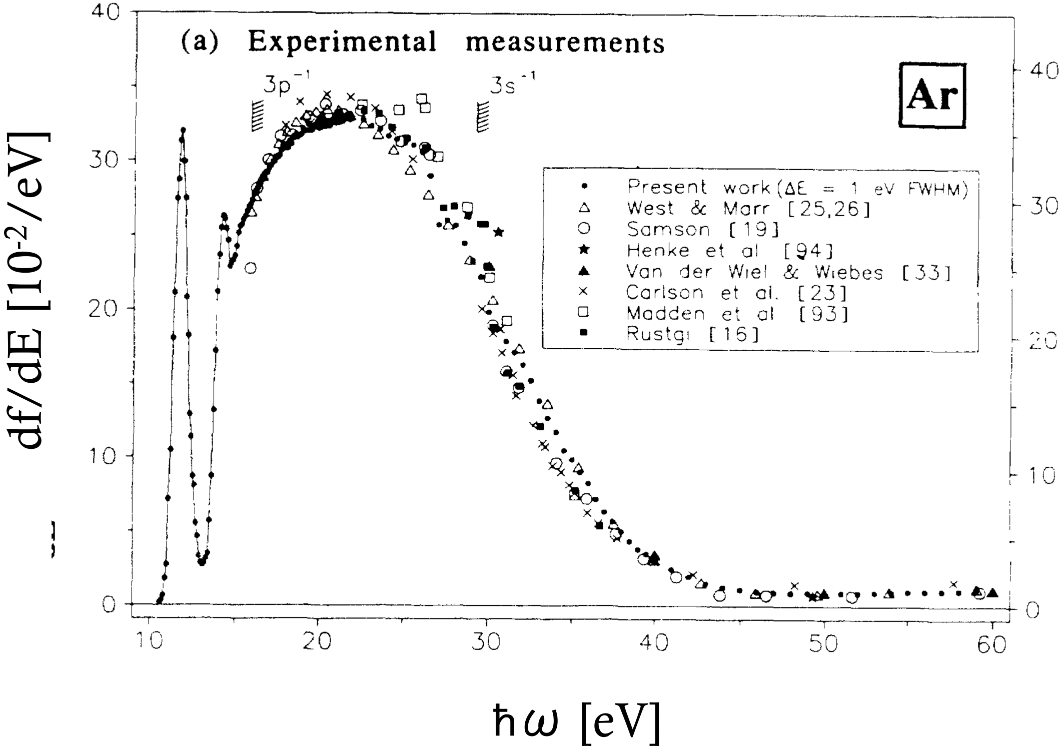


PHYSICAL REVIEW A VOLUME 46, NUMBER 1 1 JULY 1992

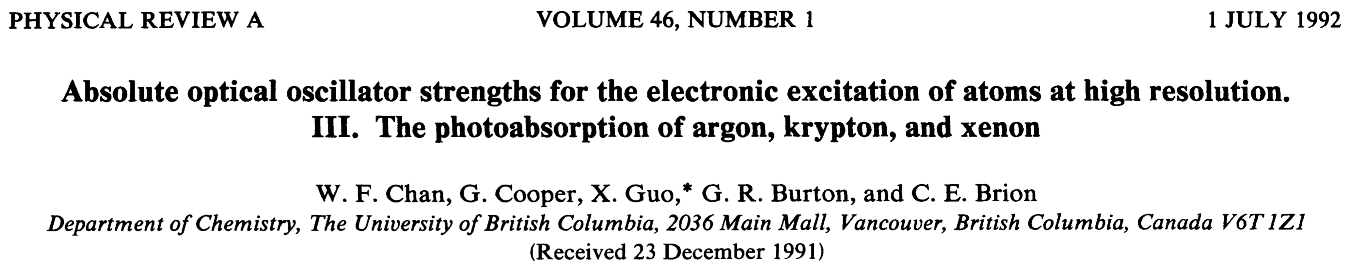
Absolute optical oscillator strengths for the electronic excitation of atoms at high resolution.
III. The photoabsorption of argon, krypton, and xenon

W. F. Chan, G. Cooper, X. Guo,* G. R. Burton, and C. E. Brion
Department of Chemistry, The University of British Columbia, 2036 Main Mall, Vancouver, British Columbia, Canada V6T 1Z1

(Received 23 December 1991)



XUV Absorption



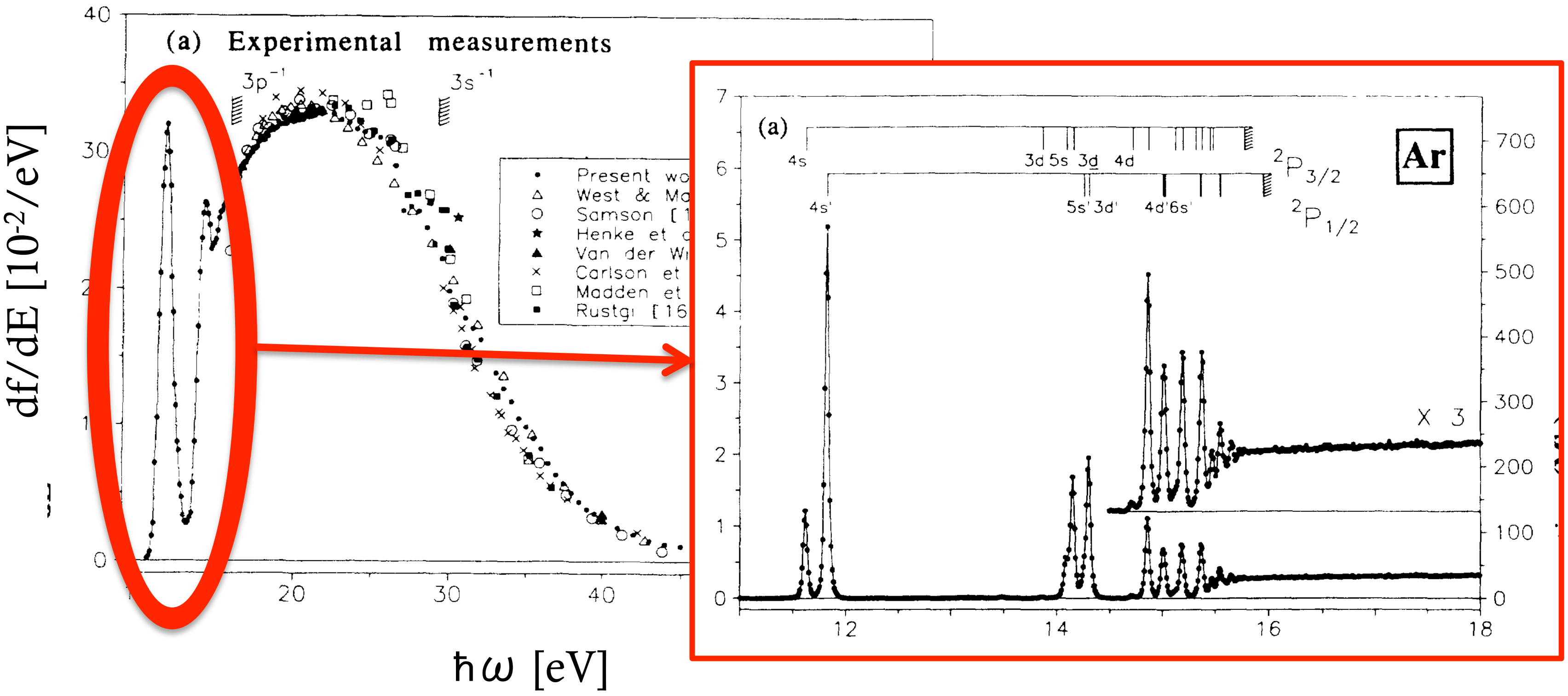
PHYSICAL REVIEW A VOLUME 46, NUMBER 1 1 JULY 1992

Absolute optical oscillator strengths for the electronic excitation of atoms at high resolution. III. The photoabsorption of argon, krypton, and xenon

W. F. Chan, G. Cooper, X. Guo,* G. R. Burton, and C. E. Brion

Department of Chemistry, The University of British Columbia, 2036 Main Mall, Vancouver, British Columbia, Canada V6T 1Z1

(Received 23 December 1991)

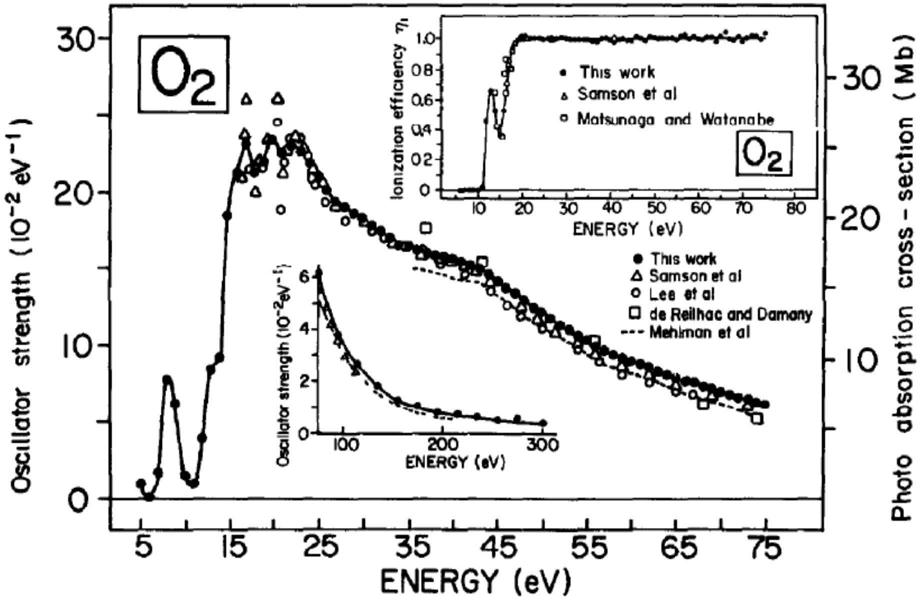


Extreme Ultraviolet Spectroscopy

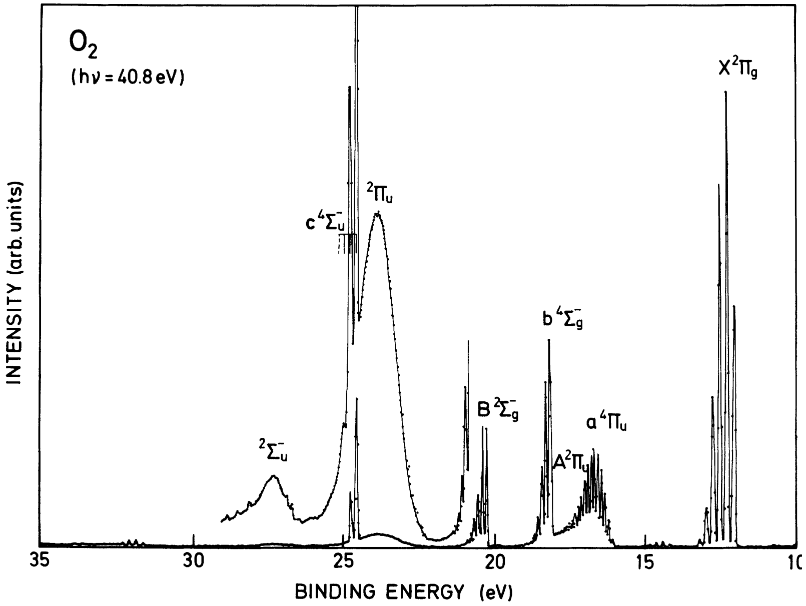
- VUV and XUV spectroscopy has told us much of what we know about atoms, molecules, and quantum mechanics (e.g. Spectroscopy of Hydrogen, Helium, Simple molecules)

- Most of the Oscillator strength of atoms, molecules, or solids lies in the VUV and XUV, in Rydberg excitations and the photoionization continuum.

<u>Photoabsorption</u>



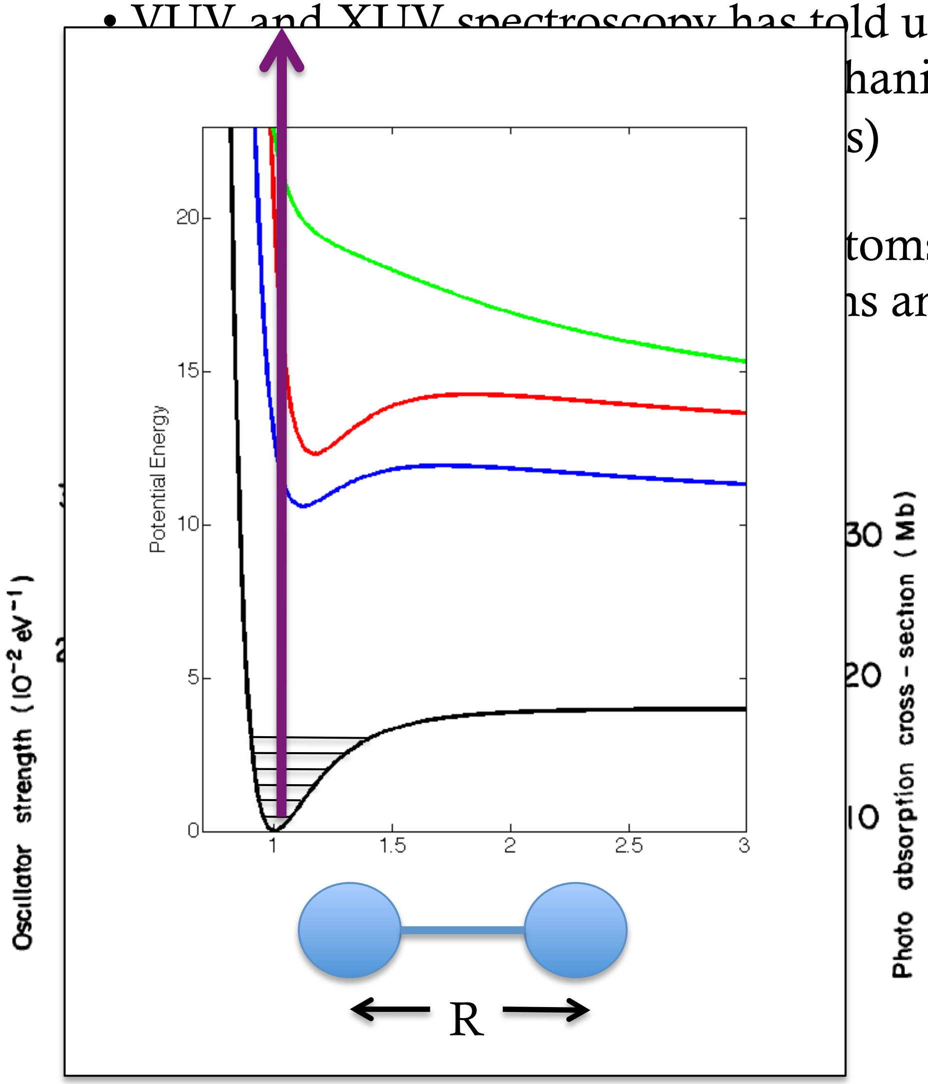
<u>Photoelectrons</u>



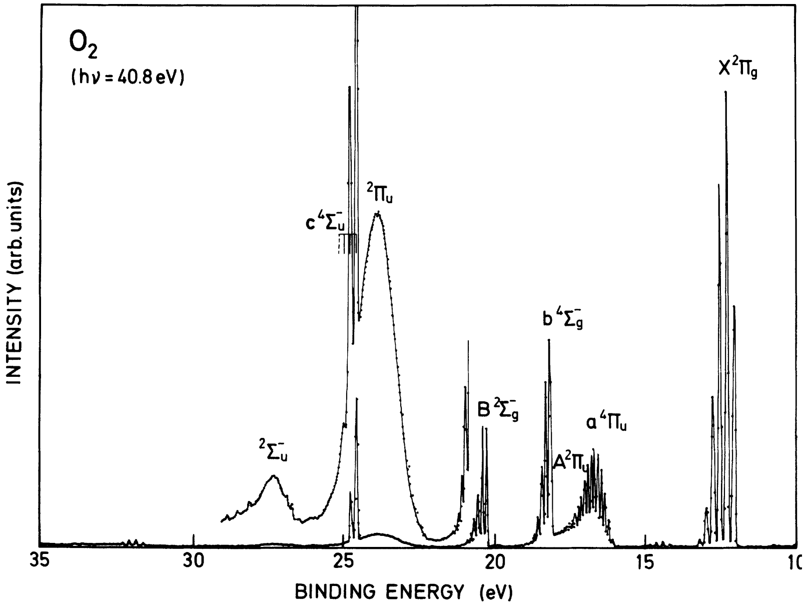
Photoionization spectroscopy now quite sophisticated! (e.g. COLTRIMS, VMI)

Extreme Ultraviolet Spectroscopy

- VUV and XUV spectroscopy has told us much of what we know about hanics (e.g. Spectroscopy of s)
- toms, molecules, or solids lies in the ns and the photoionization continuum.



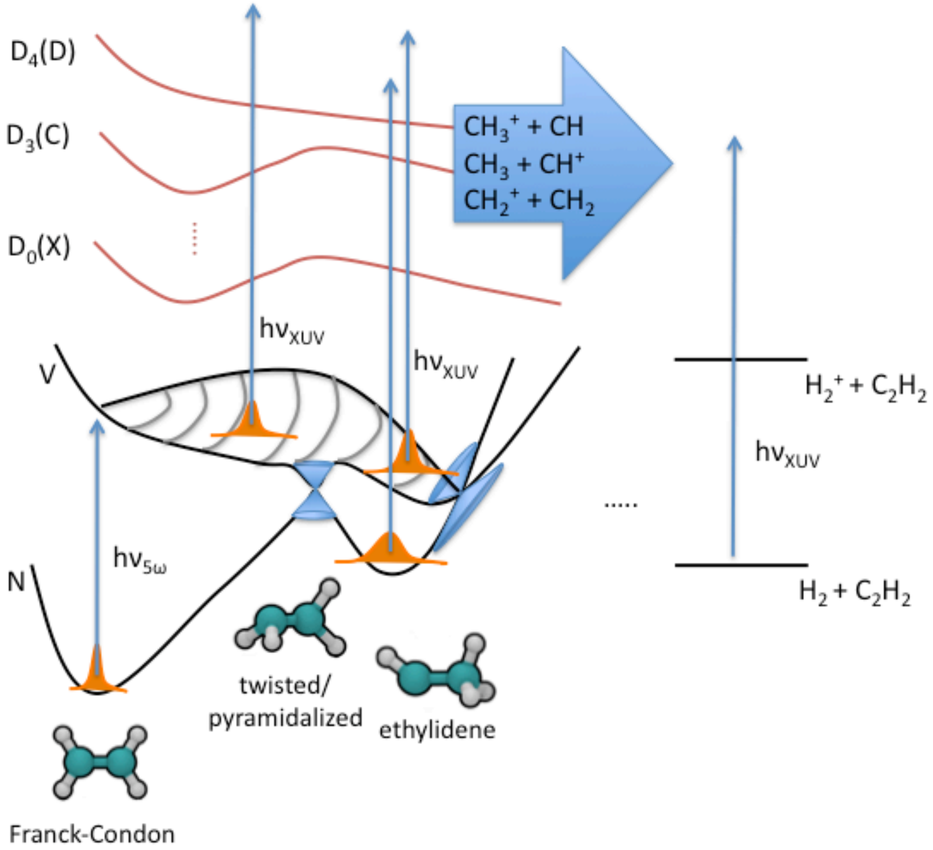
Photoelectrons

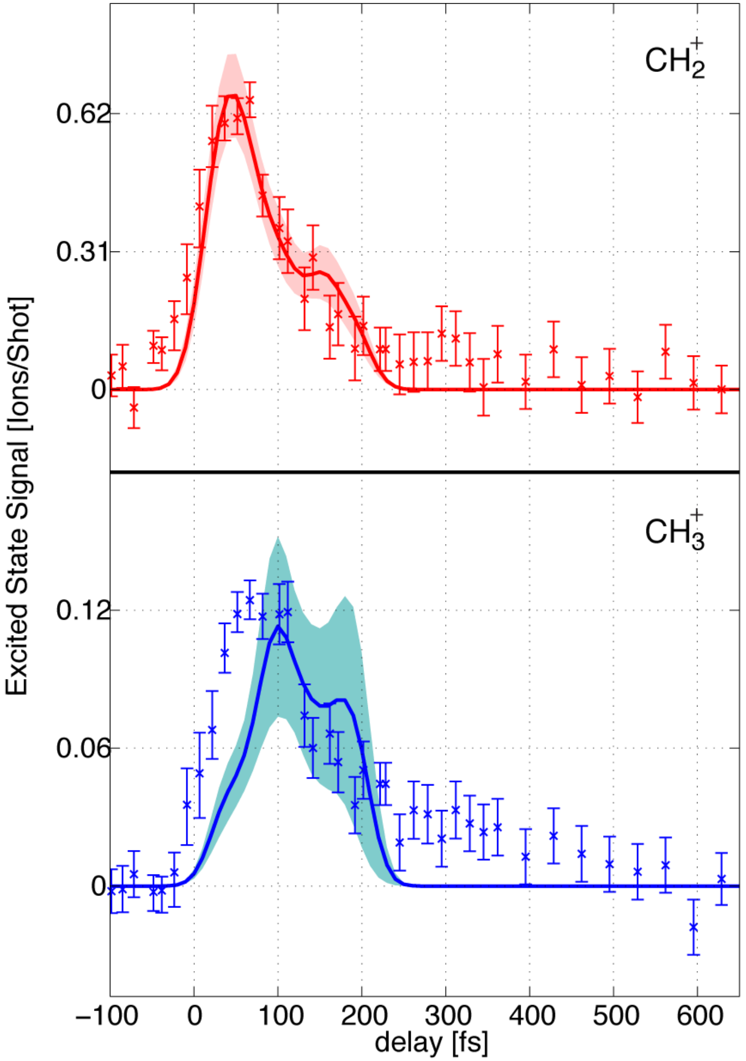


Photoionization spectroscopy now quite sophisticated! (e.g. COLTRIMS, VMI)

VUV/XUV Time Domain Studies

- Time resolved photoionization reveals molecular dynamics.

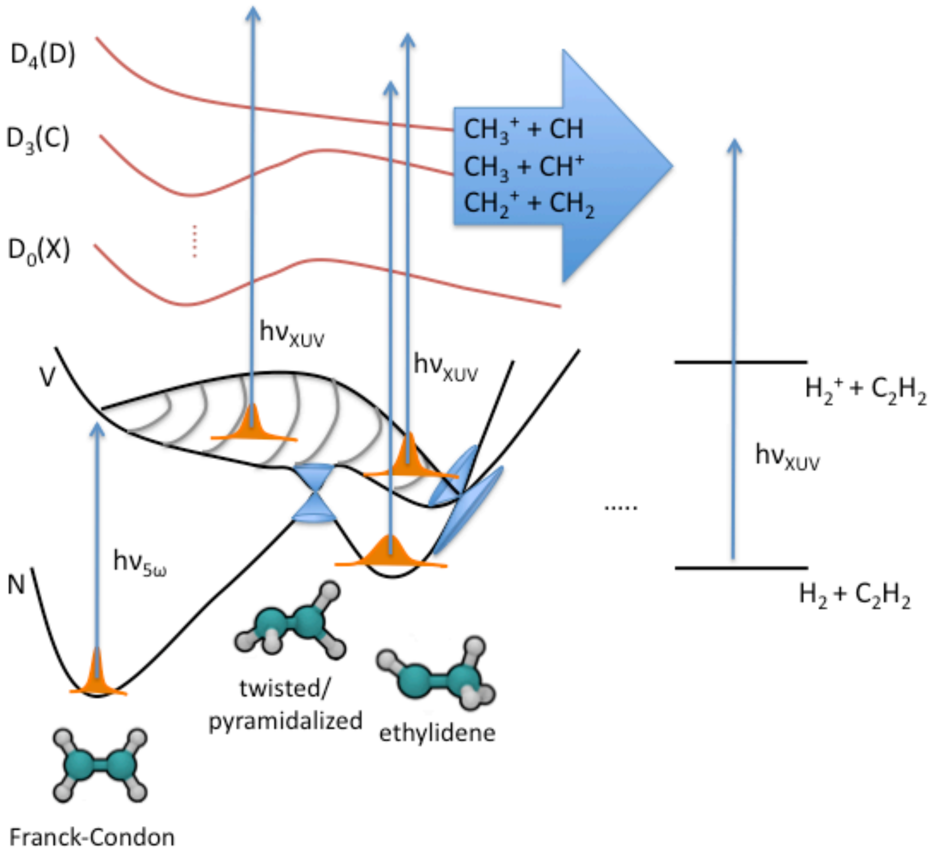


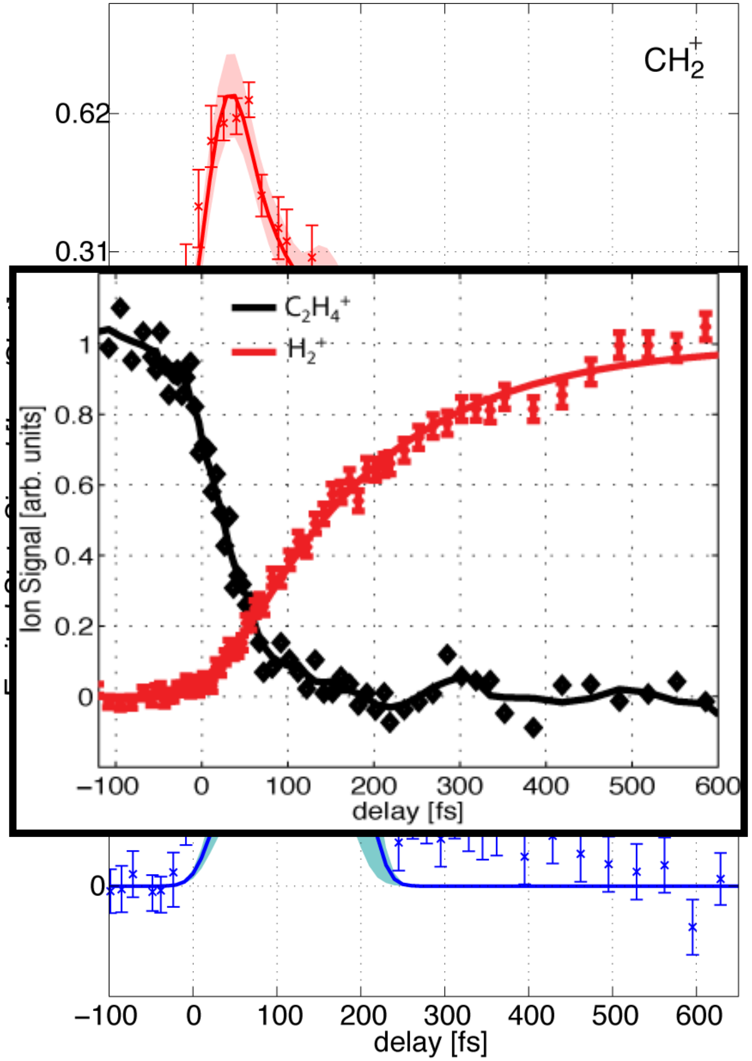


T. K. Allison et al., *Opt. Lett.* **35**, 3664 (2010)
H. Tao, T. K. Allison et al., *J. Chem Phys.* **134**, 244306 (2011)
T. K. Allison et al., *J. Chem. Phys.* **136**, 124317 (2012)

VUV/XUV Time Domain Studies

- Time resolved photoionization reveals molecular dynamics.





T. K. Allison et al., *Opt. Lett.* **35**, 3664 (2010)
H. Tao, T. K. Allison et al., *J. Chem Phys.* **134**, 244306 (2011)
T. K. Allison et al., *J. Chem. Phys.* **136**, 124317 (2012)

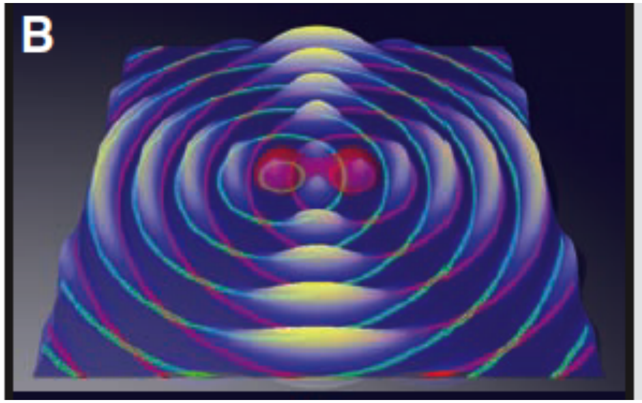
Photoelectron Imaging

- The absorption or emission of XUV light is associated with free electrons with Angstrom-scale de Broglie wavelengths.

- This can be used to probe molecular structure and dynamics.

$$KE = \frac{(\hbar k)^2}{2m_e} = h\nu \pm E_b$$

$$\Rightarrow \lambda_{DB} = \frac{12.3 \text{ Å}}{\sqrt{KE \text{ [eV]}}}$$



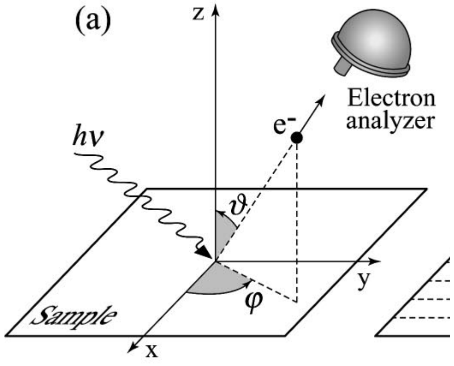
D. Akoury et al., Science (2007)

Molecular frame photoelectron angular distributions:

- Adiabatic and field-free laser alignment techniques
- COLTRIMS coincidence measurements, "post-partum" alignment.

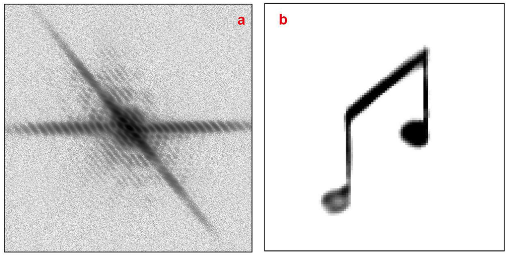
More XUV Applications

- Angle Resolved Photoemission (ARPES)
    - ○ Studying electron motion and coordination in condensed matter.



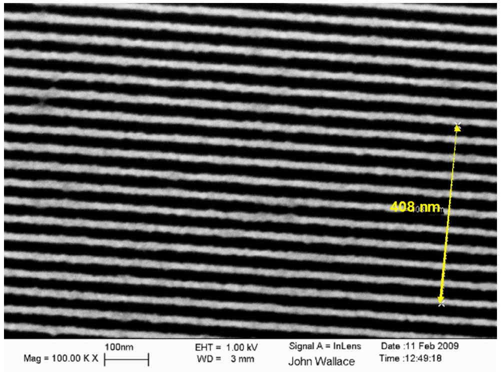
Photoemission geometry
Damascelli et al. Rev. Mod. Phys. (2003)

- Nano-scale physics
    - ○ Nano-scale imaging.
    - ○ Nano-scale heat transport.
    - ○ Magnetic domain dynamics



Coherent diffraction imaging with 32 nm light.
A Ravasio et al., *Phys. Rev. Lett. (2008)*

- EUV Lithography
    - ○ Pattern much smaller features.
    - ○ Major comercial efforts (e.g. Intel, IBM)

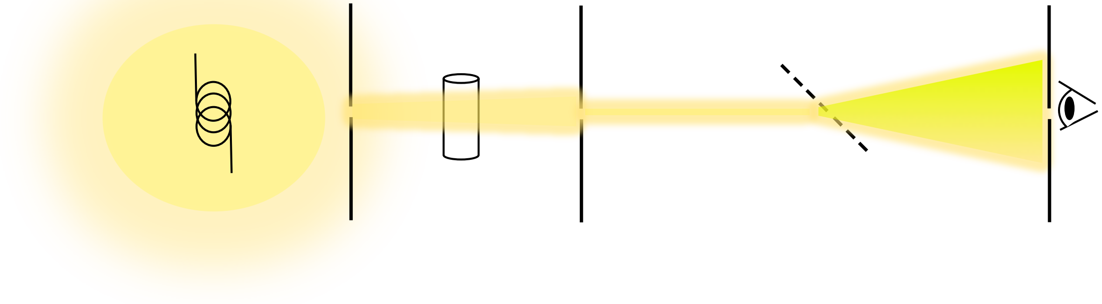


Frequency/Wavelength Metrology?

Dispersive Spectrometer

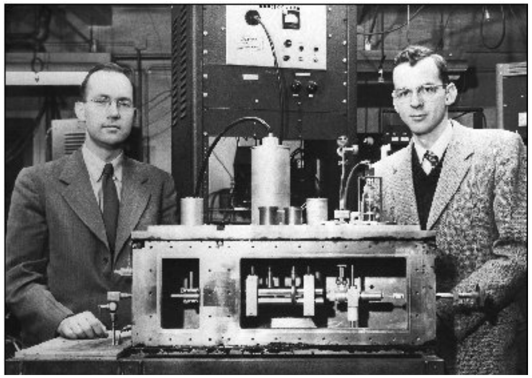
- Measure wavelength
- Resolution 10^{-6}

ca. 1670
I. Newton

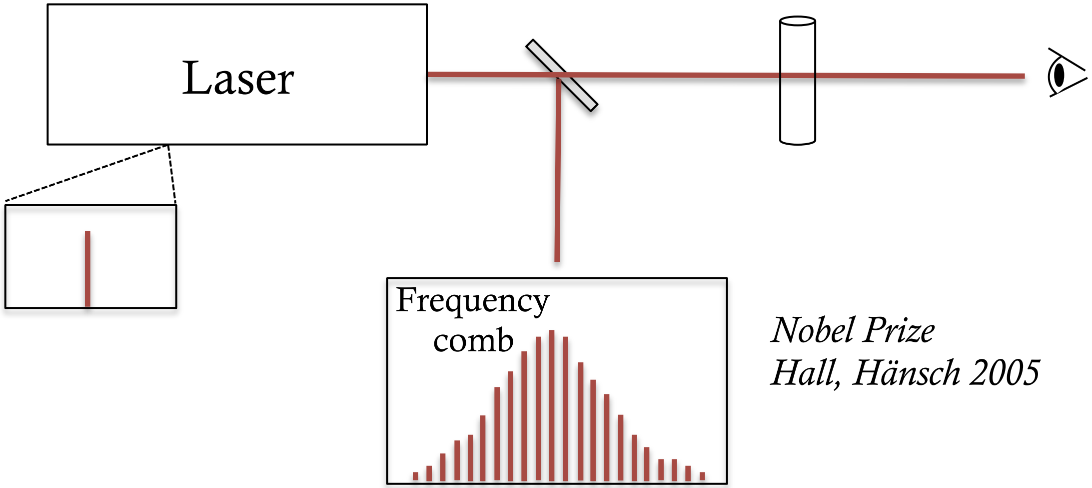


Laser spectroscopy

- Measure frequency
- Resolution $< 10^{-15}$



ca. 1960
C. Townes

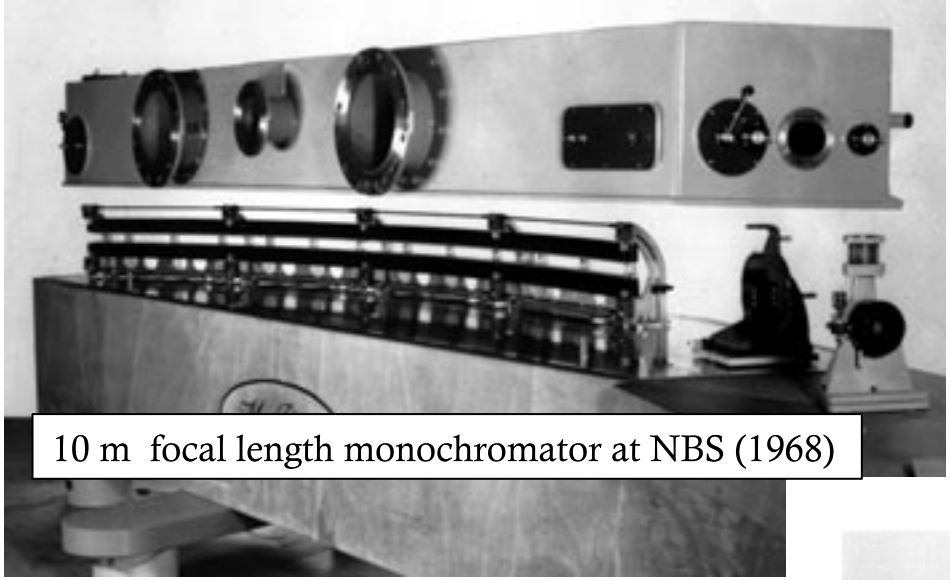


Laser

Frequency
comb

Nobel Prize
Hall, Hänsch 2005

VUV/XUV Wavelength Metrology



10 m focal length monochromator at NBS (1968)

Rowland, Lyman, Schuman, Muliken…

Madden, Codling, Ederer, Sampson…

Autoionizing states in He

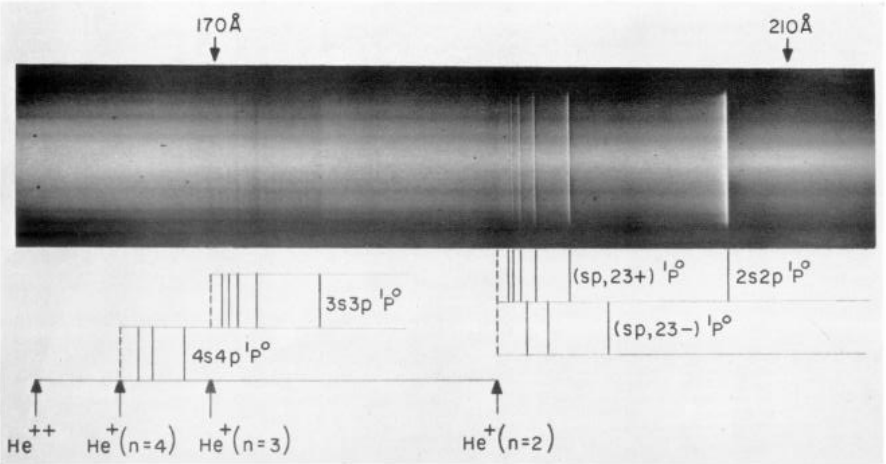
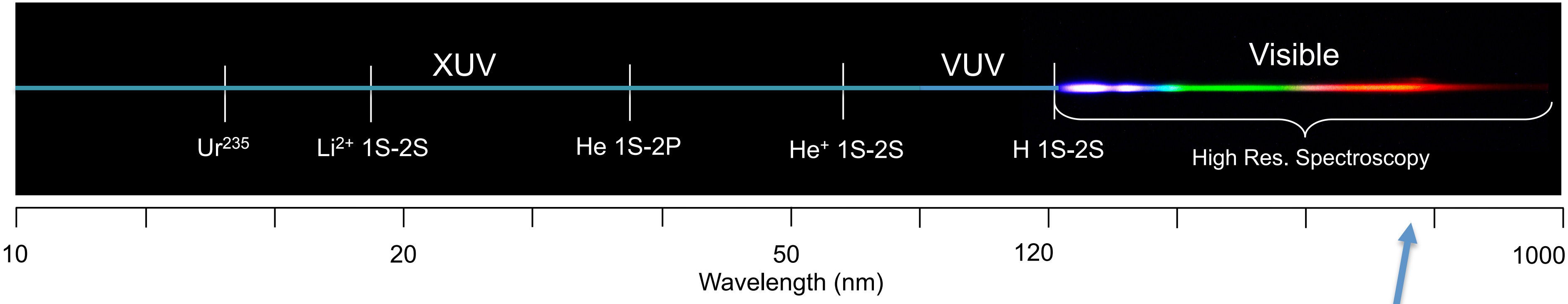


FIG. 3.—The absorption spectrum of helium between 160 and 215 Å showing the many resonances due to the existence of two-electron excitation states in neutral helium. The photograph is a positive print (black is absorption). The white portion of each resonance indicates a reduced-absorption zone or "window" in the continuous photoionization background absorption.

Metrology Motivation

Ultrahigh-resolution spectroscopy at short wavelengths



- High precision tests of QED in H, He and like ions ($\sim Z^4 - Z^6$).
- Next generation "nuclear" clocks in ^{229}Th and ^{235}U.
- Variation of fundamental constants in highly charged ions ($\sim(Z+1)^2$).

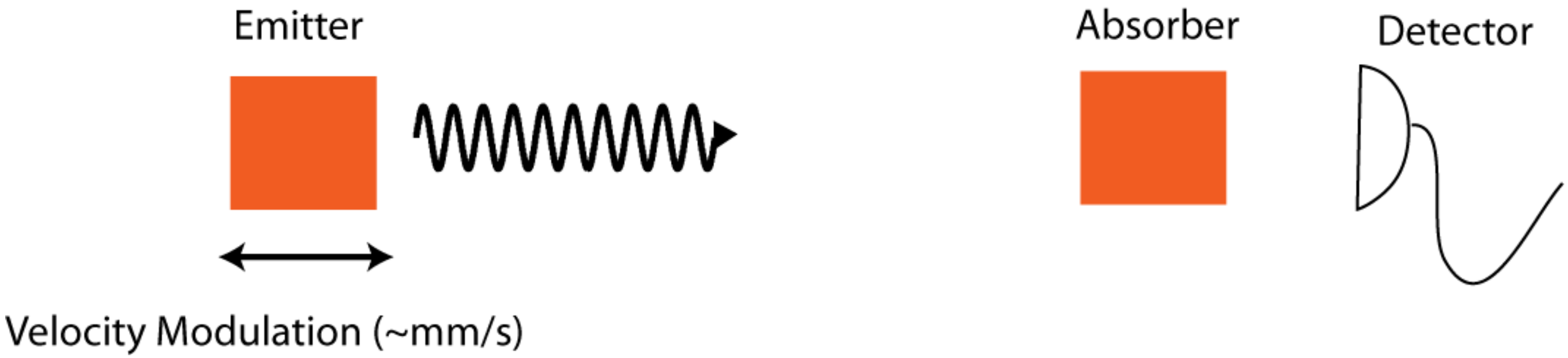
Optical Atomic Clocks

Uncertainty and stability at the $= 10^{-18}$ level.

Laser stabilization

Mössbauer Spectroscopy

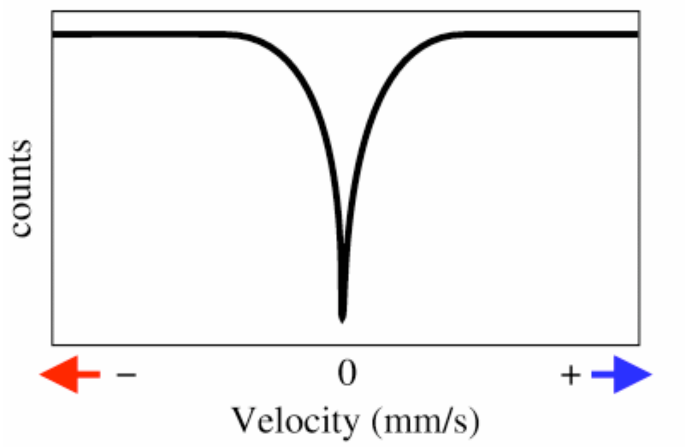
In the Mössbauer effect, a nuclear decay emits a photon without recoil, resulting in exceptionally narrow x-ray and γ-ray lines.

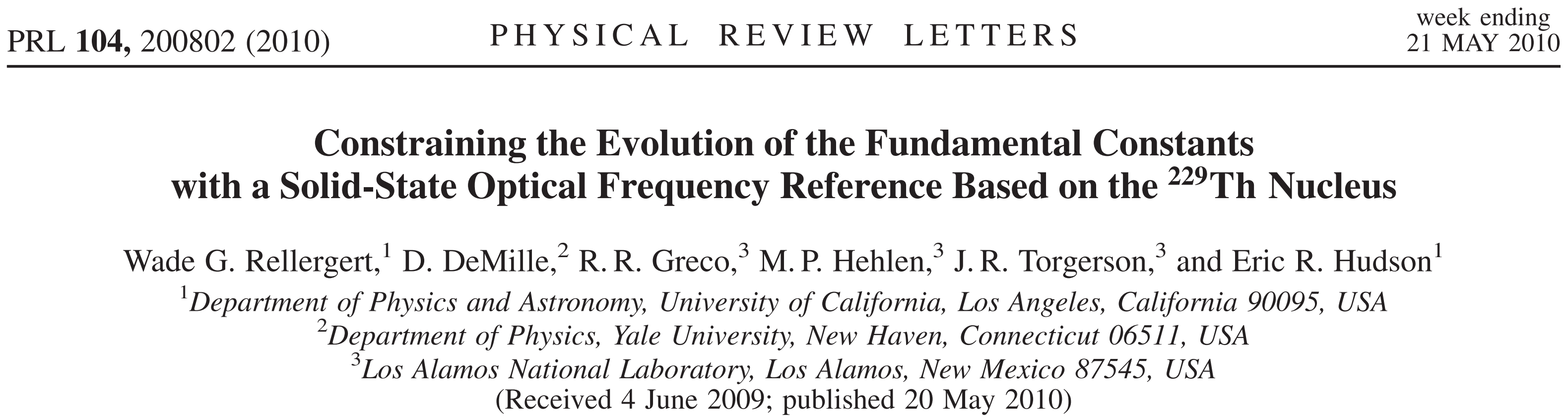


- Can be a sensitive probe of chemical environment, innermost electron orbitals.

- Linewidths and shifts are resolved by introducing a doppler shift to the emitter or source, but absolute position is known to much lower accuracy.

- Linewidths can be MHz to kHz
- (e.g. ^{181}Ta with $\hbar\omega$ = 6.2 keV, Δf = 37 kHz, $Q \sim 4\times10^{13}$)



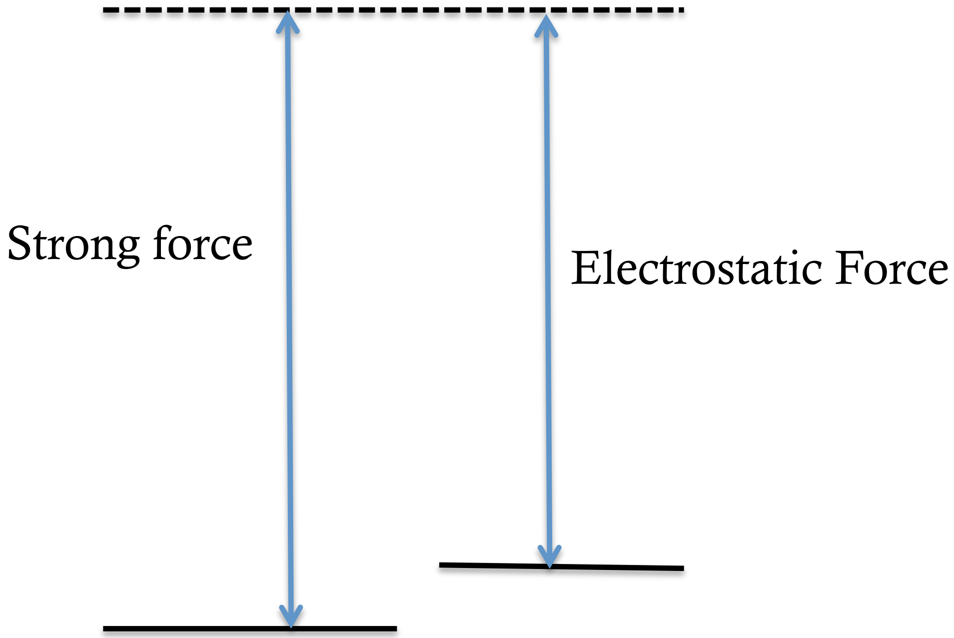


PRL **104,** 200802 (2010) PHYSICAL REVIEW LETTERS week ending 21 MAY 2010

Constraining the Evolution of the Fundamental Constants with a Solid-State Optical Frequency Reference Based on the ^{229}Th Nucleus

Wade G. Rellergert,[1] D. DeMille,[2] R. R. Greco,[3] M. P. Hehlen,[3] J. R. Torgerson,[3] and Eric R. Hudson[1]
[1] *Department of Physics and Astronomy, University of California, Los Angeles, California 90095, USA*
[2] *Department of Physics, Yale University, New Haven, Connecticut 06511, USA*
[3] *Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

(Received 4 June 2009; published 20 May 2010)



Strong force

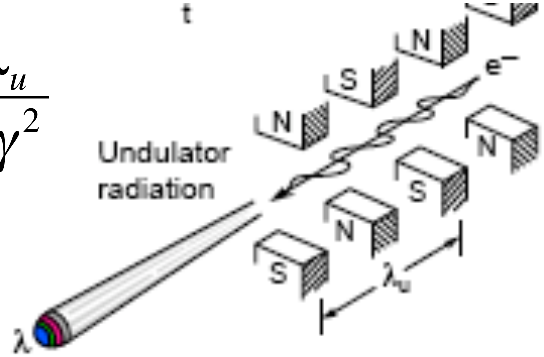
Electrostatic Force

Despite low energy excitation, MeV scale physics is at play

⇒ Big "lever arm" for seeing for seeing changes in fundamental constants.

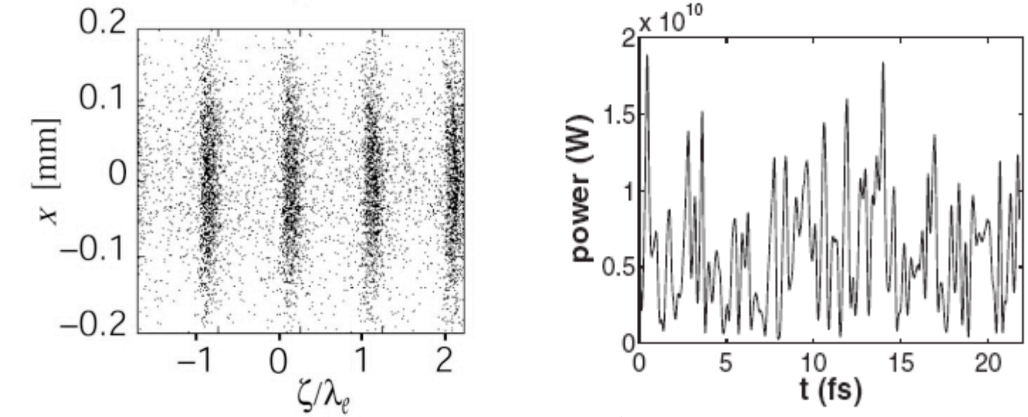
Undulator Radiation

$$\lambda \approx \frac{\lambda_u}{2\gamma^2}$$



Can be partially spatially coherent, but still essentially
a temporally incoherent "light bulb"

<u>XUV and X-ray Free electron lasers?</u>

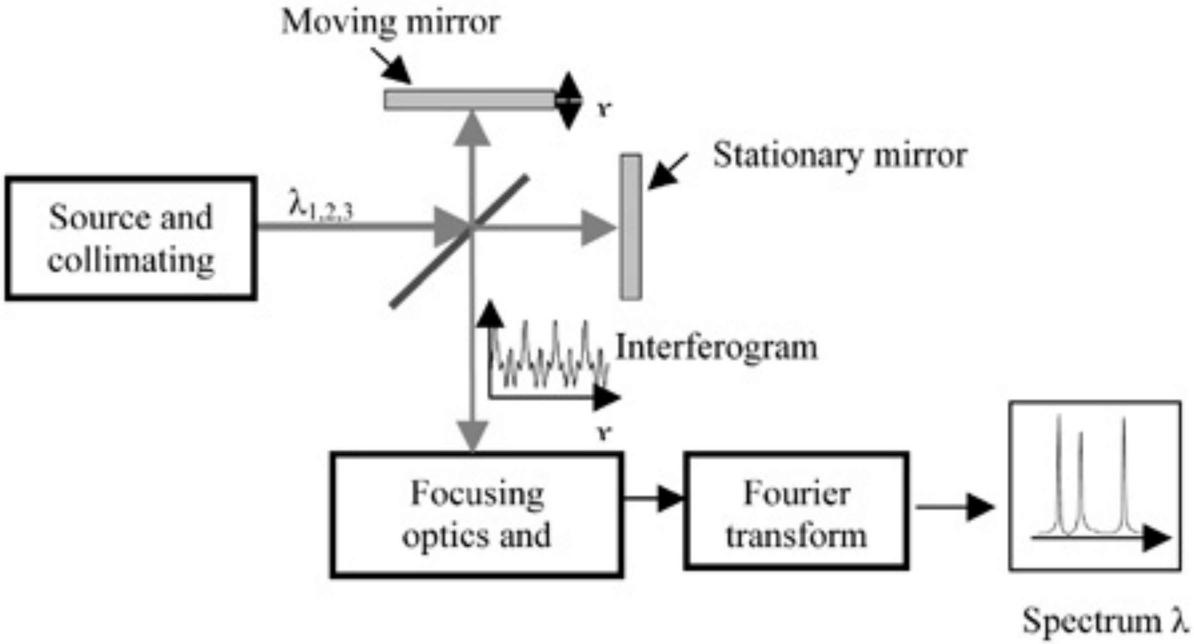


Z. Huang and K. Kim, PRSTAB (2007)

$$L_{cooperation} \equiv \lambda_r \frac{L_G}{\lambda_u} \sim 10-100 \text{ nm}$$

$$\Rightarrow T_{coh} = \frac{L_{cooperation}}{c} < 1 \text{ fs}$$

Fourier Transform Spectroscopy?

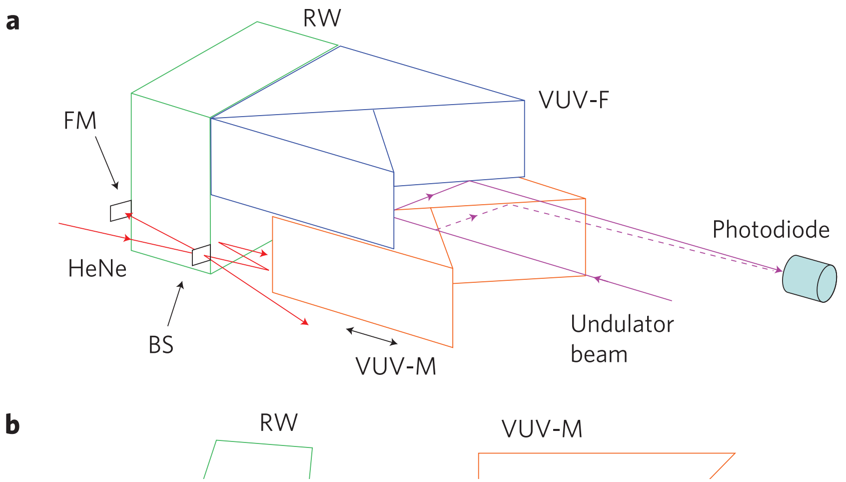


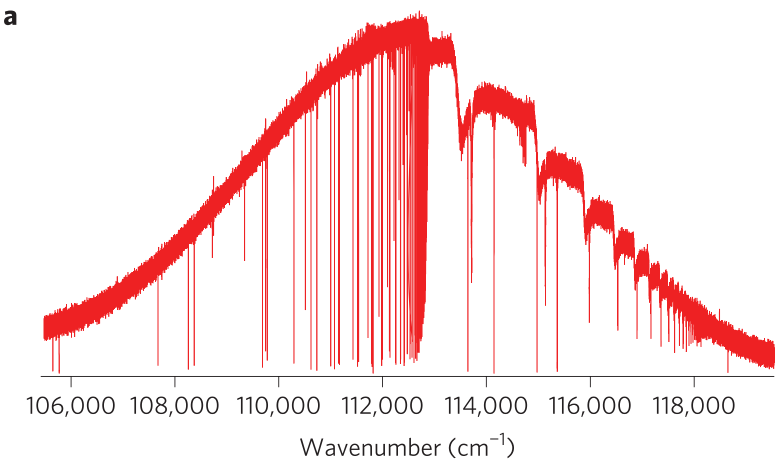
- In Principle, yes!
- In Practice, very difficult because optics are hard and wavelength is short.
- Resolution ∝ 1/path length difference ⇒ Can only go but so far.

nature photonics
LETTERS
PUBLISHED ONLINE: 23 JANUARY 2011 | DOI: 10.1038/NPHOTON.2010.314

High-resolution broad-bandwidth Fourier-transform absorption spectroscopy in the VUV range down to 40 nm

Nelson de Oliveira[1]*, Mourad Roudjane[1†], Denis Joyeux[1,2], Daniel Phalippou[2], Jean-Claude Rodier[2] and Laurent Nahon[1]





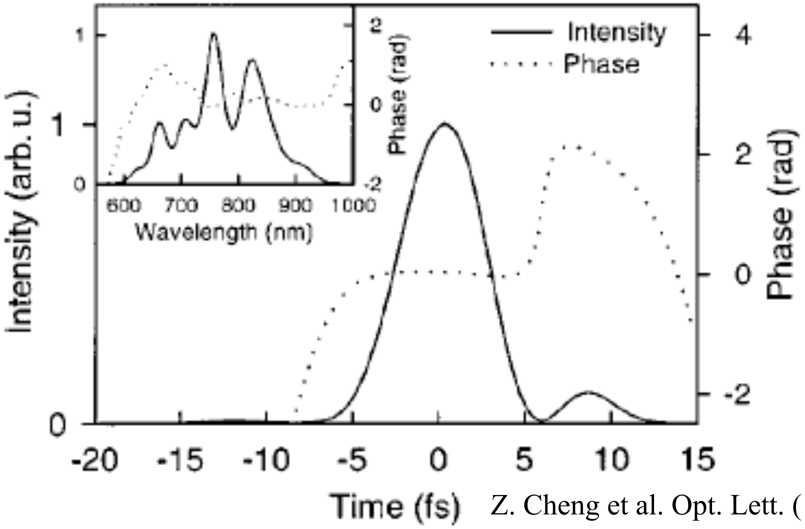
~ GHz resolution FT Spectroscopy

Lasers

Spatially and *Temporally* coherent.

Time Domain: Complete control over ultrashort pulses with ~100 THz bandwidth.

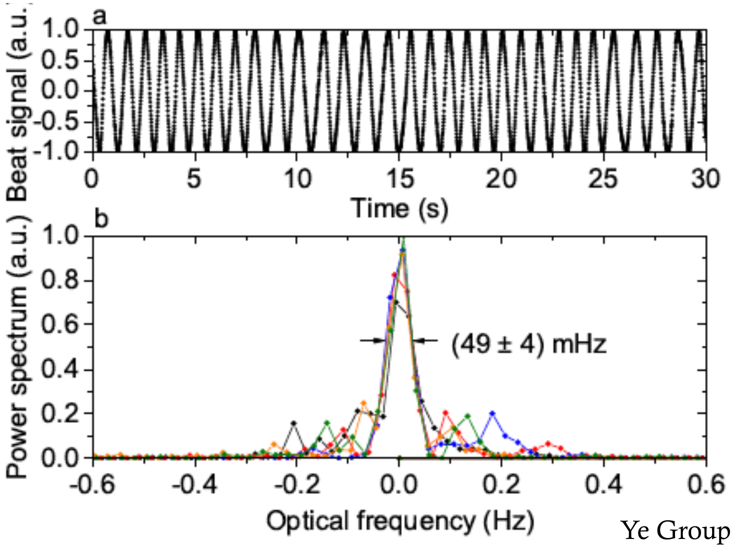
- Femtochemistry
- Coherent control
- Extreme nonlinear optics



Z. Cheng et al. Opt. Lett. (1999)

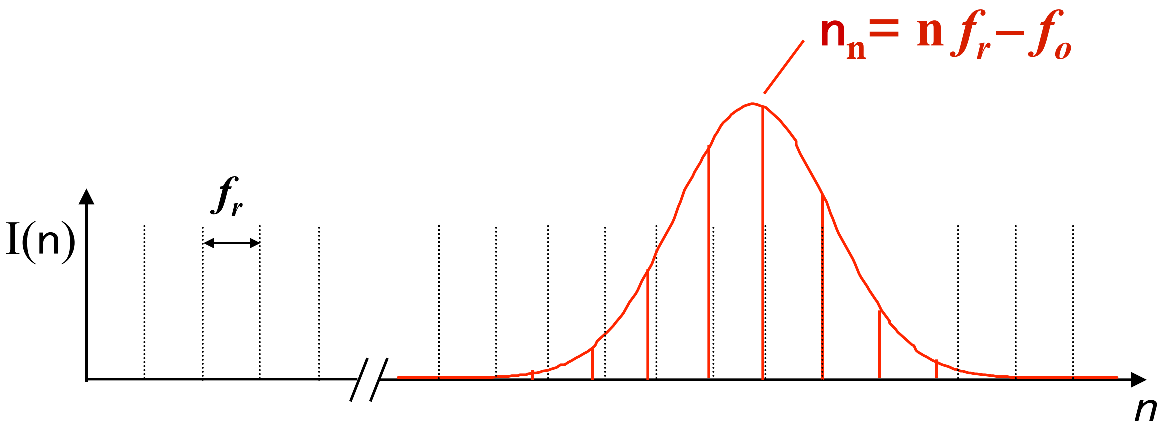
Frequency domain: Stable, precise, and tunable optical frequencies.

- High-resolution spectroscopy
- Laser cooling et al.
- Precision Measurement
  ~ 1 part in 10^{15} accuracy!



Ye Group (2011)

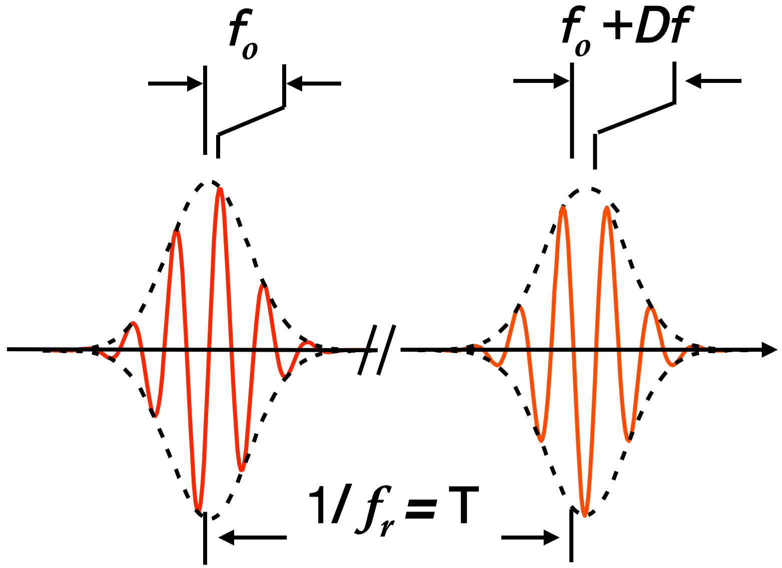
Frequency Combs



f_r = Comb spacing

f_o = Comb offset from harmonics of f_r

$\Delta\phi$ = Phase slip b/t carrier & envelope each round trip



Directly relate optical frequencies to RF

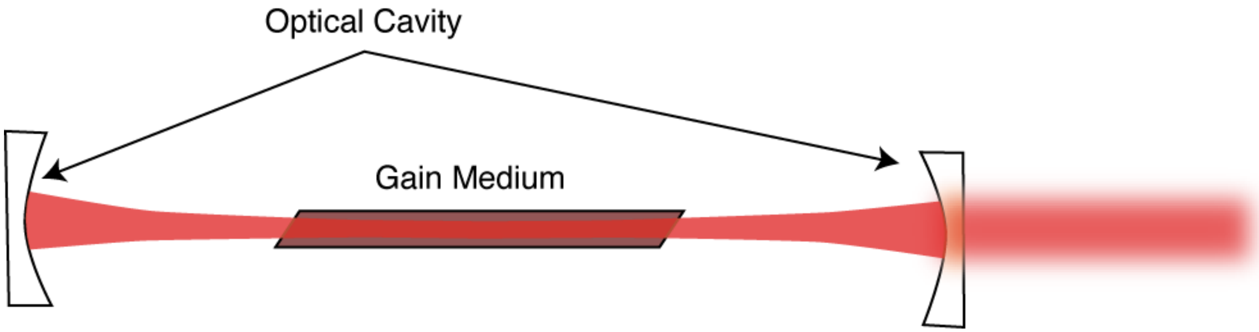
⇒ Optical clocks.

⇒ Precision measurement.

Also essential for attosecond physics through carrier envelope phase control.

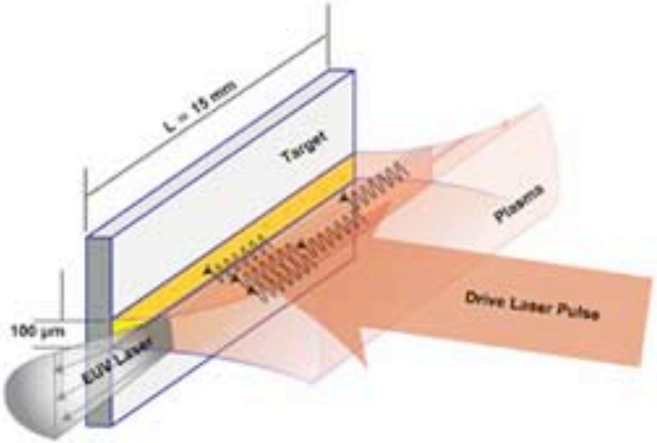
Comb tooth linewidth determined by coherence time of pulse train, not the pulse duration!

XUV Lasers?



In the XUV, lasers are hindered by:

- Poor optics performance (R < 50%) ⇒ Single pass only.
- No transparent materials ⇒ Only gas/plasma gain media.
- If this weren't difficult enough, $P_{pump} \propto \lambda^{-4} - \lambda^{-5}$ necessitates pulsed operation.



⇒ Not a precision instrument in the way we normally think of lasers.

Nonlinear Optics

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

Linear optics: absorption, refraction, scattering

$\chi^{(2)}$ processes: frequency doubling, sum and difference frequency generation, optical rectification.

$\chi^{(3)}$ processes: Four wave mixing, self focusing, self phase modulation.

$$E = E_0 \cos(\omega t) \Rightarrow P^{(2)} = \chi^{(2)} E_0^2 \cos^2(\omega t)$$

$$= \frac{1}{2} \chi^{(2)} E_0^2 [1 + \cos(2\omega t)]$$

The second harmonic!

Just turn up the power?

- Perturbation description fails when $E_0 \sim 1$ V/Å, $I \sim 10^{13}$-10^{14} W/cm^2
- Need to consider field ionization: “Extreme Nonlinear Optics”

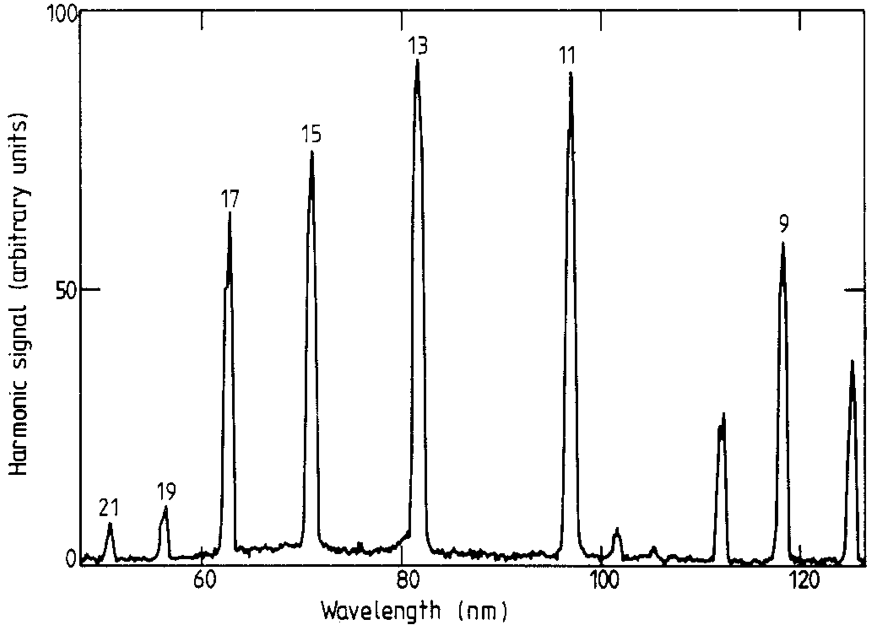
J. Phys. B: At. Mol. Opt. Phys. **21** (1988) L31-L35. Printed in the UK

LETTER TO THE EDITOR

Multiple-harmonic conversion of 1064 nm radiation in rare gases

M Ferray, A L'Huillier, X F Li, L A Lompré, G Mainfray and C Manus
Service de Physique des Atomes et des Surfaces, 91191 Gif sur Yvette, Cédex, France

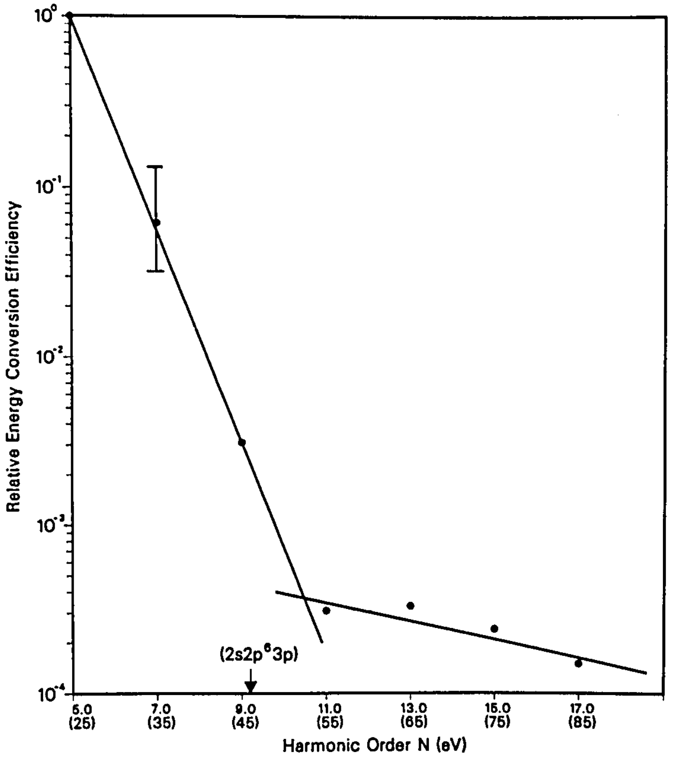
Received 2 November 1987



McPherson *et al.*
Vol. 4, No. 4/April 1987/J. Opt. Soc. Am B

Studies of multiphoton production of vacuum-ultraviolet radiation in the rare gases

A. McPherson, G. Gibson, H. Jara, U. Johann, T. S. Luk, I. A. McIntyre, K. Boyer, and C. K. Rhodes
Department of Physics, University of Illinois at Chicago, P.O. Box 4348, Chicago, Illinois 60680



VOLUME 68, NUMBER 24 PHYSICAL REVIEW LETTERS 15 JUNE 1992

High-Order Harmonic Generation from Atoms and Ions in the High Intensity Regime

Jeffrey L. Krause, Kenneth J. Schafer, and Kenneth C. Kulander
Physics Department, Lawrence Livermore National Laboratory, Livermore, California 94550

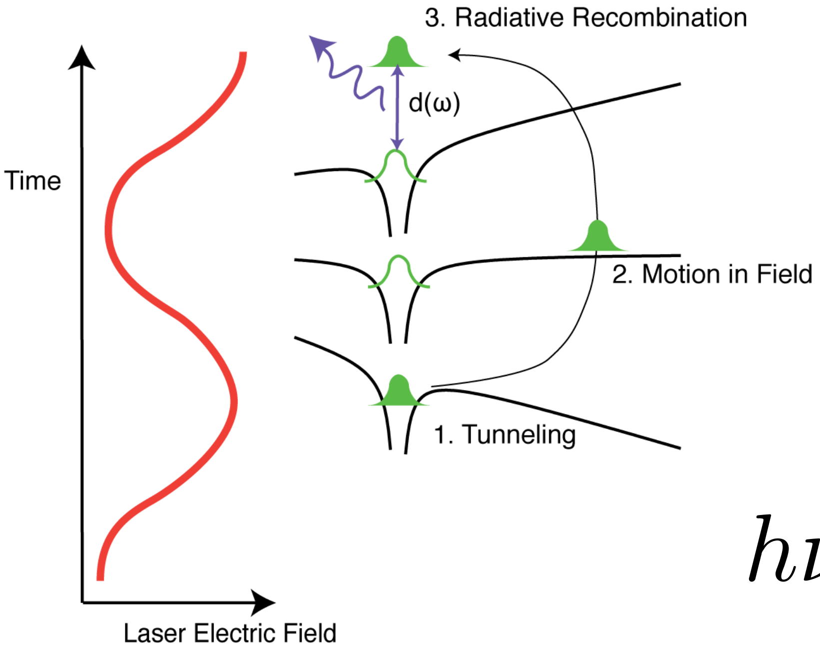
(Received 6 March 1992)

VOLUME 71, NUMBER 13 PHYSICAL REVIEW LETTERS 27 SEPTEMBER 1993

Plasma Perspective on Strong-Field Multiphoton Ionization

P. B. Corkum
National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6

(Received 9 February 1993)



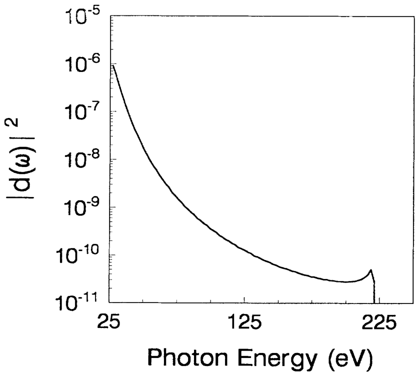


FIG. 4. Calculated value of the square of the dipole moment (measured in atomic units) plotted as a function of E_h of the harmonic. The calculation was performed for 1.06 μm fundamental radiation interacting with helium with an intensity of 6×10^{14} W/cm^2. The parameters were chosen to match those in Ref. [6].

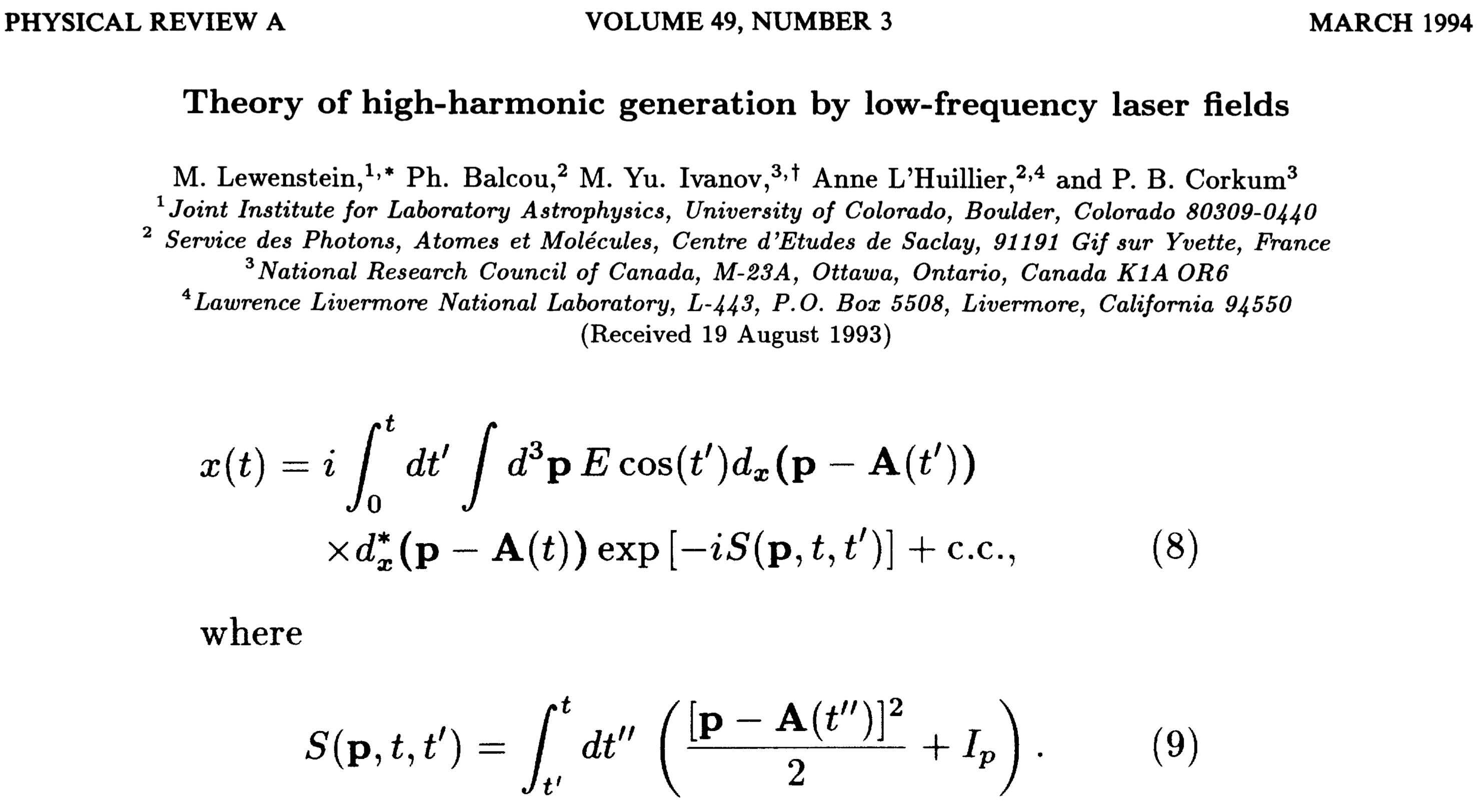
$$h\nu_{\text{cutoff}} = 3.17U_p + I_p$$

The Pondermotive Potential

$$\vec{F} = m\vec{a} = -e\vec{E}(t) = -e\vec{E}_0 \cos(\omega t)$$

$$U_p = \left\langle \frac{1}{2} m v^2 \right\rangle = \frac{e^2 E_0^2}{4 m \omega^2}$$

At 800 nm, 1 × 10^{14} W/cm^2 U_P = 6 eV, E = 2 V/Å



PHYSICAL REVIEW A VOLUME 49, NUMBER 3 MARCH 1994

Theory of high-harmonic generation by low-frequency laser fields

M. Lewenstein,[1,*] Ph. Balcou,[2] M. Yu. Ivanov,[3,†] Anne L'Huillier,[2,4] and P. B. Corkum[3]
[1] *Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colorado 80309-0440*
[2] *Service des Photons, Atomes et Molécules, Centre d'Etudes de Saclay, 91191 Gif sur Yvette, France*
[3] *National Research Council of Canada, M-23A, Ottawa, Ontario, Canada K1A OR6*
[4] *Lawrence Livermore National Laboratory, L-443, P.O. Box 5508, Livermore, California 94550*

(Received 19 August 1993)

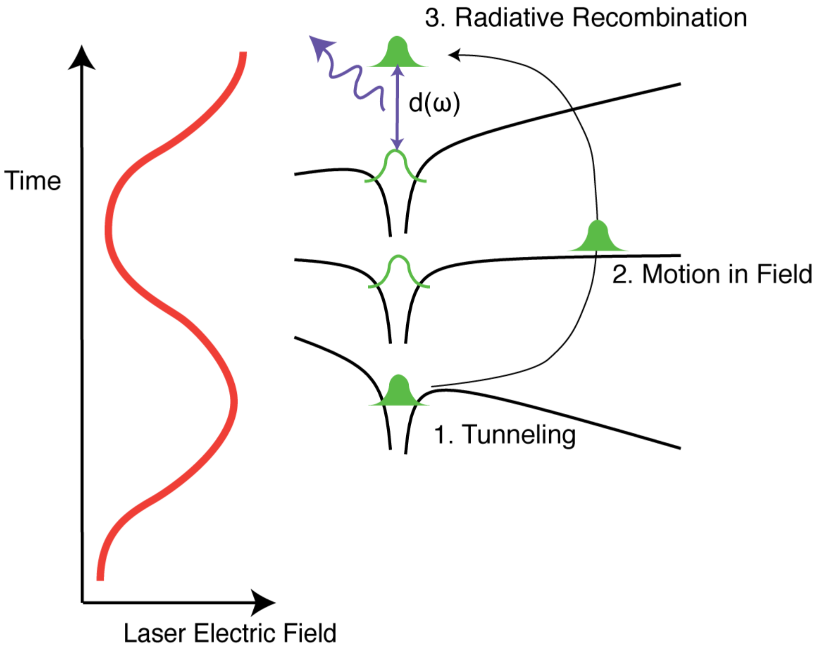
$$x(t) = i\int_0^t dt' \int d^3\mathbf{p}\, E\cos(t') d_x(\mathbf{p}-\mathbf{A}(t')) \times d_x^*(\mathbf{p}-\mathbf{A}(t)) \exp\left[-iS(\mathbf{p},t,t')\right] + \text{c.c.}, \qquad (8)$$

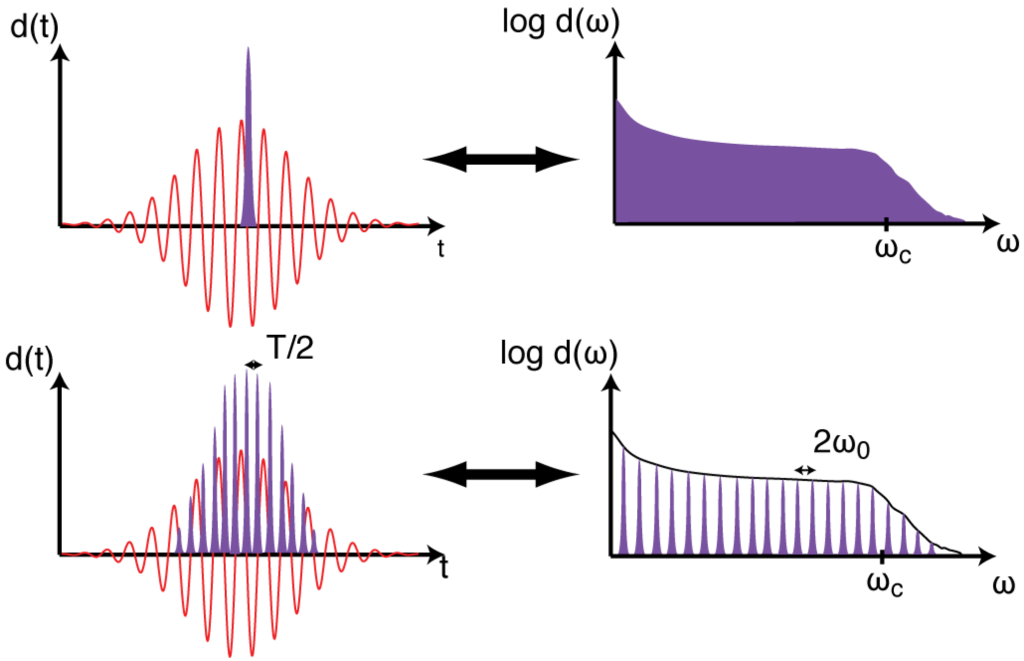
where

$$S(\mathbf{p},t,t') = \int_{t'}^{t} dt'' \left(\frac{[\mathbf{p}-\mathbf{A}(t'')]^2}{2} + I_p \right) . \qquad (9)$$

Quantum theory can also explain cutoff harmonics, phases, “atto-chirp”

High-Order Harmonic Generation (HHG)

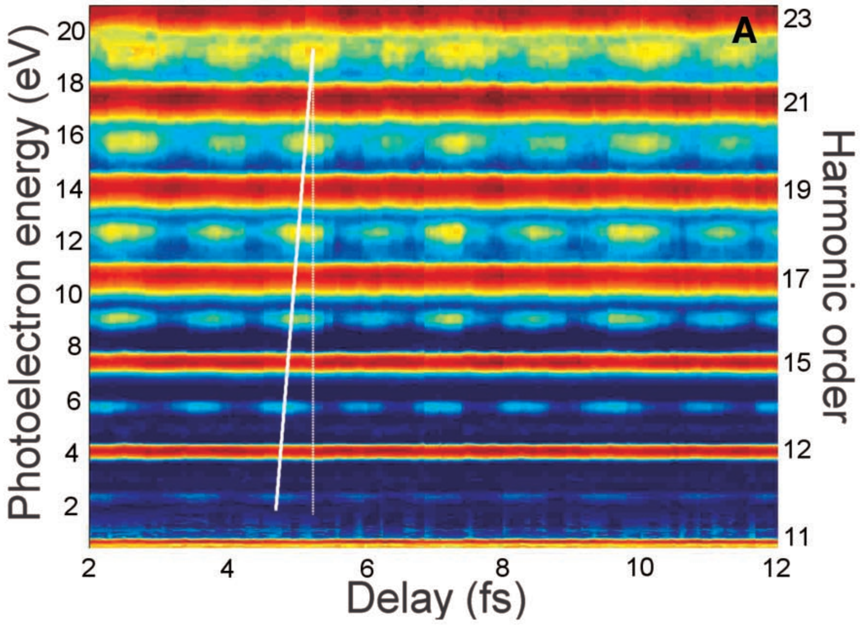


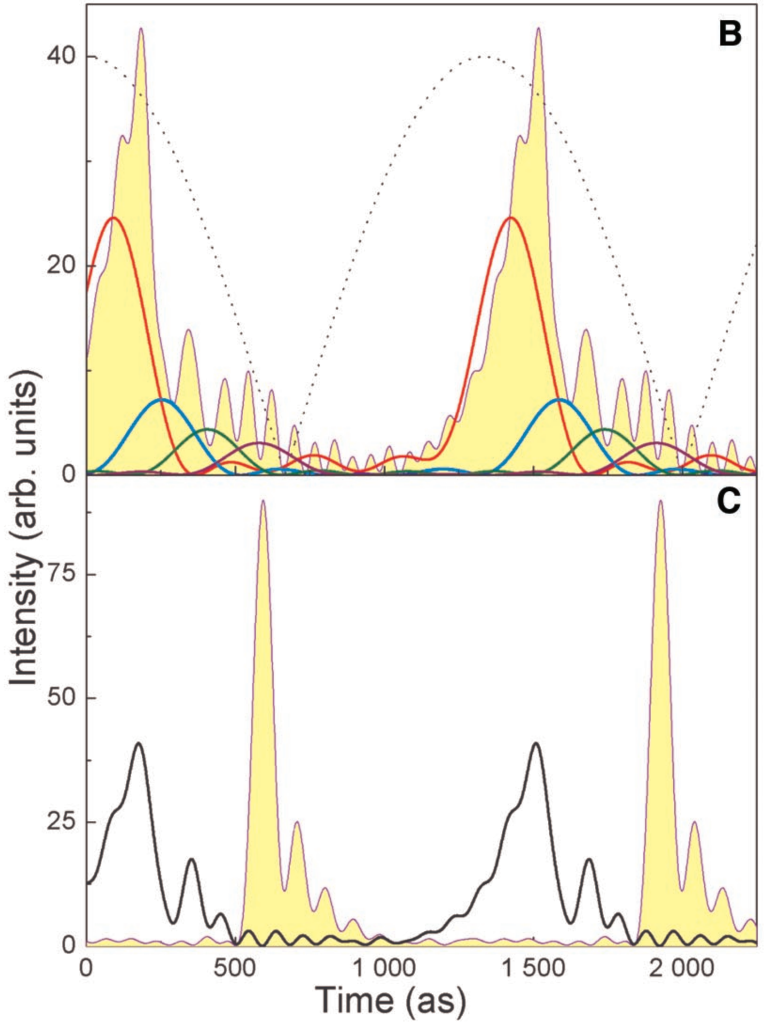


- Wavelength scaling as $P_{pump} \propto 1/\lambda$ (cf. $1/\lambda^4$ for an XUV laser)
- No particle accelerator.
- Definite phase relationship between XUV light and IR driving laser.
- Continues to surprise after > 20 years of research.

Attosecond Synchronization of High-Harmonic Soft X-rays

Y. Mairesse,[1] A. de Bohan,[1] L. J. Frasinski,[2] H. Merdji,[1]
L. C. Dinu,[3] P. Monchicourt,[1] P. Breger,[1] M. Kovačev,[1] R. Taïeb[4]
B. Carré,[1] H. G. Muller,[3] P. Agostini,[1] P. Salières[1]



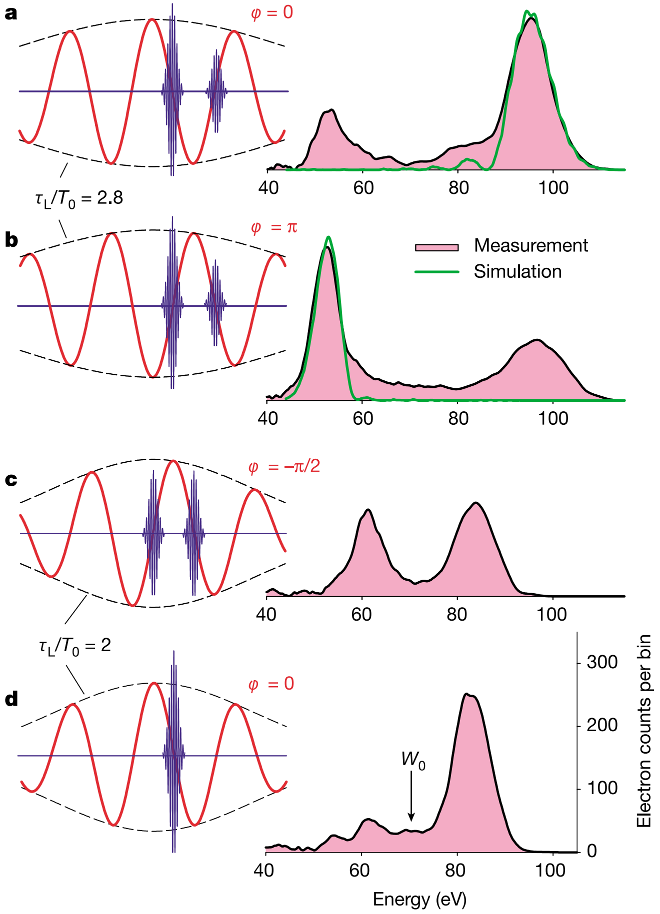


Atomic transient recorder

R. Kienberger[1]*, E. Goulielmakis[1]*, M. Uiberacker[1]*, A. Baltuska[1], V. Yakovlev[1], F. Bammer[2], A. Scrinzi[1], Th. Westerwalbesloh[3], U. Kleineberg[3], U. Heinzmann[3], M. Drescher[3] & F. Krausz[1,4]

[1] *Institut für Photonik, Technische Universität Wien, Gusshausstraße 27, A-1040 Wien, Austria*
[2] *Institut für Spanlose Fertigung und Hochleistungslasertechnik, Technische Universität Wien, Franz-Grillstr. 1, Arsenal Obj. 207, A-1030 Wien, Austria*
[3] *Fakultät für Physik, Universität Bielefeld, D-33615 Bielefeld, Germany*
[4] *Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, D-85748 Garching, Germany*

Many variants of this, e.g. FROG-CRAB etc.



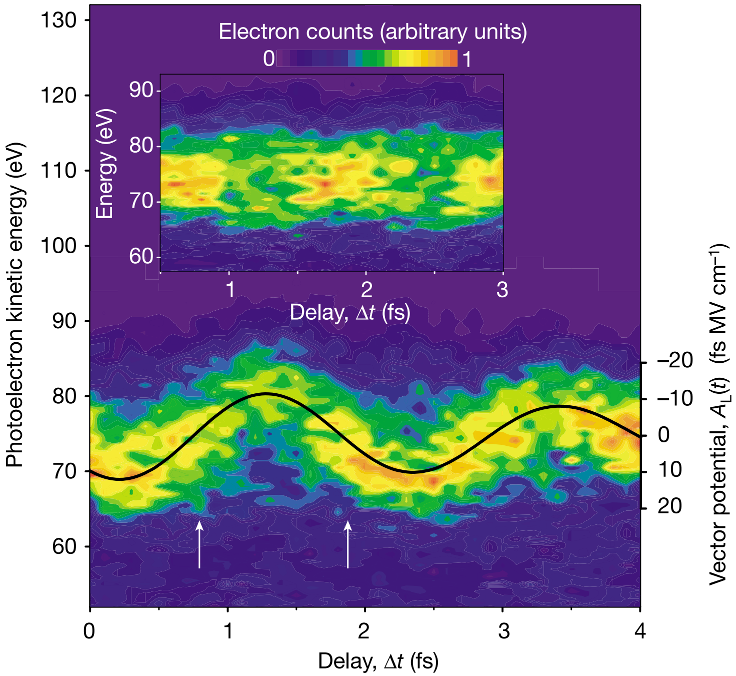
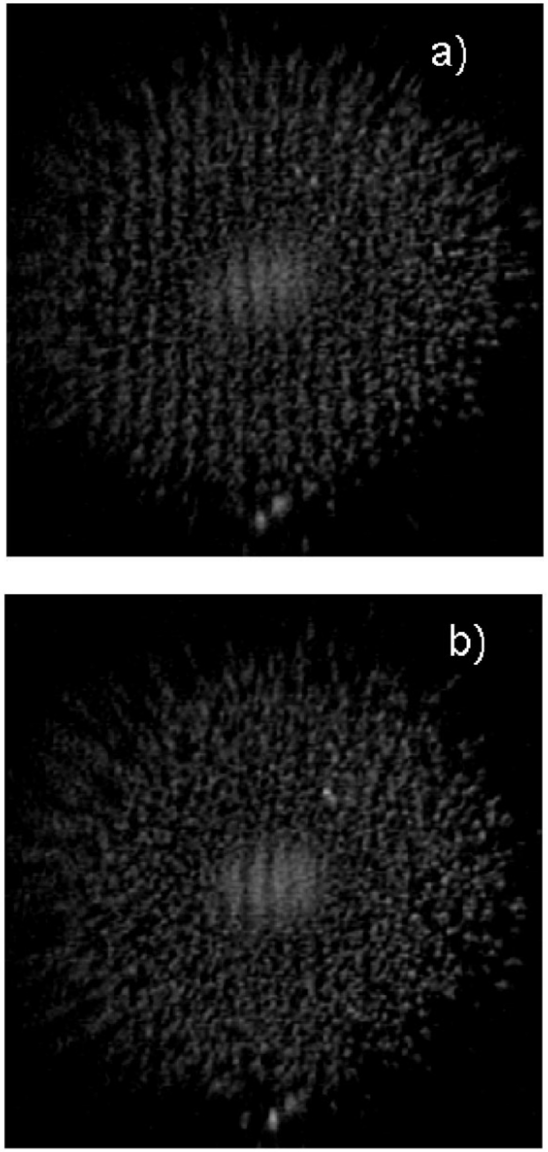
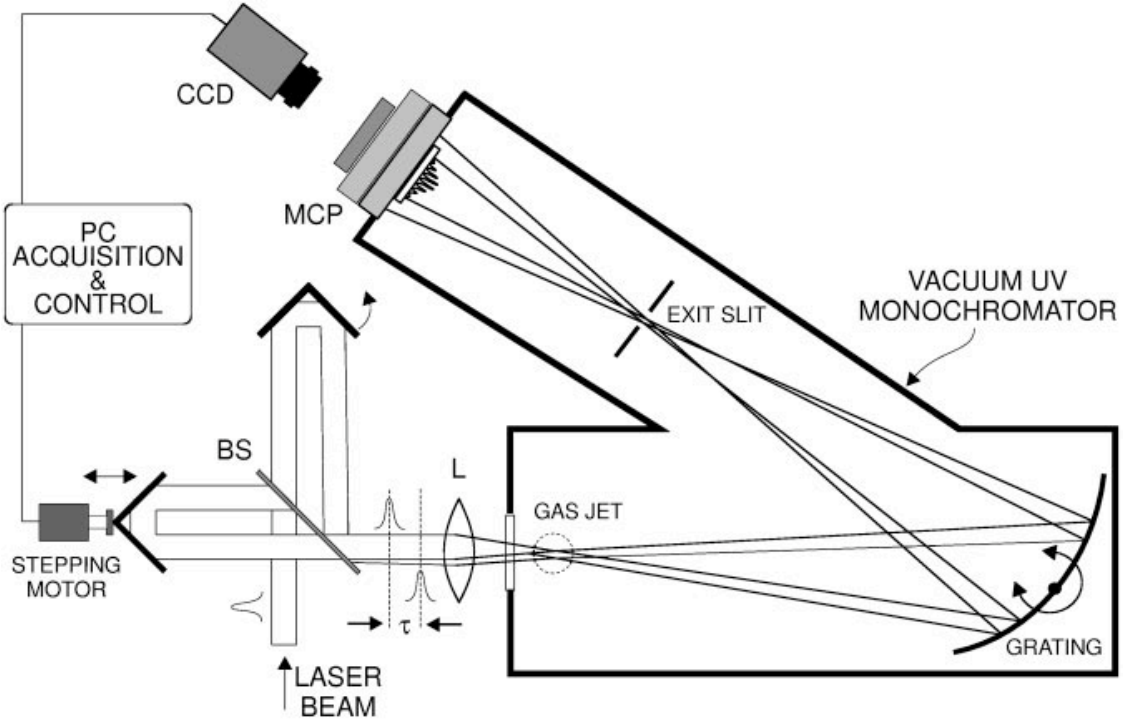


Figure 4 ATR measurement: a series of tomographic projections (streaked kinetic energy spectra) of the initial time–momentum distribution of photoelectrons knocked out by a single sub-fs XUV pulse (in false-colour representation). A few-cycle laser pulse with a cosine waveform and a normalized duration of $\tau_L/T_0 = 2$ was used for both generating the single 93-eV sub-fs excitation pulse and for probing photoelectron emission in the atomic transient recorder. Black line, $A_L(t)$ of the probing field evaluated from the peak shift of the streaked spectra (see scale on the right hand side). From $A_L(t)$ the electric field of the light wave can also be determined by using the relationship $E_L(t) = -\mathrm{d}A_L/\mathrm{d}t$. The electric field vector points towards the electron detector for $E_L > 0$. Inset, streaked spectra obtained under the same experimental conditions except for the carrier-envelope phase of the few-cycle pulse, which was left unstabilized.

High-Order Harmonic Generation (HHG)





Early work established that High Harmonics are temporally coherent with a definite phase relation to the driver pulse, with longer coherence time for the short trajectories.

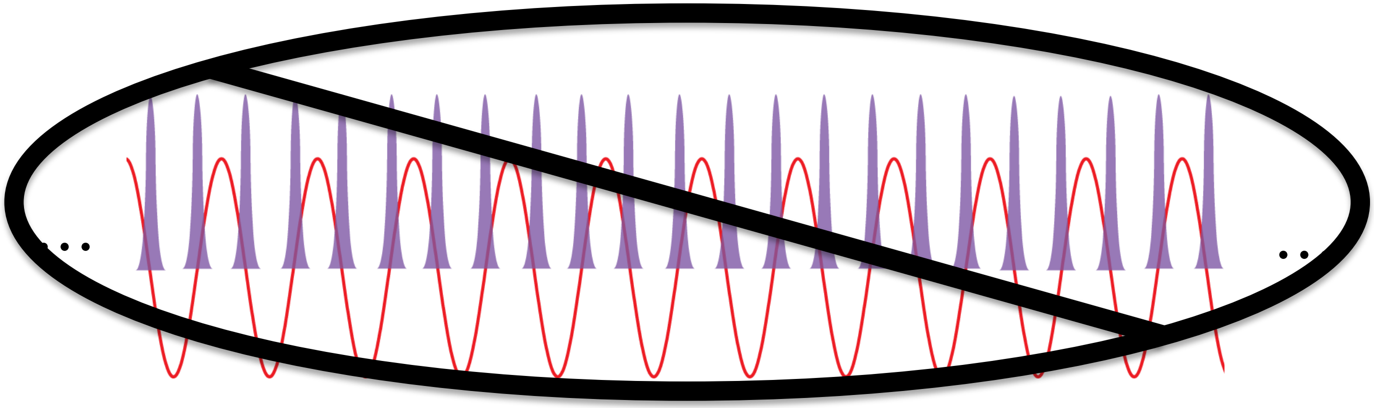
Bellini et. al. Phys. Rev. Lett. **81** 297 (1998)

Continous HHG?

$$\Delta\nu = \frac{1}{\pi\tau_c} \Rightarrow$$

- A narrow linewidth light source must be on for a long time.
- State of the art CW lasers can have $\Delta\nu < 40$ mHz ($\tau_c \sim 10$ s)

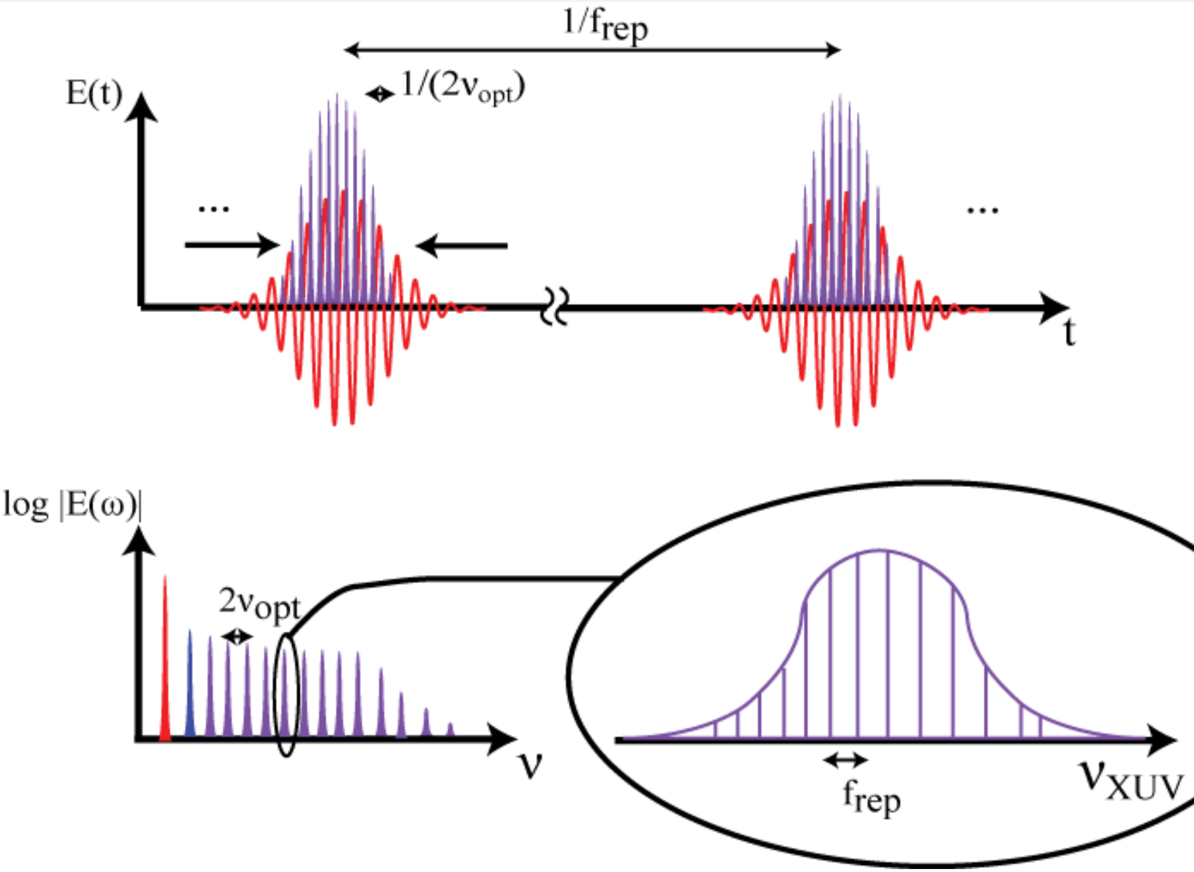
Why not just do HHG with a very stable CW laser?



- Depletion of the medium through ionization.
- GigaWatt average drive laser power.

⇒ HHG intrinsically needs ultrashort pulses.

XUV Frequency Comb

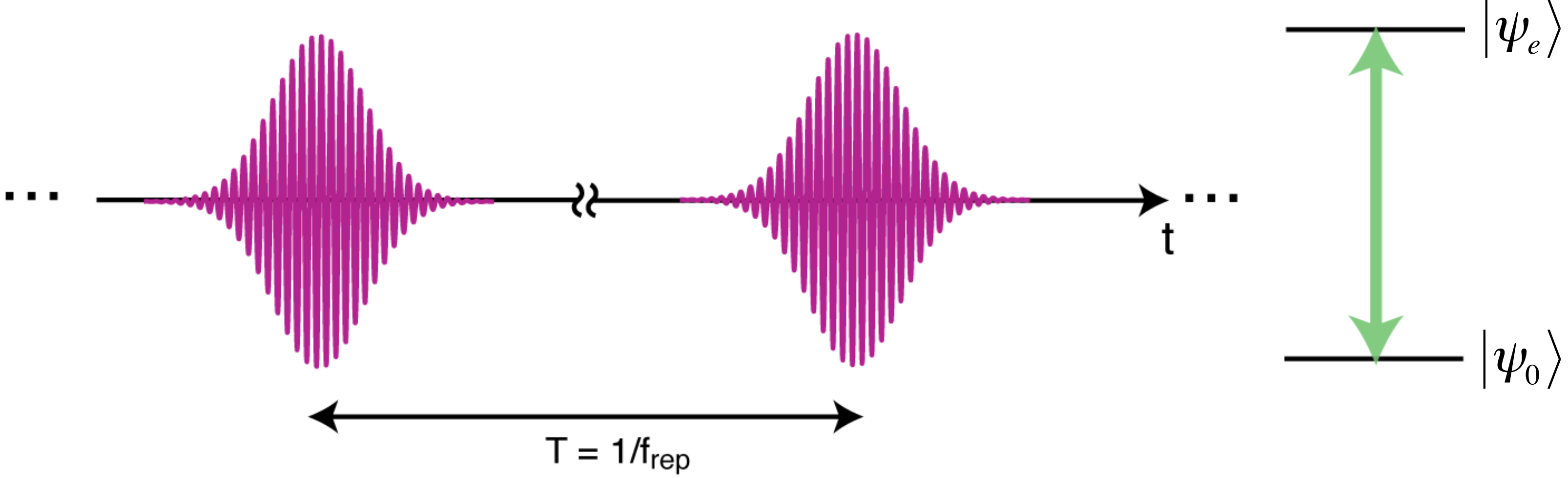


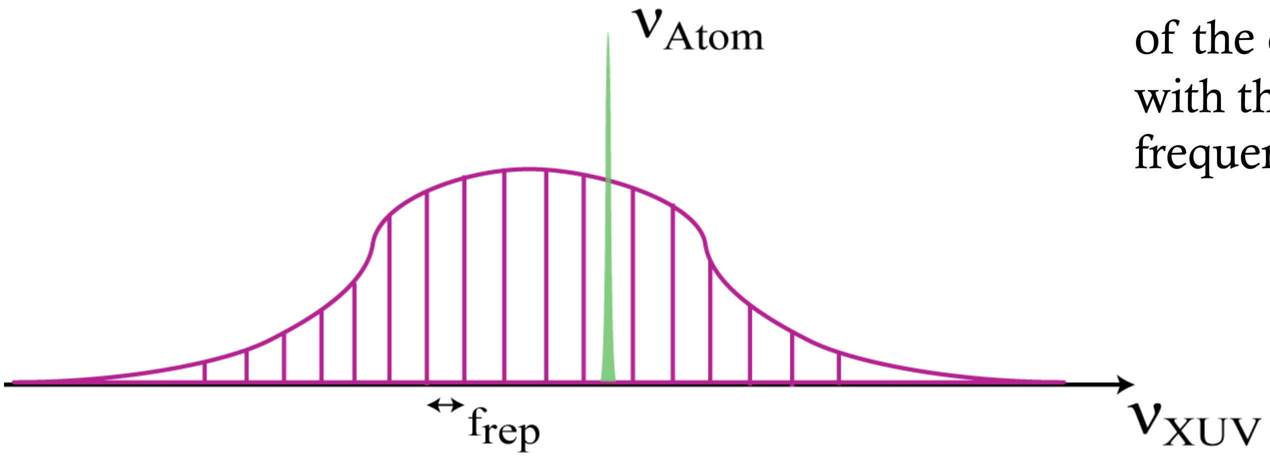
$$\nu_{IR} = nf_{rep} + f_0$$

$$\nu_{XUV} = mf_{rep} + qf_0$$

Direct Frequency Comb Spectroscopy

$$\langle \psi_e | \psi(t) \rangle \approx e^{-i(\Delta E/\hbar)t} + e^{-i[(\Delta E/\hbar)(t-T)+q\Delta\phi]} + e^{-i[(\Delta E/\hbar)(t-2T)+2q\Delta\phi]} + \cdots$$



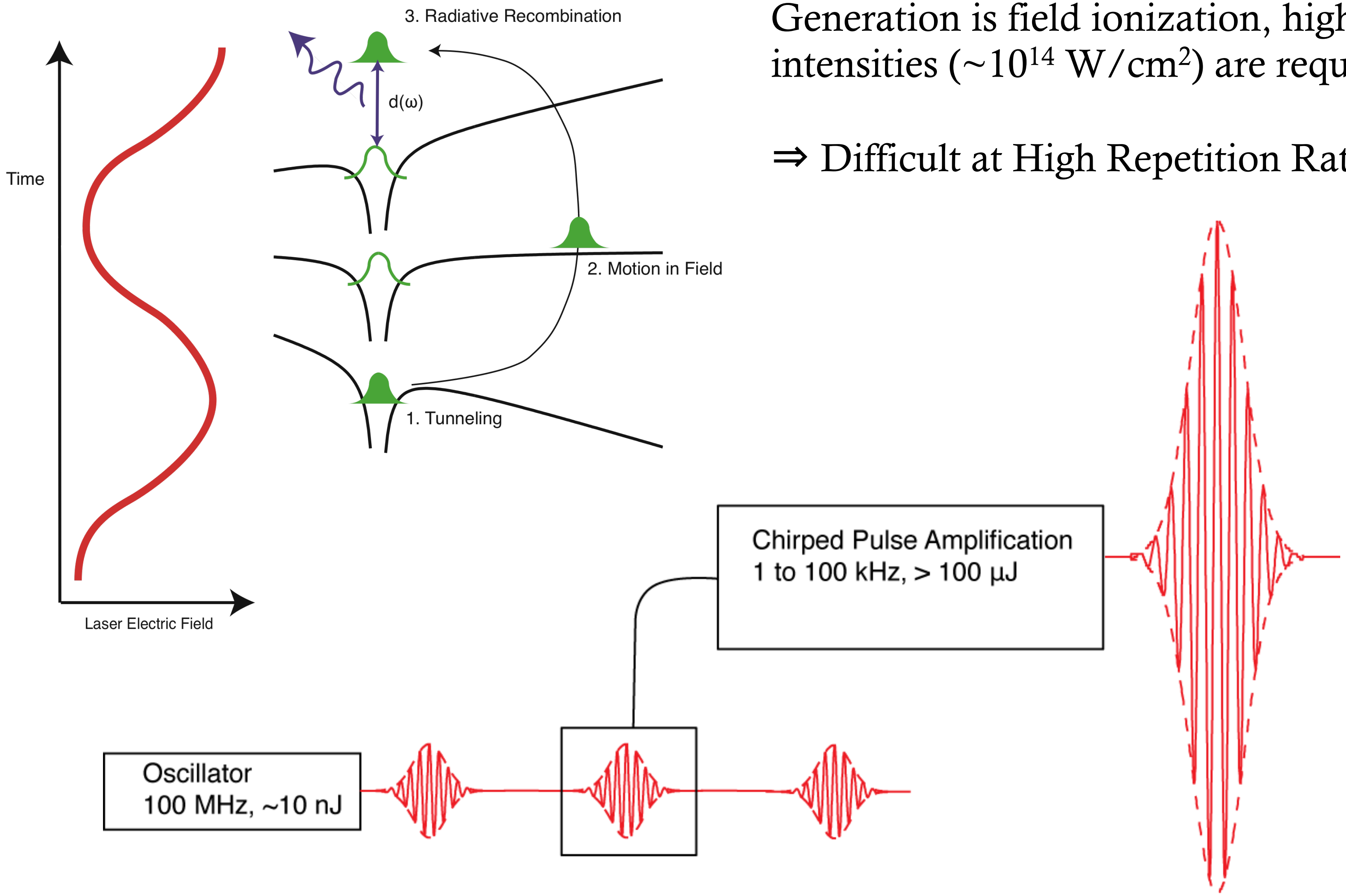


- Resonance occurs when one of the comb lines coincides with the atomic transition frequency.

Amplified Systems?

Since the first step in High Order Harmonic Generation is field ionization, high intensities (~10^{14} W/cm^2) are required.

⇒ Difficult at High Repetition Rate



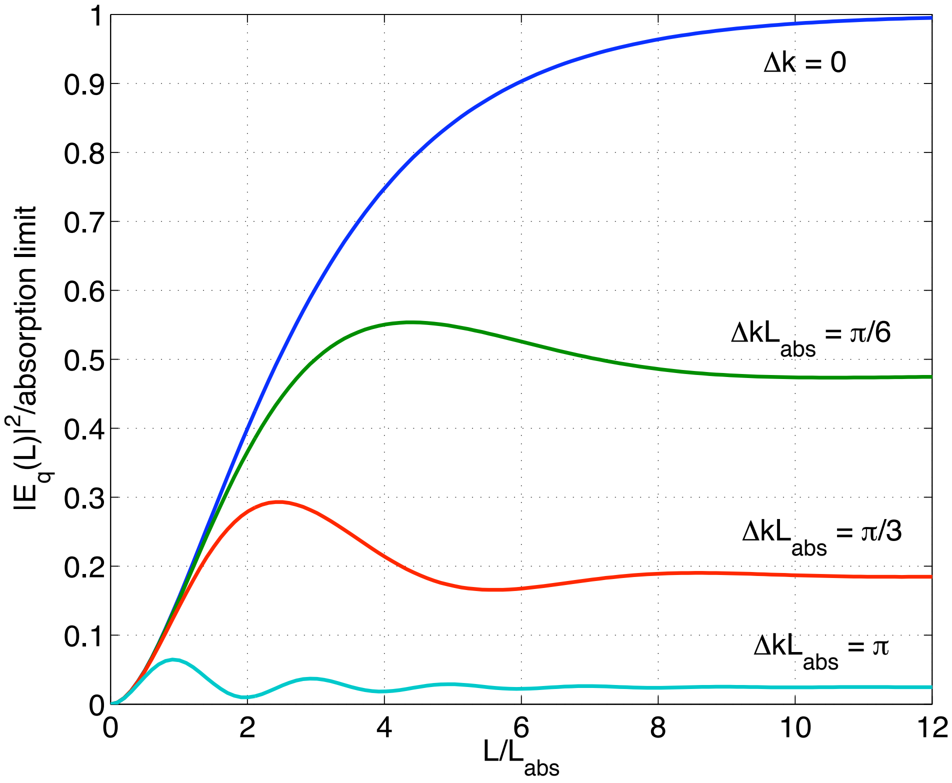
Focus Tighter?

- Single atom response is weak, so you must coherently add the signals from many atoms in order to get a strong signal, i.e. phase matching.

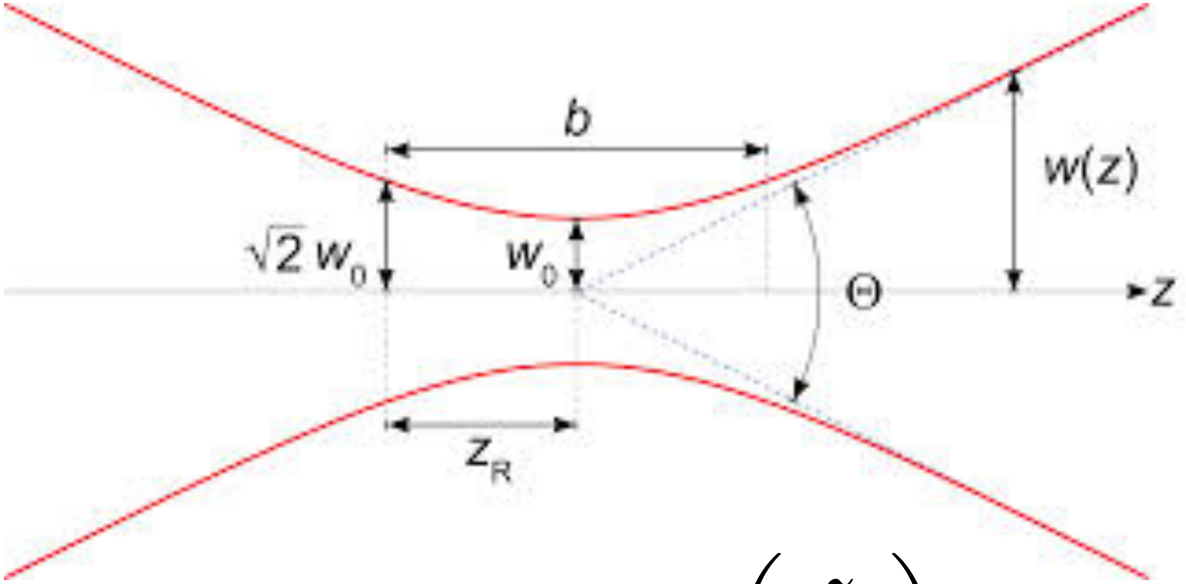
$$\Delta k = qk_1 - k_q$$

Due to…

- Gouy phase
- Neutral dispersion
- Plasma
- Intensity depend phase


1D Phase Matching model with absorption

Gouy Phase



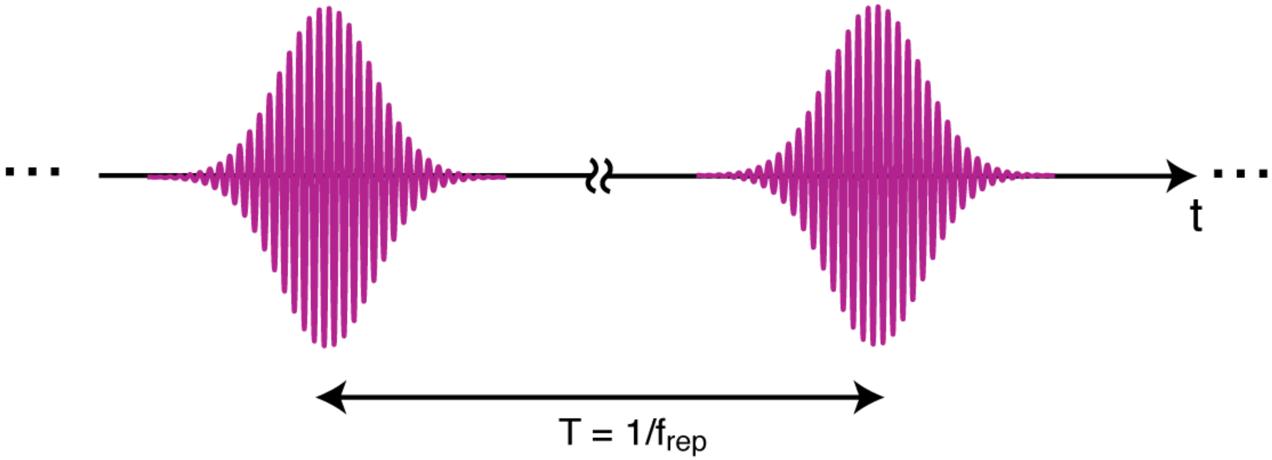
$$\phi(z) = -\tan^{-1}\left(\frac{z}{z_R}\right)$$

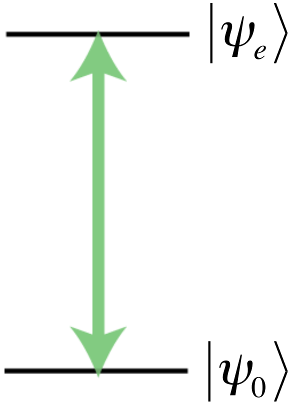
z_R for the harmonics is much larger than the fundamental…

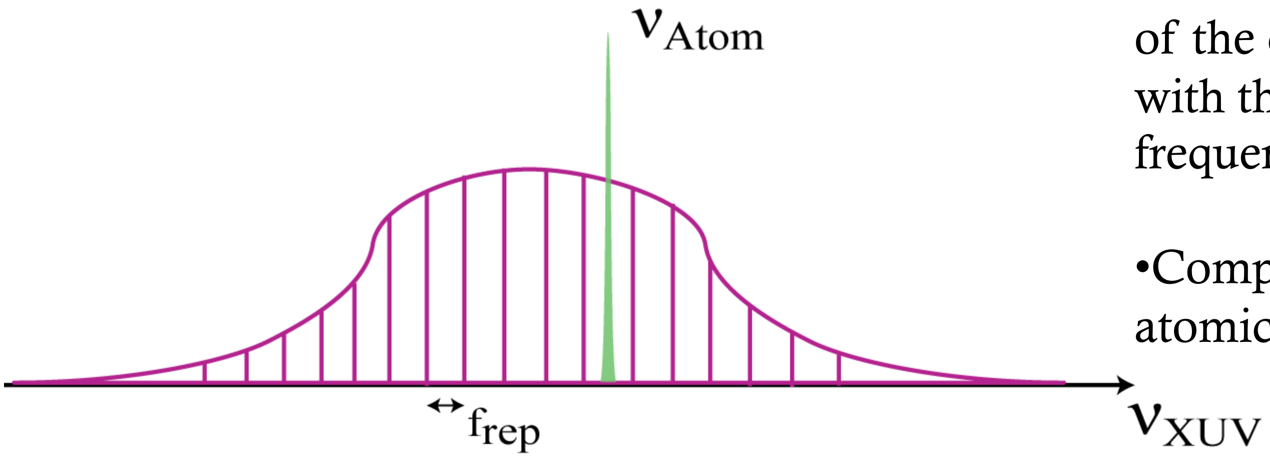
⇒ Phase mismatch in very tight focusing. Should have medium substantially smaller than the Rayleigh range

Direct Frequency Comb Spectroscopy

$$\left\langle \psi_e \middle| \psi(t) \right\rangle \approx e^{-i(\Delta E/\hbar)t} + e^{-i\left[(\Delta E/\hbar)(t-T)+q\Delta\phi\right]} + e^{-i\left[(\Delta E/\hbar)(t-2T)+2q\Delta\phi\right]} + \cdots$$







- Resonance occurs when one of the comb lines coincides with the atomic transition frequency.

- Compare comb linewidth to atomic linewidth.

The Challenge

Doing HHG with high enough repetition rate, and in an extremely controlled way, to be a useful XUV Comb.

Solutions?

Wait till *Part 2*