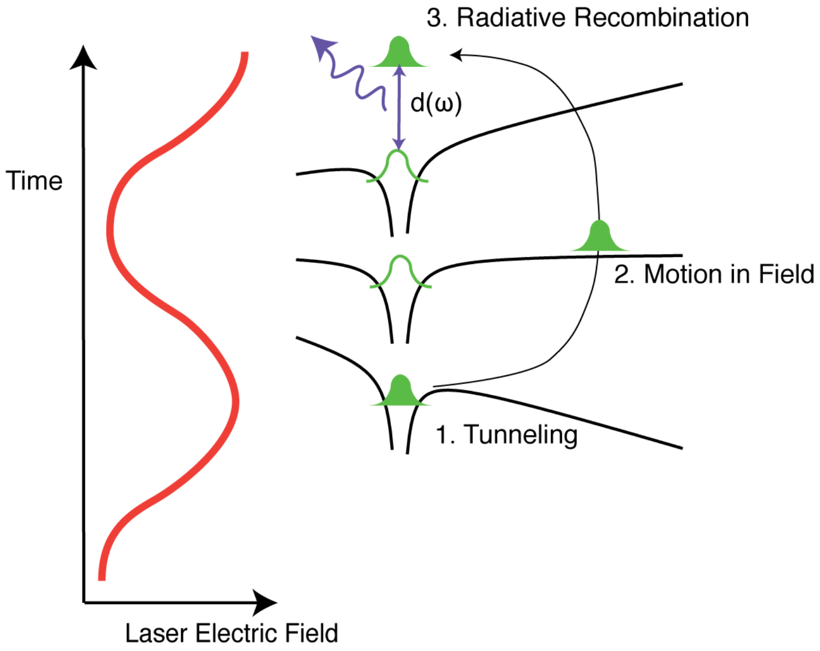
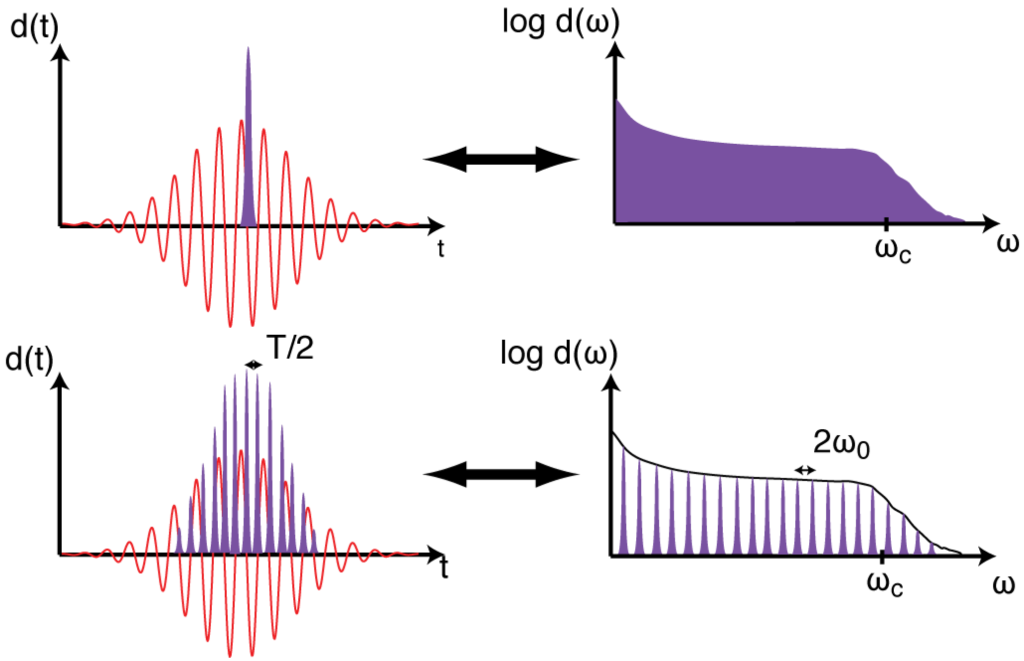
Extreme Ultraviolet Frequency Combs: Principles and Applications: *Part 2*

Thomas K. Allison
Stony Brook University

ICTP Winter College on Optics
Feb. 18, 2016

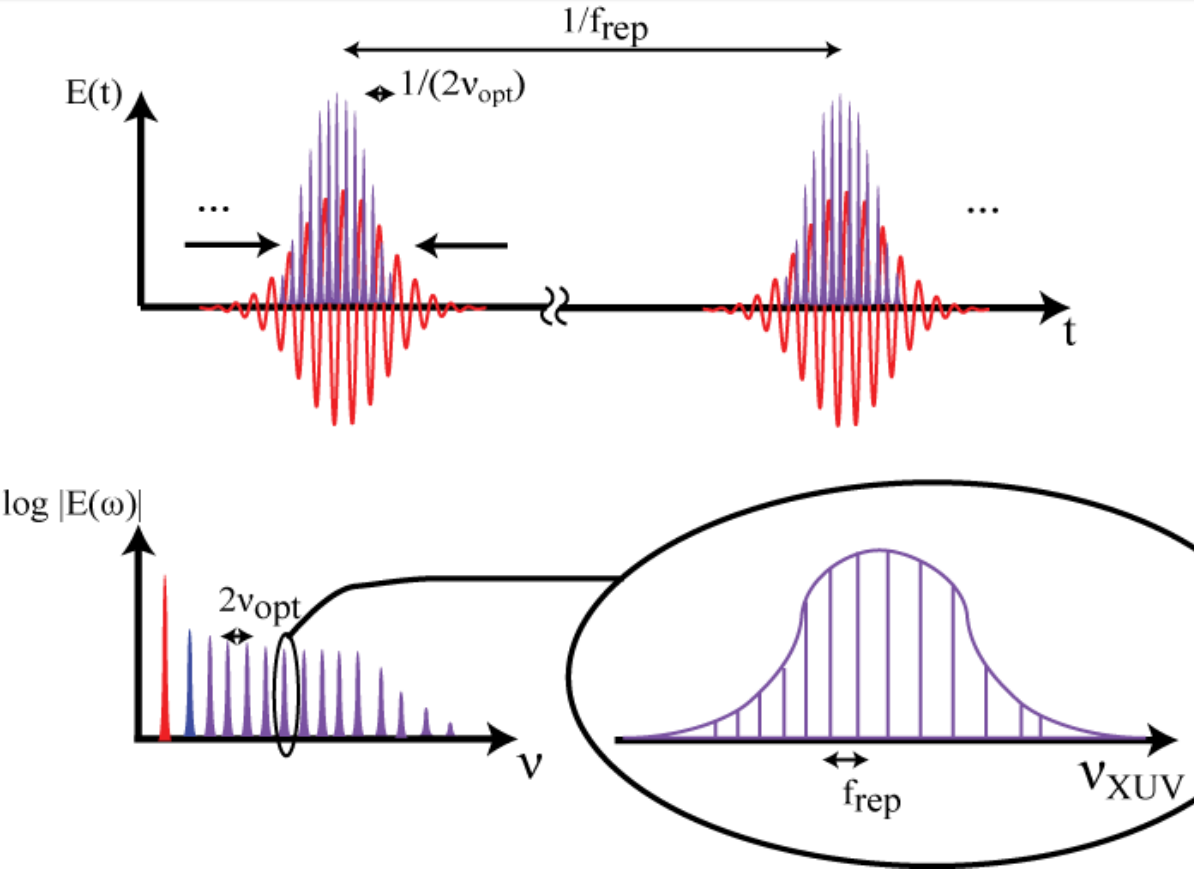
High-Order Harmonic Generation (HHG)





- 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.

XUV Frequency Comb

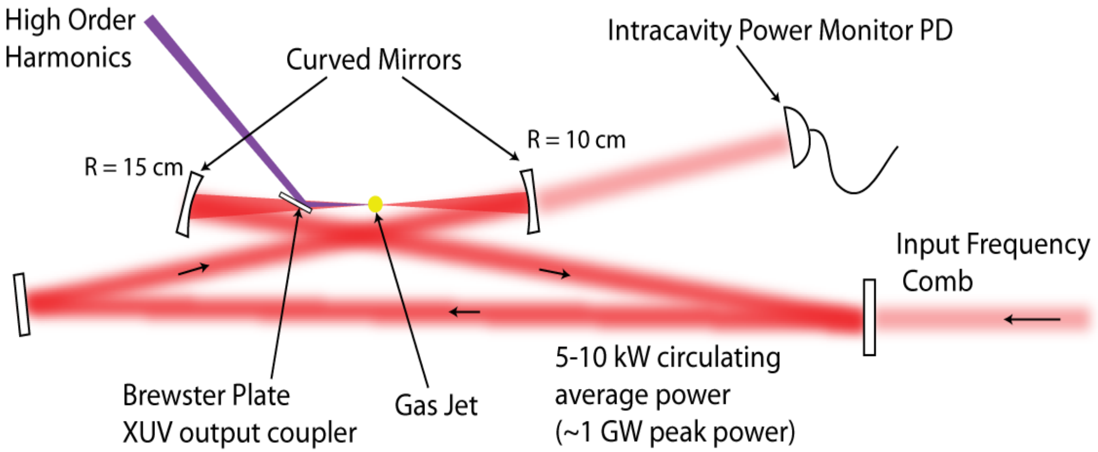


$$\nu_{IR} = nf_{rep} + f_0$$

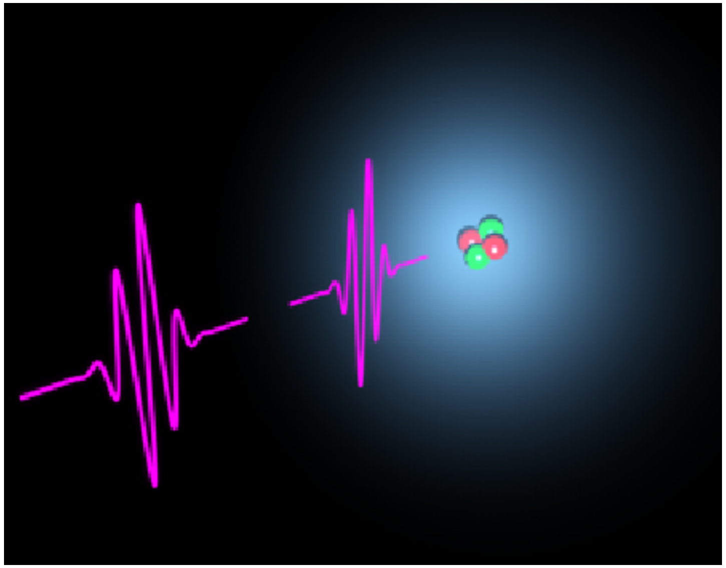
$$\nu_{XUV} = mf_{rep} + qf_0$$

Two Approaches

1. Cavity-Enhanced HHG



2. Two-Pulse “Ramsey-Comb”

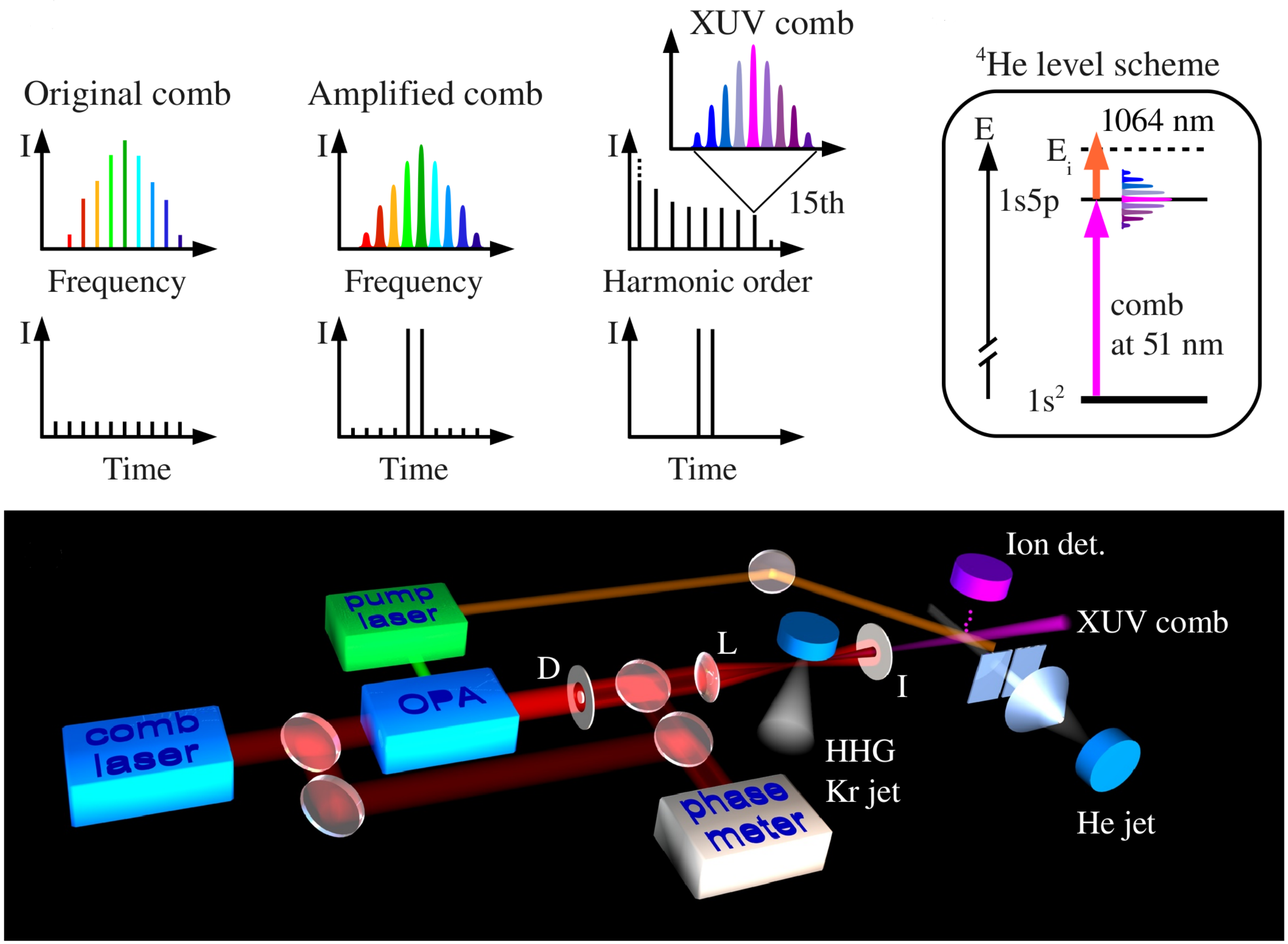


PRL **105,** 063001 (2010) PHYSICAL REVIEW LETTERS week ending 6 AUGUST 2010

Extreme Ultraviolet Frequency Comb Metrology

Dominik Z. Kandula, Christoph Gohle,* Tjeerd J. Pinkert, Wim Ubachs, and Kjeld S. E. Eikema†
Institute for Lasers, Life and Biophotonics Amsterdam, VU University, De Boelelaan 1081, 1081HV Amsterdam, The Netherlands

(Received 28 April 2010; revised manuscript received 24 June 2010; published 2 August 2010)



A physical delay line in the pump laser provided the required two pump pulses

PRL **105,** 063001 (2010) PHYSICAL REVIEW LETTERS week ending 6 AUGUST 2010

Extreme Ultraviolet Frequency Comb Metrology

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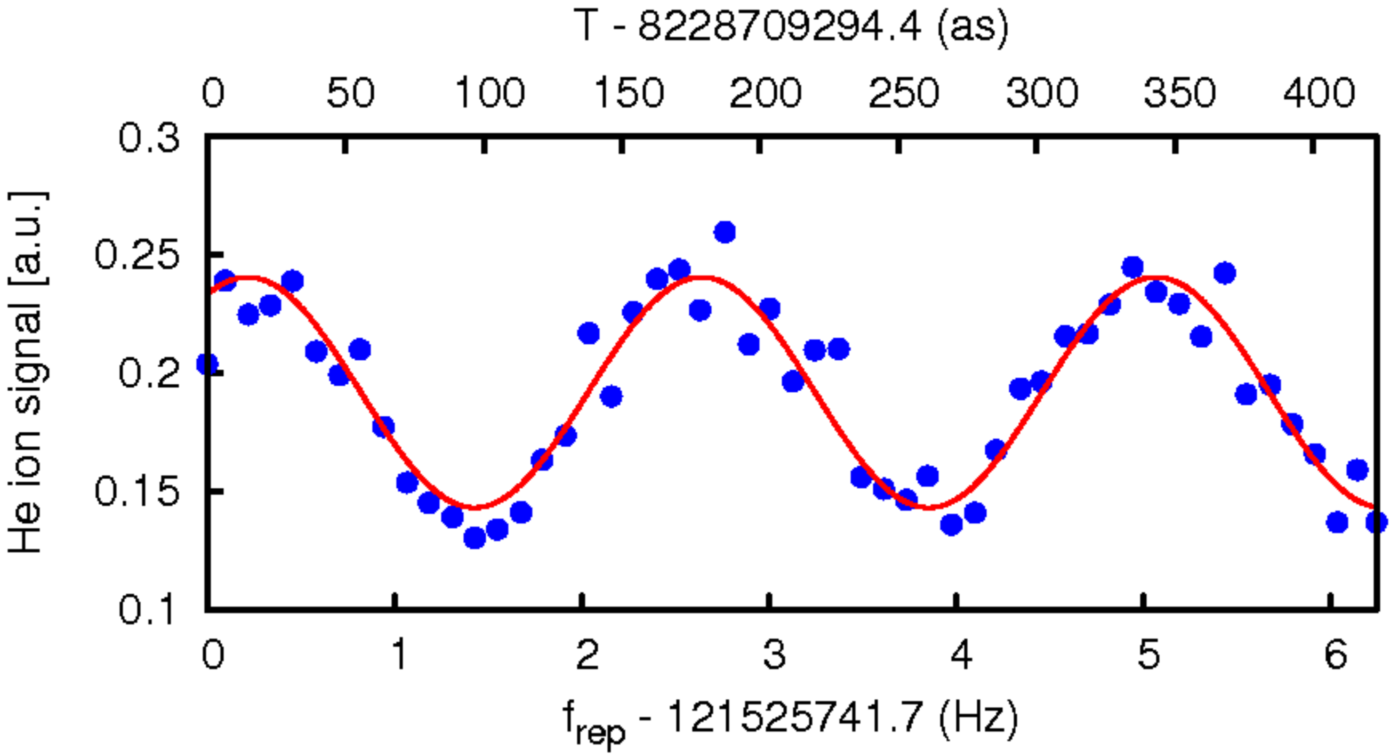
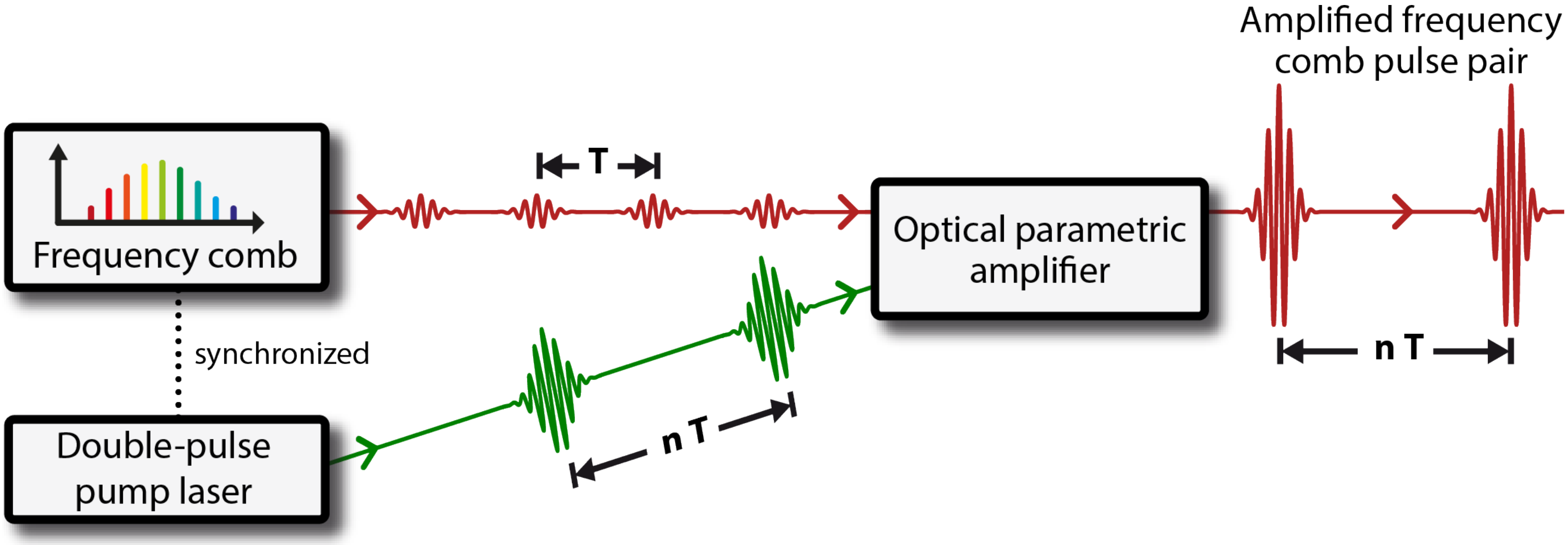


Excitation with 2 amplified and upconverted FC pulses.
Accuracy 6 MHz at 51 nm

D. Kandula et al. PRL **105**, 063001 (2010)
D. Kandula et al. PRA **84**, 062512 (2011)

Dominik Kandula

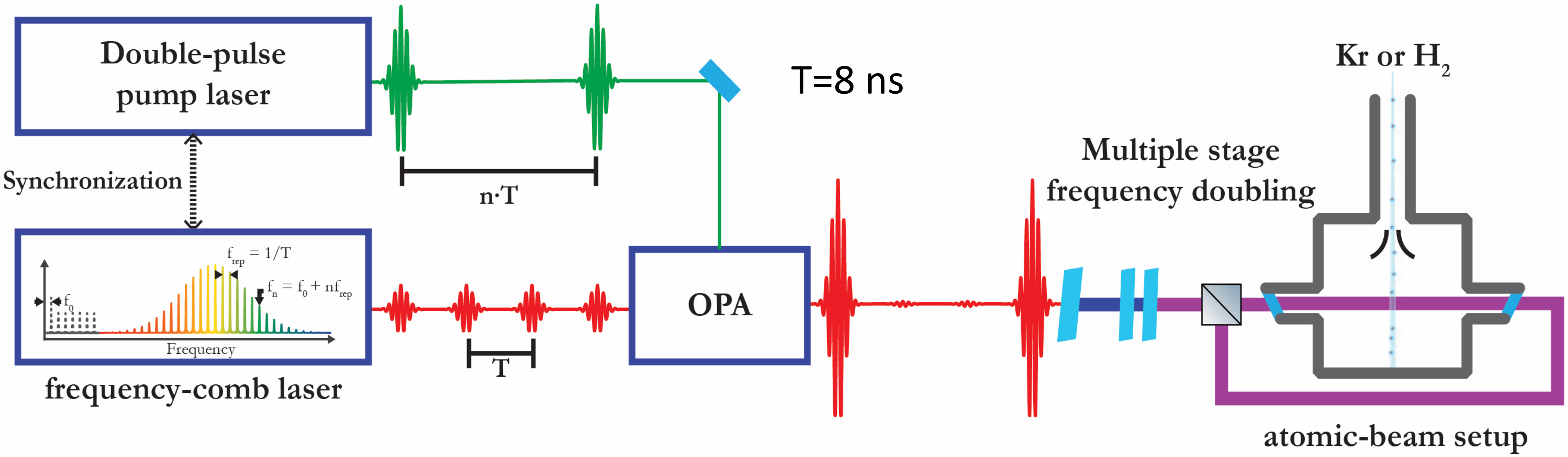
Christoph Gohle



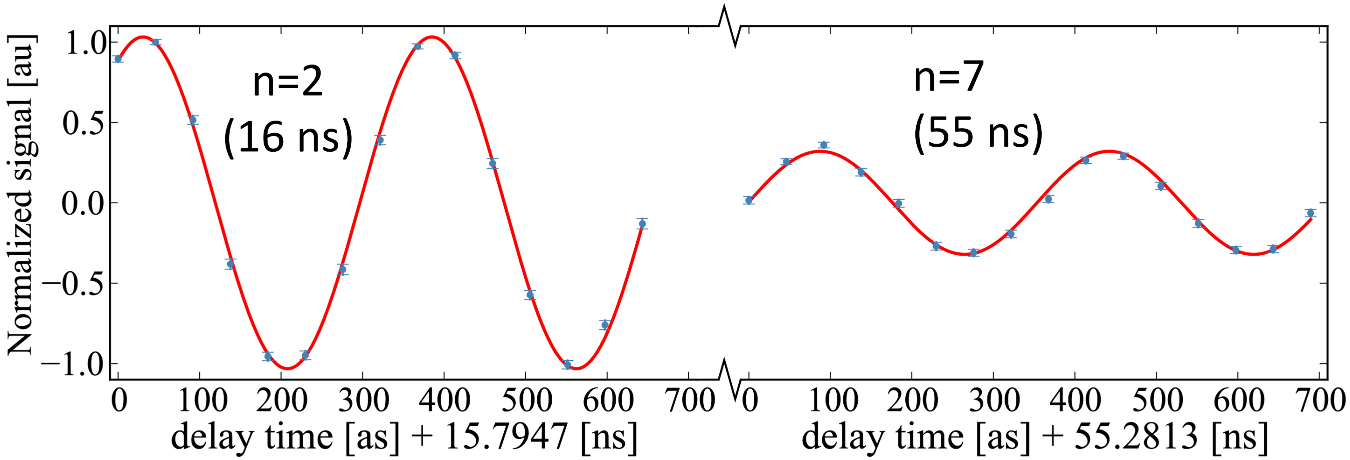
New pump laser: pulse-picking and a high-gain amplifier.
Output: amplified comb pulses 5 mJ, phase control ~ few mrad
Enables kHz-level or better accuracy by varying nT over ns to ms.

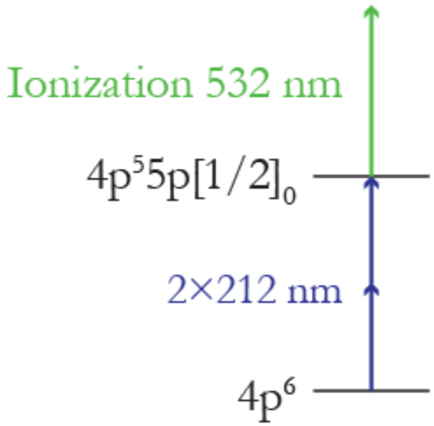
J. Morgenweg, K.S.E. Eikema, Laser Physics Letters **9**, 781-785 (2012)
J. Morgenweg, K.S.E. Eikema, Optics Express **21**, 5275-5286 (2013)
J. Morgenweg, I. Barmes, K.S.E. Eikema, Nature Physics **10**, 30-33 (2014)

- **Demonstrated at 760 nm and 212 nm. Outlook: via HHG also in the XUV (<100 nm)**

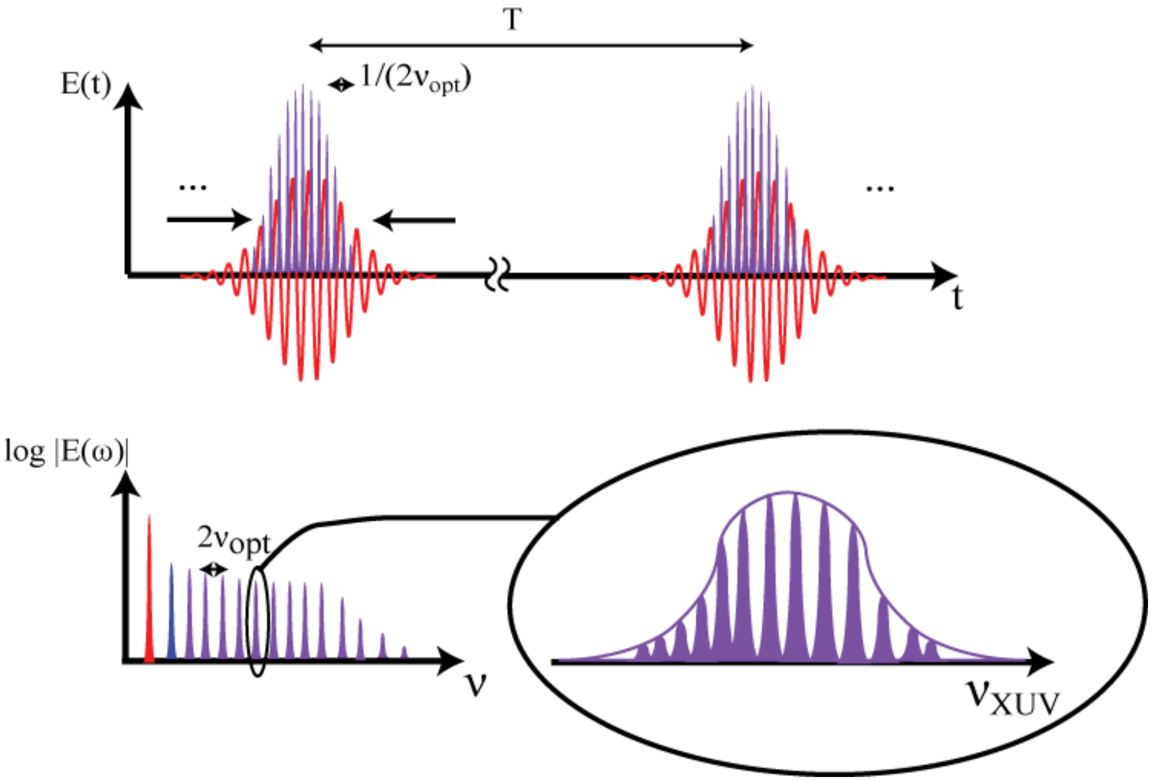


Ramsey-comb signal in Kr at 212 nm gives f=2820833101684(100) kHz: **35x improved**!

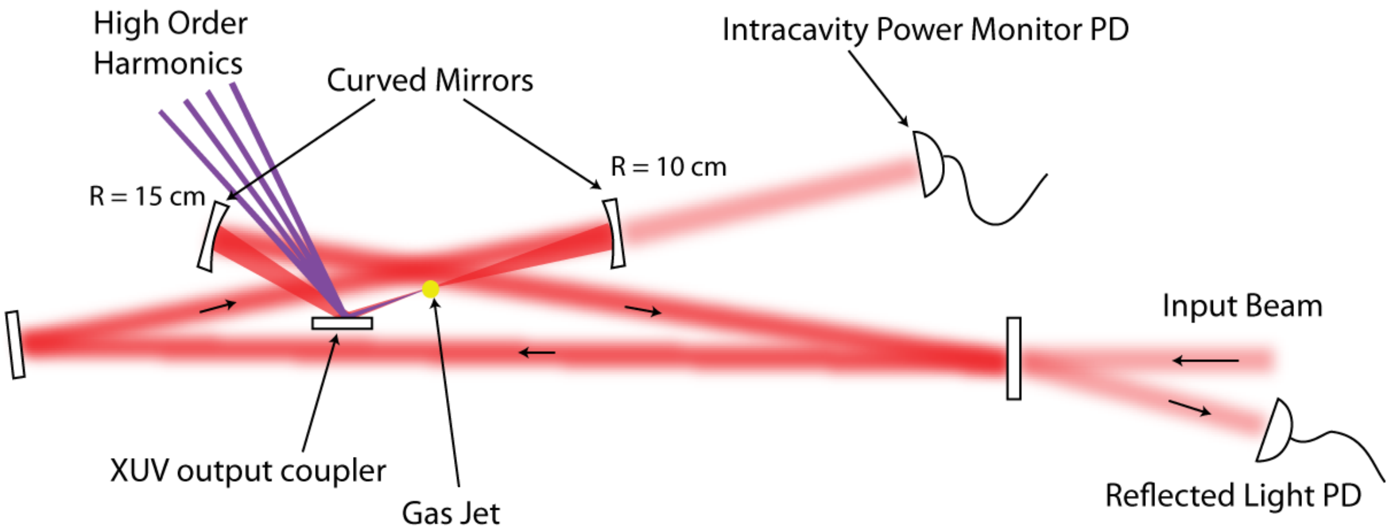




Phase Shifts



- Differential Phase shifts between are the main problem…Not in “steady state”
- Generally easier to measure frequency than phase.
- Power per “comb mode” decreases as resolution increases.
- But it is easily tunable, and could work well for two-photon spectroscopy



First Demonstrations:

<u>Garching</u>: C. Gohle et al. *Nature* **436**, 237 (2005)
<u>JILA</u>: R. J. Jones et al. *Phys. Rev. Lett.* **94**, 193201 (2005)
} ~100 pW/harmonic

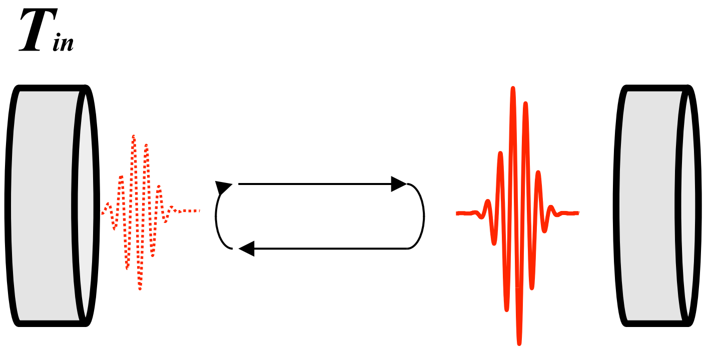
Since then, we have improved the power x10^6!

<u>JILA</u>:	T. K. Allison et al. *Phys. Rev. Lett.* **107**, 183903 (2011)	
	A. Cingöz et al. *Nature* **482**, 68 (2012)	
	D. C. Yost et al., *Opt. Exp.* **19**, 23483 (2011)	
<u>U. of Arizona</u>:	D. R. Carlson et al., *Opt. Lett.* **36**, 2991 (2011)	
	J. Lee et al., *Opt. Exp.* **19**, 23315 (2011)	>100 μW/harmonic
<u>UBC:</u>	Hammond et al. *Opt. Exp.* **19**, 24871 (2011)	
<u>MPQ Garching:</u>	I. Pupeza et al. *Nat. Phot.* **7**, 608 (2013)	
	I. Pupeza et al. *Phys. Rev. Lett.* ***112,*** 103902 (2014)	

Recent Review: A. K. Mills et al., *J. Phys. B.* **45**, 142001 (2012)

Coherent Accumulation in Buildup Cavity

Time Domain

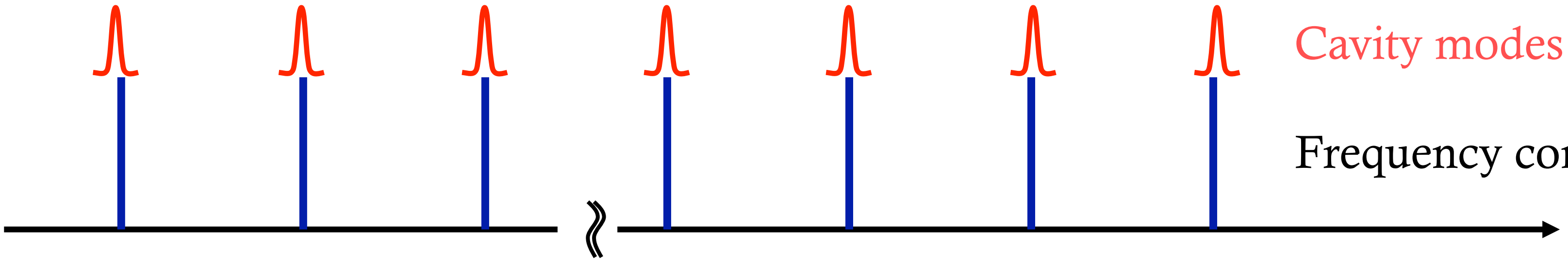


Cavity enhancement:

$$N = \frac{4T_{in}}{L^2} = 4T_{in}\left(\frac{F}{2\pi}\right)^2$$

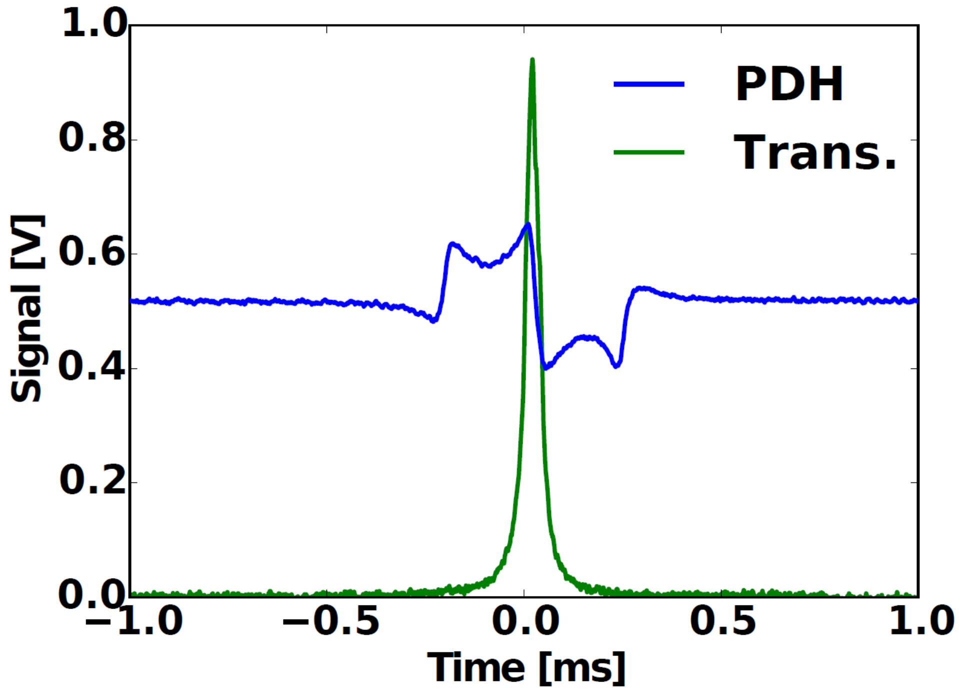
Finesses of > 1000 with cavity enhancements of several hundred are possible with bandwidth $\Delta\,\lambda \sim 30$ nm

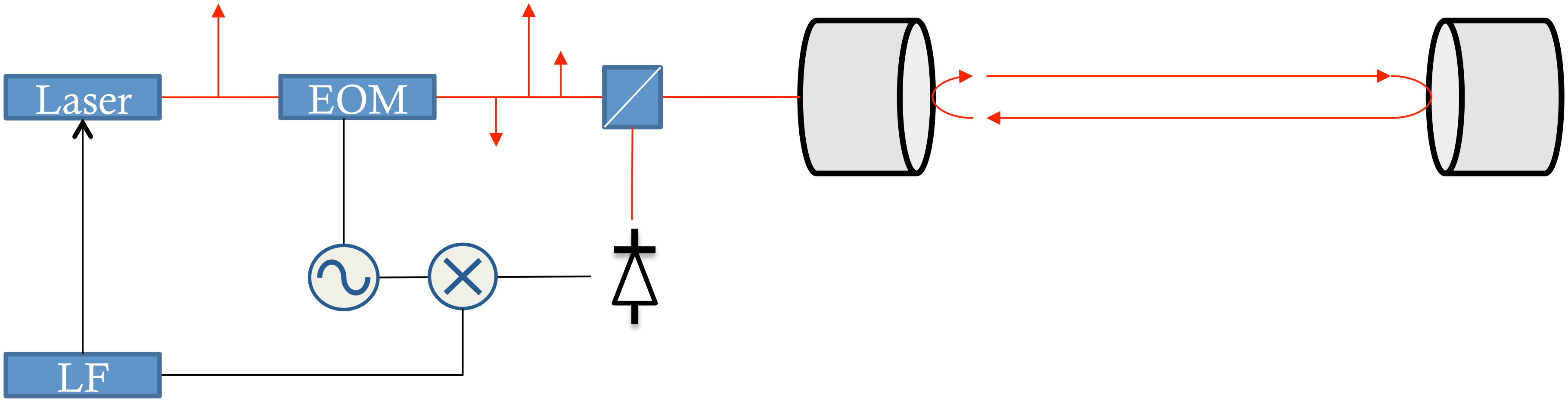
Frequency Domain



⇒ 10s of Watts can give kW intracavity power!
Signals from repetitive experiments can be added coherently!

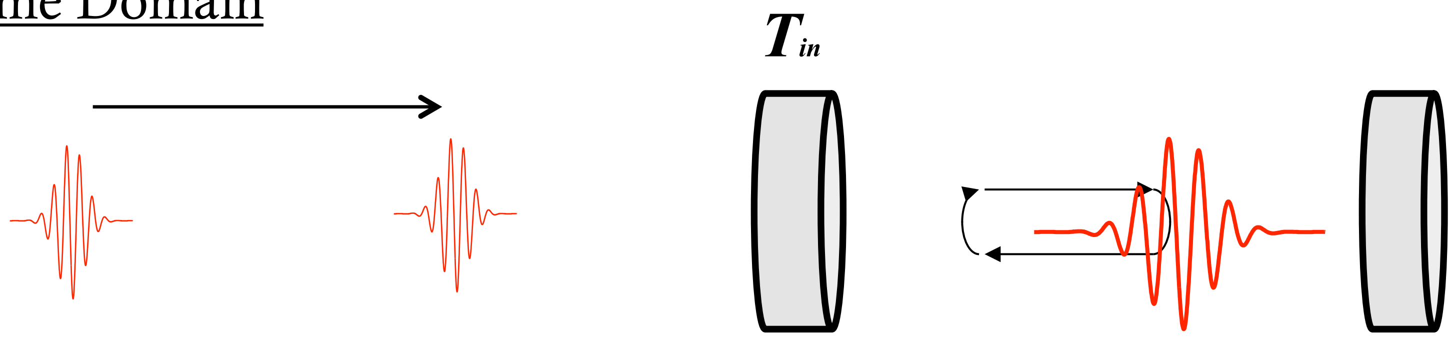
Pound-Drever-Hall Locking





Intracavity Dispersion

Time Domain



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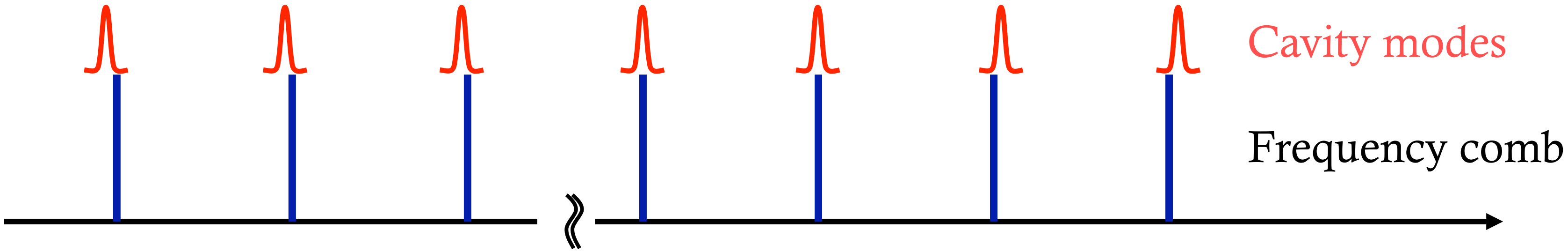
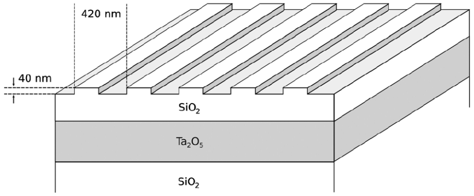
Frequency Domain



⇒ 10s of Watts can give kW intracavity power!
Signals from repetitive experiments can be added coherently!

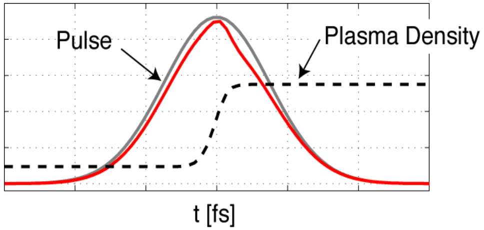
Challenges for Intracavity HHG

multi-kW average power.
⇒ Severe constraints on optics: High finesse, low dispersion, low nonlinearity, and high damage threshold

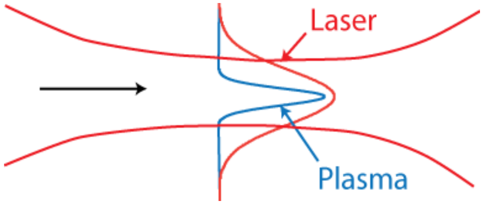


Need to get the harmonics out the cavity. No transparent materials in the XUV.

Plasma does not clear from focal region within one cavity round trip. Large steady-state plasma shifts the cavity resonance ⇒ Optical bistabillity.

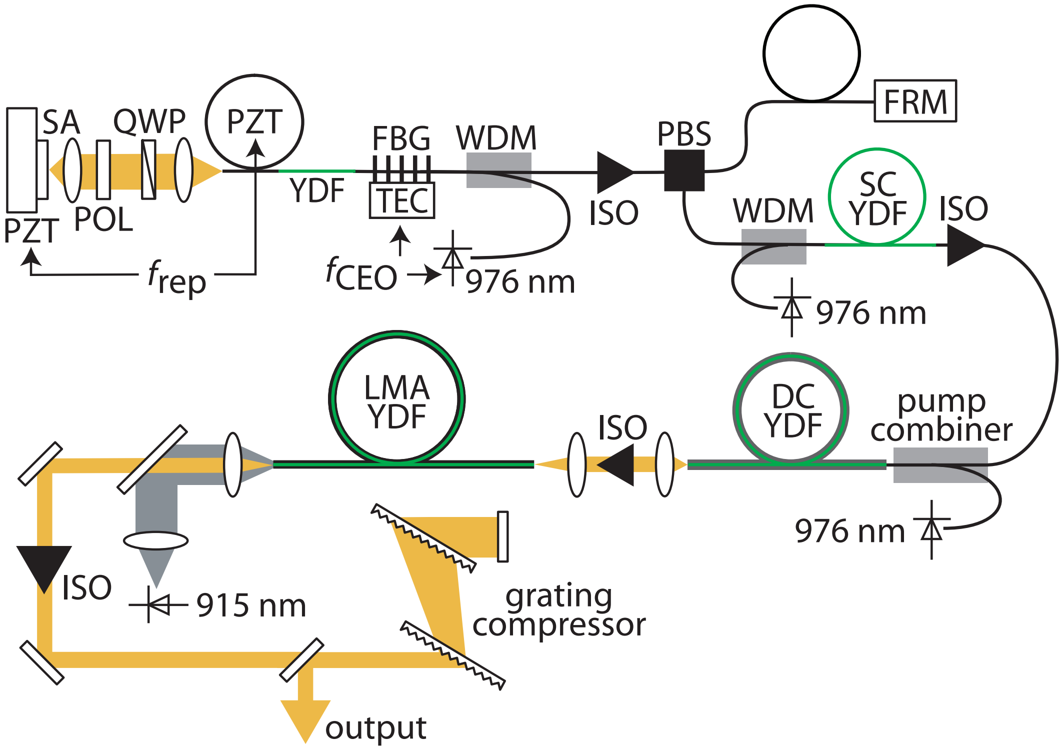


Index of refraction decreases during the pulse as the plasma density grows ⇒ Self phase modulation and blueshifting.



Plasma density is highest on axis and lower on the edges of the focus ⇒ plasma lensing, beam defocusing.

Yb:Fiber Lasers

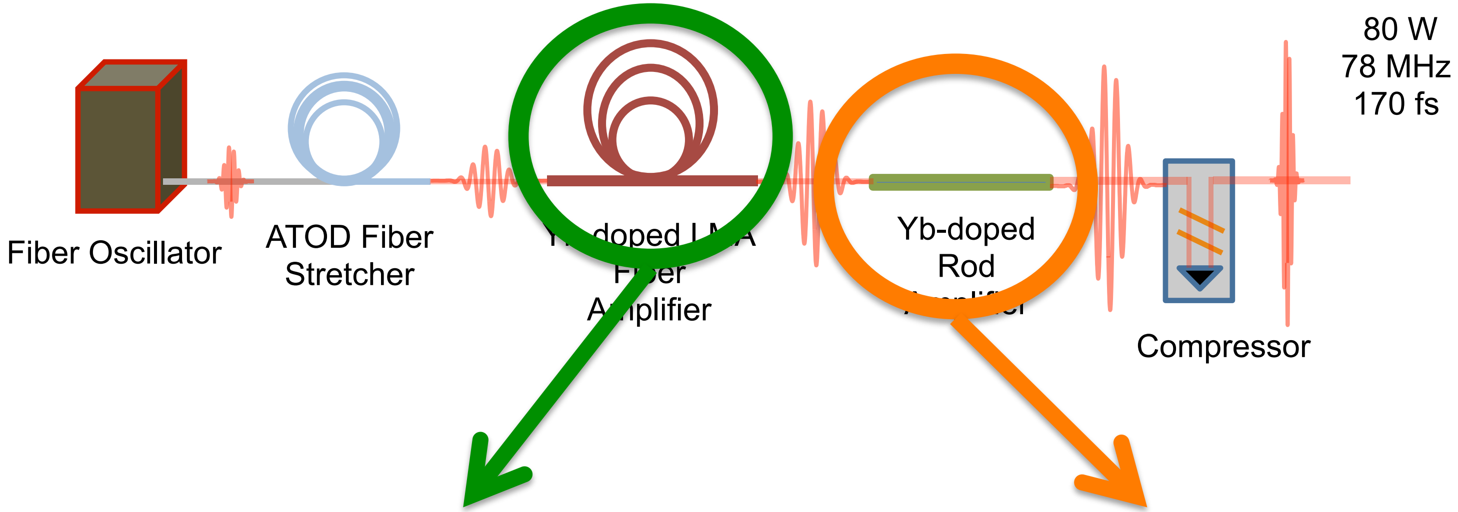


- 1070 nm central wavelength
- 154 MHz repetition rate
- 120 fs pulses
- 80 W average power (0.5 µJ/pulse)
- Very stable f_{ceo}
  - < 15 kHz free running linewidth
  - Very slow drift

Collaboration with IMRA America:
Axel Ruehl, Ingmar Hartl and Martin Fermann

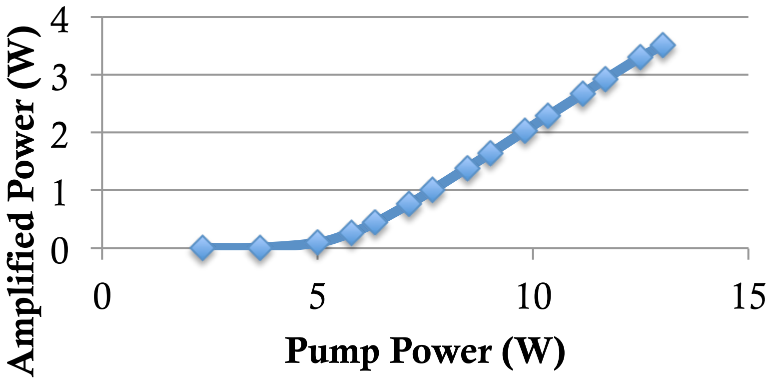
Hartl et al., *Opt. Lett.* **32** 2870 (2007)
Schibli et al., *Nat. Phot.* **2** 355 (2008)
Ruehl et al., *Opt. Lett.* **35** 3015 (2010)

Stony Brook High Power Yb:Fiber

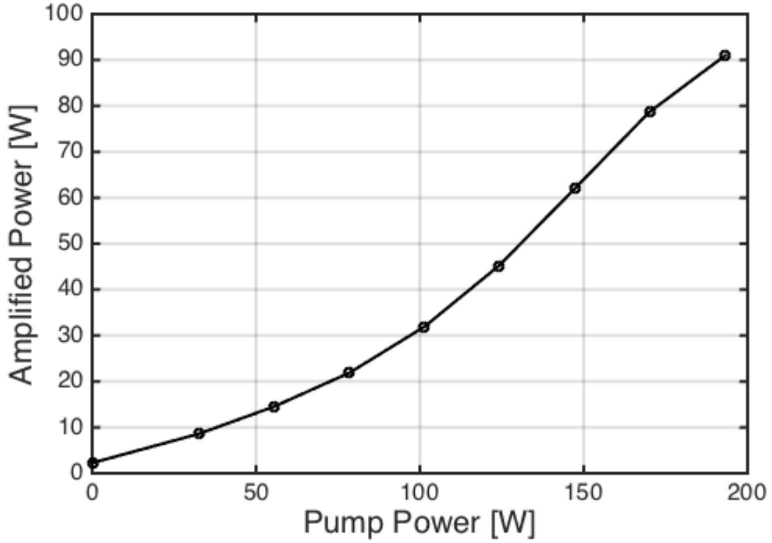


Xinlong Li
Phys. Grad.

Preamplifier (15 mW seed)



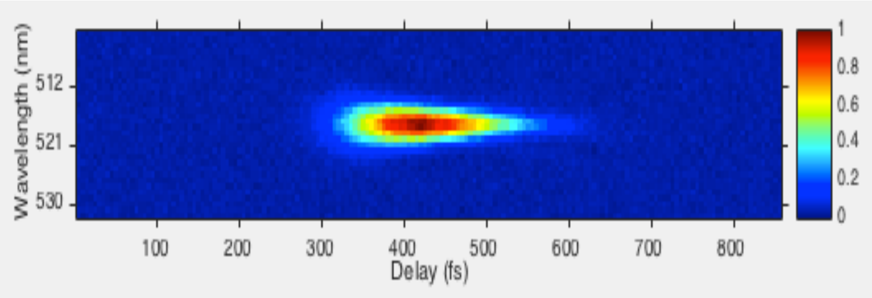
Power Amplifier (5 W seed)

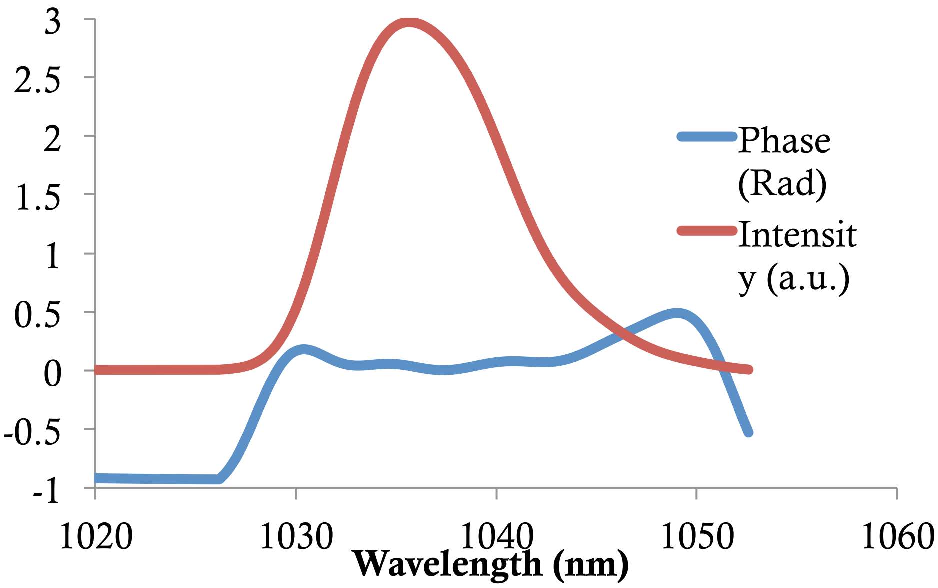


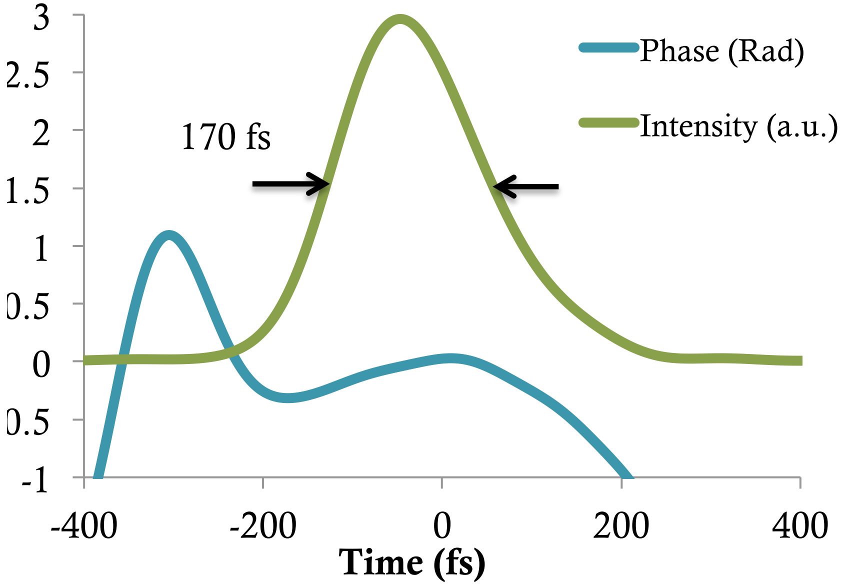
Pulse Duration

- Pulses compressed to very near the transform limit.
- FROG trace is roughly independent of power!

Raw FROG trace

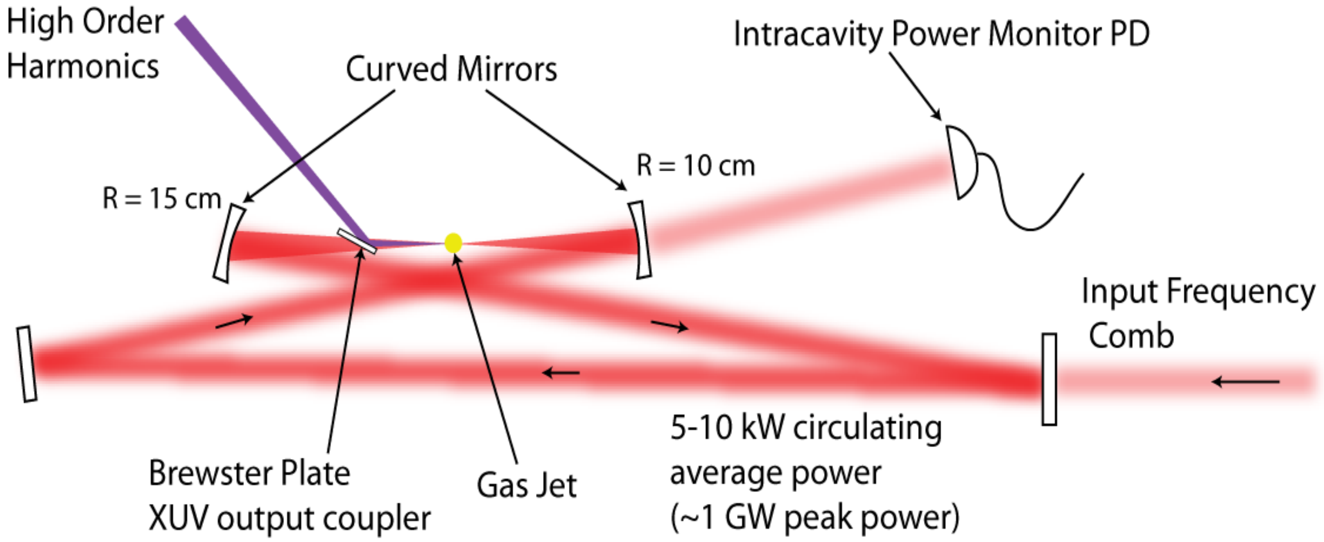






X. L. Li et al. In Preparation

Output Coupler



High Order Harmonics

Curved Mirrors

Intracavity Power Monitor PD

R = 15 cm

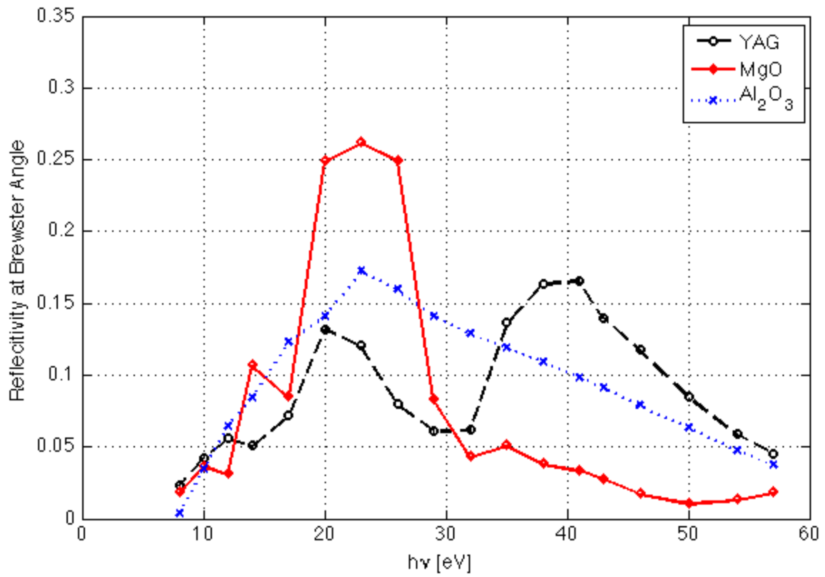
R = 10 cm

Input Frequency Comb

Brewster Plate XUV output coupler

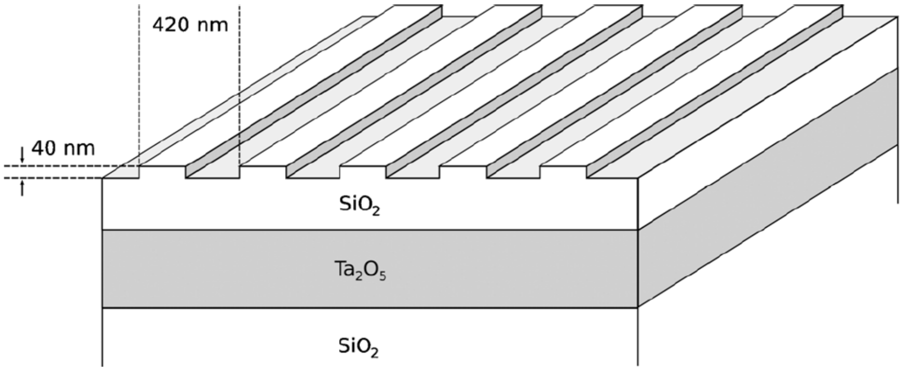
Gas Jet

5-10 kW circulating average power (~1 GW peak power)

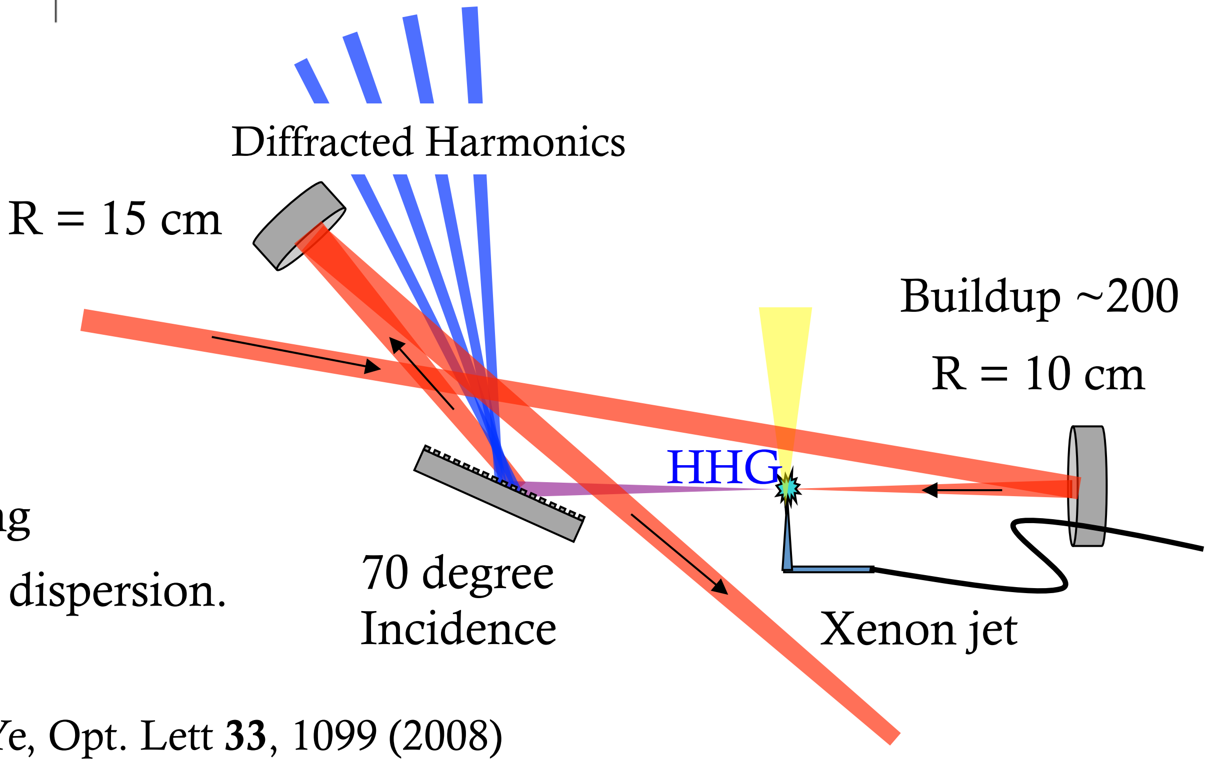


But, dispersion, nonlinearity, thermal lensing….

Output Coupler



Shallow Diffraction Grating ion beam etched into a grazing incidence high reflector



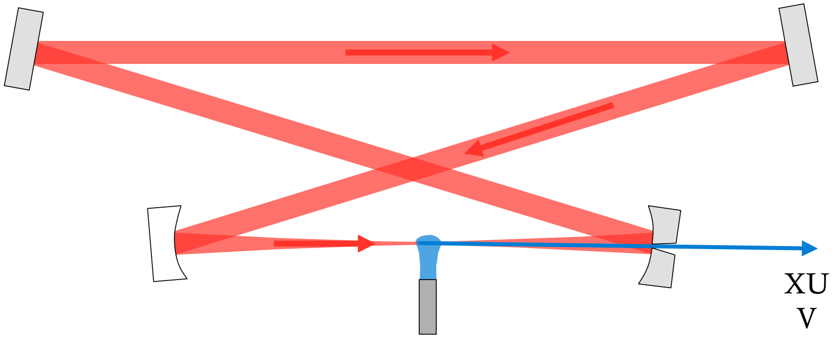
⇒ No self phase modulation

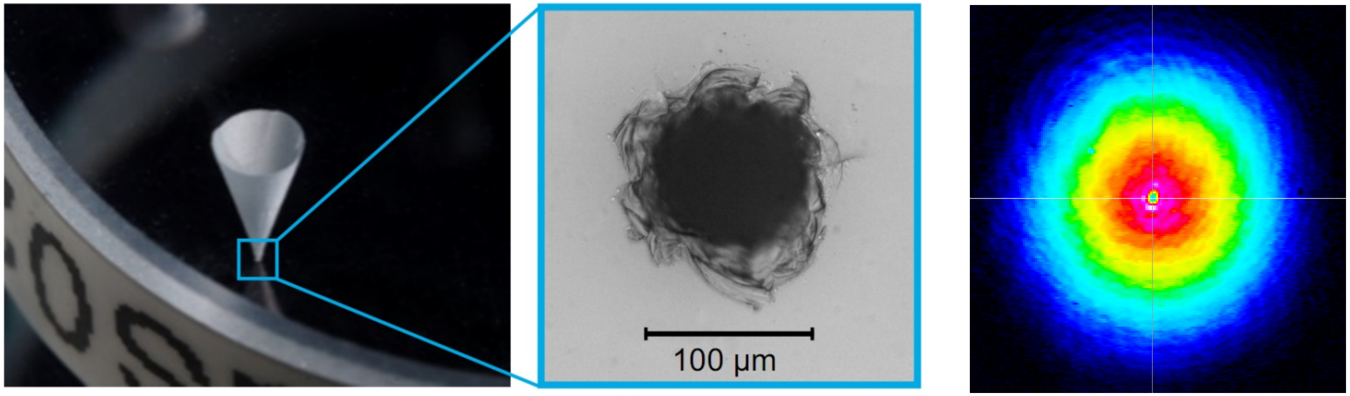
⇒ Roughly 10% output coupling

⇒Very low intracavity loss and dispersion.

D. C. Yost, T. R. Schibli, and Jun Ye, Opt. Lett **33**, 1099 (2008)

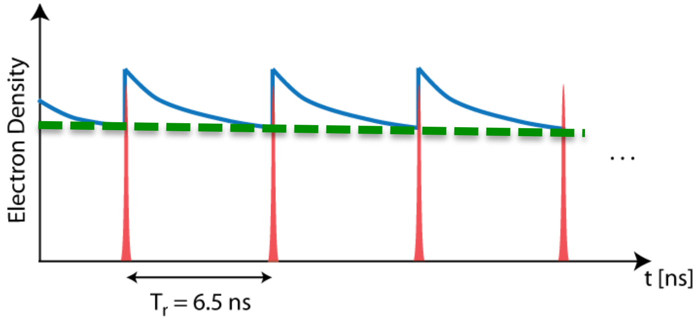
Geometric XUV Output Coupling





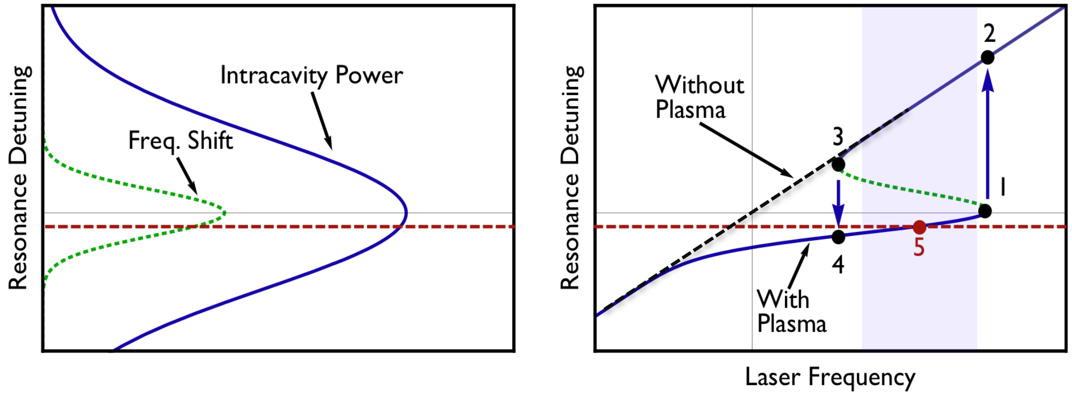
Pierced mirror: Esser et al., Opt. Express **21** (26797) 2013

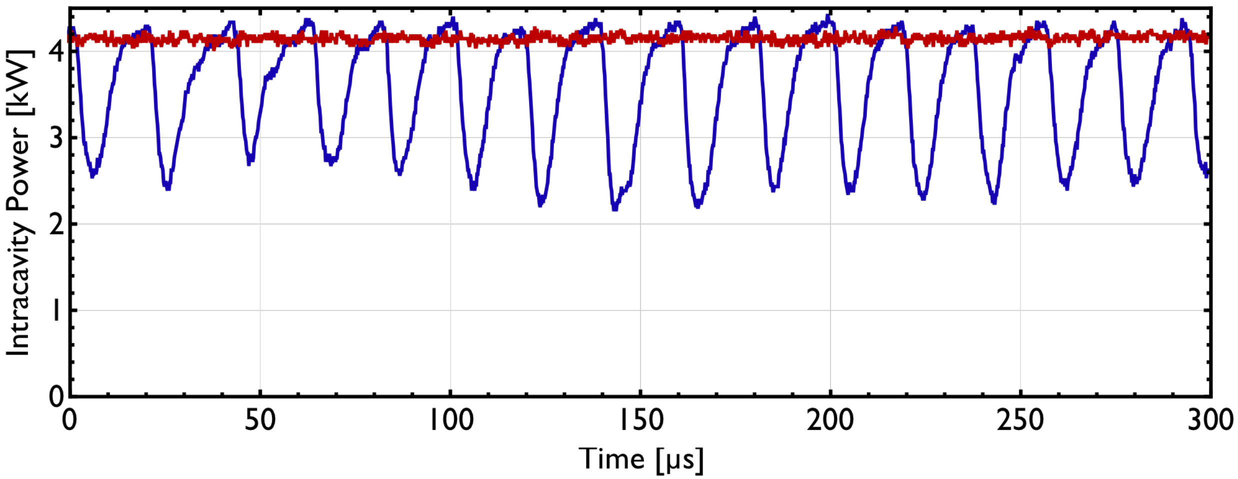
Optical Bistability



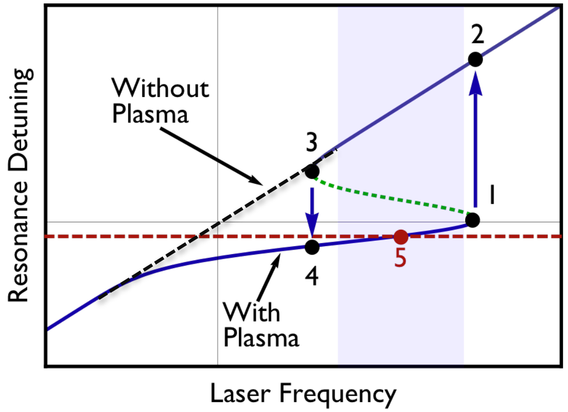
Plasma decay is fast compared to the characterstic time scale of the intracavity light ($F/\pi f_{rep}$ ~ 800 ns) ⇒ Steady-state plasma effectively tracks the intracavity power.

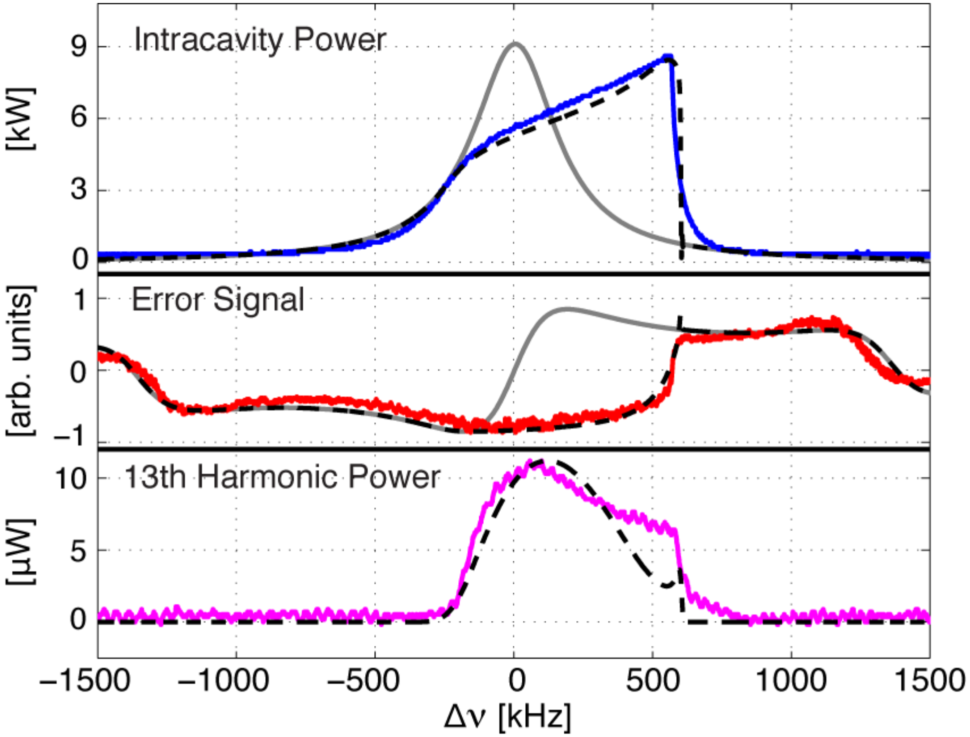
⇒ An intensity dependent phase shift





Self Locking



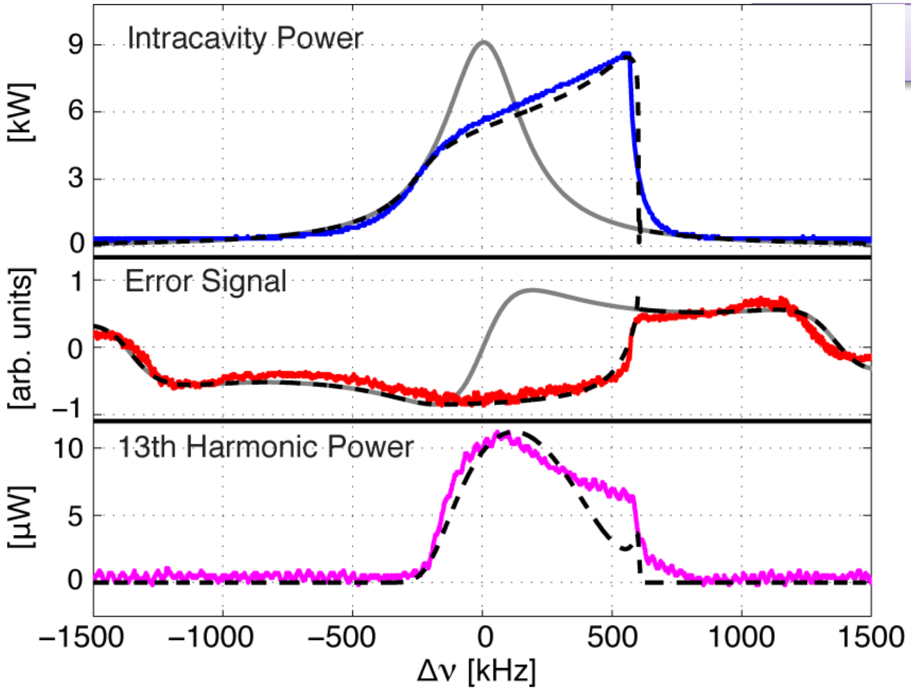


Scanning the cavity length across the resonance condition, the plasma compensates the detuning, and the system "self-locks"

T. K. Allison et al., *Phys. Rev. Lett.* **107**, 183903 (2011)
D. C. Yost et al., *Opt. Exp.* **19**, 23483 (2011)

Theory

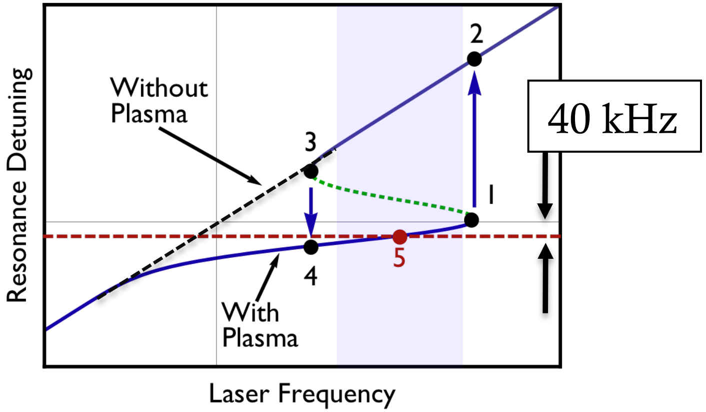
$$\frac{dA(\omega)}{dT} = \underbrace{\sqrt{\delta}E(\omega)}_{\text{Pumping Field}} \underbrace{-\frac{1}{2}(\delta+\gamma)A}_{\text{Cavity Losses}} + \underbrace{i\theta A}_{\text{Detuning}} + \underbrace{i\phi(A)A}_{\text{Steady State Plasma}} + \underbrace{f(A)}_{\text{SPM and Loss Dynamic Plasma}}$$



Black Dashed Lines are Model Results from sweeping θ across resonance!

T. K. Allison et al., *Phys. Rev. Lett.* **107**, 183903 (2011)
D. C. Yost et al., *Opt. Exp.* **19**, 23483 (2011)

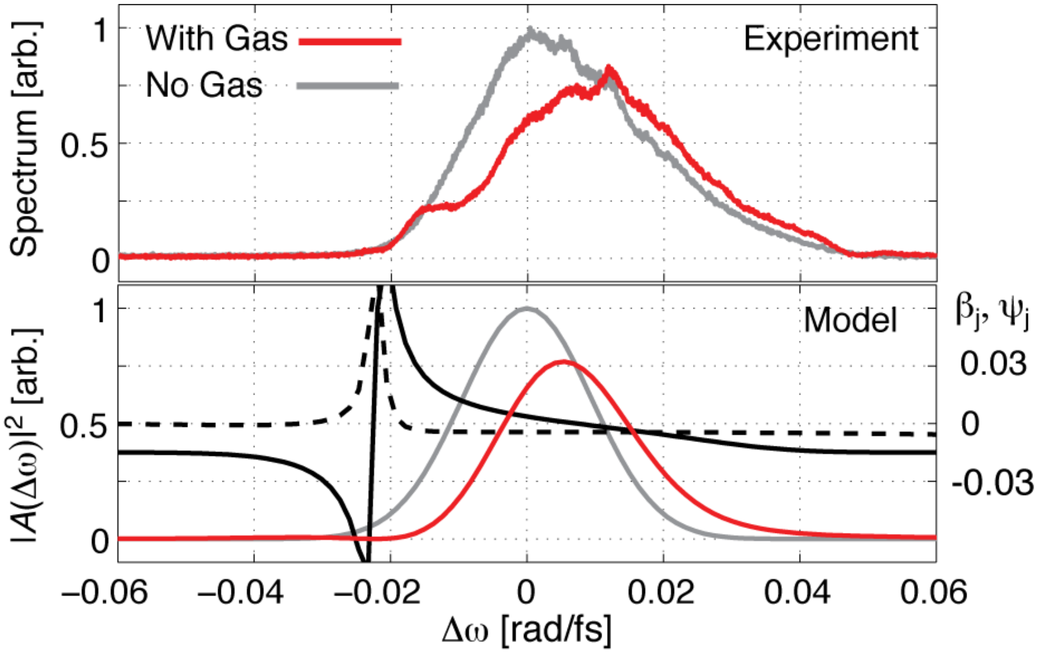
Self Phase Modulation



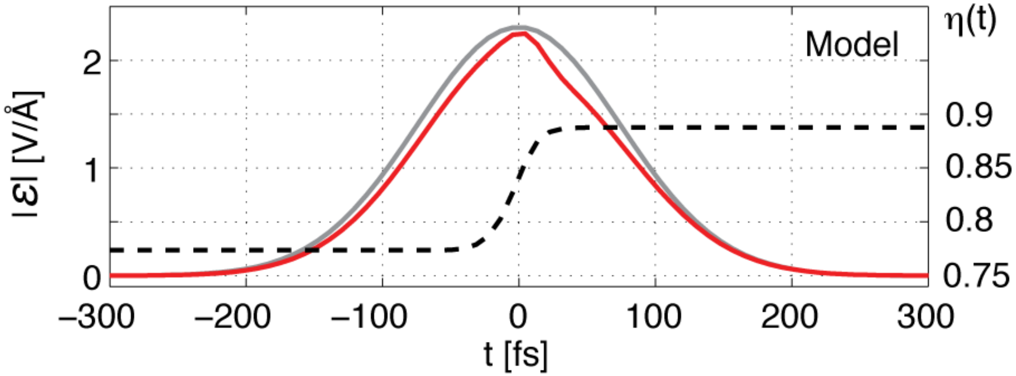
$$f(A)=\left[i\psi-\frac{\beta}{2}\right]A$$

$\beta(\omega)$ ——

$\psi(\omega)$ - - - -



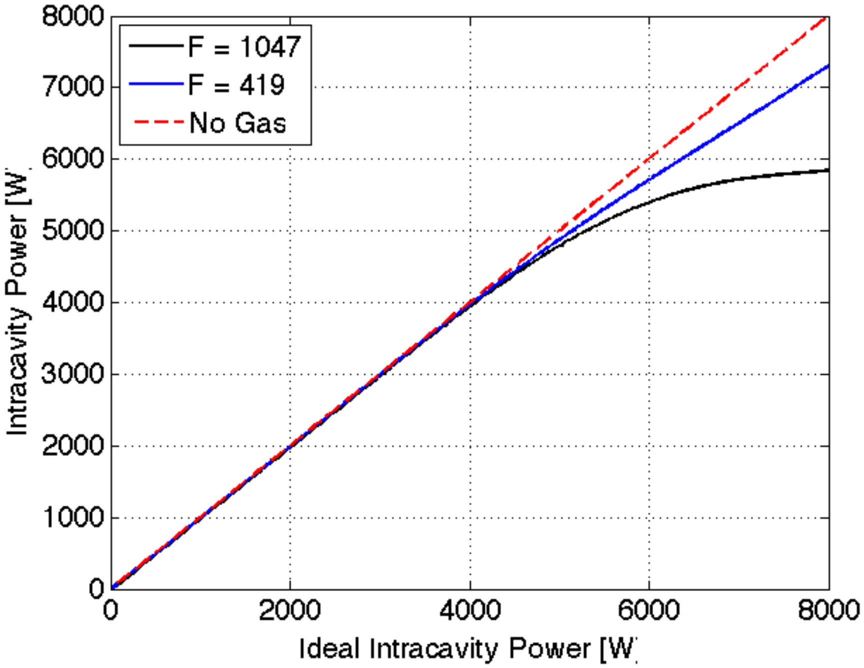
- Stable solution shows significant self phase modulation and a reduction in power.
- Very similar results if loss terms in calculation are omitted.

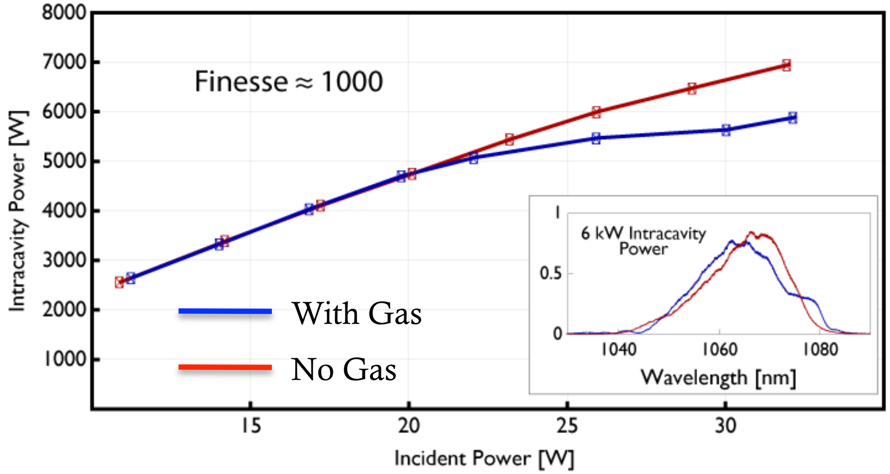


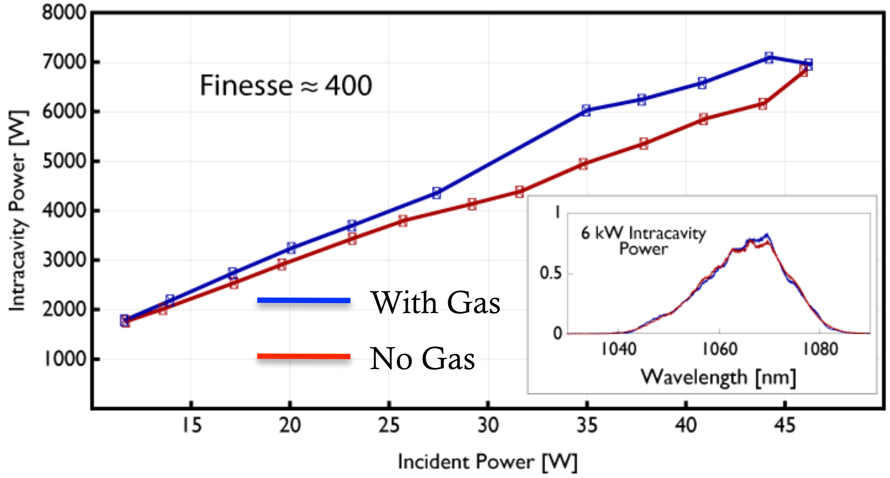
T. K. Allison et al., *Phys. Rev. Lett.* **107**, 183903 (2011)
D. C. Yost et al., *Opt. Exp.* **19**, 23483 (2011)

Self Phase Modulation

Power clamping due to SPM is strongly dependent on finesse.







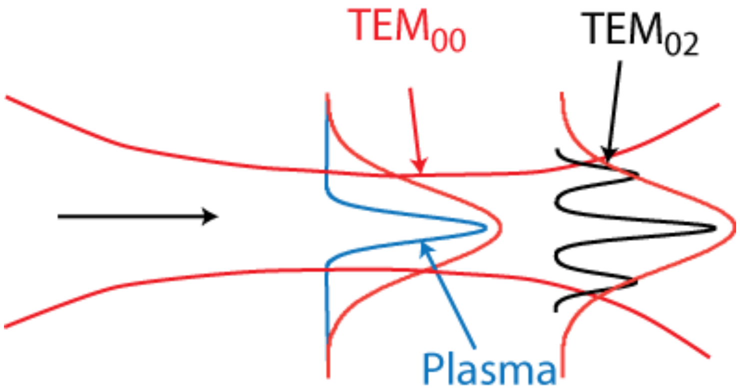
T. K. Allison et al., *Phys. Rev. Lett.* **107**, 183903 (2011)
D. C. Yost et al., *Opt. Exp.* **19**, 23483 (2011)

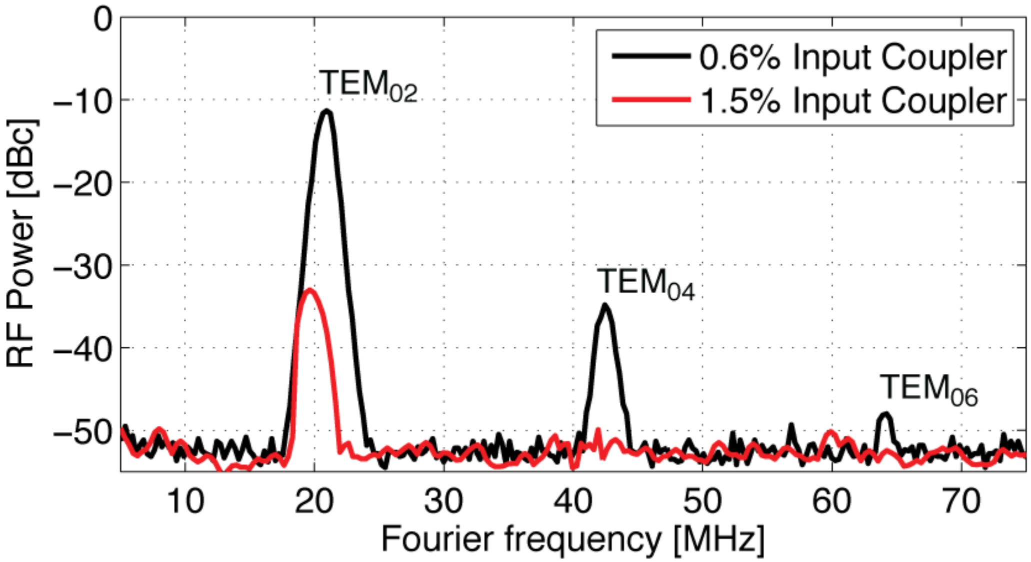
Higher Order Mode Excitation

- Plasma has higher spatial frequency content than the TEM_{00} mode of the cavity and can couple power into higher order modes ⇒ Multimode operation.

- Higher order modes have slightly different resonant frequencies
⇒ mode beating.

- Usually occurs at gas flows and intracavity powers beyond those optimal for HHG





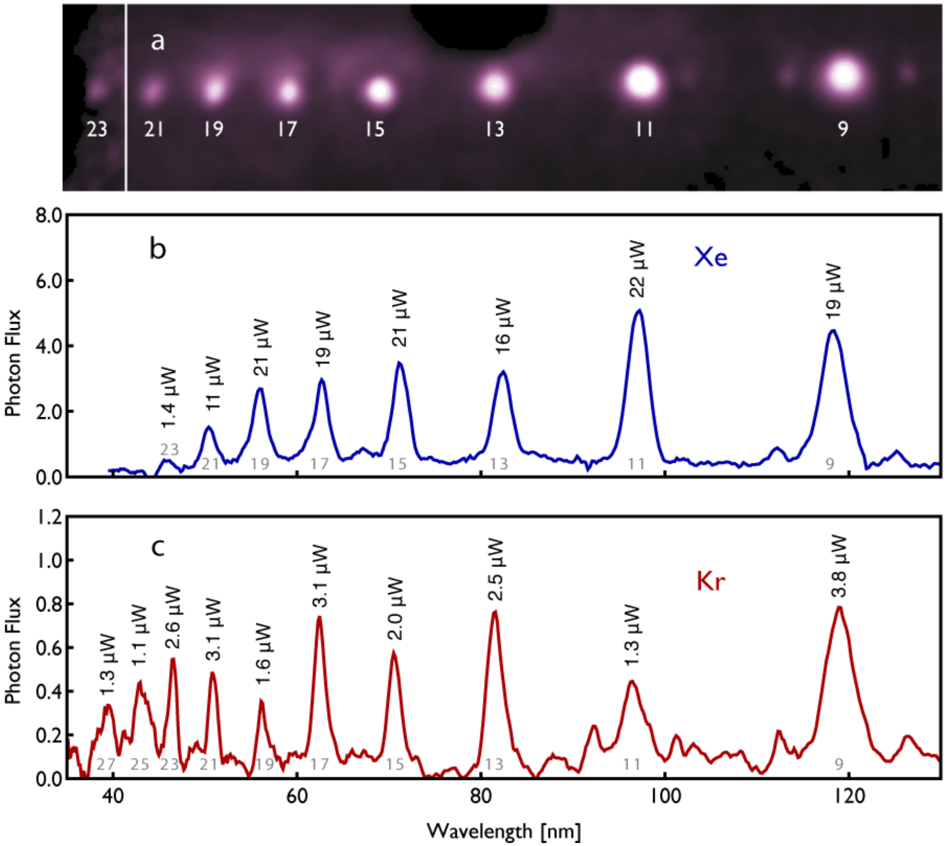
T. K. Allison et al., *Phys. Rev. Lett.* **107**, 183903 (2011)
D. C. Yost et al., *Opt. Exp.* **19**, 23483 (2011)

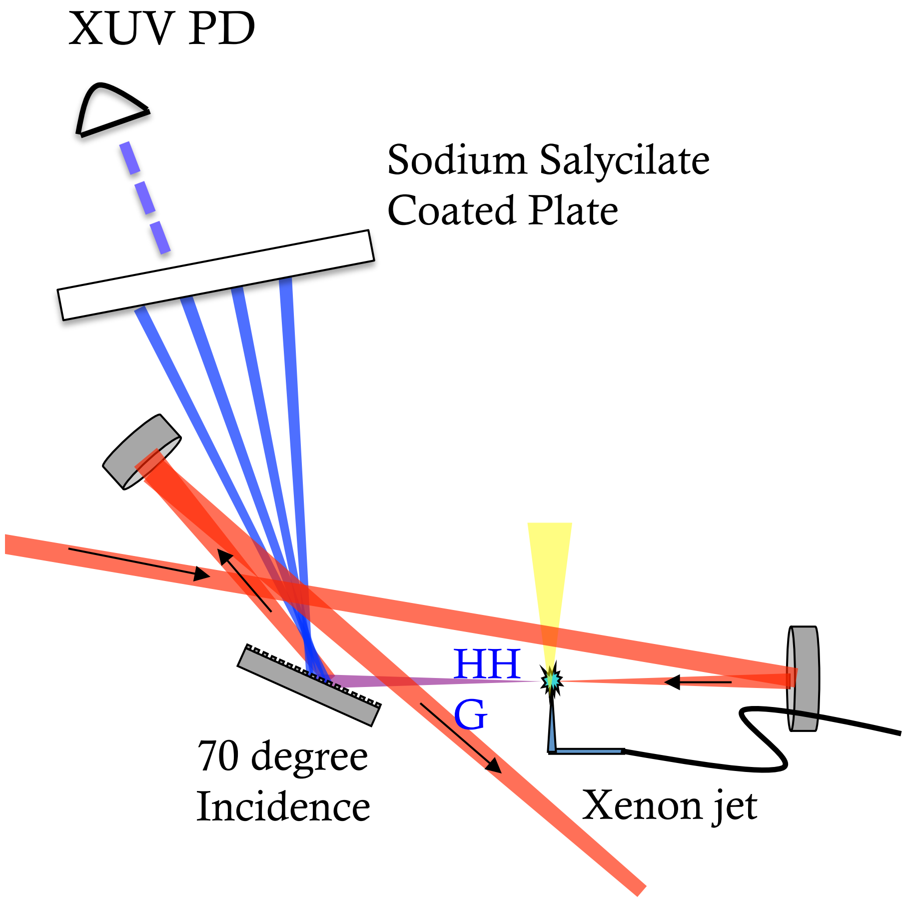
Power Scaling Results

- In Xe, > 200 μW/harmonic, or 7×10^{13} photons/harmonic/second. (10^{6} improvement)

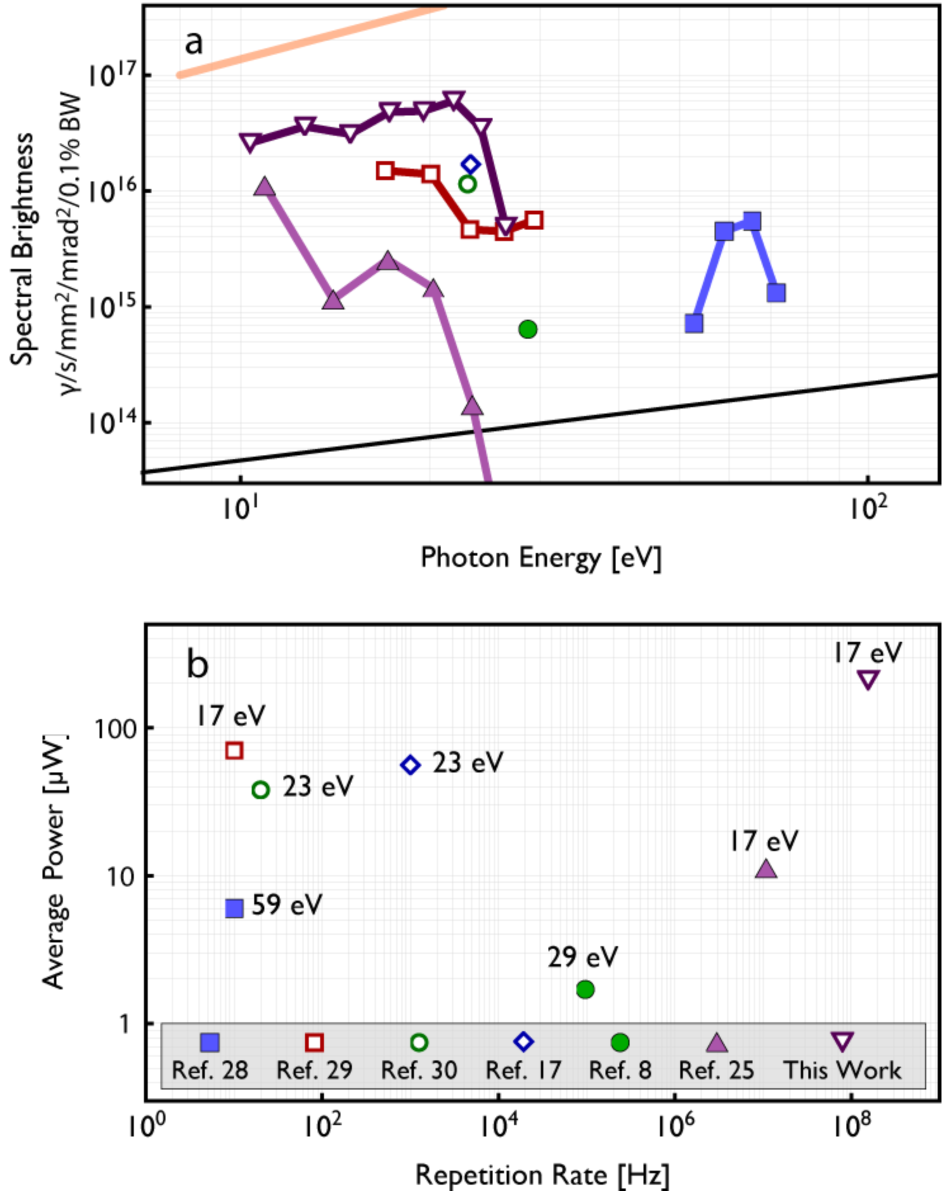
- In brightness units, ~4×10^{16} ph/s/mm^2/mrad^2/0.1% BW generated, about 1/10 XUV beamlines of the ALS.

- With Kr HHG, > 10 μW in the 27th harmonic.



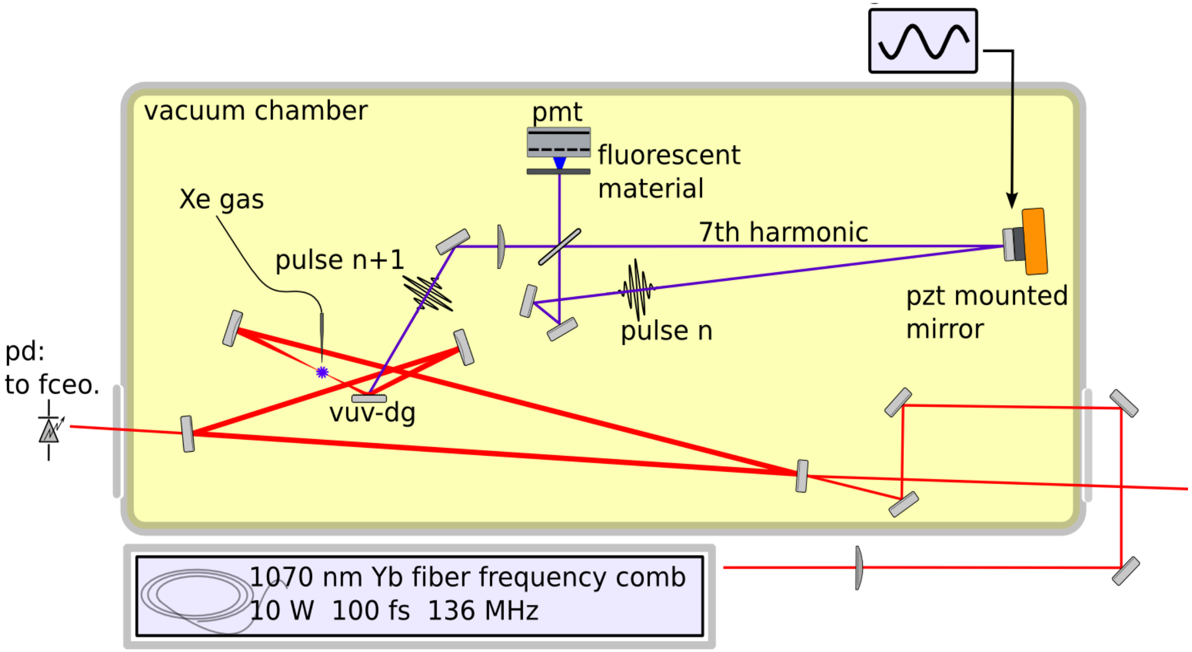


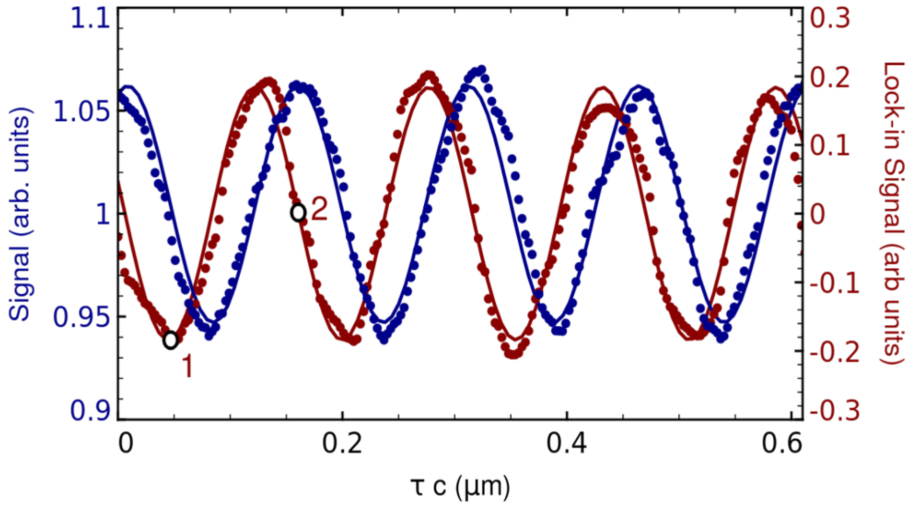
Power Comparison



Among the highest average power HHG sources demonstrated to date.

Pulse to Pulse Coherence



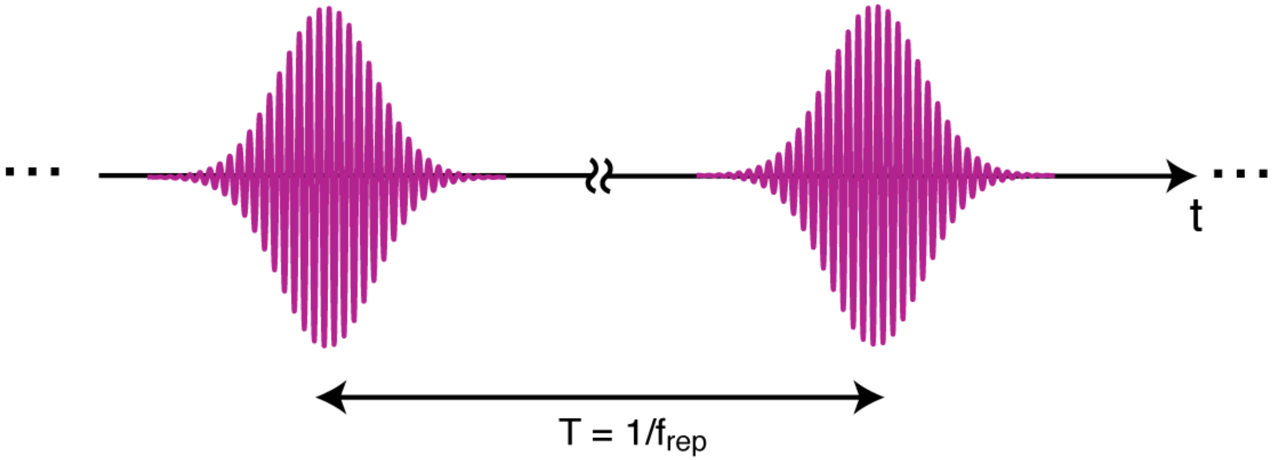


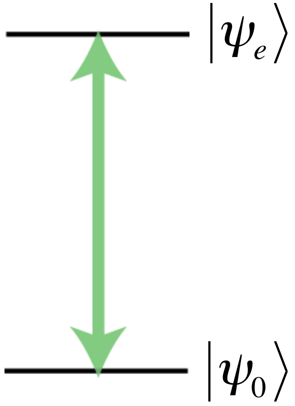
- Only the 7th harmonic (below threshold).
- Establishes that the coherence time is longer than the rep rate.
- Poor contrast in the interferogram makes interpretation difficult.

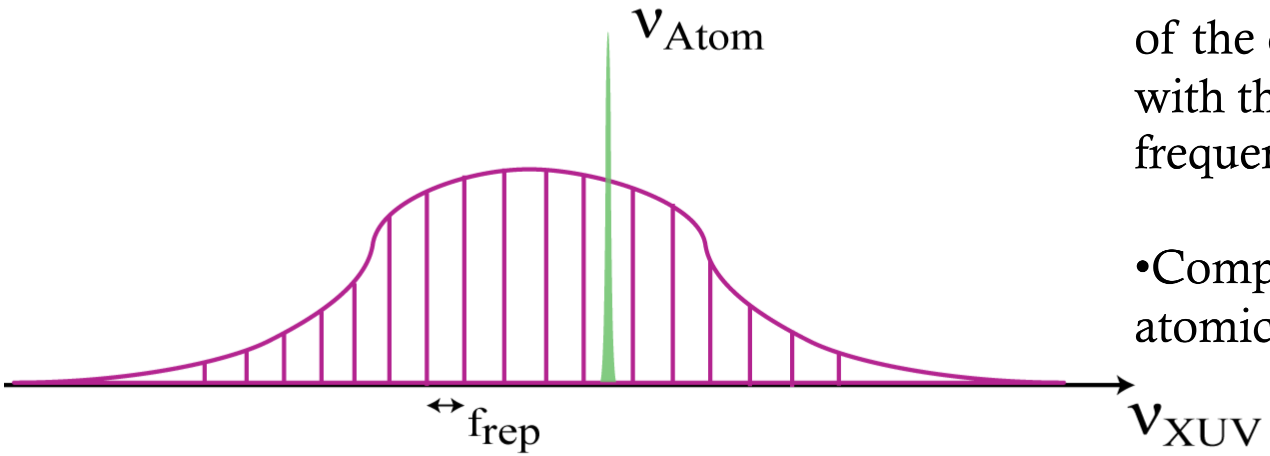
Yost et. al., Nat. Phys. **5**, 815 (2009)

Direct Frequency Comb Spectroscopy

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





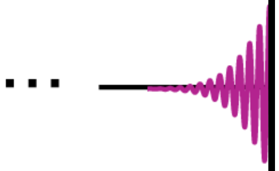


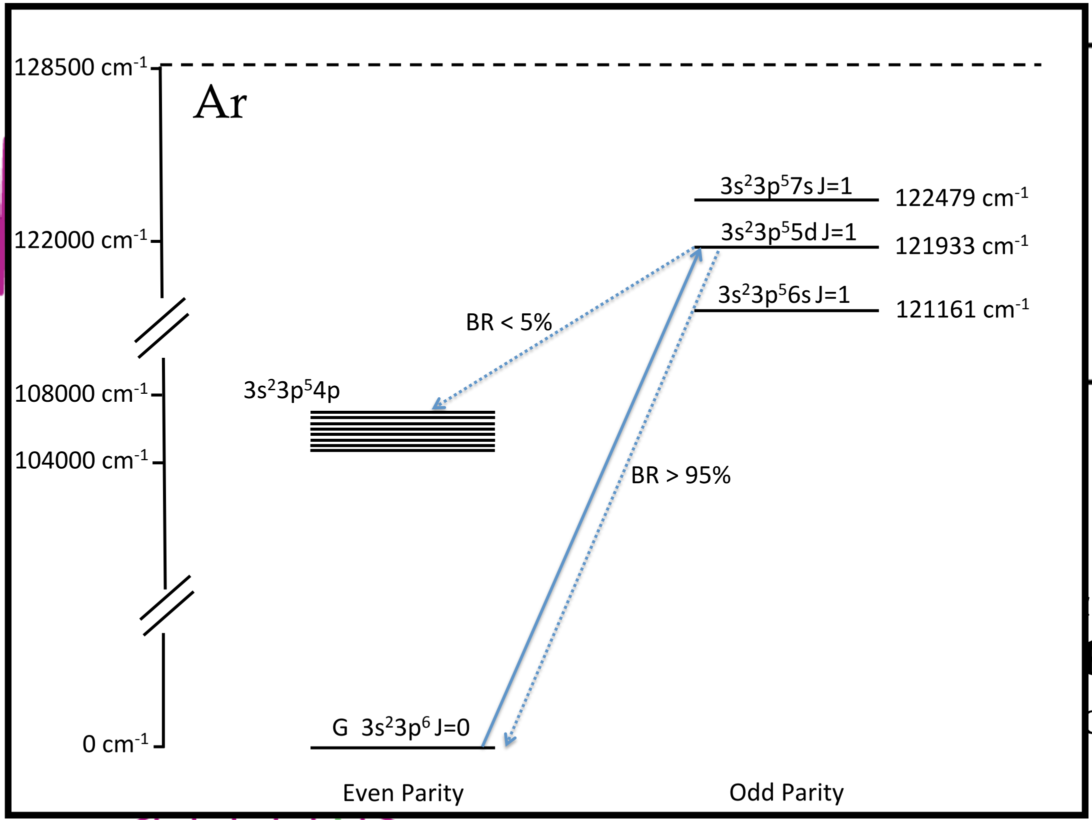
- Resonance occurs when one of the comb lines coincides with the atomic transition frequency.

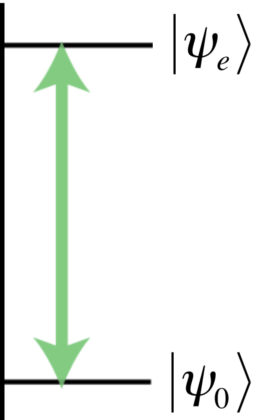
- Compare comb linewidth to atomic linewidth.

Direct Frequency Comb Spectroscopy

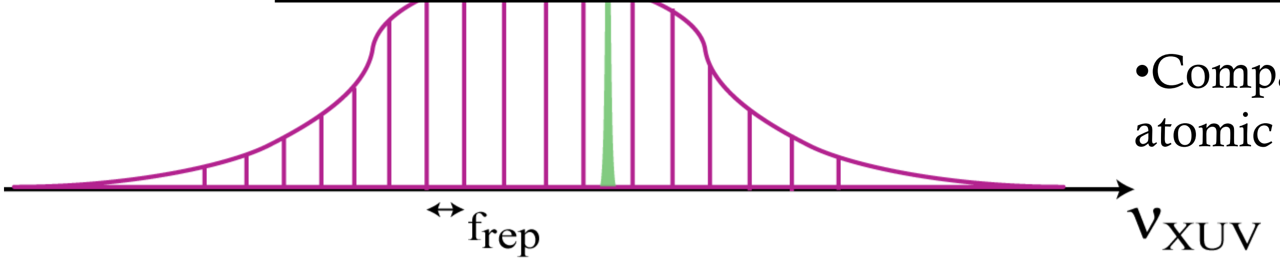
$$\langle \psi_e | \psi(t) \rangle \approx e^{-i(\Delta E/\hbar)t} + e^{-i[(\Delta E/\hbar)(t-T)+q\Delta\phi]} + e^{-i[(\Delta E/\hbar)(t-2T)+2q\Delta\phi]} + \cdots$$





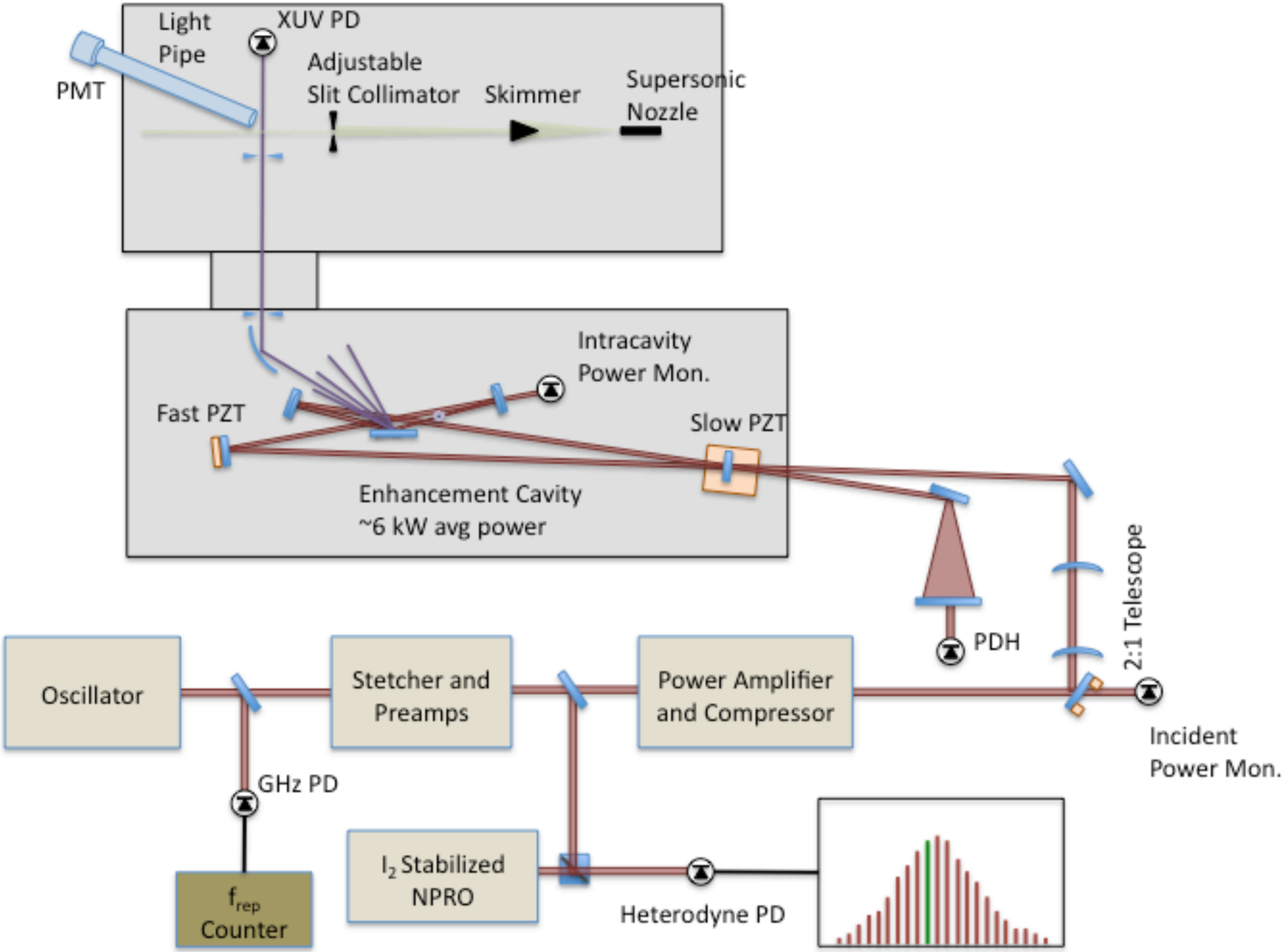


e occurs when one
b lines coincides
omic transition

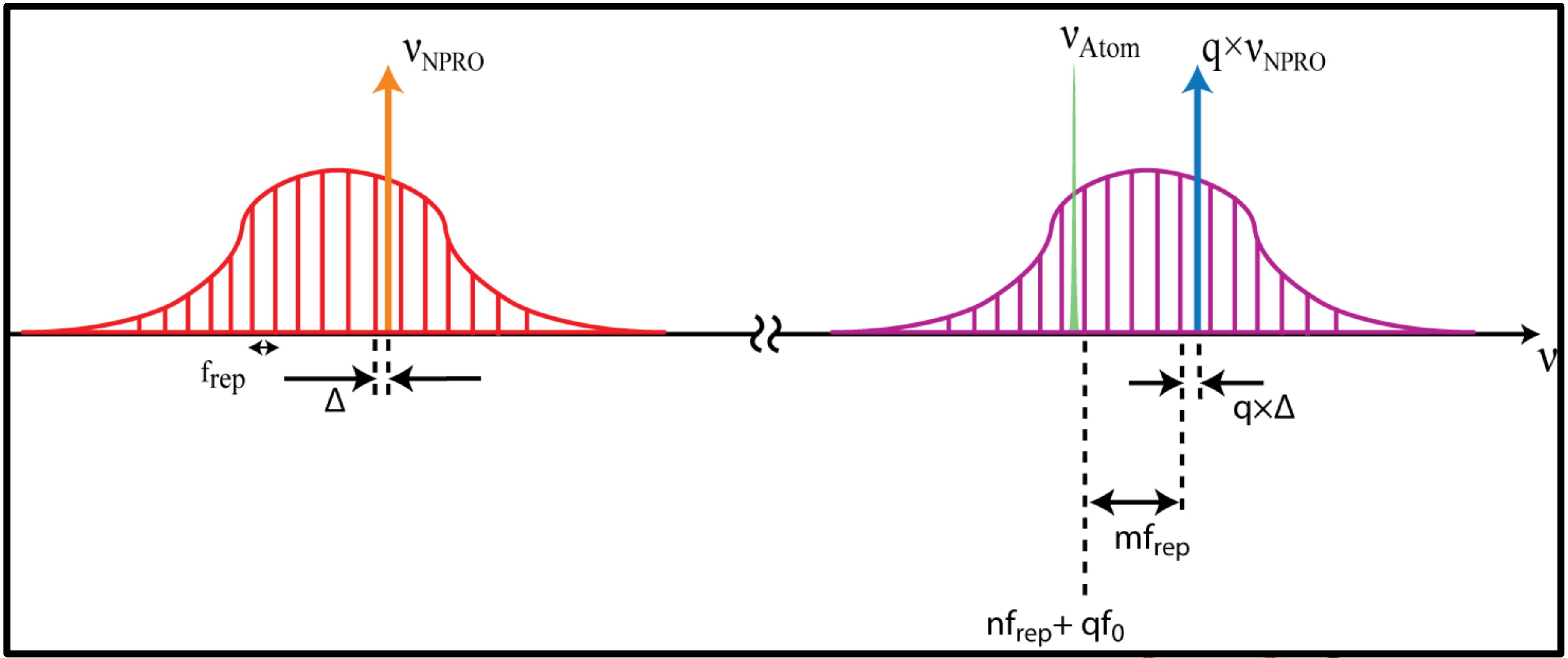


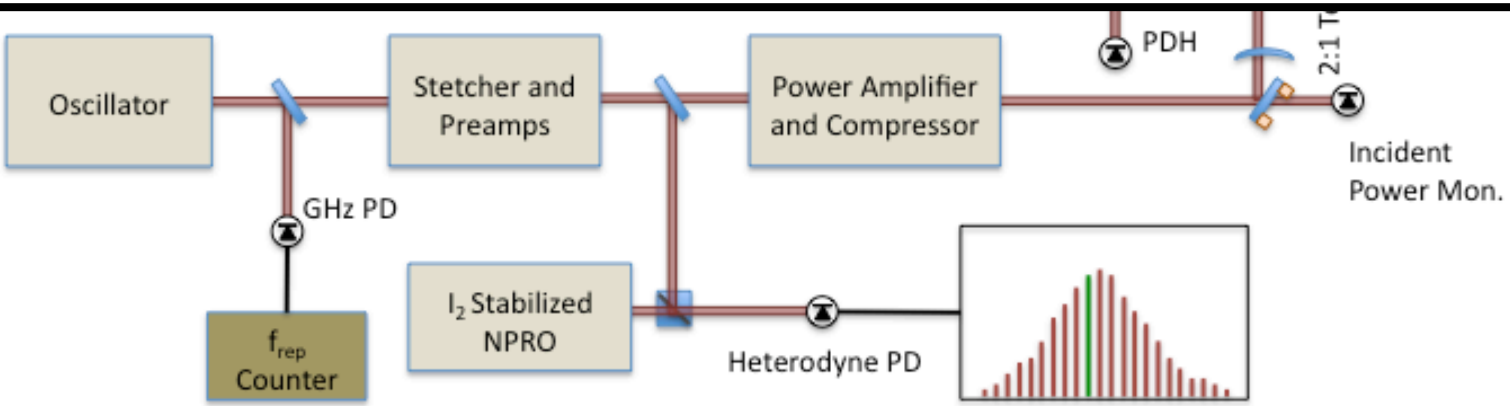
•Compare comb linewidth to atomic linewidth.

Apparatus


Light Pipe
XUV PD
PMT
Adjustable Slit Collimator
Skimmer
Supersonic Nozzle
Intracavity Power Mon.
Fast PZT
Slow PZT
Enhancement Cavity
~6 kW avg power
PDH
2:1 Telescope
Oscillator
Stetcher and Preamps
Power Amplifier and Compressor
Incident Power Mon.
GHz PD
f_{rep} Counter
I_2 Stabilized NPRO
Heterodyne PD

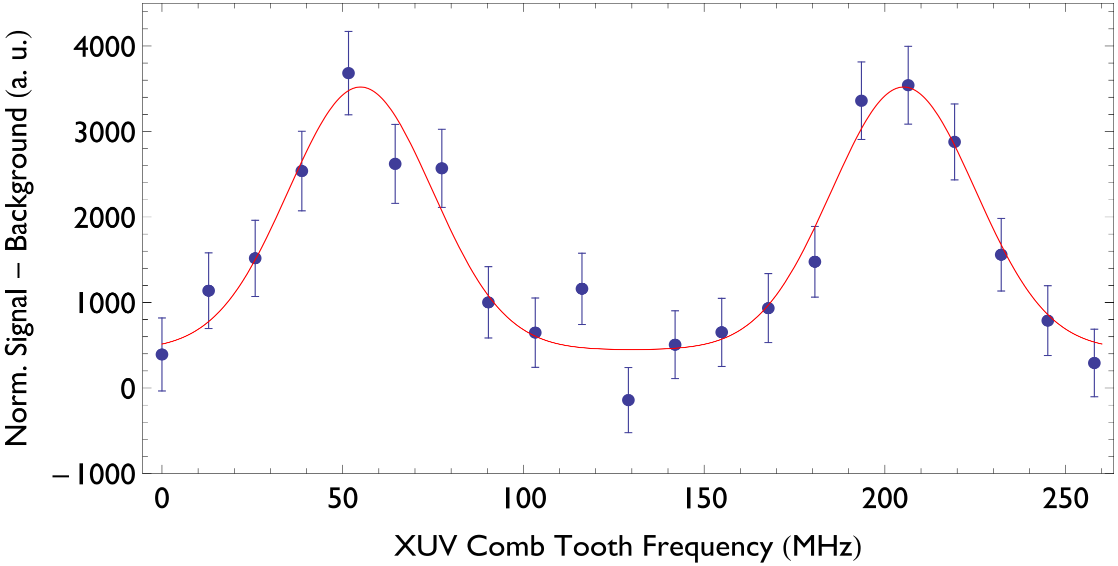
Apparatus


ν_{NPRO}
ν_{Atom}
$q\times\nu_{NPRO}$
f_{rep}
Δ
ν
$q\times\Delta$
mf_{rep}
$nf_{rep} + qf_0$


PDH
2:1 T
Oscillator
Stetcher and Preamps
Power Amplifier and Compressor
Incident Power Mon.
GHz PD
f_{rep} Counter
I_2 Stabilized NPRO
Heterodyne PD

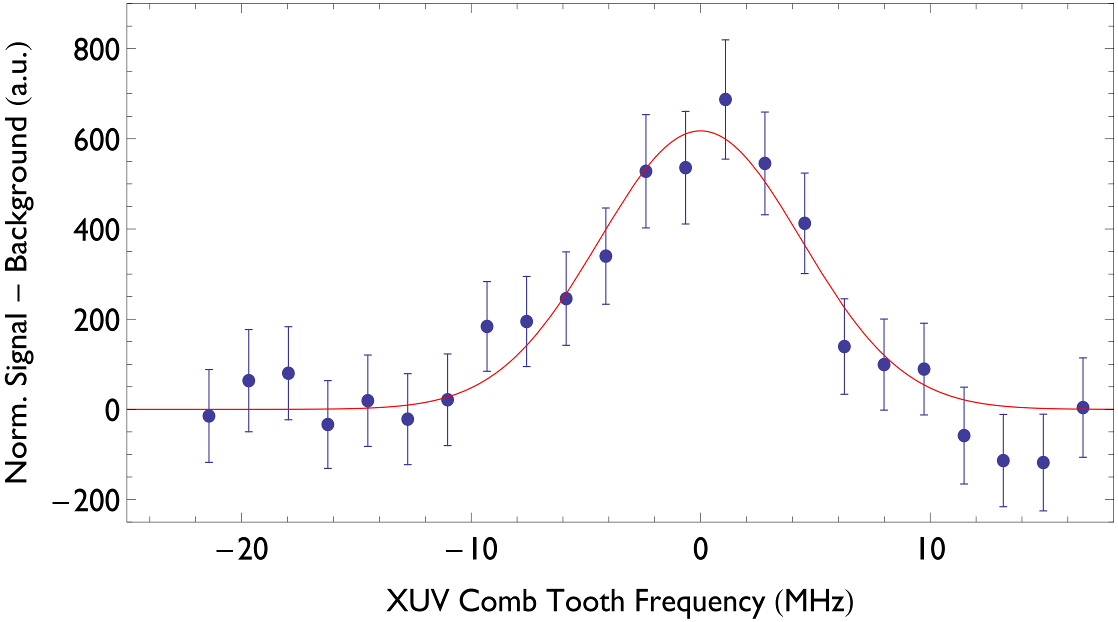
Argon Spectroscopy Results

Wide open slit, ~ 40 MHz doppler width. Scan 1.5 FSR.



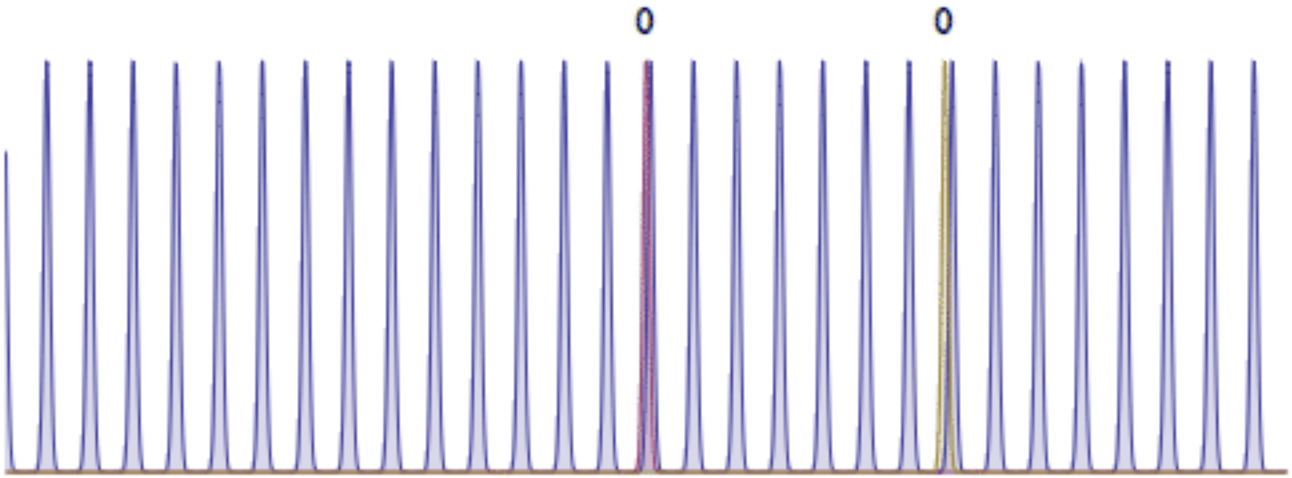
- Narrow slit, ~ 10 MHz doppler width.

⇒Comb Linewidth < 10 MHz



Cingöz, et al. Accepted to *Nature*

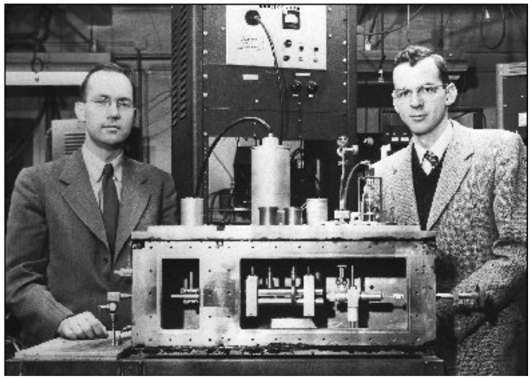
Comb Tooth Determination



13th Harmonic of I_2 stabilized comb mode

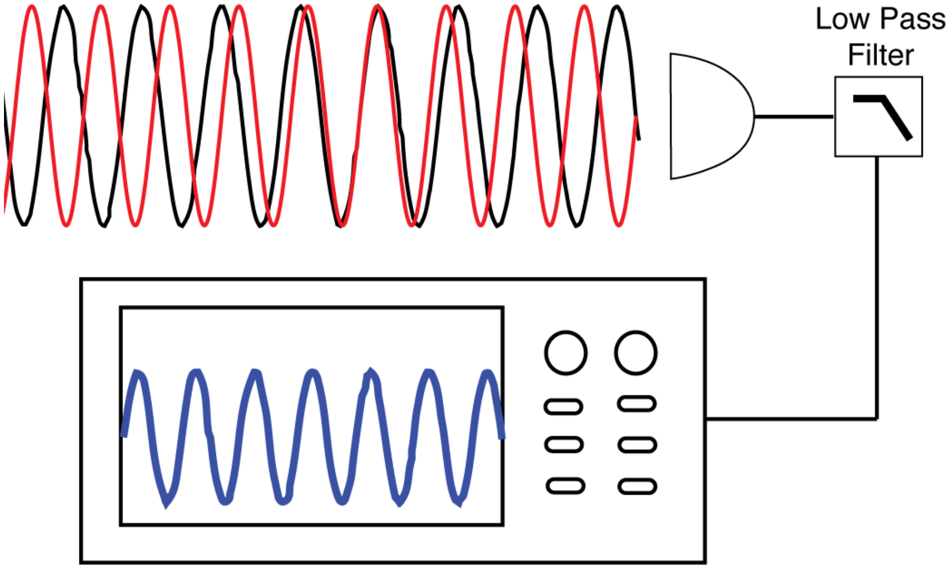
- Absolute comb tooth number of 23,801,529
- Final transition frequency 3,655,454,073±3 MHz
- 10^3 more accurate than ‘wavelength’ spectroscopy

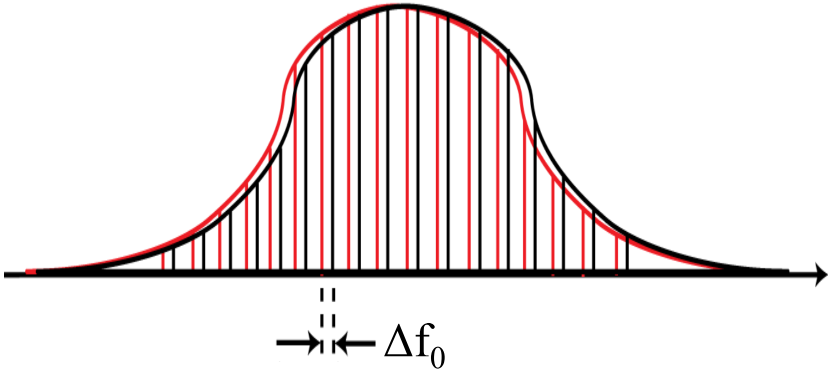
Heterodyne Beat



- 6 months after the invention of the MASER, Townes and Gordon built a second MASER to make a heterodyne beat measurement and characterize its performance.

- This is the standard method for characterizing optical phase noise.

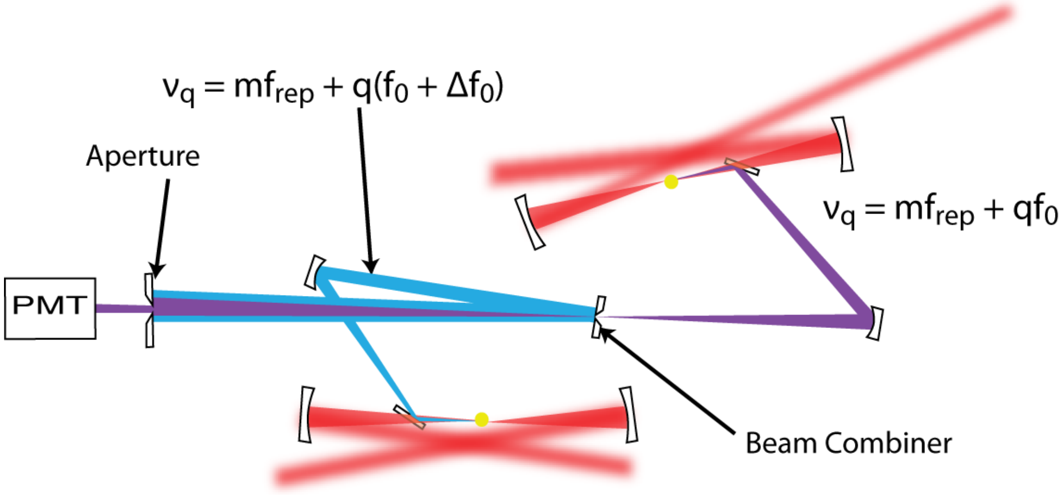




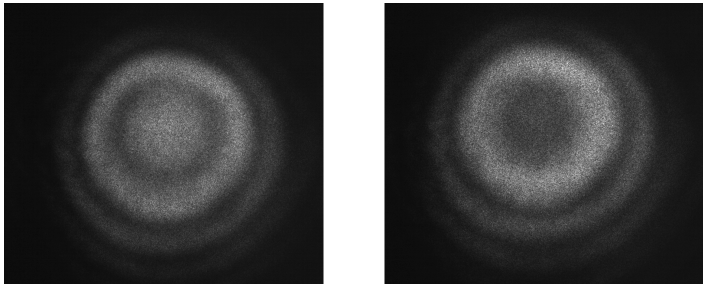
$$\cos[\omega_1 t+\phi_1(t)]+\cos[\omega_2 t+\phi_2(t)]$$

$$\Rightarrow \cos[(\omega_1-\omega_2)t+\phi_1(t)+\phi_2(t)]$$

XUV Interferometer



Examples with HeNe



In Phase

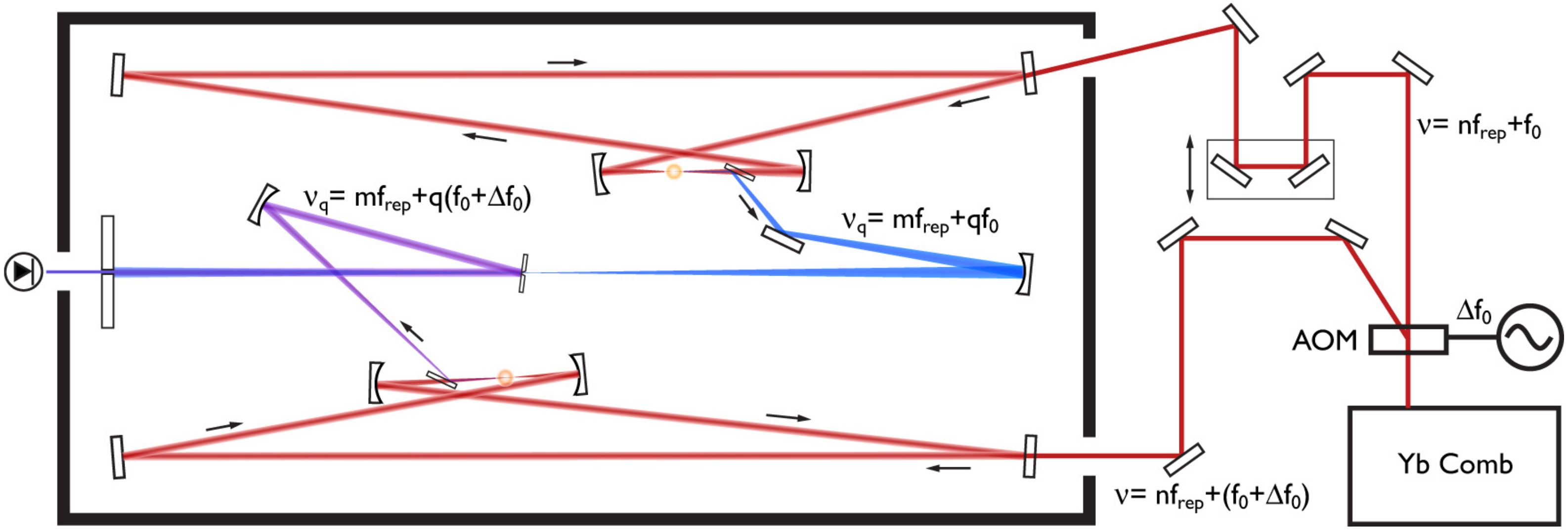
Out of Phase

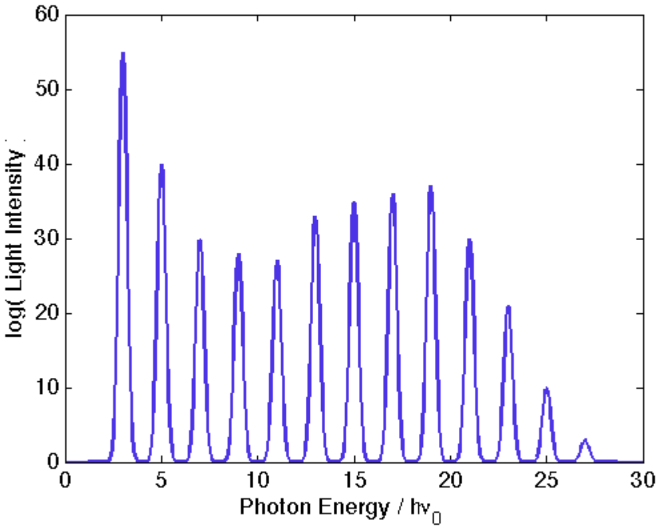
- Beams are combined on a mirror with a pyramidal hole in it.

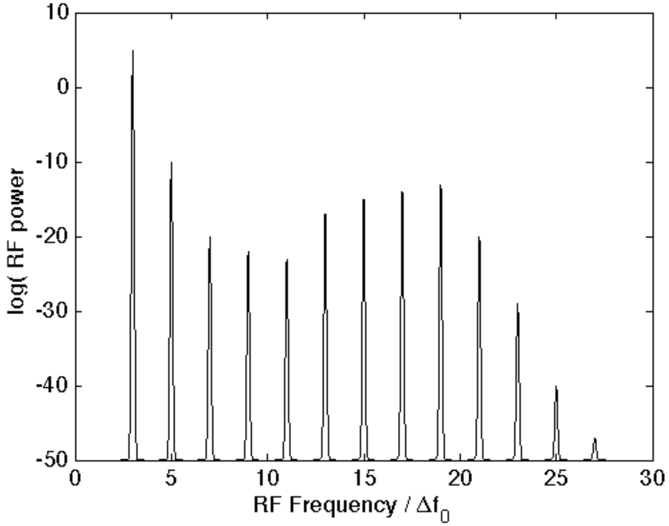
- A circular finge patter develops in the far field. Central portion filtered out with second aperture.

- The size of the central maximum/minumum scales only as $\lambda^{1/2}$. This allows the interferomter to work reasonably well over a broad range of wavelengths.

Heterodyne Beat

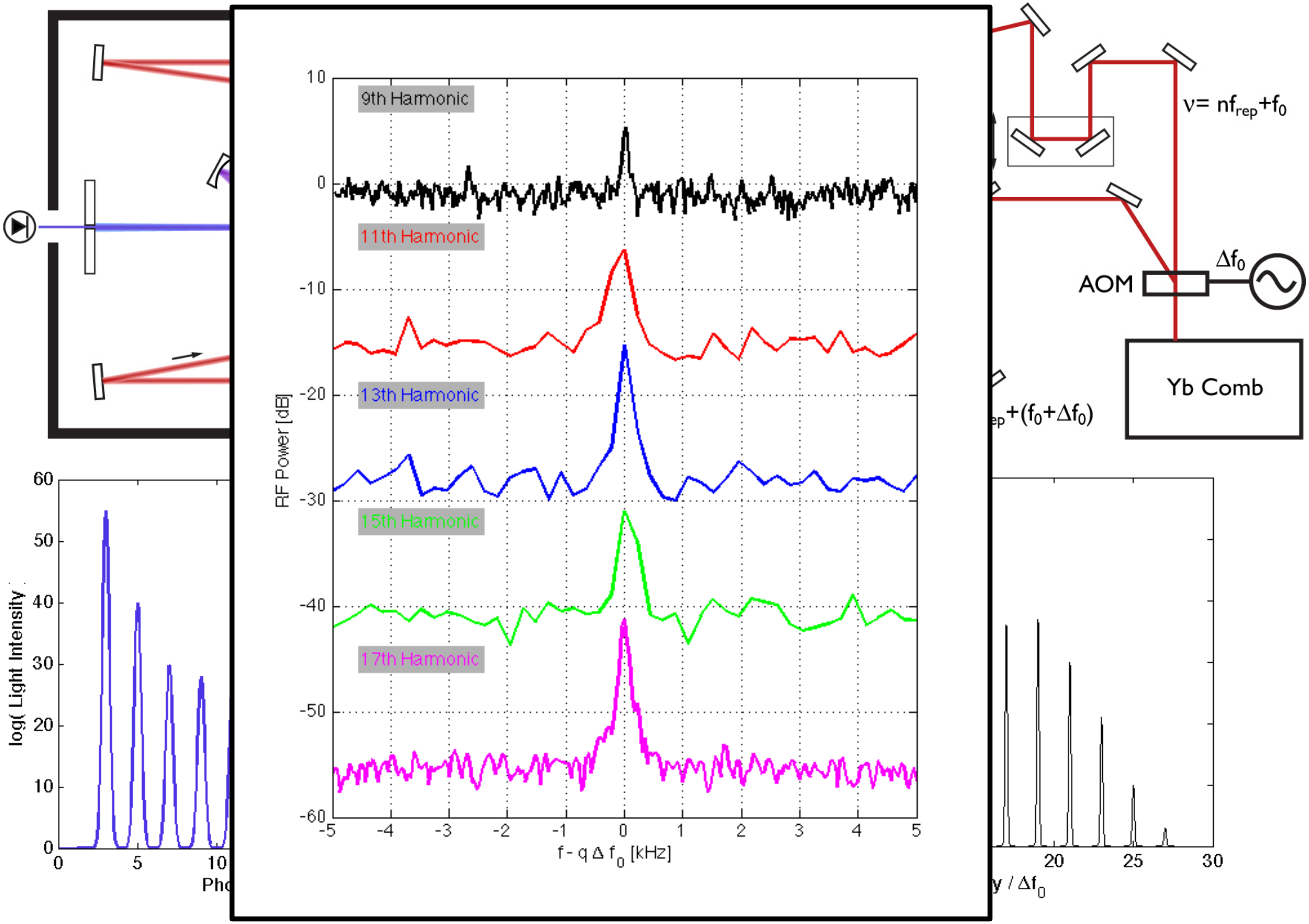






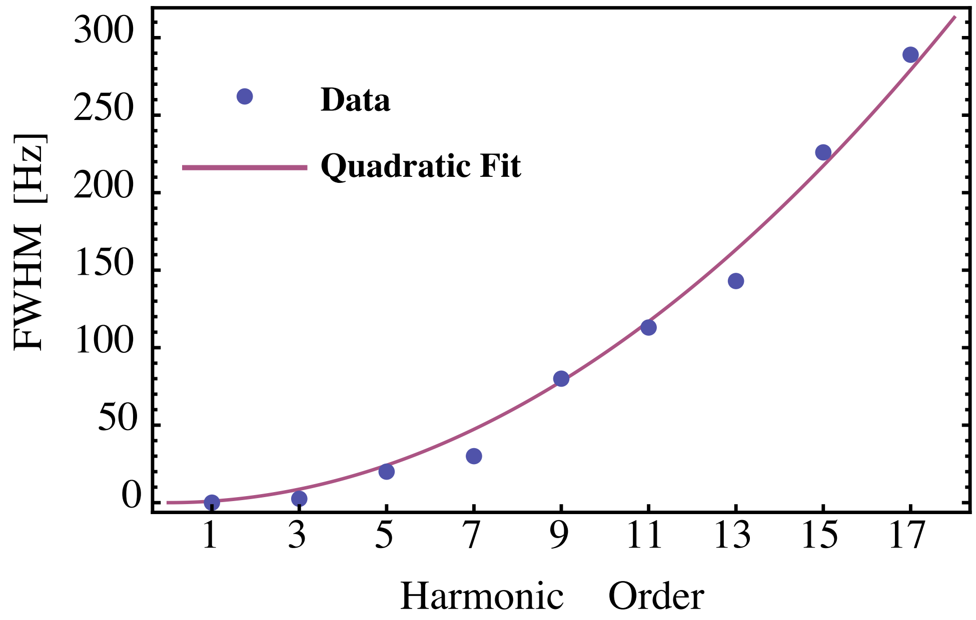
Benko *et al.*, *Nature Photonics* **8**, 530 (2014)

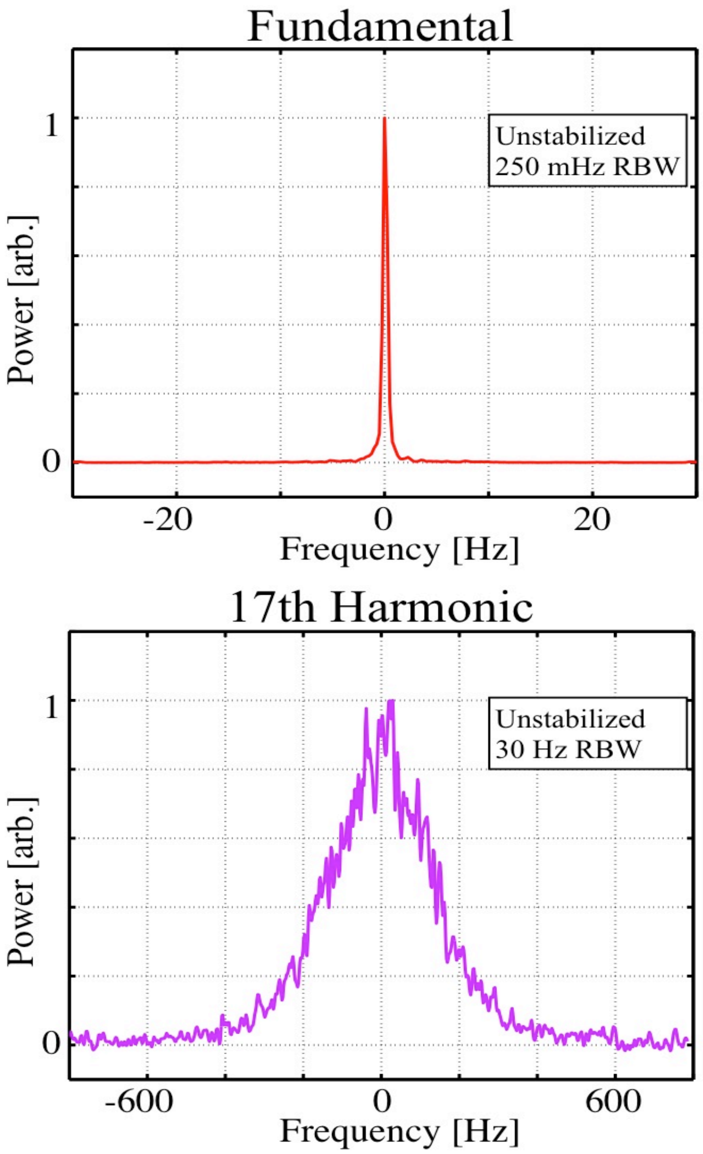
Heterodyne Beat



Benko *et al.*, *Nature Photonics* **8**, 530 (2014)

Heterodyne Beat

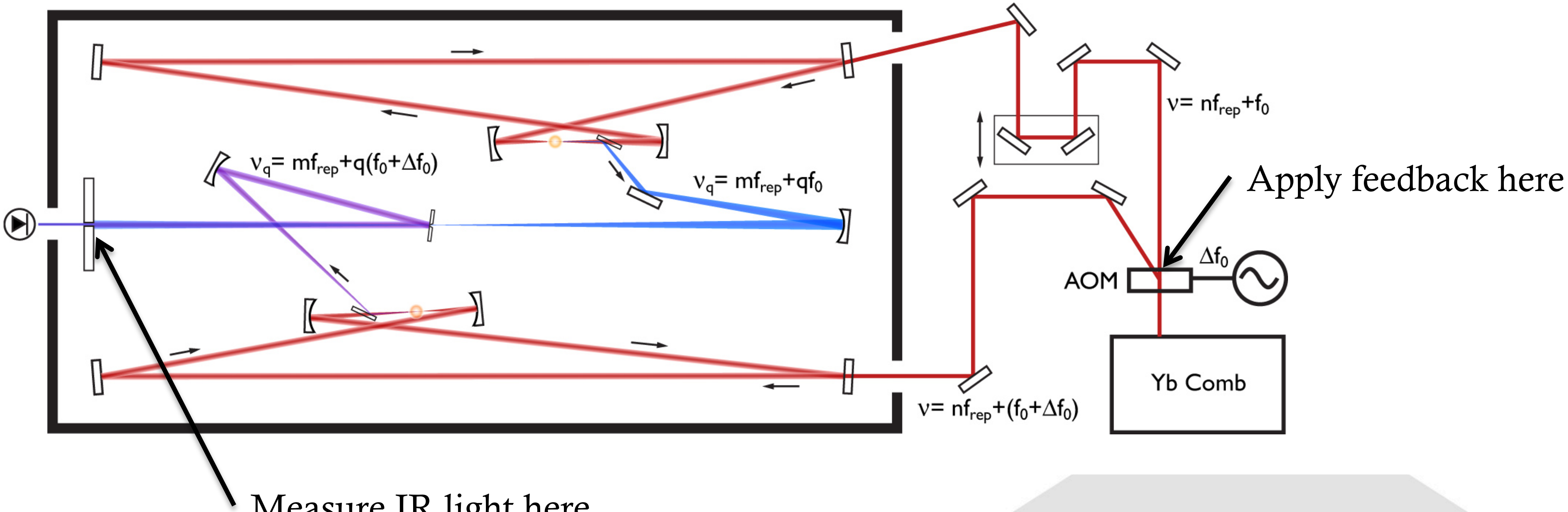




⇒ Harmonic linewidths scale quadratically with harmonic order, as in noiseless classical frequency multiplication.

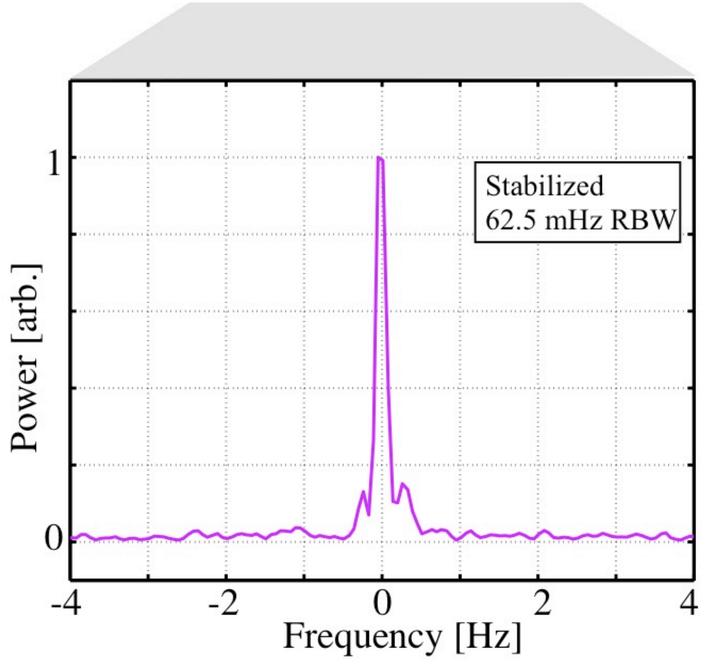
Benko *et al.*, *Nature Photonics* **8**, 530 (2014)

Sub-Hertz Linewidths



Apply feedback here

Measure IR light here

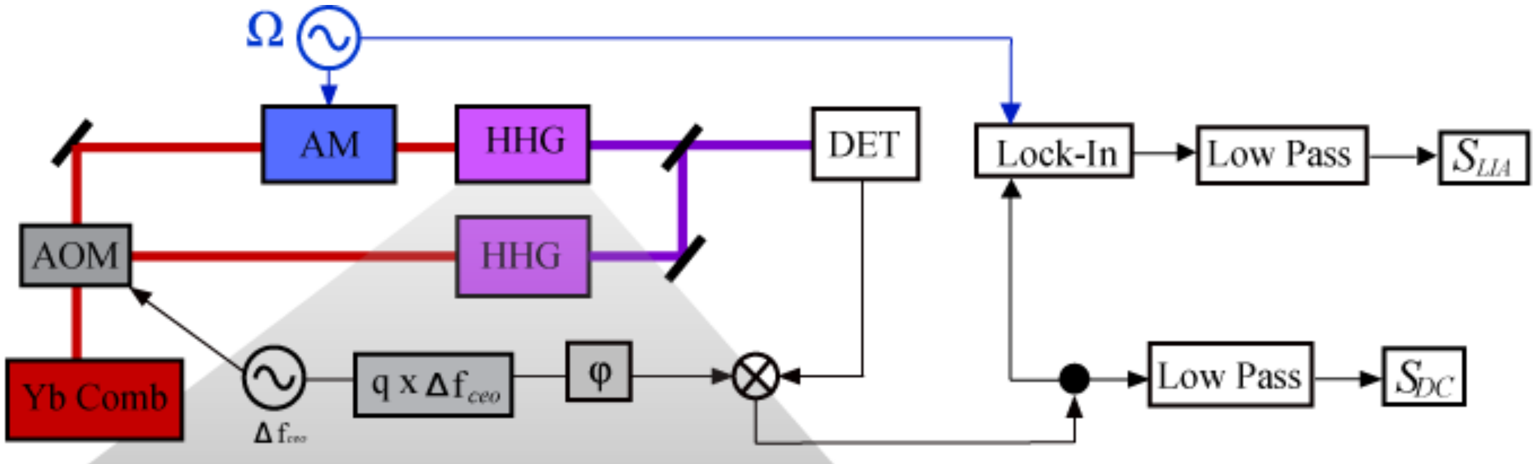


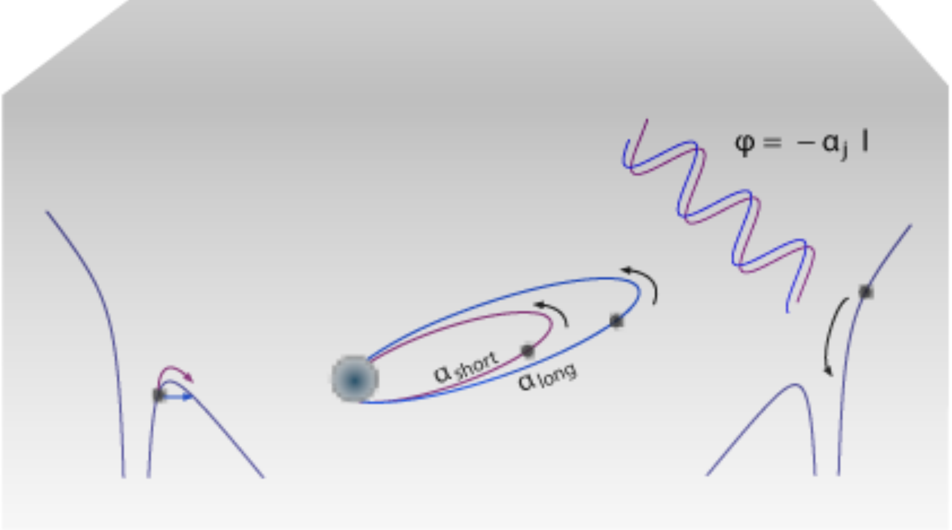
⇒ Intracavity HHG faithfully transfers coherence of IR light into the XUV.

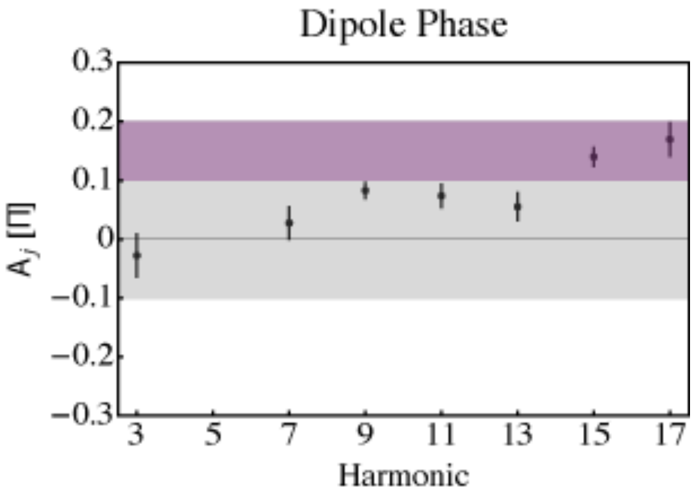
⇒ Can support precision measurement in the XUV and X-ray regime.

Benko *et al.*, *Nature Photonics* **8**, 530 (2014)

High Harmonic Phases





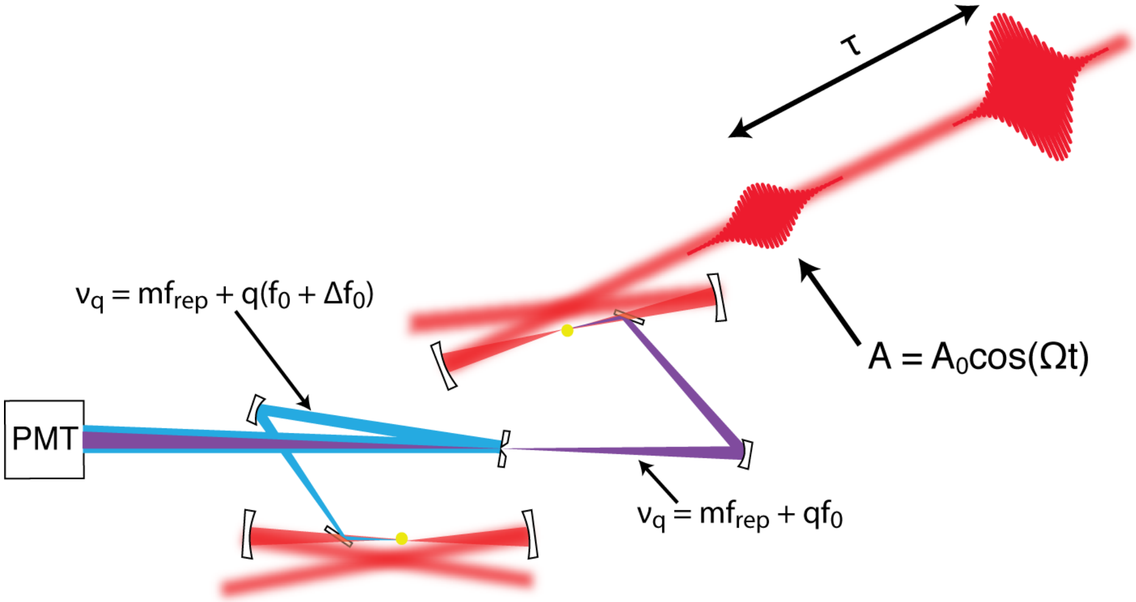


Gray region = Hostetter PRA **82**, 023401 (2010

Purple region = Standard Semi-Classical Model

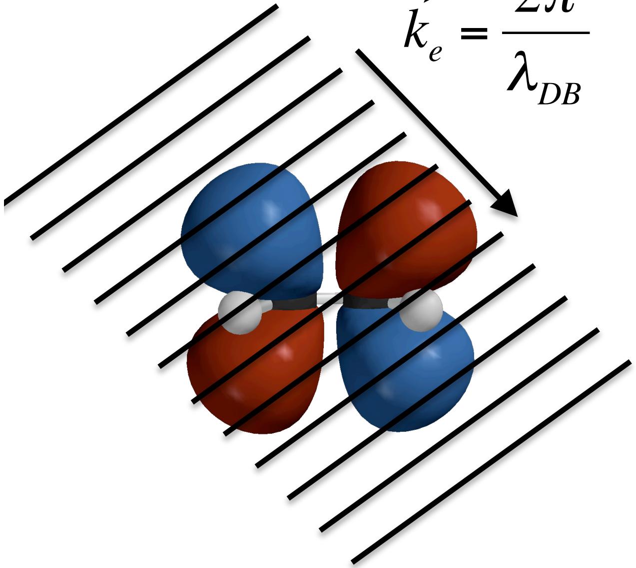
Benko *et al.*, *Nature Photonics,* In press. arXiv:1404.3779

HHG Spectroscopy



$$\vec{D}(\omega,\theta) = \kappa(\vec{k})a(\vec{k})\vec{d}(\vec{k})$$

$$\vec{k}_e = \frac{2\pi}{\lambda_{DB}}$$



$$\overbrace{\left(A_1 + \Delta A_1(\tau)\cos(\Omega t)\right)\cos\left[\omega_1 t + \Delta\phi_1(\tau)\cos(\Omega t)\right]}^{\text{Modulated HHG Signal}} \quad + \quad \overbrace{\cos\left[\omega_2 t\right]}^{\text{Local Oscillator}}$$

$$\Rightarrow A_2\left(A_1 + \Delta A_1(\tau)\cos(\Omega t)\right)\cos\left[(\omega_1 - \omega_2)t + \Delta\phi_1(\tau)\cos(\Omega t)\right]$$

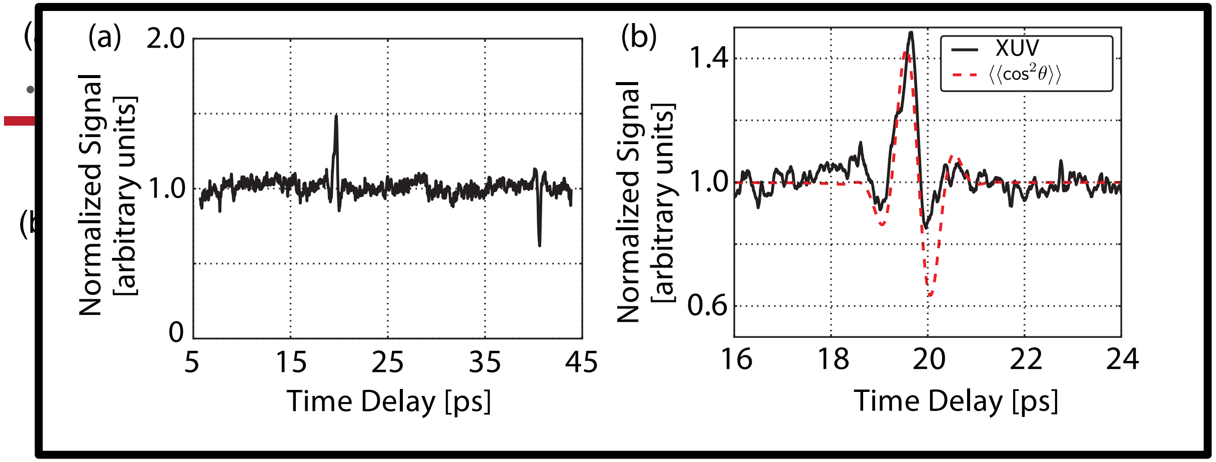
⇒ Molecular information encoded in RF waveform!

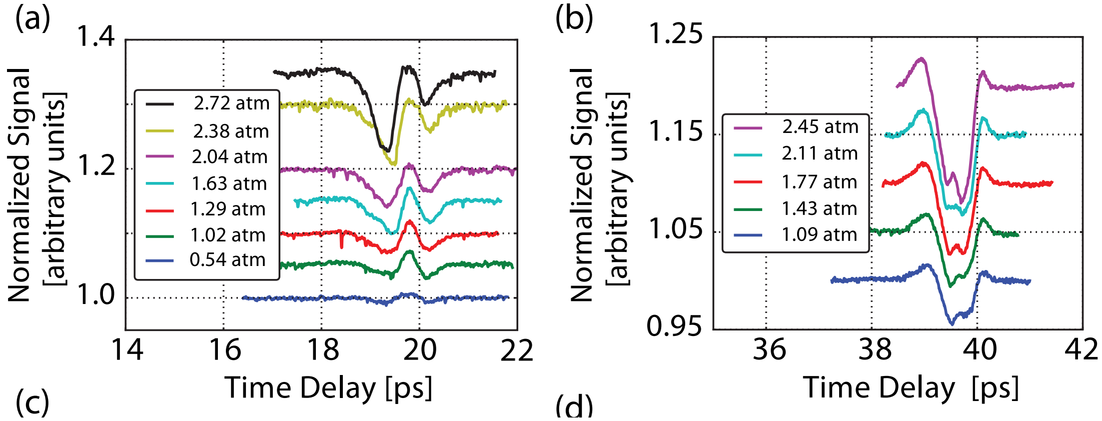
PRL **114,** 153001 (2015) PHYSICAL REVIEW LETTERS week ending
17 APRIL 2015

Cavity-Enhanced Field-Free Molecular Alignment at a High Repetition Rate

Craig Benko,[1,*] Linqiang Hua,[1,2] Thomas K. Allison,[3] François Labaye,[1] and Jun Ye[1,†]
[1] *JILA, NIST and the University of Colorado, 440 UCB, Boulder, Colorado 80309-0440, USA*
[2] *State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, China*
[3] *Departments of Chemistry and Physics, Stony Brook University, Stony Brook, New York 11794-3400, USA*

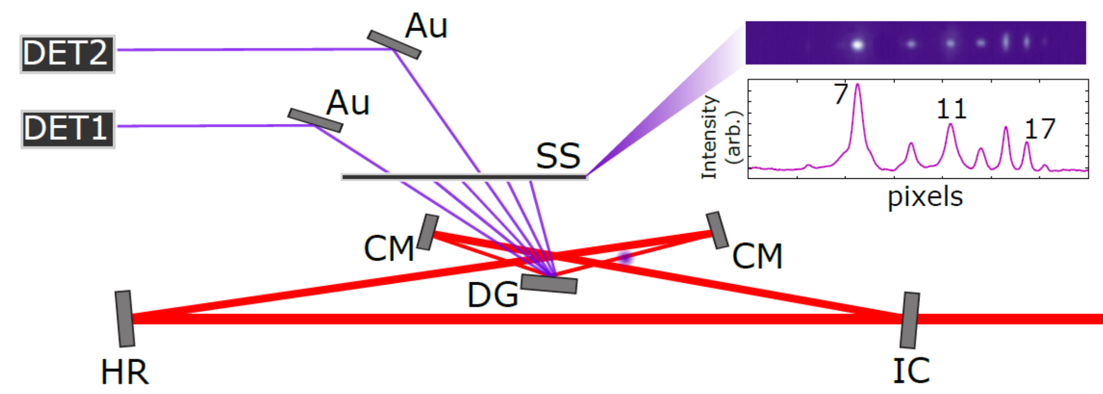
(Received 7 January 2015; published 14 April 2015)





The XUV Laser Pointer

Harmonic	**Target parameters**	**Generated power**	**Outcoupled power**	**Photons/s generated**
11th , 12.7 eV	100 μm, 100 PSI, 4:1 mix	0.7 mW	60 μW	
17th, 19.7 eV	100 μm, 100 PSI, 4:1 mix	0.63 mW	60 μW	
11th , 12.7 eV	50 μm, 400 PSI, 4:1 mix	0.84 mW	71 μW	
11th , 12.7 eV	50 μm, 275 PSI, 9:1 mix	0.93 mW	78 μW	



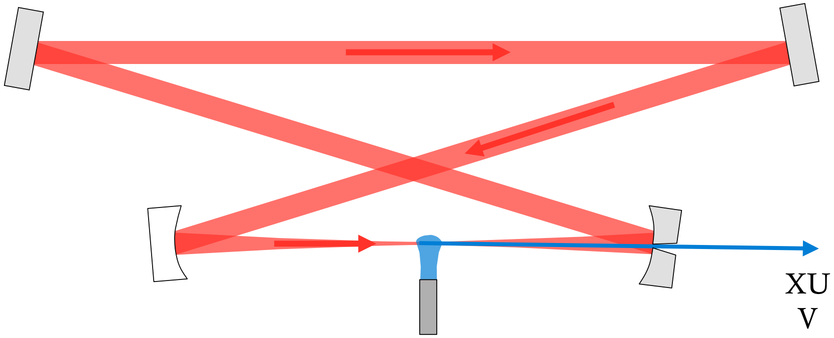
1.2 x 10^{17} photons /[s mm^2 mrad^2 .1% bandwidth]
¼ brightness of the ALS

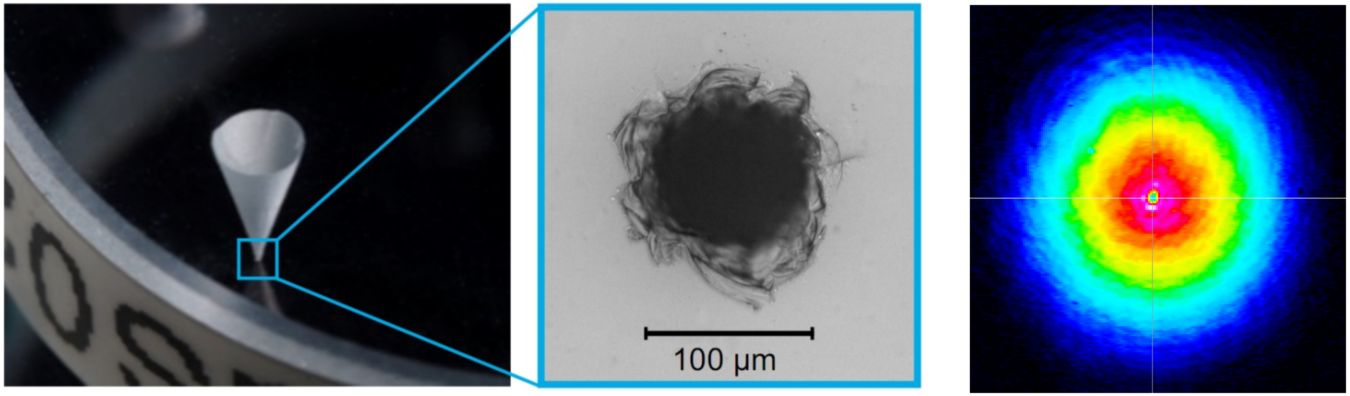
Benko *et al.* In preparation. 2015

The Enhancer Crew in Garching

Geometric XUV Output Coupling

→

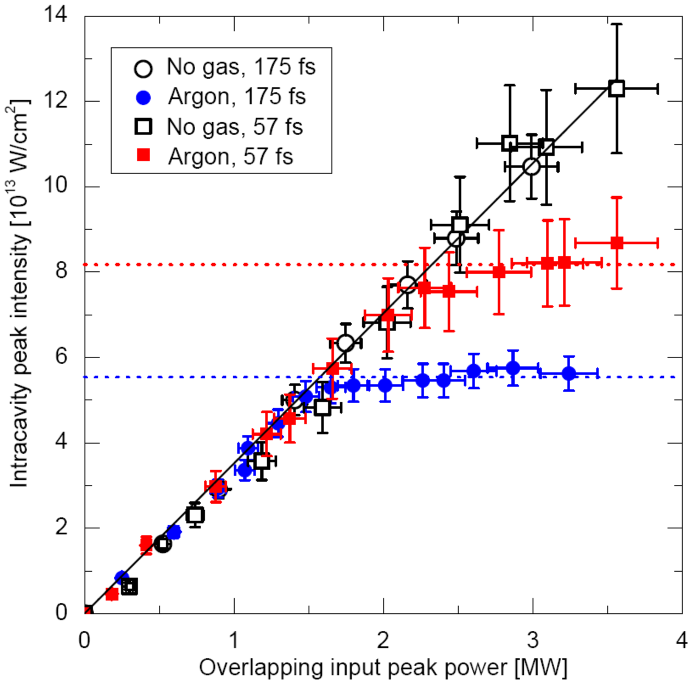


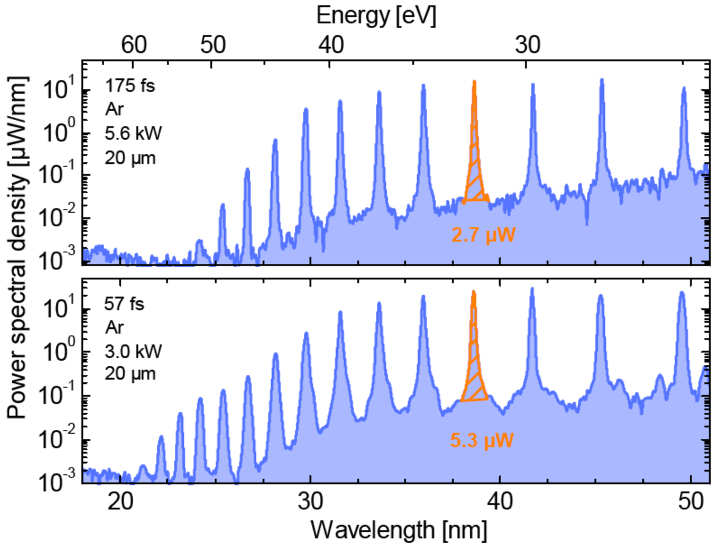


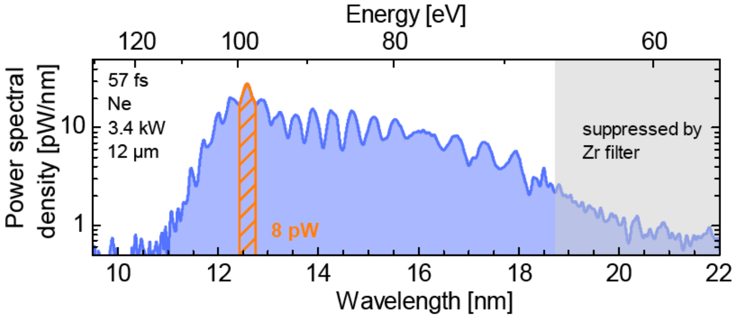
Pierced mirror: Esser et al., Opt. Express **21** (26797) 2013

Intensity Clamping, Generated Harmonics

- Nonlinearly compressed (PCF + chirped mirrors) Yb:fiber pulses → 57 fs, 3.4 kW



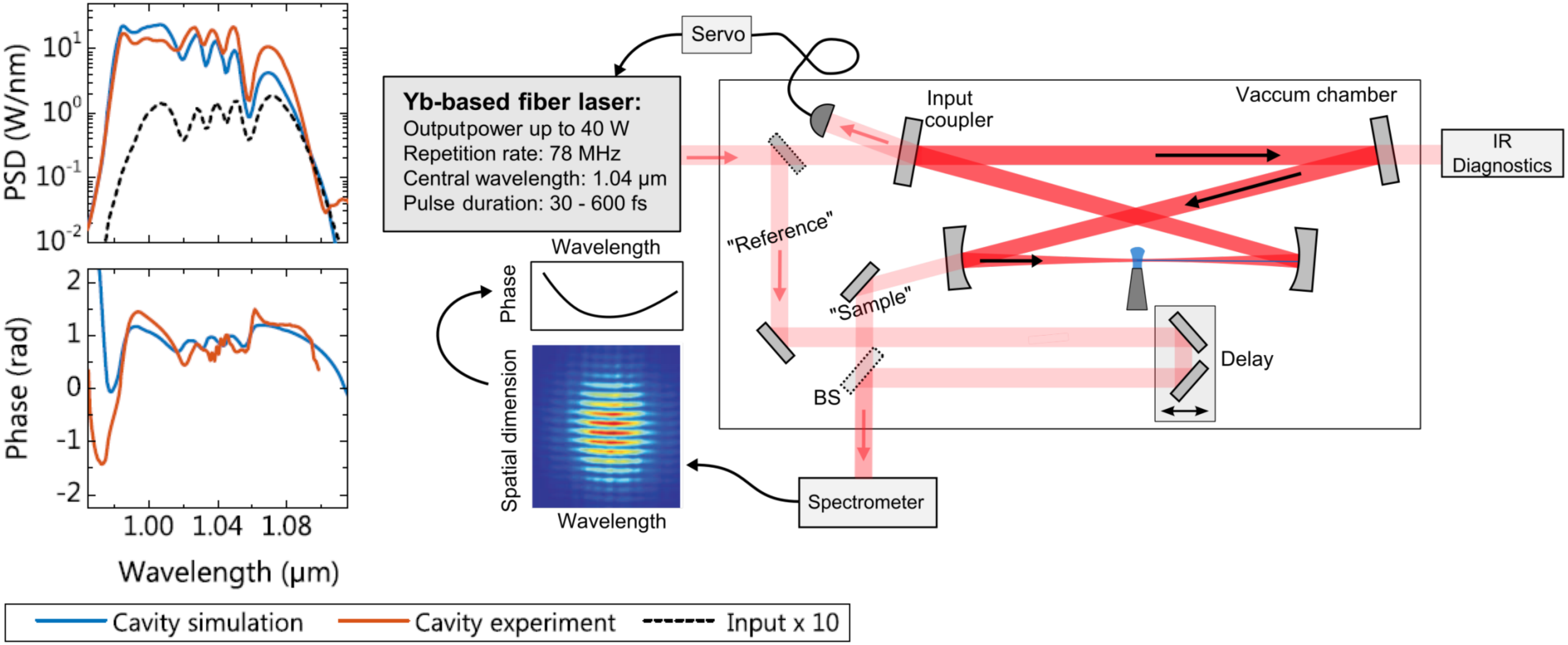




- At 78 MHz: l_{cutoff} = 11 nm
- Shorter pulses: higher intensities, higher conversion efficiency

I. Pupeza et al., Nature Photonics **7**, 608 (2013)

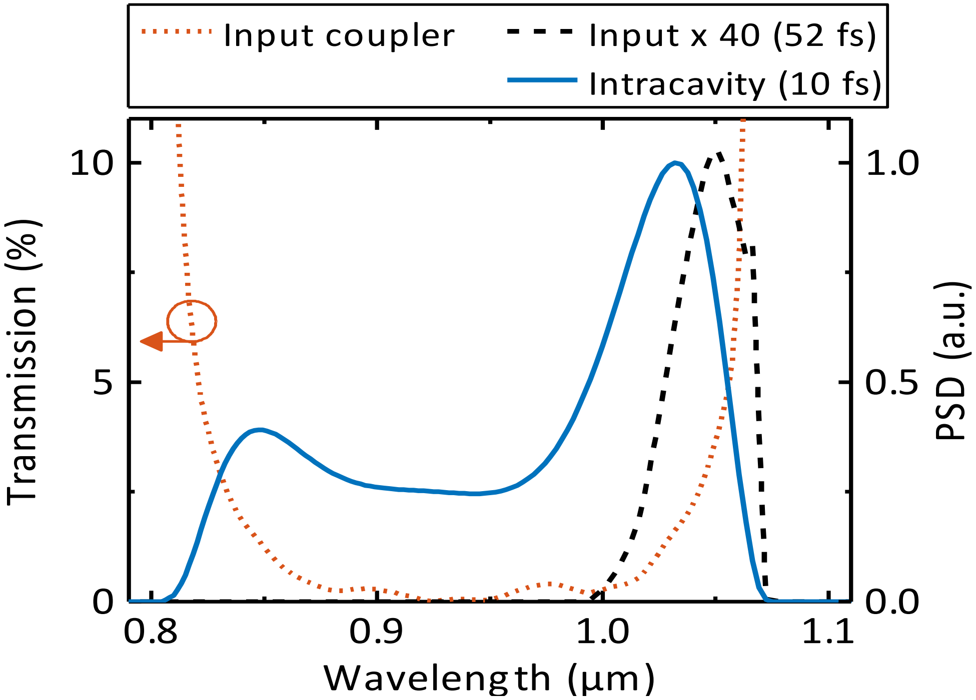
Intracavity Nonlinearities



Yb-based fiber laser:
Outputpower up to 40 W
Repetition rate: 78 MHz
Central wavelength: 1.04 µm
Pulse duration: 30 - 600 fs

S. Holzberger et al., PRL **115**, 023902 (2015)

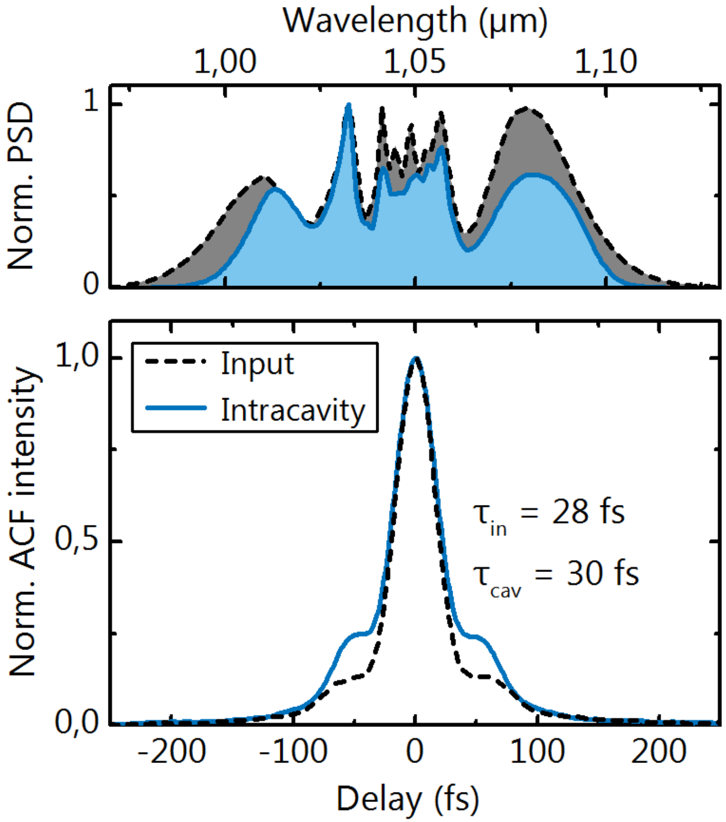
Beyond standard-approach ECs



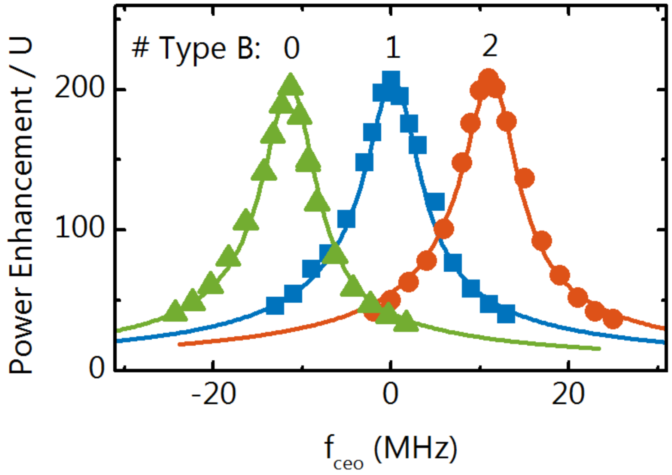
- Average power enhancement: 100 vs. 10
- Peak power enhancement: 450 vs. 10
- Calculated at 8×10^{13} W/cm² in Xe for phase-matched HHG

S. Holzberger et al., PRL **115**, 023902 (2015)

ECs for 0-Offset-Frequency Combs



- Included CEP properties in mirror optimization algorithm.
- Two designs with otherwise the same dispertion properties: type A for type B

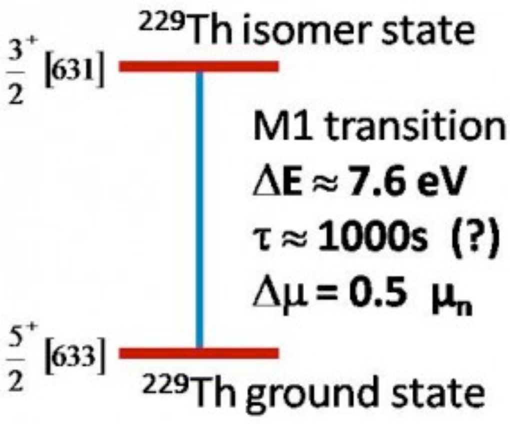


- Bandwidth
- Damage threshold

S. Holzberger et al., Opt. Lett. **40**, 2165-2168 (2015)

Intracavity HHG Applications

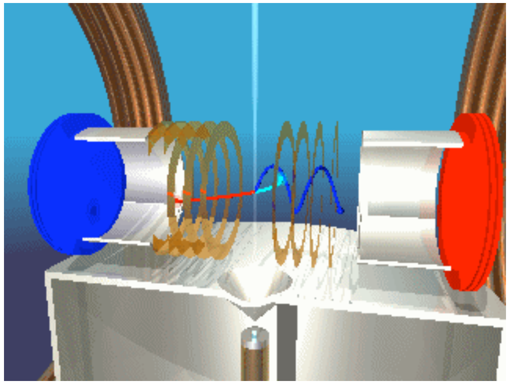
- Precision measurement in the XUV
    - Atoms: He, He$^+$, Li$^+$ … (Lamb Shift $\propto Z^4$)
    - Nucleii: ^{229}Th (7.6 eV), ^{235}U (~75 eV)



- High average power femtosecond XUV light source.
    - Current conversion efficiency only 4 × 10^{-8} (cf. ~ 10^{-5} – 10^{-6} for amplified systems).

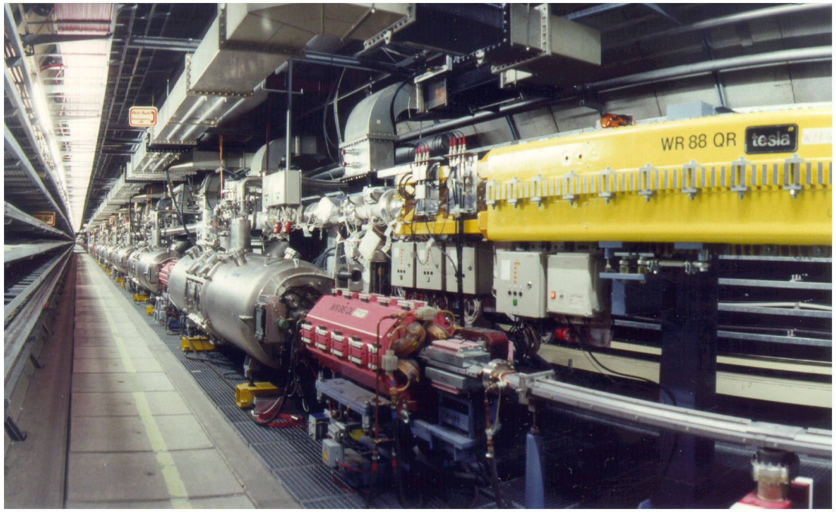
⇒ Still a lot of room to improve!

- High repitition rate applications.
    - Coincidence measurements (e.g. COLTRIMS)
    - Time Resolved ARPES without space charge limitations
    - HHG “heterodyne spectroscopy”



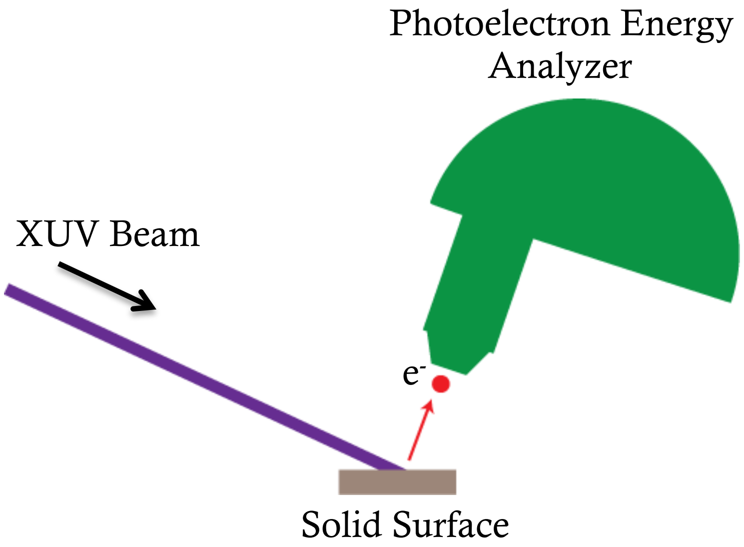
XUV Sources

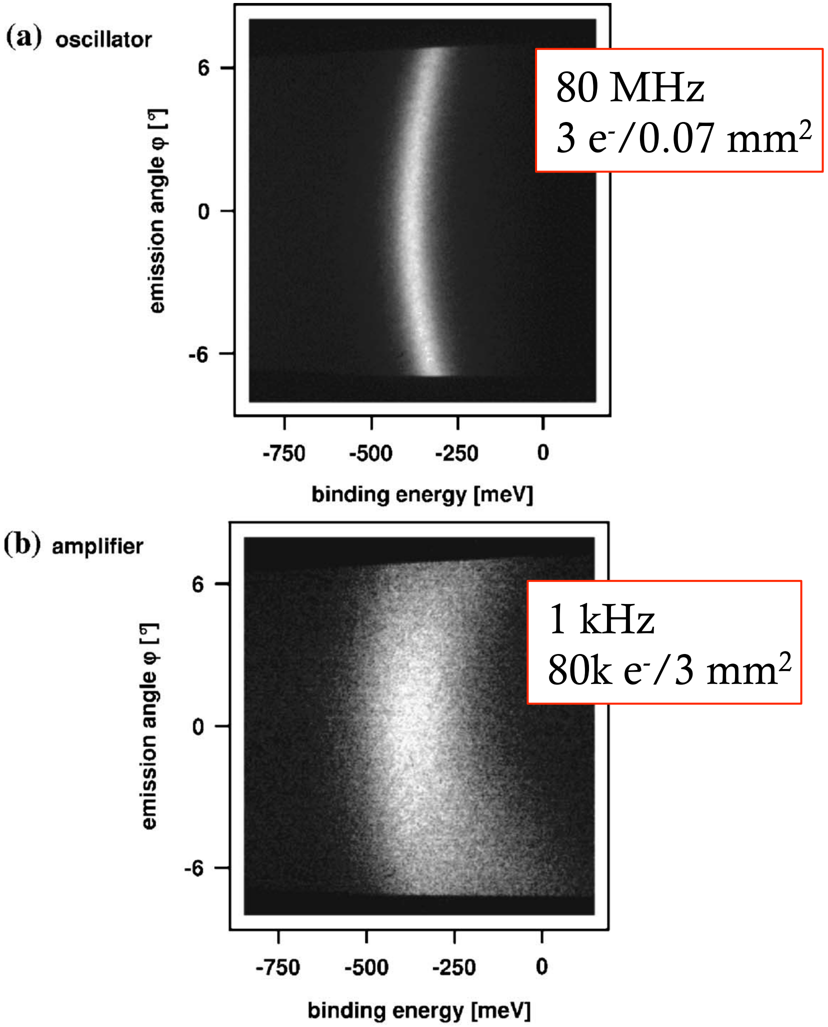
⇒



⇒

Surface Photoelectron Spectroscopy



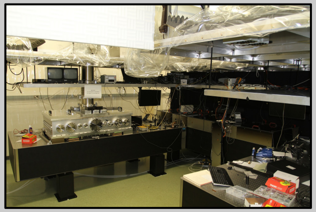


80 MHz
$3\ e^-/0.07\ \mathrm{mm}^2$

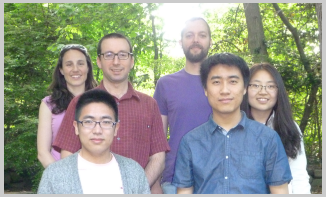
1 kHz
$80\mathrm{k}\ e^-/3\ \mathrm{mm}^2$

S. Passlack *et al.*, *J. Appl. Phys.* **100**, 024912 (2006)

⇒ Surface experiments should be conducted at as high a repetition rate as possible to avoid space charge effects.

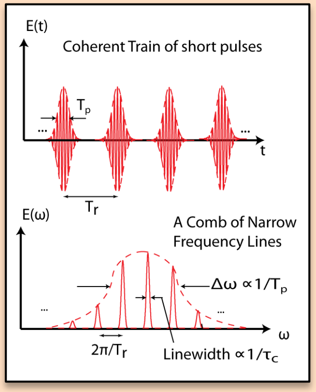


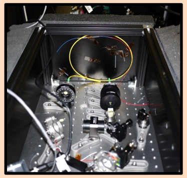
Allison Lab @Stony Brook

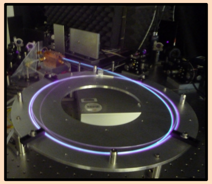


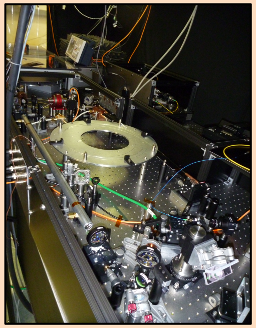
Frequency Comb Lasers

- Optical frequency waveform synthesizer.
- Fiber lasers enable high power.
- Pulses can be coherently added and stored in a passive optical resonator.
- Can convert to ***Mid-IR, XUV, Soft X-ray…***





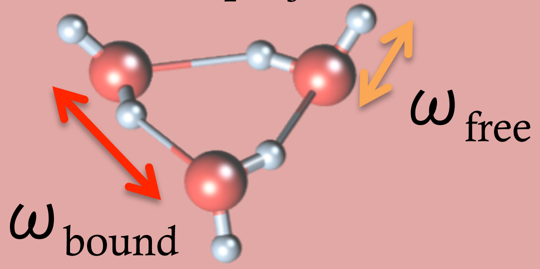




Ultrafast Vibrational Dynamics

- How are vibrational motions coupled in H-bond networks: can we learn from clusters?

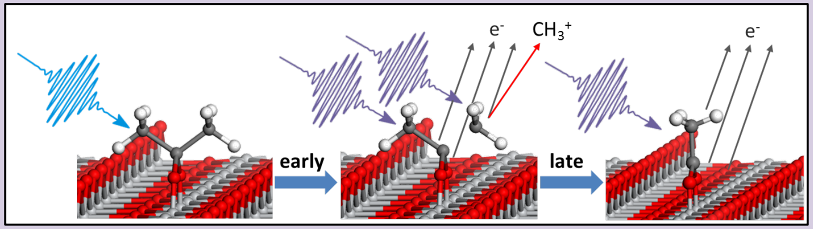
e.g. $(H_2O)_3$



Ultrafast Electronic Dynamics

- How does electronic excitation efficiently drive a chemical reaction?

e.g. Semiconductor Photocatalysis



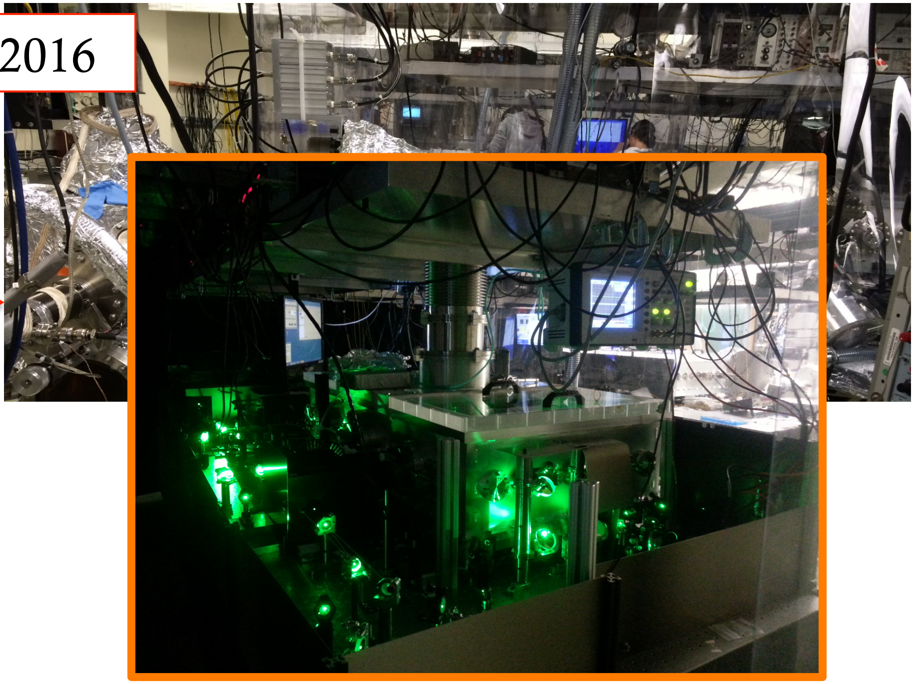
Allison Lab @ Stony Brook

May 2013

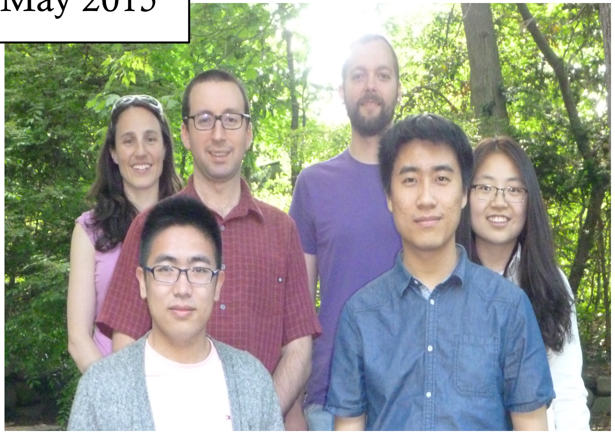
January 2013

Allison Lab @ Stony Brook


May 2013


2016


January 2013


May 2015

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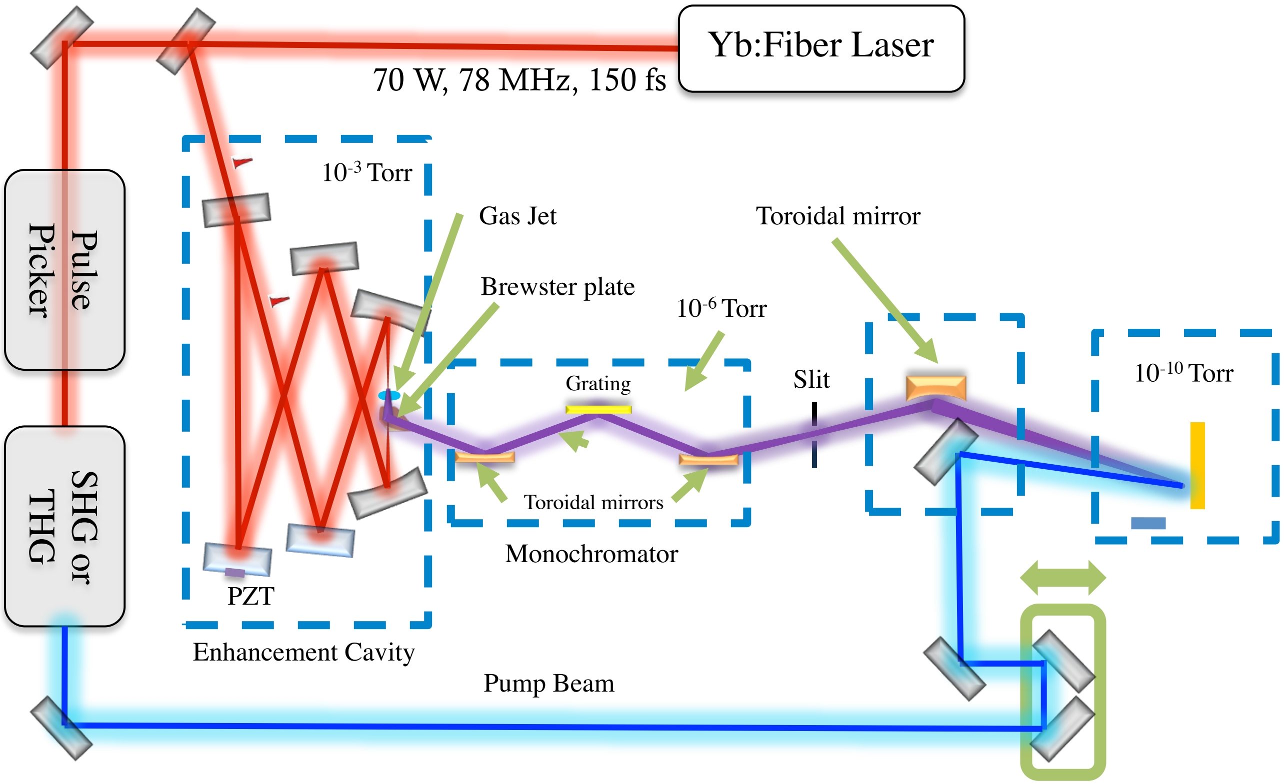


Yb:Fiber Laser

70 W, 78 MHz, 150 fs

Pulse Picker

SHG or THG

10^{-3} Torr

Gas Jet

Brewster plate

PZT

Enhancement Cavity

Grating

10^{-6} Torr

Toroidal mirrors

Monochromator

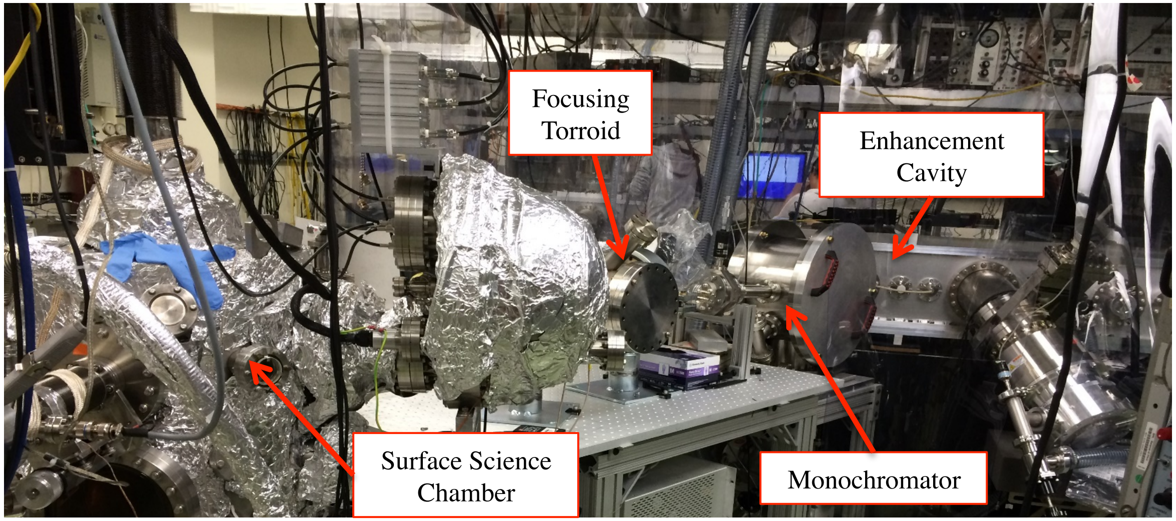
Slit

Toroidal mirror

10^{-10} Torr

Pump Beam

Mike White
BNL and SBU



Focusing Torroid

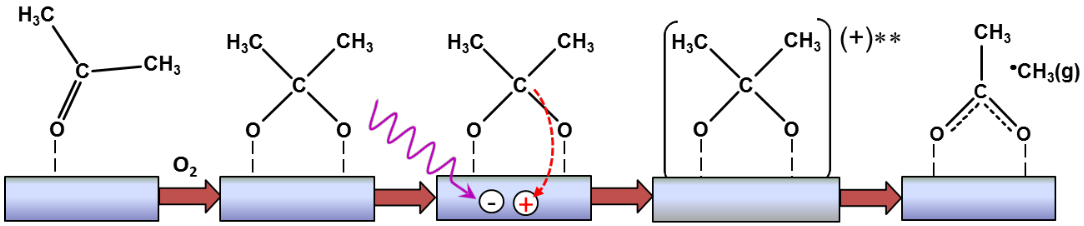
Enhancement Cavity

Surface Science Chamber

Monochromator

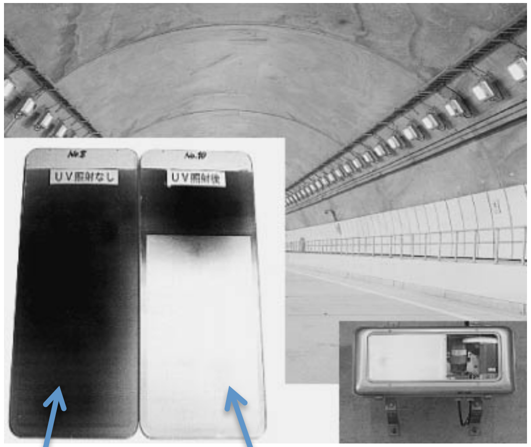
Time-Resolved Photocatalysis

Example: Photooxidation of organics.



Mike White
BNL and SBU

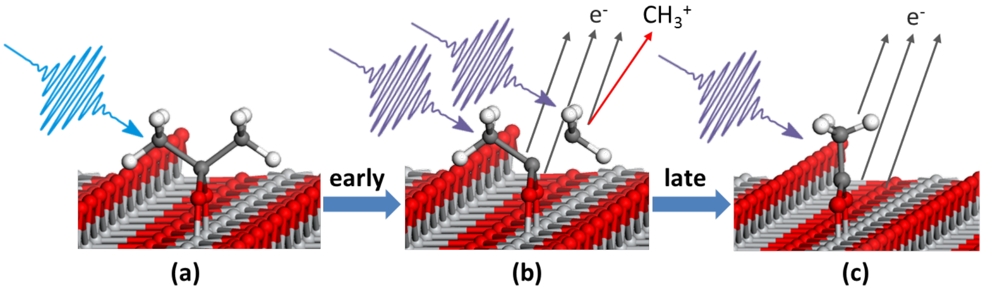
Self Cleaning Surfaces



Glass Light Cover

With TiO_2

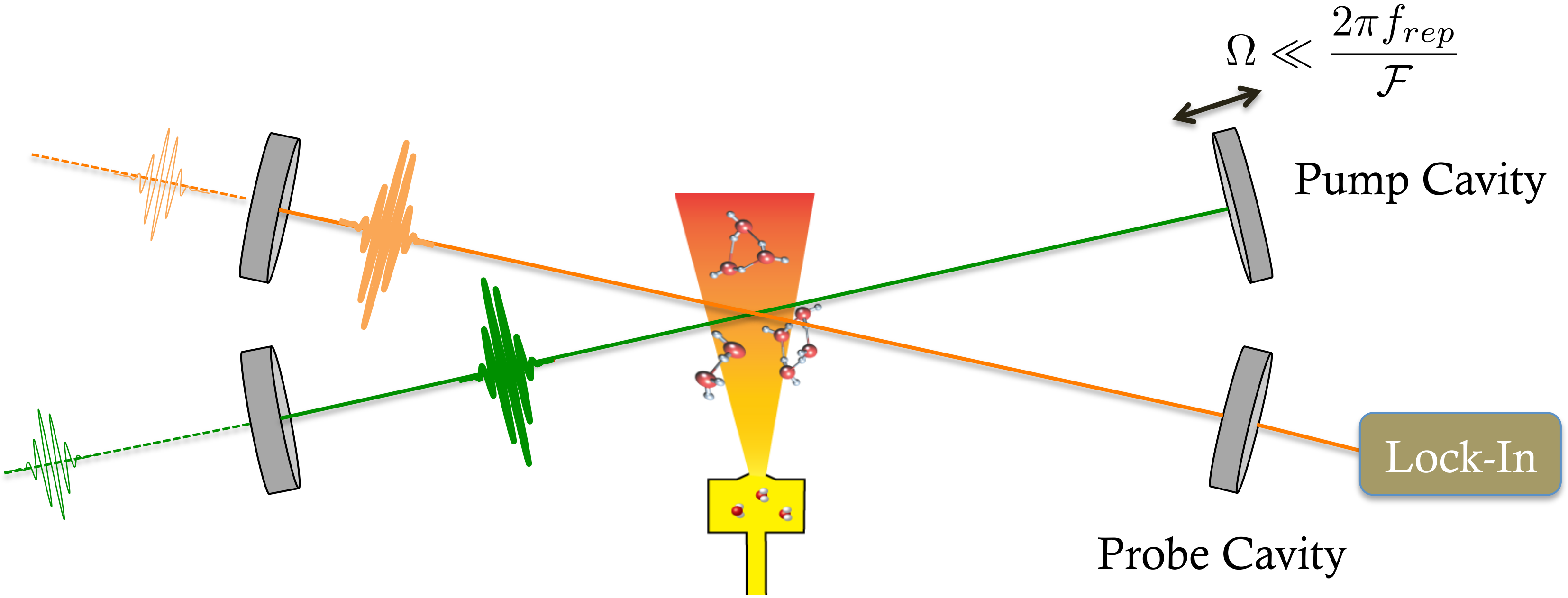
- Many intermediate steps/states!
- Linear spectra show broad features, making inference of the dynamics difficult.



Hashimoto *et al.*, J. J. Appl. Phys. **44**, 8269 (2005)

Technical Goal: Ultrasensitive Femtosecond Spectroscopy

Cavity **E**nhanced **T**ransient **A**bsorption **S**pectroscopy (CE-TAS)



$\Omega \ll \frac{2\pi f_{rep}}{\mathcal{F}}$

Pump Cavity

Lock-In

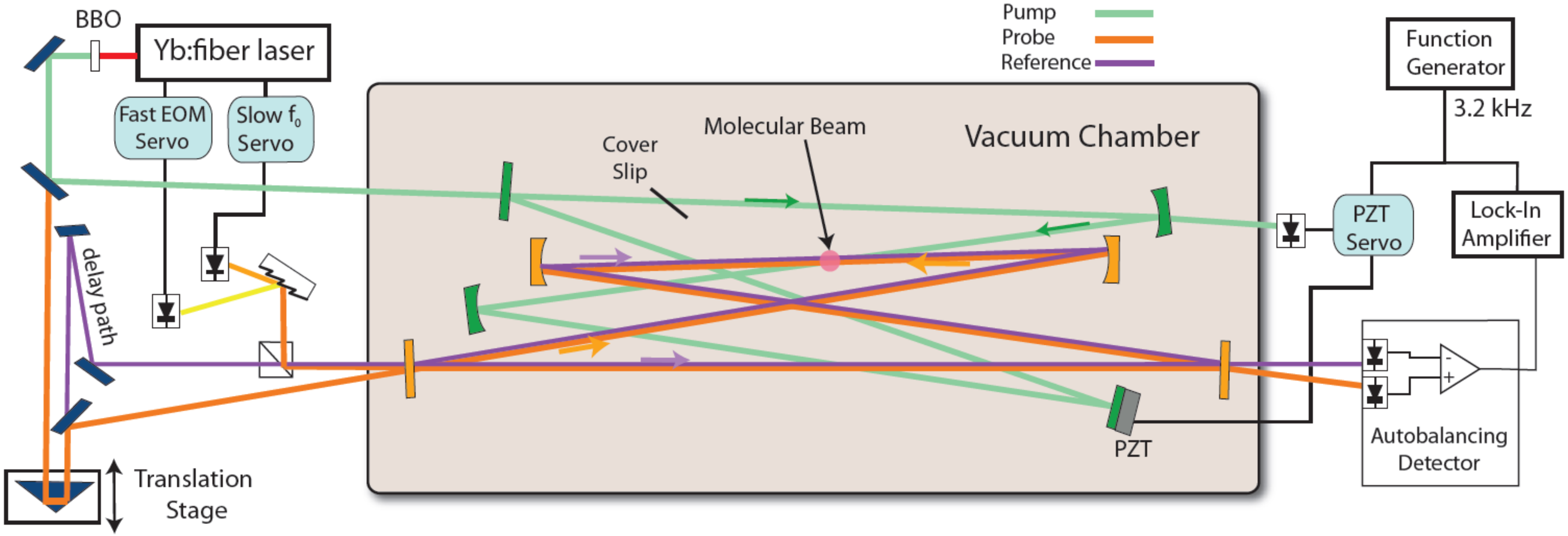
Probe Cavity

- Gain a factor of $2\mathcal{F}_{\text{pump}}/\pi$ for the pump power
- Gain a factor of $\mathcal{F}_{\text{probe}}/\pi$ for the probe sensitivity

$\Bigg\} \Rightarrow$ Signal Enhancement $\propto \mathcal{F}_{\text{pump}}\mathcal{F}_{\text{probe}}$

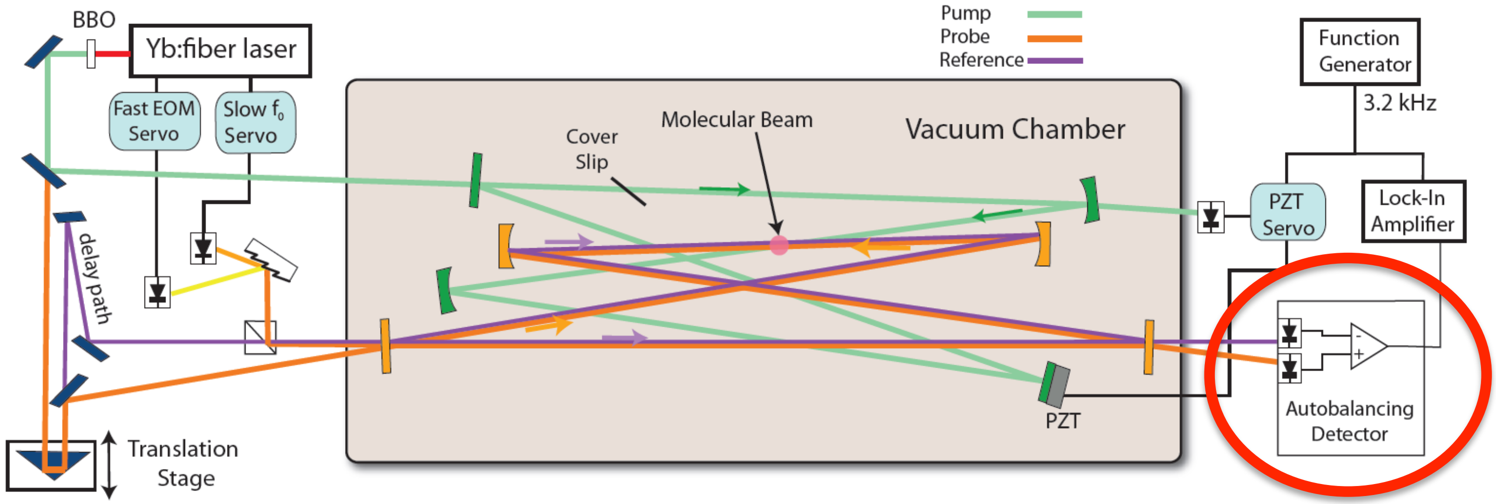
But… cannot turn up pump power arbitrarily. Want ~ 1% sample excitation

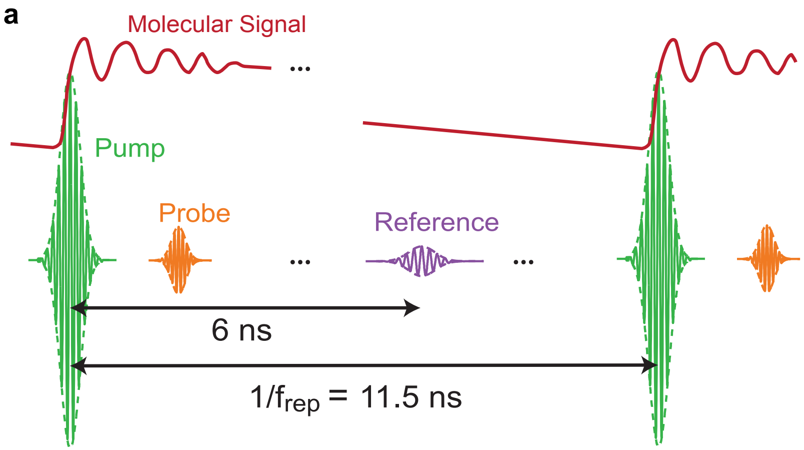
Cavity-Enhanced Ultrafast Transient Absorption Spectrometer: CE-TAS

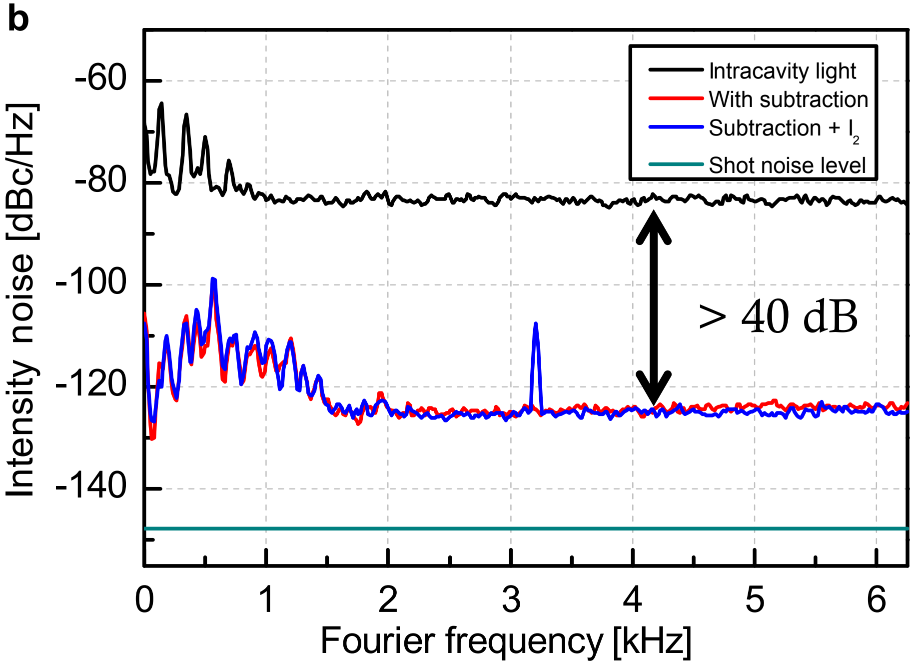


$\mathcal{F}_{probe} = 370$ (Impedance matched ⇒ 120 enhancement)

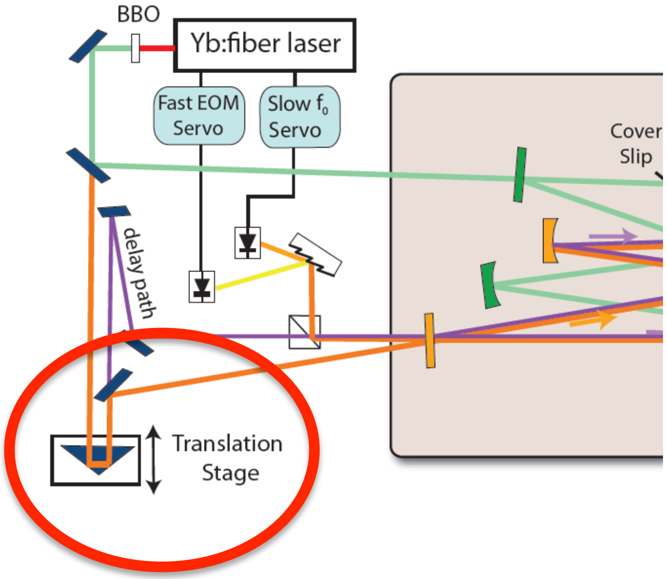
$\mathcal{F}_{pump} \sim 200$ (Overcoupled ⇒ ~50 W average power ⇒ few percent excitation)

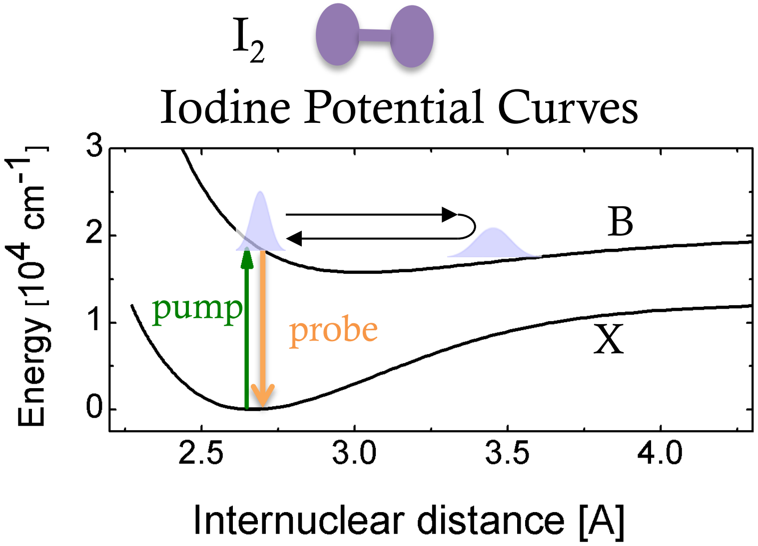

BBO
Yb:fiber laser
Fast EOM Servo
Slow f_0 Servo
Pump
Probe
Reference
Function Generator
3.2 kHz
Molecular Beam
Cover Slip
Vacuum Chamber
delay path
PZT Servo
Lock-In Amplifier
PZT
Autobalancing Detector
Translation Stage


a
Molecular Signal
...
Pump
Probe
Reference
...
...
6 ns
$1/f_{rep}$ = 11.5 ns

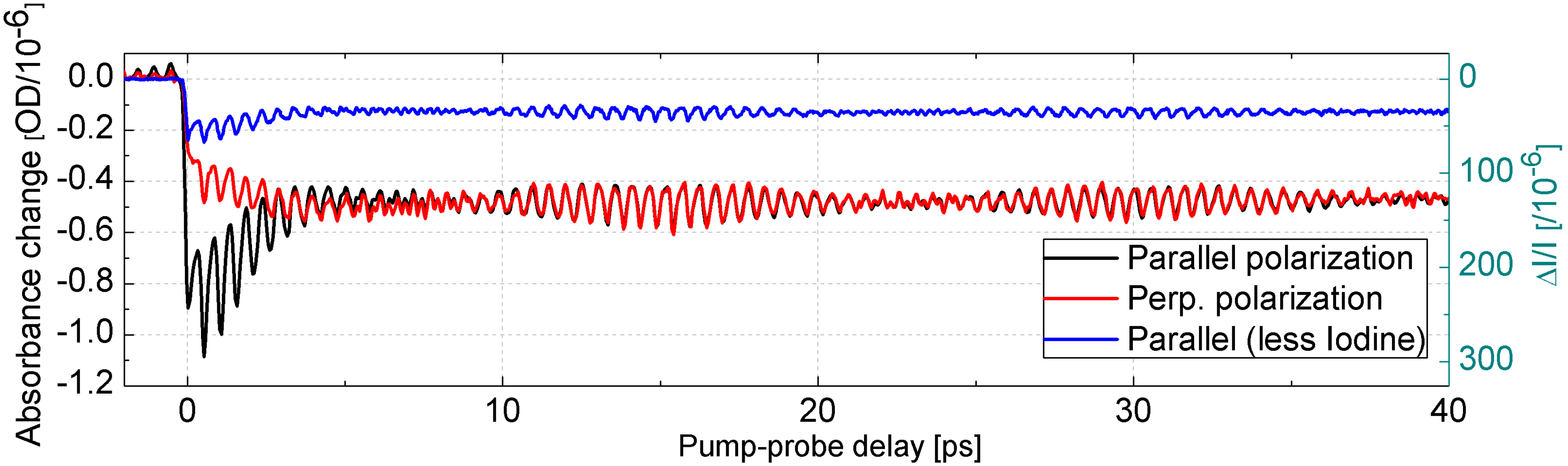

b
Intensity noise [dBc/Hz]
-60
-80
-100
-120
-140
0
1
2
3
4
5
6
Fourier frequency [kHz]
Intracavity light
With subtraction
Subtraction + I_2
Shot noise level
> 40 dB

Transient Absorption Results




I_2

Iodine Potential Curves

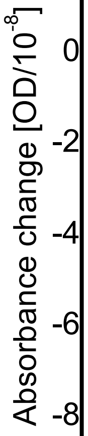


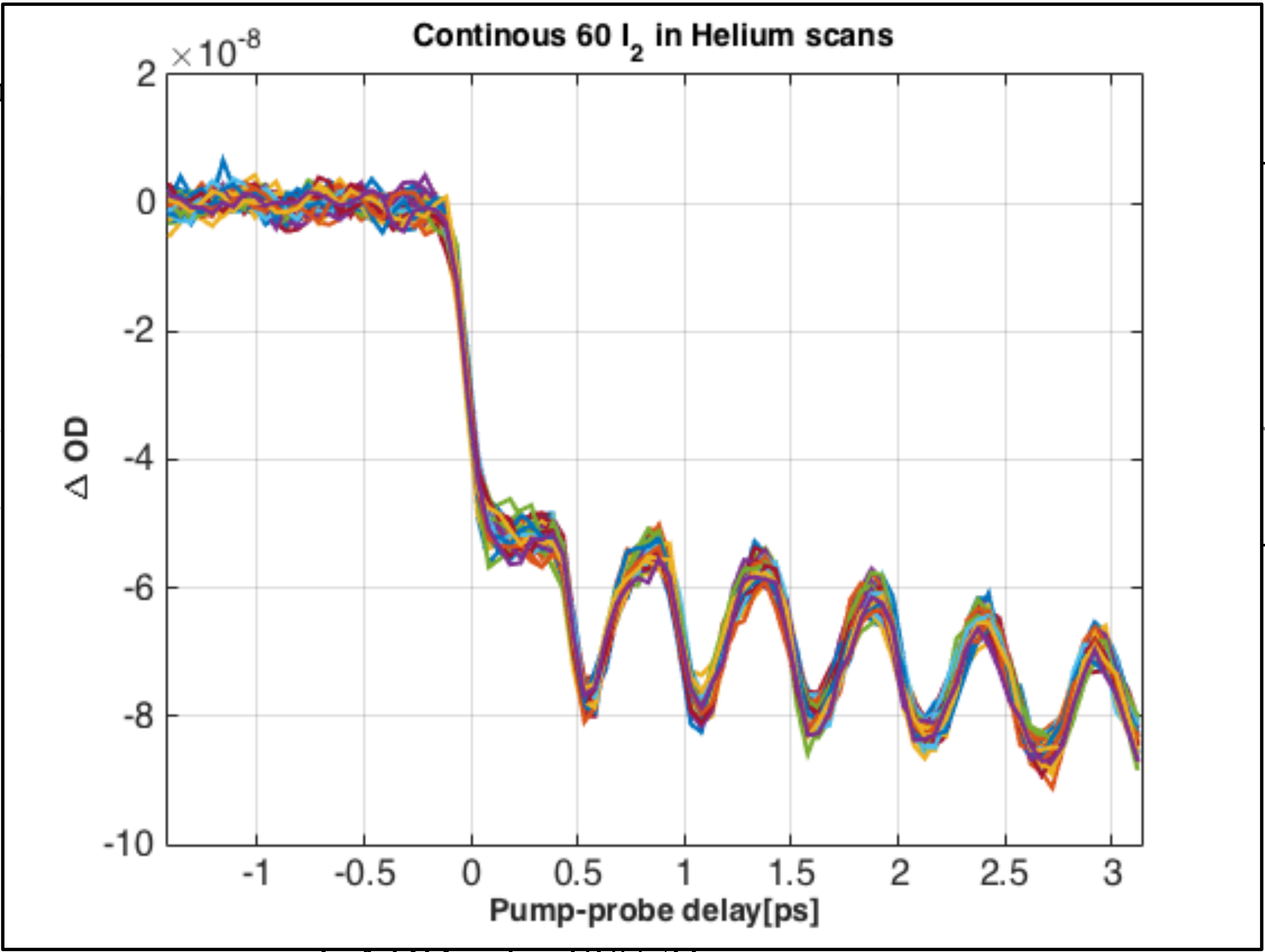
Noise Performance

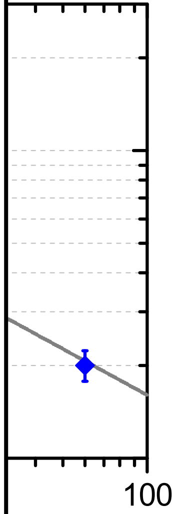
O

Er

-1/2

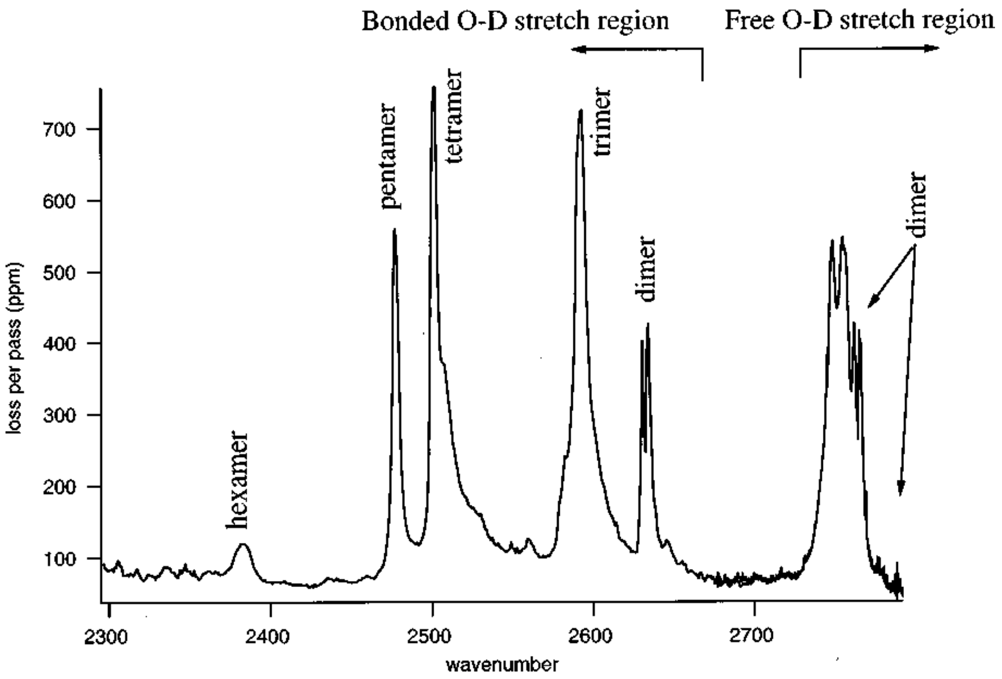




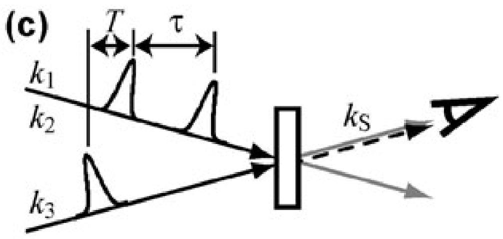


$\Rightarrow$ Δ OD = 1 x $10^{-9}/\sqrt{Hz}$

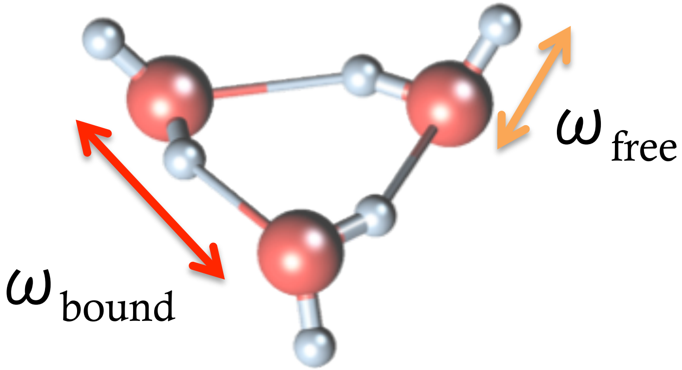
Future: 2DIR

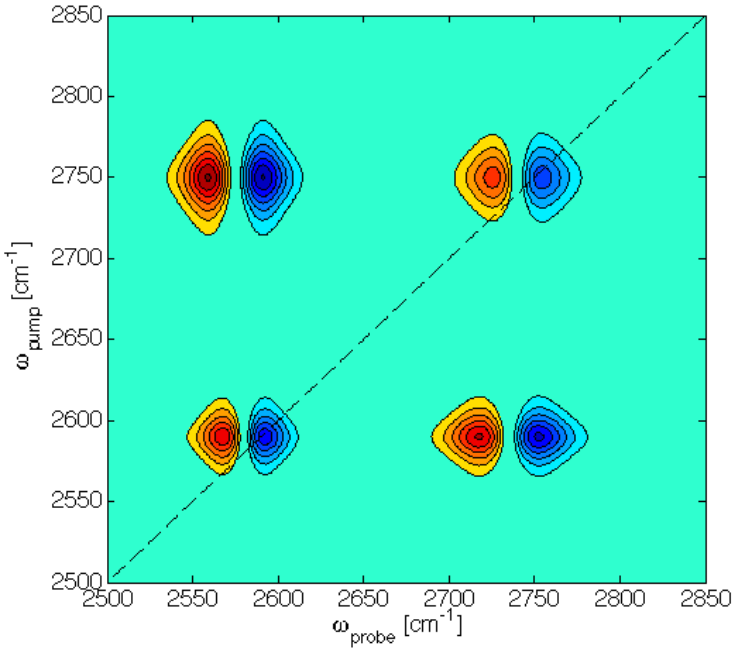


Paul *et al.,* J. Chem Phys **109**, 10201 (1998)

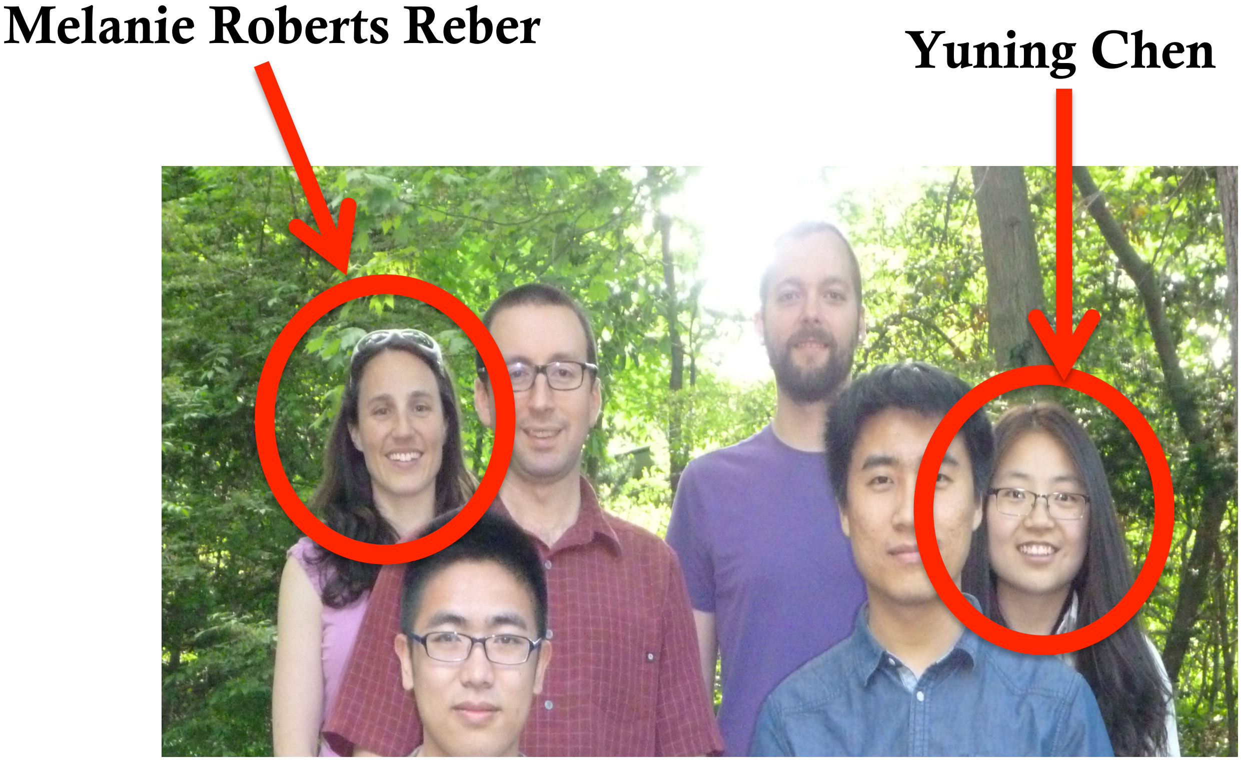


S.H. Shim and M.T. Zanni, PCCP **11**, 784 (2009)





Acknowledgements!



Equipment Loans: Trevor Sears and Mike White (SBU/BNL)

Paper: M. A. R. Reber, Y. Chen, and T. K. Allison, arXiv:1511.02973 (2015)
Recently accepted at *Optica*