Backward lasing in air from oxygen, nitrogen, hydrogen and argon

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Workshop on Oxygen Plasma Kinetics
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Financial support: ONR and MetroLaser

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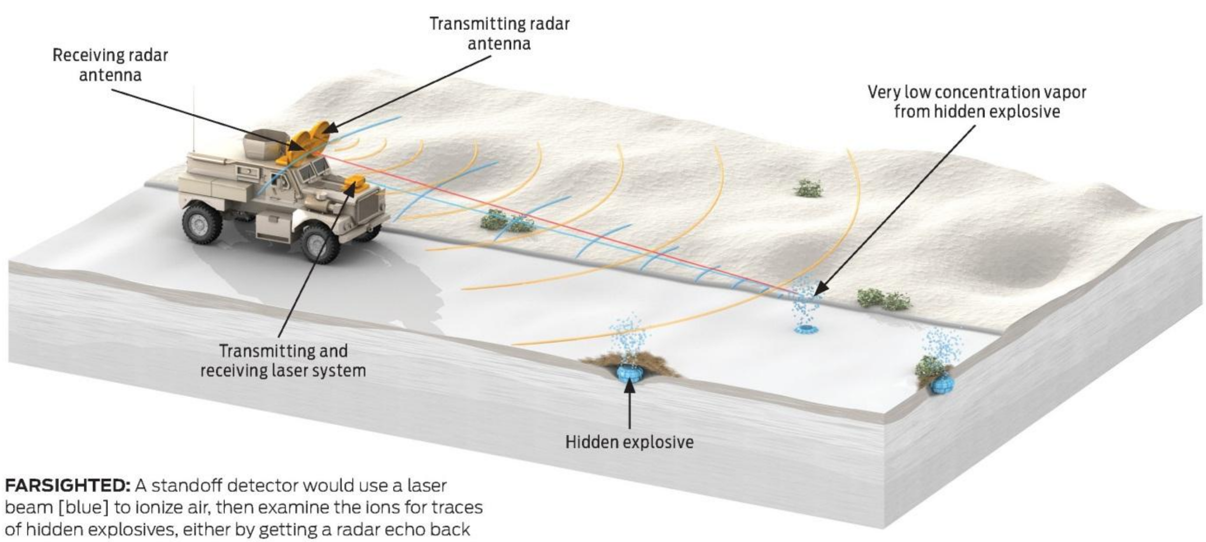
IEEE Spectrum — INSIDE TECHNOLOGY



FARSIGHTED: A standoff detector would use a laser beam [blue] to ionize air, then examine the ions for traces of hidden explosives, either by getting a radar echo back to a dish on top of the vehicle or via a backward-propagating "air laser" beam [red]. *ILLUSTRATION: BRYAN CHRISTIE DESIGN*

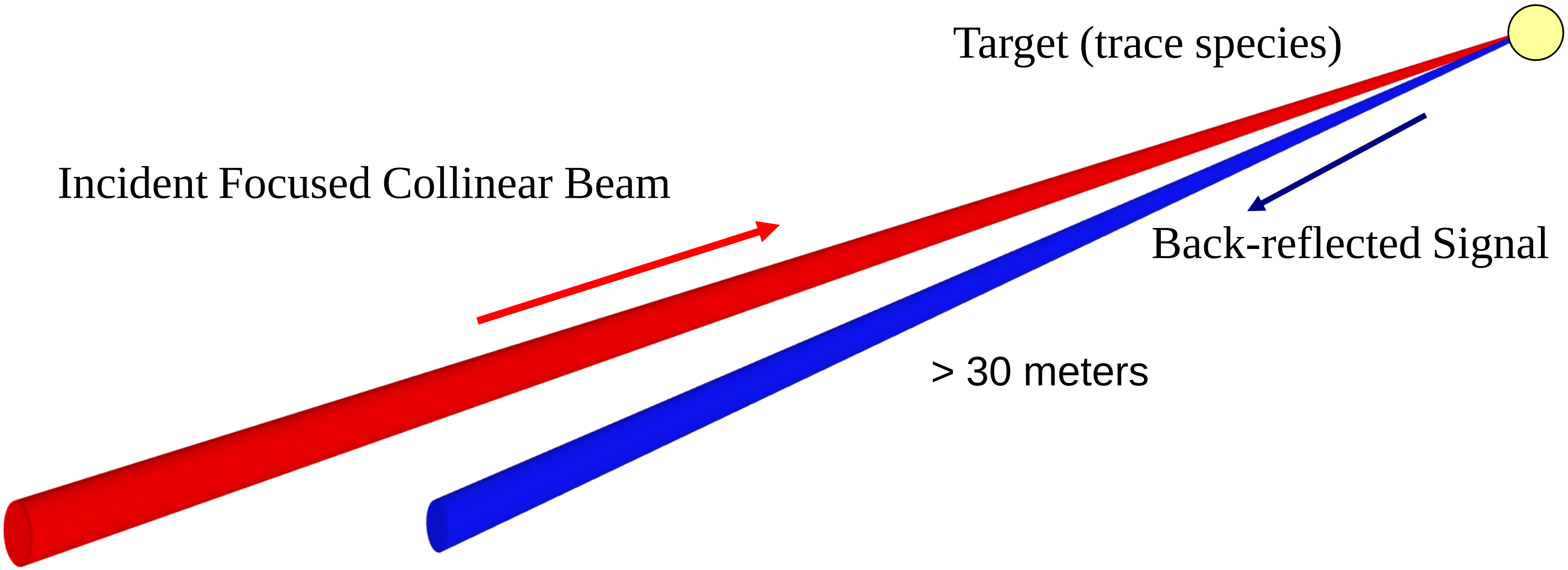
WALK SOFTLY: A mine-clearer's work still depends on metal detectors and sniffer dogs, with all the dangers that this entails. Technology may provide a better way to do the job. PHOTO: UNITED STATES MARINE CORPS

Bringing BOMBS to Light

With a mighty zap, a laser can detect fumes from hidden explosives

BY RICHARD B. MILES, ARTHUR DOGARIU & JAMES B. MICHAEL

Motivation for remote air lasing



- Laser-based remote trace species detection methods rely on backscattered light
- Incoherent light is non-directional, direct coherent light has the wrong direction!
- Need for coherent light source at the target – **remote laser source**

High Gain Backward Lasing in Air

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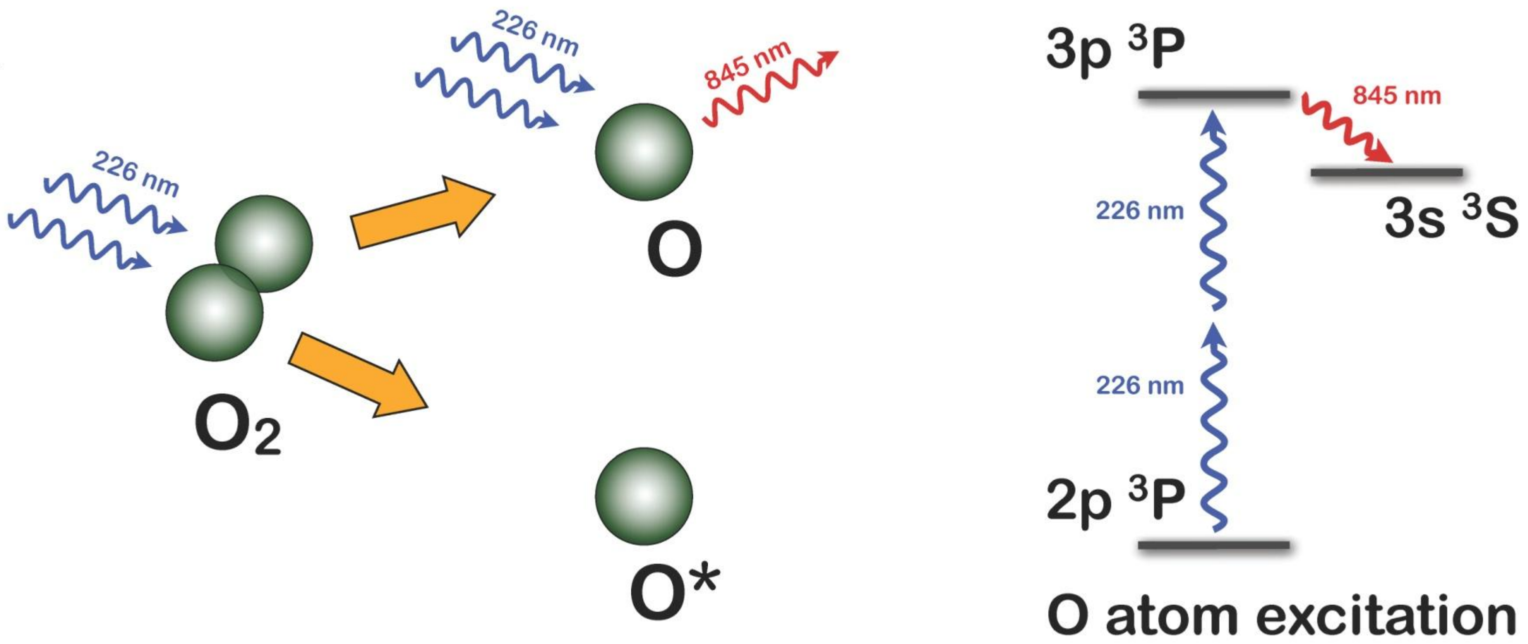

Two-photon dissociation of O_2 molecule and excitation of the O atom,
All with a single 226 nm UV pulse.

Science AAAS

High-Gain Backward Lasing in Air
Arthur Dogariu, *et al.*
Science **331**, 442 (2011);
DOI: 10.1126/science.1199492

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Dissociation of O2 with 225.67 nm

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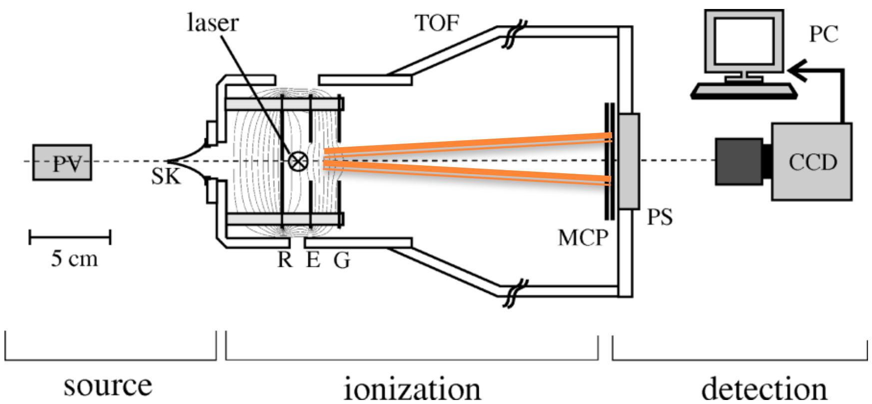
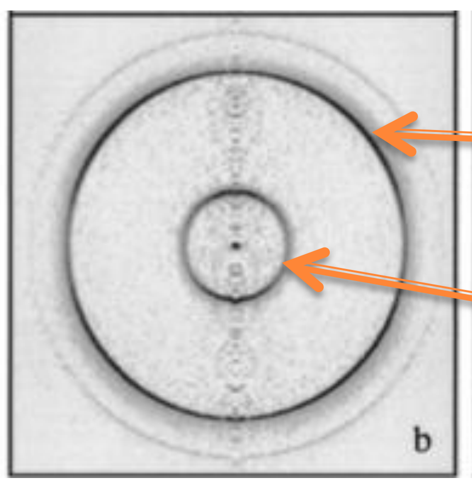
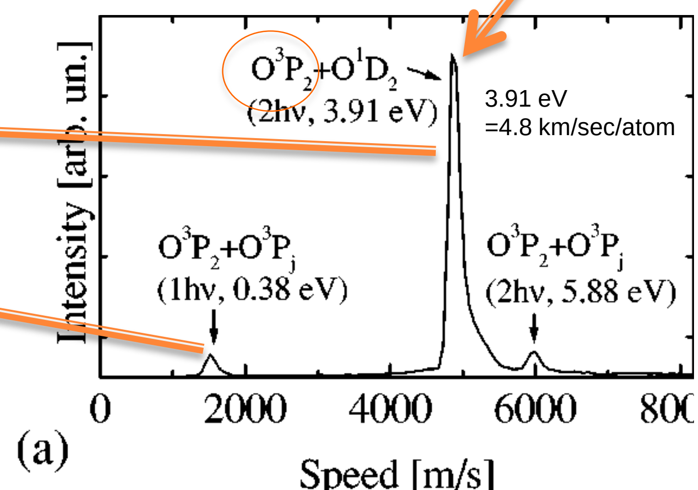


225.67 nm = 5.494 eV > O_2 dissociation

Dissociation is enhanced by two photon resonance with pre-dissociative Rydberg states

Kinetic energy release ion imaging –the radius of each circle is proportional to the kinetic energy of the ions





B. Buijsse, et al, "Angular distributions for photodissociation of O_2 in the Herzberg Continuum," J. Chem. Phys., Vol. 108, No. 17, 1 May 1998

The Oxygen Air Laser

2 photon Dissociation and 2 photon pumping all with a single laser pulse at ~226 nm

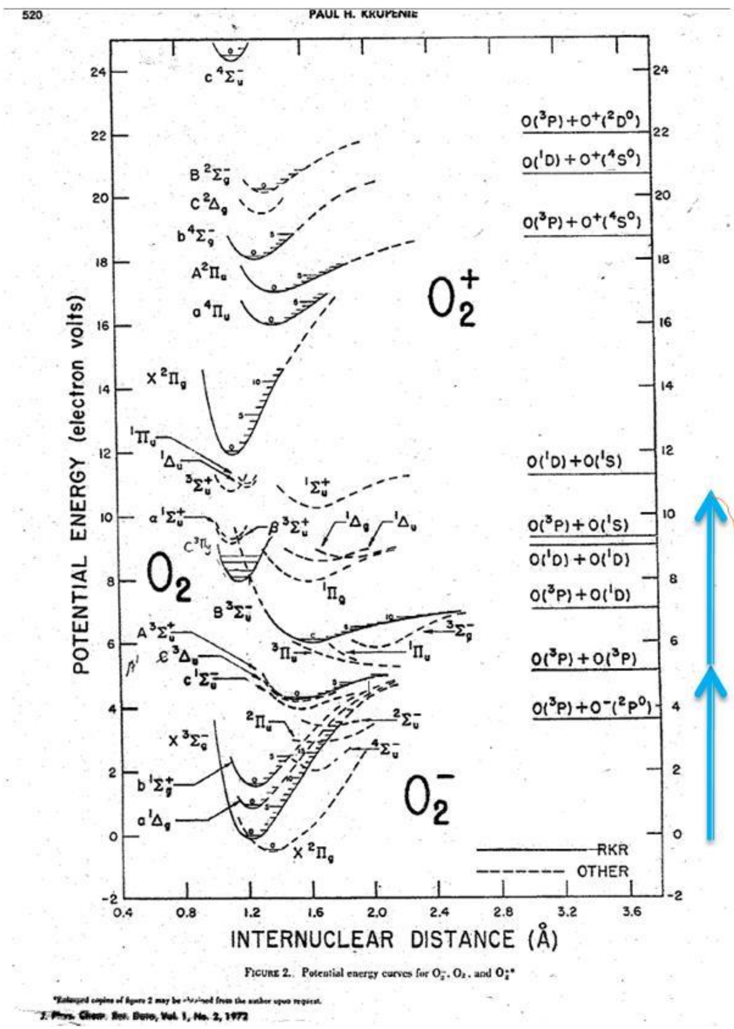
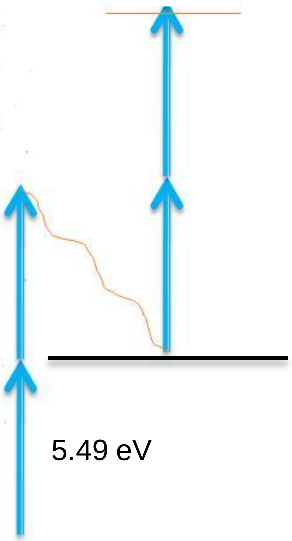


FIGURE 2. Potential energy curves for O_2^-, O_2, and O_2^+



Experimental Setup

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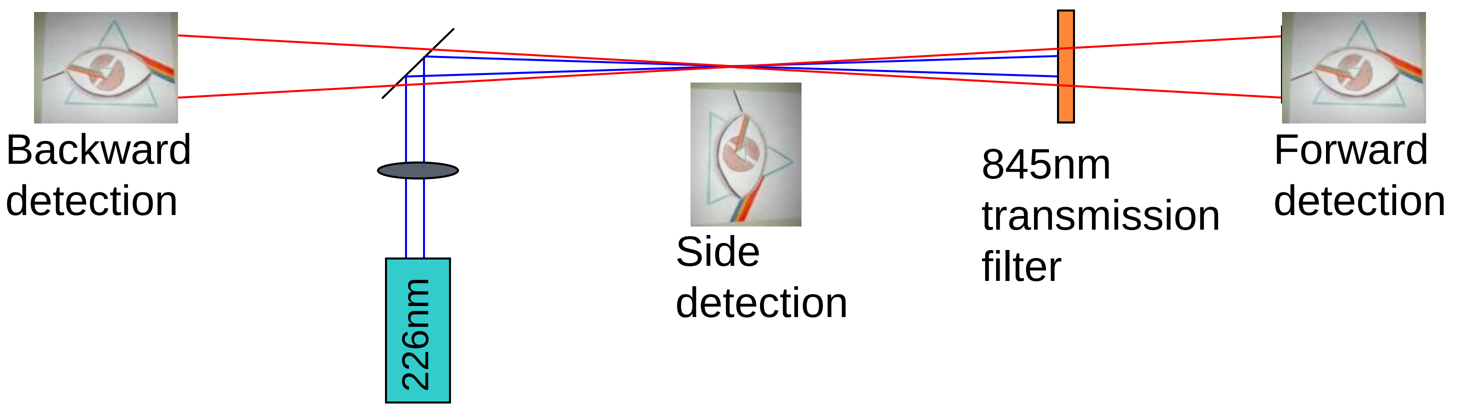
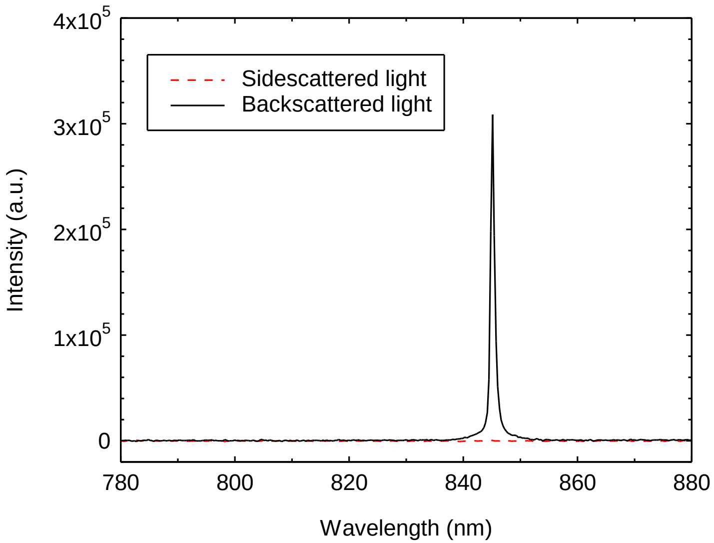
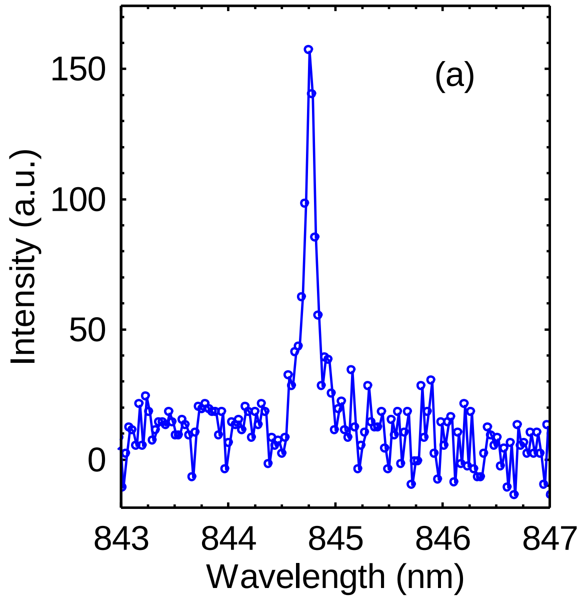


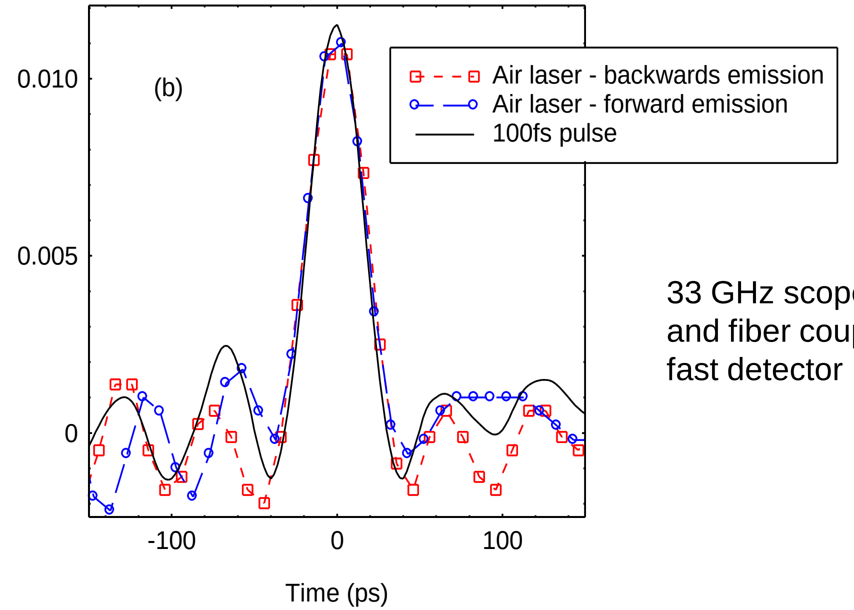
Detection = CCD, detector, PMT, spectrometer



- Emission at 844.6 nm (atomic oxygen line)
- Coherent emission along the pump laser line (forward and backward)
- Very low level detected from the side

845 nm atomic oxygen emission





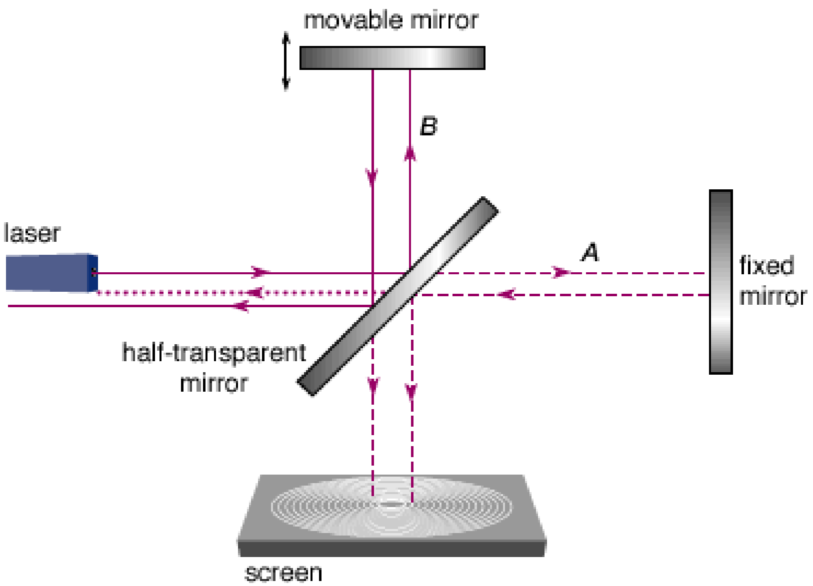
33 GHz scope and fiber coupled fast detector

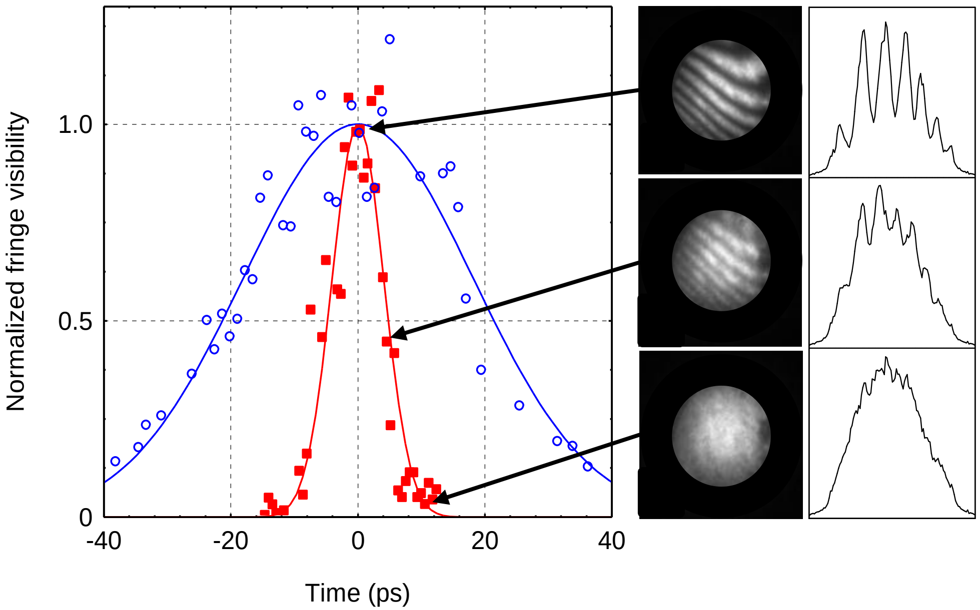
- 100 picosecond pump laser
- Backward propagating atomic oxygen emission measured spectrum (a) and pulse in time (b).
- Pulse width < 30ps
- Atomic oxygen lifetime: 34ns!

→ Coherent emission

Temporal Coherence

- Michelson - Morley interferometer – first order autocorrelation, measures coherence time (given by the laser bandwidth).





Bandwidth limited pulses!

10 cm focusing – 10 ps coherence time

30 cm focusing – 35 ps coherence time

Coherence time ≅ pulse width → transform limited

Donut vs. Gaussian mode

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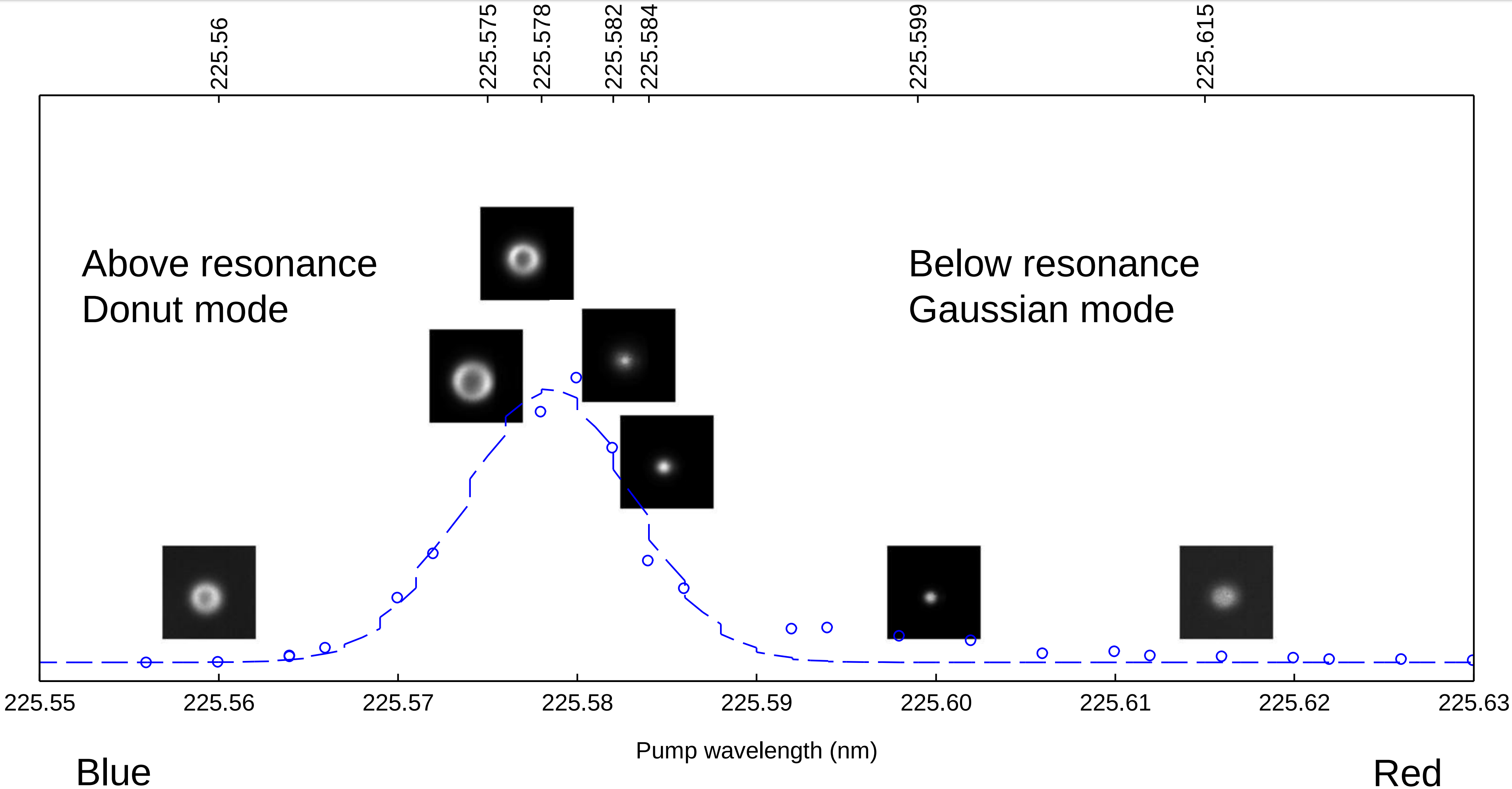
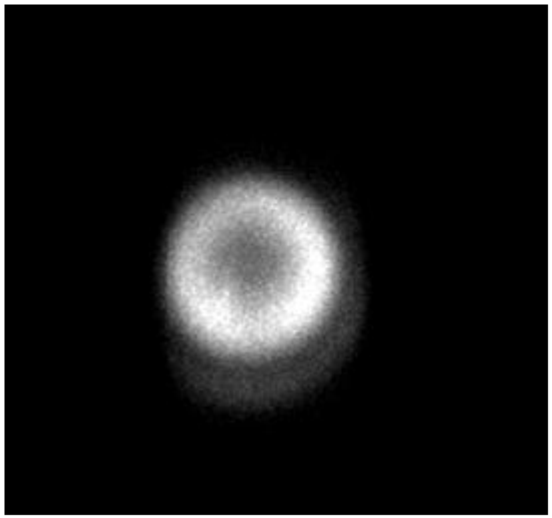
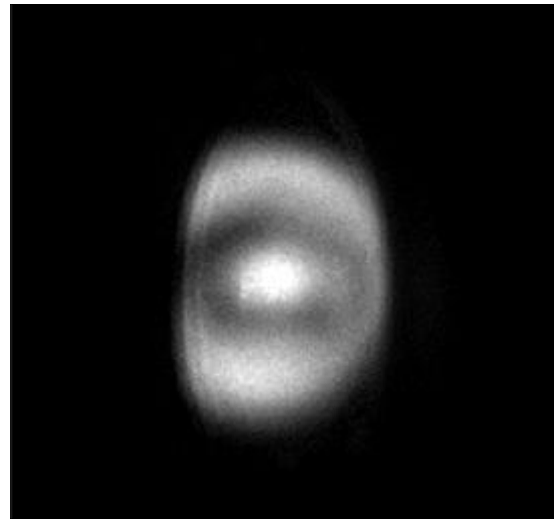


Excitation Spectrum

Beam profiles – back emission



Air



Air with pre dissociation

Backscattered oxygen laser beam at 845nm measured 30cm away from the 226nm laser focused in air (left), and in air with a 532nm pre-pulse (right).

Pre-pulse (5μs before resonant UV pulse) dissociates oxygen molecule and generates 100 times stronger atomic oxygen lasing emission.

Dogariu et al., "High Gain Backwards Lasing in Air," *Science* **331**, 442 (2011).

Beam divergence – Spatial Coherence

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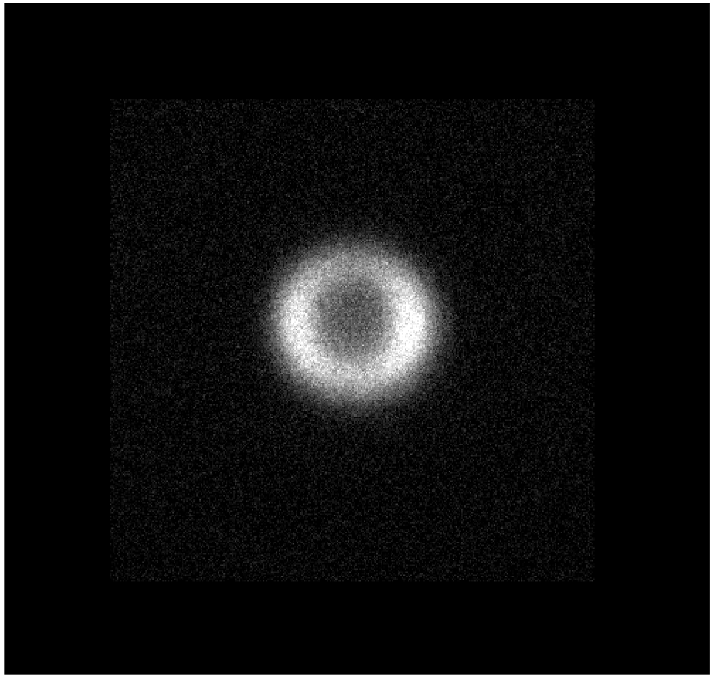

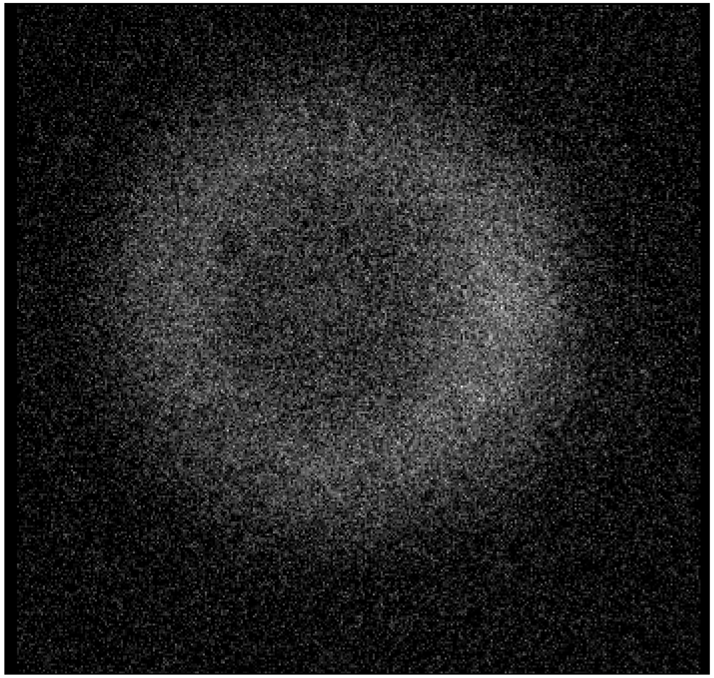

Distance from source	25 cm	75 cm
O emission beam size	1.1 cm	3.1 cm
UV pump beam size	3 mm	9 mm

Divergence: 4.7 degrees oxygen laser @ 845nm
1.43 degrees pump laser @ 226nm

$\theta_{oxygen} / \theta_{UV} = 3.3$

$\lambda_{oxygen} / \lambda_{UV} = 3.7$

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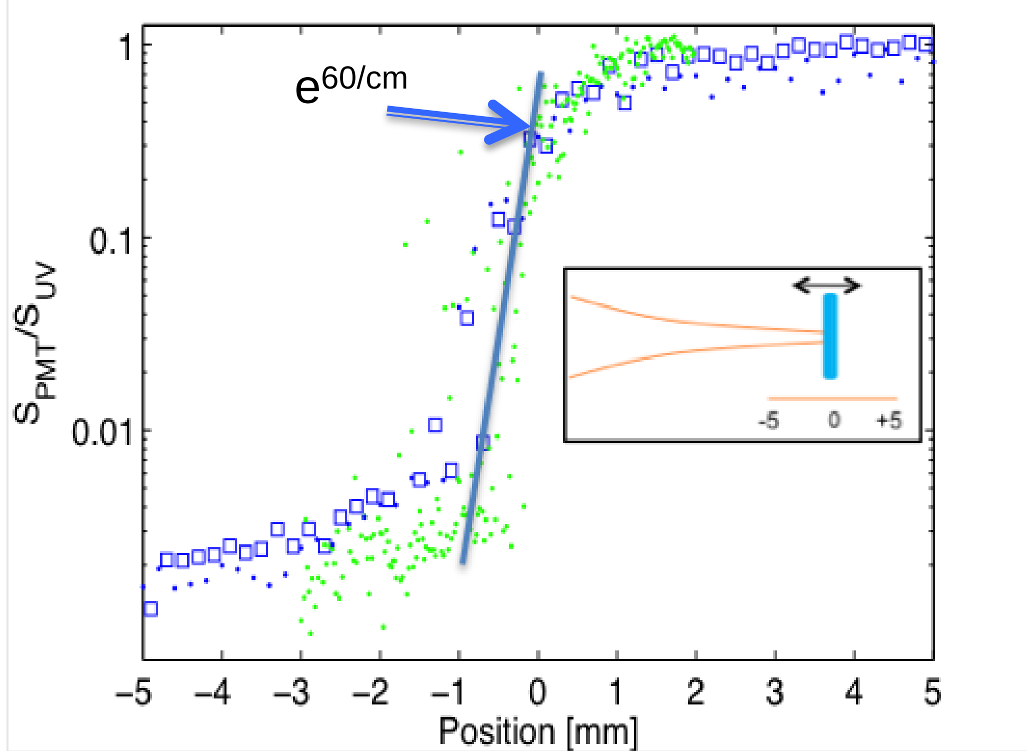
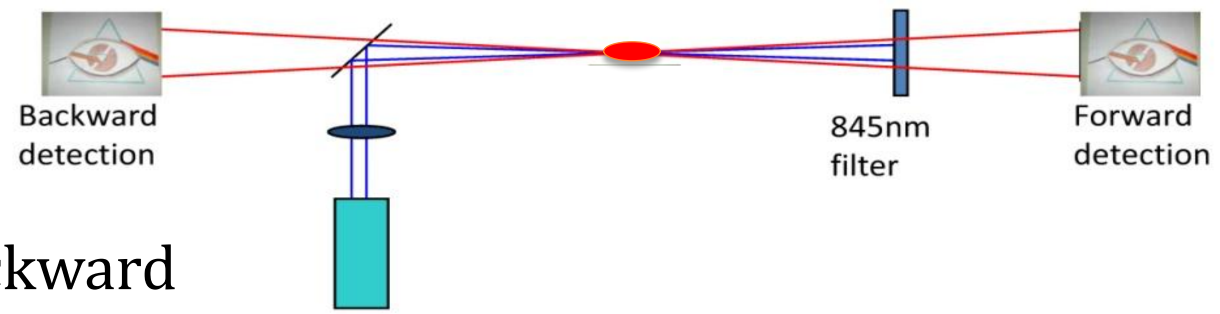
Back Emission vs. gain length

L/d = 100 - 500

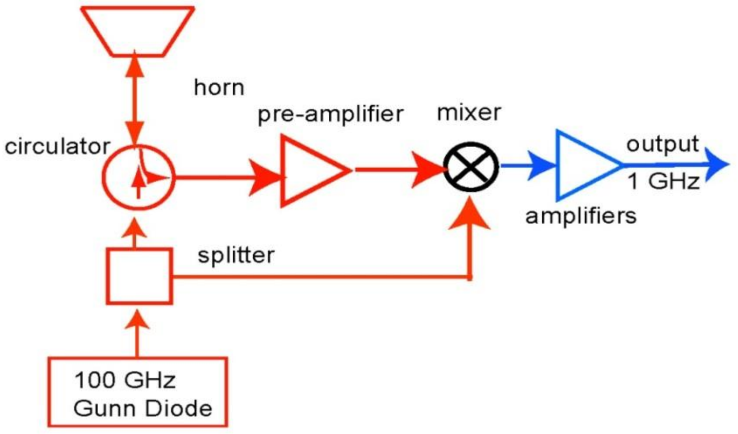


- Backwards emission signal normalized by the maximum backward lasing pulse energy vs. the position of the glass slide gain termination location.
- A glass slide used to terminate the pump beam propagation is scanned through the focal zone range of the pump beam while the backwards emission is monitored.
- The rapid growth in the signal moving from a position of -1 to 0 mm (at least two orders of magnitude) shows the nonlinearity with the gain path length. Gain coeff. 40-80 cm^{-1}

Air laser and Radar REMPI:
Lasing relative to excitation population



- Forward and backward detectors monitor the emission (lasing)
- The 100 GHz microwave system monitors the Radar REMPI signal (ionization)
- The Radar REMPI signal measures the density of excited oxygen atoms



REMPI – Resonantly Enhanced Multi-Photon Ionization
RIS – Resonance Ionization Spectroscopy

2+1 REMPI probes excited state

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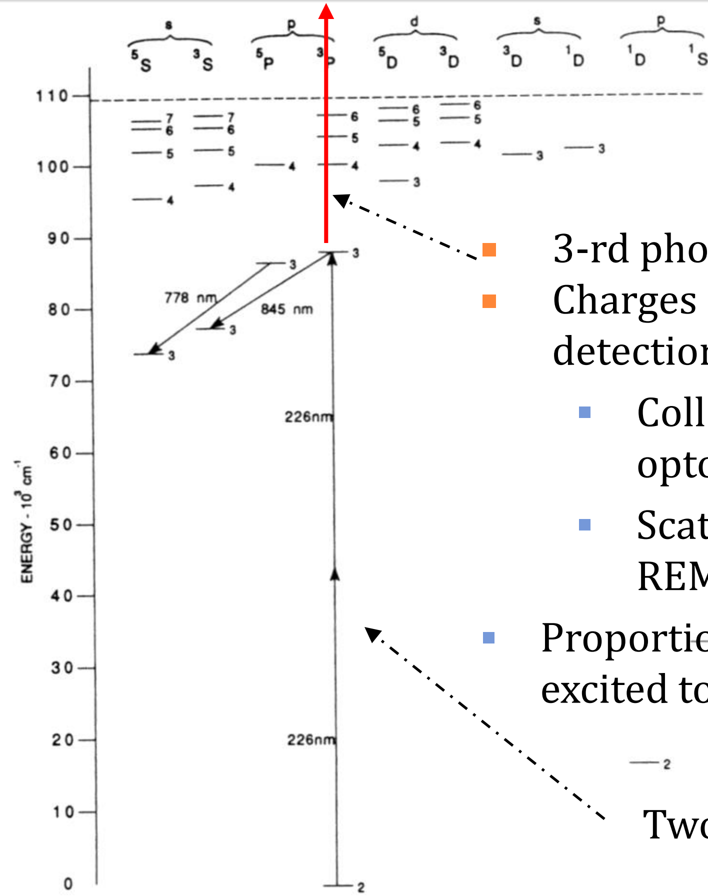
Resonantly Enhanced Multi-Photon Ionization

- An intense laser beam ionizes the atom and creates charges/plasma.
- The ionization is strongest when the photon(s) energy equals the energy difference between excited and ground state.
- Extra photons bring the energy above the ionization energy of the atom (the energy required to remove one electron from an isolated, gas-phase atom).
- Oxygen: 2+1 REMPI = 2 photons to excite and 1 to ionize.



- 3-rd photon produces ionization
- Charges provide means of detection:
  - Collected using electrodes – opto-galvanic spectroscopy*
  - Scatter microwave – Radar REMPI
- Proportional to the oxygen atoms excited to the resonant state

Two-photon excitation

*J. E. M. Goldsmith, "Resonant multiphoton optogalvanic detection of atomic oxygen in flames," *J. Chem. Phys.* **78**, 1610-1611 (1983).

Exponential Power Scaling

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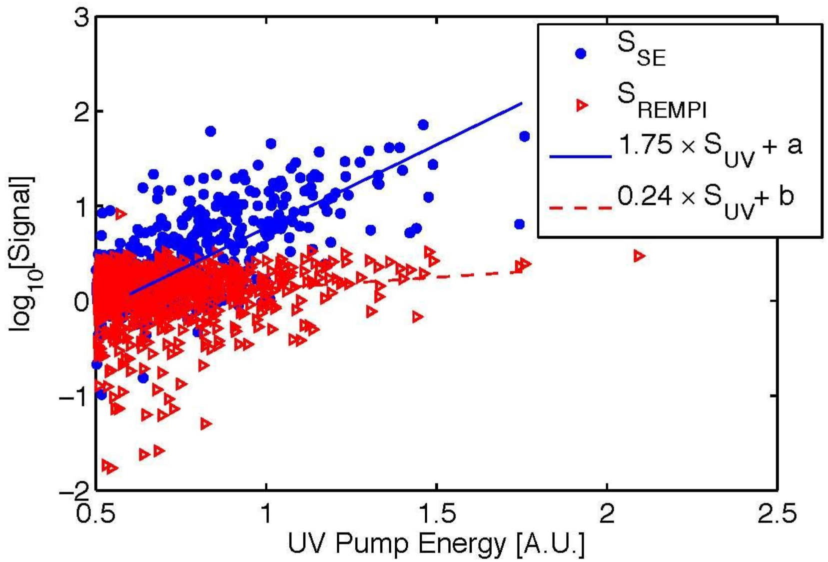

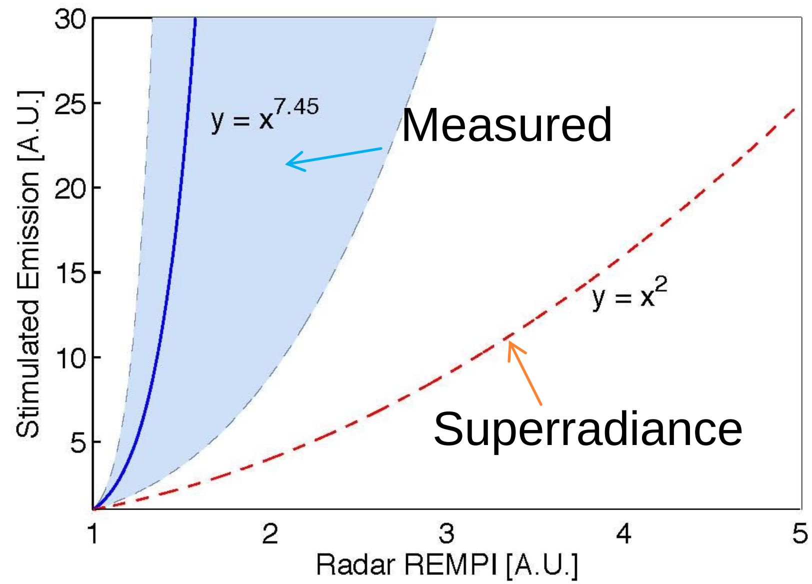

- $\text{Signal} \sim UV^k$
- Radar REMPI $k = 0.24$
- Stimulated Emission $k = 1.75$

- Radar REMPI (ionization) signal $\sim N$
- Stimulated emission signal $\sim e^{\propto N}$

Radar REMPI is a measure of number of the atomic oxygen atoms (verified in flames), the backward lasing scaling is >> quadratic.
The exponential behavior suggests stimulated emission

Air lasing from oxygen and nitrogen

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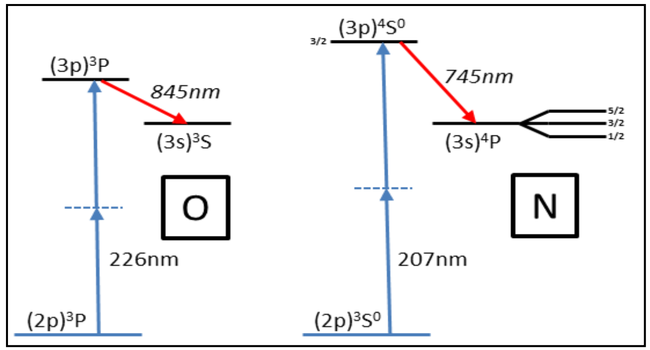
Conversion efficiency η ~ 10^{-4}
Photon conversion efficiency ~ 0.01-0.1%

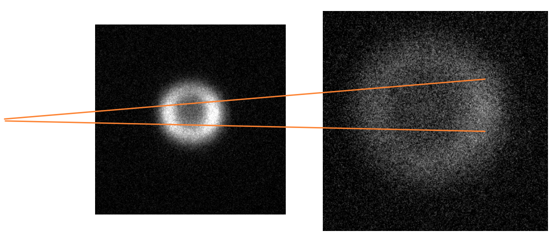
O Pump: 226nm
O Emission: 845nm
Dissociation Energy = 5.15 eV
(pre dissociation enhances pulse energy)



N Pump: 207nm
N Emission: 745nm
Dissociation Energy = 9.79 eV
(pre dissociation required)

Beam Divergence: Gaussian Propagation

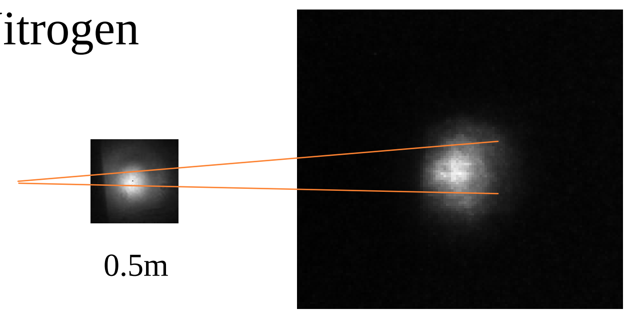
Oxygen



$$\theta_{oxygen} / \theta_{UV} = 3.3$$

$$\lambda_{oxygen} / \lambda_{UV} = 3.7$$

Nitrogen



0.5m

A

6.5m

$$\theta_{nitrogen} = 2mrad$$

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CLEO (2011, 2012, 2013, 2015)

Air lasing – pulsewidth measurement

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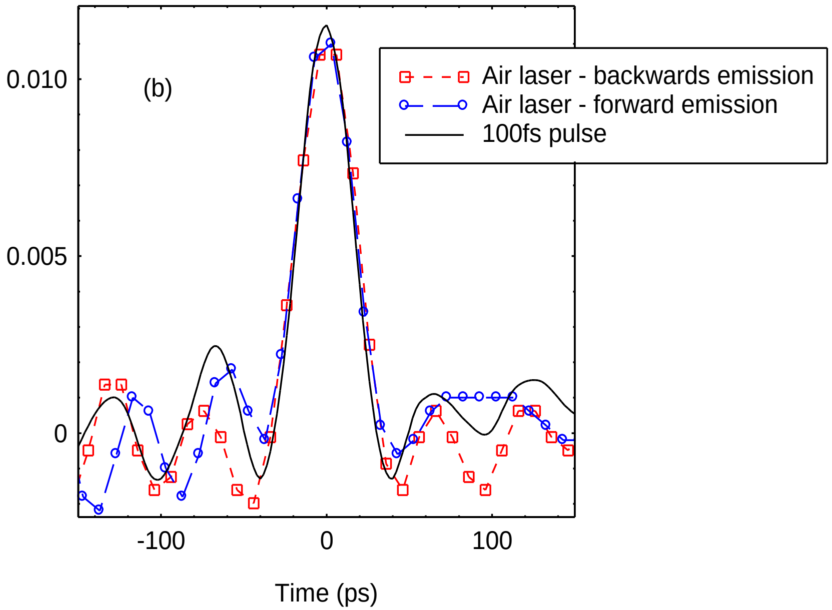
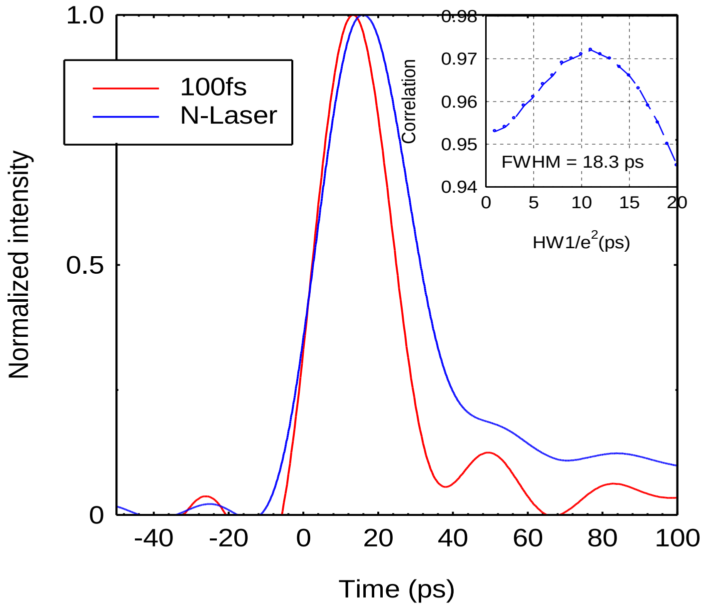
Oxygen



Pulse-width < 30ps
(Spectral width infers pulse >10ps)
Atomic oxygen lifetime: 34ns!

Nitrogen



Pulse-width ~ 20ps
Atomic nitrogen lifetime: 43ns!

Lasing from hydrogen in air

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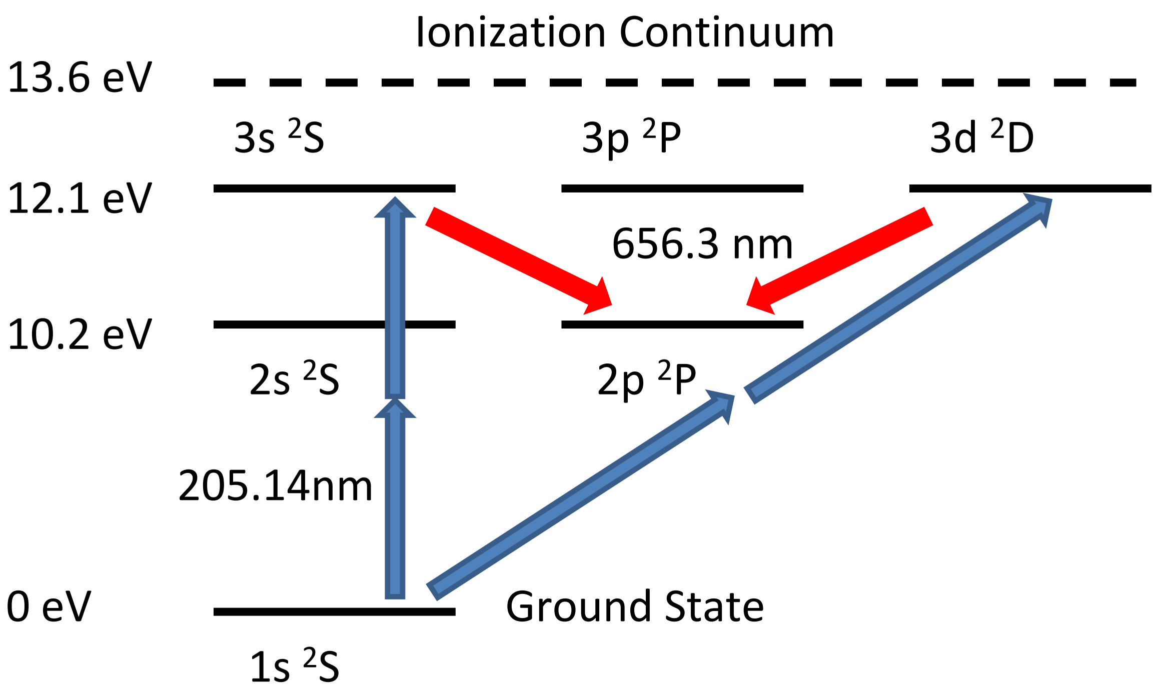
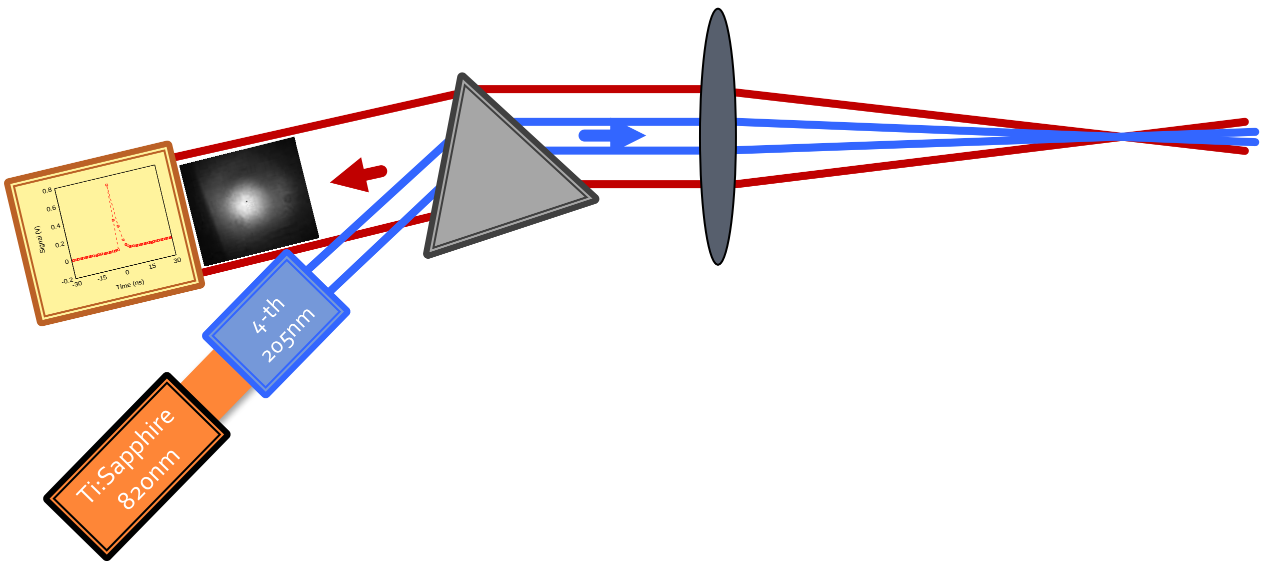
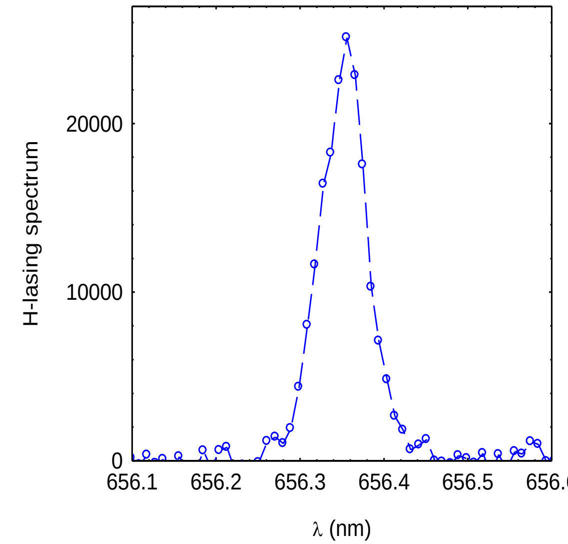
- Two photon excitation at 205.14 nm
- Balmer-alpha emission at 656.281 nm (wavelength in air)



Hydrogen backwards lasing



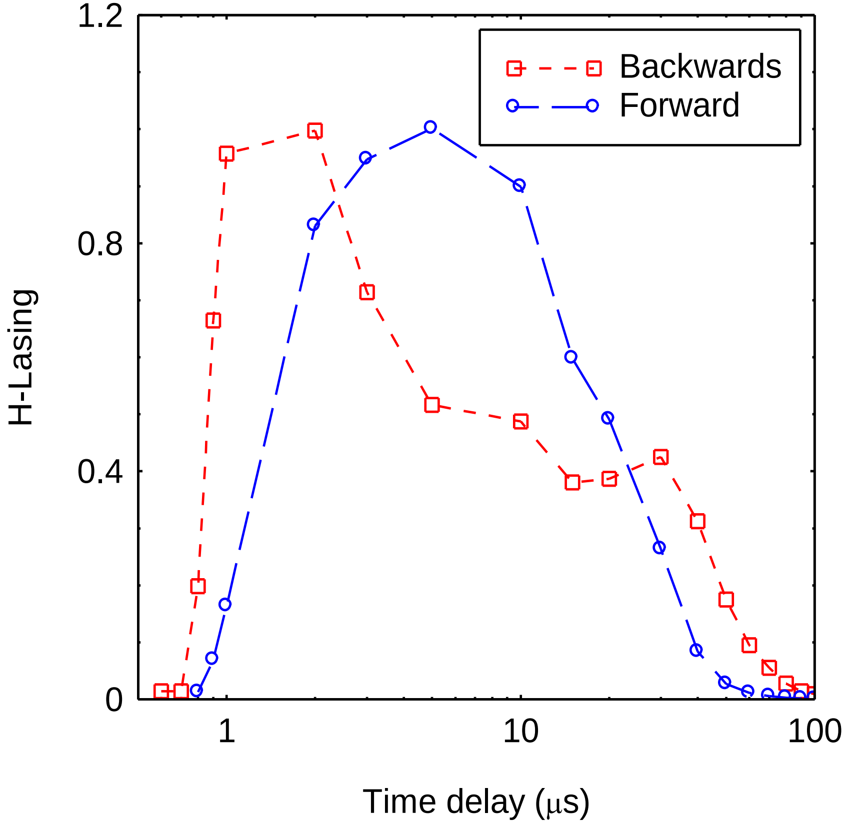


Preliminary dissociation of water vapor required

- Double pulse of the 205 nm laser
- Second laser (Nd:YAG)

- H-atom lasing – 2-photon excitation (similar to O, N)
- Strong backwards visible (red) lasing

Pre-dissociation with ns laser



Nanosecond Nd:Yag laser pulse (100mJ) produces H atoms by dissociating the water molecules.

205nm pulse is delayed – H-atoms can be pumped up to 100 microseconds after Nd:YAG pulse

Optimum delay is 5-10 microseconds

Backwards atmospheric water lasing

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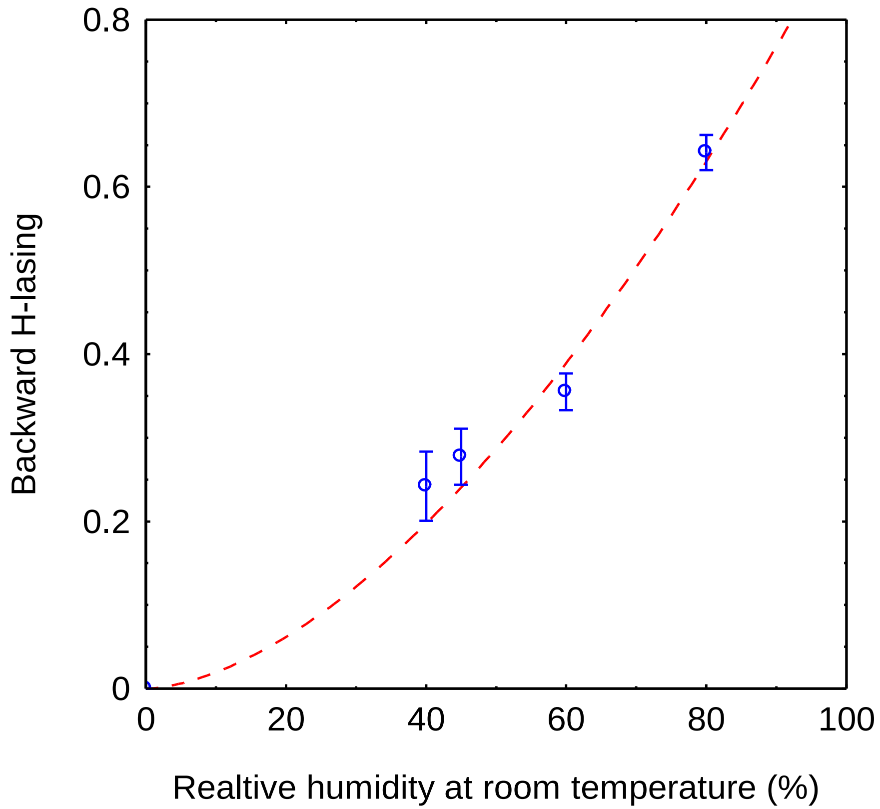
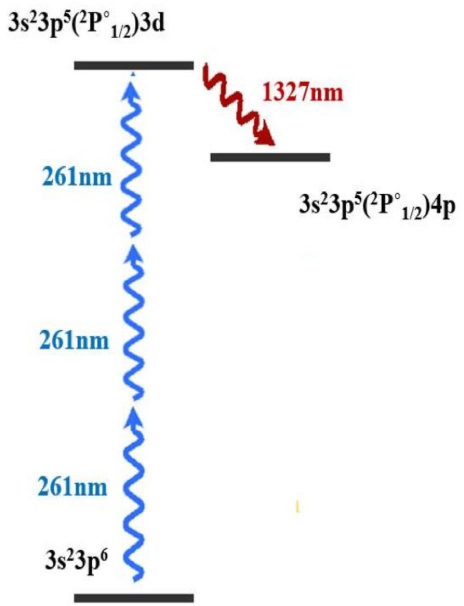
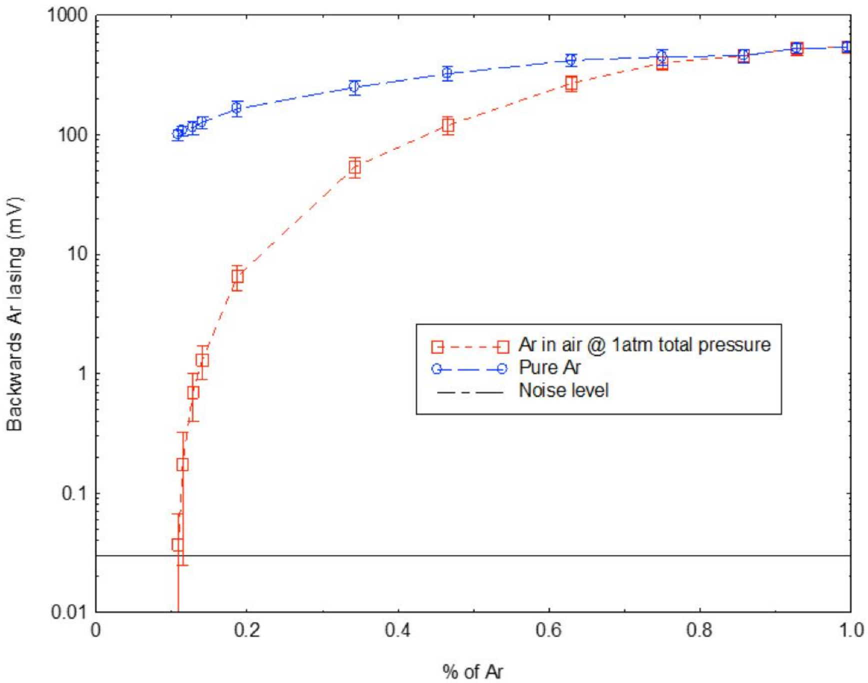


- IR laser pulse dissociates the water molecules in air to create hydrogen atoms:
  - 100mJ @ 1064nm from a ns Nd:YAG laser, or
  - 1mJ @ 800nm from a fs Ti:Sapphire laser
- UV pulse excites H via two-photon absorption
- H-lasing (backwards and forward) increases nonlinearly with the water molecule concentration
- 45% humidity = 1% water in air

Argon Backward Lasing

- Three photon pumping required
- No dissociation necessary
- 0.8% in the atmosphere
- So far success down to 10% argon in air
- Pumping with 50 fsec laser demonstrated

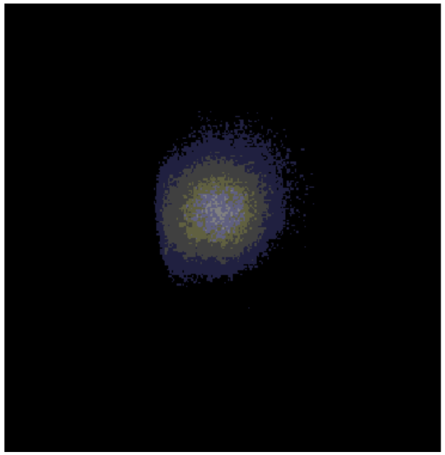


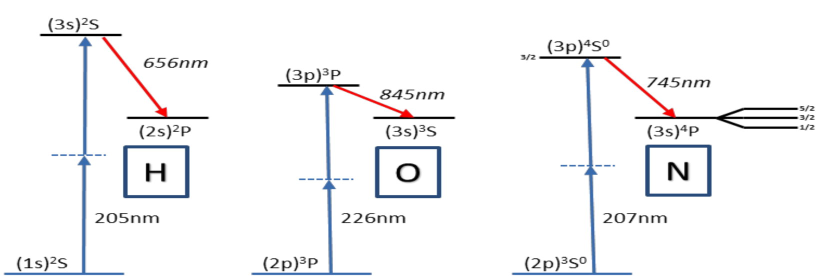


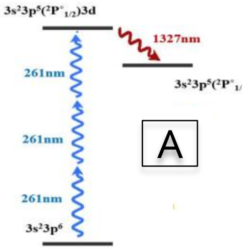
A. Dogariu and R. B. Miles, “Three-photon femtosecond pumped backwards lasing in Argon,” *Opt. Express* **24**, A544-A552 (2016).

Summary: Backwards lasing in air

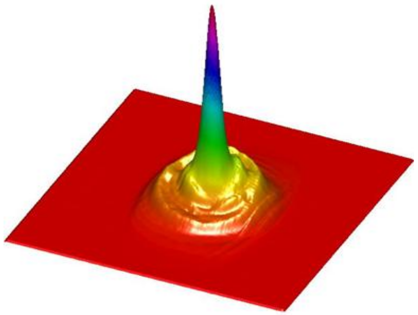
- Atmospheric lasing (major species): Molecular dissociation followed by two-photon excitation of the atomic fragments (N, O)
  - Forward and backward emission with low divergence.
  - Short coherent pulses, bandwidth limited: 10-20ps
  - Transverse spatial coherence
  - Temporal coherence
  - Exponential Gain: high optical gain ($60 cm^{-1}$)
  - Pulse length ≅ gain medium length
  - Photon conversion efficiency >0.1%
- Backwards water lasing in air (minor species)
  - Atomic H lasing (same as N,O)
  - Dissociation of water molecules (or any H-based molecule - methane)
  - Strong red backwards emission (656nm)
- Backwards lasing in Argon – no dissociation required.
  - 3-photon excitation (ps, fs) @261nm, emission@1327nm.
  - Lasing down to 10% Ar in air







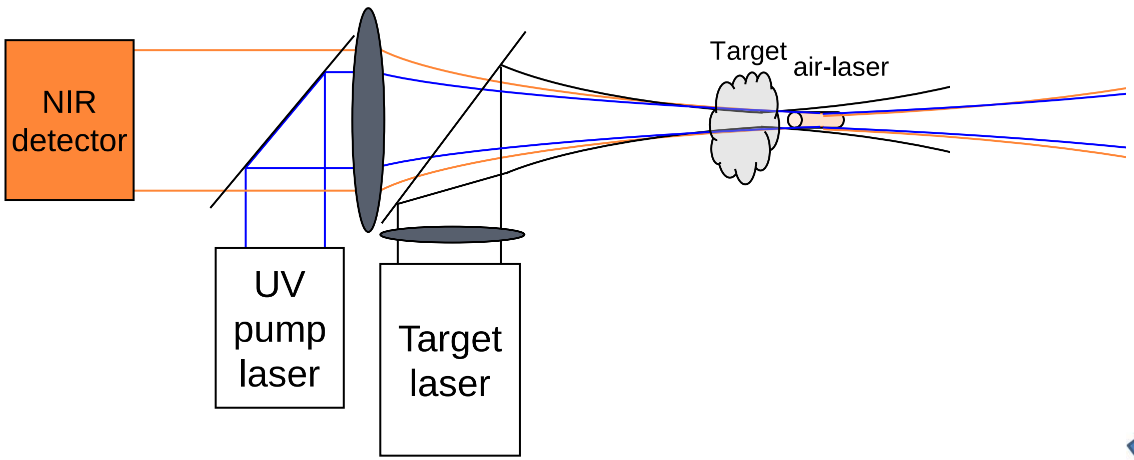
10% Ar in Air

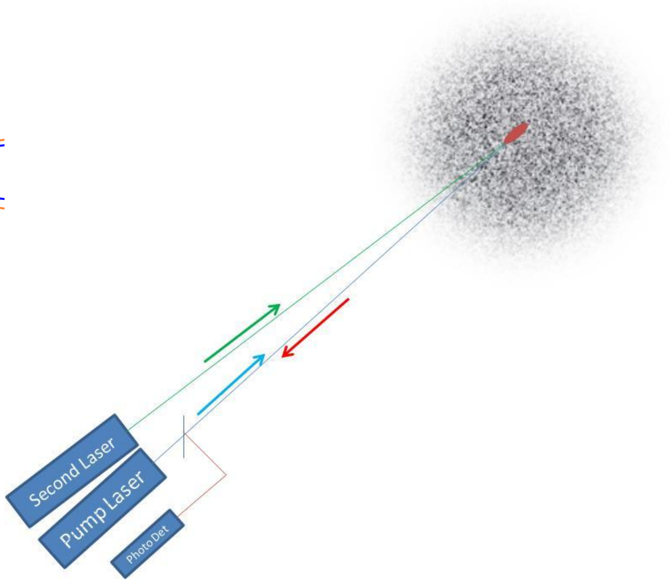


Thank you!

Questions?

Remote trace detection using air lasing





- The UV pump laser creates the backwards emitting air laser
- The "Second Laser" interacts resonantly with the target cloud, modulating the pump and air lasers:
  - **Differential index change**: small changes in the pump beam translate in big changes for the air laser (highly nonlinear)
  - **Raman gain**: Target laser tuned to provide stimulated Raman scattering (SRS) for the air laser

Pulse to pulse reference: Dual air laser

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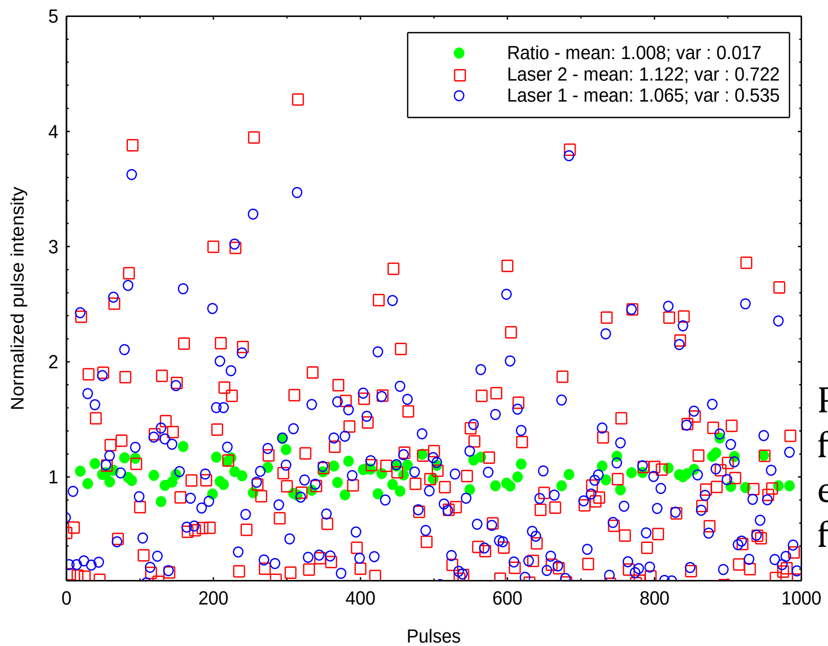
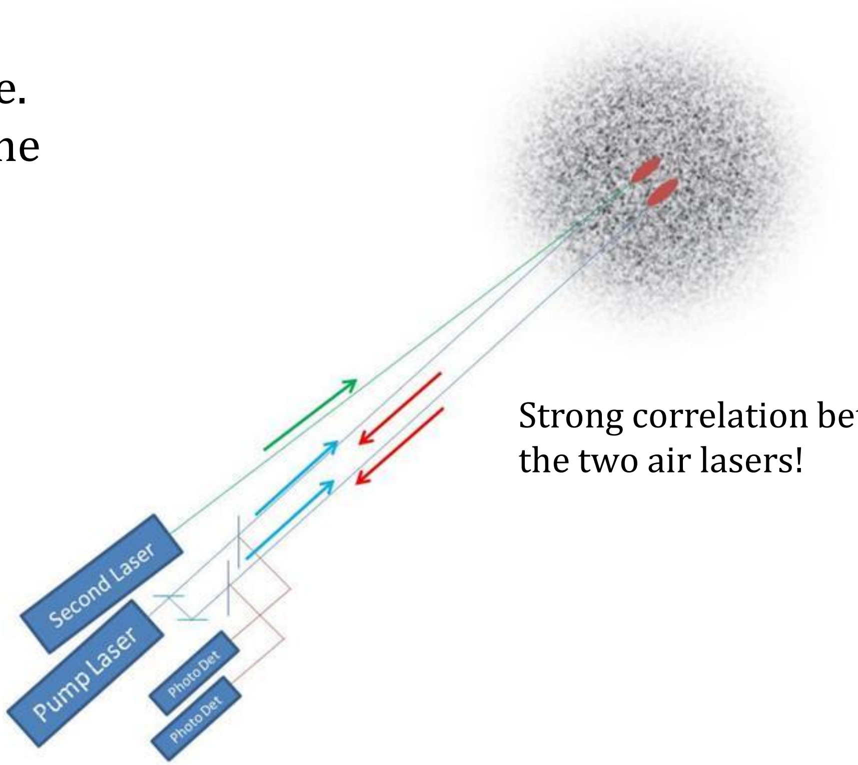
A second backward propagating air laser created by the same pump acts as a reference.

- Minimizes pulse to pulse fluctuations of the pump laser.
- Minimizes distortion due to propagation through the air.





Strong correlation between the two air lasers!

Pulse variance reduced from 50% and 70% for each laser, to less than 2% for their ratio.

Princeton University

Chemring
Sensors & Electronic Systems