Single-Electron Ionization of Diatomic Molecules by Femtosecond Laser Pulses

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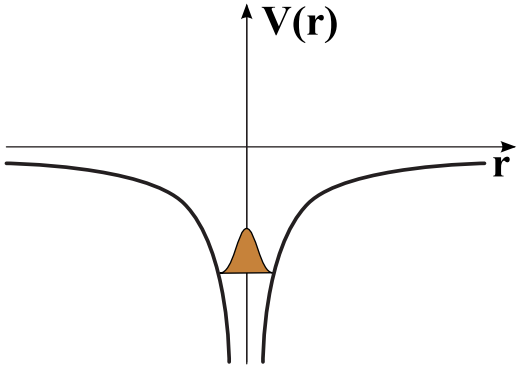
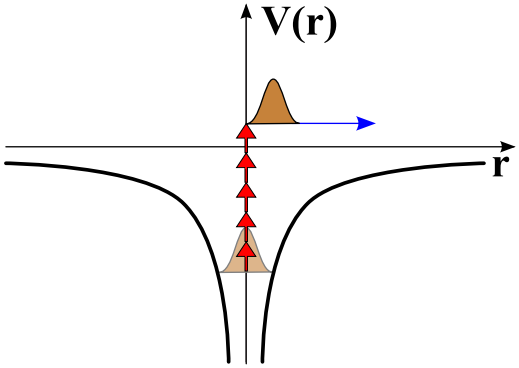
Outline

1. Introduction & Motivation
2. Experimental Details
3. Intensity Calibration
4. Analysis & Data Preparation
5. Results
6. Conclusions & Outlook

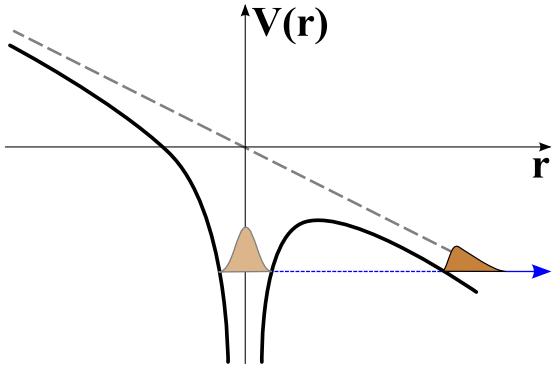
Introduction - current ionization models



Static field



Multi-photon
$(\gamma \gg 1)$



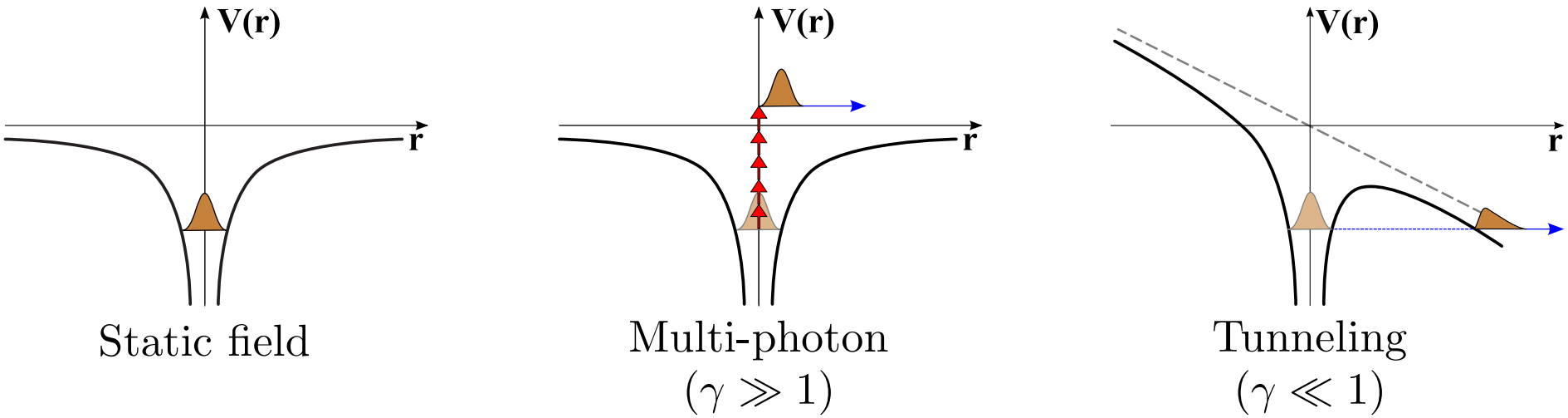
Tunneling
$(\gamma \ll 1)$

Ionization regimes can be classified according to the *Keldysh parameter* γ:

$$\gamma = \sqrt{\frac{I_p}{2U_p}}, \qquad \text{where} \quad U_p = \frac{I_0}{4\omega^2}$$

U_p – Ponderomotive energy
I_p – Ionization potential
I_0 – Peak intensity
ω – Frequency of laser field

Introduction - current ionization models



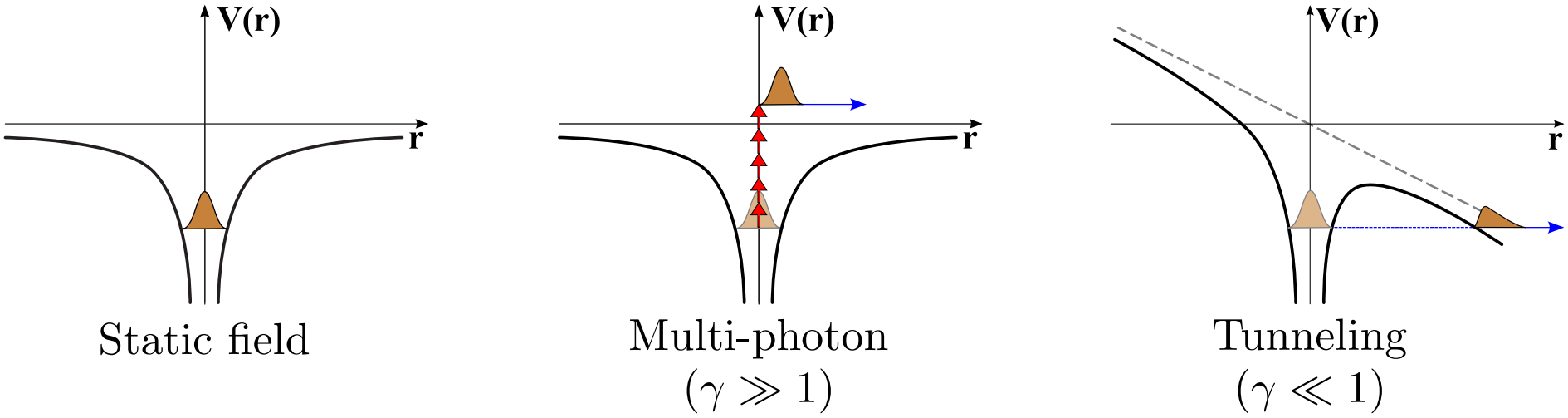
In this talk, we will mention two common **atomic** ionization theories:

1) The Perelomov-Popov-Terent'ev (**PPT**) model [1]
2) The Ammosov-Delone-Krainov (**ADK**) model [2]

[1] A. Perelomov, V. Popov, and M. Terent'ev, Sov. Phys. JETP **23**,924 (1966).

[2] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).

Introduction - current ionization models



PPT is based on the tunneling ionization theory, but is modified to accommodate time-dependent fields ($\mathscr{E} = \mathscr{E}_0 \cos \omega t$)

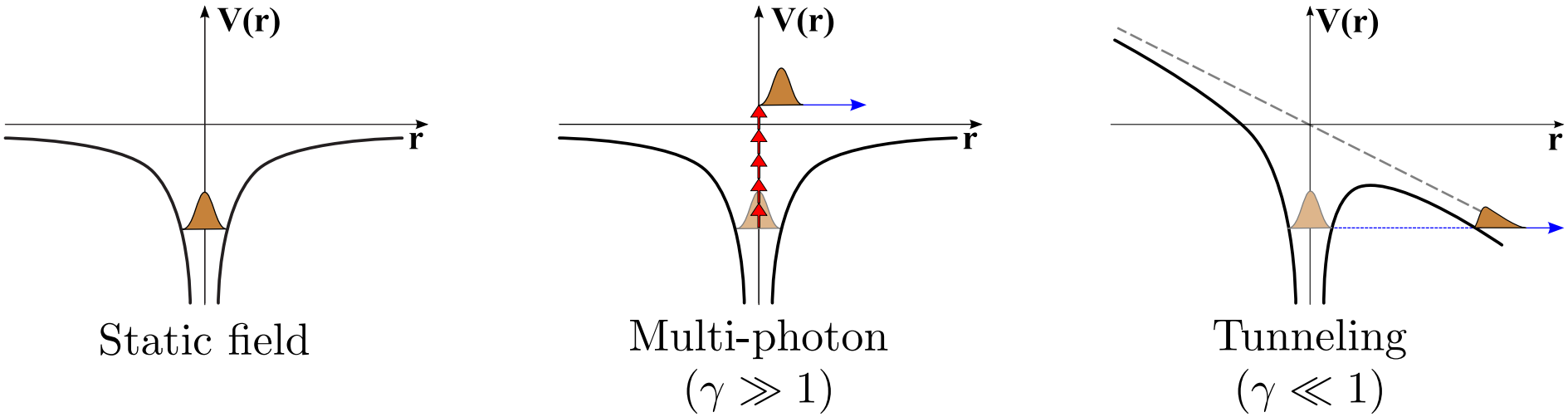
PPT is accurate in both **multi-photon** and **tunneling** ionization regimes [7-9]

[7] F. Yan-Zhuo, Z. Song-Feng, and Z. Xia-Xin, Chin. Phys. B **21** 113101 (2012)

[8] I. Barth and O. Smirnova, Phys. Rev. A **84**, 063415 (2011)

[9] C. Cornaggia and Ph. Hering, Phys. Rev. A **62** 023403 (2000)

Introduction - current ionization models



ADK is a *simplified* version of PPT, which assumes that the only factor which influences the ionization rate is the ionization potential of the **atom**

- This approximation assumes that the intermediate states are unimportant, and the critical parameter determining the ionization rate is the ionization potential

ADK is only accurate in the **tunneling** regime [7-9]

[7] F. Yan-Zhuo, Z. Song-Feng, and Z. Xia-Xin, Chin. Phys. B **21** 113101 (2012)

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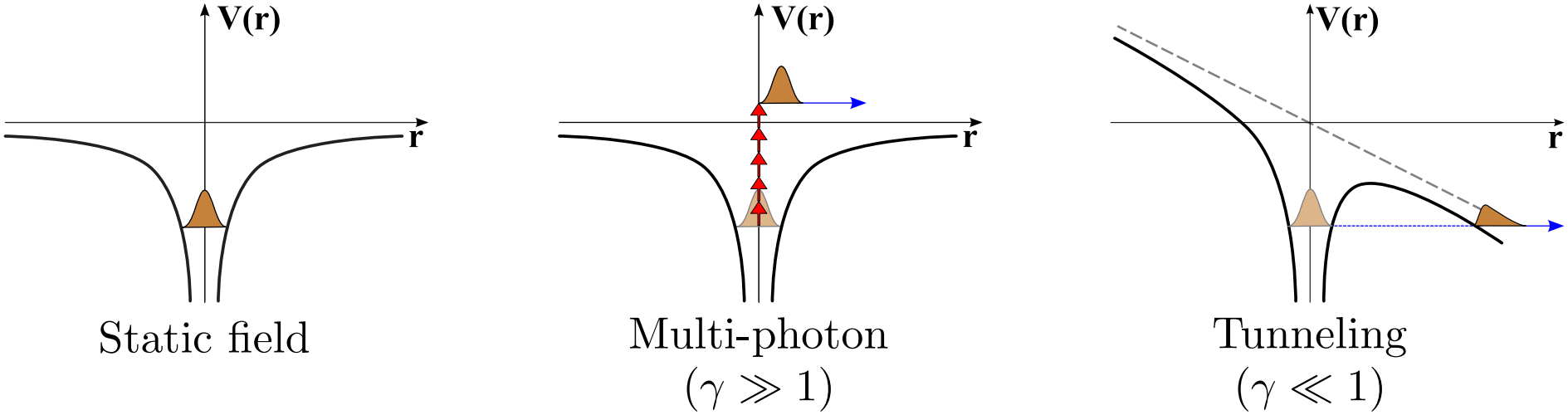

Recently, the ADK **atomic** ionization model has been extended to work for **molecules**

- Called the **molecular tunneling theory** [3] (aka MO-ADK), and is a modification of the ADK theory to account for the non-spherical symmetry of **molecules**

Like ADK, MO-ADK is only accurate in the tunneling regime, and does not agree well with experiment in the low-intensity multi-photon limit ($\gamma \gg 1$)

We use MO-ADK as a <u>benchmark</u> in our studies

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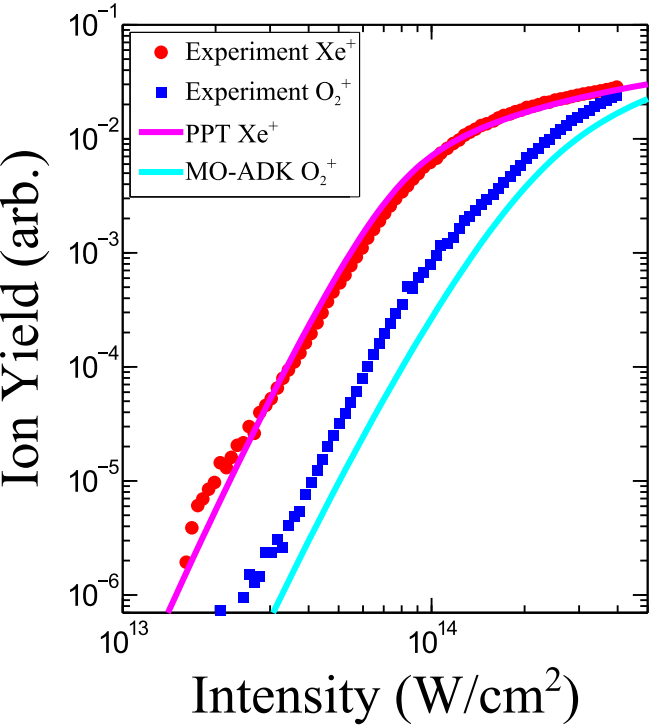
[3] X. Tong, Z. Zhao, and C. Lin, Phys. Rev. A **66** 033402 (2002).

Introduction & Motivation

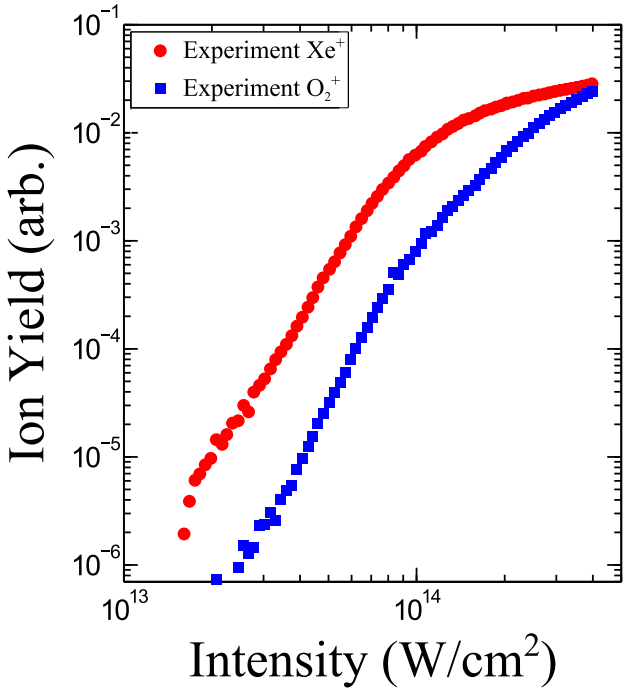
- Current **molecular** ionization theories are lacking in accuracy (*e.g.* MO-ADK)
- Existing **atomic** models work very well (*e.g.* PPT) and are in good agreement

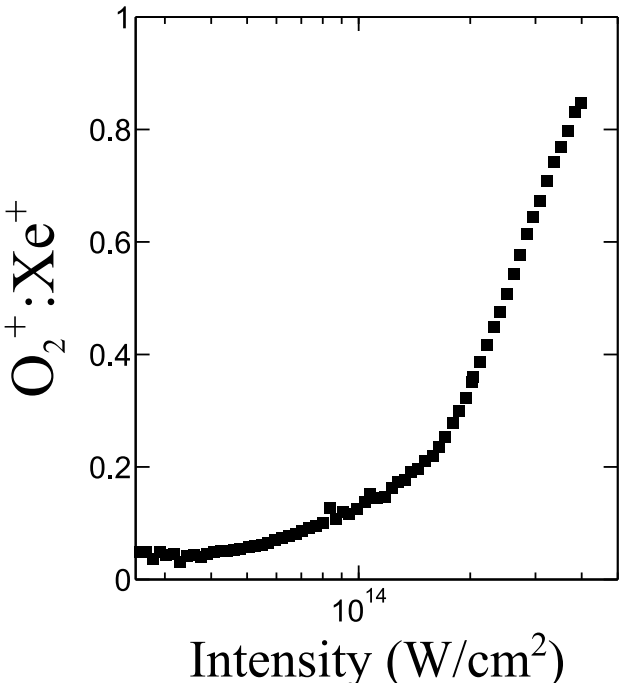


Our goal is to provide a **comprehensive set of data** which can help theorists **improve molecular** ionization models

This will allow theorists to **build upon existing theories** for **atomic** ionization to develop more accurate models for **molecular** ionization

Our method: **simultaneously** measure the singly-charged ion yield for a **molecule** paired with an **atom** which has a similar ionization potential and taking the **ratio** of the yields $\mathbf{M}^+/\mathbf{A}^+$





Taking the **ratio** of these **simultaneously** measured yields has the benefit of factoring out common experimental conditions which are **independent** of the physical quantity we are measuring

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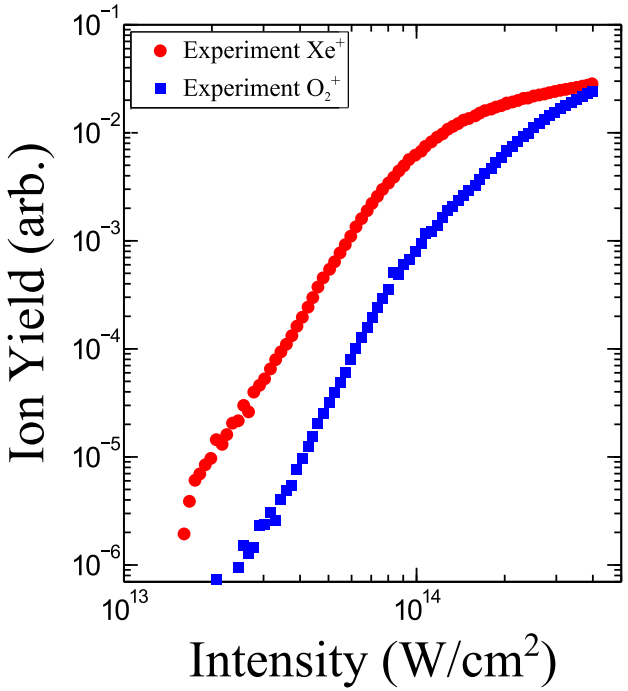
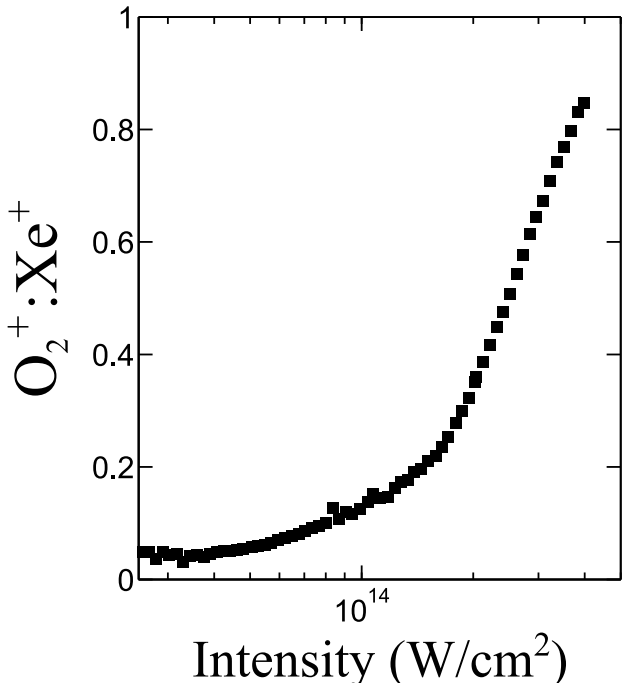
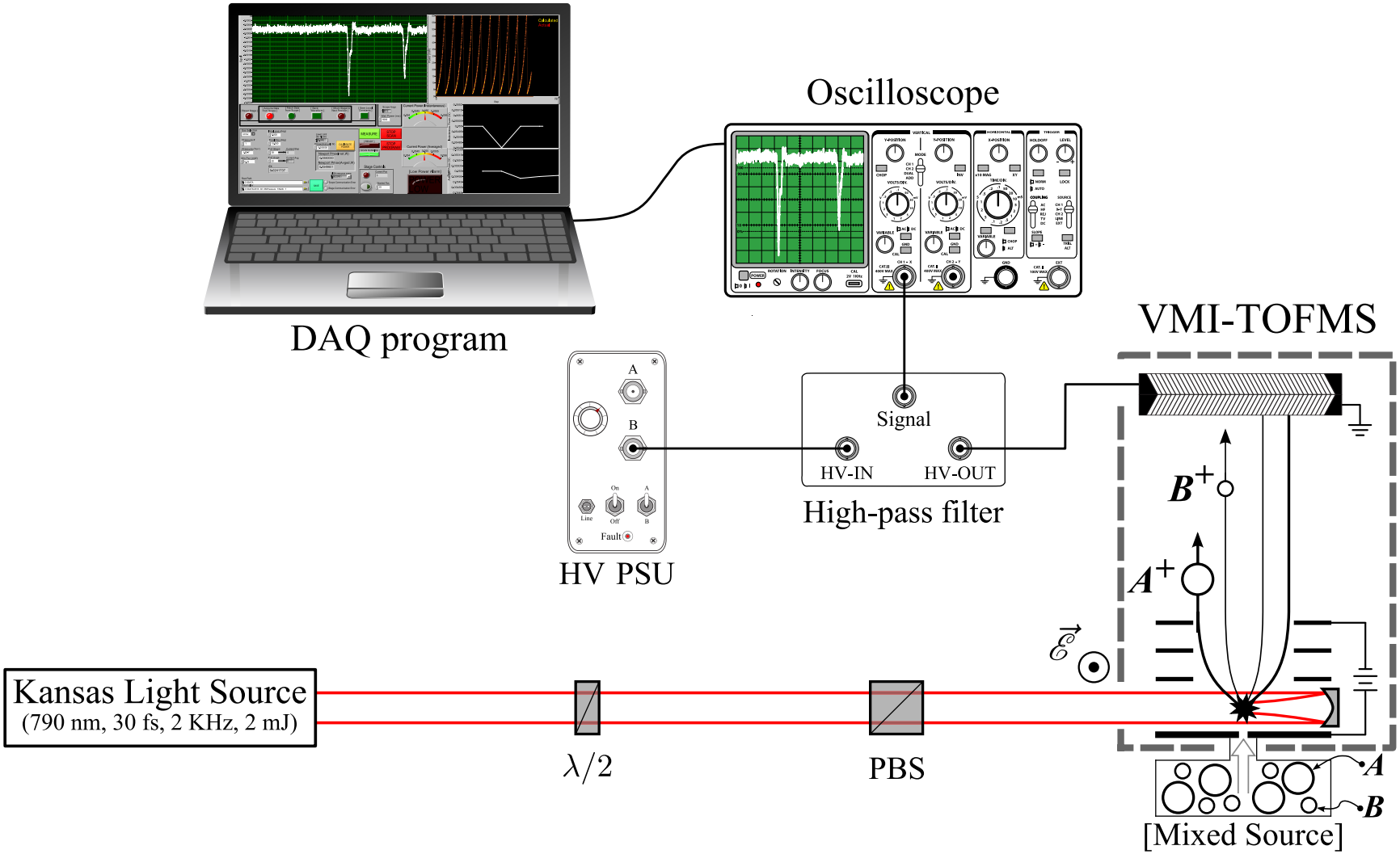




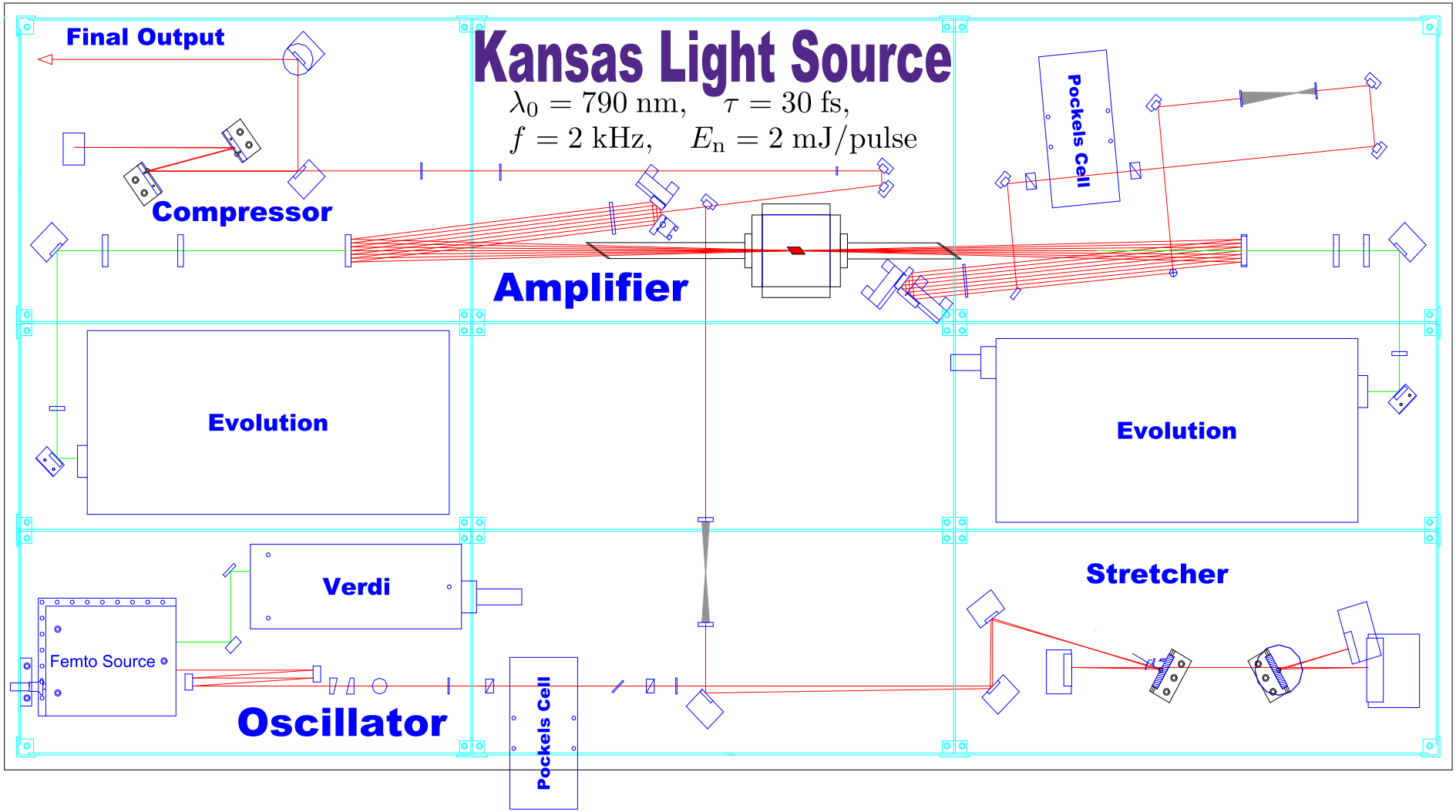
We study three **molecule**/**atom** pairs:

molecule	I_p (eV)	**Atom**	I_p (eV)
N_2	15.58	Ar	15.76
CO	14.01	Kr	14.00
O_2	12.07	Xe	12.13

Overview of our detection scheme



We use the *Kansas Light Source*



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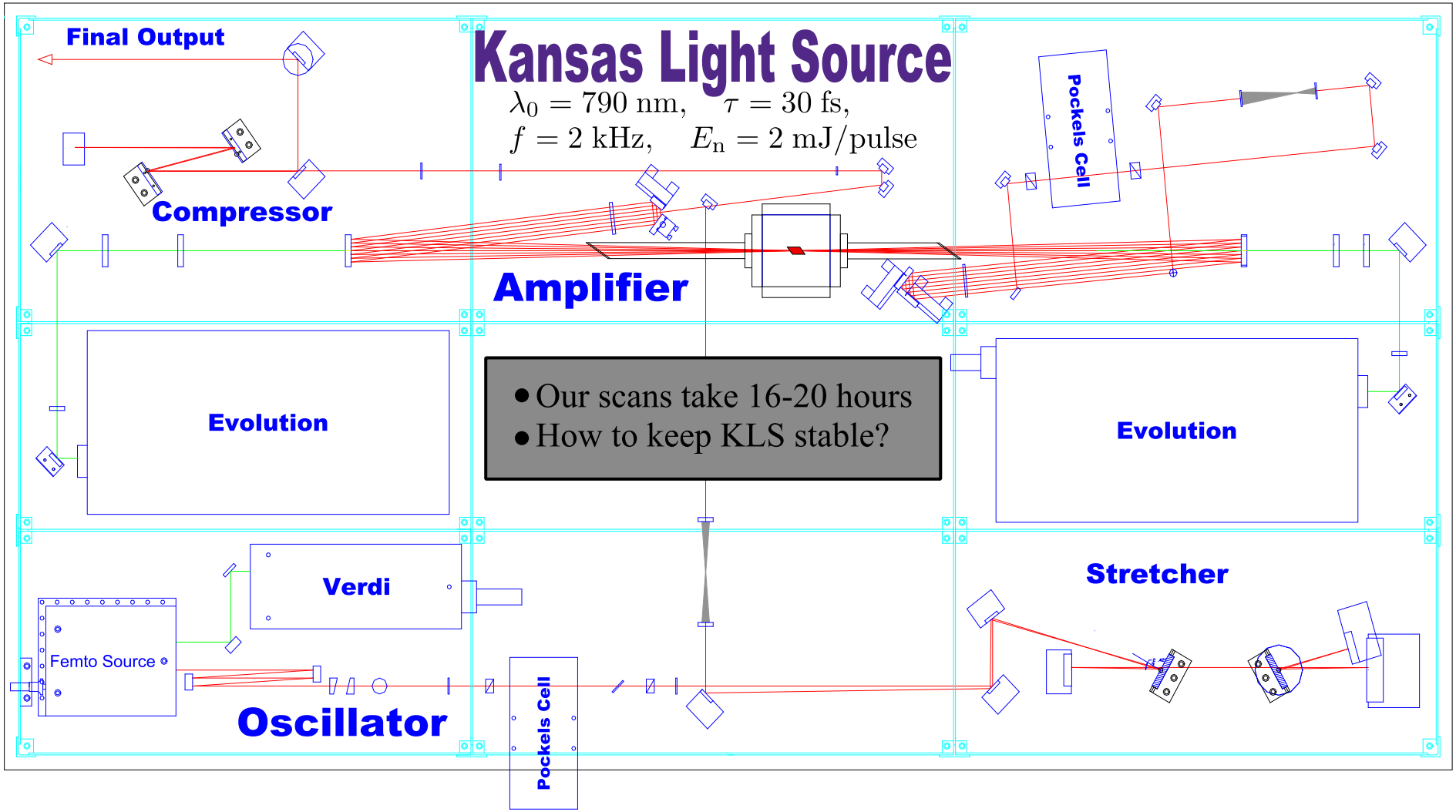
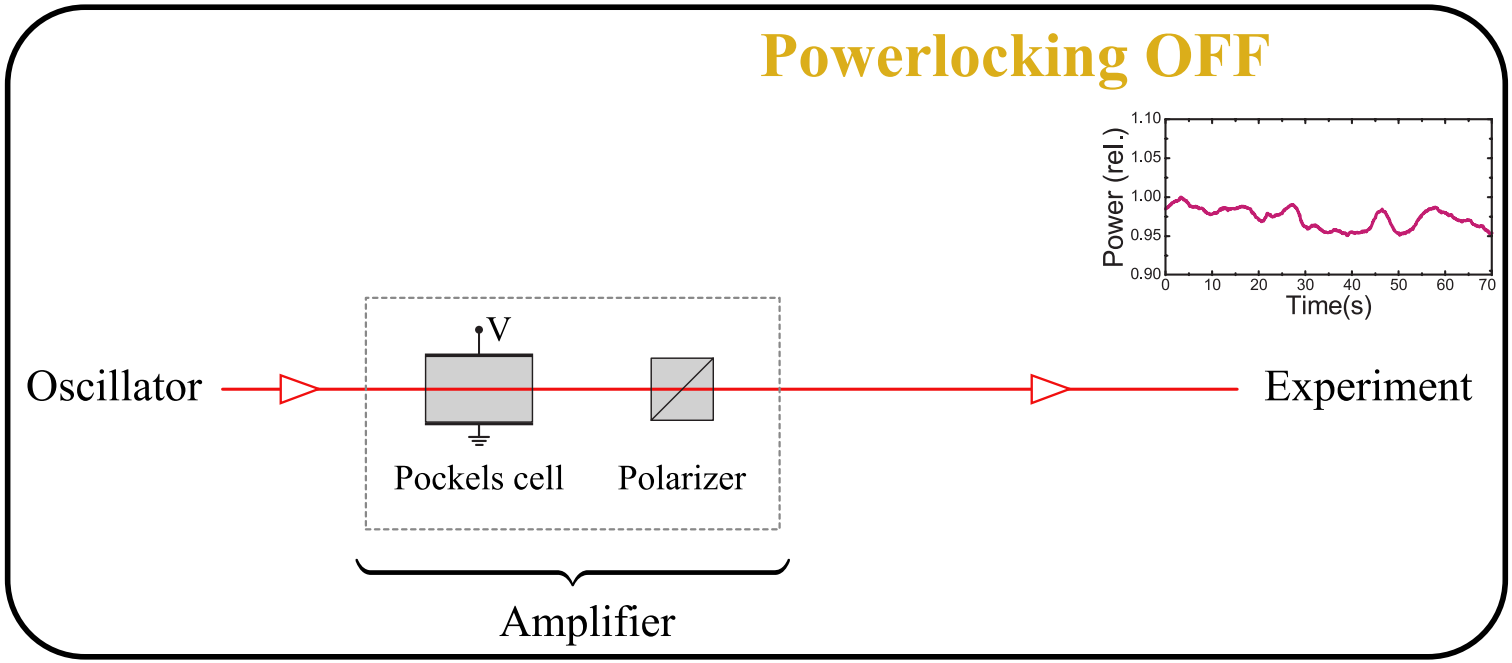


- Our scans take 16-20 hours
- How to keep KLS stable?

Laser stabilization - we used *power-locking*

Power-locking actively stabilizes the output power from KLS by implementing a closed-loop feedback and control system

- Performed using a **P**roportional-**I**ntegral-**D**erivative (**PID**) controller
- The power at the amplifier output is **measured** and compared to the user-specified **setpoint** on the PID



[4] H. Wang *et al.*, Appl. Phys. B: Lasers and Optics **89**, 275 (2007).

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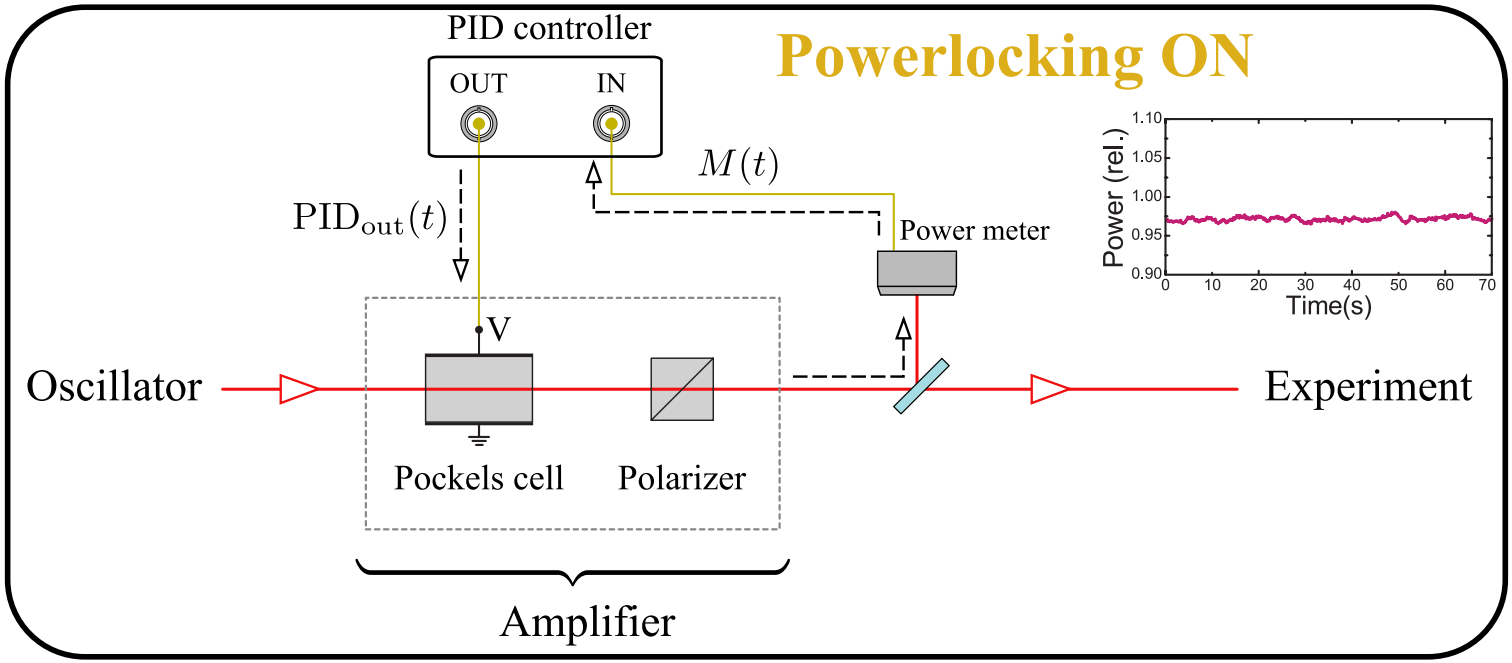
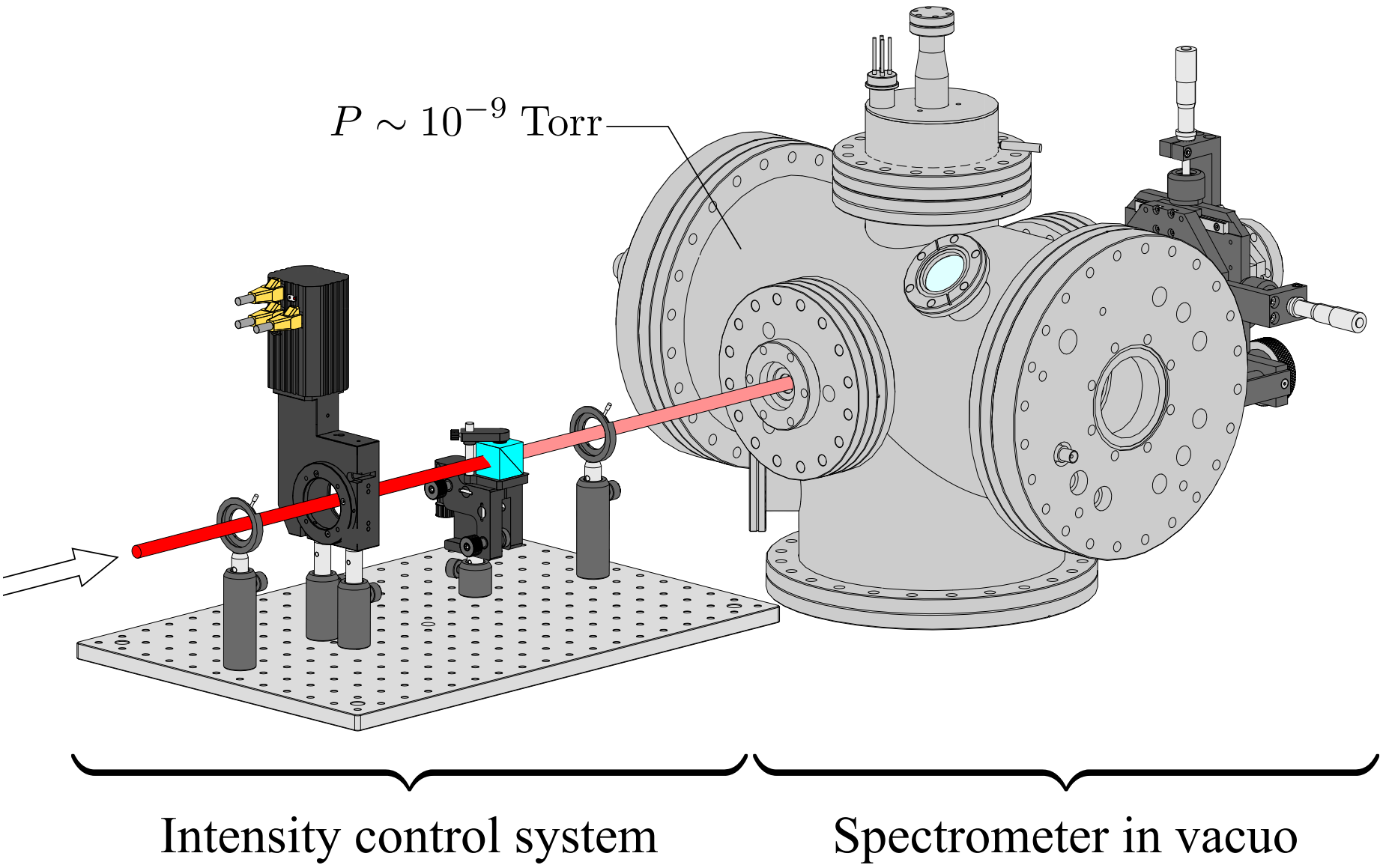
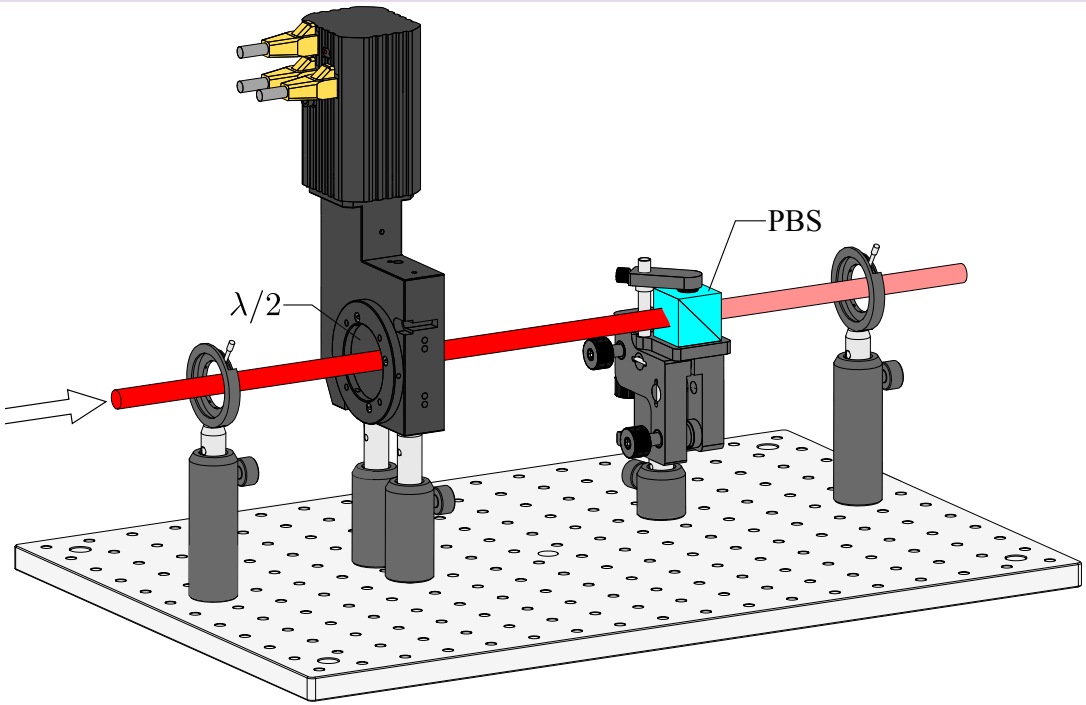


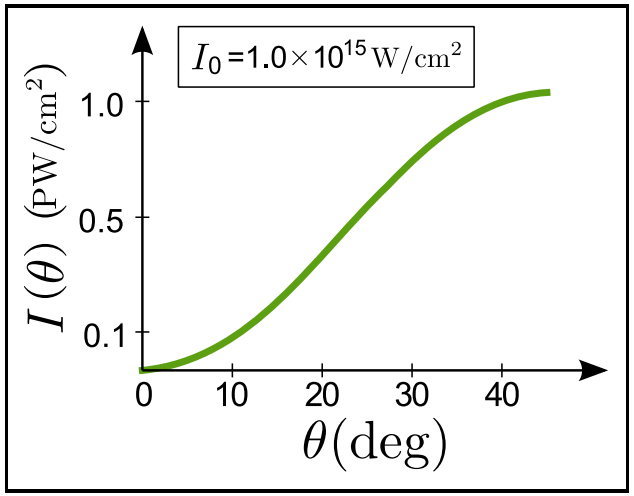
[4] H. Wang *et al.*, Appl. Phys. B: Lasers and Optics **89**, 275 (2007).

Main components of our experiment

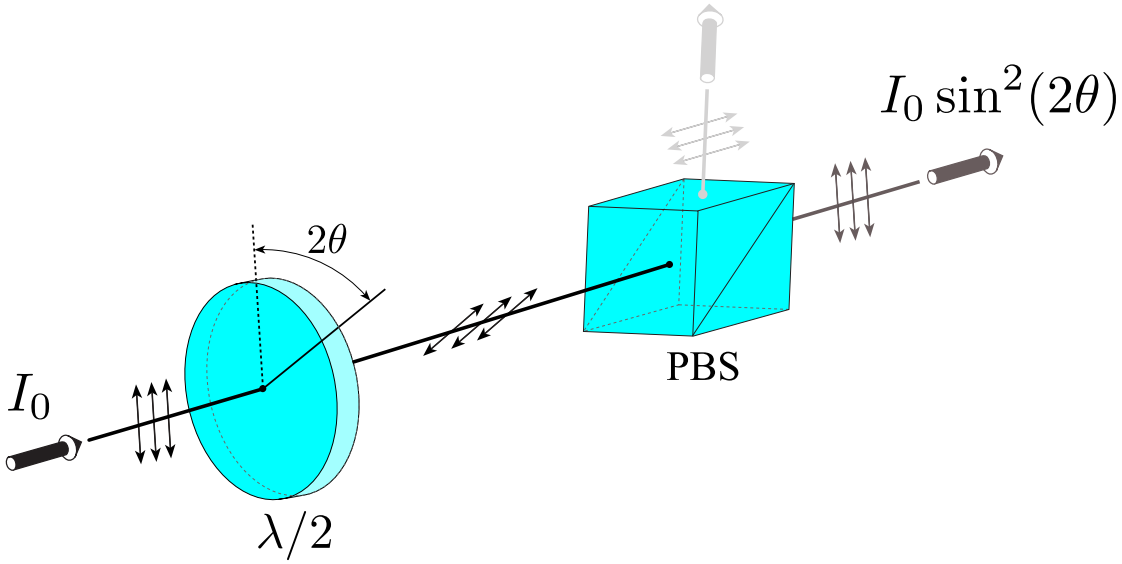


Intensity control system

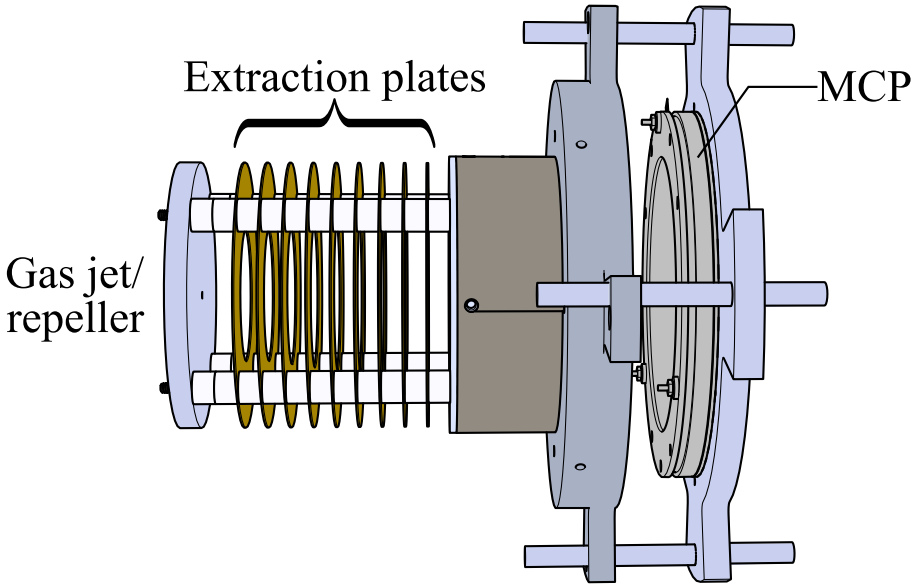


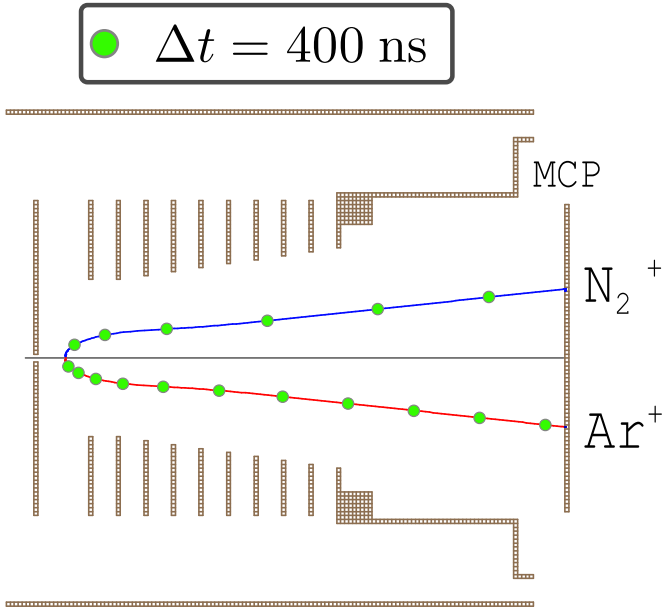


Malus' Law: $I(\theta) = I_0 \sin^2(2\theta)$



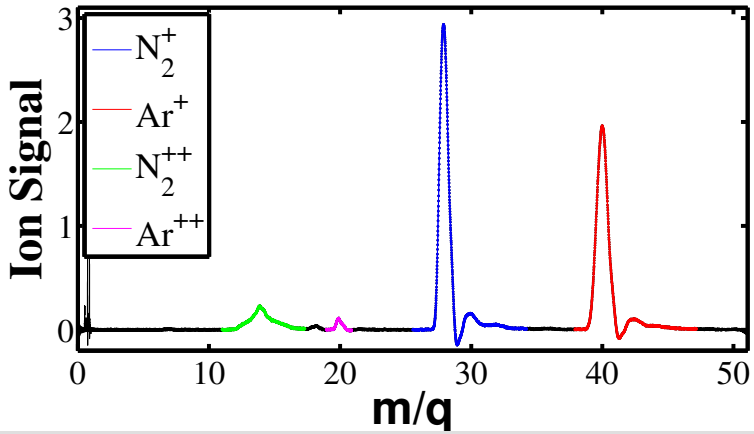
Spectrometer in vacuo



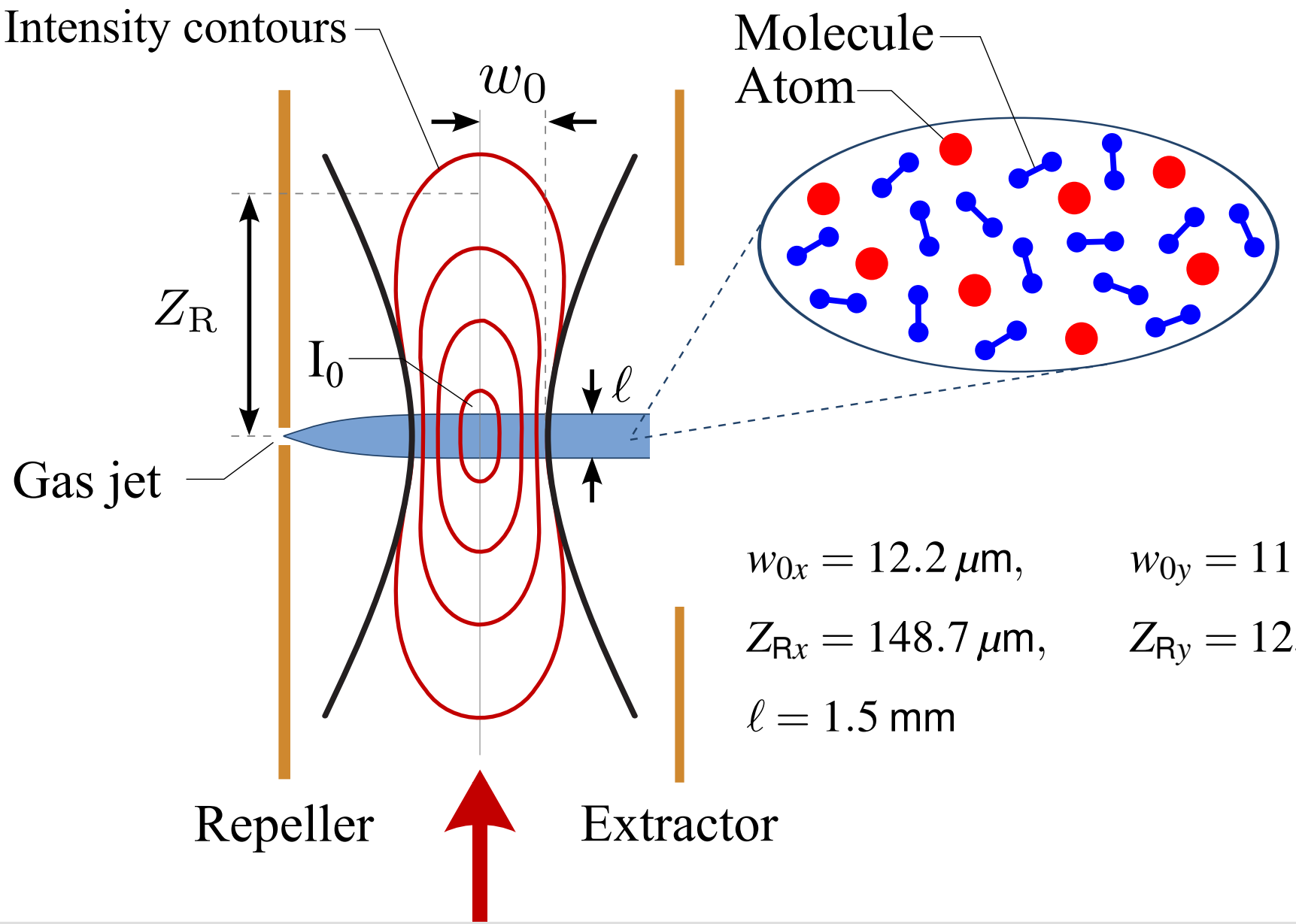


$$t \propto \sqrt{m/q}$$

$m_{N_2} = 28$ amu, $m_{Ar} = 40$ amu



At the interaction region...



$w_{0x} = 12.2\,\mu\text{m}$, $w_{0y} = 11.3\,\mu\text{m}$

$Z_{Rx} = 148.7\,\mu\text{m}$, $Z_{Ry} = 125.8\,\mu\text{m}$

$\ell = 1.5\,\text{mm}$

Gas ratio optimization

In order to take the **ratio** of the **molecule** to the **atom**, we need to have signal from both species across the entire intensity range

- **Atoms** tend to ionize easier than their companion **molecules**
- To ensure uniform detection across both species at **low intensities**, we need to compensate by increasing the density of **molecules** at the interaction region
  - We control the number of particles by changing the **pressure ratio** in the gas mixture

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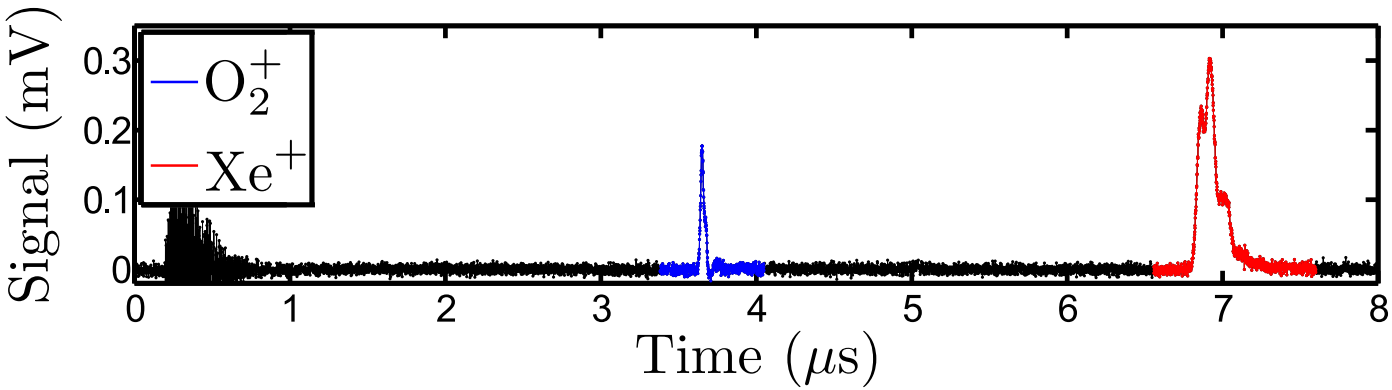
Example: Oxygen and Xenon



$$\frac{P_{O_2}}{P_{Xe}} = 2.47$$

Determining the intensity

$$I(r,z,t,\theta) = \underbrace{\overbrace{\frac{4E_{\mathrm{n}}^{(\mathrm{max})}}{\pi w_0^2 \tau}\sqrt{\frac{\ln 2}{\pi}}\sin^2(2\theta)}^{\textit{Malus' Law}}}_{I_0} \times \exp\left[-\frac{r^2}{w_0^2[1+(z/Z_{\mathrm{R}})^2]} - \frac{4\ln 2\, t^2}{\tau^2}\right],$$

where:

$E_{\mathrm{n}}^{(\mathrm{max})}$ – Pulse energy measured at $\theta = 45^{\circ}$

τ – Pulse duration (FWHM)

w_0 – Minimum spot size (e^{-2} half-width)

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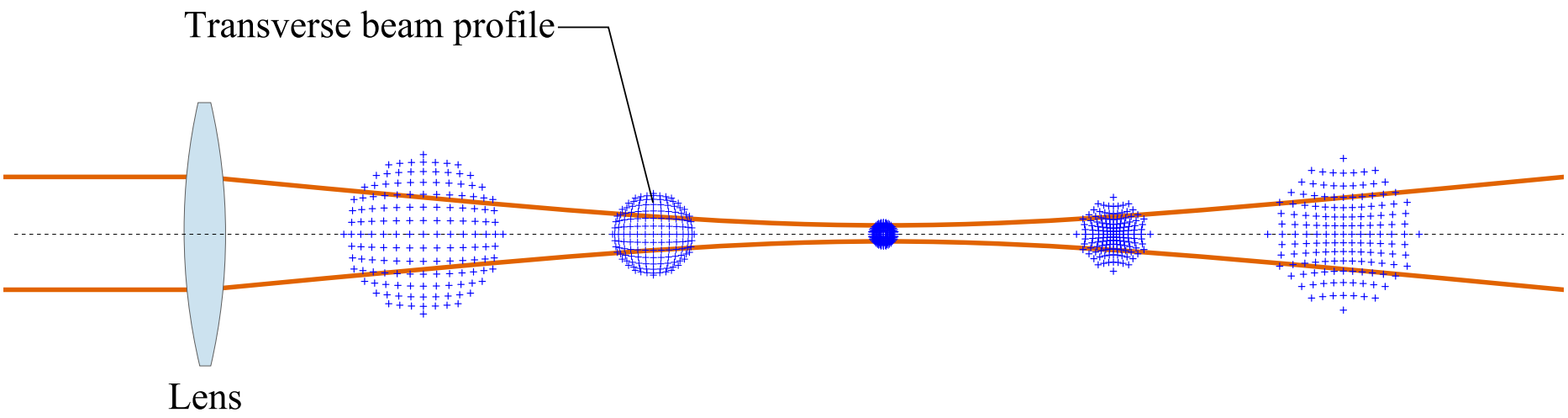
where: $E_{\mathrm{n}}^{(\mathrm{max})}$ – Pulse energy measured at $\theta = 45^{\circ}$

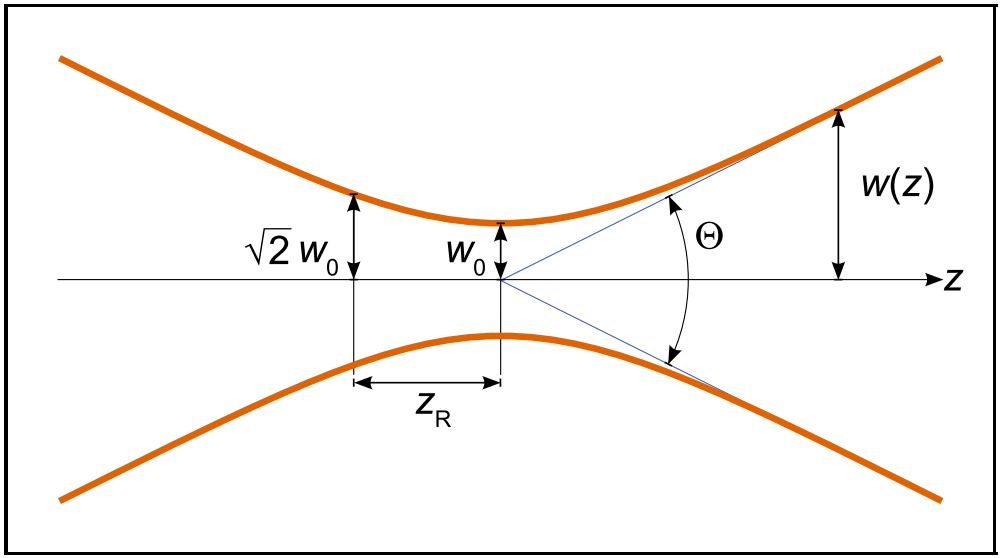
τ – Pulse duration (FWHM)

w_0 – Minimum spot size (e^{-2} half-width)

- The **pulse energy** was obtained by measuring the *average power* with a power meter: $E_{\mathrm{n}} = T \times P_{\mathrm{avg}}$, where T is the repetition period of the laser
- The **pulse duration** measured with a SH-FROG
  - we measured the transform-limited pulse to have $\tau = 30$ fs
- The **minimum spot size** was measured in a separate experiment that we invented for this purpose...

Measuring the minimum spot size - review of Gaussian optics

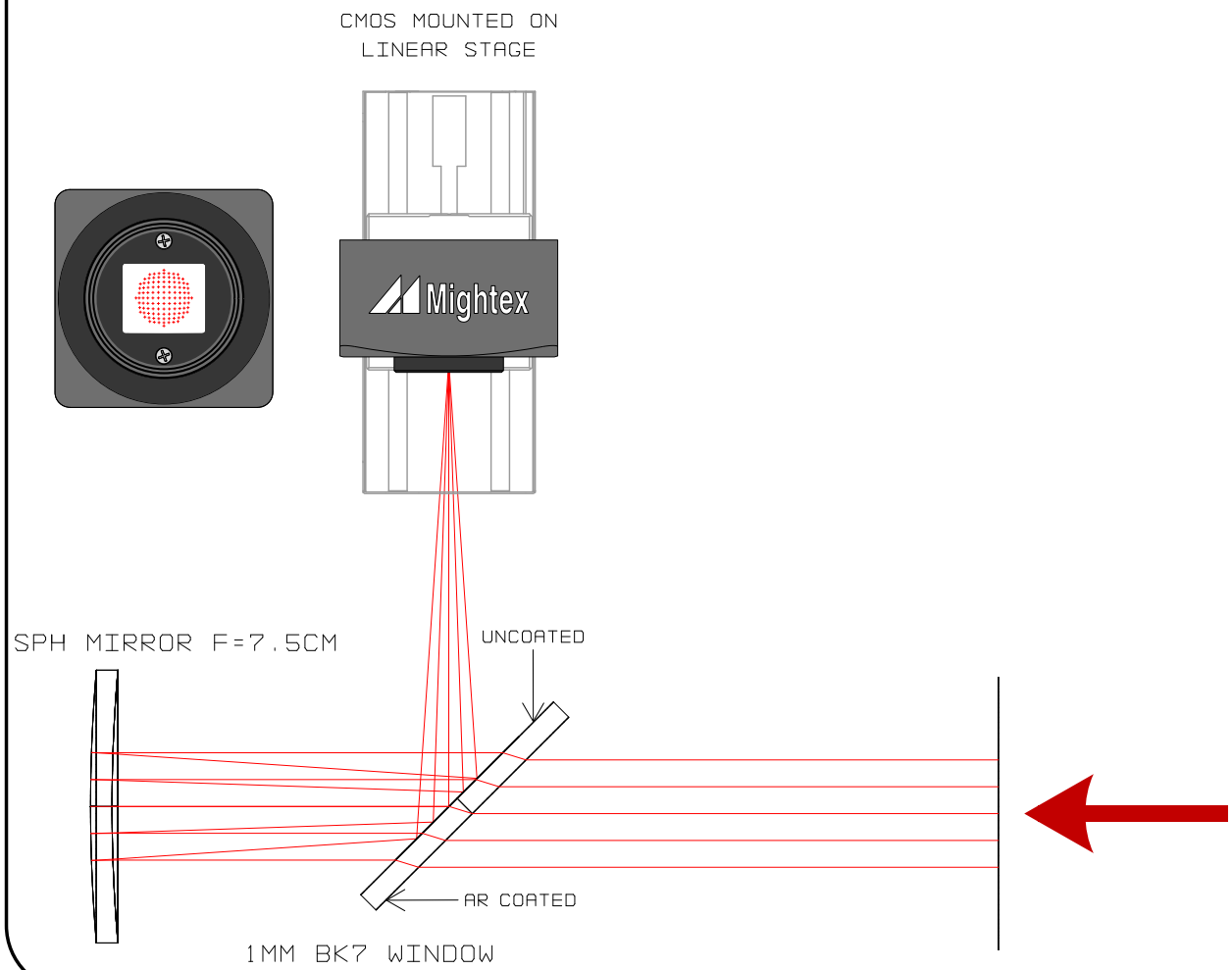




$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{Z_R}\right)^2}$$

$$Z_R = \frac{\pi w_0^2}{\lambda} \quad (\textit{Rayleigh range})$$

Measuring the minimum spot size (w_0)



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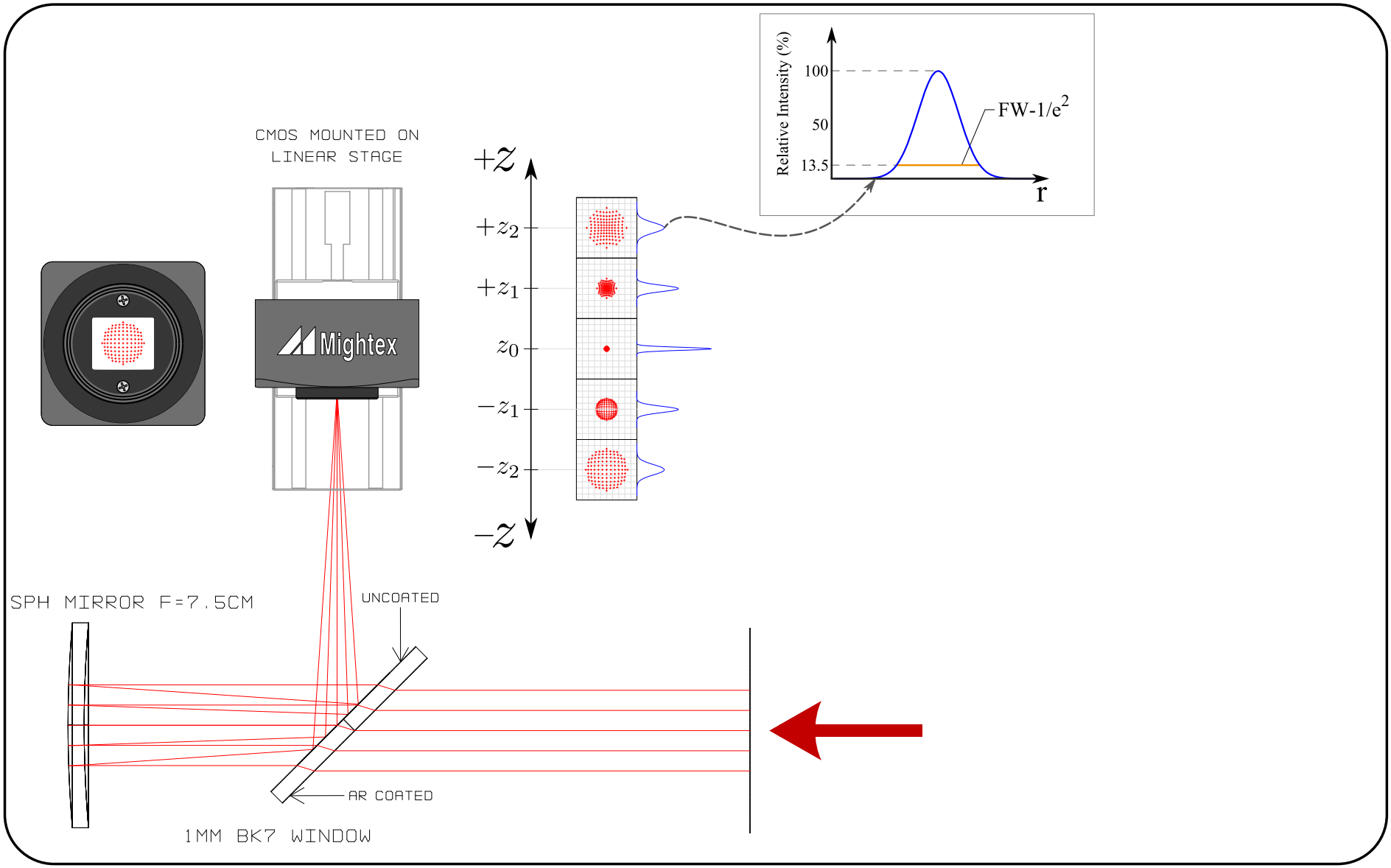

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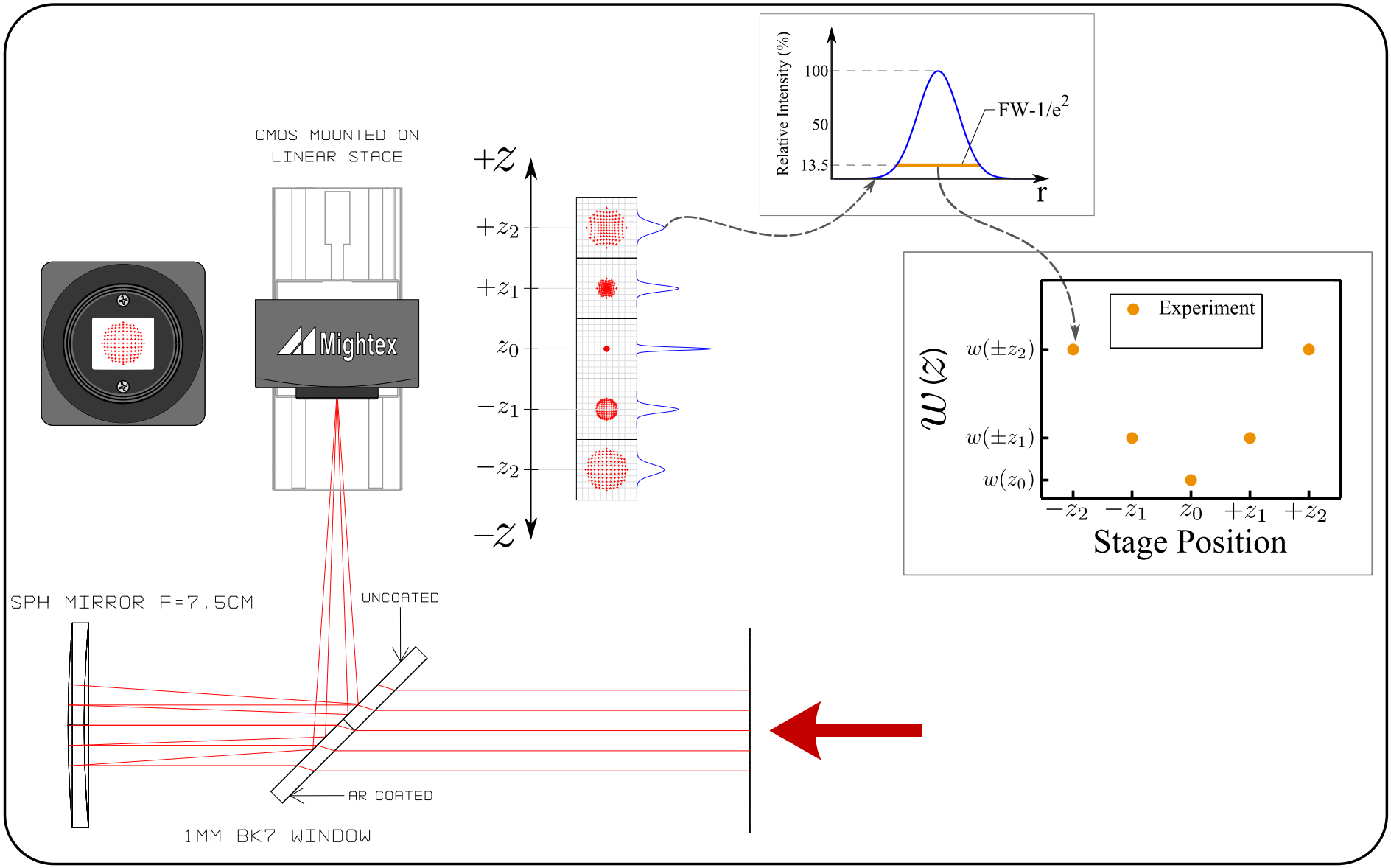

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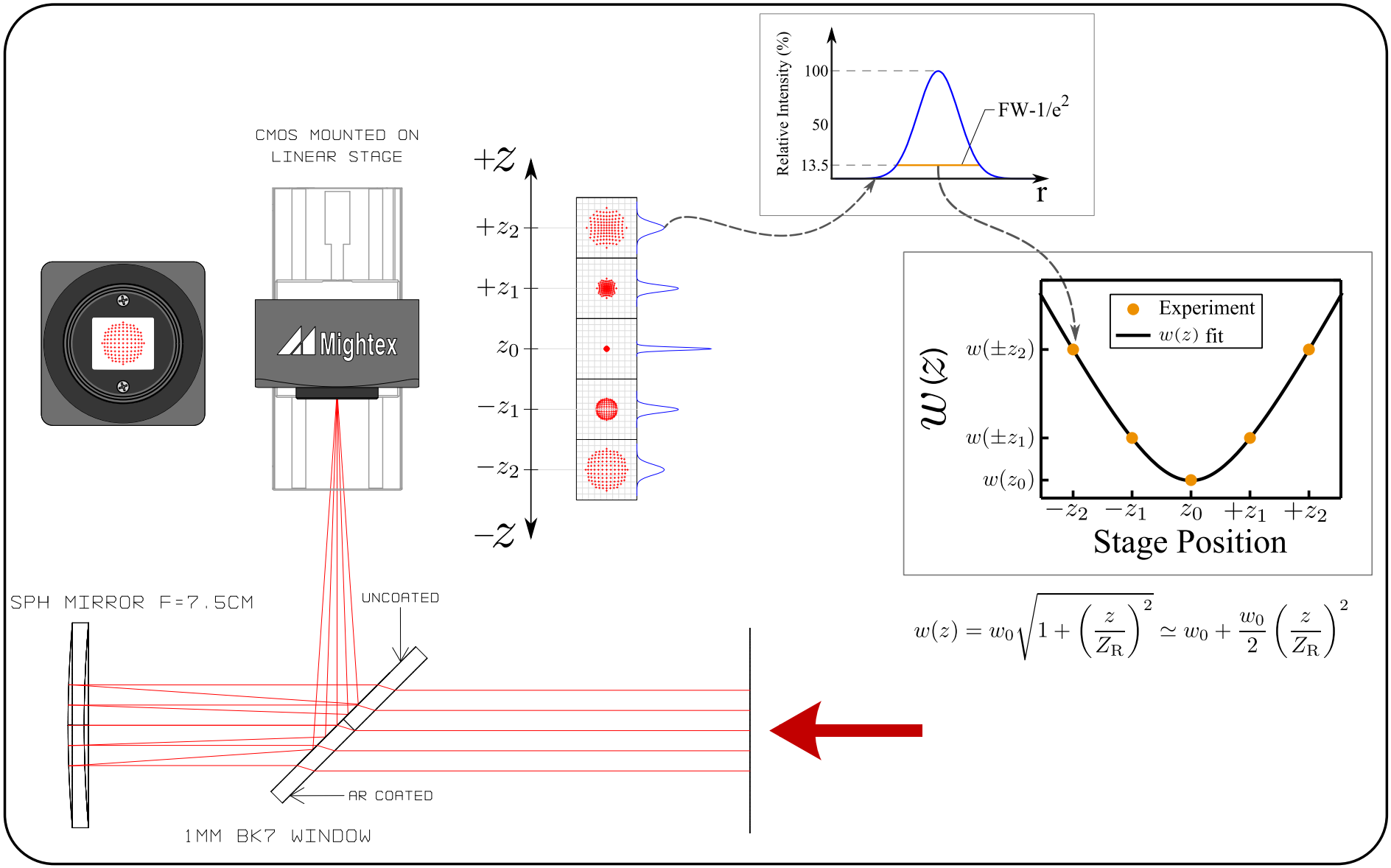
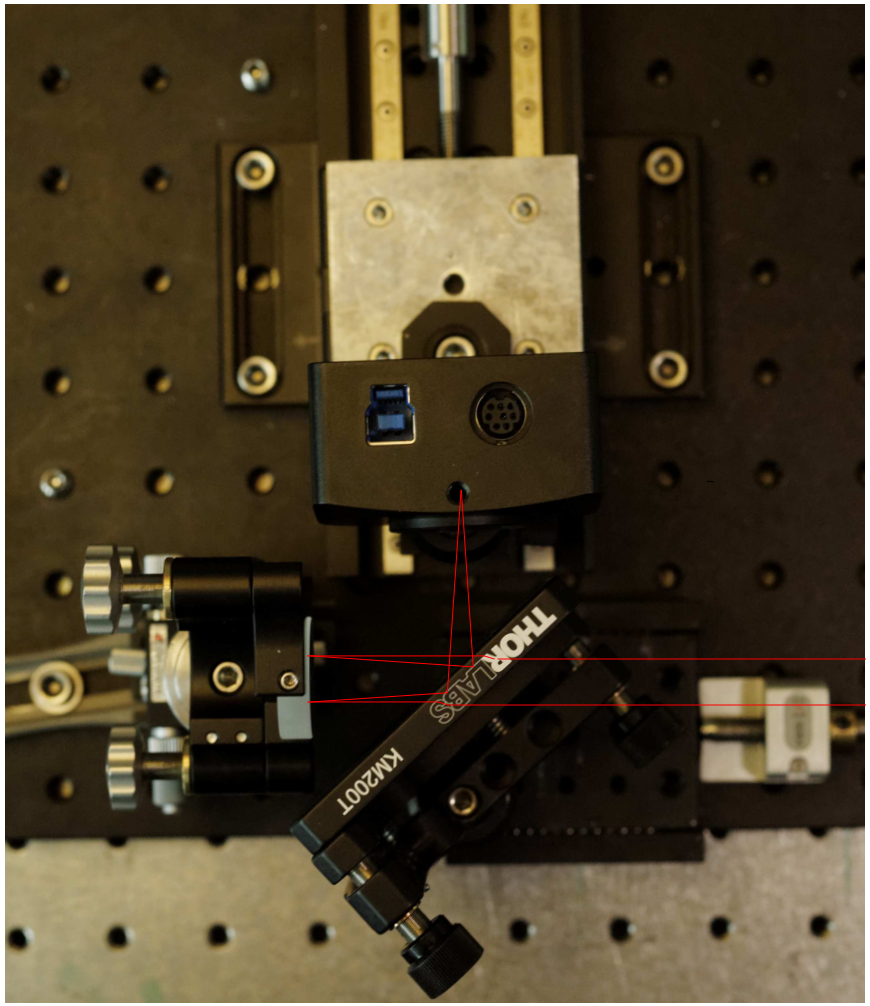
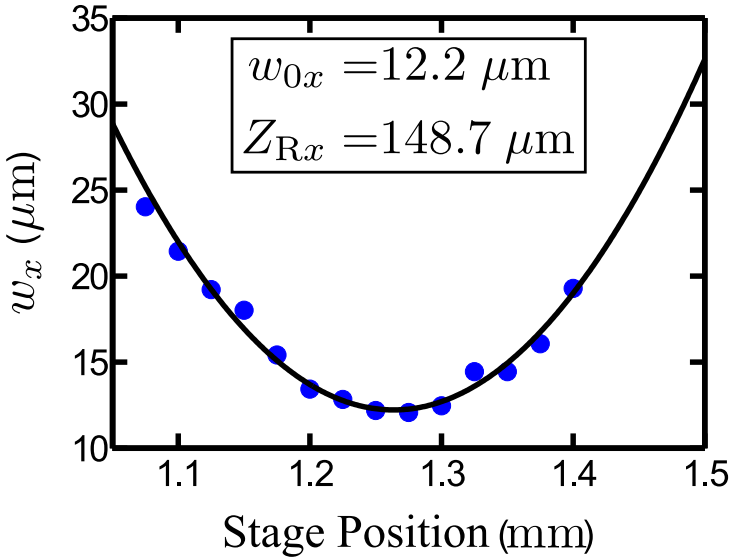
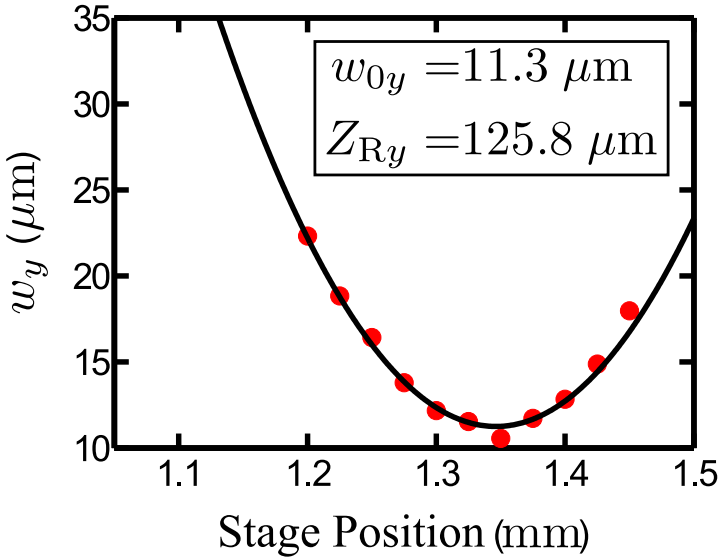


$$w(z) = w_0\sqrt{1+\left(\frac{z}{Z_\mathrm{R}}\right)^2} \simeq w_0 + \frac{w_0}{2}\left(\frac{z}{Z_\mathrm{R}}\right)^2$$

Measuring the minimum spot size (w_0)

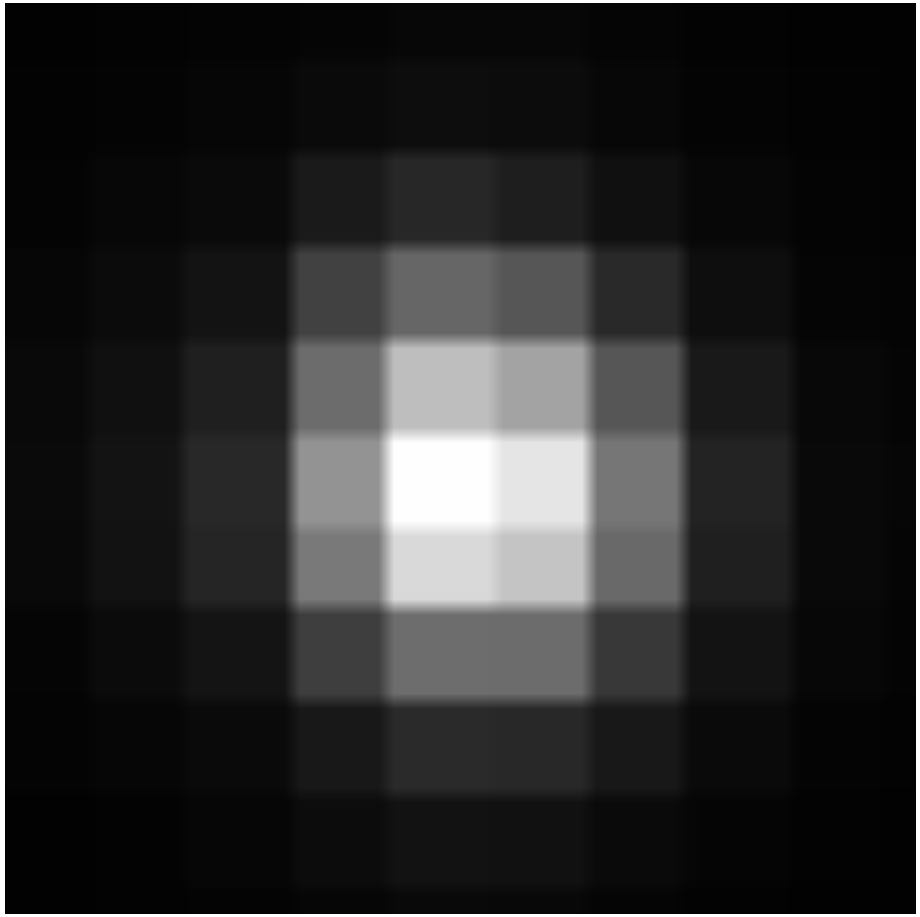


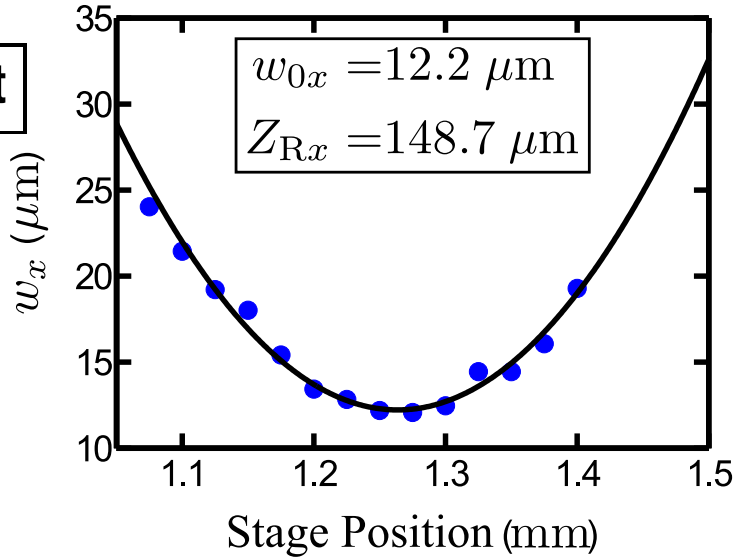


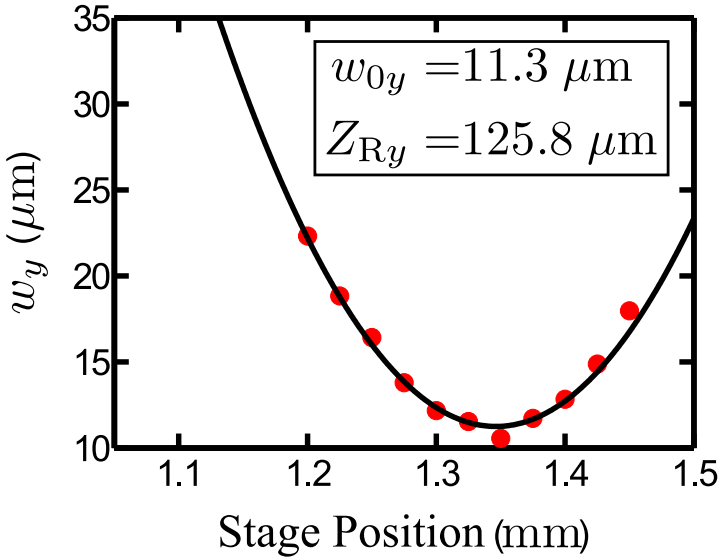


Measuring the minimum spot size (w_0)

We obtain w_0 by finding Z_R from the **fit**



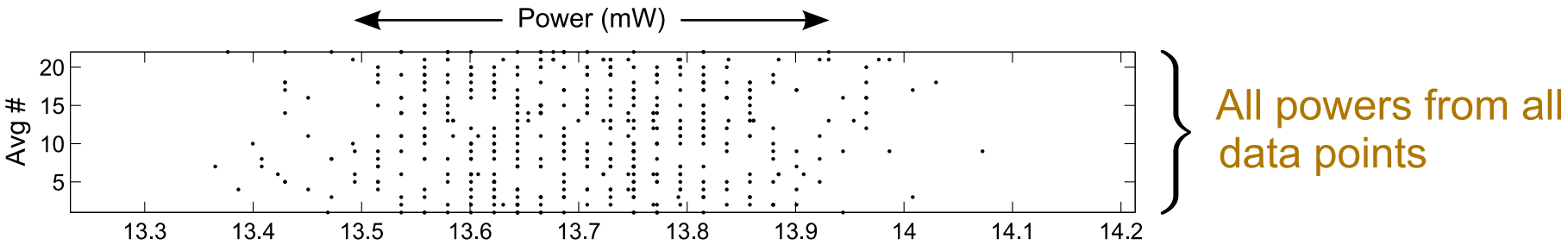




We only consider *valid* data points

Even though power locking helps stabilize the power, small fluctuations still exist, which will adversely affect our results

- We can solve this in the post-data collection analysis by **throwing out** data points corresponding to fluctuations in laser power



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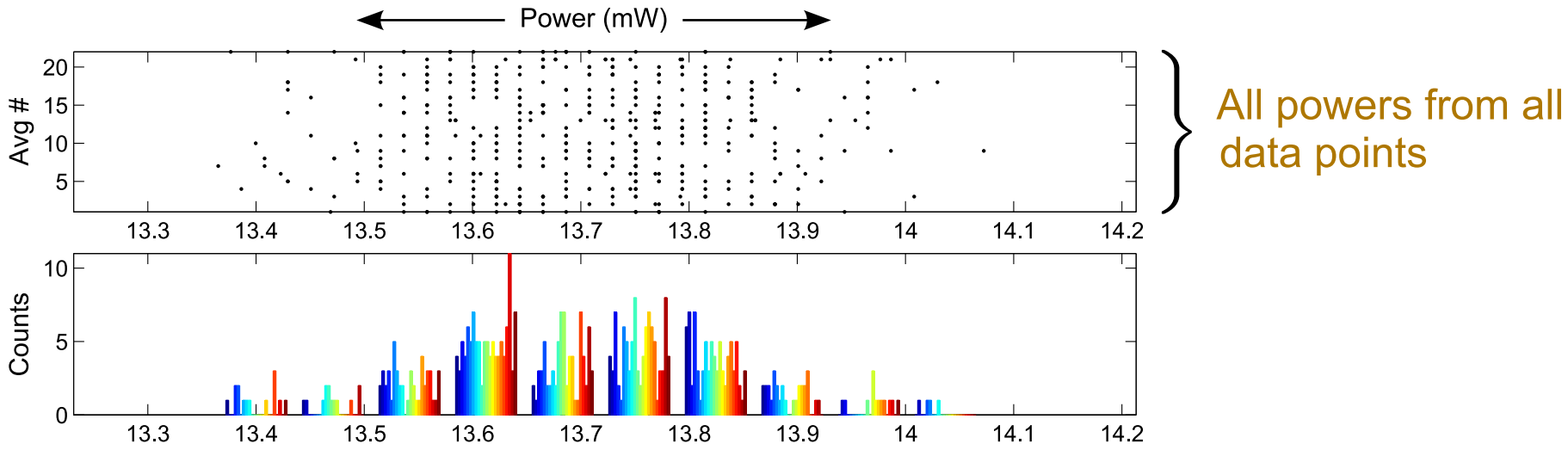

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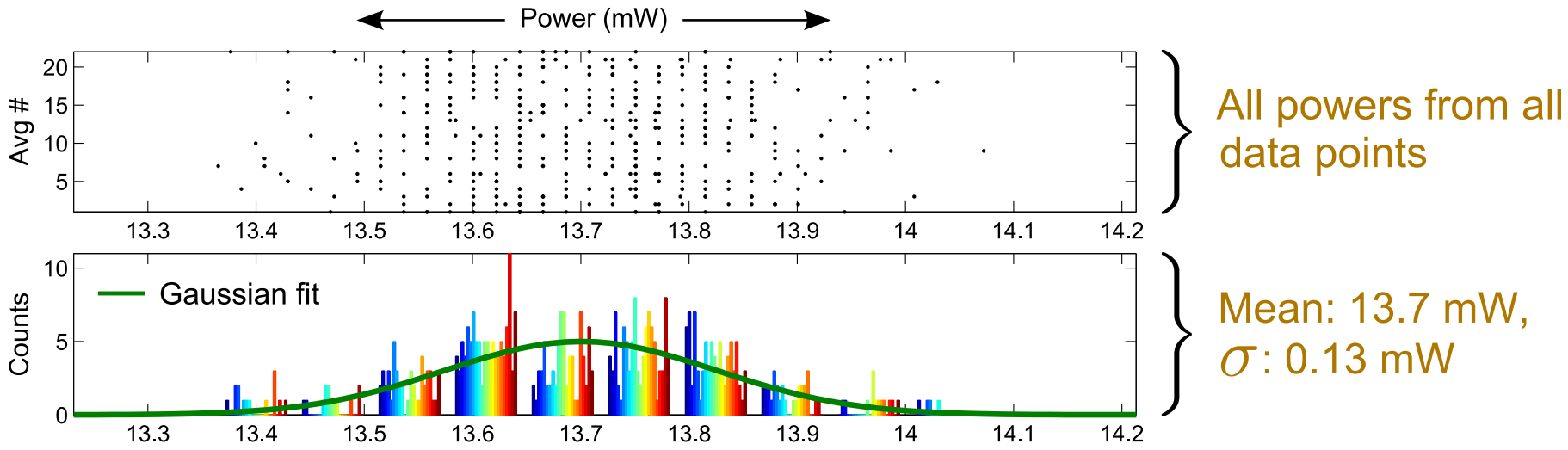

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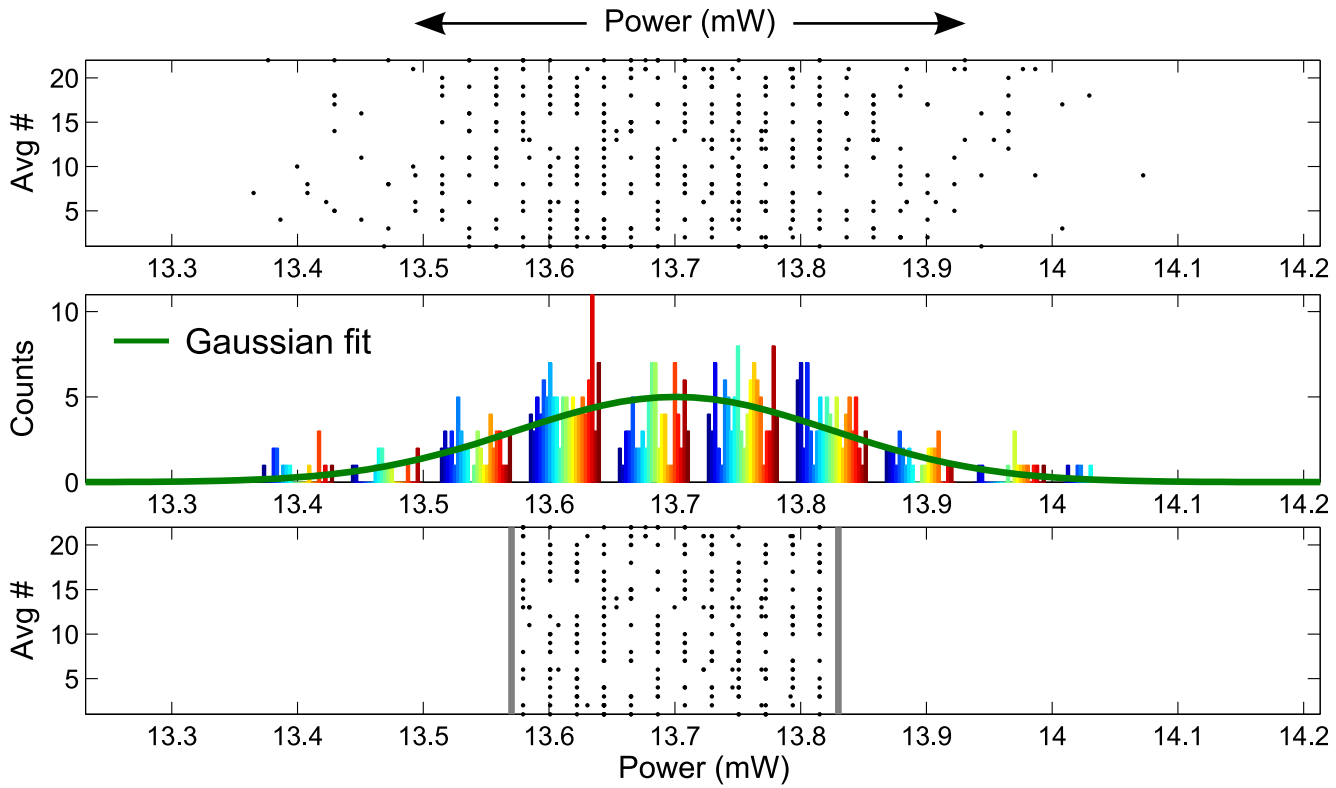


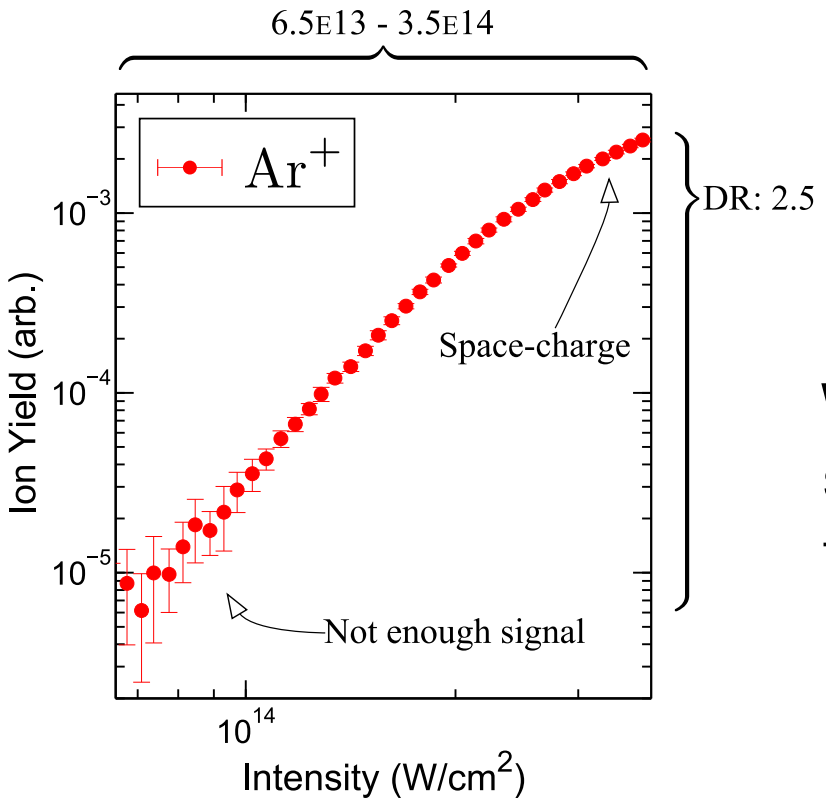
All powers from all data points

Mean: 13.7 mW, σ: 0.13 mW

151 out of 484 points are outside of +/-1σ

The *dynamic range* (DR) of our detection system limits our ability to take scans over a large range of intensities

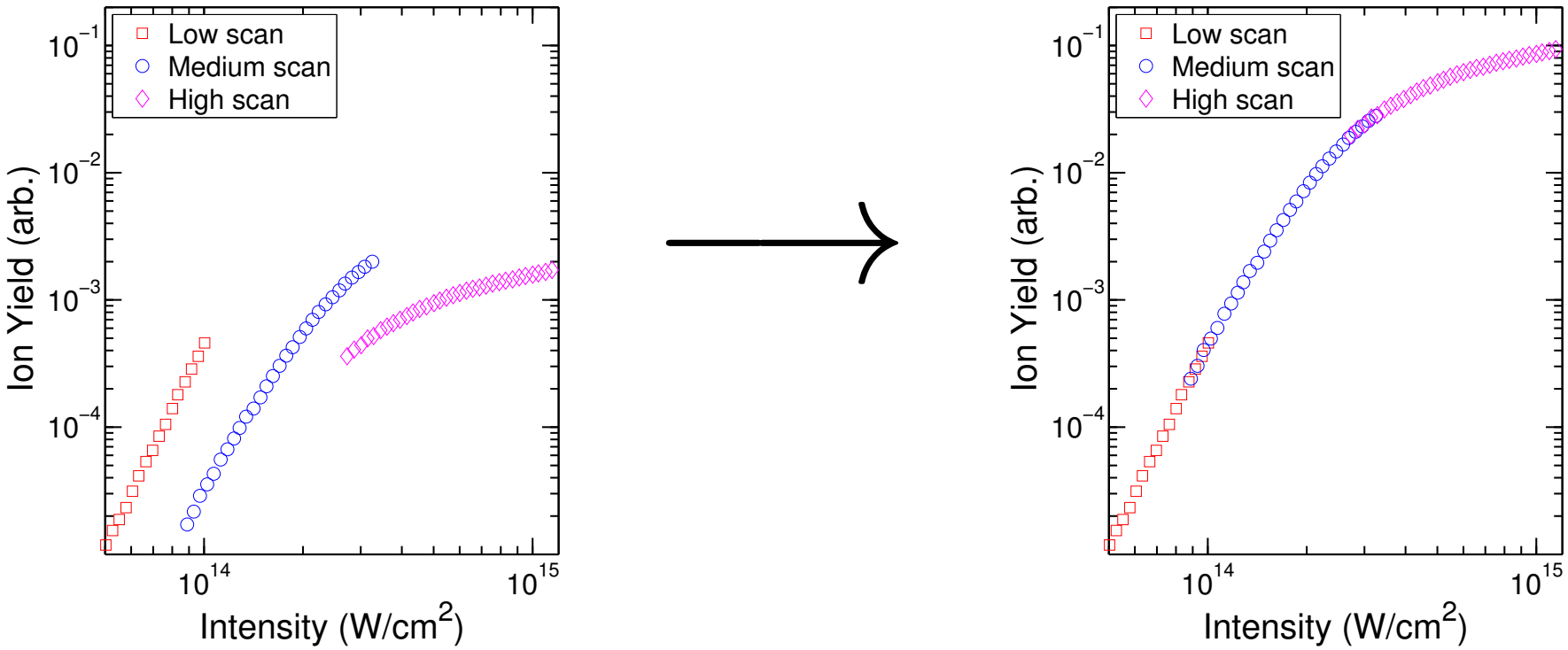
- At **low intensities** the onset of a measurable signal occurs when there are enough ionization events to register a count on the detector
- At **high intensities**, *space-charge* presents due to too many ionization events



This is not good enough

We can circumvent this limitation by taking several *different* scans and **stitching** them together

Stitching different scans

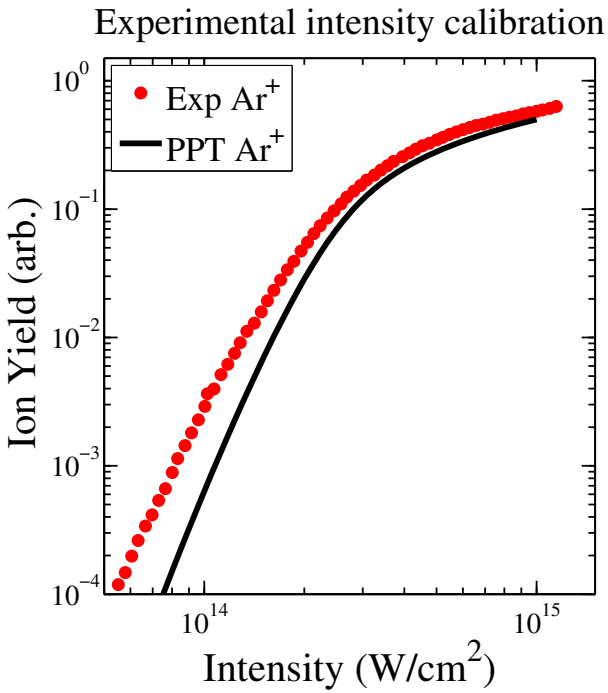


Scan	Intensity Range (W/cm^2)	Dynamic Range	Pressure (Torr)
Low	5.0E13 – 1.0E14	1.6	1.1E-5
Medium	8.8E13 – 3.3E14	2.1	3.2E-7
High	2.7E14 – 1.1E15	0.7	4.9E-8
$\sum$	5.0E13 – 1.1E15	3.9	-

Absolute intensity calibration

We applied an **absolute intensity calibration** on our data

- This is achieved by *shifting* the intensity scale of our **atomic** ion yield to the $\boxed{PPT}$ rate for the same ion



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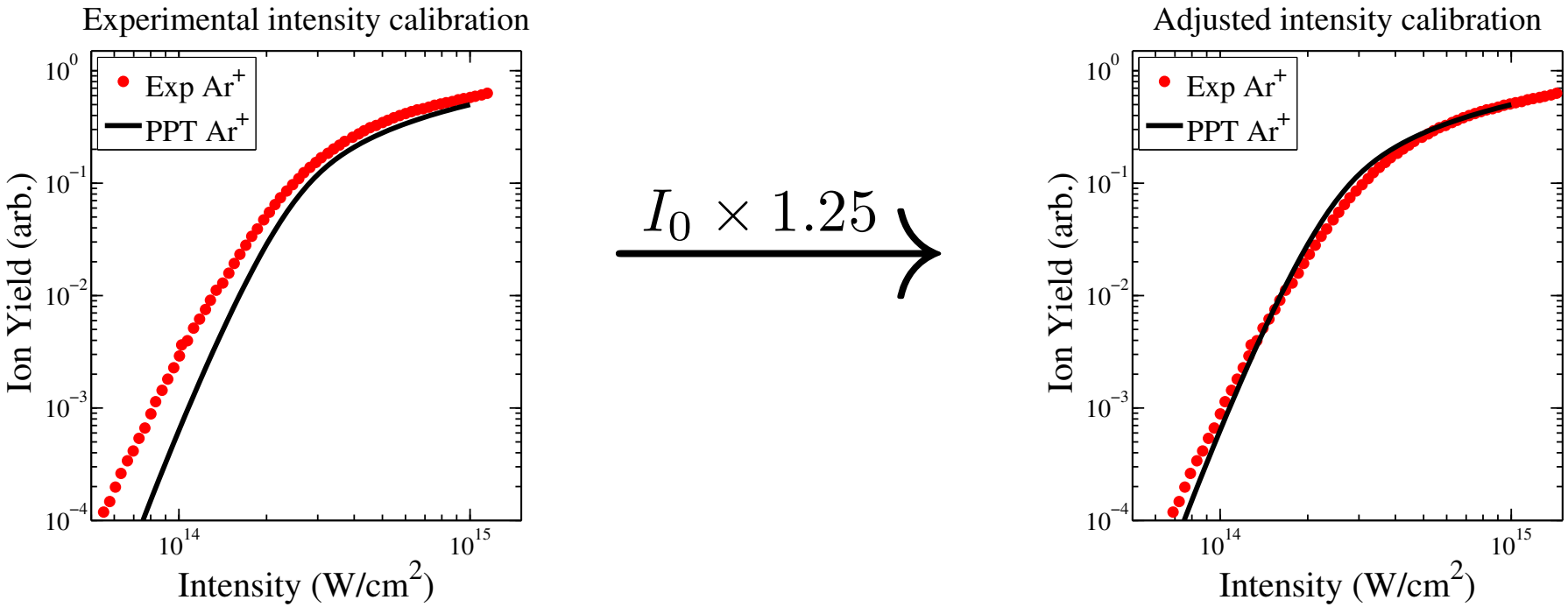
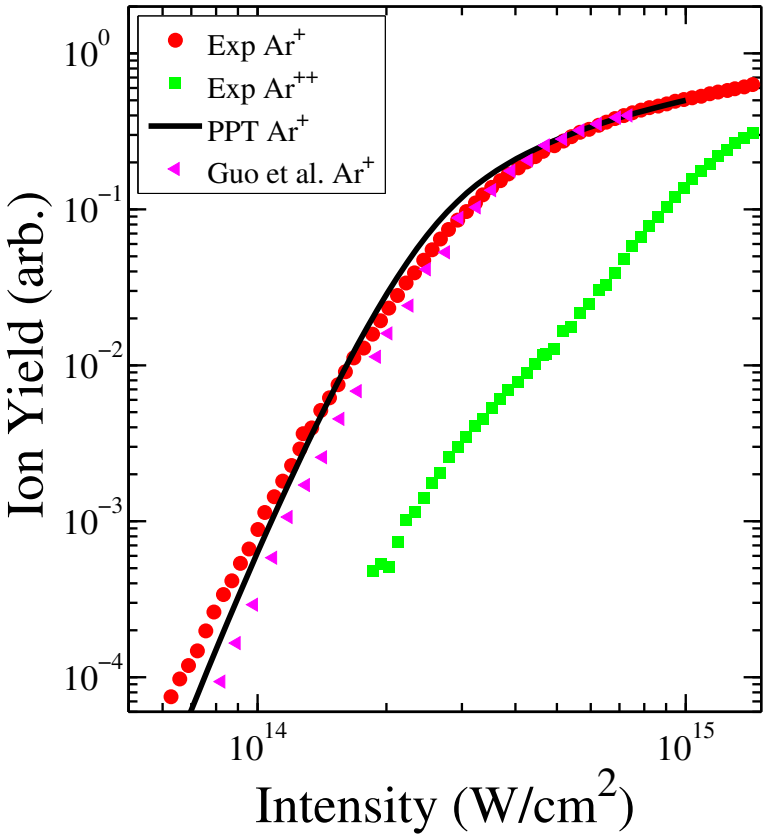
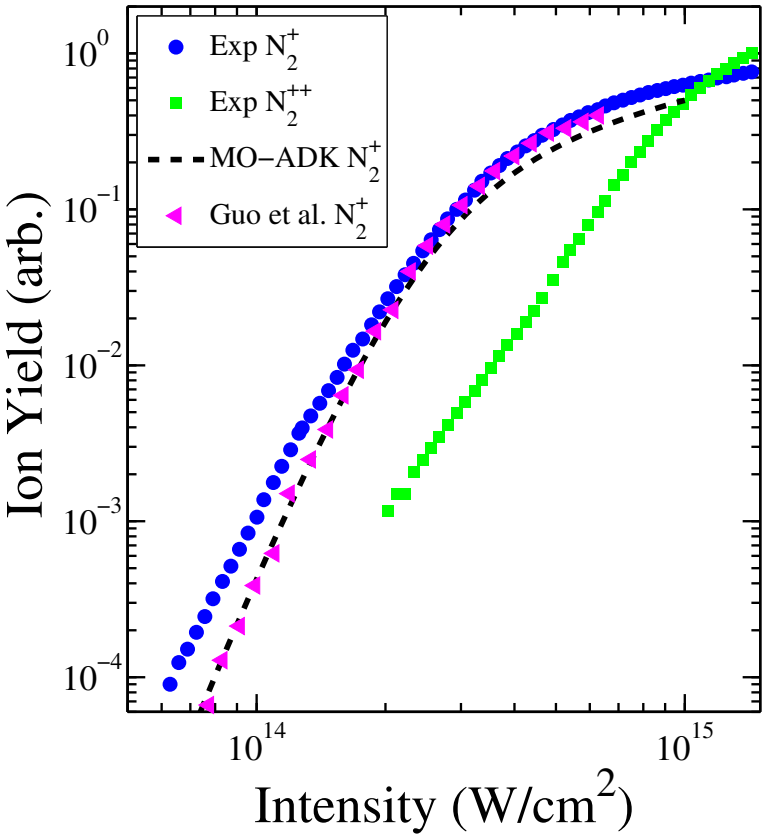


- This shift factor is applied to the simultaneously measured **molecular** yield curve
- This calibration is repeated for each **molecule**/**atom** pair

Results for N_2/Ar



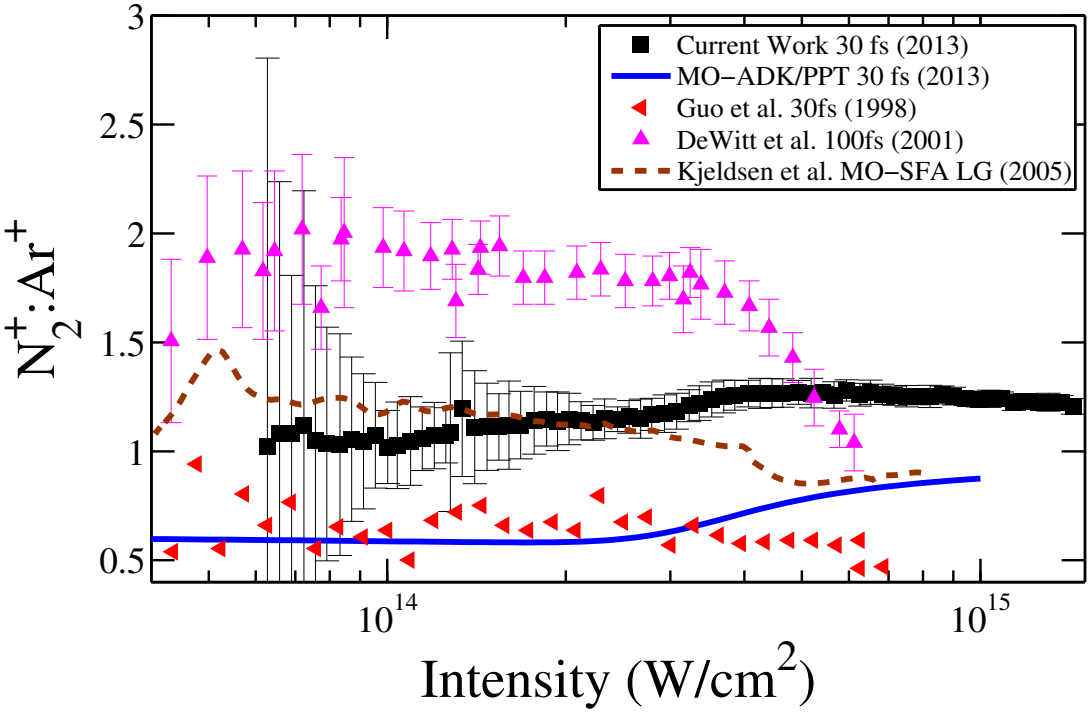


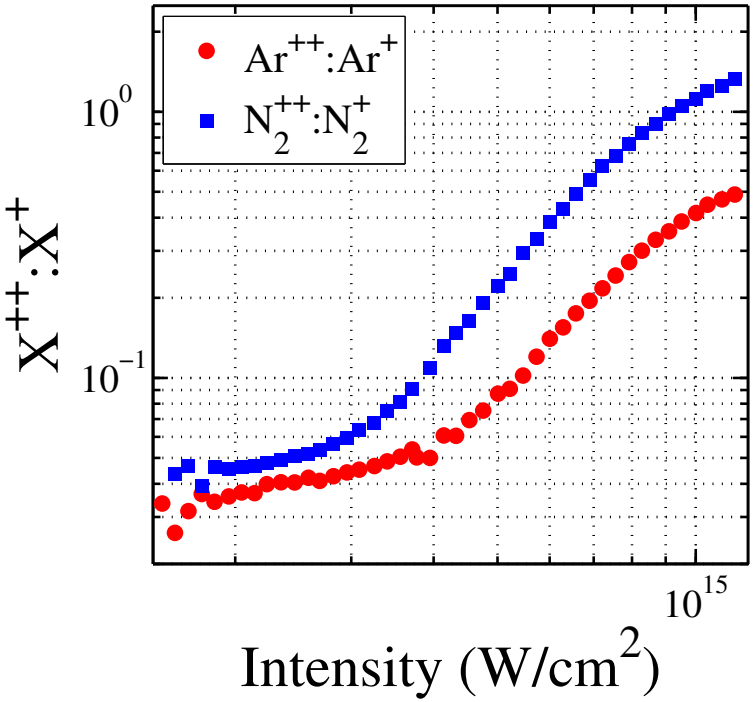
- Guo *et al.* [5] used **ADK** to calibrate the intensity

[5] C. Guo, M. Li, J. Nibarger, and G. N. Gibson, Phys. Rev. A **58**, R4271 (1998).

Results for N_2/Ar

$I_{\text{cut-off}} \sim 4 \times 10^{14}$ W/cm^2





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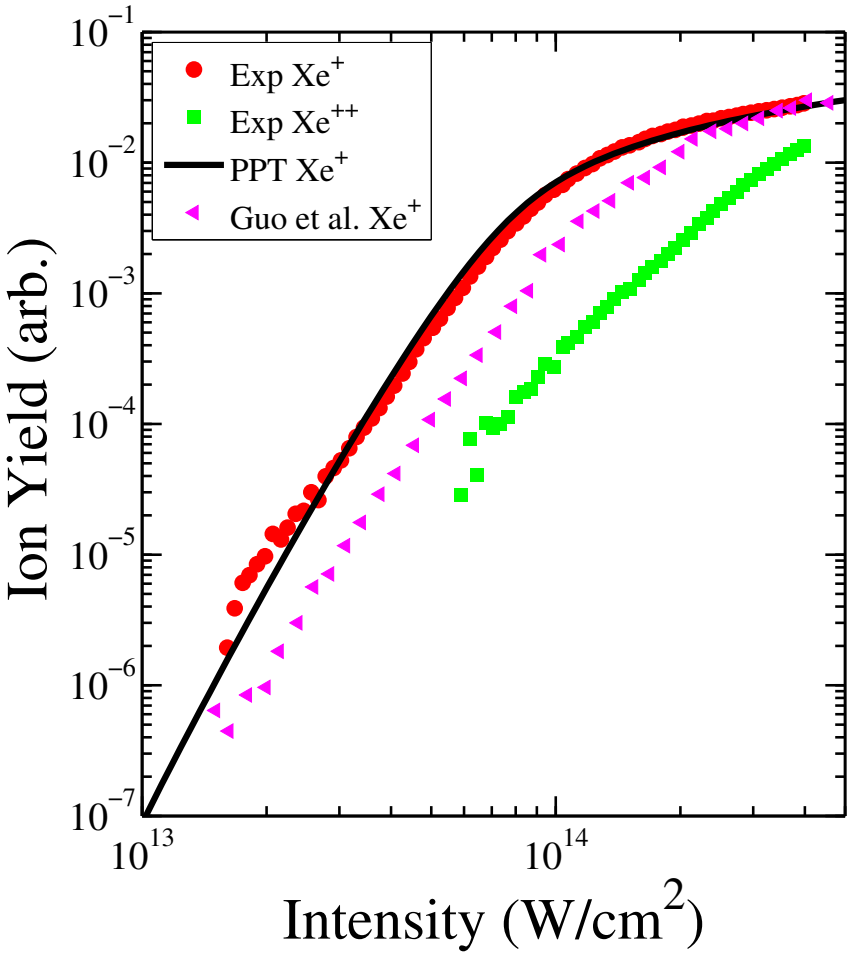
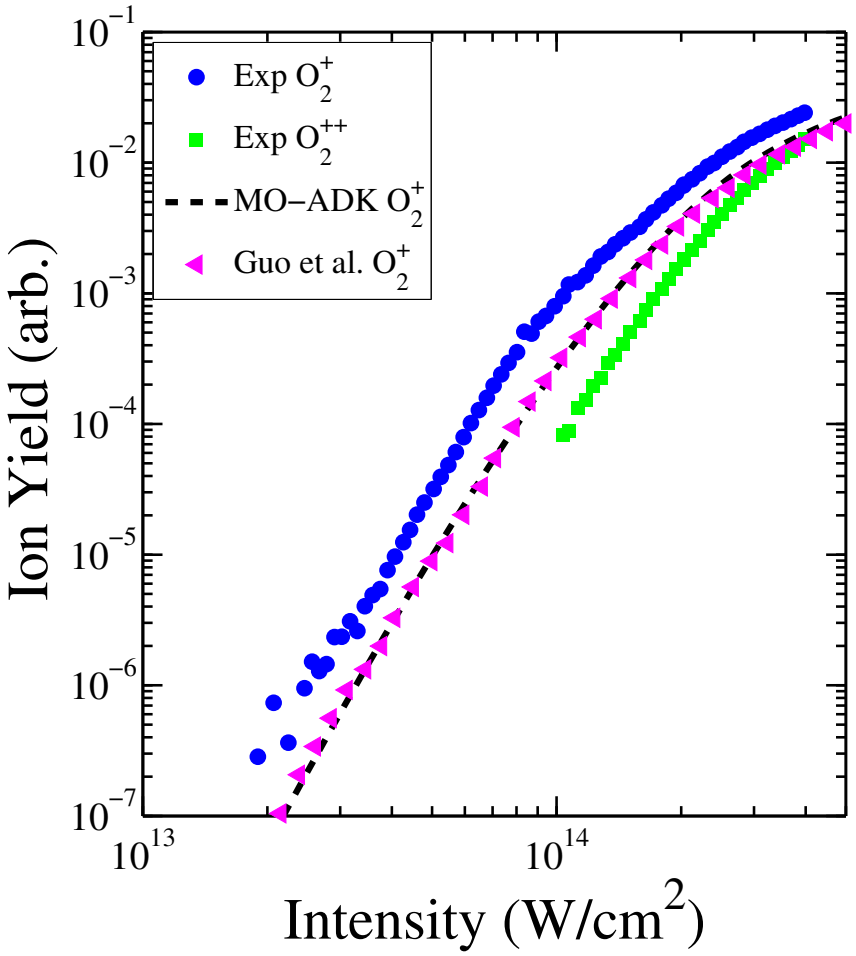
[7] T. K. Kjeldsen and L. B. Madsen, Phys. Rev. A **71**, 023411 (2005).

Results for O_2/Xe

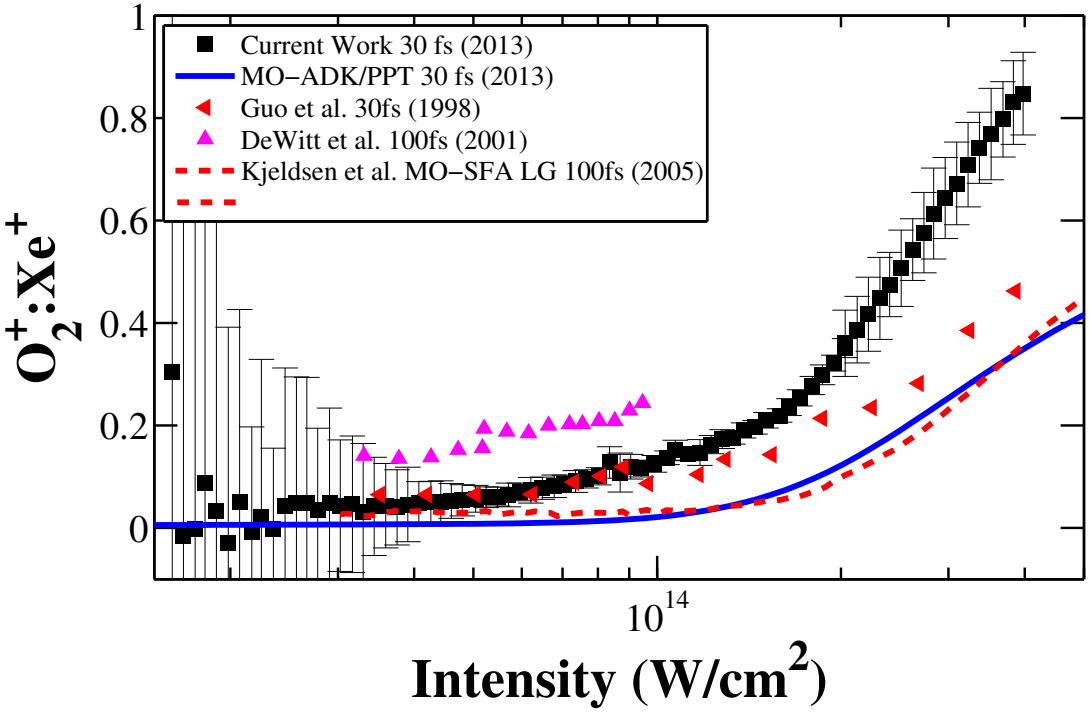


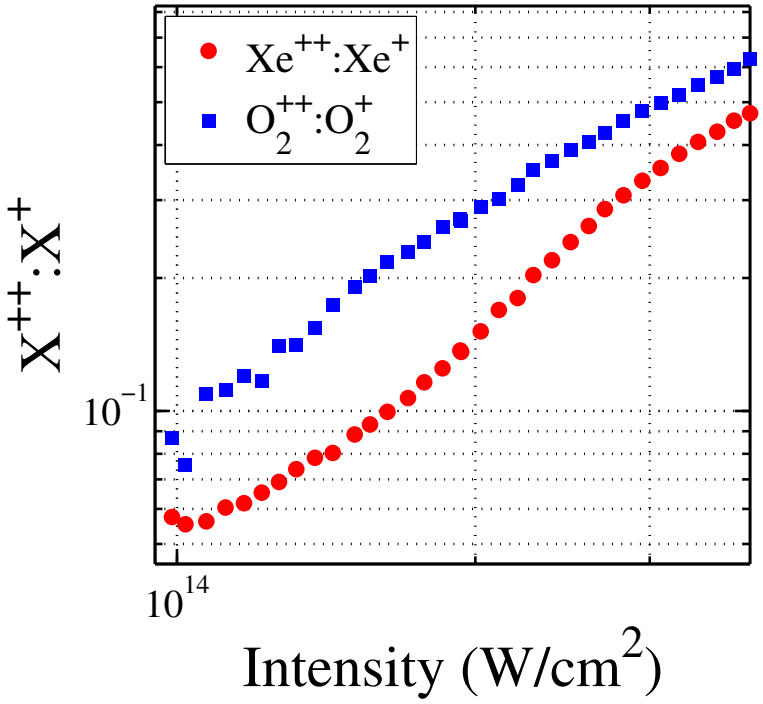


[5] C. Guo, M. Li, J. Nibarger, and G. N. Gibson, Phys. Rev. A **58**, R4271 (1998).

Results for O_2/Xe

$I_{\text{cut-off}} \sim 1 \times 10^{14}$ W/cm^2





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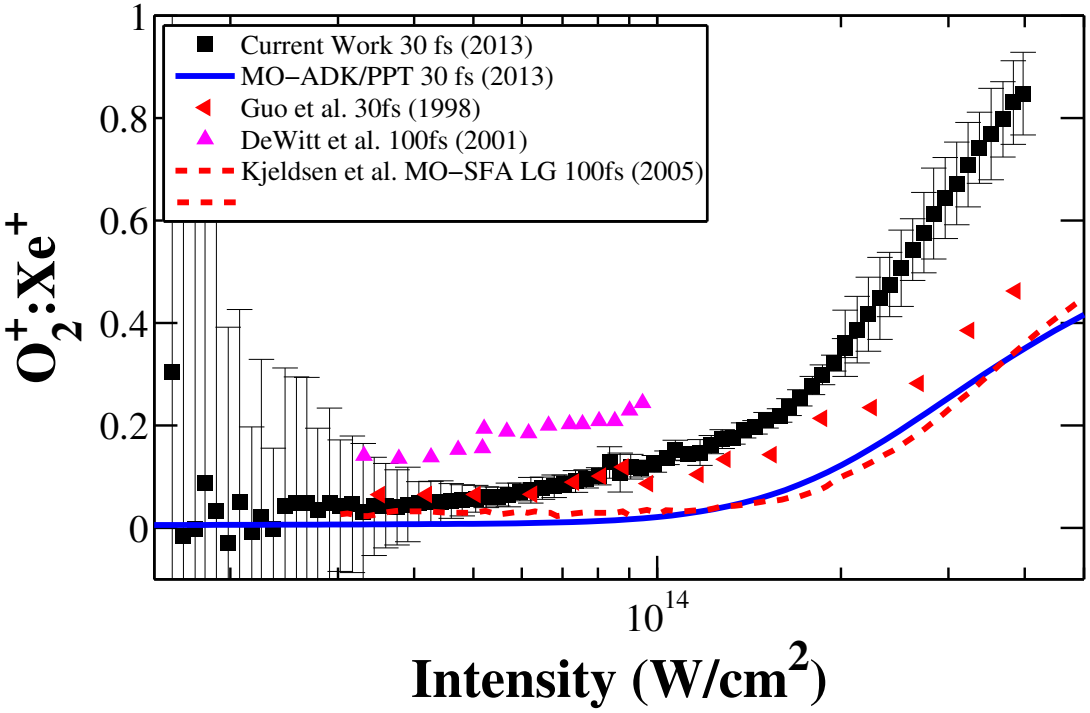

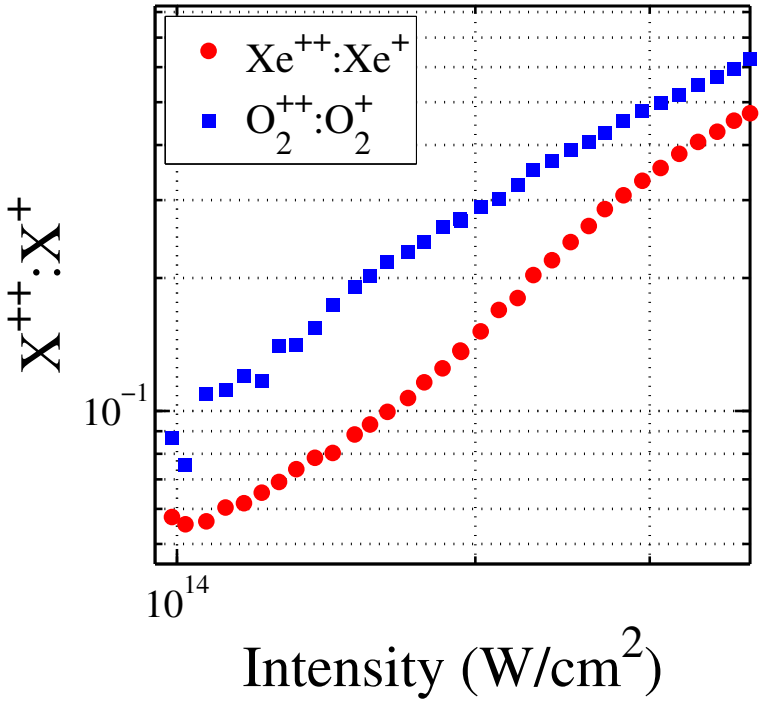

Ionization suppression is present in O_2!

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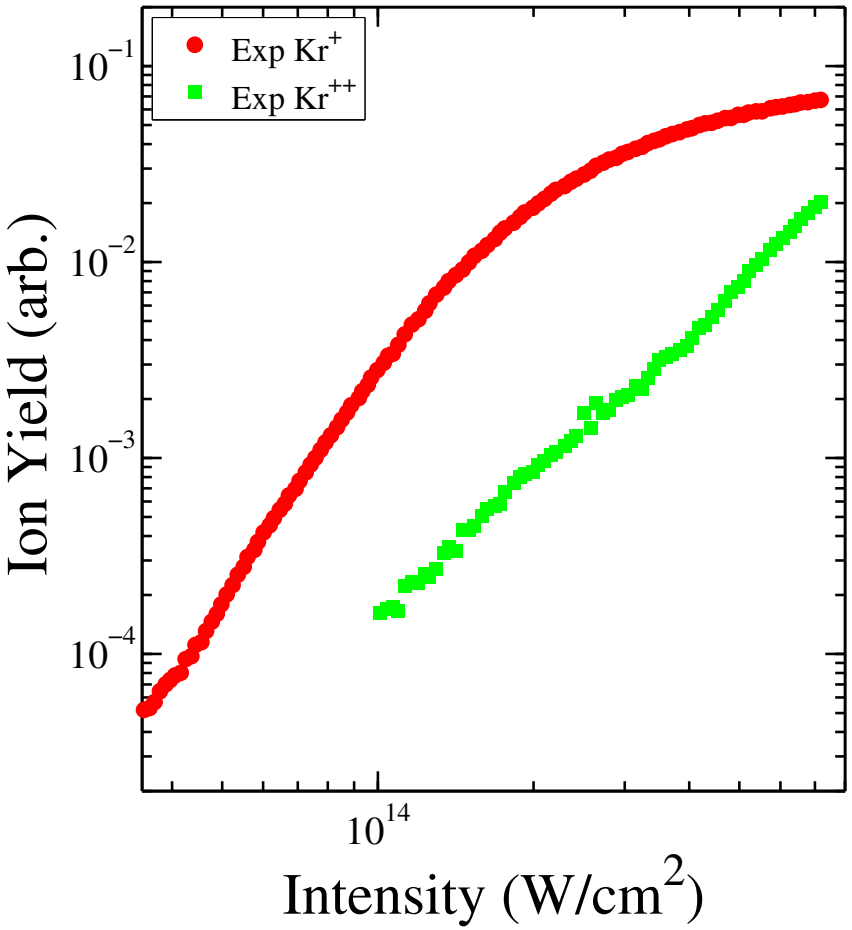
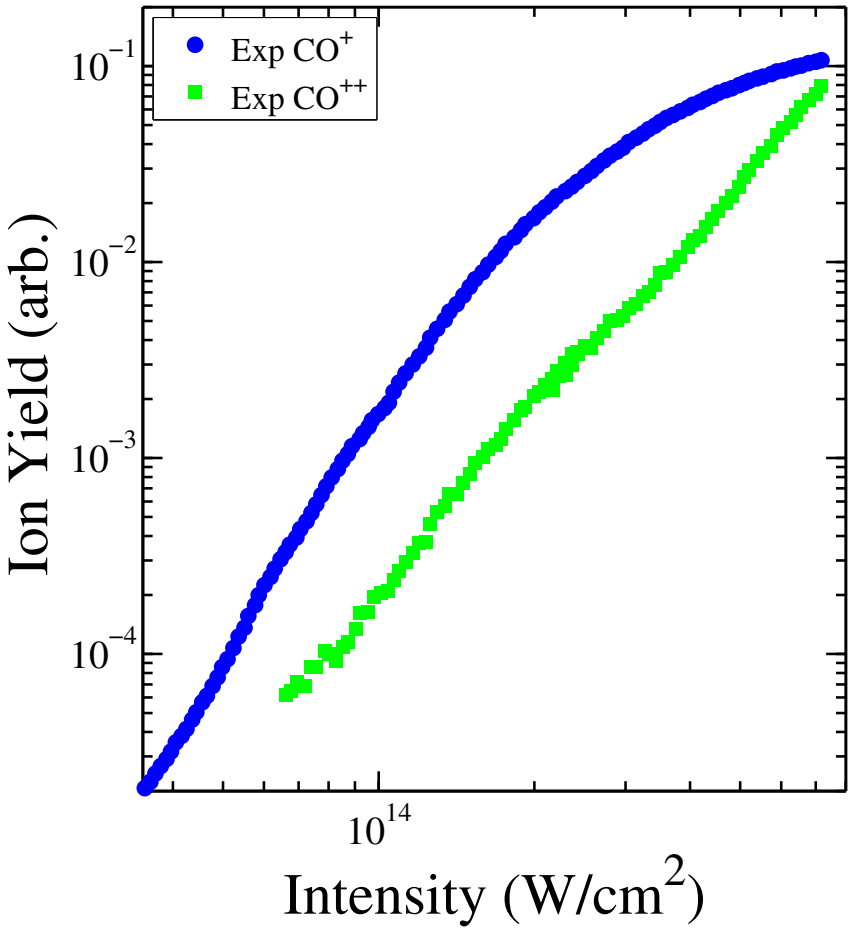
Results for CO/Kr





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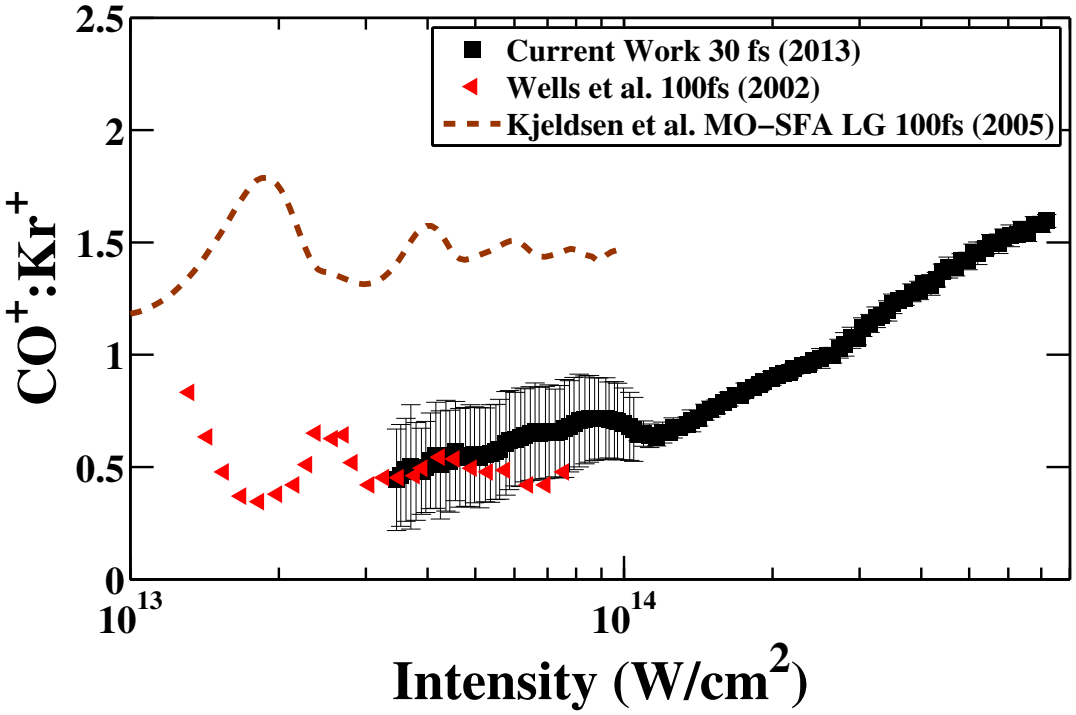

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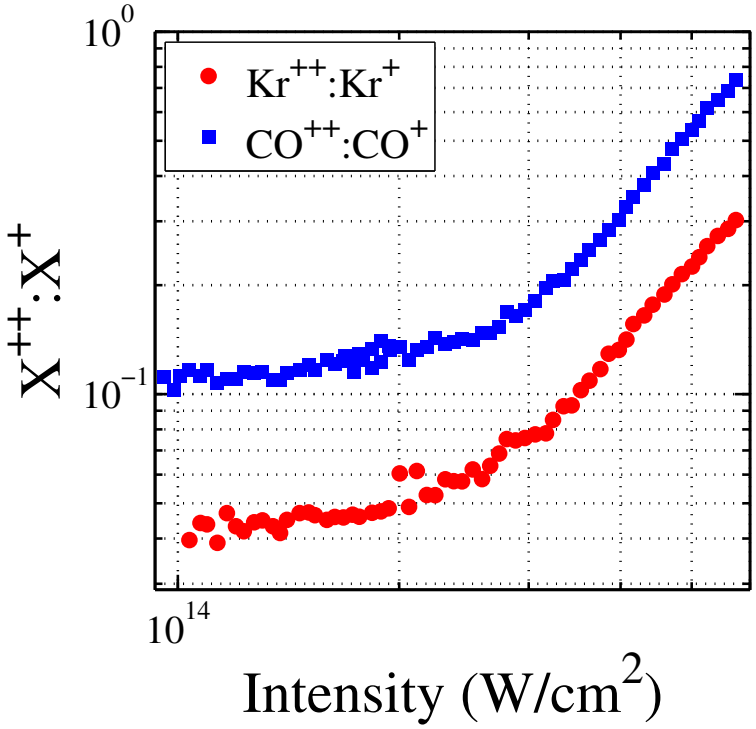

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