fs-TALIF in RAID
ENR project report 2025

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Outline

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1. Introduction
2. Implementation
3. Experiments
4. Other studies
5. Scientific output
6. Conclusions and outlook

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Introduction

Our objective is

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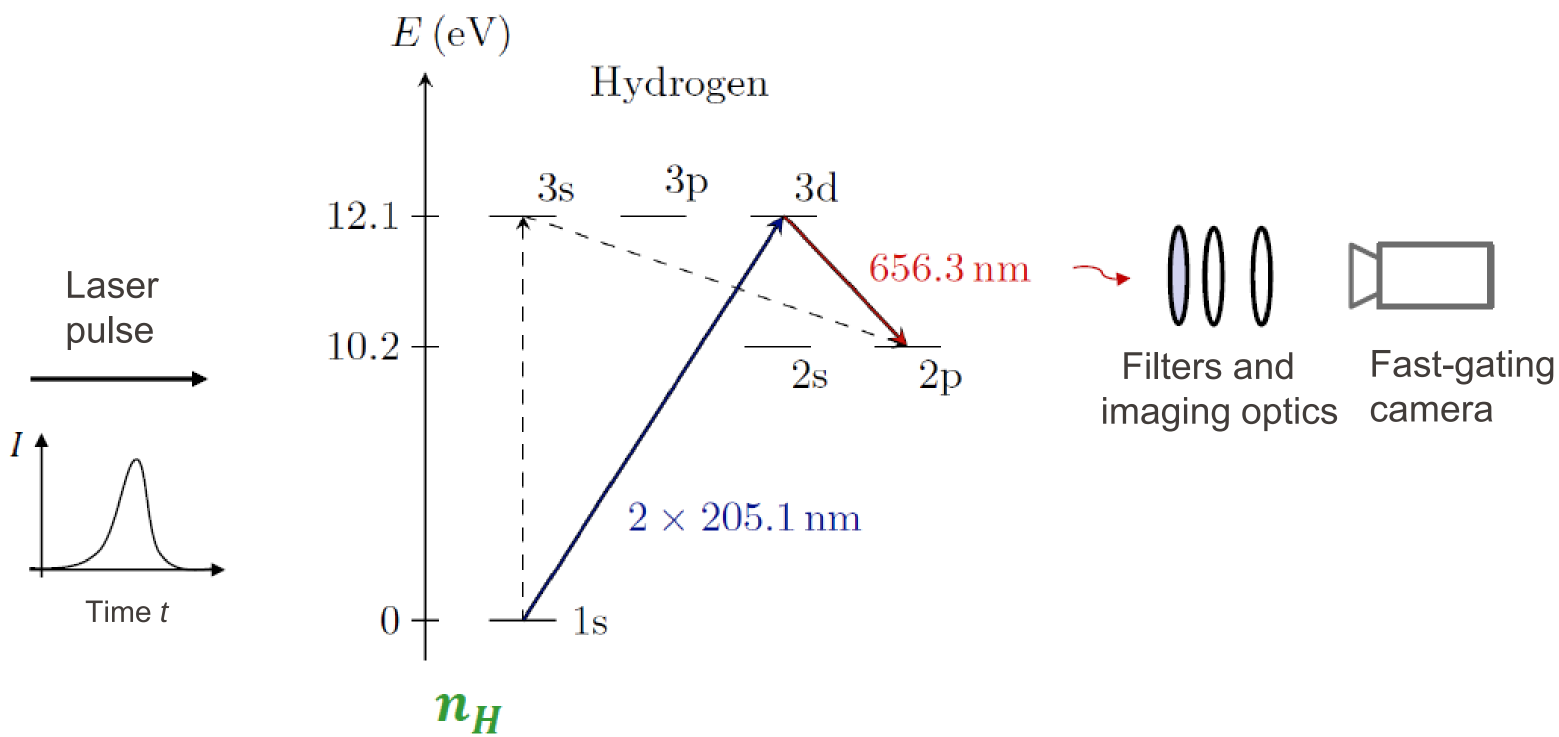
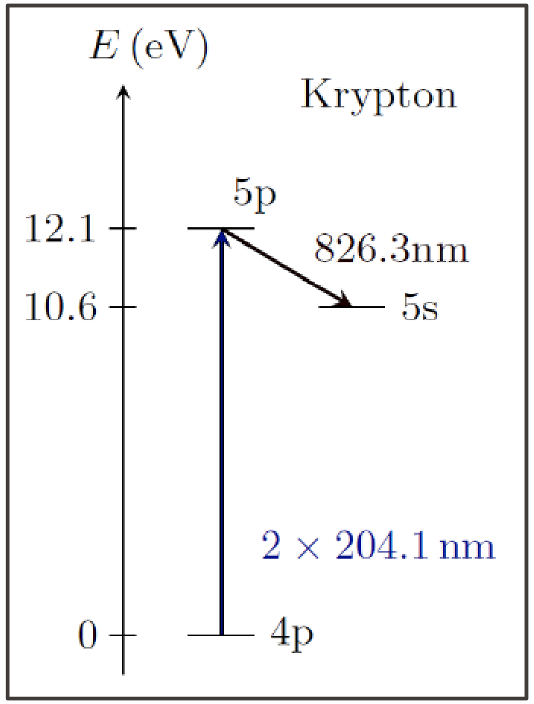
*Obtain **single laser-pulse** measurements of H densities in a dense hydrogen plasma using fs-TALIF.*

Provide an assessment of the feasibility of this method for application in magnetic confinement fusion research.

We use the Two-photon absorption laser-induced fluorescence (TALIF) technique



TALIF in Kr
(for calibration)



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We use the Two-photon absorption laser-induced fluorescence (TALIF) technique

$$S_{\mathrm{H}}(\nu_L) = n_{\mathrm{H}}\, \beta_{\mathrm{H}}\, a_{\mathrm{H}}\, g_{\mathrm{H}}(\nu_L)\, \frac{G\, \sigma_{\mathrm{H}}^{(2)}}{(h\nu_L)^2} \int_{\mathrm{obs.}V} \int_{\mathrm{pulse}} I^2(\mathbf{r}', t')\, dt'\, d\mathbf{r}'$$

- $S_{\mathrm{H}}(\nu_L)$: Total fluorescence photons from observation volume in one laser pulse
- n_{H}: H density
- β_{H}: Detection efficiency (includes geometry, transmission, etc.)
- a_{H}: Fluorescence branching ratio
- $g_{\mathrm{H}}(\nu_L)$: Normalized absorption line profile
- G: Photon stat. factor
- $\sigma_{\mathrm{H}}^{(2)}$: Two-photon absorption cross section
- $I^2(\mathbf{r}', t')$: Laser intensity (squared)

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We use the Two-photon absorption laser-induced fluorescence (TALIF) technique

Total fluorescence photons from observation volume in one laser pulse

Fluorescence branching ratio

Photon stat. factor

Two-photon absorption cross section

$$S_{\mathrm{H}}(\nu_L) = \eta_{\mathrm{H}}\, \beta_{\mathrm{H}}\, a_{\mathrm{H}}\, g_{\mathrm{H}}(\nu_L)\, \frac{G\, \sigma_{\mathrm{H}}^{(2)}}{(h\nu_L)^2} \int_{\mathrm{obs.}V} \int_{\mathrm{pulse}} I^2(\mathbf{r}', t')\, dt'\, d\mathbf{r}'$$

Valid only for "sufficiently" low laser intensities

H density

Detection efficiency (includes geometry, transmission, etc.)

Normalized absorption line profile

Laser intensity (squared)

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TALIF in Kr gas is used to calibrate the H TALIF signal

H density

"Known" ratio of absorption cross sections

Ratio of relative absorptions at ν_L

Ratio of pulse energies (squared) used in H and Kr measurements

$$\frac{n_{\mathrm{H}}}{n_{\mathrm{Kr}}} = \frac{\beta_{\mathrm{Kr}}}{\beta_{\mathrm{H}}} \left(\frac{\sigma^{(2)}_{\mathrm{Kr}}}{\sigma^{(2)}_{\mathrm{H}}}\right) \left(\frac{a_{\mathrm{Kr}}}{a_{\mathrm{H}}}\right) \left(\frac{g_{\mathrm{Kr}}(\nu_L)}{g_{\mathrm{H}}(\nu_L)}\right) \left(\frac{E^2_{\mathrm{pulse,Kr}}}{E^2_{\mathrm{pulse,H}}}\right) \left(\frac{S_{\mathrm{H}}(\nu_L)}{S_{\mathrm{Kr}}(\nu_L)}\right)$$

Ratio of detection efficiencies

Branching ratio of H fluorescence

Ratio of TALIF signals in H and in Kr

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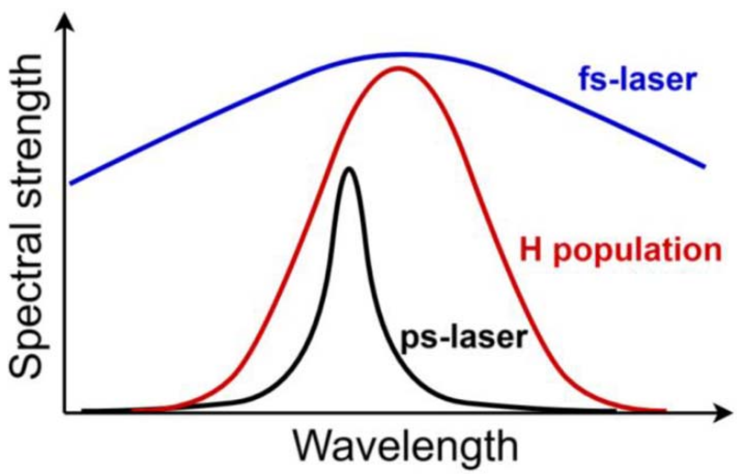
We investigate TALIF with fs pulses

- fs pulses are spectrally very broad and significantly larger than the absorption linewidth of H in a **tokamak divertor or SOL** ($T_e \lesssim 10$ eV, $B \approx 1$ T).

They can excite the entire H population with a single laser pulse.



- Furthermore, fs pulses can be extremely intense at similar pulse energies as ps lasers.

- Since fs pulses are spectrally very broad, a Kr TALIF signal can in principle be obtained with ν_L tuned to the H TALIF resonance.

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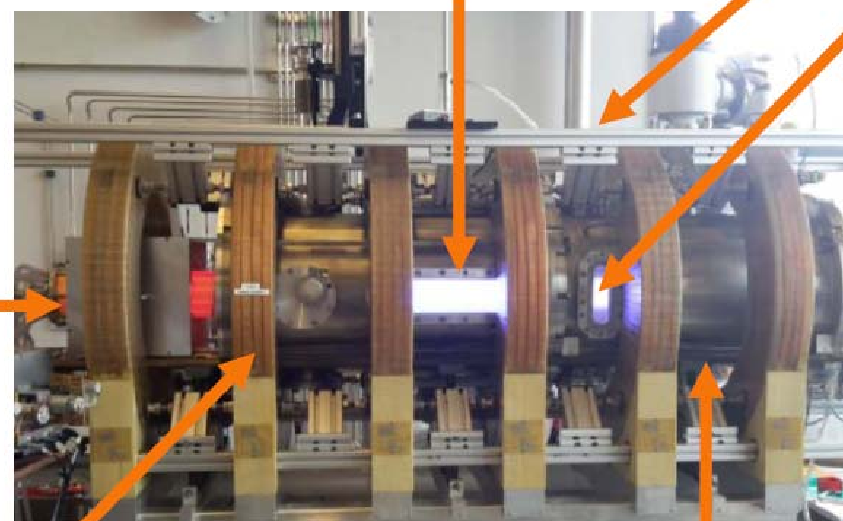
Introduction

Experiments are carried out in the Resonant Antenna Ion Device (RAID) at SPC

$B_0 = 100 - 800\,\mathrm{G}$
$p_{H2} = 0.1 - 2.0\,\mathrm{Pa}$
$P_{RF} \leq 2 \times 10\,\mathrm{kW}$
$n_e \leq 5 \cdot 10^{18}\ \mathrm{m}^{-3}$ (for H)
$T_e = 1 - 5\,\mathrm{eV}$ (for H)

Helicon plasma column



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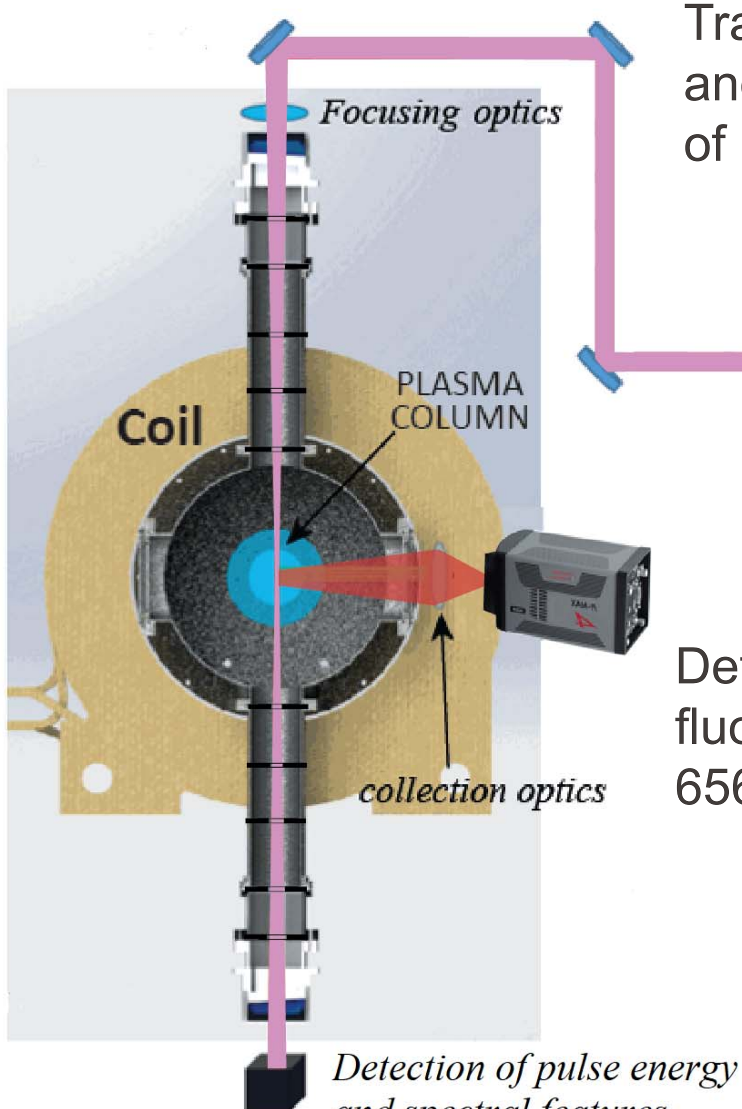
Introduction Proposed system



Transport and injection of UV beam

Generation of laser pulses at 205 nm (UV)

Detection of fluorescence at 656 nm (vis)

Diagnosis of laser pulses

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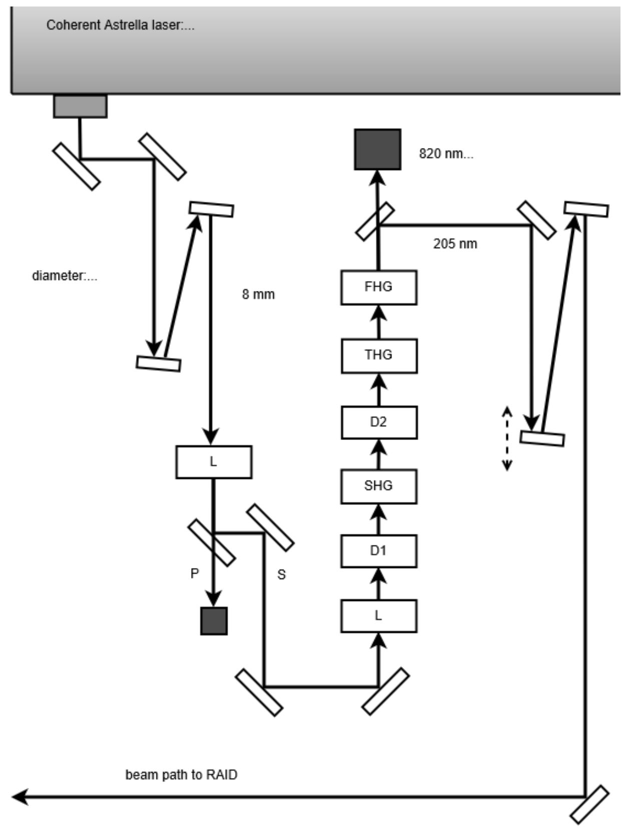
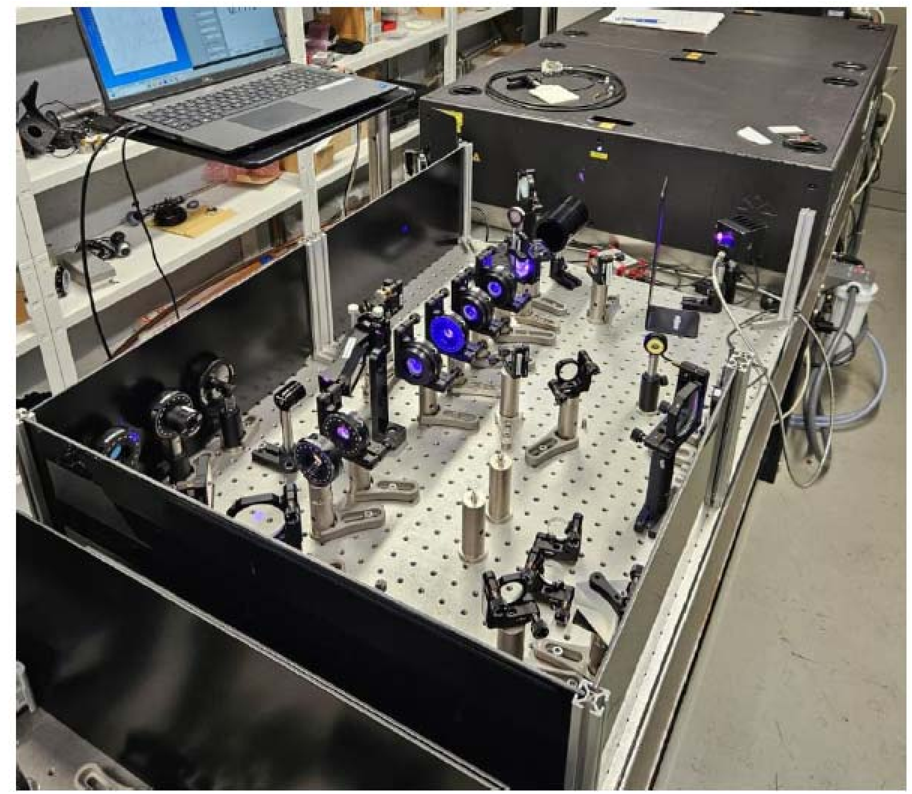
Implementation of laser (T1) FHG module and Astrella upgrade

- We built a fourth harmonic generator (FHG) to obtain 205 nm from an upgraded Astrella system (1kHz rep-rate, 7 mJ per pulse at 820 nm, pulse duration = 70 fs).
- The FHG is based on the design proposed by Susnjar et al. *Opt. Comm.* (2023)





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Implementation of laser (T1)
Characterization of UV fs pulses

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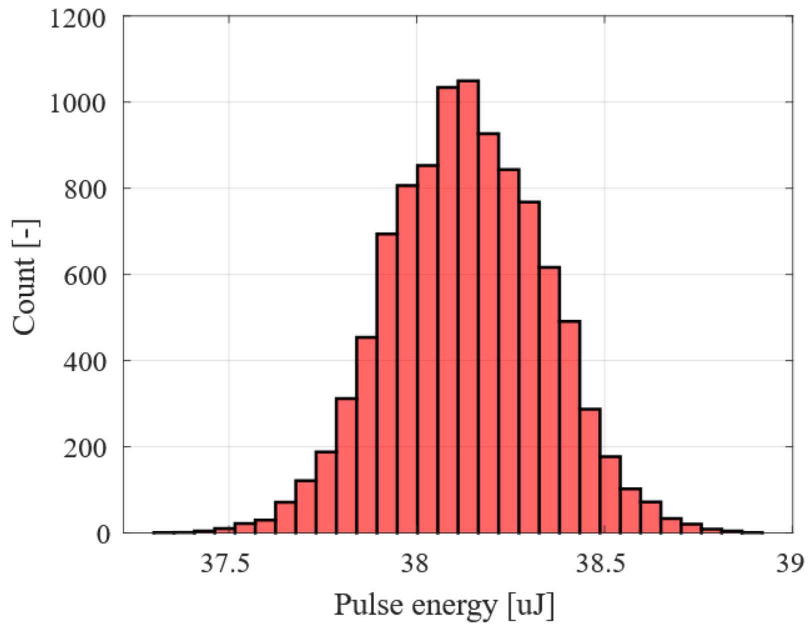
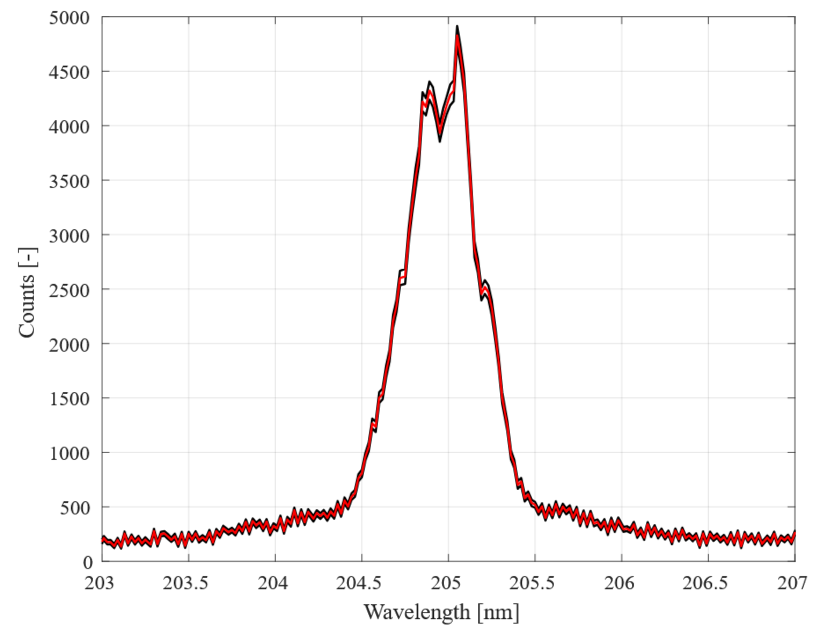
- FHG and Astrella laser were configured for maximum energy while obtaining a wide and symmetric spectrum centered at 205.1 nm.





- Histogram of energy of 10000 pulses shows stable values.

- Stable short-term spectral features. Red curve is the average of 1000 consecutive spectra, each one obtained from 100 pulses.

Implementation of laser (T1)
Characterization of UV fs pulses

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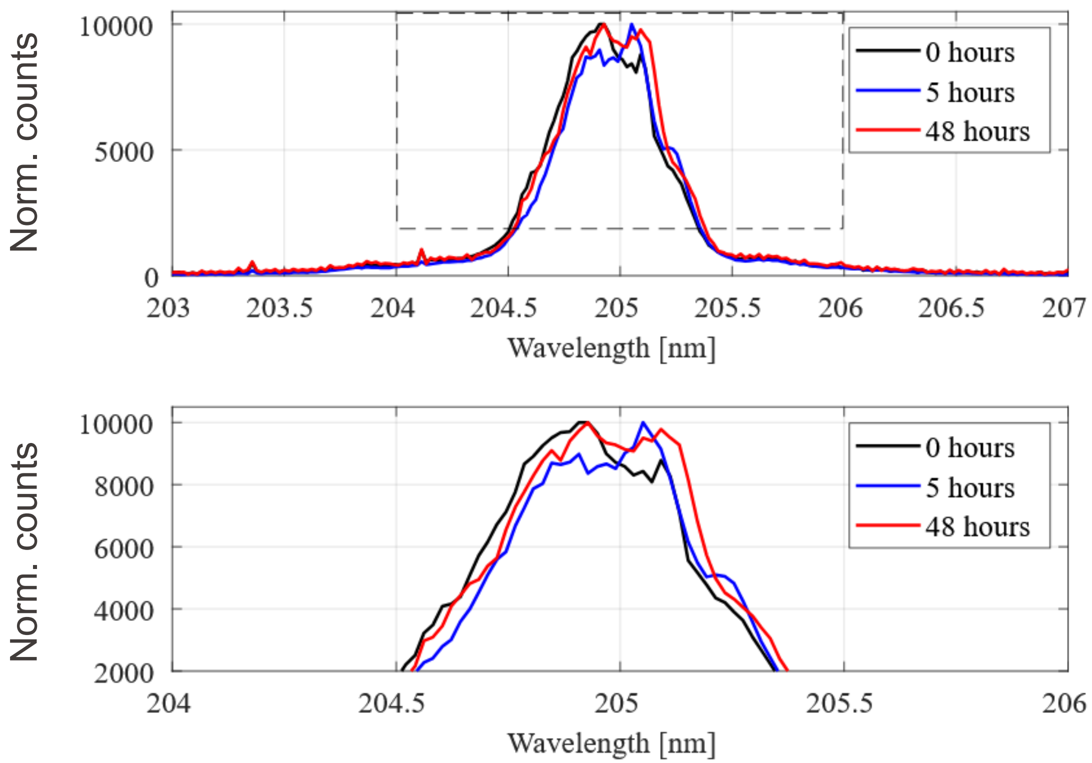
- FHG and Astrella laser were configured for maximum energy while obtaining a wide and symmetric spectrum centered at 205.1 nm.



- Long-term spectral features are stable but require monitoring.

Implementation of beam path (T3)

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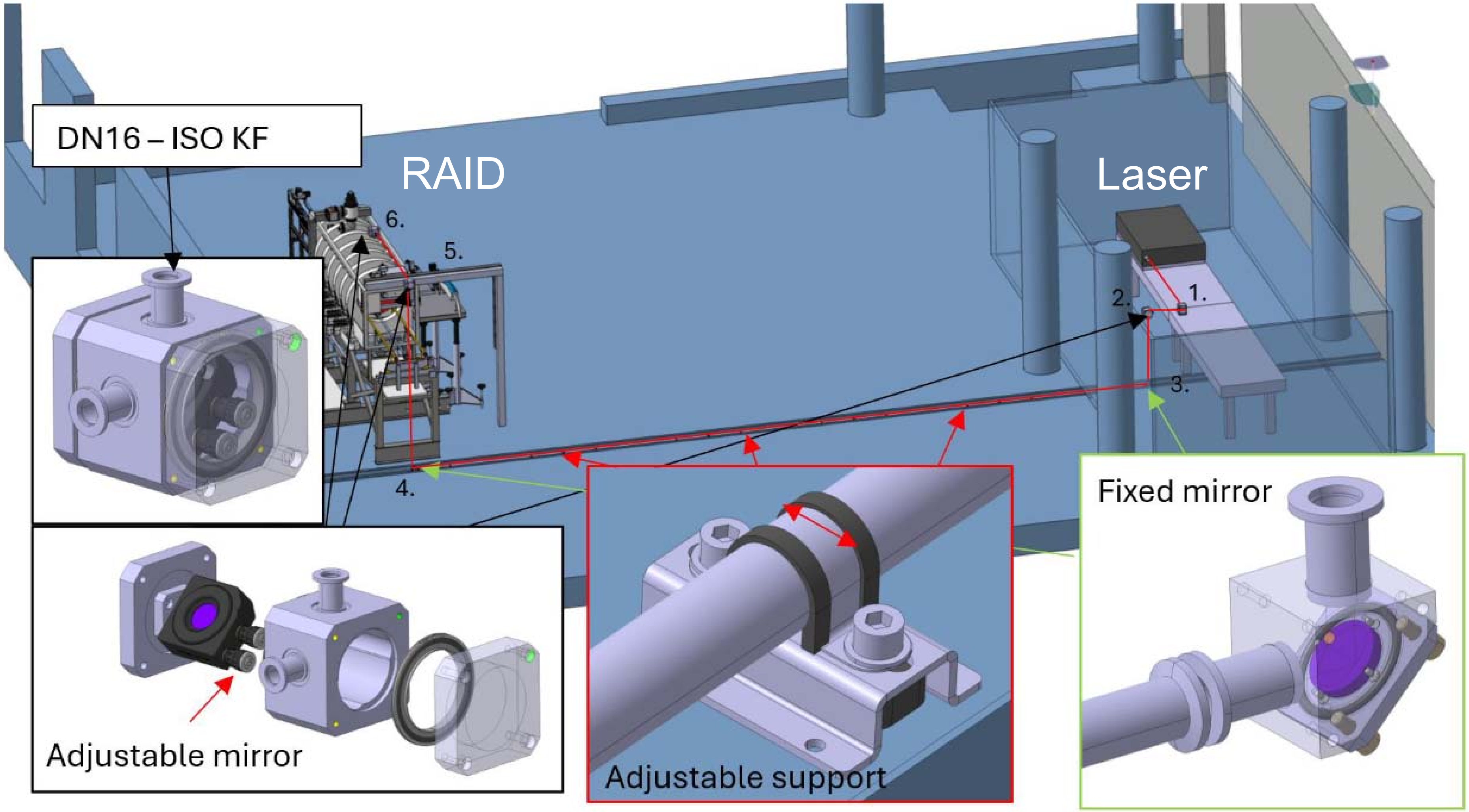

Implementation of beam path (T3)

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- Sealed beam path (designed for vacuum).
- Total length ≈ **19 m**
- Refractive elements minimized to reduce dispersion and avoid damage of optics.
- Beam is loosely focused. Waist size can be changed by means of a reflective telescope at entrance of beam path.

Laser at exit of beam path

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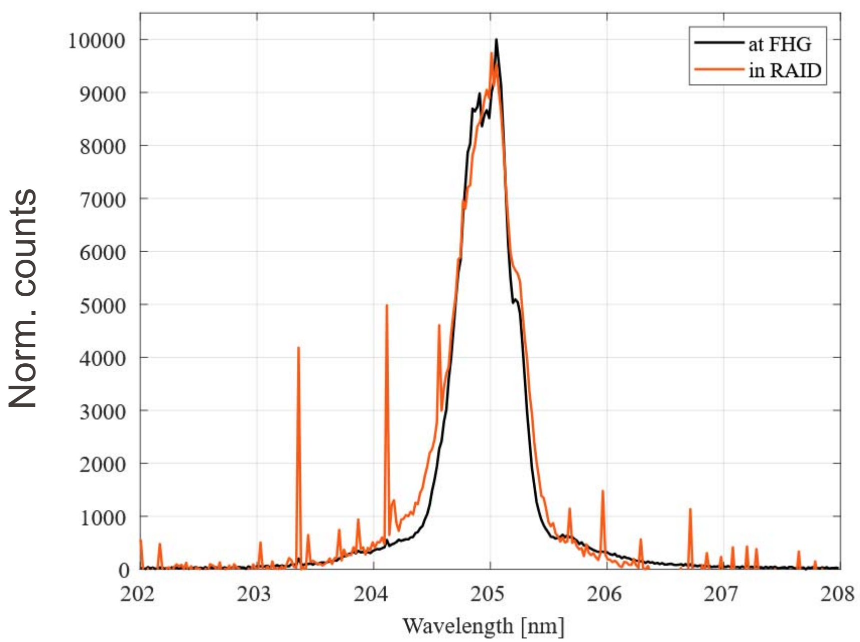
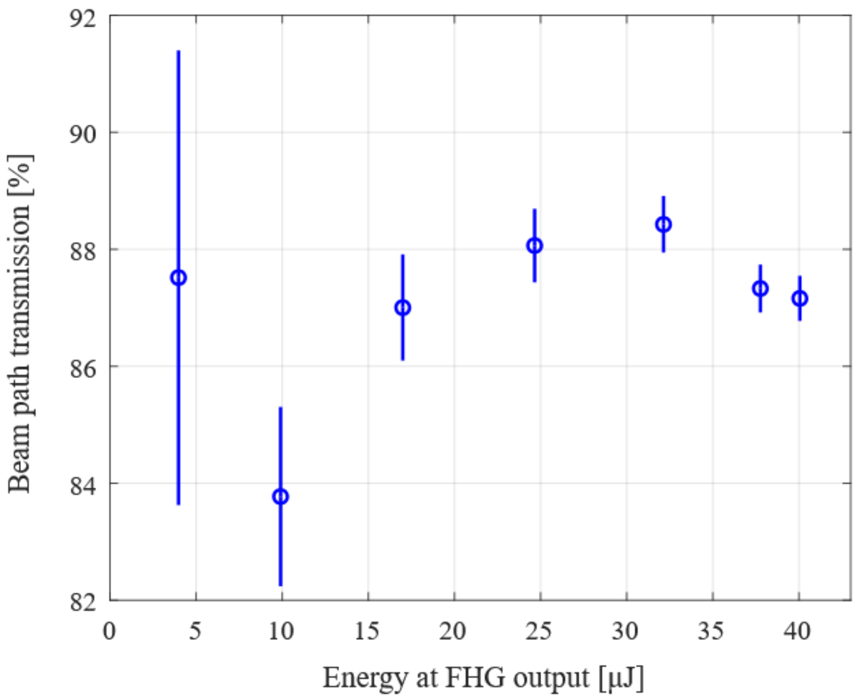
- Beam path is very efficient at transmitting the pulses and preserves their spectral features (temporal features will be studied in more detail).
- Refractive optics (windows, lenses, etc.) must be avoided as much as possible.



- Spectra measured at entrance and exit of beam path.



- Transmitted/input energy. Only ≈13% of the pulse energy is lost in beam path.

Laser inside RAID

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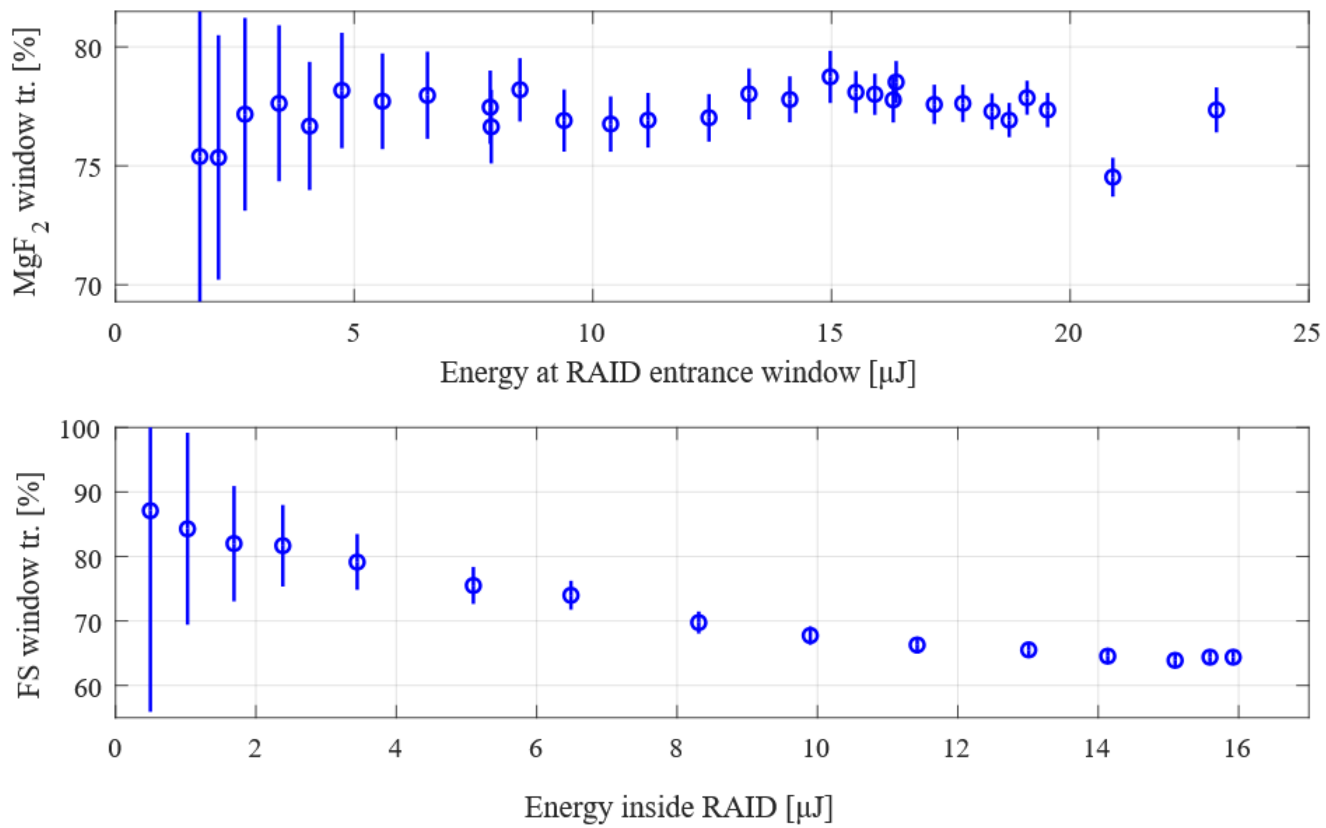
- Windows lead to losses.



- Energy measurements are made after FS window (60% efficiency at highest energies).
- Pointing of laser may vary by a few mm over a few hours. We think this is due mainly to heating of the RAID vacuum chamber.

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Detection of fluorescence (T2) ICCD camera

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- Detection system based on imaging optics, optical filtering and a very short gating time (down to 3ns) ICCD camera.

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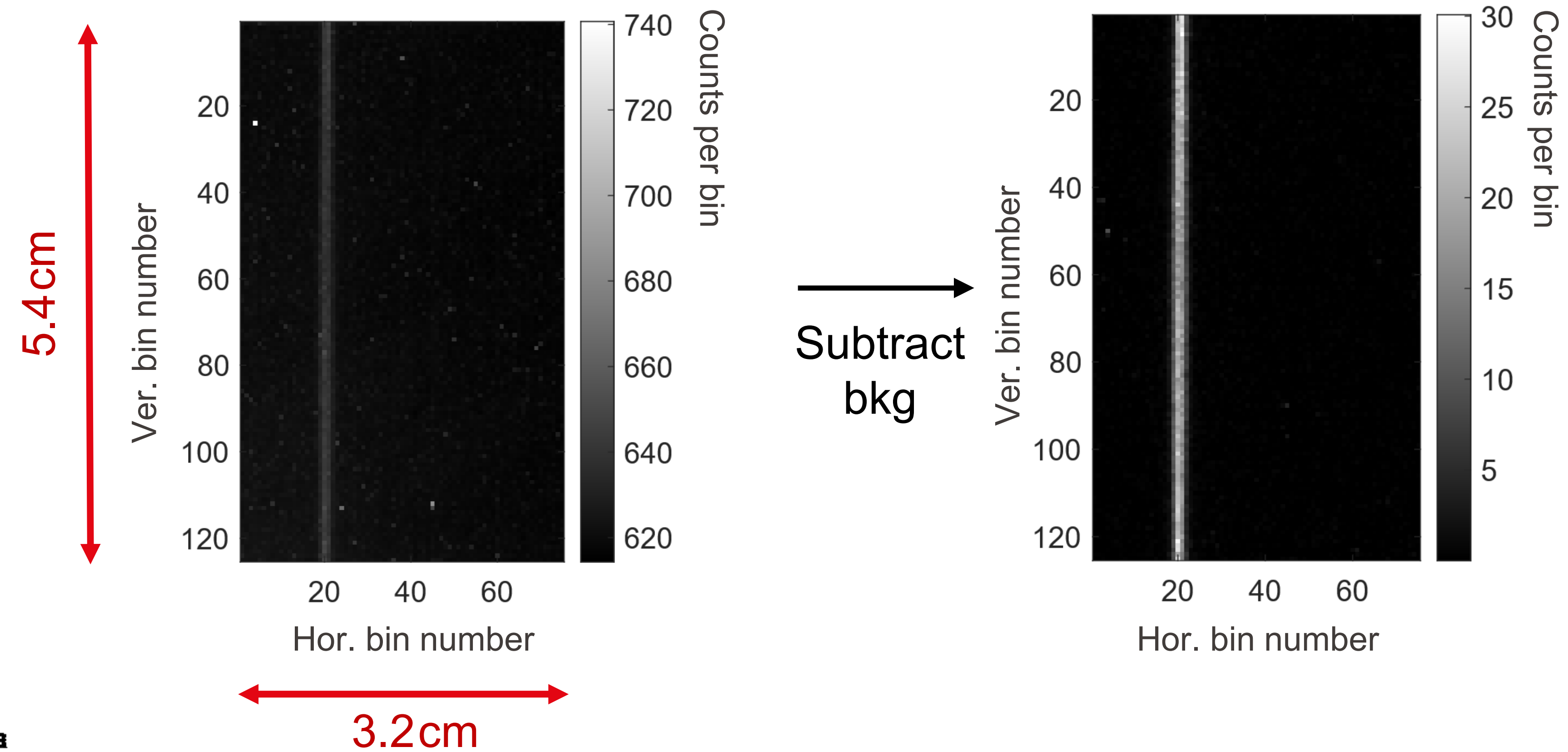
fs-TALIF in Kr gas
We obtain a signal with the laser tuned to 205.1 nm

- **No** plasma
- Gas fill pressure: 5.37 Pa
- Laser pulse energy: 10.3 µJ (measured; 17.1 µJ in RAID)
- 200 on-chip accumulations, 1000 frames averaged



- Notice that beam is collimated within field of view.
- Diameter of beam ($1/e^2$) is **≈ 3mm**
- Signal is very well resolved in 1D

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fs-TALIF in Kr gas

Signal has expected fluorescence decay rate and seems to follow quadratic scaling with Intensity

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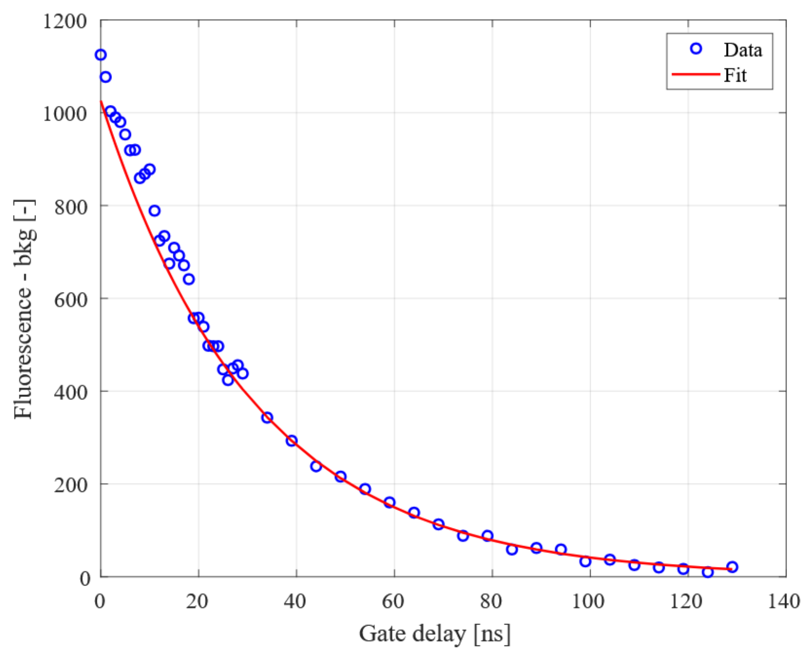

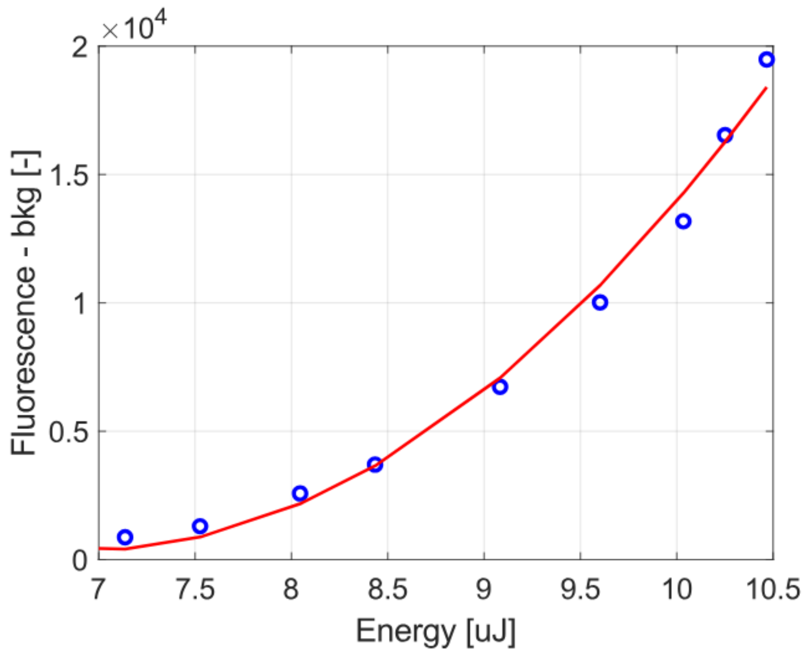

- Decay rate of fluorescence ≈31ns, close to expected value from literature (Kadi et al. *PPCF* (2024), Gazeli et al. *PoP* (2021)).
- Fluorescence scales quadratically with laser pulse energy starting from ≈ 6µJ (measured). That value includes the entire spectrum of the pulse.

fs-TALIF in H plasma

Clear signal with on-chip accumulation and frame averaging

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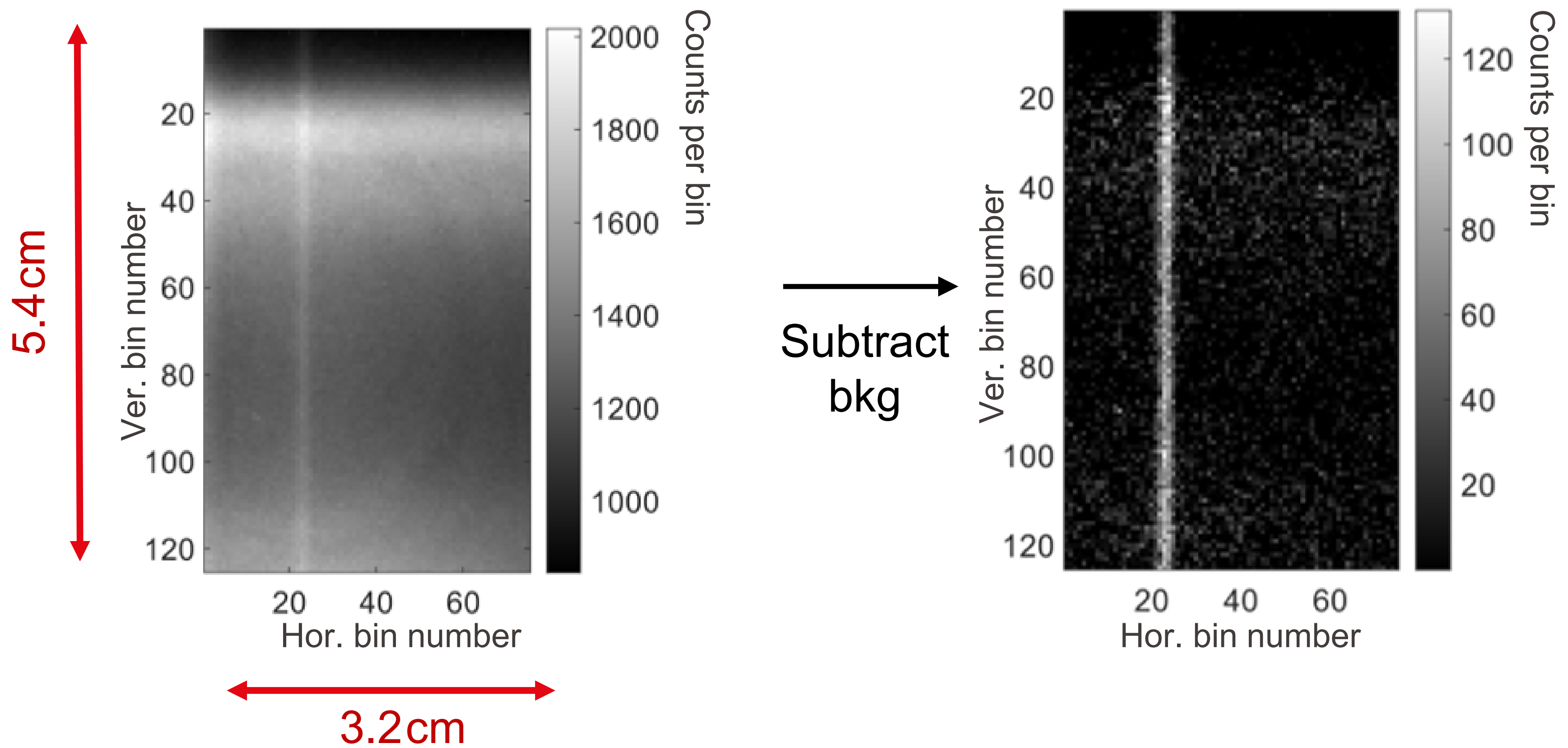
- H plasma, P_{RF} = 1x1kW, B_{field} = 260 G
- Gas fill pressure: 1.02-1.18 Pa
- Laser pulse energy: 10.3 µJ (measured; 17.1 µJ in RAID)
- 30 on-chip accumulations, 1000 frames averaged

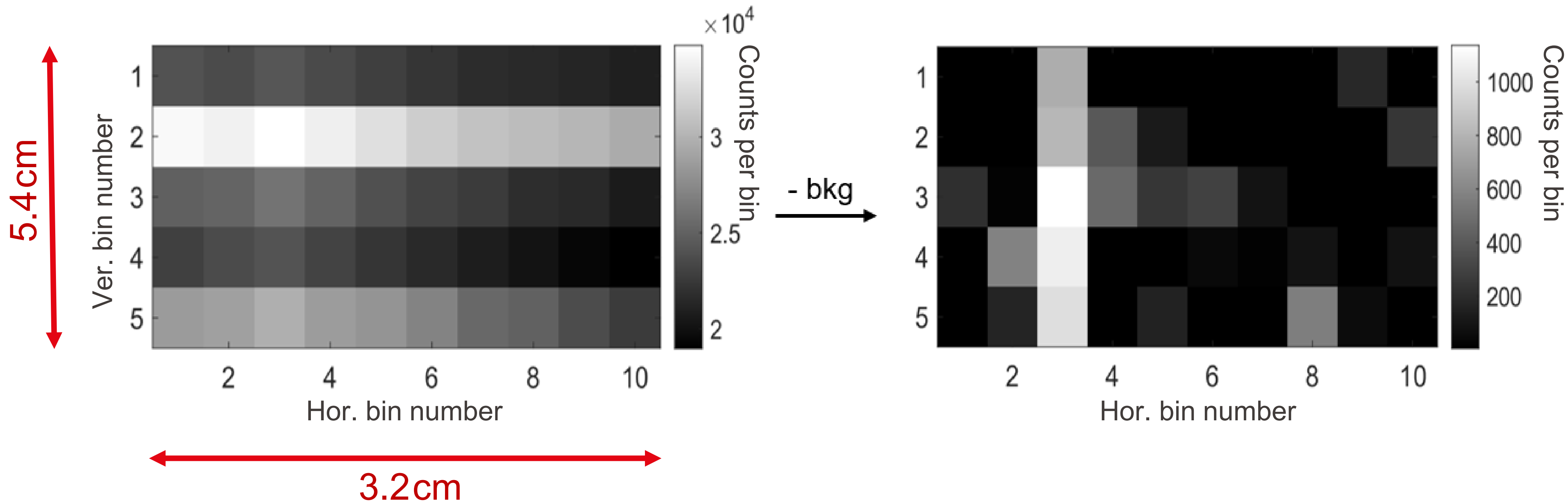


- Beam is collimated within field of view.
- Diameter of beam ($1/e^2$) is **≈ 3mm**
- Signal is very well resolved in 1D

fs-TALIF in H plasma

Faster acquisition with larger bins

- H plasma, P_{RF} = 1x1kW, B_{field} = 260 G
- Gas fill pressure: 1.02-1.18 Pa
- Laser pulse energy: 10.3 µJ (measured; 17.1 µJ in RAID)
- 10 on-chip accumulations, 100 frames averaged



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fs-TALIF in H plasma

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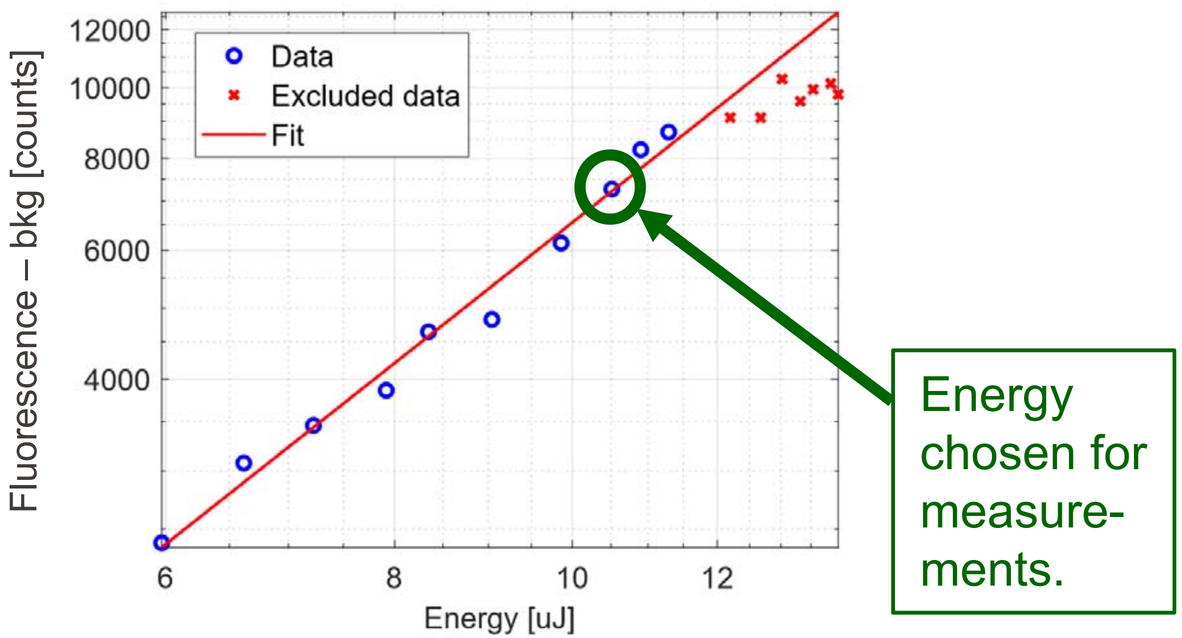
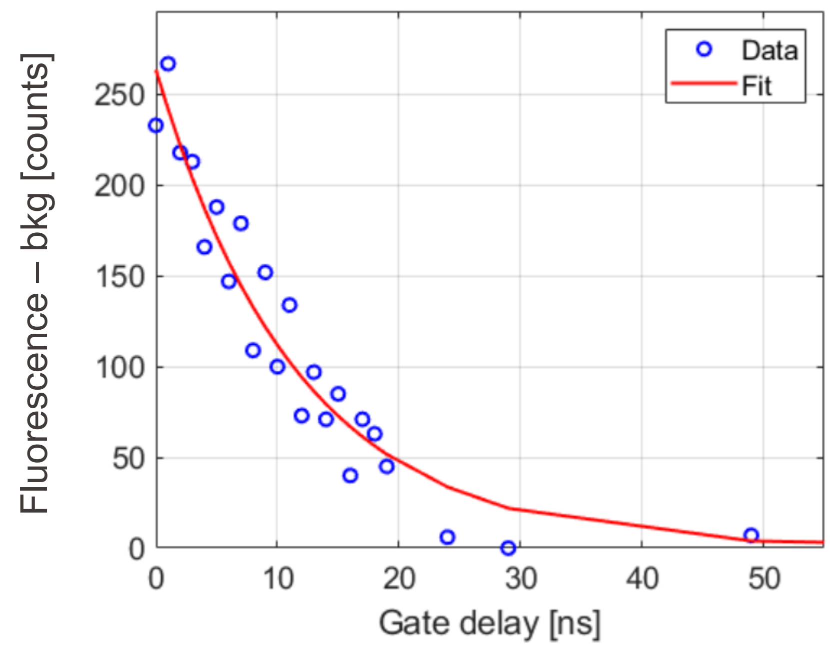
Beam waist chosen for maximum signal while staying in quadratic regime





- Fluorescence signal scales quadratically with pulse energy up to 12 µJ (measured; 20 µJ in RAID).

- Decay rate of fluorescence ≈ 10ns, close to expected value from literature (Kadi et al. *PPCF* (2024)).

fs-TALIF in D plasma

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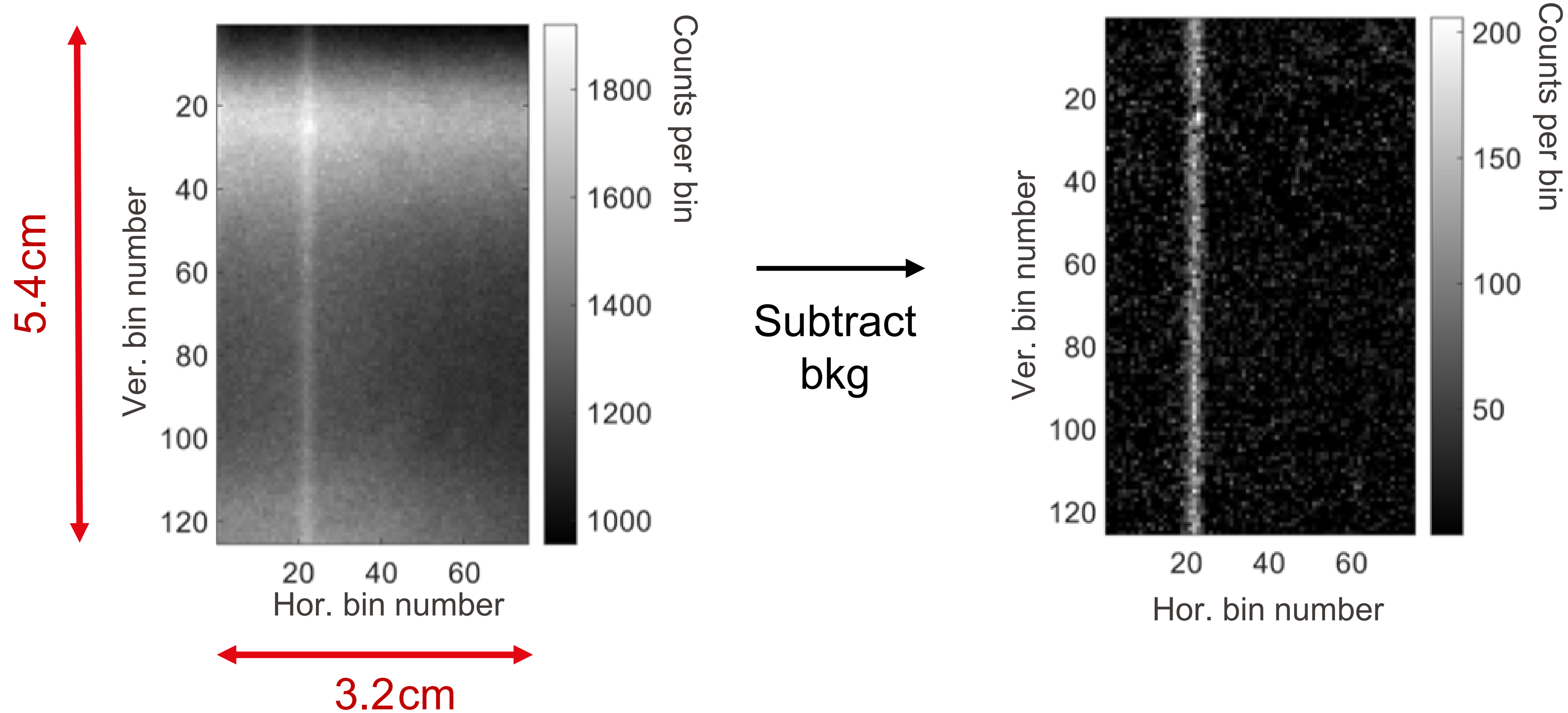
Results similar to H

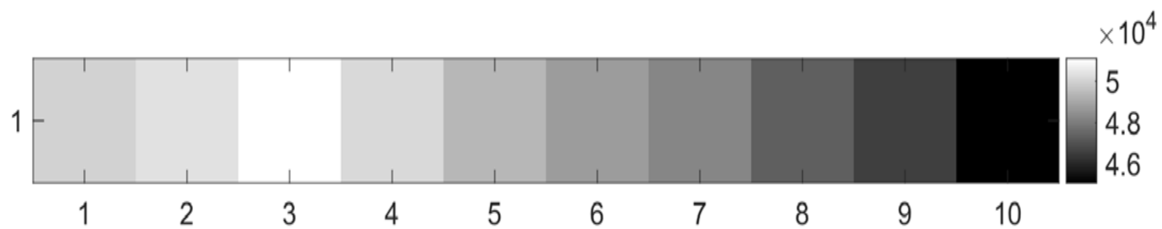
- D plasma, P_{RF} = 1x1kW, B_{field} = 260 G
- Gas fill pressure: 1.1 Pa
- Laser pulse energy: 10.3 µJ (measured; 17.1 µJ in RAID)
- 30 on-chip accumulations, 1000 frames averaged



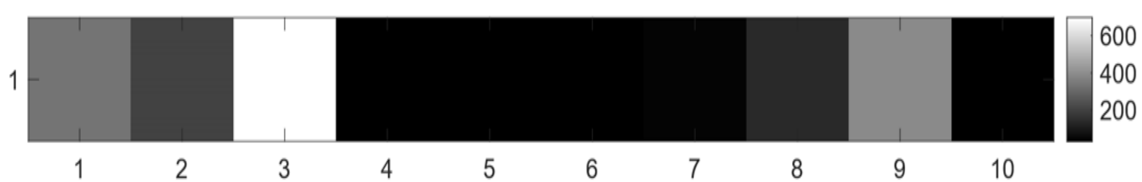
fs-TALIF in D plasma

With coarser binning we can do

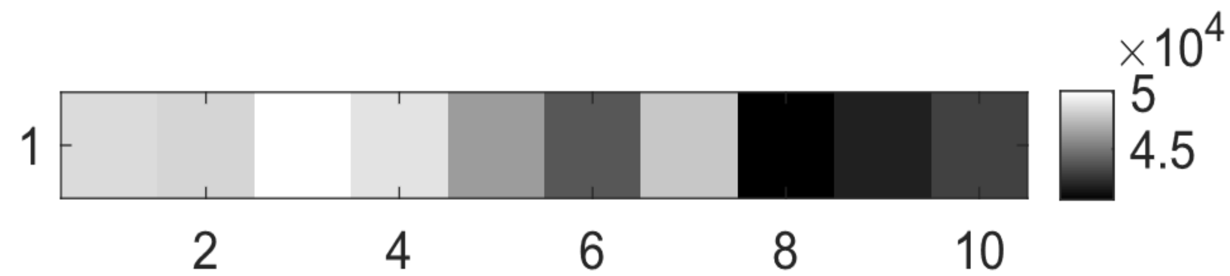
- D plasma, fill pressure: 0.58 Pa
- Full vertical bins
- 1 on-CCD accumulation, 100 frm. avg.



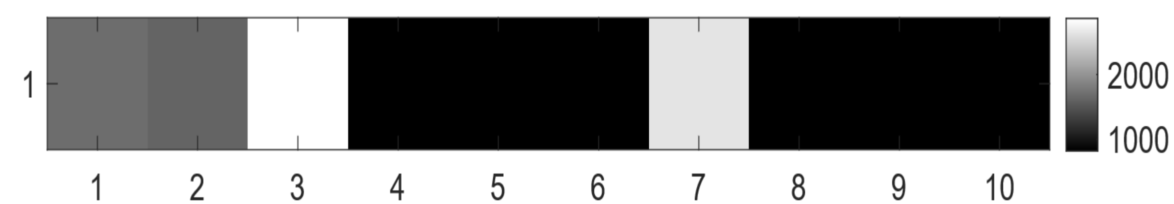
- bkg



- D plasma, fill pressure: 0.58 Pa
- Full vertical bins
- 1 on-CCD accumulation, **no** frm. avg.



- bkg



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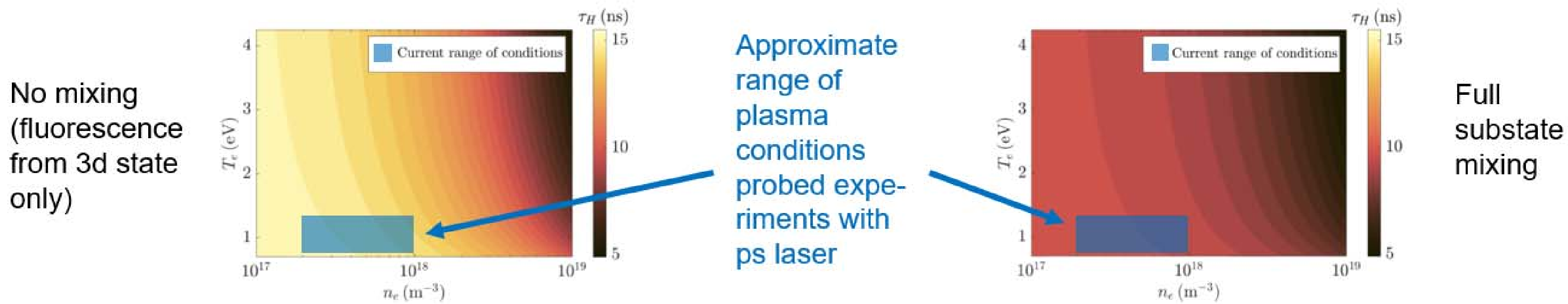
Studies of n=3 substate mixing (T5)

- Simulations of H fluorescence lifetimes suggest that electron collisional processes are not sufficient to explain observations made in (separate) experiments with our ps laser (Kadi et al. *PPCF* (2024)).



- If one includes full substate mixing of n=3 (i.e. all m levels are quickly populated statistically upon laser excitation), lifetimes become compatible with observations.

- If indeed present, full substate mixing makes value of branching ratio very robust up until $n_e \approx 3 \times 10^{18}\ \mathrm{m}^{-3}$.
  - Furthermore, the value of the branching ratio changes. Its impact in the calculation of n_{H} leads to an increase of ≈26% in the density.

- Further theoretical and experimental studies are required.

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Theoretical studies of laser pulse absorption (T5)

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- Even though widely used, the model for two-photon absorption given in introduction may have limitations in fs regime:
  - Pulse intensities are very large
  - Rate equations may neglect important quantum effects
  - Laser photon statistics may differ compared to ps and ns cases
- Using a quantum model (based on Bloch equations) we have established that model does apply for laser intensities up to (at least) a few GW/cm^2 .
- From literature, laser photon statistics factor may have a different value.
- Further theoretical studies are being pursued.
- Experimental studies with pulse compressor will allow us to study effects of pulse duration.

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Conference contributions

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Authors	Conference name	Title
S.P.H. Vincent, et al.	67th Annual Meeting of the APS Division of Plasma Physics (2025)	Poster: Femtosecond TALIF for H density measurements
I. Furno, et al.	30th IAEA Fusion Energy Conference (2025)	Poster: Active spectroscopy for atomic H and D measurements in fusion
M. Baquero-Ruiz, et al.	21st International Symposium on Laser-Aided Plasma Diagnostics (2025)	Poster: Femtosecond TALIF of atomic H in RAID
M. Goddijn, et al.	6th European Conference on Plasma Diagnostics (2025)	Poster: Femtosecond TALIF diagnostic on the RAID linear device
M. Goddijn, et al.	Joint Annual Meeting of the Austrian ahd Swiss Physical Societies (2025)	Oral: Femtosecond TALIF in fusion-relevant hydrogen plasmas

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Conclusions

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- We have developed a fs TALIF system to study H (and/or D) densities in RAID
  - Laser pulses are stable in energy and spectral features.
  - Frequent monitoring may nevertheless be required in applications.
- Using averaging in detection, we obtain clear fs-TALIF signals resolved in 1D
  - Laser beam is collimated over field of view.
  - Successful observations in H plasma
  - Successful observations in D plasma
  - Successful observations in Kr gas. Calibrations seem to be possible without changing wavelength.
- Very coarse binning allows us to run fast
  - Nonetheless, single laser pulse measurements are **not yet possible**.
  - We believe that we are at the limit of what we can achieve with the ICCD.

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Outlook

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- We will pursue the measurements of n_{H}
  - We believe that the results presented here show very good potential.
- We will install a streak camera
  - Resolving the background immediately prior to the laser pulse may allow us to better distinguish signal over noise.
  - Camera and optics are ready. Experiments are planned for early 2026.
- It may be possible to increase pulse energies while having the spectrum centered at 205.1 nm
  - With system tuned closer to 206 nm we obtain at least twice the energy.
  - We think that thicker delay crystals in FHG may allow us to obtain this at 205.1 nm.
- Studies of full substate mixing and pulse absorption will continue
  - Installation of pulse compressor and experiments will be interesting to study effect of pulse duration.

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Thank you for your attention