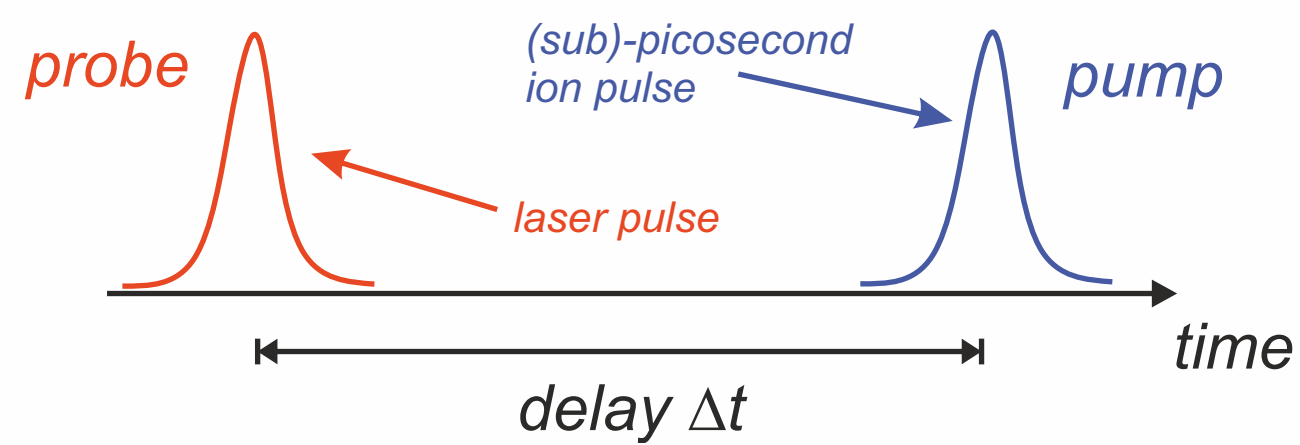


## Motivation

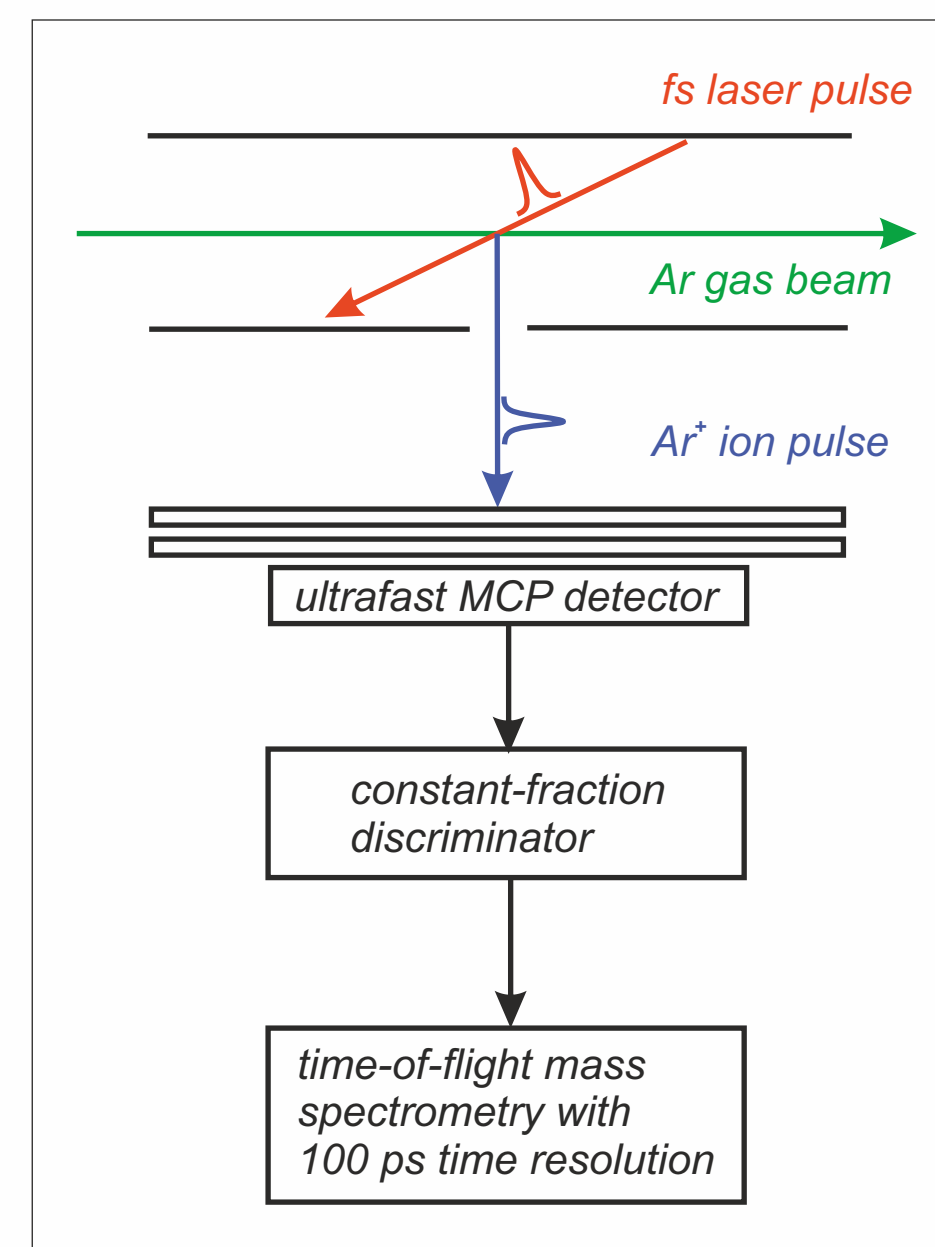
- generation of highly localized non-equilibrium states via ion impact onto a surface
- observation of dynamics on a (sub-) picosecond timescale via pump-probe technique



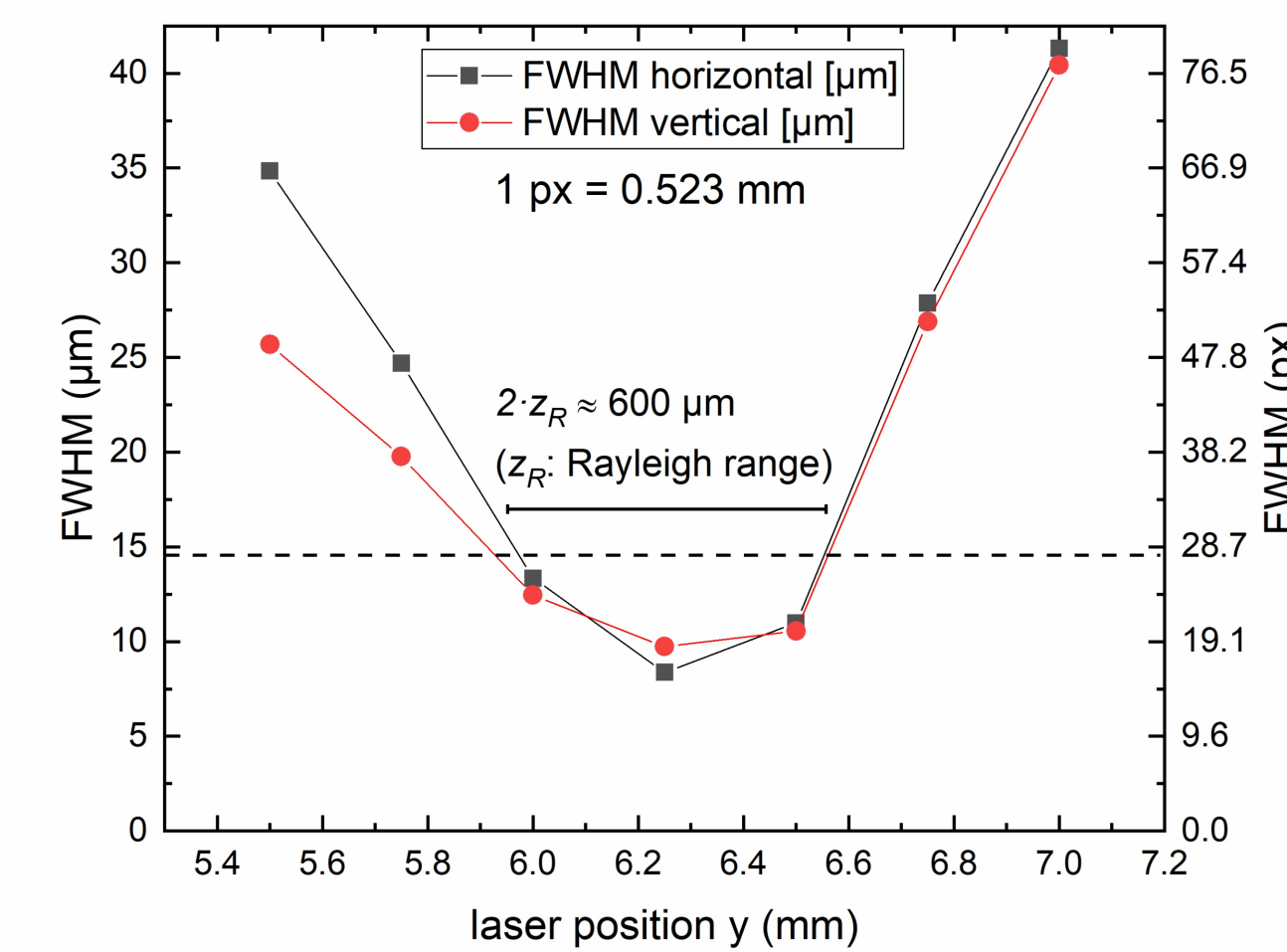
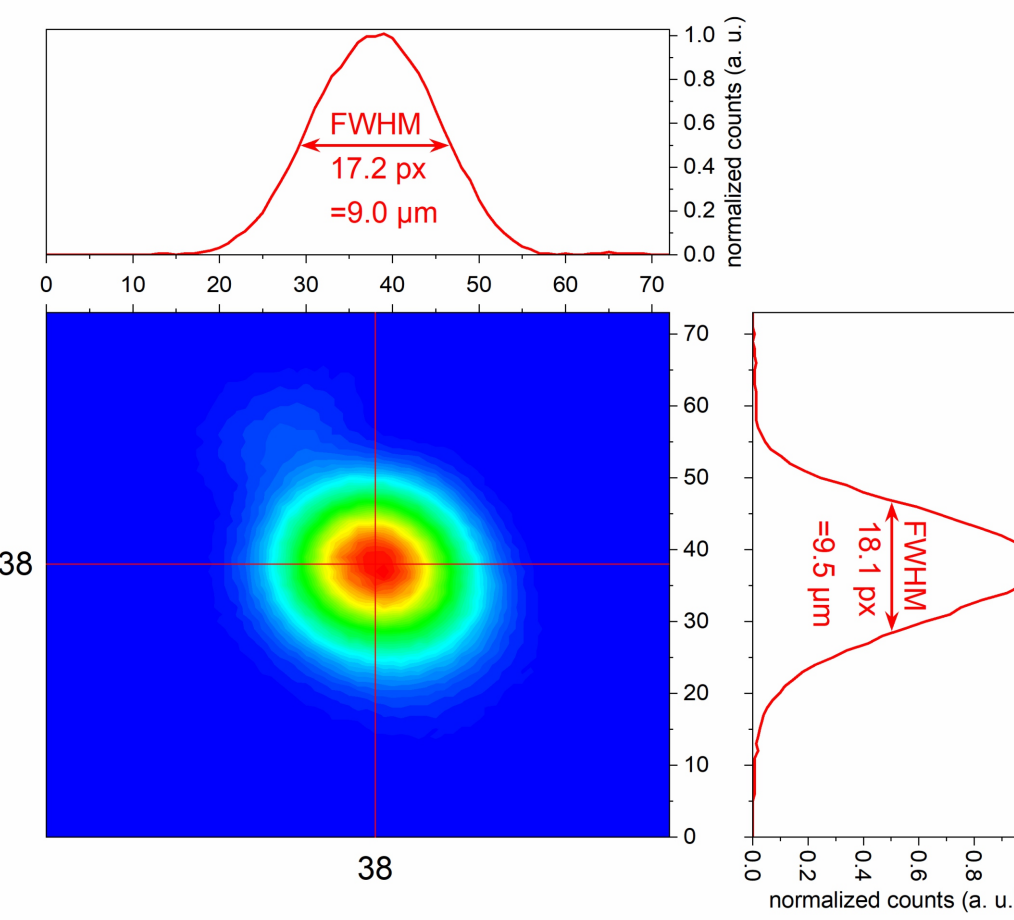
- generation of ultra-short ion pulses: difficult task due to space-charge broadening and velocity distribution of ions at a given temperature

⇒ supersonic gas expansion - control over number density and temperature distribution

- characterization of such a supersonic gas expansion of Argon via Excimer laser



## Characterization of fs laser system via CCD camera



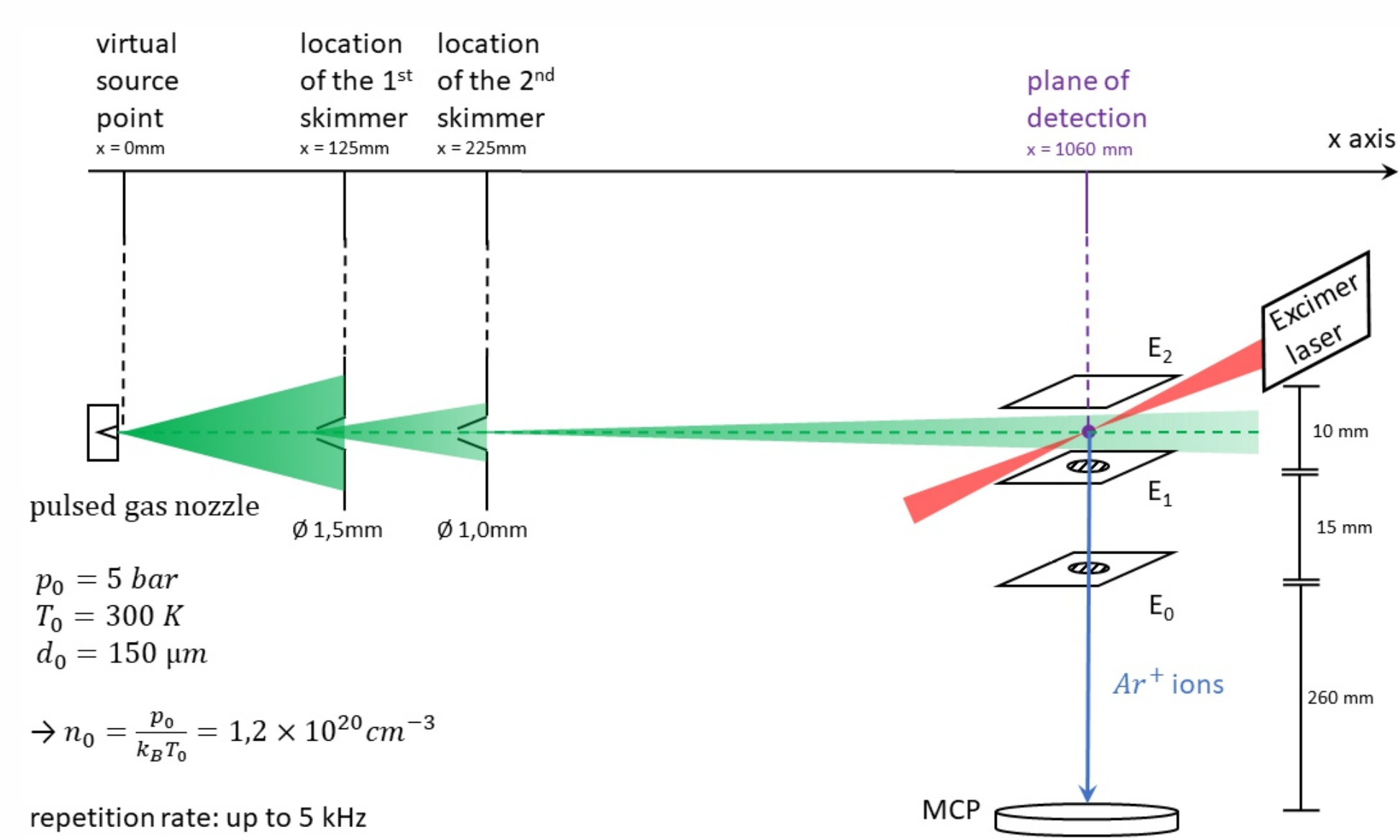
beam properties:

800 nm  
50 fs  
1.5 mJ  
1 kHz

- nearly symmetrical laser spot profile with diameter < 10 μm and Rayleigh range of  $z_R \approx 300 \mu\text{m}$
- measured spot profile leads to a peak intensity  $I_0 \approx 4.0 \times 10^{16} \text{ W/cm}^2$

⇒ intensity allows in principle to create  $\text{Ar}^{q+}$  with charge state  $q = 1, \dots, 6$

## Experimental setup



beam properties:

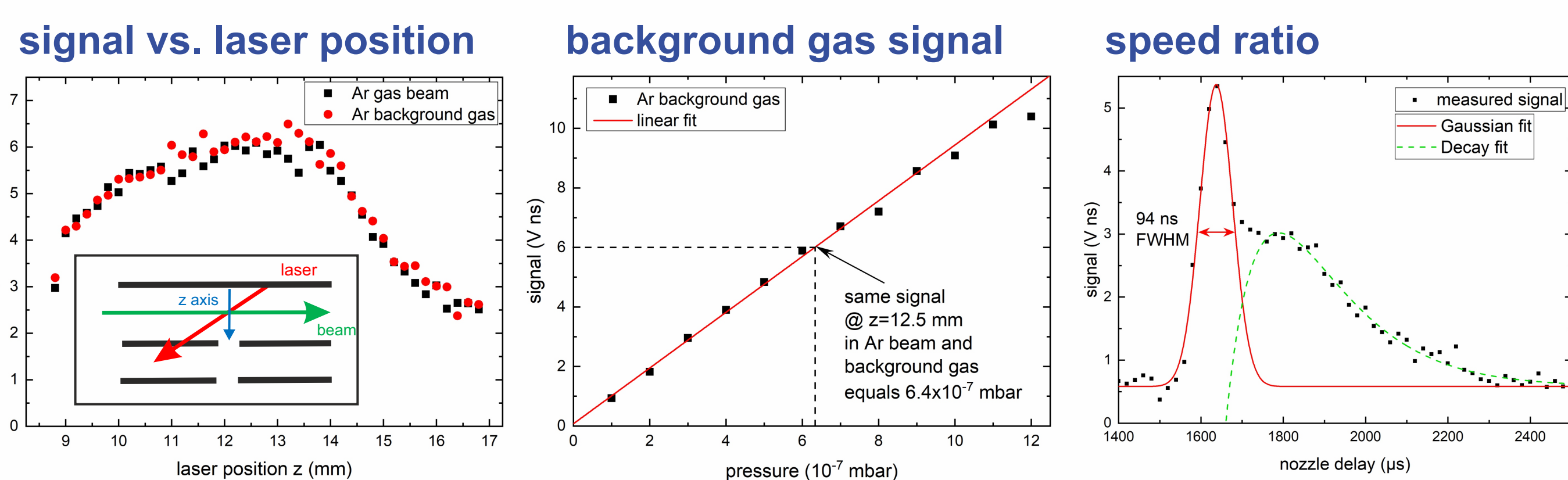
157 nm  
5 ns  
3.0 mJ  
up to 200 Hz

exit profile:  
3 mm x 6 mm

lens focus:  
384 mm

focal profile:  
400 μm x 800 μm

## Measurement of beam density



- measured ion pulses generated by **laser induced photoelectrons** in ionization volume!

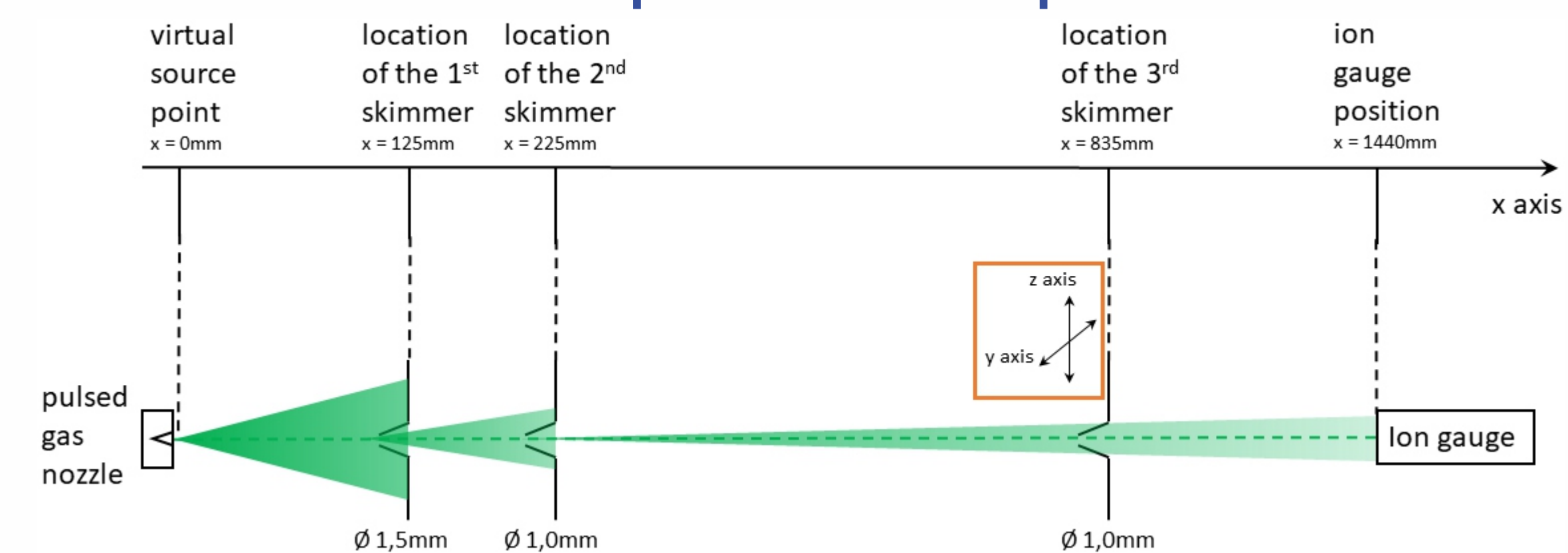
flight time yields a terminal parallel velocity of  $v_{||,\infty} = \frac{\Delta x}{\Delta t} = \frac{1060 \text{ mm}}{1640 \mu\text{s}} = 646 \frac{\text{m}}{\text{s}}$

FWHM of 94 μs yields a (deconvoluted) standard deviation of  $\sigma_t = 37 \mu\text{s}$

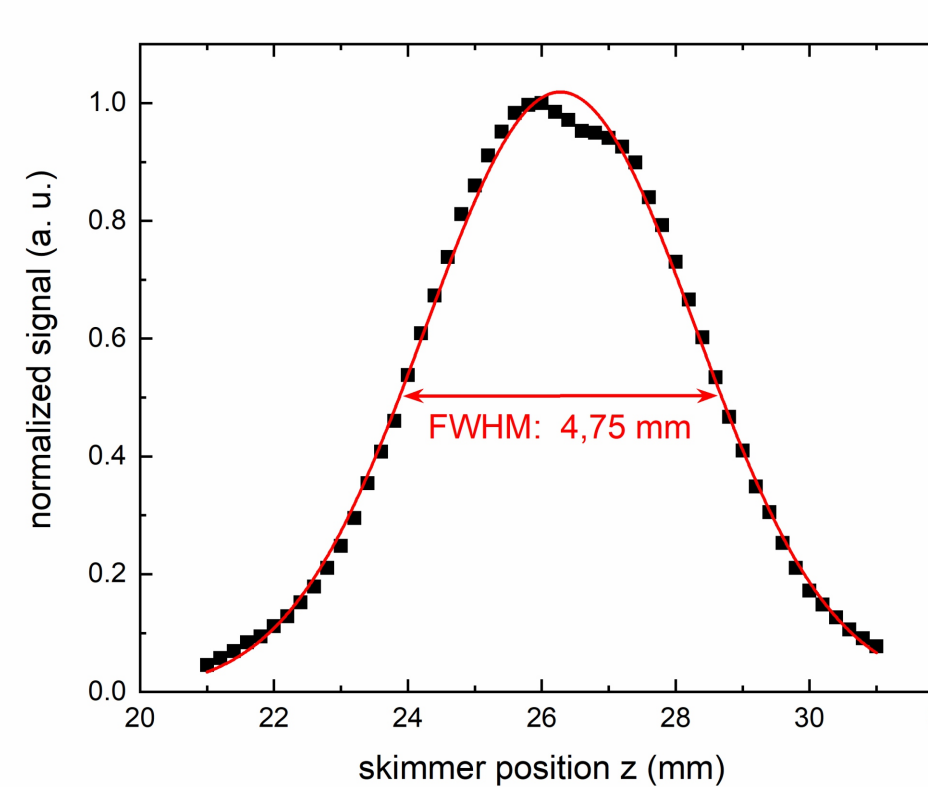
⇒ resulting speed ratio of  $S = 44$  leads to parallel temperature of  $T_{||,\infty} = 512 \text{ mK}$

⇒ combines to a spatial standard deviation of  $\sigma_x = 24 \text{ cm}$  along flight direction

## Measurement of beam profile and temperature distribution



## Beam profile and temperature distribution



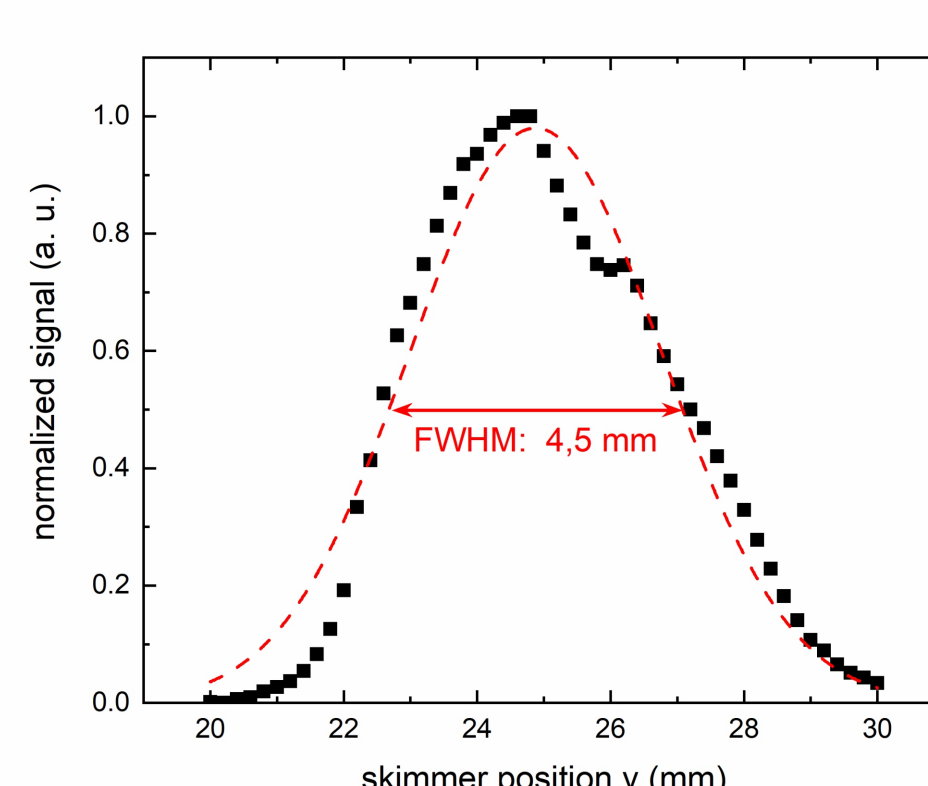
allows to quantify the center beam density with condition

$$N_{\text{Bulk}} = N_{\text{Beam}} = \iiint \rho_0 f_x(x) f_y(y) f_z(z) dV,$$

$$f_x(x) \cong 1, f_y(y) = e^{-\frac{y^2}{2\sigma_y^2}}, f_z(z) = e^{-\frac{z^2}{2\sigma_z^2}}$$

yields a center line density of  $\rho_0 = (6.0 \pm 0.12) \times 10^{10} \text{ cm}^{-3}$

difference to theoretical prediction of an order of magnitude due to the presence of skimmers blocking the central intensity of the beam



estimation of the velocity distribution perpendicular to the beam according to

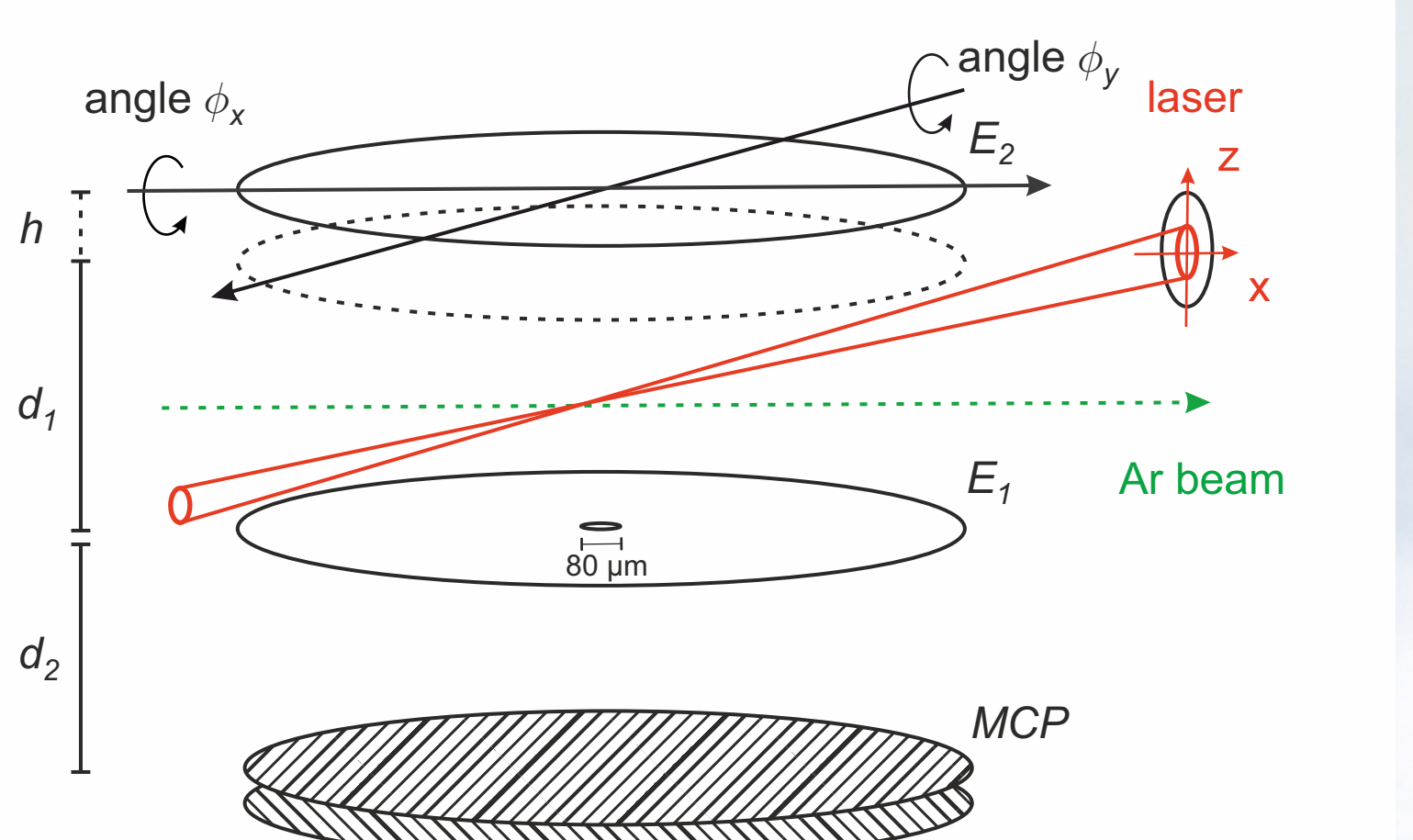
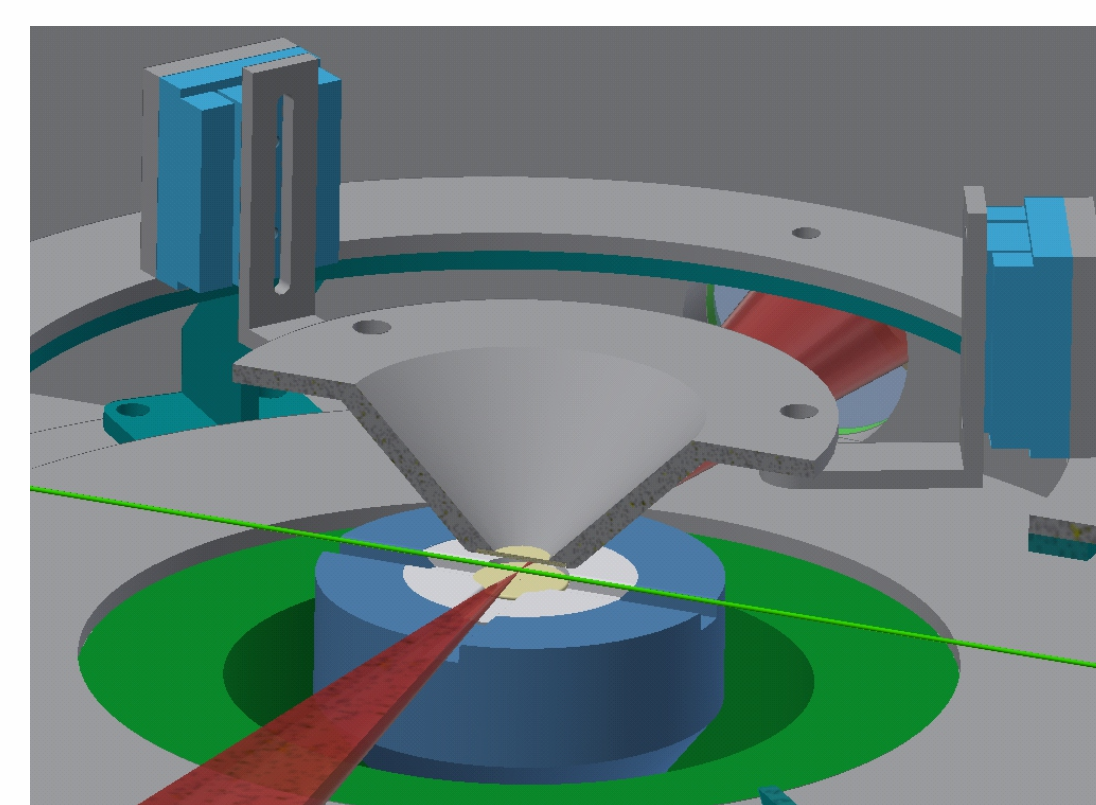
$$e^{-\frac{mv_{\perp}^2}{2k_B T_{\perp}}} = e^{-\frac{z^2}{2\sigma_z^2}} \text{ with the geometric relation } \frac{v_{\perp}}{v_{||,\infty}} = \frac{z}{\Delta x}$$

for laser with an estimated Rayleigh range of  $\approx 400 \mu\text{m}$  and distance  $\Delta x = 380 \text{ mm}$  between skimmer 3 and plane of detection:  $v_{\perp} \leq 1 \text{ m/s} \rightarrow T_{\perp} \leq 2.3 \text{ mK}$

upper limit of 10 mK in simulations already leads to a time resolution of sub-picoseconds → unproblematic

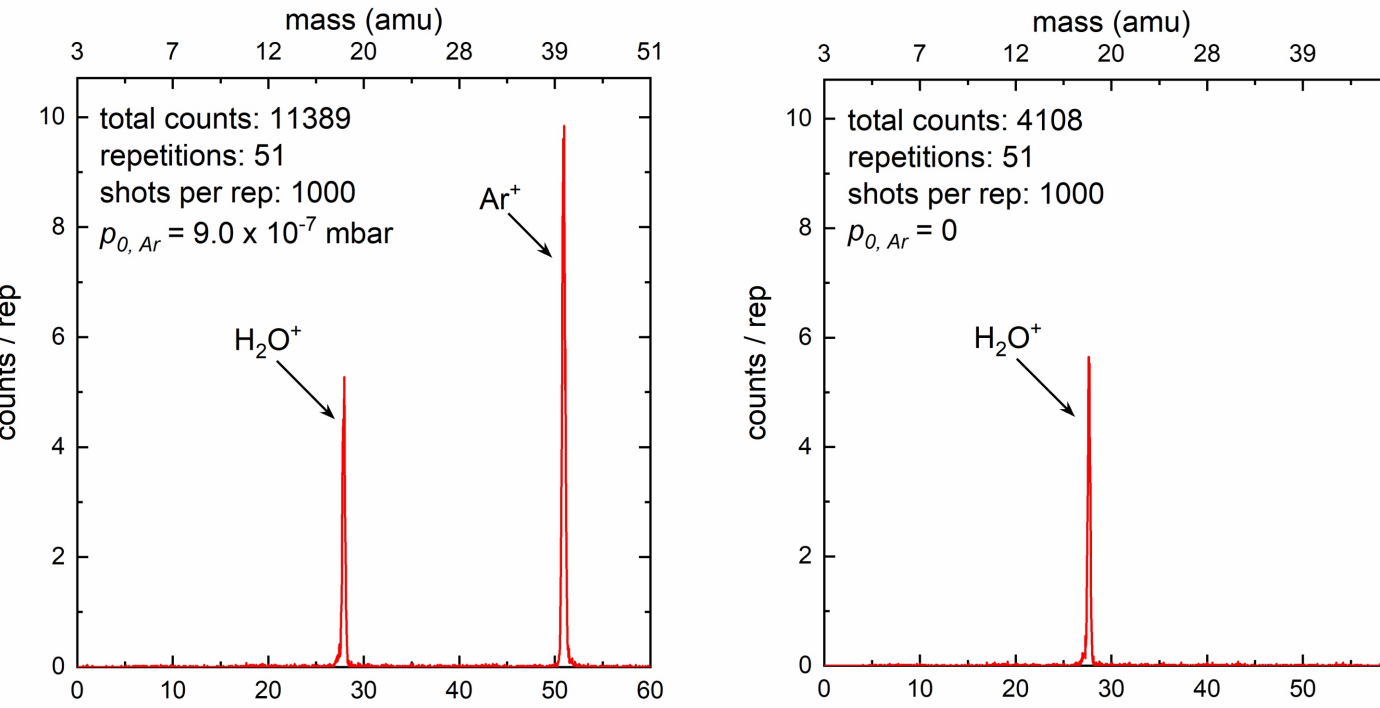
## Characterization of geometry: Argon background gas

### Detection geometry

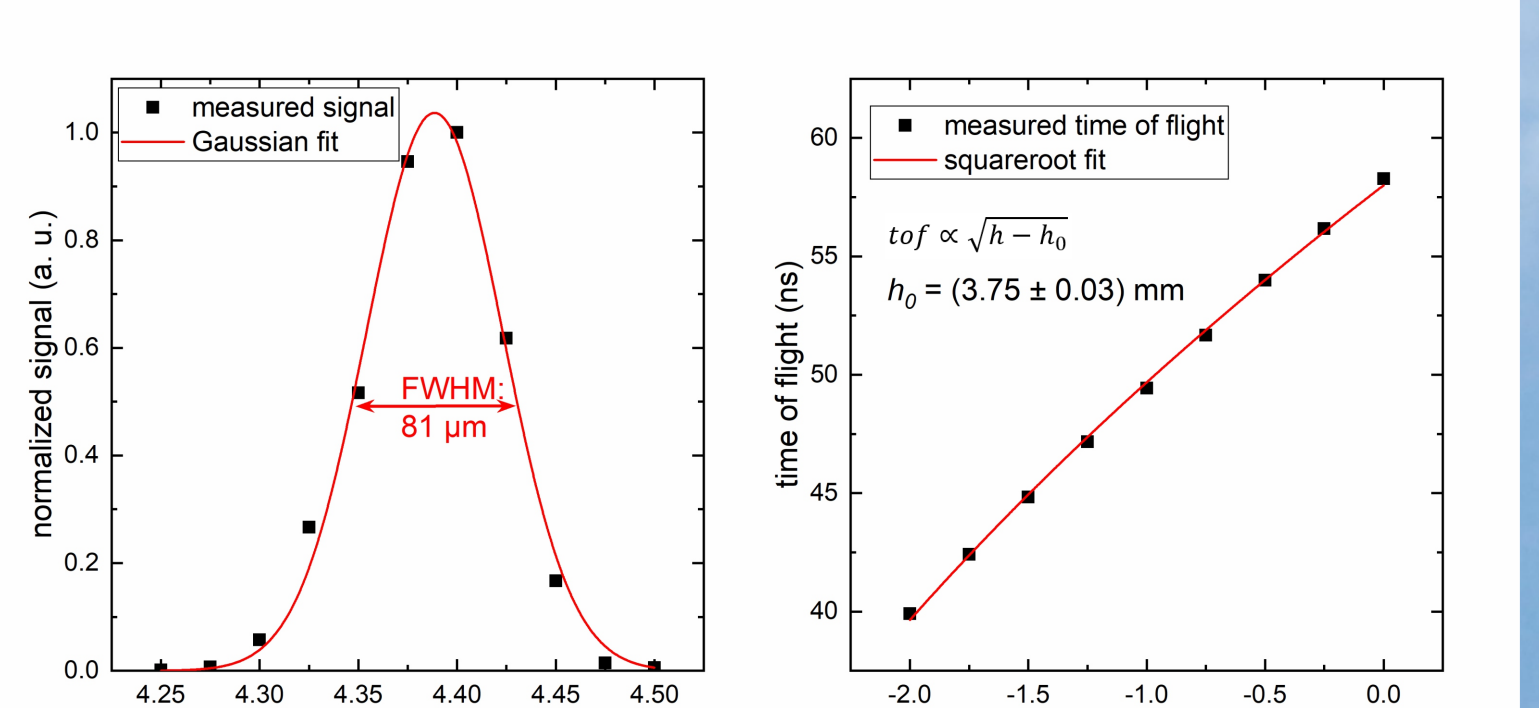


- 3 piezo motors allow to move the upper electrode twofold: absolute height  $h$  and angular tilt  $\phi_{x,y}$
- vacuum chamber filled with Argon background gas at  $p_0 = 9.0 \times 10^{-7} \text{ mbar}$

### Time-of-Flight mass spectra



### geometric boundaries

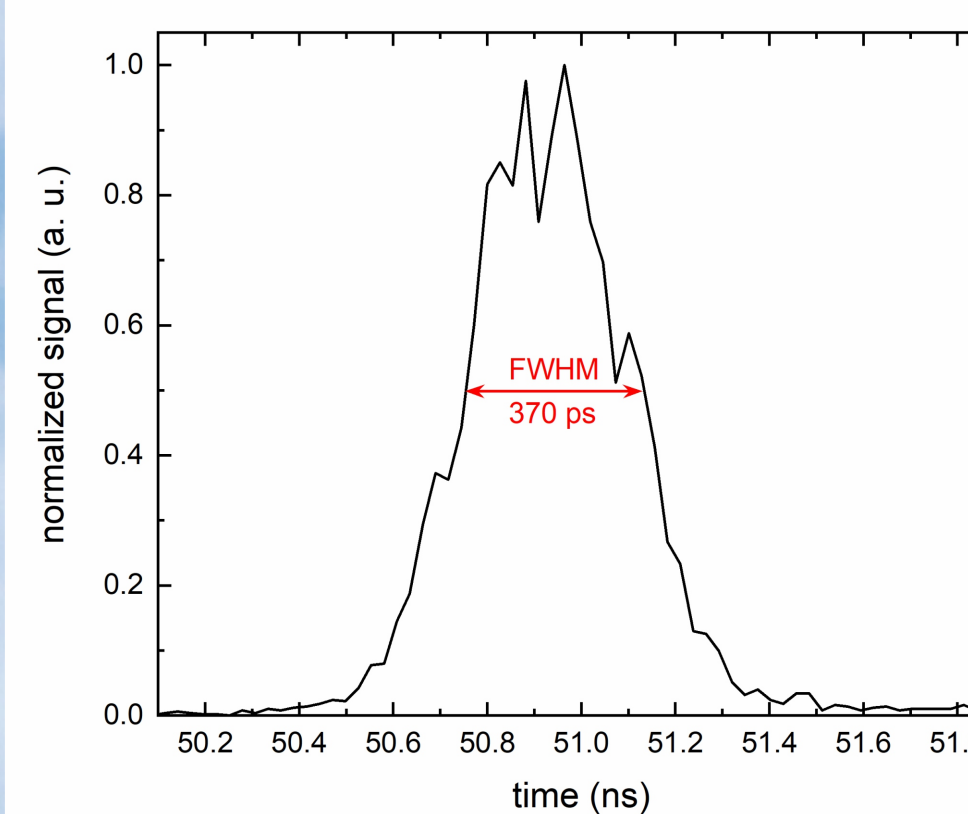


- detection of ionized Argon via Time-of-Flight mass spectrometry (ToF)
- allows to quantify absolute flight time, pulse height and pulse width for each mass species

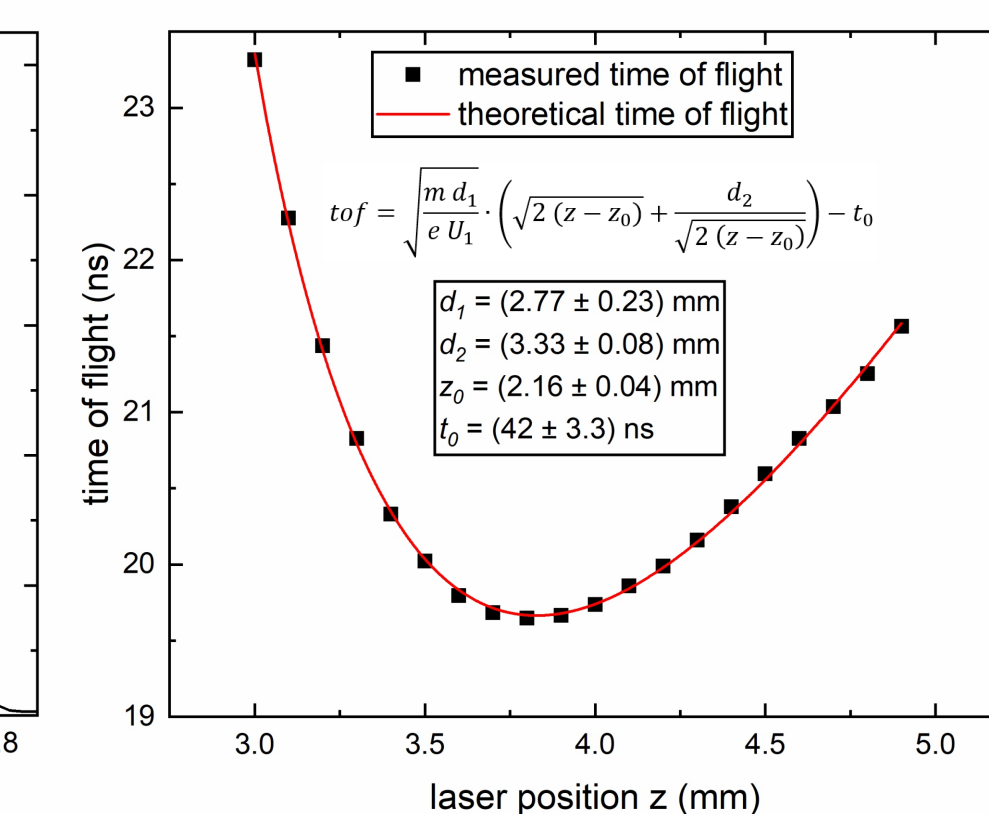
- x-scan of laser allows to image the transmitting hole in electrode  $E_1$
- time of flight as function of height  $h$  of electrode  $E_2$  yields starting height  $h_0 = 3.75 \text{ mm}$

## Time-of-Flight focus and sub-400 ps ion pulses

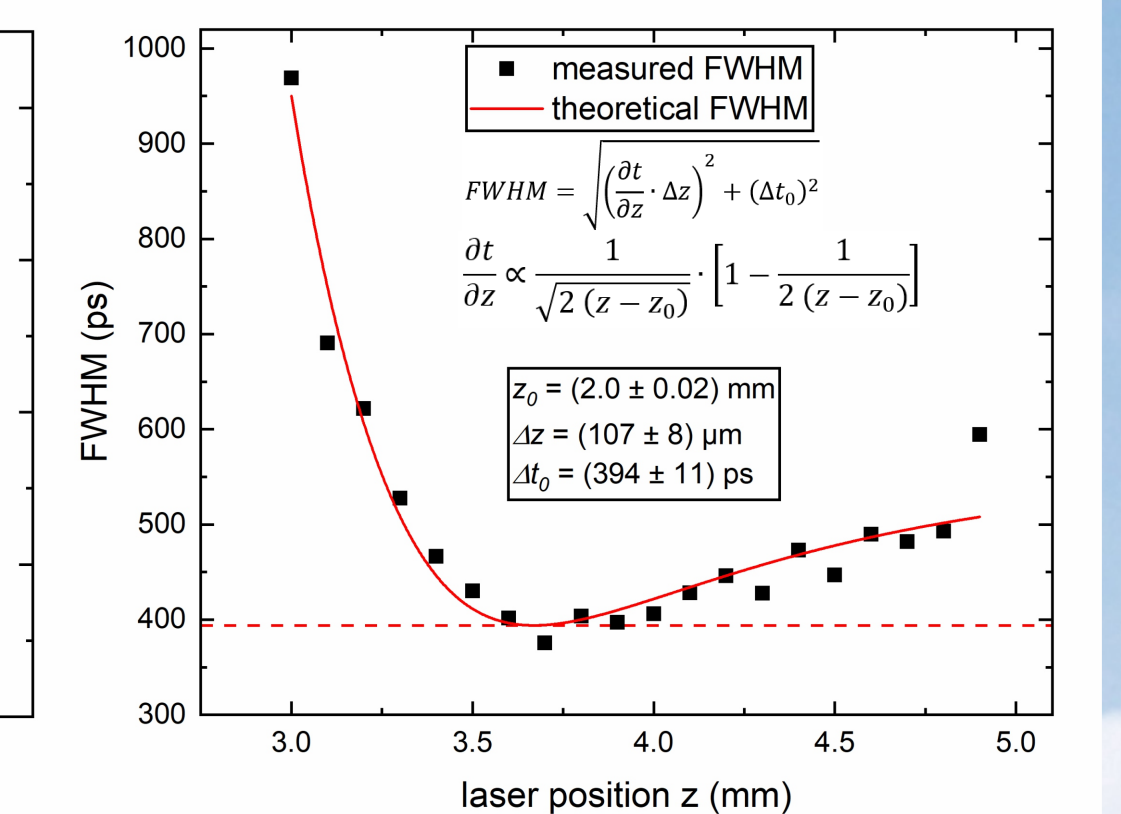
### pulse shape



### ToF vs. laser position



### FWHM vs. laser position



- the shortest pulse duration yields a FWHM of **370 ps** for the  $\text{Ar}^+$  pulse of the bulk gas
- time of flight as a function of laser position matches the theoretically expected behaviour  
⇒ allows to extract distances  $d_1$ ,  $d_2$  and  $z_0$  in geometric arrangement
- FWHM of Argon ion pulse is minimized at the Time-of-Flight minimum →  $\Delta t_{\text{min}} \approx 0.25 \text{ ps}$   
⇒ offset  $\Delta t_0$  must originate from:
  - thermal starting velocity spread →  $\approx 200 \text{ ps}$  at  $T = 300 \text{ K}$
  - temporal measurement jitter / detector time resolution
- FWHM allows to estimate the starting width  $\Delta z \approx 100 \mu\text{m}$  → expanded laser focus

## Current status

- distance  $d_1$  can be reduced without altering detection characteristic
- adjusting the laser focus to diameter < 10 μm  
⇒ reduces the behaviour of the FWHM by an order of magnitude  
⇒ produces charge states for  $\text{Ar}^{q+}$  for  $q > 1$  in mass spectra
- crossing the gas and laser beam for ion signal from gas beam  
⇒ reducing thermal broadening to shorten pulse width even further!

