Laser scattering techniques applied to cold atmospheric plasmas : trends and pitfalls

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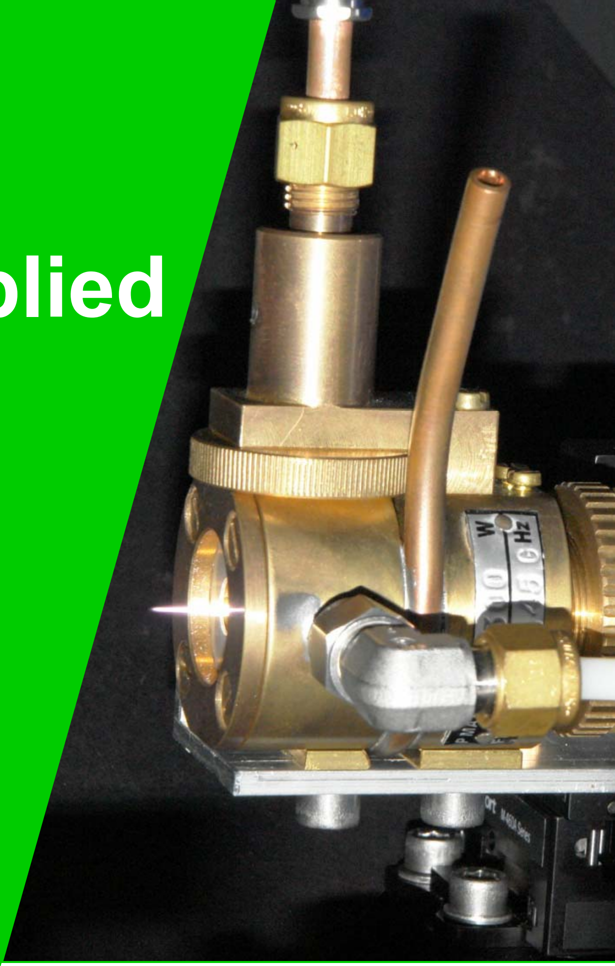
Laser scattering techniques applied to cold atmospheric plasmas: trends and pitfalls

J M Palomares, E A D Carbone, S Hübner, A F H van Gessel
and J J A M van der Mullen



EPG

TU/e Technische Universiteit Eindhoven University of Technology

Discharges working in open air

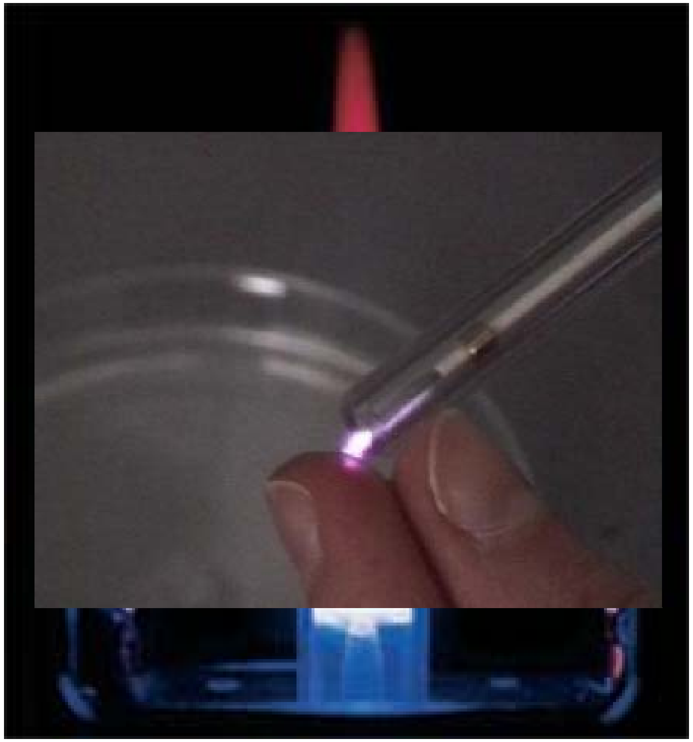
DBD, Arc, Jets, ICP, TIA…

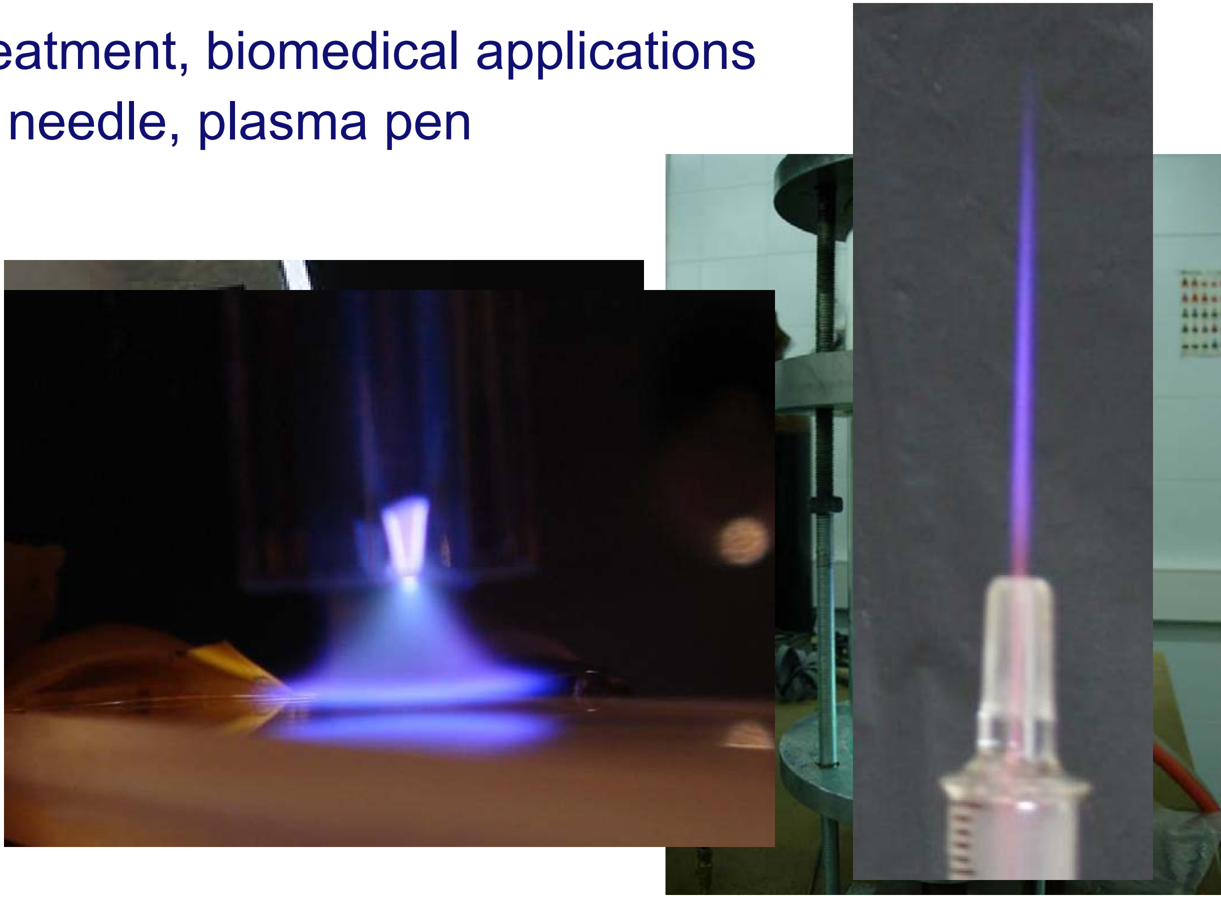
Cool Atmospheric Plasmas (CAPs)

Easy and safe to operate

Applications: surface treatment, biomedical applications

Micro-plasmas, plasma needle, plasma pen





Cold atmospheric plasmas (CAPs)

Deviations from equilibrium

- $T_e/T_g \sim 50$
- Filamentary micro plasmas
- Strong gradients
- Environment perturbations

Common OES not applicable

Laser scattering techniques:

- Thomson scattering → T_e n_e
- Rayleigh scattering → T_g
- Raman scattering → T_{rot} molecule concentrations
- Spatial and temporal resolution

Outline

Introduction to CAPs

Laser scattering on surfatron torch

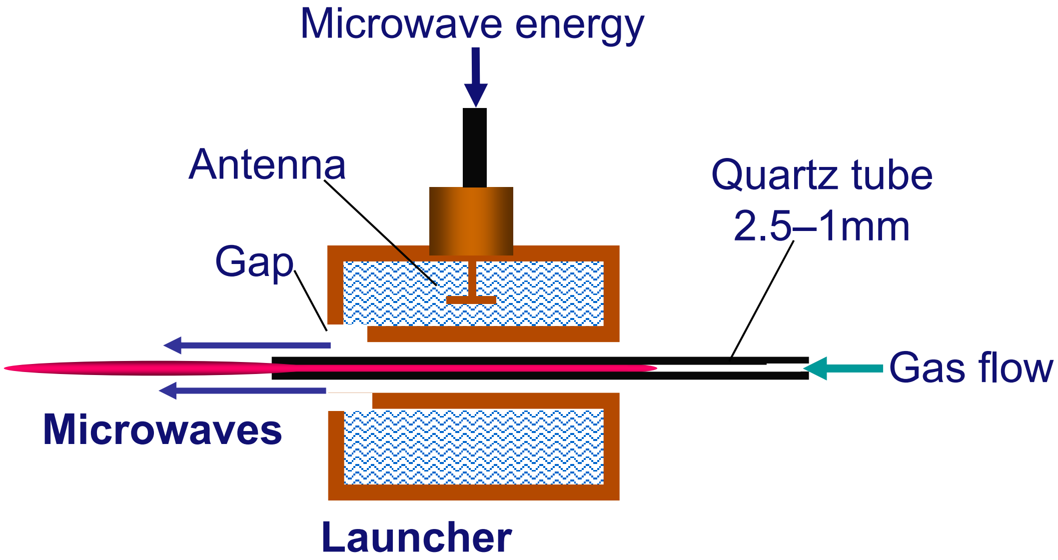
Thomson-Raman separation

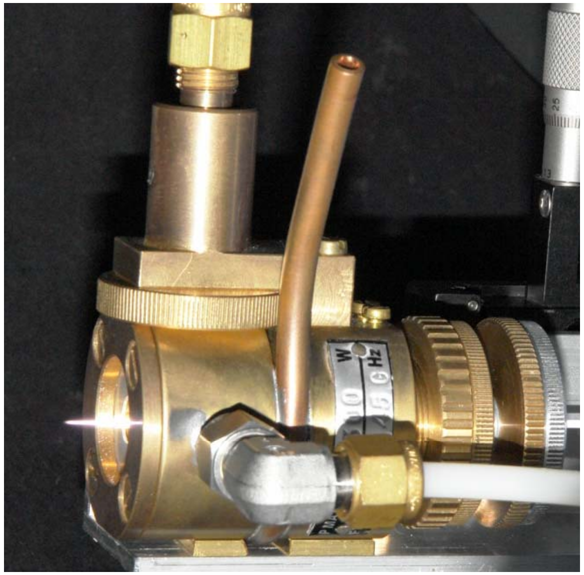
Stray light rejection

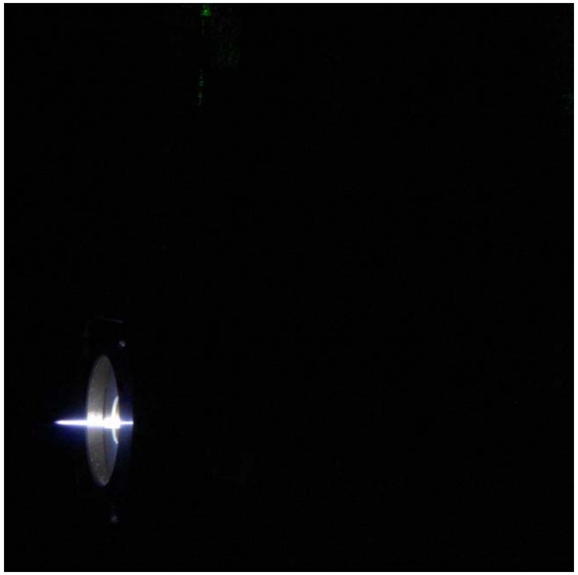
Laser perturbations

Maxwell deviations

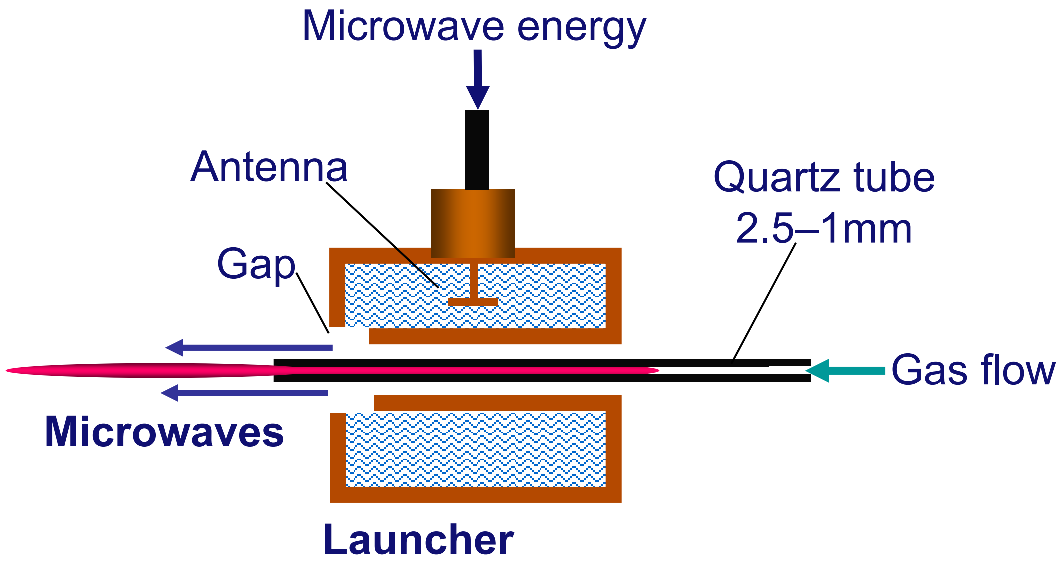
Atmospheric surfatron







Atmospheric surfatron

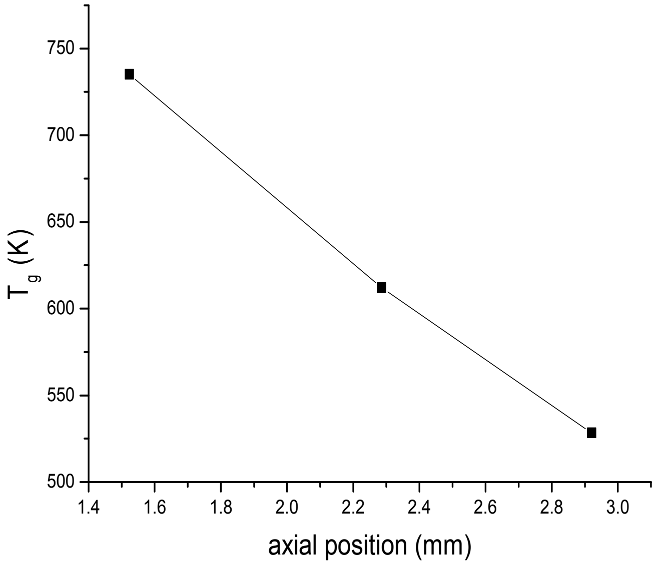


Surfatron torch ~ semi-CAP

Approaches CAP conditions down stream

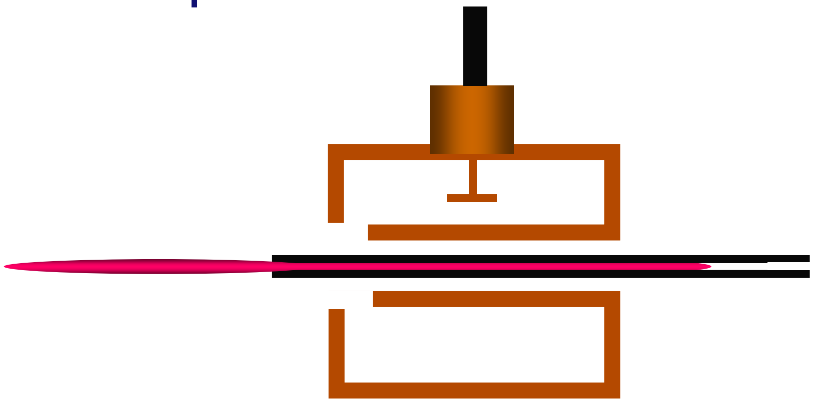
n_e: 10^{20} - 10^{21} m^{-3}

T_e: 1 – 3 eV



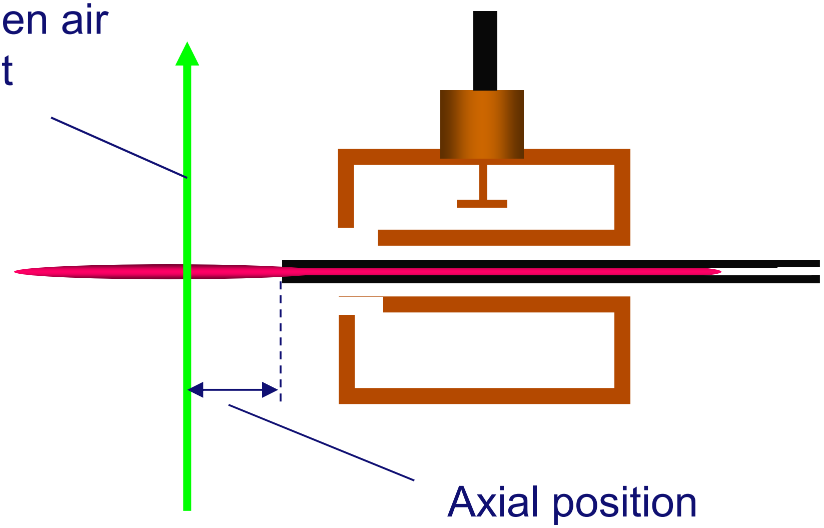
Laser scattering with iCCD

Laser scattering with spatial resolution



Laser scattering with iCCD

Laser scattering with spatial resolution



laser shooting in open air
easy alignment

Axial position

Nd:YAG – 532nm	
~ 8 ns	
10 Hz	5KHz
~ 100 mJ/pulse	~ 4 mJ/pulse

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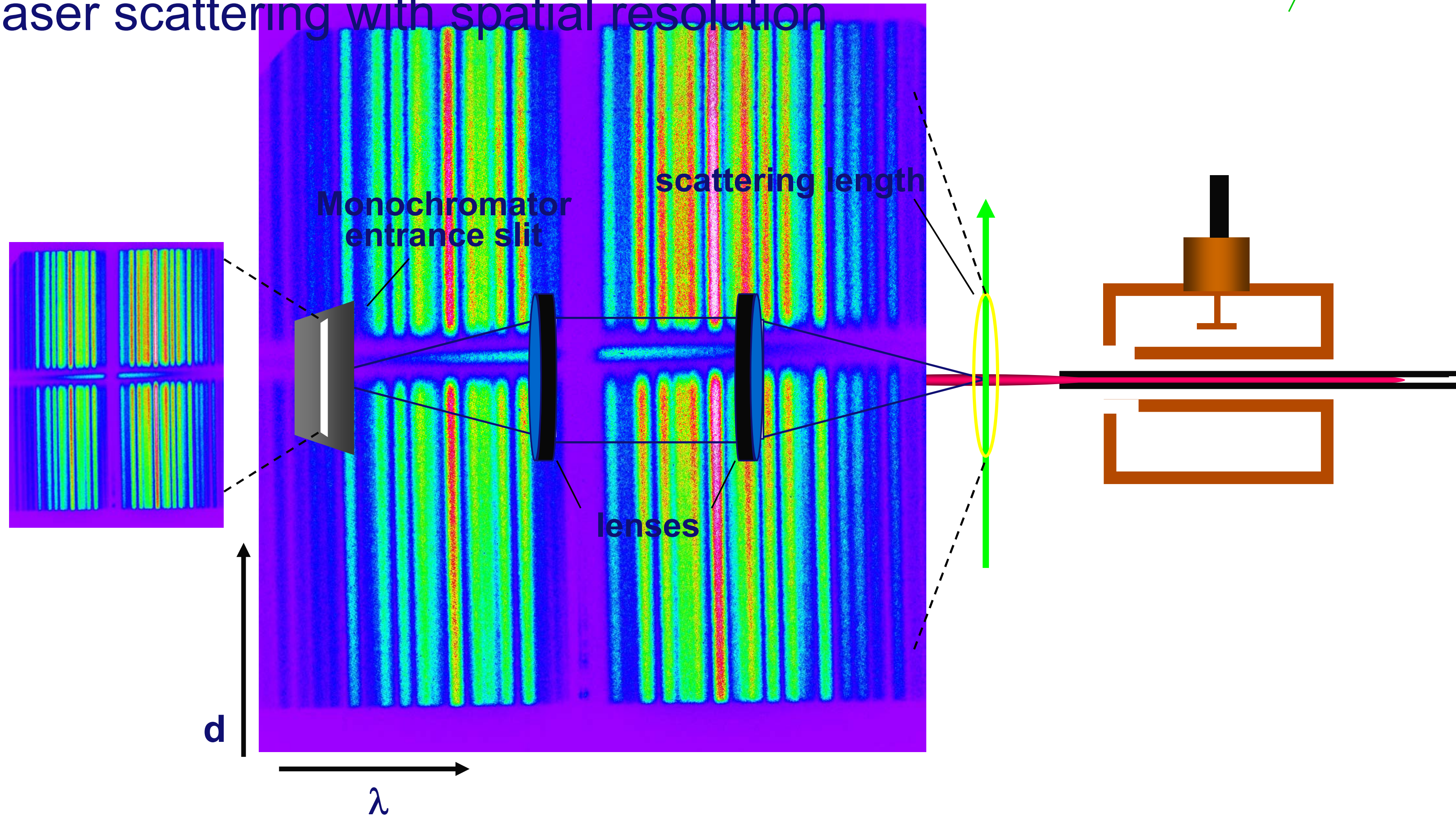
2010 *J Phys D Appl Phys* **43** 395202

Laser scattering with iCCD

Laser scattering with spatial resolution



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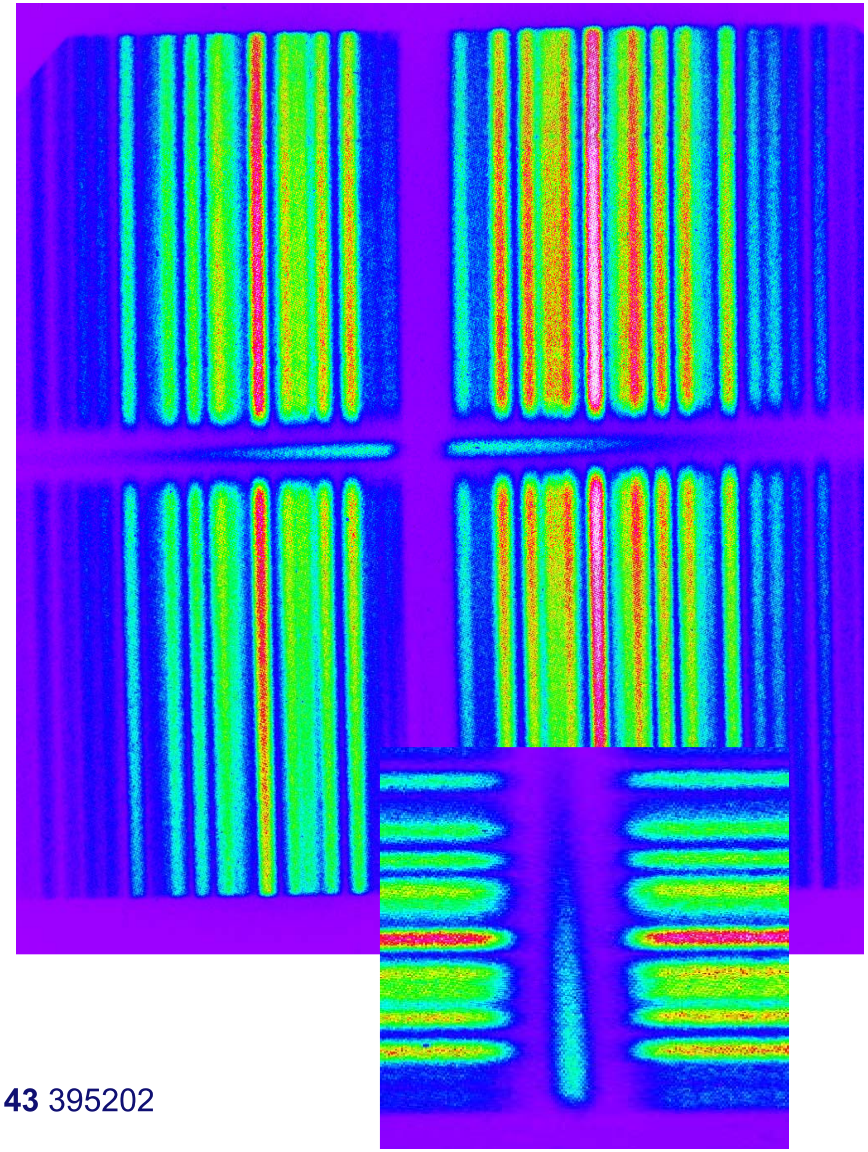
2010 *J Phys D Appl Phys* **43** 395202

Laser scattering with iCCD



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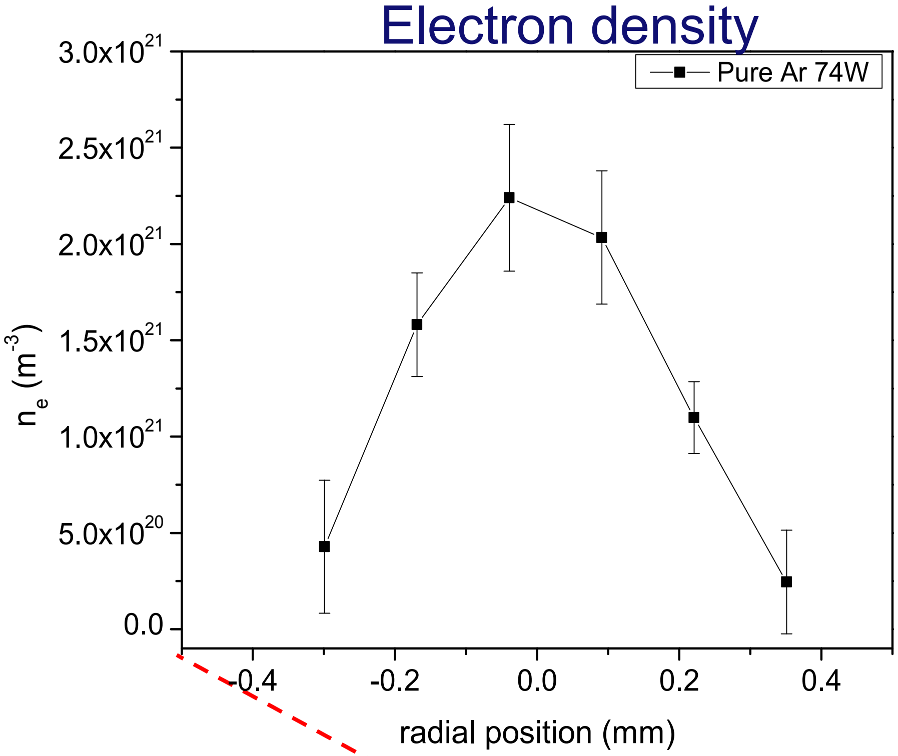
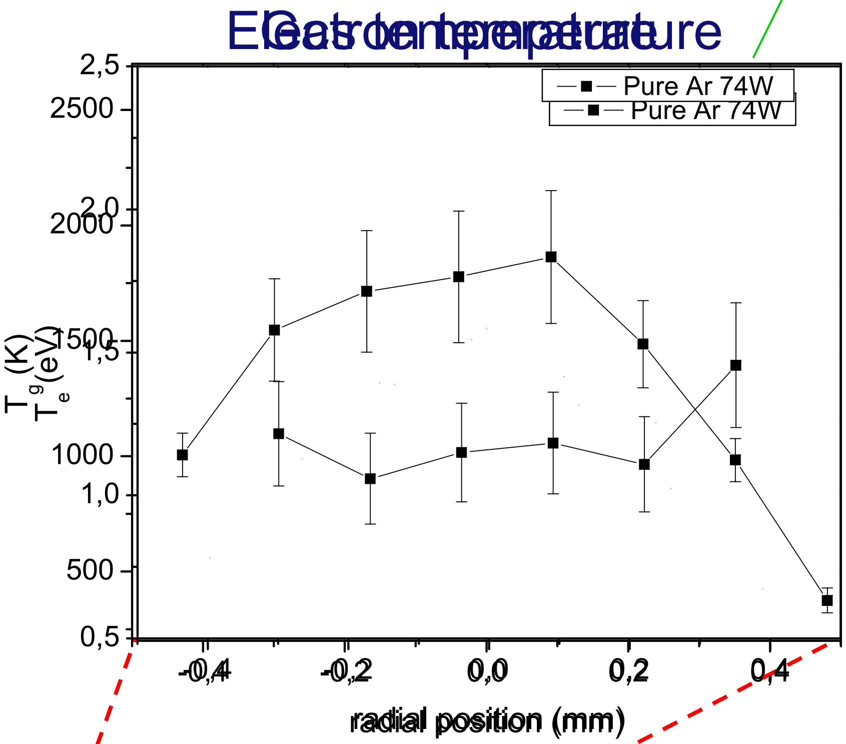
Laser scattering with iCCD

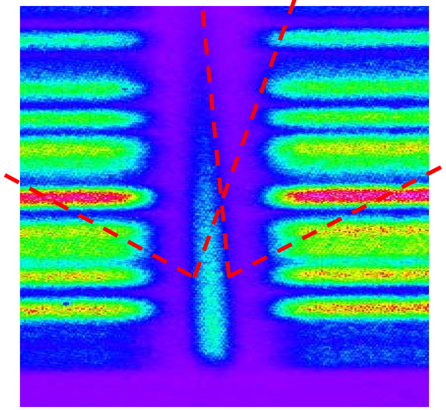


Electron density



Electron temperature

Gas temperature



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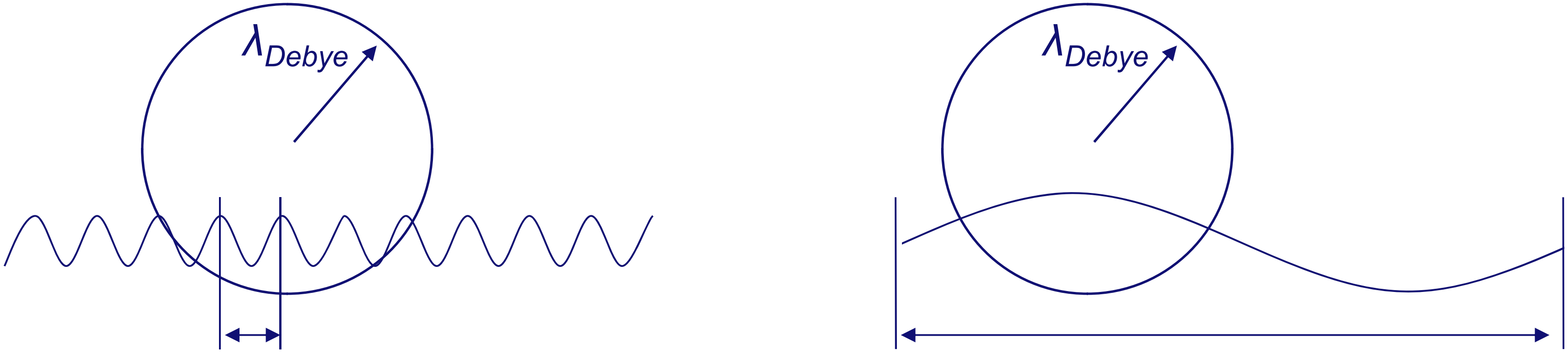
Laser scattering with iCCD

Nature of the scattering

Surfatron torch: $n_e = \leq 10^{21} m^{-3}$, $T_e = 1\text{-}3 eV$ → $\alpha < 0.2$

Scattering parameter: $\alpha \equiv 1/k\lambda_{Debye}$



$2\pi/k$

Incoherent scattering

$2\pi/k$

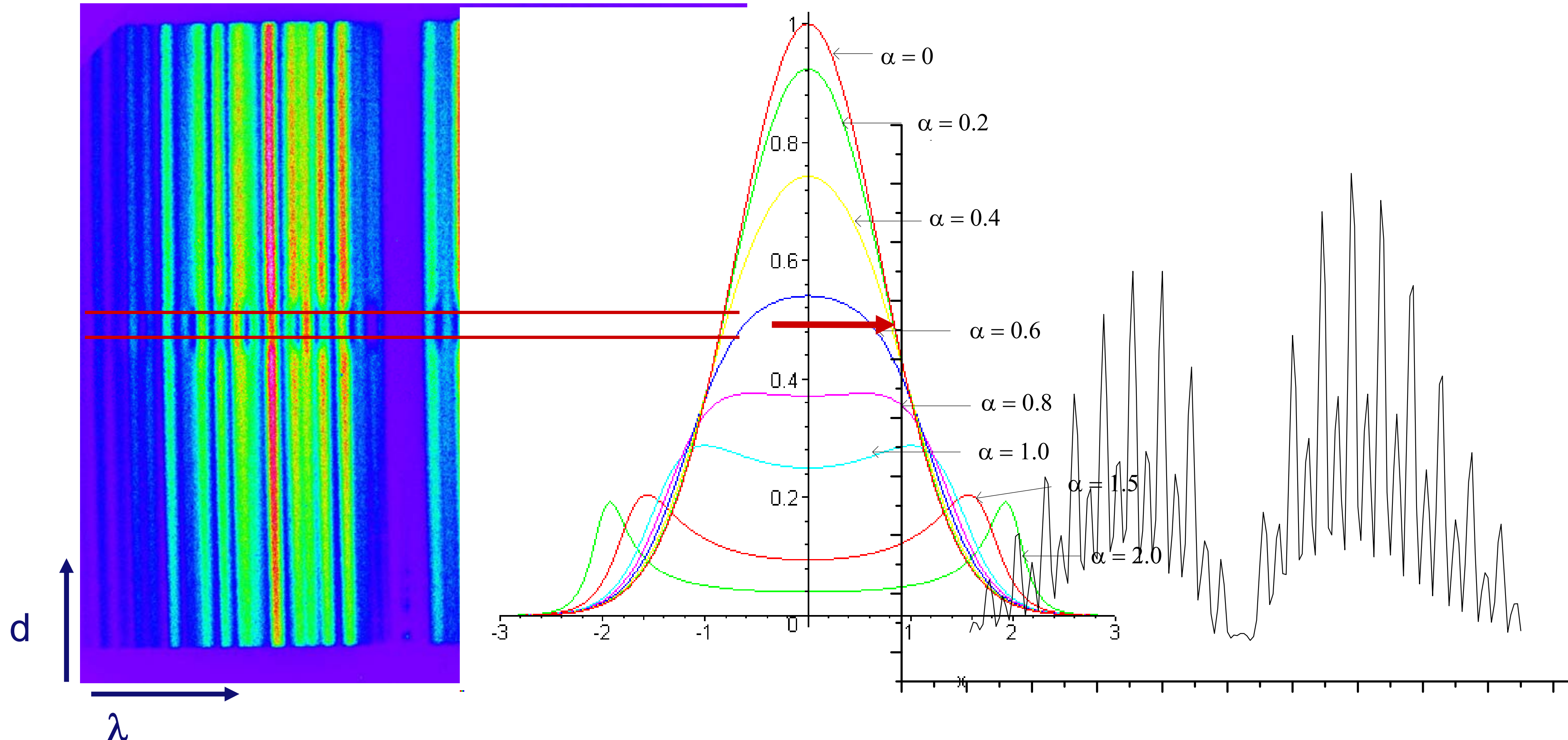
Coherent scattering

Laser scattering with iCCD

Nature of the scattering

Surfatron torch: n_e= 10^{20}-10^{21}m-3, T_e= 1-3eV → α < 0.2

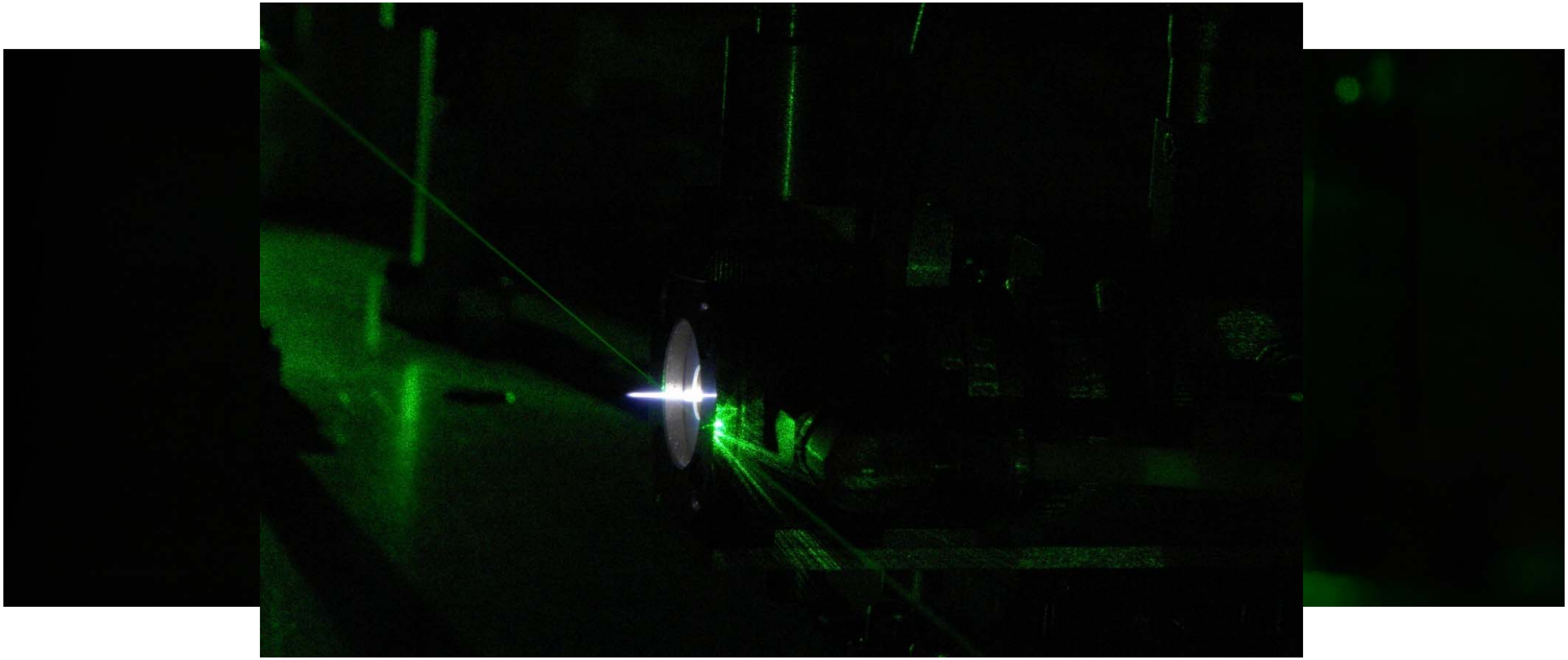
Incoherent scattering → absolute calibration → Raman scattering



Stray light rejection

Stray light: laser beam (or side beam) reflections on mirrors, lenses, windows, surfaces….

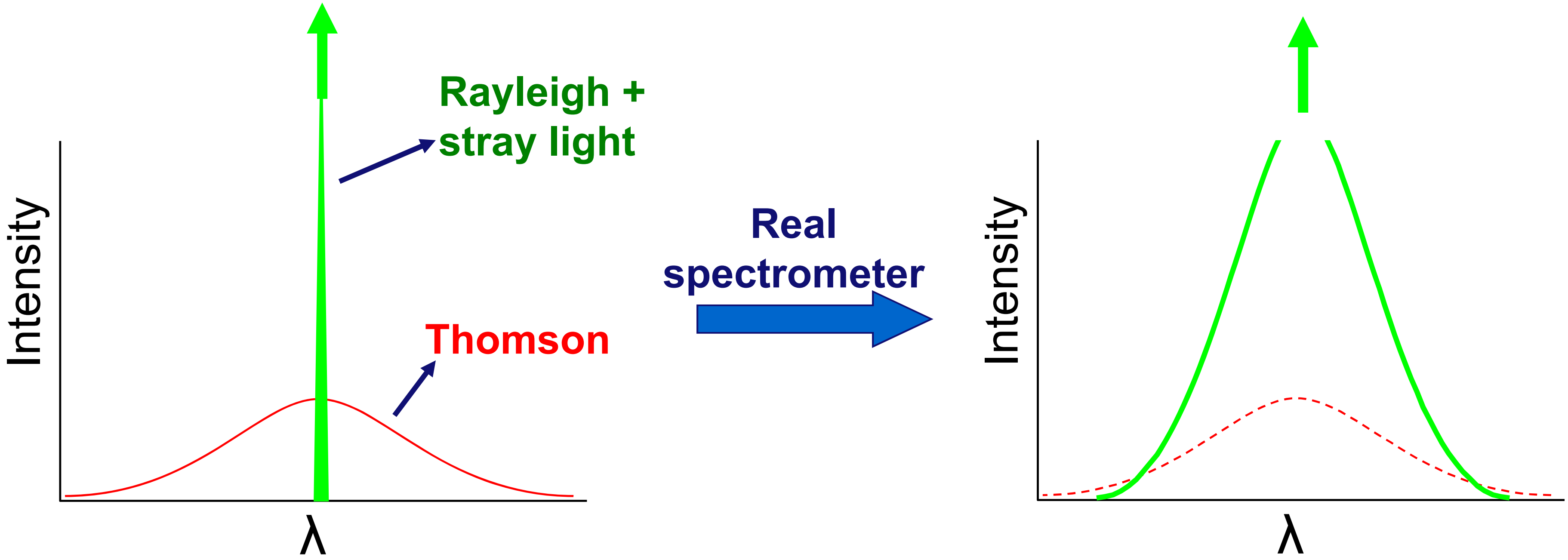
Even in plasmas working in open air



Stray light rejection

Rayleigh scattering + stray light >> Thomson scattering

In Ar CAPs ($n_e / n_a \sim 10^{-5}$) → $I_{TS}/I_{Ray} \sim 10^{-3}$



Notch filter needed

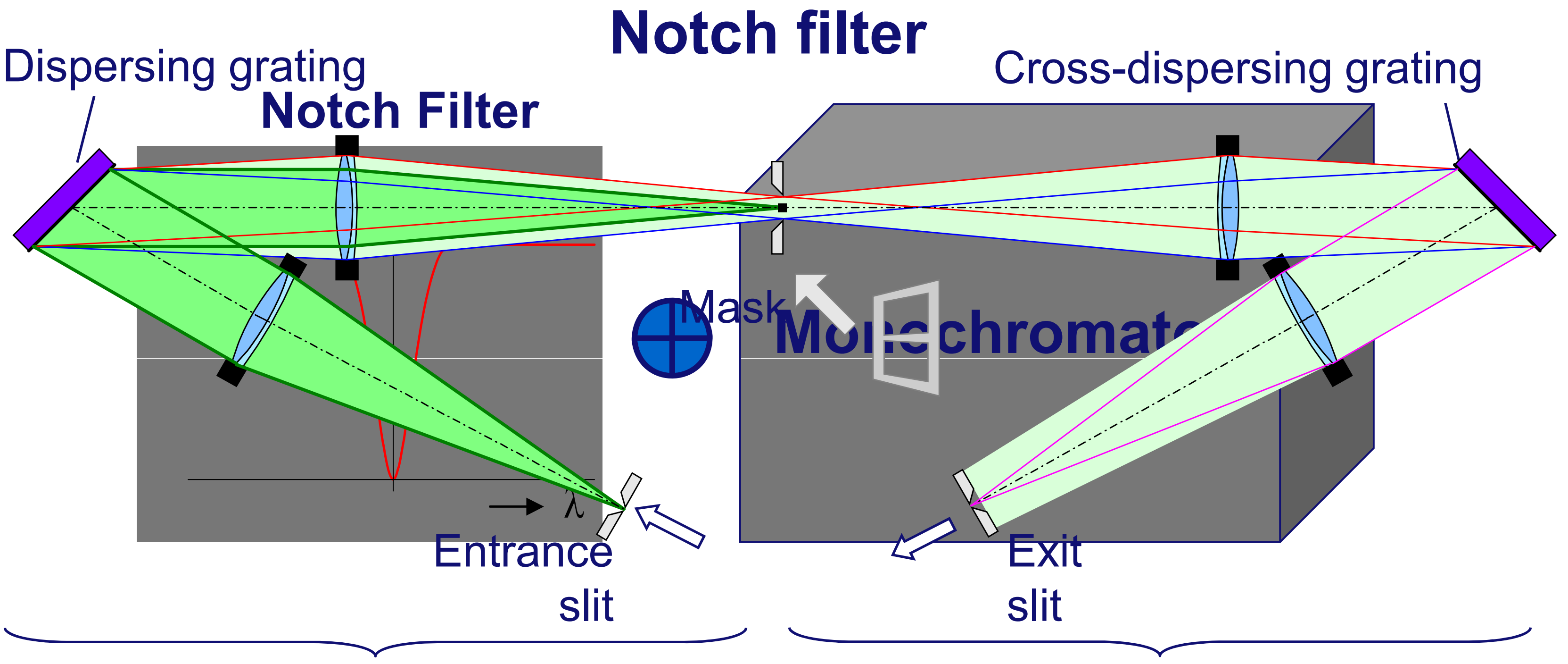
Triple Grating Spectrograph

TGS

Stray light rejection

Triple Grating Spectrograph

TGS



Monochromator I

Monochromator II

|||

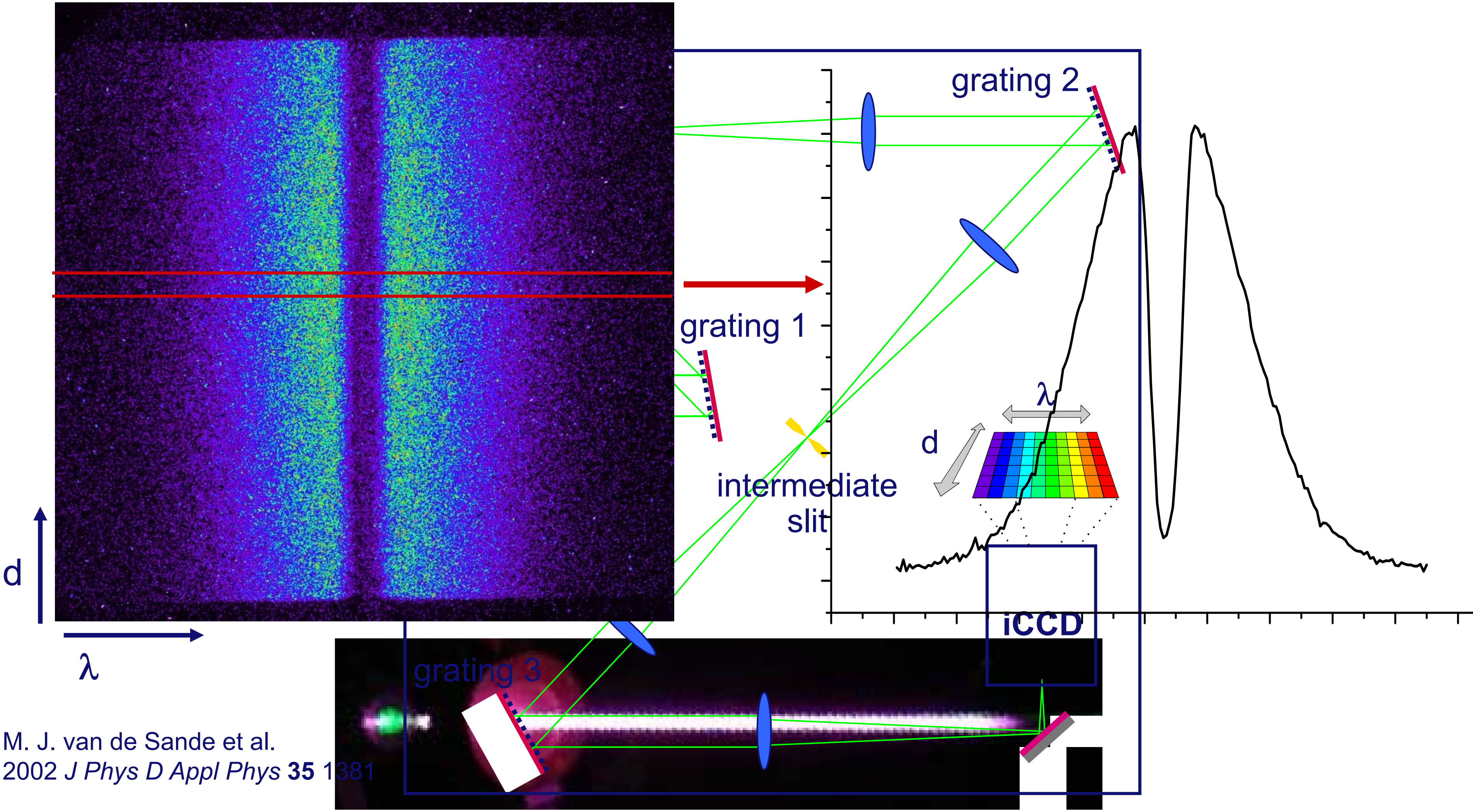
(Monocromator I)$^{-1}$

M. J. van de Sande et al.
2002 *J Phys D Appl Phys* **35** 1381

Stray light rejection

Triple Grating Spectrograph

TGS



M. J. van de Sande et al.
2002 *J Phys D Appl Phys* **35** 1381

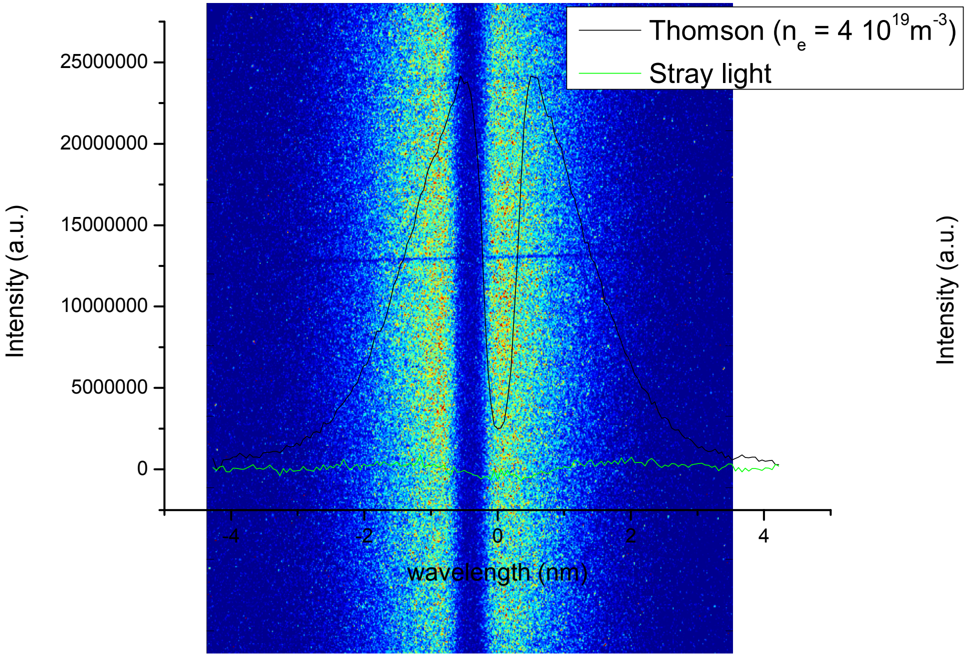
Stray light and detection limit

Detection limit of TGS:

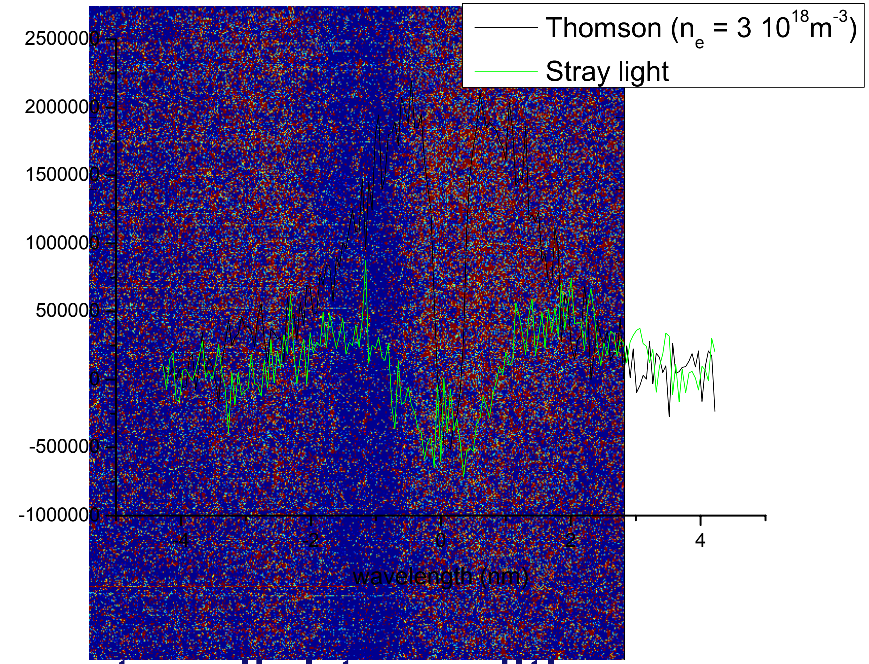
fraction collected (solid angle) ~ 10^{-3}
fraction detected (optics + iCCD) ~10^{-1}-10^{-2}
scattering length ~ 10^{-2} m

laser energy, plasma light, noise…

Thomson signal



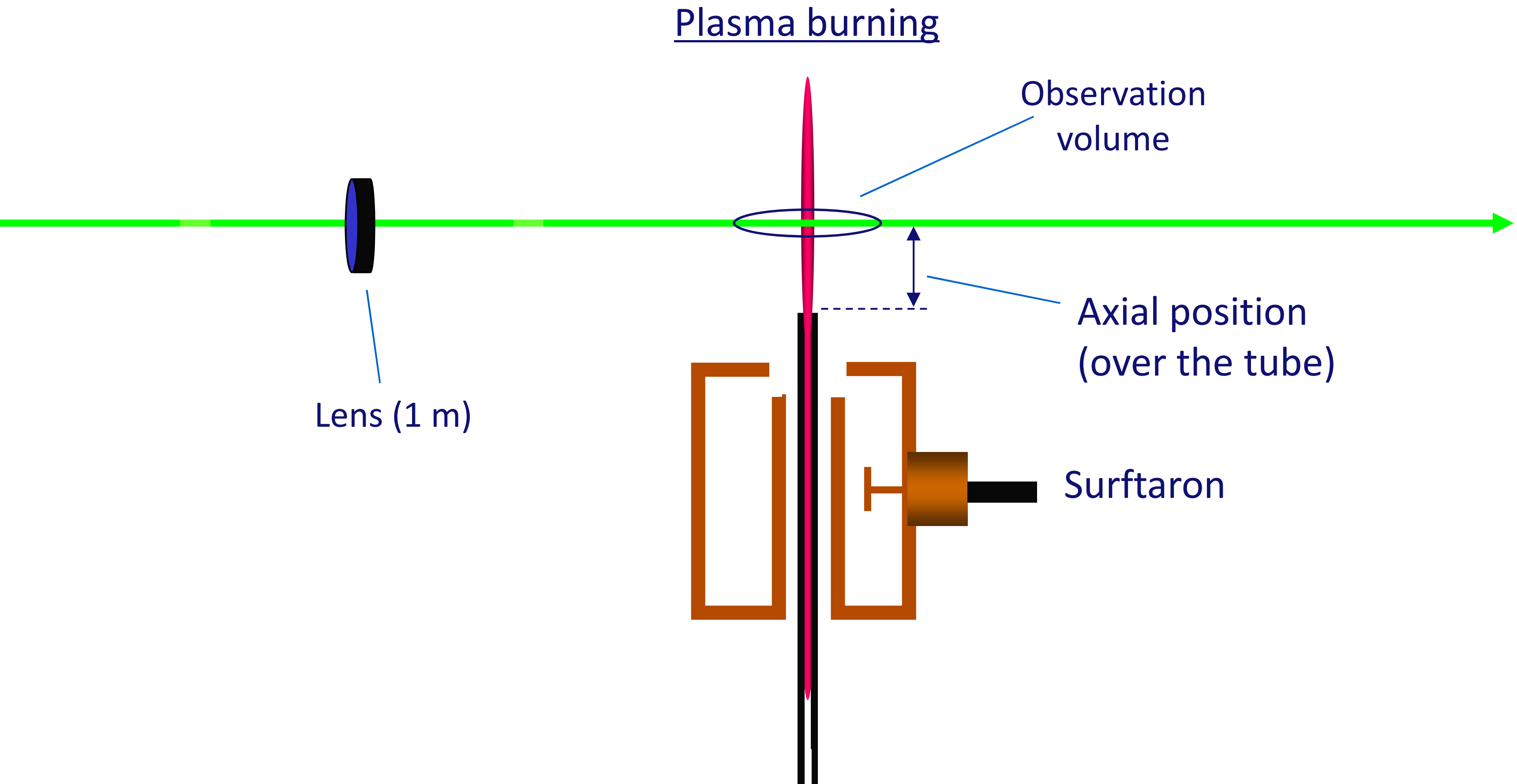
Stray light (x100)



Detection limit → depends on stray light conditions

Stray light and Rayleigh

Collected signal: Stray light + Rayleigh

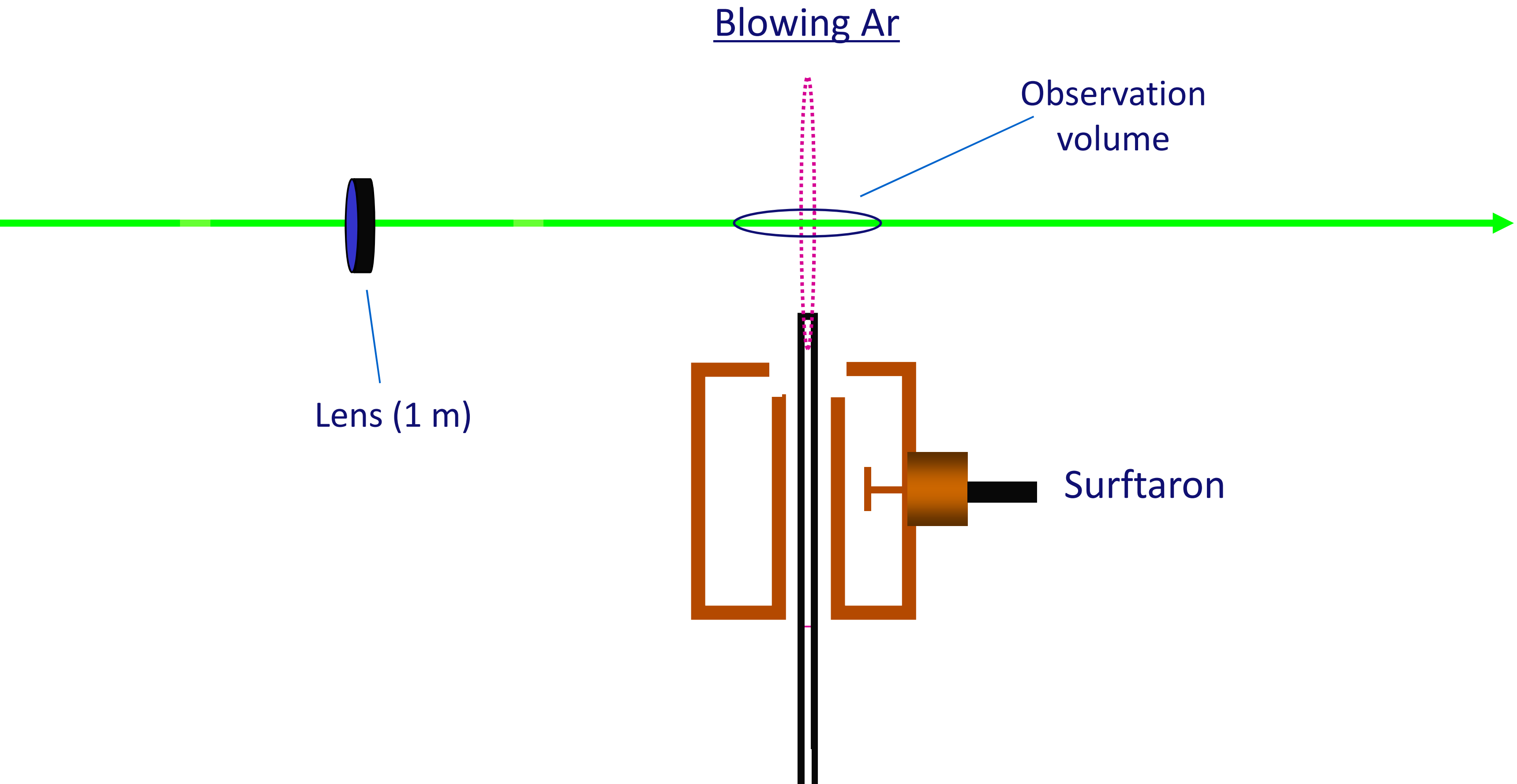


Stray light and Rayleigh

Collected signal: Stray light + Rayleigh

Stray light = constant

Rayleigh = *f*(pressure, gas)



Stray light and Rayleigh

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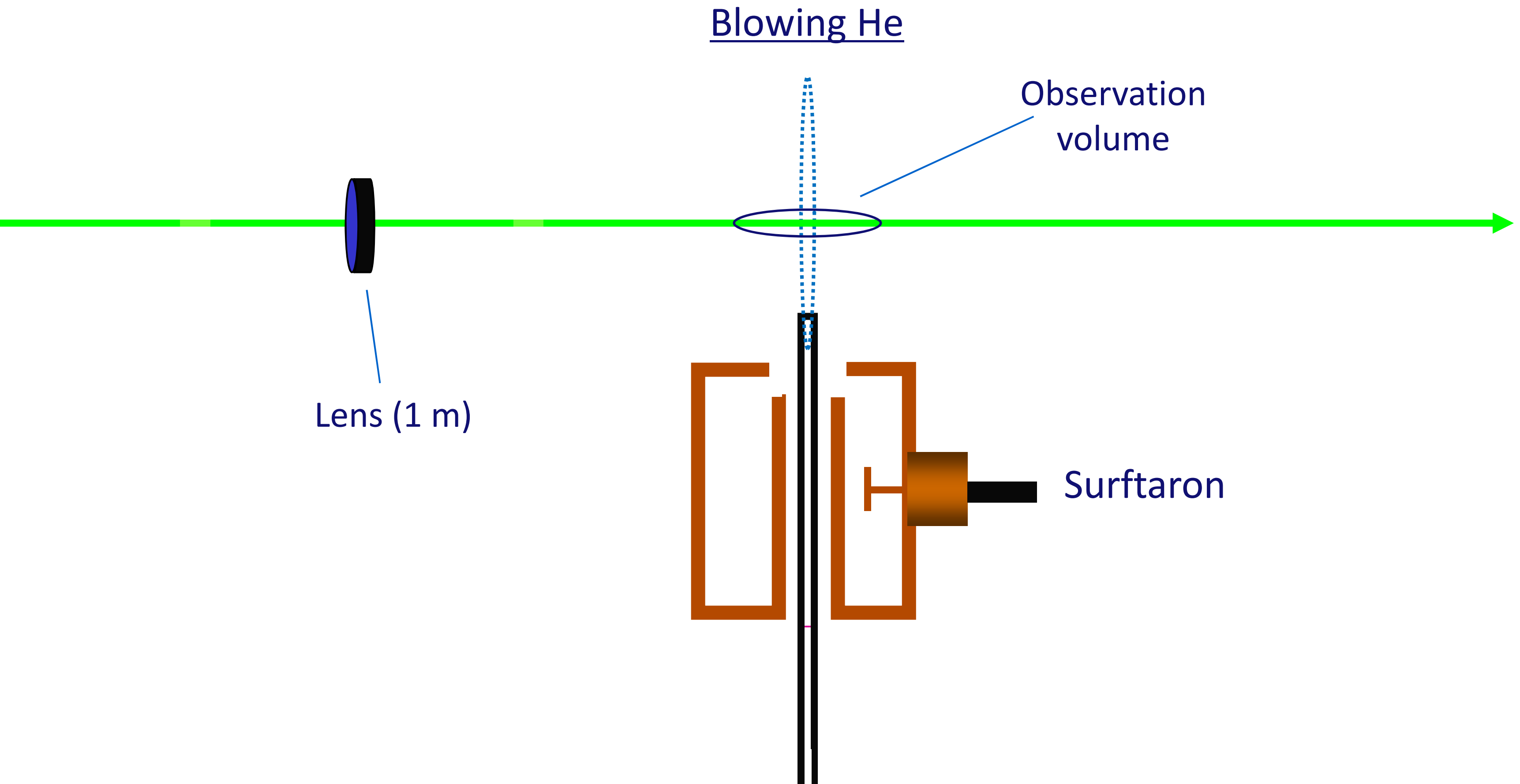
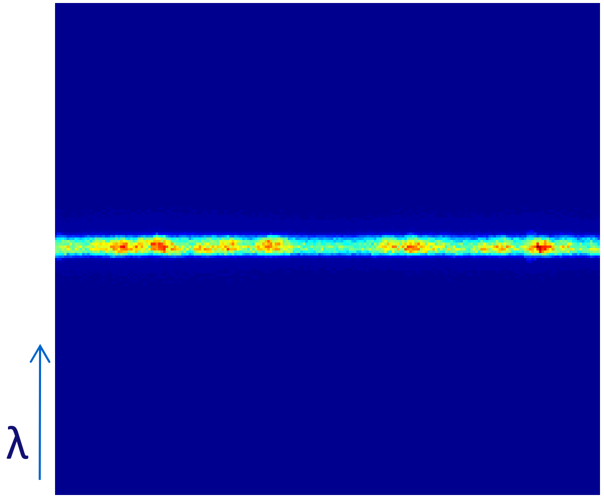
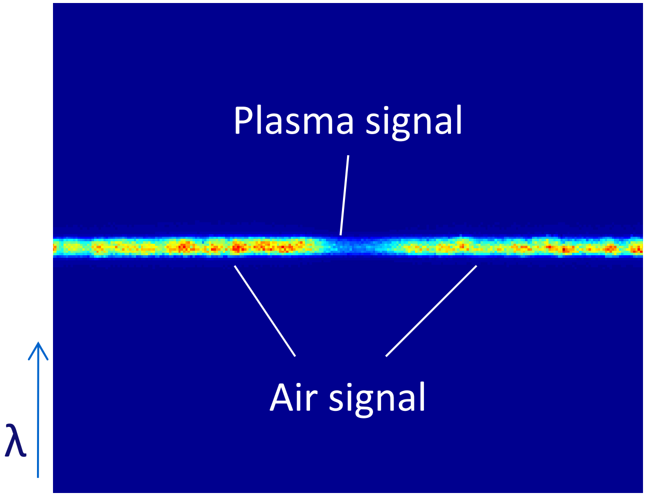


Stray light and Rayleigh

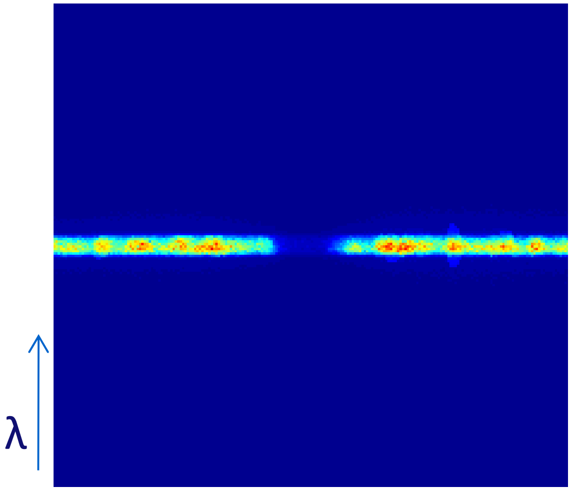
Blowing Ar

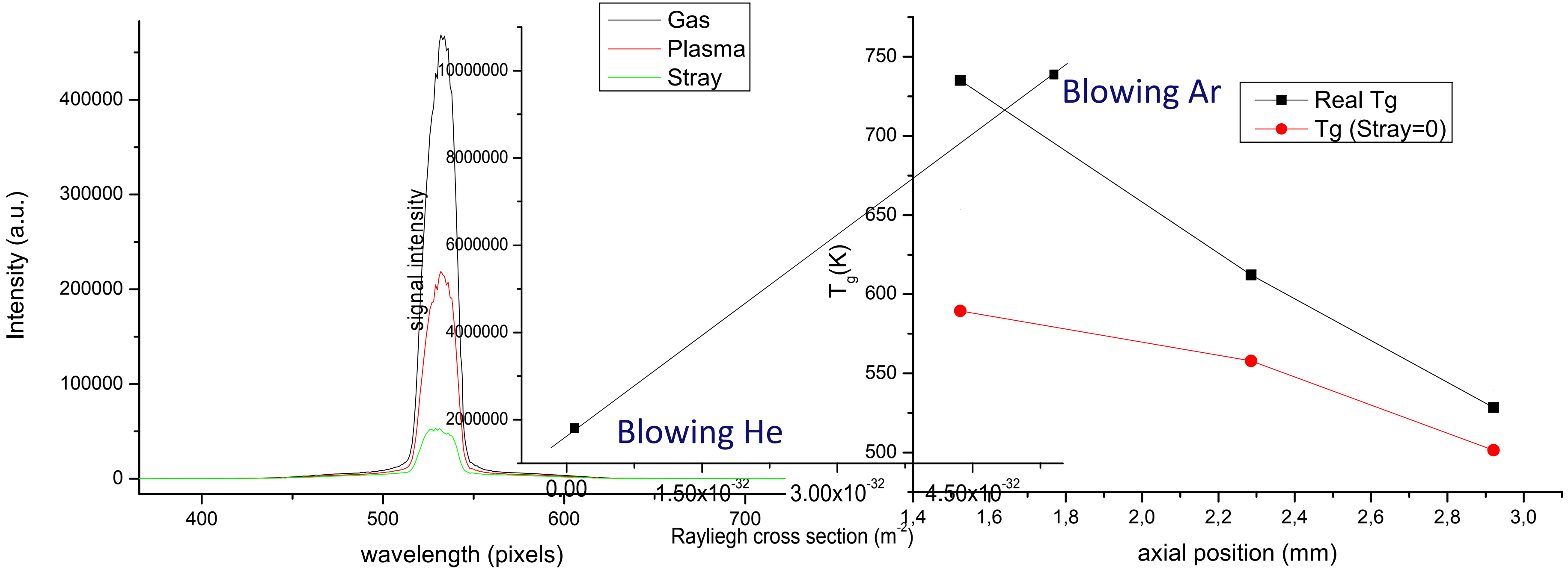


Ar Plasma



Blowing He



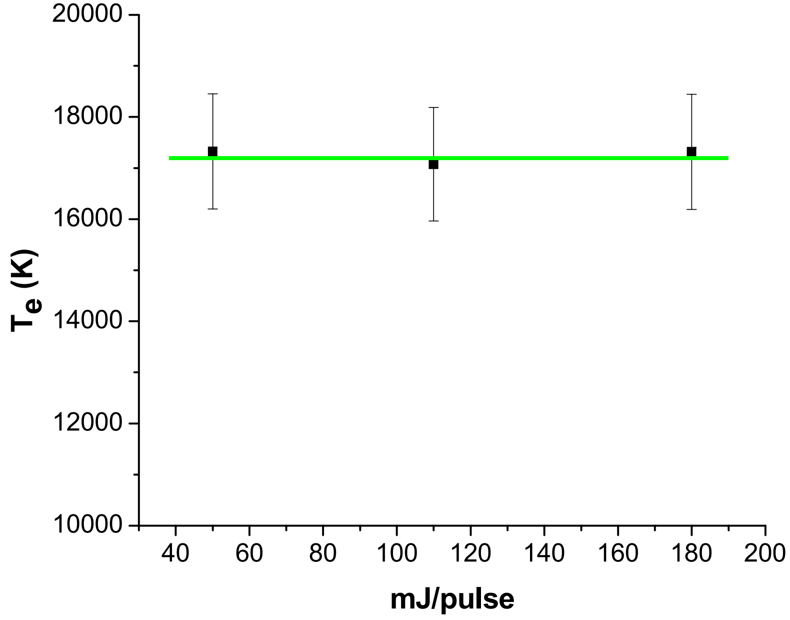


Laser perturbations

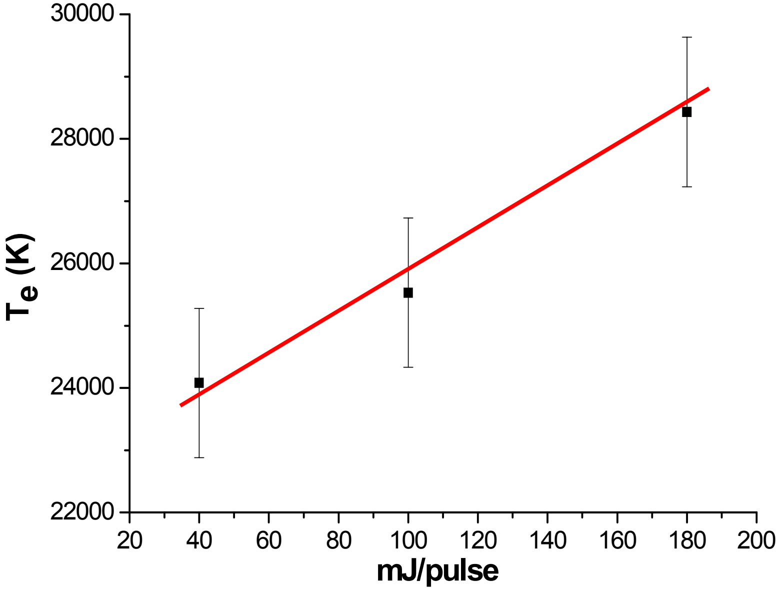
Electron heating via inverse Bremsstrahlung ↑ T_e

Photo ionization: direct or multi-photon absorption ↑ n_e

Low pressure



Atmospheric pressure



Laser perturbations

Electron heating by inverse Bremsstrahlung

Normally considered only electron-ion interactions

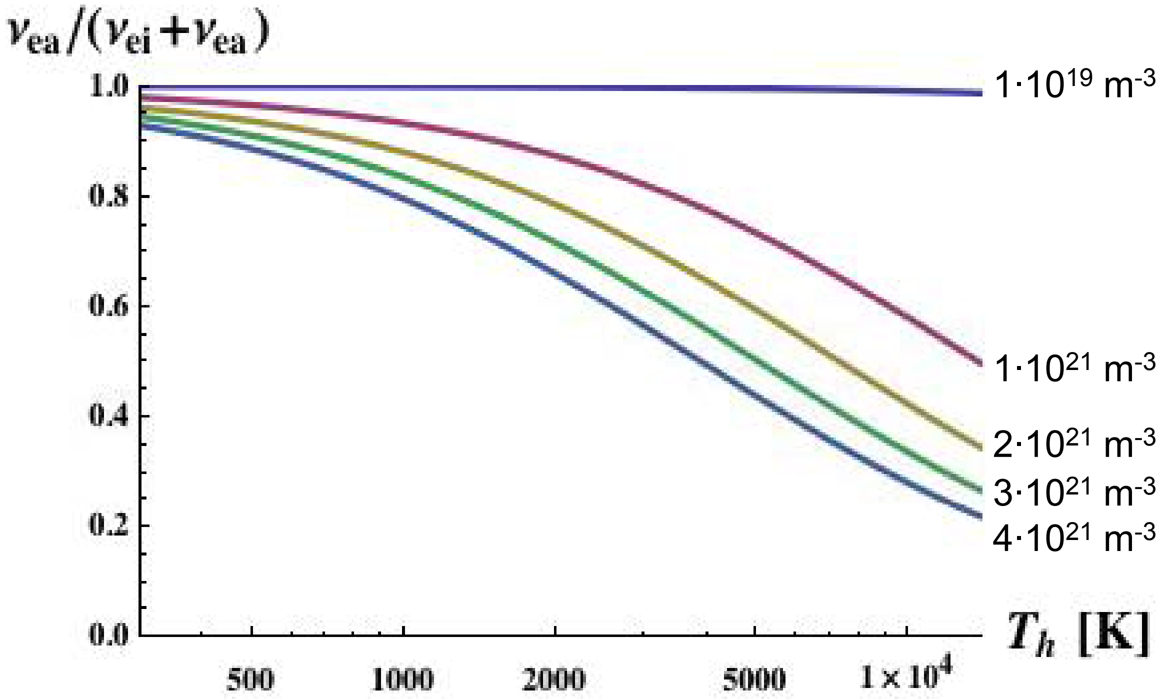
electron-atom collisions can be important in CAPs conditions

$$\frac{\Delta T_e}{T_e} \propto F \times \nu_{ei}$$

F: laser fluency (J/m^2)

ν_{ei}: electron-ion collision frequency

ν_{ea}: electron-atom collision frequency



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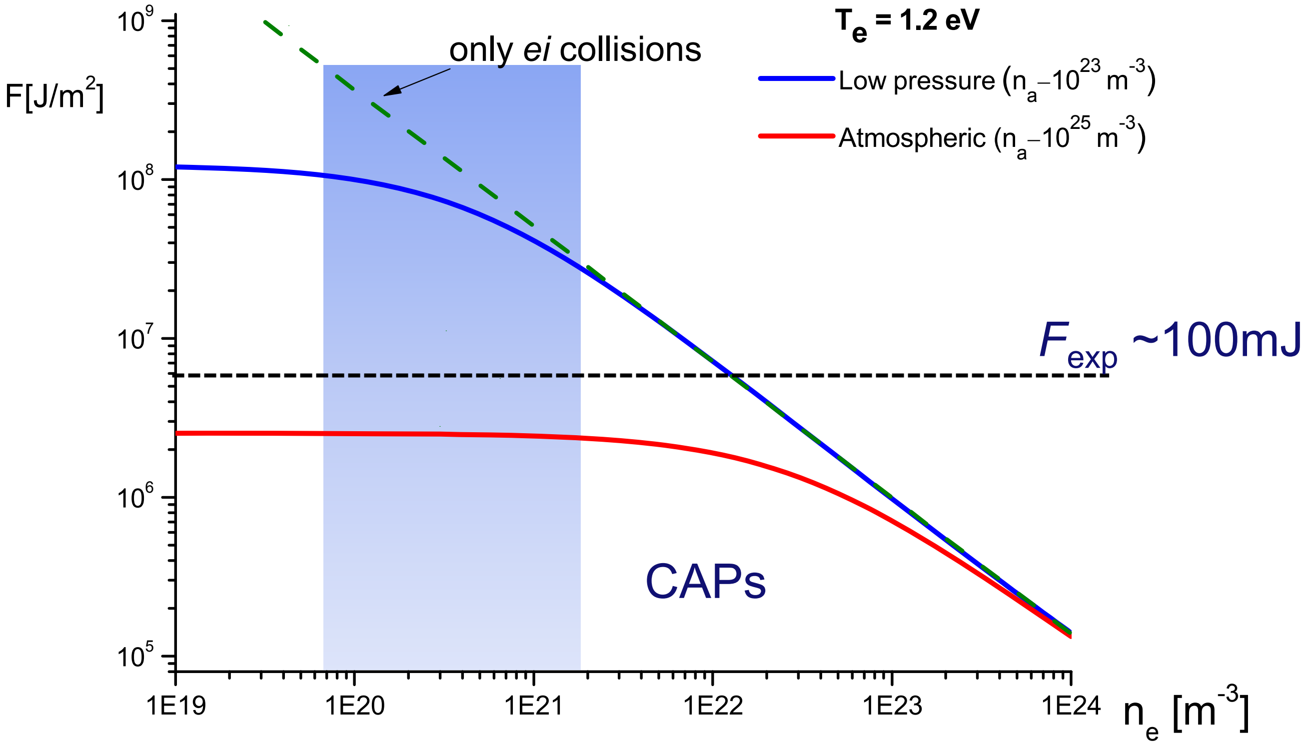
Session VII Poster P1-3

Laser perturbations

Electron heating by inverse Bremsstrahlung

- Normally considered only electron-ion interactions
- electron-atom collisions can be important in CAPs conditions

Laser fluency needed for a 10% heating ($\Delta T_e / T_e = 0.1$)



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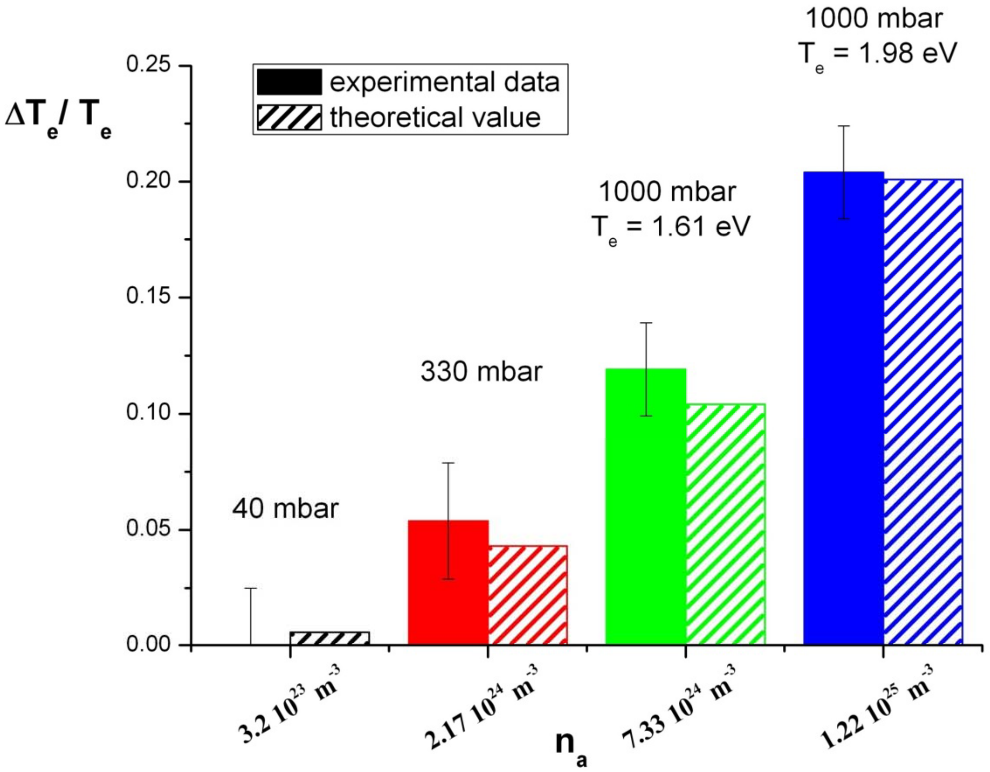
Laser perturbations

Electron heating by inverse Bremsstrahlung

- Normally considered only electron-ion interactions
- electron-atom collisions can be important in CAPs conditions

Experimental validation



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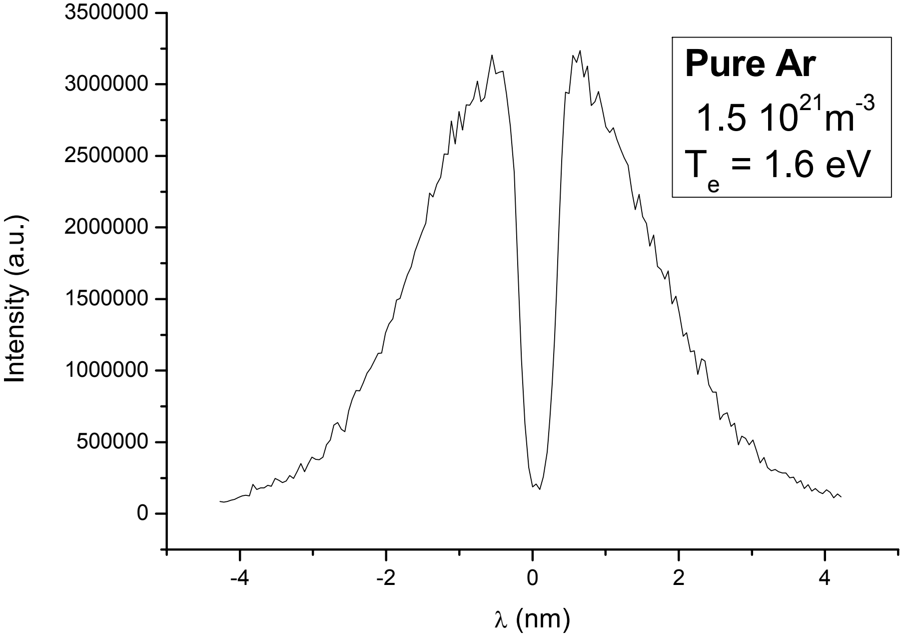
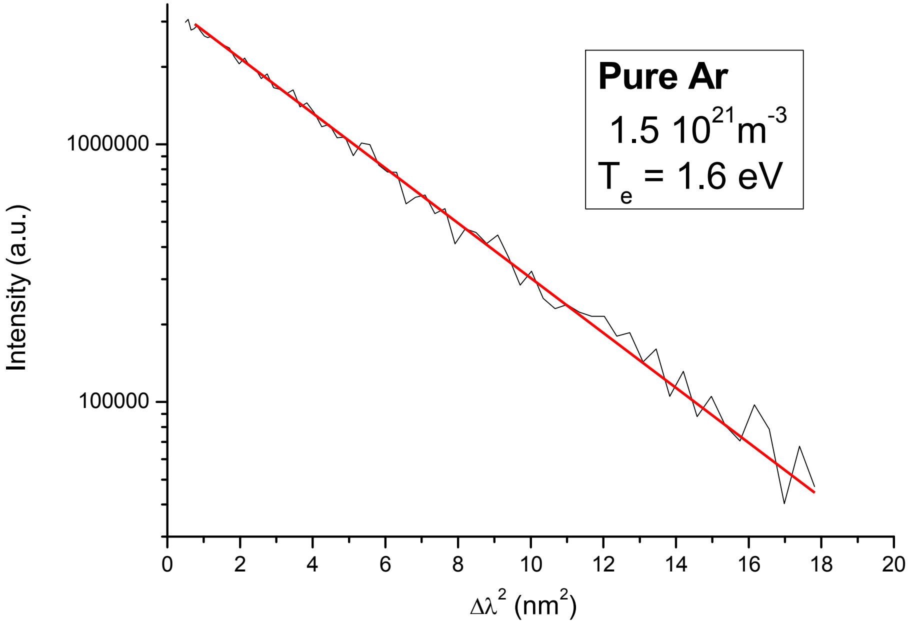
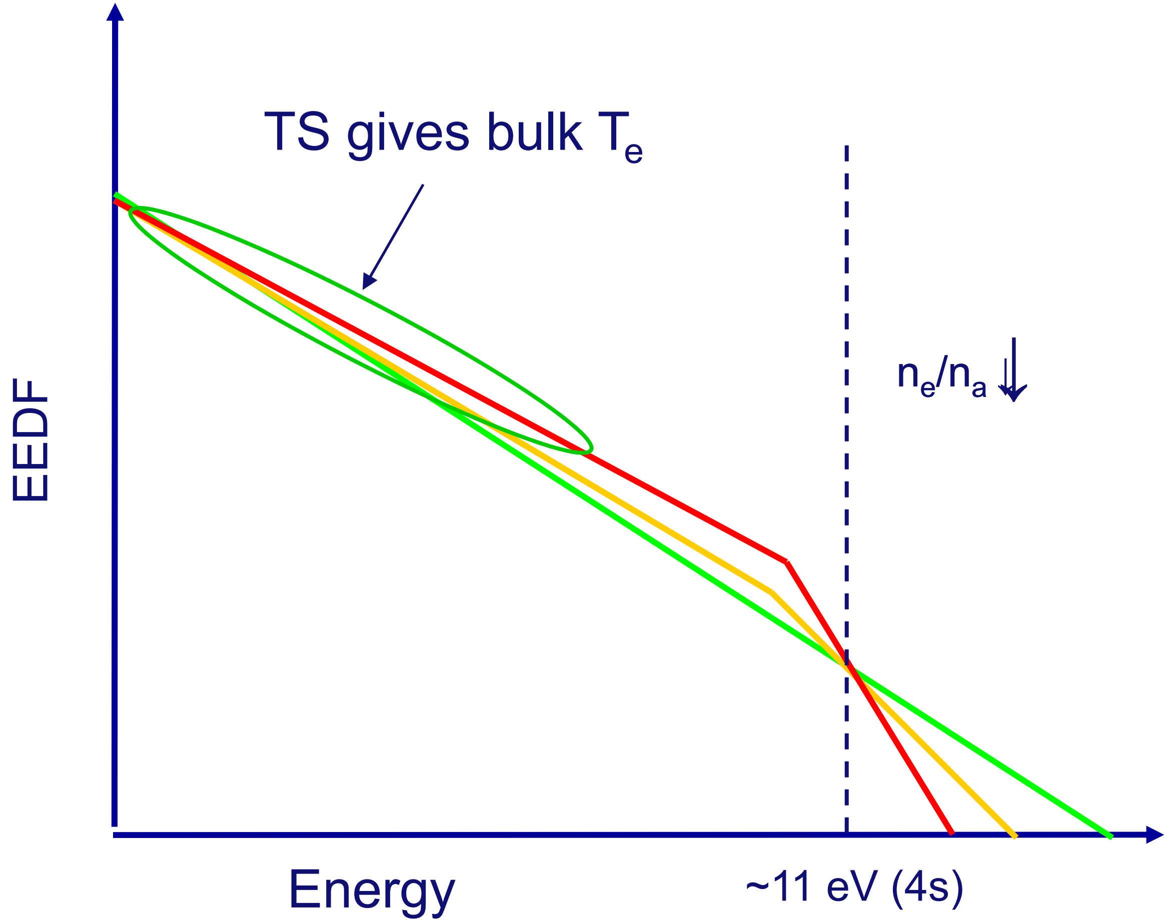
EEDF deviations

- EEDF deviations on argon plasmas:
  - Low pressures (<100mTorr)
  - Gas mixtures (Ar-He, Ar-CF_4)
- Atmospheric pressure - pure argon plasma
- However, TS only “sees” the bulk of the EEDF
- Deviations can occur on the tail





EEDF deviations

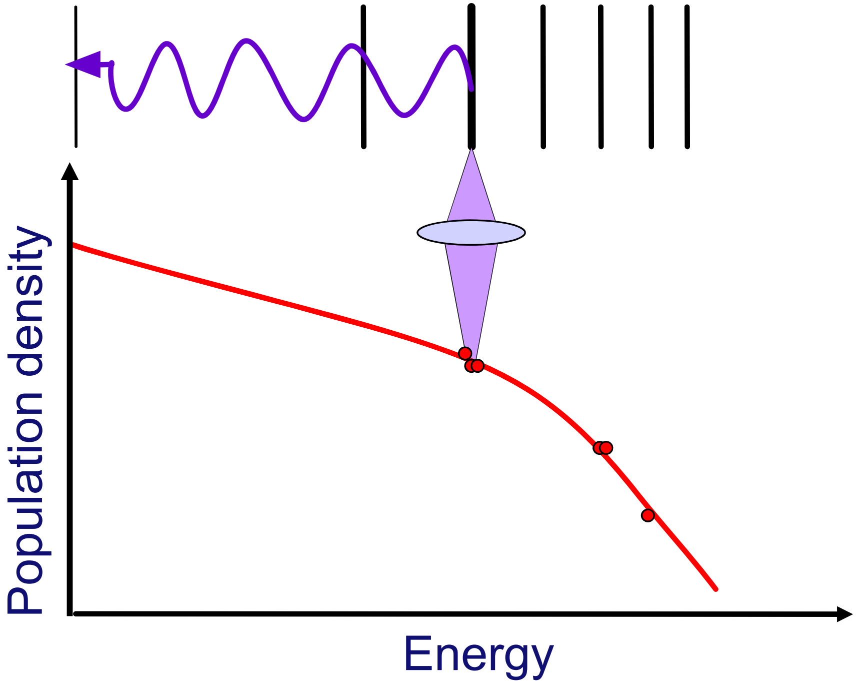


EEDF deviations

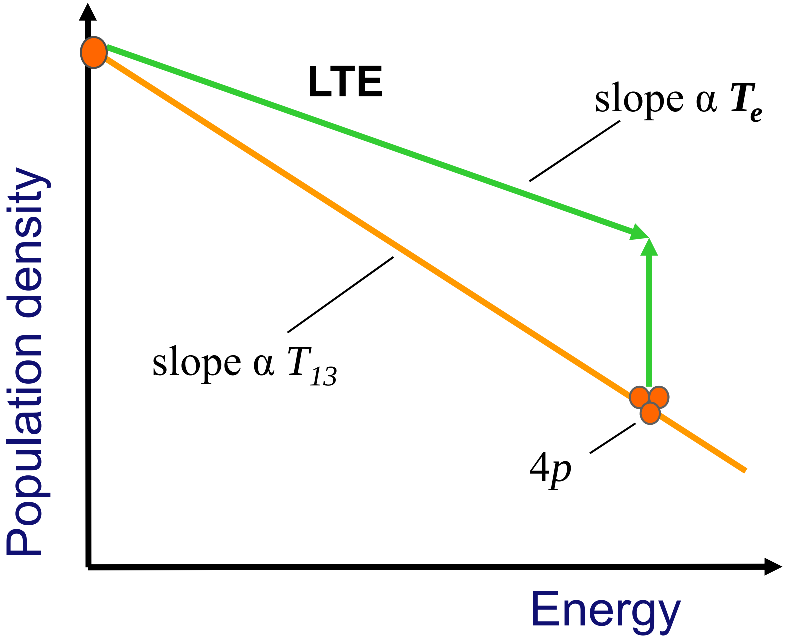
Measuring a global temperature

Creation temperature: T_e(creation)

Line intensity →ASDF

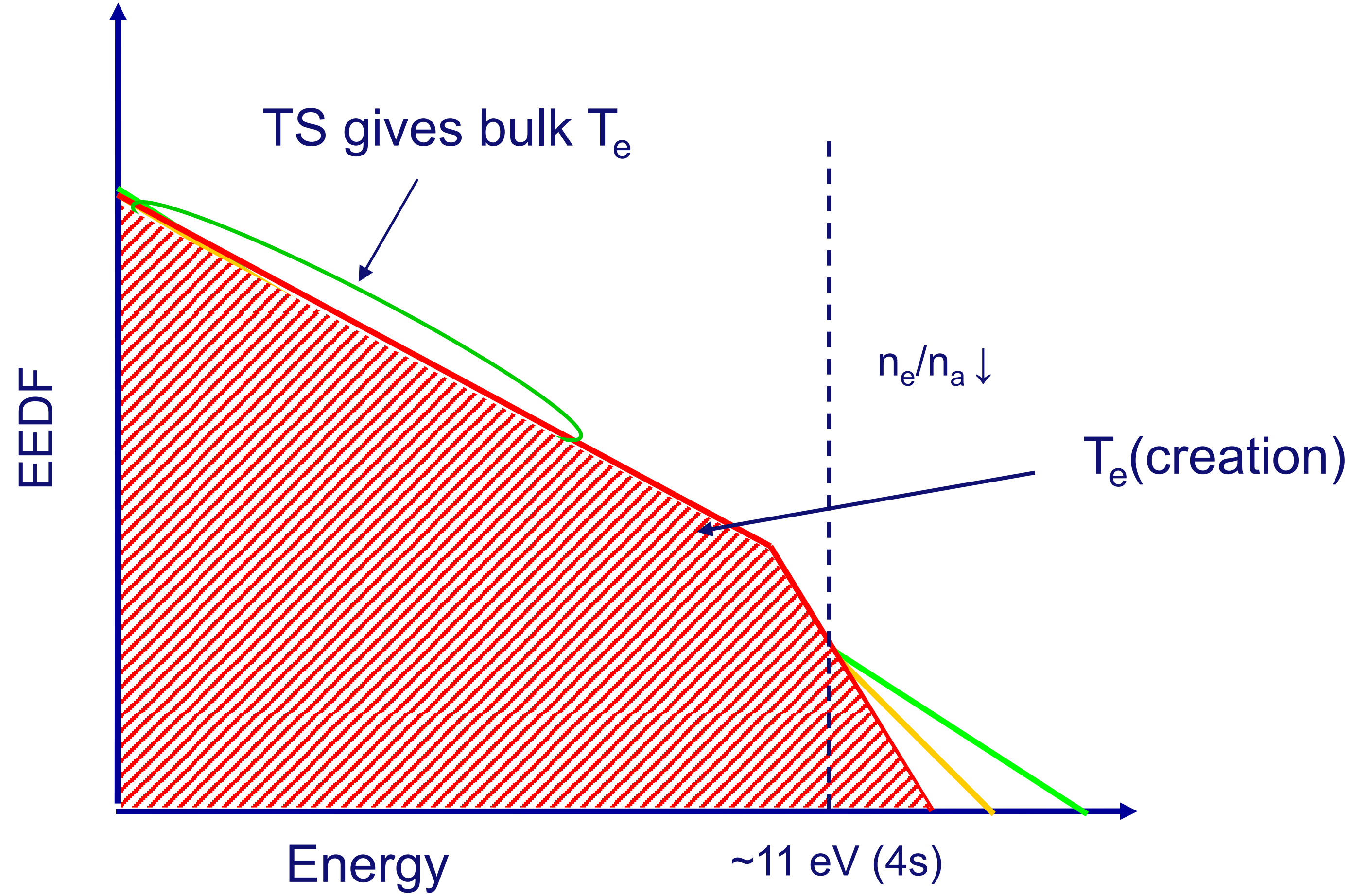


Collisional Radiative Model (CRM)



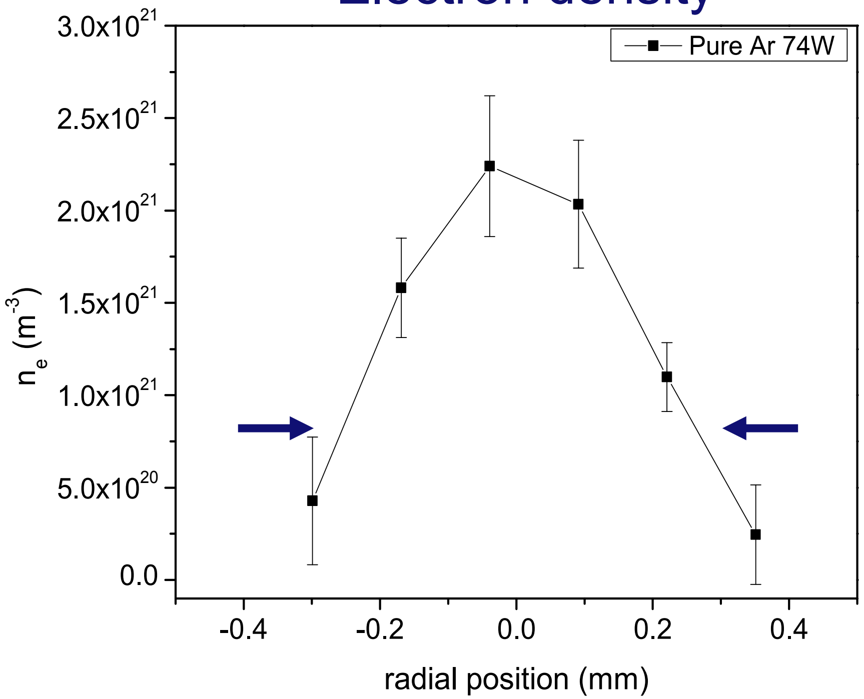
E Iordanova et al.
2009 *J Phys D Appl Phys* **42** 155208

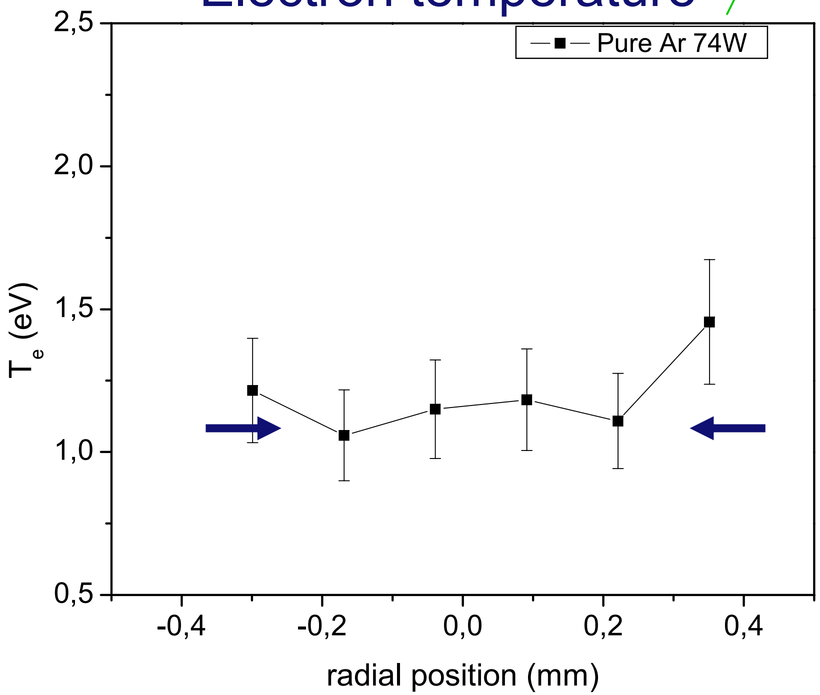
EEDF deviations



T_e(TS) ≥ T_e(creation)

EEDF deviations


Electron density

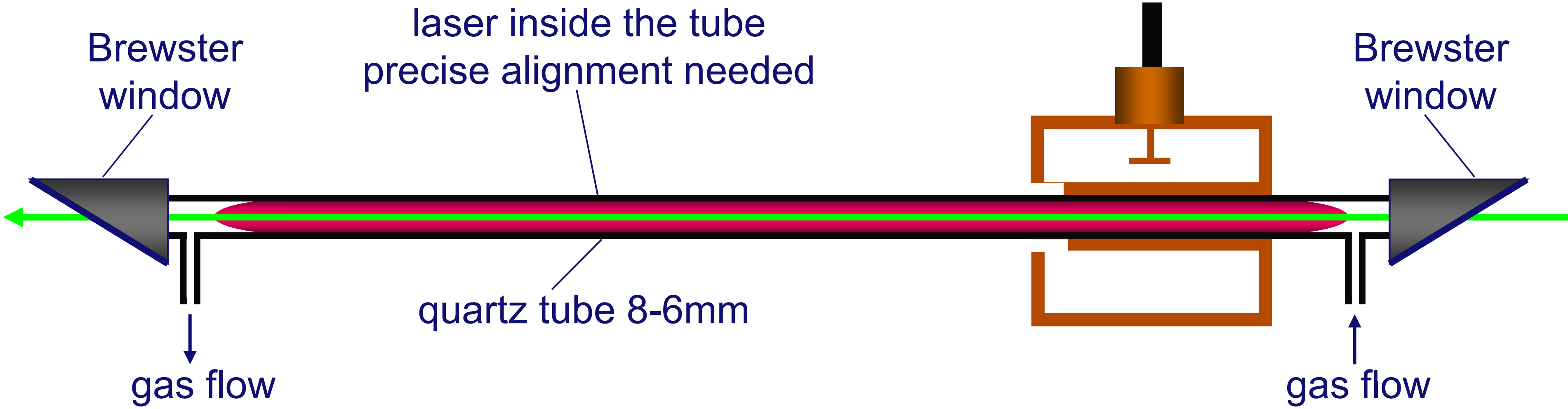

Electron temperature

air contamination

Presence of molecules → higher losses → higher T_e

EEDF tail depletion

EEDF deviations



Brewster window

laser inside the tube
precise alignment needed

Brewster window

quartz tube 8-6mm

gas flow

gas flow

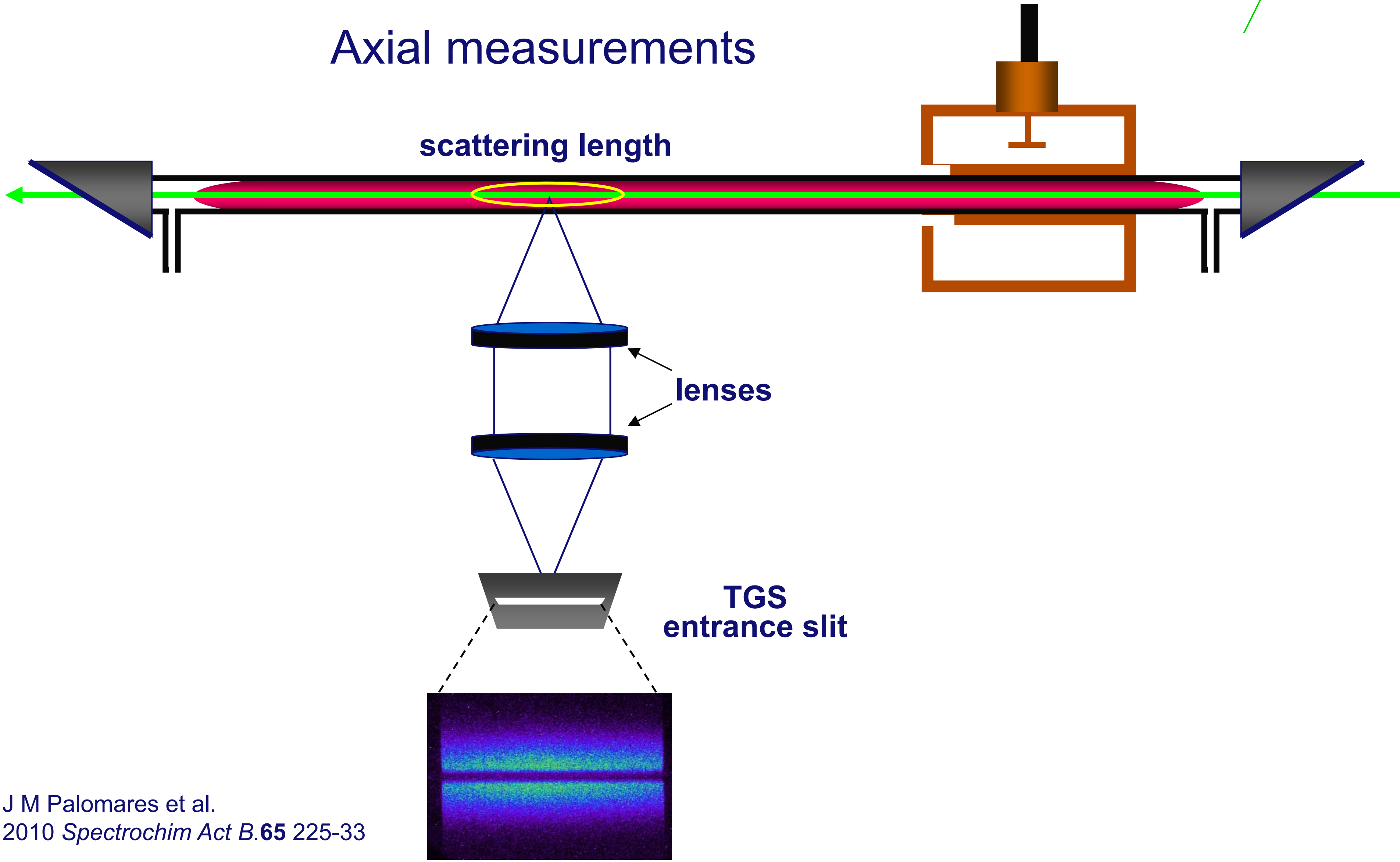
Argon gas

Pressure: 6 – 20mbars

J M Palomares et al.
2010 *Spectrochim Act B*.**65** 225-33

EEDF deviations

Axial measurements



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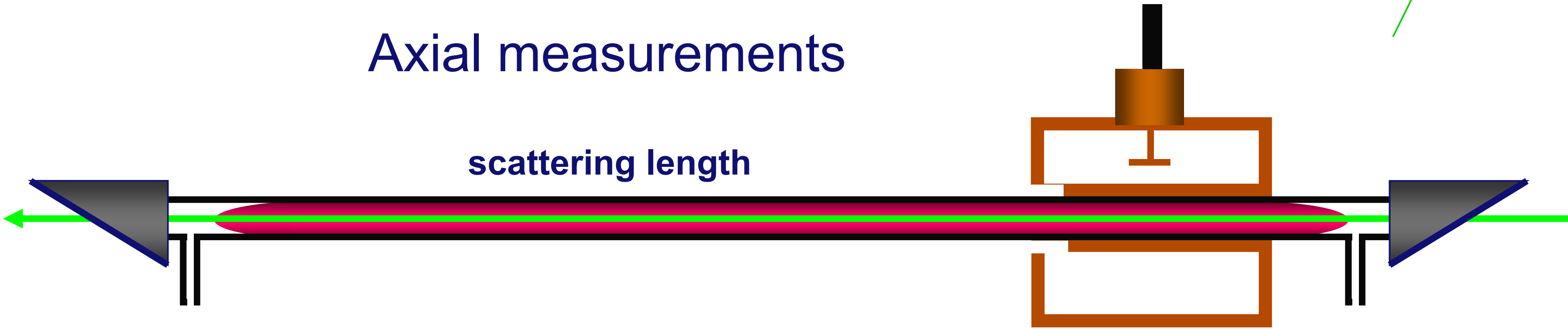
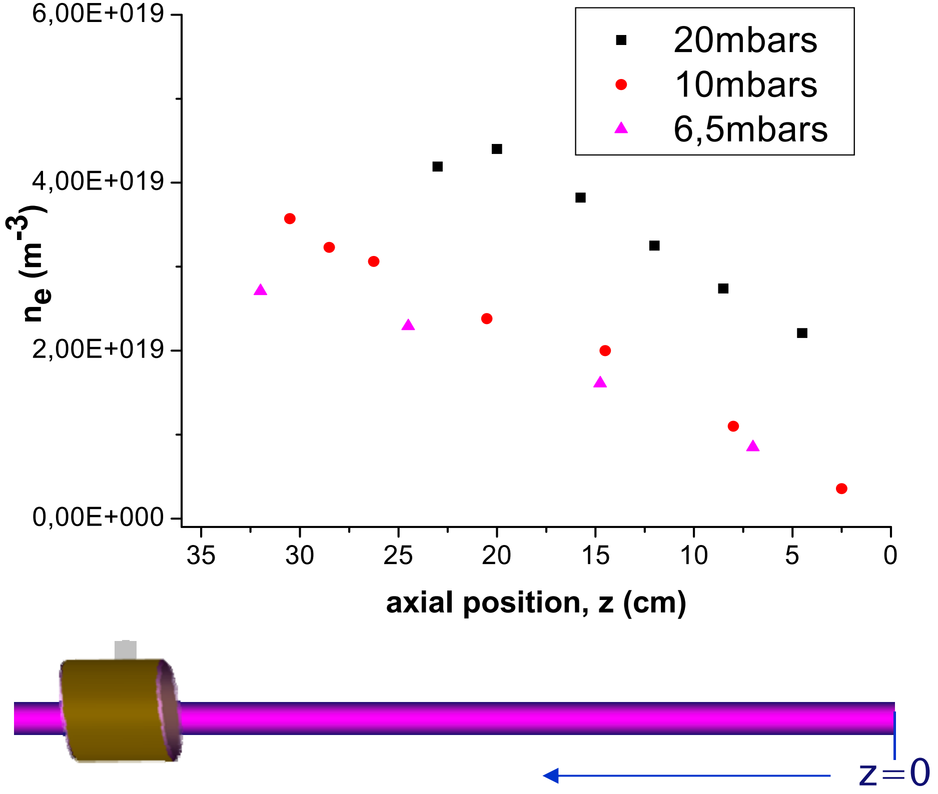
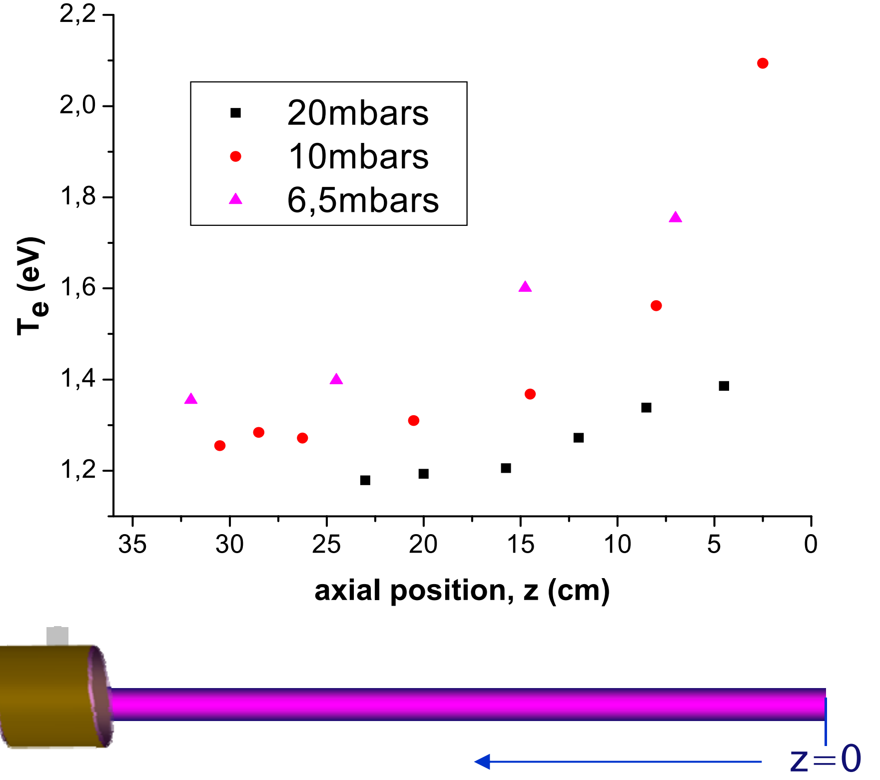
EEDF deviations

Axial measurements

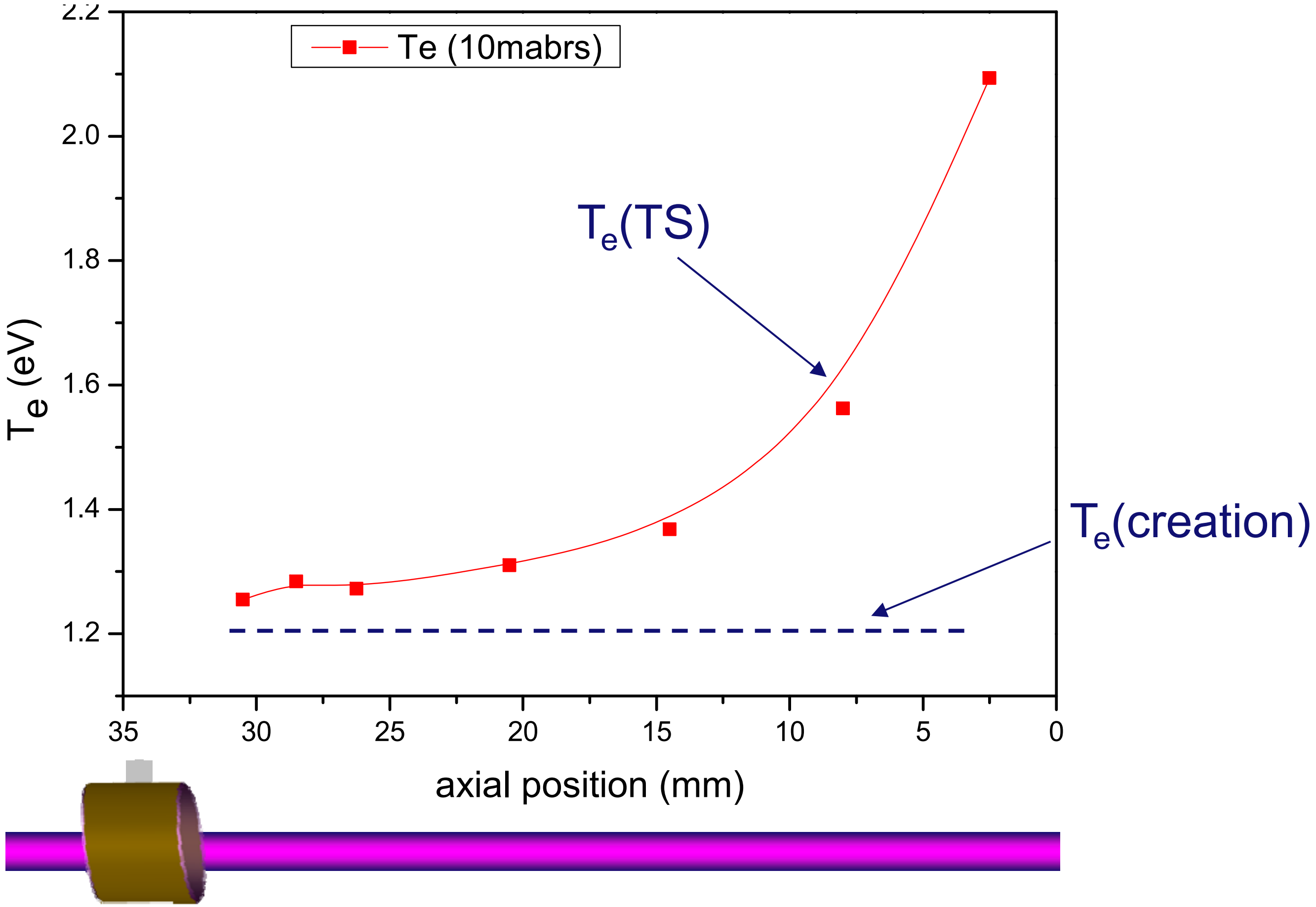






EEDF deviations

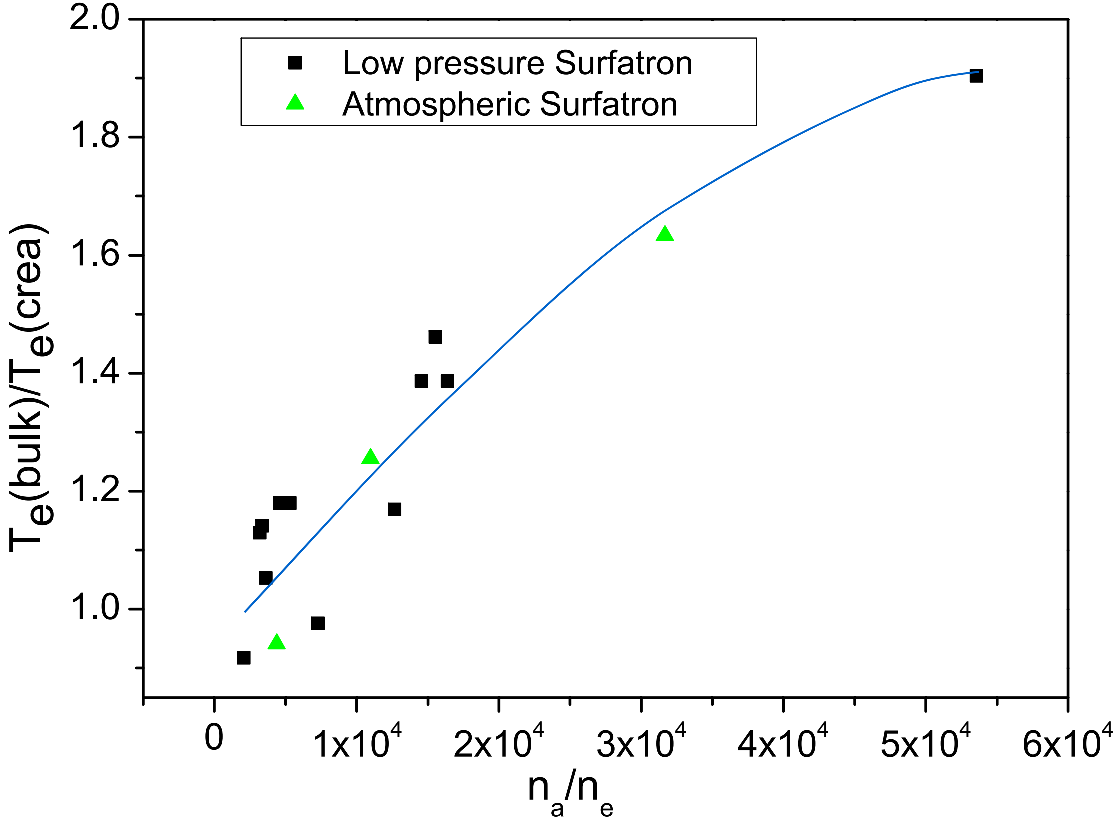
Low pressure surfatron



$\longrightarrow$ $n_e \downarrow$ $\longrightarrow$

$\longrightarrow$ $n_e/n_a \downarrow$ $\longrightarrow$

EEDF deviations



$$T_e(TS)/T_e(creation) = f(n_e/n_a)$$

Conclusions

Laser scattering on CAPs

- Precise measurements of T_e, $n_{e,}$ T_g
- Spectral and spatial resolution (iCCD)

Challenges:

Stray light

- It can affect the detection limit
- Its rejection is fundamental for Rayliegh scattering

Laser perturbation

- electron-atom interactions are important in CAPs

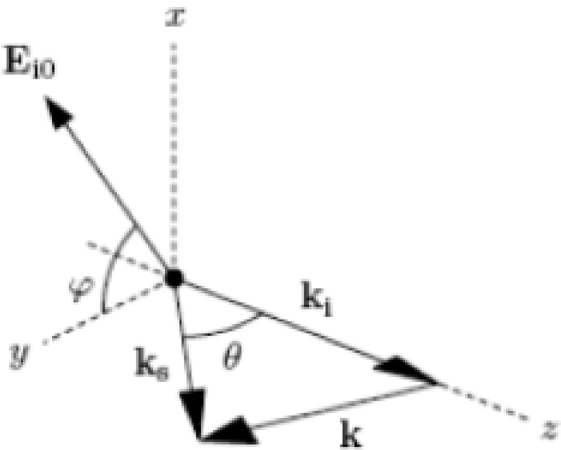
Maxwell deviations

- Thomson scattering provides T_e(bulk)
- Deviations can appear on the EEDF tail

Thanks for your attention

$$\frac{d\sigma_T}{d\Omega} = \frac{1}{S}\frac{dP_s}{d\Omega} = \mathrm{r}_e^2(1 - sen^2(\theta)\cos^2(\varphi))^2$$

$$\sigma_T = \frac{8\pi r_e^2}{3} = 6.65 \cdot 10^{-29} m^2$$



$$w_s = (w_i - \mathbf{k}_i\mathbf{v}) + \mathbf{k}_s\mathbf{v},$$

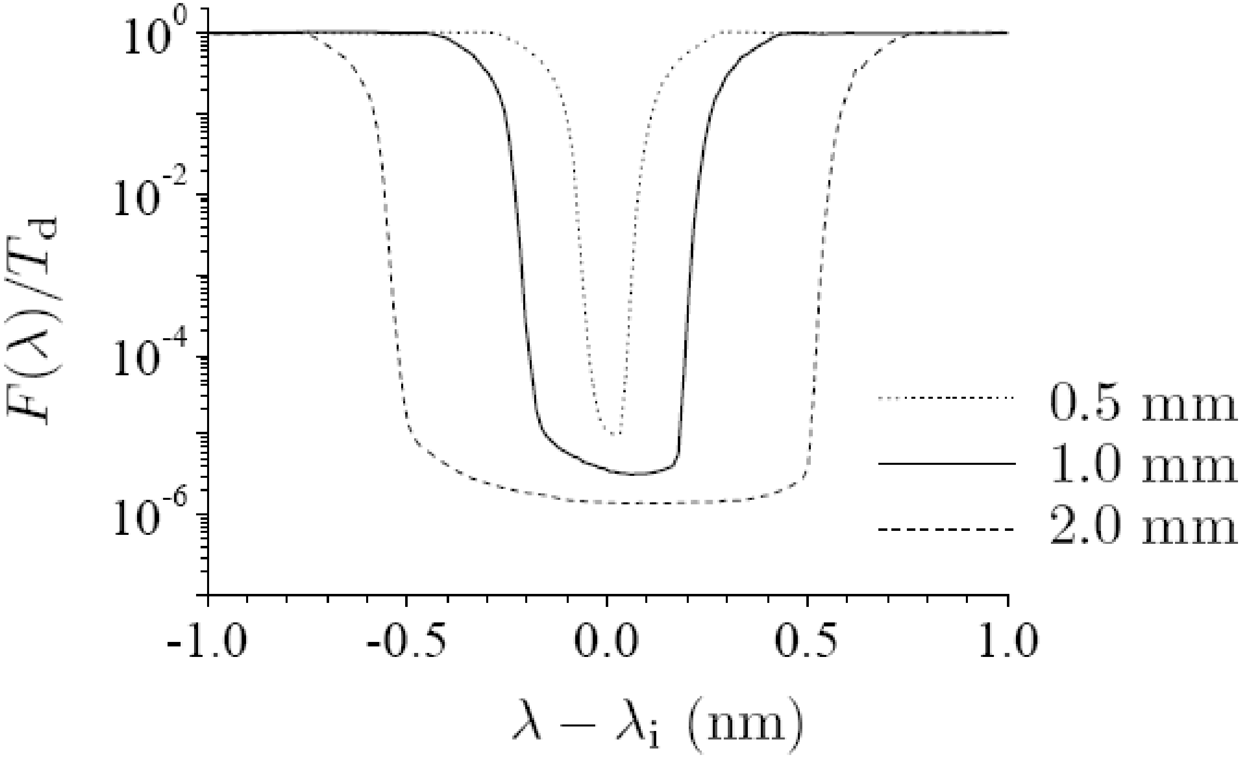
$$\Delta\omega = \omega_s - \omega_i = \mathbf{kv}$$

$$k = |\mathbf{k}_s - \mathbf{k}_i| \approx 2 \cdot k_i \cdot sen(\theta/2)$$

$$\Delta\omega/\omega_i = 2sen(\theta/2) \cdot v_k/c$$

$$\frac{\Delta T_e}{T_e} \approx 3.38 \cdot 10^{-39} \frac{n_i Z^2}{(k_B T_e)^{3/2}} \lambda_i^3 \left\{1 - \exp\left(-\frac{h\nu_i}{k_B T_e}\right)\right\} Q_i$$

Instrumental parameters		Single spectrograph characteristics	
grating constant	$n = 1800$ grooves/mm	Bandpass	$\Delta\lambda_{\mathrm{bp}} = 0.22$ nm
angle of incidence	$\alpha = 15°$	Dispersion	$d = 1.52$ mm/nm
angle of diffraction	$\beta = 45°$	Solid angle	$\Delta\Omega = 0.0197$ sr ($f/6.3$)
focal length	$f = 600$ mm		
lens diameter	$\phi = 95$ mm		
collimated beam	$a = 600$ mm		
slit width	$s_{\mathrm{ent}} = 250\ \mu$m		



Low efficiency

cross section: $6.65\ 10^{-29}\ m^2$

$n_e \sim 10^{19} m^{-3}$

Scattering length ~ 10^{-2} m

→ Scatter fraction $\tau_s \sim 10^{-11}$

fraction collected; → $\tau_c \sim 10^{-3}$

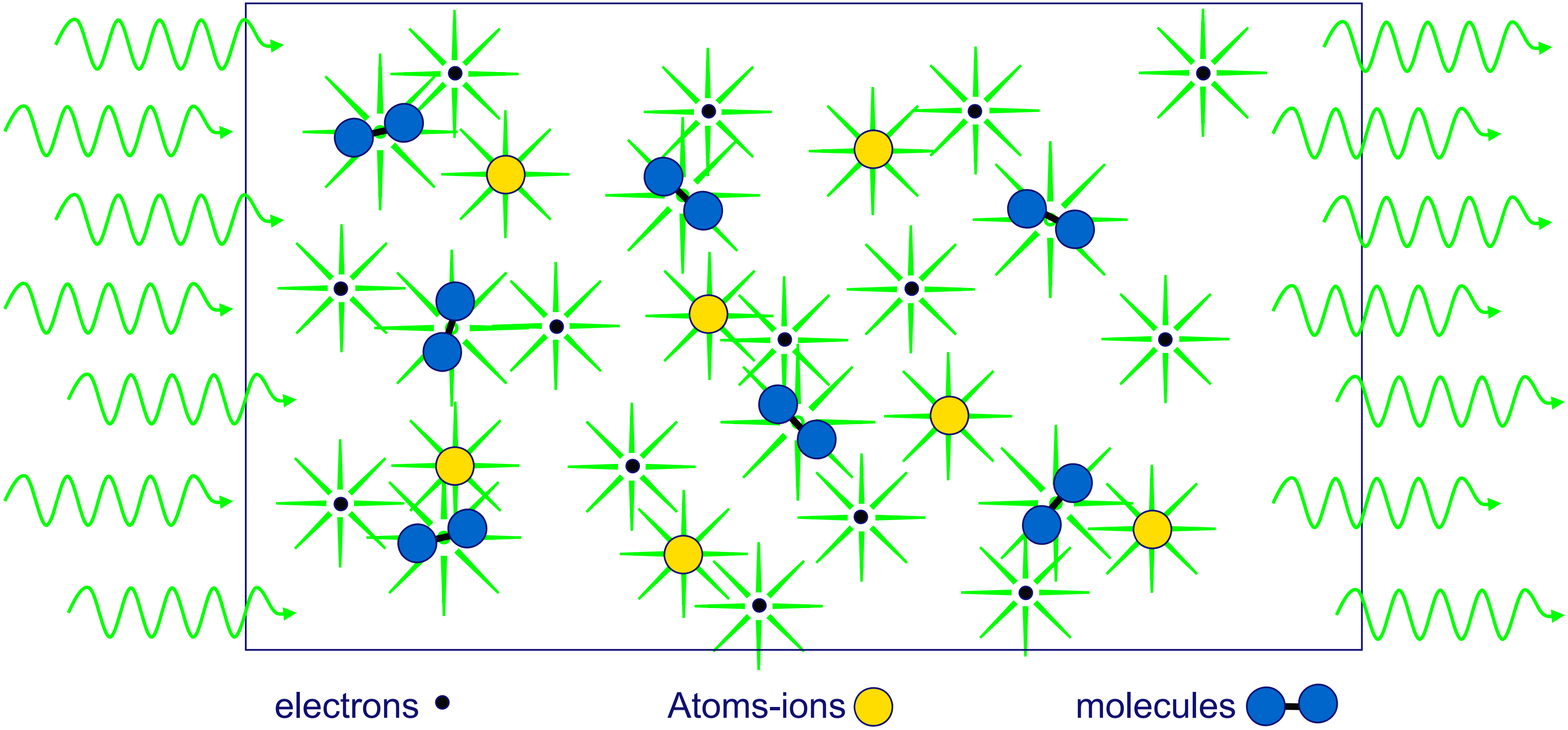
fraction detected (optics + iCCD); → $\tau_D \sim 10^{-2}$

Total fraction detected; $\tau_s \cdot \tau_c \cdot \tau_c \sim$ **10^{-16}-10^{-15}!!!**

1mJ (532nm)=$2{,}6 \cdot 10^{15}$ photons

C-Precision II laser	**Edgewave laser**
~150 mJ/pulse	~5 mJ/pulse
~10 ns	~10 ns
10Hz	5000Hz
1,5 W	**~25 W**

Laser scattering



Rayleigh and Raman Scattering

Scattering by bound electrons

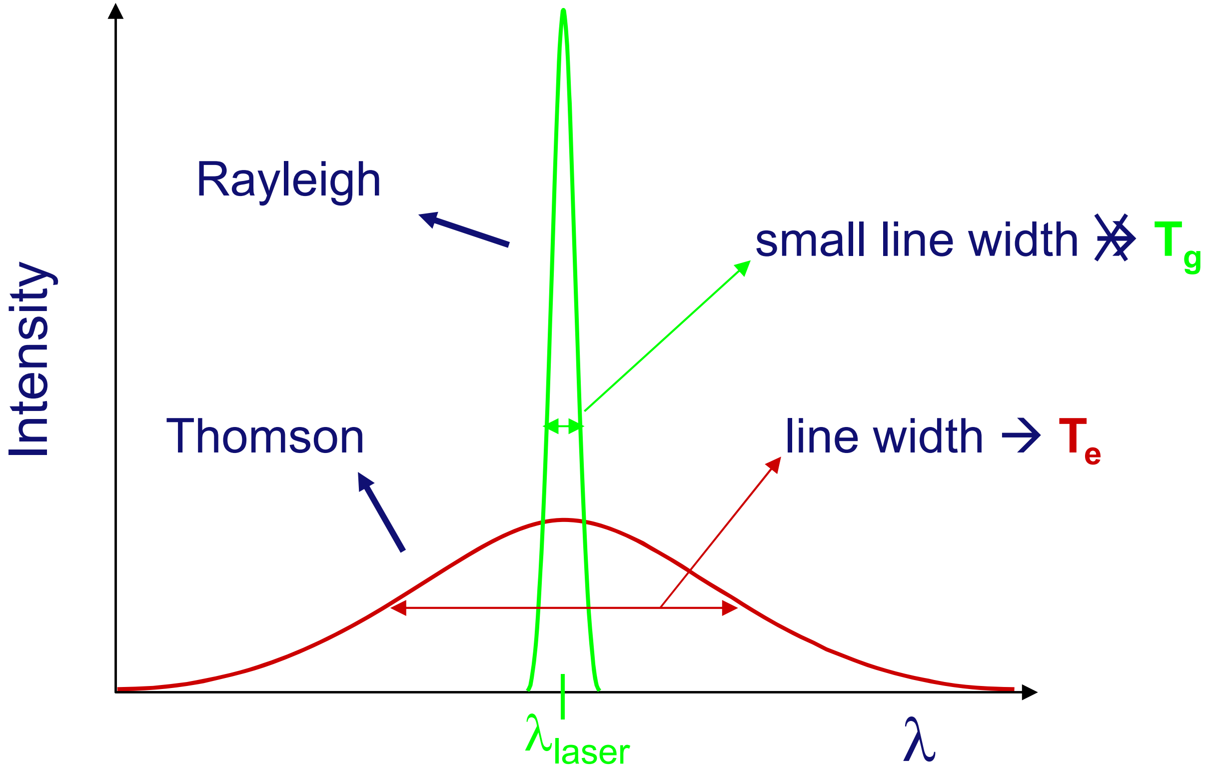
Thomson Scattering

Scattering by free electrons

Laser scattering

Thomson and Rayleigh elastic scattering: $\lambda_{laser} == \lambda_{scattering}$

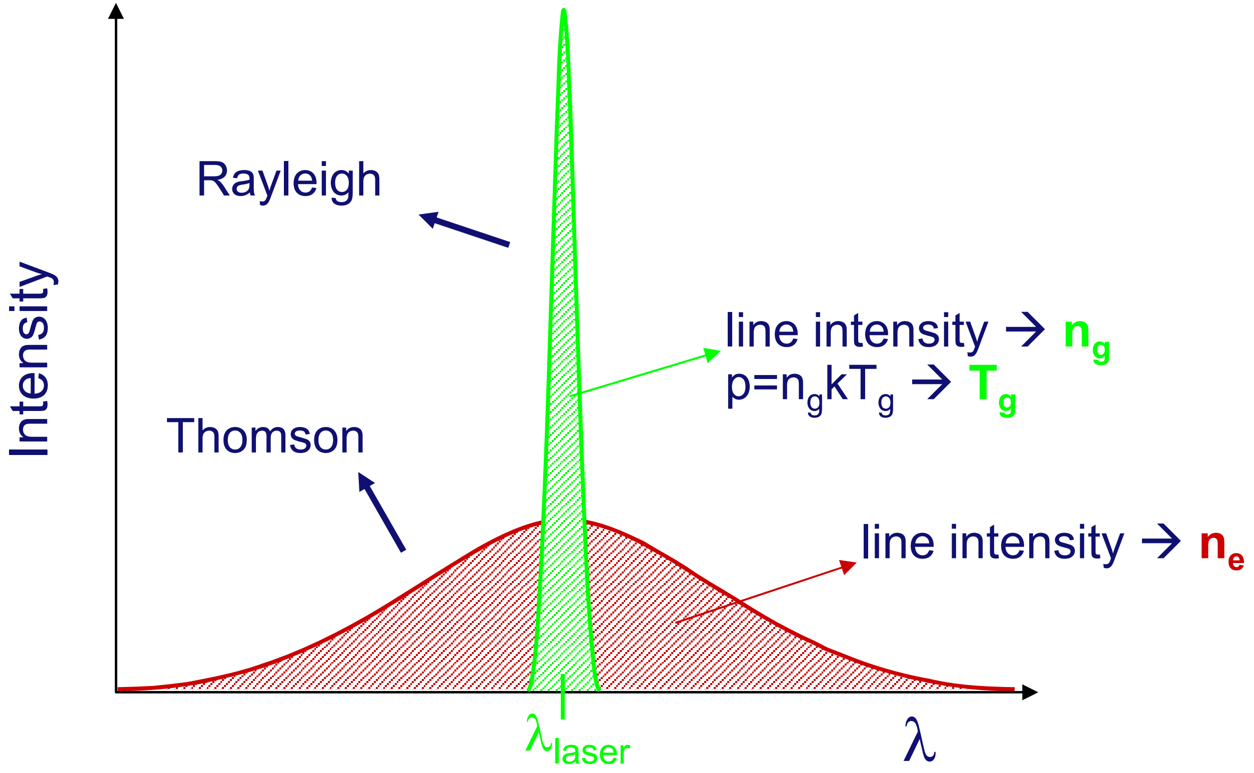
Doppler broadening → temperature



Laser scattering

Elastic scattering: $\lambda_{laser} == \lambda_{scattering}$

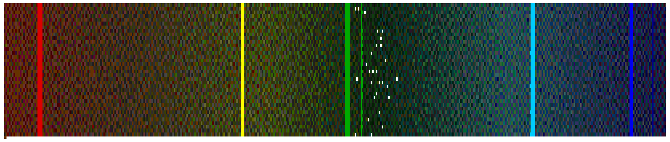
Intensity scattered → particle density



Absolute intensity measurements (ALI)

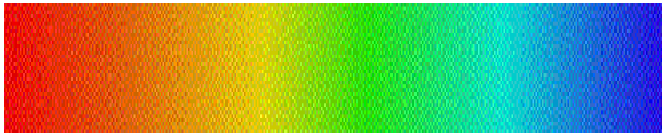
Emission spectrum

line radiation



used to determine T_e

continuum radiation



used to determine n_e

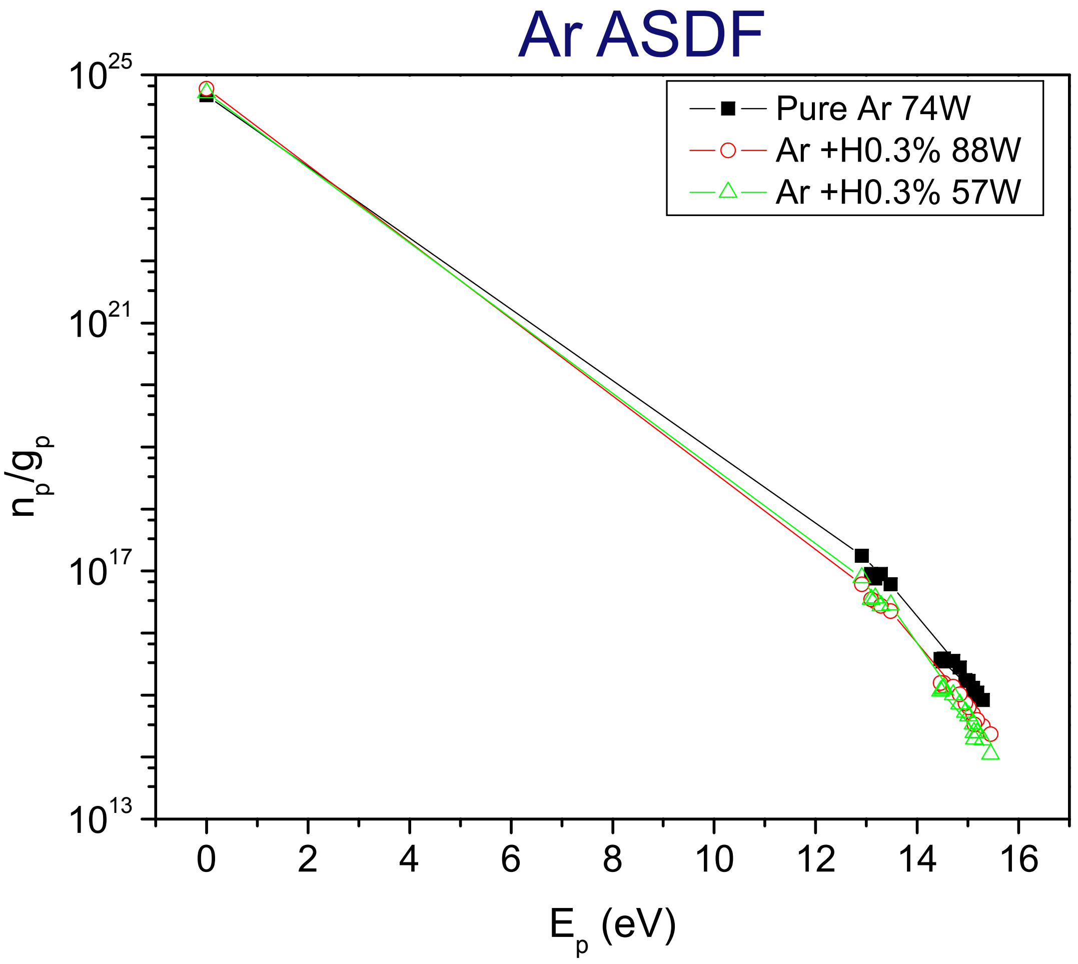
Absolute continuum intensity (ACI)

(free electrons)

Total continuum emission coefficient $j^{total}_{cont} = j^{ea}_{ff} + j^{ei}_{fb} + j^{ei}_{ff}$

$$j^{ea}_{ff} = c_2 \frac{n_e n_a}{\lambda^2} T_e^{3/2} Q^{Ar}(T_e)\left(1 + \left(1 + \frac{hc}{\lambda k T_e}\right)^2\right)\exp\left(-\frac{hc}{\lambda k T_e}\right)$$

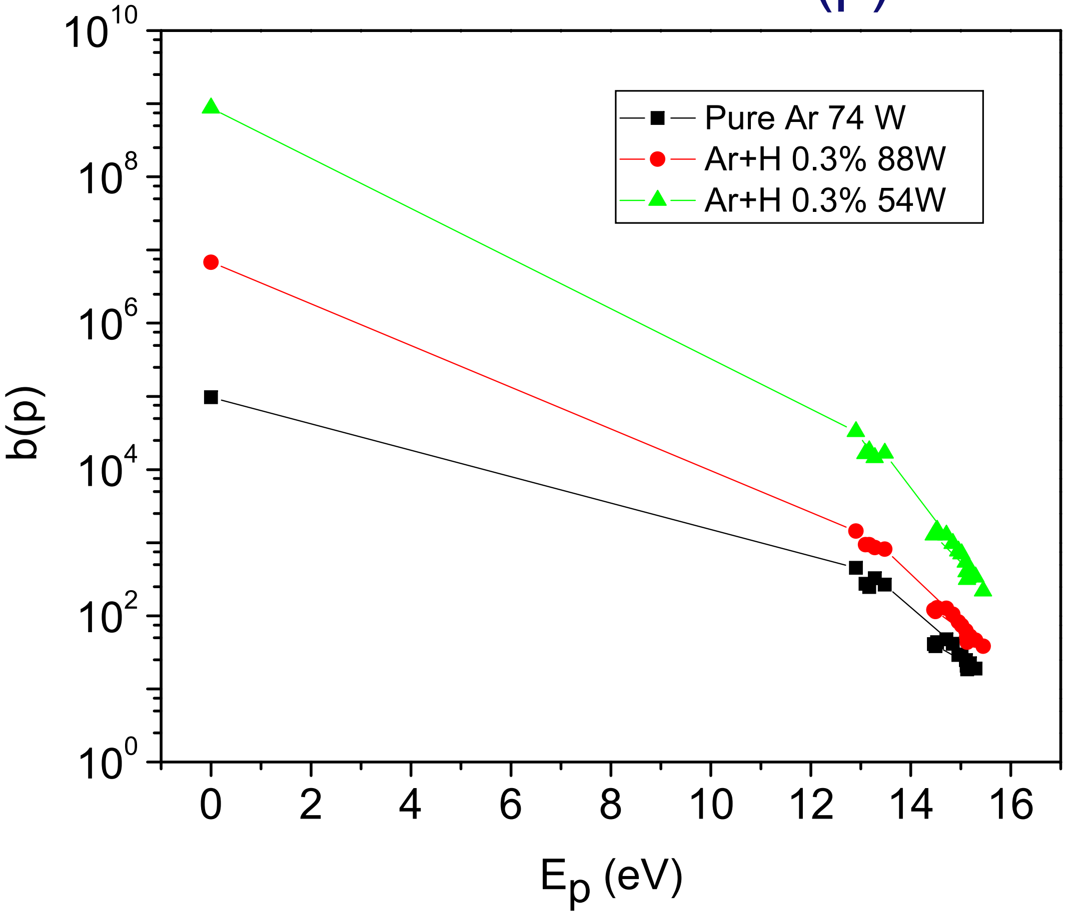
Approaching CAPs



H_2 disturb the Ar ASDF → 4p population drops

Approaching CAPs

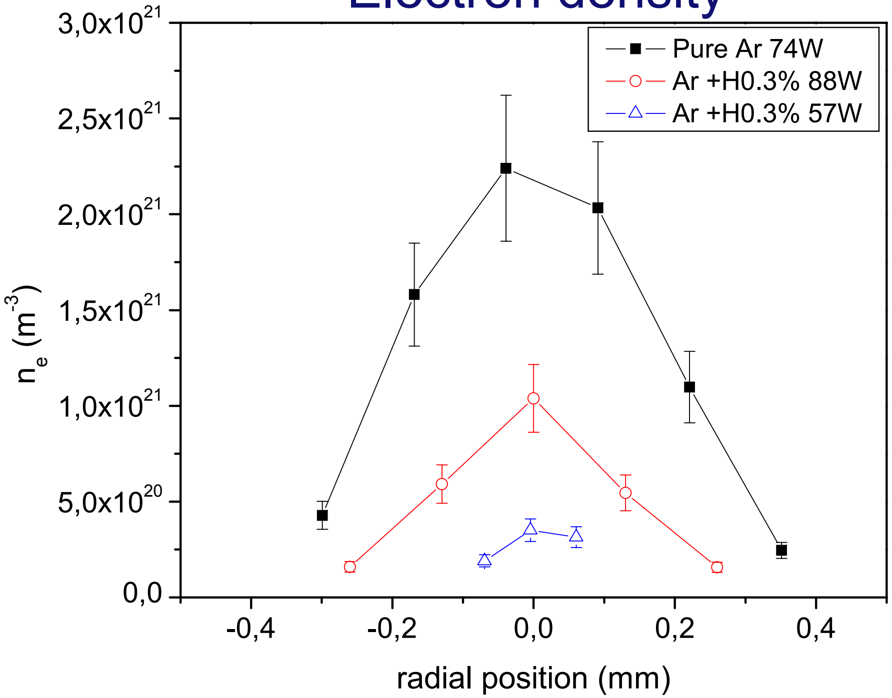
Saha factors b(p)



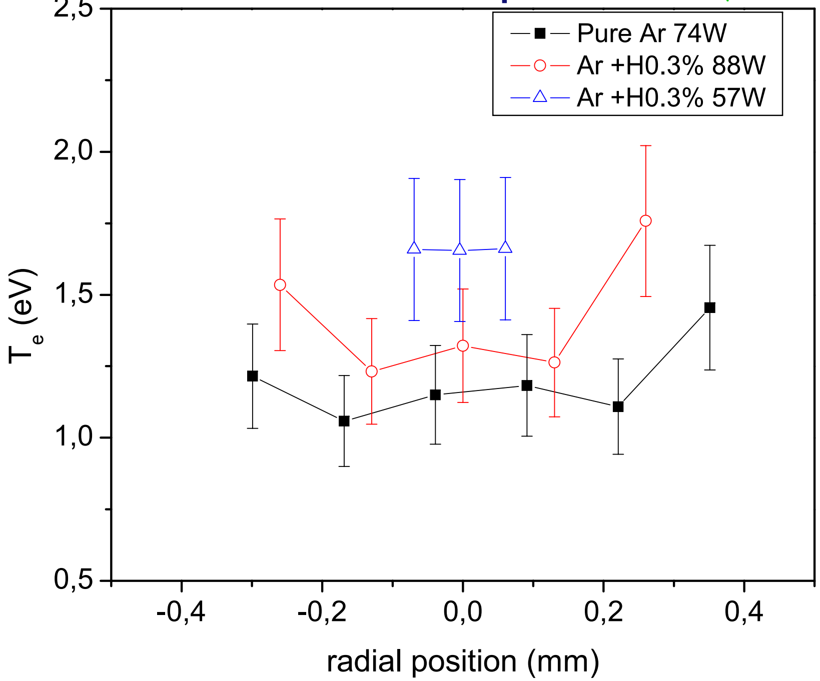
H_2 increases deviations from equilibrium

Laser scattering with iCCD

Electron density



Electron temperature

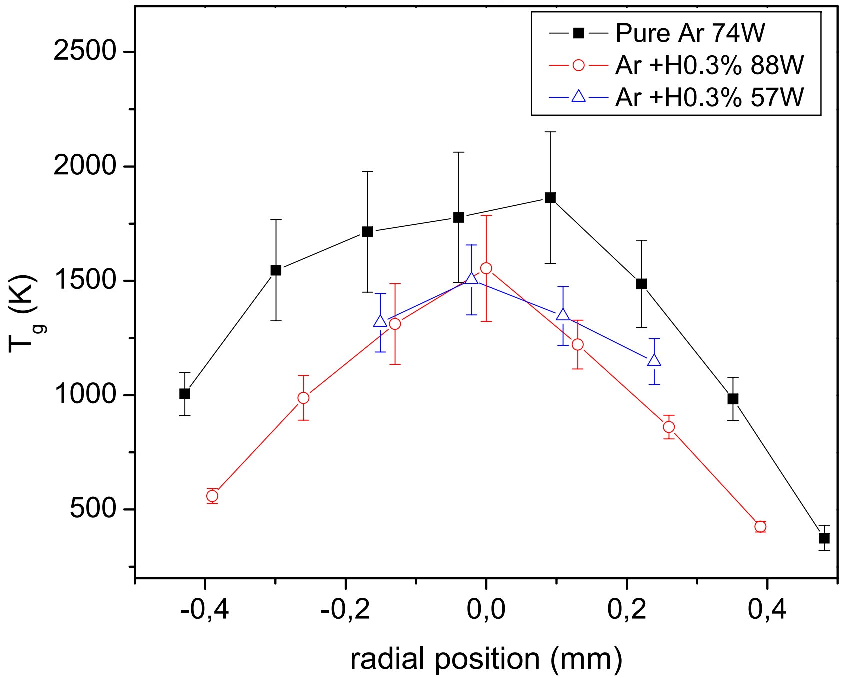


H_2 introduction

n_e drops, T_e rises: new loss channels (molecular recombination)

Laser scattering with iCCD

Gas temperature



T_g shows wider profiles

H_2 introduction → T_g drops