Electron density measurement in atmospheric pressure micro-plasmas by optical techniques: Stark broadening of spectral lines, continuum spectra emission and Thomson scattering

N. Sadeghi

LIPHY & LTM, Université Grenoble-Alpes & CNRS Grenoble (France)

E-mail: nader.sadeghi@univ-grenoble-alpes.fr

Introduction

In atmospheric pressure plasmas, the commonly used techniques for the characterization of low pressure plasmas cannot be applied:

- Due to their small dimensions, these plasmas are perturbed by the probe

-The sheath around the Langmuir probe is strongly collisional and modeling of the V-I characteristic is not obvious.

- Resonance features of hairpin and microwave cavities are strongly broadened by e-neutral collisions.

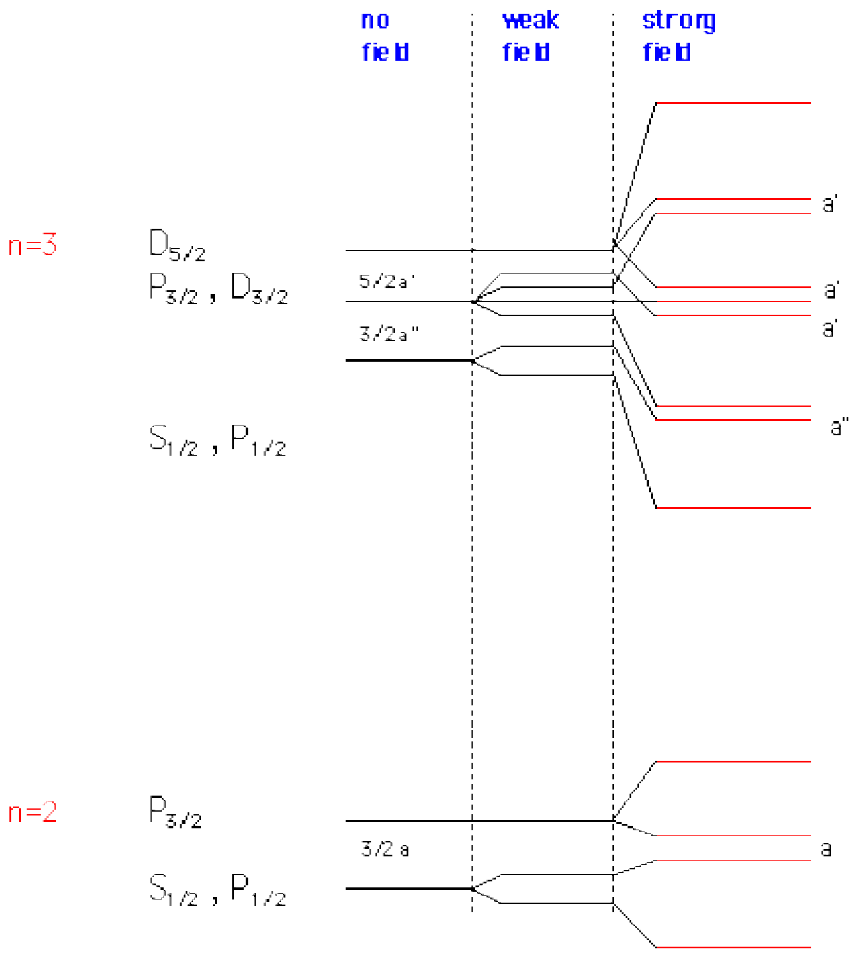
Solution: Optical techniques:

- **Broadening of spectral lines**
- **Absolute intensity of the continuum emission (Brremsstrahlung)**
- **Thomson scattering**

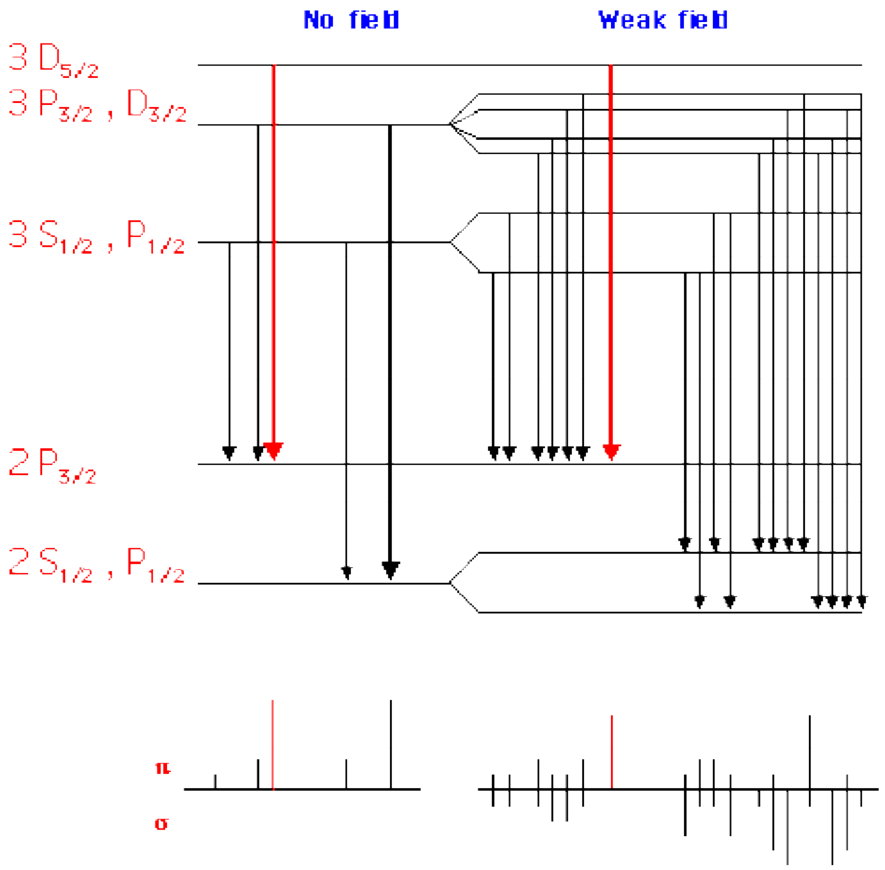
Stark Broadening of spectral lines

The Stark effect results from the splitting and displacement of atomic energy levels by and external electric field.

Stark effect of Hydrogen for n=3 and n=2



Fine structure and
Weak field Stark effect for Hydrogen Hα



Stark Broadening of spectral lines

The Stark effect results from the splitting and displacement of atomic energy levels by and external electric field.

In plasmas, electrons and ions surrounding an emitter atom generate a time varying random E-field on it, leading to the broadening of the emission (or absorption) line.

The theory of the phenomenon is well known [1] and the broadening coefficients of many atomic and ionic lines of large number of atoms are reported in [2].

Hydrogen H(β) line has the largest sensitivity and its HWHM is given by [3]:

$$\Delta\lambda_S(\mathrm{nm})=2.5\ 10^{-10}\ \alpha(n_e,T_e)\ n_e^{2/3}\ (\mathrm{cm}^{-3})$$

However, for (T_e å 1 ï 10 eV, n_e å 10^{13} ï 10^{14} cm^{-3}), $\alpha(n_e,T_e)=0.077$ *within 5%* [1]

But for other atoms than H [2], $\Delta\lambda_S$ **(FWHM) å** $K(T_e.n_e).\ n_e$

[1] P. Kepple and H. R. Griem, Phys. Rev. 173, 317 (1968)
[2] N. Konjevic, A. Lesage, J.R. Fuhr and W.L. Wiese, J. Phys. Chem. Ref. Data 31, 819 (2002)
[3] M. A. Gigosos and V. Cardenoso, J. Phys. B 29, 4795 (1996)

Pressure Broadening of spectral lines

But in atmospheric pressure plasmas, one should also consider the pressure broadening of the line, which can more than the Stark broadening .

Its theory has been widely developed (see [4]) and can be divided in two categories:

1- Resonance broadening : When the upper or lower level of the transition is optically connected to the ground sate of the perturber [5, 6],

$\Delta\lambda_R$ **åground state atoms density [N] åP/T**

2- Van der Waals broadening: When the perturber is another atom, or when the emitter levels are not optically connected to the ground state of perturber (triplet lines of He perturbed by ground state He atoms) [5, 6].

$\Delta\lambda_{vdW}$ **åground state atoms density [N].**$T^{0.3}$**å**$P/T^{0.7}$

[4] N. Allard and J. Kielkopf, Rev. Mod. Phys. 54, (1982) 1103
[5] NIST: http://physics.nist.gov/Pubs/AtSpec/node20.html
[6] G. Nayak, M.S. Simeni, J. Rosato, N. Sadeghi et P.J. Bruggeman, J. Appl. Phys. 128, 243302 (2020)

Line profile at high pressure: need for T_g determination

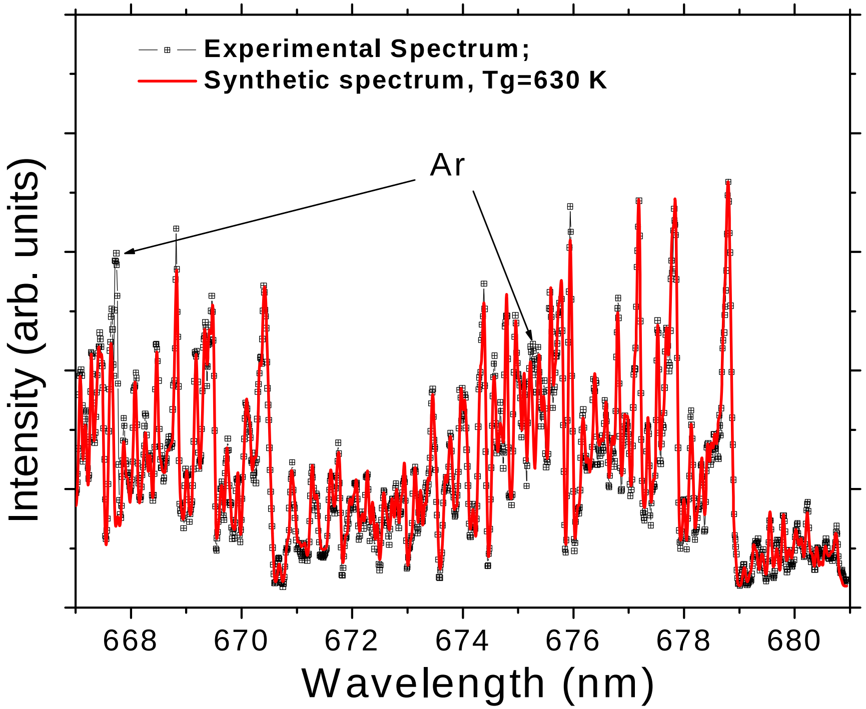
- **At atmospheric pressure, the line profile is a Voigt function with:**
  - **-its Lorentzian component** $\Delta\nu_L = \Delta\nu_{VdW} + \Delta\nu_S$
  - **-its Gaussian component** $\Delta\nu_G = (\Delta\nu_D^2 + \Delta\nu_A^2)^{1/2}$

  where $\Delta\nu_A$ is the apparatus function

But, both $\Delta\nu_L$ and $\Delta\nu_G$ depend on gas temperature

$\Delta\nu_{Doppler}$ **(FWHM) = 7.16 10^{-7} ν $(T_g/M)^{1/2}$**

$\Delta\nu_{VdW}$ **(FWHM) $\approx$ *K*. *p* $(T_g)^{-0.7}$**



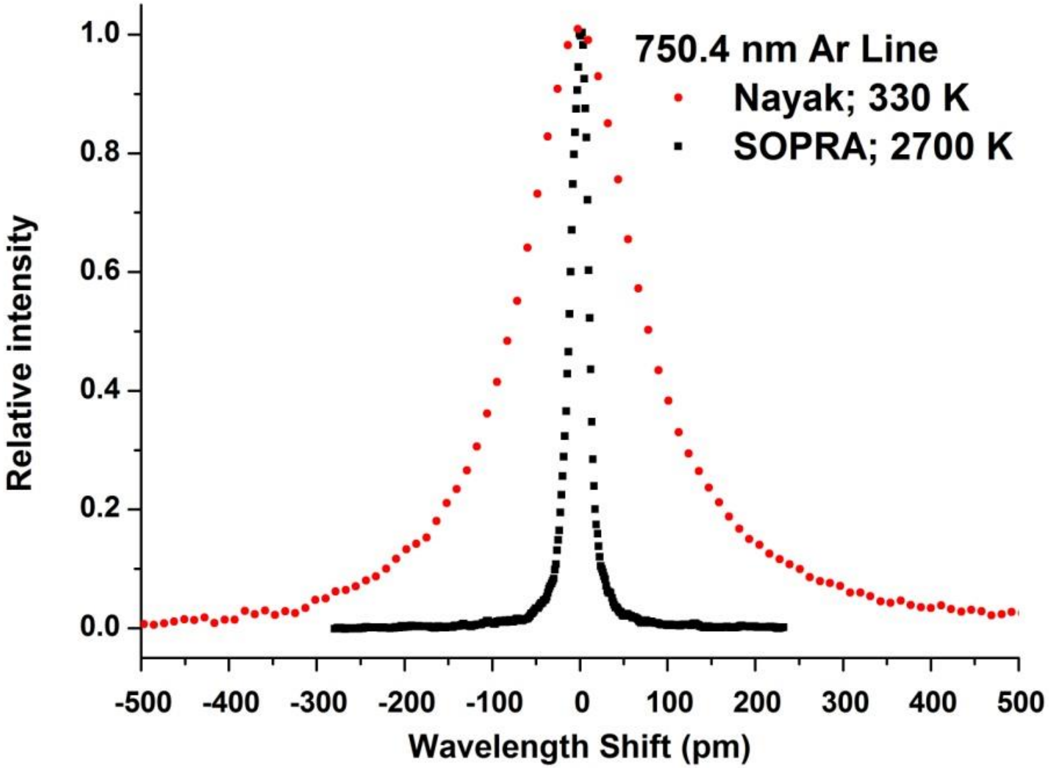
Trace of N2 was added to argon and the emission spectra was recorded

$B^3\Pi_g(v'=4\&5) \rightarrow A^3\Sigma_u^+(v''=1\&2)$ bands of nitrogen 1^+ system

T_g can also be deduced from the pressure broadening of a line for which the broadening coefficient K is well known

Line profile at high pressure: need for T_g determination

T_g deduced from the pressure broadening width of 750.4 nm resonance broadened argon line for which the broadening coefficient K has been measured



Line profile at high pressure: Need for new experimental pressure broadening coefficient

$$\Delta\nu_{\mathrm{Doppler}}\ (\mathrm{FWHM}) = 7.16\ 10^{-7}\ \nu\,(T_g/M)^{1/2}$$

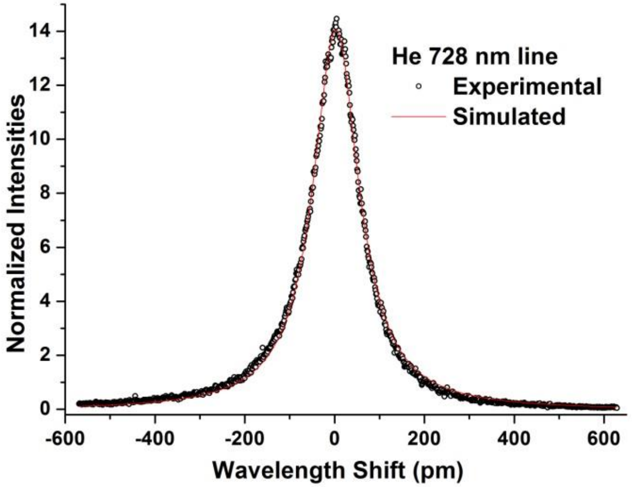
$$\Delta\nu_{\mathrm{VdW}}\ (\mathrm{FWHM})\ \text{å}\ K.\ p\ (T_g)^{-0.7}$$

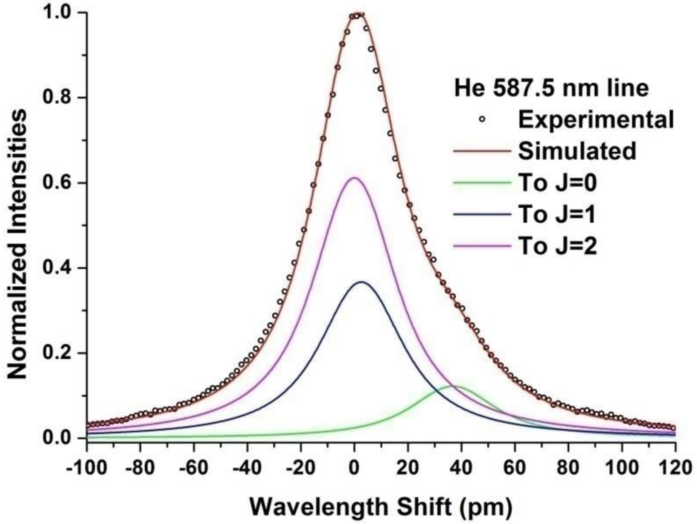
-If the correspondence between $\Delta\nu_{\mathrm{Doppler}}$ and Tg is straight, for $\Delta\nu_{\mathrm{VdW}}$ the knowledge of the broadening coefficient *K* is needed.

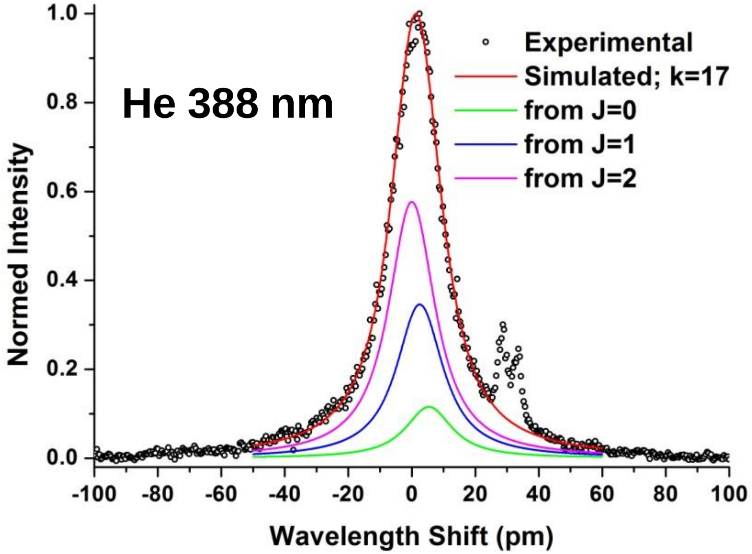
- Almost 99% of the published *K* coefficients of different spectral lines have been measured in ñThermal plasmasò, in which the Stark broadening also exists and the gas temperature is barely well defined.

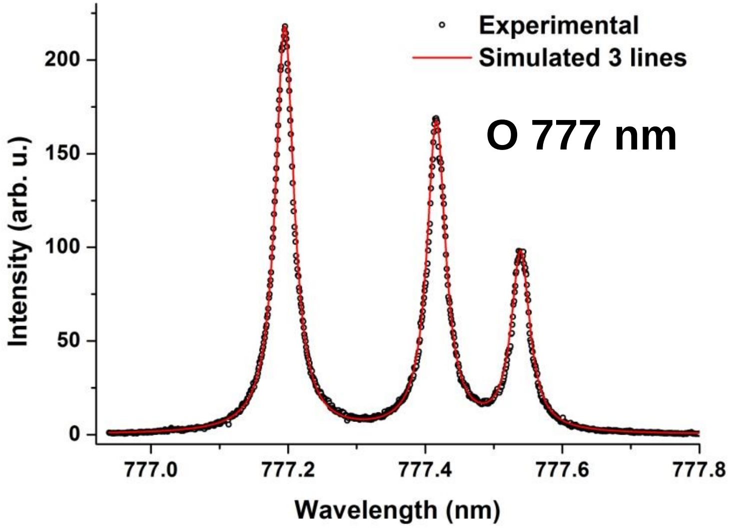
- So, to be used for the precise determination of Tg, *K* coefficients need to be revisited in better known experimental conditions.

Few examples of He and O emission line profiles recorded from 1 bar helium DBD plasma at 300 K with SOPRA spectrometer

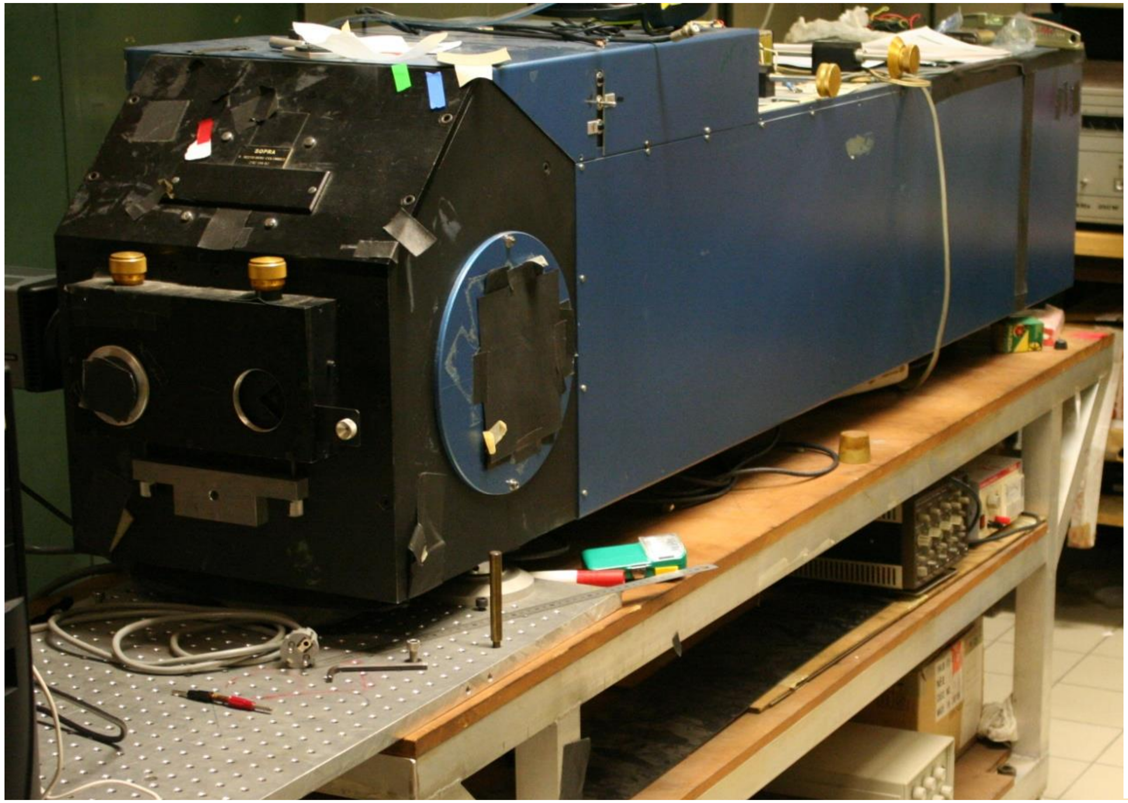








High resolution 2 m focal SOPRA spectrometer



Equipped with a 1200 groves/mm grating and working at 2nd to 5th diffraction order. When backed with a 13 µm pixels CCD camera it provides spectral resolution down to 2 pm

It is now located at LSPM (Villetaneuse) and is available to RPF members

Pressure broadened linewidths (FWHM) measured by emission spectroscopy in AP helium DBD plasma with 2 m SOPRA spectrometer

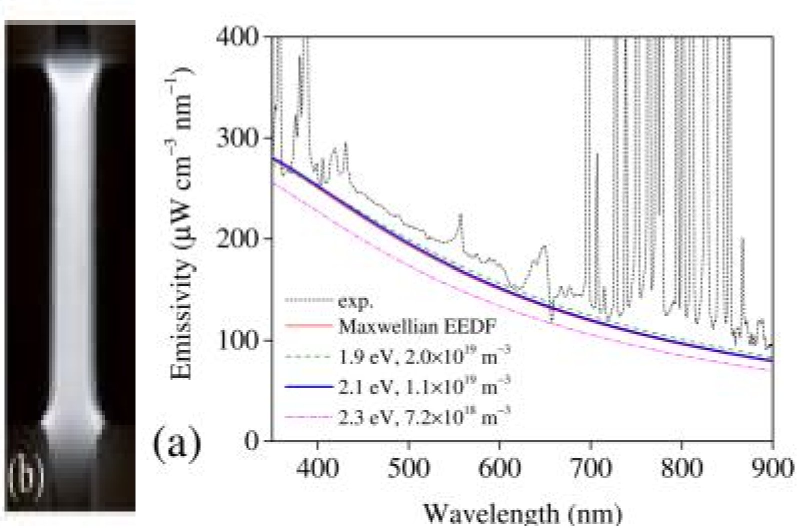
Line (nm)	Transition	Exp width (pm)	Theory vdW (pm)	Theory Resonance (pm)
He 501.5	$3\,^1P_1 - 2\,^1S_0$	33.6 ± 0.5	23.2 (27.7 GHz)	12.0 (14.4 GHz)
He 504.7	$4\,^1S_0 - 2\,^1P_1$	145 ± 25 VN	38.8 (45.6 GHz)	48.3 (56.9 GHz)
He 667.8	$3\,^1D_2 - 2\,^1P_1$	105.0 ± 0.5	34.1 (22.9 GHz)	87.4 (58.8)
He 728.1	$3\,^1S_0 - 2\,^1P_1$	123.9 ± 1.2	47.3 (26.8 GHz)	104.0 (58.8 GHz)
He 388.8	$3\,^3P_J - 2\,^3S_1$	18.5 (17.0) ± 0.6	13.6 (26.9 GHz)	
He 471.3	$4\,^3S_1 - 2\,^3P_J$	88.6 ± 4.8 VN	31.6 (42.7 GHz)	
He 587.5	$3\,^3D_J - 2\,^3P_{J'}$	44.7 (35.0) ± 0.5	26.9 (23.4 GHz)	
He 706.5	$3\,^3S_1 - 2\,^3P_J$	78.8 (65) ± 0.6	40.5 (24.3 GHz)	
O 777		(30.7) ± 0.6	21.2 (10.5 GHz)	
H 486.1		60.1 ± 0.5	45.9 (58.3 GHz)	
H 656.3		63.8 ± 0.4	45.4 (31.6 GHz)	

Absolute intensity of continuum emission (Brremsstrahlung)

In AP pure argon discharge, strong continuum radiation, visible as white color is observed.

It is produced by the deceleration of free electrons during their interaction with Ar atom.

The emissivity, $\varepsilon_{ea}(\lambda)$ is expressed as [7]:



$$\epsilon_{ea} = 4\pi \times \sqrt{\frac{2}{m_e}}\frac{n_e n_a}{\lambda^2}\frac{hc}{4\pi}\int_{h\nu}^{\infty}\sigma_{ea}^{B}(\lambda, E)\sqrt{E}f(E)\,dE,$$

n_e and n_a are electron and atom densities, *E* and *f(E)* are the energy and EEDF of electrons and *ů*$^{B}_{ea}$ *(ə, E)* is the scattering cross section, given by [8] :

$$\sigma_{ea}^{B}(\lambda, E) = \frac{8\alpha}{3\pi}\frac{E}{m_e c^2}\left(1 - \frac{hc}{2\lambda E}\right)\sqrt{1 - \frac{hc}{\lambda E}}\,\sigma_{ea}^{mom}(E),$$

Where ů$^{mom}_{ea}$ (E) is the cross section for momentum transfer and $\alpha = \dfrac{e^2}{2\epsilon_0 hc}$

With these equations the density and energy of electrons can be deduced.

[6] G. Nayak, M.S. Simeni, J. Rosato, N. Sadeghi et P.J. Bruggeman, J. Appl. Phys. 128, 243302 (2020)
[7] K. T. A. L. Burm, Plasma Sources Sci. Technol. 13, 387 (2004)
[8] V. Kasyanov and A. Starostin, Sov. Phys. JETP 21, 15 (1965)

Absolute intensity of continuum emission -2

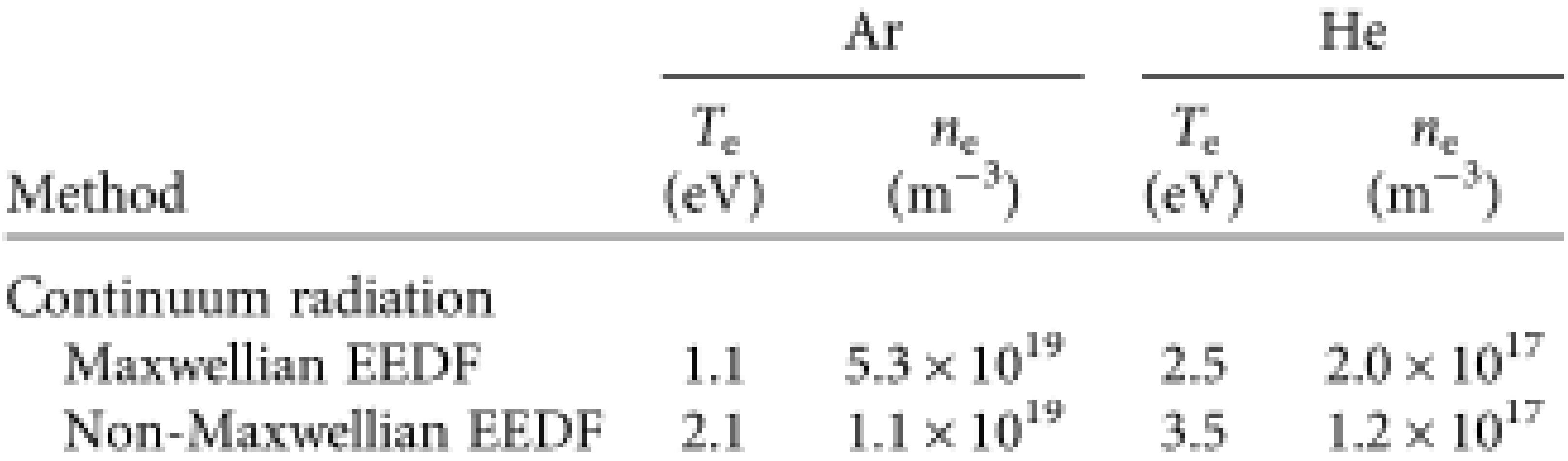
Requirement:

- A low spectral resolution, broadband spectrometer to record λ from 300 to 900 nm (Avantes 2048, with a spectral resolution of 0.6 nm in [6]).

-Perfect spectral and intensity calibration of the detection system from source to detector (using a blackbody source).

-This absolute intensity calibration ,combined with the needed assumption on the shape of EEDF, introduce large uncertainty ondetermined values.

-Table from [6] on deduced ne and Te in Ar and He radiofrequency plasmas



	Ar		He	
Method	T_e (eV)	n_e (m^{-3})	T_e (eV)	n_e (m^{-3})
Continuum radiation				
Maxwellian EEDF	1.1	5.3×10^{19}	2.5	2.0×10^{17}
Non-Maxwellian EEDF	2.1	1.1×10^{19}	3.5	1.2×10^{17}

Thomson Scattering

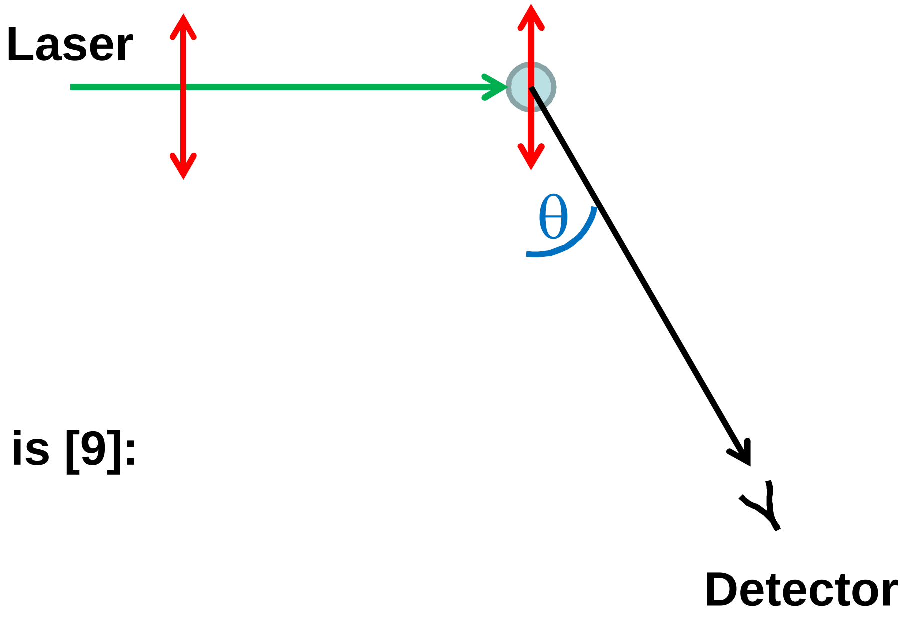
Under the Lorentz force of the E-field of the laser beam, an electron of the plasma oscillates and can emit a photon, whose wavelength is Doppler shifted by

$$\Delta\lambda=\lambda_0.v/c,$$

V being its velocity component along the observation axis.

The differential scattering cross-section of the radiation on electrons is:

$$\frac{d\sigma}{d\Omega}=\left(\frac{e^2}{4\pi\epsilon_0 m_e c^2}\right)^2 Sin\theta^2$$



Where θ is the angle between the laser polarization and the observation direction.

The total Thomson scattering cross-section is [9]:

$$\sigma_T=\frac{8\pi}{3}\cdot\left(\frac{e^2}{4\pi\varepsilon_0 m_e c^2}\right)$$

[9] Kempkens H and Uhlenbusch J *Plasma Sources Sci. Technol.* 9 (2000) 492
[10] Carbone E and Nijdam S *Plasma Phys. Control. Fusion* 57 (2015) 014026

Thomson Scattering-2

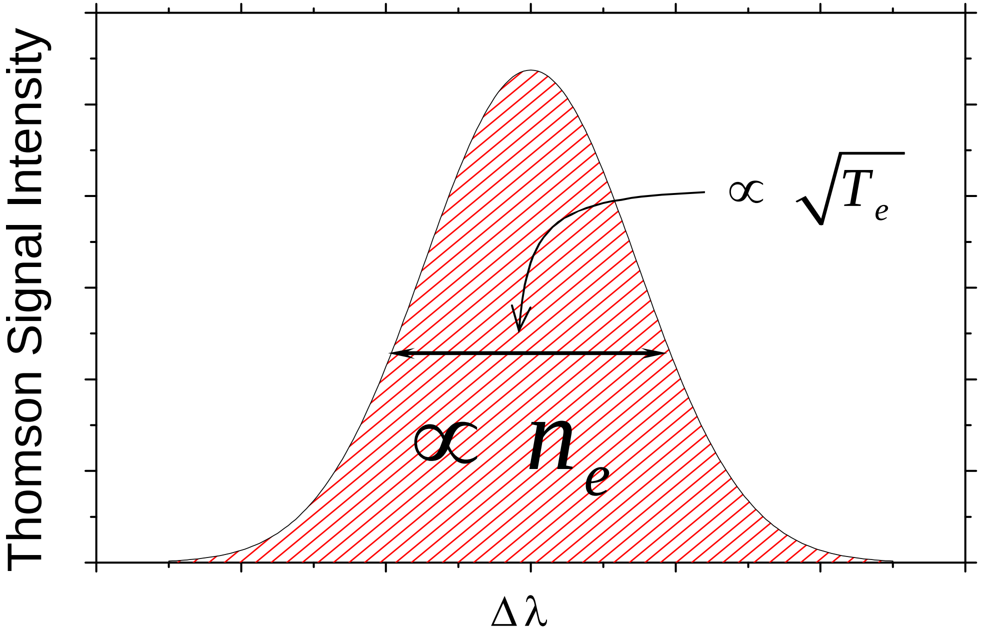
With free non-relativistic electrons the scattered electromagnetic radiation experiences a Doppler shift $\Delta\lambda/\lambda = v_e/c$

For $T_e = 3$ eV $\Rightarrow$ $v_e \sim 10^8$ cm/s then $\Delta\lambda/\lambda \sim 1/300$

For Maxwellian EEDF

$$f(\Delta\lambda) = \frac{2}{\Delta\lambda_{1/2}} \cdot \sqrt{\frac{\ln 2}{\pi}} \cdot \exp\left(-4 \cdot \ln 2 \cdot \left(\frac{\Delta\lambda}{\Delta\lambda_{1/2}} \right)^2 \right)$$

$$\Delta\lambda_{1/2} = \lambda_i \cdot 4 \cdot \sqrt{2 \cdot \ln 2 \cdot \frac{T_e}{m_e \cdot c^2}}$$



Thomson Scattering-2

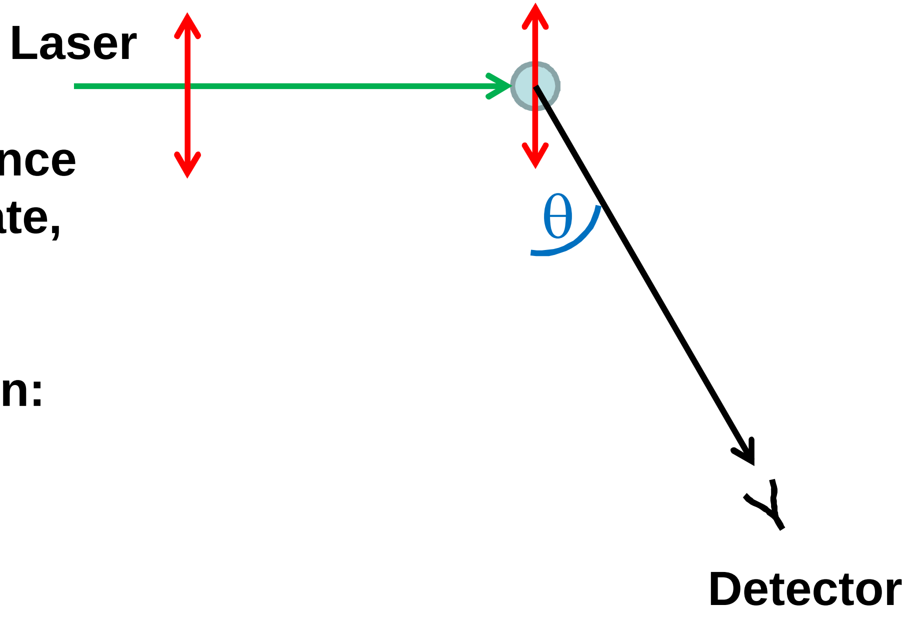
But in atmospheric pressure plasmas, one should also deal with the light scattered by atoms (Rayleigh scattering) and also the stray laser light which often are orders of magnitude stronger than the Thomson signal.

Rayleigh Scattering

Similarly, a bound electron of an atom diffuses the laser light (Rayleigh Scattering), but with negligible frequency shift. Corresponding cross-section is:

$$\sigma_R = \sigma_T \left(\frac{\omega^2}{(\omega_0^2 - \omega^2)^2 + (\gamma_0 \omega)^2} \right)^2$$

$\Delta\lambda/\lambda = v_a/c$



In which ω_0 *is* the frequency of the resonance transition of atom and γ_0 *is* the damping rate, *related to its polarizability* α.

For $\omega < \omega_{0}$, σ_R can also be defined by relation:

$$\sigma_R = \frac{128\,\pi^5}{3} \cdot \frac{\alpha^2}{\lambda^4}$$

For argon atom, $\alpha = 1.62\ 10^{-30}$ m^3 leading to

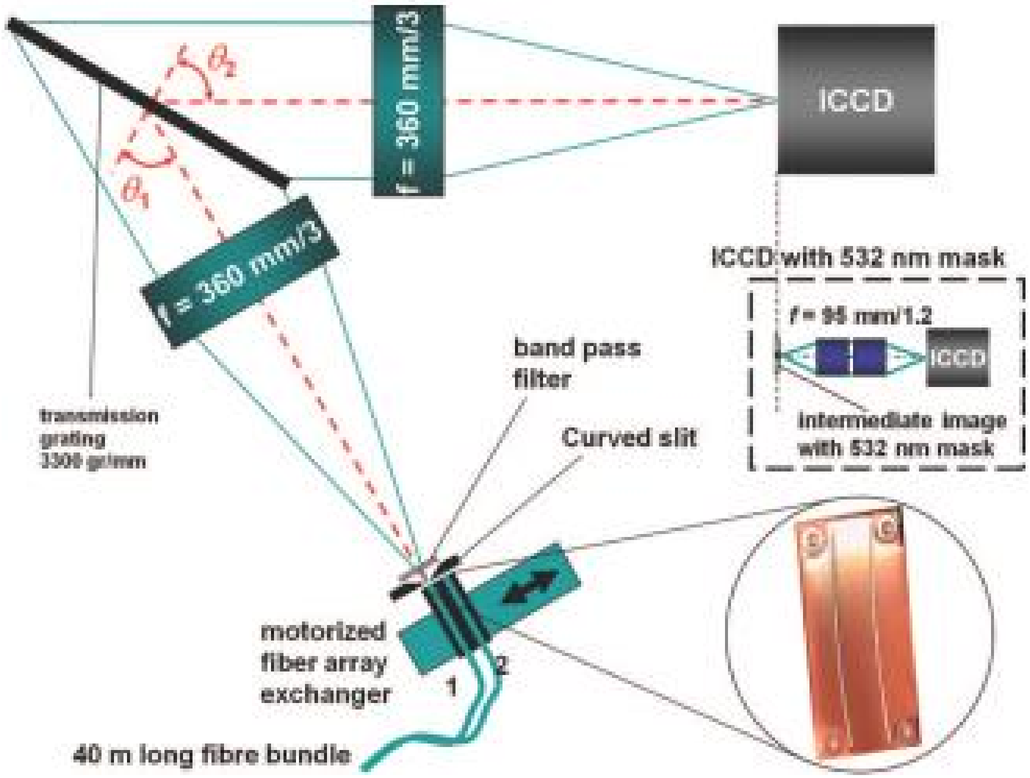
$\sigma_R = 4.28\ 10^{-31}$ m^2 **for the 532 nm Nd:Yag laser line**

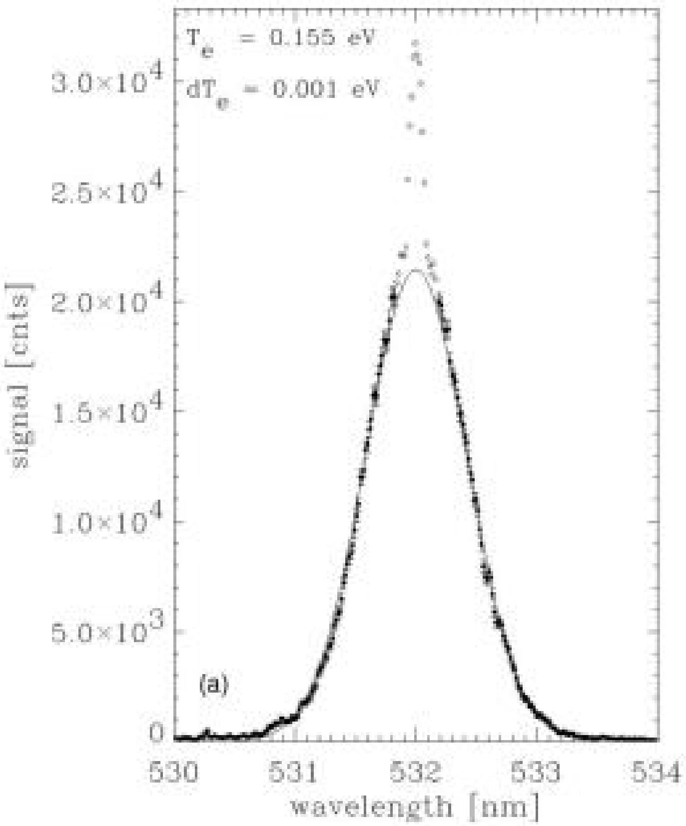
To be compared to $\sigma_T = 6.65\ 10^{-29}$ m^2

σ_R is ~100 times σ_T but the width of TS is few 1000 times larger than that of RS

Use of a single spectrometer with a transmission grating

In low pressure (1.4 Pa, [Ar]=3 10^{20} m^{-3}) argon plasma, in the expanding arc discharge chamber ([11] FOM Institute DIFFER-EURATOM)





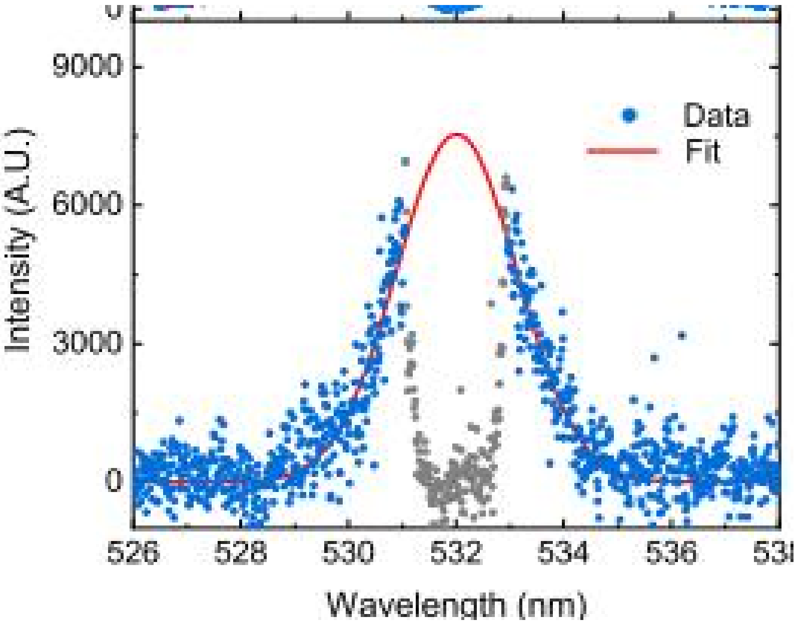
n_e= 1.2 10^{19} m^{-3}, T_e=0.155 eV

[11] van der Meiden H J et al, Rev. Sci. Instrum. 83 (2012) 123505

Use of a single spectrometer with a mask in front of CCD

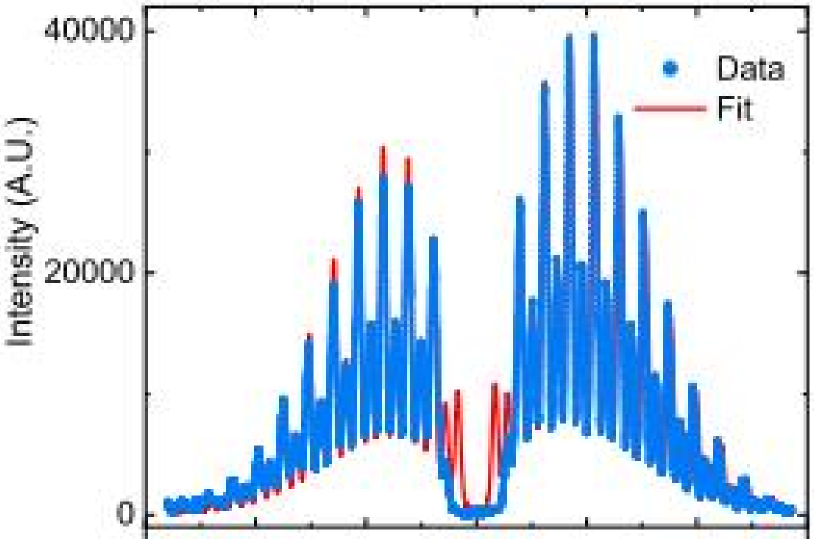
A commercial Acton 0.5 m focal; 2400 gr/mm spectrometer is used and a solid mask is placed in front of the CCD at 532 nm light position [12].

The technique was extended to 60 Torr He ([He] ~ 2 10^{24} m^{-3}). He has a σ_R ~10 times smaller than for Ar, thus 100 times smaller RS signal.



Thomson spectra at 60 Torr He

n_e= 2.2 10^{18} m^{-3}, T_e=2.5 eV

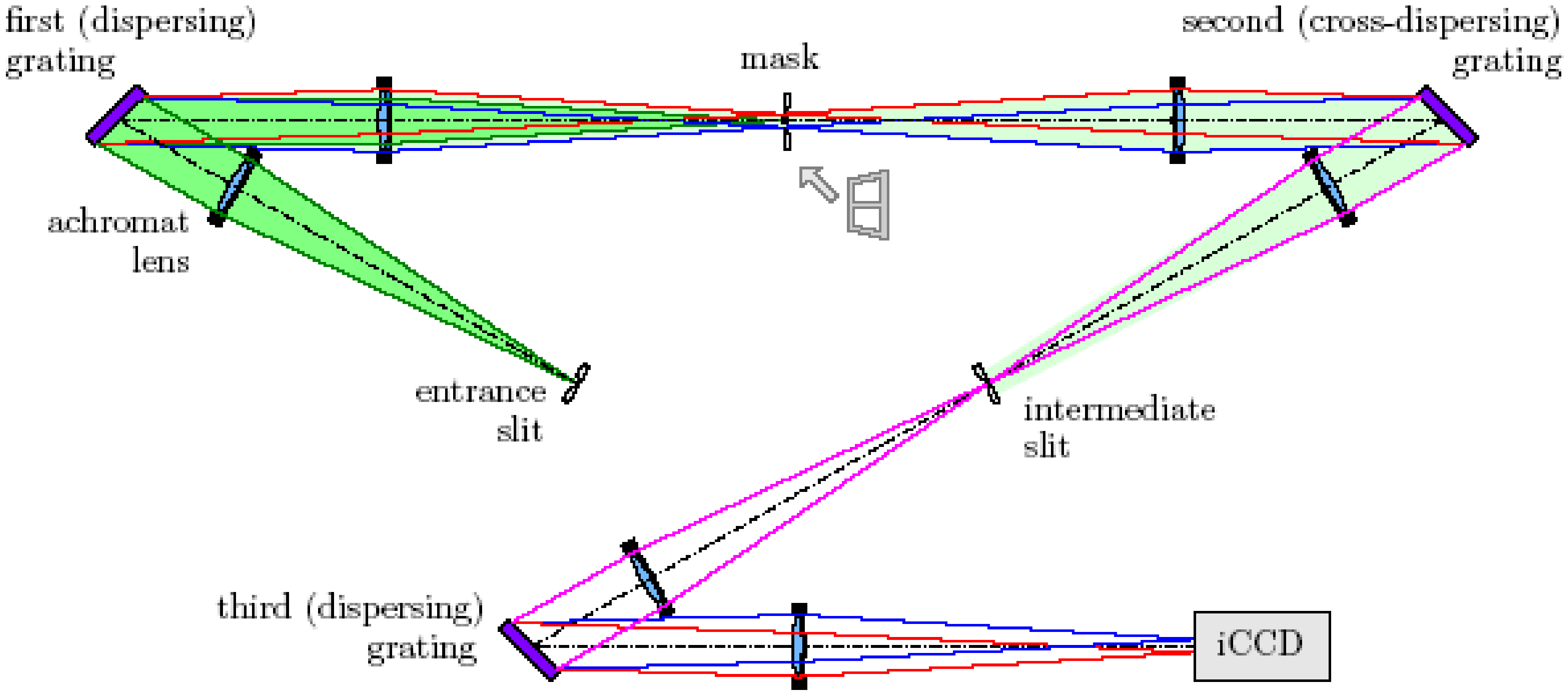


Raman spectra from 60 Torr N2 @ 300 K

[12] Timothy Y Chen et al, J. Phys. D: Appl. Phys. 52 (2019) 18LT02

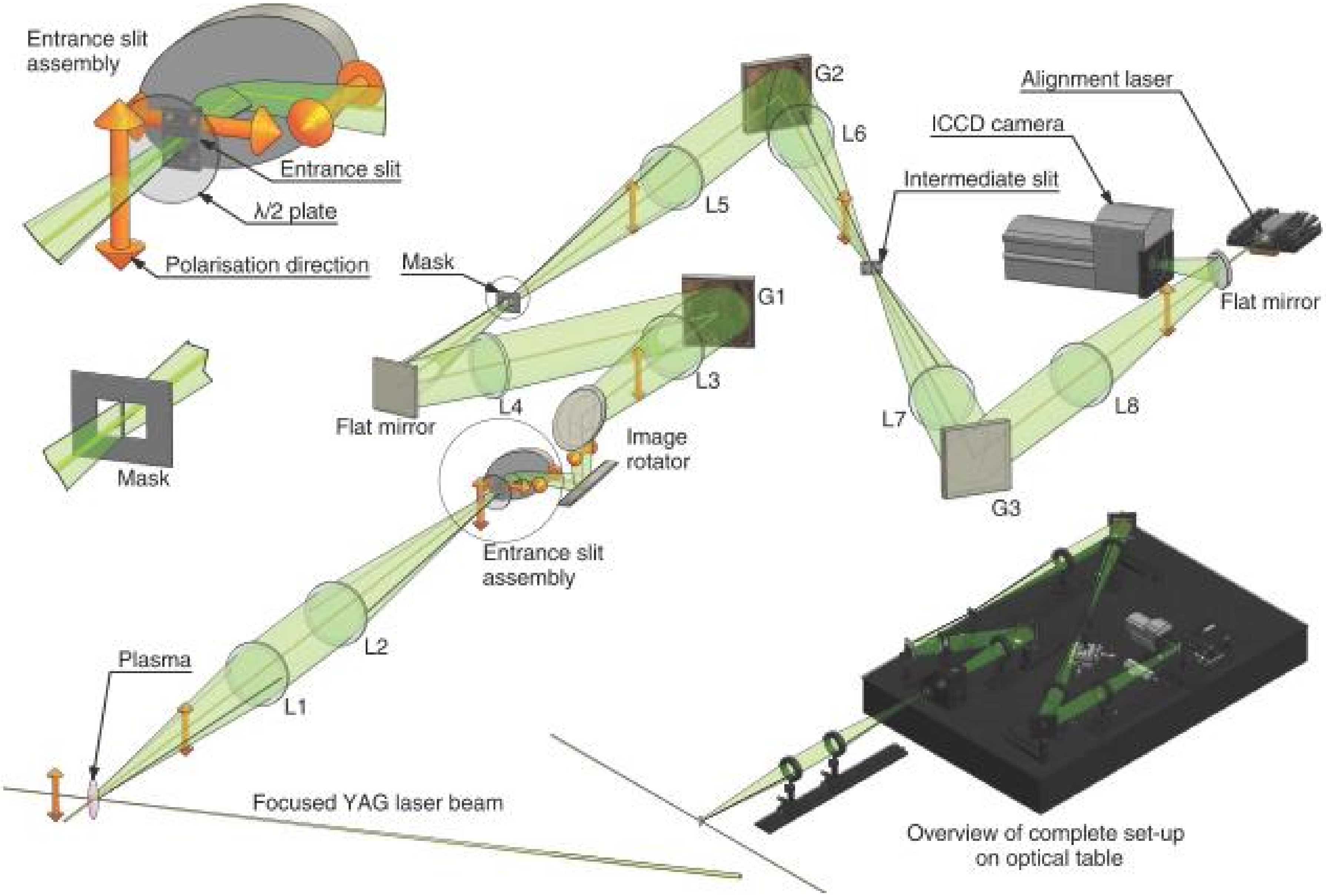
Use of a triple grating spectrometer for reducing
Rayleigh signal and scattered light at λ_0

TRIPLE GRATING SPECTROMETER



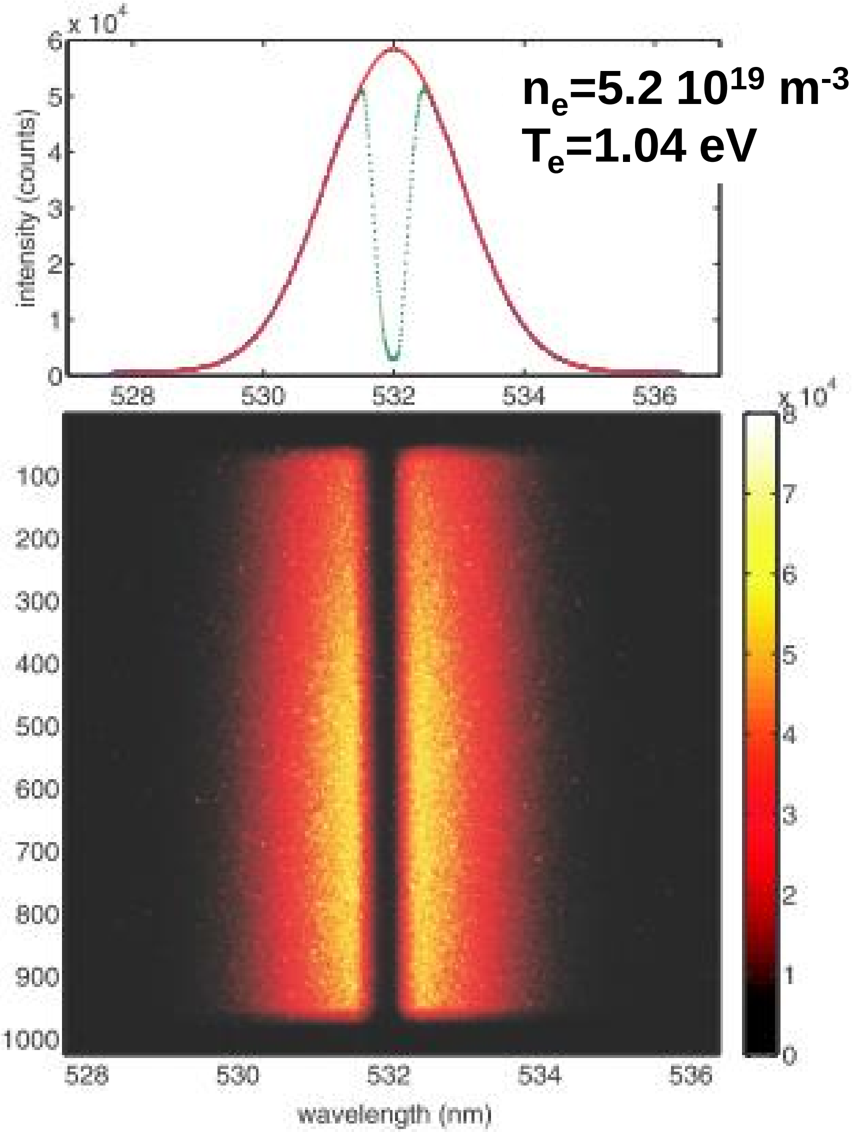
Triple gratings spectrometer for 2-D measurements

Emile Carbone and Sander Nijdam, Plasma Phys. Control. Fusion 57 (2015) 014026



Thomson and Raman spectra

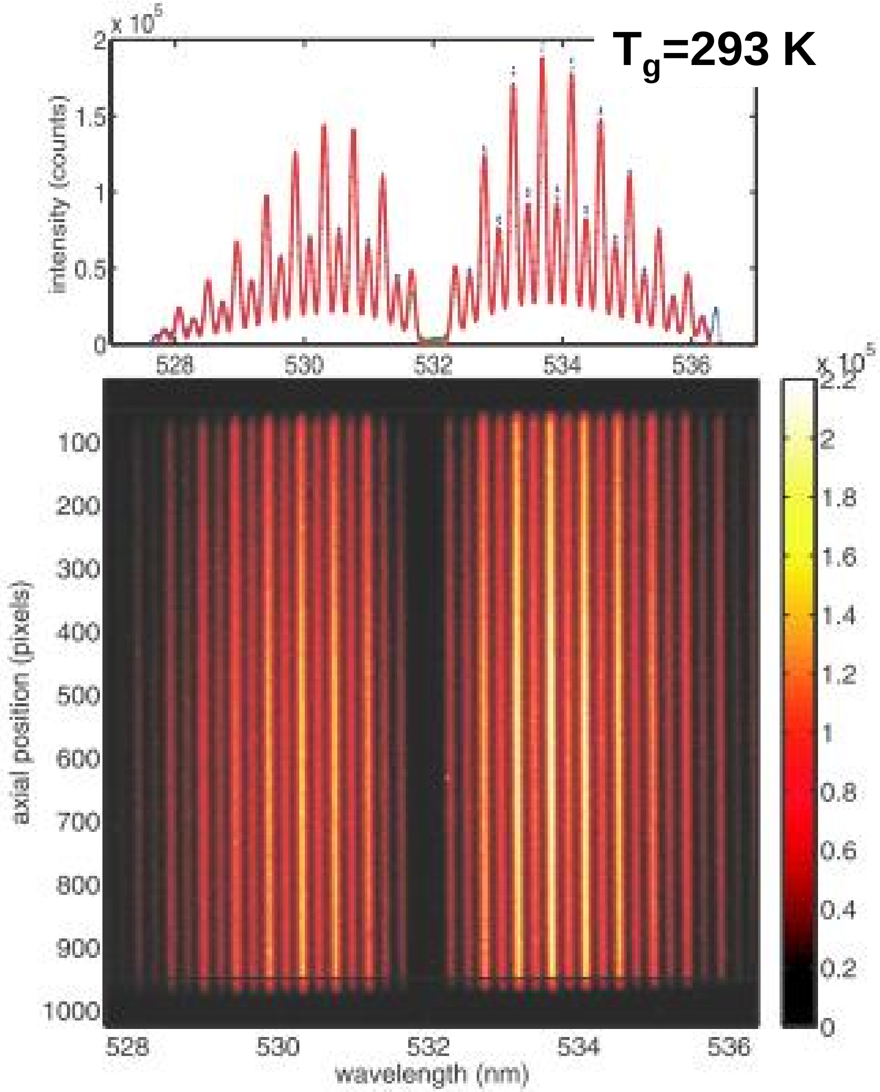
From: Emile Carbone and Sander Nijdam



n_e=5.2 10^{19} m^{-3}

T_e=1.04 eV

Surfatron plasma @ 400 Pa



T_g=293 K

Pur N_2 @ 600 Pa

Use of notch Bragg filter to reduce the Rayleigh signal and scattered light at λ_0

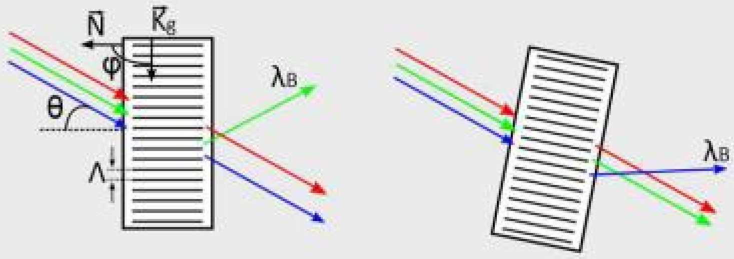


Fig. 1 (a) Schematic of a transmission grating

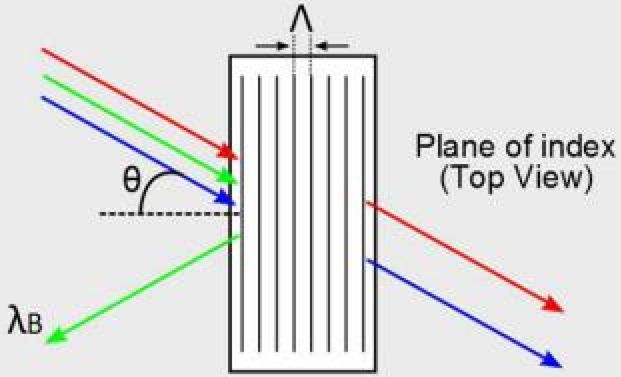
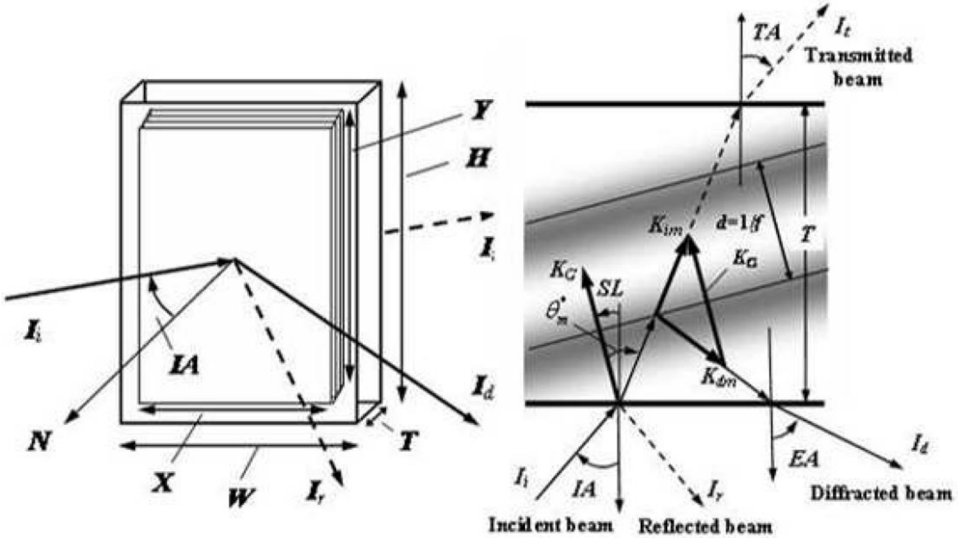
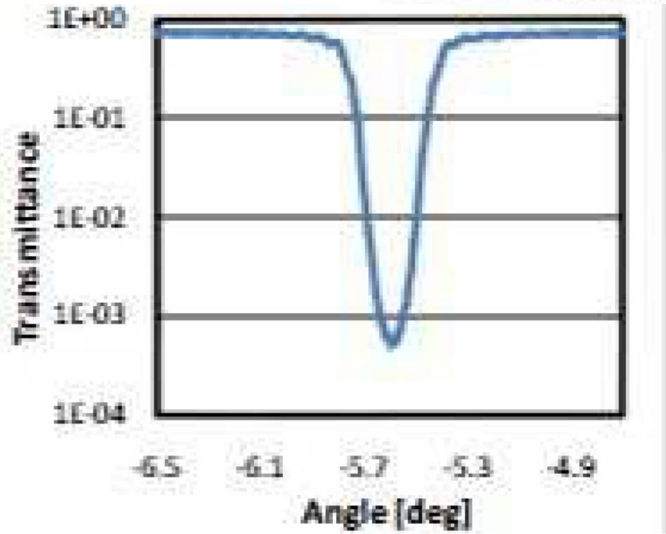


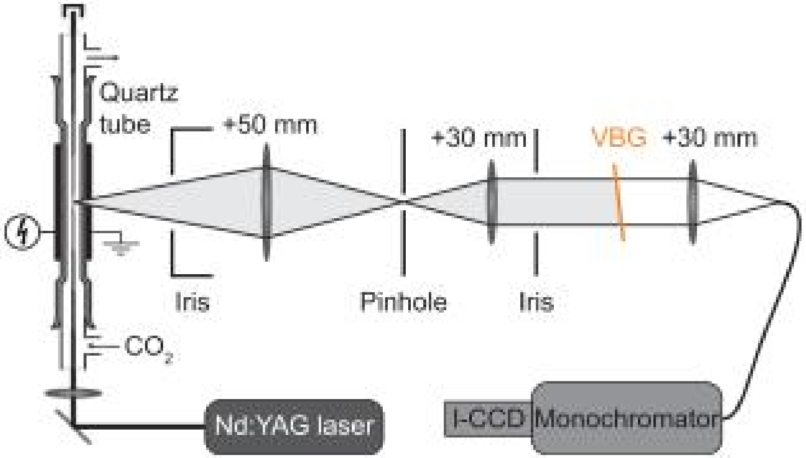
Fig. 1 (b) Schematic of a reflection grating

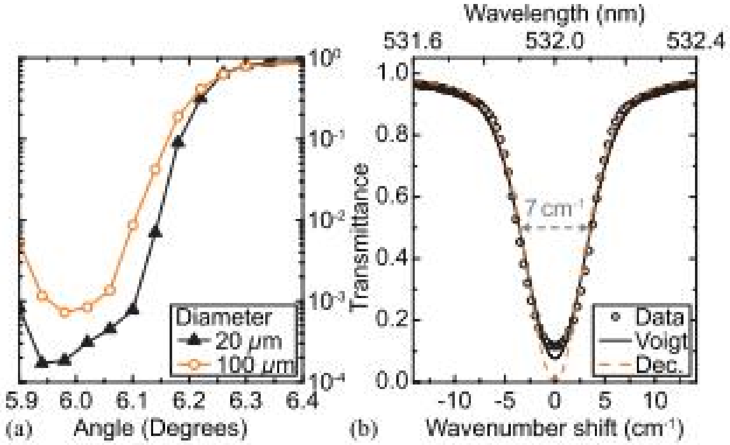


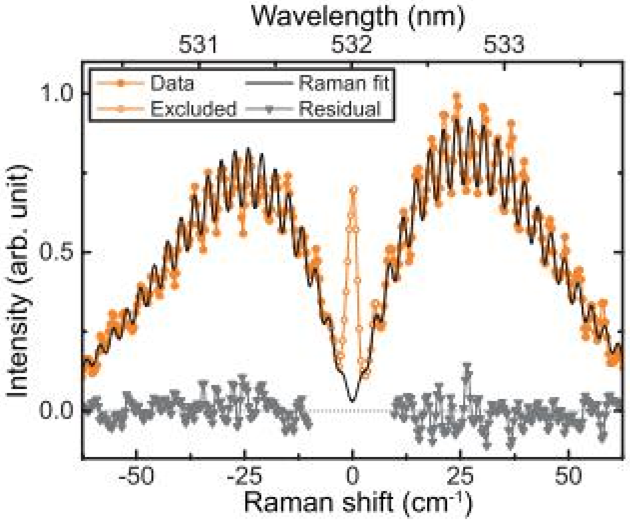


Rotational temperature of CO_2 measured in a CO_2 conversion reactor by Raman scattering with Bragg filter

Klarenaar B L M, et al, *Rev. Sci. Instrum. 86 , 046106 (2015)*







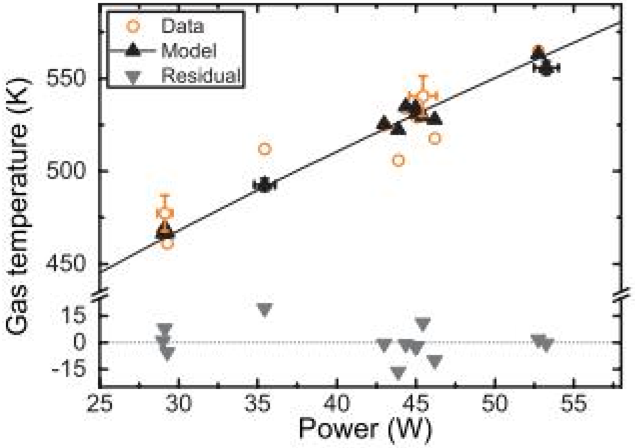
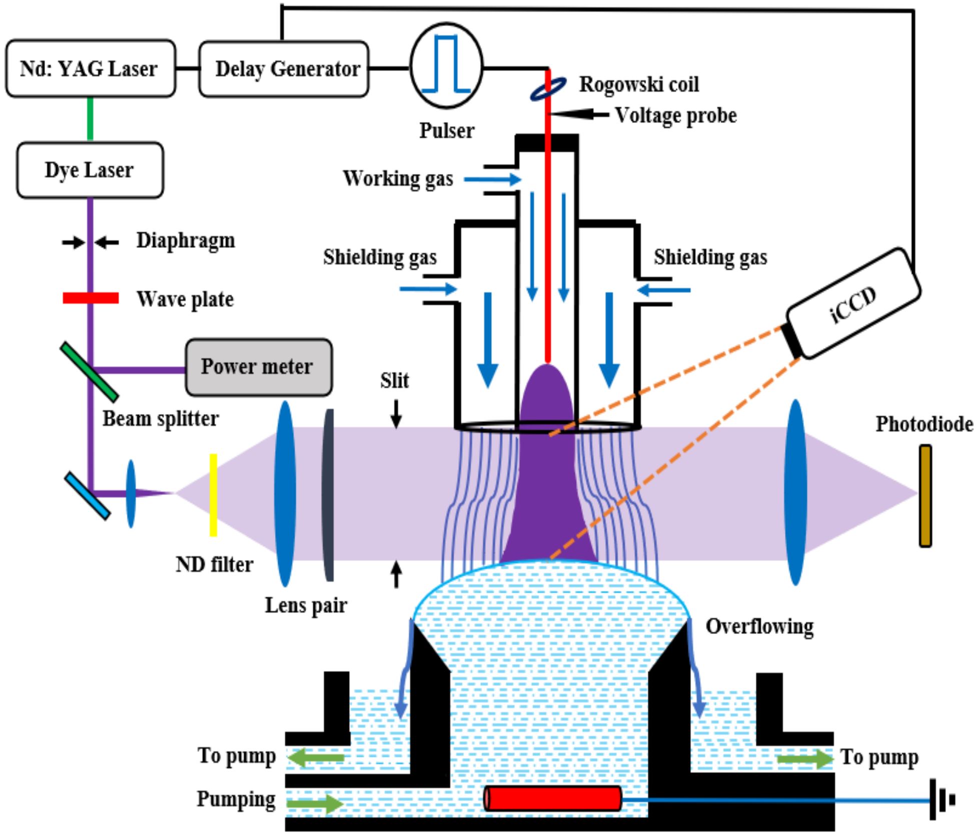


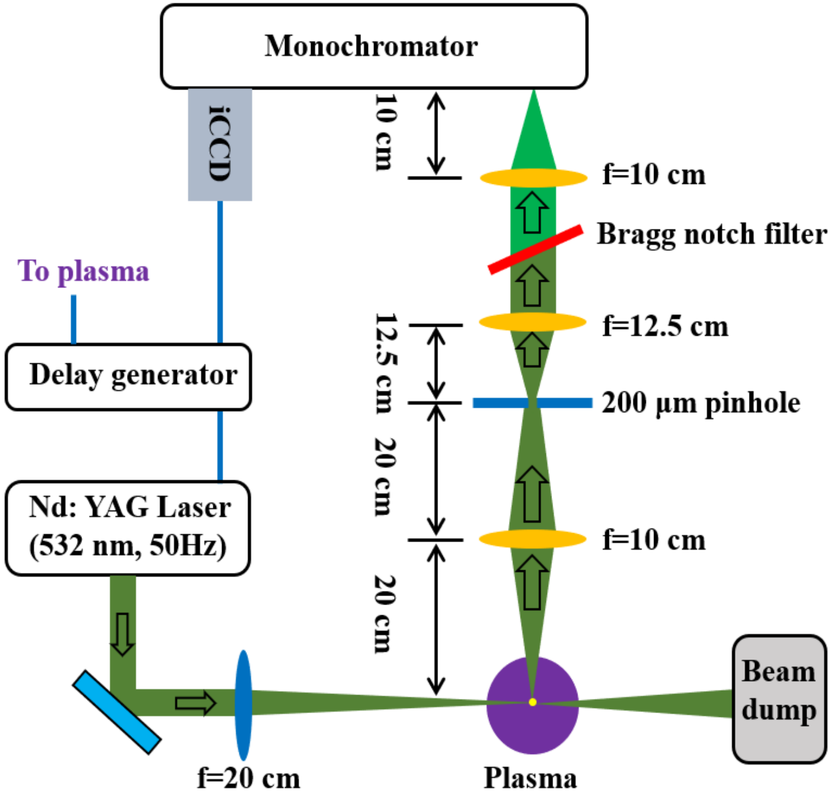
FIG. 4. Gas temperature versus the power dissipated in the plasma.

TS measurement with Bragg filter in AP helium plasma jet impacting in liquid

Y. Yue, V.S.S Kondeti, N. Sadeghi et P.J. Bruggeman, PSST 30 (2021)



He plasma jet scheme



Thomson set up

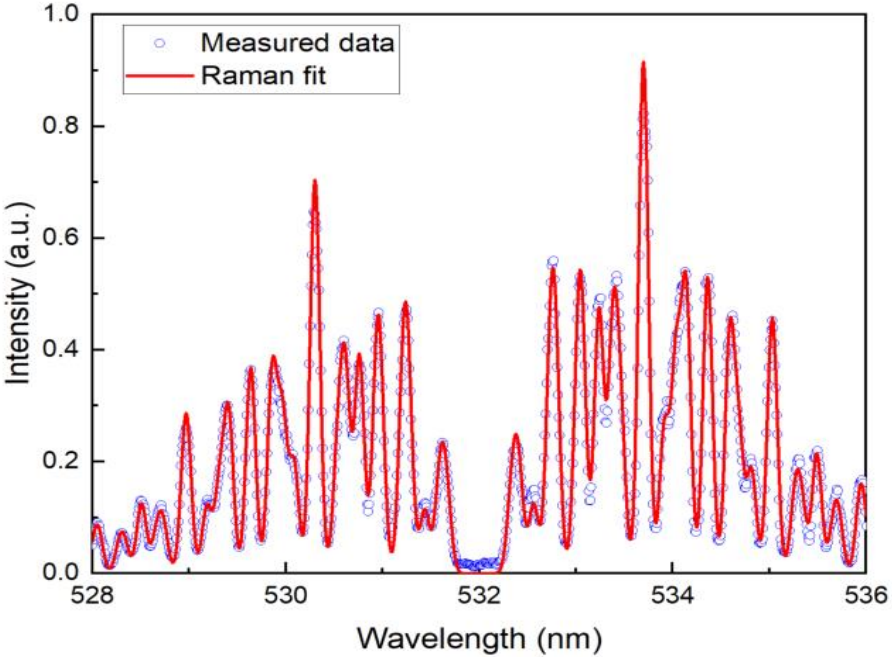
TS measurement with Bragg filter in AP helium plasma jet impacting in liquid

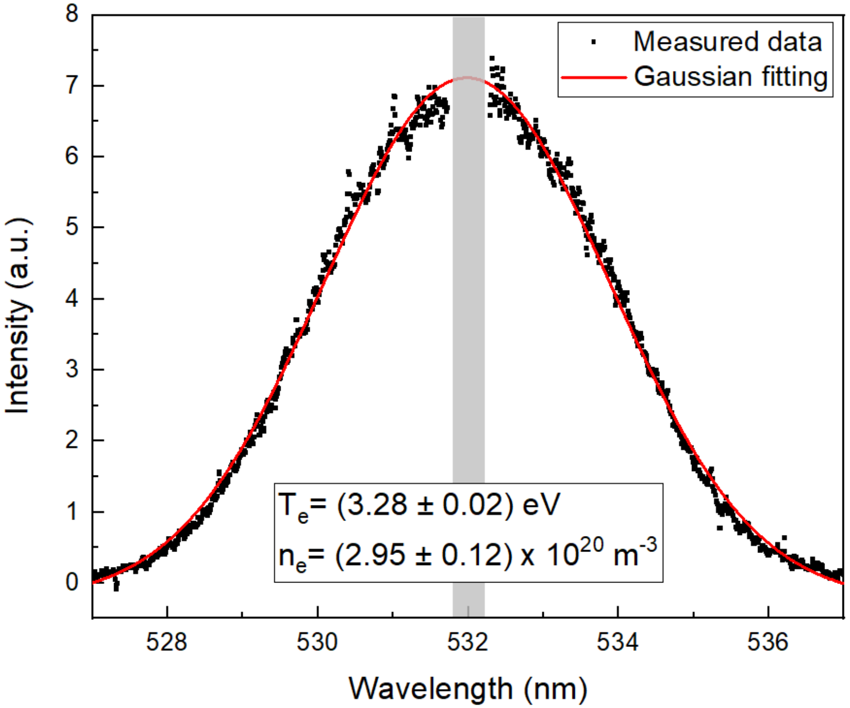
Y. Yue, V.S.S Kondeti, N. Sadeghi et P.J. Bruggeman, PSST 30 (2021)

Raman signal in air @ 300 K

Thomson signal in He jet

n_e=2.95 10^{20} m^{-3}, T_e=3.28 eV





Comparing 3 different techniques

✓ **Line broadening**:

pros: easy with a high resolution spectrometer (SOPRA)

cons: need the knowledge of Tg and pressure broadening coeff

✓ **Continuum emission**:

pros: easy and need a low resolution spectrometer

cons: difficulty for the absolute intensity calibration and EEDF shape estimation: Large uncertainty on n_e and T_e

✓ **Thomson Scattering**:

pros: Cost effective , provides n_e and T_e with good precision

cons: technical complexity, need for the absolute intensity calibration by Rayleigh or Raman scattering

Comparing different Thomson scattering techniques

✓ **Single spectrometer**:

pros: easy with a medium resolution spectrometer

cons: cannot be applied in atmospheric pressure plasmas

✓ **Triple grating spectrometer**:

pros: good precision on n_e and T_e , can provide 1D measurements

cons: need the acquisition (or construction) of a 3 grating spectro

✓ **Notch Bragg filter**:

pros: good precision on n_e and T_e , cost less than TGS

cons: no possibility of 1D recording of the signal

Thank you for your attention