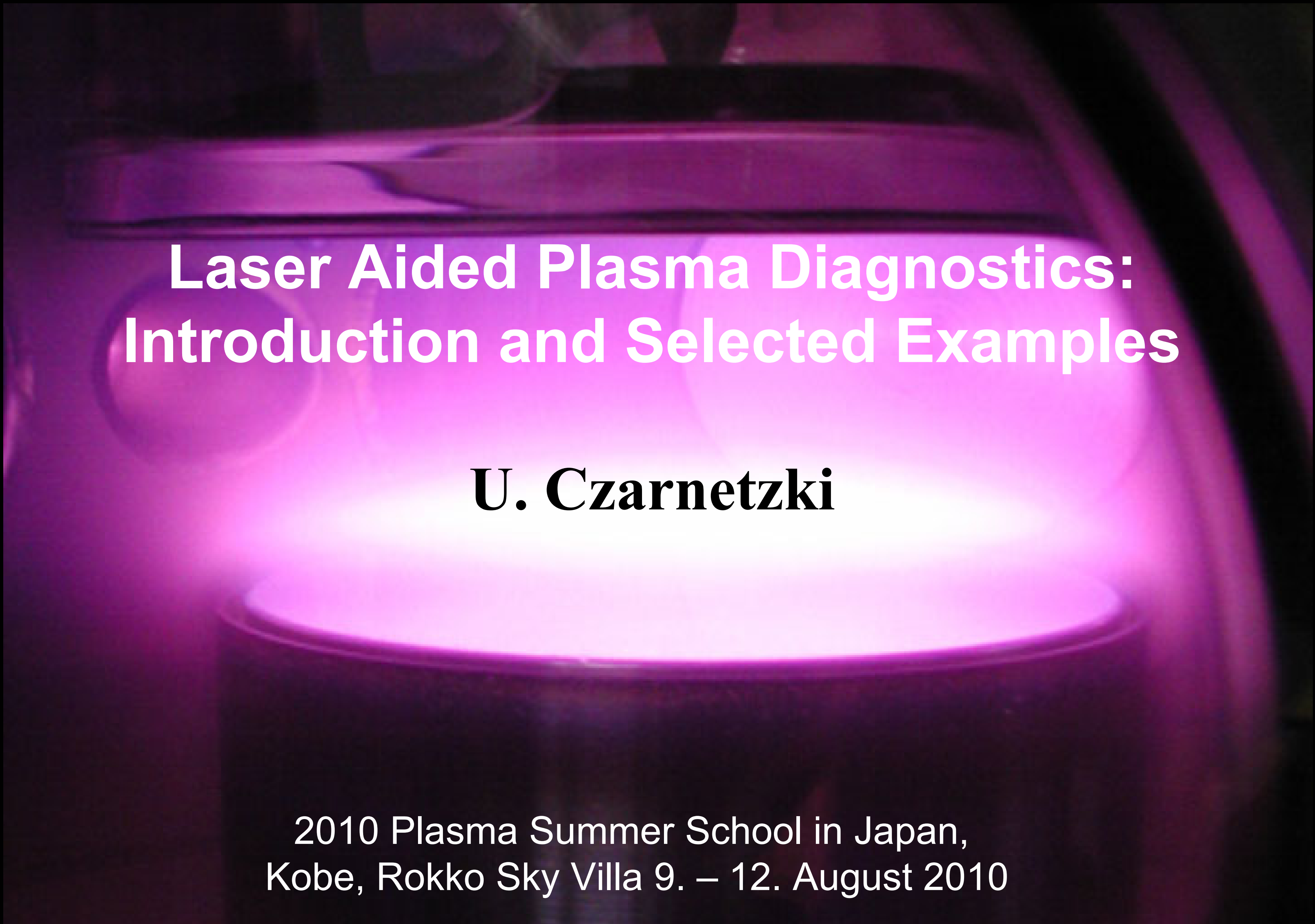

Laser Aided Plasma Diagnostics: Introduction and Selected Examples

U. Czarnetzki

2010 Plasma Summer School in Japan,
Kobe, Rokko Sky Villa 9. – 12. August 2010

Outline

1) Introduction
2) Equipment
3) Thomson Scattering
4) Laser Induced Fluorescence Spectroscopy (LIF/TALIF)
5) Diode Laser Absorption Spectroscopy
6) Summary

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Why should we do diagnostics?

- It is almost never just for the measurement of the parameter the diagnostic is delivering directly.
- This parameter has to be put into a context. The context is a physical picture of the plasma.
- The main insight is given mostly not by the numerical value of the particular parameter but by giving proof to a theory or discovery of an alternative concept.
- Of course, there is need for an initial survey on a completely new devices characterizing its operational regime. But then the work is not finished but just starts!
- In any case, diagnostics without concepts about the physics of the plasma under investigation is a poor business.

The answer is: We want to gain understanding!

Suggesting laser spectroscopy to a plasma physicist

Laser spectroscopy on plasmas is

- expensive (typically > 100 k€)
- difficult (operation, alignment, maintenance, time consuming)
- providing rich and direct information on plasma parameters
- ideal for fundamental studies
- not well suited for process control
- best if combined with other diagnostics (probes, emission, etc.) and models/simulation (!)
- fun!!!

What can laser spectroscopy do other diagnostics can not?

Laser measurements are

- local (from µm scale to line integration)
- fast (sub ns to ms)
- selective (species and state)
- non-intrusive (mostly)
- direct (not depending on plasma models)
- sometimes the only alternative (e.g. non-radiative states)

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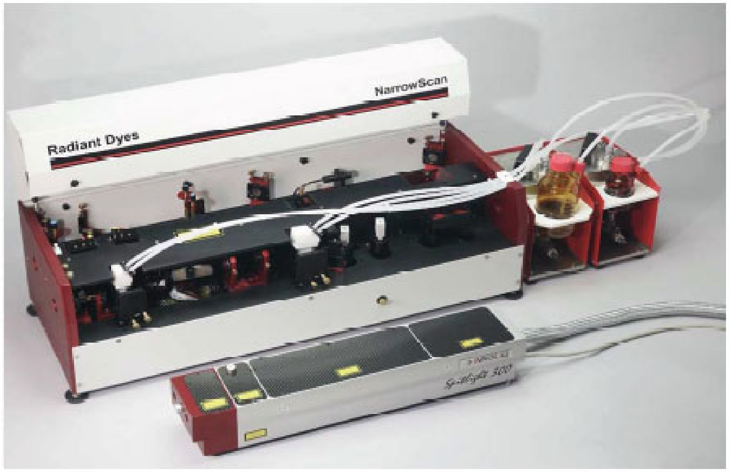
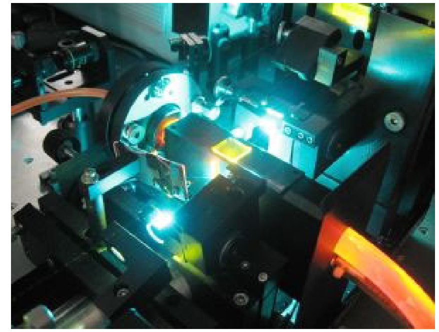
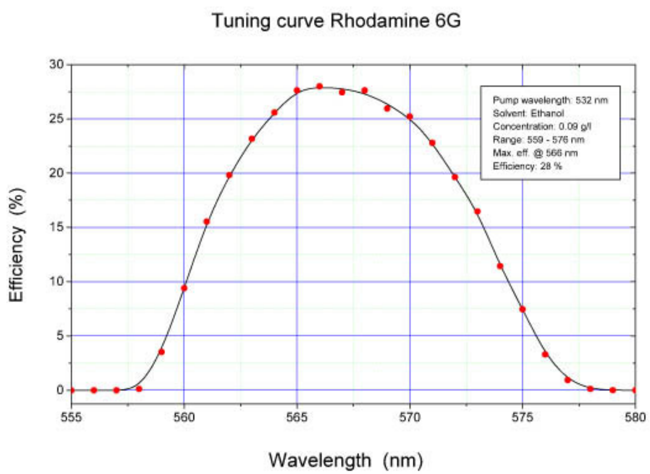
Dyes and Dye-Lasers



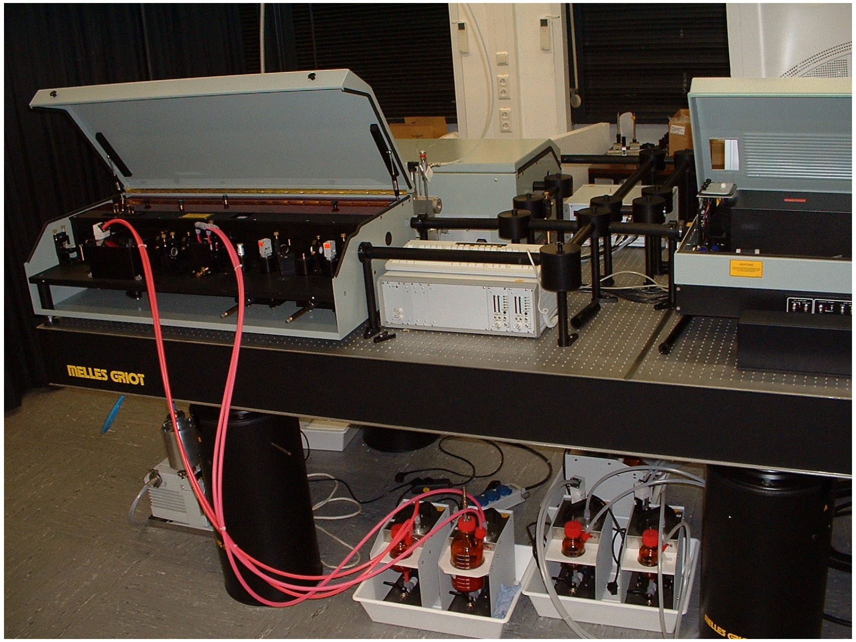




Dyes can be tuned over certain spectral regions. A wide variety of dyes with different central wavelengths is available.

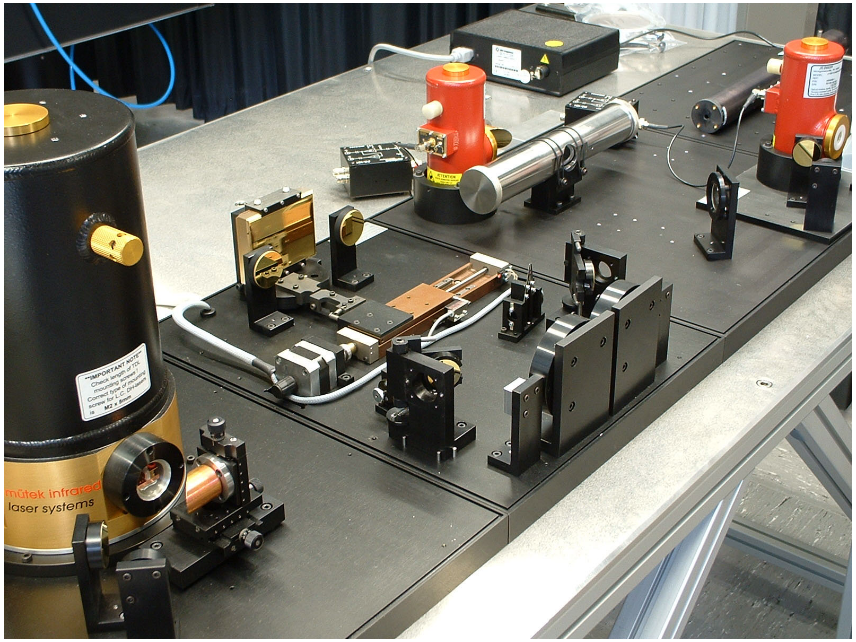
Dyes are usually solved in alcohol.

Dye-Lasers are usually pumped by either Excimer or Nd:YAG lasers



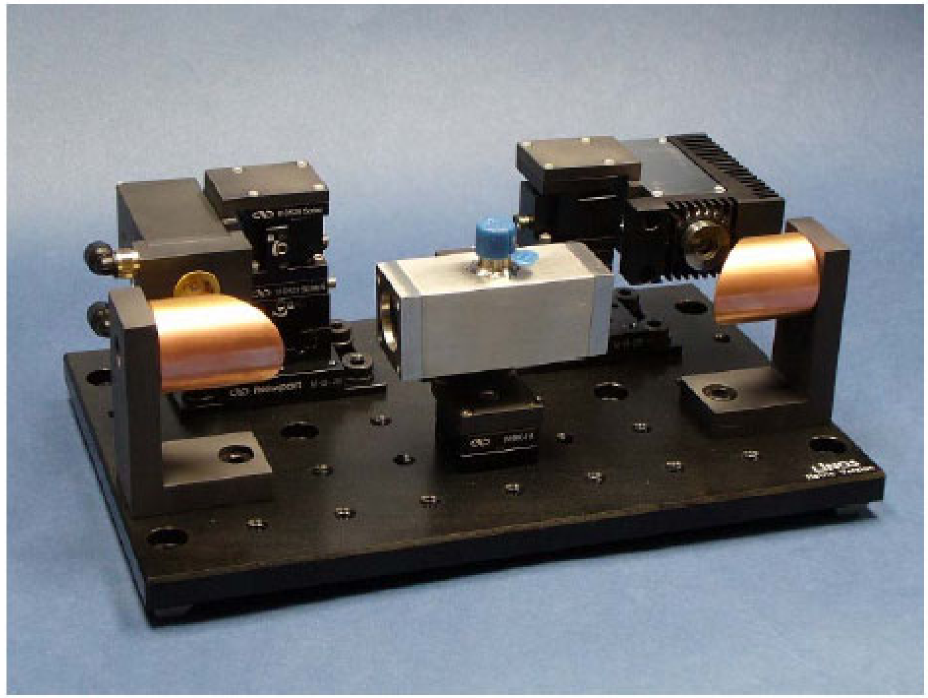
Nd:YAG lasers are also a common source for Thomson scattering and photo-detachment.

IR Lasers are usually based on lead-salt diodes



Liquid nitrogen cooling is essential for lasers and detectors.

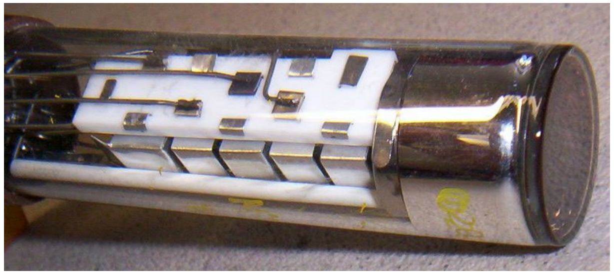
Recently quantum cascade lasers (QCL) and new detectors have become available

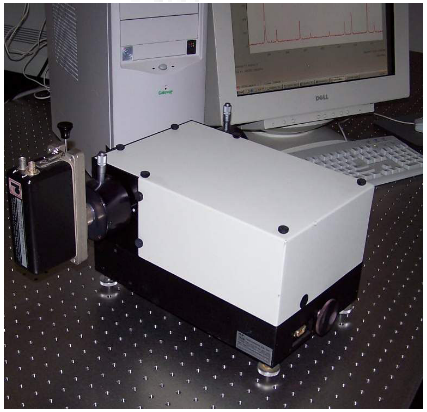


New generation of compact QCLAS Equipment

Very compact, no cooling of lasers and detectors.
However, very narrow tuning range (one laser – one species).

Detectors and Spectrometers



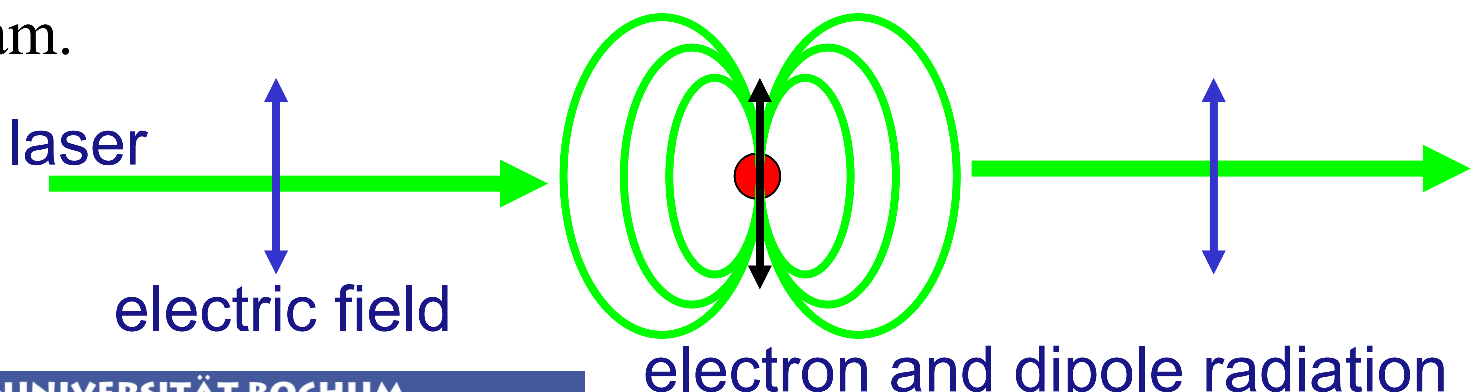


Usually photomultiplier tubes or ICCD cameras are used as detectors.
ICCD cameras allow spatial imaging and fast gating (ns) but are expensive.
Spectrometers or filters are often used for reduction of background radiation.

Outline

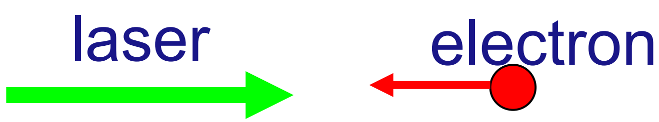
Electrons in oscillating fields

- A linearly polarized laser beam is characterized by a local electric field oscillating at a frequency ω in the direction of the polarization.
- An electron in the laser beam is forced by the field to oscillate at the same frequency and in the same direction.
- The oscillating electron emits dipole radiation with the axis of the dipole in the direction of the polarization of the laser beam.
- The emitted power is strongly anisoptropic. There is no power on the axis and there is maximum power perpendicular to the axis.
- Further, the emitted radiation is linearly polarized. If observed perpendicular to the axis, the polarization is identical to the laser beam.

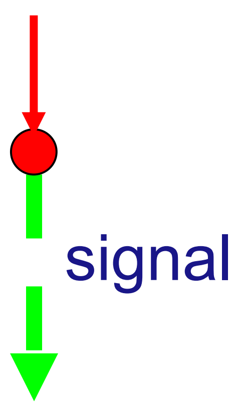


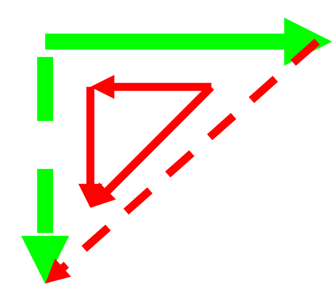
Doppler Effect in Laser Scattering on Electrons

- Light is scattered on electrons not ions due to the large difference in mass.
- An electron moving in the direction of the laser beam absorbs light at a Doppler shifted frequency according to its speed in this direction.
- The electron radiates at the same frequency where it absorbs.
- When the electron moves with respect to the observer in the laboratory there is a second Doppler effect in receiving this light.
- Therefore, the measured velocity component is determined by the angle between the laser and the observation direction.



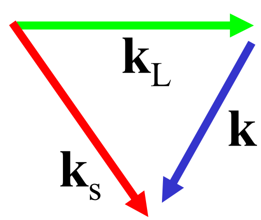
$$\frac{\Delta\omega}{\omega} = \frac{v}{c}$$





Density fluctuations

- When scattering occurs on an ensemble of many electrons, the individual contributions can interfere.
- Taking into account this ensemble effect leads to the concept of density fluctuations in the plasma. A homogeneous density is Fourier decomposed into individual electrostatic waves with a wave vector $k = 2\pi / \lambda$.
- The physical picture is that of scattering on a moving density grating.
- The angle θ between the laser und the detection direction determines the k-vector (the wavelength) of the plasma wave.
- In the scattering process, energy and momentum have to be conserved.



$$\omega = \omega_s - \omega_L$$

$$\vec{k} = \vec{k}_s - \vec{k}_L$$

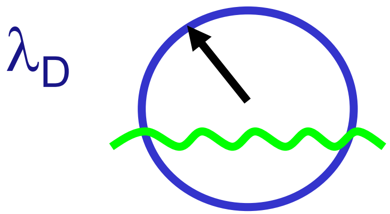
Scattering parameter α

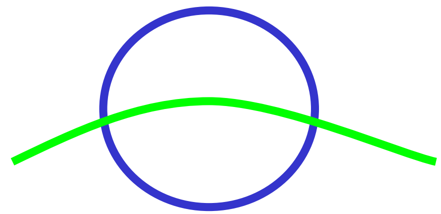
The critical parameter in Thomson scattering is α, the ratio between the wavelength of the density fluctuation to the Debye length:

$$\alpha = \frac{1}{k\,\lambda_D} = \frac{\lambda_L}{4\pi\,\lambda_D \sin(\theta/2)}$$

The nature of the scattering process is fundamentally different if either

α << 1 or α >> 1.





$$\lambda = \frac{\lambda_L}{2\sin(\theta/2)}$$

In the former case, electrons in the Debye sphere can react individually to the local electric field and the individual radiation intensities sum to the total intensity.

In the latter case all electrons radiate in phase. Radiation is emitted coherently. The individual electric fields sum to the total electric field.

Characteristics of the scattering Regimes

a) $\alpha << 1$ (incoherent scattering):

λ_L is small and/or n_e is low.
k is large.
$\lambda << \lambda_D$
Scattering on free electrons within a Debye sphere.
Typical situation in low pressure plasmas with $\theta = 90°$.

b) $\alpha >> 1$ (coherent or collective scattering):
λ_L is large and/or n_e is high.
k is small.
$\lambda >> \lambda_D$
Scattering on collective fluctuations beyond a Debye length.
Typical situation in arcs or microwave scattering in fusion research with $\theta << 1$.

Incoherent Scattering ($\alpha << 1$)

- The velocity distribution function of the electrons in the direction of the wave vector of the density fluctuations is represented as a spectral distribution in the scattered light.
- The scattered intensity is proportional to the electron density.
- From the measured spectrum, the electron velocity distribution function can be obtained, e.g. Maxwell distribution gives a Gaussian spectrum.
- The integral over the spectrum is proportional to the density.
- The absolute density can be obtained after calibration by either Rayleigh (typically Argon) or Raman (typically Nitrogen) scattering at a gas with known density and scattering cross section.

Challenges

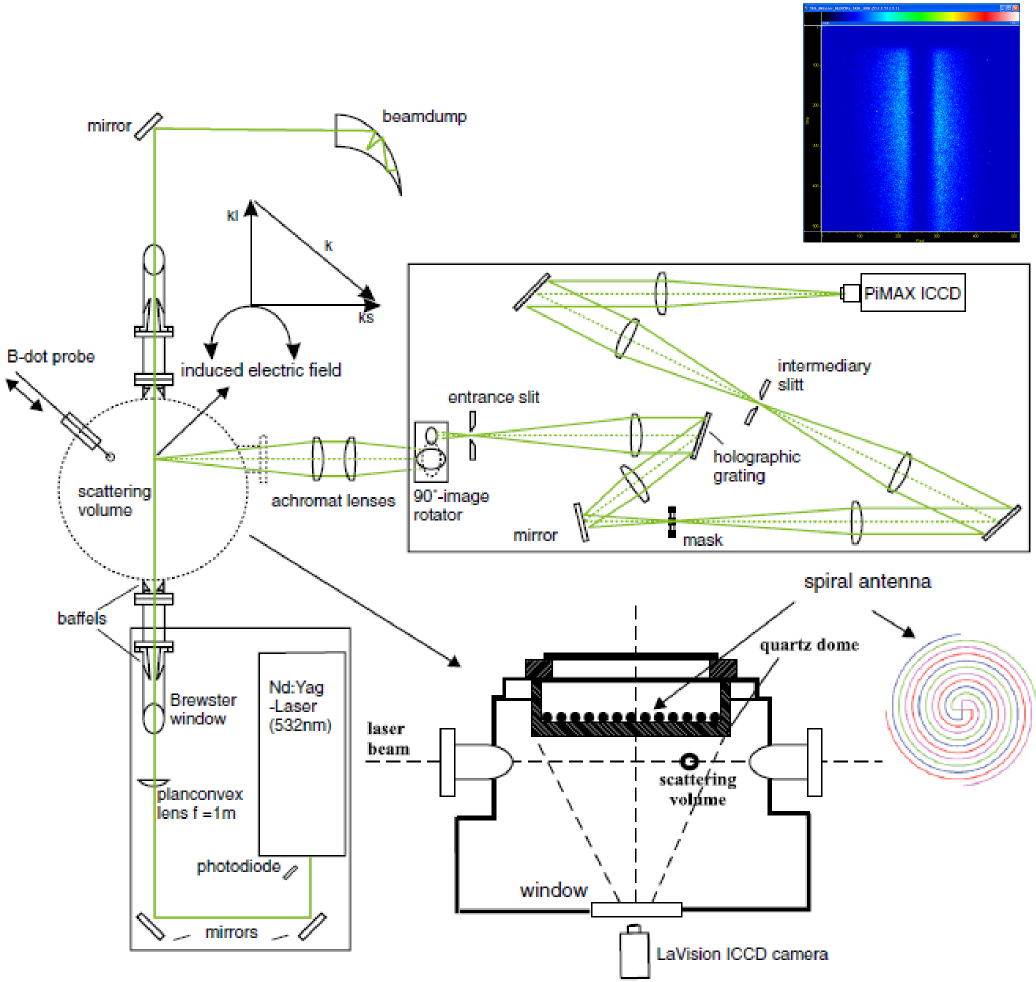
The main problems in Thomson scattering are usually:

- Small signals due to small scattering cross section.
- High signal at the laser wavelength by straylight and Raighleigh scattering (at higher neutral gas densities).
- High background by plasma emission.

Further problems:

- In processing plasmas with molecules, the laser light might lead to Raman scattering that is superimposed on the Thomson spectrum.
- Ionization out of metastable atoms or detachment of negative ions can lead to the generation of additional free electrons.

Incoherent Thomson Scattering Setup



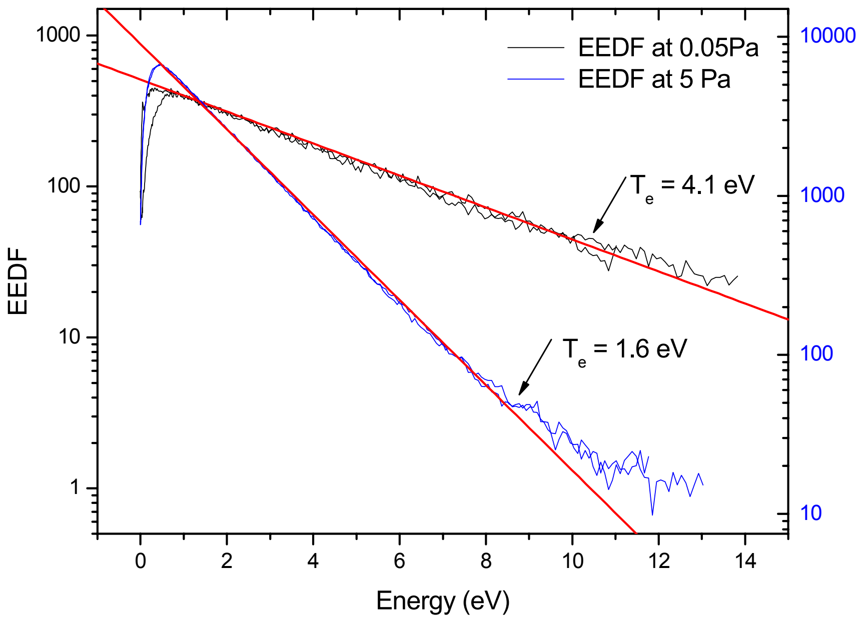
High power laser (30 W) with high repetition rate (50 Hz).

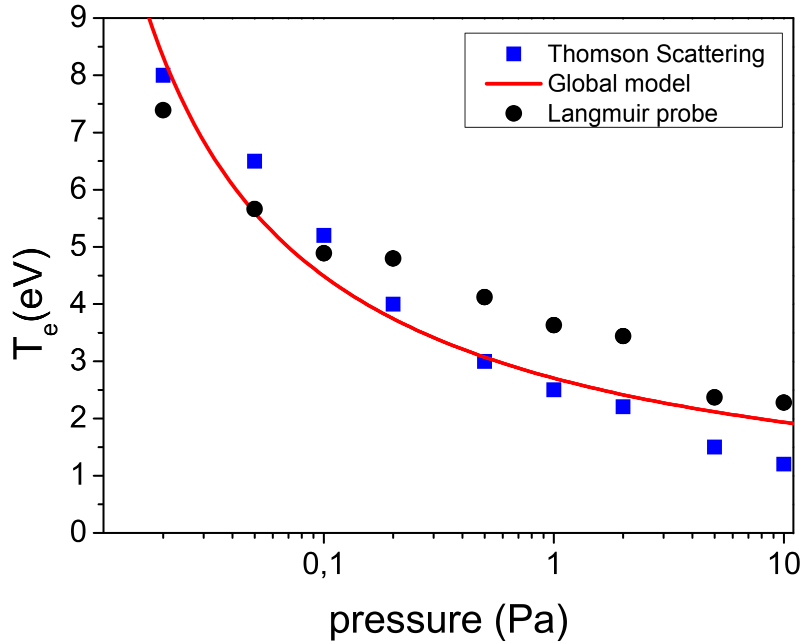
Triple grating spectrometer for suppression of light at the fundamental frequency

ICCD camera with GaAs cathode.

D.L. Crintea, D. Luggenhölscher, V.A. Kadetov, Ch. Isenberg, and U. Czarnetzki Journal of Physics D: Applied Physics 41, 082003 (2008)

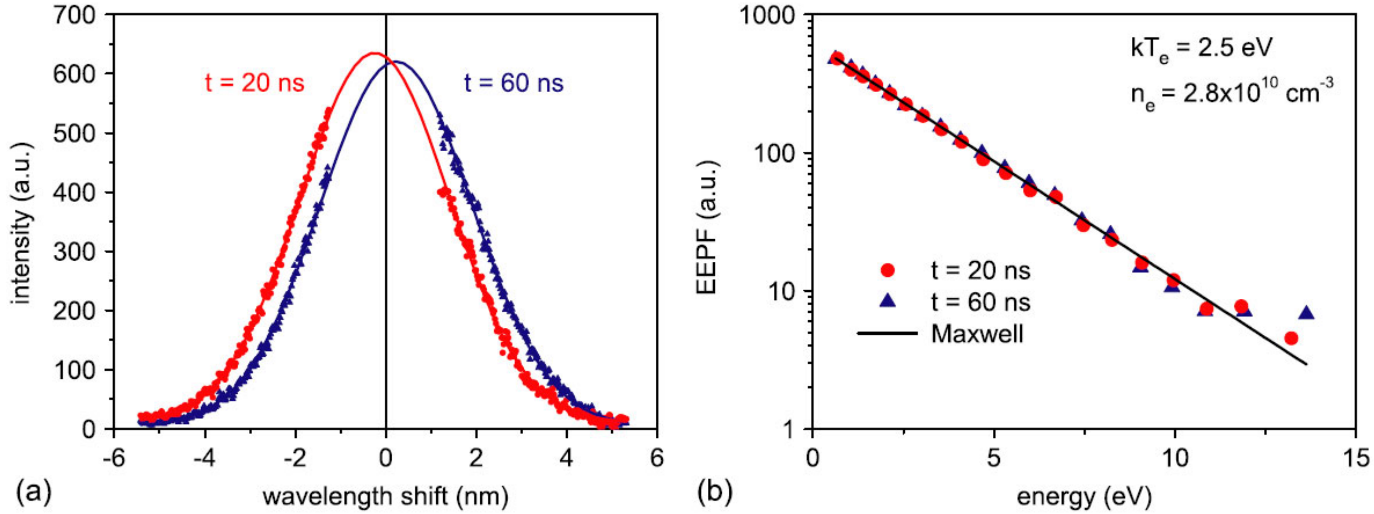
EVDF and Electron Temperature

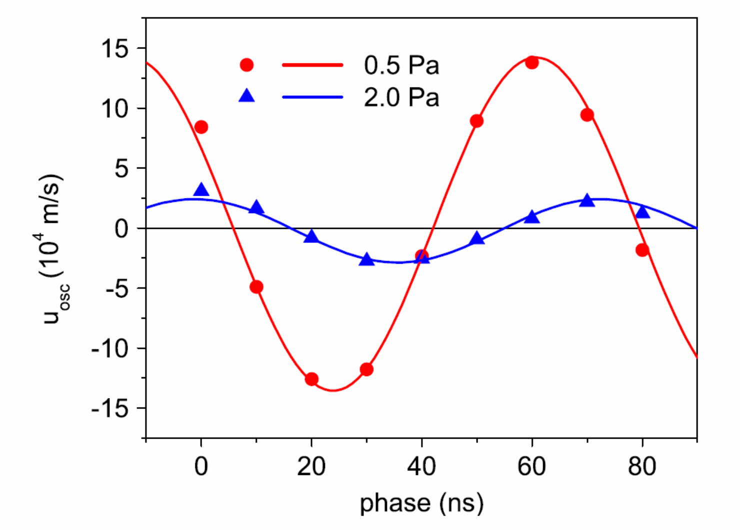




Maxwellian energy distribution
(due to Coulomb collisions).

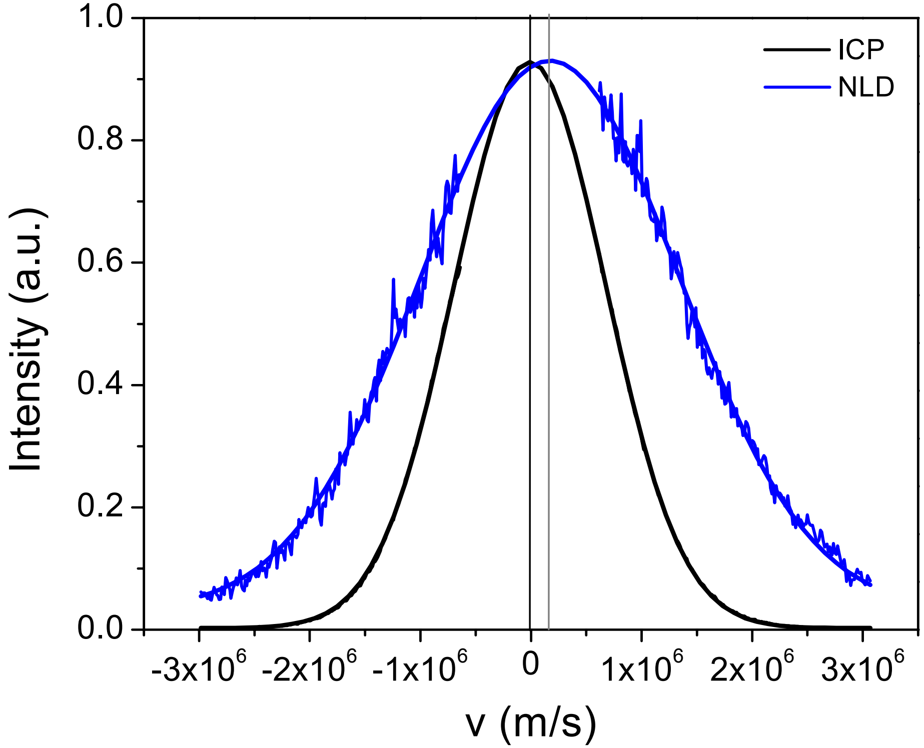
Phase Resolved Measurements in an RF-ICP

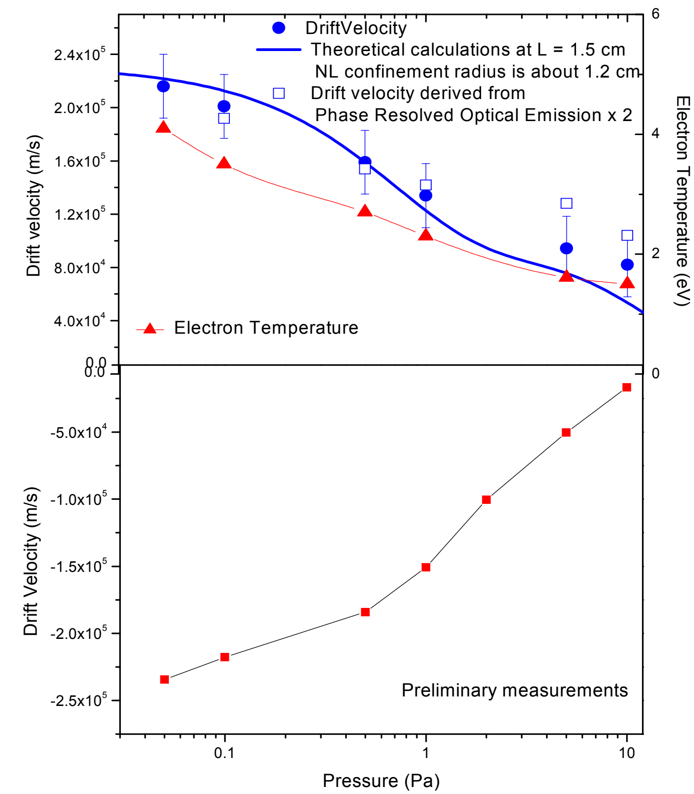




- Displacement of the velocity distribution by the oscillatory drift velocity.
- Via the absolute measurement of the density also the current density is determined (62 mA/cm^2).
- Further, the local electric field can be determined (0.67 V/cm at 0.5 Pa).

Diamagnetic Drift Measured with Thomson Scattering



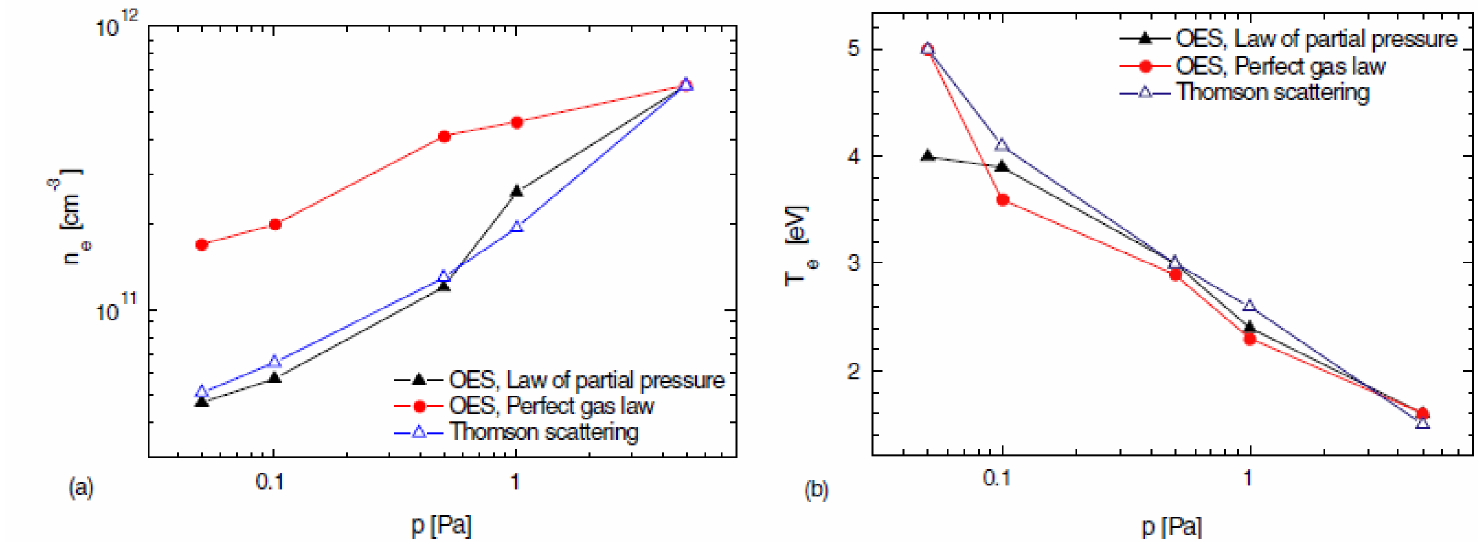


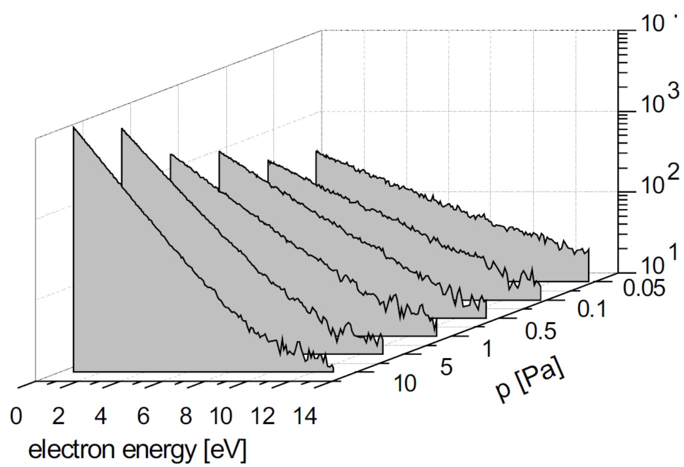
shifted velocity distribution

drift velocity

Dragos Crintea, PhD Thesis, Ruhr-University Bochum (2009)

Comparing Thomson Scattering to a Novel Collisional-Radiative Model in Argon





For the CR emission spectroscopic scheme it was important to ensure the distribution is Maxwellian and to have reference data (T_e, n_e) to compare to. Further, it could be demonstrated that the electron pressure reduces the neutral gas density.

D L Crintea, U Czarnetzki, S Iordanova, I Koleva and D Luggenhölscher, J. Phys. D: Appl. Phys. **42** (2009) 045208

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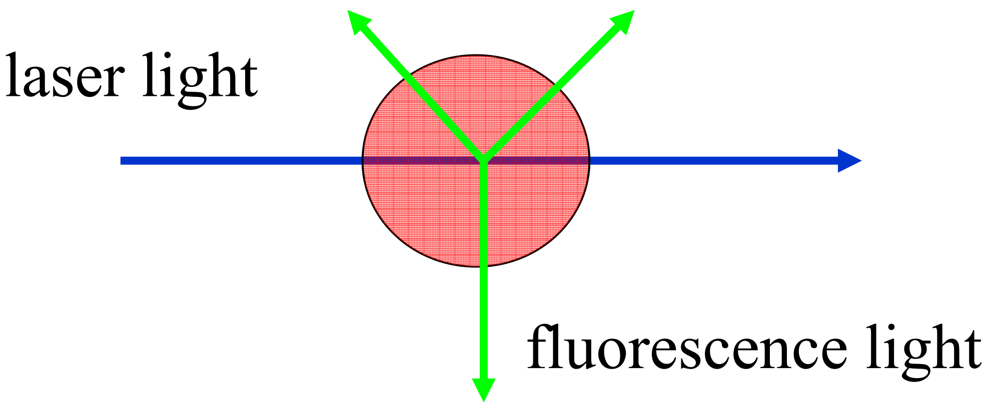
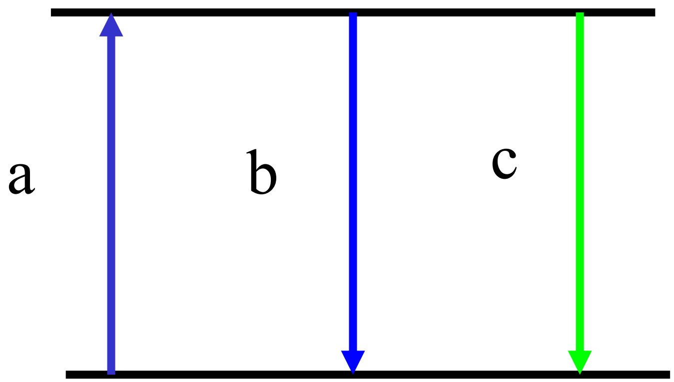
Laser Induced Fluorescence Spectroscopy (LIF)

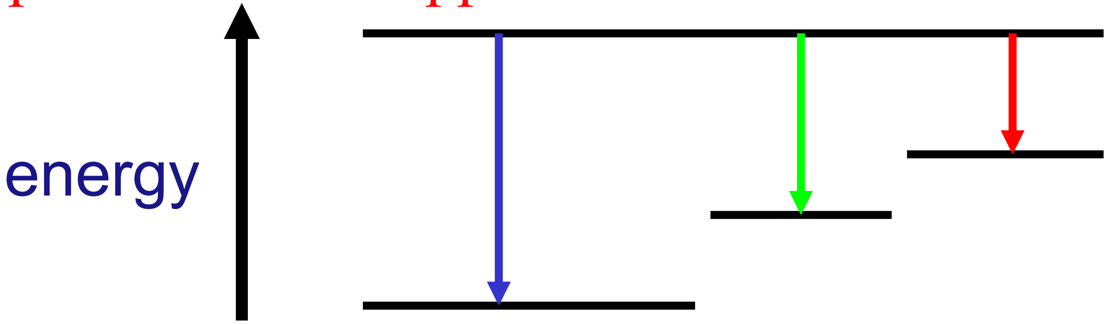




Three processes are possible in the interaction of light with atoms:
a) absorption, b) stimulated emission, c) spontaneous emission
Processes a) and b) are proportional to the laser intensity.
Spontaneous emission is emitted isotropically (from a randomly oriented ensemble of atoms or molecules)

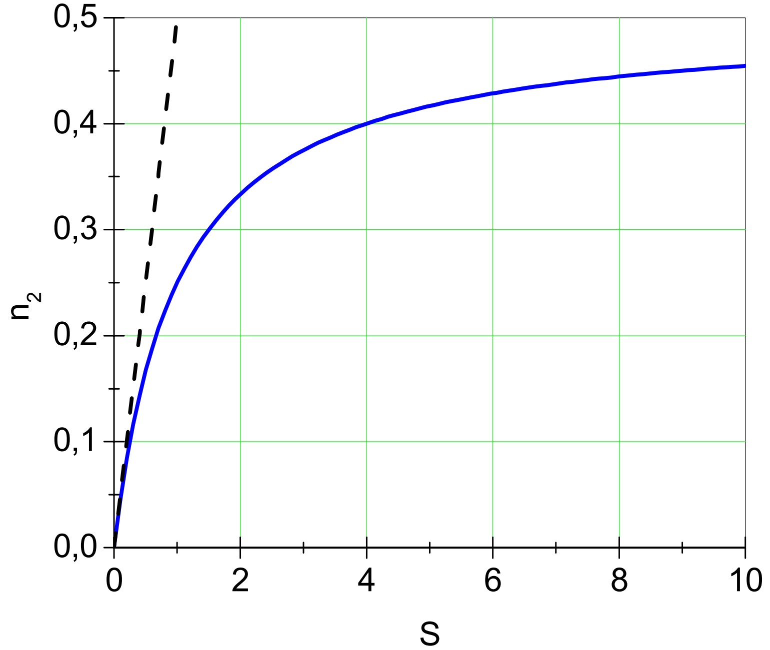
Characteristics of Fluorescent Light

- The spontaneous transition from an excited state to a lower state is connected directly to the emission of photons.
- The wavelength of these photons depends on the energy difference between the upper and the lower state.
- The transition might lead back to the initial state or some other intermediate state (branching).
- Fluorescence light is effectively isotropic.
- The observed fluorescence intensity is directly proportional to the population in the upper state.



Saturation in a Two-Level System

- The population of the upper level depends on the laser intensity I.
- At low intensities it scales linear, at high intensities it saturates.
- Low intensities (S << 1): induced emission negligible
- High intensities (S >> 1): spontaneous emission negligible
- Definition of a dimensionless saturation parameter S:



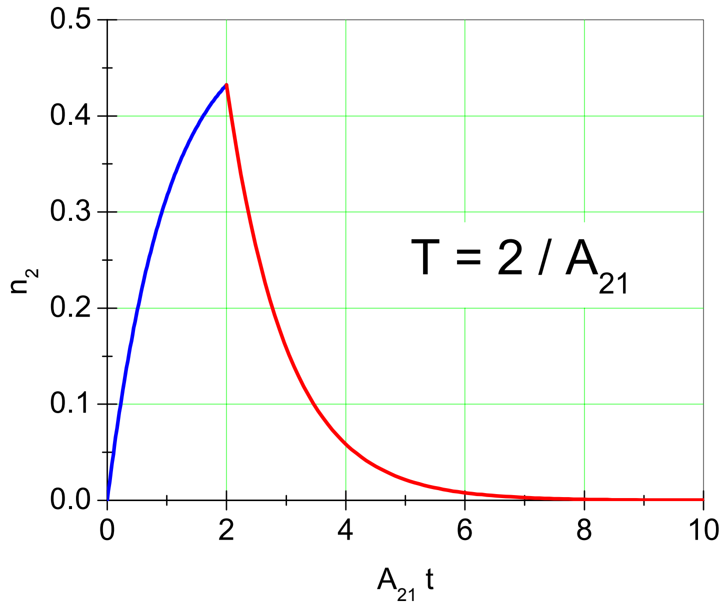
$$S = \frac{B_{12} + B_{21}}{A_{21}} u_\nu = \frac{\lambda^3}{8\pi h c} \frac{I}{\Delta\nu}$$

$$n_2 = \frac{1}{2} \frac{S}{S+1} \qquad n_1 + n_2 = 1$$

B_{12}, B_{21}, A_{21}: Einstein coefficients
u_ν: spectral energy density
$\Delta\nu$: spectral width of the laser radiation
λ: laser wavelength
n_1, n_2: relative populations

Pulsed Excitation

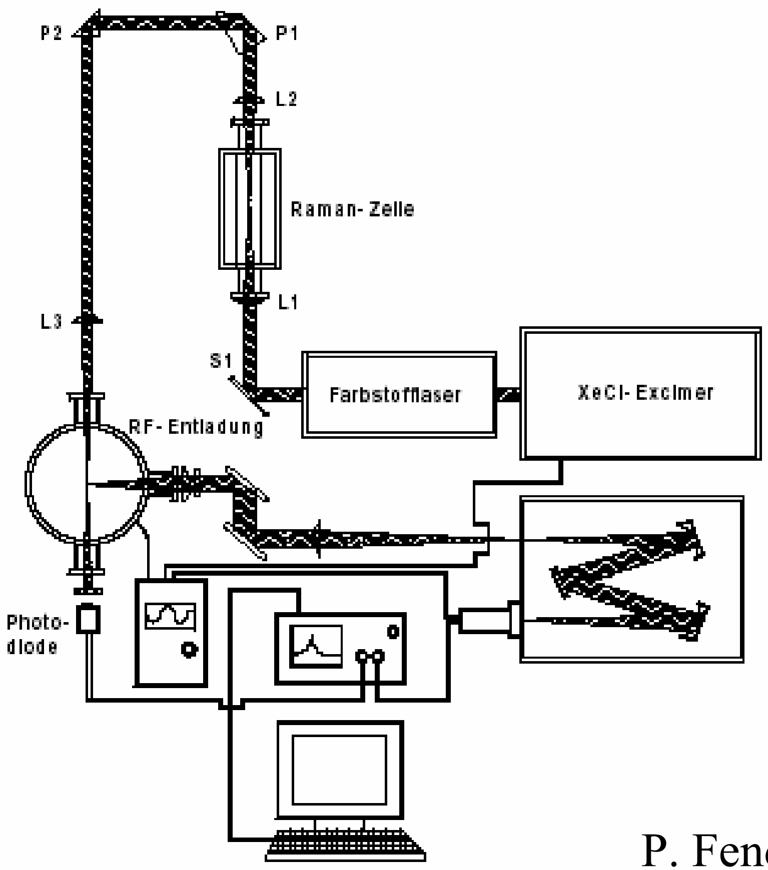
This saturation behavior is exactly true only for cw excitaiton. In many cases short laser pulses of T = only a few ns are used. This is much shorter than radiative lifetime $\tau = 1/A_{21}$ of the upper state.
Then there might not be enough time during the laser pulse for the population to come to an equilibrium with the radiation field, i.e. at the end of the laser pulse the population can be lower than under cw conditions.

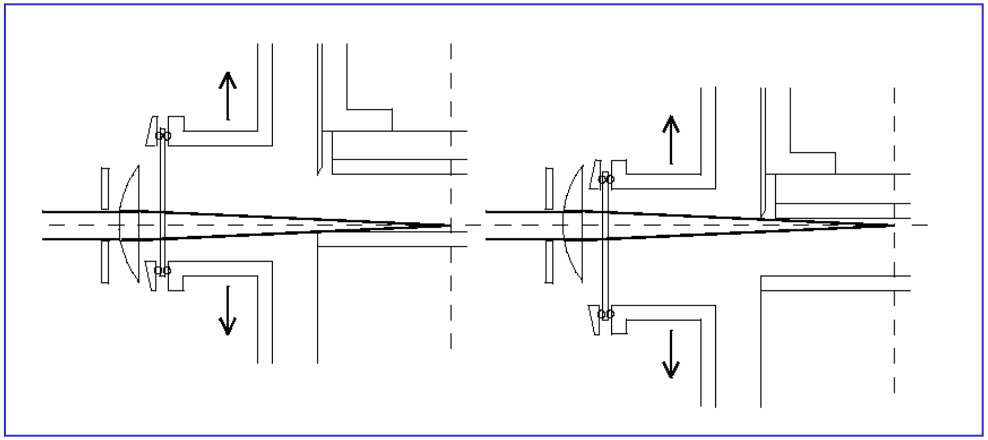


$$n_2 = \begin{cases} \frac{1}{2}\frac{S}{S+1}\left(1-e^{-(s+1)A_{21}t}\right) & 0 \le t \le T \\ n_T\, e^{-A_{21}(t-T)} & t > T \end{cases}$$

Consequently, even for S >> 1 the population can still be far from saturation if $T << 1/A_{21}$ (S+1).

CF and CF_2 densities in a RF-CCP-Discharge with CF_4

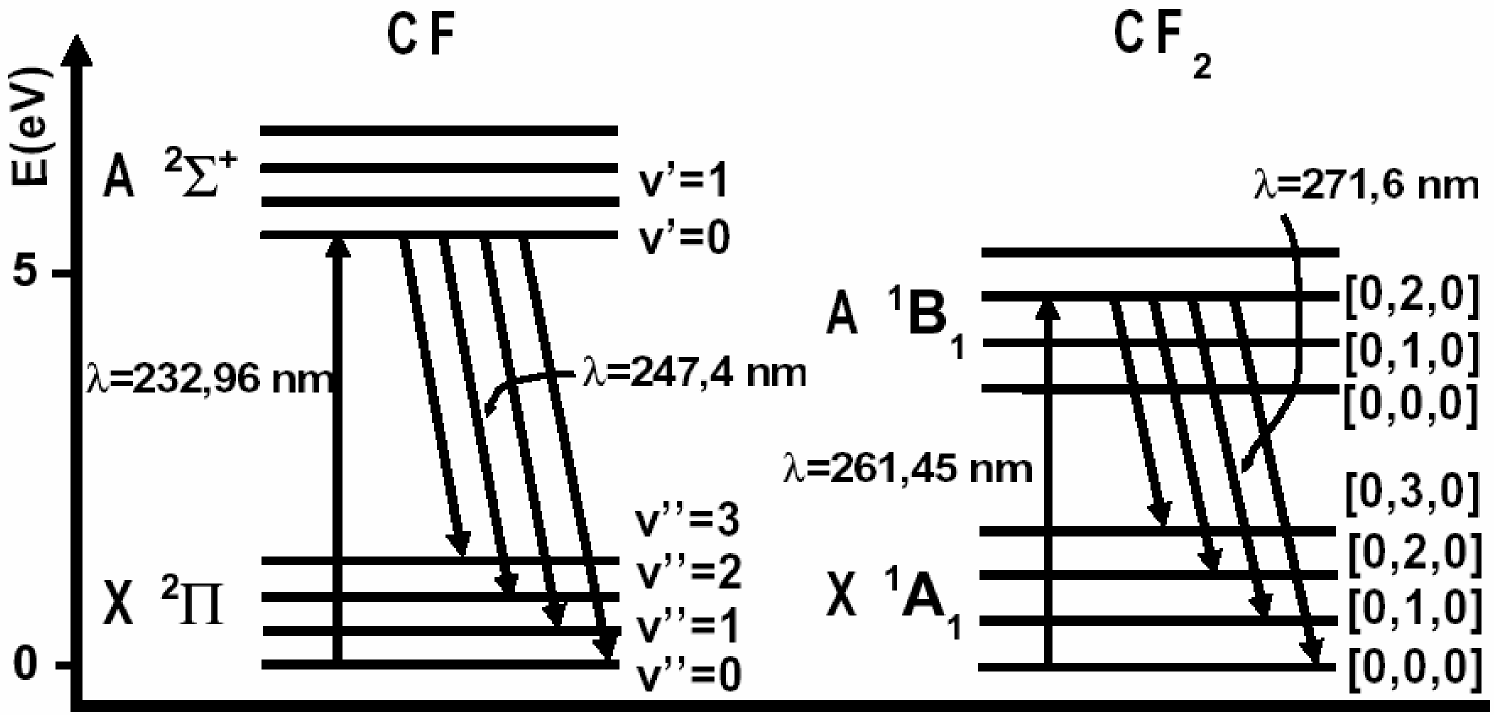




spatial (axial) profiles: radical sources,
temporal decay (pulsing): diffusion constants and chemical reactions

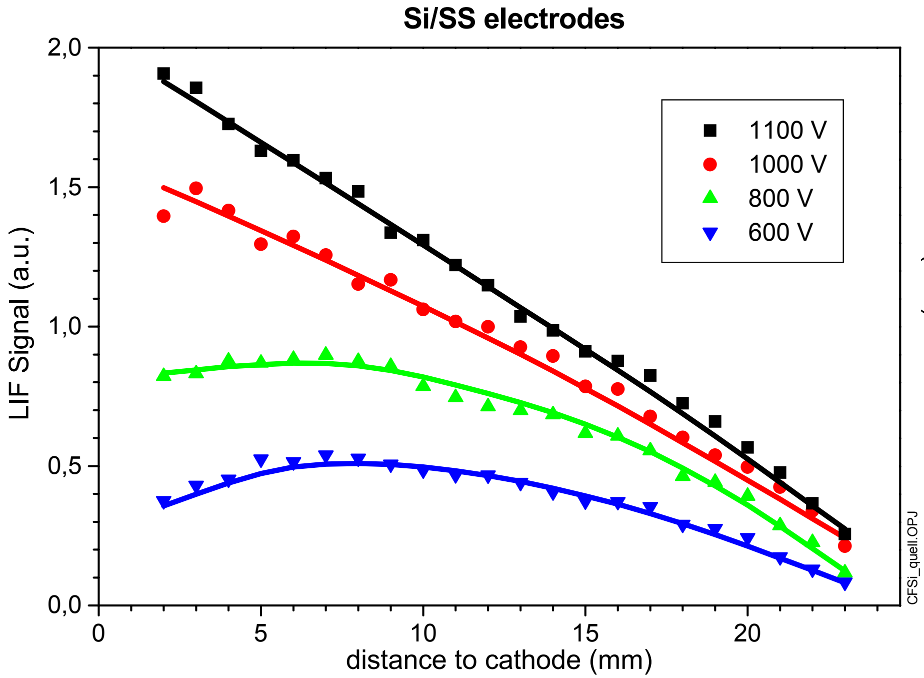
P. Fendel, A. Francis, U. Czarnetzki
Plasma Sources Science and Technology 13, 1 (2004)

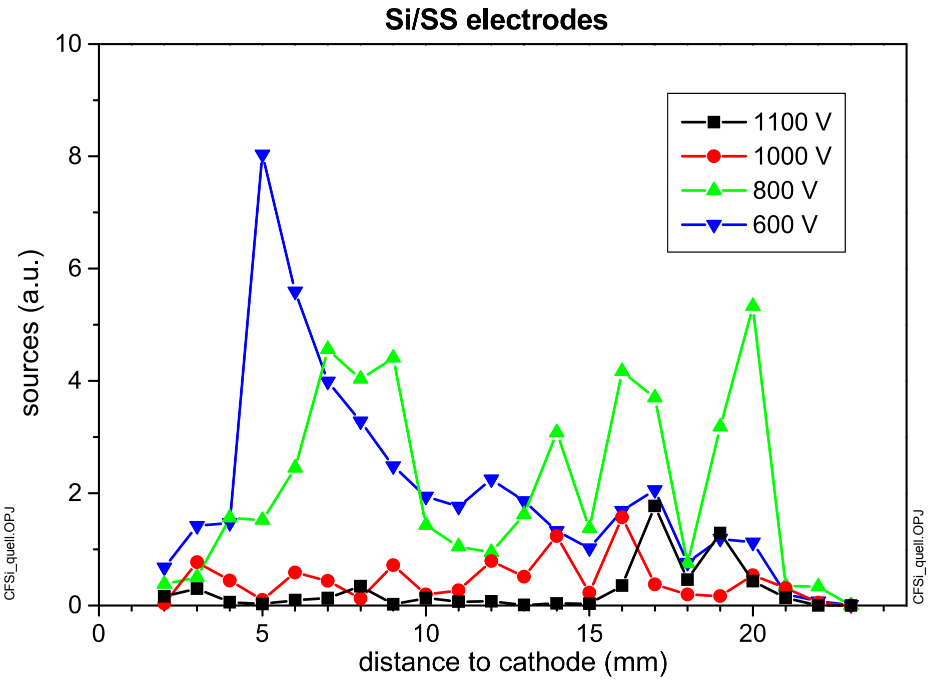
Detection of CF and CF_2 Densities by Laser Induced Fluorescence (LIF)



Various opportunities for excitation and emission exist. One has to chose carefully transitions which provide optimum signal-to-noise ratios.

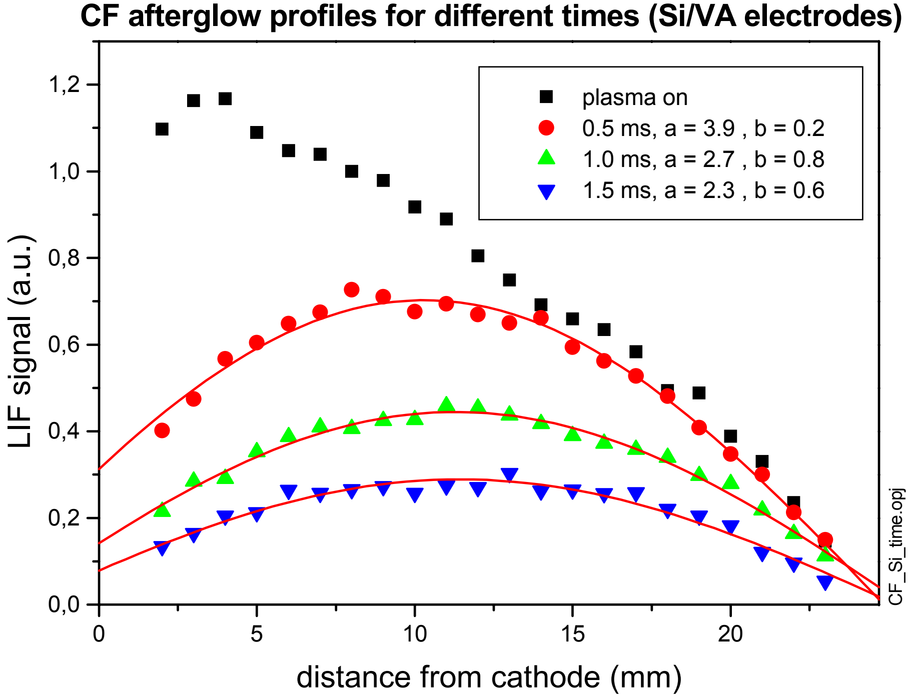
CF Sources Inferred from Spatial Density Profiles

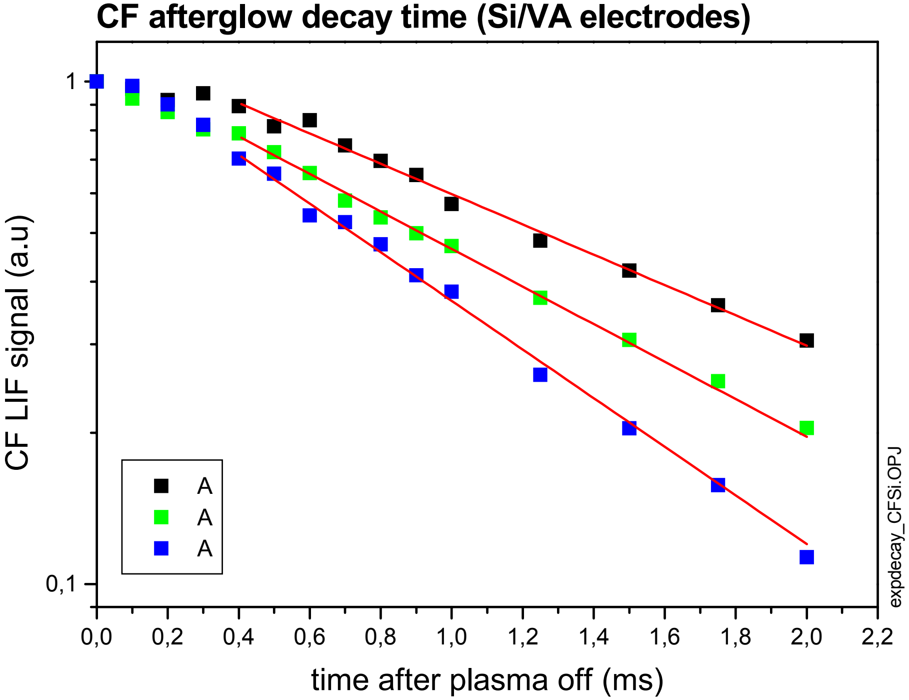




The source distribution is related to the spatial density distribution by the diffusion equation.

Decay Measurements in the Afterglow

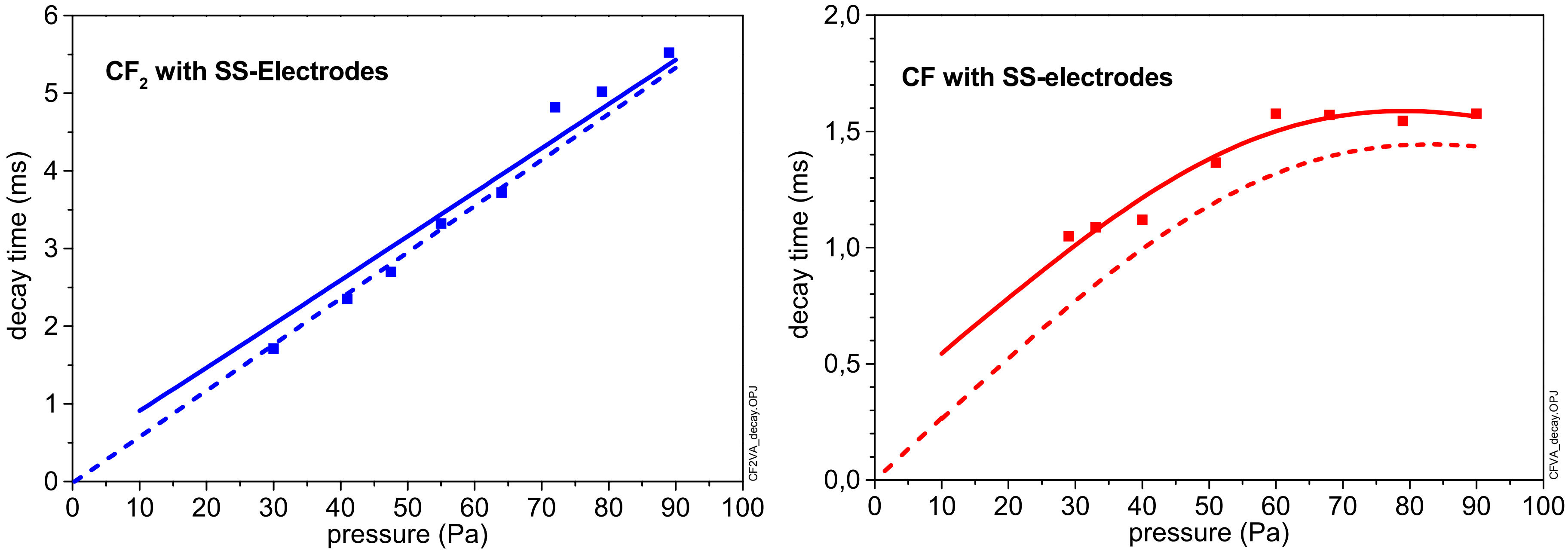




A diffusion model is fitted to all profiles. From the fit, the sticking coefficients at both electrodes are inferred. These coefficients are used in the further processing of the data.

Typically ns laser pulses are much shorter than the corresponding processes in the plasma (at least at low pressures).

Radical Density Decay in the Afterglow

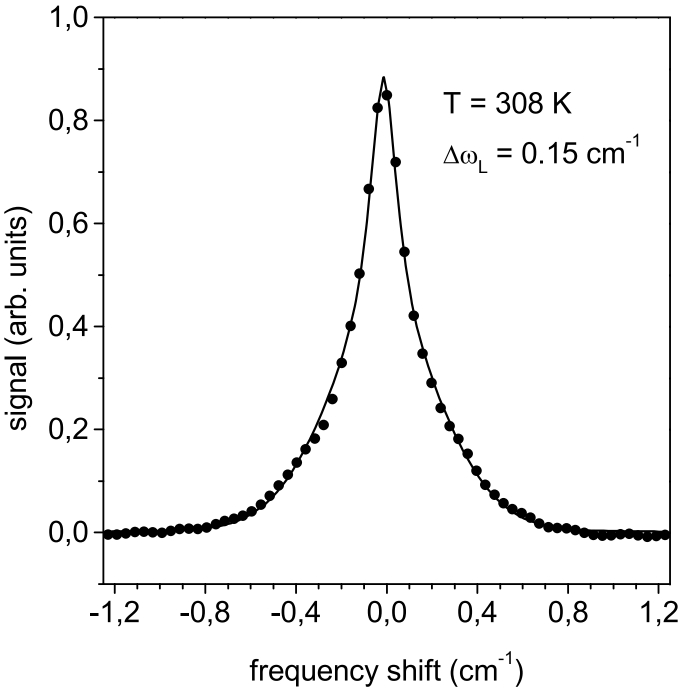


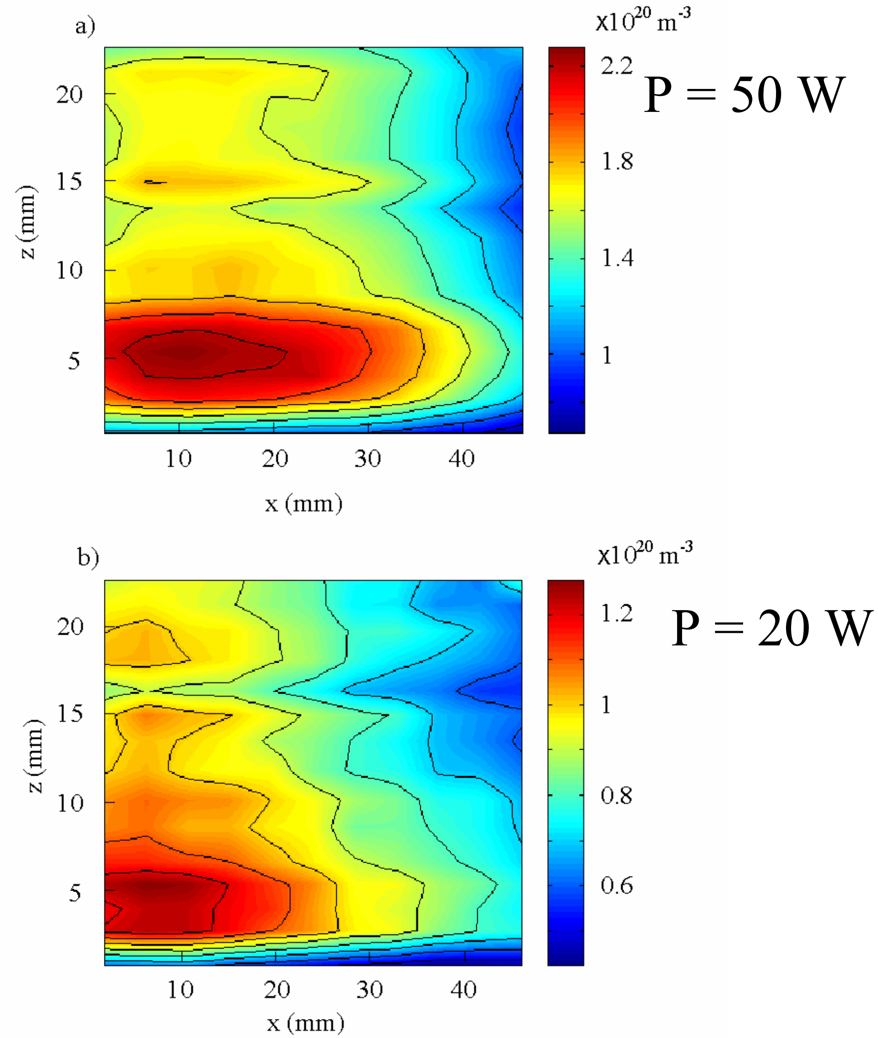
CF and CF_2 decay times behave very differently with pressure.
The solid lines represent fits of a diffusion-reaction model and the dashed lines show the calculated behaviour for perfect sticking at the electrodes.
The non-linear behaviour of CF is explained by a chemical volume reaction.

Two-Photon Absorption Laser-Induced Fluorescence Spectroscopy (TALIF)

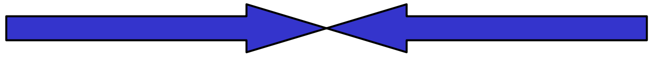
- The general principle is similar to LIF.
- However, two photons are absorbed at the same time.
- This allows excitation to higher states.
- Selection rules are different from single-photon transitions.
- Population in the upper state is now proportional to I^2.
- Much higher intensities are required.
- Saturation usually by three-photon ionization.
- Typically, maximum intensity of the order of 10^9 W/cm^2.

Atomic Hydrogen Densities in an RF Discharge





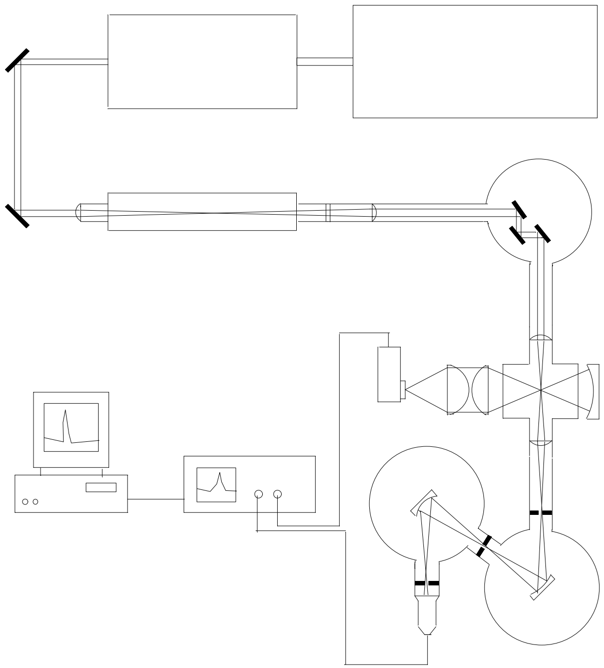
Doppler-free excitation to n=3 with 2 x 205 nm for increased sensitivity with an unfocused laser beam.

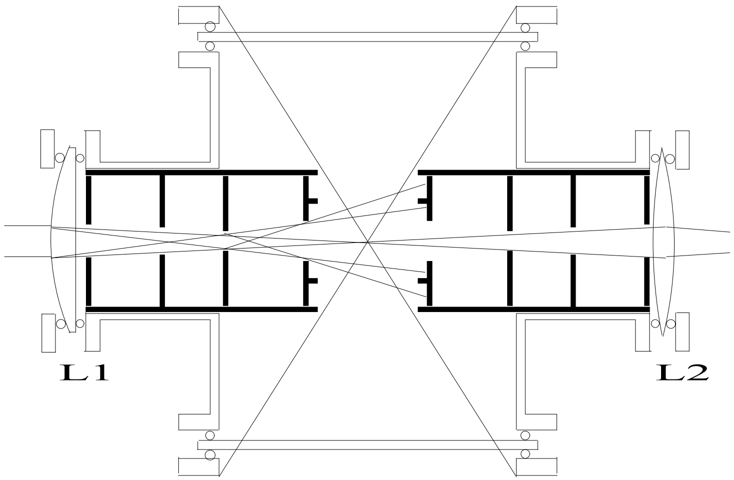


L. Cherigier, U. Czarnetzki, D. Luggenhölscher, V. Schulz-von der Gathen, and H.F. Döbele, J.Appl.Phys., **85** (1999) 696-702

Experimental Determination of Quenching Rates

In addition to radiative transitions, population can also be lost by collisions with other atoms or molecules. This radiationless loss of population is called quenching.



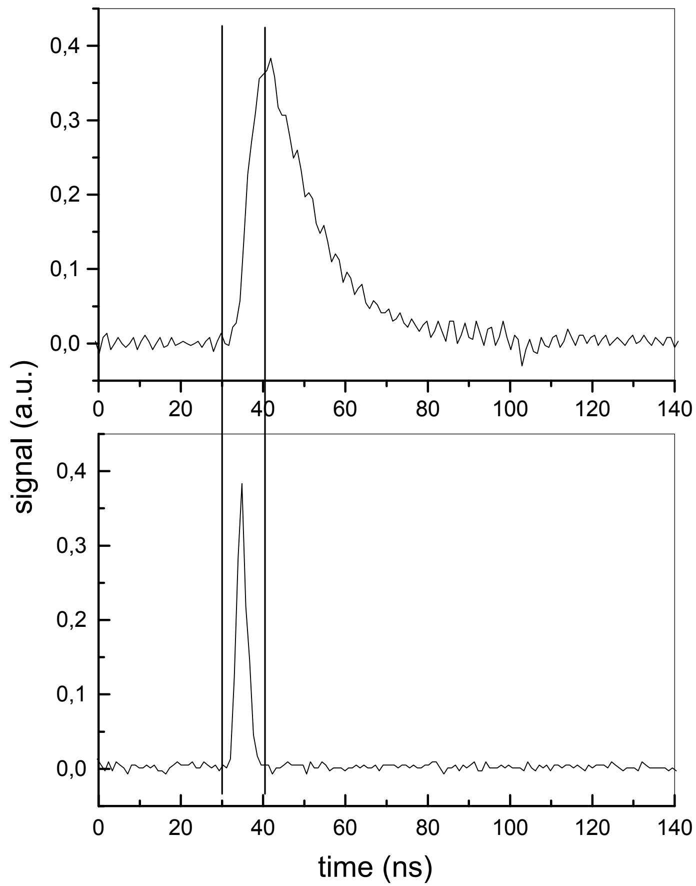


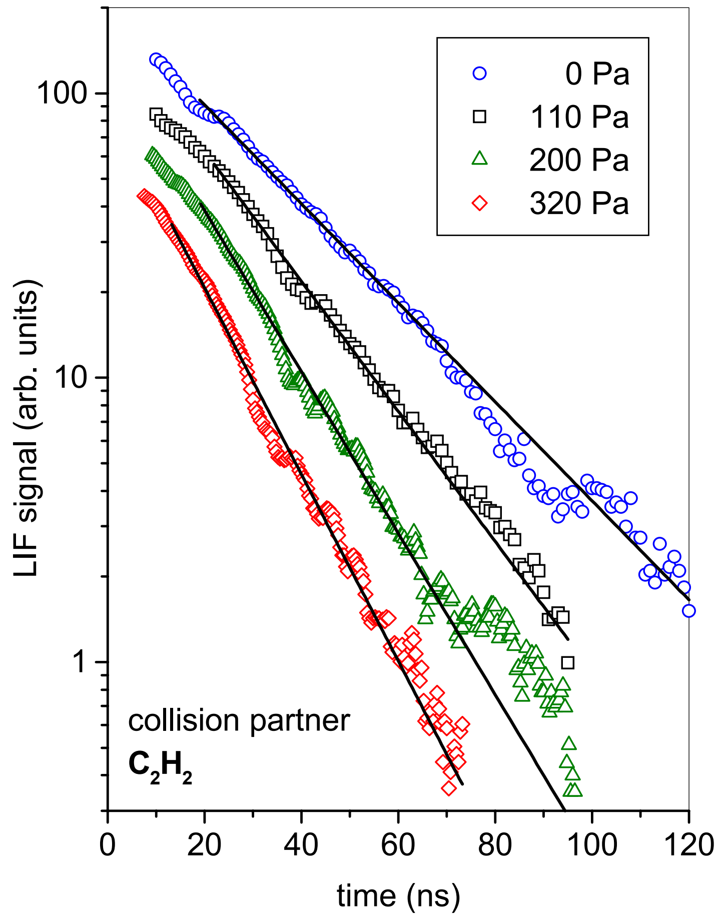
Argon two-photon excitation in the VUV.

λ (nm)	E (μJ)	I (MW/cm²)
184	9	50
186,8	0,3	2
188,3	0,5	3
189,4	0,6	3

N. Sadeghi, D.W. Setser, A. Francis, U. Czarnetzki, H.F. Döbele, J. Chem. Phys. 115, 3144 (2001)

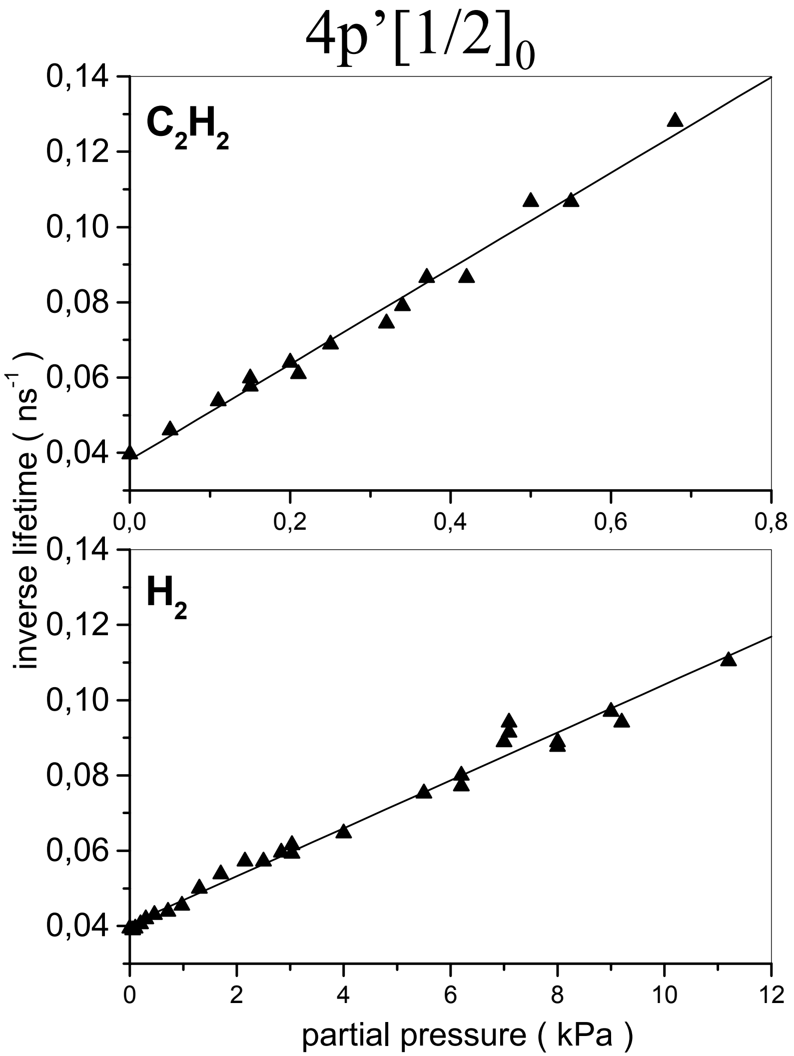
Determination of Lifetimes





Short (ns) excitation pulses are necessary.
The subsequent decay of the emission is exponential.
The decay time is reduced by quenching.

Stern-Volmer Plots



The slope gives directly the quenching rate coefficient k and the interception with the ordinate the radiative decay rate A_{total}.

$$\frac{1}{\tau_j} = \sum_i A_{ij} + k_{qj}\, n_q$$

Example:

With CH_4 at 100 Pa already 1/3 of the population in the excited Argon state (4p’[1/2]$_0$) is lost due to quenching.

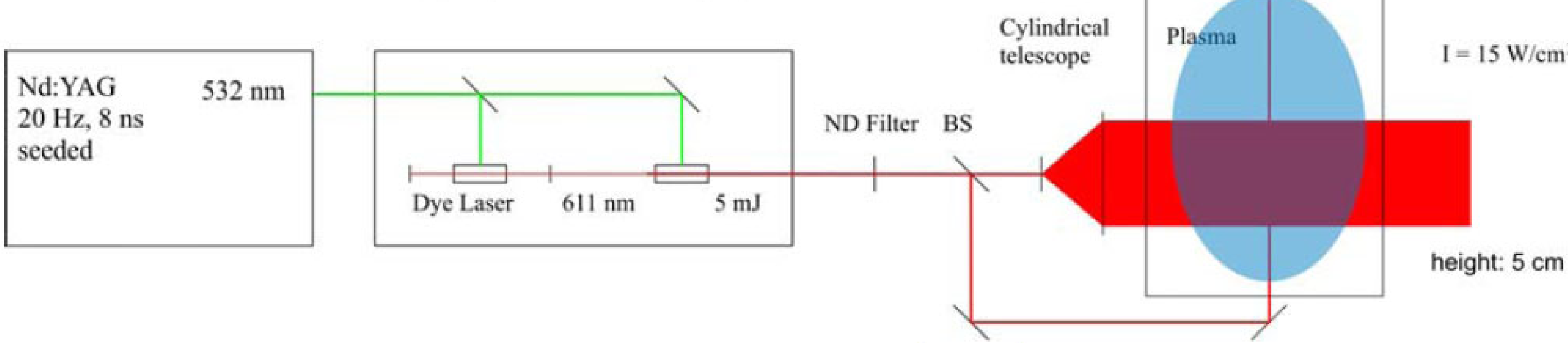
Ion Velocity Distribution Measurement

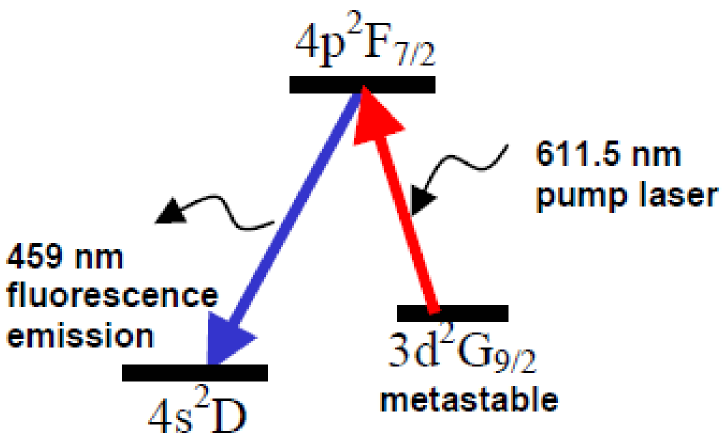
- Ion velocity distributions can be obtained by measuring the spectral excitation profile.
- The Doppler effect shifts the individual resonances and similar to Thomson scattering an ensemble distribution can be measured.

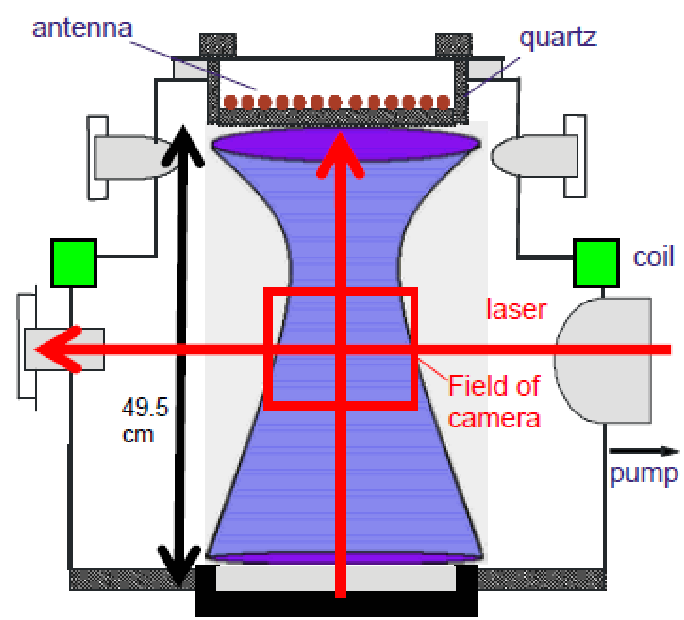
- Ion velocity distributions are important for understanding transport phenomena in plasmas.
- They are closely related to the electronic properties since electrons and ions have to diffuse at identical rates (at least globally).
- In equilibrium, ionization and particle loss must be balanced.

Experimental Setup and Excitation Scheme

Setup and Principle

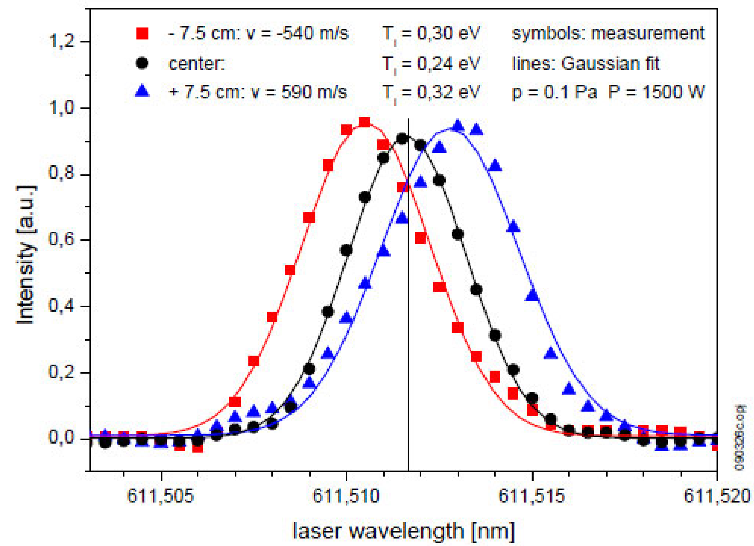


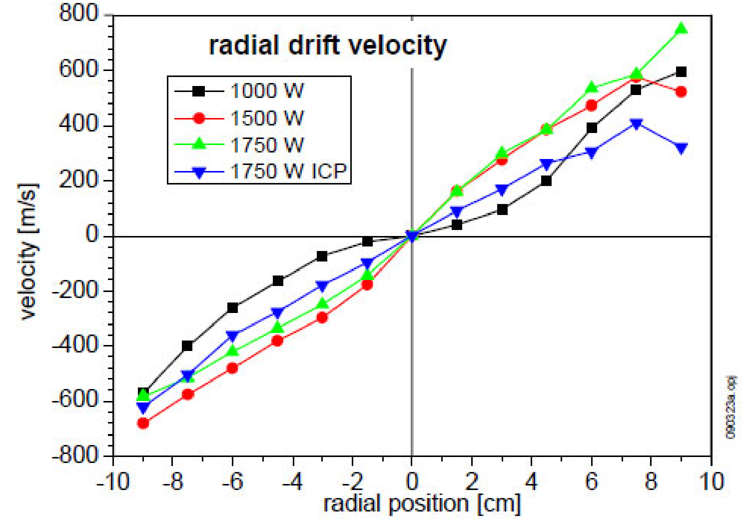


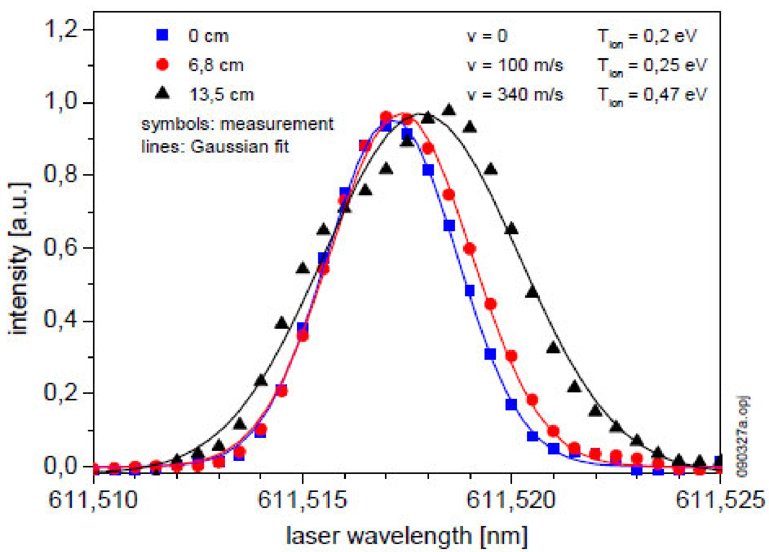


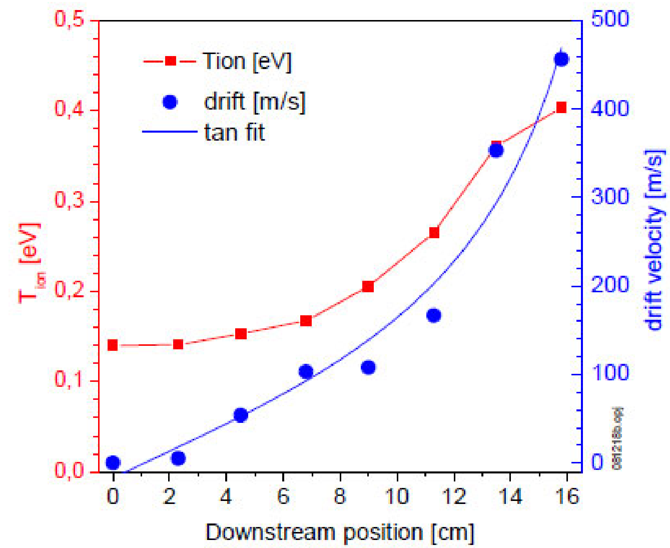
D Luggenhölscher, Y Celik, Y K Pu and U Czarnetzki, Journal of Physics: Conference Series **227** (2010) 012035

Radial and Axial Drift Velocities



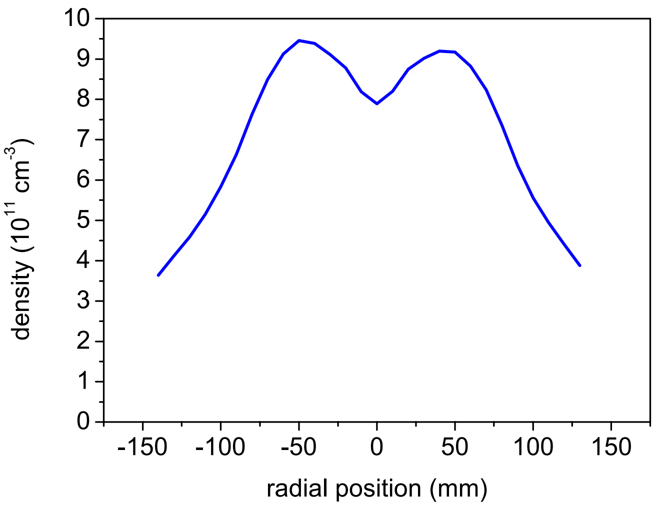


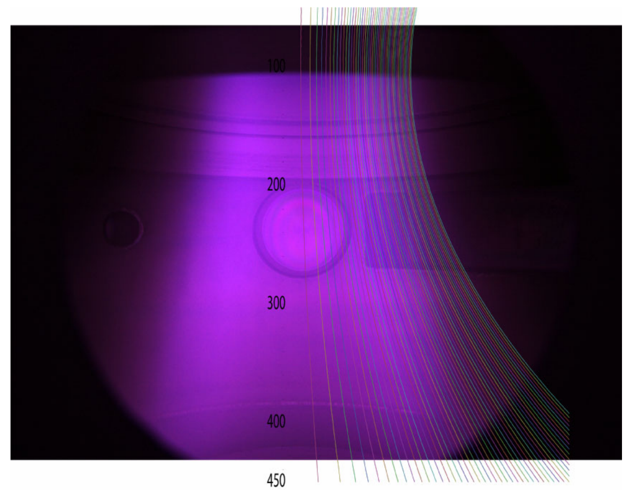




Transport Phenomena

The radial drift velocity increase per length is much higher than expected from the size of the chamber.
However, the radial density profile also decreases much faster.





This is due to confinement of electrons by the axial magnetic field and cooling during diffusion across the field.
Heating is only in the radial centre where the Helicon wave propagates.
Outside electrons are cold and ionization is no longer effective.
This causes the density profile to drop quickly and finally gives the radial shape a sharper profile. Consequently, ions are accelerated on a shorter distance by the ambipolar field build up in the density gradient.

Electric Field Measurement in Plasmas: Why are electric fields in plasmas important?

Electric fields are related closely to:

- power transfer to (heating of) electrons
- currents (electrons and ions), transport
- waves
- electron and ion energy distribution function
- ion bombardment of surfaces

Microfields are a measure of the plasma density.

$\Rightarrow$ Electric fields are a key parameter!

Electric Fields in Low Pressure Discharges

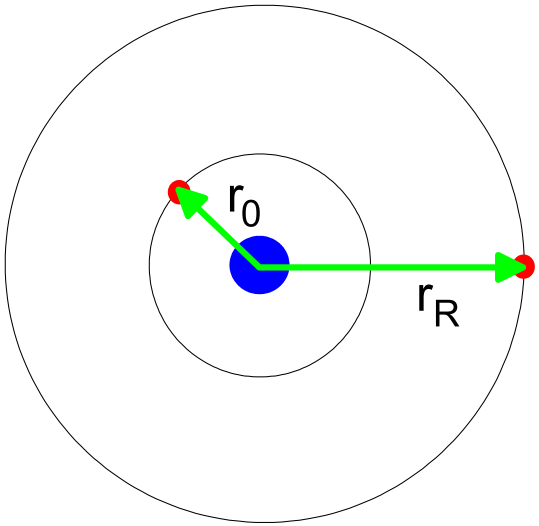
DC, HF, RF discharges:

- high-voltage sheath: 100 V/ cm - 1000 V/cm
- Debye sheath: 1 V/cm - 100 V/cm
- drift (induced) fields in the plasma bulk: 0.1 V/cm - 10 V/cm
- ambipolar fields in the plasma bulk: 0.1 V/cm - 1 V/cm

microwave fields: 10 V/cm - 100 V/cm

microfields: (stochastic fluctuations of the local charge density) 1 V/cm - 100 V/cm

Atomic Electric Fields (Hydrogen)



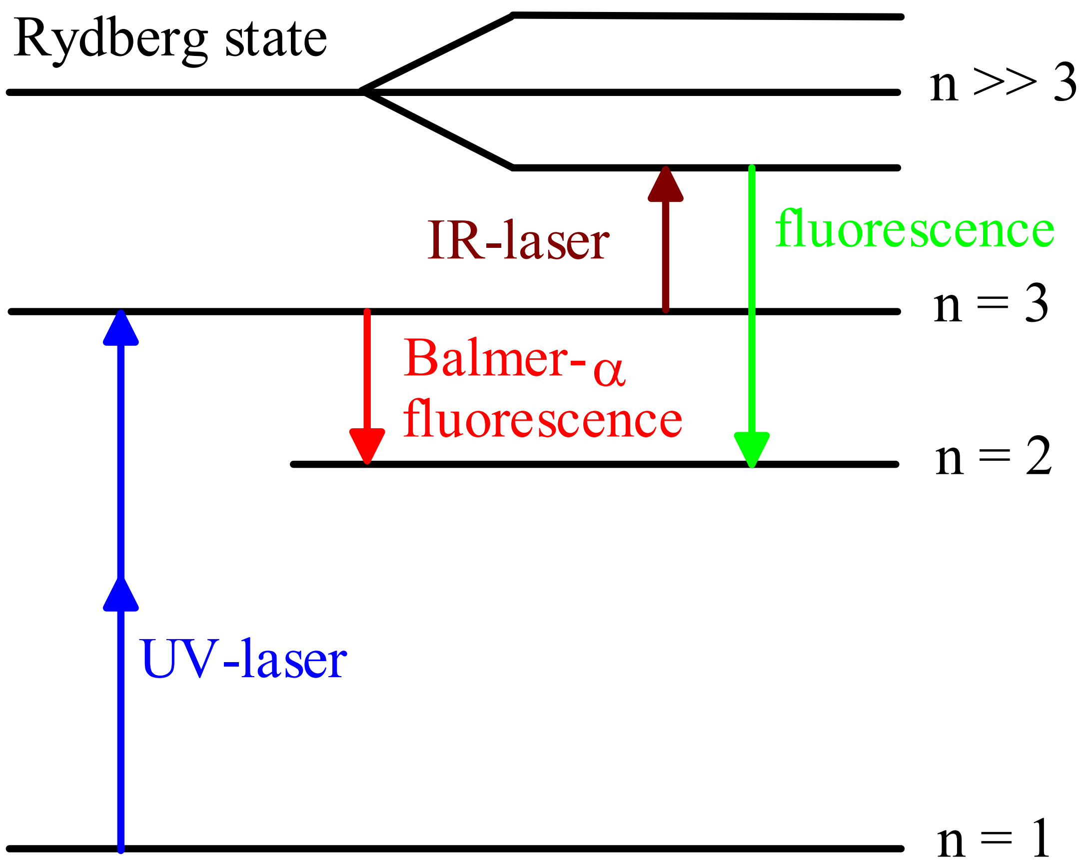
$$E(r) = \frac{e}{4\pi\varepsilon_0} \cdot \frac{1}{r^2}$$

$$\left\langle \frac{1}{r^2} \right\rangle \propto \frac{1}{n^2}$$

n = 1	r = 0.05 nm	E = 6 10^9 V/cm
n = 50	r = 125 nm	E = 900 V/cm

Rydberg states (n >> 1) are very sensitive to external fields.

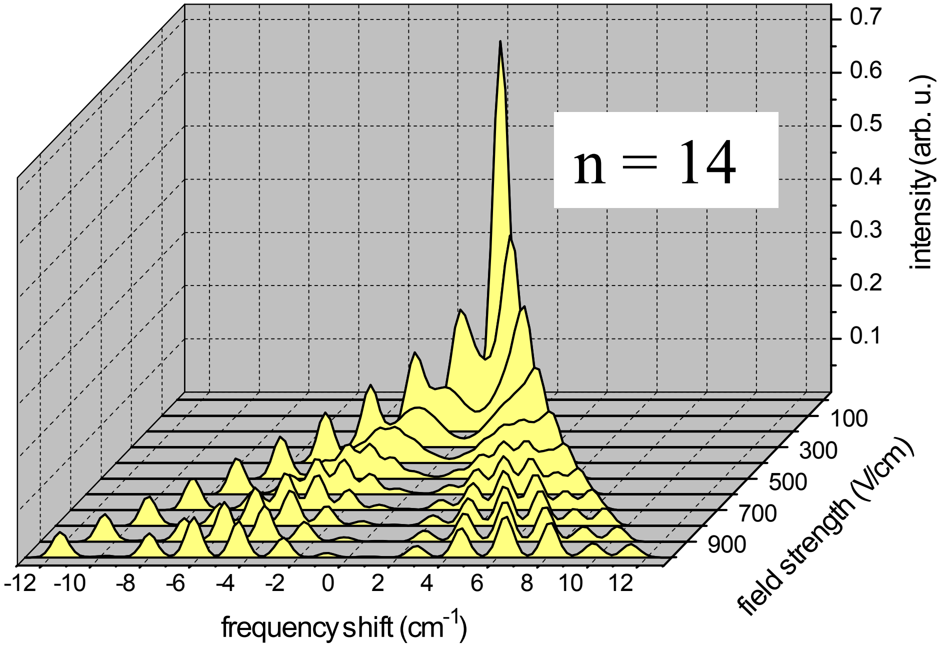
Spectroscopic Scheme: Fluorescence-Dip Spectroscopy



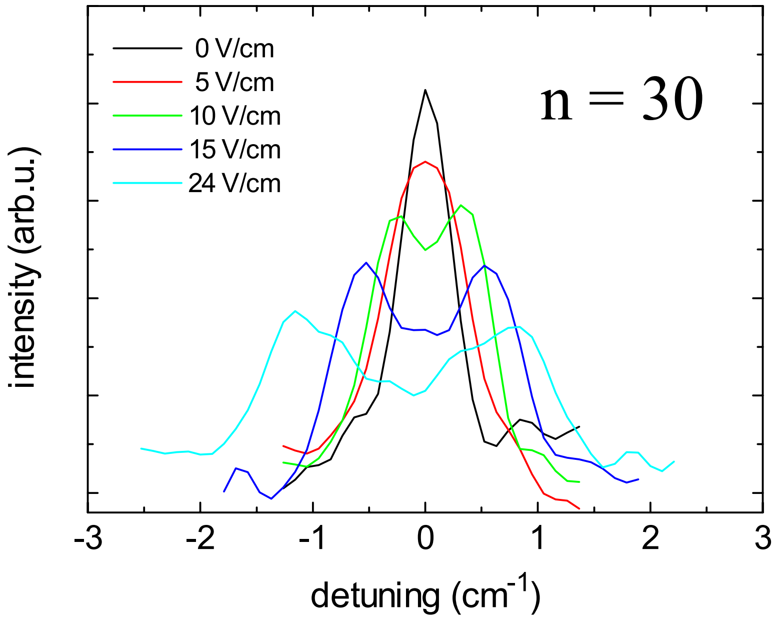
U. Czarnetzki, D. Luggenhölscher, and H.F. Döbele, Phys.Rev.Lett., **81** (1998) 4592

How is the measured spectrum related to a defined electric field?

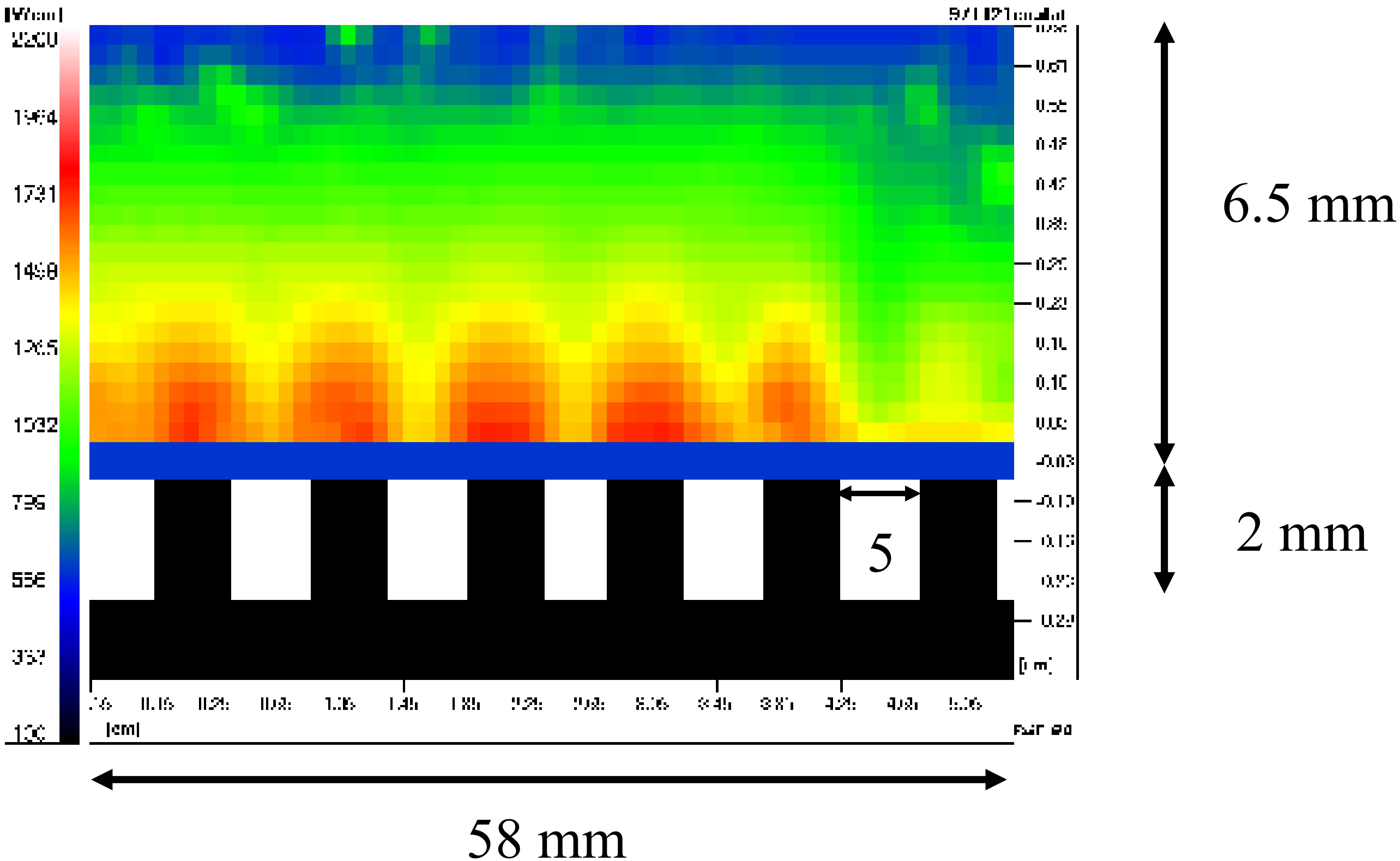
calculation



calibration

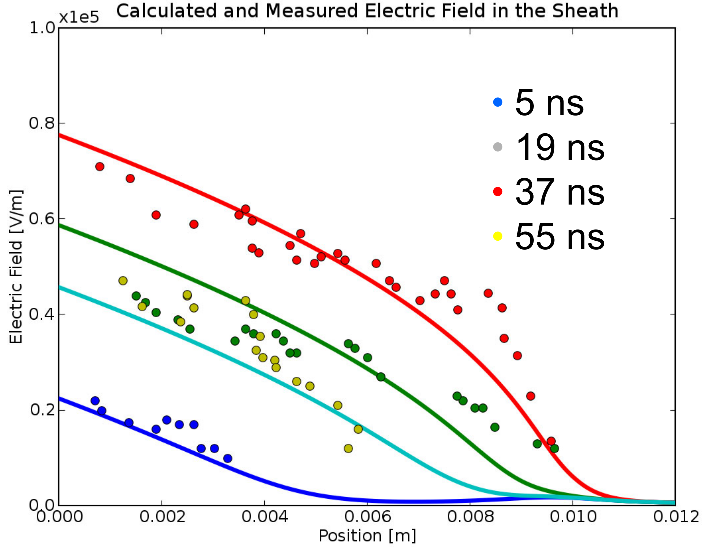


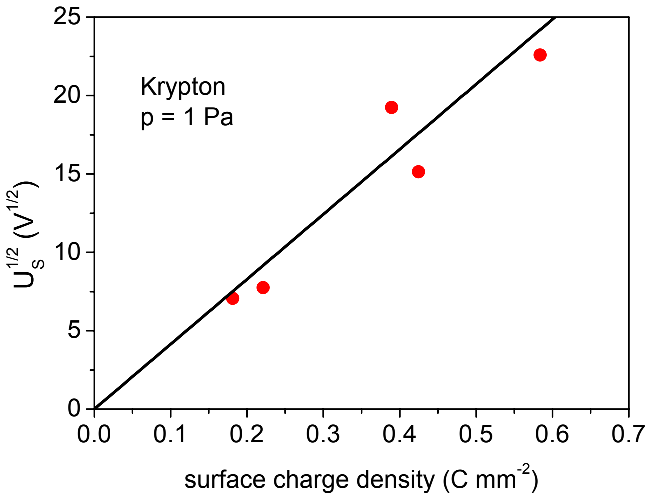
Two-Dimensional Structures in a CCP Discharge



U. Czarnetzki, G. A. Hebner, D. Luggenhölscher, H. F.Döbele, and M. E. Riley, IEEE Transactions on Plasma Science, **27**(1998) 70-71

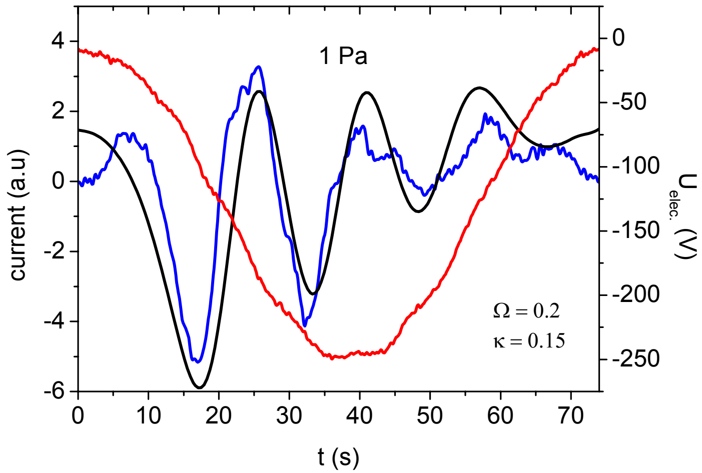
RF-CCP Sheaths: Measurement, Model, and Non-linear Oscillations





1 Pa Krypton

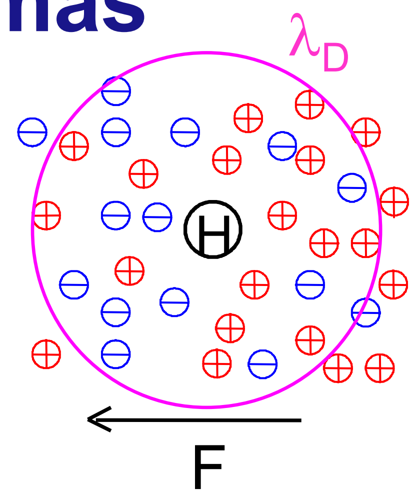
	Model	Experiment
T_e =	3.3 eV	3.5 eV
N_e =	1.8×10^9 cm^{-3}	2×10^9 cm^{-3}
V_{RMS} =	180 V	181 V

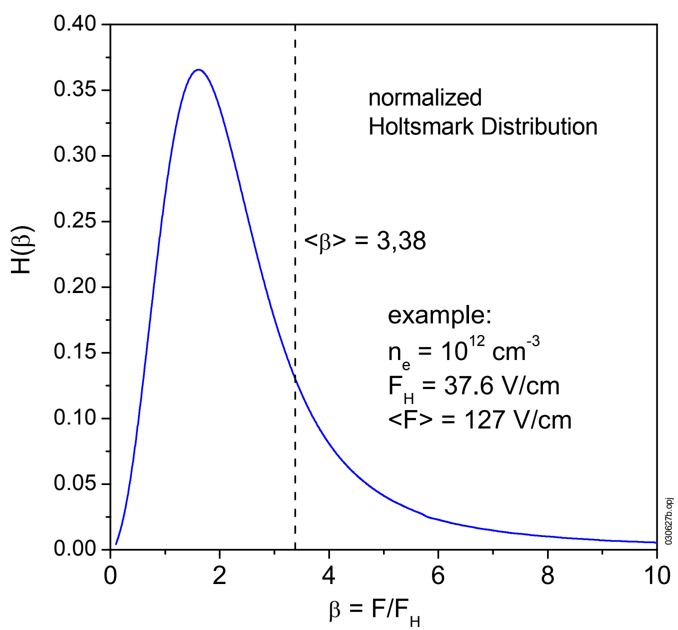


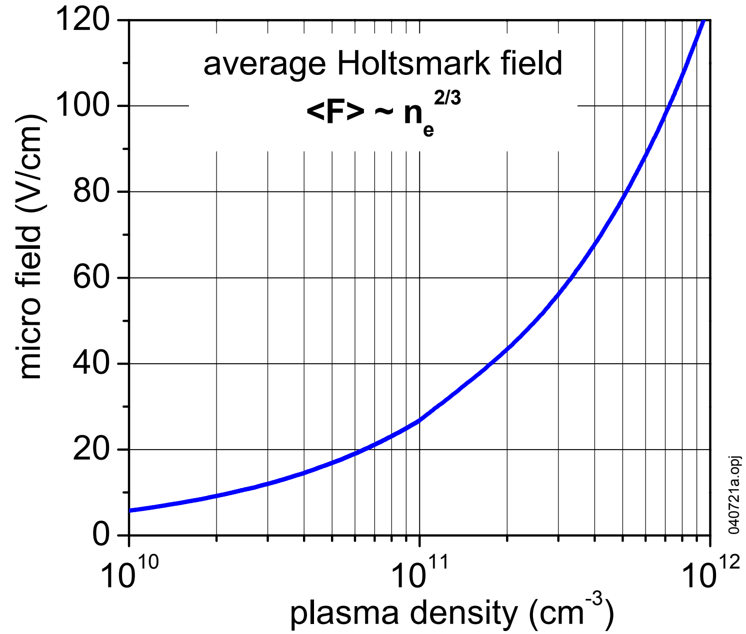
J. Schulze, B.G. Heil, D. Luggenhölscher, R.P. Brinkmann and U. Czarnetzki Journal of Physics D: Applied Physics 41, 195212 (2008)

Micro-Fields in Plasmas

- Random distribution of the charged particle distribution within the Debye sphere cause a local electric field distribution.
- Ions produce a quasi static field.
- Electrons act by collisions (negligible here).
- The ionic field distribution is described by the Holtsmark distribution.



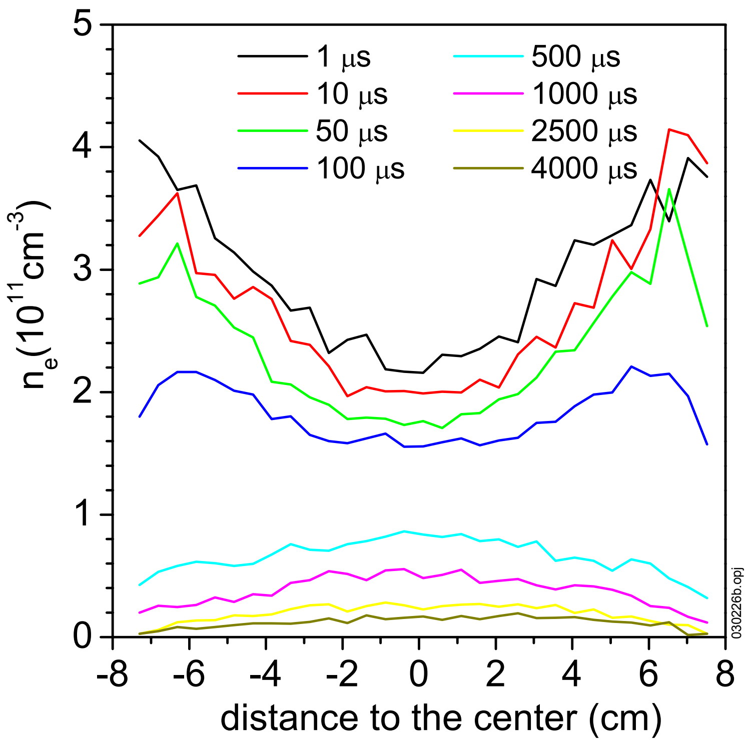


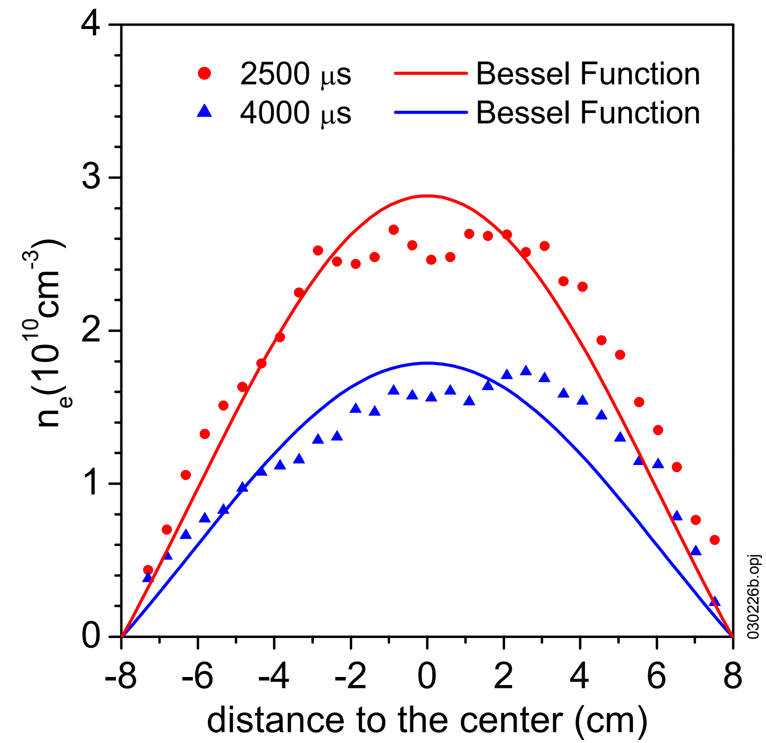


Density decay in argon dominated plasma

Argon dominated (90 % Ar, 10 % H_2): p = 30 Pa , f = 200 Hz, 20 % on

Argon needs less power ⇨ symmetric profile





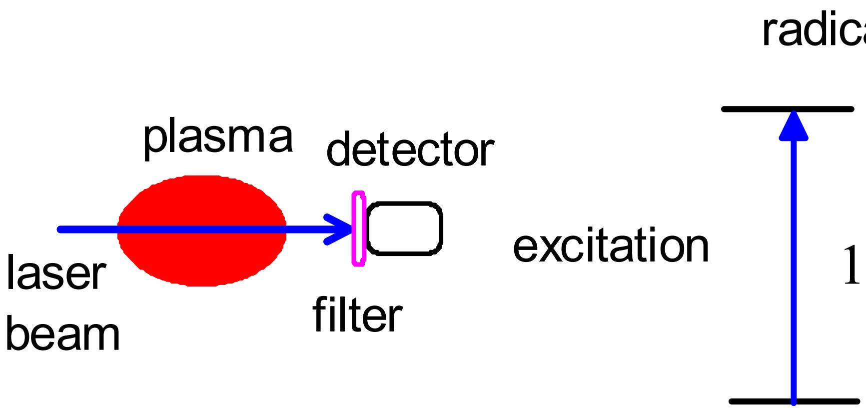
$$n(r) = n_0 J_0\left(2.41\frac{r}{R}\right)$$

U. Czarnetzki and K. Sasaki, Journal of Plasma and Fusion Research, 83, 215-227 (2007)

Outline

1) Introduction
2) Equipment
3) Thomson Scattering
4) Laser Induced Fluorescence Spectroscopy (LIF/TALIF)
5) Diode Laser Absorption Spectroscopy
6) Summary

Detection of Atomic and Molecular Radicals by Laser Absorption Spectroscopy

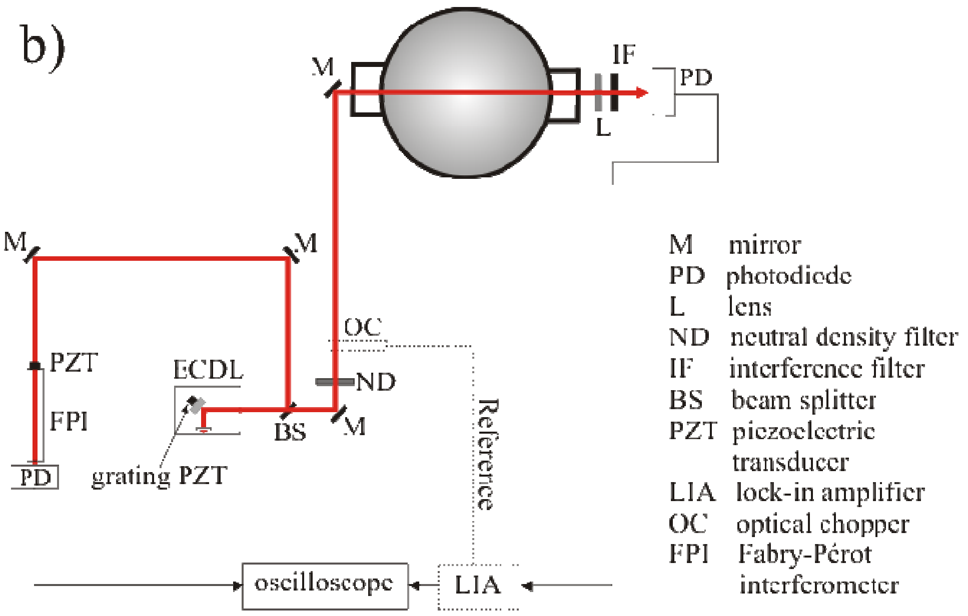


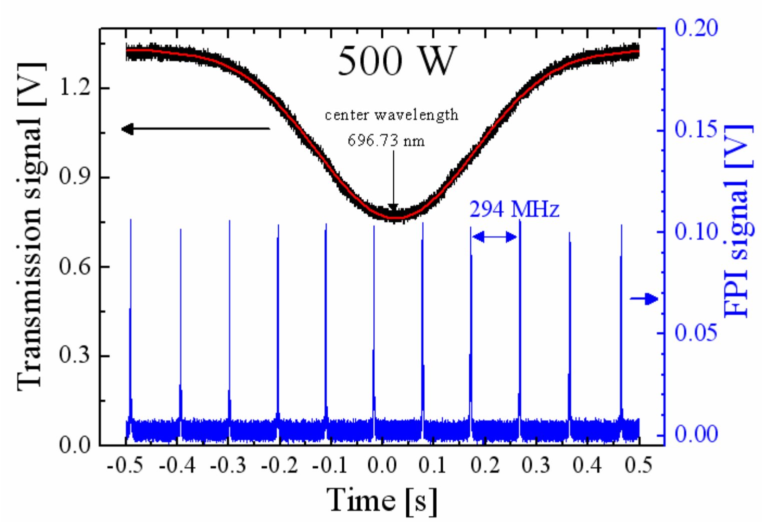
$$1-\frac{I(x)}{I_0}=1-e^{-x/\lambda}\approx\frac{x}{\lambda}=N\sigma x$$

small molecules with absorption from the UV to the IR

Absorption measurements provide information on the line-of-sight integrated densities. Knowledge of the absorption strength is required. Further knowledge about the spatial profile is at least useful if not obligatory for interpretation. Sometimes the profile can be inferred by linearly resolved measurements and Abel inversion.

Diode Laser Absorption Spectroscopy on Metastable Argon Atoms

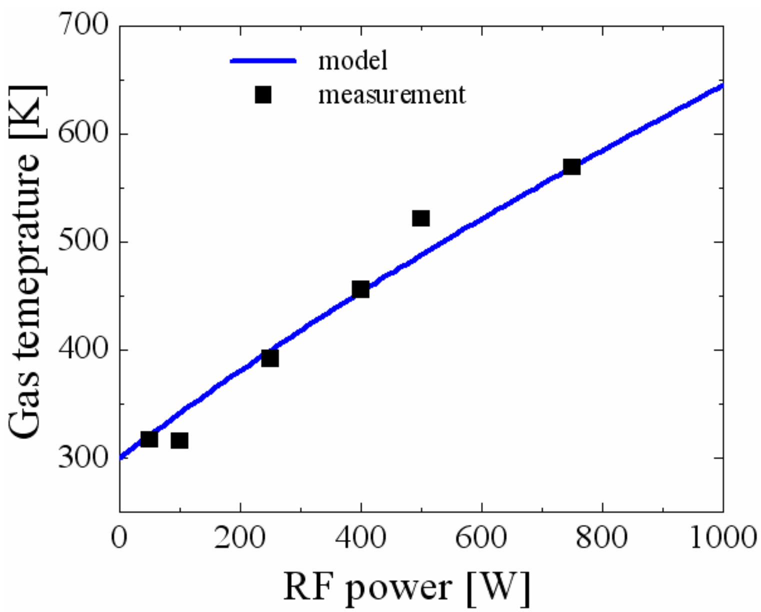


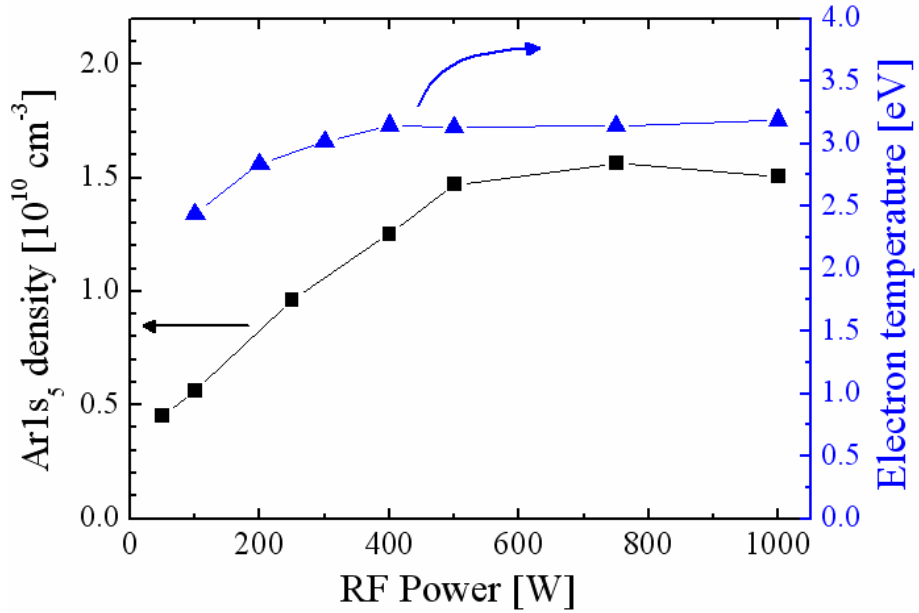


The measurement delivers directly the gas temperature (from the Doppler width) and the metastable density (from the area).

Y. Celik, M. Aramaki, D. Luggenhölscher, U. Czarnetzki, submitted to Plasma Sources Science and Technology (2010)

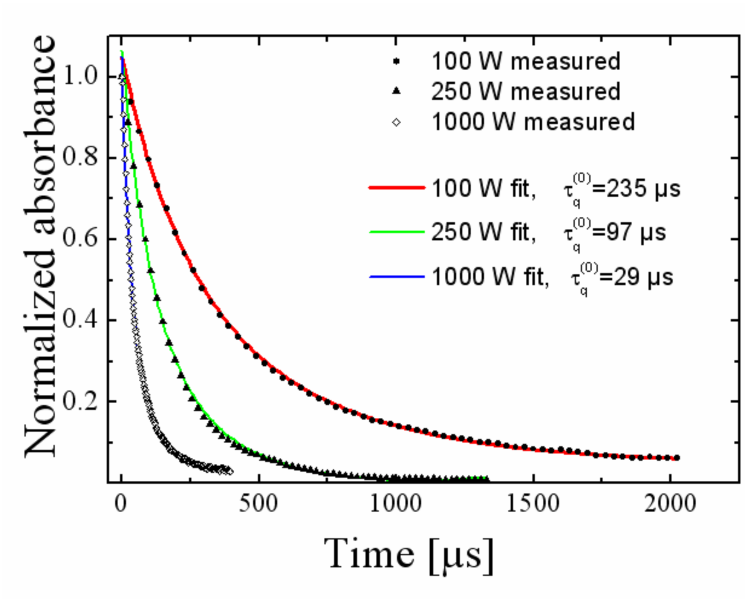
Direct Results

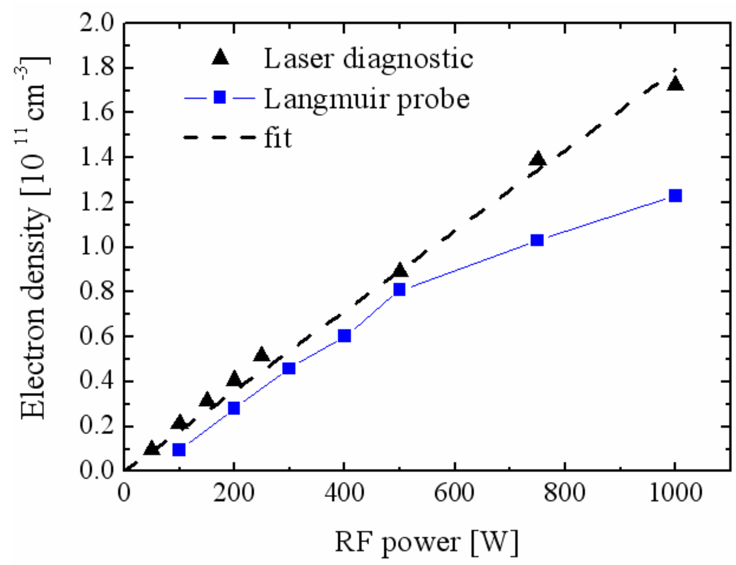




Gas temperature increase leads to reduction of the neutral gas density. This results in higher transport which must be balanced by higher electron temperature to increase ionization. Consequently also the metastable density increases.

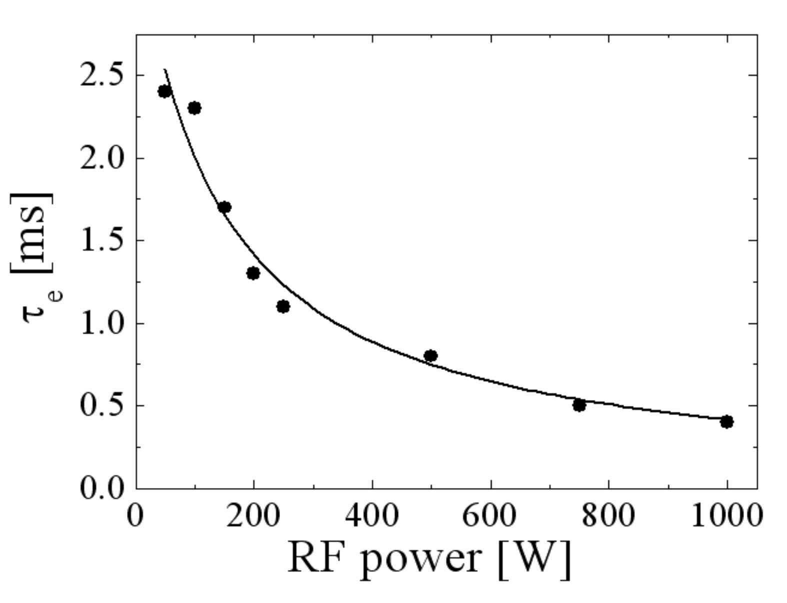
Electron Density Determination

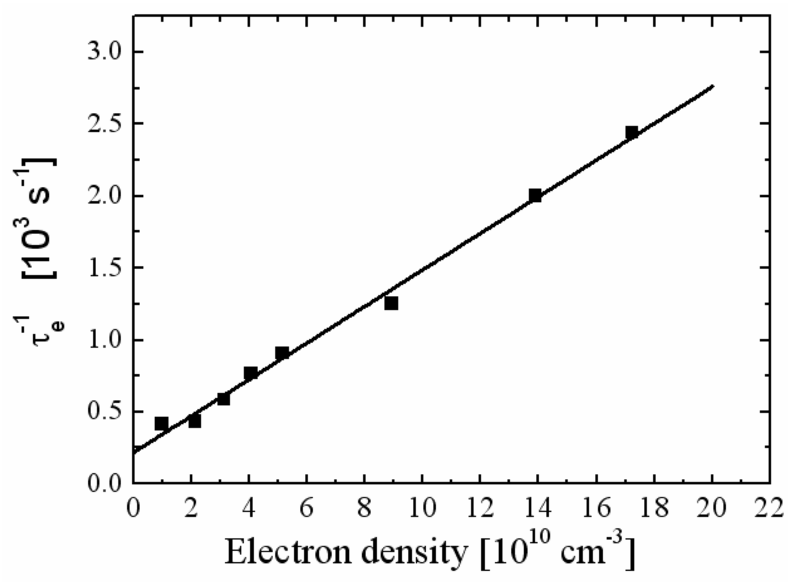




Pulsing of the discharge leads to rapid cooling of electrons in the afterglow by inelastic collisions. Then metastable production terminates but metastable quenching continues. The decay time is a measure of the electron density.

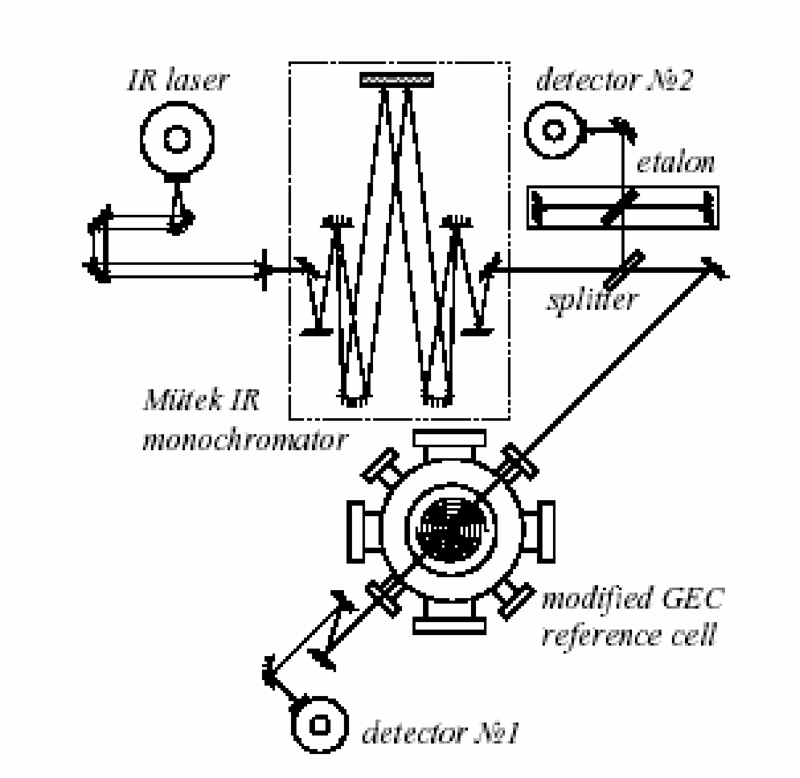
Electron-Ion Recombination and Diffusion





A model fit to the measurement yields also the relaxation time of the electron density τ_e. It is determined by diffusion to the wall and recombination. The slope of the inverse relaxation time is the recombination constant and the interception the inverse diffusion rate.

IRLAS in an ICP Discharge in CF_4



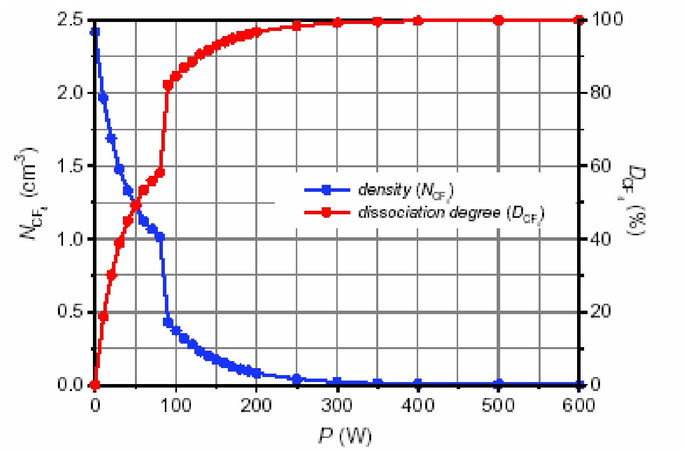
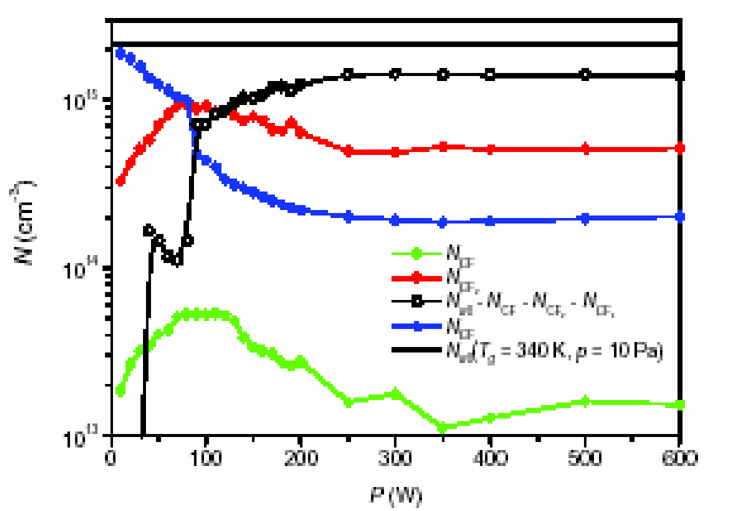


Fig.8. Concentration and dissociation degree of



By one diode laser measurement of CF, CF_2, (CF_3), CF_4 at around 1240 cm^{-1}.

V. A. Kadetov and U. Czarnetzki, internal report (2003)

Gas Temperature Effect in IRLAS

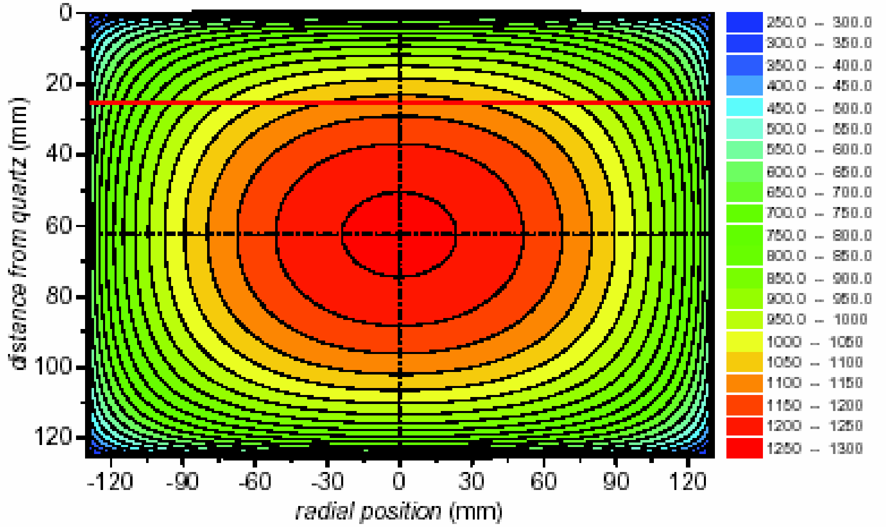


Fig.16. Calculation of the gas temperature profile in the GEC reference cell for ICP application (without bottom electrode).

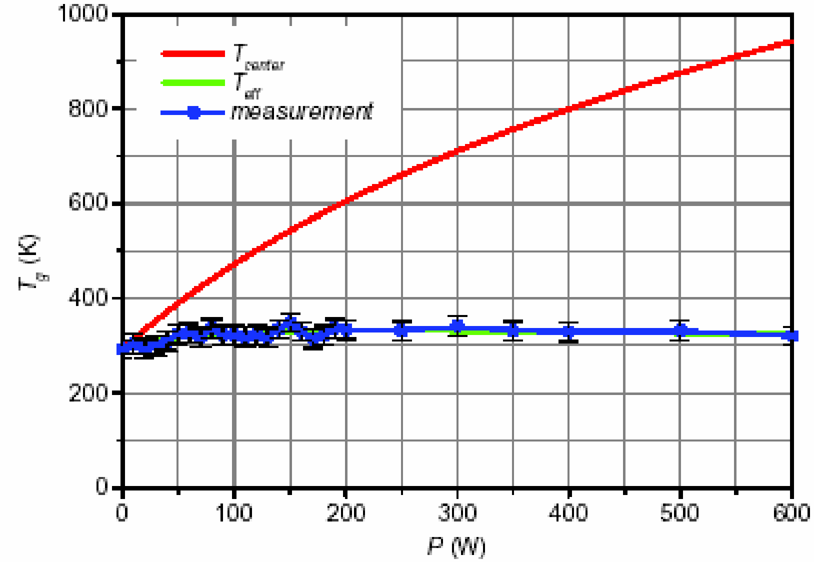


Fig.20. Gas temperature at the discharge axis and the temperature deduced from the CF4 absorption line profile.

$$-\nabla(T_g \nabla T_g) \sim \frac{P_\xi \overline{T}_g}{V T_g}.$$

$$\Rightarrow T_g \propto \sqrt{P}$$

Care has to be taken about inhomogeneous temperature distributions at high power densities. Then mostly the cold edge contributes to absorption.

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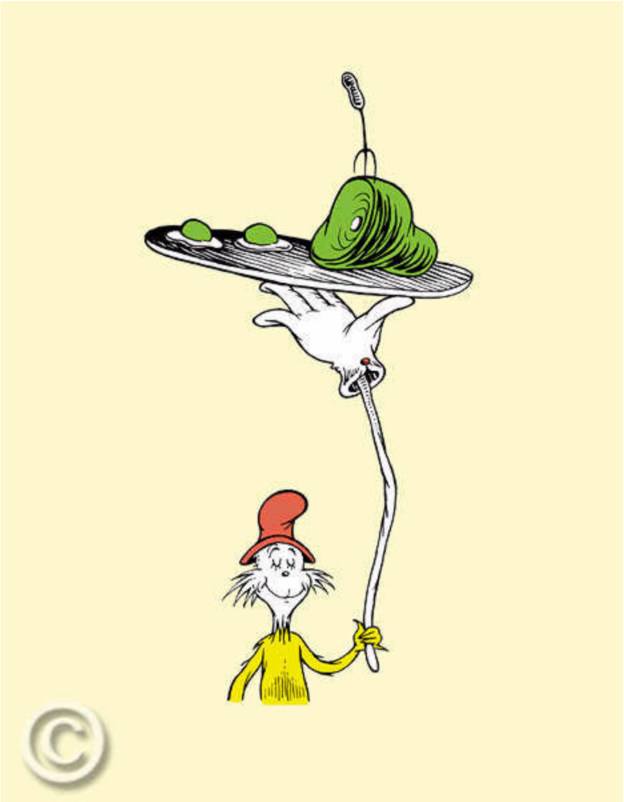
Summary

- Laser spectroscopic and optical measurements allow a detailed insight into physical processes in plasmas.
- Plasma parameters can often be determined directly.
- Combined with some simple model assumptions an even wider spectrum of physical quantities can be derived.
- A whole arsenal of techniques is available.
- There is still potential and need for further development of novel techniques.

Be smart, have fun, just do it!



"You will like them,
You will see…
Try them! Try them!
And you may.
Try them and you may, I say."

Dr. Seuss