

L'Université d'Orléans
des centres universitaires pour étudier et vivre

GREMI



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“Scattering” spectroscopy in thermal plasmas



Stéphane Pellerin

GREMI, University of Orleans/CNRS, Bourges, France



JAGIELLONIAN
UNIVERSITY
IN KRAKOW

Krzysztof Dzierzega

M. Smoluchowski Inst. of Physics, Jagellonian University, Cracow, Poland



Bartłomiej Pokrzywka

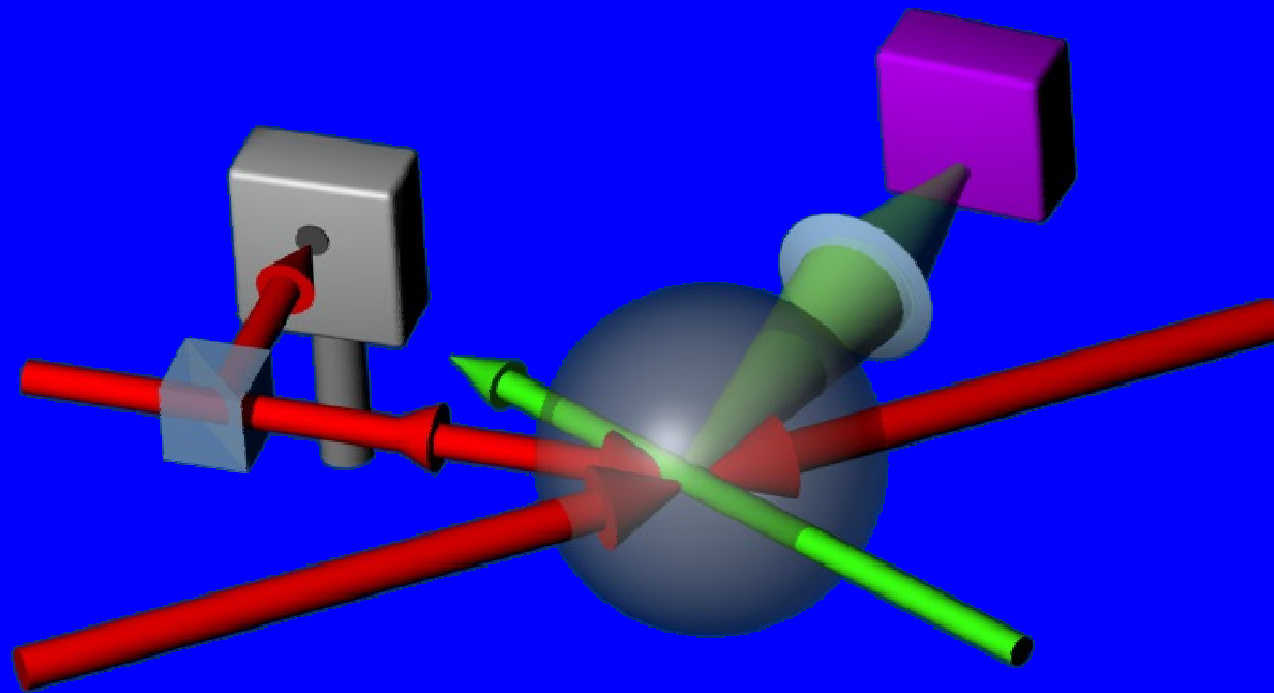
Mt. Suhora Obs., Inst. of Physics, Pedagogical Univ. of Cracow, Poland

Plan of the Presentation

1. Background: Spatially and Temporally resolved thermal plasma diagnostics
 - a. Thermal plasma
 - b. Methods of plasma diagnostics
2. A brief theoretical description
 - a. Scattering of waves in plasma: Rayleigh, Thomson
 - b. Thomson scattering in different plasmas
3. Thomson scattering in thermal arc plasma
 - a. Experimental procedure and data interpretation
 - b. Plasma disturbance by laser pulse
 - c. Spatial and Temporal averaging over laser pulse
4. Thomson scattering in laser induced plasma (LIB)
 - a. What is LIB ?
 - b. Laser induced sparks in gases: Ar, Air, He
 - Experimental procedure: Imaging, emission spectrum, data treatment
 - Plasma disturbance by laser pulse and Determination of initial T_e
 - Ion feature of TS spectrum: Determination of ion temperature
 - Rayleigh scattering: evolution of , shock wave
 - c. LIP generated on Al target
5. Summary and Conclusions

Background:

Spatially and Temporally resolved thermal plasma diagnostics



Thermal plasma

Isothermal balance close to the equilibrium $\Rightarrow T_e \approx T_h$

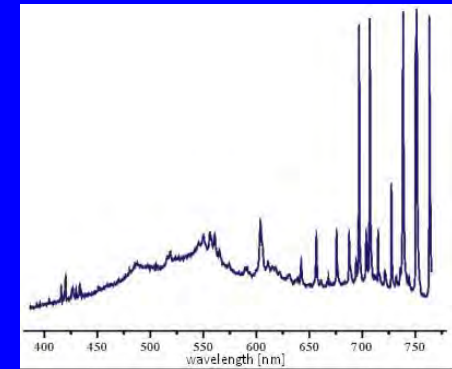


High electron density, pressure, temperature and ionization degree

For argon arc plasma at $p = 10^5$ Pa LIE is achieved for $N_e \sim 10^{22} \text{ m}^{-3}$ $T_e \sim 10^4$ K

Spectrum

- intense continuous radiation
- lines appreciably broadened due to Stark broadening
- important contribution of Doppler broadening

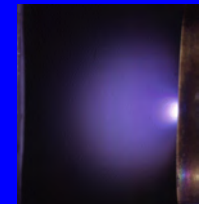


Examples analyzed in this talk:

Arc plasma at atmospheric pressure – Stationary plasma



Plasma induced by pulsed laser – Transient plasma



Plasma diagnostics requirements

Main goal:

Plasma parameters:
Plasma composition

N_e, T_e, T_h
(Quantitative analysis)

Ideal method:

- Temporal resolution adequate to a time scale of the plasma evolution
- **Spatially resolved - good spatial resolution necessary to map plasma parameters**
- Non-intrusive → plasma state not disturbed
- **No assumptions about the plasma state**

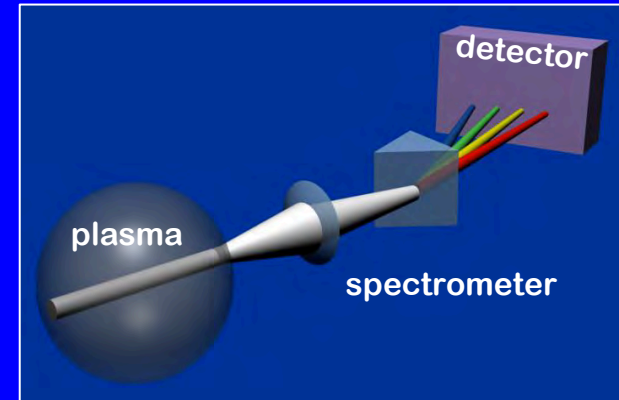
There is no such method which meets all of these requirements

Inhomogeneous plasma ⇒ Spatial resolution is crucial factor affecting quality of diagnostics

Methods of plasma diagnostics

Optical emission spectroscopy (passive)

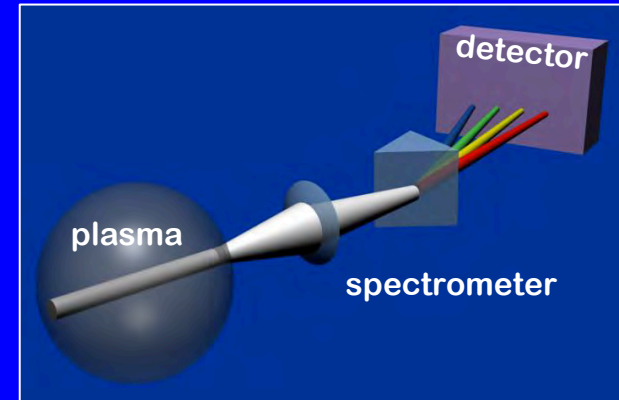
- ☹️ **only spatially integrated intensity along the line of sight can be directly measured** (local values need Abel transformation)
- possible reabsorption
- *only excited particles can be probed*
- *required assumptions about the plasma equilibrium state*



Methods of plasma diagnostics

Optical emission spectroscopy (passive)

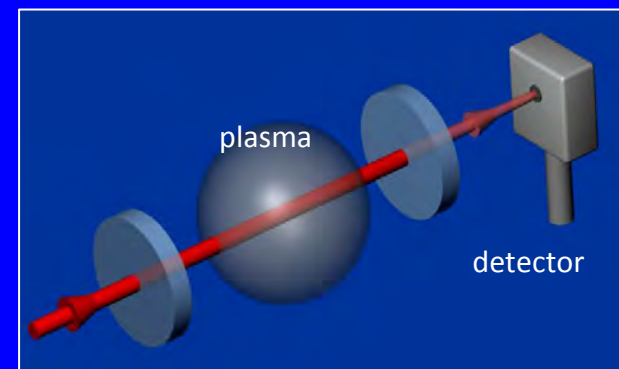
- ☹️ **only spatially integrated intensity along the line of sight can be directly measured** (local values need Abel transformation)
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Methods based on transmitted light analysis (active)

- ✓ Laser absorption spectroscopy
- ✓ Polarization spectroscopy
- ✓ Cavity ring-down absorption
- ✓ Interferometry

☹️ **Non-local methods!**



Spatially resolved laser based techniques



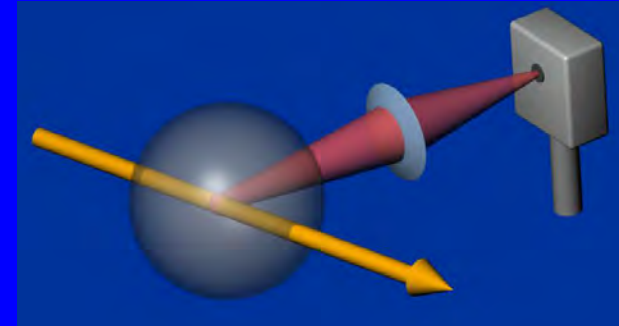
Laser based, local methods

✓ Laser induced fluorescence (LIF, TALIF)

☹ **Not useful at high electron density**

- ✦ Signal diminished by non-radiative decay
- ✦ Masked in strong plasma radiation background

Pump – probe technique



Spatially resolved laser based techniques



Laser based, local methods

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✓ Non-linear spectroscopy

FWM: Coherent third order non-linear process

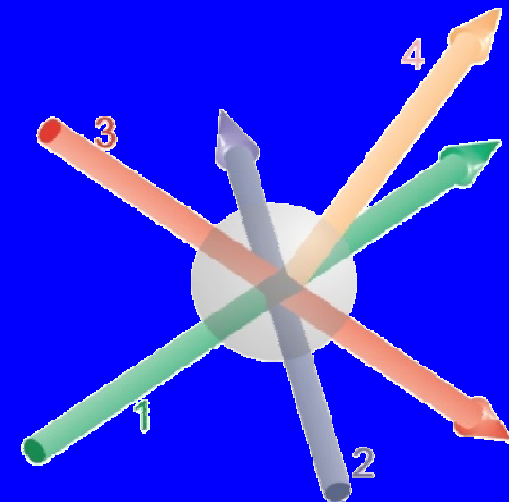
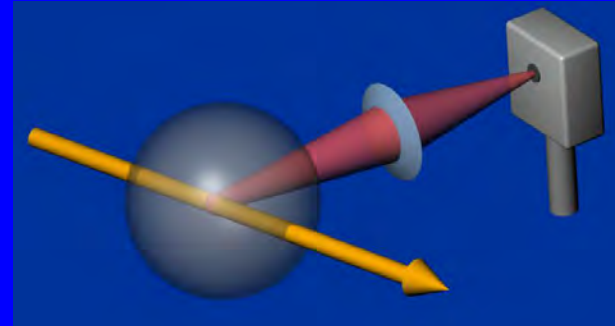
$$\omega_4 + \omega_3 = \omega_1 + \omega_2$$

$$\vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{k}_4$$

$$\omega_4 = \omega_1 + \omega_2 - \omega_3$$

Phase matching condition

Pump – probe technique



Spatially resolved laser based techniques



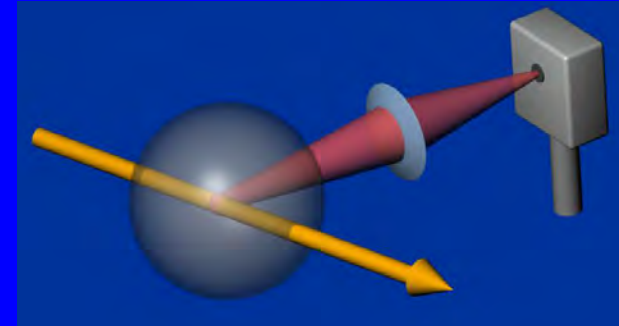
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Pump – probe technique



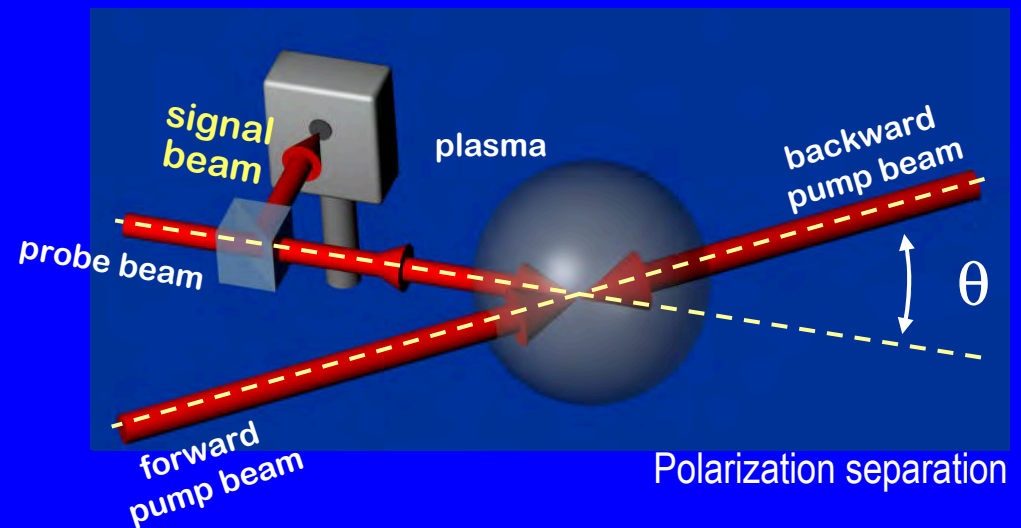
✓ Non-linear spectroscopy → PC-Degenerate Four Waves Mixing

$$\omega_1 = \omega_2 = \omega_3 = \omega_0 \Rightarrow \omega_4 = \omega_0$$

Degenerate mixing

$$\vec{k}_1 = -\vec{k}_2 \Rightarrow \vec{k}_4 = -\vec{k}_3$$

Phase-conjugate



Spatially resolved laser based techniques



Laser based, local methods

✓ Laser induced fluorescence (LIF, TALIF)

☹ **Not useful at high electron density**

- ✦ Signal diminished by non-radiative decay
- ✦ Masked in strong plasma radiation background

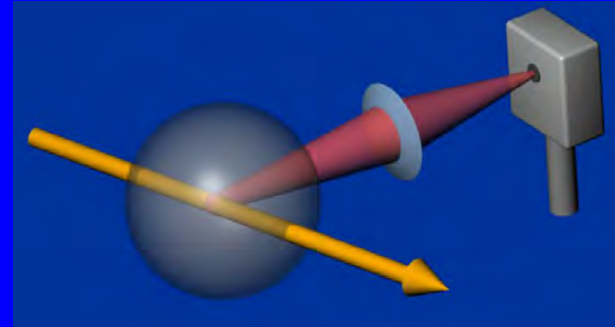
✓ Non-linear spectroscopy

✓ Scattering of laser radiation

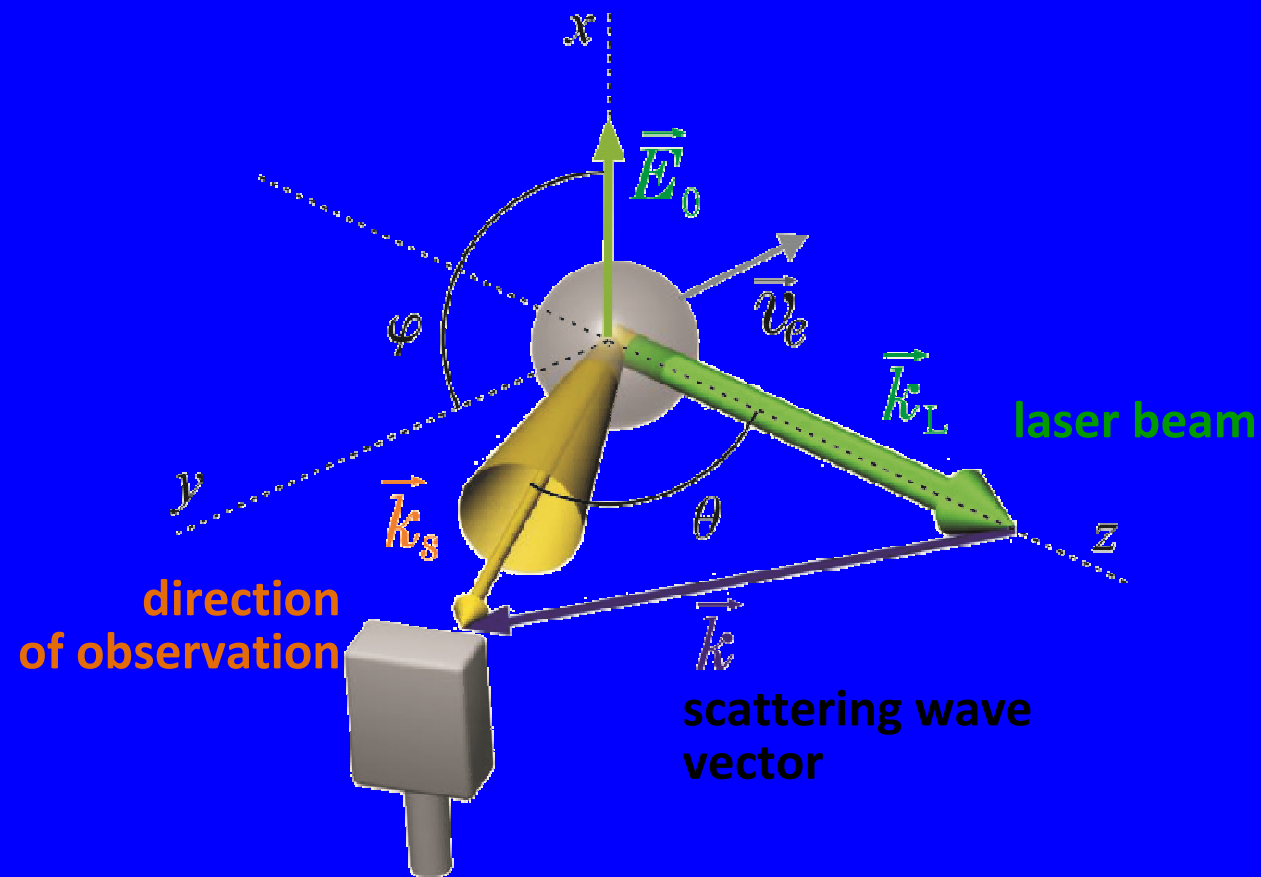
Remains to consider:

☺ **Thomson scattering**

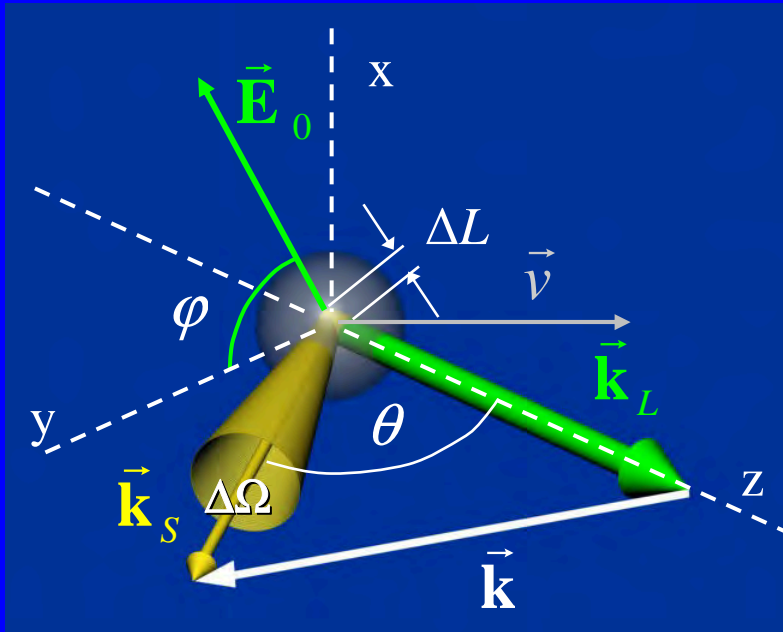
Pump – probe technique



Scattering of laser radiation: A brief theoretical description



Non-relativistic scattering of laser radiation



$$\Delta P_S = P_L \Delta L \Delta \Omega N \frac{d\sigma}{d\Omega} \underbrace{S(\mathbf{k}, \Delta\omega)}_{\text{spectral density function}} \Delta\omega_S$$

$$\frac{d\sigma}{d\Omega} = \frac{3}{8\pi} \sigma (1 - \sin^2 \theta \cos^2 \varphi)$$

spectral density function

$$|\mathbf{k}| = |\mathbf{k}_S - \mathbf{k}_L| \cong \frac{4\pi}{\lambda_L} \sin(\theta/2) \quad \text{scattering vector}$$

Motion of scattering centers

$$\Delta\omega \equiv \omega_L - \omega_S = (\mathbf{k}_S - \mathbf{k}_L) \cdot \mathbf{v} = \mathbf{k} \cdot \mathbf{v}$$

Elastic scattering

- Rayleigh scattering
- Thomson scattering

Inelastic scattering

- Raman scattering

Non coherent scattering on an ensemble

$$S_k(\Delta\omega) d\omega_s = F_k(v_k) dv_k = k^{-1} F_k(\Delta\omega/k) d\omega_s$$

Spectral density function reproduces velocity distribution of moving scattering centers

From Rayleigh, Thomson and Raman scattering we have information on electron and heavy particles density and their temperatures → PLASMA DIAGNOSTICS

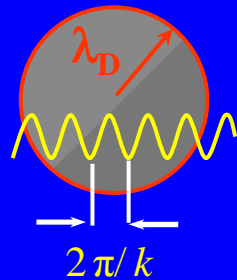
Incoherent and coherent Thomson scattering

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

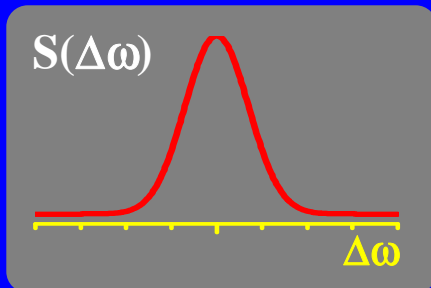
$$\lambda_D = \left(\frac{\epsilon_0 k_B T_e}{e^2 N_e} \right)^{1/2}$$

$$\widehat{\omega}_{pl} = \sqrt{e^2 n_e / (\epsilon_0 m_e)}$$

$\alpha \ll 1 \rightarrow$ Non-collective scattering

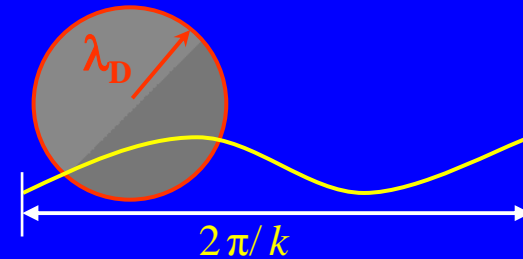


Spectral distribution gives EEDF



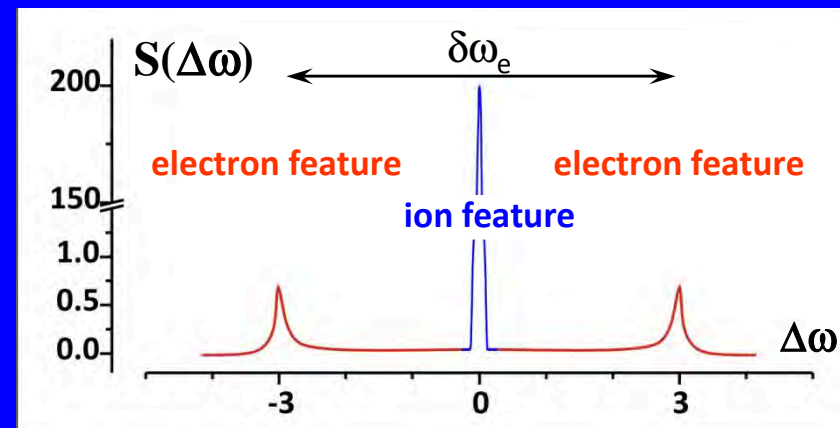
**At optical wavelength
and thermal plasma $\alpha \sim 1$**

$\alpha \gg 1 \rightarrow$ Collective / Coherent scattering



- ✓ Coupling with Langmuir plasma waves
- ✓ Coupling with ion acoustic waves

$$\delta\omega_e = \sqrt{\widehat{\omega}_{pl}^2 + 3k^2 k_B T_e / m_e} \quad \delta\omega_i = k \sqrt{2k_B T_i / M_i}$$

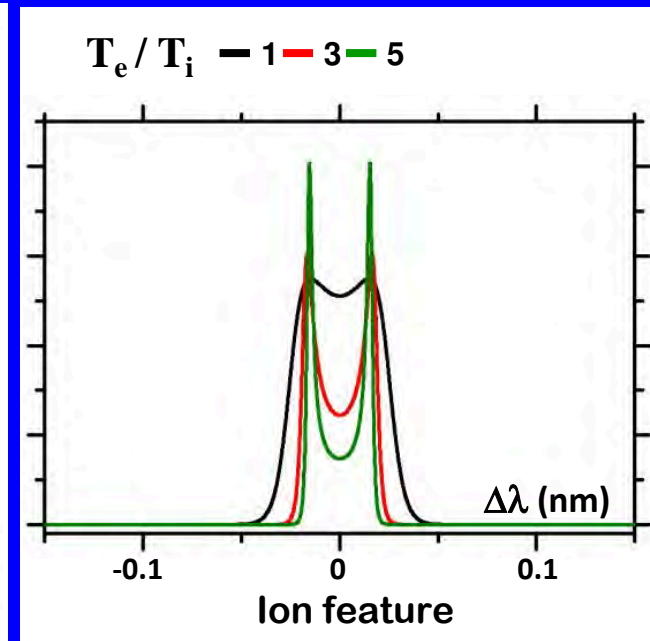
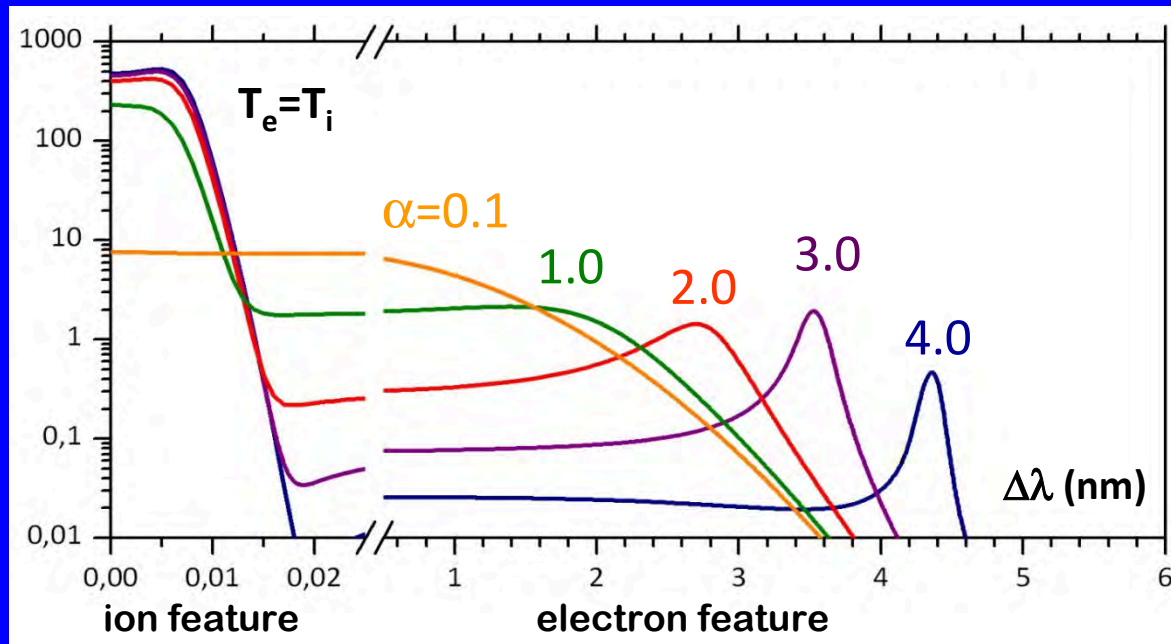


Thomson scattering – Partially collective case ($\alpha \approx 1$)

$$S(k, \Delta\omega) = \frac{1}{k v_e} \Gamma_\alpha(\Delta\omega/k v_e) + \frac{1}{\sqrt{\pi} k v_i} \frac{\alpha^2}{1 + \alpha^2} \Gamma_\beta(\Delta\omega/k v_i)$$

electron feature

ion feature



- ☺ Electron feature contains information about electron density and electron temperature.
- ☺ Ion feature contains information about ratio of electron temperature and heavy species temperature.
- ☺ Straightforward interpretation, no LTE assumption required

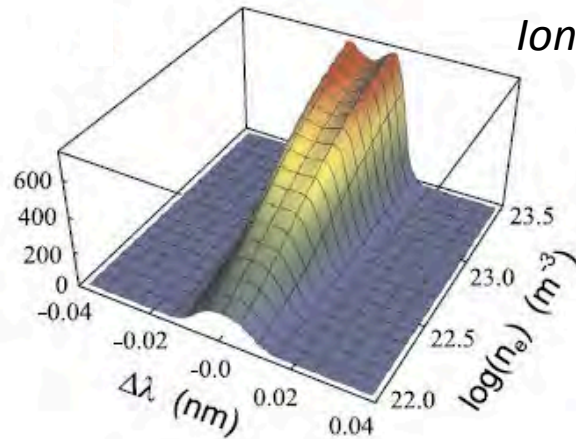
Thomson scattering – Partially collective case ($\alpha \approx 1$)

↳ The spectral form factor

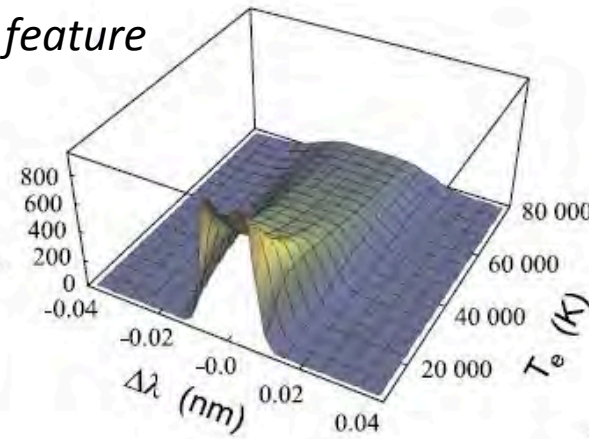
$T_e = 20\,000\text{ K}, T_e/T_i = 1$

$n_e = 1.0 \times 10^{23}\text{ m}^{-3}, T_e/T_i = 1$

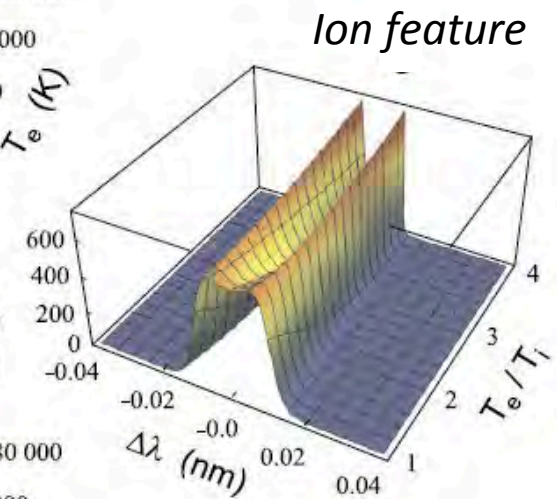
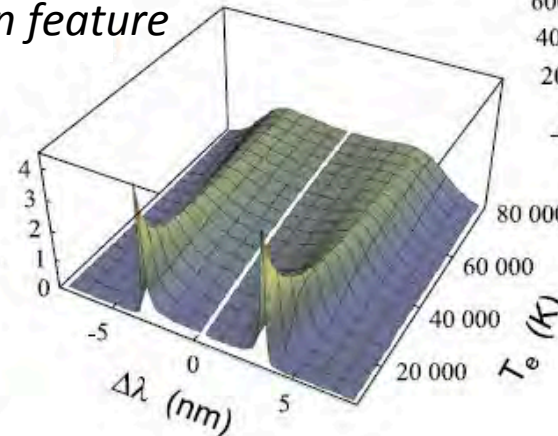
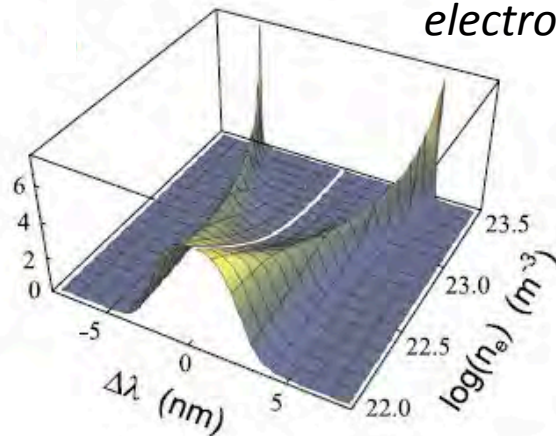
$T_e = 20\,000\text{ K}$
 $n_e = 1.0 \times 10^{23}\text{ m}^{-3},$



Ion feature



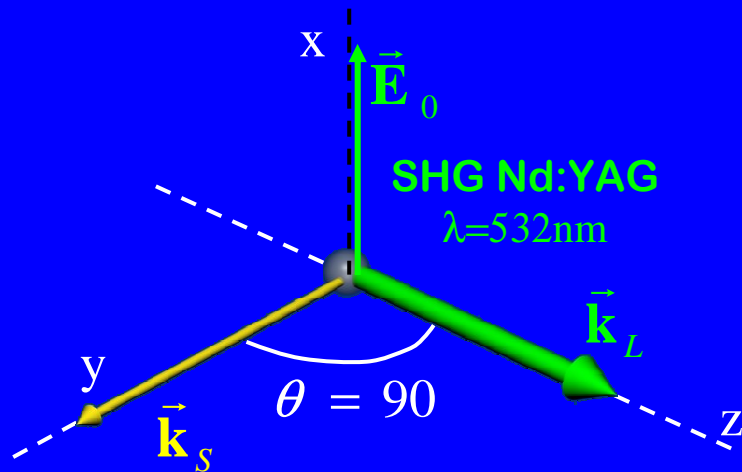
electron feature



Ion feature

$\lambda_L = 532\text{ nm}, \phi = \theta = \pi/2, Z = 1$

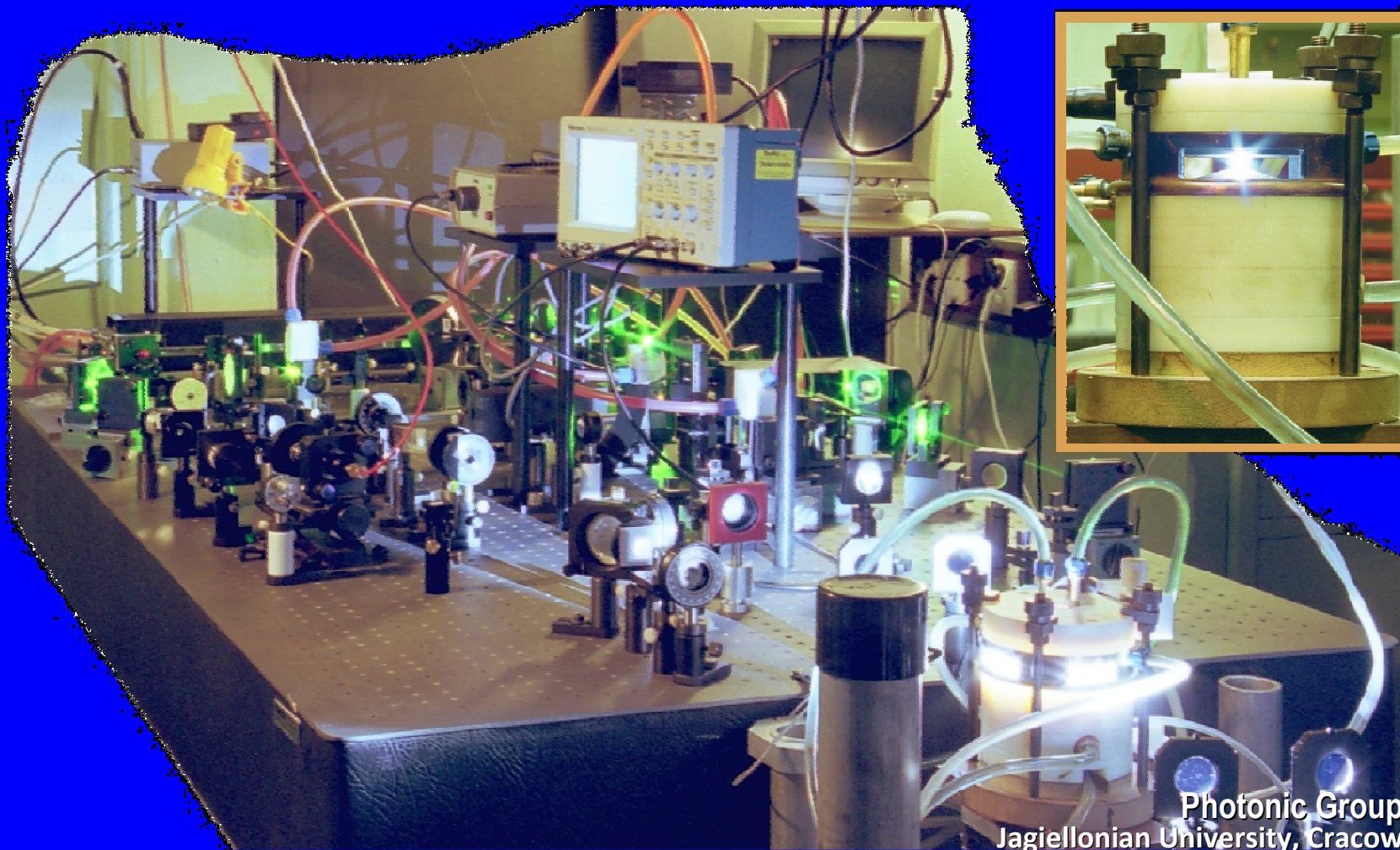
Thomson scattering in different plasmas



Plasma source	N_e [m^{-3}]	T_e [K]	α
<u>Tokamak</u>	10^{20}	10^7	0.006
<u>Glow discharge</u>	10^{18}	10^4	0.02
<u>Argon arc at atmospheric pressure</u>	10^{23}	10^4	3.0
<u>Laser induced plasma</u>	10^{25}	10^5	6.0

- Always collective scattering dominates !
- N_e, T_e can be determined based on the single Thomson scattered spectrum

Thomson scattering in thermal arc plasma



Photonic Group
Jagiellonian University, Cracow

Thomson scattering in thermal arc plasma

Plasma generated in a transferred arc,
burning in pure (99.995%) argon
at atmospheric pressure
with 40 - 160A arc current

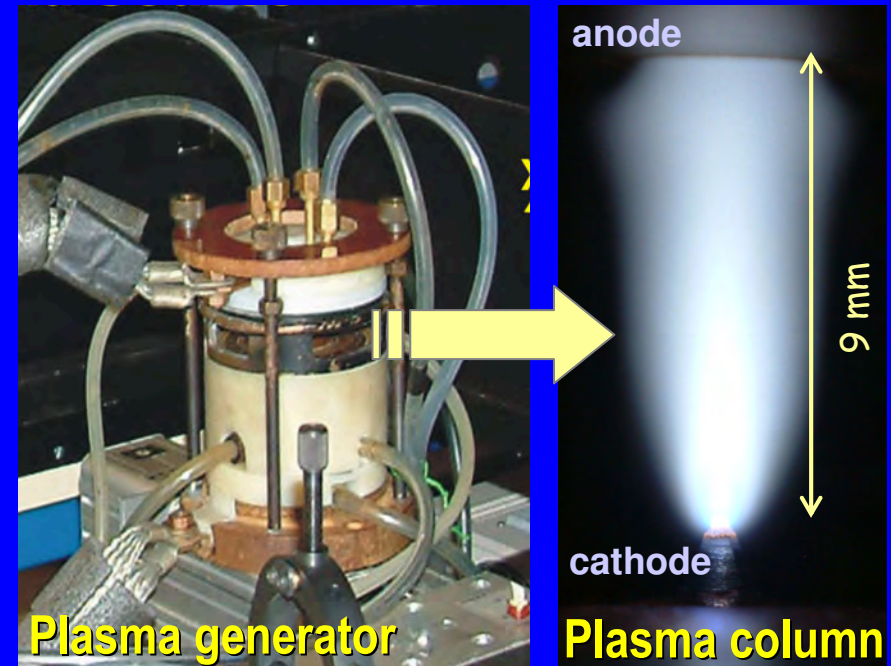
- Electron temperature $T_e : 10000 - 24000 \text{ K}$
- Electron concentration $N_e : 10^{22} - 10^{23} \text{ m}^{-3}$
- Stark broadening dominates
- Long term stability

➤ Large gradients of plasma parameters

➤ Strong continuous emission

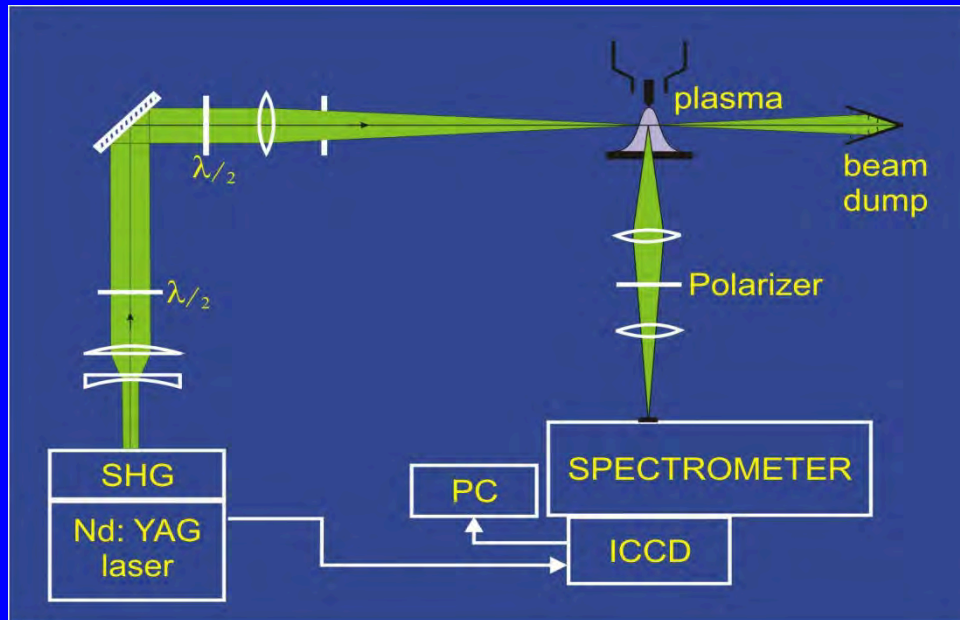
$$\propto N_e^2 / \sqrt{T_e}$$

➤ Domination of non-radiative processes



Thomson scattering in thermal arc plasma

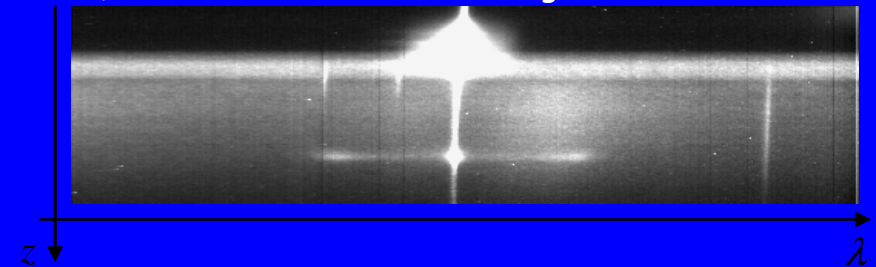
Experimental set-up



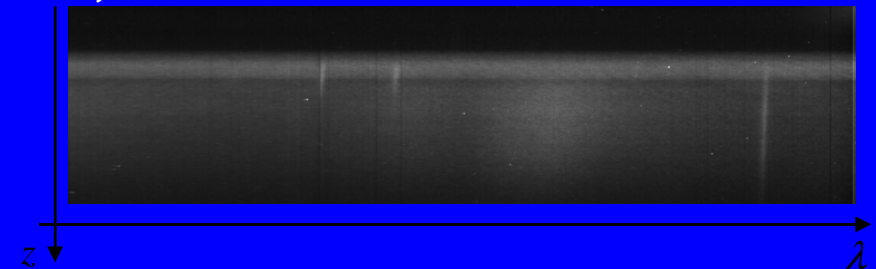
Arc plasma, pure argon at $p=10^5\text{Pa}$, $I=200\text{A}$

Measurement procedure

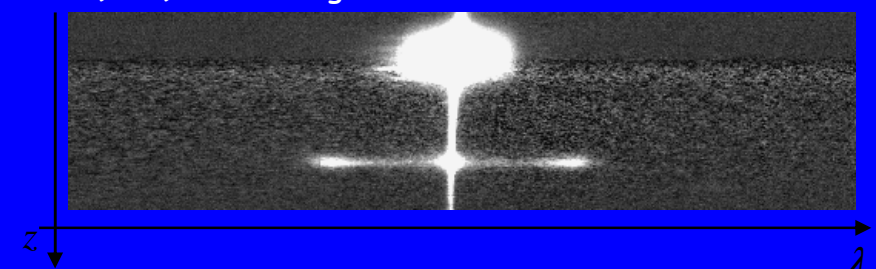
A) Plasma radiation + Scattering



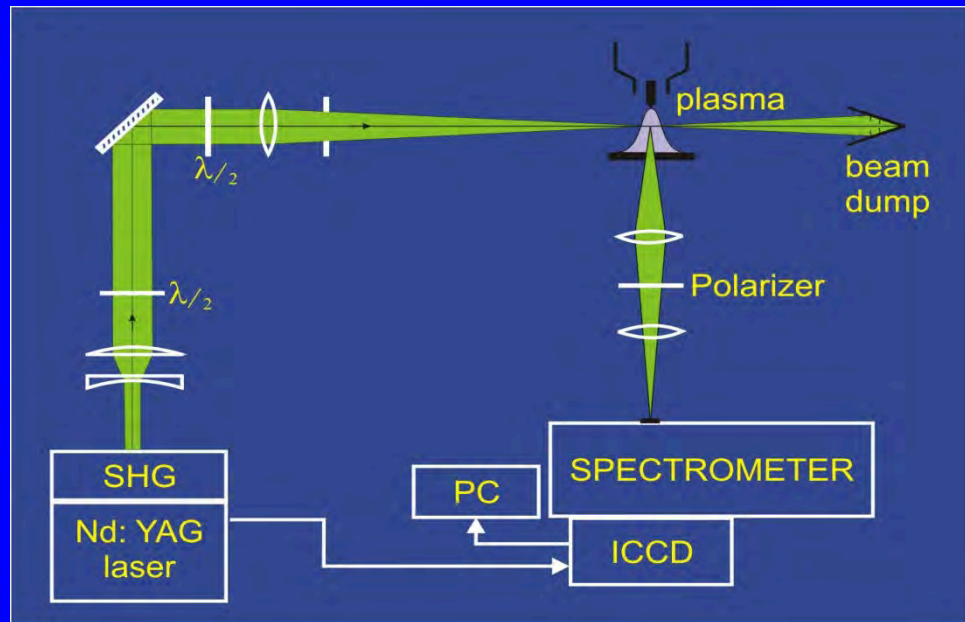
B) Plasma radiation



A) - B) Scattering



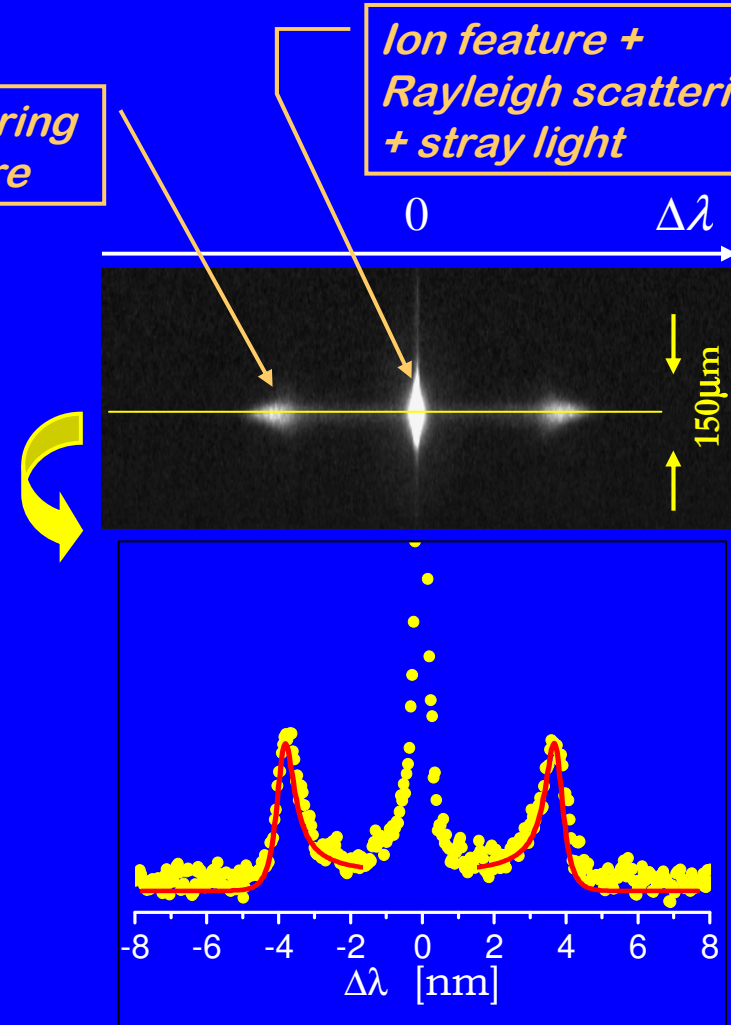
Thomson scattering in thermal arc plasma



Arc plasma, pure argon at $p=10^5\text{Pa}$, $I=200\text{A}$

Thomson scattering
- electron feature

Ion feature +
Rayleigh scattering
+ stray light



$S(\mathbf{k}, \Delta\omega)$

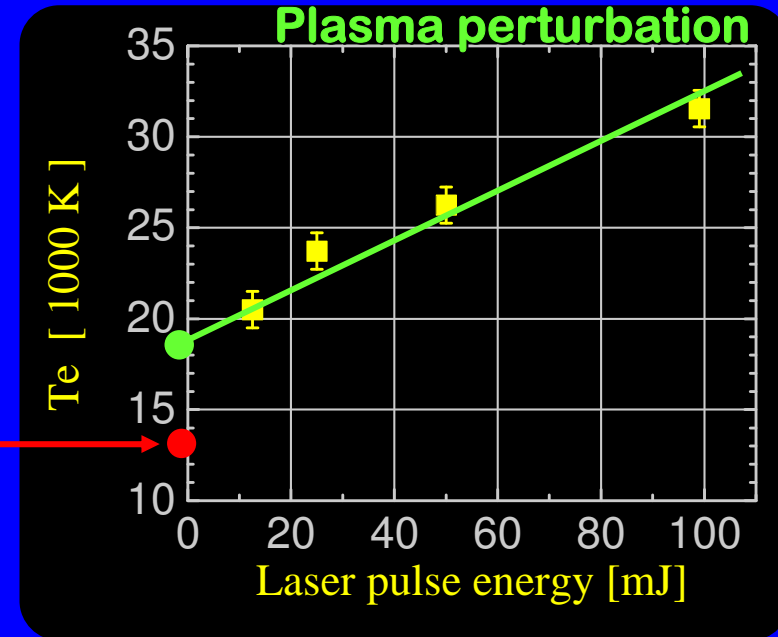
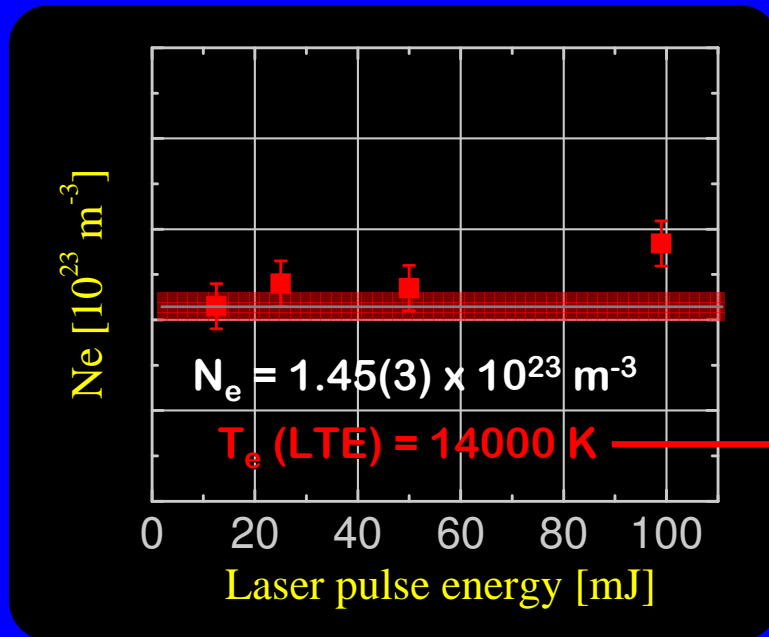


$N_e = 1.1 \times 10^{23} \text{ m}^{-3}$

$T_e = 21000 \text{ K}$

Thomson scattering in thermal arc plasma

N_e and T_e versus energy of the laser pulse



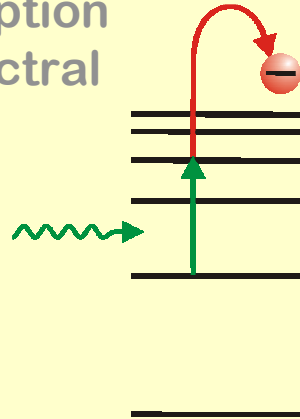
- ▶ T_e is higher than T_{LTE} obtained from N_e , by few thousands kelvins, which contradicts the LTE plasma model
- ▶ Either most of theoretical and experimental studies are false ..
OR interpretation of TS results is incorrect ???

Thomson scattering in thermal arc plasma

↳ Plasma disturbance by laser pulse

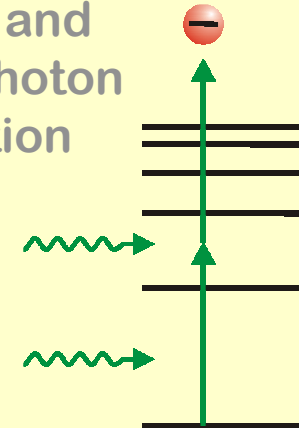
Possible absorption processes

Absorption at spectral lines



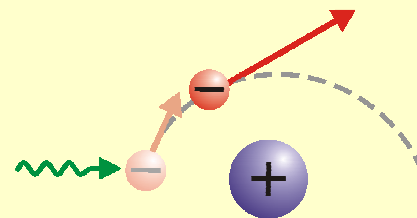
Increase of n_e

Single and multiphoton ionization



*Electron heating,
Increase of T_e*

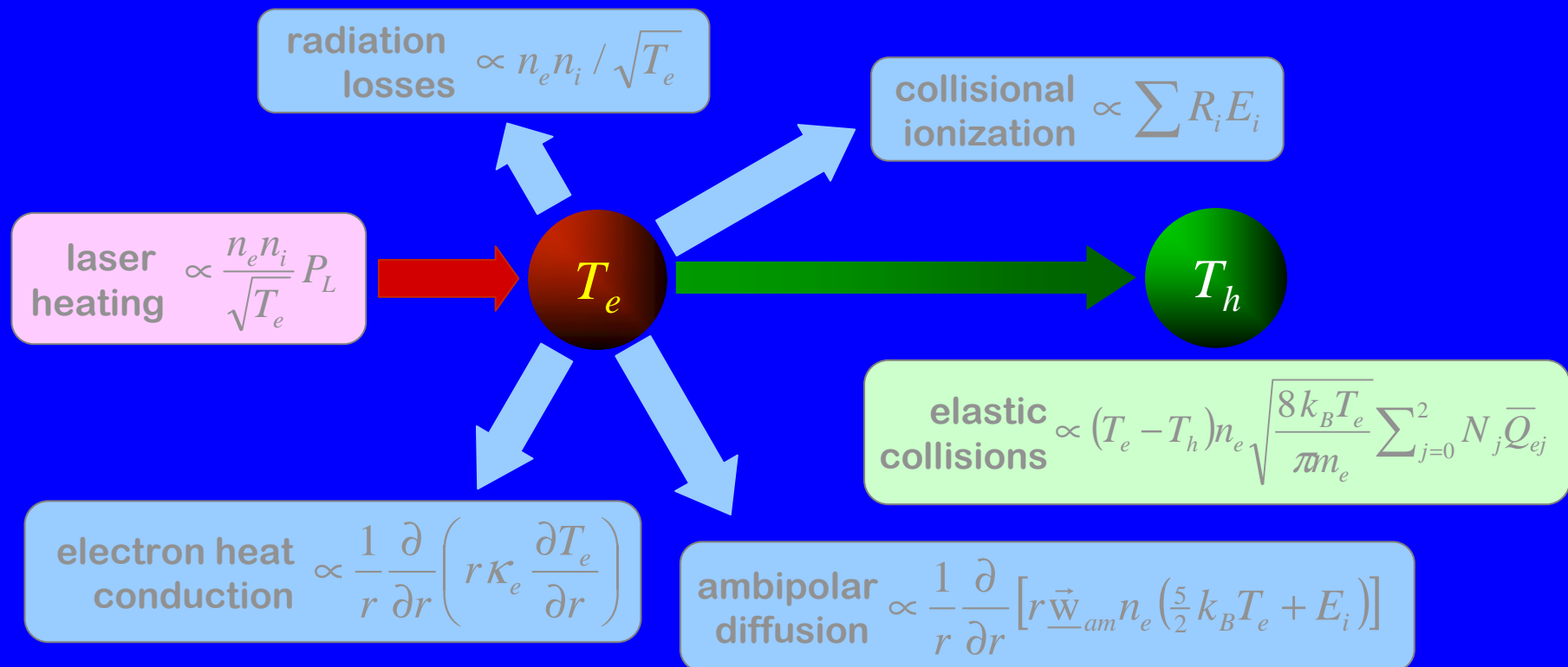
Inverse bremsstrahlung



Thomson scattering in thermal arc plasma

↳ Electron heating by laser pulse

► Plasma perturbed (heated) in the inverse bremsstrahlung process !!



A.B. Murphy; PRL 89, 025002 (2002); Phys Rev E 69, 016408 (2004)

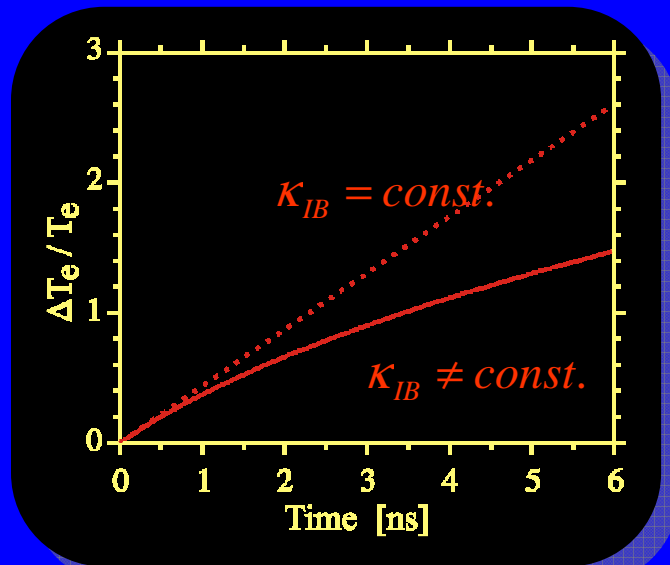
Thomson scattering in thermal arc plasma

↳ Electron heating by laser pulse

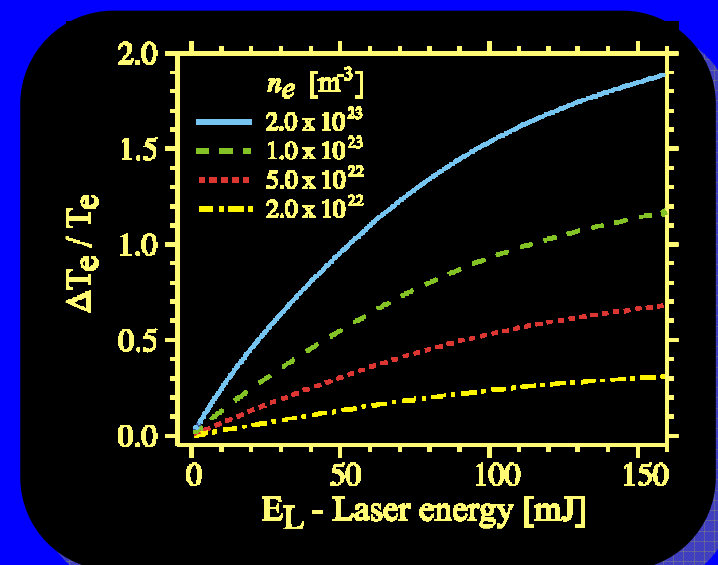
► Plasma perturbed (heated) in the inverse bremsstrahlung process !!

$$\frac{\Delta T_e}{T_e} = \frac{2}{3} \frac{\kappa_{IB} E_L}{k_B T_e n_e \pi r_0^2}$$

$$\frac{\Delta T_e}{T_e} = 6.6 \times 10^{-5} \frac{\sum_Z Z^2 n_{i,Z}}{T_e^{3/2}} \frac{E_L}{\pi r_0^2} \times \bar{g}_{ff}(\lambda_L) \lambda_L^3 (1 - \exp(-hc/k_B T_e \lambda_L))$$



$E_L = 100 \text{ mJ}$, $T_e = 17400 \text{ K}$, $n_e = 2 \times 10^{23} \text{ m}^{-3}$



Initial temperature : $T_e = 11600 \text{ K}$

A.B. Murphy; PRL 89, 025002 (2002); Phys Rev E 69, 016408 (2004)

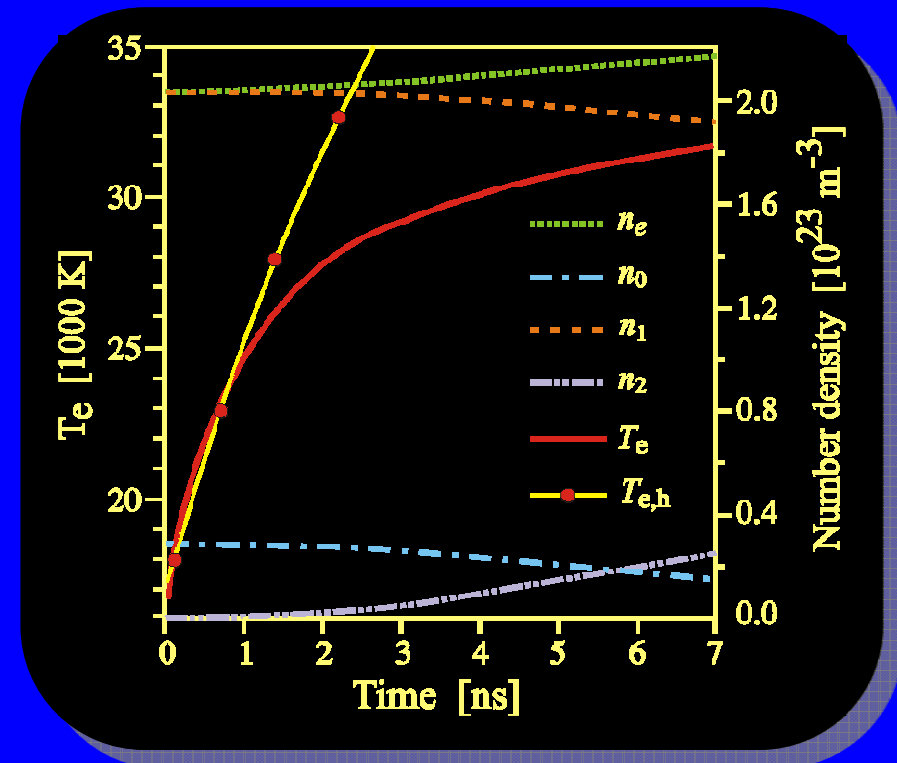
Thomson scattering in thermal arc plasma

↳ Electron heating by laser pulse

MHD model, argon thermal plasma heated by the square, 7ns long laser pulse

➔ *Electron heating is a strongly nonlinear function of the laser power*

so the linear extrapolation to zero pulse energy is invalid !



$T_{e,h}$ – assuming no cooling channels

A.B. Murphy; PRL 89, 025002 (2002); Phys Rev E 69, 016408 (2004)

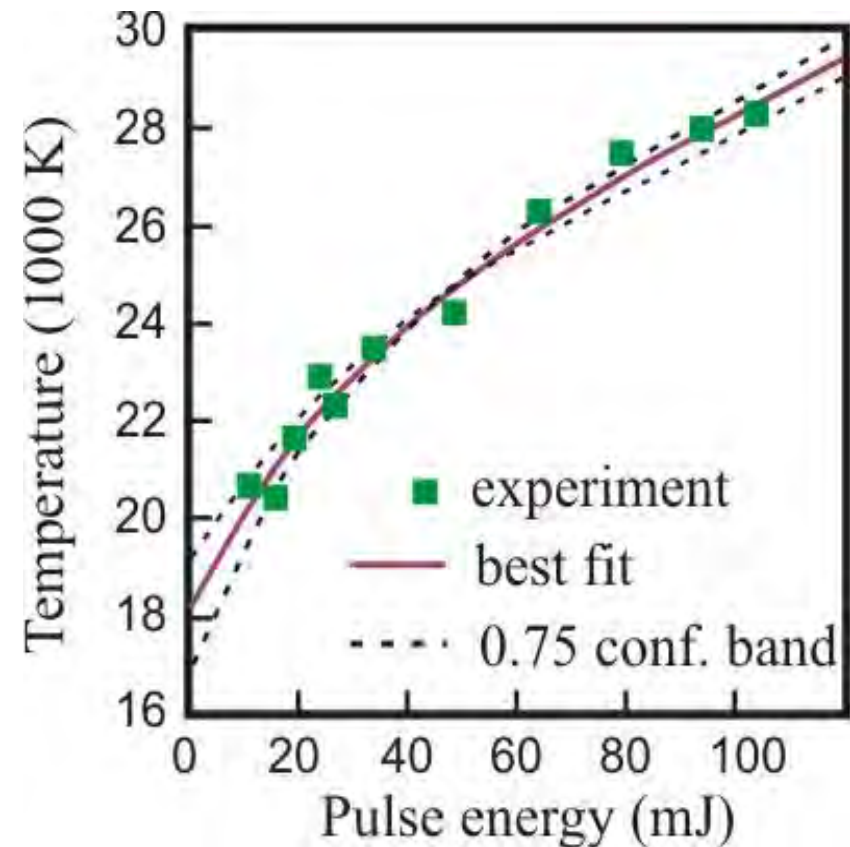
Thomson scattering in thermal arc plasma

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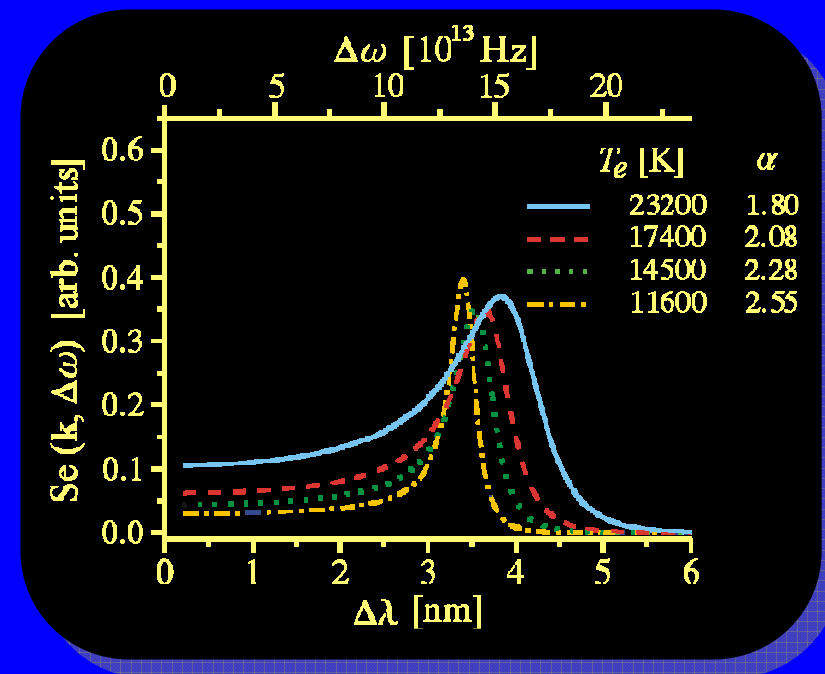
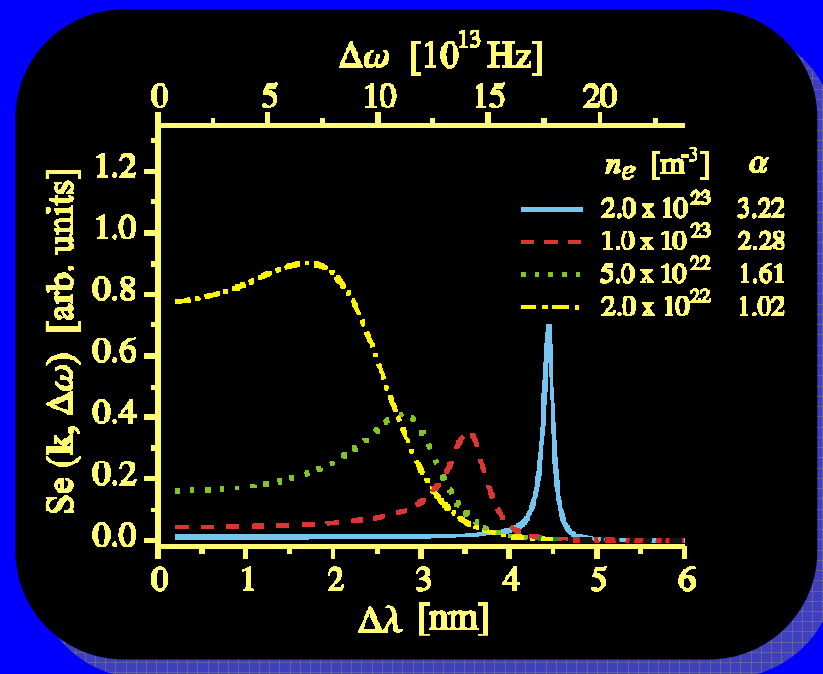


A.B. Murphy; PRL 89, 025002 (2002); Phys Rev E 69, 016408 (2004)

Thomson scattering in thermal arc plasma

↪ Averaging over laser beam cross section and over duration of laser pulse

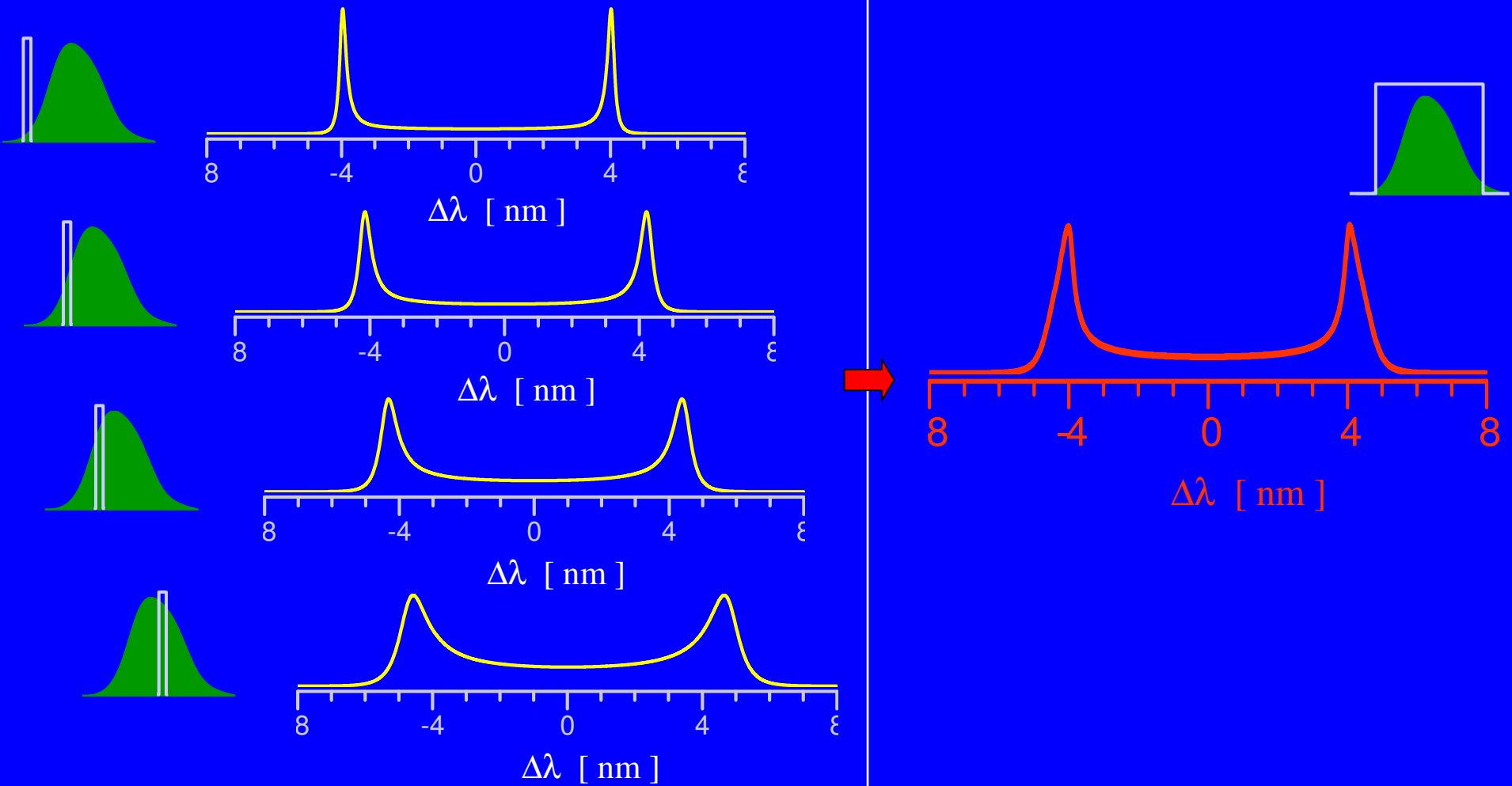
TS spectra for different n_e and T_e values



Thomson scattering in thermal arc plasma

↪ Averaging over laser beam cross section and over duration of laser pulse

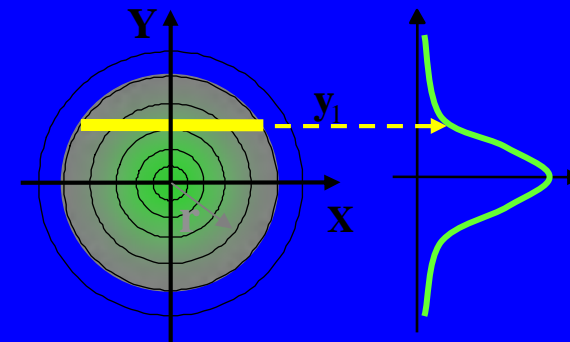
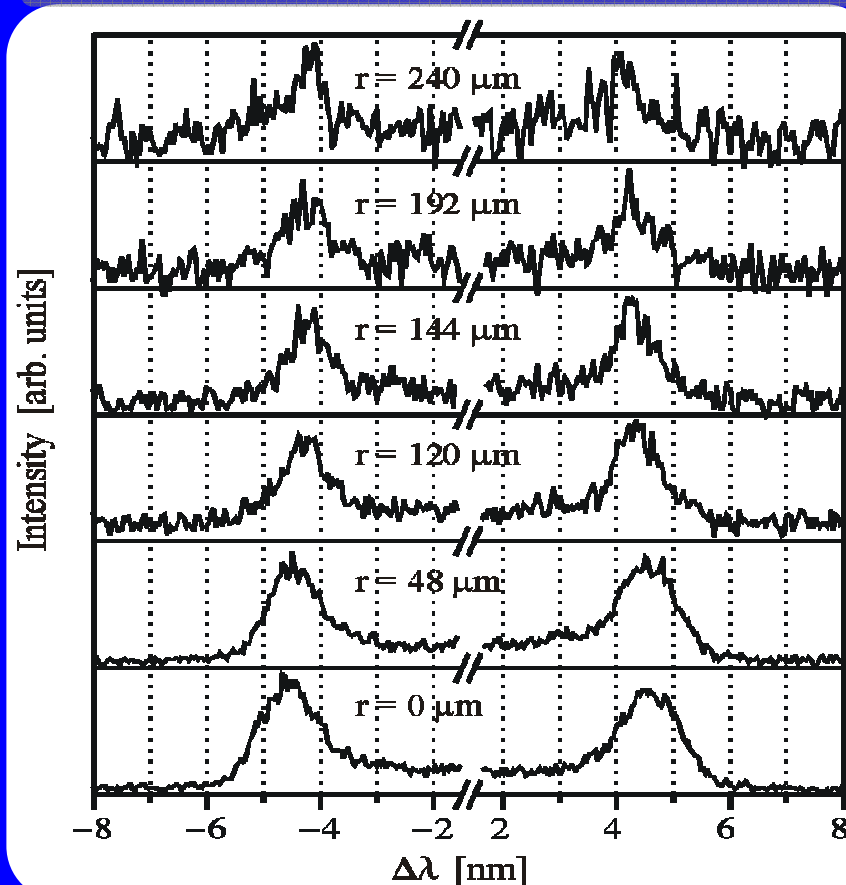
Effect of electron heating by laser pulse



Thomson scattering in thermal arc plasma

↪ Averaging over laser beam cross section
(Experimental results: spatially resolved results)

TS spectra (electron terms) across the laser beam



$$I(\lambda, y_1) = \int I(\lambda, y_1, x) dx$$

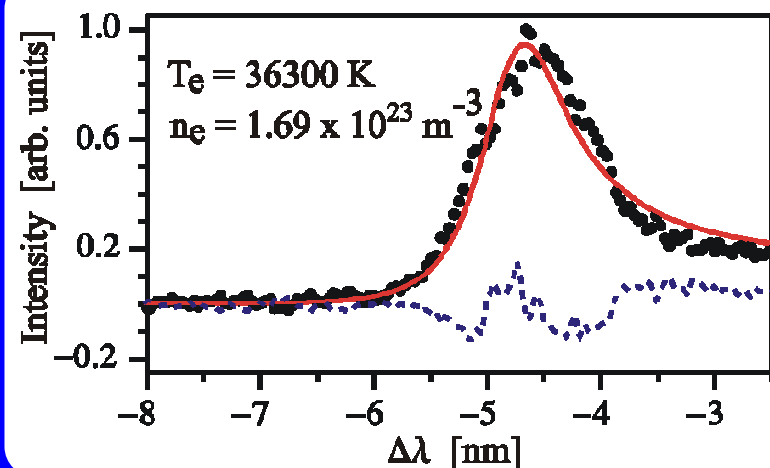
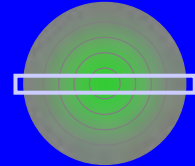
Abel transformation

$$I(\lambda, r) = \int_r^R \frac{dI(\lambda, y) / dy}{\sqrt{y^2 - r^2}} dy$$

Thomson scattering in thermal arc plasma

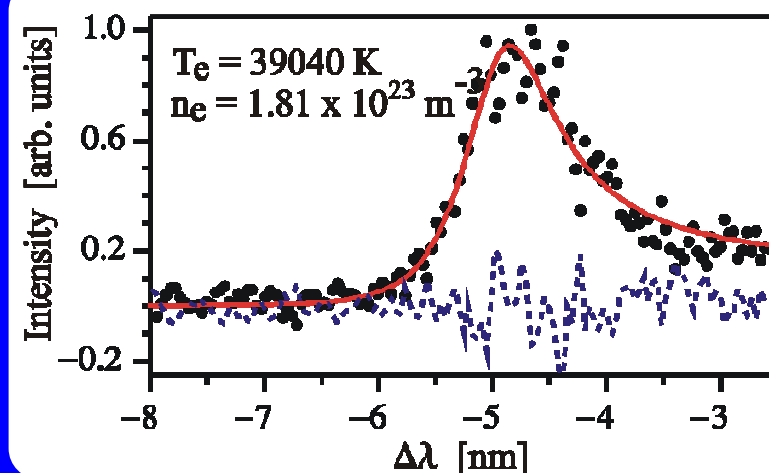
↪ Averaging over laser beam cross section
(Experimental results: spatially resolved results)

Before Abel transformation
TS @ $y=0$



systematic errors dominate

After Abel transformation
TS @ $r=0$

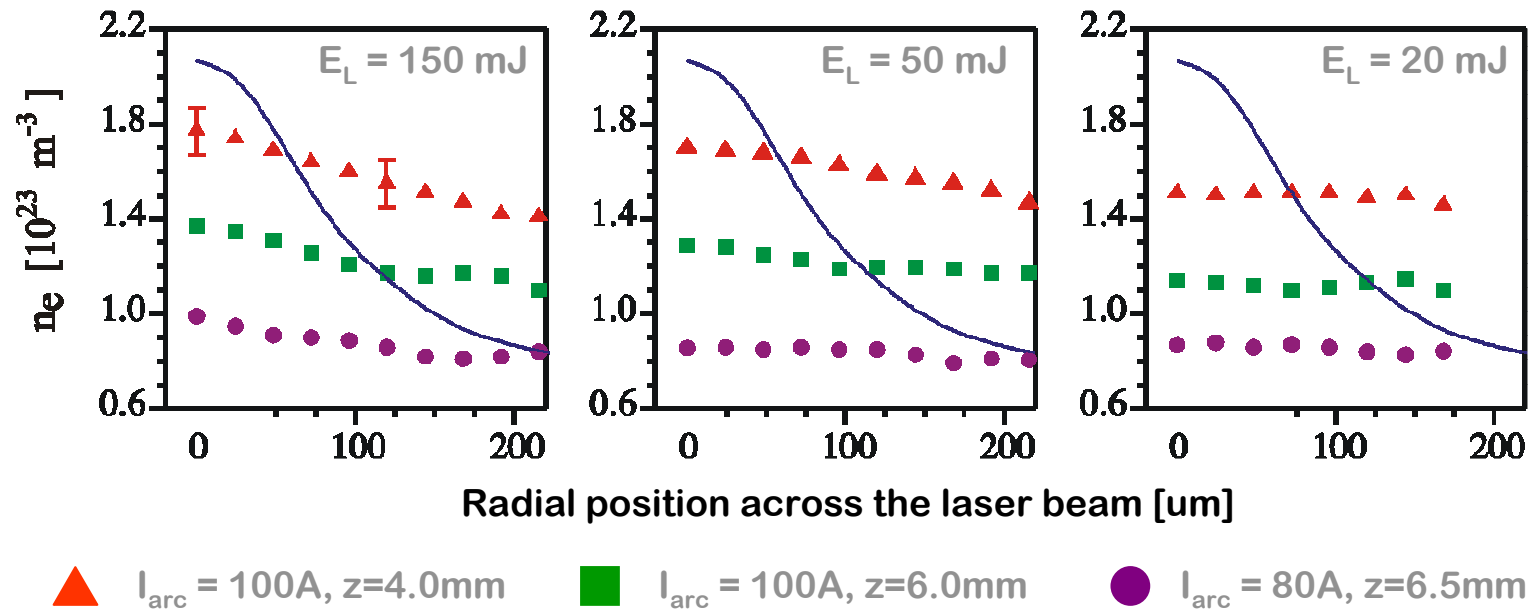
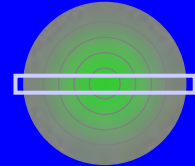


statistical errors dominate

Thomson scattering in thermal arc plasma

↪ Averaging over laser beam cross section
(Experimental results: spatially resolved results)

Local values (across the laser beam) of n_e

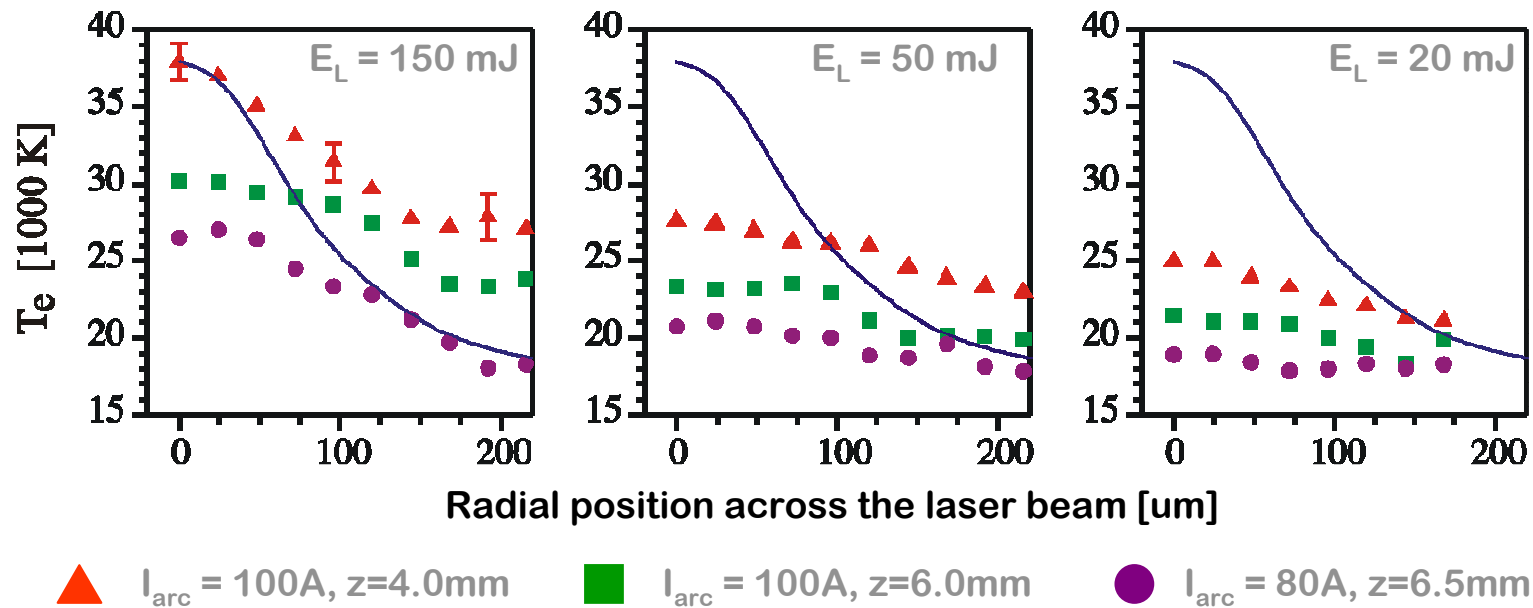
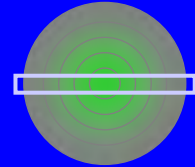


➔ *No variations of n_e at lower laser energies
and at the edges of the laser beam*

Thomson scattering in thermal arc plasma

↪ Averaging over laser beam cross section
(Experimental results: spatially resolved results)

Local values (across the laser beam) of T_e

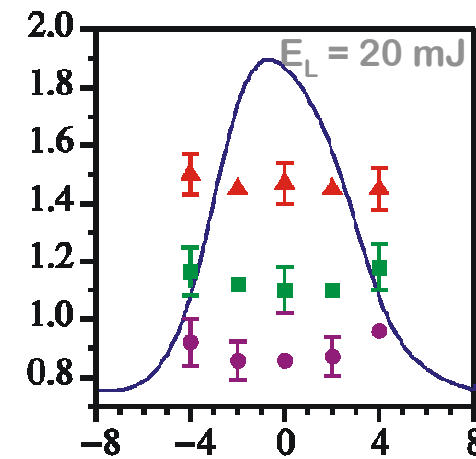
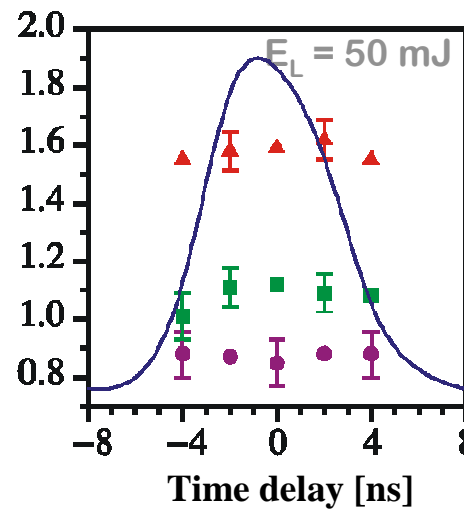
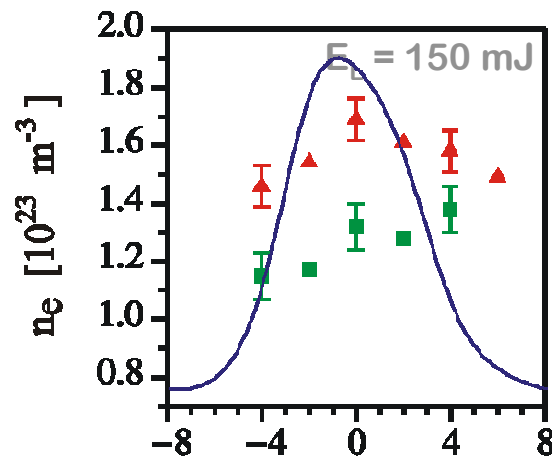
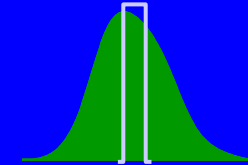


➔ *Increase of T_e depends on laser power and initial plasma conditions, i.e. the electron number density and temperature*

Thomson scattering in thermal arc plasma

↪ Averaging over laser beam cross section
(Experimental results: temporally resolved results)

Temporal evolution of n_e during the laser pulse



▲ $I_{\text{arc}} = 100\text{A}, z=4.0\text{mm}$

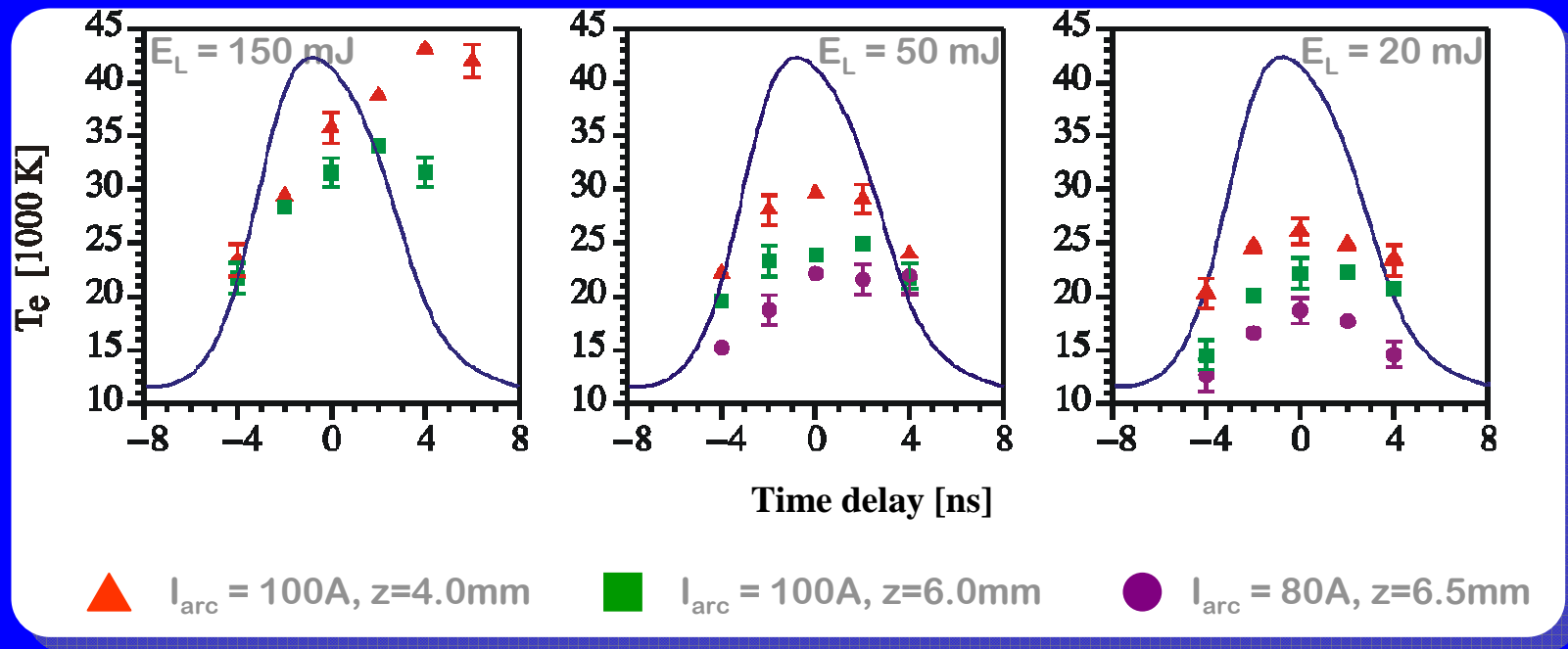
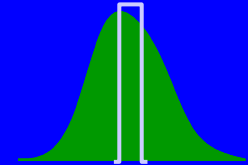
■ $I_{\text{arc}} = 100\text{A}, z=6.0\text{mm}$

● $I_{\text{arc}} = 80\text{A}, z=6.5\text{mm}$

Thomson scattering in thermal arc plasma

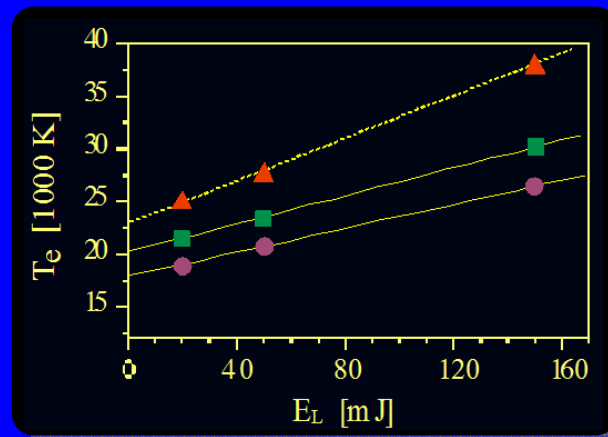
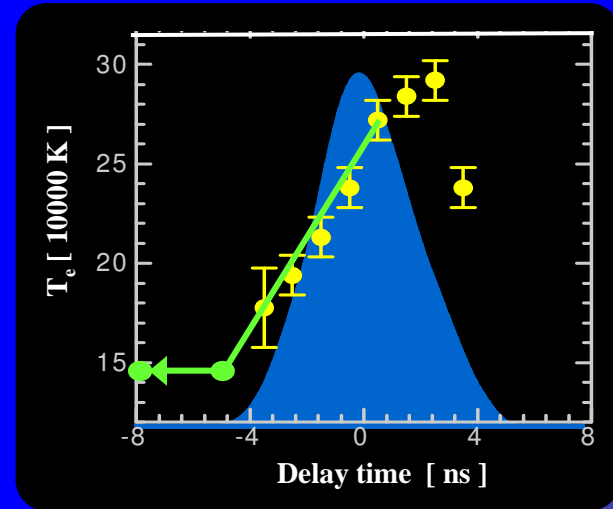
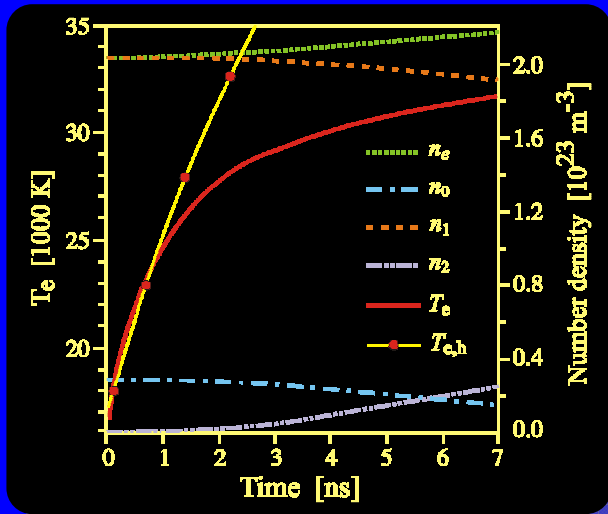
↪ Averaging over laser beam cross section
(Experimental results: temporally resolved results)

Temporal evolution of T_e during the laser pulse



Thomson scattering in thermal arc plasma

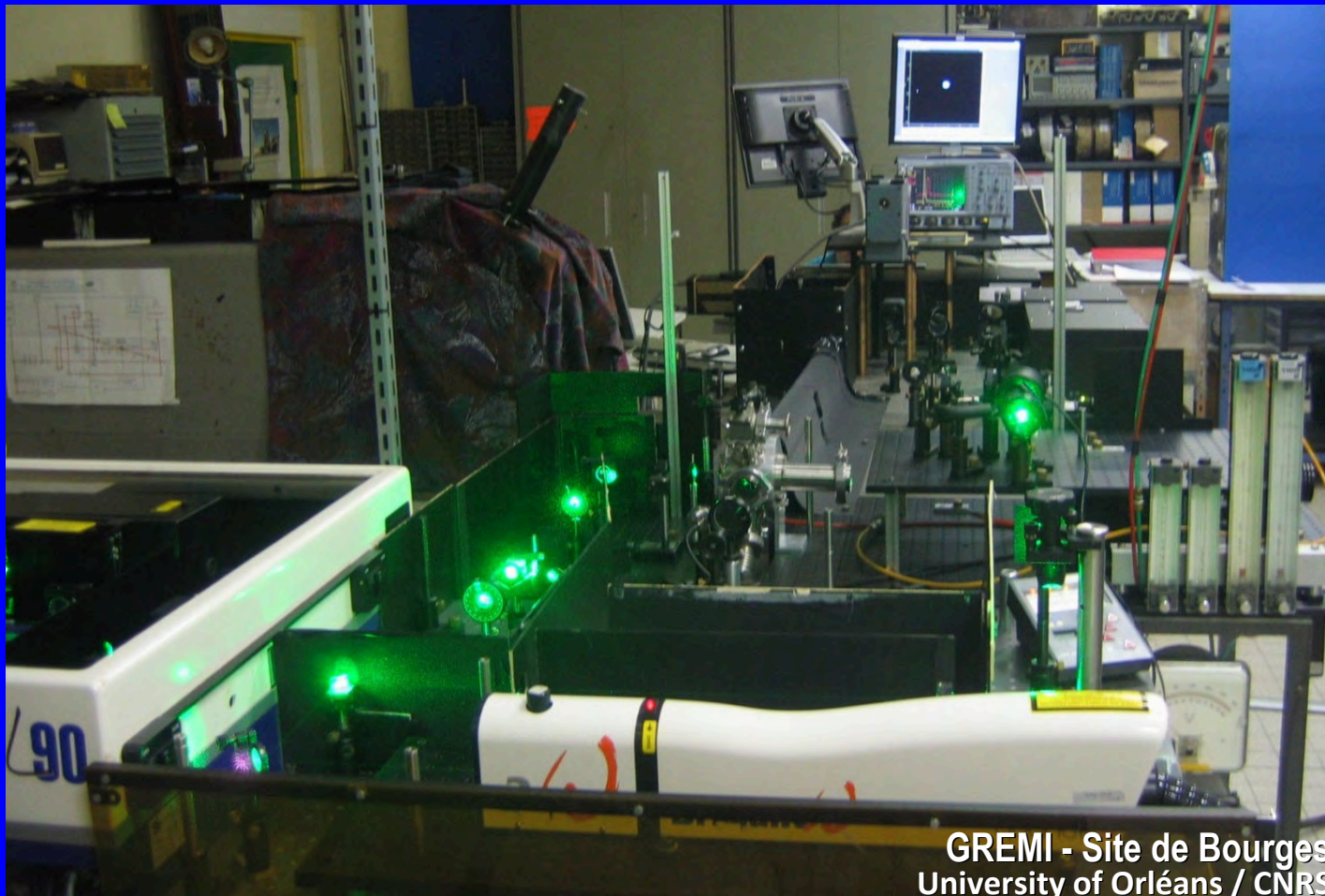
Determination of initial T_e



	N_e [10^{23} m^{-3}]	T_e [LTE]	T_e [lin.extr.]	T_e [origin]
▲	1.46	13900	22780	14000
■	1.14	13050	20090	12200
●	0.89	12500	17760	10000

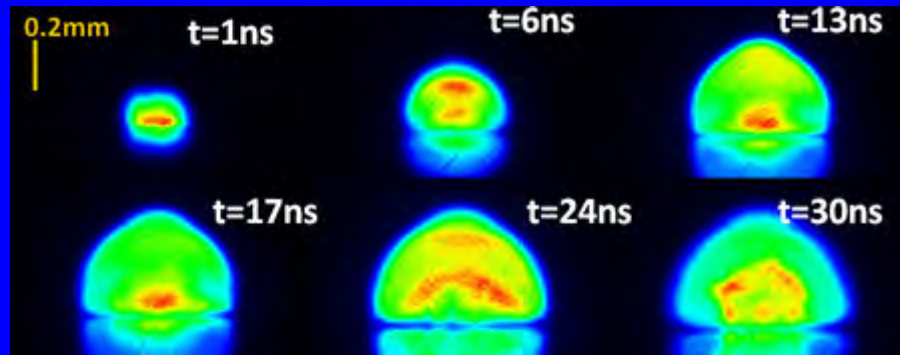
➔ Measurement of the temporal evolution of T_e and then **extrapolation to the origin of the pulse** is more adequate method than extrapolation to the zero laser energy

Thomson scattering in laser induced plasma



GREMI - Site de Bourges
University of Orléans / CNRS

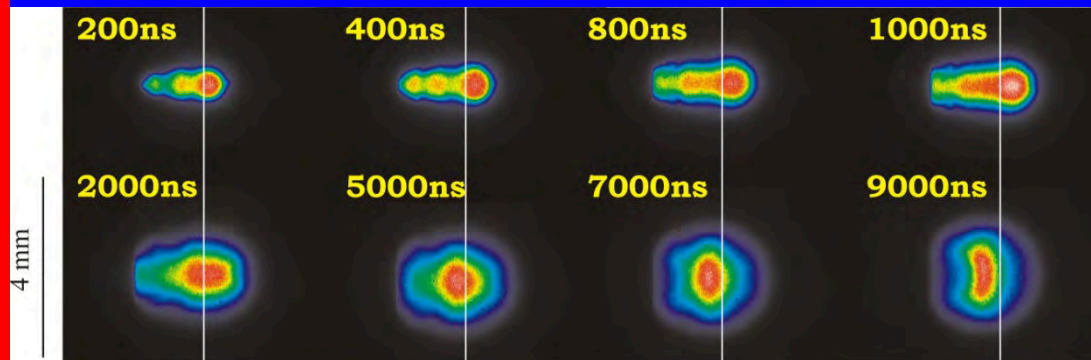
Laser induced plasmas: two main types



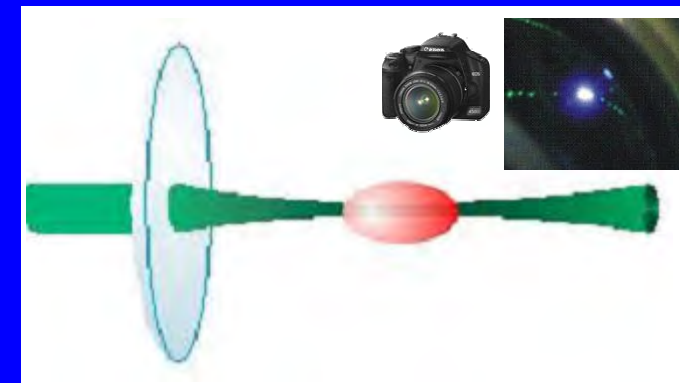
A. Mendys et al. / Spectrochimica Acta Part B 96 (2014) 61–68



Solid target (ablation)
Application: film deposition, analysis...



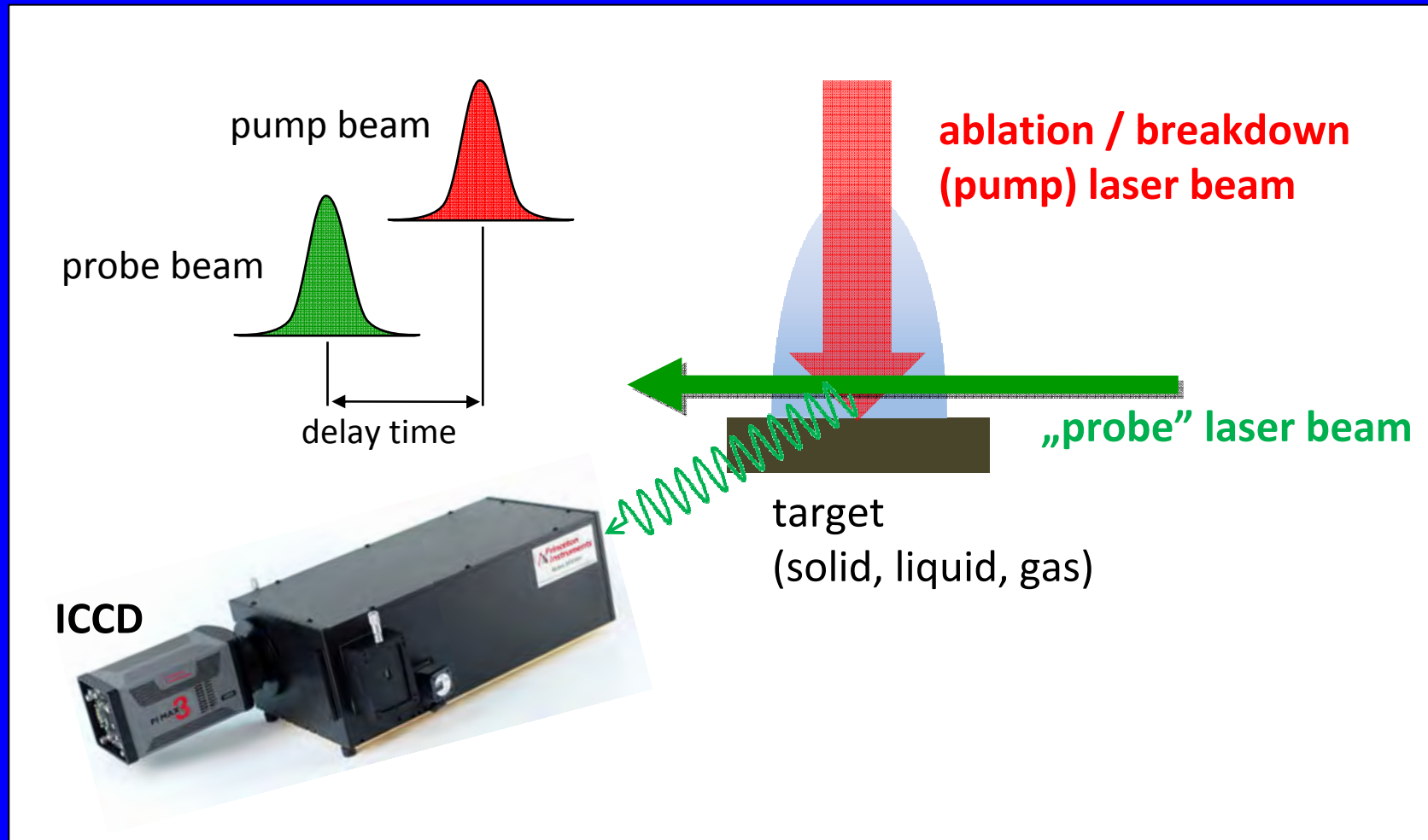
A. Mendys, et al., Spectrochim. Acta B (2011) 66, 691



Gaseous target (breakdown)
Application: plasma igniters...

Laser induced plasmas

↳ „Pump – probe” experiments



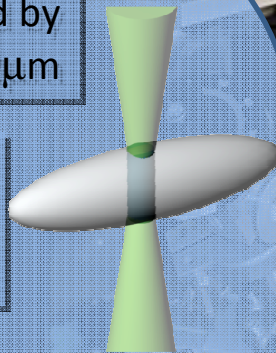
Laser induced sparks in gases

↪ **Experimental setup** to study laser induced spark created and probed by ns Nd:YAG lasers

spatial resolution

radially limited by laser beam: $\approx 150\mu\text{m}$

axially determ. by slit width $\approx 10\text{-}20\mu\text{m}$



4ns Nd:YAG @ 532nm, 40 J/cm^2
single-mode $\Delta\sigma < 0.01\text{ cm}^{-1}$

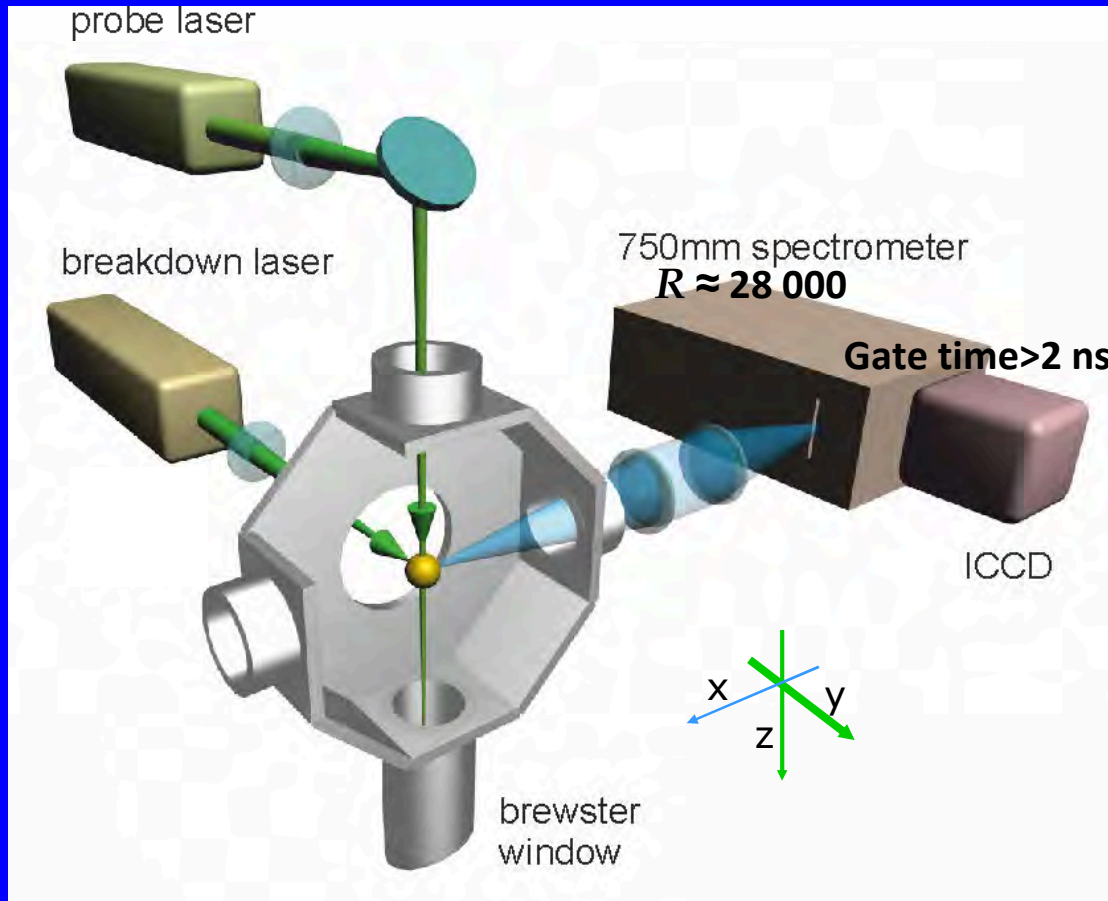
6ns Nd:YAG @ 532nm,
 2.0 kJ/cm^2 , $\Delta\sigma < 1.0\text{ cm}^{-1}$

to spectrometer + ICCD
 $R \approx 28\ 000$

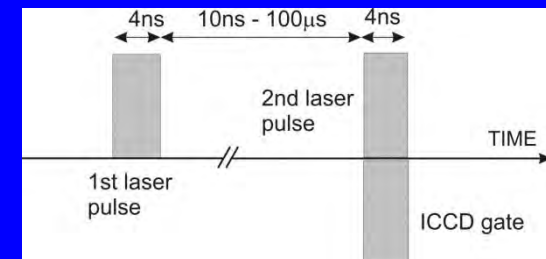
Laser induced sparks in gases

➤ **Experimental setup** to study laser induced spark created and probed by ns Nd:YAG lasers

Breakdown laser Nd:YAG @ 532nm, 2.0 kJ/cm^2 , $\Delta\tau = 6\text{ns}$, single-mode $\Delta\sigma < 1.0 \text{ cm}^{-1}$
 Probe laser Nd:YAG @ 532nm, up to 40 J/cm^2 , $\Delta\tau = 4\text{ns}$, single-mode $\Delta\sigma < 0.01 \text{ cm}^{-1}$



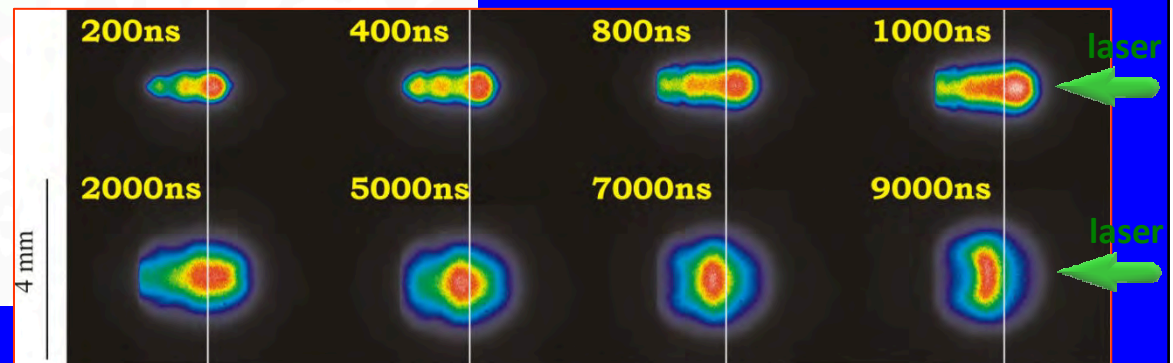
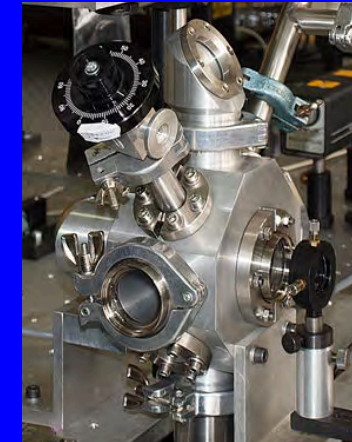
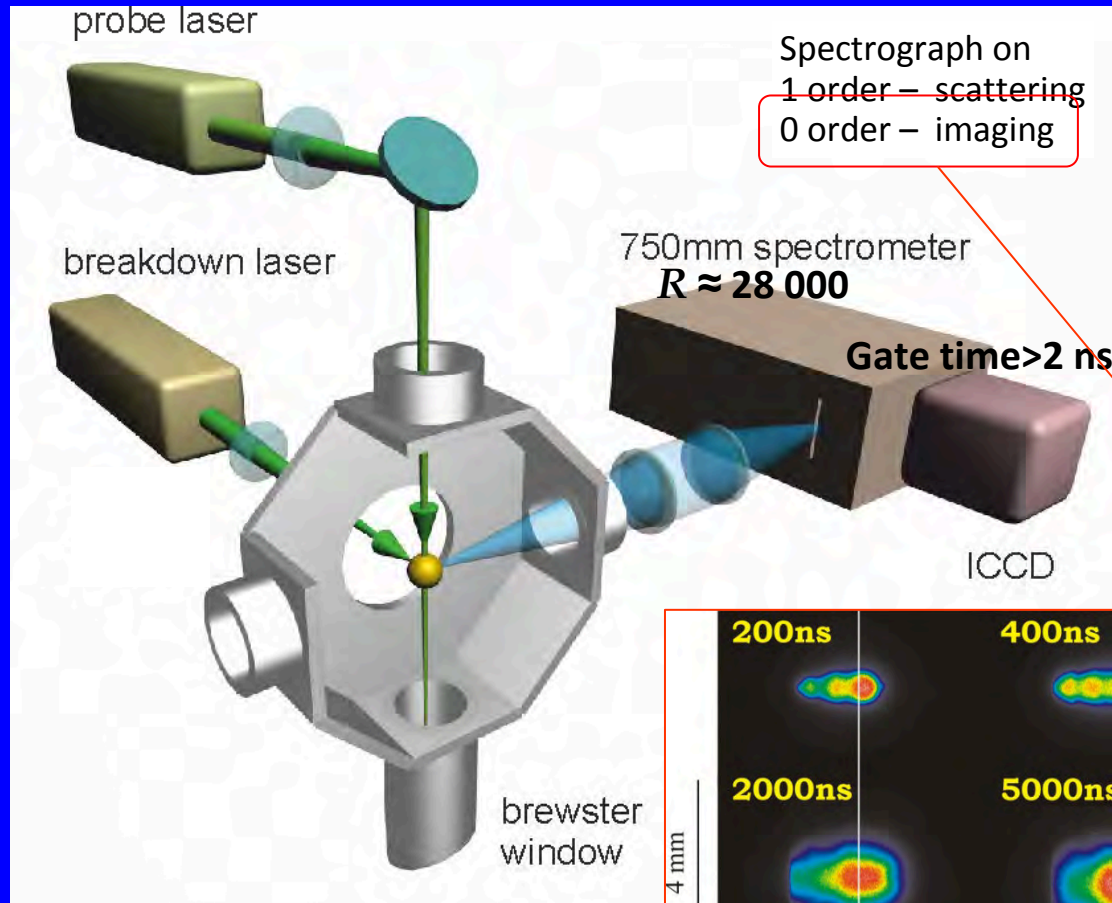
ICCD:
 delay → $5\text{ns} - 100\mu\text{s}$
 gate → $2\text{ns} - 500\text{ns}$



Laser induced sparks in gases

➤ **Experimental setup** to study laser induced spark created and probed by ns Nd:YAG lasers

Breakdown laser Nd:YAG @ 532nm, 2.0 kJ/cm^2 , $\Delta\tau = 6\text{ns}$, single-mode $\Delta\sigma < 1.0 \text{ cm}^{-1}$
Probe laser Nd:YAG @ 532nm, up to 40 J/cm^2 , $\Delta\tau = 4\text{ns}$, single-mode $\Delta\sigma < 0.01 \text{ cm}^{-1}$



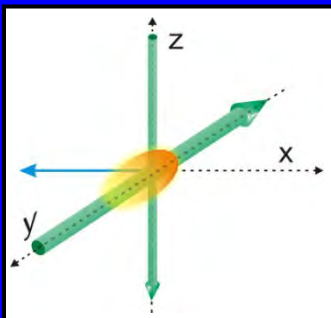
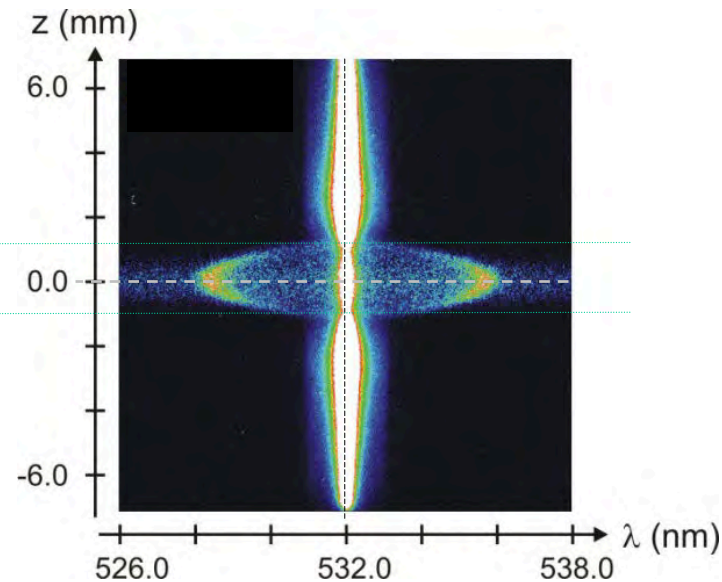
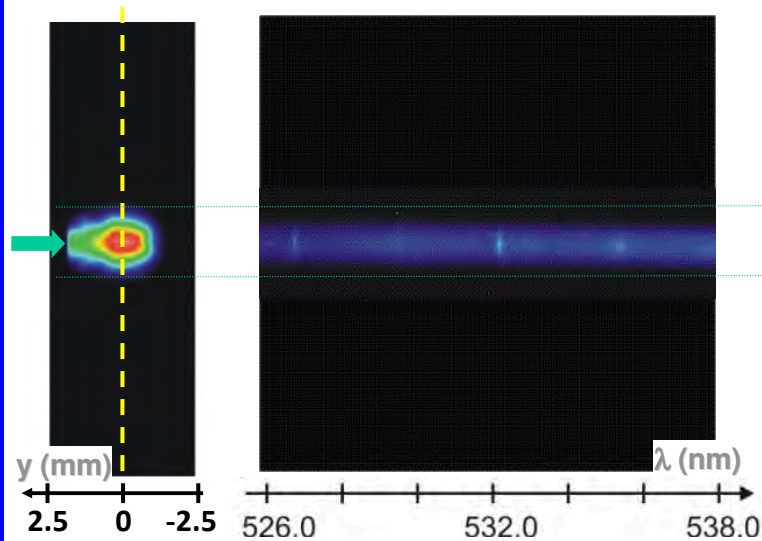
Laser scattering from laser spark in Ar at P_{atm}

plasma image

plasma emission

scattered probe laser light

$\Delta\tau = 3000\text{ns}$, $x=0$; $y=0$, exposure time = 4ns



Pump fluence – $2\text{kJ}/\text{cm}^2$

Probe fluence – $18\text{J}/\text{cm}^2$

$\theta = 90^\circ$

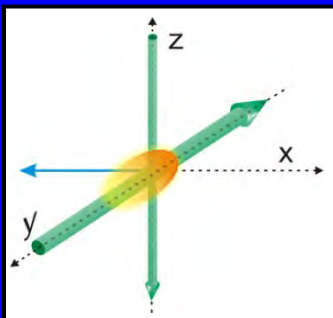
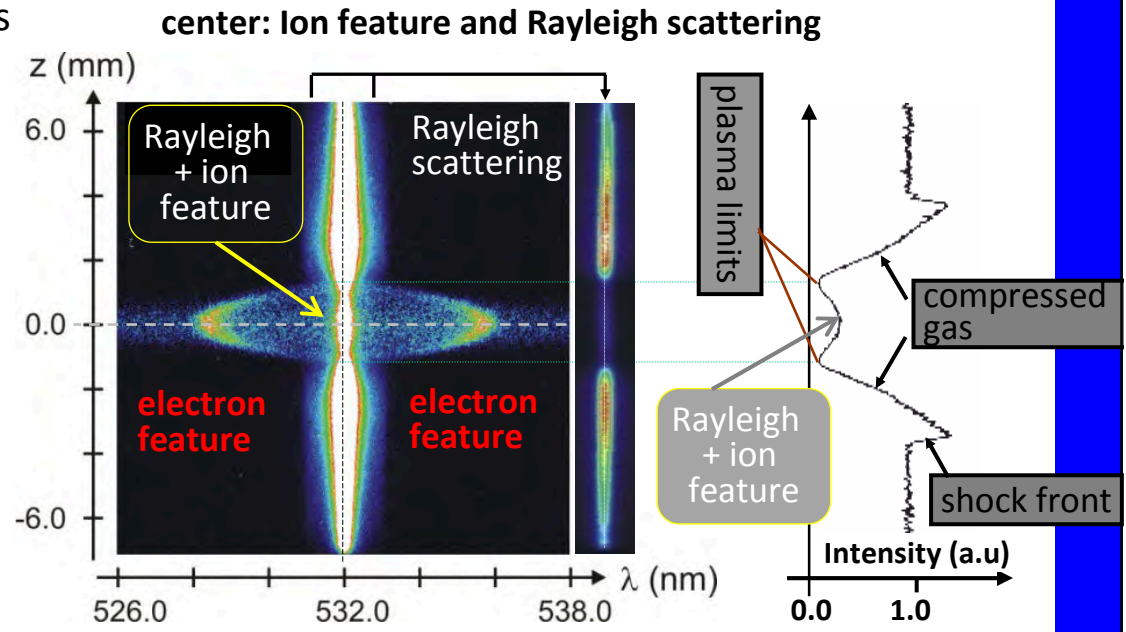
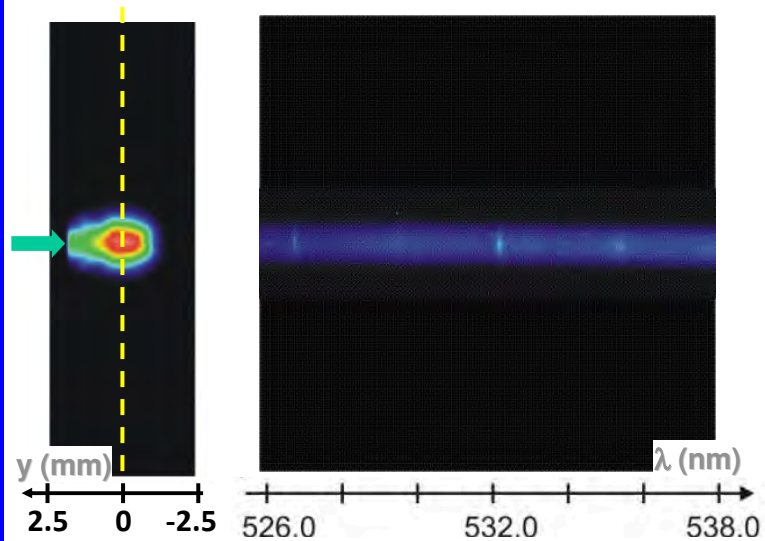
Laser scattering from laser spark in Ar at P_{atm}

plasma image

plasma emission

scattered probe laser light

$\Delta\tau = 3000\text{ns}$, $x=0$; $y=0$, exposure time = 4ns



Pump fluence – 2kJ/cm^2
 Probe fluence – 18J/cm^2
 ICCD gate – 10 ns

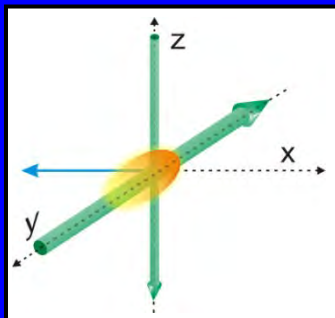
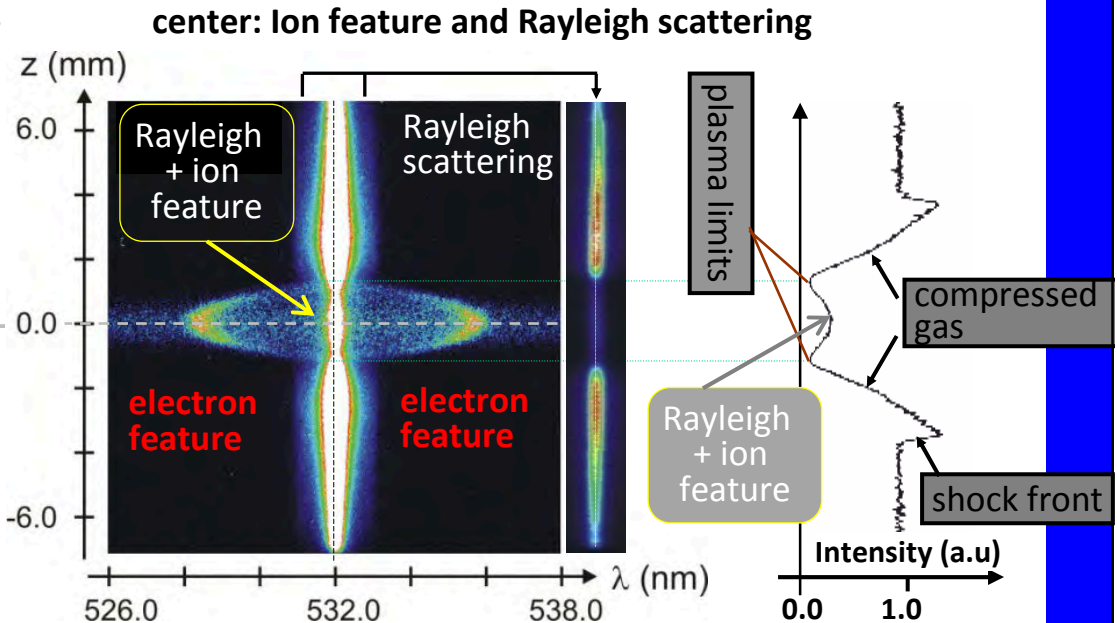
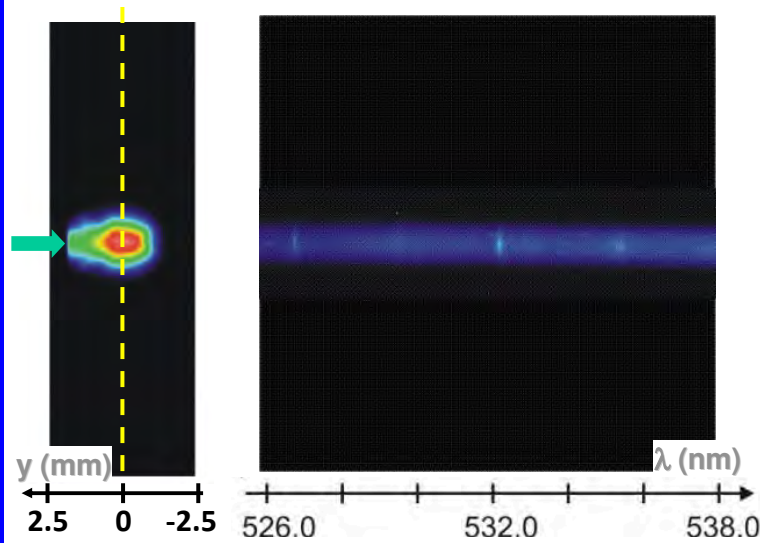
Laser scattering from laser spark in Ar at P_{atm}

plasma image

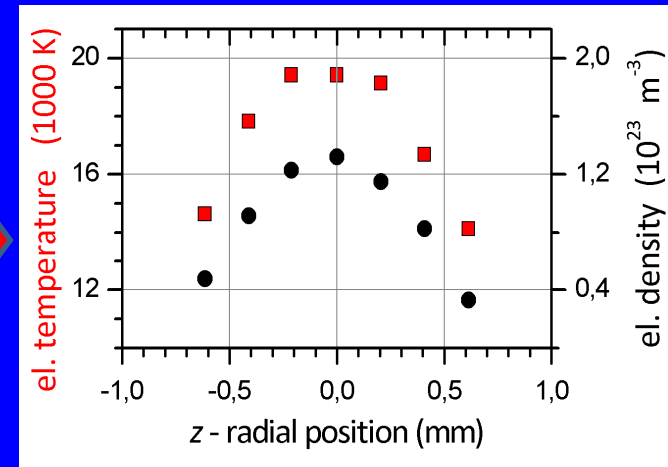
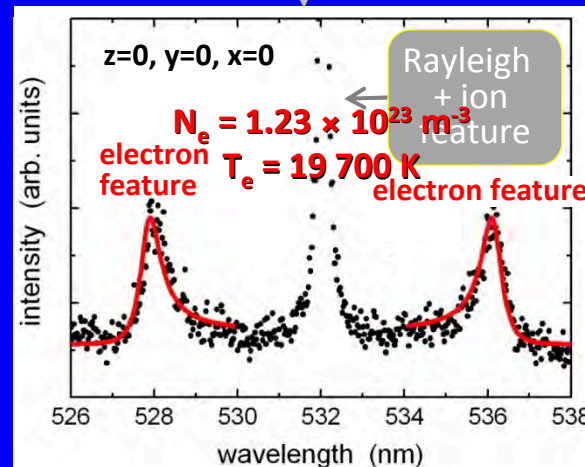
plasma emission

scattered probe laser light

$\Delta\tau = 3000\text{ns}$, $x=0$; $y=0$, exposure time = 4ns

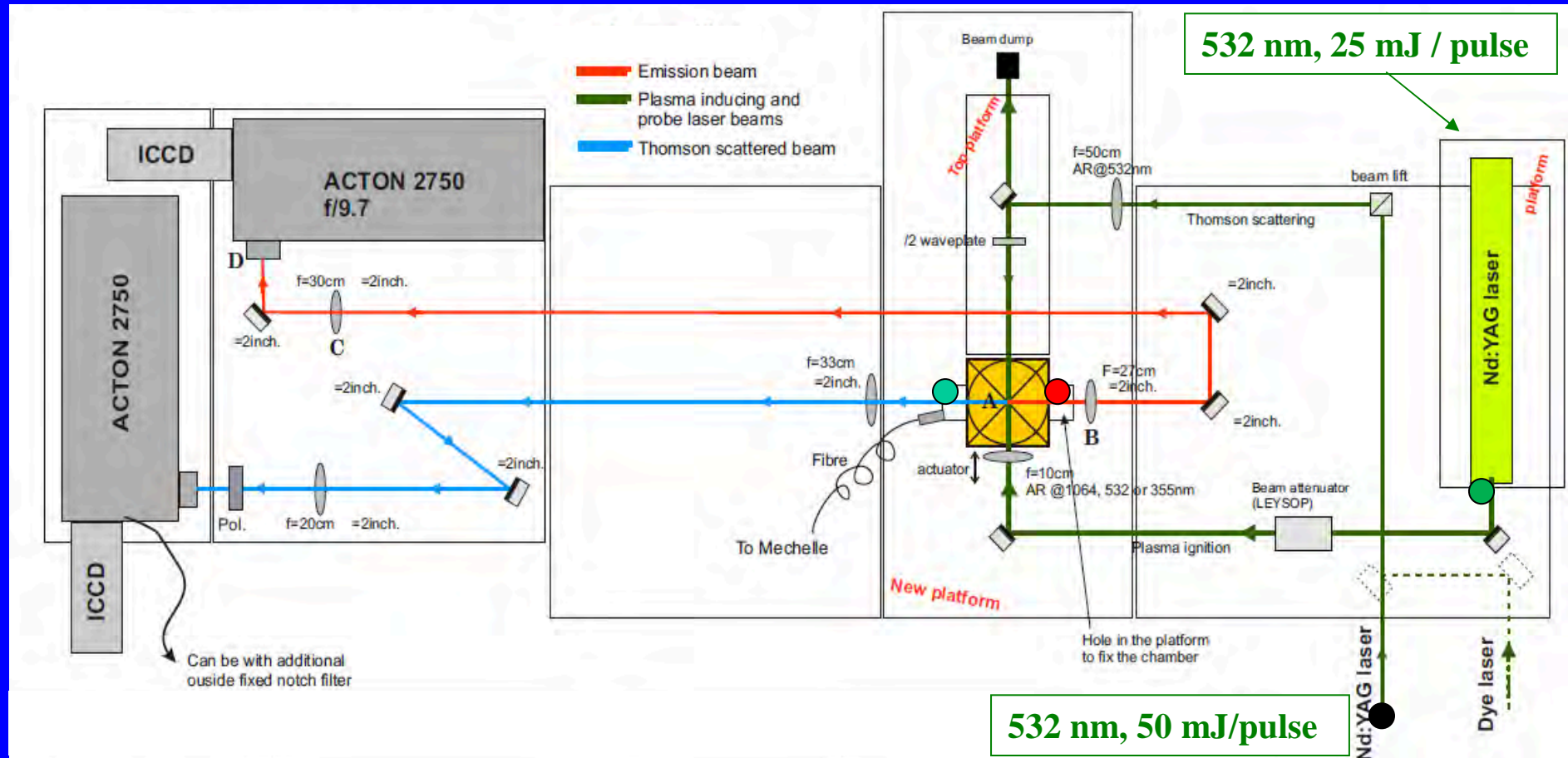


Pump fluence – 2kJ/cm^2
 Probe fluence – 18J/cm^2
 ICCD gate – 10 ns



Laser scattering from laser spark in Ar at P_{atm}

↳ Comparison with OES

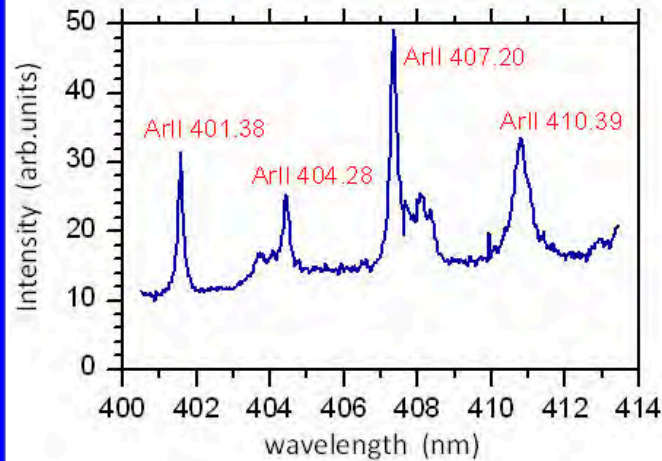
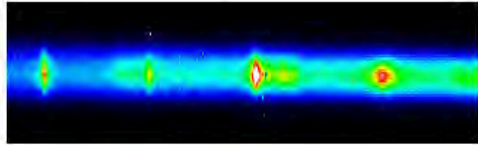


- Imaging of the plume
- Optical Emission Spectroscopy (OES)
- Thomson Scattering (TS)

Laser scattering from laser spark in Ar at P_{atm}

↳ Comparison with OES

emission from LIP in Argon
at 400ns after breakdown pulse

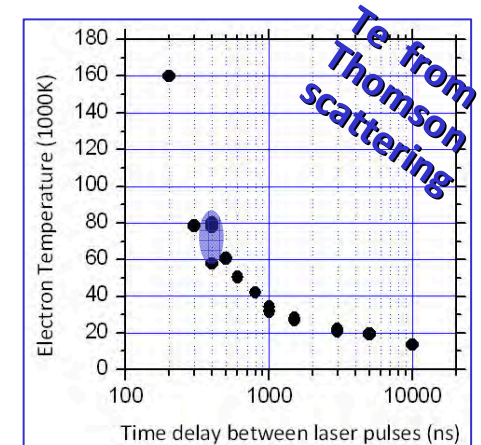
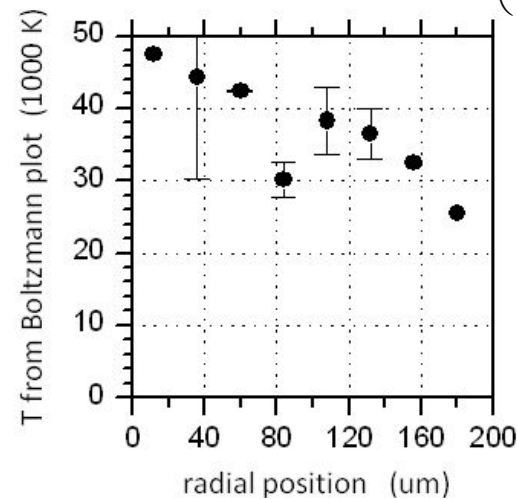


↳ Boltzmann plot

$$\epsilon_{ij} = A_{ij} \frac{h\nu}{4\pi} n_i \Rightarrow I_{ij} \propto n_i \quad \text{if plasma optically thin}$$

$$n_i = n \frac{g_i}{Q(T)} \exp(-E_i / k_B T) \quad \text{if excitation equilibrium}$$

$$\Rightarrow Y = \ln \left(\frac{\lambda_{ij} I_{ij}}{A_{ij} g_i} \right) = -E_i / k_B T + C$$

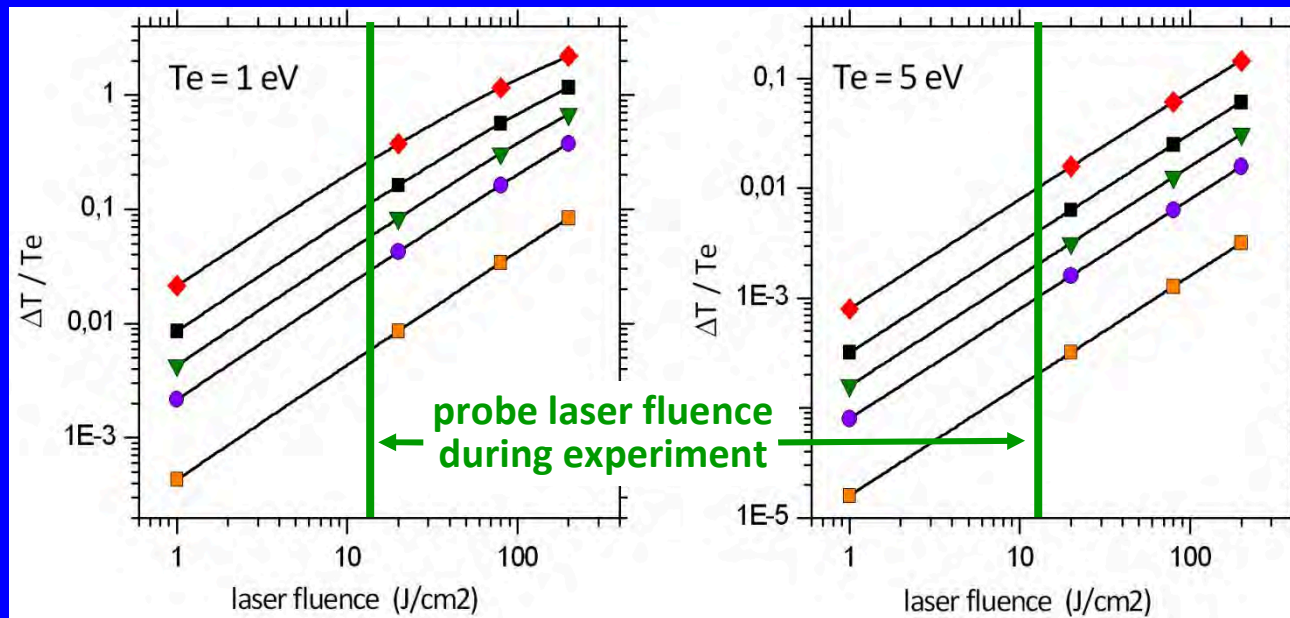


! Excitation temperature defined under assumptions of LTE plasma !

Laser scattering from laser spark in Ar at P_{atm}

↳ **Electron temperature perturbation**
due to electron heating by the probe pulse in
the **process of inverse bremsstrahlung** ?

LIBS = thermal plasmas: low T_e and high N_e \Rightarrow very susceptible to el. heating



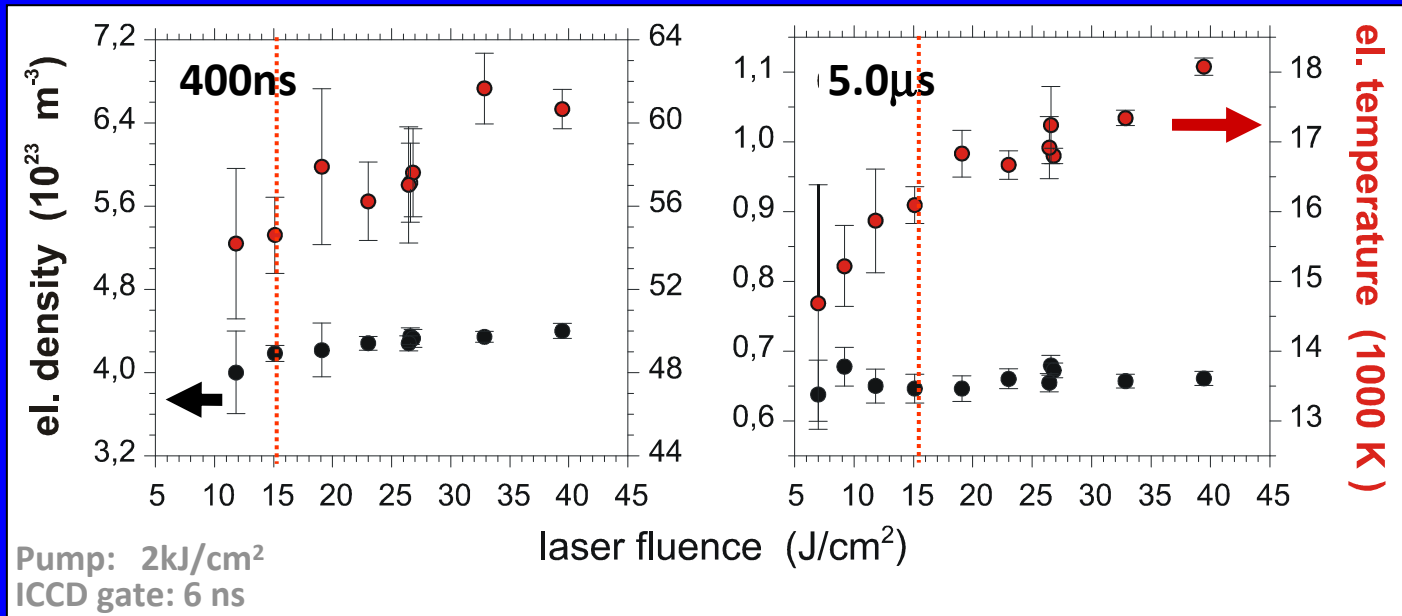
- ◆ $5.0 \times 10^{23} \text{ m}^{-3}$
- $2.0 \times 10^{23} \text{ m}^{-3}$
- ▼ $1.0 \times 10^{23} \text{ m}^{-3}$
- $5.0 \times 10^{22} \text{ m}^{-3}$
- $1.0 \times 10^{22} \text{ m}^{-3}$

Heating LIP plasma by probe laser strongly
depends on its initial conditions

Laser scattering from laser spark in Ar at P_{atm}

↳ **Electron temperature perturbation:
Influence on probe laser fluence on T_e**

LIP in argon at 1atm



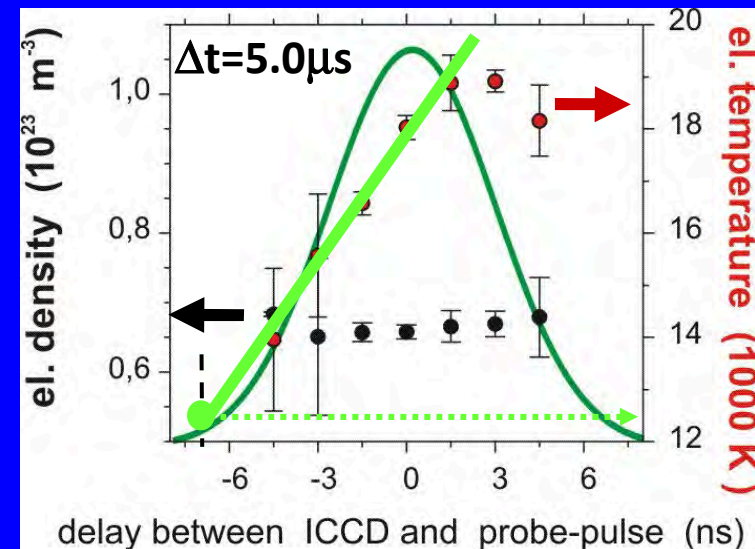
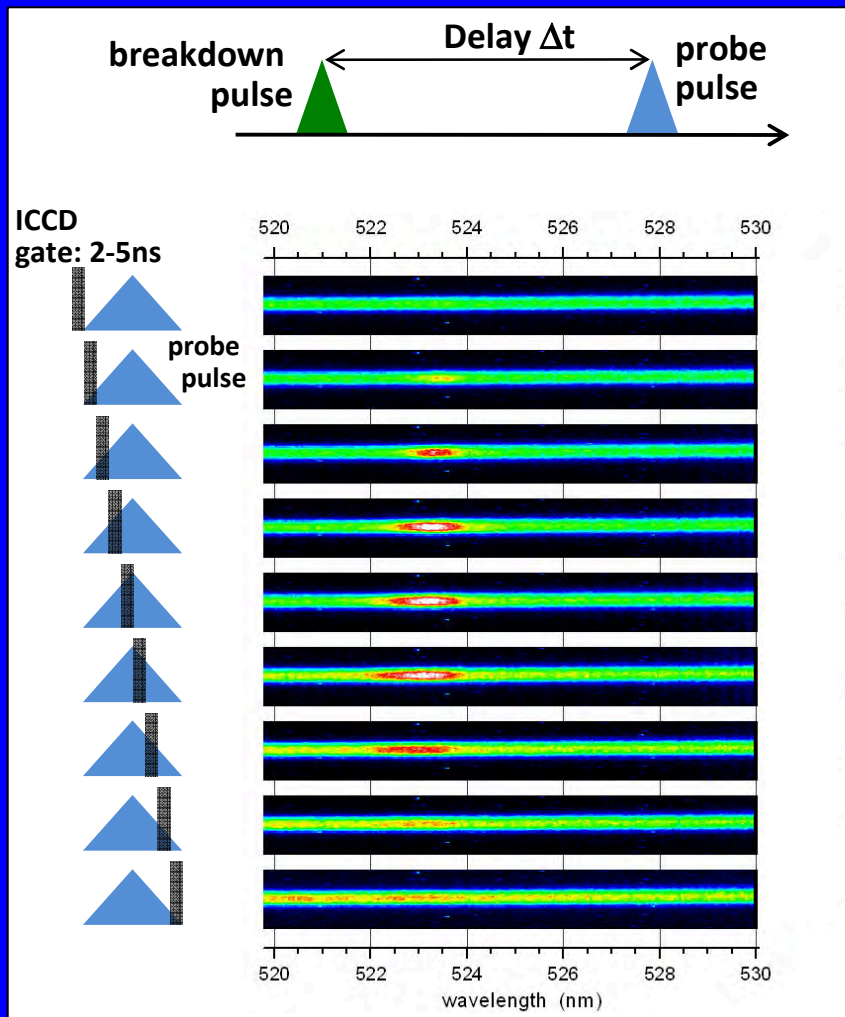
- Below some value of laser fluence no significant change of T_e is observed but only **uncertainties are rapidly growing**
- For high values of laser fluence, **Electron temperature can be significantly elevated by the probe laser pulse**

A. Mendys, et al., *Spectrochim. Acta B* (2011) **66**, 691; doi:10.1016/j.sab.2011.08.002

Laser scattering from laser spark in Ar at P_{atm}

↳ **Electron temperature perturbation:
 T_e evolution during the probe laser pulse**

Probe laser fluence: $50\text{J}/\text{cm}^2$,
ICCD gate width: 3ns

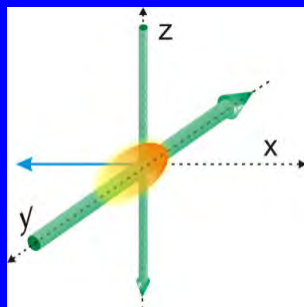
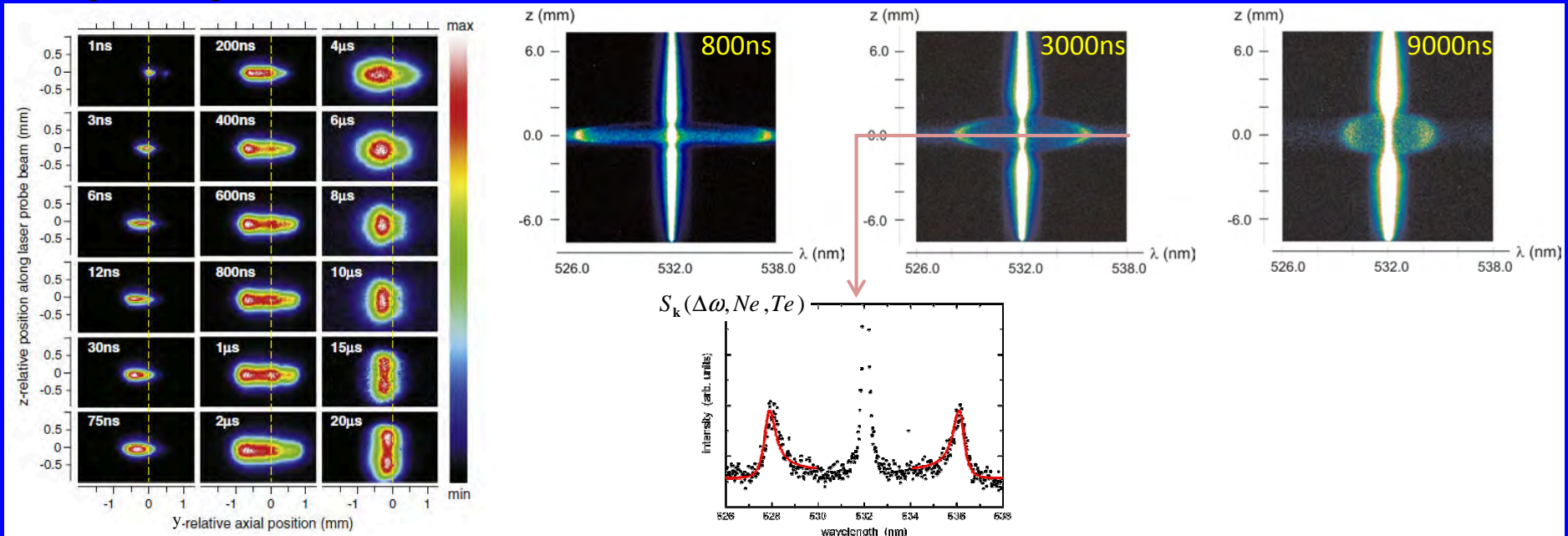


Unlike N_e , T_e is significantly disturbed as the result of much shorter time scale of inverse bremsstrahlung process

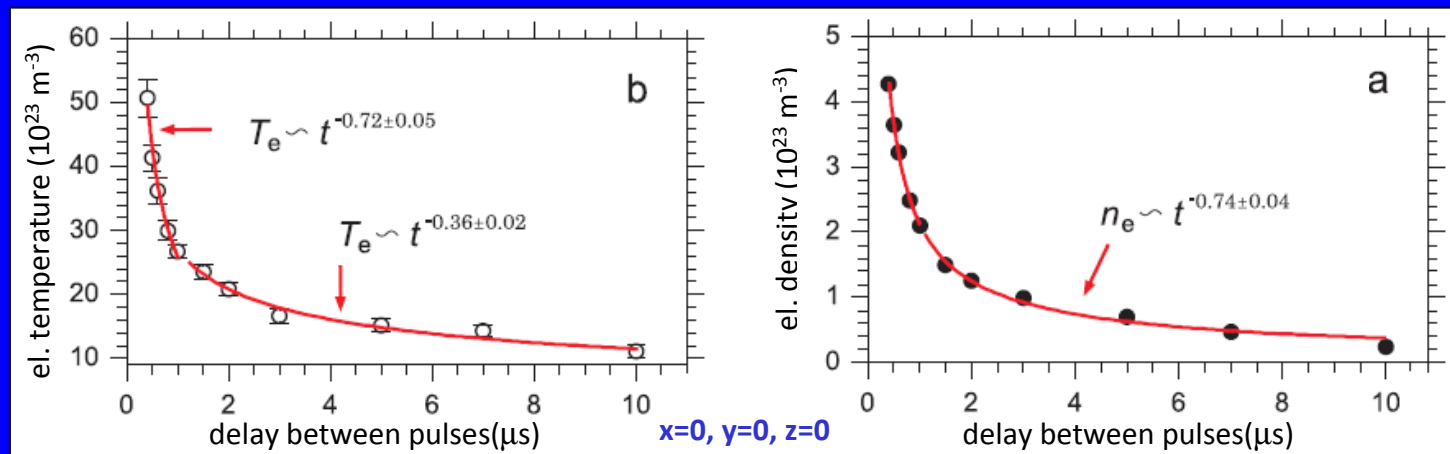
A. Mendys, et al., *Spectrochim. Acta B* (2011) **66**, 691; doi:10.1016/j.sab.2011.08.002

Laser scattering from laser spark in Ar at P_{atm}

N_e and T_e evolution on axis of LIP generated in argon by 6ns pulse of $2\text{kJ}/\text{cm}^2$ fluence



Pump : $2\text{kJ}/\text{cm}^2$
 Probe : $16\text{J}/\text{cm}^2$
 ICCD gate – 6 ns

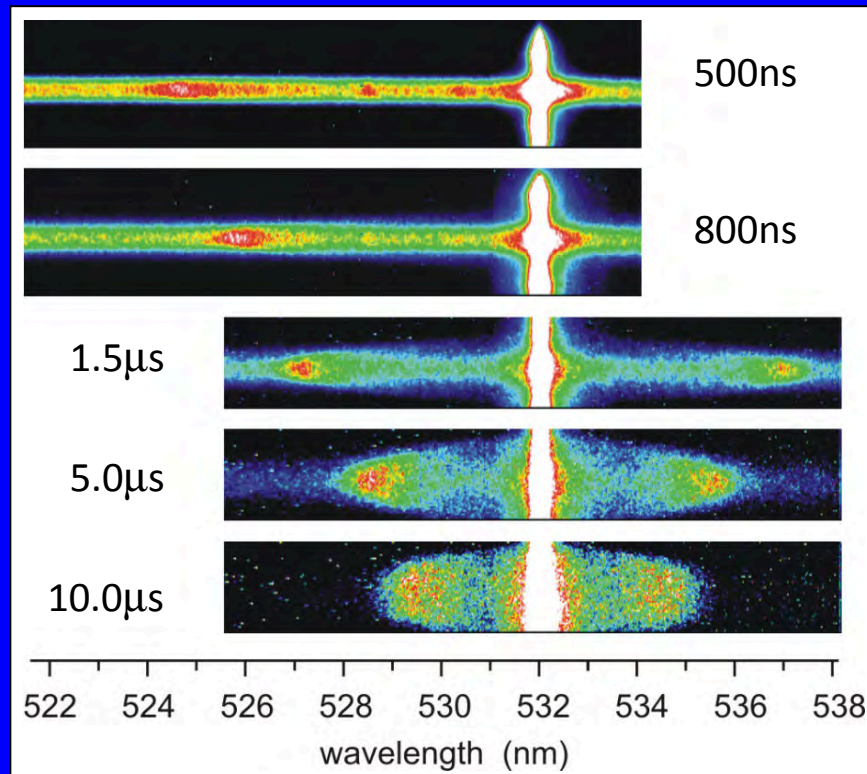
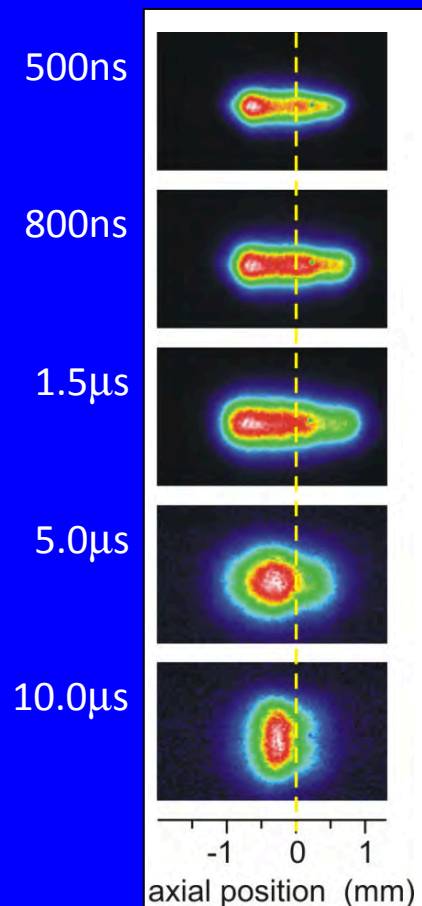


A. Mendys, et al., *Spectrochim. Acta B* (2011) **66**, 691; doi:10.1016/j.sab.2011.08.002

Laser scattering from laser spark in Ar at P_{atm}

N_e and T_e evolution on axis of LIP generated in argon by 6ns pulse of $2\text{kJ}/\text{cm}^2$ fluence

↳ **Electron feature of TS diminishes at short delays due to high continuum plasma radiation**

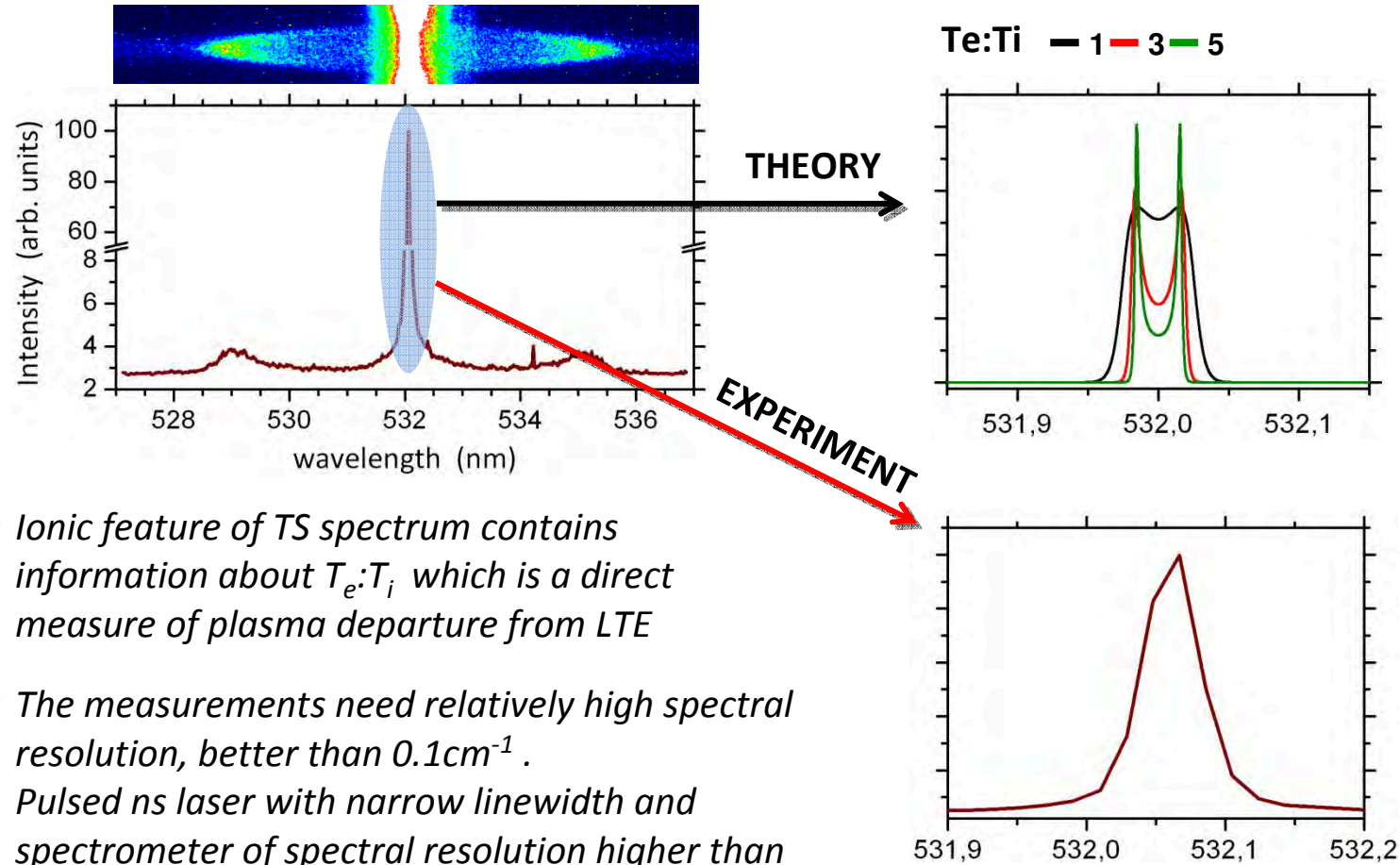


High continuum radiation of plasma significantly reduces S/N of electron feature of TS signal, only ion feature can be considered



Ion feature of TS spectrum: Determination of the ion temp.

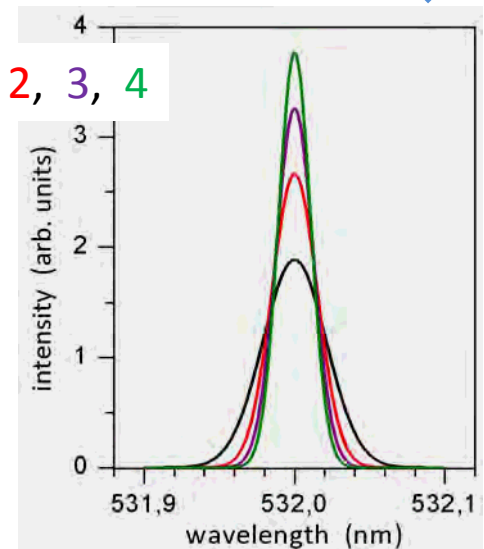
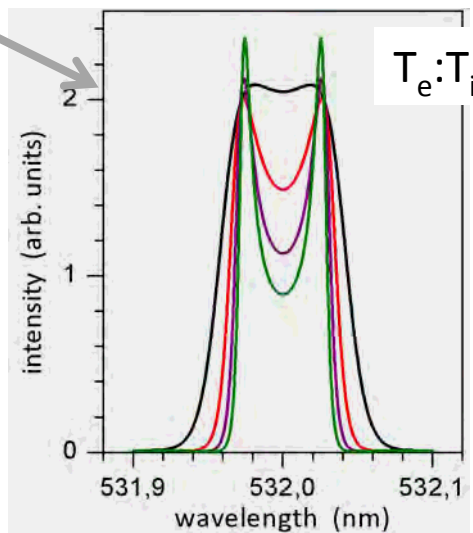
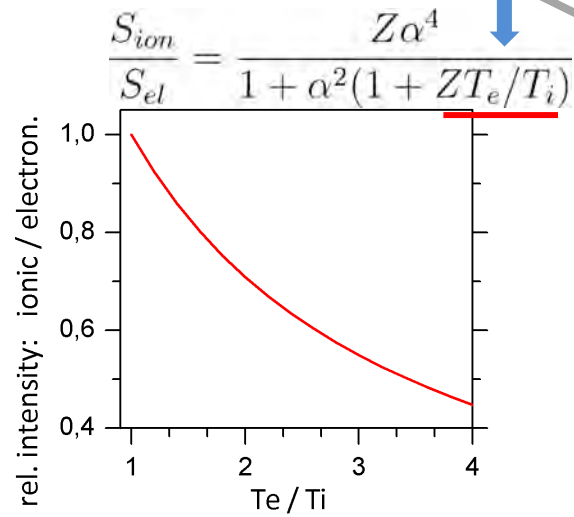
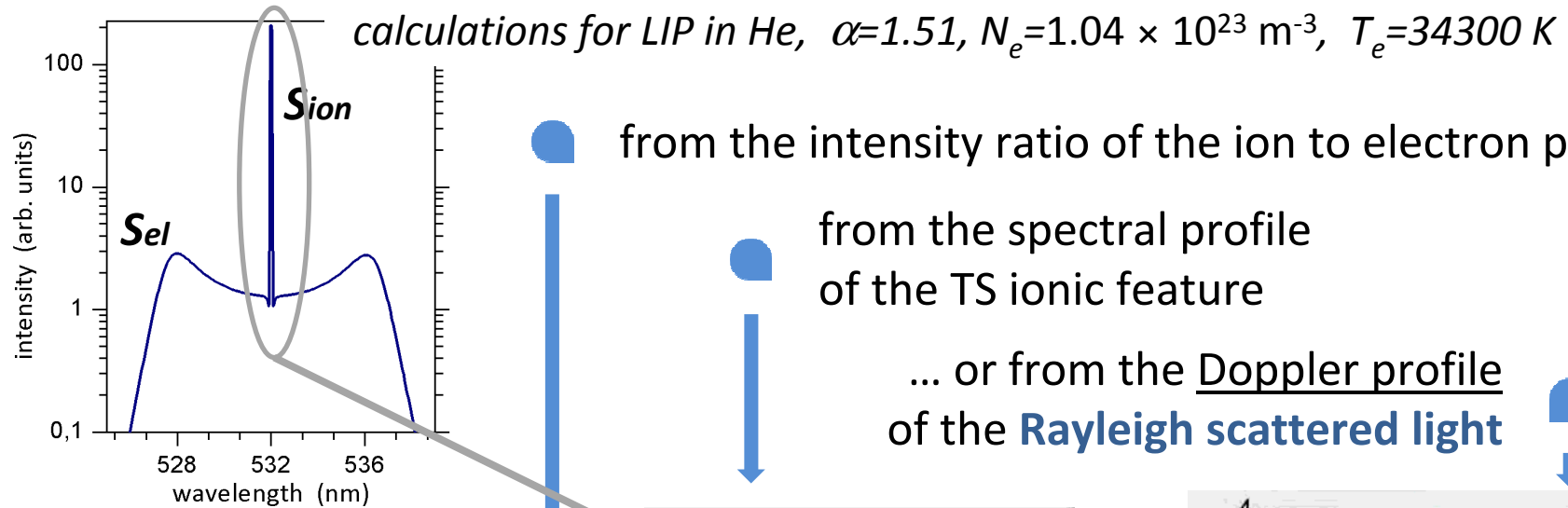
↳ indispensable to verify the thermodynamic equilibrium of LIP



- Ionic feature of TS spectrum contains information about $T_e:T_i$ which is a direct measure of plasma departure from LTE
- The measurements need relatively high spectral resolution, better than 0.1cm^{-1} . Pulsed ns laser with narrow linewidth and spectrometer of spectral resolution higher than 100 000 are indispensable

Ion feature of TS spectrum: Determination of the ion temp.

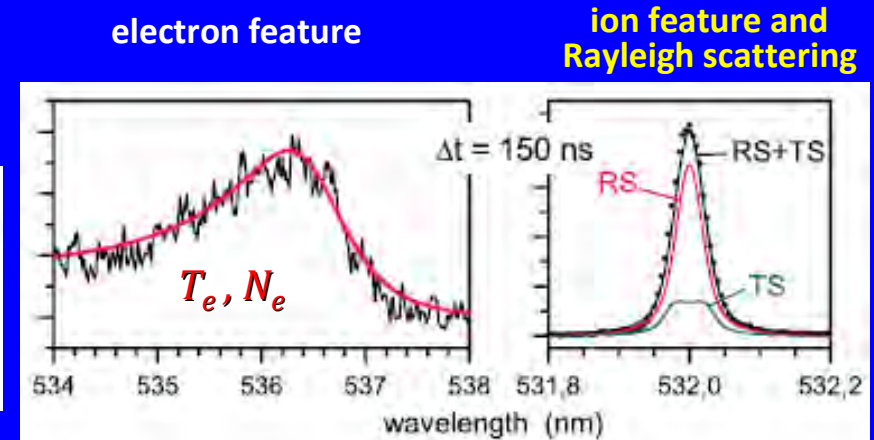
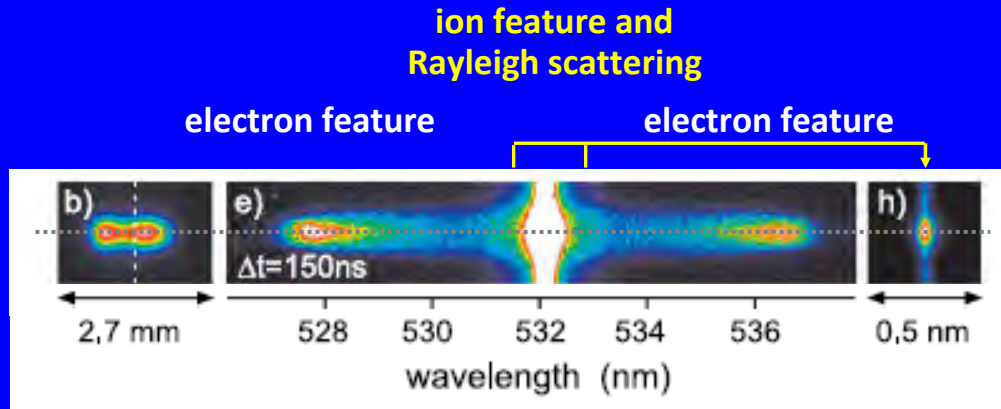
↳ indispensable to verify the thermodynamic equilibrium of LIP



Ion feature of TS spectrum: Determination of the ion temp.

Example: Helium

pump fluence – 1.2 kJ/cm²; probe fluence – 9.5 J/cm².



Data treatment

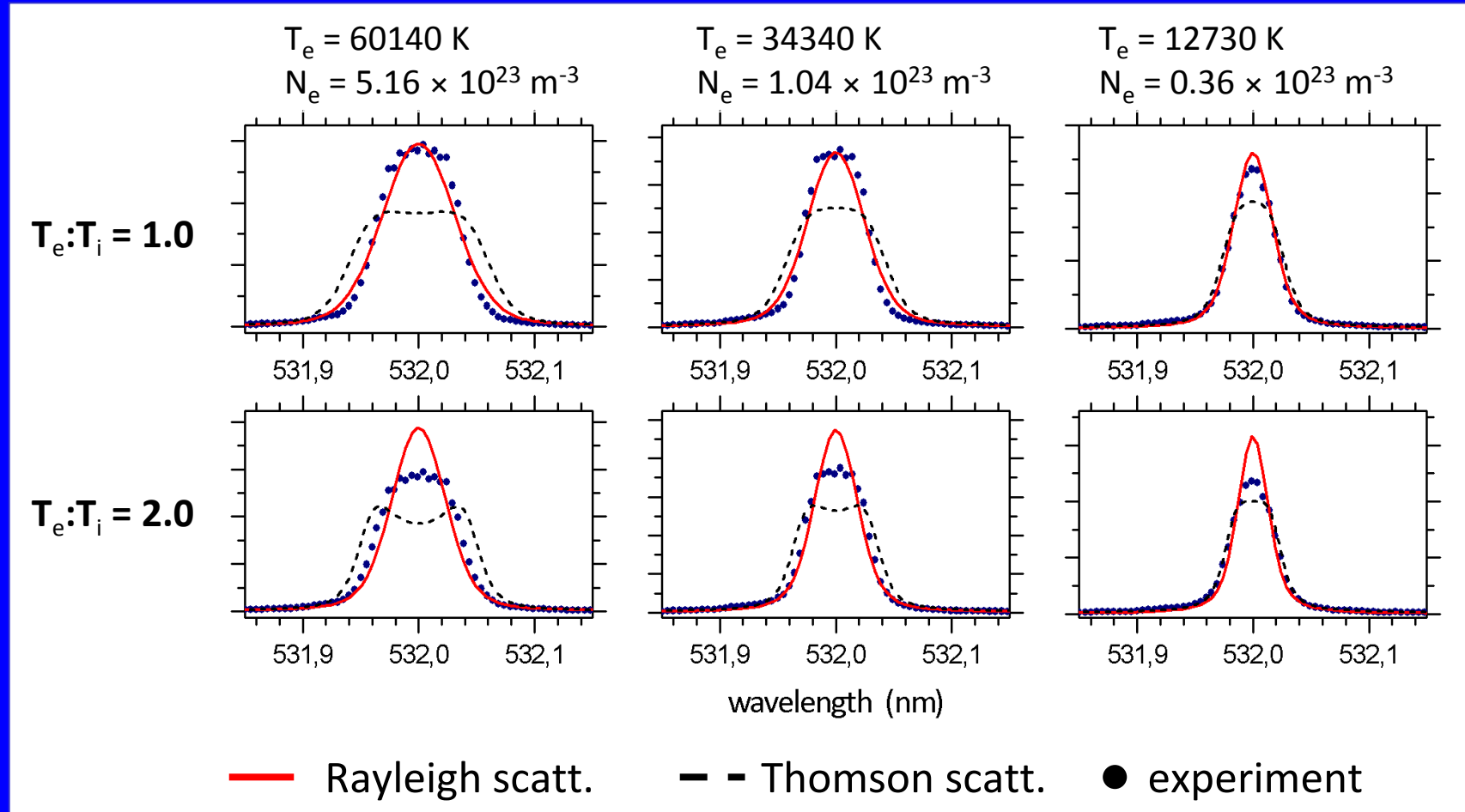
1. Determination of T_e and N_e from electronic feature
2. Deconvolution of the apparatus profile (FWHM = 0.028 nm) from central peak (h)
3. Fitting ion feature of TS and Rayleigh spectra with T_i as a parameter

K.Dzierzega et al. Appl. Phys. Lett. **102**, 134108 (2013)

Ion feature of TS spectrum: Determination of the ion temp.

Example: Helium

pump fluence – 1.2 kJ/cm²; probe fluence – 9.5 J/cm².



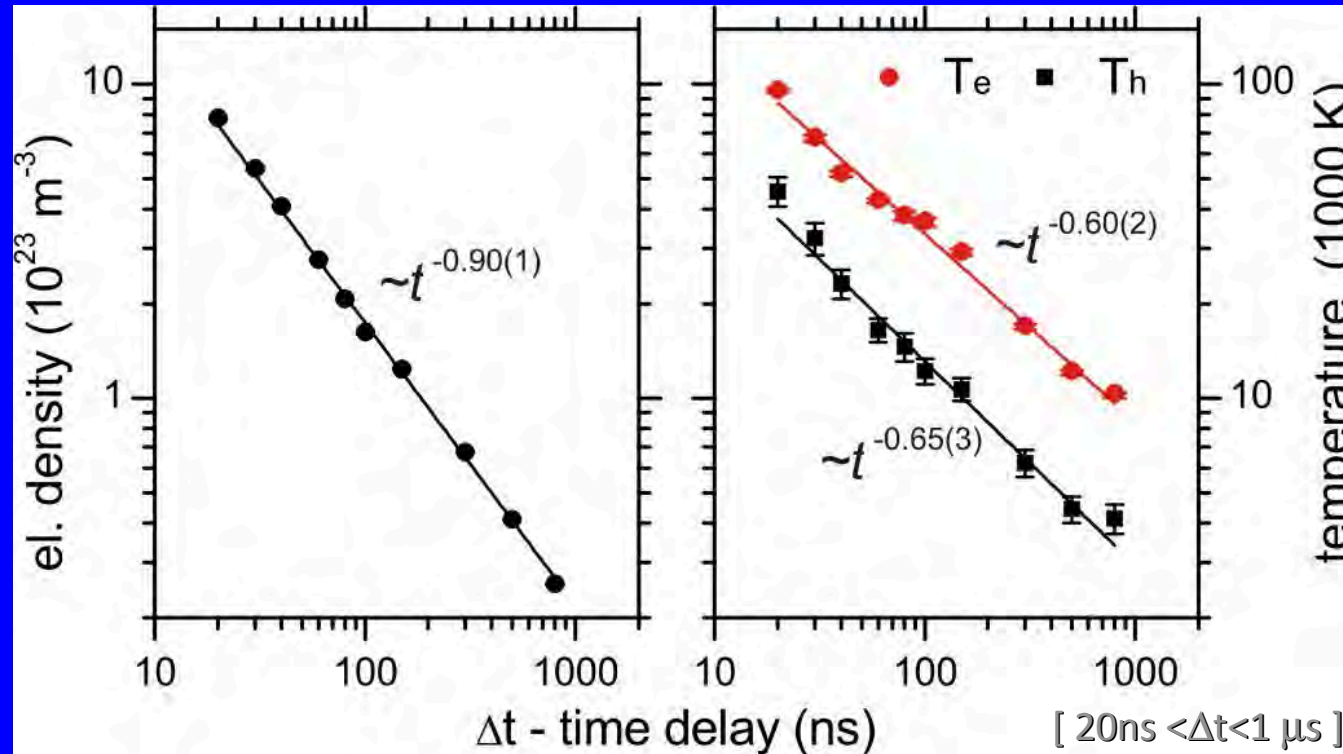
☞ Ion temperature measurement: difficult to discriminate Thomson from Rayleigh contribution

K.Dzierzega et al. *Appl. Phys. Lett.* **102**, 134108 (2013)

Ion feature of TS spectrum: Determination of the ion temp.

Example: Helium

pump fluence – 1.2 kJ/cm²; probe fluence – 9.5 J/cm².



$10^{24} \text{ m}^{-3} > N_e > 10^{22} \text{ m}^{-3}$
 $100 \cdot 10^3 \text{ K} > T_e > 10 \cdot 10^3 \text{ K}$
 $50 \cdot 10^3 \text{ K} > T_h > 2 \cdot 10^3 \text{ K}$

→ Deviation from isothermal equilibrium

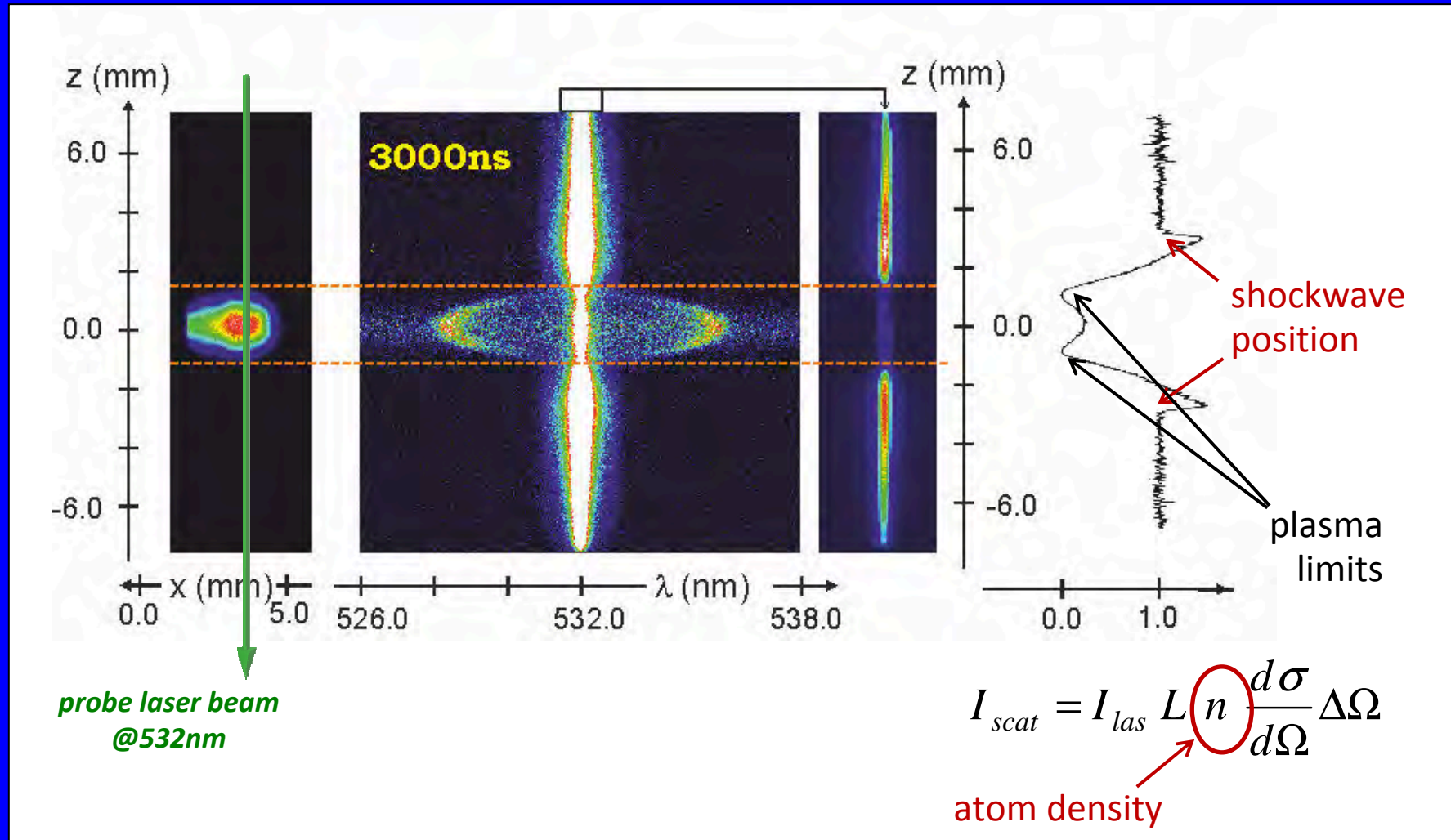
Large discrepancy between electron and heavy particle temperatures which indicates two-temperature plasma out of the local isothermal equilibrium.

Results do not rely on assumptions about the equilibrium state of the plasma!

K.Dzierzega et al. Appl. Phys. Lett. 102, 134108 (2013)

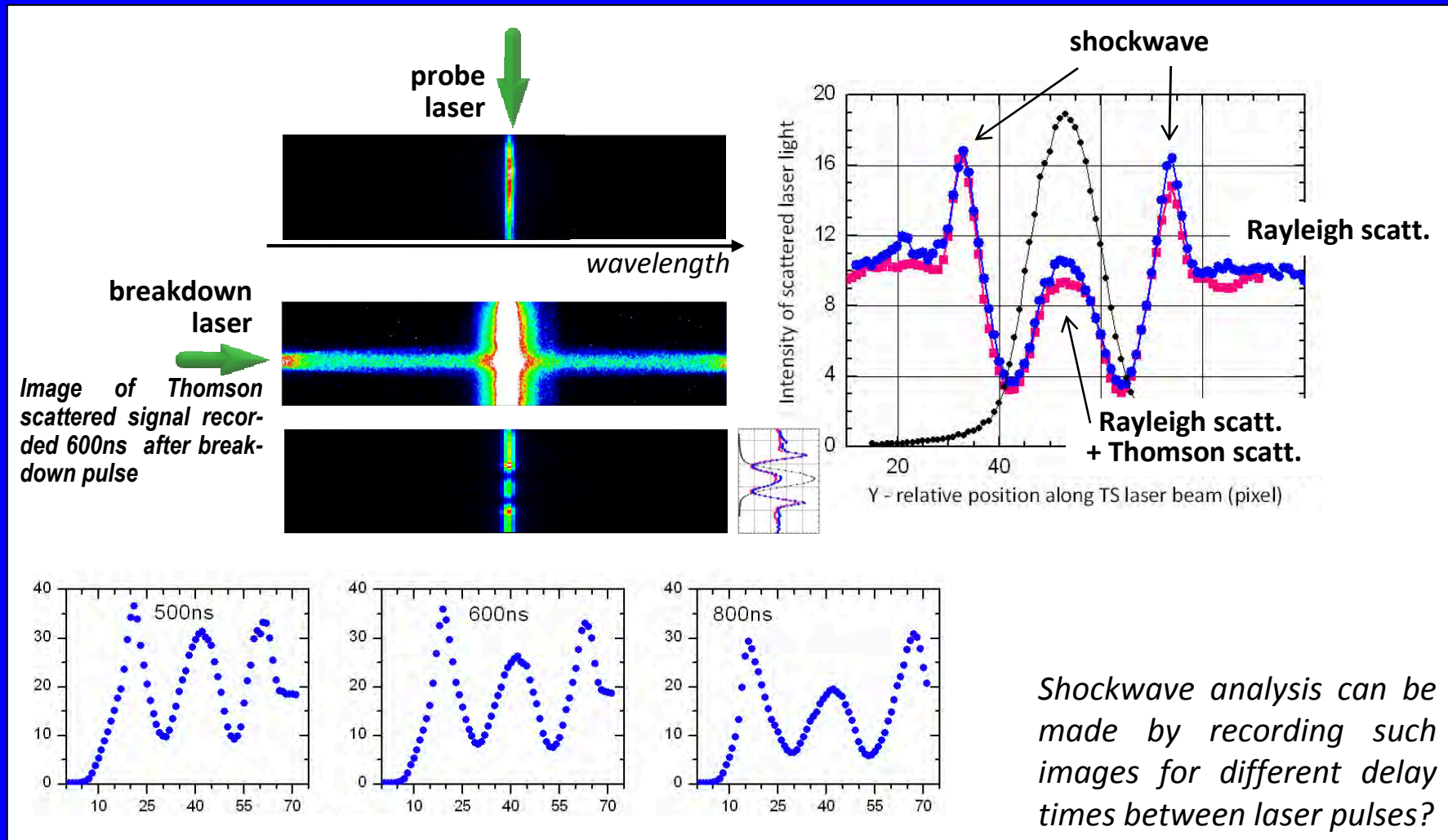


Rayleigh scattering from laser induced plasma



B.Pokrzywka et al., Spectrochim. Acta B (2012) 74–75, 24

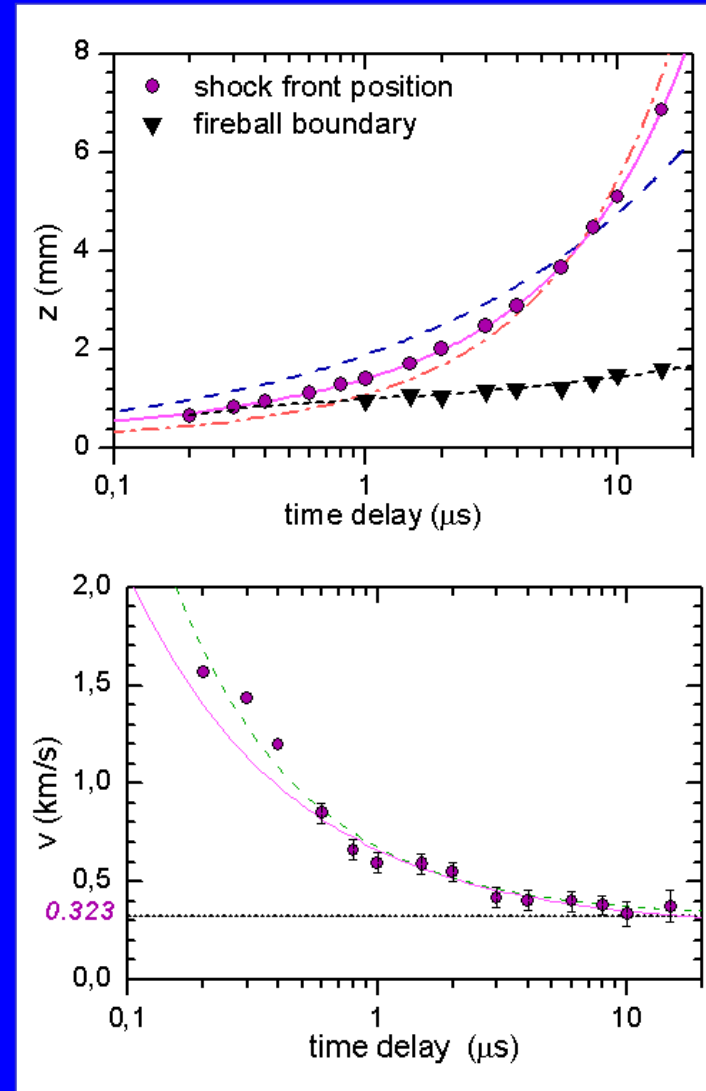
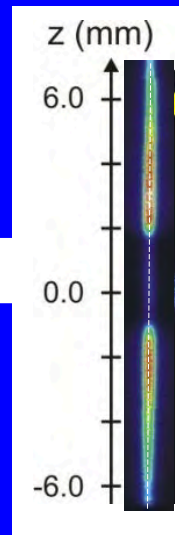
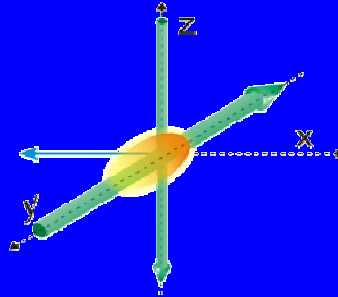
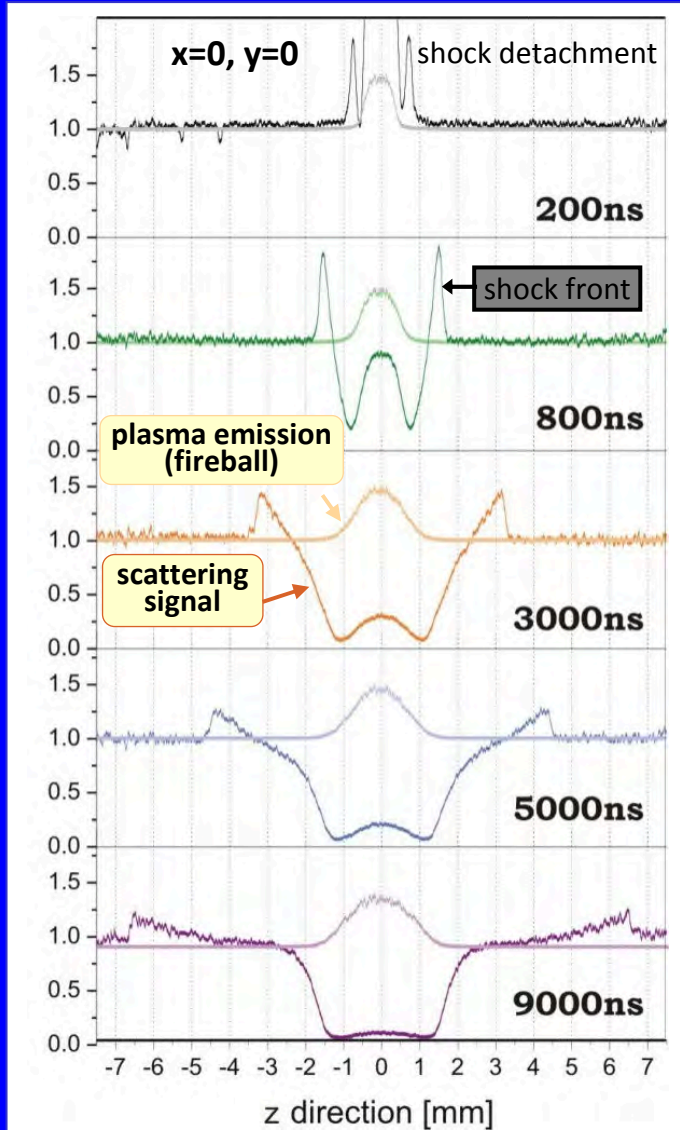
Rayleigh scattering from laser induced plasma



B.Pokrzywka et al., Spectrochim. Acta B (2012) 74–75, 24

Rayleigh scattering from laser induced plasma

Shockwave evolution on axis of LIP generated in argon by 6ns pulse of 2kJ/cm² fluence

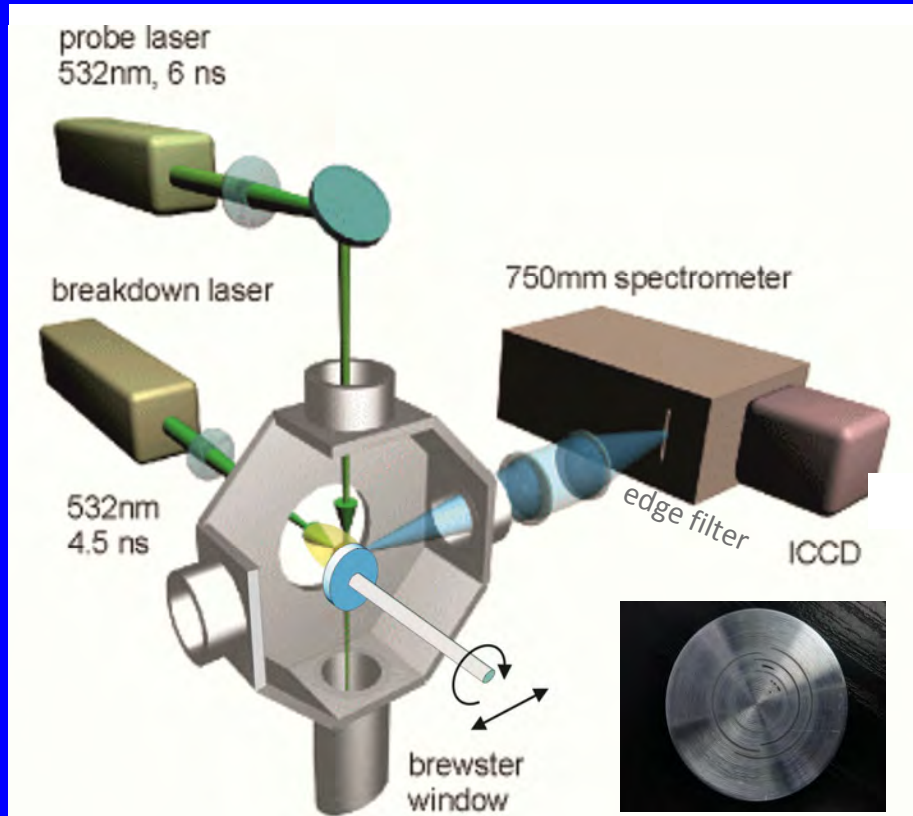


B.Pokrzywka et al., *Spectrochim. Acta B* (2012) 74–75, 24

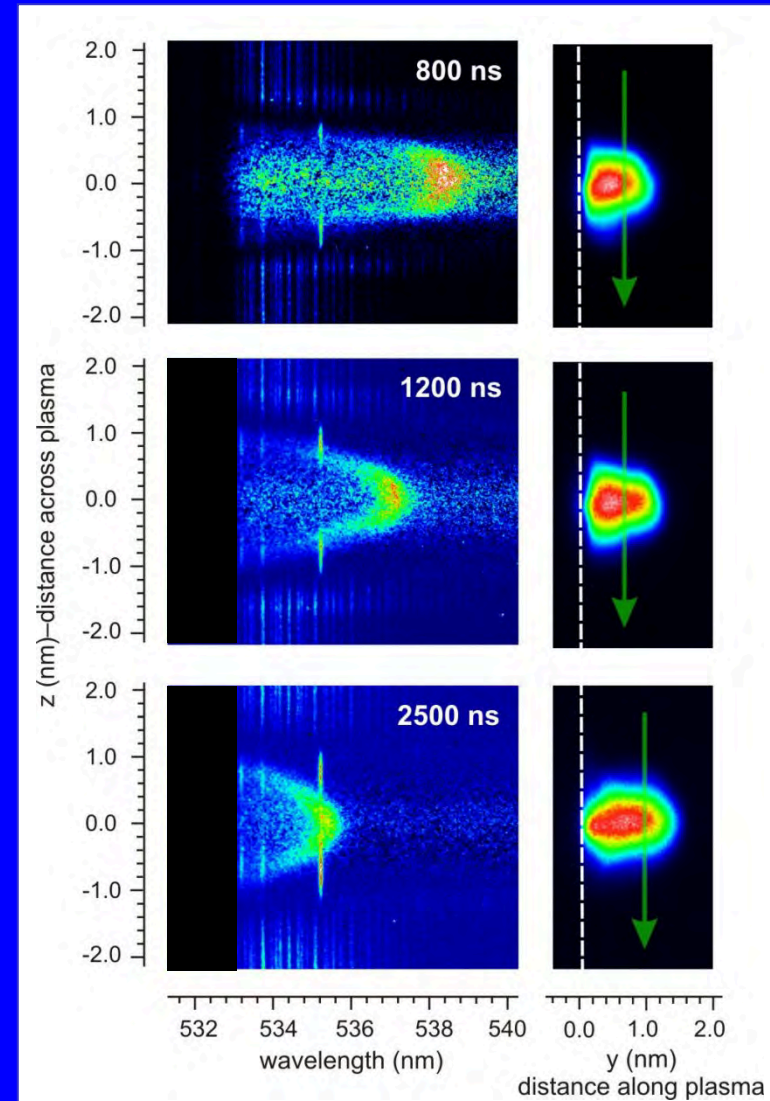


LIP generated on Al target in air

↳ Set-up and Optical scattering



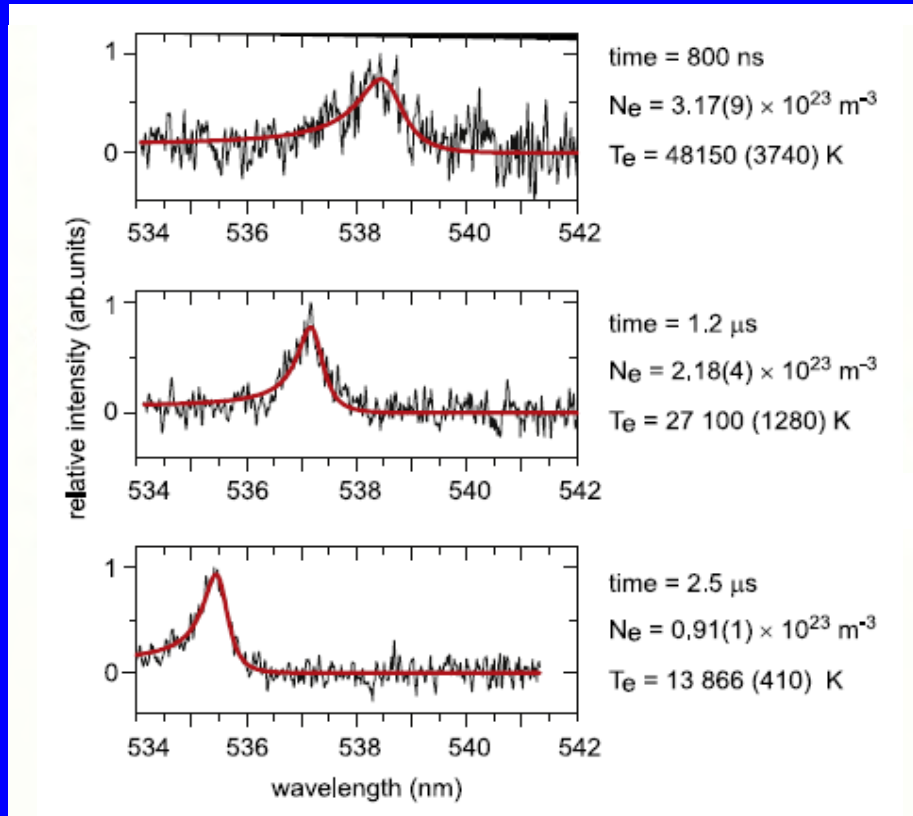
ablating laser: fluence 30 J/cm^2 ($6.7 \times 10^9 \text{ W/cm}^2$)
probe beam: fluence 100 J/cm^2 ($16.6 \times 10^9 \text{ W/cm}^2$)
Spatial resolution: $200\mu\text{m}$ (radial) \times $30\mu\text{m}$ (axial)
ICCD gate: 8 ns - 2000 laser shots



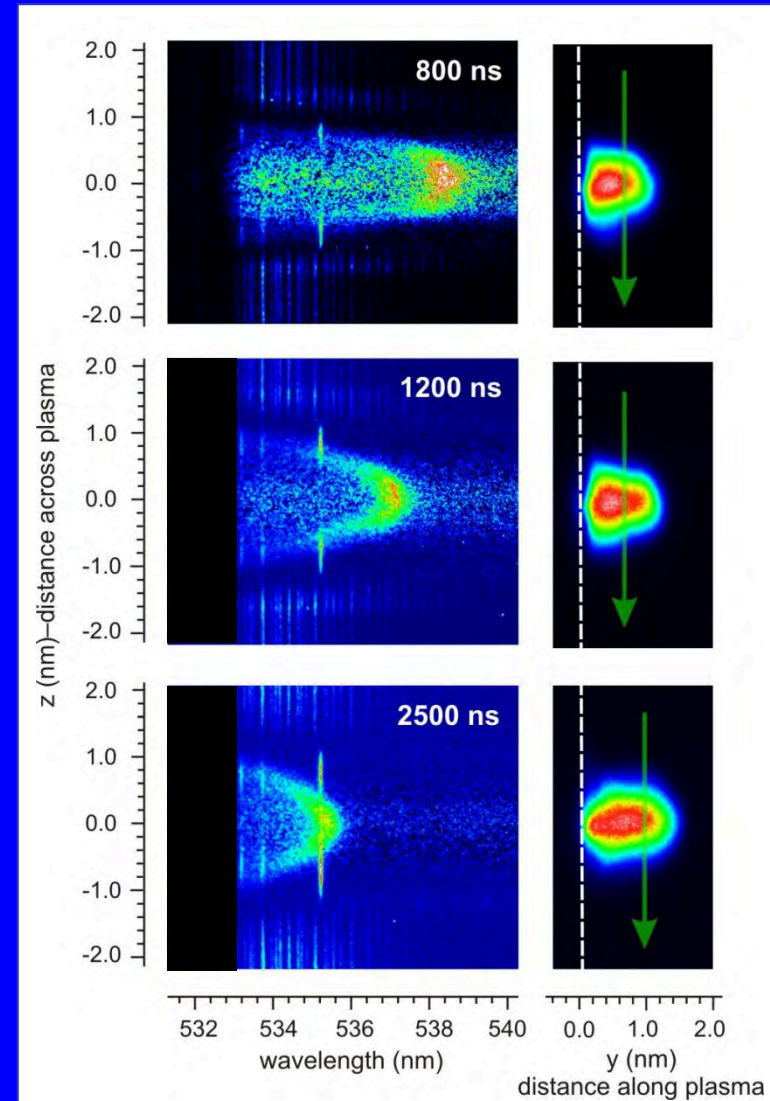
A. Mendys, et al. *Spectrochim. Acta B* **96**, 61–68, (2014) doi:10.1016/j.sab.2014.03.009

LIP generated on Al target in air

↳ Set-up and Optical scattering



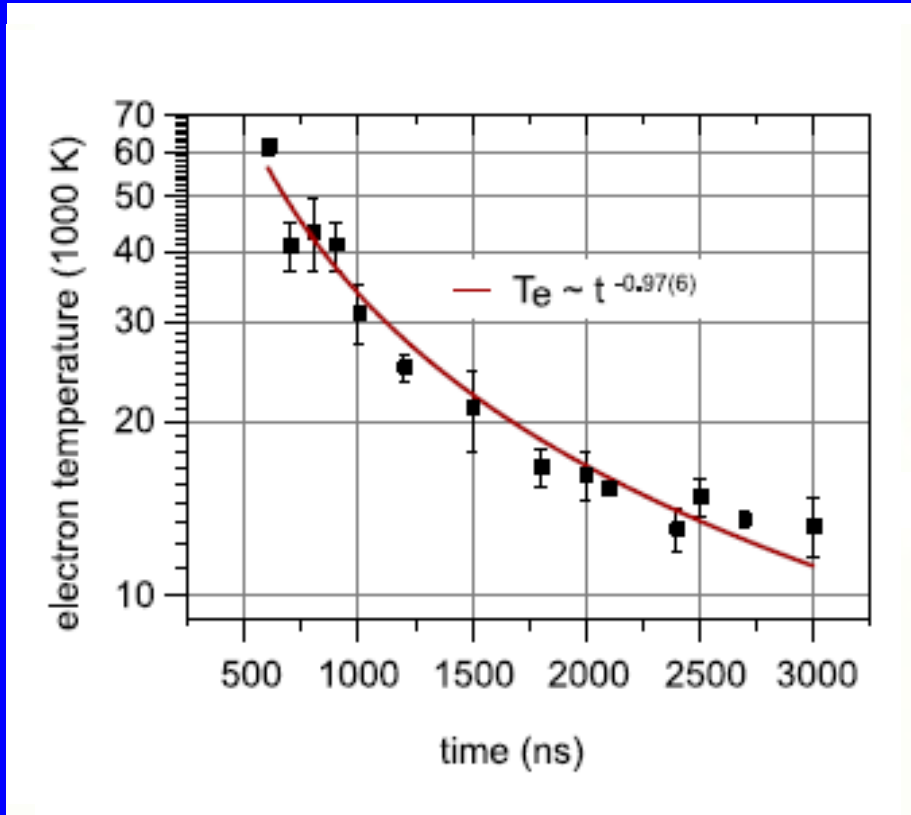
ablating laser: fluence 30 J/cm^2 ($6.7 \times 10^9 \text{ W/cm}^2$)
probe beam: fluence 100 J/cm^2 ($16.6 \times 10^9 \text{ W/cm}^2$)
Spatial resolution: $200\mu\text{m}$ (radial) \times $30\mu\text{m}$ (axial)
iCCD gate: 8 ns - 2000 laser shots



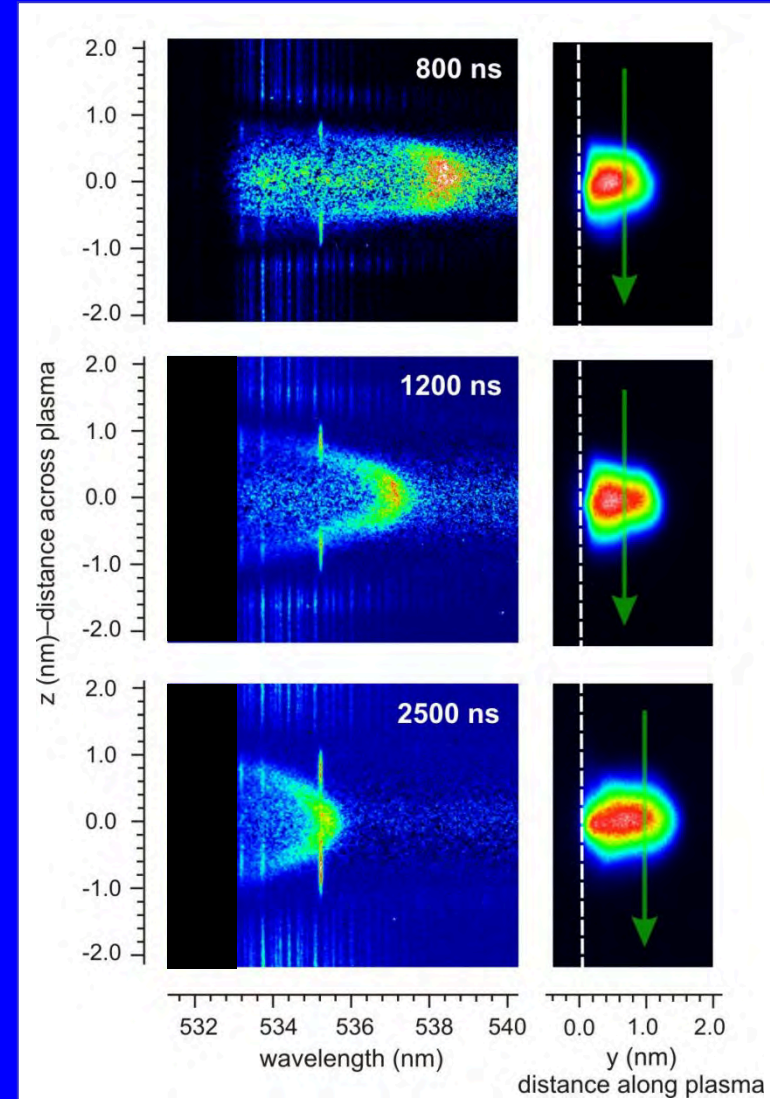
A. Mendys, et al. *Spectrochim. Acta B* **96**, 61–68, (2014) doi:10.1016/j.sab.2014.03.009

LIP generated on Al target in air

↳ Set-up and Optical scattering



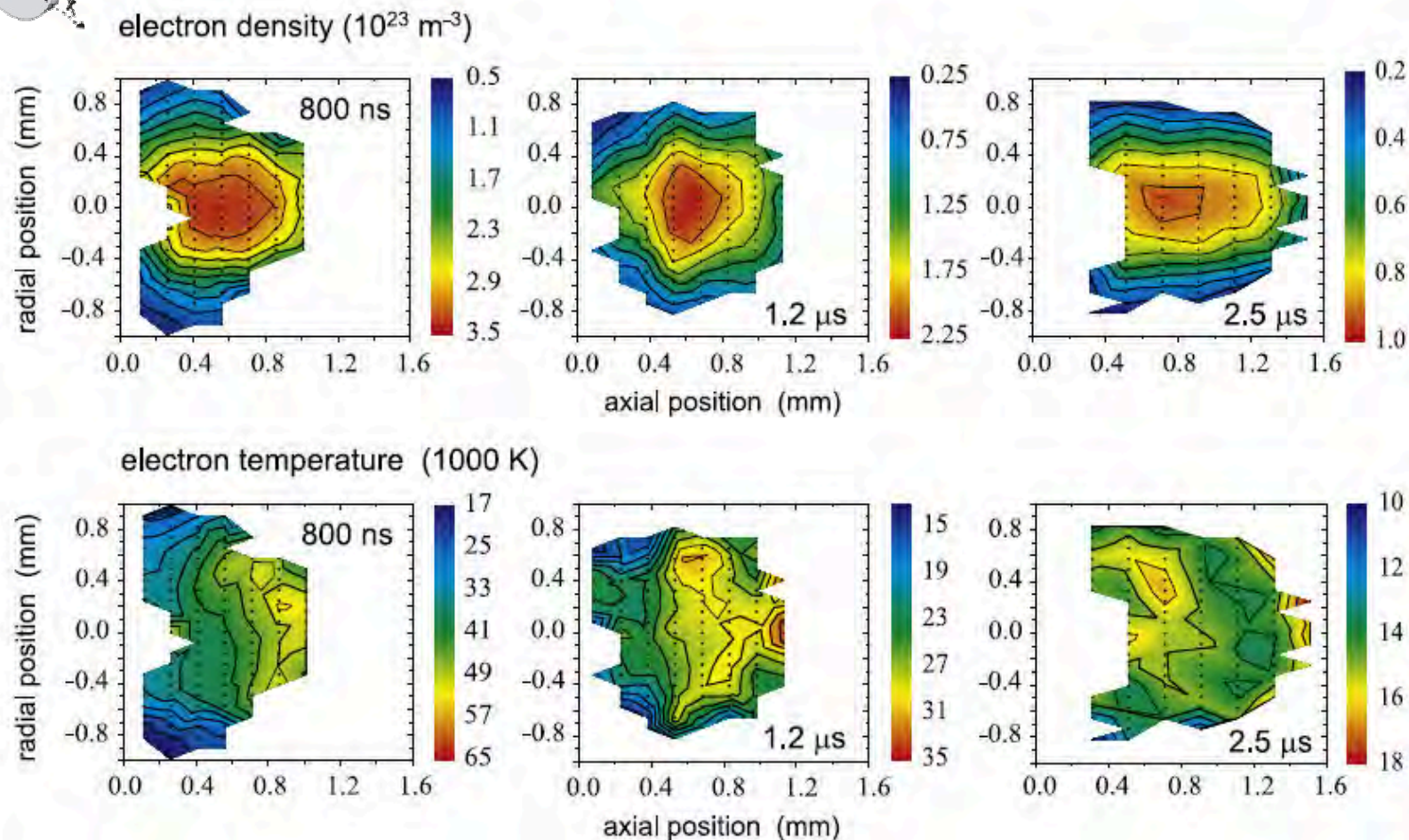
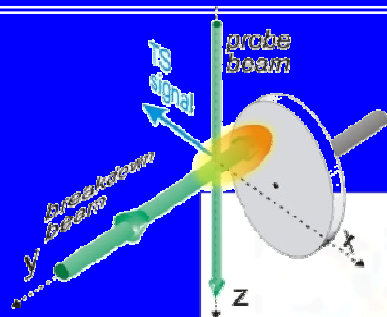
ablating laser: fluence 30 J/cm^2 ($6.7 \times 10^9 \text{ W/cm}^2$)
probe beam: fluence 100 J/cm^2 ($16.6 \times 10^9 \text{ W/cm}^2$)
Spatial resolution: $200\mu\text{m}$ (radial) \times $30\mu\text{m}$ (axial)
iCCD gate: 8 ns - 2000 laser shots



A. Mendys, et al. *Spectrochim. Acta B* **96**, 61–68, (2014) doi:10.1016/j.sab.2014.03.009

LIP generated on Al target in air

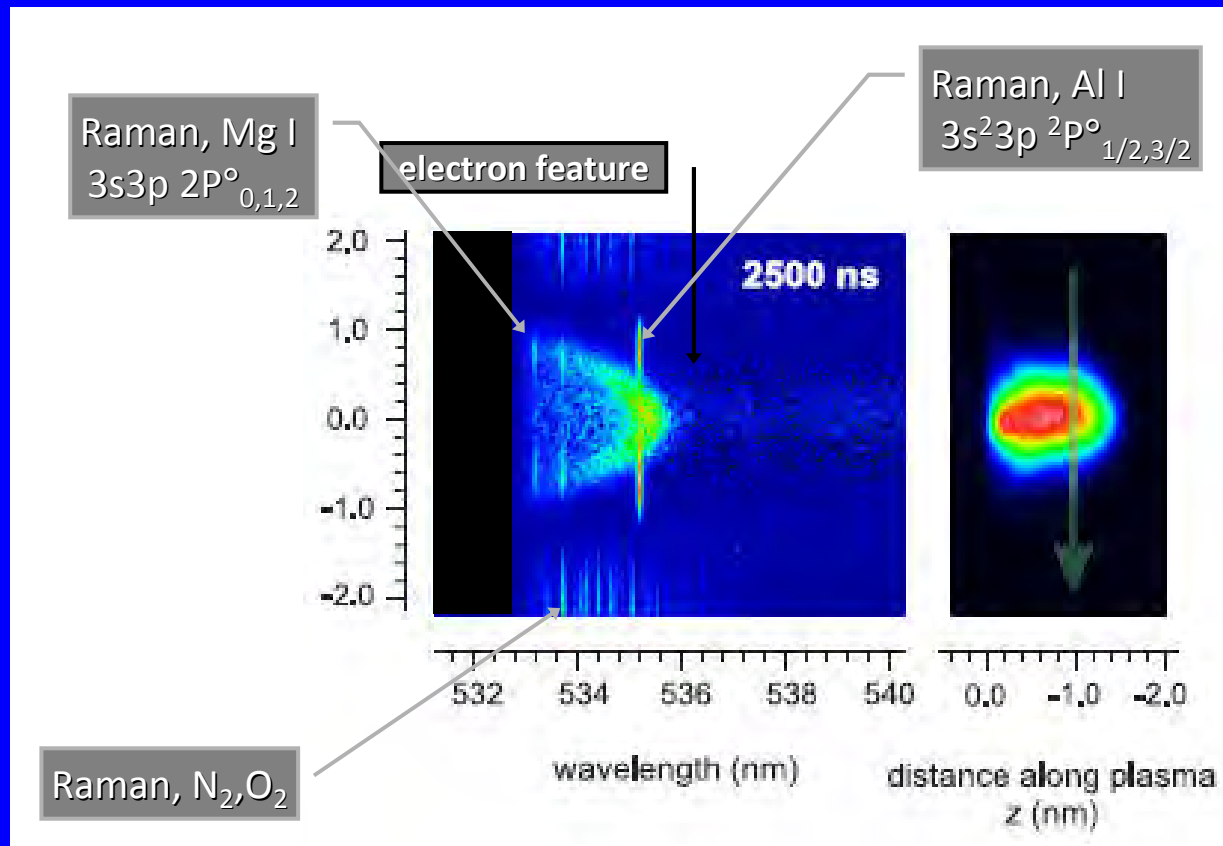
↪ N_e , T_e distribution in the plume



A. Mendys, et al. *Spectrochim. Acta B* 96, 61–68, (2014) doi:10.1016/j.sab.2014.03.009

LIP generated on Al target in air

↳ Perspective: Raman scattering



Ref: A.Delserieys, F.Y.Khattak, S.Sahoo, G.F.Gribakin, C.L.S.Lewis, and D.Riley, *Raman satellites in optical scattering from a laser-ablated Mg plume; Physical Review A* **78** (2008) 055404

➡ Raman scattering could be used for diagnosing the relative populations of excited levels in a partially ionized plasma.

Summary and Conclusions

- Laser scattered light from LIBS plasma gives a **lot of information on plasma, especially on transient one.**
- **Simultaneous measurement** of plasma parameters
 - Scattering of laser radiation is **local method – high spatial resolution**
- Direct prediction of plasma parameters **without assumption of thermodynamic equilibrium**
 - Derivation of the most important plasma parameters like N_e and T_e from TS spectra is straightforward and usually does not need further **calibration**
- **BUT Elevation of the electron temperature (heating)** in Thomson scattering experiment, fortunately this effect can be corrected

↳ "What can we learn about laser-induced plasmas from Thomson scattering experiments" (Review),
K. Dzierzega, A.Mendys B.Pokrzywka, *Spectrochimica Acta Part B* 98 (2014) 76–86

Peoples

Group founder and former group lieder

Prof. dr hab. Karol Musioł

Smoluchowski Institute of Physics, Jagiellonian University, Kraków, Poland



Krzysztof Dzierzega

M. Smoluchowski Inst. of Physics, Jagellonian University, Cracow, Poland



Bartłomiej Pokrzywka

Mt. Suhora Obs., Inst. of Physics, Pedagogical Univ. of Cracow, Poland



Stéphane Pellerin

GREMI, Université d'Orléans/CNRS, Bourges, France



Other coworkers



Agata Mendys, Tomasz Pieta, Łucasz Bratasz, M.Grabiec, S.Łabuz , W.Zawadzki
Smoluchowski Institute of Physics, Jagiellonian University, Kraków, Poland



Ali Farah-Sougueh, Emanuelle Thouin-Leduc, Flavien Valensi
GREMI, Université d'Orléans/CNRS, Bourges, France

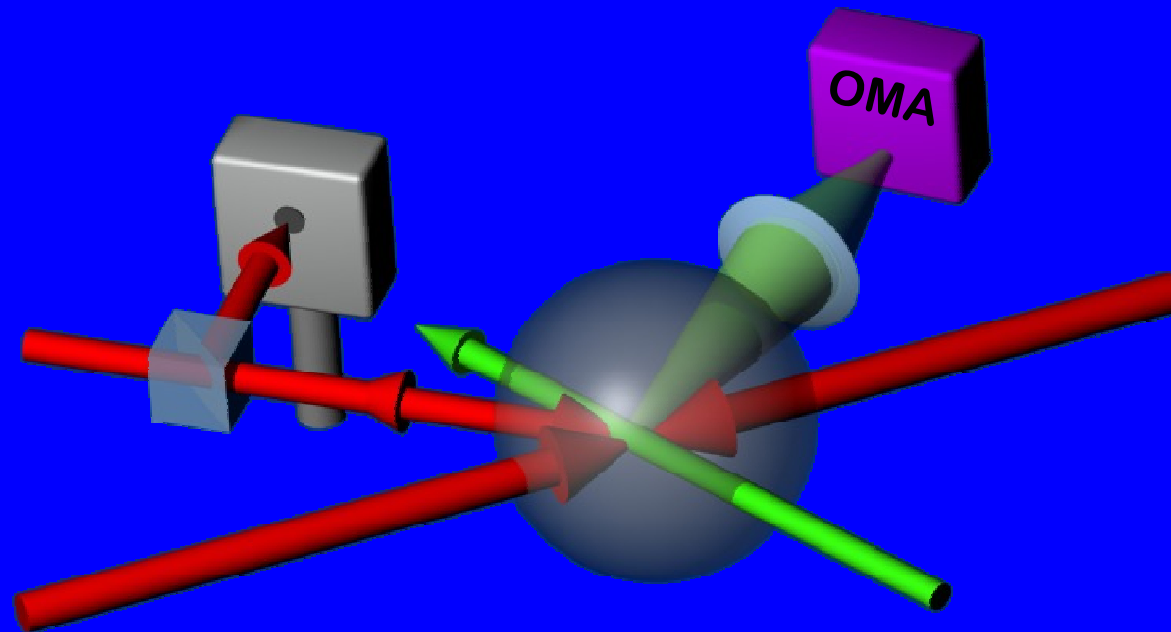


Katarzyna Bajan, Marta Krawczyk,
Institute of Physics, Pedagogical University of Cracow, Kraków, Poland



Wiesław Olchawa, Ewa Pawelec
Institute of Physics, Opole University, Opole, Poland

Thank you
for your attention



Questions ?