Laser spectroscopy with Diode Lasers

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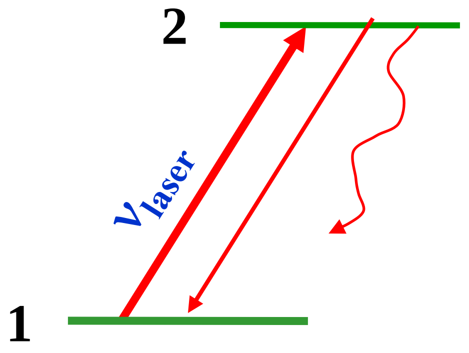
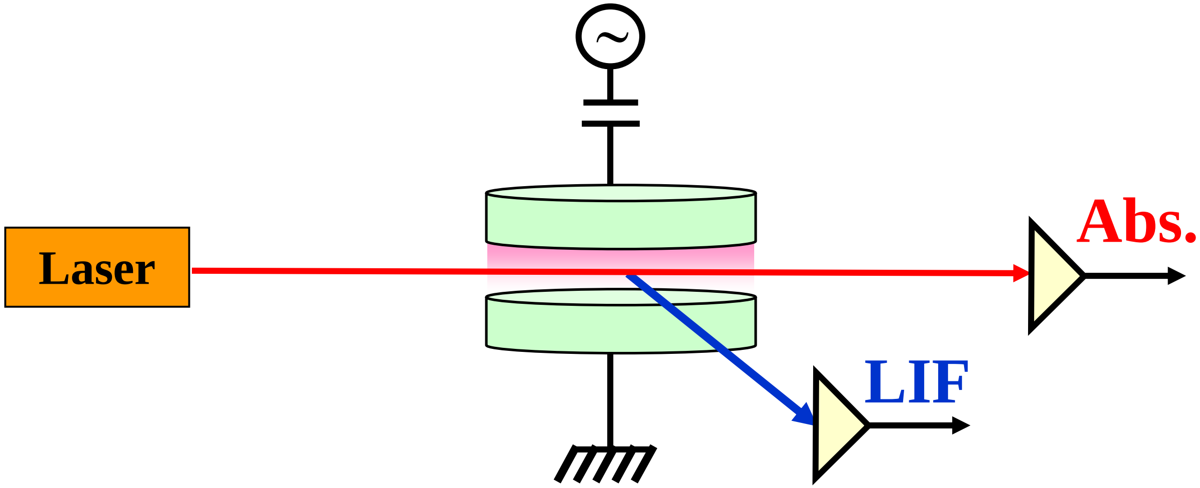
Application domain of Laser Spectroscopy

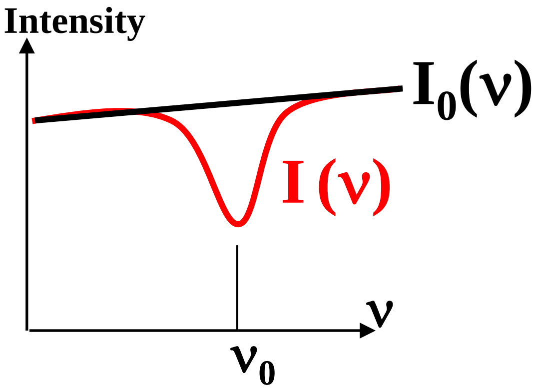
- Spectroscopy of atoms, molecules, ions, clusters:
  - Energy levels (electronic, vibration, rotation)
  - Lifetime of states
  - Collisional properties of the levels
- Determination of species density & temperature (abs.)
  - **Optical saturation phenomena**
- Other artifacts encountered in Laser Spectroscopy

Principle of Laser Spectroscopy

Laser frequency is tuned to a specific transition of interest



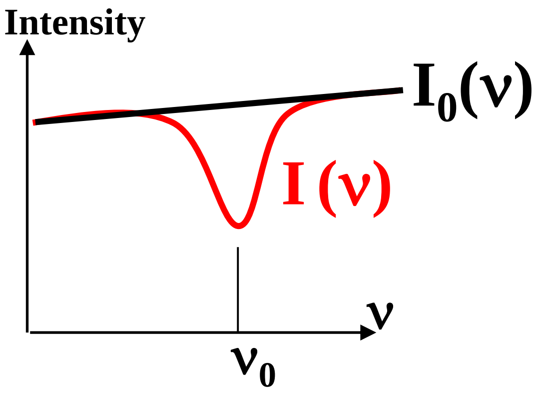




Either in Laser Absorption or in Laser Induced Fluorescence techniques, spectral information comes from the first step:

absorption of photons

Principle of Laser Spectroscopy



-From absorption signal

$$Ln\left(\frac{I_0(\nu)}{I(\nu)}\right) = l.\alpha(\nu)$$

-From LIF signal

$$I_{LIF}(\nu) \approx \Phi(P_{Laser}).\alpha(\nu)$$

Absorption Coefficient

$$\alpha(\nu) = \frac{4hB_{12}}{\lambda\gamma}\left(n_1(\nu) - \frac{g_1}{g_2}n_2(\nu)\right)$$

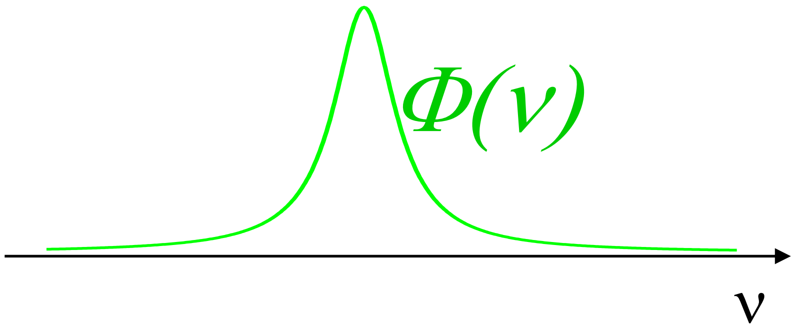
What can we learn by measuring $\alpha(\nu)$?

$$\alpha(\nu) = N k \;\Phi(\nu)$$

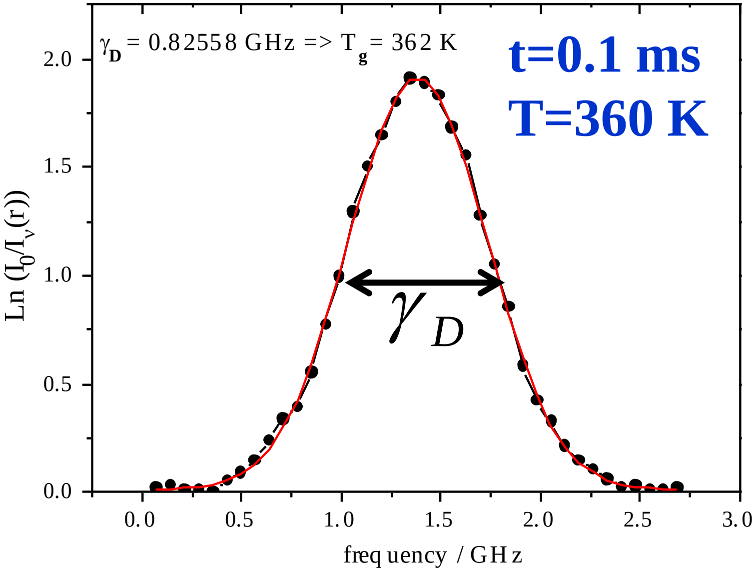
Density
traces Detection

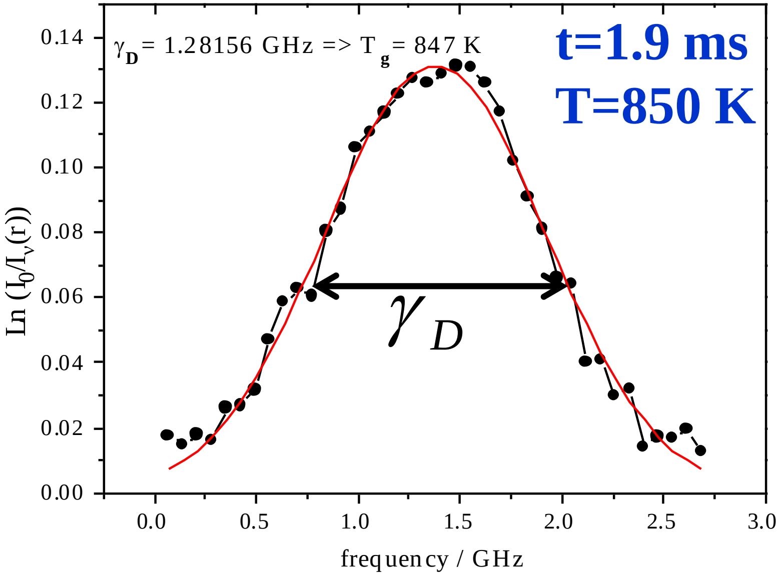
transition
probability

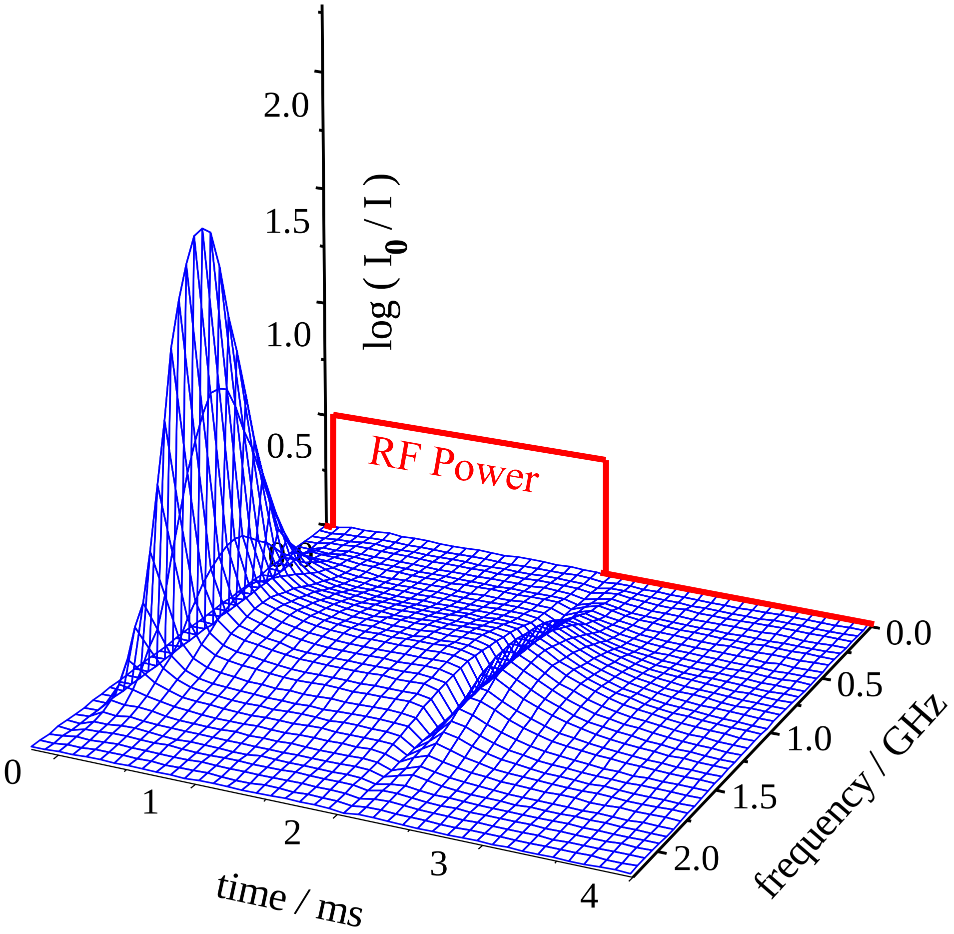
Line profile
Analysis



Density and Temperature of Ar* atoms in a high density pulsed Helicon discharge







$$\gamma_D(t) = \frac{2\sqrt{Ln2}}{\lambda_0}\sqrt{\frac{RT(t)}{M}}$$

Units used in spectroscopy

1 eV = 1.6 x 10^{-19} J = 8065 cm^{-1} **Wavenumber**

300 kT = 207 cm^{-1}

1 cm^{-1} = 30 GHz

$$\nu = \frac{c}{\lambda} \qquad \Delta\nu = \frac{c * \Delta\lambda}{\lambda^2}$$

Photon energy:

λ = 500 nm **$1/\lambda$ = 20000 cm^{-1}**

Linewidth:

At 563 nm, **$\Delta\lambda$ =1 nm → $\Delta\nu$ = 1000 GHz**

Spectral line profiles

Homogeneous linewidth: For a given transition it is identical for all atoms:

Its shape is a Lorentzian

$$\phi_L(\nu-\nu_0)=\frac{1}{2\pi}\frac{\delta\nu_L}{(\nu-\nu_0)^2+(\delta\nu_L/2)^2}$$

Natural linewidth:

$$\delta\nu_n(FWHM)=\frac{1}{2\pi\tau}$$

Power broadening:

$$\delta\nu_S=\delta\nu_n\sqrt{1+S_0}$$

S_0 at the line center ν_0

Inhomogeneous linewidth: results from collective effects, for example thermal motion of atoms (**Doppler broadening**):

Its shape is a Gaussian

$$\phi_D(\nu-\nu_0)=\frac{2\sqrt{\ln(2)/\pi}}{\delta\nu_D}\exp-4\ln(2)\frac{(\nu-\nu_0)^2}{(\delta\nu_D)^2}$$

Doppler linewidth:

$$\delta\nu_D(GHz)=(2\nu_0/c)\sqrt{2\ln 2(RT/M)}=7.16\cdot10^{-16}\nu_0\sqrt{T/M}$$

For sodium 589.1 nm line (τ=16 ns) at 500 K:

$\delta\nu_n$=0.01 GHz

$\delta\nu_D$=1.7 GHz

Different type of Lasers

Frequency fixed: (often used as pump laser)

- Ar^+, Kr^+, Nd-Yag, Excimer (XeCl), Cu, HeNe ….. $\Delta\nu_L \cong$ 10 GHz

Tunable lasers:

- **Pumped by a laser: Dye, Ti:Sa, OPO (tuning range 10 to 100 nm)**

  Lasers available from 400 nm to µm (+ frequency doubling)

  - **Pulsed lasers: P up to 10 mJ, $\Delta t \approx$ 3 to 30 ns, $\Delta\nu_L \geq$ 1 GHz**

    Convenient for frequency doubling and n photon transitions

  - **CW lasers: P= up to a few W, $\Delta\nu_L \approx$ 0.001 GHz**

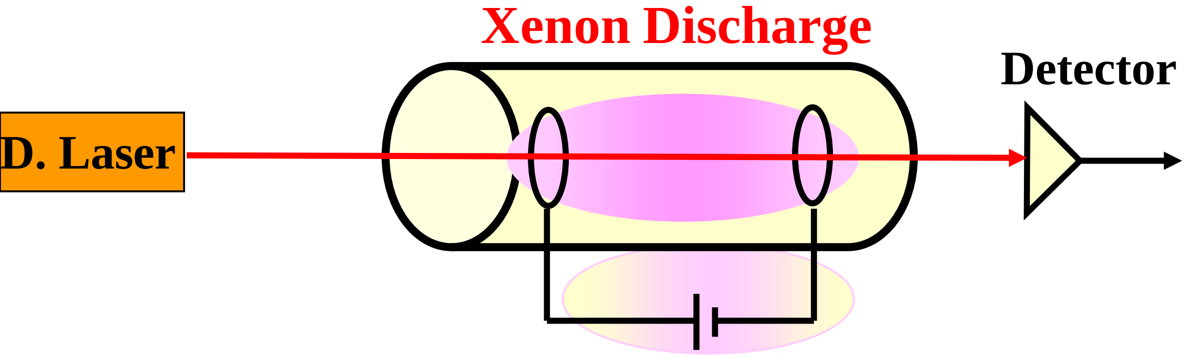
    Convenient for high resolution spectroscopy

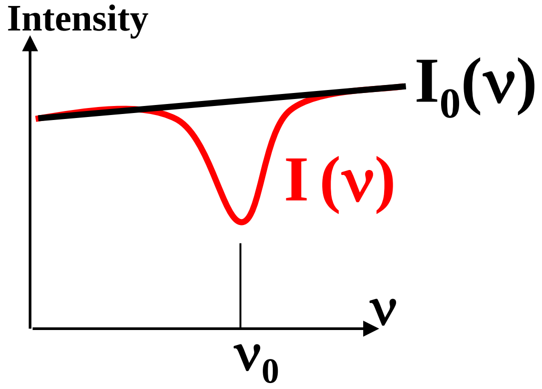
- **Diode laser: lasers available from 400 nm to 10 µm with**

  - **tuning range up to 10 nm**
  - **P= up to a few 10 mW, $\Delta\nu_L \approx$ 0.001 GHz (if single mode)**

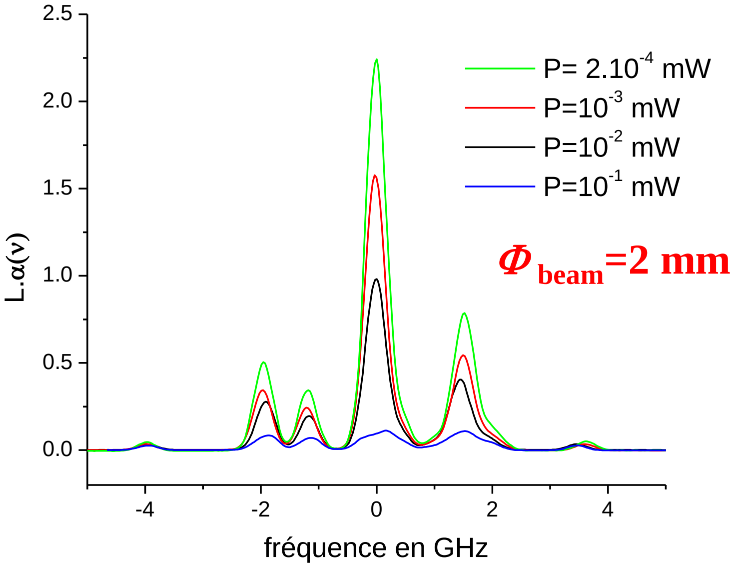
  They are more compact, easier to run and cheaper

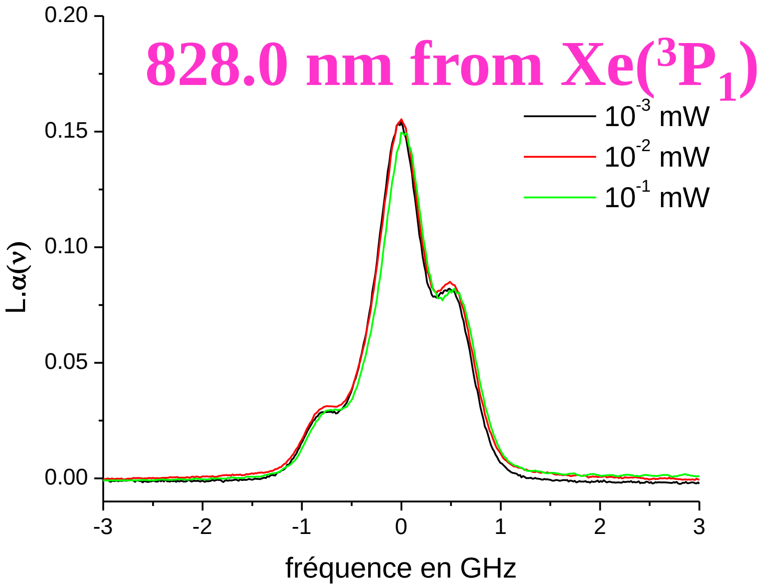
Influence of laser power on line profile and on measured absorption rate





823.1 nm from Xe($^3P_2$)





- *** Complex line structures result from Isotope Shifts and Hyperfine Structure.**
- *** Power saturation on 823.1 nm line because Xe*($^3P_2$) depletion.**
- *** No saturation on 828 nm line because Xe*($^3P_1$) lifetime ≅ 10 ns**

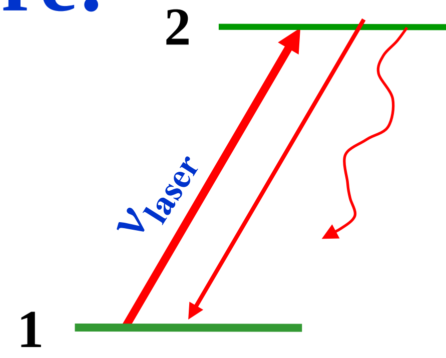
Origin of optical saturation

$$\alpha(\nu) = \frac{4hB_{12}}{\lambda\gamma}\left(n_1(\nu) - \frac{g_1}{g_2} n_2(\nu) \right)$$

α becomes no more proportional to n_1

- **1- Laser beam transfers a significant number of atoms from the lower to the upper state and n_2 becomes no more negligible compared to n_1. (short pulse lasers)**
- **2- Atoms in the upper state are lost by radiation, or collisional transfers, to a 3rd state and atoms in the lower state are not renewed fast enough: the lower state becomes depleted. (cw lasers)**

Rate equations governing the population densities N_1 and N_2 of states $|1\rangle$ and $|2\rangle$ are:



$$dN_1/dt=(B_{21}\rho+A_{21})N_2-(B_{12}\rho+1/\tau_1+\sum_q k_{1,q}M_q)N_1+C_1$$

$$dN_2/dt=B_{12}\rho N_1-\left(B_{21}\rho+A_{21}+A_{23}+\sum_q k_{2,q}M_q\right)N_2+C_2$$

$$B_{21}=\frac{g_1}{g_2}B_{12}=\frac{\lambda_0^3}{8h\pi}A_{21}$$ is the Einstein coefficient for stimulated emission

we assume $g_1 = g_2$, ρ is the energy density of the beam,

C_i accounts for the repopulation of state $|i\rangle$ from different paths, including diffusion transport into the laser volume and radiative cascades

$$\Re_1=1/\tau_1+\sum_q k_{1,q}N_q$$ and $$\Re_2=\sum_{i=lower}A_{2i}+\sum_q k_{2,q}N_q$$

are the total relaxation rates of the states in steady state, ($dN_i/dt=0$) the density difference of states |1⟩ and |2⟩ is:

$$\Delta N = N_1 - gN_2 = \Delta N^0 \Big/ \left(1 + S\frac{\Re_2 - A_{21} + g\Re_1}{\Re_1 + \Re_2}\right)$$

Where $\Delta N^0 = N_1^0 - gN_2^0 = \frac{C_1}{\Re_1} - \frac{C_2}{\Re_2}(g - A_{21}/\Re_1)$ is in the absence of laser beam ($\rho=0$),

and

$$S = B_{12}\rho/\Re^*$$

is **The saturation parameter**

related to the **mean relaxation rate**

$$\Re^* = \Re_1\Re_2/(\Re_1 + \Re_2)$$

:

The resulting population density in the lower state is:

$$N_1 = \frac{C_1(gS + R_2/\Re^*) + C_2(gS + A_{21}/\Re^*)}{S[R_2 - A_{21} + gR_1] + (R_1 + R_2)}$$

When $C_2 \searrow 0$

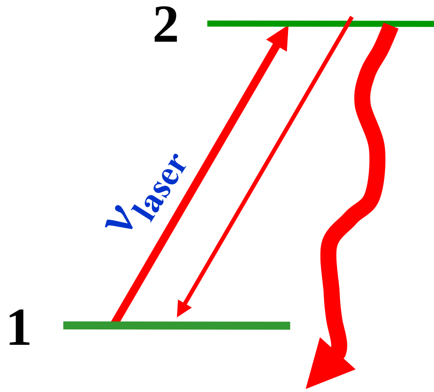
$$N_1 = \frac{C_1(gS + R_2/\Re^*)}{S[R_2 - A_{21} + gR_1] + (R_1 + R_2)}$$

Larger S is, lower the measured population will be

When $\rho \to 0$, $N_1 = \frac{C_1}{\Re_1}$ For $\rho \to \infty$ $N_1 = \frac{g(C_1 + C_2)}{\Re_2 - A_{21} + g\Re_2}$

Estimation of saturation parameter S

For a cw laser:



λ= 590 nm ;

τ_2=16 ns ; **hence** **$R_2=1/\tau_2=6.25\ 10^7\ s^{-1}$**

But **$A_{21}=6\ 10^6\ s^{-1}$;**

R_1=(1/transit time inside a beam of ϕ=2 mm)=(0.5 km.s^{-1})/(2 mm)=**$2.5\ 10^5\ s^{-1}$**

Laser power: P=1 mW; **$\Delta\nu_L$= 1 MHz <<1/($2\pi\tau_2$) ;**

Laser beam diameter =2 mm

We can calculate

$$S = \frac{B_{12} * \rho}{R_1} = 20$$

Hence the density measured by absorption will not be correct with so large S value

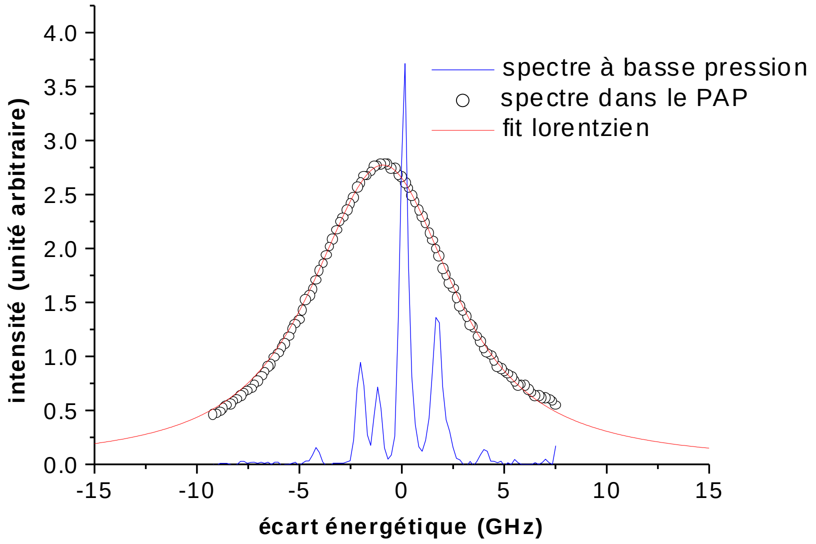
because $N_1=N_1^0/(1+S)$ **However,**

as $\delta\nu_S = \delta\nu * \sqrt{1+S} = 4.5 * \delta\nu = 45$ **MHz** **is much smaller than the Doppler width (1.7 GHz), the line profile can still provide the gas temperature**

Line Profiles Broadened by pressure (800 mBar)

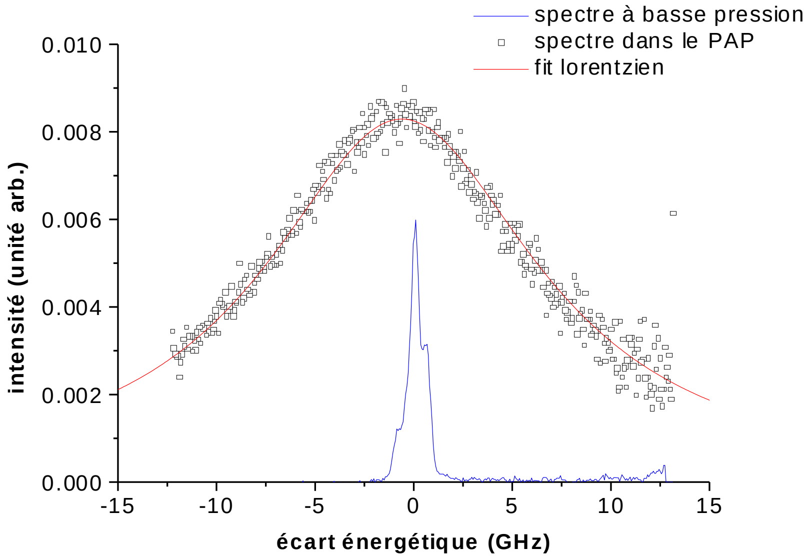
Xe 823nm line :

⇨Lorentzian profile with Δν=7,2 GHz, shifted by –1,1 GHz



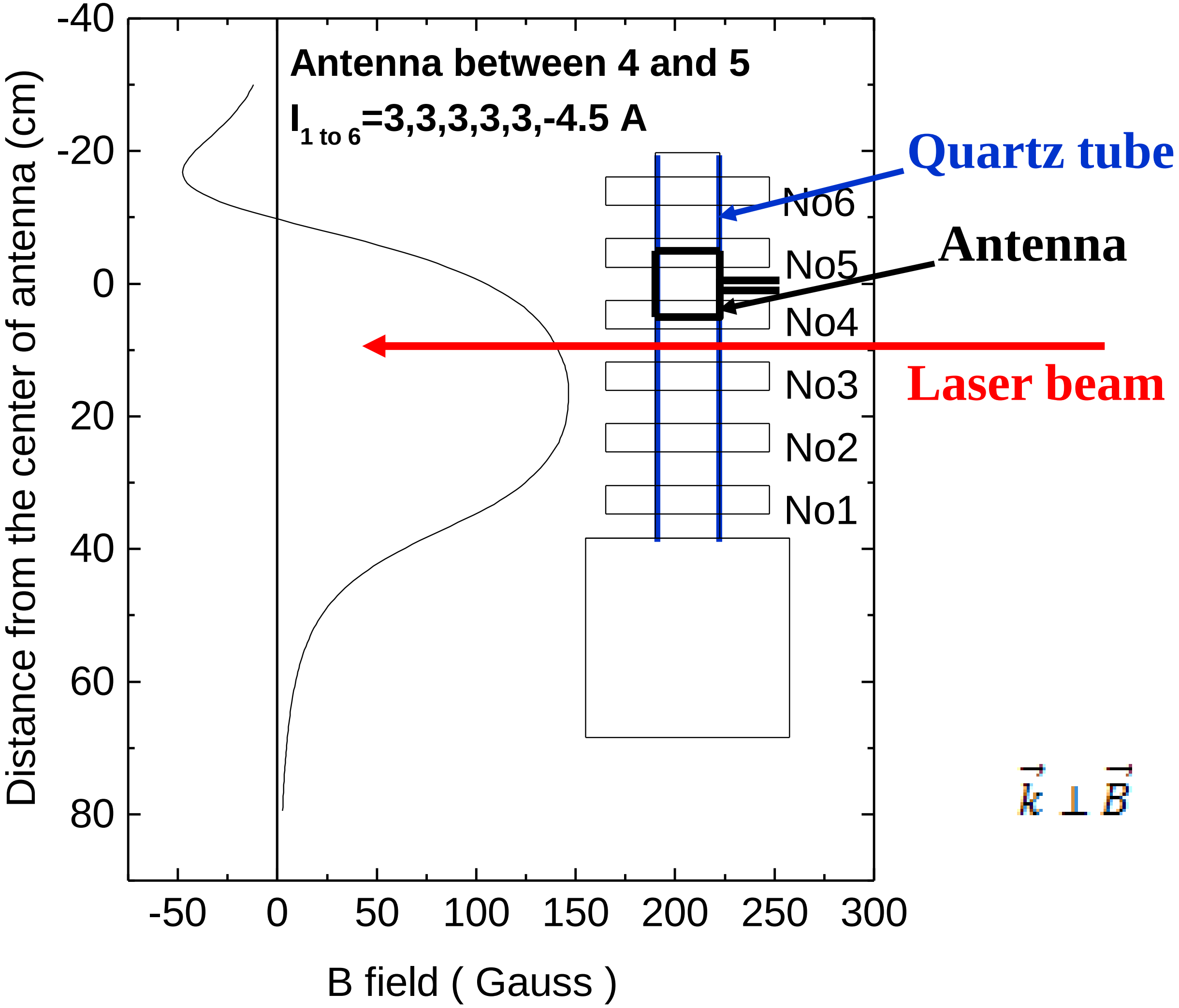
Xe 828 nm line :

⇨Lorentzian profile with Δν=16,8 GHz, shifted by –0,72 GHz



Line profile in presence of magnetic field

in a low power Helicon Argon plasma; p= 0.9 to 10 µbar

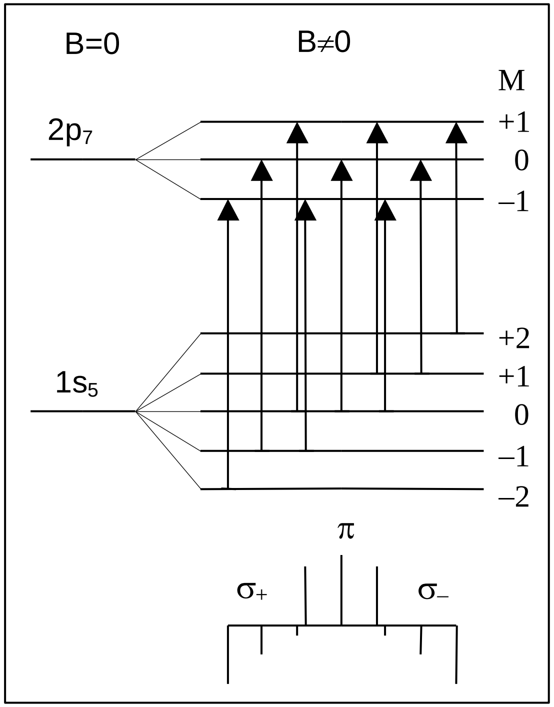


$\vec{k} \perp \vec{B}$

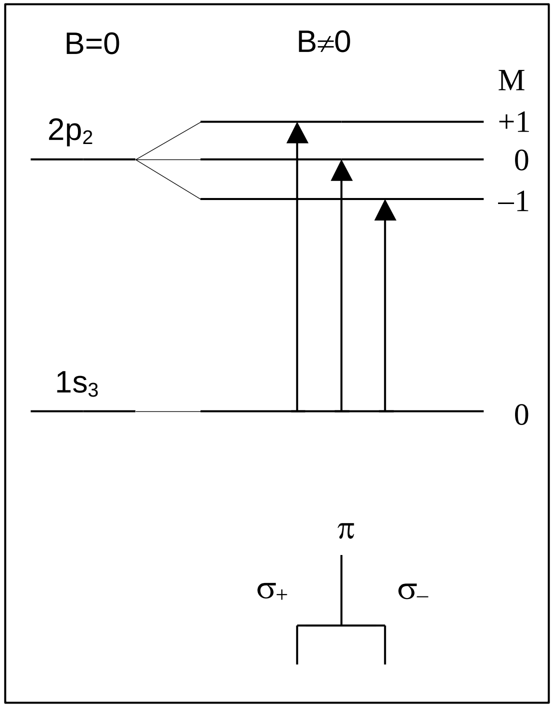
Line profile in presence of strong magnetic field

Zeeman components of the absorbing lines of Argon

722.38 nm; $2p_7 \leftarrow {}^3P_2$ line



722.42 nm; $2p_2 \leftarrow {}^3P_0$ line

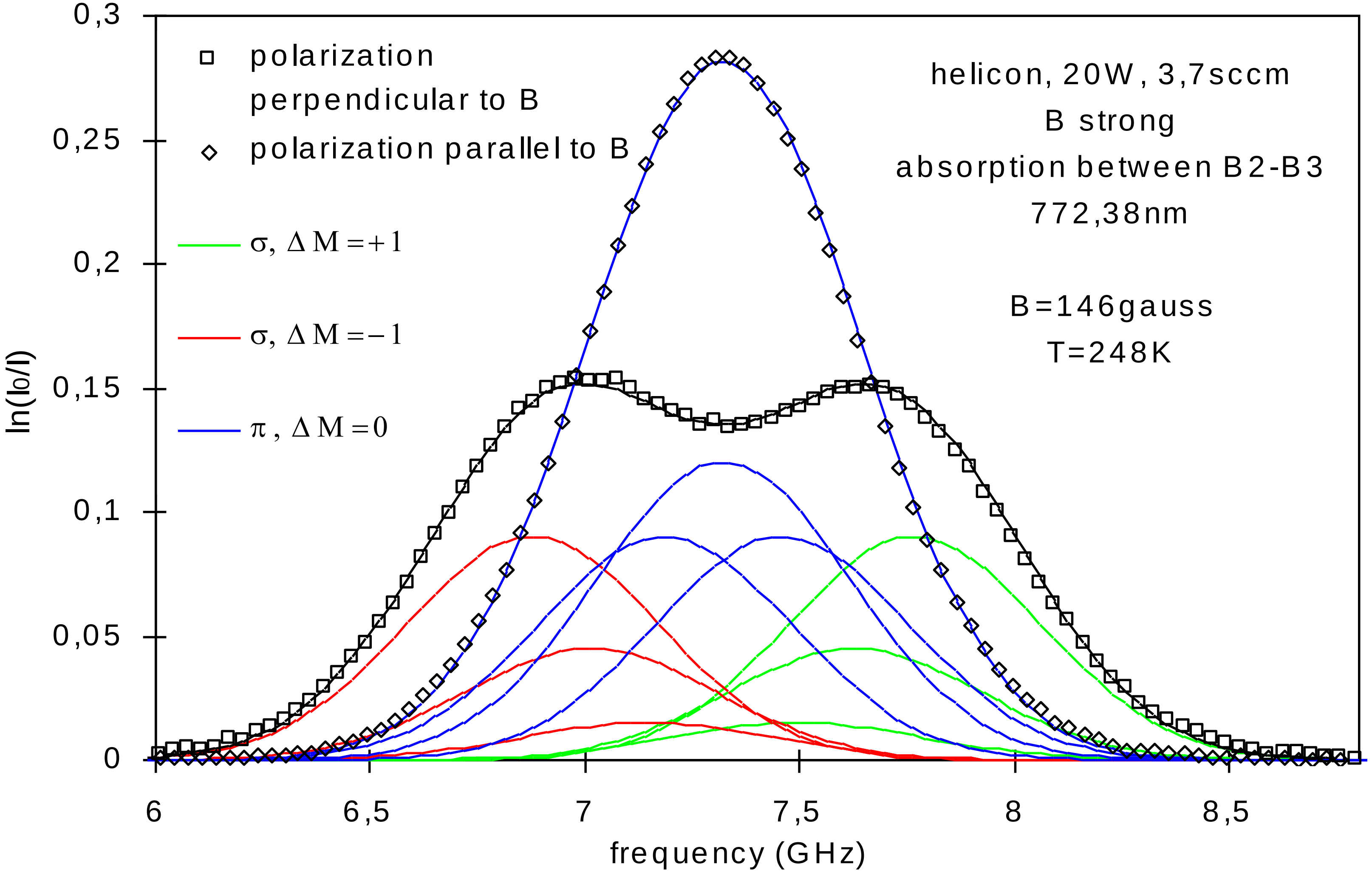


$\vec{k} \perp \vec{B}$ if $\vec{E} \perp \vec{B}$ only σ^+ and σ^- lines exist

$\vec{k} \perp \vec{B}$ if $\vec{E} // \vec{B}$ only π line (s) exist

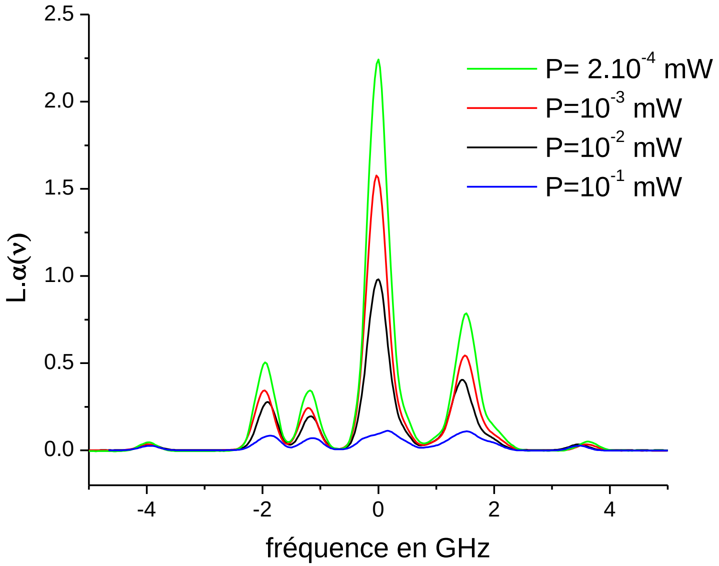
$\vec{k} // \vec{B}$ then only σ^+ and σ^- lines exist

Experimental profiles of the 722.38 nm line with two different polarization fitted with Gaussians



Concluding remarks concerning measurements by laser absorption techniques

823.1 nm from Xe($^3P_2$)



For density measurements

*** Laser intensity must be very low to avoid optical saturation:**

P $\leq$ 1 μW/ mm^2 for an strong atomic line.

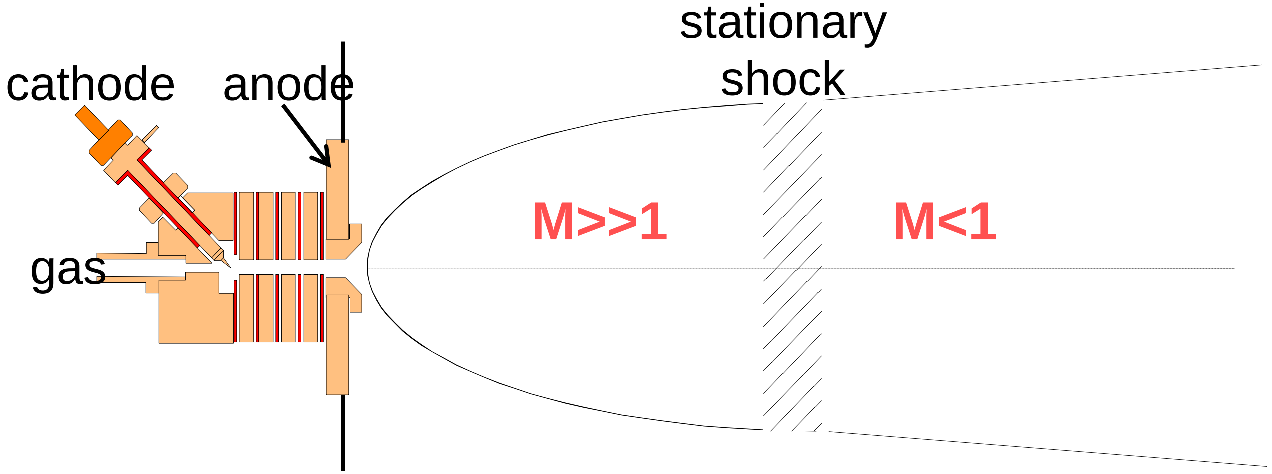
For CRDS, the laser power inside the cavity must be considered

For gas temperature measurements

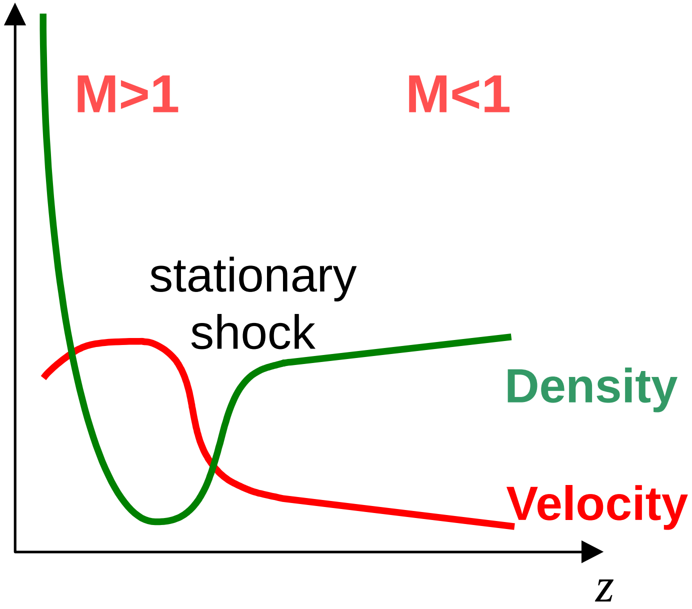
*** Atoms in interaction with the laser beam must be in collisional equilibrium with the gas bath and their mean free path much shorter than the vessel dimensions.**

Experimental determination of argon atoms *vdf* in an expanding arc jet by LIF (Eindhoven)

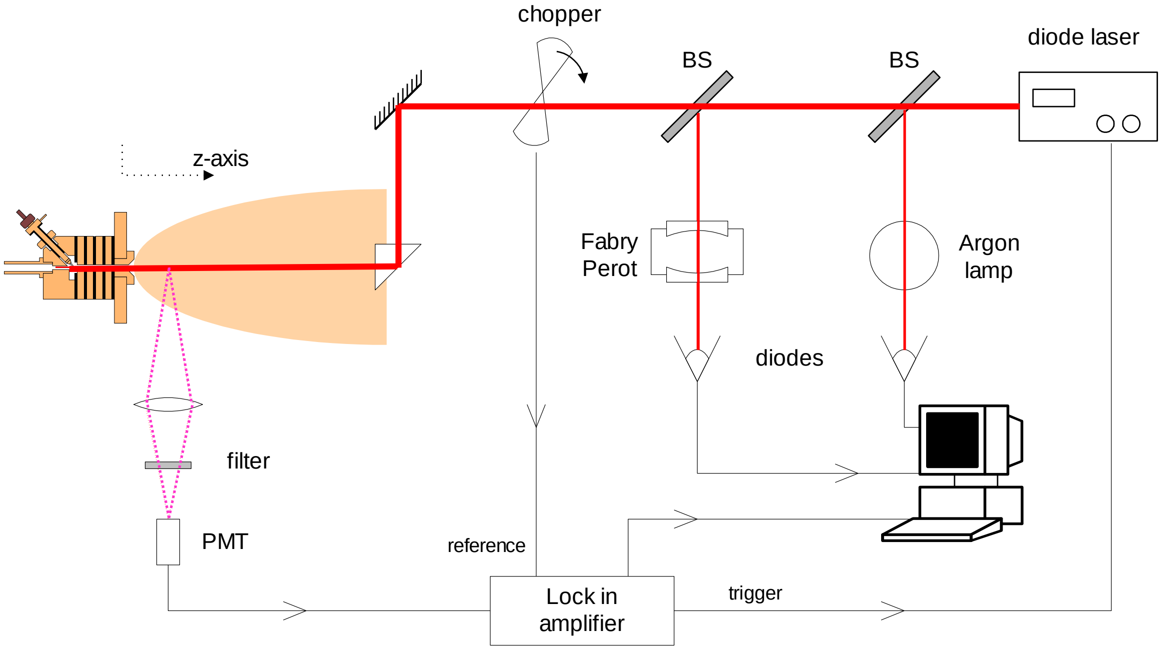
P = 5 kW
$T_e = 1$ eV
$n_e = 10^{22}\ \mathrm{m}^{-3}$
$p_{source} = 5.10^4$ Pa
$p_{bg} = 10\ldots100$ Pa

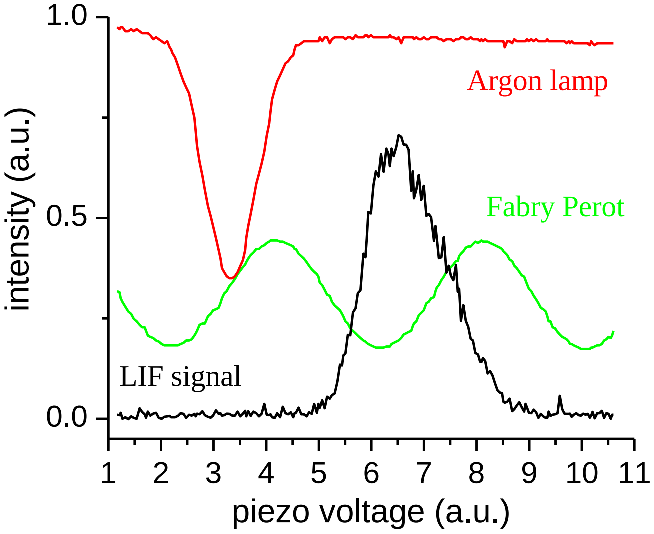


Expected density and velocity distribution along the jet axis



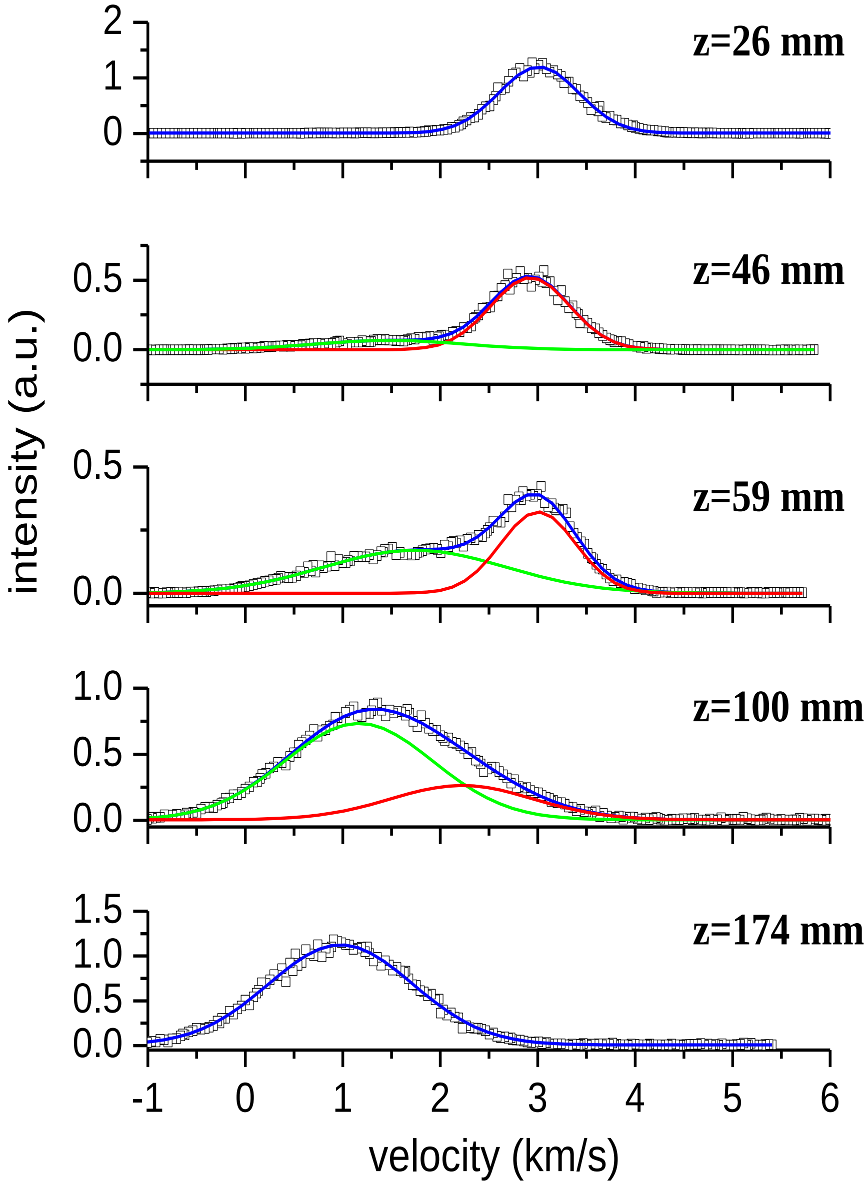
Laser Doppler-Shift setup



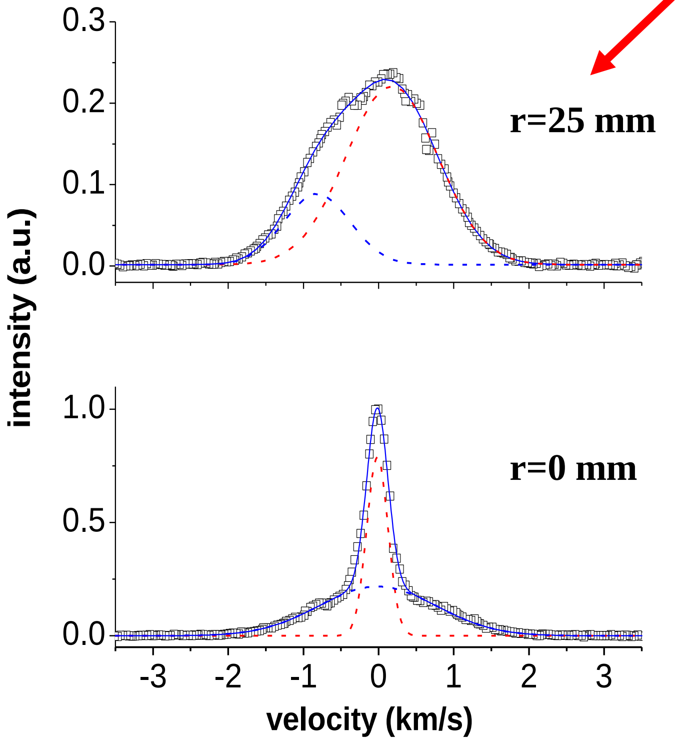


Ar* velocity distribution functions

(axial *v* at axis center)



(radial *v* at z = 50 mm)



Equipement commun du Réseau Plasmas Froids

"Système Laser à Diode"

acquis sur les crédits MRCT – CORTECH

Total des crédits reçus (2004-2007): 49551 €

- Responsables : Nader Sadeghi (LSP, Grenoble), dépositaire

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- Matériel: lasers avec cavité externe du type Littrow et du type DFB fournissant qqs 10 mW continus dans une largeur spectrale d'environ 10 MHz ($\cong 10^{-5}$ nm)

- Fournisseur: TOPTICA, représenté par OLI

DL avec cavité externe acquis par RPF

Ces DL avec cavité externe (Littrow) sont balayables sur environ une vingtaine de GHz sans saut de mode (ssm). L’accord en longueur d’onde sur la gamme de fonctionnement est obtenu en changeant la température de la DL.

- **1 Électronique de commande (Sys DL 100/19), avec: contrôle de courant, contrôle de température et tiroir de modulation (géné de fréquence).**
- **5 Têtes laser:**

396 – 399 nm;	**10 mW;**	**20 GHz (ssm)**	**Ti, Al**
402 – 407 nm	**10 mW**	**20 GHz (ssm)**	**Ga**
652 – 662 nm;	**30 mW;**	**20 GHz (ssm)**	**H***
750 – 791 nm;	**30 mW;**	**30 GHz (ssm)**	**Ar*, O***
1059 – 1090 nm;	**30 mW;**	**15 GHz (ssm)**	**He*, N_2***

DL du type DFB acquis par RPF

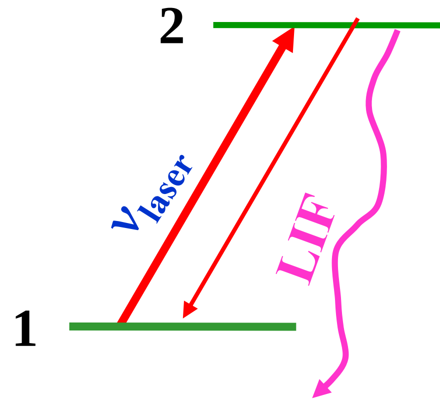
Ces DL sont sans cavité externe mais ont un réseau de Brag intégré. Leur domaine d’accordabilité n’est que d’environ 1 nm. Le balayage en fréquence peut être obtenu par le courant (environ 20 GHz ssm) ou par la température (très lent <Hz mais sur toute la gamme: environ 1 nm ≅ 1000 GHz ssm). La stabilité en fréquence n’est que ≅ 200 MHz sur quelques minutes (≅20-50 MHz pour DL Littrow) mais convient pour raies élargies à haute pression.

- **1 Électronique de commande (Sys DL- DFB 100/19), avec: contrôle de courant, contrôle de température et tiroir de modulation (géné de fréquence).**
- **2 Têtes laser:**
  - **772 – 773 nm; 70 mW; 20 GHz (ssm) Ar***
  - **1081 – 1083 nm; 70 mW; 20 GHz (ssm) He***

Matériels d’accompagnement:

- **1 Barreau en verre de 10 cm avec faces parallèles polies pour servir d'étalon Fabry-Perot ayant un intervalle spectral libre de l'ordre de 1 GHz**
- **1 Lambdamètre devant permettre la mesure de longueur d'onde avec une précision de 3 pm.** L'injection par une fibre optique monomode pose toujours quelques problèmes.
- **3 photodiodes; sensibilité≅ 1V/µW; bande passante 15 kH**

Intensity of Laser Induced Fluorescence signal



LIF signal is proportional to N_2 density given by:

$$I_{23} \propto N_2 A_{23} \propto N_1(El,V,R) A_{23} \frac{B_{12}\rho}{B_{21}\rho + \Re_2}$$

When LIF is used to determine the relative population of two different especies, m and n:

At low laser power limit, the LIF signal ratio is:

$$\frac{I_{23}(m \rightarrow 3)}{I_{23}(n \rightarrow 3)} = \frac{N_1(i)}{N_1(j)} \frac{B_{im}}{B_{jn}} \frac{A_{m3}}{A_{n3}} \frac{\tau_m}{\tau_n}$$

τ could be p and T dependent

At high laser power limit, the LIF signal ratio is:

$$\frac{I_{23}(m \rightarrow 3)}{I_{23}(n \rightarrow 3)} = \frac{N_1(i)}{N_1(j)} \left(\frac{g_i}{g_m}\right) \left(\frac{g_n}{g_j}\right) \frac{A_{m3}}{A_{n3}}$$