atelier

Plasma laser : diagnostic et modélisation

seconde partie

Plasmas en déséquilibre

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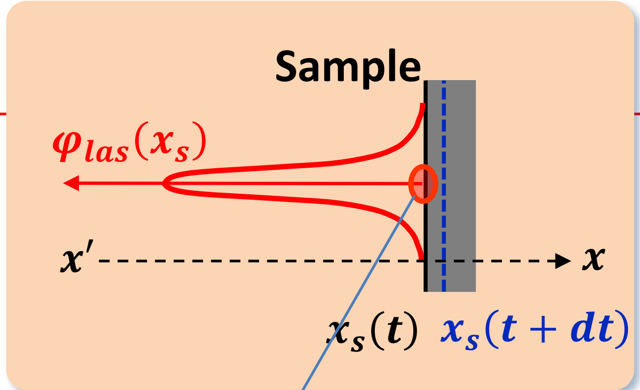
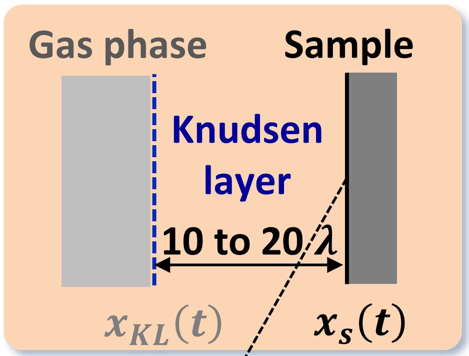
Non-equilibrium on the surface…

$T_s < 0.9\,T_c$



$$v_{rec} = \frac{dx_s}{dt}$$

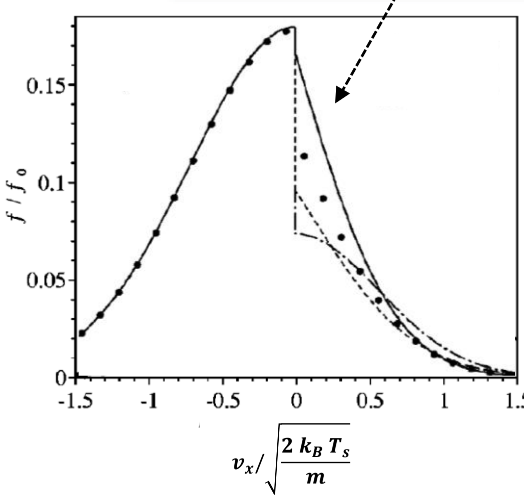
Heating within the material: due to e- at T_e
Characteristic time for e-phonons equilibrium $\sim 10\ ps$
$\Rightarrow$ e-heavies equilibrium within the material



Classical heating driven by the generalized Fourier equation

$$\frac{\partial(\rho h)}{\partial t} + v_{rec}\frac{\partial(\rho h)}{\partial x} = \frac{\partial}{\partial x}\left[k\frac{\partial T_s}{\partial x}\right] - \frac{\partial \varphi_{las}}{\partial x}$$

with φ_{las} depending on the local laser conditions
and v_{rec} the recession speed



T_s higher than in the gas phase…

A.V. Gusarov *et al.*
Phys. Fluids **14** (2002) 4242

Non-equilibrium on the surface…

$T_s < 0.9\,T_c$

Relationship between *S* and *KL* conditions
→ Mach $\mathcal{M}_{KL}$

$\mathcal{M}_{KL}$	ρ_{KL}/ρ_s	T_{KL}/T_s	p_{KL}/p_s
0	1	1	1
0.05	0.927	0.980	0.908
0.1	0.861	0.960	0.827
0.2	0.748	0.922	0.690
0.4	0.576	0.851	0.490
0.6	0.457	0.785	0.358
0.8	0.371	0.725	0.269
1.0	0.308	0.669	0.206

Clausius-Clapeyron equation

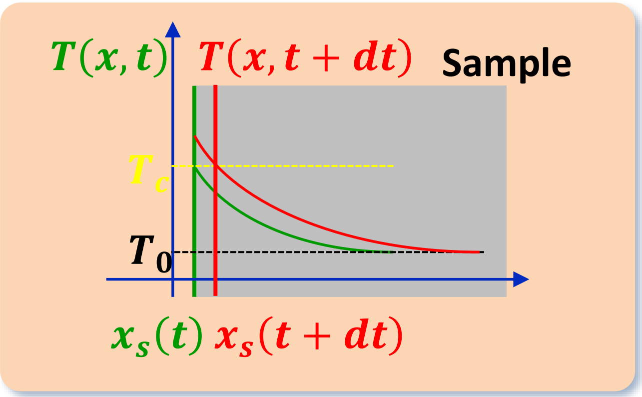
$$p_s(T_s) = p_{atm}\ exp\left[\frac{\Delta h_b\, m}{k_B}\left(\frac{1}{T_b} - \frac{1}{T_s}\right)\right]$$

$0.9\,T_c < T_s < T_c$

Formation of µ-bubbles within the liquid
→ Explosive boiling lasting more than the laser pulse

$T_s > T_c$?

Not phase change anymore → Supercritical fluid



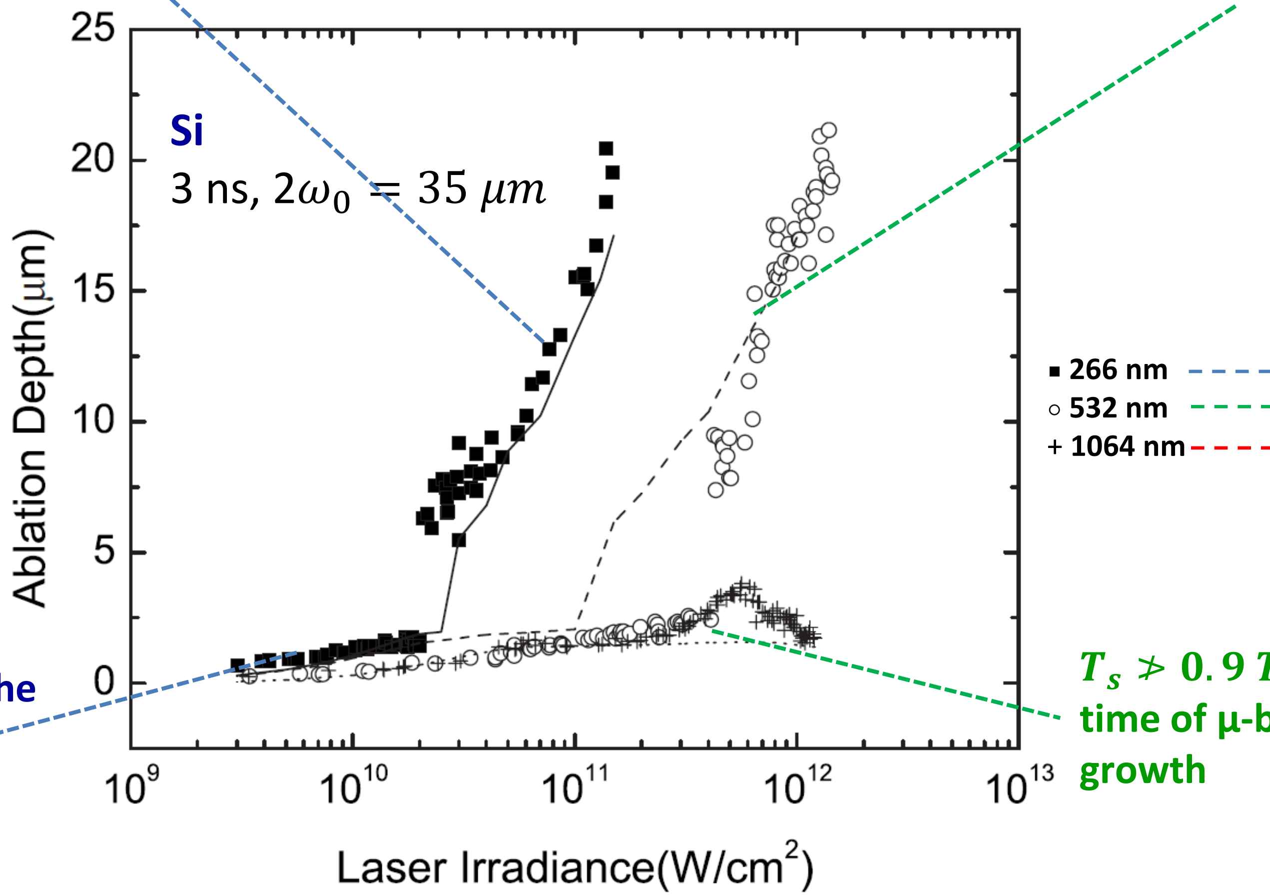
$$v_{rec} = \frac{dx_s}{dt}$$

Phase non-equilibrium

Non-equilibrium on the surface…

$T_s > 0.9\, T_c$ during the time of µ-bubbles growth

$T_s > 0.9\, T_c$ during the time of µ-bubbles growth



$T_s \ngtr 0.9\, T_c$ during the time of µ-bubbles growth

$T_s \ngtr 0.9\, T_c$ during the time of µ-bubbles growth

Q. Lu *et al.* J. Appl. Phys. **104** (2008) 083301

Non-equilibrium in the gas…

The ECHREM* code

*Eulerian CHemically REactive Multi-component plasma code

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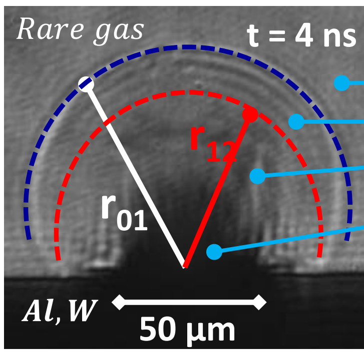
Assumptions



Hypersonic hemispherical expansion

- **(0) External gas** (rare gas: Ne, Ar, Kr or Xe)
- **(1) Shock layer** (shocked rare gas)
- **(2) Central plasma** (ablated W ou Al)

Ablated material

r_{01}	shock front radius
r_{12}	contact surface radius
v_{sf}	shock front speed

Bi-layer model
Propagation of the shockwave
Rankine-Hugoniot assumption

Atoms and ions… at T_A
Electrons… at T_e

Collisionnal-radiative source term

Balance equations

(1) Shock layer

Mass $\rho_0 v_{sf} = \rho_1 [v_{sf} - u_1(r_{01})] \Leftrightarrow \frac{d\rho(\{Rg\}_j^{Z+})}{dt} = \dot{\boldsymbol{\rho}}(\{\boldsymbol{Rg}\}_j^{Z+}) - \frac{\rho(Rg_j^{Z+})}{\rho_1}\frac{d\rho_1}{dt}$

Energy $\epsilon_0 + \frac{p_0}{\rho_0} + \frac{v_{sf}^2}{2} = \epsilon_1 + \frac{p_1}{\rho_1} + \frac{[v_{sf} - u_1(r_{01})]^2}{2}$

Momentum $p_0 + \rho_0 v_{sf}^2 = p_1 + \rho_1 [v_{sf} - u_1(r_{01})]^2$

(2) Central plasma

Mass $M_2 = \frac{2\pi}{3}\rho_2 r_{12}^3 \Leftrightarrow \frac{d\rho(\{Al,W\}_j^{Z+})}{dt} = \dot{\boldsymbol{\rho}}(\{\boldsymbol{Al,W}\}_j^{Z+}) - 3\rho(\{Al,W\}_j^{Z+})\frac{u_2(r_{12})}{r_{12}}$

Energy $E_2 = M_2(\epsilon^{Al,W} + \epsilon_2) + E_{c,2} \Leftrightarrow \frac{dE}{dt} = \rho_0 \epsilon_0 v_{sf} 2\pi r_{01}^2 - \frac{M_2}{\rho_2}(4\pi\varepsilon_{RR} + 4\pi\varepsilon_{TB} + \varepsilon'_{SE})$

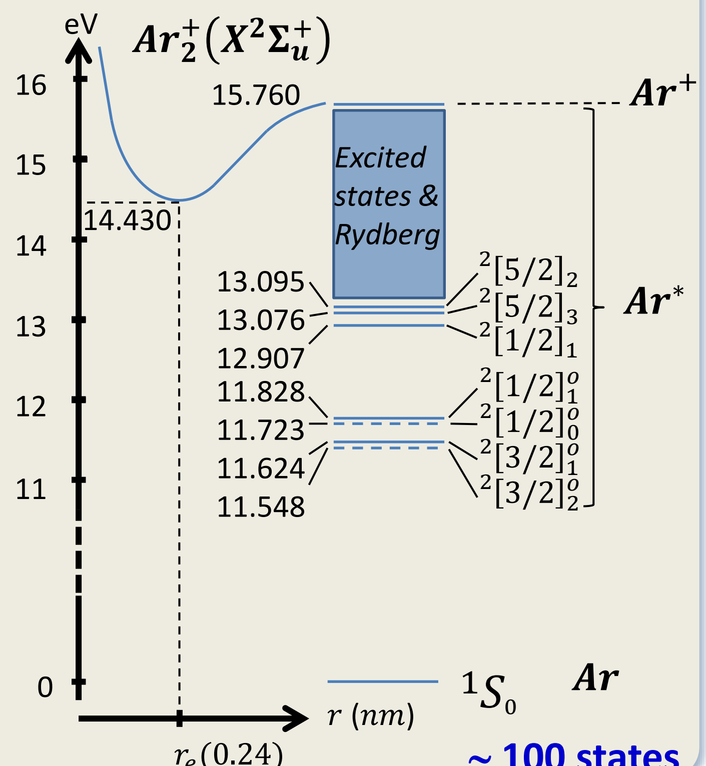
Momentum $\frac{d[u_2(r_{12})]}{dt} = \frac{8\pi}{3}\frac{r_{12}^2}{M_2}(p_2 - p_1)$

V. Morel, A. Bultel et al.
Spectrochim. Acta Part B **103-104** (2015) 112-123

Non-equilibrium in the gas…

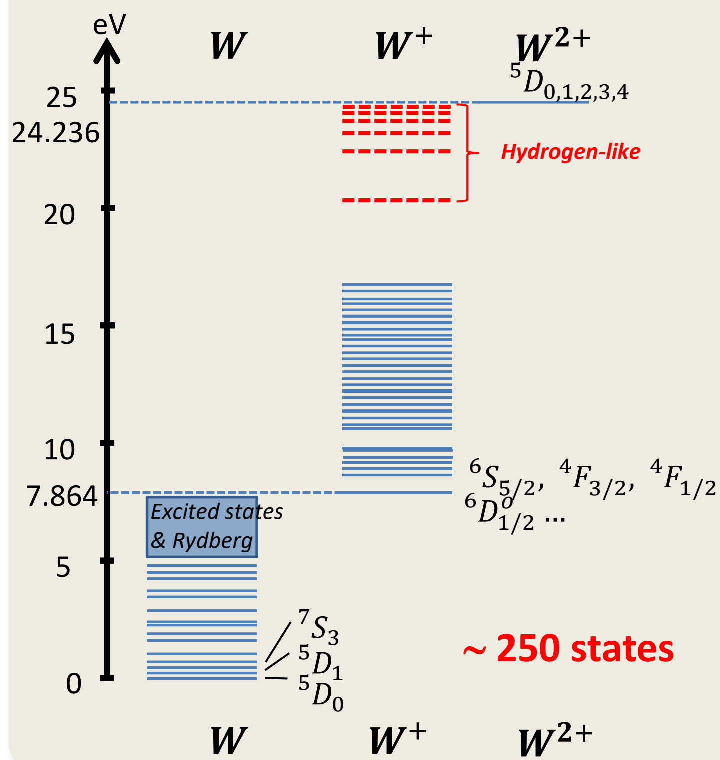
Ar-W…

Shock layer - *Argon*



~ 100 states

Central plasma - *Tungsten*



V. Morel, A. Bultel, I.F. Schneider, C. Grisolia
Spectrochim. Acta Part B **127** (2017) 7-19

GDR EMILI

Stand « données de base pour la physico-chimie des plasmas froids » !

Non-equilibrium in the gas…

The ECHREM* code

***E**ulerian **CH**emically **RE**active **M**ulti-component plasma code

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Shock layer - *Argon*

Collisional-Radiative model CoRaM-RG

$Ar + e^- \rightleftarrows Ar^* + e^-$	Exc. Elec. Impact
$Ar + Ar \rightleftarrows Ar^* + Ar$	Exc. Elec. Impact
$Ar + e^- \rightleftarrows Ar^+ + 2\,e^-$	Ioni. Elec. Impact
$Ar^* + e^- \rightleftarrows Ar^+ + 2\,e^-$	Ioni. Elec. Impact
$Ar + Ar \rightleftarrows Ar^+ + e^- + Ar$	Ioni. Heavy Impact
$Ar^* + Ar \rightleftarrows Ar^+ + e^- + Ar$	Ioni. Heavy Impact
$Ar^* + Ar^* \rightleftarrows Ar^+ + e^- + Ar$	Penning Ioni.
$Ar_2^+ + e^- \rightleftarrows Ar^* ou\ Ar + Ar$	Disso. Recomb.
$Ar^+ + e^- \rightarrow Ar^* ou\ Ar + h\nu$	Rad. Recomb.
$Ar_j \rightarrow Ar_{i<j} + h\nu$	Spont. Emiss.

30 000 elementary processes

Central plasma - *Tungsten*

Collisional-Radiative model CoRaM-W

$W_i + e^- \rightleftarrows W_{j>i} + e^-$

$W_i^+ + e^- \rightleftarrows W_{j>i}^+ + e^-$

$W_i + \Sigma_{i,Z} W_i^{Z+} \rightleftarrows W_{j>i} + \Sigma_{i,Z} W_i^{Z+}$

$W_i^+ + \Sigma_{i,Z} W_i^{Z+} \rightleftarrows W_{j>i}^+ + \Sigma_{i,Z} W_i^{Z+}$

$W_i + e^- \rightleftarrows W_j^+ + 2e^-$

$W_j^+ + e^- \rightleftarrows W^{2+} + 2e^-$

$W_i + \Sigma_{i,Z} W_i^{Z+} \rightleftarrows W_j^+ + e^- + \Sigma_{i,Z} W_i^{Z+}$

$W_j^+ + \Sigma_{i,Z} W_i^{Z+} \rightleftarrows W^{2+} + e^- + \Sigma_{i,Z} W_i^{Z+}$

$W^{2+} + e^- \rightleftarrows W_j^+ + h\nu$

$W_j^+ + e^- \rightleftarrows W_i + h\nu$

$W_j^+ \rightarrow W_{i<j}^+ + h\nu$

$W_j \rightarrow W_{i<j} + h\nu$

Thermal Bremsstrahlung

520 000 elementary processes

Collisional Database

$$k_i(T_{A,e}) = \sqrt{\frac{8\,k_B\,T_{A,e}}{\pi\,\mu}} \int_{x_0}^{+\infty} x\, e^{-x}\, \sigma_i(x)\, dx \text{ with}$$

- $\sigma_i(x)$ collisional cross section and
- $x = \frac{\varepsilon}{k_B\,T_{A,e}}$ reduced collision energy

Backward rate coefficient deduced from the ***Detailed Balance***

Radiative Database

NIST, Atomic Line List, ADAS, HULLAC…

V. Morel, A. Bultel, I.F. Schneider, C. Grisolia
Spectrochim. Acta Part B **127** (2017) 7-19

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Non-equilibrium in the gas...

The ECHREM* code

*Eulerian CHemically REactive Multi-component plasma code

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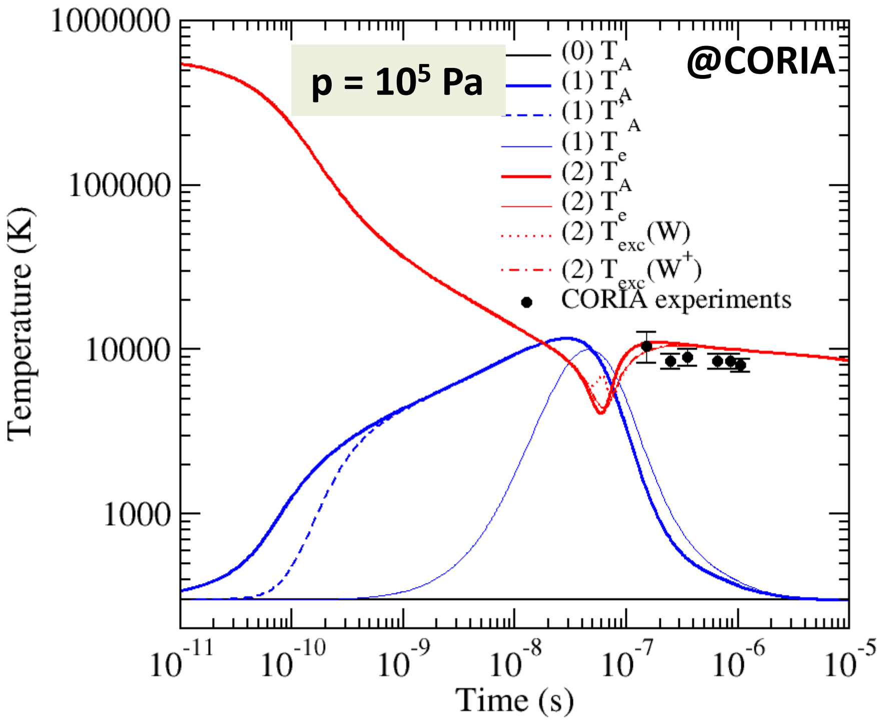

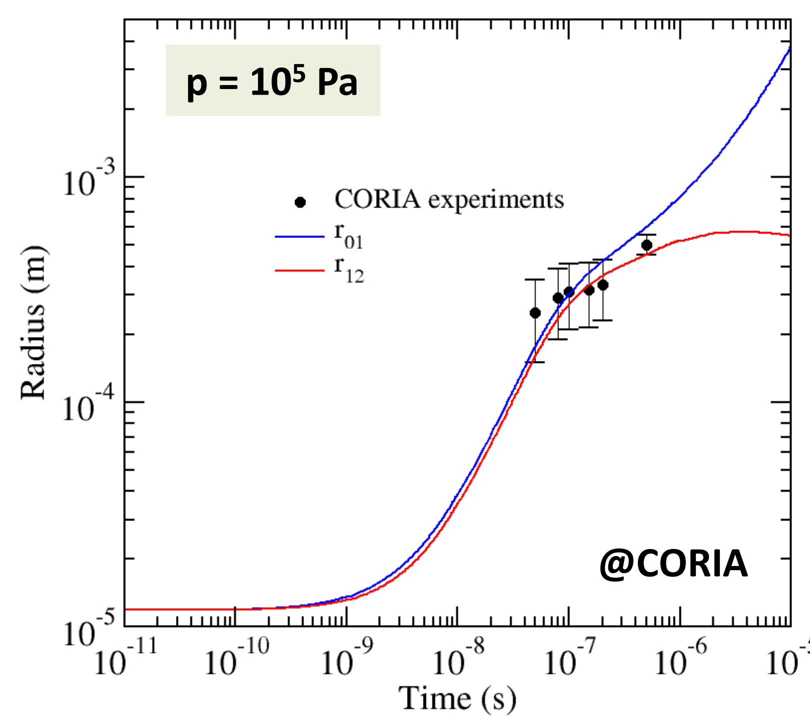

W (Ar) 10 ps 532 nm 10 J cm^{-2}

V. Morel, A. Bultel, I.F. Schneider, C. Grisolia
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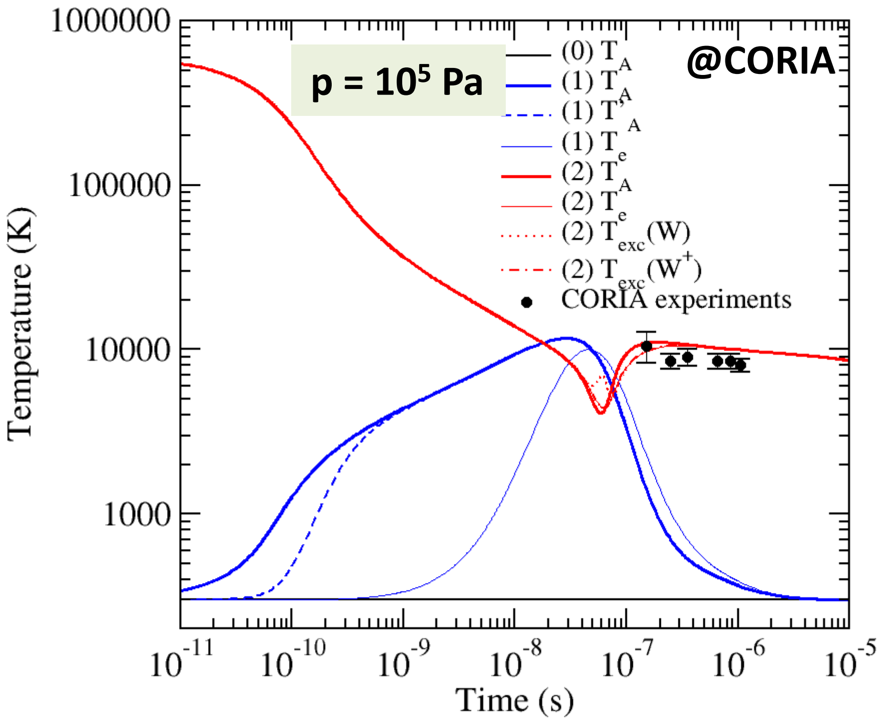
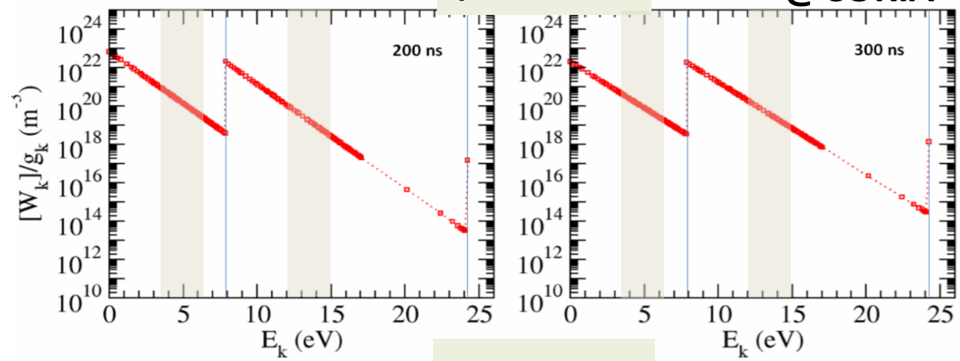


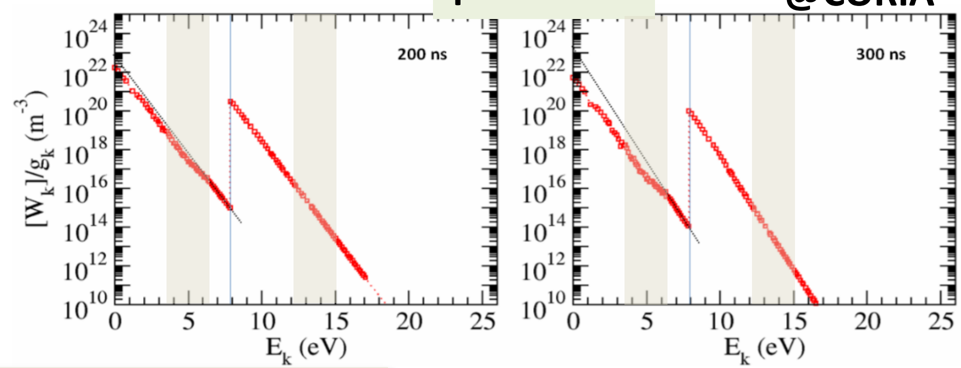
p = 10^5 Pa

@CORIA



p = 100 Pa

@CORIA



W (Ar) 10 ps 532 nm 10 J cm^{-2}

Excitation non-equilibrium

V. Morel, A. Bultel, I.F. Schneider, C. Grisolia
Spectrochim. Acta Part B **127** (2017) 7-19

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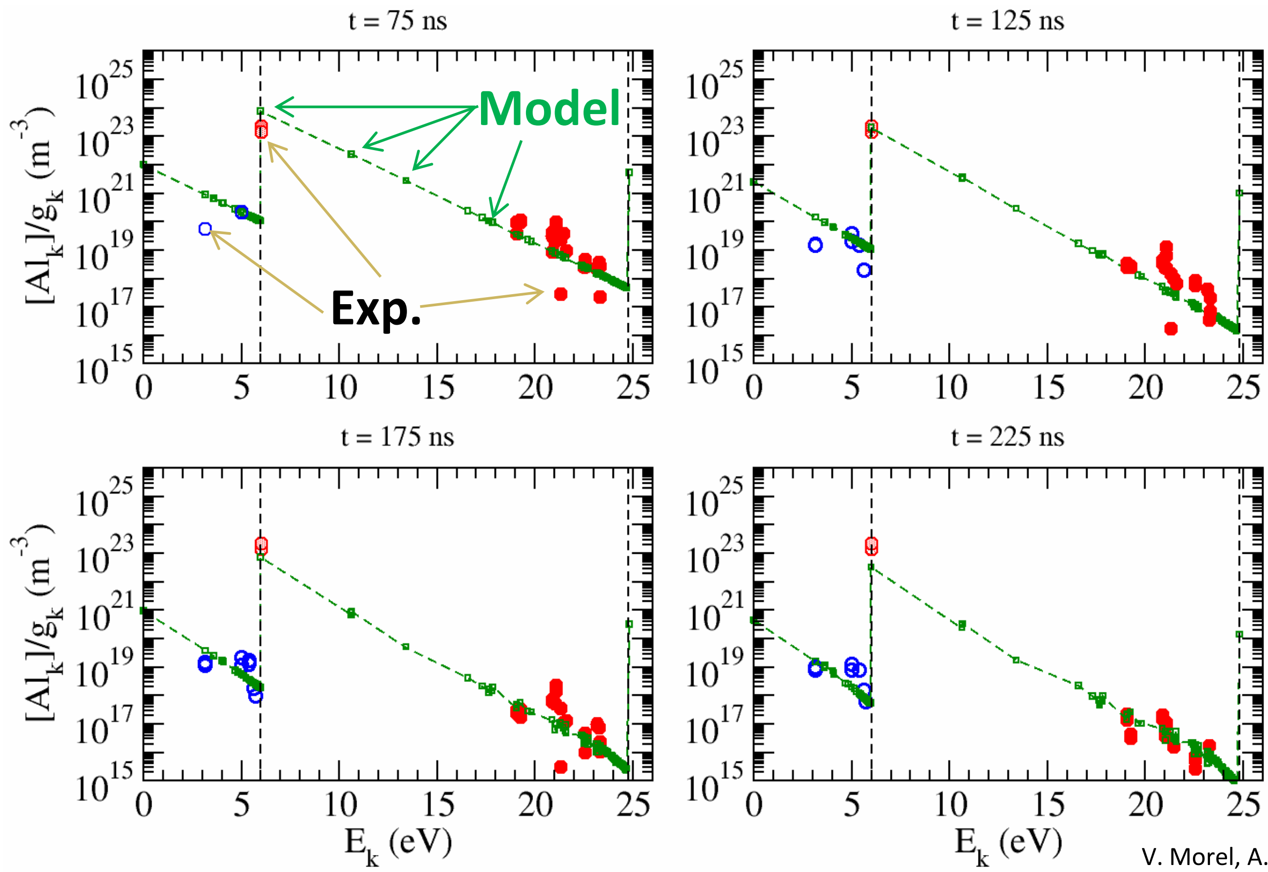
Non-equilibrium in the gas…

The ECHREM* code

*Eulerian CHemically REactive Multi-component plasma code



p = 100 Pa
@CORIA

Excitation non-equilibrium

V. Morel, A. Bultel, I.F. Schneider, C. Grisolia
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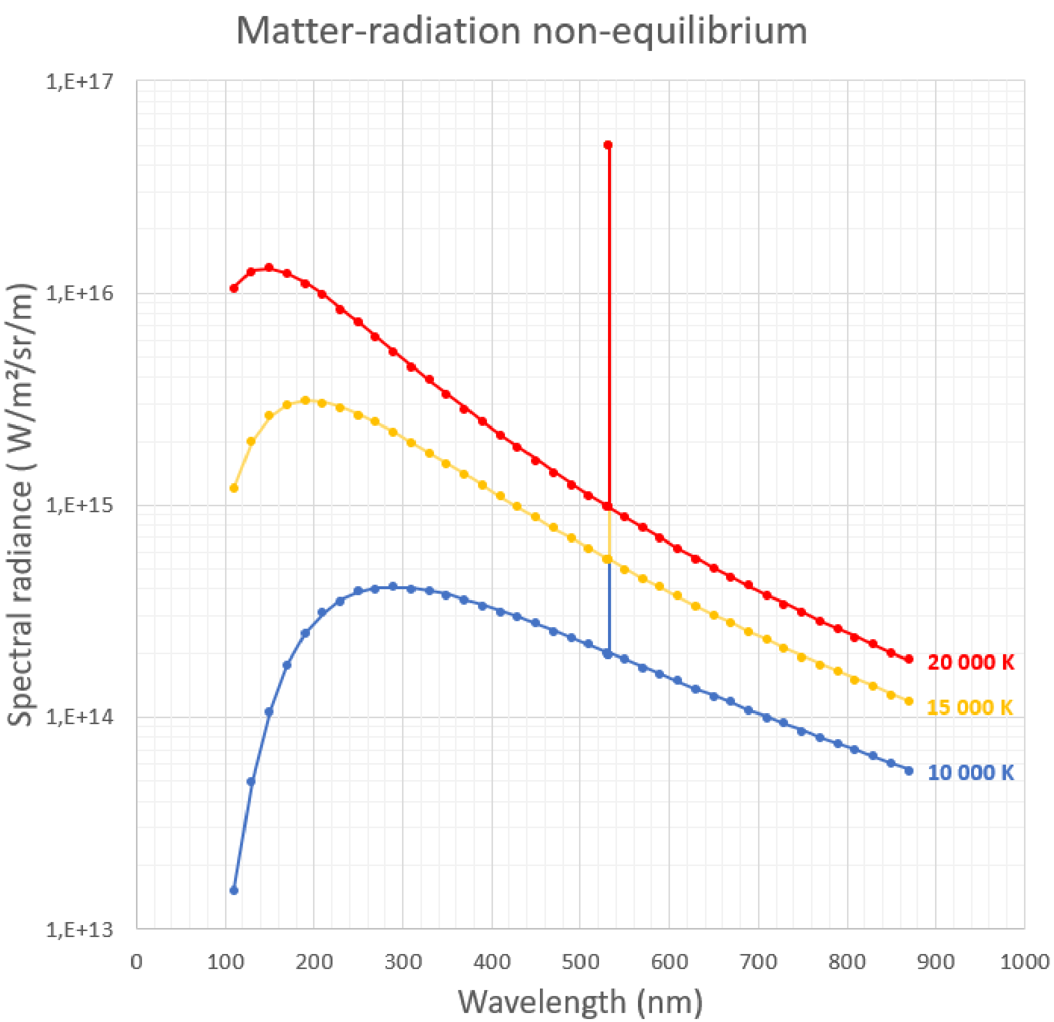
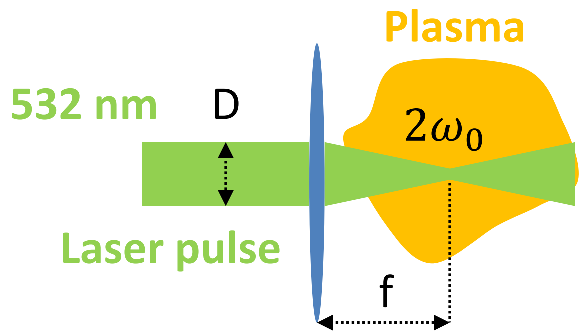
Al (N_2) 10 ps 532 nm 10 J cm^{-2}

Matter-radiation non-equilibrium…





$$L_{las} \sim \frac{E_p}{\tau_p} \frac{1}{\pi\omega_0^2} \frac{4f^2}{\pi D^2} \Delta\sigma$$

Parameter	Typical value
$E_p\ (mJ)$	10
$\tau_p\ (ns)$	5
$\omega_0\ (\mu m)$	100
$f\ (cm)$	10
$D\ (mm)$	4
$\Delta\sigma\ (cm^{-1})$	0.01
$L_{las}\ (W\ m^{-2}\ sr^{-1}\ m^{-1})$	5×10^{16}

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Matter-radiation non-equilibrium…

Radiative recombination

Radiative recombination	$A^+ + e^- \rightarrow A_i + h\nu$
Stimulated radiative recombination	$A^+ + e^- + nh\nu \rightarrow A_i + (n+1)h\nu$
Photo-ionization	$A^+ + e^- + nh\nu \leftarrow A_i + (n+1)h\nu$
Multi-photon ionization	$A^+ + e^- + nh\nu \leftarrow A_i + (n + \boldsymbol{m} \geq 2)h\nu$

Bremsstrahlung

Bremsstrahlung	$e^-\left(\frac{1}{2}m_e v_e^2\right) + (\boldsymbol{A}, \boldsymbol{A}^+) \rightarrow e^-\left(\frac{1}{2}m_e v_e'^2\right) + (\boldsymbol{A}, \boldsymbol{A}^+) + h\nu$
Stimulated Bremsstrahlung	$e^-\left(\frac{1}{2}m_e v_e^2\right) + (\boldsymbol{A}, \boldsymbol{A}^+) + nh\nu \rightarrow e^-\left(\frac{1}{2}m_e v_e'^2\right) + (\boldsymbol{A}, \boldsymbol{A}^+) + (n+1)h\nu$
Inverse Bremsstrahlung	$e^-\left(\frac{1}{2}m_e v_e^2\right) + (\boldsymbol{A}, \boldsymbol{A}^+) + nh\nu \leftarrow e^-\left(\frac{1}{2}m_e v_e'^2\right) + (\boldsymbol{A}, \boldsymbol{A}^+) + (n+1)h\nu$

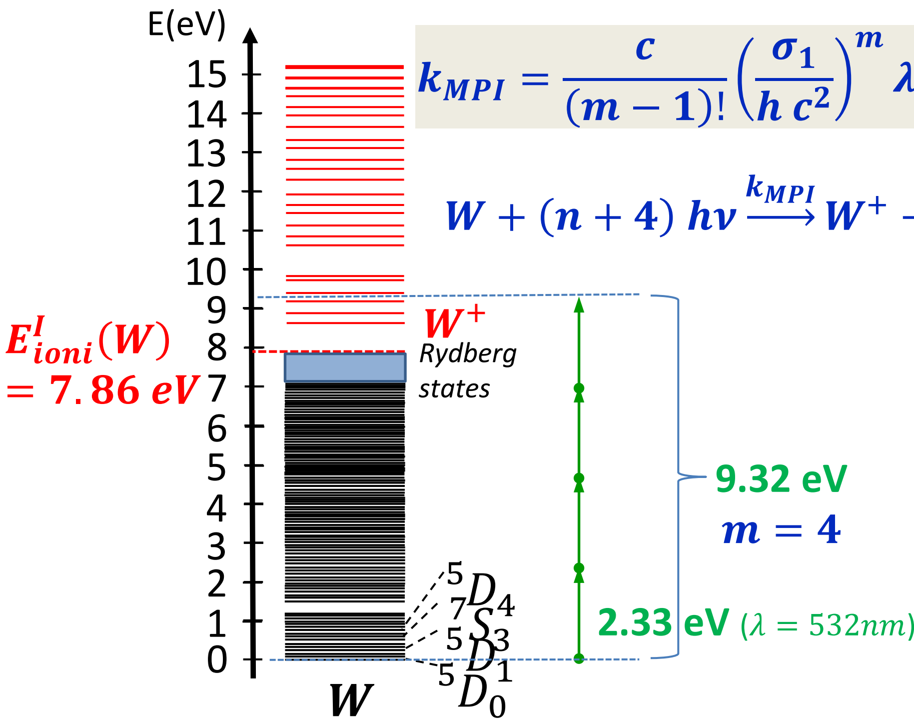
$\Rightarrow$ Absorption of the laser pulse by Inverse Bremsstrahlung and multi-photon ionization

Matter-radiation non-equilibrium…

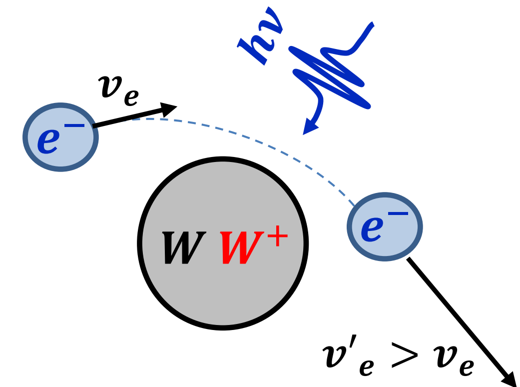
Multi-photon ionization

$$k_{MPI} = \frac{c}{(m-1)!}\left(\frac{\sigma_1}{h\,c^2}\right)^m \lambda^{2m-1}\,\varphi_{las}^m \quad (s^{-1})$$

$$W + (n+4)\,h\nu \xrightarrow{k_{MPI}} W^+ + e^- + nh\nu$$



Inverse Bremsstrahlung



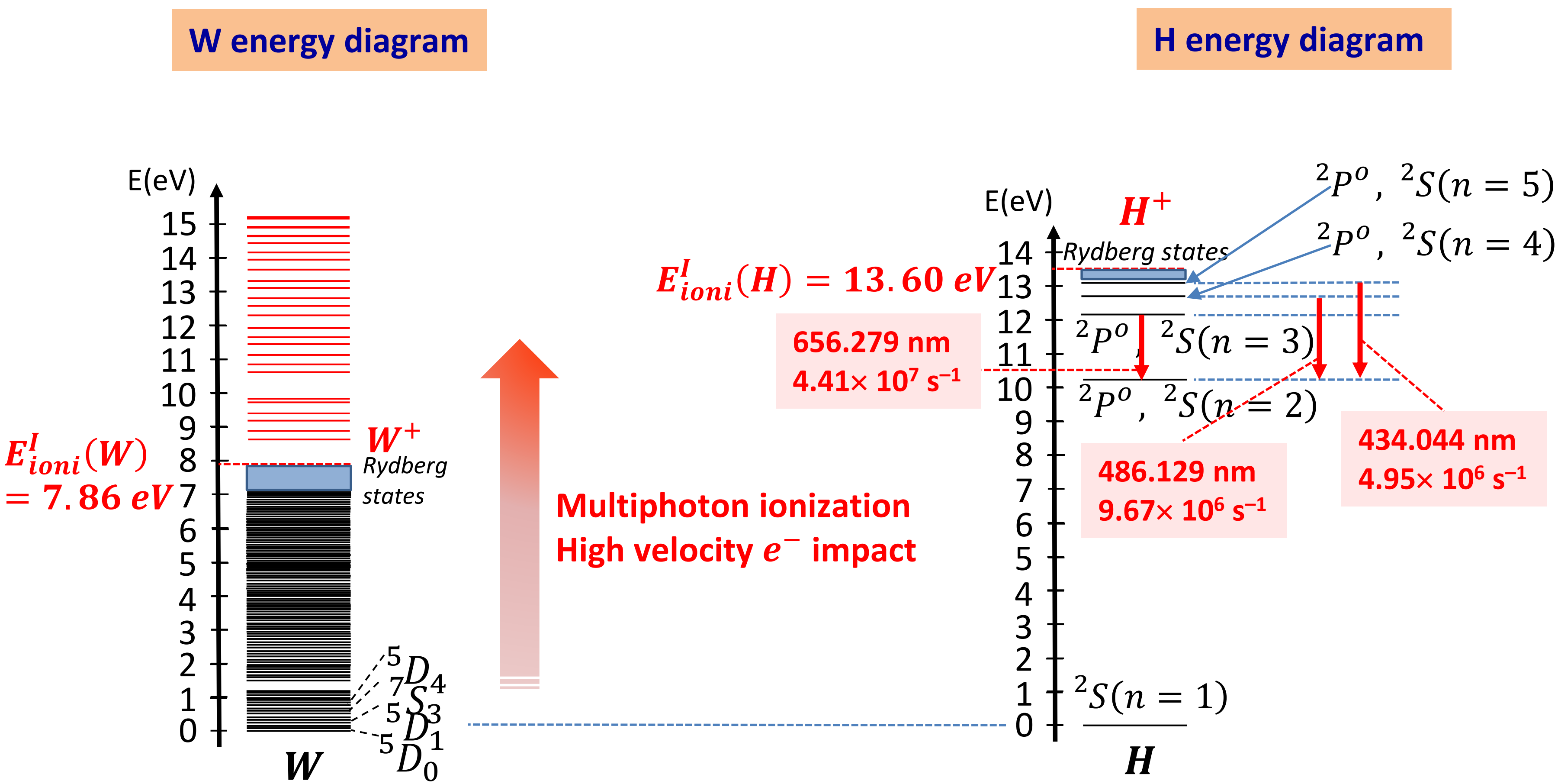
$$\frac{de_{elec}}{dt} = K_{W,W^+}\,n_e\,[W, W^+]\,\varphi_{las} \quad (W\,m^{-3})$$

$$K_W = \frac{1}{3\,m_e\,\pi\,c^3}\,\frac{e^2}{4\pi\,\varepsilon_0}\sqrt{\frac{2}{m_e}\left(\frac{hc}{\lambda}+\frac{3}{2}k_BT_e\right)}\left(1+\frac{3k_BT_e}{hc}\lambda\right)\overline{\sigma_{e-W}}\,\lambda^2\ (m^5)$$

$$K_{W^+} = \frac{4}{3}\left(\frac{e^2}{4\pi\,\varepsilon_0}\right)^3\sqrt{\frac{2\pi}{3\,m_e k_B T_e}}\,\frac{G}{m_e\,h\,c^4}\,\lambda^3 \quad (m^5)$$

$$P_{IB} \gg P_{MPI}$$

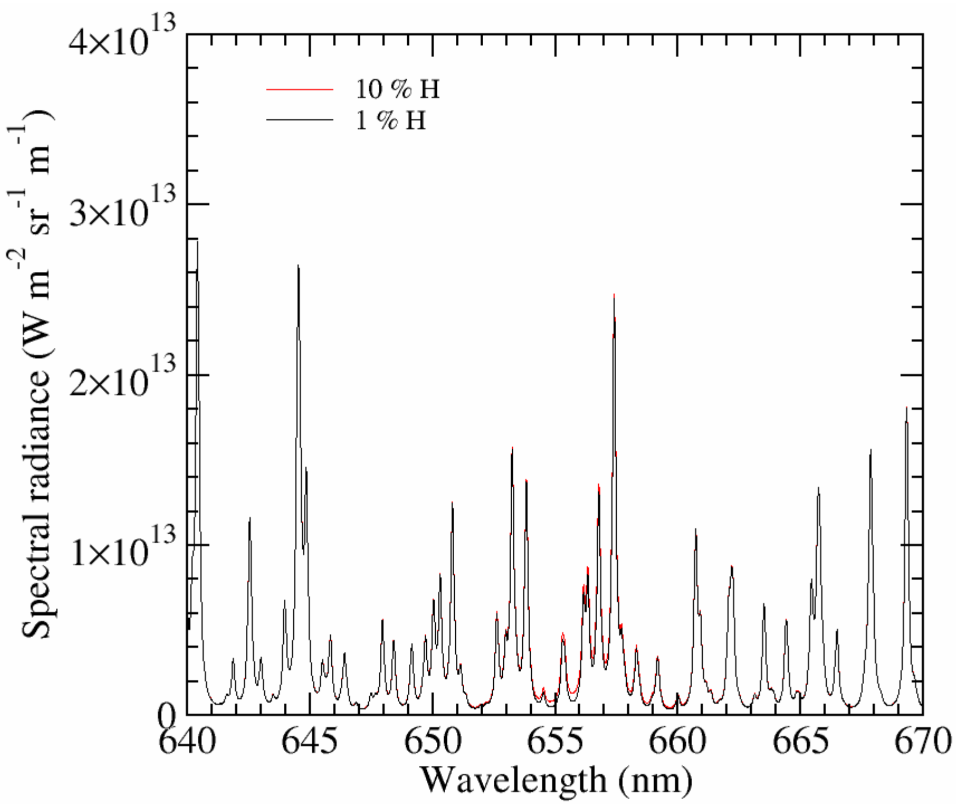
Advantages from the spectroscopic point of view…



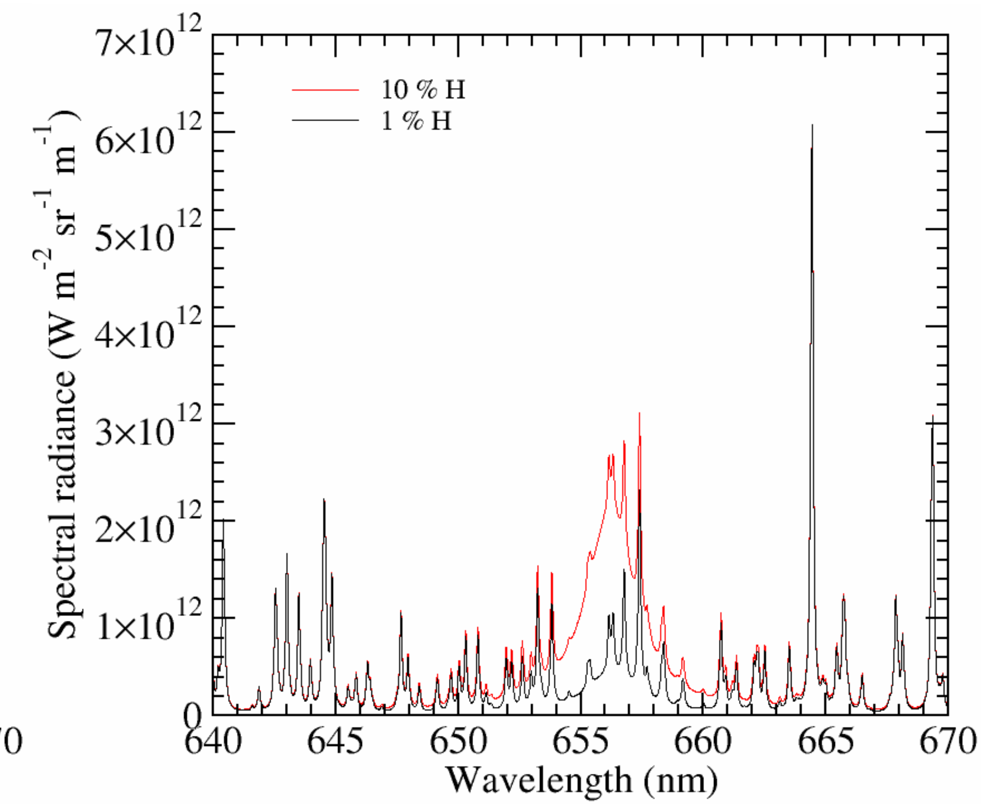
Consequences on spectra…

Multi**E**lemental **R**adiative equi**L**ibr**I**um emissio**N**

$T_e = 10\,000\ K$



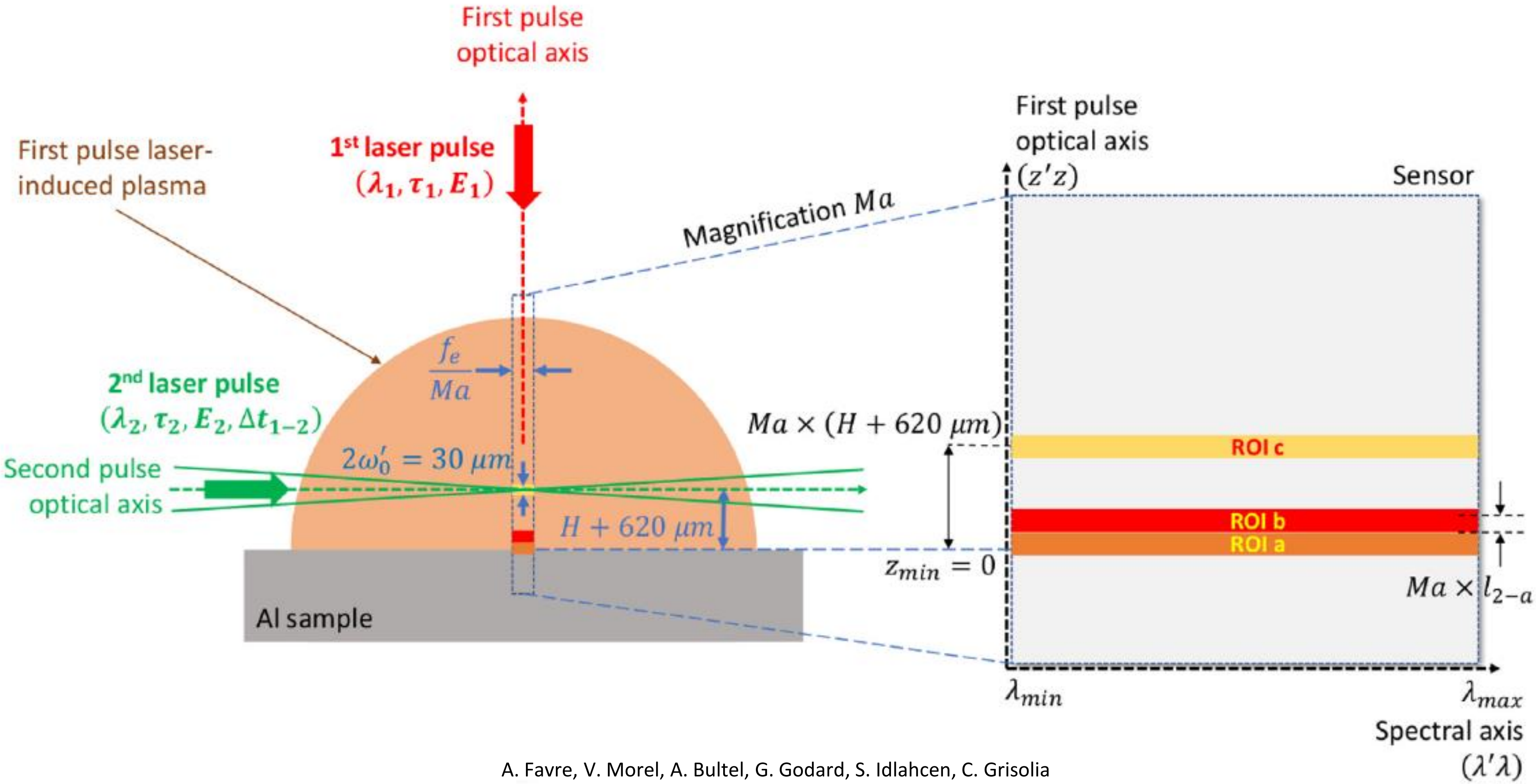
$T_e = 13\,000\ K$



A. Favre, M. Lesage, V. Morel, A. Bultel, P. Boubert
International Workshop on LIBS, Dec. 1-2, 2020, Szeged, Hungary

« Double pulse » experiments…

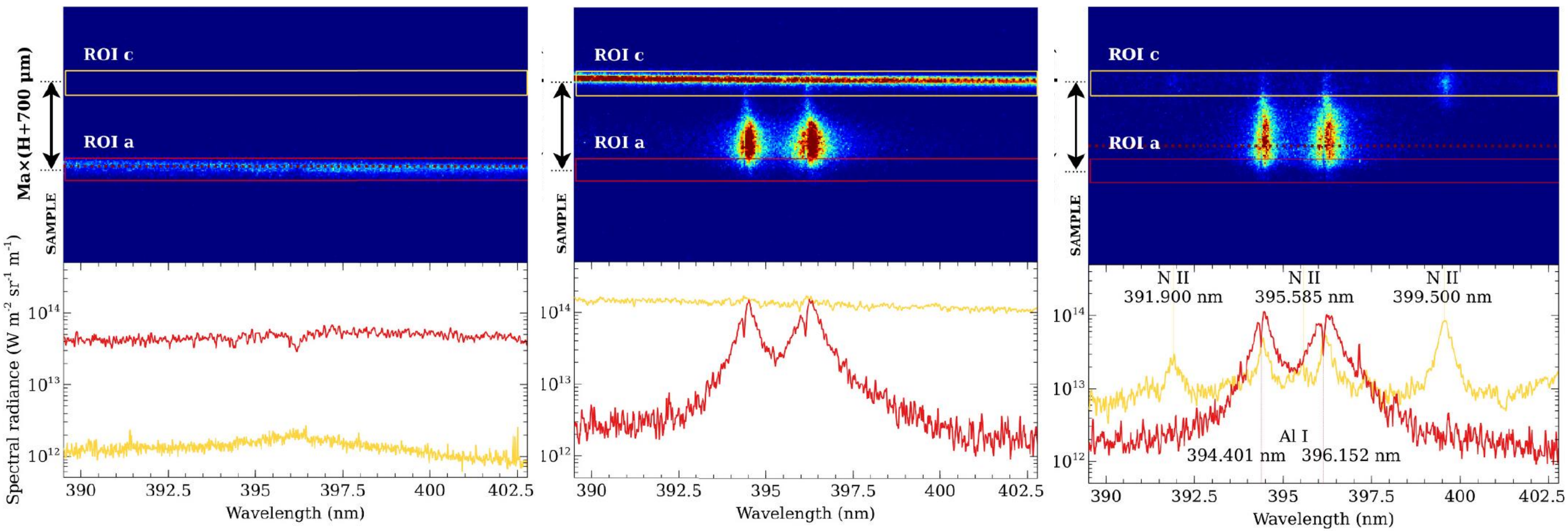
Time delay Δt_{1-2}



A. Favre, V. Morel, A. Bultel, G. Godard, S. Idlahcen, C. Grisolia
Spectrochim. Acta Part B **175** (2021) 106011

« Double pulse » experiments…

Time delay $\Delta t_{1-2} = 350\ ns$



t = 40 ns

t = 380 ns

t = 480 ns

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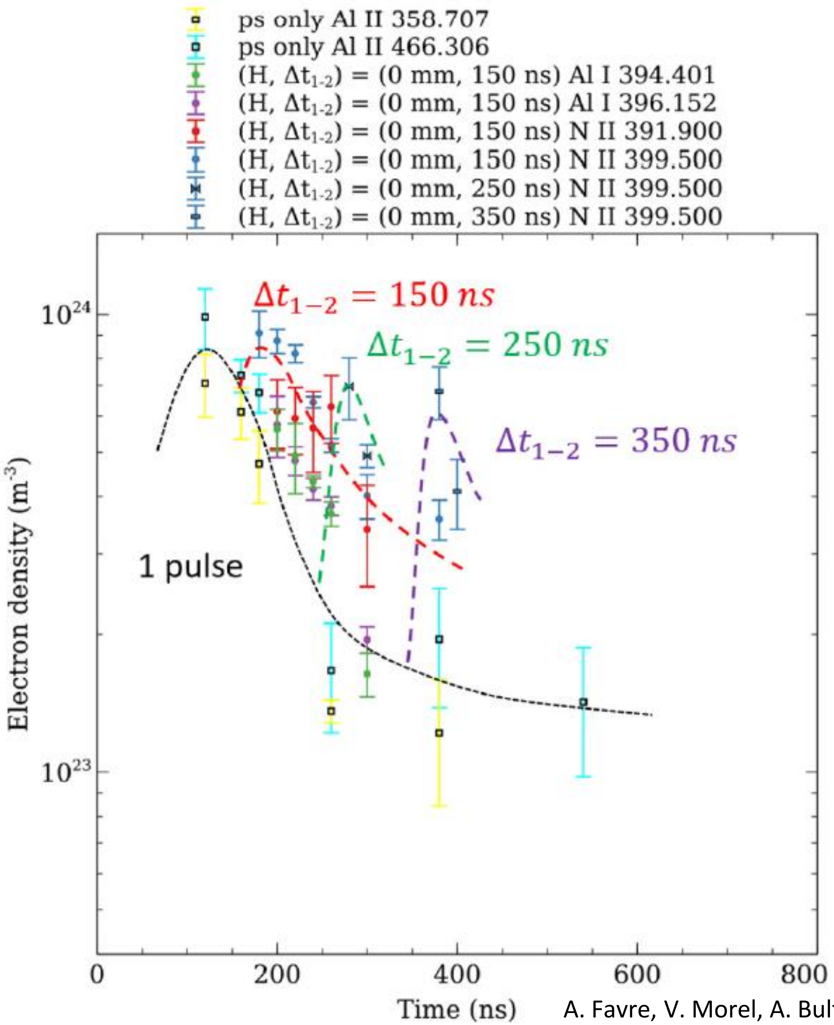
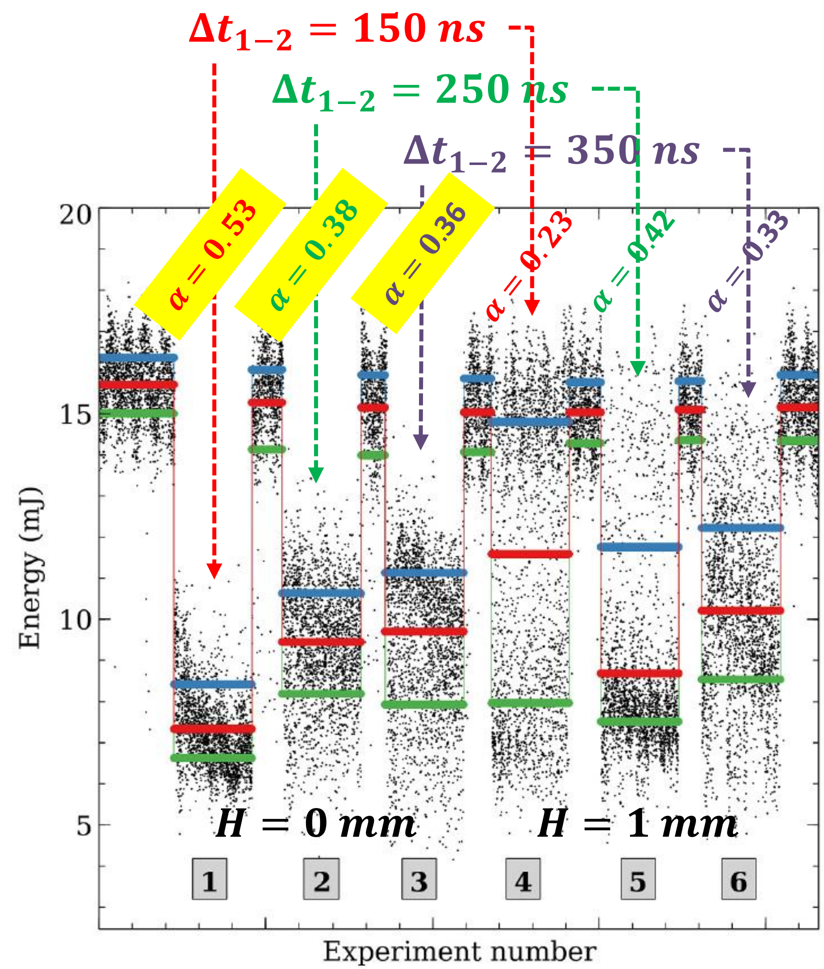
« Double pulse » experiments…

Different time delay Δt_{1-2}





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Spectrochim. Acta Part B **175** (2021) 106011

atelier

Plasma laser : diagnostic et modélisation

seconde partie

Plasmas en déséquilibre

- **Situation de déséquilibre possible à la surface, dans l'échantillon, dans le plasma**
- **Déséquilibre de phase, de translation, d'excitation, d'ionisation, chimique, entre la matière et le rayonnement**
- **Nécessite des études spectroscopiques en émission, absorption, diffusion (Thomson), LIF, etc. résolues en temps et en espace**
- **Modélisation possible à condition… de disposer de bases de données pertinentes !**