LIBS –
Principes Physiques Fondamentaux

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Outline

1. **Laser-Sample Interaction**
2. **Dynamics of the Expansion – Shockwave**
3. **Departure from Equilibrium (McWhirter and Co.)**

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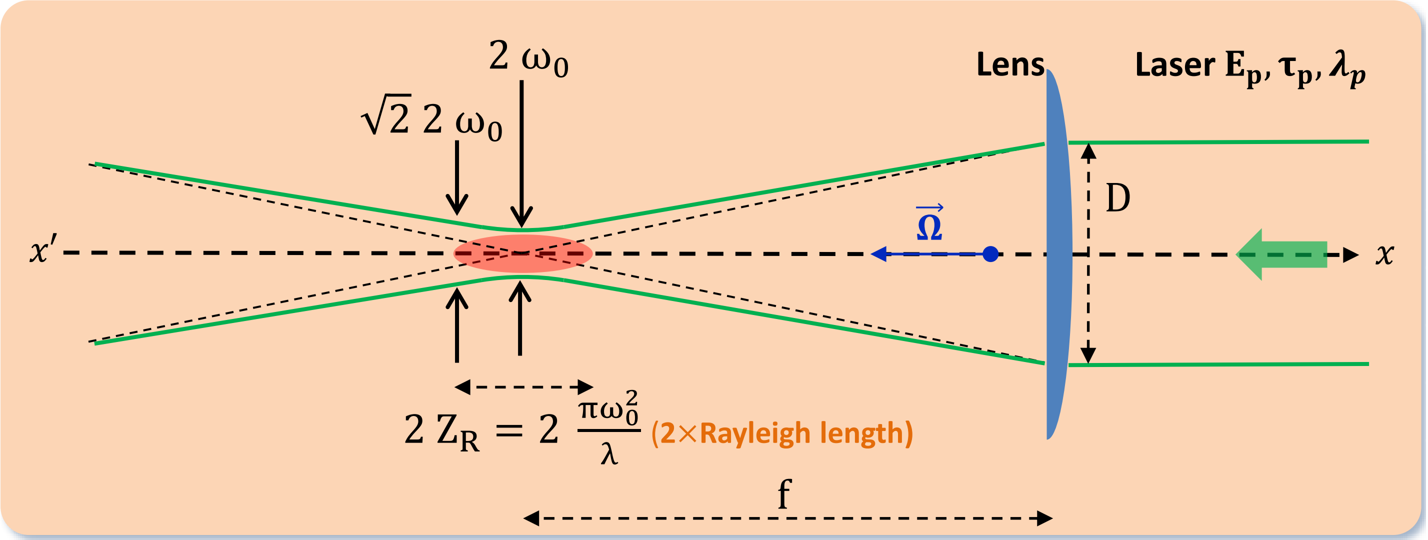
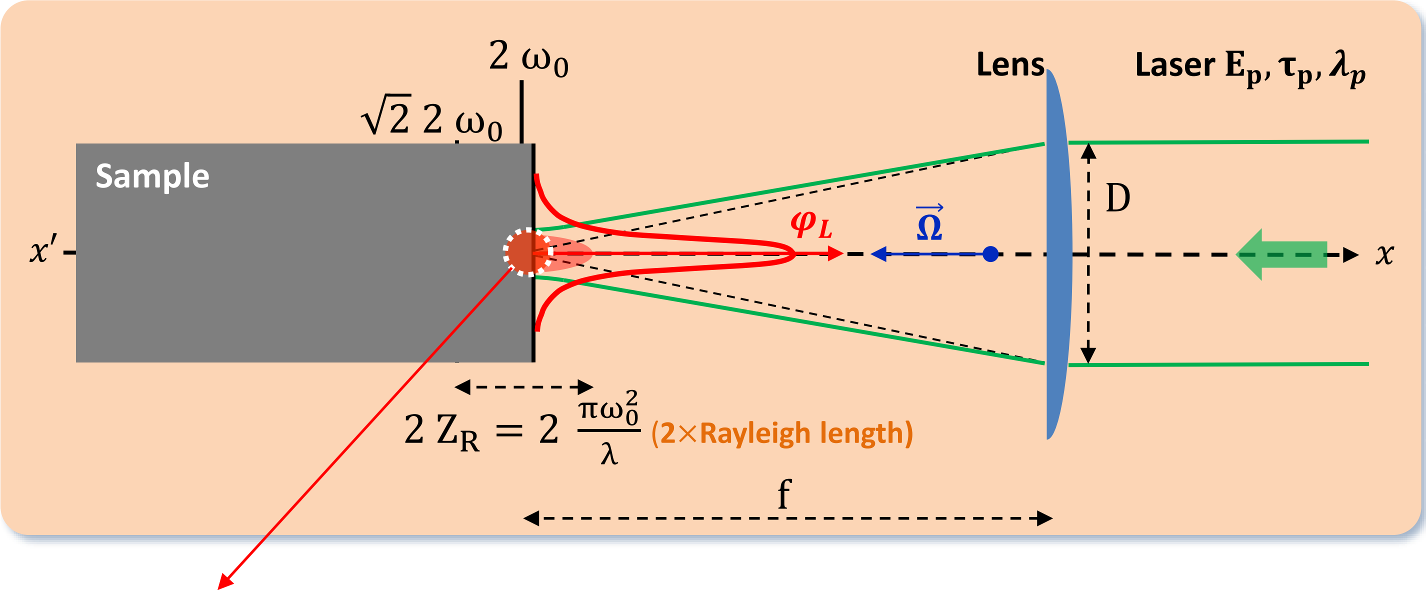


Variable	Typical values
$E_p(mJ)$	10
$\tau_p(ns)$	5
$\lambda_p(nm)$	532, 1064
$D(mm)$	4
$f(cm)$	10
$\omega_0(\mu m)$	100
$\Delta\sigma(cm^{-1})$	0.01
$\varphi_L(Wm^{-2})$	$10^{13} - 10^{14}$
L_L $(Wm^{-2}sr^{-1}m^{-1})$	$10^{16} - 10^{17}$

$$L_L \sim \underbrace{\frac{E_p}{\tau_p}\frac{1}{\pi\omega_0^2}}_{\varphi_L}\frac{4f^2}{\pi D^2}\Delta\sigma$$

Laser-sample interaction…



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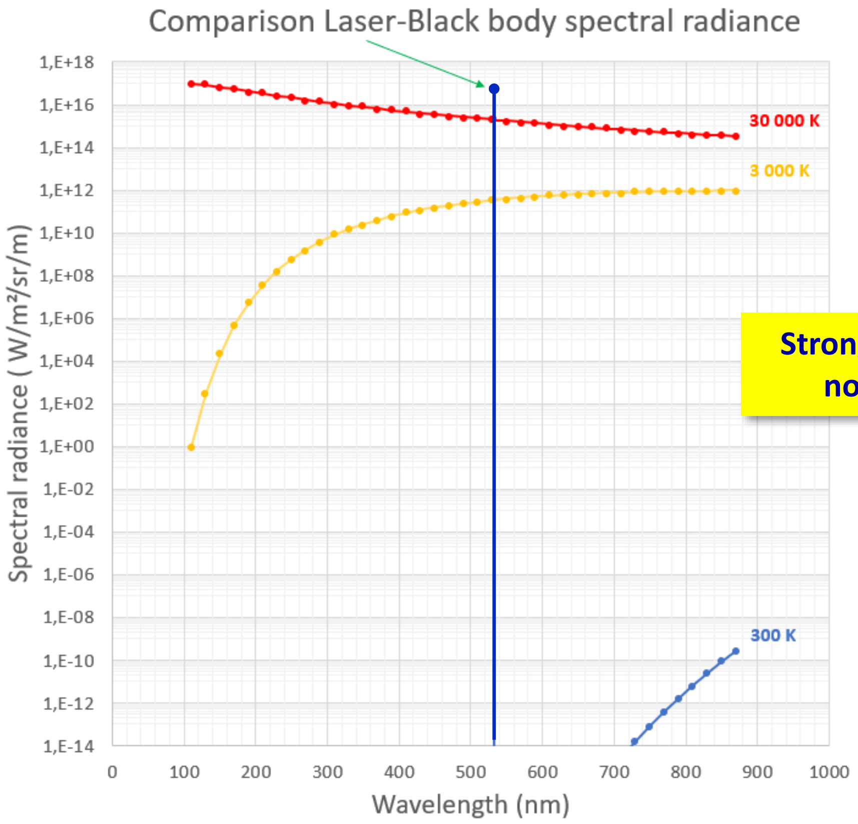
Radiative transfer within the sample…

Radiation within the sample...



Strong Matter-radiation non equilibrium...

Variable	Typical values
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Photon-particle interaction within the sample…

Photons are bosons!!!

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	$A^+ + e^- + nh\nu \leftarrow A_i + (n+1)h\nu$
Multiphoton ionization **MPI**	$A^+ + e^- + nh\nu \leftarrow A_i + (n + \boldsymbol{m} \geq 2)h\nu$

All these processes are governed by the Planck's law

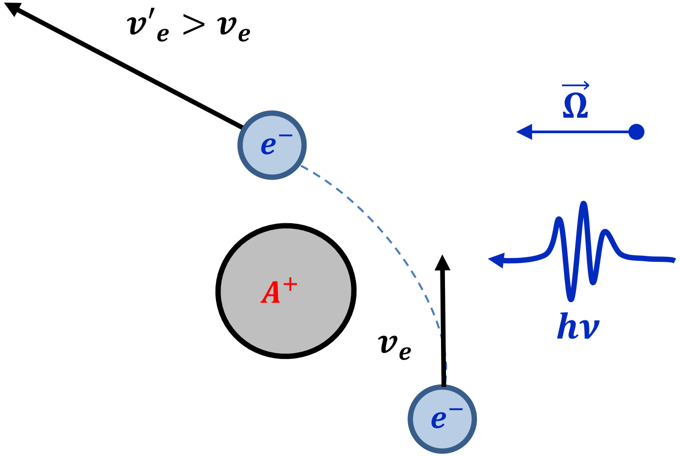
If the sample is dielectric

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

⇒ Increase in the volumic electron energy $\boldsymbol{e_e}$ …

Inverse Bremsstrahlung…



Since T_e is locally defined ($\tau_p \gg \tau_{ee} \approx 10^{-14} s$)…

$$\left(\frac{\partial e_e}{\partial t}\right)_{IB} = \frac{3}{2} n_e k_B \left(\frac{\partial T_e}{\partial t}\right)_{IB} = K_{A^+}(x,t)\, n_e\, [A^+]\, \varphi_L(x,t) \qquad (W\, m^{-3})$$

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

Since T_{A^+} is locally defined ($\tau_p \gg \tau_{A^+A^+} \approx 10^{-14} s$)…

$$\left(\frac{\partial e_{A^+}}{\partial t}\right)_{EC} = \frac{3}{2}[A^+]\, k_B \left(\frac{\partial T_{A^+}}{\partial t}\right)_{EC} = \frac{3}{2}[A^+]\, k_B\, \frac{T_e - T_{A^+}}{\tau_{A^+-e}} \quad (W\, m^{-3})$$

$$\tau_{A^+-e} = \left[n_e \frac{\sqrt{2\pi}}{m_e\, m_{A^+}} \left(\frac{e^2}{4\pi\,\varepsilon_0}\right)^2 \frac{ln(\Lambda_{A^+-e})}{\left(\frac{k_B T_e}{m_e} + \frac{k_B T_{A^+}}{m_{A^+}}\right)^{3/2}} \right]^{-1} \approx 10^{-12}\, s$$

$$\tau_{A^+-e} \ll \tau_p \; if \; ns, \tau_{A^+-e} \gg \tau_p \; if \; fs$$

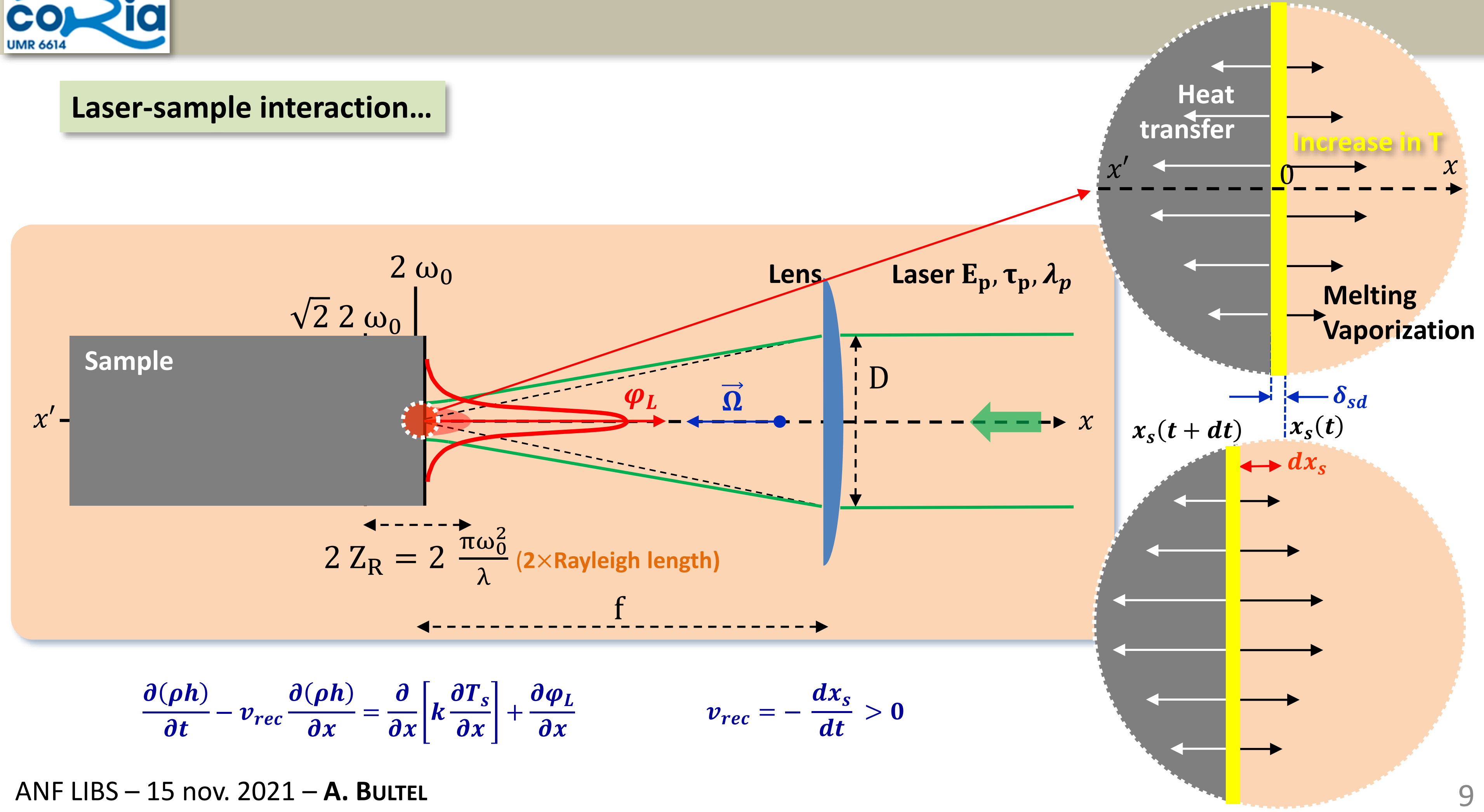
Photons disappear…

$$\frac{1}{c}\frac{\partial L_L}{\partial t} + \vec{\Omega}\; \overrightarrow{grad}\, L_L = \frac{\dot{L}_L}{c} \qquad (W\, m^{-4}\, sr^{-1}) \; with \; \dot{L}_L < 0 \; (also \; scattering \ldots)$$

$$\overrightarrow{grad}\, L_L = -\vec{\Omega}\, \frac{L_L(0,t)}{\delta_{sd}}$$

$$\delta_{sd} = \frac{\lambda_p}{2\pi\, k_{RI}} \approx 10^{-8}\, m \qquad (skin\; depth)$$

Laser-sample interaction…



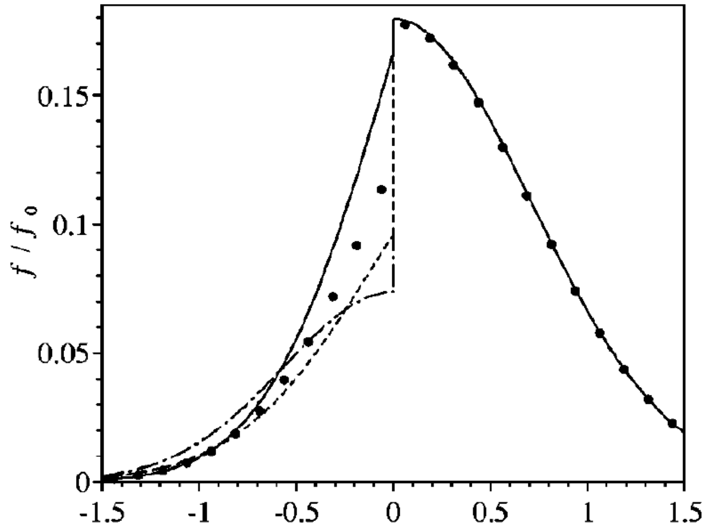
$$\frac{\partial(\rho h)}{\partial t} - v_{rec}\frac{\partial(\rho h)}{\partial x} = \frac{\partial}{\partial x}\left[k\frac{\partial T_s}{\partial x}\right] + \frac{\partial \varphi_L}{\partial x}$$

$$v_{rec} = -\frac{dx_s}{dt} > 0$$

Very fast vaporization…

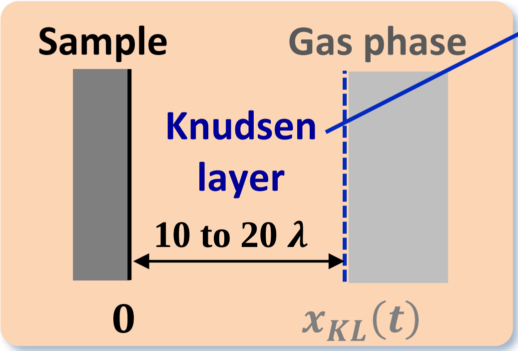
$T_s < 0.9\,T_c$

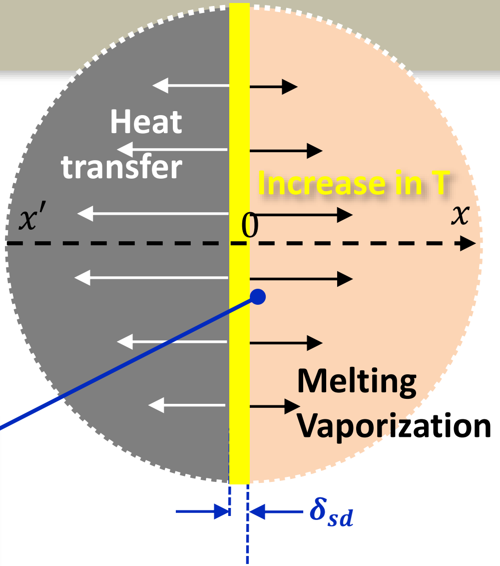
T_s much higher than the temperature in the gas phase…



$v_x / \sqrt{\dfrac{2\,k_B\,T_s(0,t)}{m}}$

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

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

$0.9\, T_c < T_s < T_c$

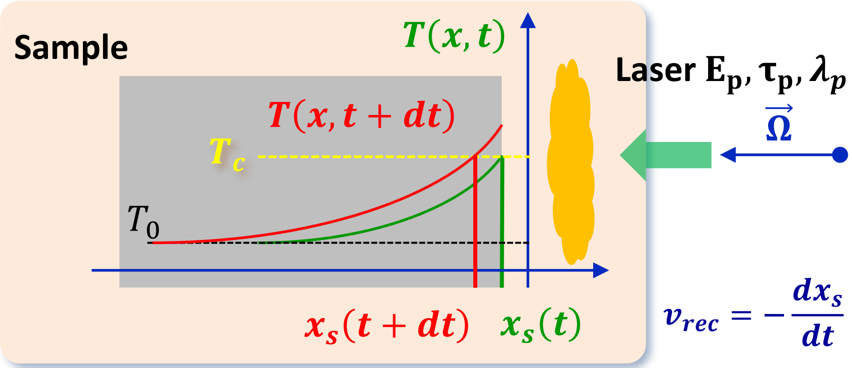
Formation of µ-bubbles within the liquid

→ Explosive boiling lasting more than the laser pulse

$T_s > T_c$?

Not phase change anymore → Supercritical fluid

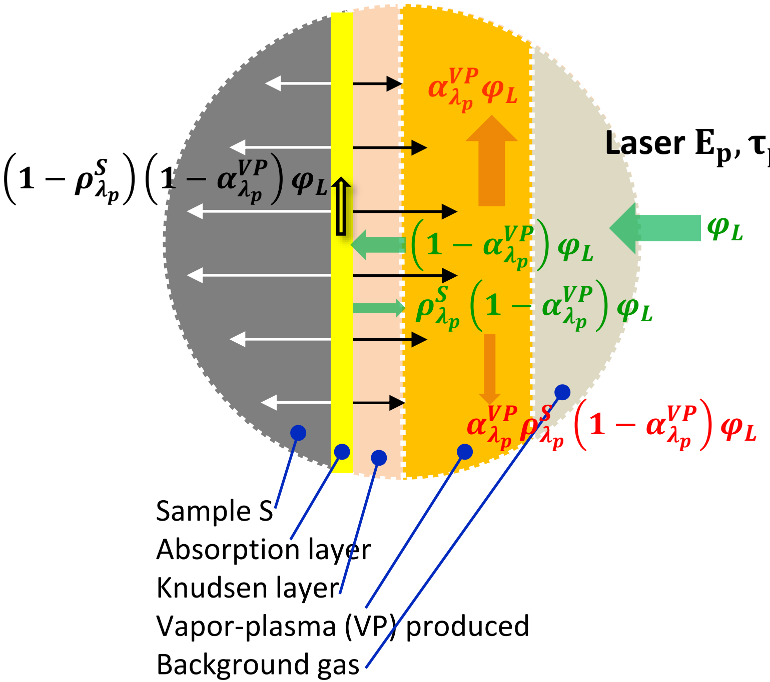
This supercritical fluid can be overheated…

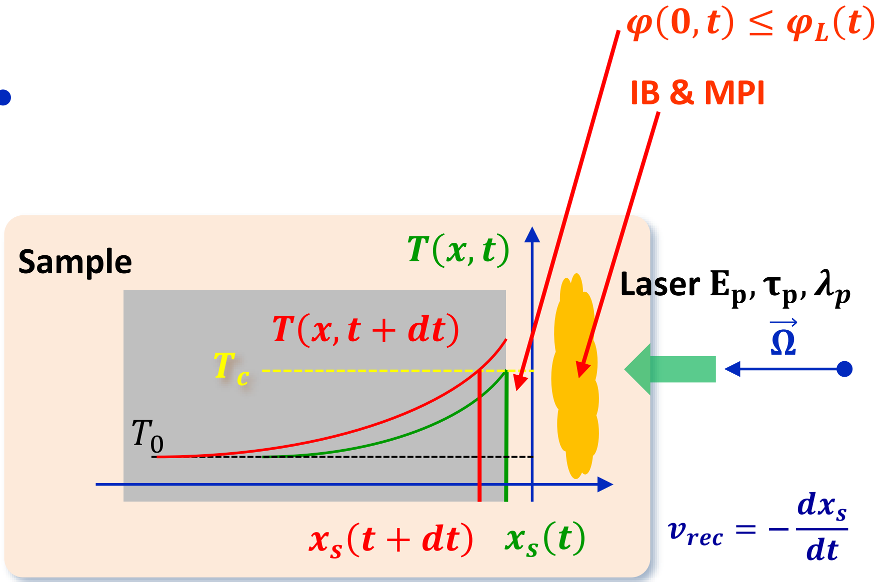


$$v_{rec} = -\frac{dx_s}{dt}$$

Phase non-equilibrium

Screening of the laser pulse by the vapor-plasma produced…

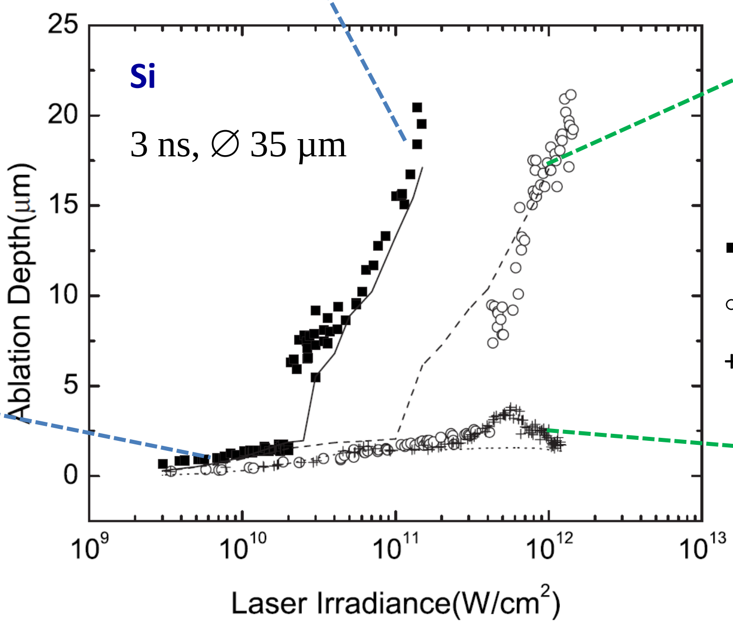




Ablation process…

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

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



▪ 266 nm

○ 532 nm

+ 1064 nm

$T_s \ngtr 0.9\,T_c$ 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

No explosive boiling for $1064\ nm$ because of IB efficiency

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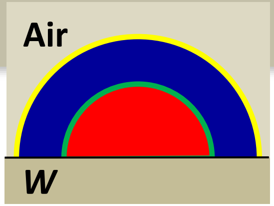
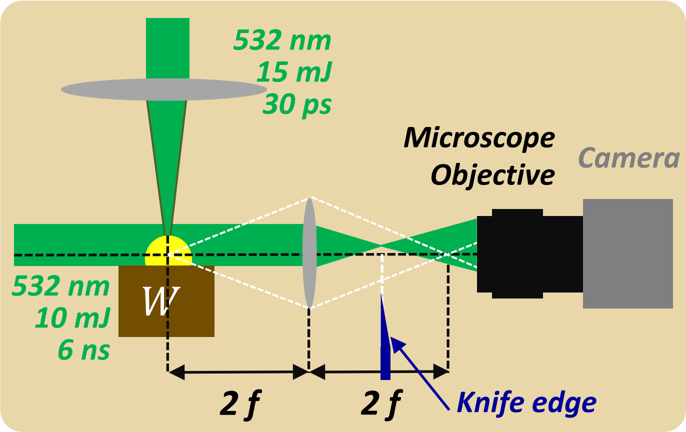
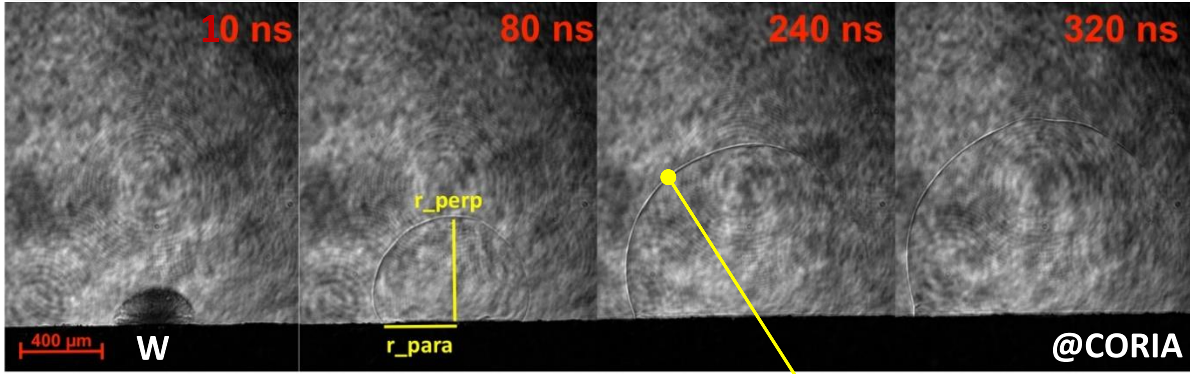


Interaction with a background gas…



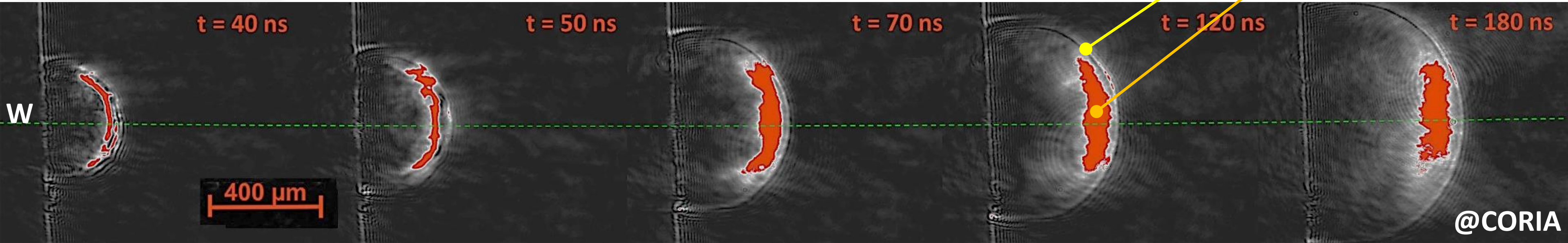
Laser shadowgraphy Sensitive to $\frac{\partial^2 n}{\partial z^2}$



Shockwave

Contact surface

Schlieren Imagery Sensitive to $\frac{\partial n}{\partial z}$



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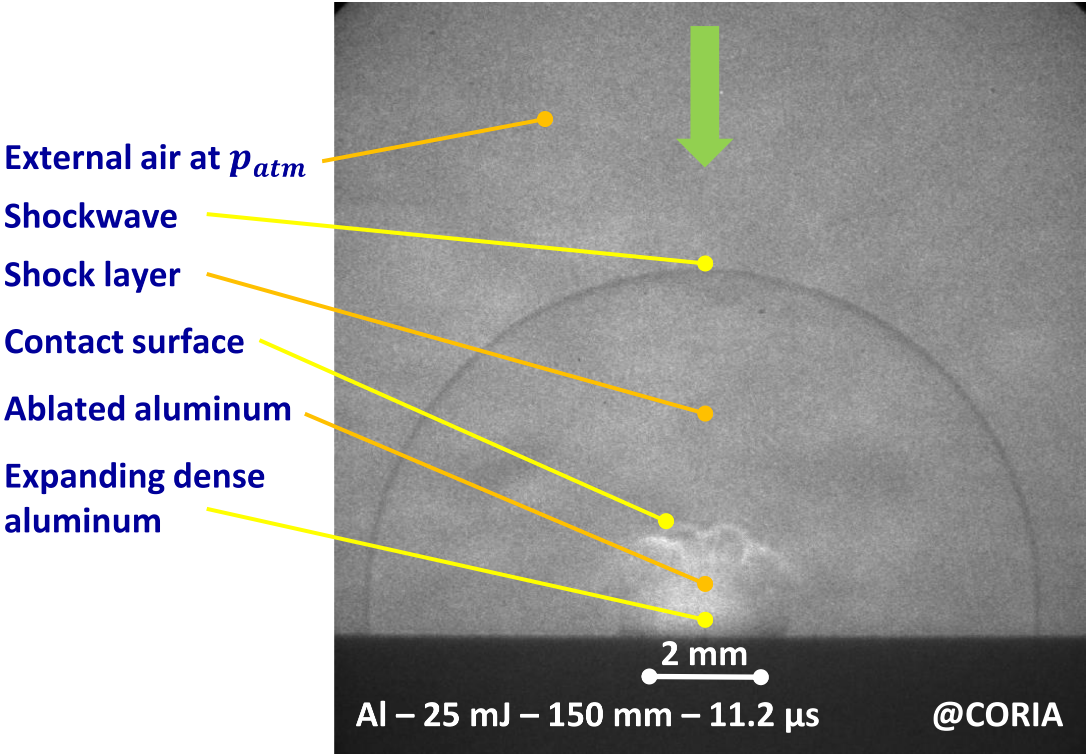
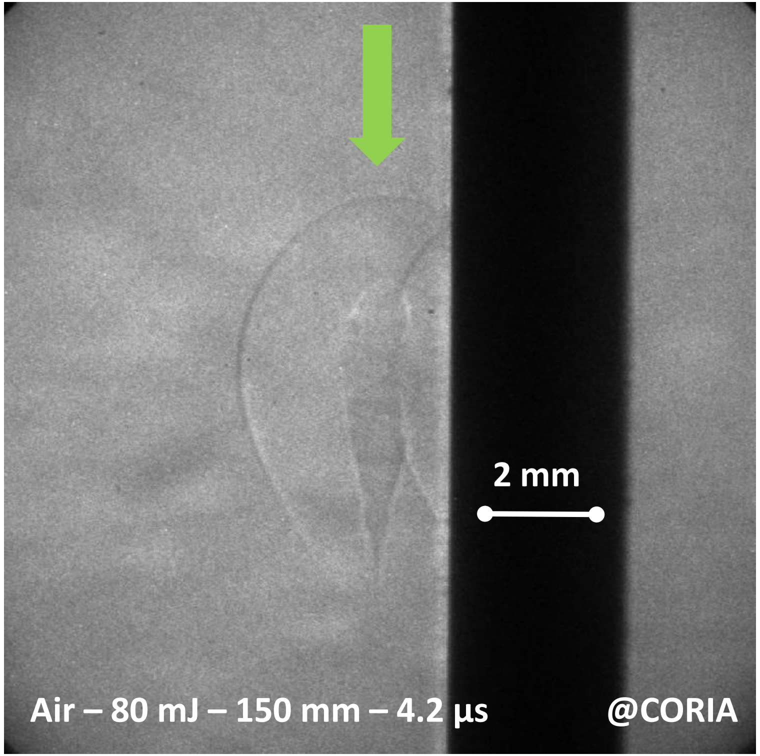
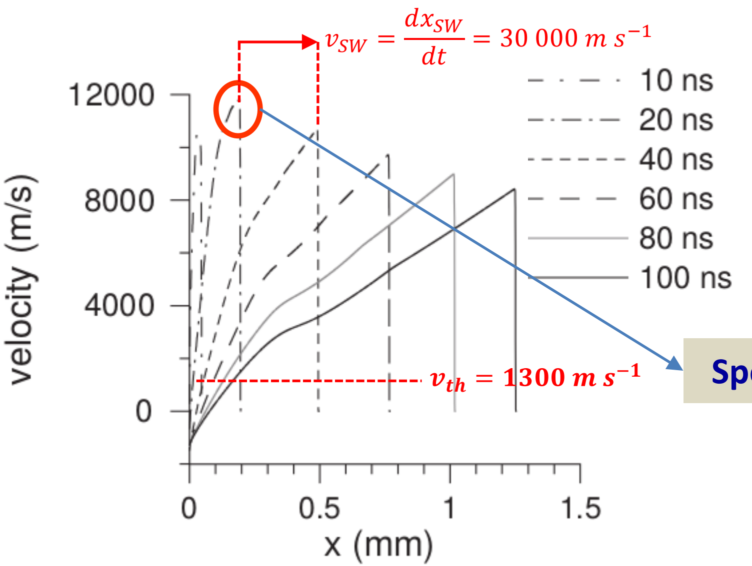




Induced expansion…

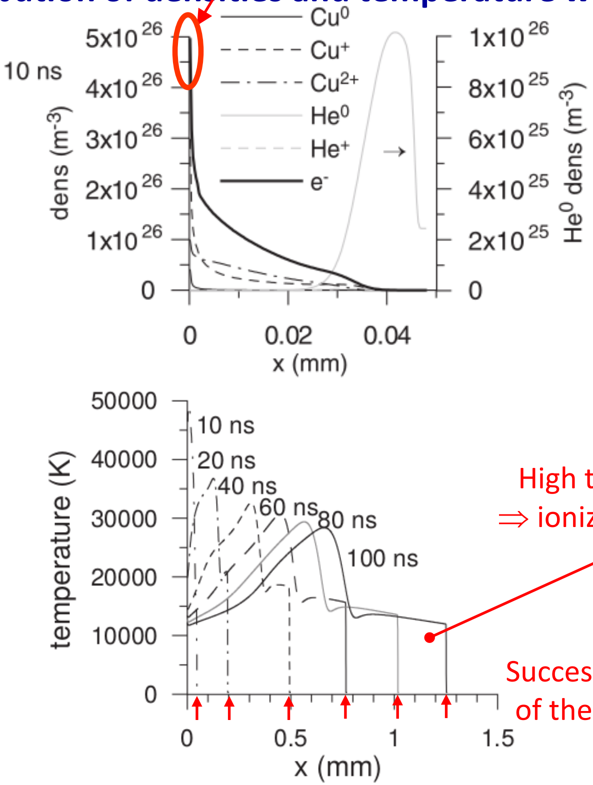
Distribution of the plasma local speed with time



Speed $\gg v_{th}$

Distribution of densities and temperature with time

Shorter times: screening by inverse Bremsstrahlung



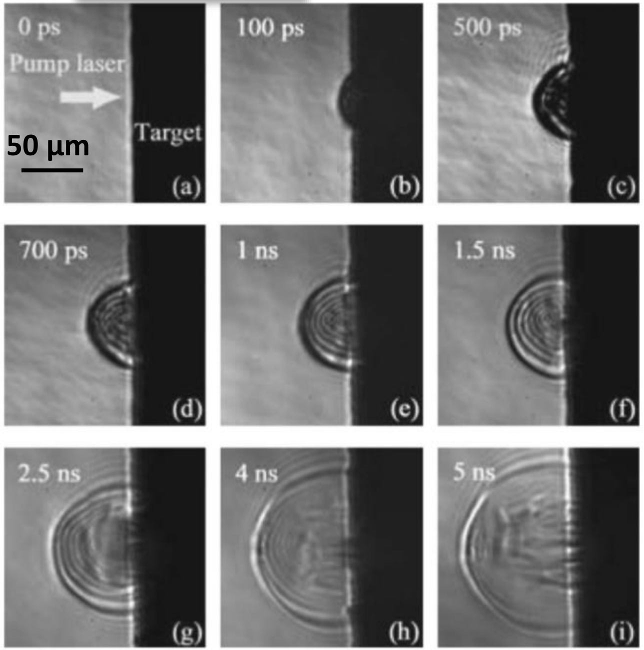
High temperature
$\Rightarrow$ ionization kinetics

Successive position of the shock front

Cu in He (p_{atm}), $\varphi_L = 10^{13}\ W\ m^{-2}$, $\lambda_L = 266\ nm$, $\tau_L = 5\ ns$

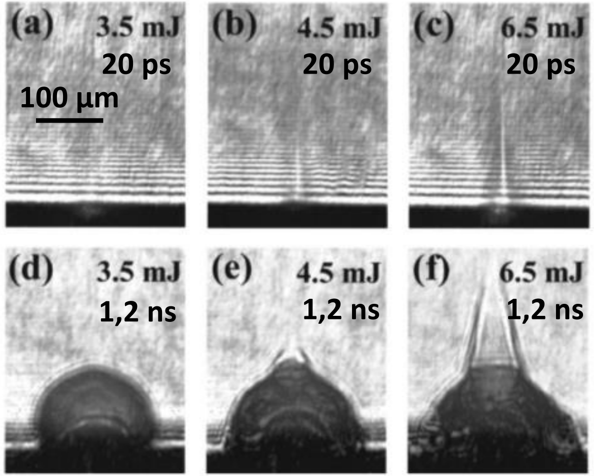
A. Bogaerts *et al.* Spectrochim. Acta Part B **60** (2005) 1280

Structure…



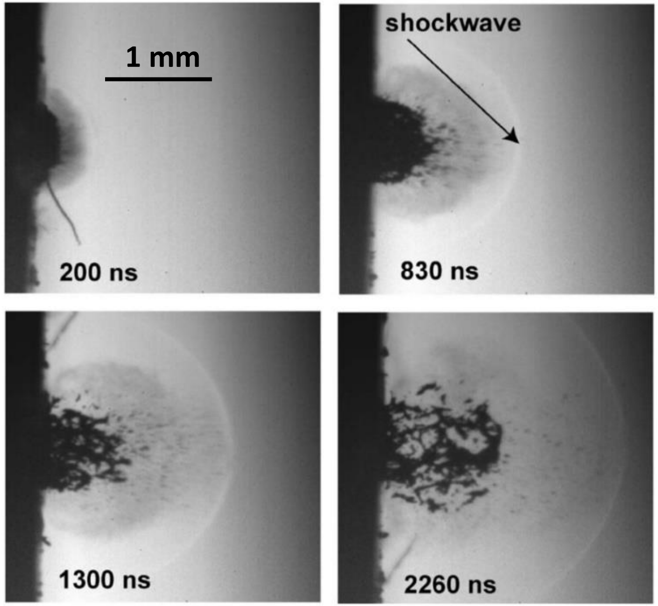
50 fs
800 nm
40 J cm^{-2}
Al

N. Zhang *et al.*
Phys. Rev. Lett. **99** (2007) 167602



35 ps
1064 nm
~ 50 J cm^{-2}
Cu

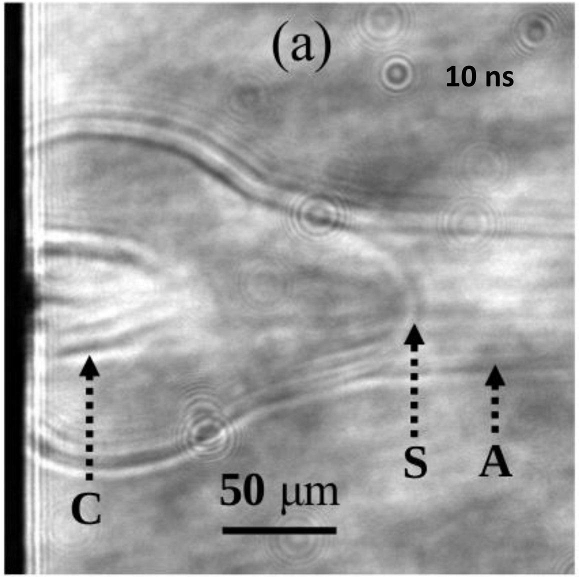
S.S. Mao *et al.*
Appl. Phys. Lett. **77** (2000) 2464

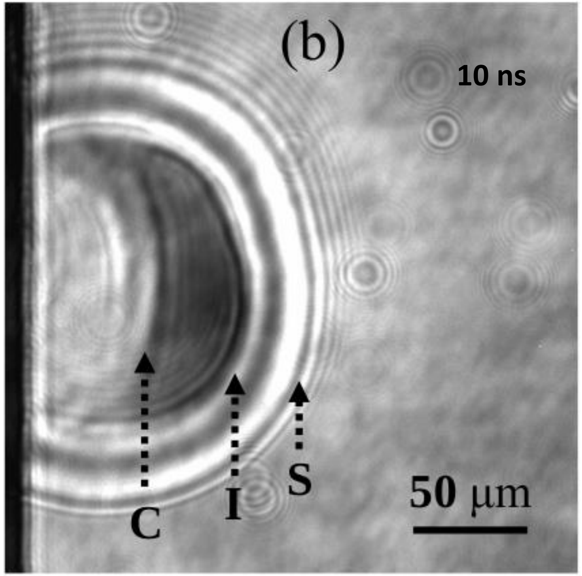


6 ns
1064 nm
6,5 J cm^{-2}
Polymère

M. Hauer *et al.*
Opt. Las. Eng. **43** (2005) 545

Structure…





100 fs
266 nm
11 J cm^{-2}
Si (air)

X. Zeng *et al.*
Lawrence Berkeley National Lab. (2004)

3 ns
266 nm
11 J cm^{-2}
Si (air)

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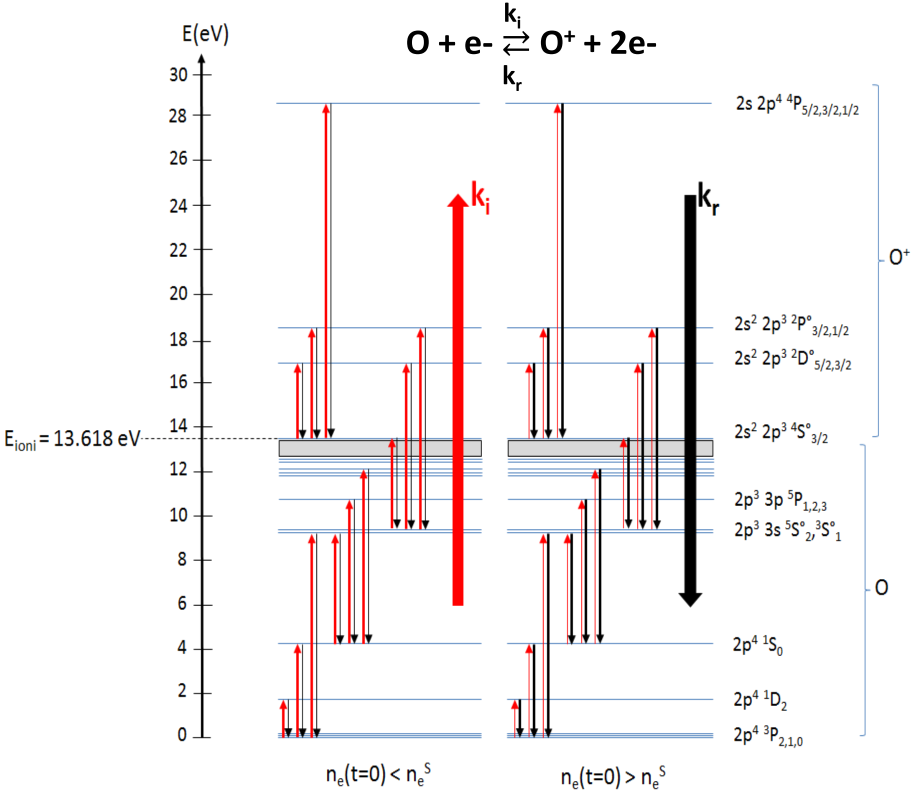
Kinetics of ionization – Example



Individual X_m variation rate

$$\frac{1}{V}\frac{dN_{X_m}}{dt} = -\sum_{n>m} k_{m\to n}\left(1-\frac{[X_n]}{[X_m]K^B_{n,m}}\right)[X_m]n_e$$

$$+\sum_{n<m} k_{n\to m}\left(1-\frac{[X_m]}{[X_n]K^B_{m,n}}\right)[X_n]n_e$$

$$-\sum_i \; k^+_{m\to i}\left(1-\frac{[X_i^+]n_e}{[X_m]K^S_{i,m}}\right)[X_m]n_e$$

Individual X_i^+ variation rate

$$\frac{1}{V}\frac{dN_{X_i^+}}{dt} = -\sum_{j>i} k_{i\to j}\left(1-\frac{[X_j^+]}{[X_i^+]K^B_{j,i}}\right)[X_i^+]n_e$$

$$+\sum_{j<i} k_{j\to i}\left(1-\frac{[X_i^+]}{[X_j^+]K^B_{i,j}}\right)[X_j^+]n_e$$

$$+\sum_m k^+_{m\to i}\left(1-\frac{[X_i^+]n_e}{[X_m]K^S_{i,m}}\right)[X_m]n_e$$

Global X variation rate

$$\frac{1}{V}\frac{dN_X}{dt} = \sum_m \frac{1}{V}\frac{dN_{X_m}}{dt} = \begin{cases} -\boldsymbol{k_i[X]n_e} \\ +\boldsymbol{k_r[X^+]n_e^2} \end{cases}$$

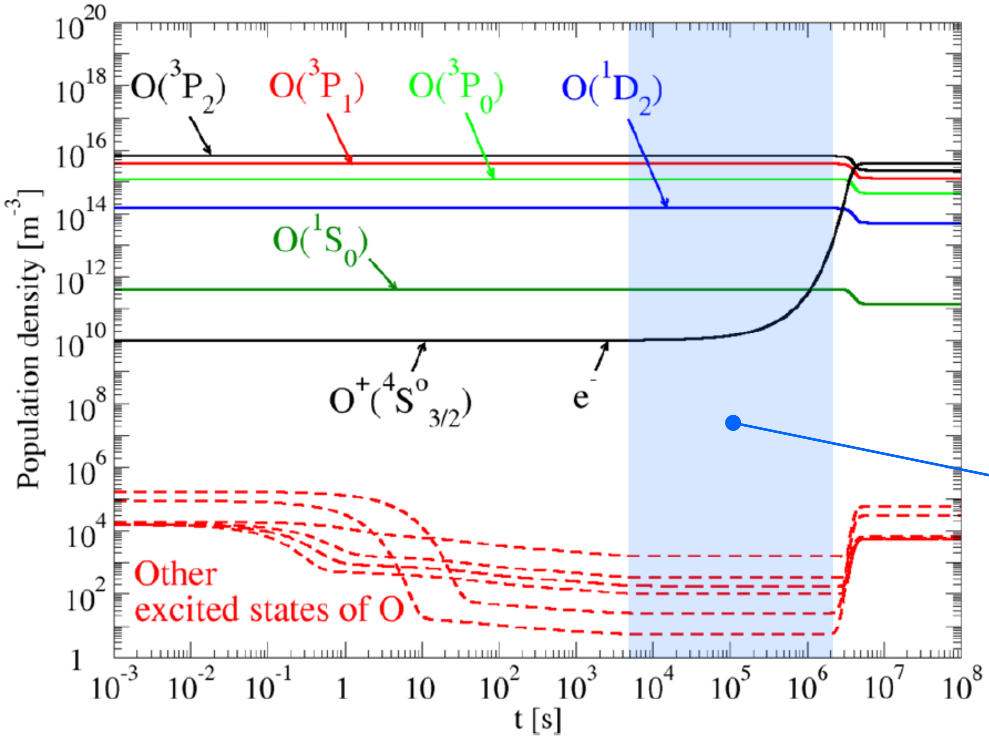
Global X^+ variation rate

$$\frac{1}{V}\frac{dN_{X^+}}{dt} = \sum_i \frac{1}{V}\frac{dN_{X_i^+}}{dt} = \begin{cases} +\boldsymbol{k_i[X]n_e} \\ -\boldsymbol{k_r[X^+]n_e^2} \end{cases}$$

J. Annaloro, V. Morel, A. Bultel *et al.*
Phys. Plasmas **19** (2012) 073515

Kinetics of ionization – Example

0D simulation



T_e = 6000 K

n_e(t=0) = 10^{10} m^{-3} p = 10^{-3} Pa

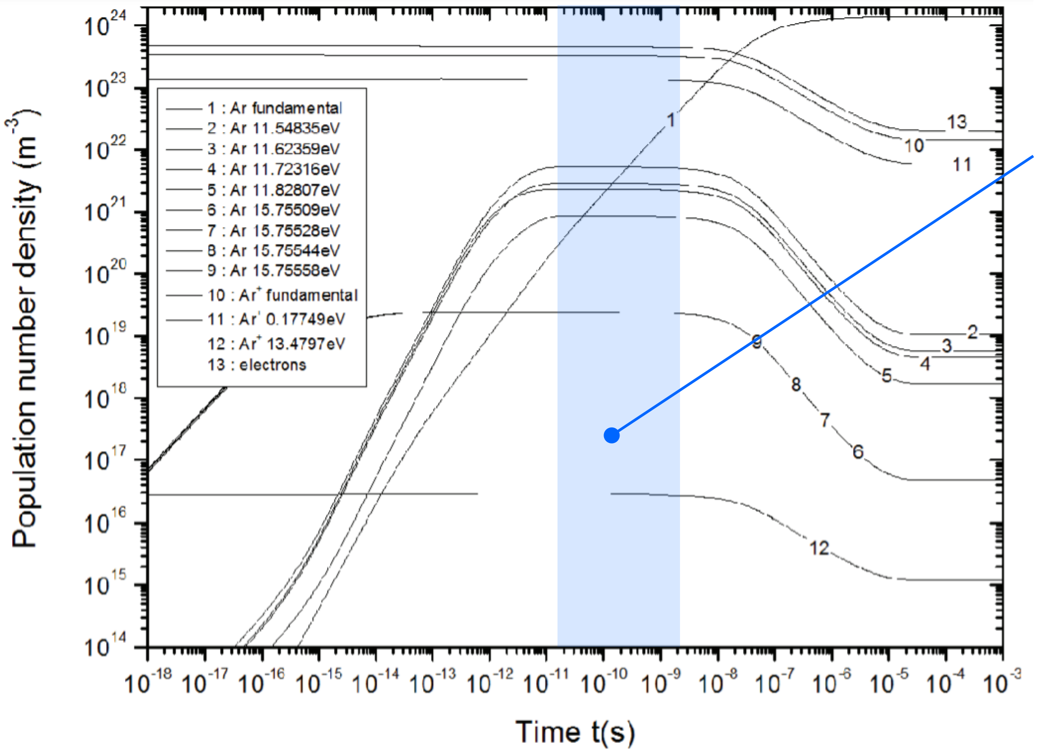
T_{exc}(t=0) = 6000 K

Quasi Steady State QSS

J. Annaloro, V. Morel, A. Bultel *et al.*
Phys. Plasmas **19** (2012) 073515

Kinetics of recombination – Example

Behaviour of the excited states



Quasi Steady State QSS

p_{atm}, **10 000 K**

J. Annaloro, P. Teulet, A. Bultel *et al.*
[Eur. Phys. J. D **71** (2017) 342]

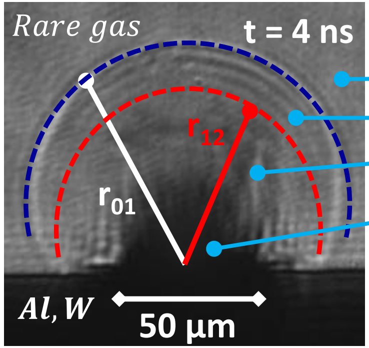
CORIA UMR 6614

The ECHREM* code

*Eulerian CHemically REactive Multi-component plasma code

Horizon 2020 — FR FCM — Investissements d'avenir — EMC3 energy materials & clean combustion center

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

Balance equations

Collisionnal-radiative source term

(1) Shock layer

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

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

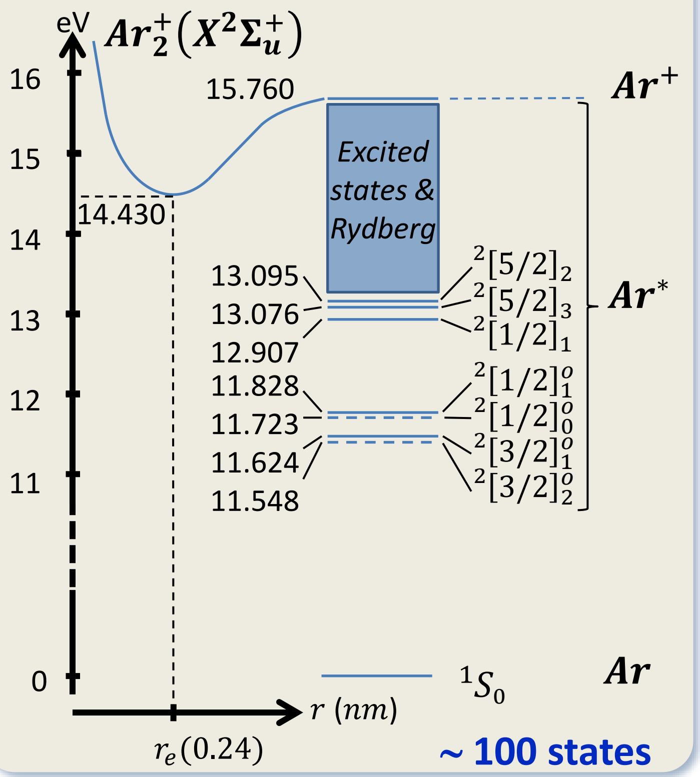
Energy $E_2 = M_2(\epsilon^{Al,W} + \epsilon_2) + E_{c,2} \Leftrightarrow \frac{dE}{dt} = \rho_0 \epsilon_0 v_{sf} 2\pi r_{01}^2 - \frac{M_2}{\rho_2}(4\pi\varepsilon_{RR} + 4\pi\varepsilon_{TB} + \varepsilon'_{SE})$

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 B **103-104** (2015) 112

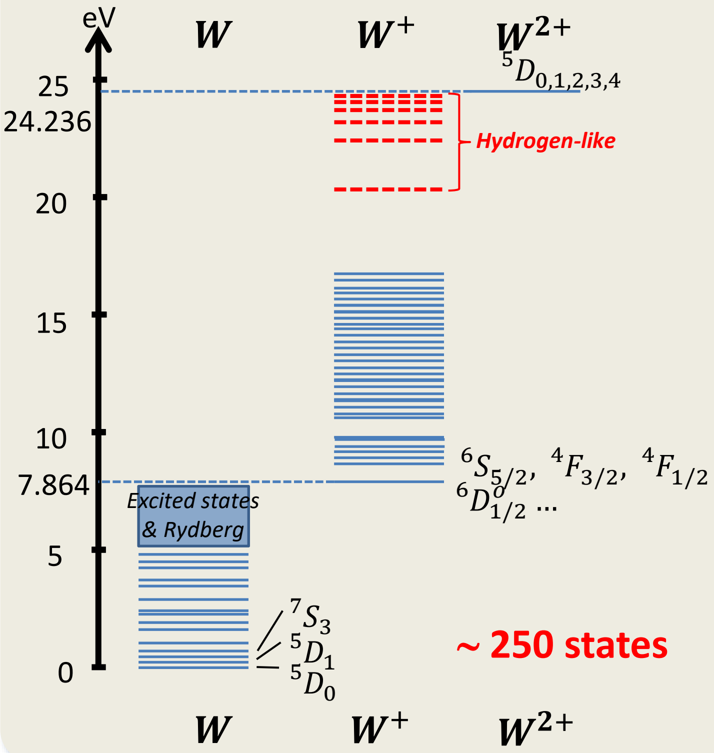
Ar-W…

Shock layer - *Argon*



~ 100 states

Central plasma - *Tungsten*



~ 250 states

The **ECHREM*** code

*Eulerian CHemically REactive Multi-component plasma code

Horizon 2020

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

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***

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

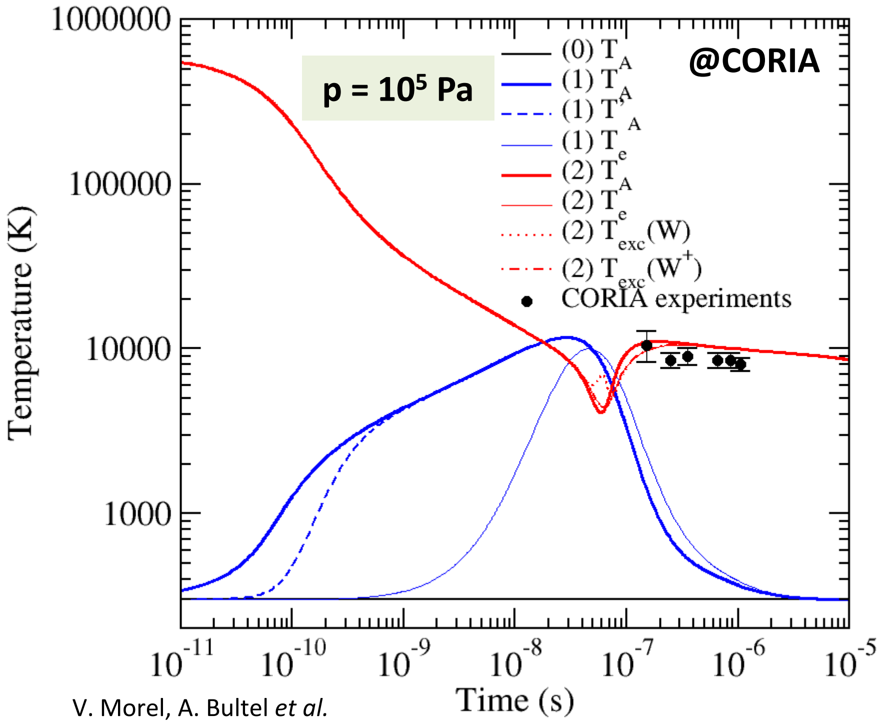
Radiative Database

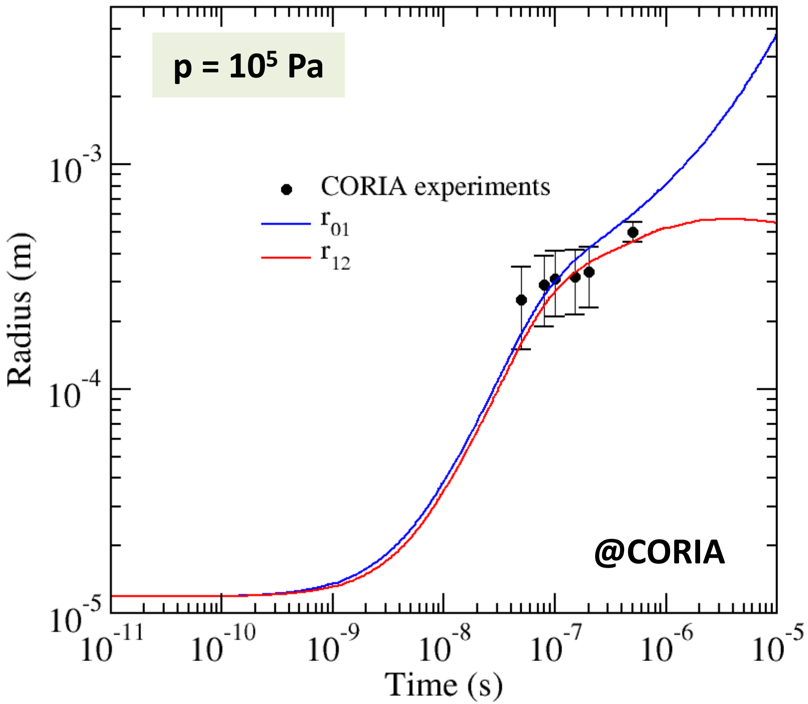
NIST, Atomic Line List, ADAS, HULLAC…

The **ECHREM*** code

*Eulerian CHemically REactive Multi-component plasma code

Horizon 2020 · FR FCM · Investissements d'avenir · EM2C energy materials & clean combustion center





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

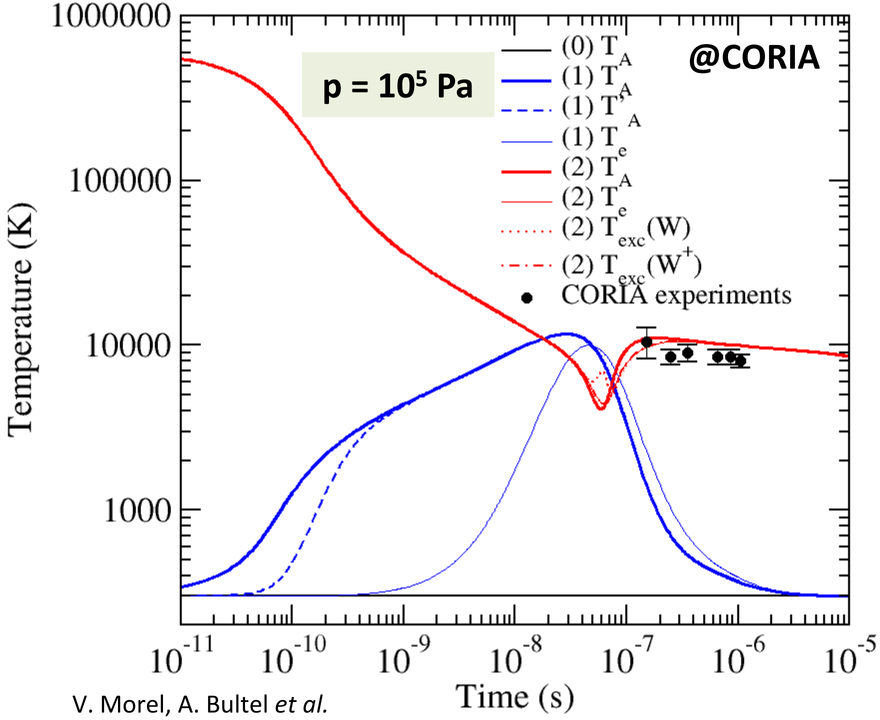
W (Ar) 10 ps 532 nm 10 J cm^{-2}

CoRIA UMR 6614

The **ECHREM*** code

*Eulerian CHemically REactive Multi-component plasma code

Horizon 2020 FR FCM EMC3 energy materials & clean combustion center



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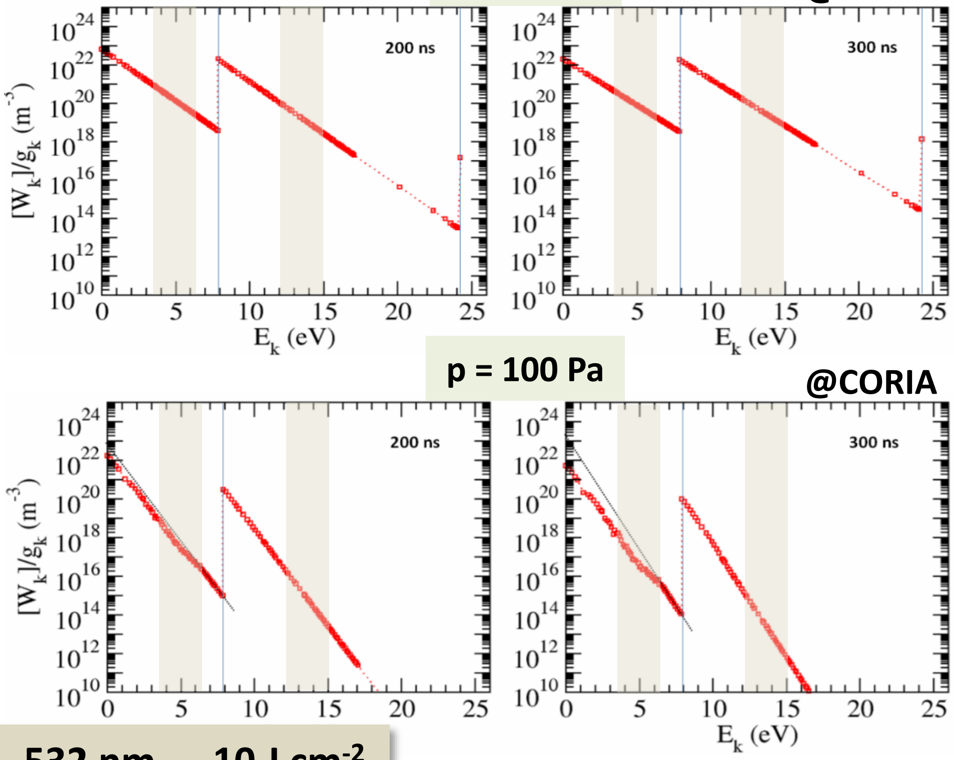


W (Ar) 10 ps 532 nm 10 J cm^{-2}

McWhirter criterion and other…

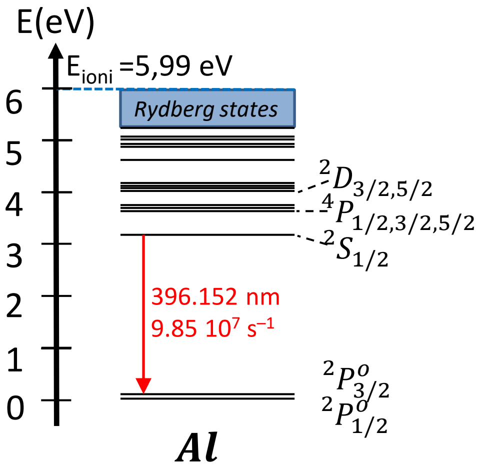
Competition collisions (e-) ↔ radiation

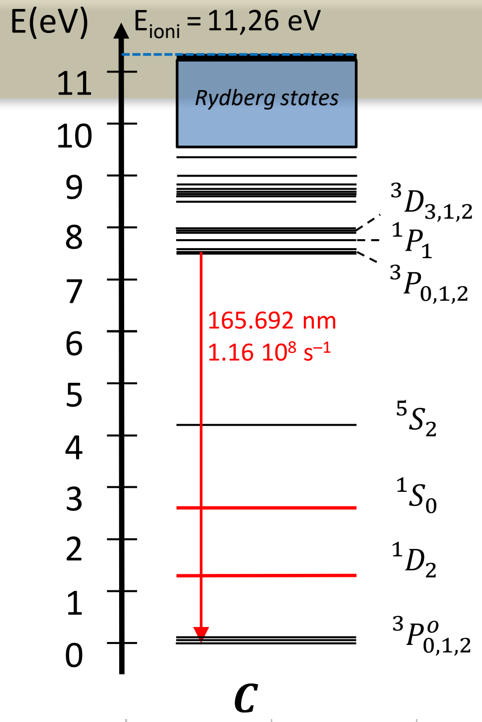
McWhirter criterion (1965)

$$n_{e,m^{-3}} > 1{,}6 \times 10^{18} \sqrt{T_{e,K}} \left(\Delta E_{ji,eV}\right)^3_{max}$$

Griem criterion (1963)

$$n_{e,m^{-3}} > 10^{17} \sqrt{T_{e,K}} \left(\Delta E_{ji,eV}\right)^3_{rés} Z^7$$



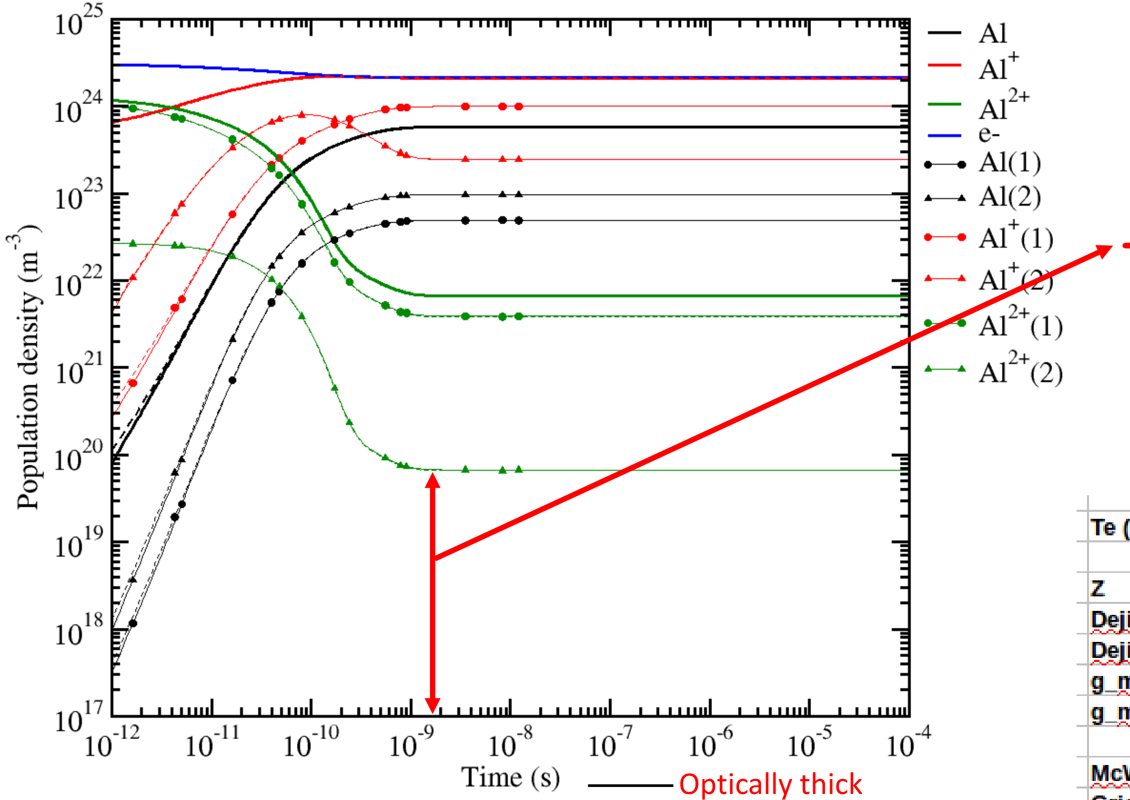


Drawin criterion (1969)

$$n_{e,m^{-3}} > 9{,}1 \times 10^{16} \sqrt{T_{e,K}} \left(\Delta E_{ji,eV}\right)^3_{max} \frac{g_{max}}{g_{min}} \times \begin{cases} 3\left[\dfrac{\left(\Delta E_{ji,eV}\right)_{max}}{T_{e,eV}}\right]^{0{,}483} & neutres \\ \left[\dfrac{\left(\Delta E_{ji,eV}\right)_{max}}{T_{e,eV}}\right]^{0{,}269} & ions \end{cases}$$

Te (K)	15000			
	Al	Al+	C	C+
Z	1	2	1	2
Deji,eV_max	3.13	4.64	3.30	5.32
Deji,eV_rés	3.14	7.42	7.48	9.29
g_max	2	1	1	2
g_min	4	1	5	4
McWhirter	6.01E+21	1.96E+22	7.04E+21	2.95E+22
Griem	3.79E+20	6.40E+23	5.13E+21	1.26E+24
Drawin	7.86E+20	1.57E+21	3.78E+20	1.23E+21

McWhirter criterion and other…

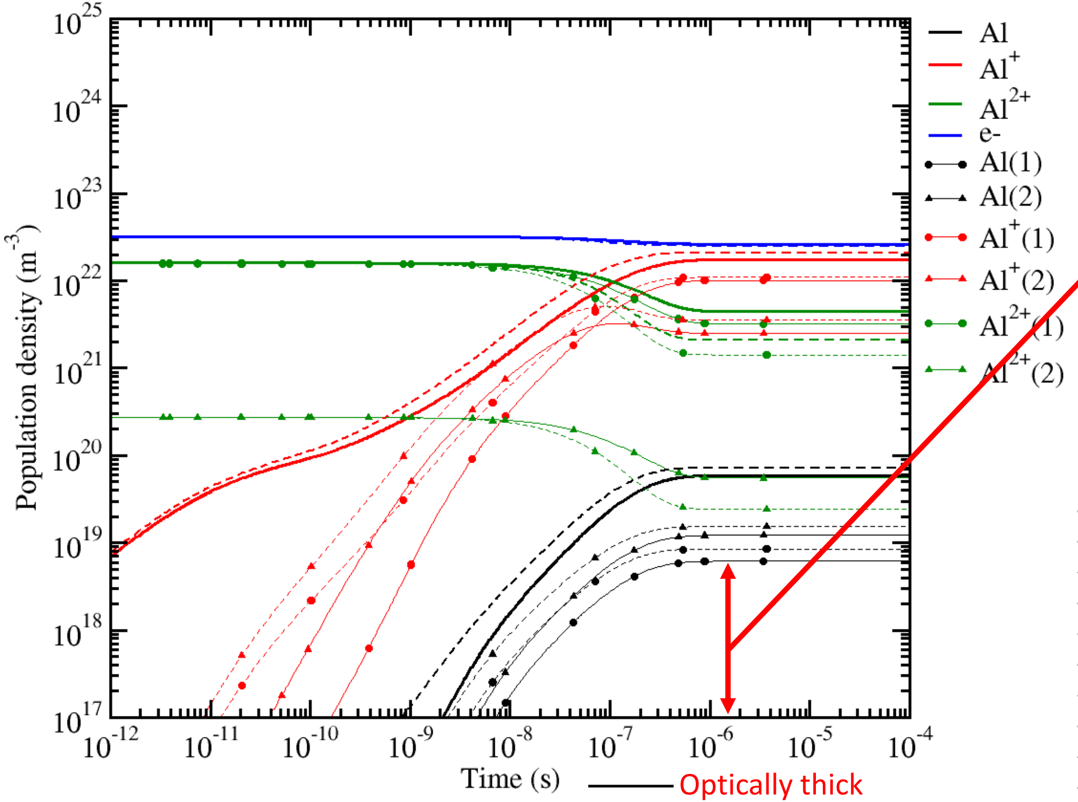


$p = 10^6 Pa, T_e = 15000\ K$

1. Some $10^{-9}s$: steady state
2. **No** radiation influence: final **equilibrium state**

Te (K)	15000	
	Al	Al+
Z	1	2
Deji,eV_max	3.13	4.64
Deji,eV_rés	3.14	7.42
g_max	2	1
g_min	4	1
McWhirter	6.01E+21	1.96E+22
Griem	3.79E+20	6.40E+23
Drawin	7.86E+20	1.57E+21

McWhirter criterion and other…



Optically thick

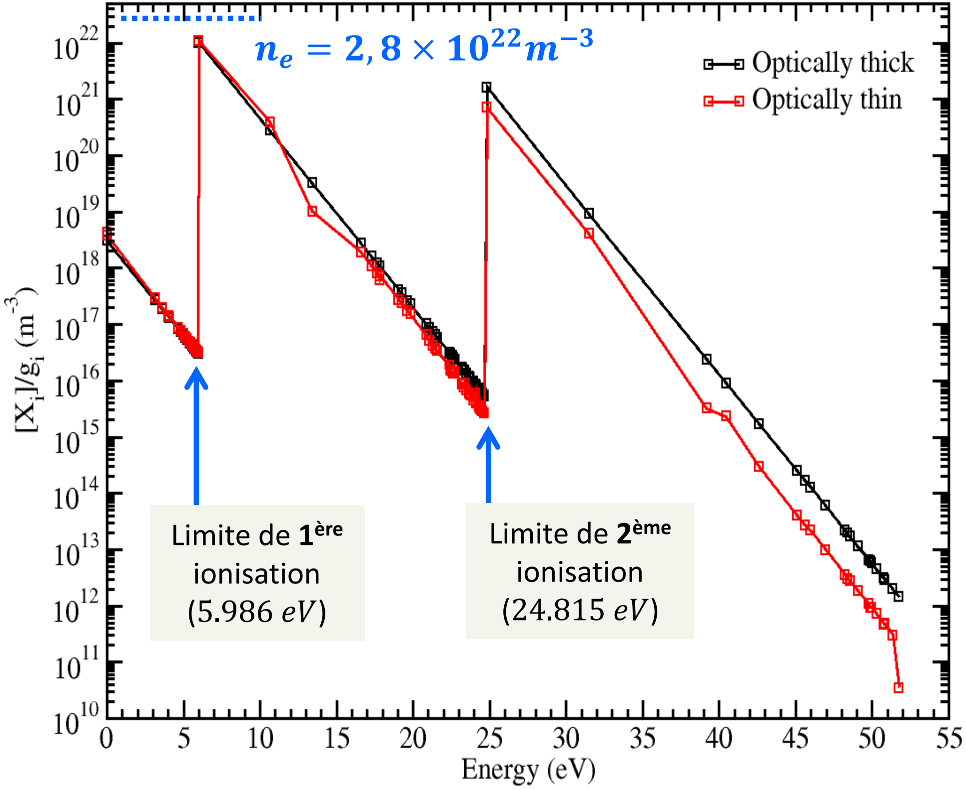
Optically thin

$p = 10^4 Pa, T_e = 15000\ K$

1. Some $10^{-6} s$: steady state
2. **Strong** radiation influence: final **non** equilibrium state

Te (K)	15000	
	Al	Al+
Z	1	2
Deji,eV_max	3.13	4.64
Deji,eV_rés	3.14	7.42
g_max	2	1
g_min	4	1
McWhirter	6.01E+21	1.96E+22
Griem	3.79E+20	6.40E+23
Drawin	7.86E+20	1.57E+21

McWhirter criterion and other…



$p = 10^4 Pa, T_e = 15000\ K, t = 1\ \mu s$

McWhirter criterion

$$n_{e,m^{-3}} > 1.6 \times 10^{18} \sqrt{T_{e,K}} (\Delta E_{ji,eV})^3_{max}$$

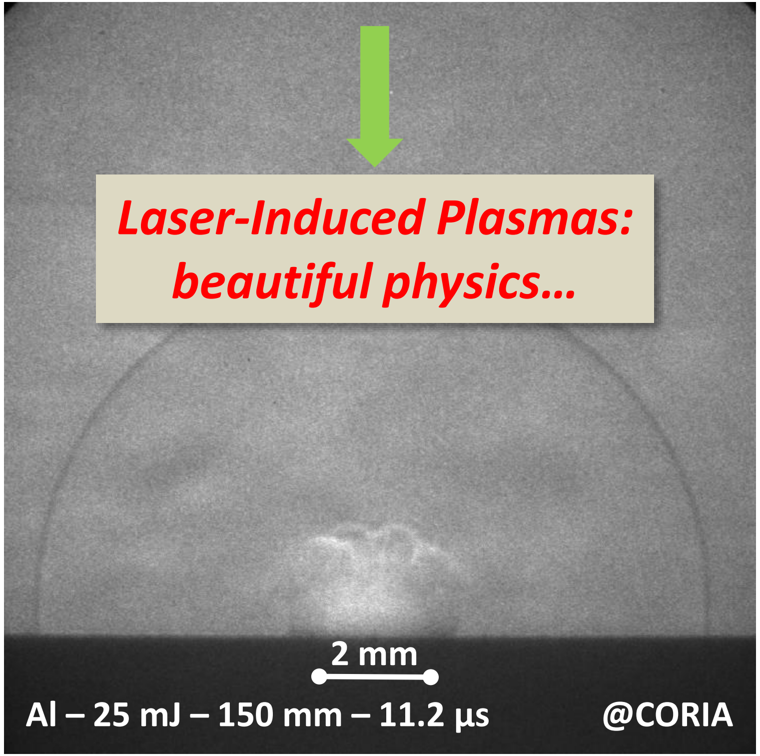
$n_e > 6,01 \times 10^{21} m^{-3}$ for Al

$n_e > 1,96 \times 10^{22} m^{-3}$ for Al^+

Criterion seems to be wrong

$T_{exc} \neq T_e$

Te (K)	15000	
	Al	Al+
Z	1	2
Deji,eV_max	3.13	4.64
Deji,eV_rés	3.14	7.42
g_max	2	1
g_min	4	1
McWhirter	6.01E+21	1.96E+22
Griem	3.79E+20	6.40E+23
Drawin	7.86E+20	1.57E+21


Laser-Induced Plasmas:
beautiful physics…
2 mm
Al – 25 mJ – 150 mm – 11.2 µs
@CORIA