The role of ablation mechanisms in the onset of ns-laser induced plasma formation

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Outline

1. Motivation: “LA-ICP-MS”

2. Case: Pulsed ns-laser ablation of copper in argon

3. Model

4. Results

   - Target: Mass removal

   - Breakdown: “0D“ Collisional Radiative Model

   - Plume: 1D Partial LTE Model

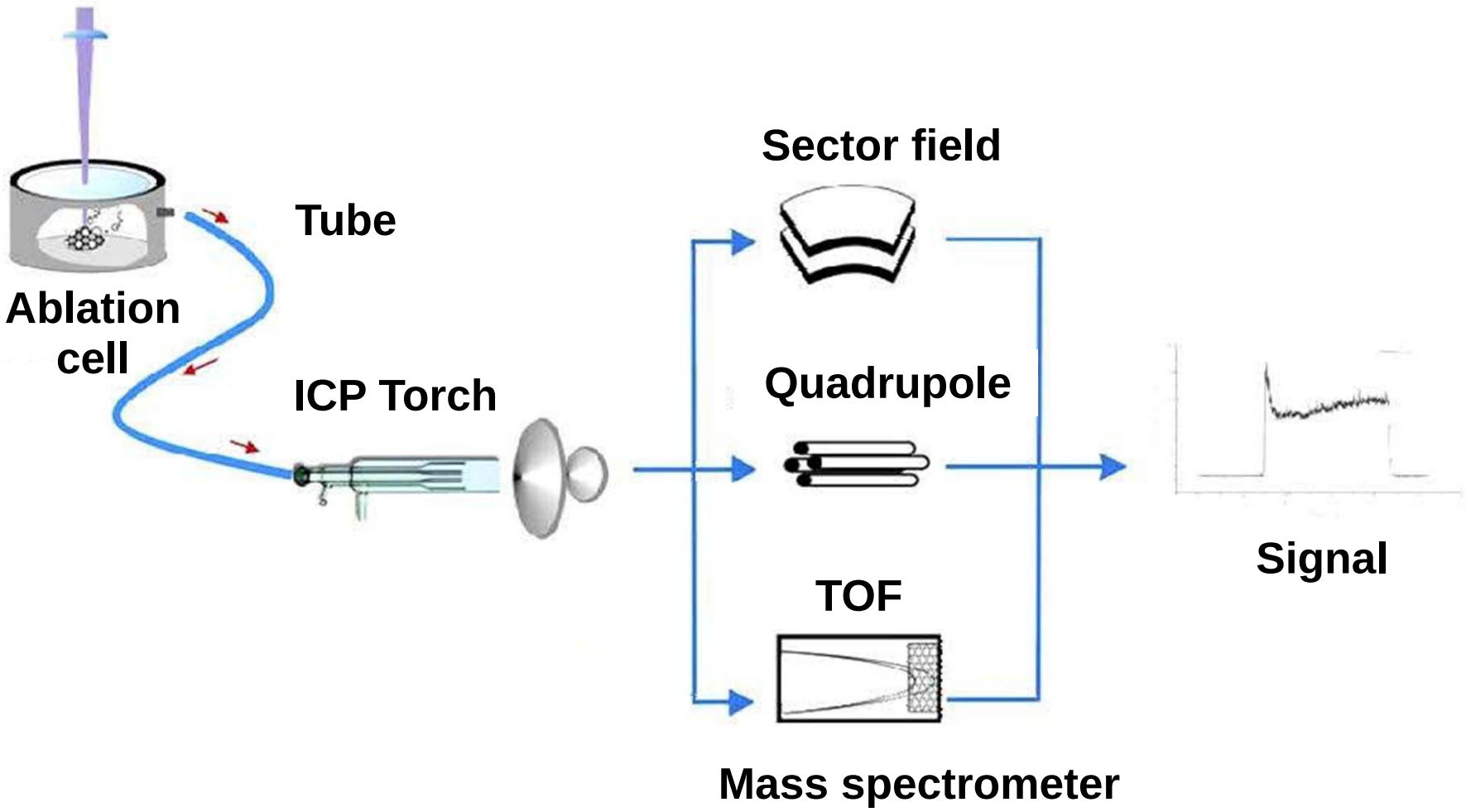
5. Experiment

6. Conclusion

Motivation: “LA-ICP-MS”

“LA-ICP-MS”:

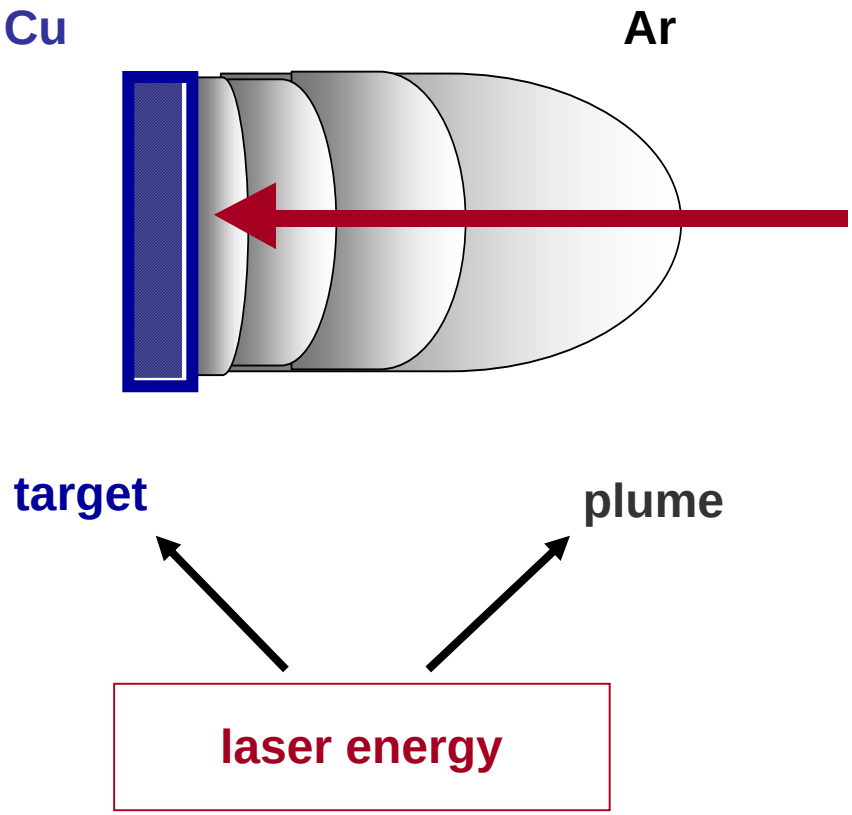
- **Laser Ablation Inductively Coupled Plasma Mass Spectrometry**
- **Determination trace-elements (Parts Per Billion)**



Case
Pulsed ns-laser ablation of Cu in Ar

What?

- 6 ns-laser pulse
- λ = 532 nm
- F = 1-10 J/cm^2
- Cu target
- 1 atm Ar



Why?

How does laser energy distribute between target and plume?

How?

1. Model laser- material interaction
2. Compare with measured transmission profiles
3. Compare with measured crater depths

Model

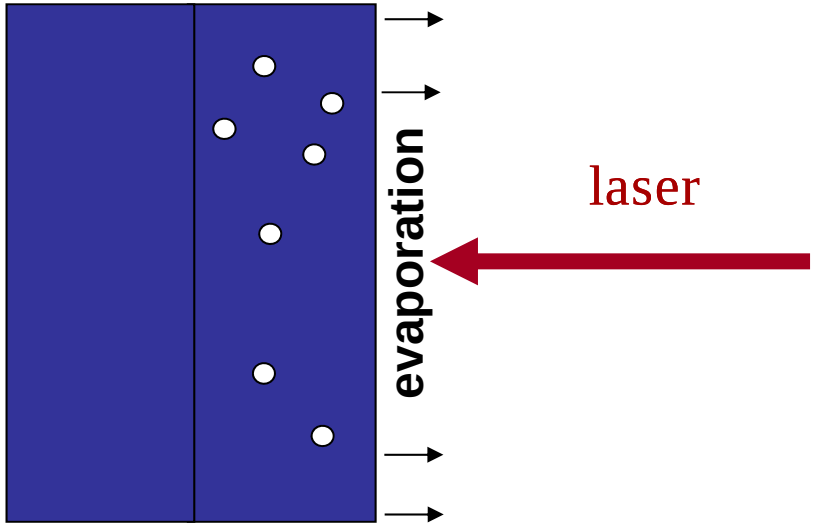
Coupled story: target ⟷ plume

1. Ablation mechanisms
2. Target properties
3. Plasma formation
4. Plume expansion

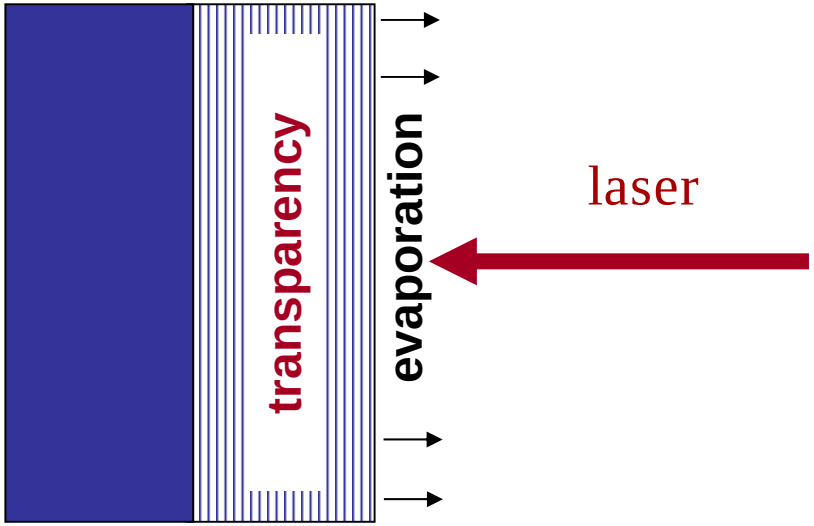
Target
Modelling approach

2 models:

Full model



"Evaporation-only" model

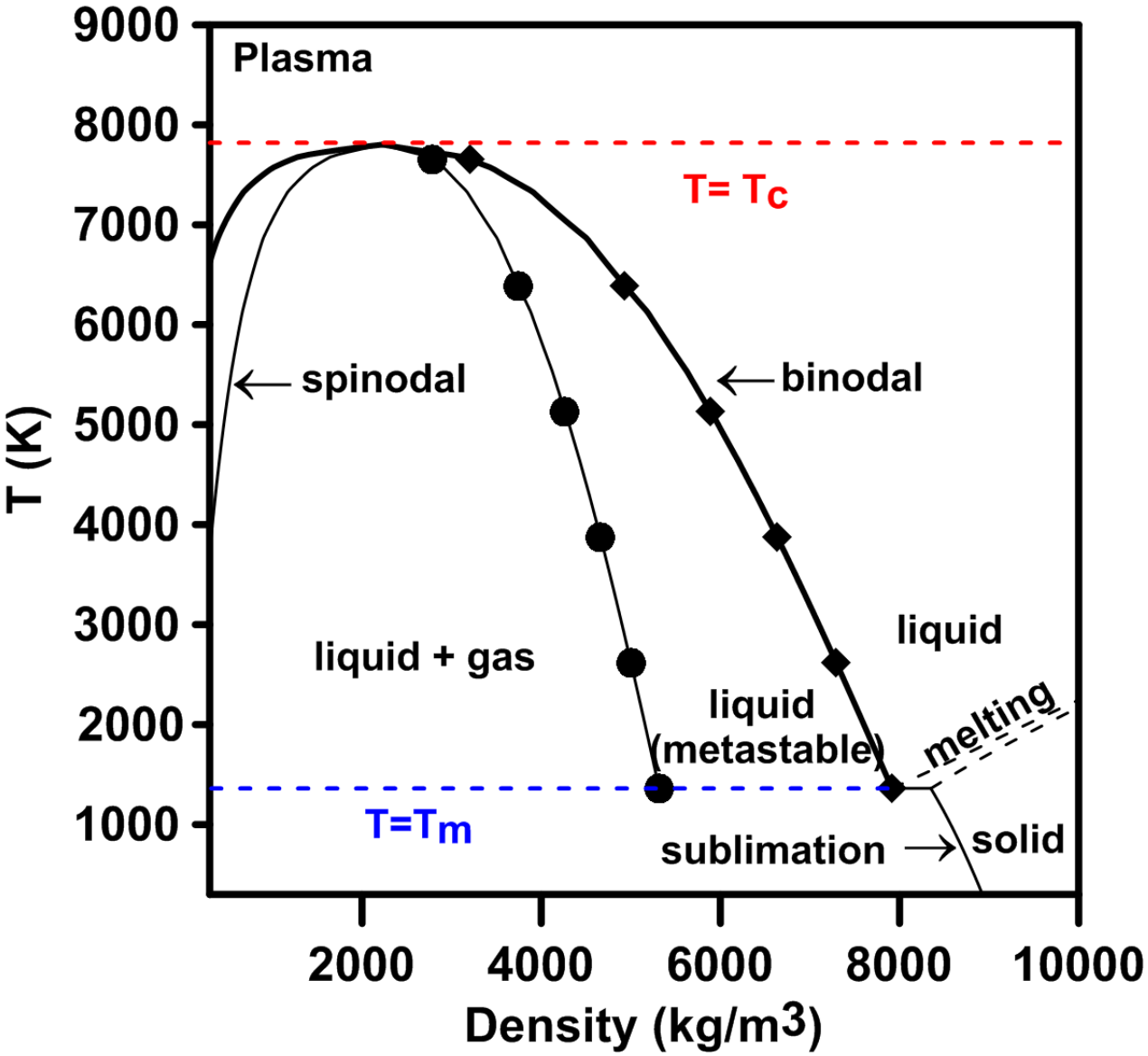


- evaporation / condensation
- volumetric mass removal

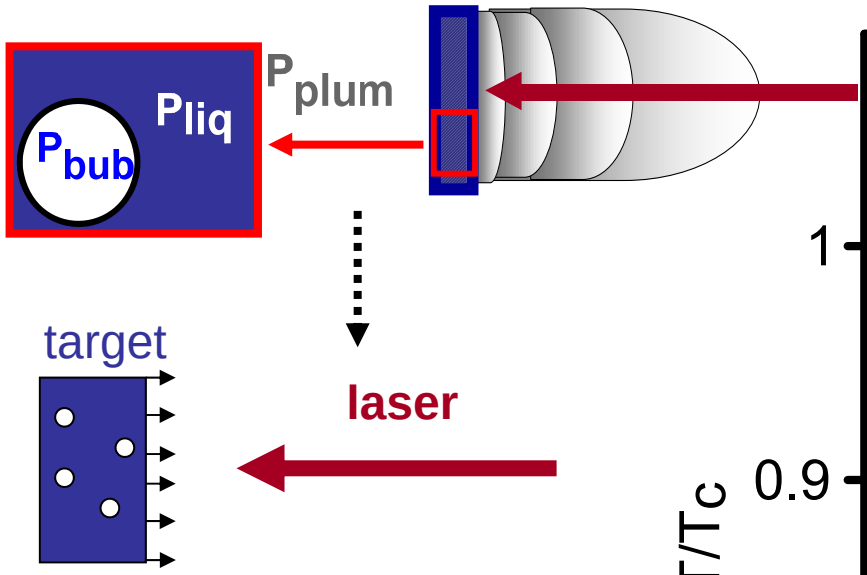
- transparency front [1]
- evaporation/condensation

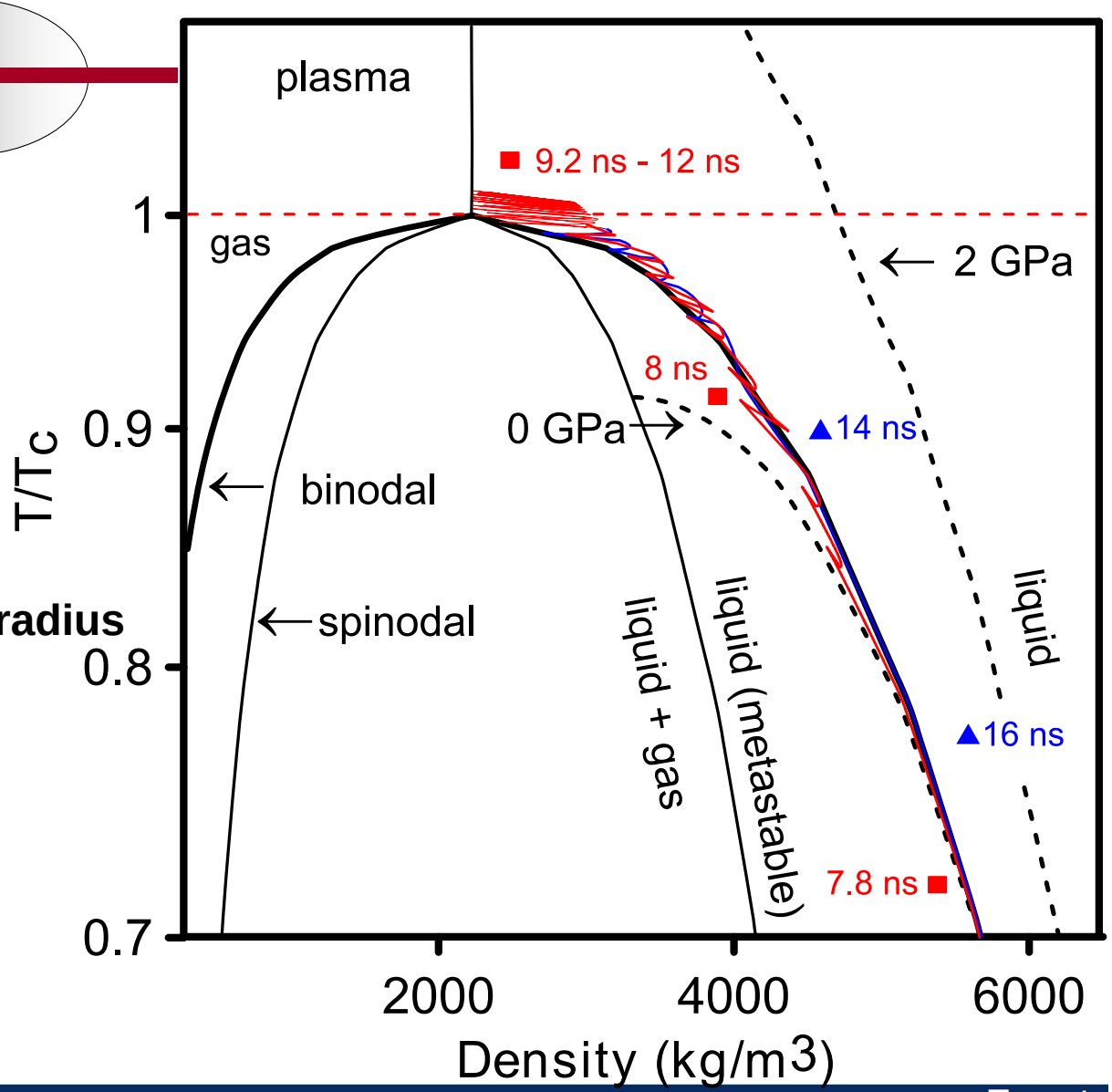
[1] V. Batanov, F. Bunkin, A. Prokhorov, and V. Fedorov, Sov. Phys. JETP 36, 311 (1973)

Target
Phase diagram copper[2]



Phase diagram
Evolution target surface cell





Bubble formation → Critical radius

$$R_{crit} = \frac{2\sigma(T)}{(P_b(T) - P_l(T))}$$

Bubble formation[3] : $\frac{P_l}{P_b} < 1$

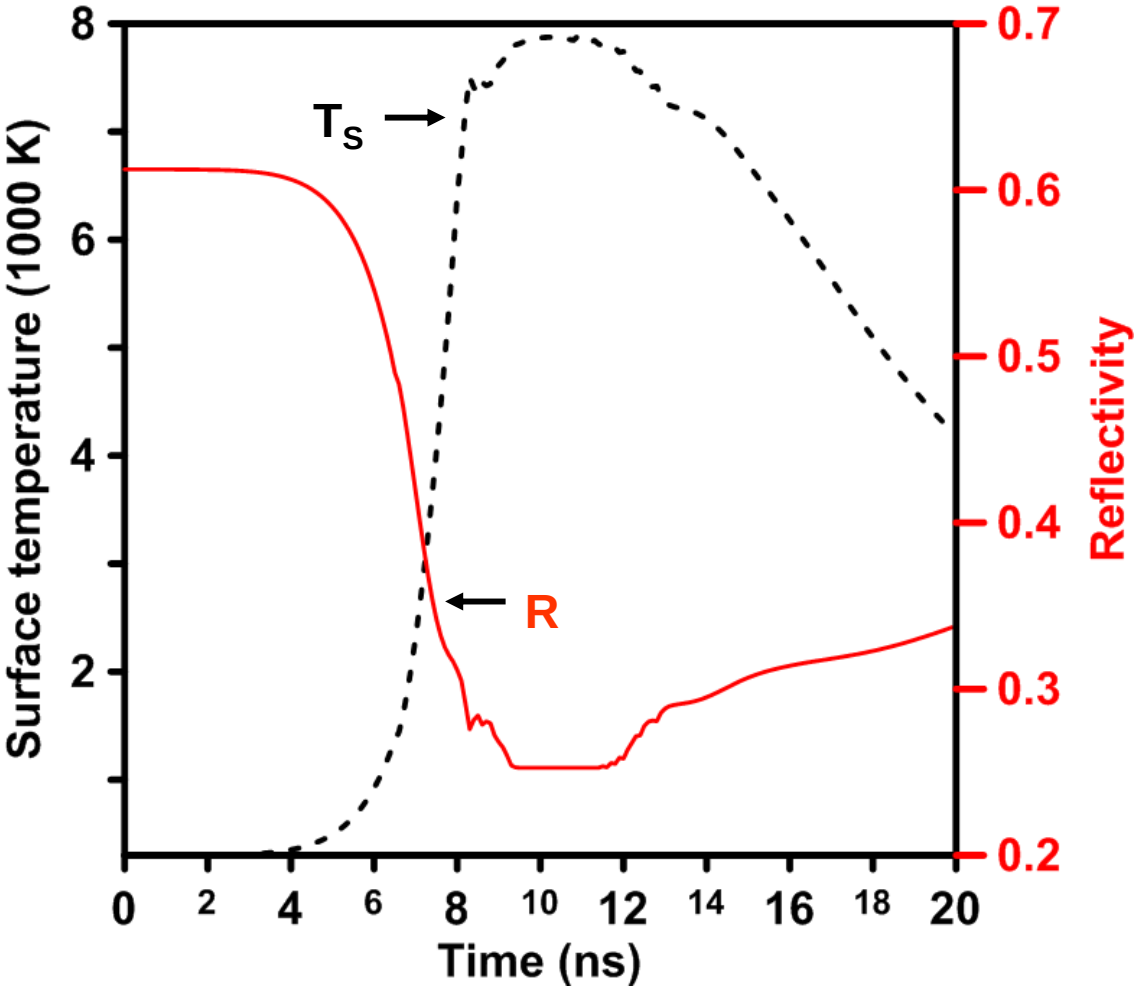
[3] N.M. Bulgakova et al., Appl. Surf. Sci., 257, 24, 10876-10882 (2011)

Target surface Reflectivity

1. temperature ↑ reflectivity R ↓

2. T~=Tcrit: not transparent

3. temperature ↓ reflectivity R ↑



Target surface
Intensity vs Temperature

Observations

1. intensity ↑ , temperature ↑

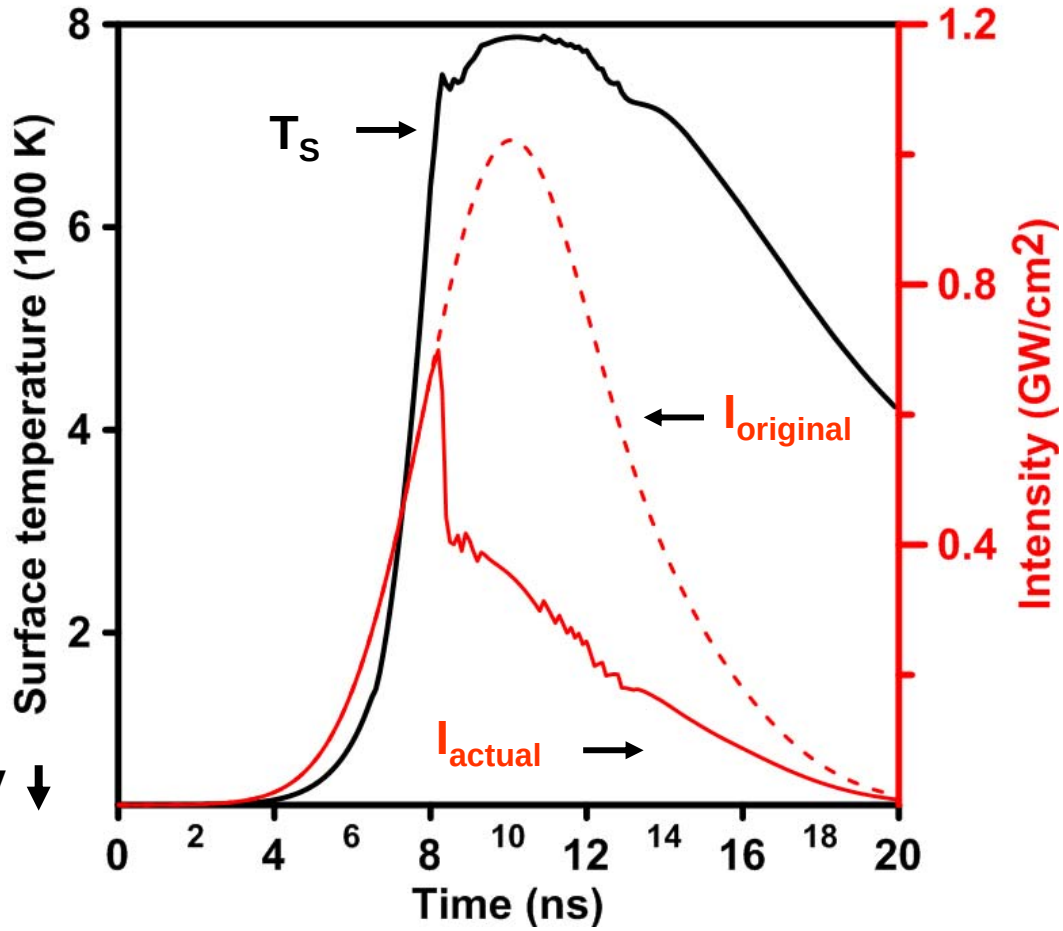
2. plasma shielding

intensity ↓ , temperature ↓

3. Volumetric ablation → oscillations

e.g. plume expands , plume density ↓

intensity ↑ , temperature ↑



Collisional radiative model

1. Collisional processes

excitation deexcitation

$Cu^* + e \leftrightarrow Cu^{**} + e$

ionization recombination

$Cu^* + e \leftrightarrow Cu^{**} + e$

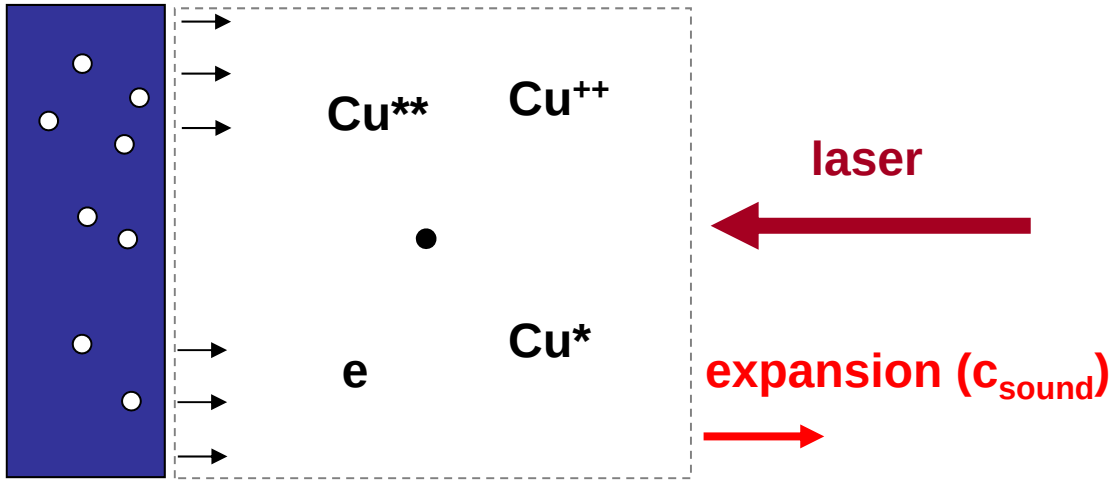
2. Radiative processes

excitation decay

$Cu^* + h\nu_{laser} \leftrightarrow Cu^{**} + e$

ionization recombination

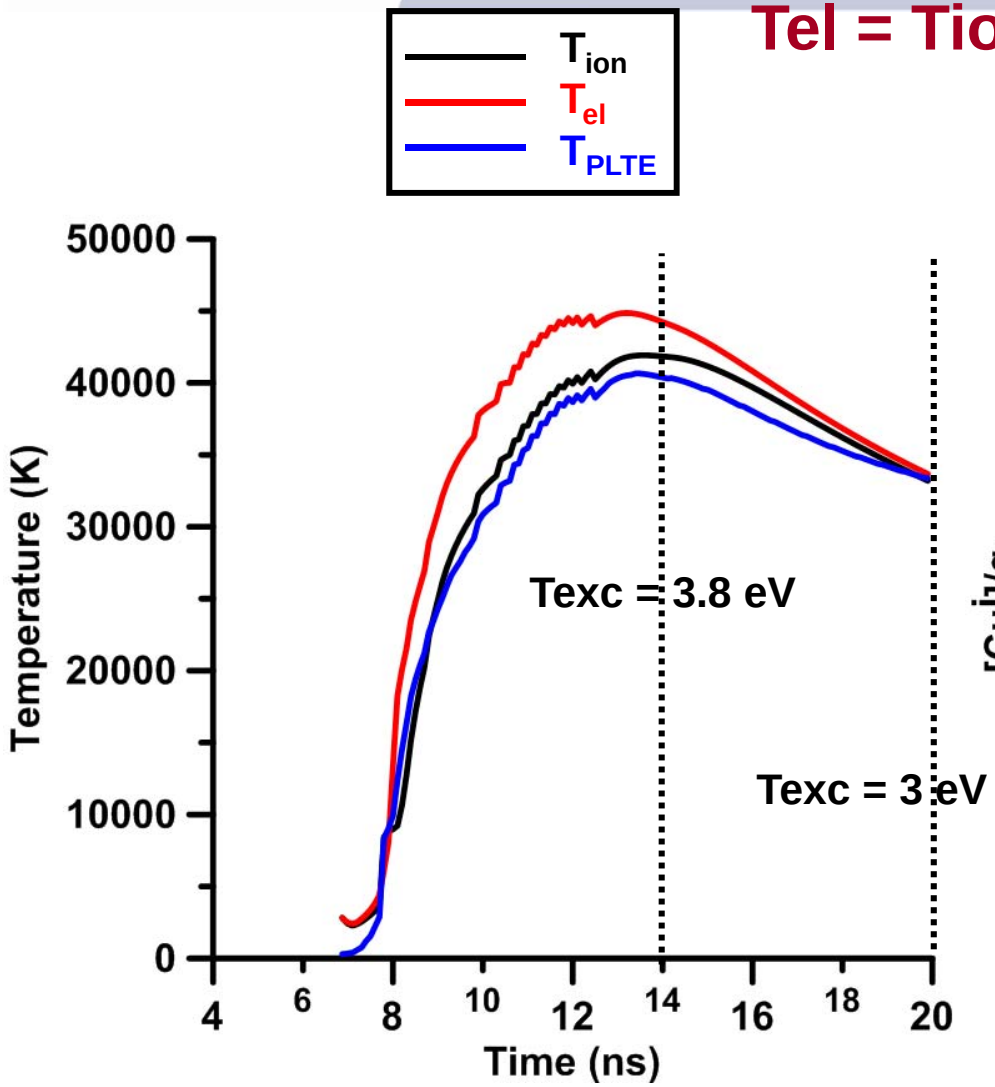
$Cu^* + n\, h\nu_{laser} \leftrightarrow Cu^{**} + e$ (n=1-3)

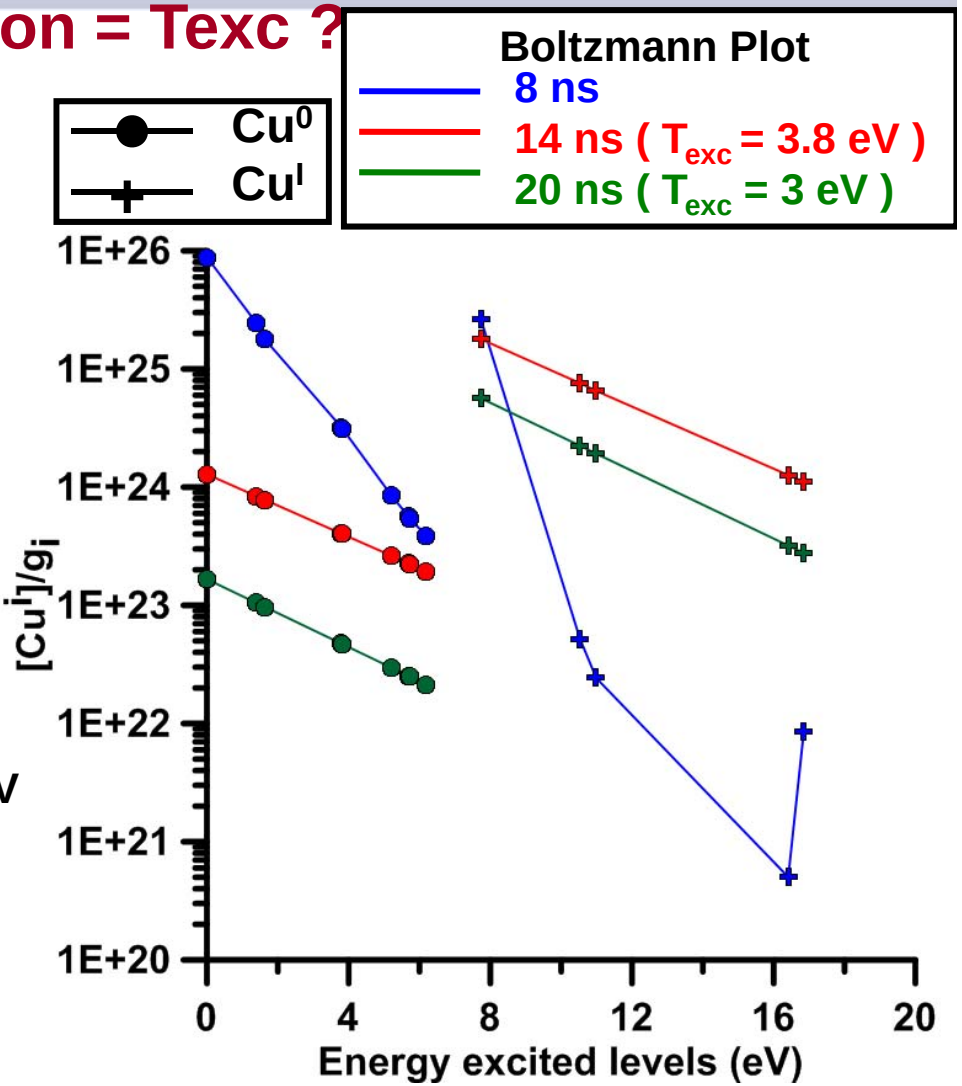


Rate equations: $$\frac{\partial N_{M^+}}{\partial t} = S_{M^+}$$

Collisional Radiative Model
Partial Local Thermodynamic Equilibrium?

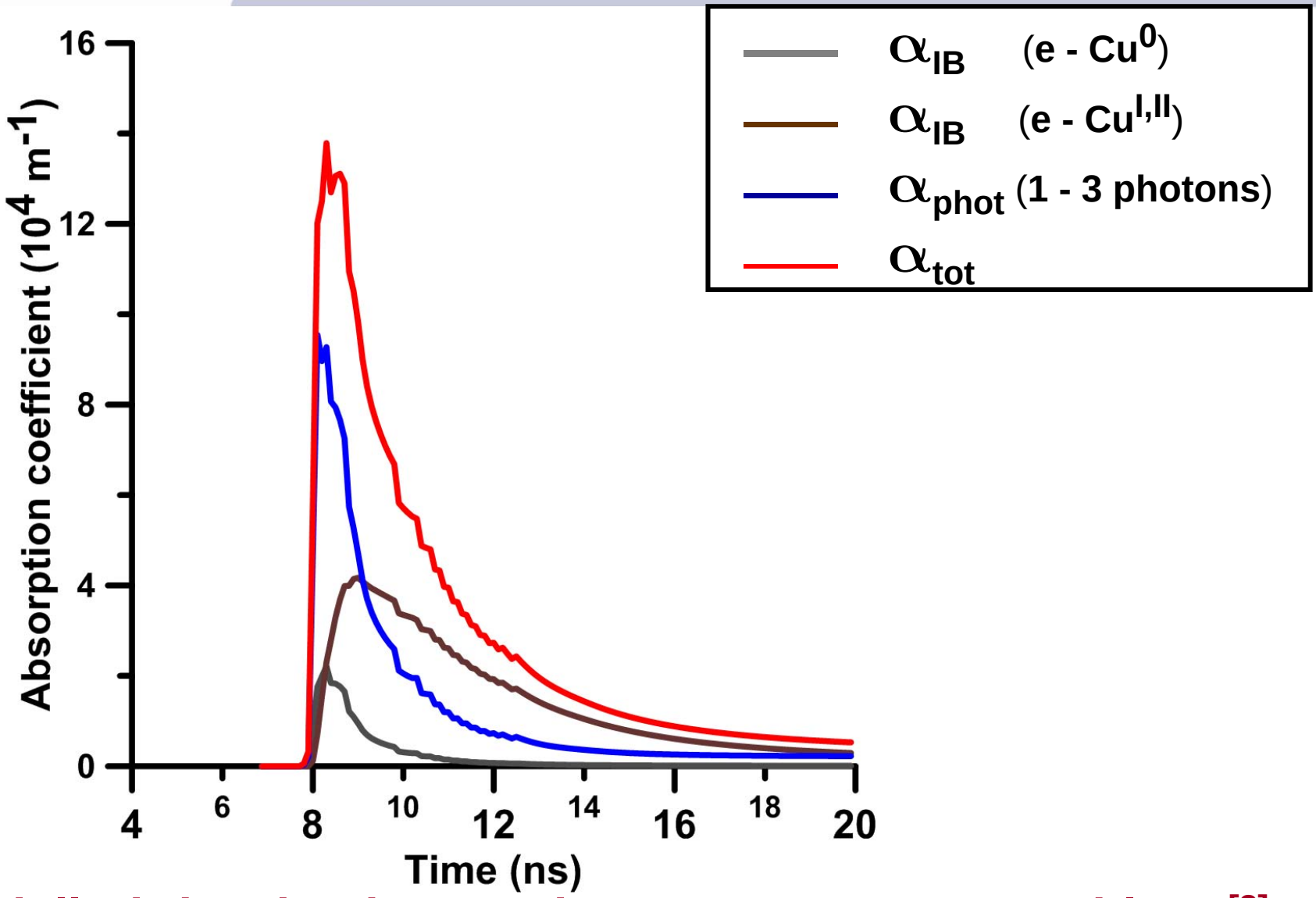
Tel = Tion = Texc ?





Laser: 532 nm - 6ns FWHM - 6 J/cm²
Gas: argon

Collisional Radiative Model
Absorption mechanisms



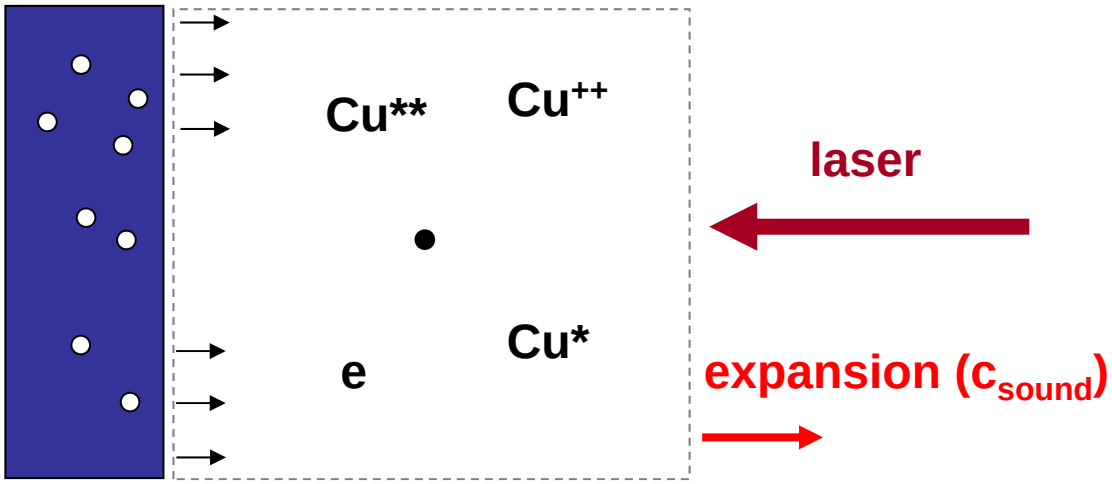
Initially (M)PI dominates, then Inverse Bremsstrahlung[3]

[3] V. Morel, A. Bultel, and B. Cheron, Spectrochim. Acta,12 Part B 65, 830 (2010).

Model

“0 D” Collisional Radiative Model

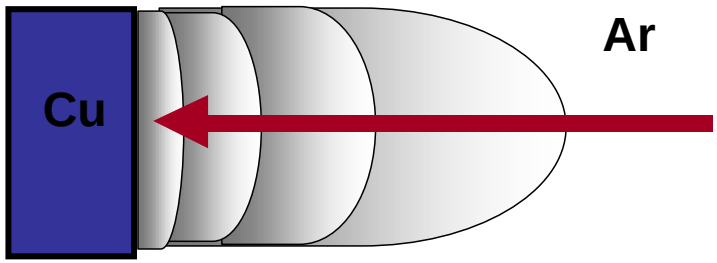
- Breakdown
- Initial expansion



Iterate

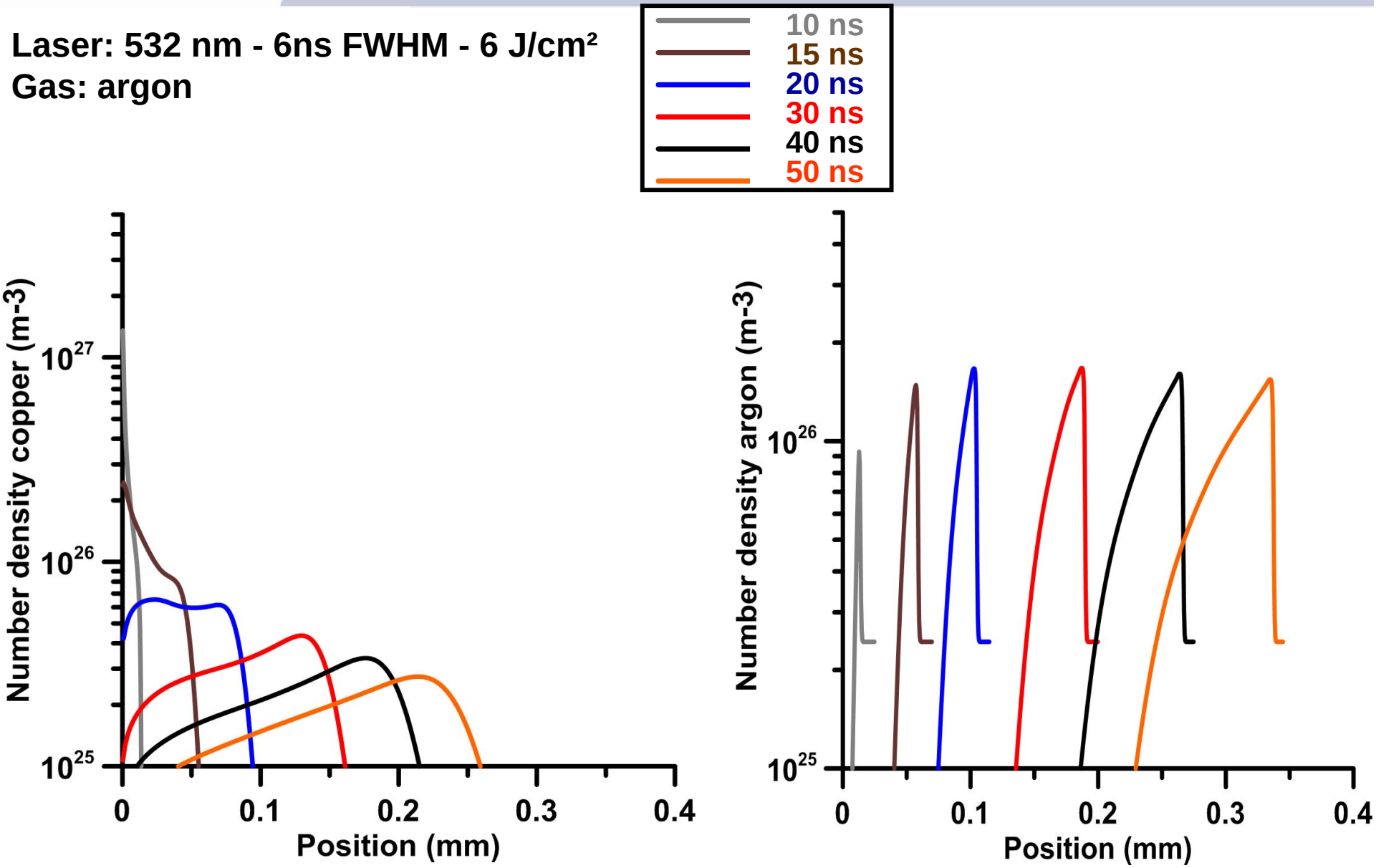
1D multiphase PLTE Model

- Plume expansion
- Mc Wirther criterion
- Saha equation



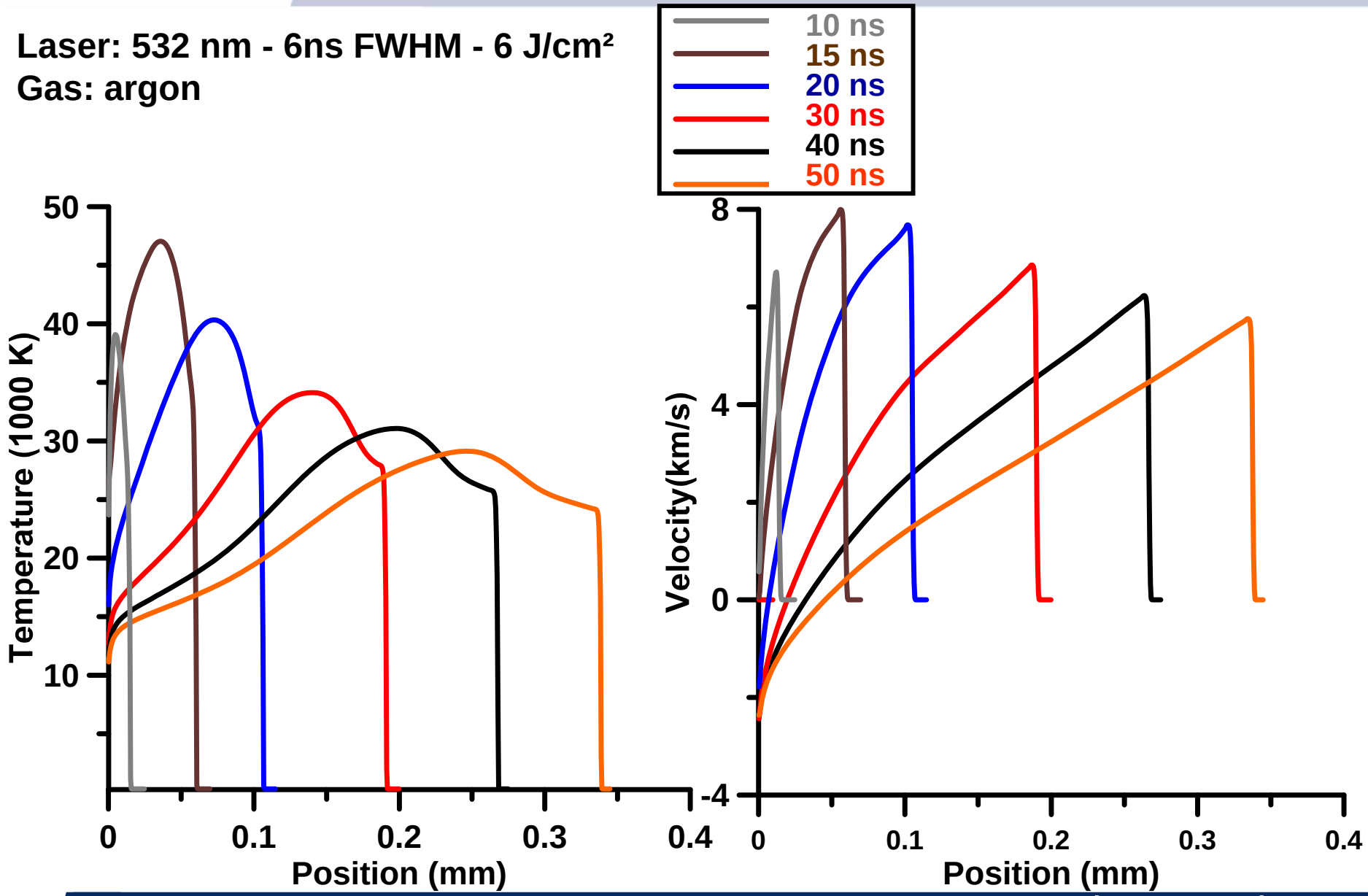
Plume
Snowplow effect

Laser: 532 nm - 6ns FWHM - 6 J/cm²
Gas: argon

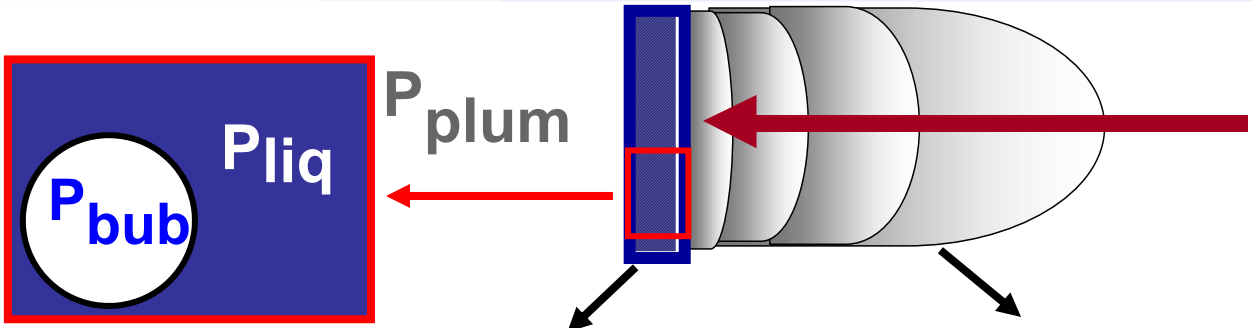


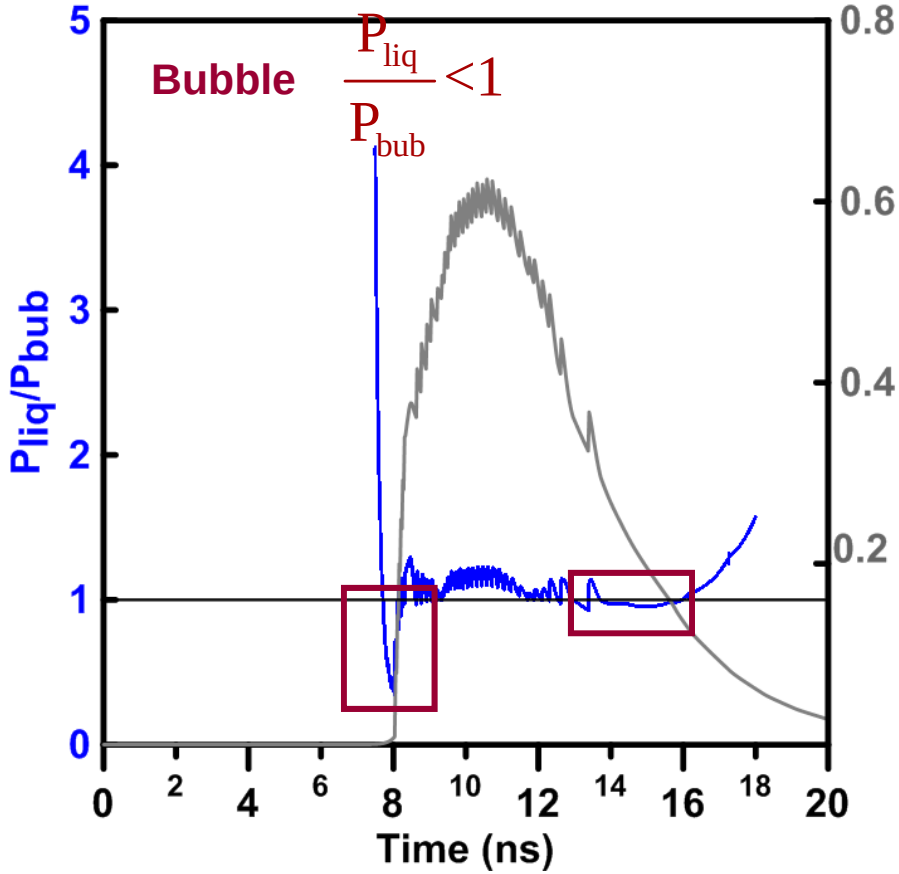
Plume
Temperature and Velocity

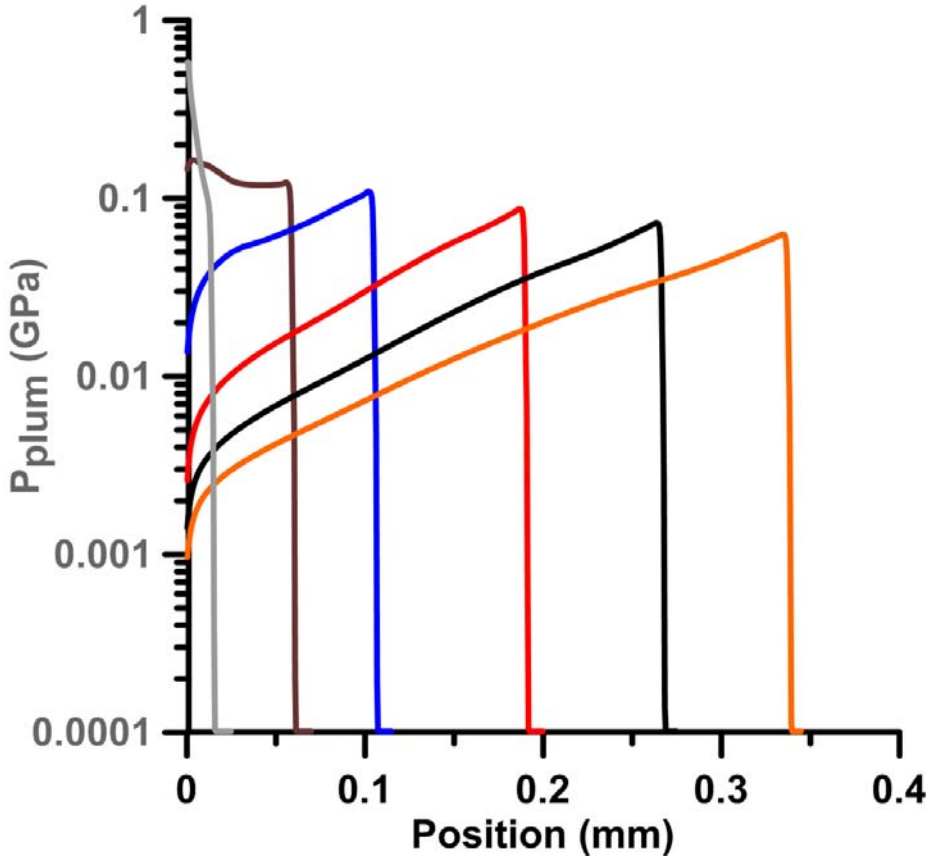
Laser: 532 nm - 6ns FWHM - 6 J/cm²
Gas: argon



Target and Plume
Interplay of diffent pressures





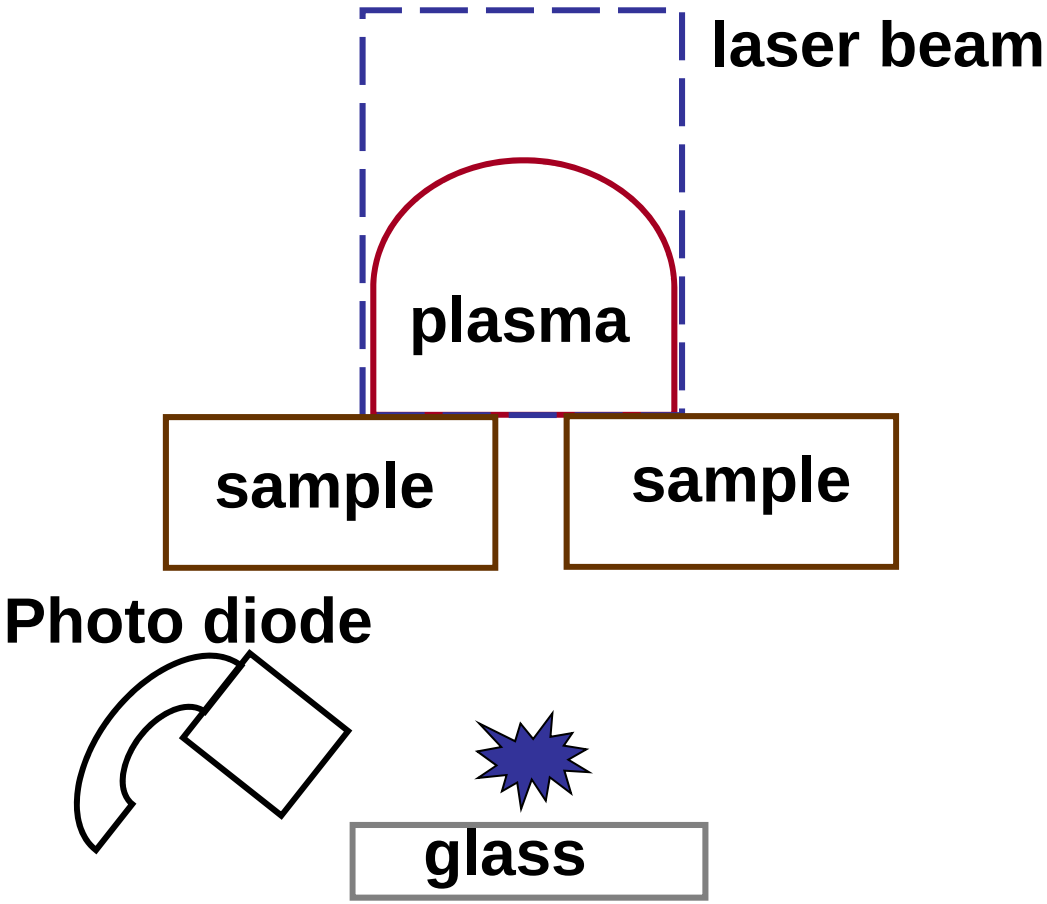


Experiment

1. Experimental setup
2. Temporal intensities
3. Transmission study
4. Ablation depth measurements

Experimental setup

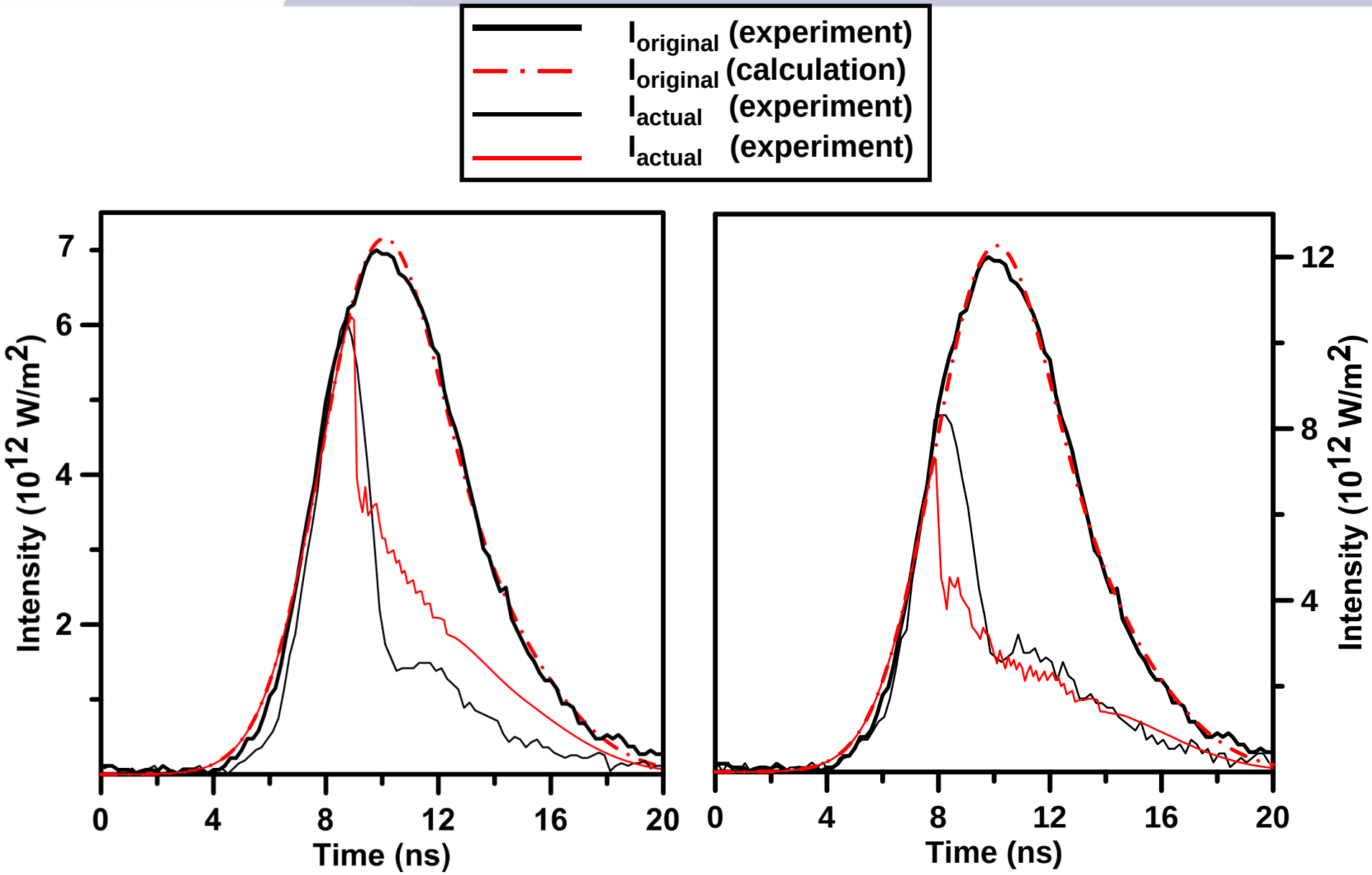
Transmission



Ablation depth

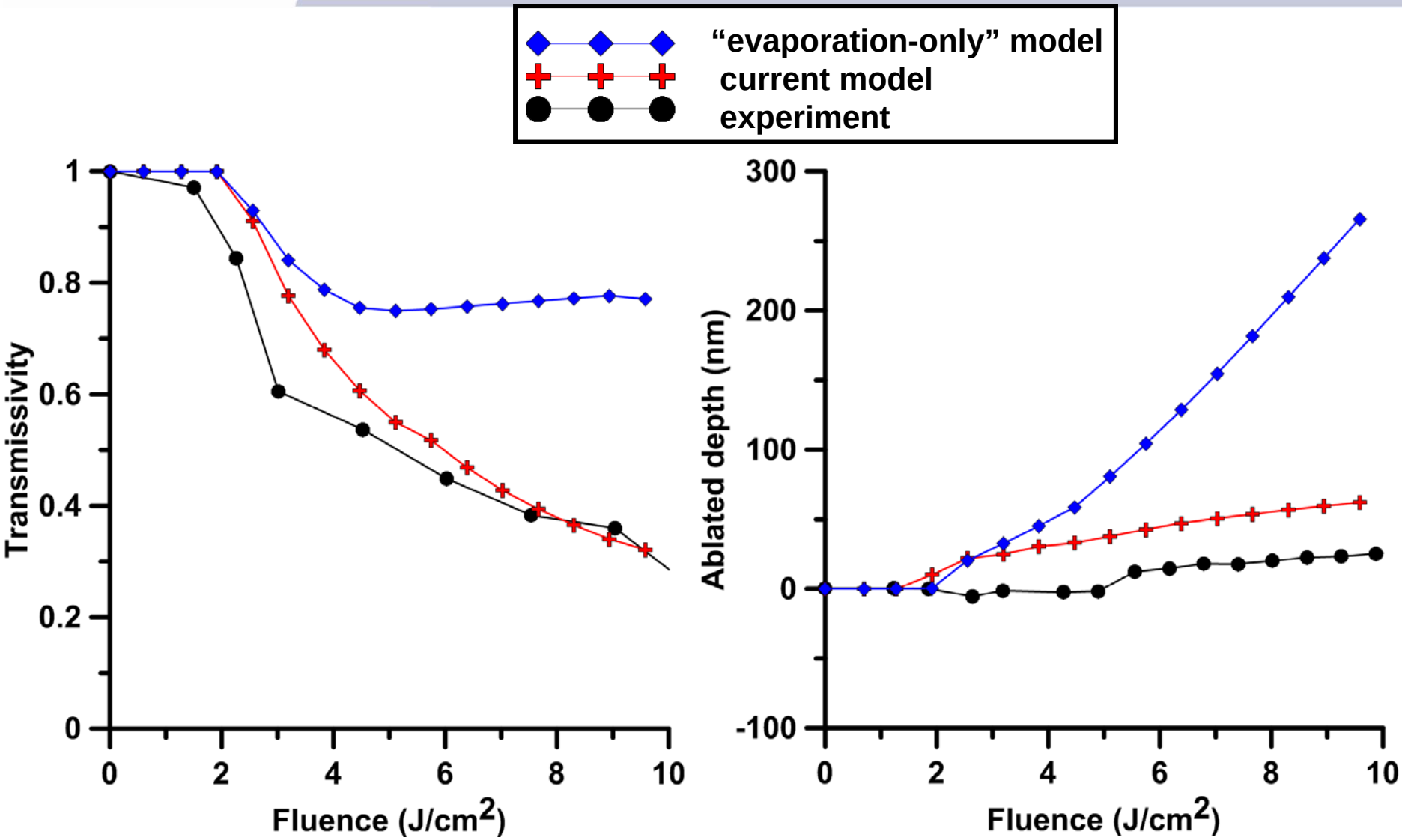
Profilometry

Temporal intensity patterns[4]



[4] experimental data dissertation G.Clair, Université. D'Aix-Marseille II –CEA, 2011

Transmission and ablation depth[4]



"Evaporation-only model" does not work !

[4] experimental data dissertation G.Clair, Université. D'Aix-Marseille II –CEA, 2011

Conclusion

Study of ns pulsed laser ablation of copper

- Tightly coupled processes: target ⟷ plume
- Surface and volumetric mass removal
- Volumetric ablation dominates ⟶ plasma formation
- Initial expansion ⟶ collisional radiative model
- Onset breakdown through 1, 2 and 3 - Photon Ionization
- Laser pulse ends ⟶ partial LTE reasonable

References

Study of ns pulsed laser ablation of copper

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