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Hydrodynamic modeling of Laser Ablation

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Outline

- Laser ablation process
- Features to be modeled
- Process modeling highlights:
  * target heating / melting / vaporization
  * Knudsen Layer
  * vapor flow / plume expansion
- Numerical simulations
- conclusion

Laser Ablation

Process of removing material using a pulsed laser beam

Some application areas

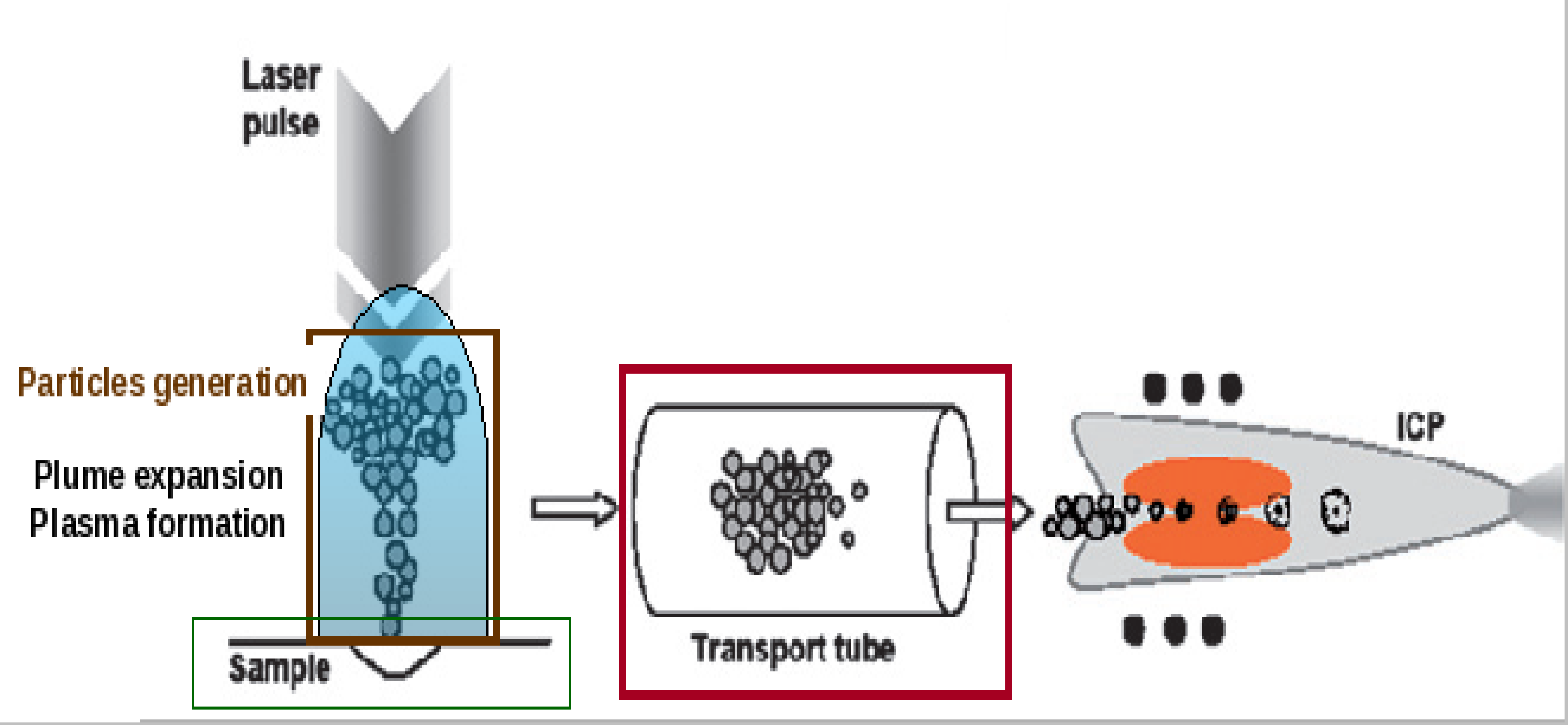
- Cutting, machining,
- Drilling, engraving,
- Bonding, coating,
- Surface cleaning,
- Surgery,dentistry,
- Nanomaterials, ...

- Laser-induced breakdown spectroscopy
- LA - Inductively coupled plasma mass spectrometry (ICP-MS)

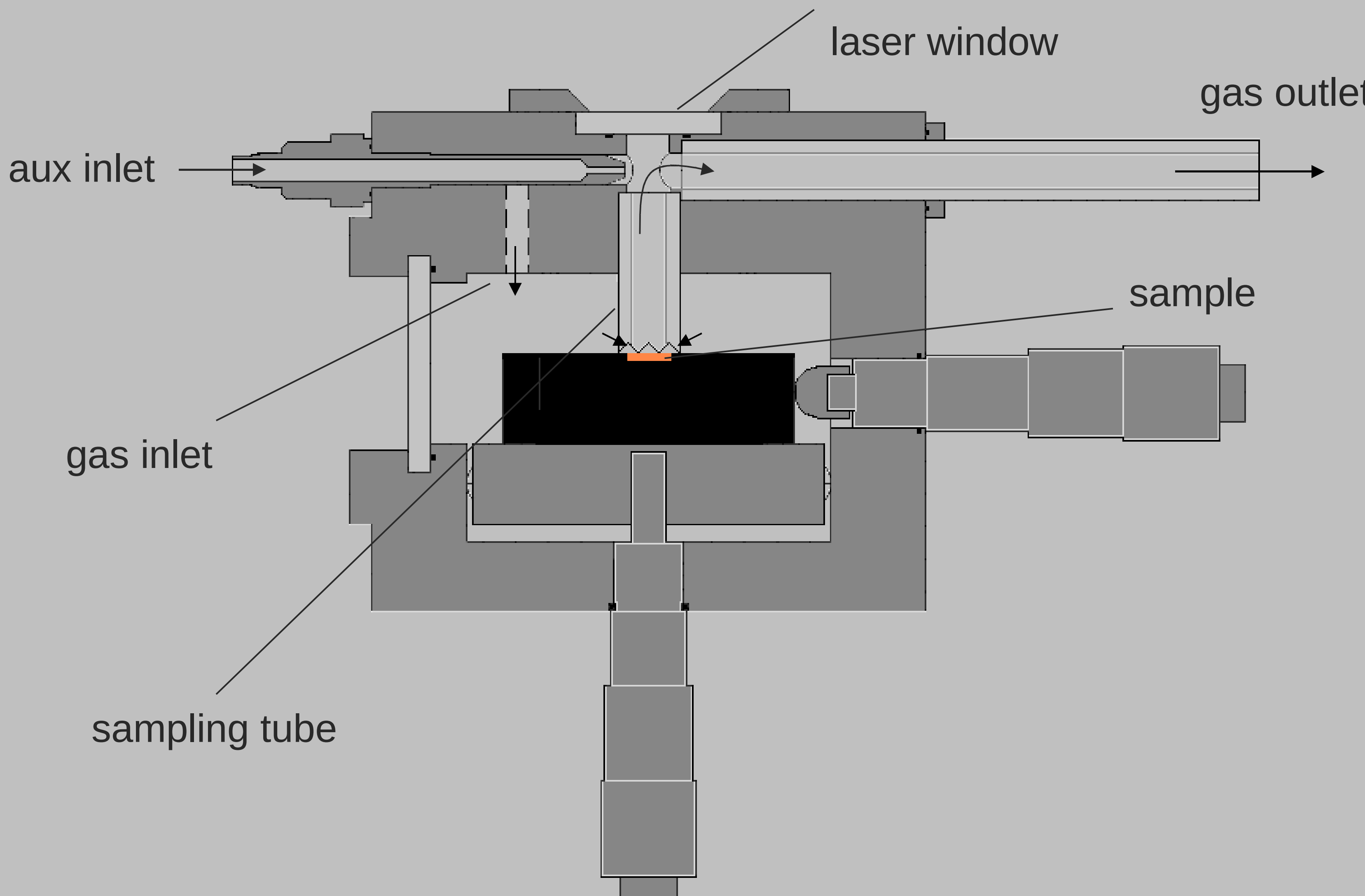
detect trace/ultra-trace elements,
as low as 2 parts per 100 billion!
Widely used in chemistry,medicine,
archaeology, geology, materials sci.

Laser Ablation - Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS)

Ultrasensitive, multi-element, essentially non-destructive (only ngrams)



Laser ablation cell



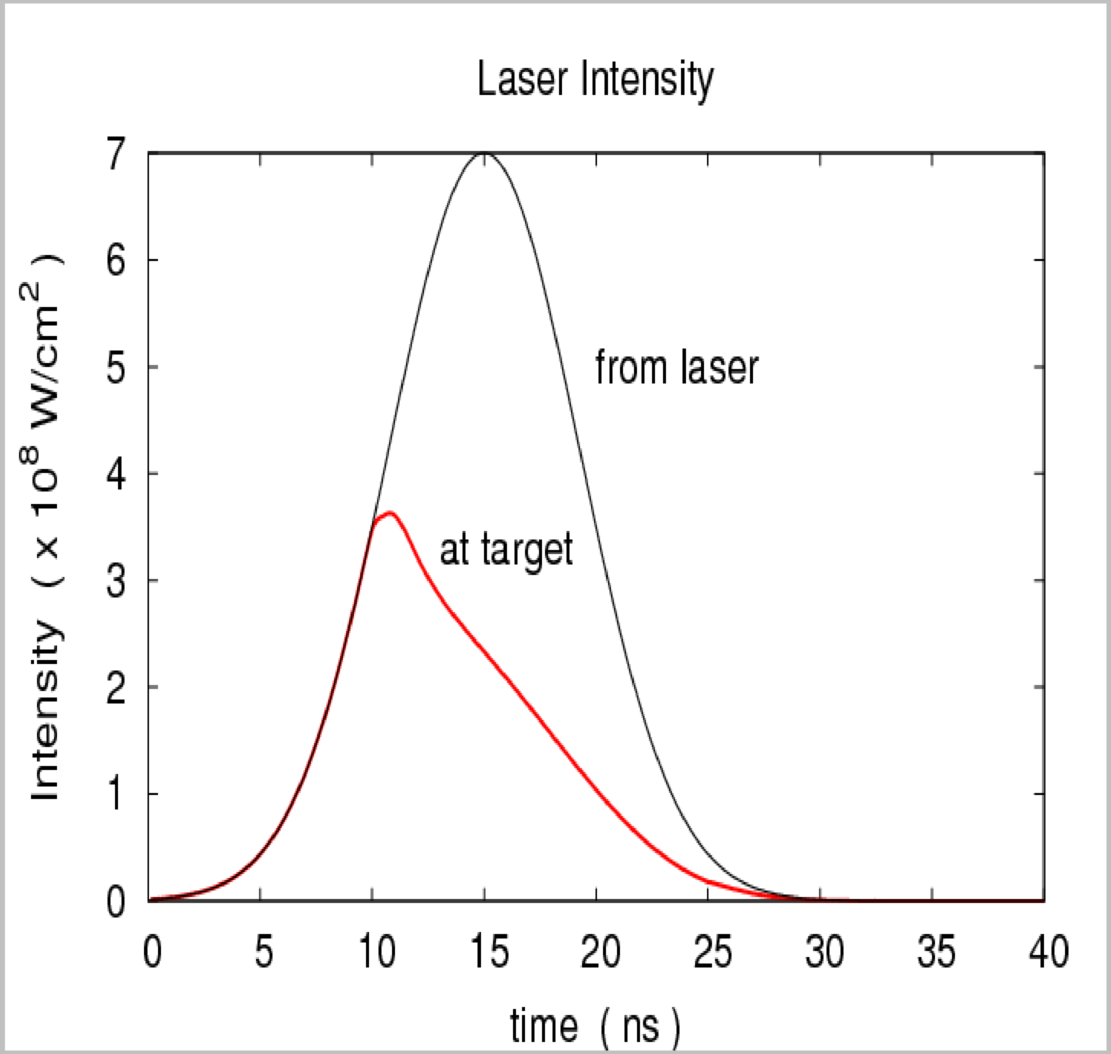
Ref: C. C. Garcia et all, SAB, 62 (2007) 13-17.

Nanosecond laser ablation

Test conditions

Laser intensity

- 8 ns (fwhm) pulse
- λ = 266 nm
- I =10^9 W/cm^2
- Cu target
- 1 atm He gas



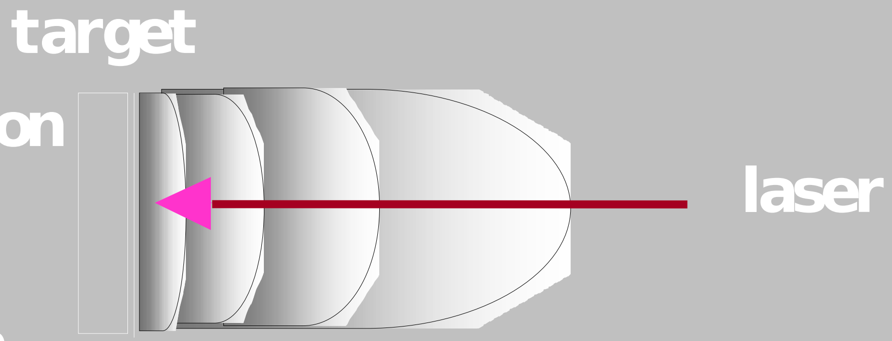
Nanosecond laser ablation

- solid target heats up, a thin layer melts (T_m=1358K)
- melt vaporizes at high T, creating a plume of vapor
- which rapidly expands (and cools somewhat)
- electrons/ions dissociate creating plasma

→ in target : heat conduction / melting / vaporization coupled to electric field (thermionic emission)

→ over target: Knudsen transition layer

→ in plume/plasma: expansion/recondensation,laser absorption



Heat conduction
→ T ↑
→ melting
→ evaporation

- **Plume expansion**
- **Laser absorption by plasma**

Nanosecond laser ablation

Features wish list

- seamless treatment of heating / melting / vaporization
- T-dependence: thermophysical properties, reflection & absorption coeffs
- vapor as non-ideal gas, need EoS up to critical T (8000 K)
- spatially non-uniform laser pulse
- thermionic emission, Coulomb explosion: coupling to electric field
- target – plume coupling via Knudsen Layer for condensation , subsonic, sonic vaporization
- determine T_{vap} , surface recession velocity
- plume expansion / recondensation, in vacuum or in background gas
- absorption of laser energy in plume / shielding of target
- plasma formation (ions and electrons)
- capture (very) strong shocks via high resolution schemes
- in 2D (axisymmetric) and 3D

Nanosecond laser ablation

Additional Challenges in modeling laser ablation

- Extreme space and time scales
- Extreme gradients: T may rise to thousands of degrees locally
- Extreme variation in thermo-physical properties:

  k varies by 12 orders of magnitude

- Coupling vaporization to gas flow via Knudsen Layer

- Need extensive thermo-physical data:
  $\rho(T,P)$, $c_p(T)$, $k(T)$, phase diagram for ***solid, liquid, vapor***
  over the range 300 K to critical T (~8000K)

Nanosecond laser ablation

Target heating / melting / vaporization

described by Heat Conduction equation (for enthalpy):

$$\frac{\partial H}{\partial t} = \nabla\bullet(k\nabla T) + S$$

$$S = I_o(t)(1-R)\alpha\exp(-\alpha z)\exp(-r^2/a^2)$$ *laser source*

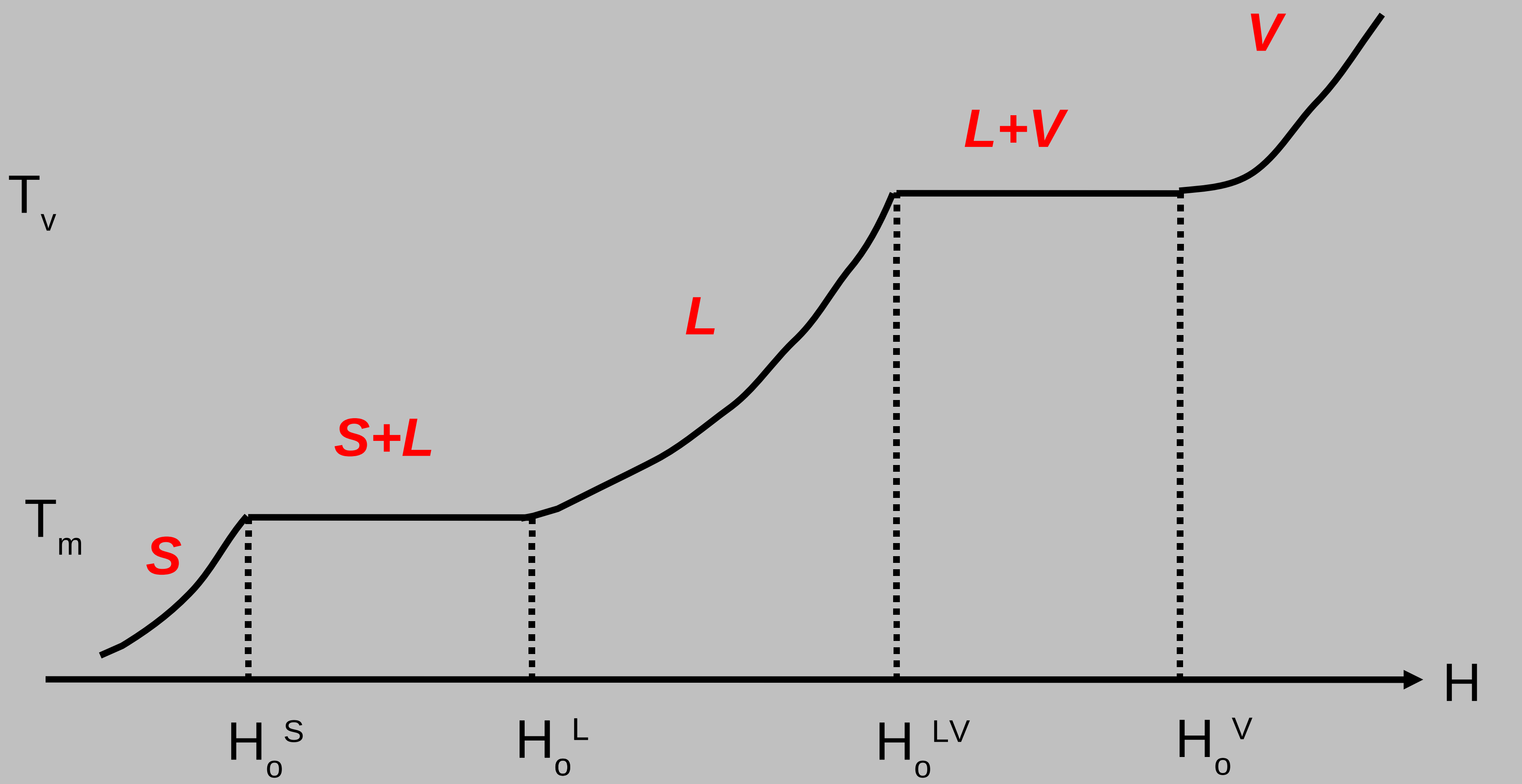
valid throughout the target, irrespectively of phase.

Phases are distinguished by value of enthalpy H via (thermodynamically consistent) *switch values*:

$H < H_o^S$: ***solid***, $H_o^S < H < H_o^L$: ***S+L***, $H_o^L < H < H_o^{LV}$: ***liquid***
$H_o^{LV} < H < H_o^V$: ***L+V***, $H_o^V < H$: ***vapor***

Thermal Equations of State

T and phase fractions from H:



$\Lambda^S=1$
$\Lambda^L=0$
$\Lambda^V=0$

$\Lambda^L=(H-H_o^L)/\Delta H^{fus}$
$\Lambda^S=1-\Lambda^L$
$\Lambda^V=0$

$\Lambda^L=1$
$\Lambda^S=0$
$\Lambda^V=0$

$\Lambda^V=(H-H_o^{LV})/\Delta H^{vap}$
$\Lambda^L=1-\Lambda^V$
$\Lambda^S=0$

$\Lambda^V=1$
$\Lambda^L=0$
$\Lambda^S=0$

Thermal EoS: *switch values*

Choose P_{ref}=1 atm, h_{ref} , set:

$$H_o^S := \rho^S h_o^S \;,\; h_o^S := h_{ref}$$

$$H_o^L := \rho^L h_o^L \;, \quad h_o^L := h_o^S + \Delta h^{fus}(T_m, P_{ref}) \;,$$

$$\Delta H^{fus}(T_m, P_{ref}) := H_o^L - H_o^S = (\rho^L - \rho^S) h_o^S + \rho^L \Delta h^{fus}$$

$$H_o^{LV}(T_v) := H_o^L + \int_{Tm}^{Tv} C_p^L(\tau) d\tau$$

$$H_o^V(T_v, P_{ref}) := \rho^V(T_v, P_{ref})\, h_o^V \;, \quad h_o^V(T_v, P_{ref}) := h_o^{LV}(T_v) + \Delta h^{vap}(T_v, P_{ref}) \;,$$

$$\Delta H^{vap}(T_v, P_{ref}) := H_o^V - H_o^{LV} = (\rho^V - \rho^L) h_o^{LV} + \rho^V \Delta h^{vap}(T_v, P_{ref})$$

$H < H_o^S$: ***solid***, $H_o^S < H < H_o^L$: ***S+L***, $H_o^L < H < H_o^{LV}$: ***liquid***

$H_o^{LV} < H < H_o^V$: ***L+V***, $H_o^V < H$: ***vapor***

Algorithm in target

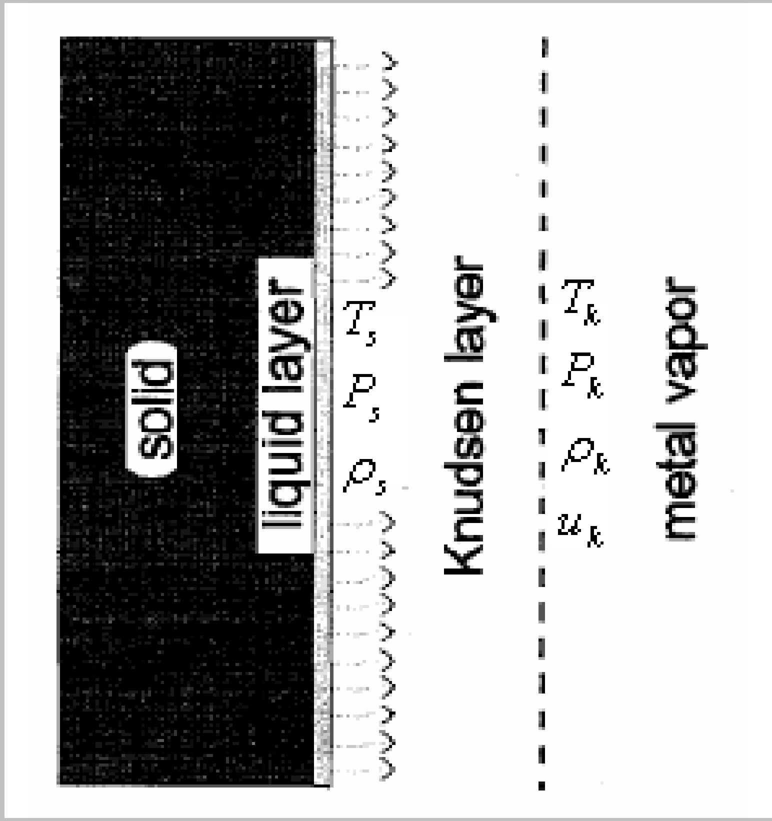
Time-explicit Finite Volume discretization

Update H from conservation law

Determine phase fractions and T from EoS

If $\Lambda^V > 0$, determine T_{vap} , $P_{sat}(T_{vap})$ at surface,

and surface recession velocity,
via energy and mass conservation
and jump conditions across the
Knudsen Layer



which couple target to plume and yield incoming boundary conditions for fluid dynamics in plume.

Knudsen Layer

Thin transition layer (a few mean free paths) between liquid and vapor where molecules are not in translational equilibrium.

T, P, ρ undergo jumps governed by local Mach number

We use [Gusarov-Smurov, 2005] treatment (messy...)

From energy and mass conservation and the KL jump conditions, we determine the following at the “outside” of the KL:

* ***Mach number*** ($\Longrightarrow$ condensation or subsonic or sonic vap)
* ***recession velocity of melt surface***
* ***vaporization T***
* ***saturation Pressure***
* ***vapor density***
* ***vapor velocity*** ($\longrightarrow$ inflow BC for CFD in plume)

Discretization: *Finite Volume, explicit in time*

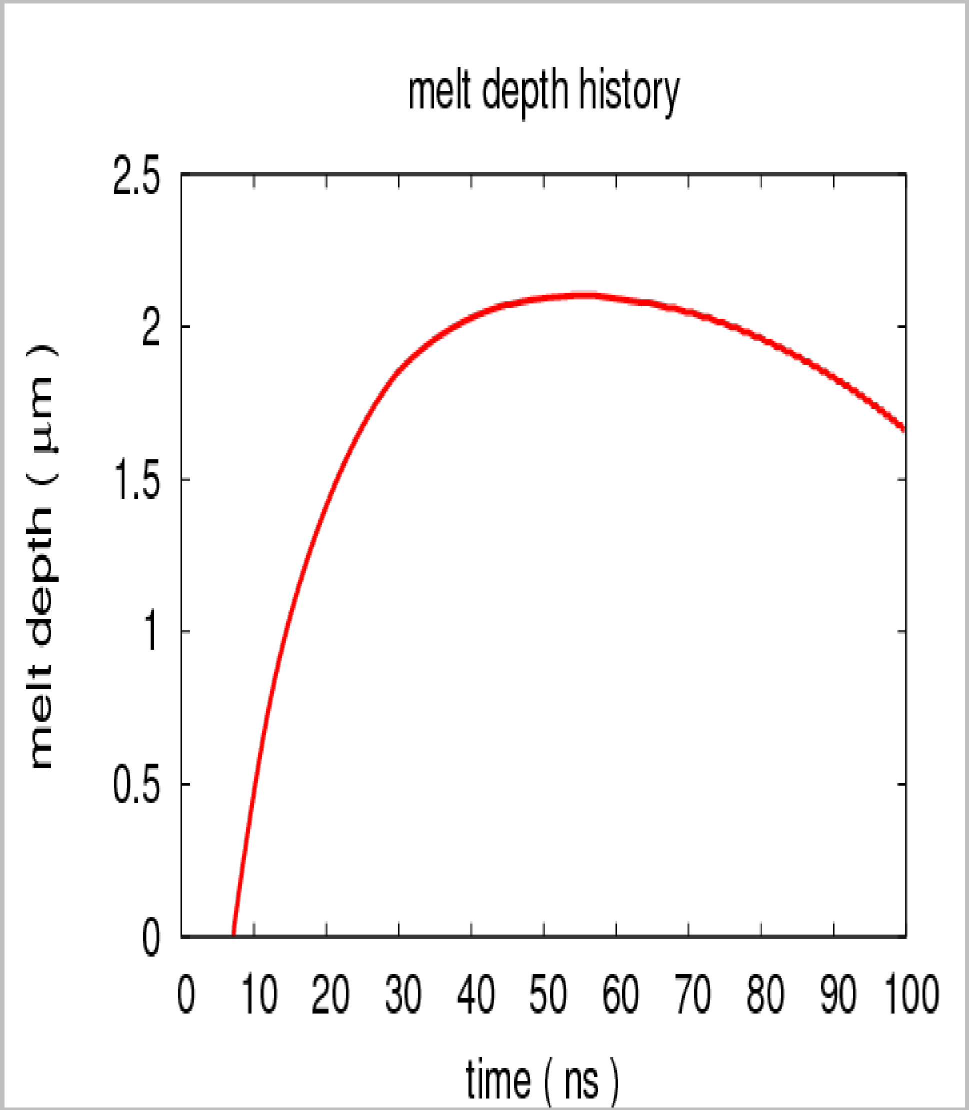
Mesh for 1-D simulation: *$\Delta x = 200$ nm, $\Delta t = 10^{-5}$ ns*

(expanding mesh)

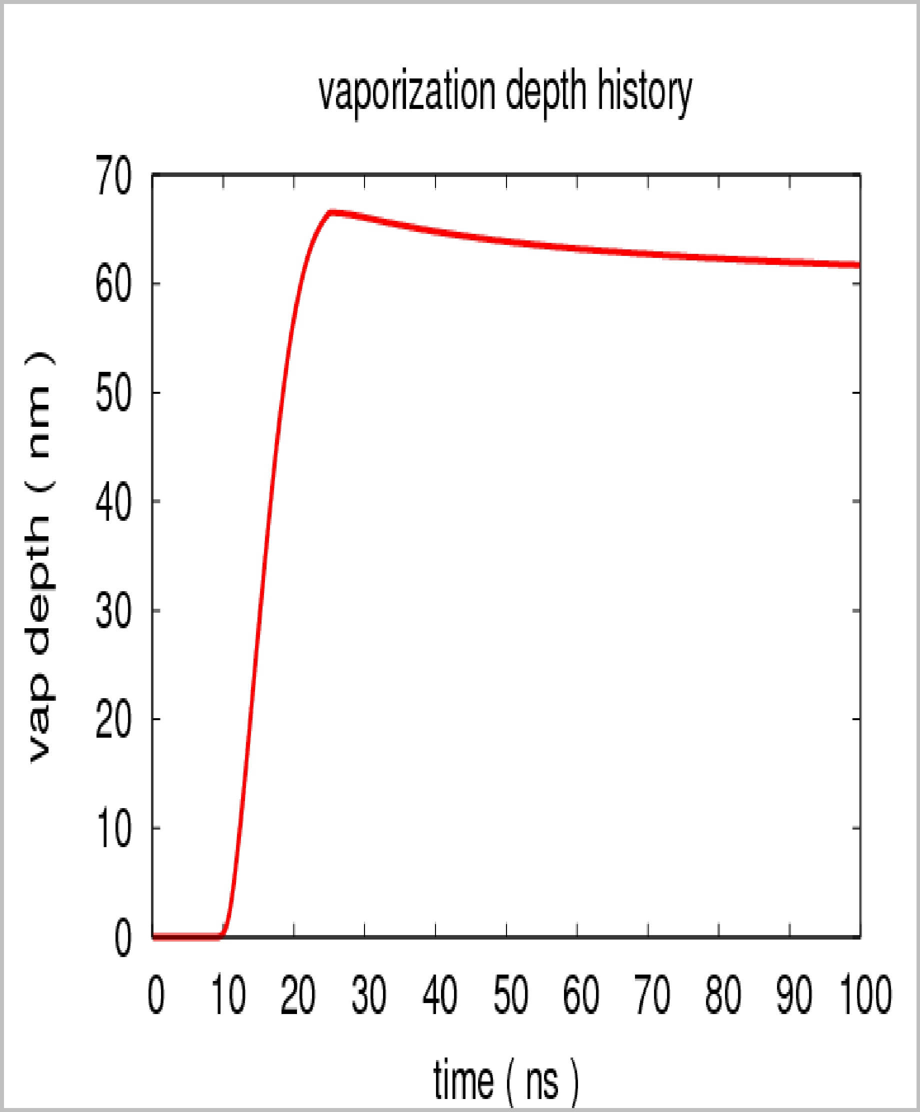
Target melting - vaporization

1-D simulation

Melt depth $\approx 2\ \mu m$



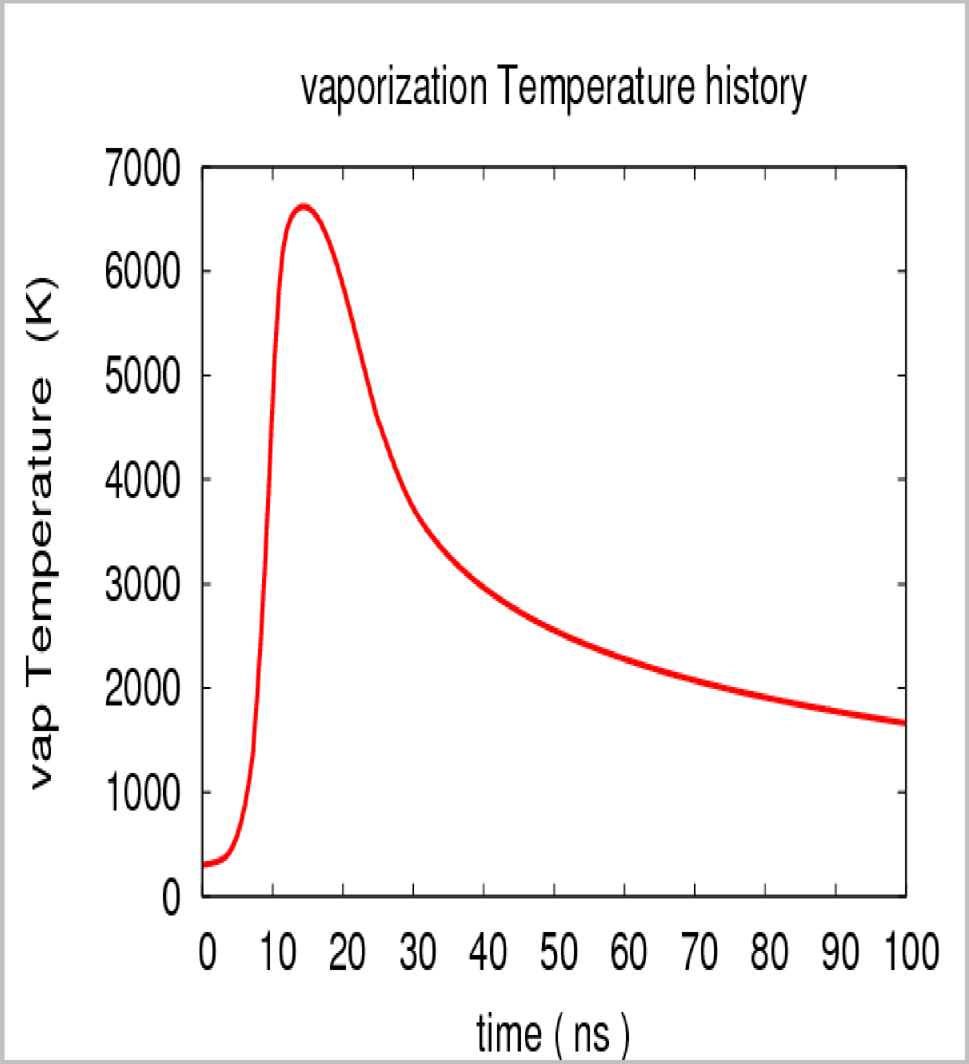
Vaporization depth $\approx 70\ nm$

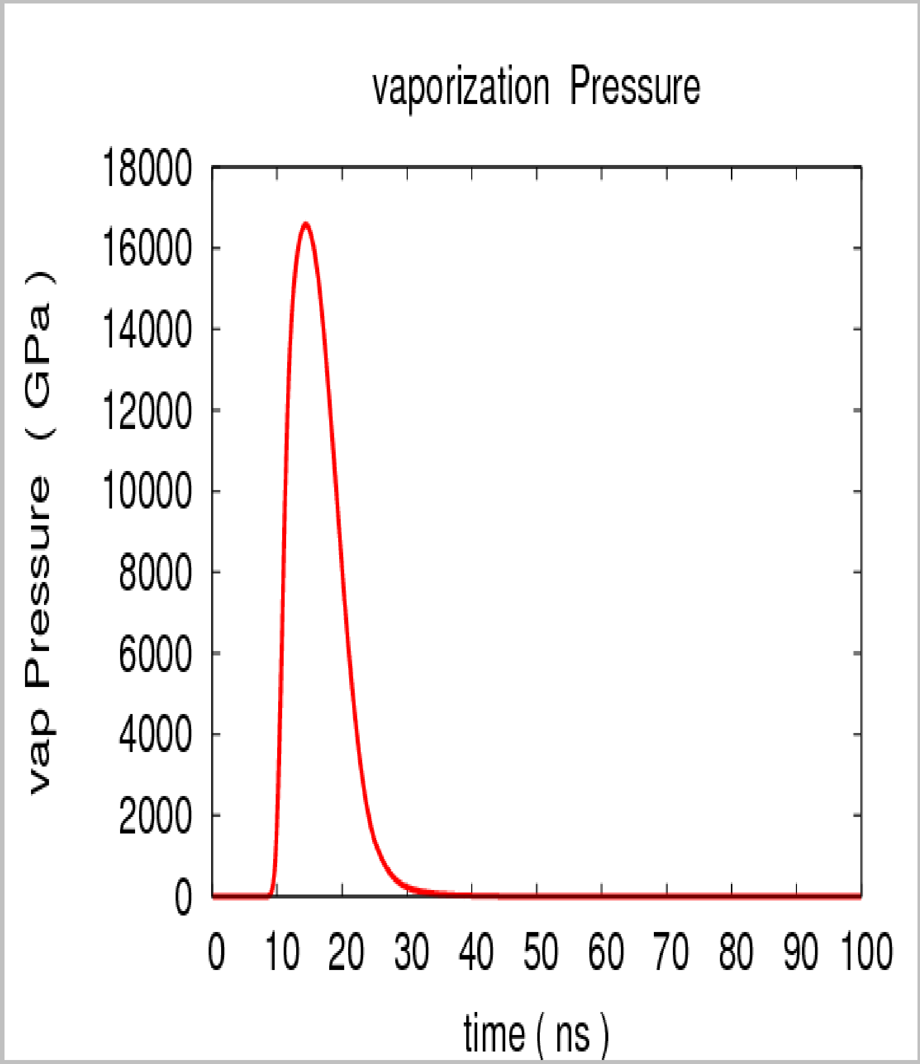


Target vaporization

1-D simulation

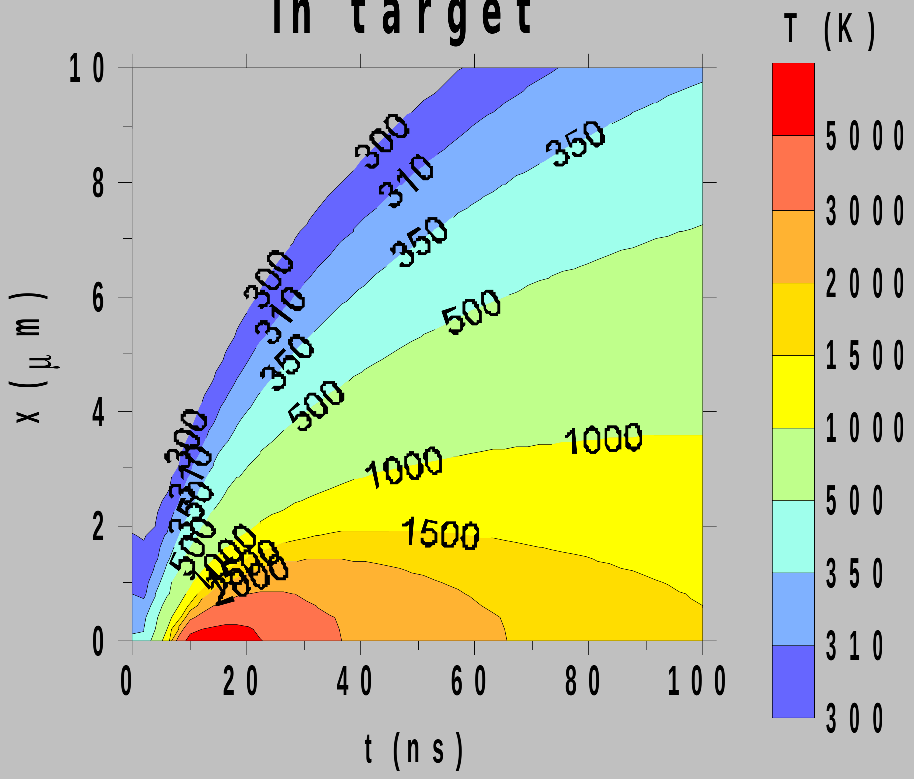
peak T_{vap} ~ 6600 K at t~15 ns peak P_{vap} ~ 160×10^6 atm



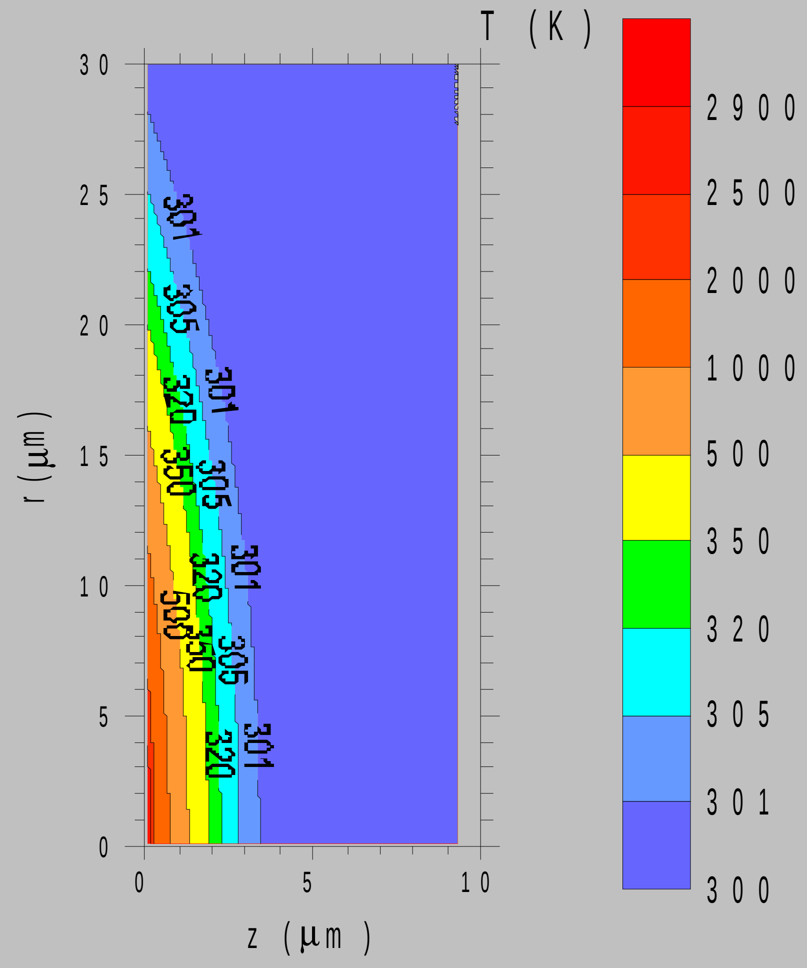


Target heating

Temperature distribution in target



1D



2D, at 10 ns

Expansion of vapor plume in background gas

Conservation laws:

Uknowns: $\rho_i, \vec{v}, T, P$

- Mass (species: Cu , He)
- Momentum
- Energy
- Equation of State

$$\frac{\partial U}{\partial t} + \nabla \cdot \vec{F} = S$$

$$U = \begin{bmatrix} \rho \\ \rho \vec{v} \\ \rho \epsilon \end{bmatrix} \quad \vec{F} = \begin{bmatrix} \rho \vec{v} \\ \rho \vec{v} \vec{v} + P 1 \\ (\rho \epsilon + P) \vec{v} + \vec{q} \end{bmatrix} \quad S = \begin{bmatrix} 0 \\ 0 \\ \Lambda \end{bmatrix}$$

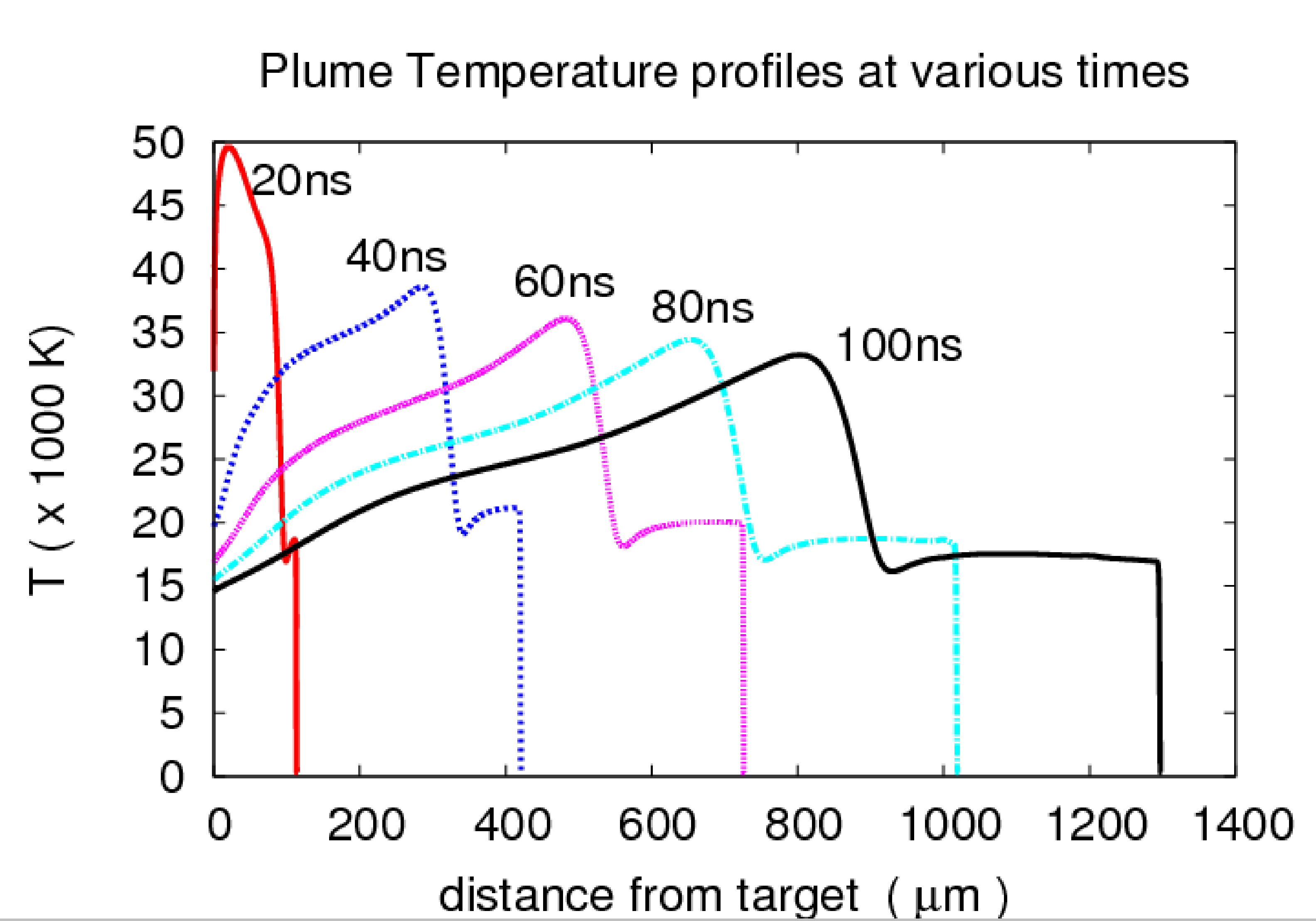
Discretization: *Finite Volume, explicit in time*
Advection scheme: *2nd order central (Tadmor) or AUSM+*

Mesh for 1-D simulation: *Δx = 200 nm, $\Delta t = 10^{-5}$ ns*
(expanding mesh: starts with 50 nodes, grows to 2500)

Expansion of vapor plume in He gas

1-D simulation

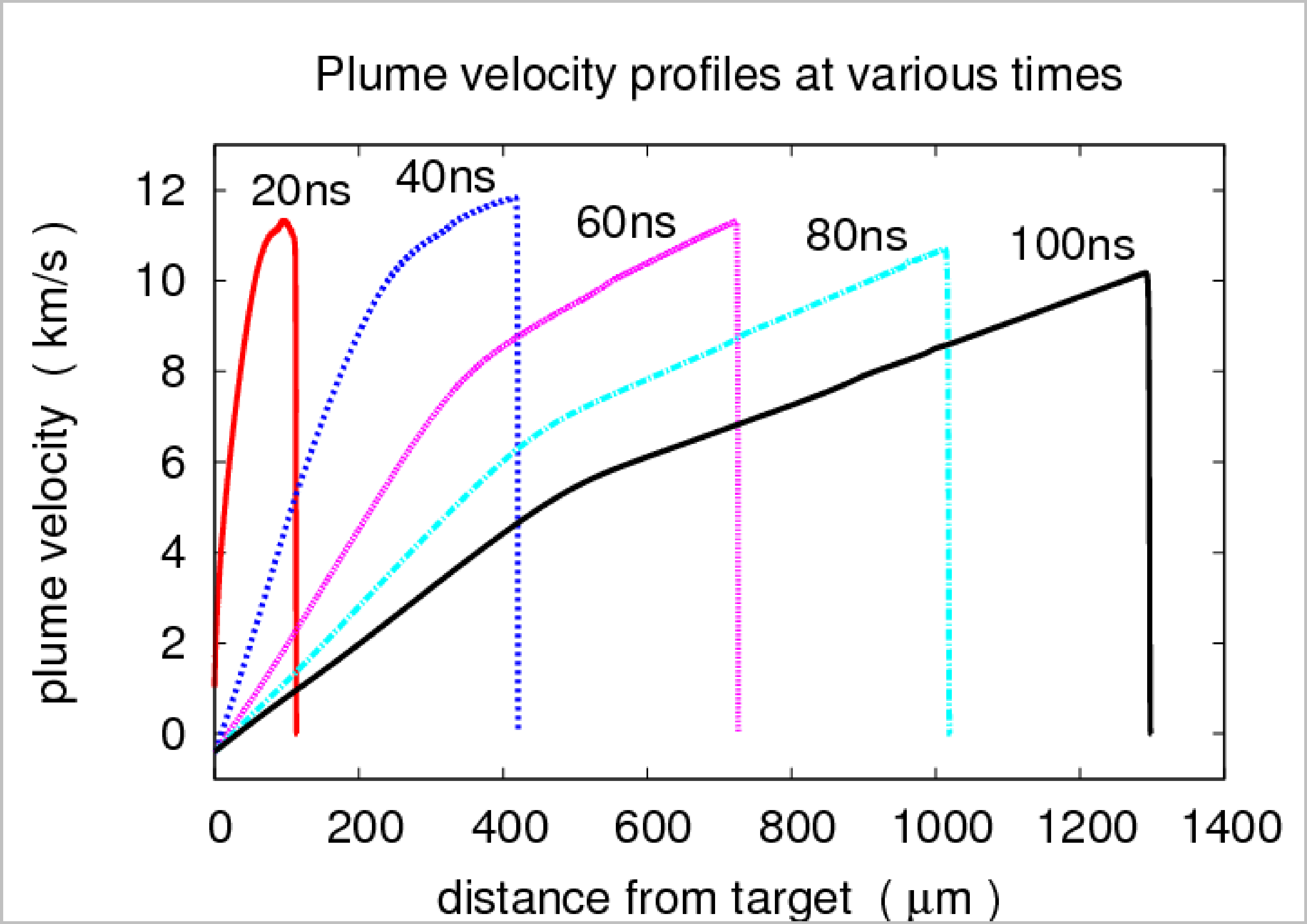
Peak plume **Temperature** ~ 50,000 K !!!



Expansion of vapor plume in He gas

1-D simulation

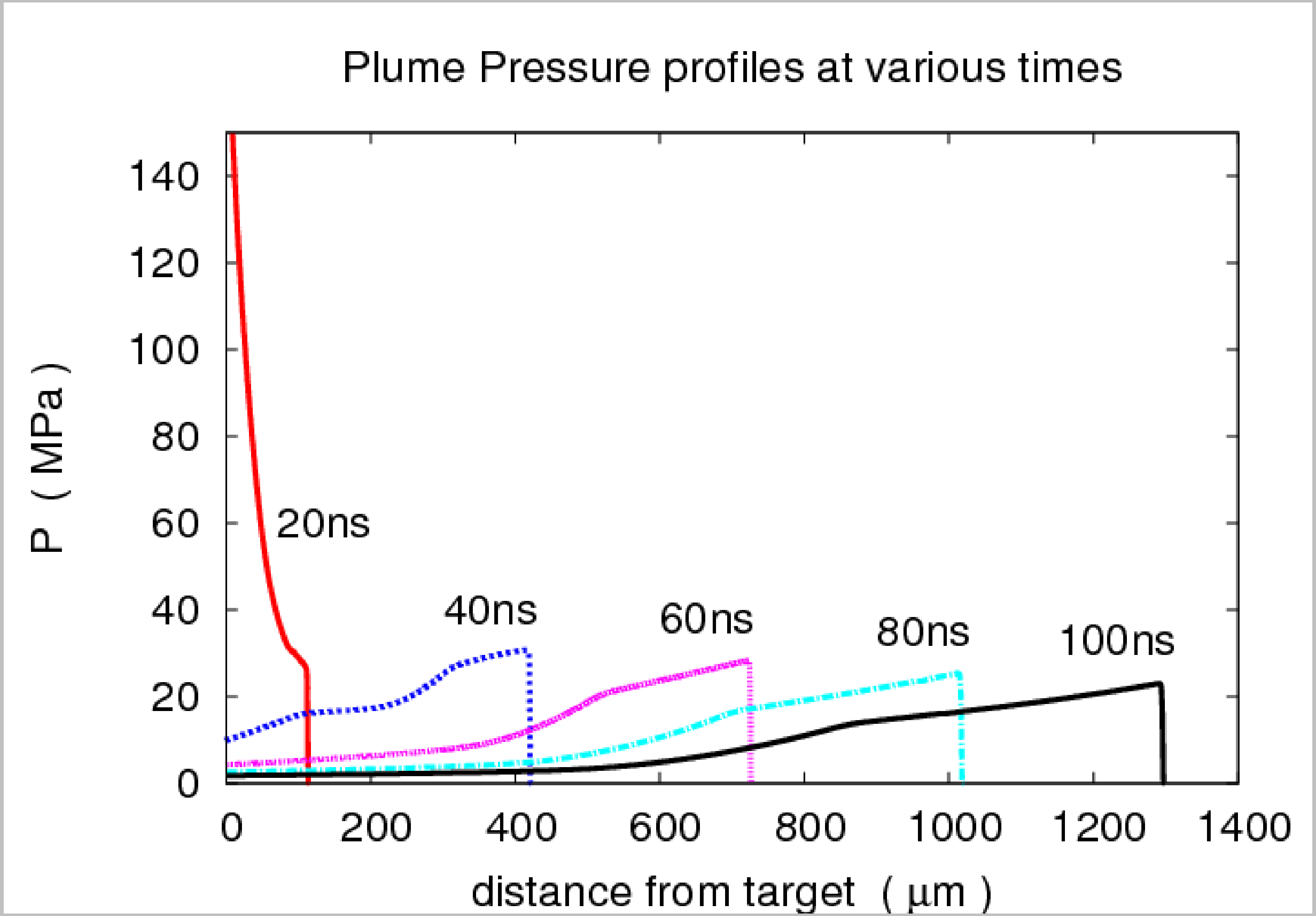
Peak plume **velocity** ~ 12 km/s !!!



Expansion of vapor plume in He gas

1-D simulation

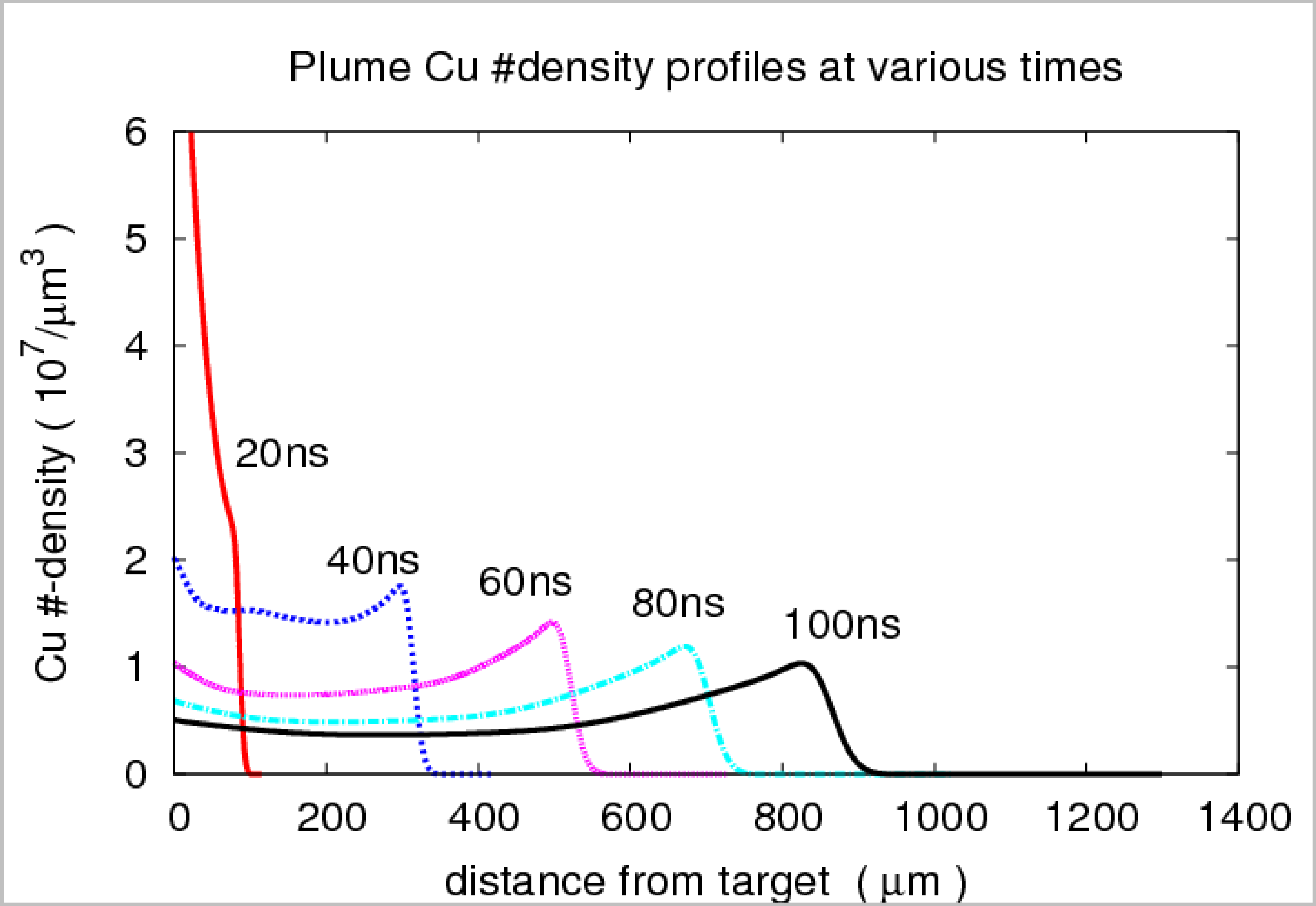
max plume **Pressure** ~ 220 MPa (~ 2200 atm)



Expansion of vapor plume in He gas

1-D simulation

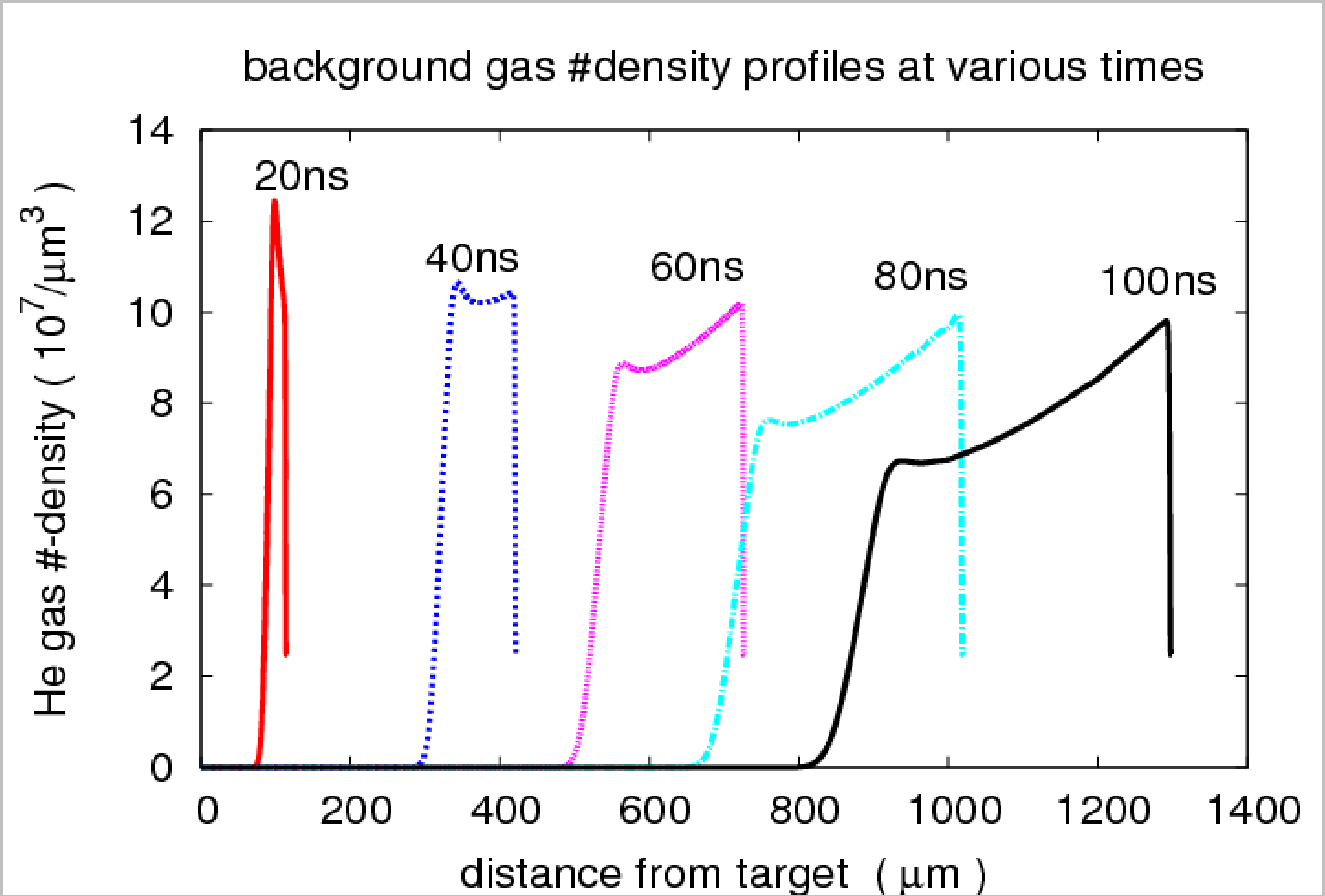
Peak **Copper vapor** Number-density ~ $27 \cdot 10^{25}/m^3$



Expansion of vapor plume in He gas

1-D simulation

Peak **He gas** number-density ~ $12 \cdot 10^{25}/m^3$



Current and future developments

- Bubble formation (Volmer-Döring theory)

- explosive boiling near critical T (Eötvös rule)
  (has major effect on melt depth due to shielding by plasma)

- Two Temperatures in plasma, for ions and electrons

- Inverse Bremsstrahlung and multi-photon ionization

Future

- fs (femptosecond) laser: all the above features and more

- extend to 2D (axisymmetric), and later to 3D

- coupling to electric field in target to model
  thermionic emission and Coulomb explosion

- Particle formation by recondensation and transport in 2D, 3D

Particle formation by condensation