Modeling of Laser Induced Breakdown in Gases

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Outline

Motivation and Goal

Physico-chemical Modeling

Numerical Method and CFD tool

Applications

Conclusions

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Why studying laser-plasma interactions (LPI)?

Scientific motivation (very interesting problem)

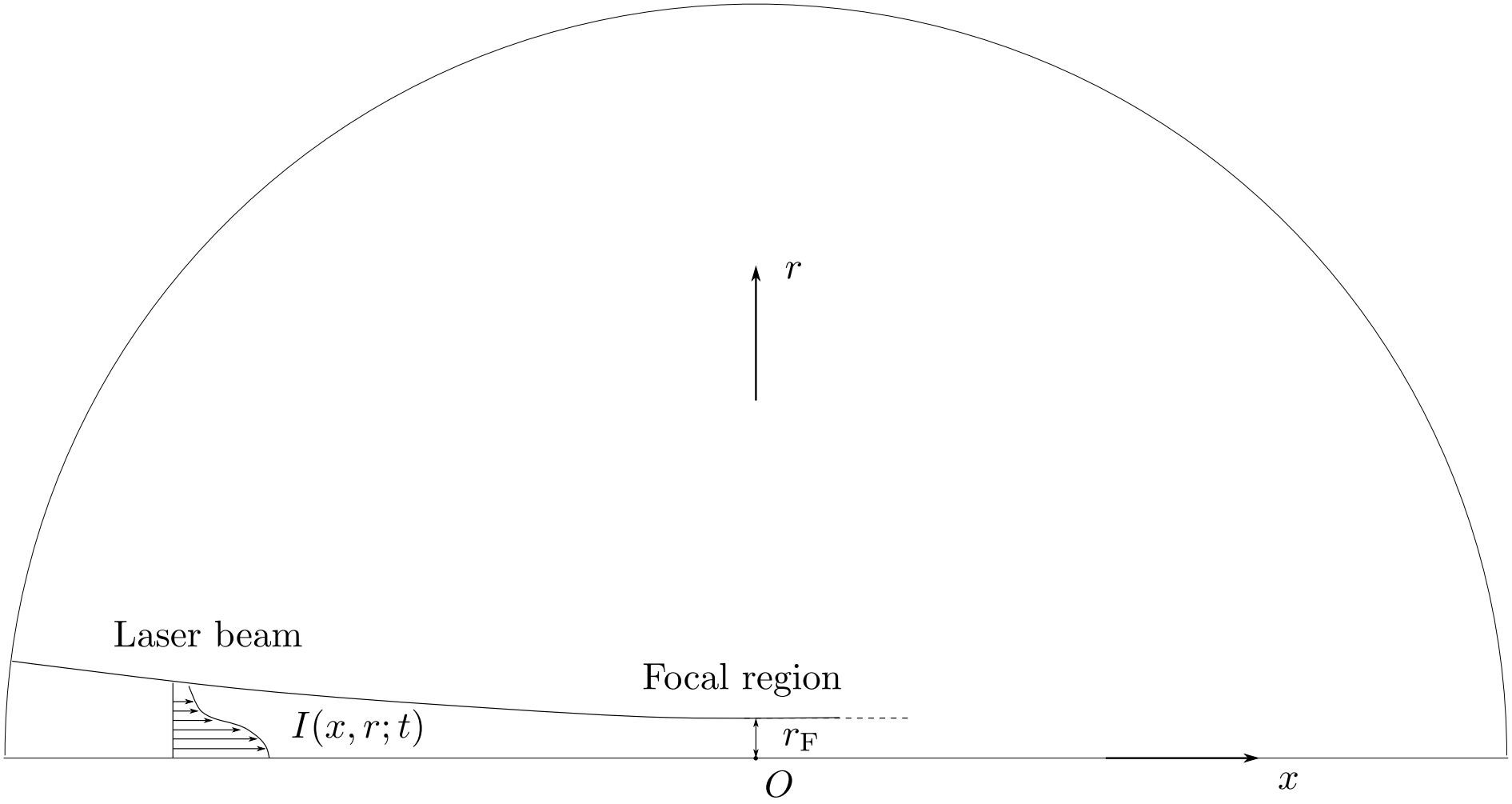
- Understand the inter-play among diverse physical phenomena (*e.g.*, ionization, self-focusing, refraction, ablation) occurring when a plasma is formed by focusing a laser beam onto a target material (*e.g.*, gas, solid)

Lasers have many applications in engineering

- Plasma-coupled combustion
- Laser-induced breakdown spectroscopy (LIBS)
- Laser welding
- ...

⇒ *last but not least, modeling of LPI poses challenges which are also faced in re-entry aerothermodynamics (possible mutual benefits)*

Problem definition: optical breakdown of gases



Large spectrum of time-scales makes modeling of LPI challenging!

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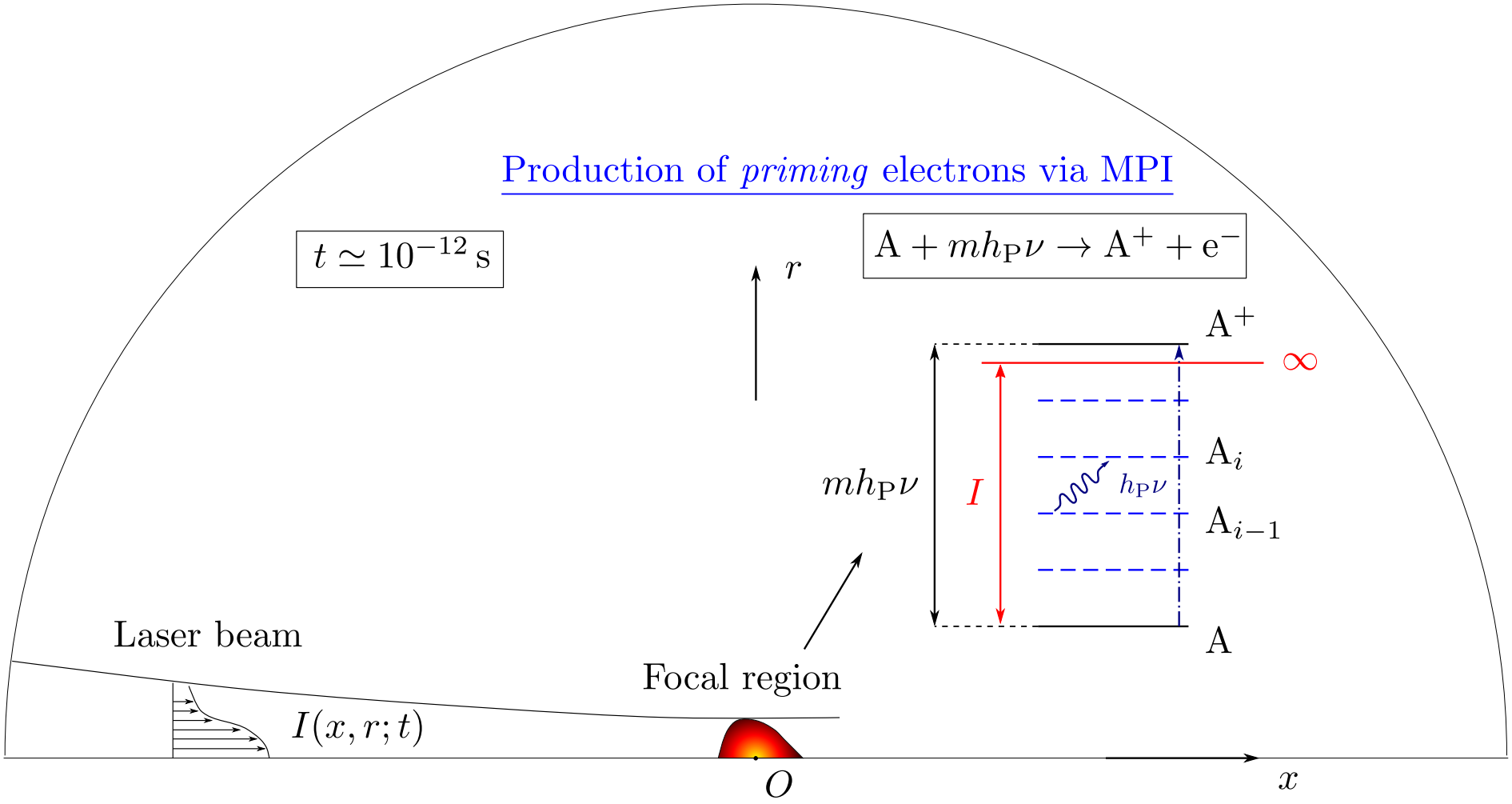

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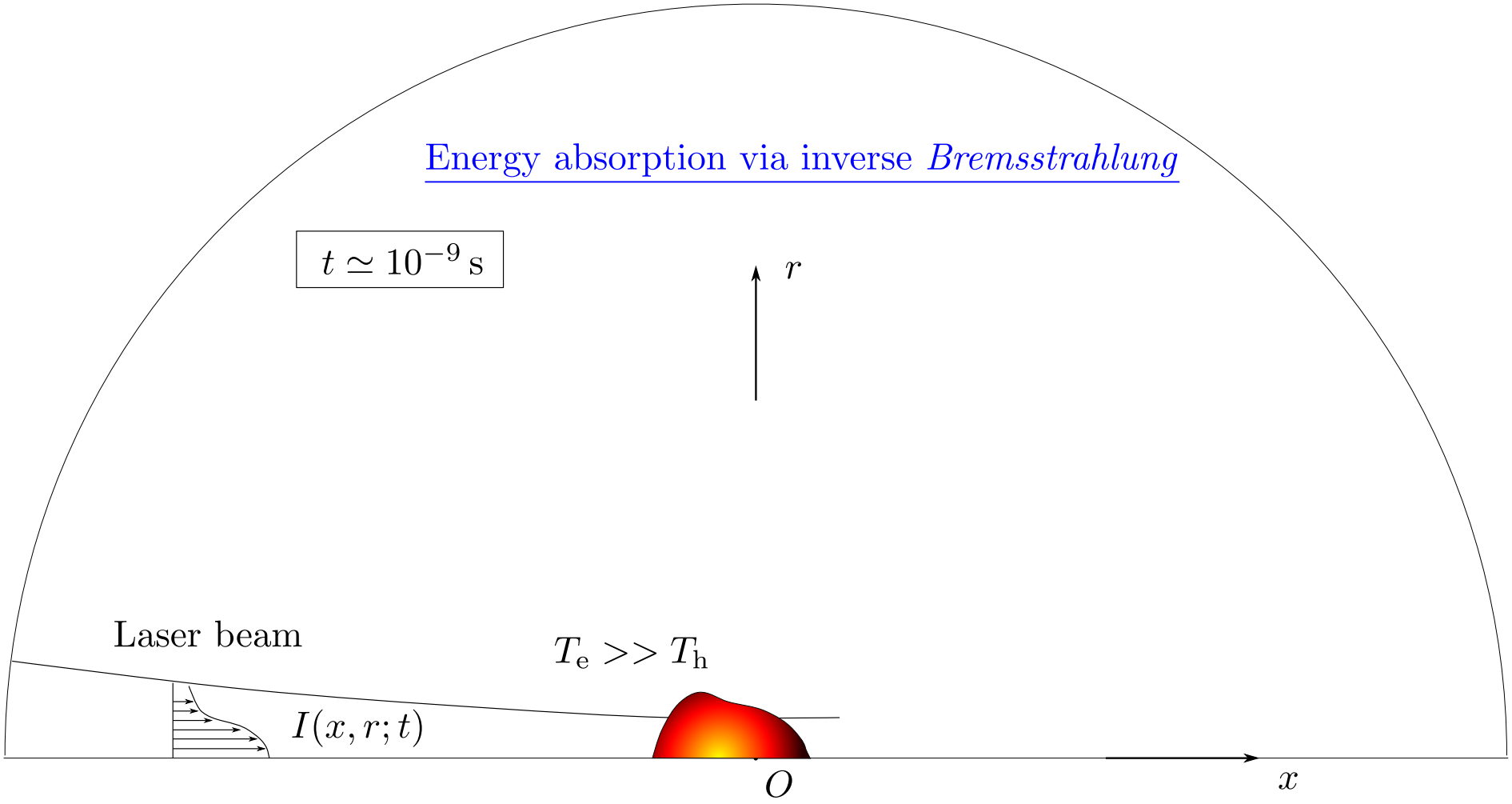

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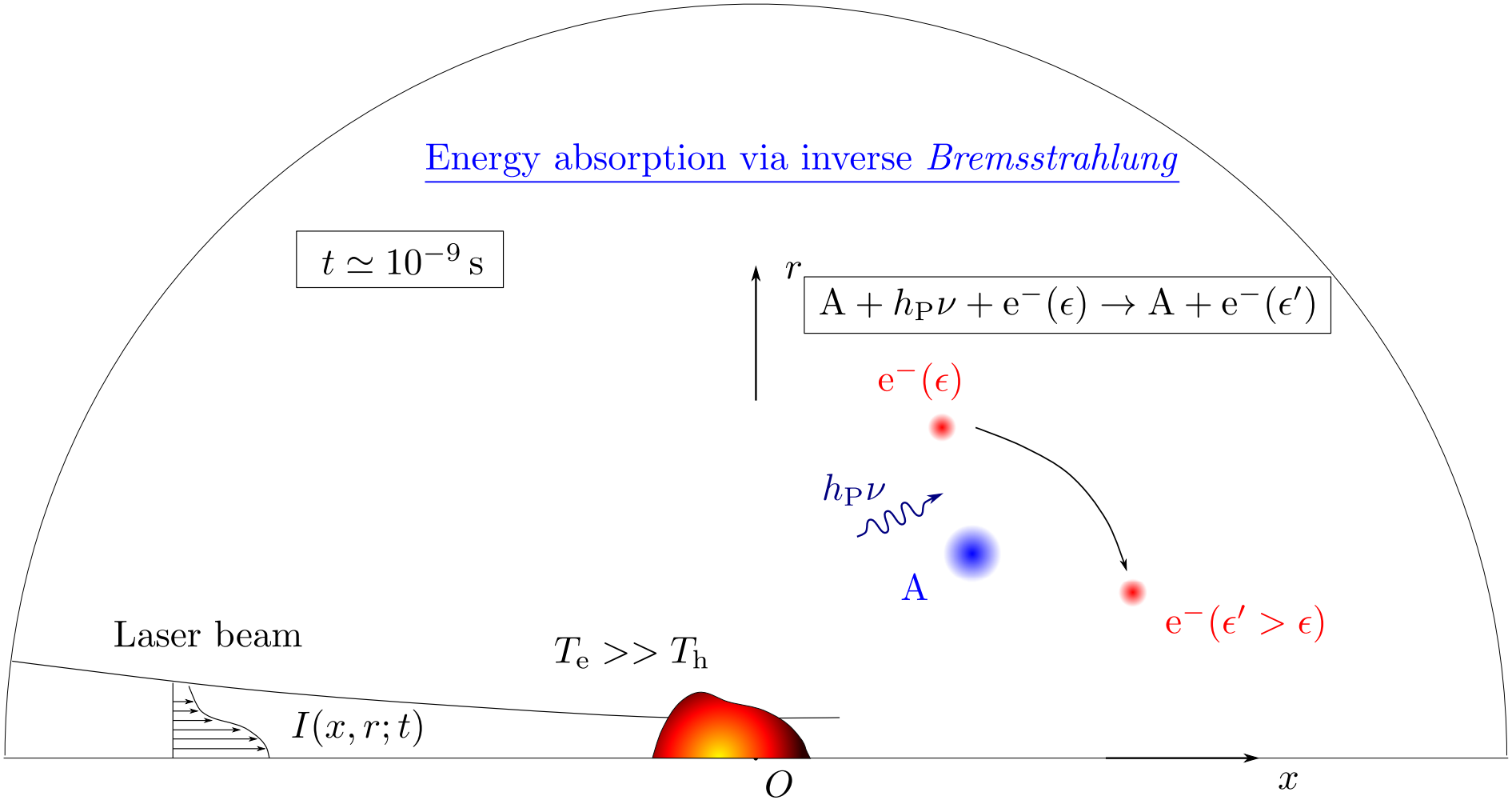

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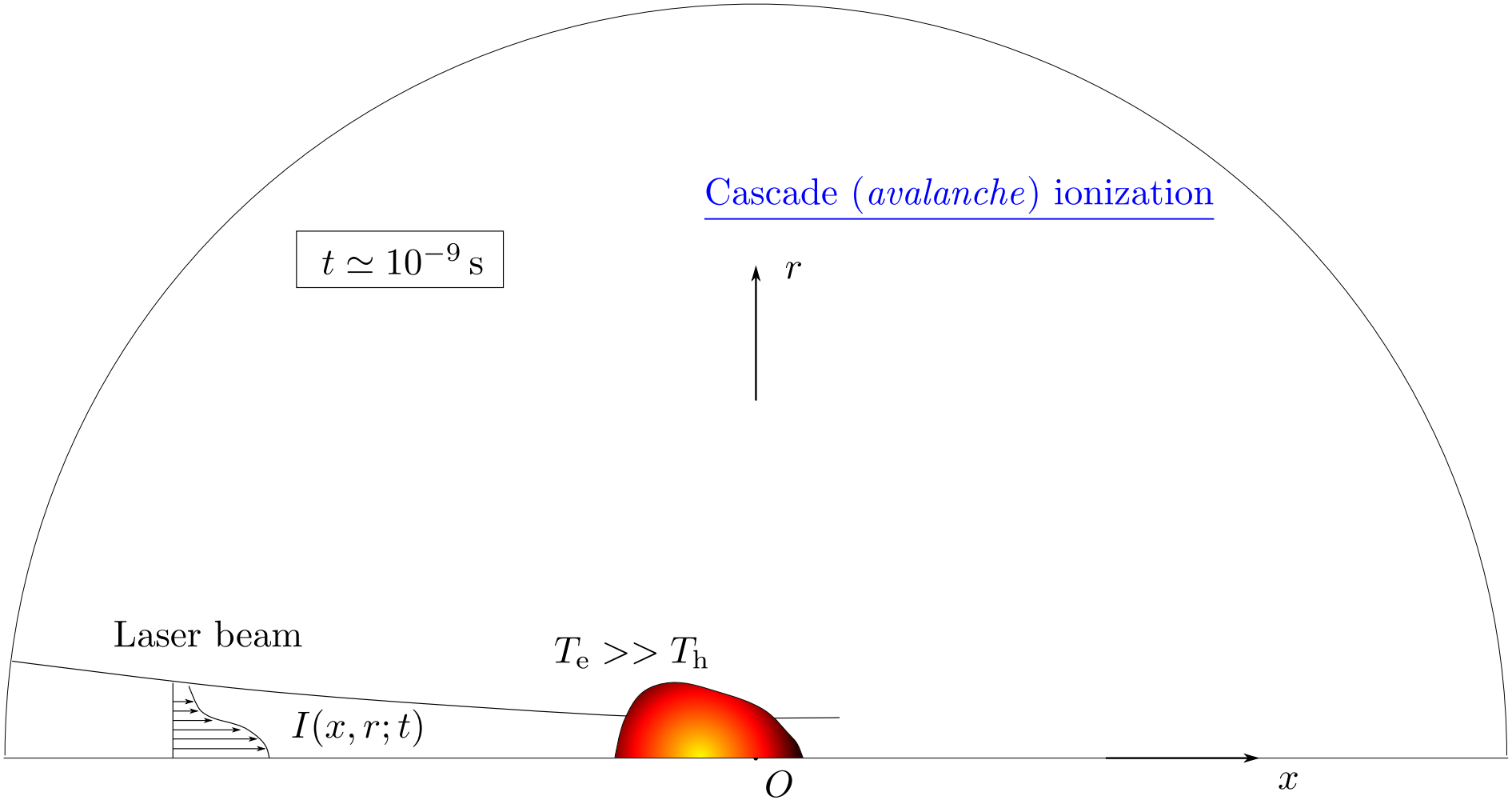

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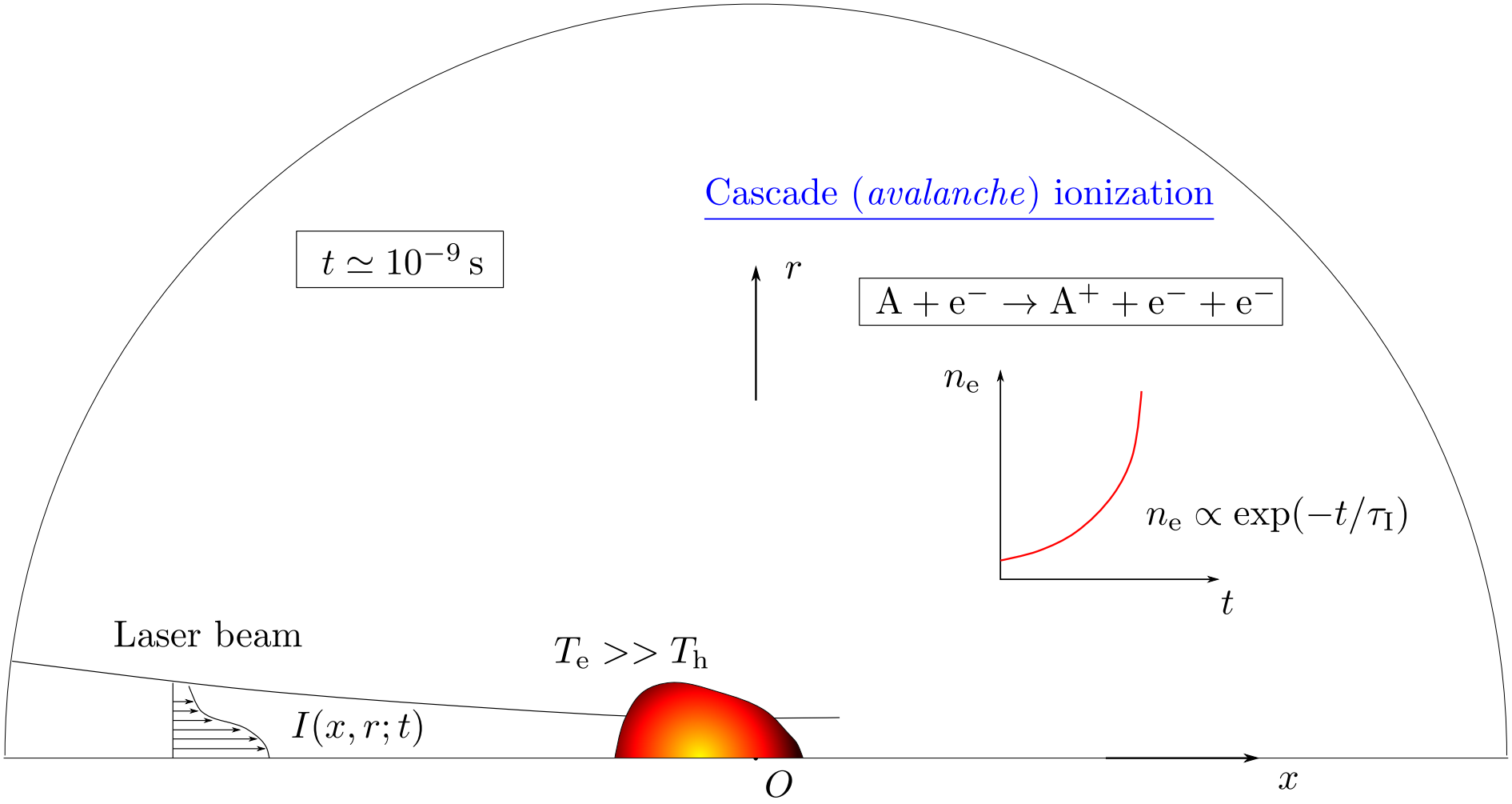

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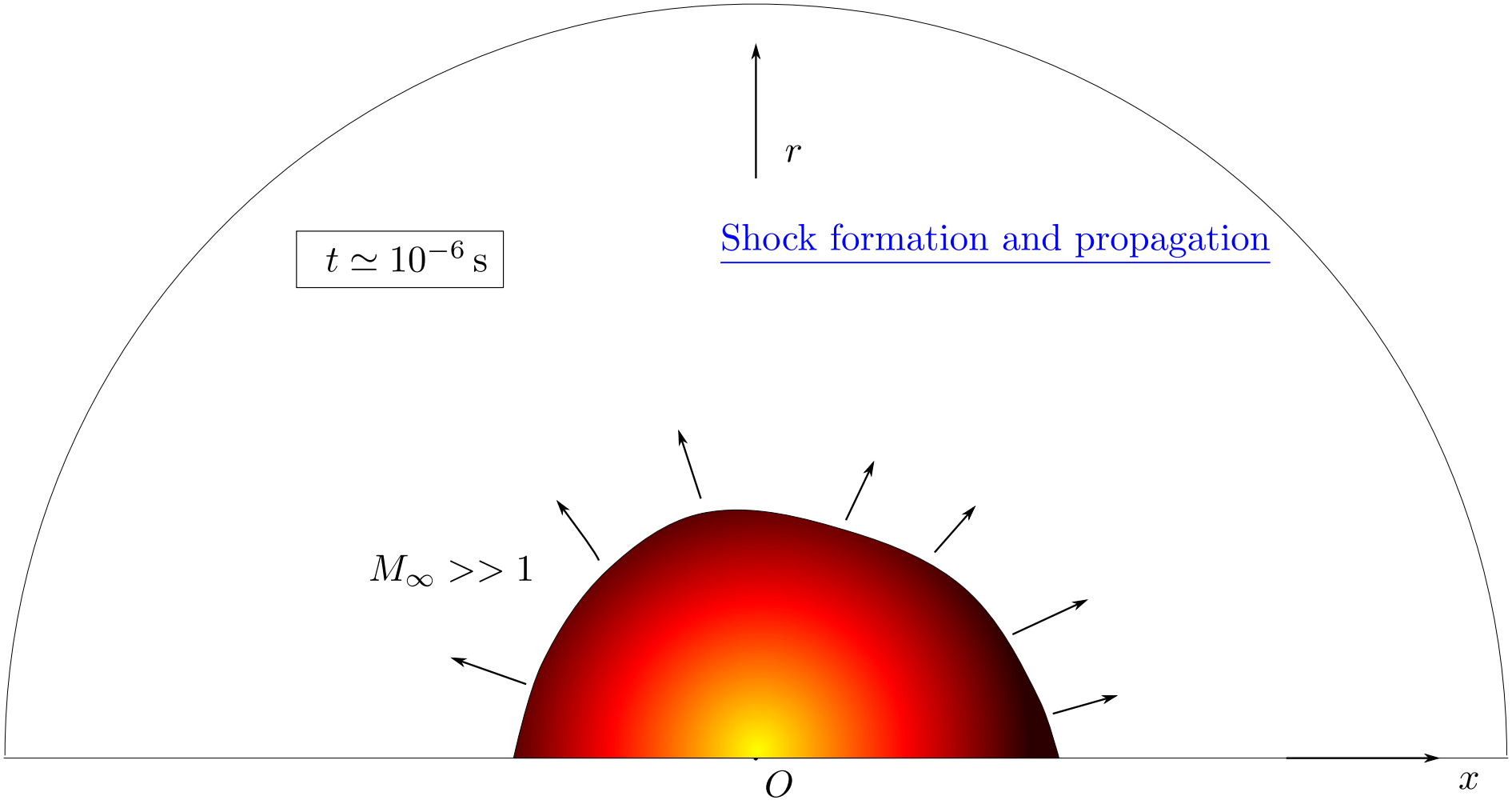

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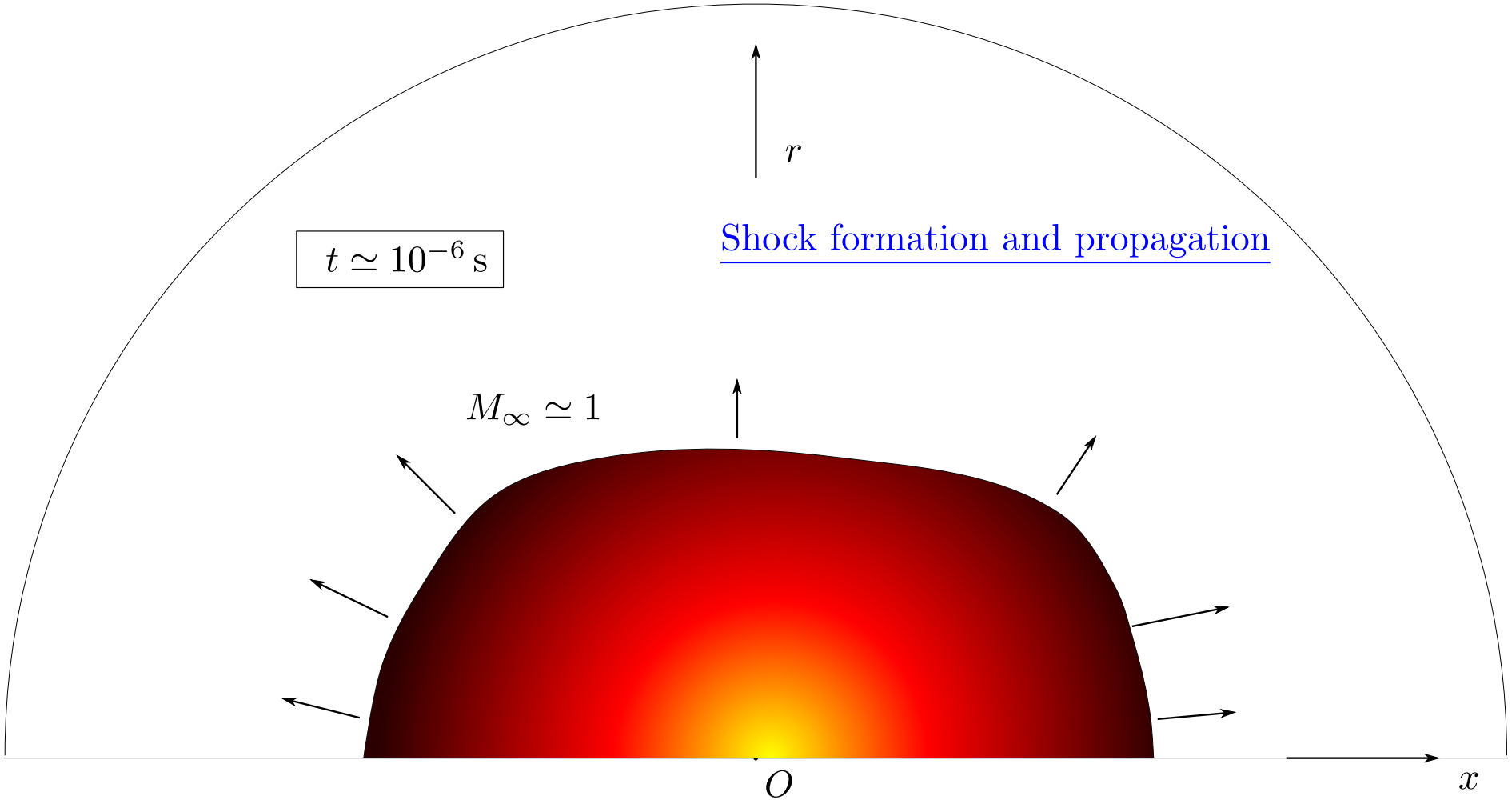

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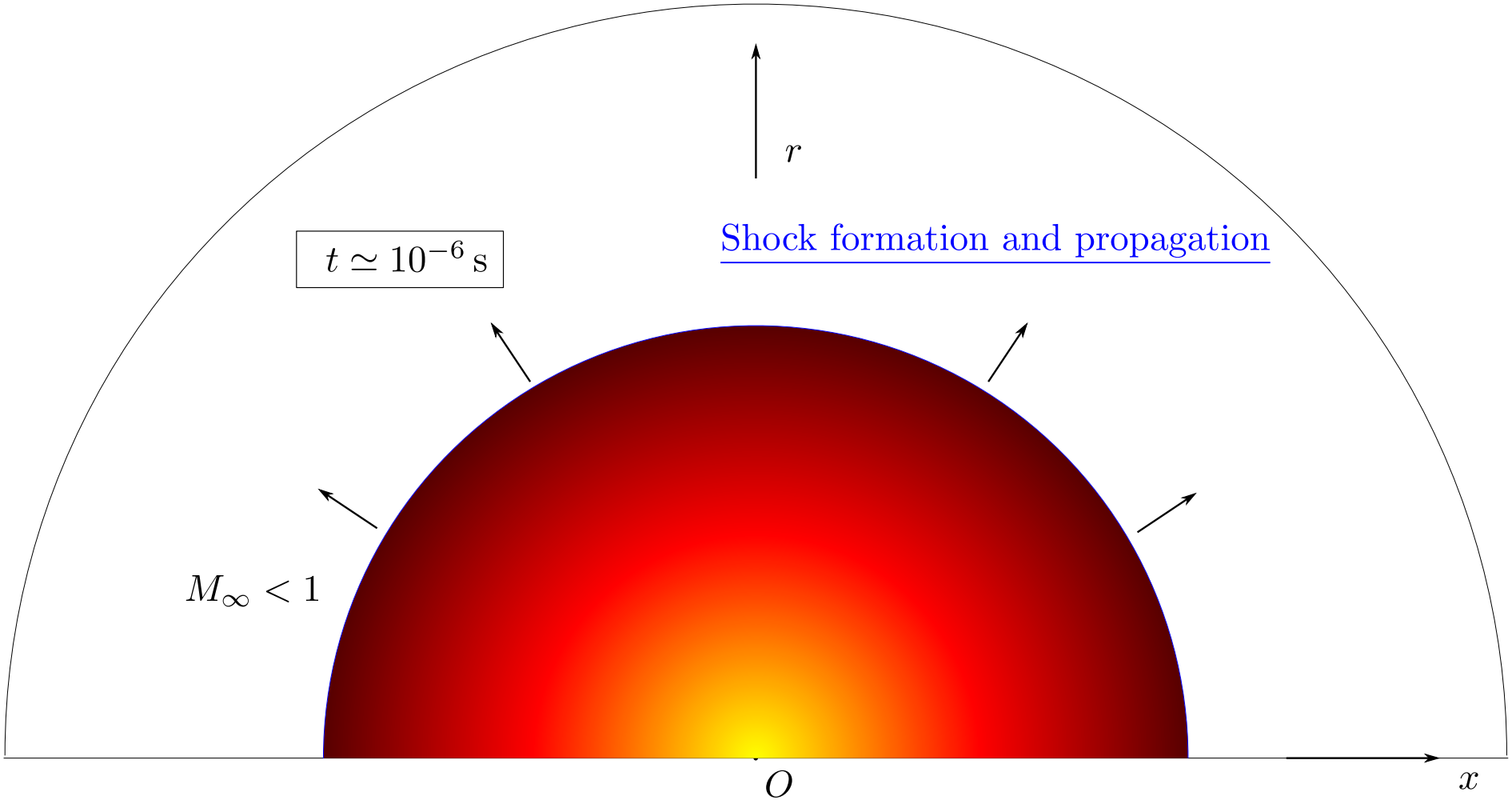

Large spectrum of time-scales makes modeling of LPI challenging!

Literature review

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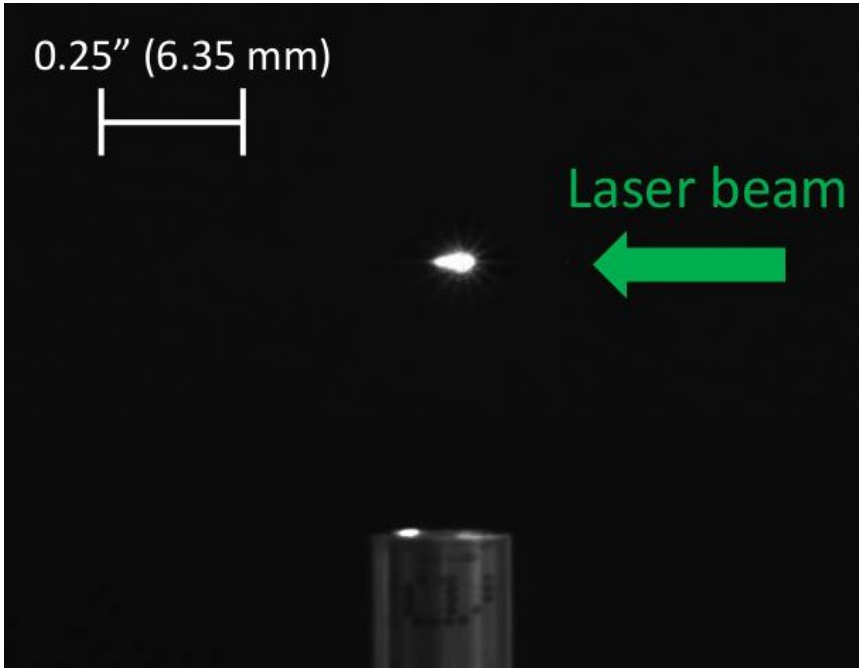
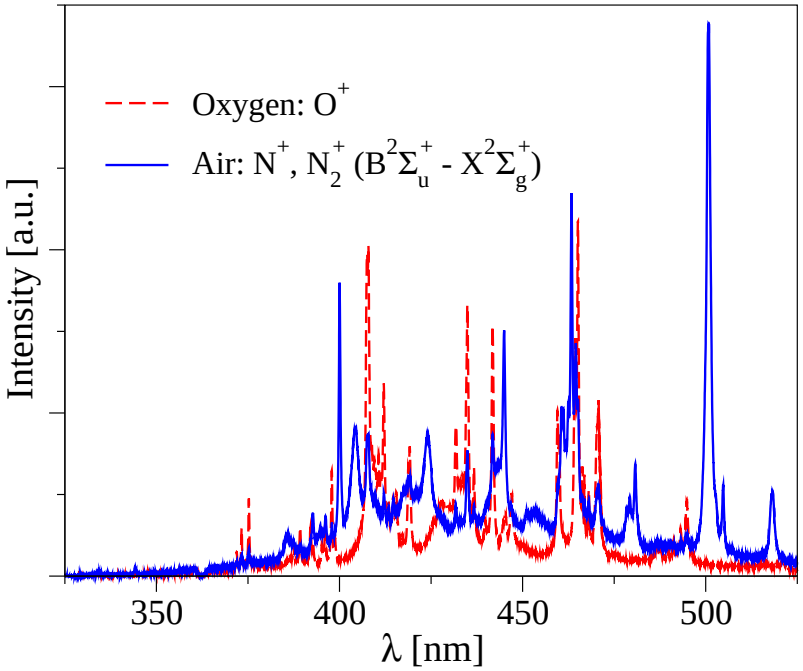
Objective

Goal

- construct and validate a predictive *physics-based* model for optical breakdown of gases



Laser discharge (M. Nishihara, UIUC)



Experimental spectra (M. Nishihara, UIUC)

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Physical model (i): material gas

Non-Local Thermodynamic Equilibrium (NLTE) fluid model

Two-temperature model: T_{h} for heavy-particles, T_{e} for free-electrons

Governing equations: NLTE Navier-Stokes

- *Mass continuity (for each species)*

$$\frac{\partial \rho_s}{\partial t} + \nabla \cdot (\rho_s \mathbf{v} + \mathbf{J}_s) = \omega_s^{\mathrm{COL}} + \omega_s^{\mathrm{RAD}}$$

- *Global momentum*

$$\frac{\partial \rho \mathbf{v}}{\partial t} + \nabla(\rho \mathbf{v} \otimes \mathbf{v} + p\mathbf{I}) = \nabla \boldsymbol{\tau}$$

- *Global energy*

$$\frac{\partial \rho E}{\partial t} + \nabla \cdot (\rho \mathbf{v} H) = \nabla{:}(\boldsymbol{\tau}\mathbf{v}) - \nabla \cdot \mathbf{q} + \Omega^{\mathrm{RAD}}$$

- *Free-electron + vibrational + electronic excitation energy*

$$\frac{\partial \rho e_{\mathsf{ve}}}{\partial t} + \nabla \cdot (\rho \mathbf{v} e_{\mathsf{ve}}) = -p_{\mathsf{e}} \nabla \cdot \mathbf{v} - \nabla \cdot \mathbf{q}_{\mathsf{ve}} + \Omega_{\mathsf{ve}}^{\mathrm{COL}} + \Omega_{\mathsf{ve}}^{\mathrm{RAD}}$$

Physical model (ii): radiation field

Possible approaches (particle and wave; classical electrodynamics)

Radiative Transfer Equation (RTE; Kinetic Theory of Photons)

$$\boldsymbol{\Omega} \cdot \nabla I_\lambda = \varepsilon_\lambda - \kappa_\lambda I_\lambda$$

+ Straightforward to include radiative processes (*e.g.*, line emission)
x Formulation becomes much more complex for refractive and dispersive media, and when accounting for scattering and polarization
x No coherence and no wave phenomena (*e.g.*, diffraction, interference)

Maxwell's equations

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t}$$

+ Wave *behavior* of light taken into account
x More difficult to include effects radiative processes
x Very fine grids required for optical region of the EM spectrum

RTE formulation

Splitting of intensity

$$I_\lambda = I_\lambda^{\mathrm{c}} + I_\lambda^{\mathrm{d}}$$

- I_λ^{c}: collimated part (*e.g.*, laser)
- I_λ^{d}: diffuse/remnant part (*i.e.*, plasma response)

Radiation transport problem is now split in two uncoupled RTEs

- Collimated (only for laser wavelength $\lambda = \lambda_{\mathrm{l}}$) *[this work]*

$$\boldsymbol{\Omega} \cdot \nabla I_\lambda^{\mathrm{c}} = -\kappa_\lambda \, I_\lambda^{\mathrm{c}}$$

- Diffuse (whole spectrum) *[on-going]*

$$\boldsymbol{\Omega} \cdot \nabla I_\lambda^{\mathrm{d}} = \varepsilon_\lambda - \kappa_\lambda \, I_\lambda^{\mathrm{d}}$$

$\Rightarrow$ *the two RTEs may be solved with different numerical methods (e.g., ray-tracing for collimated and Finite Volume for diffuse)*

⇒ *RTE for collimated beam*

$$\boldsymbol{\Omega}\cdot\nabla I_\lambda^{\mathrm{c}} = -\kappa_\lambda^{\mathrm{FF}} I_\lambda^{\mathrm{c}}, \qquad \kappa_\lambda^{\mathrm{FF}} = \sum_{s\in\mathcal{H}} \mathcal{Q}_{\mathrm{e}s}^{\mathrm{FF}}(\lambda, T_{\mathrm{e}})\, n_{\mathrm{e}} n_s \left[1 - \exp\left(\frac{h_{\mathrm{P}} c}{k_{\mathrm{B}} T_{\mathrm{e}}}\right)\right]$$

with $\mathcal{Q}_{\mathrm{e}s}^{\mathrm{FF}}(\lambda, T_{\mathrm{e}})$ being the *free-free* absorption cross-section*

Electron-ion (Coulomb potential)

$$\mathcal{Q}_{\mathrm{e}s}^{\mathrm{FF}}(\lambda, T_{\mathrm{e}}) = \frac{4}{3}\sqrt{\frac{2\pi}{3 m_{\mathrm{e}} k_{\mathrm{B}} T_{\mathrm{e}}}}\, \frac{\lambda^3}{(4\pi\epsilon_0)^3}\, \frac{Z_s^2 q_{\mathrm{e}}^6}{h_{\mathrm{P}} c^4 m_{\mathrm{e}}}$$

Electron-neutral (soft photon limit)

$$\mathcal{Q}_{\mathrm{e}s}^{\mathrm{FF}}(\lambda, T_{\mathrm{e}}) = \int_0^{+\infty} \sigma_{\mathrm{e}s}^{\mathrm{FF}}(\lambda, \varepsilon)\, f^{\mathrm{M}}(\varepsilon, T_{\mathrm{e}})\, d\varepsilon$$

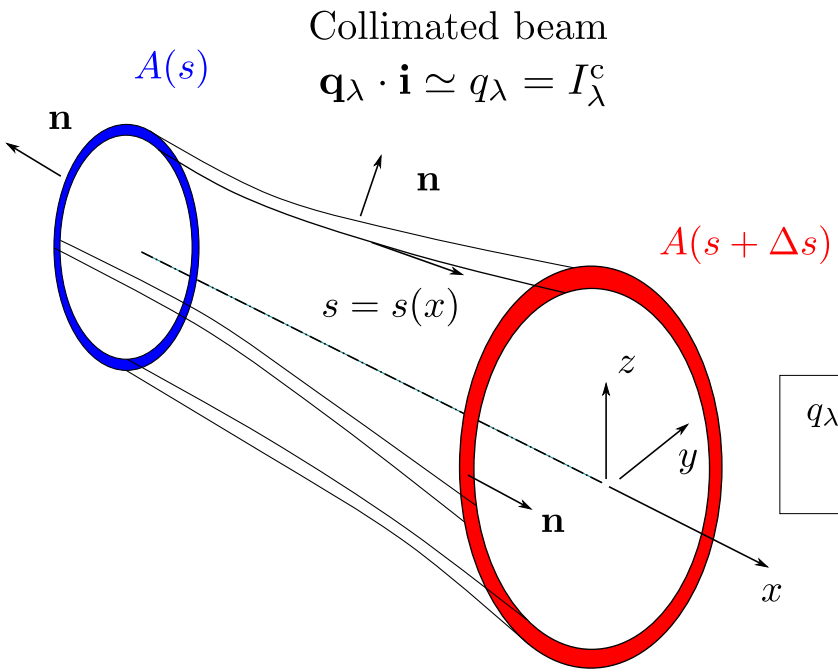
$$\sigma_{\mathrm{e}s}^{\mathrm{FF}}(\lambda, \epsilon) \simeq \frac{q_{\mathrm{e}}^2 \lambda^3}{6\pi^2 \epsilon_0\, h_{\mathrm{P}} c^4 m_{\mathrm{e}}^{3/2}} \left(\varepsilon + \frac{1}{2}\frac{h_{\mathrm{P}} c}{\lambda}\right) \sqrt{2\left(\varepsilon + \frac{h_{\mathrm{P}} c}{\lambda}\right)}\, Q_{\mathrm{e}s}^1\left(\varepsilon + \frac{1}{2}\frac{h_{\mathrm{P}} c}{\lambda}\right)$$

* Johnston, R. R., *J. Quant. Spectrosc. Radiat. Transf.* **7**, 815, 1967

RTE for laser beam: flux tube formulation

- This model reproduces the increase of the intensity/heat-flux towards the focal plane

  $\Rightarrow$ *alternative would be to solve Maxwell's equations* [on-going]



Energy conservation

$$\oint_{\Sigma_V} \mathbf{q}_\lambda \cdot \mathbf{n}\, d\Sigma = -\int_V \kappa_\lambda\, q_\lambda\, dV$$

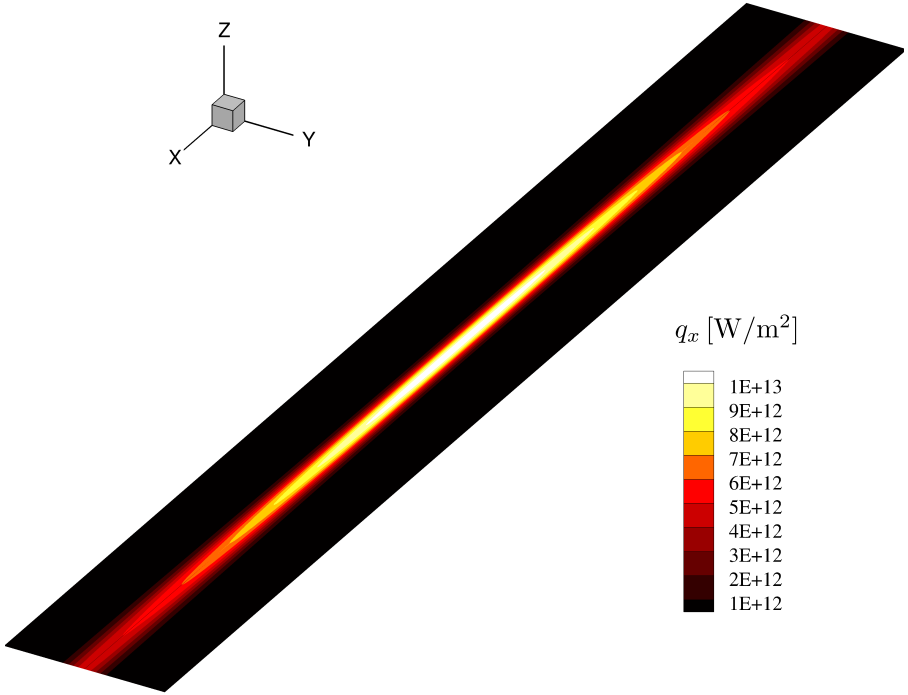
$$q_\lambda(s+\Delta s)\, A(s+\Delta s) - q_\lambda(s)\, A(s) = -\kappa_\lambda(s')\, q_\lambda(s')\, A(s')\, \Delta s$$
$$s \le s' \le\ s + \Delta s$$

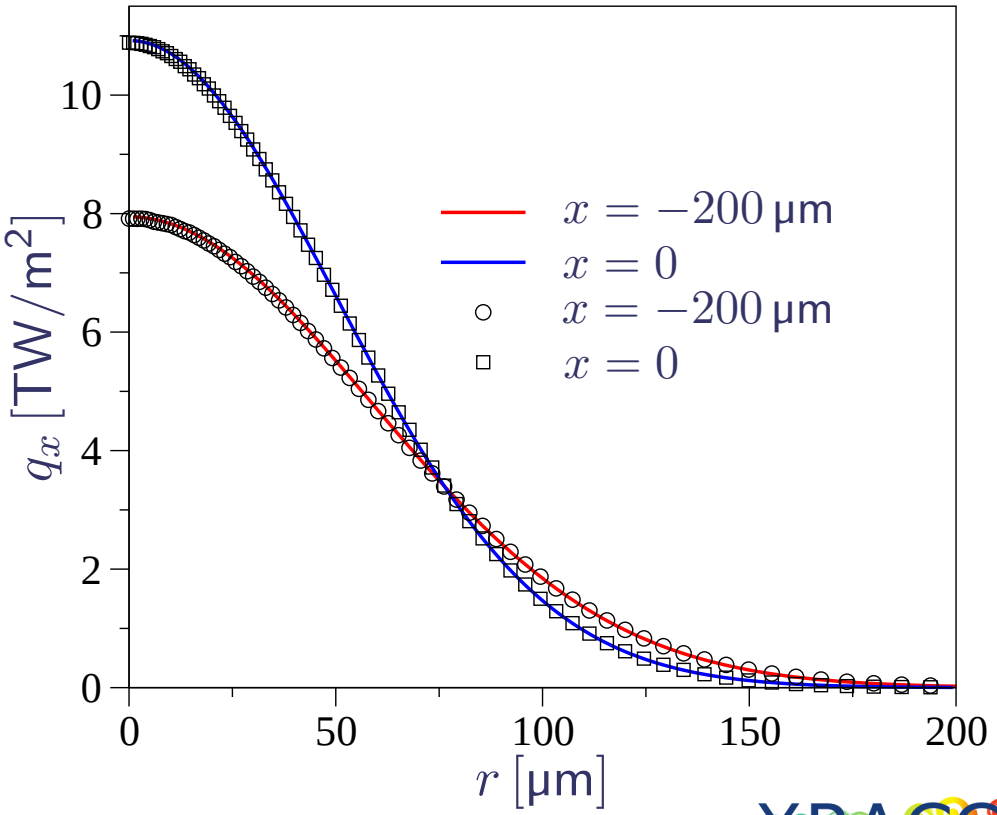
$$\frac{\partial I^{\mathrm{c}}_\lambda A}{\partial x} = -\kappa_\lambda\, I^{\mathrm{c}}_\lambda\, A, \quad ds \simeq dx \text{ (par-axial approximation}^*\text{)}$$

* Marcuse, D., *Light Transmission Optics*, van Nostrand Reinhold Company, 1982

⇒ *verification/application for a Gaussian beam (focal plane at $r = 0$)*

- Beam *discretized* using a finite number of rays/bundles
- Area distribution, $A(x)$, based on specified focal radius, focal length and beam diameter
- Left: heat-flux/intensity map
- Right: one-dimensional slices (symbols denote numerical solution)





Radiative source terms

Once the intensity known one may compute radiative mass and energy source terms (these provide coupling between gas and radiation)

- Multi-photon-ionization (MPI) $[s + k_s h_\mathrm{P} \nu_\mathrm{l} = s^+ + \mathrm{e}^-]$

$$\omega_s^{\mathrm{MPI}} = -m_s \dot{n}_s, \quad \dot{n}_s = n_s \underbrace{\sigma_s^{\mathrm{MPI}}(\nu_\mathrm{l}) I_{\lambda_\mathrm{l}}^{k_s}}_{\text{mpi rate coeff.}}$$

$$\Omega^{\mathrm{MPI}} = \sum_{s \in \mathcal{H}} \dot{n}_s \, k_s \, h_\mathrm{P} \nu_\mathrm{l}, \quad \Omega_{\mathrm{ve}}^{\mathrm{MPI}} = \sum_{s \in \mathcal{H}} \dot{n}_s (I_s - k_s h_\mathrm{P} \nu_\mathrm{l})$$

- Inverse *Bremsstrahlung* (FF)

$$\Omega^{\mathrm{FF}} = \Omega_{\mathrm{ve}}^{\mathrm{FF}} = \kappa_{\lambda_\mathrm{l}}^{\mathrm{FF}} I_{\lambda_\mathrm{l}}$$

$\Rightarrow$ *in the literature MPI is often thought to be important only for formation of priming electrons (this is not the case as simulations show)*

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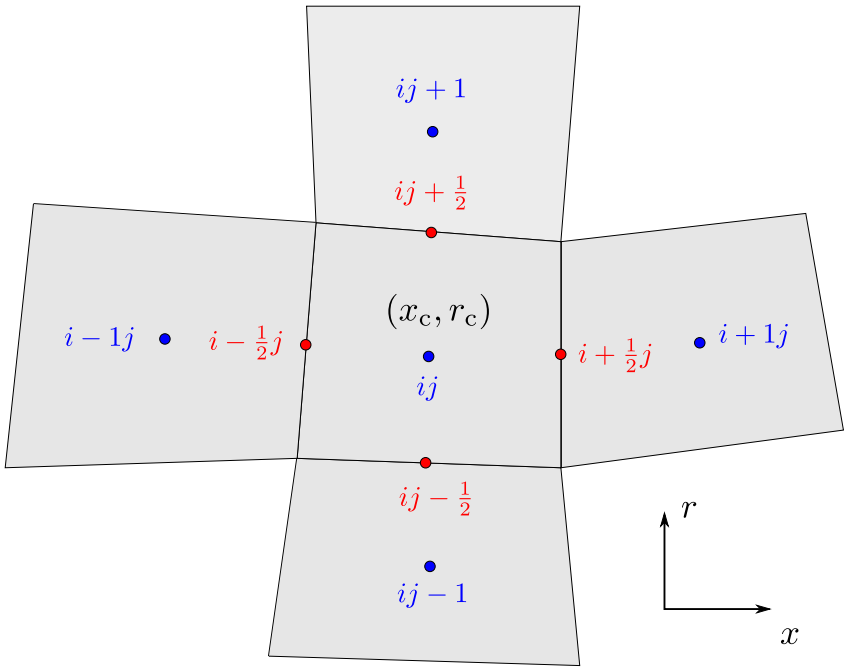
Numerical method (method-of-lines)

Space discretization: Cell-Centered Finite Volume method

Time integration: Implicit/Explicit (IMEX) dual time-stepping

- t: physical time
- τ: addition time variable to march between *discrete* physical time-levels



$$V_{ij}\left(\frac{\partial \mathbf{U}_{ij}}{\partial \tau}+\frac{\partial \mathbf{U}_{ij}}{\partial t}\right)=\underbrace{(\mathbf{C}_{ij}+\mathbf{D}_{ij}+\mathbf{S}_{ij})}_{\mathbf{R}_{ij}}$$

IMEX approach

- Implicit: diffusion $(\mathbf{D})$ + source terms $(\mathbf{S})$ due to kinetics
- Explicit: convection $(\mathbf{C})$

Two-parameter family (θ, ϕ) implicit integrator[1−2]

- n: refers to the physical time t
- k: refers to the additional time-variable τ

$$V_{ij}\left(\frac{\mathbf{U}_{ij}^{n+1,k+1}-\mathbf{U}_{ij}^{n+1,k}}{\Delta\tau_{ij}}\right)+V_{ij}\left[\frac{(1+\phi)\mathbf{U}_{ij}^{n+1,k+1}-(1+2\phi)\mathbf{U}_{ij}^{n}+\phi\mathbf{U}_{ij}^{n-1}}{\Delta t}\right]=$$
$$\theta\Big(\underbrace{\mathbf{D}_{ij}^{n+1,k+1}+\mathbf{S}_{ij}^{n+1,k+1}}_{\text{implicit}}+\underbrace{\mathbf{C}_{ij}^{n+1,k}}_{\text{explicit}}\Big)+(1-\theta)\mathbf{R}_{ij}^{n}$$

$\Rightarrow$ *solution found via linearization around* $\mathbf{U}^{n+1,k}$ *(leads to a sparse system). At convergence (i.e.,* $\mathbf{U}^{n+1,k+1}\simeq\mathbf{U}^{n+1,k}$*)* one finds

$$V_{ij}\left[\frac{(1+\phi)\mathbf{U}_{ij}^{n+1}-(1+2\phi)\mathbf{U}_{ij}^{n}+\phi\mathbf{U}_{ij}^{n-1}}{\Delta t}\right]=\theta\mathbf{R}_{ij}^{n+1}+(1-\theta)\mathbf{R}_{ij}^{n}$$

[1] Pulliam, T. H., AIAA 1993–3360

[2] Munafò, A., Alberti, A., Pantano, Freund, J. B., Panesi, M., in preparation for submission to *J. Comput. Phys.*

CFL tool: HEGEL

High fid**E**lity tool for ma**G**n**E**to gas dynamics simu**L**ations

- Originally developed in the AR department at UIUC
  [NASA grant NNX15AQ57A]

Language

- Source code written in modern `Fortran` 90/2008

Parallelization

- Domain decomposition via MPI + PETSC (*e.g.*, `DMDA`,`Mat`,`Vec`)

Discretization

- Space: *cell centered Finite Volume method*
  - High-order reconstruction: MUSCL, WENO5, MP5
- Time: *explicit, implicit, splitting*
  - Forward Euler, RK4, TVD RK2/3
  - Backward Euler (full matrix and DPLR)
  - Three-point backward + dual-time stepping
  - Operator splitting (*e.g.*, Strang)

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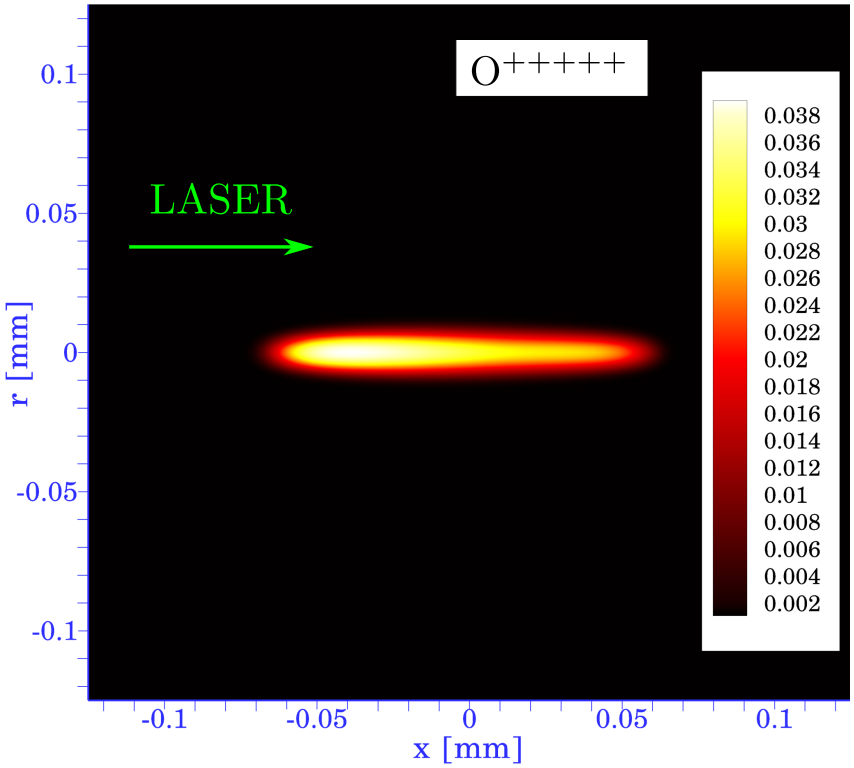
Laser induced breakdown in molecular Oxygen

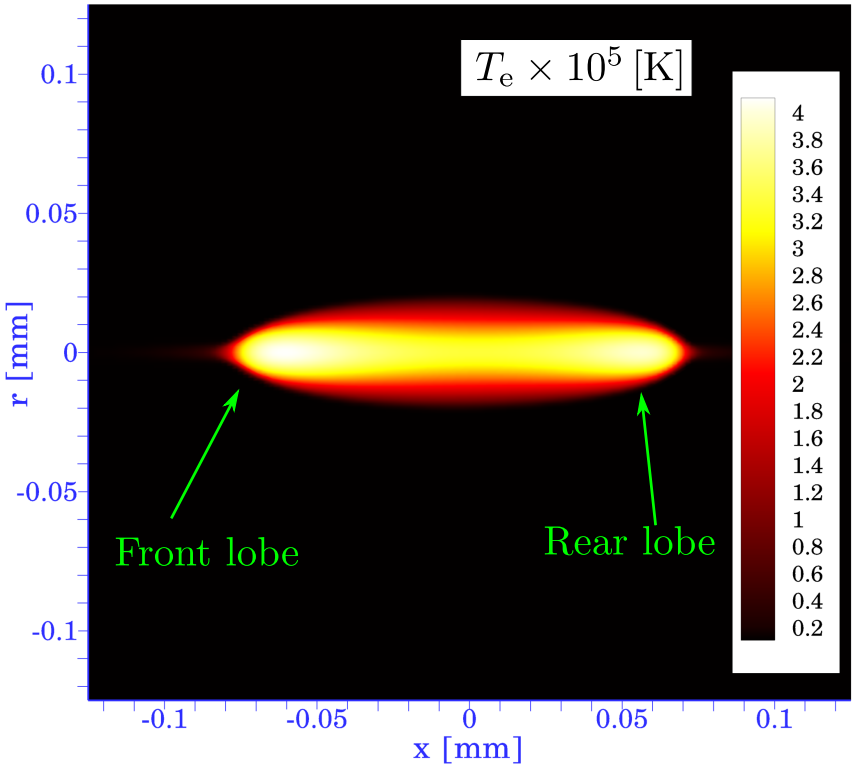
Simulation parameters

- Mixture: O_2 (+ electrons and multiply charged ions)
- $p_\mathrm{a} = 99.4\,\mathrm{kPa}$, $T_\mathrm{a} = 293.5\,\mathrm{K}$ [quiescent gas]
- $E_\mathrm{l} = 50\,\mathrm{mJ}$, $\lambda_\mathrm{l} = 532\,\mathrm{nm}$, $\mathrm{FWHM} = 7.5\,\mathrm{ns}$, $r_\mathrm{F} = 7.5\,\mu\mathrm{m}$

Degree of ionization

- Two plasma waves observed: *forward* and *backward* [$t = 5$ ns]





Proposed explanation in the literature*

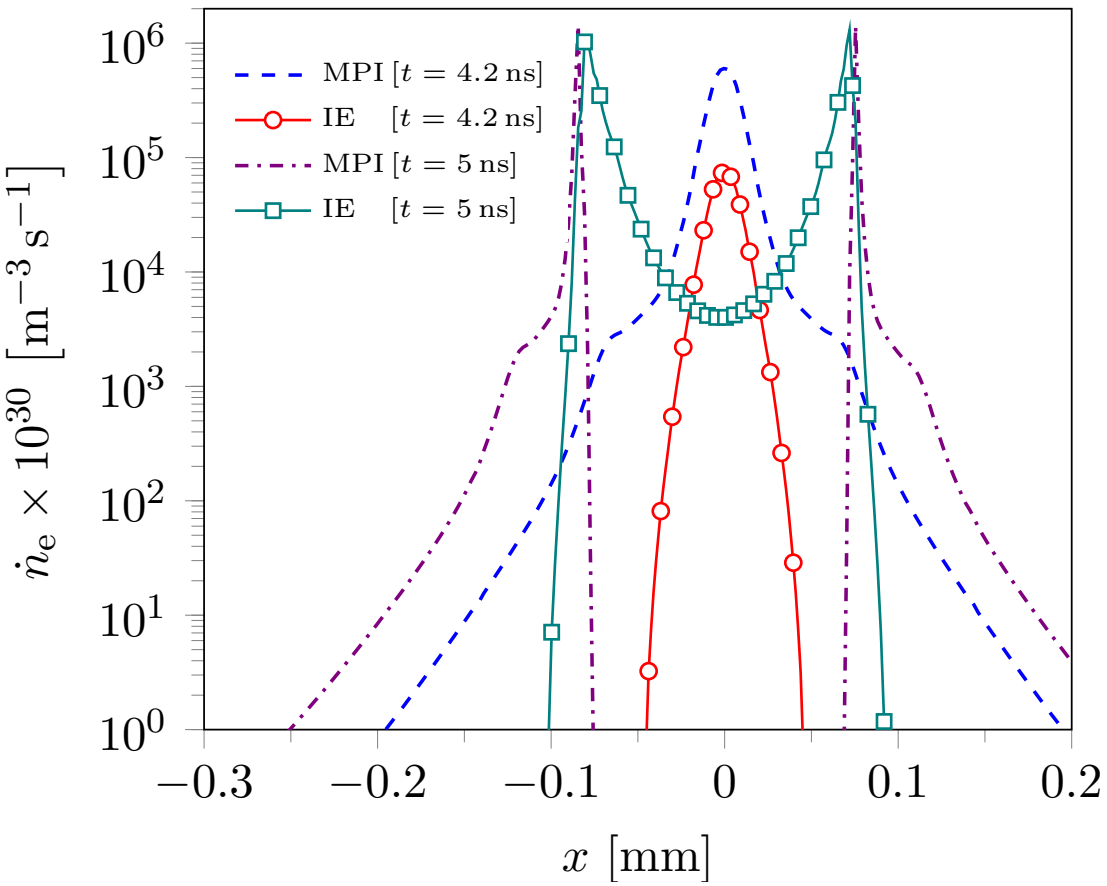
- Backward: *breakdown* wave (formulated originally by Raizer)
- Forward: *breakdown* wave due to beam re-focusing

* N. Tsuda, J. Yamada, *J. Appl. Phys.* **87**, 2122, 2000

Proposed plasma wave mechanism

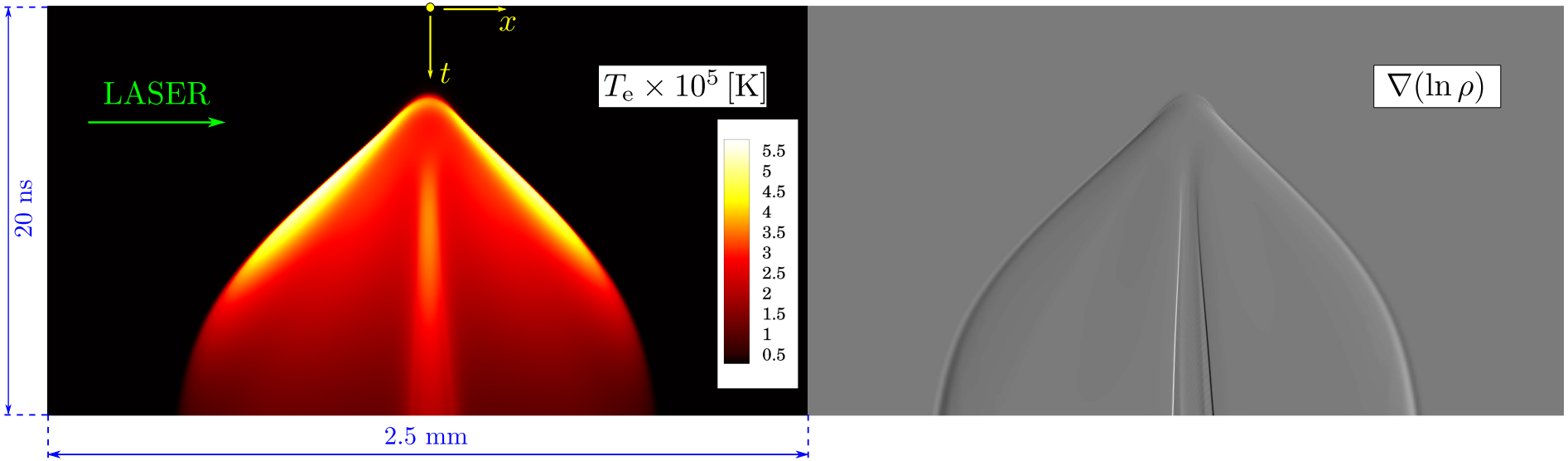
Radiation supported wave

- Triggered by MPI
- Sustained by IE and FF absorption



Plasma boundary evolution

- x-t diagram
  - Left: free-electron temperature
  - Right: Schlieren-like image from density gradient



Comparison with experimental emission images

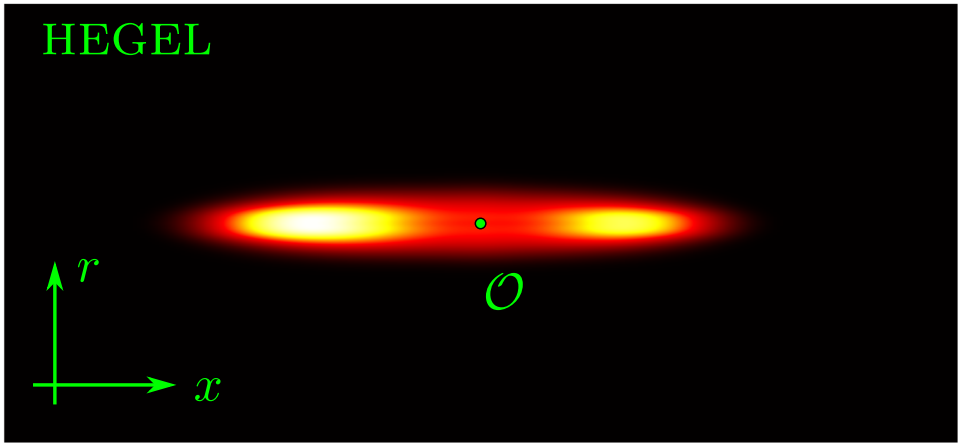
- Experiments from Jon Retter (UIUC, now SNL)

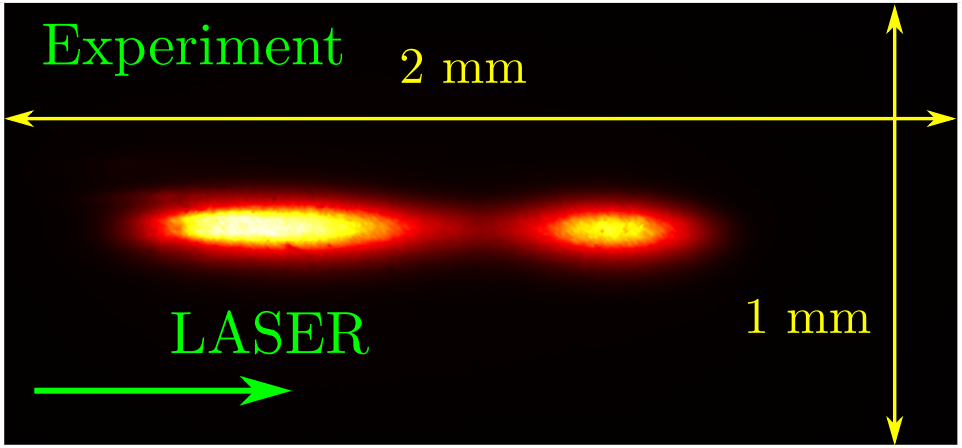
O_2 LIB

$p_a = 99.4$ kPa

$T_a = 293.5$ K

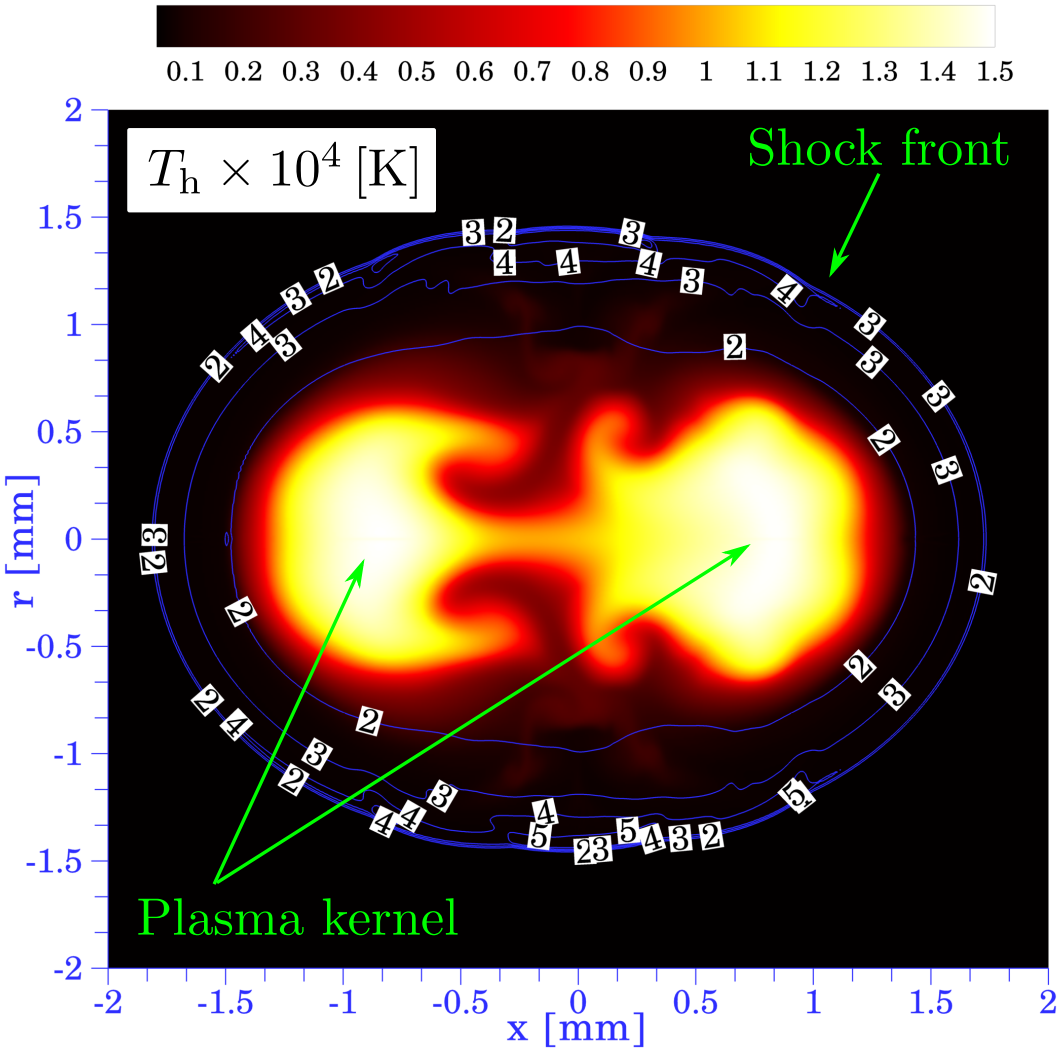
$E_{laser} = 50$ mJ





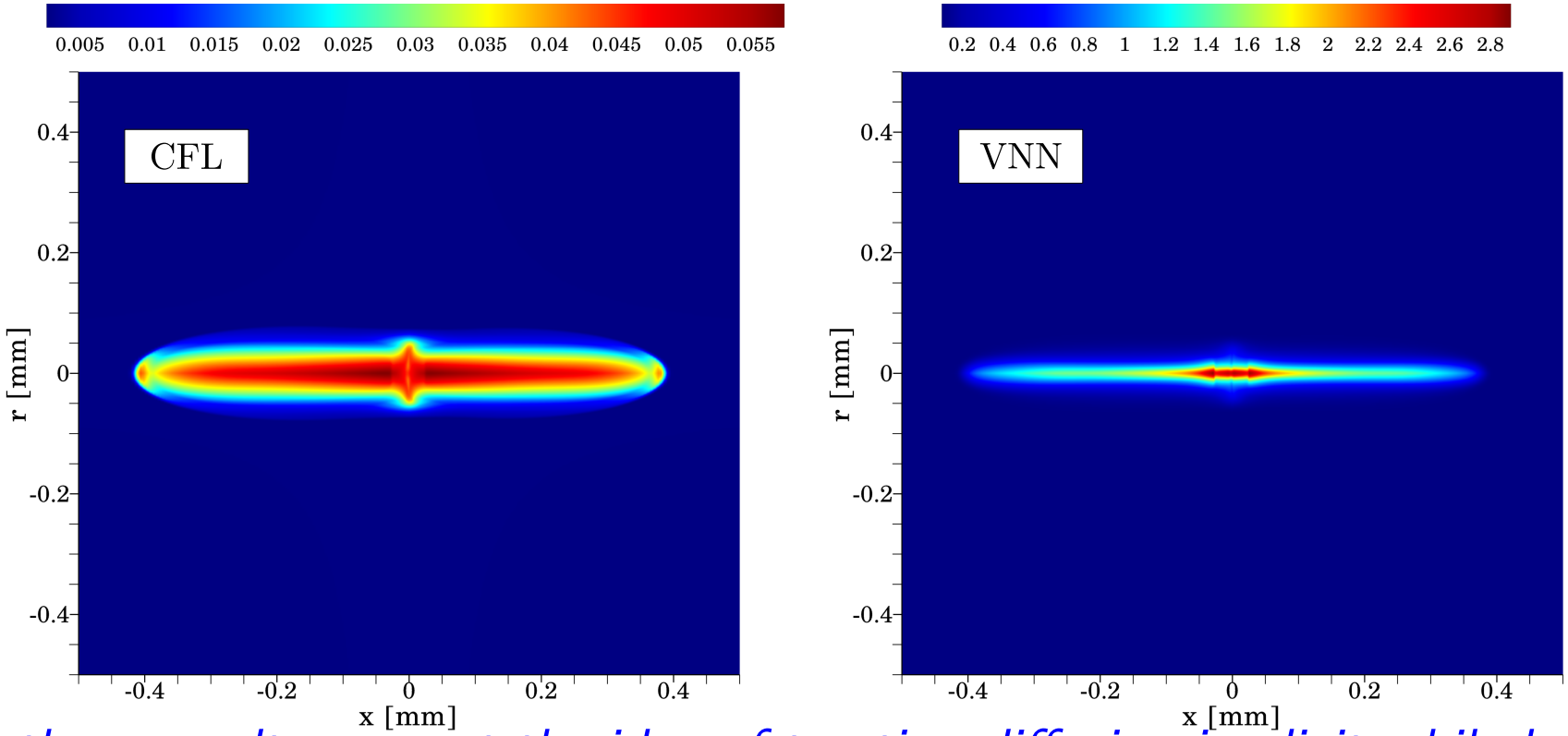
Post-breakdown evolution

- $t \simeq 1\,\mu s$



Stability considerations (CFL and VNN maps)

- $t = 10\,\text{ns}, \quad \Delta t = 0.5\,\text{ps}$
- $\text{CFL} \ll 1$ whereas VNN above linear stability bounds (0.5) due to large values of thermal conductivity of free-electrons



⇒ *these results support the idea of treating diffusion implicit while leaving convection explicit*

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Accomplishments

- Developed physics-based model for LPI
- Proposed plasma wave mechanism to explain observed *forward* and *backward* plasma waves

Future/on-going work

- Validation through comparison with experiments

  ⇒ Alberti *et al.* talk [28] (Tuesday March 26th)

- Assessment of plasma wave mechanism
- Add non-collimated radiation (*e.g.*, line/*continuum* emission)

  ⇒ Sahai *et al.* talk [41] (Wednesday March 27th)

- Develop Maxwell solver for laser beam

Thank you for your attention, Questions?