Spectroscopic Investigation of a Repetitively-Pulsed Nanosecond Discharge

by

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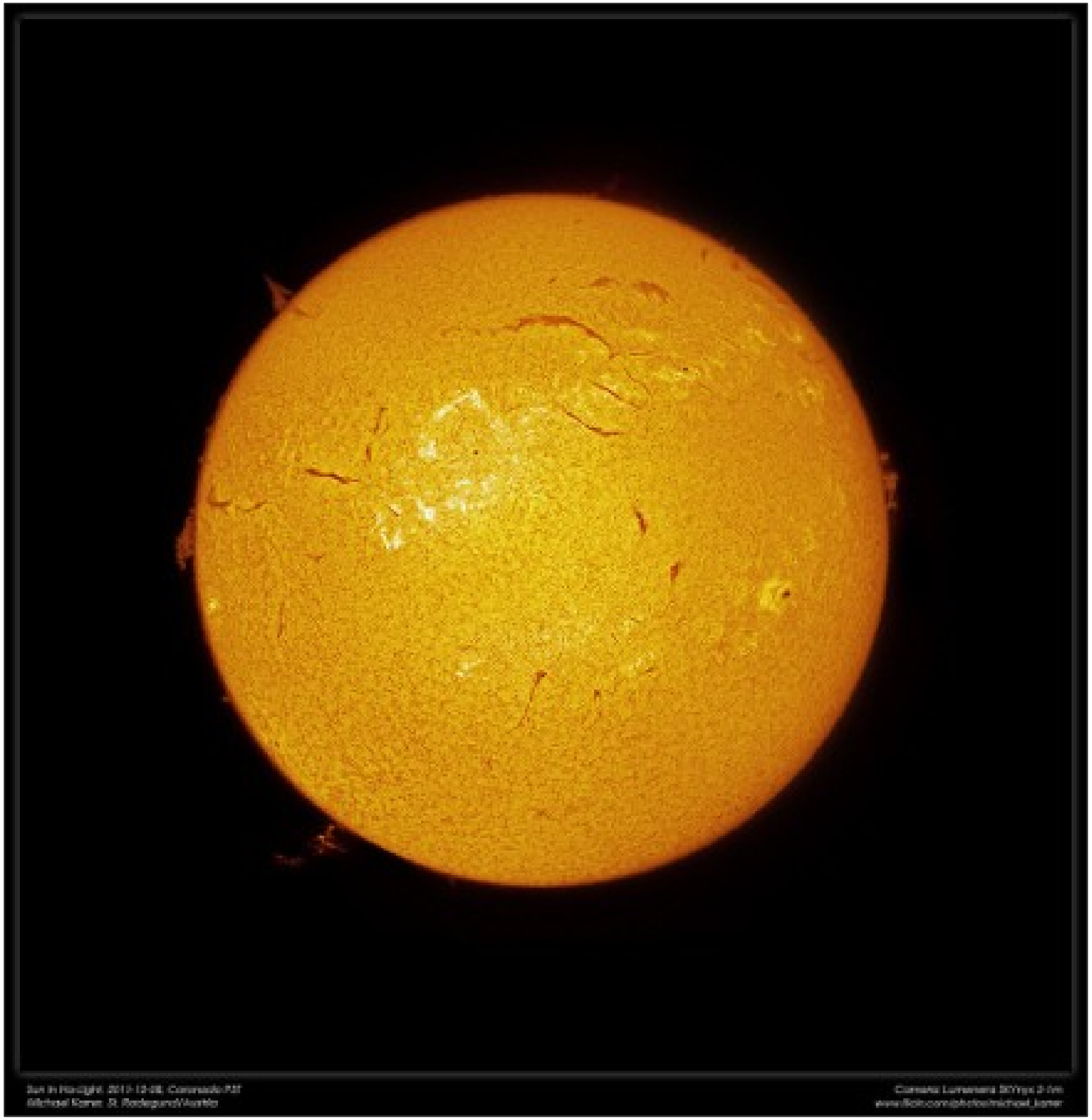
- Introduction
- Outstanding questions in RPNDs
- Metastable measurements
- Global model
- Emission measurements
- Conclusions

Introduction



Karrer, M. "Sun 2011-12-08." URL: http://www.flickr.com/photos/michael_karrer/6487199145/, Accessed: August 26, 2013.

What Is a Plasma? (Sun)

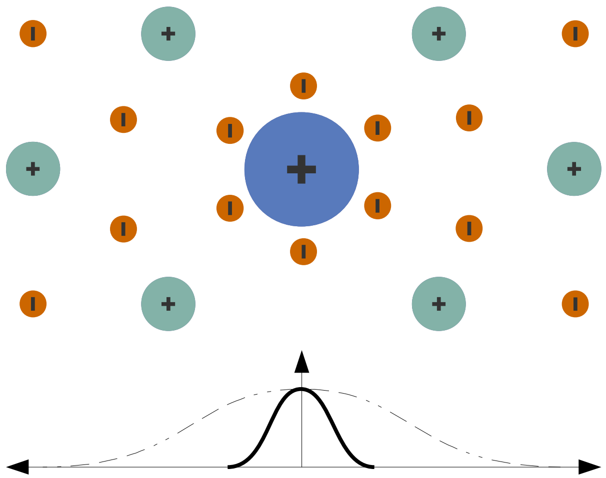
Hunt, P. "Lightning." URL: http://www.flickr.com/photos/michael_karrer/6487199145/, Accessed: Aug. 26, 2013.

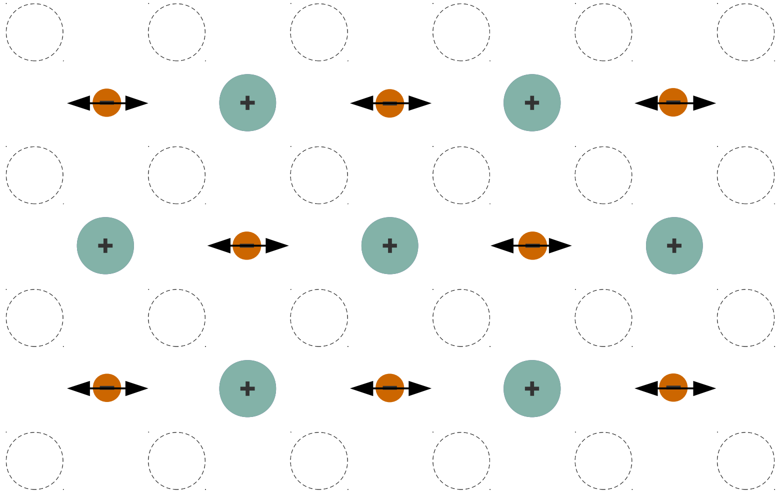
What Is a Plasma? (Lighting)

NASA, ESA, and the Hubble Heritage Team. "Star-Forming Region LH 95 in the Large Magellanic Cloud." URL: http://hubblesite.org/gallery/album/entire/pr2006055a/, Accessed: August 27, 2013.

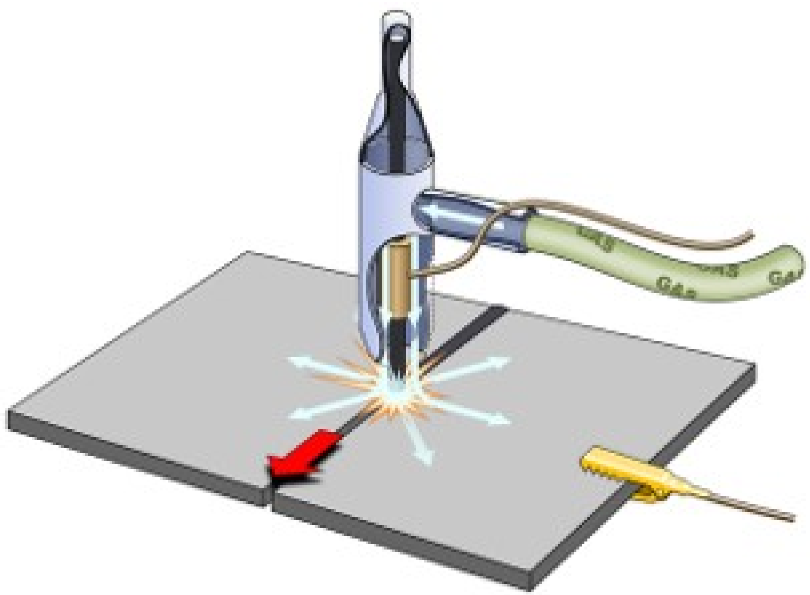
What Is a Plasma? (Interstellar Gases)

- Ionized gas
  - Neutral particles
  - Positive (and negative) ions
  - Electrons
- Exhibits large-scale electrostatic effects
  - Debye shielding
  - Electron oscillations





What Is a Plasma?



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Shigeru23. “Gas arc welding (TIG & MIG).” URL: https://commons.wikimedia.org/wiki/File:Gas_arc_welding_%28TIG_%26_MIG%29.PNG, Accessed: August 27, 2013.
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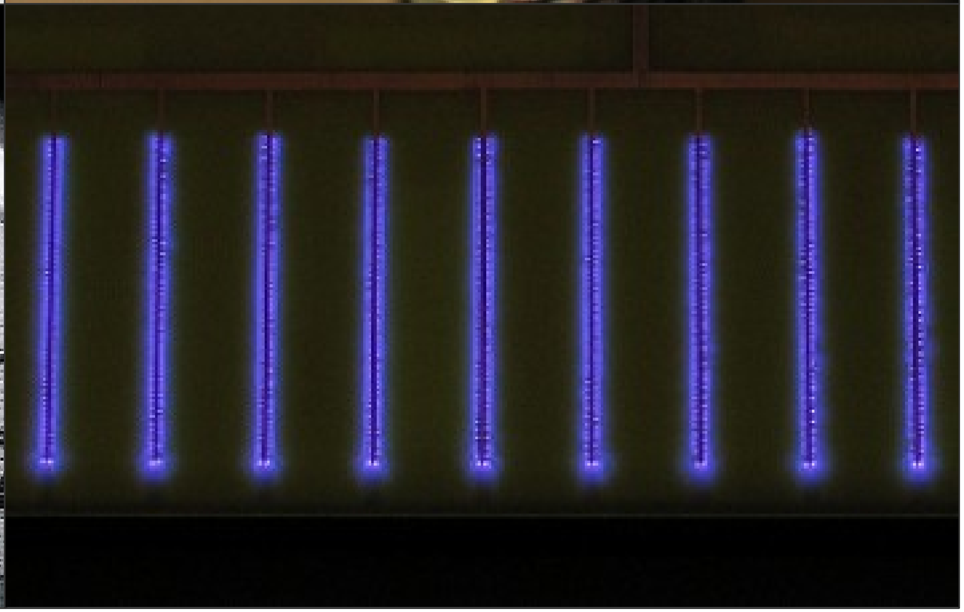
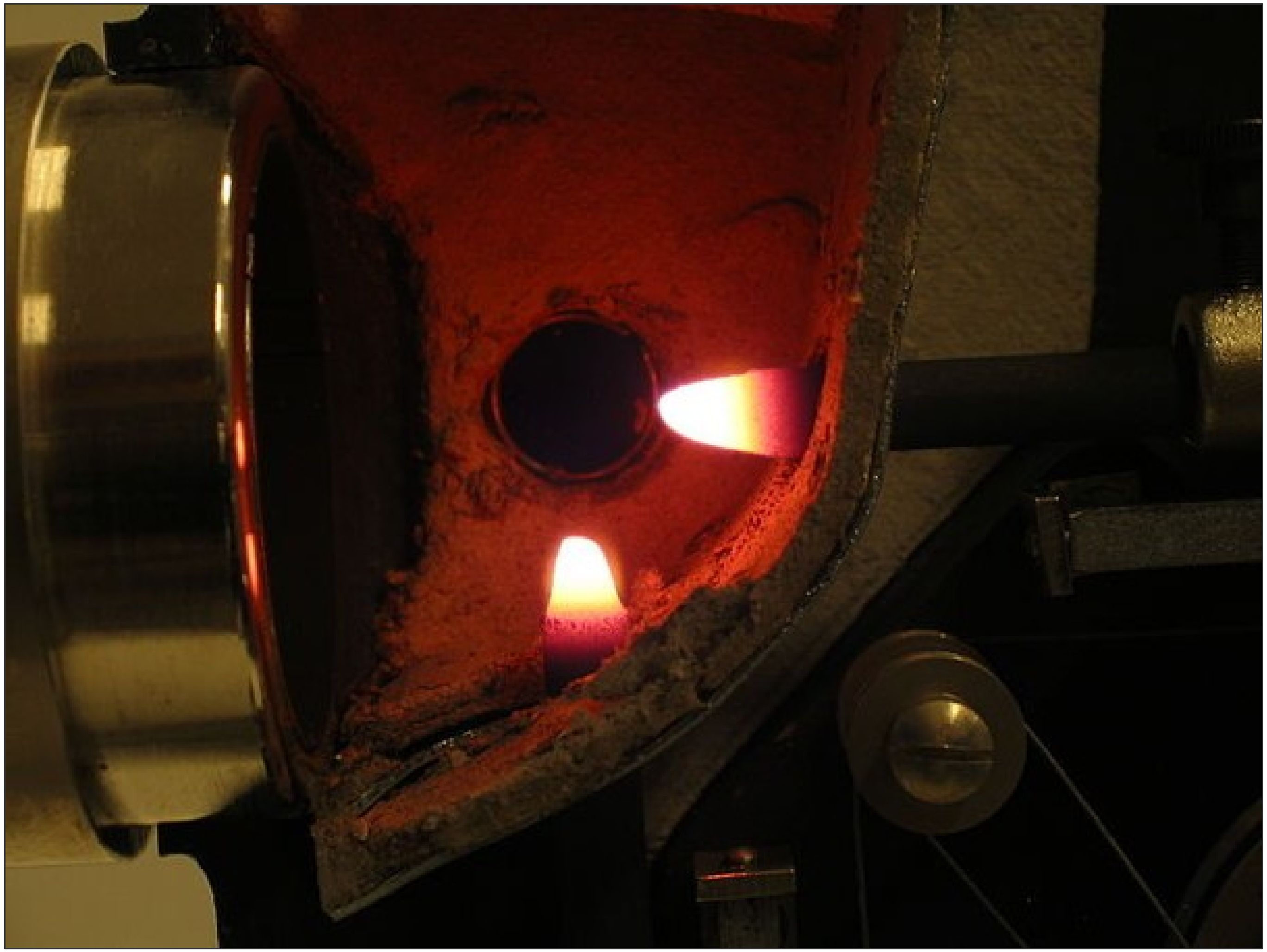
Applications



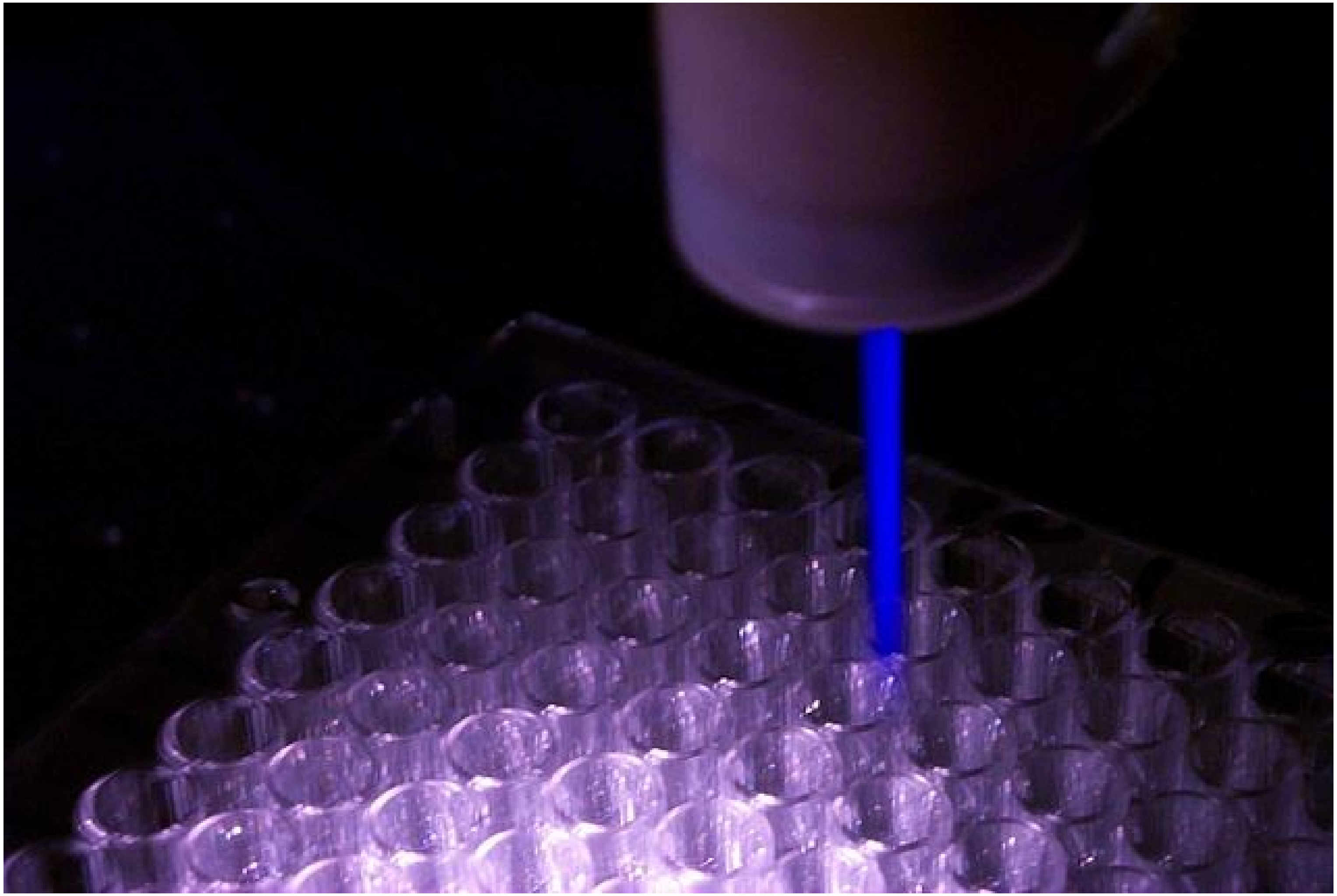
Bellay, C. “X-51A Waverider on B-52 2009.” URL: https://commons.wikimedia.org/wiki/File:X-51A_Waverider_on_B-52_2009.jpg, Accessed: August 27, 2013.
Wong, N. “Tip of a spark plug.” URL: http://www.flickr.com/photos/14029705@N00/375024452/, Accessed: August 27, 2013.
parfe_alp, “Operation.” URL: http://www.flickr.com/photos/parfe/2239523465/, Accessed: August 27, 2013.
Xunger, “Plasma glow discharge.” URL: https://commons.wikimedia.org/wiki/File:Streamwise.JPG, Accessed: August 27, 2013.

Atmospheric-Pressure Plasma Applications



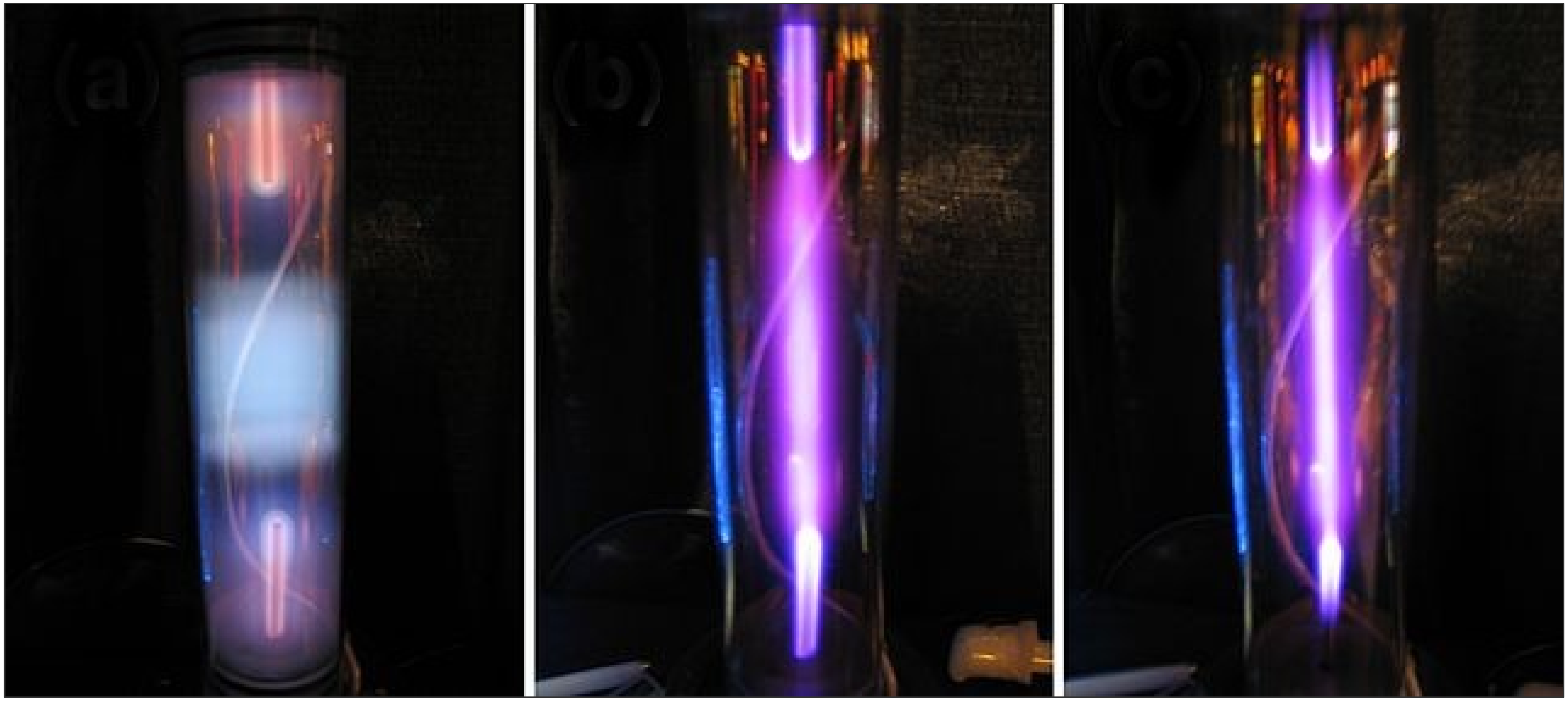
$$T_{neutrals} \sim T_{ions} \sim T_{electrons}$$

D-Kuru/Wikimedia Commons. "Afterglowing electrodes of an arc lamp." URL: https://commons.wikimedia.org/wiki/File:Arc_lamp-afterglow_2_PNr%C2%B00038.jpg, Accessed: August 28, 2013.



$$T_{\text{neutrals}} < T_{\text{ions}} < T_{\text{electrons}}$$

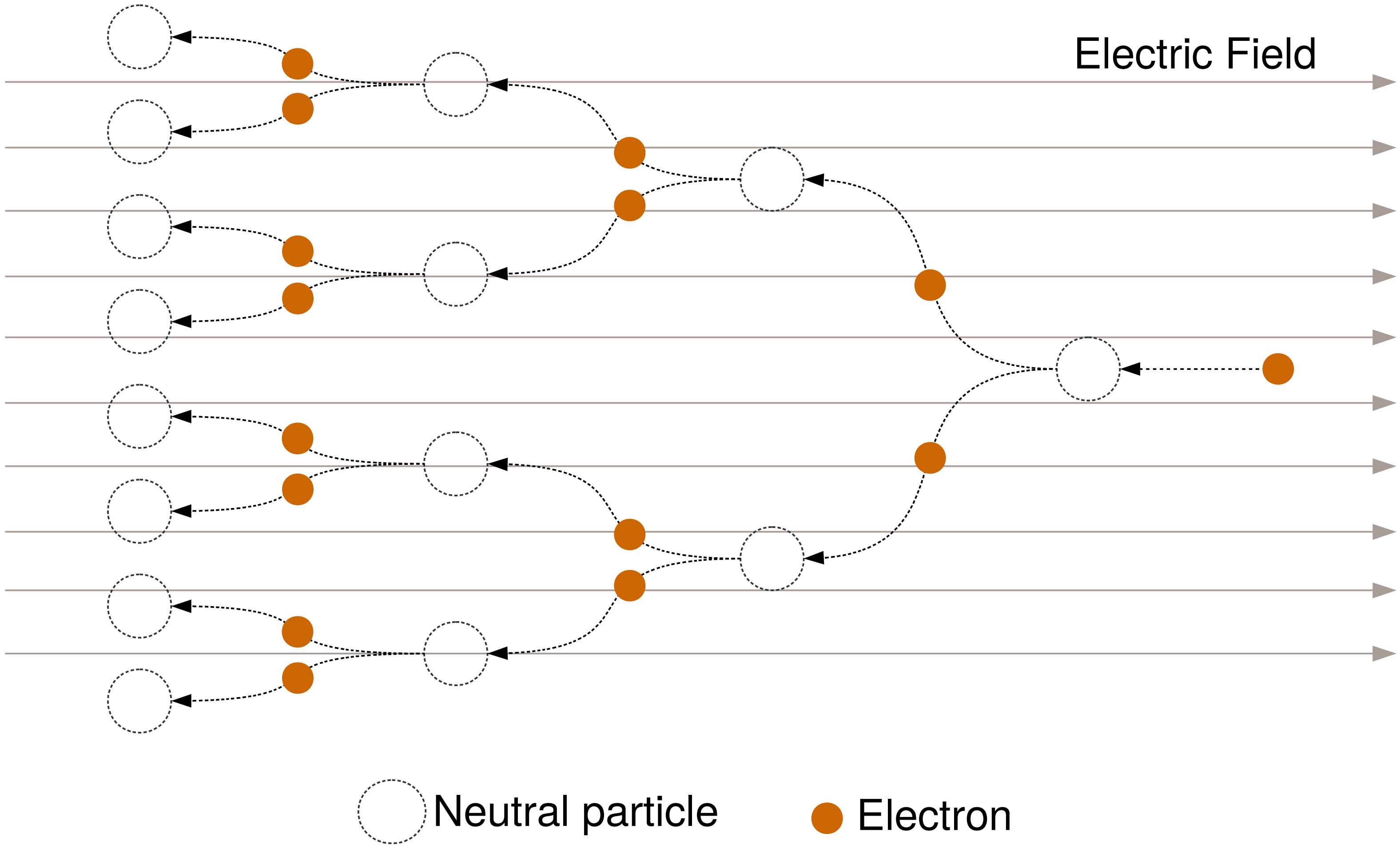
Entropi5. "A low temperature plasma jet: Plasma Pencil." URL: https://commons.wikimedia.org/wiki/File:Plasma_Pencil.jpg, Accessed: August 28, 2013.



- Increase in pressure → tendency to thermalize
- Caused by ionization instability
- Prevented by careful control of power input

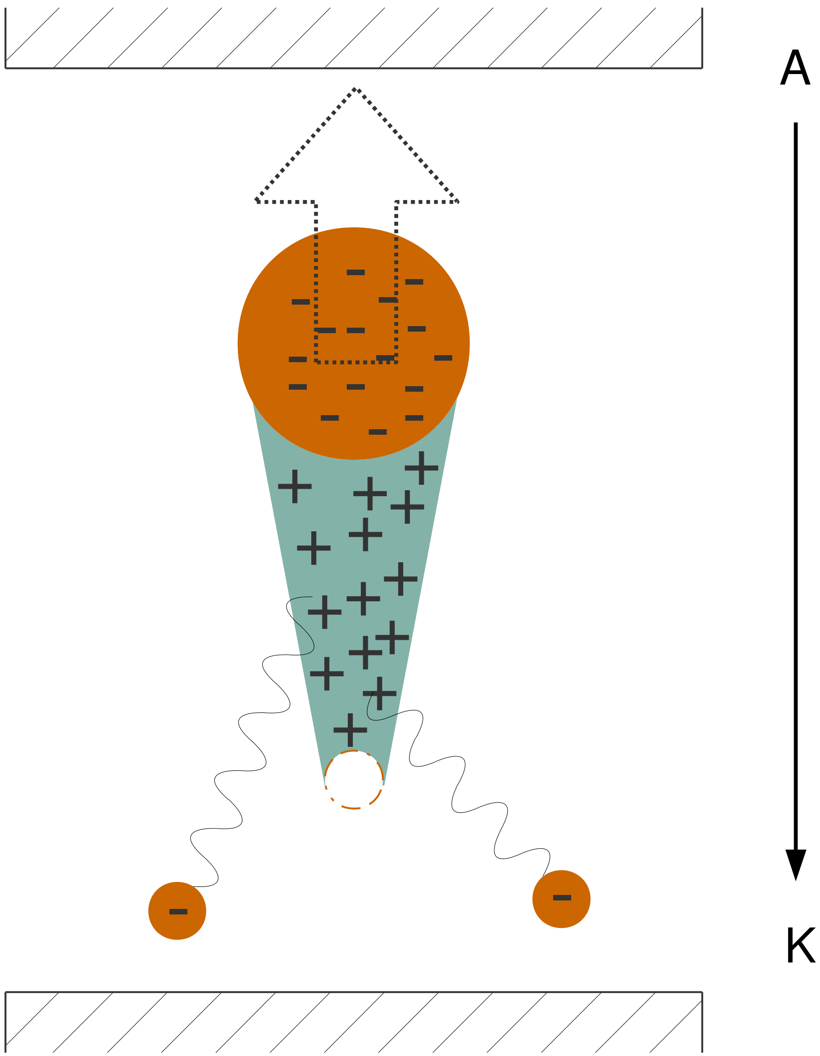
Wikigan, "Transition from a glow discharge in Argon, to an arc." URL: https://commons.wikimedia.org/wiki/File:Glow2arc.jpg, Accessed: August 28, 2013.

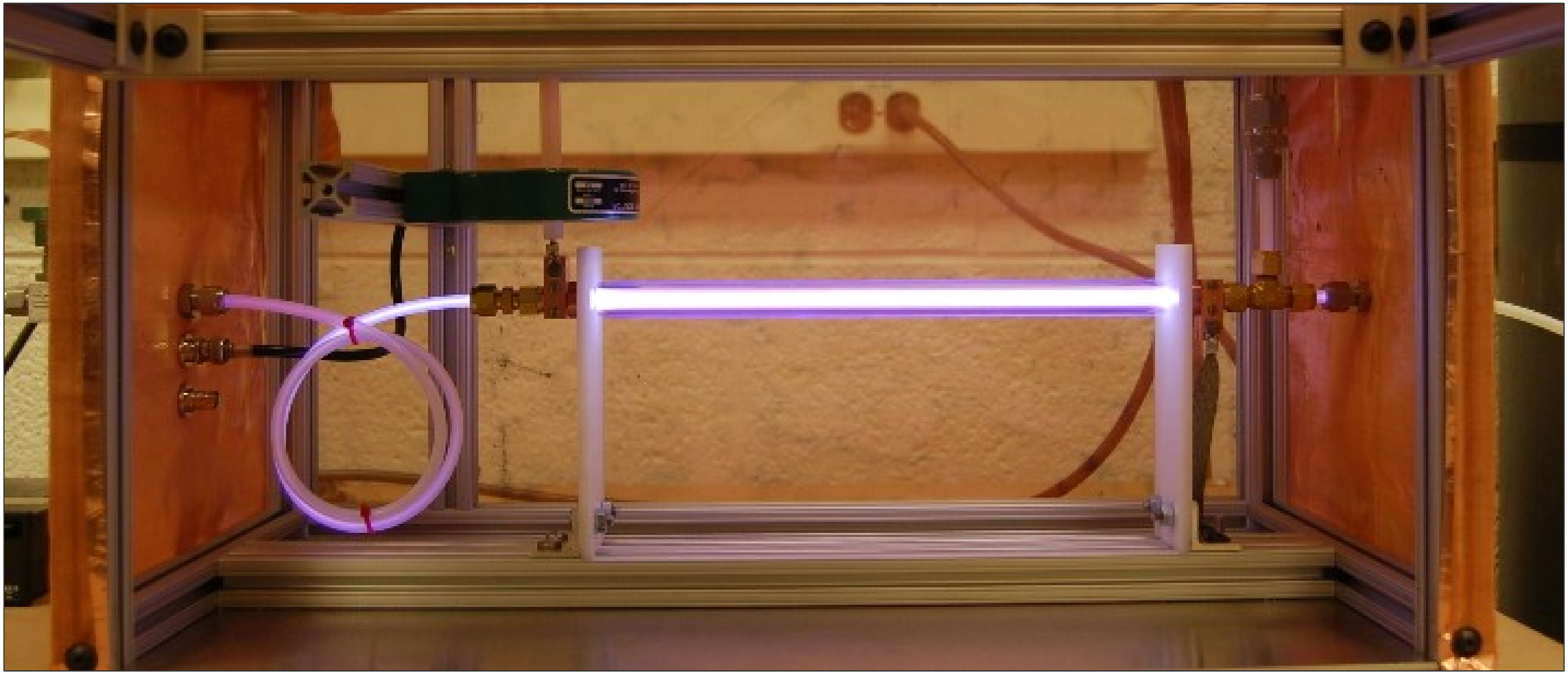
Pulsed Plasmas and Outstanding Questions



- Initial electron from cosmic rays, UV light, previous pulse, ...
- Sufficient electric field → exponential growth

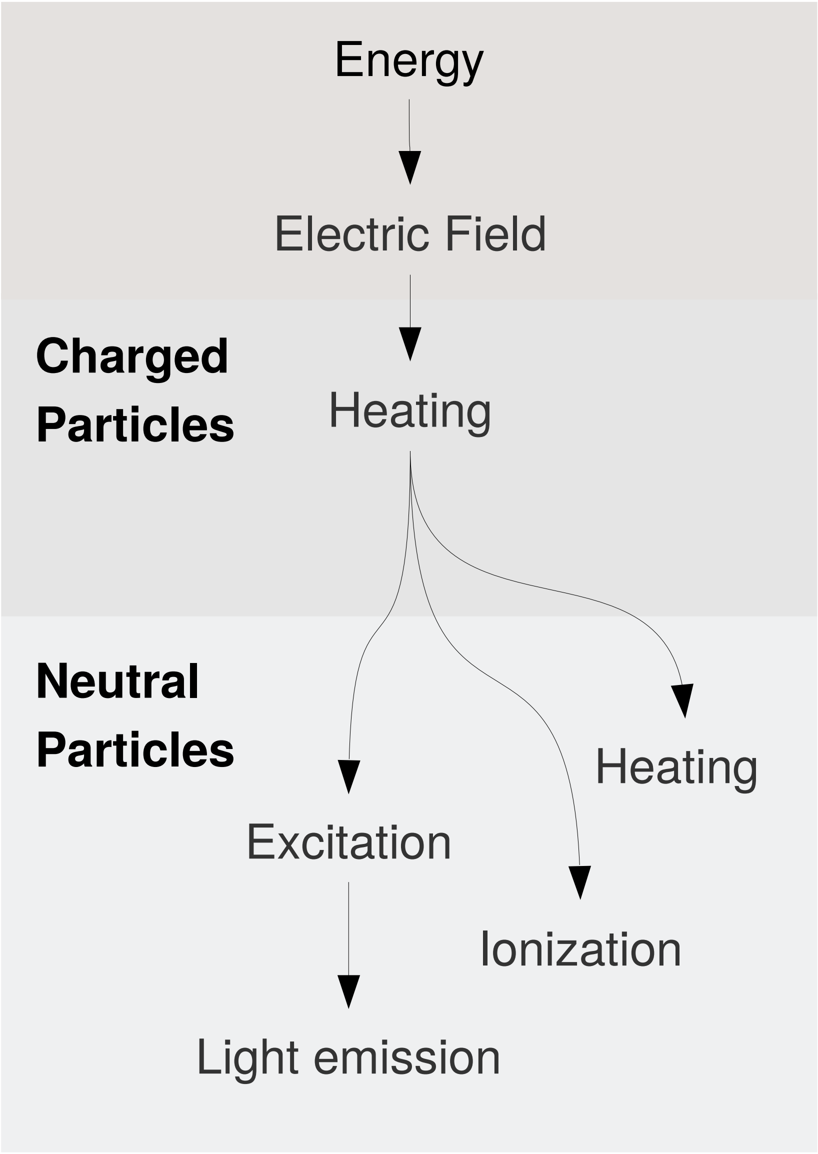
- Internal field of avalanche comparable to applied field
- Reduces energy transfer to streamer
- “Injected” electrons and photoionization become important





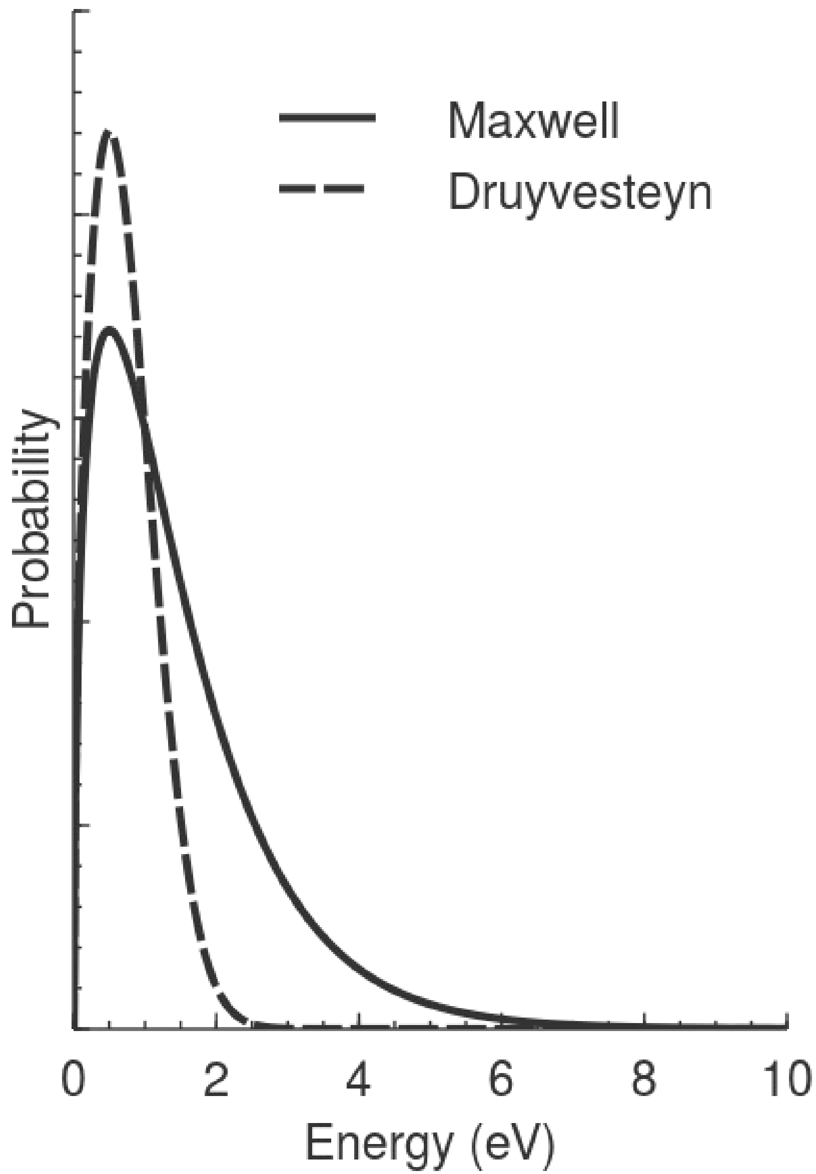
- Similar processes to streamer discharge
- Short pulse-widths: 0 – 100 ns
- High voltages: 1 – 100+ kV
- Moderate repetition rates: 1 – 100 kHz

Repetitively-Pulsed Nanosecond Discharge

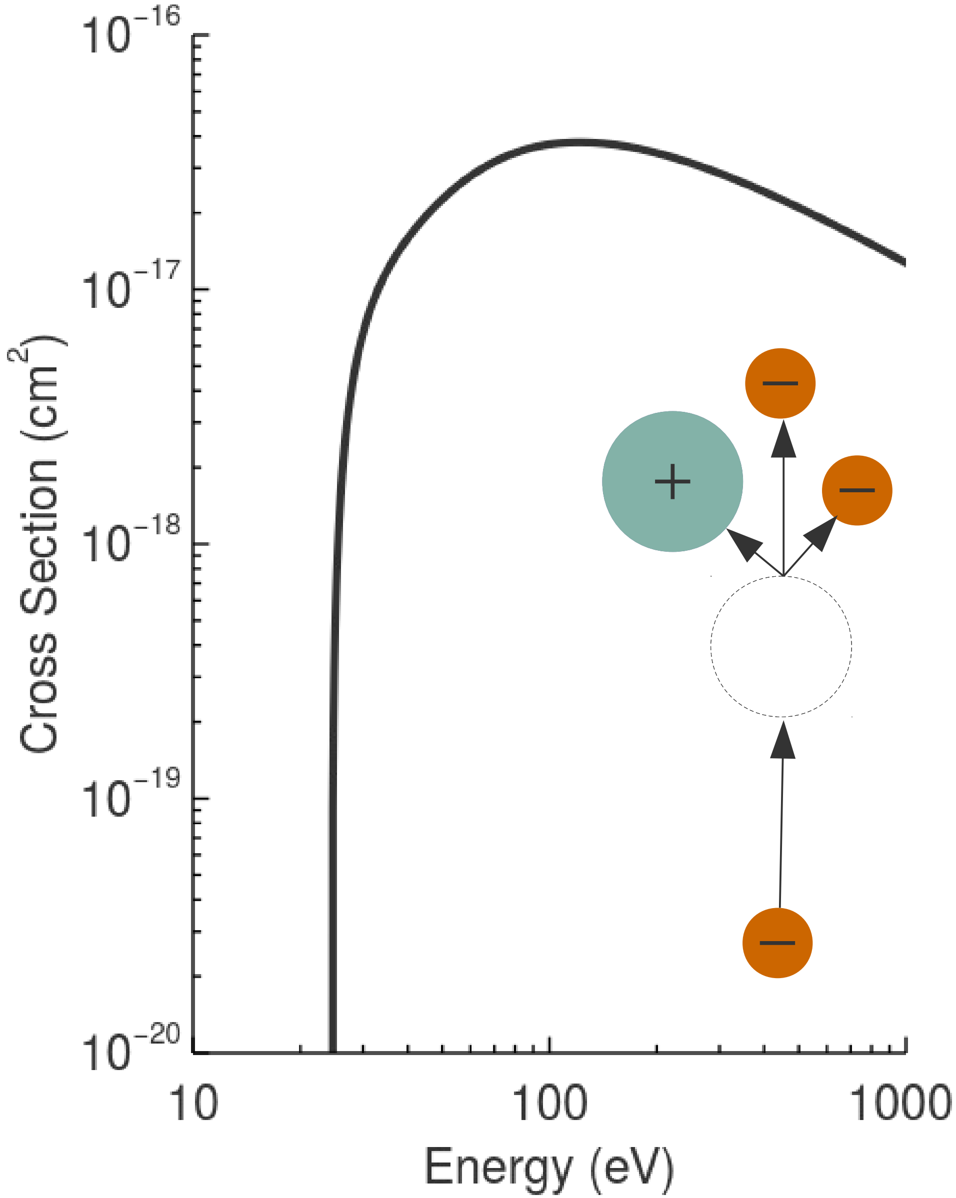


- Energy added to charged particles via electric field
- Fraction of energy heats electrons
- Remainder enters neutral particles through collisions
- How is energy divided?

- Continuous probability distribution for electron energies
- Analytic expressions and approximate calculations possible
- Assumptions may limit use for RPND
-

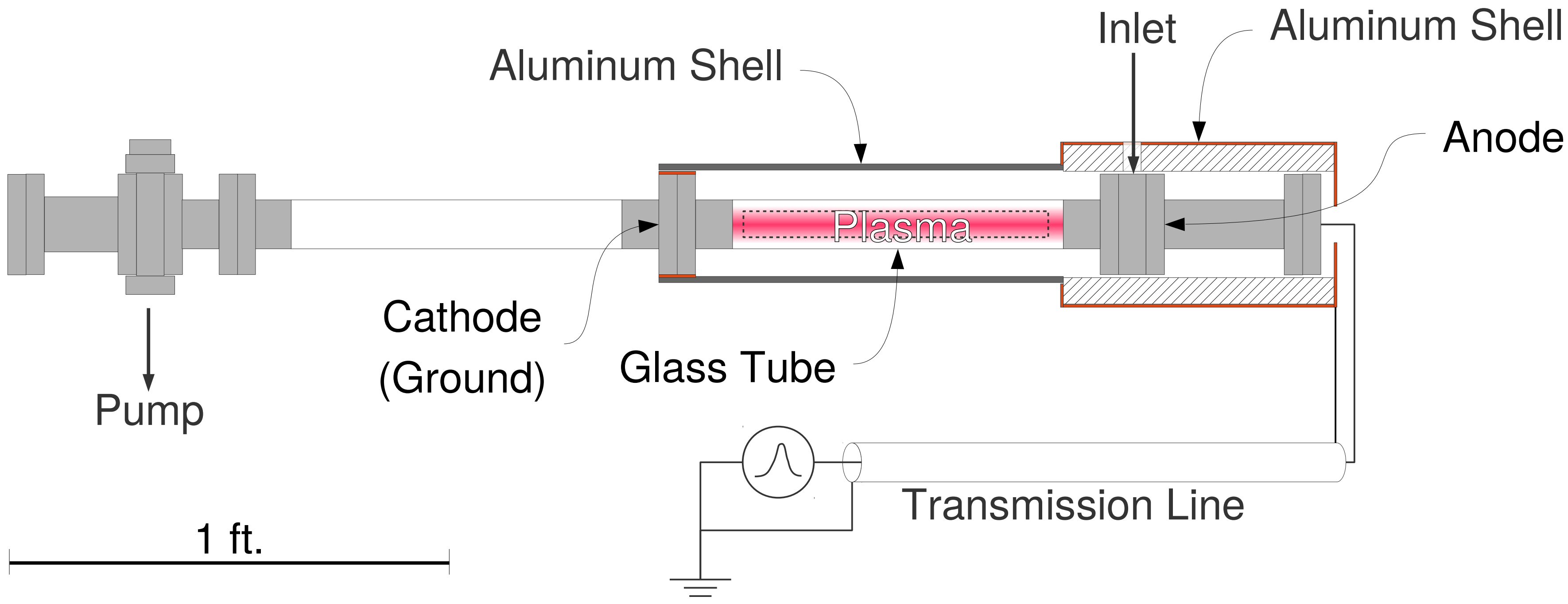


Questions: Electron Energy Distribution



- Plasma products determined by cross sections
- Probability to form a product for a given electron energy

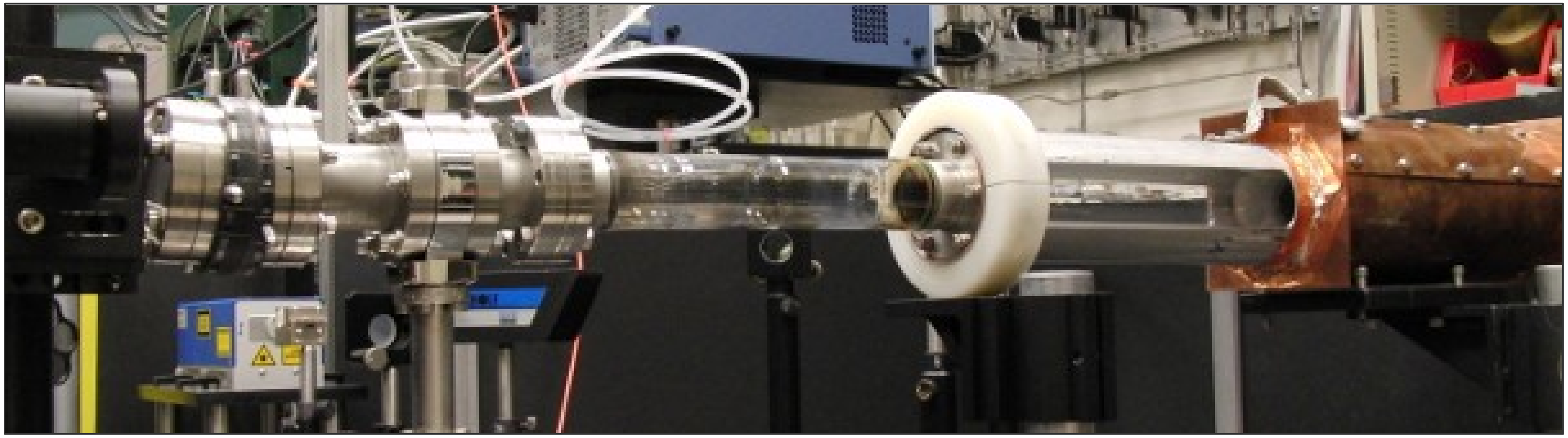
Experimental Setup



Coaxial-type geometry

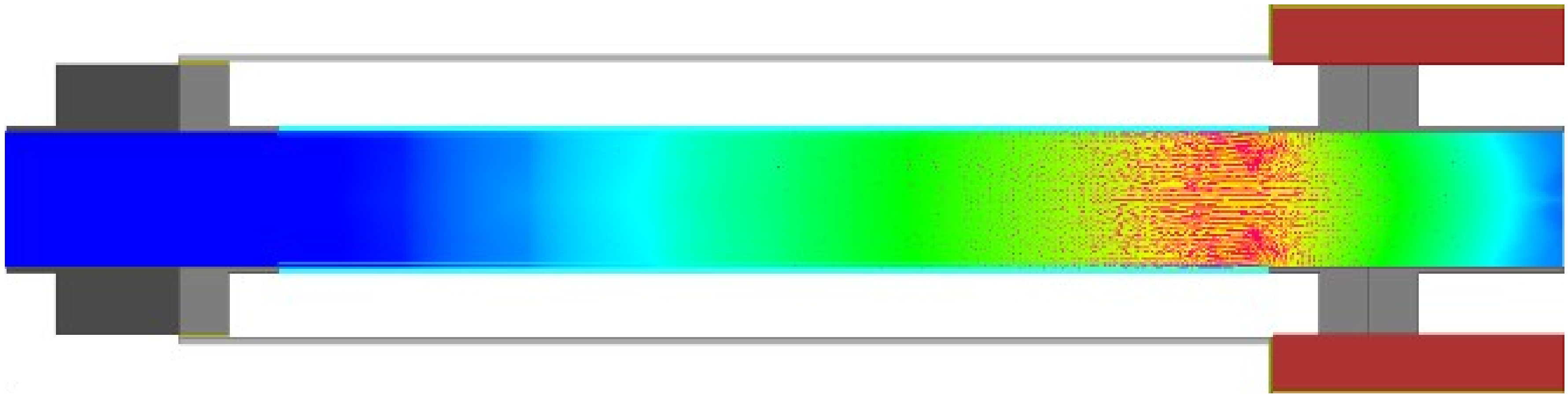
- Inner conductor: plasma
- Dielectric: borosilicate vacuum tube, air, and teflon
- Outer conductor: copper and aluminum shells

Discharge Geometry

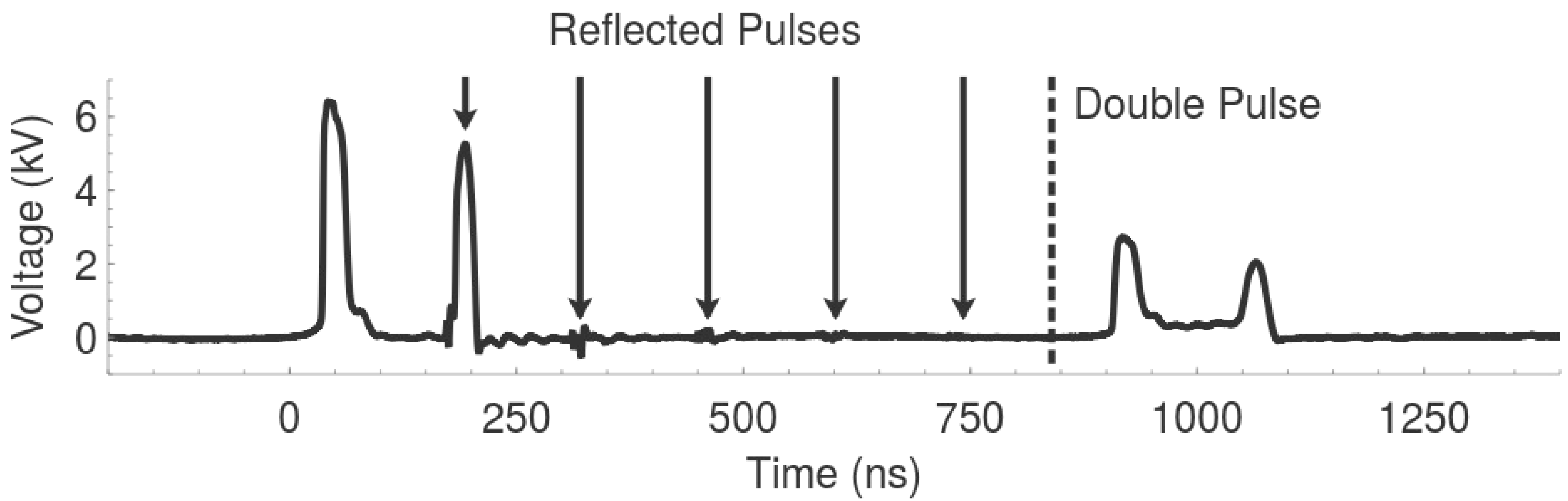


- Ultra-high purity helium: 0.3 – 16.0 Torr
- Rare gas discharges → interesting properties
  - Minimal gas heating
  - UV emissions
- Data available to create detailed population kinetics model
- Voltage pulses: 25 ns FWHM, +6.4 kV, 1.0 kHz

Experimental Setup



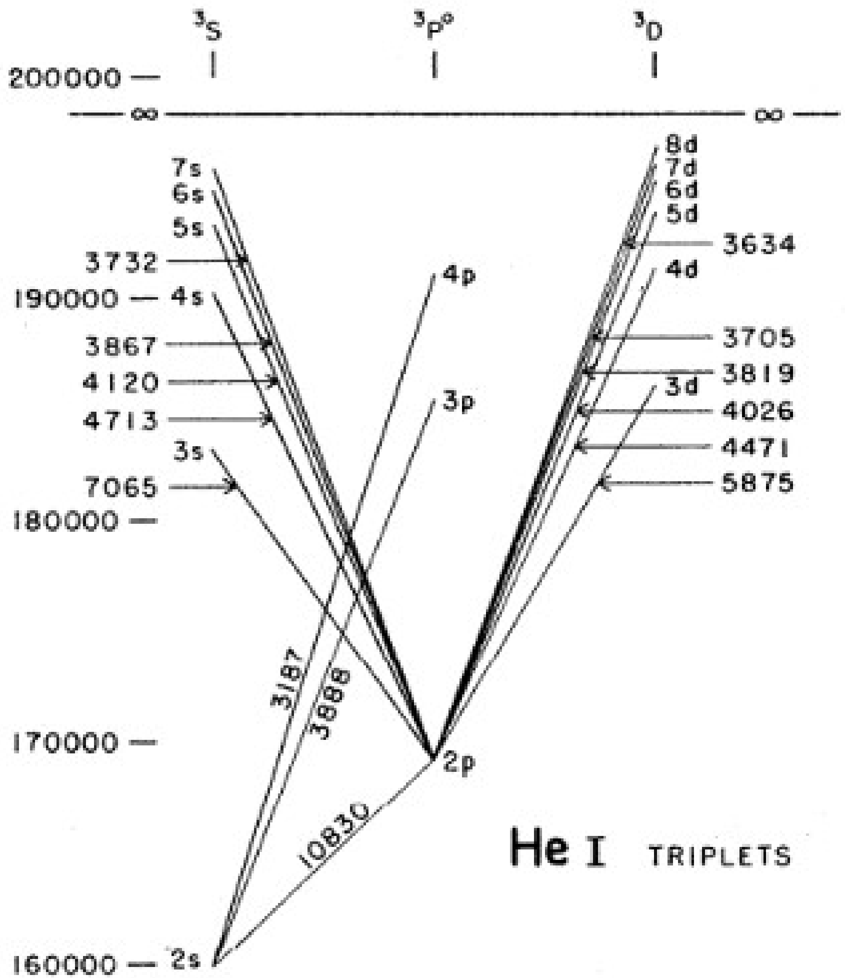
- Peak field: 3.8 kV / cm
- Non-uniform field, concentrated near anode-glass interface
- Notable radial component a result of outer conductor



- Impedance mismatch at anode should cause doubling of voltage
- Long (13 m) transmission line used to isolate incident and reflected pulse

Metastable Measurements

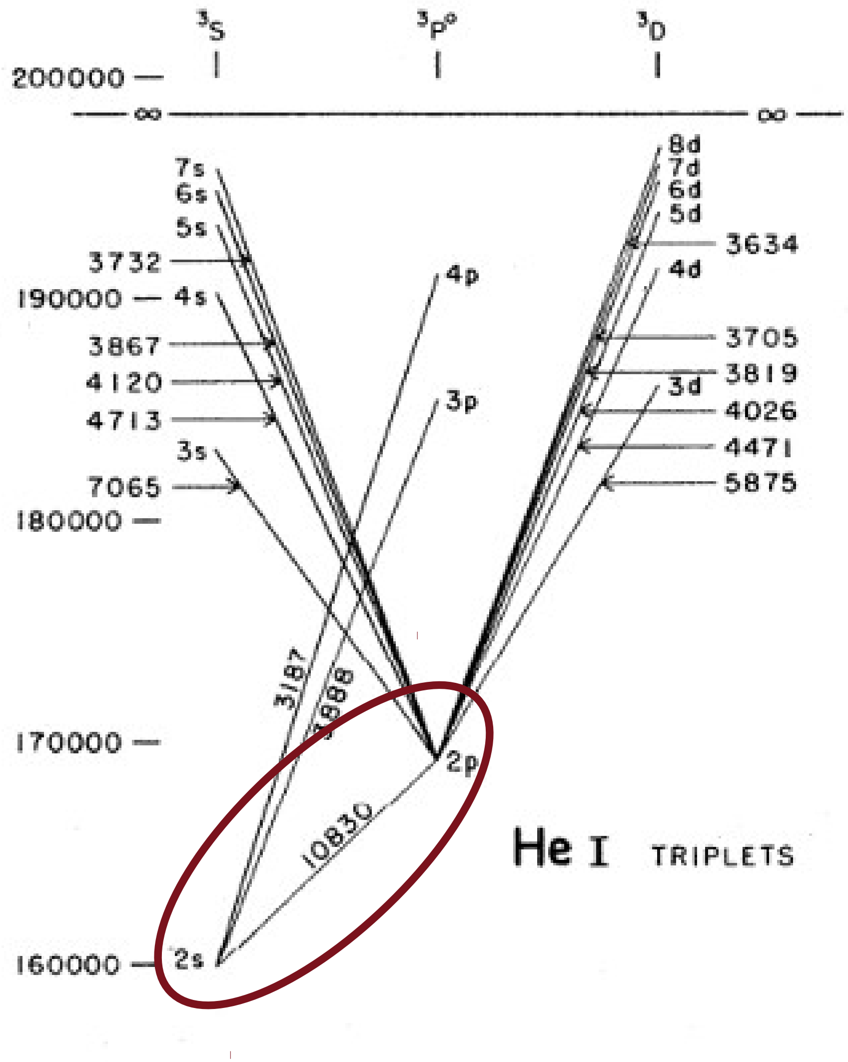
- ~~Physical probes~~
  - Perturb plasma
  - Not fast enough
  - Suggests an <u>optical</u> approach
- ~~Electrons~~
  - No detectable emissions
  - Insufficient density for laser scattering
- <u>Atoms and atomic states</u>



- Absorption of light
  - Photon energy must match energy difference between states
- Time resolution determined by detector
- Fraction of light absorbed proportional to density of lower state

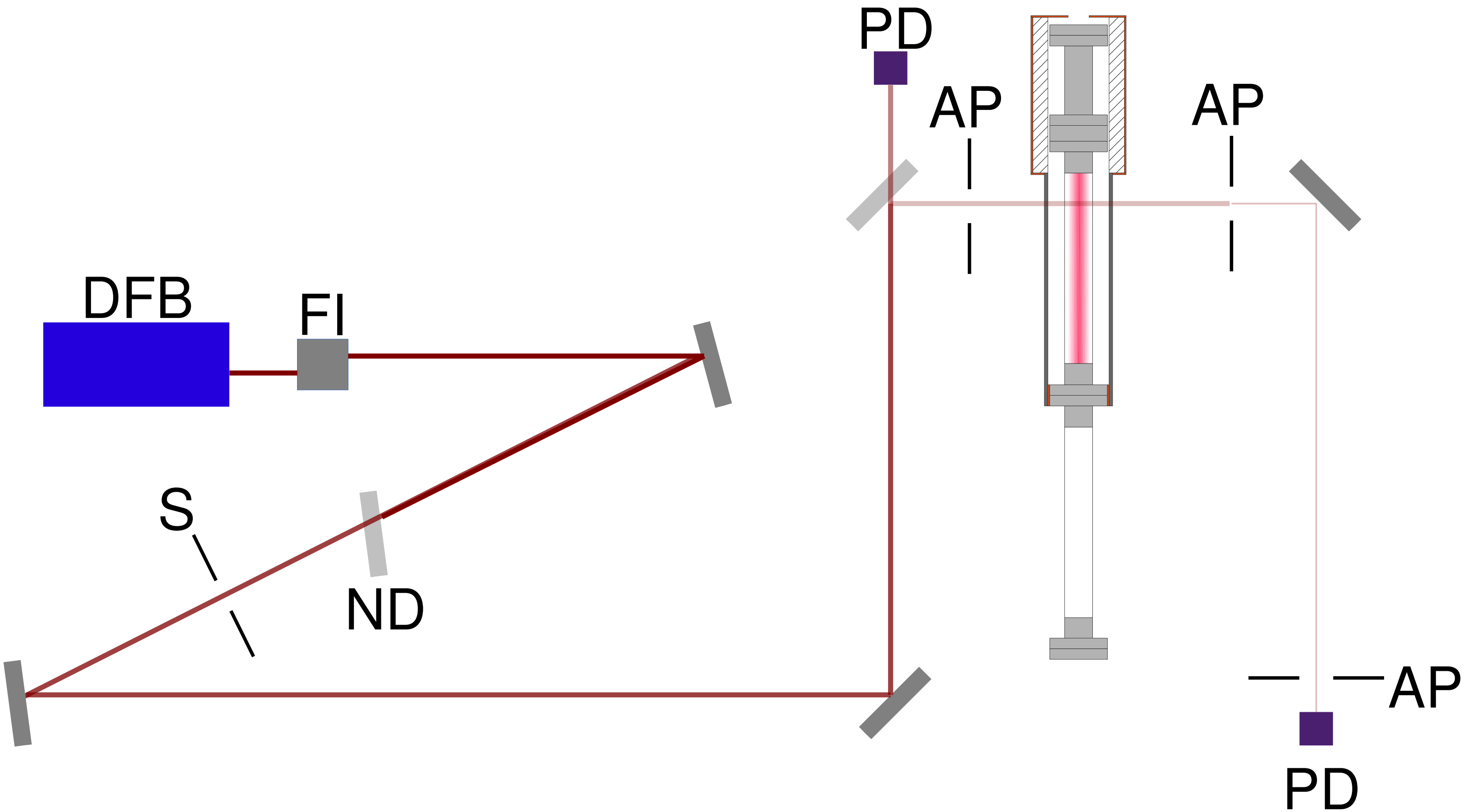
Moore, C. and Merrill, P. "Partial Grotrian Diagrams of Astrophysical Interest." National Bureau of Standards. Washington, D.C. (1968).

Excited States of Helium



- Lowest-energy excited state
- Cannot transition to unexcited state
- Reservoir for energy
- Can cause excitation and ionization long after pulse

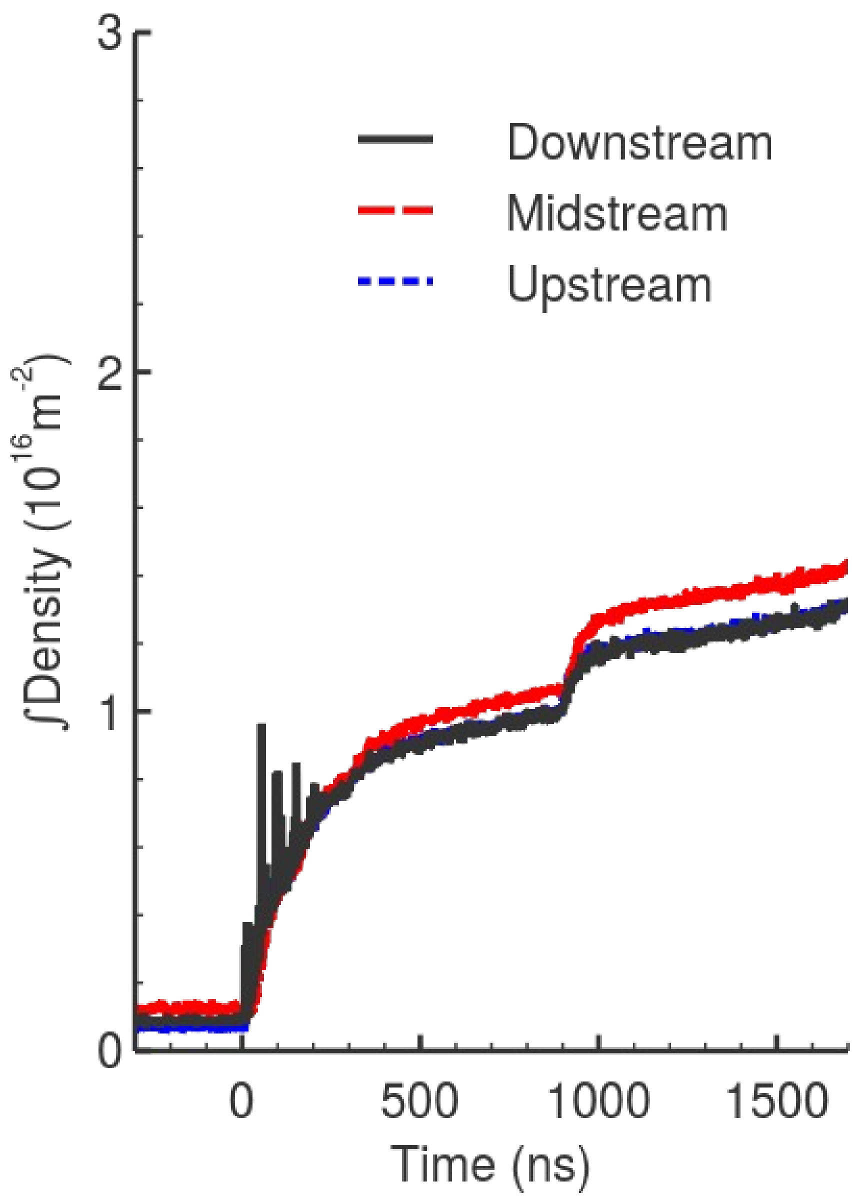
Moore, C. and Merrill, P. "Partial Grotrian Diagrams of Astrophysical Interest." National Bureau of Standards. Washington, D.C. (1968).



- 1083 nm laser diode, $2^3S \rightarrow 2^3P_0$ transition
- Pressures: 0.3, 0.5, 1.0, 2.0, 3.0, 4.0, 8.0, and 16.0 Torr
- Locations: 5.1 cm (Upstream), 12.7 cm (Midstream), and 20.3 cm (Downstream)
- Minimum detectable density: 9×10^{15} m^{-3}

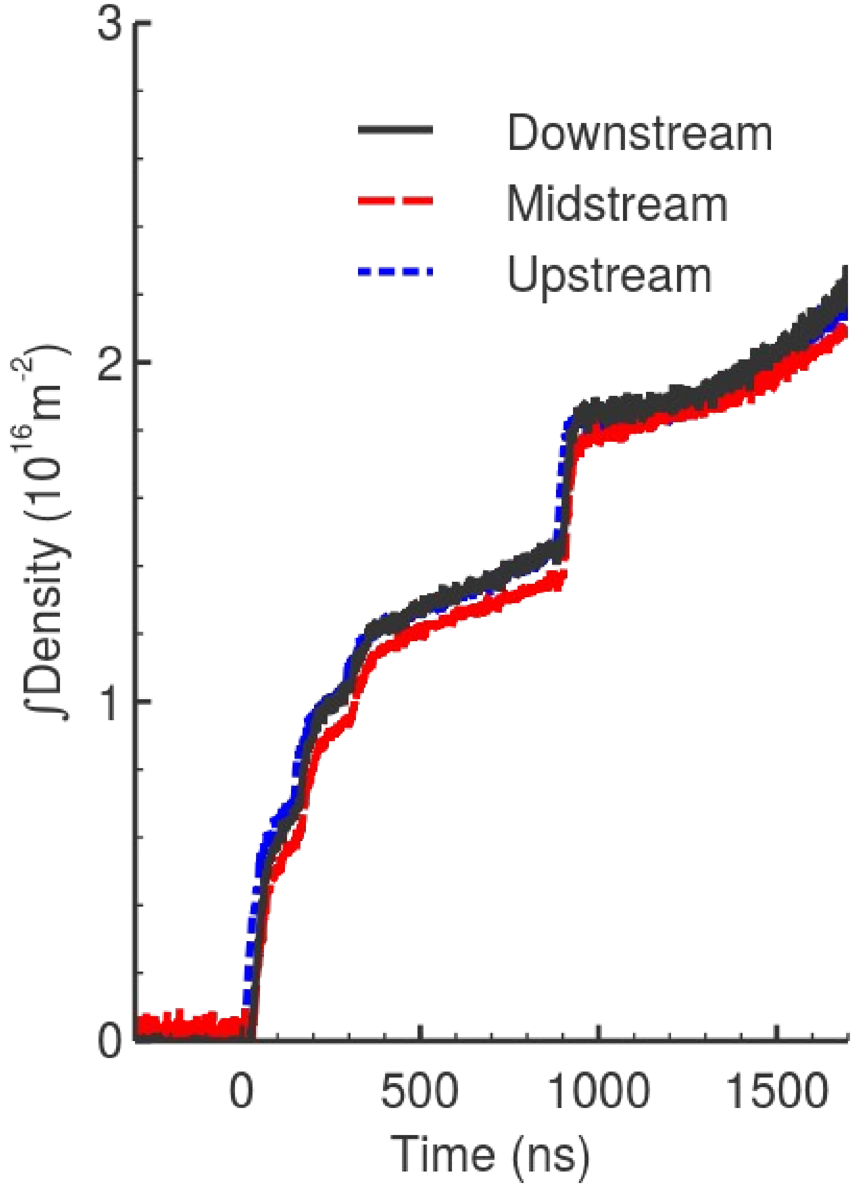
Laser-Absorption Spectroscopy Setup

- Observable metastable density before pulse
- Noise present in downstream measurements from pulse
- No significant variation with location



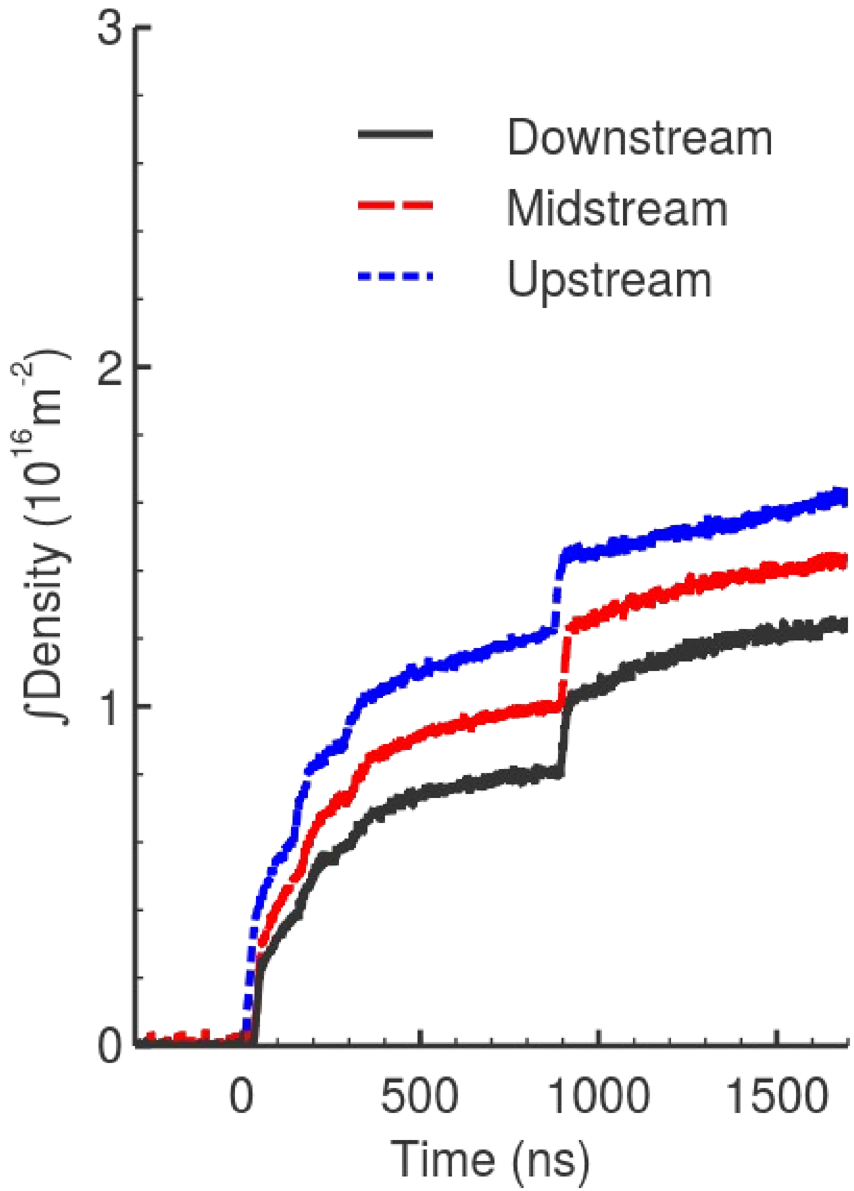
Metastable Densities (Low Pressure)

- Reduced pre-pulse density
- Increase in final density
- Reflections cause boost in metastable generation rate

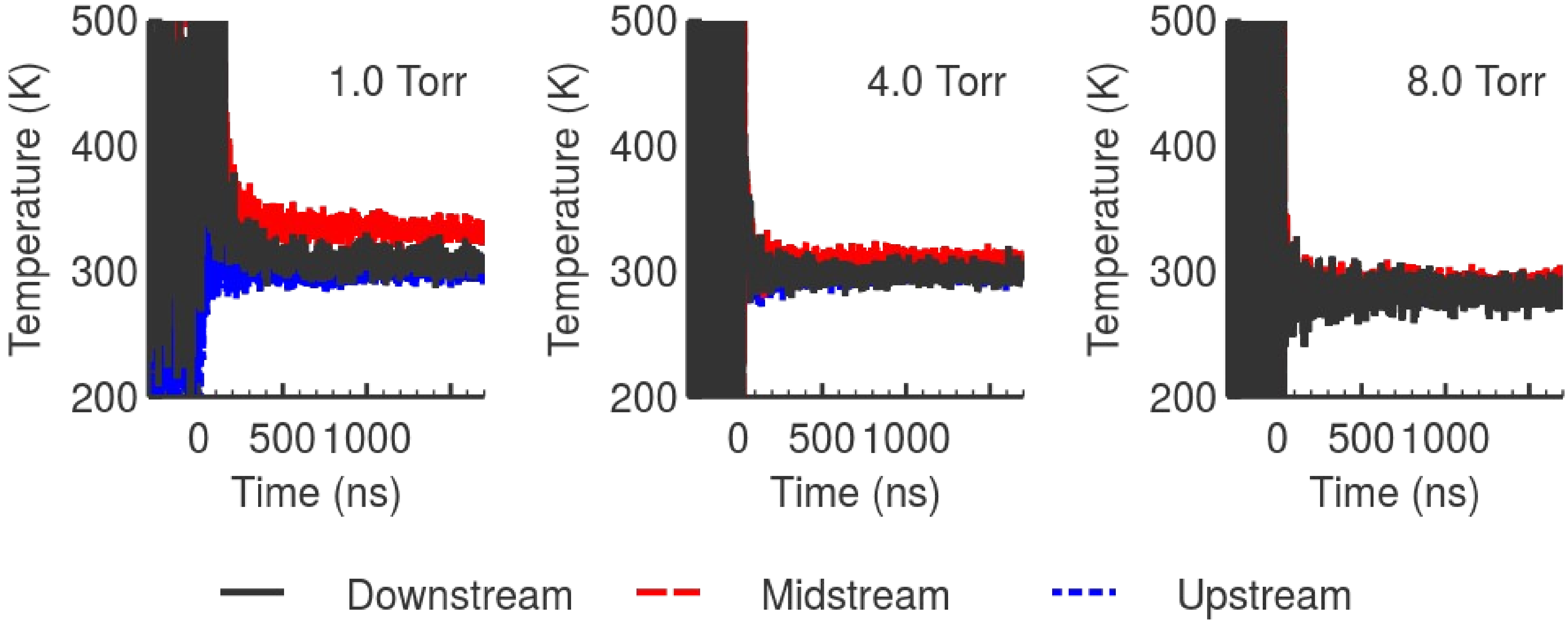


Metastable Densities (Moderate Pressure)

- No metastables before pulse
- Overall decline in metastable density
- Reflection effects still observable
- Plasma attenuation with distance from anode

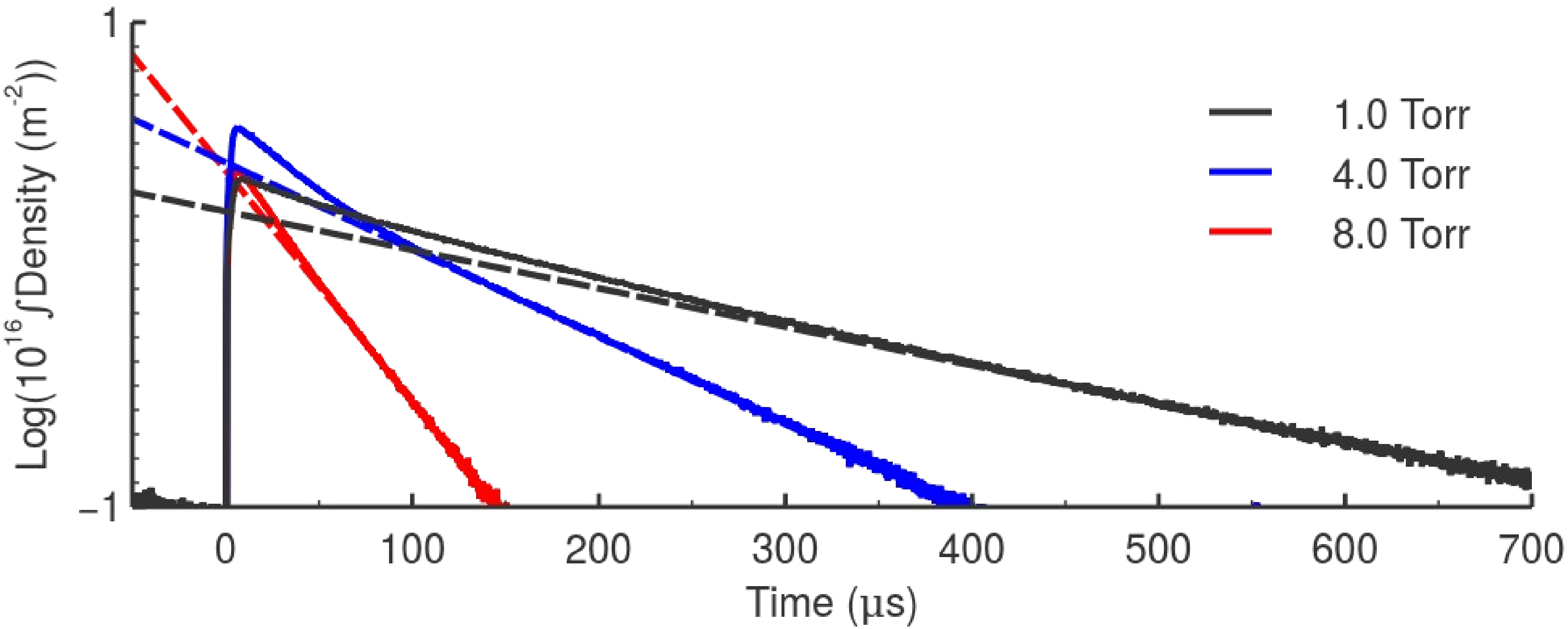


Metastable Densities (High Pressure)

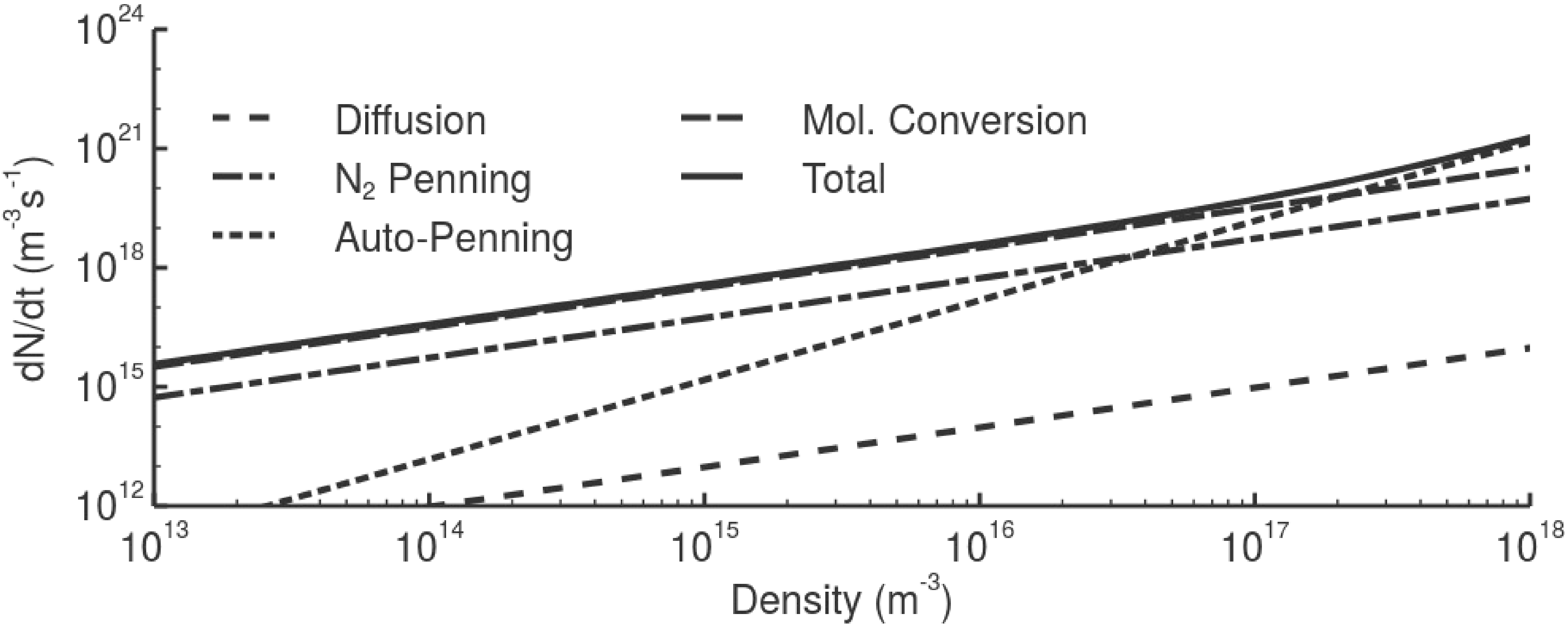


- Added benefit of laser-absorption spectroscopy
- Confirms minimal gas heating
- No significant difference from 300 K for all conditions

Temperatures



- Long-duration measurements reveal loss mechanisms
- Deviation from straight line → non-exponential process

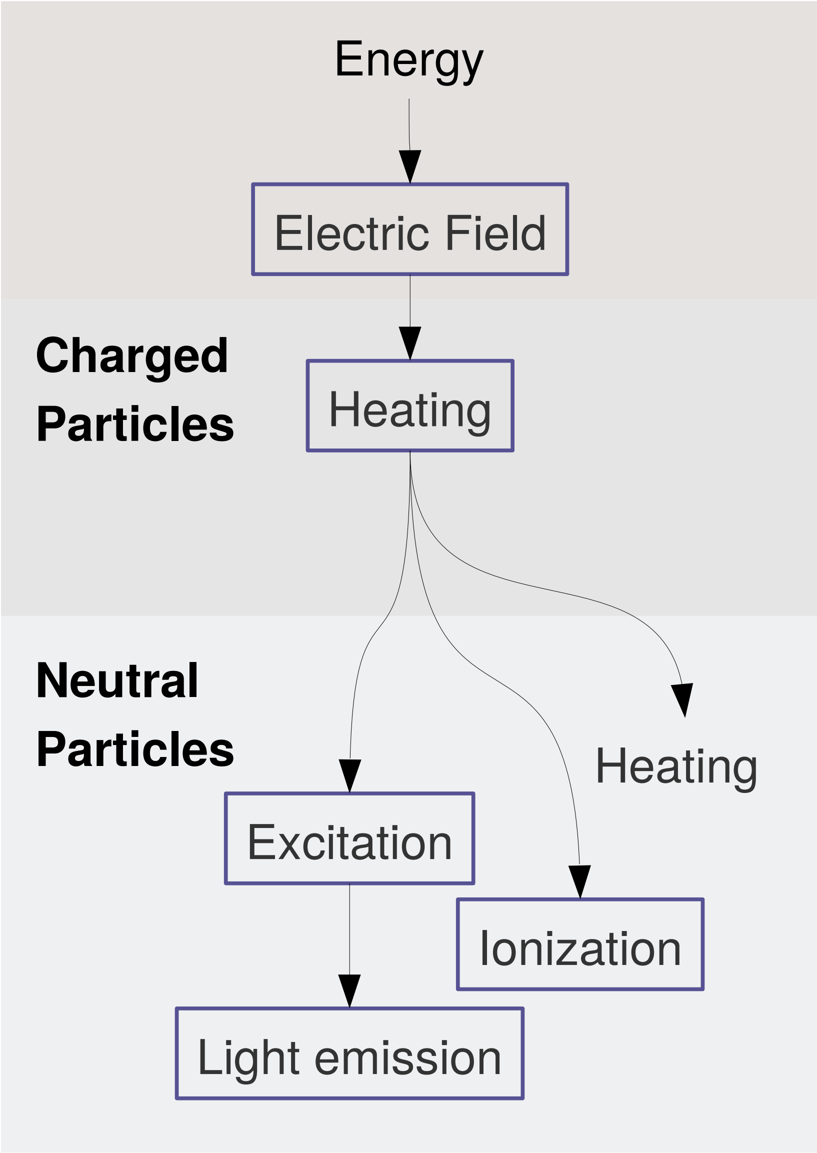


- Initially dominated by non-exponential processes
  - Superelastic electron collisions
  - Penning ionization between metastables
- Molecular conversion dominates at lower metastable densities
- Pre-pulse metastables only detectable for 4.0 Torr and below

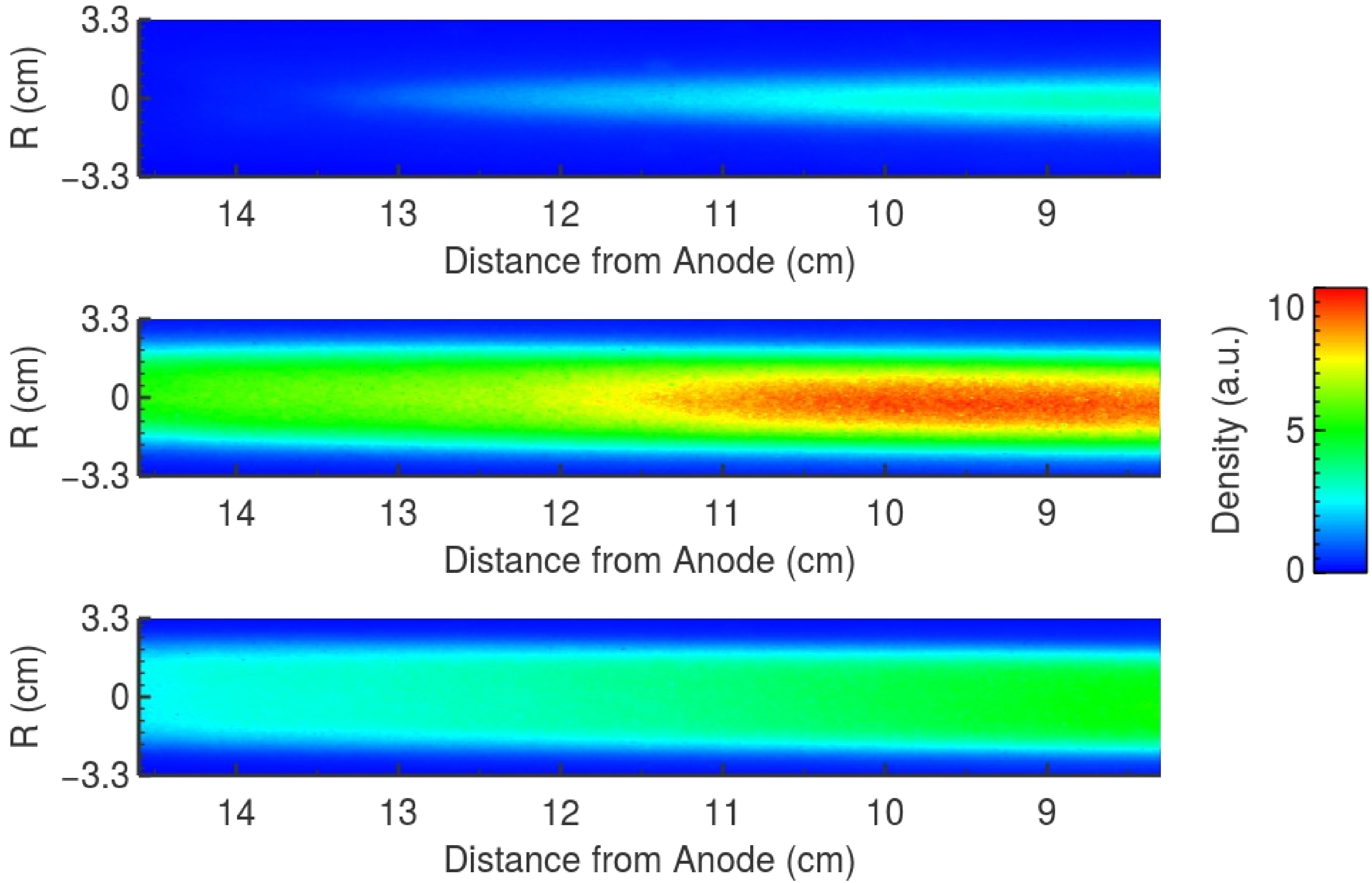
- Metastable states only persist between pulses at lower pressures
- There is an optimal pressure for metastable generation
- Significant attenuation of plasma occurs at higher pressures
- No detectable gas heating
- Losses initially dominated by superelastic electron collisions and Penning ionization between metastables

Global Model

- Metastables only one of many excited states
- Use metastable measurements to infer other properties
  - Electric fields
  - Electron densities and temperatures
  - Excited states



- Assumed spatial dependence (global model)
- Species:
  - Helium, all states $n < 5$
  - Helium ions
  - Electrons
- Reactions:
  - Elastic scattering (Pack)
  - Electron (de-)excitation (380 transitions, Ralchenko 2008)
  - Optical transitions (126 transitions, NIST ASD)
  - Excitation transfer (35 transitions, Dubreuil and Catherinot)



Weatherford, B.R., Barnat, E.V., Xiong, Z., Kushner, M., 65th Gaseous Elec. Conf., October 22-26, 2012, Austin, TX.

Spatial Dependence

$$\frac{dN_i}{dt} = n_e \left[\sum_{j \neq i} N_j K^e_{j,i}(T_e) - N_i \sum_{j \neq i} K^e_{i,j}(T_e) \right]$$

Electron (de)excitation

$$+ \left[\sum_{j > i} N_j K^o_{j,i} - N_i \sum_{j < i} K^o_{i,j} \right]$$

Radiative transitions

$$+ N_g \left[\sum_{j \neq i} N_j K^a_{j,i} - N_i \sum_{j \neq i} K^a_{i,j} \right]$$

Atomic excitation transfer

Particle Density Equation

$$\frac{d}{dt}\left(\frac{3}{2}n_eT_e\right) = \frac{e^2 n_e E(t)^2}{m_e k_m(T_e)N_g}$$

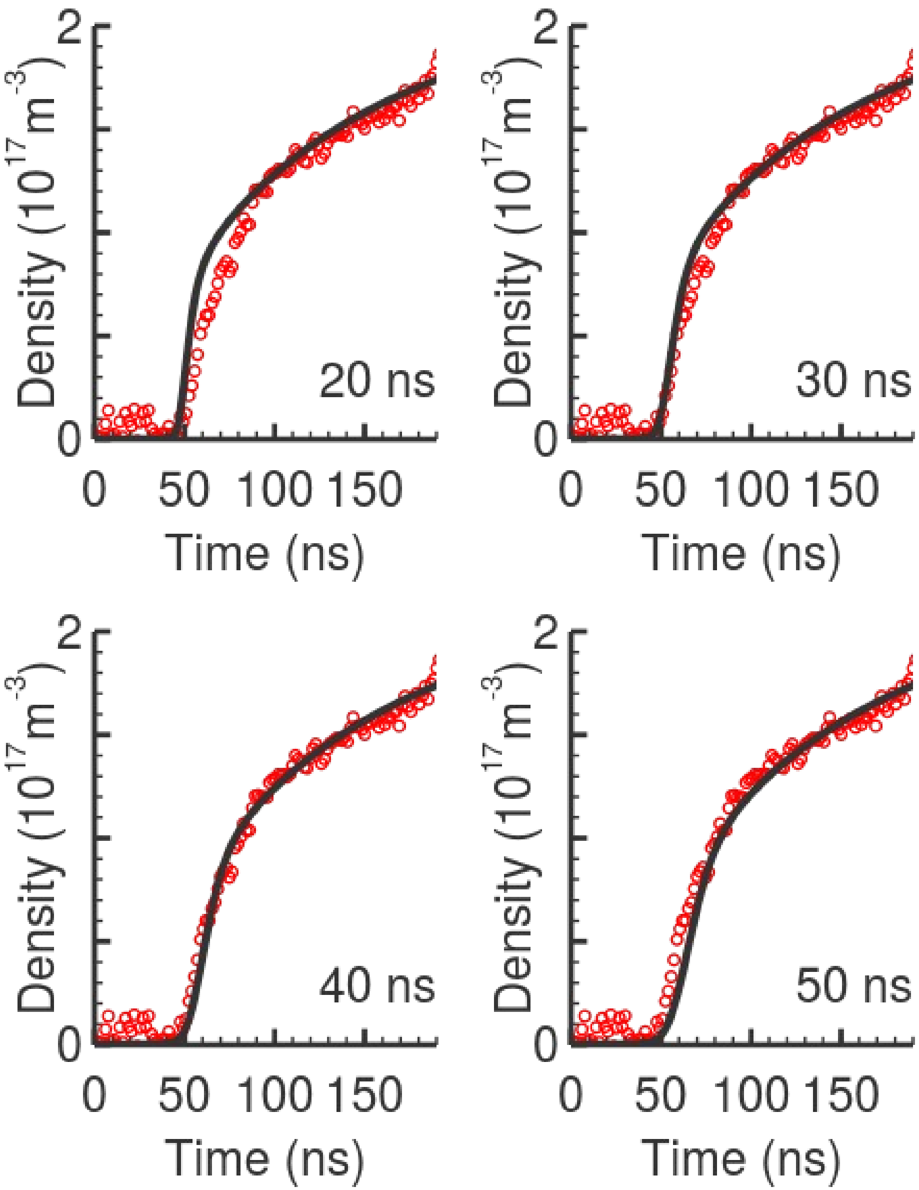
Electric field heating

$$\cdot - n_e k_m(T_e) N_g \left(\frac{3m_e}{M}\right)\frac{3}{2}(T_e - T_g)$$

Elastic collision losses

$$-n_e \sum_i \sum_{j\neq i} K_{ij}^e N_i \Delta\epsilon_{ij}$$

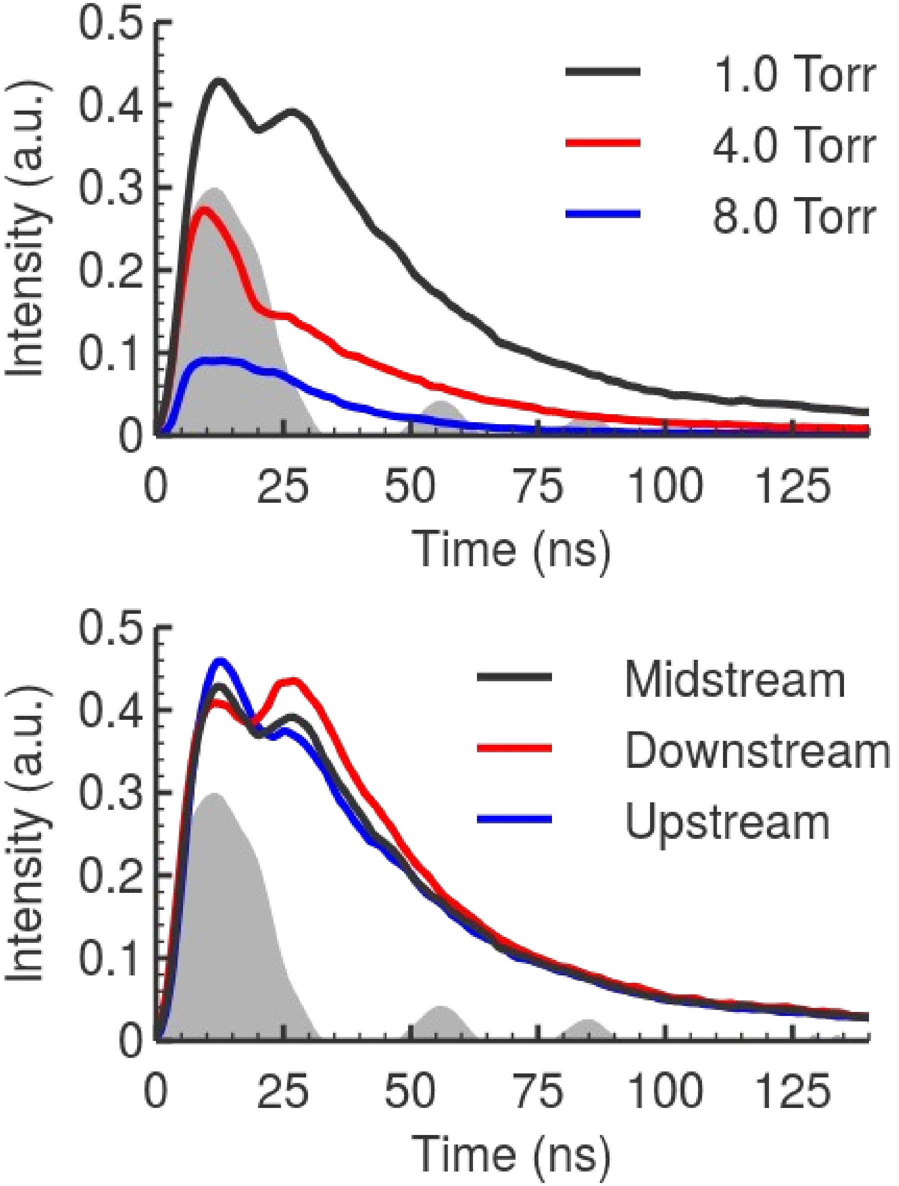
Inelastic collisions losses

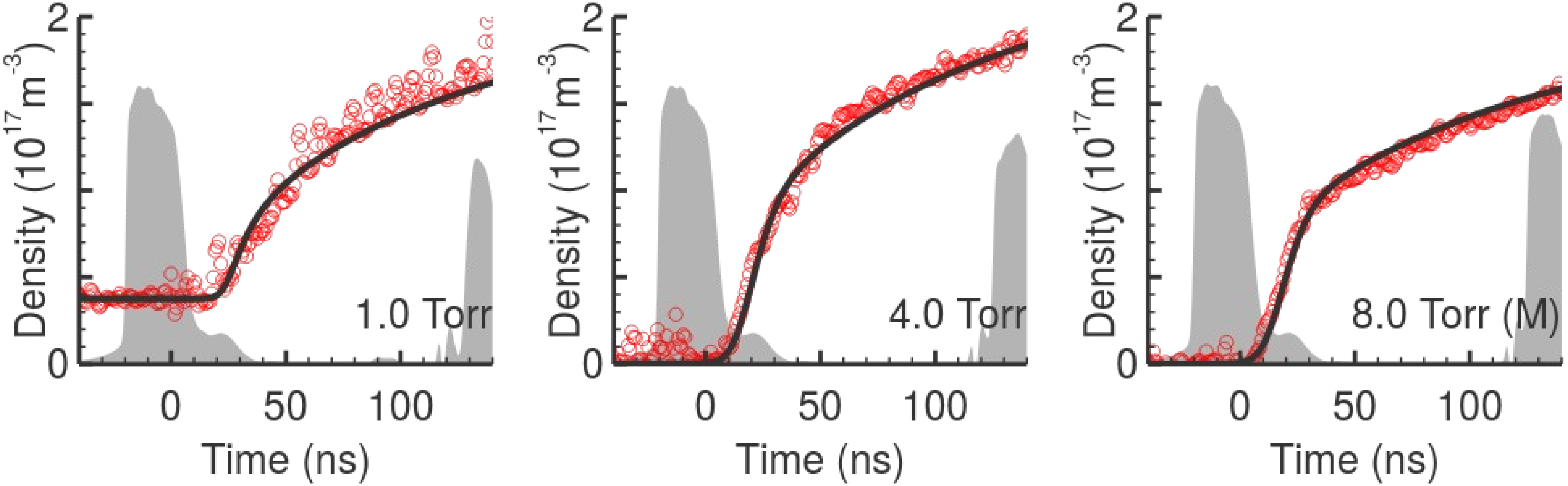


- Assumed electric field applied as a Gaussian pulse
- Free variables: electric field and pulse width
- Best fit obtained with 40 ns pulse-width, but this is longer than applied pulse?

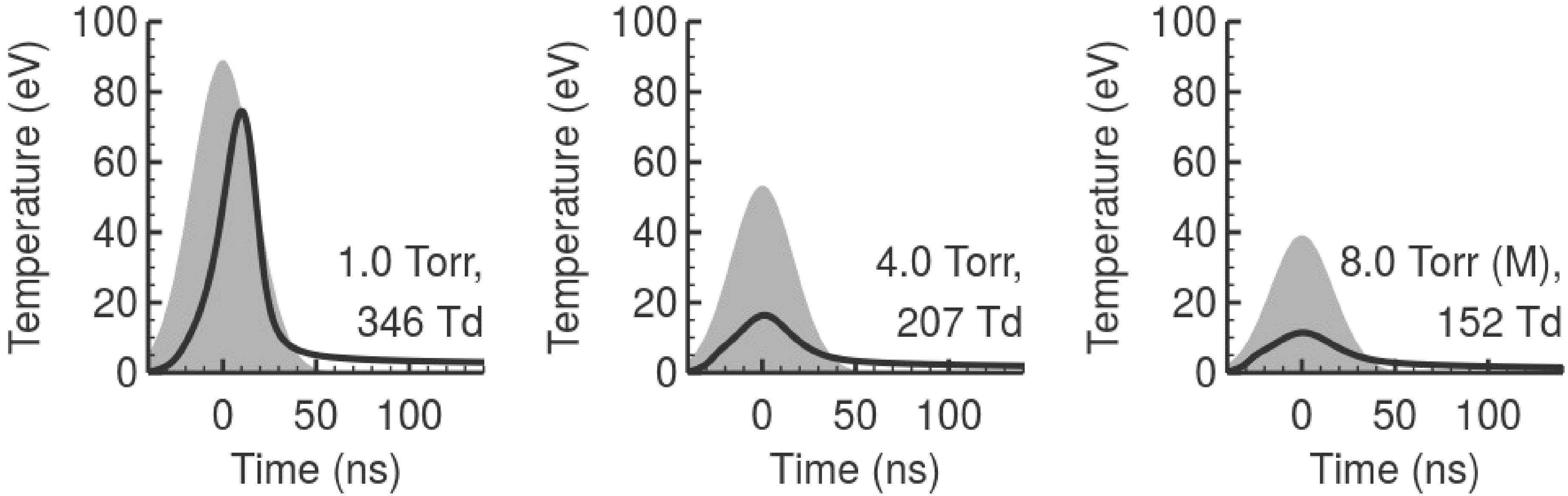
Pulse-width

- Optical emissions showed evidence of return stroke
- Double peak at several pressures
- Second peak most intense closest to ground electrode
- Excitation period effectively longer than 25 ns





- Good agreement for most cases—trends and magnitudes were consistent
- Largest discrepancies at 1.0 Torr
  - Initial rise too fast
  - Radiative cascade too slow
  - Extended excitation?



- Peak fields consistent with other studies
- 1.0 Torr fields well into range where two-term expansion solutions performed better
- 1.0 Torr temperatures are not reasonable

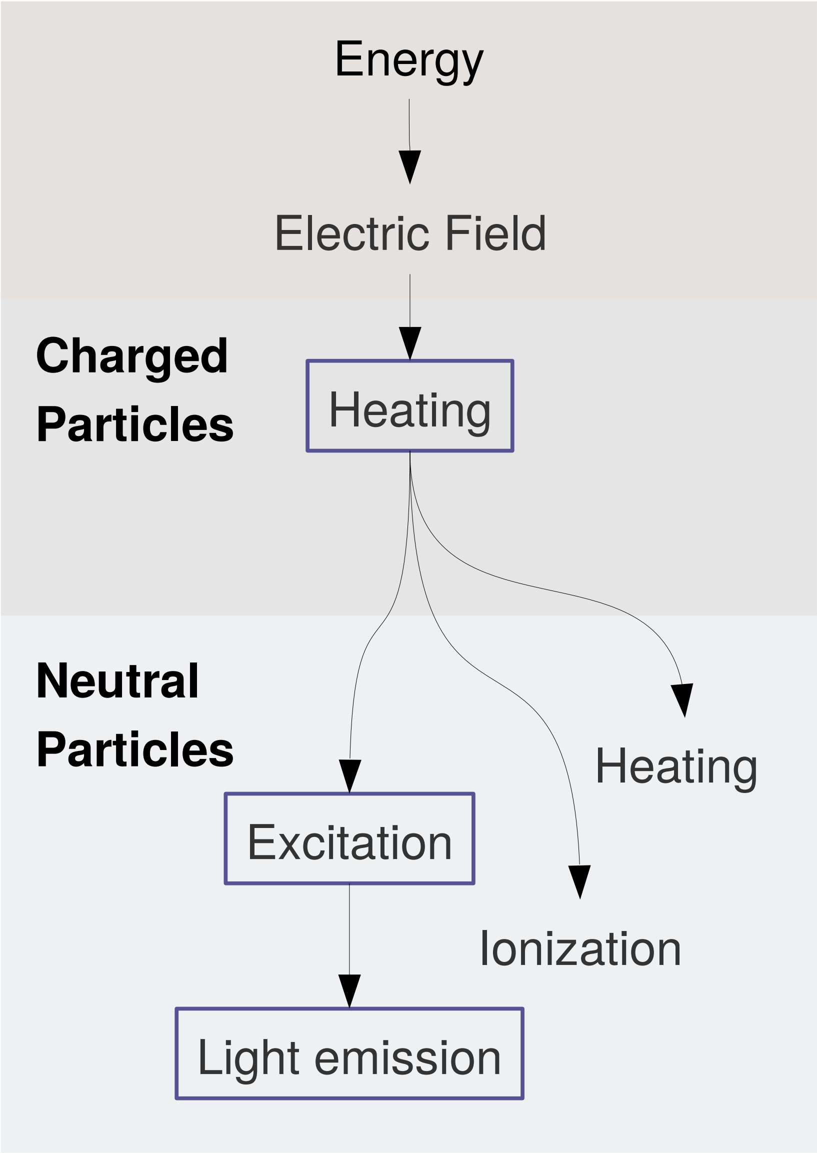
Peak Fields and Electron Temperatures

- Created and applied global model of RPND
- Observed evidence of extended excitation period
- Simulations able to reproduce metastable density dynamics
- Temperatures and fields at 1.0 Torr suggest potential violation of underlying assumptions

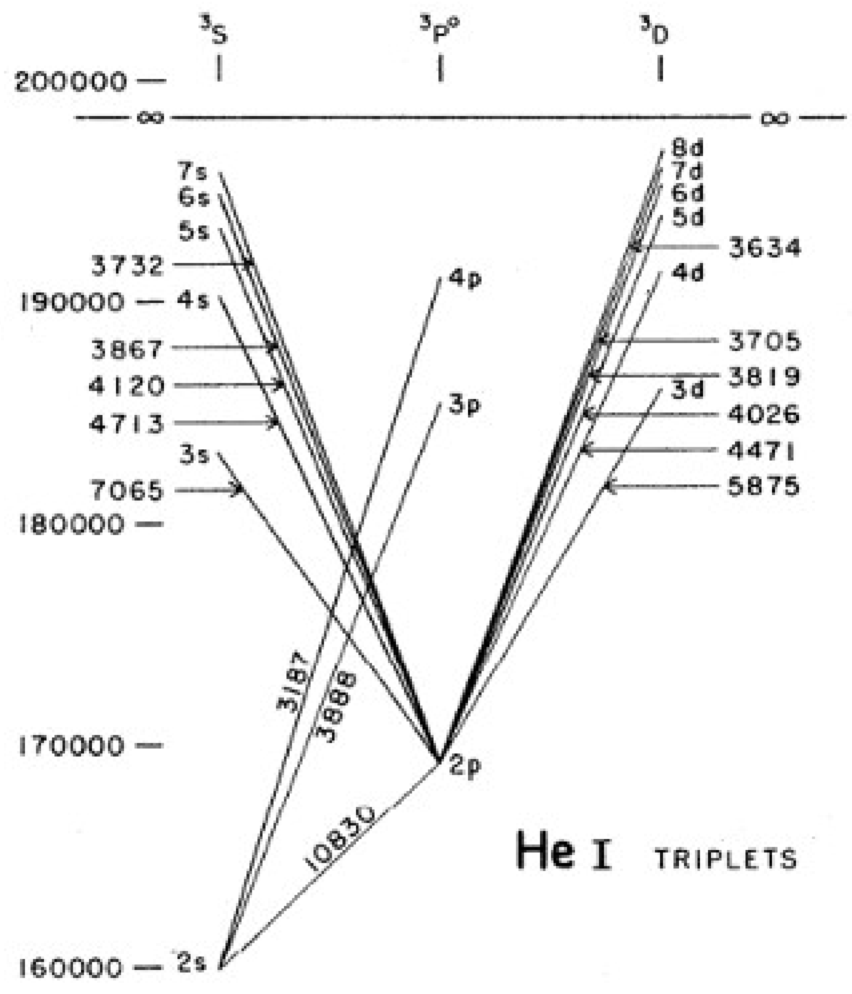
Summary of Simulation Results

Emission Measurements

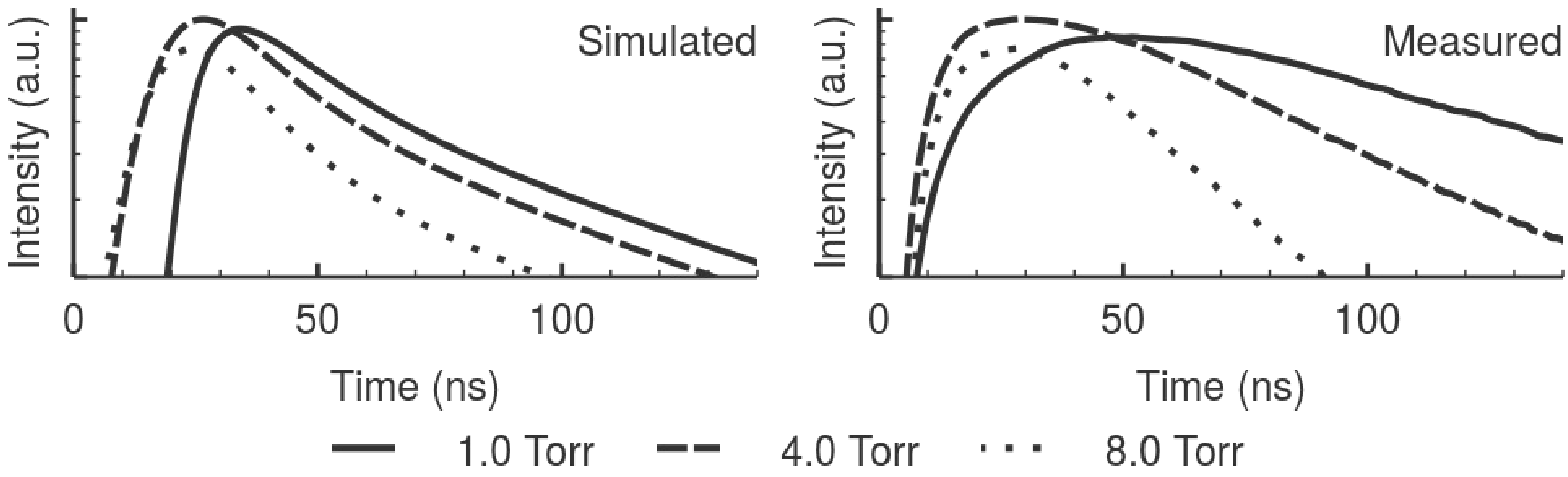
- Simulations based on metastable densities
- Optical emissions spectroscopy can check other excited states
- Can reveal other phenomena
- May be able to estimate electron temperature



Corroborating Simulations



- Observed transitions in visible (triplet and singlet)
- Calibrated relative intensities with tungsten lamp
- Same conditions as absorption spectroscopy



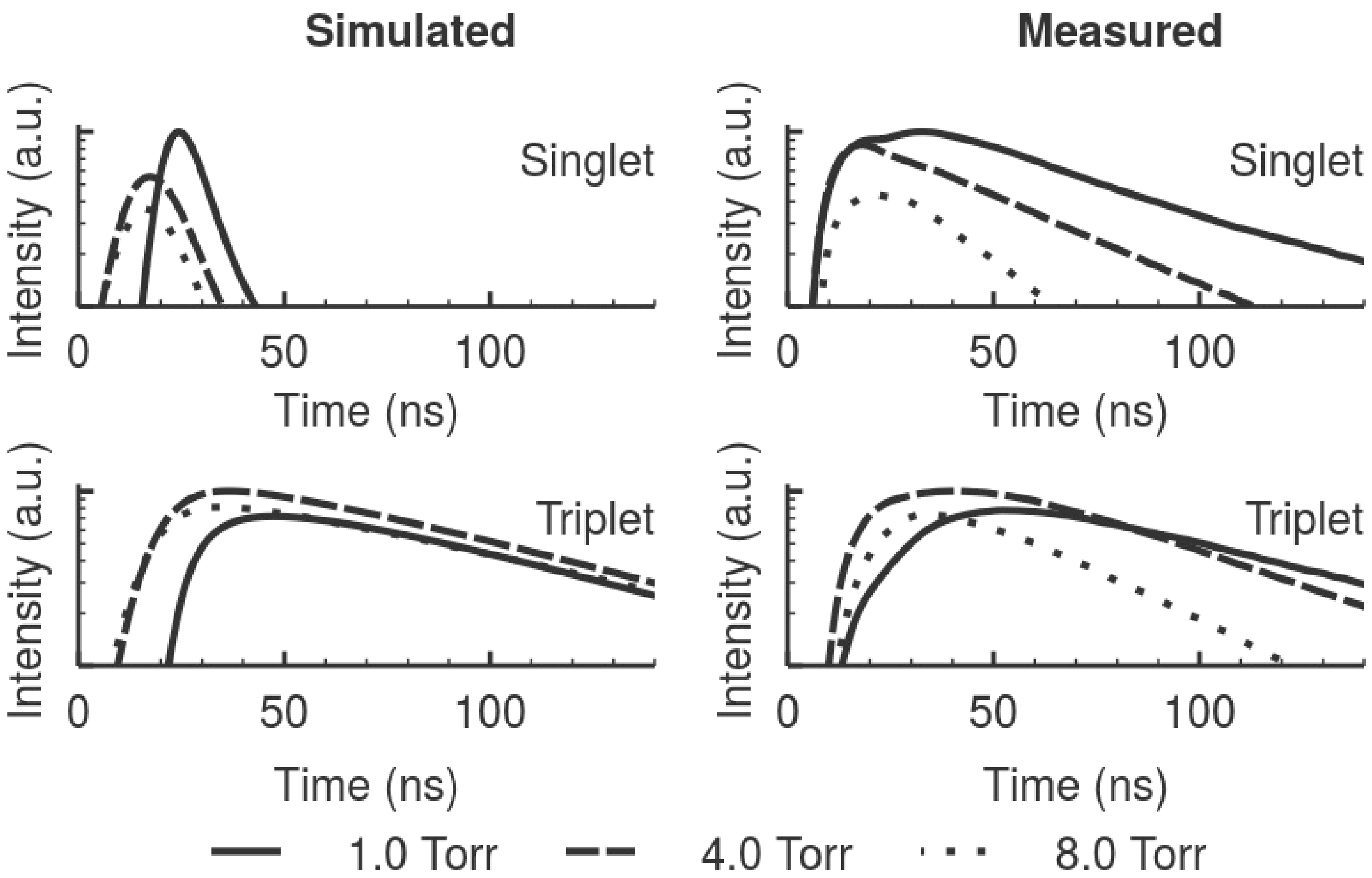
- Relative peak heights and timing similar, distinctly different in shape
- Measurements suggest pressure-dependent de-excitation process (Penning ionization of impurities?)
- Long emission lifetime suggest an extended excitation process
  - Beam-like electrons
  - Persistent electric field

Extended Excitation ($3^1D \rightarrow 2^1P^o$, 668 nm)

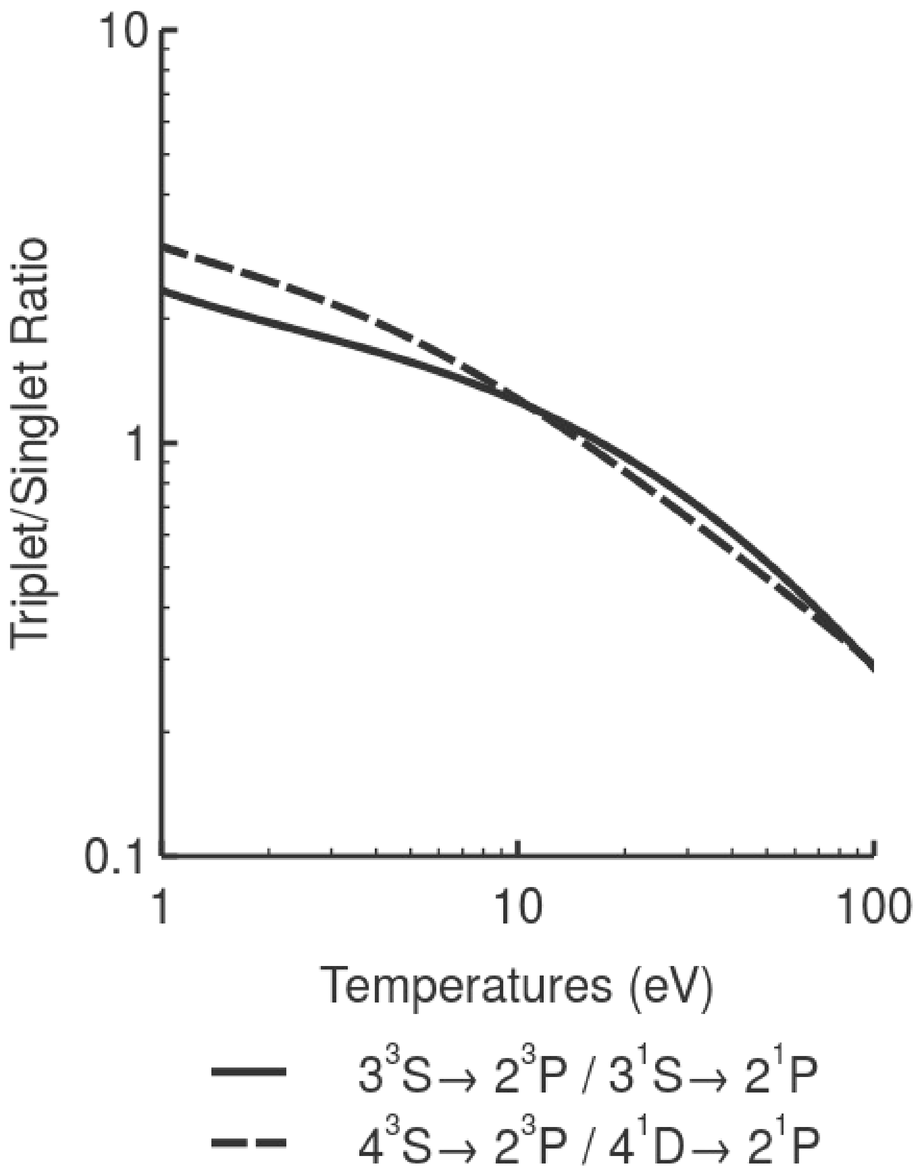
- Resonance radiation can be “trapped” in plasmas
- Increases energy residence time
- Could contribute to extended excitation

Pressure (Torr)	Trapping Factor	Effective Lifetime (s)
1.0	8,773	1.549×10^{-5}
4.0	38,031	6.715×10^{-5}
8.0	78,837	1.392×10^{-4}

Lifetime increase for $3^1P \rightarrow 1^1S$ transition

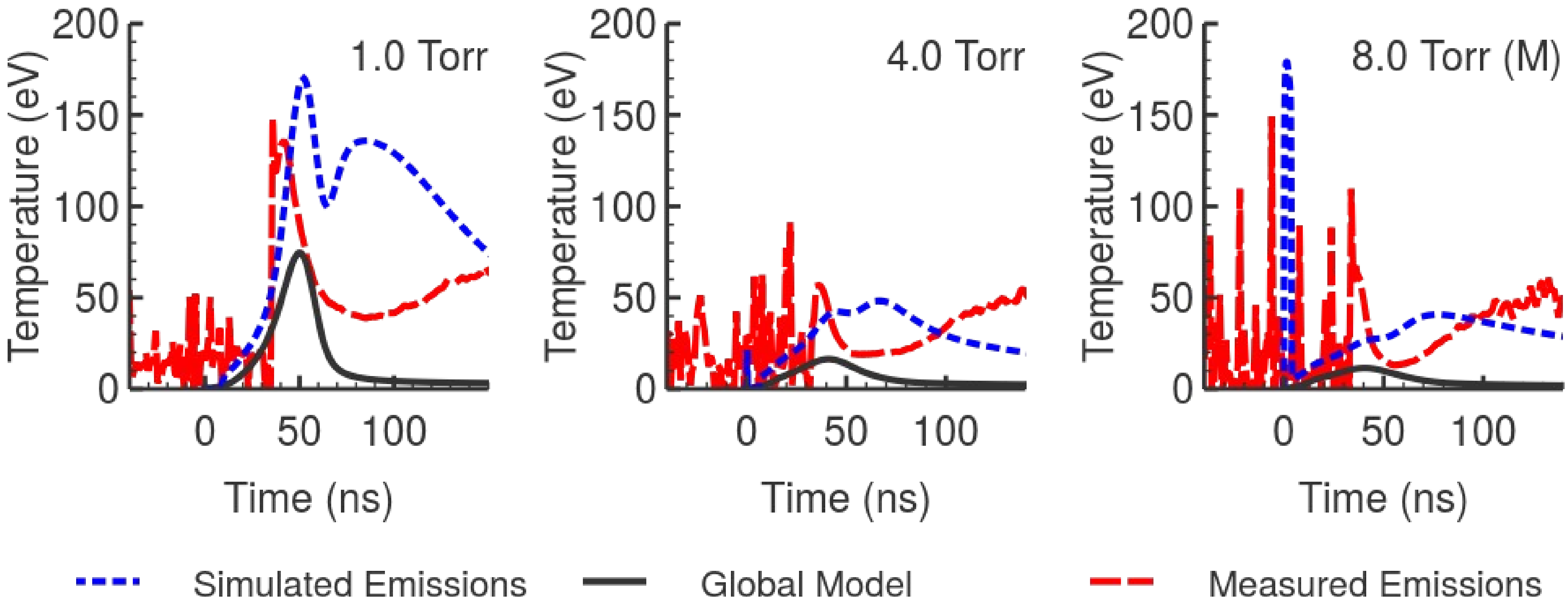


- Verify by comparison of 389 nm and 501 nm
- If radiation trapping affects system, measured lifetime of 501 line should be greater than simulation



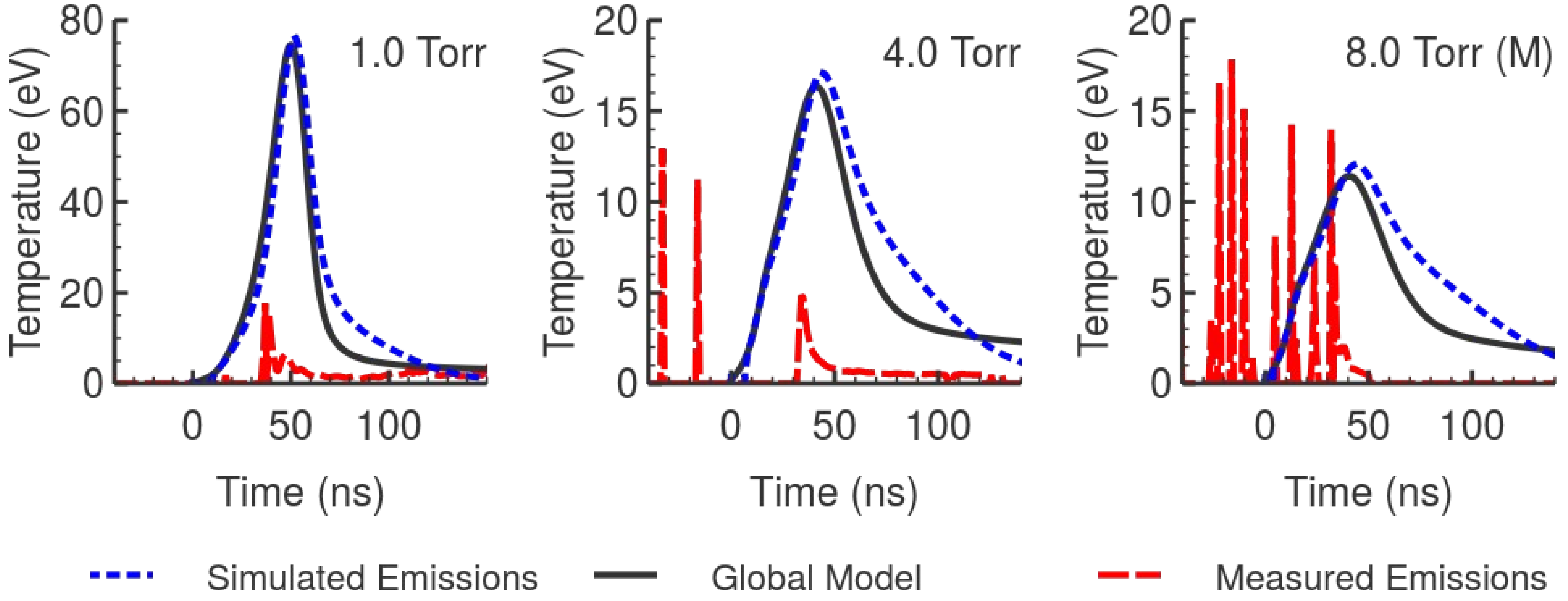
- Can be used to estimate electron temperature
- Assumes
  - Electron-ground excitation only
  - Radiative losses
- Maxwell-Boltzmann distribution

$$\frac{I_{i,j}}{I_{i',j'}} = \frac{\lambda_{i',j'} A_{i,j} \sum A_{i'} K_{0,i}(T_e)}{\lambda_{i,j} A_{i',j'} \sum A_i K_{0,i'}(T_e)}$$



- Poor match with simulated and measured emissions
- Possible need to include n = 5 states
- Collisional redistribution of n = 4 states potentially a factor

Ratio 1: $4^3S \rightarrow 2^3P$ / $4^1D \rightarrow 2^1P$



- Coronal model and global model: consistent
- Application limited by signal-to-noise ratio
- Likely not a Maxwellian temperature
- May still be useful as a measure of mean electron energy

Ratio 2: $4^3S \rightarrow 2^3P$ / $3^1S \rightarrow 2^1P$

- Global model captures some features of RPND population kinetics
- Missing physics
  - Extended excitation (distribution or field related?)
  - Radiation trapping
- $4^3S \rightarrow 2^3P^o$ / $3^1S \rightarrow 2^1P^o$ ratio may be useful indicator of electron energy in helium RPDN

Conclusions

- Developed laser-absorption spectroscopy system and analysis software
- Obtained detailed measurements of metastable dynamics in a helium RPND
- Created a global model code for helium plasmas
- Used code to infer plasma parameters from metastable density information
- Measured optical emissions of the RPND
- Found several additional phenomena that are potentially important in the description of the RPND

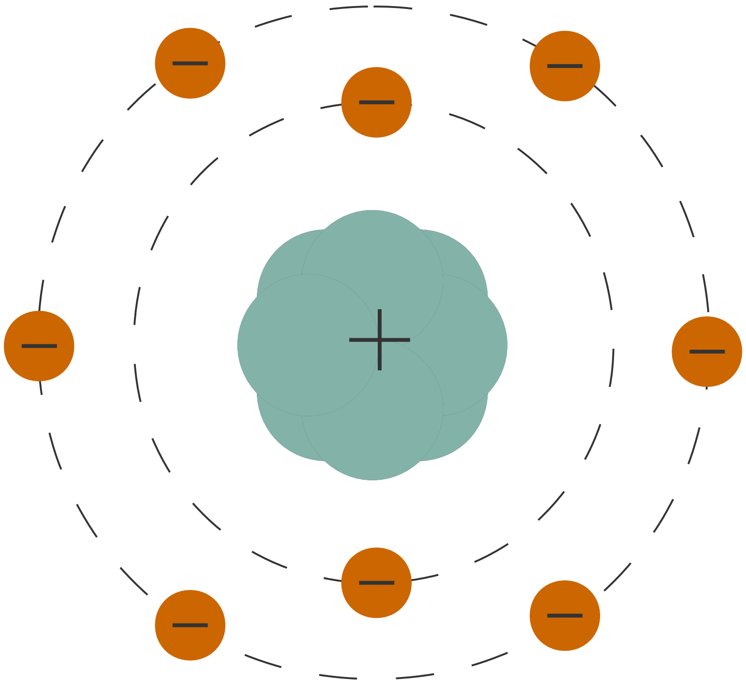
- Excited states only persist at lower pressures, what ensures consistent breakdown?
- Unexpectedly long excitation period, even accounting for return stroke
  - Slowing of beam-like electrons
  - Radiation trapping
  - Persistent electric fields
- May be possible to employ simple line ratio diagnostics for estimates of mean electron energy

- Address temporal evolution of metastable density profile (Abel inversion) $\rightarrow$ wall effects
- Incorporate additional physics (Penning ionization of impurities, radiation trapping, etc.) in present global model.
- Employ different EEDF models to better match excited state dynamics
- Move to a 0D Monte-Carlo simulation of RPND to avoid EEDF issues
-

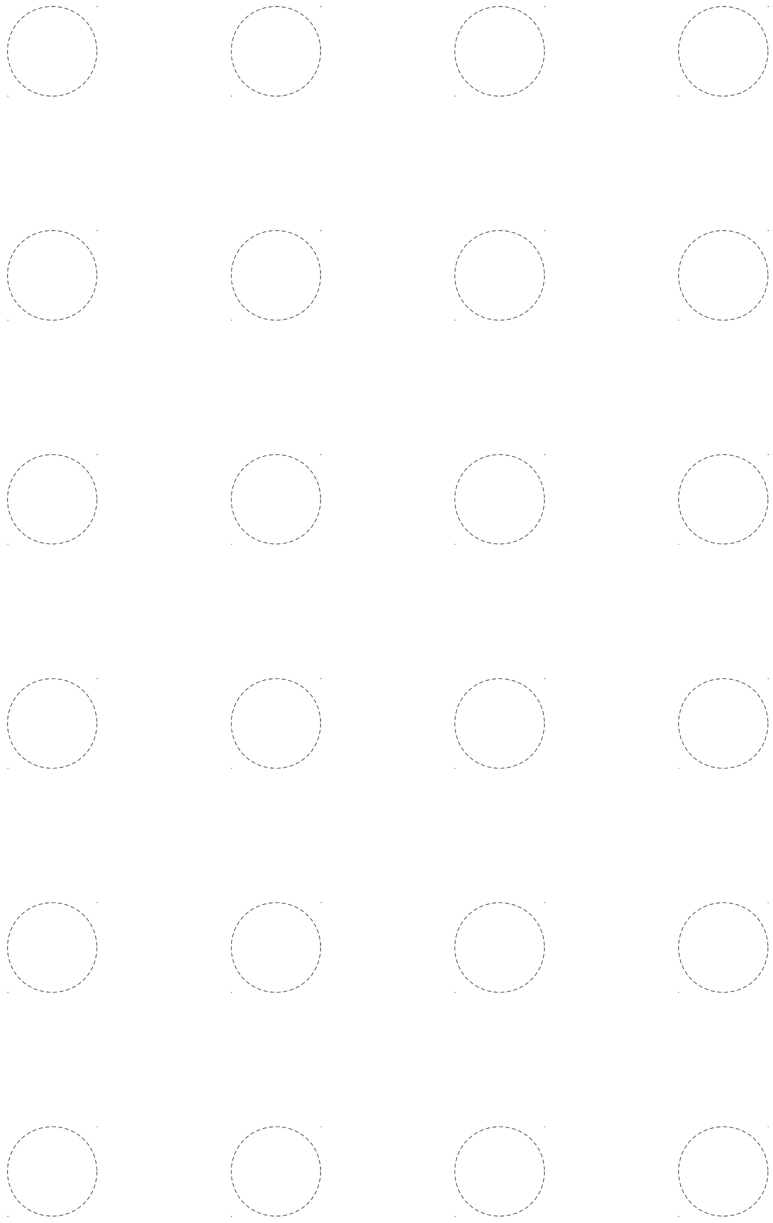
Acknowledgements

Questions

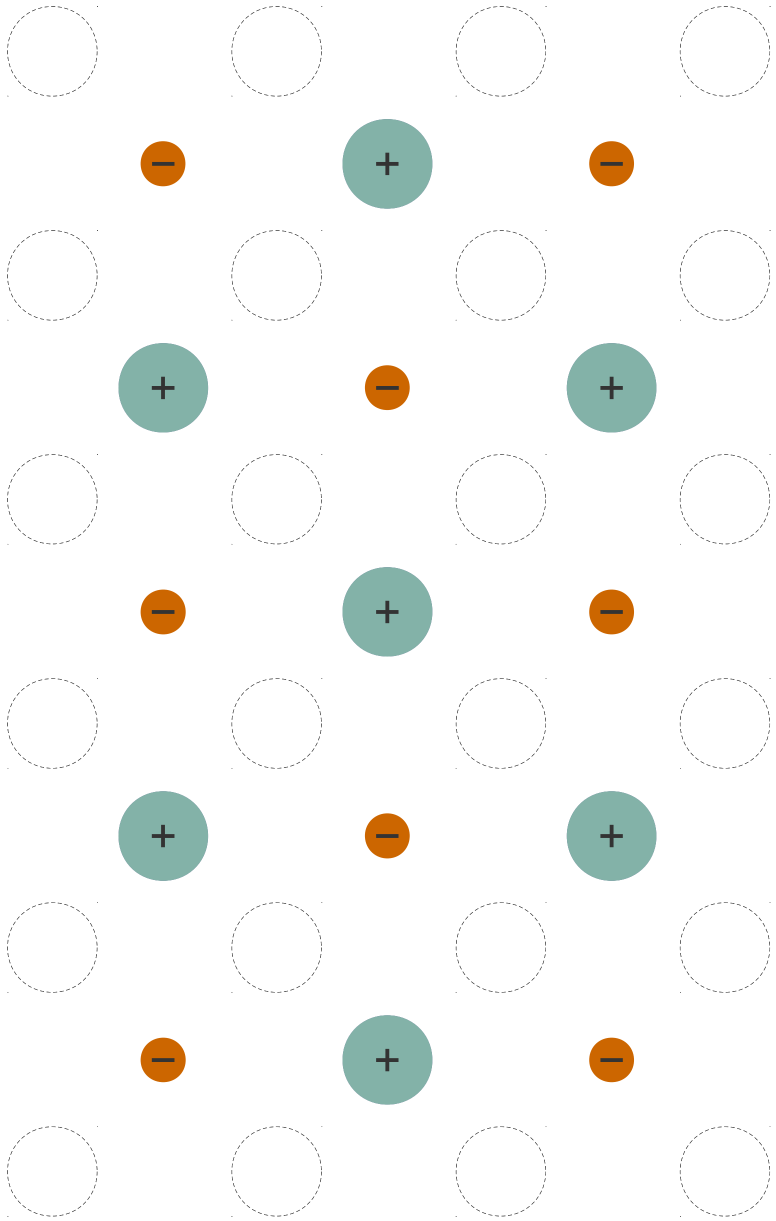
Backup Slides



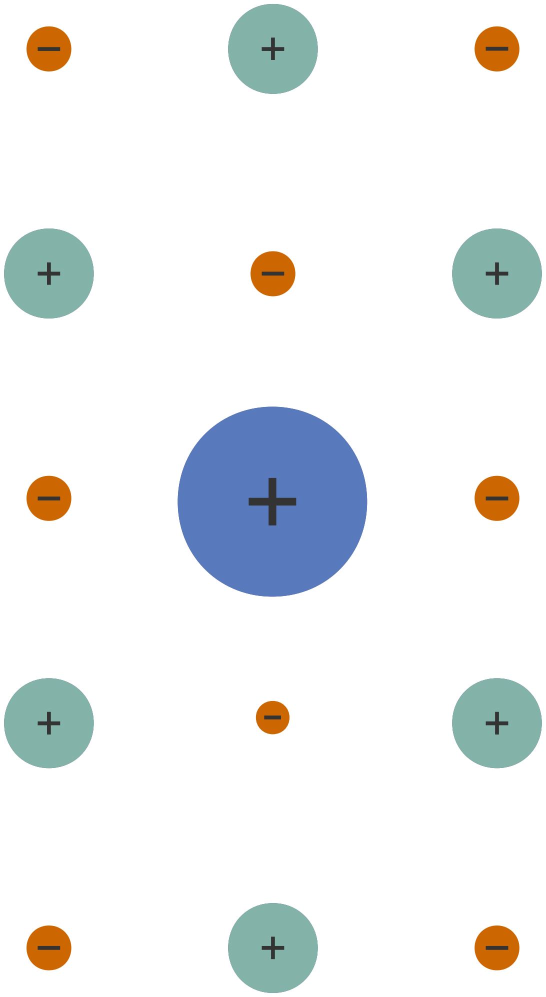
- Positively-charged nucleus
- Orbited by negatively-charged electrons
- Overall, electrically neutral



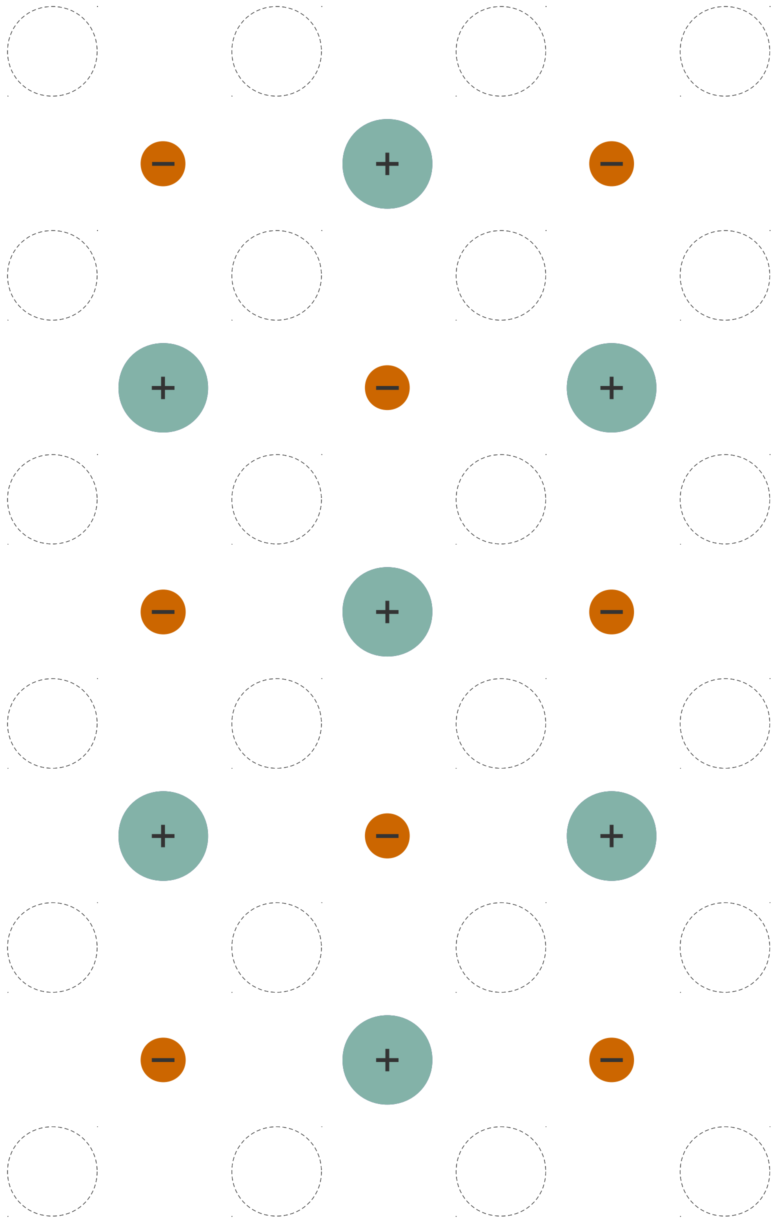
- Consider an atomic gas
- Many neutral atoms with random motion
- No electrical interaction



- Add energy to gas
  - Electricity
  - Shock
  - Light
- Electrons separate from atoms
- Collection of neutral atoms, (positive) ions, and (negative) electrons
- What makes a plasma?



Consider what happens with an electrical perturbation.



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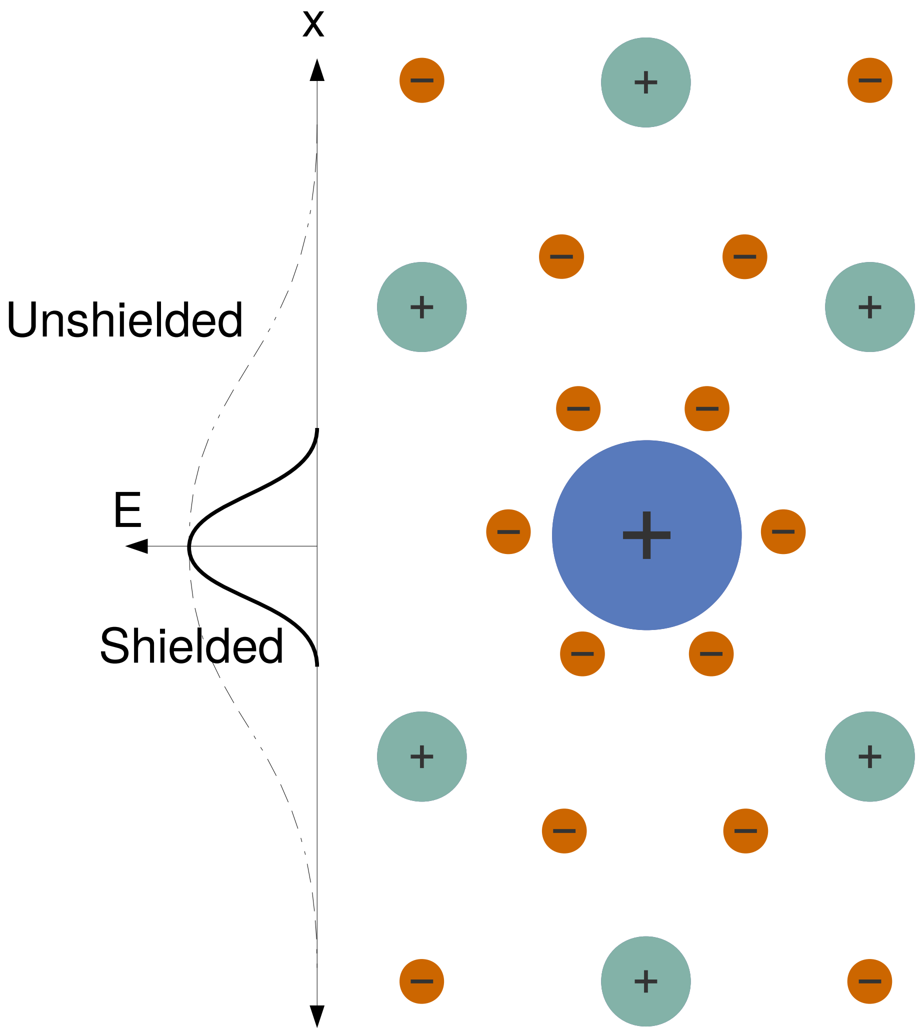
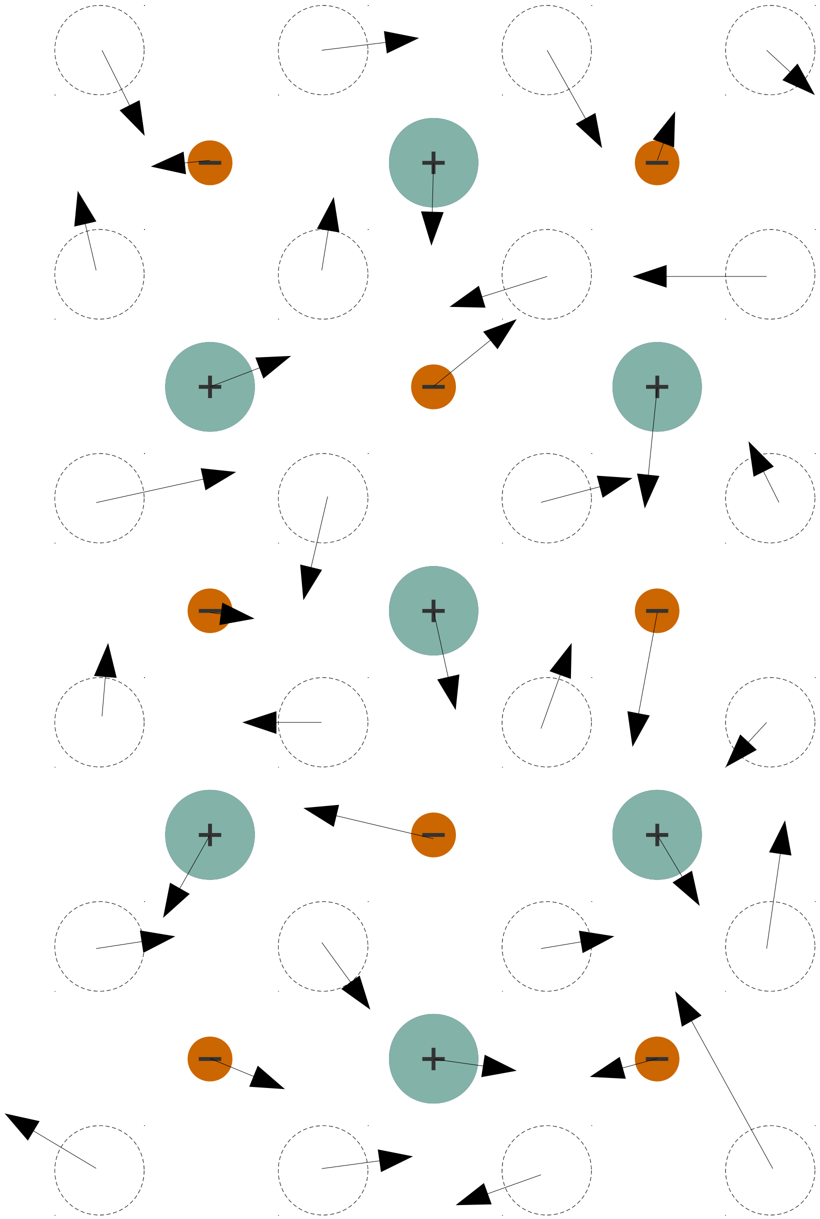
Ionized Gas

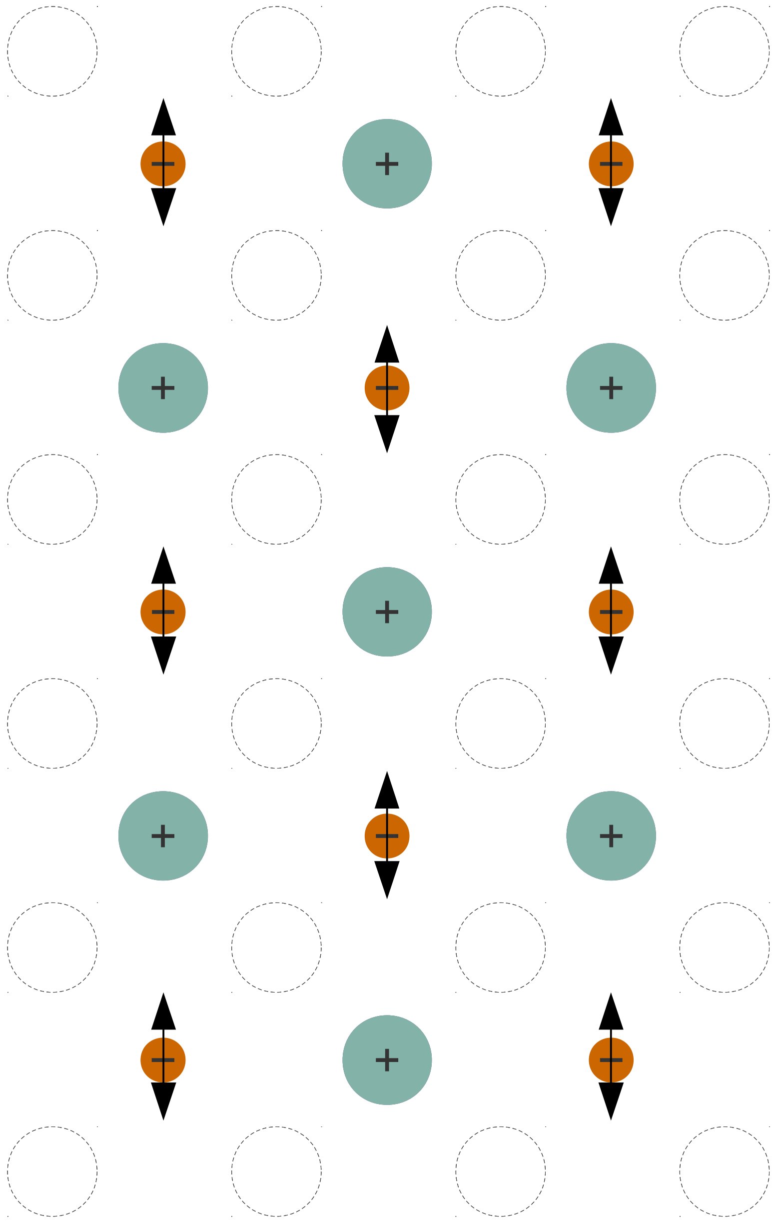


- Electrons move to shield charge
- Similar for positive and negative perturbation
- Electric field falls off quickly
- $\lambda < L$, $n\lambda^3 >> 1$



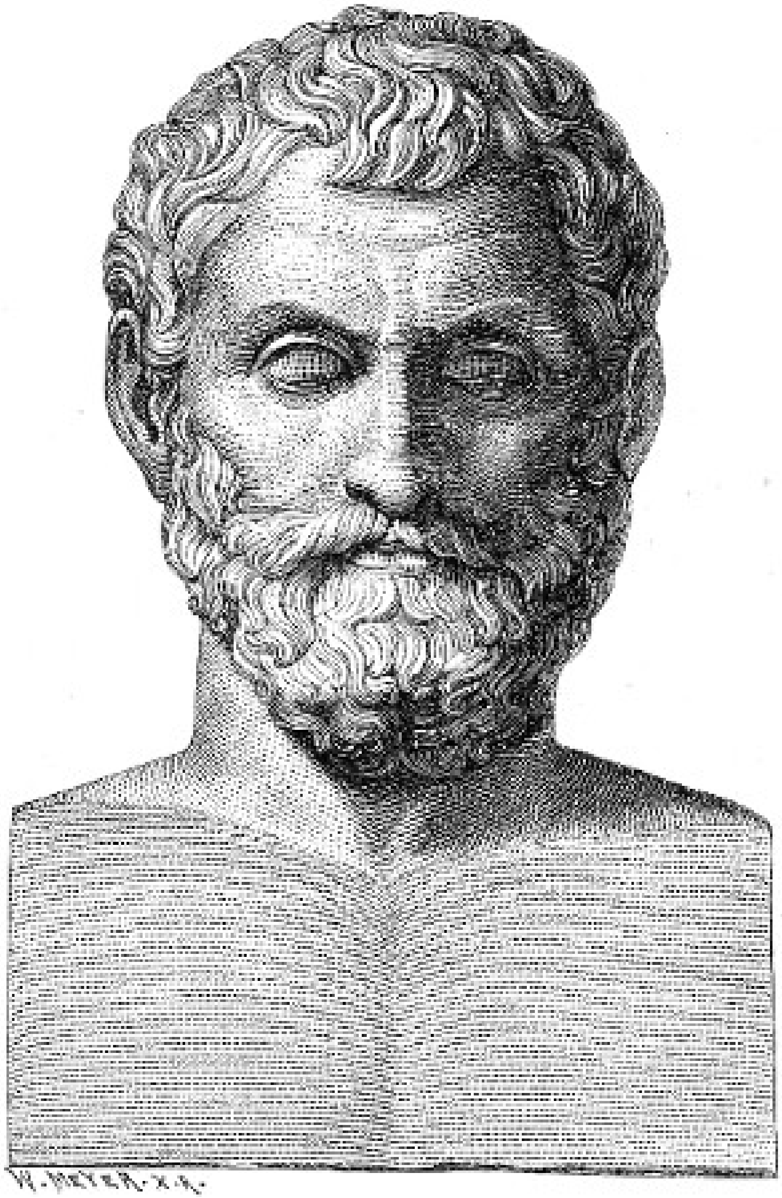
- Random collisions with neutral particles
- Neutral collisions can still determine properties of ionized gas
- Electric interactions should be dominant in plasma

Neutral Particle Interactions

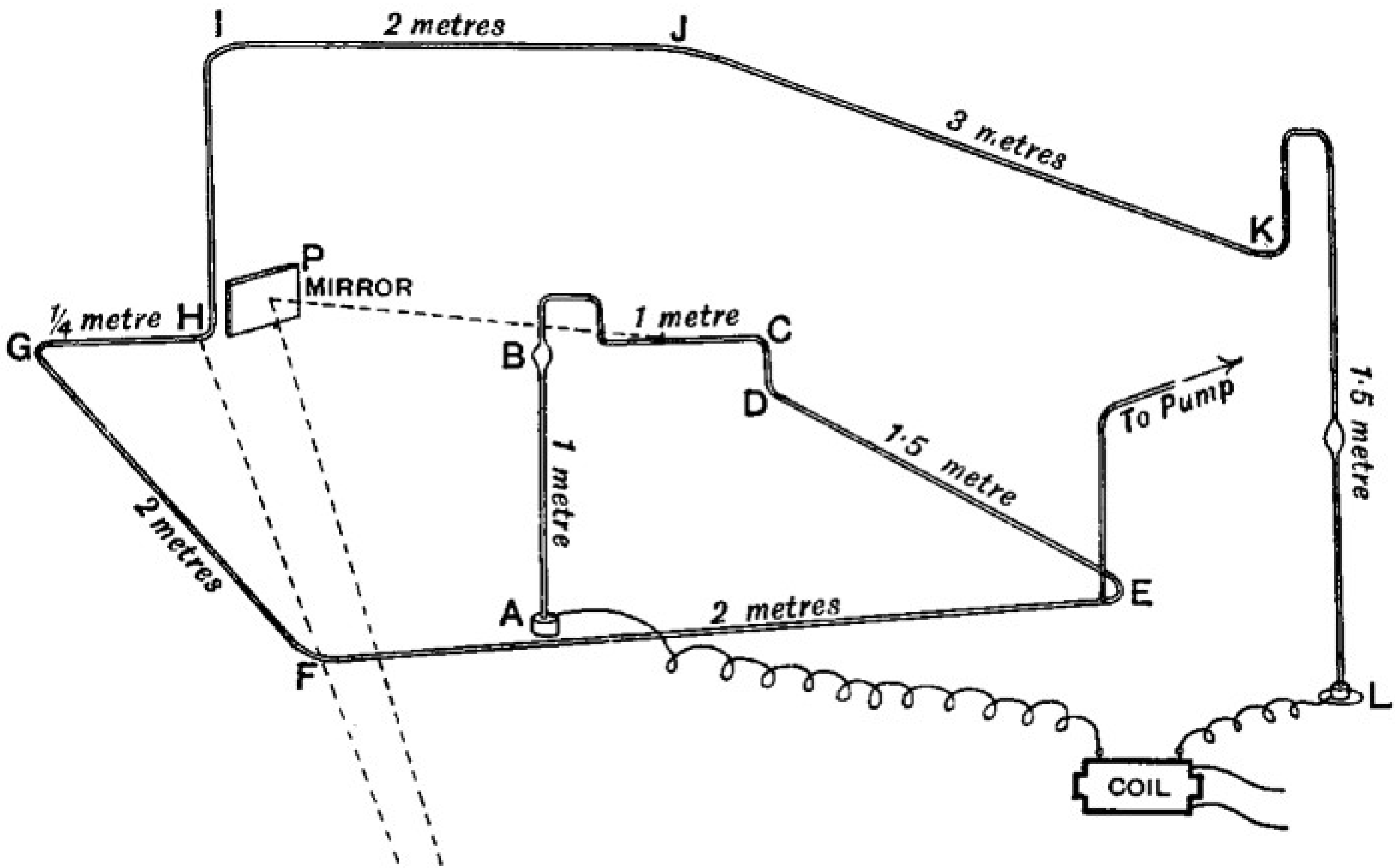


- Electrons possess natural oscillation frequency
- Characteristic of electrical interaction
- $\omega_p > \nu$

Charged-Particle Oscillations

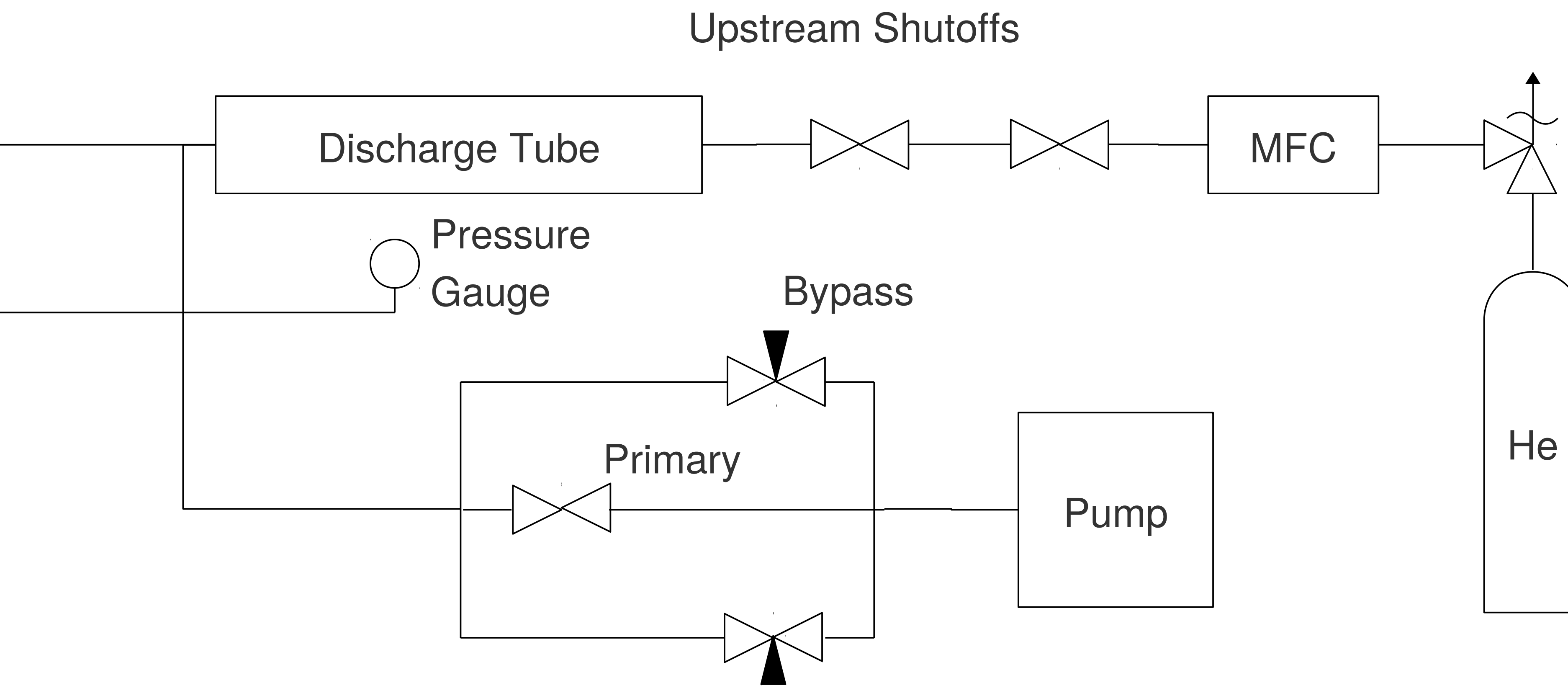


Wallis, E. et al. "Illustrerad verldshistoria utgifven av E. Wallis. Volume I." (1875).
Stotesbury, H. "J. J. Thomson." Popular Science, Vol. 56 (1899).

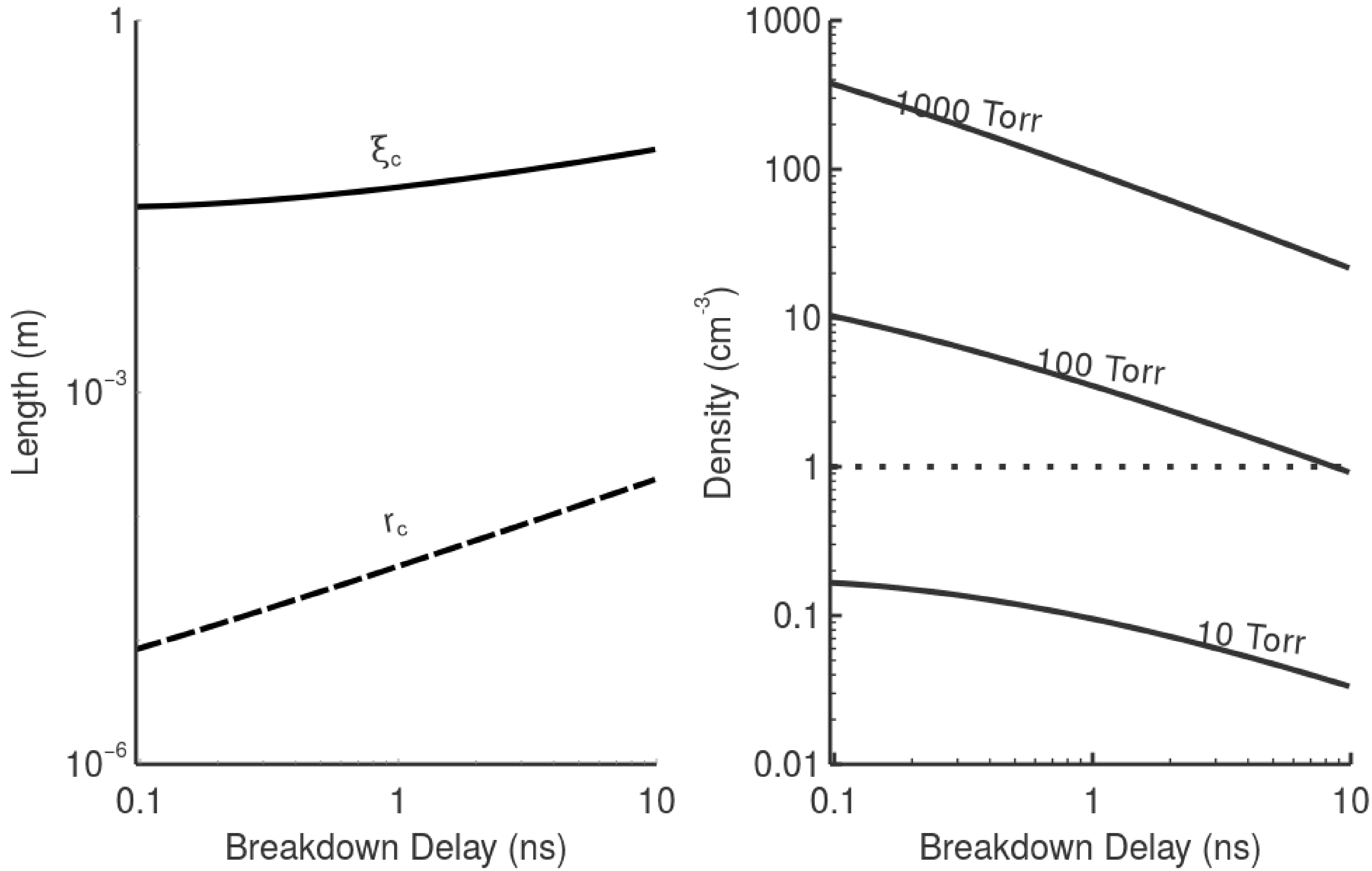


J. J. Thomson. "Notes on Recent Researches in Electricity and Magnetism." Clarendon Press, Oxford, UK, 1893.

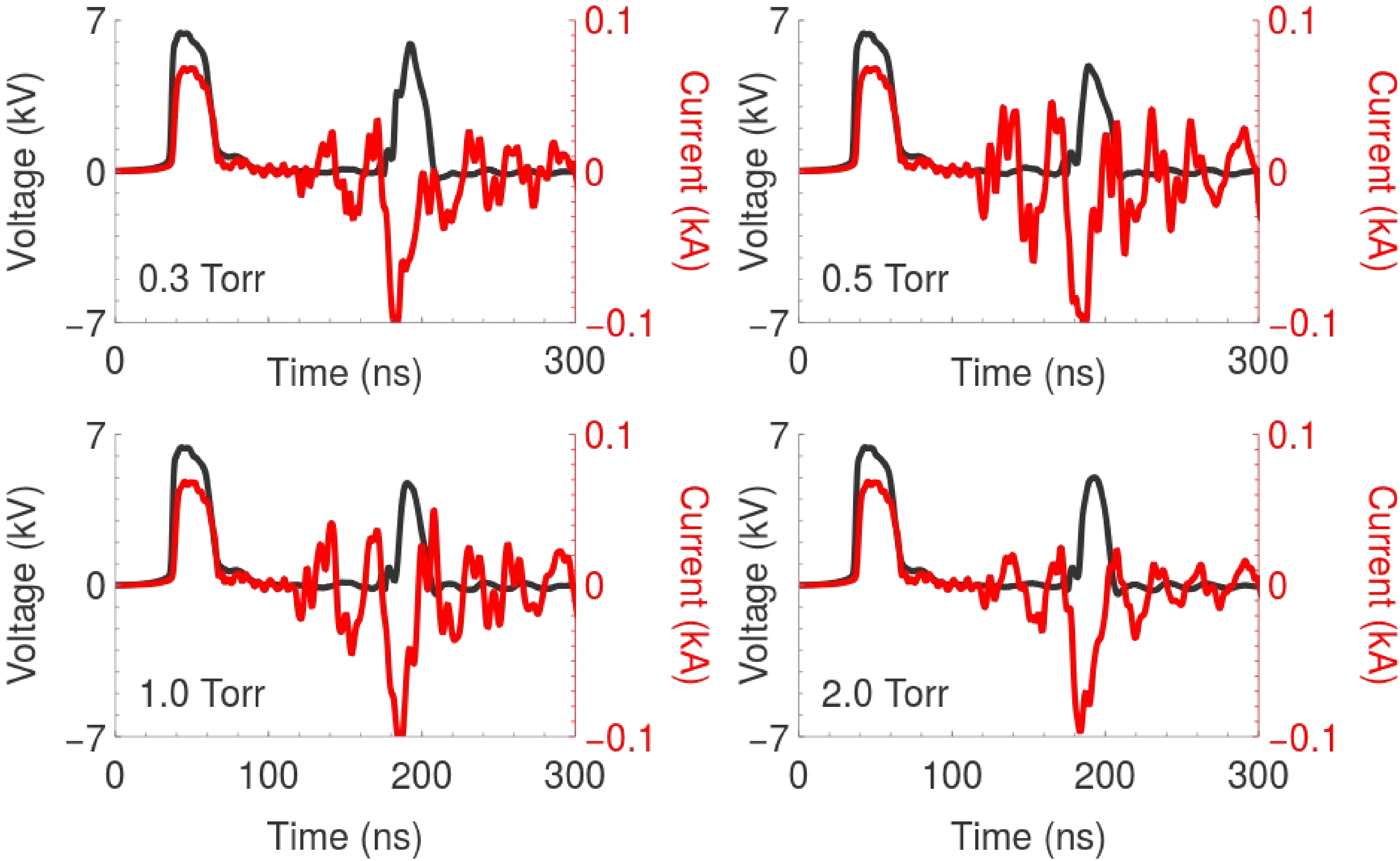
First Pulsed Plasma Measurements



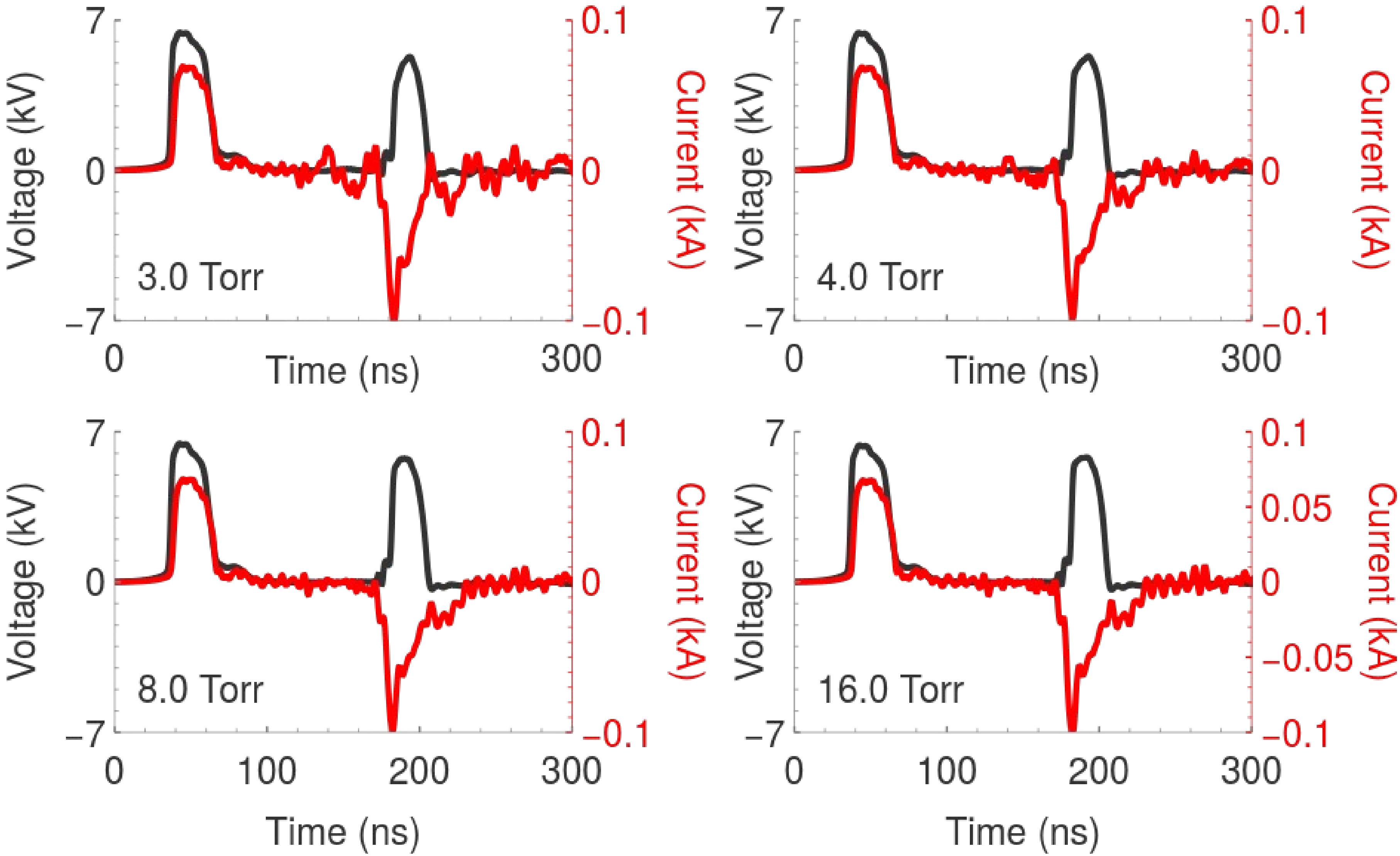
Pumping System



Homogeneous Streamers

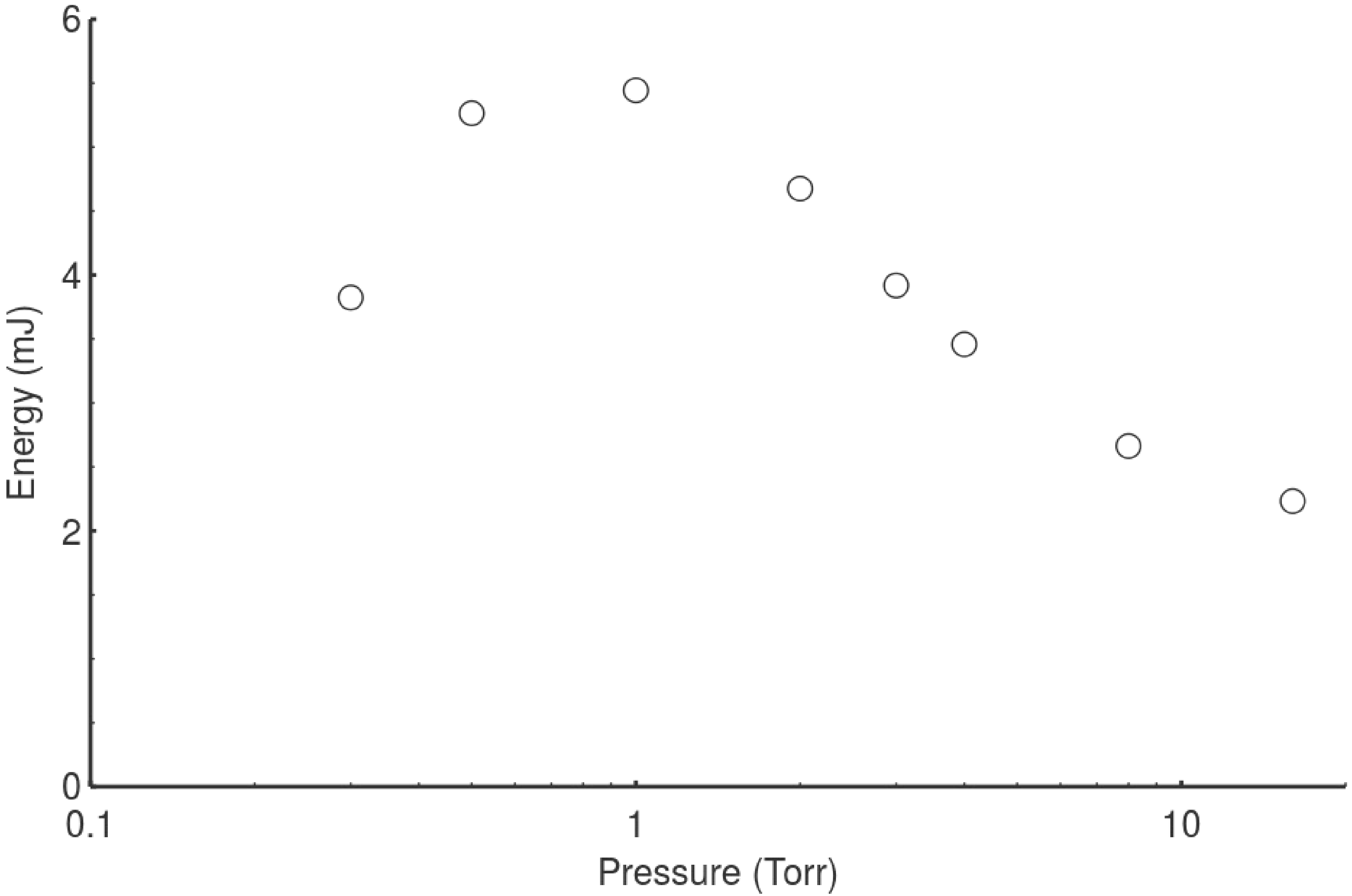

0.3 Torr
0.5 Torr
1.0 Torr
2.0 Torr
Voltage (kV)
Current (kA)
Time (ns)
7
0
−7
0.1
0
−0.1
0
100
200
300

Waveforms 1



7
0
−7
0.1
0
−0.1
Voltage (kV)
Current (kA)
3.0 Torr
0
300
Time (ns)
4.0 Torr
8.0 Torr
100
200
16.0 Torr
0.05
−0.05

Waveforms 2

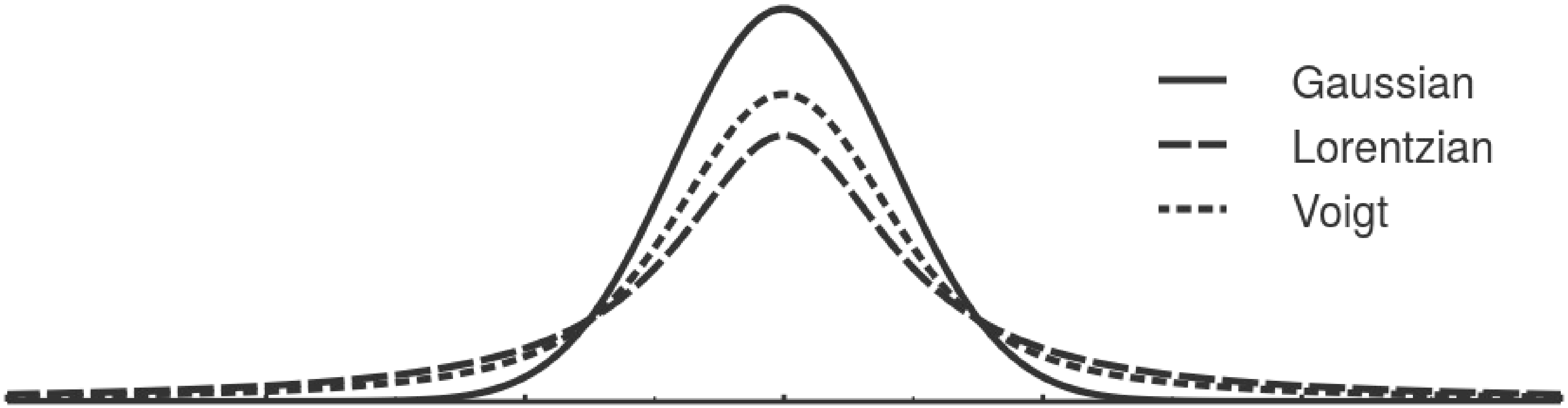


Energy Coupling

$$\Delta N = \Delta N_0 \frac{1}{1 + W\tau_{\text{eff}}} = \Delta N_0 \frac{1}{1 + I/I_{\text{sat}}}$$

$$W = \frac{\sigma I}{h\nu} \qquad \rightarrow \qquad I_{\text{sat}} = \frac{h\nu}{\sigma \tau_{\text{eff}}}$$

$$I_{\text{sat}} = \frac{2\sqrt{2} h_{21}}{\sigma \tau} = 0.45 \quad \text{mW} \cdot \text{cm}^{-2}$$



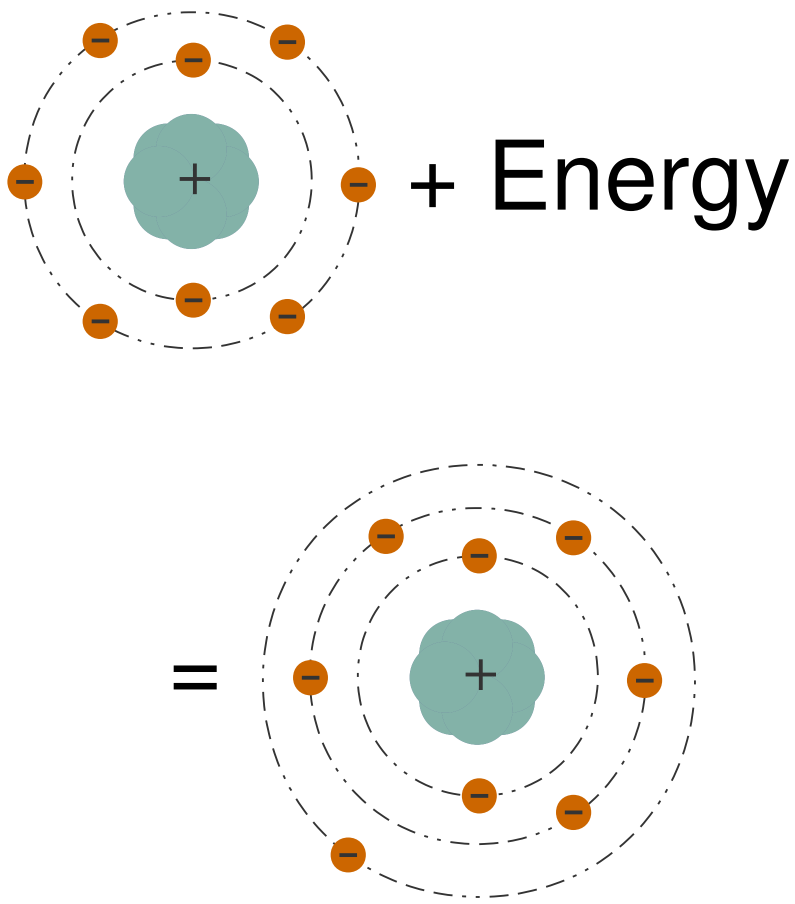
Lorentzian $$g(\omega) = -\frac{1}{4\pi^2}\frac{A\lambda^3}{\Delta\omega_a}\frac{1}{1+[2(\omega-\omega_a)/\Delta\omega_a]^2}$$

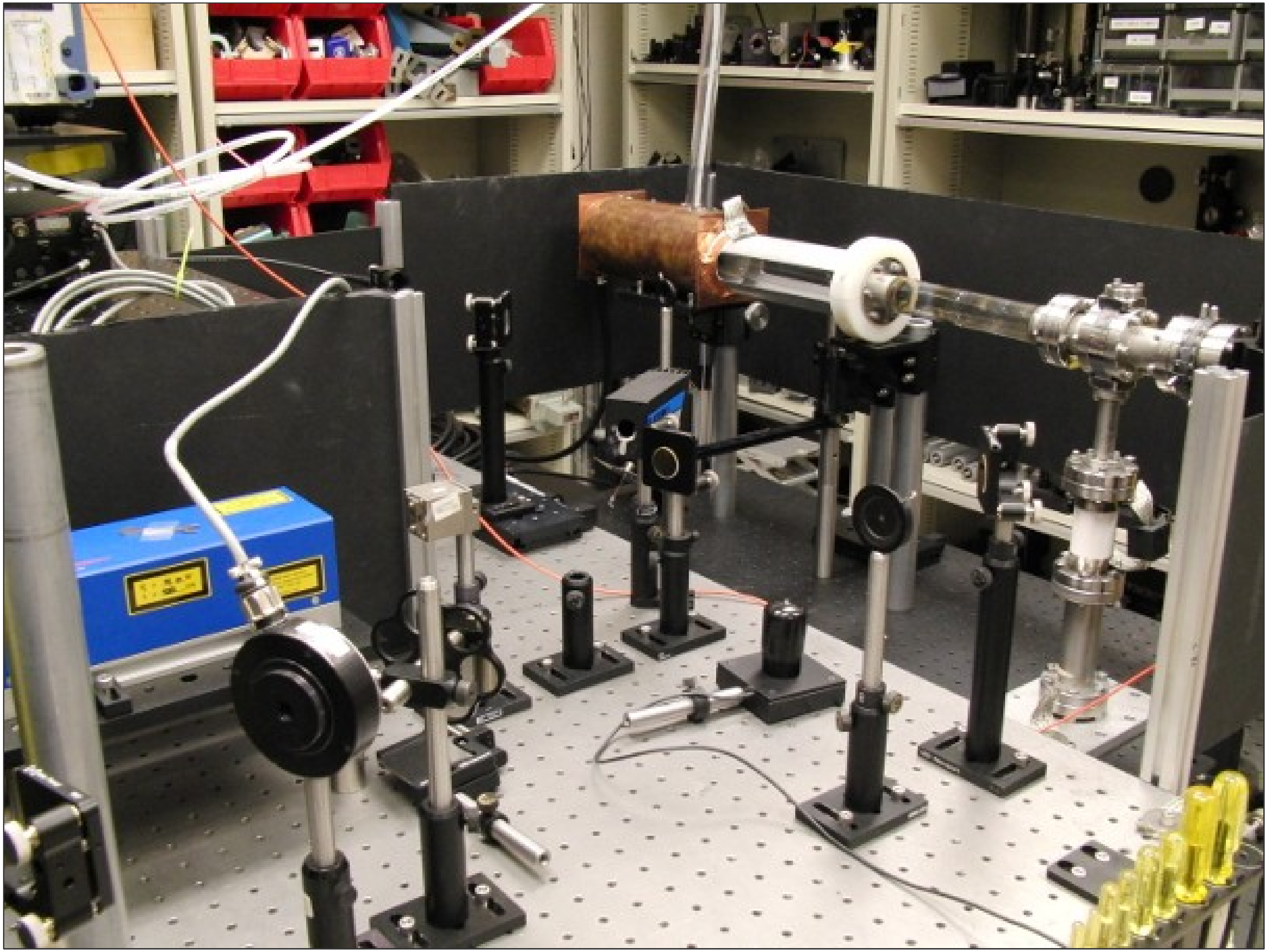
Voigt $$g(\omega) = \sqrt{\frac{2\ln 2}{\pi^3}}\frac{\Delta\omega_d}{\Delta\omega_d}\int_{-\infty}^{\infty}\frac{1}{[(\omega-\omega_a)-\omega']^2+4^2} \times \exp\left[4\ln 2\left(\frac{\omega'}{\Delta\omega_d}\right)^2\right]d\omega'$$

Gaussian $$g(\omega) = \sqrt{\frac{4\log 2}{\pi\Delta\omega_d^2}}\exp\left[-(4\log 2)\left(\frac{\omega-\omega_a}{\Delta\omega_d}\right)^2\right]$$

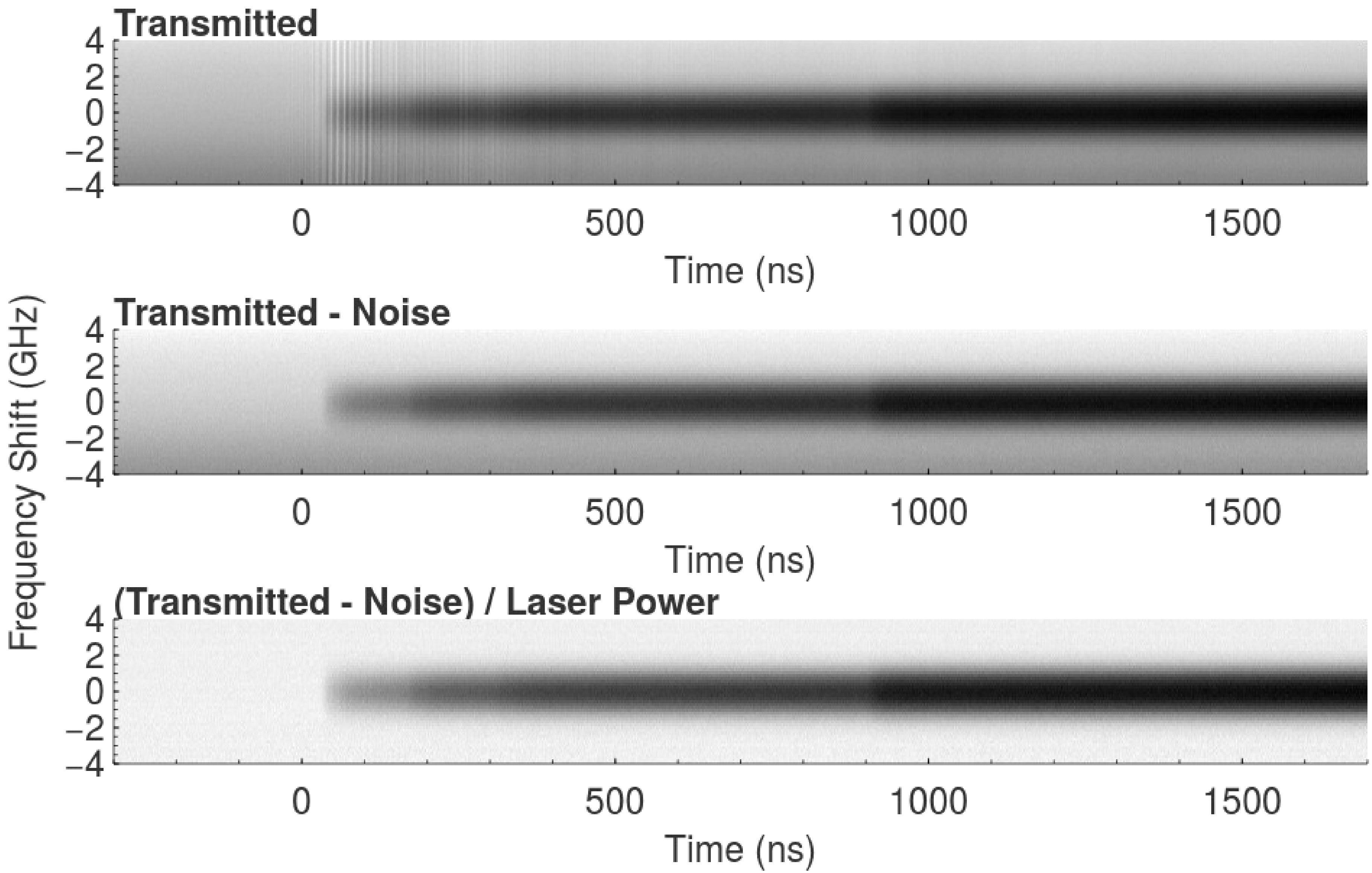
Lineshapes

- Energy added to atom
  - Collisions
  - Absorption of light
- One or more electrons to higher orbit $\rightarrow$ excited atom
- Excited state can emit photon to reach lower energy state
  - Occurs with a specific rate

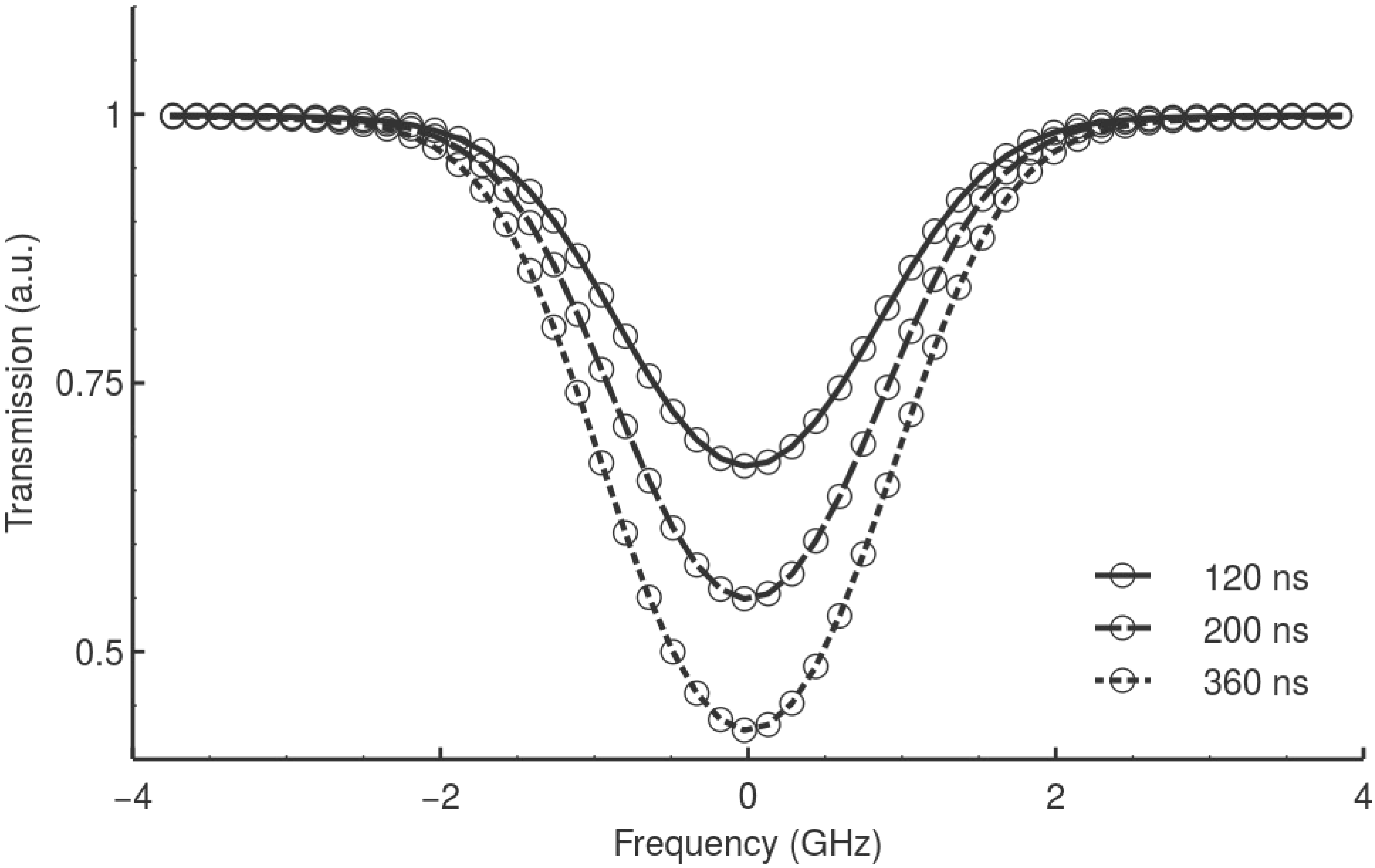




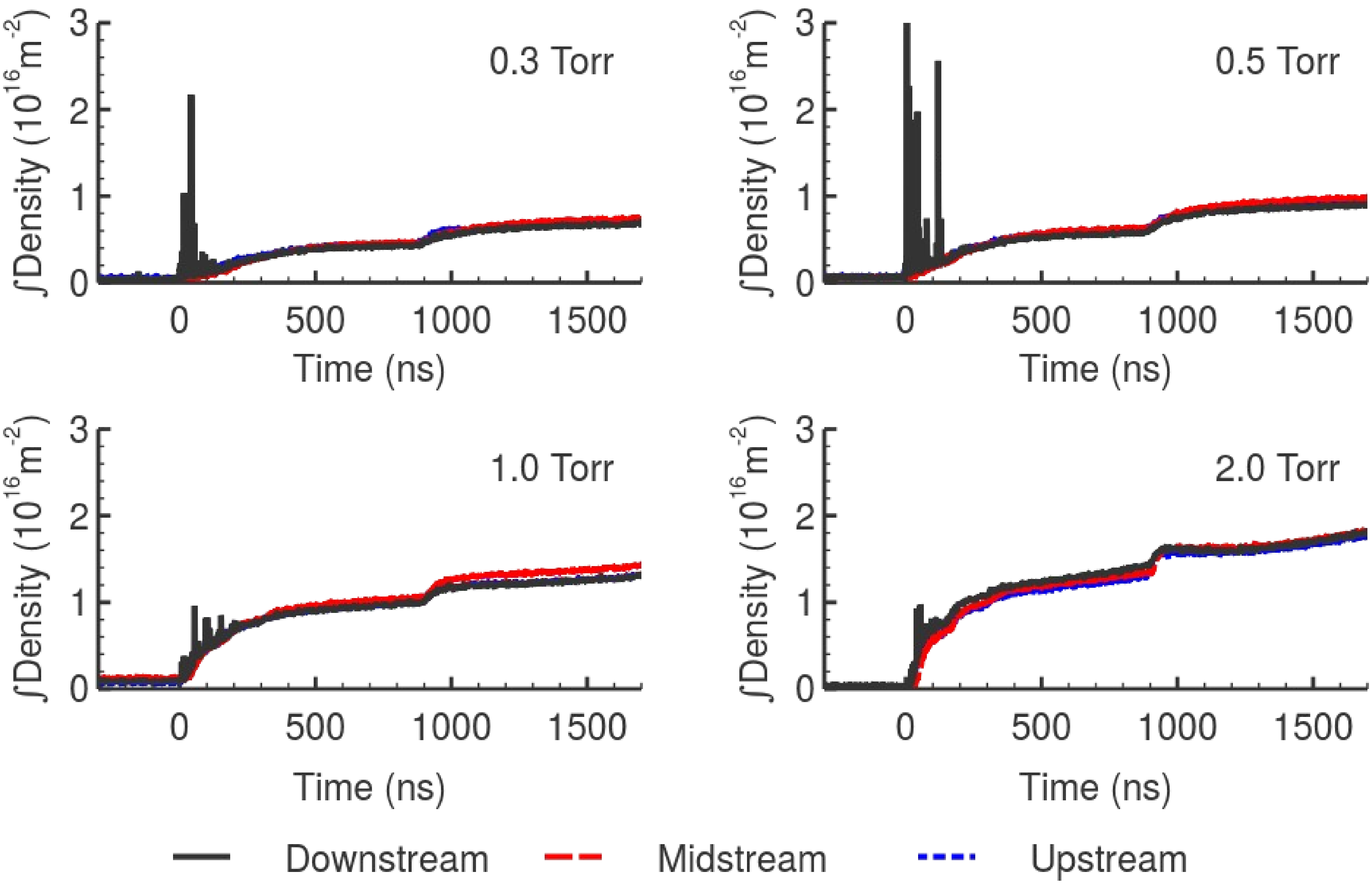
Laser-Absorption Spectroscopy Setup

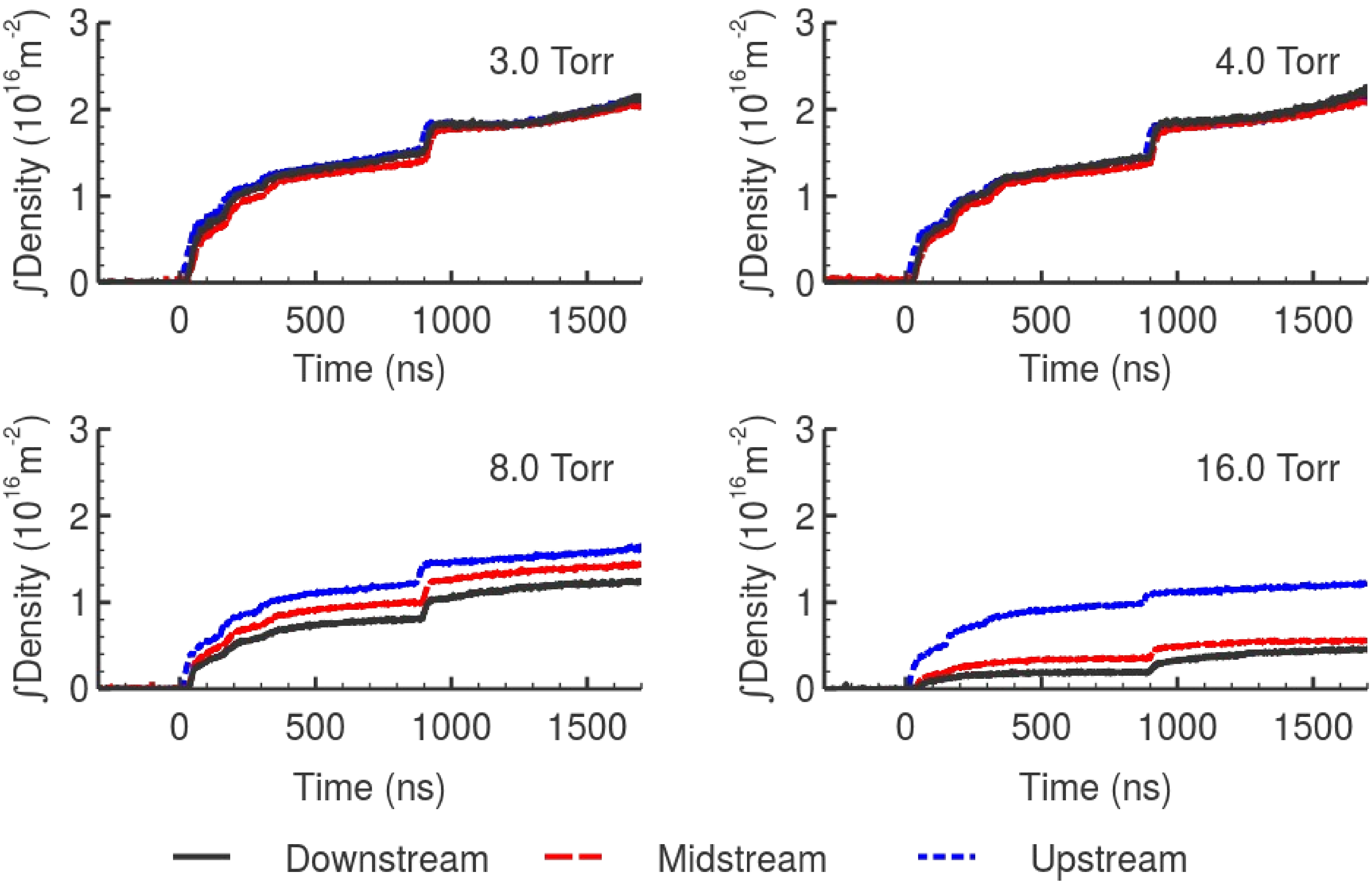


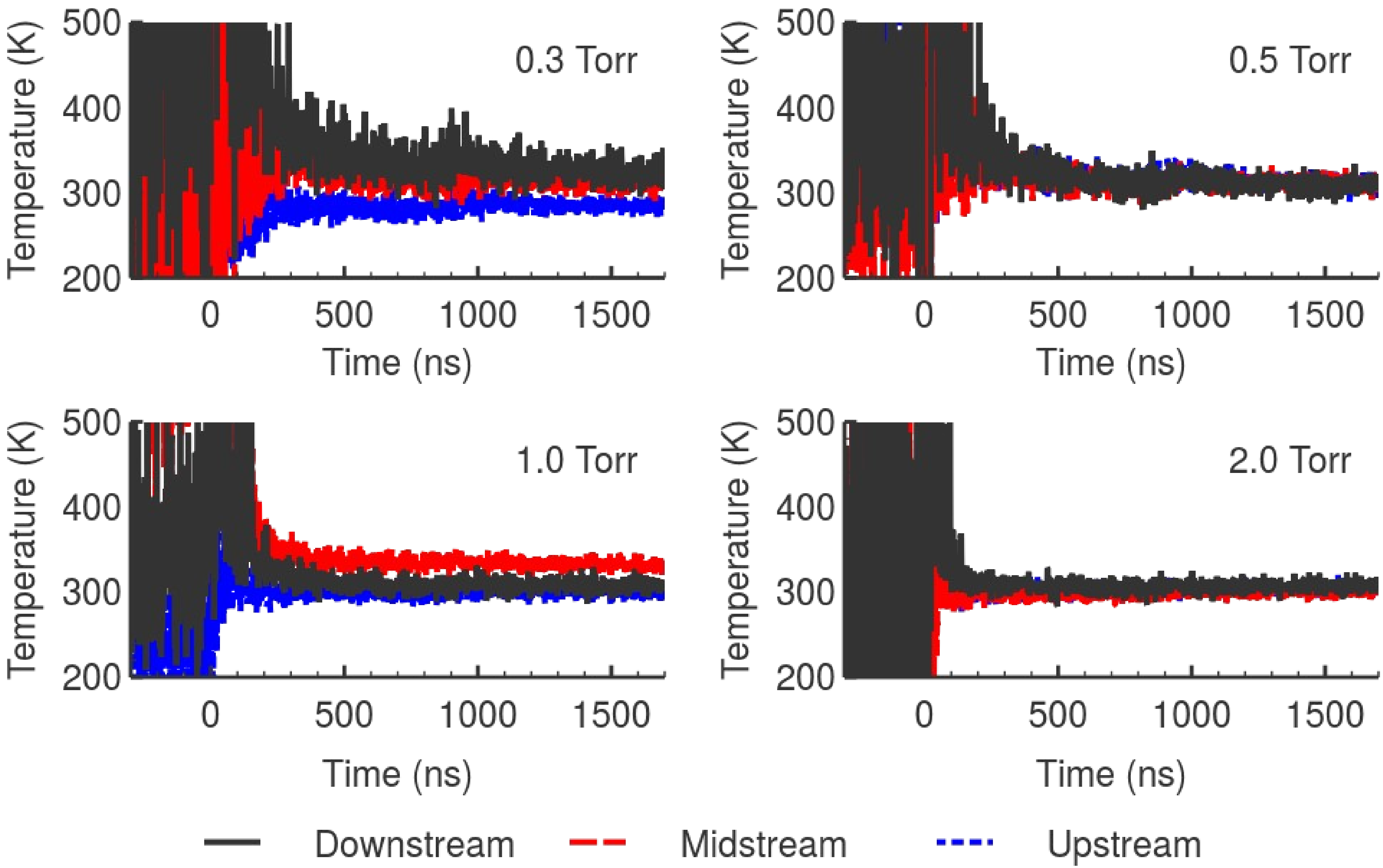
Spectral Post-Processing



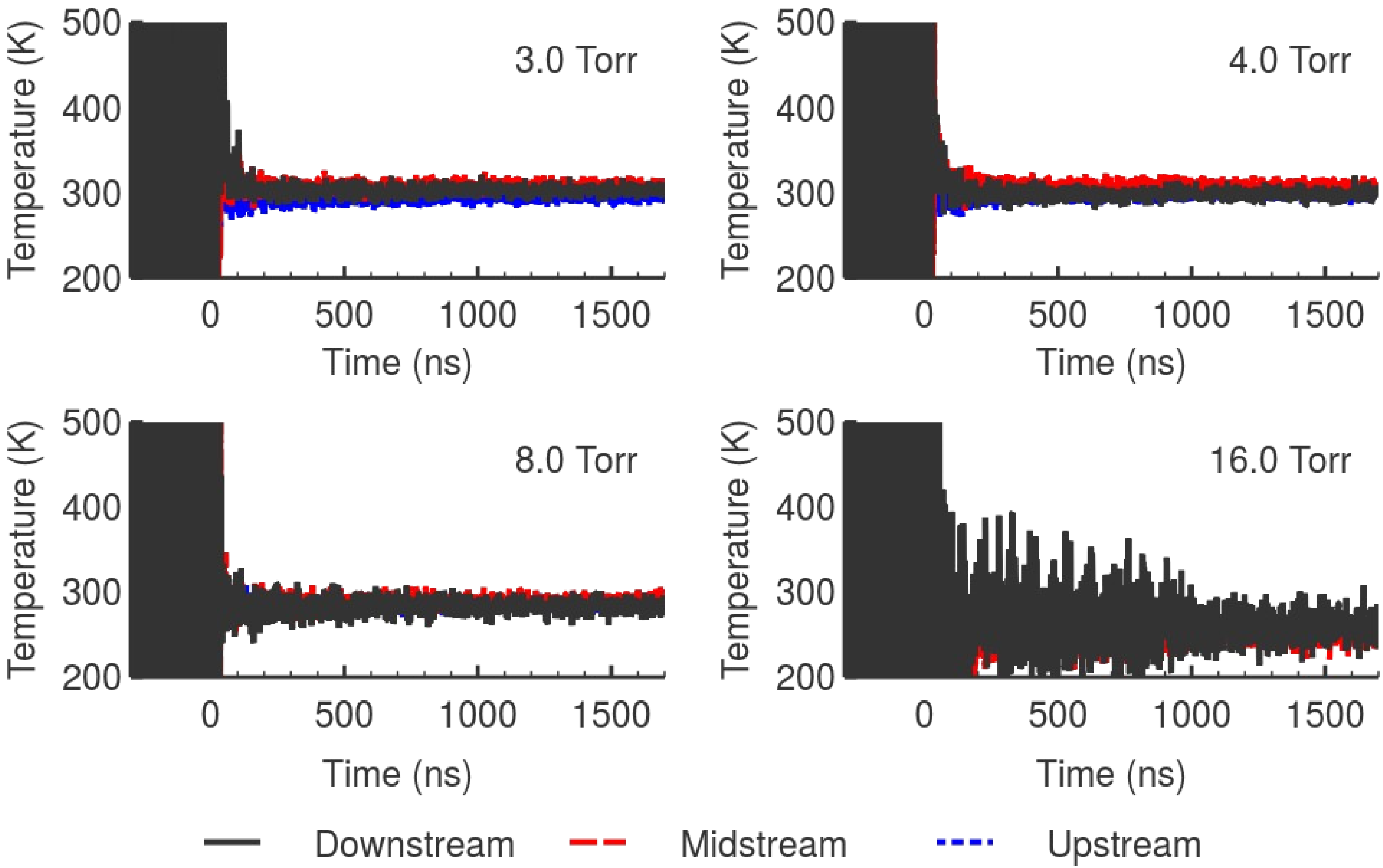
Absorption Spectra Matching

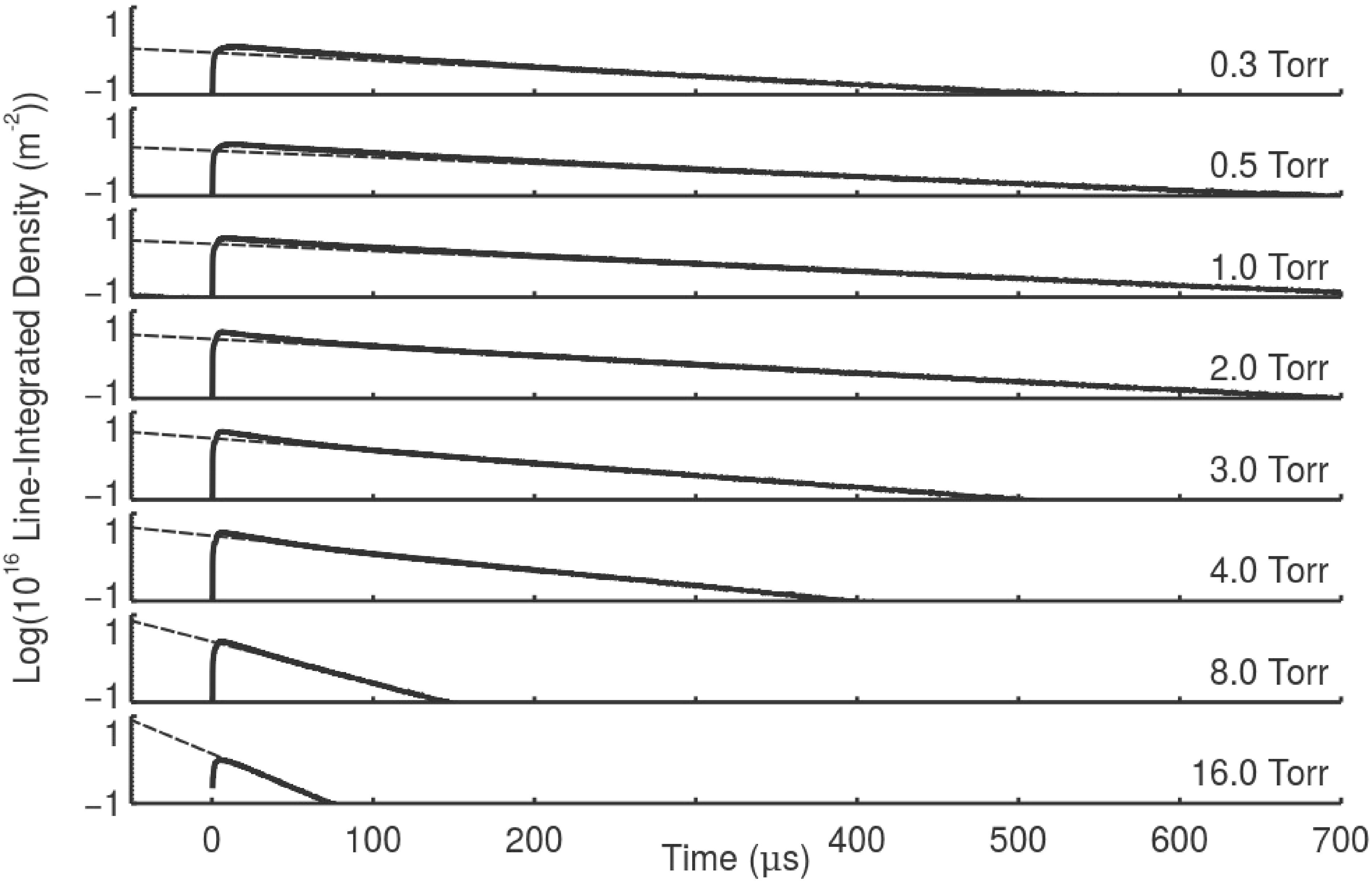

0.3 Torr
0.5 Torr
1.0 Torr
2.0 Torr
∫Density (10^{16}m^{-2})
Time (ns)
Downstream
Midstream
Upstream


3.0 Torr
4.0 Torr
8.0 Torr
16.0 Torr
∫Density (10^{16} m^{-2})
Time (ns)
Downstream
Midstream
Upstream



Temperatures 1


3.0 Torr
4.0 Torr
8.0 Torr
16.0 Torr
Temperature (K)
500
400
300
200
0
500
1000
1500
Time (ns)
Downstream
Midstream
Upstream



Metastable Decay

$$\frac{\partial f_\alpha}{\partial t} + \vec{v} \cdot \nabla f_\alpha + \frac{q_\alpha}{m_\alpha}\left(\vec{E} + \vec{v} \times \vec{B}\right) \cdot \nabla_v f_\alpha = \left(\frac{\partial f_\alpha}{\partial t}\right)_{\text{coll}}$$

- Describes the evolution of the distribution function for a particle α
- 7 independent variables
- Simple equilibrium solutions possible
  - Maxwell-Boltzmann
  - Druyvesteyn
- Generally difficult to solve otherwise

$$\frac{\partial n_\alpha}{\partial t} + \nabla \cdot (n_\alpha \vec{u_\alpha}) = G_\alpha - L_\alpha .$$

- Boltzmann equation can be converted into moments by integrating over velocity space
- Introduces particle density *n* and mean fluid velocity, *u*
- *G* – source terms
- *L* – loss terms

First Moment: Continuity Equation

$$m_\alpha n_\alpha \left[\frac{\partial \vec{u_\alpha}}{\partial t} + (\vec{u_\alpha} \cdot \nabla)\vec{u_\alpha} \right] = q_\alpha n_\alpha (\vec{E} + \vec{u_\alpha} \times \vec{B}) - \nabla \cdot \vec{\Pi} + \vec{f}|_{\text{coll}}$$

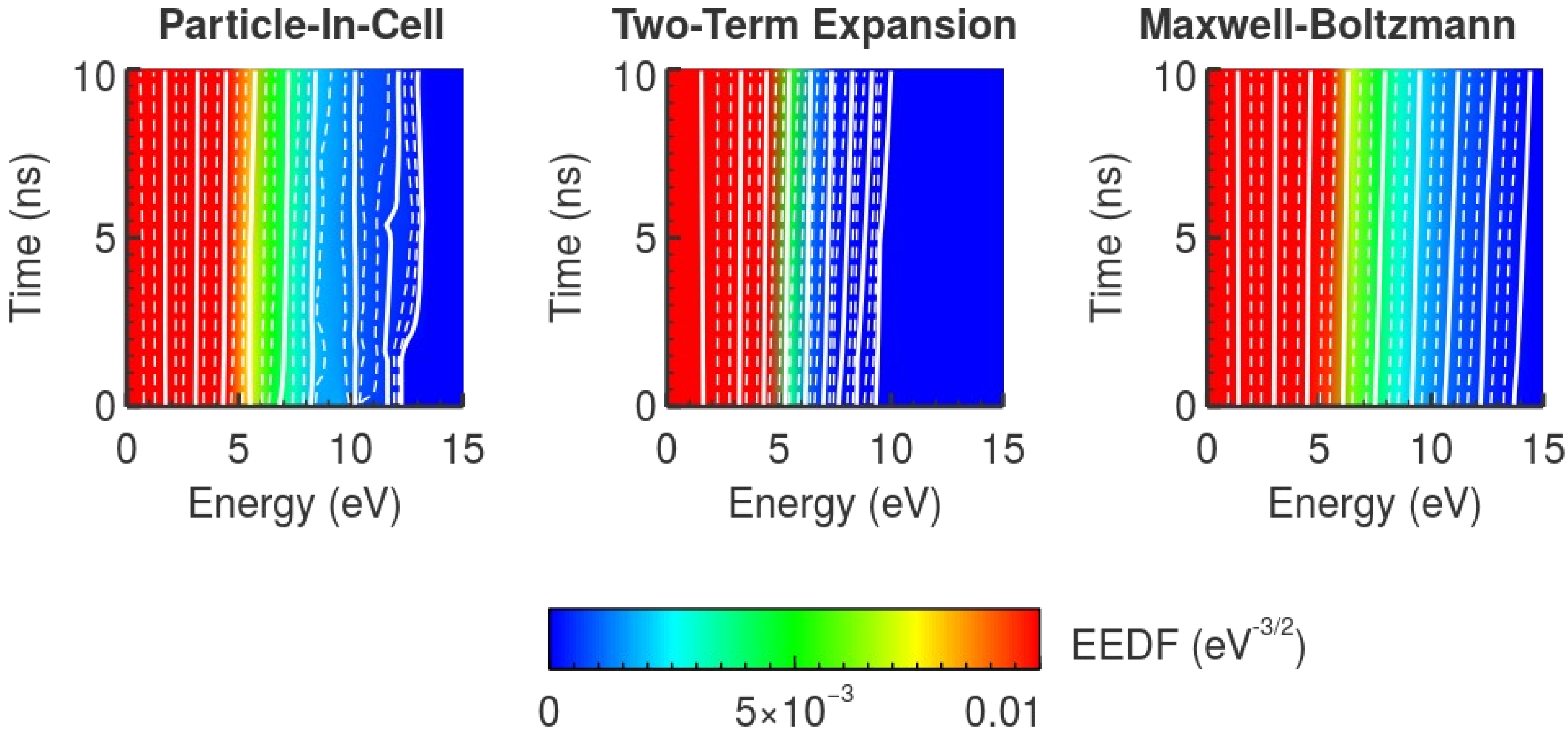
- Second momentum obtained by multiplying by velocity and integrating over velocity space
- Describes changes in fluid velocity over space
- Introduces pressure tensor Π.
- $f|_{\text{coll}}$ expresses change in momentum due to collisions

Second Moment: Conservation of Momentum

$$\frac{\partial}{\partial t}\left(\frac{3}{2}p_\alpha\right) + \nabla\cdot\frac{3}{2}(p_\alpha\vec{u}_\alpha) + p_\alpha\nabla\cdot\vec{u}_\alpha + \nabla\cdot\vec{q}_\alpha = \frac{\partial}{\partial t}\left(\frac{3}{2}p_\alpha\right)\Big|_{\mathrm{coll}}$$

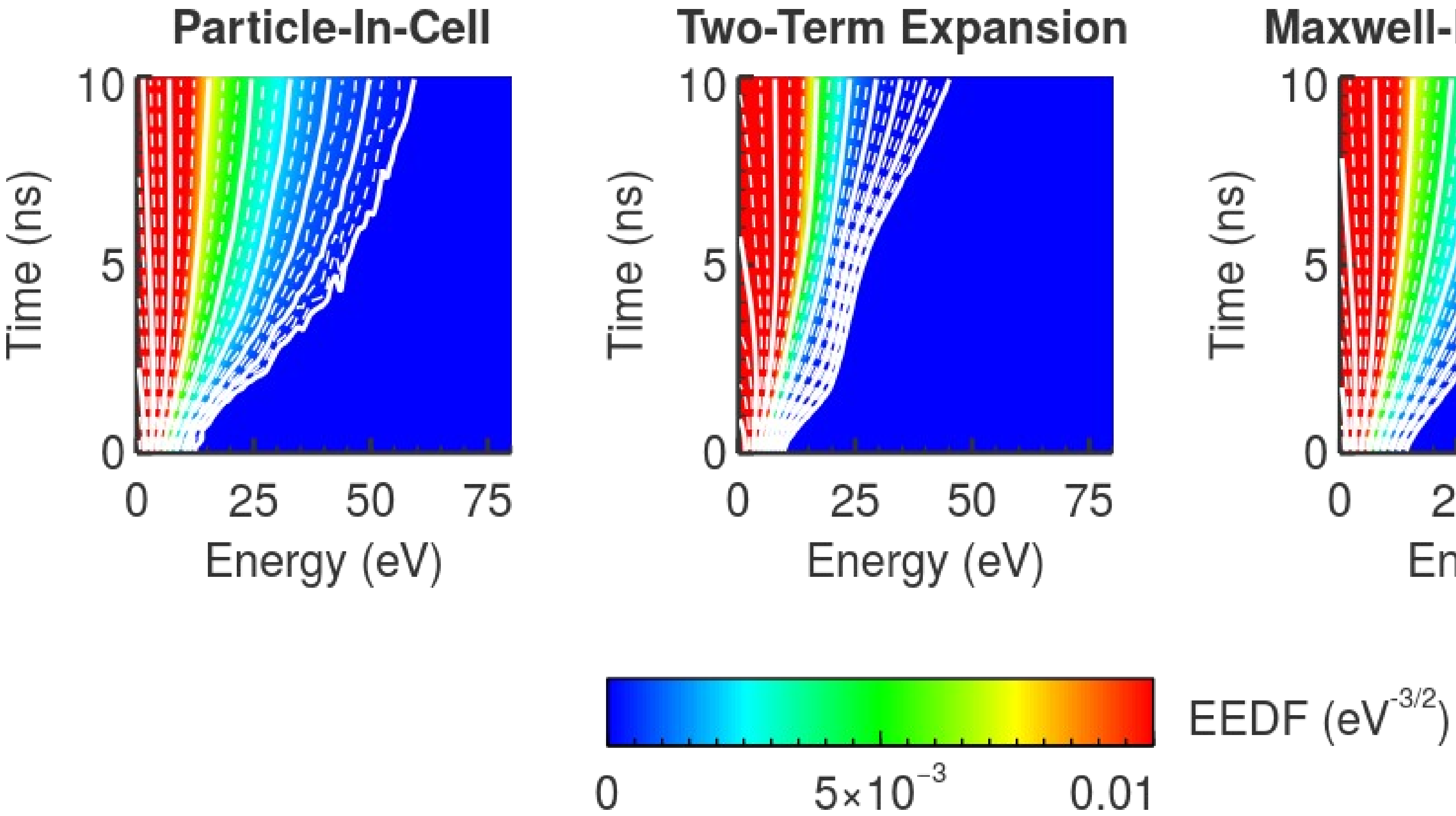
- Third momentum obtained by multiplying by energy and integrating of velocity space
- Energy expressed in terms of pressure, *p*
- RHS includes energy losses and gains resulting from collisions

- Must assume an EEDF for reaction rates
- Baseline: 0D particle-in-cell (XPDP1)
- Two approaches considered:
  - Maxwell-Boltzmann distribution
  - Solutions of two-term expansion of Boltzmann equation (BOLSIG+)



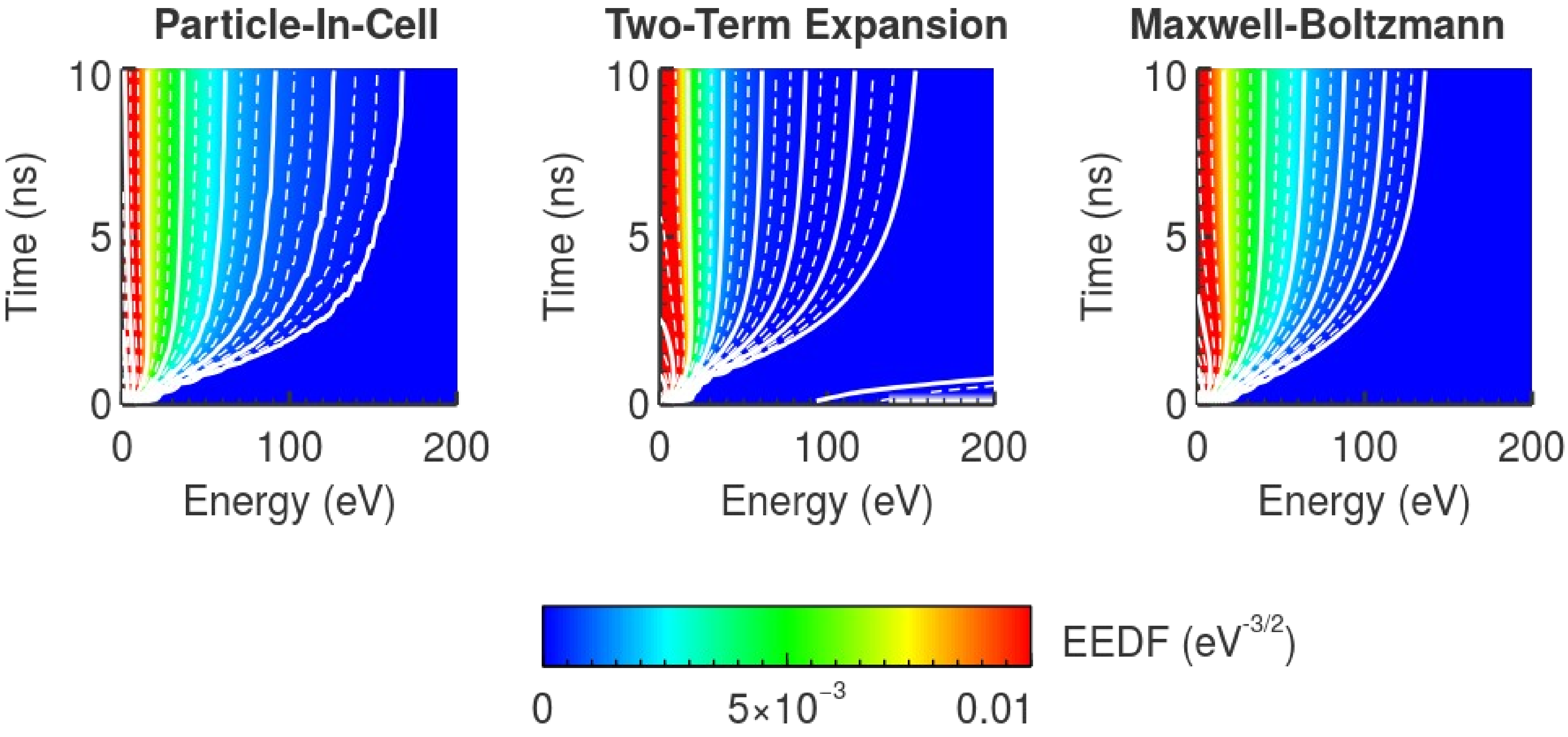
- Both approaches show reasonable agreement with particle-in-cell
- Two-term: less high-energy electrons
- Maxwell-Boltzmann: more high-energy electrons

EEDF Comparisons (10 Td)



- Unexpected contour variations in two-term expansion solutions
- Better agreement between Maxwell-Boltzmann and particle-in-cell

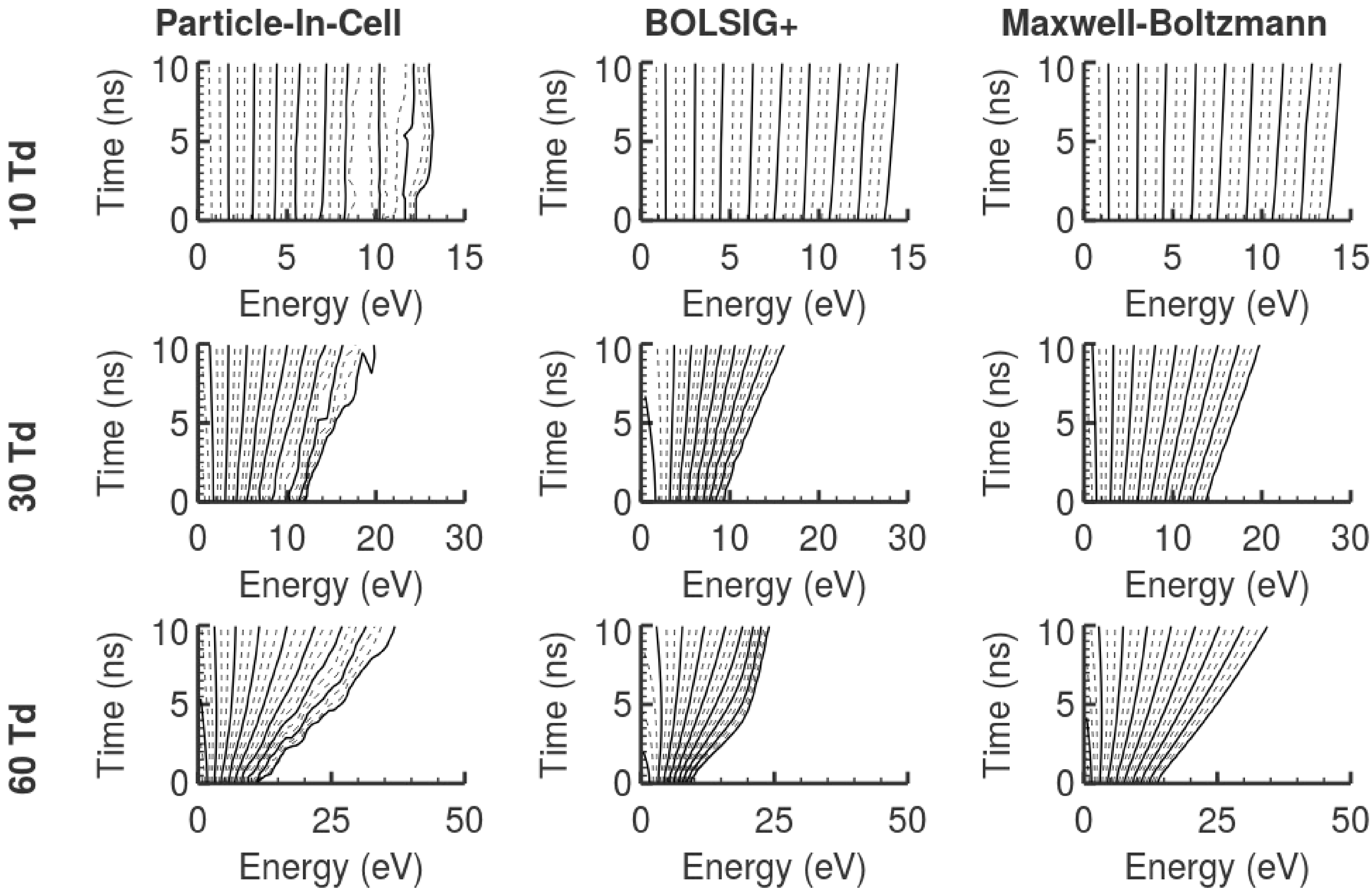
EEDF Comparisons (100 Td)



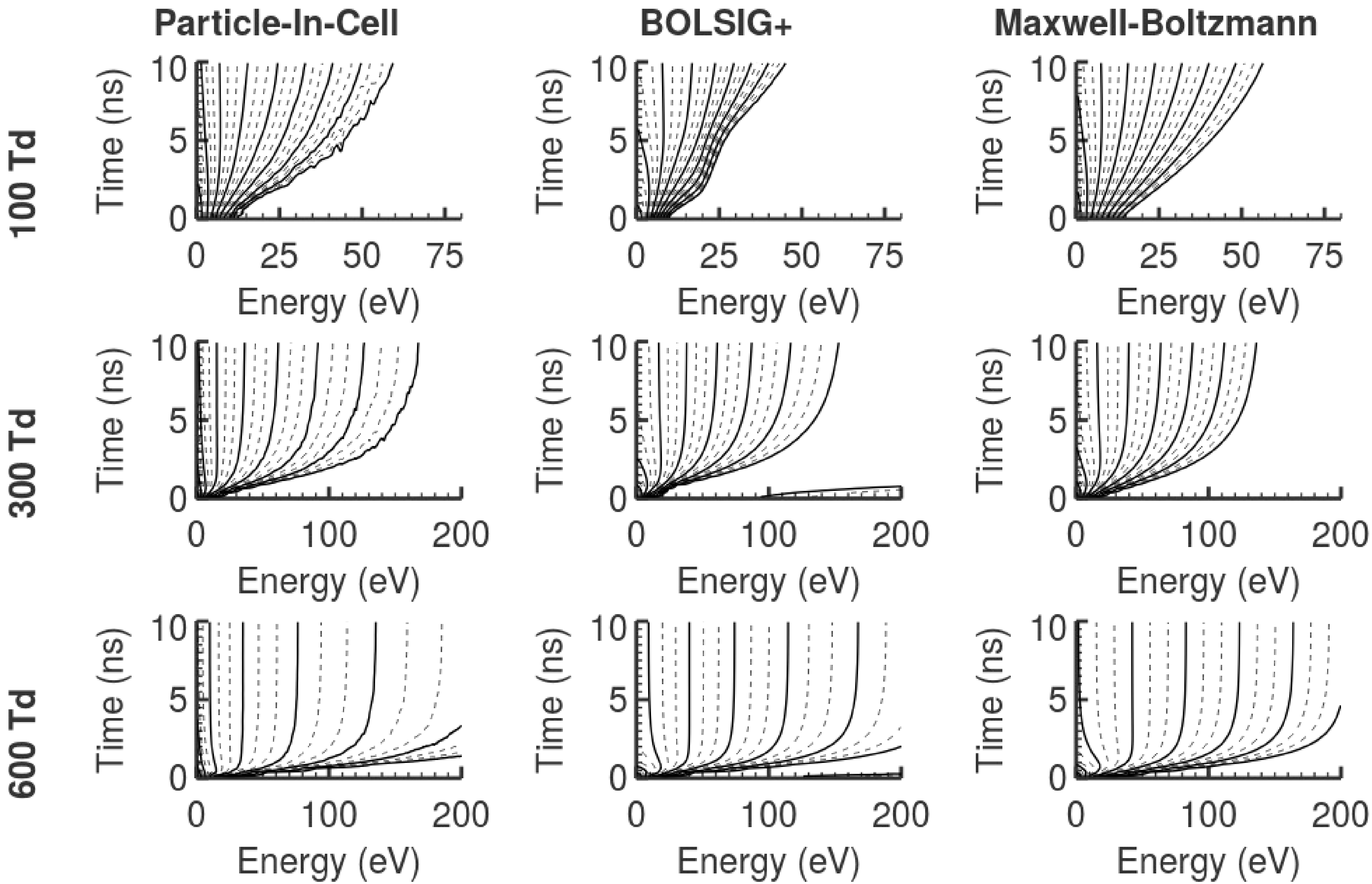
- Particle-in-cell: more 100 eV+ electrons than other results
- Two-term expansion solution best match this behavior
- Beginning of beam-like behavior?

EEDF Comparisons (300 Td)

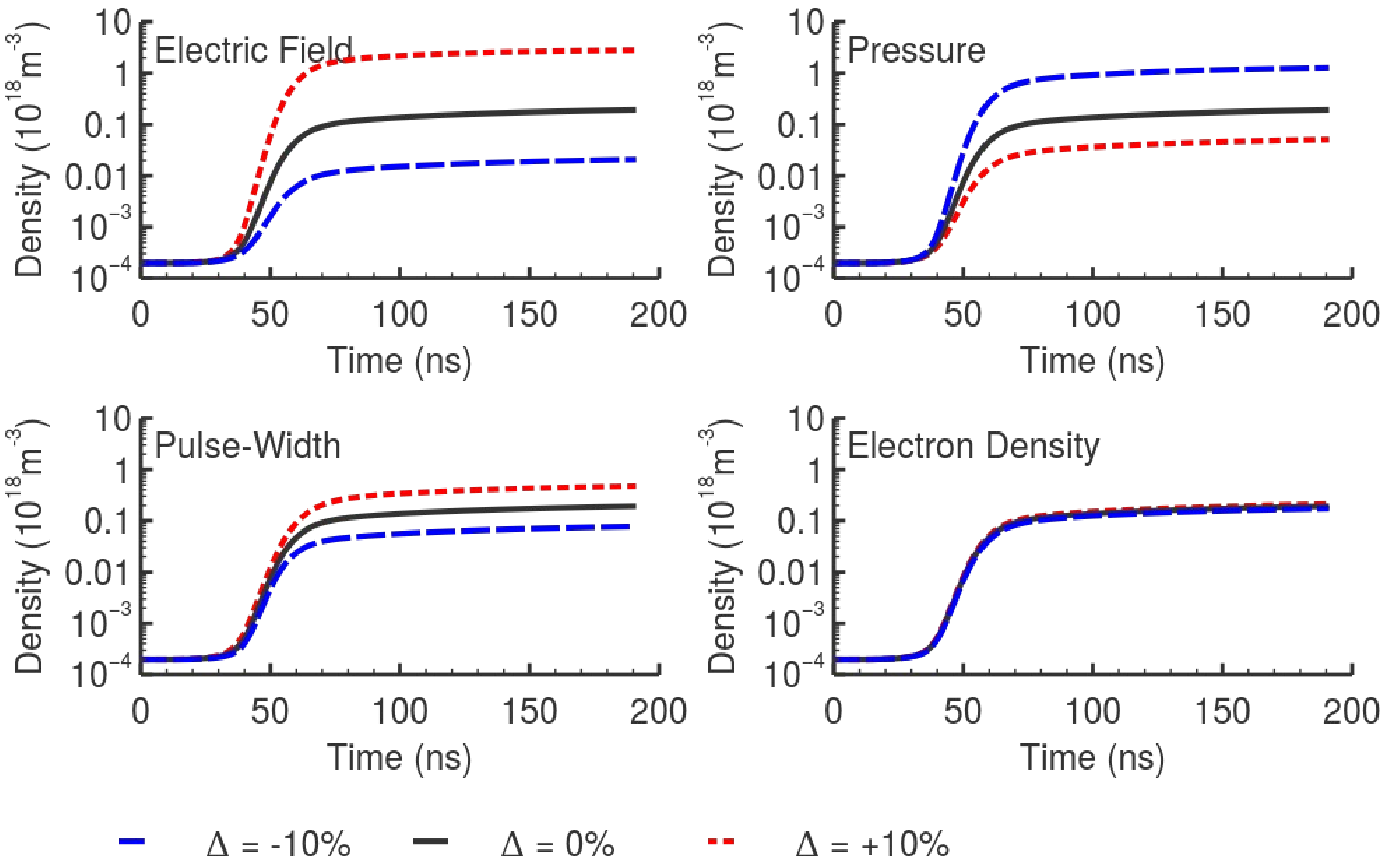
- $<$ 100 Td: Maxwell-Boltzmann gives best agreement with particle-in-cell
- $>$ 100 Td: BOLSIG+ gives best agreement with particle-in-cell
- Primary discrepancy in the number of high energy electrons: Particle-in-cell $>$ BOLSIG+ $>$ Maxwell-Boltzmann
- Better overall agreement led to use of Maxwell-Boltzmann distribution



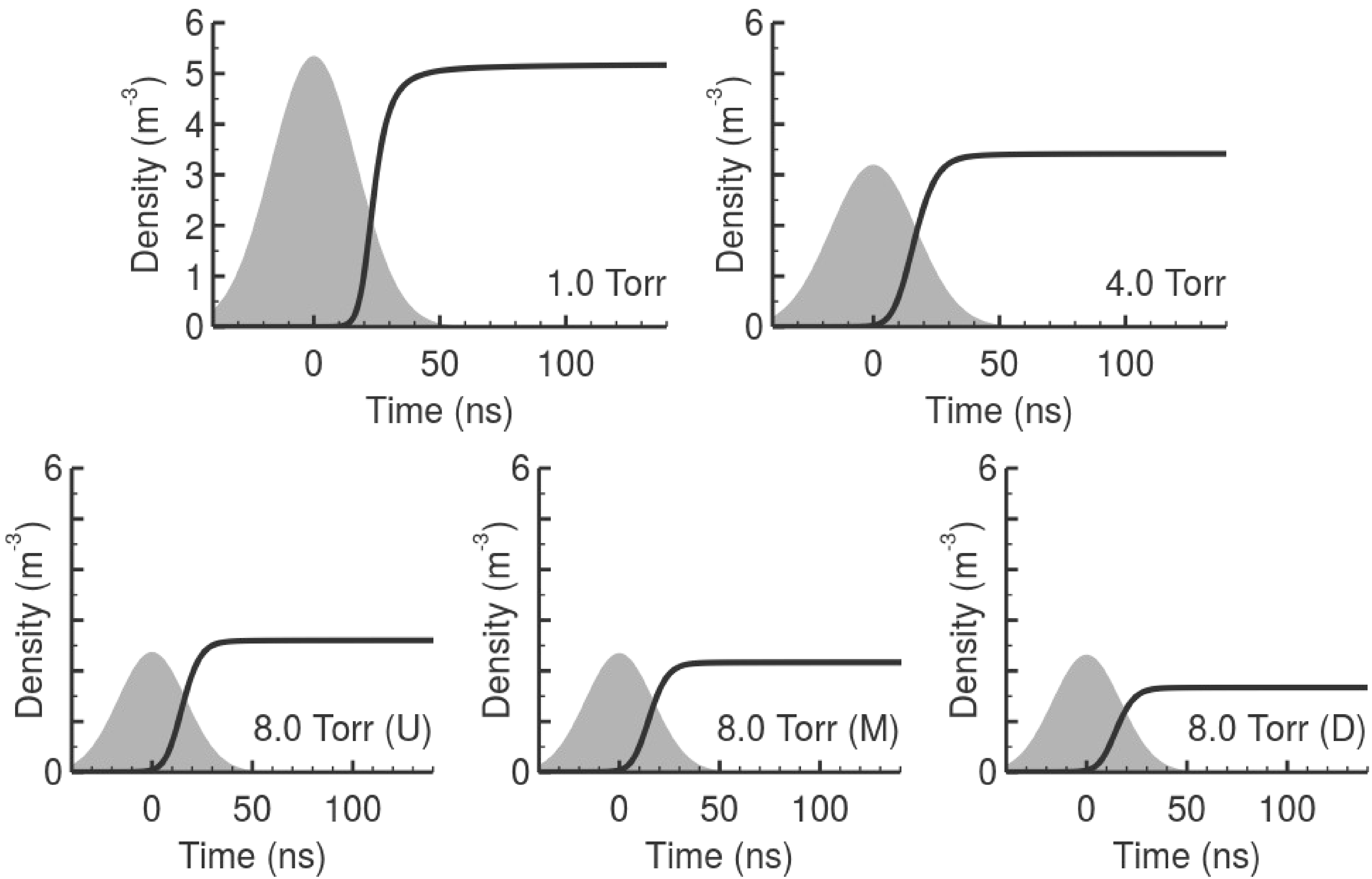
EEDF Contours 1



EEDF Contours 2

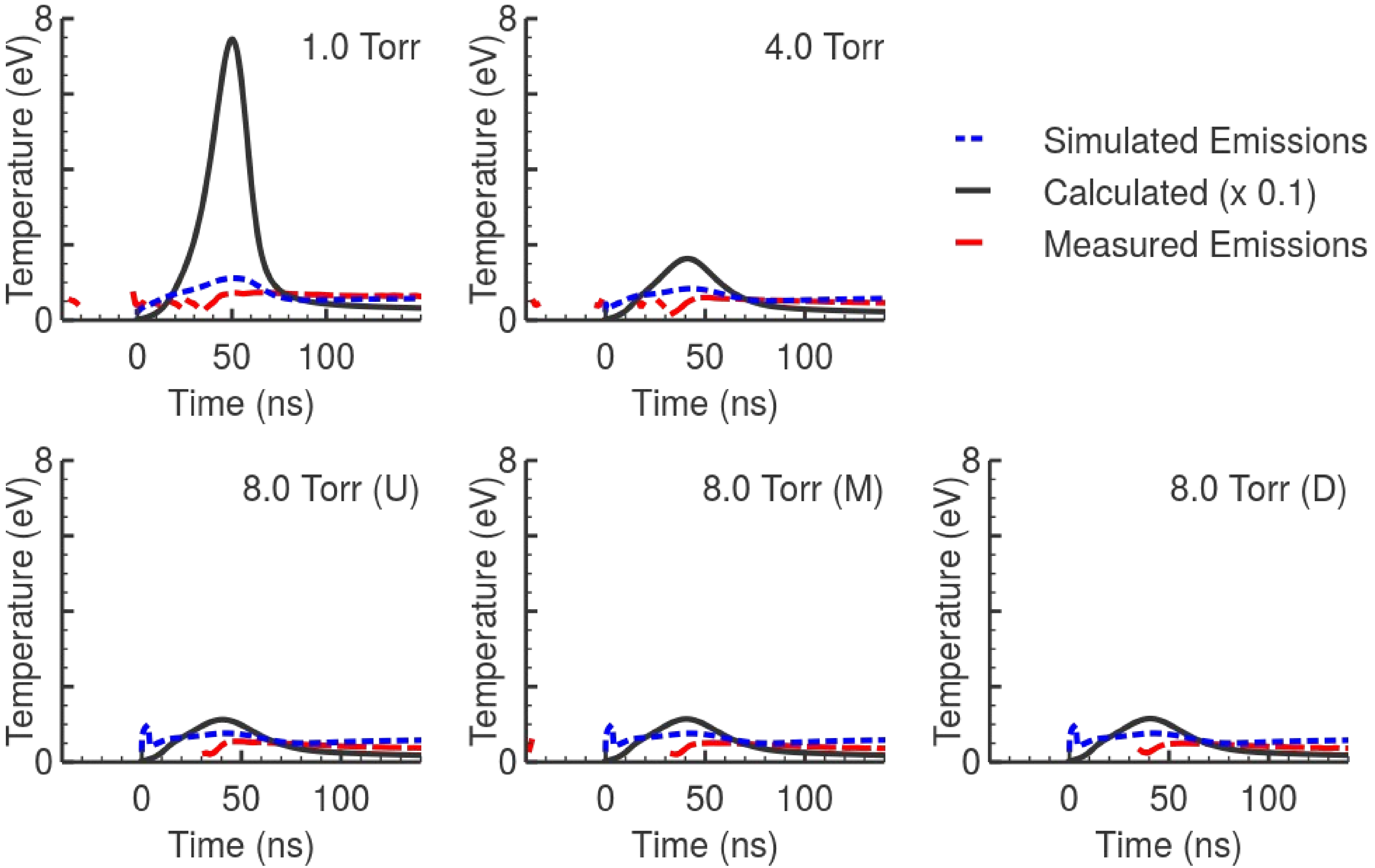


Perturbation Results



Electron Density Estimates

- Pressures: 0.3 – 16.0 Torr
- Spex HR460 monochromator, 0.88 nm bandpass
- 1,200 grooves/mm
- Photomultiplier tube, maximum 3.0 ns rise time
- Relative intensities calibrated with tungsten blackbody lamp



Boltzmann Plots