

# Spectroscopic Investigation of a Repetitively-Pulsed Nanosecond Discharge

by

Benjamin T. Yee

**Chair:** John E. Foster

- Introduction
- Pulsed plasmas and outstanding questions
- Metastable measurements
- Global model
- Emission measurements
- Conclusions

# Introduction



Karrer, M. "Sun 2011-12-08." URL: [http://www.flickr.com/photos/michael\\_karrer/6487199145/](http://www.flickr.com/photos/michael_karrer/6487199145/), Accessed: August 26, 2013.



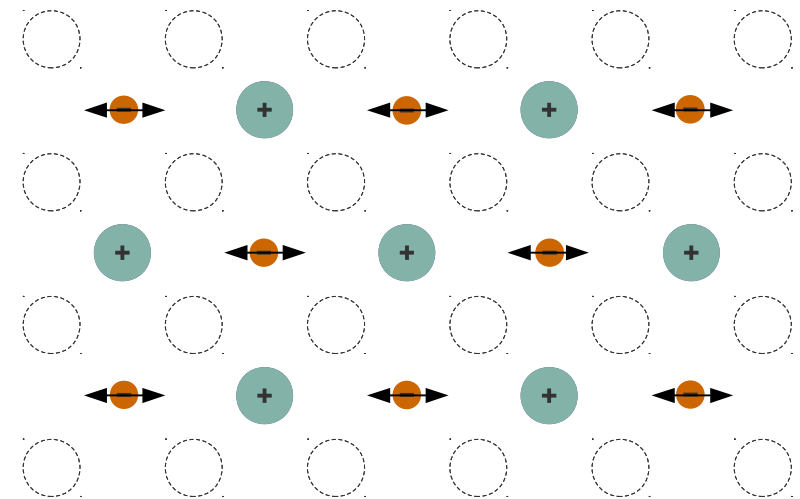
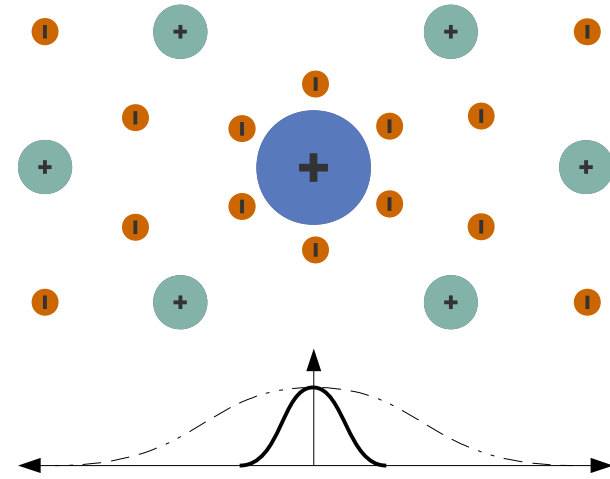
Hunt, P. "Lightning." URL: [http://www.flickr.com/photos/michael\\_karrer/6487199145/](http://www.flickr.com/photos/michael_karrer/6487199145/), Accessed: Aug. 26, 2013.



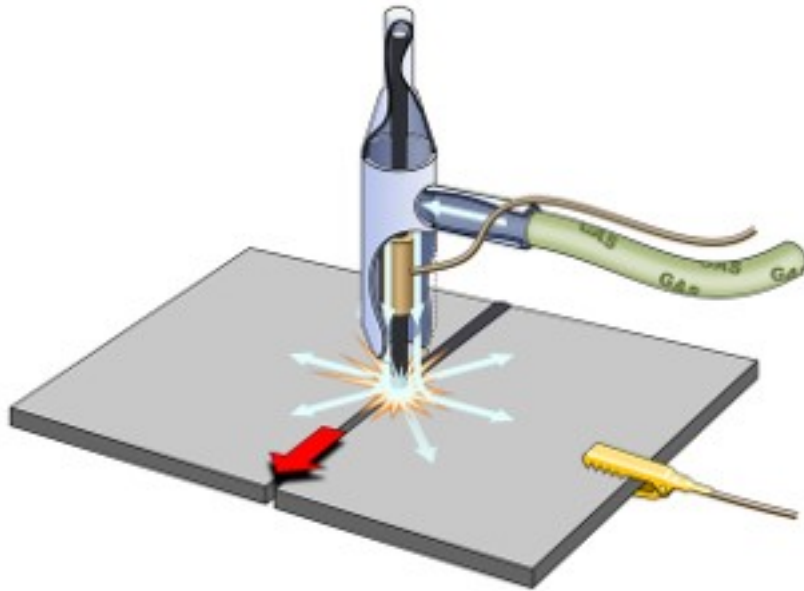
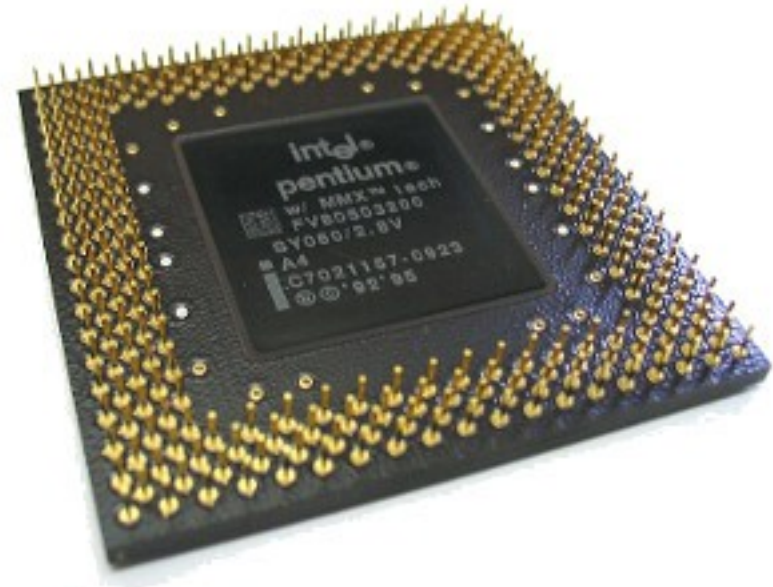
NASA, ESA, and the Hubble Heritage Team. "Star-Forming Region LH 95 in the Large Magellanic Cloud." URL: <http://hubblesite.org/gallery/album/entire/pr2006055a/>, Accessed: August 27, 2013.

# What Is a Plasma? (Interstellar Gases)

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







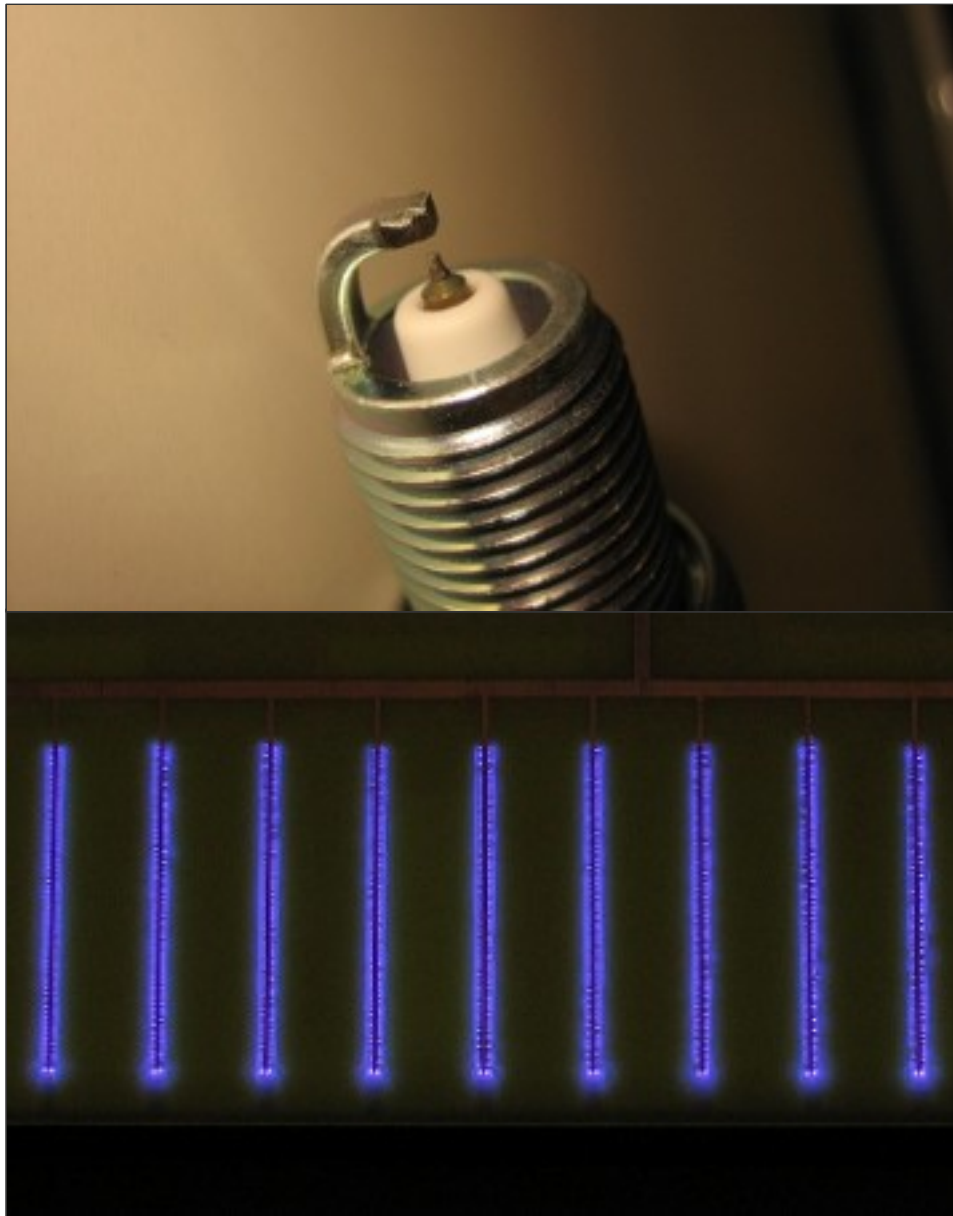
Bowden, M. "A fluorescent light bulb." URL: <http://www.sxc.hu/photo/203835>, Accessed: August 26, 21013.

Shigeru23. "Gas arc welding (TIG & MIG)." URL: [https://commons.wikimedia.org/wiki/File:Gas\\_arc\\_welding\\_%28TIG\\_%26\\_MIG%29.PNG](https://commons.wikimedia.org/wiki/File:Gas_arc_welding_%28TIG_%26_MIG%29.PNG), Accessed: August 27, 2013.

Schmid, M. "Wasabi-Chips (Migros)." URL: [https://commons.wikimedia.org/wiki/File:Wasabi-Chips\\_%28Migros%29.JPG](https://commons.wikimedia.org/wiki/File:Wasabi-Chips_%28Migros%29.JPG), Accessed: August 27, 2013.

Volant, L. "LG LV3550 15 R." URL: <http://www.flickr.com/photos/27048731@N03/5589223946/>, Accessed: August 27, 2013.

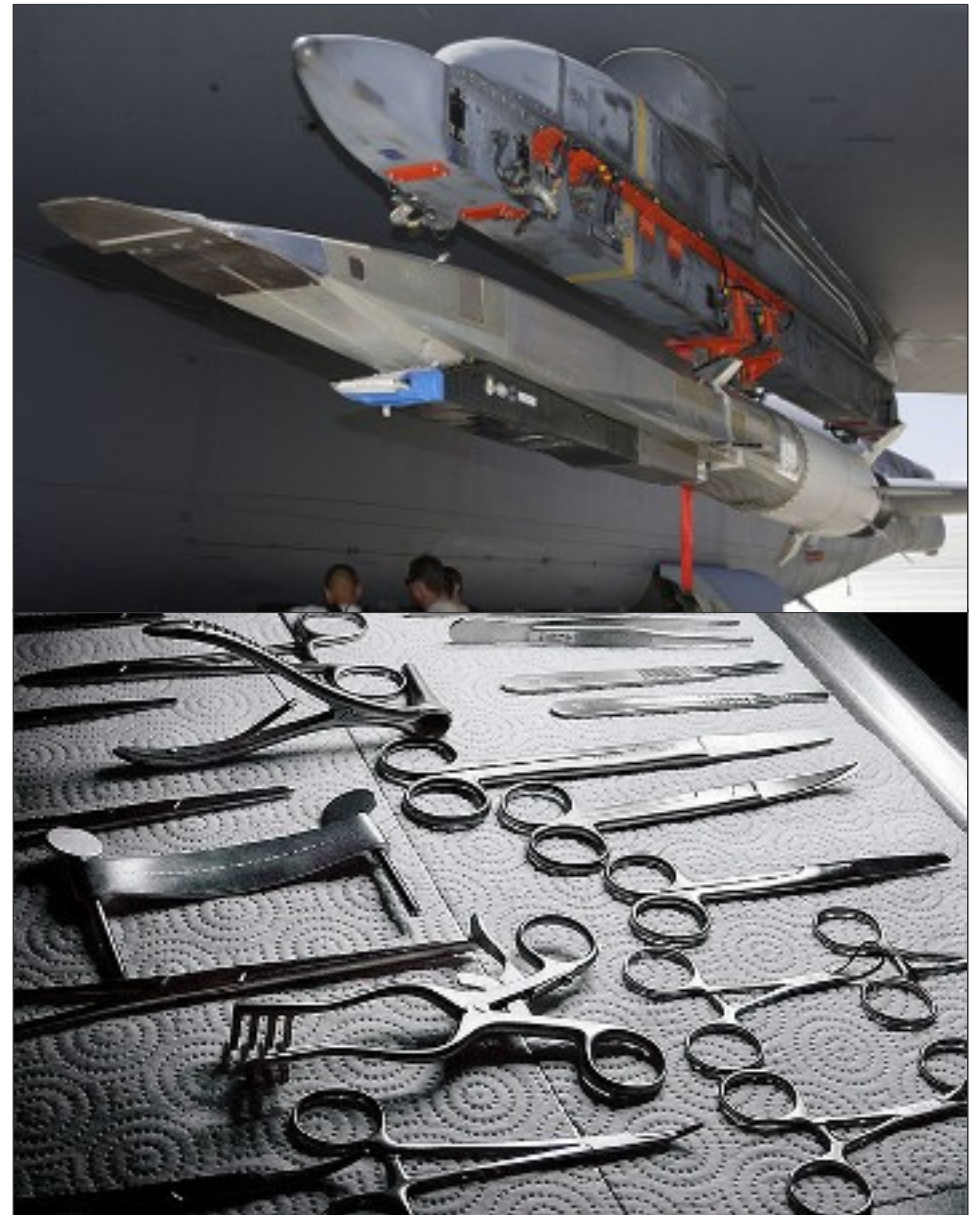




- Modification or enhancement of fuel combustion properties
- Modification of airflow over surfaces without mechanical actuators

Wong, N. "Tip of a spark plug." URL: <http://www.flickr.com/photos/14029705@N00/375024452/>, Accessed: August 27, 2013.  
Xunger, "Plasma glow discharge." URL: <https://commons.wikimedia.org/wiki/File:Streamwise.JPG>, Accessed: August 27, 2013.

- Ionization of airflow for MHD generators and energy bypasses
- Sterilization of bacterial, viral, and chemical contaminants



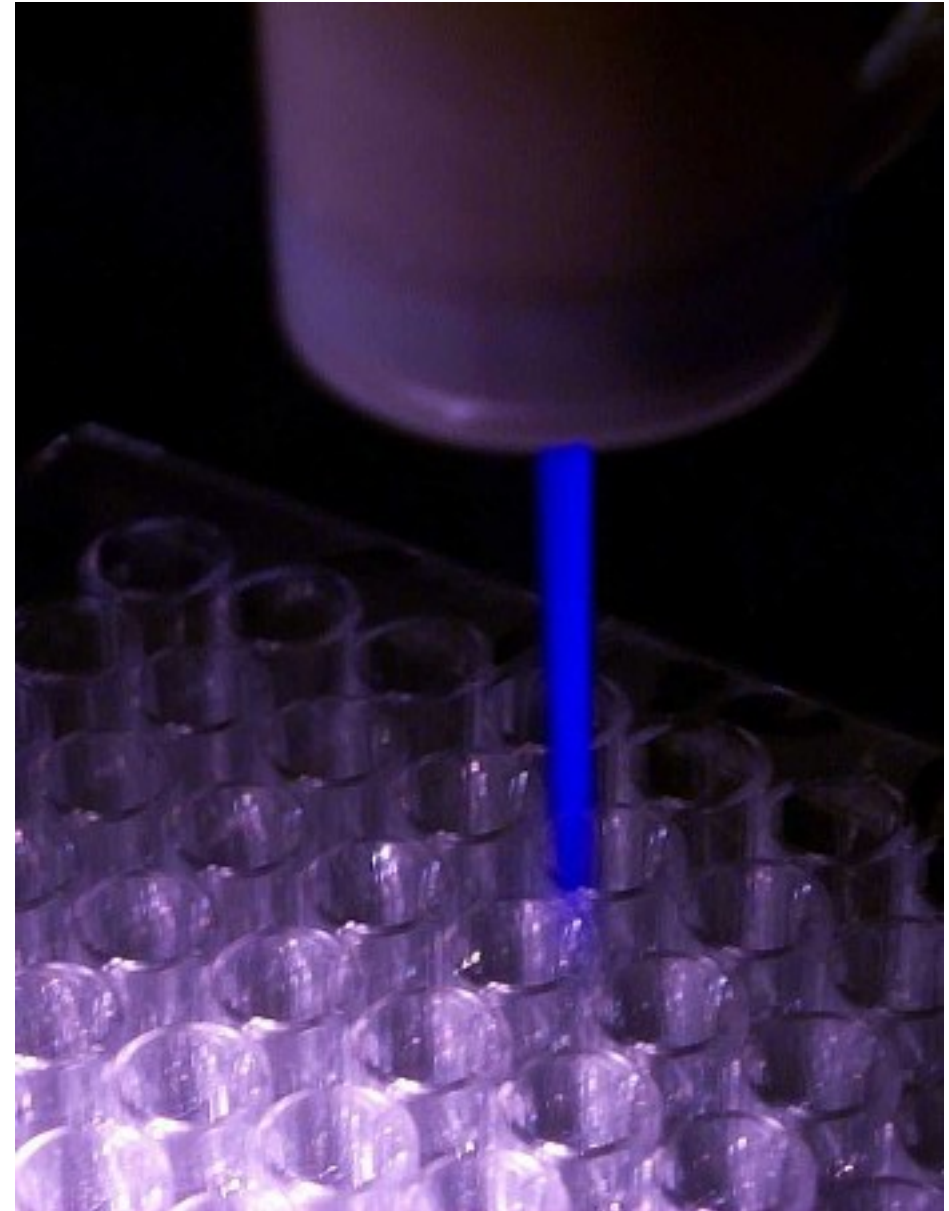
Bellay, C. "X-51A Waverider on B-52 2009." URL: [https://commons.wikimedia.org/wiki/File:X-51A\\_Waverider\\_on\\_B-52\\_2009.jpg](https://commons.wikimedia.org/wiki/File:X-51A_Waverider_on_B-52_2009.jpg), Accessed: August 27, 2013.  
parfe\_alp, "Operation." URL: <http://www.flickr.com/photos/parfe/2239523465/>, Accessed: August 27, 2013.



- Common in welding, cutting, and arc lighting
- All particle species at approximately the same temperature
  - Electrons
  - Ions
  - Neutrals
- Often several thousand degrees Celsius

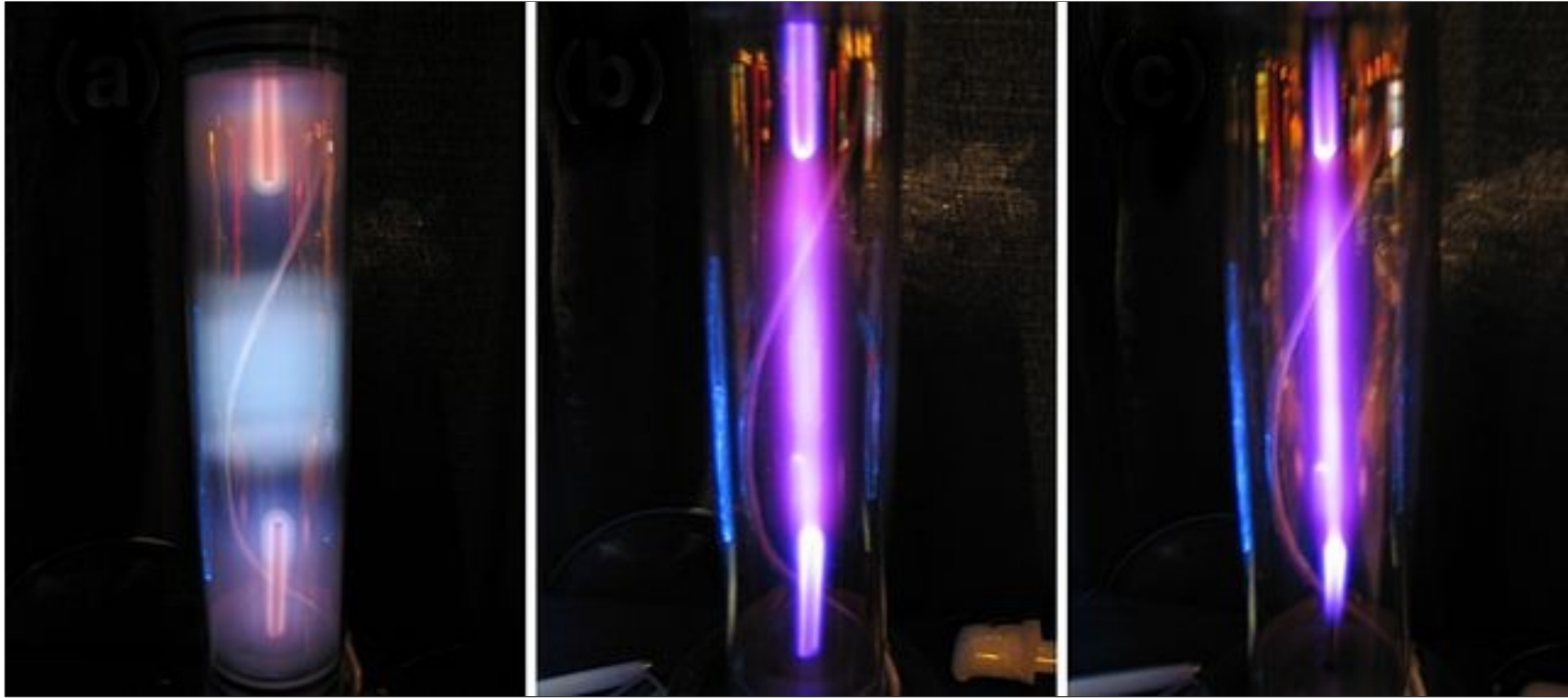
D-Kuru/Wikimedia Commons. "Afterglowing electrodes of an arc lamp." URL: [https://commons.wikimedia.org/wiki/File:Arc\\_lamp-afterglow\\_2\\_PNr%C2%B00038.jpg](https://commons.wikimedia.org/wiki/File:Arc_lamp-afterglow_2_PNr%C2%B00038.jpg), Accessed: August 28, 2013.

- Common in fluorescent lighting and polymer processing.
- Different temperatures for each species
- Often,  $T_{\text{neutrals}} \sim T_{\text{ions}} < T_{\text{electrons}}$



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



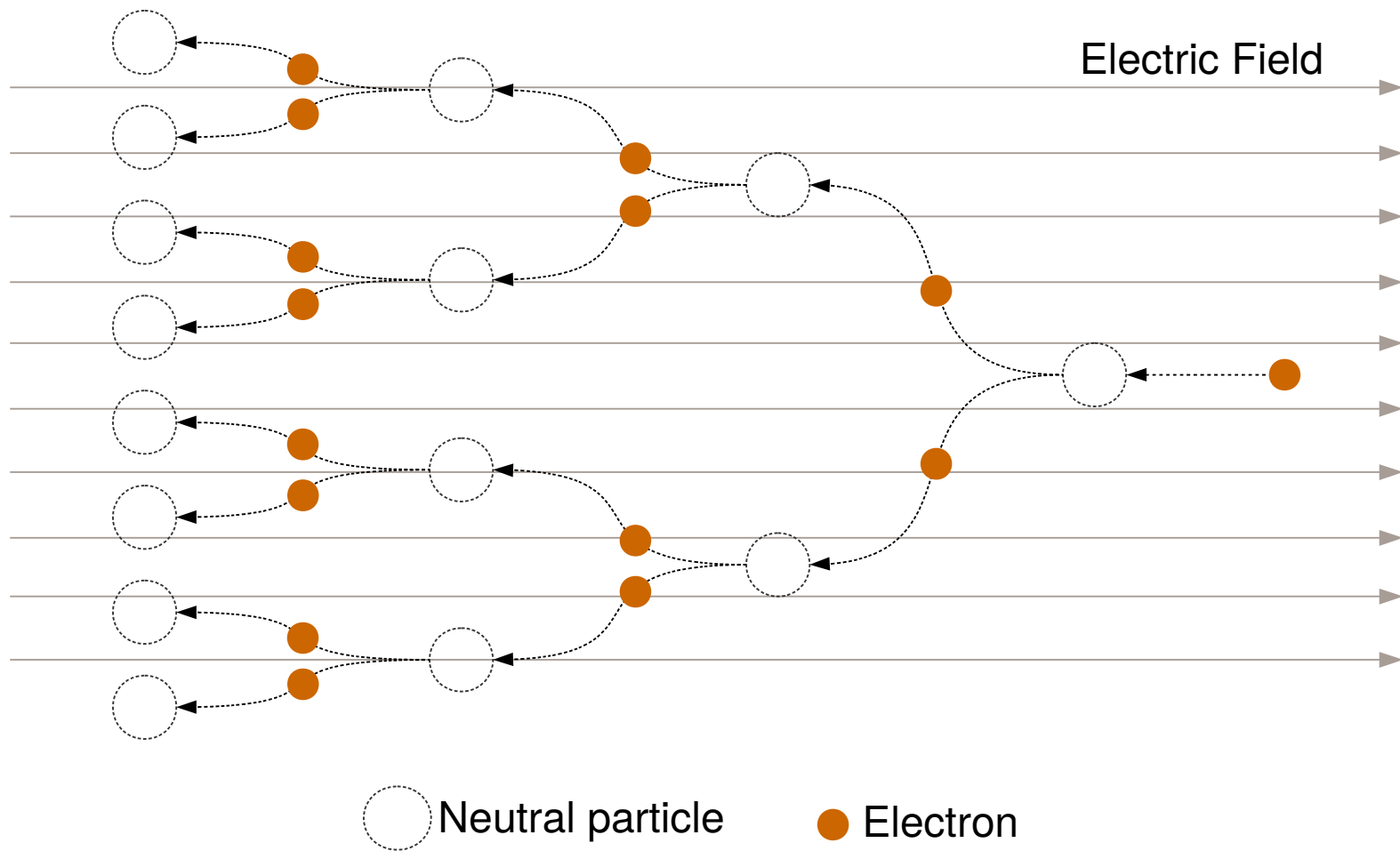


- Increase in pressure  $\rightarrow$  tendency to equilibrate
- Related to ionization instability
- Prevented by careful control of power input

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

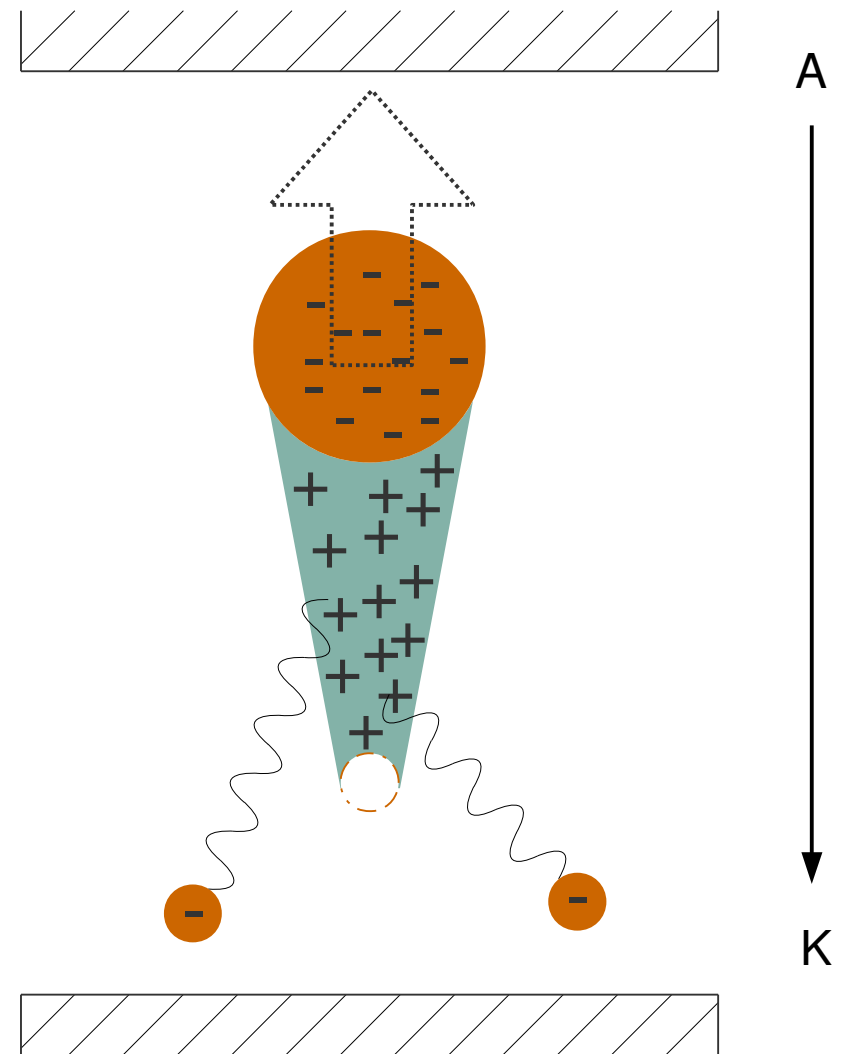
# Pulsed Plasmas and Outstanding Questions

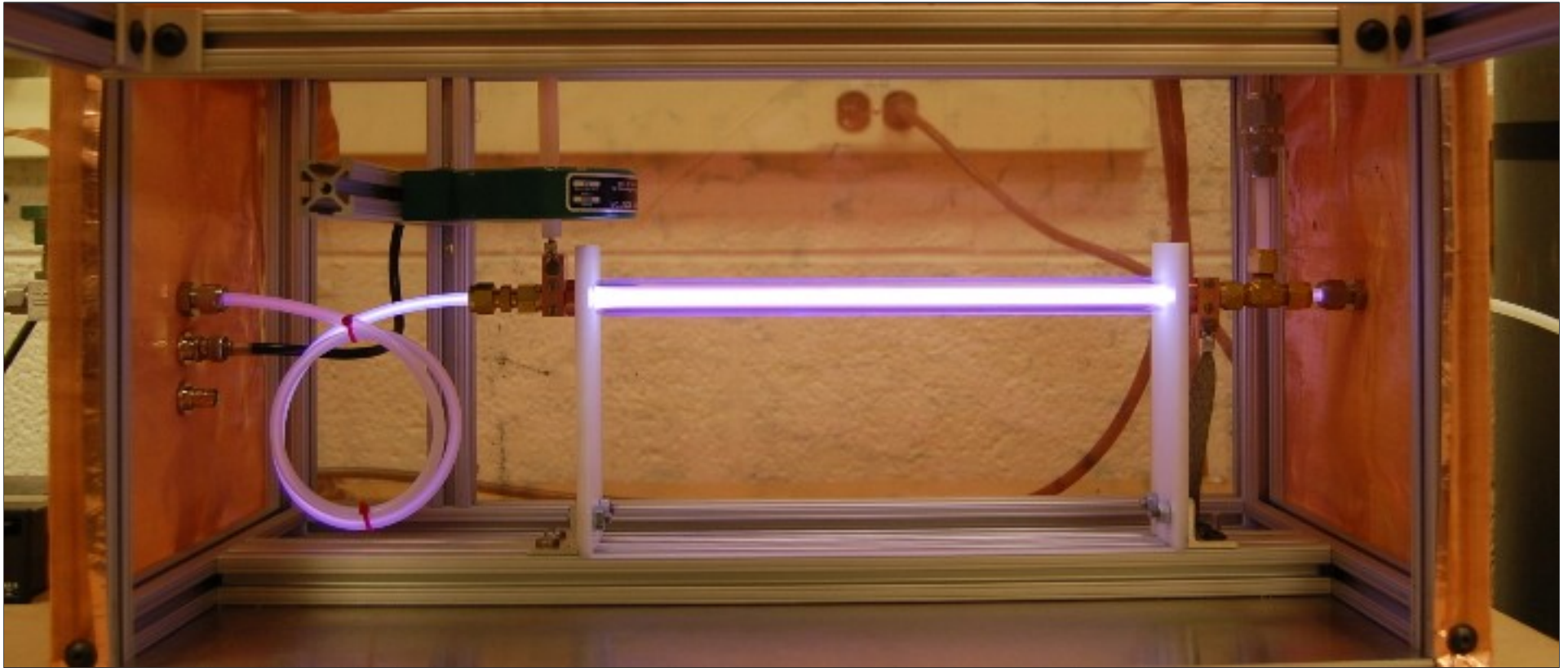




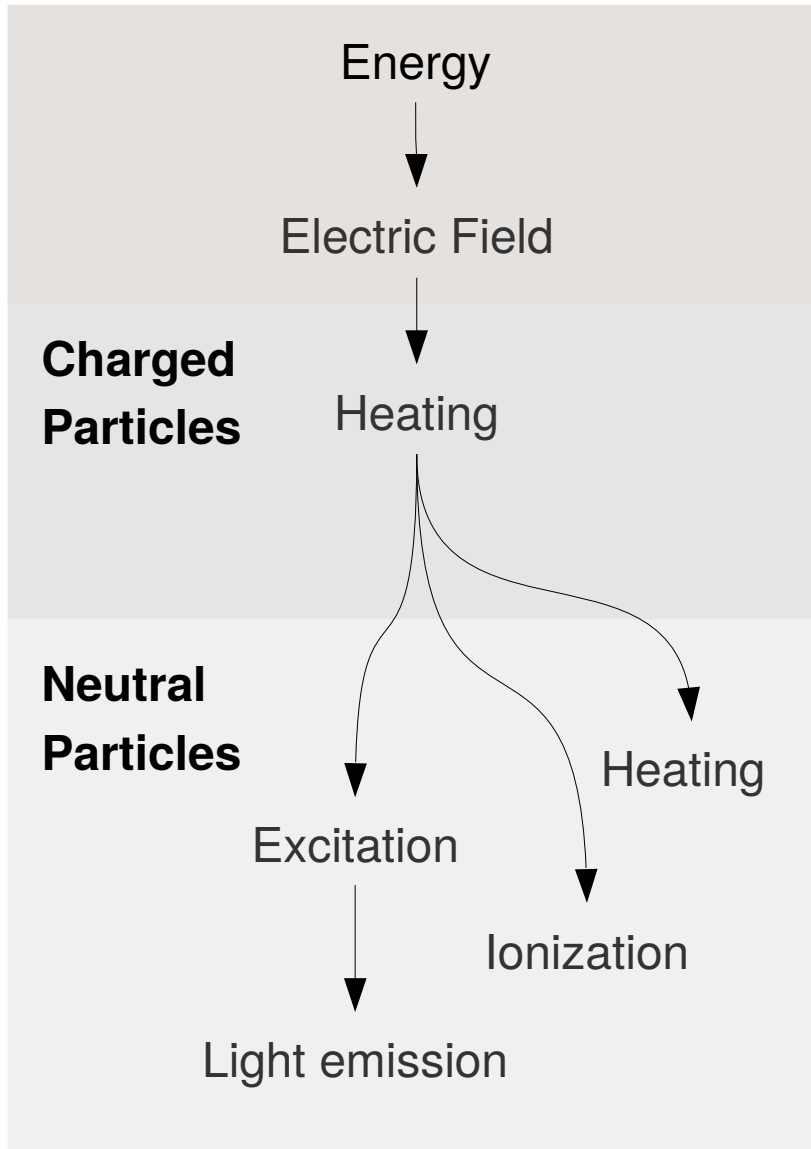
- Initial electron from cosmic rays, UV light, previous pulse, ...
- Sufficient electric field → exponential growth

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



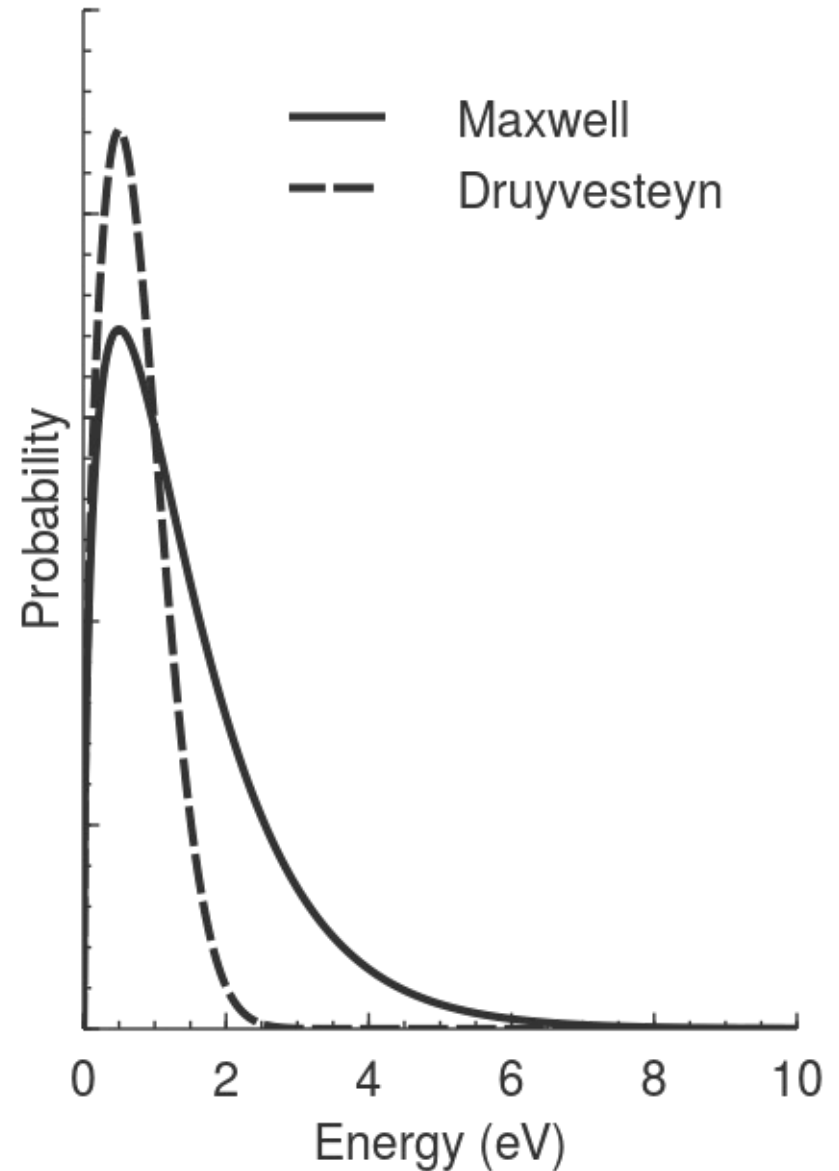


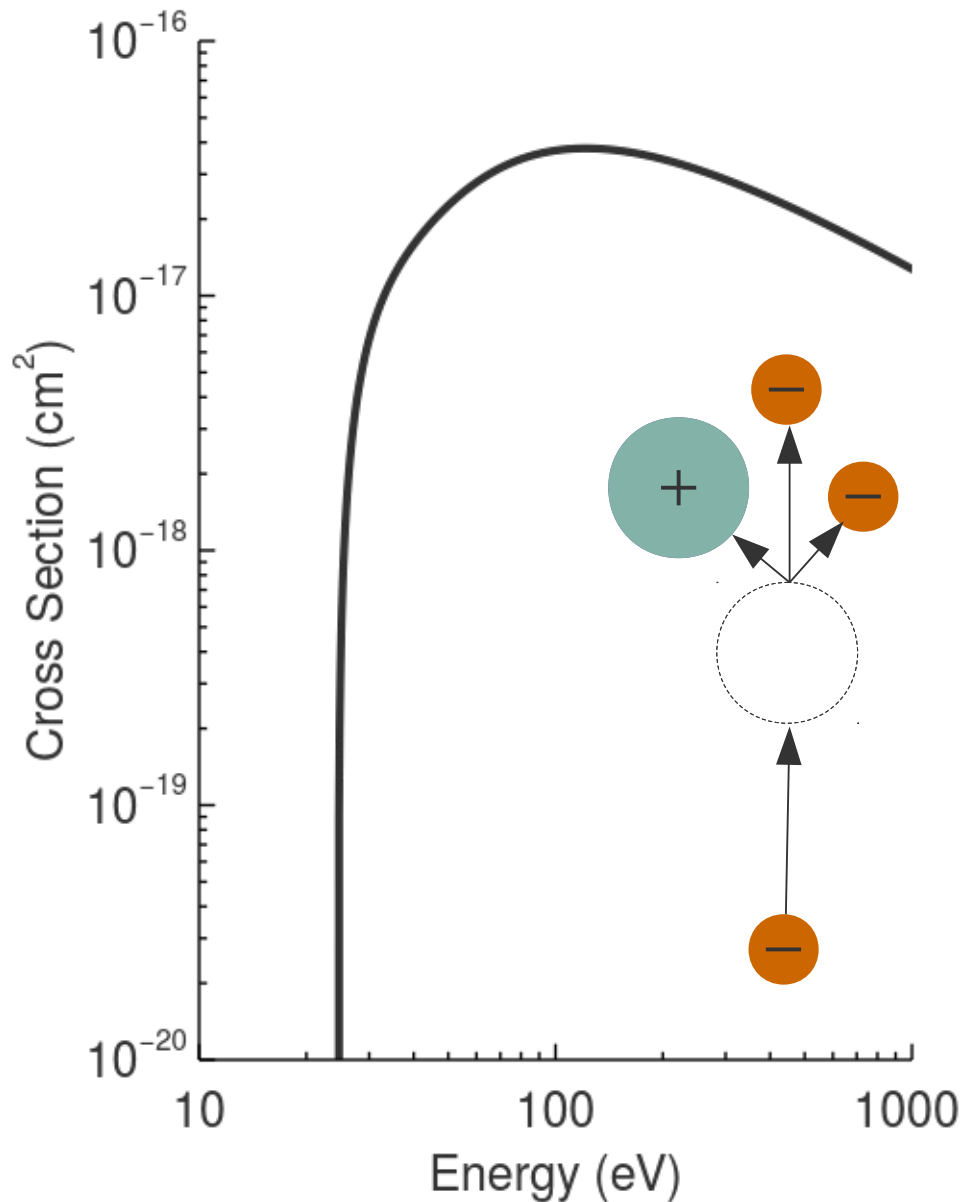
- Similar processes to streamer discharge
- Short pulse-widths: 0 – 100 ns
- High voltages: 1 – 100+ kV
- Moderate repetition rates: 1 – 100 kHz



- 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

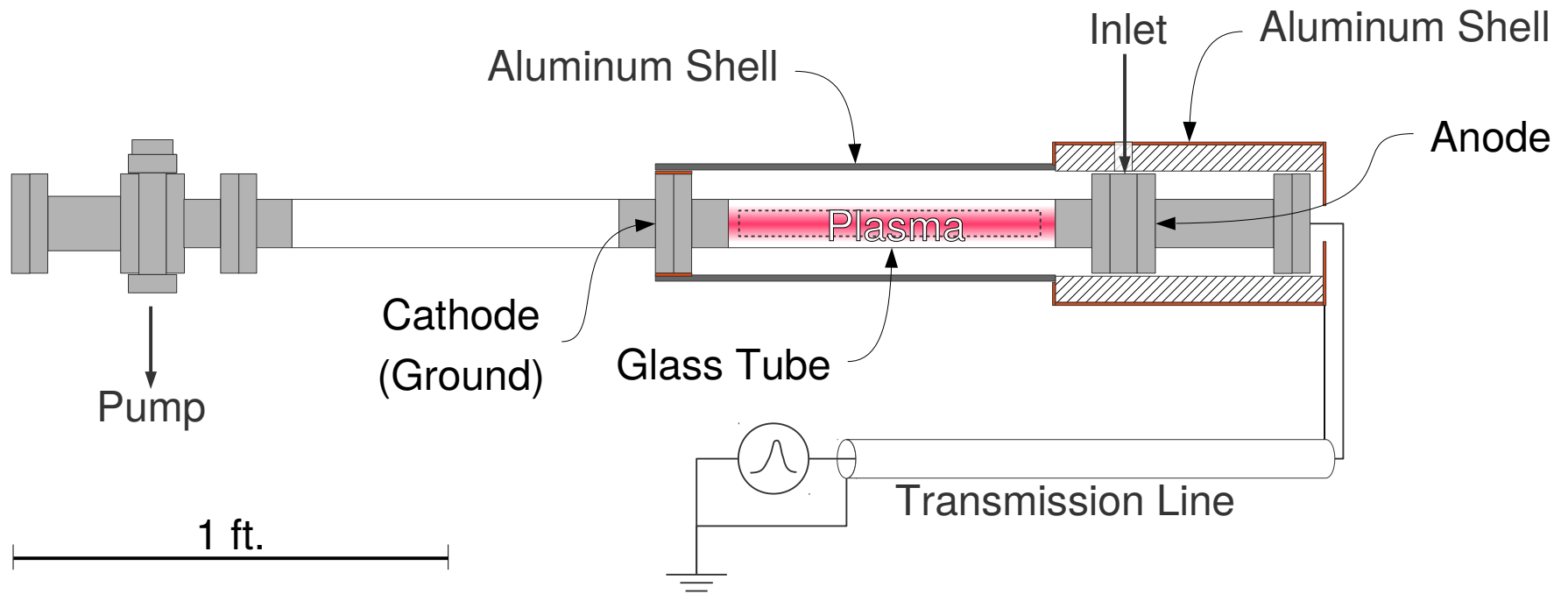




- Plasma products determined by cross sections
- Probability to form a product for a given electron energy

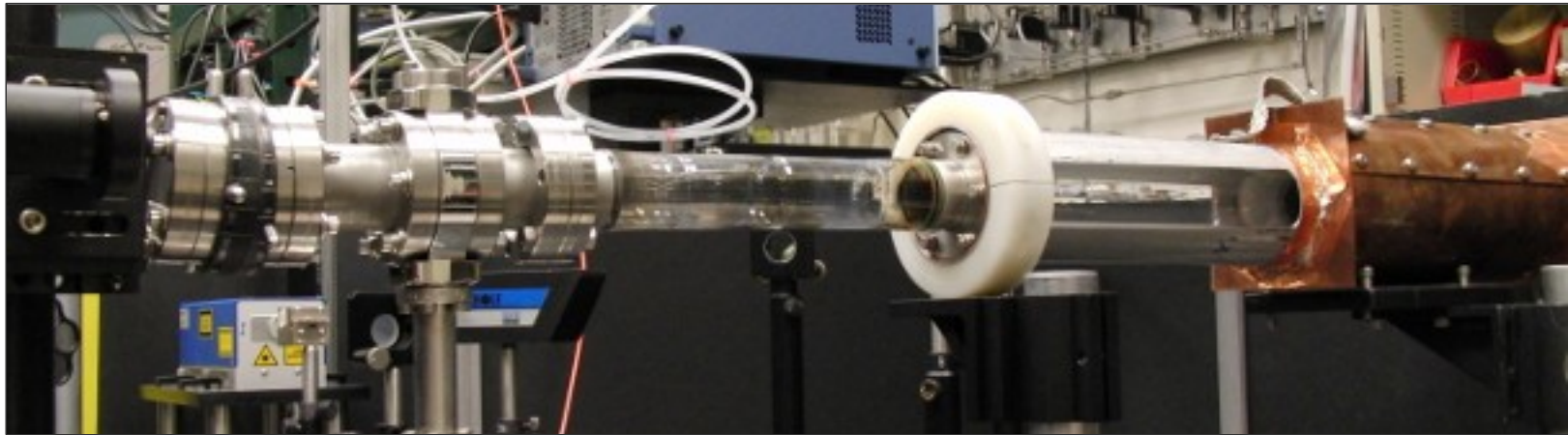


# Experimental Setup

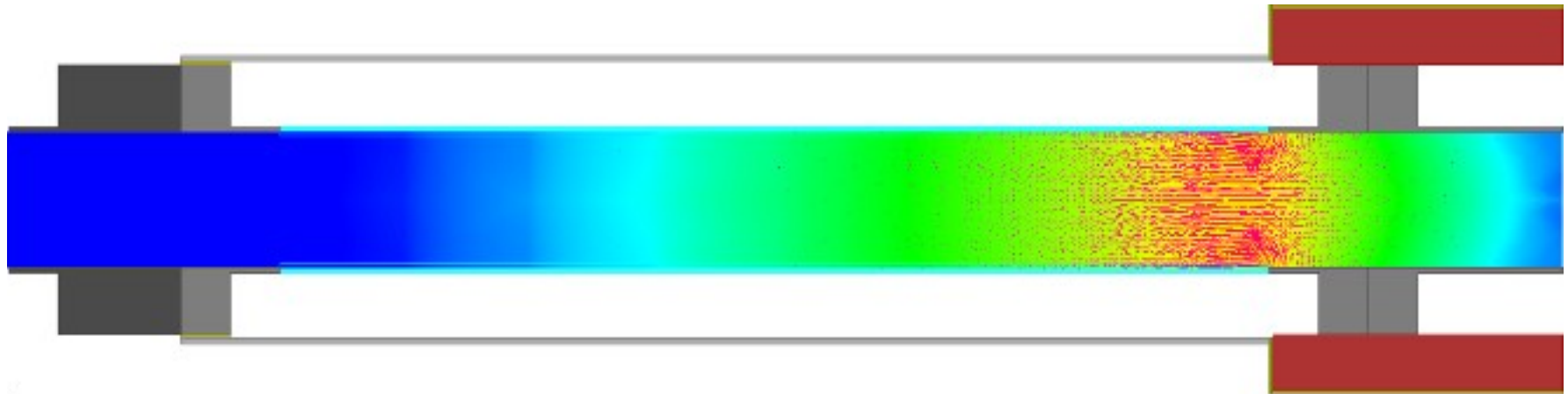


## Coaxial-type geometry

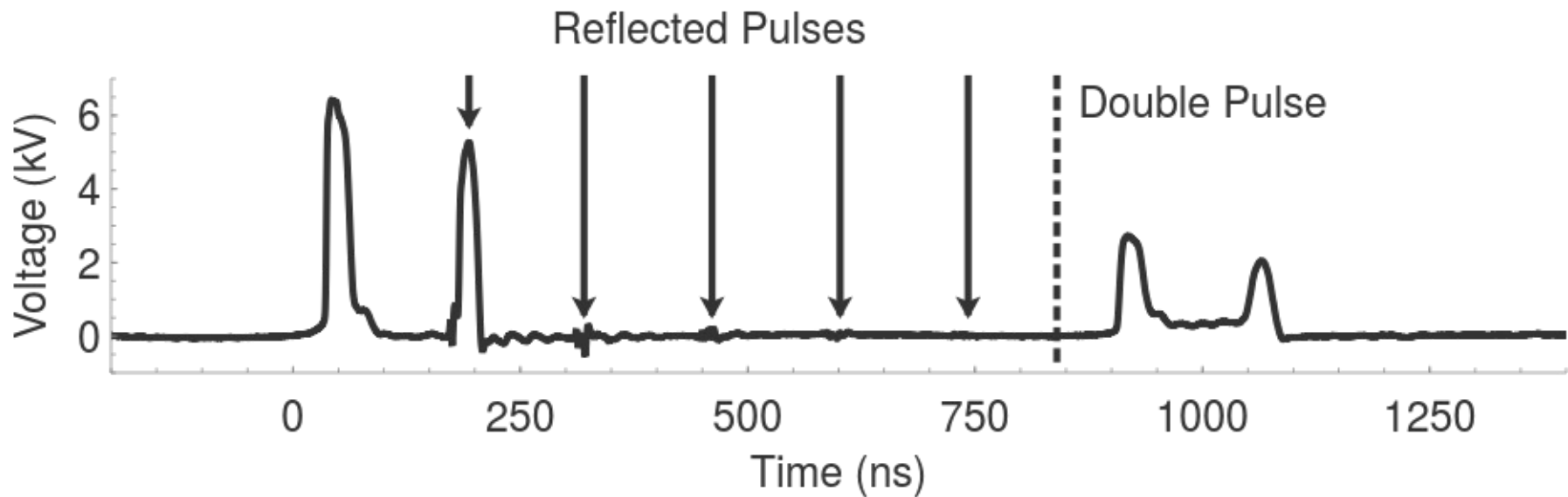
- Inner conductor: plasma
- Dielectric: borosilicate vacuum tube, air, and teflon
- Outer conductor: copper and aluminum shells



- 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



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

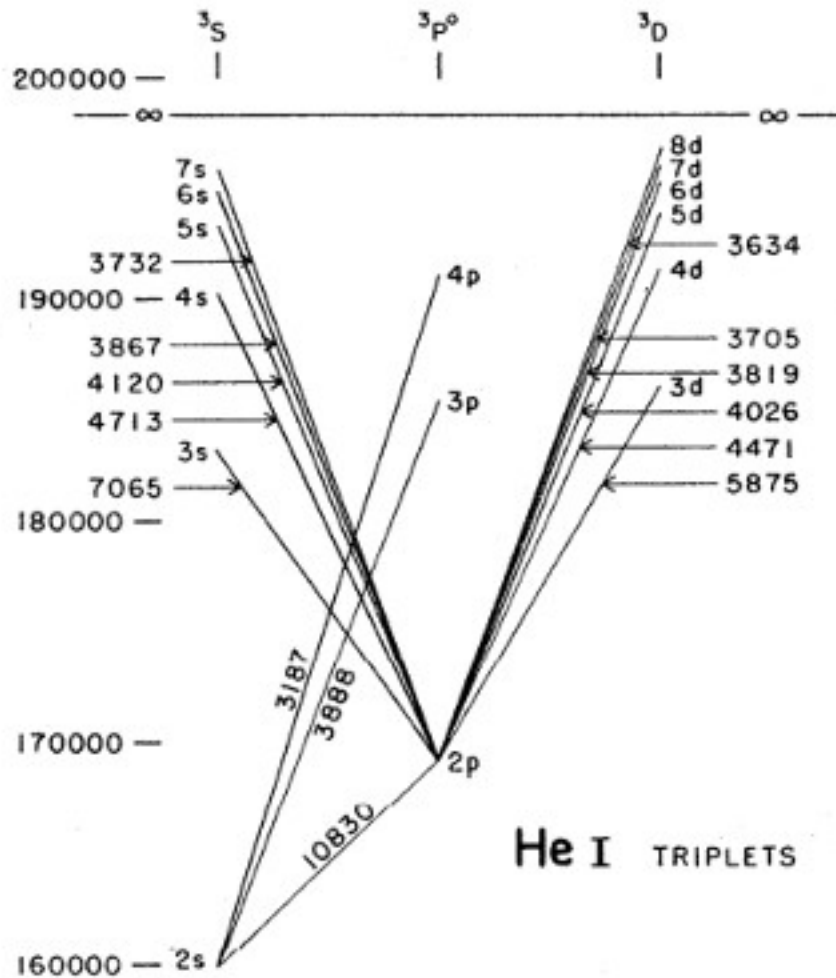


- Impedance mismatch at anode should cause doubling of voltage
- Long (13 m) transmission line used to isolate incident and reflected pulse

# Metastable Measurements

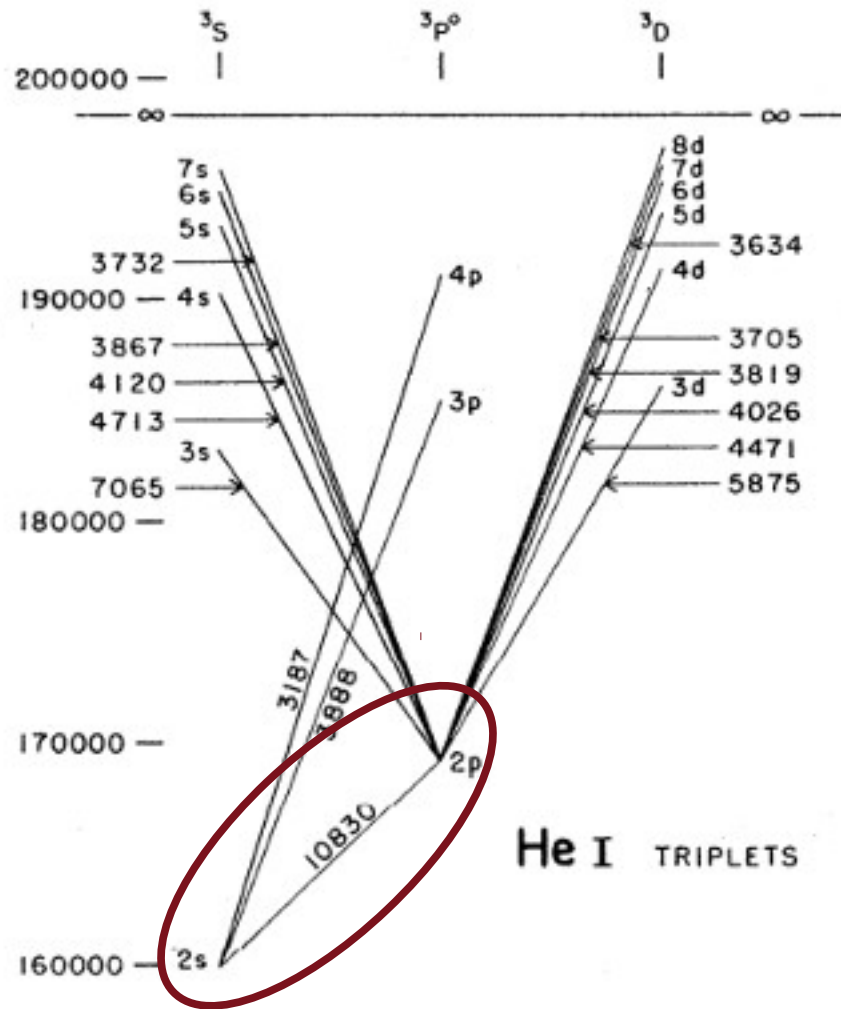


- ~~Physical probes~~
  - Perturb plasma
  - Not fast enough
  - Suggests an optical approach
- ~~Electrons~~
  - No detectable emissions
  - Insufficient density for laser scattering
- Atoms and atomic states



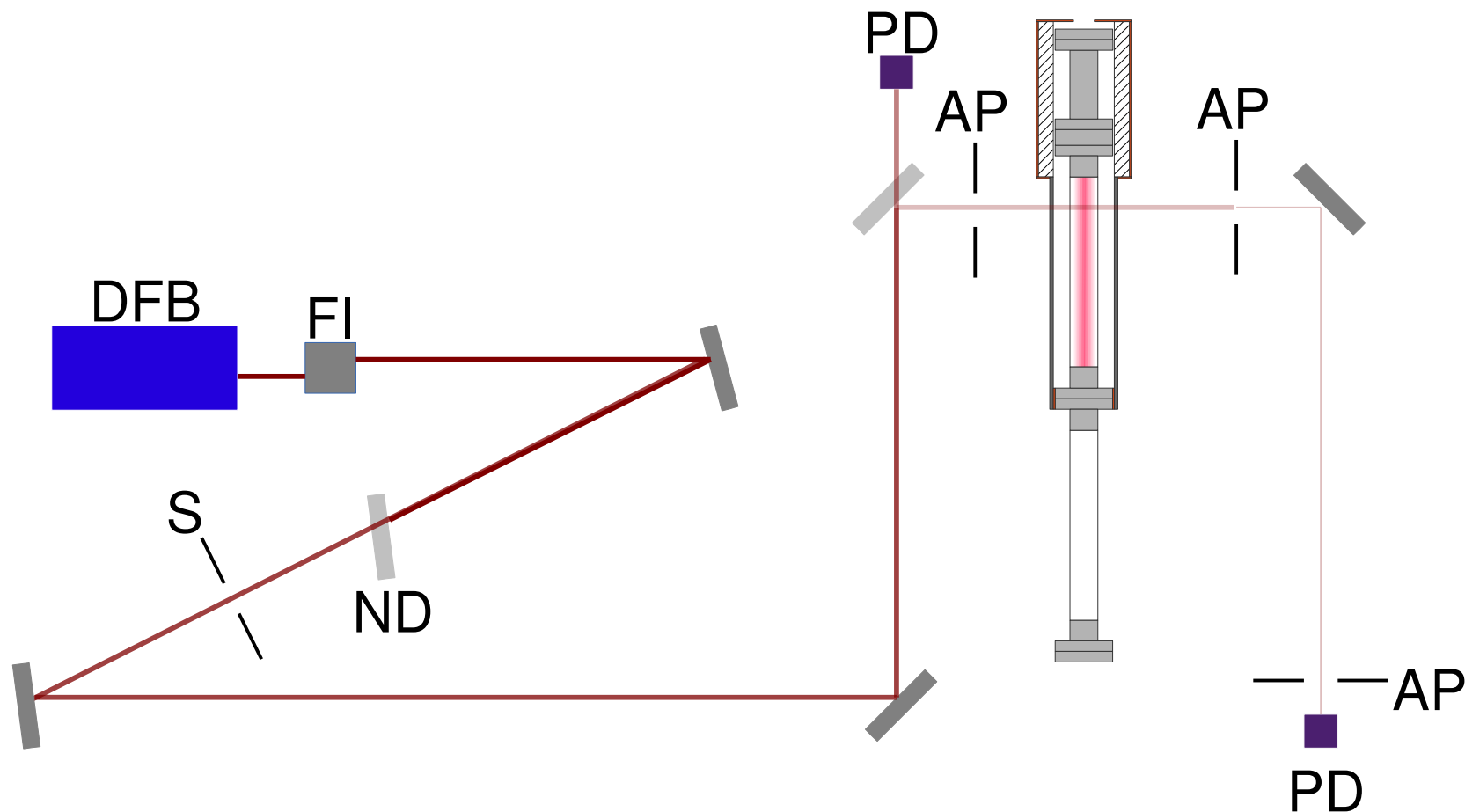
- Absorption of light
  - Photon energy must match energy difference between states
- Time resolution determined by detector
- Fraction of light absorbed proportional to density of lower state

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



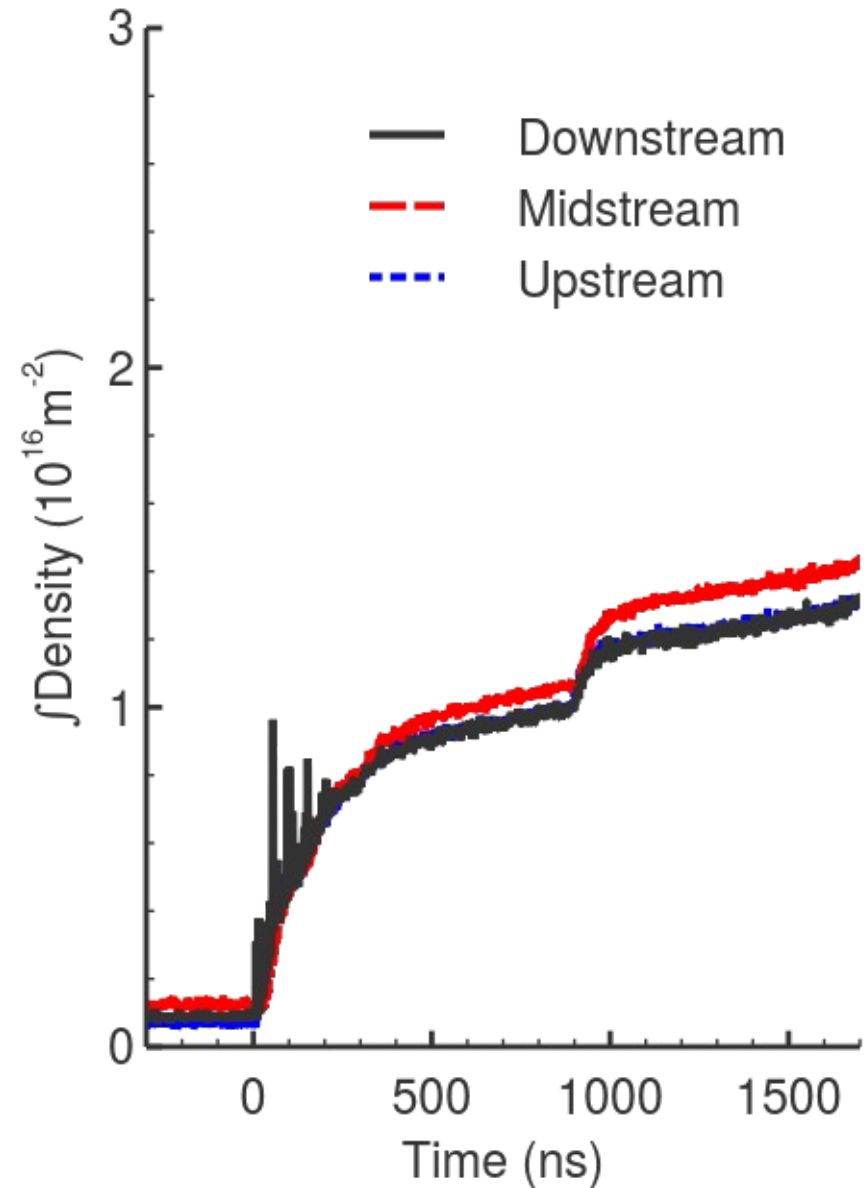
- Lowest-energy excited state
- Cannot transition to unexcited state
- Reservoir for energy
- Can cause excitation and ionization long after pulse

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

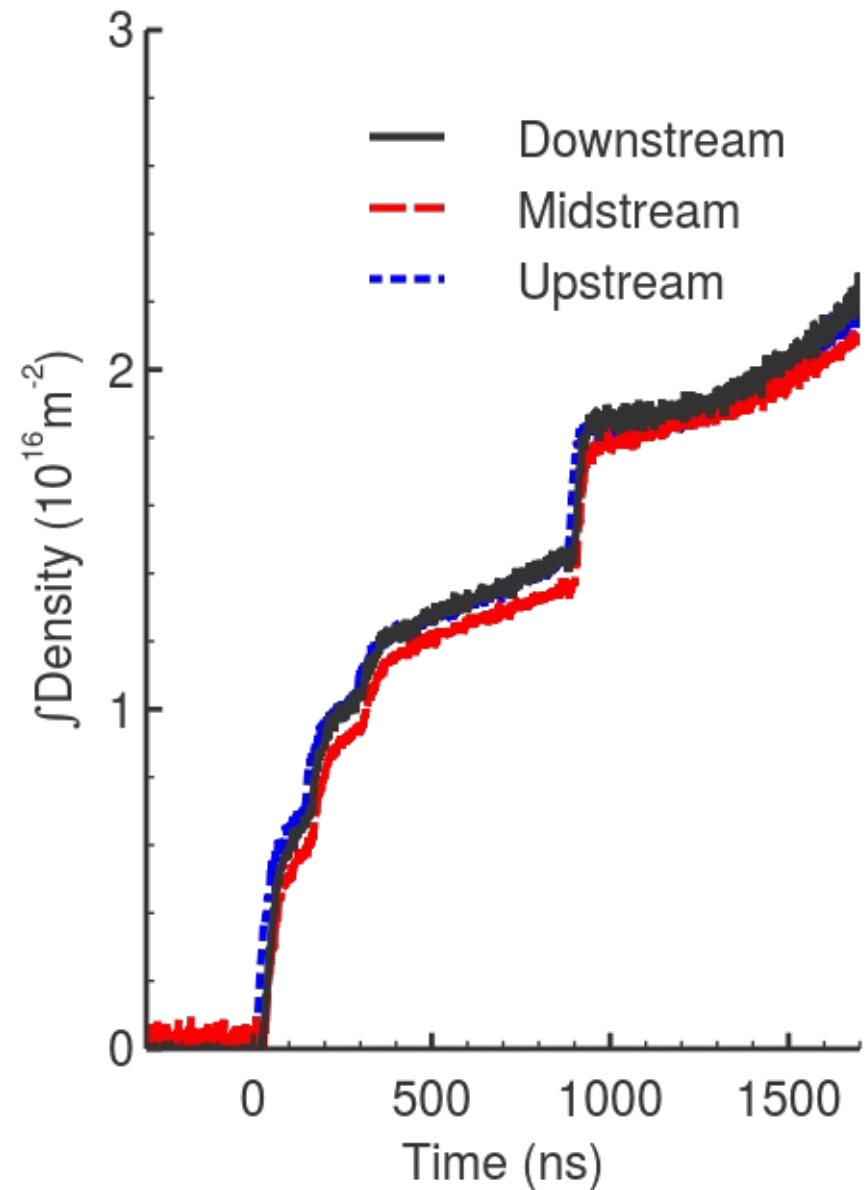


- 1083 nm laser diode,  $2^3S \rightarrow 2^3P_0$  transition
- Pressures: 0.3, 0.5, 1.0, 2.0, 3.0, 4.0, 8.0, and 16.0 Torr
- Locations: 5.1 cm (Upstream), 12.7 cm (Midstream), and 20.3 cm (Downstream)
- Minimum detectable density:  $9 \times 10^{15} \text{ m}^{-3}$

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

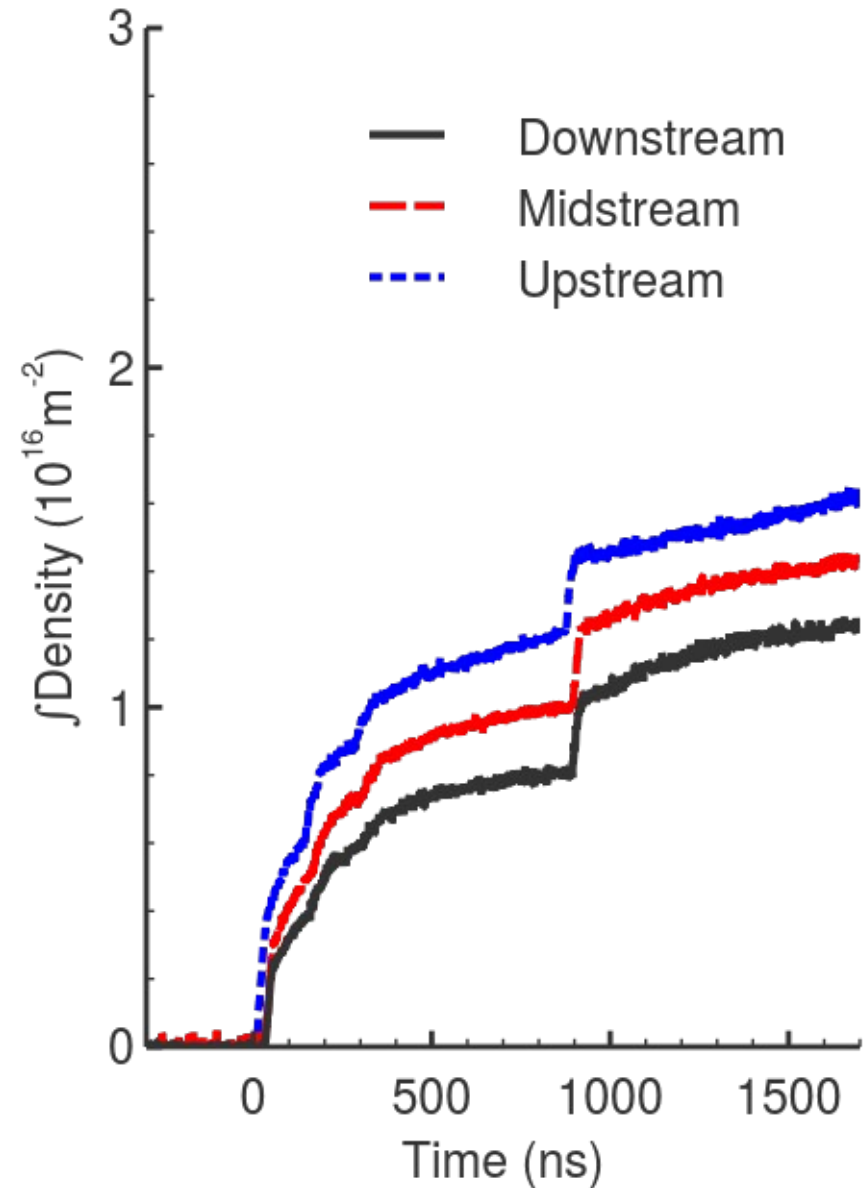


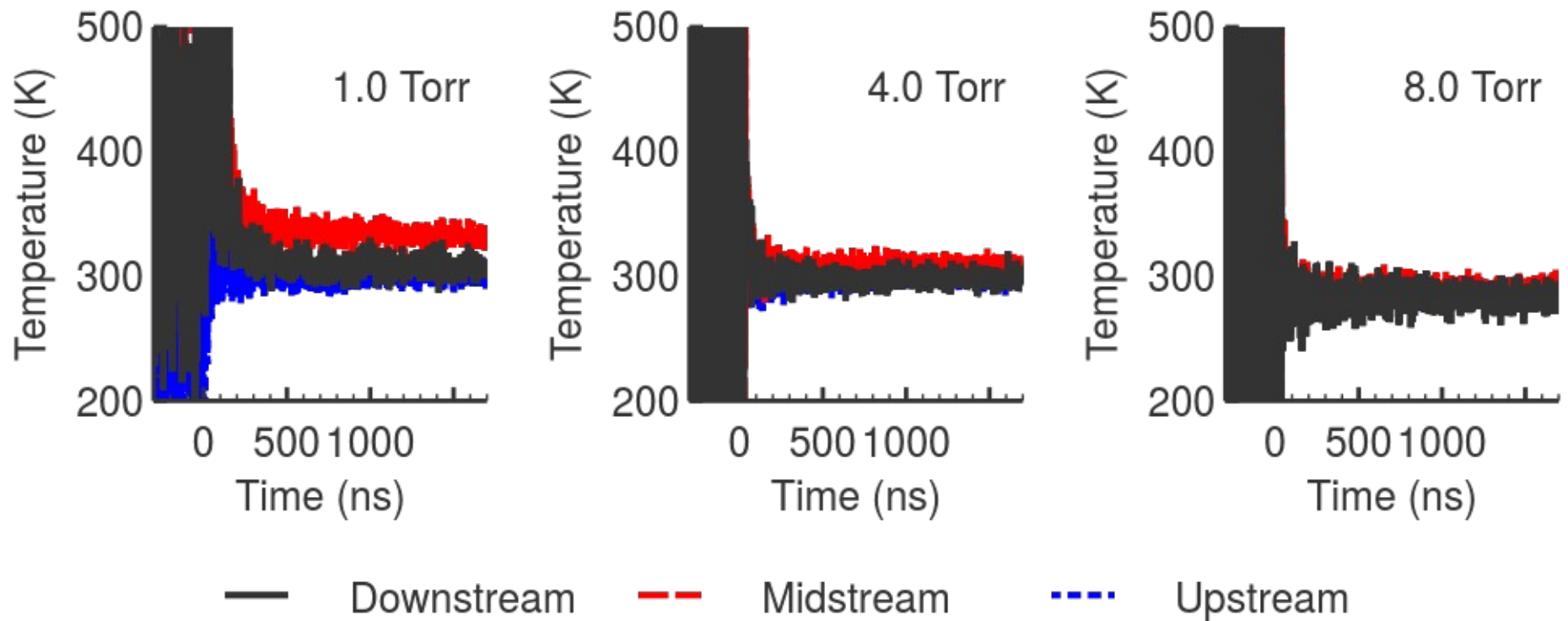
- Reduced pre-pulse density
- Increase in final density
- Reflections cause boost in metastable generation rate



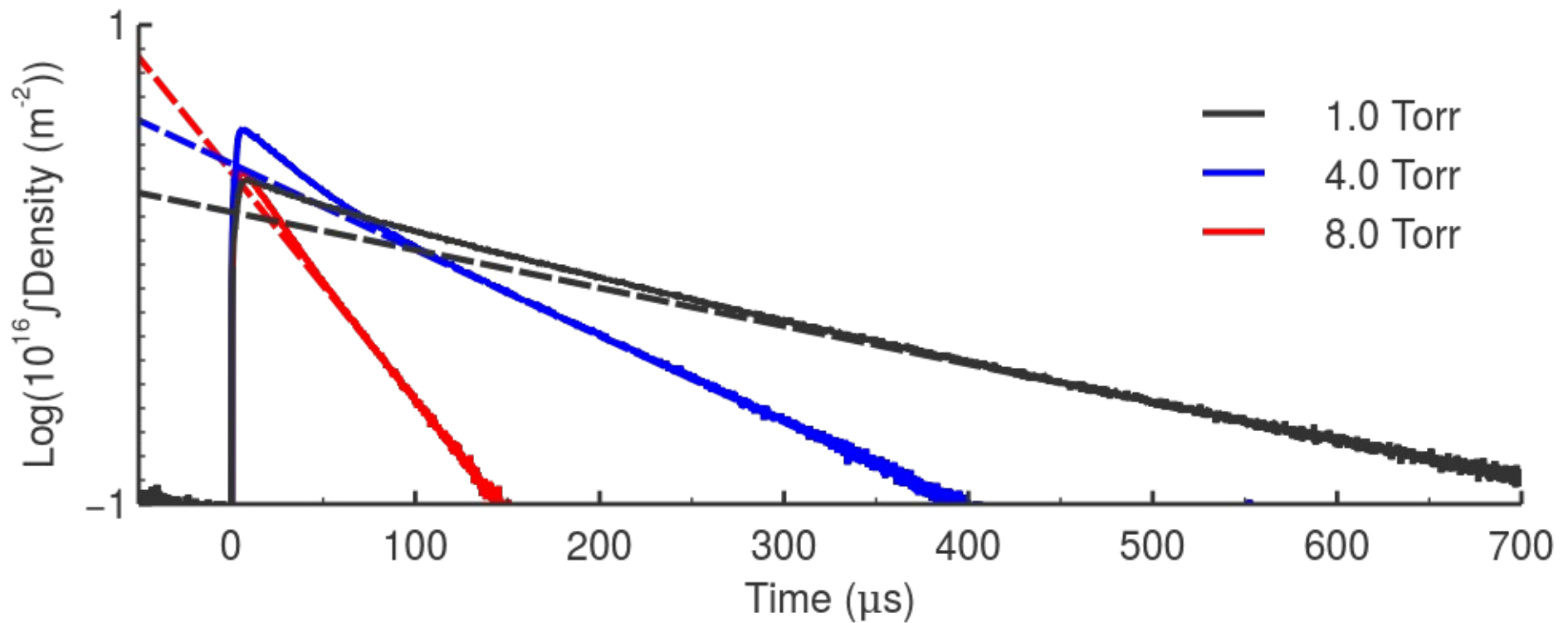


- No metastables before pulse
- Overall decline in metastable density
- Reflection effects still observable
- Plasma attenuation with distance from anode

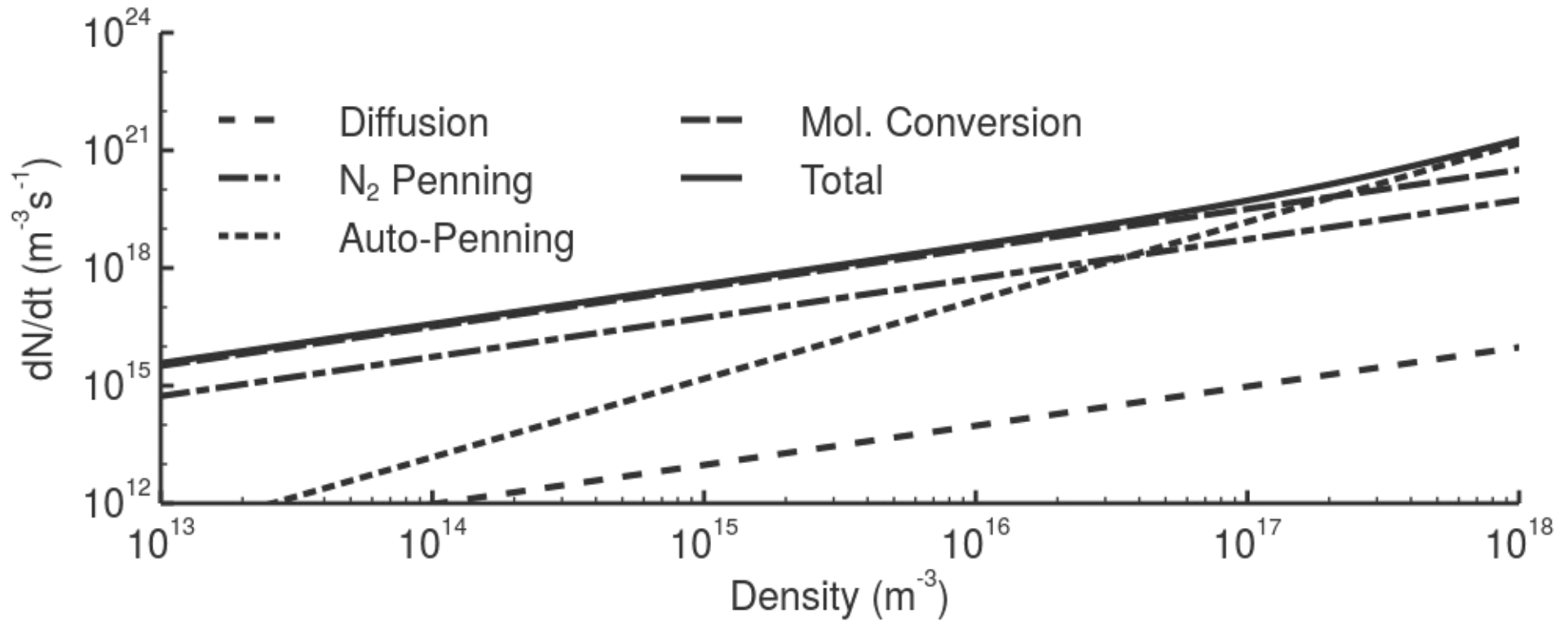




- Added benefit of laser-absorption spectroscopy
- Confirms minimal gas heating
- No significant difference from 300 K for all conditions



- Long-duration measurements reveal loss mechanisms
- Deviation from straight line  $\rightarrow$  non-exponential process

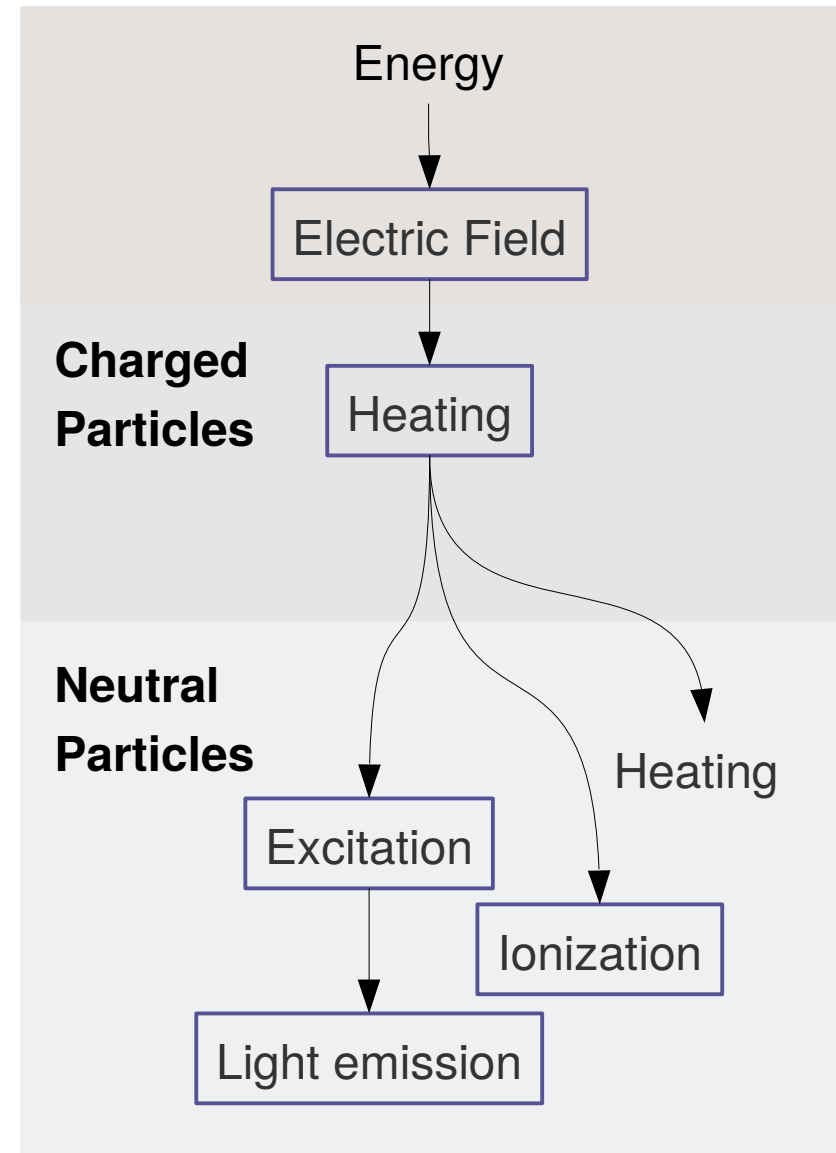


- Initially dominated by non-exponential processes
  - Superelastic electron collisions
  - Penning ionization between metastables
- Molecular conversion dominates at lower metastable densities
- Pre-pulse metastables only detectable for 4.0 Torr and below

- 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

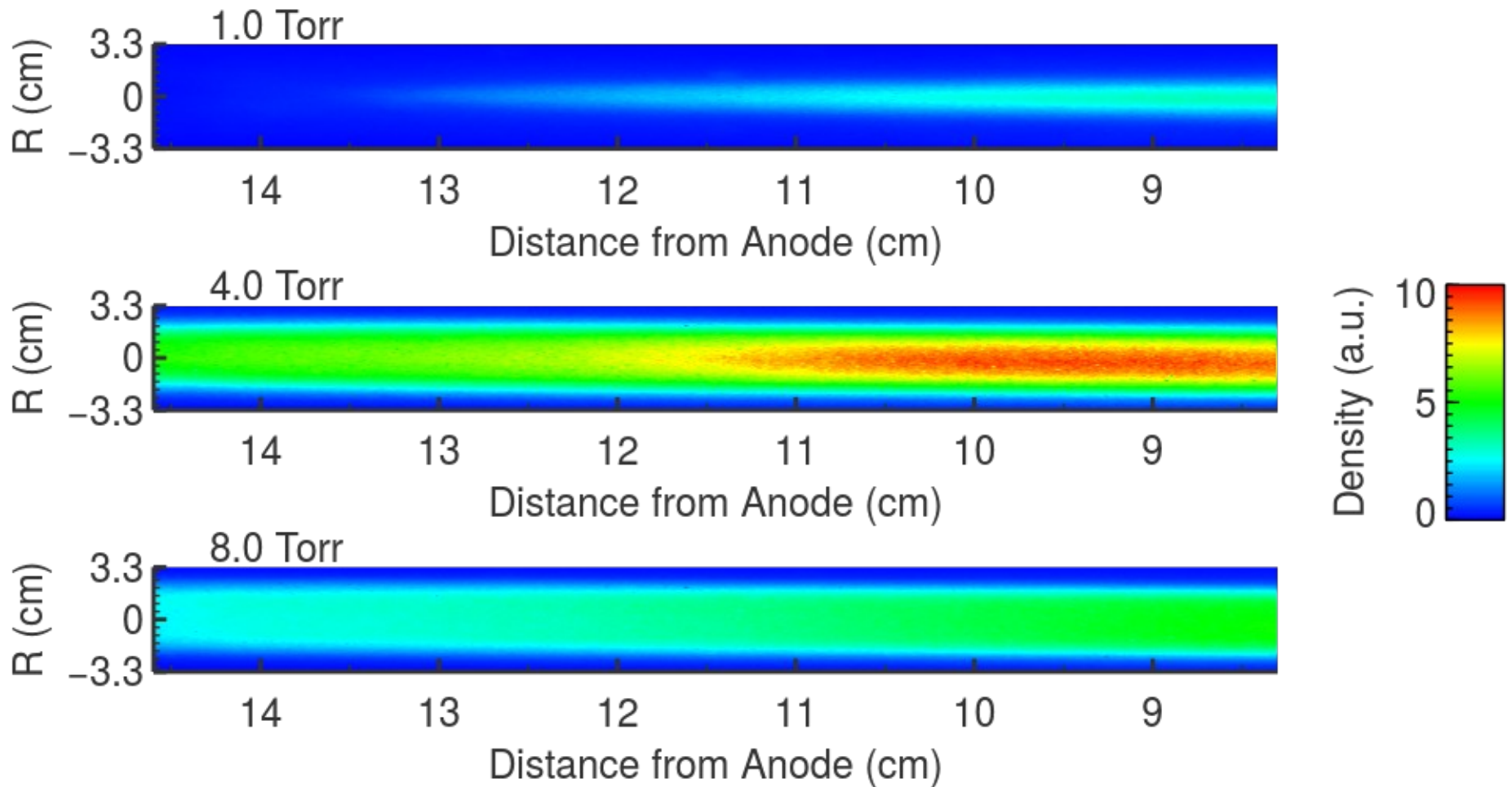
# Global Model

- 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)
- Assume Maxwell-Boltzmann EEDF based on comparisons with particle-in-cell and two-term expansion results.



- Variation with pressure, uncertain cause
- Assume uniform radial density distribution, results easily modified for different profiles

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

$$\frac{dN_i}{dt} = n_e \left[ \sum_{j \neq i} N_j K_{j,i}^e(T_e) - N_i \sum_{j \neq i} K_{i,j}^e(T_e) \right] \quad \text{Electron (de)excitation}$$

$$+ \left[ \sum_{j > i} N_j K_{j,i}^o - N_i \sum_{j < i} K_{i,j}^o \right] \quad \text{Radiative transitions}$$

$$+ N_g \left[ \sum_{j \neq i} N_j K_{j,i}^a - N_i \sum_{j \neq i} K_{i,j}^a \right] \quad \text{Atomic excitation transfer}$$

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

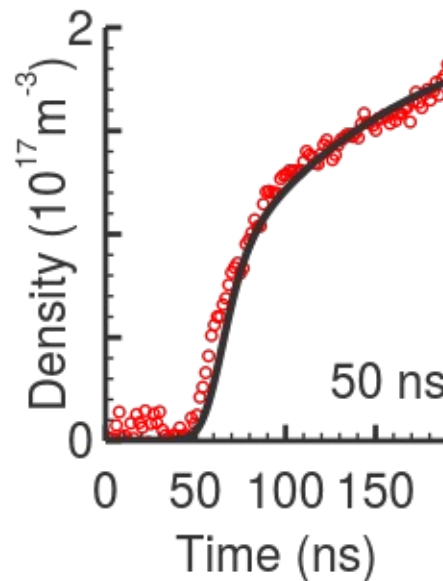
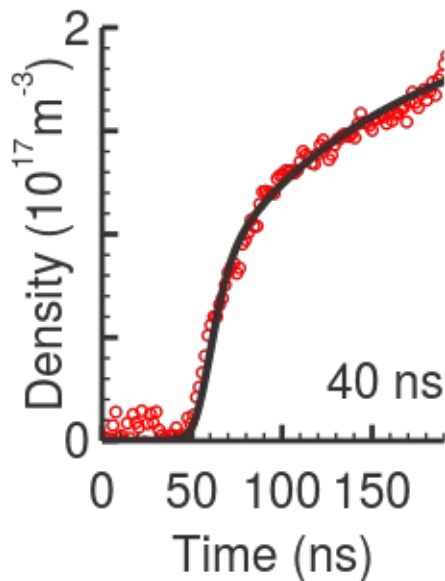
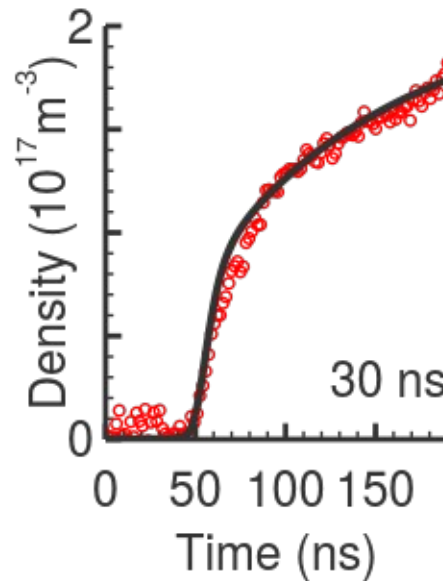
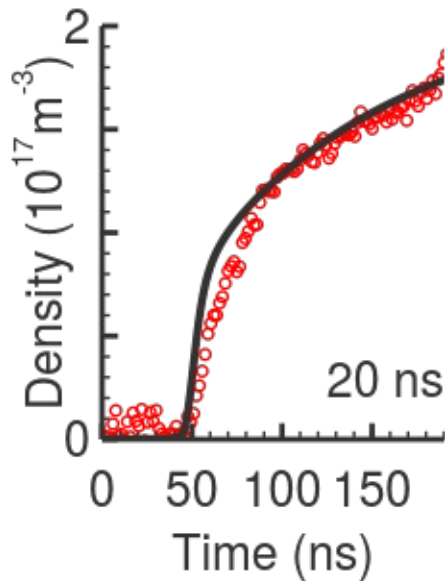
Electric field  
heating

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

Elastic  
collisions

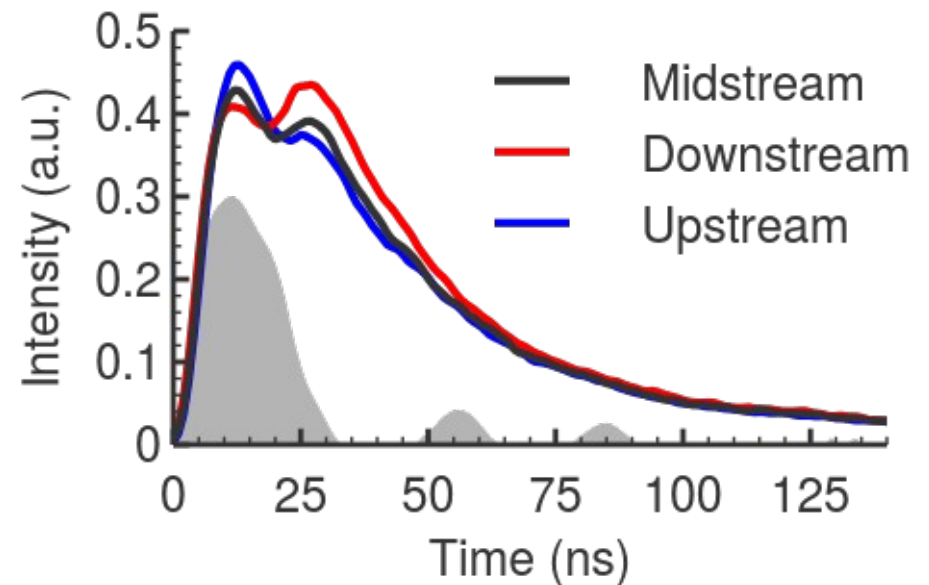
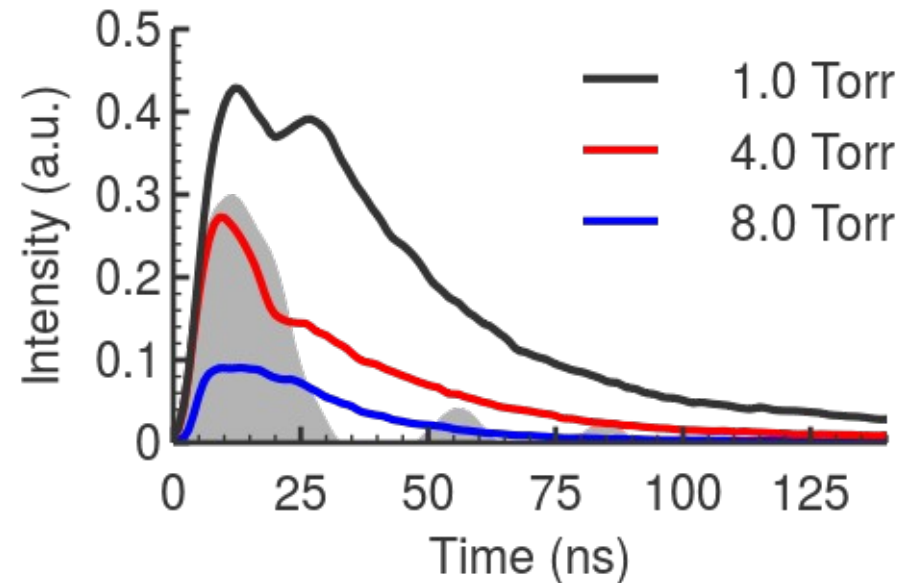
$$- n_e \sum_i \sum_{j \neq i} K_{ij}^e N_i \Delta \epsilon_{ij}$$

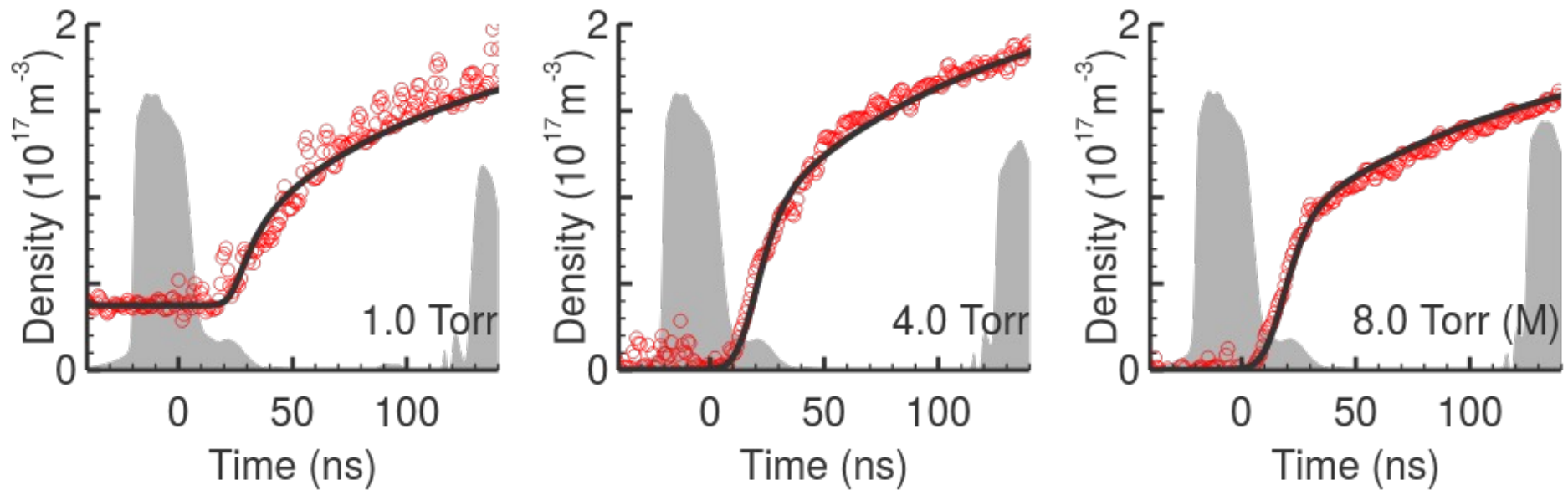
Inelastic  
collisions



- 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?

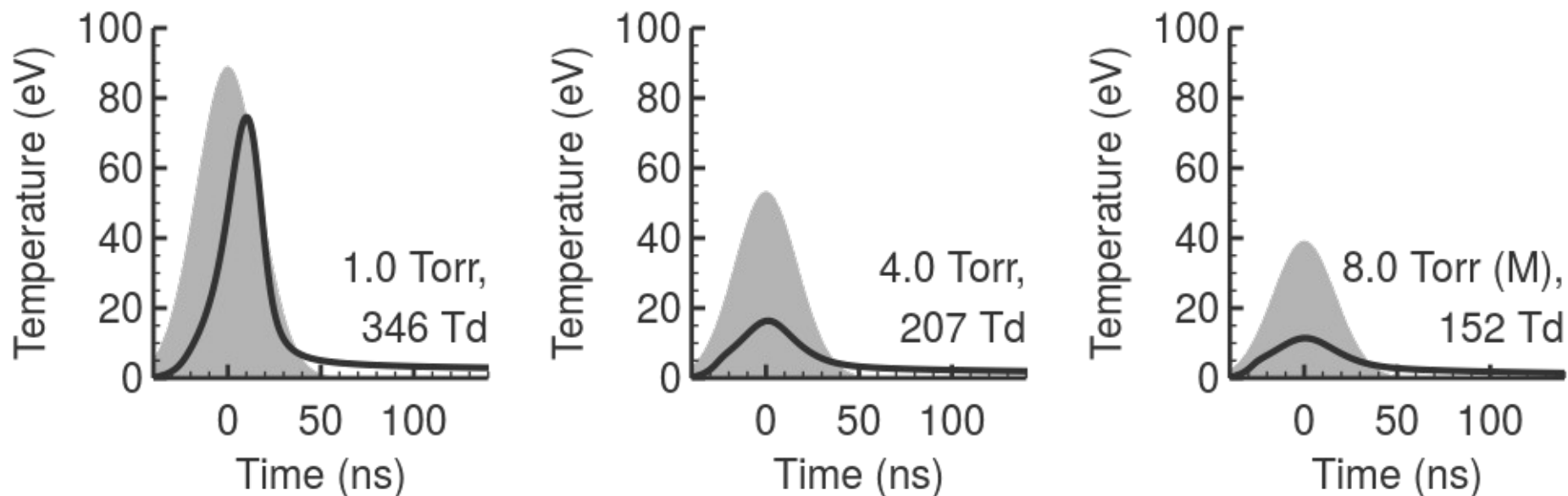
- Optical emissions showed evidence of return stroke
- Double peak at several pressures
- Second peak most intense closest to ground electrode
- Excitation period effectively longer than 25 ns





- 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?



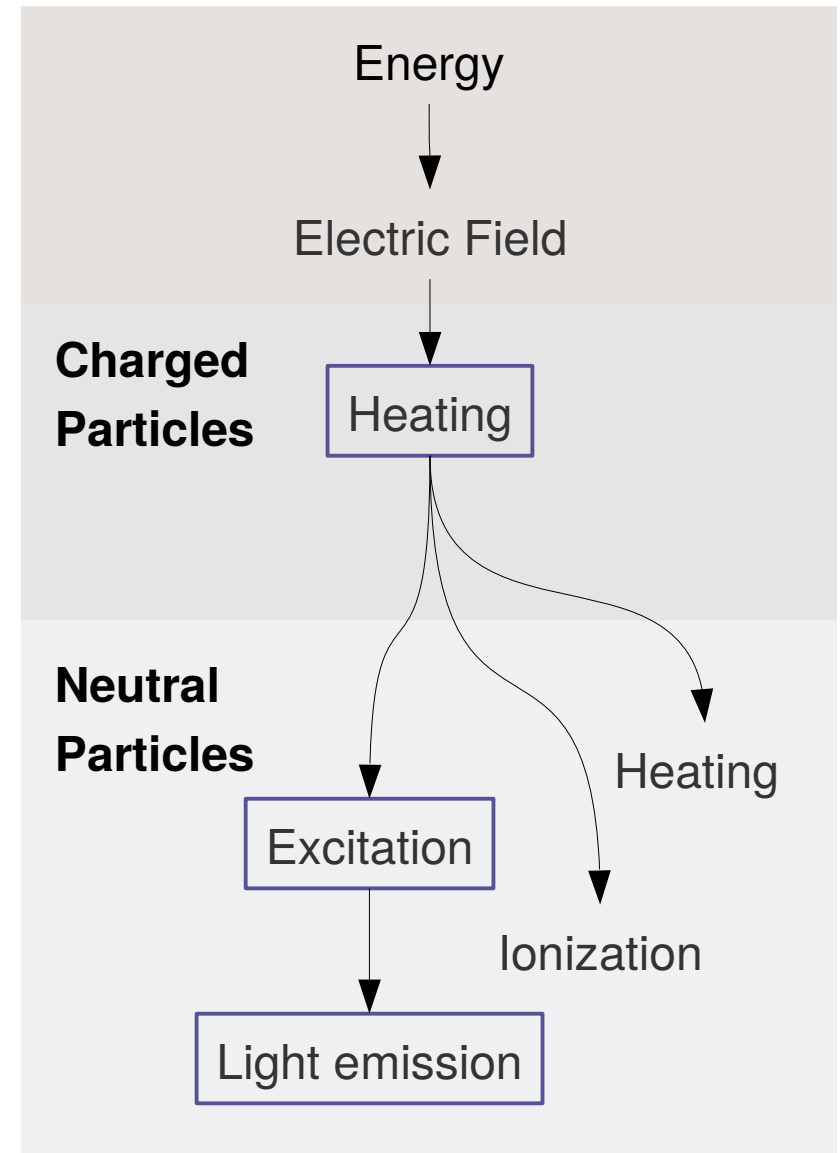


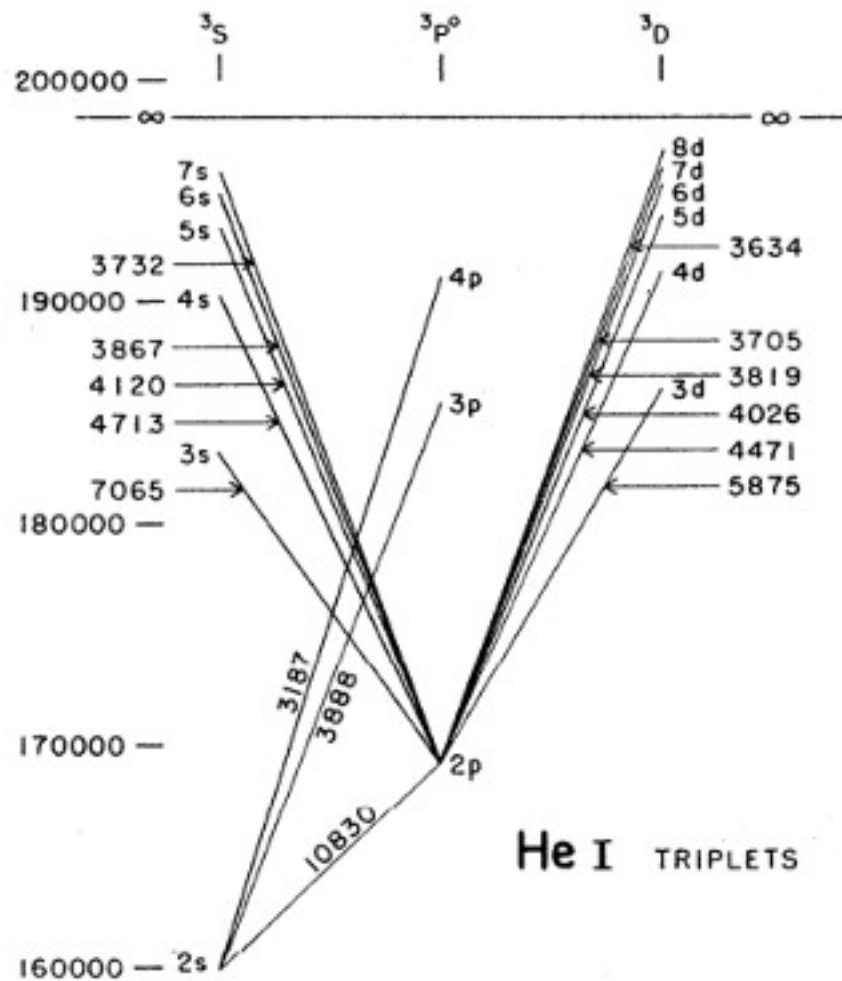
- Peak fields consistent with other studies
- 1.0 Torr fields well into range where two-term expansion solutions performed better
- 1.0 Torr temperatures are not reasonable

- 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

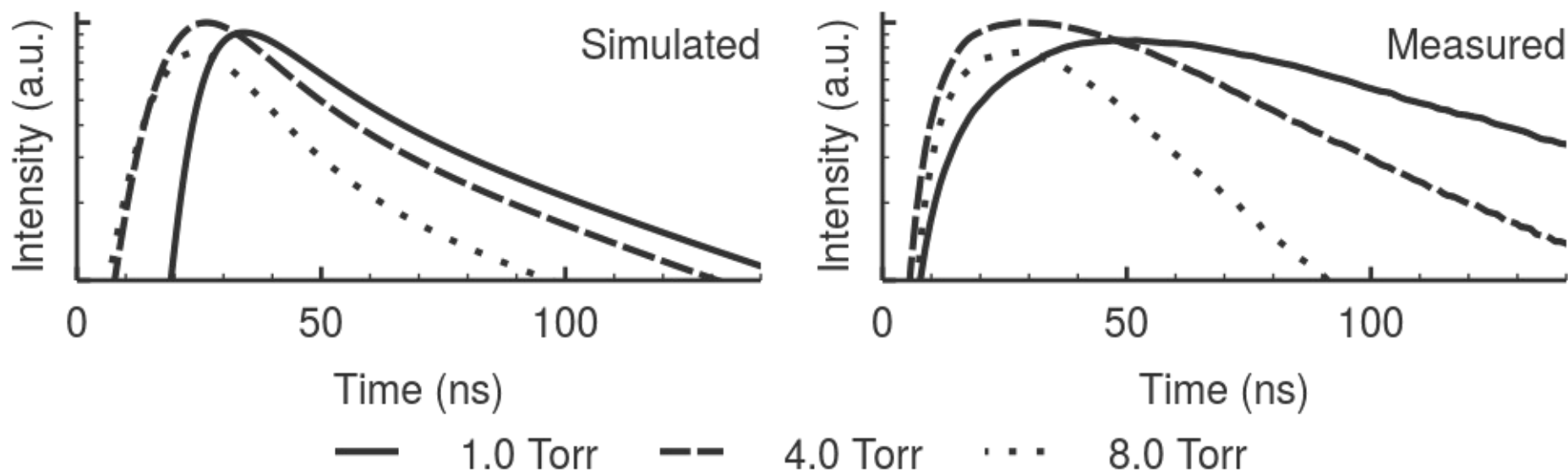
# Emission Measurements

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





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



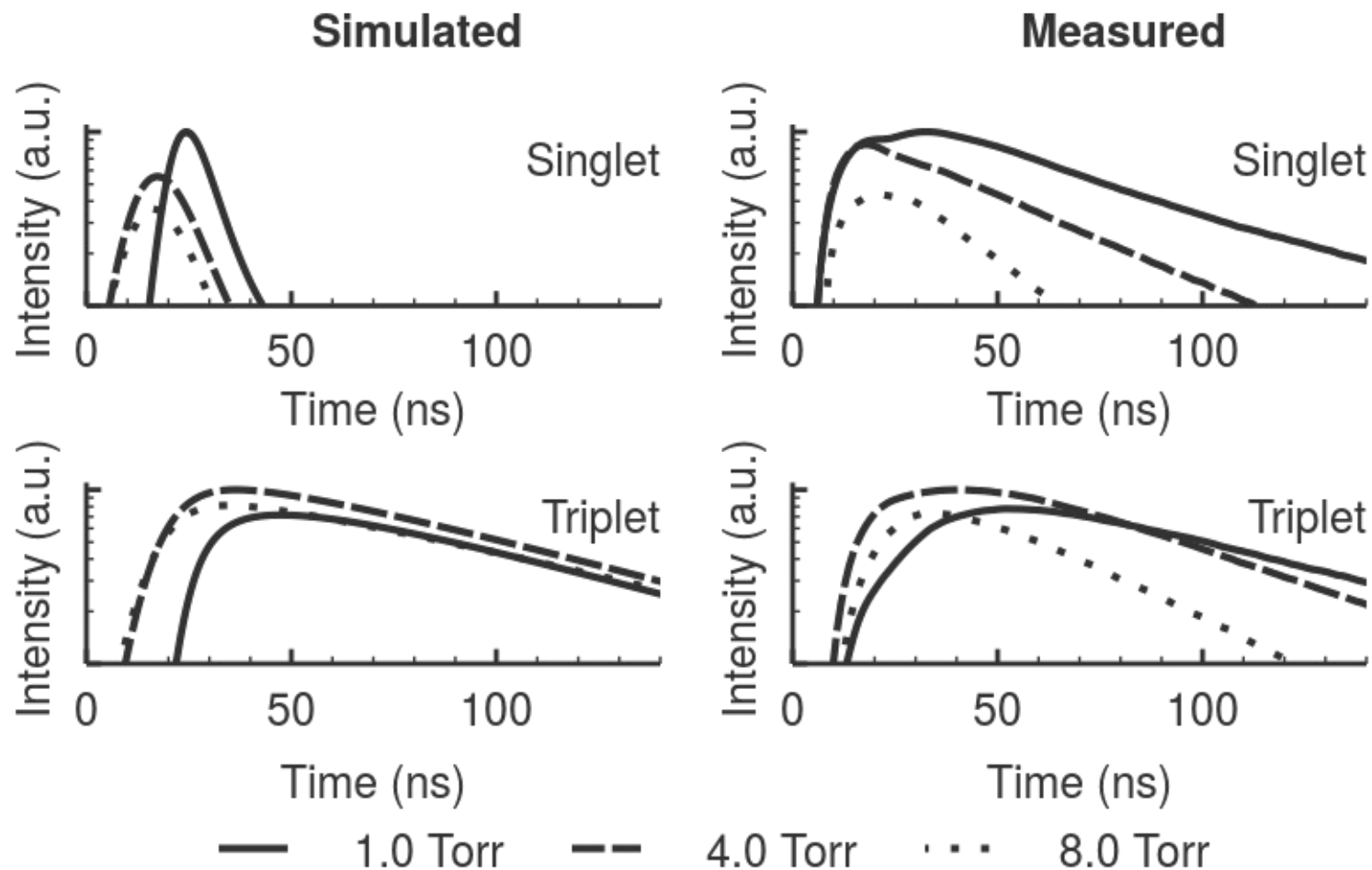
- Relative peak heights and timing similar, distinctly different in shape
- Measurements suggest pressure-dependent de-excitation process (Penning ionization of impurities?)
- Long emission lifetime suggest an extended excitation process
  - Beam-like electrons
  - Persistent electric field

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

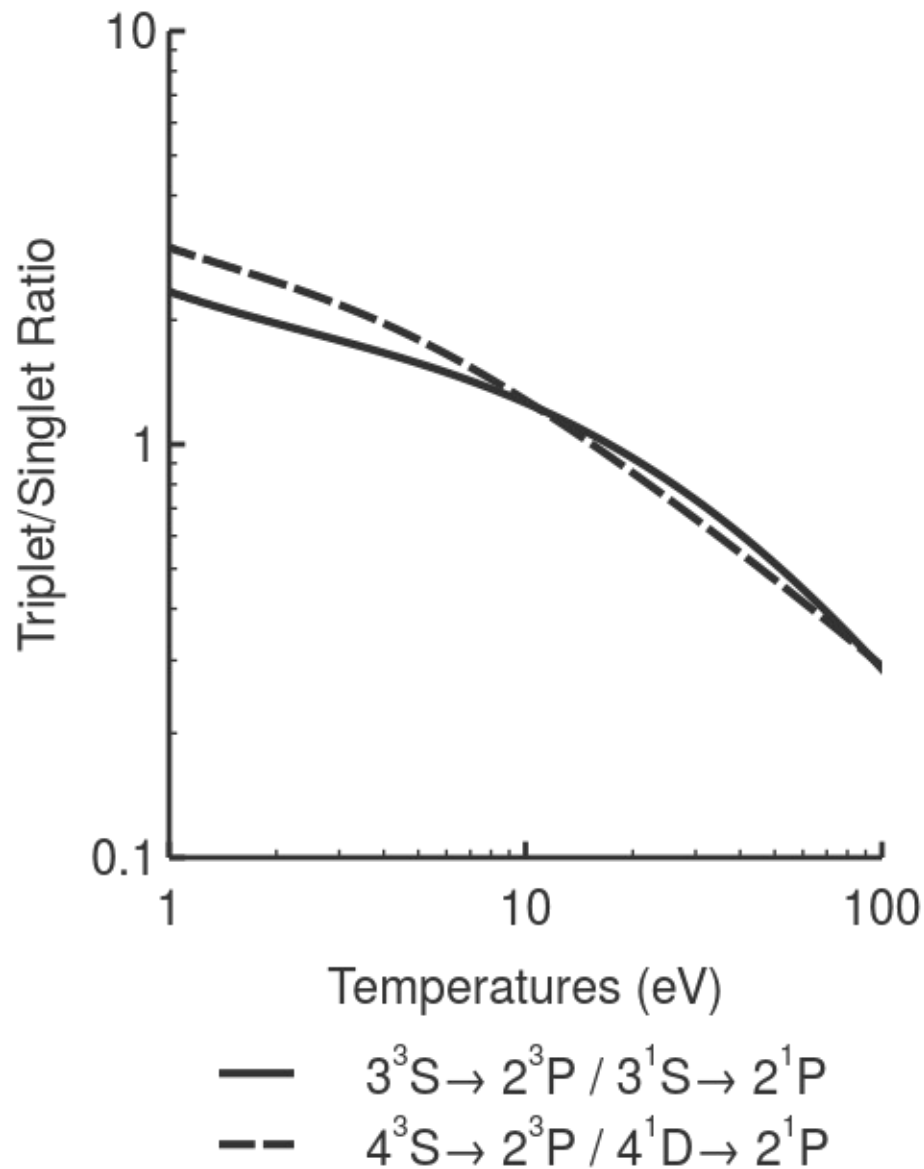
Pressure (Torr)	Trapping Factor	Effective Lifetime (s)
1.0	8,773	$1.549 \times 10^{-5}$
4.0	38,031	$6.715 \times 10^{-5}$
8.0	78,837	$1.392 \times 10^{-4}$

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



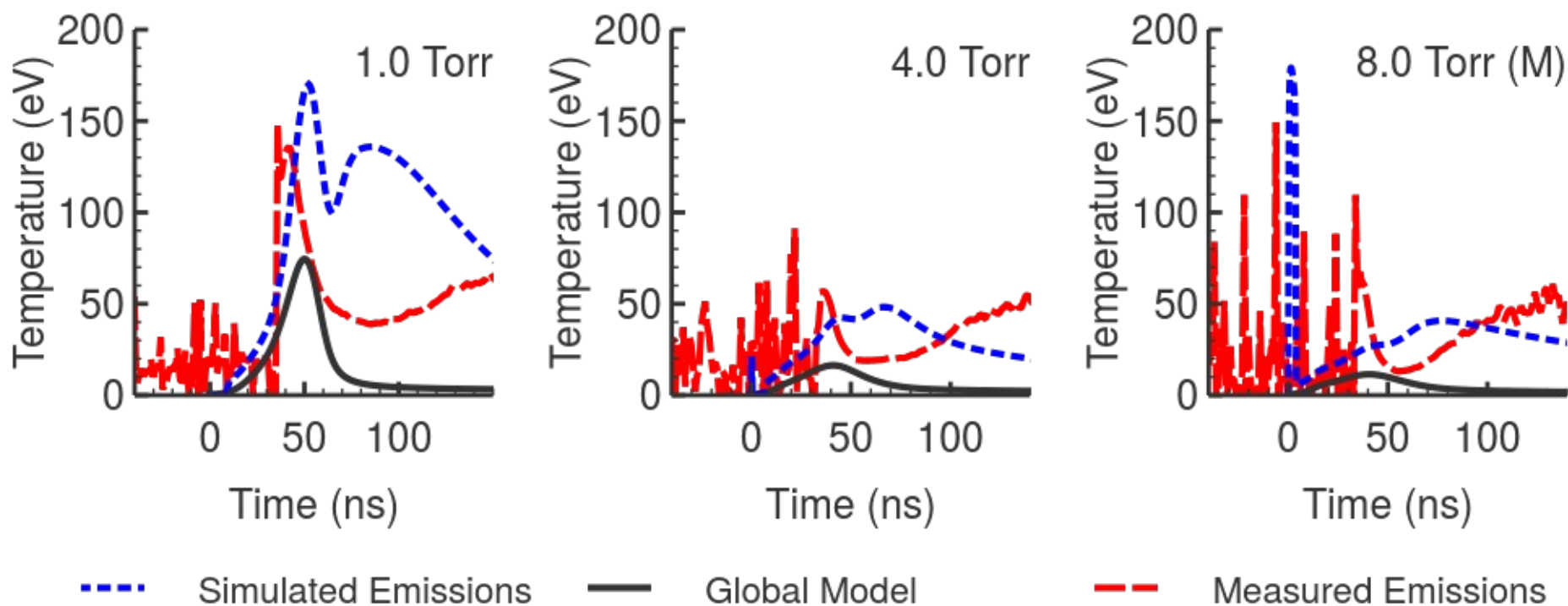


- Verify by comparison of 389 nm and 501 nm
- If radiation trapping affects system, measured lifetime of 501 line should be greater than simulation

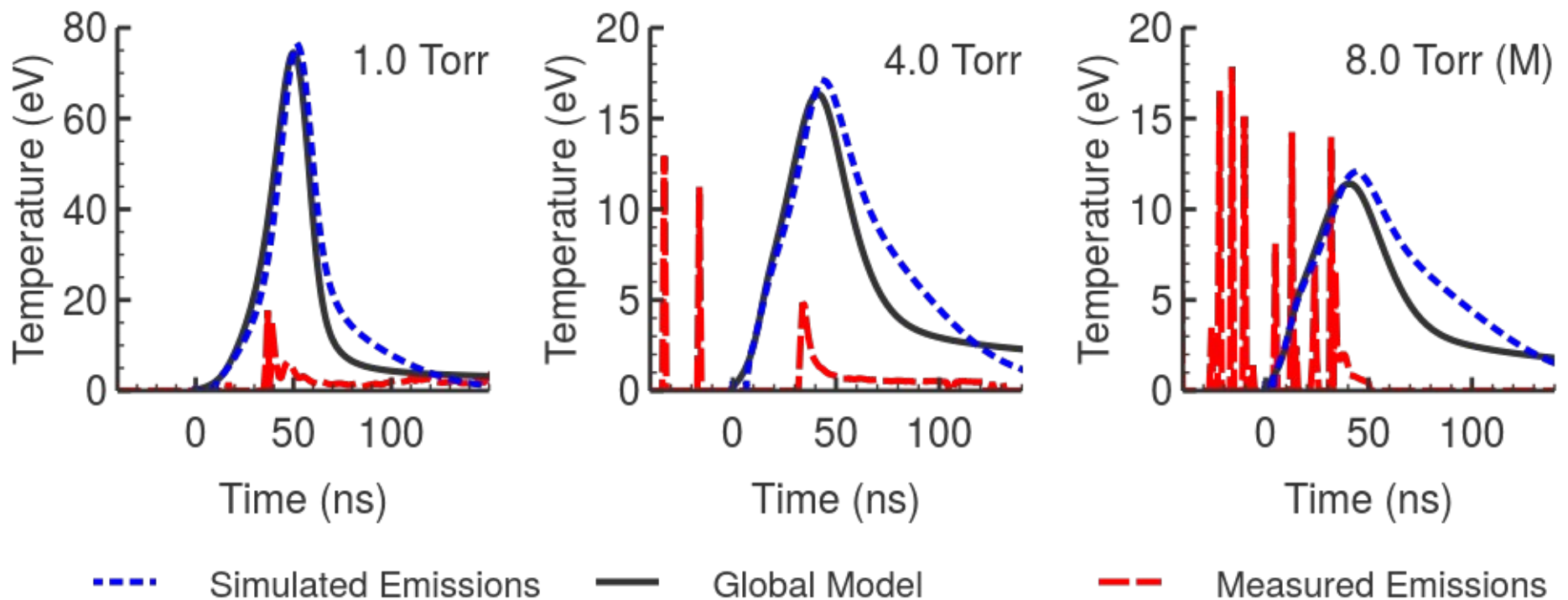


- Can be used to estimate electron temperature
- Assumes
  - Electron-ground excitation only
  - Radiative losses
- Maxwell-Boltzmann distribution

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



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



- 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

- 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 RPND

# 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

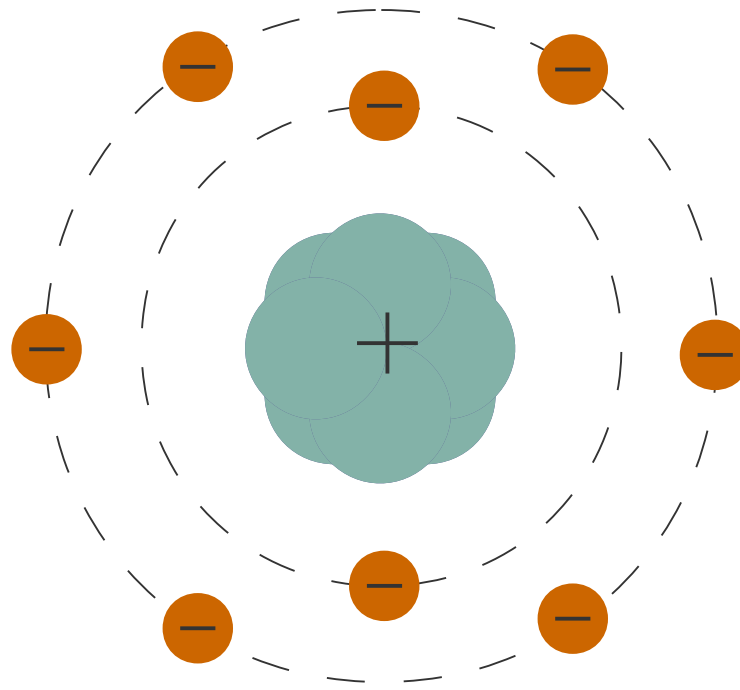


- Address temporal evolution of metastable density profile (Abel inversion) → wall effects
- Incorporate additional physics (Penning ionization of impurities, radiation trapping, etc.) in present global model.
- Employ different EEDF models to better match excited state dynamics
- Move to a 0D Monte-Carlo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

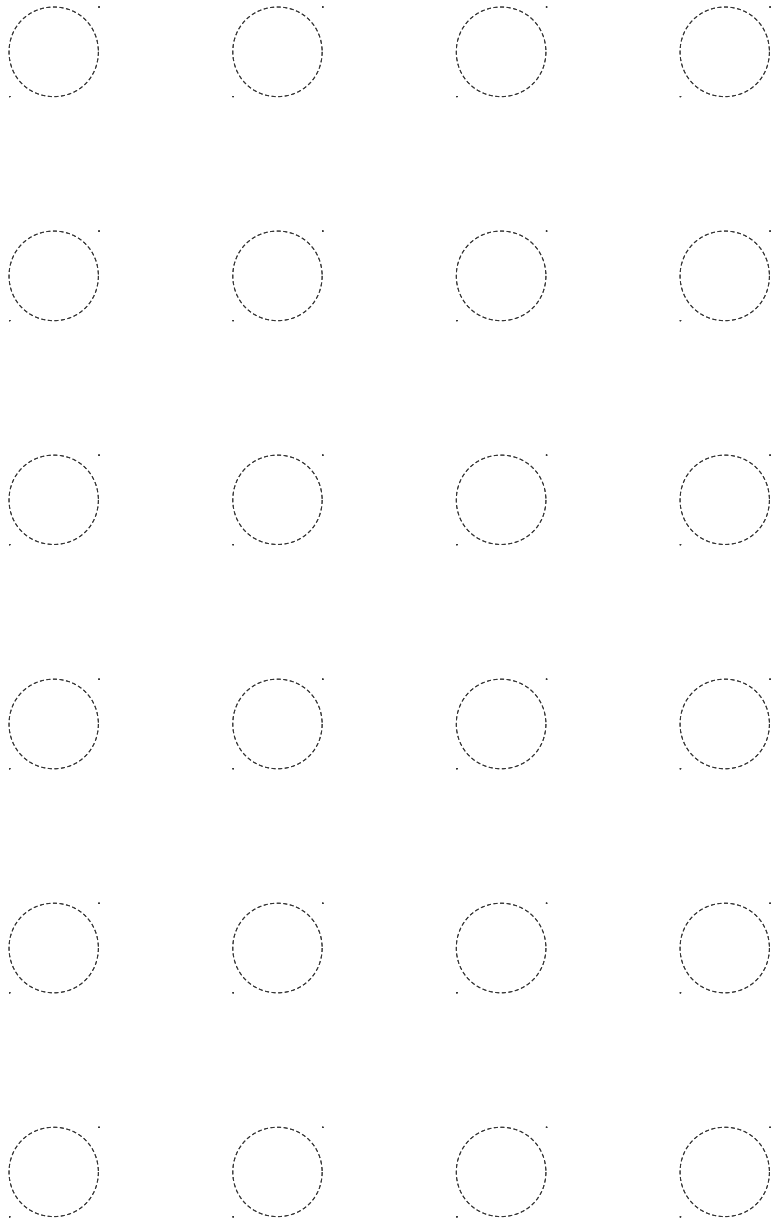
# Acknowledgements

# Questions

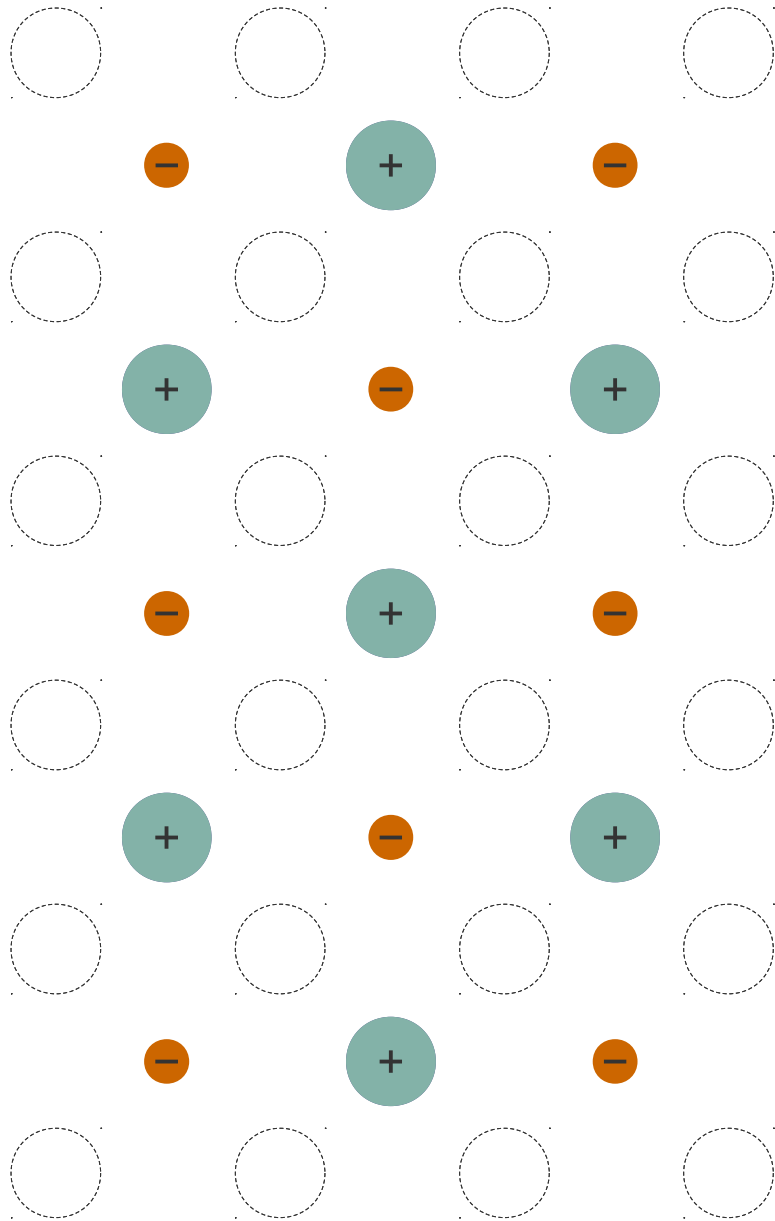
# Backup Slides



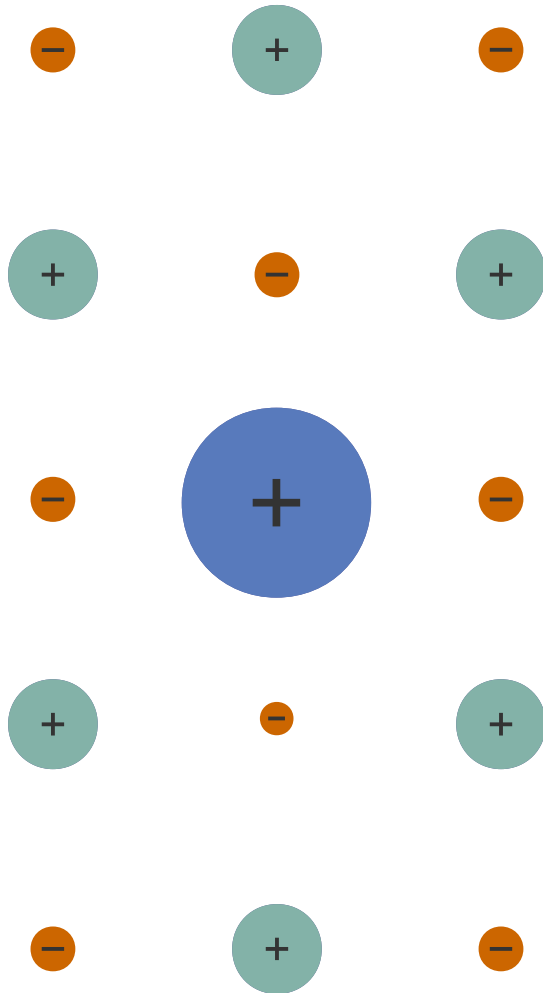
- Positively-charged nucleus
- Orbited by negatively-charged electrons
- Overall, electrically neutral



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

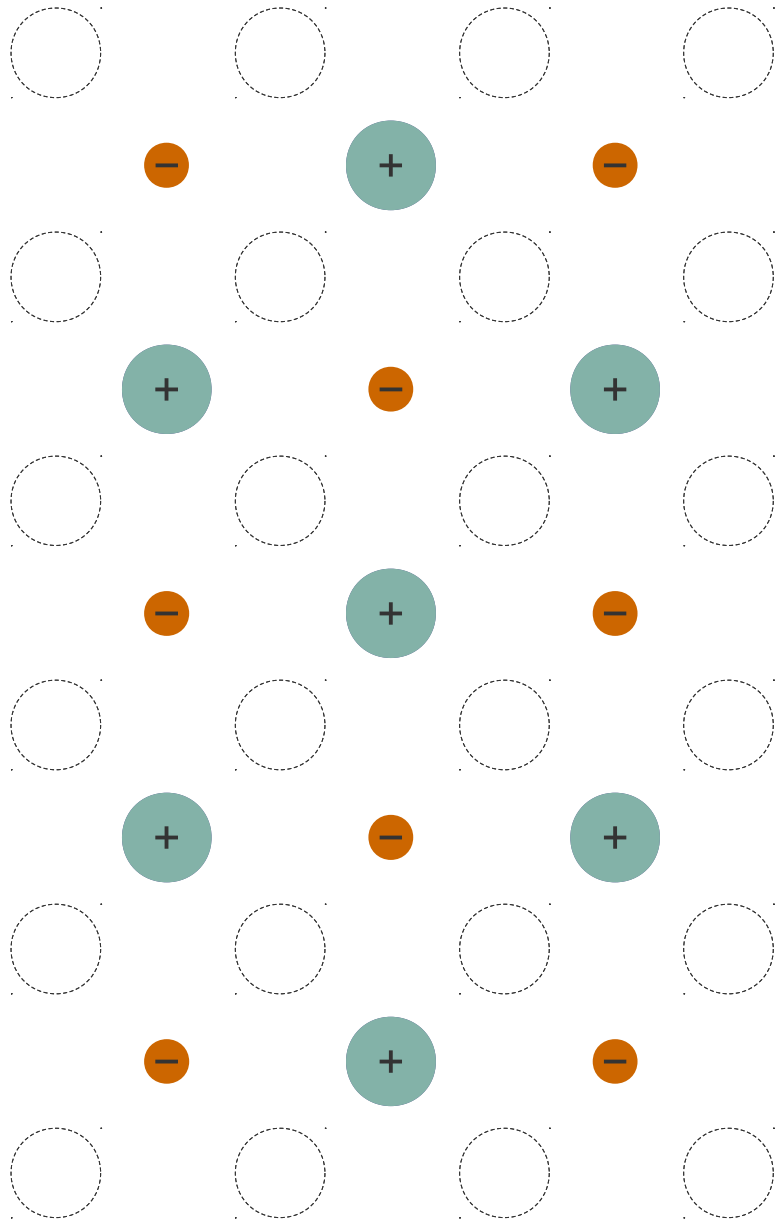


- Add energy to gas
  - Electricity
  - Shock
  - Light
- Electrons separate from atoms
- Collection of neutral atoms, (positive) ions, and (negative) electrons
- What makes a plasma?

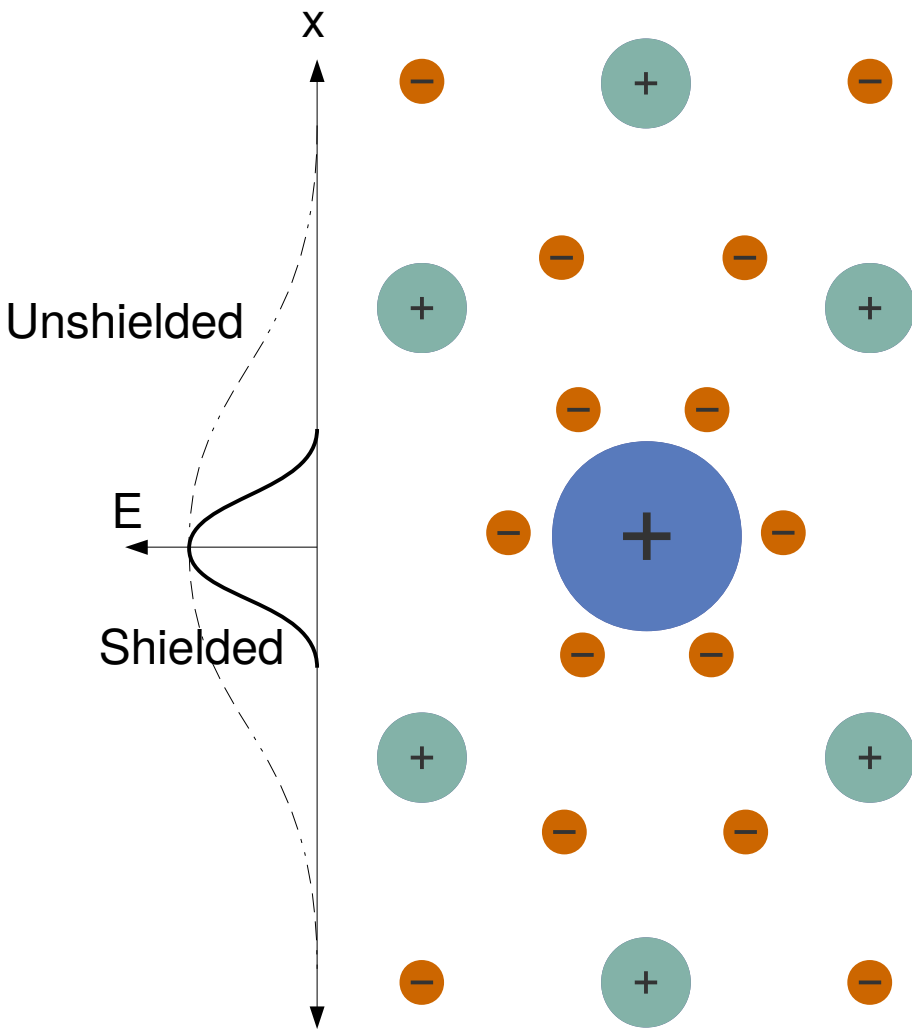


Consider what happens with an electrical perturbation.

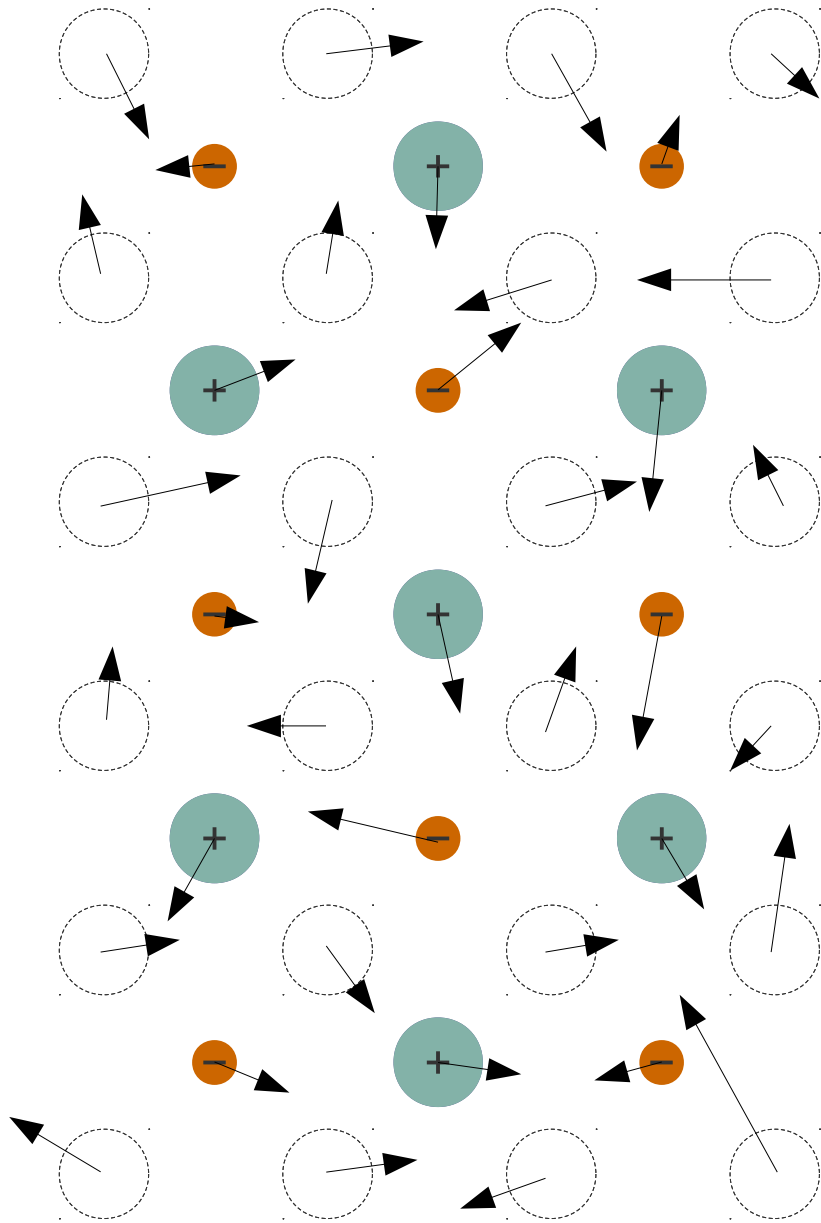




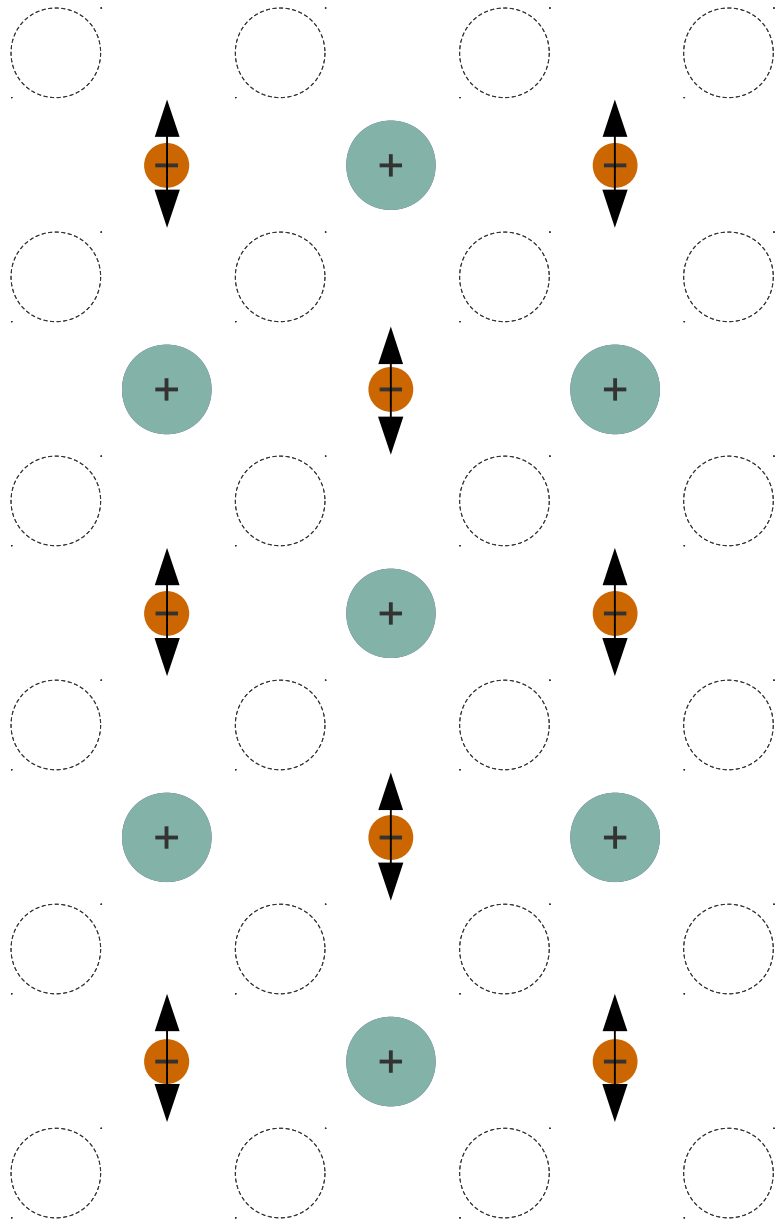
- Add energy to gas
  - Electricity
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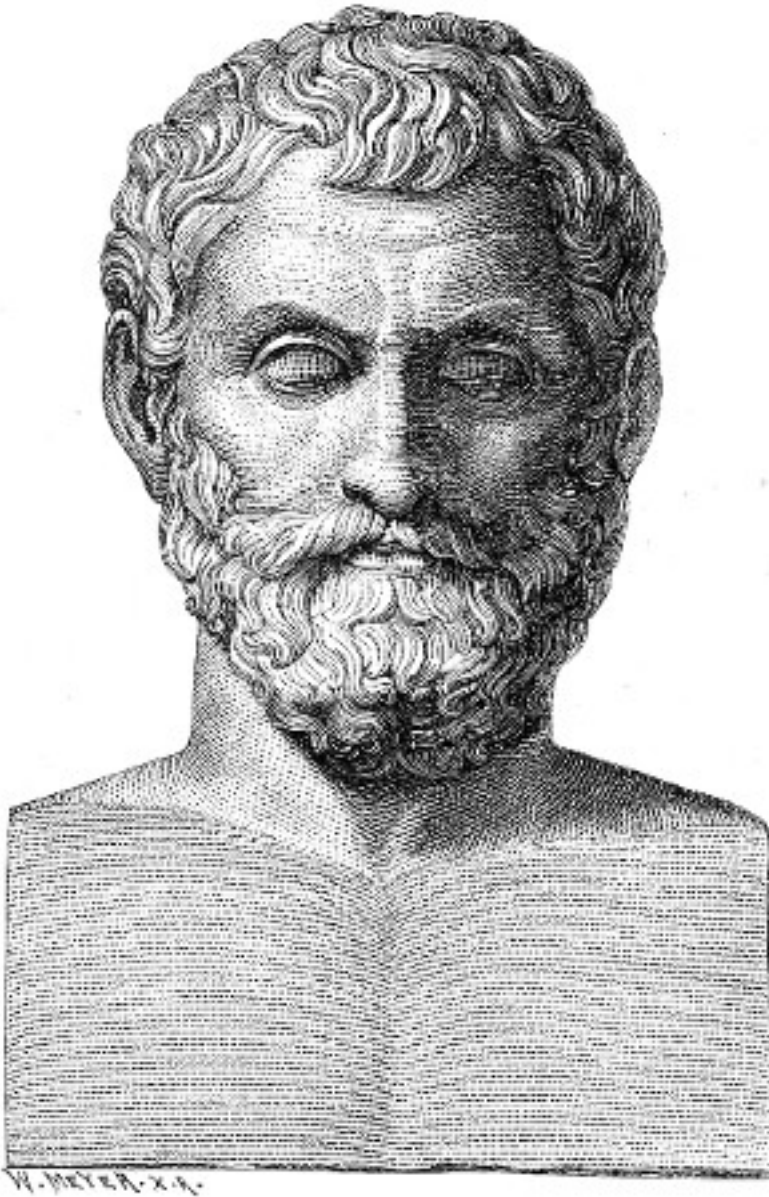
- Electrons move to shield charge
- Similar for positive and negative perturbation
- Electric field falls off quickly
- $\lambda < L, n\lambda^3 \gg 1$



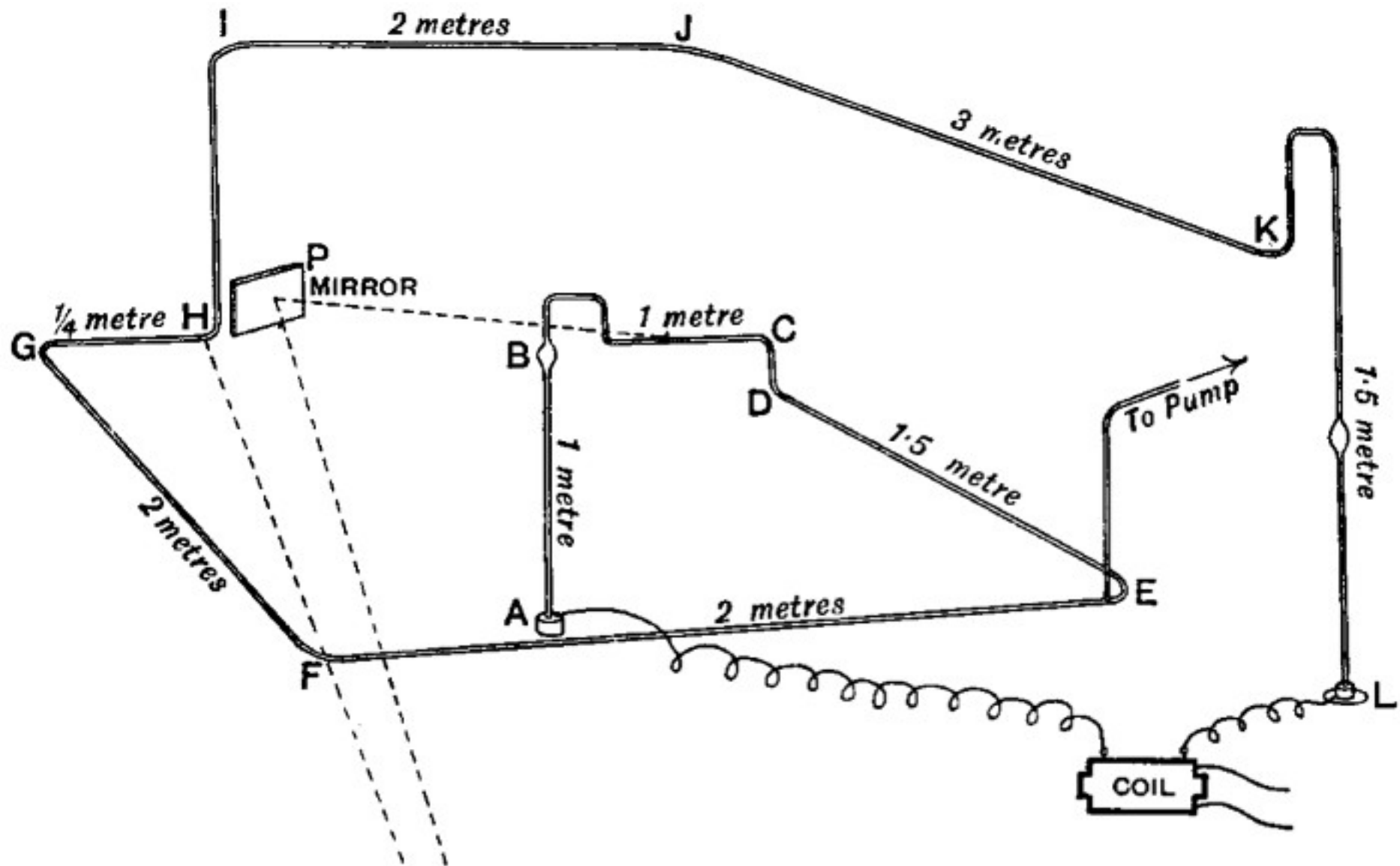
- Random collisions with neutral particles
- Neutral collisions can still determine properties of ionized gas
- Electric interactions should be dominant in plasma



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

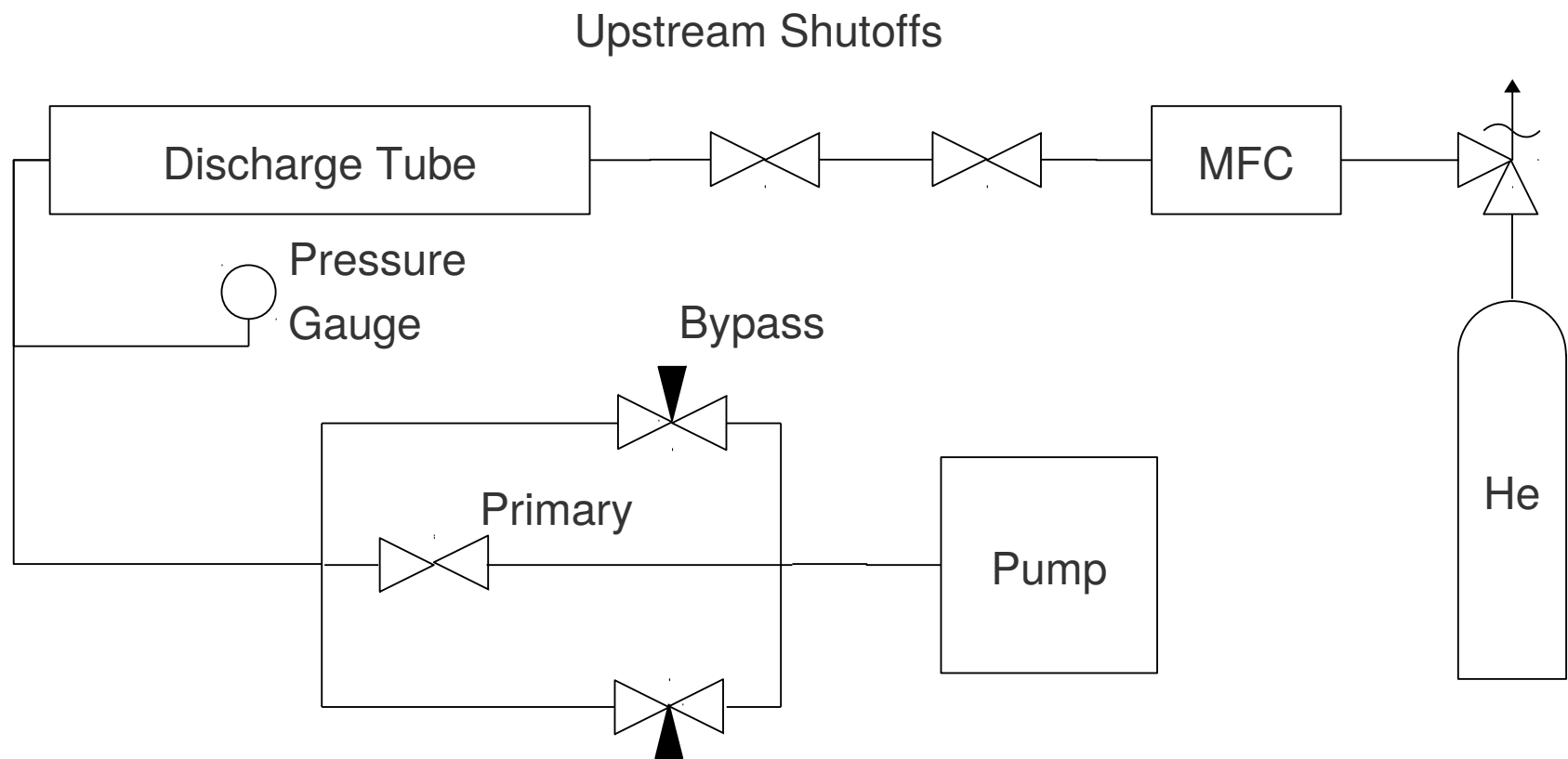


Wallis, E. et al. "Illustrerad verldshistoria utgifven av E. Wallis. Volume I." (1875).  
Stotesbury, H. "J. J. Thomson." Popular Science, Vol. 56 (1899).

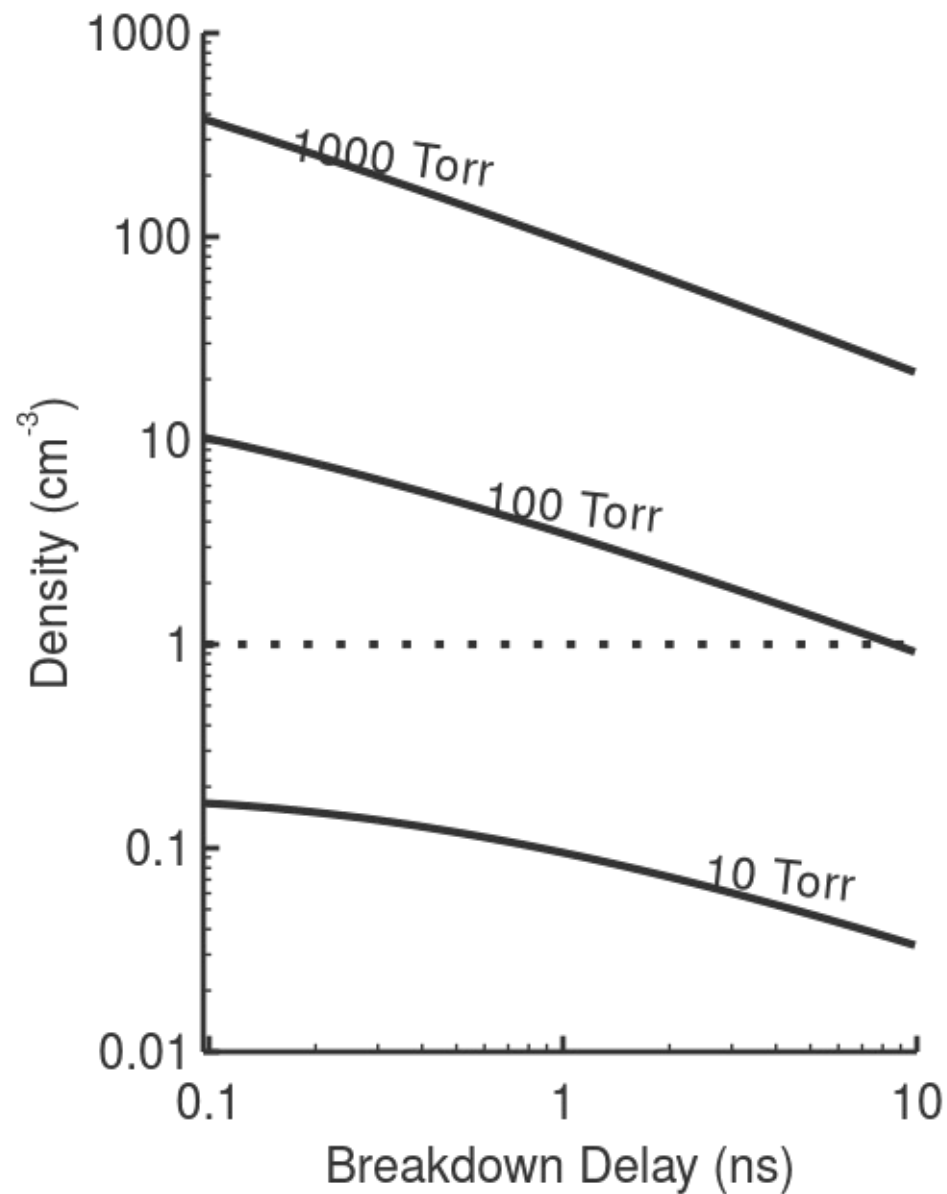
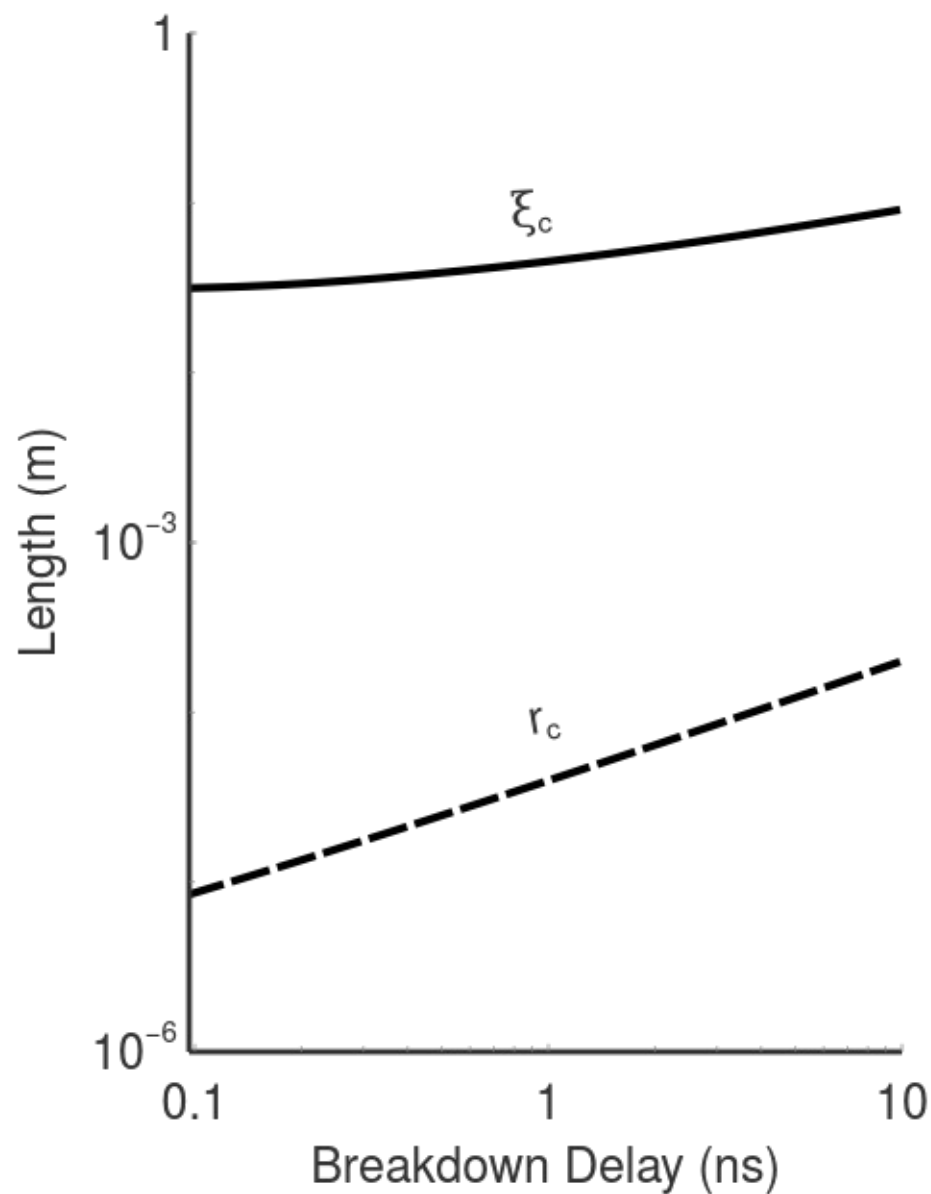


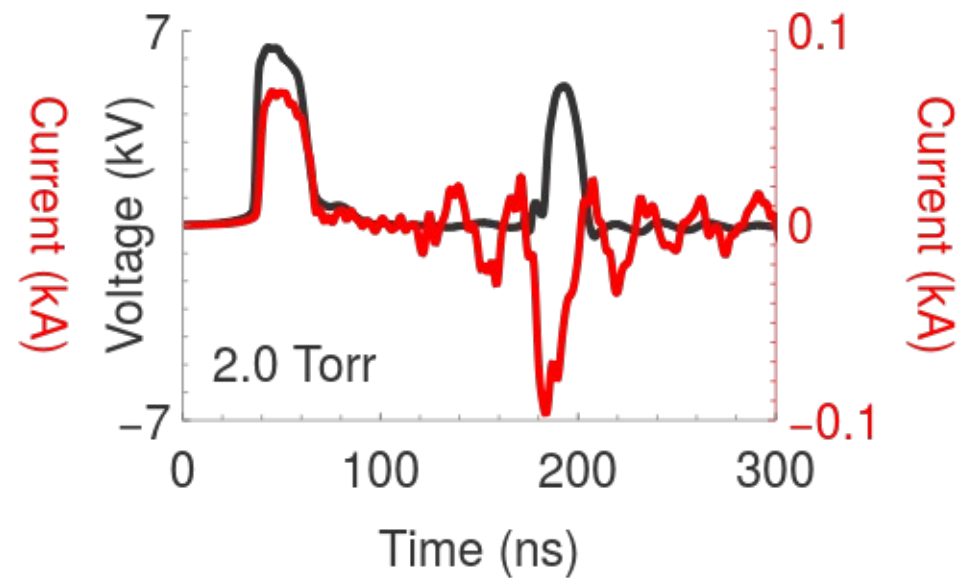
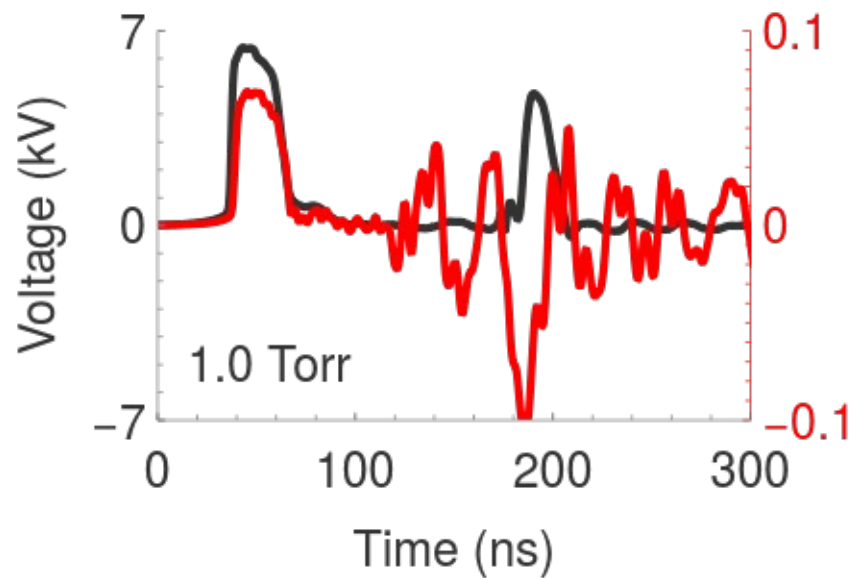
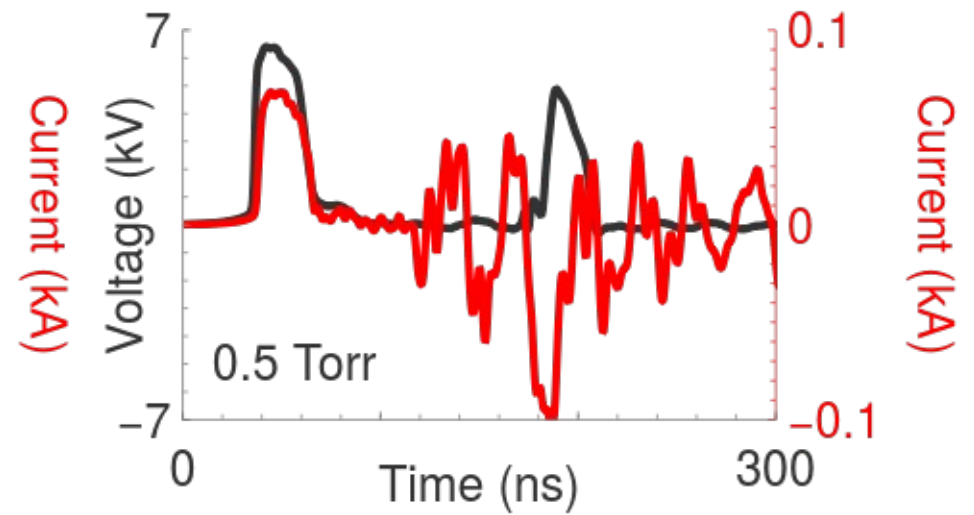
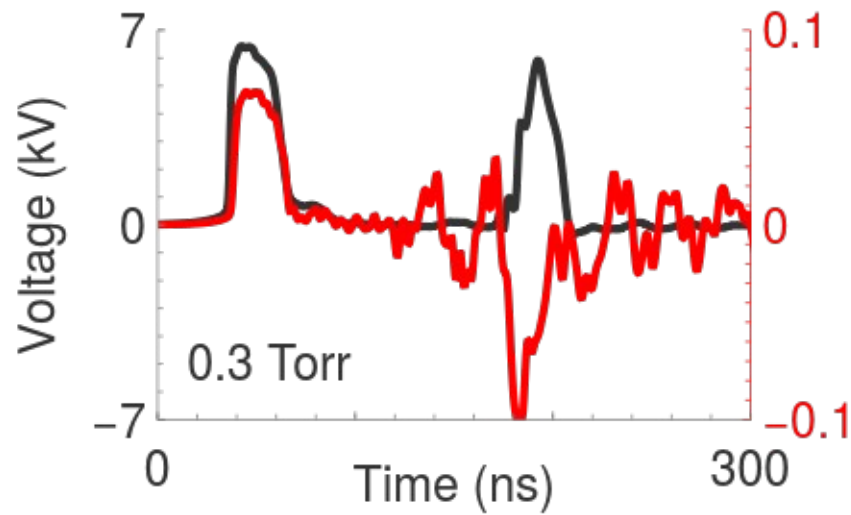
J. J. Thomson. "Notes on Recent Researches in Electricity and Magnetism." Clarendon Press, Oxford, UK, 1893.

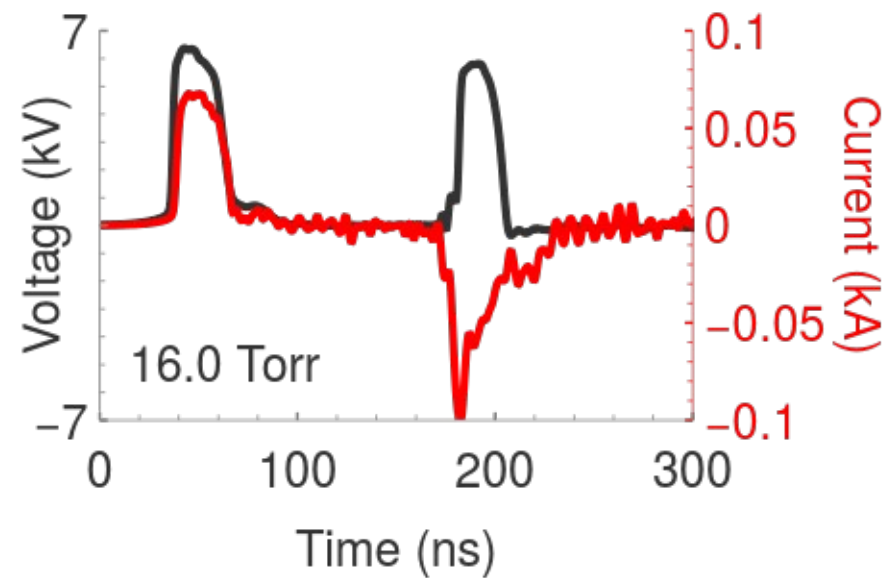
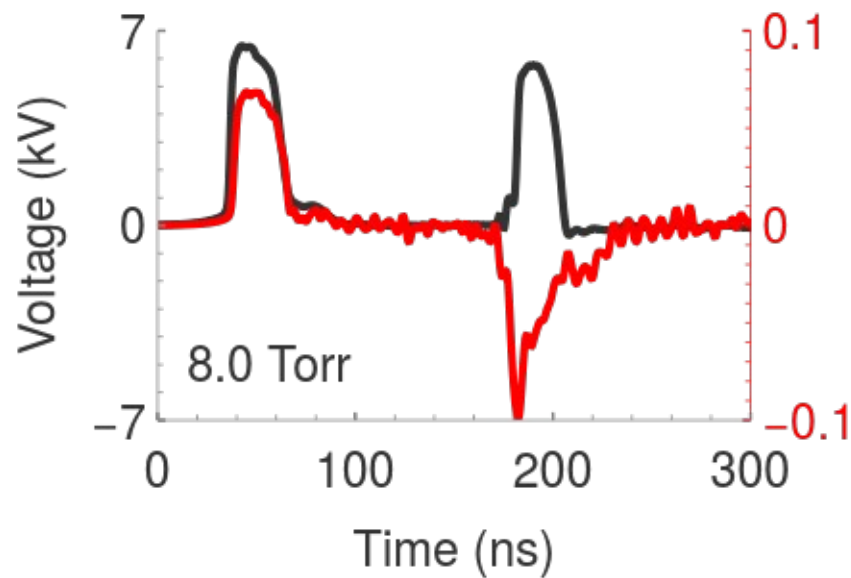
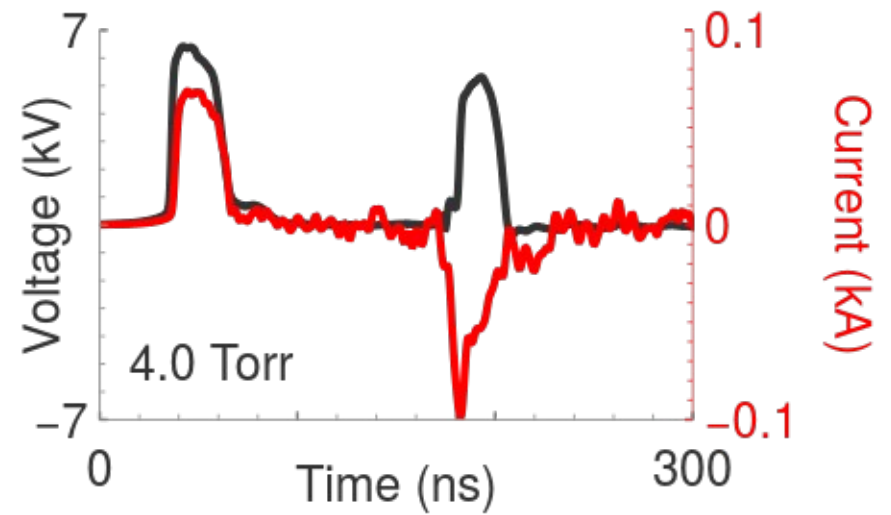
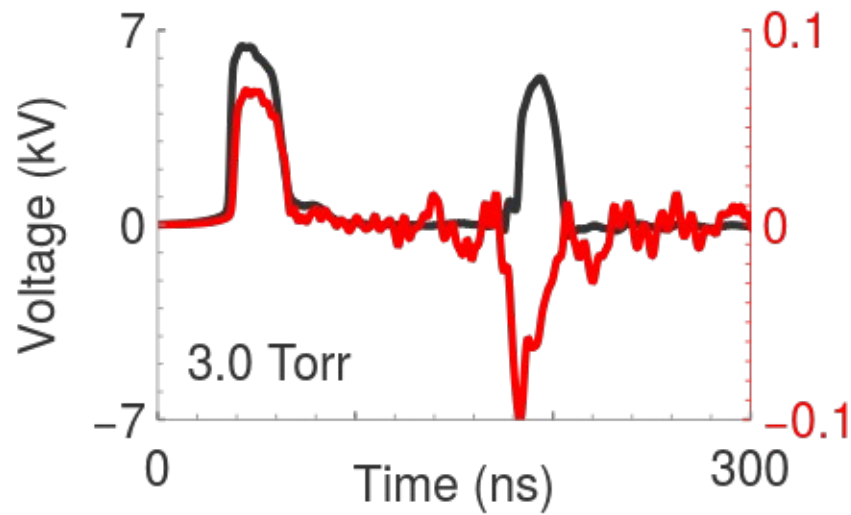
- Normally controlled by boundaries which limit gas flow out of plasma
1. Discharge causes gas heating
  2. Results in local rarefaction
  3. Increases reduced electric field
  4. Increases ionization rate and power deposition
  5. Results in increased gas heating

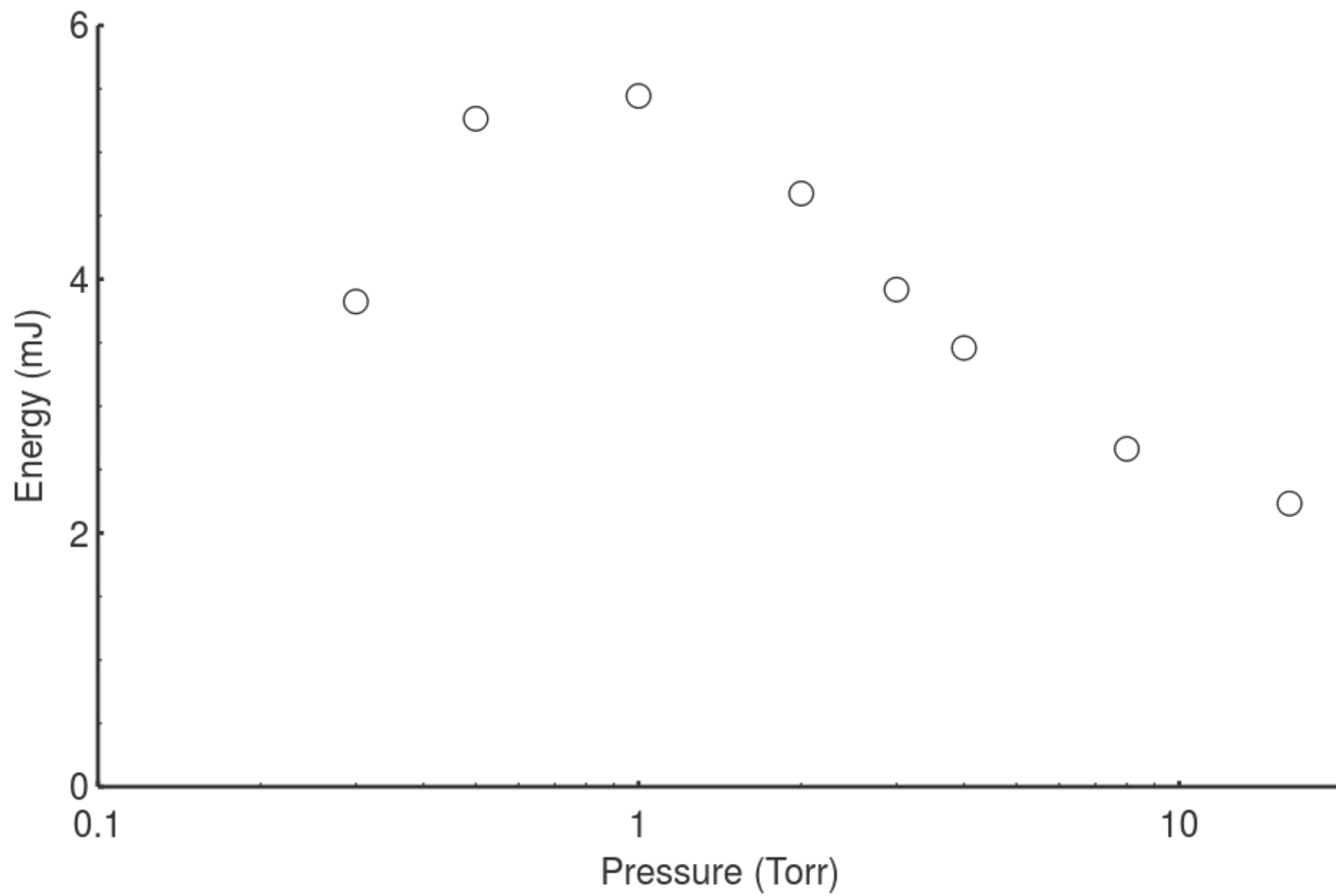








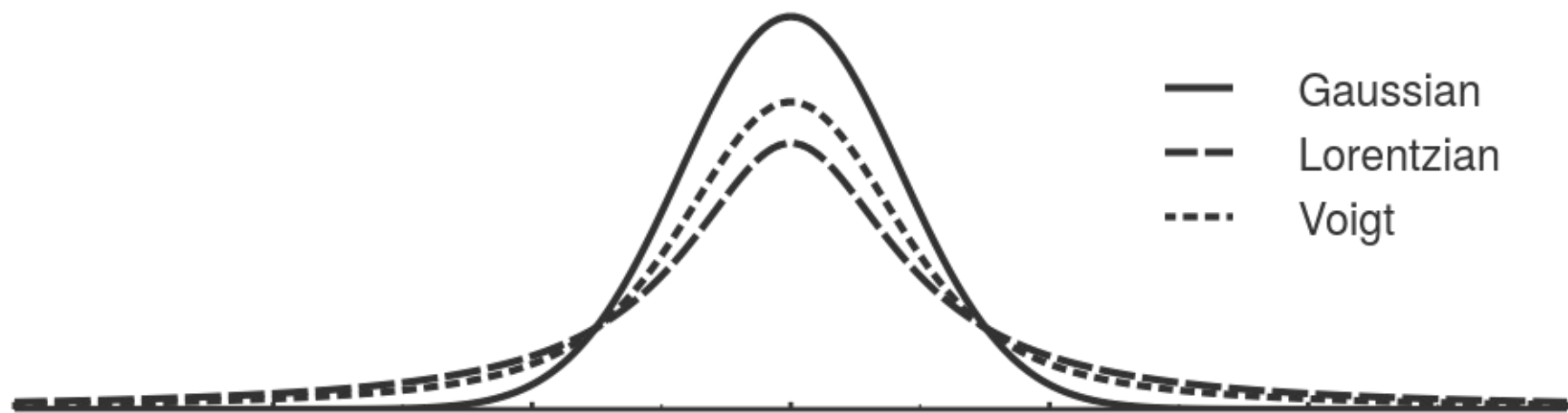




$$\Delta N = \Delta N_0 \frac{1}{1 + W\tau_{\text{eff}}} = \Delta N_0 \frac{1}{1 + I/I_{\text{sat}}}$$

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

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

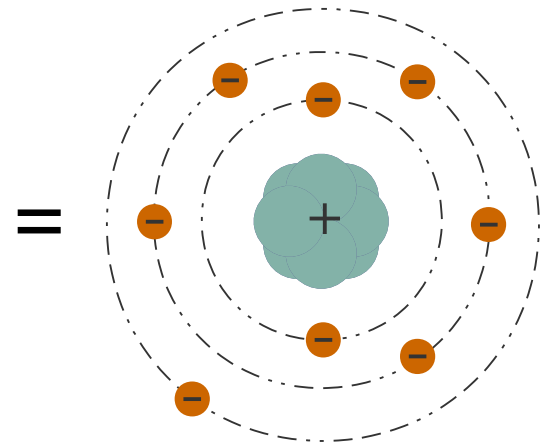
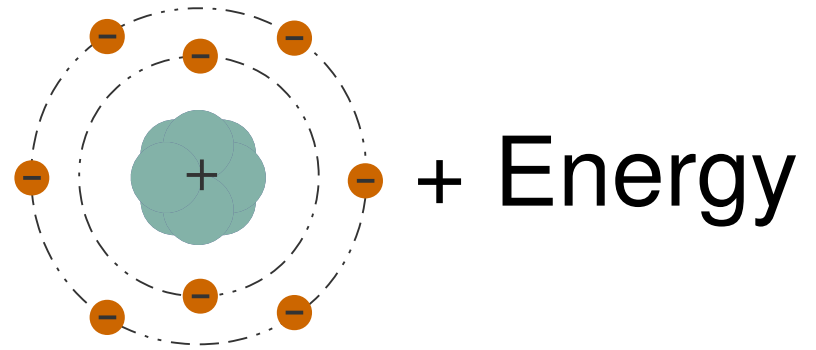


Lorentzian  $g(\omega) = -\frac{1}{4\pi^2} \frac{A\lambda^3}{\Delta\omega_a} \frac{1}{1 + [2(\omega - \omega_a)/\Delta\omega_a]^2}$

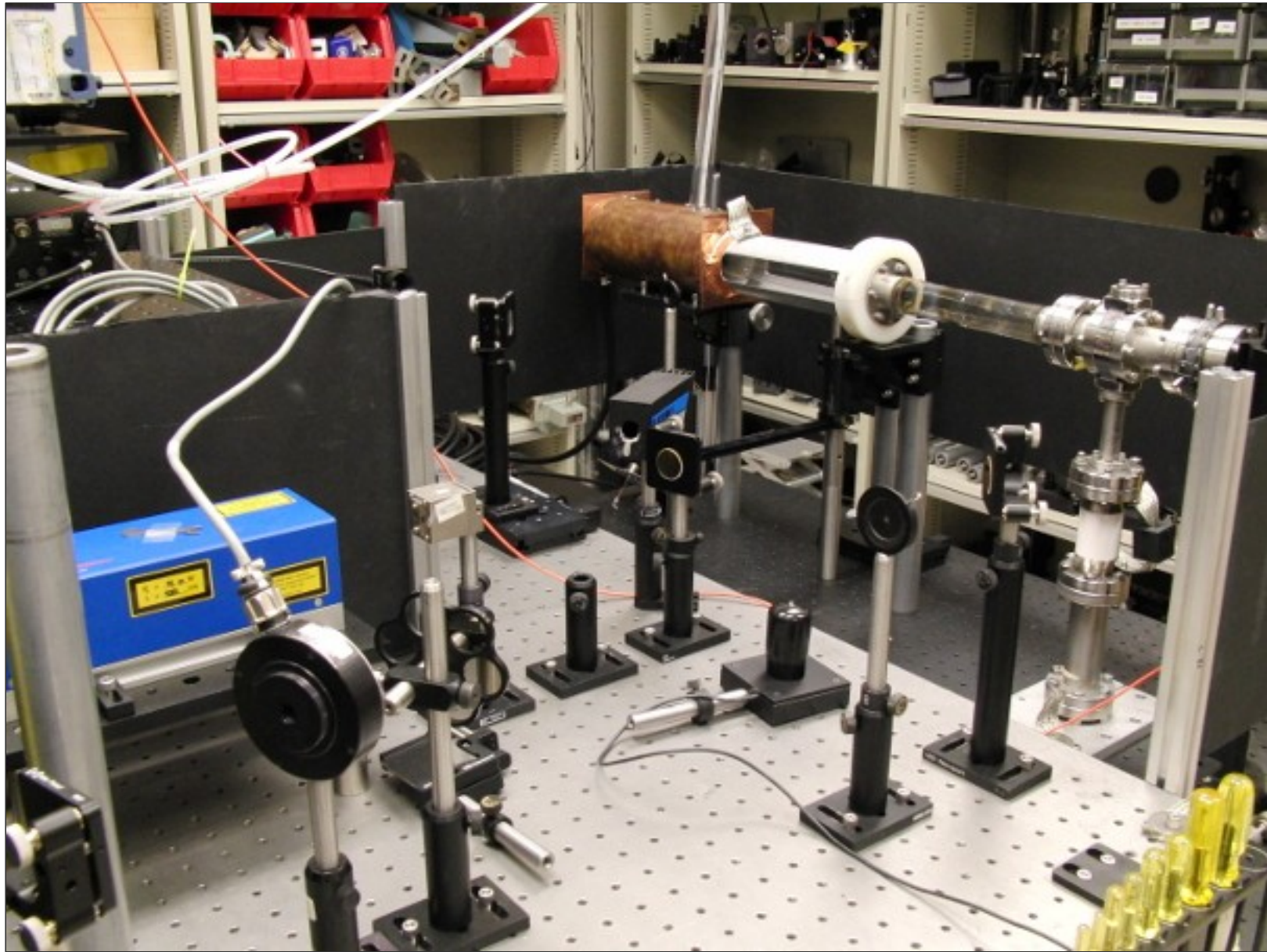
Voigt  $g(\omega) = \sqrt{\frac{2 \ln 2}{\pi^3}} \frac{\Delta\omega_d}{\Delta\omega_d} \int_{-\infty}^{\infty} \frac{1}{[(\omega - \omega_a) - \omega']^2 + 4^2} \times \exp \left[ 4 \ln 2 \left( \frac{\omega'}{\Delta\omega_d} \right)^2 \right] d\omega'$

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

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

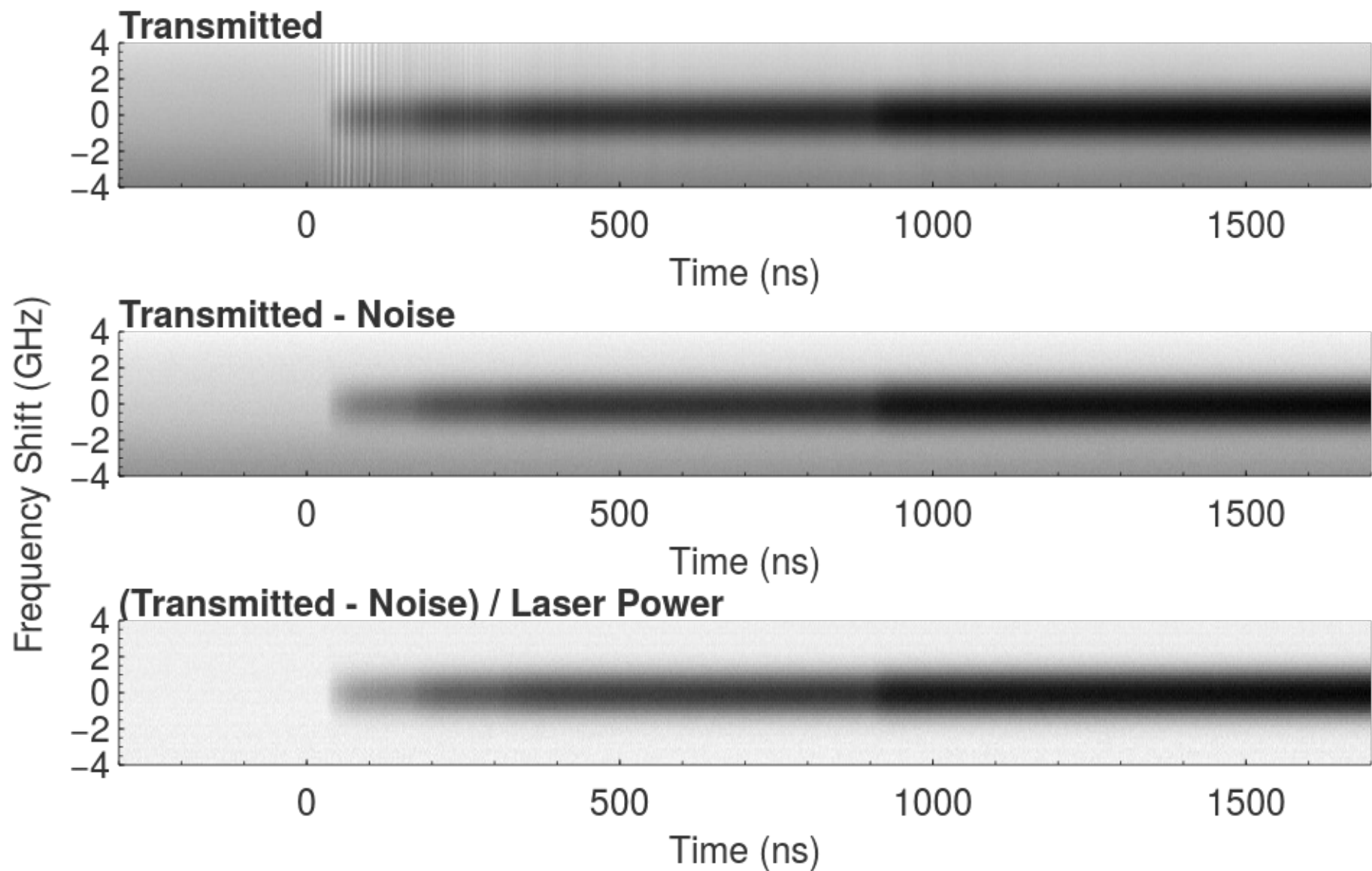


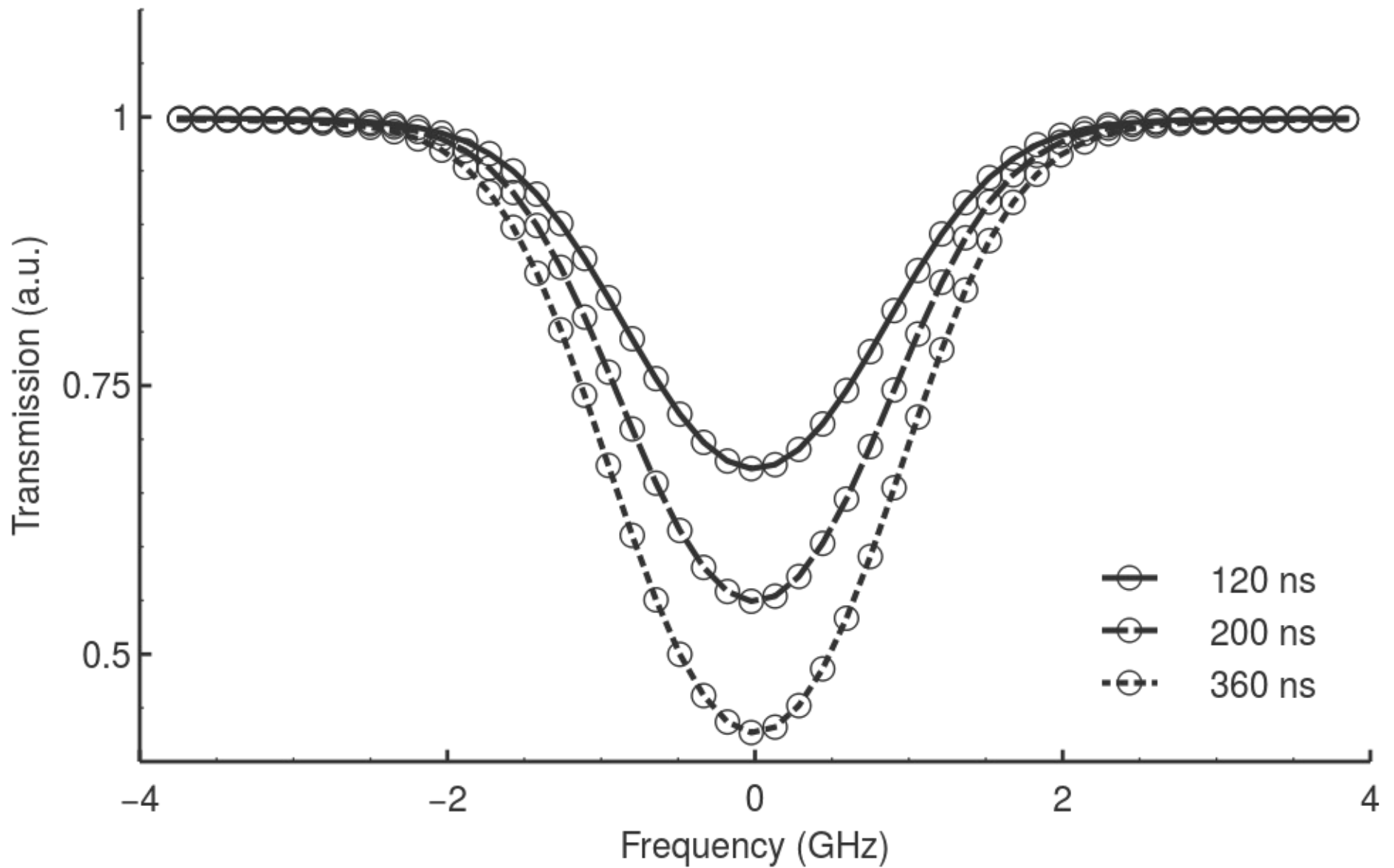


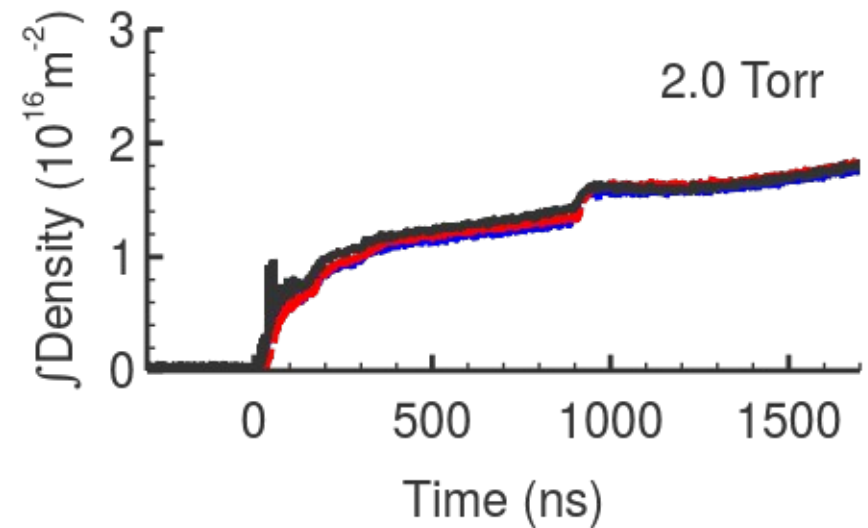
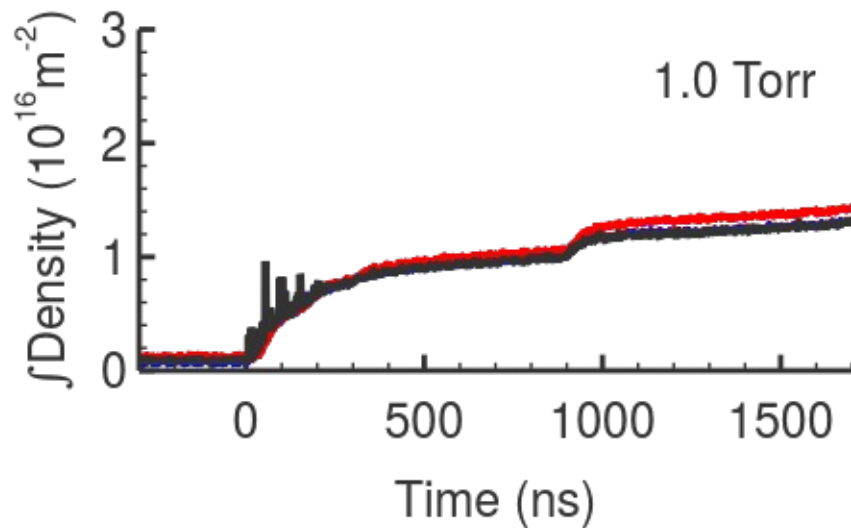
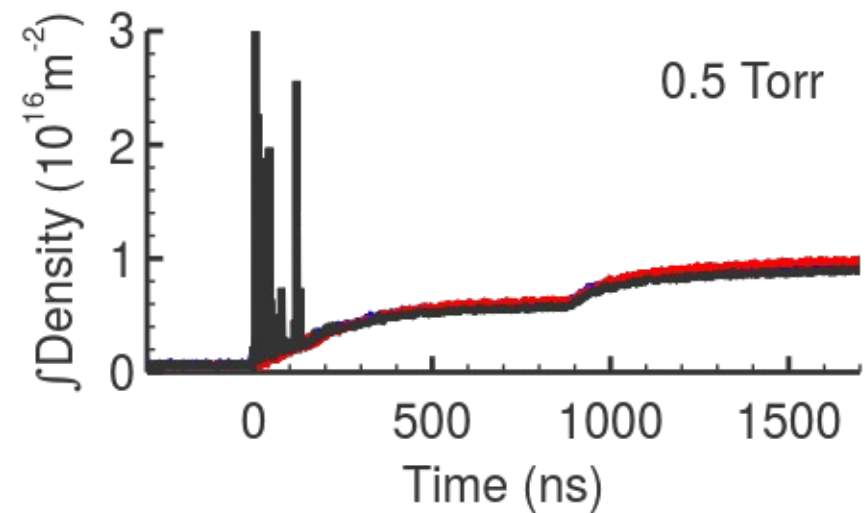
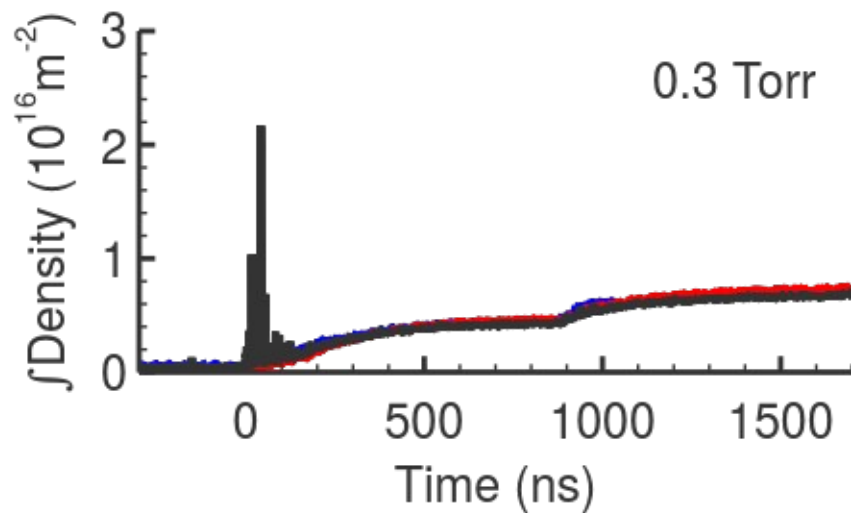


# Laser-Absorption Spectroscopy Setup

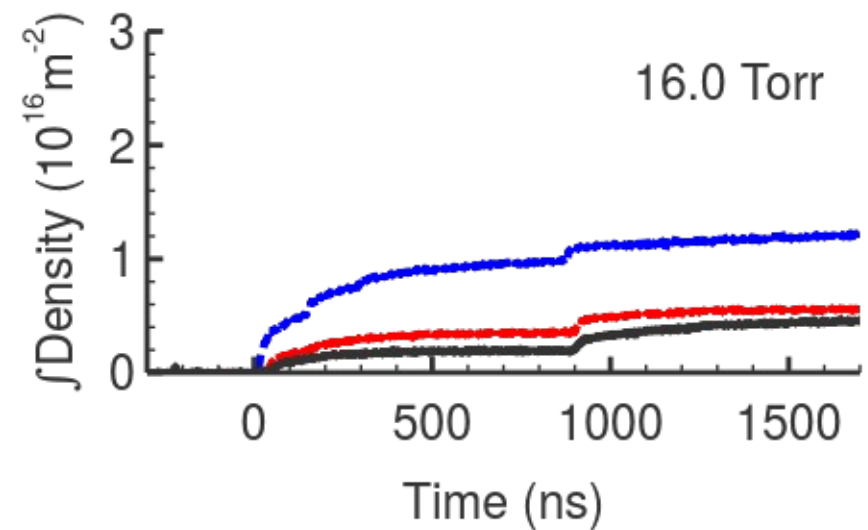
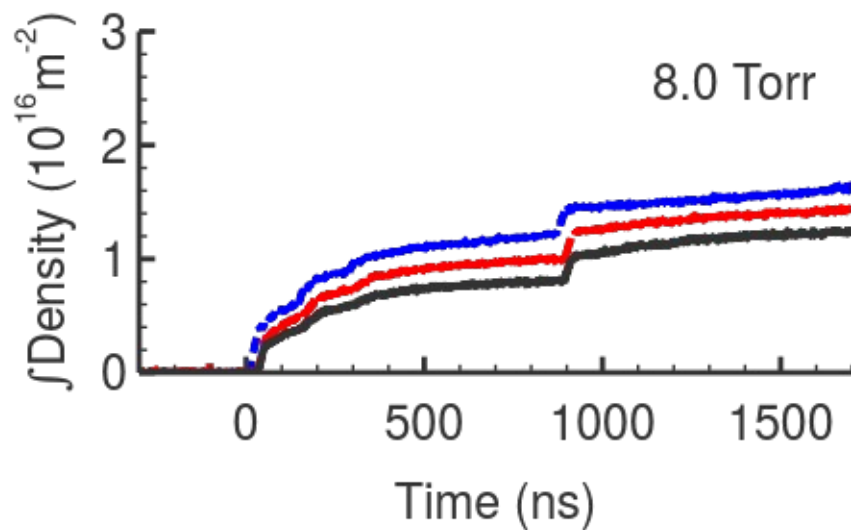
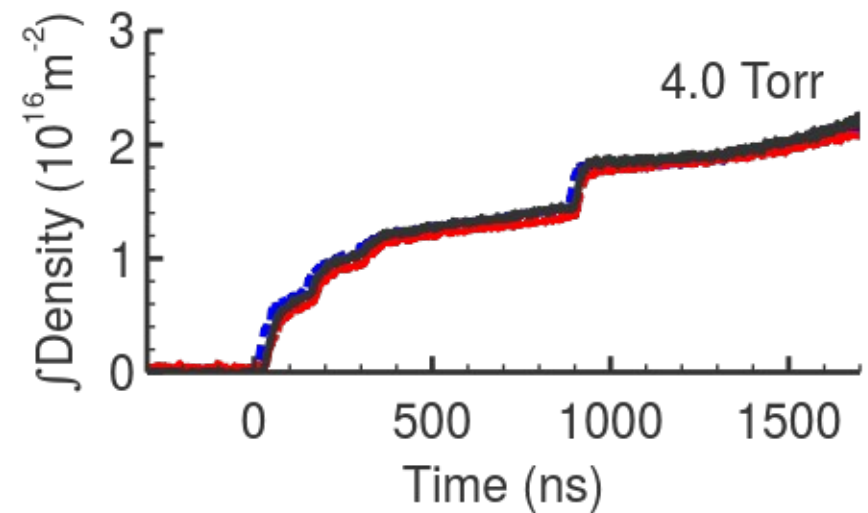
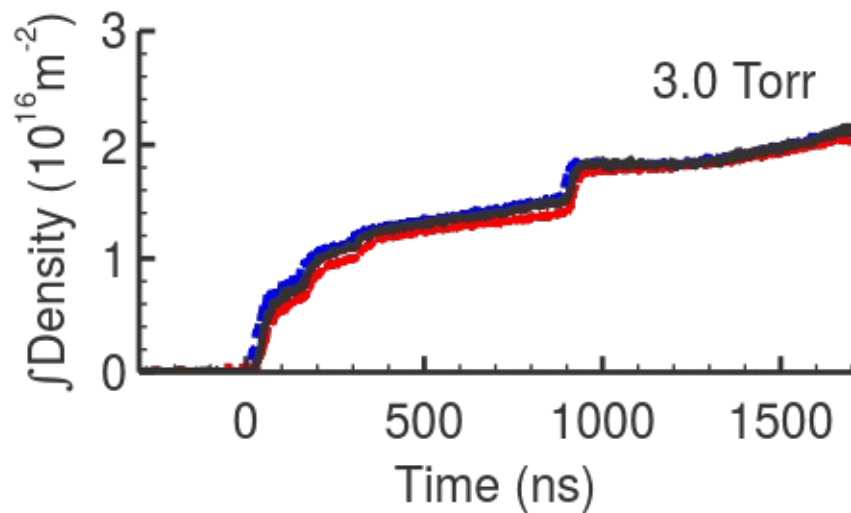




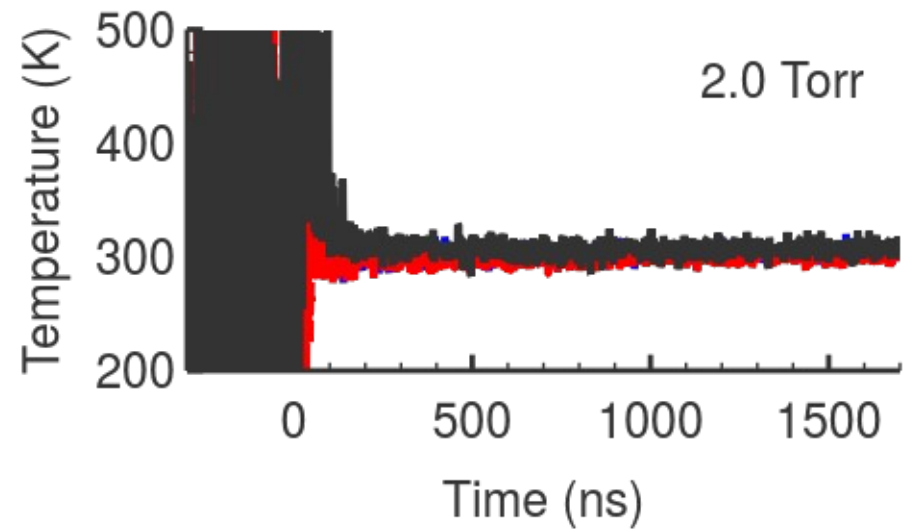
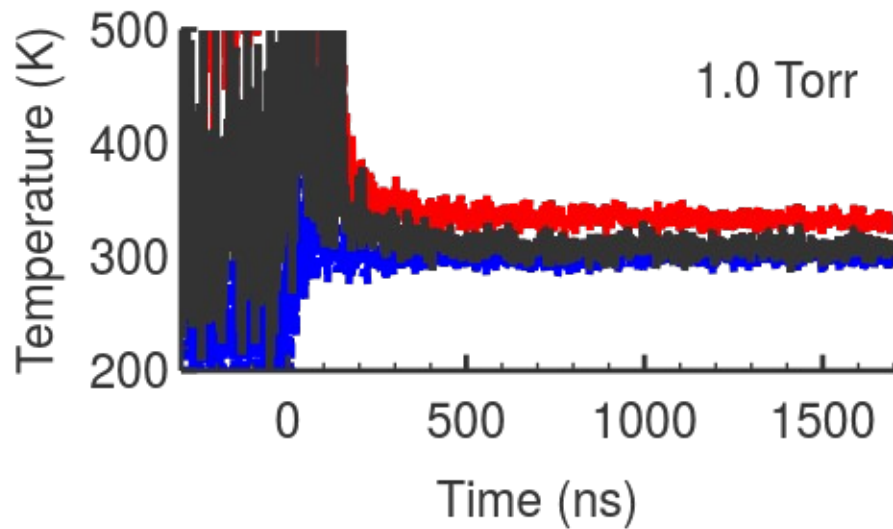
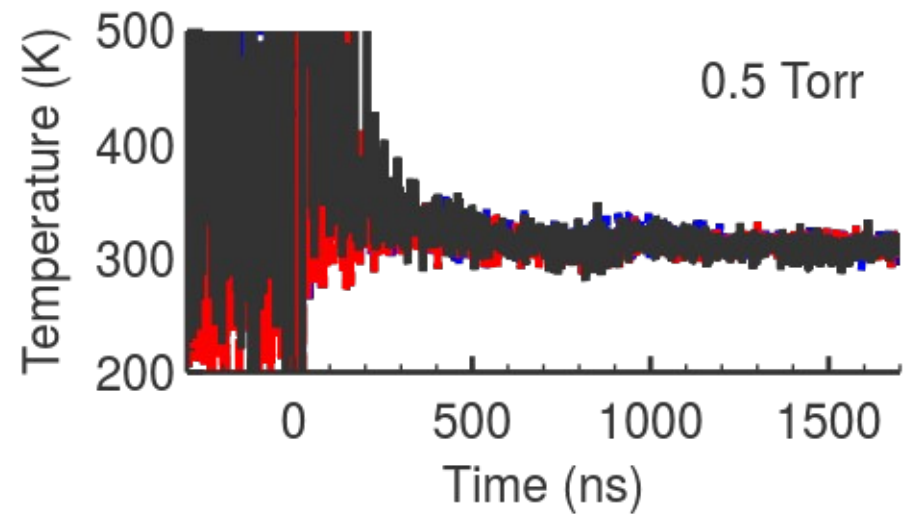
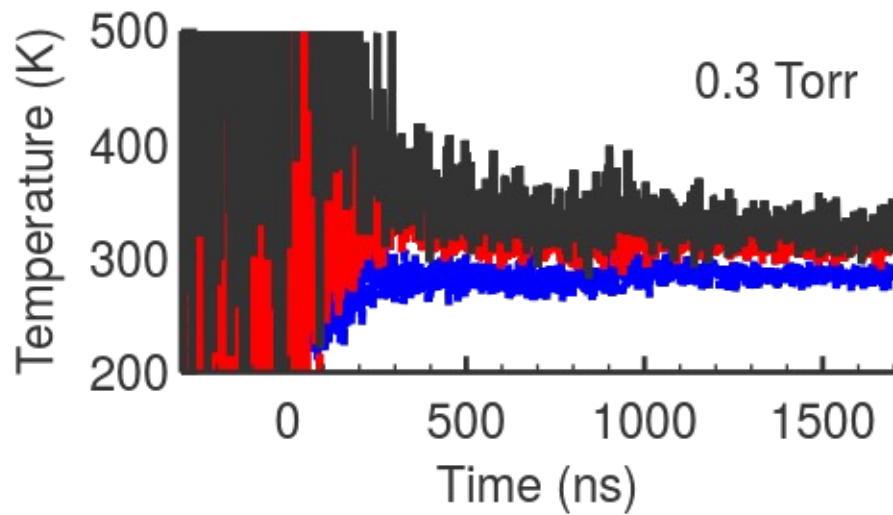




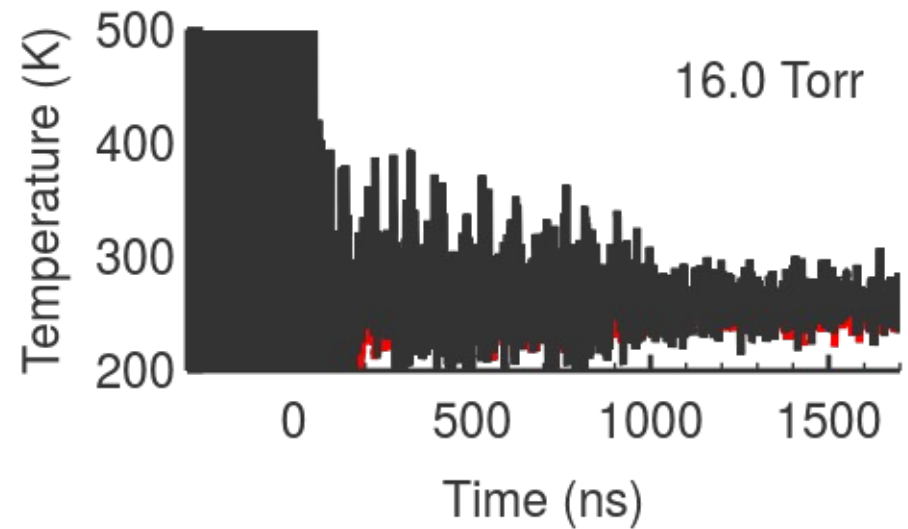
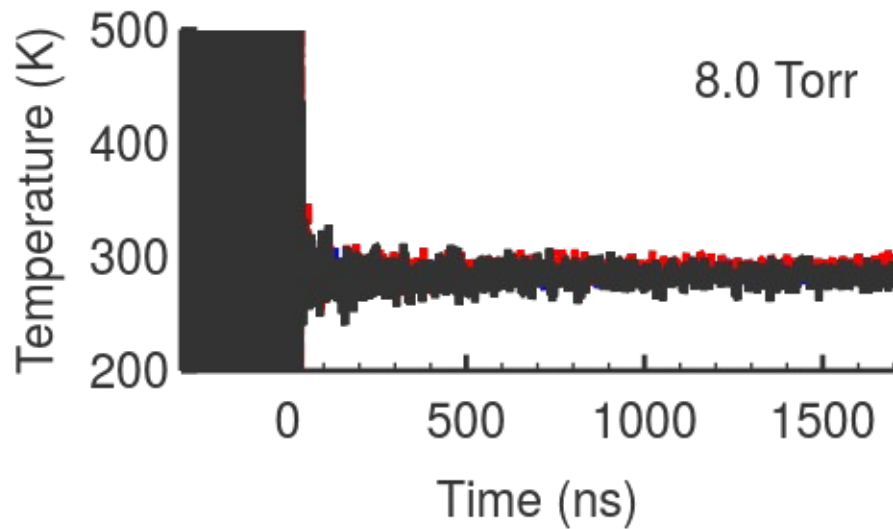
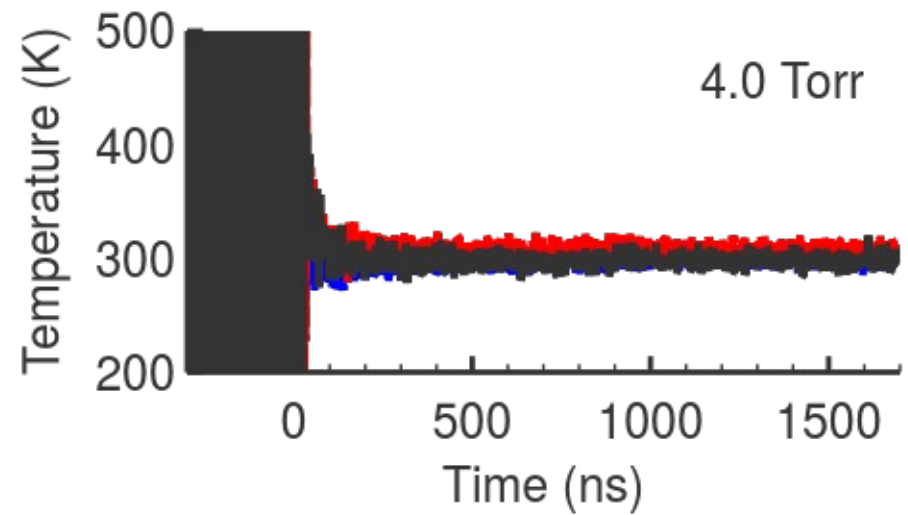
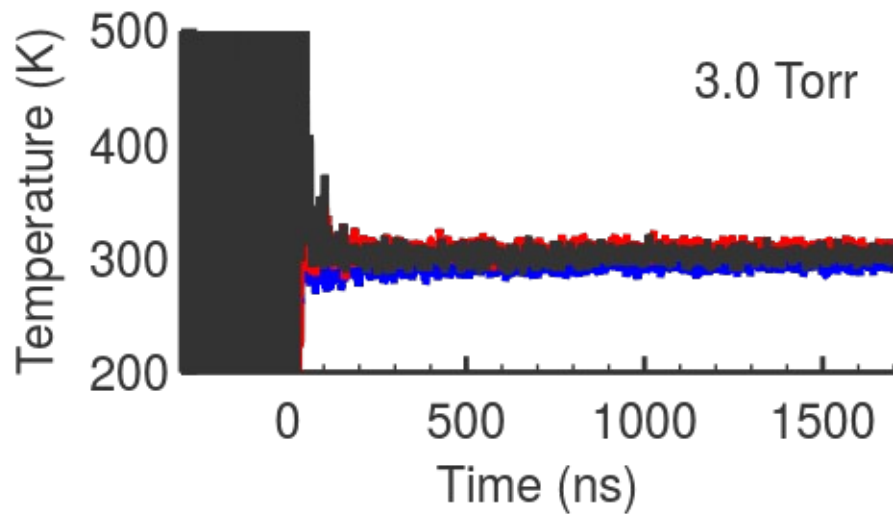
— Downstream    - - - Midstream    ··· Upstream



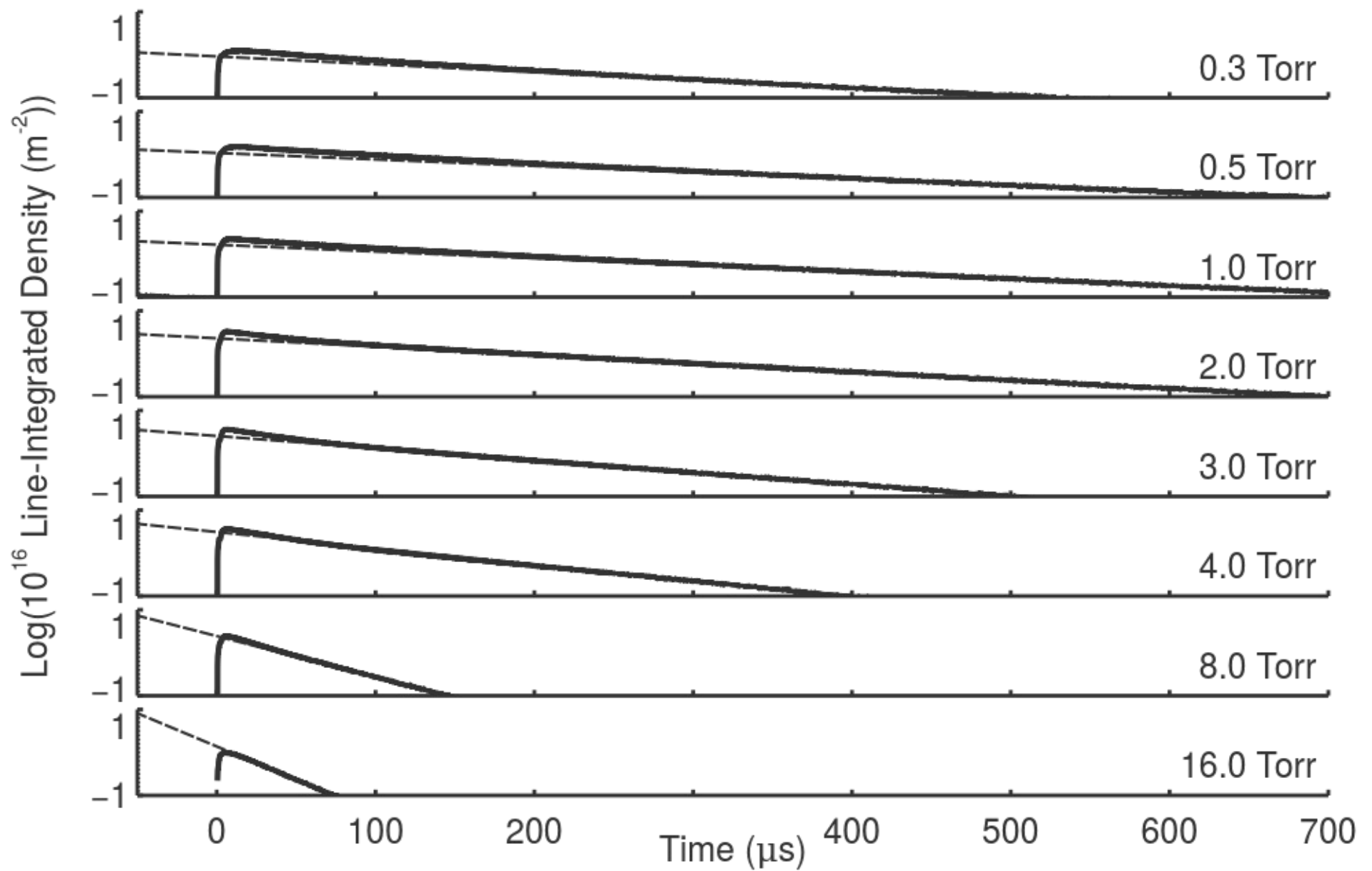
— Downstream    - - - Midstream    ··· Upstream



— Downstream    - - - Midstream    . . . Upstream



— Downstream    - - - Midstream    . . . Upstream



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

- Describes the evolution of the distribution function for a particle  $\alpha$
- 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

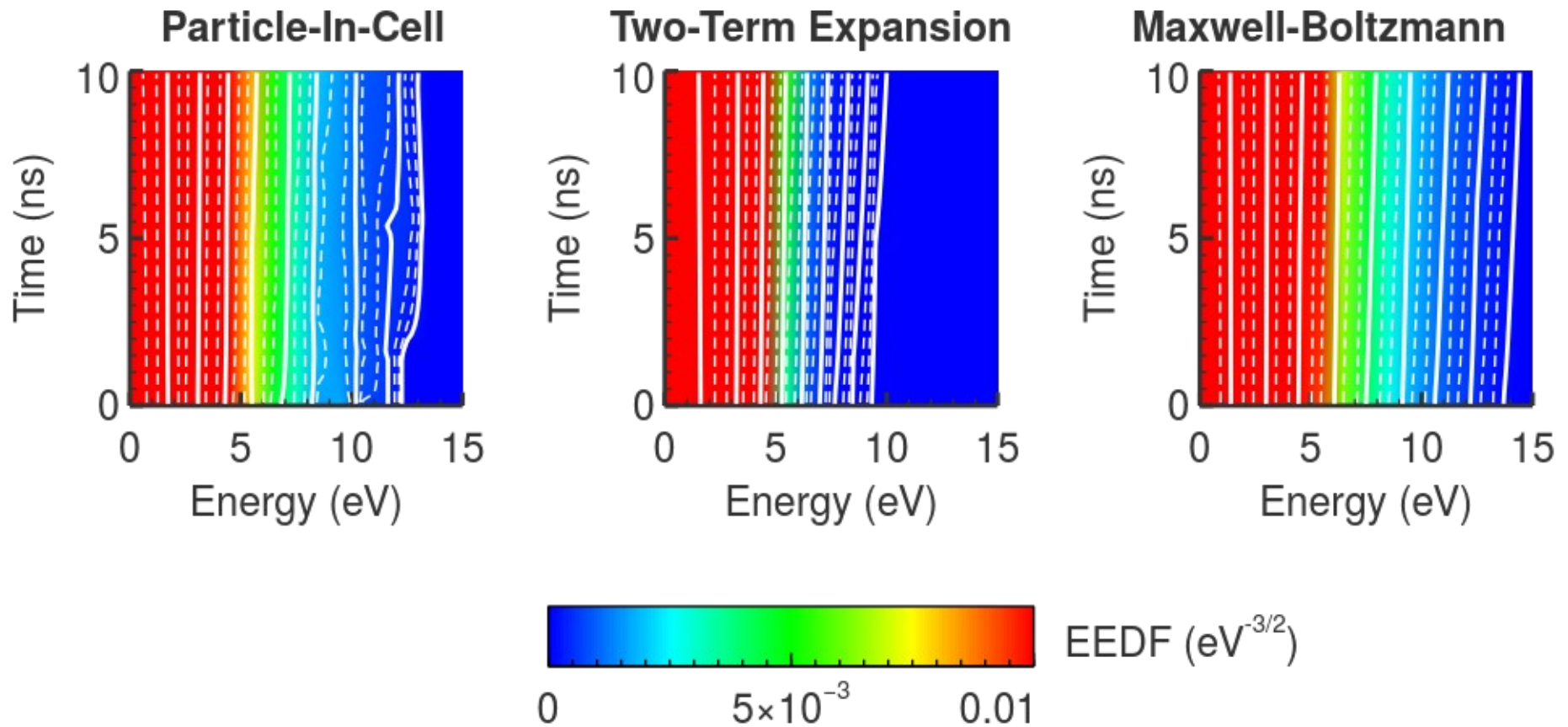
$$m_\alpha n_\alpha \left[ \frac{\partial \vec{u}_\alpha}{\partial t} + (\vec{u}_\alpha \cdot \nabla) \vec{u}_\alpha \right] = q_\alpha n_\alpha (\vec{E} + \vec{u}_\alpha \times \vec{B}) - \nabla \cdot \vec{\Pi} + \vec{f}|_{\text{coll}}$$

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

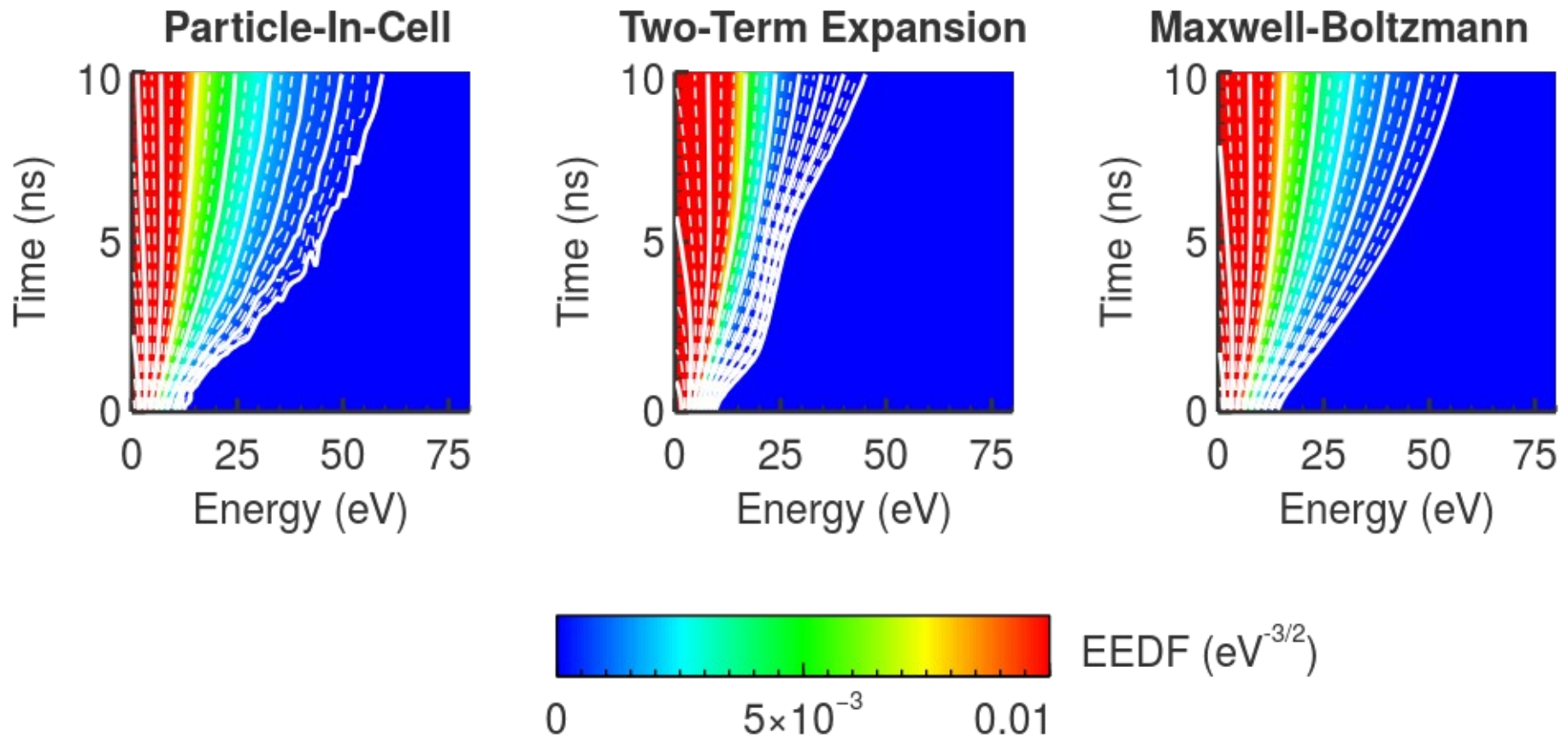
$$\frac{\partial}{\partial t} \left( \frac{3}{2} p_\alpha \right) + \nabla \cdot \frac{3}{2} (p_\alpha \vec{u}_\alpha) + p_\alpha \nabla \cdot \vec{u}_\alpha + \nabla \cdot \vec{q}_\alpha = \frac{\partial}{\partial t} \left( \frac{3}{2} p_\alpha \right) \Big|_{\text{coll}}$$

- 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

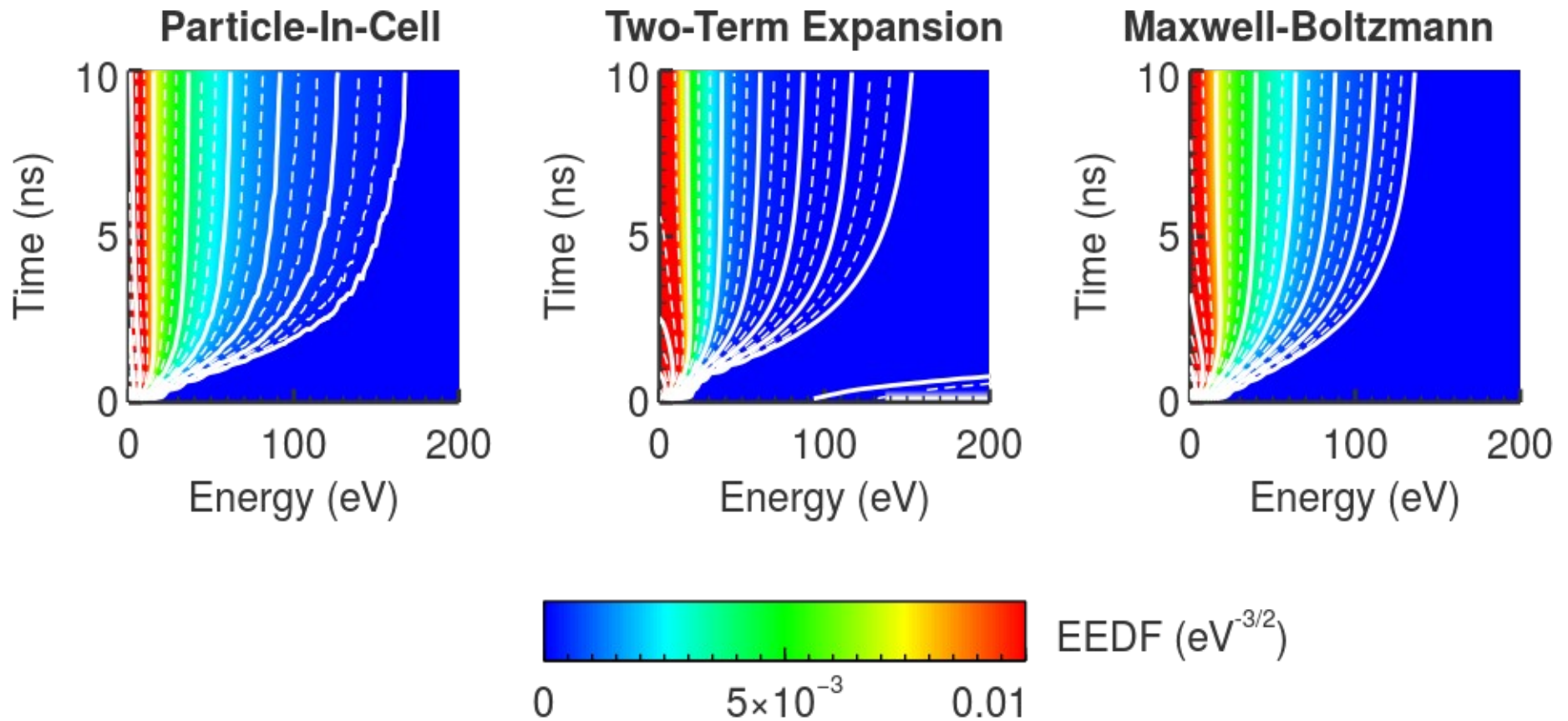
- Must assume an EEDF for reaction rates
- Baseline: 0D particle-in-cell (XPDP1)
- Two approaches considered:
  - Maxwell-Boltzmann distribution
  - Solutions of two-term expansion of Boltzmann equation (BOLSIG+)



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



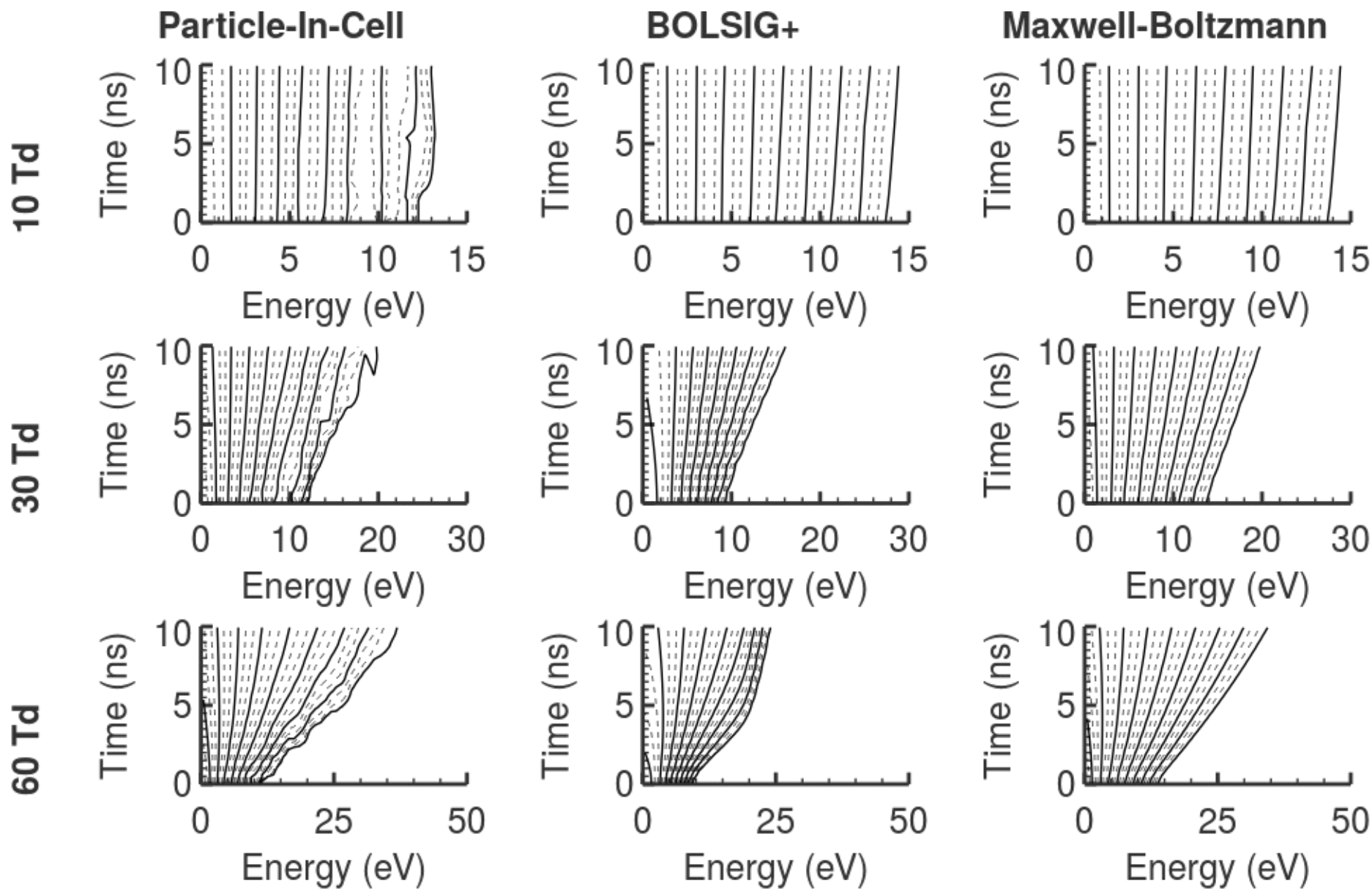
- Unexpected contour variations in two-term expansion solutions
- Better agreement between Maxwell-Boltzmann and particle-in-cell

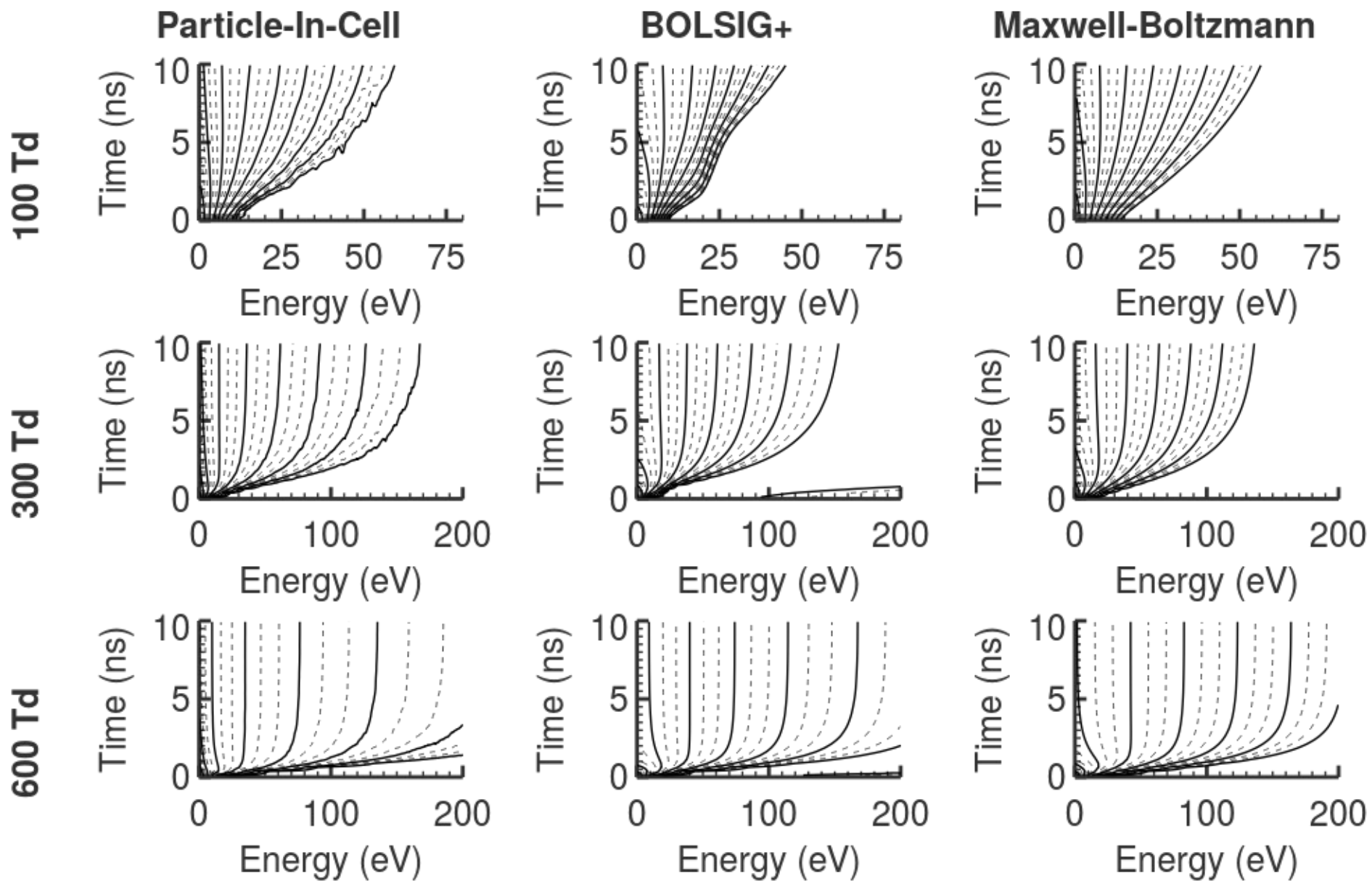


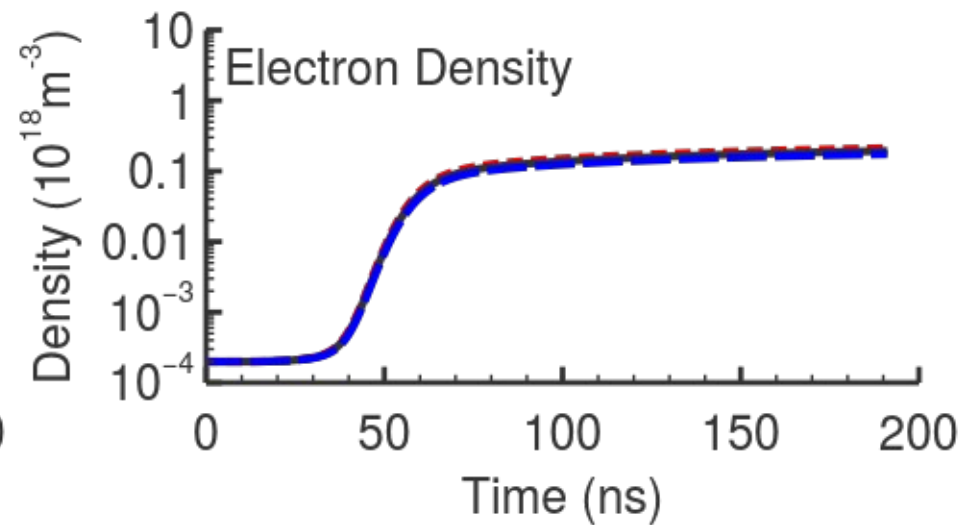
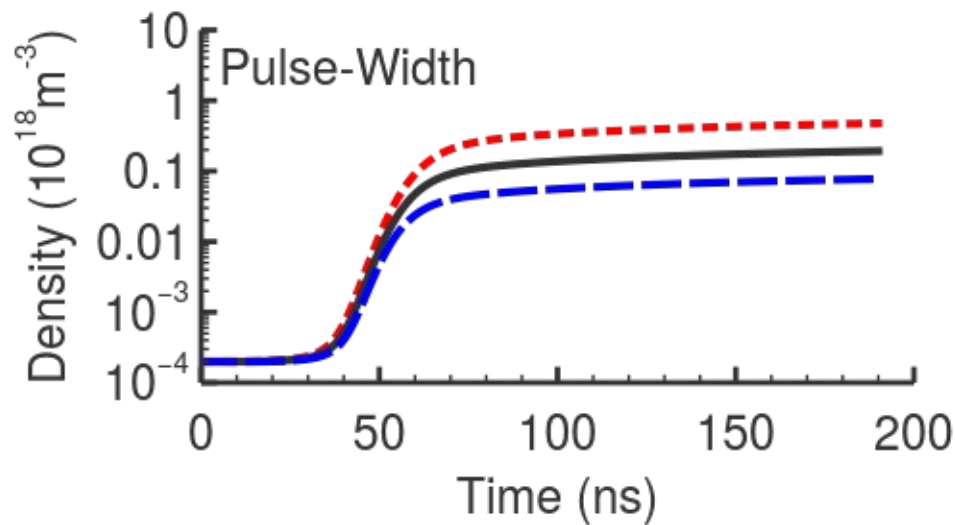
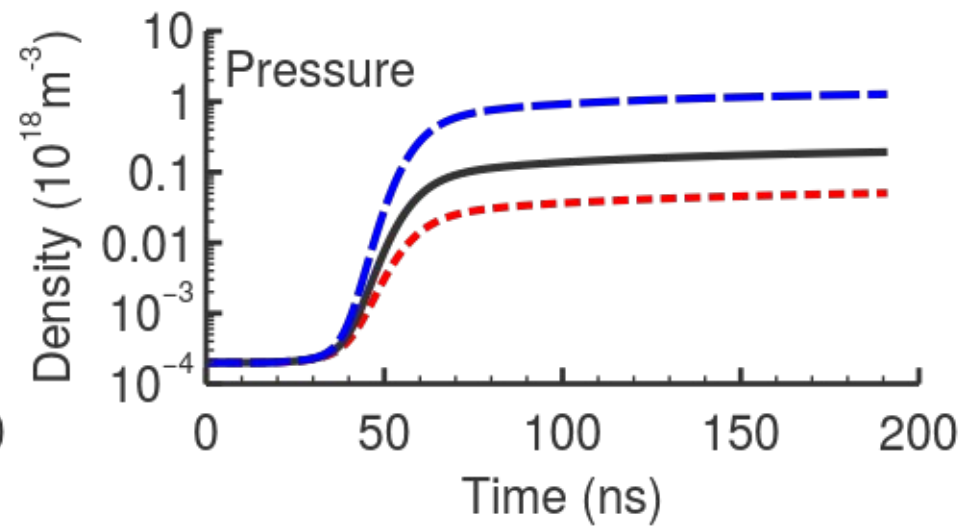
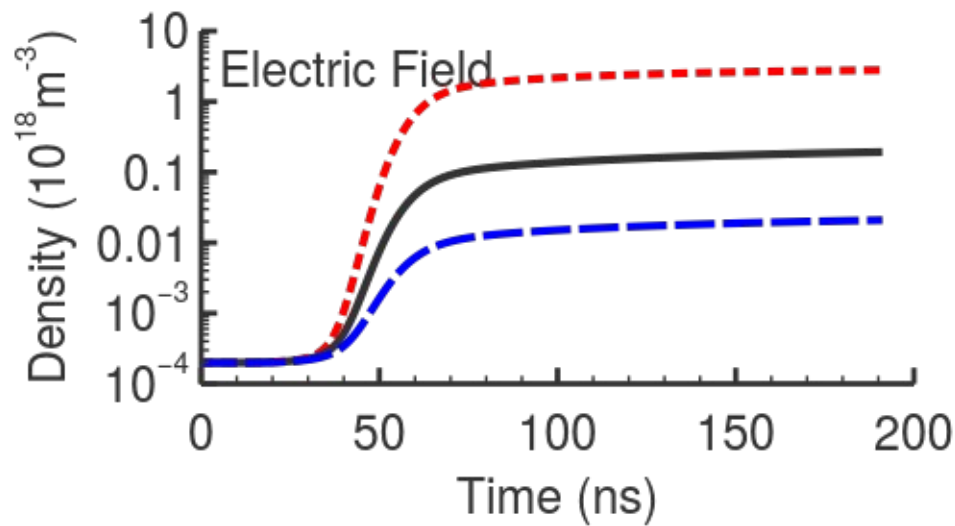
- Particle-in-cell: more 100 eV+ electrons than other results
- Two-term expansion solution best match this behavior
- Beginning of beam-like behavior?

- $< 100$  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

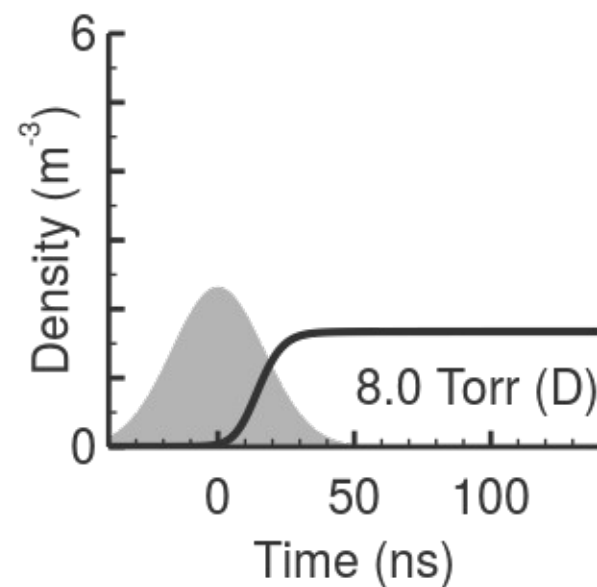
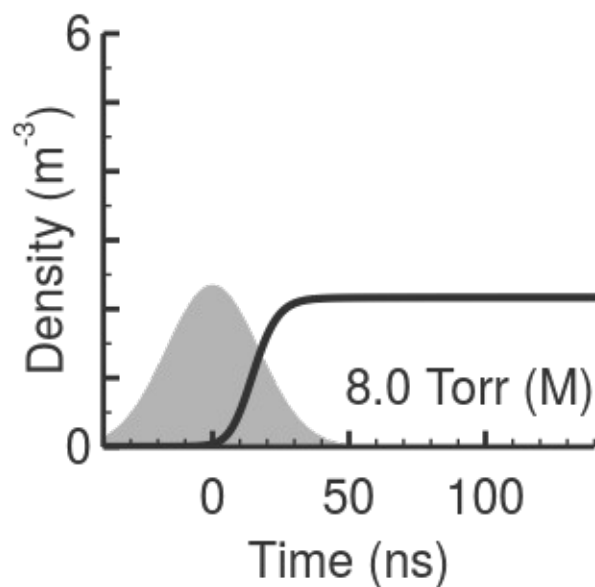
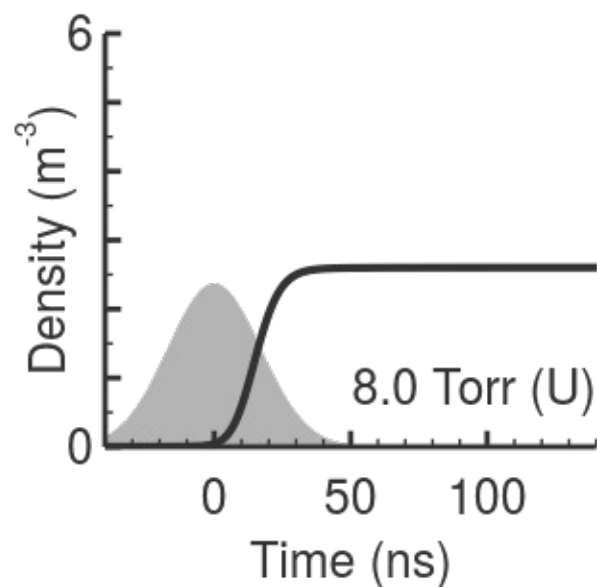
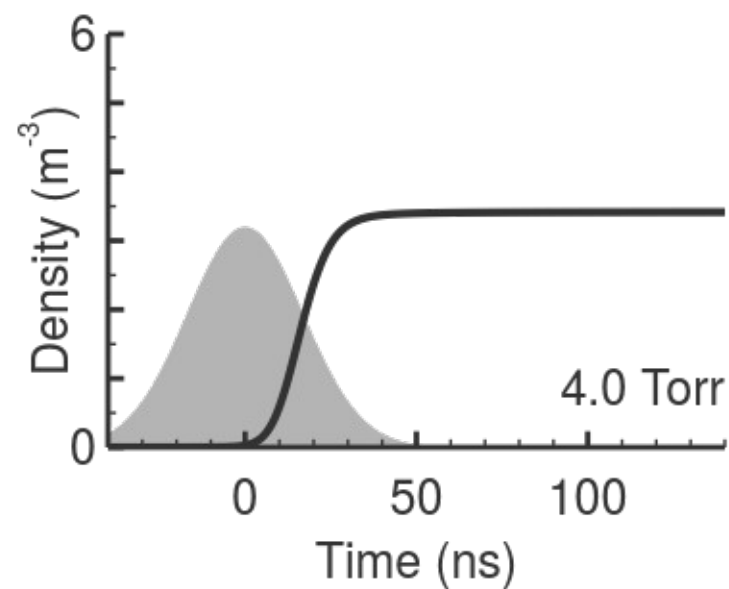
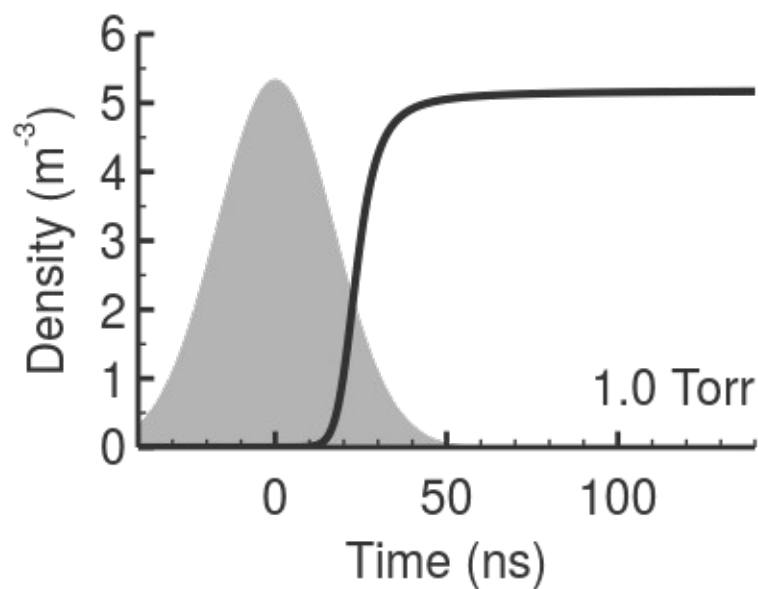




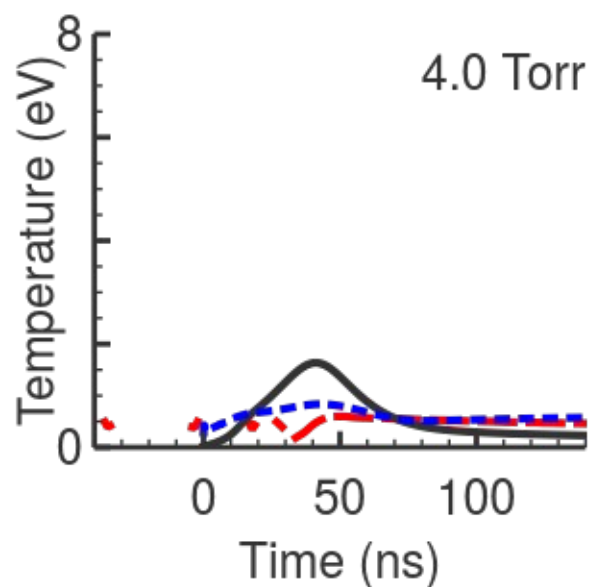
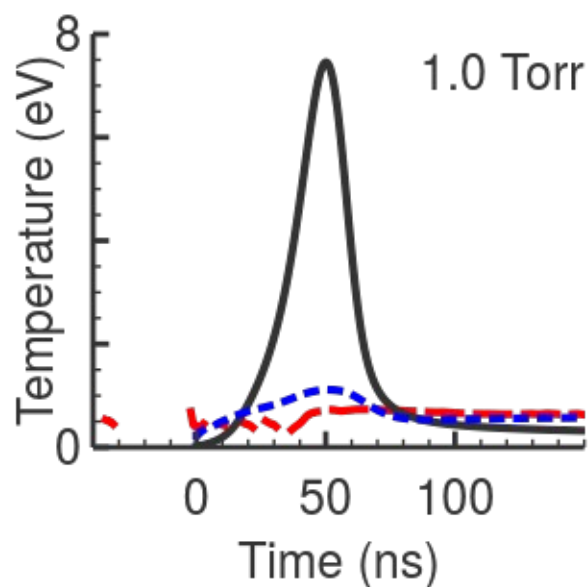




—  $\Delta = -10\%$     —  $\Delta = 0\%$     - -  $\Delta = +10\%$



- 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



--- Simulated Emissions  
— Calculated (x 0.1)  
--- Measured Emissions

