Spectroscopic Investigation of a Repetitively-Pulsed Nanosecond Discharge

by

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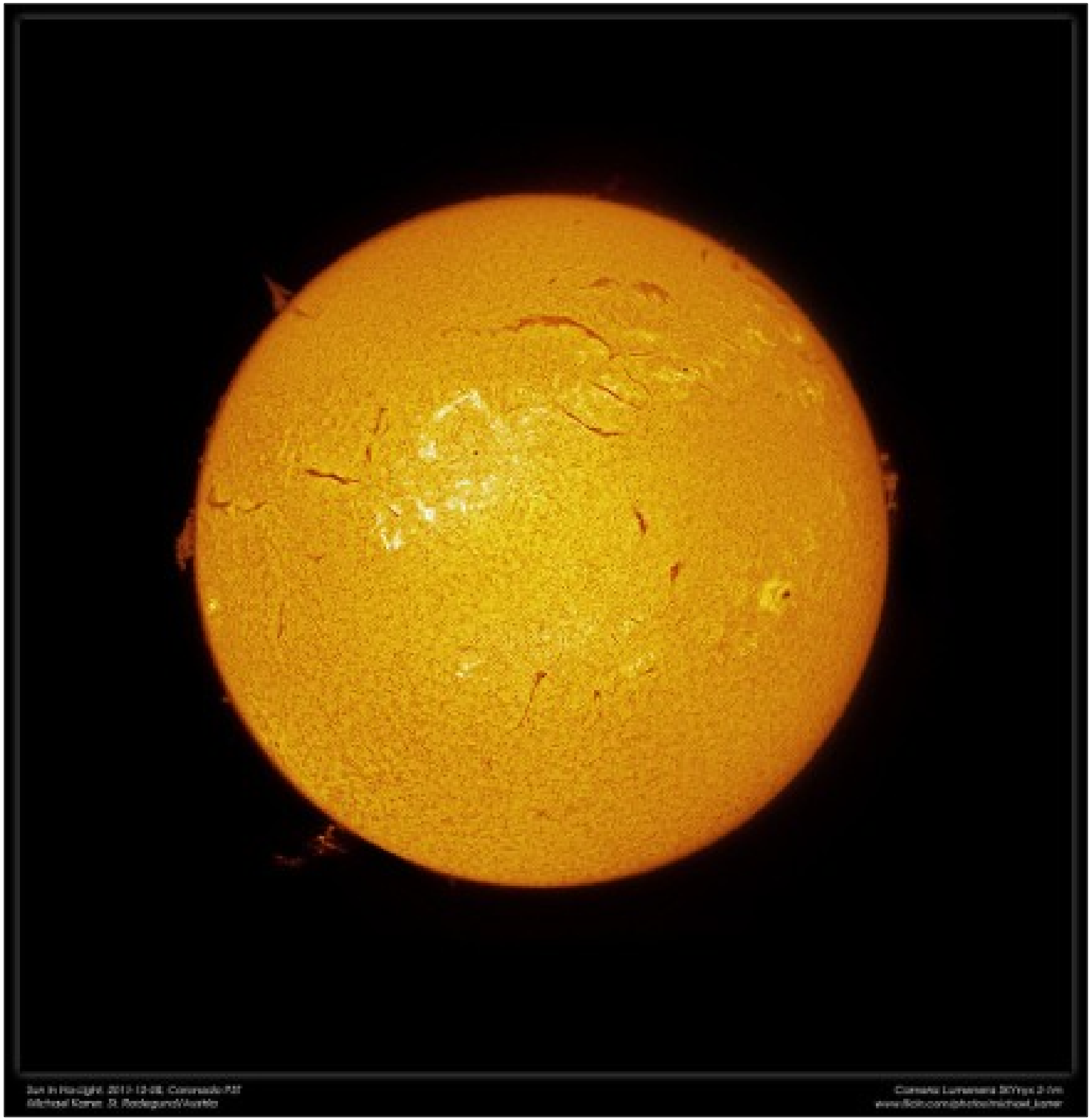
- Introduction
- Pulsed plasmas and outstanding questions
- Metastable measurements
- Global model
- Emission measurements
- Conclusions

Introduction



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

What Is a Plasma? (Sun)

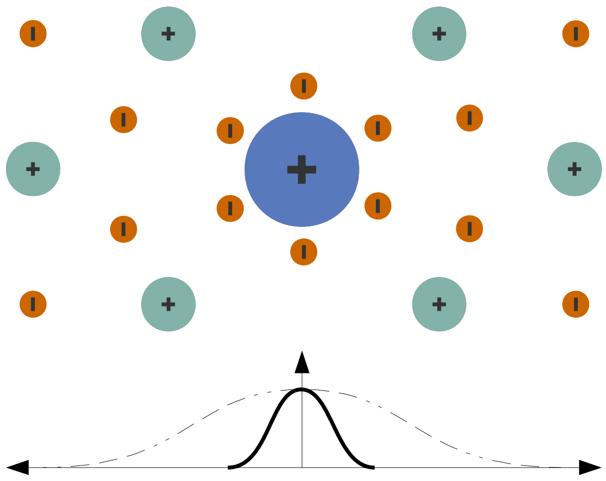
Hunt, P. "Lightning." URL: http://www.flickr.com/photos/michael_karrer/6487199145/, Accessed: Aug. 26, 2013.

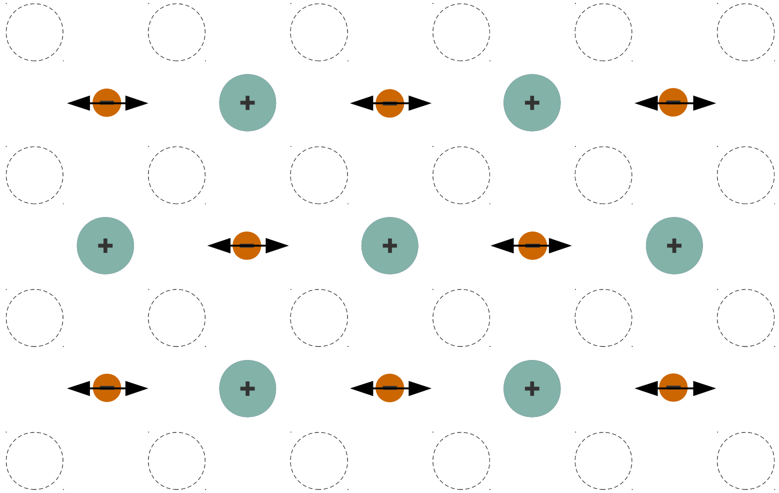
What Is a Plasma? (Lighting)

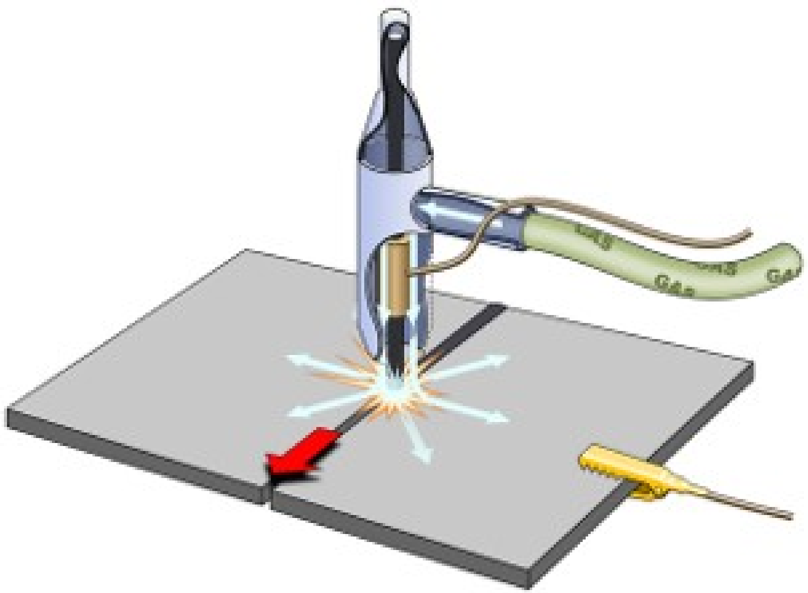
NASA, ESA, and the Hubble Heritage Team. "Star-Forming Region LH 95 in the Large Magellanic Cloud." URL: http://hubblesite.org/gallery/album/entire/pr2006055a/, Accessed: August 27, 2013.

What Is a Plasma? (Interstellar Gases)

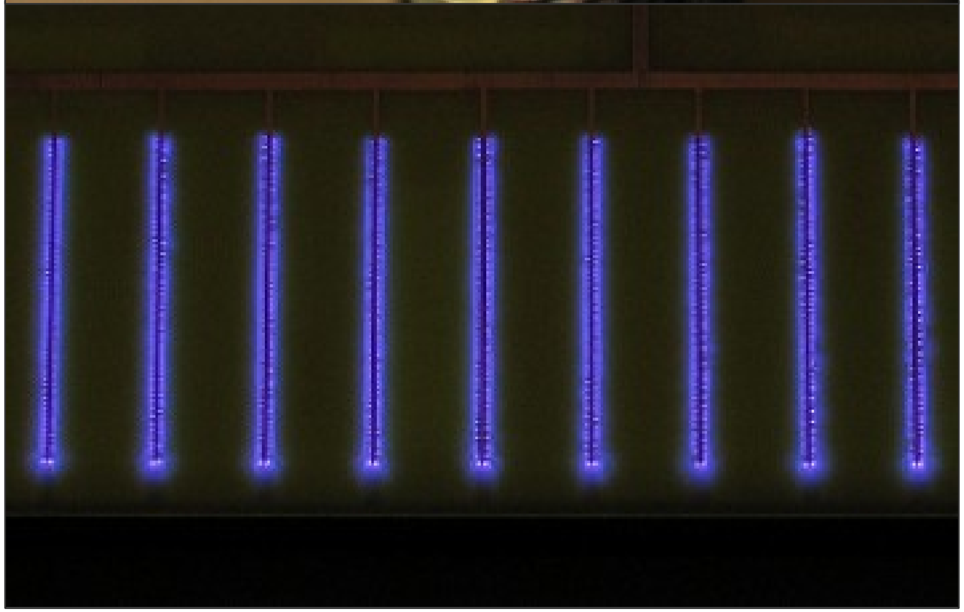
- Ionized gas
  - Neutral particles
  - Positive (and negative) ions
  - Electrons
- Exhibits large-scale electrostatic effects
  - Debye shielding
  - Electron oscillations







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, Accessed: August 27, 2013.
Schmid, M. “Wasabi-Chips (Migros).” URL: 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 gas flow for MHD generators and energy bypasses

- Sterilization of bacterial, viral, and chemical contaminants (e.g. MRSA or anthrax)

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.
parfe_alp, “Operation.” URL: http://www.flickr.com/photos/parfe/2239523465/, Accessed: August 27, 2013.

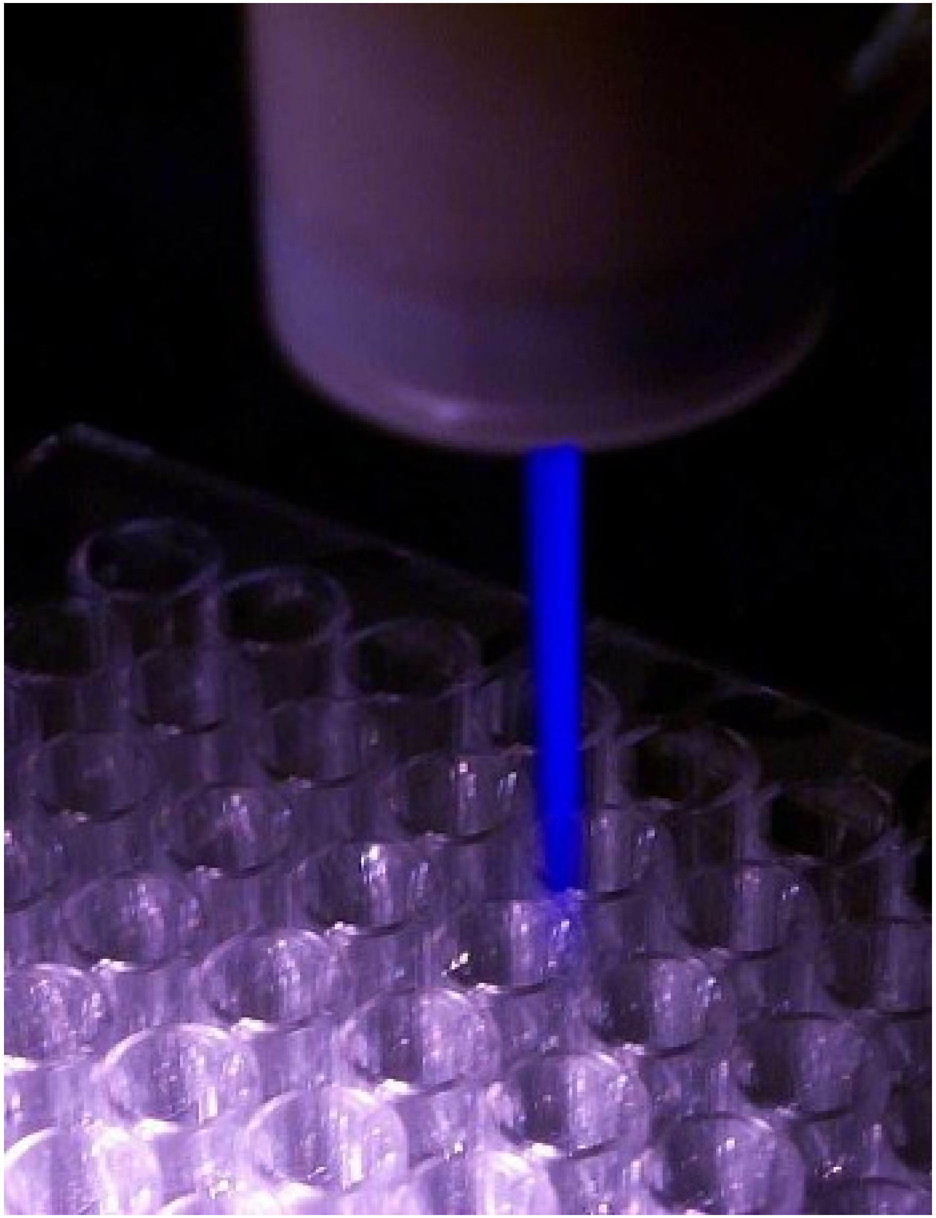
Atmospheric-Pressure Plasma Applications

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

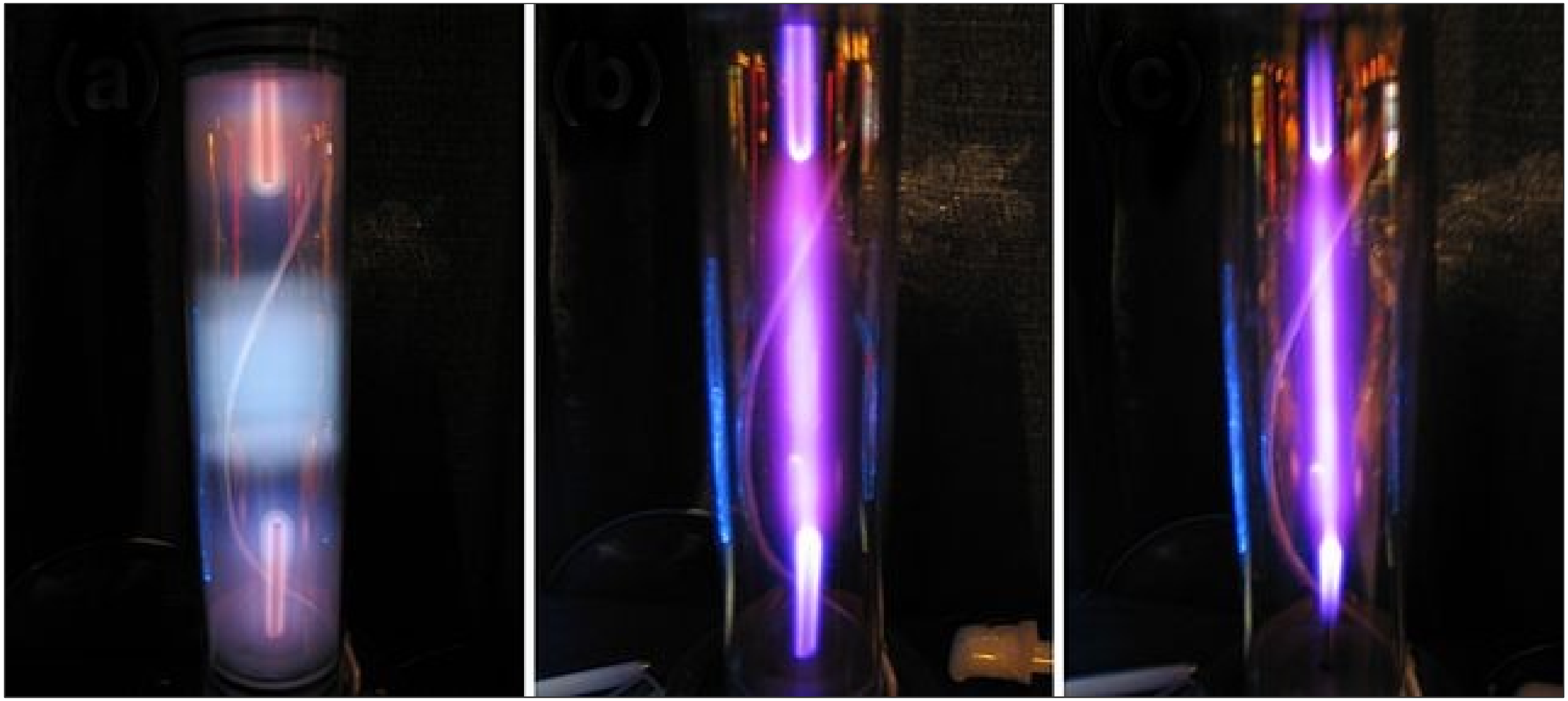
D-Kuru/Wikimedia Commons. "Afterglowing electrodes of an arc lamp." URL: https://commons.wikimedia.org/wiki/File:Arc_lamp-afterglow_2_PNr%C2%B00038.jpg, Accessed: August 28, 2013.

Equilibrium Plasmas

- Common in fluorescent lighting and material processing.
- Different temperatures for each species
  - Often, $T_{neutrals} \sim T_{ions} < T_{electrons}$
- Desirable for many high-pressure applications; high gas temperatures can cause damage



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

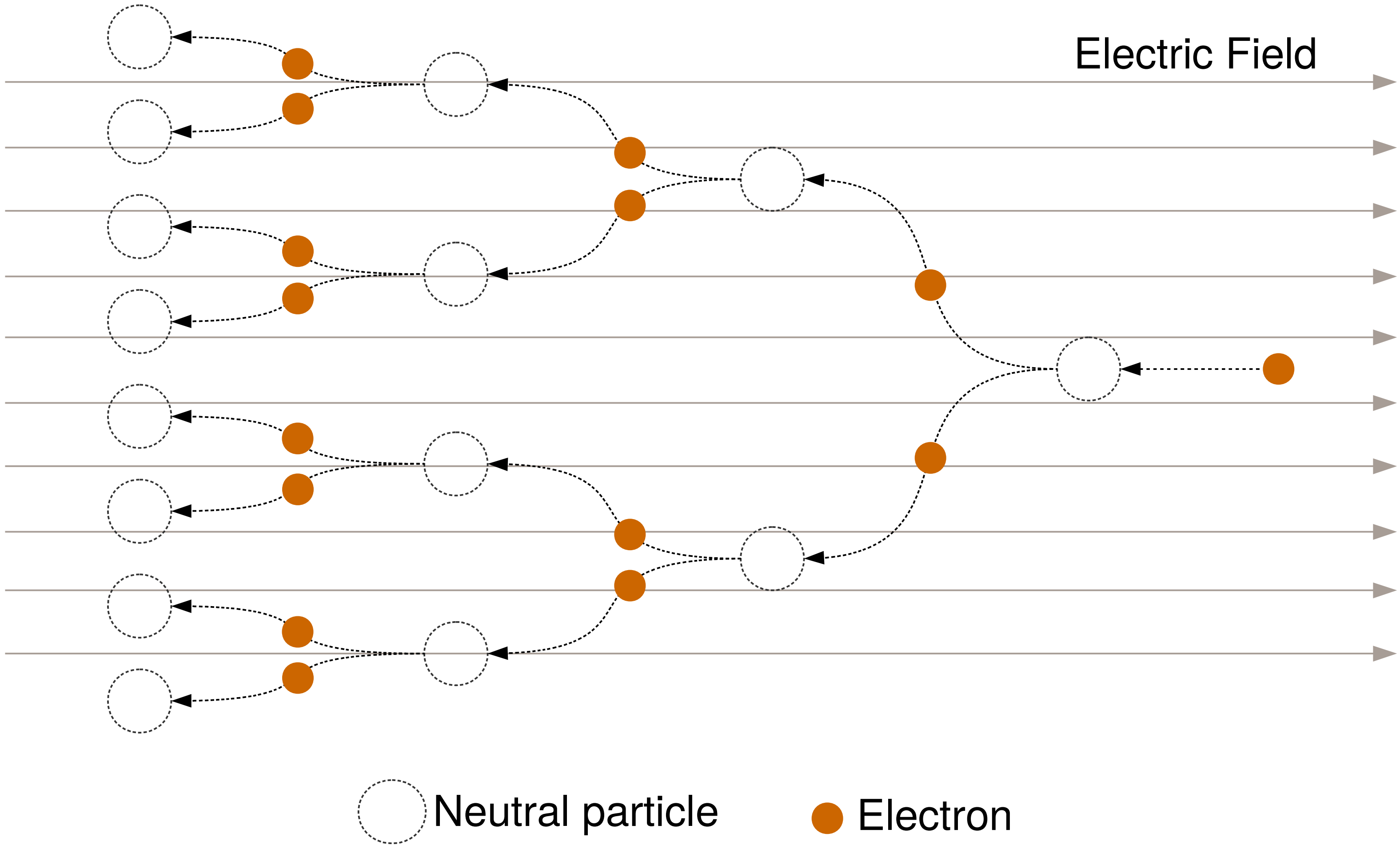


- Increase in pressure → tendency to equilibrate
- Related to ionization instability
- Prevented by careful control of power input
  - Charging of dielectric barriers
  - Pulsed discharges

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

Glow-to-Arc Transition

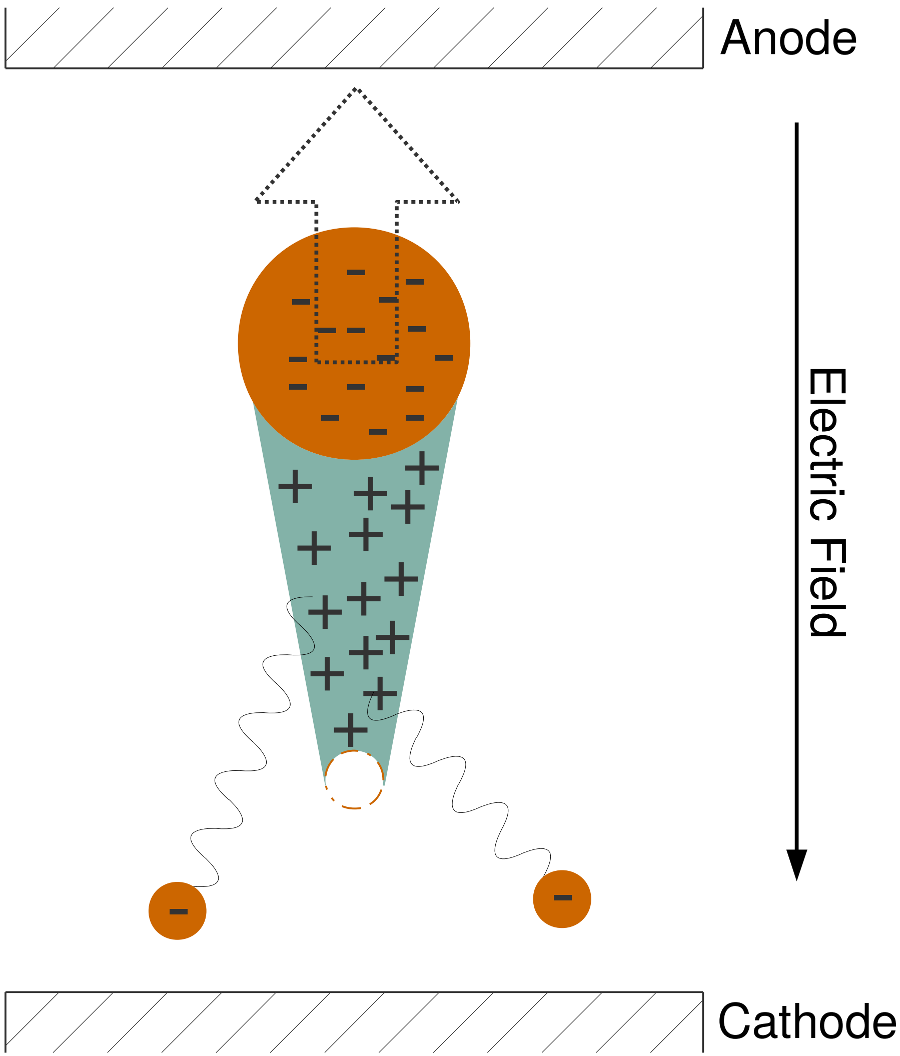
Pulsed Plasmas and Outstanding Questions



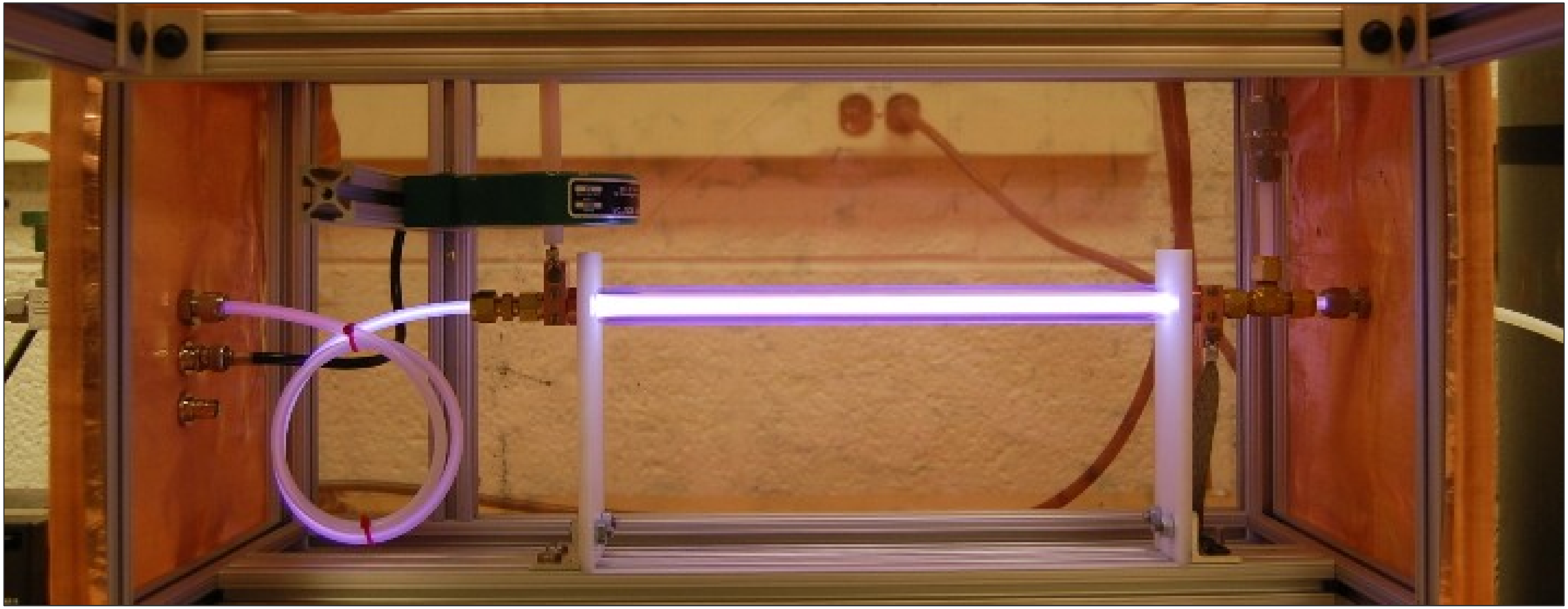
- Most pulsed plasmas initiated by electron avalanche
- Initial electron from cosmic rays, UV light, previous pulse, ...
- Sufficient electric field → exponential growth

Electron Avalanche

- In certain cases (Raether-Meek criterion), avalanche becomes streamer discharge
- Internal field of avalanche comparable to applied field
- Reduces energy transfer to streamer
- “Injected” electrons and photoionization can become important

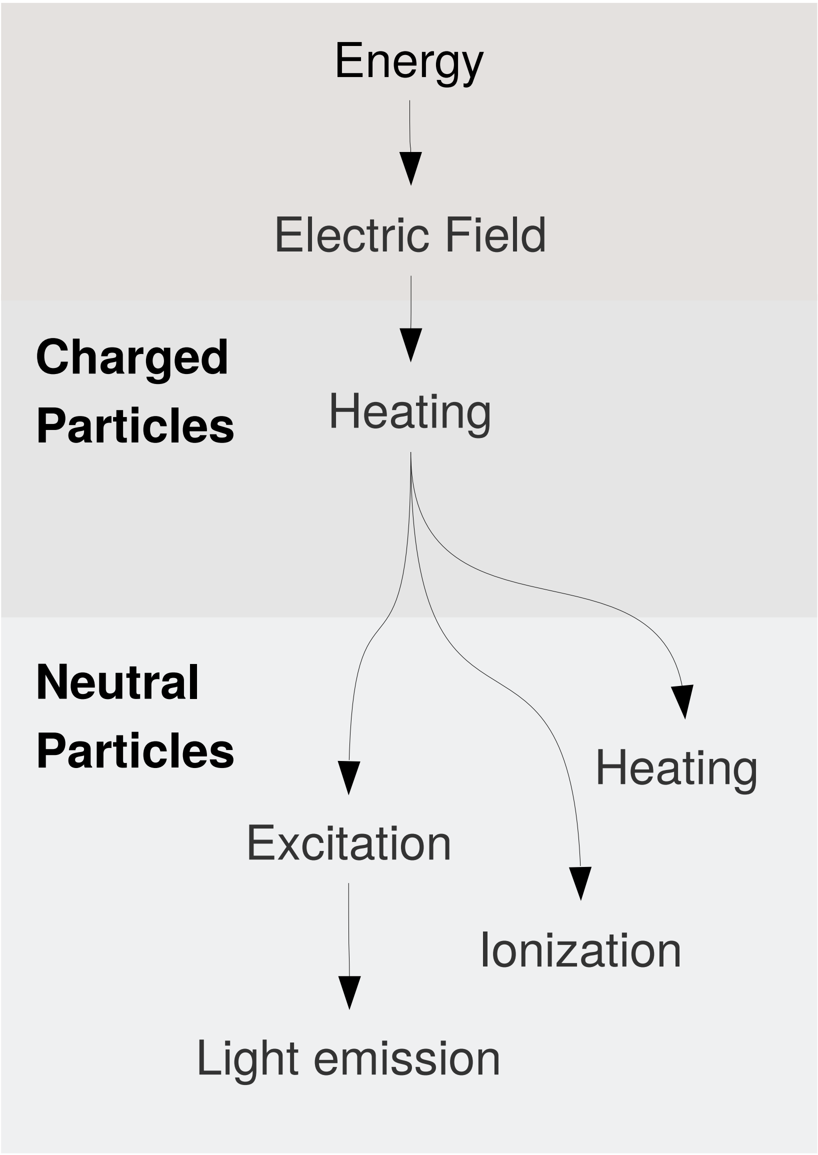


Streamer Discharge



- Repetitively pulsed nanosecond discharge (RPND)
  - Development involves significant space charge, similar to streamer
- Attractive for high-pressure applications
  - Uniform over large volumes (on the order of liters)
  - Little gas heating
- Pulse characteristics
  - Pulse-widths: 1 – 100 ns
  - Voltages: 1 – 100+ kV
  - Repetition rates: 1 – 100 kHz

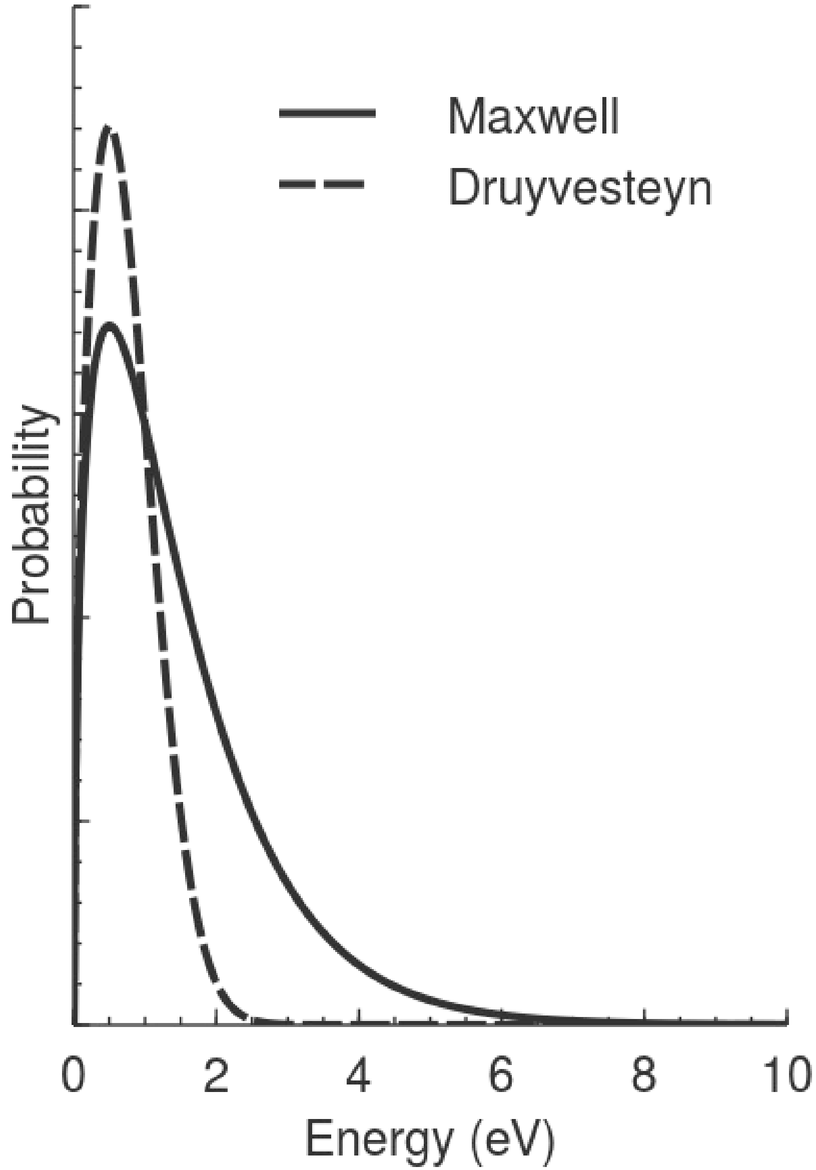
Repetitively-Pulsed Nanosecond Discharge

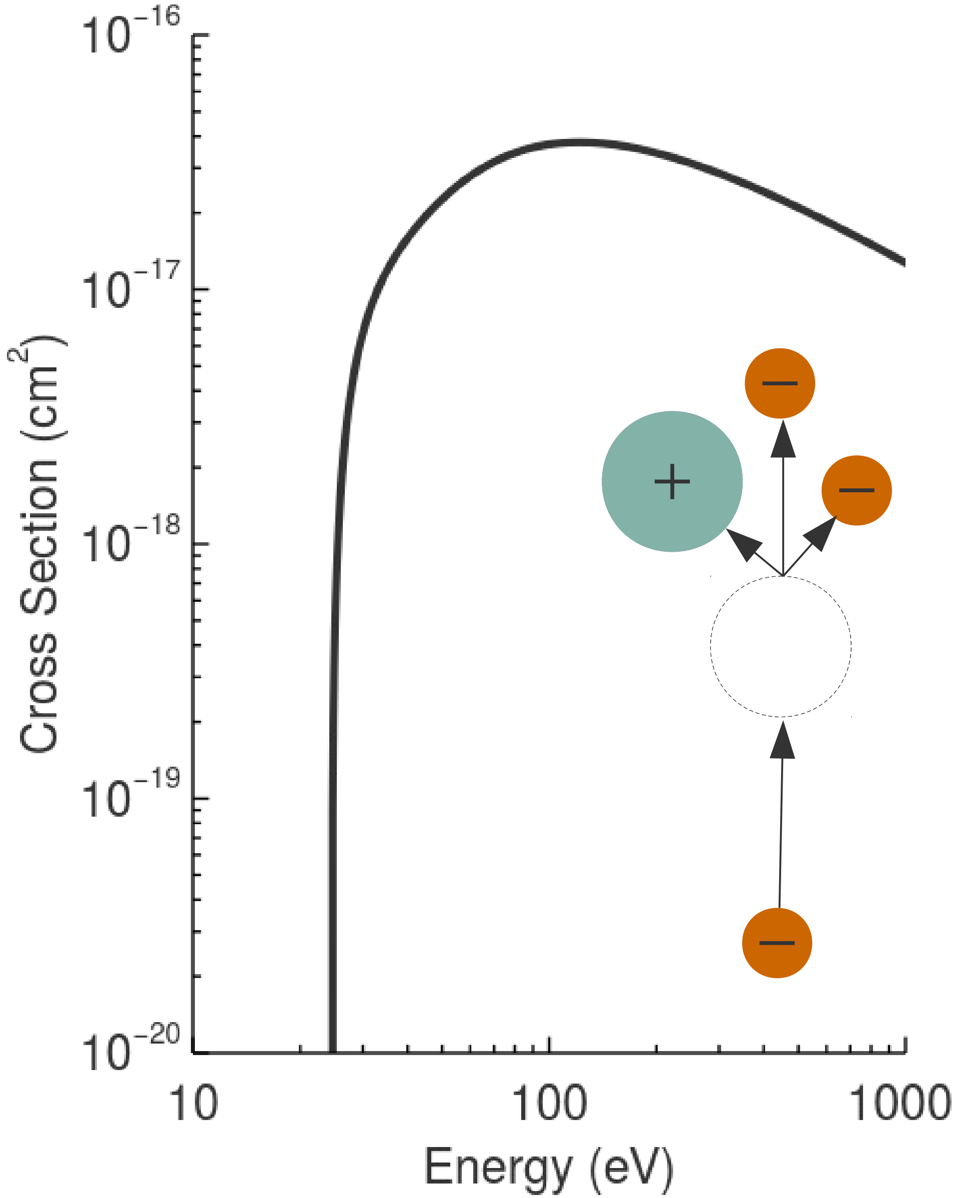


- Energy added to charged particles via electric field
- Some energy heats electrons
- Remainder enters neutral particles through collisions
- How is energy divided?
  - Important as neutral particle products are often desirable for applications

Questions: Energy Coupling

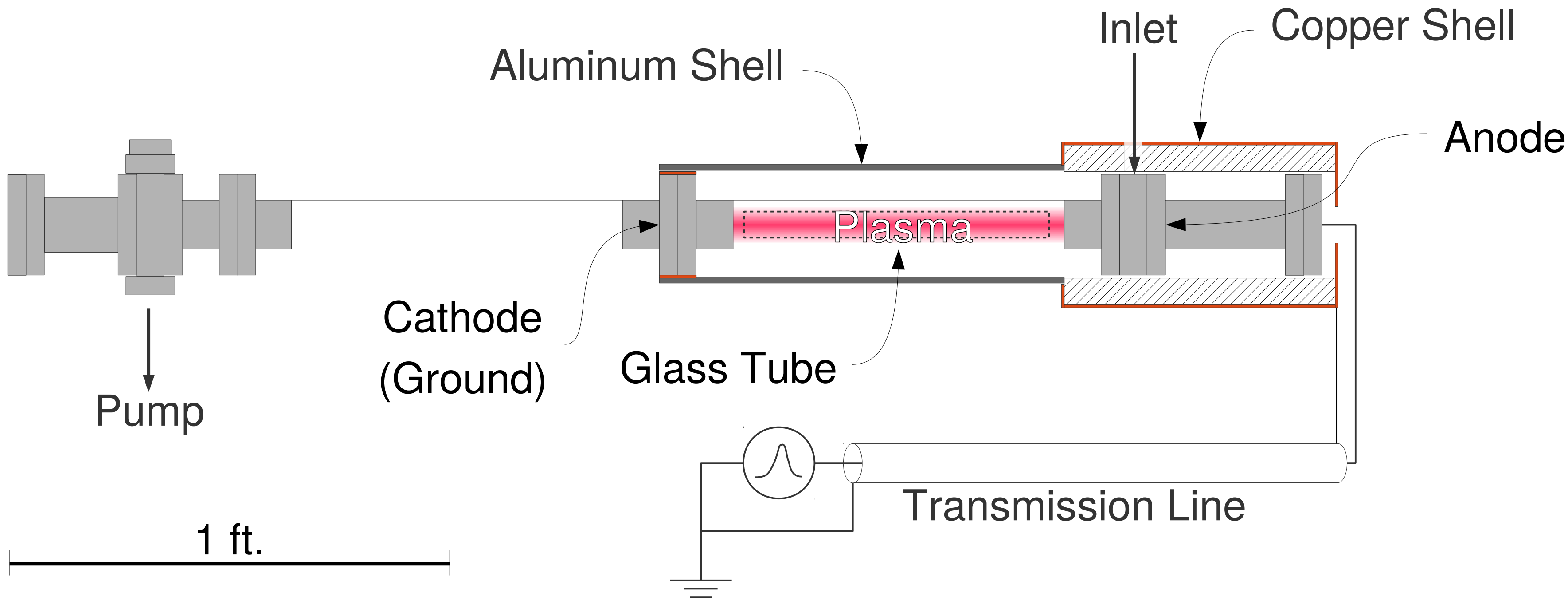
- An EEDF is a continuous probability distribution for electron energies
- With assumptions can be obtained
  - Analytically
  - Numerically
- However, assumptions may not be valid for RPND





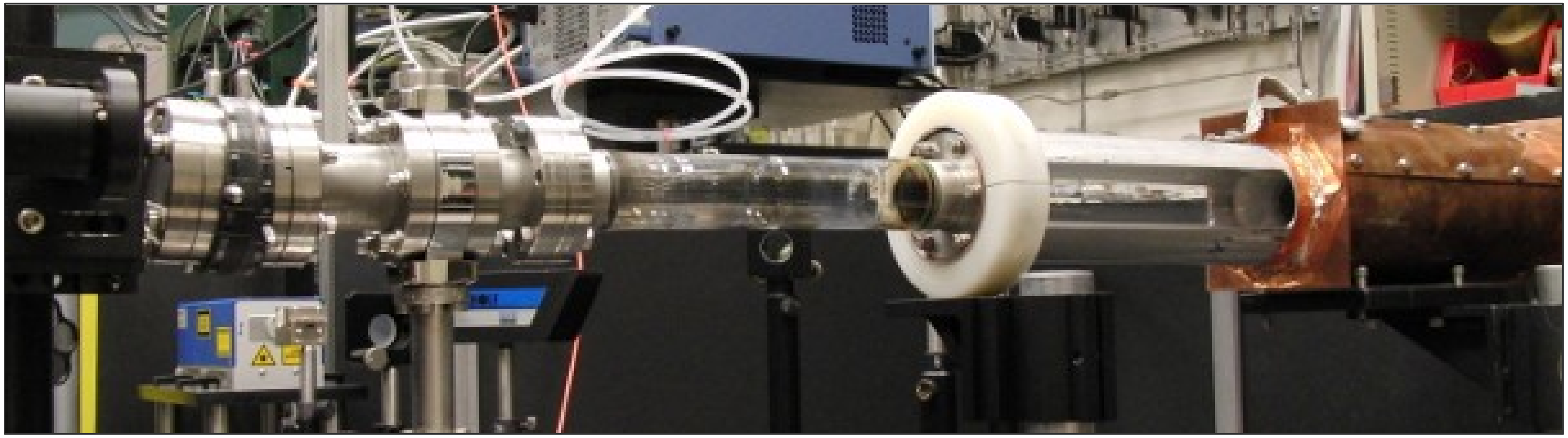
- Reactions in plasma determined by cross section and EEDF
- Cross section expresses probability of specific reactions
  - Excitation
  - Ionization
  - Dissociation
  - ...
- Must know EEDF to determine reaction rates

Experimental Setup



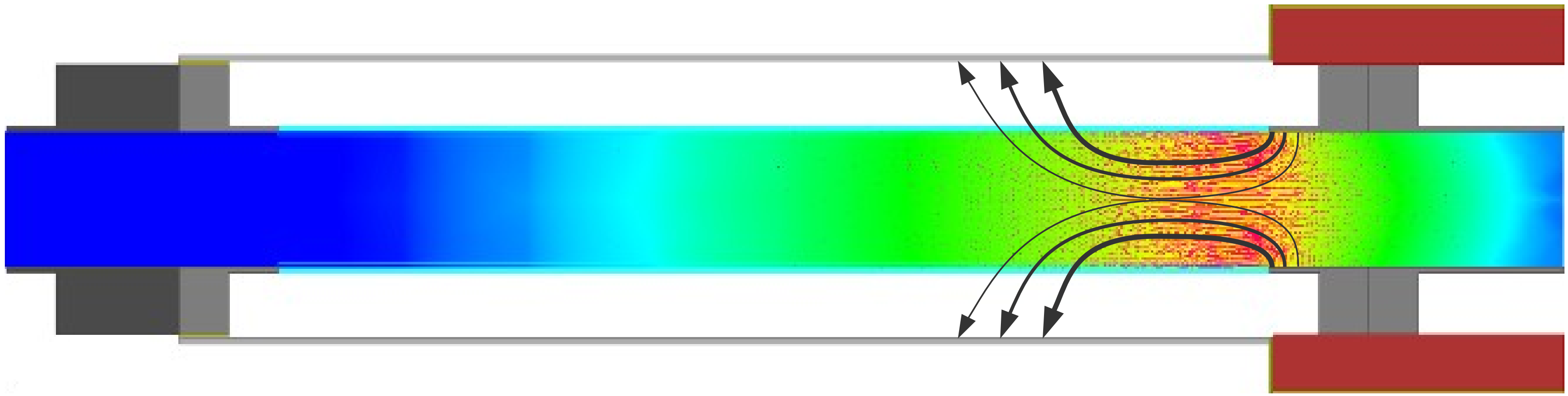
Coaxial-type geometry

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

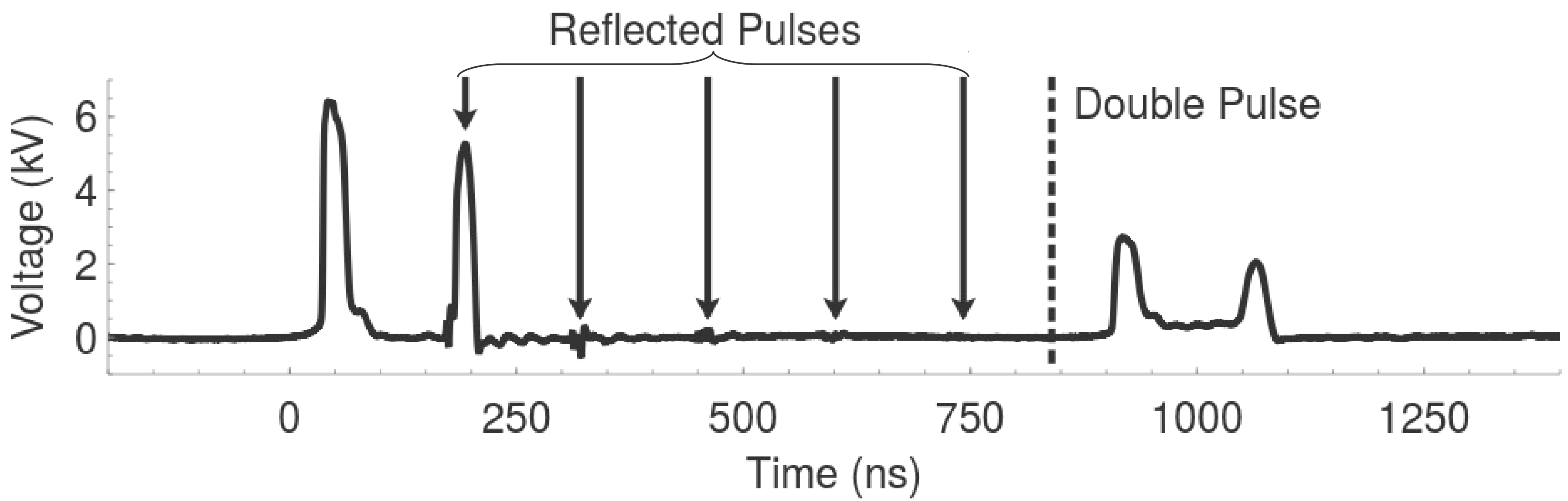


- Ultra-high purity helium: 0.3 – 16.0 Torr
- Rare gas discharges → interesting properties
  - Minimal gas heating
  - UV emissions
  - Data available to create detailed population kinetics model
- Voltage pulses: 25 ns FWHM, +6.4 kV, 1.0 kHz

Experimental Setup



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



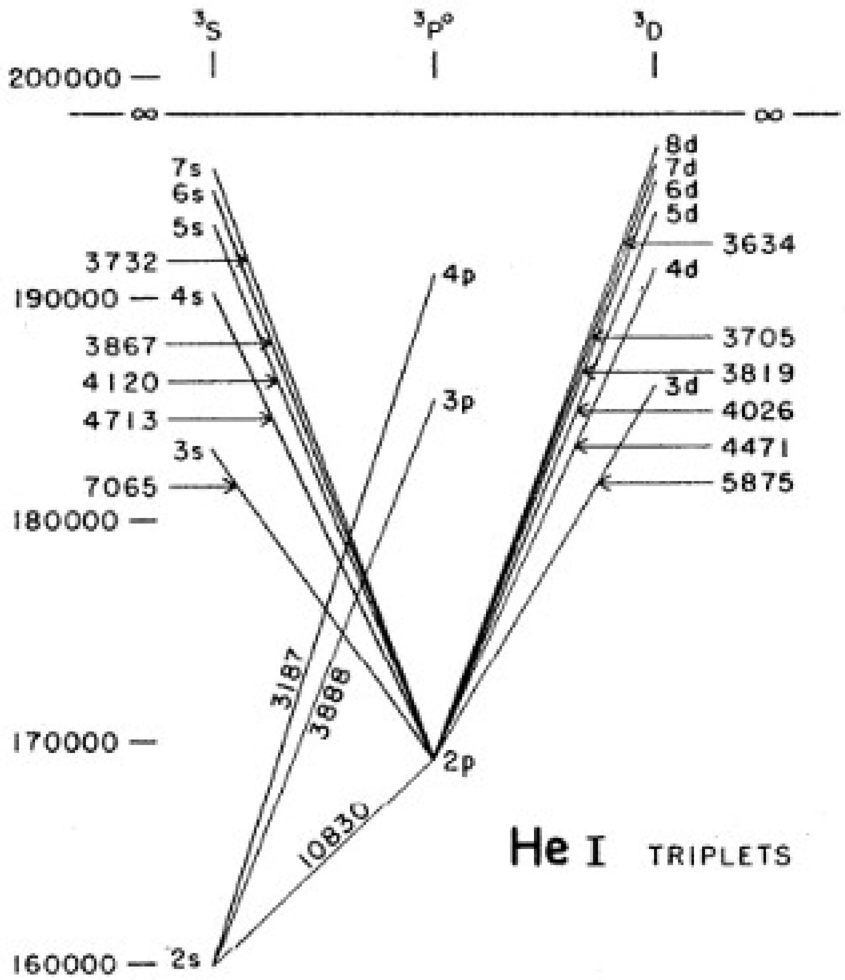
- Impedance mismatch causes numerous reflections between power supply and anode
  - Additional opportunity for energy absorption
- 13 m transmission line used to isolate incident and reflected pulses in time
- Double-pulsing by power supply observed after 800 ns

Input Waveform

Metastable Measurements

- Physical probes
  - Perturb plasma
  - Not fast enough (fastest Langmuir probes: 11 MHz)
  - Suggests an <u>optical</u> approach
- Electrons
  - No detectable emissions (i.e. bremsstrahlung or synchrotron)
  - Insufficient density for laser scattering
  - Suggests <u>atomic</u> measurements

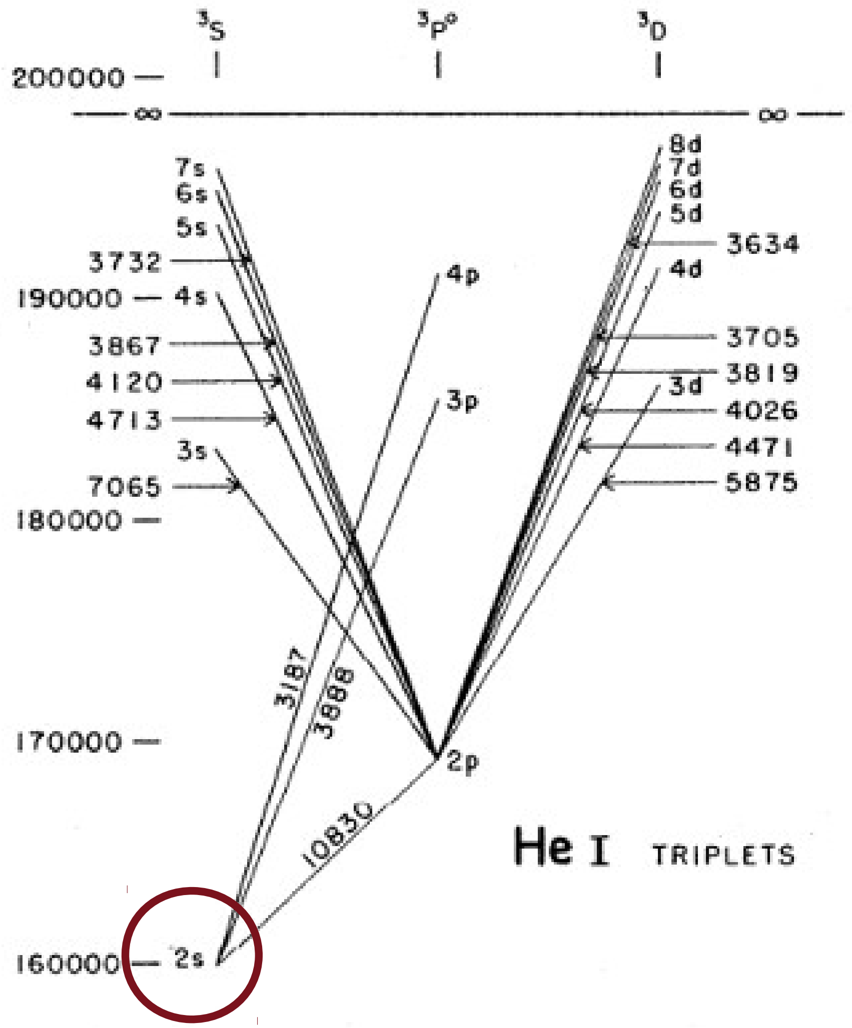
→ <u>Optical measurements of atomic states</u>



- Atomic states can absorb light with specific wavelengths
  - Light must be close to transition energy with another state
- Consider volume of gas with a mixture of states
  - Light incident on volume and matching specific transition would be absorbed
  - Absorption proportional to density of initial atomic state
- Higher temporal resolution than Langmuir prove, photodiodes available with 1+ GHz bandwidth

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

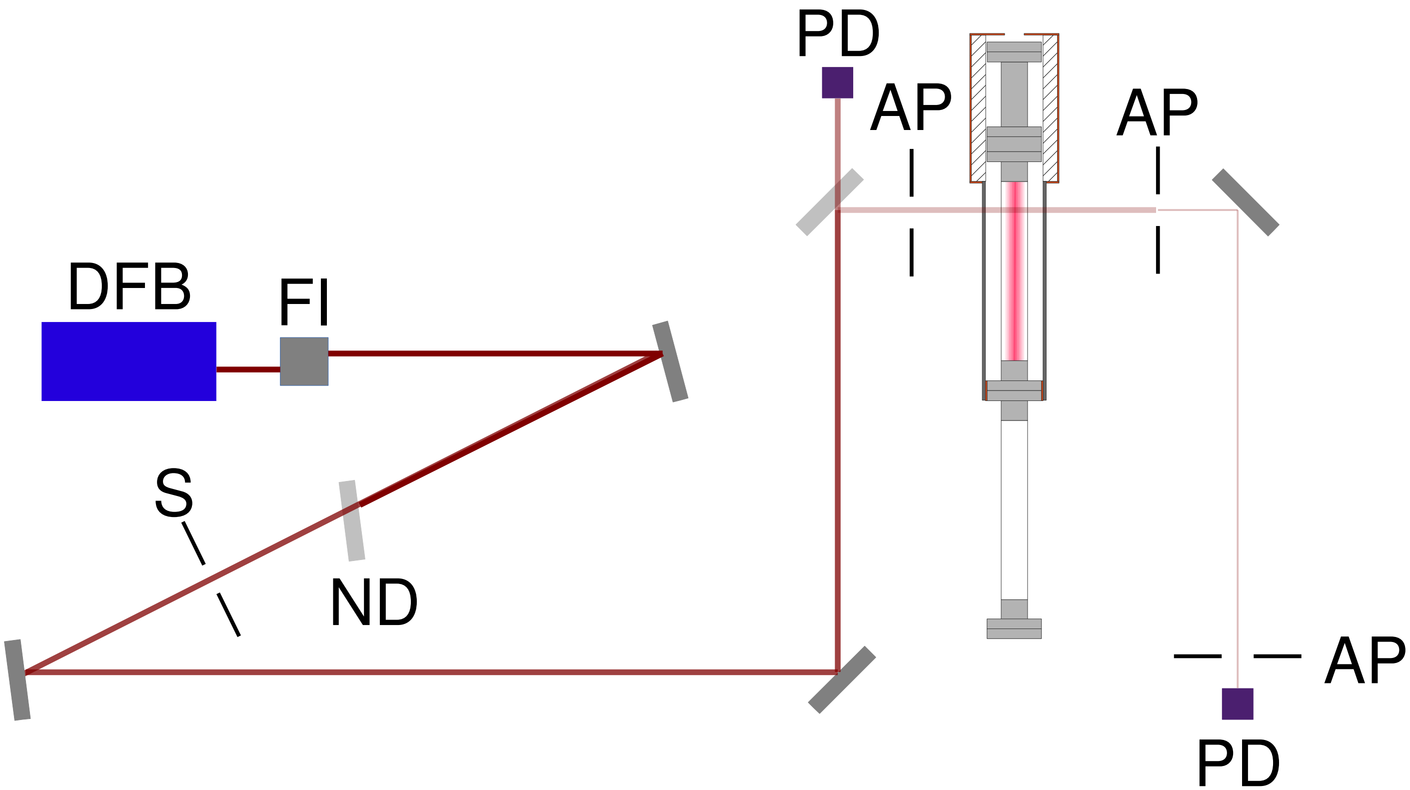
Absorption Spectroscopy



- Triplet metastable chosen for study
- Lowest-energy excited state
- Cannot spontaneously transition to ground state (spin-forbidden) → long-lived
- Reservoir for energy
- Can cause excitation and ionization long after applied voltage pulse

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

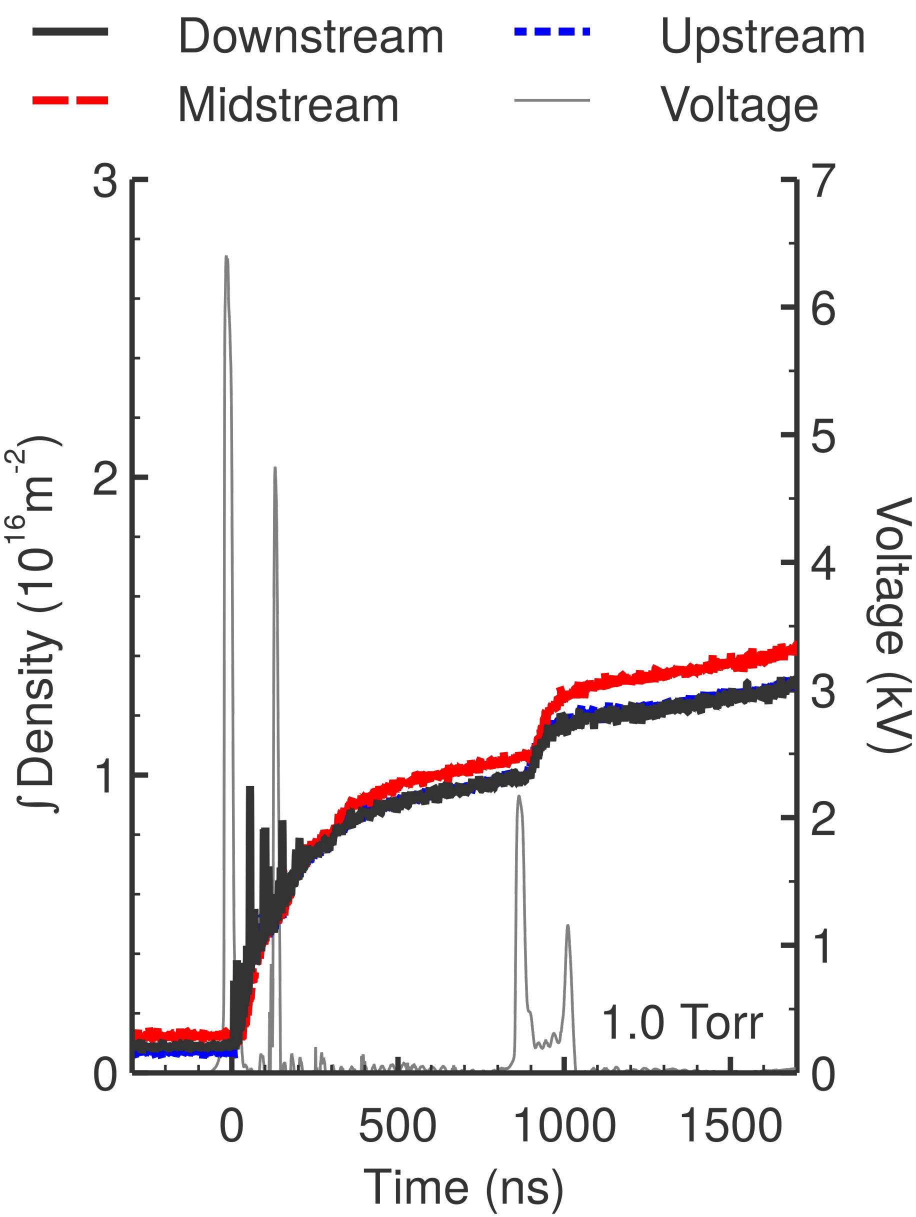
Triplet Metastables



- $2^3S \rightarrow 2^3P_0$ transition (1083 nm), across diameter of discharge
- Pressures: 0.3, 0.5, 1.0, 2.0, 3.0, 4.0, 8.0, and 16.0 Torr
  - Covered transition between low and high pressure behavior for metastable losses
- Locations: 5.1 cm (Upstream), 12.7 cm (Midstream), and 20.3 cm (Downstream)
  - Allowed observation of axial variations in plasma formation

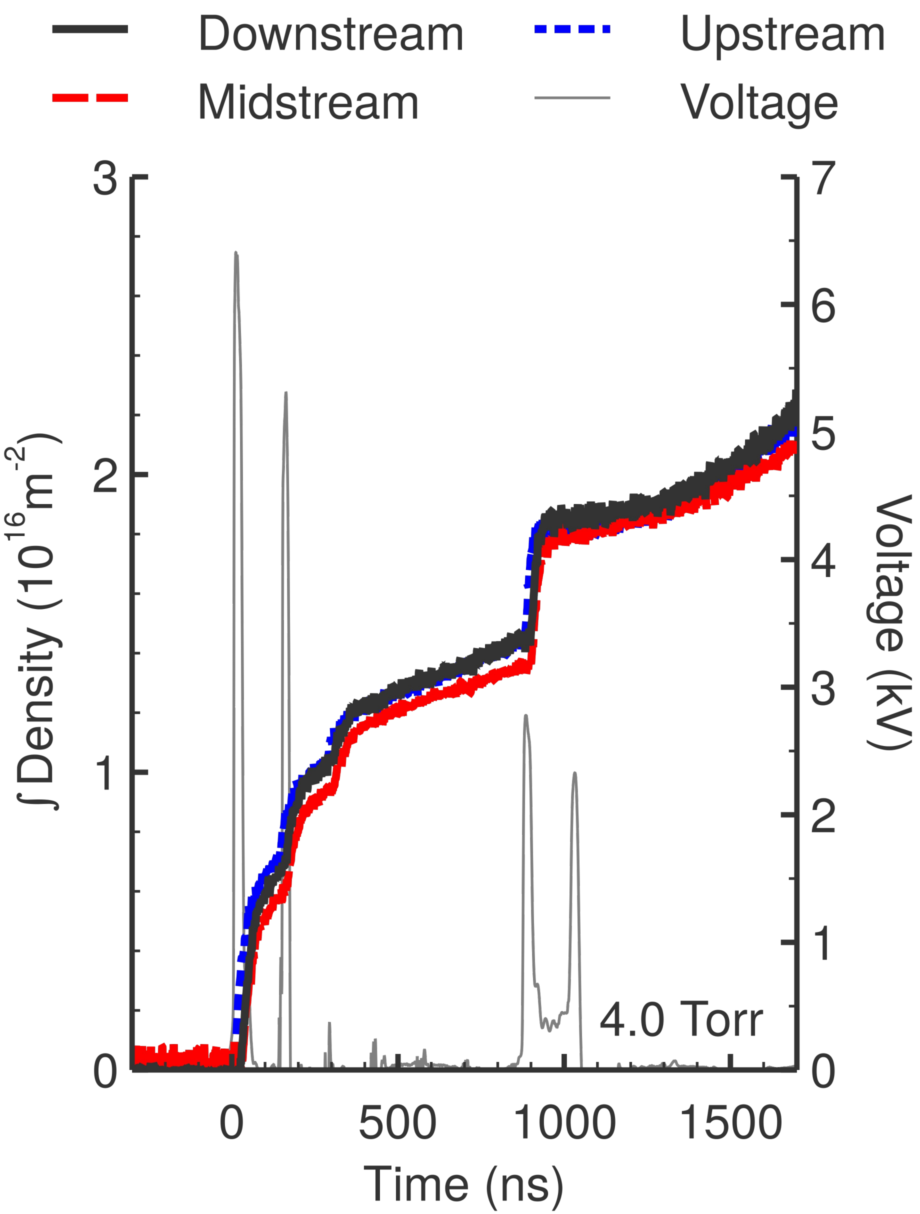
Laser-Absorption Spectroscopy Setup

- Line-integrated densities (will not assume radial distribution)
- Observed persistent metastable population between pulses
- Electrical noise present in measurements near anode
- No significant changes with location relative to anode

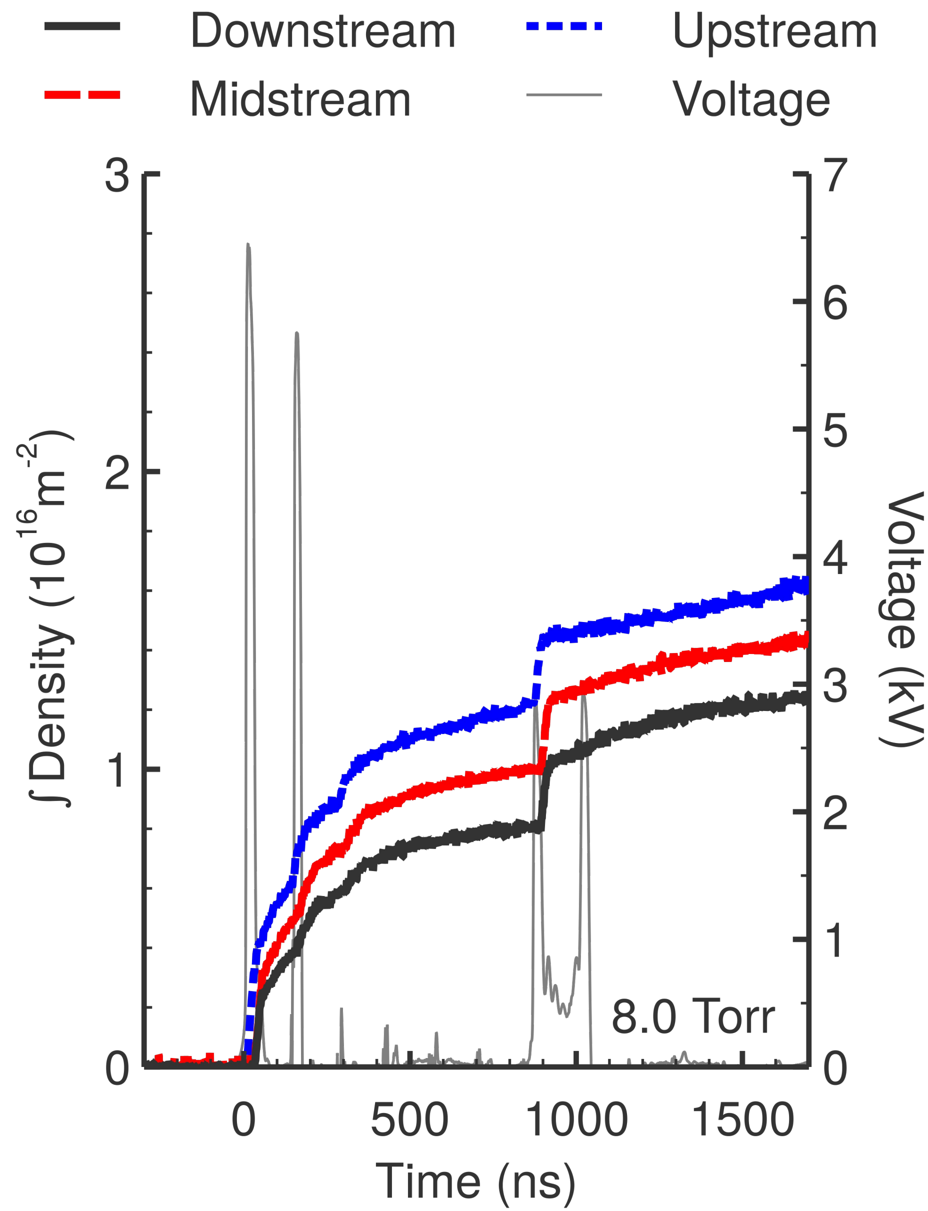


Metastable Densities (Low Pressure)

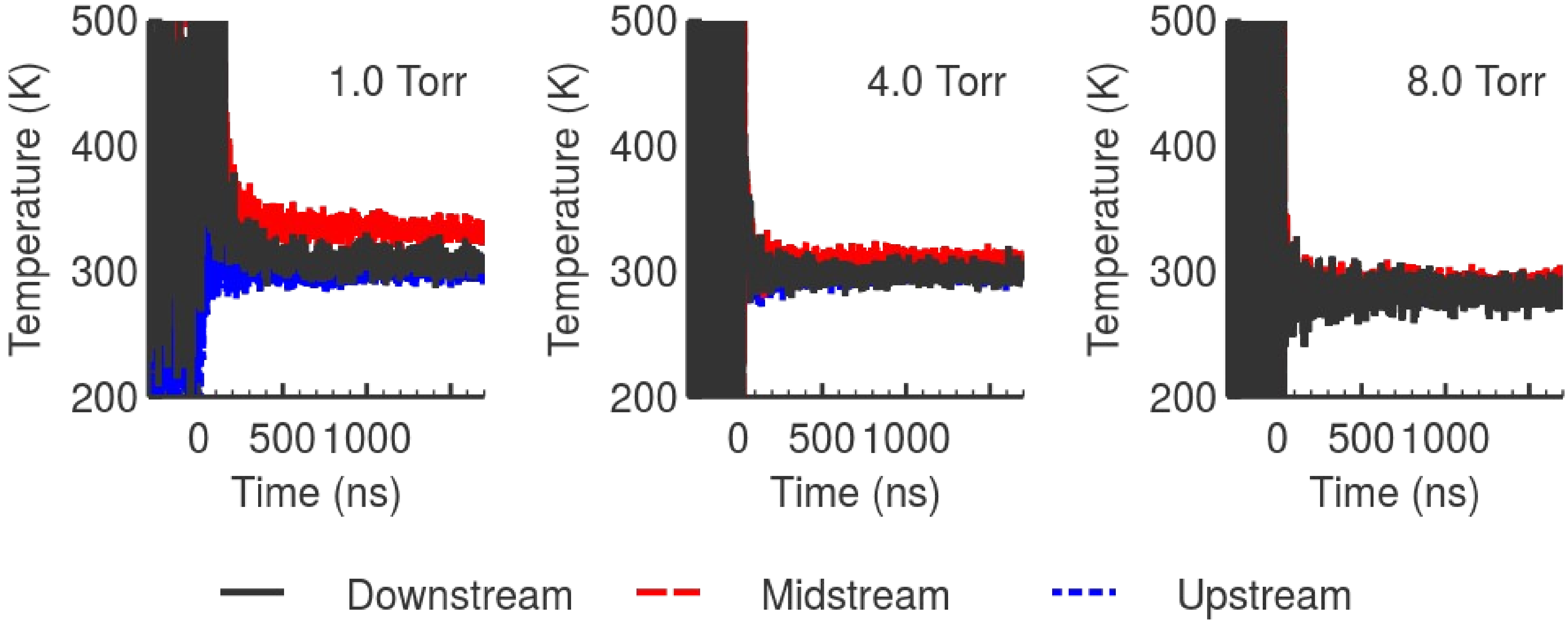
- Decrease in pre-pulse metastable population
- Peak metastable population for examined range
- Reflections from power supply deposit additional energy, boost metastable generation



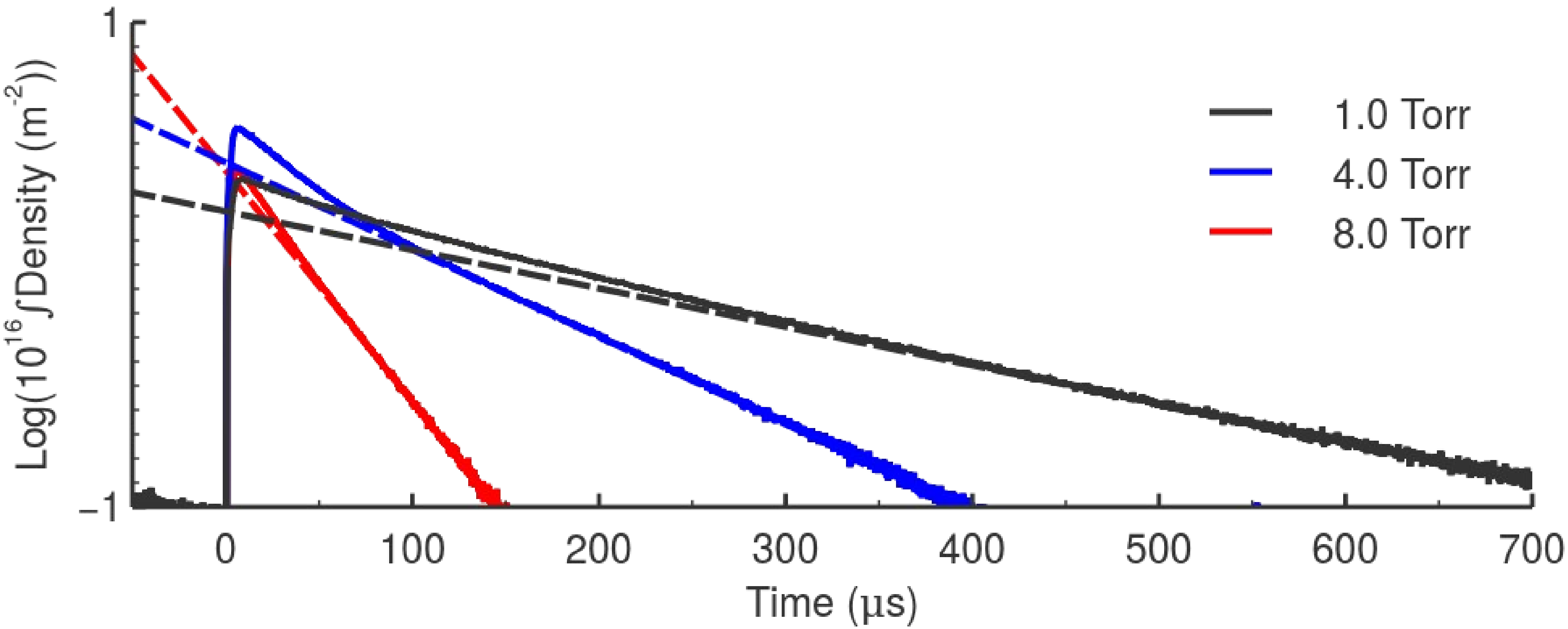
- No detectable metastables before pulse ($< 3.0 \times 10^{14}$ m^{-2})
- Overall decline in metastable density
- Reflection effects still observable
- Metastables decrease with distance from anode
  - Suggests plasma attenuation



Metastable Densities (High Pressure)

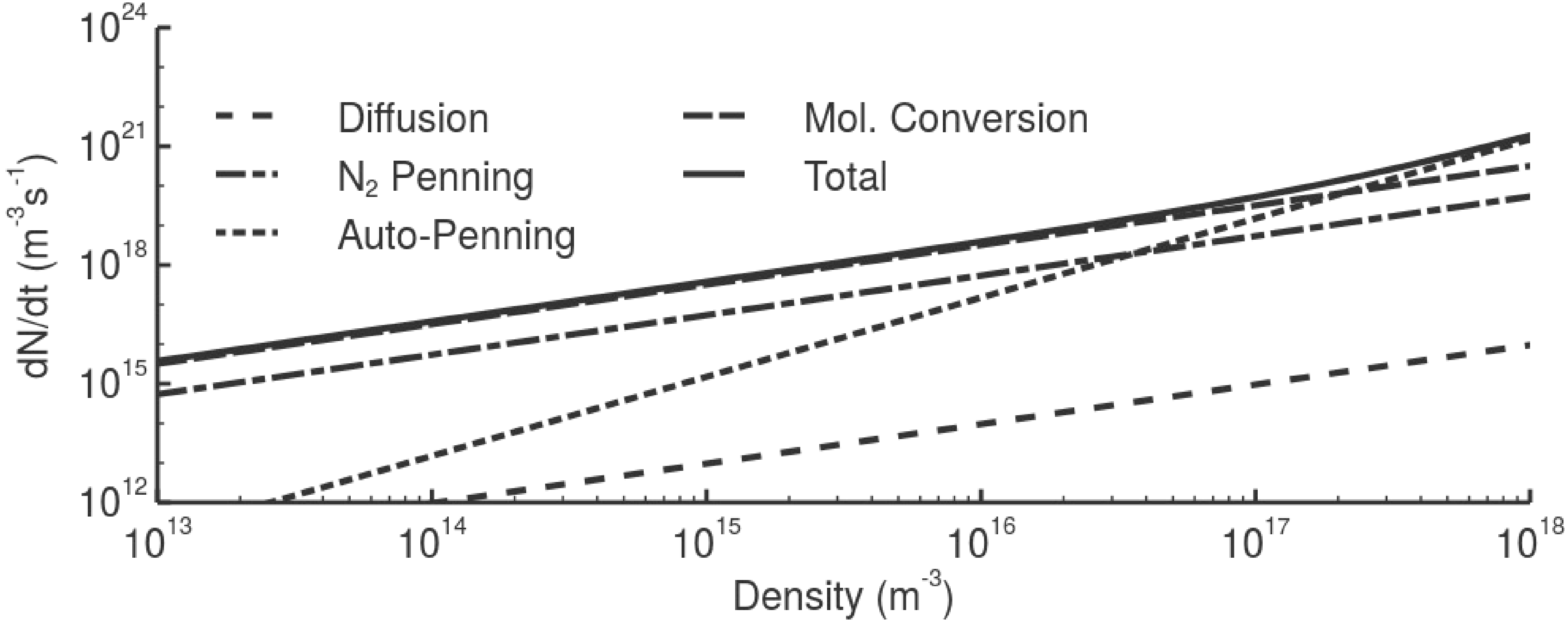


- Change in absorption resulting from small changes in laser wavelength yields gas temperatures (from Doppler broadening)
- Results indicate minimal gas heating
- No significant difference from 300 K for all conditions



- Long-duration metastable measurements can reveal loss mechanisms
- Straight line indicates exponential process (e.g. diffusion)
- Deviation from straight line implies a non-exponential process (e.g. Penning ionization between metastables)

Metastable Destruction



- Initially dominated by non-exponential processes
  - Superelastic electron collisions (previous measurements show electron density comparable to metastable)
  - Penning ionization between metastables
- Molecular conversion dominates at lower metastable densities

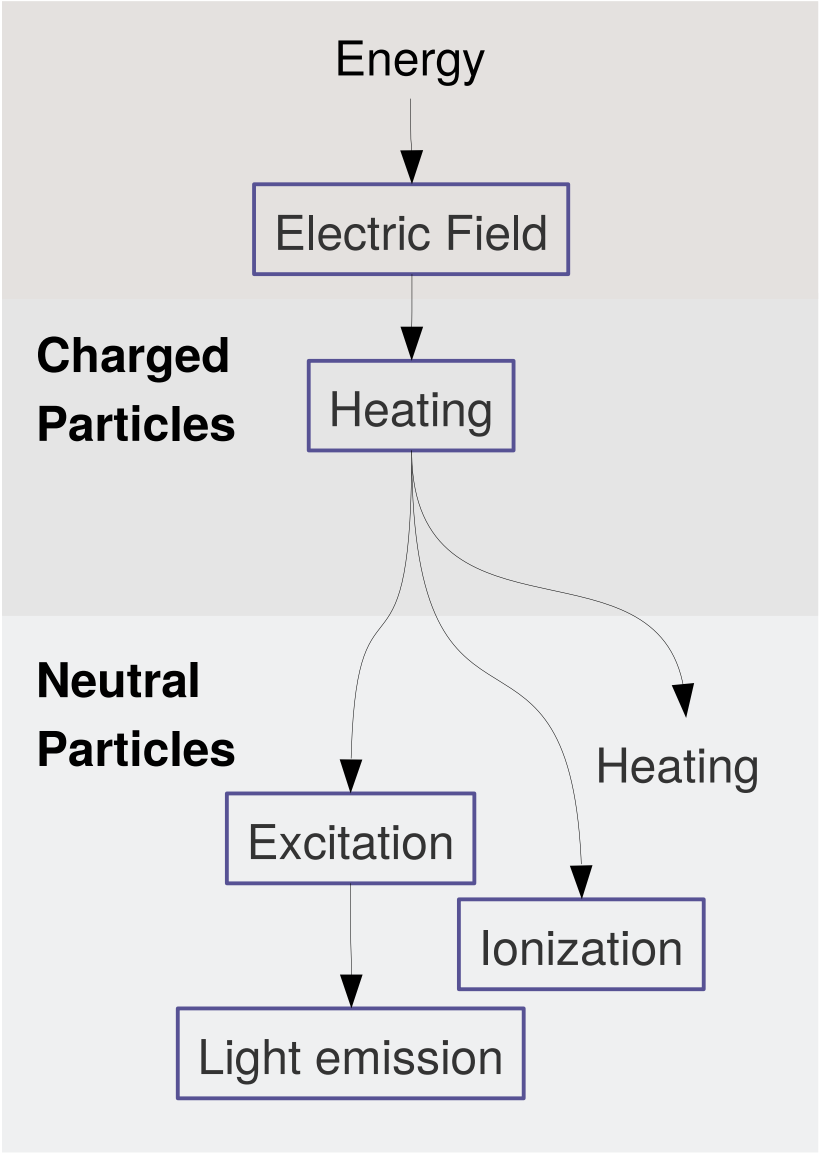
Destruction Processes

- Metastable states only persist between pulses at lower pressures
- There is an optimal pressure for metastable generation
- Additional energy deposited by pulses reflected from power supply
- Plasma appears to attenuate at higher pressures
- No detectable gas heating
- Data suggest losses initially dominated by superelastic electron collisions and Penning ionization between metastables

Metastable Summary

Global Model

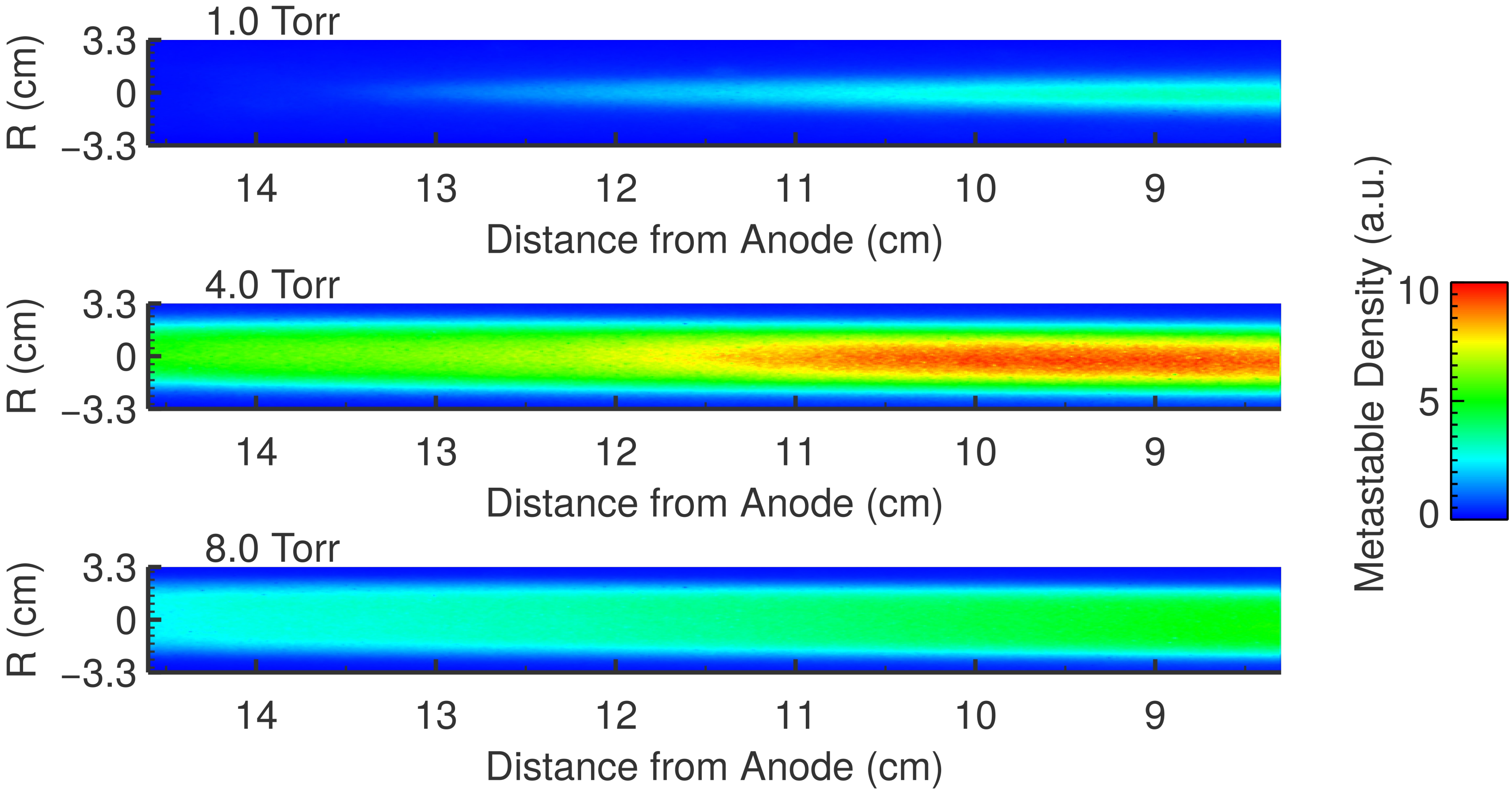
- Metastables only one of many excited states
- Use metastable measurements to infer other properties
  - Electric fields
  - Electron densities and temperatures
  - Excited states
- Can be accomplished with a numerical model of plasma



- Several approaches and degrees of complexity
  - Fluid, kinetic, or particle
  - 0,1,2, or 3 dimensions
  - Particles and reactions to include
- Used global model based on first and third moments of Boltzmann equation
  - Simple starting place
  - Allows inclusion of large number of reactions
  - Does not solve for spatial variations (homogeneous)

- Species:
  - Helium, all states $n < 5$ (multiplet averaged)
  - 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.

Model Development



- Previous LCIF measurements showed radial variations in metastable and electron densities
- Cause and pressure-dependence not well-understood
- Assume uniform radial density distribution, results can be modified for different profiles

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 (First Moment)

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

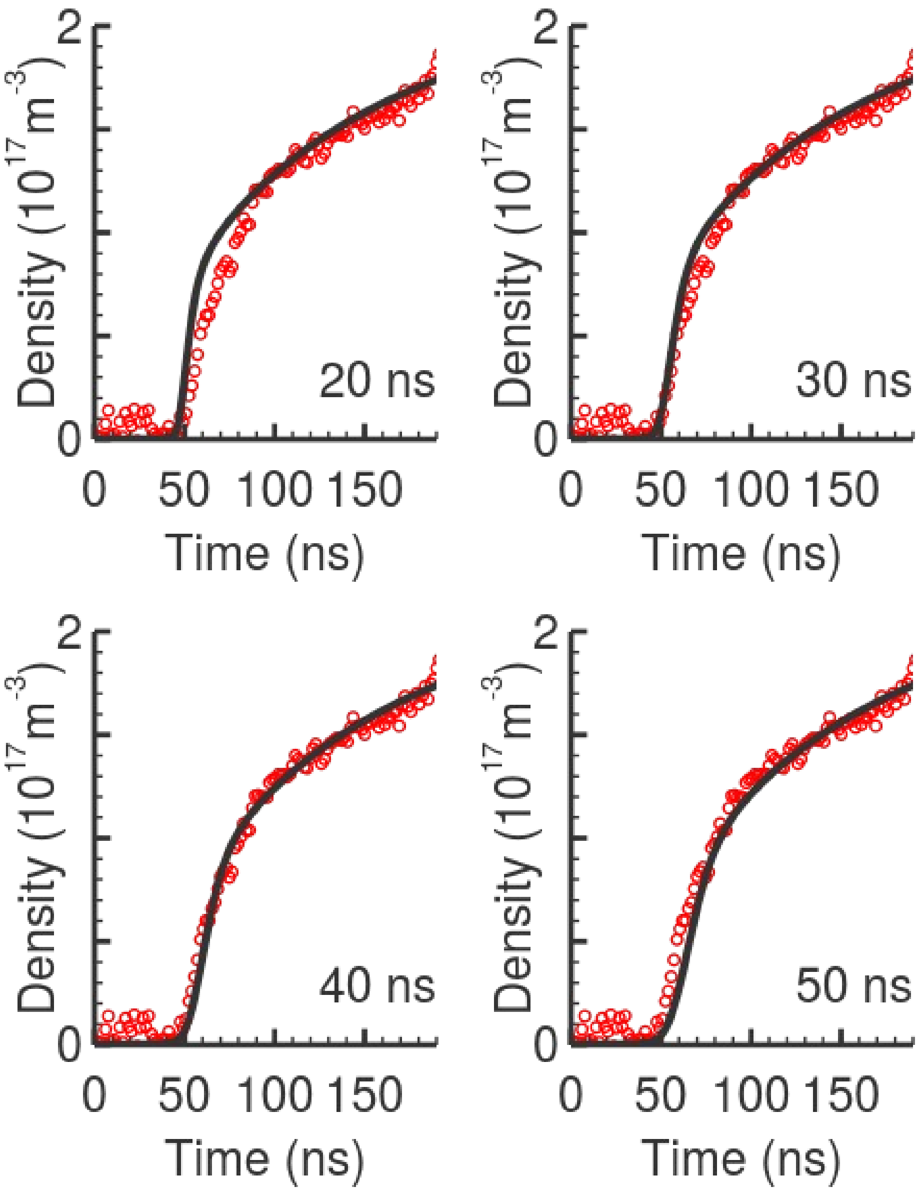
Electric field heating

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

Elastic collisions

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

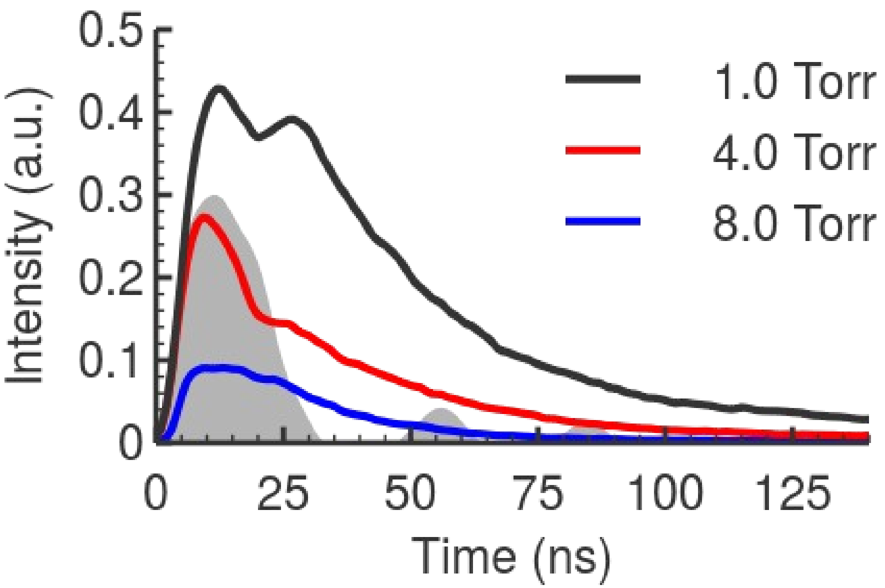
Inelastic collisions

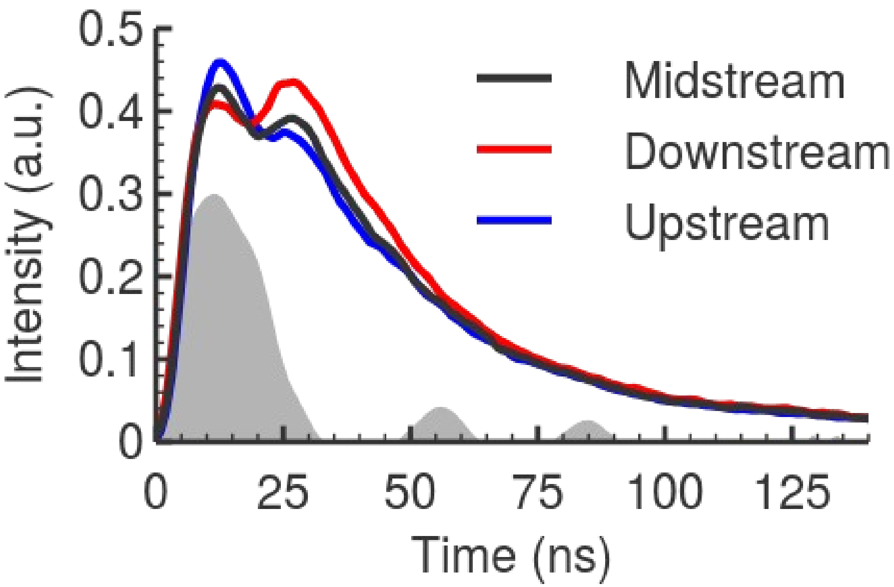


- Assumed electric field applied as a Gaussian pulse
- Free variables: peak electric field and pulse width
- Best fit obtained with 40 ns pulse-width.
- Why is this longer than the applied pulse (25 ns)?

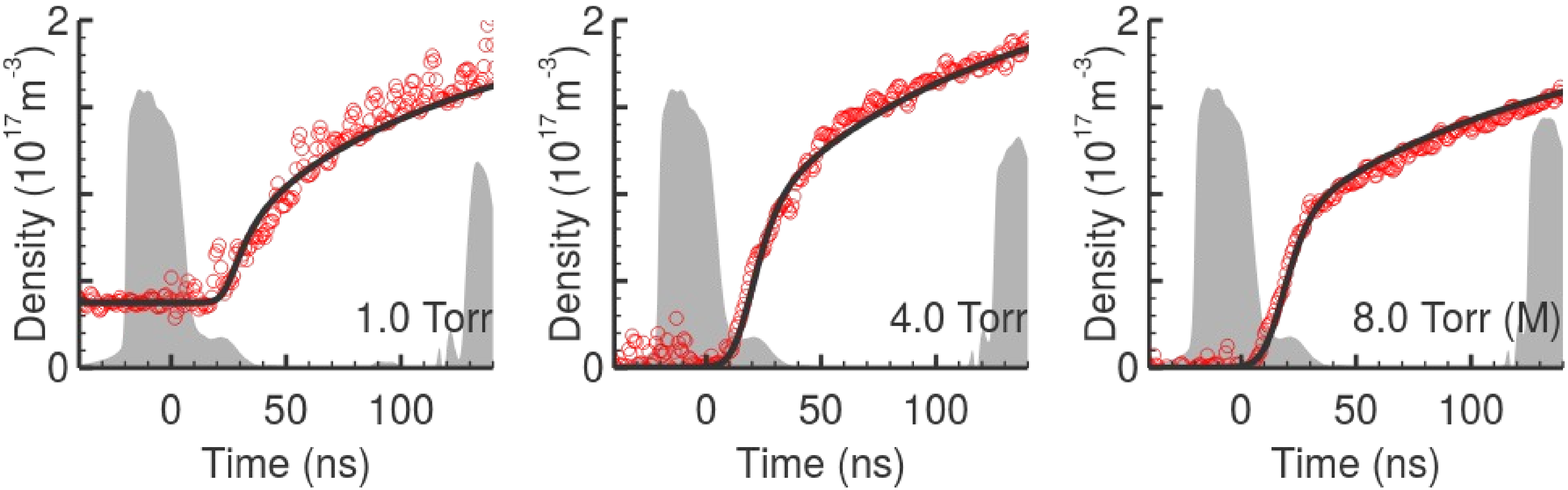
Pulse-width

- Optical emissions showed evidence of return stroke
  - Counter-propagating excitation from cathode to anode
- Double peak at several pressures
  - Different from power supply reflections
  - Second peak most intense closest to cathode
- *Effective* excitation period longer than 25 ns

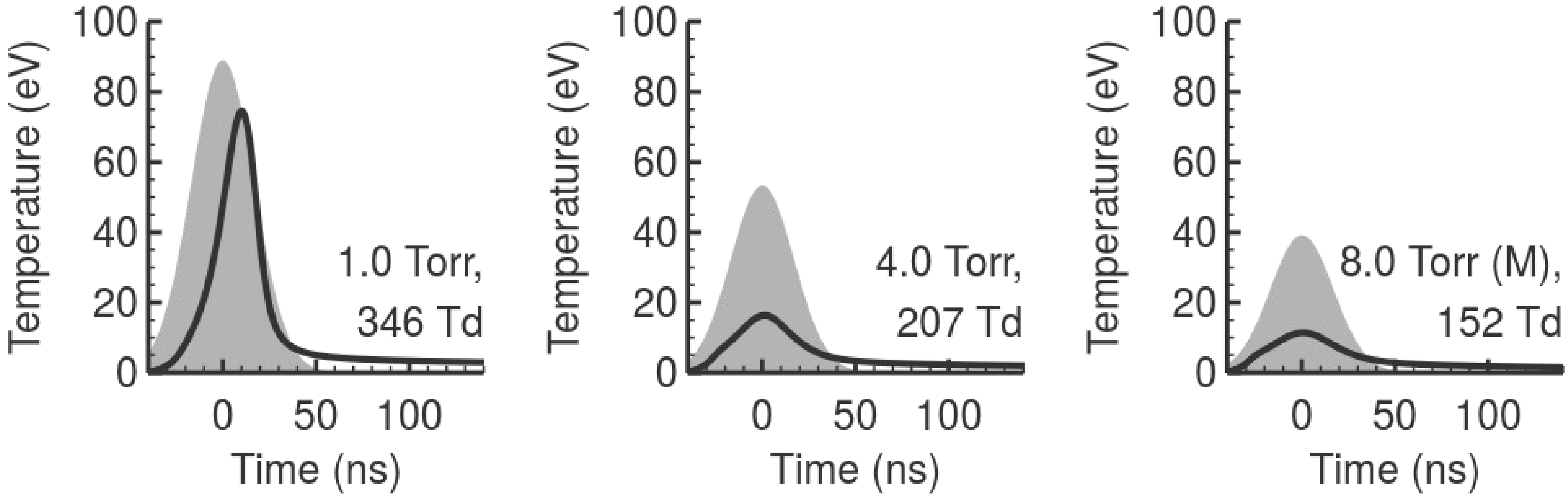




Evidence for Return Stroke



- 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?



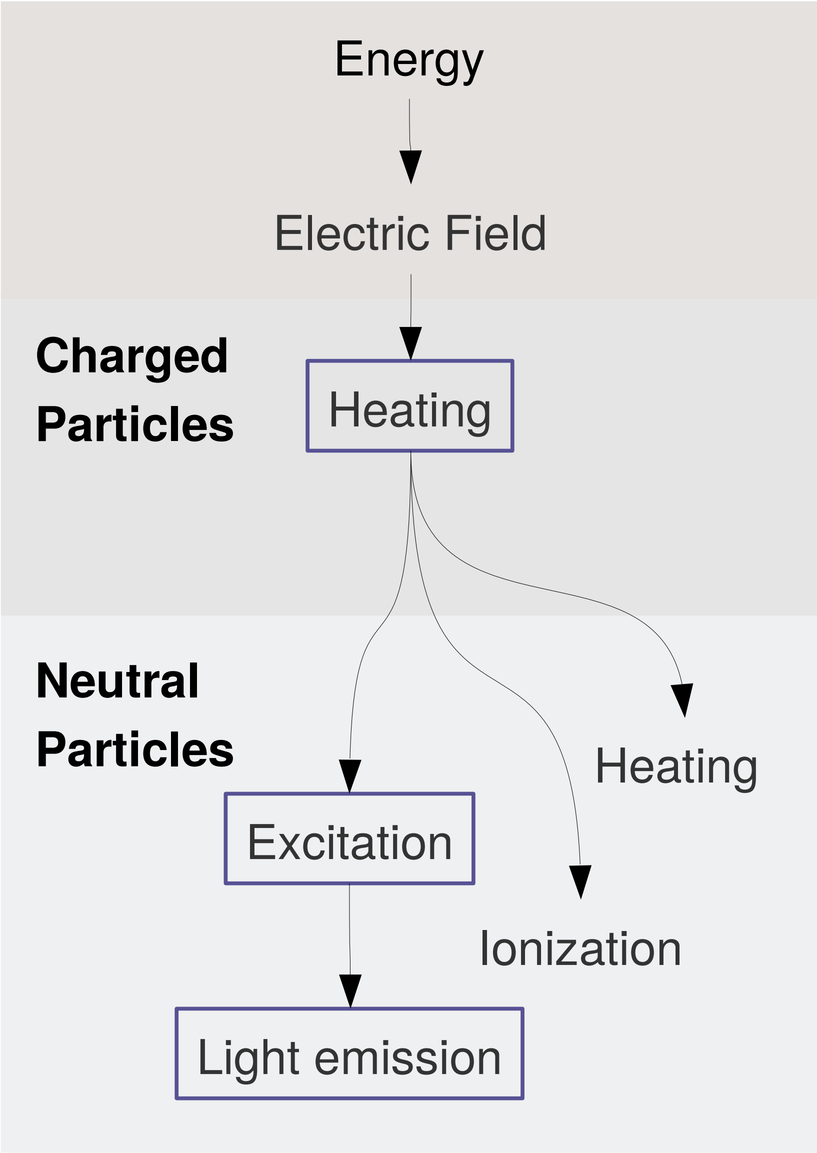
- Peak fields consistent with other studies, but in range where EEDF is not well-matched by Maxwell-Boltzmann
- Electron temperatures (particularly 1.0 Torr) are excessive
- Suggests potential violation of assumptions
- Different distribution or model necessary?

Peak Fields and Electron Temperatures

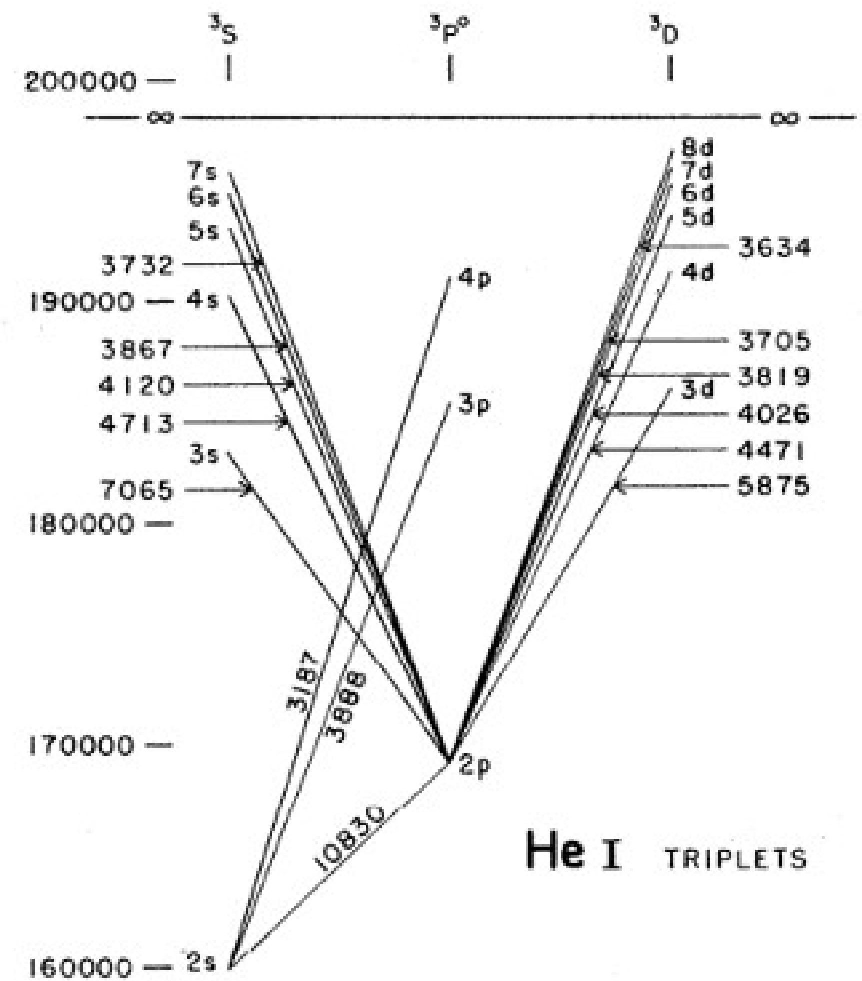
- Created global model of helium plasma and applied to RPND
- Observed evidence of extended excitation period in population of metastables
- Simulations reproduced metastable density dynamics and magnitudes
- Temperatures and fields at, especially at low pressures, suggest violation of underlying assumptions (e.g. Maxwell-Boltzmann distribution)

Emission Measurements

- Simulations based on metastable densities
  - Generated predictions of other excited state densities and emissions
- Optical emission spectroscopy
  - Experimental check of simulated emissions
  - May reveal other phenomena
  - Potential estimate electron temperature

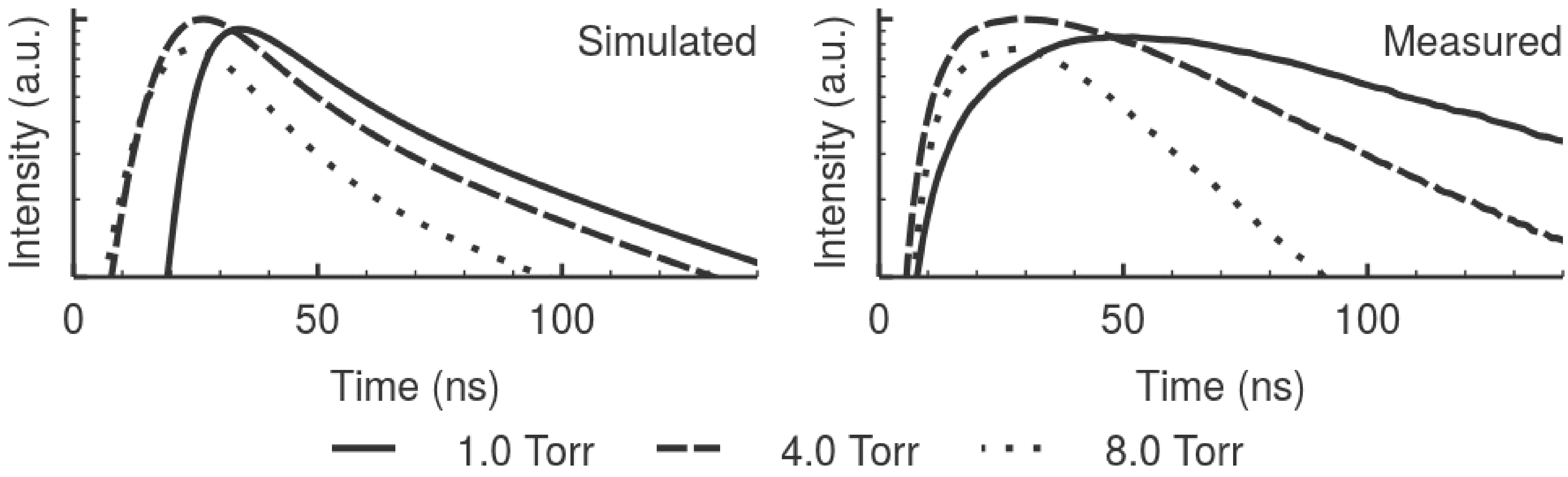


Checking the Simulations



- Observed optical emissions in visible range (triplet and singlet)
- Calibrated relative intensities with tungsten lamp
- Same conditions as absorption spectroscopy
  - 0.3 – 16.0 Torr
  - Three axial locations

Helium Emissions



- Relative peak heights and timing similar, distinctly different in shape
- Measurements suggest pressure-dependent de-excitation process
- Decay rate increases with pressure
  - Penning ionization of impurities?
  - Missing excitation transfer rates?
- Long emission lifetime suggest an extended excitation process
  - Beam-like electrons
  - Persistent electric field (seen in other experiments)

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

- Resonance radiation (transition from excited state to ground) can be "trapped" in plasmas
- Absorbed and re-emitted many times before exiting
- Increases energy residence time
- Described by "trapping factor", multiplier of normal lifetime
  - Geometry and lineshape dependent
- Could contribute to extended excitation

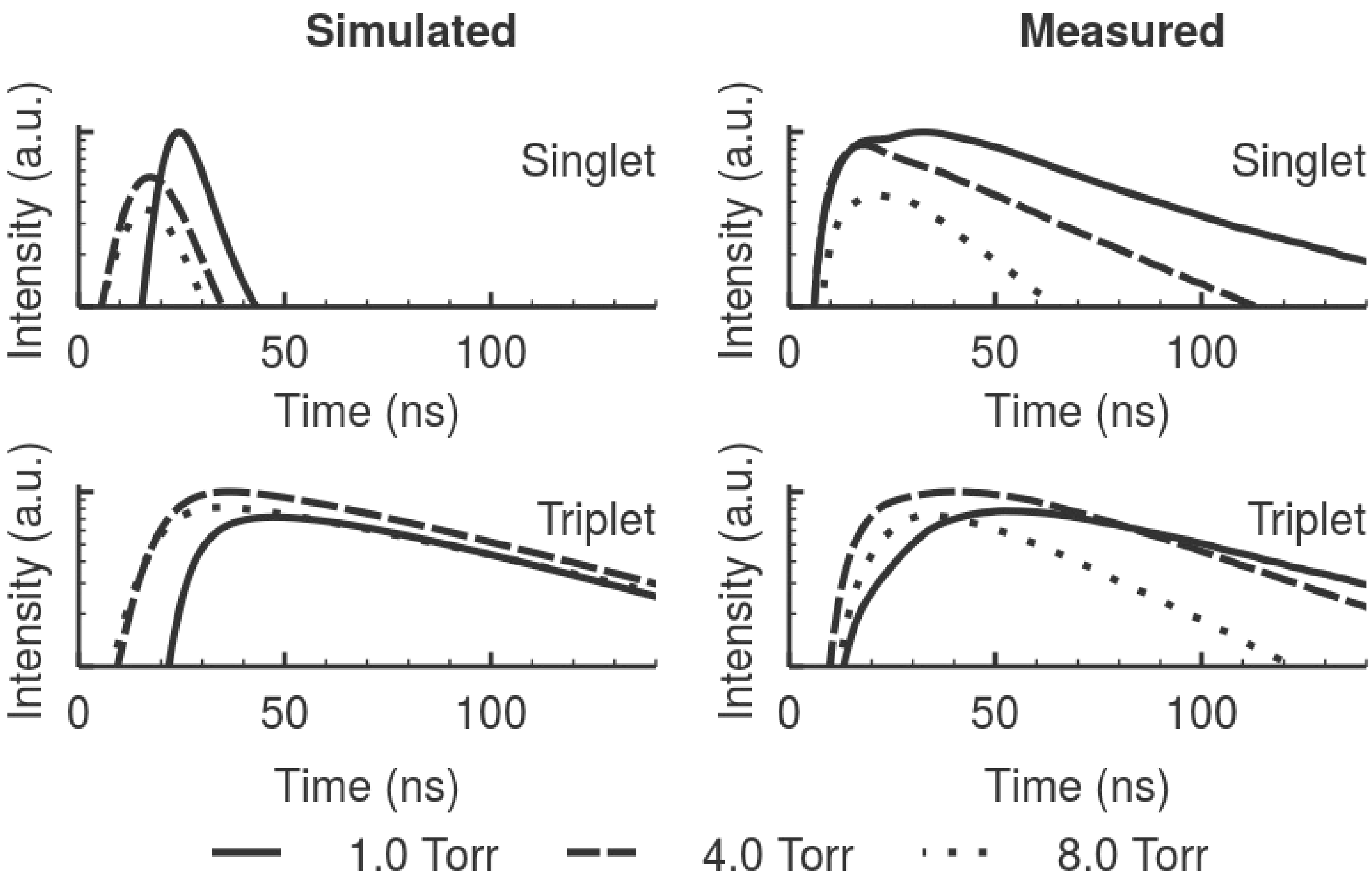
Pressure (Torr)	Trapping Factor	Effective Lifetime (s)
1.0	8,773	1.549×10^{-5}
4.0	38,031	6.715×10^{-5}
8.0	78,837	1.392×10^{-4}

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

Radiation Trapping

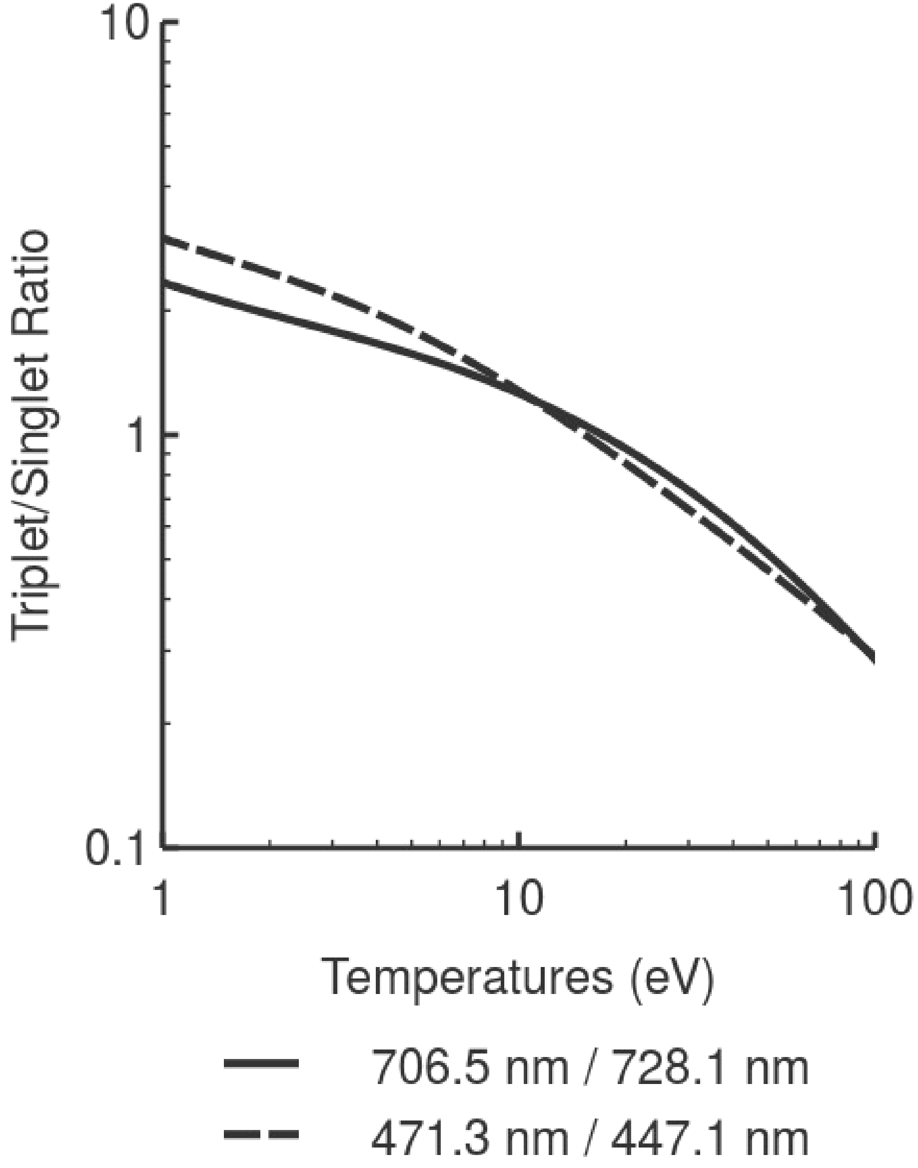
- Compare same transition within triplet and singlet manifolds at 501.5 nm and 388.8 nm ($3P \rightarrow 2S$)
  - Only 3^1P has a resonance transition, 3^3P does not
- If radiation trapping is absent: comparison of simulated and measured singlet emissions should be the same as the triplets
- If radiation trapping is present: simulated singlet emissions will decay faster than measured emissions, but triplets will be unaffected

Verification of Radiation Trapping



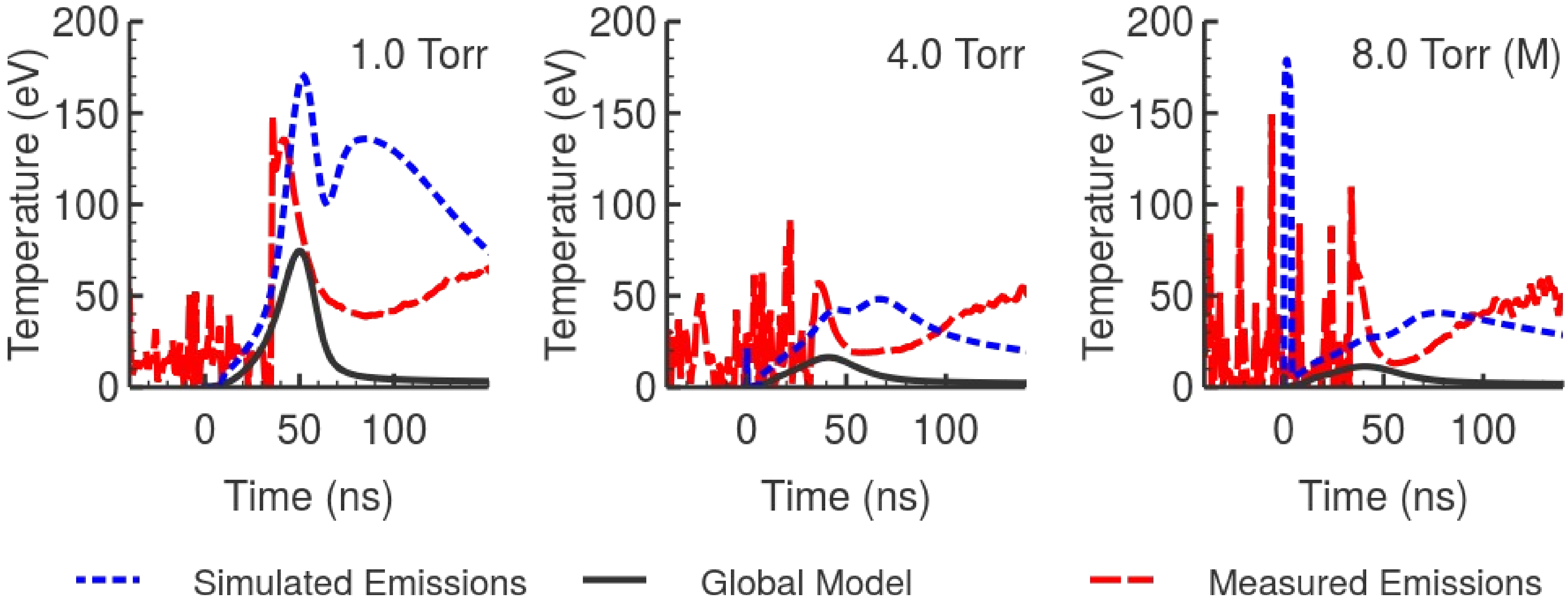
- Simulated singlet emissions fall off more rapidly than measured
- Comparison of triplet emissions show a lack of the same behavior
- Suggests importance of radiation trapping

Verification of Radiation Trapping



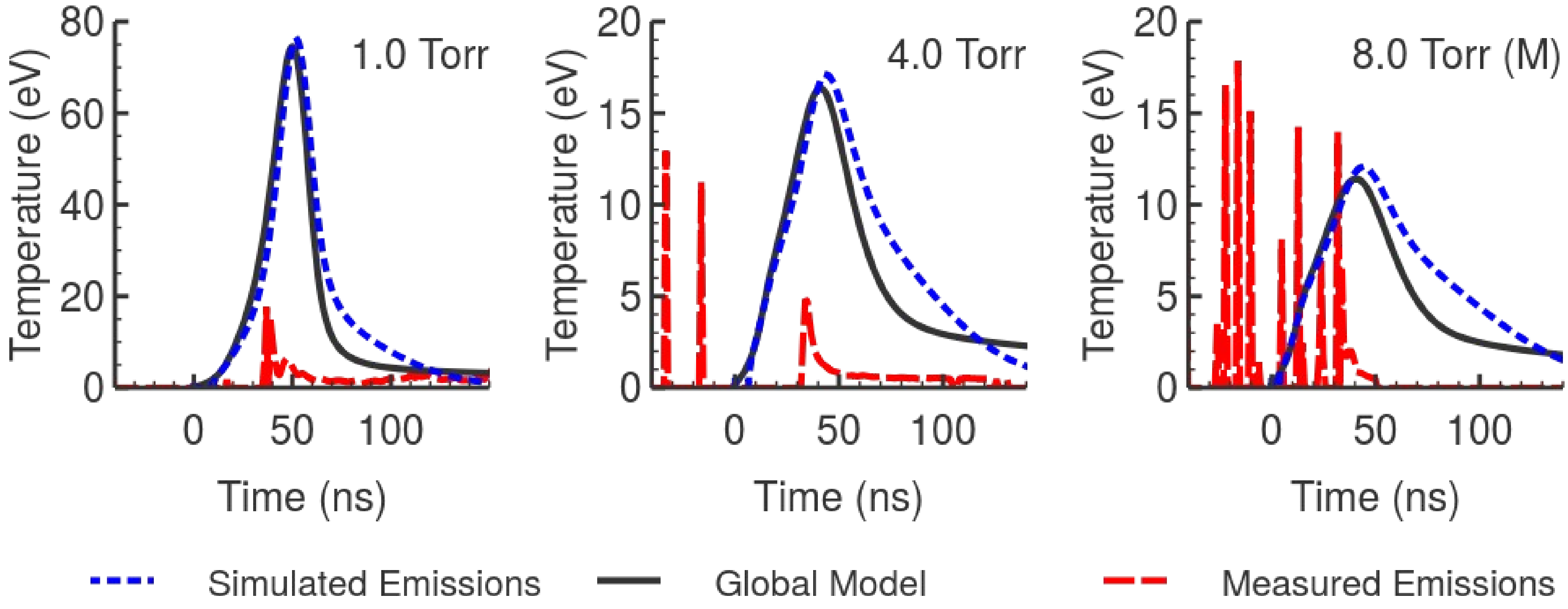
- Plasma emissions can be used to estimate electron temperature
- Several approaches, line ratios based on coronal model possibly suitable
- Assumes
  - Excited states generated by electron collisions with unexcited atoms
  - Excited states decay by radiative emission only
- Maxwell-Boltzmann distribution used for consistency, others possible
- Two candidate ratios selected for evaluation
  - 706.5 nm / 728.1 nm ($3^3S \rightarrow 2^3P^o$ / $3^1S \rightarrow 2^1P^o$)
  - 471.3 nm / 447.1 nm ($4^3S \rightarrow 2^3P^o$ / $4^1D \rightarrow 2^1P^o$)

Coronal Model



- Compared temperatures from electron energy equation with those predicted by simulated line ratios and measured line ratios
- Temperatures from simulated line ratios did not match solutions of electron energy equation
  - Suggests coronal model does not apply for these transitions
- Possible cause is collisional redistribution of n = 4 states
- Agreement may be improved by inclusion of n = 5 states

Ratio 1: 471.3 nm / 447.1 nm



- Simulated emissions consistent with solutions of electron energy equation
  - Coronal model may be applicable
- Application to measured emissions was limited by signal-to-noise ratio
- Distribution likely not Maxwell-Boltzmann (concept of temperature not applicable)
  - May still be useful as a measure of mean electron energy

Ratio 2: 706.5 nm / 728.1 nm

- Global model captures some features of RPND population kinetics
- Missing physics
  - Extended excitation (distribution or field related?)
  - Radiation trapping
- $3^3S \rightarrow 2^3P^o$ / $3^1S \rightarrow 2^1P^o$ 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

- Pulse reflections can deposit significant amounts of additional energy
- Metastable states only persist between pulses at lower pressures
- Excitation period lasts significantly longer than expected
  - Partially a result of a return stroke
  - Additional physics necessary to completely account for behavior
- May be possible to employ simple line ratio diagnostics for estimates of mean electron energy

- Address temporal evolution of metastable density profile (Abel inversion)
  - Provides insight on the relation between wall effects and radial profiles
- Incorporate additional physics (Penning ionization of impurities, radiation trapping, etc.) in present global model
  - Would address whether post-pulse behavior can be completely described by mechanisms unrelated to EEDF
- Employ different EEDF models to better match excited state dynamics
  - May provide alternative explanation for extended excitation in RPND
- Develop a 0D Monte-Carlo simulation of RPND
  - Could provide better insight on EEDF evolution under high electric fields

Future Work

Acknowledgements

Questions