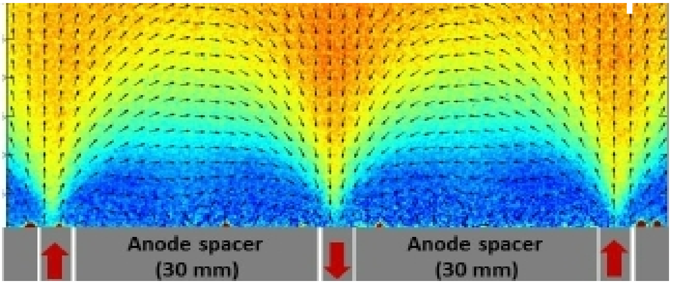
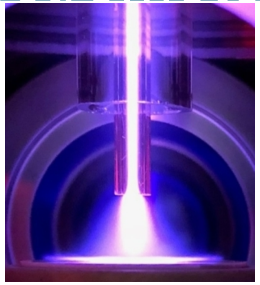
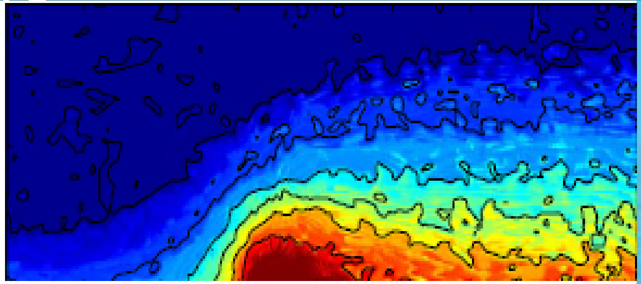
SAND2021-12565C

Laser-Produced Aluminum Plasmas Expanding in an Applied Electric Field: Plasma Generation in Single Particle Aerosol Mass Spectrometers







74th Annual Gaseous Electronics Conference

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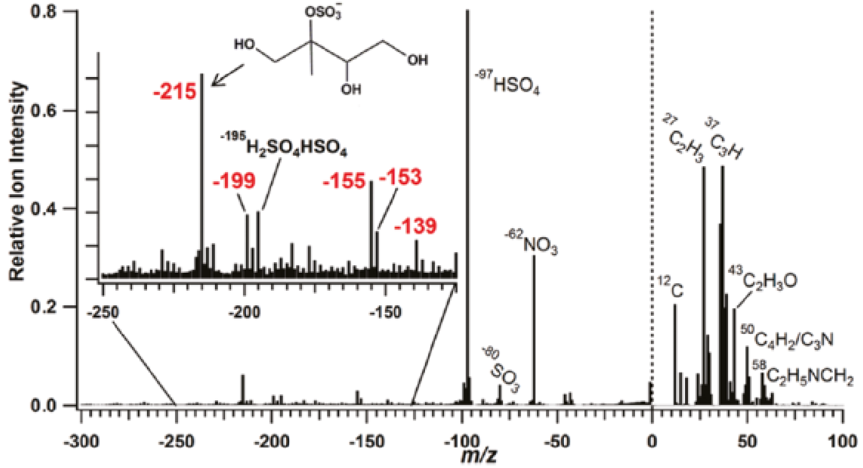
Agenda

- Single Particle Aerosol Mass Spectrometry
- Model Description
- Global Plasma Modeling
- Effect of Applied Field
- Comparison with Experimental Data

Aerosol Mass Spectrometry

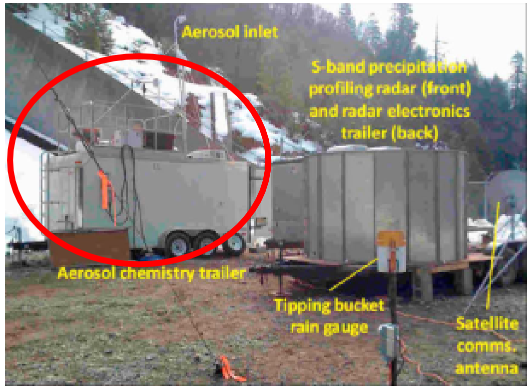
- Aerosol mass spectrometry is critical for
  - Climate science: Cloud condensing nuclei, precipitation
  - Atmospheric monitoring: Pollution, dust storms, forest fires
- The chemical composition of particles can indicate their origin or hazardousness.
- Real-time measurements provide accurate information on reactive particles.
  - Small size and weight desirable for field/vehicle tests.
- Measuring a mass spectrum for individual particles provides more information and is especially useful for source attribution.

Aerosol mass spectrum collected in Atlanta, GA

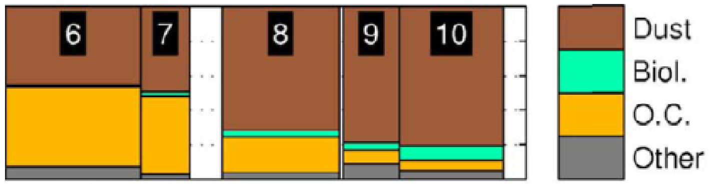


L. E. Hatch et al., Environ. Sci. Technol. 45, 5105 (2011).

Aerosol mass spectrometer in a climate study in the Sierra Nevada Mountains



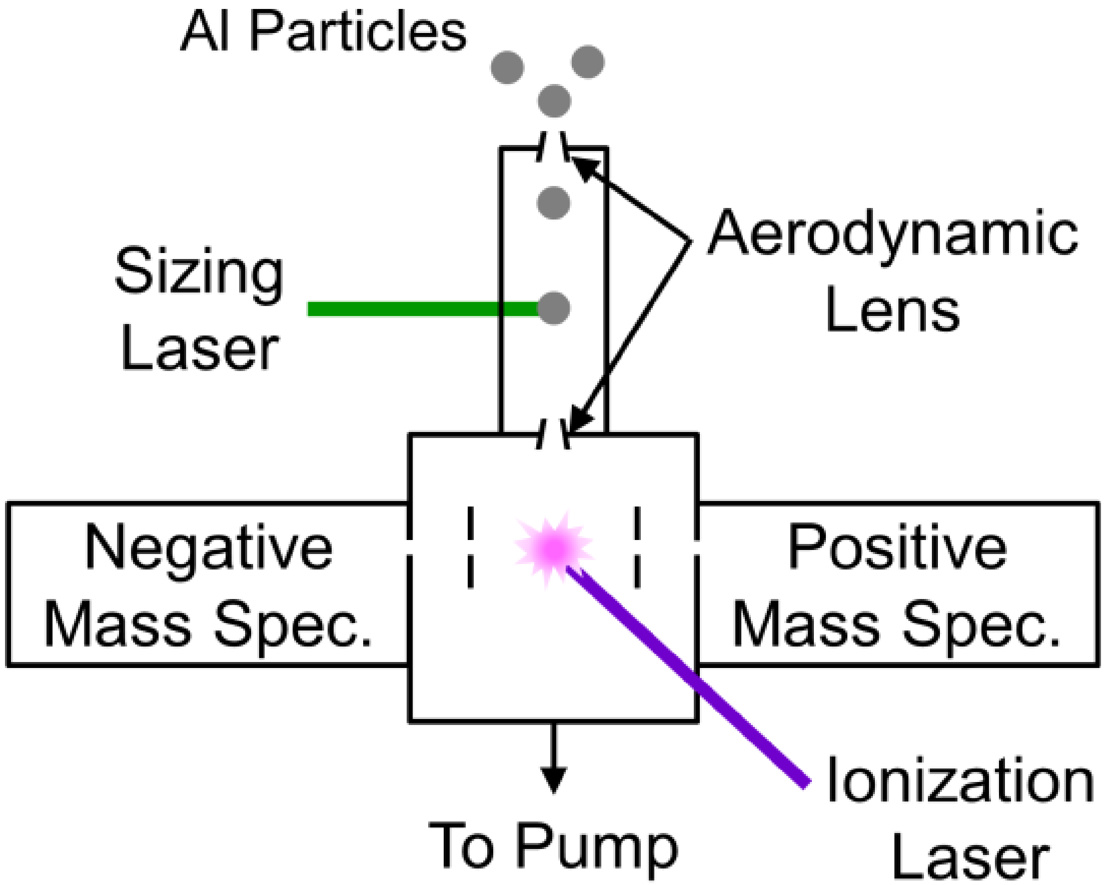
Single particle spectra facilitate categorization of particles:



A. P. Ault, et al., J. Geophys. Res. 116, D16205 (2011).

Single Particle Aerosol Mass Spectrometer (SPAMS)

- Conditions based on SPAMS 3.0 by Livermore Instruments
- An aerodynamic lens focuses the incoming particles into a beam into a differentially pumped chamber.
- First sizing laser (405 nm, continuous) detects particle and measures its aerodynamic diameter.
- Ionization laser (248 nm, 8 ns) vaporizes and ionizes.
- An applied field (2.8×10^5 V/m) separates negative and positive ions.
- In this study, we focused on spherical Al particles.



- **Goal: Better understand ionization mechanisms in SPAMS systems using numerical modeling.**
  - Results may aid in **future designs** and **analysis** of results.

Global Model Description

- 0-D, well-stirred reactor approximation.
  - Feasible to achieve μs timescales and address some problem-specific issues.
- Al, e^-, Al^+, 6 excited states.
- 90 reactions:
  - Electrons: excitation, ionization, recombination, superelastic
  - Photons: excitation, ionization, inverse Bremsstrahlung radiation
  - Penning ionization, quenching
- Two temperatures (T_e, T_g).
- Maxwellian energy distributions.
- Uniform photon flux.

Global Plasma Model

Reactions — Transport — Expansion

$$\frac{\partial n_e}{\partial t} = \sum_j^{rxns} \left(a_{e,j}^{RHS} - a_{e,j}^{LHS}\right) R_j - \Gamma_{e,diff}\frac{A}{V} - n_e u_e \frac{\pi R^2}{V} - \frac{n_e}{V}\frac{dV}{dt}$$

$$\frac{\partial\left(\frac{3}{2}k_B n_e T_e\right)}{\partial t} = P_{ohmic} + \sum_i^{rxns} \Delta\epsilon_{e,i} R_i$$

$$-\frac{A}{V}\left(\frac{3}{2}k_B T_e\right)\Gamma_{e,diff} - \left(\frac{3}{2}k_B n_e T_e\right) u_e \frac{4R}{3} - \gamma\frac{\left(\frac{3}{2}k_B n_e T_e\right)}{V}\frac{dV}{dt}$$

n_i = number density of species I
$\Gamma_{e,diff}$ = electron diffusion flux
R = plasma radius
V = plasma volume
k_j = reaction rate of reaction j
$\Delta\varepsilon$ = energy change

$$R_i = k_i \prod_j^{LHS} n_j$$

Including Electric Field Effects in 0D

- Expansion into vacuum causes plasma density and radii to rapidly vary over many orders of magnitude.
- We must address multiple limits of behavior:
  - Plasma is dense enough to shield the $E_{applied}$.
  - Plasma density is too low to shield $E_{applied}$.
    - Electrons undergo many collisions across the plasma.
    - Electrons are accelerated out of the plasma collisionlessly.
- Electron momentum equation:

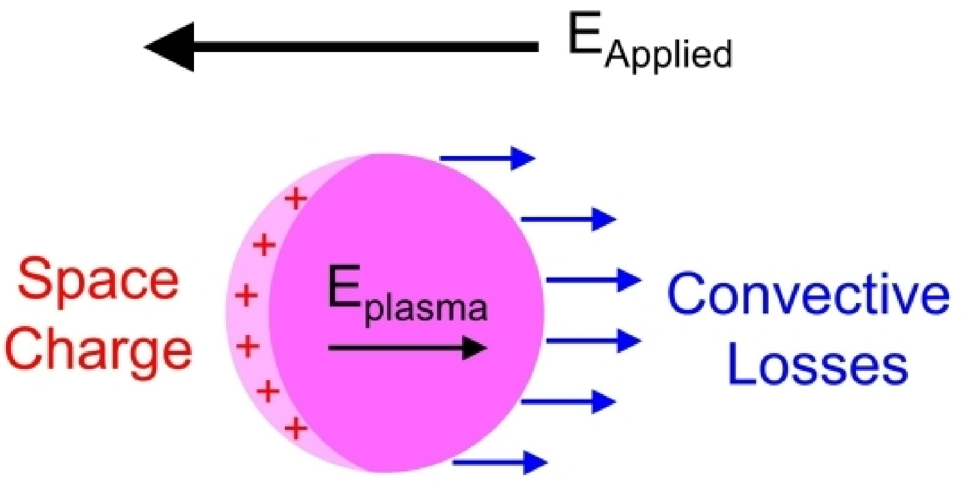
Field Acceleration Collisions

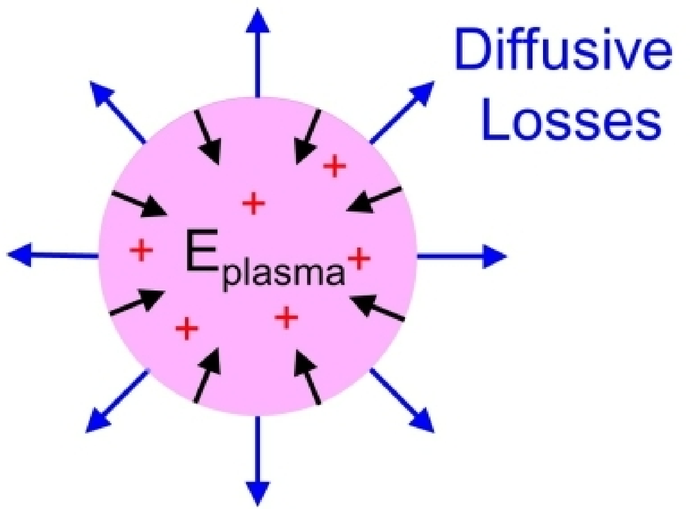
$$\frac{dm_e n_e u_e}{dt} = n_e e\left(E_{applied} - \frac{2(\sum_i q_i n_i)R}{3\varepsilon_0}\right) - m_e n_e u_e \nu_m - m_e n_e u_e^2\frac{3}{4R} - m_e u_e \Gamma_{e,diff}\frac{3}{R} - \frac{m_e n_e u_e}{V}\frac{dV}{dt}$$

Convection/Expansion

$$\Gamma_{e,diff} = \frac{1}{4} n_e \sqrt{\frac{8k_B T_e}{\pi m_e}}\, exp\left(-\frac{e\Phi_{plasma}}{k_B T_e}\right) \qquad \Phi_{plasma} = \frac{Q}{4\pi\varepsilon_0 R} = \frac{\sum_i q_i n_i}{4\pi\varepsilon_0 R}V$$

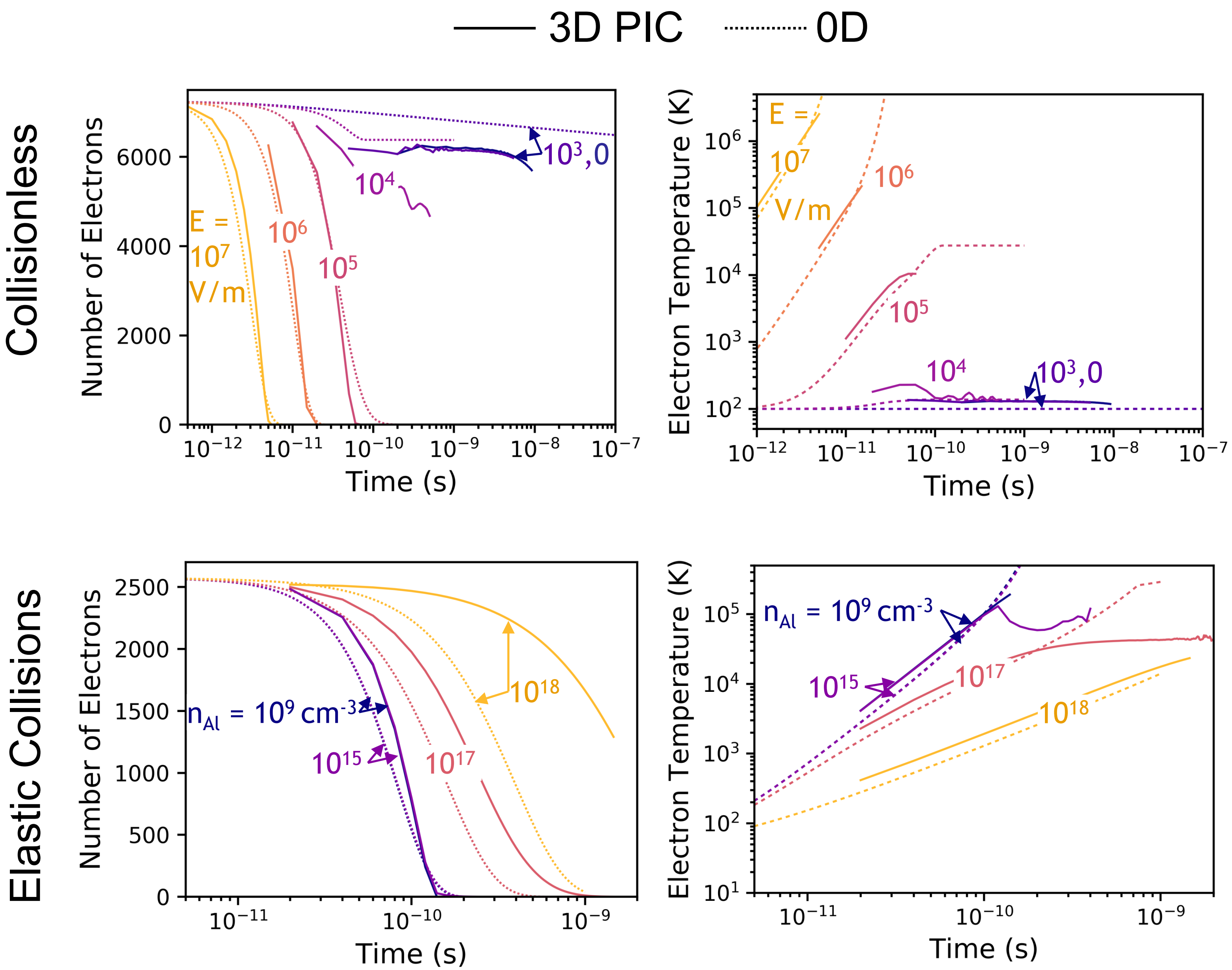
- Once e^- leave the control volume, they are ignored:





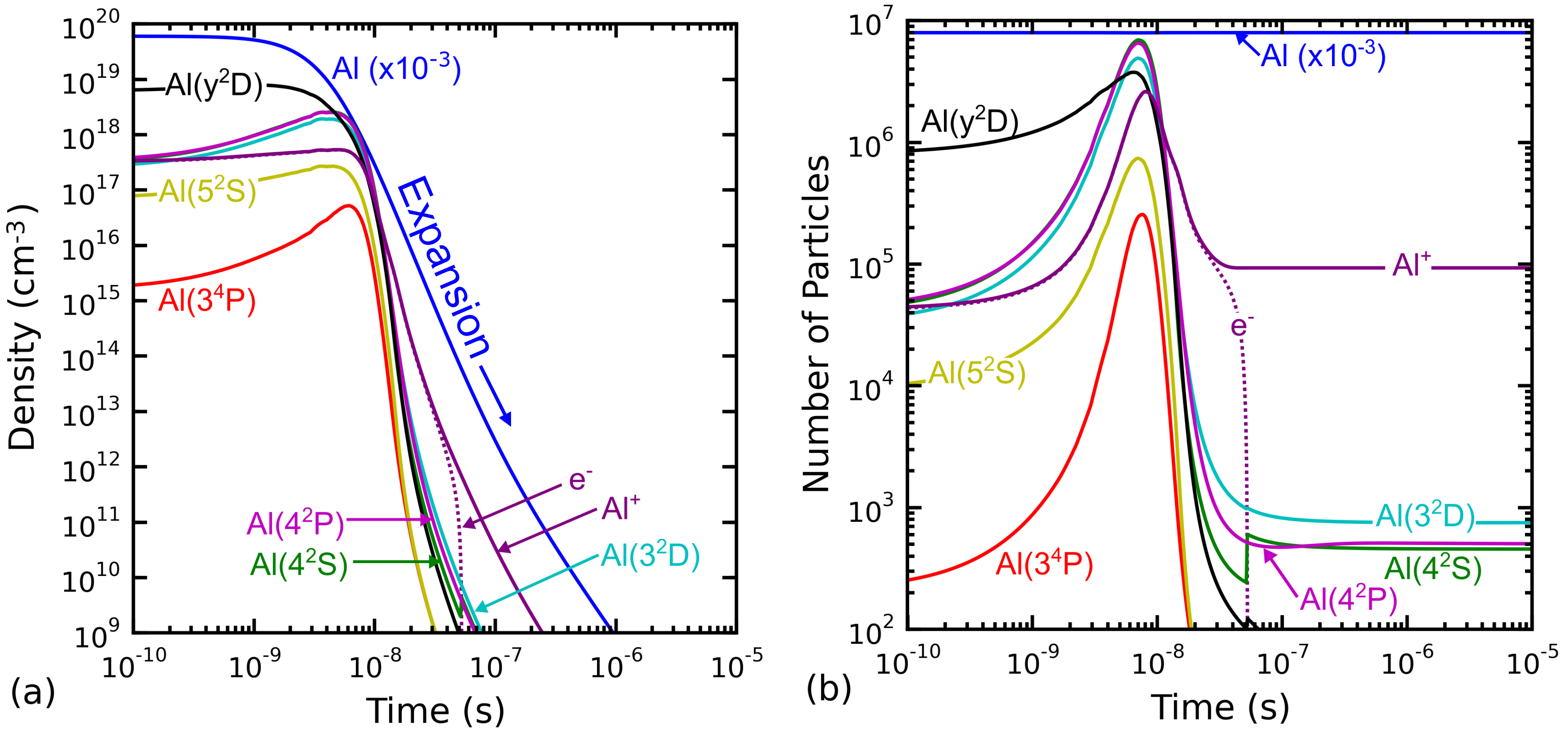
Benchmarking Electric Field Effects

- Toy problems to check the approach: no chemistry, no expansion.
- Aleph, electrostatic PIC-DSMC model
- 3D wedge
- Collisionless:
  - R = 12 μm, $n_e = 10^{12}$ cm^{-3}
  - T_e = 100 K, T_i = 0 K
- Elastic collisions:
  - R = 85 μm, $n_e = 10^{9}$ cm^{-3}
  - T_e = 40 K, T_i = 0 K
  - Elastic collision, $\sigma(v) \propto 1/v$
- Runtime:

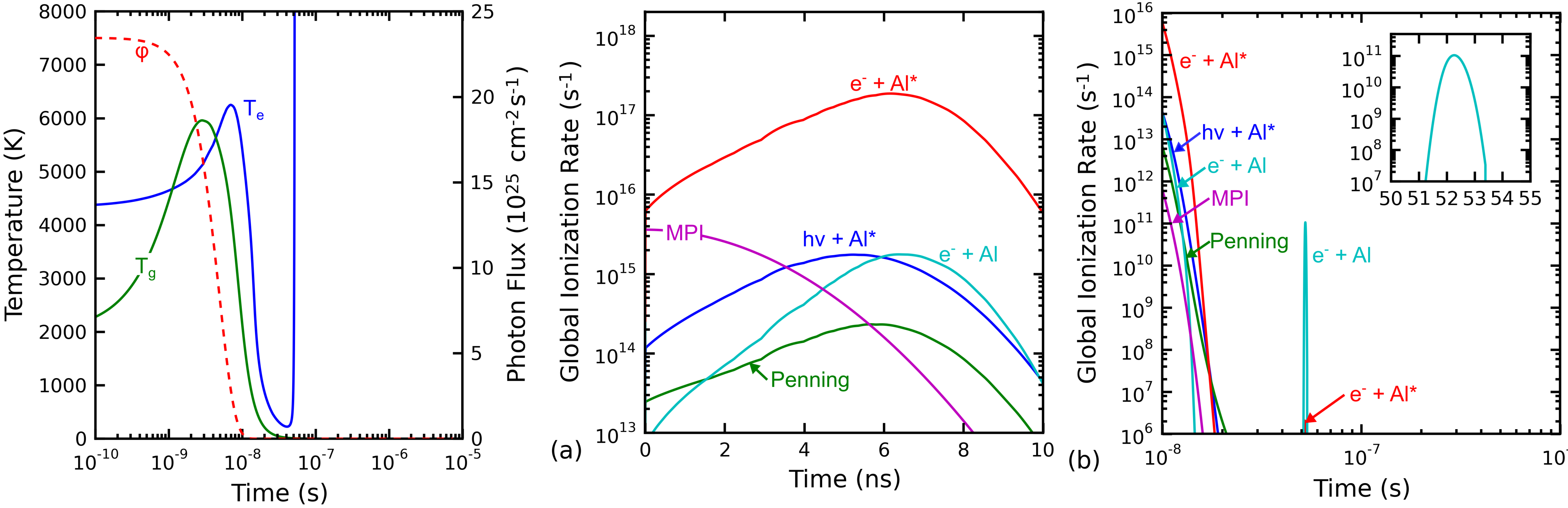


Species Densities

- $E = 2.8 \times 10^5$ V/m
- 1 μm Al
- 8 mJ, 248 nm
- Electric field removes electrons from the plasma at 20-50 ns.
  - Decreasing the laser pulse duration would not improve the mass resolution.
- Removal of electrons interrupts rapid three-body recombination.



Ionization Mechanisms



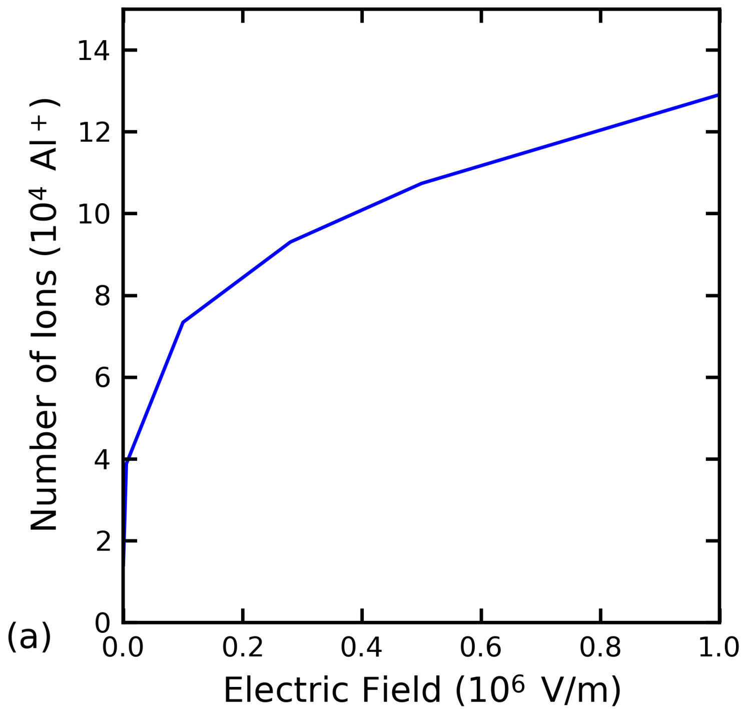
- As applied field accelerates electrons T_e increases rapidly.
- T_g continues to cool due to adiabatic expansion.

- Global ionization rate = volumetric rate × plasma volume.
- The primary ionization mechanism is:

$$e^- + Al^* \rightarrow Al^+ + e^- + e^-$$

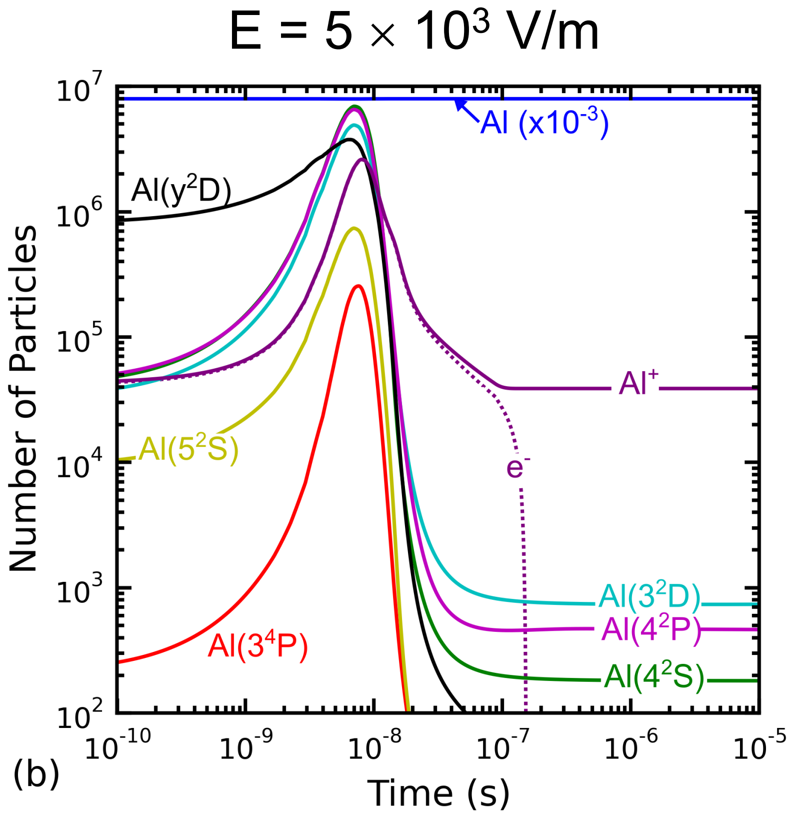
- As e^- are accelerated out of the plasma, some electron impact ionization occurs.
- This process only produces ~ 100 ions. (<< 10^5 ions present)

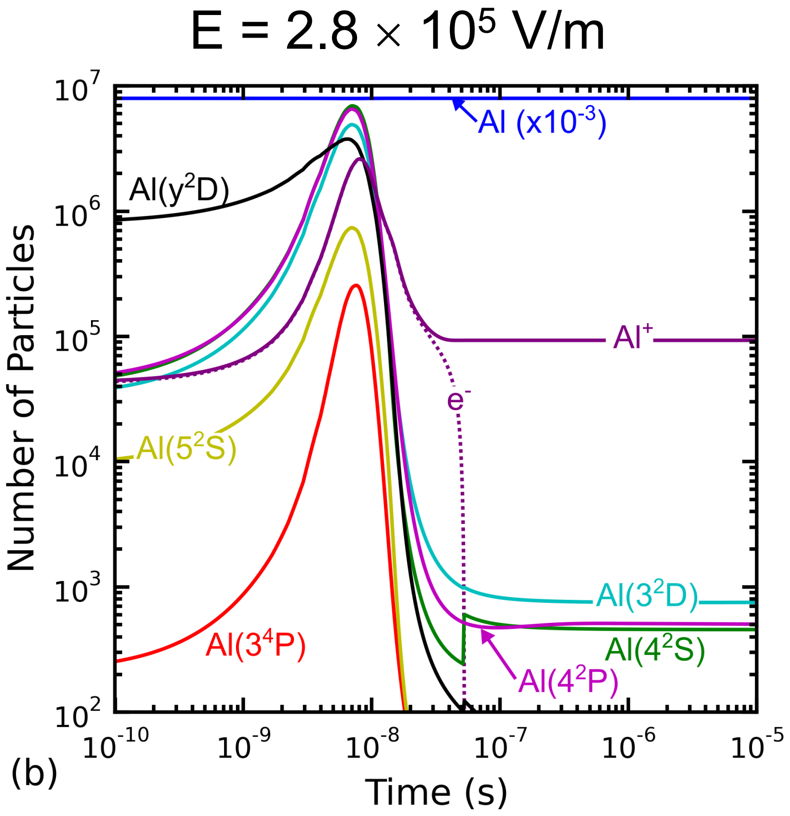
Vary Applied Field



- Total number of Al^+ ions at 1 μs
- 1 μm Al particle.

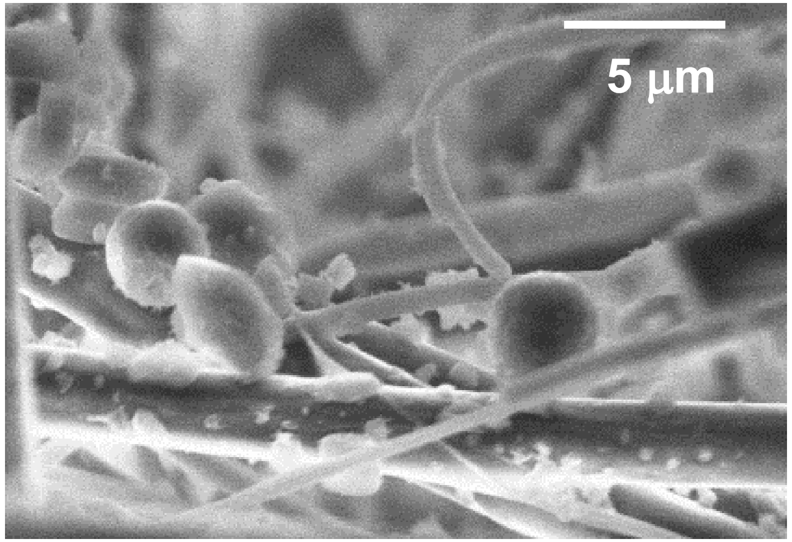
- Applied field has negligible impact before charge separation.
- Higher field means earlier charge separation.
- Total ion count increases as the electrons are removed from the plasma earlier in time, preventing more recombination.

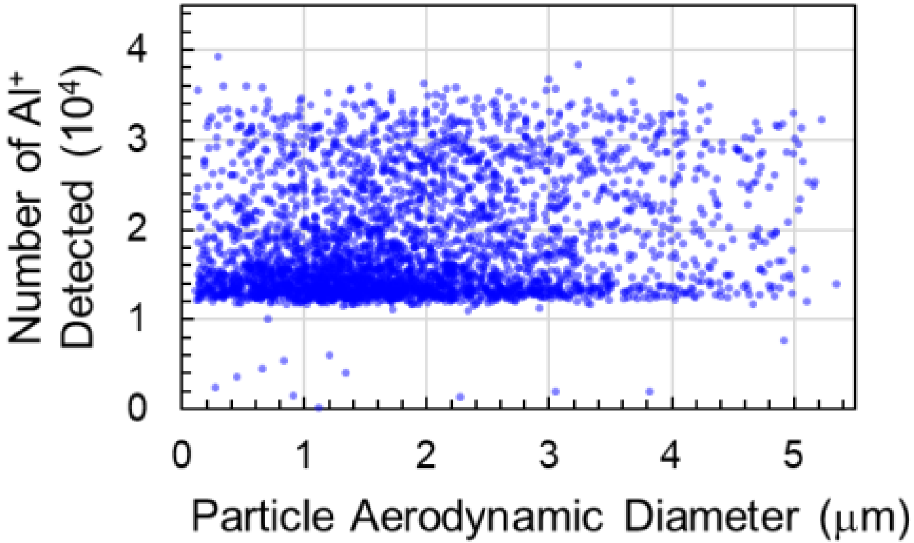


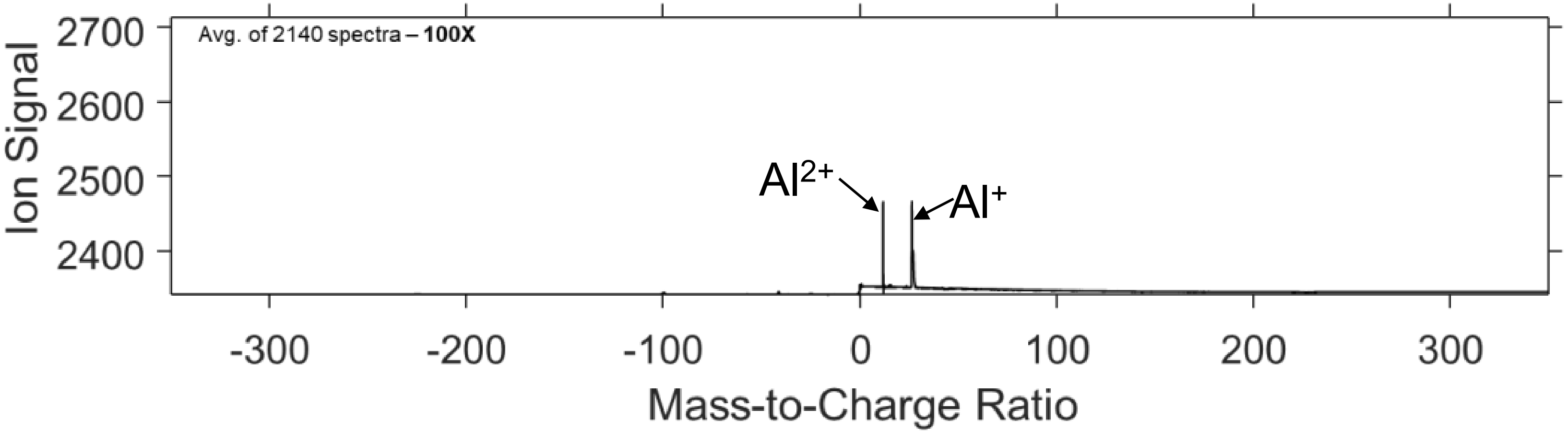


Comparison with Experimental Data

- Spherical Al particles were generated using an $Al(AcAc)_3$ precursor.
- Conversion from signal to total number of ions has significant uncertainty.
- Significant Al^{2+} was unexpected!
- Spectra collected with Livermore Instruments SPAMS 3.0.



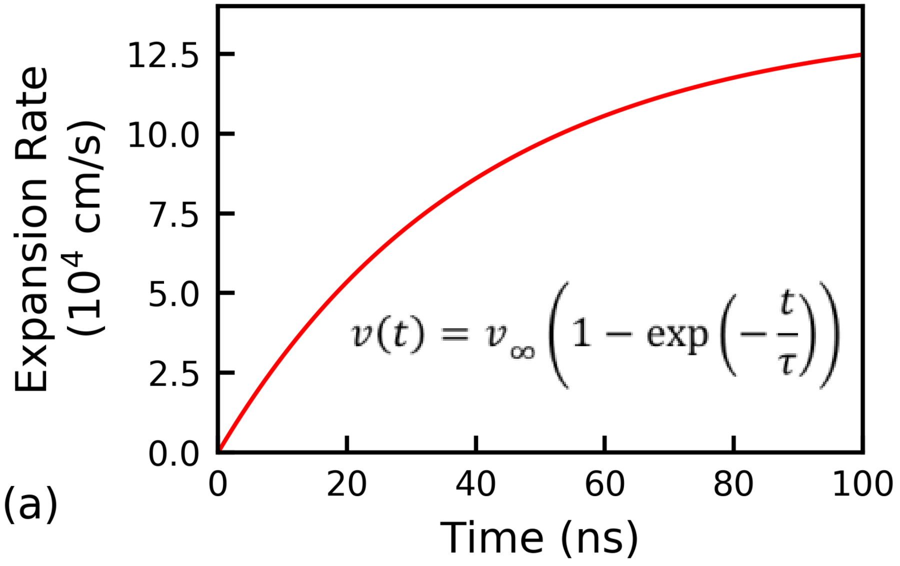


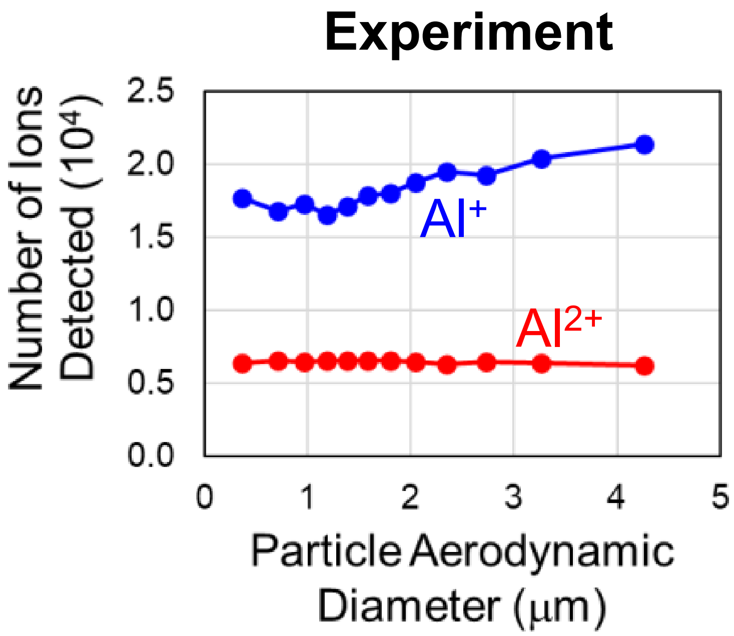


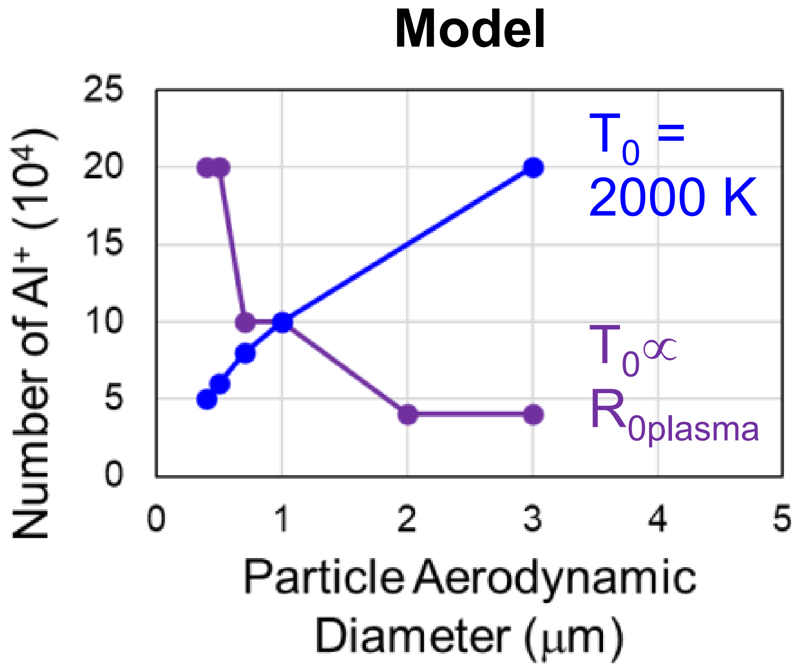
Comparison with Experimental Data

- $E = 2.8$ V/m
- 8 mJ, 248 nm
- Expansion timescales where tuned ($\tau = 40$ ns) to match the magnitude of the experimental data at 1 μm.
- In model, scaling with particle radius depends on the details of the expansion parameters (e.g. value of v_∞).

$$v_\infty = \sqrt{\frac{3k_B T_0}{m_{Al}}}$$

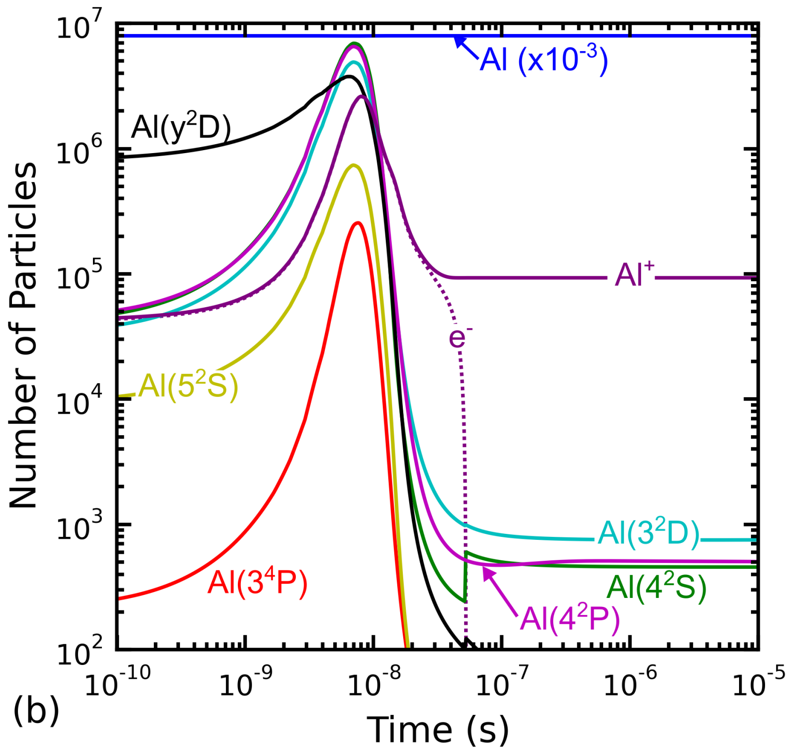






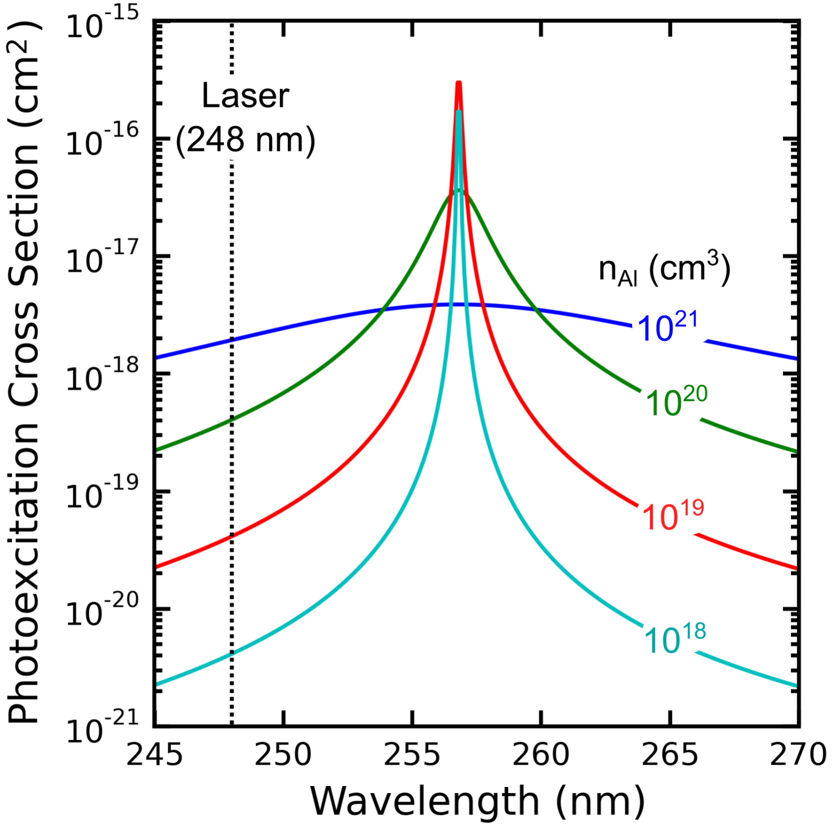
Concluding Remarks

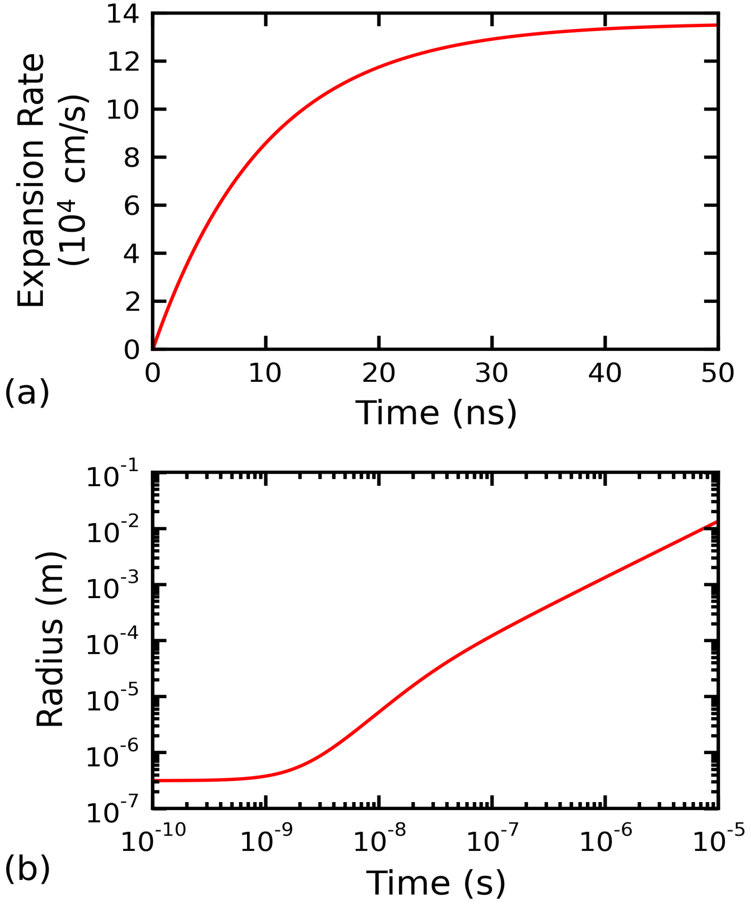
- A method for approximating electron heating and extraction from a plasma expanding into vacuum was developed and tested.
- Ionization mechanisms in a single particle aerosol mass spectrometer (SPAMS) have been modeled.
  - The dominant ionization mechanism is $e^- + Al^*$.
  - The primary role of the electric field is to extract electrons, interrupting recombination.
  - Negligible ionization occurs due to the applied field accelerating the electrons.
- Experimentally, ion counts are only weakly dependent on particle size.
- Important questions:
  - How does 3-body recombination scale with vacuum expansion?
  - What are the timescales, limiting behavior for the expansion into vacuum?
  - Where is the Al^{2+} coming from?



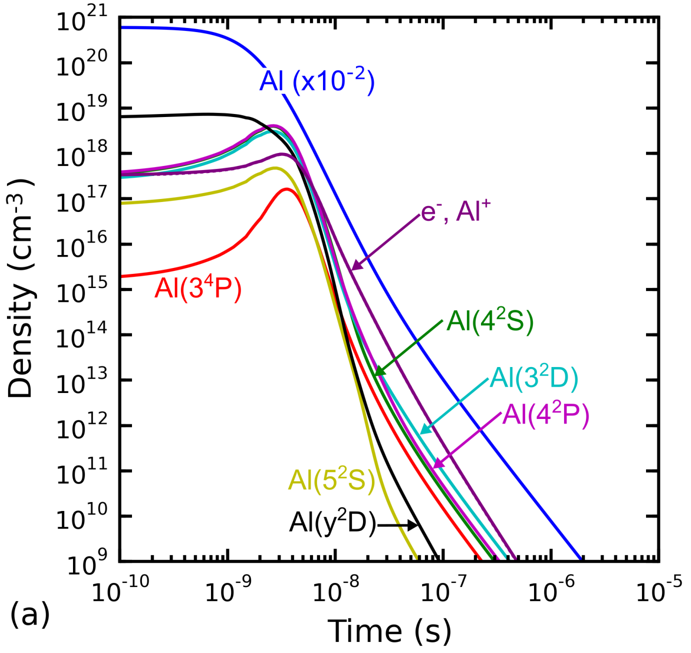
BACKUP

Photoexcitation and Expansion

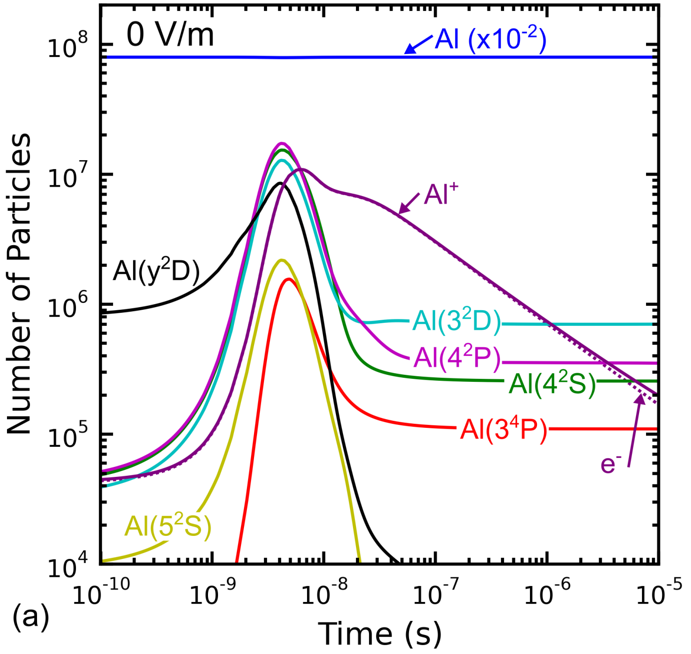




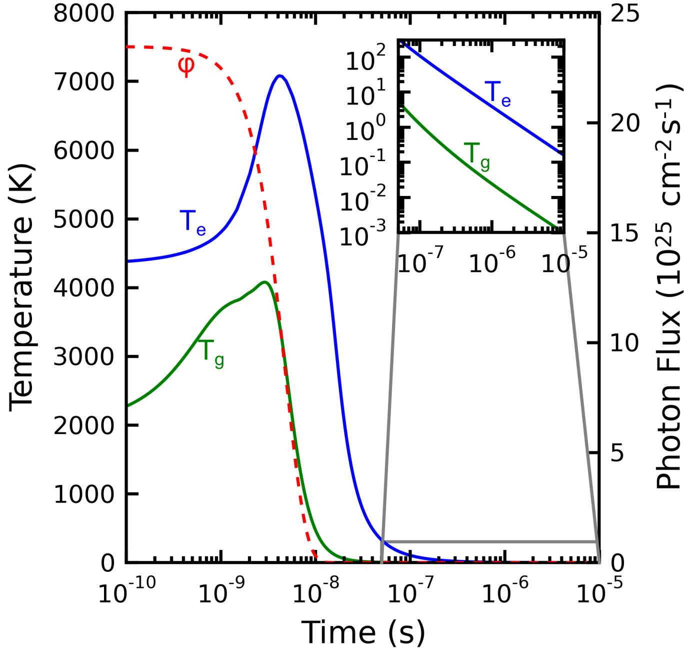
a



Density (cm-3)
Time (s)
Al (x10-2)
e-, Al+
Al(34P)
Al(42S)
Al(32D)
Al(42P)
Al(52S)
Al(y2D)
(a)



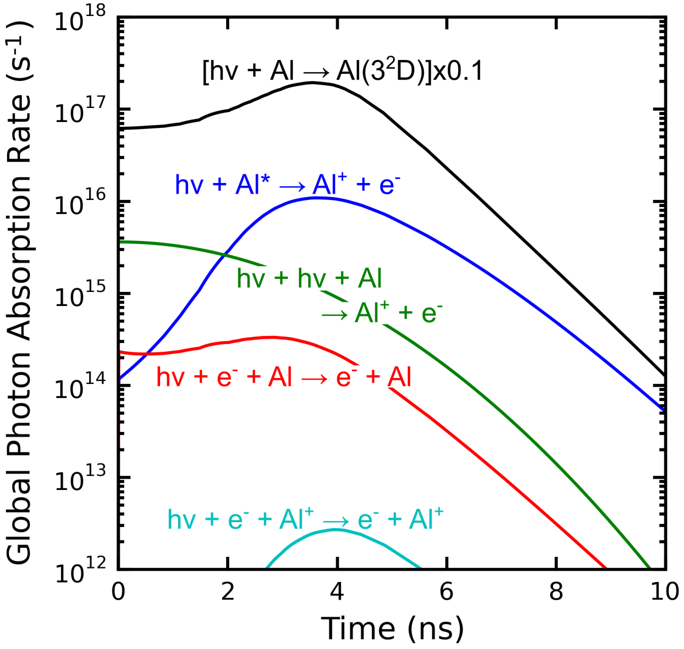
0 V/m
Number of Particles
Time (s)
Al (x10-2)
Al+
Al(y2D)
Al(32D)
Al(42P)
Al(42S)
Al(34P)
e-
Al(52S)
(a)



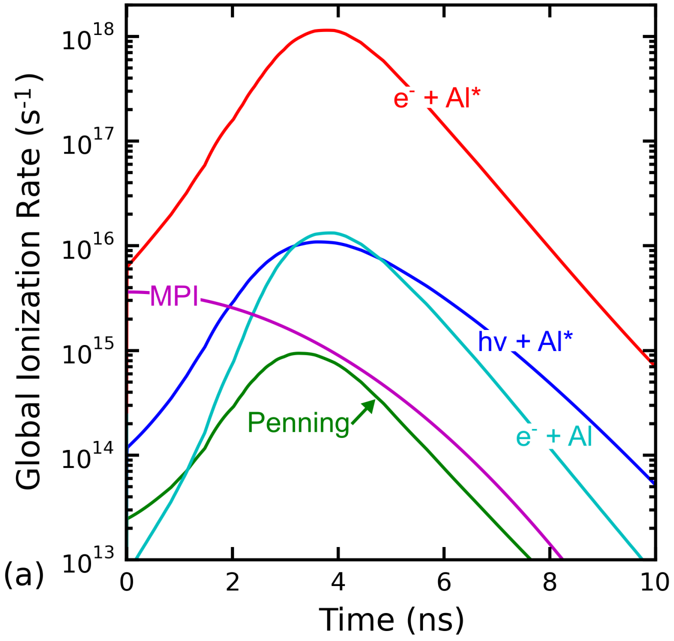
Temperature (K)
Photon Flux (1025 cm-2 s-1)
Time (s)
φ
Te
Tg

a

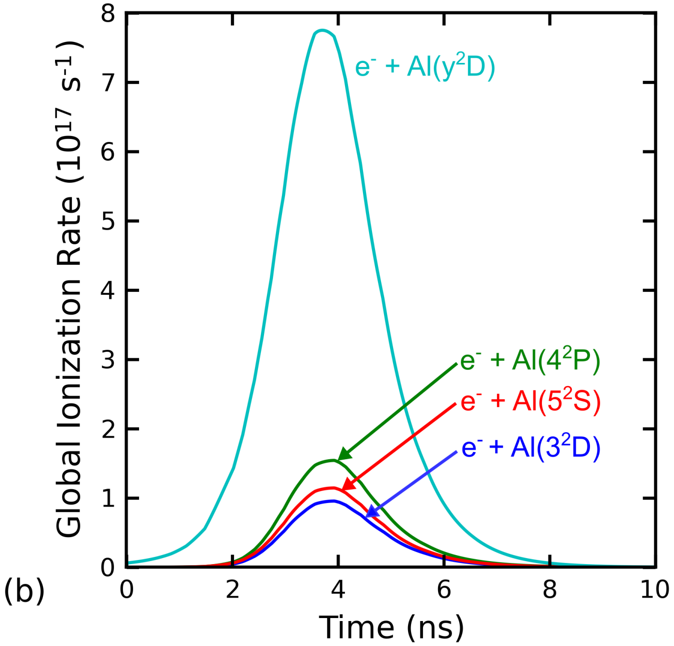
•



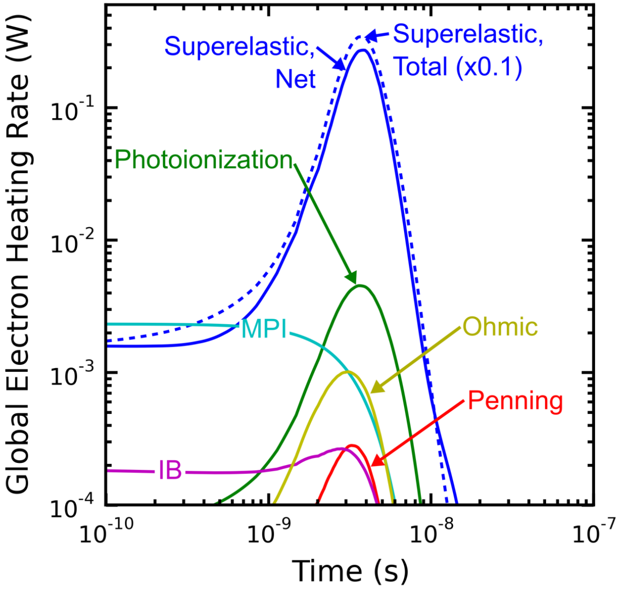
[hv + Al → Al(3^{2}D)]x0.1
hv + Al* → Al$^+$ + e$^-$
hv + hv + Al → Al$^+$ + e$^-$
hv + e$^-$ + Al → e$^-$ + Al
hv + e$^-$ + Al$^+$ → e$^-$ + Al$^+$
Global Photon Absorption Rate (s^{-1})
Time (ns)



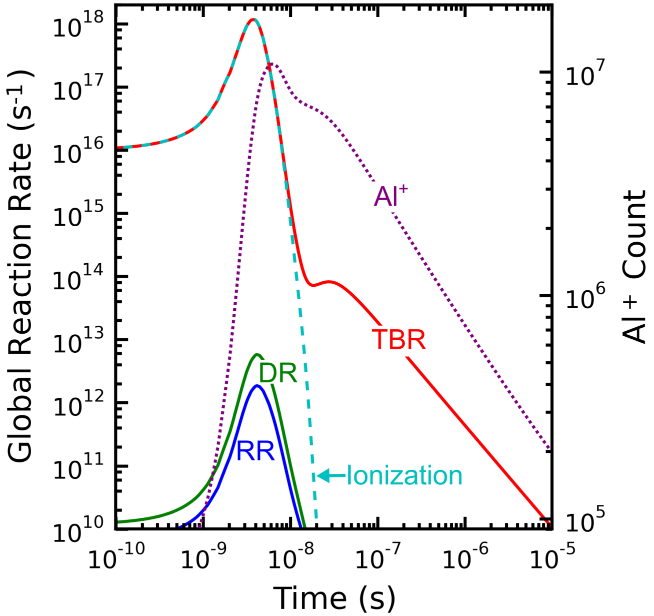
(a)
e$^-$ + Al*
MPI
hv + Al*
Penning
e$^-$ + Al
Global Ionization Rate (s^{-1})
Time (ns)



(b)
e$^-$ + Al(y^{2}D)
e$^-$ + Al(4^{2}P)
e$^-$ + Al(5^{2}S)
e$^-$ + Al(3^{2}D)
Global Ionization Rate (10^{17} s^{-1})
Time (ns)

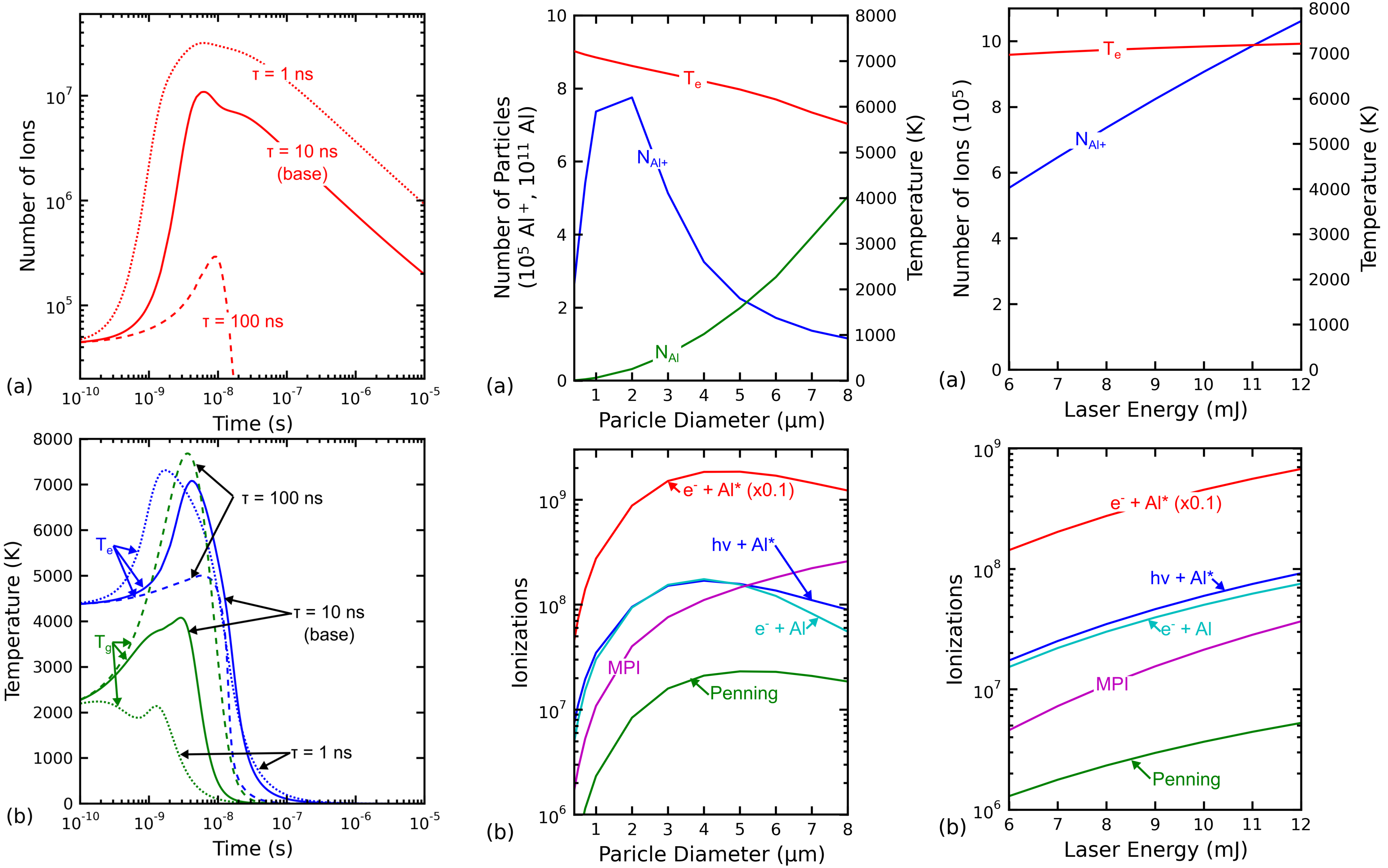


Superelastic, Net
Superelastic, Total (x0.1)
Photoionization
MPI
Ohmic
Penning
IB
Global Electron Heating Rate (W)
Time (s)



Al$^+$
TBR
DR
RR
Ionization
Global Reaction Rate (s^{-1})
Al$^+$ Count
Time (s)

a


Number of Ions
τ = 1 ns
τ = 10 ns (base)
τ = 100 ns
(a)
Time (s)
Temperature (K)
Te
Tg
τ = 100 ns
τ = 10 ns (base)
τ = 1 ns
(b)
Time (s)
Number of Particles (10^5 Al+, 10^11 Al)
Temperature (K)
Te
NAl+
NAl
(a)
Paricle Diameter (μm)
Ionizations
e- + Al* (x0.1)
hv + Al*
e- + Al
MPI
Penning
(b)
Paricle Diameter (μm)
Number of Ions (10^5)
Temperature (K)
Te
NAl+
(a)
Laser Energy (mJ)
Ionizations
e- + Al* (x0.1)
hv + Al*
e- + Al
MPI
Penning
(b)
Laser Energy (mJ)