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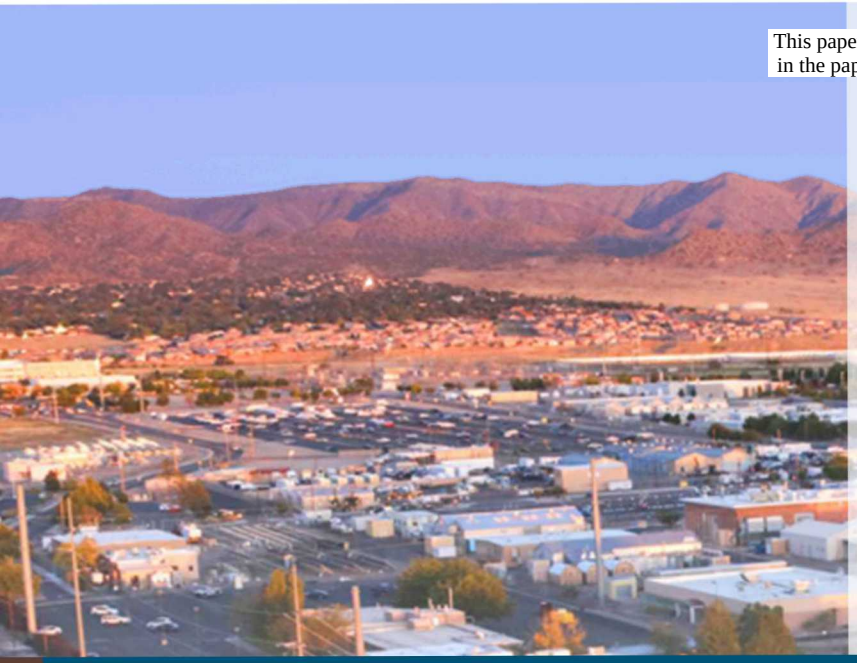

Employing Reduced Chemistry Models on E-Beam/Gas Systems

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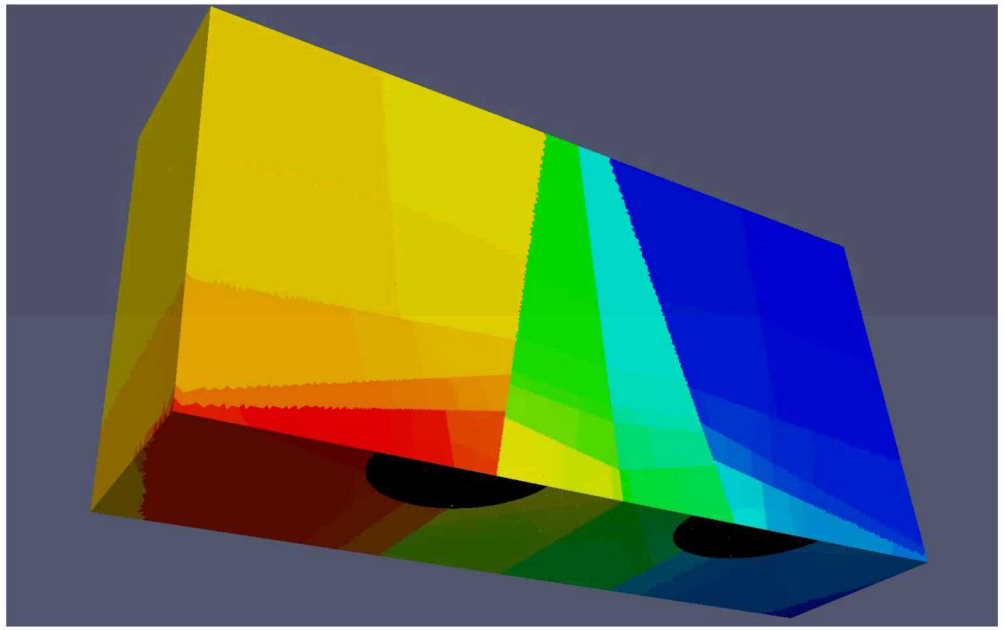
Goals

Electron-neutral interactions are part of many low temperature plasma studies. Most of the simulation techniques used employ energy-dependent cross sections to describe the binary electron-neutral interactions. However, either for computational efficiency purposes, or at a minimum due to lack of data, some inelastic interactions are omitted. To mitigate for the missing interactions an effective elastic collision cross section can be computed that includes the missing inelastic cross sections. While this does not account for the loss of electron energy due to the excitation energy, it does account for some energy loss (to the background gas) that reduces the cumulative energy gain from an applied field. This work aims to demonstrate the impact of not properly accounting for missing cross sections in a system involving the ionization of a low-pressure background argon gas from an injected electron beam.

This work:

- Investigates the importance of more vs. less detailed plasma chemistry models.
- Uses existing electrostatic capability (Aleph) to answer some questions, setting the stage for an electromagnetic capability (EMPIRE) in the future.
- Develops high fidelity model for comparison to future benchtop experiments (from B. Yee).

Aleph Simulation Tool



- 1, 2, or 3D Cartesian
- Unstructured FEM (compatible with CAD)
- Massively parallel
- Hybrid PIC + DSMC (PIC-MCC)
- Electrostatics
- Fixed B field
- Solid conduction
- Advanced surface (electrode) models
- e- approximations (quasi-neutral ambipolar, Boltzmann)
- Collisions, charge exchange, chemistry, excited states, ionization
- Photon transport, photoemission, photoionization
- Advanced particle weighting methods
- Dual mesh (Particle and Electrostatics/Output)
- Dynamic load balancing (tricky)
- Restart (with all particles)
- Agile software infrastructure for extending BCs, post-processed quantities, etc.
- Currently utilizing up to 64K processors (>1B elements, >1B particles)

Aleph Simulation Tool

Basic algorithm for one time step of length Δt:

1. Given known electrostatic field $\mathbf{E}^n$, move each particle for $\frac{\Delta t}{2}$ via:

$$v_i^{n+1/2} - v_i^n + \frac{\Delta t}{2}\left(\frac{q_i}{m_i}\mathbf{E}^n\right)$$

$$x_i^{n+1} = x_i^n + \Delta t v_i^{n+1/2}$$

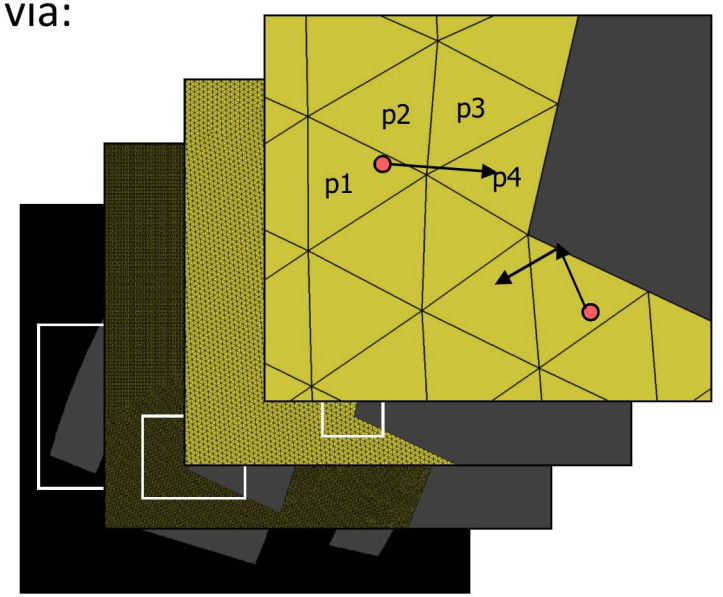
2. Compute intersections (non-trivial in parallel).
3. Transfer charges from particle mesh to static mesh.
4. Solve for $\mathbf{E}^{n+1}$,

$$\nabla \cdot \left(\epsilon \nabla V^{n+1}\right) - -\rho(\mathbf{x}^{n-1})$$

$$\mathbf{E}^{n+1} = -\nabla V^{n+1}$$

5. Transfer fields from static mesh to dynamic mesh.
6. Update each particle for another $\frac{\Delta t}{2}$ via:

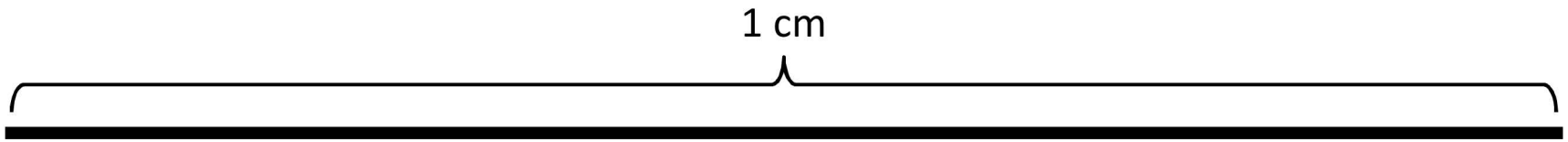
$$v_i^{n+1} - v_i^{n+1/2} + \frac{\Delta t}{2}\left(\frac{q_i}{m_i}\mathbf{E}^{n-1}\right)$$



7. Perform DSMC collisions: sample pairs in element, determine cross section and probability of collision. Roll a digital die, and if they collide, re-distribute energy.
8. Perform chemistry: for each reaction, determine expected number of reactions. Sample particles of those types, perform reaction (particle creation/deletion).
9. Reweight particles.
10. Compute post-processing and other quantities and write output.
11. Rebalance particle mesh if appropriate (variety of determination methods).

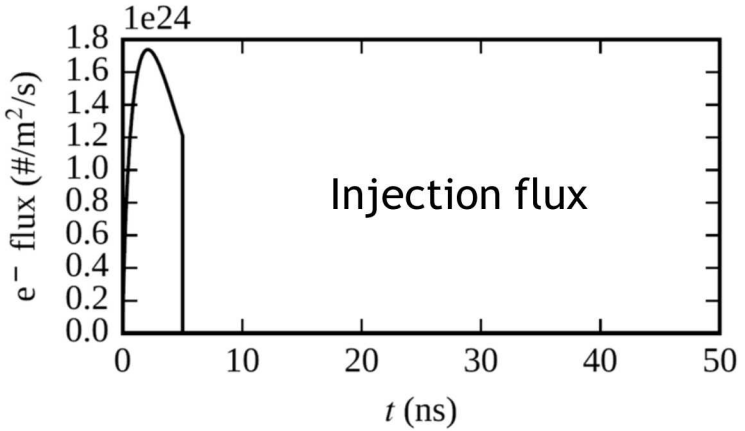
1D Model Setup

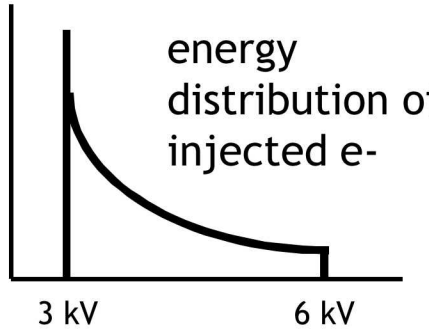
Gap repopulated with 100 mTorr 300 K argon gas



e- injection surface
- $V = 0$
- 3 kV – 6 kV energy distribution
- Flux informed by vacuum current measurements
- Injection time = 5 ns
- All non-Ar particles exit; Ar is specular

target surface
- $V = 0$
- All non-Ar particles exit; Ar is specular





Fullest Argon Chemistry Model (1/3)

IST-Lisbon argon cross sections retrieved from LXCat (includes contributions from multiple sources):

- e- + Ar elastic scattering
- e- + Ar → e- + Ar+ + e- ionization
- 43 e- + Ar → e- + Ar(*x* eV) excitations for:

Ar(3P2)	Ar(4p[5/2]3)	Ar(4p'[3/2]1)	Ar(3d[7/2]4)	Ar(5p[5/2]3)	Ar(5p'[3/2]1)	Ar(6s)
Ar(3P1)	Ar(4p[5/2]2)	Ar(4p'[3/2]2)	Ar(3d[7/2]3)	Ar(5p[5/2]2)	Ar(5p'[1/2]1)	Ar(4d')
Ar(3P0)	Ar(4p[3/2]1)	Ar(4p'[1/2]1)	Ar(3d[3/2]1)	Ar(5p[3/2]1)	Ar(5p'[3/2]2)	
Ar(1P1)	Ar(4p[3/2]2)	Ar(4p'[1/2]0)	Ar(3d'[5/2]2)	Ar(5p[3/2]2)	Ar(5p'[1/2]0)	
Ar(4p[1/2]1)	Ar(4p[1/2]0)	Ar(3d[3/2]2)	Ar(5p[1/2]1)	Ar(5p[1/2]0)	Ar(4d)	

Grouped states: Ar(3d[1/2]0+3d[1/2]1, Ar(3d[5/2]2+5s[3/2]2), Ar(3d[5/2]3+5s[3/2]1), and Ar(3d'[5/2or3/2]3or2+5s'[1/2]0or1)

- These are represented in Aleph according to their excitation energy (from IST-Lisbon), e.g.,
  - species Ar(3P2) maps to Aleph species Ar(11.5480)
  - species Ar(3P1) maps to Aleph species Ar(11.6230)
  - species Ar(3d'[5/2or3/2]3or2+5s'[1/2]0or1) maps to Aleph species Ar(14.2300)
  - species Ar(4d') maps to Aleph species Ar(14.9670)

Fullest Argon Chemistry Model (2/3)

In addition to this we track all the transitions amongst all excited states and the ground state with their Einstein *A* coefficients,

- Ar(11.5480) -/-> (metastable)
- Ar(11.6230) -> Ar(0), 1.320e+08
- Ar(11.7230) -/-> (metastable)
- Ar(11.8280) -> Ar(0), 5.32e+08
- Ar(12.9070) -> Ar(11.5480), 1.89e+07
- Ar(12.9070) -> Ar(11.6230), 5.4e+06

Some grouped states have their rates split and averaged (better ideas?):

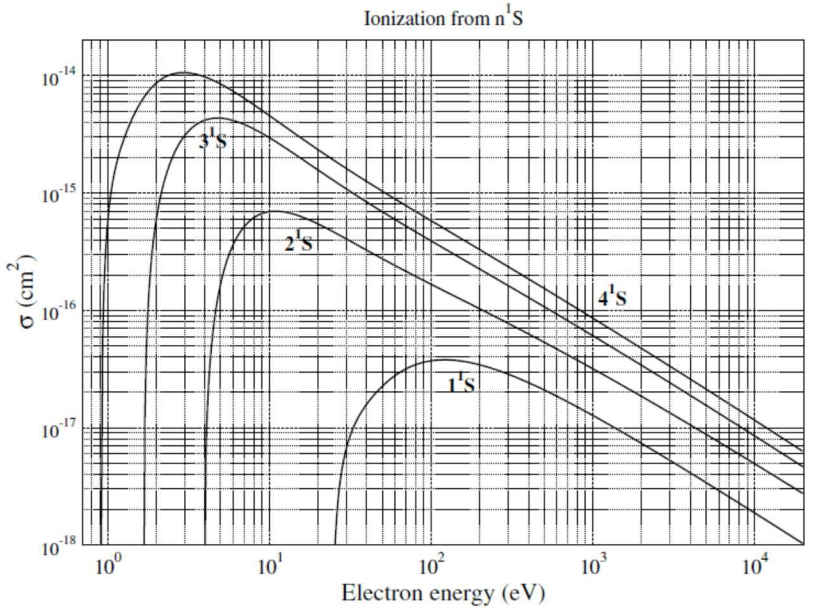
- Ar(14.0900) -> Ar(13.0760), 0.5 * 3.1e+06
- Ar(14.0900) -> Ar(13.0950), 0.5 * (2.0e+06 + 8.9e+06)
- Ar(14.2300) -> Ar(13.1530), 0.25 * (3.69e+05 + 2.22e+06 + 2.8e+05)

A total of 149 transitions.

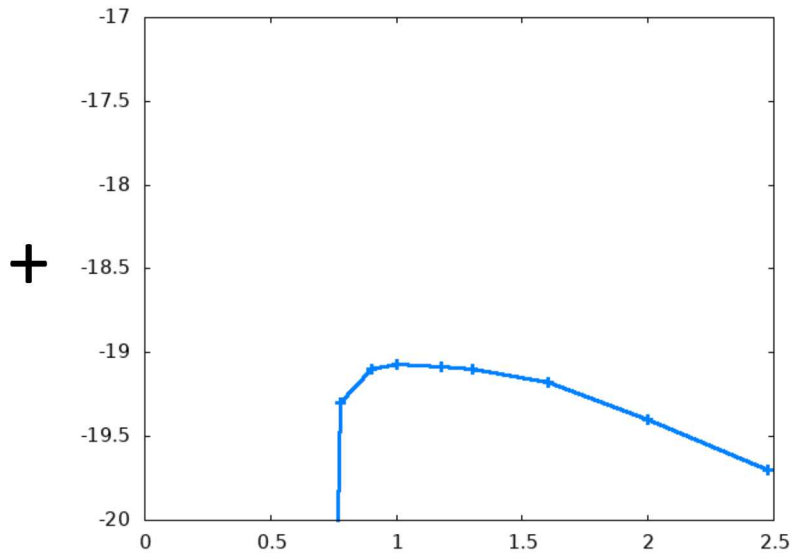
Fullest Argon Chemistry Model (3/3)

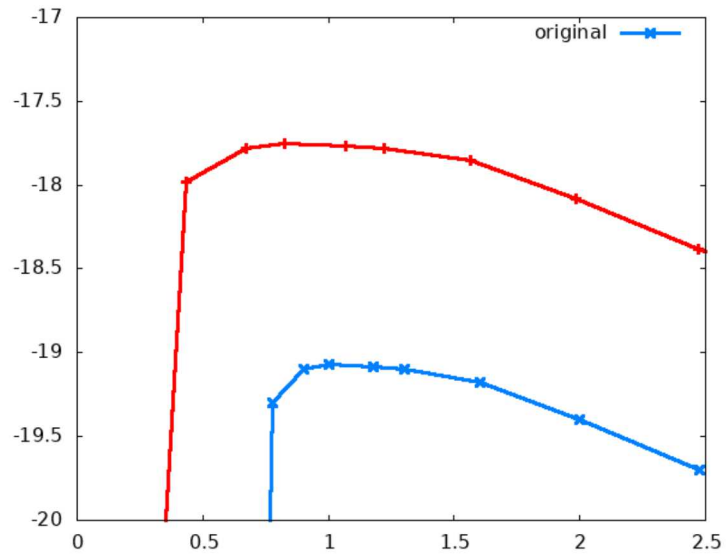
To account for multistep ionization we construct an ionization cross section for every excited state. Most of these do not exist so these are artificially derived.

- Shift cross section excitation threshold to ionization energy for excited state.
- Multiply entire cross section by factor derived from Ralchenko helium work.



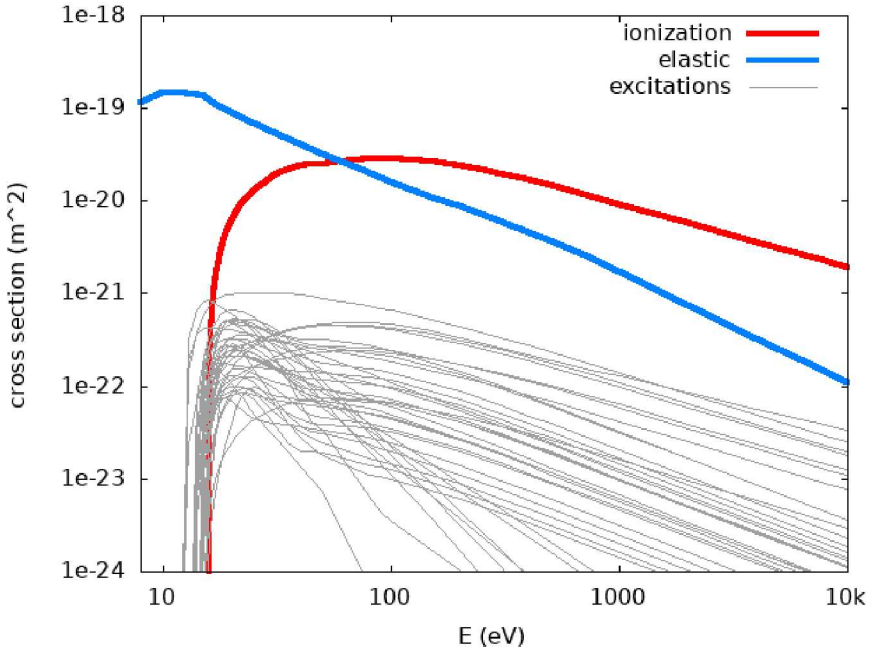
Graph 41. Recommended electron-impact ionization cross sections from atomic terms of He I with $n \leqslant 4$.





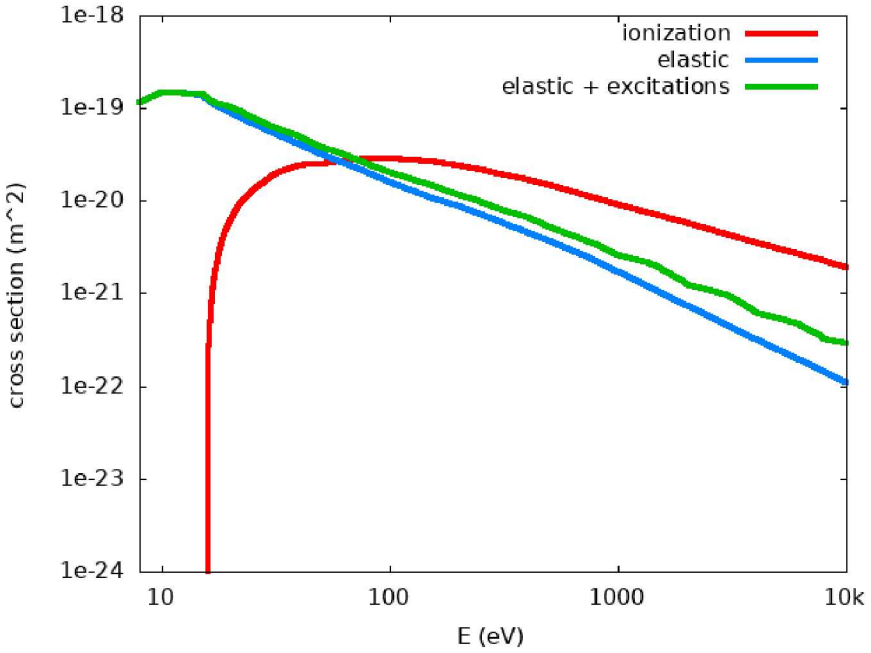
Argon Chemistry Models

Full chemistry



- 43 excited states and excitation cross sections
- 1 + 43 ionization cross sections
- 149 radiative transitions

Simplified and Summed



- 0 excited states

Simplified:

- Use raw elastic cross section

Summed:

- Add excitation cross sections to elastic cross section for a summed elastic cross section

- All chemistries include enhanced recombination, e- + Ar+ + e- → e- + Ar, rate = 8 x 10^{-32} m^6/s

Simulation Constraints

Constraint	Requirement
Debye length	$\Delta x < \lambda_D = \sqrt{\frac{k_B T_e \varepsilon_0}{n_e q_e^2}}$
Collision mfp	$\Delta x < \lambda_c = \frac{1}{n_{bg}\sigma_{max}}$
Particle CFL	$\Delta t < \frac{v_{max}}{\Delta x}$
Plasma frequency	$\Delta t < 2/\omega_p = 2\sqrt{\frac{\varepsilon_0 m_e}{n_e q_e^2}}$
Collision frequency	$\Delta t < 1/\omega_c = \frac{1}{n_{bg}\sigma_{max} v_{max}}$

At T_e = 2 eV, n_e = 10^{19} m^{-3}, and n_{bg} = 3.2 x 10^{21} m^{-3}, v_{max} = 4.6 x 10^7 m/s (6 keV), and $(\sigma v)_{max}$ = 3.2 x 10^{-13} m^3/s

λ_D = 3.3 µm, λ_c = 1600 µm → Δx = 3 µm

$\Delta x/v_{max}$ = 65 fs, $2\pi/\omega_p$ = 35 ps, $1/\omega_c$ = 990 ps → Δt = 60 fs

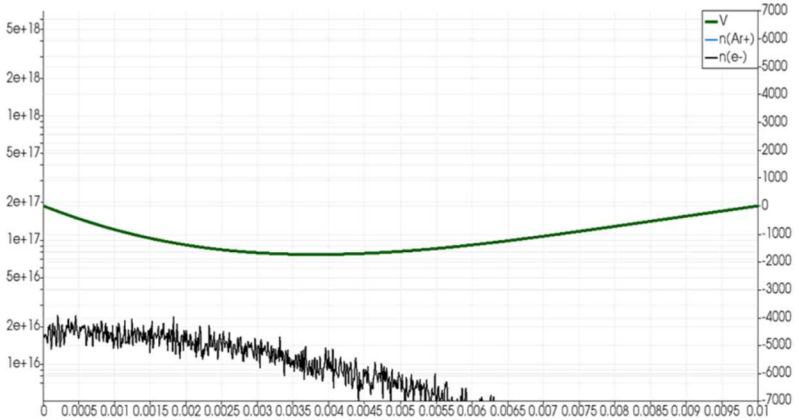
64 e-/cell, 10 Ar/cell, 32 other heavy species/cell

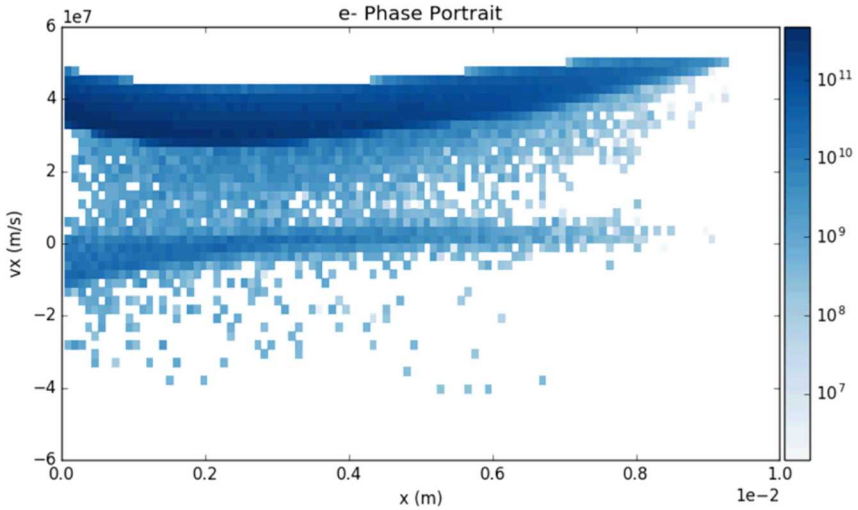
512 cores, direct solver, 26h for full chemistry to 500 ns

(results compared to Δx = 1 µm and Δt = 6 fs simulations, and with different random number seeds)

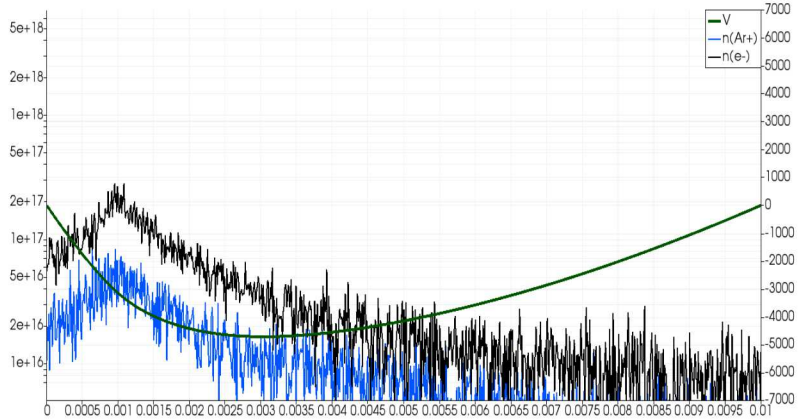
Simulation Evolution (full chemistry)

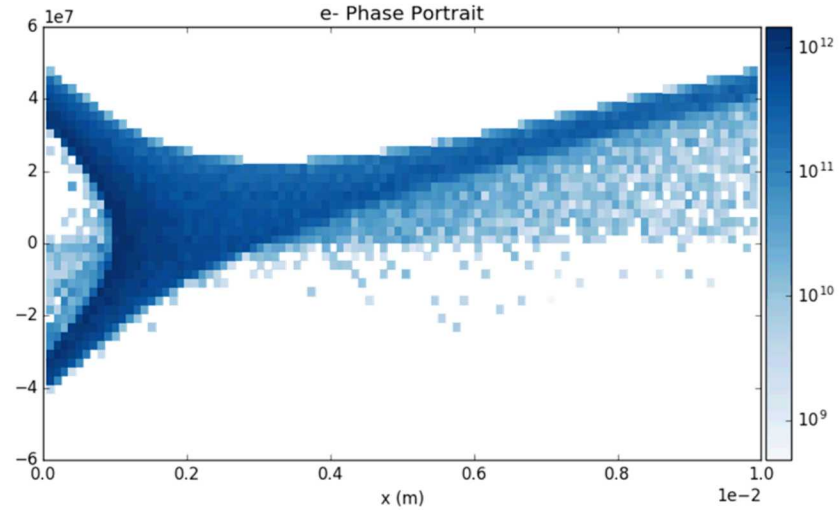
t = 0.2 ns



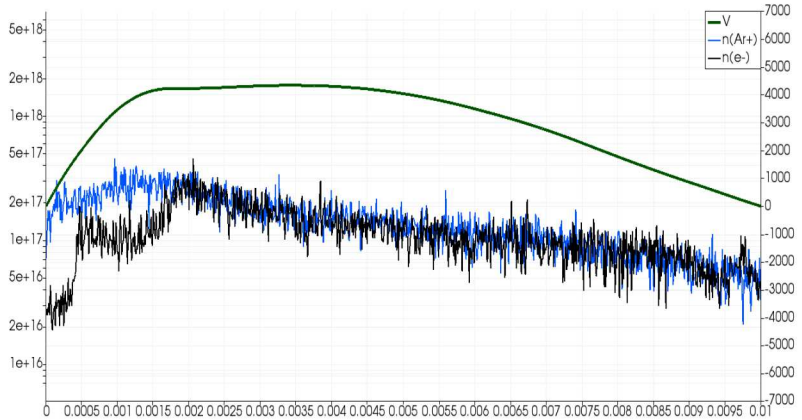


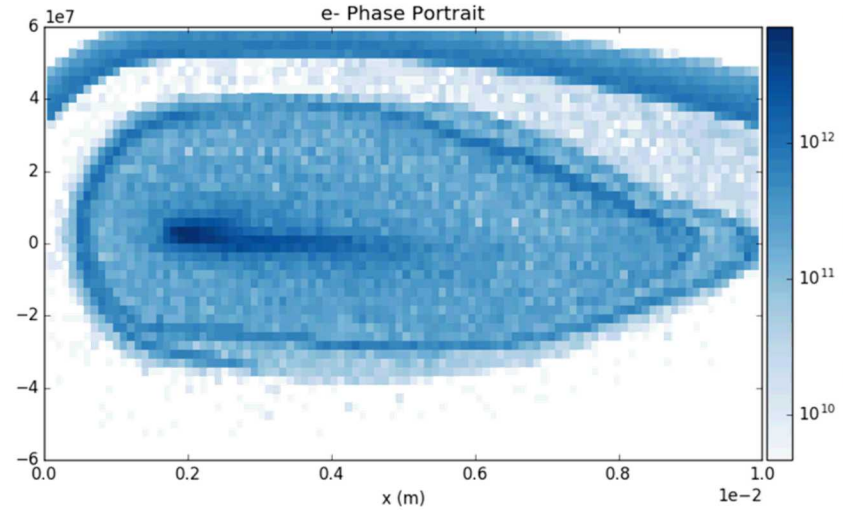
t = 1 ns





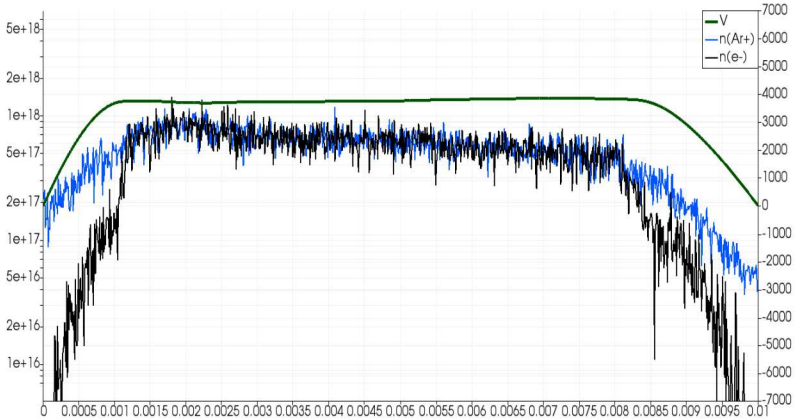
t = 5 ns (beam off)

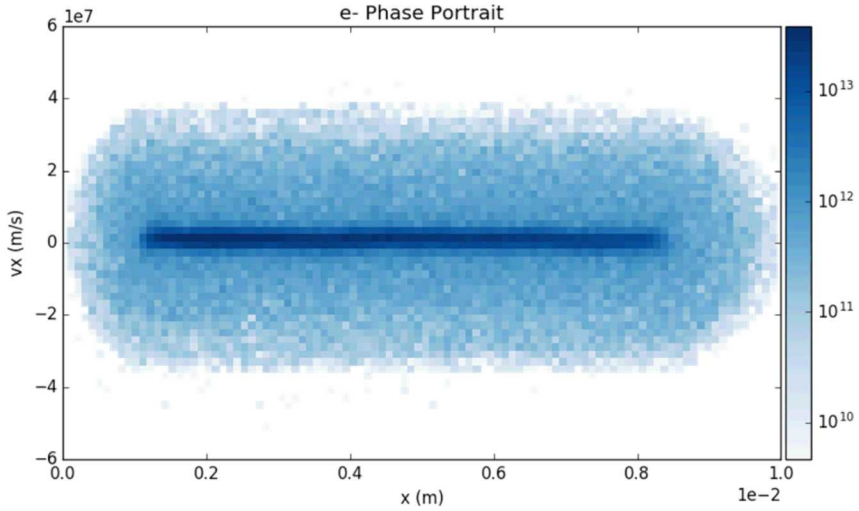




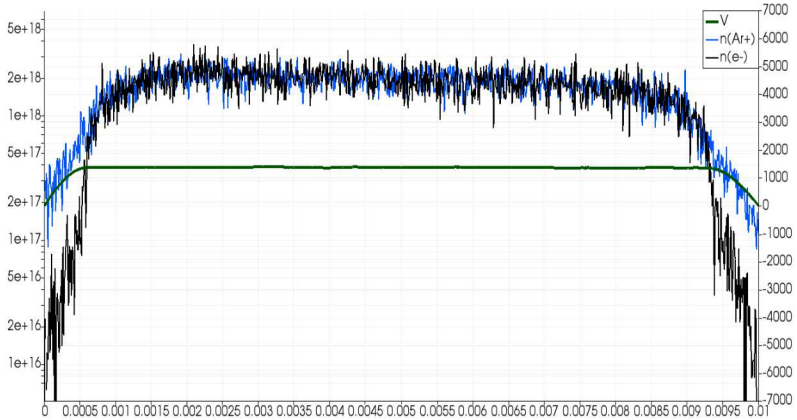
Simulation Evolution (full chemistry)

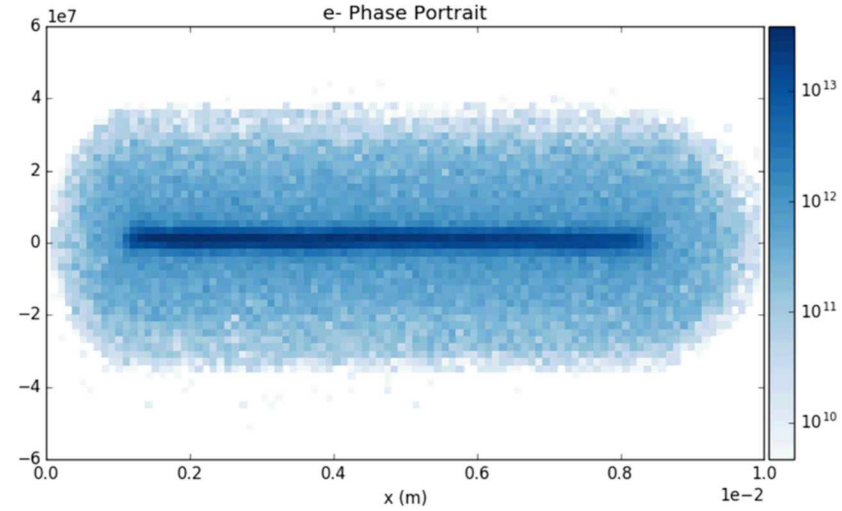
t = 10 ns



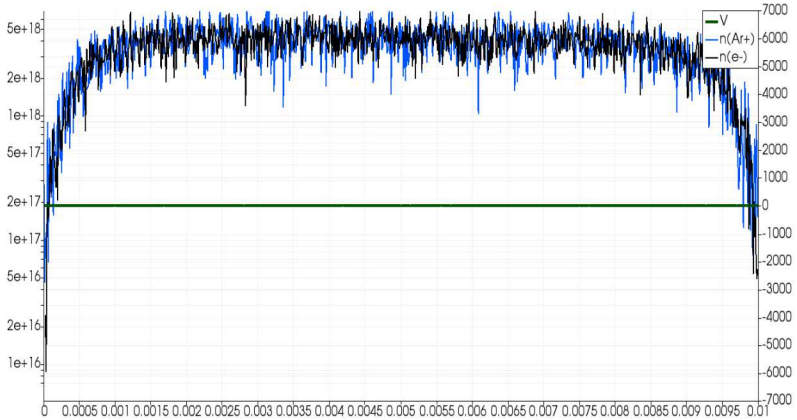


t = 20 ns

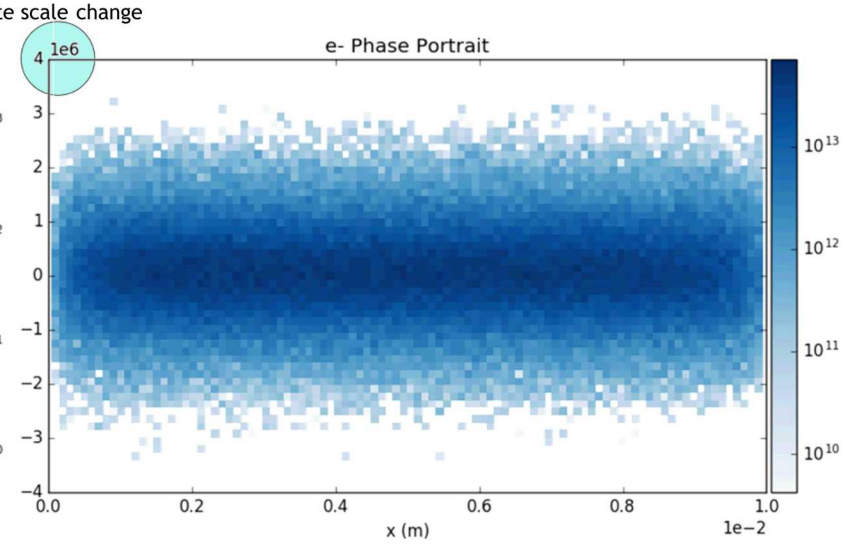




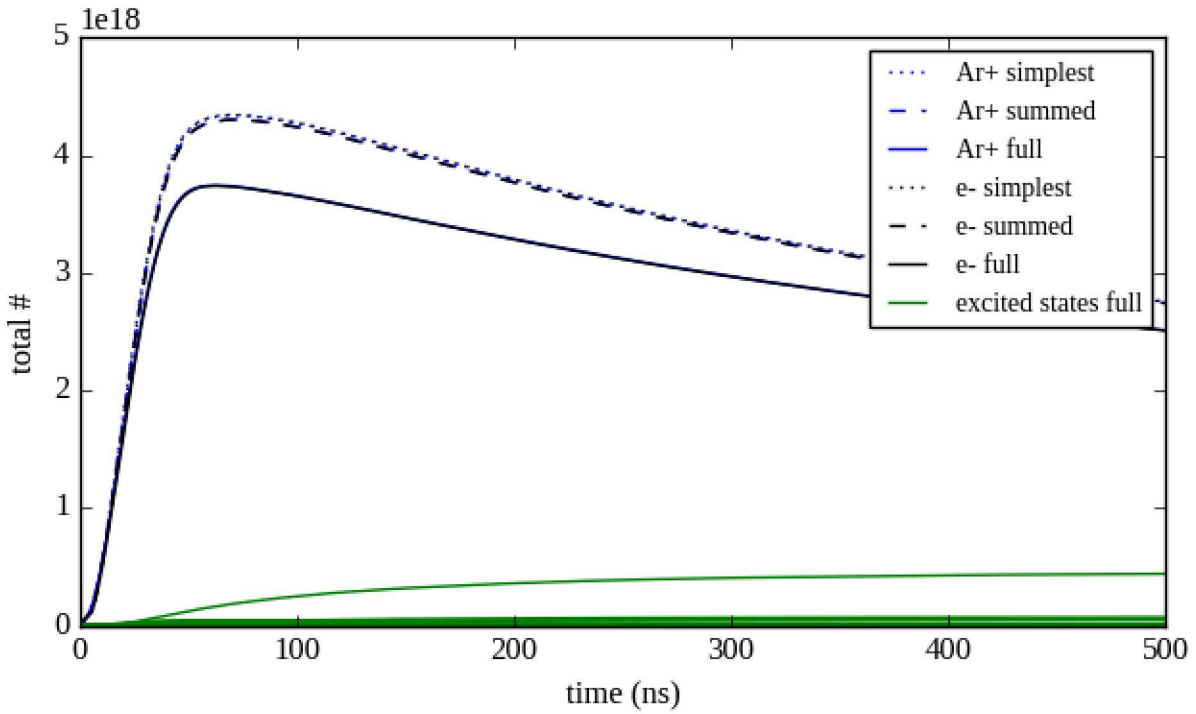
t = 100 ns

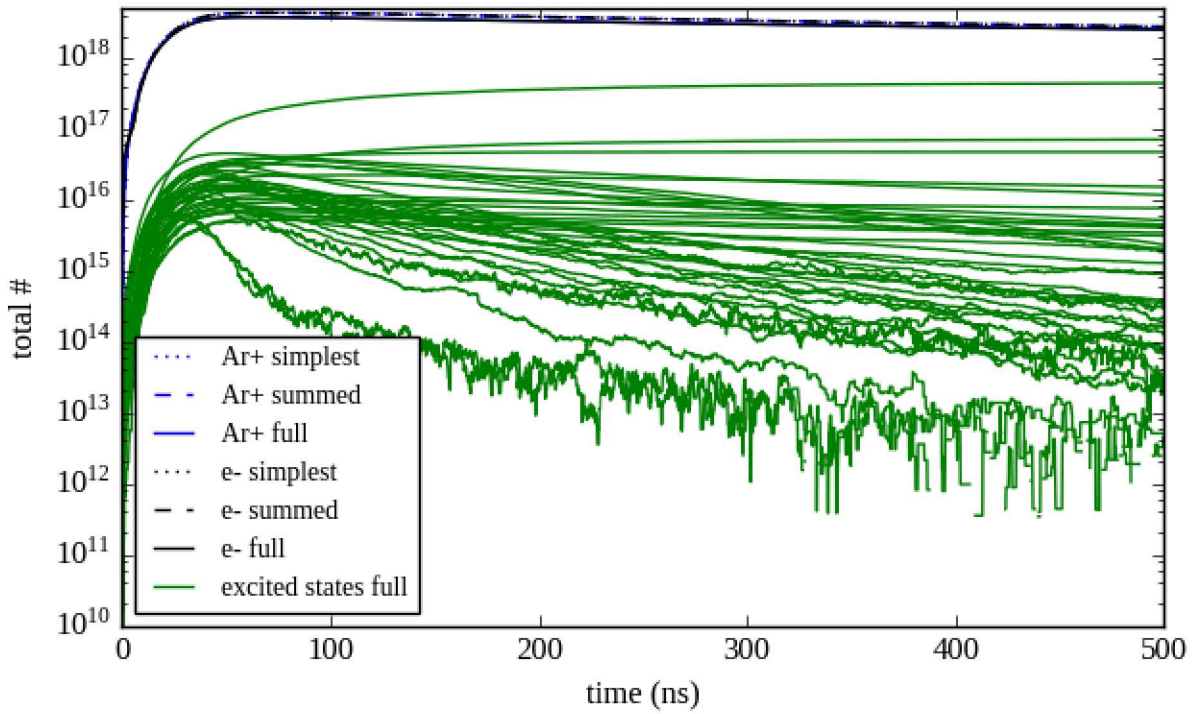


Note scale change



Results





Linear (left) and logscale (right) species counts over entire 1 cm domain. Note the large family of excited states in the full chemistry system. Excited states are reduced primarily via radiative decay although each species can ionize through electron impact.

Observations

- The simplest chemistry and the summed excited state chemistry produced nearly identical results. This was moderately surprising.

- The full chemistry produced slightly less plasma (e.g., lower n_e), likely due to loss of energy to tracked inelastic processes.

- Plasma generation is primarily caused by the slowing down of the e- population due to large negative space charge, not due to high energy e- “cascades”. The e- energies get reduced to much lower energies (and in fact some e- are reflected) where the cross sections are higher.

- Pulse length and cavity gap are critical for creating populations of slower e-.

Thank You!