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Comparison of Aleph and BOLSIG+ Results for Electron-Nitrogen Chemistry

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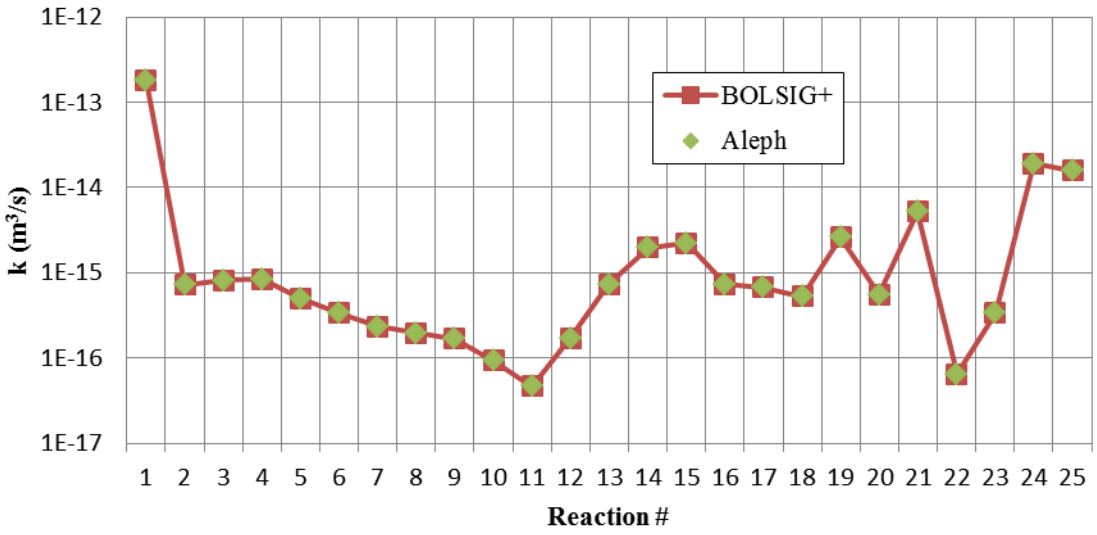
EXECUTIVE SUMMARY

The goal of this report is to document the current status of Aleph with regards to electron collisions under an electric field. Aleph and the community-accepted BOLSIG+ code are both used to compute reactions rates for a set of 25 electron-nitrogen interactions. A reasonable comparison is found (see below) providing evidence that Aleph is successfully simulating or implementing:

- Particle-particle collision cross-sections via DSMC methodology
- Energy balance for simple particle interactions
- Electron energy distribution function (EEDF) evolution



Comparison of BOLSIG+ and Aleph reaction rates.

INTRODUCTION

The purpose of this report is to document the comparison between Sandia's Aleph code and the community-accepted BOLSIG+ code [1] (http://www.bolsig.laplace.univ-tlse.fr/) in computing reaction rates for a set of electron-nitrogen interactions. A set of 25 interactions from the lxcat database (and referenced to [2] and [3]) is used,

1. $e^- + N_2 \rightarrow e^- + N_2$ (elastic)
2. $e^- + N_2 \rightarrow e^- + N_2^*$(rotational)
3. $e^- + N_2 \rightarrow e^- + N_2^*$(v=1,resonance)
4. $e^- + N_2 \rightarrow e^- + N_2^*$(v=1)
5. $e^- + N_2 \rightarrow e^- + N_2^*$(v=2)
6. $e^- + N_2 \rightarrow e^- + N_2^*$(v=3)
7. $e^- + N_2 \rightarrow e^- + N_2^*$(v=4)
8. $e^- + N_2 \rightarrow e^- + N_2^*$(v=5)
9. $e^- + N_2 \rightarrow e^- + N_2^*$(v=6)
10. $e^- + N_2 \rightarrow e^- + N_2^*$(v=7)
11. $e^- + N_2 \rightarrow e^- + N_2^*$(v=8)
12. $e^- + N_2 \rightarrow e^- + N_2^*(A^3\Sigma, v=0\text{-}4)$
13. $e^- + N_2 \rightarrow e^- + N_2^*(A^3\Sigma, v=5\text{-}9)$
14. $e^- + N_2 \rightarrow e^- + N_2^*(B^3\Pi)$
15. $e^- + N_2 \rightarrow e^- + N_2^*(W^3\Delta)$
16. $e^- + N_2 \rightarrow e^- + N_2^*(A^3\Sigma, v>9)$
17. $e^- + N_2 \rightarrow e^- + N_2^*(B'^3\Sigma)$
18. $e^- + N_2 \rightarrow e^- + N_2^*(a'^1\Sigma)$
19. $e^- + N_2 \rightarrow e^- + N_2^*(a^1\Pi)$
20. $e^- + N_2 \rightarrow e^- + N_2^*(w^1\Delta)$
21. $e^- + N_2 \rightarrow e^- + N_2^*(C^3\Pi)$
22. $e^- + N_2 \rightarrow e^- + N_2^*(E^3\Sigma)$
23. $e^- + N_2 \rightarrow e^- + N_2^*(a''^1\Sigma)$
24. $e^- + N_2 \rightarrow e^- + N_2^*$(sum of singlet states)
25. $e^- + N_2 \rightarrow e^- + N_2^+$ (ionization)

Aleph and BOLSIG+ both incorporate these interactions and arrive at estimates of each reaction rate. BOLSIG+ uses very different solution methods than Aleph so a good comparison between them is stronger evidence that they are operating as intended than two codes that implement the same solution methodology (e.g., one DSMC code vs. another DSMC code). BOLSIG+ uses PDE-based solution methods and will not be discussed further. See their information for more details.

SETUP

A 3D mesh domain is utilized, a $(100\ \mu m)^3$ cube discretized into 12 tetrahedral elements (9 nodes). The positions of all particles remain fixed throughout the simulation, but the velocities of electrons are allowed to evolve in time converging to a stationary electron energy distribution function (EEDF). Because all positions are fixed we are simulating a 0D box of interacting particles – a 0D3V simulation.

Boundary conditions are set to impose a fixed electric field of 3.21883 MV/m by setting a 0V boundary condition at one face and 321.883V at the other one 100 μm away. The Poisson solve is only performed at initialization and then held fixed to mimic the fixed E-field assumption in BOLSIG+. A background of neutral N_2 particles is uniformly distributed throughout the domain with a density of 3.21883 x $10^{24}/m^3$ and temperature of 0K. A background of electron particles is also uniformly distributed throughout the domain with a density of $10^{16}/m^3$ and a temperature of 0K. See Figure 1 for a graphical representation of the model. The final EEDF should be independent of the initial electron temperature (unless it is set so high that it bypasses all of the cross-sections). The final EEDF should also be essentially independent of the neutral temperature, as collision dynamics are based on center-of-mass energies and the accelerated electron energies will dominate.

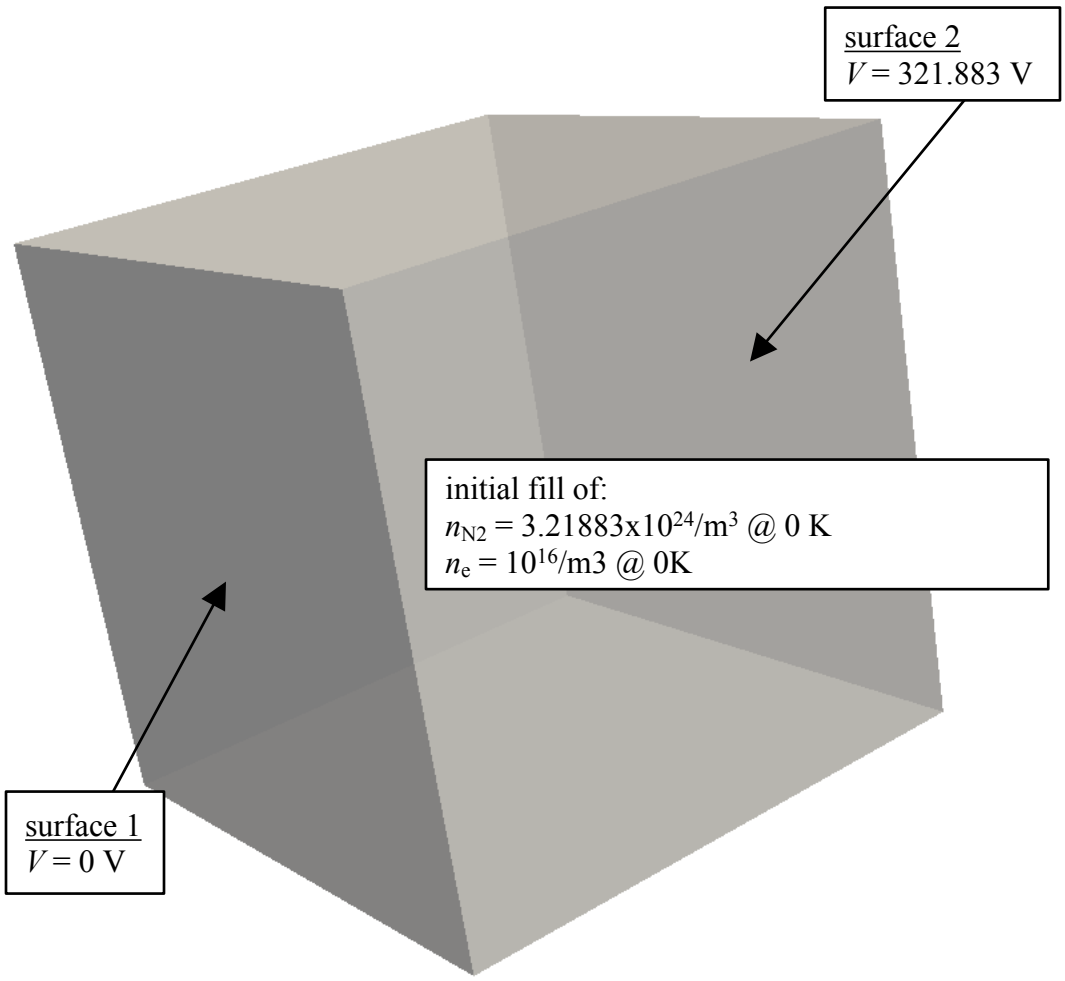


Figure 1. Geometry, boundary conditions, and initial condition for model problem.

The electrons are under constant acceleration (adding energy), and colliding with the background N_2 particles according to the cross-sections 1-25 above (losing energy). The EEDF will converge to a steady state EEDF (but of course include statistical variations because our solution method is

stochastic). The frequency of successful collision events for each interaction is tracked, averaged over time, and then converted into a reaction rate for comparison to BOLSIG+.

An earlier modeling approach did not used fixed electron background density, but instead injected electrons at the low potential side of a 3D (100 μm) x (100 μm) x (1 mm) extended brick domain (see Figure 2). The injection model would necessarily have a "relaxation" phase where the EEDF evolves from an assigned one (electron injection conditions) to another EEDF near the outflow (high potential) surface. Because we compute global interaction frequencies, it would always be necessary to include this transition domain in our calculations if we used this 3D3V model. Therefore, we switched to the 0D3V model described earlier.

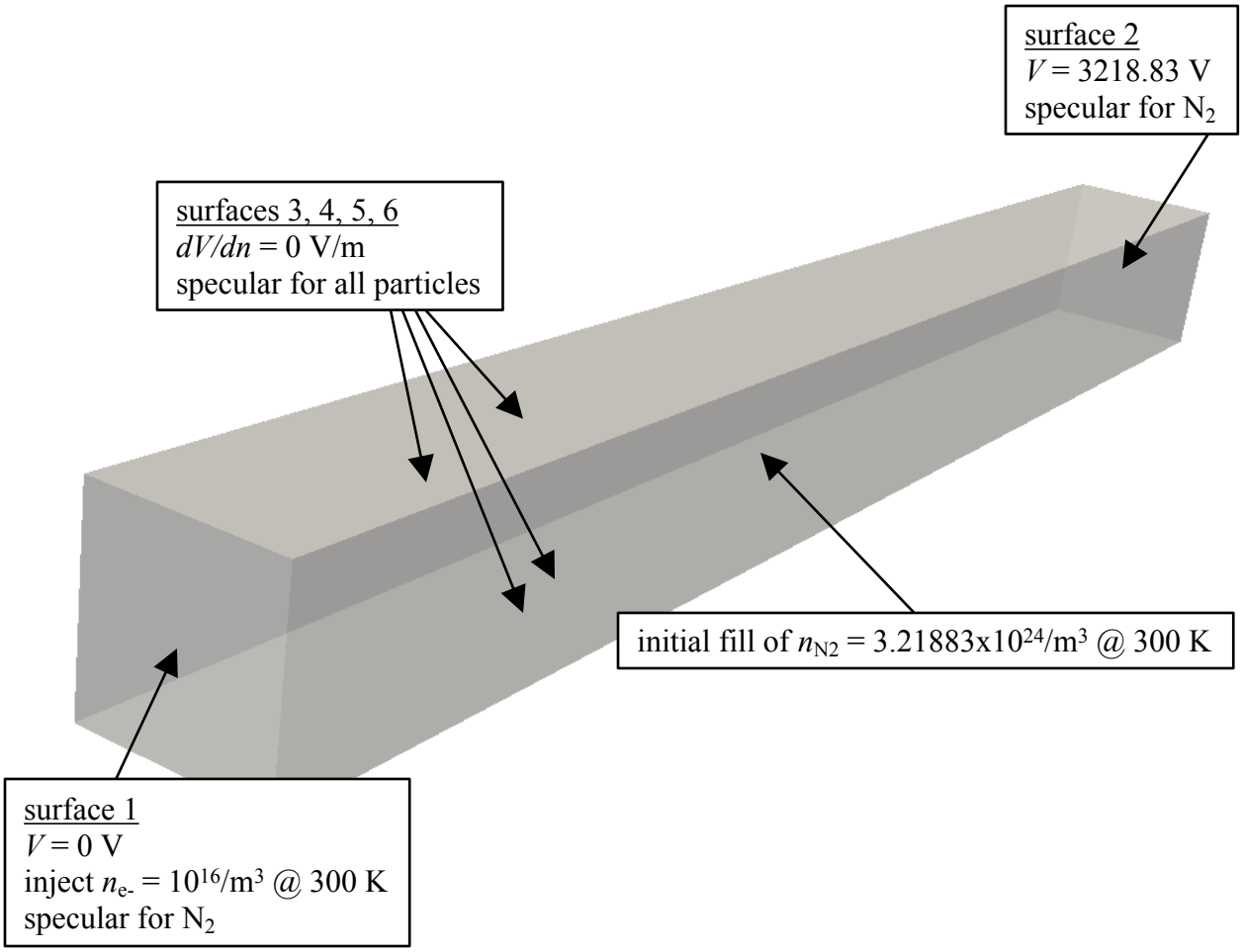


Figure 2. Geometry, boundary conditions, and initial condition for model problem.

Specific input deck lines motivating additional comments are provided here. The full input deck appears on page 12.

```
5   particle position update = off
```

This is the special Aleph line command that turns off all particle updates. The velocities are updated (both due to collisions and acceleration), but the position updates are discarded.

```
11  timestep size = 1e-13
```

The neutral density is $\sim 10^{24}/m^3$. If each electron-N_2 interaction were considered separately, a typical low collision rate would have 10^8 collisions/s, while a typical high collision rate would have 10^{11} collisions/s. Our timestep of 100 fs (10^{-13} s) would then produce 1 collision every ~100,000 timesteps for the low rate and 1 collision every ~100 timesteps for the high rate. We resolve the highest collision frequency (by approximately a factor of 100), even though the DSMC method does not explicitly require it.

```
12  total number of timesteps = 1000000
```

A critical question is how much time is required for the EEDF to reach a stationary solution (one that is centered statistical variation about a converged mean). The smallest frequency reaction would indicate a timescale of 10 ns per collision, if it were the only interaction. However, the real time to converge the EEDF is a function of all of the interactions – the other 24 interactions greatly influence the availability of electrons of appropriate energies for the lowest frequency reaction (and it technically influences the convergence of all the other reaction rates). We take 1,000,000 time steps resulting in a full simulation time of 100 ns, or 10 times the slowest interaction timescale. The averaging begins at 50 ns, or 5 times the slowest timescale.

```
18  exodus output stride =          off
```

Unlike most simulations, we're only looking at total interaction counts and rates, so we don't include any exodus output (e.g., for visualization).

```
19  potential field solve stride = initial
```

Usually we would solve for an electric field every timestep, but here we enforce a fixed electric field by solving the electric field only once at the beginning of the simulation and then use the same field for the duration of the simulation.

```
20  interaction stride =             1
```

An interaction stride of 1 gives the greatest collision interaction fidelity.

```
22  restore energy stride =         off
```

Because of our special interaction "products" we will not need to account for energy loss. Every electron-N_2 interaction is handled by performing the probabilistic interaction, but at the conclusion all N_2 products (ions, excited states, etc.) are ignored and only the original N_2 is retained. This guarantees we have a proper constant background of N_2.

```
26  initial N2 density = 3.218830e24, T = 0, exact_number = true
27  initial e- density = 1.0e16, T = 0, exact_number = true
```

We seed the initial domain with uniform N_2 and e- particles via the `exact_number = true` option command. This insures that we get the most accurate number of particles instead of the default purely probabilistic particle creation.

```
34  particle weighting for N2 = 3.218830E+8
35  particle weighting for e- = 1E0
```

Because we are looking for reaction rates (and not absolute densities of anything) the particle weight for e- is a matter of convenience, and using a weight of 1.0 results in simpler arithmetic in the rate calculations. Similarly, the weight for N_2 was chosen to produce the correct density with a good number of neutrals. There are 10,000 each of e- and N_2 in the simulation.

```
44  interact e- + N2,            interaction_name = 1,
    interaction_model = elastic_isotropic_scattering,
    cross_section_file = 1.data,
    fixed_heavy_particle_properties = true
45  interact e- + N2,            interaction_name = 2,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 2.data,  heat_of_reaction = 3.204353E-21,
    fixed_heavy_particle_properties = true
.
.
.
67  interact e- + N2,            interaction_name = 24,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 24.data, heat_of_reaction = 2.082830E-18,
    fixed_heavy_particle_properties = true
68  interact e- + N2 -> e- + N2, interaction_name = 25,
    interaction_model = ionization,
    cross_section_file = 25.data, heat_of_reaction = 2.499395E-18,
    fixed_heavy_particle_properties = true
```

The input lines for each of the 25 interactions with the data file name #.data and heat of reaction (electron energy loss) for the inelastic and ionization collisions. The first is elastic scattering, the last is ionization, and all the others are inelastic scattering. The use of `fixed_heavy_particle_properties = true` forces the simulation to leave the neutral collision participant (the heavy one of the pair) with the same energy it had before the collision. This ensures the original neutral energy distribution is preserved. The use of `e- + N2 -> e- + N2` for the ionization instead of the expected `e- + N2 -> e- + e- + N2+` means we don't produce any actual ions and again leave the product neutral species alone.

```
71  output global computational_count for e-, name = e-_comp_count,
    window = discrete, size = {banner_stride}, stride = {banner_stride}
72  output global computational_count for N2, name = N2_comp_count,
    window = discrete, size = {banner_stride}, stride = {banner_stride}
```

Looking at the number of computational particles is helpful to ensure the simulation is operating properly and not creating any new particles. These counts remain fixed over the entire simulation.

```
73  output global interaction_frequency, interaction_name = 1,  name =
    c1,  window = discrete, size = {banner_stride}, stride =
    {banner_stride}
.
.
.
97  output global interaction_frequency, interaction_name = 25, name =
    c25, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
```

These lines provide the output for each reaction in the form of a global interaction frequency. By computing them on a stride less than the total number of timesteps we can see any transient behavior present, and by averaging over the same time step interval we ensure we average over the full simulation time. In the output the last 50 lines of collision frequency data is cut-and-pasted for further calculations, representing the second 50 ns of simulation.

RESULTS

The simulation took approximately 2 hours on 1 core of a Linux blade using version 4336 of Aleph (December 18, 2013). 95% of the computation time was spent in particle calculations.

The comparison is presented in Figure 3. Linear differences are presented in Figure 4, and a logarithmic version is included in Figure 5 (this plot is in log units to be more compatible with Figure 3).

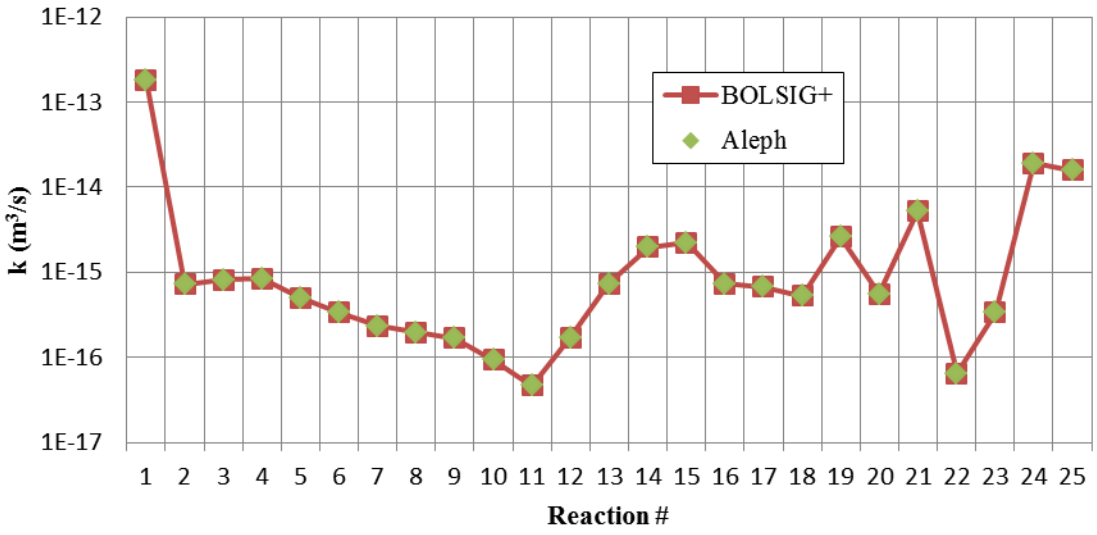


Figure 3. Comparison of BOLSIG+ and Aleph reaction rates.

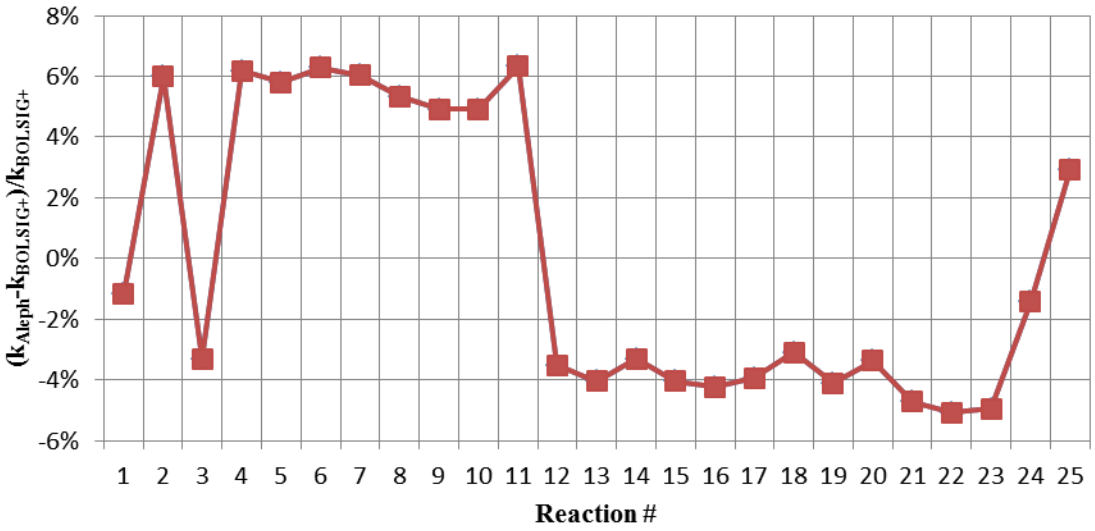


Figure 4. Difference between BOLSIG+ and Aleph reaction rates.

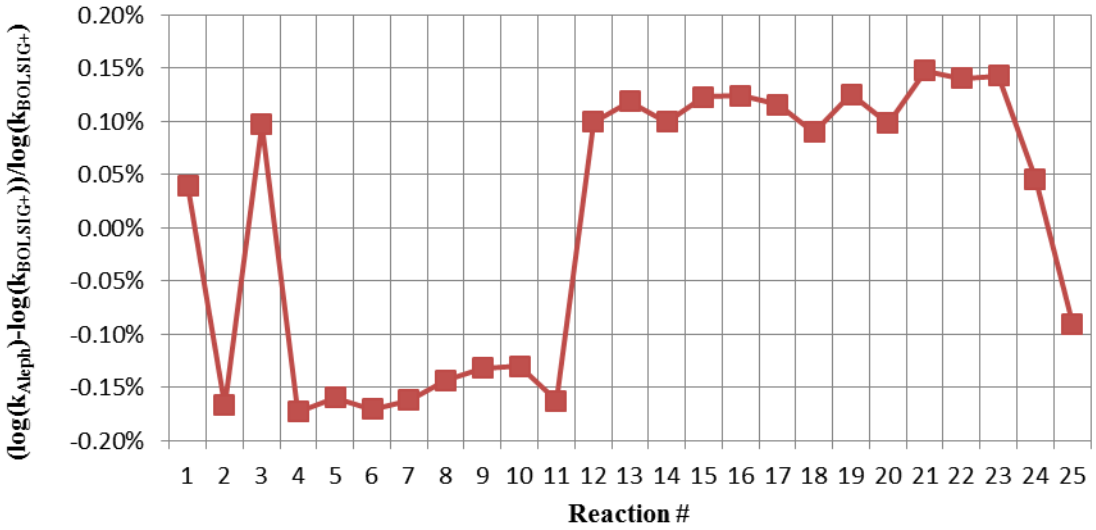


Figure 5. Difference between log(k) reaction rates for BOLSIG+ and Aleph.

FUTURE WORK

There are a number of ways this work could be extended:

1. Ensure better compatibility between BOLSIG+ assumptions and Aleph assumptions, for example:
    a. Post-collision electron energy distribution
    b. Cross-section interpolations (logarithmic vs. linear and extrapolation values off both ends of the energy range).
2. Analyze the convergence of the EEDF and/or reaction rates. This may be a substantial exercise.

3. Investigate the influence of convergence solution parameters in both Aleph (e.g., number of particles, time steps, and time step size) and BOLSIG+ (e.g., accuracy, tolerance, and grid).
4. Implement a cumulative Evaluation structure in Aleph – counting the number of collisions in a large time interval and then dividing by the time interval is better than computing frequencies every timestep and then averaging them (a sample of averages behaves better than an average of samples).

As a reminder, the goal of this work is to document the current performance of Aleph on this problem, not to make an exhaustive study (i.e., no or very few questions that arose during this comparison were explored). In practical use, the cross-section data error bars and other uncertainties will easily exceed the differences we found in this study.

ALEPH INPUT DECK

```
1   Sensitivity Level = UUR
2
3   random number generator seed = 13
4
5   particle position update = off
6
7   # UNITS
8   units = SI
9
10  # TIME
11  timestep size = 1e-13
12  total number of timesteps = 1000000
13  # banner_stride = {banner_stride = 10000}
14
15  # STRIDES
16  particle dump stride = {banner_stride}, file name =
    particles.h5part, file type = h5part
17  restart stride =               off
18  exodus output stride =         off
19  potential field solve stride = initial
20  interaction stride =             1
21  reweighting stride =           off
22  restore energy stride =        off
23  rebalance stride =             off
24
25  # INITIAL CONDITIONS
26  initial N2 density = 3.218830e24, T = 0, exact_number = true
27  initial e- density = 1.0e16, T = 0, exact_number = true
28
29  # BOUNDARY CONDITIONS
30  BC for voltage on nodelist_1 is dirichlet V = 0
31  BC for voltage on nodelist_2 is dirichlet V = 321.883
32
33  # PARTICLE WEIGHTING
34  particle weighting for N2 = 3.218830E+8
35  particle weighting for e- = 1E0
36
37  # PARTICLE TYPE DEFINITIONS
38  define particle N2,  mass = 4.651747E-26, charge = 0, category =
    neutral, diameter = 0.0, polarizability = 0.0
39
40  # INPUT
41  input mesh file name = 0D.e
42
43  # PARTICLE INTERACTIONS
44  interact e- + N2,            interaction_name = 1,
    interaction_model = elastic_isotropic_scattering,
    cross_section_file = 1.data,
    fixed_heavy_particle_properties = true
```

```
45  interact e- + N2,            interaction_name = 2,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 2.data,  heat_of_reaction = 3.204353E-21,
    fixed_heavy_particle_properties = true
46  interact e- + N2,            interaction_name = 3,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 3.data,  heat_of_reaction = 4.646312E-20,
    fixed_heavy_particle_properties = true
47  interact e- + N2,            interaction_name = 4,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 4.data,  heat_of_reaction = 4.662334E-20,
    fixed_heavy_particle_properties = true
48  interact e- + N2,            interaction_name = 5,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 5.data,  heat_of_reaction = 9.452842E-20,
    fixed_heavy_particle_properties = true
49  interact e- + N2,            interaction_name = 6,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 6.data,  heat_of_reaction = 1.409915E-19,
    fixed_heavy_particle_properties = true
50  interact e- + N2,            interaction_name = 7,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 7.data,  heat_of_reaction = 1.874547E-19,
    fixed_heavy_particle_properties = true
51  interact e- + N2,            interaction_name = 8,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 8.data,  heat_of_reaction = 2.355200E-19,
    fixed_heavy_particle_properties = true
52  interact e- + N2,            interaction_name = 9,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 9.data,  heat_of_reaction = 2.819831E-19,
    fixed_heavy_particle_properties = true
53  interact e- + N2,            interaction_name = 10,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 10.data, heat_of_reaction = 3.300484E-19,
    fixed_heavy_particle_properties = true
54  interact e- + N2,            interaction_name = 11,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 11.data, heat_of_reaction = 3.765115E-19,
    fixed_heavy_particle_properties = true
55  interact e- + N2,            interaction_name = 12,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 12.data, heat_of_reaction = 9.885429E-19,
    fixed_heavy_particle_properties = true
56  interact e- + N2,            interaction_name = 13,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 13.data, heat_of_reaction = 1.121524E-18,
    fixed_heavy_particle_properties = true
57  interact e- + N2,            interaction_name = 14,
    interaction_model = inelastic_isotropic_scattering,
    cross_section_file = 14.data, heat_of_reaction = 1.177600E-18,
    fixed_heavy_particle_properties = true
```

```
58 interact e- + N2,            interaction_name = 15,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 15.data, heat_of_reaction = 1.179202E-18,
   fixed_heavy_particle_properties = true
59 interact e- + N2,            interaction_name = 16,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 16.data, heat_of_reaction = 1.249698E-18,
   fixed_heavy_particle_properties = true
60 interact e- + N2,            interaction_name = 17,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 17.data, heat_of_reaction = 1.307376E-18,
   fixed_heavy_particle_properties = true
61 interact e- + N2,            interaction_name = 18,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 18.data, heat_of_reaction = 1.345828E-18,
   fixed_heavy_particle_properties = true
62 interact e- + N2,            interaction_name = 19,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 19.data, heat_of_reaction = 1.369861E-18,
   fixed_heavy_particle_properties = true
63 interact e- + N2,            interaction_name = 20,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 20.data, heat_of_reaction = 1.424335E-18,
   fixed_heavy_particle_properties = true
64 interact e- + N2,            interaction_name = 21,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 21.data, heat_of_reaction = 1.767201E-18,
   fixed_heavy_particle_properties = true
65 interact e- + N2,            interaction_name = 22,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 22.data, heat_of_reaction = 1.901784E-18,
   fixed_heavy_particle_properties = true
66 interact e- + N2,            interaction_name = 23,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 23.data, heat_of_reaction = 1.962666E-18,
   fixed_heavy_particle_properties = true
67 interact e- + N2,            interaction_name = 24,
   interaction_model = inelastic_isotropic_scattering,
   cross_section_file = 24.data, heat_of_reaction = 2.082830E-18,
   fixed_heavy_particle_properties = true
68 interact e- + N2 -> e- + N2, interaction_name = 25,
   interaction_model = ionization,
   cross_section_file = 25.data, heat_of_reaction = 2.499395E-18,
   fixed_heavy_particle_properties = true
69
70 # OUTPUT
71 output global computational_count for e-, name = e-_comp_count,
   window = discrete, size = {banner_stride}, stride = {banner_stride}
72 output global computational_count for N2, name = N2_comp_count,
   window = discrete, size = {banner_stride}, stride = {banner_stride}
```

```
73 output global interaction_frequency, interaction_name = 1,  name =
   c1,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
74 output global interaction_frequency, interaction_name = 2,  name =
   c2,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
75 output global interaction_frequency, interaction_name = 3,  name =
   c3,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
76 output global interaction_frequency, interaction_name = 4,  name =
   c4,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
77 output global interaction_frequency, interaction_name = 5,  name =
   c5,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
78 output global interaction_frequency, interaction_name = 6,  name =
   c6,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
79 output global interaction_frequency, interaction_name = 7,  name =
   c7,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
80 output global interaction_frequency, interaction_name = 8,  name =
   c8,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
81 output global interaction_frequency, interaction_name = 9,  name =
   c9,  window = discrete, size = {banner_stride}, stride =
   {banner_stride}
82 output global interaction_frequency, interaction_name = 10, name =
   c10, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
83 output global interaction_frequency, interaction_name = 11, name =
   c11, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
84 output global interaction_frequency, interaction_name = 12, name =
   c12, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
85 output global interaction_frequency, interaction_name = 13, name =
   c13, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
86 output global interaction_frequency, interaction_name = 14, name =
   c14, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
87 output global interaction_frequency, interaction_name = 15, name =
   c15, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
88 output global interaction_frequency, interaction_name = 16, name =
   c16, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
89 output global interaction_frequency, interaction_name = 17, name =
   c17, window = discrete, size = {banner_stride}, stride =
   {banner_stride}
```

```
90  output global interaction_frequency, interaction_name = 18, name =
    c18, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
91  output global interaction_frequency, interaction_name = 19, name =
    c19, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
92  output global interaction_frequency, interaction_name = 20, name =
    c20, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
93  output global interaction_frequency, interaction_name = 21, name =
    c21, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
94  output global interaction_frequency, interaction_name = 22, name =
    c22, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
95  output global interaction_frequency, interaction_name = 23, name =
    c23, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
96  output global interaction_frequency, interaction_name = 24, name =
    c24, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
97  output global interaction_frequency, interaction_name = 25, name =
    c25, window = discrete, size = {banner_stride}, stride =
    {banner_stride}
98  status report name = stdout, columns = timestep, simulation_time,
    c1, c2, c3, c4, c5, c6, c7, c8, c9, c10, c11, c12, c13, c14, c15,
    c16, c17, c18, c19, c20, c21, c22, c23, c24, c25, e-_comp_count,
    N2_comp_count, stride = {banner_stride}
```

BOLSIG+ SETTINGS

These were the settings used in the BOLSIG+ simulations (in case someone wants to rerun at a future date):

1. “Effect of electron production = Not included”
2. “Energy sharing after ionization = One electron takes all”
3. “Extrapolate cross sections” = off
4. # of grid points = 100
5. “Grid type = automatic”
6. Precision = 1e-10
7. Convergence = 1e-4
8. Max # of iterations = 1000

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[2] A. V. Phelps and L. C. Pitchford, *Phys. Rev. A 31,* p. 2932, 1985.

[3] A. V. Phelps and L. C. Pitchford, "JILA Information Center Report No. 26: Anisotropic Scattering of Electrons by N2 and its Effects on Electron Transport: Tabulations of Cross Section and Results," University of Colorado, Boulder, 1985.


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