Critical assessment of reaction mechanisms using the LisbOn KInetics tool suit

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Introduction

Predictive tools for non-equilibrium low-temperature plasmas (LTPs) should properly describe the kinetics of both the electrons and the heavy-species, the former responsible for inducing plasma reactivity and the latter providing the pathways for reaction mechanisms.

Here, we focus on plasmas produced in N2-O2 gaseous mixtures, aiming to deliver a KInetic Testbed for PLASMa Environmental and Biological Applications (KIT-PLASMEBA) [1], comprising the development of the LisbOn KInetics (LoKI) simulation tool, and the critical assessment of collisional-radiative data embedded in state-of-the-art kinetic schemes (KITs) for various gases and gas mixtures.

Verification and validation procedures are mandatory to ensure the quality of the tool and the results it provides. The validation roadmap includes the critical assessment of the collisional, radiative and transport mechanisms and data describing the kinetics of a gas/plasma system. Within this analysis, we are retrieving experimental data originally used in the validation of early model-versions, to evaluate and improve the current quality of model predictions.

Code implementation

LoKI is a user-friendly, scalable and upgradable tool suit, developed under MATLAB® with an object-oriented design. It comprises two modules, a Boltzmann solver, LoKI-B [2], and a chemistry solver, LoKI-C.

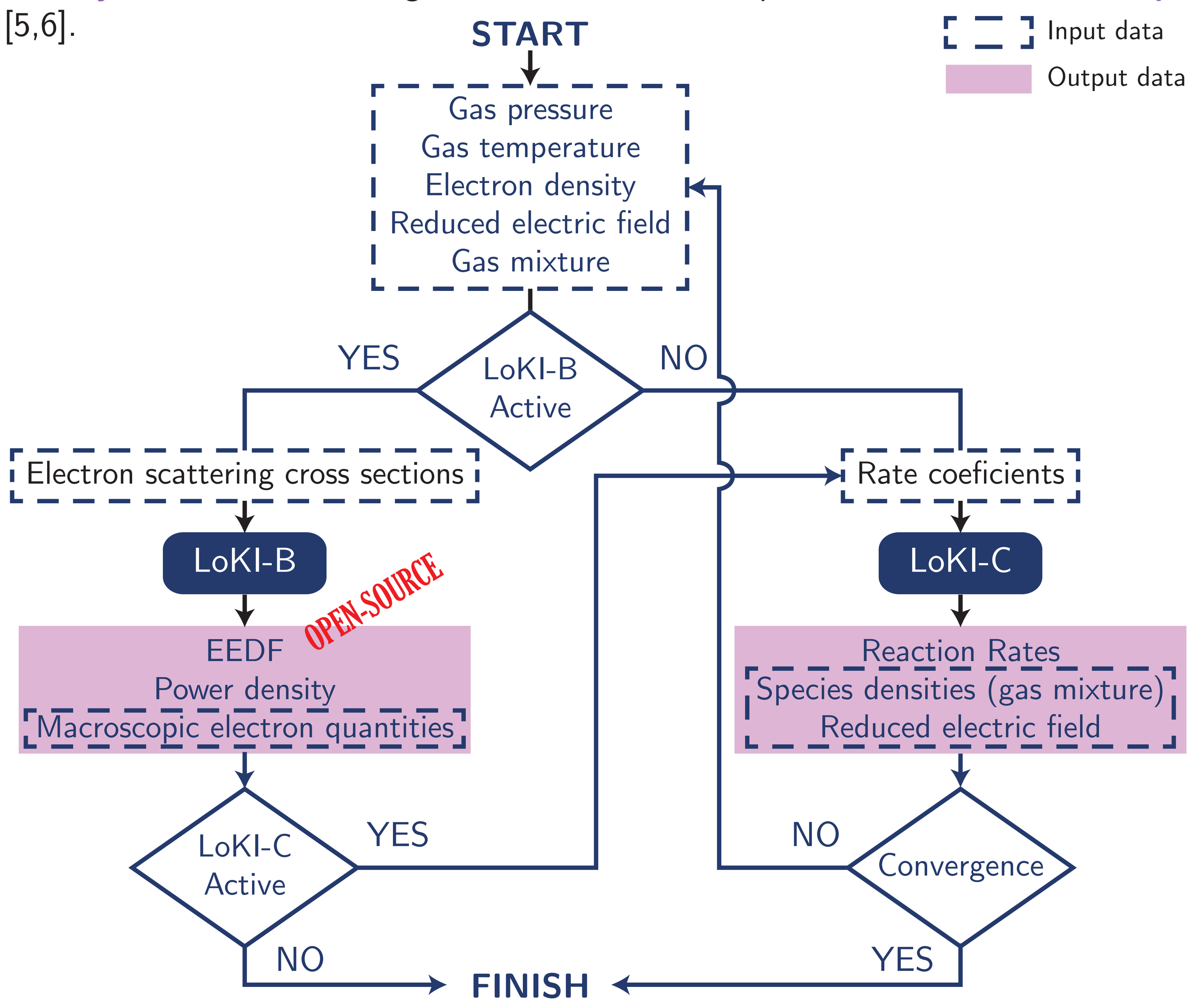
- LoKI-B (released as open-source [3]) solves a time and space independent form of the two-term electron Boltzmann equation (EBE), using electron-scattering cross sections that can be downloaded from the LXCat open-access website [4].

- LoKI-C gives the solution to the system of zero-dimensional (volume average) rate balance equations for the heavy species (charged and neutral) present in the plasma, receiving as input data the KIT(s) for the gas/plasma system under study, and using several modules to describe the mechanisms (collisional, radiative and transport) controlling the creation / destruction of species.

LoKI handles simulations in any atomic / molecular gas mixture, considering collisions with any target state (electronic, vibrational and rotational), specified in the reaction mechanism adopted. As output, the tool provides the electron energy distribution function and the corresponding electron macroscopic parameters (if LoKI-B is activated), along with the densities of species and the corresponding creation / destruction rates.

Code workflow

The following flow chart describes the workflow of LoKI. For stationary discharges, when both modules are activated, the reduced maintenance electric field (or an equivalent parameter, such as the electron temperature) is self-consistently calculated as an eigenvalue solution to the problem, under the assumption of quasi-neutrality [5,6].



Critical assessment of the Nitrogen KIT

Here, we show the results of a set of simulations for pure nitrogen DC discharges, considering the following working conditions: infinitely long tube of radius $1\ \mathrm{cm}$, gas pressures and temperatures in the ranges p~0.5-2 Torr and T_g~400-700 K, respectively, and discharge currents I~5-100 mA.

The simulations were performed with the LoKI tool suit using three different kinetic schemes:

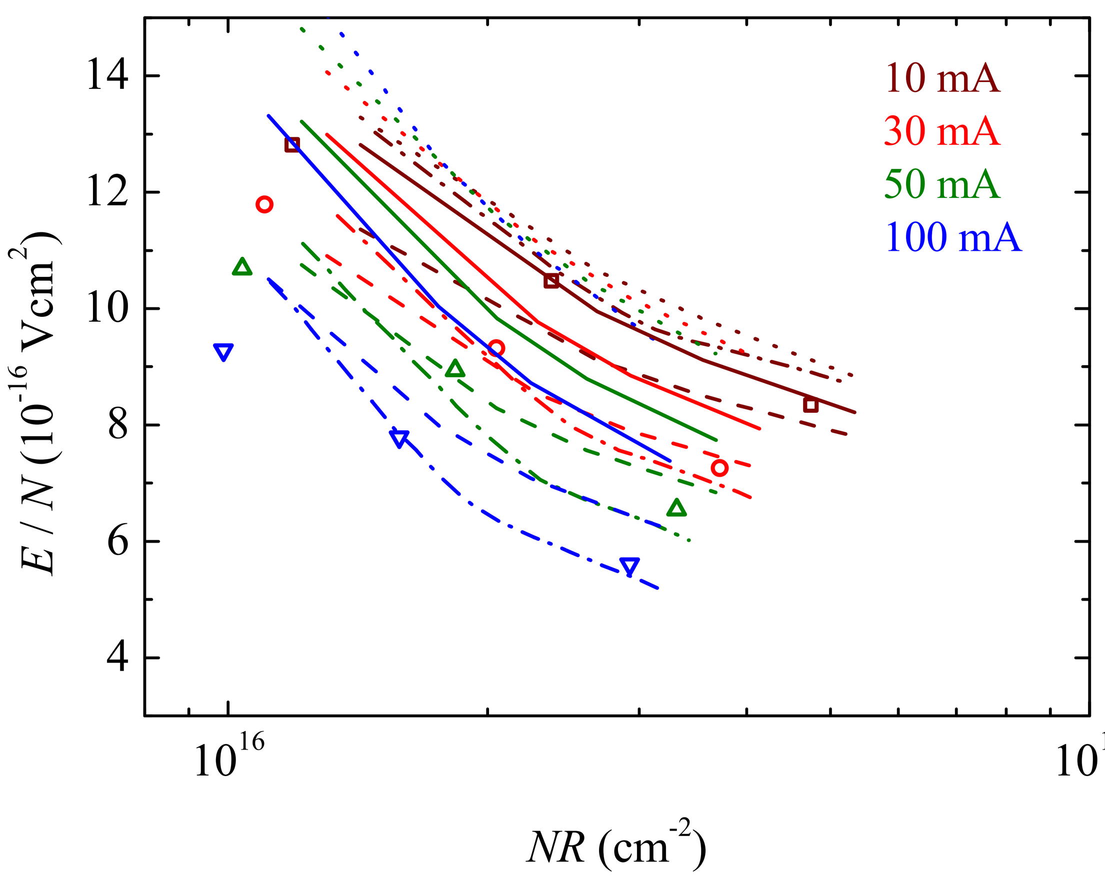
1) Most up-to-date kinetic scheme of our group [7] (solid lines)

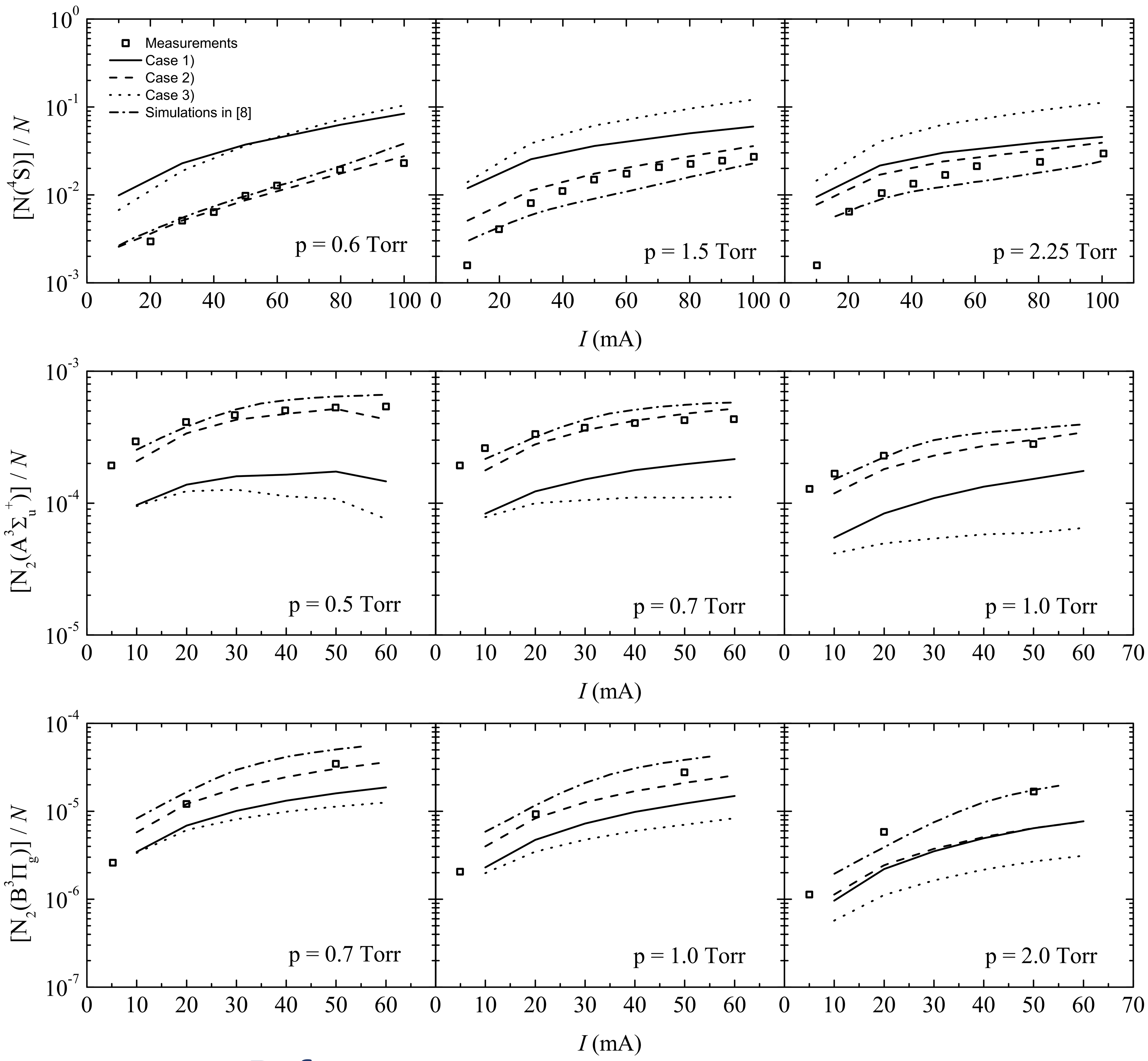
2) Previous kinetic scheme of our group [8], considering electronic excitations/ionisations from all $N_2(X,v)$ states (dashed lines)

3) Previous kinetic scheme of our group [8], considering electronic excitations/ionisations only from $N_2(X,v=0)$ (dotted lines)

The results of the simulations have been compared with experimental mesurements from [9] (symbols) and with the simulation results reported in [8] performed with a previous in-house code (dash-dotted lines).

The figure below shows, for various currents, the discharge characteristics, corresponding to the plot of the maintenance reduced electric field, E/N, versus the product of the gas density, N, and the tube radius, R. The figures on the right show, for various pressures, the relative densities of atoms, $N(^4S)$, and the molecular excited species $N_2(A^3\Sigma_u^+)$ and $N_2(B^3\Pi_g)$.





Conclusions

The LisbOn KInetics tool suit has been used to perform a critical analysis of different kinetic schemes proposed by our group for pure nitrogen comparing with experimental measurements of the discharge characteristic and the relative densities of $N(^4S)$, $N_2(A^3\Sigma_u^+)$ and $N_2(B^3\Pi_g)$, all for a DC discharge.

Even though for most of the kinetic schemes tested we obtained a reasonable agreement with the experiments, the best agreement is found with the more simple kinetic scheme reported in [8] considering excitations/ionisations from all $N_2(X,v)$ states. Indeed, after studying multiple modifications of the kinetic schemes, we have found that the biggest influence in the results is due to considering (or not) excitations/ionisations from the complete manifold $N_2(X,v)$.

These results evidence the importance of pursuing the critical analysis of mechanisms and data. Yet, the first calculations obtained with LoKI and the updated kinetic scheme of [7] already give good predictions for the parameters studied.

Our next steps in the validation process of the kinetic scheme of N_2 will be the assesment of the excitations/ionisations from the individual states of the manifold $N_2(X,v)$ since, right now, we are assuming that the rate coefficients from excited states are equal to those of the ground state $N_2(X,v=0)$.

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