Training in data calculations for environmental applications: Cross sections for excitation and ionization of NO_x molecules by electron impact for control and reduction of atmospheric pollution by NO_2/N_2O.

Scientific field: Chemical physics, quantum physics, basis on mathematical modeling and simulation, applied physics, engineering sciences
Key words: molecular dynamics, collisions, cross sections, atmospheric pollution
Collaborations: Pr. V. Kokoouline at *University of Central Florida, Orlando, USA.*
Salary: 600 €/month
Period: from September 2023 for 6 months

Ionization of the nitrogen dioxide NO_2 plays an important role in a number of plasma applications [R.K. Janev (1995) and X. Guo, et al. (2001)]. The molecule is also a common pollutant in combustion engines exhausts and contributes significantly to chemistry in the stratosphere and the troposphere. In stratosphere, NO_2 reaction cycles are important mechanisms in the formation and maintenance of ozone. In troposphere, NO_2 is formed due to the high temperatures of combustion of airplane fuel that dissociate oxygen and nitrogen molecules into their atomic constituents, which subsequently recombine. Although NO_2 is an essential contributor to the chemistry of upper atmosphere, it is an undesirable pollutant in the troposphere [J.H. Seinfeld and S.N. Pandis (1998)].

In addition, during last years, in large cities such as Paris or London, the NO_2 fraction in the NO_x exhaust of diesel cars has been continuously increasing due to the introduction of oxidative after-treatment technologies (see Fig. 1 as an example of the impact of NO_x emission on average NO_2 concentrations in Paris). As a direct effect, NO_2 ambient concentrations close to the emission source increase.

Many previous works have been done to elucidate nitrogen-dioxide molecule properties and their interaction with photons and electrons. However, only a few works on the electron ionization of $_{NO2}$ have been reported so far. Hence accurate ionization cross sections for NO_2 are important for monitoring of this species in the atmosphere and plasma modeling. The experimental determination of ionization cross sections includes those of Rapp and Englander-Golden (1965) and Lopez et al. (2003). As for the theoretical point of view, calculations for open shell molecule such as NO_2 are particularly challenging. The rigorous quantum mechanical approach for the calculations for molecules is limited to the application of simple molecules. Contrary to it, there now exists binary encounter Bethe formalism by Kim et al. (1997), semi-empirical formalism by Khare et al. (1989), and DM-formalism by Probst et al. (2001).

Research subject, work plan:

The aim of this project is to contribute to enrich the database of the IAEA by cross sections for excitation of N_2O and NO_2 molecules by electron impact. The cross sections will be evaluated using molecular dynamics approach. In particular, the UK R-matrix code [J. Tennyson *et al*. (2007)] will be employed. The obtained cross sections of collision processes will be introduced into databases used for modeling the dynamics of destruction of NO_x particles. ***Collaboration with the University of Central Florida with a possibility of stay is planned for this project***.

In this project the work will consist of two main tasks:

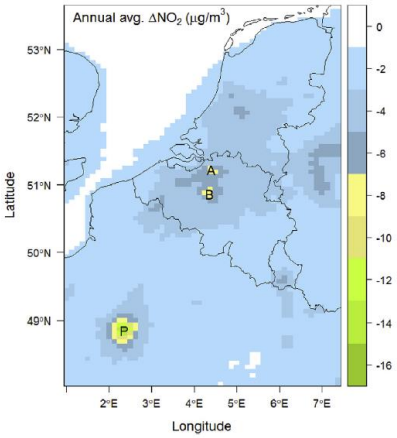

Fig. 1: Impact of NO_x *emission levels on annual* NO_2 *concentrations in the Benelux zoom area. City locations are A: Anwerp, B: Brussels and P: Paris. (Bart Degraeuwe, et al. 2016).*

Task 1 – Using Quantemol software and the implemented molecular dynamics approach, based on the UK R-matrix formalism, we will determine vibrational and electronic excitation and ionization cross sections of N_2O and NO_2 molecules. The UK R-matrix method [J. Tennyson (1996)] is a suite of electron-scattering codes based on first principles used for calculating low-energy electron-molecule scattering cross sections.

Task 2- The above-mentioned calculations will be compared and validated with experiments performed in laboratory. These data will be also included in the atomic and molecular databases for plasma modelling such as ADAS (http://open.adas.ac.uk/) and KIDA (http://kida.obs.u-bordeaux1.fr/).

References:

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