Development of Geant4-DNA for atmosphere simulations

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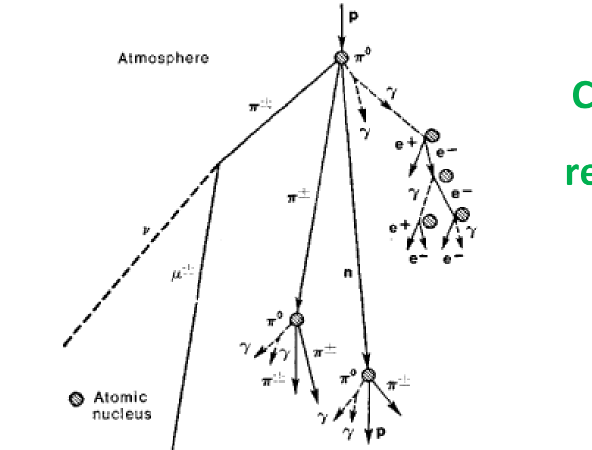
20 June 2022

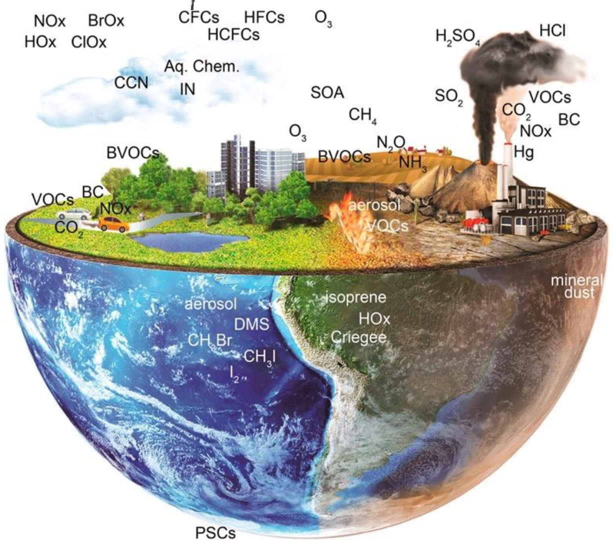
Motivation

<u>What is the impact of cosmic rays and ions on atmospheric chemistry</u>?



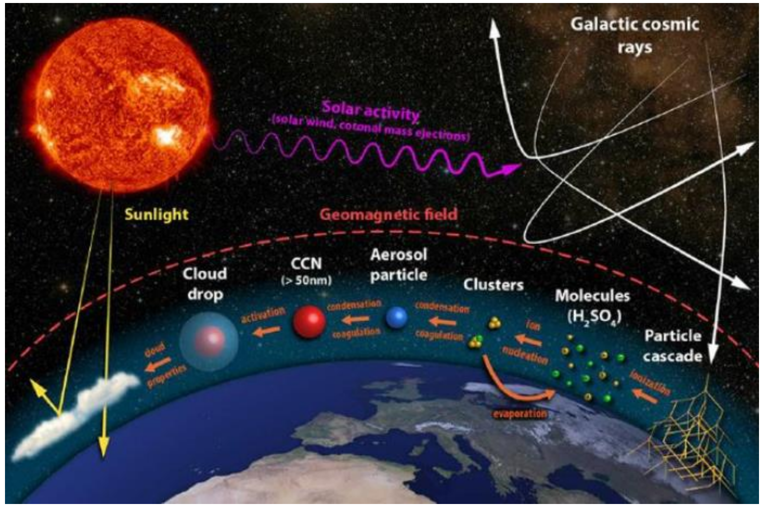
CRs represent the main source of atmospheric ionization and related physical-chemical changes in the low-mid atmosphere

(Bazilevskaya et al. 2008).



precipitation
(Kniveton 2004)

aerosol formation
(Shumilov et al. 1996 Mironova and Pudovkin 2005; Kazil et al. 2006)



ion-induced nucleation
(Svensmark et al. 2007)

cloud cover
(Voiculescu et al. 2006)

Ozone deplation
(Enghoff et al2012)

Motivation

What is the impact of cosmic rays and ions on atmospheric chemistry?

Chemical networks

Incomplete in the ions description

Physical Machinary

How many ions are produced?
In what state?
What spatial distribution?

Climatological models

The predictivity of current models is still incomplete..

The proposed project aim at a very accurate characterization of ions state and distribution in the atmosphere.

Ions in the atmosphere

- Ions affect the atmospheric composition either destroying or producing neutral molecules by faster reactions
  - **ion-molecule reaction rate up to 10 orders higher**

- Even a small amount of substance can make a difference:
  - **non-linear chemical system**
  - catalytic processes

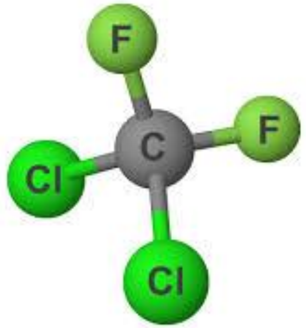
- Spatial distribution of ions is **extremely inhomogeneous**

Production of dangerous greenhouse gases

$$O_3^+ + N_2 \longrightarrow O_2^+ + N_2O$$

F. Cacace et al. ***Angew. Chem. Int. Ed. Engl.*** ***2001***, *40, 1938*

$CFCl_3$



The connection of cosmic rays with ions and the climate parameters is a challenging topic.

CR induced Ionisation models: State of the Art

Monte-Carlo simulations:

Oulu CRAC:CRII (CORSIKA+FLUKA)

Usoskin et al., J. Atm. Solar-Terr. Phys, (2004).
Usoskin, Kovaltsov, J. Geophys. Res., (2006, 2010).

Bern model ATMOCOSMIC (GEANT-4)

Desorgher et al., Int. J. Mod. Phys. A, (2005)
Scherer et al. Space Sci. Rev. (2006).

AtRIS (GEANT-4)

Banjac et al., JGR Space Physics (2018)

RUSCOSMICS

Maurchev et al., Bull. Russ. Acad. Sci. Phys. (2019)

Physics behind: Monte-Carlo simulation of the cascade, all species and processes included down to lower stratosphere (below ~20 km),

Output of the models: average production rate of ion pairs (ions cm-3 s-1)

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Physics behind: Monte-Carlo simulation of the cascade, all species and processes included down to lower stratosphere (below ~20 km),

Output of the models: average production rate of ion pairs (ions cm-3 s-1)

BUT…

- **The interaction of low-energy secondary radiation** with molecules **are not included;**
- **They can not provide the exact concentration of the ions produced, their spatial distribution, and ionization state.**

!

Geant4-DNA for atmosphere

The AIM:

Accurately describe the **amount and state of ionisation, and the spatial distribution of ions produced by CR** interaction in the atmosphere;

The HOW:

By **including in Geant4-DNA models** for particle-impact interactions with relevant molecules for climatology

Now: e- impact on N2 and O2

- **Ionisation:** Relativistic Binary Encounter Bethe model;
- **Elastic:** ELSEPA code;
- **Electronic excitation**: WORK IN PROGRESS.

Next: $\boldsymbol{CO_2, N_2O, O_3, CH_4, ..}$

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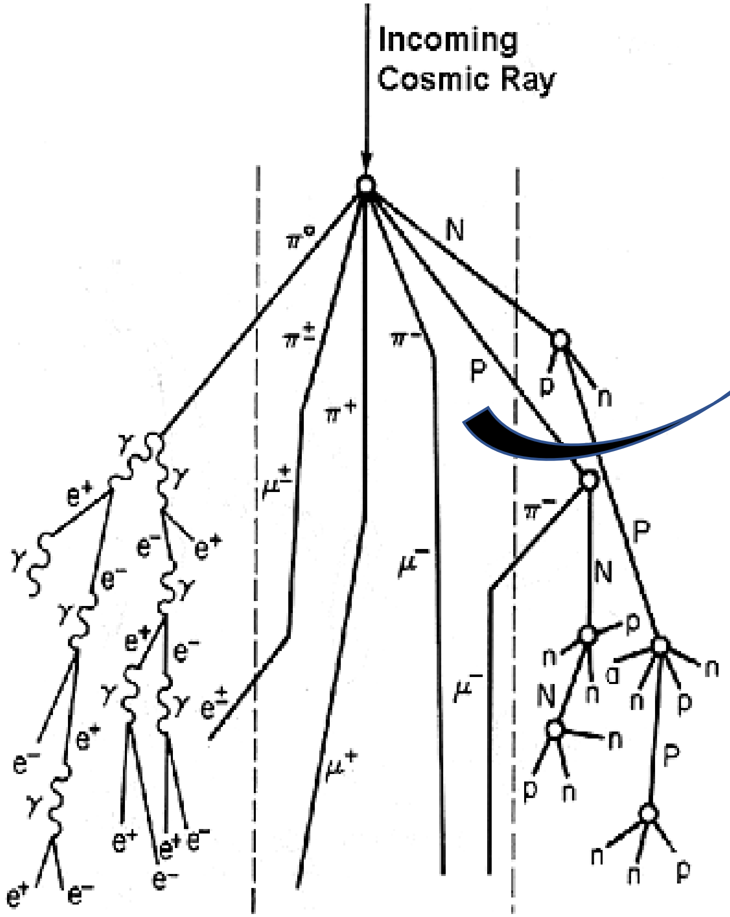
External Collaborators:

- **A. Cartoni, M. Satta** (Sapienza Chemistry, CNR, Rome)
- **Dott.ssa B. Caccia** (National Health Instituite, Rome)
- **S.Incerti, H.N. Tran** (CNRS / IN2P3, France)
- **D. Emfietzoglou, I. Kyriakou** (Ioannina, Greece)

Geant4-DNA for atmosphere :final goal



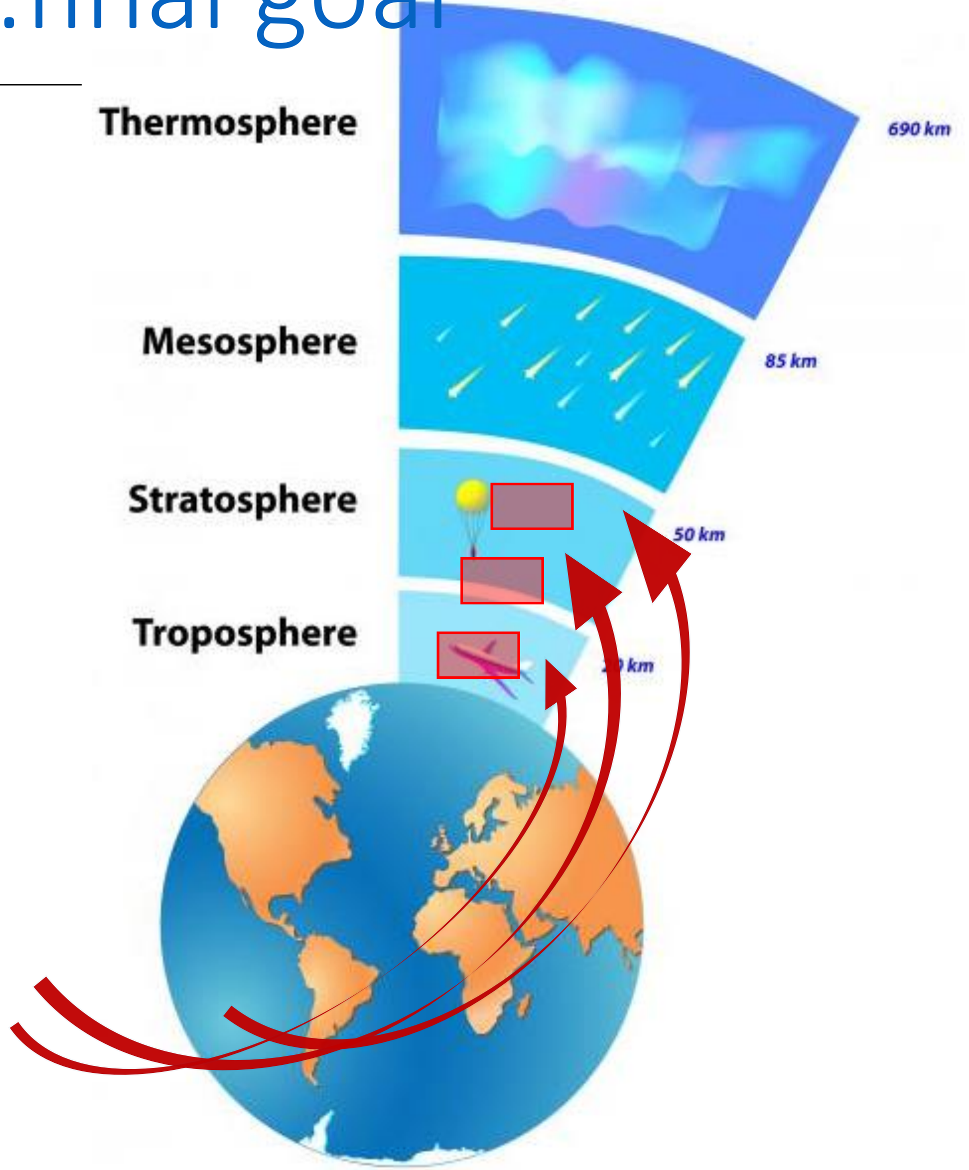
Cosmic ray spectra
Cascade: Monte-Carlo simulation
(ATMOCOSMICS, RUCOSMICS..)

- Geant4 models:

  p,γ,μ,k interactions with matter

- Our “DNA” models:

  Accurate e- interaction description in small volumes at different altitudes in the troposphere and lower Stratosphere.



Ionisation - RBEB

- Electron impact ionization
- **Method**: Relativistic Binary Encounter Bethe (RBEB)
  (Kim, 2000);
- **Energy range**: threshold – 1 GeV.
- **Advantages**:
  - depends only on B, U, N for each MO;
  - allows energy loss to be randomly sampled without using tables;
  - Applicable to different targets (H2O, DNA, gases..)

SDCS for Molecular Orbital:

$$\frac{d\sigma_{ion,MO}}{dW} = \frac{\pi a_0^2 \alpha^4 N}{(\beta_t^2 + \beta_u^2 + \beta_b^2)2b'} \Big\{ [ln(\frac{\beta_t^2}{1-\beta_t^2}) - \beta_t^2 - ln(2b')][\frac{1}{(w+1)^3} + \frac{1}{(t-w)^3}] - \frac{1}{t+1}(\frac{1}{w+1} + \frac{1}{t-w})[\frac{1+2t'}{(1+t'/2)^2}] + \frac{1}{(w+1)^2} + \frac{1}{(t-w)^2} + \frac{b'^2}{(1+t'/2)^2} \Big\}$$

B: Binding Energy
U: Average Kinetic Energy
N: Electron Occupation Number

W: ejected e^- energy;
T: incident e^- energy;
U=u/B, w=W/B, t=T/B;
$S = (4\pi a_0^2 N R^2)/B^2$

GEANT4-DNA ELECTRON Ionisation (alternative models to default one)

Material	Corresponding model	Class name	Energy range	Type
H_2O	G4DNACPA100IonisationModel	BEB*	11 eV - 255 keV	interpolated
Gold	G4DNARelativisticIonisationModel	MRBEBV**	8.3 eV - 1 GeV	analytical

* Binary Encounter Bethe
** Modified Relativistic Binary Encounter Bethe Vriens
(for alkali metals - low binding energy regime)

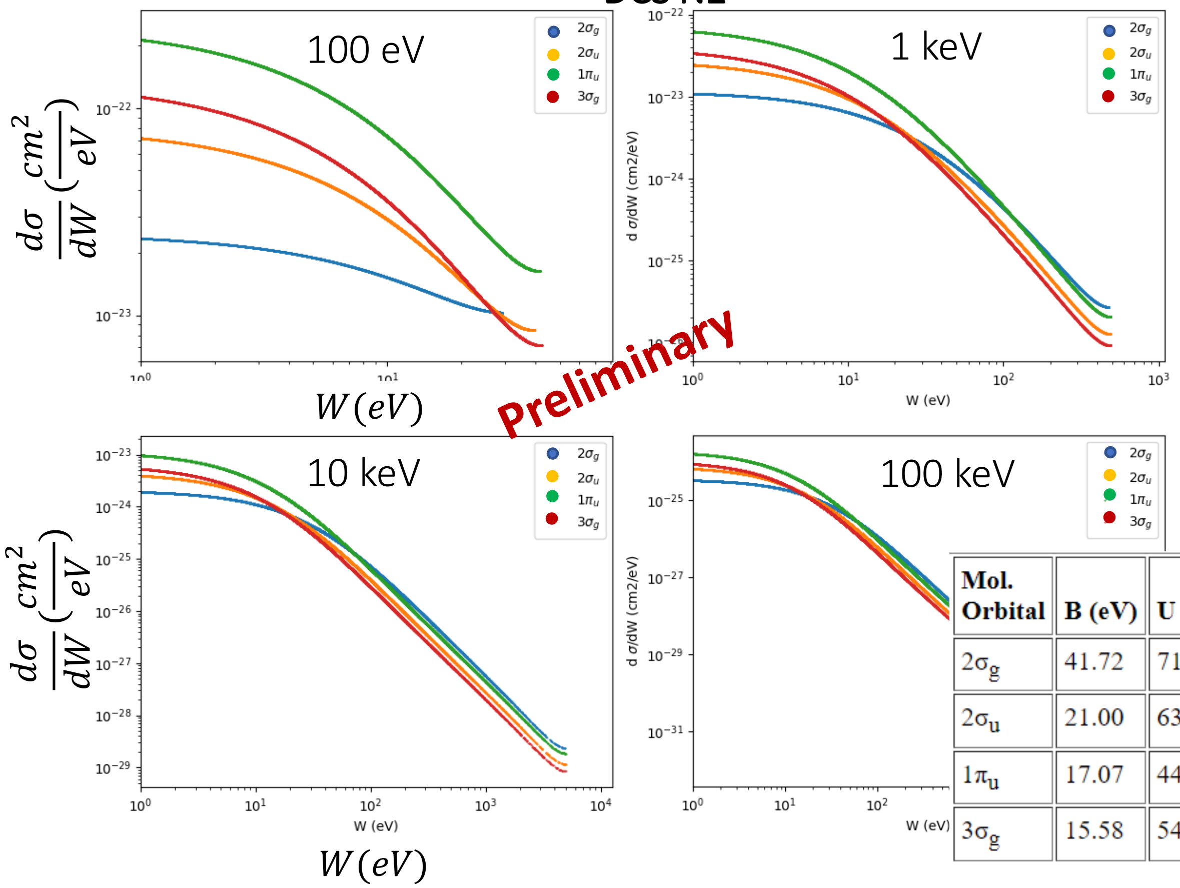
RBEB - implementation in GEant4

e^- impact ionization of N2, O2:

- **Energy range:** threshold - 1GeV
- **Type** : analytical

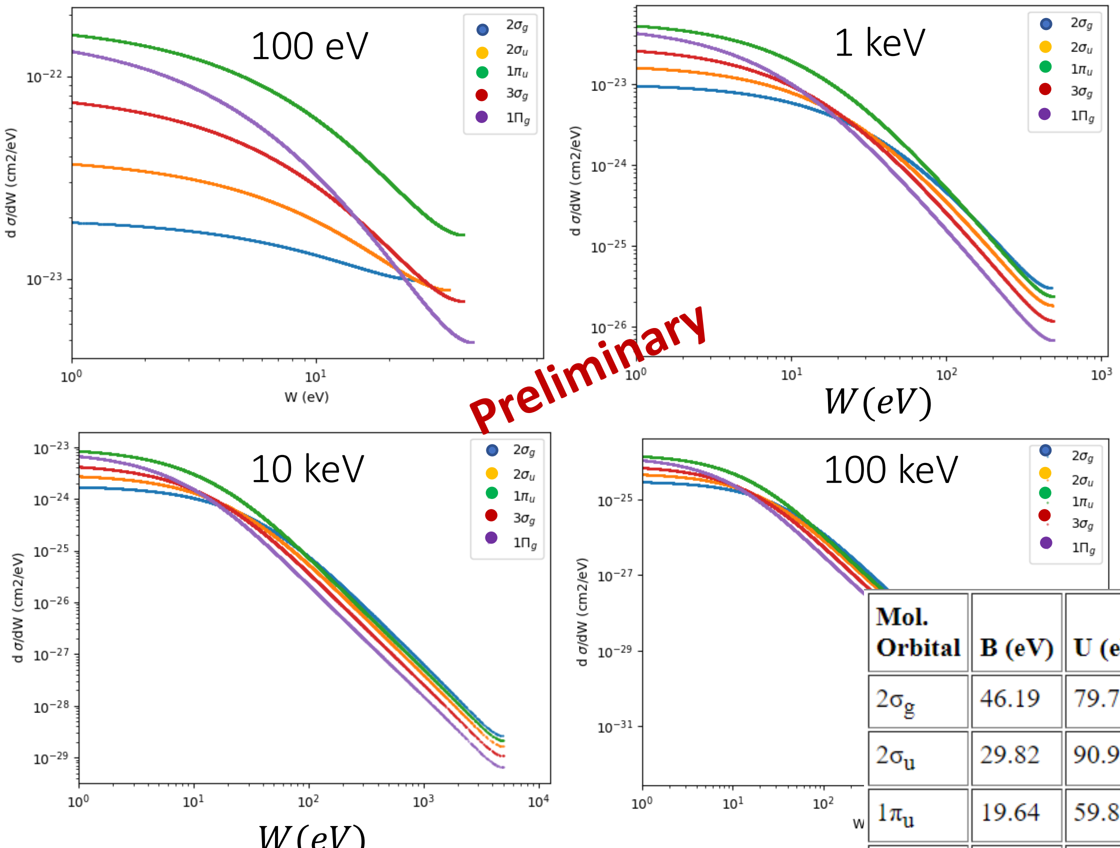
B: Binding Energy
U: Average Kinetic Energy
N: Electron Occupation Number

DCS N2



Mol. Orbital	B (eV)	U (eV)	N
$2\sigma_g$	41.72	71.13	2
$2\sigma_u$	21.00	63.18	2
$1\pi_u$	17.07	44.30	4
$3\sigma_g$	15.58	54.91	2

DCS O2



Mol. Orbital	B (eV)	U (eV)	N
$2\sigma_g$	46.19	79.73	2
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$1\pi_g$	12.07	84.88	2

* B, U values from Hwang et al. (1996)

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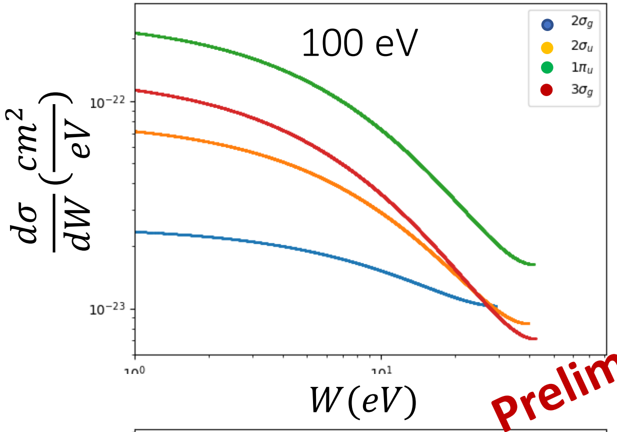
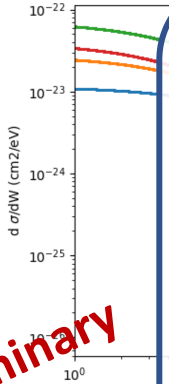
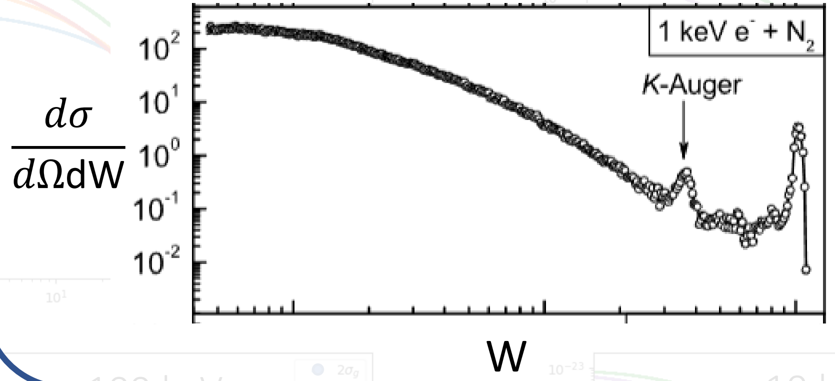




Preliminary

- **N and O inner k-shells not yet included**



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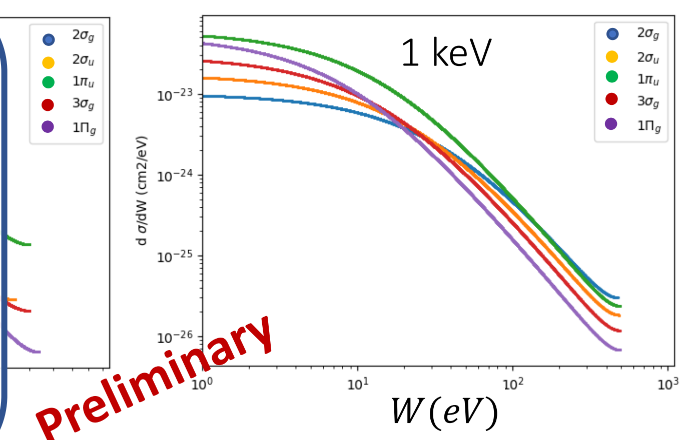

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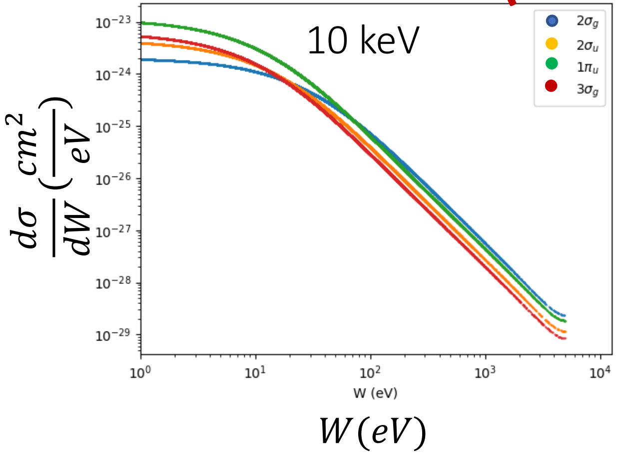

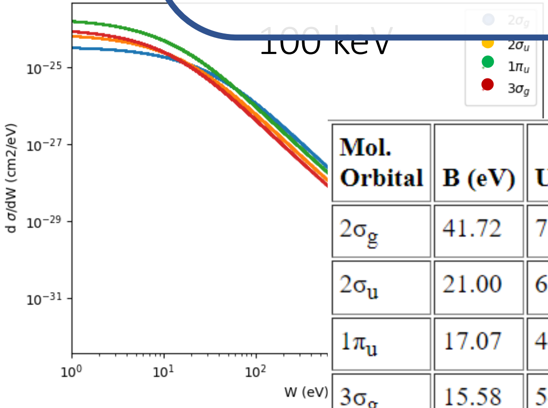

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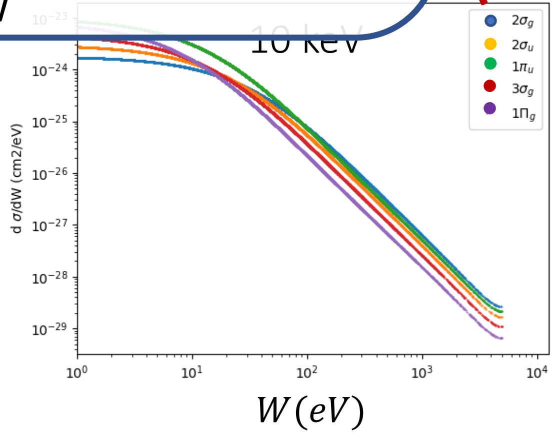

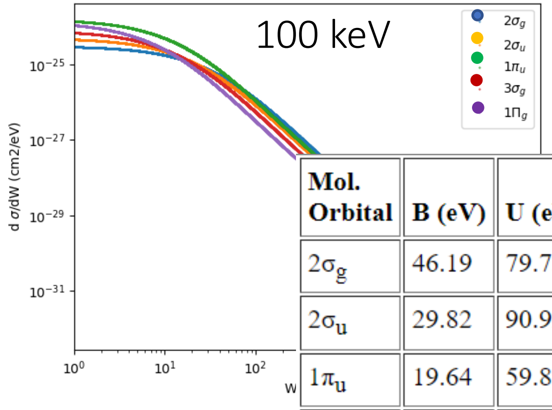

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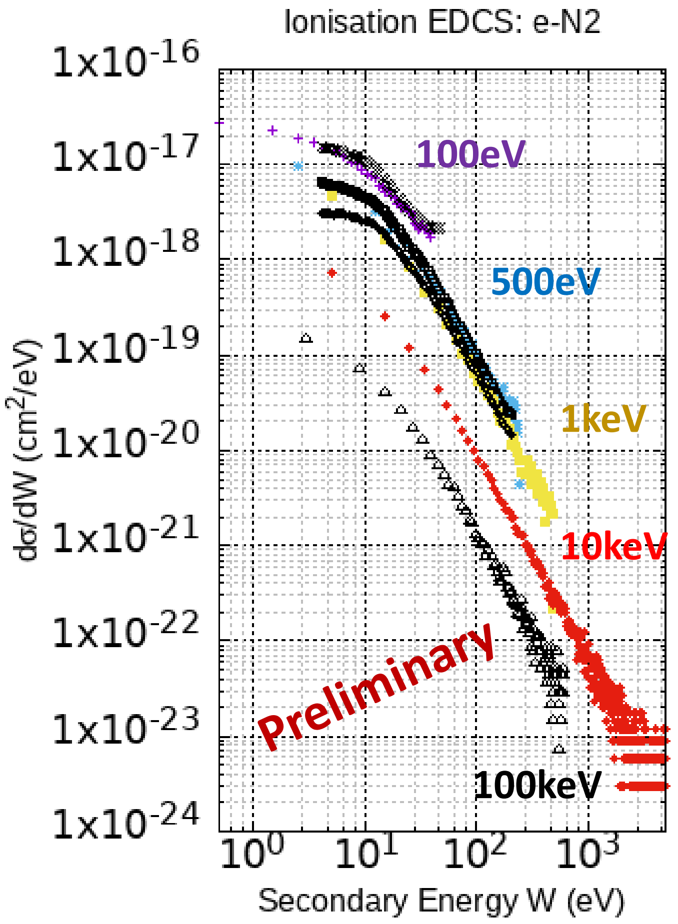
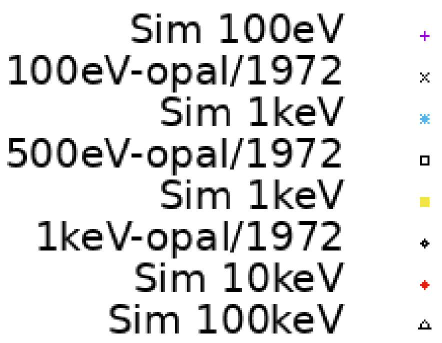
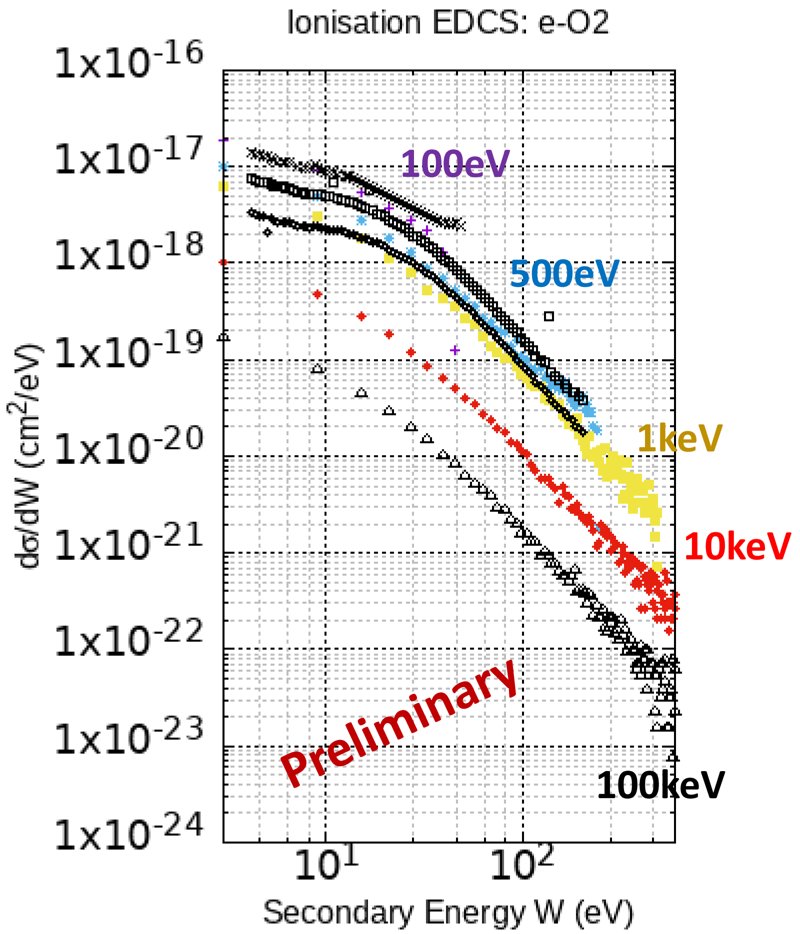






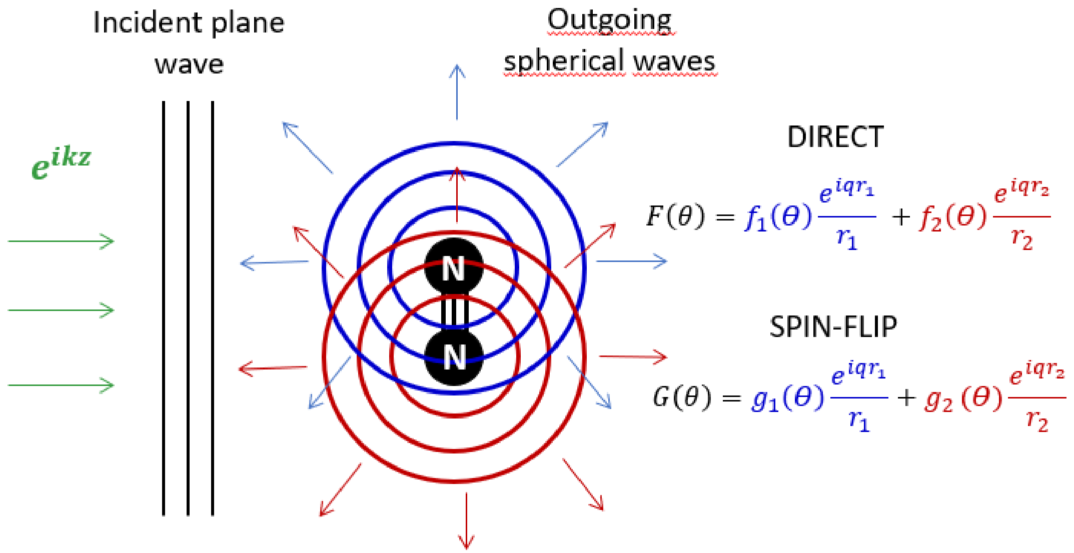
Other DCS data sets:

- C.B. Opal, E.C. Beaty, W.K. Peterson, Atomic Data Tables 299.(1972) 209.
- R.D. DuBois, M.E. Rudd, Phys. Rev. A 17 (1978) 843.
- T.W. Shyn, Phys. Rev. A 27 (1983) 2388.

Elastic scattering – ELSEPA code

"ELastic Scattering of Electrons and Positrons by neutral Atoms" code developed by Salvat et al.
Freely available at *https://github.com/eScatter/elsepa*

- **Method**: Indipendent Atom Model (Mott and Massey 1965)
  - Relativistic partial wave analysis
  - Molecular **SDCS** as a coherent sum of atomic scattering amplitudes
- **Energy range:** tens of eV – 1 GeV
- **Advantages**:
  - Easy to change calculation parameters and interaction potential models;
  - Allows to calculate DCS in a variety of materials;



$$\left.\frac{d\sigma}{d\Omega}\right|_{N2} = \langle\, |F(\theta)|^2 + |G(\theta)|^2 \,\rangle_{or}$$

GEANT4-DNA ELECTRON Elastic (alternative model to default one)

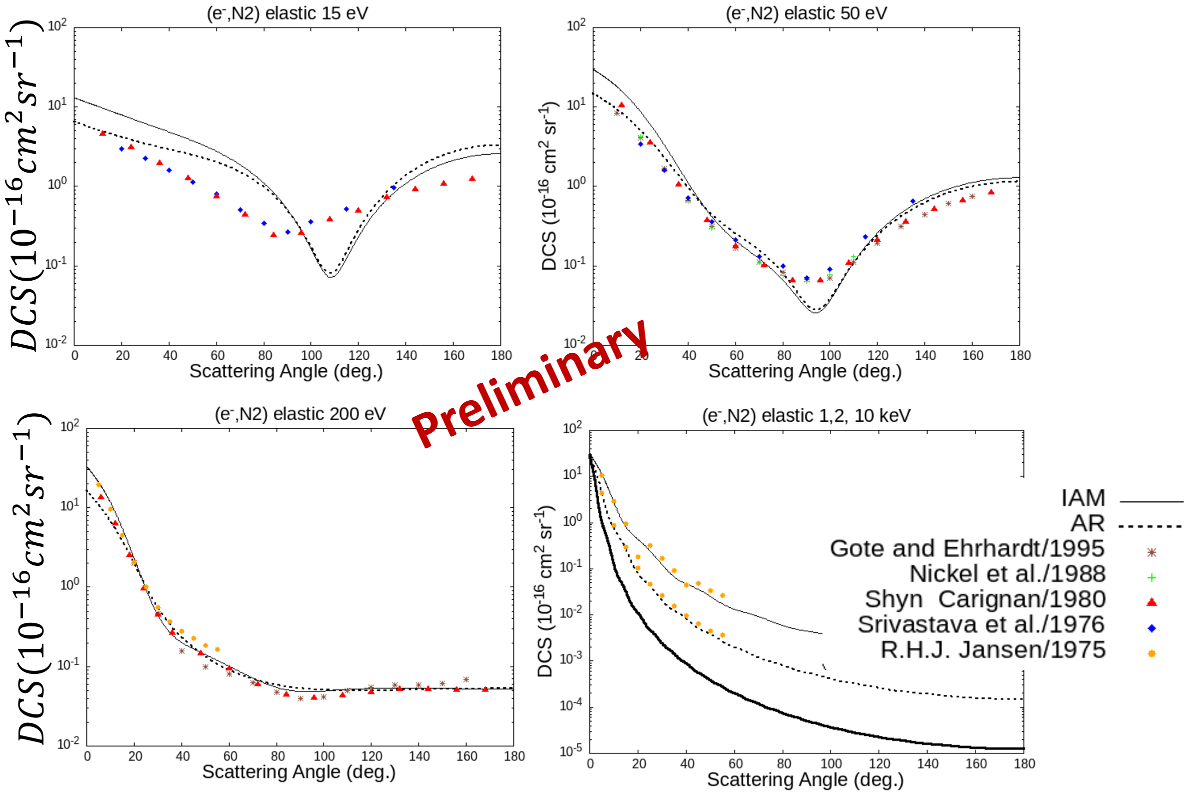
Material	Corresponding model	Class name	Energy range	Type
Gold	Relativistic PW (ELSEPA)	G4DNAELSEPAElasticModel	10 eV - 1 GeV	interpolated

ELSEPA – implementation in Geant4

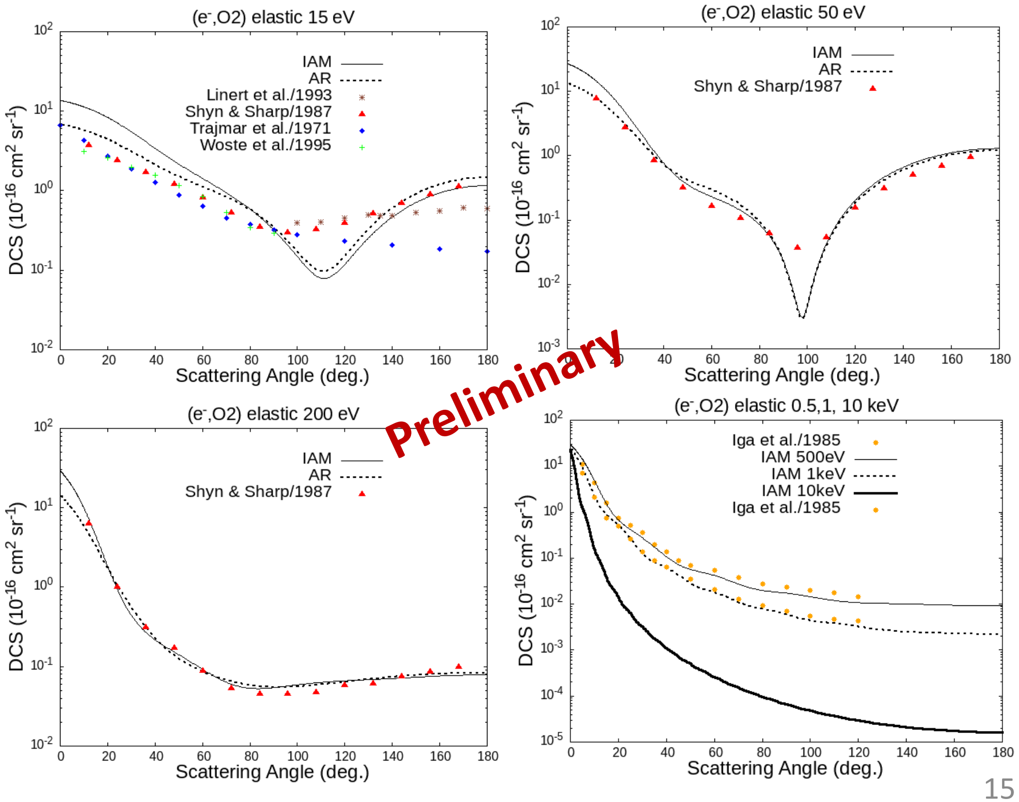
e^- elastic scattering on N2, O2:

- **Energy range:**
  - 100 eV - 1GeV : ELSEPA data
  - < 100eV : weighted average of exp data (work in progress)
- **Type** : interpolated

DCS N2

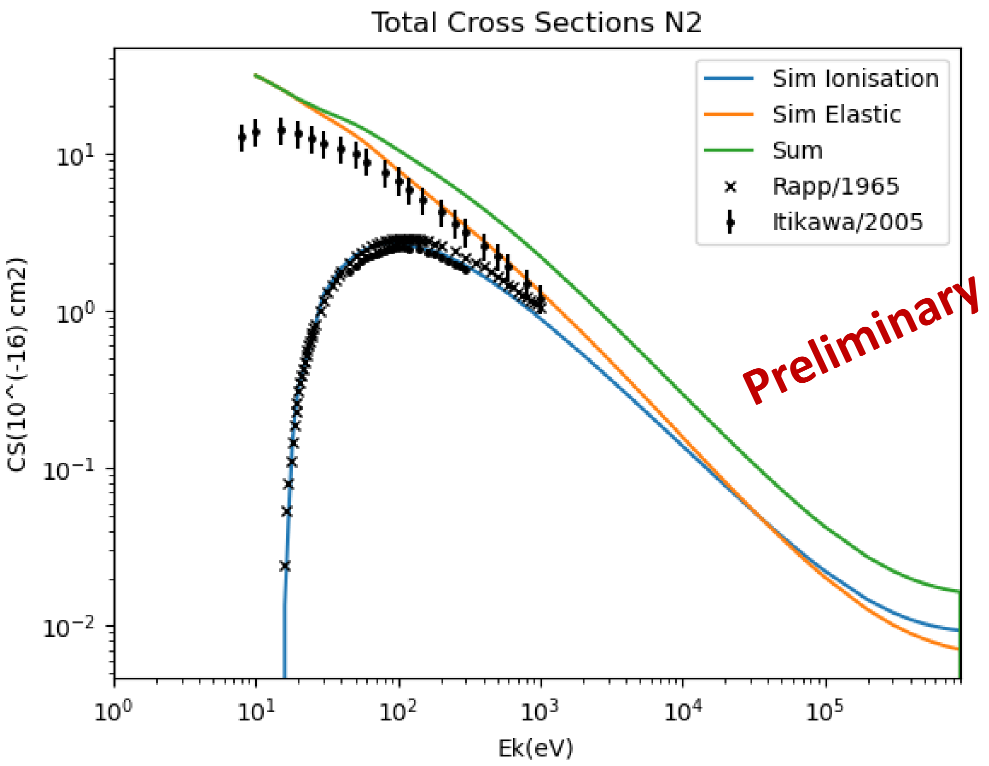


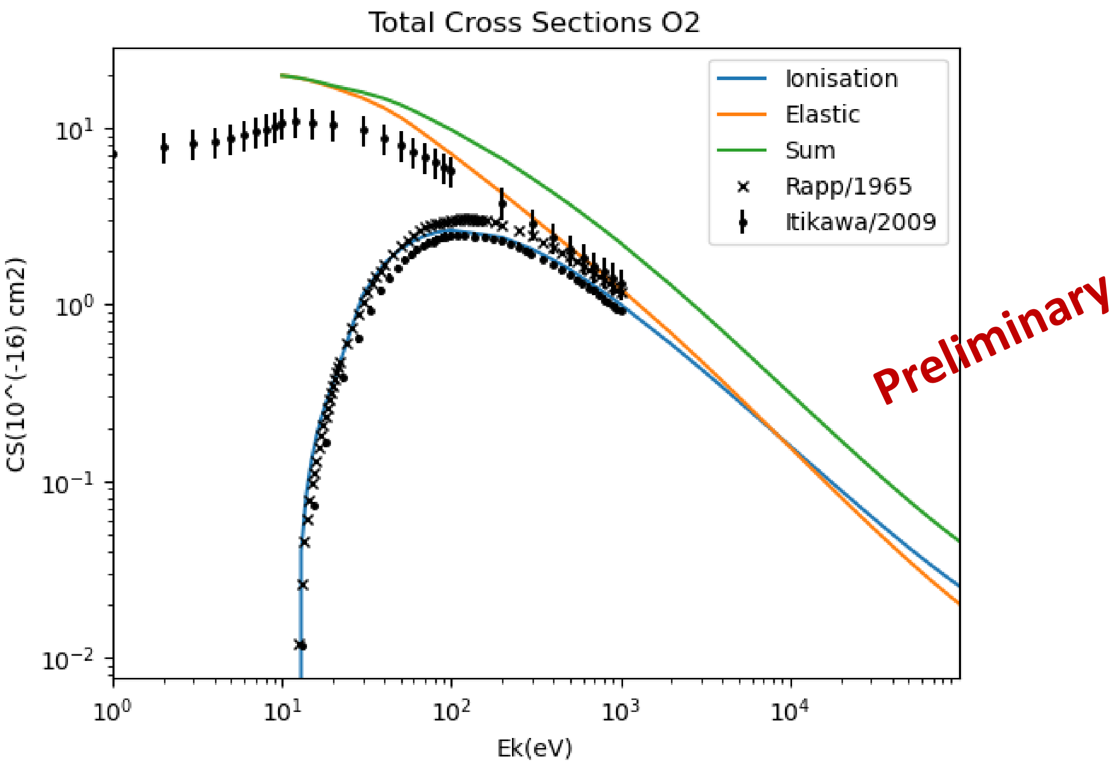
DCS O2



Ionisation/Elastic – Total CS

CS N2,O2:

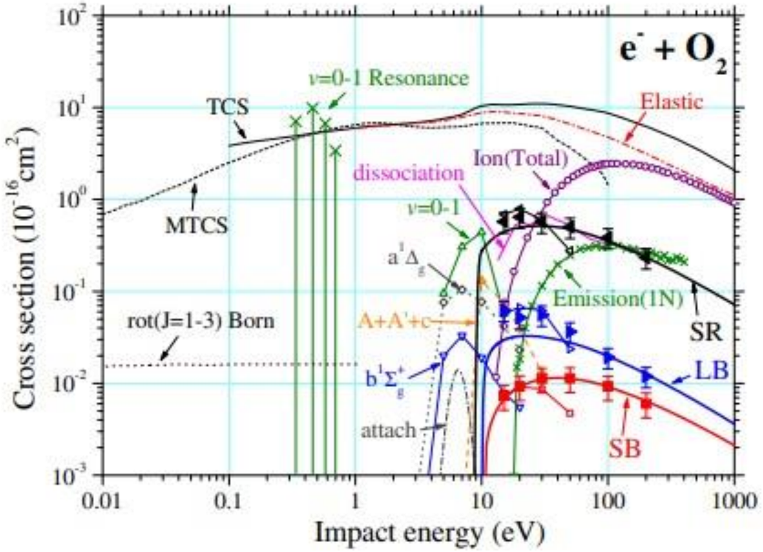




N_2 ionization threshold: 15.58 eV

O_2 ionization threshold: 12.07 eV

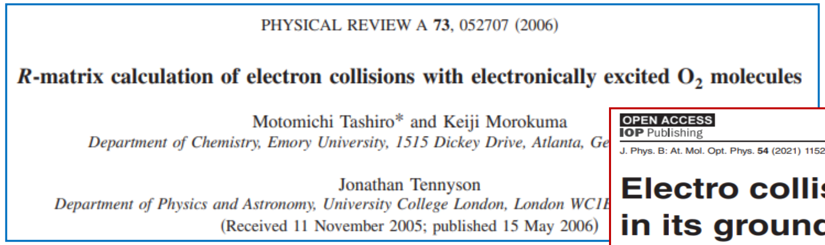
Excitation CS



- **BEf-scaled Born method:**
  - For electric dipole-allowed transitions
  - It depends by exp. Or theo. Quantities

- Ab-initio **R-matrix** close-coupling **method**
  - Accurate for TCS in the low energy range (5eV – 100 eV);
  - UKRmol+ code for the calculations.

- Experimental data


PHYSICAL REVIEW A 73, 052707 (2006)

***R*-matrix calculation of electron collisions with electronically excited O_2 molecules**

Motomichi Tashiro* and Keiji Morokuma
Department of Chemistry, Emory University, 1515 Dickey Drive, Atlanta, Ge
Jonathan Tennyson
Department of Physics and Astronomy, University College London, London WC1E
(Received 11 November 2005; published 15 May 2006)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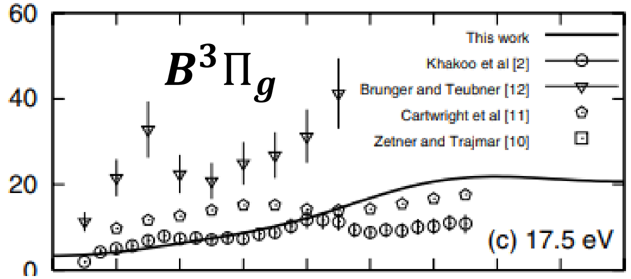
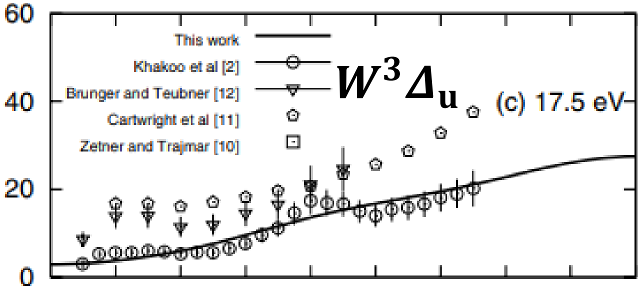
https://doi.org/10.1088/1361-6455/abf9f0

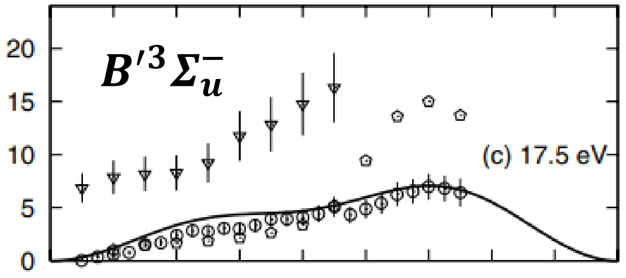
Electro collisions with molecular nitrogen in its ground and electronically excited states using the R-matrix method

He Su[1], Xinlu Cheng[2], Hong Zhang[1,*] and Jonathan Tennyson[3,*]

Tashiro and Murukuma (2007):







Summary

- ✓ The ionisation state and the spatial distribution of ions produced by cosmic rays can significantly change chemical reaction rates by orders of magnitude;

- ✓ Our project is trying to better characterizing ions in the atmosphere by extending Geant4DNA with new interaction models with molecules in the atmosphere;

- ✓ We are starting with N_2 and O_2 but our goal is to find models for all molecules of climatological interest ($CO_2, N_2O, O_3, CH_4, ..$)

Thank you for your attention!

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Backup

ELSEPA interaction potential

Optical potential model:

$$V(r) = V_{st}(r) + V_{ex}(r) + \boldsymbol{V_{cp}(r)} - iW_{abs}(r)$$

Electrostatic potential $V_{st}(\mathrm{r})$

- **Potential model:**

  Nuclear charge: Fermi distribution;

  Electron density: Dirac–Fock distribution.

Exchange potential $V_{ex}(r)$

Potential model:

Furness–McCarthy potential.

Correlation-polarization potential $\mathbf{V_{st}(r)}$

Influence at small scattering angles and E < 500eV

- **Potential model:**
  - Buckingham potential + LDA correlation (Perdew and Zunger)
- **Free parameters:**
  - static polarizability α_d = *1.562E-24(O2), 1.710E-24(N2) [cm^3]*
  - cut-off parameter $b_{pol}^2 = \max[(E - 20\ \mathrm{eV})/\mathrm{eV},\ 1].$

Inelastic absorption potential $-\mathbf{iW}_{abs}(\mathrm{r})$:

Influence at intermediate and large scattering angles

- **Potential model:**
  - LDA potential (Salvat)
- **Free parameters**:
  - lowest excitation energy $\in_1 =$ 0.98 eV(O2), 7.63 (N2)
  - absorption strength $A_{abs} = 2$

<u>N.B</u>. Empirical parameters are **validated for noble gases** and mercury

Work in progress and next steps

- Ionisation: Inclusion of inner N,O k-shells and deexcitation;

- Elastic scattering:
  - implementation of weighted average of exp data for E< 100eV;
  - Systematic study to find the best empirical parameters of the optical interaction potential , b_{pol}, A_{abs};

- Electronic excitation: ??