Development of Geant4-DNA for atmosphere simulations

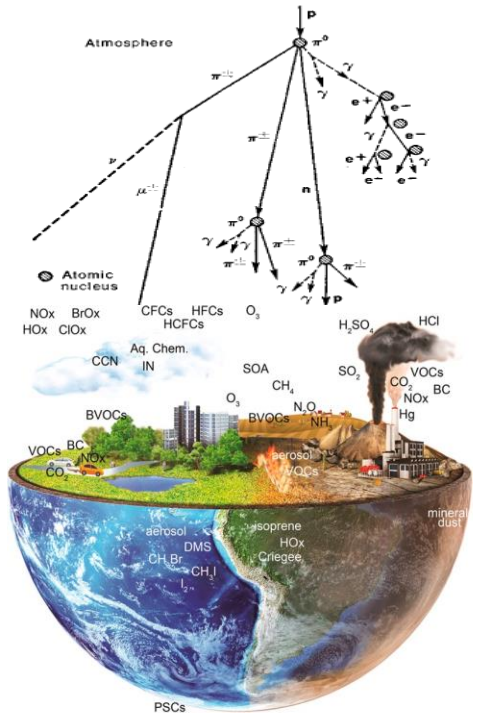
F. Nicolanti, B. Caccia, A. Cartoni, D.Emfietzoglou, R. Faccini, S. Incerti, I. Kyriakou, M. Satta, H. N. Tran, C. Mancini-Terracciano

Istituto Nazionale di Fisica Nucleare

Contact: francesca.nicolanti@uniroma1.it

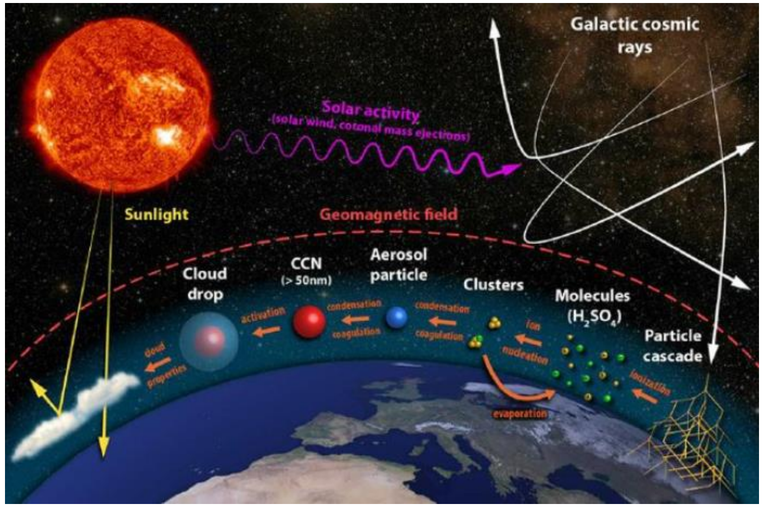
Ions in the atmosphere

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



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
(Kniveton 2004)

- The predictivity of current climatological models is still incomplete..
- Many questions open: How many ions are produced? In what state? What spatial distribution?

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**

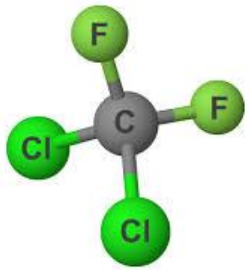
- Spatial distribution of ions is **extremely inhomogeneous**

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.

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CR induced Ionisation models

Monte-Carlo codes:

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)

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:

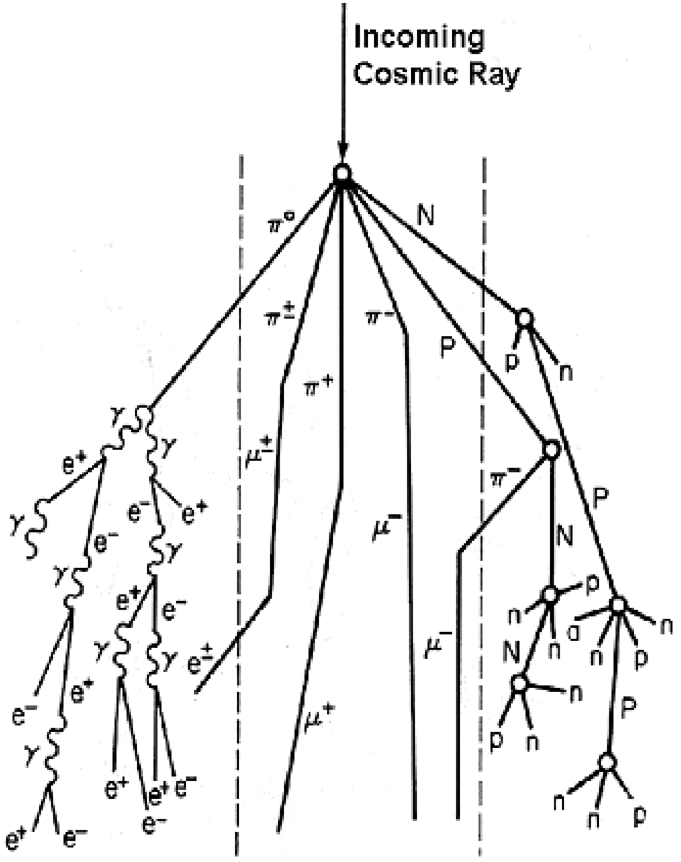
By **including in Geant4-DNA new models** for particle-impact interactions with relevant molecules for climatology

Under study:

- e- impact on N2 and O2

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

Geant4-DNA for atmosphere



Geant4 codes (ATMOCOSMICS /RUSCOSMICS..):

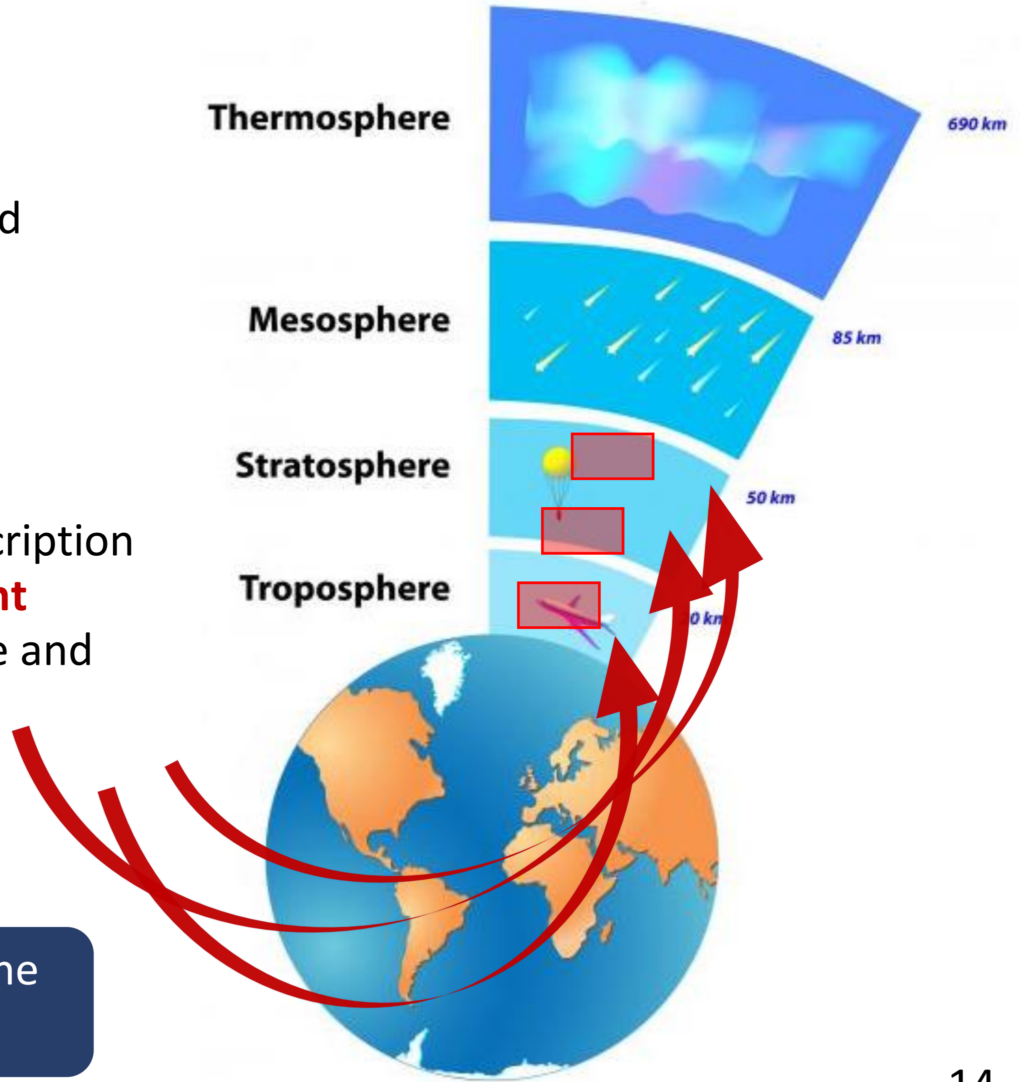
- Simulation of the CR induced atmospheric cascade

Our "DNA" models:

- Accurate e- interaction description <u>in small volumes</u> **at different altitudes** in the troposphere and lower Stratosphere.

↓

High characterization of the ions produced



Electron impact on N_2, O_2

1. Ionisation:

- **Method**: Relativistic Binary Encounter Bethe – **RBEB** (Kim, 2000)
- **Energy range**: threshold – 1 GeV
- EDCS for Molecular Orbital (4 Outer shells for N_2, 5 outer shells for O_2)

2. Elastic scattering:

- **Method**: Indipendent Atom Model (Mott and Massey 1965) calculated by means of the **ELSEPA code****
  - spherical complex optical potential formalism
- **Energy range**: tens of eV – 1 GeV
- Angular DCS

3. Electronic excitation: Work in progress

- probably different models will be used to cover the whole energy range and electronic states.

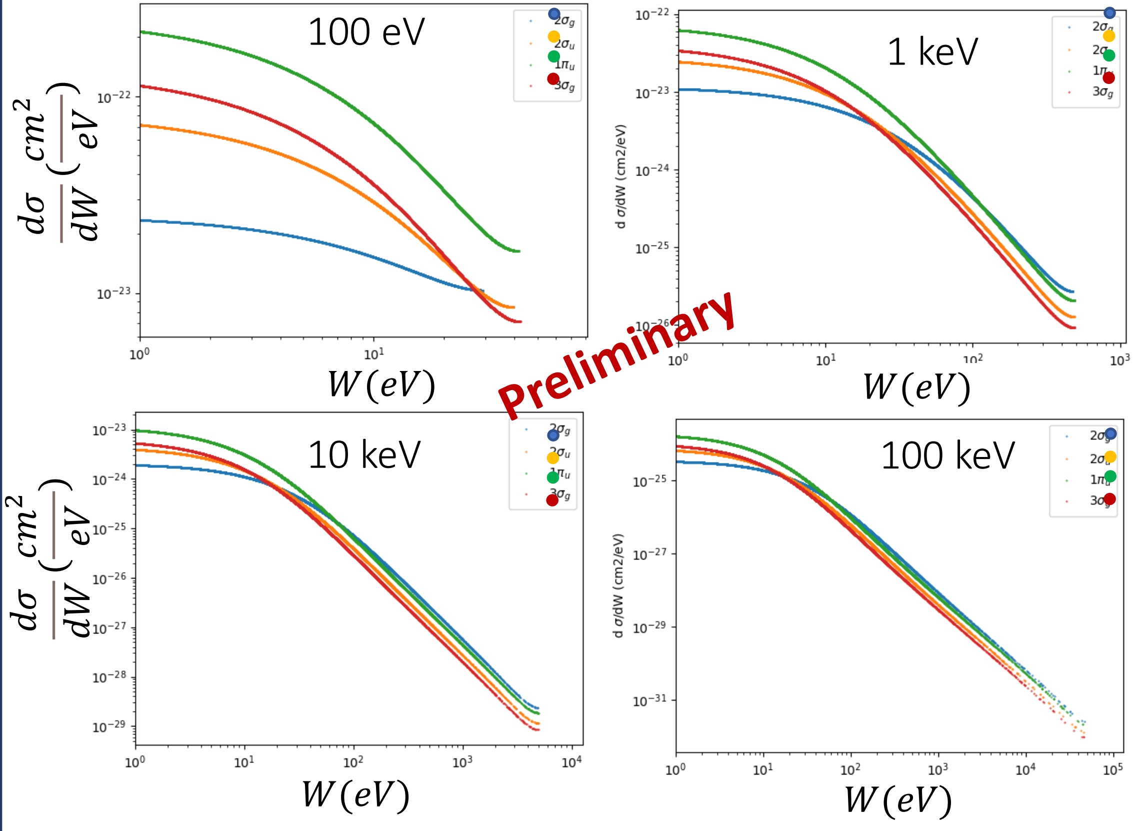
** "ELastic Scattering of Electrons and Positrons by neutral Atoms" code developed by Salvat et al. Freely available at *https://github.com/eScatter/elsepa*

RBEB implementation in GEant4DNA - N2

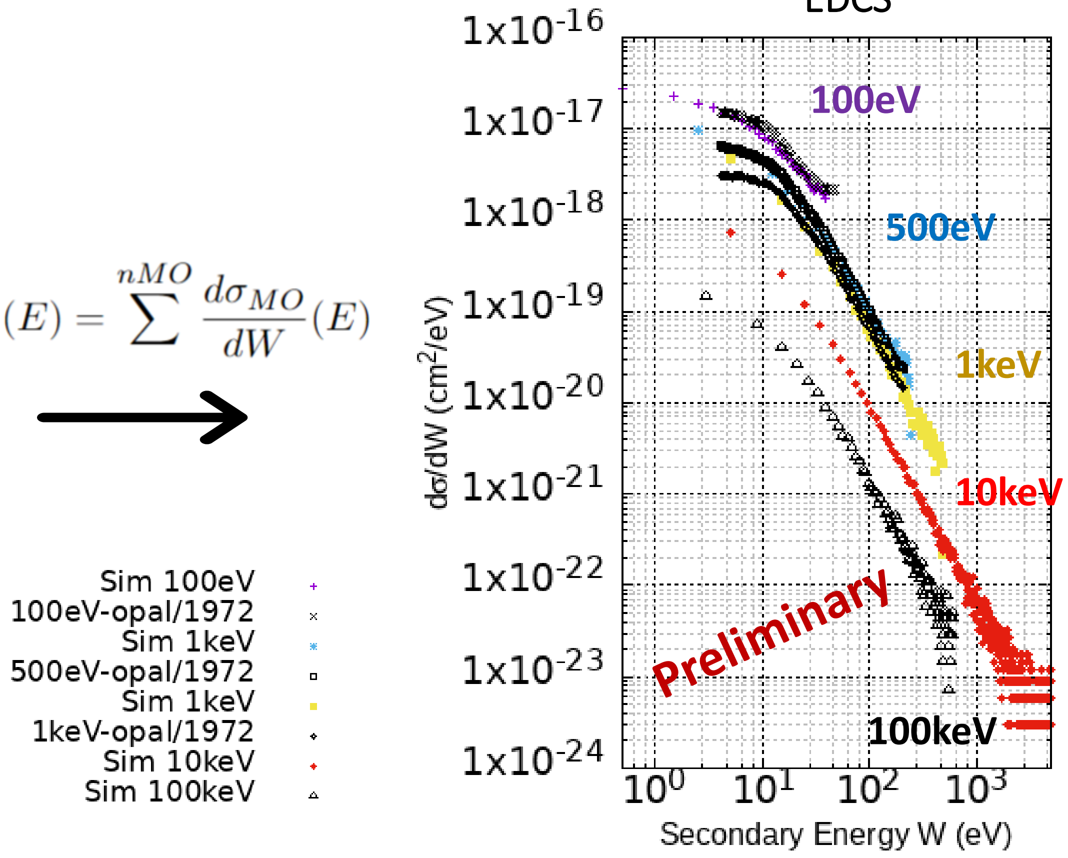
e^- impact ionization of N2:

- **Energy range:** threshold - 1GeV
- **Type** : analytical
- Model class: G4DNARBEBGas

EDCS for Molecular Orbital



$$\frac{d\sigma}{dW}(E) = \sum^{nMO} \frac{d\sigma_{MO}}{dW}(E)$$

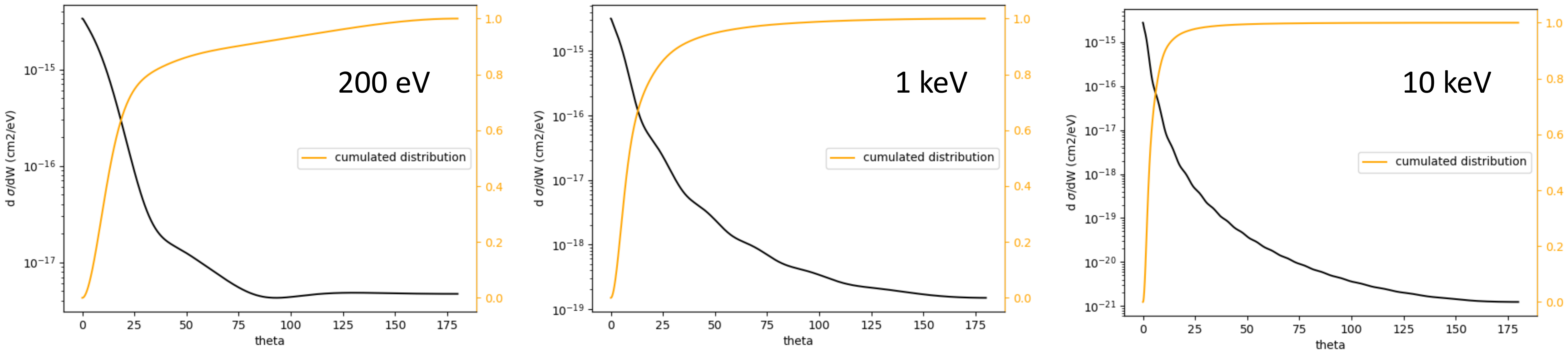


ELSEPA – implementation in Geant4DNA

e^- elastic scattering on N2, O2:

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

N2: DCS Cumulated distribution



Data files →

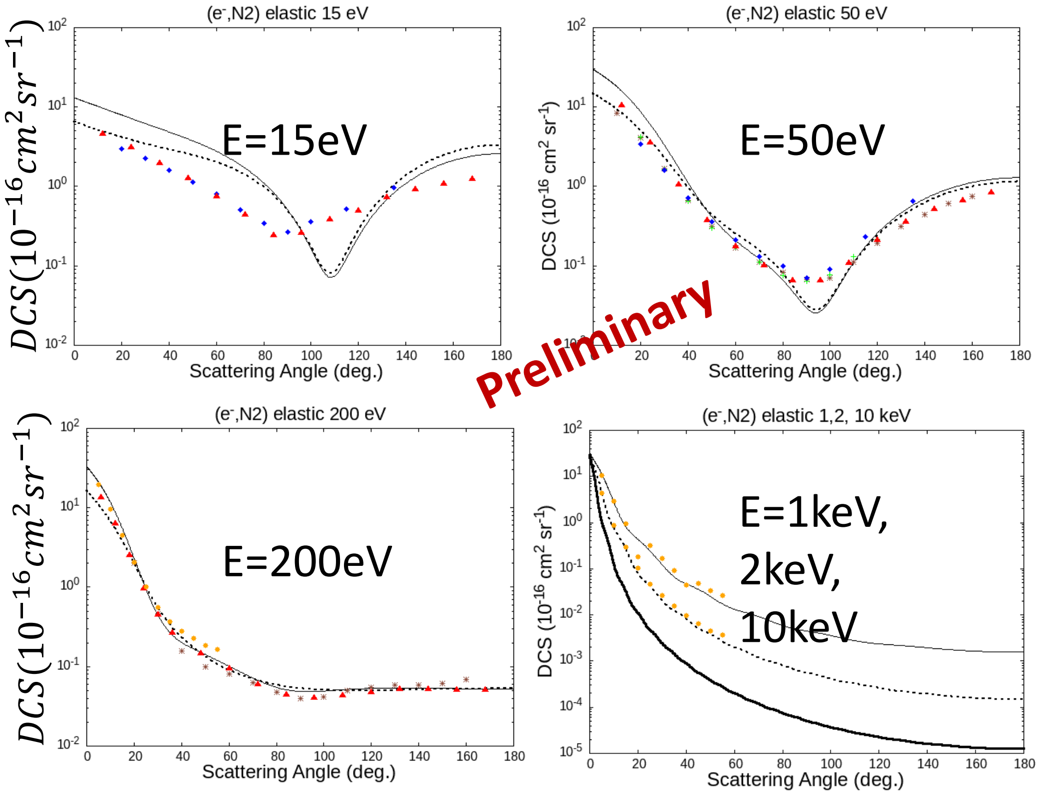
Sigmadiff_cumulated_elastic_e_N2_elsepa.dat
Sigmadiff_cumulated_elastic_e_O2_elsepa.dat

ELSEPA – implementation in Geant4DNA

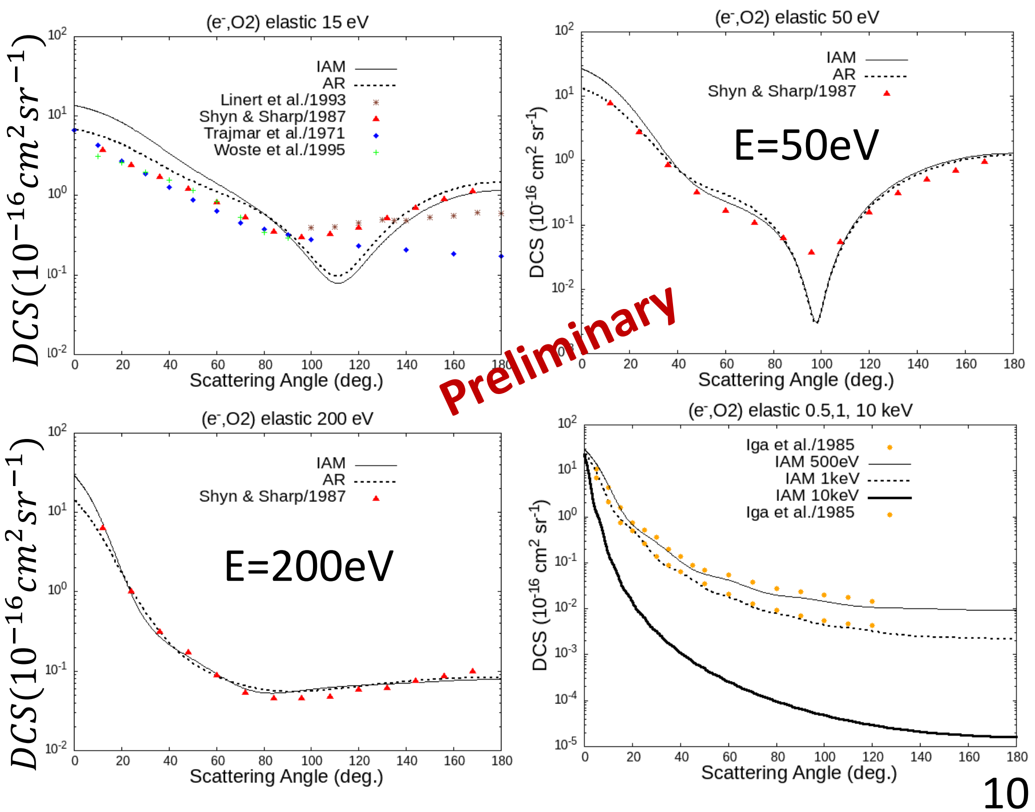
e^- elastic scattering on N2, O2:

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

DCS N2 per Molecular Orbital



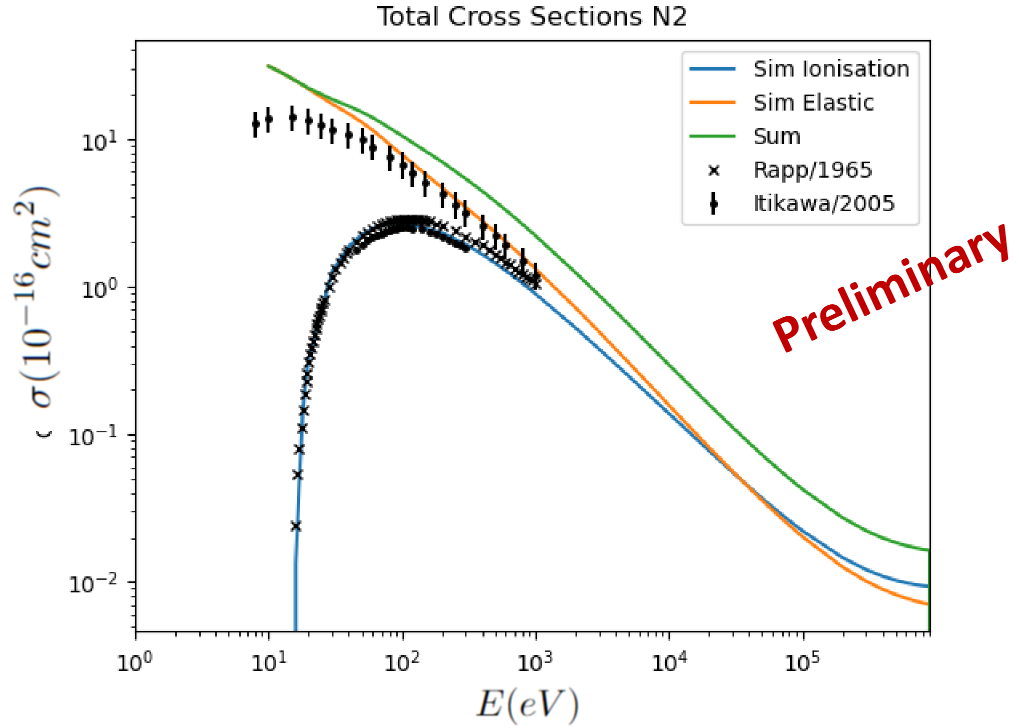
DCS O2 per Molecular Orbital



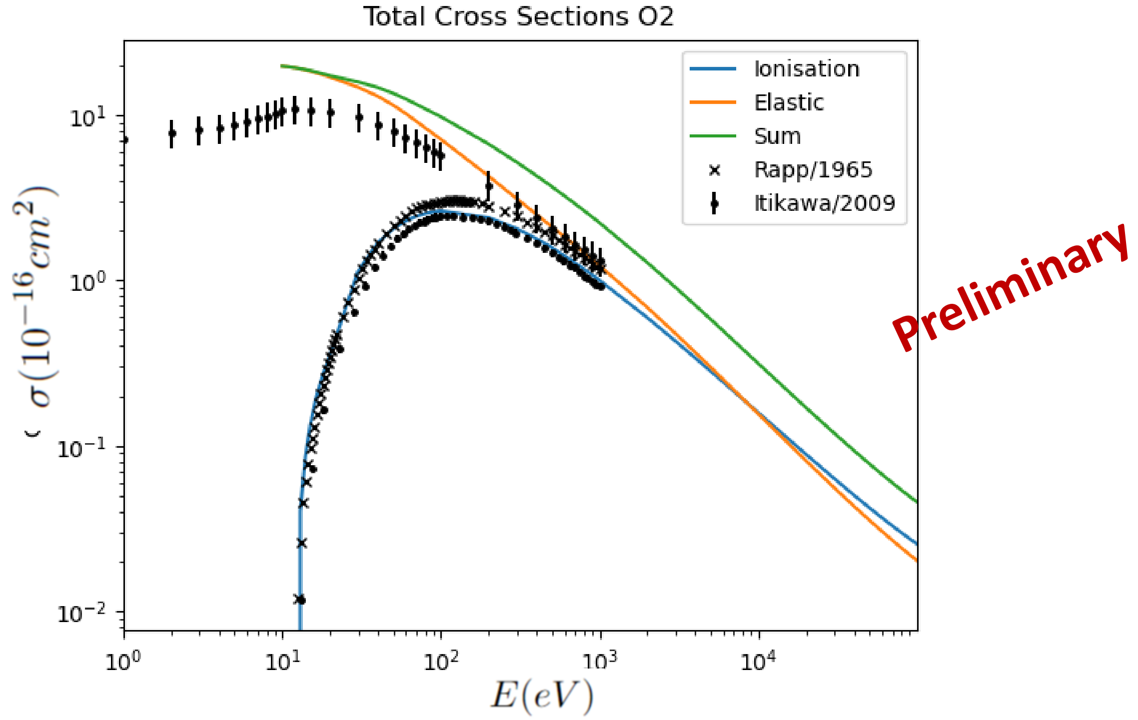
Ionisation/Elastic – Total CS

Ionisation CS → $$\sigma = \int_{W=0}^{Wmax} \frac{d\sigma}{dW}$$

Elastic CS → $$\sigma = \int_{0}^{2\pi} \frac{d\sigma}{d\Omega} 2\pi sin\theta d\theta$$



N_2 ion. threshold: 15.58 eV



O_2 ion. threshold: 12.07 eV

Work in progress and next steps

1. **Electronic excitation**: implementation in Geant4DNA
   - BEf-scaled Born method

     → for dipole-allowed transitions
   - Ab-initio R-matrix close-coupling method (UKRmol+ code)

     → For very low energies and resonances
   - Experimental data

2. **Testing and validation against reference data and other MC models:**
   - Comparison with predefined models from Geant4
   - Range calculation - using /dna/range example – and comparison with data
   - Stopping power calculation- using /dna/spower example – and comparison with data

Conclusions

- **New G4DNA models** to simulate electron impact **ionization and elastic scattering on N2 and O2 molecules** have been implemented in the energy range from threshold up to 1 GeV

- The results are in **good agreement with experimental data**

- Models suitable for new molecules in Geant4DNA:
  - **Potential impact on the physics of the atmosphere**
  - Open the way for **new space-related studies** regarding chemistry and exobiology
  - **Can be applied to any kind of Geant4-DNA simulation.**

Thank you for your attention !

francesca.nicolanti@uniroma1.it

Backup slides

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:

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

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)

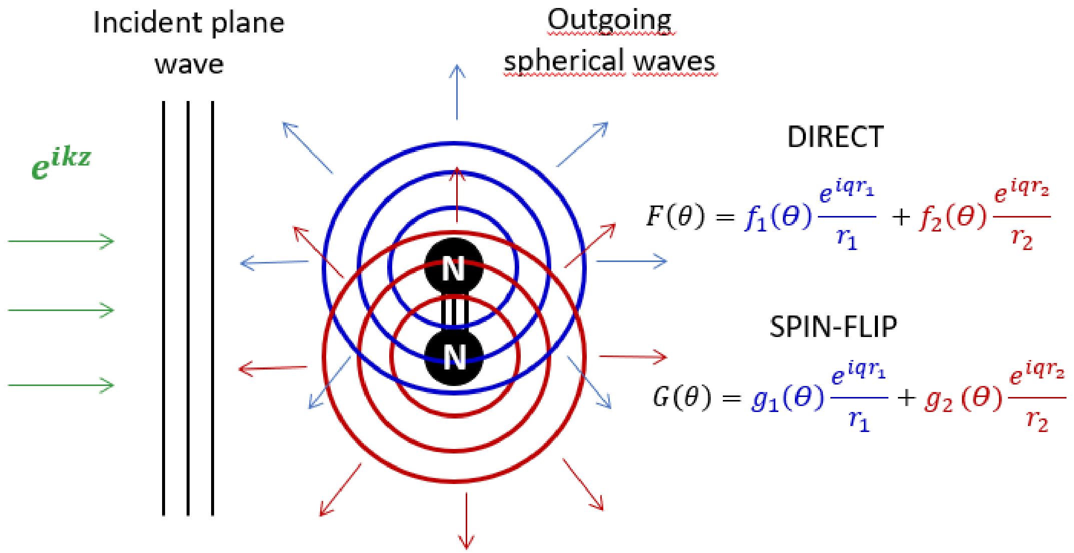
Material	Corresponding model	Class name	Energy range	Type
H_2O	G4DNACPA100IonisationModel	BEB*	11 eV - 255 keV	interpolated
Gold	G4DNARelativisticIonisationModel	MRBEBV**	8.3 eV - 1 GeV	analytical

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

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 interaction potential

Optical potential model:

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

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

- Potential model:

  Nuclear charge: Fermi distribution;

  Electron density: Dirac–Fock distribution.

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

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 $b^2_{pol} = \max[(E - 20\text{ eV})/\text{eV}, 1]$.

Exchange potential $V_{ex}(r)$

Potential model:

Furness–McCarthy potential.

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

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 $A_{abs} = 2$

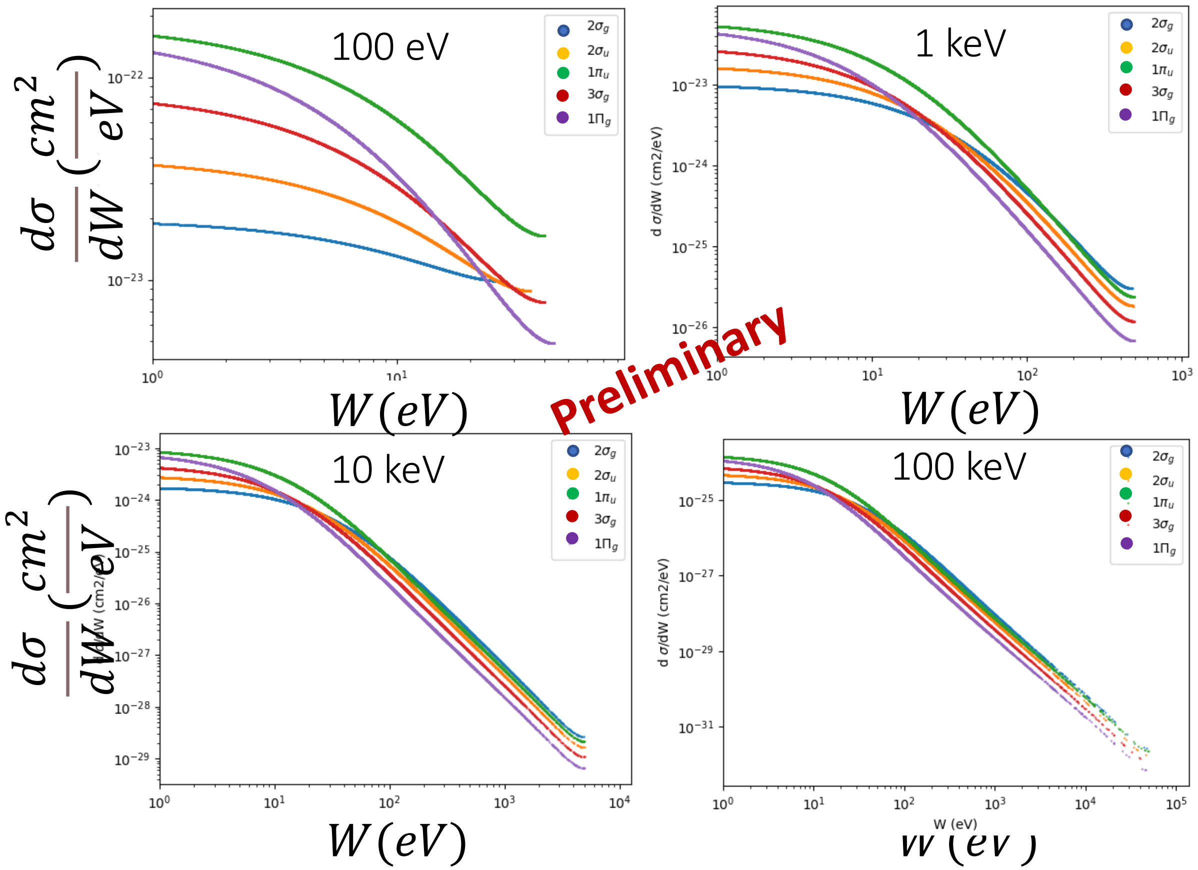
<u>N.B</u>. Empirical parameters are **validated for noble gases** and mercury

RBEB - implementation in GEant4DNA O2

e^- impact ionization of O2:

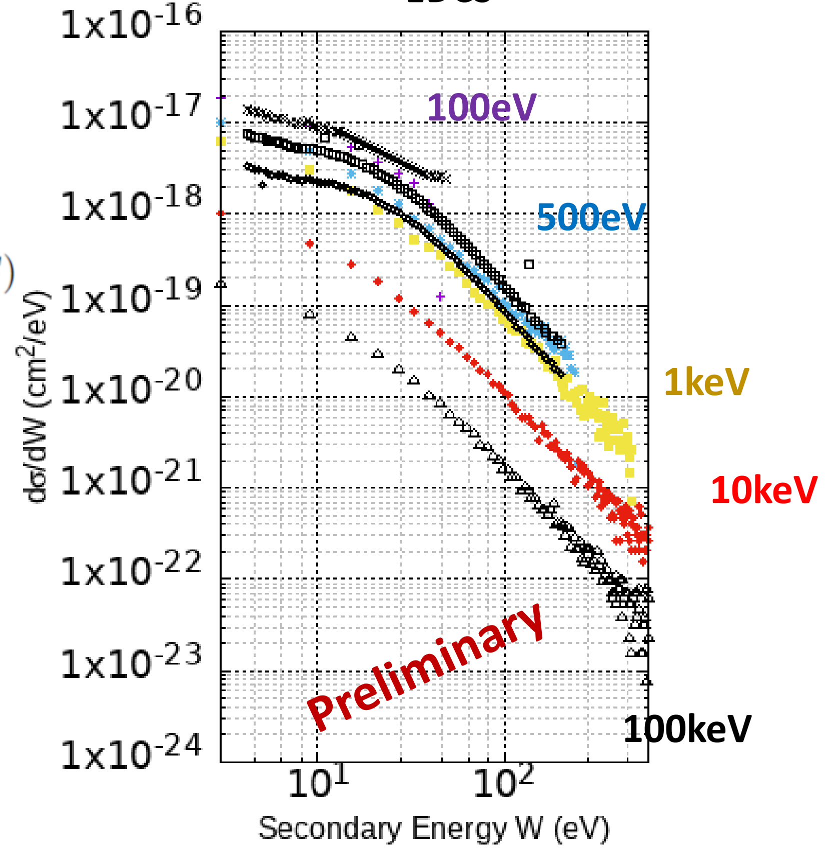
- **Energy range:** threshold - 1GeV
- **Type** : analytical
- Model class: G4DNARBEBGas

EDCS for Molecular Orbital



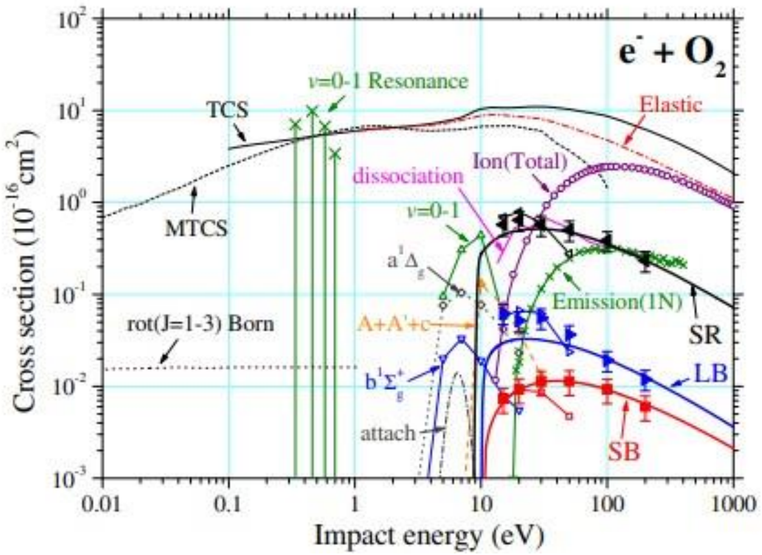
$$\frac{d\sigma}{dW}(E) = \sum^{nMO} \frac{d\sigma_{MO}}{dW}(E)$$

EDCS



Excitation CS

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



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

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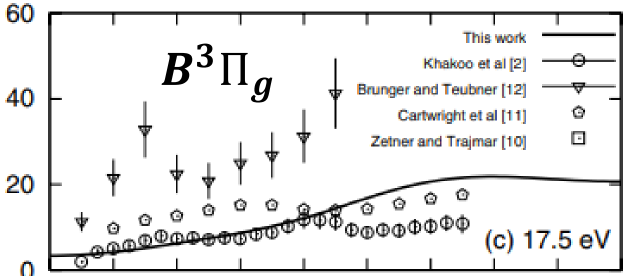
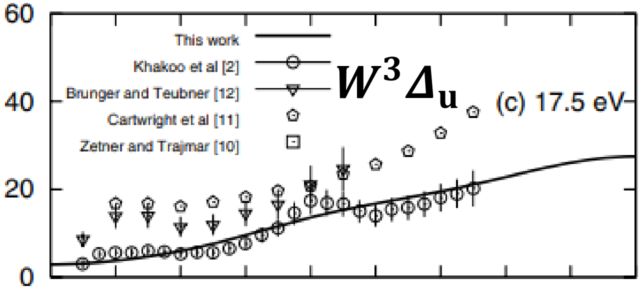
Electro collisions with molecular nitrogen in its ground and electronically excited states using the R-matrix method

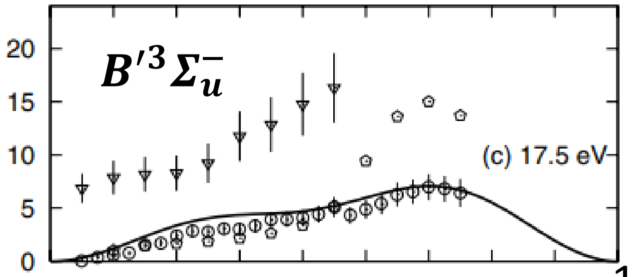
He Su[1], Xinlu Cheng[2], Hong Zhang[1,*] and Jonathan Tennyson[3,*]

Tashiro and Murukuma (2007):

- Experimental data







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