Vibrational excitation cross sections calculations with Quantemol Electron Collisions (QEC) - 1.2

Quantemol breakfast seminar, 25/11/2020

M.Tudorovskaya

Overview

- Electron scattering data for plasma physics
- The theory: R-matrix method
- Quantemol Electron Collisions software
- Vibrational excitation

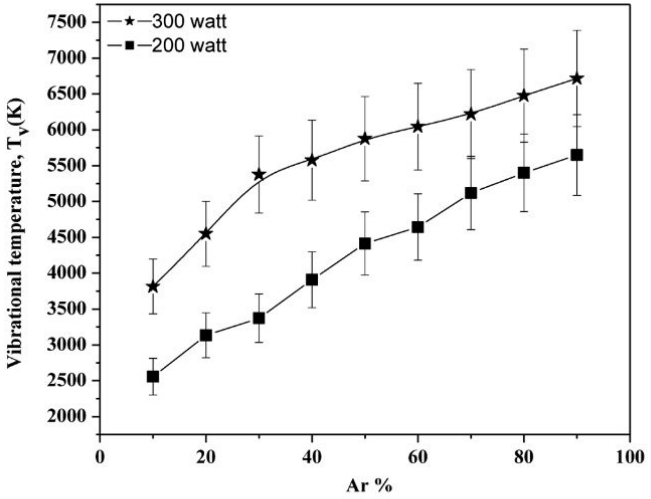
Vibrational excitation in plasma

Example:

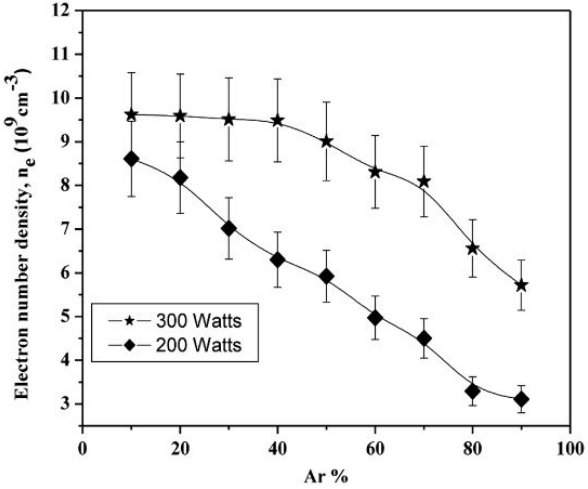
Effect of Excitation and Vibrational Temperature on the Dissociation of Nitrogen Molecules in Ar-N_2 Mixture RF Discharge

F. U. Khan , N. U. Rehman , S. Naseer , M. Y. Naz , N. A. D. Khattak & M. Zakaullah

Spectroscopy Letters: An International Journal for Rapid Communication, 44:3, 194-202



Vibrational temperature versus argon percentage in the mixture for input powers of 200 watt and 300 watt with 30-Pa filling gas pressure.



Electron number density as a function of argon percentage with 0.3-mbar filling gas pressure, input powers of 200 watt and 300 watt.

Processes of interest for plasma chemistry

Elastic scattering (also: momentum transfer)

$AB + e \longrightarrow AB + e$

Electron-impact electron excitation (be electron excited state)

$AB + e \longrightarrow AB^* + e$

Electron-impact vibrational excitation (to the first excited state)

$AB + e \longrightarrow AB + e$

Electron-impact rotational excitation (by rotational state)

$AB + e \longrightarrow AB + e$

Dissociative electron attachment (may require additional considerations)

$AB + e \longrightarrow AB^- \longrightarrow A + B^-$

Ionization (and dissociative ionization)

$AB + e \longrightarrow AB^+ + \cancel{e} \longrightarrow A + B^+$

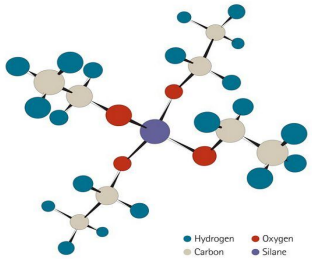
Dissociation

$AB + e \longrightarrow AB + \cancel{e} \longrightarrow A + B + e$ (different fragments)

Electron scattering R-matrix theory

e^-

Inner region



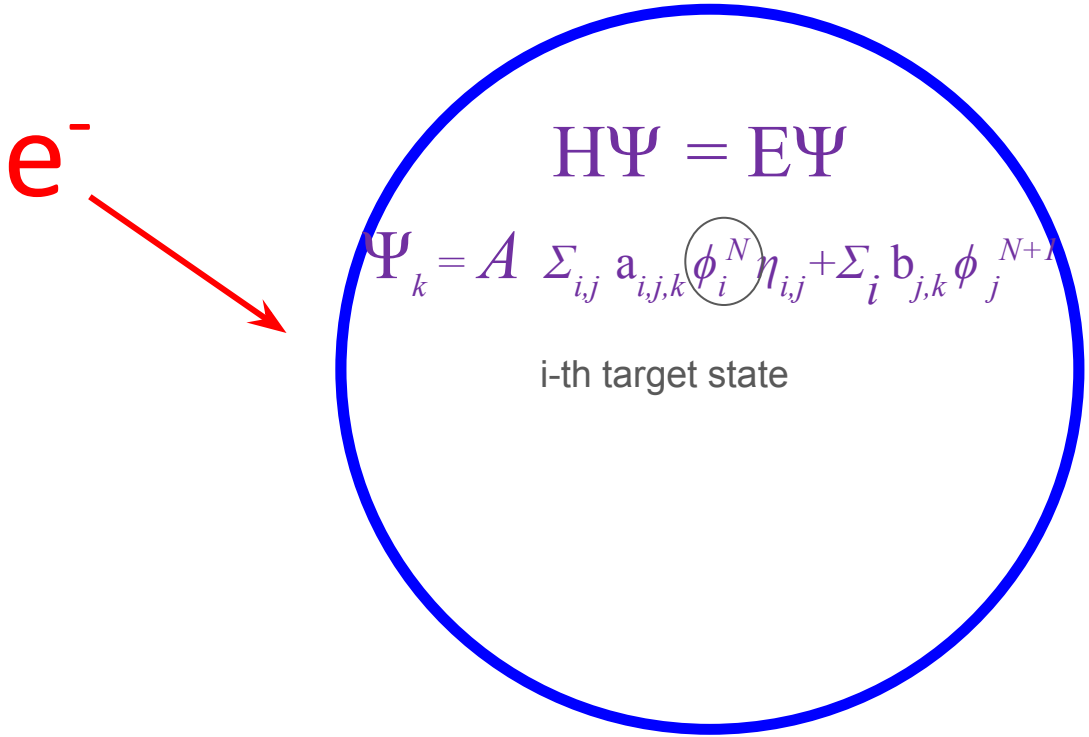
Boundary:

solutions matched;
Coefficients are contained in the R-matrix

Outer region

Radial solution in effective potential

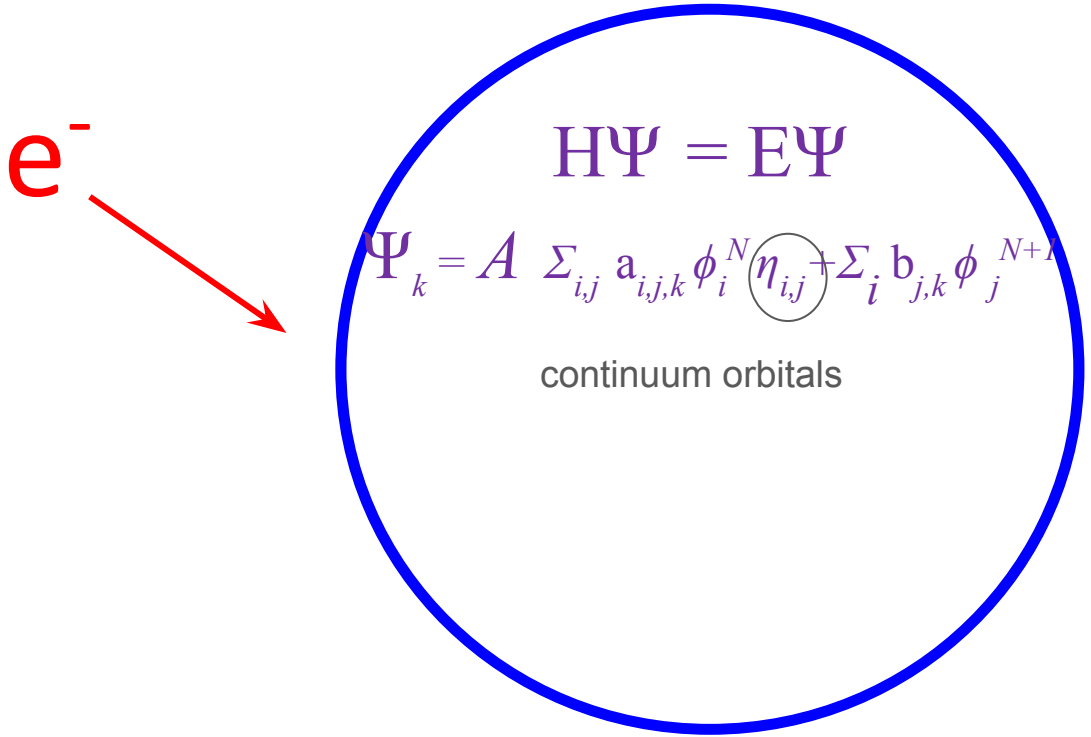
Electron scattering R-matrix theory



The N-electron molecule - “target” - is modeled assuming the nuclei do not move

N+1 electron problem for the time-independent Schrödinger equation

Electron scattering R-matrix theory



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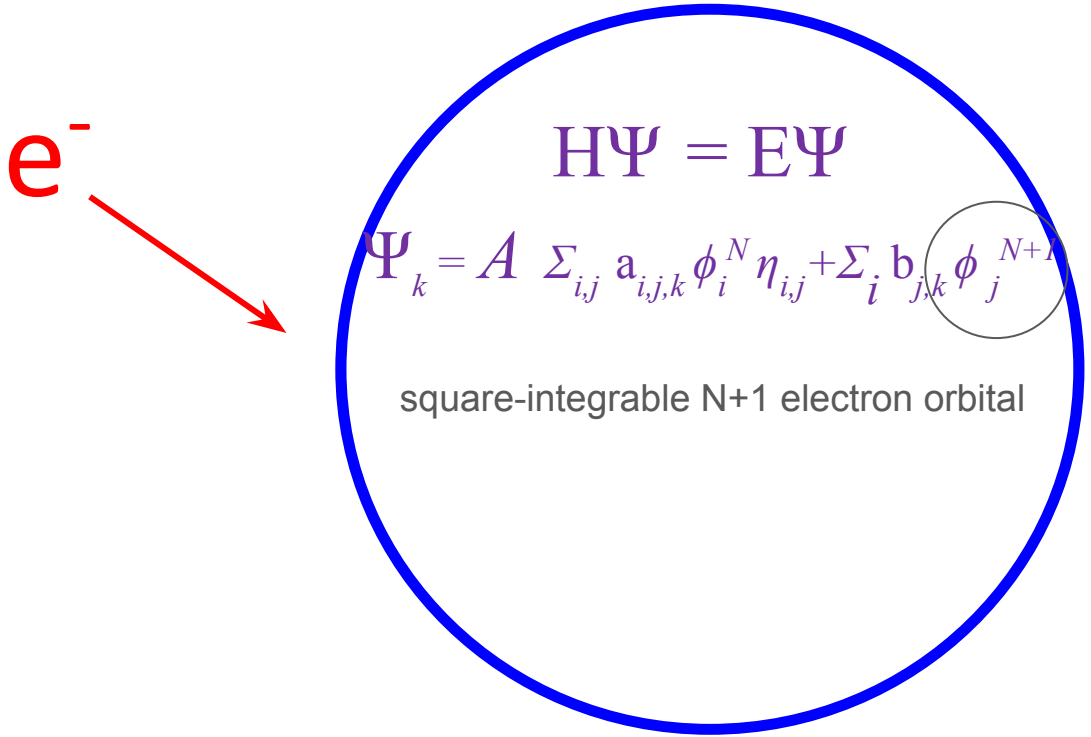

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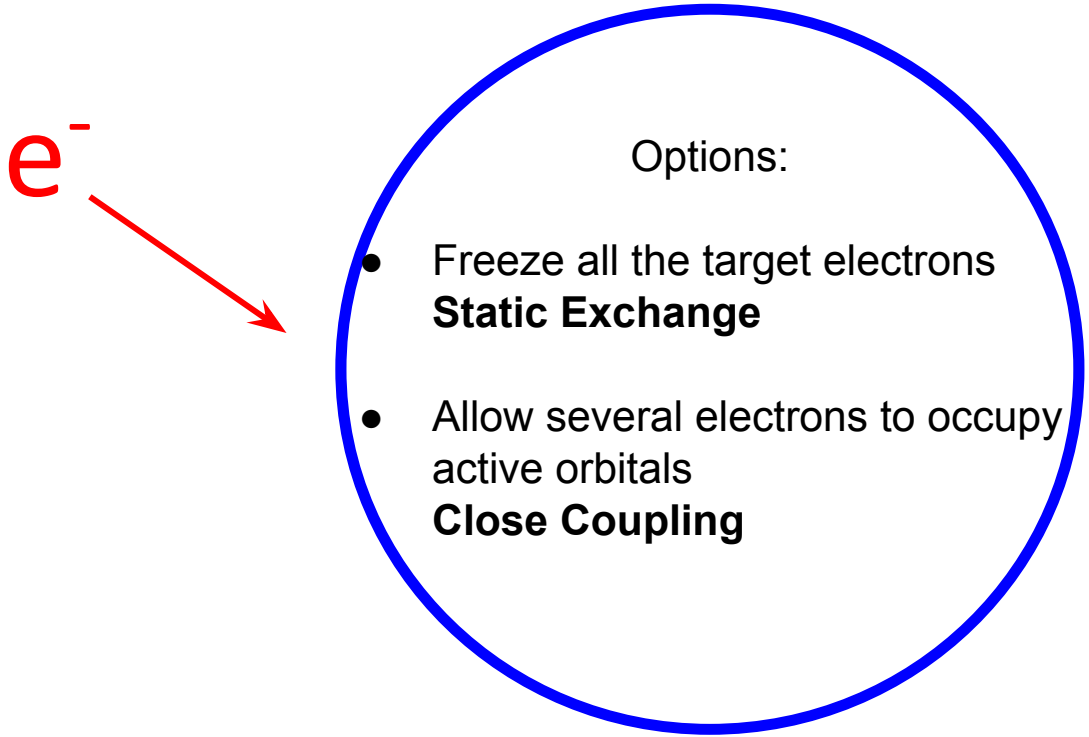

Quantemol Electron Collisions (QEC)

UKRMol+/Python

An upgraded product for faster calculations & bigger molecules

Quantemol Electron Collision: an expert system for performing UKRmol+ electron – molecule collision calculations

B. Cooper, M. Tudorovskaya, S. Mohr, A O'Hare, M. Hanicinec, A. Dzarasova, J. Gorfinkiel, J. Benda, Z. Masin, A. Al-Refaie, P J Knowles, J. Tennyson

Atoms 2019, **7**, 97

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Atoms 2019, **7**, 97

Watch the demo...

Vibrational excitation: theory

(Viatcheslav Kokoouline and collaborators, University of Central Florida)

- Rotation is neglected
- Small changes in the geometry for the partial derivatives

$$<\sigma> = \frac{\pi\hbar^2}{4m\epsilon} \sum_{i=1}^{3N-5} \sum_{ll'\lambda\lambda'} \left| \frac{\partial S_{l\lambda,l'\lambda'}}{\partial q_i} \right|^2$$

Scattering matrix;
q - effective coordinate

$$\hat{S} = \frac{\hat{1} + i\hat{K}}{\hat{1} - i\hat{K}}$$

Data from QEC!

K-matrices for stretched geometries for each mode contain the outer-region asymptotic solutions

[1] Rolf Güsten, et al. Nature 568, (2019) 357–359
[2] S. Fonseca dos Santos, N. Douguet, V. Kokoouline, A. E. Orel, J. Chem. Phys. 140, (2014), 164308
[3] J. Tennyson, et al. J. Phys.: Conf. Ser. 86, (2007), 012001
[4] Mehdi Ayouz, Viatcheslav Kokoouline, Atoms. 4(4) (2016) 30

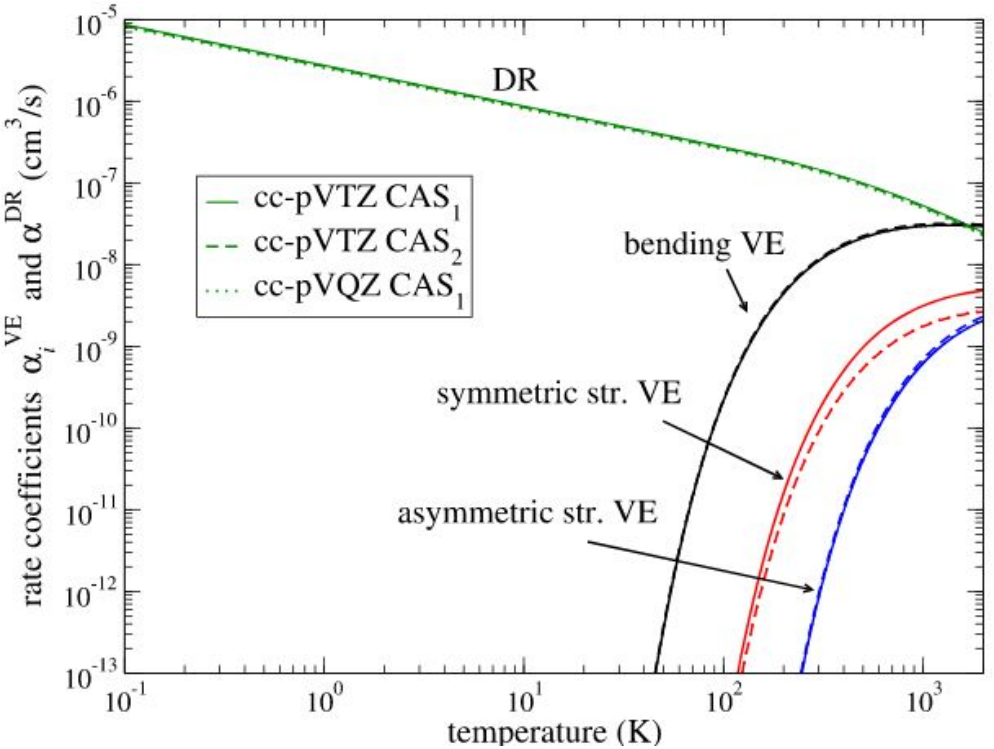
Vibrational excitation

Viatcheslav Kokoouline et al
2018 Plasma Sources Sci. Technol.27 115007

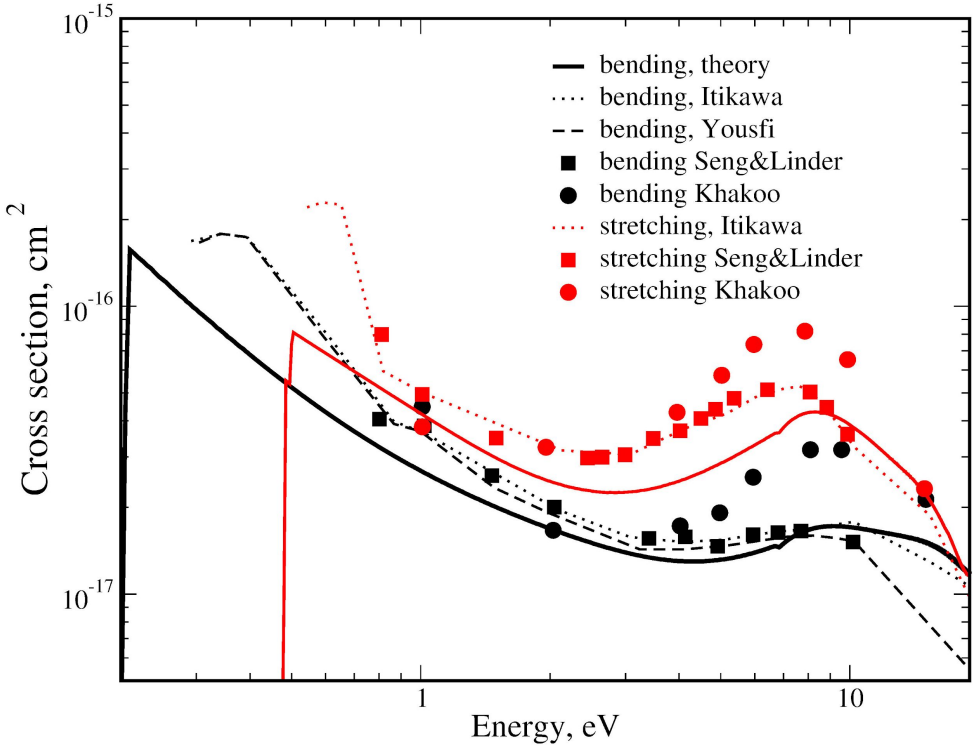
BF_3^+:

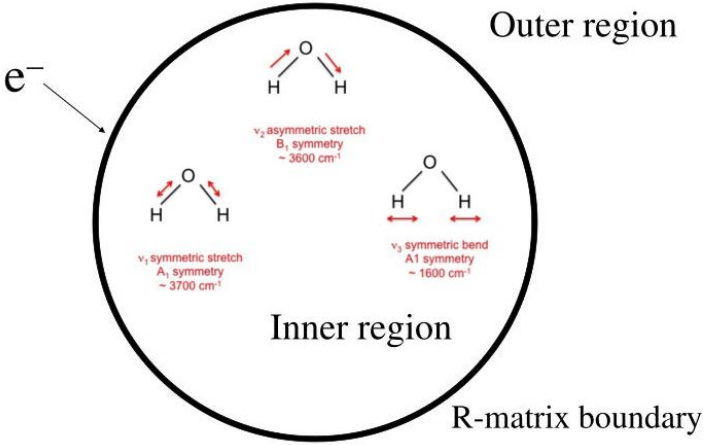
- Dissociative recombination(DR)
- Vibrational excitation (VE)

Different modes (bending, stretching)
Different basis sets



Vibrational excitation: H_2O

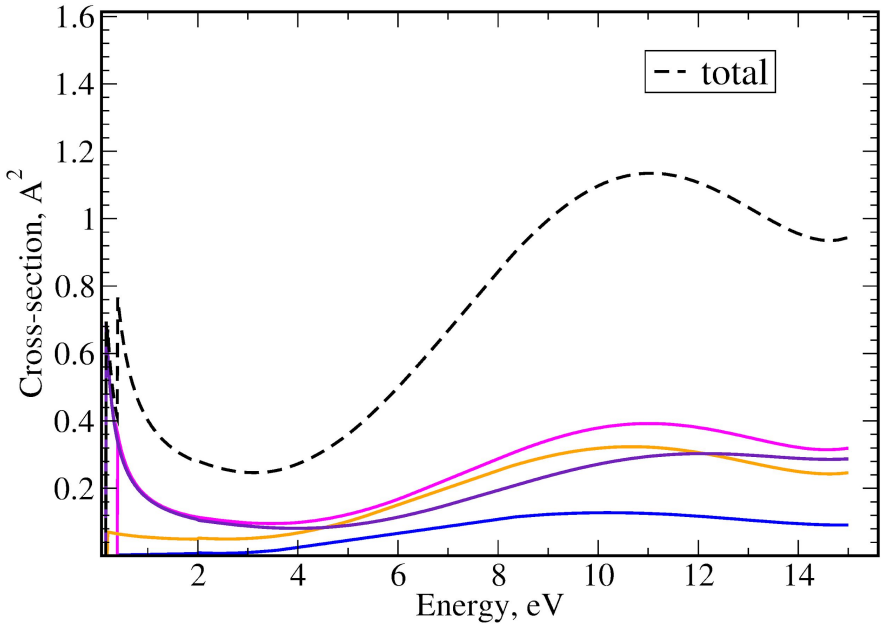




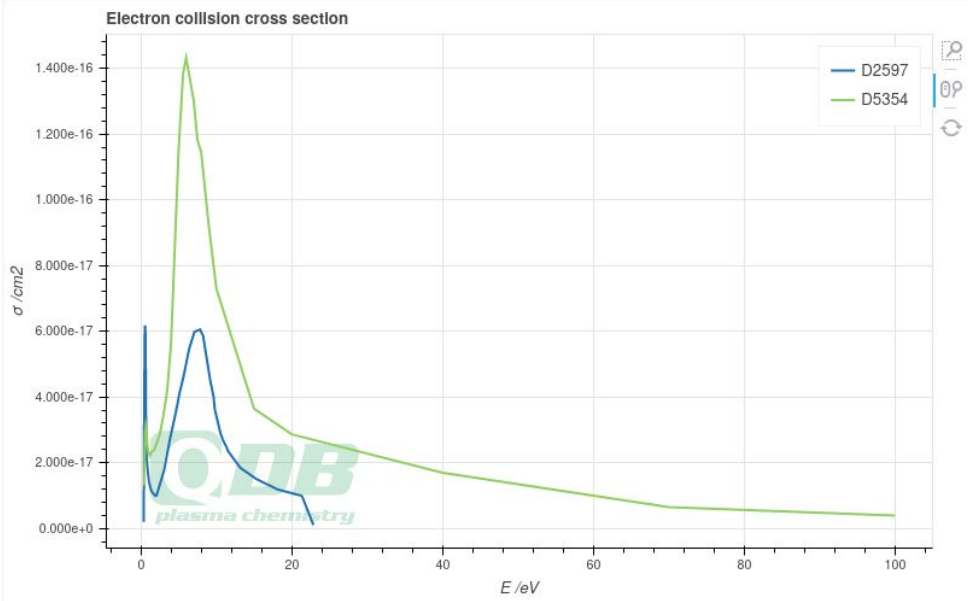
The theory also allows to distinguish all the modes:

Generally, different modes lead to different dissociation paths

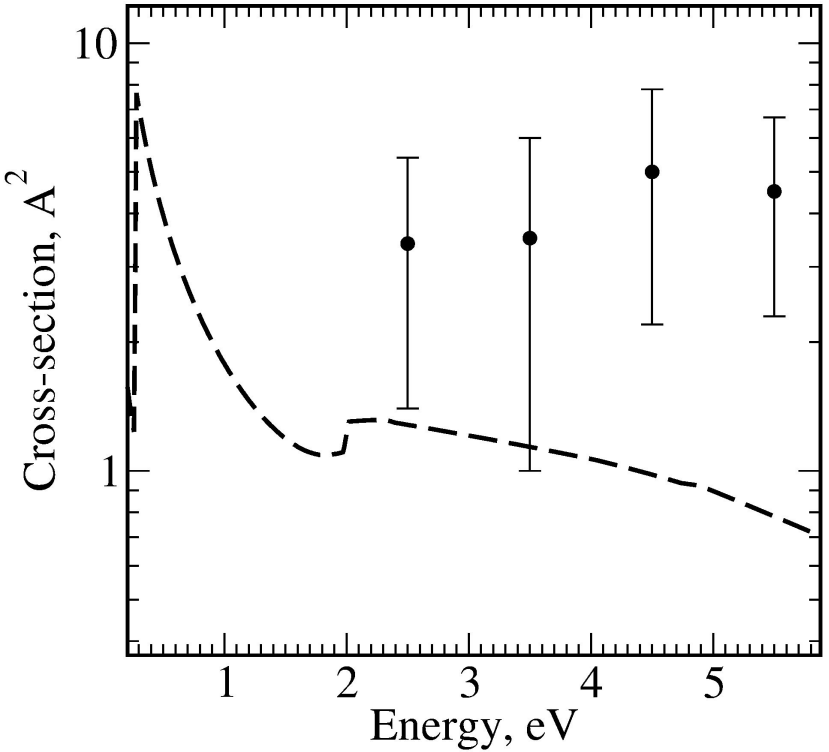
Vibrational excitation: CH_4



www.quantemoldb.com



Vibrational excitation: OCS



Can be used as an estimator for systems with resonances

Thanks &
and listen about further development

Free trial version is available

Email support@quantemol.com