Electron (and positron) scattering cross sections for low temperature plasmas: experimental and semiempirical

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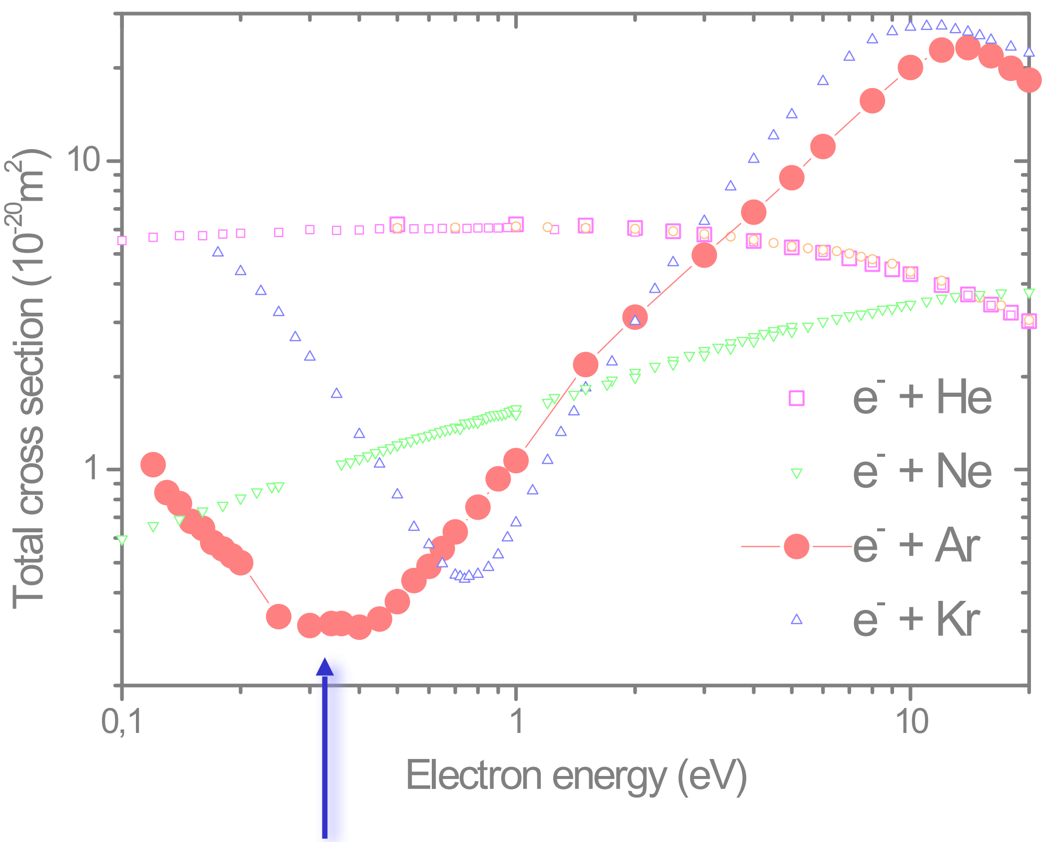
IAEA Experimental Network Meeting,
Wien, 19-21.11.2018

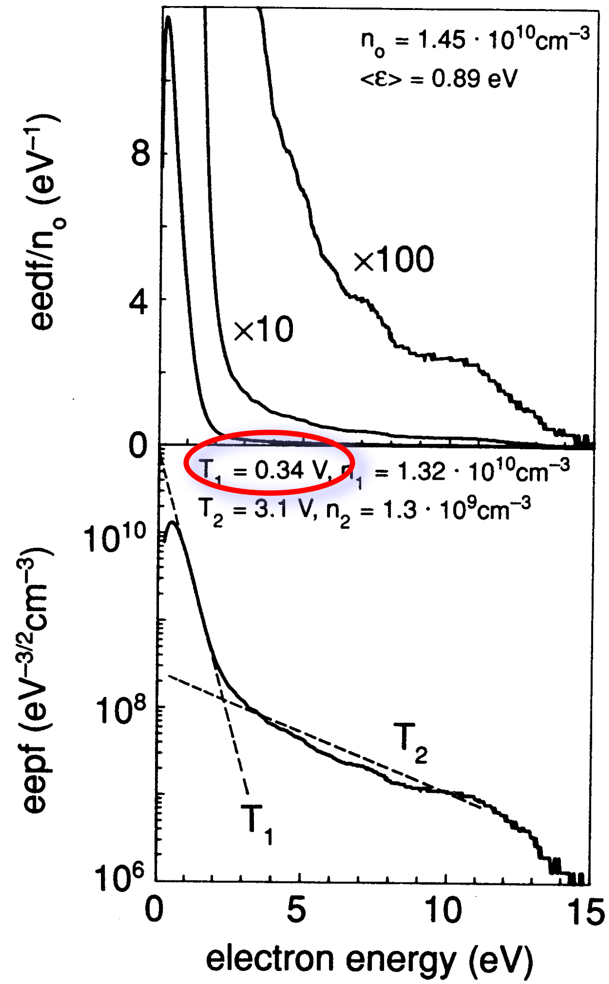
Electron scattering:

- Why we need cross sections?
- Which cross sections do we need?
- Total scattering cross sections
- Ionization cross sections
- Electronic excitation – state of art
- Polar molecules – what can be done?

Plasma temperature ← integral cross sections

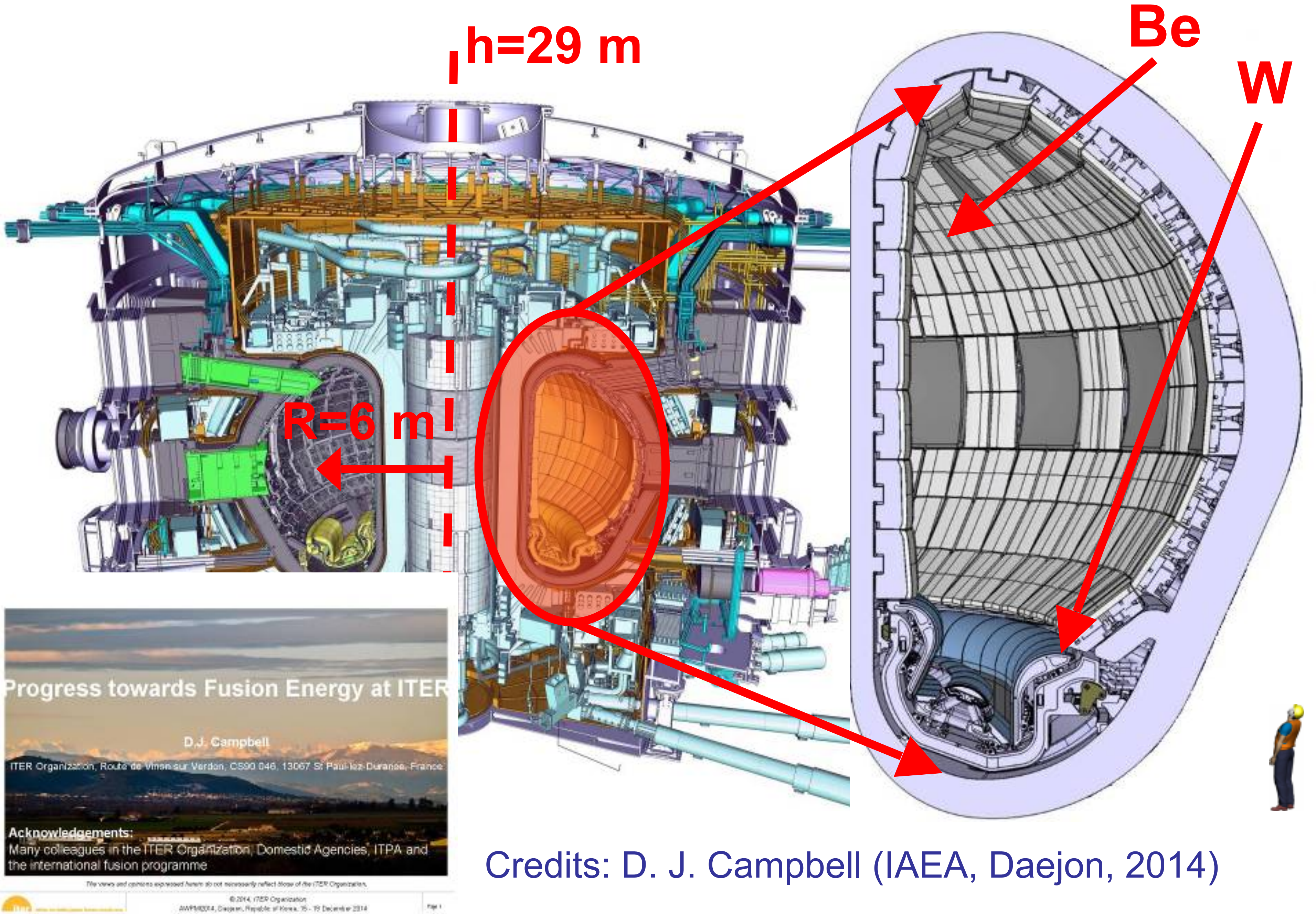


Ramsauer minimum (zero in *s*-wave)



V. Godyak, Sendai 2006

The ITER Tokamak



Progress towards Fusion Energy at ITER

D.J. Campbell

ITER Organization, Route de Vinon sur Verdon, CS90 046, 13067 St Paul-lez-Durance, France

Acknowledgements:
Many colleagues in the ITER Organization, Domestic Agencies, ITPA and the international fusion programme

The views and opinions expressed herein do not necessarily reflect those of the ITER Organization.

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Credits: D. J. Campbell (IAEA, Daejon, 2014)

Rationale: edge and divertor plasma

Influence of atomic physics on EDGE2D-EIRENE simulations of JET divertor detachment with carbon and beryllium/tungsten plasma-facing components

Table 3. Atomic and molecular reactions included in the physics models used in EIRENE (also valid for D).

NIMBUS-like model	Kotov-2008 model
(1) $e + H^0 \rightarrow 2e + H^+$	Same reactions as default plus:
(2) $H^+ + H^0 \rightarrow H^0 + H^+$	(9) $H_2 + H^+ \rightarrow H^+ + H_2$
(3) $e + C^0 \rightarrow 2e + C^+$	(10) $H_2 + H^+ \rightarrow H_2^+ + H^0$
(4) $e + H_2 \rightarrow 3e + 2H^+$	(11) $e + H_2 \rightarrow 2e + H_2^+$
(5) $e + H_2 \rightarrow e + 2H^0$	(replacing (4))
(6) $e + H_2 \rightarrow 2e + H^+ + H^0$	(12) $e + H_2^+ \rightarrow e + H^0 + H^+$
(7) $e + H^+ \rightarrow H^0$	(13) $e + H_2^+ \rightarrow 2e + 2H^+$
(8) $2e + H^+ \rightarrow e + H^0$	(14) $e + H_2^+ \rightarrow 2H^0$
No CRM[a] for (4), (5) and (6)	CRM[a] for (11), (5) and (6)

[a] Collisional Radiative Model.

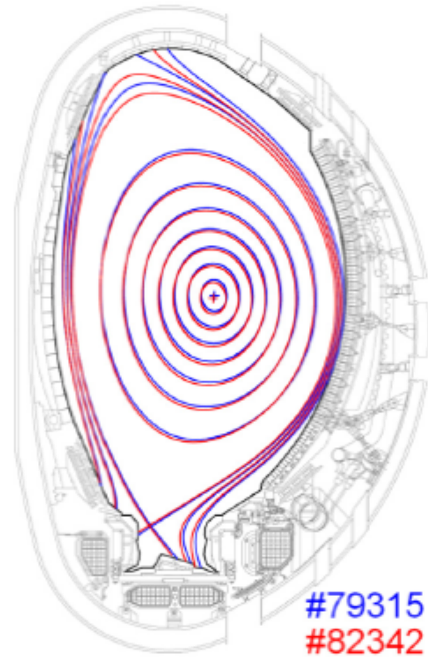


Figure 1. Magnetic equilibria for the shots #79315 and #82342 at 20 s and 13 s, respectively.

Guillemaut et al. Nucl.Fusion **54** (2014) 093012

Data needed:
I Neutrals (H, C, C_2, Be, BeH_2, CH_4)

1. Total cross section
2. Partial cross sections:

elastic scattering $e + A \rightarrow e + A$

rotational excitation $e + CH_4\ (J=0) \rightarrow e + CH_4\ (J=2)$

vibrational excitation $e + AB\ (v=0) \rightarrow e + AB\ (v>0)$

electron attachment (dissociative) $e + AB \rightarrow A^- + B$

electronic excitation $e + A \rightarrow e + A^*$

emission lines: $A^* \rightarrow A + h\nu$

neutral dissociation $e + AB \rightarrow A + B + e$

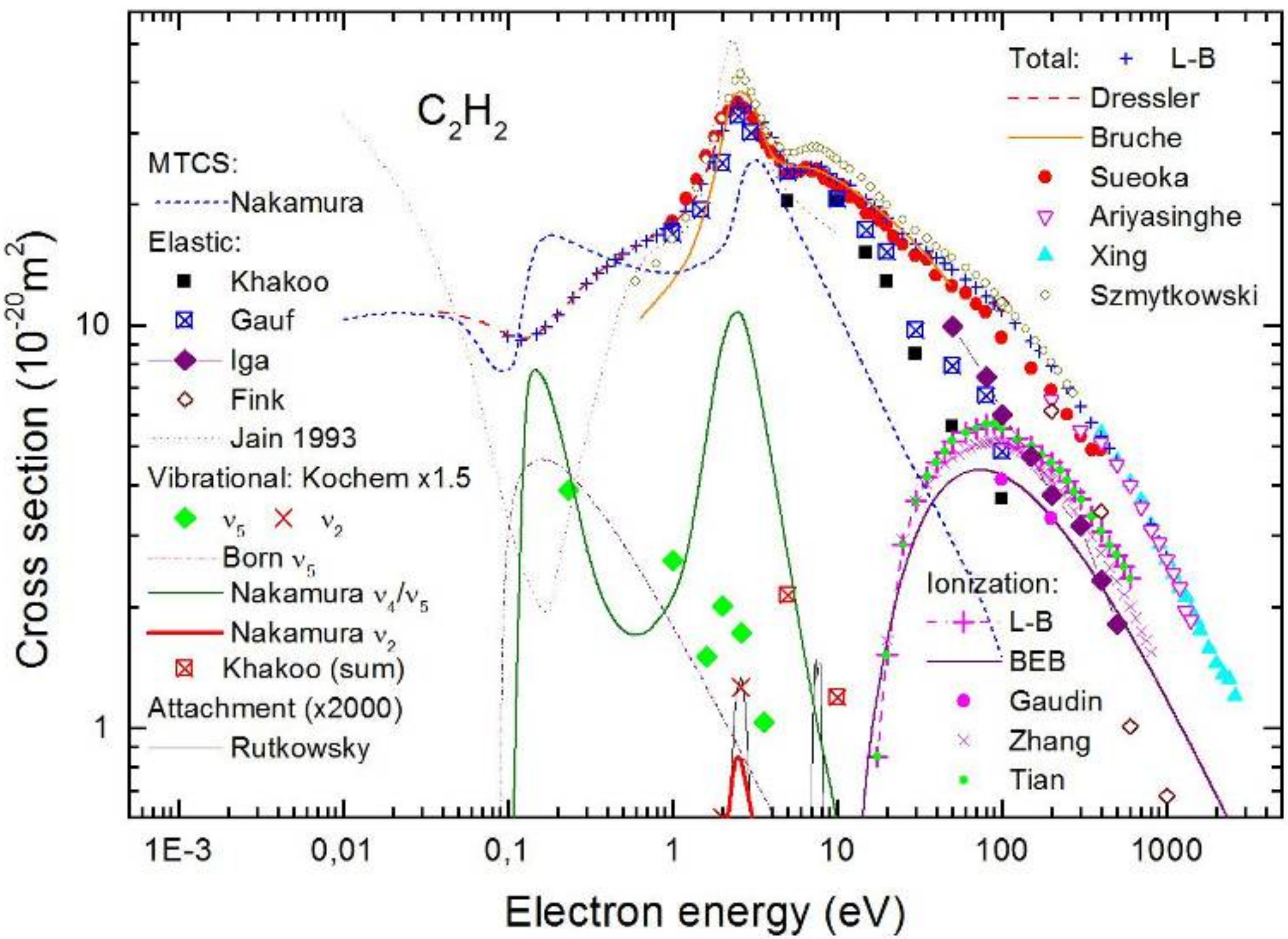
emission from dissociation $e + AB \rightarrow A^* + B + e + h\nu$

ionization $e + A \rightarrow A^+ + 2e$

dissociative ionization $e + AB \rightarrow A + B^+ + 2e$

ionization into excited states $e + A \rightarrow (A^+)^* + 2e$

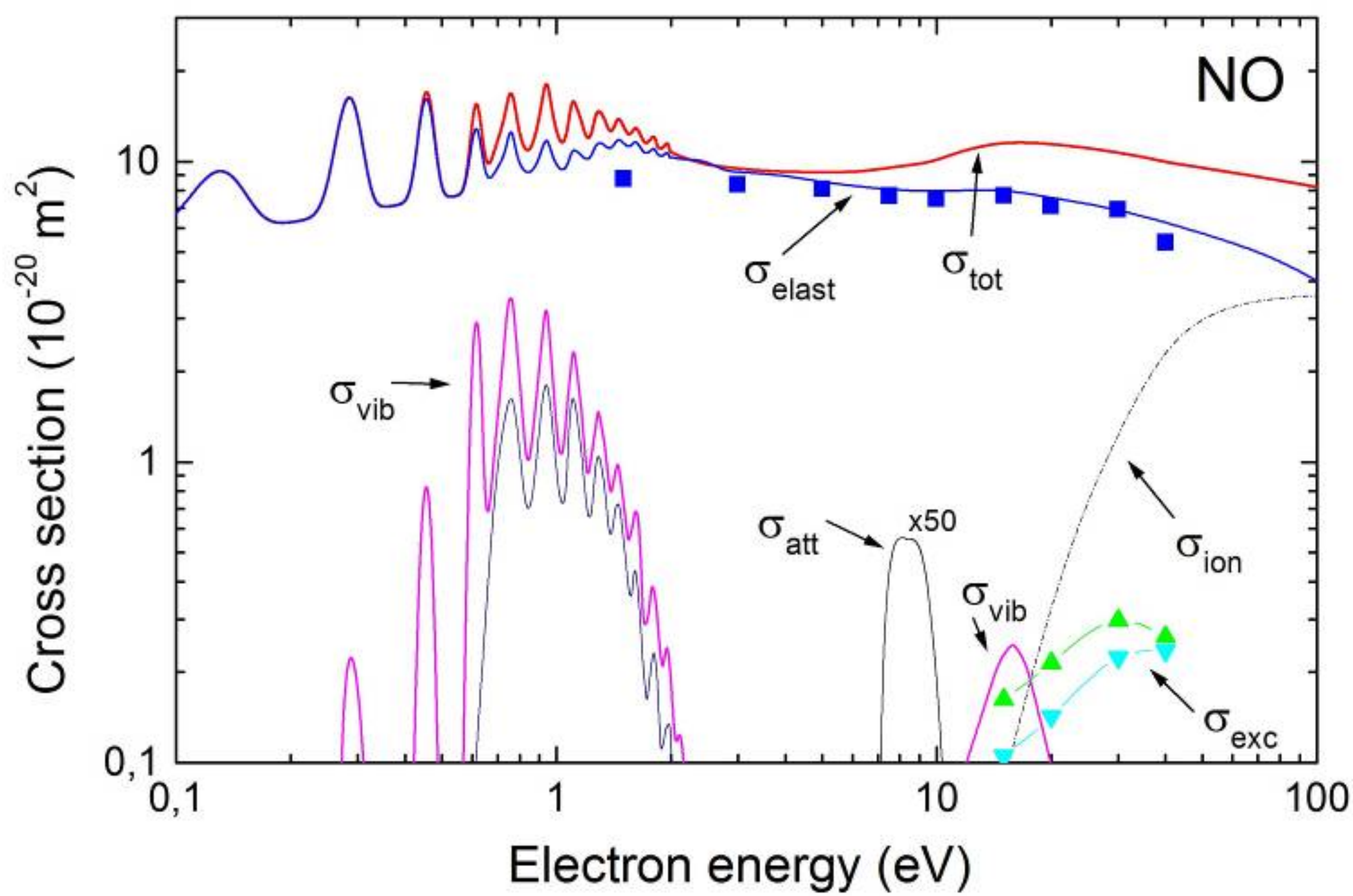
C_2H_2 - review



For some processes, like vibrational excitation different determinations (beam, swarm, semi-empirical) seriously disagree

M.-Y. Song, J. S. Yoon, H. Cho, Y. Itikawa, G. Karwasz, V. Kukooulin, Y. Nakamura, J. Tennyson, J. Phys. Chem. Ref. Data 46 (2017) 013106

NO review - preliminary

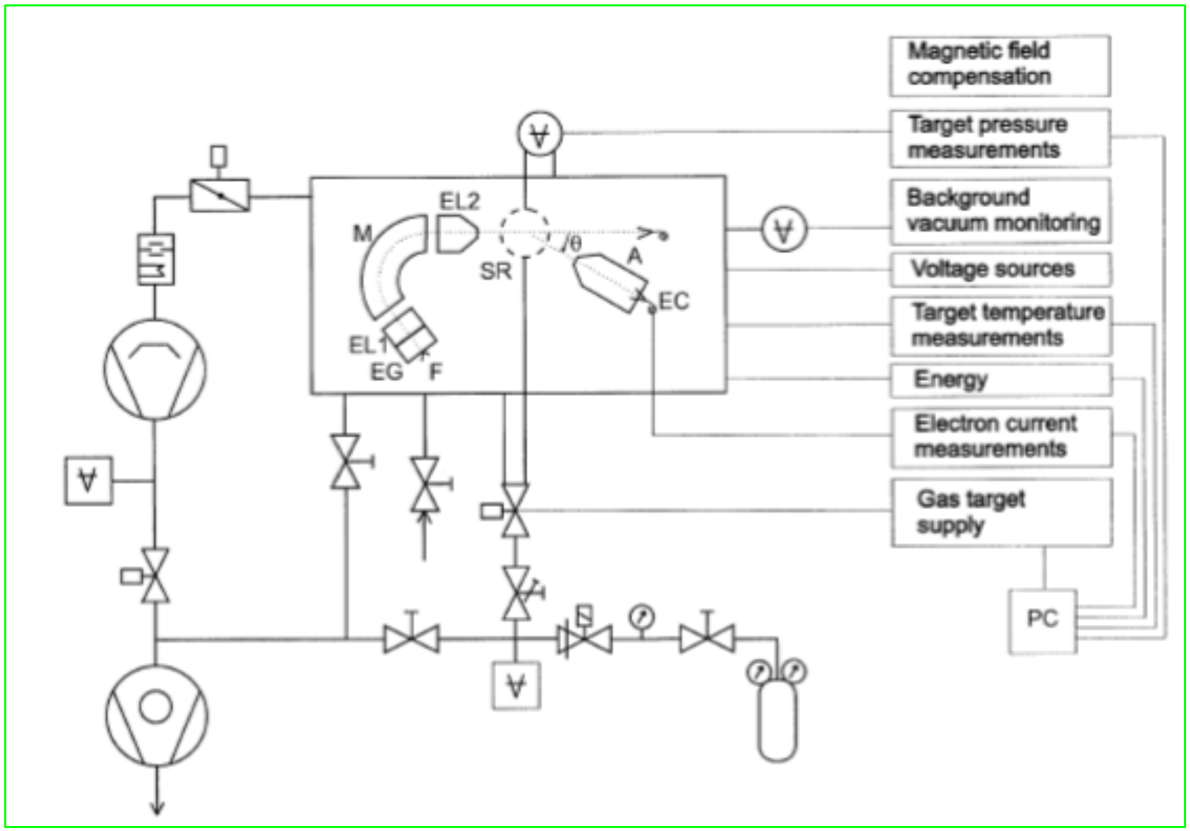


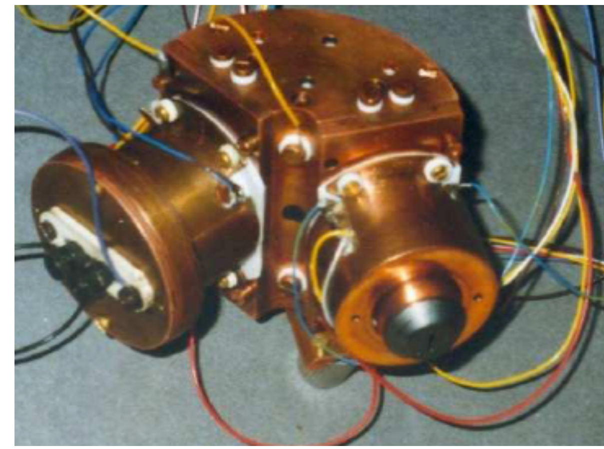
Electronic excitation needs some re-analysis, before its use in plasma modelling

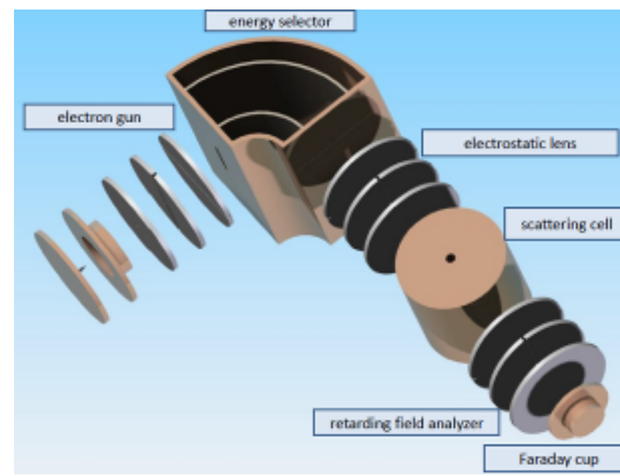
M.Y. Song et al., in preparation

Experimental total: electrostatic analyser

attenuation method $I = I_0 \exp(-\sigma n L)$; precision <5%
(apart from the angular resolution error)





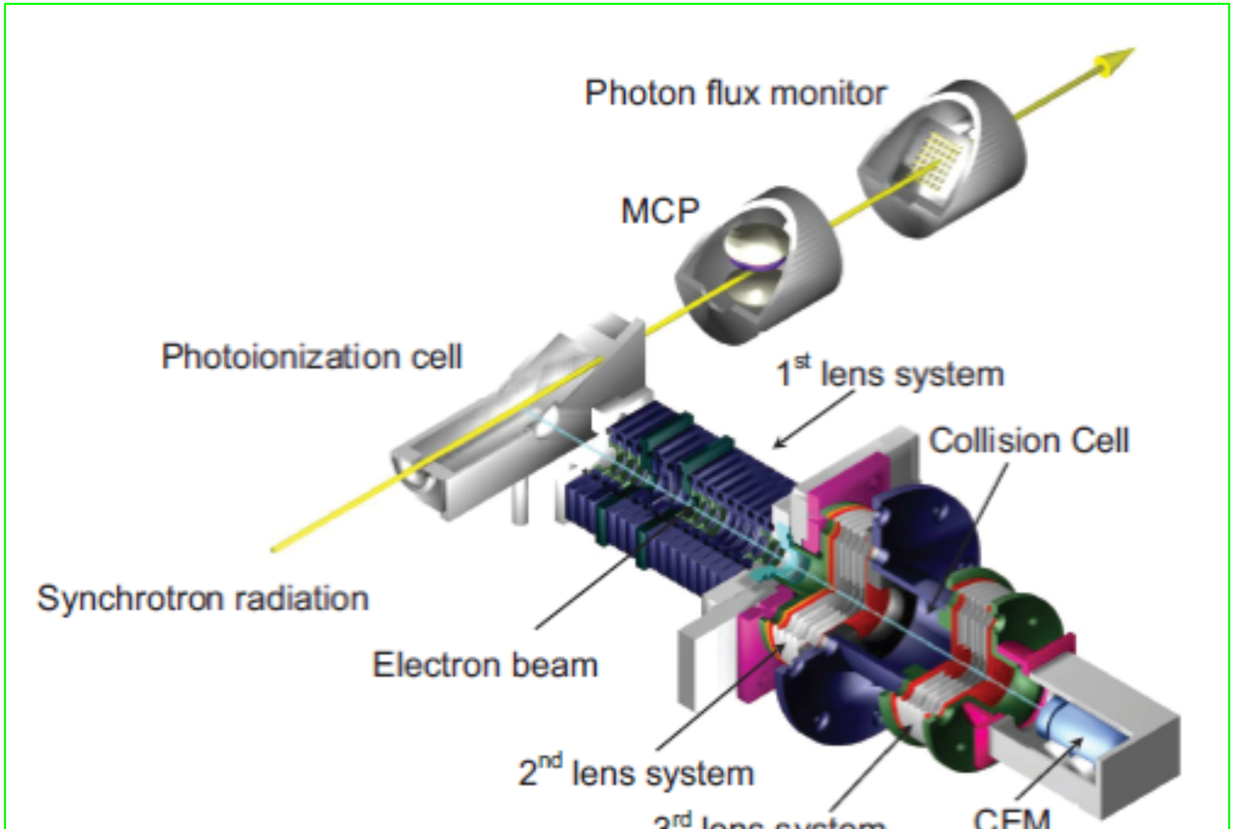


Gdańsk Technical University
Szmytkowski & collaborators > 1982

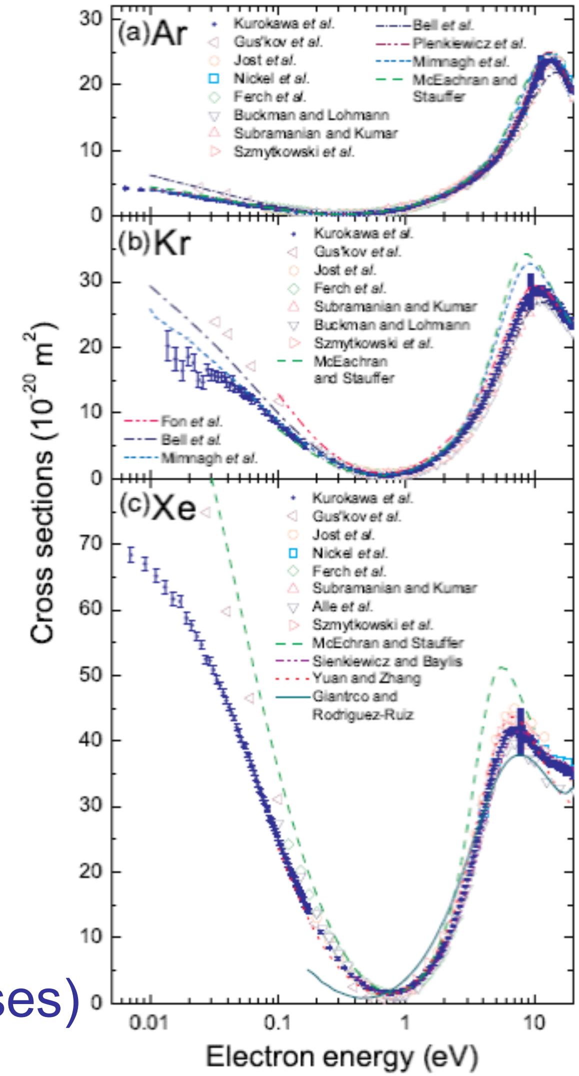
Courtesy: P. Możejko, A. Domaracka, PhD

Cz. Szmytkowski, K. Maciąg, G. Karwasz, Chem. Phys. Lett 107 (1984) 481

Experimental total at E< 1eV



Tokyo Technical University

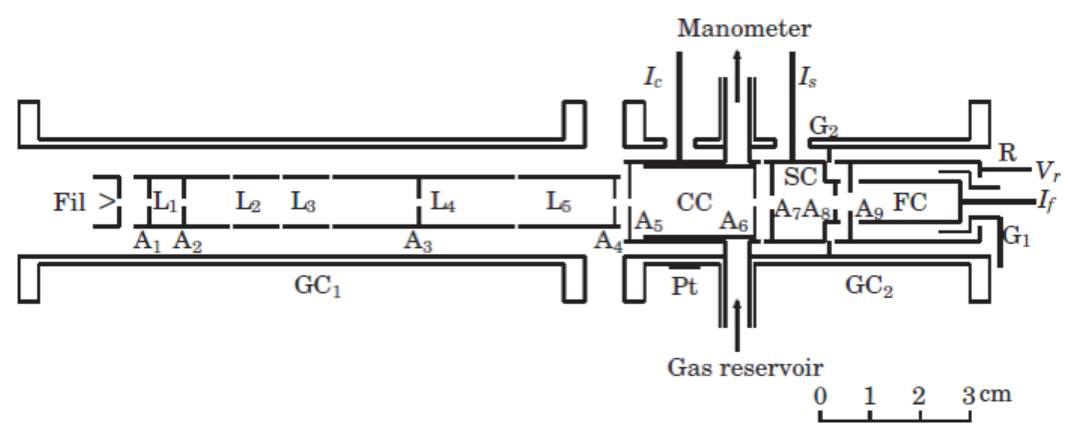


Excellent quality data at very low energies (noble gases)

K. Kitajima et al., Eur. J. Phys. D **63** (2012) 130

Experimental total: magnetic guiding

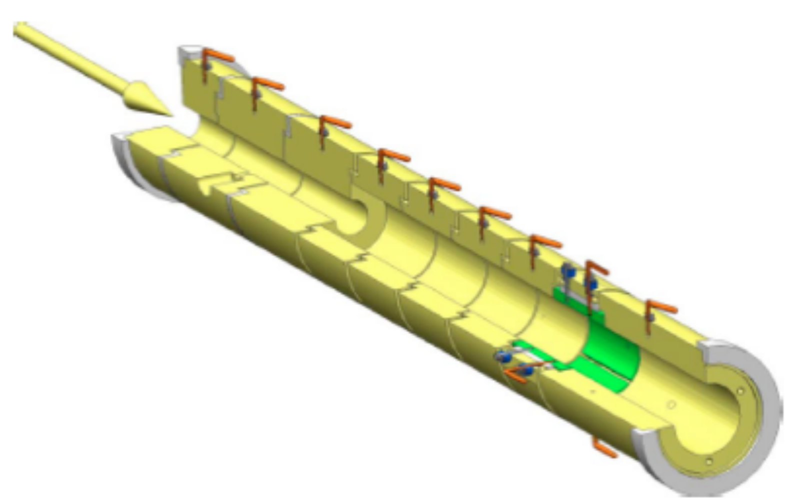
Problem: deconvolution of data vc angle is needed



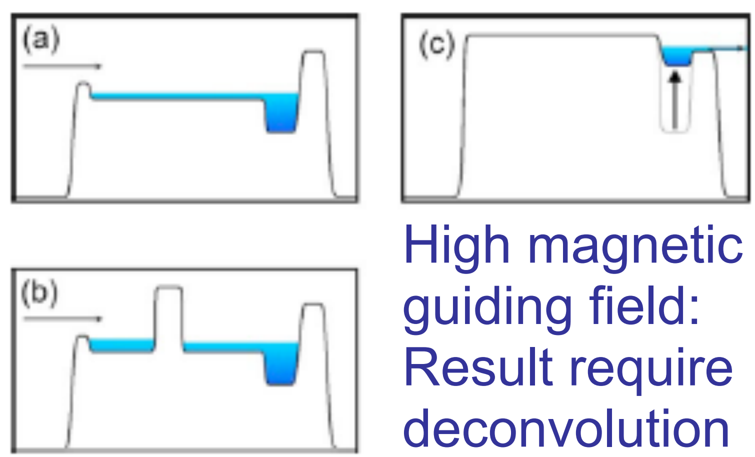
H. Nishimura et al., J.Phys. Soc. Japan **72** (2003) 1080

Linear transmission:
Good quality data
at about 100 eV

113105-2 Sullivan *et al.*



Rev. Sci. Instrum. **79**, 113105 (2008)

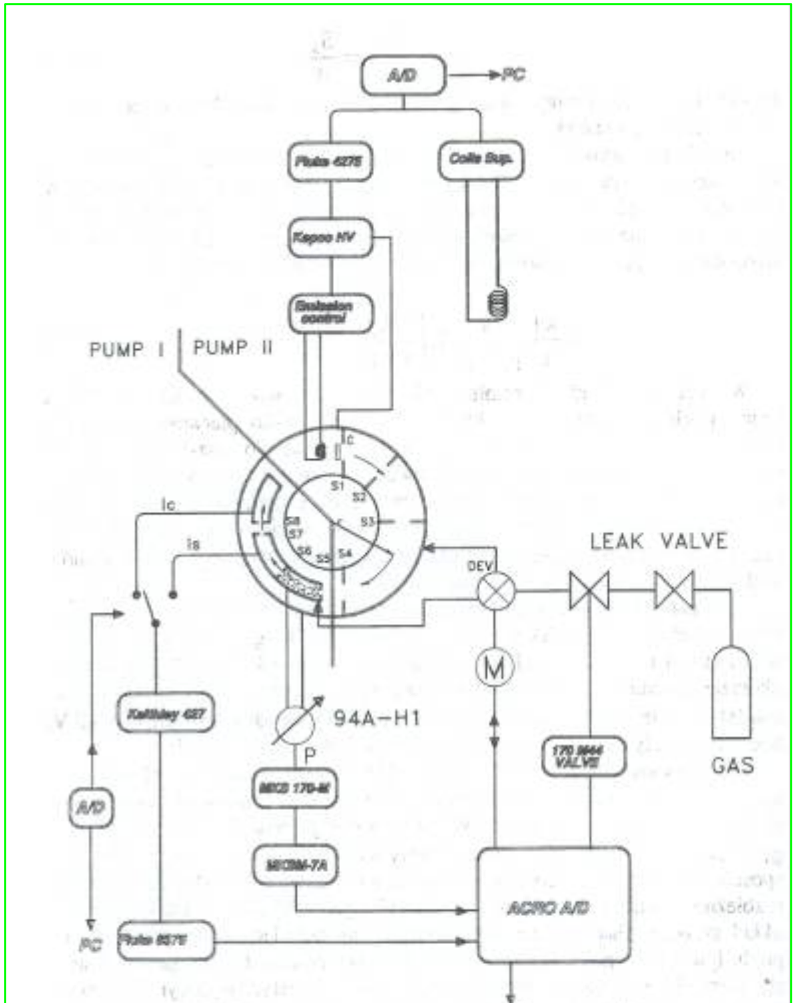


High magnetic
guiding field:
Result require
deconvolution

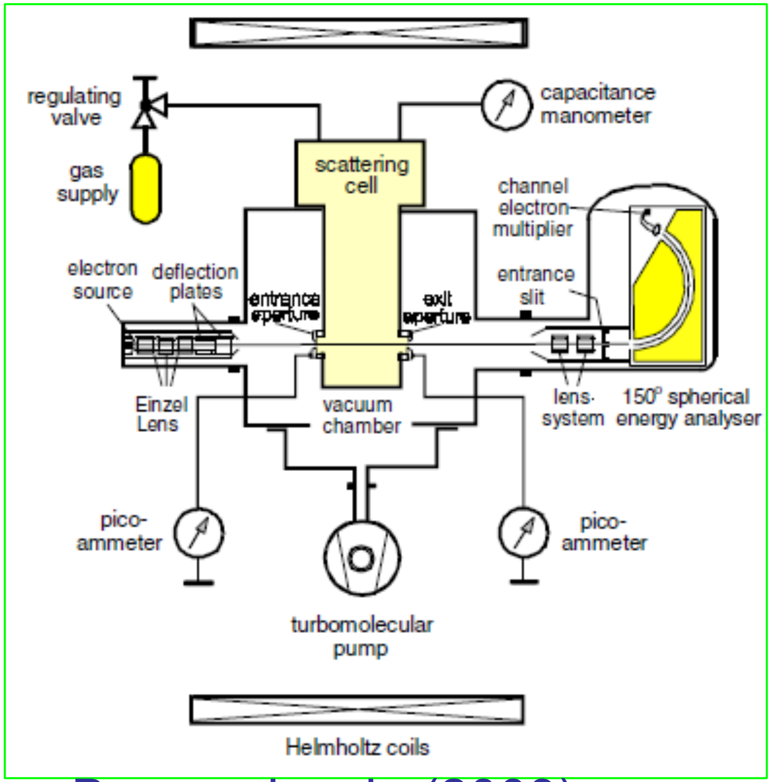
J. P. Sullivan et al., Rev. Sci. Instr. 79 (2008) 113105

High-energy total cross sections

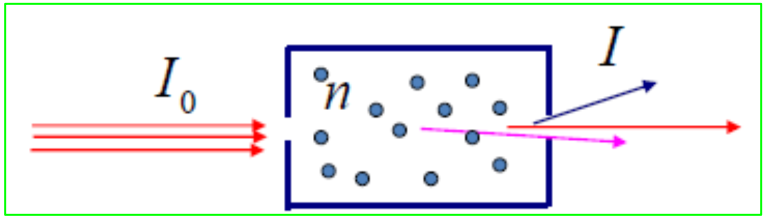
Baek W Y and Grosswendt B, J. Phys. B **36** (2003) 751



Trento (1985-2000)



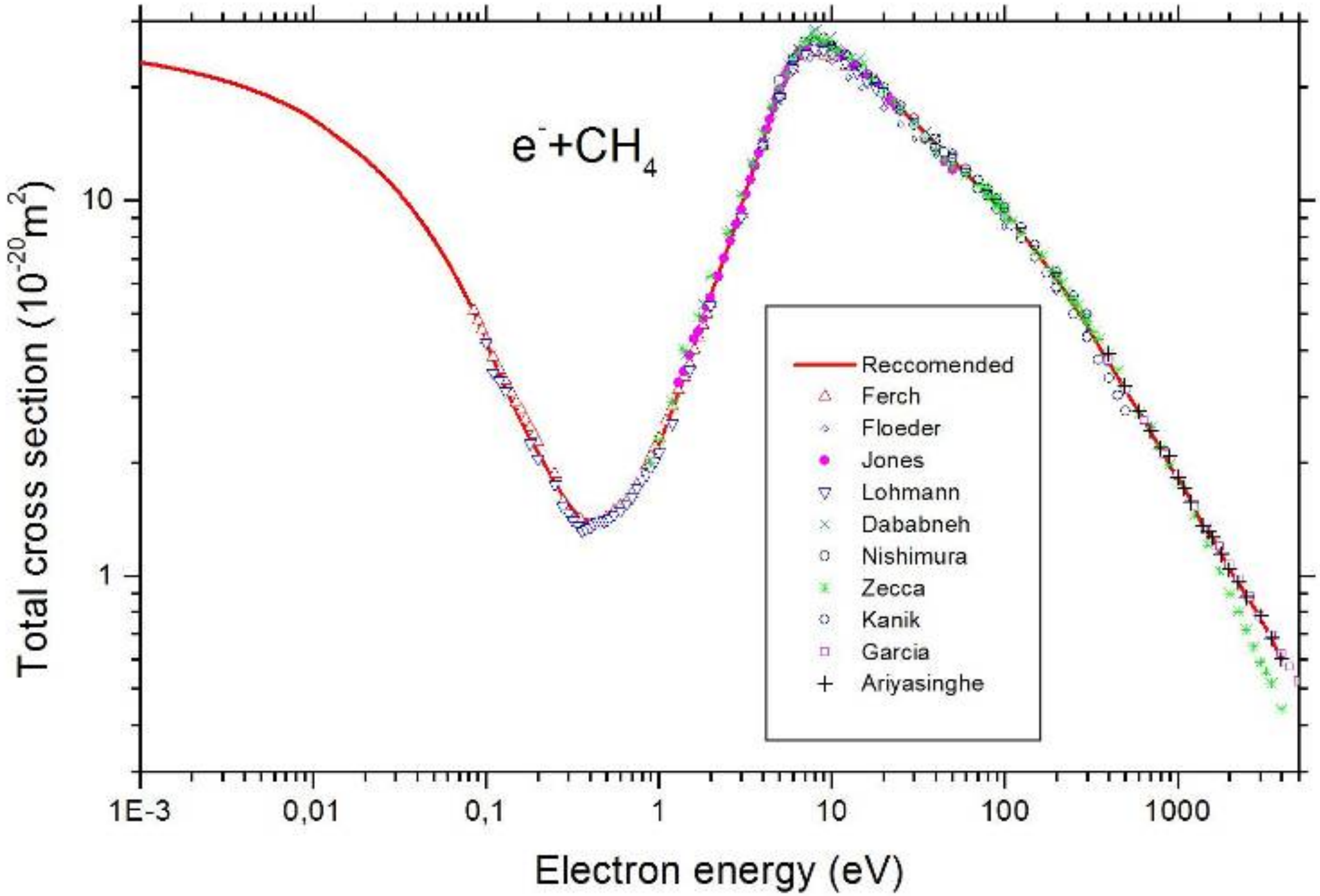
Braunschweig (2003)



Angular resolution error !

Review case study: CH_4

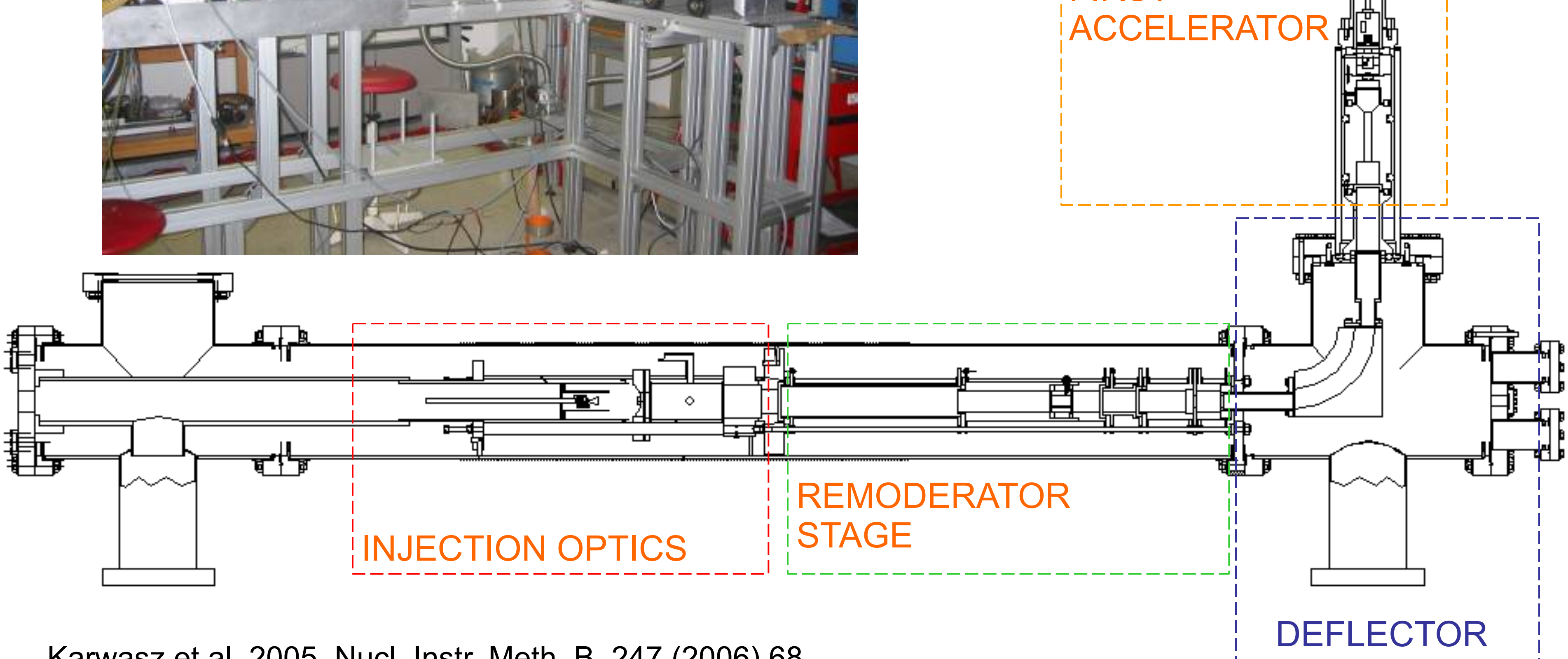
Excellent (±5%) agreement for total CS, but after some semi-empirical refinement at very low and at high energies (see later)



M.-Y. Song, J. S. Yoon, H. Cho, Y. Itikawa, G. Karwasz, V. Kukooulin, Y. Nakamura, J. Tennyson, J. Phys. Chem. Ref. Data, 44 (2015) 023101

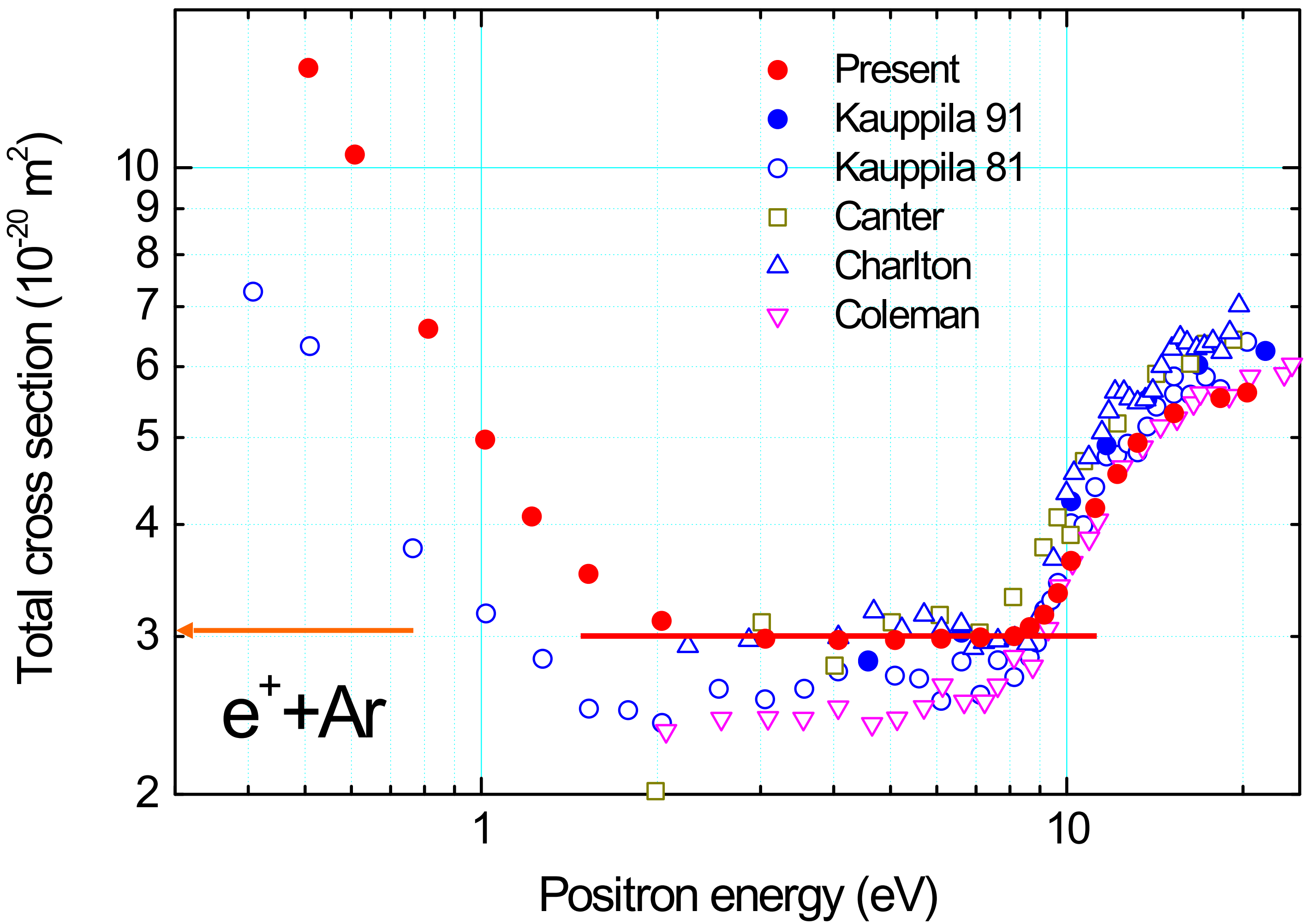
Positron – total cross sections

Easy to be calculated, difficult to measure: a careful experiment may serve as benchmark for theory



Karwasz et al. 2005, Nucl. Instr. Meth. B, 247 (2006) 68

Argon: a flat cross section up to Ps threshold ?

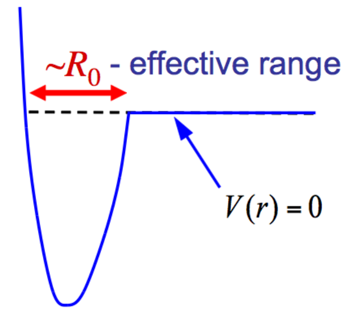


Karwasz, Obergugl, June 2004

Modified effective range theory

Standard effective-range expansion for finite range potential

$$k \cot \delta_0(k) = -\frac{1}{a} + \frac{1}{2} R_0 k^2 + 0(k^4)$$



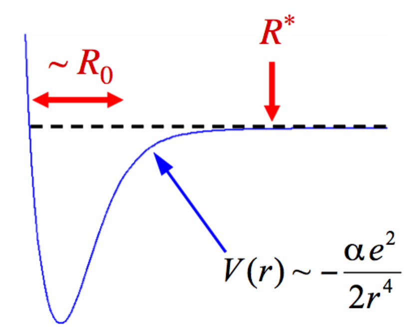
Modified effective range expansion (MERT) for r^{-4} potential

$$\tan \delta_l = \frac{s^2 - \tan \eta^2 + B_l \tan \eta (s^2 - 1)}{\tan \eta (1 - s^2) + B_l (1 - s^2 \tan^2 \eta)}$$

O'Malley, Spruch, Rosenberg, Journal of Mathematical Physics 2, 4 (1961).

$$B_l = \tan(\phi_l + l\tfrac{\pi}{2})$$

η, s – parameters given by analytic solutions (Mathieu functions)



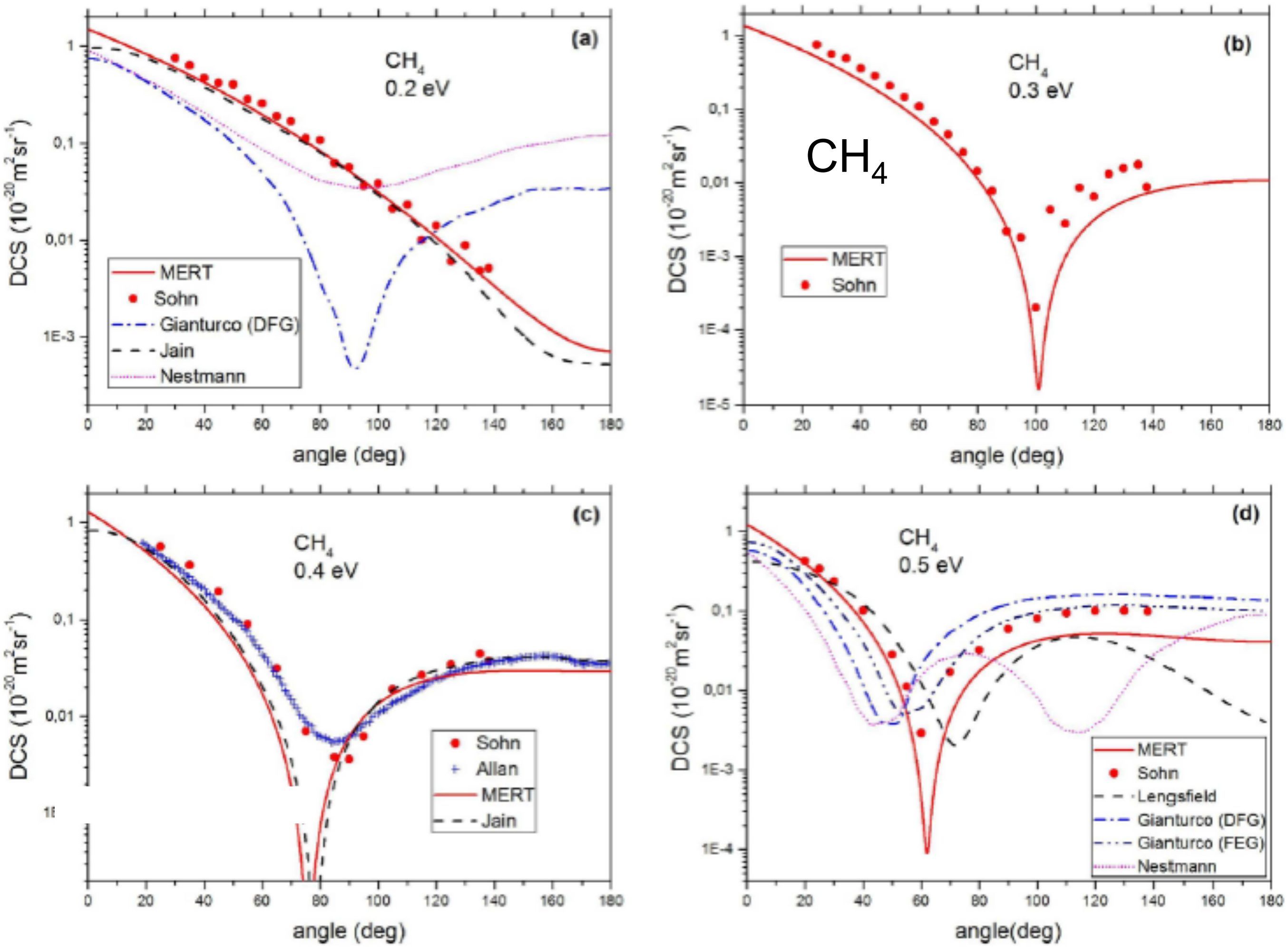
Contribution from the short-range potential

$$B_l(k) = B_l(0) + \frac{1}{2} R^* R_0 k^2 + 0(k^4)$$

R_0 – effective range

Z. Idziaszek, G Karwasz, Phys. Rev. A 73, 064701 (2006)

Very low energies: Modified Effective Range Theory



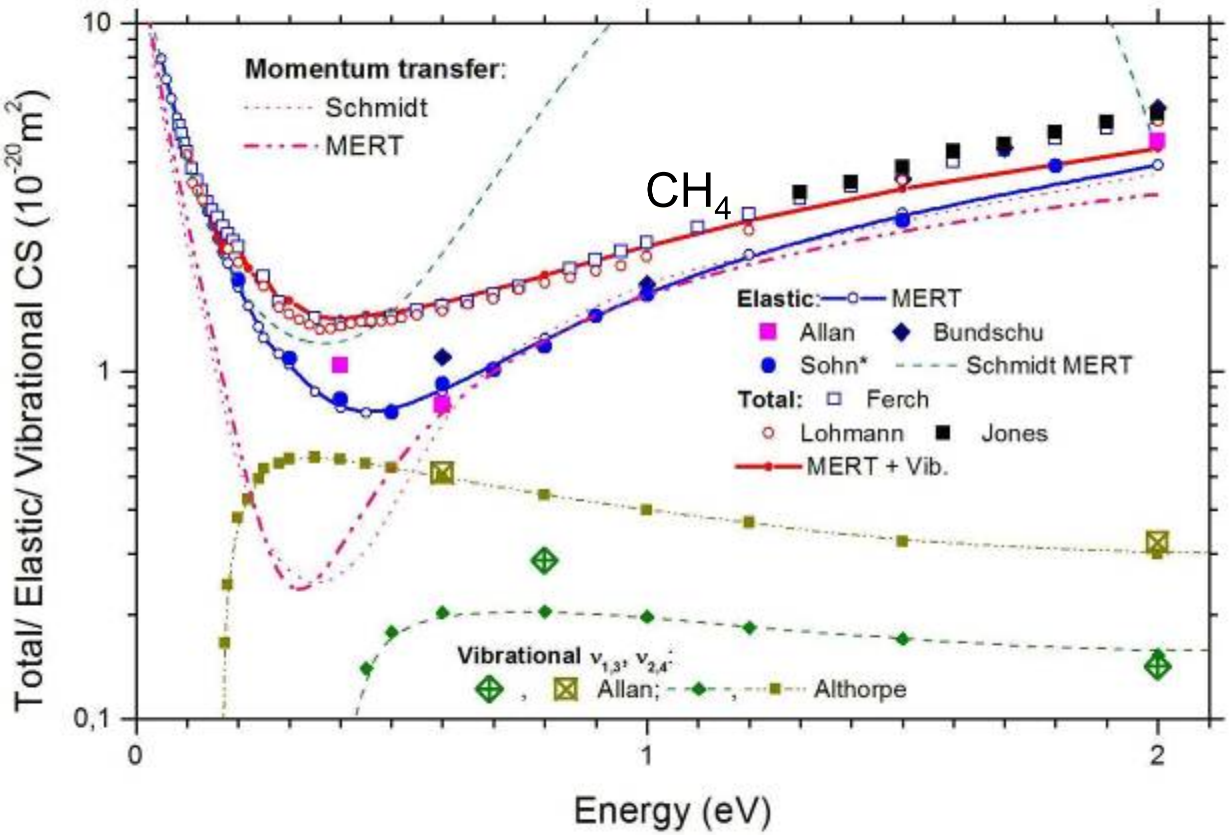
Two fitting parameters:
A – scattering length
R – effective range
(for *s* and *p* partial waves)

Analysis of DCSs allows check of consistency for other results

K. Fedus, G. Karwasz, Eur. J. Phys. D **68**: 93 (2014),

Semi-empirical methods: elastic (MERT)

Link between elastic, total, MTCS: for spherical targets at low energies



MERT links elastic, total, MTCS:

for atoms, spherical molecules @ energies < few eV

K. Fedus, G. Karwasz, Eur. J. Phys. D 68 (2014) 93

Positron total in H_2: experiment vs theory

KAMIL FEDUS, JAN FRANZ, AND GRZEGORZ P. KARWASZ

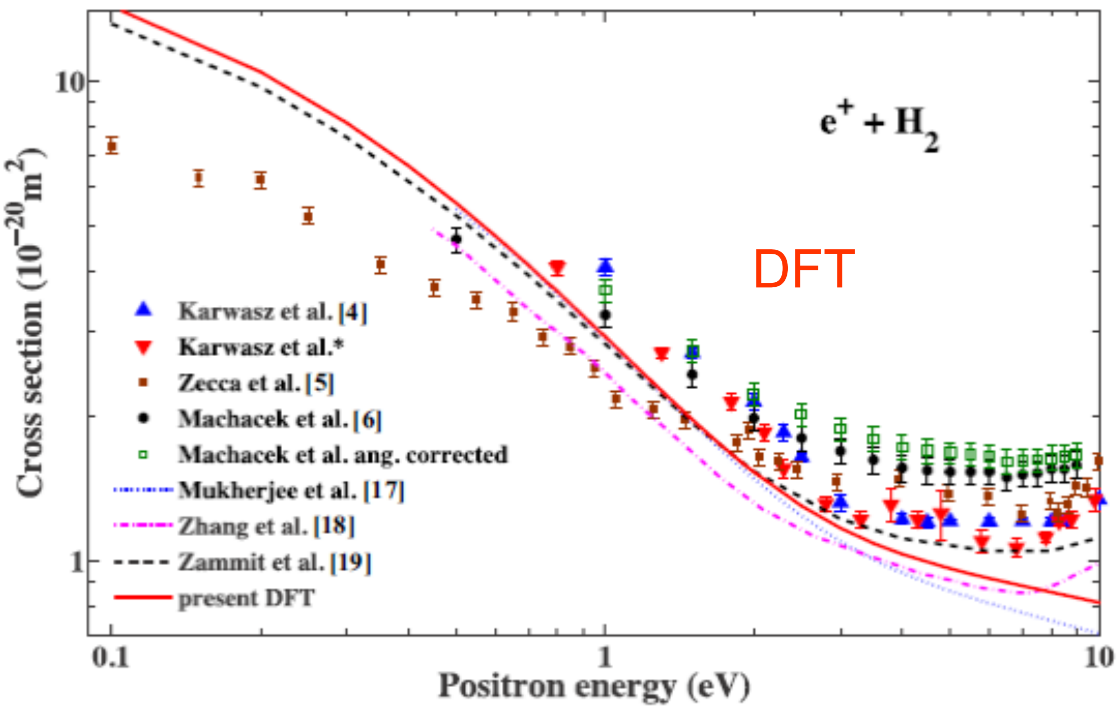
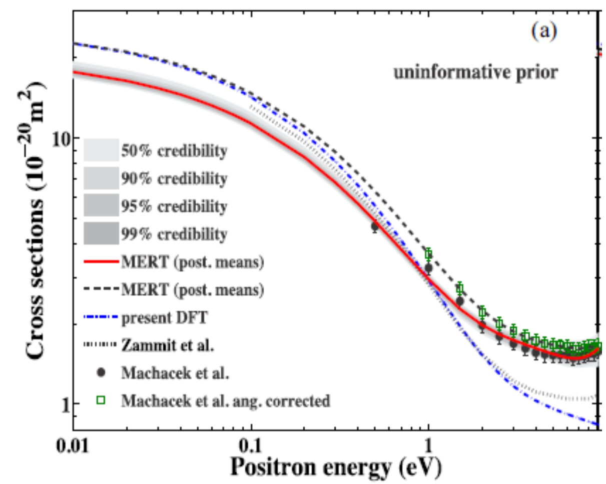
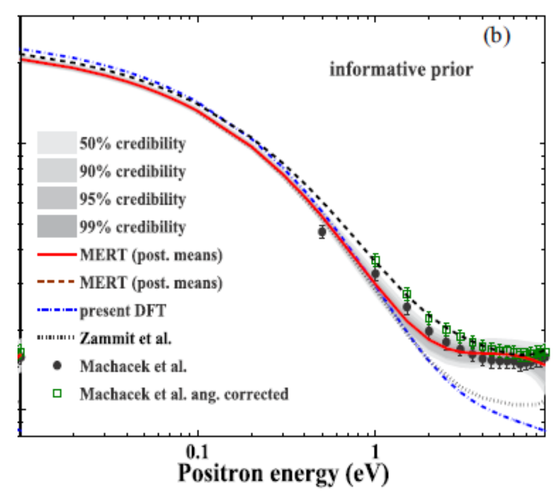


FIG. 1. (Color online) Total cross sections for positron scattering from the ground state of H_2. Present DFT results are compared with the calculations of Mukherjee *et al.* [17], Zhang *et al.* [18], Zammit *et al.* [19], and experiments of Karwasz *et al.* published in 2006 [4] and the same set presently reanalyzed (Karwasz *et al.**), Zecca *et al.* [5] and Machacek *et al.* [6] (directly measured and corrected).





PRA 91 (2015) 062701

Do positrons measure atomic radii?

Eur. Phys. J. D (2016) 70: 155
DOI: 10.1140/epjd/e2016-70100-3

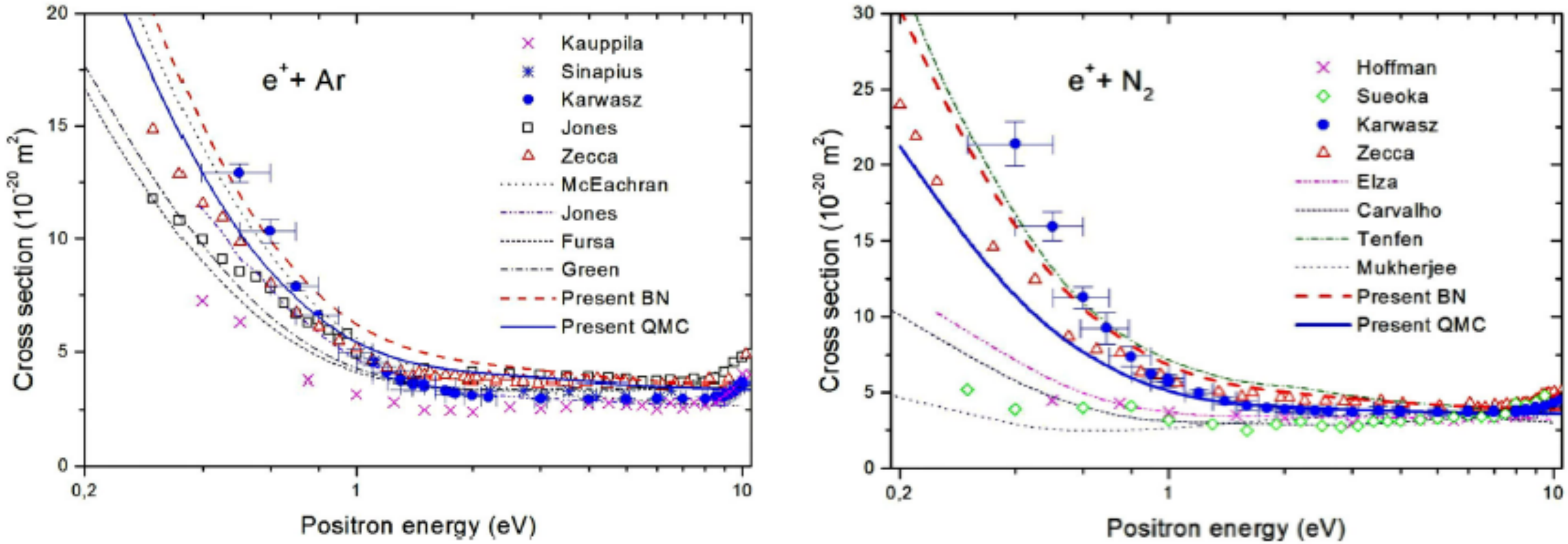
THE EUROPEAN PHYSICAL JOURNAL D

Regular Article

Do positrons measure atomic and molecular diameters?*

Jan Franz[1], Kamil Fedus[2,a], and Grzegorz P. Karwasz[2]

DFT calculations



Quantum mechanics explains the classical-like result

Eur. Phys. J. D (2016) 70: 261
DOI: 10.1140/epjd/e2016-70452-6

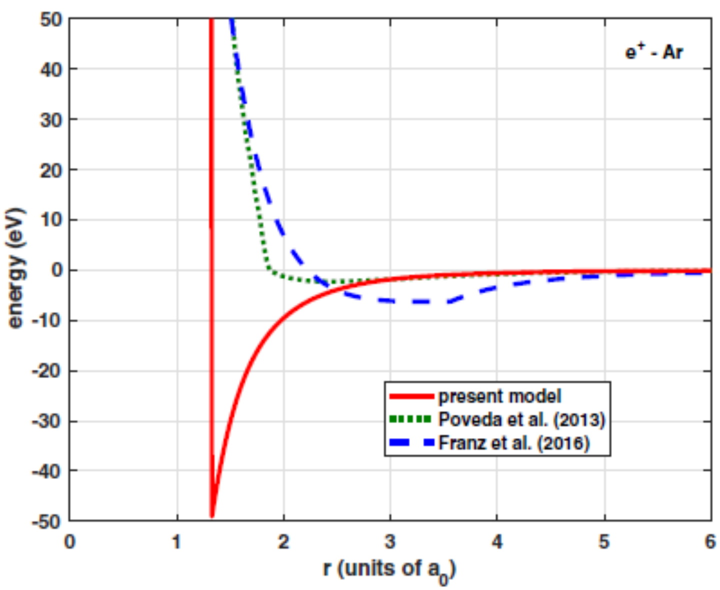
THE EUROPEAN PHYSICAL JOURNAL D

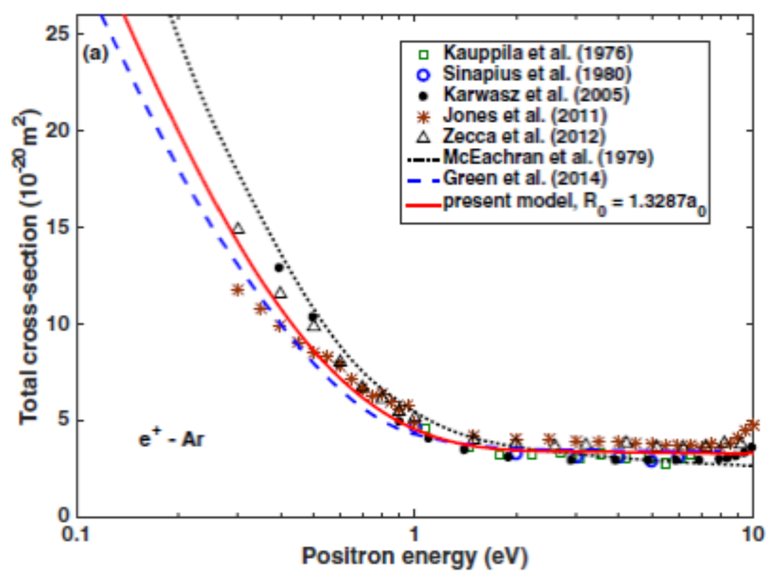
Regular Article

But this is still only a model, not mechanism: not hard, but sticky ball

A rigid sphere approach to positron elastic scattering by noble gases, molecular hydrogen, nitrogen and methane

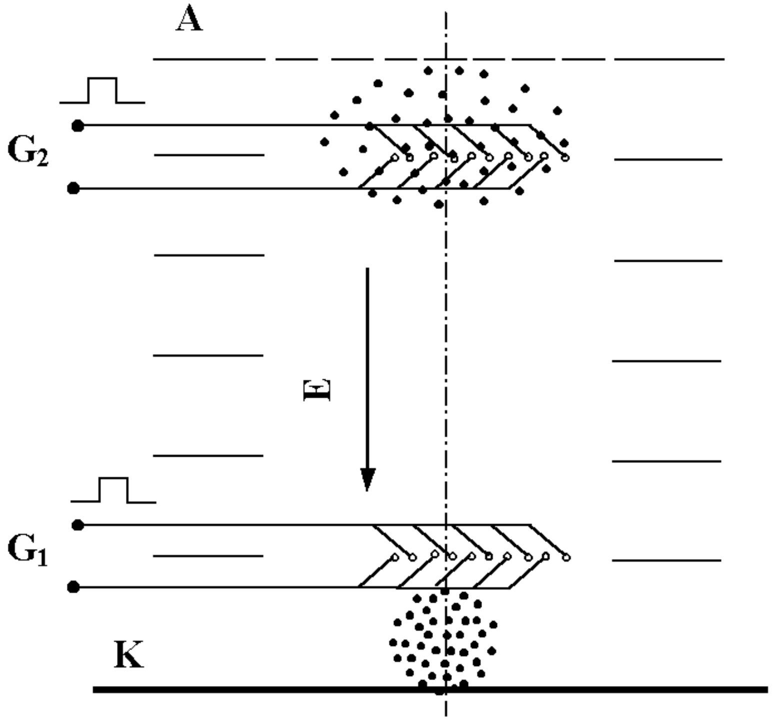
Kamil Fedus[a]





Swarm experiments: diffusion coefficients → cross sections

$$\frac{\partial}{\partial t} n_e(\mathbf{r},t) = -w \frac{\partial}{\partial z} n_e(\mathbf{r},t) + D_T \left[\frac{\partial^2}{\partial x^2} n_e(\mathbf{r},t) + \frac{\partial^2}{\partial y^2} n_e(\mathbf{r},t) \right] + D_L \frac{\partial^2}{\partial z^2} n_e(\mathbf{r},t)$$



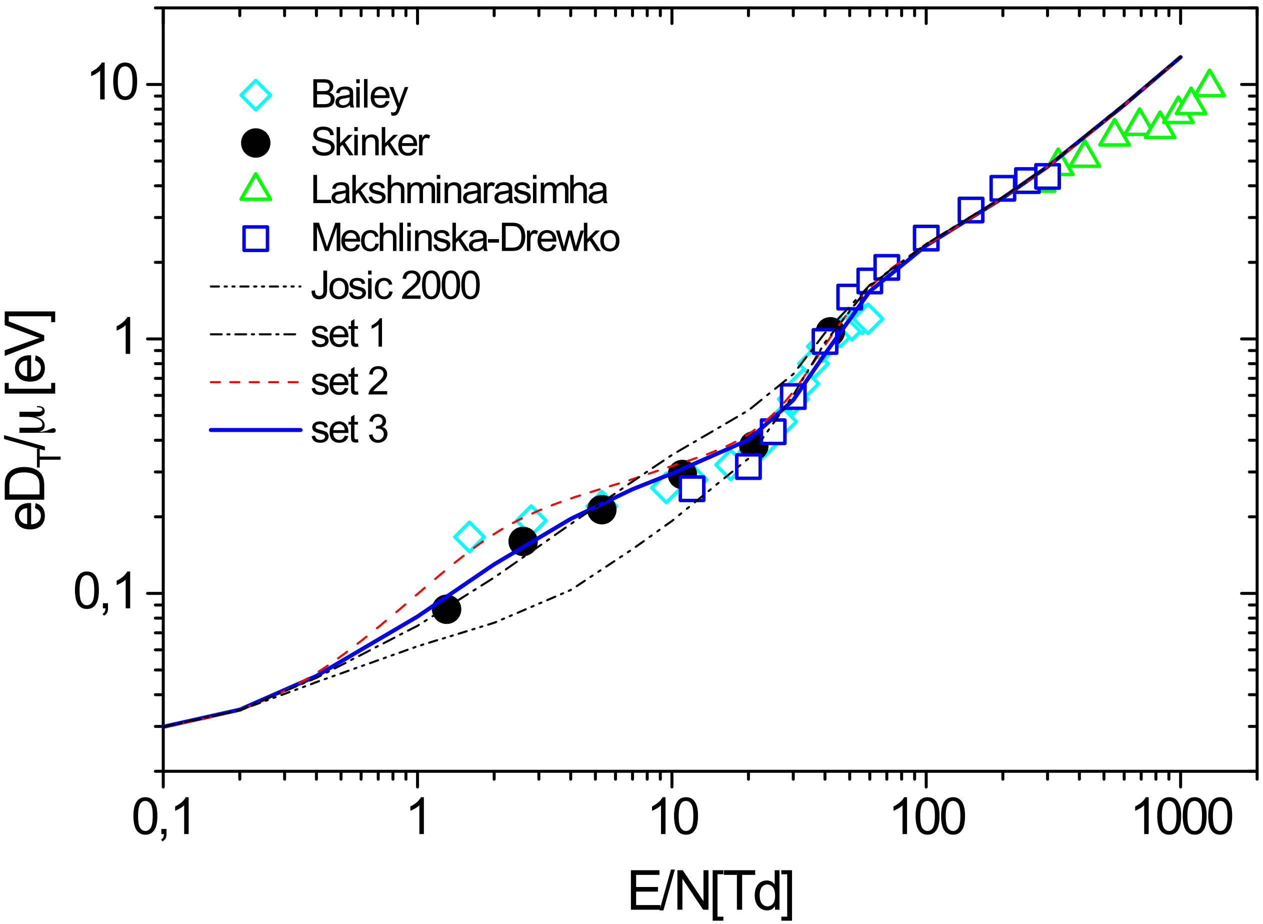
$$w = -\left(\frac{2}{m}\right)^{1/2} \frac{eF}{3N} \int_0^\infty \frac{E}{\sigma_m(E)} \frac{df_0(E)}{dE} dE$$

$$D_T = \left(\frac{2}{m}\right)^{1/2} \frac{1}{3N} \int_0^\infty \frac{E}{\sigma_m(E)} f_0(E) dE$$

Experiment is simple, but requires fitting cross sections, that can form a non-unique set

W. Roznerski, K. Leja, J. Mechlińska-Drewko (Gdańsk Technical University)

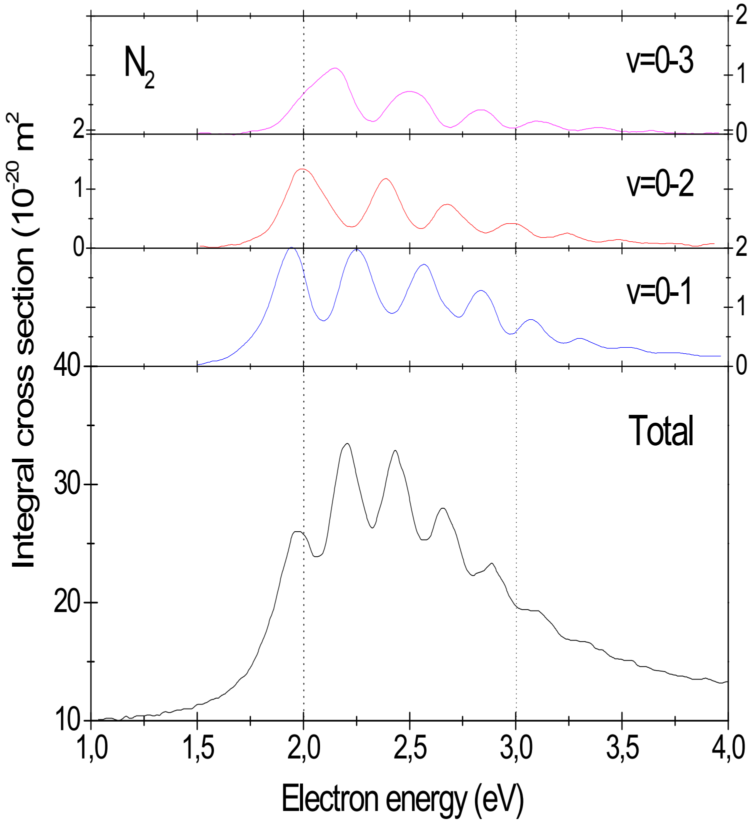
Analogy: NO swarm data

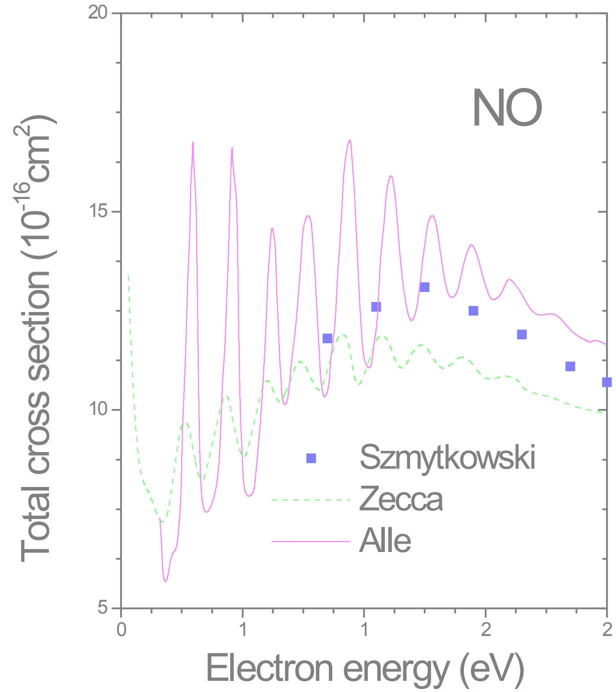


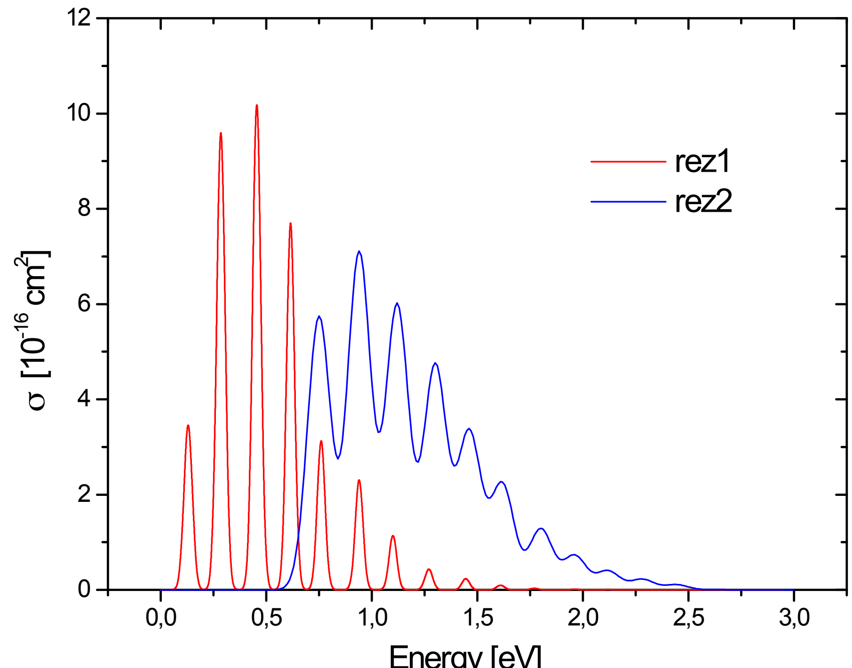
L. Josić, T. Wróblewski, Z. Lj. Petrović, J. Mechlińska- Drewko and G.P. Karwasz, Chem. Phys. Lett. 350 (2001) 318

NO resonances – search for analogies

N_2 and O_2 – like

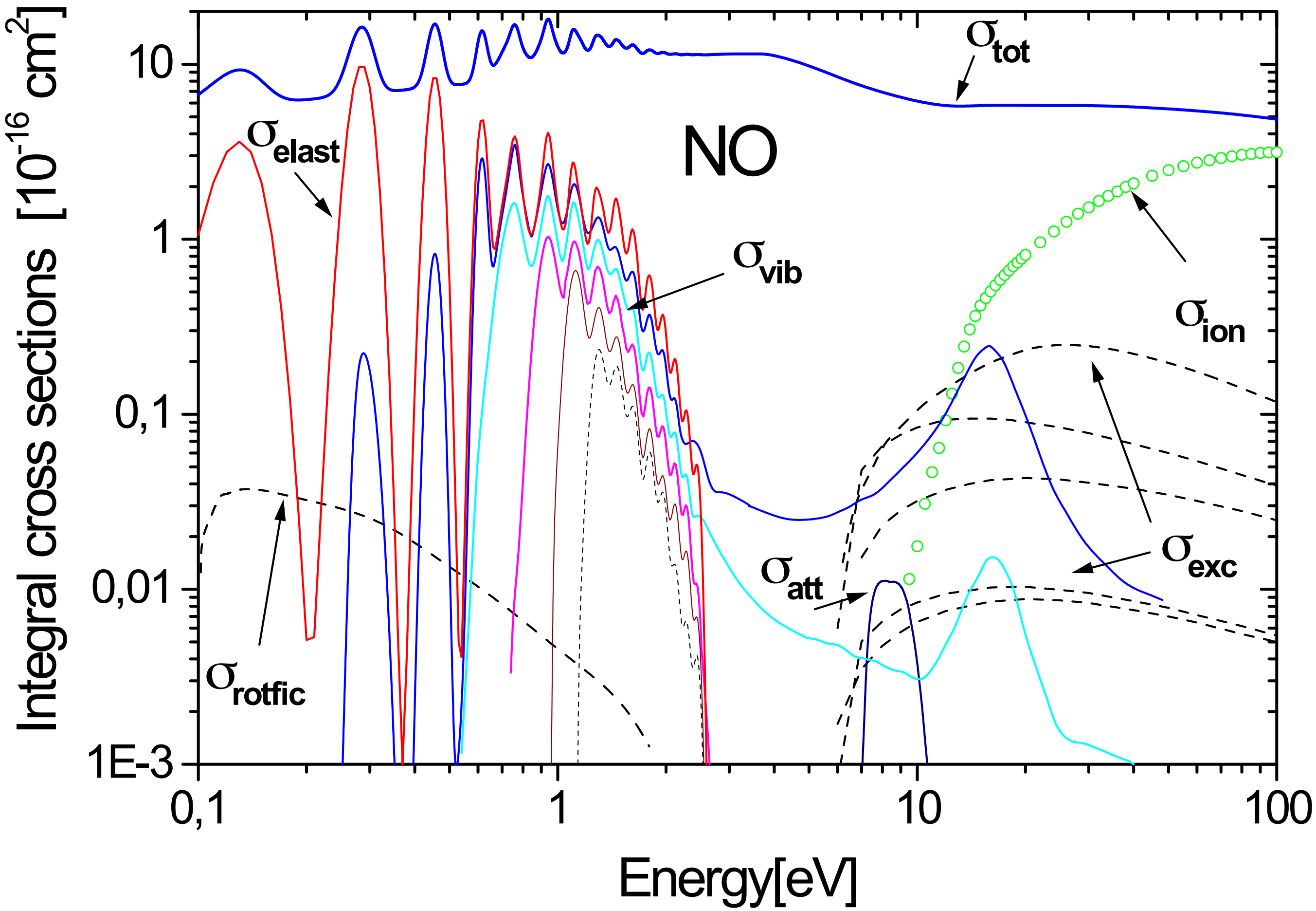






Josic et al. Chem. Phys. Lett. (2003)

NO – a congruent set of cross sections



Confirmed by beam experiments (ANU Canberra, Fribourg Uni)

Resonances: theory allows more precise prediction

Plasma Sources Sci. Technol. **21** (2012) 055018 V Laporta *et al*

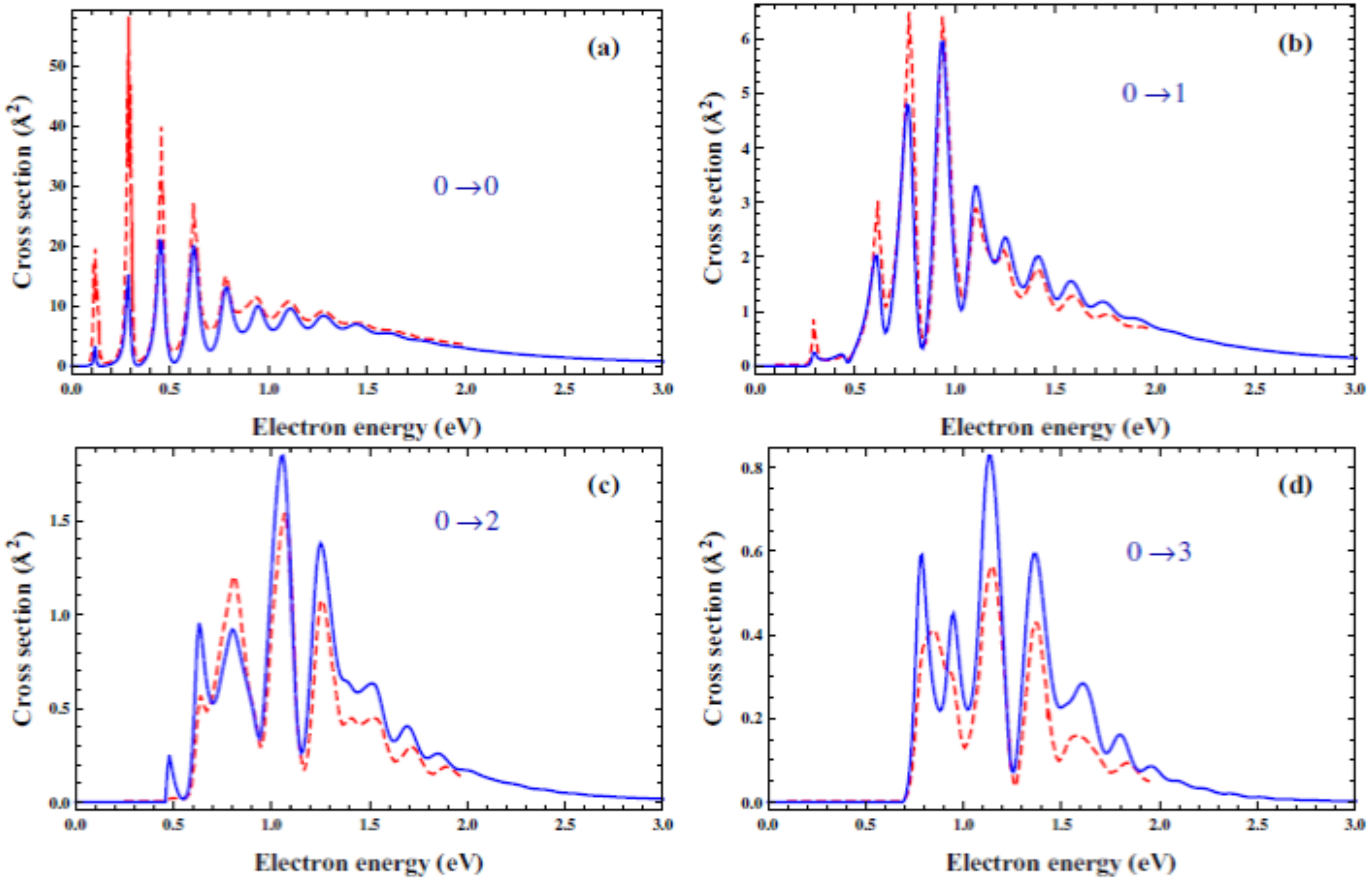
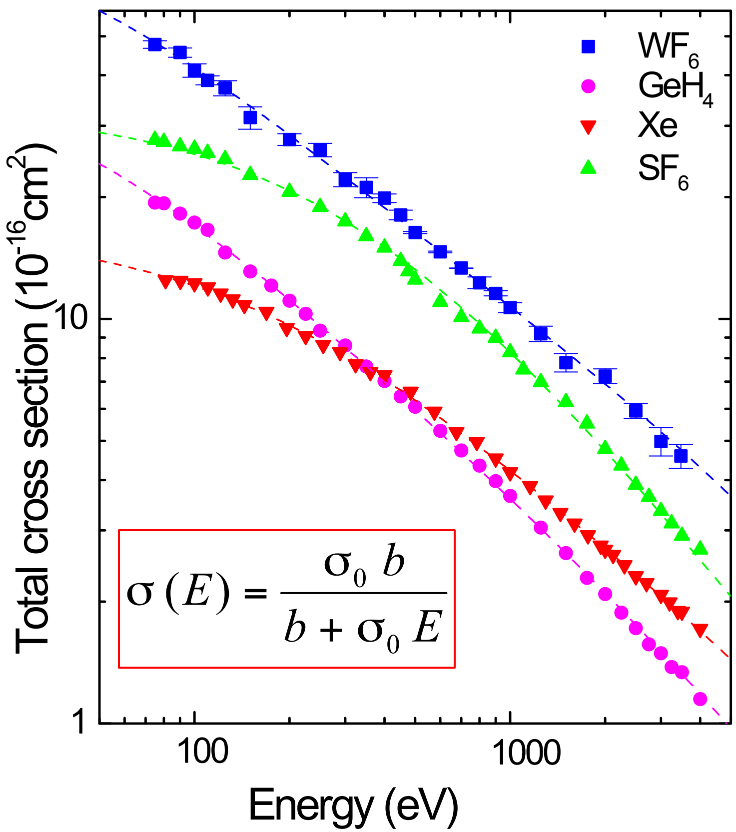


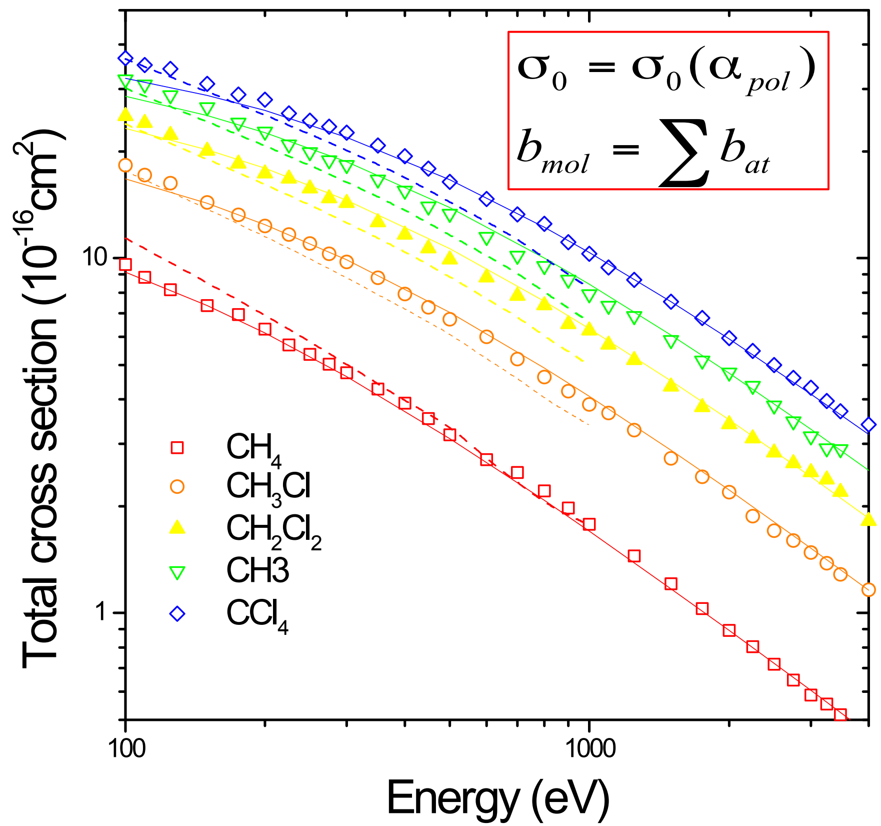
Figure 7. Electron–NO comparison of the present RVE calculations (full lines) with those of Trevisan *et al* [18] (dashed lines) for the vibrational transitions $0 \rightarrow 0, 1, 2, 3$.

J. Tennyson & collaborators (2012)

High energies total: in search for additivity rule

CH_4, CF_4, SiH_4, ... WF_6 → CH_2F_2, SiF_4 ...
→ H, C, Si, ... W

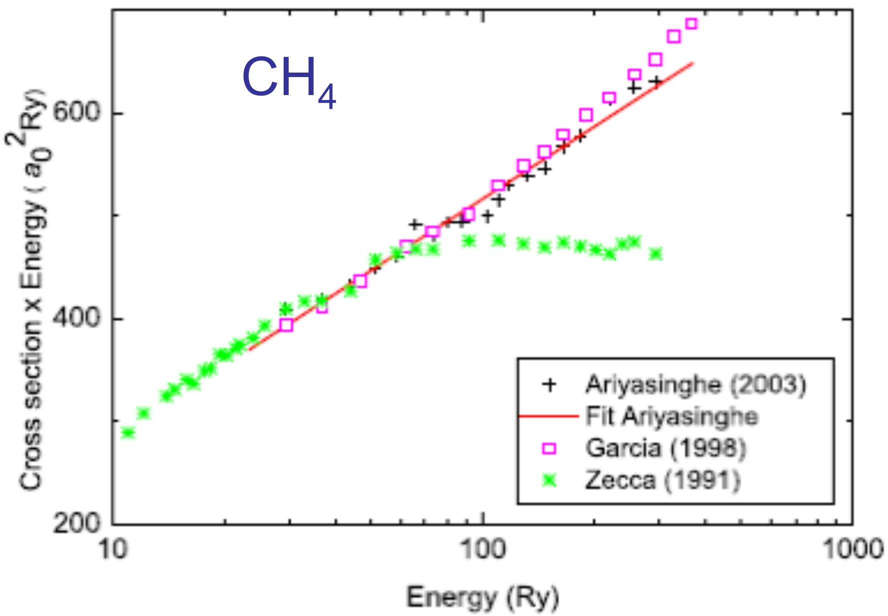


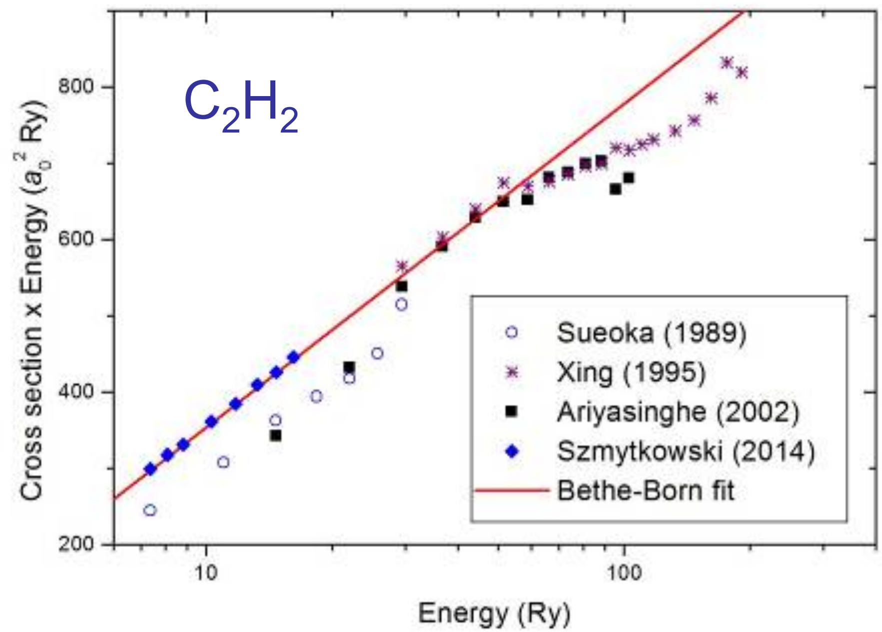


G. Karwasz et al., Phys. Rev. A **59** (1999) 1341

High energy limit (Born-Bethe plot)

$\sigma(E) = A / E + B \log(E)/E$

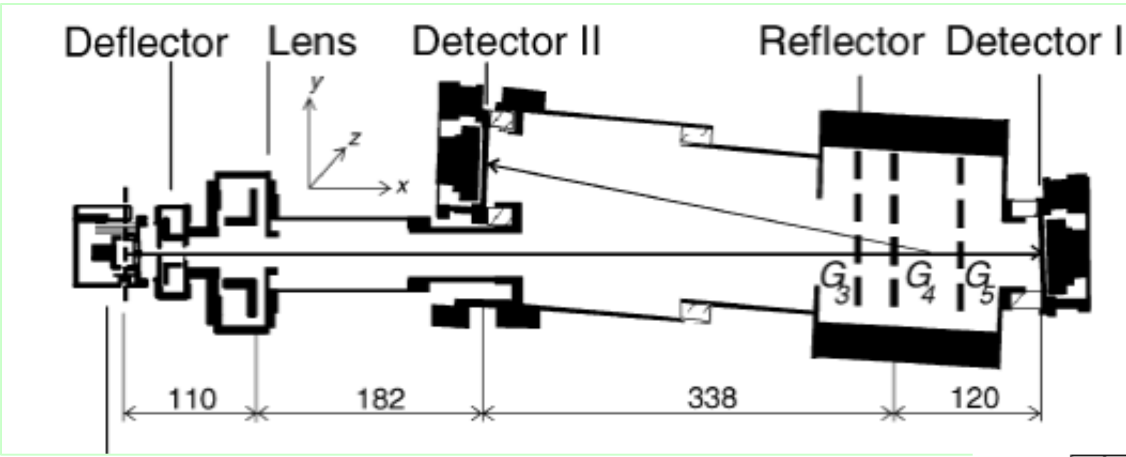




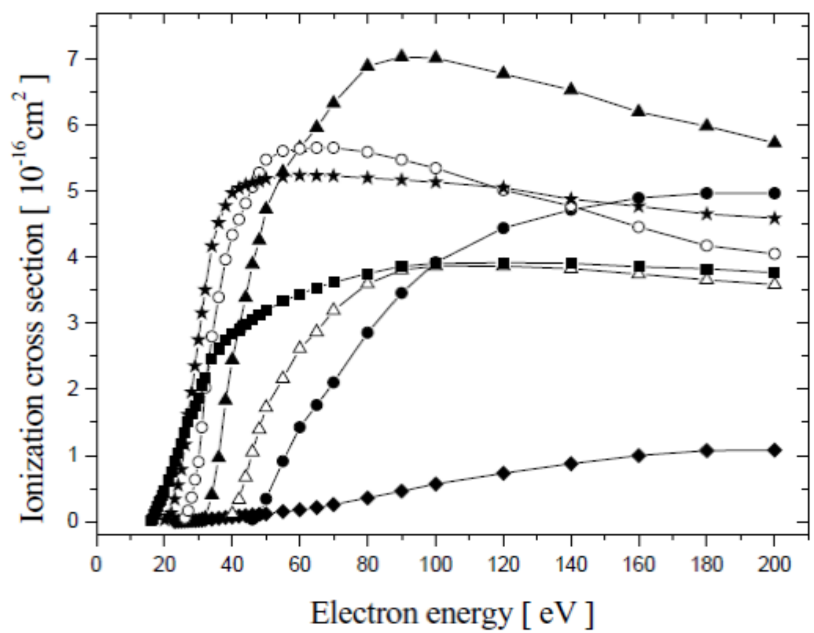
1. Trento experiment was underestimated in high-energy limit (>1000 eV)
2. We still have no idea, how do parameters link to other molecular features

Mi-Young Song et al., JPCRD (2015) & (2017)

Experimental methods: ionization

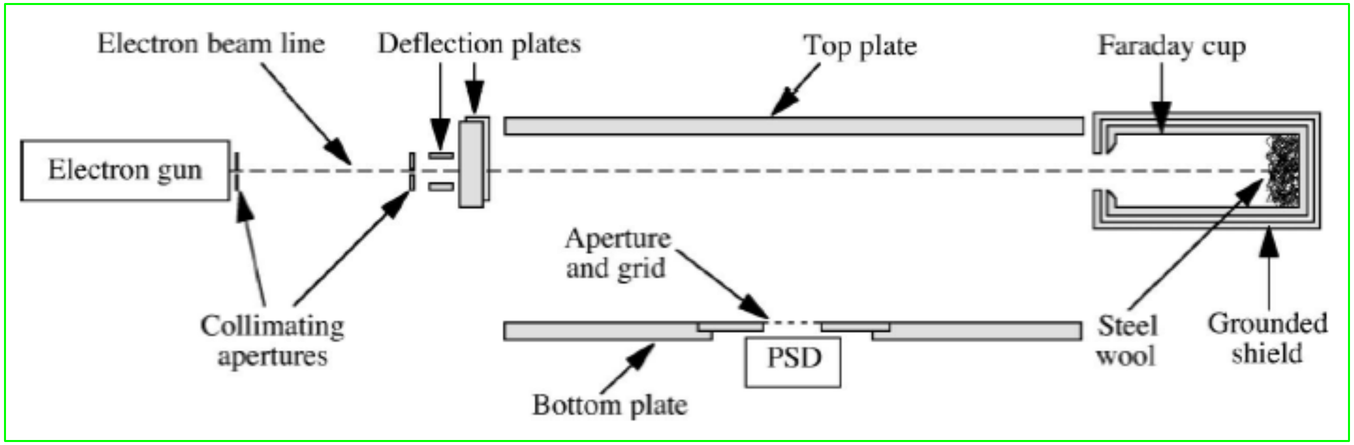


If two ions are formed
(for example $CH_4 \rightarrow CH_3^+ + H^+ + 2e$)
the ionization is counted twice

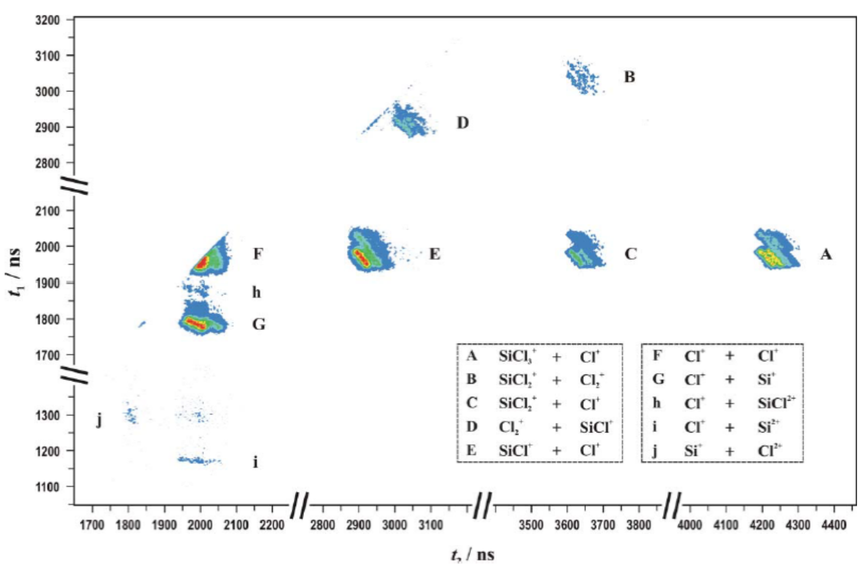


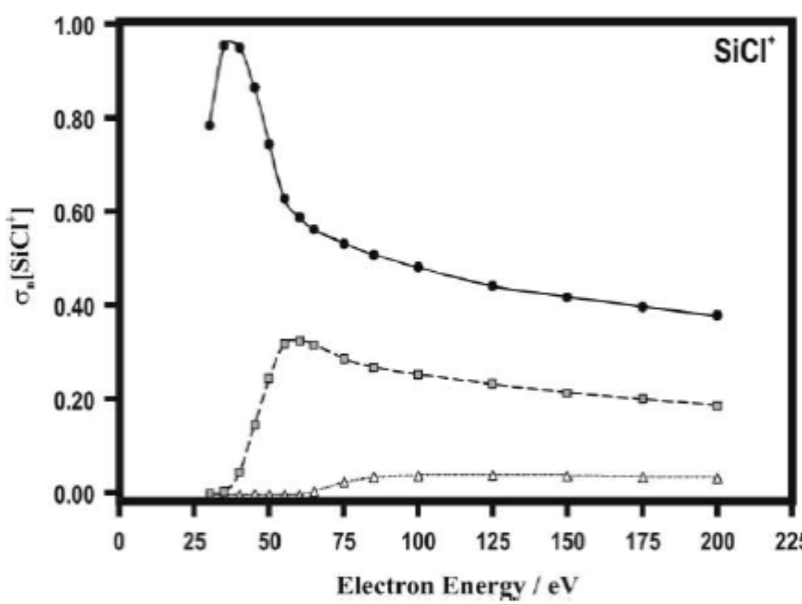
R. Basner, M. Schmidt, K. Becker, Int. J. Mass Spectr. 233 (2004) 25

Experimental methods: ionization (2)



SiCl$^+$ from single, double, triple ionization of $SiCl_4$

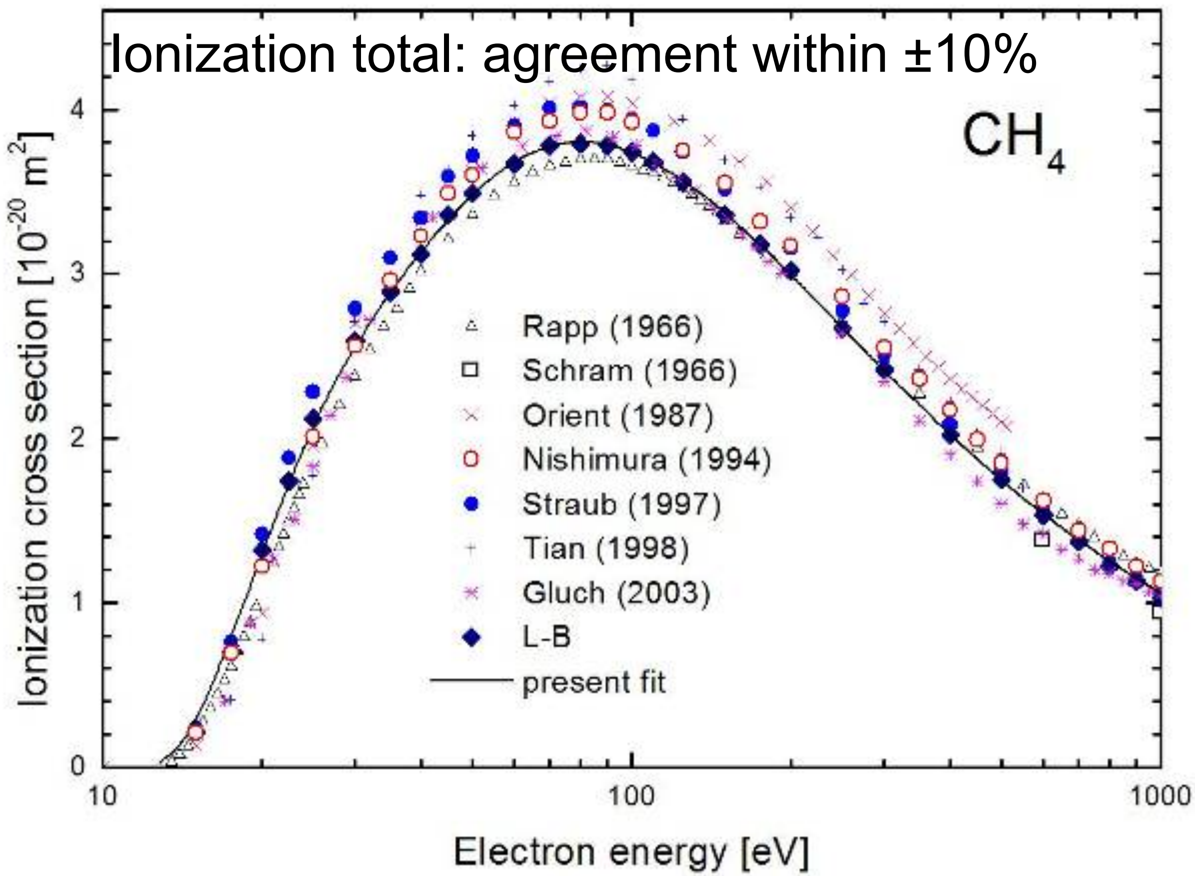




B. G. Lindsay et al., JCP **129** (2004), S J King nad S D Price, JCP**134** (2011) 074311

Ionization (excellent!): CH_4

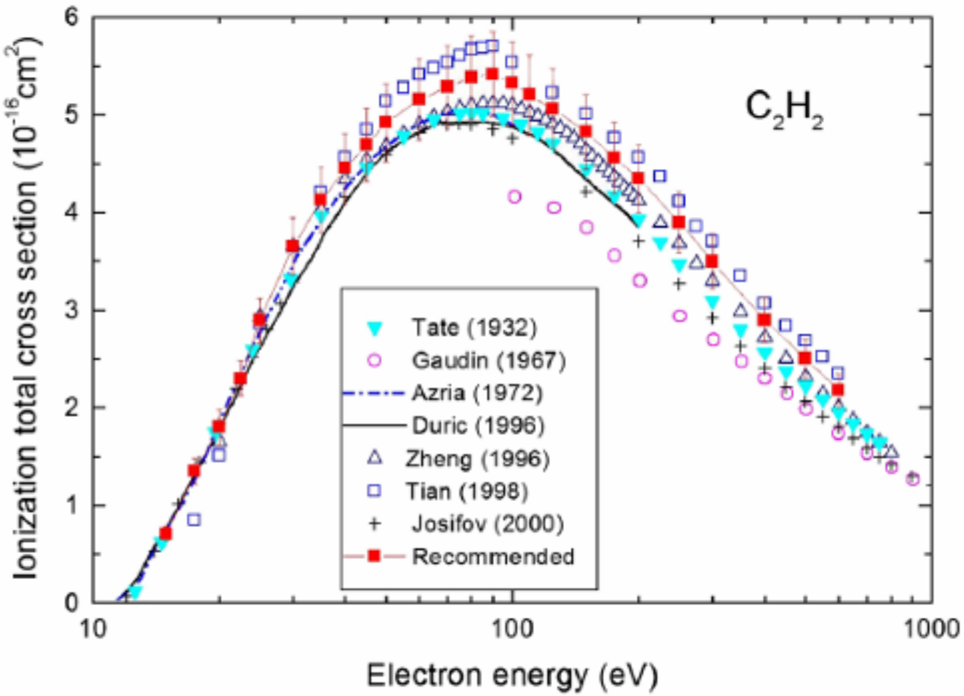
Semi-empirical (BEB model) allows to find-out error in experiments

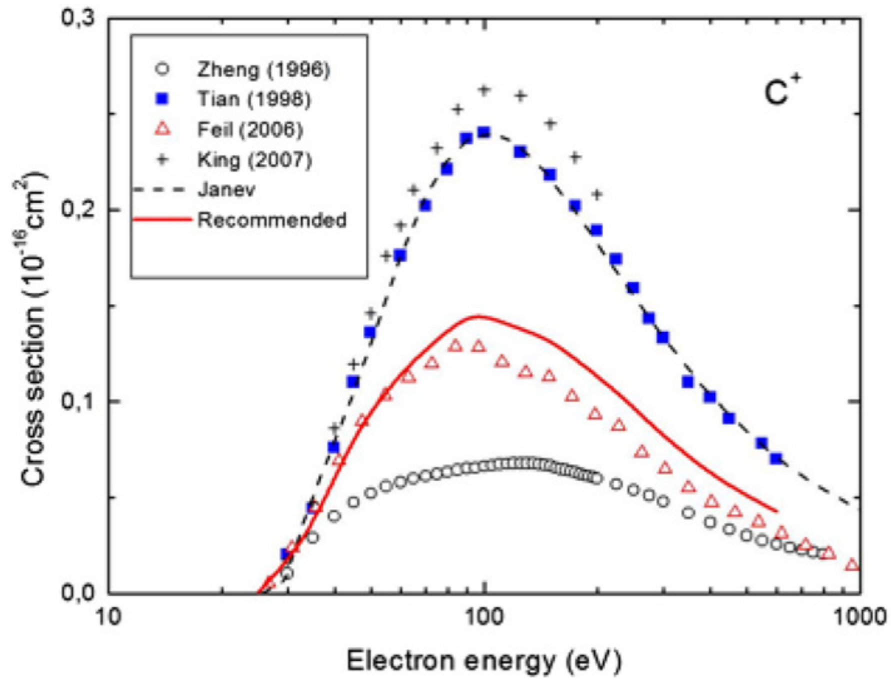


M.-Y. Song, J. S. Yoon, H. Cho, Y. Itikawa, G. Karwasz, V. Kukooulin, Y. Nakamura, J. Tennyson,

Ionization (fairly good): C_2H_2

We do not know channels via which the fragmentation occurs: Quantum dynamics is needed

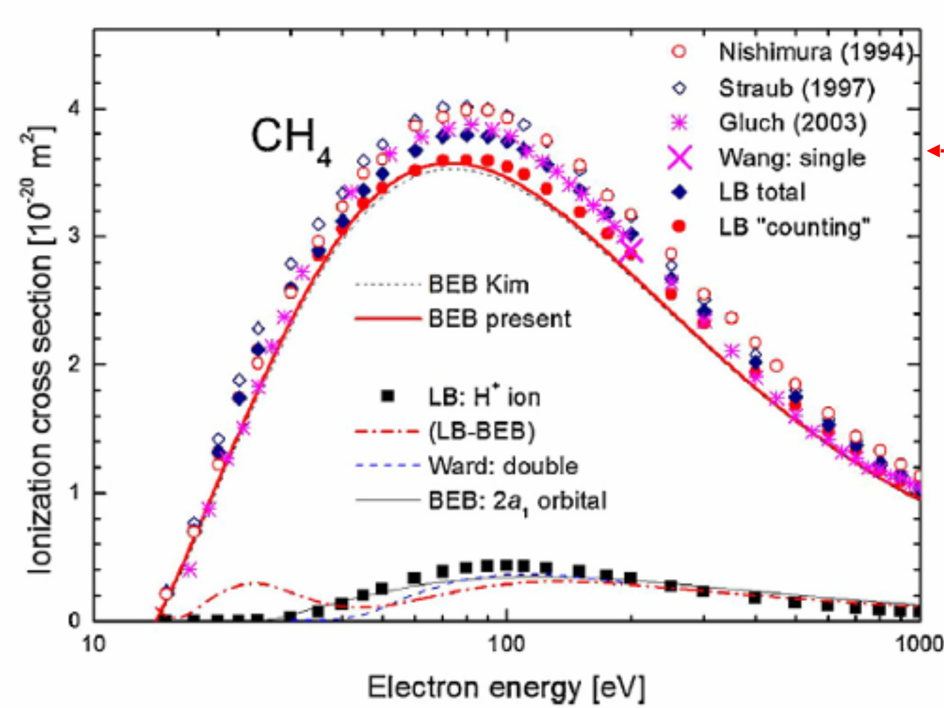




Poor agreement for C^+ production from $e^- + CH_4$ process:King normalized to Tian

M.-Y. Song, J. S. Yoon, H. Cho, Y. Itikawa, G. Karwasz, V. Kukooulin, Y. Nakamura, J. Tennyson, J. Phys. Chem. Ref. Data 46 (2017), 013106

Ionization: BEB formula – check of experiments & prediction



Experiment overestimates the *counting* ionization (due to double counting of the dissociative ionization)

$$\sigma = \sum_n 4\pi a_0^2 \xi_n \left(\frac{R}{I_n}\right)^2 \frac{1}{t+u_n+1}\left\{1-\frac{1}{t}+\frac{\ln t}{2}\left(1-\frac{1}{t^2}\right)-\frac{\ln t}{t+1}\right\}$$

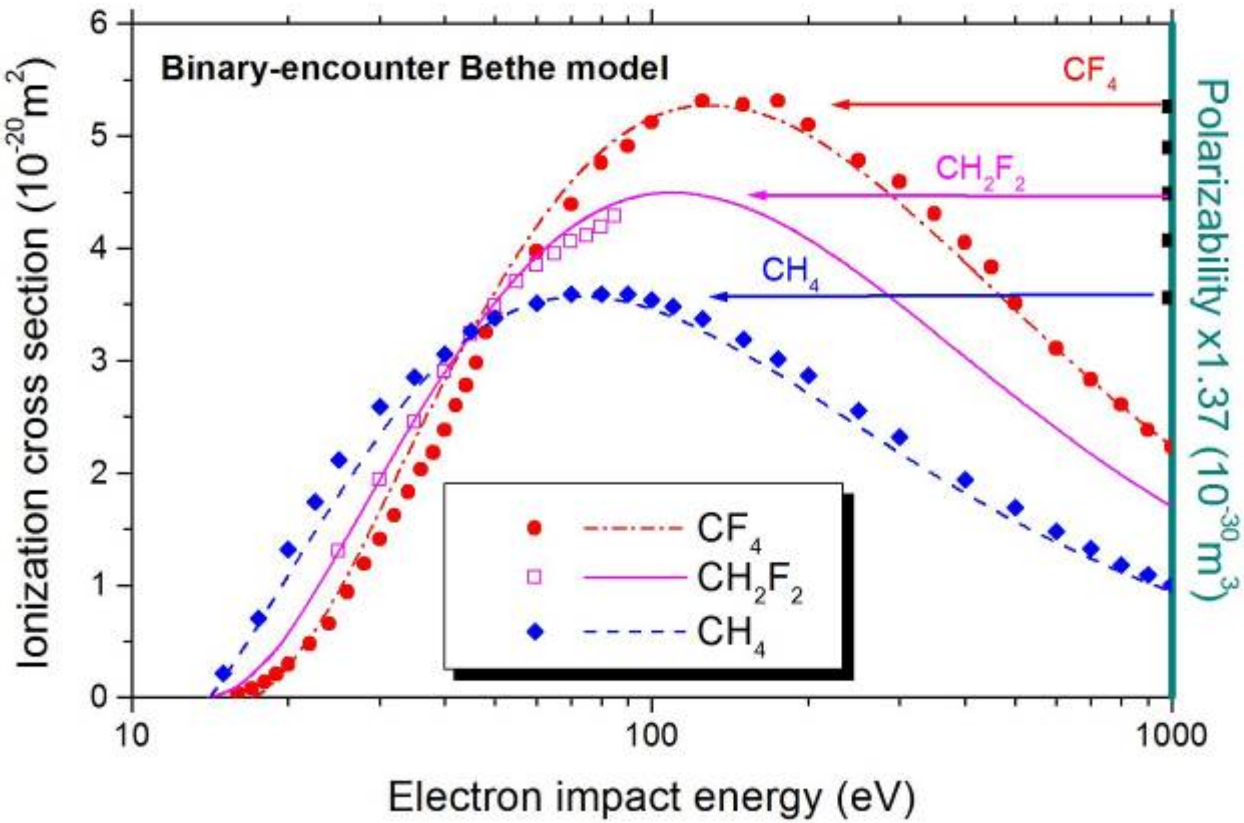
Normalized energies: $t = E/I_n$, $u_n = E_{kin}/I_n$

Only two values needed from Quantum Chemistry

G. Karwasz, P. Mozejko, M.-Y. Song, Int. J. Mass Spectrometry 365/366 (2014) 232

Ionization (BEB): CH_4, CH_3F, ... CF_4

Scalling observed for the methane analogues



Thumb rule (?)

$$\sigma_{max} = 4/3\ \alpha$$

G. Karwasz, P. Mozejko, M.-Y. Song, Int. J. Mass Spect. 365 (2014) 232

Ionization (Fe^{+24}): CCC theory

Scalling with the number of electrons?

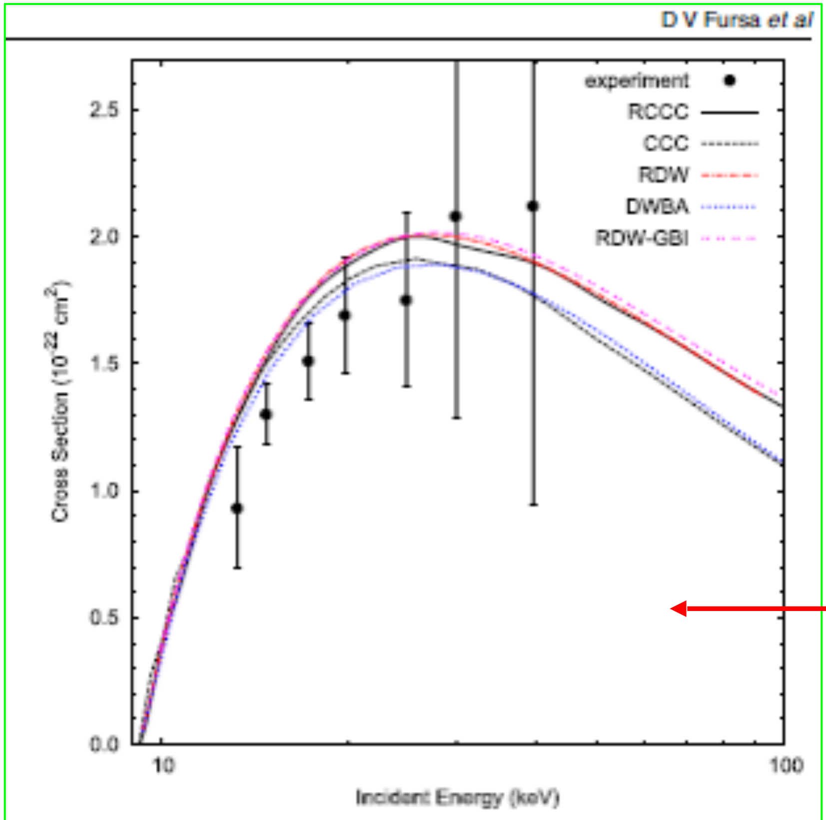


Figure 1. Electron-impact ionisation cross section for Fe^{25+} ion. The present RCCC, CCC, RDW and DWBA calculations are described in the text. The RDW-GBI calculations are due to [21]. The experiment is due to [25].

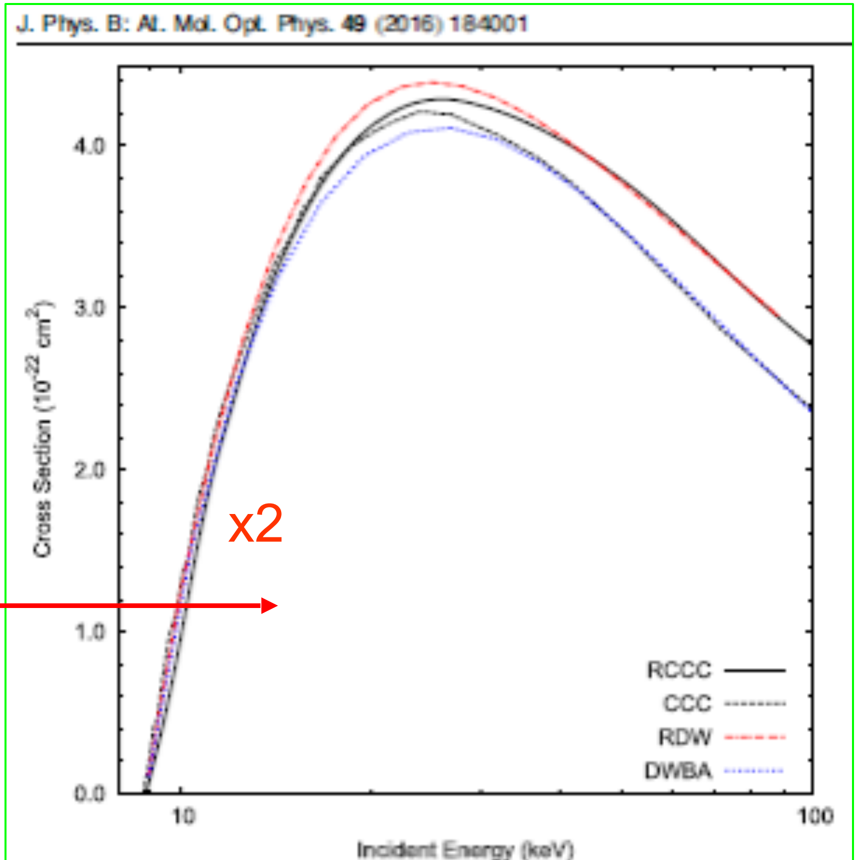
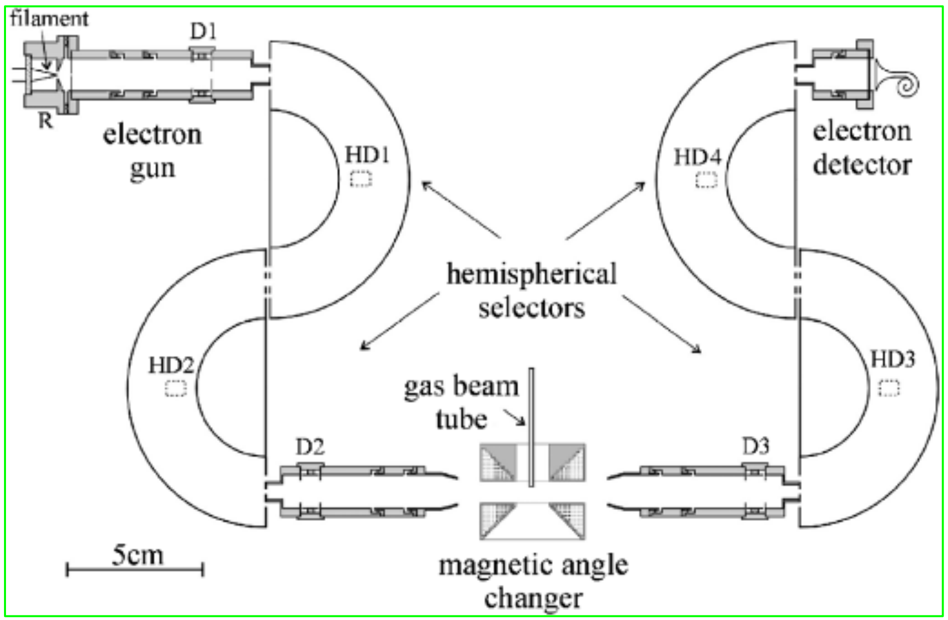


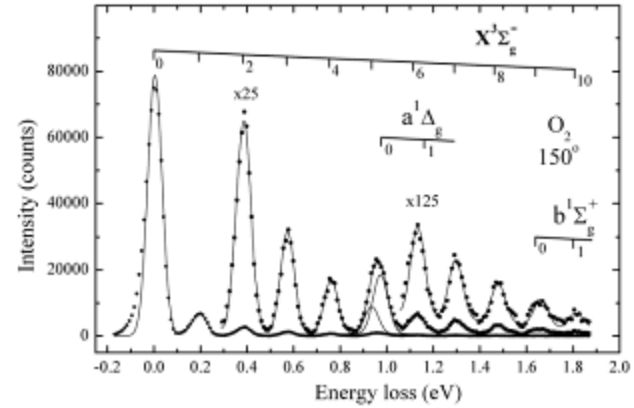
Figure 3. Electron-impact ionisation cross section for Fe^{24+} ion. The theoretical calculations are as in figure 1.

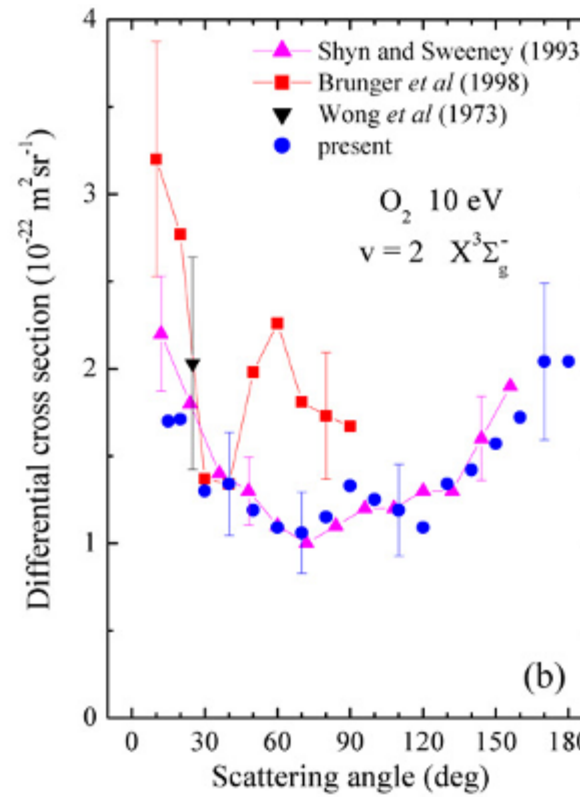
Fursa et al. J. Phys. B **40** (2018) 93 184001

Experimental methods: excitation (electronic, vibrational)

These are difficult experiments: require careful normalization and extrapolation
→ uncertainty >20%







Experiments by:

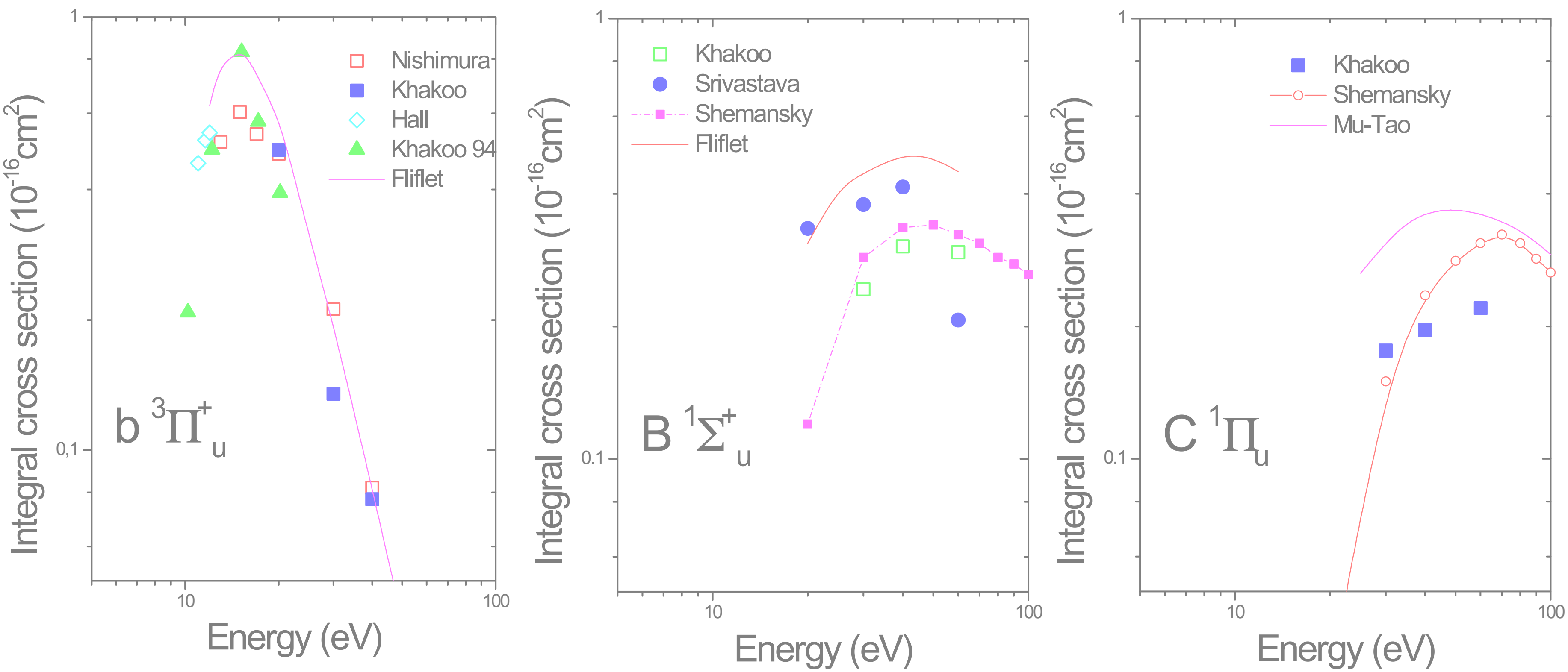
I. Linert, M. Zubek (Gdansk) J. Phys. B **39** (2006)

M. Khakoo et al. (Fullerton California)

M. Allan (Freiburg University)

$e^- + O_2$ (v=0) – $e^- + O_2$ (v=0, 1, 2, e

Electronic excitation H_2: state of art in 1996



A. Zecca, G. Karwasz, R.S. Brusa, Riv. Nuovo Cimento 19, No. 5 (1996), 1-146

Electronic excitation: H_2 (experiment)

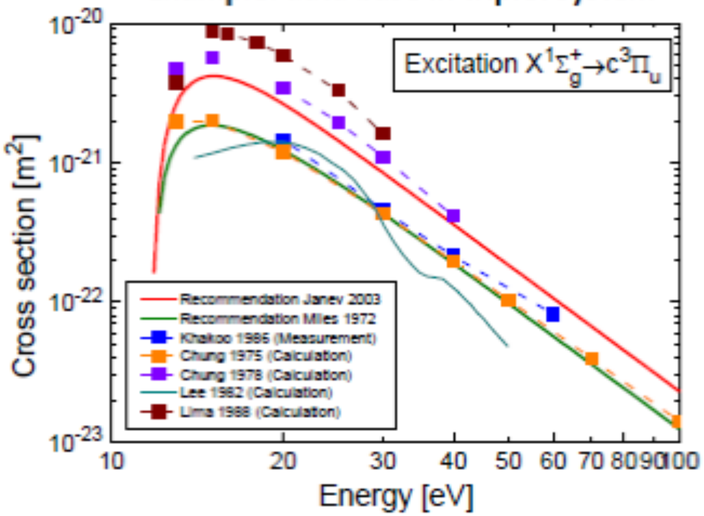
Molecular emission in negative hydrogen ion sources for fusion

IPP

Collisional radiative model for H and for H_2: Yacora

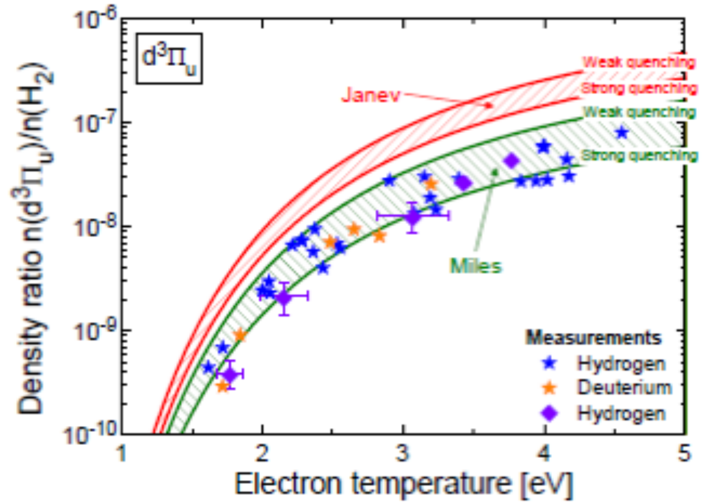
Important parameter: density ratio H/H_2 obtained from the intensity ratio H_γ / Fulcher band

Example: data base in triplet system



Differences by 5-folds

Benchmark with experiments:
better agreement for
the semi-empiric Miles data
but
uncertainty of a factor of 3 remains

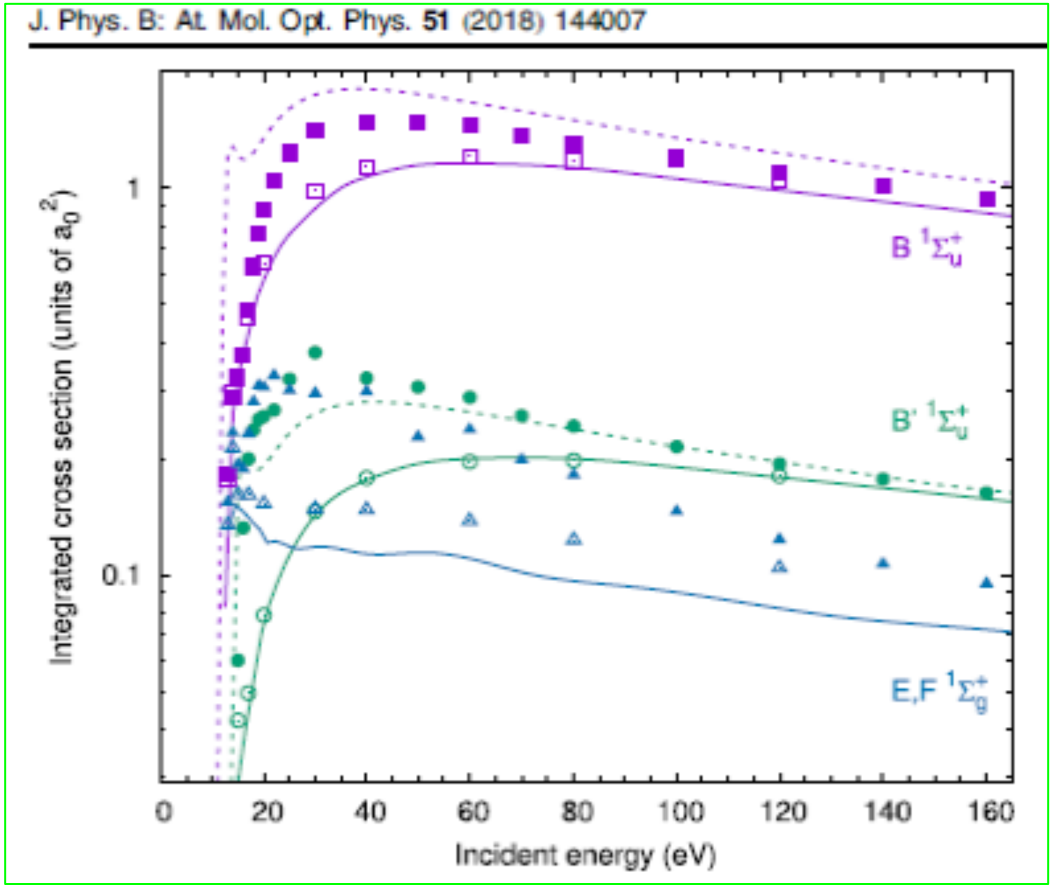


© Ursel Fantz, 19

IAEA TM, A+M+PMI, December 15–19,2014

Courtesy: Ursel Fantz (IAEA Meetings, Daejon, 2016)

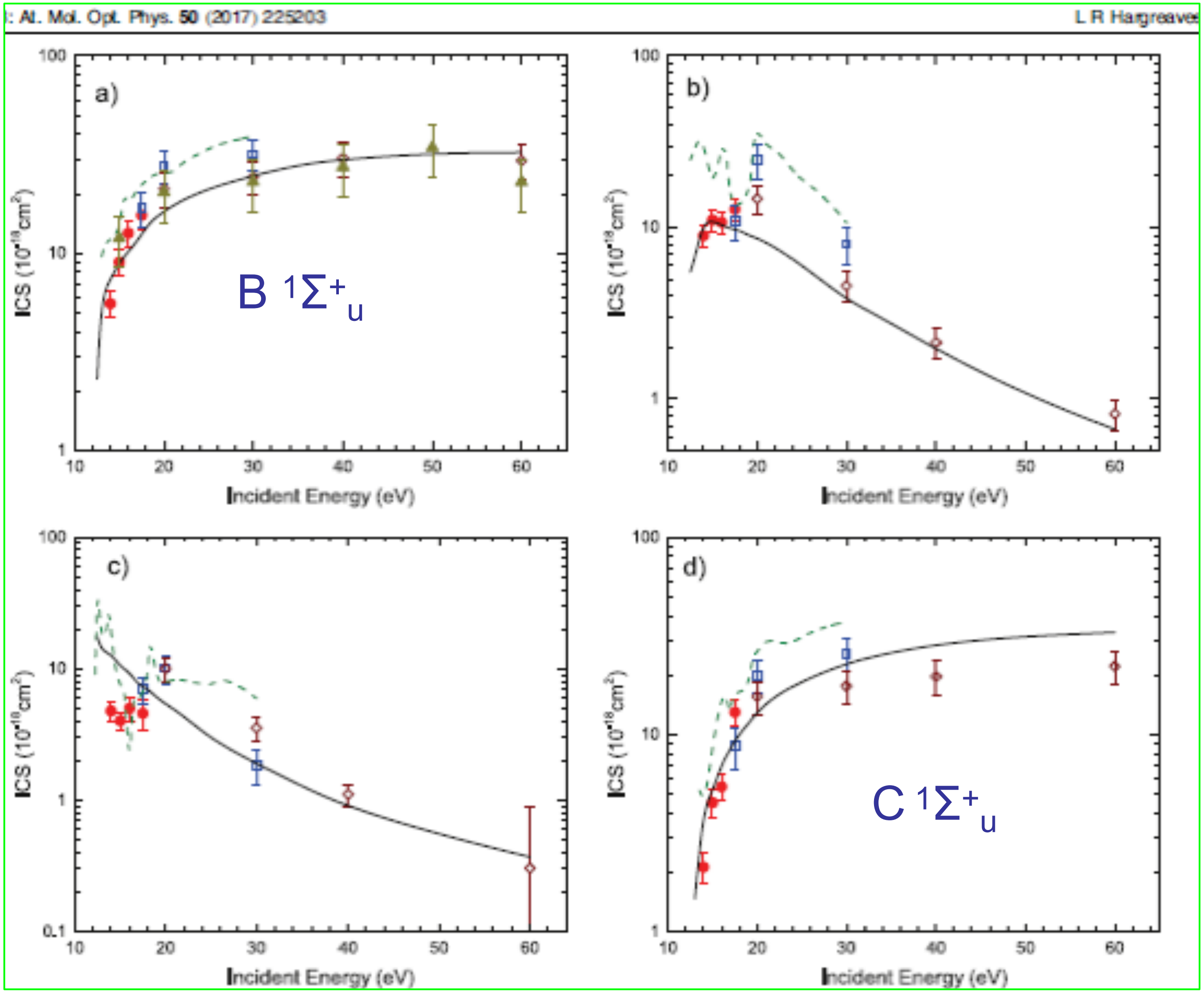
Electronic excitation: H_2 (theory)



Agreement within error bar with experiments

Convergent close coupling:
J. K. Tapley,.. D. Fursa, I. Bray, J. Phys. B 51 (2018) 144007

Electronic excitation: H_2 (theory)



Hargreaves et al.. J. Phys. B (2017)

Agreement within error bar with experiments

Electronic excitation: CH_4

Electronic excitation: reasonable agreement between dissociation into-neutrals experiment and R-Matrix calculation but theory shifted down in energy by 3 eV

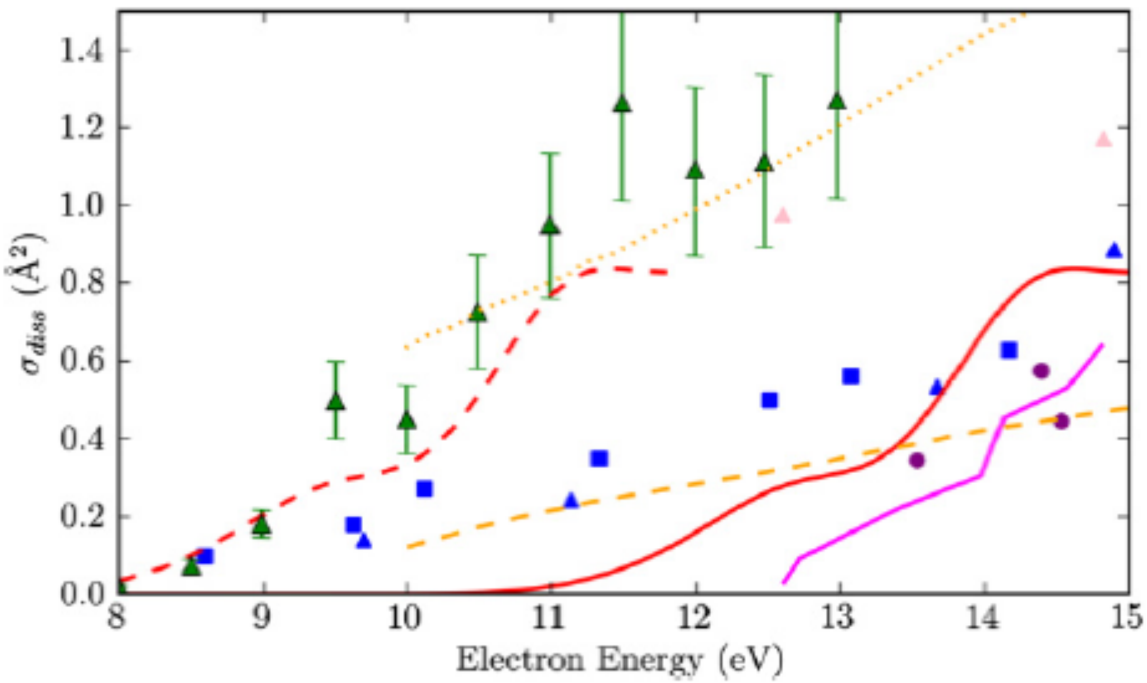
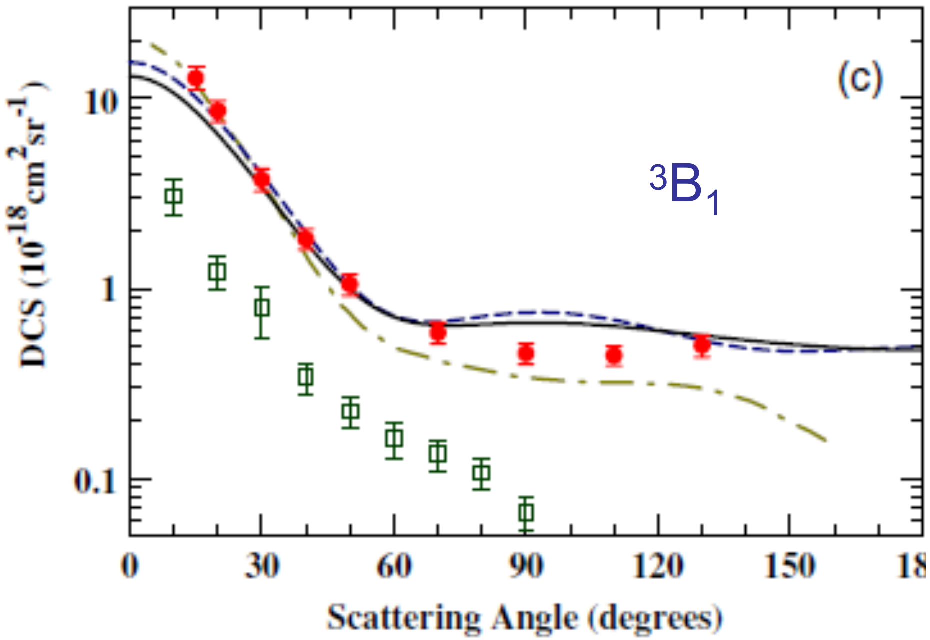
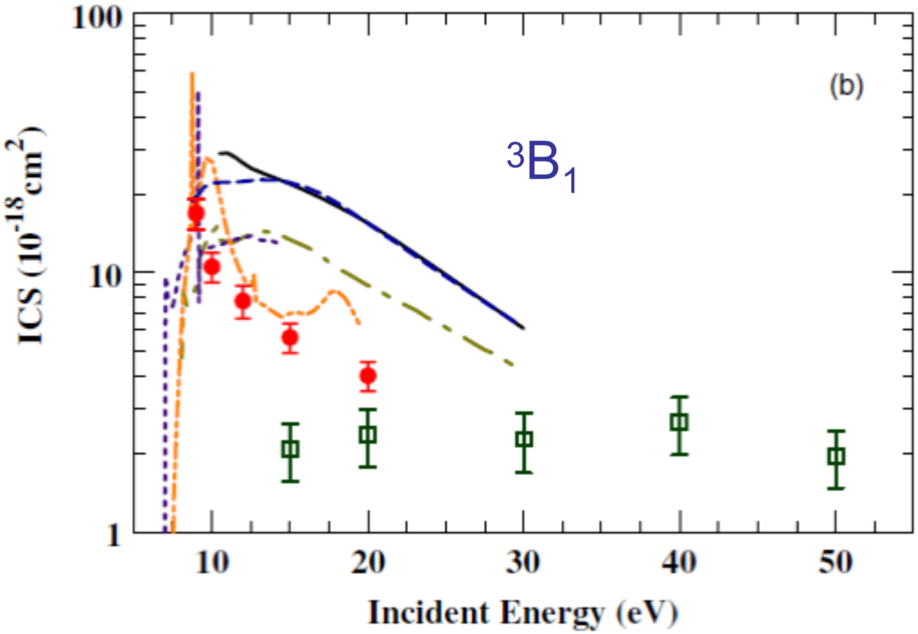


Figure 13. Electron impact dissociation cross section. Theory: red solid line: present work; red dashed line: present work, shifted to lower energy by 3.2 eV; purple solid line: Hayashi (1991); orange dashed line: CH_2 of Ziółkowski *et al* (2012); orange dotted line: CH_3 of Ziółkowski *et al* (2012). Experiment: blue squares: CH_2 of Nakano *et al* (1991); blue triangles: CH_3 of Nakano *et al* (1991); green triangles: CH_3 of Makochekanwa *et al* (2006); pink triangles: CH_3 of Motlagh and Moore (1998); purple circles: Winters (1975).

W. J. Brigg, J. Tennyson, M. Plummer
J. Phys. B **47** (2014) 185203
R-Matrix

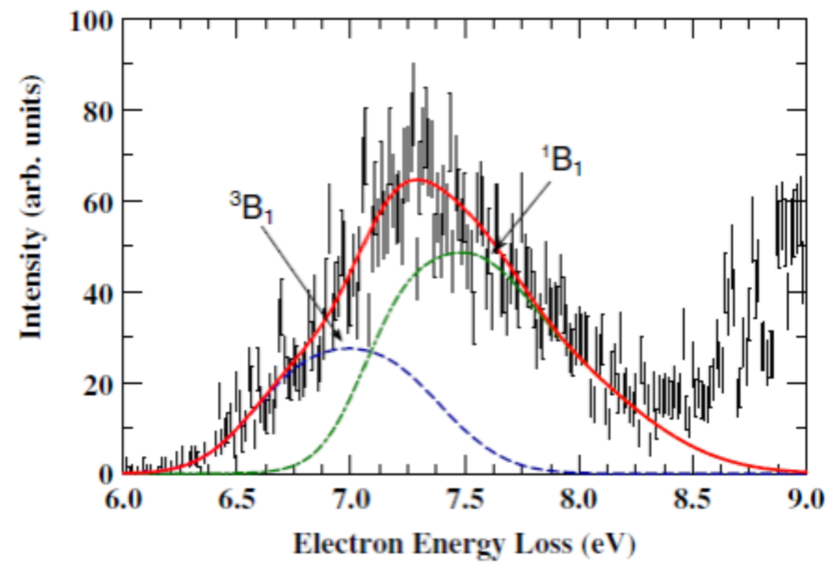
Electronic excitation: H_2O



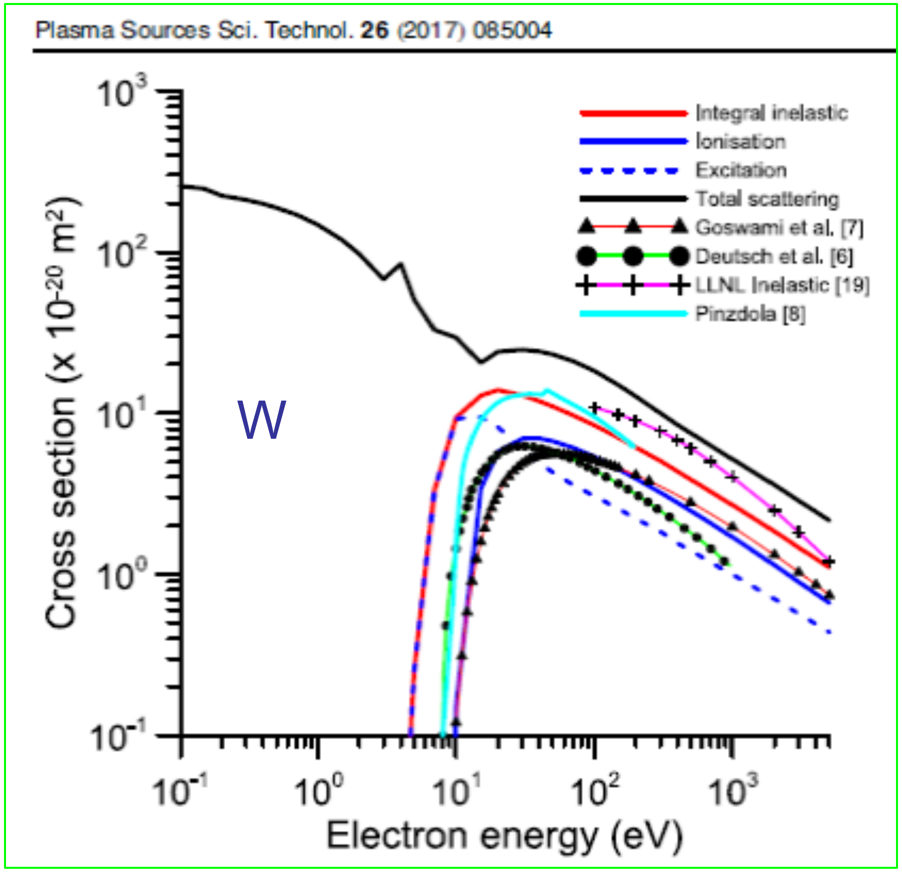


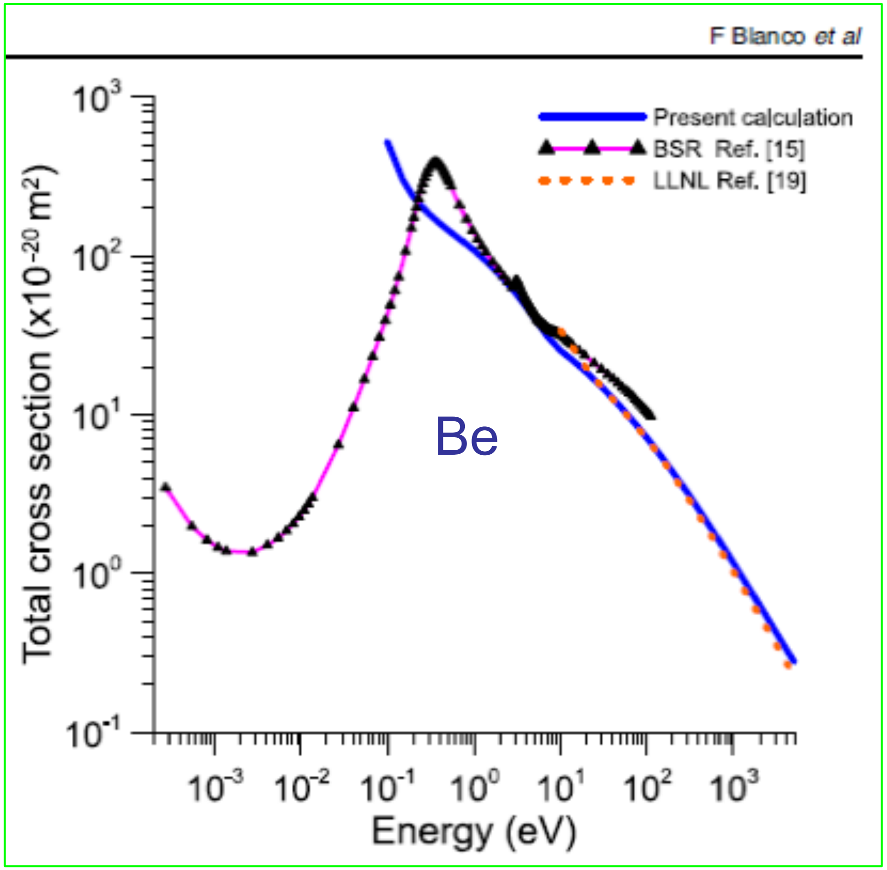
L. Hargreaves et al., J. Phys. B 45 (2012) 201001 (Fullerton, CA)
squares: Thorn et al. J. Chem. Phys, 126 (2007) 064306 (Adelaide)

Reason for discrepancy: wrongly resolved energy-loss spectra



Tungsten, berillium (model potentials)

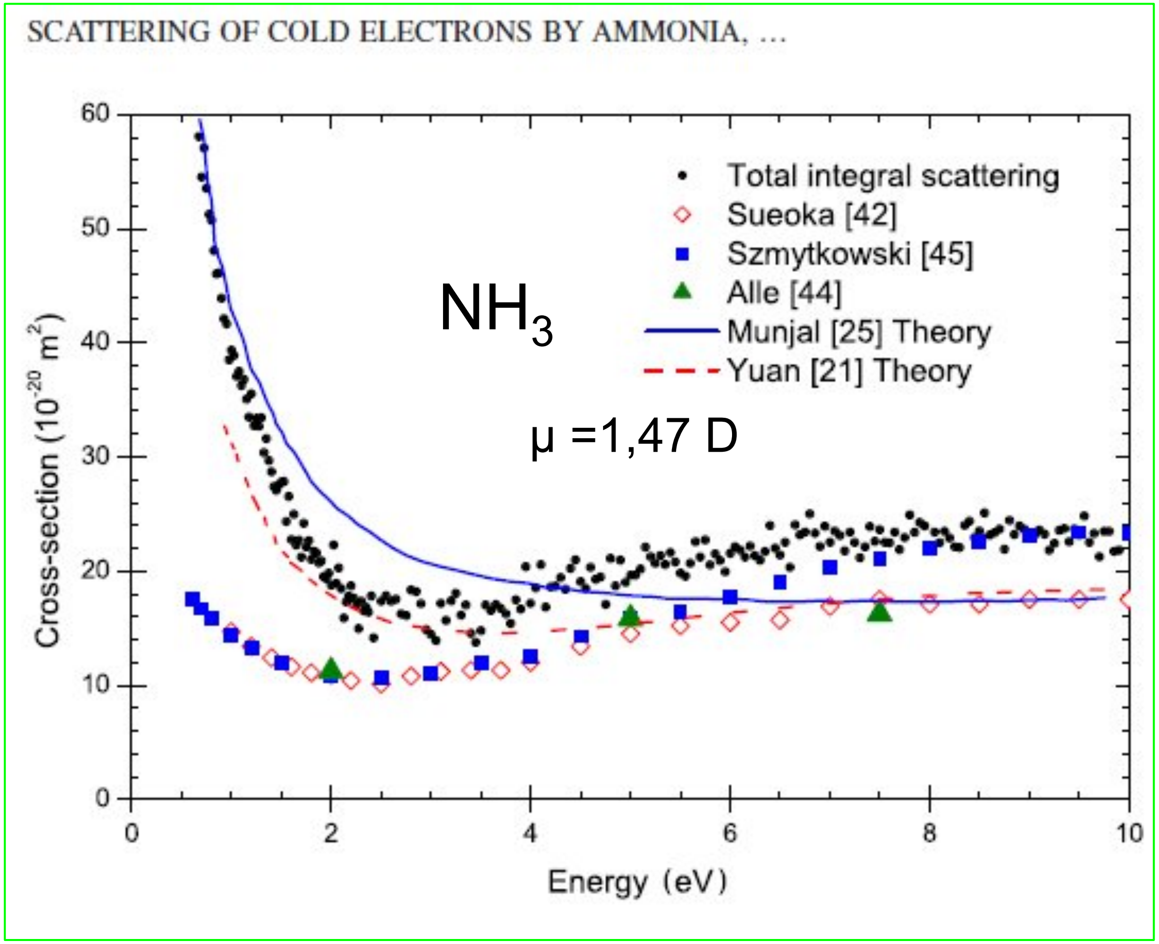




[15] Zatsarinny (2015)

F. Blanco et al. Plasma Sources Sci. Technol. 26 (2017) 085004

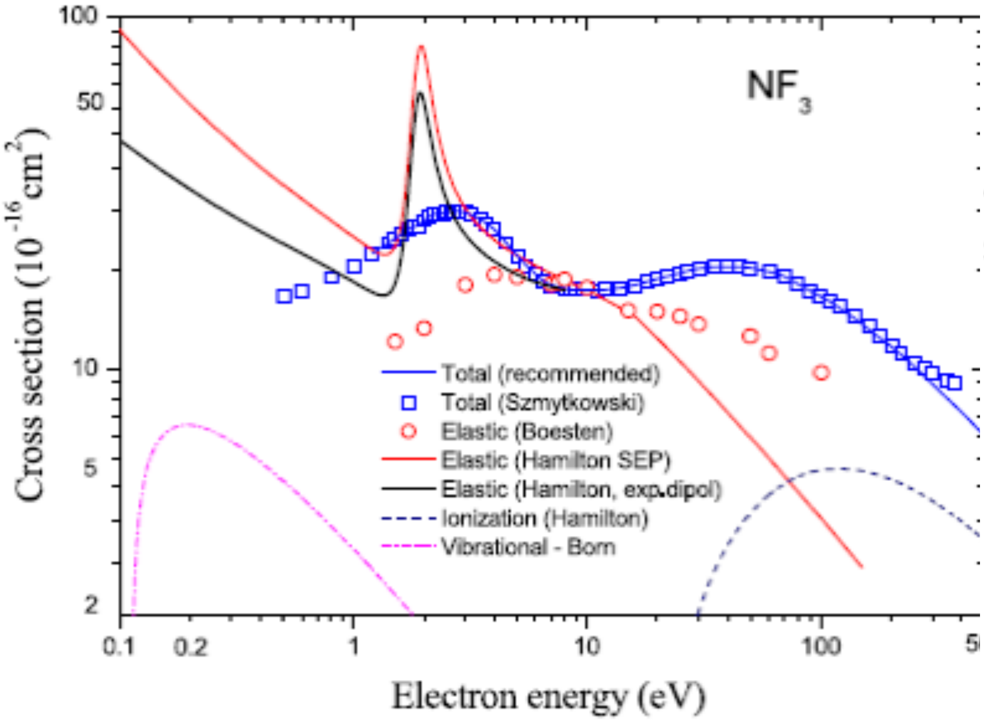
ITER-like: NH_3 total cross section

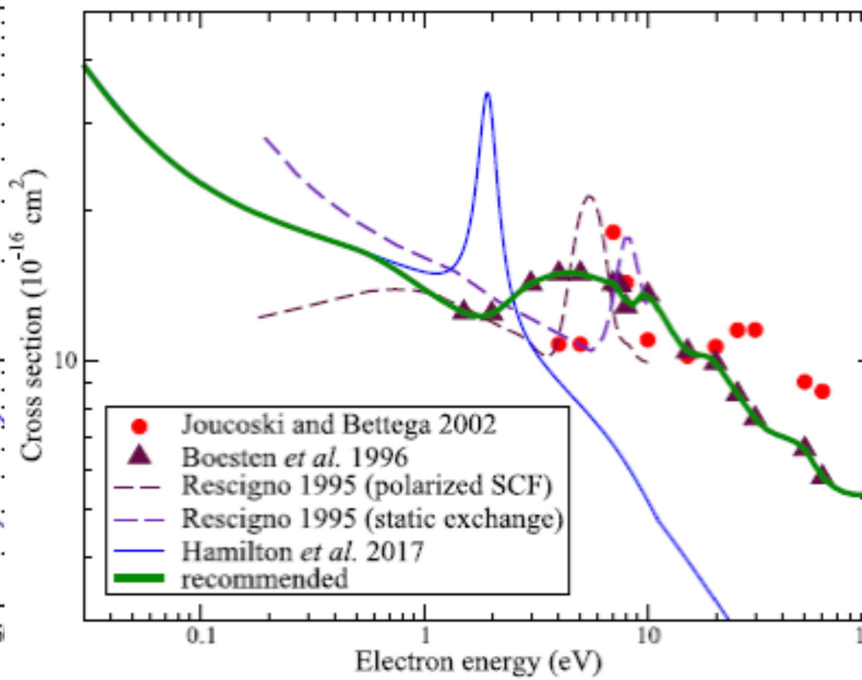


A serious disagreement between different beam experiments
Gdańsk TU and synchronous-radiation very low energy data (Jones)

Jones et al. PRA **78**, 042714 (2008)

Weakly polar (NF_3)





Poor agreement between experiment total, theory and swarm-derived momentum cross sections (and no MERT developed)

M.-Y. Song et al.. JPCRD 46 (2017) 043104

Total $e^+ + H_2O$: ANU measurements

Recent „corrected” ANU & Trento [Zecca] do not agree

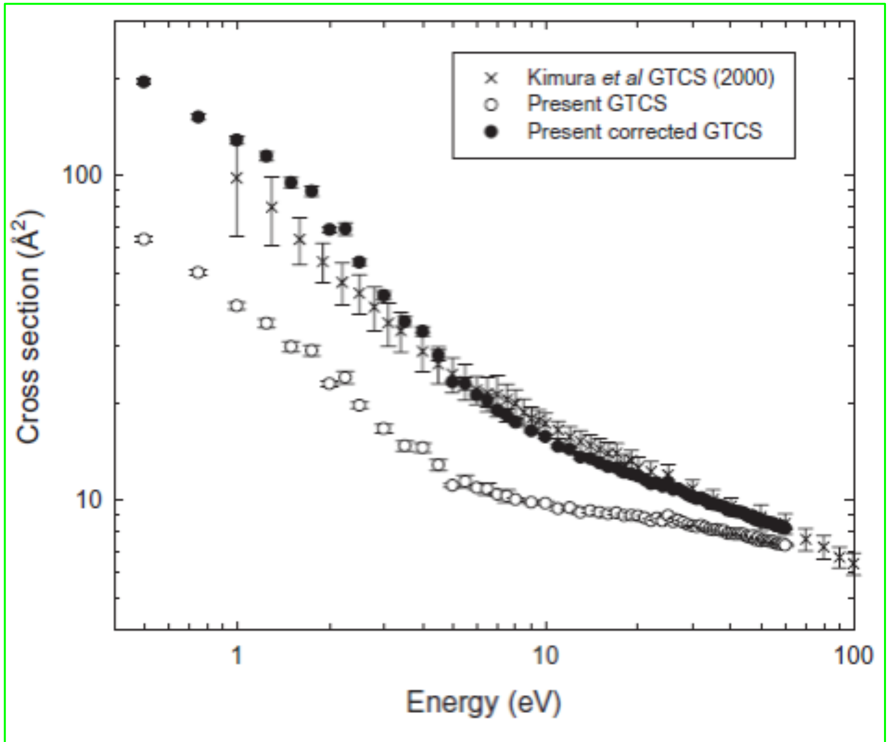


Figure 8. Present positron impact H_2O GTCS before and after co

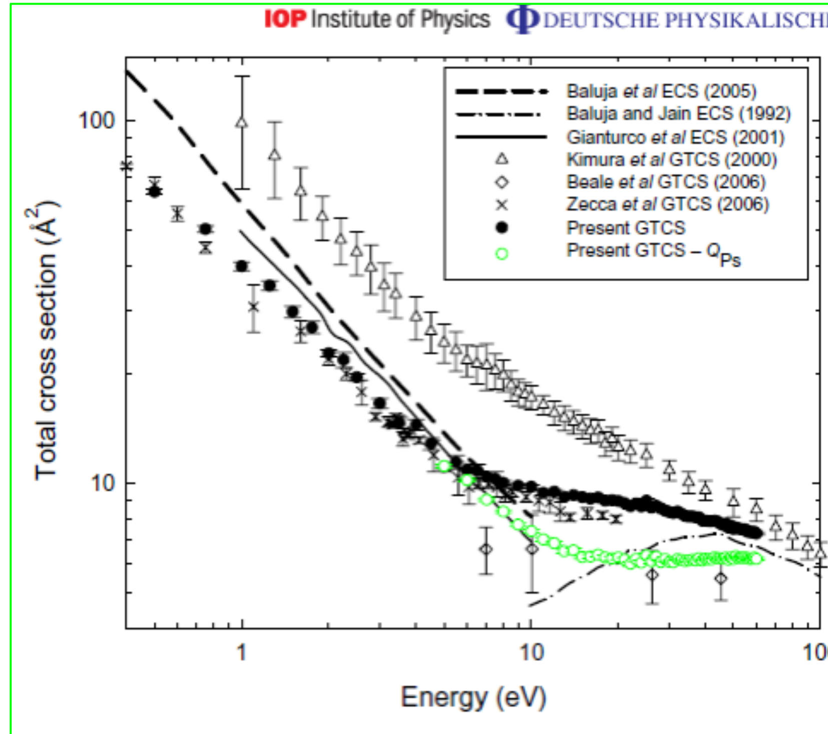


Figure 1. Present positron impact H_2O GTCS and GTCS – Q_{Ps} c

Difference by almost a factor of 3x

C. Machochekanwa et al., New J. Phys. 11 (2009) 103036

Polar molecules (aceton): low energies

- Experiment (total): angular resolution error
- Similar errors in other polar molecules including NH_3, H_2O
- Theory: two models (Schwinger mutlichannel + Born and Independent Atoms) differ much!

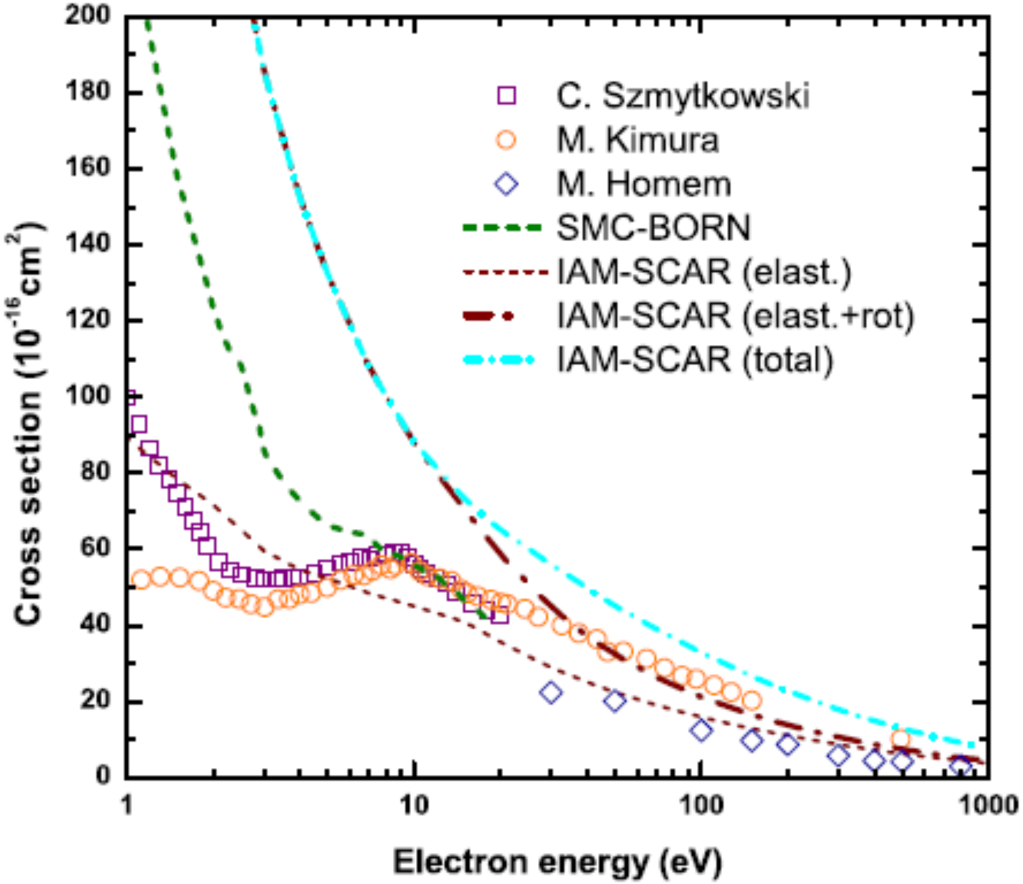
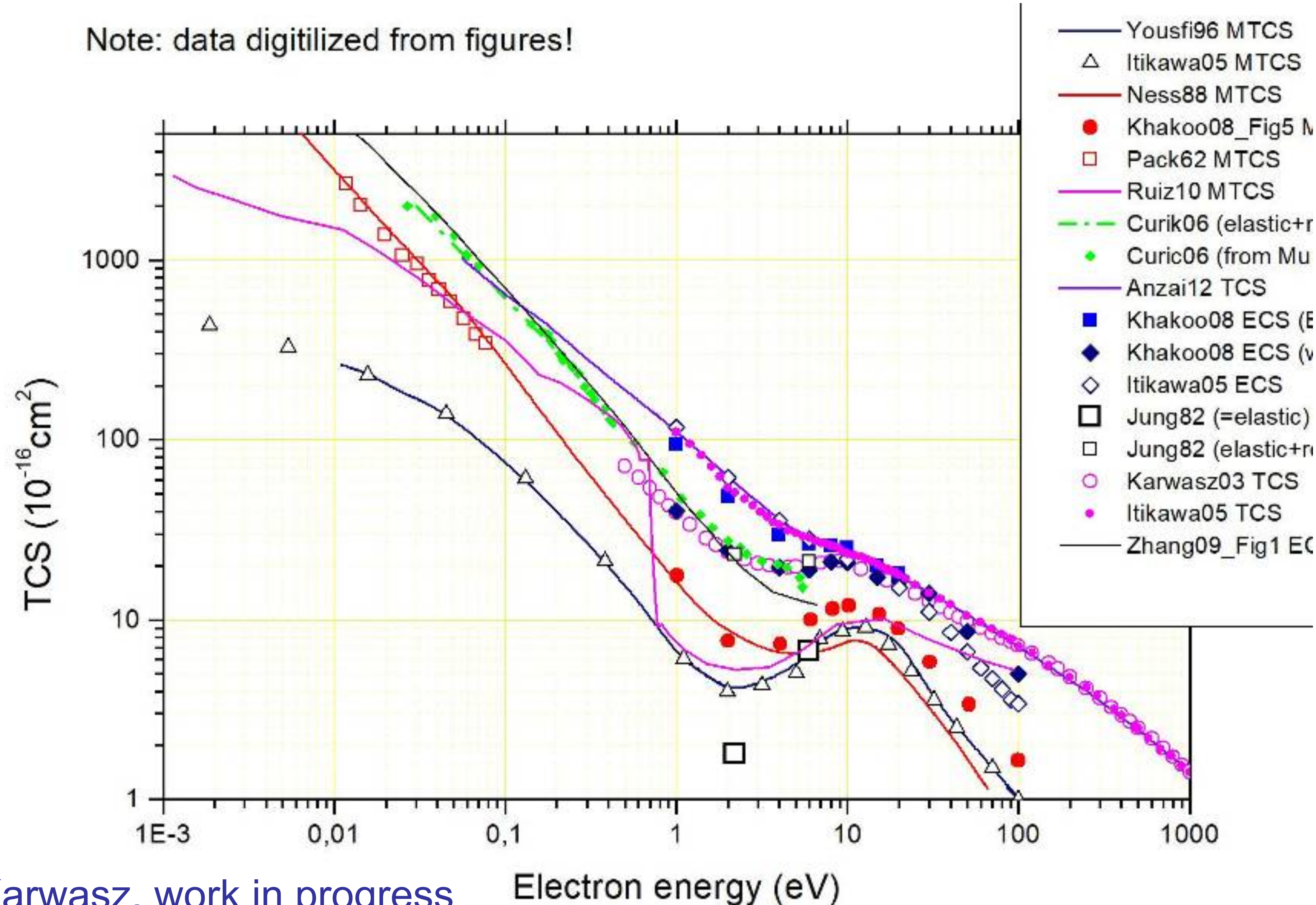


FIG. 2. Integral cross section for electron collisions with acetone. Our SMC-SEP results with Born-closure procedure, dashed line; total cross section of Szmytkowski [15], squares; total cross section of Kimura *et al.* [16], circles; integral cross section of Homem *et al.* [18], diamonds.

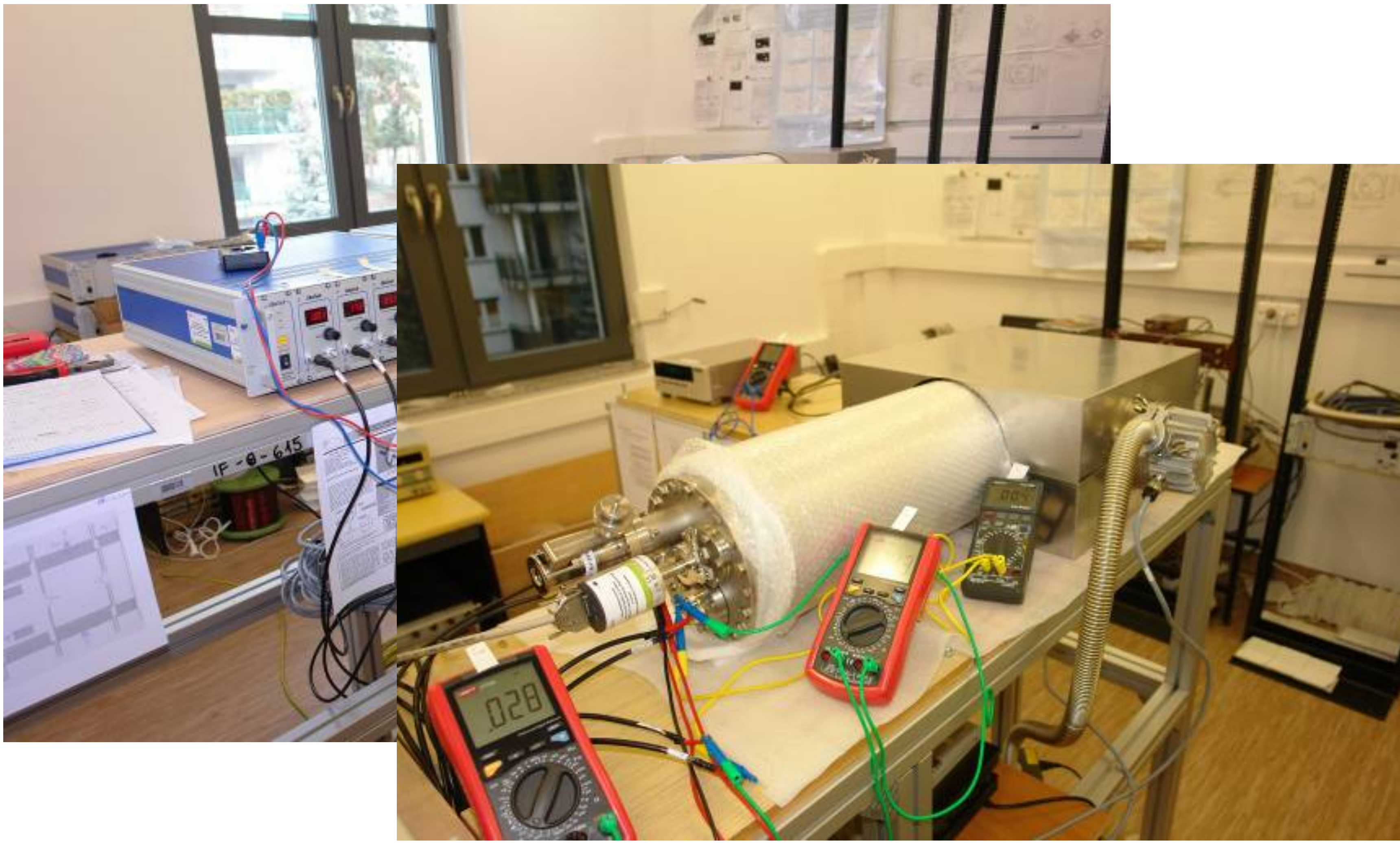
Pastega et al.. PRA 93 (2016) 032708

Total $e^- + H_2O$: state of art



Karwasz, work in progress

Work in progress (Toruń lab)



A set for very-low energy total cross sections on polar molecules

Some pending ideas:

- Still no matching between cross sections from different methods (swarm, beam) in important ITER-like targets like NH_3, W – compunds etc.

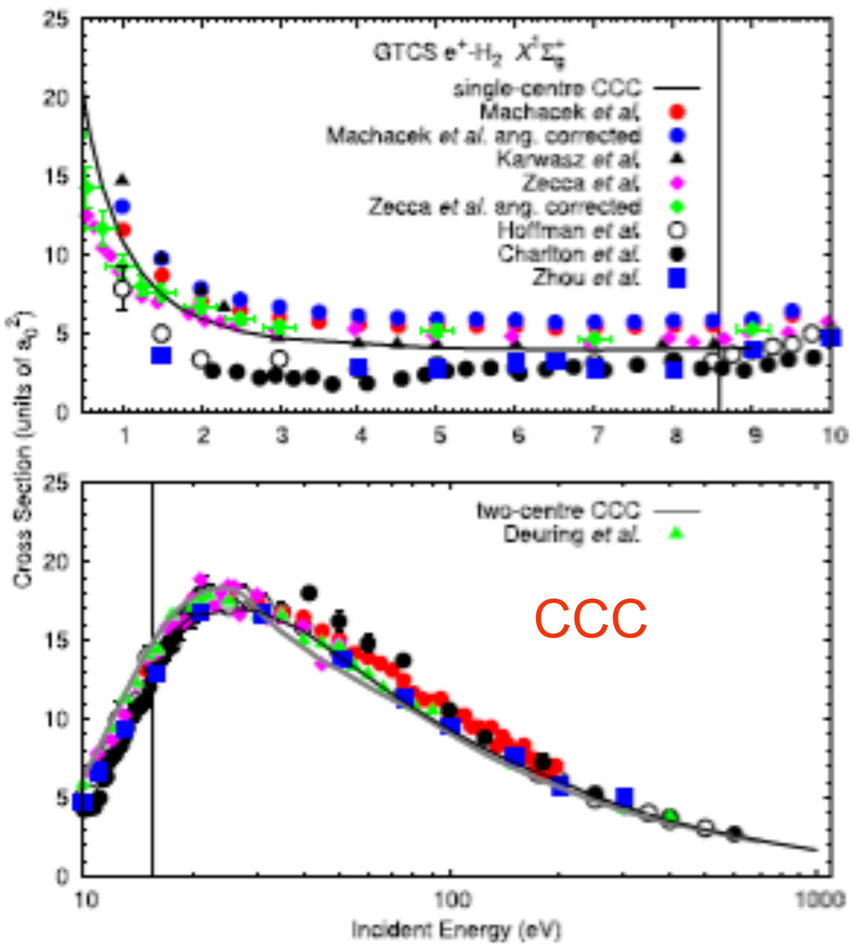
- Semi-empirical models still in use:

- in ionization total
- in ionization partial (?)
- in total at high energies (a possible additivity rule)
- in total at very low energies (MERT) – but only spherical
- in vibrational outside resonances (i.e. near threshold)
- in polar molecules - search for scaling at low energies (?)

- **significant progress in theory has been done**

Optimistic resumé: excellent state of art is $e^+ + H_2$ (total, Ps, ionization)

J. Phys. B: At. Mol. Opt. Phys. 50 (2017) 123001 Tutorial



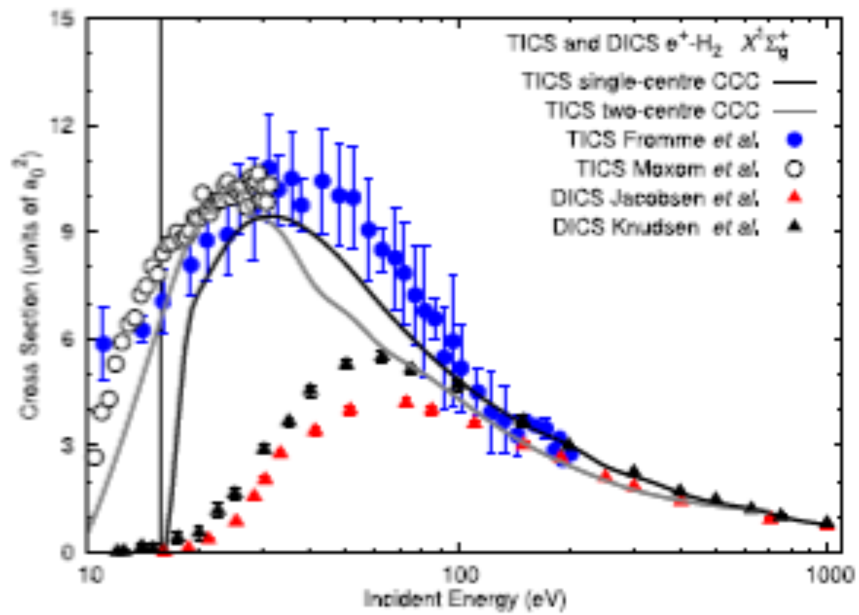


Figure 10. CCC results of the total (single) ionisation cross section (TICS) for positron scattering from the ground state of H_2. The mean internuclear distance $R_m = 1.448\ a_0$ single-centre CCC (present) TICS are compared with the $R_0 = 1.4\ a_0$ two-centre CCC TICS [109], the measurements of Fromme *et al* [128] and Moxom *et al* [129], and the direct ionisation cross section (DICS) measured by Jacobsen *et al* [130] and Knudsen *et al* [131]. The dashed–dotted vertical line at 15.4 eV indicates the ionisation threshold of H_2 in the ground state.

M. Zammit et al. J. Phys. . 50 (2017) 123001 2017

Acknowledgments:

International Atomic Energy Agency
Atomic Molecular Data Services
Provided by the Nuclear Data Section

Evaluation group: M.-Y. Song, J.-S. Yoon (NFRI) H. Cho (Chungham Uni, S. Korea), Y. Itikawa (Sagamihara, Japan), J. Tennyson (UC London), V. Koukulin (Orlando), Y. Nakamura (Japan)

Gdańsk group: Cz. Szmytkowski, J. Mechlińska-Drewko, M. Zubek, P. Możejko, J. Franz

Trento group:
A. Zecca, R. S. Brusa

Toruń group:
K. Fedus (also Fullerton)
A. Karbowski

Thank you for the attention!