Electron scattering on molecules - partial (and total) cross sections: search for uncertainties and errors in experimental procedures

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Nicolaus Copernicus University,
Toruń, Poland

IAEA Meeting, Wien, 19.12.2016

Data needed:

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

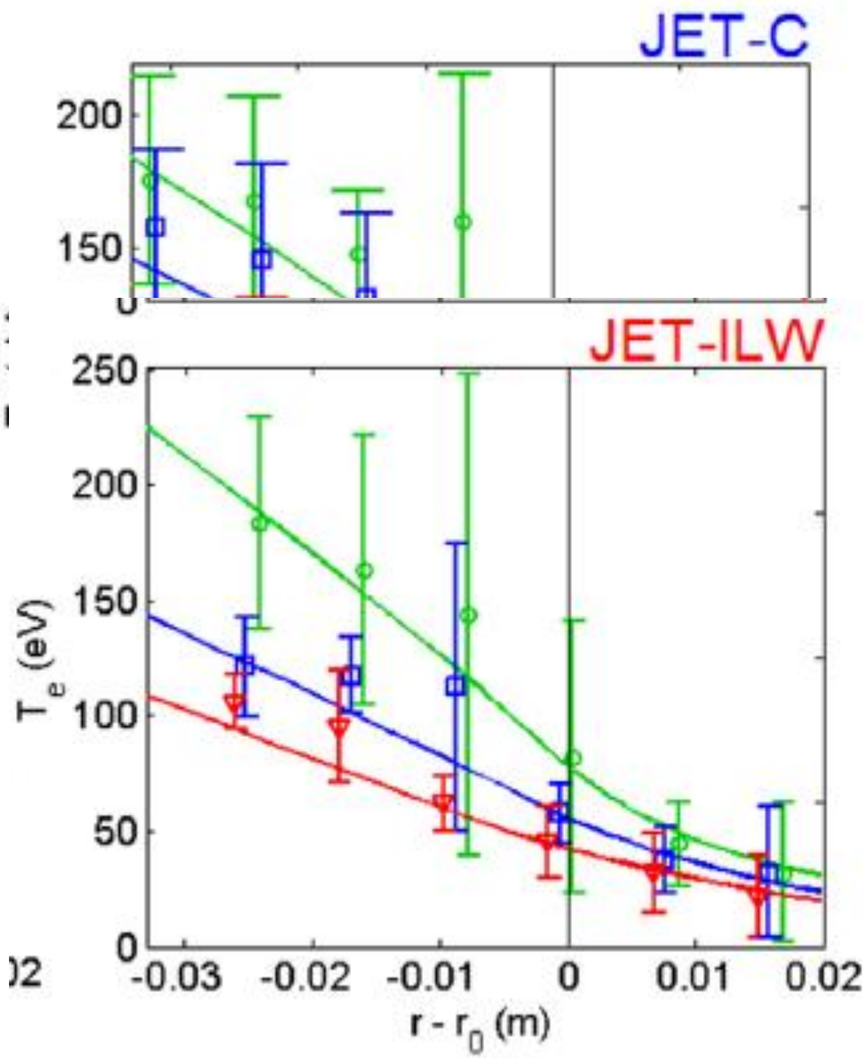
emisison from dissociation $e + AB \rightarrow A^* + B + e + h\nu$

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

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

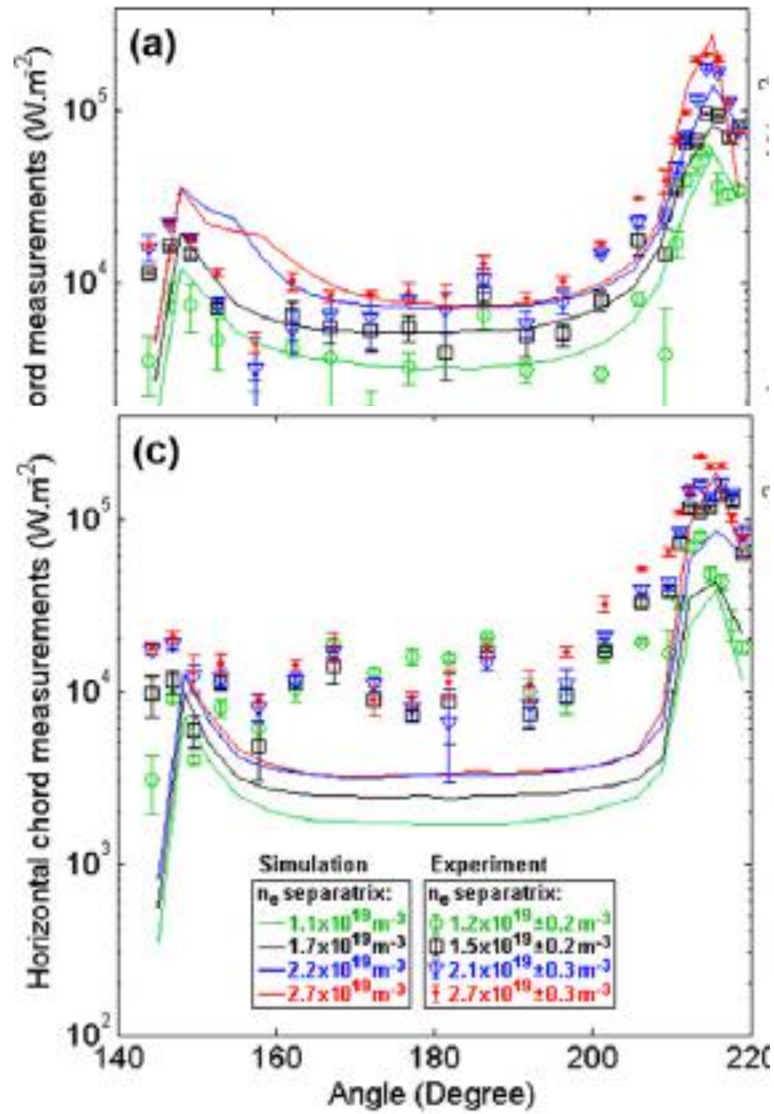
ionization into excited states $e + A \rightarrow (A^+)^* + 2e$

¿ITER: electron T and power irradiated



Electron temperature (and density) during three points of density ramp

Guillemaut et al. Nucl.Fusion (2014)



Power irradiated (0.5-1.5 MW) simulation:
JET-C <10% **JET-ILW factor 3!**

ITER:
wall sputtering

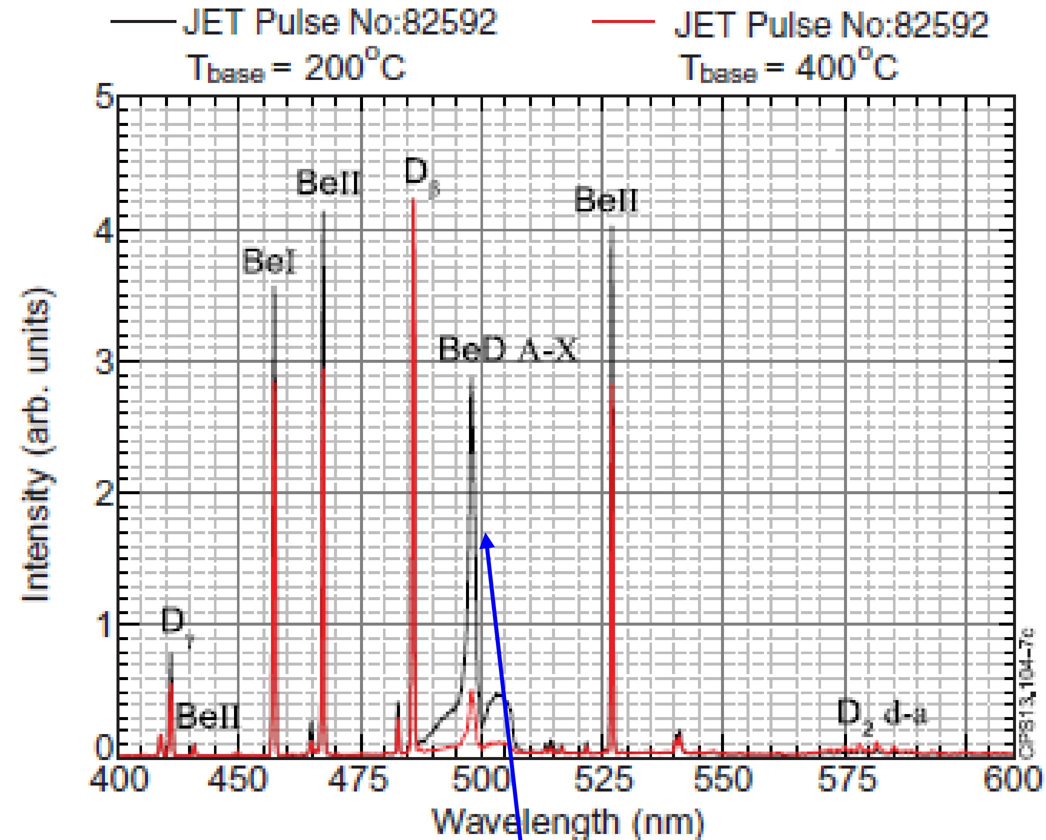


Figure 7. Emission spectra recorded at the Be limiter at the first and last discharge of the series in experiment I representing low T_{base} [JPN #82592] and high T_{base} [JPN #82626] conditions.

BeD

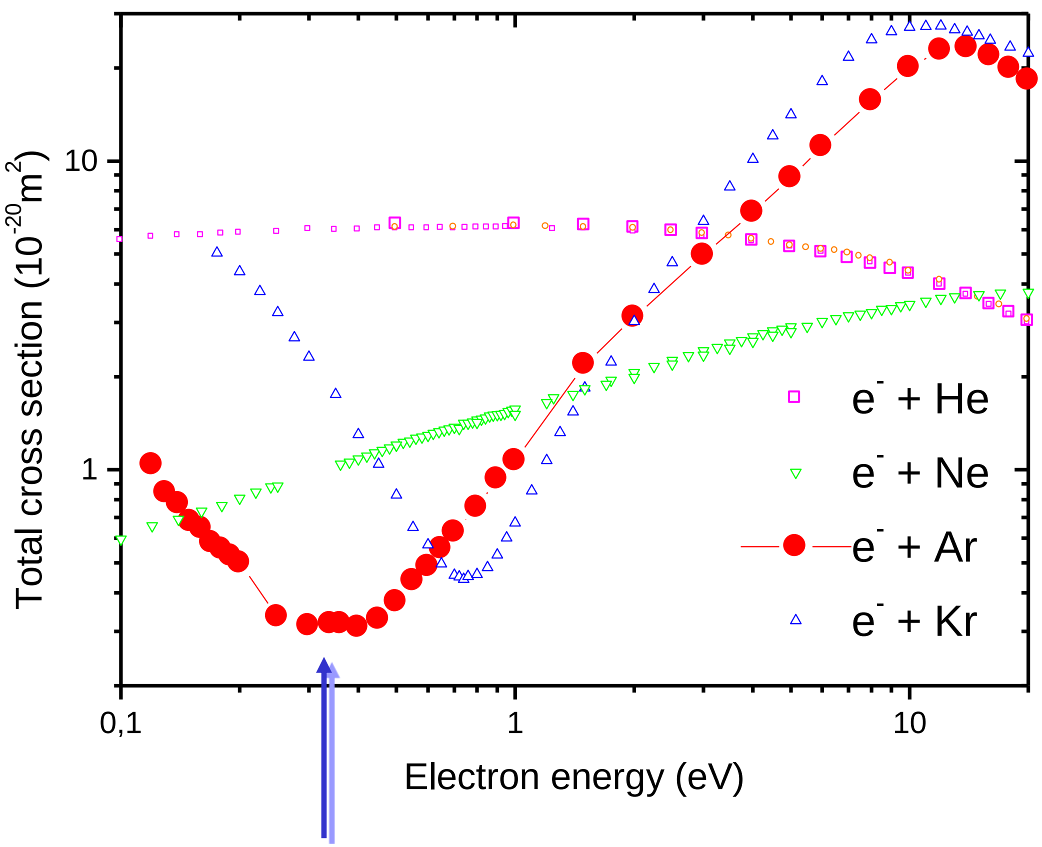
IOP Publishing | International Atomic Energy Agency Nuclear Fusion

Nucl. Fusion **54** (2014) 103001 (11pp) doi:10.1088/0029-5515/54/10/103001

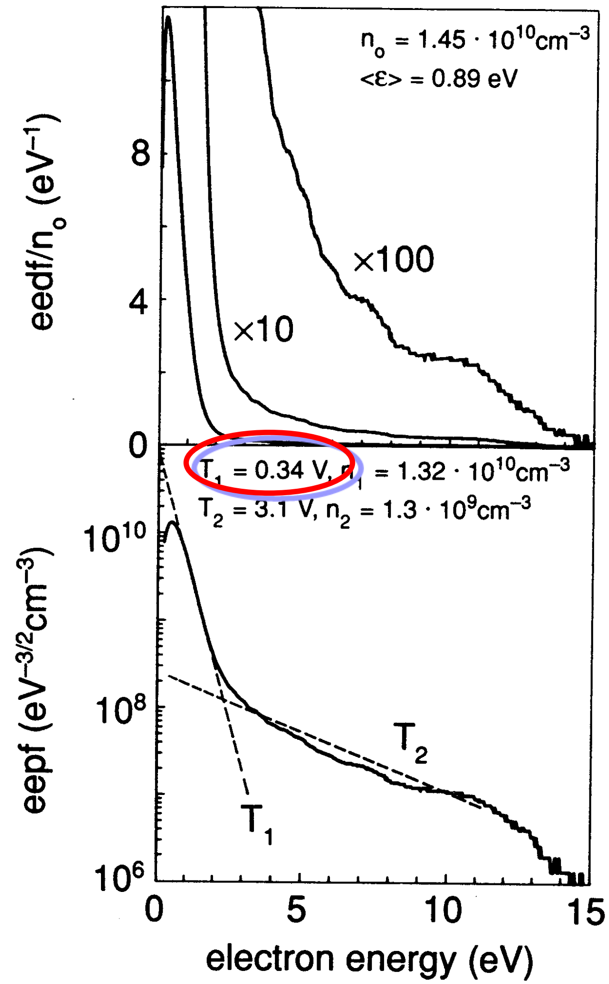
Study of physical and chemical assisted physical sputtering of beryllium in the JET ITER-like wall

S. Brezinsek[1], M.F. Stamp[2], D. Nishijima[3], D. Borodin[1], S. Devaux[4], K. Krieger[4], S. Marsen[4], M. O'Mullane[2], C. Bjoerkas[5], A. Kirschner[1] and JET EFDA contributors[a]

Plasma temperature ← integral cross sections



Ramsauer
minimum
(zero in *s*-wave)



V. Godyak, Sendai 2006

Radiation damage in biological tissues

112 *M.C Fuss et al. / Chemical Phys*

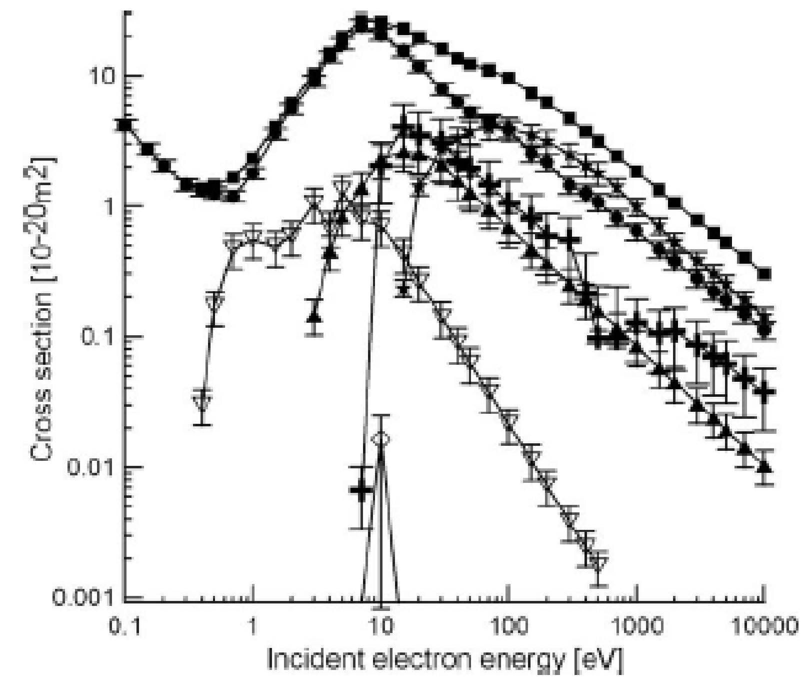
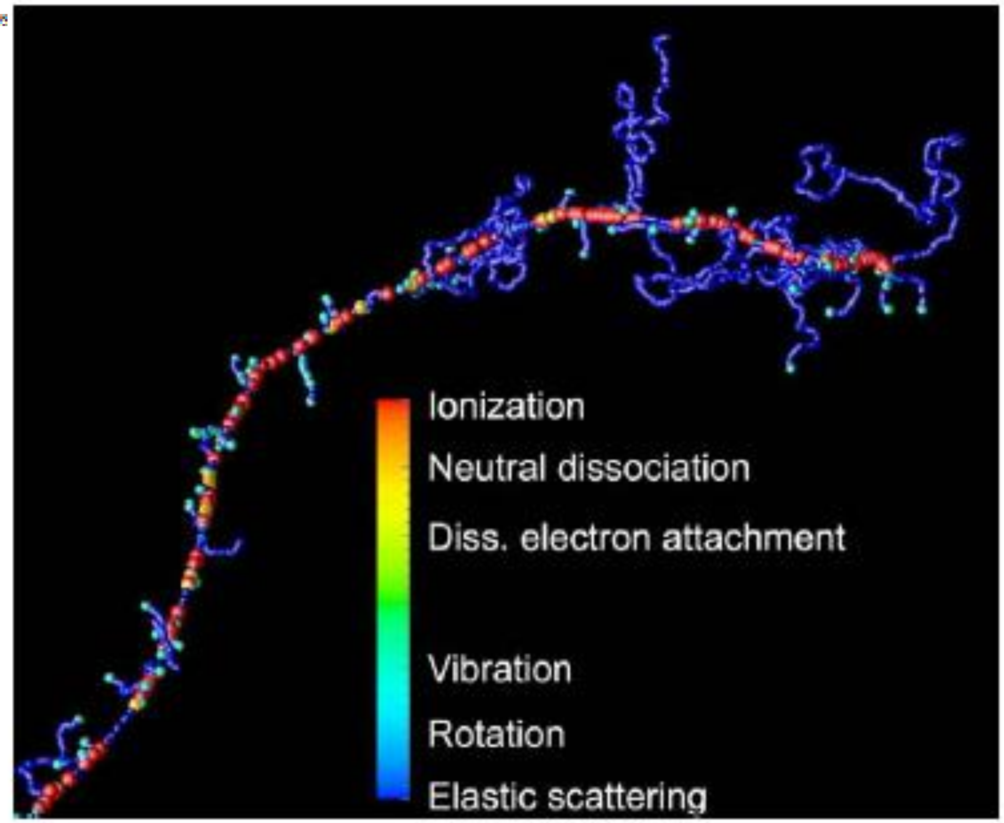


Fig. 1. Integral electron–methane interaction cross sections used in the present work (data of Table 1): ■, total interaction cross section; ●, elastic collision; ★, ionization; ▲, rotational excitation; ▿, vibrational excitation; +, neutral dissociation; ◇, dissociative electron attachment. For the total interaction cross section, error bars fall within the size of the symbol used.



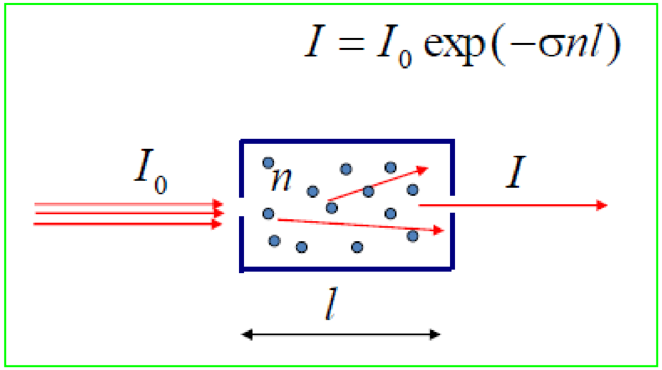
ig. 6. Detail showing the final part (~1 cm) of one electron track. The colour does ..ere indicate the type of interaction undergone, including elastic collisions. The production of multiple secondary electrons can be discerned along the primary particle's path.

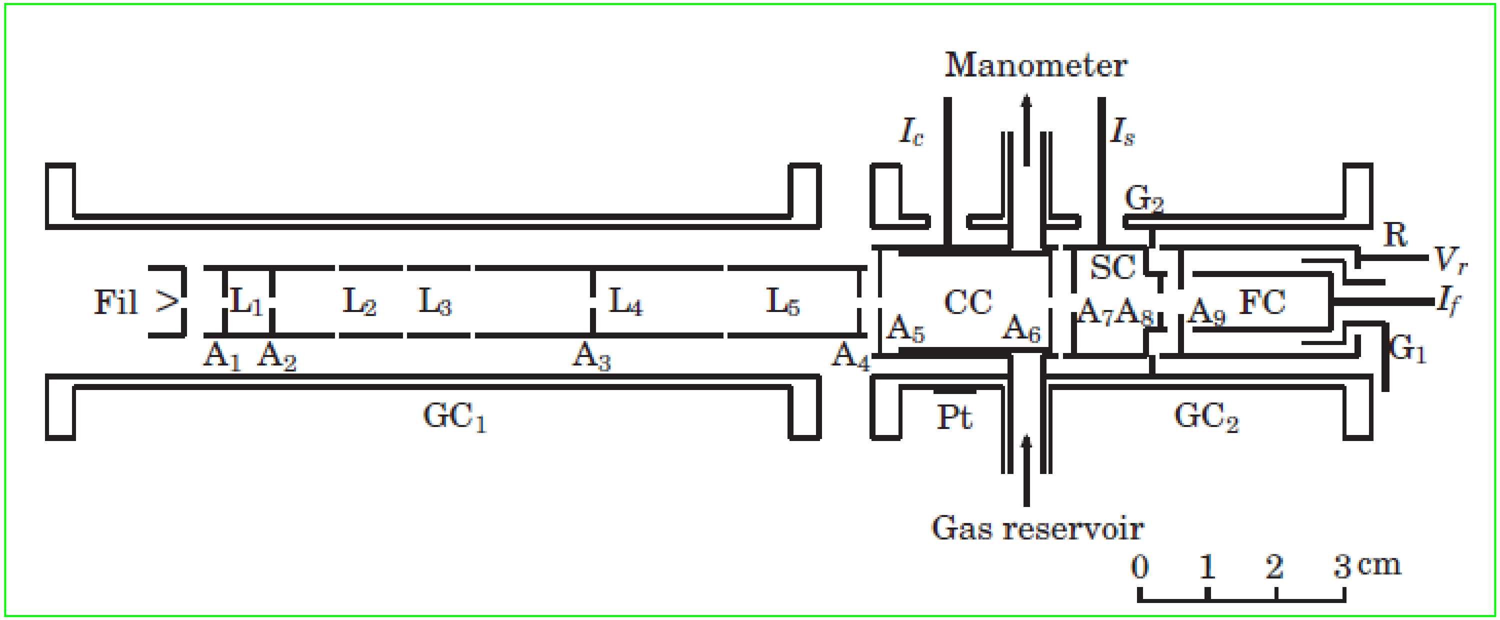
M. C. Fuss, ... G. Garcia Chem. Phys. Lett. 486 (2010) 110

Experimental methods: total

attenuation method $I = I_0 \exp(-\sigma n L)$

precision <5%, unless...



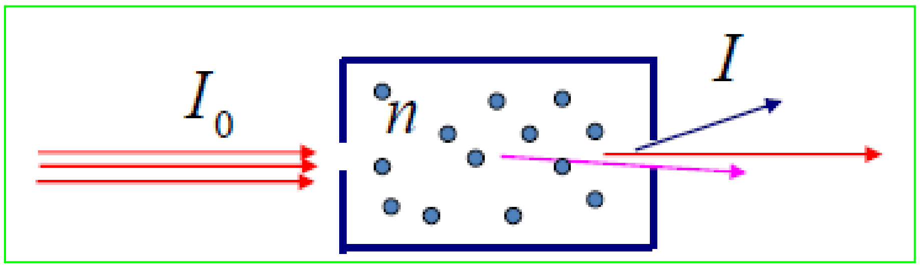


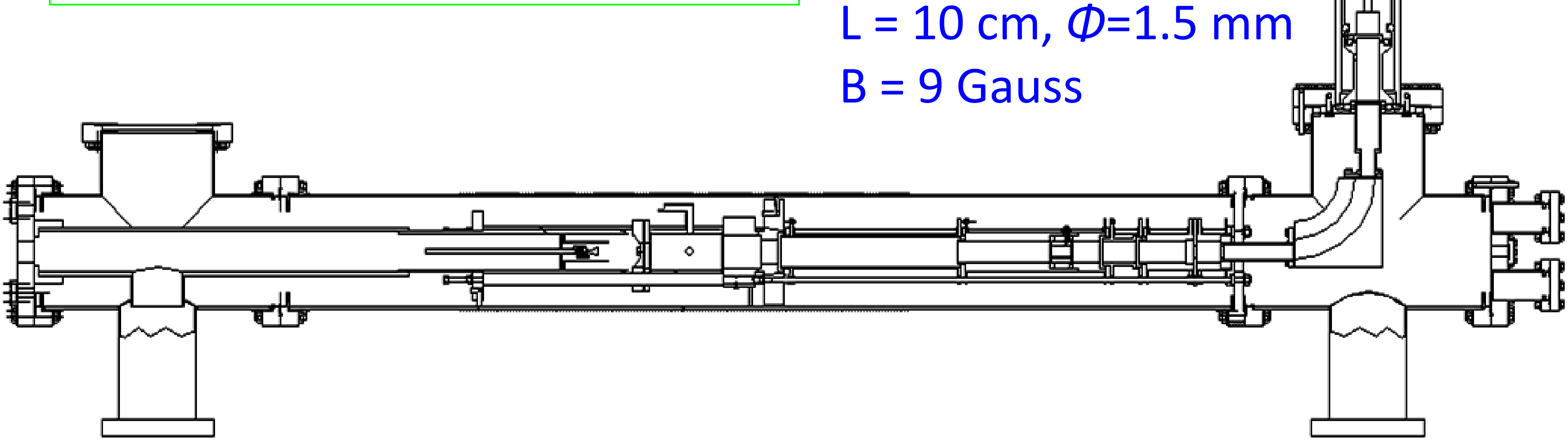
H. Nishimura et al., J.Phys. Soc. Japan **72** (2003) 1080

Experimental methods: total

Angular resolution error, leading to underestimation of TCS

→ avoid guiding magnetic field, use long scattering cells with small apertures





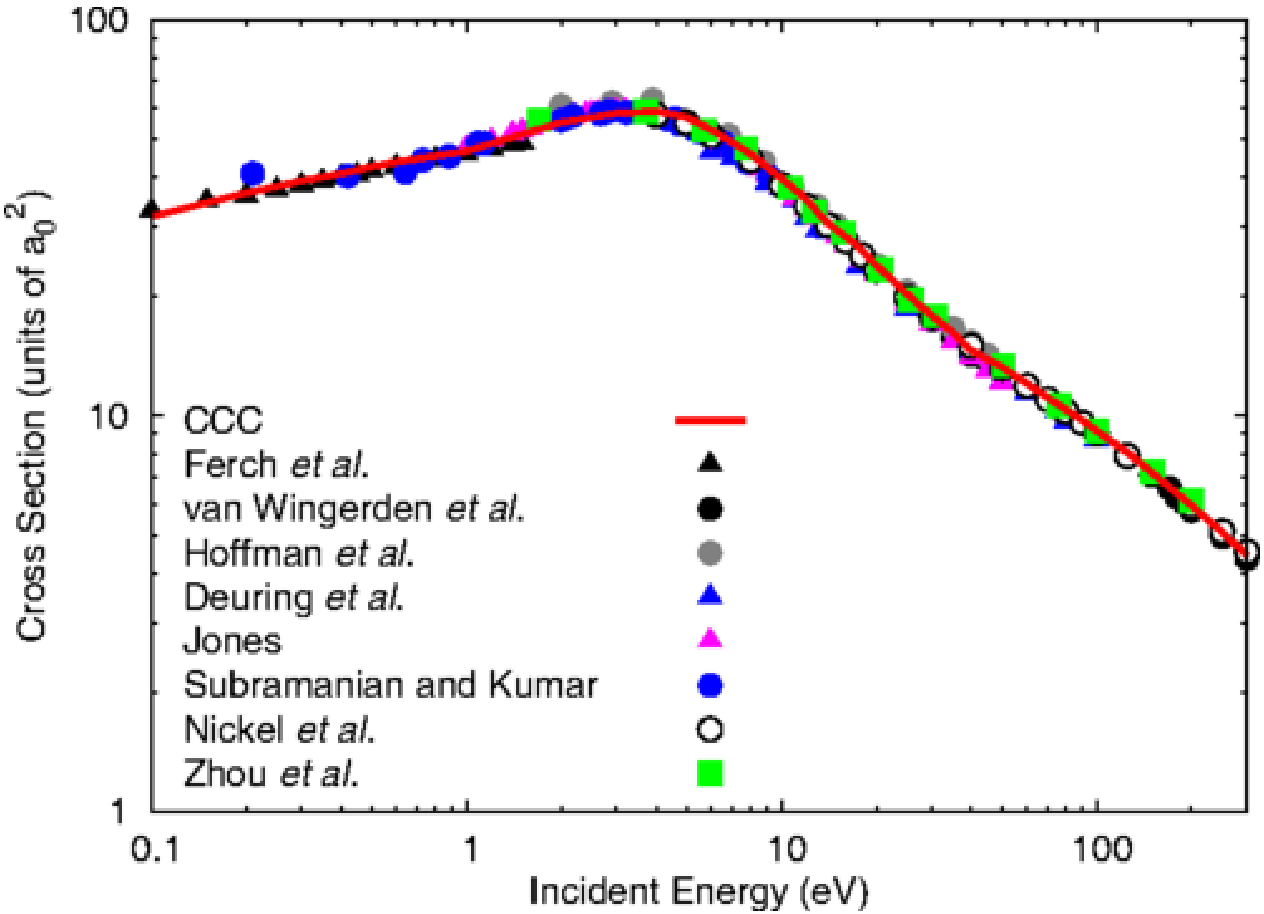
L = 10 cm, Φ=1.5 mm

B = 9 Gauss

G. Karwasz, R. S. Brusa, M. Barozzi, A.Zecca, Nucl. Instr. Meth. Phys. B 171 (2000) 178

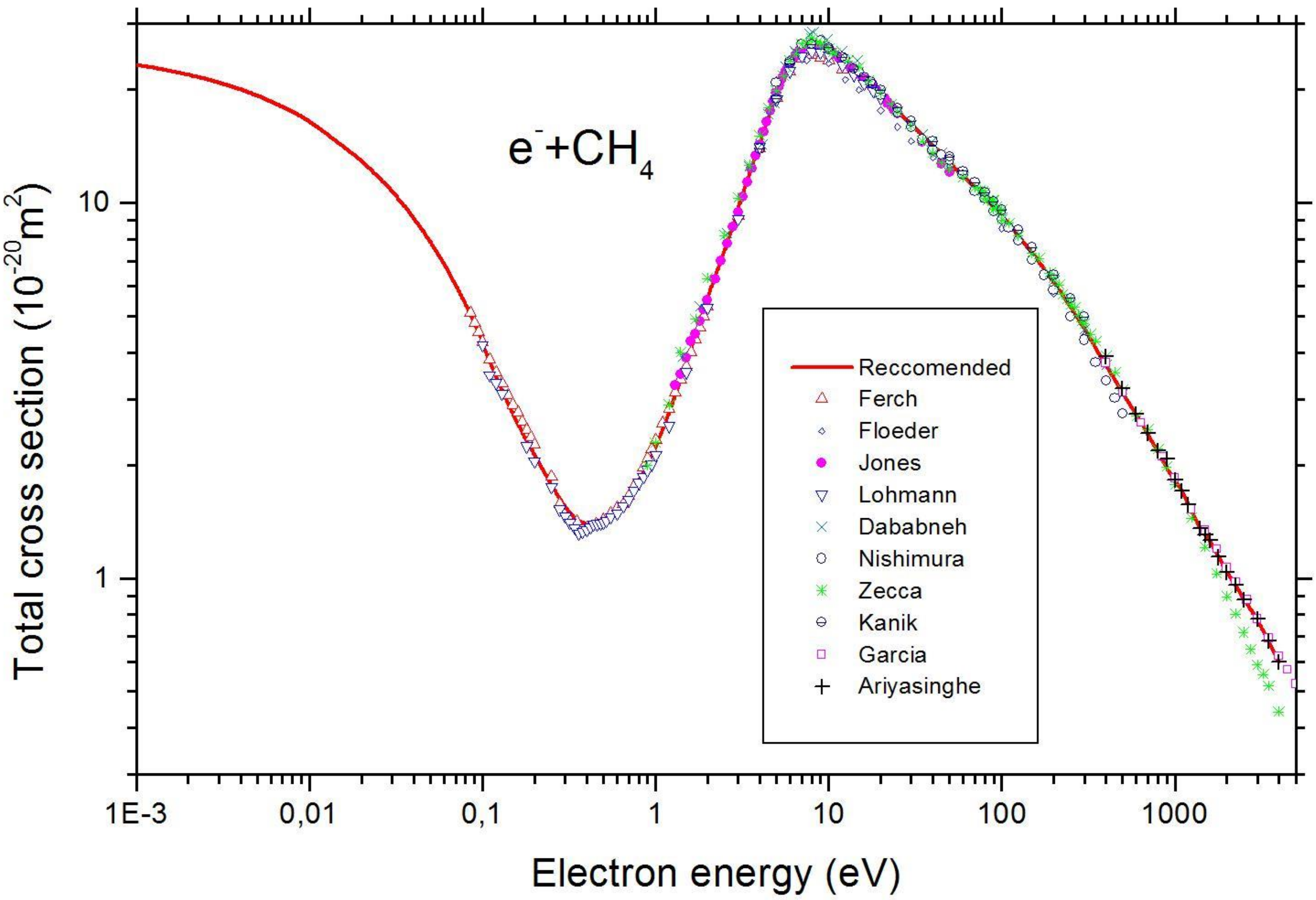
Trento 2005/ Torun 2010

Hydrogen – total: experiment vs theory



Mark C. Zammit, Jeremy S. Savage, Dmitry V. Fursa, and Igor Bray
Phys. Rev. Lett. 116, 233201
http://journals.aps.org/prl/abstract/10.1103/PhysRevLett.116.233201

Experimental methods: total



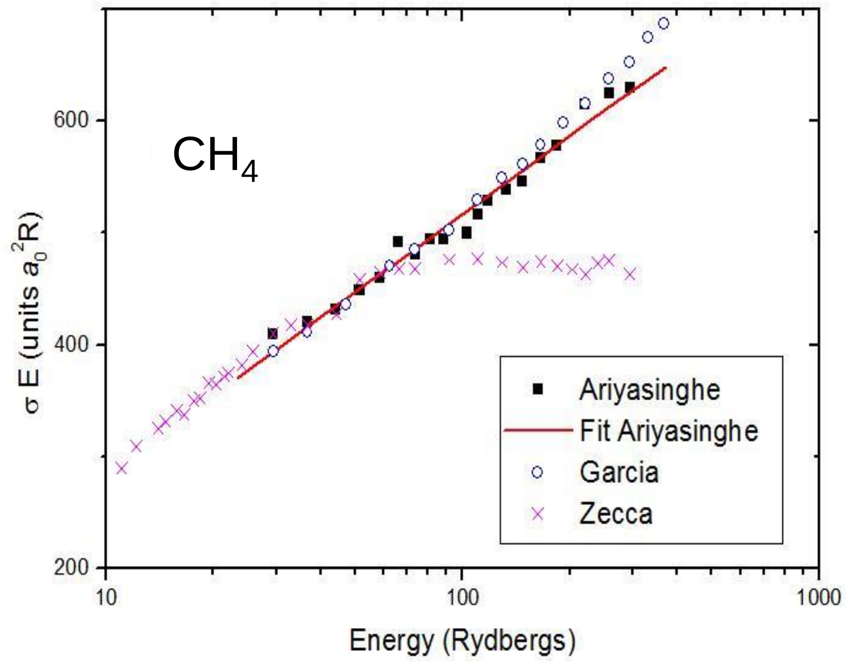
Agreement generally within ±5%

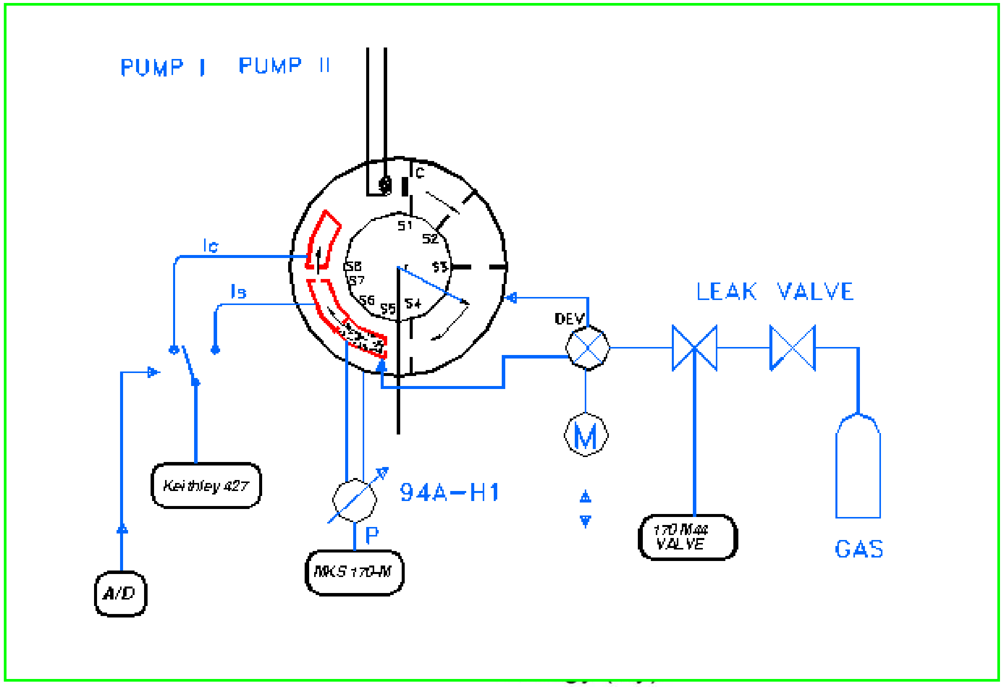
Apart from high *E* (and polar)

Mi-Young Song, Jung-Sik Yoon, Hyuck Cho, Yukikazu Itikawa, G. P. Karwasz, V. Kokoouline, Yoshiharu Nakamura, Jonathan Tennyson, J. Phys. Chem. Ref. Data, 44 (2015) 023101

Total @ high energies: Born-Bethe fit

$\sigma(E) = A + B \ln E$



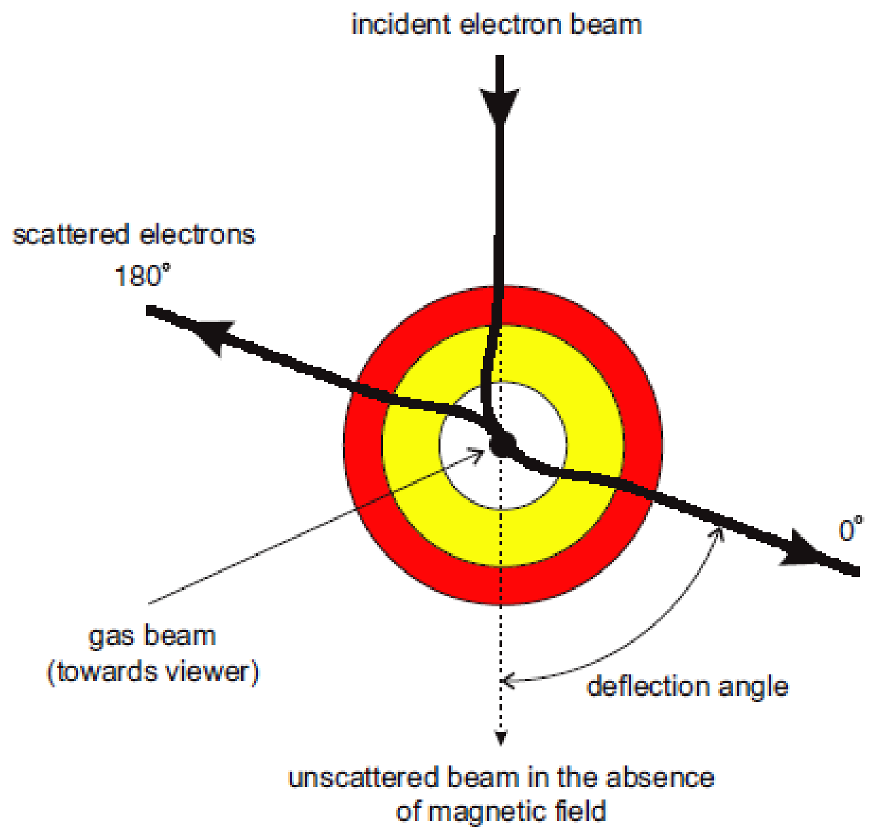


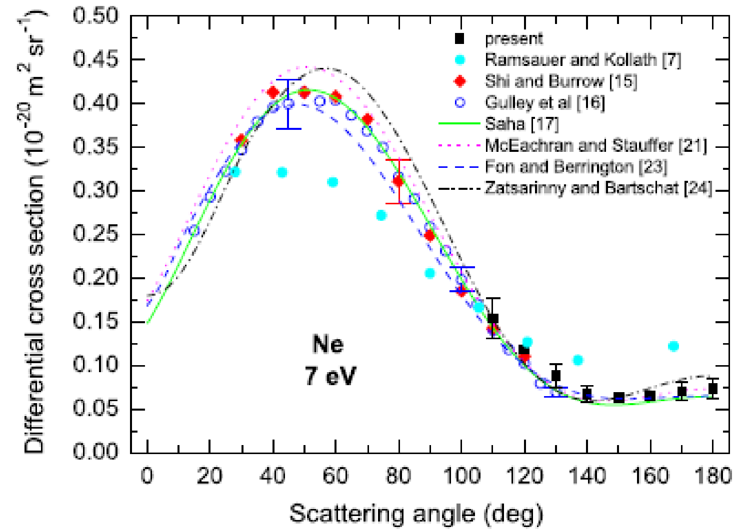
„In the high energy limit present (GK, Zecca) measurements are affected by angular resolution error. In order to evaluate it, differential cross sections at low angles would be needed. A rough evaluation from Born approximation for the *elastic* channel gives an error of a few percent. Note that the error in TCS can be higher, as Trento apparatus does not perform discrimination against inelastically forward-scattered electrons.

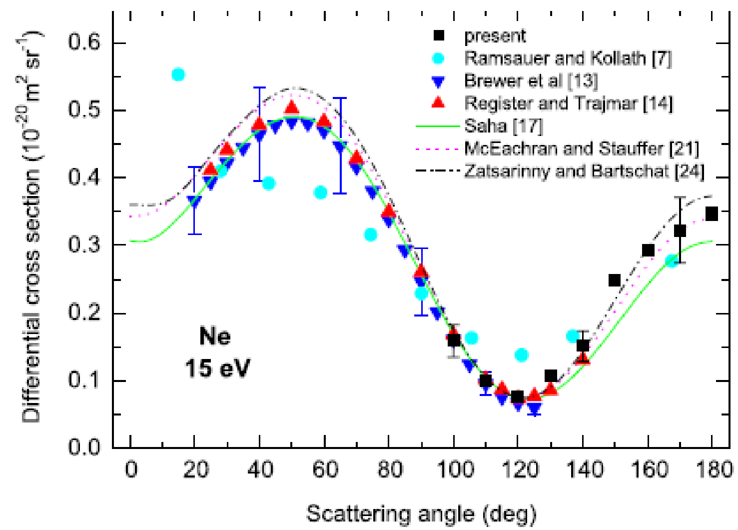
Fig.4. Born-Bethe fit $(\sigma/a_o^2)\,(E/\mathrm{R}) = A + B \ln(E/\mathrm{R})$ to TCS from Ariysainghe: A=52.31±17.3, B=232.2±8.6 where Rydberg constant is R=13.6 eV and the cross sections is expressed in atomic units $a_0^2 = 0.28\times10^{-20}\mathrm{m}^2$

Experimental methods: elastic

PHYSICAL REVIEW A 74, 042701 (2006)

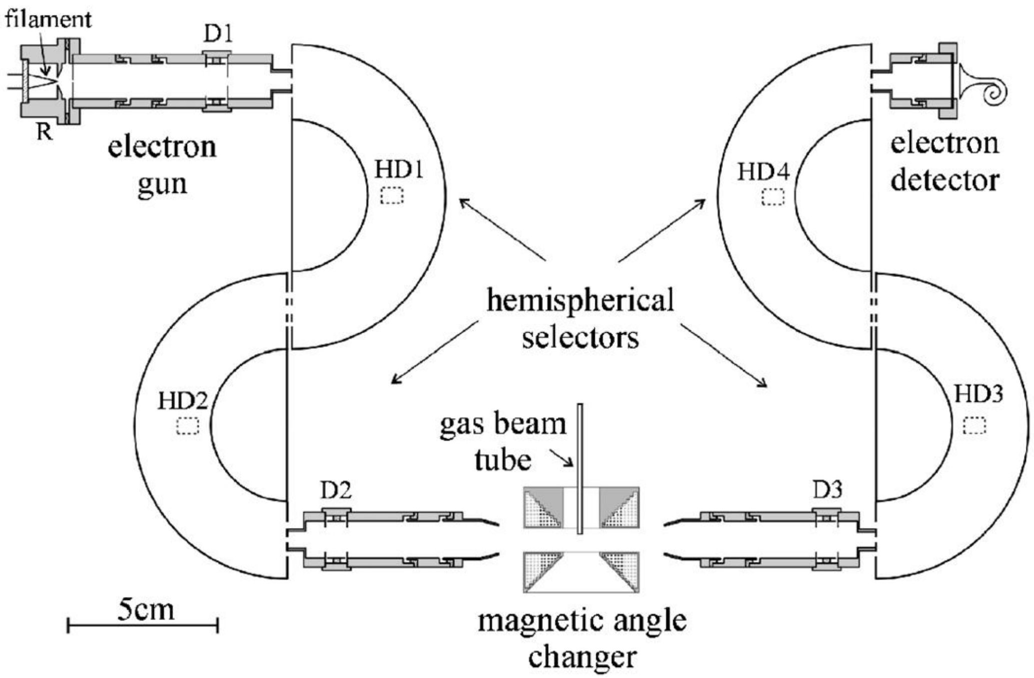


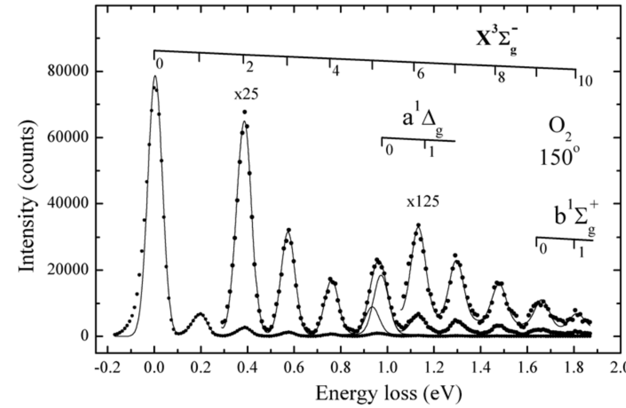


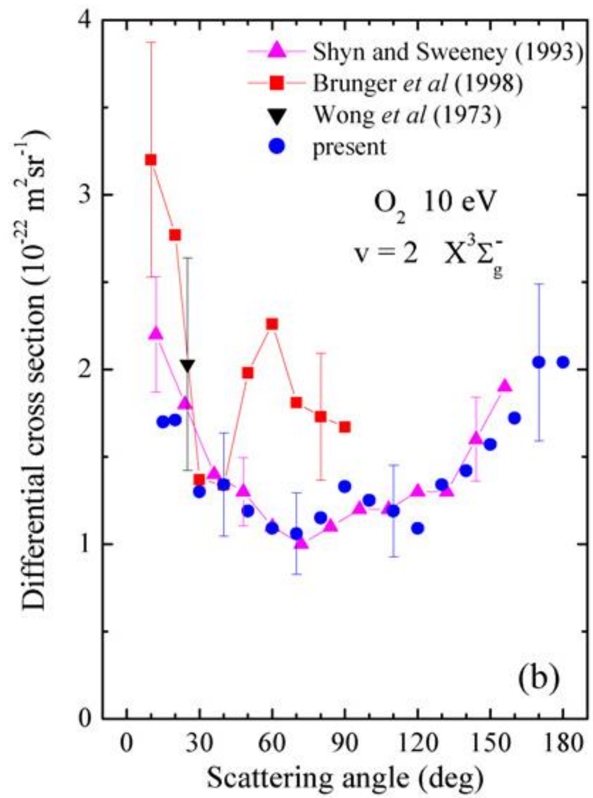


I. Linert, B. Mielewska, G. King, and M. Zubek, PRA (2006)

Experimental methods: excitation (electronic, vibrational)







accuracy ±20-40%

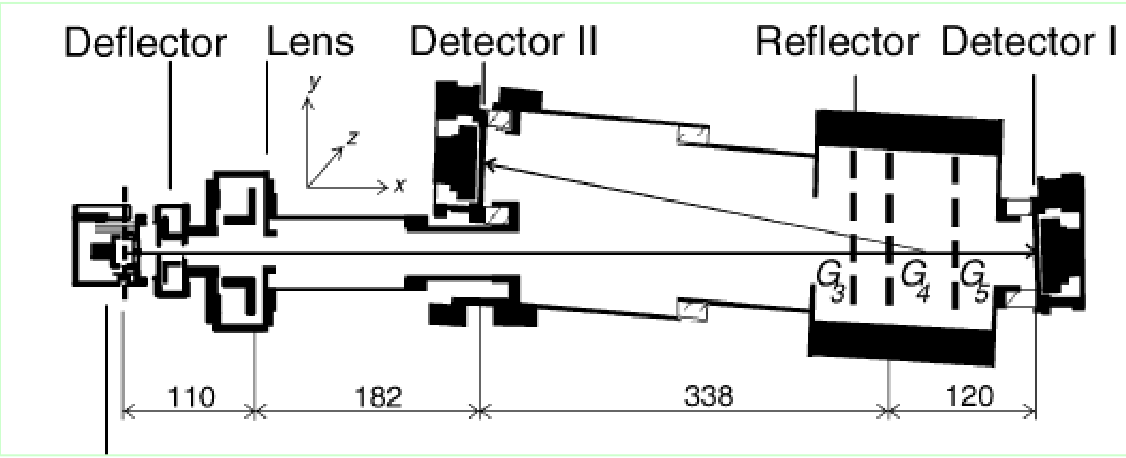
Experiments by:

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

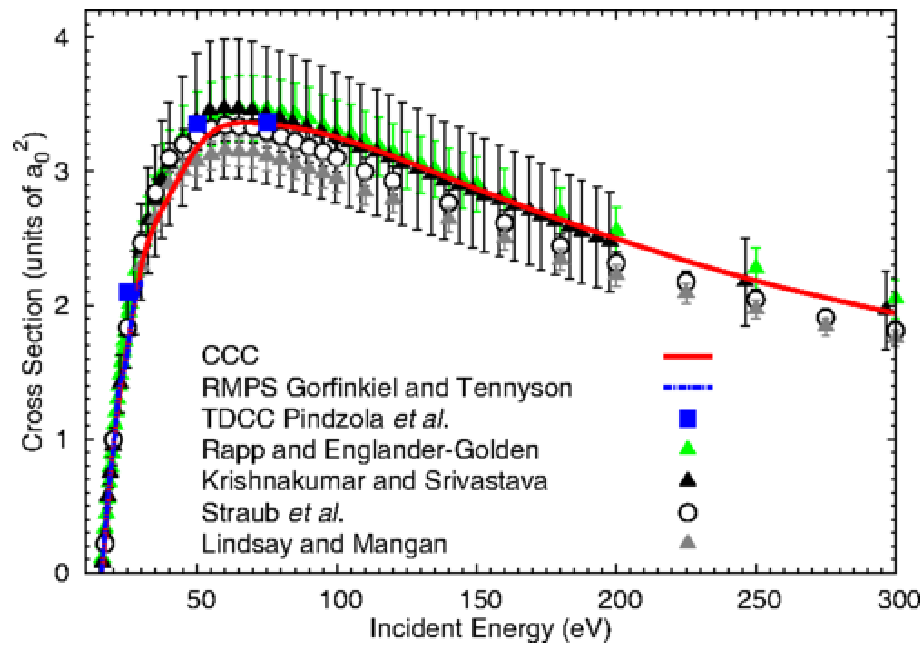
M. Khakoo et al. (Fullerton California)

M. Allan (Freiburg University)/ J. Fedor (Prague)

Experimental methods: ionization, total

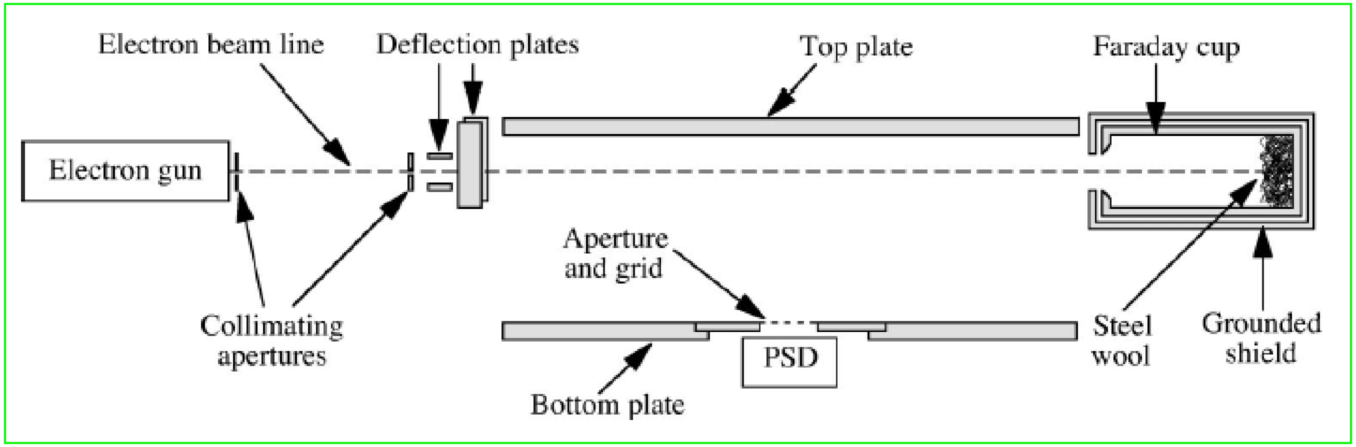


Accuracy: ±10-15%

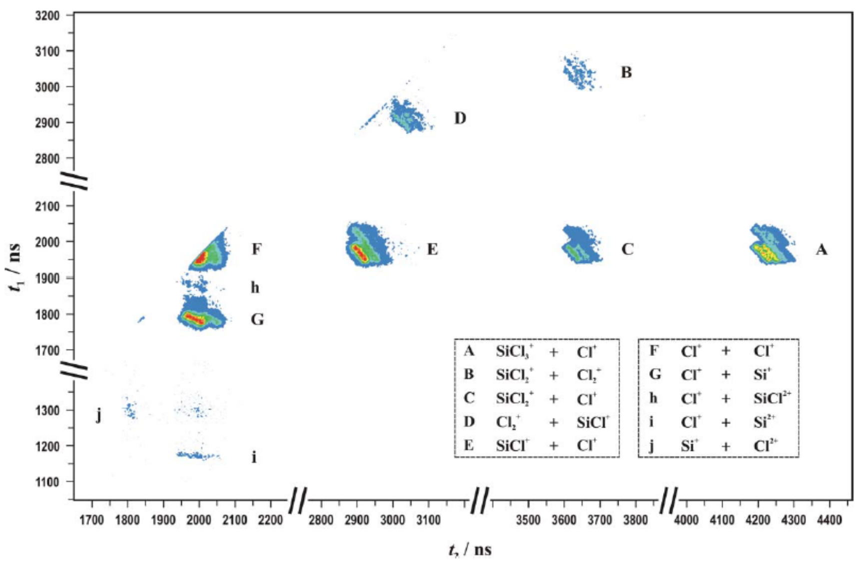


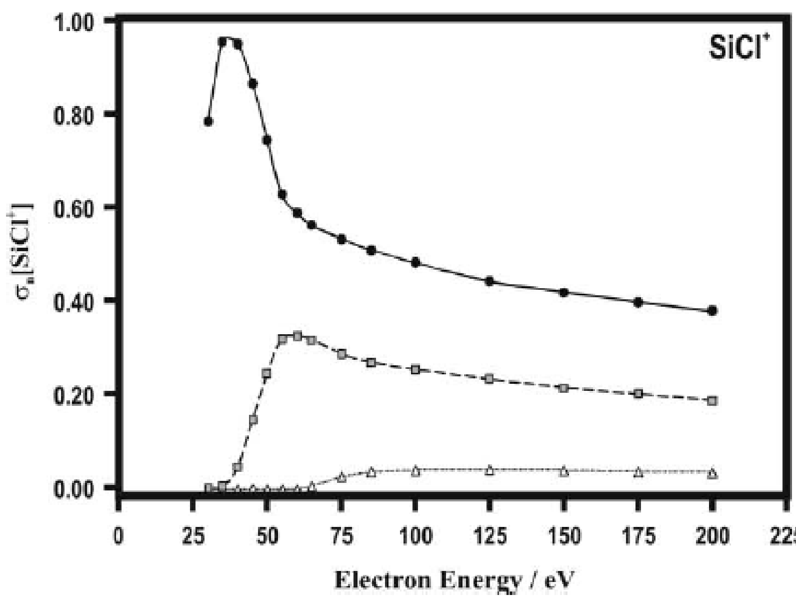
Zammit et al.. PRL 2016

Experimental methods: ionization (2)



Accuracy: ±15-20%

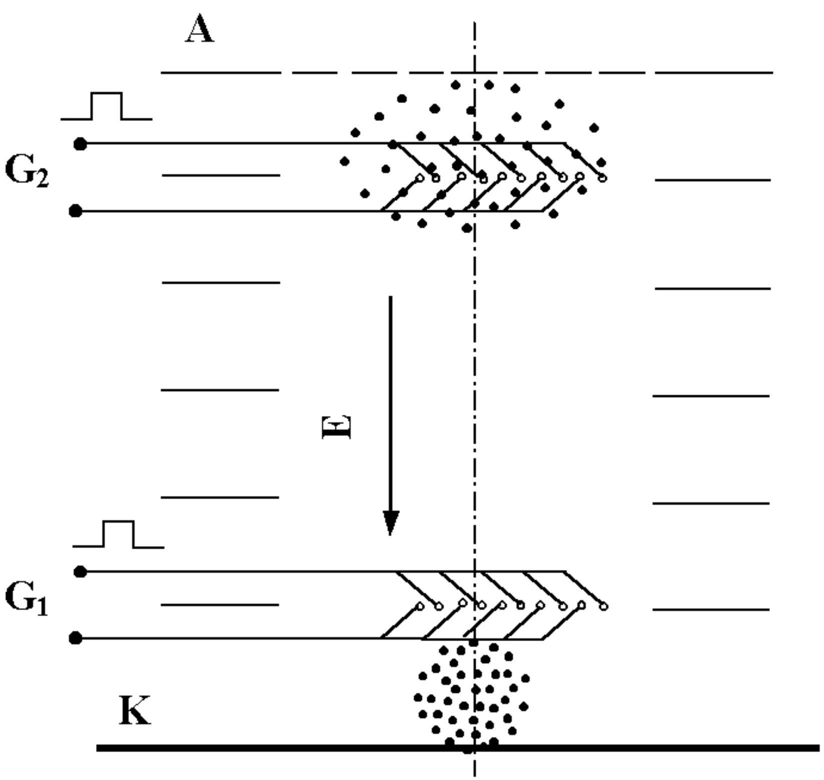




B. G. Lindsay et al., JCP **129** (2004), S J King nad S D Price, JCP**134** (2011) 074311

Diffusion coefficients → electronic distribution function n_e (***r***, ***v***, *t*)

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



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

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

Accuracy: ±5-10%

Non-unique modelling

W. Roznerski (+), J. Mechlinska-Drewko (+), Y. Nakamura

Hydrogen – electronic excitation (modelling)

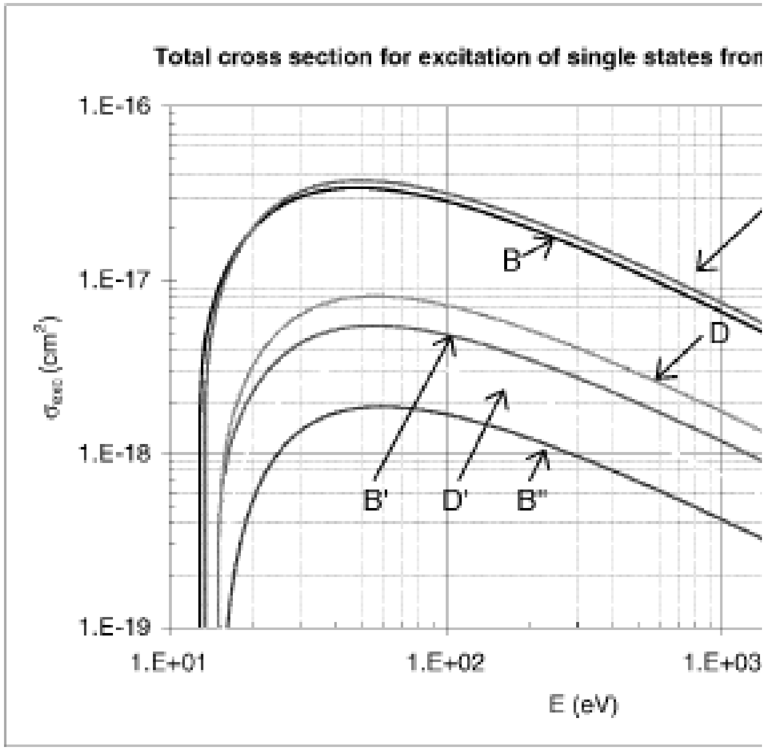


Figure 22: Total electron impact excitation cross sectio singlet electronic states of H_2 from the $H_2(X^1\Sigma_g^+; v =$

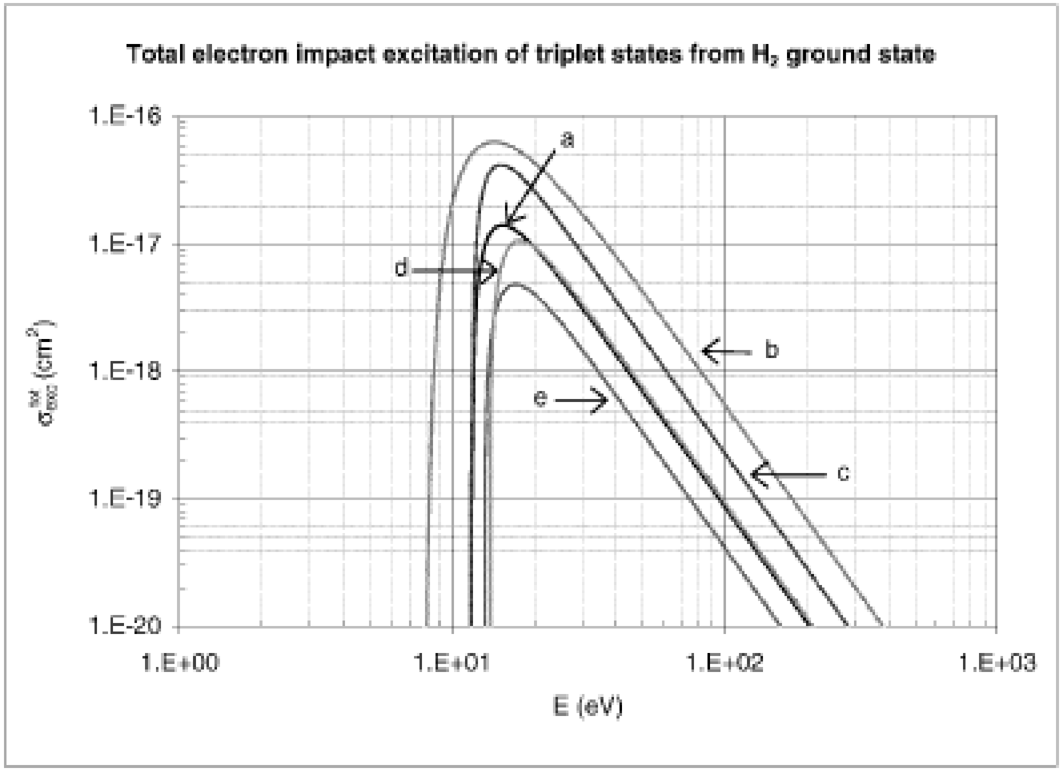


Figure 25: Total electron impact excitation cross section of a,b,c,d and e triplet states of H_2 from its $(X^1\Sigma_g^+; v = 0)$ ground state, Eq. (95).

Collision Processes in Low-Temperature Hydrogen Plasmas

Jül-4105
Dezember 2003
ISSN 0944-2952

Ratko K. Janev, Detlev Reiter, Ulrich Samm

Hydrogen – electronic excitation (2010)

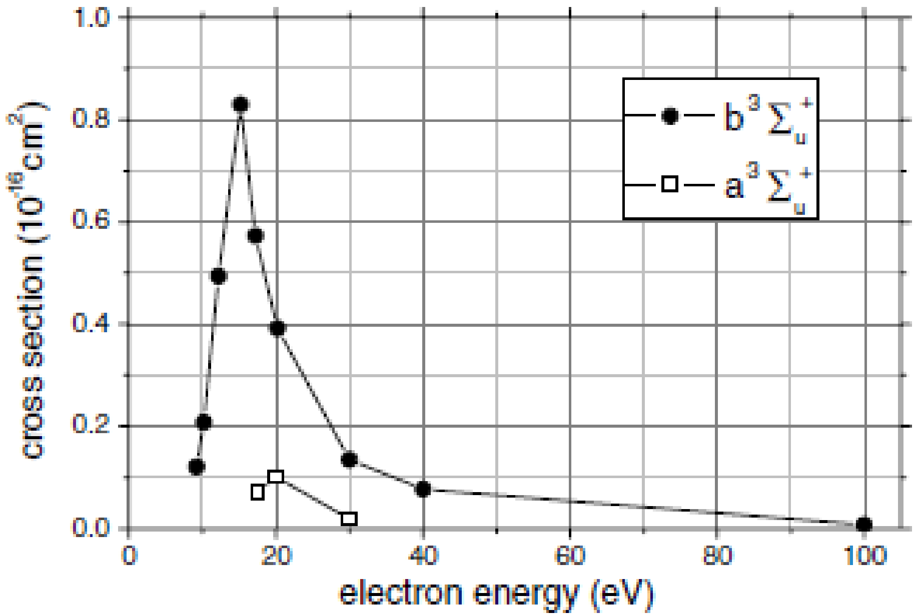


Figure 7. The electronic excitation cross sections for $b\,^3\Sigma_u^+$ and $a\,^3\Sigma_g^+$ states of H_2 as given by Yoon *et al* [36]. These cross sections

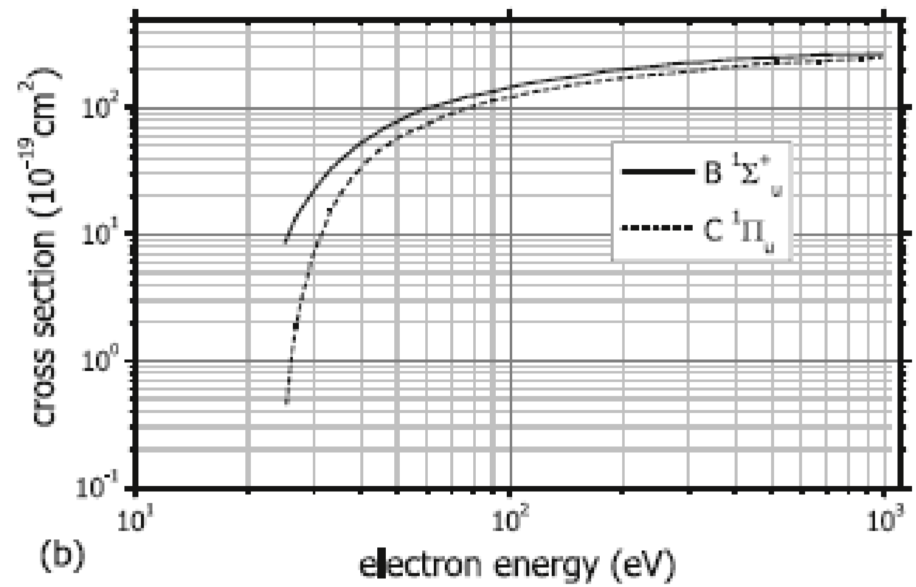


Figure 8. (*a*) Electron-impact excitation cross sections of Lyman and Werner bands of D_2. Solid curve: Lyman bands; dots: Werner bands. (*b*) Log–log plot for cross sections of $B\,^1\Sigma_u^+$ (solid) and $C\,^1\Pi_u$ (dots) in the threshold energy region [53].

Jung-Sik Yoon[1], Young-Woo Kim[1], Deuk-Chul Kwon[1], Mi-Young Song[1], Won-Seok Chang[1], Chang-Geun Kim[2], Vijay Kumar[3] and BongJu Lee[1]

Rep. Prog. Phys. **73** (2010) 116401

Jung-Sik Yoon, Mi-Young Song, Jeong-Min Han, Sung Ha Hwang, Won-Seok Chang, and BongJu Lee

Journal of Physical and Chemical Reference Data Volume 37, Issue 2 > 10.1063/1.2838023

Electronic excitation – deconvolutions of spectra (H_2)

J Wrkich *et al*

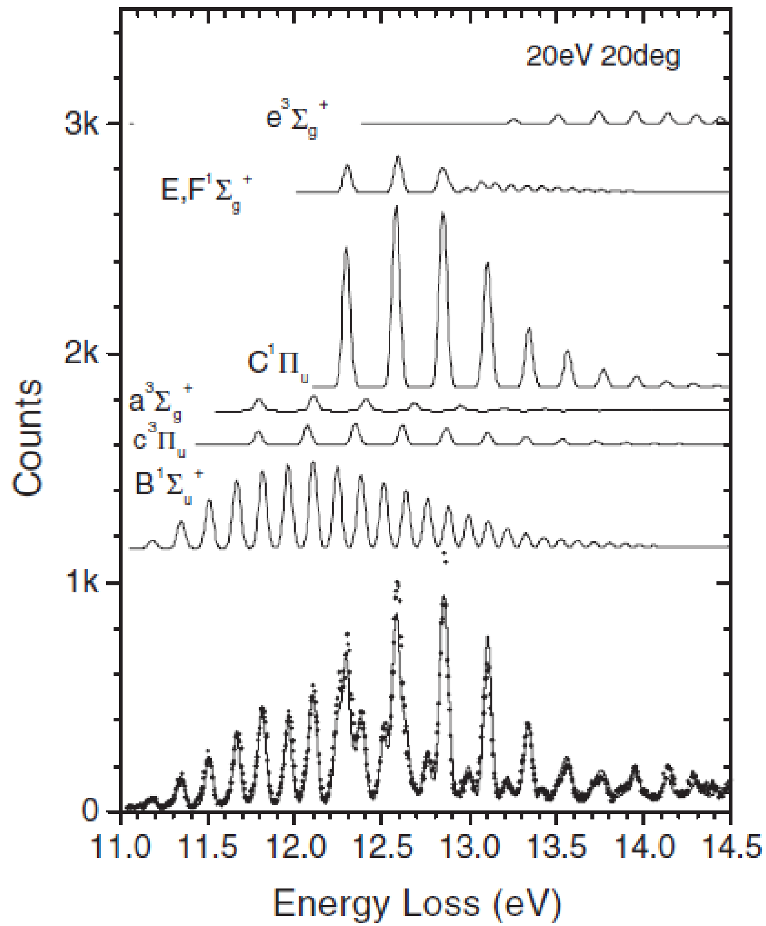
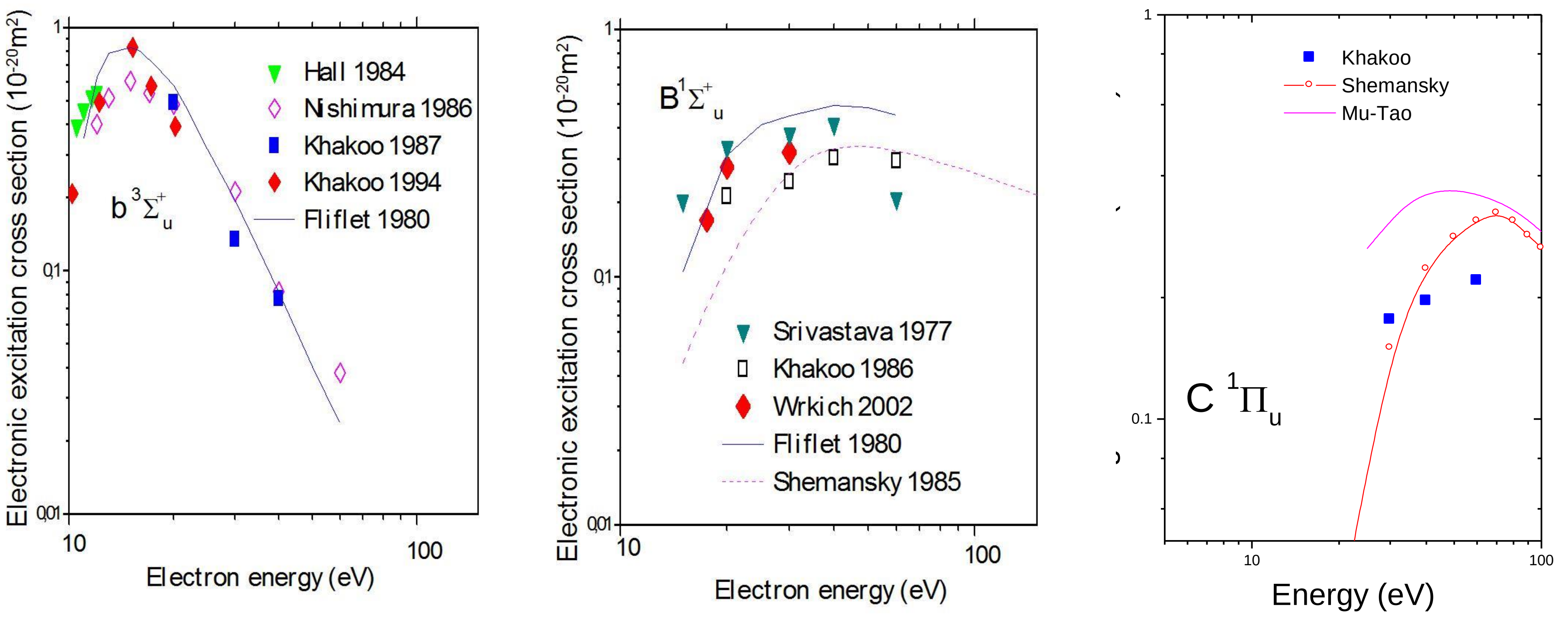


Figure 1. Experimental electron energy loss spectrum of H_2 (dots) taken at $E_0 = 20$ eV and $\theta = 20°$ with a fitted spectrum (curve) and showing the positions of the unfolded vibrational manifolds as a result of the fitting.

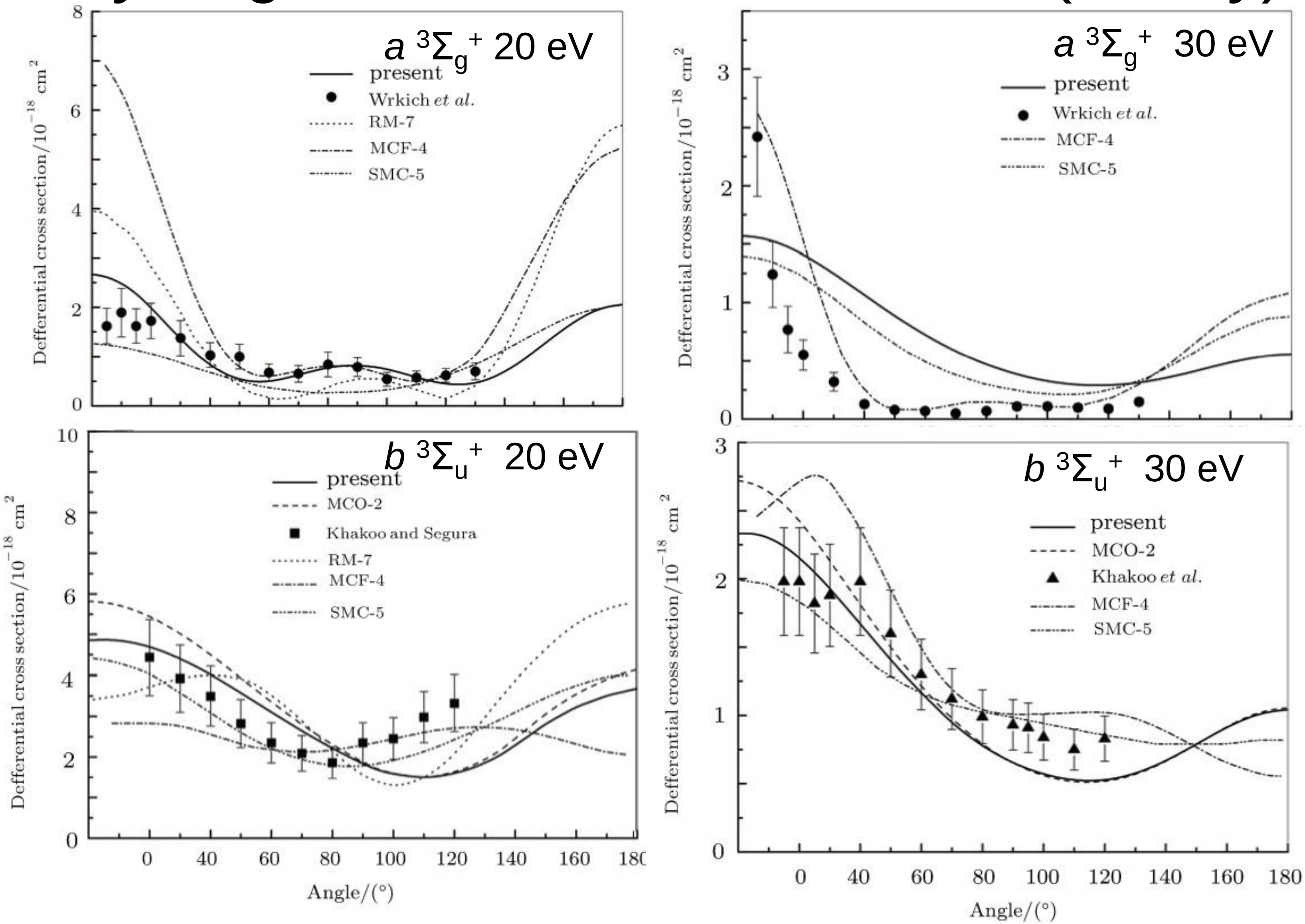
Wrkich *et al.* JPB (2002)
Fulletron University (CA)

Electronic excitation – dipole allowed and forbidden in H_2

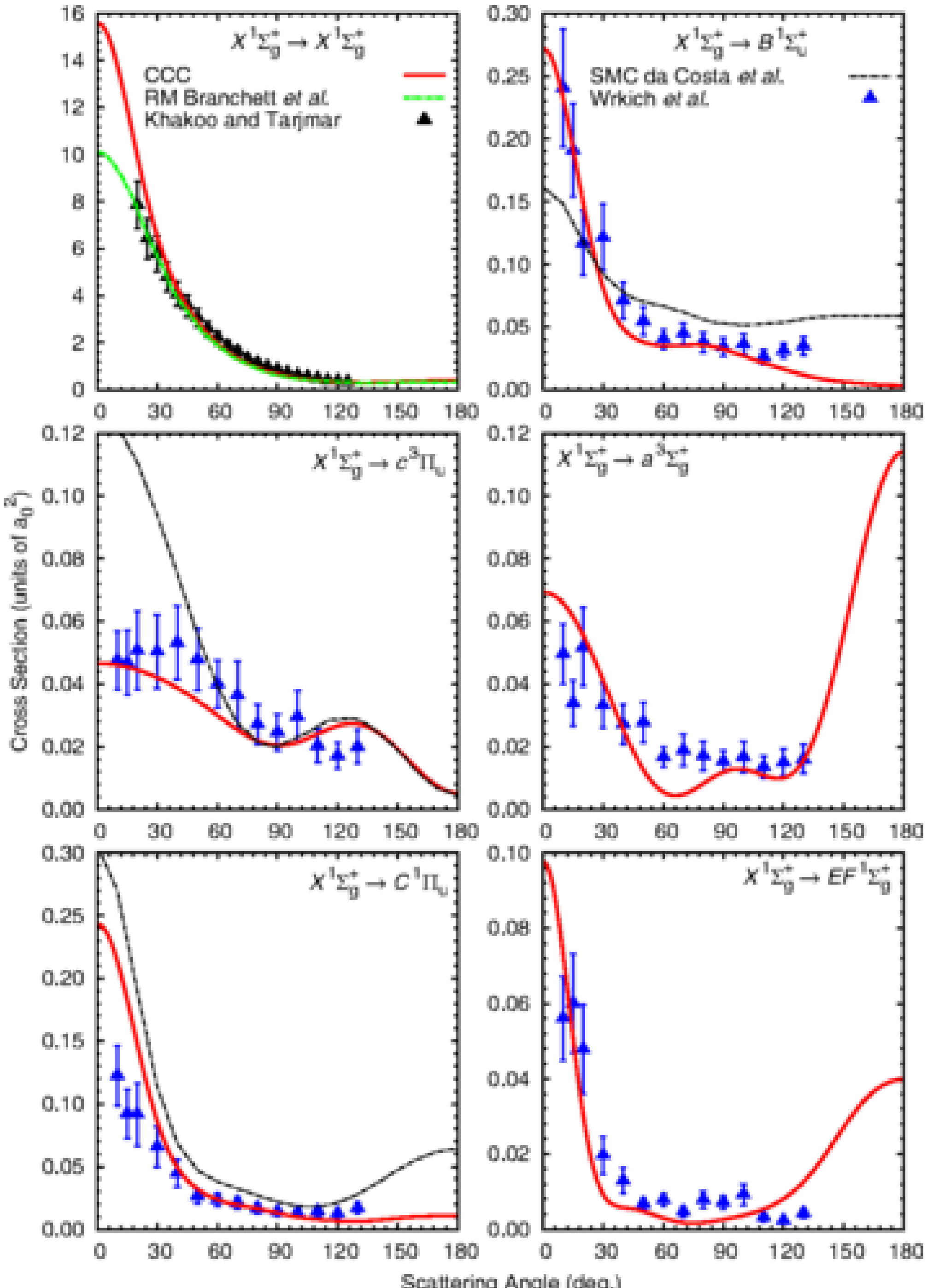


A. Zecca, G. Karwasz, R.S. Brusa, Nuovo Cimento (1996) updated

Hydrogen – electronic excitation (theory)



Wang Yuan-Cheng, Ma Jia, Zhou Ya-Jun, Momentum-space multichannel optical model
Chinese Physics B, 2016, 25(4): 043401



Hydrogen – a complete set of electronic excitations (singlet and triplet states)

Perfect agreement, i.e. within experimental total uncertainties

Zammit et al. PRL 2016

Nitrogen – electronic excitation

J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 225202 C P Malone *et al*

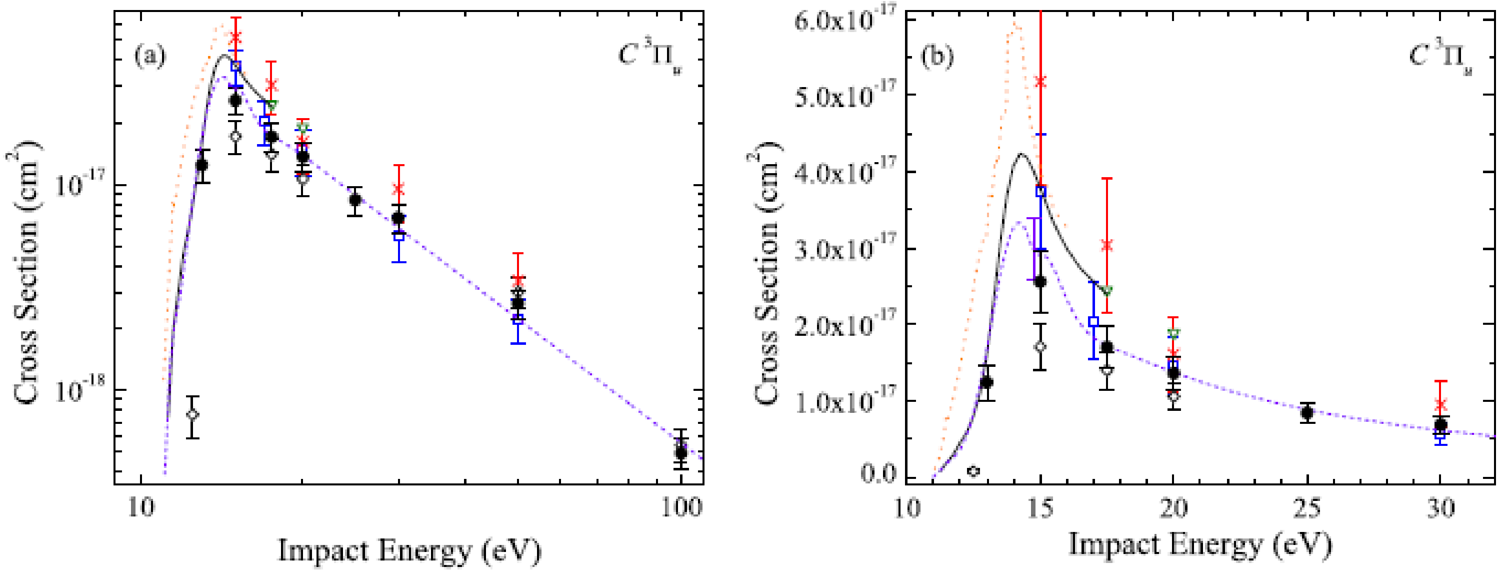


Figure 2. ICSs for electron-impact excitation of the $C\,^3\Pi_u$ state of N_2. The plots illustrate (a) shape and (b) magnitude. Black solid circles, present data; black open diamonds, Johnson *et al* [1]; blue open squares, Trajmar *et al* [13]; green inverted open triangles, Zubek and King [4]; red crosses, Campbell *et al* [11]; black solid line, Zubek [27]; purple dashed line, Shemansky *et al* [10] (see the text); red dotted line, Poparić *et al* [28].

Good agreement between experiments, few theories

C.P. Malone *et al.* J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 225202

Nitrogen – electronic excitation

J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 225202 C P Malone *et al*

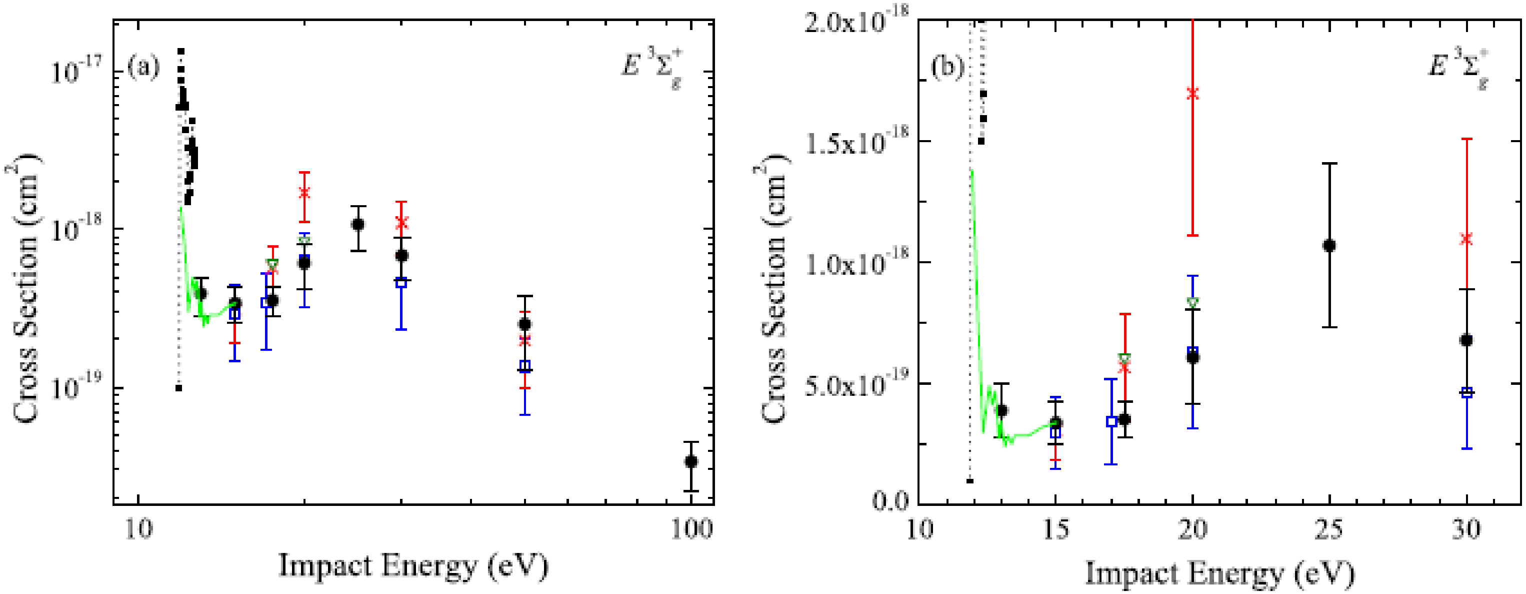


Figure 3. ICSs for electron-impact excitation of the $E\,^3\Sigma_g^+$ state of N_2. Black solid circles, present data; blue open squares, Trajmar *et al* [13]; green inverted open triangles, Zubek and King [4]; red crosses, Campbell *et al* [11]; light green solid line, Zubek [26]; black solid squares with dotted line, Brunger *et al* [25].

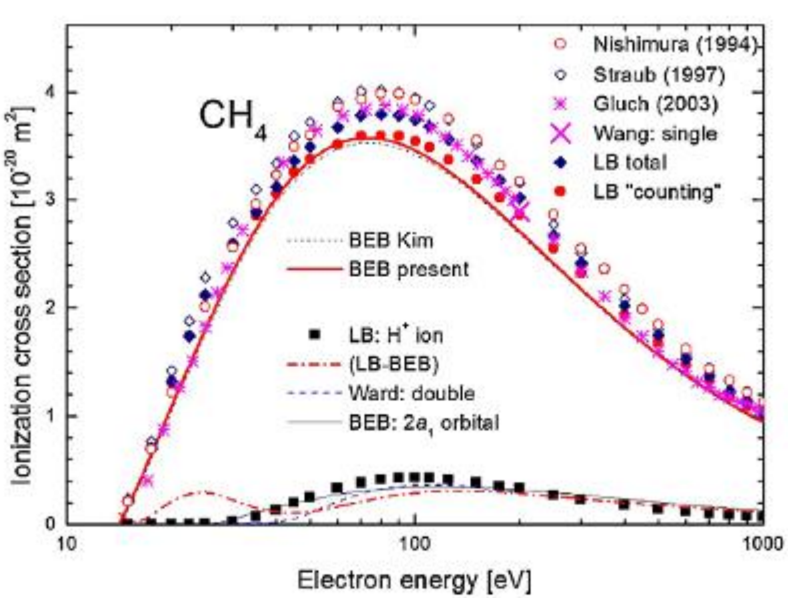
Qualitative agreement between experiments, few theories

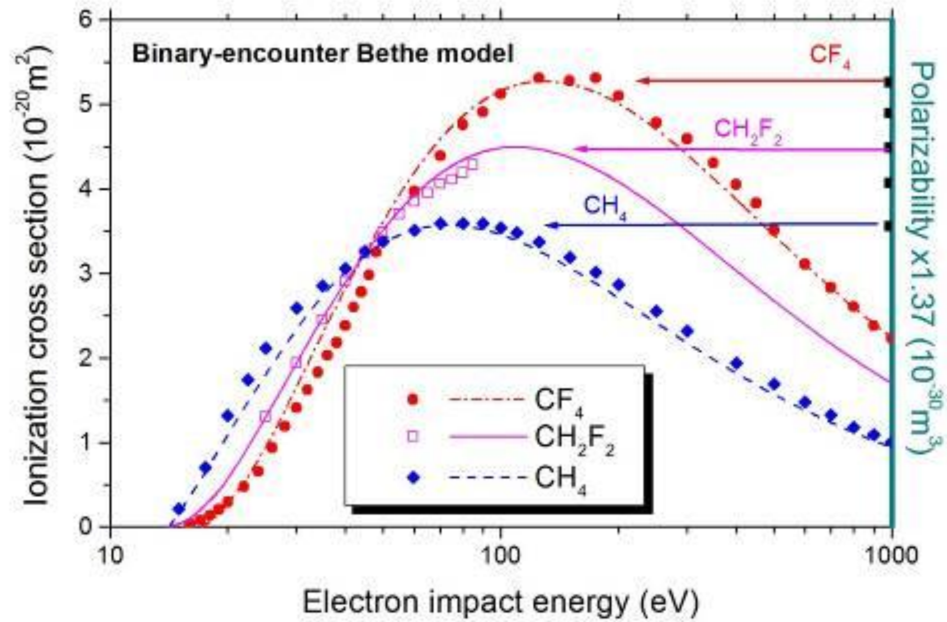
C.P. Malone *et al.* J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 225202

Ionization: semiempirical formulae

$$\sigma[\mathrm{cm}^2] = \frac{10^{-13}}{IE}\left\{A_1 \ln(E/I) + \sum_{i=2}^{N} A_i \left(1 - \frac{I}{E}\right)^{i-1}\right\},$$

R. K. Janev, D. Reiter, Phys. Plasmas 9, 4071 (2002);





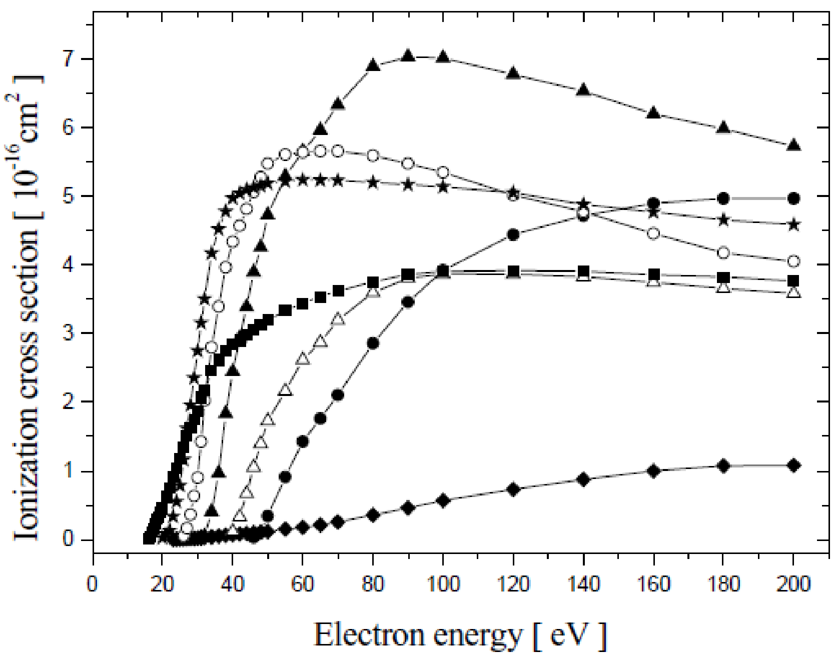
$$\sigma = \sum_n 4\pi a_0^2 \xi_n \left(\frac{R}{I_n}\right)^2 \frac{1}{t + u_n + 1}\left\{1 - \frac{1}{t} + \frac{\ln t}{2}\left(1 - \frac{1}{t^2}\right) - \frac{\ln t}{t+1}\right\}$$

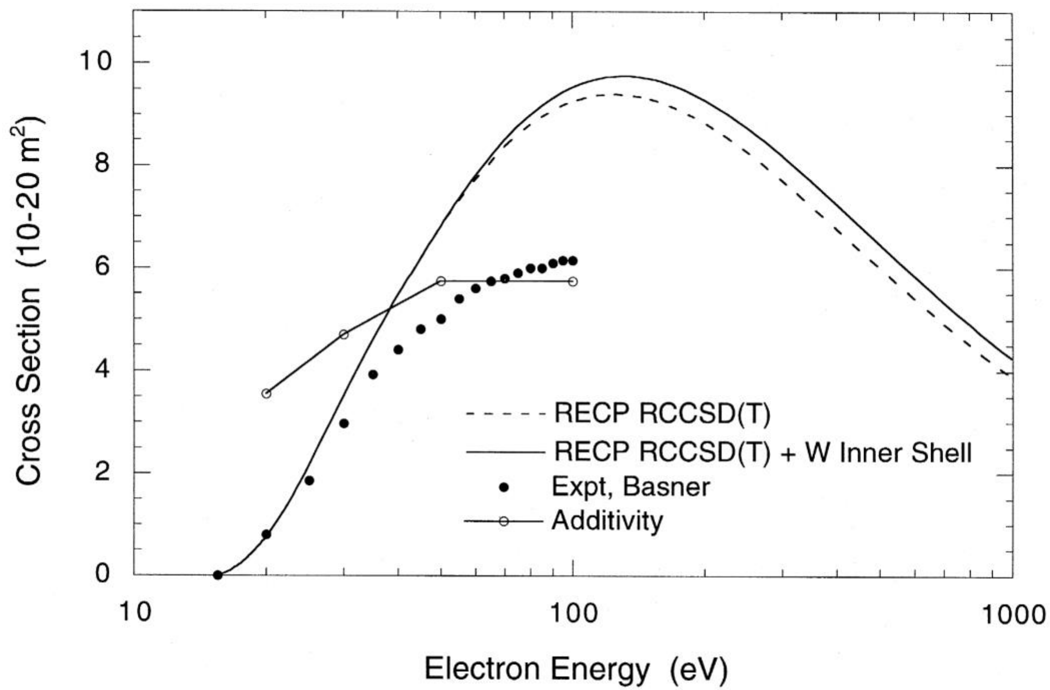
Normalized energies: $t = E/I_n$, $u_n = E_{kin}/I_n$ Only two values needed from QCh

Y.-K. Kim and M. E. Rudd, Phys. Rev. A 50 (1994) 3954
G. Karwasz, P. Mozejko, M.-Y. Song, Int. J. Mass Spectrometry (2014)

Partial (and total) ionization: WF_6

■ WF_5^+; ⋆ WF_4^+ (x10); o WF_3^+ (x10); ▲ WF_2^+ (x10)

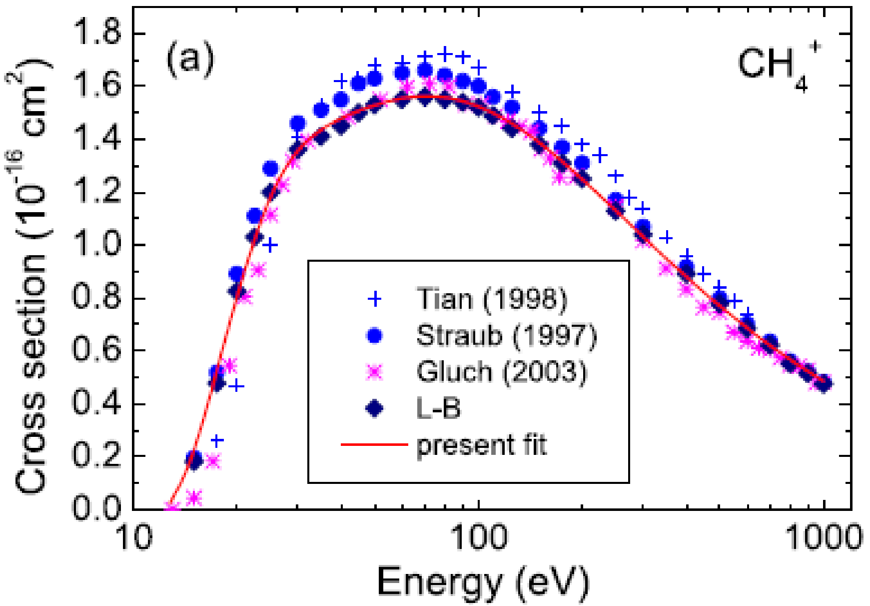




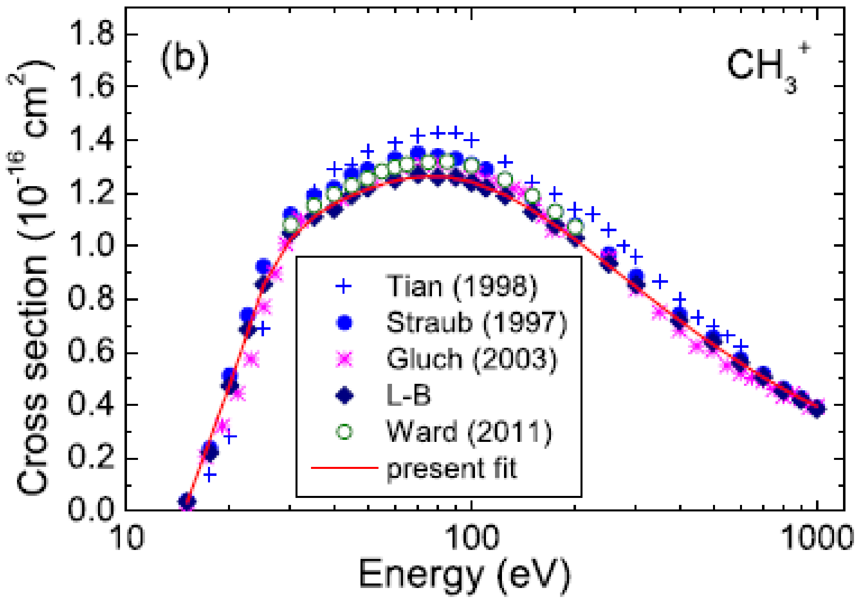
Total ionization in **serious** (50%) disagreement with relativistic BEB

R. Basner, M. Schmidt, K. Becker, Int. J. Mass Spectr. 233 (2004) 25
W.M. Huo, Y.-K. Kim, Chemical Physics Letters 319 (2000) 576–586

Partial ionization: CH_4

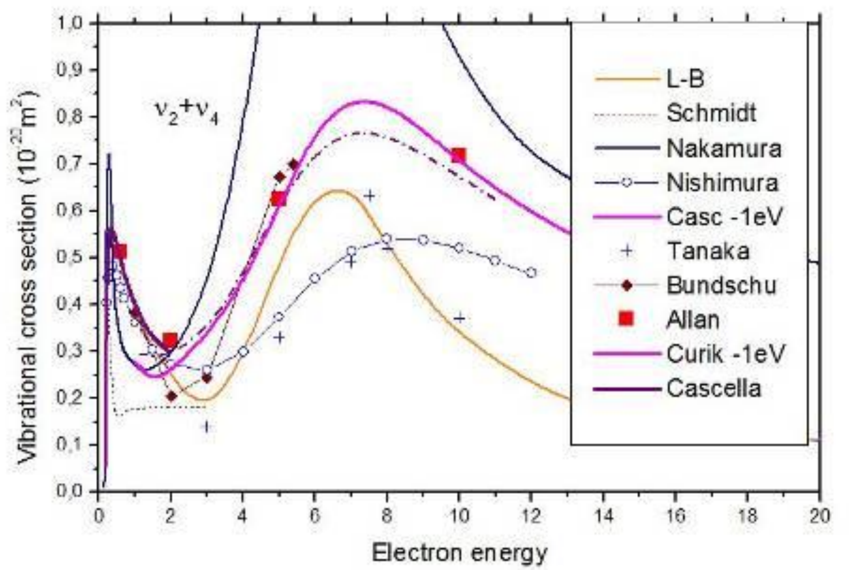


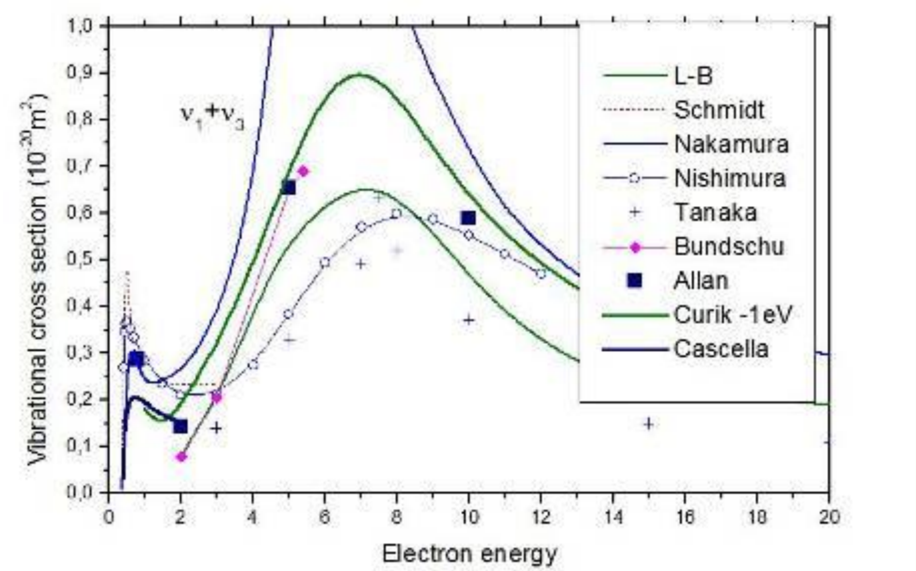
Agreement within 15-20%; unless some cases, like H^+ ions



Mi-Young Song *et al*.. JPCRD 2015

Vibrational: resonant scattering in CH_4

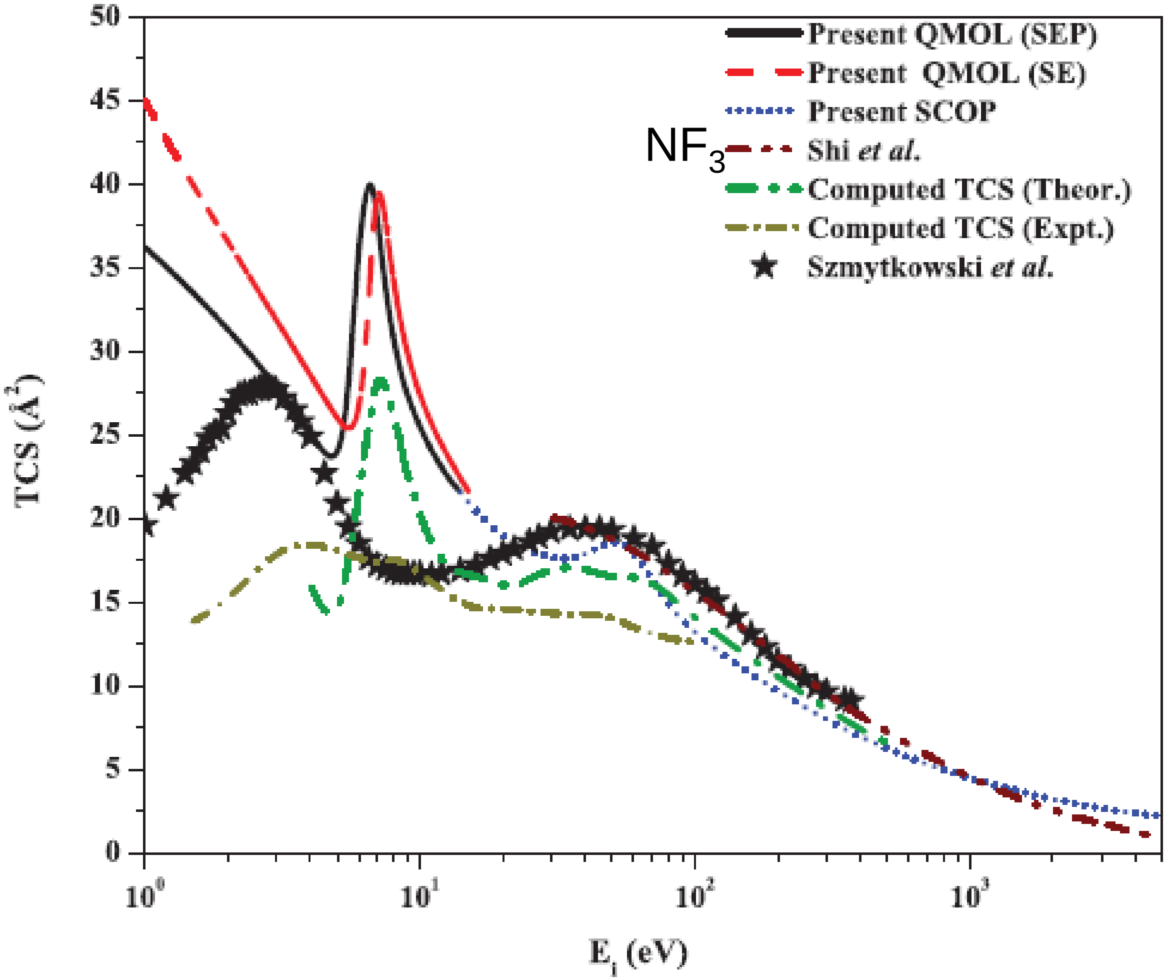




Serious (by few folds) disagreement between swarm-derived, beam-measured and theoretical values

M.-Y. Song, J. S. Yoon, H. Cho, Y. Itikawa, G. Karwasz, V. Kukooulin, Y. Nakamura, J. Tennyson, JPCRD 2015

„Shape” resonances: experiment vs. theory (NF_3)

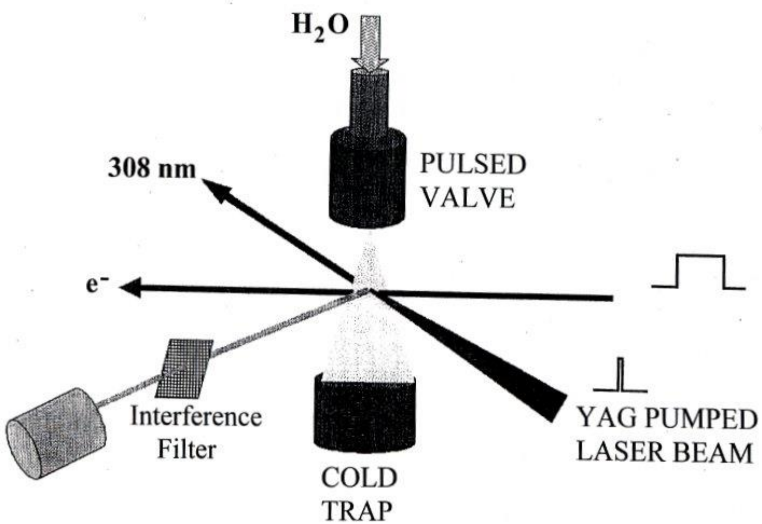


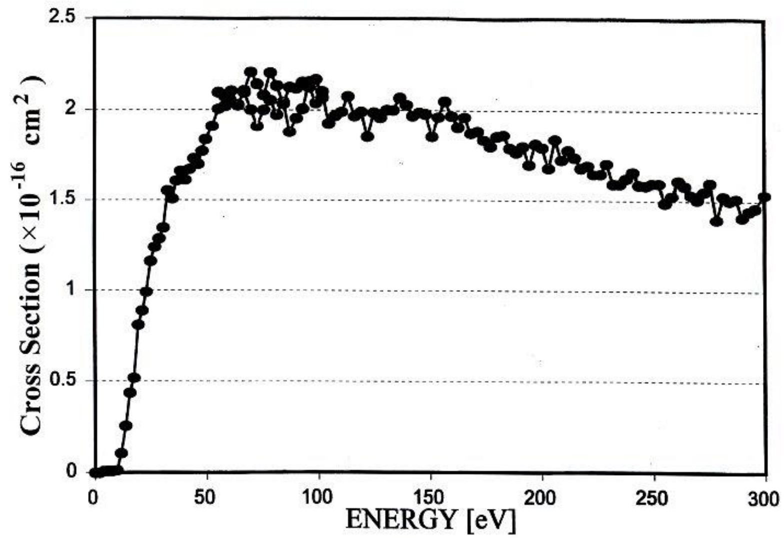
Calculations do not yield XS for resonant vibrational excitation (which is essentially unknown due to lack of experiments)

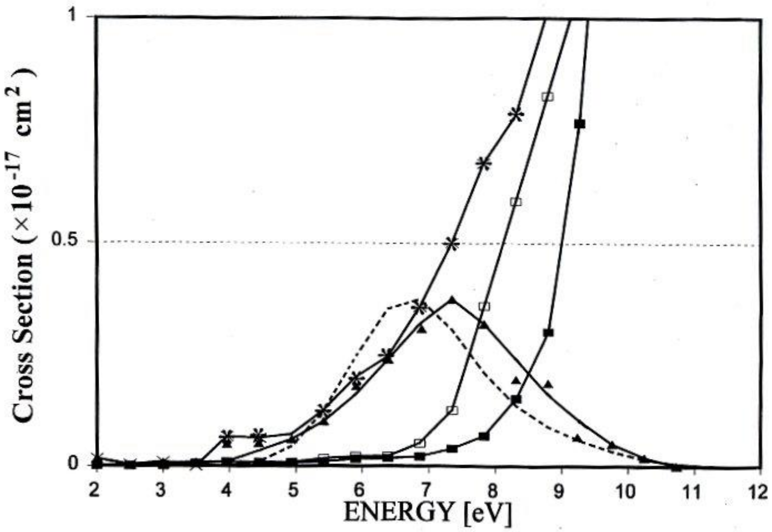
B. Goswami et al. PRA **88** (2013) 032707

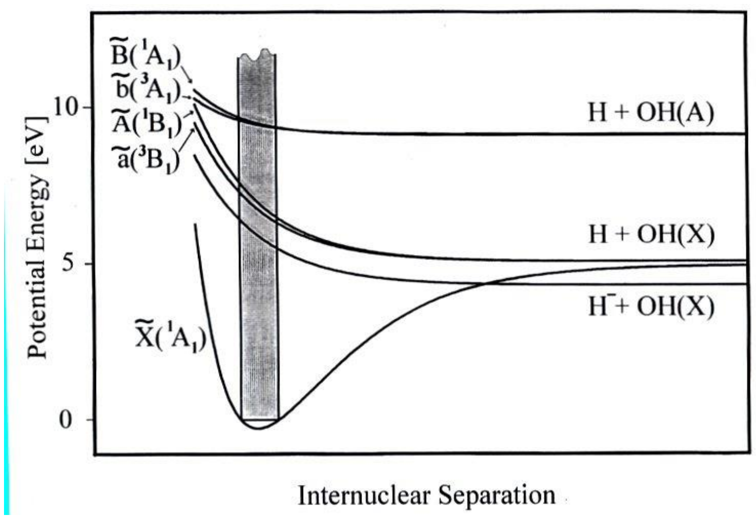
Dissociation into neutrals (H_2O)

Laser-induced fluorescence





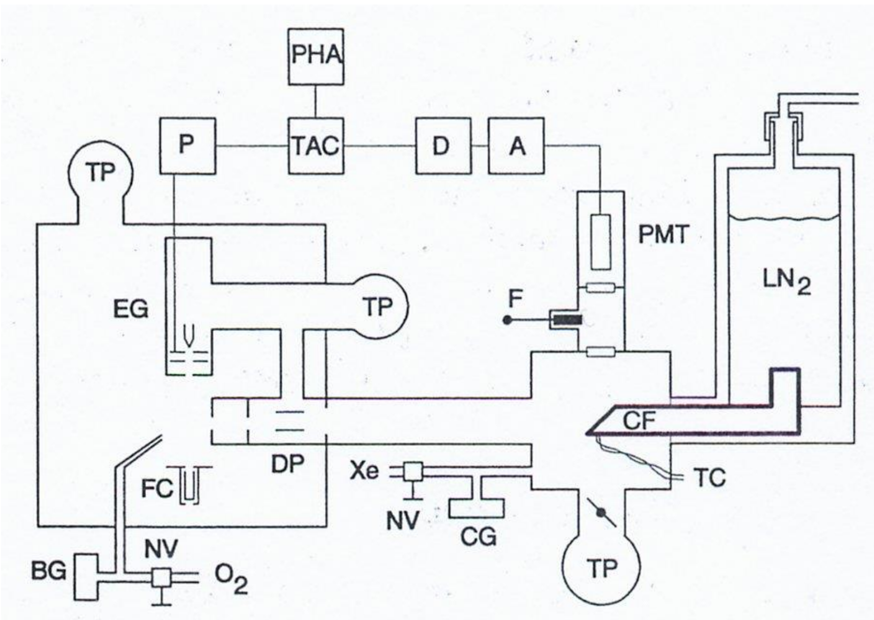




Herb and McConkey

Dissociation into neutrals (N_2O)

XeO* excimer decay



$N_2O \rightarrow O(^1S_0)$

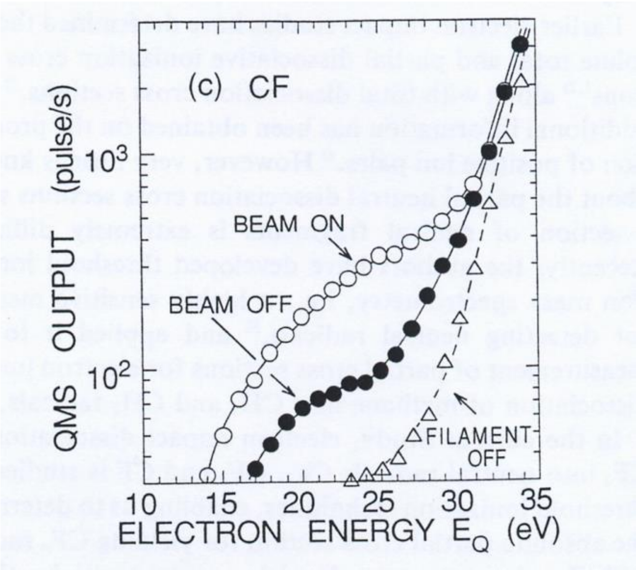
LeClair and McConkey JCP 99 (1993) 4566

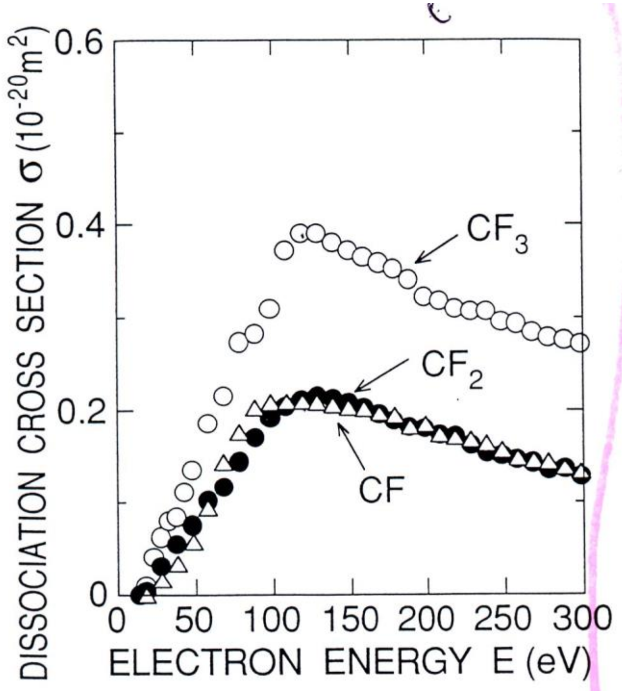
TABLE I. Absolute cross sections for the production of $O(^1S)$ following electron impact on $O(^3P)$, $O_2(X\,^3\Sigma_g^-)$, and $N_2O(X\,^1\Sigma^+)$ in units of 10^{-18} cm^2. The atomic oxygen data are taken from Ref. 57.

E (eV)	O	O_2	N_2O
4.4	0.54	···	···
4.8	1.30	···	···
5	1.60	···	···
6	2.57	···	···
7	3.02	···	···
8	3.22	···	···
9	3.28	···	···
10	3.27	···	···
12	3.15	···	5.12
14	2.97	···	9.52
16	2.79	0.35	13.8
20	2.44	0.92	18.1
24	···	1.26	20.5
28	···	1.47	21.7
32	···	1.61	22.2
36	···	1.73	22.4
40	1.37	1.82	22.4
45	1.21	1.92	22.5
50	1.07	1.97	22.4
60	···	2.04	22.2
70	0.69	2.07	21.7
80	···	2.08	21.1
90	···	2.06	20.6
100	0.38	2.04	20.1
120	···	1.98	18.9
140	···	1.90	18.0
160	···	1.82	17.1
180	···	1.75	16.3
200	0.08	1.67	15.6
250	···	1.53	13.9
300	···	1.39	12.8
350	···	1.26	11.8
400	···	1.16	11.0
450	···	1.08	10.3
500	···	1.02	9.60
600	···	0.92	8.63
700	···	0.82	7.91
800	···	0.75	7.39
900	···	0.69	6.88
1000	···	0.65	6.49

Dissociation into neutrals (CF_4)

Two electron beams: dissociation & ionization





Nakano and Sugai, Jpn. J. Appl. Phys. 31 (1992) 2919

Dissociation into neutrals (CF_4, CH_3F…)

„Volatile organotellurides”

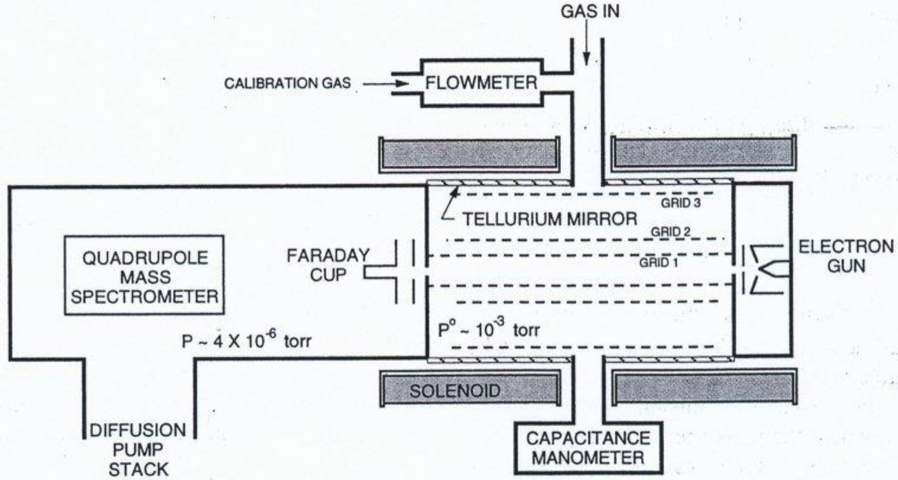


FIG. 1. Schematic diagram of the apparatus.

Motlagh and Moore JCP 109 (1988) 432

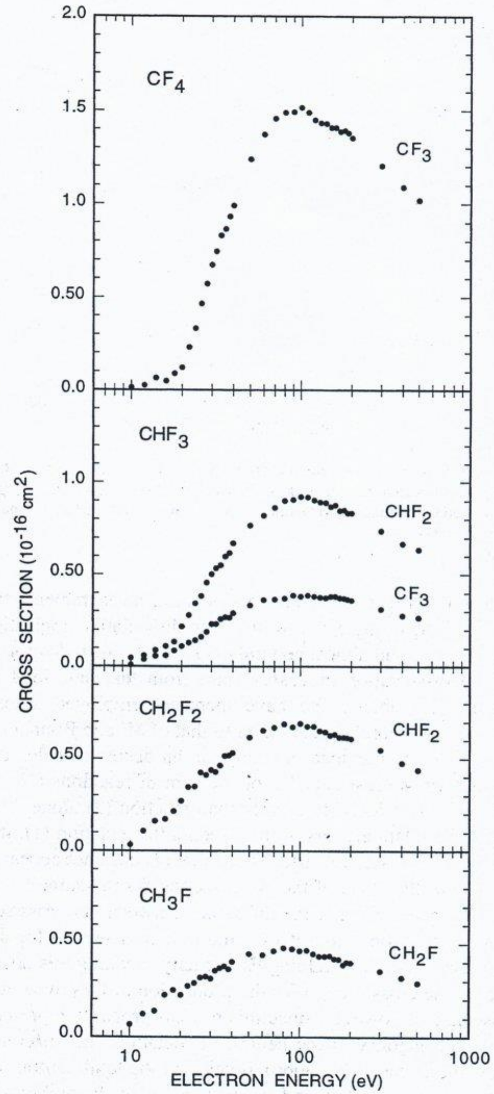


FIG. 5. Cross sections for the production of fluoromethyl radicals by neutral dissociation (n.d.) and dissociative ionization (d.i.) from electron impact on fluoromethanes.

$CF_4 \rightarrow CF_3 + F$
(4 sites possible)

$CHF_3 \rightarrow CHF_2 + F$
(3 sites possible)

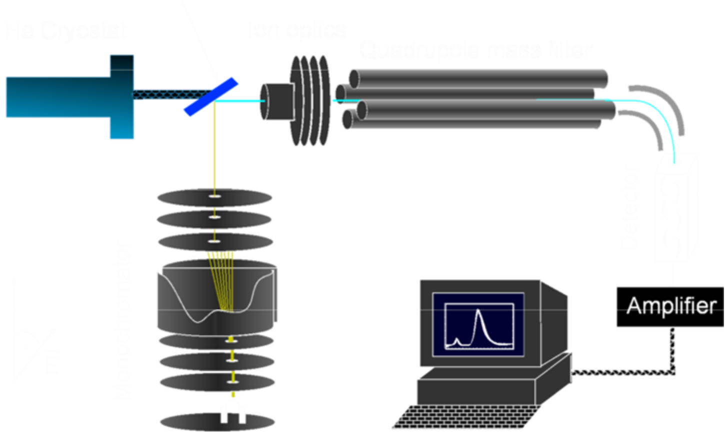
$CHF_3 \rightarrow CF_3 + H$
(1 site possible)

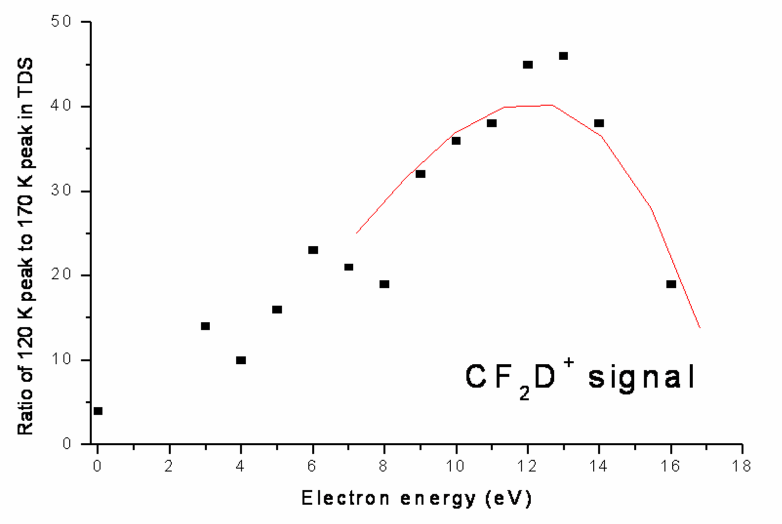
$CH_2F_3 \rightarrow CHF_2 + F$
(2 sites possible)

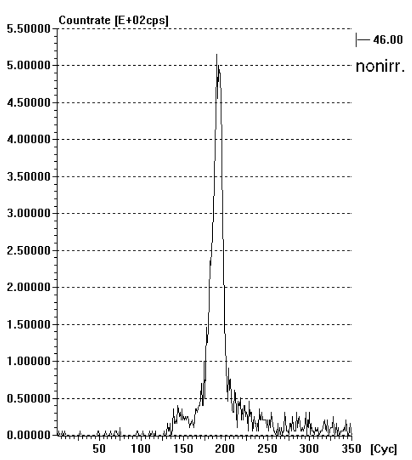
?

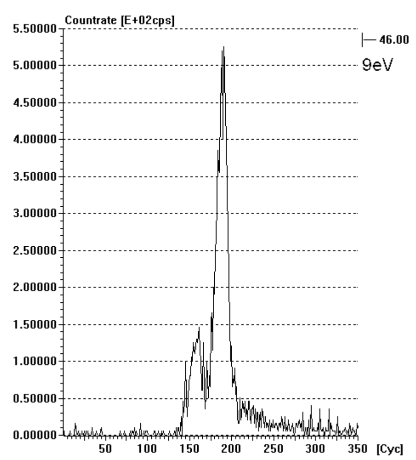
Dissociation into neutrals (CF_3COOH)

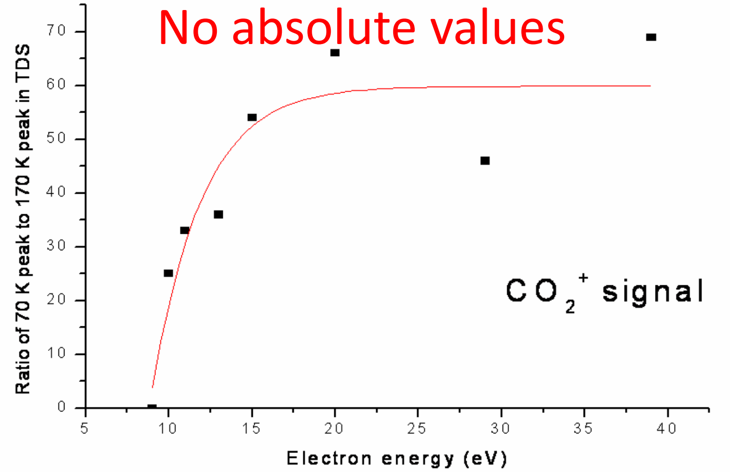
Cold deposition/ Electron irradiation/ Thermal desorption





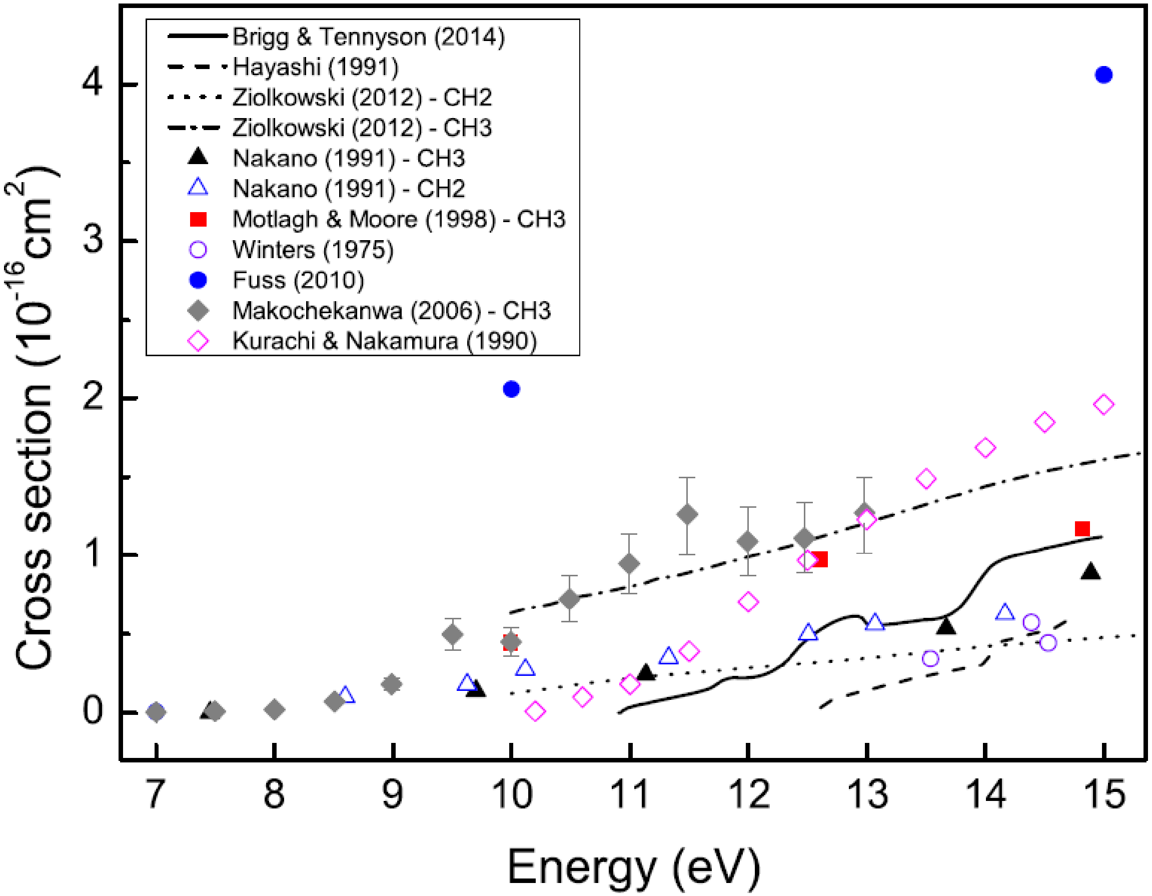






Reactions in nanofilms of trifluoroacetic acid (CF_3COOH) driven by low energy electrons, M. Orzol, T. Sedlacko, R. Balog, J. Langer, G. P. Karwasz, E. Illenberger, A. Lafosse, M. Bertin, A. Domaracka, R. Azria, Int. J. Mass Spectr. 254 (2006) 63

Dissociation into neutrals/ electronic excitation – theory & experiment (CH_4)

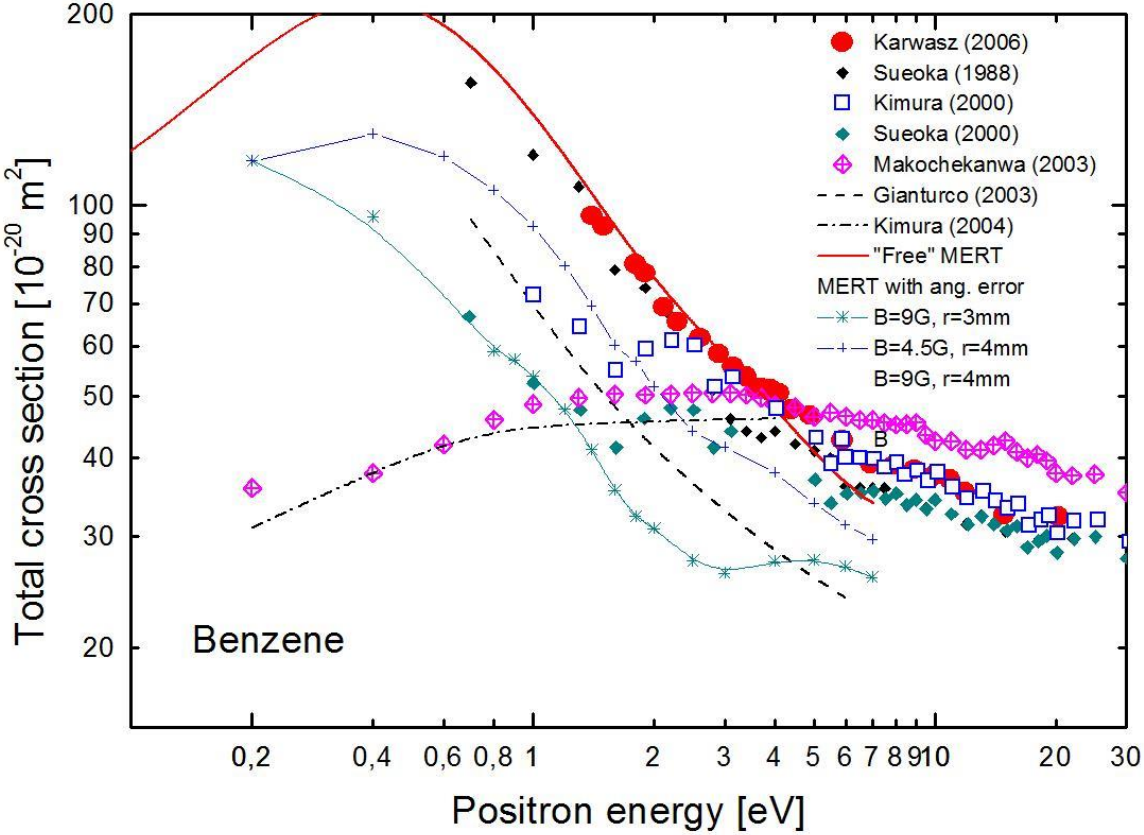


Experiments in serious disagreements;
Calculations Ziółkowski shifted by -3eV;
Briggs underestimated;

No recommended values were given

Mi-Young Song *et al*.. JPCRD 2015

Total: Positron scattering (C_6H_6)

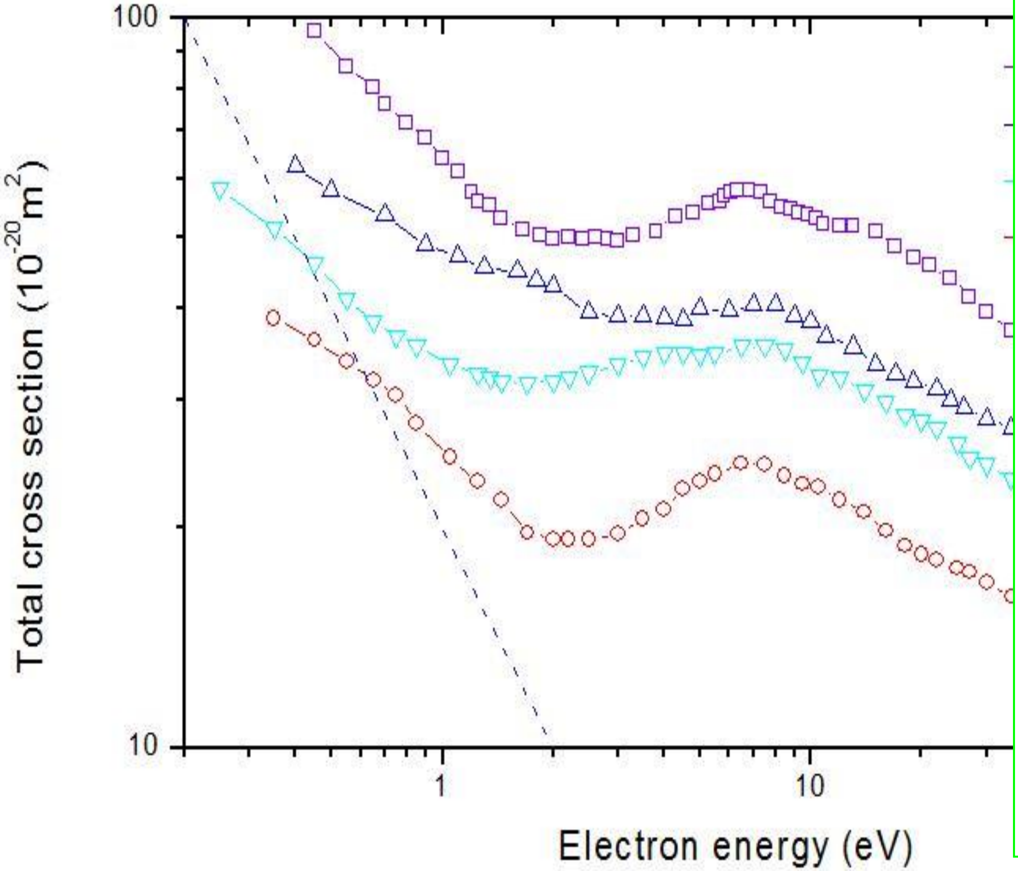


Modified effective range theory used to correct experimental data of Sueoka et al.

Kimura, Makochekanwa data come from Suoeka, but they published data obtained with a **higher guiding magnetic field**

G.P.Karwasz, A. Karbowski, Z. Idziaszek, R. S. Brusa, Nucl. Instr. and Meth. B, **266/3** (2008) 471

Total: polar molecules



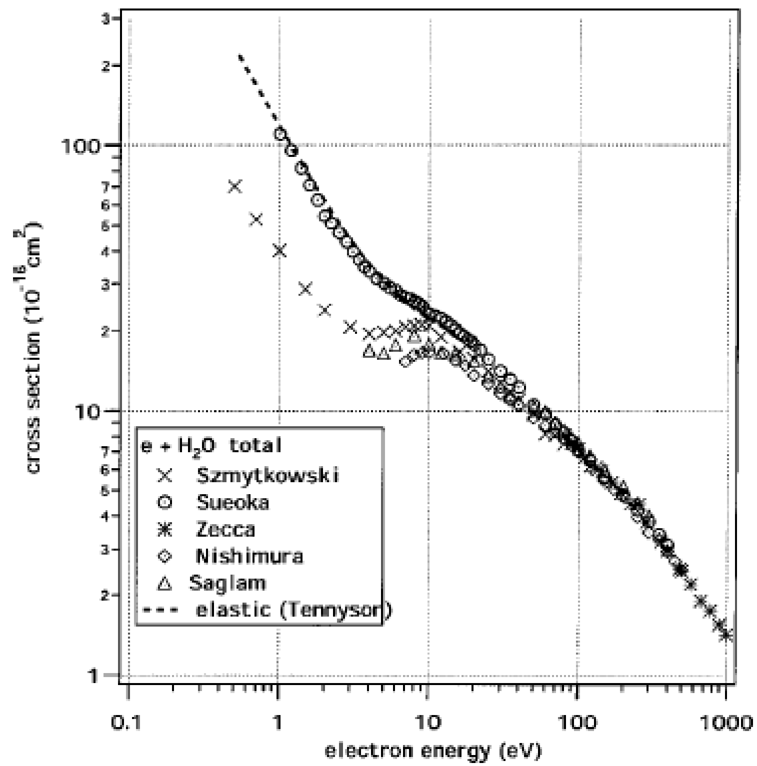
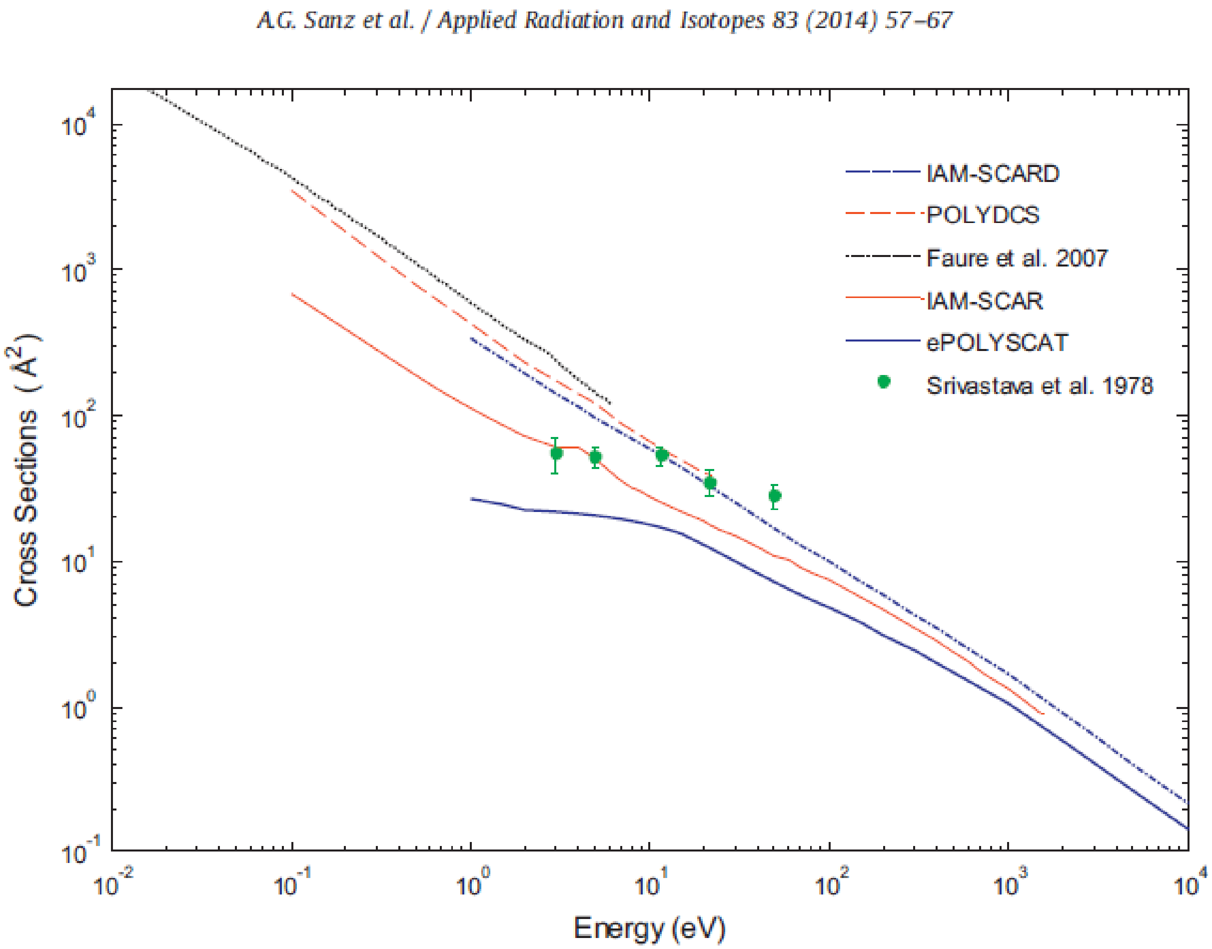


FIG. 2. Total scattering cross section, Q_T, of H_2O. A comparison is made of the experimental cross sections obtained by Szmytkowski (Ref. 21), Sueoka *et al.* (Ref. 26), Zecca *et al.* (Ref. 22), Nishimura and Yano (Ref. 23), and Saglam and Aktekin (Refs. 24 and 25). The theoretical elastic cross section obtained by Tennyson *et al.* (Ref. 28) is also shown for comparison.

Szmytkowski and collaborators

Y. Itikawa, N. Mason, JPCRD 34/1 (2005)

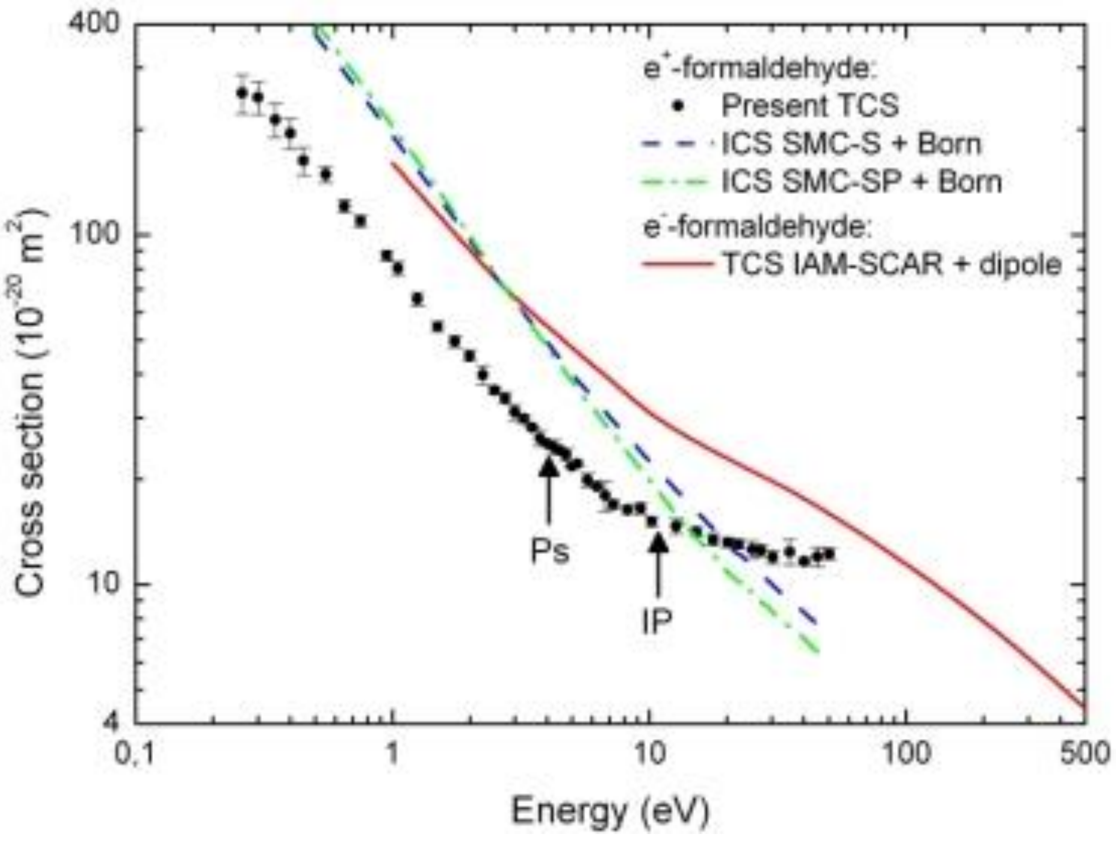
Total: polar molecules (HCN)



As experimentalist
I would believe more
in theory than in
experiment

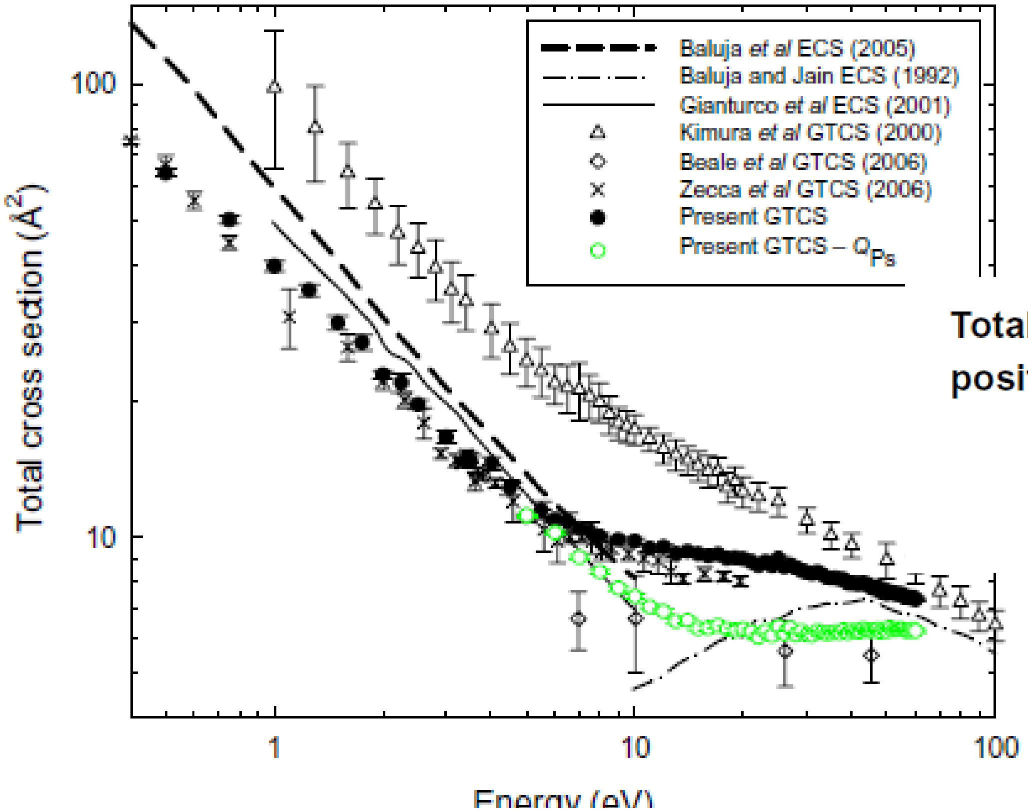
A. G. Sanz, Applied Radiation and Isotopes 83 (2014) 57–67

Polar molecules (e^+/e^- + HCOH)



As experimentalist
I would believe more
in theory than in
experiment

Independent atom model-screened additivity rule / Schwinger multichannel
A Zecca, E Trainotti, L Chiari, G García, F Blanco, M H F Bettega, M T do N Varella, M A P Lima and M J Brunger
Journal of Physics B: Atomic, Molecular and Optical Physics, Volume 44, Number 19



s ($e^+ + H_2O$)

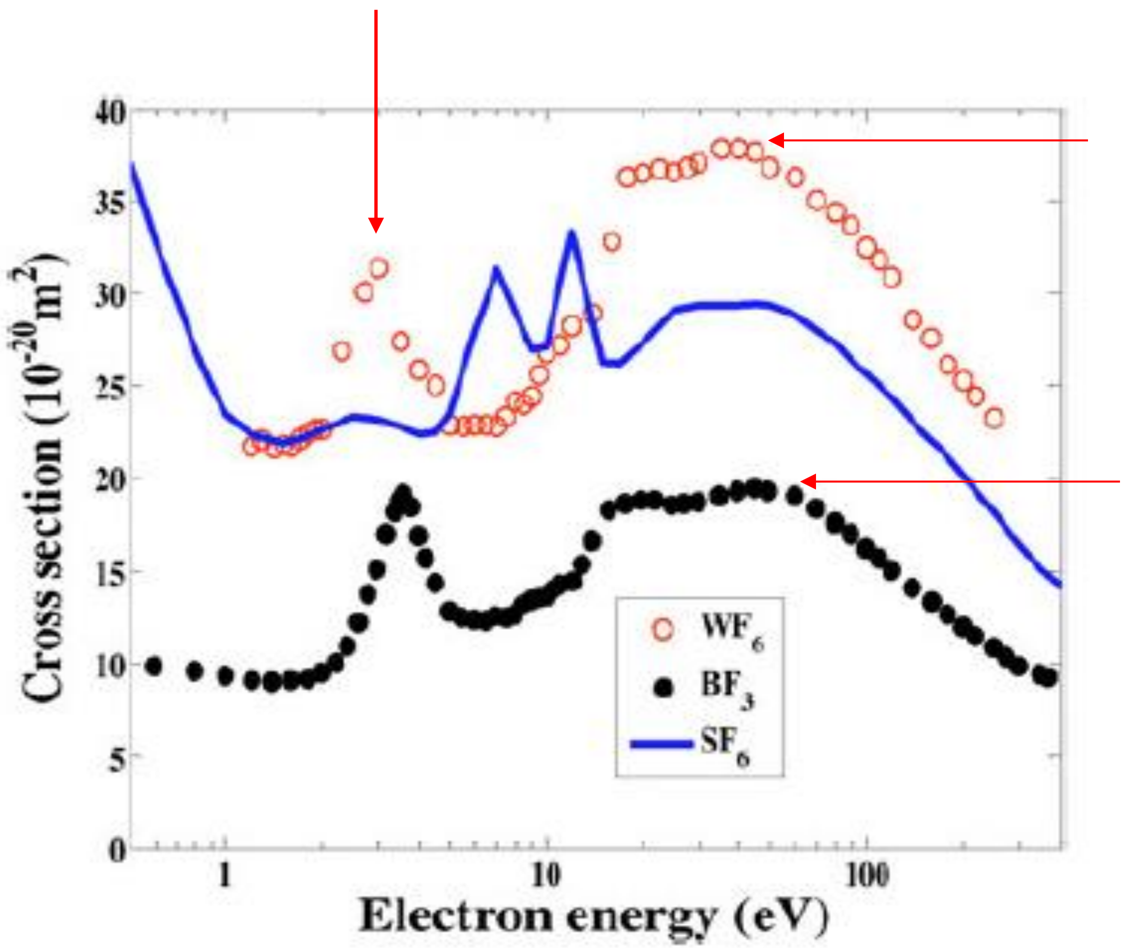
Total and positronium formation cross sections for positron scattering from H_2O and HCOOH

Casten Makochekanwa[1,5], Ana Bankovic[2], Wade Tattersall[1,3], Adric Jones[1], Peter Caradonna[1], Daniel S Slaughter[1], Kate Nixon[4], Michael J Brunger[4], Zoran Petrovic[2], James P Sullivan[1] and Stephen J Buckman[1]

Table 2. H_2O positron impact GTCS (10^{-16} cm^2). Numbers in parentheses are the values after the forward scattering effect correction. Errors are as explained in the text.

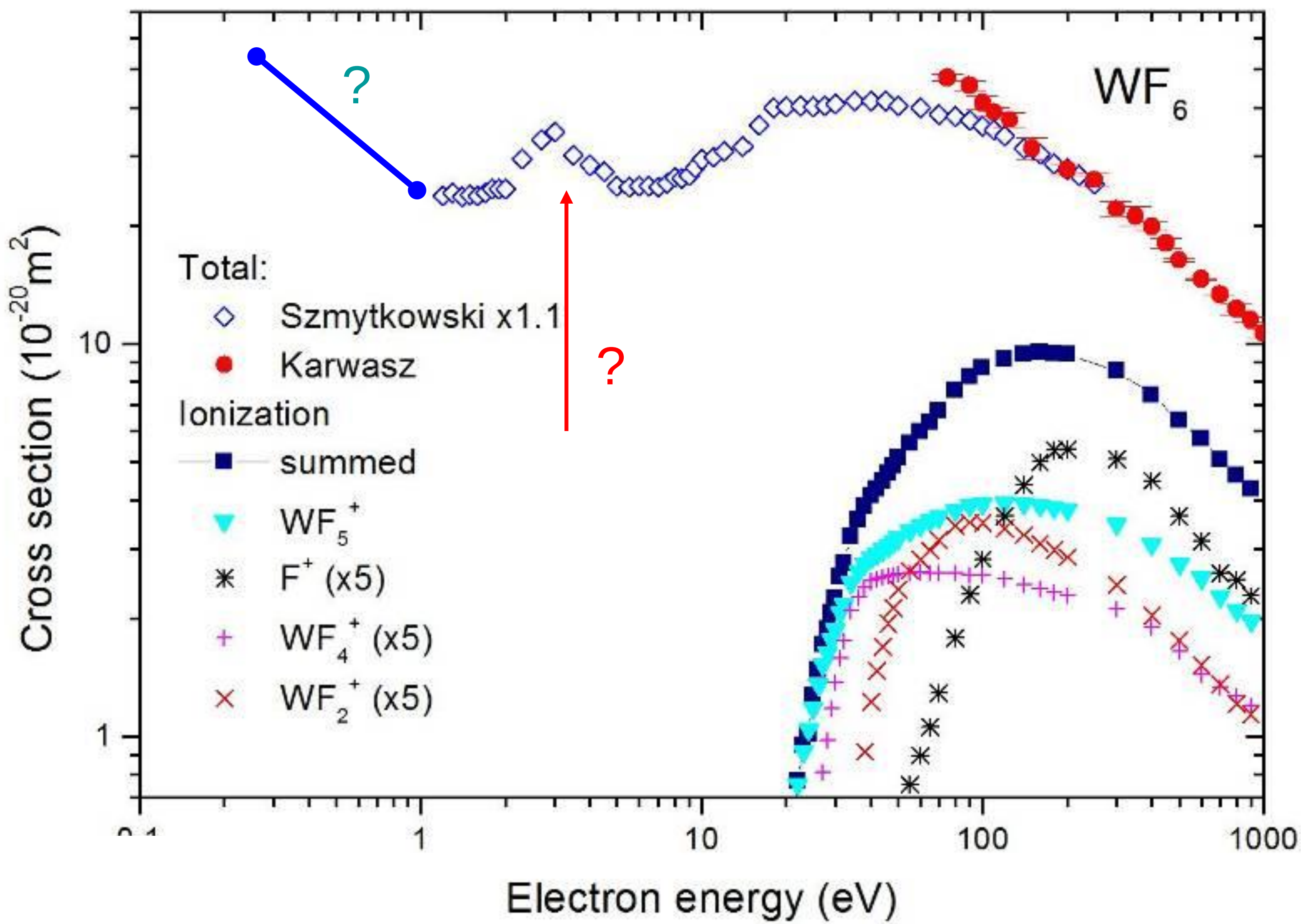
Energy (eV)	GTCS	Energy (eV)	GTCS
0.5	63.756(194.45) ± 1.148(3.50)	25	8.985(11.38) ± 0.119(0.15)
0.75	50.348(151.73) ± 1.019(3.07)	26	8.598(10.81) ± 0.120(0.15)
1	39.751(128.87) ± 1.050(3.40)	27	8.694(10.86) ± 0.125(0.16)
1.25	35.157(115.00) ± 1.080(3.53)	28	8.569(10.64) ± 0.136(0.17)
1.5	29.762(95.30) ± 1.076(3.45)	29	8.432(10.41) ± 0.125(0.15)

„Resonances" in total cross sections: WF_6



G. Karwasz, K. Fedus, FS&T (2013), experimental data: Szmytkowski and collaborators

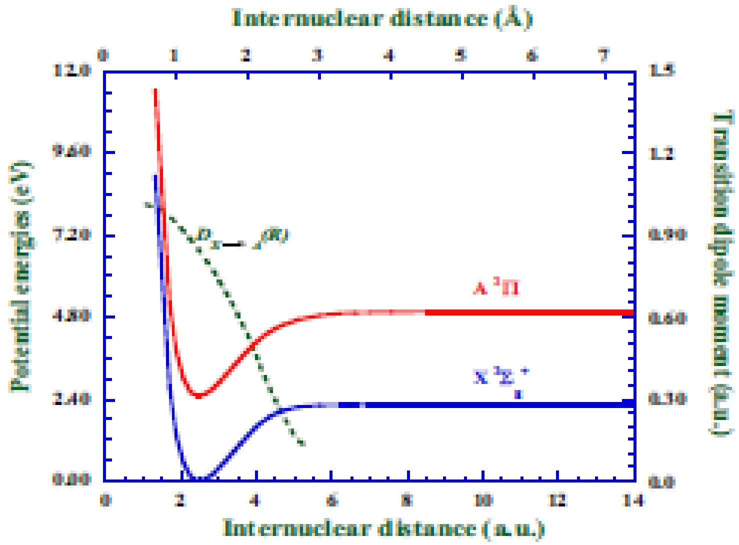
WF_6 - few data

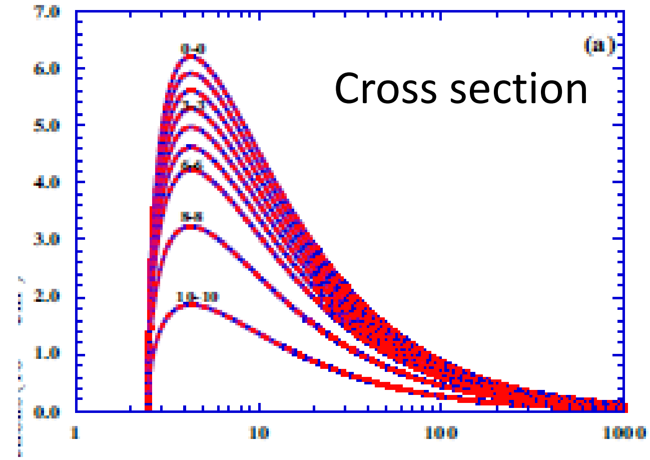


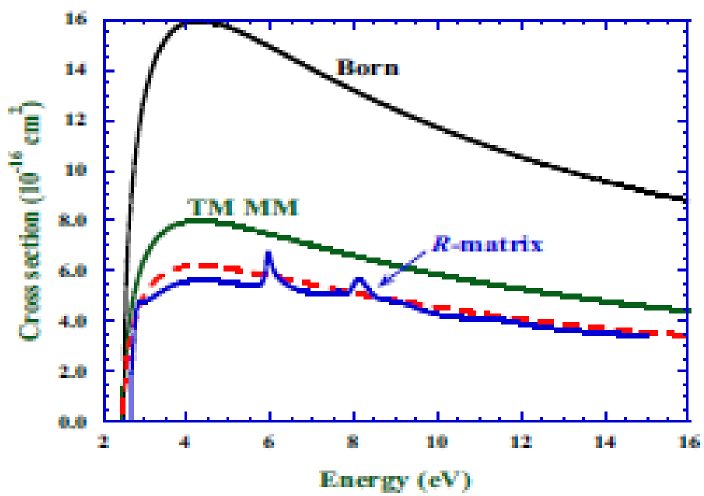
GK, work in progress

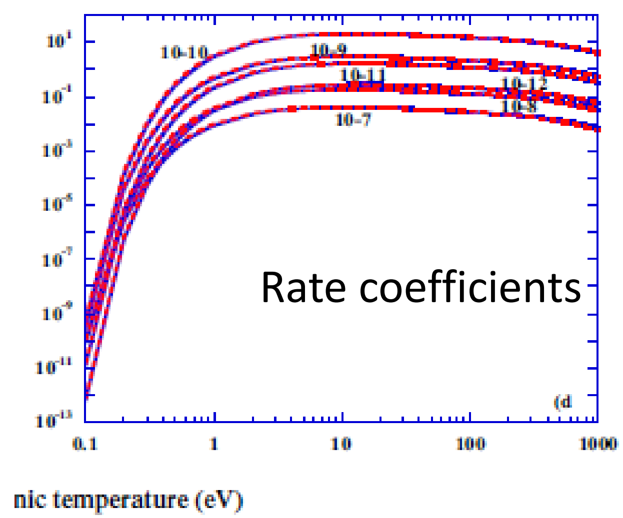
BeH: electronic and vibrational excitation

$X\,^2\Sigma^+\,(v{=}0) \rightarrow A\,^2\Pi\,(v')$





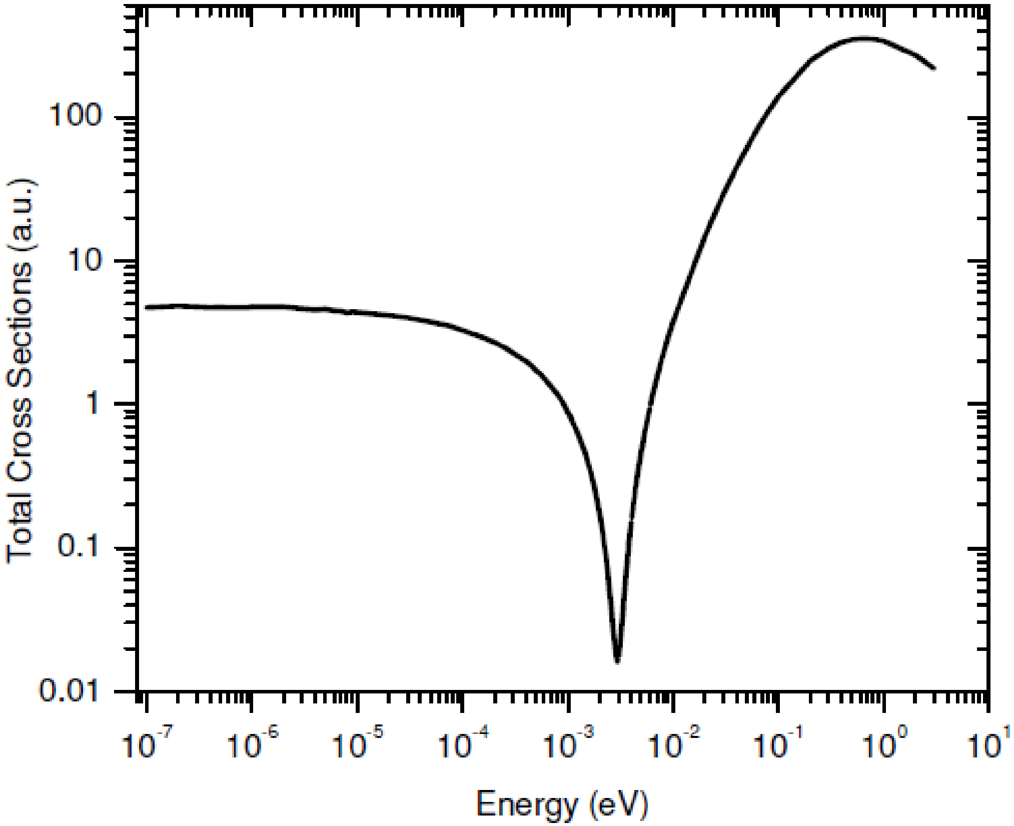




R Celiberto, K L Baluja and R K Janev, Plasma Sources Sci. Technol. **22** (2013) 015008

Mott-Massey Schr. eq.

Beryllium



Ramseuer minimum in Be

Figure 1. Total cross sections for low-energy electron scattering from atomic beryllium.

D. R. Reid, J. M. Wadehra, J.Phys. B 47 (2014)

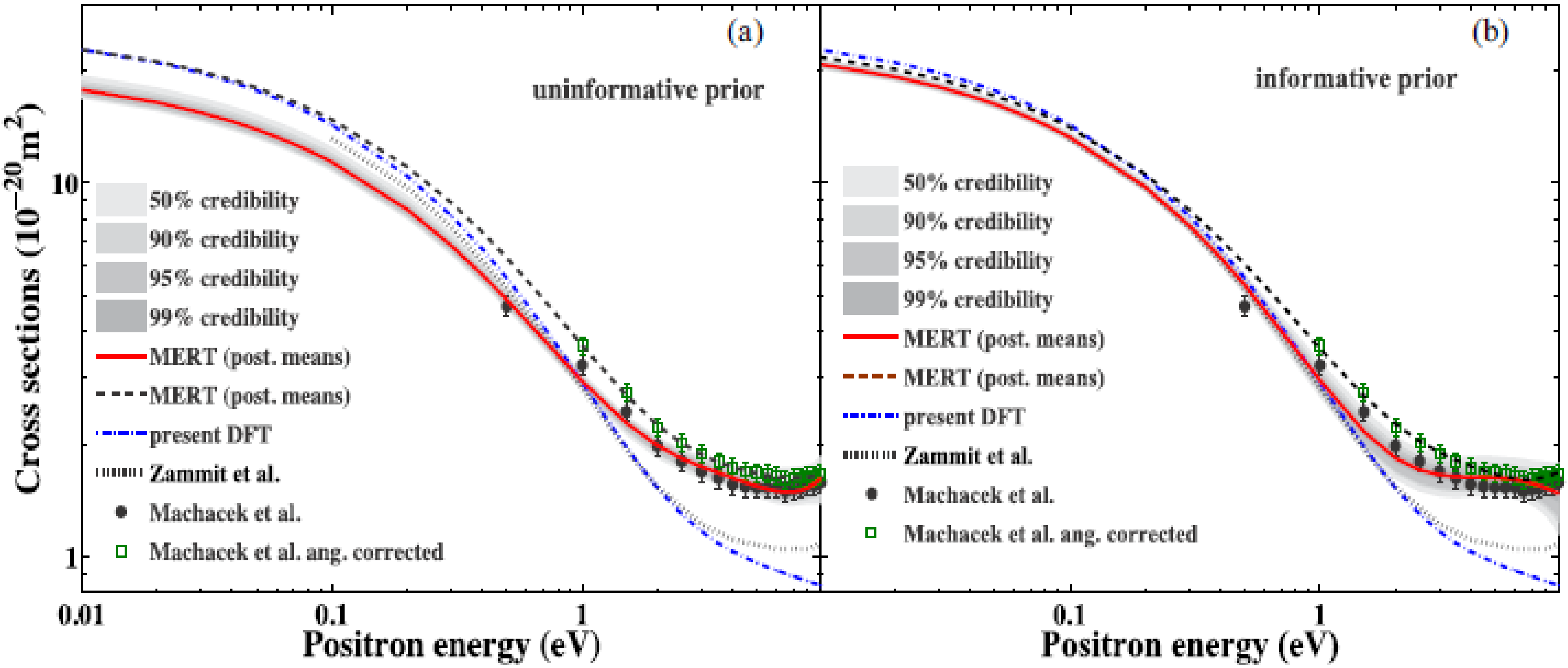
Positron + H_2: Bayesian analysis

PHYSICAL REVIEW A **91**, 062701 (2015)

Positron scattering on molecular hydrogen: Analysis of experimental and theoretical uncertainties

Kamil Fedus,[1,*] Jan Franz,[2,†] and Grzegorz P. Karwasz[1,‡]

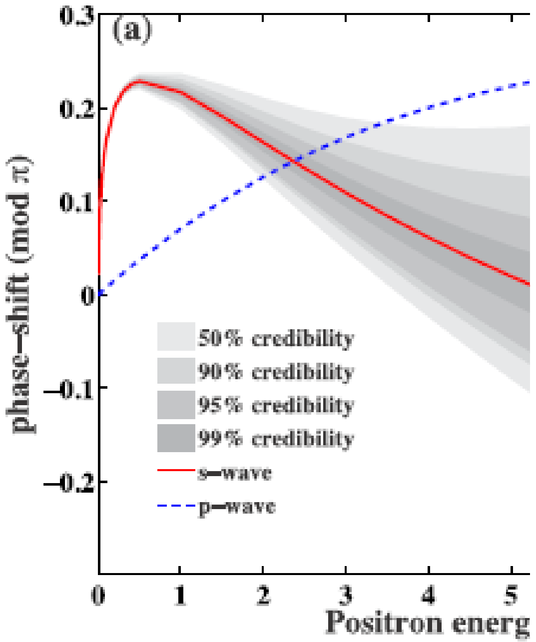
Total cross section



Positron + H_2: Bayesian analysis

Phase shifts

KAMIL FEDUS, JAN FRANZ, AND GRZEGORZ P. KARWASZ



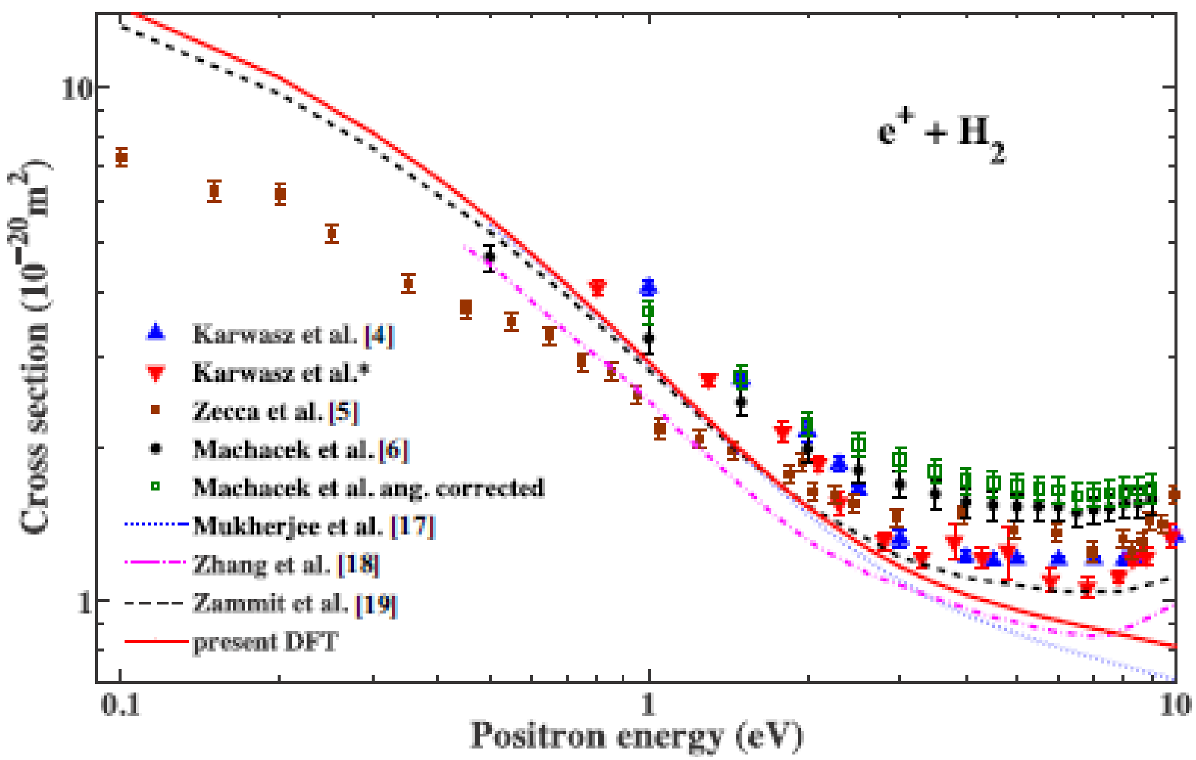
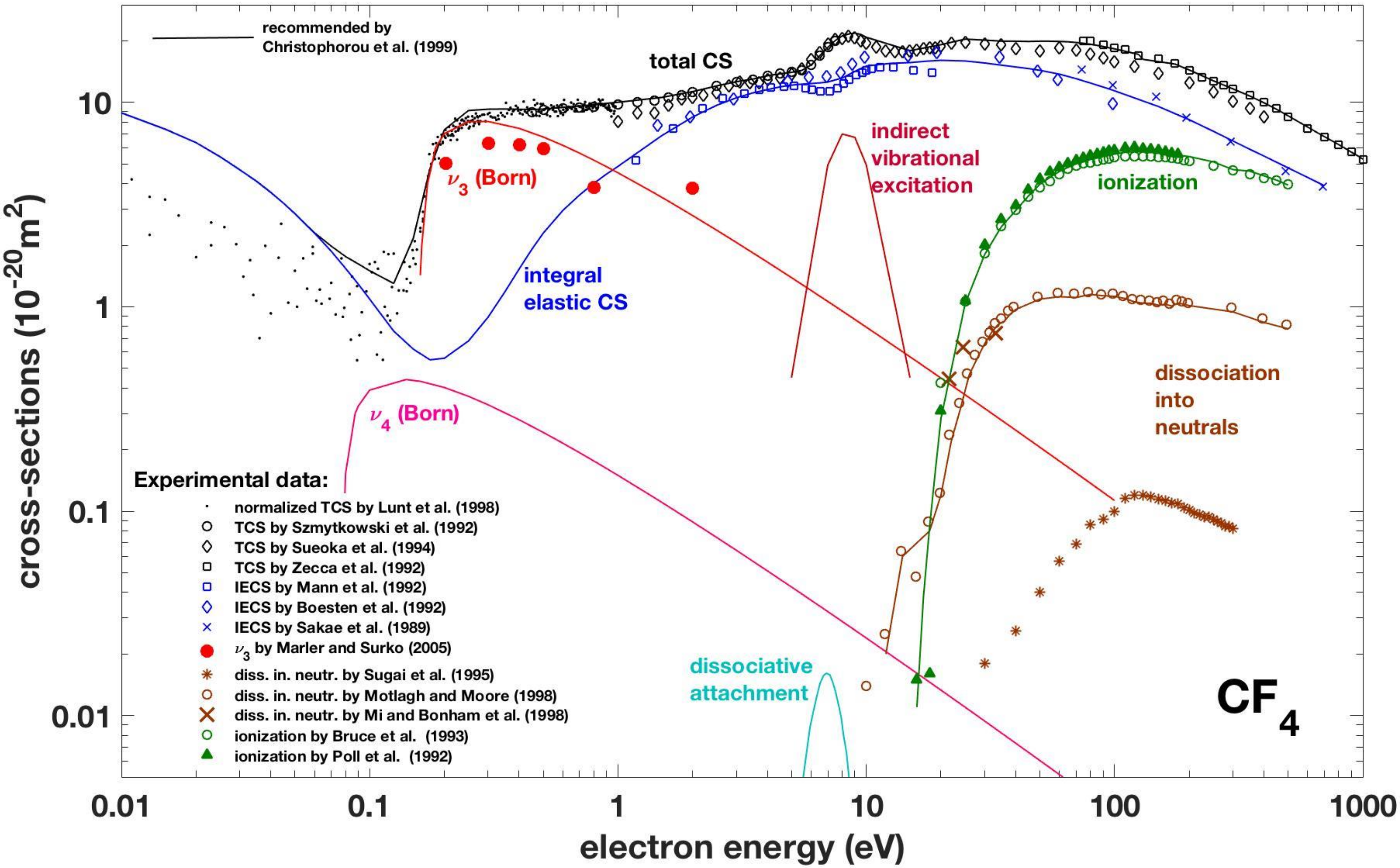


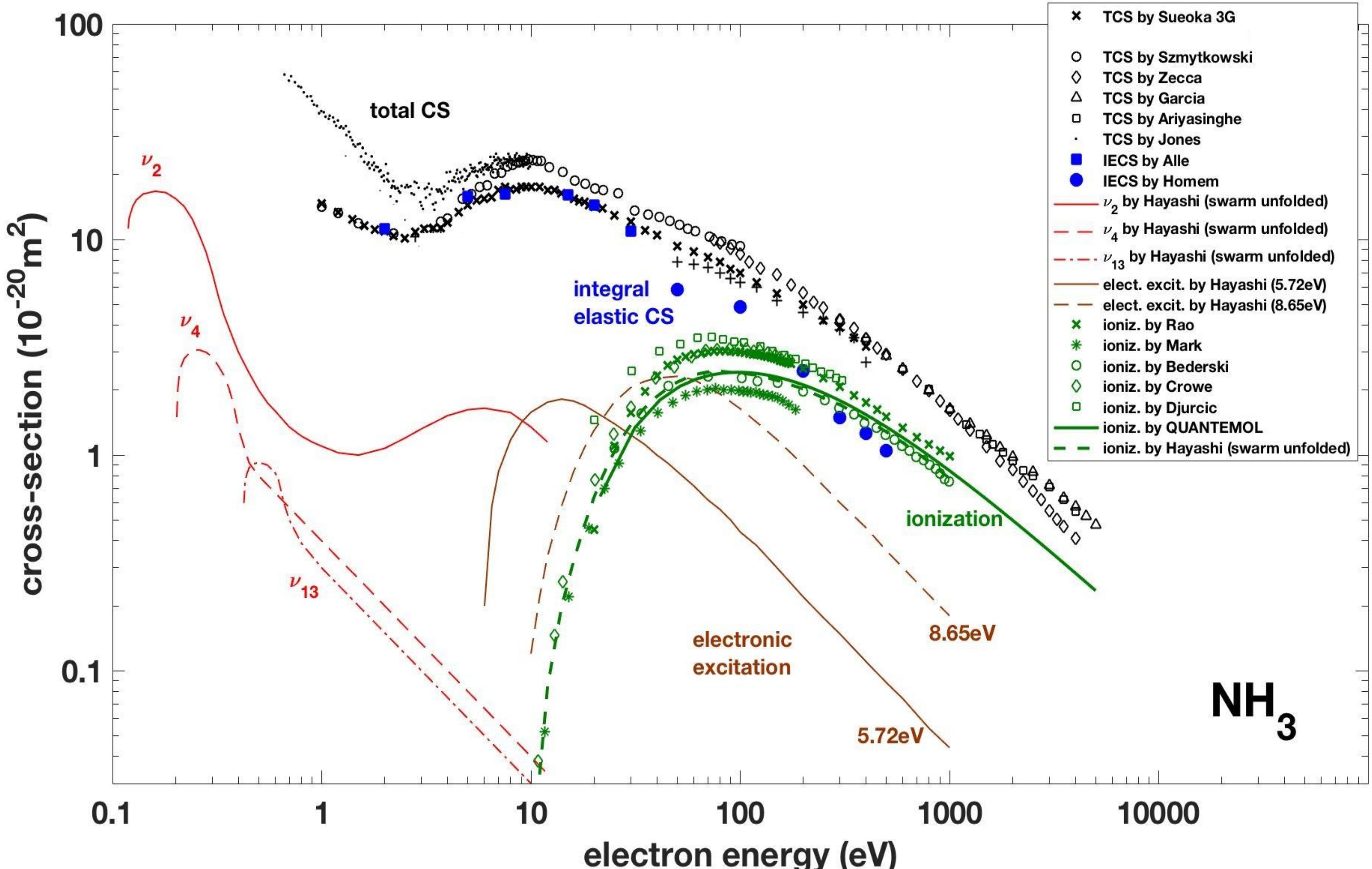
FIG. 3. (Color online) MERT-derived
sections by Machacek *et al.* [6]: (a) direct
phase shift only posterior mean values are given. The gray areas in the plot correspond to 50%, 90%, 95%, and 99% posterior regions due to uncertainties of MERT parameters.

Bayesian analysis does not help much when experiments are uncertain

Check of congruence: CF_4 (√)

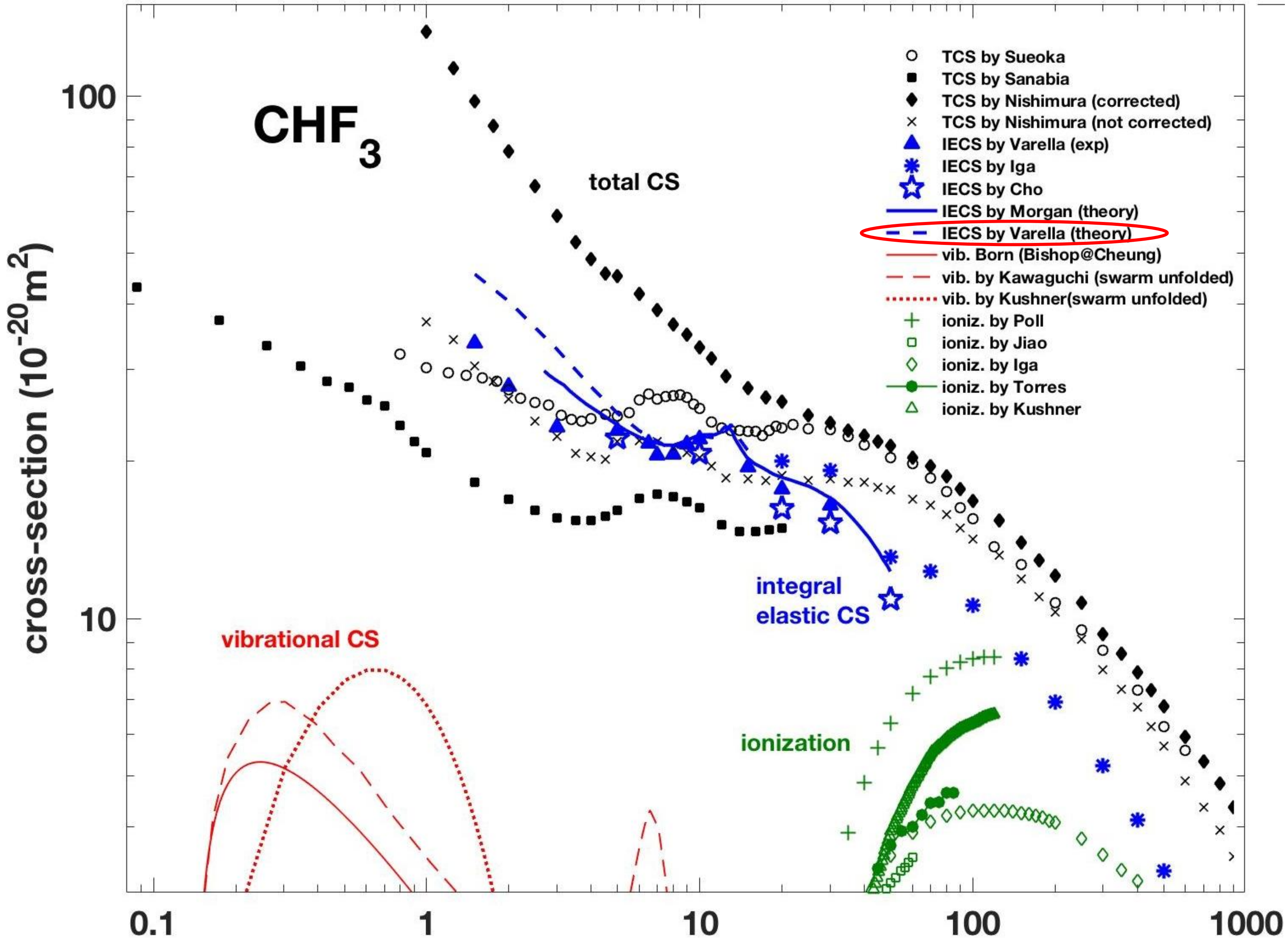


Check of congruence: NH_3 (✗)



Really poor agreement...

Check of congruence: CHF_3 ✗ ✗



Experimental uncertainties for electron scattering on molecules

- Total, in majority cases, within ±5%

but no data for BeH, WH_2, few WF_6

- Ionization: total within ±10%; in agreement with theories

but partial ±15%

- Electronic excitation: good agreement between experiment and theory only of H_2

- Vibrational excitation: poorly understood at resonances

- Dissociation into neutrals desperately needed

Conclusions (II)

- Some targets possible for theory, other for experiments
- Solution: **commissioning measurements;**
- - NH_3 vibrational and electronic excitation (Fullerton California?)
- - BeH_2 elastic theoretical (Prague University?)
- - BeH_2 electronic excitation (?)
- - polar molecules (NH_3) at low energies (UNC Toruń?)
- - H vs defects in tungsten (positron beam: Trento University, TUV München, UNC Toruń)

Thank for your attention,
and IAEA staff for welcome