Evaluation of cross section for electron collisions with Methane

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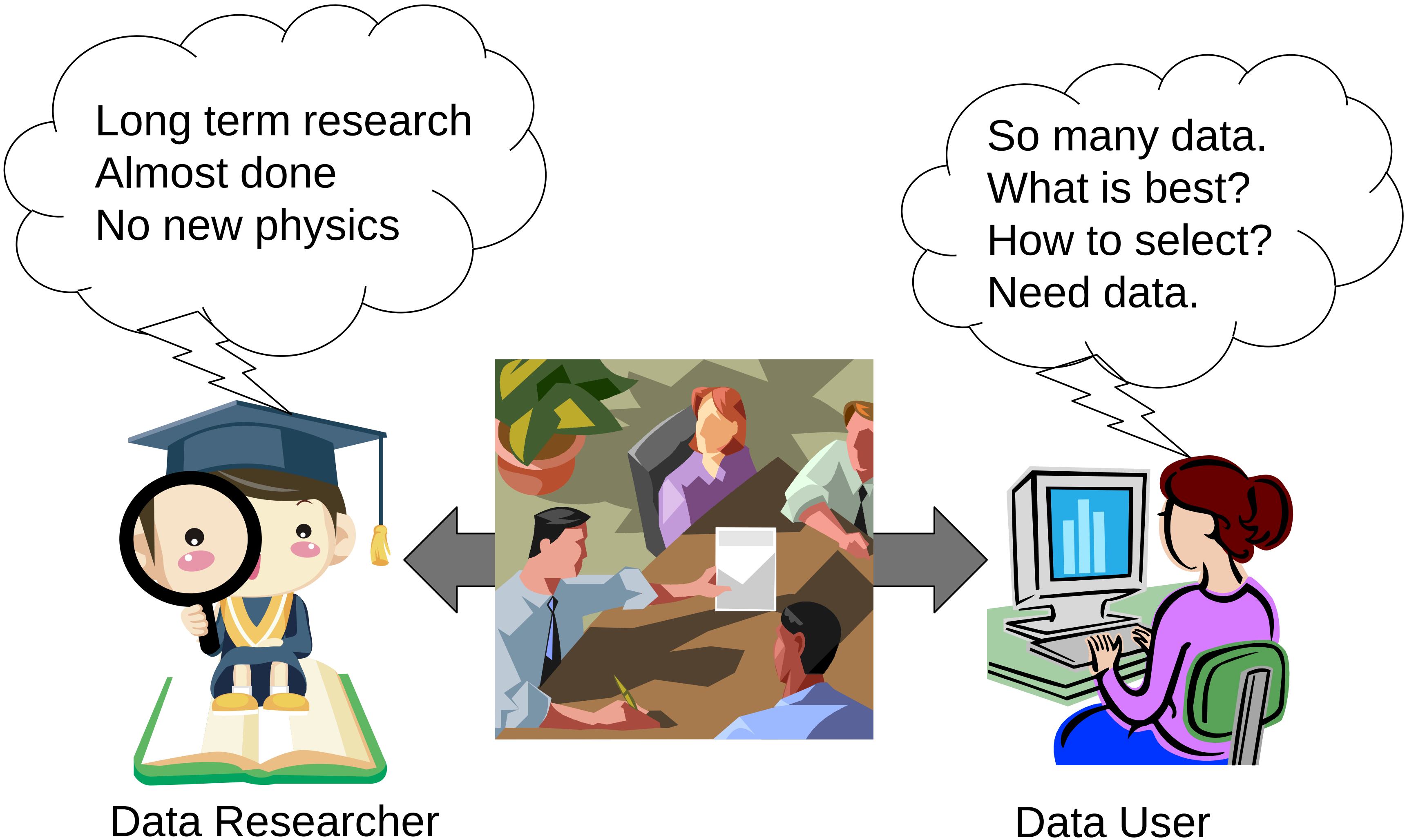
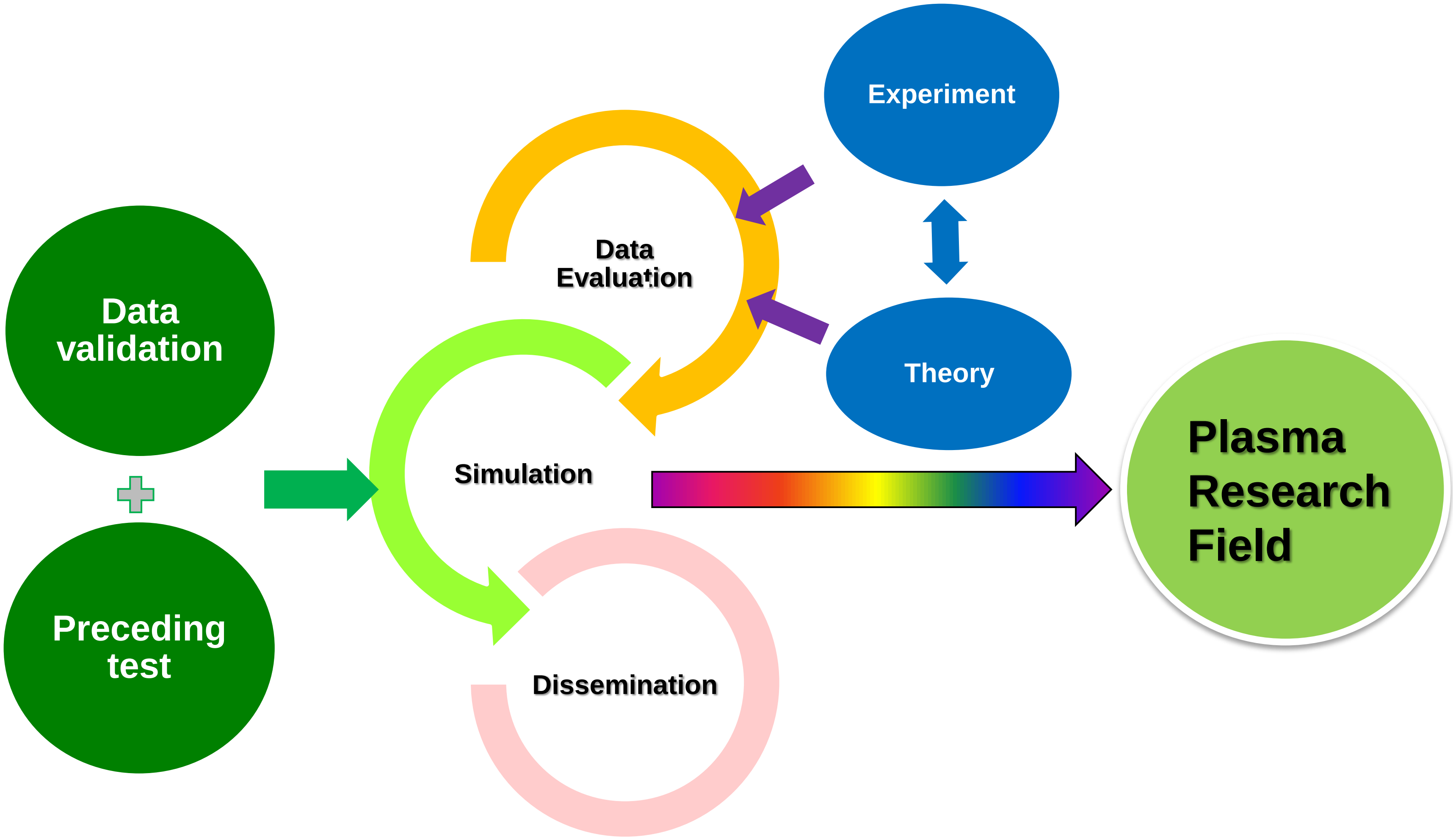
Outline

- Introduction
- Organization of Evaluator Group
- Evaluation Procedures
- Evaluation of CH_4 Collision data
- Summary

1. Introduction




Data validation
Preceding test
Data Evaluation
Experiment
Theory
Simulation
Dissemination
Plasma Research Field

2. Organization of Evaluator Group

- This work decide at the Joint IAEA-NFRI Technical Meeting (TM) on Data Evaluation for Atomic, Molecular and Plasma Material Interaction Processes in Fusion in September 2012
- Participants recommended group member and molecule at that time.
- Group Members:
  - Y. Itikawa (Japan)
  - Grzegorz P. Karwasz (Nicolaus Copernicus University),
  - J. Tennyson (University College London)
  - Viatcheslav kokoouline(University of Central Florida)
  - H. Cho(Chung-Nam National University)
  - Y. Nakamura (Tokyo Denki University)
  - J.-S. Yoon, M.-Y. Song (National Fusion Research Institute)
- **Our purpose: To establish the internationally agree standard reference data library for AM/PMI data**

1) Experiment (Cho, Karwasz, Nakamura)

- The experimentalist cover all processes
- We must check carefully systematic uncertainty of the experimental data what we will be doing before we evaluate the experimental data
- We need swarm experimentalist to evaluate vibrational excitation cross section
- We need experienced person to evaluate electronic excitation cross section

2) Theory (Itikawa, Tennyson, Kokouline)

- The theorists cover all processes
- Theory should cover some processes because of experiment is so difficult. But theorists don’t give uncertainty. How to solve these problems.
- Few researchers can measure electronic excitation cross section but they cannot analysis because they don’t know radical state. They need theoretical data

→1st GM : 23 - 25 January 2013, Gunsan, South Korea

→ 2nd GM: 25 -27 June 2013, Deajeon, South Korea

→3rd GM : 23-24 September 2013, Open university. UK

3. Evaluation Procedures

Why Methane?

- Methane (CH_4) is the simplest hydrocarbon molecule and has attracted significant interest as a target for low-energy electron collision studies.
- It has many technological and atmospheric applications as well as a fundamental importance as one of the testing grounds for the collision theories.
- Reflecting this importance, a number of compilation of cross section data have been published. Those publications, however, are rather old and a considerable number of new cross sections are available now.

1. To review a previous evaluation paper

1) W.L. Morgan, "Critical evaluation of low-energy electron impact cross sections for plasma processing modeling. II: CF_4, SiH_4, and CH_4 ", Plasma Chem. Plasma Process. 12, 477 (1992)
2) I.Kanik, S. Trajmar, and J.C. Nickel "Total electron scattering and electronic state excitations cross-sections for O_2, CO, and CH_4" , J. Geophys. Res. 98, 7447 (1993)
3) G. P. Karwasz, R. S. Bursa, and A. Zecca, "One century of experiments on electron－atom and molecule scattering: a critical review of integral cross－sections II. Polyatomic molecules" La Rivista del Nuovo Cimento 24, 1 (2001)
4) T. Shirai, T. Tabata, H, Tawara and Y. Itikawa, "Analytic cross sections for electron collisions with hydrocarbons: CH_4, C_2H_6, C_2H_4, C_2H_2, C_3H_8, and C_3H_6", Atomic Data Nucl. Data Tables 80, 147 (2002)
5) R.K. Janev and D. Reiter, "Collsion processes of CH_y and CH_y^+ hydrocarbons with plasma electrons and protons", Phys. Plasmas 9, 4071 (2002) [Revised in: D. Reiter and R.K. Janev, "Hydrocarbon collision cross sections for magnetic fusion: The methane, ethane and propane families" Contrib. Plasma Phys. 50, 986 (2010)
6) M.C. Fuss, A. Munoz, J.C. Oller, F. Blanco, M.-J. Hubin-Franskin, D. Almeida, P. Limao-Vieira, and G. Garcia, "Electron-methane interaction model for the energy range 0.1-10000 eV " Chem. Phys. Lett. 486, 110 (2010)
7) LB Vol17C
8) H. Tanake et al, NIFS-DATA 108, 2009

2. Define working Scope

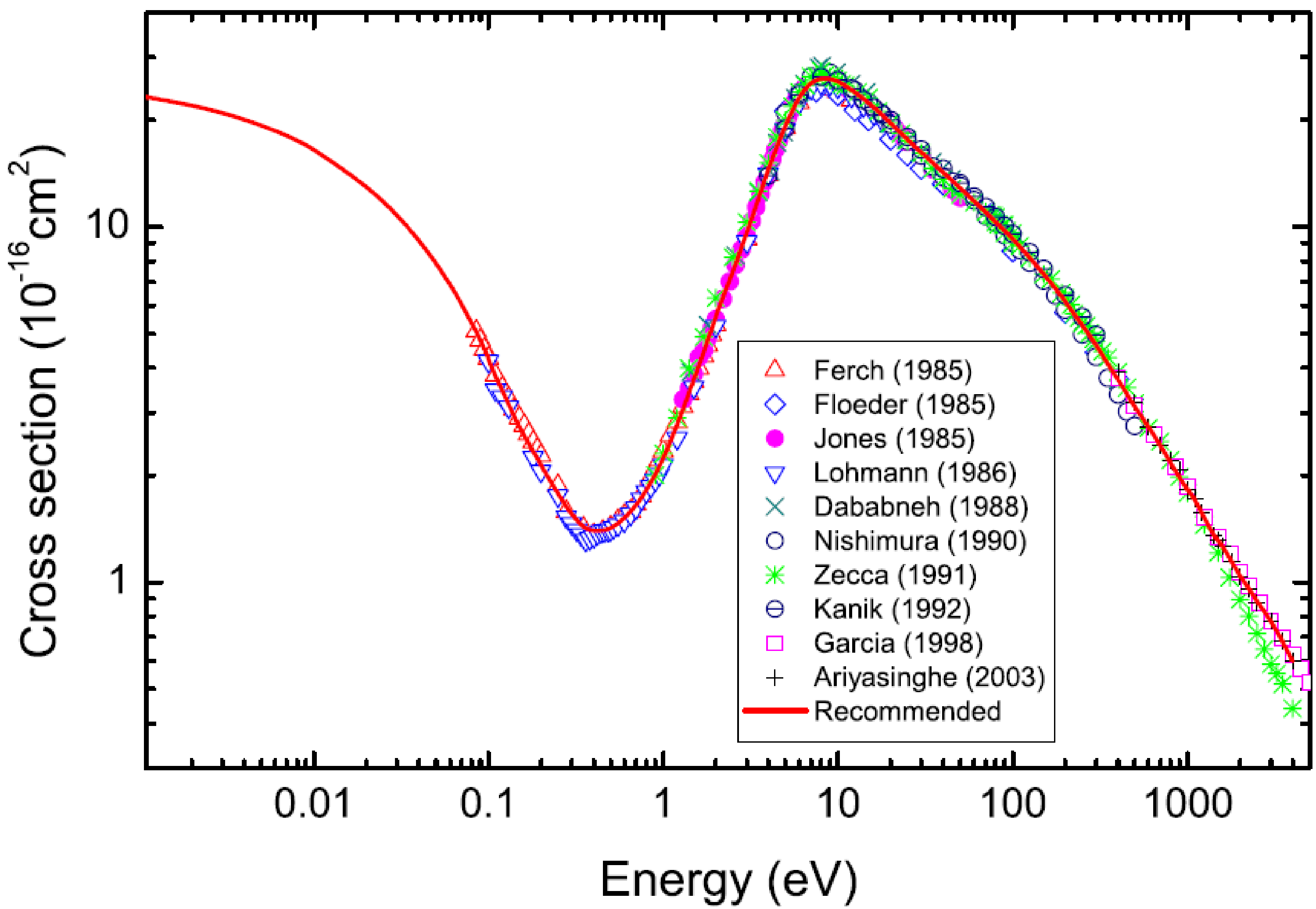
- We don’t have all collision processes and decide working scope. (Electron collision, Photon collision cross section)
- We list up processes according to Prof. Itikawa’s comment processes LIST

3. Define Main evaluator for each process

- We shard working part from the processes list. All coworker decide working part.
- we specified main evaluator for each process in order to arrange the contents of evaluation. He will collects and adjusts other evaluator’s opinion.

  (1). Ionization (dissociative ionization) – [**Karwasz**]
  (2). Total cross section- [**Karwasz**]
  (3). Electron Attachment **[Cho]**
  (4). Elastic + DCS [**Cho,** Itikawa]
  (5). Momentum transfer + DCS [**Nakamura,** Karwasz, Cho, Itikawa,]
  (6). Vibrational excitation + DCS [**Kokoouline**, Karwasz, Nakamura]
  (7). Rotational excitation + DCS [**Itikawa,** Nakamura, Kokoouline, Tennyson]
  (8). Electron excitation & Dissociation [**Tennyson**, Kokoouline, Cho, NFRI]

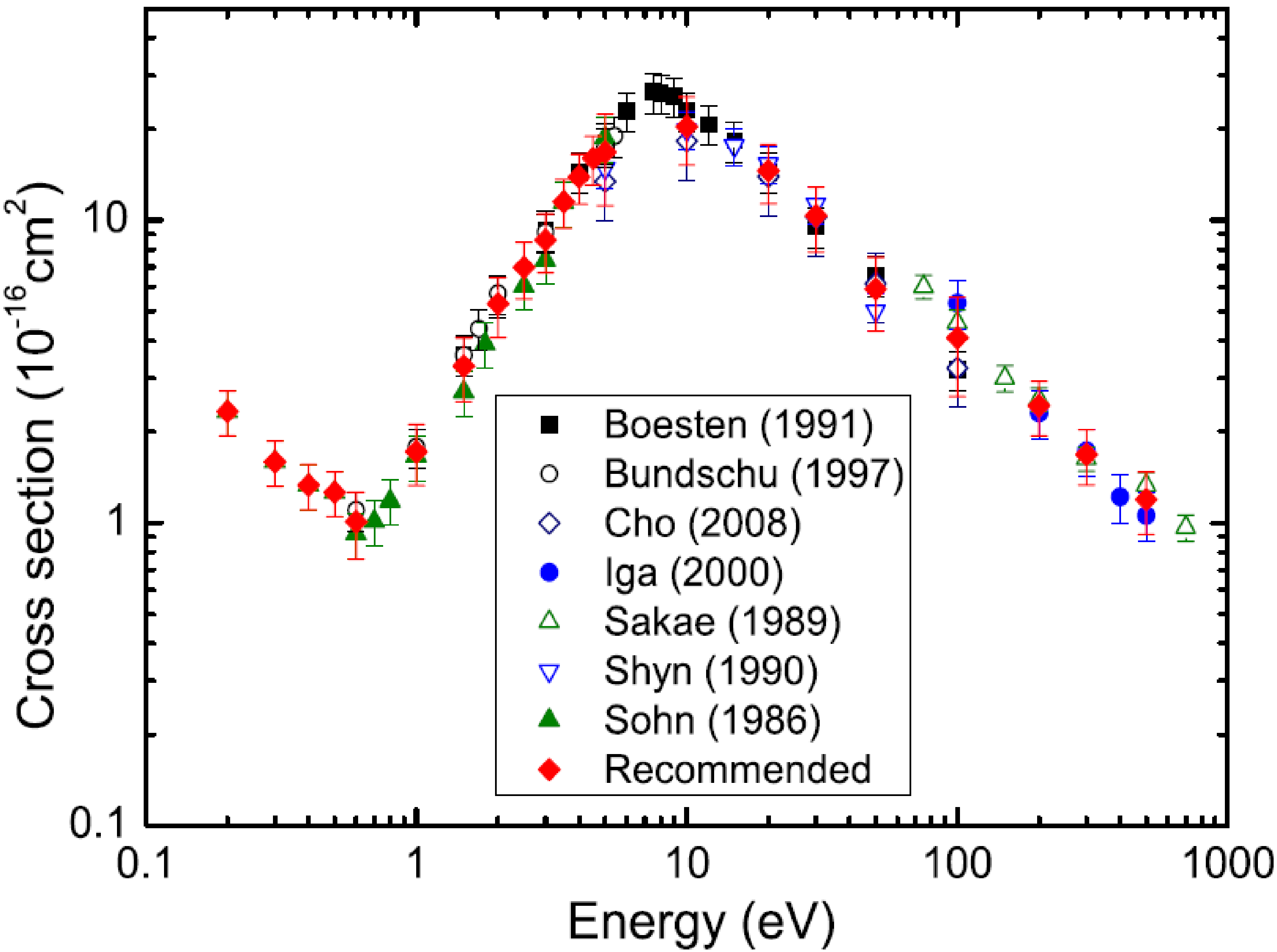
1) Total scattering cross section



Authors	Energy range Declared errors	SC length SC apertures	Method
Ferch *et al* (1985)[12]	0.085-12eV, 1-4.5% stat., systematic $< 1\%$		absolute, time of flight
Lohmann and Buckman (1986)[13]	0.1-20 eV, 3-5% stat.	255 mm	absolute, time of flight
Jones (1985)[14]	1.3-50 eV, 3-9% total	380 mm, $\phi = 2.0$ mm	absolute, time of flight
Szmytkowski (1991, from Zecca *et al*[15])	0.4-100 eV, 3% stat, 4% syst.	30.5 mm, 0.35×0.7 mm, $2 \times 10^{-3} sr$	absolute, linear transmission, cylindrical monochromator
Dababaneh *et al* (1988)[20]	1.5-500 eV, 5% + ang. Resolution	109 cm, $3 - 28^{\circ}$	absolute, transmission, magnetic guiding
Nishimura and Sakae (1990)[17]	5-500 eV, 13% syst., 1-5 % stat.	25 mm + 1 cm drift $\phi = 1.0 - 2.0$mm	absolute, linear transmission, with drift cell, angular resolution extrapolated to zero
Kanik *et al* (1992)[16]	4-300 eV, 2.1-3.3%		total absolute, linear transmission, electrostatic gun
Zecca *et al* (1991)[15]	75-4000 eV, 2.5% sys., 3% stat.; -30% ang. res. at 4 keV	140 mm + 120 mm drift, 3×5mm exit $3.4 \times 10^{-4} sr$	absolute, Ramsaer-like high-energy experiment; double scattering cell, no retarding-field analyzer
Ariyasinghe *et al* (2004)[19]	300-4000 eV, 4% total	245 mm, $\phi = 1.0$ mm	absolute, linear transmission, with retarding-field analyzer
Garcia and Manero (1998)[18]	400-5000 eV, 2% stat., 3% syst.	70-127 mm, $\phi = 1.0$ mm, $0.35 \times 10^{-4} sr$	absolute, linear transmission, with retarding-field analyzer
Barbarito *et al* (1979)	0.05-16 eV	4 cm, $\phi = 1.0$ mm	absolute, linear transmission, electrostatic gun
Sueoka and Mori (1986)	1-400 eV	6.38 mm (7.17 mm effective)	normalized, linear, magnetic guiding, positron and electron scattering measurements
Floeder *et al* (1985)	5-400 eV, 5% stat., 3% syst.		absolute, linear transmission, weak magnetic focusing, positron and electron scattering measurements

- The recommended values were obtained as weighted average values out of all experiments considered, with weight equal to the total experimental errors declared.
- The high energy limits of the experiments—above 100 eV by Nishimura
- and Sakae and above 1000 eV by Zecca et al. were excluded from the averaging procedures. As this procedure is identical to the one used in the Landolt–Bornstein review, the set of data is also identical in the energy range 0.1–1000 eV.
- At low energies, in the range of 0–0.1 eV, the TCS (which is equal to the integral elastic cross sections) is based on the modified effective range model by Fedus and Karwasz obtained from elastic differential measurements by Allan and checked with momentum transfer cross sections obtained from the analysis of electron swarm parameters.
- The uncertainty on such an evaluation is ±5%.
- At high energies, in the range of 1–4 keV, our recommended
- values are based on measurements by Ariyasinghe et al. who used a linear transmission method with a retarding-field analyzer.

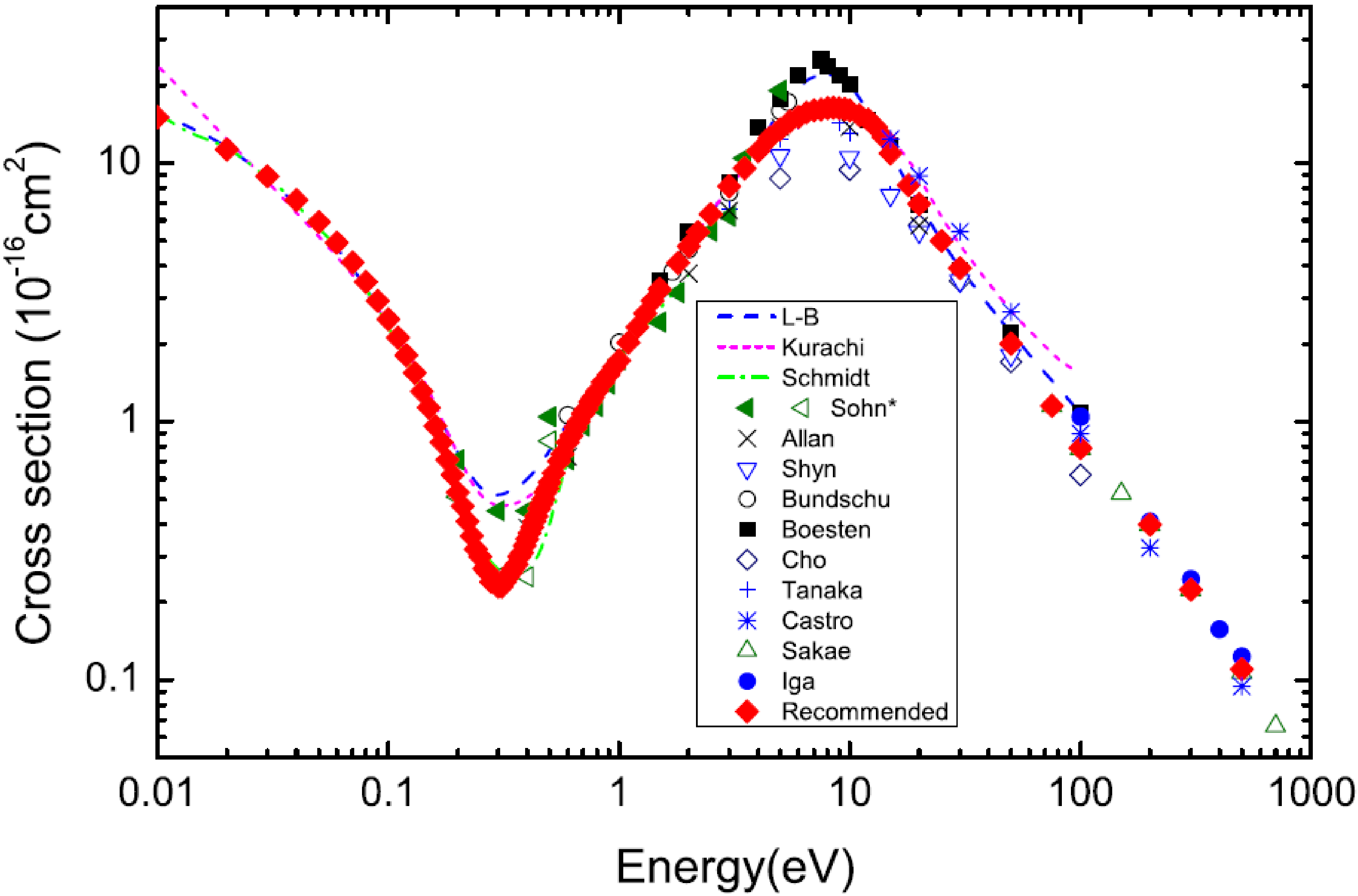
2. Elastic scattering cross section



Authors	Energy & angular Range measured	Remarks
Boesten & Tanaka (1991)	1.5-100 eV 10°-130°	spectrometer, normalization , Phase shift analysis
Bundschu et al (1997)	0.6-5.4 eV 12°-132°	magnetic filed angle changer , Phase shift analysis
Cho et al (2008)	5-100 eV 10°-180°	magnetic filed angle changer
Iga et al (2000)	100-500 eV 10°-135°	electrostatic analyzer, Manual extrapolation following Sakae
Sakae et al (1989)	75-700 eV 5°-135°	Cylindrical mirror analyzer, Extrapolated by fitting the square of t he Legendre polynomials to the experimental values.
Shyn & Cravens (1990)	5-50 eV 12°-156°	electrostatic analyzer, Exponentially extrapolating to 180°
Sohn et al (1986)	0.2-5 eV 15°-138°	spectrometer, normalization , Phase shift analysis
Allan(2007)	0.4-20eV 10°-180°	magnetic filed angle changer

- After collecting available elastic cross sections measured experimentally, the following data sets are excluded from the further considerations:
  - ✓ relative measurements
  - ✓ data with no uncertainties
  - ✓ the data points which are too far off the general pattern
  - ✓ the data points which have no other data points overlapped in the angular and energy region of interest
- We agree the average of 8 all data to derive the recommended data sets
- We estimated the uncertainties of the recommended data

3. Momentum Transfer cross section



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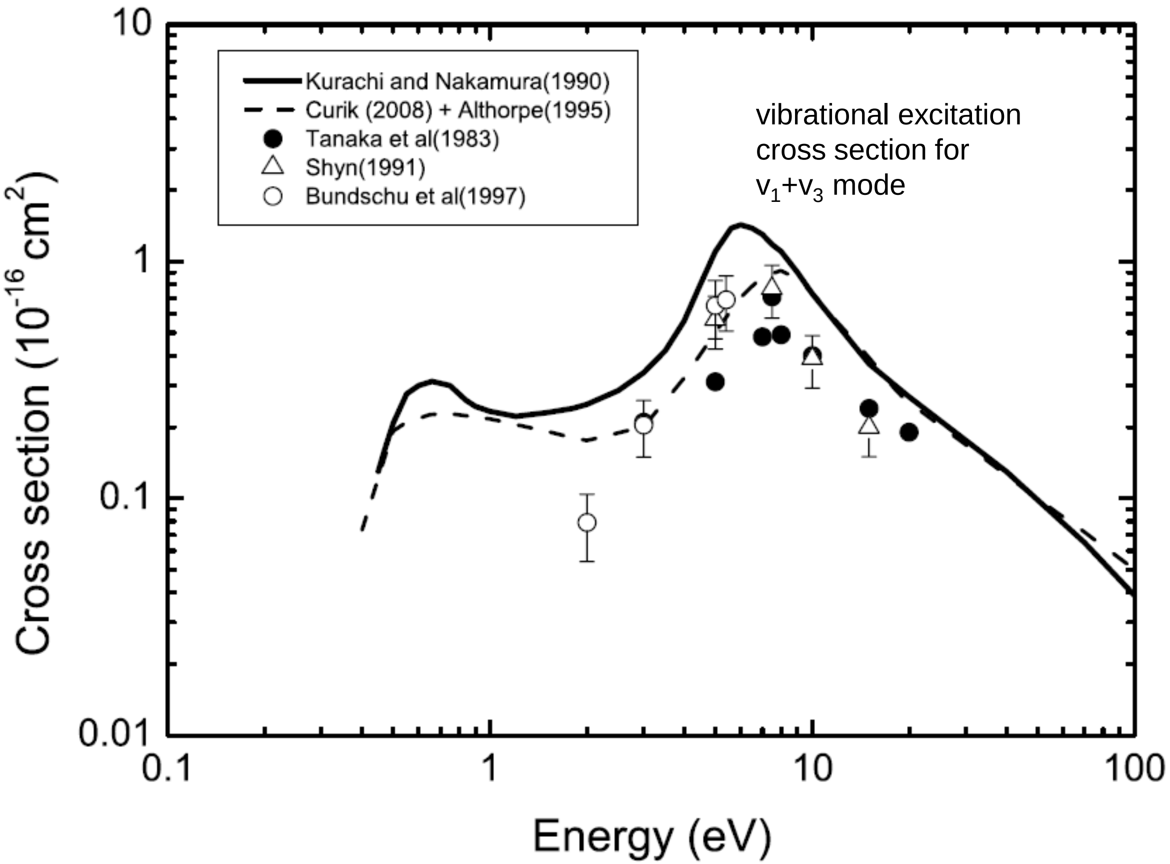
Sohn et al (1986)	0.2-5 eV 15°-138°	spectrometer, normalization , Phase shift analysis
Allan(2007)	0.4-20eV 10°-180°	magnetic filed angle changer
Castro(2010)	0-180	scaling quasi-free scattering model
Kurachi(1990), Schmidt(1991)		swarm analysis

- Similar confidence criteria apply to momentum transfer as to integral elastic cross sections.
- Additionally, momentum transfer cross sections can be obtained from analysis of swarm experiments
- Momentum transfer cross sections (MTCS) are more sensitive to uncertainties at high scattering angles than the integral elastic cross sections. Therefore, experiments at high angles are important.

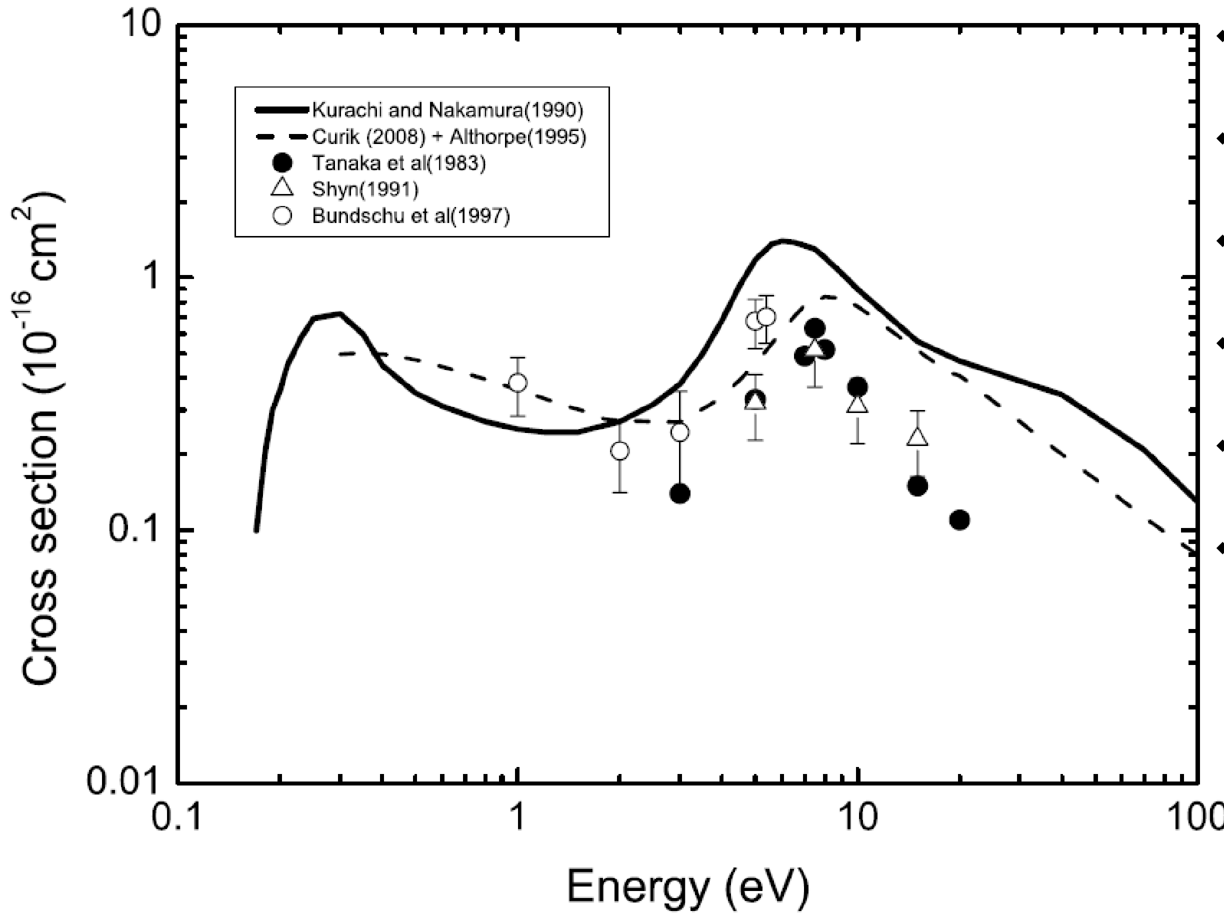
- 0.001 eV – 1 eV on the modified effective range theory analysis of elastic differential, and of total cross section by Fedus and Karwasz.
- 1 eV – 12 eV on swarm-derived data by Kurachi and Nakamura, which are in agreement with recent beam experiment by Allan in $0-180^{\circ}$ angular range;
- 15 eV - 50 eV on the recommended Landolt- B¨Ornstein data;
- 75 eV – 300 eV on beam experiment by Sakae et al.
- We estimate the uncertainty bar on the recommended values in the whole energy region considered as $\pm 10\%$.

- In the very-low energy region, modeling of swarm parameters is very sensitive to the correct choice of MTCS.
- However, due to the high value of the dipole polarizability, the applicability of this method in methane is limited to energies below 0.5eV. (the Ramsauer–Townsend (R–T) minimum of the MTCS)

4. Vibrational Excitation cross section



- Tanaka83: J. Phys. B 16 (1983) 2861
- Shyn91: J. Phys. B 24 (1991) 5169
- Bundschu97: J. Phys. B 30 (1997) 2239
- Curik08: J. Phys. B 41 (2008) 115203
- Althorpe95: J. Phys. B 28 (1995) 4165
- Kurachi90: Proc. 13th Symp. on Ion Sources and Ion-Assisted Technoligy, Tokyo (1990) 205



- ❖ Tanaka83: J. Phys. B 16 (1983) 2861
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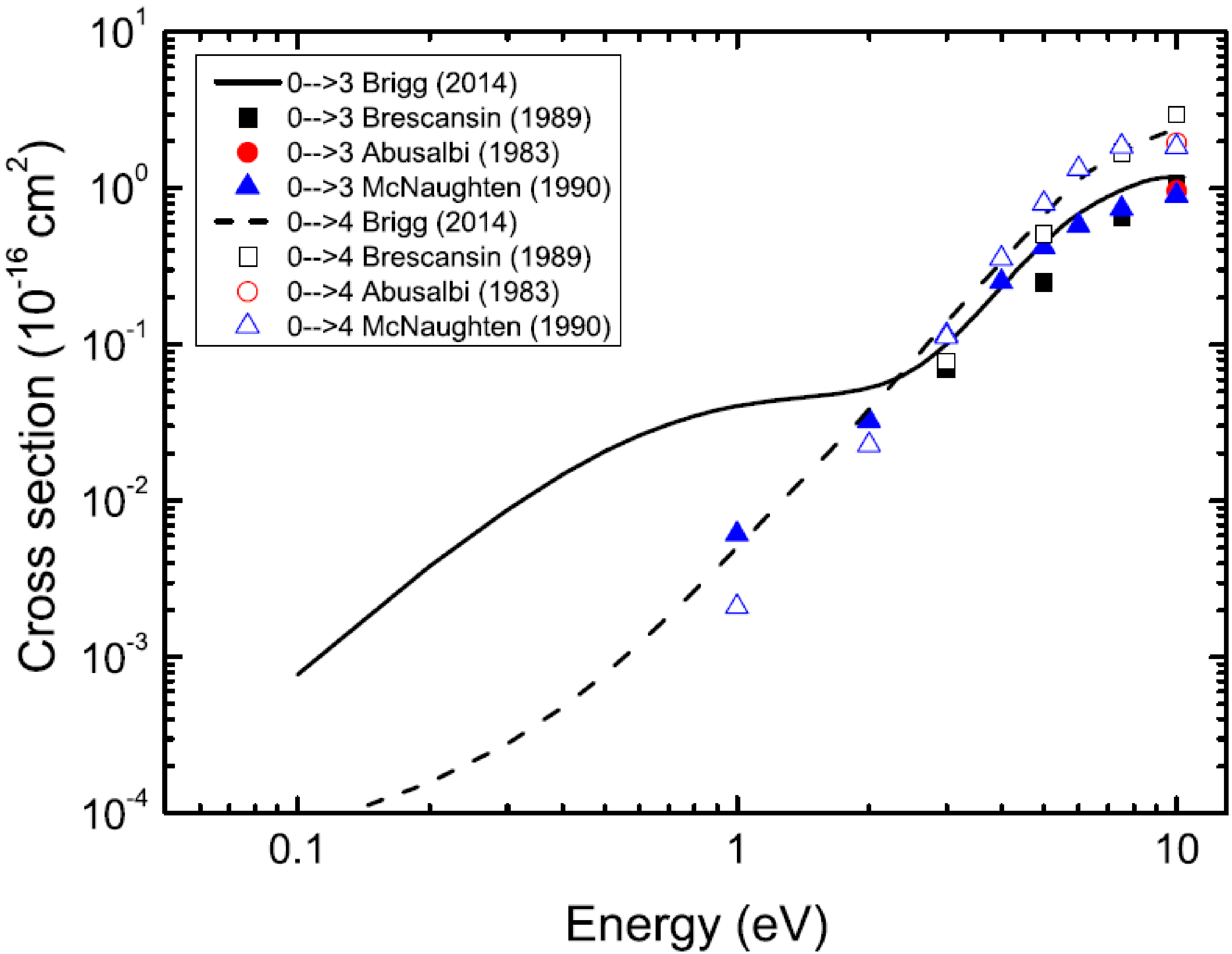
- ❖ Althorpe95: J. Phys. B 28 (1995) 4165
- ❖ Kurachi90: Proc. 13th Symp. on Ion Sources and Ion-Assisted Technoligy, Tokyo (1990) 205

Source	Method	Energy (eV)
Althorpe et al (1995)	multi-center DFT-HF for target and CIDRE for scattering	0.3-12
Cascella et al (2001)	Close-coupling Schrödinger equation, non-adiabatic couplings have been approximated	0.3-12
Curik et al (2008)	Lippmann-Schwinger equation, discrete momentum representation, optical potential for incident electron, normal modes for vibrations.	0.5-20
Kurachi et al (1990)	Swarm method.	0.5-100
Tanaka et al (1983)	Crossed beam, spectrometer, normalization Measurement angle: 10-130	
Shyn et al (1990)	Crossed beam, electrostatic analyzer. Measurement angle: 0-168	5-50
Bundschu et al (1997)	Crossed beam, magnetic field angle changer. Measurement angle: 12-132	0.6-5.4

- There is insufficient experimental data on vibrational excitation cross sections to cover the energy interval from 0.1 eV to 20 eV.
- Three different experiments gave results, which disagree with each other by between 10% and 100%.
- We recommend the swarm-derived cross sections for electron collision vibrational excitations of the methane molecule. The uncertainties in vibrational excitation cross section are about 15% at its peak and higher elsewhere.
- The swarm-derived cross sections
  - Analytical procedures for calculating these swarm parameters from electron collision cross section data, such as Boltzmann equation analysis and Monte Carlo calculation, are well established.

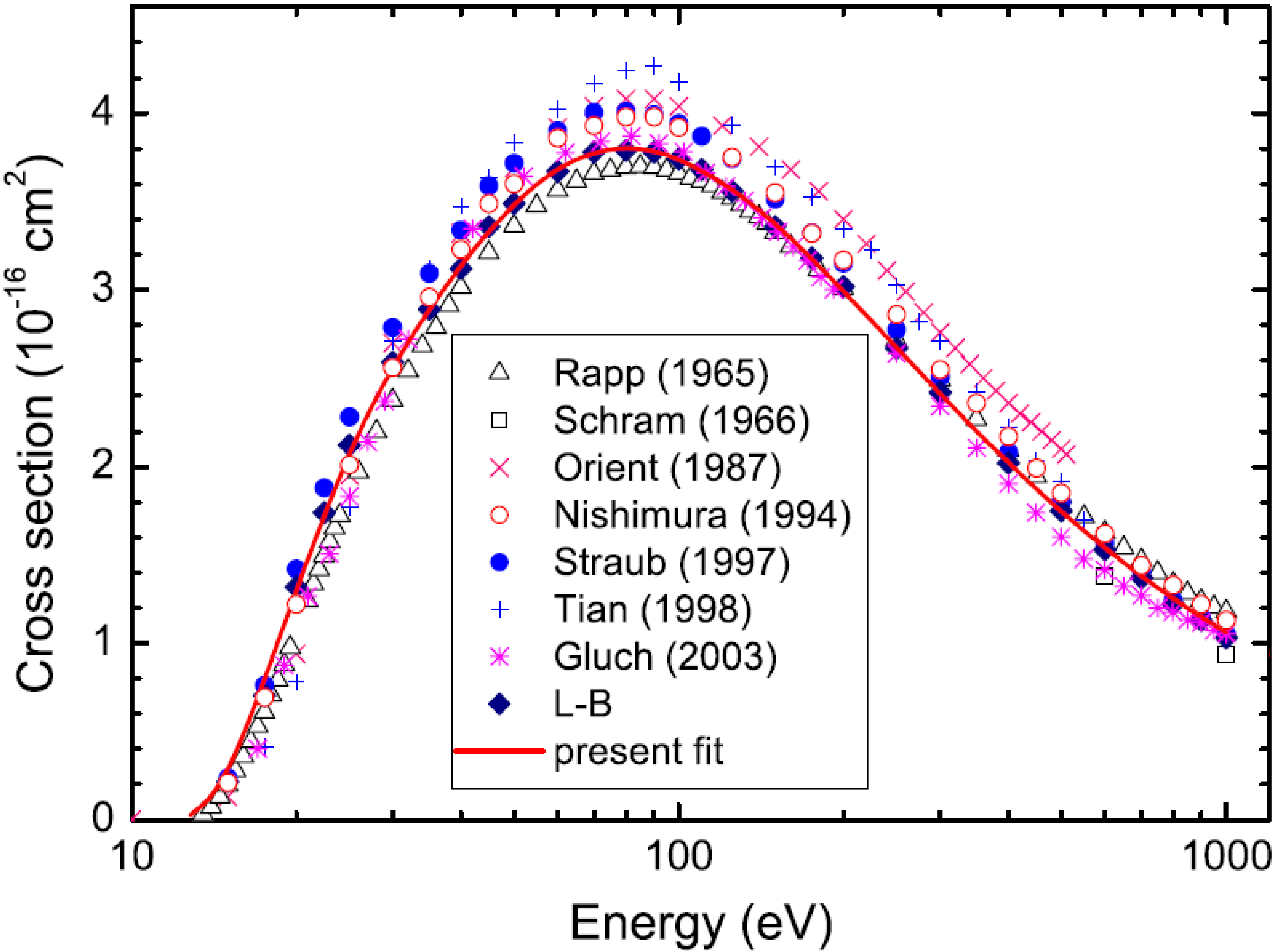
5. Rotational Excitation cross section

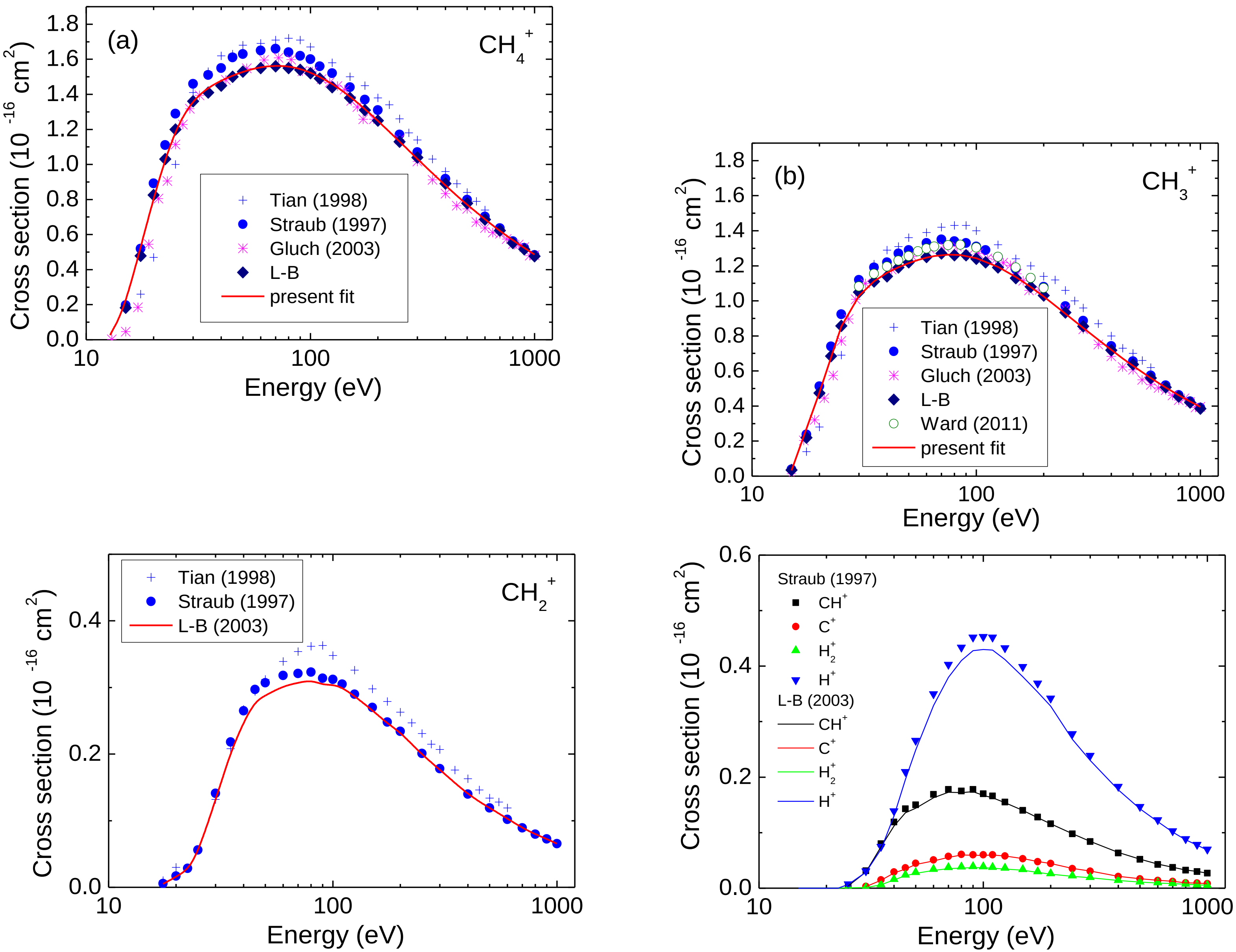


Source	Method	Energy (eV)
Müller et al. (1985)	Crossed Beam DCS in the range 15 – 150◦ at 0.5 and 7.5 eV, and for 75 – 150◦ at 5 and 10 eV. They give no integral cross Section.	0.5, 7.5, 5, 10
McNaughten et al (1990)	Close-coupling calculation, adopted a more elaborate model of the interaction potential	1-20
Brescansin et al. (1989)	Schwinger multichannel variational method	3-20
Abusalbi et al. (1983)	Close-coupling calculation.	10
Brigg et al(2014)	R-matrix study of rotational excitation below 10eV	0.1- 10

- The lowest non-vanishing multipole moment is the octupole moment. Therefore,in collisions where vibrational modes are not excited, when the initial rotational state is J = 0, the possible final rotational states should have J ≥ 3.
- Recently Brigg et al.(2014) have performed an R-matrix study of rotational excitation below 10 eV.
- Their results for the total cross section are in reasonable agreement with those of Brescansin et al.(1989) and Abusalbi et al.(1983)
- we recommend to use the data by Brigg et al(2014) because they are obtained in high-accuracy calculations and agree reasonably well with the previous calculations.
- The agreement with the experimental and other theoretical results suggests that the uncertainty of the calculated results is probably about 10%.

6. Ionization cross section

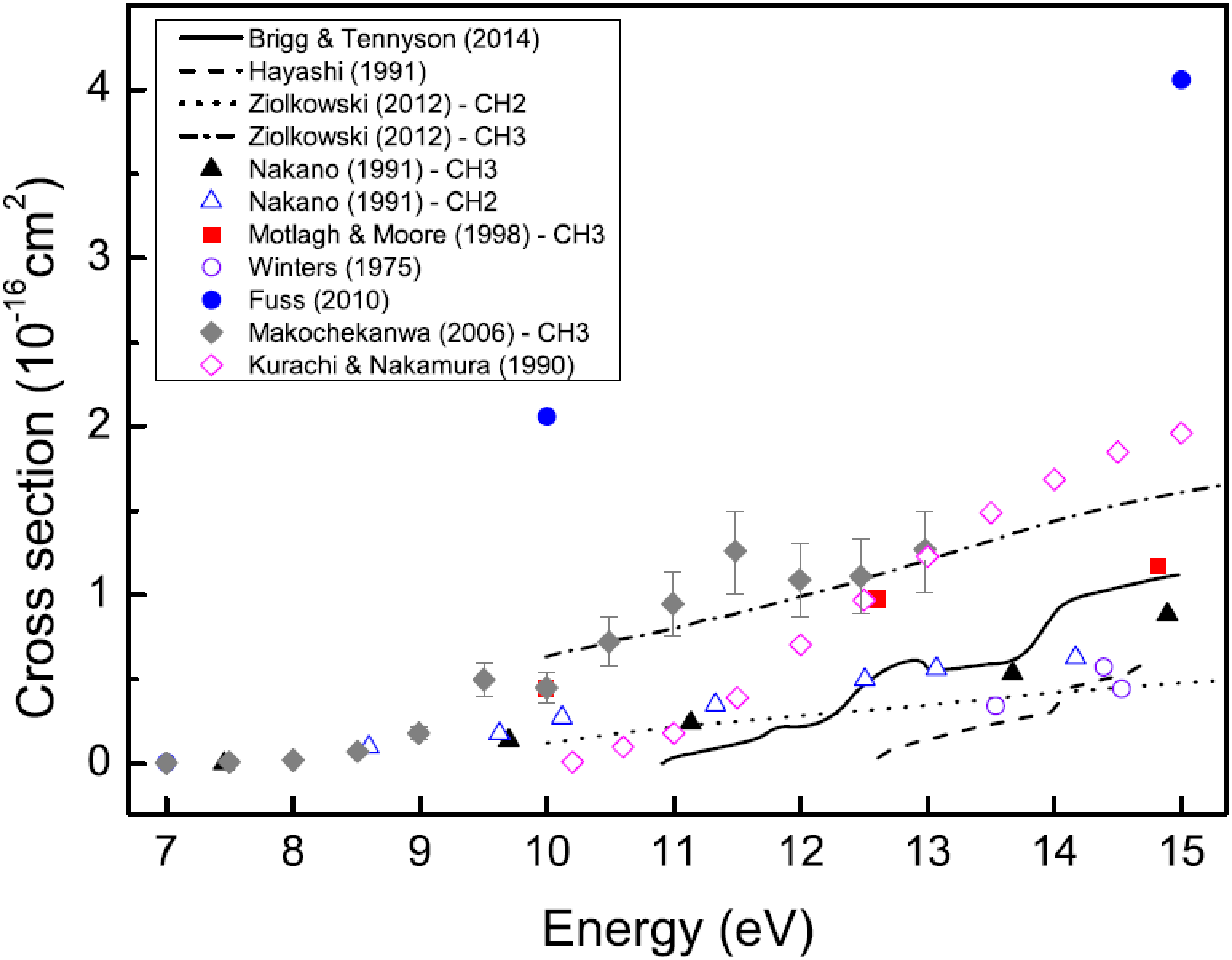



(a)
CH4+
Tian (1998)
Straub (1997)
Gluch (2003)
L-B
present fit
Cross section (10 -16 cm2)
Energy (eV)
(b)
CH3+
Tian (1998)
Straub (1997)
Gluch (2003)
L-B
Ward (2011)
present fit
Cross section (10 -16 cm2)
Energy (eV)
CH2+
Tian (1998)
Straub (1997)
L-B (2003)
Cross section (10 -16 cm2)
Energy (eV)
Straub (1997)
CH+
C+
H2+
H+
L-B (2003)
CH+
C+
H2+
H+
Cross section (10 -16 cm2)
Energy (eV)

Source	Method	Energy (eV)
Rapp and Englander-Golden (1965)	Pressure measurement, Normalized their CH4 results to their own absolute measurements in H2	Threshold - 1000
Schram(1966)	Pressure measurement using McLeod gauge.	600-12000
Orient(1987)	Crossed beam and relative flow technique normalized value.	10~500
Nishimura(1994)	Parallel plate ion collector method with a magnetically confined linear electron beam. absolute cross sections with an 8% systematic uncertainty	15~3000
Straub(1997)	Time-of-flight and position-sensitive detector. the data were put on absolute scale by pressure measurements	15~200
Gluch(2003)	Partial cross sections using a double-focusing mass spectrometer and normalized at 100 eV to the total cross section by Rapp and Englander-Golden	11 ~ 1000
Tian and Vidal(1998)	Time-of-flight spectrometer and normalized their partial cross sections to the Ar+ cross section of Straub et al	17~600
Lindsay(2003)	Based on Straub et al but are lower by about 10% for all ions at 100 eV and by 1 – 2% at 1000 eV	15~1000

- ❖ Recommended cross sections by Lindsay and Mangan from Landolt–Bornstein collection are based on Straub et al. but are lower by about 10% for all ions at 100 eV and by 1%–2% at 1000 eV.
- ❖ We adopted the database of Lindsay and Mangan also as our present recommended data, see A rough evaluation of the uncertainties is $\pm$5% for total ionization cross section and $\pm$10% for partial cross sections.

7. Dissociation cross section

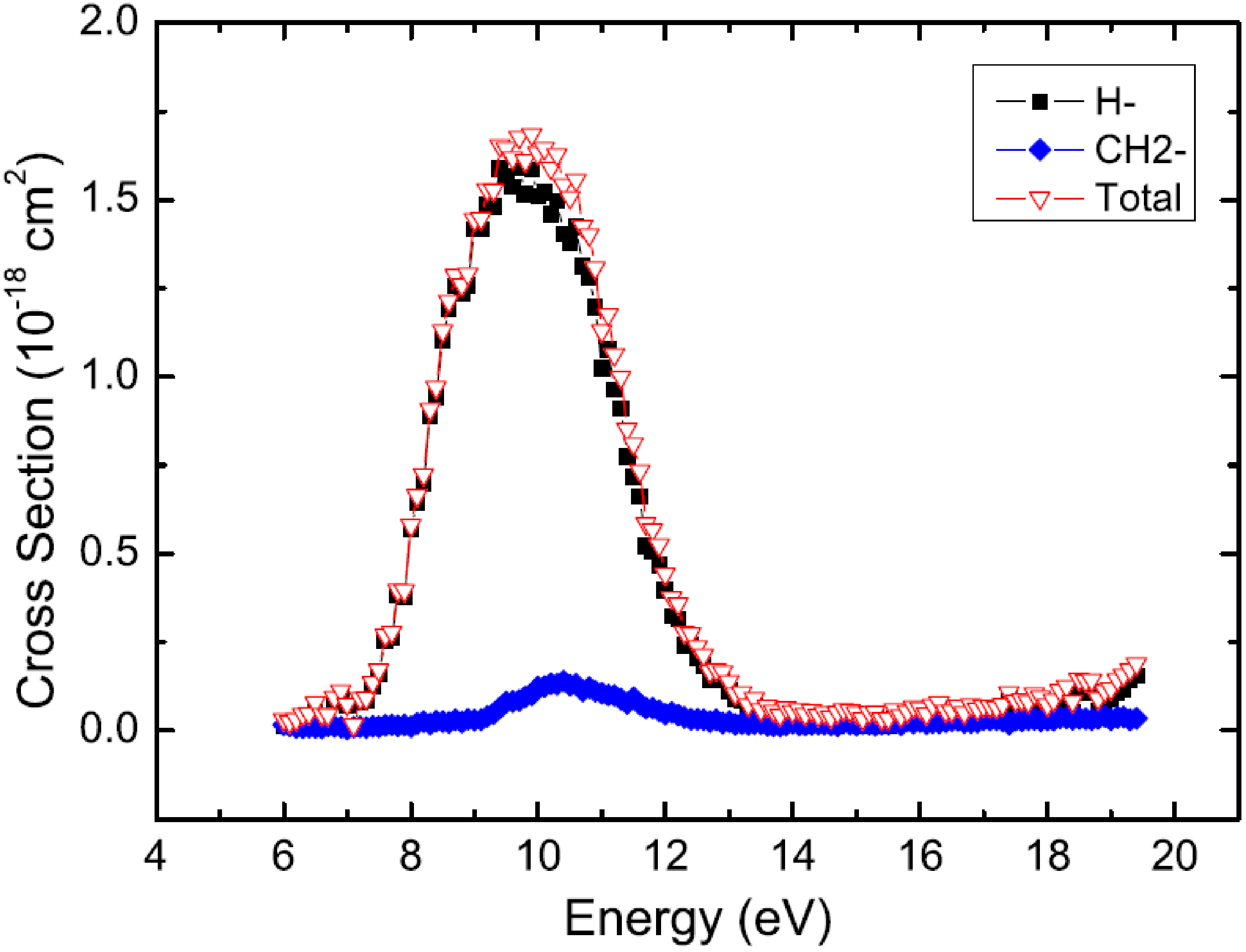


- Electron impact dissociation of methane, and other molecules, generally proceeds via electron impact electronic excitation.
- In particular, for methane, all the low-lying electronic states are dissociative so are only very short lived when excited by electron impact.
- In order of increasing energy threshold, the dissociation products that need to be considered are CH3 + H (channel 1), CH2 + H2 (channel 2), CH2 + 2H (channel 3), CH + H2 + H (channel4)
- In practice, there are no available measurements that distinguish between channel 2 and channel 3.

- At this time, it is not possible to make a clear recommendation for either total or partial electron-impact dissociation cross sections for methane.
- This problem, which is important for a number of topics including plasma-aided combustion, clearly requires further study.

Source	Method	
Fuss et al(2010)	based on electron beam measurements (Nakano et al. and Makochekanwa et al)	recommended
Nakano et al. (1991)	electron beam measurements	Measurement
Makochekanwa et al.(2006)	electron beam measurements	Measurement
Winstead et al(1993)	ab initio calculations	Calculation
Hayashi(1991)	swarm studies	Measurement
Kurachi and Nakamura (1990)	swarm studies	Measurement
Ziołkowski et al (2012)	ab initio calculations	Measurement
Brigg et al.(2014)	R-Matrix	Calculation
Winter (1975)	electron beam measurements	Calculation
Motlagh and Moore(1998)	electron beam measurements	Measurement

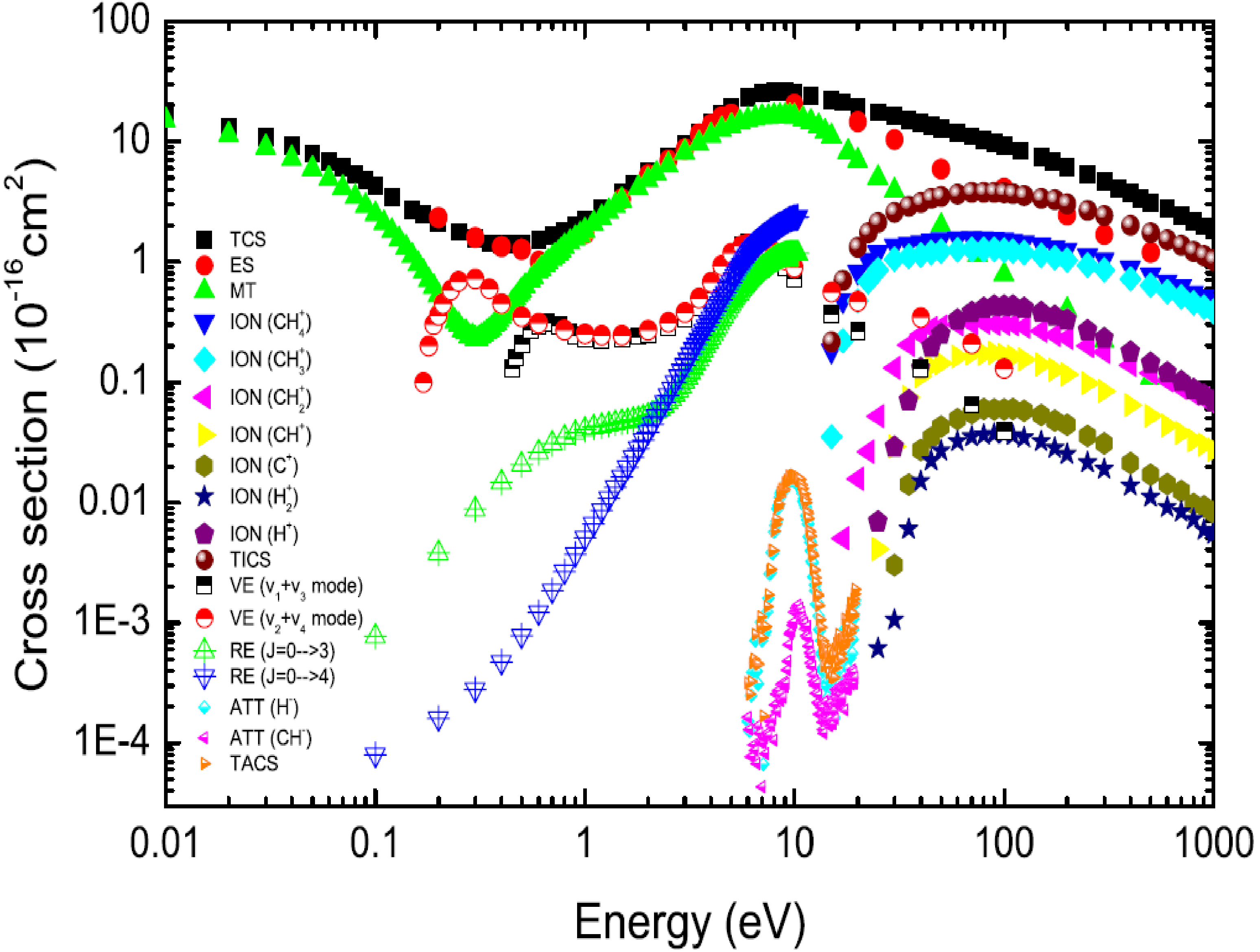
8. Dissociative electron attachment cross section



Source	Method	
Sharp and Dowell(1967)	Using the total ionization method and normalized their data to the positive ion cross section	H^-,CH_2^-
Rawat et al.(2008)	Absolute result	H^-,CH_2^-
Hoshino et al (2011)	Relative results	H^-, C^-, CH^-, CH_2^-, CH_3^-

- ❖ Peak positions of CH−2 of Sharp and Dowell(1967), Rawat et al.(2008), and Hoshino et al.(2011)agree very well.
- ❖ We recommend the data of Rawat et al.(2008) because they are much more recent and used a more advanced apparatus compared to the experiment of Sharp and Dowell and Rawat et al.(2008) reported uncertainty, while Sharp and Dowell(1967) did not.

The summary of cross section for electron collisions with methane



- ✓ There is considerable variation in the reliability of the available data.
- ✓ Total, momentum transfer, elastic scattering, and ionization cross section
  - ➢ it is possible to recommend values over an extended energy range with small uncertainties, typically 5%–10%.
- ✓ For electron impact rotational excitation
  - ➢ we rely on predictions from ab initial calculations. Because of the high symmetry of methane, these cross sections are small and hard to determine empirically
  - ➢ but experimental work on this process would be welcome.
- ✓ For the vibrational excitation cross sections
  - ➢ We recommend the vibrational excitation cross sections determined from swarm measurements but note that this is only and indirect measurement for which it is hard to establish true uncertainties
  - ➢ Reliable beam measurements of this process would be very helpful.

- ✓ Electron impact dissociation of methane is an important process
  - ➢ but we are unable to recommend a good set of data for this process.
- ✓ The dissociative electron attachment process
  - ➢ we recommend using the most recent experimental data but are not able to provide estimated uncertainties.
- ✓ This evaluation is the first in series of systematic evaluations of electron collision processes for key molecular targets

Cross Sections for Electron Collisions with Methane

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Cross section data are compiled from the literature for electron collisions with methane (CH_4) molecules. Cross sections are collected and reviewed for total scattering, elastic scattering, momentum transfer, excitations of rotational and vibrational states, dissociation, ionization, and dissociative attachment. The data derived from swarm experiments are also considered. For each of these processes, the recommended values of the cross sections are presented. The literature has been surveyed through early 2014. © *2015 AIP Publishing LLC.* [http://dx.doi.org/10.1063/1.4918630]

Key words: attachment; dissociation; electron collisions; evaluation; ionization; total cross sections.

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Summary of Evaluation Procedures

Preparatory stage

- Review of previous evaluation paper
- Collection of new paper.
- Define working Scope
- Contents of report
- To shard working part

Evaluation stage

- analysis method of experiment and theory (characteristics, limitation, uncertainty, method)
- Comparisons of different research group
- Combine different collision processes

Certified stage

- Check uncertainty
- Define recommended data of each collision processes
- Agreement of each evaluator

Certified stage

Calculation of Uncertainty

$x_1, x_2, x_3 \ldots\ldots x_n$ (independent value)

$$\bar{x} = \frac{x_1 + x_2 + \ldots + x_n}{n} \text{ (average)}$$

$$s(x) = \sqrt{\sum_{i=1}^{n} \frac{(x_i - \bar{x})^2}{n-1}}, \text{(standard deviation)}$$

$$s_p^2(x) = \frac{\sum_{i=1}^{n} v_i s_i^2}{\sum_{i=1}^{n} v_i}, \text{(combined standard deviation, } v_i \text{ : degree of freedom)}$$

$$\mathrm{u_s} = \frac{a}{\sqrt{3}}, \text{(systematic effects)}$$

$$\mathrm{u_m} = \sqrt{\frac{s_p^2(x)}{n} + u_s} \text{ (combined standard uncertainty)}$$

$$\mathrm{U} = \mathrm{k} \times u_m \text{(additional uncertainty)}$$

Evaluation tip

- **TCS, MTCS, VECS: Ramsauer-Townsend (R-T) minimum, To need swarm analysis.**
- **MTCS & ESCS : To need 0-180 degree DCS measurement data.**
- **TICS,PICS : To check experimental method (source, collision region, detection)**
- **DNCS : No experiment data, To need calculated EXCS.**
- **EXCS : No experiment , to need calculation data and uncertainty**
- **DACE: To understand structure. How to get data from mass spectrum data.**

Currently evaluation

- **2014**
  - ✓ C_2H_2 evaluation will be done in next week.
- **2015**
  - ✓ NF_3 evaluation still doing
- **2016**
  - ✓ CO_2,CO or NO, NO_2