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

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Data needed:

1. Total cross section
2. Partial cross sections:

- elastic scattering $\text{e}+A \rightarrow \text{e}+A$
- rotational excitation $\text{e}+\text{CH}_4 \ (J=0) \rightarrow \text{e}+\text{CH}_4 \ (J=2)$
- vibrational excitation $\text{e}+\text{AB}(v=0) \rightarrow \text{e}+\text{AB}(v>0)$
- electron attachment (dissociative) $\text{e}+\text{AB} \rightarrow \text{A}^- + \text{B}$
- electronic excitation $\text{e}+\text{A} \rightarrow \text{e}+\text{A}^*$
  - emission lines: $\text{A}^* \rightarrow \text{A} + h\nu$
- neutral dissociation $\text{e}+\text{AB} \rightarrow \text{A} + \text{B} + \text{e}$
  - emission from dissociation $\text{e} + \text{AB} \rightarrow \text{A}^* + \text{B} + \text{e} + h\nu$
- ionization $\text{e}+\text{A} \rightarrow \text{A}^+ + 2\text{e}$
- dissociative ionization $\text{e}+\text{AB} \rightarrow \text{A} + \text{B}^+ + 2^\circ$
- ionization into excited states $\text{e} + \text{A} \rightarrow (\text{A}^+)^* + 2\text{e}$
¿ITER: electron T and power irradiated?

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

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

Guillemaut et al. Nucl. Fusion (2014)
ITER: wall sputtering

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

S. Brezinsek¹, M.F. Stamp², D. Nishijima³, D. Borodin¹, S. Devaux⁴, K. Krieger⁴, S. Marsen¹, M. O’Mullane⁵, C. Bjoerkas⁶, A. Kirschner⁷ and JET EFDA contributors⁸
Plasma temperature ← integral cross sections

V. Godyak, Sendai 2006
Radiation damage in biological tissues

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.

Fig. 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.

Experimental methods: total attenuation method \( I = I_0 \exp(-\sigma n L) \)

precision <5%, unless...

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
Hydrogen – total: experiment vs theory

![Graph showing cross section vs incident energy for hydrogen.]

Mark C. Zammit, Jeremy S. Savage, Dmitry V. Fursa, and Igor Bray
Phys. Rev. Lett. 116, 233201
Experimental methods: total

Agreement generally within ±5%

Apart from high $E$ (and polar)

Total @ high energies: Born-Bethe fit

\[ \sigma(E) = A + B \ln E \]

CH₄

"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 \( (\sigma/a_o^2) (E/R) = A + B \ln (E/R) \) to TCS from Ariysainghe: \( A=52.31\pm17.3, B=232.2\pm8.6 \) where Rydberg constant is \( R=13.6 \) eV and the cross sections is expressed in atomic units \( a_o^2 =0.28\times10^{-20}m^2 \).
Experimental methods: elastic

I. Linert, B. Mielewska, G. King, and M. Zubek, PRA (2006)
Experimental methods: excitation (electronic, vibrational)

Experiments by:
M. Khakoo et al. (Fullerton California)
M. Allan (Freiburg University)/ J. Fedor (Prague)

accuracy ±20-40%
Experimental methods: ionization, total

Accuracy: ±10-15%

Zammit et al.. PRL 2016
Experimental methods: ionization (2)

Electron beam line  Deflection plates  Top plate  Faraday cup
Electron gun  Collimating apertures
Aperture and grid  Bottom plate

Accuracy: ±15-20%

B. G. Lindsay et al., JCP 129 (2004), S J King nad S D Price, JCP 134 (2011) 074311
Diffusion coefficients $\rightarrow$ electronic distribution function $n_e(r, v, t)$

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

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

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

Accuracy: $\pm 5-10\%$

**Non-unique** modelling

W. Roznerski (+), J. Mechlinska-Drewko (+), Y. Nakamura
Hydrogen – electronic excitation (modelling)

Figure 22: Total electron impact excitation cross section of singlet electronic states of $H_2$ from the $H_2(X^1\Sigma_g^+; v = 1)$ ground state, Eq. (95).

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

Ratko K. Janev, Detlev Reiter, Ulrich Samm

Jül-4105
Dezember 2003
ISSN 0944-2952
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 are shown as solid and open squares, respectively. The solid line represents the calculated cross sections, while the dashed line represents the experimental data.

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 $C^1\Pi_u$ (dotted) in the threshold energy region [53].

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, 2010
Electronic excitation – deconvolutions of spectra ($H_2$)

Figure 1. Experimental electron energy loss spectrum of $H_2$ (dots) taken at $E_0 = 20$ eV and $\theta = 20^\circ$ with a fitted spectrum (curve) and showing the positions of the unfolded vibrational manifolds as a result of the fitting.
Electronic excitation – dipole allowed and forbidden in $\text{H}_2$

Hydrogen – electronic excitation (theory)

Wang Yuan-Cheng, Ma Jia, Zhou Ya-Jun, Momentum-space multichannel optical model
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

Figure 2. ICSs for electron-impact excitation of the $C^3Π_u$ 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

Qualitative agreement between experiments, few theories

Ionization: semiempirical formulae

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

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

Normalized energies: \( t = \frac{E}{I_n}, \quad u_n = \frac{E_{\text{kin}}}{I_n} \)

Only two values needed from QCh

Partial (and total) ionization: \( \text{WF}_6 \)

- \( \text{WF}_5^+ \)
- \( \star \text{WF}_4^+ \) (x10)
- \( \circ \text{WF}_3^+ \) (x10)
- \( \blacktriangle \text{WF}_2^+ \) (x10)

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

Partial ionization: \( \text{CH}_4 \)

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

„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)
Dissociation into neutrals (H$_2$O)

Laser-induced fluorescence
Dissociation into neutrals (N$_2$O)

LeClair and McConkey JCP 99 (1993) 4566
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$_3$F...)

„Volatile organotellurides“

Motlagh and Moore JCP 109 (1988) 432

CF$_4$ → CF$_3$ + F
(4 sites possible)

CHF$_3$ → CHF$_2$ + F
(3 sites possible)

CH$_2$F$_3$ → CHF$_2$ + F
(2 sites possible)

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.
Dissociation into neutrals (CF$_3$COOH)

Cold deposition/ Electron irradiation/ Thermal desorption


No absolute values
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.
Kimura, Makochekeanwa data come from Suoeka, but they published data obtained with a higher guiding magnetic field.

Total: polar molecules

Fig. 2. Total scattering cross section, $Q_T$, of H$_2$O. 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.
Total: polar molecules (HCN)

As experimentalist I would believe more in theory than in experiment
Polar molecules \((e^+/e^- + \text{HCOH})\)

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*
Total and positronium formation cross sections for positron scattering from \( \text{H}_2\text{O} \) and \( \text{HCOOH} \)

Table 2. \( \text{H}_2\text{O} \) positron impact GTCS \((10^{-16} \text{ cm}^2)\). Numbers in parentheses are the values after the forward scattering effect correction. Errors are as explained in the text.

<table>
<thead>
<tr>
<th>Energy (eV)</th>
<th>GTCS</th>
<th>Energy (eV)</th>
<th>GTCS</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>63.756(194.45) ± 1.108(3.50)</td>
<td>25</td>
<td>8.985(11.38) ± 0.119(0.15)</td>
</tr>
<tr>
<td>0.75</td>
<td>50.348(151.73) ± 1.019(3.07)</td>
<td>26</td>
<td>8.598(10.81) ± 0.120(0.15)</td>
</tr>
<tr>
<td>1.0</td>
<td>39.751(128.87) ± 1.050(3.40)</td>
<td>27</td>
<td>8.694(10.86) ± 0.125(0.16)</td>
</tr>
<tr>
<td>1.25</td>
<td>35.157(115.00) ± 1.080(3.53)</td>
<td>28</td>
<td>8.569(10.64) ± 0.136(0.17)</td>
</tr>
<tr>
<td>1.5</td>
<td>29.762(95.30) ± 1.076(3.45)</td>
<td>29</td>
<td>8.432(10.41) ± 0.125(0.15)</td>
</tr>
</tbody>
</table>
„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

\[ \chi^2 \Sigma^+ (v=0) \rightarrow A^2 \Pi (v') \]

Cross section

Rate coefficients


Mott-Massey Schr. eq.
Beryllium

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

Positron + H₂: Bayesian analysis

Total cross section

![Graph showing cross sections for different positron energies with uninformative and informative priors.](image-url)
Bayesian analysis does not help much when experiments are uncertain.
Check of congruence: $\text{CF}_4$ (√)
Check of congruence: NH$_3$(x)
Check of congruence: $\text{CHF}_3 \times \times$
Experimental uncertainties for electron scattering on molecules

- Total, in majority cases, within ±5% but no data for BeH, WH₂, few WF₆
- Ionization: total within ±10%; in agreement with theories but partial ±15%
- Electronic excitation: good agreement between experiment and theory only of H₂
- Vibrational excitation: poorly understood at resonances
- Dissociation into neutrals desperately needed
Conclusions (II)

- Some targets possible for theory, other for experiments
- Solution: commissioning measurements;
- \( \text{NH}_3 \) vibrational and electronic excitation (Fullerton California?)
- \( \text{BeH}_2 \) elastic theoretical (Prague University?)
- \( \text{BeH}_2 \) electronic excitation (?)
- Polar molecules (\( \text{NH}_3 \)) 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