Evaluation of Data for Collisions of Electrons with Nitrogen Molecule and Nitrogen Molecular Ion

Summary Report of an IAEA Consultants Meeting

IAEA Headquarters, Vienna, Austria

5–6 December 2013

Prepared by

Hyun-Kyung Chung and Nigel J. Mason

February 2014
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Abstract
A Consultants’ Meeting (CM) on Evaluation of Data for Collisions of Electrons with Nitrogen Molecules and Nitrogen Molecular Ions was held at IAEA Headquarters in Vienna, Austria, from 5th to 6th December 2013. The meeting was organized in collaboration between the European eMOL project led by Prof N. J. Mason of the Open University, UK, and the Atomic and Molecular Data Unit of the IAEA. Seven experts from six countries participated in the meeting to evaluate currently available electron scattering data for nitrogen and nitrogen molecular ions and to develop general guidelines for data evaluation as a structured small group activity.

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1. Introduction

A Consultants’ Meeting (CM) on Evaluation of Data for Collisions of Electrons with Nitrogen Molecules and Nitrogen Molecular Ions was held at IAEA Headquarters in Vienna, Austria, from 5th to 6th December 2013. The meeting was organized in collaboration with the European eMOL project led by Prof N. J. Mason of the Open University, UK, and the Atomic and Molecular Data Unit at IAEA to evaluate the current status of electron collisional data for nitrogen molecules and molecular ions and recommend the best available data sets.

The Atomic and Molecular Data Unit (AMD Unit) is interested in providing evaluated and recommended data for atomic, molecular and plasma-surface interaction processes relevant to fusion and other plasma applications. The EU-funded eMOL group aims to carry out evaluations of electron-molecule scattering data and to develop the methodology for evaluations as a group activity. The Unit has collaborated with the eMOL group in order to motivate the atomic and molecular physics community to develop guidelines for critical assessment and recommendation of scattering data.

Nitrogen is often used as an edge plasma cooling gas in tokamak experiments. The nitrogen is injected as a molecular gas, which rapidly dissociates and ionizes. For the modelling of those experiments, due to the lack of recommended data for electron – nitrogen molecule scattering processes the nitrogen source is treated as pure atoms. As a consequence one cannot very well simulate the line emissions in the injection region. In order to address the data needs, seven participants from six countries were invited to evaluate currently available electron scattering data of nitrogen and nitrogen molecular ions.

A secondary objective of this meeting is to share experiences and develop general guidelines for data evaluation as a structured small group activity.

The evaluation procedure specified by eMOL as a group activity is as follows. Articles and data subject to the evaluation were assembled at Open University and distributed to the evaluators prior to the meeting. For each process, three evaluators were assigned to review the assembled data sets. During the two day meeting, evaluators discussed the reviewed data and worked towards consensus on the uncertainties and the recommended data set. At the conclusion of the meeting the evaluators returned home with a well-defined set of tasks to finish the evaluation. One follow-up meeting (video conference) is planned to review the homework and resolve remaining issues. Upon the completion of evaluation a journal article will be prepared for submission to European Physical Journal D, which has agreed to receive evaluations from the eMOL project. The eMOL project plans to conduct 12-15 such evaluations during the period of 2013-2015.

The following scattering processes of electrons with nitrogen and nitrogen molecular ions were reviewed:

- Total cross sections of electron – nitrogen molecule collisions
- Elastic cross sections of electron – nitrogen molecule collisions
- Momentum transfer cross sections of electron – nitrogen molecule collisions
- Vibrational and rotational excitation (resonances) cross sections of electron – nitrogen molecule collisions
- Ionization cross sections of electron – nitrogen molecule collisions
- Electron excitation and dissociation cross sections of electron – nitrogen molecule collisions
- Electron collisions with nitrogen molecular ions

This proceeding report contains short summaries of discussions on electron scattering data of seven processes in Section 2, and conclusions and recommendations on the general practices and guidelines of critical evaluation of electron molecule scattering data in Section 3. The list of participants is provided in Appendix 1 and the meeting agenda in Appendix 2.
2. Proceedings of the Meeting

The Head of Nuclear Data Section, Dr R. Forrest welcomed participants to the meeting and emphasized the importance of evaluation activities of atomic and molecular data in the context of fusion applications. Dr Braams described the meeting objectives that the AMD Unit is interested in developing data evaluation procedures in a systematic way. The role of nitrogen molecules in fusion plasma control was described. The evaluation of the currently available electron scattering data sets and proposed recommended data sets of seven processes are summarized in this section.

2.1 D. Field, G. Garcia and B. Antony: Total cross sections

The total cross-section is defined as the sum over all angles, that is 4\pi steradians, and for all processes for scattering of electrons by N\textsubscript{2}. Since N\textsubscript{2} has no dipole moment, at low energy below \approx 1 eV the great majority of the cross-section is elastic. There is a small contribution from rotationally inelastic quadrupole scattering that only becomes non-negligible at the very lowest energies of collision and is even then small. At higher energy, strong vibrationally inelastic scattering is encountered especially in the energy range 2 to 3 eV.

The energy scale for collisions has been set by electron time-of-flight measurements for N\textsubscript{2} scattering, aided by time-of-flight measurements involving electron scattering by O\textsubscript{2}. The energy scale so-determined has been shown to be consistent with calibrations involving Born-type vibrational excitation on other species than N\textsubscript{2}, such as CS\textsubscript{2}. The energy scale at low energy, below a few hundred meV is correct to within \pm 1 to 2 meV and, at around 2 eV and above, to better than \pm 5 meV. The accuracy of cross-sections is set essentially by absolute pressure measurement and a conservative estimate would be \pm 10%.

There have been a very large number of measurements of total scattering cross-sections reported in the literature between \approx 1.5 eV and 6 eV covering the range of energy on the so-called ‘resonance region’. Recommended data in this brief report supersedes those given by R. E. Kennerly [1], the data recommended in the review of Y. Itikawa [2]. There is in fact excellent agreement between these data and the accompanying recommended data with regard to absolute (maximum) cross-section values (amended from the meeting). However the energy resolution in the accompanying data is considerably better than in Kennerly as can be seen in the figure appended.

At lower energy the recommended data are those in M Sulc et al. [3]. Data at higher energy from the latter group (unpublished) match very well the data of Kennerly, reinforcing our choice of Kennerly’s data.

Recommended values for rotationally inelastic scattering are given by theoretical values given in M Sulc et al. [3]. Data for total scattering cross-sections are shown in the figure and the table which accompany this report. Data for momentum transfer cross-sections will be given at a later date. These require some computation from existing data on total and backward scattering data for N\textsubscript{2}.

2.2 G. Garcia, J. Tennyson and V. Kokoouline: Elastic cross sections

From the experimental point of view, elastic electron scattering cross sections from N$_2$ has been extensively studied [1-6] for low and intermediate energies (0.55-100 eV) and especially in the resonance region (1-5 eV). Buckman et al. [7] reviewed all these experiments in 2003 providing a recommended set of data with uncertainties, for the integral cross section (ICS) values, of about 20%. Itikawa’s review [12], published in 2006, maintained that recommendation and extended the ICS data up to 1000 eV by averaging the experimental results of Shyn and Carrigan and DuBois [8] and Rudd [9]. This study was incorporated with the calculations based on our IAM-SCAR [10] method which has demonstrated for a wide variety of molecular targets its reliability to provide integral cross section data for intermediate and high energies, i.e. from 10-10000 eV. This calculation showed a good agreement, within 20%, with the recommended values up to 1000 eV. It is recommended that calculated values should extend the range from 1000 to 10000 eV, maintaining Itikawa’s recommendation below 1000 eV.

Above 10000 eV recommended values tend to those derived from the Born approximation with an independent atom representation being therefore consistent with the expected asymptotic behaviour. Concerning differential cross section (DCS) data, the IAM-SCAR calculation agrees with experimental values (within 20 %) for energies above 30 eV although for small angles calculated data tend to be lower than the experimental ones, due to an internal normalization procedure [11]. Below 30 eV there is a general agreement between all the available experimental data but the IAM-SCAR, as expected, does not reproduce well the angular dependence of the cross sections. Consequently, recommended electron DCS are the calculated IAM-SCAR values from 30 to 10000 eV (verifying the low angle data for the lower energies) and the average of the experimental data for energies below 30 eV.

2.3 L. Alves, J. Tennyson and B. Antony: Momentum transfer

The (elastic) momentum-transfer cross section $Q_{MT}$ is the $(1 - \cos \theta)$ weighted, angle-averaged differential cross section for elastic scattering, where $\theta$ is the scattering angle, which can differ significantly from the elastic integral cross section $Q_{el}$, obtained by direct integration of the corresponding differential cross section over all angles.

In plasma modelling, input to the two-term electron Boltzmann equation requires both $Q_{MT}$ and the total momentum-transfer cross section $Q_{MT, tot}$, which includes the contribution of elastic, excitation and ionization processes. These cross sections appear naturally in the collision term in the Boltzmann equation, when the angular dependence of the velocity is expanded in Legendre polynomials. However, anisotropies in the inelastic cross sections are often neglected, introducing the approximation $Q_{MT, tot} \sim Q_{MT, eff} = Q_{MT} + Q_{exc} + Q_{ion}$, where the so-called effective momentum-transfer cross section $Q_{MT, eff}$ sums the elastic momentum-transfer to the integral cross sections for excitation $Q_{exc}$ and ionization $Q_{ion}$.

Compared are $Q_{MT}$, presented in the review papers of Itikawa [1] and Tabata et al [2], with the measurements of Linert and Zubek [3] and of Allan [4], the calculations of Shi et al [5] and of Feng et al [6], and the effective cross section $Q_{MT, eff}$ with the IST-LISBON database at LXcat [7].

Recommendation is for using the measurements of D. Field et al [8] below 0.8 eV, the data in [4] (for both elastic and vibrational excitation processes) between 0.8 and 5 eV, the data in [3] between 5 and 30 eV, and the values calculated in [5] above 100 eV. Further comparisons will be done with $Q_{MT, eff}$ of [7] and the reference data of [1].

2.4 V. Kokouline, D. Field and J. Tennyson: Vibrational and rotational excitation (resonances)

Vibrational excitation

The previous evaluation by Itikawa [1] was based on an earlier evaluation by Brunger, Buckman, and Elford [2]. Since these evaluations, new and more accurate experimental data have been published.

For the vibrational excitation from the ground $v=0$ to the first excited level $v=1$ of $N_2$, the recommended differential cross-sections are the ones measured by M. Allan [3]. In this study, the differential cross-section for the $v=0 \rightarrow v=1$ excitation was measured for scattering angles $\theta=45^\circ$, $135^\circ$, and $180^\circ$ for energies between 0.3 and 5.5 eV. The measured cross-section was not resolved
rotationally, i.e. a sum over all rotational levels has been measured. The region of resonances between 1.9 and 3.7 eV is well resolved. The same study gave also the differential cross sections for the ν=0 → ν=1 excitation as a function of the scattering angle for several values of scattering energies, E=0.8, 1.988, and 5 eV.

A special attention should be taken for energies near the N2⁺ (R²Σg⁺) resonance situated around scattering energy of 11.4 eV. This region has been carefully measured by Hoffmann et al. [4]. The available data are for seven scattering angles between 10⁰ and 180⁰ and for the energy interval between 11.35 and 11.95 eV. Therefore, these data are recommended for the region between 11.35 and 11.95 eV. The authors of the study provided also an analytical formula for the differential cross sections near the resonance with parameters obtained by a fit for the measured data. This formula can be used instead of numerical data obtained in the experiment.

Another experimental study by Linert and Zubek, [5] gives the differential cross sections for the ν=0 → ν=1 excitation as a function of the scattering angle between angles 0°=20° and 180° for four values of the scattering energy 5, 10, 15, and 20 eV. For the scattering energy of 5eV covered in both studies (by Allan and by Linert and Zubek), the two measurements agreed with each other within 20% accuracy. Therefore, the cross sections measured by Linert and Zubek are recommended for energies 10, 15 and 20 eV. For energies between 5 eV and 11.3 eV as well as between 12 eV and 20 eV, the recommended cross sections should be interpolated because Linert and Zubek do not provide data for intermediated energies (in contrast to the study by Allan, in which he reported the cross sections as functions of energies). The estimated uncertainty of the measurements is about 20%.

Rotational excitation

The previous evaluation by Itikawa [1] for rotational excitation from j=0 → j=2 and from j=0 → j=4 used the data from an earlier evaluation by Brunger, Buckman, and Elford, [2], which was based on swarm experiment data. Since these evaluations, another accurate theoretical calculation by Sulc et al. [6] has been published. The resulting cross section for the j=0 → j=2 transition is in good agreement with the previous evaluation for energies <1 eV. Therefore, we recommend to keep the data recommended by Itikawa [1] for the j=0 → j=2 and j=0 → j=4 transition.

New measurements and/or theoretical calculations of the rotational excitation of N2 are needed for low (<1 eV) and higher (>1 eV) energies starting with ground and excited rotational level of N2.


2.5 B. Antony, N.J. Mason, and L. Alves: Ionization

The partial and total ionization cross section for the ground state neutral N2 molecule by the impact of electron has been studied extensively by Itikawa [1], Lindsay and Mangan [2] and Tabata et al [3] in their reviews. These reviews are based on the experimental work of Straub et al [4], Rapp and Englander-Golden [5], Tian and Vidal [6], Hudson et al [7] and Krishnakumar and Srivastava [8]. Among all the reviews, Itikawa has done the most comprehensive work on N2 molecule, surveying literature till the end of 2003.

After going through previous reviews, evaluating existing data and deliberations through discussions, it was accepted during the meeting that the measurements of Straub et al [4] will be adopted as the recommended data set for partial (N2⁺, N⁺ and N2⁺) and total ionization cross section. Straub et al [4] have used a time-of-flight mass spectrometer to detect product ions within an uncertainty of 3.5%.
However, below 25 eV, the data reported by Straub et al is fragmentary and hence, the mean of Straub et al and Rapp and Englander-Golden [5] are considered in this energy range as they gives very good agreement with Straub et al. Above 50 eV, the data reported by Rapp and Englander-Golden is slightly on the higher side of the recommended data. Rapp and Englander-Golden [5] reports an uncertainty of 7% in their measurement. However, the overall uncertainties estimated are 5% for the production of N_2^+ and N^+ ions and 6% for the N”^+ ion [1]. It is to be noted that N” may also include N_2”^+, as measurements cannot resolve same-charge-to-mass ratio fragments. However, a rough estimate by the authors [1] shows that the contribution may be quite small (< 0.5%) and hence can be neglected. For the production of other fragments like N”^+ + N, N_2”^+ + N and N” + N_2”, the data reported by Tian and Vidal [6] are considered in this energy range as they gives very good agreement with Straub et al. Rapp and Englander-Golden [5] reports an uncertainty of 7% in their measurement. However, the overall uncertainties estimated are 5% for the production of N_2^+ and N^+ ions and 6% for the N”^+ ion [1]. It is to be noted that N” may also include N_2”^+, as measurements cannot resolve same-charge-to-mass ratio fragments. However, a rough estimate by the authors [1] shows that the contribution may be quite small (< 0.5%) and hence can be neglected. For the production of other fragments like N”^+ + N, N_2”^+ + N and N” + N_2”, the data reported by Tian and Vidal [6] with an uncertainty of 10% was recommended. The recommended data set for the total ionization cross section was obtained by adding all the individual partial ionization cross sections with an estimated uncertainty of 5% [2].

Besides the works reported above, various other experimental and theoretical data also exist in the literature. Krishnakumar and Srivastava (with 10% uncertainty) and Hudson et al (with 5% uncertainty) have also measured total ionization cross section of N_2 by electron impact. Toth et al [9], Hwang et al [10] and Joshipura et al [11] have calculated total ionization cross section of e-N_2 using distorted wave Born approximation (DWBA), binary-encounter-bethe (BEB) and complex scattering potential-ionization contribution (CSP-ic) methods respectively. In general, all these data agrees reasonably well with the recommended data.


2.6 N.J. Mason, V. Kokouline and G. Garcia: Electron excitation and dissociation

There is a long standing controversy about the electronic excitation of nitrogen – with an unresolved disagreement between theory and experiment (and between different experiments) both for integral and differential cross sections for most of the energy range studied. The disagreement is both qualitative and quantitative and is attributable to the complexities of performing both the experiments and calculations. The main experimental difficulty lies in deconvoluting the contribution to the excitation cross section from each electronic state. The set of recent experiments by the group of Khakoo [4] provides the largest and most self-consistent set of differential and integral cross sections which are also in broad agreement with earlier data of Trajmar et al [5] and with the earlier recommended data of Itikawa [2].

However these data are not in good agreement with the most recent theoretical data, in particular the R-matrix method implemented within the UKRmol suite appears to overestimate the cross sections. This suggests that it is necessary to revisit the potential energy surfaces of N_2. It is also proposed that the most direct method for comparing experiment and theory is to use the theoretical data to simulate
Electron Energy Loss Spectra (EELS) at different energies and angles. This will avoid the need to deconvolute the experimental data.

Emission spectra from many of the electronic states of nitrogen are important in both the terrestrial and several planetary atmospheres. Ajello et al [3] have recently updated their earlier data [1] but there remain several inconsistencies with the EEL derived cross sections which provide challenges to current atmospheric models and plasma diagnostics.


2.7 J. Tennyson: Electron collisions with N₂⁺

There are many processes involving electron collisions with the nitrogen molecular ion for which there appears to be no data from either experiment or theory. These processes include electron impact rotational excitation, vibrational excitation and ionisation.

There are measurements for electron impact excitation from the ground X state to the second excited, B, state and a number of theoretical calculations on this process. Remarkably little has been published on excitation of the first excited, A, state by electron collisions even though these cross sections are actually contained in the various theoretical calculations that have been performed.

Dissociative recombination studies are difficult with N₂⁺ because it is hard to cool. There are a number of studies and the most recent storage ring results are recommended. These are not for 100% ground state N₂⁺ but at least the vibrational distribution is characterised. This work also yields branching ratios and parallel studies also provide data on electron impact dissociation.

3. Meeting Conclusions and Recommendations

Participants agreed that the recommended data set should be finalized for each process by the end of February and compiled by March with a video conference. The LXcat project (www.lxcat.net) or VAMDC project (www.vamdc.eu) will be explored as a possible “active database” hosting the recommended data so that data sets can be easily updated as data of better quality is available and archived with a version history.

The IAEA ALADDIN database (https://www.amdis.iaea.org/ALADDIN), not an active database, will host the most current recommended data set.

Participants shared their views on general guidelines and protocols of evaluation procedures of electron scattering data. The process of critical evaluation of data on wavelengths and energy levels developed by A. Kramida at NIST (Fusion Science and Technology, vol. 63, page. 313 (2013)) was discussed as an example of such a protocol.

This review of electron nitrogen scattering data was the fourth in the eMOL series. (Previous reviews were centered on water, tetrahydrofuran and CF₃I). These data reviews are revealing several requirements:
• A compilation of the data (ideally by a dedicated bibliometrician)
• The data best reviewed by a set of experts who can both review the data and propose recommended values.
• The need to establish an online database that is accessible through a universal access portal.
• The need to update the database and recommendations as subsequent papers are published.
• The eMOL project is developing this method and has established a wider ‘Review Board’. Face to face meetings are recognized as being an important part of such reviews with at least one such meeting per data review being recommended.
• Criteria for recommended data sets are identified:
• Recommended data sets should be self-consistent with the sum of individual cross sections being consistent with the total cross section and partial excitation / ionization cross sections being consistent with sum/integral cross sections.
• All data sets must contain uncertainty estimates - both for experimental and theoretical data. In the case of theoretical data it is necessary to provide a set of guidelines for the community. These guidelines must be adopted by publishers if they are to be effective.
• In many cases semi-empirical calculations may provide data sets on a more rapid timescale than more detailed calculations and where experiments are not practical (e.g for many radical species). Such methods must be benchmarked such that any systematic effects are recognized and uncertainty estimates quantified.

It was emphasized that the data compilations and recommended data should be integrated within larger atomic and molecular data compilations and online databases and portals such as the VAMDC initiative which is developing a ‘Roadmap’ for atomic and molecular data assembly, curation, recommendation and dissemination.

Currently, a few projects such as the NFRI (National Fusion Research Institute, Korea) methane evaluation or the European eMOL project are carried out to provide community agreed evaluation and recommendation of the electron scattering data. The experiences and findings from these evaluation projects should be shared and compiled to produce unified guidelines for group evaluation procedures.

It was recommended that the IAEA Atomic and Molecular Data Unit should take the coordinator role to produce a technical document to outline the general guidelines and protocols of evaluation procedures of electron scattering data. The scope of the document should be sufficiently technical so that it can be accepted by journals as a guideline for a peer review process.
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IAEA Consultants’ Meeting on Evaluation of Data for Collisions of Electrons with Nitrogen Molecule and Nitrogen Molecular Ion

5–6 December 2013, IAEA Headquarters, Vienna, Austria

Scientific Secretary: Ms Hyun-Kyung Chung
Chairman: Mr N.J. Mason

Agenda

Thursday, 5 December

Meeting Room: F0817

09:30 – 09:50  R. Forrest, B.J. Braams, N.J. Mason, H-K Chung: Welcome, meeting objectives, introduction of participants, administrative matters

09:50 – 11:00  D. Field, G. Garcia and B. Antony: Total cross sections

11:00 – 11:10  Break

11:10 – 12:20  G. Garcia, J. Tennyson and V. Kokoouline: Elastic cross sections

12:20 – 13:40  Lunch

13:40 – 14:50  L. Alves, J. Tennyson and B. Antony: Momentum transfer

14:50 – 16:00  D. Field, J. Tennyson and V. Kokoouline: Vibrational and rotational excitation (resonances)

16:00 – 16:10  Break

16:10 – 17:30  N.J. Mason, B. Antony and L. Alves: Ionisation

Friday, 6 December

Meeting Room: F0817

09:00 – 10:10  N.J. Mason, V. Kokoouline and G. Garcia: Electron excitation and dissociation

10:10 – 11:30  J. Tennyson: Electron collisions with N₂⁺

11:30 – 12:30  All: Review processes not finished earlier

12:35 – 14:00  Lunch

14:00 – 16:00  All: Review, homework assignments and sketch of meeting report

17:00 –  Close of meeting