CHAPTER 6   IONIZATION OF N_2

"In this chapter, we investigate the interactions of electrons with molecular N_2 over an extensive energy range, starting from the ionization threshold and extending to 5 KeV. We have computed inelastic CSs (Q_{inel}), elastic CSs (Q_{el}) and total CSs (Q_T) through SCOP formalism. The continuous and discrete aspects of the Q_{inel} have been determined in relation to ionization (Q_{ion}) and excitation ($\sum Q_{exc}$) cross-sections, by utilizing the CSP-ic approach. Furthermore, the measured Double differential CSs (DDCS) of secondary electrons emitted during the ionization of N_2 by collisions with incoming electrons. Their energy lies between 3 Kev to 5 KeV. The measurements were done in the laboratory of molecular physics at TIFR- Mumbai, as a part of our collaboration with Prof. Lokesh Tribedi and theoretical work on ionization of N_2 is done by us. The measured DDCS were used to estimate total ionization cross sections".

6.1 Introduction

Molecular nitrogen (N_2) is a ubiquitous part of the atmosphere of Earth and it has major impact on atmospheric dynamics. Understanding auroral and ionospheric effect in the uppermost atmosphere requires electron collisions with N_2. The detection of newly generated N_2^+ in titan's torus by using the "Cassini Plasma Science (CAPS)" instrument highlighting the importance of atmospheric sputtering and providing important information on plasma-atmosphere interactions [2].

The release of harmful compounds and greenhouse gases into the atmosphere presents a massive worldwide challenge, with unanswered scientific questions, chief among them the accuracy of the chemical-physical methods used in climate change forecasting models [3]. Even though ions are very important to many processes in the atmosphere, such as nucleation, precipitation, and the formation of aerosols through ion-induced. we are just starting to understand how cosmic rays and ions affect climate, so more in-depth research is desirable [4].

There were huge amounts of results on electron collision with molecular N_2, as highlighted in a review by Brunger and Buckman [6]. Approximately three decades ago, total CSs were measured by Hoffman and coworkers [7], Dalba and coworkers [8], Blaauw and coworkers [9], and Garcia and coworkers [10]. Furthermore, Karwasz and coworkers [11] and Nogueira and coworkers [12] investigated Q_T at elevated energies. Jain and Baluja [13] employed calculations to determine various total CSs for electron collision with N_2 at threshold to high energies. Itikawa and coworkers [14] have provided results for diverse total CSs concerning the ground state of N_2.

Table 6.1 Molecular properties of N_2

Schematic	I.P. (eV)	Polarizability ($Å^3$)
	15.60[a]	11.70[a]

[a] www.chemspider.com

6.2 Experimental Setup

The Double Differential Cross-Sections (DDCS) for the emission of electrons from N_2 have been experimentally obtained utilizing electron projectiles with energy levels under 10 keV. The experiment has been be performed at the existing electron spectroscopy setup at TIFR-Mumbai and graphical representation is shown in figure 6.4. The energy of projectile electrons ~10 keV has obtained from an existing electron gun assembly. The entire experiment is performed under high vacuum condition, about 10^{-7} torr. The projectile electrons are guided up-to the collision region using sets of two einzel lens (E1 & E2), deflectors (D1 & D2) and several apertures of different dimensions. The well collimated electron beam interacts with the target gas in the scattering chamber. For the present work, N_2 is used as the target. The energy of secondary electrons emitted from N_2 are analyzed through a "Hemispherical Electrostatic energy Analyzer" and subsequently detected by a "Channel Electron Multiplier (CEM)". The spectrometer comprises of two "hemispherical electrodes" which are biased at different voltages corresponding to the electron energy to be detected. The secondary electrons with energies from 1eV to few hundreds are detected.
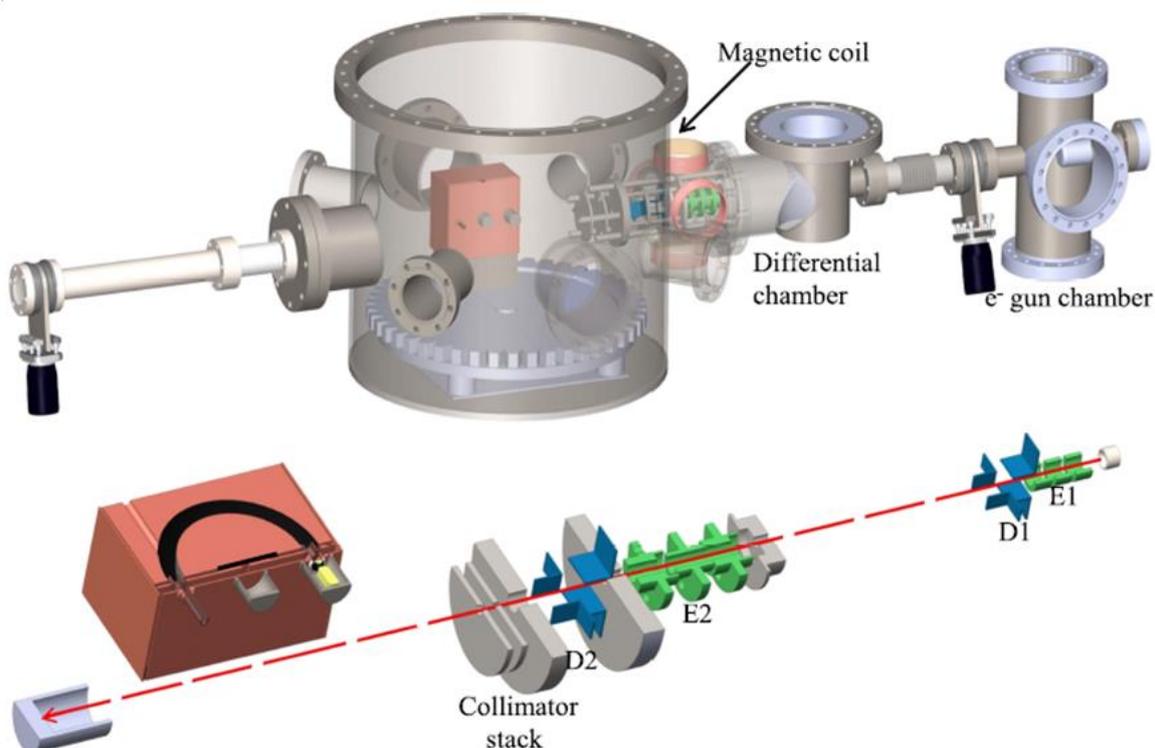

Figure 6.1 The graphical view of experimental setup

The detection of electrons with low energy is extremely difficult and for this purpose it has to be ensured that there is no presence of electric or magnetic field near the interaction region. Extreme cleanliness has to be maintained inside the chamber. The scattering chamber contains two sets of μ-metal sheets that reduce the Earth's magnetic field in its collision region. The spectrometer is placed on a rotatable turntable and hence data can be collected for different electron emission angles.

6.3 Results of molecular nitrogen (N₂)

The N_2 serves as an effective benchmark for evaluating the current methodology. The construction of the molecular charge density involves a single centre expansion at the molecular mass centre [5]. This charge density serves as the fundamental input for the generation of all potentials and, consequently, the computation of scattering CSs. The essential target parameters for the N_2 molecule in these calculations are highlighted in table 6.1.
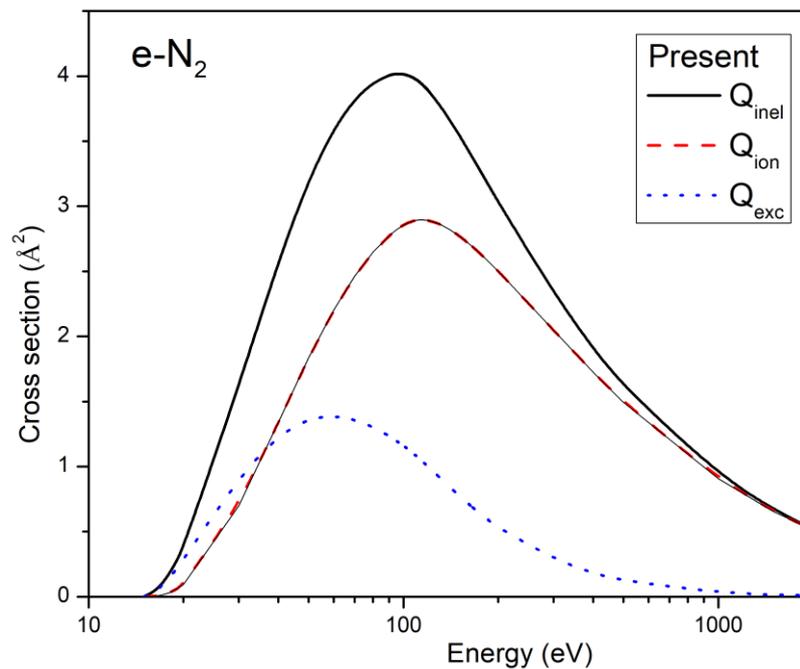
6.3.1 Inelastic contributions

Figure 6.2 Qinel for N₂

Present:- Solid: Qion , Dash: Qion, Dot: Qexc

The CSs calculated in the current study, along with comparative data, are depicted in Figures 6.1, 6.2 and 6.3 for electron scattering with N_2 molecule. Numerous experiments were performed to investigate electron impact CSs for N_2 molecule [7-12, 15-21]. In Figure 6.1, include present Q_{inel} (inelastic), total Q_{ion} (ionization), and $\sum Q_{exc}$ (excitation) CSs, and figure 6.2 represent present Q_{ion} compare with other findings.
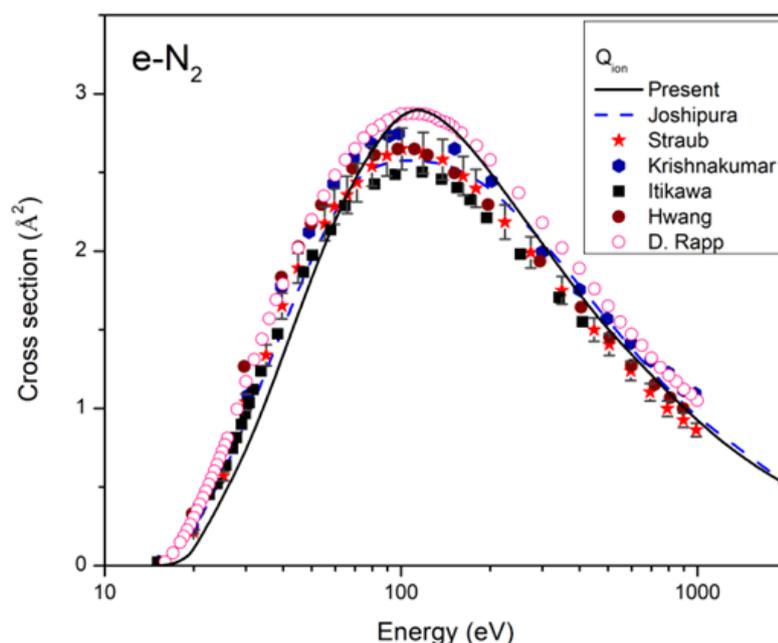
6.3.2 Ionization contributions

Figure 6.3 Q_{ion} for N_2

Q_{ion}: Solid-Present, Dash-Joshipura and coworkers, Star- Straub and coworkers[21], Hexagon- Krishnakumar[19], Square- Itikawa and coworkers[14], filled Circle- Hwang and coworkers[22], pink circle- Rapp and coworkers [20]

In Figure 6.2, our computed results for the total ionization CSs exhibit excellent matching with Krishnakumar and Srivastava [19]. At the peak, our present findings quite overestimate the other results [14,19,22]. Furthermore, our data aligns well with the experimental data of Straub

and coworkers [21], whereas the data from Joshipura and coworkers are comparatively lower than our data, especially below 100 eV. The present Q_{ion} also shows good accord with the Binary Encounter Bethe results reported by Hwang and coworkers [22]. However, the experimental results of Rapp and coworkers [20] shows excellent matching with entire energy range.

Authers [23] obtained the differential CSs in the electron emission energy as well as the emission angles. The measured DDCS, by taking integration over the emission angles or emission energies we have obtained the SDCS and upon further integration we get the TCS of N_2 in collisions with 10 keV electrons.
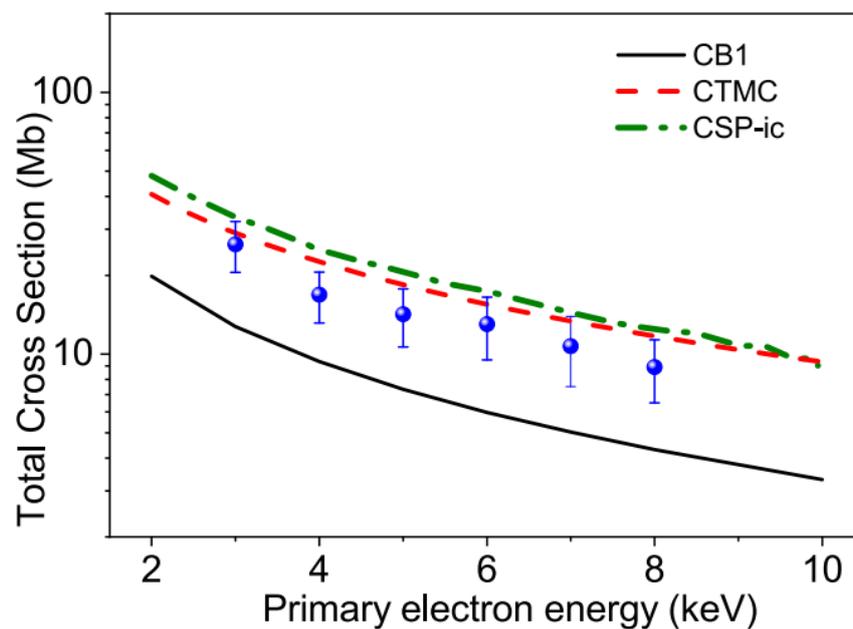

Figure 6.4 Total ionization CSs of N_2

Solid: CB 1 [23], Dash: CTMC [23], Blue circle (present experimental) and Dash dot: CSP-ic (present)

The first-born approximation CB1 model [23] consistently underestimates the experimental result [23], yet it exhibits excellent accord with the observed energy dependence. Moreover, the CTMC [23] (classical trajectory Monte Carlo) model tends to be slightly higher value compared to others, but generally falls under experimental uncertainties, which are approximately 22%–27%. The data of CTMC [23] consistently fall below the present data, and this difference increases at higher energies. In contrast, the CB1 [23] is closer to the present measurements [23] but exhibits a noticeable deviation from the available findings [24].

The total cross-section (TCS) values calculated using the CSP-ic model tend to overestimate the experimental findings across all energy ranges, although the model effectively captures the qualitative energy dependence. This discrepancy may be attributed to factors such as the spherical charge distribution of the N_2 molecule and the approximations inherent in the semi-empirical model [25]. Notably, the TCS values obtained from the two ab initio models, CB1 [23] and CTMC [23], fall slightly below and slightly above the experimental data, respectively. Among these, the CTMC [23] model shows the closest agreement with the present results.

6.3.3 Elastic and Total contributions

The total CSs is the summed of all elastic and inelastic CSs. In figure 6.3, we have shown the complete set of total CSs (Q_T) and elastic CSs (Q_{el}) electron impact energy range threshold to 2000 eV for e-N_2.
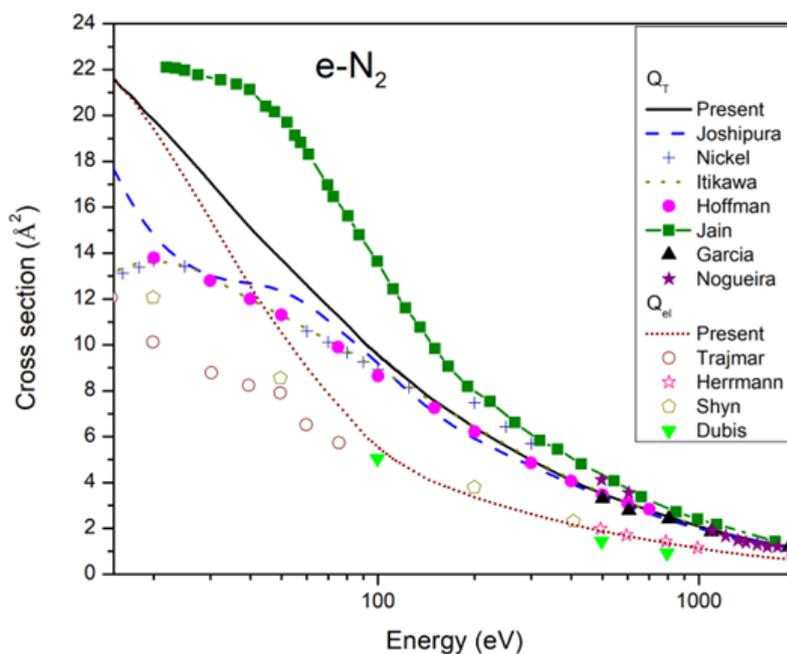

Figure 6.5 Q_{el} and Q_T for N_2

Q_T: Solid-Present, Dash-Joshipura, Plus-Nickel, Dot- Itikawa *et al.* [14], Circle-Hoffman *et al.* [7], Solid green square- Jain and Baluja [13], Triangle- Garcia *et al.* [10], Star- Nogueira *et al.* [12], Q_{el}: Short dot-Present, Wine circle- Trajmar *et al.* [18], Pink Star- Herrman *et al.* [17], Pentagon- Shyn [14], Green Triangle- Dubois [16]

Our calculated total (Q_T) and elastic (Q_{el}) CSs exhibit good agreement with experimental findings from [7-12] above 100 eV. Moreover, the theoretical results from Jain and Baluja [13] overestimates CSs at lower energies. Our calculated Q_{el} values are consistent with experimental data from Shyn and Carignan [15], Dubois and Rudd [16], Hermann and coworkers [17], and Trajmar and coworkers [18] above 100 eV. Furthermore, the Q_{el} values from reference [14] align with our results above 100 eV.

6.4   Chapter summary

This chapter covered the entire set of theoretical CSs data for electron-induced molecular nitrogen across the energy-range starting from the threshold to 5 KeV. Additionally, we utilised measured DDCS data of ionization of molecular N_2 with incoming electrons energies varying from 3 KeV to 5 KeV. Molecular nitrogen (N_2) is the predominant molecule in Earth's environment. Additionally, it plays essential roles in various technological processes. For the computation of Q_{inel}, Q_{el} and Q_T, we have utilised SCOP formalism and the discrete and continuum contributions of the Q_{inel} have been determined in terms of ionization (Q_{ion}) and summed excitation ($\sum Q_{exc}$) CSs, by employing the CSP-ic formalism. Our calculated data are in a reasonably good accord with the available data. The measured total cross-sections (TCS) were compared with ab initio CB 1 and CTMC models, along with semi-empirical CSP-ic method. The energy dependence of CB 1 and CSP-ic is qualitatively similar whereas within the experimental uncertainties, the CTMC model provides an excellent fit to the measured TCS data.

6.5 Bibliography

[1]   H.T. Smith, M. Shappirio, E.C. Sittler, D. Reisenfeld, R.E. Johnson, R.A. Baragiola, F.J. Crary, D.J. McComas, D.T. Young, Geophys. Res. Lett. 32 (2005) L14S03

[2]   H.T. Smith, R.E. Johnson, V.I. Shematovich, Geophys. Res. Lett. 31 (2004) L16804

[3]   T. D. Mark and G. H. Dunn, *Electron Impact Ionization,* Springer (1985)

[4]   K. Bartschat and M. J. Kushner, *Proc. Natl Acad. Sci.* **113** (2016) 7026.

[5]   S. Hara, J. Phys. Soc. Jpn. 22 (1967) 710

[6]   M. J. Brunger and S. J. Buckman, Phys. Rep. **357** (2002) 215, 40-47

[7]   K. R. Hoffman, M. S. Dababneh, Y. F. Hsieh, W. E. Kaupppila, V. Pol, J. H. Smart, T. S. Stein, Phys. Rev. A., **25** (1982) 1393

[8]   G. Dalba, P. Fornaisini, R. Grisenti, G. Ranieri, A. Zecca, J. Phys. B., **13** (1980) 4695

[9]   H. J. Blaauw, R. W. Wegenaar, D. H. Barends, F. J. Deheer, J. Phys. B. **13** (1980) 359

[10]   G. Garcia, A. Perez, J. Campos, Phys. Rev. A. **38**, (1988) 654

[11]   G. Karwasz, R. S. Brusa, A. Gasparoli, A. Zecca, Chem. Phys. Lett., **211** (1993) 529

[12]   J. C. Nogueira, I Iga, C. J. Eduardo, *Revista Brasiliera de Fisica*, **15**, (1985) 224

[13]   A. Jain, K. L Baluja, Phys. Rev. A **45**, (1992) 202

[14]   Y. Itikawa, M. Hayashi, A. Ichimura, K. Onda, K. Sakimoto, K. Takayangi, M. Nakamura, H. Nishimura, T Takayangi, J. Phys. Chem. **15** (1986) 985

[15]   T. W. Shyn, G. R. Carignan, Phys. Rev. A, **22** (1980) 923

[16]   R. D. Dubois, M. E. Rudd, J. Phys. B., **9** (1976) 2657

[17]   D. Herrmann, K. Jost, J. Kessler, M Fink, J. Chem. Phys., **64** (1976) 1

[18]   S. Trajmar, D. F. Register, A. Chutjian, Phys. Rep, **97** (1983) 219

[19]   E. Krishnakumar, S. K. Srivastava, J. Phys. B.**23** (1990) 1893

[20]   D. Rapp, P. Englander Golden, D. Briglia, J. Chem. Phys., **43** (1965) 1465

[21]   H. C. Straub, P. Rrnault, B. G. Lindsay, K. A. Smith, R. F. Stebbings, Phys. Rev. A., **54** (1996) 2146.

[22]   W. Hwang, Y. K. Kim, M. E. Rudd, J. Chem. Phys., **104** (1996) 2956.

[23]   M. R. Chowdhury, D. Chauhan, C. Limbachiya, K. Tokesi, C. Champion, P. F. Weck and L. C. Tribedi, J. Phys. B.*,* **53** (2020) 235201

[24]   B. L. Schram, F. J. De Heer, M. J. Van der Wiel and J. Kistemaker, Physica, **31**(1965) 94

[25]   J. E. Hudson, M. L. Hamilton, C. Vallance and P. W. Harland, Phys. Chem. Chem. Phys. **5** (2003)