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Differential cross sections for electron elastic scattering and vibrational v=1 excitation in nitrogen in the energy range from 5 to 20 eV measured over an angular range 10º-180º

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Abstract

Absolute differential cross sections for elastic electron scattering and vibrational, v=0→1 excitation in nitrogen have been measured in the scattering angle range from 10º to 180º and at incident electron energies between 5 eV and 20 eV. The magnetic angle-changing technique has been applied to access the range of scattering angles between 90º and 180º. Integral and momentum cross sections have also been derived by integration of the measured differential cross sections. A comparison with recent theoretical calculations of elastic and vibrational cross sections is presented.

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1. Introduction

Molecular nitrogen being the most abundant molecule in the Earth's atmosphere (constituting 78% of it) is, apart from molecular hydrogen, probably the most often selected target for electron collision studies and measurements of scattering cross sections. This interest was certainly born by the important role nitrogen plays in atmospheric processes such as aurora and dayglow [1]. For example, the experimentally determined cross sections have been used in numerical modelling of electron cooling process in the ionosphere [2]. This interest is also due to a need for accurate cross sections in models which lead to a deeper understanding of electrical discharges in atmospheric gases and other technological processes [3]. Nitrogen, with its relatively simple electronic configuration, also represents a suitable target for fundamental tests of descriptions of electron-molecule interactions [4]. In these aspects it is vital to form a comprehensive data base for differential and integral cross sections of the electronic elastic and inelastic processes in nitrogen. The magnetic angle-changing technique that has been recently developed [5, 6] allows the complete scattering angle range from close to 0^o up to 180^o to be accessed and this has enabled the cross section data base to be expanded.

The present article reports differential and integral cross sections for elastic electron scattering and vibrational $v = 0 \rightarrow 1$ excitation in nitrogen for energies of 5, 10, 15 and 20 eV measured over the scattering angle range from 10^o to 180^o. The aims of this work are to, (1) revise previous measurements made in our group in the region of backward scattering (110^o-180^o) at 5 eV [8] in view of the recent measurements of Allan [7] and some inconsistencies between both works at high scattering angles, (2) to extend the previous study to higher energies and a much wider range of scattering angles. Also, the purpose for this work was to carry out measurements in the regions of some disagreement between previously published cross sections, for example in the forward (20^o-40^o) elastic scattering at 5 and 10 eV. The present measurements, providing differential cross sections (DCS) in a wide scattering angle range, up to 180^o, also test extrapolation techniques used over a region of high scattering angles in the determination of integral cross sections. The energy region of the present studies, 5-20 eV, spans the regions between the $^2\Pi_g$ and $^2\Sigma_u^+$ negative ion resonances.

In the past, elastic DCSs for low and intermediate energies have been measured typically in the scattering angle range between 20^o and 135^o. Measurements in this angular region were presented by Srivastava *et al* [9] (5-75 eV), who also gave a summary of the earlier experimental work. The results of Srivastava *et al* were later renormalized by Trajmar

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et al [10]. More recent elastic DCSs are those of Sohn *et al* [11] (0.1-1.5 eV), Nickel *et al* [12] (20-100 eV), Brennan *et al* [13] (1.5-5 eV), Shi *et al* [14] (0.55-2.2 eV) and Sun *et al* [15] (0.8-10 eV). Very recently Muse *et al* [16] (1-30 eV) reported measurements carried out with a novel thin aperture gas source. DCSs in a wider angular range were obtained by Dubois and Rudd [17] (20-800 eV), Shyn and Carignan [18] (1-400 eV) and Gote and Ehrhardt [19] (10-200 eV) who measured their cross sections up to 150^o, 156^o and 160^o, respectively. Recently, Allan [7] using magnetic angle-changing technique extended his measurements in nitrogen up to180^o at energies between 0.8 and 5 eV (the region of the $^2\Pi_g$ resonance).

Much less work has been performed in the measurements of DCSs for vibrational excitation. In the scattering angle range up to 135^o measurements have been carried out by Truhlar *et al* [20–22] (5–75 eV), Tanaka *et al* [23] (3–30 eV) and Middleton *et al* [24] (20–50 eV) for excitation of the v=1 level and by Pavlovic *et al* [25] (10–40 eV) and Brennan *et al* [13] (1.5-5 eV) for excitation of the v = 1, 2, 3 levels. Allan [7] studied vibrational excitation (v=1) of nitrogen in the region of the $^2\Pi_g$ resonance (0.8–5 eV) in the scattering angle range up to 180^o.

Several theoretical calculations of elastic scattering and vibrational excitation have been reported in the energy range 5-20 eV. Recent elastic scattering calculations of Feng *et al* [26] concentrated on an accurate treatment of correlation-polarisation interaction in a parameter-free adiabatic approach. These calculations continue the earlier converged vibrational close-coupling calculations of Sun *et al* [15]. In the earlier works; Sawada *et al* [27] applied a formulation based on the independent-particle model and fixed-nuclei approximation, Chandra and Temkin [28] utilised hybrid theory combining vibrational close-coupling with the adiabatic nuclei approximation, Siegel *et al* [29] employed continuum multiple-scattering method, Morrison and Collins [30] solved electron scattering equations using model exchange potentials whereas Onda and Truhlar [31,32] performed rotational close-coupling calculations. Also available are the later calculations of Gillan *et al* [33] who used the *R*-matrix method and those of Huo *et al* [34] who employed the Schwinger multichannel method. Calculations of vibrational excitation were performed by Chandra and Temkin [28], Gillan *et al* [33], Truhlar *et al* [21] and by Onda and Truhlar [31,32]. A comprehensive review of the work in nitrogen was done by Brunger *et al* [35] and recommended cross sections were compiled recently by Itikawa *et al* [36].

Despite the large number of measurements and calculations there are still discrepancies between the various results especially those obtained in the regions of forward and backward scattering. From the experimental point of view, studies in the forward

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scattering are undoubtedly challenging due to the presence of the incident electron beam and the effect of lens focusing on the scattering volume correction. In the theoretical calculations it is necessary to adequately account for long-range polarization and short-range correlation effects which, as pointed out by Gillan *et al* [33] are important in forward and backward scattering, respectively. The present measurements extending the available DCSs up to 180^o provide a test case for theoretical calculations carried out for backward scattering.

2. Experimental

Electron spectrometer used in the present measurements consists of a source of selected electron beam, a scattered electrons analyzer and a magnetic angle changer, which encompasses the source of the target gas beam. The spectrometer has been described in detail elsewhere [37]. Briefly, the electron beam source contains double hemispherical selector (mean radius 37.5 mm) and two systems of triple-electrode cylindrical lenses. The scattered electron analyzer is equipped with a double hemispherical selector identical to that of the incident electron beam source. It can be rotated around the target beam axis over the angular range from -90^o to 90^o with respect to the direction of the incident electron beam. The gas beam is produced by a stainless steel capillary with 0.6 mm inner diameter. The scattering angles from 90^o to 180^o are made accessible with a help of the magnetic angle changer [38]. The angle changer produces a static, localized magnetic field that is perpendicular to the scattering plane. The electrons are deflected by the magnetic field which separates the backward scattered electrons from the incident electron beam and therefore enables detection of electrons scattered at high angles. The scattering angle scale has been determined with an uncertainty of ±2.0^o against position of a minimum in the elastic electron scattering in argon at 117.5^o at 10 eV [39]. The angular resolution of the spectrometer is estimated from the electron optics computations to be less than 5^o.

In the measurements of elastic electron scattering the relative flow technique [40,41] has been applied to obtain absolute values of the DCSs. The ratios of scattered electron intensities in nitrogen to that in helium were recorded for fixed incident electron energies at scattering angles in the range from 10^o to 180^o at 10^o intervals. The corresponding gas flow rates in both gases were determined from the measurements of pressure increase in a section of gas line equipped with a Baratron pressure gauge. The absolute DCSs in nitrogen were obtained using helium theoretical cross section of Nesbet [42] in the energy range 5-15 eV and that of Saha [43] at 20 eV. To fulfill the condition of equal mean-free-path lengths for

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both gases the ratio of driving pressures of nitrogen to that of helium in the gas line was maintained at 0.34. The driving pressure of nitrogen behind the capillary was kept below 0.3 mbar. To achieve stability of the spectrometer operation a mixture of both gases was always present in the vacuum chamber. While one gas was flowing to the interaction region through capillary the other was let directly to the vacuum chamber through a side valve. Elastic scattering intensities (and energy loss spectra, see below) were corrected for the background contributions which were determined by admitting the target gas directly to the vacuum chamber.

In the studies of vibrational excitation, energy loss spectra were recorded for fixed incident electron energies at each scattering angle. From the energy loss spectra, using a peak fitting procedure, ratios of intensities of the v=1 peak to that of the elastic peak have been determined. In the case of differing widths of elastic and inelastic peaks areas of the peaks were taken instead of intensities. Absolute DCSs for excitation of the v=1 level have been derived from the above ratios taking present elastic cross sections. The energy loss spectra for scattering angles above 90^o were recorded using magnetic angle changer at a fixed position of the electron analyzer. This introduces an additional uncertainty in the angular scale of the inelastic scattering of 1.5^o for 5eV and less than 1^o at higher energies resulting in a total uncertainty for vibrational excitation of ±3.5^o at 5 eV and less than ±3.0^o at other energies.

Electron energy was calibrated against the position of the $^2P_{3/2}$ negative ion resonance in Ar at 11.103 eV [44] with an uncertainty of ±30 meV. The energy resolution in the present measurements was 40–60 meV.

3. Results and discussion

3.1. Cross sections for elastic scattering

The DCSs obtained for elastic electron scattering at energies of 5, 10, 15 and 20 eV are presented and compared with other available experimental and theoretical results in figures 1(a)-(d). Their numerical values are listed in table 1. The statistical uncertainties in the present DCSs are estimated to be 13% for 5 eV, 10% for 10 eV and 15 eV and 15% for 20 eV. These values include uncertainties in the cross sections of Nesbet [42] and Saha [43] for helium which from comparison with other calculations were taken to be 2% and 2.5%, respectively.

The DCS obtained at 5 eV (figure 1a) are in agreement with most of the earlier experimental data in the angular range from 60^o to 120^o but exceptional agreement is found

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with the results of Allan [7] and earlier results of Srivastava *et al* [9] (later renormalized by Trajmar *et al* [10]). Agreement with the results of Allan extends beyond 120^o however above 160^o our results are slightly below Allan's although within the experimental uncertainty. The results of Shyn and Carignan [18] start to deviate from the present and Allan's results above 120^o being 25% higher at 156^o. Also the extrapolated cross section of Muse *et al* [16] tends to overestimate the cross section in the range 130^o-180^o. In the region below 50^o measurements from different laboratories show differences. At 30^o the present cross section places between the lowest cross section of Allan and the highest of Brennan *et al* [13] which differ from the present result by 15% and 8%, respectively. The results of theoretical calculations shown in figure 1a except those of Huo *et al* [34] in general reproduce well the angular behavior of the experimental cross sections in the range 60^o-180^o. Here, the best agreement with experiment is shown by the calculations of Feng *et al* [26] and Sun *et al* [15]. In both works polarization-correlation effects were treated in the most detailed way. In the region below 60^o the theoretical results except those of Gillan *et al* [33] display the angular dependences in accord with experiment but most of them overestimate the cross section. Deviation of the results of Gillan *et al* from the experimentally determined angular shape of the cross section has been attributed to inadequate representation of long-range dipole polarization.

The DCS measured at 10 eV (figure 1b) agree well with the experimental data of Sun *et al* [15] and those of Muse *et al* [16] in the angular range they overlap. The extrapolated cross section of Muse *et al* at 180^o is higher than the present result by about 30%. The results of Shyn and Carignan [18] and Gote and Ehrhardt [19] at 150^o-160^o exceed our results by about 12% and 20%, respectively. The renormalized data of Srivastava *et al* [9] are 18% higher in the whole angular range. In the range of forward scattering the cross sections of Gote and Ehrhardt and Shyn and Carignan are higher than the present measurements, at 20^o by about 15%. The theoretical calculations show in general poorer agreement (figure 1b) with experimental data than for 5 eV and display inconsistencies in the region of backward scattering. For the forward scattering, except the results of Sun *et al* [15], they are above the experimental cross sections. These discrepancies indicate need for adequate representation of long-range polarization and short-range correlation interactions.

The 15 eV DCS (figure 1c) obtained in the range 15^o-180^o coincide well with the previous measurements which are generally in good accord with each other although below 20^o they slightly diverge remaining within their uncertainty limits. At 15 eV (and 20 eV) the present results confirm extrapolated cross sections of Muse *et al* determined above 130^o. The

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available theoretical data of Sawada *et al* [27] and Siegel *et al* [29] tend to be higher than the experimental cross sections and above 70^o diverge in their angular behaviour from the measured cross sections (see inset of figure 1c).

The 20 eV DCS (figure 1d) measured from 10^o to 180^o are in general accord with the compared experimental results except those of DuBois and Rudd [17]. Their cross section, below 50^o, is lower than the other results by about 30%. Also, the results of Gote and Ehrhardt [19] and Shyn and Carignan [18] at the high scattering angles (150^o-160^o) deviate from the present results and are 13% and 15% lower, respectively. Turning to comparison with theoretical calculations, results of Onda and Truhlar [32] show good agreement with experiment in the whole angular range, while those of Siegel *et al* [29] tend to be above measured cross sections and moreover show different angular dependence as further shown in the inset of figure 1d.

3.2. Integral and momentum transfer cross sections for elastic scattering

The integral and momentum transfer cross sections were determined by integration of the DCSs in the angular range 0^o–180^o. The measured cross sections were extrapolated down to 0^o and at 10, 15 and 20 eV linear extrapolation was applied. At 5 eV the theoretical data of Sun *et al* [15] were used, after normalizing to the present results in the range 25^o–60^o. Deduced cross sections are listed in table 2 and are shown in figures 2(a) and (b). The associated uncertainties in both integrated cross sections are equal to those determined for the elastic cross sections since the integration and extrapolation techniques introduced uncertainties of less than 0.5 %.

The integral cross section is compared (figure 2a) with previous results derived from experimental and theoretical DCSs and with measured total cross sections [15,45-47]. There is general consistency between various determinations although the cross sections of Srivastava *et al* [9] and Shyn and Carignan [18] at 7 and that of Srivastava *et al* at 10 eV are higher. At 5 eV the elastic integral cross section coincides with the total cross section since contribution of excitation (vibrational) to the total cross section is small, less than 0.1×10^{-20} m^2 [36]. This contribution increases with energy and is about 15 % at 15 eV. The momentum transfer cross section (figure 2b) is compared with available data which include cross section of Ohmori *et al* [48] derived from swarm experiment (for other references of swarm determinations see Robertson *et al* [49]). Again at 7 and 10 eV cross sections of Srivastava *et al* [9] and Shyn and Carignan [18] are above the present results. Very good agreement is

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found between the present cross section and those of Sun *et al* [15] and the swarm determination of the momentum cross section by Ohmori *et al*.

3.3 Differential cross sections for vibrational excitation

The DCSs for vibrational (v=0→1) excitation measured at 5, 10, 15 and 20 eV are presented in figures 3(a)–(d) and compared with available experimental and theoretical data. Their numerical values are given in table 3. The statistical uncertainties in the DCSs obtained at 5 eV are estimated to be 22% in the angular range 25^o–40^o and 19% in the remaining angular range. The uncertainties at 10, 15 and 20 eV are 17%, 14% and 18% in the whole angular range, respectively. These total uncertainties result from statistical uncertainties in the measured inelastic to elastic intensity ratios and those in the DCSs for elastic scattering.

At the incident energy of 5 eV, the present DCS (figure 3a) displays two minima, at 55^o and 125^o, and is asymmetric with respect to a maximum at about 85^o. It is in the best agreement with results of Brennan *et al* [13] measured up to 130^o. The DCS of Allan [7] show more symmetric angular shape with respect to 85^o than the present results. However, the differences between both results at 25^o and 180^o are within uncertainty limits. These results indicate considerable contribution from the $^2\Pi_g$ resonance at 5 eV. The R-matrix calculations of Gillan *et al* [33] which apart from $^2\Pi_g$ symmetry also accounted for five other scattering symmetries generated lower cross sections in the forward than in the backward scattering.

At 10 eV, the present DCS are compared in figure 3(b) with the measurements of Tanaka *et al* [23] and Truhlar *et al* [21]. The present measurements show a maximum in the angular dependence at about 50^o which is not clear in the previous results of Tanaka *et al*. The results of Gillan *et al* reflect well the experimental angular shape of the DCS although are lower in their absolute value.

Figure 3(c) shows DCS at 15 eV measured in the scattering angle range 15^o-180^o. The results of Tanaka *et al* are much higher than the present cross section. A single value of DCS obtained by Tronc and Malegat [50] at 90^o coincides very well with the present results.

Figure 3(d) presents vibrational DCS at 20 eV. This energy is close to the center (23 eV) of the $^2\Sigma_u^+$ shape negative-ion resonance which is the d-wave resonance with a contribution of the p-wave. The present results are in reasonable agreement with the measurements of Tanaka *et al* [23] and Middleton *et al* [24] although below 90^o are approximately 20% lower. The single point of Tronc and Malegat [50] at 90^o is again in good accord with present measurements. Good overlap with the earlier results of Truhlar *et al* [20]

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(as normalised by Onda and Truhlar [32]) is also noticed. The present cross section shows a minimum near 85° reproduced well in the calculations of Gillan *et al* [33] and Onda and Truhlar [32], which however, greatly underestimate the DCS.

3.4 Integral cross section for vibrational excitation

Integral cross section for the v=0→1 excitation in nitrogen is presented in figure 4. The numerical values are included in table 3. The uncertainties in the integral cross section reach 20% at 5 eV, 17% at 10 eV, 14% at 15 eV and 18% at 20 eV.

The present results are in good agreement with previous determinations of Allan [7], Tanaka *et al* [23] and Truhlar *et al* [21] at 5 eV and that of Truhlar *et al* [21] at 10 eV. Above 15 eV the integral cross section begins to rise due to the contribution of the $^2\Sigma_u^+$ resonance. At 15 eV the result of Tanaka *et al* is much larger than the present results most likely as a result of their higher DCS. At 20 eV the result of Truhlar *et al* [20] agrees well with the present result, whereas the cross section of Tanaka *et al* is again larger than the present value. The most likely reason for this difference is the shape of the extrapolation function used by Tanaka *et al* above 130°. The calculations of Feng *et al* [26] agree with the present cross section at 10 eV and with the result of Tanaka *et al* at 7.5 eV while at lower energies their cross section rises rapidly with decreasing energy.

4. Conclusions

Absolute differential cross sections for electron elastic scattering and excitation of the vibrational v = 1 level of molecular nitrogen measured over a wide scattering angle range, 10°–180°, at incident electron energies of 5, 10, 15 and 20 eV are reported. The magnetic angle-changing technique has been employed to access the scattering angle range 90°-180°. Excellent agreement has been found with elastic DCS of Allan [7] at 5 eV in most region of the backward scattering. A small difference is still observed close to 180°. Agreement between available data in the elastic scattering is generally better in the 60°-120° angular range and the angular range of agreement becomes wider for higher energies. For excitation of the vibrational v=1 level of nitrogen agreement between available data is much less satisfactory. The present results indicate that some extrapolation techniques used to extend the DCSs towards 180° do not give results in accordance with measurements. The present DCSs allowed integral cross sections for elastic scattering and vibrational excitation to be

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determined avoiding extrapolation over wide angular region of scattering in the backward directions.

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Table 1. Differential cross sections in units of 10^{-20} m^2/sr for electron elastic scattering in nitrogen at incident energies of 5, 10, 15 and 20 eV.

Scattering angle (degree)	Energy (eV)			
	5	10	15	20
10	–	–	–	6.824
15	–	–	3.874	–
20	–	2.147	3.171	4.137
25	1.734	–	–	–
30	1.761	1.918	2.202	2.472
40	1.685	1.688	1.661	1.642
50	1.464	1.386	1.205	1.036
60	1.178	1.058	0.802	0.633
70	0.947	0.726	0.543	0.347
80	0.734	0.510	0.311	0.204
90	0.618	0.357	0.263	0.205
100	0.587	0.380	0.285	0.261
110	0.574	0.439	0.388	0.356
120	0.596	0.542	0.533	0.473
130	0.638	0.650	0.672	0.577
140	0.689	0.788	0.815	0.715
150	0.732	0.910	0.935	0.914
160	0.811	1.008	1.098	1.064

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170	0.815	1.031	1.110	1.147
180	0.861	1.016	1.131	1.223

Table 2. Elastic integral (σ_i) and momentum transfer (σ_m) cross sections in units of 10^{-20} m^2 in nitrogen at incident energies of 5, 10, 15 and 20 eV.

	Energy (eV)			
	5	10	15	20
σ_i	11.62	10.96	11.14	11.29
σ_m	9.268	8.568	8.086	7.284

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Table 3. Differential cross sections in units of 10^{-23} m^2/sr and integral cross section (σ_i) in units of 10^{-23} m^2 for vibrational v=0→1 excitation in nitrogen at incident energies of 5, 10, 15 and 20 eV.

Scattering angle (degree)	Energy (eV)			
	5	10	15	20
10	–	–	–	25.45
15	–	–	3.801	–
20	–	3.440	2.392	19.86
25	13.34	–	2.232	–
30	10.77	2.212	1.714	14.30
40	6.815	1.784	1.504	11.57
50	4.805	1.859	1.590	9.555
55	4.630	–	–	–
60	5.015	1.792	1.568	8.643
70	5.755	1.509	1.638	6.660
80	5.900	1.137	1.605	4.711
90	4.890	0.682	1.493	5.301
100	4.320	0.589	1.454	6.770
110	4.030	0.398	1.428	8.676
120	3.065	0.556	1.162	9.536
130	3.300	0.647	1.094	10.36
140	5.070	0.880	1.154	10.81
150	7.660	1.209	1.606	13.35
160	10.25	1.478	2.354	16.55
170	12.13	1.666	3.155	17.92

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180	13.56	1.816	3.250	19.14
σ_i	76.07	15.99	20.22	122.5

Figure captions

Figure 1
Differential cross sections for elastic electron scattering in molecular nitrogen at electron incident energies of: (a) 5 eV, (b) 10 eV, (c) 15 eV and (d) 20 eV. The insets in figures (c) and (d) show differential cross sections in the angular regions 60^o-160^o and 50^o-160^o, respectively.

Figure 2
(a)Integral cross sections for elastic electron scattering in molecular nitrogen. Comparison is also made with results of total cross section (TCS) measurements. (b) Momentum transfer cross sections for elastic electron scattering in molecular nitrogen.

Figure 3
Differential cross sections for excitation of the vibrational v=1 level of molecular nitrogen at incident electron energies of: (a) 5 eV, (b) 10 eV, (c) 15 eV and (d) 20 eV. Note that in figure (d) the results of the theoretical calculations have been multiplied by a factor of 4.

Figure 4
Integral cross sections for excitation of the vibrational v=1 level of molecular nitrogen.

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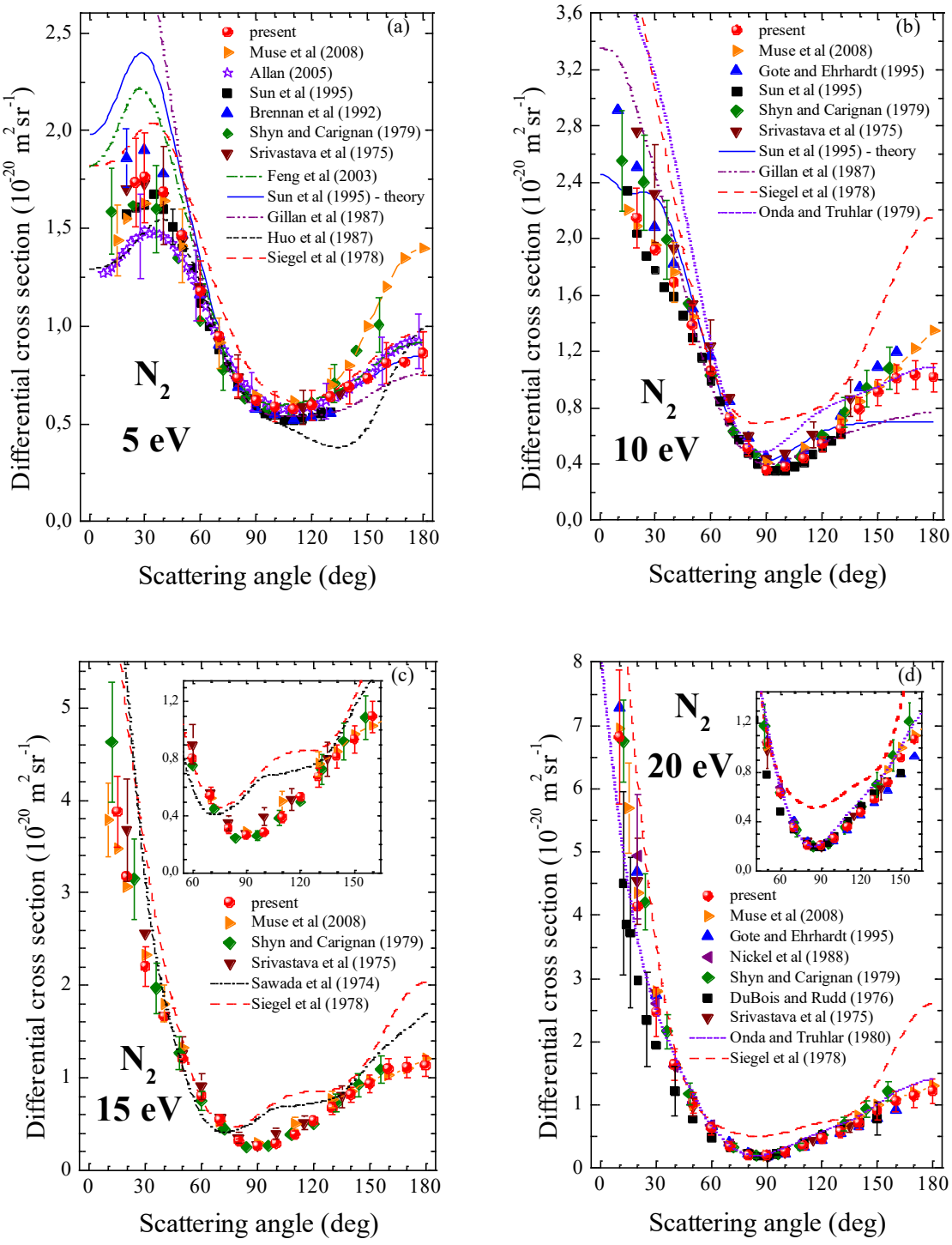

Fig. 1

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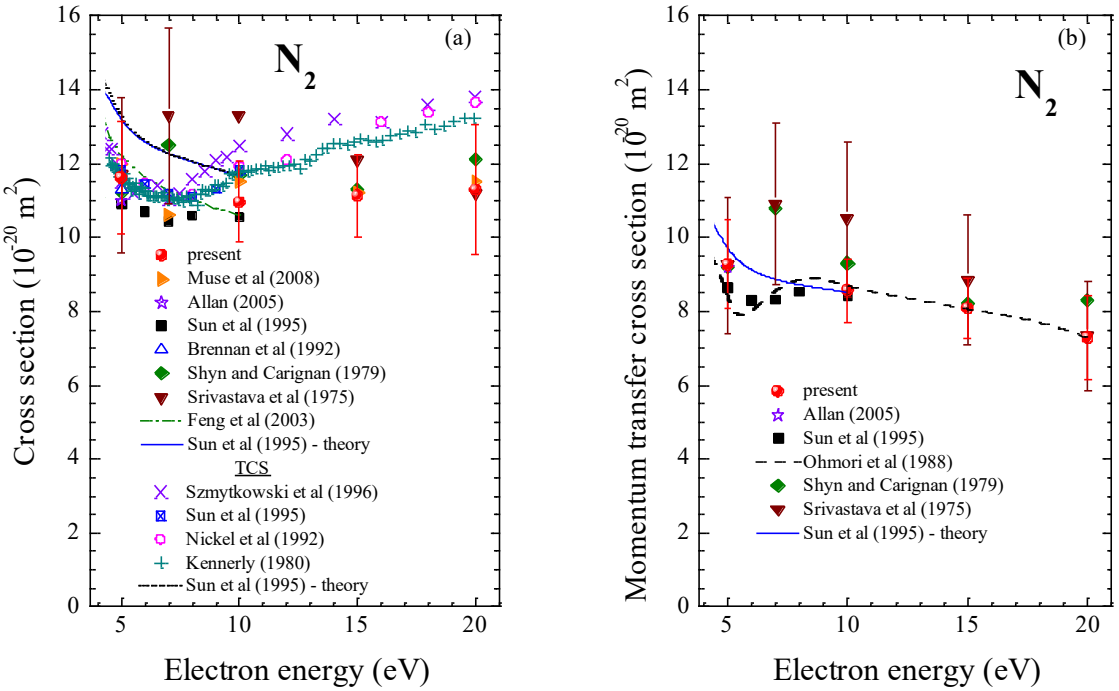

Fig. 2

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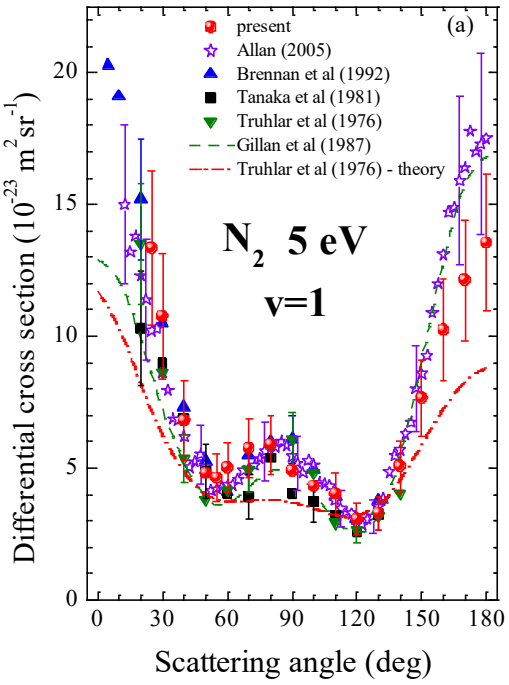

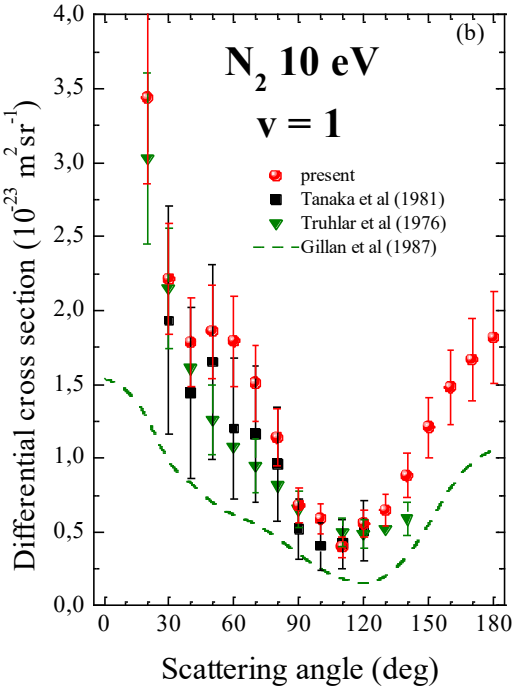

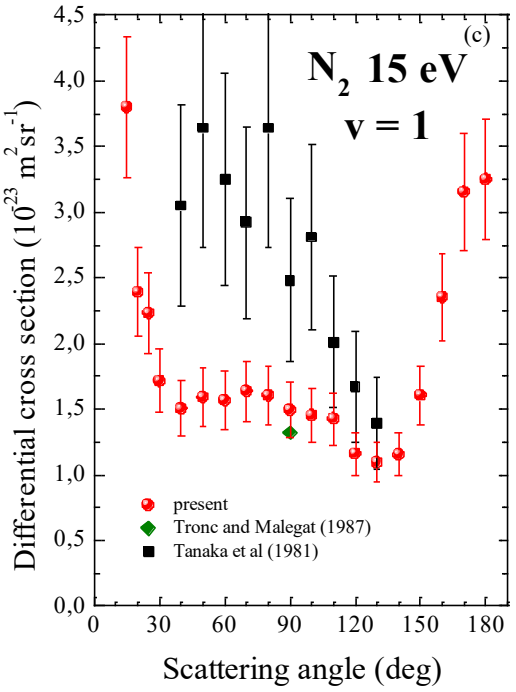

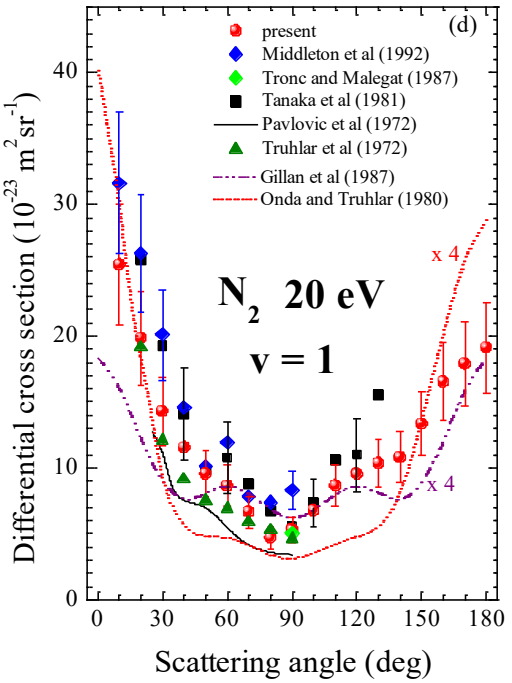

Fig.3

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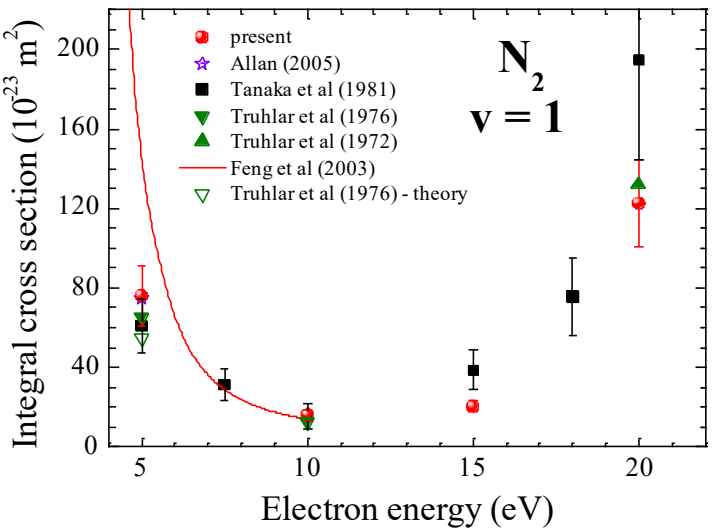

Fig. 4

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