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Absolute elastic cross sections for electron scattering from N_2O

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Abstract. Absolute elastic differential cross sections are presented for electron scattering from nitrous oxide (N_2O) for incident energies between 5 eV and 80 eV. Integral and momentum transfer cross sections have also been determined by extrapolation of the differential cross section data to 0° and 180°. Particular attention has been paid to the measurement of differential cross sections below 10 eV as in this region only one earlier measurement at 5 eV has been reported. Previous works investigating vibrational excitation have shown evidence for a negative ion resonance near 8 eV, however no manifestation of this resonance has been found in the elastic scattering channel in this work. A comparison is also made between the differential cross sections of N_2O and CO_2 as previously it has been found that the cross sections of isoelectronic molecules (i.e. N_2 and CO) exhibit very similar shapes and values.

1. Introduction

In recent years, nitrous oxide (N_2O) has attracted increasing scientific interest due to its role in a number of important processes. For example, N_2O lasers have been used as a secondary frequency standard in areas of spectroscopy within the 10 μm region where the frequency range of the CO_2 laser is inadequate (Fox and Reid 1985). Nitrous oxide has also been found to be increasingly important in the chemistry of the upper atmosphere where it may play an important role in the destruction of the ozone layer (Hahn and Junge 1977, Wang and Sze 1980, Wayne 1991). It has also been noted that electron scattering cross sections for molecules with a similar structure show similar characteristics. For example Jain (1982) has reported that for incident electron energies between 40 eV and 800 eV the elastic differential, integral and momentum transfer cross sections for electron scattering from N_2 and CO are similar. As N_2O and CO_2 are isoelectronic and linear in their ground states, they may also be expected to follow this trend.

Within the last ten years, several cross sections for electron scattering from N_2O have been reported. Kwan *et al* (1984) and Szmytkowski *et al* (1984, 1989) have reported total cross sections and Kubo *et al* (1981) and Marinković *et al* (1986) have published differential cross sections. In addition, Azria *et al* (1975), Tronc *et al* (1981) and Andrić and Hall (1984) have reported vibrational differential cross sections for incident energies in the 2 eV and 8 eV regions. Recently, Barnett *et al* (1991) have reported the observation of metastable N_2 and O from electron impact dissociative excitation of N_2O. Several theoretical calculations and assignments of the electronic states of N_2O have been performed (Chutjian and Segal 1972, Fridh *et al* 1978, Nakatsuji 1983) but only one attempt has been made to calculate electron scattering

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cross sections (Dubé and Herzenberg 1975) in which the adiabatic nuclei approximation to calculate vibrational excitation cross sections at low incident energies ($E < 4$ eV) was used.

In this work we present differential, integral and momentum transfer cross sections for electrons scattered from N_2O for incident energies between 5 and 80 eV using an experimental technique different to that used in the two previous works which have reported cross sections in this energy range (Kubo *et al* 1981, Marinković *et al* 1986). Particular attention has been given to the measurement of cross sections below 10 eV as there is reported evidence for a shape resonance in the 8 eV region (Tronc *et al* 1981, Andric and Hall 1984).

2. Experimental details

A brief outline of the apparatus and procedures used to obtain the cross sections presented in this work is given below. A more detailed description can be found in Johnstone and Newell (1991). The apparatus used was based around a conventional electron spectrometer. Electrons emitted from a thoriated tungsten filament were energy selected using a 180° hemispherical selector. The resultant beam was then focused to the centre of an interaction region where it was crossed at 90° with a gas beam. Electrons scattered from the gas beam were detected by an analyser and a reference detector. In the analyser, the scattered electrons were focused into a second 180° hemispherical selector and their energy analysed before subsequent detection by a single channel electron multiplier (CEM). The analyser was able to detect electrons scattered over an angular range between 10° and 120° and with a solid angle of 6×10^{-3} sr. The intensity of the incident beam was typically 15 nA and the overall resolution of the spectrometer (i.e. incident electron beam and analyser combined) was 60–65 meV. The angular divergence of the incident beam was typically 2°. The reference detector was positioned at a fixed scattering angle of 90° and was composed of a three-element electrostatic lens followed by a CEM. Its solid angle, subtended at the centre of the interaction region, was the same as that of the analyser. By applying a negative voltage to the centre of the lens it was possible to select only those electrons elastically scattered from the gas beam. The energy resolution of the analyser and the reference detector was such that it was not possible to distinguish between elastically scattered electrons and those which had excited rotations in the N_2O molecule.

The gas beam was produced using a capillary tube 25 mm long and with an internal diameter of 0.6 mm. The contact potential was determined by measuring the position of the 19.37 eV He resonance and comparing it to the value quoted by Brunt *et al* (1977). To check for any shift in the contact potential which arose from changing between helium and nitrous oxide, the transmission profile of the reference detector was measured before and after changing the gas. This was done by ramping the voltage applied to the centre element of the three element lens in the detector and measuring the intensity of the signal from the channeltron. Any shift in the position of the profile was attributed to a change in the contact potential caused by admitting N_2O to the system. Using this procedure the incident energy of the electron beam could be set to an accuracy of ±75 meV.

The absolute elastic differential cross sections were measured in two steps. First, the relative differential cross sections (normalized to 90°) were measured. The reference detector was used to account for any change in the electron or gas beam intensities

while making the angular measurements. In addition, by using a procedure known as the subtraction technique (Newell *et al* 1981, Johnstone and Newell 1991) any contributions from electrons scattered from sources other than the gas beam were also accounted for. The relative differential cross sections were then normalized to an absolute scale at 90° measured using the relative flow technique (Kanik *et al* 1989) and using the helium cross sections of Nesbet (1979) and Register *et al* (1980b) as reference standards. For these measurements the reference detector alone was used to measure the scattered electron count rate.

The errors in the absolute differential cross sections arose from two sources: the measurement of the relative differential cross section (8%) and the determination of the absolute cross section at 90° (9.5%). Thus the total error in any differential cross section value did not exceed 13%.

3. Results

3.1. differential cross sections

The absolute elastic differential cross sections measured in the present work are tabulated in tables 1 and 2 and a representative selection are shown in figures 1 and 2. The data of Kubo *et al* (1981) and Marinković *et al* (1986) are also shown for

Table 1. Absolute elastic differential cross sections for electron scattering from N_2O for incident energies between 5 eV and 12 eV ($\times 10^{-16}$ cm^2 sr^{-1}).

	Incident energy (eV)					
θ	5	7.5	8.0	8.5	10	12
10	0.80	1.89	2.71	2.73	4.43	8.19
15	0.60	1.4	1.91	2.12	3.77	5.44
20	0.8	0.99	1.37	1.52	2.95	4.58
25	0.82	0.82	1.18	1.38	2.45	
30	0.91	0.82	1.10	1.30	2.22	3.08
35	1.02	0.83	1.06	1.19	1.93	
40	1.06	0.86	1.07	1.16	1.59	2.00
45	1.16	0.86	1.05	1.09	1.45	
50	1.21	0.84	1.04	1.05	1.31	1.50
55	1.17	0.81	0.99	1.04	1.19	
60	1.16	0.76	0.97	0.99	1.09	1.20
65	1.12	0.66	0.92	0.93	0.95	
70	0.98	0.60	0.88	0.88	0.84	0.98
75	0.67	0.66	0.81	0.81	0.80	
80	0.95	0.70	0.76	0.71	0.72	0.73
85	0.87	0.65	0.68	0.65	0.64	0.63
90	0.80	0.61	0.60	0.53	0.56	0.55
95	0.72	0.52	0.51	0.51	0.47	
100	0.60	0.45	0.48	0.44	0.44	0.41
105	0.54	0.38	0.42	0.44	0.41	0.40
110	0.46	0.36	0.42	0.46	0.44	0.41
115	0.40	0.36	0.41	0.44	0.50	0.47
120	0.36	0.38	0.46	0.59	0.61	0.60
Error	12%	12%	13%	12%	12%	12%

Table 2. Absolute elastic differential cross sections for electron scattering from N_2O for incident energies between 15 eV and 80 eV ($\times 10^{-16}$ cm^2 sr^{-1}).

	Incident energy (eV)				
θ	15	20	30	50	80
10	8.26	33.79	29.97	23.17	18.29
15	6.43	8.89	12.88	13.27	8.73
20	5.40	4.50	5.54	4.88	1.78
25	4.12				
30	3.28	2.83	2.50	1.96	0.65
35	2.68				
40	2.10	1.65	1.21	0.85	0.28
45	1.72				
50	1.45	1.13	0.73	0.46	0.14
55	1.27				
60	1.12	0.82	0.44	0.24	0.084
65	0.96				
70	0.85	0.57	0.24	0.15	0.070
75	0.74	0.47	0.17	0.14	0.068
80	0.64	0.37	0.16	0.13	0.063
85	0.54	0.28	0.17	0.12	0.062
90	0.43	0.26	0.18	0.10	0.054
95	0.36	0.26	0.21	0.11	0.050
100	0.34	0.24	0.24	0.10	0.051
105	0.34	0.31	0.27	0.13	0.057
110	0.41	0.37	0.30	0.15	0.070
115	0.50				0.076
120	0.64	0.46	0.34	0.27	0.10
Error	12%	12%	12%	12%	13%

comparison. Both these previous works obtained absolute cross sections in a different manner to that employed in the present work. Kubo *et al* (1981) used the relative flow technique to determine the cross section at each angle, while Marinković *et al* (1986) measured relative differential cross sections, which they then extrapolated to 0° and 180° and normalized to total cross sections. These authors quote errors of 30% and 20% respectively which should be compared with the experimental error in the present work of 13%. Figures 1(*a*) to 1(*c*) show that as the energy increases from 5 eV to 10 eV the shape of the differential cross section undergoes a significant change. At 5 eV, the cross section has a broad maximum around 60° and, as the energy increases, the maximum disappears and the cross section becomes forward peaked, with the minimum moving to 100°. For incident energies greater than 10 eV, this trend continues with the small angle cross sections increasing while the minimum deepens and continues to move to smaller scattering angles. Between 10 eV and 20 eV, there appears to be a slight shoulder in the 30° to 90° region and this was also reported by Marinković *et al* (1986) at 12 eV where it was particularly pronounced.

The agreement between the present results and those reported by Kubo *et al* (1981) and Marinković *et al* (1986) is reasonable. The largest discrepancies are found at the higher scattering angles, where the cross sections in the present work are generally larger. It should also be noted that the minimum seen in the data of Marinković *et al* (1986) is generally deeper than that seen in the present work or that of Kubo *et al*

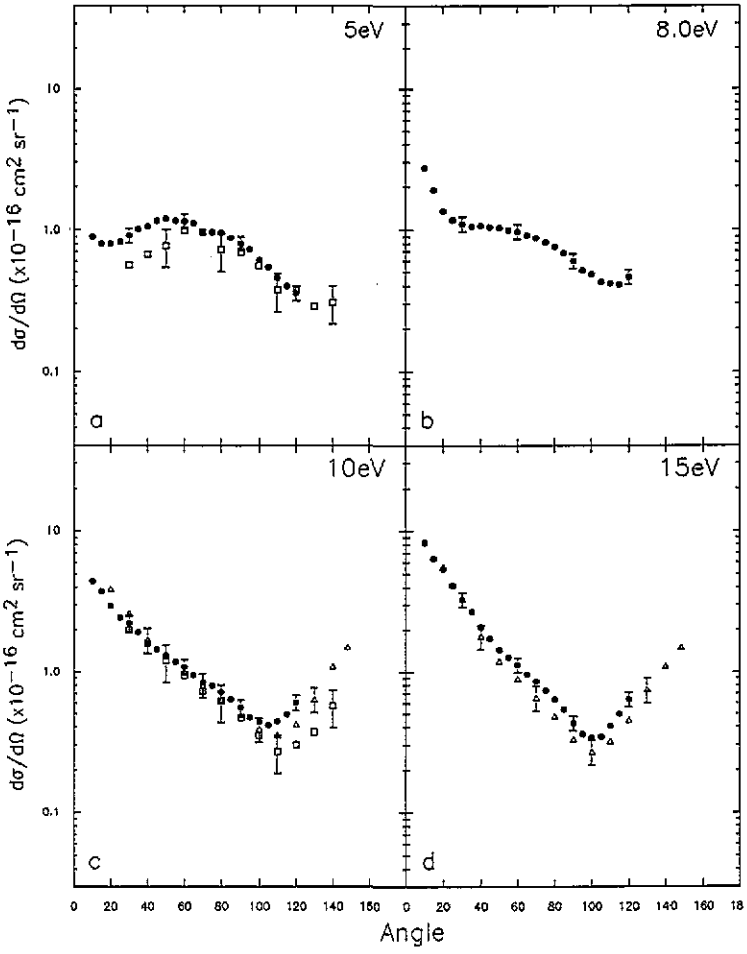

Figure 1. Absolute elastic differential cross sections for electron scattering from N_2O ($\times 10^{-16}$ cm^2 sr^{-1}); (*a*) 5 eV, (*b*) 8 eV, (*c*) 10 eV, (*d*) 15 eV. ●, present work; □, Kubo *et al* (1981); △, Marinković *et al* (1986).

(1981). As the data of Marinković *et al* (1986) are also slightly higher at the smaller scattering angles, this suggests that the angular resolution of their apparatus may have been superior to that used in the present work or by Kubo *et al* (1981). This is borne out by a comparison of the solid angles of the spectrometer used in the present work, 6×10^{-3} sr, and that used by Marinković *et al* (1986) who quote a value of 1×10^{-3} sr.

Two resonances have been detected in low energy electron scattering from N_2O at 2 eV and 8 eV. At 2 eV a shape resonance has been observed and its effects have been seen in a variety of scattering processes. For example, Kwan *et al* (1984) and Szmytkowski *et al* (1984) have observed an enhancement in the total cross section, while Azria *et al* (1975), Tronc *et al* (1981) and Andrić and Hall (1984) have all measured increased vibrational excitation. Tronc *et al* (1977) have also reported a rise in O^- production formed by dissociative attachment at this energy. The second higher lying resonance at 8 eV has only been seen by Tronc *et al* (1981) and Andrić and Hall (1984) in their work on vibrational excitation. From their measurements of the angular behaviour of the vibrational excitation cross sections, Andrić and Hall (1984) postulated

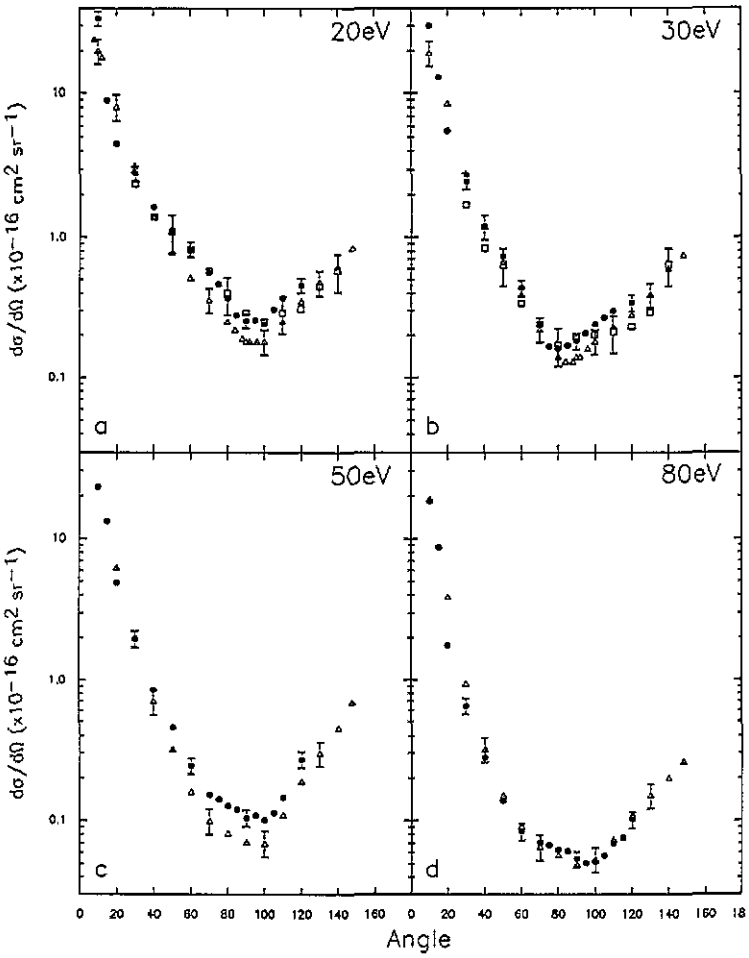

Figure 2. Absolute elastic differential cross sections for electron scattering from N_2O ($\times 10^{-16}$ cm^2 sr^{-1}); (*a*) 20 eV, (*b*) 30 eV, (*c*) 50 eV, (*d*) 80 eV. ●, present work; □, Kubo *et al* (1981); △, Marinković *et al* (1986).

the formation of a resonance at 8 eV, where the $l = 1$ partial wave was dominant. In a previous study of electron scattering from sulphur hexafluoride (Johnstone and Newell 1991) such resonances were seen to give significant enhancements to the elastic scattering cross sections. However, no such evidence was seen in the present data collected at 7.5, 8.0 and 8.5 eV and therefore the 8 eV resonance appears to be weak with the elastic cross sections dominated by non-resonant scattering.

It is clear from figures 1(*a*) and 1(*b*) that the elastic cross section does not display the characteristic p-wave dependence, but is rather more characteristic of isotropic s-wave scattering. Consequently, the s wave would appear to be the dominant partial wave in the scattering at 5 and 8 eV. The p-wave characteristic in the differential cross section develops quickly at higher energies of 10 eV and greater, see figures 1(*c*), 1(*d*) and 2. However, since there is no significant change in the cross section in the 8 eV region, where the resonance was postulated, we can only conclude that any resonance at this energy is very weak, independent of the partial-wave channel in which it appears.

3.2. Integral and momentum transfer cross sections

In addition to the differential cross sections, integral (σ_t) and momentum (σ_m) transfer cross sections have also been derived in the present work. The integral cross section, which represents the probability of an electron with energy E_0 being elastically by an atom or molecule, is obtained by integrating the differential cross section $d\sigma(E_0, \theta)/d\Omega$ over all angles and is defined by

$$\sigma_t = 2\pi \int_0^{\pi} d\sigma(E_0, \theta)/d\Omega \sin\theta \, d\theta. \tag{1}$$

The momentum transfer cross section is defined similarly by

$$\sigma_m = 2\pi \int_0^{\pi} d\sigma(E_0, \theta)/d\Omega(1-\cos\theta) \sin\theta \, d\theta \tag{2}$$

and is a measure of the average forward momentum lost by an electron when it collides with a molecule. To obtain the integral and momentum transfer cross sections in the present work, the measured differential cross section data are extrapolated from 10° to 0° and from 120° to 180° using a fitting procedure based on an expansion of Legendre polynomials. The errors associated with this process are large due to the variation of the differential cross section with angle and the large angular range over which the differential cross section had to be extrapolated; typically the errors for σ_t and σ_m were 20% and 25% respectively. The values of σ_t and σ_m obtained are shown in table 3. Total cross sections (σ_T) were derived by addition of the ionization cross sections of Rapp and Englander-Golden (1965) to values of σ_t given in table 3 and these are displayed in figure 3. Below 10 eV the contribution from vibrational excitation may be significant but no integral cross section values for this process are available. Therefore the values of the total cross sections shown in figure 3 derived from the extrapolated differential cross sections should be taken as minimum values. Also presented are the direct measurements of the total cross section by Kwan *et al* (1984) and Szmytkowski *et al* (1984, 1989). It is observed that there is good agreement between the total cross sections derived from the present work and those obtained from a direct measurement

Table 3. Absolute integral and momentum transfer cross sections for electron scattering from N_2O ($\times 10^{-16}$ cm^2). The numbers shown as percentages represent the contribution to values of σ_t and σ_m from the extrapolated values of the differential cross sections.

	Incident energy (eV)					
	5	7.5	8.0	8.5	10	12
σ_t	9.7	9.1	10.8	11.6	13.7	17.0
	12%	27%	25%	27%	12%	26%
σ_m	7.8	8.9	10.0	11.1	11.7	14.2
	27%	50%	49%	53%	53%	58%
	15	20	30	50	80	
σ_t	16.2	14.8	12.3	9.3	4.5	
	23%	17%	16%	17%	16%	
σ_m	12.3	8.6	6.4	4.8	2.1	
	55%	53%	56%	62%	64%	

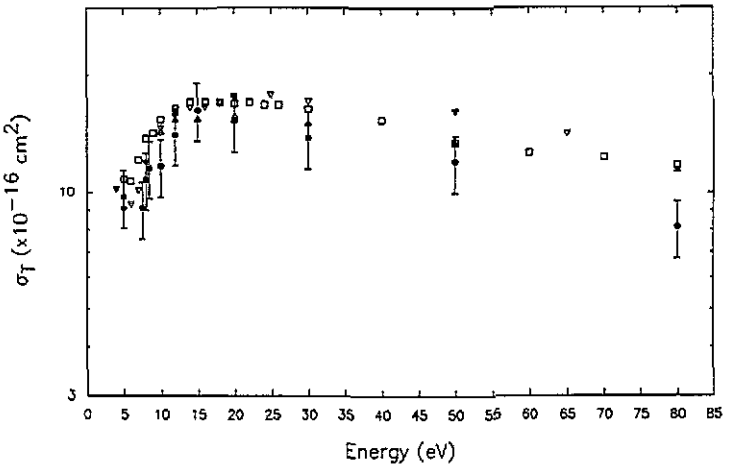

Figure 3. Absolute total cross sections for electron scattering from N_2O ($\times 10^{-16}$ cm^2). ●, present work; △, Marinković *et al* (1986); ▽, Kwan *et al* (1984); □, Szmytkowski *et al* (1984) and (1989).

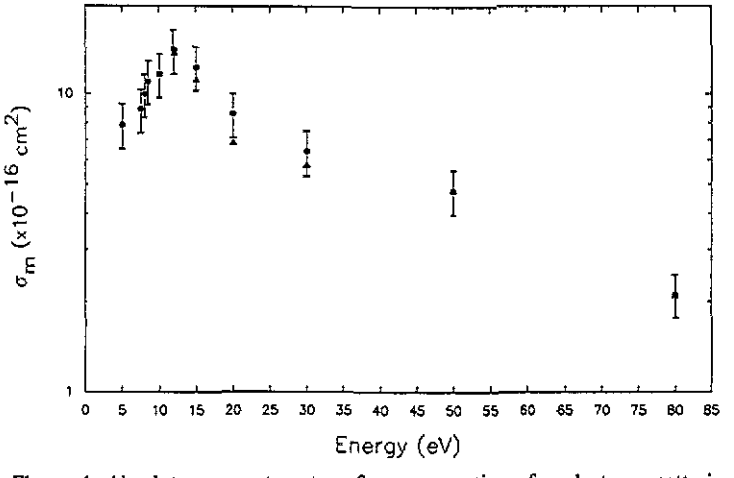

Figure 4. Absolute momentum transfer cross sections for electron scattering from N_2O ($\times 10^{-16}$ cm^2). ●, present work; △, Marinković *et al* (1986).

in that they all show a maximum at 15 eV. The momentum transfer cross sections are shown in figure 4 and the general trend of this cross section is similar to that of σ_t (see table 3), with the cross section rising to a peak at 12 eV and then falling away as the incident energy increases.

4. Comparison between N_2O and CO_2 cross sections

Different molecules are known to have similar electron scattering properties. In particular investigations of the electron scattering cross sections from CO and N_2 have revealed several similarities between the shape and magnitude of the cross sections. Kwan *et al* (1984) have shown that the total cross sections for N_2 and CO agree to within 5% for energies greater than 20 eV, while Jain (1982) has noted that the elastic

differential cross sections are similar in magnitude and shape for energies ≥ 40 eV. These striking similarities have been attributed to the similar structure and geometries of these two molecules.

N_2O and CO_2 are isoelectronic (containing the same number of bound electrons) and are linear in their ground states. Therefore, as with N_2 and CO, the electron scattering cross sections might be expected to show similar characteristics. Kwan *et al* (1984) were the first to investigate this possibility when they compared their own total cross section measurements for N_2O and with the CO_2 total cross sections of Hoffman *et al* (1982) and Kwan *et al* (1983) and found good agreement between the σ_T measurements of the two molecules for incident energies of 15 eV and above. Figure 5 compares the N_2O elastic differential cross sections measured in the present work and the CO_2 cross sections of Register *et al* (1980a) at 10 eV and those of Kanik *et al* (1989) for 20 eV and 50 eV. At 10 eV the CO_2 cross sections are lower than the N_2O cross sections for all angles below 90° and this may be a result of the permanent dipole moment of the N_2O molecule which could increase the differential cross sections at the smaller scattering angles. At 20 eV and 50 eV the agreement between the N_2O and CO_2 differential cross sections is good. Not only do the shapes of the cross sections agree well but the magnitudes are also in good agreement.

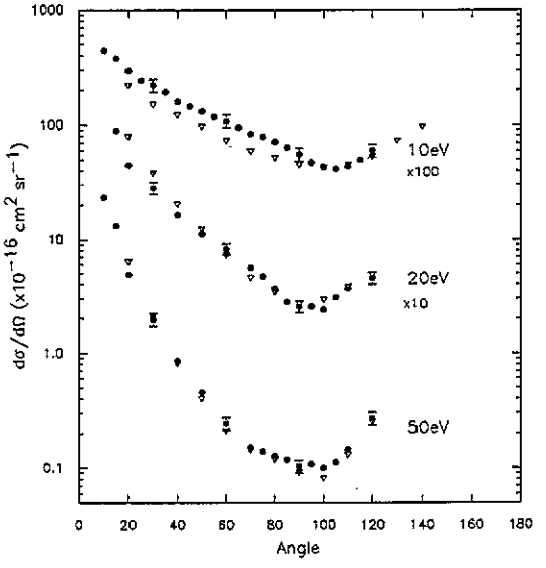

Figure 5. Comparison of the differential cross sections for electron scattering from N_2O and CO_2 ($\times 10^{-16}$ cm^2 sr^{-1}). 10 eV: ●, N_2O, present work; ▽, CO_2, Register *et al* (1980). 20 eV: ●, N_2O, present work; ▽, CO_2, Kanik *et al* (1989). 50 eV: ●, N_2O, present work; ▽, CO_2, Kanik *et al* (1989).

5. Conclusion

In this paper, the elastic differential, integral and momentum transfer cross sections have been presented. In general, there was good agreement between the present work and the previously published works of Kubo *et al* (1981) and Marinković *et al* (1986).

The largest disagreements between all three works were found around the minimum in the angular differential cross sections where the data of Marinković *et al* (1986) were generally found to be lower than the present work or those of Kubo *et al* (1981). The angular differential cross sections were also compared with those of CO_2 and found to be in good agreement for energies above 15 eV; this is believed to be a result of the similar electronic structure of the N_2O and CO_2 molecules.

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