

## Total electron scattering cross-sections for nitrous oxide at low electron energies

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MS received 15 December 1997; revised 13 January 1998

**Abstract.** Absolute total electron scattering cross sections for nitrous oxide have been measured at low electron energies using a photoelectron source. The measurements have been carried out at 19 electron energies varying from 0.73–9.14 eV with an accuracy of  $\pm 3\%$ . The cross sections obtained in the present experiment have been compared with other measurements.

**Keywords.** Electron scattering; total cross sections; nitrous oxide.

PACS No. 34.80

The scattering of low energy electrons by atoms and molecules has been studied for many years. It has been found to be important for the better understanding of weakly ionized plasma. The electron collision studies of nitrous oxide molecules are of particular importance as N<sub>2</sub>O lasers are now being used as a secondary frequency standard in areas of spectroscopy [1]. Also, nitrous oxide plays an important role in stratospheric chemistry and eventually affects the Earth's climate [2].

Total electron scattering cross sections for nitrous oxide were measured many years ago by Brüche [3] at electron energies larger than 1.0 eV and by Ramsauer and Kollath [4] at energies varying from 0.1–1.2 eV. Since then, only three more measurements have been reported using high energy resolution electron spectrometers. Zecca *et al* [5] used a retarding potential difference electron monochromator to obtain normalized total cross sections at electron energies ranging from 0.75–7 eV whereas a linear transmission method was employed by Szymkowski *et al* [6] to obtain absolute cross section values in the energy range 0.8–40 eV. Absolute total cross sections for 1–500 eV electron scattering of nitrous oxide were also reported by Kwan *et al* [7] by a beam transmission method. However, some relative transmission measurements have been carried out by Boness *et al* [8] and Sanche and Schulz [9] in the energy range 0–6 eV, but unfortunately, it is not possible to obtain cross sections from these results. Also, no theoretical computations have so far been reported in the literature for obtaining the absolute total electron scattering cross sections for nitrous oxide. In a nutshell, there are only four measurements available for a possible comparison of cross section values at electron energies from 0.7–10 eV. Although the general shape of the cross section curves reported by Szymkowski *et al* [6], Kwan *et al* [7], Zecca *et al* [5] and Brüche [3] is more or less

similar with respect to electron energy, the cross section values in some cases are different. There is a large discrepancy in cross section values at and around the well-known  $2\sum^+$  shape resonance centered at 2.3 eV energy [10]. Around the peak of the shape resonance, the cross section value reported by Szmytkowski *et al* [6] is about 26% higher than that obtained by Brüche [3] and is larger by a factor of 2.3 than the normalized cross section given by Zecca [5]. However, there is satisfactory agreement in cross section values reported by Kwan *et al* [7] and Szmytkowski *et al* [6] at the peak of the shape resonance. Also, there is a large discrepancy in the results obtained by the four groups on both sides of the shape resonance at energies ranging from 1.5–3.5 eV. In view of this, we decided to measure the absolute total scattering cross sections for nitrous oxide at low electron energies.

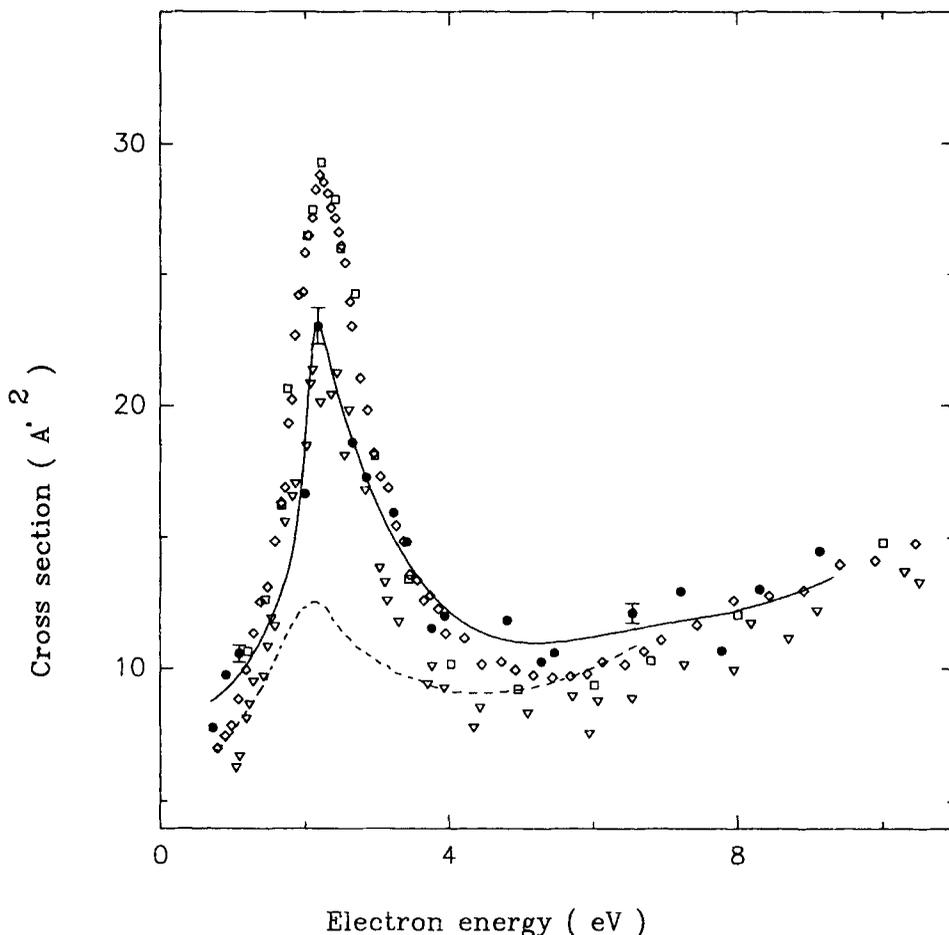
This paper is part of the ongoing research programme taken up to measure total electron scattering cross sections for atoms and molecules at low electron energies using a photoelectron source. Previously, absolute total cross sections for helium, neon [11], argon, krypton, xenon [12], molecular hydrogen [13] and molecular oxygen [14] have been measured at electron energies from 0–10 eV. In this paper, we present measurements of scattering cross sections for electrons scattered by nitrous oxide with projectile energy varying from 0.73–9.14 eV.

A detailed description of the experimental set-up used for the study, the method for analysis of the data and error analysis has been reported previously [11,12]. In brief, the experiment consists of measuring the intensities of the peaks in the photoelectron spectra of the source gas such as argon, krypton, xenon or nitrous oxide itself. The photoelectrons thus produced by the source gas are scattered by the target gas, the cross section of which is to be determined. Each peak in the photoelectron spectrum of the source gas provides one point in the electron energy scale. Complete scanning of the electron energy is performed by varying the energy of the ionizing radiation as well as changing the source gas. Using different combinations of photons of three different wavelengths (HeI, 58.4 nm, NeI, 73.6 nm and 74.4 nm) and four different source gases and neglecting the energy points where the statistics for the photoelectron intensity were poor, it was possible to measure scattering cross sections at 19 electron energies.

The electron scattering cross sections for nitrous oxide were measured using the method described previously. When source and target species are different, the electron scattering cross section  $\sigma$  can be evaluated by using the following equation:

$$\ln\left(\frac{I_{e2} I_{\lambda 01}}{I_{e1} I_{\lambda 02}}\right) = \frac{P_1 - P_2}{760} [n_0 \sigma x + k(al_1 + l_2)]. \quad (1)$$

Here  $I_{e1}$  and  $I_{e2}$  are the amplitudes of the photoelectron peaks at two different gas pressures  $P_1$  and  $P_2$ ,  $I_{\lambda 01}$  and  $I_{\lambda 02}$  are the incident photon intensities monitored during the experiment by the beam monitor at the two pressures,  $l_1$  is the distance from the centre of the beam splitter to the circular aperture covering the ionization region,  $l_2$  is the distance from the same aperture to the actual ionization region defined geometrically by different slits in the acceleration region and cylindrical mirror analyser,  $x$  is the scattering path length,  $a$  is the ratio of the pressures of the target gas outside and inside the ionization region and  $k$  is the photoabsorption coefficient of the target gas at a particular wavelength. The total photoabsorption cross sections at the three incident photon wavelengths



**Figure 1.** Total electron scattering cross section for nitrous oxide as a function of incident electron energy from 0.73–9.14 eV obtained by different researchers. The symbols denote as follows:  $\nabla$ , Brüche [3]; — — —, Zecca [5];  $\diamond$ , Szmytkowski *et al* [6];  $\square$ , Kwan *et al* [7];  $\bullet$ , present work; ———, cross section curve after proper curve fitting (present work).

have been taken from Samson and Yin [15]. The ratios of  $I_{\lambda 01}$  and  $I_{\lambda 02}$  were determined from the beam splitter as described previously by Kumar *et al* [11].

When source and target gas species are the same, the electron scattering cross sections could be calculated by the equation given below:

$$\ln\left(\frac{I_{e2} I_{\lambda 01} P_1}{I_{e1} I_{\lambda 02} P_2}\right) = \frac{P_1 - P_2}{760} [n_0 \sigma x + k(al_1 + l_2)]. \quad (2)$$

The electron scattering cross sections for nitrous oxide were measured using equations (1) and (2) respectively in the two cases described above. In both these equations, all parameters  $I_{e1}$ ,  $I_{e2}$ ,  $I_{\lambda 01}$ ,  $I_{\lambda 02}$ ,  $P_1$  and  $P_2$  could be determined experimentally and cross sections could be deduced.

All errors in the measurement of electron-scattering cross sections have been discussed in detail previously by the present authors. In the present experiment, the most probable error was estimated to be  $\pm 3\%$ . The problem of inadequate discrimination against forward scattered electrons has been attempted to estimate the magnitude of this effect on the measured cross sections in the experimental set-up used by us. A Monté Carlo method was developed and the electron trajectories originating from the ionization region and terminating at the detector were numerically studied using five random generators i.e. the finite length of the ionization region where photoelectrons are produced, random angle of projection within the allowed cone of ejection determined by the geometry, spread in energy of the photoelectrons determined by thermal broadening, place where a chosen fraction of the emitted photoelectrons are undergoing forward scattering and deflection in angle within a specified cone due to forward scattering. Assuming a differential scattering cross section of  $100 \text{ \AA}^2$  per steradian, the error in total cross section due to lack of discrimination of forward scattered electrons was found to be dependent on the overall cone angle of emergence. For our experimental set-up where the overall cone angle of emergence has been estimated to be  $6^\circ$ , the maximum error has been found to be about 0.5%.

The total electron-scattering cross sections for nitrous oxide as measured in the present experiment are shown in figure 1 for electron energies ranging from 0.73–9.14 eV along with error bars at three energies only. Also shown in the figure are the measured cross sections of Brüche [3], Zecca *et al* [5], Szmytkowski *et al* [6] and Kwan *et al* [7]. The cross section values reported by Ramsauer and Kollath [4] is not shown in the figure as there are only 10 cross section values available at energies from 0.1–1.2 eV and 3 values between 0.7 to 1.2 eV. The energy width for electron beams used by Kwan *et al* [7] was about 100 meV and the experimental error for absolute total cross section measurement was about 5% at 1.2 eV and 3% at higher electron energies.

Szmytkowski *et al* [6] reported an overall experimental error (calculated as the linear sum of all single contributions) of 10% at the lowest electron energy, rising to 12% at the sharp maxima and decreasing to 8% at and above 10 eV. The energy spread of the electron beam used by their group was about 70 meV. Relative total scattering cross sections were measured by Zecca *et al* [5] in an attempt to locate broad resonance, if any, and to determine the absolute size of the sharp resonances observed by Sanche and Schulz [9]. However, their relative cross sections were normalized at 4 eV to the value obtained by Brüche [3] at the same energy. Also shown in figure 1 is the cross section curve after proper curve fitting to the results obtained in the present experiment. This has been carried out using an appropriate mathematical function in the library of large number of such functions available in the Tablecurve-2D (Jandel Scientific) software for which the regression coefficient is closest to unity. It may be pointed out here that the theoretically computed electron scattering cross sections for nitrous oxide have not been shown in figure 1 as these have not been reported so far in literature by any research group.

For the sake of discussion, the cross section curve (figure 1) in the entire electron energy range from 0.73–9.14 eV has been divided into two regions. These include regions below 5.5 eV and between 5.5 and 9.14 eV. In the first energy region a sharp increase in cross section up to the maximum of the shape resonance centered around 2.3 eV is noticed followed by a sharp decrease up to about 5 eV energy. The temporary state

involved was the  $^2\Sigma^+$  of the  $N_2O^-$  ion. From figure 1, it is clear that the cross section values at the maximum of the resonance peak and also at the wings on both sides of the peak show large discrepancies. To have a clear comparison, the cross section curves reported by Brüche [3], Zecca *et al* [5], Szmytkowski *et al* [6] and Kwan *et al* [7] along with the curve obtained in the present experiment have been made to undergo proper curve fitting procedure. This has been done with the help of Tablecurve-2D (Jandel Scientific) software. The energy and the cross section value at the peak of the shape resonance thus obtained are 2.3 eV and  $28.8 \text{ \AA}^2$  [6], 2.3 eV and  $29.1 \text{ \AA}^2$  [7], 2.1 eV and  $12.5 \text{ \AA}^2$  [5], 2.3 eV and  $21.9 \text{ \AA}^2$  [3], and 2.2 eV and  $23.2 \text{ \AA}^2$  (present work) respectively. The electron energy value at the peak of the shape resonance has been reported by different research groups to be the same but the cross section values have been found to be different. The cross section values reported at the peak by Szmytkowski *et al* [6] and Kwan *et al* [7] are same within the stipulated experimental errors but these values are larger by about 25% as compared to the cross section obtained in the present measurement. The cross section value measured in the present work is about 6% higher than that obtained by Brüche [3] whereas the value given by Zecca *et al* [5] is about 46% lower than that reported in the present work. It has been mentioned earlier in the text that the relative data of Zecca *et al* [5] has been normalized at 4.0 eV to the cross section value reported by Brüche [3]. It is difficult to visualize why the normalization was carried out at 4.0 eV because this energy neither corresponds to the maximum nor the minimum of the resonance peak. Also, the ratio of the cross section values at the maximum and the minimum ( $9.1 \text{ \AA}^2$  at 4.4 eV electron energy) of the resonance peak after normalization is extremely small i.e. about 1.4. In comparison, such a ratio varies from 2.1 to 3.2, the smallest and the largest being obtained from data reported in the present experiment and by Kwan *et al* [7]. Incidentally, the shape resonance passes through the minima at 5.4 eV, 5.1 eV, 5.0 eV and 5.2 eV having cross section values 9.6, 9.0, 8.3 and  $11.0 \text{ \AA}^2$  as given by Szmytkowski *et al* [6], Kwan *et al* [7], Brüche [3] and the authors in the present work respectively. Except for Zecca's results, all other four measurements are fairly reliable. However, there are two sets of cross section values available from the four measurements for the peak of the shape resonance. One set of values, from Szmytkowski *et al* [6] and Kwan *et al* [7] is about 28% higher than that given by Brüche [3] and authors in the present work. At this stage, it may be worthwhile to discuss about error in all the four measurements associated with incomplete discrimination against projectiles at small angles in the forward direction. In the present experiment, the maximum error because of lack of such discrimination has been found to be about 0.5% whereas such an error in the measurements by Kwan *et al* [7] is also small, of the order of 0.5 to 1%. Brüche [3] has not discussed about this potential source of error associated with their instrument, whereas Szmytkowski *et al* [6] have found an error of about 1% at 60 eV and even smaller at lower energies. In view of the above discussion, it is clear that two sets of cross section values exist for the peak of the shape resonance and this controversy needs to be resolved in future.

In the energy region from 5.5–9.14 eV, the cross section curve after passing through a minimum on the decreasing wing of the main shape resonance peak shows an increasing trend in cross sections at higher energies. In the measurements reported by Zecca *et al* [5], the cross sections have been measured up to 7 eV only. All measurements including those reported by Zecca *et al* [5] show that the cross section values differ slightly in each

**Table 1.** Total electron scattering cross sections for nitrous oxide at various electron energies. Also given are the photon wavelengths and the source gas and the photoion state for the corresponding electron energy.

Ionizing line (Å)	Source gas	Photo-ion state	Electron energy (eV)	Cross section (Å <sup>2</sup> )
744	Ar	$^2P_{1/2}$	0.73	7.76
744	Ar	$^2P_{3/2}$	0.91	9.74
736	Ar	$^2P_{3/2}$	1.09	10.56
744	Kr	$^2P_1$	2.00	16.66
736	Kr	$^2P_1$	2.18	23.02
744	Kr	$^2P_{3/2}$	2.66	18.58
736	Kr	$^2P_{3/2}$	2.85	17.28
744	Xe	$^2P_{1/2}$	3.23	15.94
736	Xe	$^2P_{1/2}$	3.41	14.82
744	N <sub>2</sub> O	$\tilde{X}^2\Pi(\nu = 0)$	3.76	11.52
736	N <sub>2</sub> O	$\tilde{X}^2\Pi(\nu = 0)$	3.94	12.00
584	N <sub>2</sub> O	$\tilde{A}^2\Sigma^+(\nu = 0)$	4.81	11.84
584	Ar	$^2P_{1/2}$	5.28	10.25
584	Ar	$^2P_{3/2}$	5.46	10.61
584	Kr	$^2P_{1/2}$	6.55	12.11
584	Kr	$^2P_{3/2}$	7.22	12.92
584	Xe	$^2P_{1/2}$	7.78	10.68
584	N <sub>2</sub> O	$\tilde{X}^2\Pi(\nu = 0)$	8.31	13.01
584	Xe	$^2P_{3/2}$	9.14	14.48

case, the difference being more than the stipulated experimental errors. The cross section values reported in the present experiment in this electron energy region are the highest whereas those reported by Brüche [3] are the lowest.

The values of scattering cross sections for nitrous oxide as measured in the present experiment are given in table 1 along with the photon wavelength, source gas and the photon state for the corresponding electron energy.

As nitrous oxide and carbon dioxide molecules are isoelectronic and linear in their ground state, it may be interesting to find out the difference in the electron scattering cross section curves for the two molecules. The cross section measurements for the CO<sub>2</sub> have also been carried out by the present authors and these would be published elsewhere in detail. Our measurements in the case of these two molecules show two broad features. Qualitatively, the shape of the cross section curves of N<sub>2</sub>O and CO<sub>2</sub> has striking similarities, but quantitatively, it has been found that the cross section values for CO<sub>2</sub> are smaller than those obtained for N<sub>2</sub>O at all electron energies below 10 eV. Secondly, the resonance peaks have been found at different electron energies i.e. 2.3 and 3.8 eV for N<sub>2</sub>O and CO<sub>2</sub> respectively. It may be worthwhile to make an attempt to explain at least the first broad feature. N<sub>2</sub>O is a polar molecule with a permanent dipole moment whereas CO<sub>2</sub> is a non-polar molecule. It has been found that at smaller angles, the differential cross sections at intermediate energies are larger in the case of polar molecules than those for the non-polar molecules [16, 17]. It is presumed that the same is true at low electron energies also. In that case, the total cross sections would also be larger in case of molecules having permanent dipole moment.

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