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1989 J. Phys. B: At. Mol. Opt. Phys. 22 525

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## Total absolute cross section measurements for electron scattering on NH<sub>3</sub>, OCS and N<sub>2</sub>O

Czesław Szmytkowski<sup>†</sup>, Krzysztof Maciąg<sup>†</sup>, Grzegorz Karwasz<sup>‡</sup> and Dušan Filipović<sup>§</sup>

<sup>†</sup> Department of Physics, Technical University of Gdańsk, Gdańsk, Poland

<sup>‡</sup> Polish Academy of Sciences, IMP PAN, Gdańsk, Poland

<sup>§</sup> Institute of Physics, Beograd-Zemun, Yugoslavia

Received 13 June 1988, in final form 26 September 1988

**Abstract.** The absolute total electron scattering cross sections for NH<sub>3</sub>, OCS and N<sub>2</sub>O were measured using a linear transmission technique. The presented results cover the range of impact energy from 1 to 80 eV for NH<sub>3</sub> and from 40 to 100 eV for OCS and N<sub>2</sub>O. The most prominent feature of the cross section for NH<sub>3</sub> is the very broad hump centred at about 10 eV with a maximum value of  $23.5 \times 10^{-20} \text{ m}^2$ . Above 10 eV the present results on NH<sub>3</sub> are in excellent agreement with recent calculations of Jain.

### 1. Introduction

Absolute total cross sections for electron scattering were some of the first experimentally studied data for electron-atom and electron-molecule scattering. In spite of this good tradition, the agreement between experimental results from different laboratories may be considered satisfactory only for noble gases and a very few molecules. Visible discrepancies between cross sections are caused by apparatus effects of different measurement techniques, difficult to grasp and estimate, and often exceed the declared experimental errors. On the other hand, for a number of molecules available data are only fragmentary and are limited to low energies where shape resonance structures are dominant.

For the ammonia molecule the known total cross sections are absolute cross sections, below 50 eV, measured by Brüche (1929) on a Ramsauer-type apparatus and most recently, normalised data of Sueoka *et al* (1987) obtained with the help of a TOF transmission method.

Jain (1988) has recently carried out calculations of the total (elastic + absorption) cross sections for the ammonia molecule using a parameter-free spherical complex optical potential. The differences between the results obtained using different models for the potential components have been so distinct that their comparison with the experiment could give a hint as to which of the proposed electron-molecule interaction models better describes the scattering process. Large discrepancies, up to 15%, between the available experimental data make it difficult to draw such a conclusion.

Measurements of total cross sections for OCS have so far been carried out in the lower energy range. Total absolute cross sections, below 40 eV, were presented by

Szmytkowski *et al* (1984) and Dababneh *et al* (1985), and normalised, integrated total cross sections in the range 0.3–5 eV by Sohn *et al* (1987).

The continuum multiple-scattering method employed by Lynch *et al* (1979) for calculations of vibrationally elastic cross sections in OCS, describes fairly well the qualitative behaviour of the cross section, especially the resonance structure at low energies. The evaluation of the degree of usefulness of this model for  $e^-$ -OCS scattering in a wider energy range is impossible due to the lack of experimental data for intermediate energies.

In the present experiment we have measured total absolute cross sections for electron scattering on  $\text{NH}_3$ , OCS and  $\text{N}_2\text{O}$  using a non-magnetic linear transmission method. We compare our results with the previous experimental data and theoretical calculations.

## 2. Experimental

The experimental procedure was based on the relation between the attenuation of an electron beam passing through the gas under study and the total cross section as given by the de Beer-Lambert law (for detailed discussion see the review of Bederson and Kieffer 1971). In the reported experiment energy selected electrons covered a distance  $L$  in the gas target. The electrons which succeeded in leaving the scattering region through the exit orifice of the chamber were collected by a Faraday cup.

The total cross section  $\sigma(E)$  was evaluated for each energy  $E$  using a formula:

$$\sigma(E) = \frac{\ell(T_m T_c)^{1/2}}{L \cdot p_m} \ln \frac{I_0(E)}{I_g(E)} \quad (1)$$

where  $\ell$  is the Boltzmann constant and  $I_0/I_g$  is the ratio of electron transmitted currents without and with the gas target in the collision chamber, respectively. Among the experimentally measured quantities,  $p_m$  is the gas pressure in the scattering volume as measured with a Baratron gauge, and  $T_m$  denotes the temperature at which the manometer sensor is held while  $T_c$  is the collision chamber temperature. Since the temperature of the manometer head  $T_m = 322 \pm 1$  K was usually higher than that of the gas scattering chamber the expression (1) takes into account the correction for the thermal transpiration effect (Knudsen 1910).

In the course of the experiment, the target gas was let alternately into the scattering volume in such a way that the background pressure outside the scattering chamber was kept at the same value (below  $2 \times 10^{-4}$  Pa) in both modes of operation. This enabled us to reduce the influence of the target gas, effusing from the collision chamber, on the intensity of the primary electron beam and therefore on the measured cross section.

Using effective path length calculations of Nelson and Colgate (1973) we deduced that the effect of density drop across the orifices of the scattering cell is less than 0.4%, and the geometrical distance between them (30.5 mm) was adopted as the absorption length  $L$ .

The other experimental details have been described elsewhere (see e.g. Szmytkowski *et al* 1984, 1987).

In order to lessen the influence of apparatus effects, the measurements were carried out for a given energy in a series of runs using different sets of electron beam controlling parameters and a range of target pressures ( $0.07 < p_m < 0.4$  Pa).

To obtain the final total cross section at a particular energy, the average values from different series of runs for the same energy were themselves averaged and weighted according to their uncertainties.

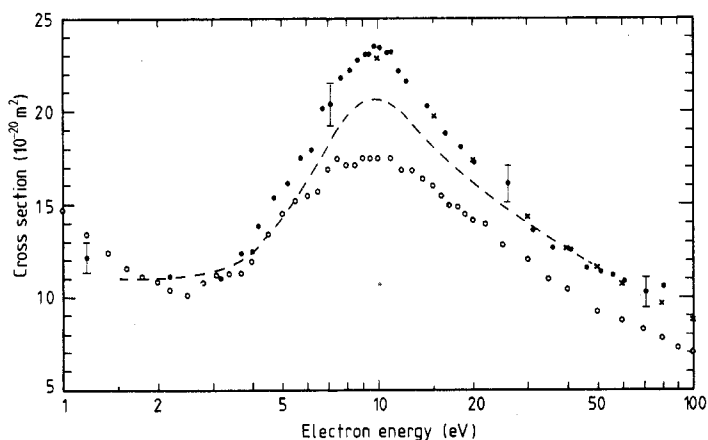
The statistical uncertainties (one standard deviation of weighted mean values) did not exceed 2% over the whole energy range.

In order to estimate the systematic error due to incomplete discrimination against detection of electrons which experienced scattering in the small-angle forward direction, we used a procedure similar to that of Jost and Ohnemus (1979). It is worth stressing that in these calculations we have considered not only the electrons scattered on the beam axis but also those scattered beyond it, which due to the finite dimensions of the incident beam and the exit orifice of chamber may, after collision, reach the collector. Differential cross sections measured by Harshbarger *et al* (1971), Ito *et al* (1983) and Marinković *et al* (1986) for  $\text{NH}_3$ ,  $\text{OCS}$  and  $\text{N}_2\text{O}$  molecules, respectively, were used to estimate the corresponding errors. The lowering of the magnitude of measured total cross sections for all studied molecules does not exceed 1% for low energies, but around 100 eV it can be as high as 4%.

Pessimistic estimation of the overall systematic error (the direct sum of errors of all measured quantities) gives values equal to 8% below 2.5 eV, decreasing to 6% in the range 5–20 eV and increasing again up to 10% at the highest applied energies.

### 3. Results

The results of measurements of total cross section for electron scattering on  $\text{NH}_3$  molecules are shown in figure 1. The most prominent feature of the cross section is a very broad structureless hump around 10 eV with a maximum value of  $23.5 \times 10^{-20} \text{ m}^2$ . Experiments on vibrational excitation of the  $\text{NH}_3$  electronic ground state (Ben Arfa and Tronc 1985, Cvejanović *et al* 1987b) have indicated that a very broad shape resonance occurs around 6–7 eV. This short-lived negative-ion state of  $\text{NH}_3$  is created



**Figure 1.** Total cross sections for electron-ammmonia scattering. Experiment: ●, present results, absolute; ---, Brüche (1929), absolute; ○, Sueoka *et al* (1987), normalised. Theory: +, Jain (1988). The error bars correspond to the overall experimental uncertainties at some selected points.

by the trapping of an extra electron in the unoccupied  $\sigma^*$  valence orbital. This shape resonance overlaps another,  $^2A_2''$   $\text{NH}_3^-$  Feshbach resonance, localised between 5 and 6 eV. Evidence of its existence was observed mainly in cross sections for negative ion production (Compton *et al* 1969, Sharp and Dowell 1969, Stricklett and Burrow 1986, Tronc *et al* 1988) and in residual energy spectra (Cvejanović *et al* 1987a).

It is worth noting that the general energetic dependence of the cross section for  $\text{NH}_3$  is similar to cross sections in isoelectronic  $\text{H}_2\text{O}$  (Sueoka *et al* 1987, Szmytkowski 1987) and  $\text{CH}_4$  (Sueoka and Mori 1986, Lohmann and Buckman 1986) molecules. Cross sections for all these molecules exhibit very broad humps centred around 10 eV. Similarly, as in  $\text{NH}_3$ , this increase of cross sections is attributed to the formation of resonant states at impact energies of 5–7 eV. The differences in cross sections, visible for lower energies, are probably partly connected with significant differences of permanent dipole moments of these molecules. As the dipole moment decreases, the minimum in the cross section becomes deeper and shifts towards lower energies.

From comparison of  $\text{NH}_3$  ionisation cross section data (Djurić-Preger *et al* 1976, Crowe and McConkey 1977, Märk *et al* 1977, Bederski *et al* 1980, Orient and Srivastava 1985) with our total cross sections it follows that the role of ionisation in the  $e^-$ - $\text{NH}_3$  scattering process increases gradually from threshold to about 100 eV, and at 100 eV amounts to at least 30%.

In figure 1 we have presented for comparison the experimental results of Brüche (1929) and the latest measurements of Sueoka *et al* (1987). The general run of all curves, in the common energy range, is similar, but the magnitudes of cross sections obtained with different techniques are different. The biggest discrepancies can be seen around the maximum, where the data of Brüche (1929) and Sueoka *et al* (1987) are smaller than the present results by about 13 and 29%, respectively. For energies below 4 eV and above 30 eV the agreement with Brüche's results is good, while for higher energies the discrepancy with data of Sueoka *et al* is on average about 20%. A similar discrepancy between the data of Sueoka's group and that from other laboratories is seen also for some other investigated targets. The systematic lowering of Sueoka's results may be partly connected with the normalisation procedure and, especially at higher energies, with the forward scattering effect in their experiments. The effective scattering length in Sueoka's measurements was usually determined from the normalisation of the  $e^+$ - $\text{N}_2$  results to those of Hoffman *et al* (1982). However, Sueoka and Mori (1984) noticed that if the normalisation is performed for  $e^-$ - $\text{N}_2$  data, the normalised total cross section would be 10% higher. Unfortunately, such details on the normalisation procedure in the case of  $\text{NH}_3$  (Sueoka *et al* 1987) are not given. The forward scattering effects were also not counted in Sueoka's measurements. By comparison with other TOF experiments (e.g. Hoffman *et al* 1982) we expect that the lowering of measured cross sections could be several per cent at energies above 50 eV.

A comparison (figure 1) of the total cross section calculated by Jain (1988), using the correlation-polarisation potential along with the polarised electronic density in the imaginary (absorption) part of the potential, with the present measurements shows an excellent agreement between theory and experiment.

We have also carried out measurements of total cross section for OCS and  $\text{N}_2\text{O}$ . For energies below 40 eV, the present results are, within experimental uncertainties, in very good agreement with our previous measurements (Szmytkowski *et al* 1984), with characteristic resonant maxima attributed to short-lived low-energy shape resonant states. Results for energies from 40 to 100 eV are presented in numerical form in table 1 along with the available data of other authors.

**Table 1.** Total cross sections for  $OCS$  and  $N_2O$  (in  $10^{-20} \text{ m}^2$ ).

Electron energy (eV)	OCS			N <sub>2</sub> O		
	Present	Dababneh <i>et al</i> (1985)	Lynch <i>et al</i> (1979) (elastic theory)	Present	Kwan <i>et al</i> (1984)	Brüche (1927)
40	26.8	24.2	20.26	14.5	17.3	14.1
45	26.0		18.95	14.0		13.2
48						12.9
50	24.7		17.92	13.6	16.6	
60	23.6		16.39	12.7		
70	22.6		15.34	12.4		
77					14.5	
80	21.5		14.56	11.8		
90	20.9		13.82	11.1		
100	20.2		12.95	10.5	13.0	

The present results for  $N_2O$  in the range 25–50 eV are in satisfactory agreement with measurements of Brüche (1927), but are generally about 15% lower than results of Kwan *et al* (1984). One of the reasons, which might cause a significant lowering of results, is the unsatisfactory discrimination of electrons scattered in the forward direction. However, in our experiment the greatest possible estimated lowering of the cross section due to this effect does not exceed 4%. It is worth noting that the discrepancy between the present results and those of the Detroit group is not systematic, because in the case of  $OCS$ , results from both laboratories show an opposite tendency to that for  $N_2O$ . For  $OCS$ , at 40 eV, our result is about 10% higher than measurements of Dababneh *et al* (1985). For comparison of the present total cross sections with elastic ones calculated by Lynch *et al* (1979), we need quantitative information on the inelastic processes. However, if due to the lack of such data one assumes that the contribution of ionisation (the most dominant inelastic process near 100 eV) in the scattering for  $OCS$  is similar, on average, to that for other triatomic molecules, then the agreement of the theory with experiment for intermediate energies seems to be quite satisfactory.

### Acknowledgment

This work has been supported in part by the Polish Ministry of National Education within Programme CPBP 01.06 under Project 3.01.

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