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Elastic scattering of low-energy electrons from ammonia

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Abstract. We report absolute differential cross section measurements for vibrationally elastic electron scattering from NH_3 at incident energies from 2–30 eV. The present results, from a crossed electron–molecular beam apparatus, represent the first comprehensive experimental attempt to quantify the elastic electron–NH_3 scattering process. At each energy studied we have integrated our differential cross section data to generate total elastic and elastic momentum transfer cross sections and a critical comparison of both our differential and integral cross sections against previous experiment and theory is provided. We also report our observation of a strong Feshbach resonance in the elastic channel at an energy of 5.59 ± 0.05 eV.

1. Introduction

The study of electron scattering from ammonia (NH_3) molecules is of practical interest due to its applications in space physics (such as in the modelling of planetary atmospheres), plasma chemistry, gas-discharge lasers, synthetic chemistry and switching devices. In particular, the discovery of NH_3 molecules in the interstellar medium has increased the attention paid to its spectroscopic and dynamical properties (Gianturco 1990), whilst in the field of plasma chemistry, NH_3 is of importance as it is a source of nitrogen atoms for the fabrication of nitride films and the fabrication of other nitrogen compounds (Sato *et al* 1986).

Previous experimental investigations into the electron–NH_3 scattering process have been limited. At the grand total cross section level there is the time-of-flight (TOF) experiment of Sueoka *et al* (1987) and the linear transmission experiment of Szmytkowski *et al* (1989), whilst Hayashi (1981, 1991) reports momentum transfer cross sections which are based on the drift velocity measurements of Pack *et al* (1962) and the calculations of Altshuler (1957) and Jain and Thompson (1983). Angular distributions for elastic scattering and the excitation of the normal vibrational modes of NH_3 in the energy regime 12–50 eV were reported by Furlan *et al* (1990). Unfortunately, these data are not absolute and so no quantitative conclusions can be gleaned from them. The only absolute differential cross sections currently available in the literature, for the energy regime of the present study, are the measurements of Ben Arfa and Tronc (1987) for elastic scattering and the excitation of the ν_1, ν_3 and ν_4 normal modes of vibration at 7.5 eV. Their measurements were placed on an absolute scale using a relative flow normalization technique with the N_2 elastic cross section of Srivastava *et al* (1976) serving as a reference standard. Hence, given

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this sparsity of available data for low-energy electron scattering from NH_3, it is quite clear that the present study is well justified.

From a theoretical perspective there exist several calculations for the elastic scattering of electrons from NH_3. The earlier work is well summarized by Gianturco and Jain (1986) and Gianturco (1990) and, in the interest of brevity, we do not discuss it further here. More recently there have been the Schwinger variational calculations of Pritchard *et al* (1989), the parameter-free model calculations by Gianturco (1990), Jain and Thompson (1991) and Baluja and Jain (1991), and the complex Kohn variational calculations of Lengsfield *et al* (1991). In each of these cases the cross sections were computed within the framework of the fixed-nuclei approximation (FNA) which leads to severe convergence problems in the calculation of electron–polar polyatomic molecule cross sections for the higher order partial waves (Pritchard *et al* 1989). As a result Pritchard *et al* (1989) only report intermediate and backward angle differential elastic scattering data where the first few partial waves dominate the description of the interaction. On the other hand, Gianturco (1990), Jain and Thompson (1991) and Lengsfield *et al* (1991) have attempted, within the FNA formalism, to circumvent this problem. This has been achieved by the spatial partitioning of the scattering problem such that a description of the higher order partial waves ($l > 6$) is given by the first Born approximation, whilst for $l \leqslant 6$, the solution is obtained by the usual close-coupling (Gianturco, Jain and Thompson) or Kohn (Lengsfield *et al*) calculation. Lengsfield *et al* have also included polarization and correlation effects via the use of an *ab initio* optical potential.

In the next section we discuss the details of the experimental procedure, including the normalization of the angular distributions, employed in the current measurements. Our results, and a discussion of them, are presented in section 3 with conclusions being drawn in section 4.

2. Experimental apparatus and procedures

The electron monochromator employed in the present measurements, and its principles of operation, were described in detail previously by Brunger *et al* (1991) (and references therein) and so we do not go into detail again here. Briefly, however, a beam of anhydrous NH_3 effusing from a multichannel capillary array is crossed with a beam of monoenergetic electrons of desired energy E_0. Vibrationally elastic (referred to as 'elastic' throughout this paper) scattered electrons at a particular scattering angle (θ) are energy analysed and detected. The data accumulation of the present relative angular distributions at each E_0 is performed entirely under computer control. This involves the control of the routing of the gas to the capillary array for the signal plus background measurement, and then to an alternative capillary at the periphery of the vacuum chamber for the background measurement; the regular measurement of the gas pressure; the angular positioning of the analyser; and the counting and analysis of the scattered intensity levels. Typical measurement times varied between 6 and 12 h with the stability of the apparatus being excellent throughout. This latter point is emphasized in that while each final angular distribution represents the weighted average of a number, in some cases as many as 16 individual distributions, the standard deviation of the combined data set, at any given θ, never exceeded 0.2%.

The scattered electron analyser can be rotated about the molecular beam allowing access to an angular range of $-20°$ to $125°$. The true $0°$ position was determined as

that about which the intensity of elastically scattered electrons was symmetric. The estimated error in this determination was $\pm 1°$. In the present series of measurements we have calibrated our energy scale by detecting, in the elastic channel, the 2^2S resonance in helium at 19.367 eV which has enabled us to determine the absolute energy scale to within 50 meV.

The energy resolution in the present measurements was typically 60 meV (FWHM). As excitation of the two lowest order normal modes of vibration of the NH_3 molecule (ν_2, ν_4) require only 118 and 202 meV respectively, it was possible that, given our energy resolution, electrons that had excited these transitions could have contributed to the elastic intensity. Consequently, at each E_0 studied, we measured at a representative forward, middle and backward angle, energy loss spectra which encompassed the elastic peak and the normal modes of vibration of the NH_3 molecule. It was found that in each case the contribution of the $\nu_{2,4}$ normal modes of vibration to the elastic intensity was negligible.

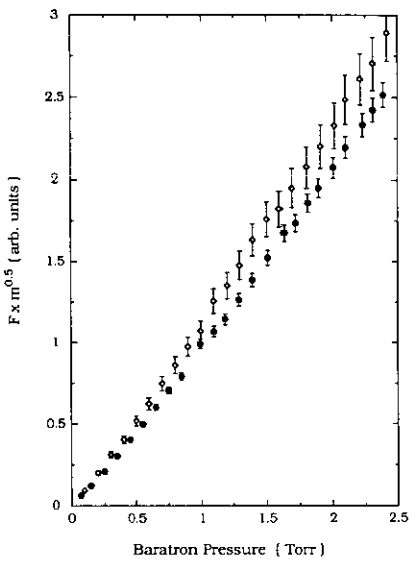

Figure 1. Relative flow rates for ammonia (○) and helium (●) as a function of the capillary driving pressure.

As in our previous studies of electron scattering from homonuclear diatomics (Brunger *et al* 1991, H_2; Brennan *et al* 1992, N_2) the absolute scale of the elastic NH_3 cross section, at a given E_0, was determined by a relative flow technique (see for example Nickel *et al* (1989)). To this end we have, in a separate series of experiments, measured the respective relative flow rates of ammonia and helium as a function of their capillary driving pressures (P). The technique used in these measurements is very similar to that of Khakoo and Trajmar (1986) and so we do not go into further detail here. The results of a typical calibration for NH_3 and He are given in figure 1, the driving pressures being measured by an MKS Baratron capacitance manometer.

The basis of the relative flow technique is that the elastic differential cross section for NH_3 is related to that for helium via

$$\frac{d\sigma}{d\Omega}(\theta)_{NH_3} = \frac{I_{He}}{I_{NH_3}} \frac{N_e(\theta)_{NH_3}}{N_e(\theta)_{He}} \frac{(Fm^{0.5})_{He}}{(Fm^{0.5})_{NH_3}} \frac{d\sigma}{d\Omega}(\theta)_{He} \tag{1}$$

where I are the electron beam currents, $N_e(\theta)$ are the scattered electron count rates, F the respective flow rates through the capillary array, m the respective molecular weights and $d\sigma/d\Omega(\theta)_{He}$ the known elastic cross section for helium. The choice of the standard helium cross section is discussed in detail elsewhere by Brunger *et al* (1992) but, in summary, for electron energies below the onset of the $n = 2$ inelastic threshold in helium we have employed the *ab initio* variational calculation of Nesbet (1978), whilst for the energies above this threshold we have used data previously measured in these laboratories (Brunger *et al* 1992).

In the case of our studies on homonuclear diatomics two important criteria were met.

(i) Up to a critical value of the capillary driving pressure, P_c, a plot of data for $(Fm^{0.5})_x$ against P_x ($x \equiv N_2, H_2, He$) lie on the same straight line. The importance of this result is that in (1) the ratio $(Fm^{0.5})_{He}/(Fm^{0.5})_{NH_3}$ could then simply be replaced by the easily measured ratio P_{He}/P_{NH_3}.

(ii) The value of this critical pressure, P_c, was sufficiently large that it was experimentally feasible, in terms of the scattered electron count rate, to perform the measurements of the required angular distributions for capillary driving pressures $P < P_c$.

From figure 1 it is apparent that for NH_3 and He the critical value of the driving pressure for which condition (i) is satisfied is less than about 0.4 Torr. Unfortunately, this value is too low for condition (ii) to be met. Consequently, we have used data such as in figure 1, at pressures less than 2 Torr, to derive a calibration curve of $(Fm^{0.5})_{He}/(Fm^{0.5})_{NH_3}$ as a function of P_{He}/P_{NH_3}. This enables us to determine the appropriate value for $(Fm^{0.5})_{He}/(Fm^{0.5})_{NH_3}$ to be substituted into (1) for any value of the ratio of the capillary driving pressures. Other constraints, such as the requirement to keep the mean free paths of the two gases identical at the entrance to the capillary array and the need to operate in the Clausing flow regime (Nickel *et al* 1989), place tight limits on the actual range of P_{He}/P_{NH_3} used. However, it is worth noting that for $P > P_c$, differences in the value of $(Fm^{0.5})_{He}/(Fm^{0.5})_{NH_3}$ compared with that determined from the simple ratio of the driving pressures, P_{He}/P_{NH_3}, can be significant and lead to an appreciable error in the absolute value of the cross section derived from the angular scattering data if not properly accounted for.

Having obtained the elastic differential cross sections at each energy studied, we then extrapolated the data to 0° and to 180° using the *shape* of the theoretical cross sections of Lengsfield *et al* (1991) as a guide. The only exception to this is at 30 eV where the calculation of Baluja and Jain (1991) is currently the only one available in the literature and consequently at this energy we have used this calculation to extrapolate to both the upper and lower limit of integration. These data were then integrated to give the experimental total elastic (Q_{el}) and momentum transfer (Q_{MT}) cross sections as defined by:

$$Q_{el} = 2\pi \int_0^{\pi} \left(\frac{d\sigma(\theta)}{d\Omega}\right)_{el} \sin\theta \, d\theta \tag{2}$$

$$Q_{MT} = 2\pi \int_0^{\pi} \left(\frac{d\sigma(\theta)}{d\Omega}\right)_{el} \sin\theta(1 - \cos\theta) \, d\theta. \tag{3}$$

3. Results and discussion

Differential cross sections for elastic scattering of NH_3 by electron impact are given in table 1. These differential cross sections have been determined at six incident energies between 2.0 and 30.0 eV, and at scattering angles in the range 10°–125°. The uncertainty in the present measurement of the elastic differential cross section is estimated to be ±8% (one standard deviation). This arises from the addition, in quadrature, of uncertainties associated with the flow rate calibration curve (6%), counting statistics in the angular distribution (< 1%), statistical uncertainty in the determination of the ratio of the NH_3 to He cross sections (4%), variations in gas pressure (1%) and electron beam current (2%) and an uncertainty in the helium cross section (2%).

Table 1. Differential cross sections for elastic electron scattering from ammonia (units of 10^{-16} cm^2 sr^{-1}).

	Energy (ev)					
θ_e^0	2.0	5.0	7.5	15	20	30
10	—	—	—	—	—	11.99
15	—	—	—	—	10.21	6.94
20	3.92	3.40	4.50	6.20	7.16	5.31
25	2.62	2.77	3.63	4.70	5.07	3.85
30	1.72	2.03	2.71	3.48	3.56	2.58
35	1.24	1.63	2.07	2.59	2.53	1.73
40	0.89	1.26	1.53	1.84	1.79	1.18
45	0.63	1.09	1.27	1.46	1.26	0.81
50	0.53	1.08	1.14	1.17	0.92	0.59
55	0.45	1.08	1.07	0.95	0.70	0.44
60	0.39	1.04	0.97	0.77	0.56	0.34
65	0.34	1.04	0.93	0.65	0.45	0.28
70	0.31	1.05	0.94	0.62	0.38	0.24
75	0.29	1.05	0.95	0.55	0.34	0.21
80	0.27	1.03	0.90	0.51	0.31	0.18
85	0.24	0.93	0.83	0.45	0.27	0.16
90	0.22	0.87	0.77	0.43	0.25	0.14
95	0.20	0.79	0.72	0.39	0.24	0.14
100	0.18	0.68	0.61	0.37	0.22	0.13
105	0.16	0.55	0.50	0.32	0.20	0.12
110	0.14	0.44	0.41	0.33	0.19	0.13
115	0.11	0.33	0.34	0.31	0.19	0.14
120	0.08	0.22	0.24	0.31	0.21	0.15
125	—	0.24	0.26	0.36	—	—

Representative examples of the present data at 2.0, 5.0, 7.5 and 20.0 eV can be found in figures 2–5 respectively. It is immediately apparent from these figures that the level of agreement between the present work and the calculations of Pritchard *et al* (1989), Gianturco (1990), Lengsfield *et al* (1991) and Jain and Thompson (1991) improves markedly as the incident electron energy increases.

At 2 eV (figure 2), the present absolute differential cross section is compared with the calculations of Gianturco (1990), Jain and Thompson (1991) and Lengsfield *et al* (1991). It bears little resemblance to either of the close coupling model calculations (Jain and Thompson 1991, Gianturco 1990), although it is in better accord with the

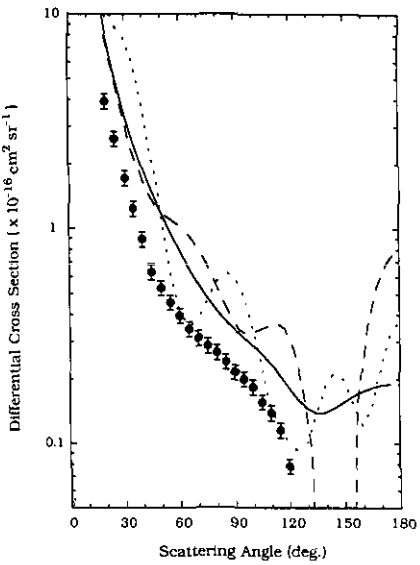

Figure 2. Differential cross section for elastic electron scattering from NH_3 at 2 eV: present data, ●; Gianturco, – – –; Jain and Thompson, - - -; and Lengsfield *et al*, ——.

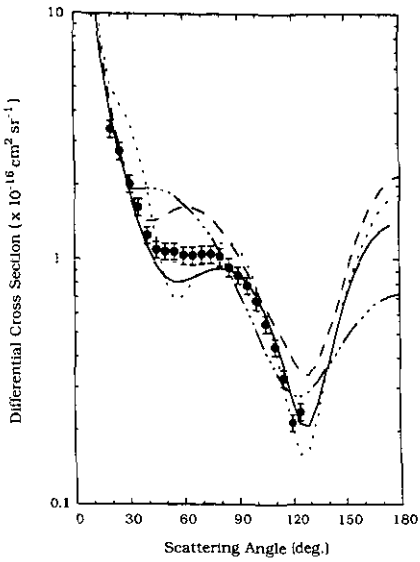

Figure 3. Differential cross section for elastic electron scattering from NH_3 at 5 eV: present data, ●; Gianturco, – – –; Jain and Thompson, - - -; Pritchard *et al*, — ··· —; and Lengsfield *et al*, ——.

shape of the calculation of Lengsfield and co-workers. We note the oscillatory structure in the calculation of Jain and Thompson which we do not observe in the present experiment. This sort of oscillatory behaviour is usually indicative of convergence problems associated with the calculation and/or incorrect averaging of the T-matrix elements. We do not believe it is physical.

At 5.0 eV (figure 3), the level of agreement between the present experiment and theory improves considerably, particularly for forward scattering angles and particularly in the case of Lengsfield and co-workers. With the exception of a small angular region around 60° where this calculation is a little lower in absolute magnitude, the overall agreement between it and the experiment is very good.

At 7.5 eV (figure 4), in addition to comparing the present data with the calculations of Gianturco (at 7 eV), Jain and Thompson and Lengsfield and co-workers, we also show a comparison with what, to our knowledge, is the only other absolute determination of a low-energy differential elastic cross section in NH_3 by Ben Arfa and Tronc (1987). The agreement with this earlier measurement is excellent at all angles except for a few in the forward direction. As discussed earlier, Ben Arfa and Tronc placed their data on an absolute scale by employing the relative flow technique and using the absolute cross section of Srivastava *et al* (1976) for elastic scattering from N_2 as a standard. The cross section of Srivastava *et al* has been superseded by that of Trajmar *et al* (1983) with the major difference being that this latter measurement is somewhat more peaked in the forward direction than that of Srivastava and co-workers. Hence, the disagreement between the present measurement and Ben Arfa and Tronc at the forward angles may simply be an artefact of the N_2 cross section that they employed in making their measurements absolute. Once again the best overall agreement is found with the complex Kohn calculation although it overestimates the

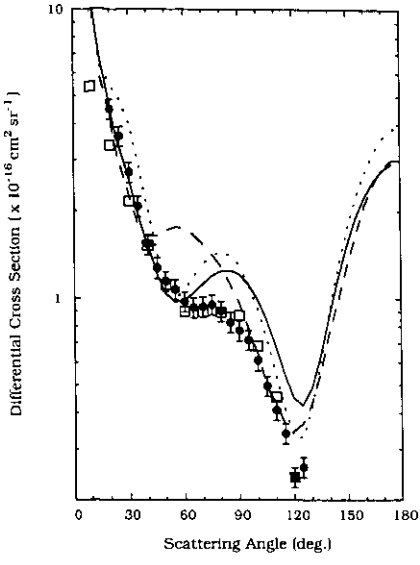

Figure 4. Differential cross section for elastic electron scattering from NH_3 at 7.5 eV: present data, ●; Ben Arfa and Tronc, □; Gianturco at 7 eV, – – –; Jain and Thompson, - - -; and Lengsfield *et al*, ——.

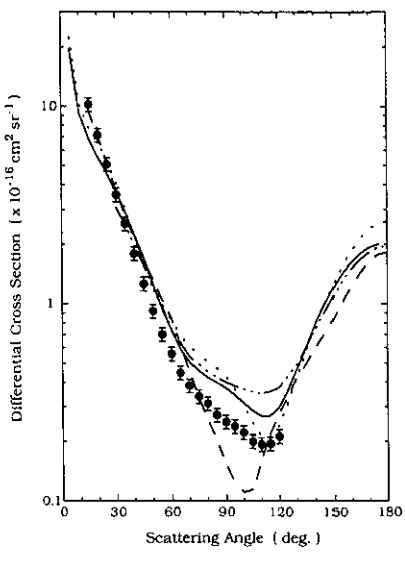

Figure 5. Differential cross section for elastic electron scattering from NH_3 at 20 eV: present data, ●; Gianturco, – – –; Jain and Thompson, - - -; Pritchard *et al*, — · · · —; and Lengsfield *et al*, ——.

cross section beyond about 60°. There is also a fair level of agreement with the calculations of Gianturco and Jain and Thompson at angles less than 60°.

Finally, we compare our 20 eV elastic differential cross section data (figure 5) with the calculations of Gianturco, Jain and Thompson and Lengsfield and co-workers. We would again characterize the overall level of agreement between the present data and these theories as being fair and, in particular, we highlight the excellent agreement at forward angles ($\theta \leqslant 35°$) between ourselves and Gianturco. We also note that, even though there are differences in the absolute magnitude of the cross sections of Pritchard *et al* and Lengsfield *et al* and the measured data, the shape of both calculated cross sections at middle angles is very similar to our own.

In the energy region between about 3 and 10 eV, electron–NH_3 scattering is strongly influenced by the formation of temporary negative ion resonances. Ben Arfa and Tronc (1987) demonstrated how both elastic scattering and vibrational excitation are affected by a broad d-wave shape resonance centred at around 7.5 eV. Although we do not show all of our elastic differential cross section data in the figures, the general trend at both low (2 eV) and high ($\geqslant$ 15 eV) energies is for a forward peaked, but smoothly varying, cross section. However, at 7.5 eV the effect of the d-wave resonance in the elastic differential cross section can be clearly seen (figure 4), and it can also be seen in the 5 eV data (figure 3). Also, in the energy region between 5 and 6 eV, a series of sharp, overlapping structures attributed to the vibrational levels (ν_2 inversion mode) of a NH_3^- $^2A_2''$ Feshbach resonance has been observed in both electron transmission experiments (Stricklett and Burrow 1986) and in the negative ion (NH_3^-, NH_2^- and H^-) yields produced by (dissociative) attachment of low-energy electrons (Stricklett and Burrow 1986, Tronc *et al* 1988). Our own studies of the energy dependence of the elastic cross section at an angle of 20° indicate the

presence of a strong (about 10% of the non-resonant cross section) resonance peak at 5.59 ± 0.04 eV. This energy is in good agreement with that of the dominant structure observed by Stricklett and Burrow in the transmission experiments (5.61 ± 0.03).

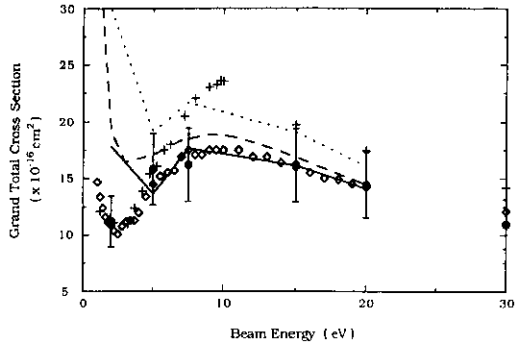

Figure 6. Grand total cross section for electron scattering by NH_3: present total elastic cross section, ●; Sueoka *et al*, ◇; Szmytkowski *et al*, +; Jain and Thompson, - - -; Gianturco, – – –; and Lengsfield *et al*, ——.

Table 2. Total elastic and elastic momentum transfer cross sections for ammonia (units of 10^{-16} cm^2).

E_0(eV)	Q_{ELAS}	Q_{MT}
2	11.22	2.24
5	15.83	8.75
7.5	16.21	9.15
15	16.16	10.03
20	14.43	6.40
30	10.97	5.73

In table 2 and figures 6 and 7, we present our total elastic and momentum transfer cross sections respectively. The intrinsic problems associated with the extrapolation procedure which is necessary to calculate these cross sections from the present differential measurements have been discussed previously by Brunger *et al* (1991). In the present case, however, they are potentially more serious than that considered by Brunger *et al* (1991) due to the strong forward peaking of the electron–polar molecule elastic cross section and the not insignificant strength at backward angles. The theoretical cross sections of Lengsfield *et al* have been chosen for the extrapolation procedure because they show the best overall agreement in both shape and absolute magnitude to the present results. It is difficult to accurately estimate the uncertainty associated with such an extrapolation procedure. At each energy we have calculated the contribution to the integrands in equations (2) and (3) from the angular regions that we cannot access in the present measurement. These can be as high as 50%, and, if we use the variation amongst the theoretical cross sections as an estimate of the range of possible extrapolations, we estimate the uncertainty in the integral cross sections arising from the extrapolation procedure to be 10–15%. This results in an overall estimated uncertainty in the integral cross sections of 20–25%.

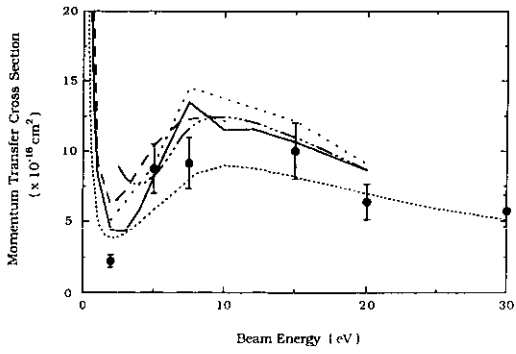

Figure 7. The momentum transfer cross section for elastic scattering by NH_3: present cross section, ●; Hayashi, ---; Gianturco, – – –; Pritchard *et al*, — · · · —; Jain and Thompson, - - -; and Lengsfield *et al*, ——.

Referring to figure 6, it is apparent that for $E_0 \leqslant 5$ eV the present derived total cross section is in good agreement with both the TOF and linear transmission determinations of the grand total cross section. Thereafter, our data and the calculations of Gianturco and Lengsfield and co-workers favours the TOF determination of Sueoka *et al* (1987) over that measured in a linear transmission experiment by Szmytkowski *et al* (1989). However, we must point out here that we cannot be more definitive about this comparison, as at energies $E_0 > 5$ eV there are many more open channels in addition to the elastic channel we have specifically investigated, which contribute to the grand total cross section. Consequently our total elastic cross section represents a lower bound on the grand total cross section and we would suggest that an independent TOF or linear transmission experiment be undertaken in an attempt to resolve the discrepancy between the data of Sueoka and co-workers and that of Szmytkowski and co-workers.

In figure 7 we compare the present elastic momentum transfer cross sections with the swarm determination of Pack *et al* (1962), as reported by Hayashi (1981), and again with the theoretical calculations of Pritchard and co-workers, Gianturco, Jain and Thompson and Lengsfield and co-workers. There is good agreement between the present momentum transfer cross section and that of Hayashi for energies $E_0 \geqslant 5$ eV. At 2 eV it is clear that the present data indicate a deeper minimum in the cross section than had been previously reported. With respect to the theories we find good agreement with them all at 5.0 eV but otherwise the present measurement is generally lower throughout the entire energy range.

4. Conclusions

The present data represent the first comprehensive set of elastic differential cross section measurements for electron scattering from NH_3. At energies above 2 eV, there is generally fair agreement between the present measurements and theory, particularly the recent Kohn variational calculation of Lengsfield *et al* (1991). The agreement between our data and the only previous experimental determination of an elastic differential cross section in NH_3, by Ben Arfa and Tronc (1987), is excellent. There is also good agreement between the total cross section derived from the present results and the TOF measurements of Sueoka *et al* (1987) at all energies studied. The

comparison of the integral cross sections with the various theoretical calculations reveals a reasonable level of agreement at energies above 2 eV, particularly, in the case of the total cross section, with Gianturco and Lengsfield and co-workers. This may be due, in part, to the use of the latter to extrapolate the present data to forward and backward angles and also to the fact that in those angular regions of the differential cross section which contribute most strongly to the total cross section, the agreement with the theory is quite good.

Acknowledgments

We dedicate this paper to Mr K B Roberts to mark the occasion of the 20th anniversary of his loyal service to the Ion Diffusion Unit and the Electron Physics Group. We also thank Professors Gianturco, Jain and Rescigno for providing us with their tabulated cross sections prior to publication and R W Crompton and M T Elford for their comments on the manuscript. As usual we also gratuitously acknowledge Dr M J 'Sinestra' Brennan, in this case for providing us all with entertainment.

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