

## Low Energy Electron Scattering From H<sub>2</sub>\*

M. J. Brunger, Stephen J. Buckman and D. S. Newman

Electron Physics Group, Research School of Physical Sciences,  
Australian National University, G.P.O. Box 4, Canberra, A.C.T. 2601, Australia.

### Abstract

Absolute differential and integrated total cross sections for elastic electron scattering and vibrational excitation of molecular hydrogen have been measured at an incident energy of 1.5 eV. The results are presented and discussed with particular reference to a long-standing impasse which has existed between and within experiment and theory for the near-threshold excitation of the first vibrational state of H<sub>2</sub>. The integral vibrational cross section is in good agreement ( $\pm 10\%$ ) with previous beam experiments and theory but is some 60% higher than the cross section derived from a swarm analysis.

### 1. Introduction

Although molecular hydrogen represents the simplest electron-molecule scattering system available, from both an experimental and theoretical point of view, there exists a long-standing discrepancy concerning the magnitude and shape of the near threshold (less than about 3 eV) vibrational excitation ( $v=0-1$ ) cross section (Morrison *et al.* 1987 and references therein). These differences exist not only between experiment and theory, but also within the two main experimental approaches which are used to obtain low energy electron scattering cross sections, the so-called swarm and beam methods.

In principle, both methods can provide low energy absolute cross sections for elastic scattering and excitation events in atoms and molecules, even though they constitute a completely different approach to the problem. On the one hand, the beam measurements are single collision experiments where direct information about the scattering cross section is obtained by studying discrete scattering events under conditions of zero electric field and low target gas pressure. On the other hand, the swarm technique involves the study of the collective motion of a large number of electrons in a high pressure gas under the influence of an electric field. Cross section information is then obtained by an iterative process in which comparisons of the experimental transport parameters are made with those derived using the Boltzmann equation. Each technique has its inherent strengths and weaknesses, most of which have been discussed in some detail previously (see for example Crompton 1983;

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Morrison *et al.* 1987). However, regardless of the relative attributes of the two techniques, the level of accuracy which is claimed for both is such that there ought to be reasonable agreement between their resultant cross sections. This is particularly the case when one considers that recent years have seen an increasing level of sophistication and hopefully, accuracy, being attained in beam experiments which has been matched by similarly sophisticated developments in the transport theory used to analyse the swarm results.

In some cases, such as the lighter rare gases helium and neon, the level of agreement has been excellent to good between both experiment and theory (e.g. Buckman and Lohmann 1986). In other cases, such as the heavier rare gases, nagging discrepancies remain between results from beam and swarm experiments, and between experiment and theory, for the 'grand' total scattering cross section (which includes elastic scattering and all energetically favourable excitations). Such targets, for which only elastic scattering is possible at low energies, should be well understood, but the level of disagreement (5–30%) is well outside expected experimental uncertainties. To some extent the level of discrepancy between the results of the beam (attenuation experiments) and swarm techniques in this case is hard to quantify, as each experiment results in different cross sections (total and momentum transfer respectively) which are only equivalent either at zero energy or if the angular scattering is isotropic. The derivation, for purposes of comparison, of total cross sections from momentum transfer and vice versa via modified effective range theory (MERT), has been discussed in detail by Buckman and Mitroy (1989).

However, by far the most significant discrepancy for a collision system of fundamental importance occurs for low energy vibrational excitation of  $\text{H}_2$ , where the discrepancy between the various experiments and theory is as large as 60%. In this case the discrepancy lies in the total cross section for ro-vibrational ( $v = 0-1$ ,  $\Delta j = 0, \pm 2$ ) excitation which can be derived *directly* from the swarm analysis. This cross section can be compared directly with that obtained by an appropriate beam measurement. The object of the present work was to provide a test measurement, using single collision techniques, of the absolute differential cross section for ro-vibrational excitation of  $\text{H}_2$  at 1.5 eV. From this cross section, and a corresponding measurement for vibrationally elastic scattering, the total ro-vibrational cross section and the total momentum transfer cross section can be obtained and compared with the swarm-derived cross sections.

In the next section we briefly summarise the current status of both experiment and theory for low energy electron-hydrogen scattering. Section 3 contains a description of the apparatus used for the present measurements and an outline of the experimental procedures and normalisation techniques employed. The results are presented and discussed in Section 4 and we provide some concluding remarks in Section 5.

## **2. Current Status of Experiment and Theory for Low-energy Electron-Hydrogen Scattering**

Considering the fundamental nature of the collision problem, there have been relatively few determinations of low energy electron scattering cross sections for molecular hydrogen in recent years, particularly for near-threshold

vibrational excitation. Differential elastic scattering measurements have been carried out by Ehrhardt *et al.* (1968), Linder and Schmidt (1971), Shyn and Sharp (1981), Nishimura *et al.* (1985), Furst *et al.* (1984) and Sohn (1986). The measurements of Linder and Schmidt and of Sohn also included studies of discrete rotational excitation in the ground vibrational state. Differential vibrational excitation cross sections have been measured by Ehrhardt *et al.*, Linder and Schmidt and Nishimura *et al.*, while the total vibrational cross section has been measured by Burrow and Schulz (1969). In addition there have been several measurements of the total collision cross section (Golden *et al.* 1966; Dalba *et al.* 1980; Ferch *et al.* 1980; Jones 1985; Subramanian and Kumar 1989). Swarm studies, from which the momentum transfer cross section for elastic scattering and total cross sections for rotational and ro-vibrational excitation have been derived, are reported in papers by Engelhardt and Phelps (1963), Phelps (1968), Crompton *et al.* (1969, 1970), Haddad and Crompton (1980), Petrovic and Crompton (1987), Morrison *et al.* (1987) and England *et al.* (1988). There has been little corresponding theory, and the major work has been limited to a few groups (eg. Henry and Lane 1969; Gibson and Morrison 1984; Morrison *et al.* 1984, 1987).

In summarising the current level of agreement between the various experiments and theories we will restrict ourselves to the discussion of reasonably recent results where applicable. The early results have been thoroughly reviewed by Crompton *et al.* (1970) and Linder and Schmidt (1971), while more recently there are the reports of Morrison *et al.* (1987) and McConkey *et al.* (1988). Surprisingly the latter, in reviewing what is known about near-threshold ro-vibrational excitation, do not make mention of the swarm work or of the differences between beam and swarm results.

### 2.1 Elastic Scattering and Rotational Excitation

In general there is good agreement between the various experiments and the theoretical calculations of Morrison *et al.* for both rotationally elastic scattering and for discrete rotational excitation. This applies to both differential cross sections (Linder and Schmidt 1971; Sohn 1986; Jung *et al.* 1987) and total cross sections for both  $j=0-2$  and  $j=1-3$  excitations. In particular, for the total rotational cross sections at energies below 2 eV there is excellent agreement between the beam (Linder and Schmidt), swarm (England *et al.*) and theoretical results.

### 2.2 Total Scattering

Results from beam experiments for the grand total cross section are also in good agreement with the close coupling calculations at energies from 0.1 to 10 eV. In particular the results from the time-of-flight technique (Ferch *et al.* 1980; Jones 1985), which can be obtained with high accuracy, are in excellent agreement with the theory. There is also excellent agreement between the theory and swarm experiment for the total momentum transfer cross section at energies up to 2 eV (see Morrison *et al.* 1987).

### 2.3 Vibrational Excitation

As outlined in the previous section the major, unresolved, controversy for electron-molecular hydrogen scattering occurs over the magnitude and shape of the near threshold ( $<3$  eV) excitation cross section for the first vibrational state. It arises from the direct comparison of the beam measurements of Ehrhardt *et al.* and of Linder and Schmidt with the swarm derived cross section of Crompton *et al.* (1969, 1970). At energies below about 2 eV, there are marked differences between the respective cross sections. For example at 1.5 eV, the beam measurements are 60 and 90% higher respectively than the swarm cross section. Similar differences exist between the data of Ehrhardt *et al.* all the way down to the threshold at 0.52 eV.

In making this comparison it should be stressed that the major thrust of both of the above beam experiments, particularly that of Ehrhardt *et al.*, was to study the effect of negative ion resonances on the excitation cross sections rather than to obtain accurate absolute scattering cross sections. Nonetheless both of these experiments *do* provide an absolute total ro-vibrational cross section which can be *directly* compared with the swarm result and as such it is perhaps worth while to comment briefly on the techniques which were employed in them.

The cross section measurements of Ehrhardt *et al.* were carried out with a conventional crossed electron-molecular beam spectrometer with an energy resolution sufficient to resolve vibrational but not rotational excitations. Their differential vibrational excitation cross sections were placed on an absolute scale by measuring the ratio of the elastic to inelastic scattering intensities and then normalising their integrated elastic differential cross sections to the 'known' total cross section. In addition to the differential measurements at a number of energies, they measured excitation functions over an energy range from threshold to about 5 eV at a few selected angles. In placing the energy dependence of the  $v=0-1$  vibrational cross section on an absolute scale they relied on their observation that their excitation functions were independent of scattering angle (which is equivalent to the angular distributions being independent of energy), and used the measured energy dependence at  $20^\circ$  as representative of the total cross section. They gave no estimate of the absolute uncertainty of this cross section.

Linder and Schmidt used the same apparatus for their measurements but with much higher energy resolution (30 meV) to enable individual rotational and ro-vibrational processes to be resolved in the energy range 0.3–15 eV and angular range  $20^\circ$ – $120^\circ$ . Their results were placed on an absolute scale by extrapolating all of their differential measurements at each energy to  $0^\circ$  and  $180^\circ$  and integrating to obtain relative total cross sections whose sum was then normalised to the grand total cross section measurement of Golden *et al.* (1966). They estimated the errors in their differential and total cross sections to vary between 10 and 20%.

Following the earliest swarm studies of Engelhardt and Phelps (1963) and Phelps (1968), Crompton and co-workers have performed a number of measurements of electron transport coefficients for electron swarms in  $H_2$  to derive low energy electron scattering cross sections. Their initial studies, carried out in both normal and para-hydrogen to assess the effect of initial

rotational state population, have been complemented and extended in recent years by further measurements of transport parameters (drift velocity and transverse diffusion coefficient) in hydrogen-rare gas mixtures (Haddad and Crompton 1980; Petrovic and Crompton 1987; England *et al.* 1988). In all cases the subsequent measurements have confirmed the earlier data and there are only small differences between the latest swarm-derived cross section set for H<sub>2</sub> (England *et al.* 1988) and the original work of Crompton *et al.* (1969, 1970).

There has been an extensive investment of effort in theoretical calculations by Morrison and co-workers over the past decade for the vibrational excitation cross sections as well as the cross sections for rotational and elastic scattering. A detailed summary of these calculations has been given by Morrison *et al.* (1987). These calculations predict a value of the total vibrational excitation cross section at 1.5 eV which is in good agreement with both the beam measurements of Ehrhardt *et al.* and of Linder and Schmidt but not with the swarm cross section of England *et al.* Closer to threshold (<0.75 eV), the theoretical and swarm cross sections begin to converge (the difference is reduced to about 25% at 0.7 eV), although both cross sections are considerably lower than that of Ehrhardt *et al.*

In light of this longstanding, serious discrepancy, the resolution of which has important ramifications for either the theory and the beam experiments or for the swarm experiments and/or their subsequent analysis, the present measurements were intended to present data from another beam experiment which was expressly set up to provide absolute scattering cross sections for low energy electron-molecule collisions.

### 3. Experimental Apparatus and Techniques

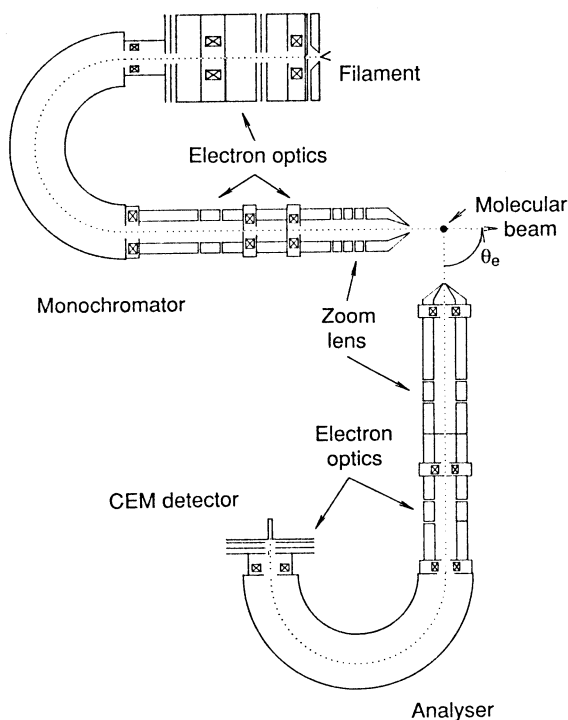
#### 3.1 Apparatus

The present experiments were carried out on a recently constructed, high resolution crossed electron-molecular beam apparatus. This apparatus will be described in full detail in a forthcoming paper (Brunger *et al.* 1990) but, given the degree of controversy surrounding the level of agreement between existing results, it is appropriate here to give a brief description of both the apparatus and, in particular, the experimental techniques employed.

The incident electron beam was produced by a conventional thermionic source, and transported and energy resolved by a combination of electrostatic electron optics and a hemispherical electrostatic energy analyser (Fig. 1). The optical elements, which consist of both aperture and tube lenses, were designed specifically with the production of low energy beams (0.1–5 eV) in mind. At these energies, electron beams with an energy spread of less than 30 meV are readily achievable but, for the measurements presented here, where resolution of rotational structure was not required, the energy resolution is typically 80–100 meV. A typical energy loss spectrum for H<sub>2</sub> from the present apparatus is shown in Fig 2. For the present study at 1.5 eV, beam currents of 0.5–5 nA were used, and the diameter of the electron beam in the interaction region was typically less than 1 mm.

Scattered electrons were transported and energy analysed by a similar combination of electrostatic electron optics and hemispherical energy analyser,

and detected by a single channel electron multiplier (Mullard B310). A channeltron was favoured for these measurements over a channelplate-position sensitive detector combination in order to avoid the additional problems of absolute determination of variations of spatial efficiency with the latter. Once again the electron lenses were specifically designed to transport low energy (0–5 eV) scattered electrons with high efficiency. The operation of the zoom lens immediately adjacent to the target region will be discussed in detail in a later section. The analyser and associated optics were mounted on a rotatable turntable whose axis was aligned with that of the molecular beam. This provides access to scattering angles between  $-20^\circ$  and  $+135^\circ$ .

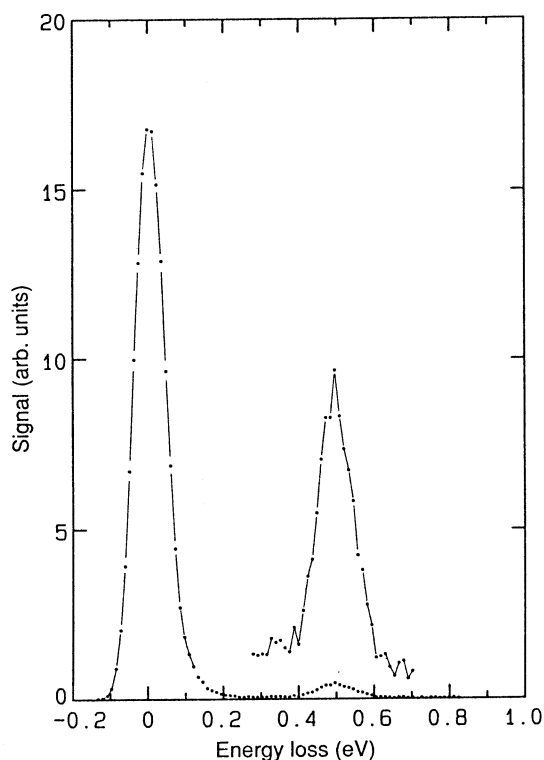


**Fig. 1.** Schematic diagram of the electron spectrometer.

All components of the electron spectrometer were constructed from molybdenum if exposed to the electron beam, or grade 310 non-magnetic stainless steel. The spectrometer was housed in a vacuum chamber, pumped by a 550 litre/sec turbomolecular pump, to a base pressure  $5 \times 10^{-9}$  mbar. The chamber was located at the centre of a set of three mutually orthogonal Helmholtz coil pairs which, when combined with magnetic shielding inside the vacuum manifold, reduced the ambient magnetic field to less than  $10^{-7}$  T.

The molecular beam was formed by effusive flow through a multichannel capillary array which has an active diameter of 1.0 mm and contains 280 capillaries which are 40  $\mu\text{m}$  in diameter and 1.0 mm long. The array source was constructed by fixing a commercially available glass array (Galileo Electro-Optics Corp.) to a glass tube. The array and tube were gold coated to avoid charging

by the electron beam. The driving pressure of gas behind the capillary array, as measured with a calibrated capacitance manometer (MKS Baratron), was typically in the range 0.5–2.5 Torr (1 Torr  $\equiv$  133 Pa). The justification for this range of operating pressures will be discussed in the next section. The interaction volume, formed by the intersection of the electron and molecular beams, was about 1.5 mm above the capillary array.



**Fig. 2.** Energy loss spectrum for molecular hydrogen at an incident electron energy of 1.5 eV and a scattering angle of 60°. The upper spectrum of the  $\nu=0-1$  feature has been enhanced by a factor of 20.

Whilst the technique we employ for assigning absolute values to our measured angular distributions does not require a knowledge of the absolute sizes of the electron and molecular beams or their overlap, it was critically important that, in order to avoid the application of angle-dependent corrections to the measured scattered intensity, the analyser viewed *all* of the interaction volume at *all* scattering angles. By careful alignment of the apparatus, and selection of appropriate sizes and positions for entrance apertures to the analyser, we were able to ensure that this condition was met. Experimental measurements to verify this are presented in the next section. The angular resolution of the apparatus was estimated to be 1.5°.

The experiment can be run either manually or under computer control. The computer controls the position of the analyser via a stepper motor, the movement of a retractable Faraday cup for the measurement of electron beam

current, the scanning of the analyser energy loss voltage, the measurement of the source driving pressure as well as the chamber pressure, and the transfer of data from a multichannel analyser used for data acquisition.

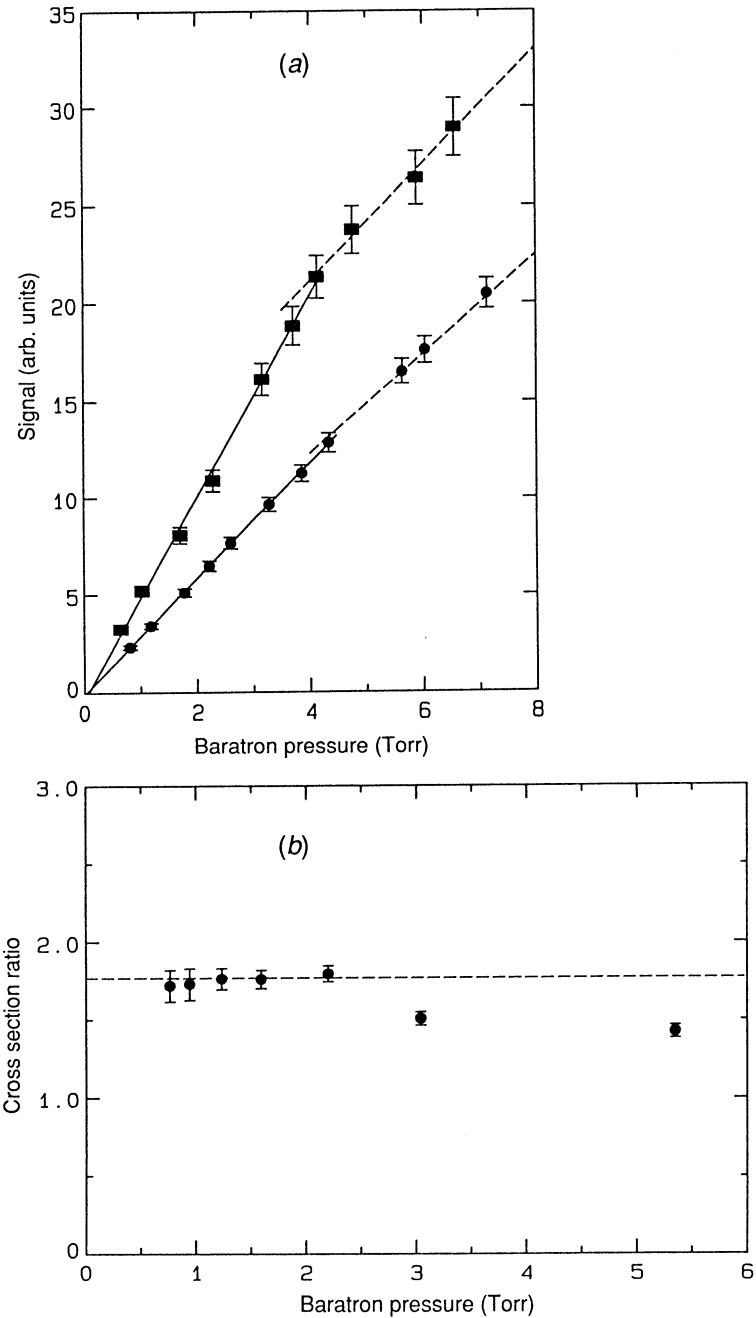
### 3.2 *Experimental Techniques and Normalisation Procedure*

For both elastic scattering and excitation of the  $\nu=0-1$  vibrational band at least four relative angular distributions were carried out under a variety of experimental conditions, although always with experimental parameters within certain bounds which were established by a series of independent measurements. Background contributions at each scattering angle were determined by measurements of the scattered intensities when the gas entered the vacuum chamber at an equal flow rate via a second capillary, located away from the scattering volume. These angular distributions were combined, with appropriate weighting for their statistical uncertainty, to give final relative angular distributions for these two processes. The final relative distribution for elastic scattering was converted to an absolute elastic differential cross section by comparison with a 'standard' cross section for elastic electron scattering from helium (Nesbet 1978). This comparison involved the measurement of the ratio of elastic scattering from helium to that from  $H_2$  and the use of the relative flow technique (Srivastava *et al.* 1975) which is described in Subsection 3.2.3. Given the absolute elastic differential cross section, previously determined ratios of the elastic to the  $\nu=0-1$  intensities could then be utilised to convert the relative  $\nu=0-1$  angular distribution to an absolute differential cross section. However, a number of experimental conditions need to be established and verified for this procedure to be valid.

**3.2.1 Molecular Beam Conditions.** The relative flow normalisation technique requires the measurement of scattered intensities under identical target conditions for both the gas under study and the reference gas. For identical target conditions to apply, the magnitude of the driving pressure behind the capillary source must be such that collisions of gas molecules with other gas molecules do not adversely effect the spatial distribution of the molecular beam. This is strictly only true when the mean free path for the molecules,  $\lambda$ , is much larger than the largest dimension of the capillary tube, or alternatively when the Knudsen number based on the tube length  $L$  ( $K_L = \lambda/L$ ) is less than one. In our case this would correspond to a driving pressure for  $H_2$  of less than 0.3 Torr, a pressure which makes statistically accurate scattering experiments very difficult to perform. However, it has been demonstrated (Olander and Kruger 1970) that the spatial characteristics of a molecular beam produced by a capillary may still be well understood at pressures up to an order of magnitude higher than this, i.e.  $K_L < 10$ .

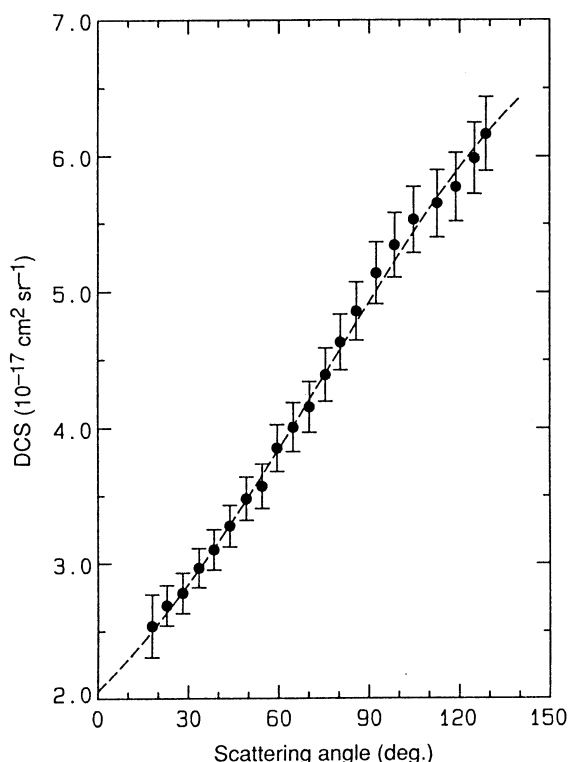
We have investigated the effects of driving pressure for our experimental arrangement by measuring (i) the scattered signal as a function of driving pressure for both  $H_2$  and He at a range of scattering angles, and (ii) the variation of the ratio of the elastically scattered signal at  $90^\circ$  to that at  $20^\circ$  as a function of source driving pressure. These results are shown in Figs 3a and 3b respectively. In Fig. 3a it is evident that there is an observable change of slope in both the  $H_2$  and He curves at a driving pressure of about 4 Torr. This corresponds to the pressure region where the mean free path for collisions





**Fig. 3.** (a) Elastically scattered electron signal at 1.5 eV as a function of capillary driving pressure for He (circles) and H<sub>2</sub> (squares). (b) The ratio of elastically scattered electron signal at 1.5 eV for a scattering angle of 90° to that for 20° as a function of capillary driving pressure. The dashed line represents the theoretical prediction (Nesbet 1978) for this ratio.

becomes smaller than the capillary diameter, and is an indication that the target number density and/or the spatial distribution of the molecular beam is being affected by gas collisions. Fig. 3*b*, on the other hand, provides a more direct indication of changes in the spatial distribution of the molecular beam. This results from the fact that at forward scattering angles, the size of the collision volume as seen by the detector is determined by the electron beam, which is the smaller of the two beams. However at 90°, the collision volume is determined by the size of the molecular beam and, if this beam expands outside of the viewing cone of the scattered electron analyser as a function of driving pressure, then we may expect to see a decrease in the ratio of the measured signal at 90° to that at a forward angle like 20°. This decrease is indeed observed at a driving pressure in excess of about 2.5 Torr.



**Fig. 4.** Absolute differential cross section for elastic electron scattering from He (in units of  $10^{-17} \text{ cm}^2 \text{ sr}^{-1}$ ) at 1.5 eV: circles, present results; dashed curve, Nesbet (1978).

Having established from the above that data collection should be restricted to driving pressures less than 2.5 Torr (all of the  $\text{H}_2$  measurements described here were carried out at driving pressures of less than 1.5 Torr), we further need to establish that the scattering geometry is well defined at these pressures, i.e. the analyser views the entire scattering volume at all scattering angles. This has been achieved by measuring elastic angular distributions for helium and comparing the results with the ab-initio, fully variational calculation of Nesbet (1978) which, we believe, represents the most accurate determination of

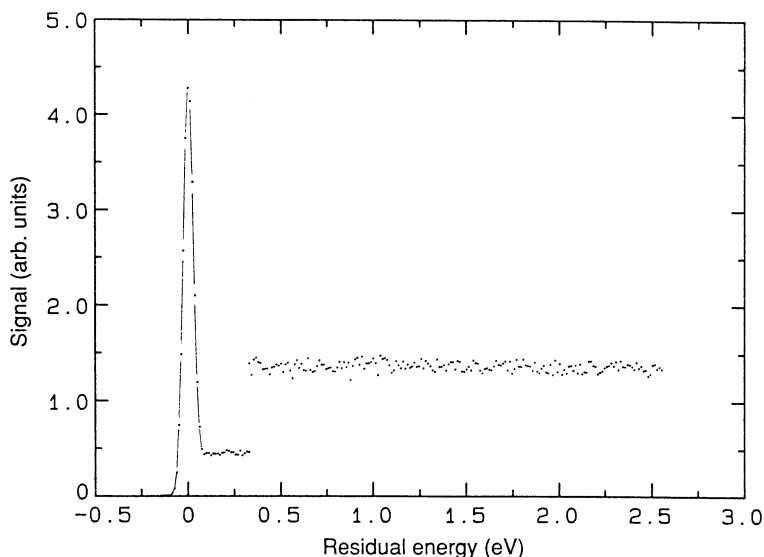
the cross section for electron helium elastic scattering below the first inelastic threshold. These results are shown in Fig. 4. The experimental data have been placed on an absolute scale by a phaseshift analysis (Allen 1986; Allen and McCarthy 1987) and it can be seen that the agreement with theory, both in the shape and the absolute magnitude of the differential cross section, is excellent. This implies that there is no need for the application of any angular dependent scaling of the cross sections measured with the present apparatus.

**3.2.2 Analyser Transmission.** The application of the normalisation technique discussed in the next section to an inelastic scattering cross section requires a knowledge of the ratio of inelastic to elastic scattering. This in turn requires a measurement of the efficiency of the scattered electron analyser and optics as a function of energy. For the present measurements, allowing for the energy resolution of the apparatus, the scattered electrons of interest have energies between about 1.55 and 0.95 eV and their optimal transmission through the analyser requires significantly different voltages on the elements of the analyser zoom lens (see Fig. 1). The relative efficiency over this energy range has, in our case, been optimised by changing the voltages on these elements synchronously with the energy loss voltage.

The relative transmission of the analyser and optics at these energies can, in principle, be measured by investigating the yield of electrons (ejected and scattered) from helium following near-threshold ionisation (Pichou *et al.* 1978). As a consequence of the Wannier theory for near-threshold ionisation (Wannier 1953; Rau 1971) the yield of detected electrons is expected to be both isotropic and independent of energy for values of the excess energy (i.e. the difference between the incident electron energy and the ionisation potential) up to some value  $E_{\max}$  above the ionisation threshold. Thus, in a measurement of the yield of scattered and ejected electrons resulting from electron impact ionisation at an appropriately low energy, one may expect to observe a flat spectrum in all situations where the excess energy is less than  $E_{\max}$ . Experimental verification of this, in particular the value of  $E_{\max}$ , is still uncertain and, to some extent, controversial. The range of validity of the Wannier theory has been tested in both differential and integral measurements of the ionisation cross section for excess energies up to 8 eV (see for example Pichou *et al.* 1978; Schubert *et al.* 1981; Read 1984; Hammond *et al.* 1985). Some measurements (Schubert *et al.*; Sohn *et al.* 1983) indicate that even when the excess energy is as high as 6 eV the Wannier theory appears to hold, whilst others (Hammond *et al.*) show that there are departures from this theory of the order of 5% at excess energies as low as 0.075 eV. A recent study (Jones *et al.* 1989) indicates that the Wannier model does not adequately describe (e,2e) experiments at an excess energy of 4 eV, although the extent of the disagreement and the ramifications for the present requirements are not clear.

One can only conclude that the present situation regarding the range of validity of the Wannier model is not well established, and that the extent to which it is uncertain at any particular energy is also not well known. Its use therefore as an indicator of the performance of a spectrometer is subject to a substantial uncertainty. Nonetheless we have carried out such a measurement for an incident electron energy of 30.08 eV (i.e. 5.5 eV above the ionization threshold) and an analyser energy loss range of 24 to 30.5 eV with the analyser

and electron optical voltages set for optimum transmission at 0.98 and 1.5 eV as outlined above. The spectrum of ejected electrons, expressed in terms of residual energy, is shown in Fig. 5. In the area of interest, indeed over most of the residual energy range, it is essentially flat. In particular, the intensities of electrons of 0.98 and 1.5 eV are equal to within less than 1%. The sharp peak at zero residual energy is an electron optical artefact indicating the high efficiency with which near-zero energy electrons can be detected. We estimate that the uncertainty in this relative efficiency determination may be as high as 10%. Measurements to investigate this problem further are underway and will be reported in detail in a future paper (Brunger *et al.* 1990).



**Fig. 5.** Yield of near threshold electrons from helium, as a function of residual energy, following electron impact ionisation at 30.08 eV. The spectrum above 0.3 eV residual energy has been enhanced by a factor of 3.0.

**3.2.3 Normalisation—The Relative Flow Technique.** The angular distribution for vibrationally elastic scattering (henceforth referred to as ‘elastic scattering’) has been placed on an absolute scale by use of the relative flow technique (Srivastava *et al.* 1975; Khakoo and Trajmar 1986; Nickel *et al.* 1989). This method relates the scattering intensity for the gas of interest to that of a reference gas, in our case helium. The differential cross section for elastic scattering from H<sub>2</sub> was obtained from the relationship

$$\frac{d\sigma(\theta)_{\text{H}_2}}{d\Omega} = \frac{d\sigma(\theta)_{\text{He}}}{d\Omega} \frac{\dot{N}(\theta)_{\text{H}_2}}{\dot{N}(\theta)_{\text{He}}} \left( \frac{m_{\text{He}}}{m_{\text{H}_2}} \right)^{\frac{1}{2}} \frac{F_{\text{He}}}{F_{\text{H}_2}}, \quad (1)$$

where  $\dot{N}$  is the rate of elastically scattered electrons for each gas,  $m$  the molecular weight and  $F$  the flow rate of each gas through the capillary.

The measurements of the elastically scattered signal rates were carried out under identical electron optical conditions for each gas and at driving pressures of less than 1.5 Torr for both gases. In addition the driving pressures were

carefully adjusted for each measurement such that the mean free path for each gas at the entrance to the capillary array was identical. Many such measurements were carried out for a range of driving pressures, electron optical settings and scattering angles. Although the measurement of the relative flow rates is a straightforward procedure with our gas handling system, it is nevertheless a time consuming exercise. To avoid such measurements on a regular basis during data collection the relative flow rates have been calibrated against the capillary driving pressure for each gas, which was readily measured by the capacitance manometer. A detailed description of this procedure will be given in a forthcoming paper (Brunger *et al.* 1990).

From these measurements the absolute differential elastic cross section for  $H_2$  can be obtained from equation (1) by using the theoretical value of the differential elastic scattering cross section for helium at 1.5 eV from Nesbet's (1978) calculation.

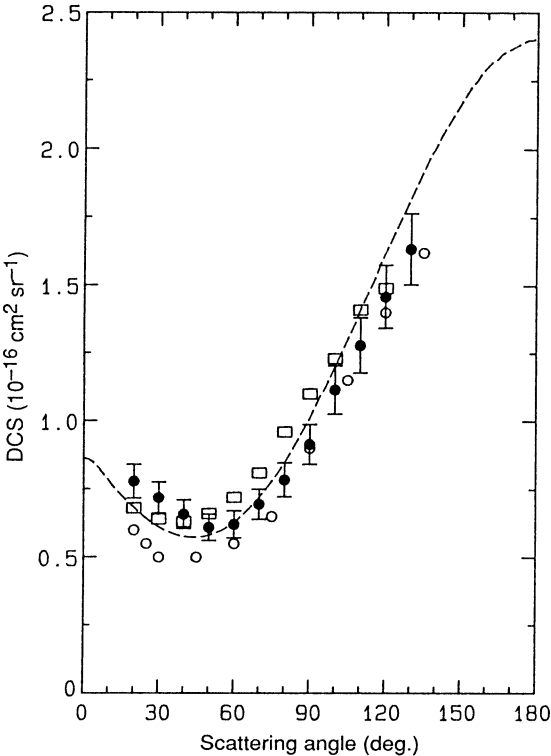
The angular distribution for vibrational excitation was placed on an absolute scale by measuring, at various scattering angles, the ratio of the scattered intensity for  $v=0-1$  to that for elastic scattering. Provided the precautions outlined in Section 3.2 are met, this ratio is equivalent to the ratio for the differential cross sections for these processes, at those scattering angles. Thus, from the previously determined elastic differential cross section, the differential cross section for ro-vibrational excitation could also be placed on an absolute scale. These ratios were measured at a number of forward, middle and backward scattering angles. The absolute differential cross sections obtained from each set of measurements were consistent.

**3.2.4 Experimental Errors.** As it was our intention that the present results should be used as a test measurement of the vibrational excitation cross section for  $H_2$ , we have paid particular attention to the identification and quantification of all possible sources of experimental error. For the elastic scattering measurements, the uncertainty in the differential cross section is believed to be 8% at all scattering angles. This figure is comprised of 2% arising from the normalisation to the helium standard cross section, 5% from the application of the relative flow technique, counting statistics (<2%) in the angular distribution, the statistical uncertainty in the ratio of  $H_2/He$  cross sections (4%) and the uncertainty and variation in gas pressure (1%) and electron beam current (2%) during data collection. The differential measurements for vibrational excitation are normalised via the elastic results and thus inherit the 8% uncertainty plus a further uncertainty due to counting statistics (<3%), similar uncertainties in pressure and current measurements to those above, an uncertainty of 5% in the determination of the ratio of elastic to vibrational excitation intensity and an uncertainty of 10% in the estimation of the relative analyser transmission between 1.55 and 0.95 eV. This results in an overall uncertainty of 14% in these data. The corresponding integral cross sections for both elastic scattering and vibrational excitation are both estimated to have an uncertainty of less than 20%.

#### 4. Results and Discussion

The absolute differential cross sections for elastic scattering from  $H_2$  at 1.5 eV are shown in Fig. 6 and Table 1. The present results, which cover the

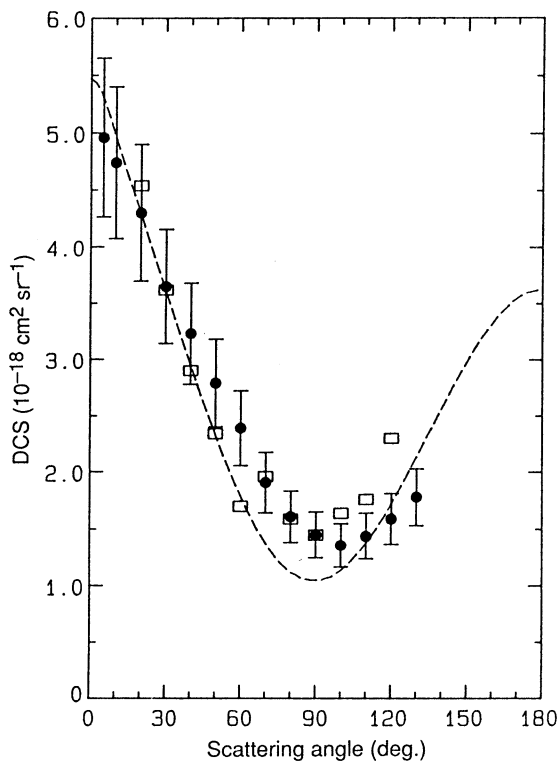
angular range 20–130°, are compared and contrasted with those from theory (Snitchler *et al.* 1990) and experiment (Linder and Schmidt 1971; Sohn 1986). In general the agreement between the theory and present experiment is very good, as is that with the data of Linder and Schmidt. The theoretical curve lies outside the present experimental error bars at only a few scattering angles in the forward and backward directions. The cross section of Sohn *et al.* is for rotationally elastic scattering ( $\Delta j = 0$ ) and can therefore be expected to be slightly smaller in magnitude than both of the other measurements and theory.



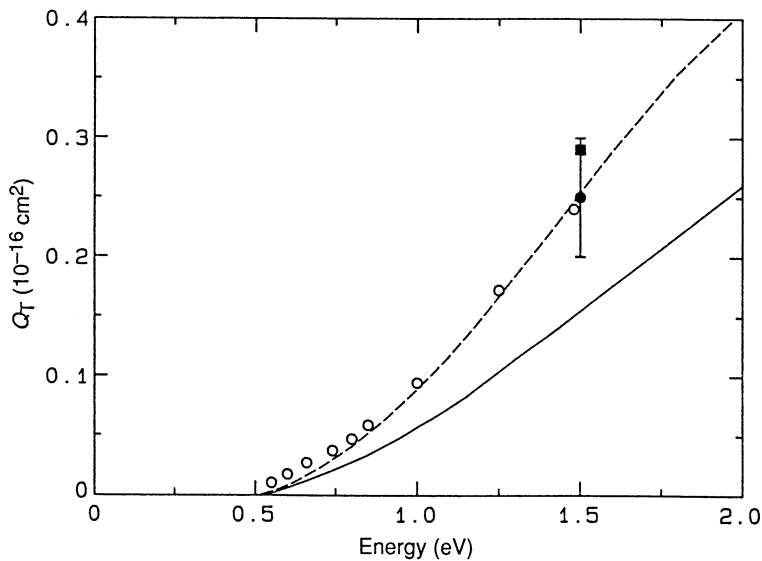
**Fig. 6.** Absolute differential cross section for vibrationally elastic scattering from H<sub>2</sub> (in units of 10<sup>−16</sup> cm<sup>2</sup> sr<sup>−1</sup>) at 1.5 eV: solid circles, present results; open squares, Linder and Schmidt (1971); open circles, Sohn (1986); and dashed curve, Snitchler *et al.* (1990).

**Table 1.** Absolute differential cross sections for vibrationally elastic scattering (10<sup>−16</sup> cm<sup>2</sup> sr<sup>−1</sup>) and ro-vibrational ( $\nu = 0-1$ ) excitation (10<sup>−18</sup> cm<sup>2</sup> sr<sup>−1</sup>) of H<sub>2</sub>  
Estimated uncertainties are  $\pm 8\%$  and  $\pm 14\%$  respectively

Angle (deg.)	Elastic	$\nu = 0-1$	Angle (deg.)	Elastic	$\nu = 0-1$
5	—	4.96	70	0.70	1.91
10	—	4.74	80	0.79	1.61
20	0.78	4.30	90	0.92	1.45
30	0.72	3.65	100	1.12	1.36
40	0.66	3.23	110	1.28	1.44
50	0.61	2.79	120	1.46	1.59
60	0.62	2.39	130	1.63	1.78



**Fig. 7.** Absolute differential cross section for ro-vibrational excitation of H<sub>2</sub> ( $\nu=0-1$ ) (in units of  $10^{-18} \text{ cm}^2 \text{ sr}^{-1}$ ) at 1.5 eV: solid circles, present results; open squares, Linder and Schmidt (1971); and dashed curve, Snitchler *et al.* (1990).



**Fig. 8.** Total cross section for ro-vibrational excitation ( $\nu=0-1$ ) of H<sub>2</sub> (in units of  $10^{-16} \text{ cm}^2$ ) at 1.5 eV: solid circle, present result; solid square, Linder and Schmidt (1971); open circles, Ehrhardt *et al.* (1968); dashed curve, Snitchler *et al.* (1990); and solid curve, England *et al.* (1988).

For the  $\nu = 0-1$  vibrational excitation, the absolute differential cross sections at 1.5 eV and over the angular range of 5–130° are also given in Table 1 and compared with other experimental values in Fig. 7. The agreement with theory is reasonably good, there being overlap within experimental error at all angles except the region from 50 to 90°. There is also good agreement with the experimental data of Linder and Schmidt.

**Table 2. Integral cross sections ( $10^{-16} \text{ cm}^2$ ) for grand total scattering, total ro-vibrational ( $\nu = 0-1$ ) excitation of  $\text{H}_2$  and total momentum transfer**

The estimated uncertainty in each of the present determinations is  $\pm 20\%$

	Grand total	Total ro-vibrational	Total momentum transfer
<b>Theory</b>			
Morrison <i>et al.</i>	15.0	0.25	18.73
<b>Swarm</b>			
England <i>et al.</i>	—	0.15	18.14
<b>Beam</b>			
Jones	14.6	—	—
Ehrhardt <i>et al.</i>	—	0.25	—
Linder & Schmidt	—	0.29	—
<b>Present</b>	14.1	0.25	17.2

As the main thrust of this work was to make a comparison with cross sections obtained from the swarm analysis, we have integrated both of these differential cross sections to obtain the total vibrational excitation cross section for  $\nu = 0-1$  and the total momentum transfer cross section. We have also calculated the grand total cross section for comparison with TOF experiments. These integrations require an extrapolation of the measured differential cross sections to 0° and 180°. We have used the shape of the theoretical cross sections as a guide for this extrapolation. The errors involved in this extension of the experimental data are difficult to quantify accurately. However, in the case of the  $\nu = 0-1$  total cross section, less than 15% of the integrand arises from those sections of the differential cross section which we do not measure but which we derive from the extrapolation procedure. The use of several 'reasonable' alternative extrapolations for this cross section indicate that the error arising from the extrapolation is less than 5%. For the grand total and total momentum transfer cross section this error is slightly larger ( $\sim 8\%$ ), as the elastic differential cross section peaks in the backward direction resulting in a larger contribution to the integrand from that section of the differential cross section which we do not measure. These integral cross sections are summarised in Table 2 and the  $\nu = 0-1$  total cross section is shown in Fig. 8 where it can be compared with the earlier beam data (Ehrhardt *et al.* 1968; Linder and Schmidt 1971), the swarm result (England *et al.* 1988) and the theoretical result (Snitchler *et al.* 1990). The present value clearly confirms the earlier beam measurements and the theory at this energy. Although it does not arise directly from the analysis, the uncertainty of the swarm-derived total vibrational cross section at 1.5 eV is thought to be about 10% (Crompton 1990). Thus the swarm cross section and the present result are clearly incompatible.



Comparison is also made in Table 2 with a selection of other integral cross sections from beam and swarm experiments and theory. The total momentum transfer cross section, which in our case will not include small contributions from higher vibrational modes, is in agreement, within error bars, with both the swarm experiment and theory. The grand total cross section is also in good agreement with theory and with a recent TOF measurement of Jones (1985).

## 5. Conclusions

The present measurement of the vibrationally elastic and  $v = 0-1$  ro-vibrational excitation cross sections at 1.5 eV provide strong support for the previous beam measurements and recent theoretical calculations for these processes. Preliminary results at other energies, above 1.5 eV (Brunger *et al.* 1990), further support the energy dependence exhibited by these previous experiments and the theory. The present results maintain and indeed, heighten, the impasse between, on the one hand the beam measurements and theory and on the other, the swarm determinations of vibrational excitation for  $H_2$ .

In their recent detailed analysis of both experiment and theory, Morrison *et al.* concluded that further, careful investigations using beam, swarm and theoretical techniques were warranted to resolve this impasse. We believe the present results are the first step towards fulfilling the first of these requirements. As we have already discussed there have been several recent cross checks of the swarm experiment which have been entirely consistent with the original work. It is neither appropriate, nor are we qualified, to comment on either the swarm experiments or their analysis here. Indeed it is perhaps appropriate to reserve any conclusions on this situation until measurements covering the full range from threshold (0.52 eV) to 3 eV are complete, as the swarm analysis is most sensitive to the disparity which exists between beam and swarm experiments at threshold. Such measurements are presently underway in this laboratory.

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## References

- Allen, L. J. (1986). *Phys. Rev. A* **34**, 2706.
- Allen, L. J., and McCarthy, I. E. (1987). *Phys. Rev. A* **36**, 2570.
- Brunger, M. J., Buckman, S. J., Newman, D. S., and Alle, D. T. (1990). To be published.
- Buckman, S. J., and Lohmann, B. (1986). *J. Phys. B* **19**, 2547.
- Buckman, S. J., and Mitroy, J. (1989). *J. Phys. B* **22**, 1365.
- Burrow, P. D., and Schulz, G. J. (1969). *Phys. Rev.* **187**, 97.
- Crompton, R. W. (1983). Proc. XVI Int. Conf. on Phenomena in Ionised Gases, Dusseldorf (Eds W. Botticher *et al.*), p. 58 (Univ. of Dusseldorf).
- Crompton, R. W. (1990). Personal communication.
- Crompton, R. W., Gibson, D. K., and McIntosh, A. I. (1969). *Aust. J. Phys.* **22**, 715.
- Crompton, R. W., Gibson, D. K., and Robertson, A.G. (1970). *Phys. Rev. A* **2**, 1386.
- Dalba, G., Fornasini, P., Lazzizzera, I., Ranieri, G., and Zecca, A. (1980). *J. Phys. B* **13**, 2839.
- Ehrhardt, H., Langhans, L., Linder, F., and Taylor, H. S. (1968). *Phys. Rev.* **173**, 222.
- Engelhardt, A. G., and Phelps, A. V. (1963). *Phys. Rev.* **131**, 2115.
- England, J. P., Elford, M. T., and Crompton, R. W. (1988). *Aust. J. Phys.* **41**, 573.
- Ferch, J., Raith, W., and Schroder, K. (1980). *J. Phys. B* **13**, 1481.
- Furst, J., Mahgerefteh, M., and Golden, D. E. (1984). *Phys. Rev. A* **30**, 2256.
- Gibson, T. L., and Morrison, M. A. (1984). *Phys. Rev. A* **29**, 2497.
- Golden, D. E., Bandel, H. W., and Salerno, J. A. (1966). *Phys. Rev.* **146**, 40.
- Haddad, G. N., and Crompton, R. W. (1980). *Aust. J. Phys.* **33**, 975.
- Hammond, P., Read, F. H., Cvejancovic, S., and King, G. C. (1985). *J. Phys. B* **18**, L141.
- Henry, R. J. W., and Lane, N. F. (1969). *Phys. Rev.* **183**, 221.
- Jones, R. K. (1985). *Phys. Rev. A* **31**, 2898.
- Jones, T. J., Cvejancovic, S., Read, F. H., and Woolf, M. B. (1989). In 'Electronic and Atomic Collisions', XVI ICPEAC (Eds A. Dalgarno *et al.*), Abstracts of Contributed Papers, 233.
- Jung, K., Scheurlein, K. M., Sohn, W. K., Kochem, K-H., and Ehrhardt, H. (1987). *J. Phys. B* **20**, L327.
- Khakoo, M. A., and Trajmar, S. (1986). *Phys. Rev. A* **34**, 138.
- Linder, F., and Schmidt, H. (1971). *Z. Naturforsch.* **26a**, 1603.
- McConkey, J. W., Trajmar, S., and King, G. C. (1988). *Comm. At. Mol. Phys.* **22**, 17.
- Morrison, M. A., Crompton, R. W., Saha, B. C., and Petrovic, Z. Lj. (1987). *Aust. J. Phys.* **40**, 239.
- Morrison, M. A., Feldt, A. N., and Saha, B. C. (1984). *Phys. Rev. A* **30**, 2811.
- Nesbet, R. K. (1978). *Phys. Rev. A* **20**, 58.
- Nickel, J. C., Zetner, P. W., Shen, G., and Trajmar, S. (1989). *J. Phys. E* **22**, 730.
- Nishimura, H., Danjo, A., and Sugahara, H. (1985). *J. Phys. Soc. Jpn* **54**, 1757.
- Olander, D. R., and Kruger, V. (1970). *J. Appl. Phys.* **41**, 2769.
- Petrovic, Z. Lj., and Crompton, R. W. (1987). *Aust. J. Phys.* **40**, 347.
- Phelps, A. V. (1968). *Rev. Mod. Phys.* **40**, 399.
- Pichou, F., Huetz, A., Joyez, G., and Landau, M. (1978). *J. Phys. B* **11**, 3683.
- Rau, A. R. P. (1971). *Phys. Rev. A* **4**, 207.
- Read, F. H. (1984). *J. Phys. B* **17**, 3965.
- Schubert, E., Jung, K., and Ehrhardt, H. (1981). *J. Phys. B* **14**, 3267.
- Shyn, T. W., and Sharp, W. E. (1981). *Phys. Rev. A* **24**, 1734.
- Snitchler, G., Alston, S., Norcross, D., Saha, B., Danby, G., Trail, W., and Morrison, M. A. (1990). To be published.
- Sohn, W.K. (1986). Personal communication to M. A. Morrison.
- Sohn, W. K., Jung, K., and Ehrhardt, H. (1983). *J. Phys. B* **16**, 891.
- Srivastava, S. K., Chutjian, A., and Trajmar, S. (1975). *J. Chem. Phys.* **63**, 2659.
- Subramanian, K. P., and Kumar, V. (1989). *J. Phys. B* **22**, 2387.
- Wannier, G. H. (1953). *Phys. Rev.* **90**, 817.