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Single and double ionisation of helium by electron impact

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Abstract. A pulsed crossed-beam technique incorporating time-of-flight spectroscopy which was recently developed in this laboratory has been applied to measurements of the cross sections for single and double ionisation of helium. Measurements over the unusually wide energy range from near threshold to 10 000 eV provide valuable checks on previous measurements based on different experimental approaches and an assessment of the range of validity of a number of theoretical predictions.

1. Introduction

In recent work in this laboratory (Shah *et al* 1987) we developed a pulsed crossed-beam technique incorporating time-of-flight spectroscopy for measurements of the electron impact ionisation cross section of atomic hydrogen with high precision over a wide energy range. Although the technique was developed primarily to overcome the special difficulties in carrying out accurate measurements with hydrogen atoms it can also be applied with advantage to stable gas targets where the results of traditional methods often exhibit significant discrepancies. This is illustrated in the present work by use of the pulsed crossed-beam technique to study the electron impact ionisation of helium. Cross sections for both single and double ionisation have been determined for electron impact energies ranging from near threshold to 10 keV. These data, which span an unusually wide energy range, make possible a detailed assessment of the range of validity of several theoretical predictions for single ionisation. The need for a satisfactory model of double ionisation of helium by fast point-charge projectiles (cf McGuire 1982, Reading and Ford 1987) has also been highlighted in recent work.

There have been many previous studies of the electron impact ionisation of helium. Most of these have been based on beam-static-gas-target methods in which a total ionisation cross section $\sigma_t = \sigma_1 + 2\sigma_2$ (where σ_1 and σ_2 are the cross sections for single and double ionisation respectively) is determined from measurements of the total yield of secondary ions produced from a specified path length and target gas density. There are many possible sources of error and serious discrepancies arise between the results of different experiments. These have been considered in a critical review by Kieffer and Dunn (1966). Mass spectrometric analysis of the secondary ion products in the beam-static-gas approach (cf Schram *et al* 1966) can provide separate cross sections σ_1 and σ_2 but it is difficult to ensure that product ions are extracted from the region of formation and recorded with equal efficiency.

A fast crossed-beam approach has been used by Montague *et al* (1984) to obtain absolute cross sections for single ionisation up to 750 eV. In this method a keV energy beam of He atoms is prepared by charge transfer neutralisation of He^+ ions in passage through helium. The fast He beam intersects an electron beam in an arrangement where the collision volume and density profiles are carefully defined. Fast He^+ product ions are separated by magnetic analysis and recorded with high efficiency. The method obviates the need for electric or magnetic fields in the target region and thereby avoids some of the uncertainties inherent in the beam-static-target approach. However, care must be taken to ensure a negligible content of metastable atoms in the fast He beam. A fast crossed-beam method has recently been used by Wetzel *et al* (1987) for studies of single ionisation for electron energies up to 200 eV.

In the present work, short duration pulses of electrons are passed through a thermal energy beam of ground-state He atoms in a high-vacuum region. Immediately after the transit of each electron pulse through the beam, slow He^+ and He^{2+} ions are swept out of the beam intersection region by a pulsed electric field and selectively identified (in the presence of background-gas product ions) by their characteristic times of flight to a particle multiplier.

Unlike previous beam-static-gas experiments, the ionisation in our experiment takes place in the absence of electric or magnetic fields thereby reducing some of the uncertainties in calibration. In the present measurements, as in our earlier work with H atoms (Shah *et al* 1987), cross sections have been determined by reference to well established cross sections σ_c for charge transfer in H^+–He collisions and σ_i for single ionisation of He by protons (Shah and Gilbody 1985).

2. Experimental approach

2.1. General description

A detailed description of the basic apparatus and measuring procedure has been given previously (Shah *et al* 1987) and only the main features need to be summarised here.

An electron gun was triggered by a pulse generator to provide electron pulses of 100 ns duration at 10^5 pulses/s. The electrons intersected (at right angles) a beam of helium atoms effusing from a bunch of 1 mm diameter hypodermic needles housed inside a 4 mm diameter tube. In the beam intersection region, which was located 10 mm from the tip of the needle assembly, the estimated He atom beam density was about 10^{12} atoms/cm^3. The intersection region was maintained at a base pressure of about 2×10^{-7} Torr.

Immediately after the transit of each pulse of electrons through the helium beam slow He^+ or He^{2+} ions formed by ionisation were swept out of the beam intersection region by a pulsed electric field, accelerated through the same potential difference and then recorded as individual counts by a particle multiplier. The extraction field was produced by application of pulses of approximately 100 V amplitude and 500 ns duration between a pair of high-transparency grids located on either side of the beam intersection region.

The extracted He^+ and He^{2+} ions were identified and distinguished from background-gas product ions by their different times of flight to the multiplier in accordance with their charge to mass ratios. As in our previous work, great care was taken to ensure high and equal extraction efficiency irrespective of the primary electron energy. The extraction pulse generator was triggered by the main pulse generator (which operated the electron gun) via a variable delay which could be adjusted according to the transit time of the trailing edge of the electron pulse through the He beam.

A time to amplitude converter (TAC) operated with start pulses from the extraction pulse generator and with stop pulses from the particle multiplier provided time-of-flight spectra on a multichannel analyser (MCA) of the type shown in figure 1. This shows a portion of the spectrum obtained with an electron impact energy of 500 eV which can be seen to exhibit clearly resolved peaks corresponding to He^+ and He^{2+} product ions.

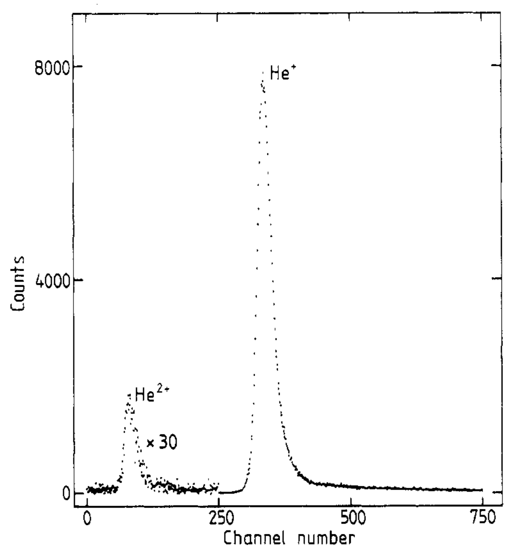

Figure 1. Time-of-flight spectra showing He^+ and He^{2+} products of ionisation observed with 500 eV electrons.

2.2. Electron beam system

The seven-element electron gun was the same as that described previously (Shah *et al* 1987) and utilised a V-shaped thoriated tungsten filament. This provided pulsed beams equivalent to between 1 and 3 nA in the continuous mode with very small angular divergence. The high sensitivity of our technique obviated the need for higher currents. The final four lens elements in the gun assembly were used to adjust the electron beam energy up to a maximum of 10 keV while maintaining an essentially unchanged beam intensity.

In the interaction region the electron beam had a diameter not exceeding 2 mm while the diameter of the helium beam was 4 mm. As in our previous work, careful checks were made to ensure that the effective collision volume was insensitive to the vertical position of the electron beam. The electron beam current was recorded by a screened Faraday cup located beyond the beam intersection region. An indication of the intensity of the helium beam was obtained from the pressure recorded by an ionisation gauge in the main vacuum chamber. The signals from both the Faraday cup and the ionisation gauge were digitised and fed into a microcomputer which also recorded the time-of-flight spectra from the MCA.

The collision energy of the electron beam can be expressed as $E = V_f - d$ where V_f is the acceleration voltage applied to the midpoint of the V-shaped filament and d is a correction parameter which allows for filament misalignment and the effect of contact potentials. A value of $d = 1.4 \pm 0.1$ V was obtained by linear extrapolation of the low-energy data to the threshold value of 24.6 eV for the ionisation of helium.

2.3. *Cross section measurements and normalisation*

At each particular electron impact energy the He^+ and He^{2+} yields were obtained from the average of several measurements of the area of the appropriate peak in the time-of-flight spectrum. Allowance was made for the very small contribution from the background gas by checking the time-of-flight spectrum in the absence of the helium.

The cross sections σ_1 and σ_2 for single and double ionisation of helium were obtained from the expressions

$$\sigma_1 = S(He^+)/k\mu \tag{1}$$

and

$$\sigma_2 = S(He^{2+})/k\mu \tag{2}$$

where $S(He^+)$ and $S(He^{2+})$ are the He^+ and He^{2+} yields per unit electron beam intensity, μ is the effective target density of the He atoms, a quantity related to the pressure recorded in the crossed-beam vacuum chamber. The constant k is the overall detection efficiency of the helium ions. In previous work (Shah and Gilbody 1985) in which we used a crossed-beam coincidence technique incorporating time-of-flight spectroscopy to study the ionisation of helium by H^+, He^{2+} and Li^{3+} impact, we have shown that the detection efficiency k was equal for He^+ and He^{2+} ions when the particle multiplier was operated with -4.5 kV on the first dynode as in the present experiment.

In order to determine σ_1 and σ_2 the product $k\mu$ in equations (1) and (2) was determined by reference to known equivelocity proton impact cross sections. As in our previous work (Shah *et al* 1987) a pulsed proton beam from a 10–100 keV accelerator could be substituted for the electron beam with the target conditions unchanged. With proton impact, He^+ product ions arise from both the single ionisation process

$$H^+ + He \rightarrow H^+ + He^+ + e \tag{3}$$

with cross section σ_i, and the charge transfer process

$$H^+ + He \rightarrow H + He^+ \tag{4}$$

with cross section σ_c. Thus the observed He^+ yield can be described in terms of the cross section sum $\sigma(He^+) = \sigma_i + \sigma_c$. Values of σ_c were obtained from the absolute measurements of Stier and Barnett (1956) which are believed to be accurate to within $\pm 5\%$. Values of σ_i were obtained from our previous measurements of proton impact ionisation (Shah and Gilbody 1985) which in turn were normalised to the values of σ_c due to Stier and Barnett (1956).

Our normalised values of σ_1 for single ionisation of He by electrons together with the present values of $(\sigma_c + \sigma_i)$ for 10–100 keV protons and previously measured cross sections σ_c and σ_i are shown in figure 2. The two sets of data can be seen to be in excellent accord with cross sections for equivelocity electrons and protons becoming equal at high velocities in accord with the predictions of the first Born approximation. Values of σ_2 for double ionisation of He were measured relative to σ_1 and the ratio $R = \sigma_1/\sigma_2$ of these cross sections is independent of our normalisation procedure. Our measured values of σ_1 and σ_2 are shown in table 1. The uncertainties associated with individual cross sections are assessed at the 67% confidence level and reflect the

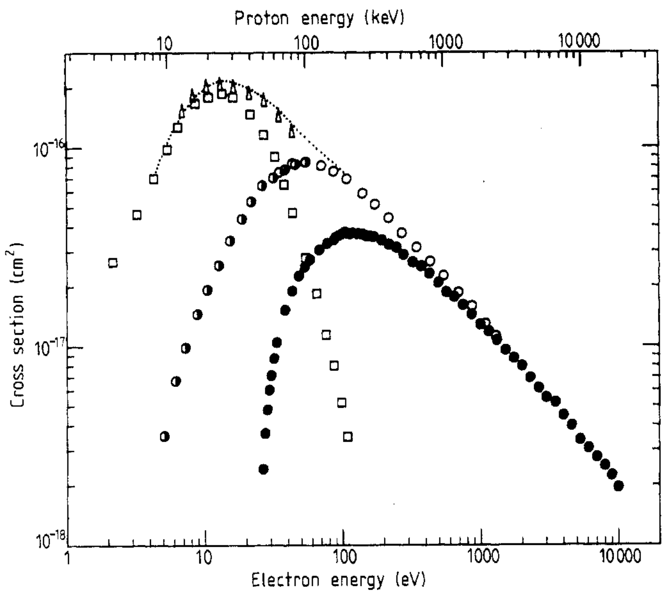

Figure 2. Equivelocity cross sections for He^+ production in collisions of protons and electrons with hydrogen atoms. ●, cross sections σ_1 for single ionisation of He by electron impact (present work); ○, ◑, cross sections σ_i for single ionisation of He by proton impact (Shah and Gilbody 1985 and unpublished data); □, cross sections σ_c for charge transfer in collisions of protons with He (Stier and Barnett 1956); △, cross section sum $\sigma_i + \sigma_c$ for proton impact (present work); . . . , cross section sum $\sigma_i + \sigma_c$ with σ_i taken from ○ above and σ_c taken from □.

degree of reproducibility of the values in terms of the various experimental parameters and statistical fluctuations. Additional estimated uncertainties of ±6% and ±7% in the values of σ_1 and σ_2, respectively, are associated with our normalisation procedure.

3. Results and discussion

3.1. Single ionisation

Figure 3 shows our measured cross sections σ_1 for single ionisation of helium by electrons in the energy range 26.6-10 000 eV. We also show for comparison some results which are intended to be representative of the many previous measurements. A plot of the fractional deviation of these results from the present values is also shown to facilitate comparison. At energies below 1000 eV, agreement in most cases is within the maximum combined estimated uncertainties. We note, in particular, that the results of Montague *et al* (1984), based on the crossed-beam method employing a fast He atom beam, which extend to 750 eV, are in practically perfect agreement with the present data. However the more recent results of Wetzel *et al* (1987) for energies up to 200 eV, which are also based on the use of a fast He atom beam, can be seen to deviate from the present values by up to 9%.

Of the many previous measurements based on the beam-static-gas approach, the measurements of Rapp and Englander-Golden (1965) which extend to 1000 eV are believed to be amongst the most reliable. Their results, which are strictly measurements

Table 1. Cross sections σ_1 and σ_2 for the single and double ionisation of helium by electrons.

Energy (eV)	σ_1 (10^{-17} cm^2)	σ_2 (10^{-19} cm^2)	Energy (eV)	σ_1 (10^{-17} cm^2)	σ_2 (10^{-19} cm^2)
26.6	0.242 ± 0.008		280	2.89 ± 0.03	1.35 ± 0.06
27.6	0.366 ± 0.009		325	2.65 ± 0.03	1.31 ± 0.06
28.6	0.480 ± 0.013		375	2.53 ± 0.03	1.33 ± 0.04
29.6	0.604 ± 0.012		430	2.32 ± 0.03	1.23 ± 0.05
30.6	0.715 ± 0.016		500	2.09 ± 0.03	1.16 ± 0.05
32.1	0.871 ± 0.022		570	1.87 ± 0.02	0.990 ± 0.040
33.6	1.05 ± 0.03		650	1.77 ± 0.02	0.896 ± 0.027
38.6	1.52 ± 0.03		750	1.61 ± 0.02	0.836 ± 0.025
43.6	1.90 ± 0.03		870	1.44 ± 0.02	0.696 ± 0.036
48.6	2.26 ± 0.05		1 000	1.28 ± 0.02	0.597 ± 0.015
53.6	2.50 ± 0.04		1 150	1.19 ± 0.02	0.540 ± 0.018
58.6	2.73 ± 0.06		1 320	1.07 ± 0.02	0.453 ± 0.025
68.6	3.05 ± 0.07		1 520	0.955 ± 0.012	0.403 ± 0.015
78.6	3.29 ± 0.06		1 750	0.872 ± 0.010	0.363 ± 0.026
88.6	3.45 ± 0.06		2 010	0.796 ± 0.009	0.313 ± 0.017
90.2	3.53 ± 0.05	0.063 ± 0.012	2 300	0.693 ± 0.008	0.259 ± 0.009
95.2	3.60 ± 0.05	0.095 ± 0.013	2 650	0.615 ± 0.009	0.218 ± 0.006
100	3.67 ± 0.08	0.172 ± 0.025	3 000	0.551 ± 0.007	0.187 ± 0.006
105	3.74 ± 0.09	0.255 ± 0.020	3 500	0.520 ± 0.013	0.176 ± 0.010
110	3.70 ± 0.05	0.302 ± 0.015	4 000	0.448 ± 0.006	0.142 ± 0.003
115	3.67 ± 0.05	0.388 ± 0.025	4 600	0.398 ± 0.005	0.122 ± 0.005
120	3.70 ± 0.04	0.439 ± 0.031	5 300	0.337 ± 0.004	0.102 ± 0.006
130	3.69 ± 0.05	0.561 ± 0.030	6 100	0.308 ± 0.003	0.087 ± 0.003
140	3.67 ± 0.04	0.748 ± 0.040	7 000	0.276 ± 0.003	0.073 ± 0.002
150	3.60 ± 0.04	0.806 ± 0.036	8 000	0.250 ± 0.003	0.068 ± 0.003
160	3.58 ± 0.04	0.864 ± 0.036	9 000	0.224 ± 0.003	0.059 ± 0.003
170	3.55 ± 0.05	0.937 ± 0.045	10 000	0.195 ± 0.005	0.051 ± 0.003
195	3.42 ± 0.05	1.16 ± 0.05			
220	3.25 ± 0.04	1.23 ± 0.04			
250	3.13 ± 0.04	1.32 ± 0.04			

of $(\sigma_1 + 2\sigma_2)$ where σ_2 is relatively small (see figure 3), can be seen to exhibit deviations from the present values by up to 8%. The higher-energy data due to Schram *et al* (1966) and Nagy *et al* (1980) can be seen to exhibit much more serious deviations from the present values by up to 18%.

In figure 4 our cross section σ_1 for single ionisation of helium may be compared with a number of theoretical estimates based on different approximations. Calculations by Bell and Kingston (1969) based on the first Born approximation employ a six-parameter correlated wavefunction for the ground state of helium with polarised-orbital wavefunctions for the $l = 0$, 1 and 2 partial waves of the continuum state and with Hartree–Fock wavefunctions for the $l = 3$, 4 and 5 partial waves of the continuum state. Both length and velocity formulations of the Born matrix element were used to provide results which differ by only about 3% in the region of the cross section maximum and 1% above 600 eV. In figure 4 we show only the slightly larger values obtained from the velocity formulation. At impact energies above 1000 eV, these theoretical values are in good agreement with our measured cross sections.

At intermediate energies in the region of the cross section maximum there is generally poor agreement between experiment and the theoretical predictions. Values

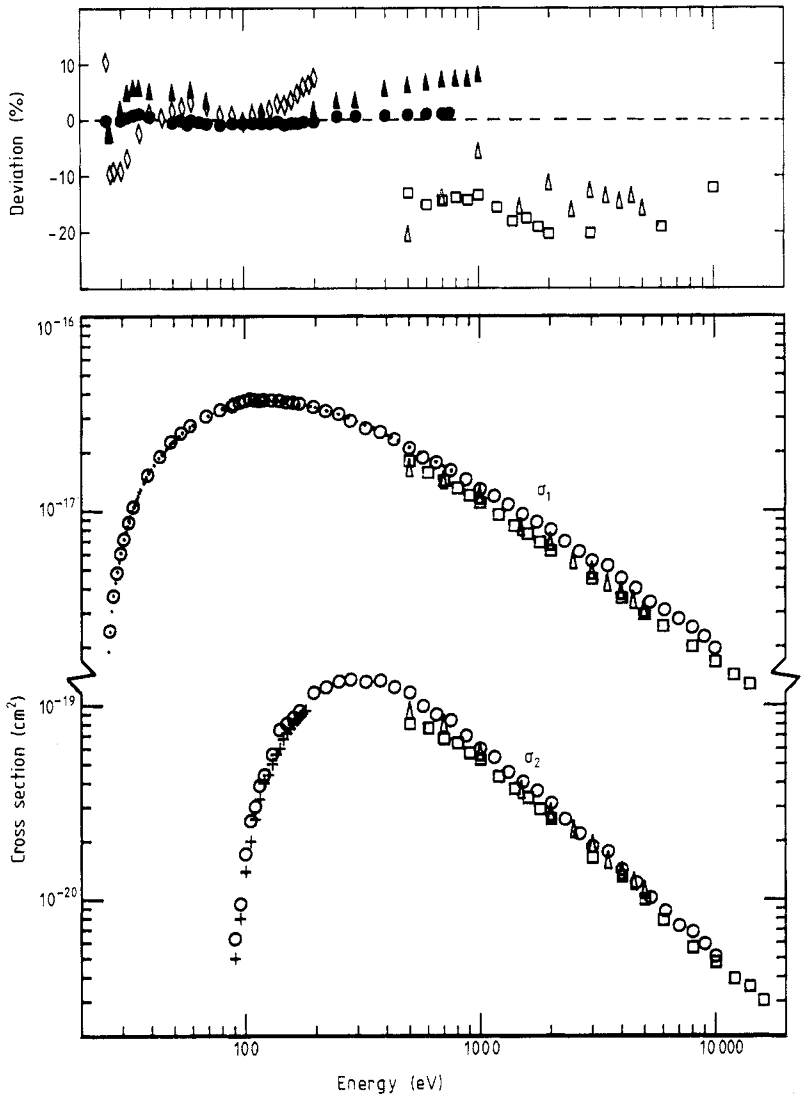

Figure 3. Cross sections σ_1 and σ_2 for single and double ionisation of helium by electron impact. ○, present work; •, ●, Montague *et al* (1984); △, Nagy *et al* (1980); □, Schram *et al* (966); +, Stephan *et al* (1980); ▲, Rapp and Englander-Golden (1965); ◇, Wetzel *et al* (1987). The upper plot shows the fractional deviation of previously measured values of σ_1 from the present values.

calculated by Bell and Kingston (1969) are up to 70% larger than experiment. Values up to 60% larger than experiment have been calculated by McGuire (1971) from generalised oscillator strengths using approximate one-electron wavefunctions. Recent calculations by Bartschat and Burke (1987) based on the use of the *R*-matrix method provide cross sections up to 34% larger than experiment. The results obtained by Peach (1966) using the Ochkur approximation seem to agree best with experiment in the region of the cross section maximum. Values calculated by Peach (1966) using the Born approximation with allowance for electron exchange are rather smaller than

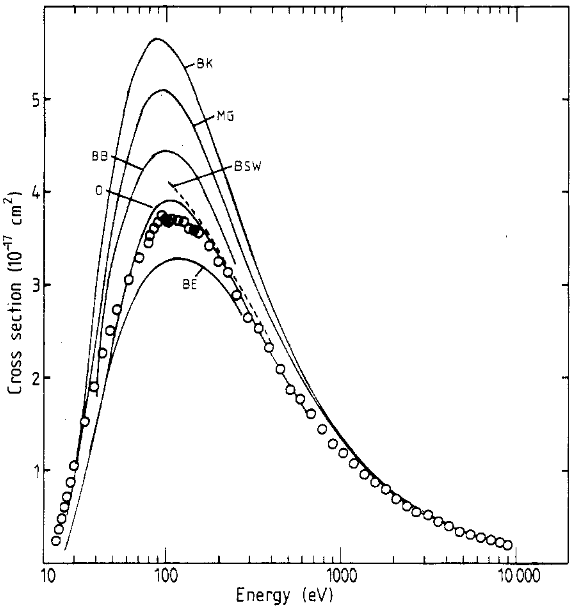

Figure 4. Comparison of measured cross sections for single ionisation of helium by electrons with theoretical estimates. ○, present measurements; BK, Bell and Kingston (1969), Born approximation; MG, McGuire (1971), generalised oscillator strength approach; BB, Bartschat and Burke (1987), *R*-matrix method; BSW Bransden *et al* (1979), distorted-wave method; O, Peach (1966), Ochkur approximation; BE, Peach (1966), Born exchange.

experimental values. Cross sections calculated by Bransden *et al* (1979) based on the use of a distorted-wave model can be seen to be in reasonable accord with experiment only at energies above the cross section maximum.

The fact that our measurements extend to impact energies of 10 keV allows us to examine the extent to which our cross sections σ_1 are described by the Bethe (1930) approximation. This predicts that at sufficiently high impact energies, E, cross sections, σ_1, can be expressed in the form

$$\sigma_1 E = A \log E + B$$

where A and B are constants for a particular atom. In figure 5 our values of the product σ_1 plotted against log E exhibit the expected tendency to approach the Bethe asymptote at high energies. The best fit to the experimental data indicates values of $A = 590 \pm 20$ and $B = 325 \pm 75$. These compare favourably with the values of $A = 567$ and $B = 311$ calculated by Bell and Kingston (1969) using their velocity formulation in the first Born approximation.

3.2. *Double ionisation*

Our measured cross sections σ_2 for double ionisation of helium (figure 3) are about three orders of magnitude smaller than the corresponding values of σ_1. Previous measurements by Schram *et al* (1966) and by Nagy *et al* (1980) based on the mass spectrometric analysis of ions in the beam-static-gas-target approach are included in figure 3. Values due to Schram *et al* (1966) are smaller than our values by 35% at

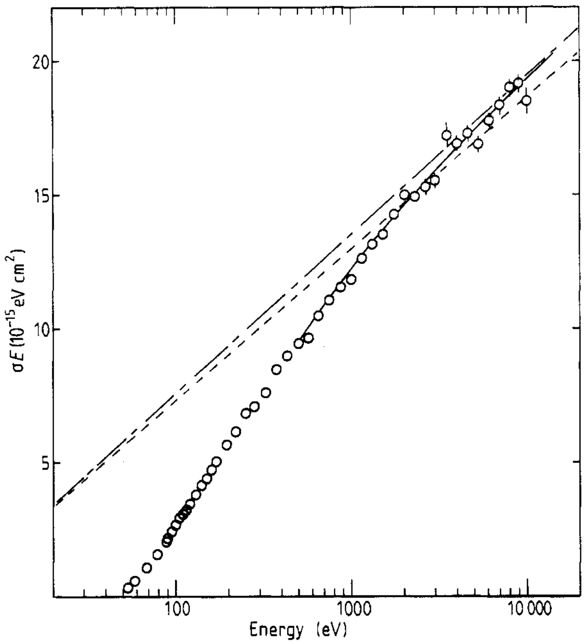

Figure 5. Bethe plot of $\sigma_1 E$ against log E for electrons on helium. ○, present data; — — —, best asymptotic fit to present data; - - -, theoretical prediction, Bell and Kingston (1969).

their lowest energy of 500 eV and by 8% at our highest energy of 10 000 eV. The results of Nagy *et al* (1980) which span the range 500–5000 eV are in good agreement with our values at the higher energies but fall below our values at the lower impact energies; at 500 eV their cross section is 17% smaller. Stephan *et al* (1980) have carried out a mass spectrometric analysis of the ions from a Nier ion source to obtain ratios σ_2/σ_1 in the range 80–180 eV. Separate values of σ_2 obtained by normalisation to the total cross sections measured by Rapp and Englander-Golden (1965) can be seen (figure 3) to be in excellent agreement with our values.

McGuire (1982) has considered the double ionisation of helium in terms of the 'shake-off' (SO) and 'two-step' (TS) mechanisms. The SO mechanism, which corresponds to single ionisation followed by shake-off of the second electron due to change of the electronic screening of the nucleus, should be dominant at high velocities when $R = \sigma_2/\sigma_1$ should become independent of velocity. In the TS mechanism, which is dominant at lower velocities, both electrons are ejected as a result of a single collision with the projectile. The probability of the TS process can be determined from the product of the probabilities of removing each electron separately. The observation that the ratio R for electrons is up to about a factor of two larger than that for ionisation by equivelocity protons has also stimulated further theoretical and experimental studies with other projectiles including antiprotons (cf Andersen *et al* 1987, Reading and Ford 1987).

In figure 6 we show the present values of R for electron impact compared with previously measured values. We also show for comparison corresponding values of R for equivelocity protons previously measured by ourselves (Shah and Gilbody 1985) and by Andersen *et al* (1987). At intermediate velocities, much larger values of R are

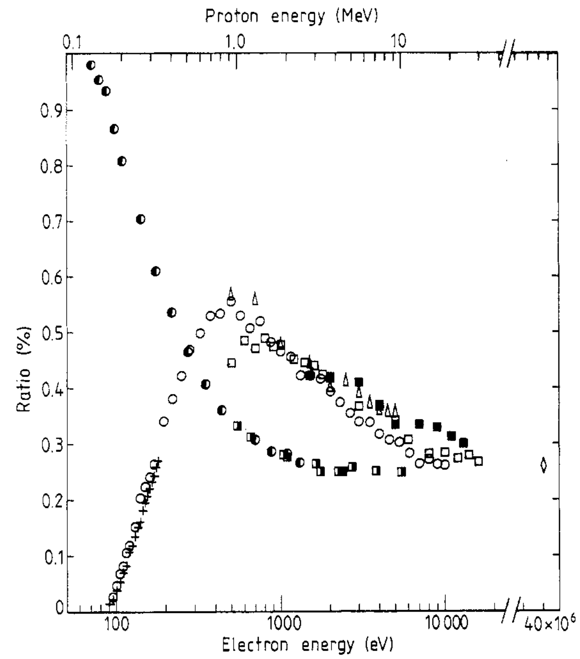

Figure 6. Ratio R of cross sections for double to single ionisation of helium for electron and equivelocity proton impact. Electron impact: ○, present results, △, Nagy *et al* (1980); □, Schram *et al* (1966); ■, Andersen *et al* (1987); +, Stephan *et al* (1980); ◇, Muller *et al* (1983). Proton impact: ◐, Shah and Gilbody (1985); ◧, Andersen *et al* (1987).

apparent for electrons, a feature which McGuire (1982) has tried to explain in terms of the different contributions to the cross sections as a result of interference between the probability amplitudes associated with the TS and SO mechanisms. At high velocities, values of R for both electron and proton impact can be seen to be approaching the common velocity-invariant value of about 2.4×10^{-3} predicted by McGuire (1982). For 40 MeV electrons Muller *et al* (1983) measured a value of $R = 2.6\times10^{-3}$. However, it can be seen that values of R for electrons previously measured at intermediate velocities exhibit significant scatter. The present values of R exhibit a much smoother approach to the asymptotic value than, for example, the recent results of Andersen *et al* (1987) which are about 20% larger at 10 keV.

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