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Total cross sections for the production of metastable neon atoms by electron impact

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Abstract. Total cross sections have been measured for the production of the metastable $1s_3$ and $1s_5$ states in neon at incident electron energies from threshold to 500 eV. A time of flight technique was used to determine relative cross sections which were made absolute by comparison with the known cross sections for metastable production in helium. Reasonable agreement is found between the present results and those of Phillips *et al* who used different techniques to measure relative cross sections and to normalise their data. This good agreement has implications for the mechanism of the excitation of the metastable states.

1. Introduction

Apart from the intrinsic interest in the cross sections for the production of metastable states in neon, these cross sections play an important role in the study of gas discharges in neon and in model calculations of various laser systems. Nevertheless information on these cross sections, both theoretical and experimental, has been very scarce. The theoretical problem is compounded in this case by the choice of a coupling scheme to describe the excited states. Bransden and McDowell (1978) note that whereas *LS* coupling can be used to describe the excited states in helium, the spin-orbit interaction in the excited states in neon is sufficiently strong to demand a j-l coupling scheme. In addition Blum *et al* (1980) have questioned the validity of *LS* coupling in the description of the interaction mechanism in neon. Veldre *et al* (1965) have used the first Born approximation to calculate excitation cross sections of the excited states in neon. In particular they found that the cross section for direct excitation of the $1s_3$ and $1s_5$ states was zero. This can be readily understood as they used an *LS* coupling basis and made no allowance for exchange scattering. In this basis both the $1s_3$ and $1s_5$ states are triplet and can only be excited by exchange scattering.

Taylor *et al* (1983) have calculated cross sections for metastable excitation in neon using an R-matrix theory in an *LS* coupling basis. This theory has successfully predicted the positions of several of the observed resonances in the cross section at energies up to 2.5 eV above threshold (Buckman *et al* 1983).

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Machado *et al* (1984) have calculated total and differential cross sections for several excited states in neon using a first-order many-body theory. This theory belongs to the class of distorted-wave approximations and omits several coupling effects which are features of the R-matrix approach. These calculations included spin-orbit coupling effects in the target states.

The determination of absolute experimental cross sections has, in the main, also relied heavily on the *LS* coupling scheme to describe the excited states. For example, de Heer *et al* (1979) have assumed that the $1s_3$ and $1s_5$ states are triplet and estimate that the combined cross sections for direct excitation to these states is less than $2.4\times10^{-3}\pi a_0^2$ at 40 eV. Phillips *et al* (1981a, b) use empirical arguments to justify their assumption that these states are populated by cascades for electron impact energies greater than 90 eV. They then use the cascade cross sections of Sharpton *et al* (1970) to assign absolute values to the cross sections at 90 eV. In these experiments Phillips *et al* have used a laser to excite the metastable states to fluorescing levels thereby measuring relative excitation functions of the metastable states. Buckman *et al* (1983) have normalised their relative measurements to the cross sections of Taylor *et al* (1983) at an incident energy of 18 eV.

Such normalising procedures make it impossible to test the validity of the underlying assumptions and very difficult to estimate the magnitudes of the uncertainties in the experimental cross sections.

Register *et al* (1984) have recently measured differential and total cross sections for the excitation of the forty lowest lying states in neon. These cross sections included those for the excitation of the $1s_3$ and $1s_5$ states at incident energies of 25, 30 and 50 eV. The procedure followed in these measurements was to integrate the measured relative differential cross sections; the absolute cross section was determined by comparing these cross sections with the differential cross section for elastic scattering at the energies of interest. The metastable cross sections derived from this study are therefore free from any ambiguities surrounding the validity of *LS* coupling in neon.

In the present study we have used a time of flight technique to measure relative excitation functions for the metastable states in neon. These have been made absolute by comparing them with the known cross sections for the excitation of the 2^1S state in helium. Comprehensive reviews of the relevant theoretical and experimental cross sections in helium have been made by de Heer and Jansen (1977) and by Bransden and McDowell (1978). The time of flight technique does not distinguish between metastable levels which are excited directly from the ground state or those which are populated by cascades from higher levels. Thus our data are directly comparable with those of Phillips *et al* (1981a, b) but in the present case no assumptions have been made concerning the validity of the *LS* coupling scheme in neon. Consequently comparison between the two sets of cross sections can provide information on the role of *LS* coupling in the collision.

2. Experimental procedures

The experimental procedure consisted of two parts. First a relative excitation function for the metastable states in neon was measured. These relative data were then made absolute by comparing the metastable yield in neon with that in helium. In the case of helium there is no doubt that the *LS* coupling scheme applies both in the description of the excited states and in the excitation process. Thus the 2^3S state can only be populated by exchange scattering which can be ignored at incident electron energies of 200 eV (Moiseiwitsch and Smith 1968). We have deduced the total cross section for population of the 2^1S state at an incident energy of 400 eV which was used to normalise our helium data at all energies.

2.1. *Relative cross sections*

The relative measurements were made using a time of flight apparatus which is shown schematically in figure 1. The basic design of the apparatus was similar to that which has been described by Lloyd *et al* (1972). However all of the component parts have been rebuilt.

A pulsed beam of electrons excited a known pressure of target gas. The electron beam was collected in a large Faraday cup which ensured that all of the incident current was detected. The beam current was monitored with an Ortec Model 439 current digitiser which compensated for drifts in the incident electron current and for changes in the beam current with energy.

The interaction region was defined by the intersection of the electron beam and the field of view of the detector. The products of the collision, that is photons, ions, electrons and metastable atoms, passed through a series of grids and a drift tube before reaching the detector. The ions and electrons could be stopped by appropriate electric fields between the grids. The photons and metastable atoms could be distinguished by their arrival times at the detector.

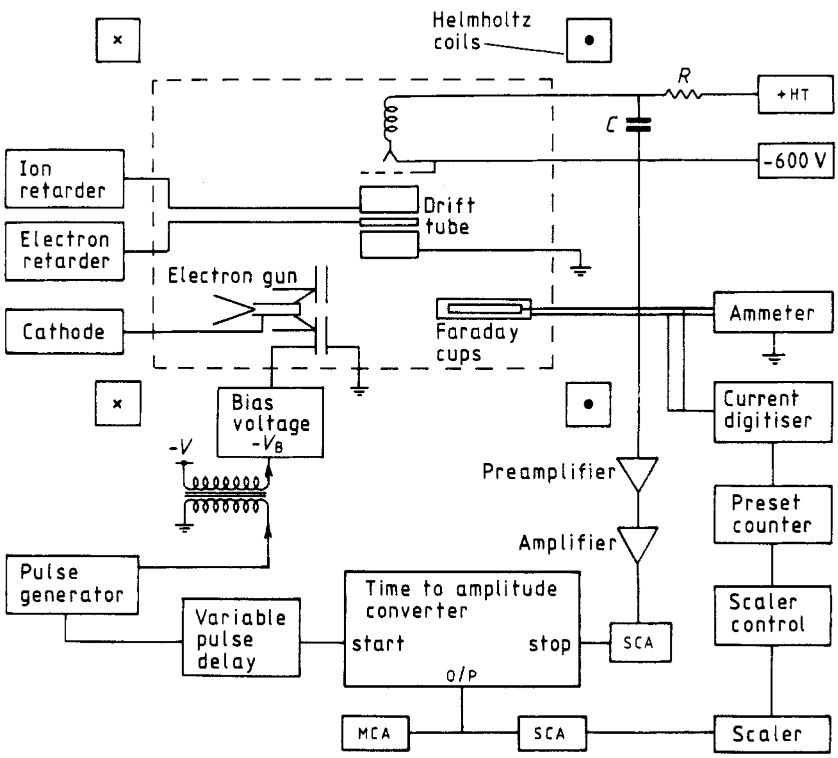

Figure 1. Schematic diagram of the apparatus.

Electrical pulses from the electron multiplier were amplified and acted as the stop pulse for a time to amplitude converter (TAC). The start pulse for the TAC was derived from the pulses which were applied to the electron gun to turn the beam on. These pulses could be delayed thereby enabling us to discriminate against events corresponding to the detection of photons. In general the time for which the electron beam was on was less than 1% of the repetition period.

The output of the TAC was monitored by a multichannel analyser (MCA). A typical time of flight spectrum for helium is shown in figure 2. The feature on the left represents events arising from the detection of photons whilst the broad feature arises from the detection of metastable atoms which leave the interaction region with a Maxwellian distribution of velocities. The experimental data have been fitted to a function of the form (Borst and Zipf 1971)

$$N = Ct^{-4} \exp(-At^{-2}) \tag{1}$$

where C is a constant depending on the geometry of the system and

$$A = \frac{mL^2}{2kT}$$

where L is the flight distance, m the atomic mass, T the ambient temperature and t the time of flight. The results obtained from the fit were consistent with the known values of these parameters namely $L = 11$ cm and $T = 293$ K.

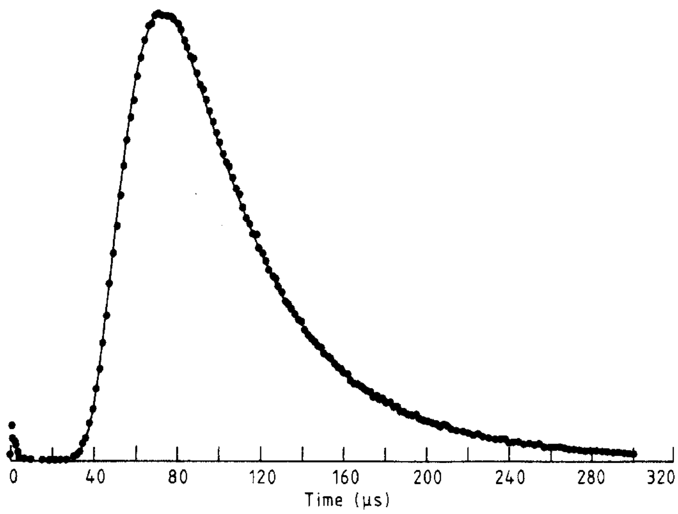

Figure 2. Time of flight spectrum for helium metastables. The line is a fit to the data by a Maxwellian function as explained in the text.

The effects of recoil on the metastable distribution were investigated by studying the time of flight distribution at energies close to threshold and at energies of the order of 200 eV. No difference was observed in the fits to the Maxwellian at any energy.

The procedure for measuring relative cross sections was to record the number of counts in the metastable peak for a constant target pressure and incident electron flux at each energy. The target pressure was measured with an ionisation gauge and the electron flux was set by the current digitiser which controlled a scalar through a preset counter. It was established that the metastable signal was linear with respect to the incident electron current and with target gas pressure.

The electron beam energy was calibrated by observing the resonance at 20.3 eV in the helium cross section (Schulz and Fox 1957) whilst the 16.91 and 18.58 eV resonances in neon (Brunt *et al* 1976) were used to calibrate the electron energy for the neon experiments. The differences between the beam energy measured in this way and that determined from the voltage placed on the cathode of the gun were attributed to contact potentials because great care was taken to ensure that all surfaces bounding the interaction region were at earth potential.

A pair of Helmholtz coils was used to reduce the magnitude of the Earth's magnetic field to 5×10^{-6} T.

2.2. *Normalisation of the cross sections*

The cross section, σ, for the production of metastable atoms is related to the metastable count rate M by

$$\sigma = M\frac{e}{I_0}\frac{4\pi}{(ld\Omega)_{\mathrm{eff}}}\frac{1}{\rho\eta\tau} \tag{2}$$

where e is the electronic charge, I_0 the incident electron beam current, $(ld\Omega)_{\mathrm{eff}}$ the effective scattering length, τ the transmission of the drift tube and electronic circuitry, η the efficiency of the detector for the metastable species and ρ the number density of target atoms.

Any attempt to use equation (2) to determine σ directly was thwarted by the fact that η was unknown for the channel electron multiplier. Therefore an alternative approach was developed which used a stainless steel plate as an emitter of secondary electrons when bombarded by the metastable atoms. The secondary electron yields for such a plate were taken to be the measured values of Dunning *et al* (1975).

The modifications to the apparatus which incorporated the stainless steel plate are indicated in figure 3. The plate was inserted at the end of the drift tube at an angle of 45° to the end of the tube. A channel electron multiplier (CEM) viewed the plate and collected the Auger electrons which were emitted when the metastable atoms struck the plate. Pulses from the CEM were treated in the same way as has been described in the relative measurements. Time of flight spectra were recorded by pulsing the electron beam in an identical fashion as for the relative measurements. The time of flight spectra recorded in this way were similar to those illustrated in figure 2. The photoelectrons from the plate were clearly identifiable and could be discriminated against by their time of flight. Geometrical considerations showed that the apparatus was not sensitive to the specular reflection of metastable atoms from the plate.

We note that the use of such an experimental arrangement to detect metastable atoms was first proposed by Lamb and Retherford (1950).

A ratio technique was then used to circumvent the additional problems implied by equation (2), namely the accurate determination of the product of $(ld\Omega)_{\mathrm{eff}}$ and τ. Helium was introduced at a known pressure into the scattering chamber and the number of Auger electrons emitted from the plate was recorded for a fixed incident electron beam flux at an energy of 200 eV. These measurements were repeated at several helium densities and a linear relationship between counts and pressure was obtained as in figure 4. The chamber was evacuated and the experiment repeated with neon. It was observed that the electron beam profile was independent of the gas in the chamber.

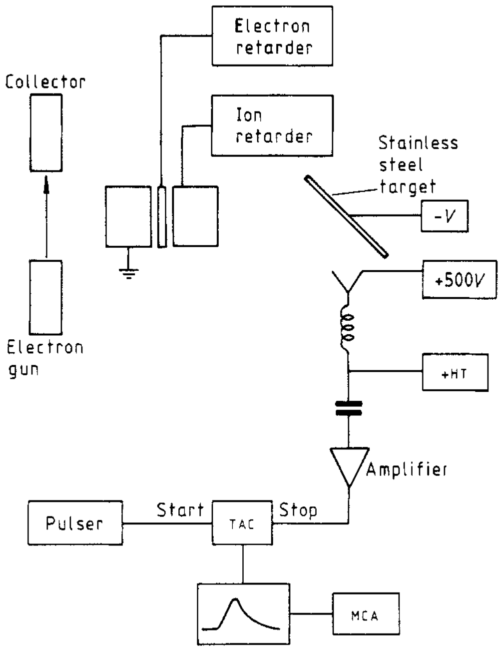

Figure 3. Schematic diagram of the apparatus incorporating the stainless steel target.

It was also established that the collection efficiency of the Auger electrons was independent of the metastable species.

The ratio of the cross sections for neon and helium was given by

$$\begin{aligned}\frac{\sigma_{\mathrm{Ne}}}{\sigma_{\mathrm{He}}}&=\frac{M_{\mathrm{Ne}}}{\rho_{\mathrm{Ne}}}\frac{\rho_{\mathrm{He}}}{M_{\mathrm{He}}}\frac{\eta_{\mathrm{He}}}{\eta_{\mathrm{Ne}}}\\&=\frac{R_{\mathrm{Ne}}}{R_{\mathrm{He}}}\frac{S_{\mathrm{Ne}}}{S_{\mathrm{He}}}\frac{\eta_{\mathrm{He}}}{\eta_{\mathrm{Ne}}}\end{aligned}\tag{3}$$

where R_i was the slope of the graph of signal against ionisation gauge reading as in figure 4 and S_i was the sensitivity of the ionisation gauge to the particular species.

The ratio of the efficiencies η_i for stainless steel between the 2^1S state in helium and the metastable states in neon was taken to be 0.87 ± 0.16 (Dunning *et al* 1975).

Although the relative sensitivities of the ionisation gauge which was used during the experiments were known in principle from the manufacturer's specifications, we calibrated the gauge by carrying out elastic scattering experiments on neon and helium with 100 eV electrons.

Equation (3) can be used for elastic scattering; however, in this case the ratio of detection efficiencies for the elastically scattered electrons at 100 eV is unity. R, obviously, is the slope of the graph of the elastically scattered electron signal against ion gauge reading and the cross sections are those for elastic scattering. In these experiments a cylindrical mirror electron spectrometer viewed the interaction region which was defined by the intersection of the electron beam with a bulk sample of gas. Electrons which had been elastically scattered at 40° were detected and counted. The electron beam was not pulsed. The pressure of helium in the interaction region was

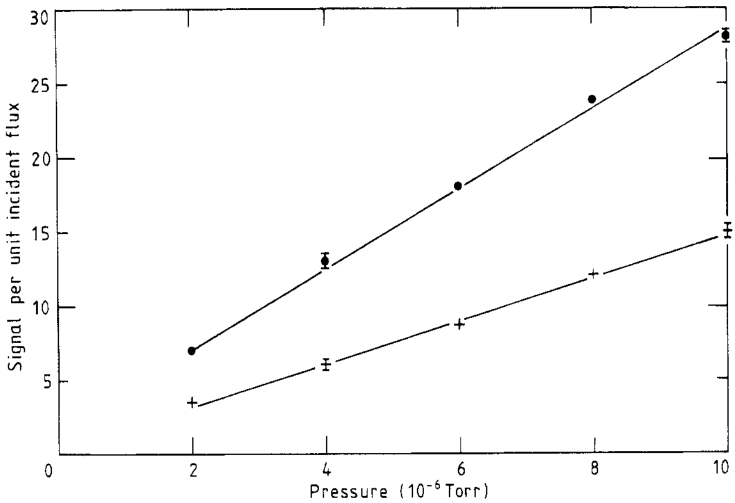

Figure 4. Relationship between metastable count rate and the pressure of helium (·) and of neon (+).

varied and the slope of the graph of the elastic scattering count rate against pressure was measured. The experiment was repeated with neon as the target gas. The ratio of the sensitivities was then calculated using the differential cross sections for elastic scattering of 100 eV electrons from helium and neon of Jansen *et al* (1976). This ratio, which agreed with the manufacturer's specifications, was determined to an overall accuracy of 7%.

The final step in the determination of the absolute value of the neon cross section required the assignment of the helium cross section. The helium metastable signal arose from two sources; direct excitation and cascades from higher n^1P states. The cascade contribution was estimated in the present case by resorting to calculated total cross sections for n^1P excitation. We note that, at an incident energy of 400 eV, there is good agreement between several theoretical approaches up to $n=4$; for example the ten-channel multichannel eikonal approximation of Flannery and McCann (1975), the second-order potential method of Berrington *et al* (1973) and the first Born approximation (Bransden and McDowell 1978) all agree to within about 5% at this energy. Thus we chose to determine these contributions at any incident energy where, on the one hand, the first Born approximation could be used but on the other hand there was still sufficient signal so that good statistical accuracy could be achieved in a reasonable counting time. An incident energy of 400 eV was therefore chosen to normalise the helium cross sections. This value of course fixed the helium cross section at all energies or at least those for which the triplet excitation could be ignored.

The measured cross section for metastable production in helium at an incident energy of 400 eV was given by

$$\sigma_{\mathrm{He}} = \sigma_{\mathrm{Direct}}(2^1\mathrm{S}) + \sum_{n=2}^{\infty} a_n \sigma(n^1\mathrm{P}) \tag{4}$$

$$= \sigma_{\mathrm{Direct}}(2^1\mathrm{S}) + a_2\sigma(2^1\mathrm{P}) + a_3\sigma(3^1\mathrm{P}) + \sum_{n=4}^{\infty} a_n\sigma(n^1\mathrm{P}). \tag{5}$$

Here the coefficients a_n are the branching ratios for the n^1P-2^1S transitions and $\sigma(n^1P)$ the cross sections for the direct excitation of the n^1P states from the ground state. The coefficients a_2 and a_3 are 0.001 and 0.024 respectively (Burger and Lurio 1971); and we note that a_n for $n > 3$ are each approximately equal to 0.03 (Weiss *et al* 1966). To estimate the infinite sum in equation (5) we follow de Heer and Jansen (1977) and assume that the cross sections $\sigma(n^1P)$ are proportional to n^{-3} for $n \geqslant 4$. Therefore

$$\sum_{n=4}^{\infty} a_n \sigma(n^1\mathrm{P}) = 0.03 \times 4^3 \left(\sum_{n=1}^{\infty} \frac{1}{n^3} - \sum_{n=1,2,3} \frac{1}{n^3} \right) \sigma(4^1\mathrm{P})$$

$$= 0.077\sigma(4^1\mathrm{P}).$$

Thus

$$\sigma_{\mathrm{He}} = \sigma_{\mathrm{Direct}}(2^1\mathrm{S}) + 0.001\sigma(2^1\mathrm{P}) + 0.024\sigma(3^1\mathrm{P}) + 0.077\sigma(4^1\mathrm{P}).$$

The cross section for the direct excitation of the 2^1S state from the ground state at 400 eV has been measured by Dillon and Lassettre (1975) to an accuracy of 5%. Similarly the cross section for 2^1P excitation is taken to be that of Dillon and Lassettre (1975) which we note is in excellent agreement with that calculated by the first Born approximation. Using the Born cross sections for $\sigma(3^1P)$ and $\sigma(4^1P)$ (Bransden and McDowell 1978), we deduce that the total cross section for production of metastable states at 400 eV is $6.65 \times 10^{-3} \pi a_0^2$. The total cascade contribution to the cross section at this energy represents only 16% of the total cross section. The cross section, at 200 eV, which was used in equation (3) to determine the neon cross section was $1.22 \times 10^{-2} \pi a_0^2$.

The neon cross section at 200 eV was determined from equation (3) thereby normalising the relative neon cross sections at all energies.

3. Discussion of experimental uncertainties

The dominant contribution to the experimental uncertainty in the neon cross section at a given energy arises from the uncertainty in the ratio of the efficiencies of detection of the helium and neon metastable atoms. The 18% in the ratio is obtained by combining the error in the measurements of Dunning *et al* (1975) and does not include any systematic effects. Perhaps the most significant possible systematic error in this regard arises from the possibility that the surface of the stainless steel used in the present experiments was different from that used by Dunning *et al* (1975). In particular one could speculate that the presence of unknown adsorbed gases on the surface could influence the secondary electron yields from the plate. In this regard we note that Dunning *et al* were unable to identify any effect of different gases in their gas cell even though they searched for such effects. In addition, Dunning *et al* (1975) give secondary emission yields for a Cu–Be surface which, although significantly different from the deduced values of Borst (1971), yield a similar ratio for the efficiencies of helium to neon. We carried out a series of ratio experiments using a particle multiplier tube with a Cu–Be surface under the same experimental conditions which have been described for the relative measurements. These experiments gave a neon cross section at 200 eV which was in good agreement with that determined from the stainless steel plate technique. The ratio $\eta_{\mathrm{He}}/\eta_{\mathrm{Ne}}$ used in this phase of the measurements was 1.15 ± 0.21 (Dunning *et al* 1975). The agreement in the results from these two techniques led us to conclude that any unknown surface effects could be accounted for by the error in the ratio.

Given such a large uncertainty in the efficiencies, the influence of the errors in the other quantities in equation (3) is of only secondary importance. The error in the helium cross section was taken to be 5%. The contribution of the various n^1P cross sections contributes only 16% to the total cross section, thus our procedure is insensitive to ambiguities concerning the validity of the Born approximation or cascade populations to the n^1P levels. The uncertainty in the calibration of the ionisation gauge of 7% arose from the combined uncertainties in the elastic scattering cross sections. The statistical error in the remainder of the quantities in equation (3) was less than 2%. The overall uncertainty in the neon cross section was estimated to be 21%.

Several other possible sources of systematic error in our results have been considered. These include the influence of contact potentials on the incident energy, of the detection of high lying Rydberg states and of the effects of a change in the electron beam geometry as a function of the electron energy. The influence of contact potentials of the beam energy has been identified by observing the onset of the resonance features in the neon cross section (Buckman *et al* 1983). From these measurements together with similar experiments in helium and argon, we conclude that there is a constant 2 eV shift between the potential placed on the oxide cathode and the beam energy. The observed width of the resonances indicated an uncertainty of ± 0.15 eV in the beam energy. This width is consistent with that obtained from oxide cathodes.

It was not possible, with our simple electron gun, to maintain the same beam focus conditions over the whole energy range of the experiments. However we took particular care to ensure that all of the incident electron flux was collected at each energy and that the change in beam diameter had a minimal effect on the effective scattering length. To first order, the cross section defined in equation (2) does not depend on the current density in the electron beam (Kuyatt 1968). This conclusion is valid if the scattering volume is cylindrical. A closer inspection of our experimental geometry reveals that the scattering volume is that generated by the intersection of a cylinder and a cone where the axis of the cylinder is parallel to the base of the cone. Estimates of the influence of this geometry on equation (2) for known electron beam diameters show that at worst a systematic error of $\pm 1.5\%$ is introduced between the extremes of electron energy used in the experiments; this arises because in each case the electron beam radius was small compared with the flight distance.

The contribution of high-lying Rydberg states to the detected signal can be dismissed by comparing the known cross sections for the excitation of such states (Schiavone *et al* 1979) with the present cross sections; for example in neon the peak in the Rydberg excitation cross section at 60 eV is less than 1% of the present cross section at that energy.

4. Results and discussion

The cross sections for the production of the metastable states in neon are shown as a function of the incident energy in table 1 where the uncertainty represents ± one standard deviation. These results are shown in figure 5 where they are compared with the combined data of Phillips *et al* for the production of the $1s_5$ state (Phillips *et al* 1981a) and for the $1s_3$ state (Phillips *et al* 1981b). The large uncertainties in both sets of data reflect the difficulties in assigning absolute values to the cross sections. In our

Table 1. Absolute neon metastable cross section.

Energy (eV)	$\sigma(10^{-2}\pi a_0^2)$	Energy (eV)	$\sigma(10^{-2}\pi a_0^2)$
17	1.23 ± 0.26	110	1.53 ± 0.32
18	1.92 ± 0.40	120	1.37 ± 0.29
19	2.33 ± 0.49	130	1.29 ± 0.27
20	2.74 ± 0.58	140	1.18 ± 0.25
21	3.15 ± 0.66	150	1.18 ± 0.25
22	3.48 ± 0.73	160	1.07 ± 0.22
23	3.79 ± 0.80	170	1.01 ± 0.21
24	3.89 ± 0.82	180	1.01 ± 0.21
25	3.96 ± 0.83	190	0.95 ± 0.20
26	4.00 ± 0.84	200	0.93 ± 0.20
27	3.95 ± 0.83	220	0.85 ± 0.18
28	3.88 ± 0.81	240	0.77 ± 0.16
29	3.77 ± 0.79	260	0.76 ± 0.16
30	3.69 ± 0.77	280	0.69 ± 0.14
31	3.68 ± 0.77	300	0.68 ± 0.14
32	3.58 ± 0.75	320	0.67 ± 0.14
33	3.42 ± 0.72	340	0.63 ± 0.13
34	3.40 ± 0.71	360	0.60 ± 0.13
35	3.37 ± 0.71	380	0.57 ± 0.12
36	3.24 ± 0.68	400	0.53 ± 0.11
37	3.18 ± 0.67	420	0.50 ± 0.11
38	3.09 ± 0.65	440	0.50 ± 0.11
39	2.99 ± 0.63	460	0.50 ± 0.11
40	2.93 ± 0.62	480	0.46 ± 0.10
50	2.62 ± 0.55	500	0.48 ± 0.10
60	2.30 ± 0.48		
70	2.17 ± 0.46		
80	1.97 ± 0.41		
90	1.78 ± 0.37		
100	1.59 ± 0.33		

case this is dominated by the relative quantum efficiencies of the detector whereas in the case of Phillips *et al* the major source of uncertainty arises from the calibration of their data to the total cascade contributions of Sharpton *et al* (1971). These errors in turn were based on the limits of experimental repeatability and do not include any systematic uncertainties in the absolute calibration. Phillips *et al* estimate a 10% error in the energy dependence of their cross sections and quote a total error of ± 25% in the absolute values for the production of the metastable states in neon.

There is reasonable agreement between the shapes of the present cross sections and those of Phillips *et al.* For example both sets of data peak at 26 eV. On the other hand the relative measurements of Theuws *et al* (1983) show that the peak in the cross section occurs at 30 eV. The absolute values of the present cross sections also agree with those of Phillips *et al* who have calibrated their data by assuming that the cross section for direct excitation is zero at 90 eV. The agreement which we observe supports this premise, which appears to be contradicted by the recent results of Register *et al* (1984). For example at 60 eV they propose a cross section for the direct excitation of the $1s_5$ state which is about a factor of five greater than that observed by Phillips *et al.* Whilst we note that this proposal is not supported by our results, it is also true

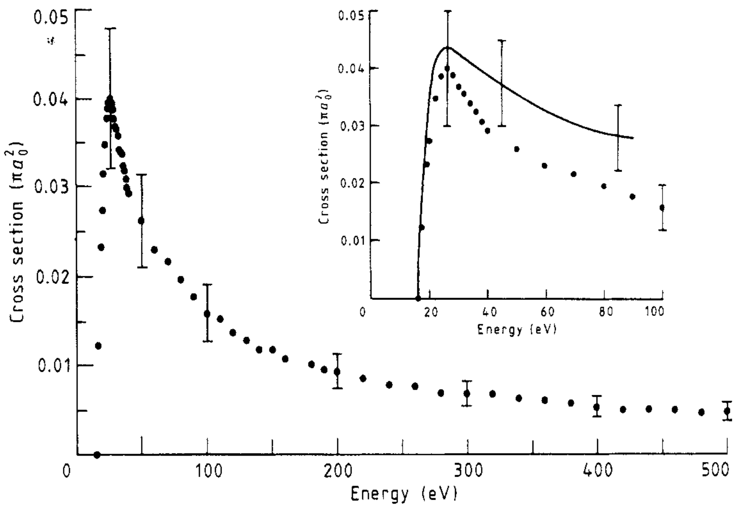

Figure 5. The total cross section for the production of metastable neon atoms as a function of the electron energy. The inset shows the present data (•) compared with those of Phillips *et al* (1981a, b) (——).

that Register *et al* (1984) have joined their data at 25, 30 and 50 eV in figure 9 by a smooth curve which in this case is misleading.

At energies close to threshold the contribution of cascade processes to our metastable signal is minimal. Therefore it is heartening to note that our cross section at 18 eV is only 10% less than that predicted by the R-matrix calculation of Taylor *et al* (1983). Acceptable agreement is found at 20 eV with the calculated cross sections of Machado *et al* (1984). The increasing importance of cascades at higher energies restricts a comparison with the 30 eV data of Machado *et al.*

The comparison between our results from total metastable production and the similar results of Phillips *et al* (1981a, b) shows that cascade processes dominate the production of metastable states in neon at energies in excess of about 40 eV. This mechanism is consistent with the premise that these states in neon are predominantly triplet and therefore can be excited by exchange scattering. Thus the *LS* coupling scheme offers a reasonable description of the excitation of these states.

Acknowledgments

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