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Multiple ionization of magnesium by electron impact

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Abstract. A pulsed crossed beam technique incorporating time-of-flight spectroscopy of the collision products, which was developed previously in this laboratory, has been successfully adapted to allow studies of multiple ionization of metallic atoms. Cross sections σ_n for the formation of $n = 1$-4 times ionized magnesium have been determined over the wide energy range 8-5300 eV by normalization to cross sections for single ionization of Mg recently measured by Freund *et al* using a fast crossed beam technique. Cross sections σ_2 are found to closely approach our values of σ_1 at our highest impact energies. Comparison is made with previous experiments based on different methods and, in the case of single ionization, results are compared with 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 ionization cross section of atomic hydrogen with high precision over a wide energy range. The technique is ideally suited to measurements with hydrogen atoms and other unstable species. However, in subsequent work with helium (Shah *et al* 1988) we showed that it can also be applied with advantage to studies of both single and multiple ionization of stable targets where the results of traditional methods sometimes exhibit serious discrepancies. The technique is also attractive for studies of ionization of metallic species where reliable measurements are not extensive. Apart from the fundamental interest, there is a need for accurate cross sections for both single and multiple ionization of many metallic species in the context of both fusion energy research and in astrophysics.

In the present work we have studied the electron impact ionization of magnesium. Cross sections σ_n for n times ionization of magnesium for $n = 1$-4 have been determined at energies within the range 8-5300 eV. For these measurements an oven source has been used to provide a ground state thermal energy beam of magnesium atoms. As in our previous work, short duration pulses of electrons are arranged to intersect the Mg atom beam in a high vacuum region. Immediately after the transit of each electron pulse through the beam, slow Mg^{n+} 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 multipler. Our method ensures a high and uniform collection efficiency for the product ions. It is also worth noting that, unlike most previous experimental approaches, the ionization takes place in the absence of both electric or magnetic fields thereby avoiding possible errors arising from distortion of primary electron beam trajectories.

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There have been a number of previous studies of the electron impact ionization of magnesium. Okudaira *et al* (1970) used a crossed beam technique in which the slow product ions were analysed using a magnetic field mass spectrometer. Cross sections σ_n for the production of $n = 1$-3 times ionized magnesium were determined within the range 12–1100 eV by normalization to the total ionization cross sections measured by Okuno *et al* (1970). In the latter experiment, a crossed beam arrangement was used in conjunction with the condenser plate method to determine the total slow ion and electron yields. The Mg atom beam density was determined by the deposition method in which the rate at which Mg atoms in the beam were deposited on a cooled surface was determined by atomic absorption spectroscopy. In similar measurements of the total ionization cross section by Vainshtein *et al* (1972) for impact energies up to 200 eV using a modulated crossed beam technique, the atom beam density was also determined by means of the deposition method.

Karstensen and Schneider (1978) have measured cross sections for single and double ionization of magnesium in the energy range 30–280 eV using a crossed beam technique. The electron beam was pulsed and the Mg^+ and Mg^{2+} product ions were separated by a time-of-flight technique. Light emitted from (4^1D-3^1P) decay of excited Mg atoms in the crossed beam region during the electron pulses provided a means of measuring the atom beam density.

In the most recent and perhaps most accurate of the previous measurements, Freund *et al* (1990) used a fast crossed beam technique in which a 3 keV Mg target beam intersected an electron beam. The Mg atom beam was produced by electron capture neutralization of Mg^+ in a gas target. The target species was chosen to maximize the formation of ground state Mg atoms through near-resonant electron capture. Cross sections for single ionization for electron impact energies up to 200 eV were determined by recording the yield of Mg^+ formed in the magnesium beam with a knowledge of the overlap between the electron and Mg beams. The Mg atom beam intensity was determined with high accuracy by the use of a pyroelectric detector calibrated with known ion beams.

2. Experimental approach

2.1. General description

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

A seven element electron gun was triggered by a pulse generator to provide electron pulses of 200 ns duration at a frequency of 5×10^4 pulses/s. The electrons intersected (at right angles) a beam of Mg atoms effusing from a specially developed oven source. The beam intersection region was maintained at a pressure of about 5×10^{-8} Torr.

Immediately after the transit of each pulse of electrons through the Mg atom beam, slow Mg^{n+} product ions formed by ionization were swept out of the beam intersection region by a pulsed electric field accelerated through a potential difference of 5 keV and then recorded as individual counts by a particle multiplier. The ion extraction field was produced by the application of pulses of about 20 V in amplitude and 2 μs in duration between a pair of high transparency grids located on either side of the beam intersection region.

The product Mg^{n+} ions were identified according to charge state n and distinguished from background gas product ions by their different times of flight to the multiplier in accordance with their charge to mass ratios. It was essential, as in our previous

work, to ensure that the product ions were extracted with high and equal efficiency irrespective of the primary beam energy. The extraction pulse generator was triggered by the main pulse generator (which triggered 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 target beam.

A time-to-amplitude converter operated with start pulses from the extraction pulse generator and with stop pulses from the particle multiplier provided time-of-flight spectra displayed on a multichannel analyser of the type shown in figure 1. The spectrum obtained at 1000 eV exhibits peaks corresponding to Mg^+, Mg^{2+}, Mg^{3+} and Mg^{4+} clearly resolved from background gas product ions.

2.2. The electron beam system

The seven element electron gun was the same as that described previously (Shah *et al* 1987). This utilized a V-shaped thoriated tungsten filament run at a temperature which provided pulsed beams equivalent to a current of about 10 nA in the continuous mode with very small angular divergence. As in our previous work, the final four elements in the gun were used to set the electron energy at values ranging from the ionization threshold up to 5300 eV while the beam intensity remained unchanged.

In the intersection region, the electron beam had a diameter of less than 2 mm while the diameter of the Mg beam was 4 mm. The electron beam current was recorded as a current to a screened Faraday cup. This signal was then digitized and fed into a microcomputer which also recorded the time-of-flight spectra.

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 mid-point of the V-shaped filament and d is

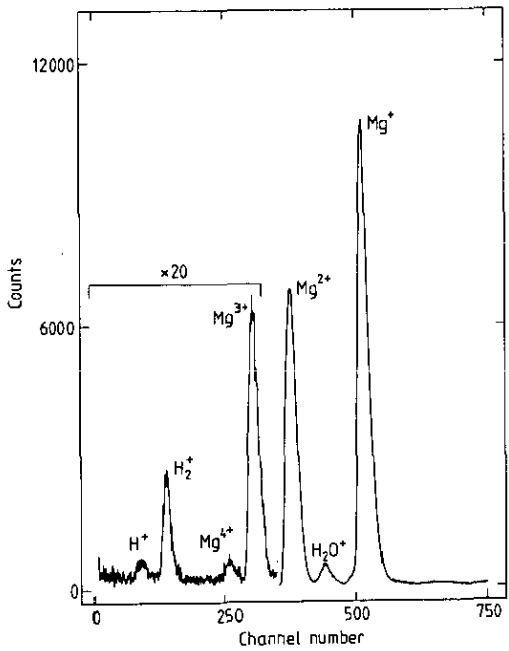

Figure 1. Time-of-flight spectra showing Mg^+, Mg^{2+}, Mg^{3+} and Mg^{4+} product ions observed in collisions of 1000 eV electrons with magnesium.

a correction parameter which allows for filament misalignment and the effect of contact potentials. A value of $d = 0.9 \pm 0.1$ V was obtained (figure 2) by extrapolation of the low energy data to the ionization threshold value of 7.64 eV for magnesium.

2.3. Magnesium oven source

Figure 3 shows a schematic diagram of the oven source which it was necessary to design to produce a reliable beam of Mg atoms of the required intensity.

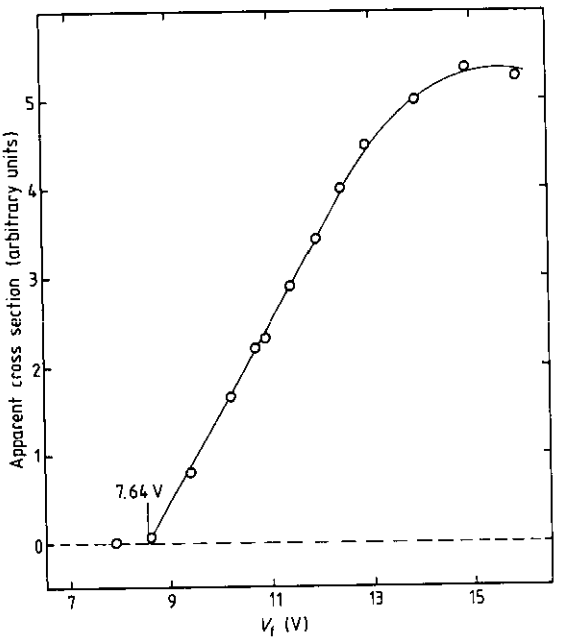

Figure 2. Dependence of Mg^+ yield on mid-filament potential V_f (see text) in collisions of electrons with magnesium. The ionization threshold of 7.64 eV is indicated.

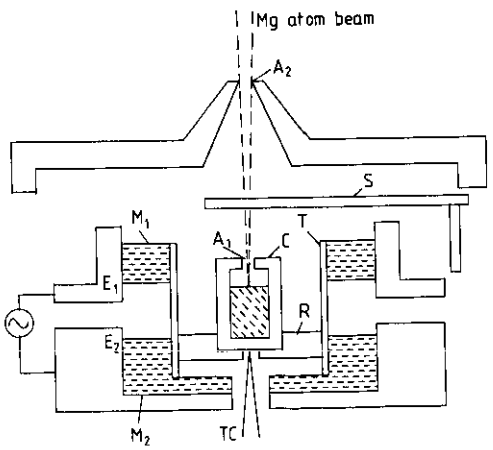

Figure 3. Schematic diagram of oven source of magnesium atoms.

A charge of about 5 g of magnesium was contained within a molybdenum crucible C which was supported by an aluminium oxide ring R within a heated tantalum tube T. The ends of the tube were held by molybdenum rings M_1 and M_2 which in turn were clamped within water cooled copper electrodes E_1 and E_2. By passing about 75 A AC through these electrodes the temperature of the tantalum tube could be raised so as to provide indirect heating of the molybdenum crucible and its contents. The temperature of the crucible was recorded by a thermocouple TC. A mechanically operated shutter S could be used to cut off the Mg beam when necessary. The magnesium atom beam emerged through a 2 mm diameter aperture A_1 in the crucible and passed via the 4 mm diameter defining aperture A_2 to the crossed beam region a distance of about 90 mm from A_1. The temperature of the oven was adjusted to provide estimated Mg atom beam densities between 10^{10} and 10^{12} atoms cm^{-3} in the crossed beam region. Operation could be sustained over a period of about 12 h.

2.4. Cross section measurements and normalization

At each particular electron impact energy the Mg^{n+} yields were determined from the average of several measurements of the area of the appropriate peak in the time-of-flight spectrum. Peaks corresponding to $n = 1$–4 could be recorded. In order to allow for any small changes in the atom beam intensity during the measurements, the Mg^+ yield per unit electron beam intensity was always measured relative to the corresponding yield at an impact energy of 100 eV.

Cross sections σ_n for n times ionization of magnesium were then obtained from the expression

$$\sigma_n = S_n / k\mu \tag{1}$$

where S_n is the Mg^{n+} yield per unit electron beam intensity and μ is the effective target thickness of the Mg atom beam. The constant k is the overall detection efficiency of the Mg^{n+} ions. A good indication of the independence of k on charge state n for the different ions was provided by observing the effect on recorded count rate when the potential difference through which the ions were accelerated was changed. The ratios of the double to single ionization and triple to single ionization count rates were found to be constant over a wide voltage range between about 5 and 7 keV.

In order to determine σ_n, the product $k\mu$ in equation (1) was determined by normalizing our relative cross sections for single ionization between 20 and 40 eV to the corresponding cross sections for single ionization recently measured by Freund *et al* (1990) using the fast crossed beam technique. The fast crossed beam method simplifies the problem of Mg atom density determination and the measurements of Freund *et al* (1990) seem very unlikely to be distorted by the presence of significant fractions of long lived excited Mg atoms. Normalization between 20 and 40 eV in the region of the σ_1 cross section maximum was considered appropriate since the effect of any small admixtures of metastable atoms on the measured cross sections by Freund *et al* (1990) would be smaller than at lower impact energies. In addition, our previous studies of the electron impact ionization of both helium (Shah *et al* 1988) and argon (McCallion *et al* 1992) have shown good agreement with measurements based on the fast crossed beam technique (Wetzel *et al* 1987, Freund *et al* 1990) in the range 20–40 eV. As noted in section 1, the beam deposition method has been used by some previous investigators to determine Mg atom beam densities. However, the uncertainties

Table 1. Cross sections σ_n for the production of n times ionized magnesium by electron impact.

Energy (eV)	σ_1 (10^{-16} cm^2)	σ_2 (10^{-17} cm^2)	σ_3 (10^{-19} cm^2)	σ_4 (10^{-20} cm^2)
7.7	0.072 ± 0.006			
8.1	0.421 ± 0.002			
8.4	0.800 ± 0.020			
9.2	1.66 ± 0.04			
9.7	2.21 ± 0.06			
9.9	2.32 ± 0.09			
10.4	2.90 ± 0.04			
10.9	3.43 ± 0.04			
11.4	4.00 ± 0.10			
11.9	4.49 ± 0.07			
12.9	5.00 ± 0.20			
13.9	5.36 ± 0.02			
14.9	5.26 ± 0.03			
17.4	5.47 ± 0.06			
19.9	5.28 ± 0.03			
22.4	5.16 ± 0.06			
24.9	5.00 ± 0.10	0.010 ± 0.001		
27.4	4.90 ± 0.10	0.036 ± 0.005		
29.9	4.82 ± 0.05	0.082 ± 0.010		
32.4	4.72 ± 0.03	0.114 ± 0.005		
34.9	4.64 ± 0.05	0.150 ± 0.020		
37.4	4.60 ± 0.10	0.172 ± 0.006		
39.9	4.41 ± 0.08	0.170 ± 0.020		
42.4	4.23 ± 0.07	0.190 ± 0.010		
44.9	4.18 ± 0.08	0.200 ± 0.020		
47.4	3.73 ± 0.02	0.200 ± 0.020		
49.9	3.66 ± 0.08	0.190 ± 0.020		
54.9	3.41 ± 0.04	0.200 ± 0.010		
59.9	3.31 ± 0.04	0.25 ± 0.02		
64.9	2.94 ± 0.04	0.30 ± 0.01		
67.4	2.84 ± 0.02	0.36 ± 0.01		
69.9	2.95 ± 0.02	0.39 ± 0.01		
74.9	2.73 ± 0.03	0.48 ± 0.02		
79.9	2.66 ± 0.03	0.57 ± 0.03		
84.9	2.51 ± 0.03	0.68 ± 0.03		
89.9	2.37 ± 0.02	0.77 ± 0.07		
94.9	2.35 ± 0.03	0.91 ± 0.03		
100	2.15 ± 0.02	0.97 ± 0.01		
105	2.10 ± 0.02	1.09 ± 0.02		
110	1.89 ± 0.05	1.11 ± 0.06		
115	1.92 ± 0.02	1.26 ± 0.05		
120	1.88 ± 0.04	1.40 ± 0.07		
130	1.87 ± 0.02	1.60 ± 0.05		
140	1.71 ± 0.02	1.70 ± 0.02		
150	1.65 ± 0.02	1.86 ± 0.02		
160	1.58 ± 0.03	1.98 ± 0.03	1.2 ± 0.3	
170	1.46 ± 0.02	2.05 ± 0.03	1.3 ± 0.5	
180	1.33 ± 0.02	2.01 ± 0.03	1.9 ± 0.5	
195	1.27 ± 0.02	2.12 ± 0.03	3.0 ± 0.3	
220	1.07 ± 0.01	2.12 ± 0.02	3.8 ± 0.3	
250	1.03 ± 0.01	2.36 ± 0.02	5.9 ± 0.4	
280	0.880 ± 0.010	2.28 ± 0.04	6.5 ± 0.8	

Table 1. (continued)

Energy (eV)	σ_1 (10^{-16} cm^2)	σ_2 (10^{-17} cm^2)	σ_3 (10^{-19} cm^2)	σ_4 (10^{-20} cm^2)
320	0.809 ± 0.005	2.38 ± 0.03	8.1 ± 0.3	
375	0.700 ± 0.020	2.40 ± 0.02	9.5 ± 0.1	1.6 ± 0.4
430	0.600 ± 0.009	2.26 ± 0.01	10.3 ± 0.2	3.0 ± 0.8
500	0.536 ± 0.006	2.24 ± 0.05	9.7 ± 0.3	3.6 ± 0.4
570	0.490 ± 0.010	2.17 ± 0.03	10.4 ± 0.2	4.6 ± 0.3
650	0.420 ± 0.010	2.03 ± 0.03	9.4 ± 0.4	5.0 ± 0.6
750	0.374 ± 0.004	1.92 ± 0.03	8.7 ± 0.5	4.5 ± 0.9
870	0.319 ± 0.002	1.76 ± 0.03	7.8 ± 0.3	4.2 ± 0.5
1000	0.309 ± 0.004	1.84 ± 0.02	7.7 ± 0.5	4.0 ± 1.0
1150	0.269 ± 0.006	1.67 ± 0.02	6.6 ± 0.3	4.2 ± 0.6
1320	0.220 ± 0.009	1.46 ± 0.02	5.5 ± 0.2	3.0 ± 0.7
1520	0.193 ± 0.002	1.37 ± 0.01	5.0 ± 0.3	3.3 ± 0.9
1750	0.166 ± 0.004	1.20 ± 0.01	4.2 ± 0.2	2.5 ± 0.3
2010	0.141 ± 0.005	1.06 ± 0.01	3.48 ± 0.05	2.3 ± 0.4
2300	0.132 ± 0.006	1.02 ± 0.01	3.30 ± 0.30	2.2 ± 0.6
2650	0.115 ± 0.002	0.93 ± 0.01	3.00 ± 0.20	2.2 ± 0.5
3000	0.102 ± 0.006	0.82 ± 0.01	2.50 ± 0.10	1.5 ± 0.4
3500	0.084 ± 0.003	0.70 ± 0.01	2.10 ± 0.20	1.5 ± 0.6
4000	0.079 ± 0.002	0.68 ± 0.01	1.92 ± 0.04	1.3 ± 0.2
4600	0.068 ± 0.003	0.61 ± 0.02	1.60 ± 0.10	
5300	0.055 ± 0.003	0.51 ± 0.01	1.40 ± 0.10	

in the sticking probability for Mg atoms on surfaces even at liquid nitrogen temperature seem very large. Vainshtein *et al* (1972) estimated a sticking probability of only 66%. Indeed studies of the deposition of magnesium on the cooled surfaces near our oven source provided clear evidence of multiple reflections. In the absence of an accurate alternative means of obtaining absolute cross sections, we believe that our normalization procedure is the most accurate method of calibration at the present time.

Our measured cross sections σ_n are shown in table 1. The uncertainties associated with individual cross sections are assessed at the 67% confidence level and reflect the degree of reproducibility of the values in terms of the various experimental parameters and statistical fluctuations. There are additional estimated uncertainties in absolute value of ±12% for σ_1 and ±13% for σ_2, σ_3 and σ_4.

3. Results and discussion

In figure 4 we show our measured cross sections σ_n for the production of up to $n = 4$ times ionized magnesium. Also included are the results of previous measurements. It is interesting to note that cross sections σ_2 for double ionization are approaching the corresponding cross sections for single ionization at our highest impact energies. In addition to the direct double ionization process, contributions to σ_2 are also believed to arise as a result of Auger transitions. In particular, the discontinuity evident in the σ_2 curve in the 40–60 eV region is believed to be (cf Peach 1970) due to the Auger transition $Mg^+ (2p^5 3s^2)^2P \rightarrow Mg^{2+}(2p^6)^1S + e$ following removal of a 2p electron at 55.8 eV. Cross sections σ_3 are nearly two orders of magnitude smaller than σ_1 while cross sections σ_4 are at least one order of magnitude less than σ_3. Signal to noise

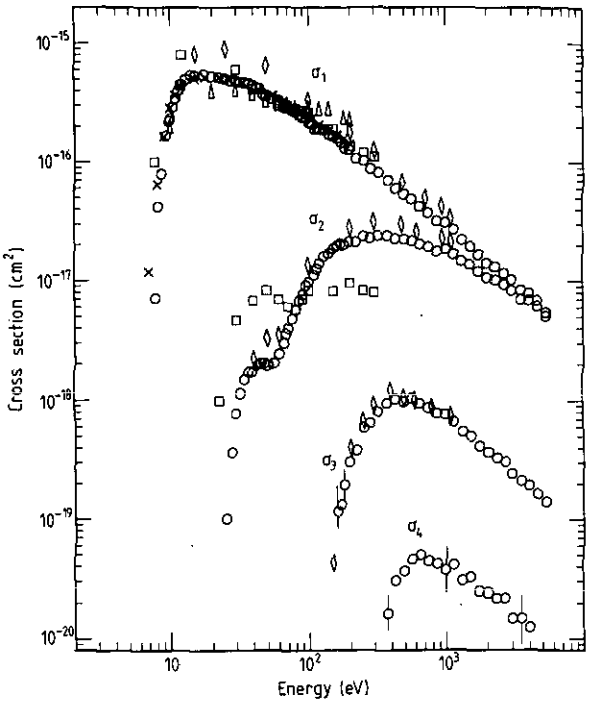

Figure 4. Cross sections σ_n for the production of n times ionized magnesium by electron impact. ○, σ_1, σ_2, σ_3 and σ_4 (present data); △, σ_1 (Vainshtein *et al* 1972); ◇, σ_1, σ_2, σ_3 (Okudaira *et al* 1970); ×, σ_1 (Freund *et al* 1990); □, σ_1, σ_2 (Karstensten and Schneider 1978).

considerations precluded an accurate study of the near threshold behaviour of the cross sections.

The single ionization cross sections measured by Freund *et al* (1990) to which our cross sections are normalized between 20 and 40 eV are in good accord with our measurements over the energy range 10 to 200 eV although it is evident that their values are becoming slightly higher with increasing energy. At 200 eV their cross section is 13% higher than our value. Below 10 eV, and particularly near threshold, there is a larger discrepancy.

The cross sections σ_1, σ_2 and σ_3 measured by Okudaira *et al* (1970) for energies up to 1200 eV may be compared with the present values. Their values of σ_1 are up to 1.8 times larger and exhibit greater scatter than our cross sections. Their values of σ_2 are also larger than our cross sections but also exhibit structure in the 40–60 eV region. For σ_3 the results of Okudaira *et al* (1970) are in reasonable agreement with our cross sections.

Cross sections σ_1 measured by Karstensen and Schneider (1978) at energies in the range 30–280 eV can be seen to be considerably larger than our values near the cross section peak. Their values of σ_2 are in poor general accord with our data in both magnitude and energy dependence indicating a problem in either measuring or calibration procedure.

The cross sections measured by Vainshtein *et al* (1972) for energies up to 200 eV, although strictly total cross sections, are dominated by the contribution from σ_1 and

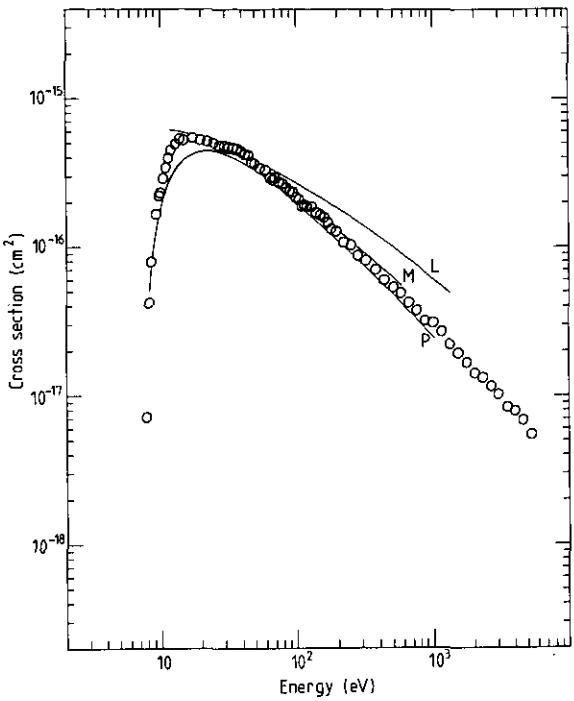

Figure 5. Cross section σ_1 for single ionization of magnesium by electron impact. O, present experimental data; P, corrected Ochkur approximation (Peach 1970); M, generalized oscillator strength approximation (McGuire 1977); L, classical-empirical formula (Lotz 1968).

can therefore be compared directly with our values. Their cross sections can be seen to exhibit a different energy dependence with good agreement only at energies near 60 eV.

Theoretical estimates have so far been confirmed to cross sections for single ionization and some predictions may be compared with the present values of σ_1 in figure 5. The results of Peach (1970) obtained using a corrected Ochkur approximation can be seen to be in good agreement with our cross sections only at energies near the ionization threshold. The calculations underestimate the peak value of the cross sections and predict high energy cross sections which fall more rapidly than those observed. More recent calculations by McGuire (1977) based on the generalized oscillator strength approximation provide cross sections which are in generally good agreement with our values over the range 15–1000 eV. We also include in figure 5 the results of the classical empirical calculations of Lotz (1968). In this case, the predicted energy dependence is in poor accord with our measurements. Good agreement in magnitude is apparent only at energies near 60 eV.

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