J. Phys. B: At. Mol. Phys. **15** (1982) 2539–2549. Printed in Great Britain

Total cross sections for electrons scattered from gases: 0.5–3.0 keV range on Ar

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Received 10 April 1981, in final form 14 April 1982

Abstract. Absolute total cross sections for electron scattering on Ar have been measured over the energy range of 0.5–3.0 keV. The correction due to the small-angle scattering was carried out using the theoretical differential elastic and inelastic cross sections. The corrected total cross sections agree well with the earlier experimental and theoretical results. The comparison with the high-energy Born theory is discussed.

1. Introduction

The total cross section for electron scattering on atoms and molecules has been of interest since 1930. Results prior to 1971 have been presented in the review of Bederson and Kieffer (1971), but are mainly electron–atom collisions in the low impact energy range. Currently, there is a considerable interest in determining cross sections for electrons colliding with atoms at intermediate energies. In recent review articles, Bransden and McDowell (1977) and Joachain *et al* (1977) discuss various theoretical approaches that are being used to calculate these intermediate-energy cross sections. It is well known that at low impact energies, exchange and polarisation effects are important in electron–atom collision phenomena. Nevertheless, at sufficiently high energies, only the static interaction will remain, with the result that the total scattering cross sections can be derived in the first Born approximation.

The Born approximation has been developed by several theoretical workers (Inokuti 1971, Inokuti *et al* 1975, Inokuti and McDowell 1974, Naon *et al* 1975) to estimate the high-energy limits of the total cross sections of electron scattering from atoms. A test of these theories requires the measurement of the total cross sections of electron–atom collisions at sufficiently high energies. However, for electron scattering from rare-gas atoms heavier than helium, the reported experimental cross sections (Wagenaar 1978, Wagenaar and de Heer 1980, Kauppila *et al* 1981) at high energies do not yet merge with the Born approximation calculation, thus indicating the need for measurements at even higher impact energies.

Another aspect of total scattering of electrons by atoms of considerable attention in recent years concerns the validity of the sum rule which is based on the forward dispersion relations of Gerjuoy and Krall. Here also, the total cross sections in the high-energy range are needed.

It is for these reasons that in the last few years our group has been interested in both theoretical and experimental studies of electron–atom and electron–molecule scattering in the intermediate- and high-energy electron impact ranges (Peixoto *et al* 1975, 1978, Peixoto and Nogueira 1976, Peixoto and Lee 1979, Lee and Freitas 1981a, b).

In this publication we present the measured total cross sections for electron scattering from argon in the incident electron energy range of 0.5 and 3.0 keV in intervals of 100 eV using the electron beam transmission method. There are no published experimental data for energies higher than 1.0 keV for electrons scattered from argon. The experimental values here reported are compared with semiempirical values of de Heer *et al* (1979). At energies below 800 eV, the experimental values are compared with results of Wagenaar and de Heer (1980) and Kauppila *et al* (1981). The sources of experimental errors are presented, together with the results and comparisons with the high-energy theoretical data and the validity of the Born approximation in the final section.

2. Experimental

The apparatus (figure 1), consists of an electron gun, a primary electron beam collector C_1, a scattering chamber and a transmitted electron beam collector C_2, mounted inside a vacuum chamber. The vacuum tank was pumped by a 6 in oil diffusion pump, equipped with a liquid nitrogen trap, chevron baffle and butterfly valve.

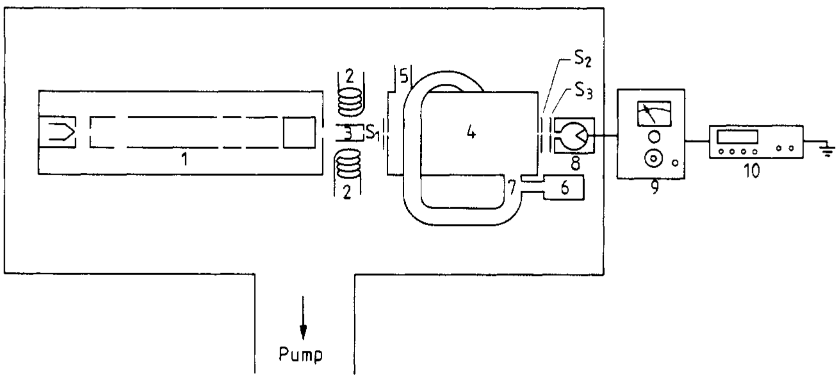

Figure 1. Schematic picture of the apparatus. 1, electron gun; 2, solenoids; 3, collector for the primary beam (C_1); 4, scattering chamber; 5, inlet for the gases; 6, Schulz–Phelps ion gauge; 7, outlet for the gases; 8, collector for the transmitted beam (C_2); 9, electrometer Keithley 602; 10, digital multimeter Keithley 173A. S_1, S_2, S_3 are apertures with diameters 1.0, 1.0 and 0.8 mm respectively.

The electron gun (1, in figure 1) which has an energy range between 20 and 3000 eV, is shielded by a sheet of μ-metal in order to minimise stray electric and magnetic fields.

The collector C_1 (3, in figure 1) is a Faraday cup, 20 mm long and with a diameter of 5 mm, with a hole at the bottom to allow the primary electron beam to pass through. From time to time a magnetic field is applied through a pair of coils (2, in figure 1), placed with their axes perpendicular to the electron beam trajectory, so that the primary beam can be completely deflected towards the collector. The magnitude of the current measured in this way was typically of the order of 10^{-7} A.

The scattering chamber (4, in figure 1) is a gas cell of length of 80 mm that can be made longer or shorter; in this experiment lengths of 80 and 130 mm were employed. The diameters of the apertures S_1 and S_2 are 1 mm.

The aperture S_3 in front of the collector C_2 (8, in figure 1) has a diameter of 0.8 mm and the distance between S_2 and S_3 is 20 mm. The angular resolution of the collector calculated from the centre of the scattering chamber ($L = 80$ mm) is 0.4°.

The transmitted beam is measured in the collector C_2 (8, in figure 1) which is connected to an electrometer (Keithley 602). A digital multimeter (Keithley 173A) is attached to the electrometer in order to minimise reading errors of the electrometer. With this arrangement the current could be recorded with three significant figures.

The gas enters the scattering chamber through an inlet (5, in figure 1) of diameter eight times the size of apertures S_1 and S_2. The outlet (7, in figure 1) has the same diameter as the inlet, and is connected to a tubular coil of diameter 8 mm and length 150 mm. The function of the outlet is to enable faster pumping of the scattering chamber, however the tubular coil kept the pressure inside the scattering chamber stable, by preventing a gas flow forming inside the scattering chamber. The gas pressure in the scattering chamber is measured by a Schulz–Phelps-type ion gauge (6, in figure 1), close to the outlet. After the ionisation gauge was calibrated by a MKS Baratron capacitance manometer in the whole working pressure range, i.e., from 15 to 70 mTorr.

The background pressure in the vacuum tank was of the order of 5×10^{-6} Torr. Argon was introduced in the scattering chamber to give a gas pressure up to 60 mTorr in the scattering region and the intensity of the transmitted electron beam was recorded for scattering chamber pressures varying from 20 to 50 mTorr. During the interval of the measurements the primary beam current in the collector C_1 (3, in figure 1) was monitored frequently.

3. Procedure

At the selected electron energies, the transmitted electron current was measured for pressures in the scattering chamber between 20 and 50 mTorr. The total cross section is obtained from the expression

$$I = I_0 \exp(-Nl\sigma) \tag{1}$$

or

$$\sigma = \ln(I_0/I)/Nl \tag{2}$$

where σ is the total cross section, I the transmitted current, and N the number of atoms in the scattering volume of length l. I_0 refers to the intensity of the primary beam current.

The total cross section is finally given by the slope of the straight-line plot of $\ln I$ against pressure (figure 2) multiplied by a constant factor. The multiplicative factor was obtained assuming the ideal-gas behaviour (a reasonable assumption at these pressures) and it is equal to $11.48/T(\mathrm{K})$. The temperature was always measured at the entrance and at the exit of the scattering chamber and was found to be 28 ± 1 °C.

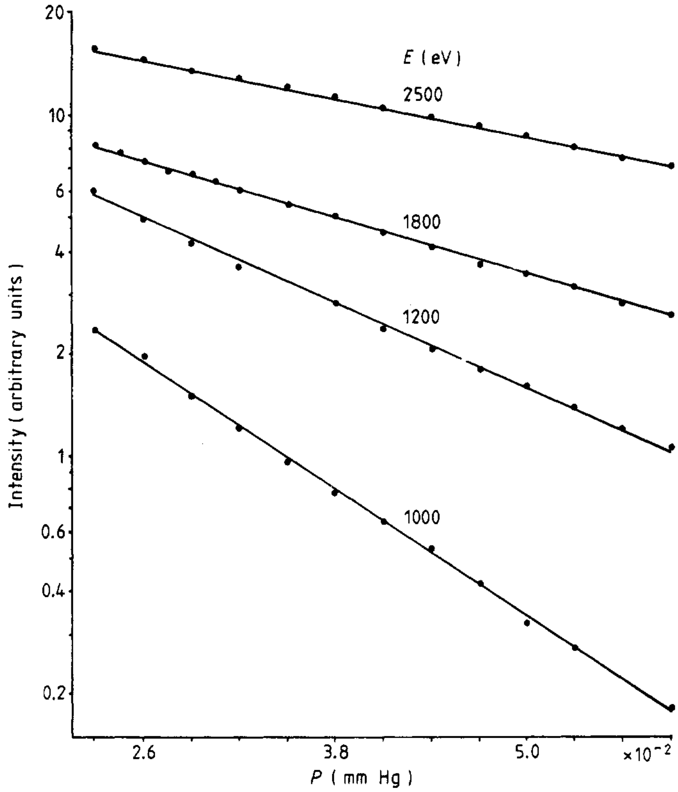

Figure 2. Intensity of the transmitted electron beam for energies (E) of 1000, 1200, 1800 and 2500 eV measured in the pressure range of 2.3 to 5.9 (10^{-2} mm Hg).

4. Sources of error

In this section the experimental systematic errors and the random statistical errors are discussed. The error introduced by the failure of the detector to discriminate between transmitted electrons and electrons scattered through small angles will be discussed in the next section.

The overall standard deviation of the total cross section is estimated from the experimental errors with the well known expression

$$\bar{d}\sigma = (\bar{d}\sigma^2)^{1/2} = \left[\left(\sum_{i=1}^{m} \frac{\partial\sigma}{\partial p_i}\Delta p_i\right)^2\right]^{1/2} \tag{3}$$

where p_i and Δp_i are, respectively, experimental parameters and their standard deviations. The derivatives $\partial\sigma/\partial p_i$ are analytically calculated from equation (2).

The electron source was switched on at least three hours before any actual measurements and fluctuations were verified by recording the intensity of the beam in the collector C_2 as a function of time. The largest observed fluctuation between the current recorded before the gas was introduced in the scattering chamber and after pumping it out following a set of measurements was 2%.

The ionisation gauge was calibrated against an MKS Baratron between 0.015 and 0.070 mm Hg. A straight line was obtained when the pressure measured by the ion gauge was plotted against the corresponding pressures obtained by the MKS Baratron. The deviation in the worst case was about 4%. The pressure was measured with an accuracy of ±0.5% and the gas used was of research grade quality, i.e., 99.99% purity. The FWHM distribution of the energy of the primary beam was 0.3 eV at 1 keV and 2.0 eV at 0.5 keV. Values of these experimental errors substituted in equation (3) give an overall standard deviation $\overline{\Delta\sigma}$ of 5% for the measured total cross sections.

A random error of 1%, which is attributed to uncertainties in the energy of the primary beam, magnetic fields and scattering by delocalised gases, is added giving the final total standard deviation of 6%.

In order to account for possible end effect problems, that is, the correct definition of the length over which the electron interacts with the electron beam, two sets of measurements were carried out with different gas cell lengths: in the first case the length was 80 mm, and in the second case 130 mm. The results obtained under these two condition sets were reproducible between the experimental error (6%).

5. Results

Our experimental values of the total cross sections for electron scattering from Ar between 0.5 and 3.0 keV are shown in column 1 of table 1 alongside the experimental measurements of Wagenaar (1978), Wagenaar and de Heer (1980) and Kauppila *et al* (1981). It can be observed that below 700 eV the agreement between our measured cross sections and those of Wagenaar (1978) and Wagenaar and de Heer (1980) is excellent, i.e. well within the experimental errors. However, the cross sections of Kauppila *et al* are about 10% lower than ours. Our measured cross sections also agree very well with the theoretical values of Joachain *et al* (1977) at impact energies below 1 keV. However, for incident energies higher than 1 keV, our experimental cross sections are found to be lower than the semiempirical results of de Heer *et al* (1979) i.e., 14% lower at 2000 eV and 19% lower at 3000 eV, showing a divergence at these high energies from the semiempirical formula derived by these authors.

This discrepancy can be attributed to the failure of our detector to discriminate between the transmitted beam current and the small-angle scattered electrons in our measurements. This effect, that always results in an underestimation of the total cross section, is worse at higher impact energies, when the elastic as well as the inelastic differential cross sections become more and more forward peaked. This could also explain this divergence from agreement at higher energies.

In order to minimise this systematic error, we introduced a correction factor to the total cross section given by

$$\sigma^{\text{corr}} = \frac{2\pi}{L} \int_0^L \mathrm{d}x \int_0^{2\tan^{-1}\{a/[2(L+S-x)]\}} \left(\frac{\mathrm{d}\sigma}{\mathrm{d}(\cos\theta)}\right)_{\text{tot}} \sin\theta \,\mathrm{d}\theta \tag{4}$$

where L is the length of the scattering chamber, in our case, $L = 80$ mm. S is the distance between the exit aperture S_2 and the aperture S_3 and is 20 mm. The diameter of the aperture S_3 is a and $\mathrm{d}\sigma/\mathrm{d}(\cos\theta)$ is the total differential cross section in the scattering angle θ, and is defined as

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}(\cos\theta)}\right)_{\text{tot}} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}(\cos\theta)}\right)_{\text{el}} + \left(\frac{\mathrm{d}\sigma}{\mathrm{d}(\cos\theta)}\right)_{\text{inel}}. \tag{5}$$

Table 1. Cross sections for electrons scattering from Ar.

E (eV)	σ (Å^2)	σ^1 ($=\sigma+\sigma^{corr}$) (Å^2)	Wagenaar (1978)	Wagenaar and de Heer (1980)	de Heer *et al* (1979)	Joachain *et al* (1977) I	Joachain *et al* (1977) II	Kauppila *et al* (1981)
500	3.76	3.88 (30)	3.59	3.66	3.50	3.84	3.75	3.31
600	3.35	3.48 (22)	3.21	3.28				2.95
700	3.09	3.23 (21)	2.91	2.97		3.08		2.72
800	2.89	3.03 (20)				2.82	2.80	2.54
900	2.70	2.85 (19)						
1000	2.51	2.66 (18)			2.23	2.42	2.42	
1100	2.25	2.39 (16)						
1200	2.00	2.15 (15)						
1300	1.81	1.96 (14)						
1400	1.68	1.83 (13)						
1500	1.59	1.74 (13)						
1600	1.49	1.64 (12)						
1700	1.41	1.56 (11)						
1800	1.32	1.47 (11)						
1900	1.26	1.40 (10)						
2000	1.19	1.33 (10)			1.35			
2100	1.12	1.26 (10)						
2200	1.07	1.21 (9)						
2300	1.01	1.15 (9)						
2400	0.963	1.10 (9)						
2500	0.919	1.06 (8)						
2600	0.882	1.02 (8)						
2700	0.853	0.99 (8)						
2800	0.823	0.96 (7)						
2900	0.801	0.94 (7)						
3000	0.786	0.92 (7)			0.972			

In this expression $(\mathrm{d}\sigma/\mathrm{d}(\cos\theta))_{\mathrm{el}}$ are elastic differential cross sections and $(\mathrm{d}\sigma/\mathrm{d}(\cos\theta))_{\mathrm{inel}}$ are total inelastic differential cross sections for electrons scattered by argon.

The total inelastic differential cross sections were obtained from the well known expression (Naon *et al* 1975)

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}(\cos\theta)}\right)_{\mathrm{inel}} = \frac{4}{K^4} S_{\mathrm{inc}}(K) \tag{6}$$

where K is the magnitude of the momentum transfer vector and $S_{\mathrm{inc}}(K)$ is the incoherent scattering function. The $S_{\mathrm{inc}}(K)$ used in this work has been calculated by Naon *et al* (1975) using the Bethe–Goldstone method. The zero-angle inelastic scattering thus obtained by Inokuti (1971) is

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}(\cos\theta)}\right)_{\mathrm{inel},\theta=0} \simeq 16k^2 S(-3) \tag{7}$$

where $S(-3)$ for argon is 3.196 (Dehmer *et al* 1975) and k is the incident energy in rydberg units.

The elastic differential cross sections employed in this work for correction purposes were obtained from the calculations of McCarthy *et al* (1977) who used the optical model.

Corrections were carried out at four energies: 0.8, 1.0, 2.0 and 3.0 keV. For the data taken at 800 eV, corrections were made using the elastic differential cross sections given by McCarthy *et al* (1977) at 750 eV. This is reasonable since it was estimated that the error caused by this replacement was less than 1% in the correction factor σ^{corr} of the total cross section, because it was observed from the calculations that the main contribution to this correction was the inelastic scattering correction which contributed more than 90% of the total correction.

The double integration in equation (4) is solved by a Simpson quadrature. An interpolation method has been employed to obtain the total differential cross sections in a regular grid. Further, for scattering angles less than 5° the elastic differential cross sections were obtained by fitting the logarithm of the cross section as a linear function of the magnitude of the momentum transfer vector.

For incident energies different from these four cases, the correction of the total cross section was obtained by standard interpolation techniques. We estimate the uncertainty to be 20% in the correction part of the total cross section σ^{corr}, due to the interpolation procedure and the general random errors in the numerical procedure. Nevertheless, this error of 20% in σ^{corr} reflects only 4% in the corrected total cross section in the limiting case. The corrected total cross section, that is, $\sigma' = \sigma^{\text{exp}} + \sigma^{\text{corr}}$, of the electron scattering by argon are presented together with associated errors in column 2 of table 1. These errors are obtained by the sum of the experimental deviation (6%) and an estimated error of 20% in the corrected portion of the total cross section ($\sigma_{\text{tot}}^{\text{corr}}$) at each incident energy. From this it is observed that the agreement between our corrected total cross section and other theoretical and experimental results is good. At incident energies below 800 eV, the corrected total cross sections agree with the experimental results of Wagenaar (1978) and Wagenaar and de Heer (1980) to within 10%. Our results are higher than those of Kauppila *et al* (1981), by about 10%. The comparison with the theoretical results of Joachain *et al* (1977) also shows good agreement. For incident energies higher than 1 keV, our corrected total cross sections agree well with the semiempirical results of de Heer *et al* (1979).

It is important to point out that the main contribution to the correction factor of the total cross section comes from the inelastic scattering; the elastic scattering contributes less than 7% throughout the whole energy range. This fact shows that very accurate total cross sections can be obtained on applying a retarding potential field in front of the detector to eliminate the inelastic small-angle scattered electrons.

6. Comparison with the high-energy-limit theories

At high incident energies the limit of the total cross section of the electron scattered by atoms is well known. The total elastic cross section can be obtained from the Born approximation (Inokuti and McDowell 1974) as

$$\sigma_{\text{el}} = \pi k^{-2}[A + (B + B_{\text{ex}})k^{-2} + (k^{-4} + \ldots)] \tag{8}$$

where

$$A = 8 \int_0^\infty |Z - F(K)|^2 K^{-3}\, \mathrm{d}K. \tag{9}$$

In this expression, K is the magnitude of the momentum transfer vector, Z is the atomic number, $F(K)$ is the x-ray form factor and

$$B = -Z^2. \tag{9a}$$

B_{ex} is derived from the Ockhur–Born exchange approximation (Bonham 1978) and is equal to

$$B_{ex} = \int_0^{2k} \frac{dK}{K} |Z - F(K)| F(K) \tag{9b}$$

and

$$C = 0. \tag{9c}$$

In this work, the parameter A is obtained from equation (9), where the $F(K)$ used were the Hartree–Fock form factor of Tavard *et al* (1967). The parameter A obtained in this procedure equals 244.02 au.

The total cross sections for inelastic scattering of electrons by atoms can be written as

$$\sigma_{inel} = 4\pi\left(\frac{R}{T}\right) |M_{tot}^2 \ln(4C_{tot}TR^{-1}) + \ldots| \tag{10}$$

where M_{tot}^2 is the modulus squared of the total dipole matrix element. The term related to the parameter C_{tot} is given as

$$M_{tot}^2 \ln C_{tot} = -2L(-1) + I_1 - I_2. \tag{11}$$

Using published data from the literature (Naon *et al* 1975, Inokuti *et al* 1975) for all the quantities in equations (10) and (11) we determined the expression for the total inelastic cross section in the high-energy limit to be

$$\sigma_{inel} = \frac{4\pi}{k^2}(4.1767 + 4.268 \ln k^2) \tag{12}$$

in atomic units.

The comparison between our corrected total cross section and the cross sections obtained from the high-energy-limit theory with and without the exchange correction is illustrated in figure 3. One can see that the theoretical data is always higher than our cross sections throughout the entire energy range. As expected, the contribution of the exchange effect to the total cross sections is not important for the incident energies considered in this work. At 500 eV the theoretical value is higher than our results by about a factor of two. Although the difference diminishs with increasing incident energy, the theoretical cross section is still higher than our corrected cross section by approximately 30% at 3000 eV.

In an attempt to explain this discrepancy, we summed the total inelastic cross section defined in equation (10) with the total elastic cross sections which are obtained from numerical integration of the differential elastic cross sections in the optical model (McCarthy *et al* 1977). The results for 1000, 2000 and 3000 eV are shown in figure 3. It can be observed that the agreement between this set of cross sections and our corrected results is significantly better than for previous comparisons. On the other hand, we have also obtained the total elastic cross sections by the partial-wave method instead of the Born approximation of equation (8). These PW total elastic cross sections

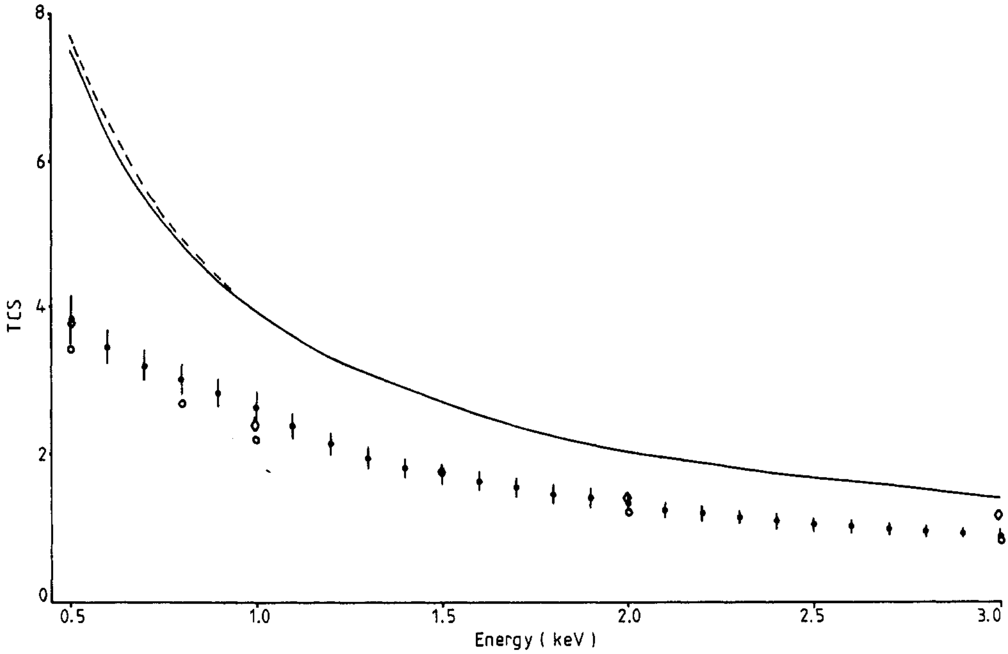

Figure 3. Total cross sections (TCS) for 0.5 to 3.0 keV electrons scattered from Ar. Comparison with the high-energy-limit theory. ♦♦♦, present measurements corrected for undiscriminated detection of small-angle scattered electrons; full curve, Born–Bethe theory without exchange correction; broken curve, Born–Bethe theory with exchange correction; calculated values of total cross sections from summation of total inelastic cross sections (given by equation (12)) with total elastic cross sections from numerical integration of optical model differential cross sections (McCarthy *et al* 1977) (open circles) and with the total elastic cross sections from the partial-wave method (crosses).

are obtained by the well known optical theorem

$$\sigma_{\mathrm{el}}^{\mathrm{tot}} = \frac{4\pi}{k_i} \operatorname{Im} f(0) \tag{13}$$

where the $f(0)$ is the partial-wave zero-angle scattering amplitude which is obtained in the static interaction potential field. For incident energies of 0.5, 1.0 and 1.5 keV, the $f(0)$ used in this work was calculated by Fink and Yates (1970). At higher energies we have calculated the PW zero-angle scattering amplitude by the numerical integration method. The total cross sections obtained by the sum of the PW $\sigma_{\mathrm{el}}^{\mathrm{tot}}$ and the $\sigma_{\mathrm{inel}}^{\mathrm{tot}}$ of equation (10) are also shown in figure 3. One can see that the agreement between this set of theoretical data and our corrected cross sections is good. This strongly implies that the high-energy limit (Born region) for elastic scattering is not reached even at 3 keV incident energy. In contrast, this limit has been reached for inelastic scattering at incident energies around 1 keV.

7. Conclusions

To summarise, we have measured the absolute total cross section of the electron scattering by argon atoms in the energy range of 500 eV–3 keV. Below 800 eV, our measured cross sections agree very well with the theoretical and semiempirical data in this energy range. At higher energies, our measurements are markedly lower than the semiempirical results of de Heer *et al* (1979). However, when the correction for the small-angle electron scattering is included, the corrected cross sections agree well with the experimental and semiempirical results throughout the entire energy range. Comparison of our data with the high-energy-limit theoretical data shows that this limit for the Born region is not reached even at 3 keV.

This study also reveals that the small-angle scattering significantly affects the measurement of total cross sections, especially in the high impact energy region. Below 800 eV, this effect of small-angle scattering on the total cross section is only a few per cent, however this increases to approximately 20% at 3 keV. As the main contributors to the magnitude of small-angle scattering are inelastic collision processes, we expect that very accurate experimental cross sections can be obtained by introducing a retarding potential field in front of the detector. This feature is at present under investigation.

Acknowledgments

The authors would especially like to thank Professor Eduardo M A Peixoto for use of experimental facilities, Professor José M Riveros for the use of the MKS Baratron micromanometer and for many fruitful discussions, and one of the referees for suggesting the small-angle correction procedure.

This work was supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

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