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Scattering of electrons by CH_4, CF_4 and SF_6 in the 75–700 eV range

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Abstract. Differential cross sections (DCS) have been measured by the crossed-beam method. The angular range is 5–135° and the electron energies are 75, 100, 150, 200, 300, 500 and 700 eV. The overall energy resolution is not sufficient to discriminate elastic scatterings from rotational and vibrational excitations. The absolute DCS are determined by using a gas chamber on the basis of the known absolute DCS for He. The experimental DCS are extrapolated to 0° and 180° scattering angles by fitting the square of the Legendre polynomials to the measured values. The integral and momentum transfer cross sections are then obtained. The errors are about 10% for all experimental results. The present data for CH_4 agree well with the previous measurements and calculations. For CF_4 and SF_6, the DCS increase steeply at the forward angles. The integral cross sections obtained are consistent with the previous results for total and inelastic cross sections.

1. Introduction

Accurate measurements of electron impact cross sections for the molecular gases CH_4, CF_4 and SF_6 are required for the practical application of these gases and the verification of theoretical scattering models. Total cross sections for them have been measured by many workers using the transmission method (for example, Lohmann and Buckman 1986, Jones 1985, 1986, Kennerly *et al* 1979), and momentum transfer cross sections in the very low-energy region have been estimated from the results of swarm experiments (for example, McCorkle *et al* 1978, Hayashi and Nimura 1984). Measurements of differential cross sections (DCS) for elastic and inelastic scattering, however, are scarce compared with the abundance of the above measurements. Experimental and theoretical works which have been carried out until now have been summarised by Trajmar *et al* (1983) and by Gianturco and Jain (1986). In most works on DCS, the impact energy of electrons is below 20 eV. In the higher-energy region, there are few experimental works (for example, Vuskovic and Trajmar 1983, Trajmar *et al* 1983) and inconsistencies remain in the absolute values of cross sections between the various results. Systematic studies of the gases are required over a broad energy range.

The CH_4 molecule is the simplest organic compound; it is used as an atmospheric gas in diffuse discharge switches and as a filling gas in proportional counters for detecting nuclear radiation. Several works have recently been reported for elastic and inelastic electron scattering by CH_4 in the energy range below 20 eV. These works have been tabulated by Curry *et al* (1985). CF_4 gas is used in plasma etching for semiconductors. We have no knowledge of measurements of DCS of electron elastic

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scattering for CF_4. SF_6 is used widely as an insulation gas in high-voltage apparatus. DCS have been reported for 0.3–10 eV by Rohr (1979) and for 5–75 eV by Srivastava *et al* (1978, and renormalised by Trajmar *et al* 1983). Inconsistencies have been pointed out by Hayashi and Nimura (1984) between the total cross section and the sum of elastic and inelastic cross sections.

In this paper, the absolute values of DCS for elastic electron scattering have been obtained experimentally for CH_4, CF_4 and SF_6. From these data, the integral and momentum transfer cross sections are estimated.

2. Experimental apparatus and procedure

Descriptions of the apparatus and experimental procedure have been given in a previous paper (Katase *et al* 1986). Therefore, they are only briefly described here.

In the measurements of the relative DCS, the crossed-beam method was used. Electrons of a definite energy were fired from an electron gun with a tungsten filament of the hairpin type and crossed a beam of target molecules. The molecular beam was formed with a multicapillary array (MCA) at the centre of a vacuum chamber. Electrons scattered by the molecules were analysed in energy with a cylindrical mirror analyser (CMA), which could be moved around the MCA and prevented electrons from exciting the molecules to electronically higher states. Electrons losing the energy corresponding to vibrational or rotational excitations were not discriminated because of an insufficient energy resolution of 1 eV at a 100 eV electron energy. Another CMA (M-CMA) was fixed at a scattering angle of $-40°$ to compensate the counting rate of the CMA for the fluctuations in the electron beam current and the target density. The counting rate of the M-CMA was proportional to the effective integral of the product of the incident electron flux density and the target molecule density over the interaction region. The CMA signals were counted, at every measurement, for a time period defined so that the number of counts of the M-CMA reached a constant preset value. Therefore, the fluctuations of both densities were automatically corrected. The angular distribution of scattered electrons at a specified electron impact energy was measured automatically by a control system with a microcomputer. The angular distribution of the background count was measured in the same way without the molecular beam. That is, electrons were detected after being scattered by the static gas at the same pressure as in the above measurement. The relative DCS were obtained from the angular distribution after the subtraction of background. The angular dependence of the size of the interaction region was used to correct the relative DCS in the same way as described in our previous paper (Katase *et al* 1986).

The relative DCS obtained were converted to absolute values at 30° by using the ratio of the elastic DCS of the target gases to that of He and the absolute values of the latter. They were given by Jansen *et al* (1976) for electron energies of 100–700 eV and Register *et al* (1980) for 75 eV. A gas chamber was used to obtain the ratio. The schematic diagram of the experimental arrangement using the gas chamber is shown in figure 1. In order to obtain the ratio, the counting rates of elastically scattered electrons for the gases were measured for a definite value of the integrated electron current as a function of absolute pressure in the gas chamber. The electron counting rate per unit absolute pressure f was expressed as an equation

$$f = C/(P + \mathrm{const}) \qquad (1)$$

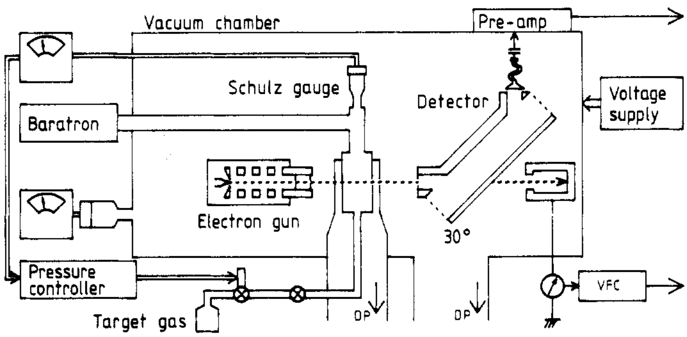

Figure 1. Schematic diagram of the experimental equipment for measuring the ratio of the elastic DCS of a molecular gas to that of He.

where C is the counting rate and P is the absolute pressure in the gas chamber. The value of f was obtained by the least-squares method, fitting equation (1) to experimental results. The value of f for He was denoted by f_{He}. An MKS Baratron gauge was used to measure the pressure in the chamber for the estimation of f_{He}. The resolution of this gauge was about 10^{-2} Pa. Since the DCS for the gas molecules were larger than those for He, the chamber pressure was made lower in the measurement of f for gas molecules than for He. The pressure was then measured by a Schulz-type ionisation gauge, which was calibrated for the gases with the Baratron gauge at a higher pressure of around 1 Pa. Descriptions of the calibration were given by Sakae *et al* (1987).

By using the values of the ratio f/f_{He}, the absolute DCS for the target gases at 30° (σ_{30}) were obtained as follows:

$$\sigma_{30} = (f/f_{He})\sigma_{RJ30} \tag{2}$$

where σ_{RJ30} was the absolute DCS at 30° for He obtained by Jansen *et al* or Register *et al.* Figure 2 shows the absolute DCS at 30° determined for all target gases as a function of electron impact energy.

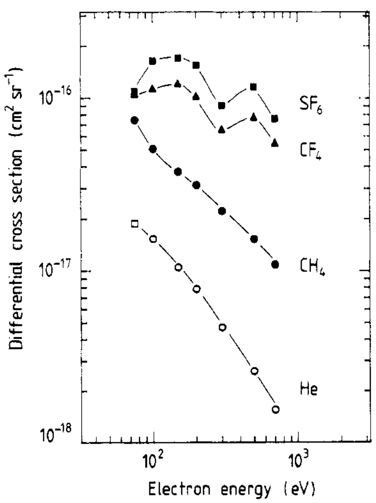

Figure 2. Absolute values of DCS for elastic scattering of electrons at a scattering angle of 30°. The open square and the open circles are the experimental results of Register *et al* (1980) and Jansen *et al* (1976), respectively.

The relative DCS were extrapolated to the forward and backward angles by fitting the square of the Legendre polynomials to the experimental values. The fitting was performed using the least-squares method. The integral and momentum transfer cross sections were obtained by integrating the relative DCS from scattering angles of 0° to 180° and normalising to the absolute values.

3. Estimation of errors in the cross sections

The errors in the absolute values of DCS orginate from several sources. They are classified into two types of errors: relative errors, which occur in the measurements of the angular distribution, and calibration errors, which arise in the process of conversion from the relative elastic DCS to the absolute values. All errors estimated are standard deviations.

(*a*) Relative errors. The relative errors of DCS originate from statistical deviations in the counting of pulses from the CMA and the M-CMA. Statistical errors in the background count add to the above errors. Since the counting was stopped after an accumulation of at least 2500 counts with the CMA and M-CMA, the statistical fluctuation of counts is less than 2% for both CMA. The ratio of the true counts when the gas beam effuses from the MCA to the background counts is about 20. The effective error due to the background counts is about 2% at most in the relative DCS. The combined error of the above three values is about 3.5% for the relative values of DCS.

(*b*) Calibration errors of DCS at a fixed angle of 30°. The errors of the absolute DCS are 6.5–6% for He at 30° as described in the references. These are the principal sources of error in the present absolute values of DCS. In one cycle of the measurement for the calibration, the fluctuation of the measured pressure is so small that the statistical error of the counting rate is dominant in the deviation of the fit by the least-squares method using equation (1). The statistical errors are evaluated to be about 4% in f/f_{He}, which is the ratio of the DCS for the molecular gases to that for He. The error of the absolute pressure in the gas chamber is about 5%. This is caused by the deviation of the indication of the Schulz-type ionisation gauge between cycles of measurements. Calibration errors are estimated to be about 9% from these error sources.

A summary of all relevant errors is shown in table 1. The overall errors of the absolute DCS include the calibration errors of DCS at 30°, the relative errors of DCS at 30° and the relative errors of DCS at every specified angle. These absolute errors are calculated to be about 10%.

Table 1. Errors in the measured DCS.

	Sources	Errors (%)
Relative errors	Counting of CMA	<2
	Counting of M-CMA	<2
	Background counting	<2
Calibration errors	Absolute DCS of He	6–6.5
	Ratio of DCS (30°)	4
	Pressure	5
Overall error		10

The integral and momentum transfer cross sections also comprise two types of errors. One of them is propagated from the relative errors of the DCS through the integration. The other one is generated from the calibration of the DCS at 30°. The overall errors of the integral and momentum transfer cross sections are estimated to be about 10%. The calibration errors are the main source of error.

4. Results and discussion

The experimental results for the DCS are plotted in figures 3, 4 and 5 for CH_4, CF_4 and SF_6, respectively†. These are data for the elastic scattering including the vibrational and rotational excitations. Full curves show the fitted polynomials as described in § 2.

The present results for CH_4 are compared with previous experiments in figure 6. The relative experimental data of Arnot (1931) for 205 eV and of Hughes and McMillen (1983) for 100 and 300 eV are normalised to the present value at an angle of 30°. The data of Vuskovic and Trajmar (1983) for 200 eV, which were calibrated to theoretical values in their paper, are shown in the figure. These data are similarly normalised to the present value. Only the data of Oda (cited by Dhal *et al* 1979) for 500 eV are absolute ones in the comparison of these cross sections in the figure. There is good agreement between the present results and the previous values except the cross sections for the backward scattering at 200 eV and the forward scattering at 500 eV. This agreement with the values obtained with good energy resolution means that the vibrational excitations are not dominant in the present results. Comparisons with the recent calculations by Jain (1986) are also shown in figure 6. In shape, the present data resemble his calculations (full curves), where the optical potential is real and inelastic channels are not considered. The differences between the present results and the theoretical curve of pure elastic DCS (broken curves) are considered to express principally the rotational excitation cross sections.

The integral and momentum transfer cross sections are listed in tables 2 and 3, respectively, for three gases. They are also plotted in figures 7, 8 and 9 with the results of previous works. For integral cross sections of CH_4, the present data are considered to be reasonable in comparison with those of Tanaka *et al* (1982). Present values are, however, smaller by 50% than those of Vuskovic and Trajmar (1983) at 200 eV. Present values are fairly large in the high-energy region of 300 eV compared with the values estimated by Floeder *et al* (1985) from the total and inelastic cross sections. The theoretical values of integral cross sections by Jain (1986) agree well with the present results. The disagreement in the momentum transfer cross sections is considered to be caused by the increase of backward scattering by the rotational excitation included in the experimental data.

For CF_4, no previous measurements of DCS are known to us, for comparison. The present results for integral elastic cross sections are considered to be reasonable in comparison with previous works on total cross sections by Mori *et al* (1985) and Jones (1986).

The present integral cross section for SF_6 at a 75 eV is about three times as large as that of Srivastava *et al* (1976) as shown in figure 9. This deviation is due to the differences in the forward DCS as shown in figure 10. These disagreements are less marked for the momentum transfer cross section than for the integral cross section.

† Tables of the present results will be mailed upon request.

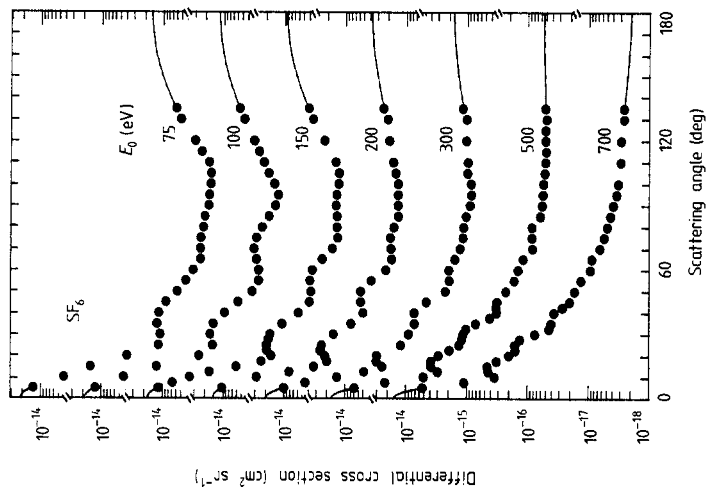

Figure 5. DCS for elastic scattering of electrons by SF_6. The full curves are those extrapolated by fitting the polynomials to the experimental data.

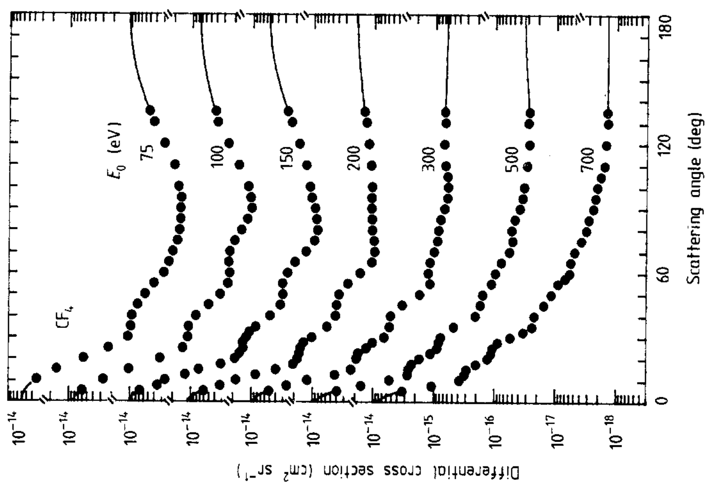

Figure 4. DCS for elastic scattering of electrons by CF_4. The full curves are those extrapolated by fitting the polynomials to the experimental data.

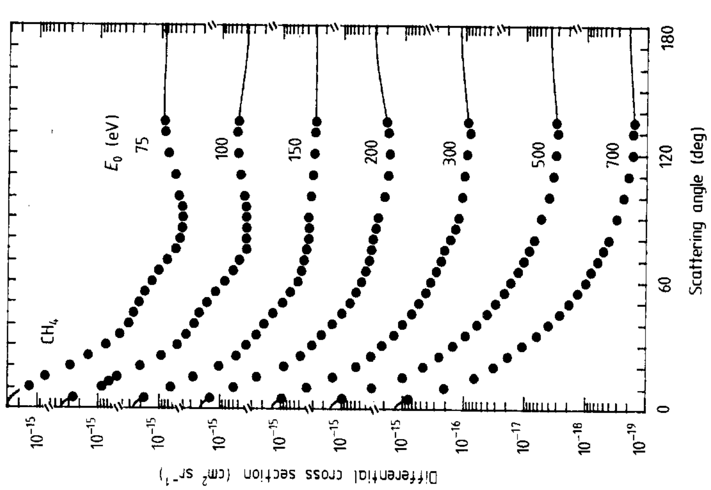

Figure 3. DCS for elastic scattering of electrons by CH_4. The full curves are those extrapolated by fitting the polynomials to the experimental data.

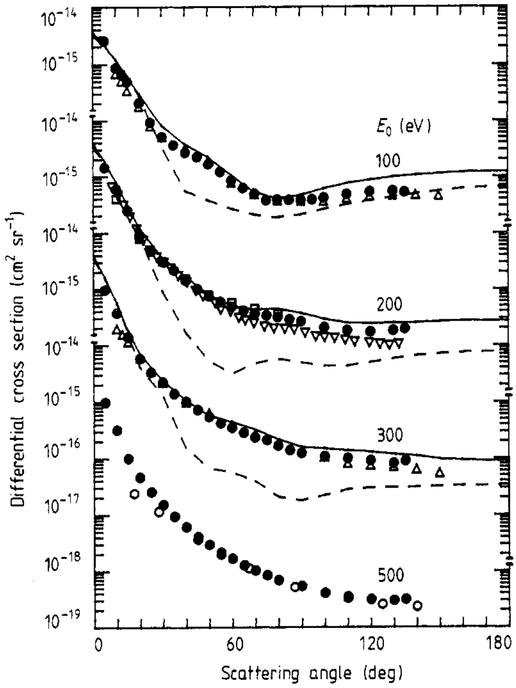

Figure 6. Comparisons of the present results (●) for CH_4 with previous exeriments and calculations. The plotted data are by Hughes and McMillen (1933) (△), Arnot (1931) (□), Vuskovic and Trajmar (1983) (▽) and Oda (cited by Dhal *et al* 1979) (○). The curves are calculations by Jain (1986). The full curves are obtained without consideration of inelastic channels, and the broken curves are the pure elastic DCS.

Table 2. Integral cross sections for elastic scattering of electrons in units of 10^{-16} cm^2. Errors are shown in brackets in per cent.

	Electron energy (eV)						
	75	100	150	200	300	500	700
CH_4	6.03 (9)	4.59 (10)	3.01 (10)	2.56 (9)	1.63 (9)	1.33 (10)	0.967 (10)
CF_4	14.7 (9)	12.2 (9)	10.8 (9)	8.56 (9)	6.46 (9)	4.68 (9)	3.90 (9)
SF_6	20.0 (9)	18.7 (10)	15.6 (9)	13.1 (9)	10.6 (9)	7.59 (8)	6.02 (8)

Table 3. Momentum transfer cross sections for elastic scattering of electrons in units of 10^{-16} cm^2. Errors are shown in brackets in per cent.

	Electron energy (eV)						
	75	100	150	200	300	500	700
CH_4	1.15 (8)	0.787 (10)	0.526 (9)	0.399 (9)	0.223 (9)	0.107 (9)	0.0663 (9)
CF_4	5.28 (9)	4.16 (9)	2.99 (8)	1.92 (8)	1.13 (8)	0.632 (8)	0.418 (9)
SF_6	6.42 (9)	5.70 (9)	4.37 (8)	2.98 (8)	1.76 (9)	1.02 (8)	0.655 (8)

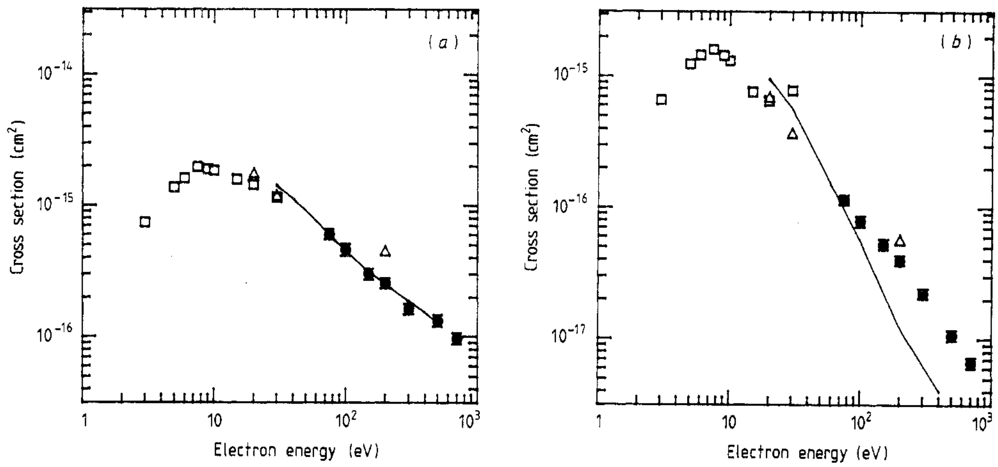

Figure 7. Integral (*a*) and momentum transfer (*b*) cross sections for elastic scattering of electrons by CH_4 as a function of electron energy. The full circles with error bars are the present results. Open squares and triangles are the data of Tanaka *et al* (1982) and Vuskovic and Trajmar (1983), respectively. The full curves are theoretical results by Jain (1986).

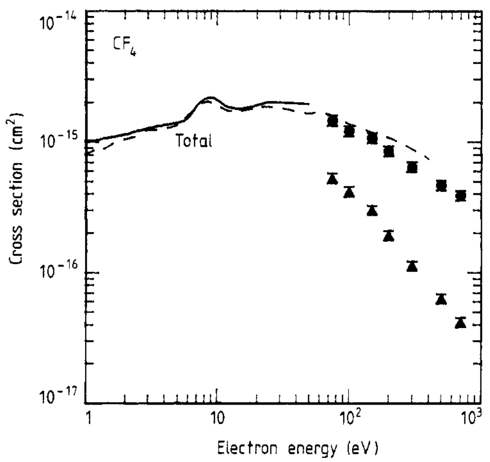

Figure 8. Integral (●) and momentum transfer (▲) cross sections for elastic scattering by CF_4 as a function of electron energy. The total cross sections by Jones (1986) and Mori *et al* (1985) are shown as a full curve and a broken curve, respectively.

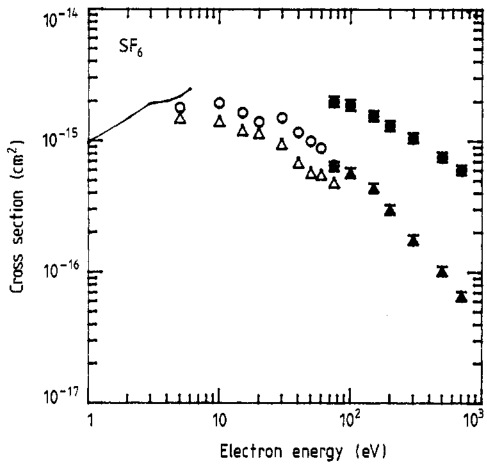

Figure 9. Integral (●) and momentum transfer (▲) cross sections for elastic scattering by SF_6 as a function of electron energy. The data measured by Srivastava *et al* (1983) are shown for integral (○) and momentum transfer (△) cross section. The full curve is the experimental result for the integral elastic cross section by Rohr (1979).

The sum of the present integral elastic cross section and the inelastic cross sections used by Hayashi and Nimura (1984) becomes about two times as large as that estimated by them. This value is reasonable in comparison with the data of Kauppila *et al* (1983) and Kennerly *et al* (1979). The inconsistency reported by Hayashi and Nimura for the sum of elastic and inelastic cross sections has thus been resolved.

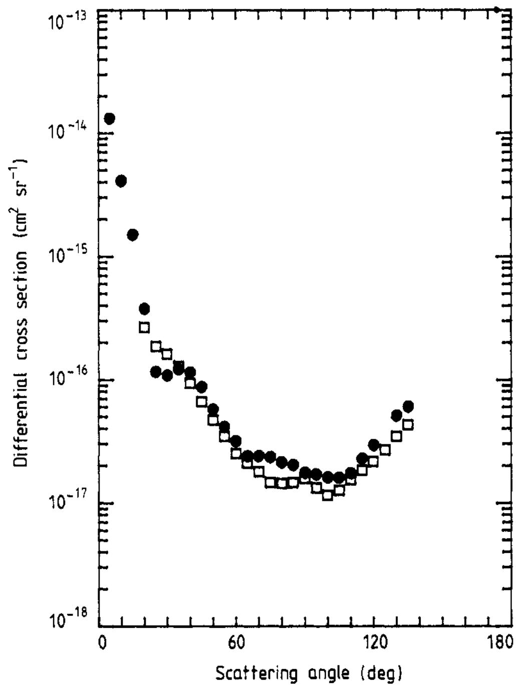

Figure 10. Comparison of the present DCS (●) for SF_6 with those of Srivastava *et al* (□). The electron energy is 75 eV.

5. Conclusions

Absolute values of the DCS have been obtained for CH_4, CF_4 and SF_6. For CH_4, there was good agreement between the present and previous results. The DCS for SF_6 were considerably different from previous results for forward scattering. The sum of elastic and inelastic cross sections agreed, however, with other results.

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