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# Absolute partial cross sections for electron-impact ionization of $SF_6$ from threshold to 1000 eV

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**Abstract**
Absolute partial cross sections for electron-impact ionization of $SF_6$ are reported for electron energies from threshold to 1000 eV. The product ions are mass analysed using a time-of-flight mass spectrometer and detected with a position-sensitive detector whose output demonstrates that all product ions are collected with equal efficiency irrespective of their initial kinetic energies. Data are presented for the production of $SF_5^+$, $SF_4^+$, $SF_3^+$, $SF_2^+$, $(SF^+ + SF_4^{2+})$, $S^+$, $F^+$, $SF_3^{2+}$, $SF_2^{2+}$ and $SF^{2+}$ and for the total cross section, which is obtained as the sum of the partial cross sections. The overall uncertainty in the absolute cross sections for singly charged ions, except $S^+$, is $\pm 5\%$; that for $S^+$ is $\pm 8\%$. The uncertainty in the cross sections for doubly charged ions is $\pm 12$ to 15%. Data are also presented for formation of $(SF_n^+, F^+)$ ion pairs. Comparison is made with prior experiments and calculations.

## 1. Introduction

Sulfur hexafluoride has many technological applications, including areas such as plasma etching. It is also known to be a potent greenhouse gas. Nevertheless, there have been very few measurements of its electron-impact ionization cross sections. The absolute total charge production cross section was measured by Rapp and Englander-Golden (1965) for energies up to 300 eV. There have also been three partial cross section measurements. However, the two earliest measurements (Stanski and Adamczyk 1983, Margreiter *et al* 1990b) both depend on normalization to the data of Rapp and Englander-Golden (1965) and furthermore neither was assigned an uncertainty. Moreover, the most recent measurement (Rao and Srivastava 1997) is of very limited scope as data are only reported at one energy. Consequently quantitative knowledge of the electron-impact ionization properties of the $SF_6$ molecule is largely based on the single study of Rapp and Englander-Golden (1965). Several calculations of the $SF_6$ total cross section have been reported (Margreiter *et al* 1990a, Hwang *et al* 1996, Tarnovsky *et al* 1998, Kim and Rudd 1999, Deutsch *et al* 2000). The available electron-impact ionization data for $SF_6$ have recently been reviewed by Christophorou and Olthoff (2000).

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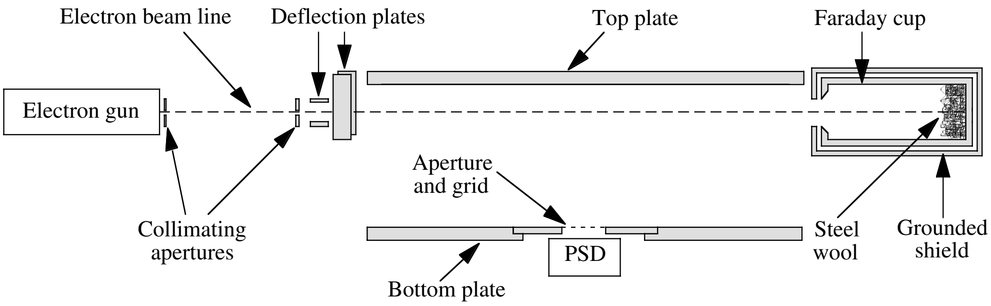


**Figure 1.** Schematic diagram of the apparatus.

In this paper absolute partial cross sections are reported for production of $SF_5^+$, $SF_4^+$, $SF_3^+$, $SF_2^+$, ($SF^+$ + $SF_4^{2+}$), $S^+$, $F^+$, $SF_3^{2+}$, $SF_2^{2+}$ and $SF^{2+}$ from threshold to 1000 eV. Only the sum of the $SF^+$ and $SF_4^{2+}$ cross sections is presented since these ions could not be fully resolved by the mass spectrometer. The total ionization cross section is obtained as the sum of the measured partial cross sections. During the course of this work it was determined that electron-impact ionization of $SF_6$ may result in production of positive ion pairs and the cross sections for ionization via these channels are also reported.

## 2. Apparatus and experimental method

The apparatus, which is shown in figure 1, consists of an electron gun, a time-of-flight mass spectrometer with a position-sensitive detector (PSD) and an absolute pressure gauge (not shown). It has been described in detail previously (Straub *et al* 1995, 1996). Briefly, during a cross-section measurement the entire vacuum chamber is filled with $SF_6$ at a pressure of $2.5 \times 10^{-6}$ Torr. The electron gun produces 20 ns long pulses at a repetition rate of approximately 10 kHz. These pulses are directed through an interaction region, located between two plates maintained at ground potential, and are collected in a Faraday cup. Approximately 250 ns after each electron pulse, a 3 kV pulse is applied to the top plate to drive any positive ions formed by electron impact toward the bottom plate. Some ions pass through a grid-covered aperture in the bottom plate. These ions are then accelerated and subsequently impact a PSD (Gao *et al* 1984), which records their arrival times and positions. The ion arrival times are used to identify their mass-to-charge ratios and the ion arrival positions are used to determine the effectiveness of product ion collection. Under conditions in which very few of the incident electrons produce an ion, the partial cross section $\sigma(\mathrm{X})$ for production of ion species X is given by

$$\sigma(\mathrm{X}) = \frac{N_{\mathrm{i}}(\mathrm{X})}{N_{\mathrm{e}} n l} \qquad (1)$$

where $N_{\mathrm{i}}(\mathrm{X})$ is the number of X ions produced by a number $N_{\mathrm{e}}$ of electrons passing a distance $l$ through a uniform $SF_6$ target of number density $n$. $\sigma(\mathrm{X})$ is then determined by measuring the four quantities on the right-hand side of equation (1) (Straub *et al* 1995, 1996). Technical details concerning the PSD detection efficiency calibration and use of the capacitance diaphragm gauge may be found in Straub *et al* (1999) and (1994) respectively.

The $SF_6$ used in this work was obtained from Matheson Gas Products and has a specified purity of 99.8%. It is transported from the gas cylinder through stainless-steel tubing and introduced to the vacuum chamber through a leak valve. The effectiveness of these procedures

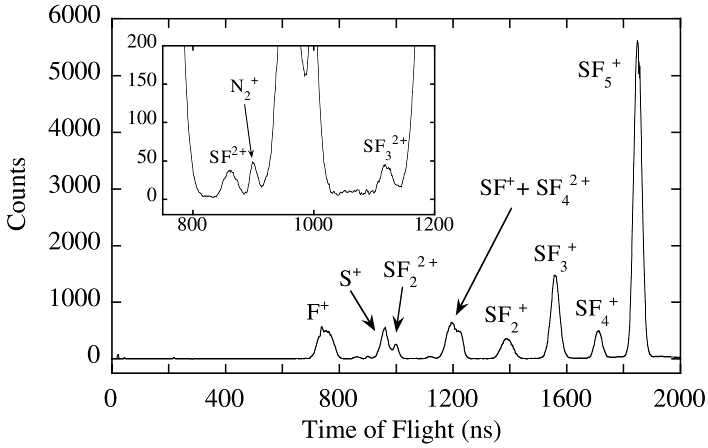


**Figure 2.** Time-of-flight spectrum for ions produced by 100 eV electron impact on $SF_6$.

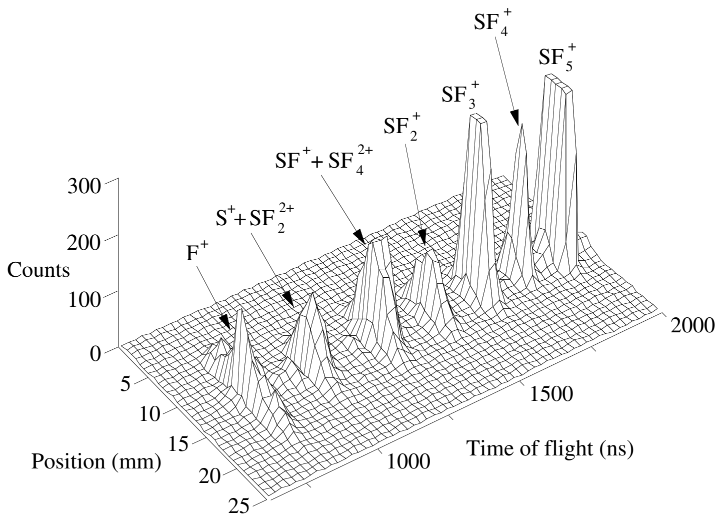


**Figure 3.** Position and time-of-flight distribution produced by 100 eV electron impact on $SF_6$. The position axis indicates the displacement of the ions perpendicular to the axis of the electron beam.

in preventing contamination can be seen from the absence of spurious peaks in the time-of-flight spectrum shown in figure 2. As noted previously by other workers, no stable $SF_6^+$ ion is observed. As the figure shows, it was not possible to resolve the $SF^+$ peak from the $SF_4^{2+}$ peak or to completely resolve the $S^+$ and $SF_2^{2+}$ peaks. In the latter case the peaks are sufficiently well separated to allow the individual cross sections to be determined by the use of a fitting procedure (Lindsay *et al* 2000).

Figure 3 shows a plot in which the product ions' transverse arrival positions at the PSD (i.e. the displacement of the ions perpendicular to the electron beam axis) have been combined with their flight times. The widths in both position and time of the various ion peaks are due to the ions' initial velocities perpendicular to the electron beam in addition to the transverse spatial extent of the electron beam itself. The width of the $SF_5^+$ peak is due primarily to the spatial extent of the electron beam, but the widths of the lightest ion peaks are determined largely by the energies with which these ions are formed. Complete collection of all product ions from the path length $l$ is demonstrated by the arrival position distribution of the ions at the PSD.

During the course of this work it was determined that electron-impact ionization of $SF_6$ may result in production of positive ion pairs via

$$e + SF_6 \rightarrow SF_n^+ + F^+ + \cdots. \tag{2}$$

While double photoionization of $SF_6$ has been observed previously (Frasinski *et al* 1986) this is, to our knowledge, the first observation of positive ion pair formation in electron collisions with $SF_6$. Although a somewhat indirect experimental approach was utilized here it is clear that there is a significant probability that, in a single collision, an $F^+$ ion may be formed together with a $SF_3^+$, $SF_2^+$, $SF^+$ or $S^+$ ion. As the instrumentation utilized here is capable of counting only one ion per electron pulse when two ions are created in the same collision the first to arrive at the detector will, if detected, preclude detection of any subsequent ions and thus lead to underestimation of their cross sections. In order therefore to measure the cross sections for all ions other than $F^+$ a gate was applied to the timing signal to suppress detection of the $F^+$ ions. This ensures that the measured partial cross section $\sigma(X)$ is for production of the specified fragment ion X irrespective of whether an $F^+$ ion is also formed. The cross section for $F^+$ ions was obtained by suppressing detection of all other ions. This approach however may still lead to an underestimation of the true $F^+$ cross section because it is possible that two $F^+$ ions may occasionally be formed in the same collision, only one of which will be counted. A similar problem in the case of $CF_4$ required that the measured $F^+$ cross section be increased by up to 6% (Sieglaff *et al* 2001).

The cross sections for ion pair production were determined by taking advantage of the fact that the counting electronics used can only process one event at a time. Thus, if a light ion Y and a heavier ion X are formed in the same collision and the timing electronics is limited to recording only the first ion detected, then the apparent cross section for production of the heavier, and thus slower, ion X, $\sigma_{app}(X)$, is given by

$$\sigma_{app}(X) = \sigma(X) - \varepsilon\sigma(X, Y) \tag{3}$$

where $\sigma(X)$ is the true cross section for production of X ions, $\sigma(X, Y)$ is the cross section for production of both an X and a Y ion in the same event, and $\varepsilon$ is the detection efficiency of the system. By measuring both the true and the apparent cross sections for production of $S^+$ ions for example and then applying equation (3) it is possible to determine the cross sections for production of ($S^+$, $F^+$) ion pairs. It should be noted that this analysis assumes that relatively few ionization events result in the formation of three positive ions, which is almost certainly warranted within the accuracy of the reported measurements.

A detailed analysis of the experimental uncertainties has been given previously (Straub *et al* 1995, 1996). However, in the light of experience gained with this apparatus over a number of years the uncertainty in the absolute cross section for ions, such as $SF_5^+$ (where the counting statistics are $\pm 1\%$ or better), is now considered to be $\pm 5\%$. The uncertainties in the $SF_4^+$, $SF_3^+$, $SF_2^+$, $SF^+$ and $F^+$ cross sections are also $\pm 5\%$. That in the $S^+$ cross section, estimated as $\pm 8\%$, is larger due to the fitting procedure necessitated by the overlap of the $S^+$ and $SF_2^{2+}$ peaks. The uncertainties in the $SF_3^{2+}$, $SF_2^{2+}$ and $SF^{2+}$ cross sections are $\pm 12$, $\pm 15$ and $\pm 12\%$ respectively. The uncertainty in the measured $F^+$ cross section is $\pm 5\%$, but note that this may only be a lower limit to the true $F^+$ cross section because of the experimental difficulties described above related to ion pair production. The uncertainties in the ($SF_3^+$, $F^+$), ($SF_2^+$, $F^+$), ($SF^+$, $F^+$) and ($S^+$, $F^+$) ion pair production cross sections are due primarily to counting statistics and are $\pm 30$, $\pm 25$, $\pm 20$ and $\pm 10\%$ respectively. Near the threshold for formation of each species the

**Table 1.** Absolute partial ionization cross sections for electron-impact ionization of $SF_6$. The uncertainties in the $SF_5^+$, $SF_4^+$, $SF_3^+$, $SF_2^+$, ($SF^+$ + $SF_4^{2+}$), $S^+$ and $F^+$ cross sections are ±5, ±5, ±5, ±5, ±5, ±8 and ±5% respectively, unless otherwise indicated.

| Energy (eV) | $\sigma(SF_5^+)$ ($10^{-16}$ cm$^2$) | $\sigma(SF_4^+)$ ($10^{-17}$ cm$^2$) | $\sigma(SF_3^+)$ ($10^{-17}$ cm$^2$) | $\sigma(SF_2^+)$ ($10^{-17}$ cm$^2$) | $\sigma(SF^+ + SF_4^{2+})$ ($10^{-17}$ cm$^2$) | $\sigma(S^+)$ ($10^{-17}$ cm$^2$) | $\sigma(F^+)$ ($10^{-17}$ cm$^2$) |
|---|---|---|---|---|---|---|---|
| 18 | $0.06 \pm 0.02$ | | | | | | |
| 20 | $0.19 \pm 0.03$ | | | | | | |
| 22.5 | 0.46 | $0.13 \pm 0.07$ | $0.12 \pm 0.06$ | | | | |
| 25 | 0.86 | $0.26 \pm 0.08$ | $0.56 \pm 0.07$ | | | | |
| 27.5 | 1.18 | $0.42 \pm 0.05$ | $1.17 \pm 0.12$ | | | | |
| 30 | 1.47 | $0.69 \pm 0.06$ | $2.21 \pm 0.13$ | | | | |
| 32.5 | 1.84 | $1.11 \pm 0.07$ | 3.43 | $0.22 \pm 0.02$ | | | |
| 35 | 2.03 | 1.30 | 4.81 | $0.55 \pm 0.04$ | | | |
| 40 | 2.36 | 1.67 | 6.23 | 1.21 | $0.52 \pm 0.03$ | 0.11 | |
| 45 | 2.57 | 1.76 | 5.89 | 1.82 | 1.59 | 0.29 | |
| 50 | 2.74 | 1.97 | 6.76 | 2.27 | 2.75 | 0.70 | $0.49 \pm 0.12$ |
| 60 | 3.01 | 2.27 | 8.08 | 2.79 | 4.03 | 1.51 | $1.34 \pm 0.16$ |
| 70 | 3.12 | 2.52 | 8.69 | 3.02 | 4.89 | 2.11 | 2.39 |
| 80 | 3.23 | 2.67 | 8.92 | 2.94 | 5.30 | 2.54 | 2.94 |
| 90 | 3.23 | 2.68 | 9.26 | 3.08 | 5.94 | 2.88 | 3.96 |
| 100 | 3.34 | 2.86 | 9.71 | 3.14 | 6.34 | 3.32 | 5.01 |
| 120 | 3.31 | 2.83 | 9.59 | 2.95 | 6.50 | 3.92 | 6.33 |
| 140 | 3.34 | 2.89 | 9.79 | 2.95 | 6.49 | 4.17 | 7.32 |
| 160 | 3.32 | 2.70 | 9.55 | 2.74 | 6.61 | 4.24 | 7.85 |
| 200 | 3.23 | 2.67 | 9.35 | 2.66 | 6.27 | 4.31 | 8.45 |
| 250 | 3.03 | 2.43 | 8.68 | 2.34 | 5.69 | 3.90 | 8.17 |
| 300 | 2.93 | 2.38 | 8.17 | 2.18 | 5.17 | 3.71 | 7.62 |
| 400 | 2.62 | 2.08 | 7.17 | 1.72 | 4.25 | 3.00 | 6.27 |
| 500 | 2.42 | 1.99 | 6.60 | 1.54 | 3.83 | 2.63 | 5.38 |
| 600 | 2.21 | 1.72 | 5.98 | 1.40 | 3.42 | 2.33 | 4.81 |
| 800 | 1.86 | 1.46 | 4.93 | 1.13 | 2.67 | 1.73 | 3.72 |
| 1000 | 1.65 | 1.27 | 4.49 | 0.98 | 2.21 | 1.51 | 2.97 |

uncertainties in the cross sections are of course greater and are given in table 1. The mean energy of the electron beam was established to within ±0.5 eV by observing the threshold for $He^+$ formation in the presence of the $SF_6$ target gas.

## 3. Results and discussion

The present cross sections for production of $SF_5^+$, $SF_4^+$, $SF_3^+$, $SF_2^+$, ($SF^+$ + $SF_4^{2+}$), $S^+$, $F^+$, $SF_3^{2+}$, $SF_2^{2+}$ and $SF^{2+}$ are given in tables 1 and 2 and are shown in figure 4. It should be noted that the cross sections shown are for production of the specified fragment ion irrespective of whether another ion is produced in the same event. As mentioned earlier there have been few prior studies in which the $SF_6$ partial cross sections were obtained. However, differences of factors of 2 or more exist between some of these cross sections, and the vast majority of the data have not been assigned any uncertainty. In these circumstances inclusion of these measurements in the figures seems of dubious utility and would probably only serve to add confusion. The only other partial cross sections, besides those presented here, which have been assigned an uncertainty are those of Rao and Srivastava (1997). Their measurements were performed at an electron energy of 100 eV and the reported uncertainty is ±10%. The present cross sections for production of lighter ions, such as $SF^+$, are considerably larger than those of Rao and

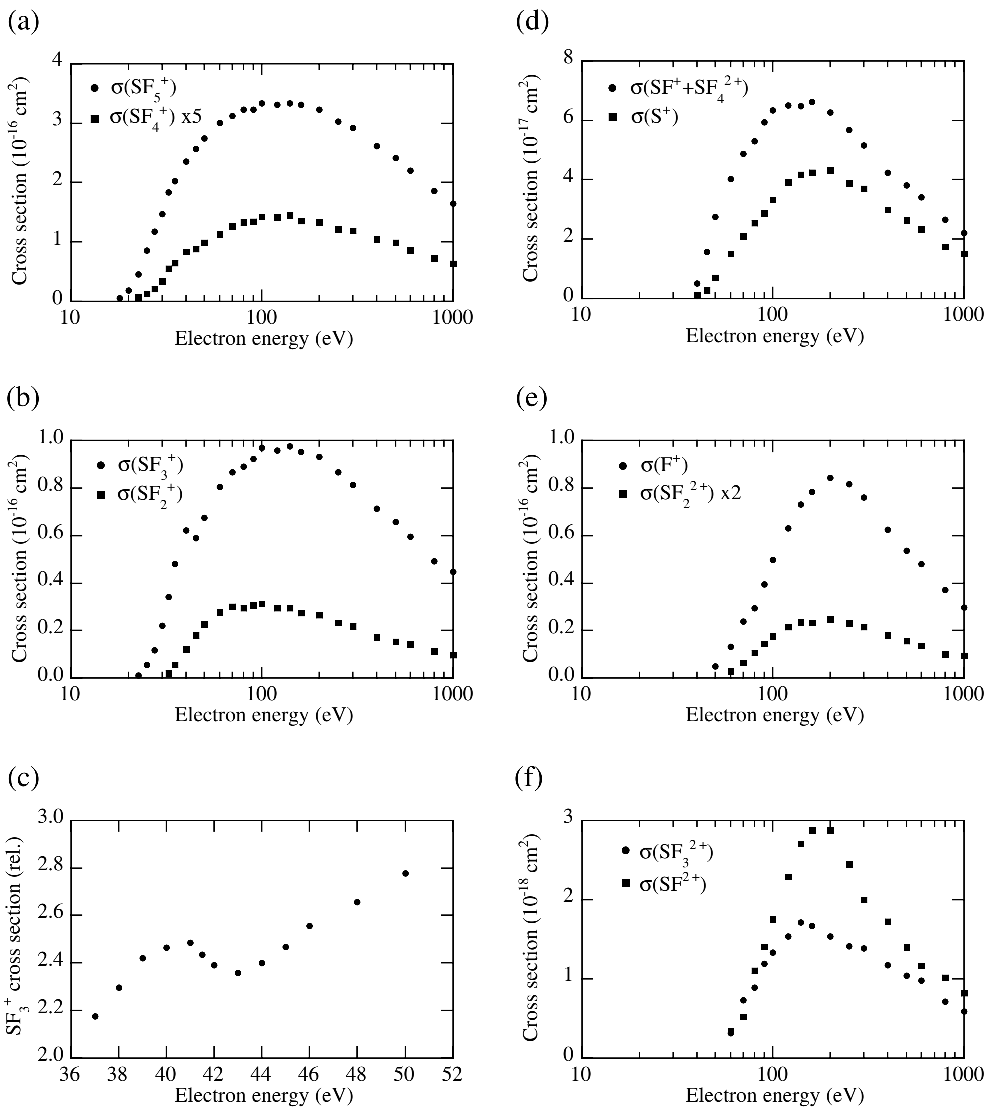


**Figure 4.** $SF_6$ partial electron-impact ionization cross sections. (*c*) The $SF_3^+$ cross section in the vicinity of 44 eV. A similar feature was observed previously by Margreiter *et al* (1990b).

Srivastava (1997) suggesting that they may have been unable to completely collect all of the energetic fragment ions.

At the energy resolution employed here the partial ionization cross sections for molecules generally do not exhibit structure. However, several features which are consistent with those seen previously by Margreiter *et al* (1990b) are apparent in the present work. The most prominent of these is shown in figure 4(*c*).

The present $SF_6$ total electron-impact ionization cross section is shown in figure 5 together with the absolute cross section of Rapp and Englander-Golden (1965). It should be noted that the data of Rapp and Englander-Golden (1965) are for total charge production whereas the present data are the sums of the partial cross sections. These data are nonetheless directly

**Table 2.** Absolute cross sections for production of doubly charged ions by electron-impact ionization of $SF_6$. The uncertainties in the $SF_3^{2+}$, $SF_2^2$ and $SF^{2+}$cross sections are ±12, ±15 and ±12% respectively, unless otherwise indicated.

| Energy (eV) | $\sigma(SF_3^{2+})$ ($10^{-18}$ cm$^2$) | $\sigma(SF_2^{2+})$ ($10^{-17}$ cm$^2$) | $\sigma(SF^{2+})$ ($10^{-18}$ cm$^2$) |
|---|---|---|---|
| 60 | 0.32 ± 0.10 | 0.15 ± 0.05 | 0.35 ± 0.07 |
| 70 | 0.73 ± 0.19 | 0.32 ± 0.09 | 0.52 ± 0.09 |
| 80 | 0.89 ± 0.18 | 0.53 ± 0.13 | 1.11 ± 0.17 |
| 90 | 1.19 ± 0.18 | 0.72 ± 0.16 | 1.41 |
| 100 | 1.33 | 0.88 ± 0.18 | 1.75 |
| 120 | 1.54 | 1.08 | 2.29 |
| 140 | 1.72 | 1.19 | 2.71 |
| 160 | 1.67 | 1.17 | 2.87 |
| 200 | 1.54 | 1.24 | 2.87 |
| 250 | 1.41 | 1.16 | 2.45 |
| 300 | 1.39 | 1.08 | 2.00 |
| 400 | 1.17 | 0.90 | 1.72 |
| 500 | 1.04 | 0.78 | 1.39 |
| 600 | 0.98 | 0.68 | 1.16 |
| 800 | 0.72 | 0.51 | 1.02 |
| 1000 | 0.59 | 0.47 | 0.82 |

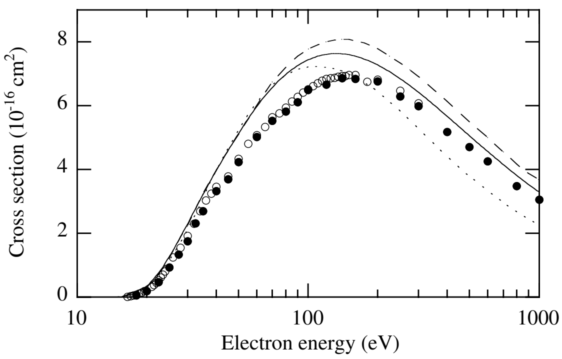


**Figure 5.** Total cross section: present results (●); Rapp and Englander-Golden (1965) (○); BEB theory from Hwang *et al* (1996) (——); BEB theory including multiple ionization from Kim and Rudd (1999) (- - - -); DM theory from Deutsch *et al* (2000) (· · · · · ·).

comparable because of the very small number of multiply charged ions observed. Figure 5 also shows a number of theoretical calculations. The work of Hwang *et al* (1996) and Kim and Rudd (1999), obtained using the binary-encounter–Bethe model (BEB), predicts the energy at which the cross section peaks more accurately than the semiclassical Deutsch–Märk (DM) formalism; however, the magnitude of the DM cross section is closer to the experimental value.

The cross sections for the various ion pair production channels are given in table 3 and plotted in figure 6. Note that any apparent structure in these cross sections is almost certainly a consequence of the large statistical uncertainty in the data. Presently no other experimental or theoretical data exist with which to compare these cross sections; however, the magnitude of these pair production cross sections is similar to those observed in the well studied case of $CF_4$ (Bruce *et al* 1992, 1994, Sieglaff *et al* 2001).

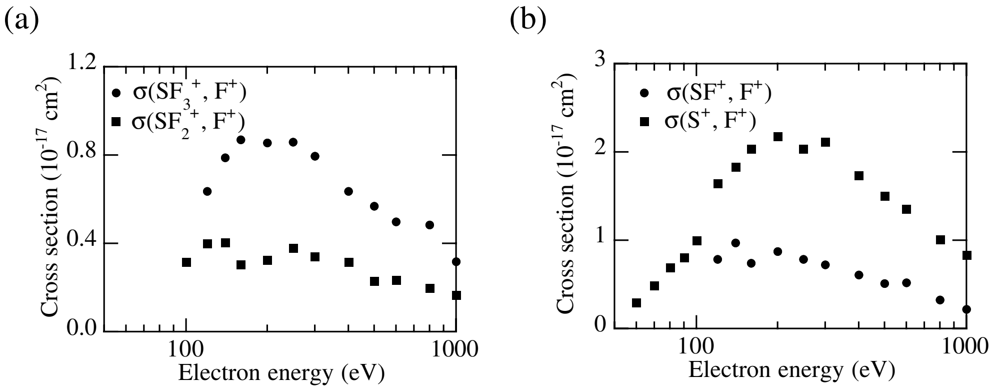


**Figure 6.** Cross sections for production of ($SF_n^+$, $F^+$) ion pairs.

**Table 3.** Absolute cross sections for production of ($SF_3^+$, $F^+$), ($SF_2^+$, $F^+$), ($SF^+$, $F^+$) and ($S^+$, $F^+$) ion pairs by electron-impact ionization of $SF_6$. The uncertainties in these cross sections are ±30, ±25, ±20 and ±10% respectively, unless otherwise indicated.

| Energy (eV) | $\sigma(SF_3^+, F^+)$ ($10^{-18}$ cm$^2$) | $\sigma(SF_2^+, F^+)$ ($10^{-18}$ cm$^2$) | $\sigma(SF^+, F^+)$ ($10^{-18}$ cm$^2$) | $\sigma(S^+, F^+)$ ($10^{-17}$ cm$^2$) |
|---|---|---|---|---|
| 60 | | | | $0.29 \pm 0.09$ |
| 70 | | | | $0.48 \pm 0.09$ |
| 80 | | | | $0.69 \pm 0.10$ |
| 90 | | | | $0.81 \pm 0.11$ |
| 100 | | $3.2 \pm 1.1$ | | $1.00 \pm 0.13$ |
| 120 | $6.4 \pm 2.3$ | $4.0 \pm 0.9$ | 7.9 | 1.64 |
| 140 | $7.9 \pm 2.7$ | $4.0 \pm 1.0$ | 9.7 | 1.83 |
| 160 | 8.7 | $3.0 \pm 0.8$ | 7.4 | 2.03 |
| 200 | 8.5 | $3.2 \pm 0.8$ | 8.7 | 2.18 |
| 250 | 8.6 | 3.8 | 7.8 | 2.04 |
| 300 | 8.0 | 3.4 | 7.2 | 2.11 |
| 400 | 6.4 | 3.1 | 6.1 | 1.74 |
| 500 | 5.7 | 2.3 | 5.1 | 1.50 |
| 600 | 5.0 | 2.3 | 5.2 | 1.35 |
| 800 | 4.8 | 2.0 | 3.2 | 1.01 |
| 1000 | $3.2 \pm 1.6$ | 1.7 | $2.2 \pm 0.7$ | 0.83 |

## 4. Conclusion

Absolute partial cross sections for the production of $SF_5^+$, $SF_4^+$, $SF_3^+$, $SF_2^+$, ($SF^+$ + $SF_4^{2+}$), $S^+$, $F^+$, $SF_3^{2+}$, $SF_2^{2+}$ and $SF^{2+}$ from electron-impact ionization of $SF_6$ are reported for energies from threshold to 1000 eV. Data are also presented for the formation of ($SF_n^+$, $F^+$) ion pairs. The apparatus geometry is of simple design embodying a short-path-length time-of-flight mass spectrometer and position-sensitive detection of the product ions, which unequivocally demonstrates that all fragment ion species are collected with equal efficiency irrespective of their initial kinetic energy. These are the only published partial cross section data which do not rely on normalization to the work of others. The present total cross section is in excellent agreement with the measurements of Rapp and Englander-Golden (1965); however, agreement with even the most recent theoretical calculations is only fair.

## Acknowledgments

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