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Isomer effects on the total cross section for electron scattering from C₄F₆ molecules

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Abstract

The absolute total cross section (TCS) for electron scattering from hexafluoro-1,3-butadiene (1,3-C₄F₆) was obtained in a transmission experiment for electron impact energies from 0.6 to 370 eV. The TCS energy function has two pronounced enhancements separated by a deep minimum located near 2.6 eV: a low-energy hump around 1 eV and a dominant very broad enhancement centred near 30 eV. The 1,3-C₄F₆ TCS results are compared with the data for hexafluoro-2-butyne (2-C₄F₆) and the influence of the structural differences on the electron-scattering TCS for isomers of the C₄F₆ molecule (*isomeric effect*) is explicitly indicated. The most pronounced role of the molecular geometry on the magnitude and shape of the TCS energy dependence is observed at the lowest energies studied. Above 40 eV the TCSs for both isomers are close to each other. Some conformities of TCSs within a series of perfluorocarbons (C₂F₄, 1,3-C₄F₆, C₆F₆) as well as in a series of their hydrocarbon counterparts (C₂H₄, 1,3-C₄H₆, C₆H₆) are also pointed out and discussed.

1. Introduction

Studies intended to indicate the influence of atom location in a target molecule on electron scattering were begun in the 1930s [1, 2]. However, differences between early total cross sections (TCS) for isomers of hydrocarbons (C₄H₁₀ and C₅H₁₂) measured at energies below 50 eV [1, 2] appeared rather imperceptible; a more convincing manifestation of the influence of molecular geometry on the magnitude and shape of the TCS was visible only below 4 eV for C₂H₆O isomers [2]. More recent investigations concerning the electron-induced ionization have not yet given the explicit answer to how the changes in molecular geometry reflect in the scattering processes. The differences between ionization cross sections for the same sets of hydrocarbon isomers obtained in various laboratories [3–8] are, if anything, rather small and not systematic. Even in the vicinity of the ionization efficiency maximum (70–80 eV),

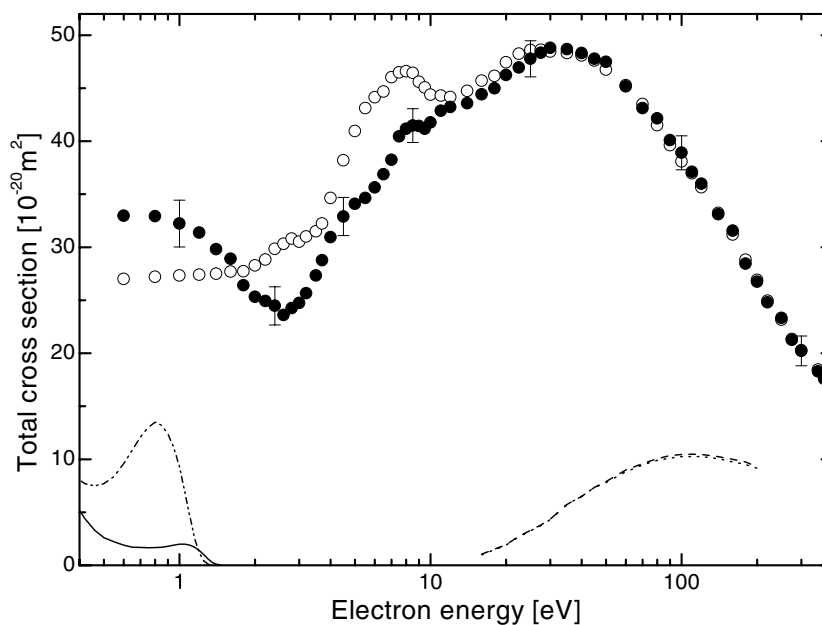


Figure 1. The energy dependence of experimental cross sections for electron- C_4F_6 scattering. Hexafluoro-1,3-butadiene: ●, TCS, present (error bars at selected energies represent overall uncertainties, statistical plus systematical); —, total electron attachment, [16]; - - -, total ionization, [9]. Hexafluoro-2-butyne: ○, TCS, [36]; — · · —, total electron attachment, [16]; · · · · ·, total ionization, [9].

where the observed ionization cross sections for hydrocarbon isomers differ mostly, the change in the magnitude due to geometry variation does not exceed a few per cent, and it is well within the limits of experimental error. Similarly small differences of experimental ionization cross sections were observed for perfluorocarbon isomers [9] (cf figure 1). The calculations employing the Deutsch-Märk formalism, though they account for the general shape and magnitude of the experimental data, also do not demonstrate any discernible influence of atom arrangement in the target molecule on the ionization cross section [10].

In contrast to those low-intermediate and intermediate energy findings, the low-energy electron-scattering experimental data indicate a very distinct isomeric effect visible in various scattering channels: elastic [11–14], electron attachment [15–19] (e.g. figure 1), vibrational [11–13, 20–24], and electronic [22–25] excitation, for selected hydrocarbons as well as for fluoro- and/or perfluoro-carbons. Also low-energy approximate calculations clearly show a pronounced isomeric effect on the elastic cross section for electron impact on simple hydrocarbons [26–28]. A variation of resonance positions for isomers of fluorinated hydrocarbons was noticed in the electron transmission spectra [29–31].

More recent TCS measurements for series of hydrocarbons carried out from 4–5 eV to intermediate energies [32, 33] have suggested that one would expect a more significant isomeric effect on the TCS around the TCS maximum location, i.e. below 10 eV. More spectacular differences in the shape and magnitude of the electron-scattering TCS, attributed to different arrangements of constituent atoms, have been found very recently between 0.5 and 12 eV for isomers of simple hydrocarbons: C_3H_6 [34], C_3H_4 [14, 35] (also for positron

scattering [14]) and for C₄H₆ [36]. For isomers of perfluorocarbons the structural effect on the electron-scattering TCS has not yet been reported.

In this paper we demonstrate the isomeric effect on the total electron-scattering cross section for two structural isomers of the C₄F₆ molecule: hexafluoro-1,3-butadiene (1,3-C₄F₆) and hexafluoro-2-butyne (2-C₄F₆). Hexafluoro-1,3-butadiene (CF₂=CF-CF=CF₂) is the simplest conjugated double-bonded perfluorocarbon species with the two trifluorovinyl groups lying in different planes [37, 38]. Its isomeric counterpart hexafluoro-2-butyne (CF₃-C≡C-CF₃) is a linear molecule with a central CC triple bond and with a trifluoromethyl group at each end of the molecule. Due to its very low global warming potential [39], hexafluoro-1,3-butadiene is considered as a substitutive component of plasmas for fabricating nanoelectronic devices [40], and therefore there is a need for comprehensive data on the electron-assisted processes in this compound. The experimental absolute total electron-scattering cross sections for 1,3-C₄F₆ presented in the following discussion have been obtained just in this work, while the TCS data for 2-C₄F₆ have already been published [36]. The results provide some insight into the role played by the structural properties of the target compound in the scattering process and, consequently, may serve as a guide for theoretical investigations.

2. Experimental details

The absolute TCS for 1,3-C₄F₆ molecule has been obtained in single-collision conditions using a linear electron-beam transmission technique [41] which is based on the measurements of the attenuation of an electron beam passing through the gaseous target under study. The apparatus and experimental procedure used in the present study are the same as those employed in the earlier 2-C₄F₆ TCS measurements [36], and they were described in detail elsewhere (e.g. [42]). A tunable-energy electron beam, formed in a 127° cylindrical dispersing element and a system of electron lenses, is directed into a reaction chamber filled with a gaseous sample. Those electrons which pass through the exit aperture of the scattering cell are energetically discriminated by the retarding field assembly and eventually collected with a Faraday cup detector. The electron energy scale is calibrated by a small admixture of N₂, yielding the well known resonant oscillatory structure around 2.3 eV. The absolute number density of target molecules was derived from the ideal gas law based on absolute measurements of the gas-target pressure in the reaction volume and its temperature, taking into account a thermal transpiration effect [43].

The final TCS value at each electron impact energy is a weighted mean of results from several (3–7) series of individual measurements (7–10 in a series). The statistical variations of the TCS (one standard deviation of the weighted mean value) are less than 1% over the whole energy range studied. The overall systematical uncertainty in the measured TCS, estimated as a combination of potential systematic errors of all quantities taken directly in the experiment, is about 8% below 2 eV, decreasing gradually to 5% in the range 5–150 eV, and amounts to about 7% at the highest applied energies. The main contribution to the resulting systematical uncertainty of the measured TCS comes from

- (i) the inability to determine correctly the distribution of the target density along the scattering volume [44],
- (ii) the inability to discriminate electrons scattered elastically or with small energy losses into the forward direction—the effect increases with electron energy increase,
- (iii) the electron current instabilities at the lowest applied energies, and
- (iv) the impurity of the gas target under study.

The sample of 1,3-C₂F₄ from Interchim, with a stated minimum purity of 97%, was used directly from the cylinder without further purification.

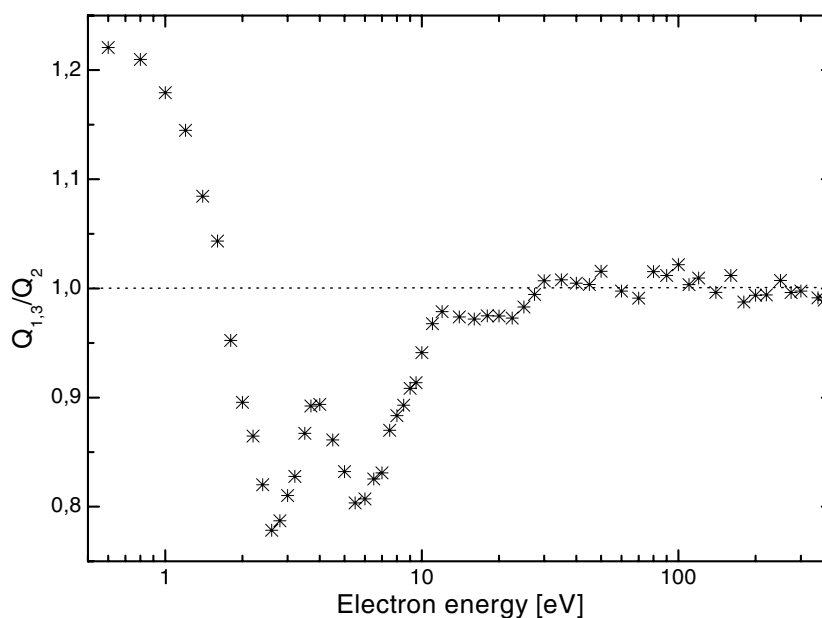


Figure 2. The ratio, $\rho = Q_{1,3}/Q_2$, of electron-scattering TCSs for 1,3- C_4F_6 ($Q_{1,3}$, present) and 2- C_4F_6 (Q_2 , [36]).

3. Results and discussion

Figure 1 shows the absolute electron-scattering TCS for 1,3- C_4F_6 ($Q_{1,3}$) measured in this work over the energy range from 0.6 to 370 eV and other quantitative data on the e^- -1,3- C_4F_6 scattering available in the literature: the electron attachment [15] and the electron-induced ionization [9] TCSs. The results on the electron scattering from 2- C_4F_6 (the absolute TCS (Q_2) [36], the electron attachment [15] and the total ionization [9] cross sections) are also included for comparison. In figure 2 the ratio, $\rho = Q_{1,3}/Q_2$, of TCSs for both C_4F_6 isomers under discussion is displayed as a function of electron energy, E .

Inspection of figures 1 and 2 clearly shows that different configuration of atoms in the isomers of C_4F_6 molecule causes the noticeable differences of the respective TCS for energies below 30 eV, and that below 10 eV this disparity becomes more expressive. In general, the TCS energy function for 1,3- C_4F_6 has two pronounced enhancements separated by a deep minimum centred near 2.6 eV: the low-energy enhancement is easily visible below 2 eV, and the second very broad one is peaked near 30 eV. In contrast, the TCS for 2- C_4F_6 has only one very broad enhancement though with two maxima located near 8 and 30 eV [36]. The broad enhancement centred around 30–40 eV has also already been found for other perfluorinated hydrocarbons studied so far (e.g. [45, 46]) and seems to be the usual behaviour of perfluorides. Weak features, probably attributable to resonant effects, discernible on the rising slope of the broad TCS enhancement, are located for both isomers at significantly different energies. The relatively high magnitude of compared TCSs—their values markedly exceed $25 \times 10^{-20} \text{ m}^2$ over the considerable energy range studied—reflects a more general phenomenon: the electron-scattering TCS increases with the geometrical size of the molecular carbon skeleton [32, 33, 47].

In the following the observed differences and similarities in the compared TCS energy dependences are discussed in more detail. Between 0.6 and 1 eV the TCS for 1,3- C_4F_6 is

about 20% higher than that for 2-C₄F₆. Low-energy experiments on the dissociative electron attachment to C₄F₆ molecules [15, 16] clearly suggest that around 1 eV the scattering for both isomers is in part resonant in character with the formation of transient negative ions, C₄F₆^{-*}, when the impacting electron is temporarily accommodated into the lowest normally unoccupied molecular orbital. Such a resonant state may decompose either via autodetachment of the extra electron or via a variety of dissociative channels leading to production of neutral products and fragment anions [16]. At electron impact energies below the electronic threshold the autoionization leaves the parent molecule in one of its vibrational levels of the electronic ground state. The temporary negative ion state, 1,3-C₄F₆⁻, formed around 1 eV is the lower of two states arising from the splitting of the π^* orbitals of the two perfluoroethylenic subunits (the π^* anion state of tetrafluoroethylene is created near 2.8–3 eV [29, 48]); the upper π^* resonant state should be located near 5 eV. The electron attachment cross section for 1,3-C₄F₆ at energy 0.8 eV (1.6×10^{-20} m² [15]) is, however, almost an order of magnitude lower than the cross section for electron attachment to the 2-C₄F₆ molecule (figure 1). On the other hand, recent calculations at the static-exchange approximation for C₄H₆ isomers [27] demonstrate that the elastic scattering for 1,3-C₄H₆ near 1 eV is roughly 20–25% more effective than for 2-C₄H₆; the experimental TCSs for these isomers near 1 eV differ by factor of 2 [36]. Supposing the same relation is true for the elastic scattering from perfluorinated analogs, C₄F₆, and taking into account the aforementioned differences in the total attachment cross sections, it is likely that the higher TCS value for 1,3-C₄F₆ near 1 eV may be attributed in part to more efficient elastic scattering and in part to inelastic scattering phenomena for this isomer. A more certain explanation of the observed difference needs, however, further accurate quantitative data for various scattering channels.

In the range from 1 to about 2.6 eV the TCS for 1,3-C₄F₆ decreases steeply and, whereas the TCS for 2-C₄F₆ is a slowly increasing function of energy in that region, the TCS for 1,3-C₄F₆ above 1.8 eV appears to be lower than that for 2-C₄F₆. In the vicinity of 2.6 eV, 1,3-C₄F₆ becomes much more transparent for electrons and in consequence its TCS curve exhibits the very deep minimum around which the TCS for 1,3-C₄F₆ is nearly 20% lower than that for 2-C₄F₆.

From the minimum at 2.6 eV up to 4 eV the TCS for 1,3-C₄F₆ rises rapidly with energy increase. In the same energy range the TCS for 2-C₄F₆ is a rather slowly varying function of energy with a well-marked shoulder spanned between 2.5 and 4 eV, and a possible weak maximum at 2.8–3 eV. Therefore, around 4 eV the difference between cross sections diminishes to about 10%. On the other hand, between 4 and 6 eV a significant shoulder can be distinguished in the TCS for 1,3-C₄F₆, presumably related to the other π^* resonant state, and the TCS for 2-C₄F₆ steeply increases, which causes the succeeding increase of the difference between both TCSs up to about 20% at 6 eV. Above that energy, the isomeric difference decreases rapidly with energy increase and near 8 eV it falls to 10% again.

In both TCSs, the energy region around 8 eV is characterized by a distinct structure: a pronounced maximum for 2-C₄F₆ and a weak bump for the 1,3-C₄F₆ isomer. At between 9 and 11 eV the TCS for 2-C₄F₆ declines quite rapidly while the TCS for 1,3-C₄F₆ increases with energy; the difference of the compared TCSs diminishes down to 2% at 12 eV. For energies above 12 eV, the TCS general tendency for both perfluorocarbon isomers becomes very similar. Near 30 eV the TCSs attain their main maxima and above that energy the TCSs for the C₄F₆ isomers nearly overlap and decrease monotonically with impact energy as E^{-a} , where $a \sim 0.47$. Such an energy dependence means that at intermediate energies the TCS is roughly proportional to the time the approaching electron spends in the vicinity of the target molecule. The conformity of the compared C₄F₆ TCSs at intermediate energies suggests that an independent atom approximation (e.g. [49]) can reasonably reproduce both TCSs at higher

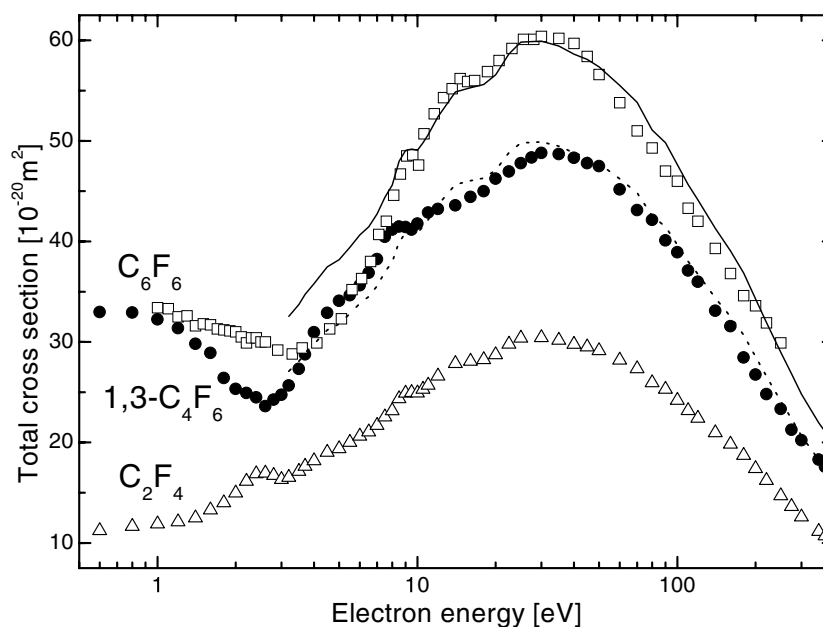


Figure 3. A comparison of the experimental electron-scattering TCS on: C_2F_4 (Δ , [48]), 1,3- C_4F_6 (\bullet , present) and C_6F_6 (\square , [50]). To demonstrate the similarity in the shape of compared TCSs, above 3 eV the TCS for C_2F_4 is multiplied by 1.6 (---) and by 2 (—).

energies. It is also worth noting that the share of ionization processes in the scattering for 1,3- C_4F_6 is, like for other perfluorides, relatively low, and around the maximum of the total ionization cross section amounts to no more than 1/3 of the TCS (cf figure 1).

It is interesting to note that above 3 eV the general shape of the TCS energy function for the 1,3- C_4F_6 compound closely resembles those found for C_2F_4 [48] and for C_6F_6 [50]. The ratio of TCS values is about 1:1.6:2 for C_2F_4 , 1,3- C_4F_6 and C_6F_6 , respectively. To aid in the visualization of that resemblance the TCS for C_2F_4 was multiplied by respective factors; then even weak structures of the compared TCSs look much the same (see figure 3). The aforementioned similarities can possibly be related to the fact that the hexafluoro-1,3-butadiene and the hexafluorobenzene molecules may be to some degree considered as composed of two and three 'perfluoroethylene subunits', respectively. Based on this somewhat speculative assumption, the observed ratio of the TCSs for a series of perfluorides can, however, be easily rationalized and well reproduced. A quite similar relationship, however only above 5 eV, is also noticed for TCS functions of the hydrocarbon analogues C_2H_4 , 1,3- C_4H_6 and C_6H_6 (figure 4); the ratio of the respective TCS amounts is 1:1.6:1.9 [36, 48, 51]. This finding suggests that the TCS for more complex molecules might be roughly estimated from TCSs of their submolecular components (*group additivity*). Further, it appeared that in the case of the considered series of perfluorides (C_2F_4 , 1,3- C_4F_6 and C_6F_6) the low-energy (below 3–5 eV) TCS curve lies below the TCS curve for a respective hydrocarbon (C_2H_4 , 1,3- C_4H_6 and C_6H_6). Such a behaviour is rather exceptional because usually that relationship is opposite and the TCS for the perfluorinated molecule is much higher in this energy range.

Finally, a few remarks should be made concerning the effect of substitution of fluorine atoms for hydrogen atoms in 1,3- C_4H_6 (*perfluorination effect*). Examination of the 1,3- C_4H_6 and 1,3- C_4F_6 TCSs compared in figure 5 reveals some similarities as well as differences:

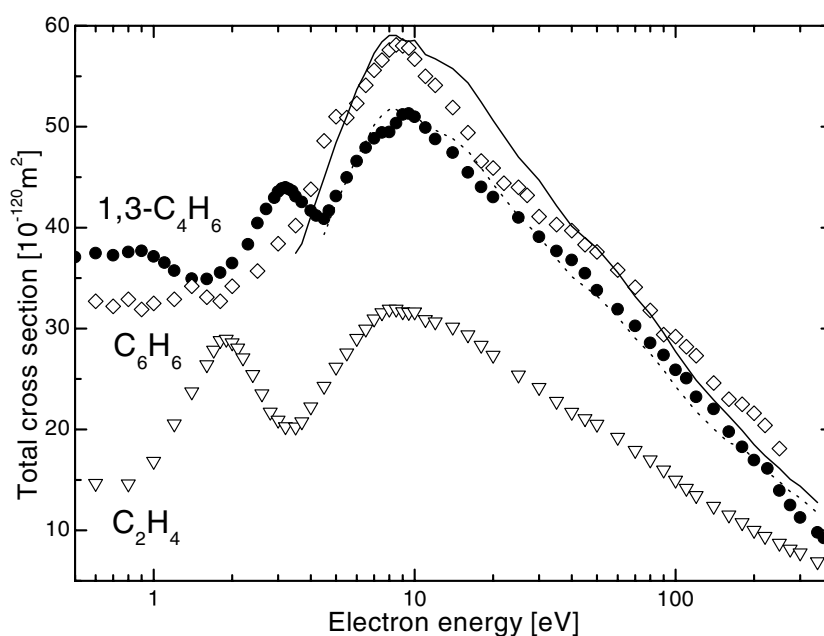


Figure 4. A comparison of the experimental electron-scattering TCS for: C₂H₄ (∇ , [48]), 1,3-C₄H₆ (\bullet , [36]) and C₆H₆ (\diamond , [51]). The TCS for C₂H₄ is multiplied by 1.6 (---) and by 1.9 (—) to show the similarity in the shape of compared TCS above 5 eV.

- (i) the low-energy enhancement in TCSs is visible below 1.5 eV for both 1,3-C₄F₆ and 1,3-C₄H₆ systems;
- (ii) the minimum in the 1,3-C₄F₆ TCS energy dependence is shifted by about 1 eV towards higher energy and is much deeper than for 1,3-C₄H₆;
- (iii) the main TCS enhancement for 1,3-C₄F₆ is much broader and is located at much higher energies (centred near 30 eV) in comparison to that for 1,3-C₄H₆ (centred near 9 eV);
- (iv) at energies below about 16 eV the TCS for 1,3-C₄F₆ is lower than that for 1,3-C₄H₆. This relation changes at intermediate energies where the TCS for 1,3-C₄F₆ becomes distinctly higher. This substituent effect is typical for all pairs of perfluorinated and perhydrogenated molecules studied so far [45, 46, 52];
- (v) the structures visible on the low-energy slope of the main enhancement are for 1,3-C₄F₆ less expressive than those for 1,3-C₄H₆.

4. Conclusion

The absolute total electron-scattering cross sections for 1,3-C₄F₆ molecules have been measured in a linear transmission experiment from 0.6 to 370 eV. To the best of our knowledge, no other electron-scattering TCS data for 1,3-C₄F₆ seem to exist in the literature. In order to study the isomeric effect, the 1,3-C₄F₆ TCS data have been compared with earlier 2-C₄F₆ results. The differences in the shape and magnitude of TCSs are visible below 30 eV and they increase with the electron impact energy decrease. To understand the role of the molecular structure on the mechanisms of the electron scattering better, further more detailed experiments and theoretical studies are necessary. We hope that due to the results presented here, some of the problems which have to be tackled appear to be more evident.

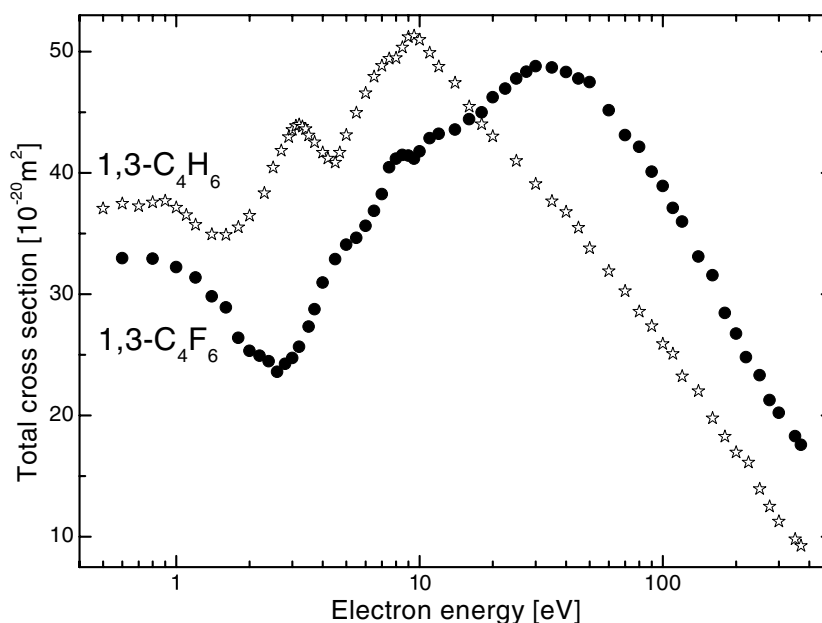


Figure 5. An illustration of the fluorination effect: experimental TCSs for electron scattering from 1,3-C₄F₆ (●, present) and 1,3-C₄H₆ (☆, [48]).

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