

ELECTRON ELASTIC COLLISIONS WITH C₃F₆ MOLECULE

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Abstract: We report calculations of differential and integral cross sections for intermediate- and high-energy (50–1000eV) elastic collisions of electrons with hexafluoropropene (C₃F₆) molecules. The calculations have been carried out using the independent atom model with static-polarization model potential. The present results are compared with elastic cross sections estimated from total and ionization experiments. Agreement between present calculations and the “experiment” is good for energies above 70eV.

Keywords: electron scattering, elastic cross section, hexafluoropropene, independent atom method

1. Introduction

Fully fluorinated hydrocarbons (*e.g.* CF₄, C₂F₆, C₃F₈) and SF₆ are gases with wide technological applications: plasma etching, deposition and cleaning, pulse power switching, gaseous dielectrics [1, 2]. These perfluorides are also of atmospheric and environmental concerns due to their rather long residence time in the environment and, in consequence, a high global warming potential (GWP) [3]. Therefore, there is an increasing demand for environmentally friendly alternatives (substitutes) for compounds used so far. One of the replacement candidates, with a relatively low GWP, is hexafluoropropene (C₃F₆) [4]. The e⁻-C₃F₆ scattering processes have received a little experimental and theoretical attention. A few experimental works concentrate on the dissociative electron attachment [5–9] and electron-induced ionization processes [7, 10–12]. Jiang *et al.* [13] calculated the total (elastic + inelastic) cross section at intermediate and high impact energies (30–3000eV) using a simple additivity rule

(AR) and an energy-dependent geometric additivity rule (EGAR). Very recently, Szmytkowski *et al.* [14, 15] have presented an absolute total cross section for electron collisions with C₃F₆ molecule measured with a transmission technique for incident electron energies between 0.5 and 370 eV. Preliminary results of integral elastic cross section calculations for energies ranging from 30 to 300 eV have been also reported [14]. We are not aware of any other calculated or measured elastic cross sections for electron scattering from C₃F₆ molecules.

2. Theory

The present calculations have been carried out using the independent-atom method (IAM) [16, 17] with static-polarization model potential. In this approximation, the electron-molecule collision problem is reduced to electron-atom collision, assuming that each atom of the molecule scatters independently, any redistribution of atomic electrons due to the molecular binding is unimportant, and multiple scattering within the molecule is neglected [16]. This approach offers reasonable approximations to elastic, momentum transfer and total cross sections for intermediate- and high-energy electron and/or positron scattering from many polyatomic molecular targets (see *e.g.* [13, 17–28] and references therein). The electron-impact ionization cross sections for a molecule are obtained as a sum of the relevant atomic components [29]. In the simple form of independent atom approximation, the differential cross section (DCS) for elastic electron scattering on a molecule, taking into account all possible orientations of the intermolecular axis, is given as:

$$\frac{d\sigma}{d\Omega} = \sum_i^N \sum_j^N f_i(\theta, k) f_j^*(\theta, k) \frac{\sin(sr_{ij})}{sr_{ij}}, \quad (1)$$

where N is the number of atoms in the molecule, θ is the scattering angle, and k is the incident electron wave number; $f_i(\theta, k)$ and $f_j(\theta, k)$ are complex scattering amplitudes due to the i^{th} and j^{th} atom of the molecule, respectively, and $s = 2k\sin(\theta/2)$ is the magnitude of the momentum transfer during the collision. r_{ij} is the internuclear distance between the i^{th} and j^{th} atom of the target molecule. In this investigation, the distances r_{ij} in the C₃F₆ molecule are obtained using an optimization procedure with GAMESS code [30], taking as the starting point the experimental geometry of the C₃F₆ molecule presented by Lowery *et al.* [31]. In all equations, atomic units are used in which $e = m_e = \hbar = 1$, although the final results of the calculations are given in SI units.

It follows from the optical theorem that the integral cross section (ICS) for electron elastic scattering on the molecule in the IAM approximation is given by:

$$\begin{aligned} \sigma(E) &= \frac{4\pi}{k} \text{Im} f(s=0, k) = \\ &= \frac{4\pi}{k} \sum_{i=1}^N \text{Im} f_i(\theta=0, k) = \sum_{i=1}^N \sigma_i(E), \end{aligned} \quad (2)$$

where $\sigma_i(E)$ is the integral cross section of the i^{th} atom of the molecule at energy $E = k^2/2$.

To obtain the atomic scattering amplitudes and elastic electron-atom cross sections, we have employed partial wave analysis and solved numerically the radial Schrödinger equation

$$\left[\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} + k^2 - 2(V_{\text{stat}}(r) + V_{\text{polar}}(r)) \right] u_l(r) = 0 \quad (3)$$

under the boundary conditions

$$u_l(0) = 0, \quad u_l(r) \xrightarrow{r \rightarrow \infty} A_l \hat{j}_l(kr) - B_l \hat{n}_l(kr), \quad (4)$$

where $\hat{j}_l(kr)$ and $\hat{n}_l(kr)$ are the spherical Bessel-Riccatti and Neumann-Riccatti functions, respectively. $V_{\text{stat}}(r)$ is the static potential of the atom determined following the procedure of Salvat *et al.* [32]:

$$V_{\text{stat}}(r) = -\frac{Z}{r} \sum_{n=1}^3 a_n \exp(-\beta_n r), \quad (5)$$

where Z is the nuclear charge, and a_n and β_n are the parameters determined by an analytical fitting procedure to Dirac-Hartree-Fock-Slater self-consistent data [32]. The polarization potential $V_{\text{polar}}(r)$ is expressed in the form proposed by Padial *et al.* [33]:

$$V_{\text{polar}}(r) = \begin{cases} v(r) & r \leq r_c \\ -\alpha/2r^4 & r > r_c \end{cases}, \quad (6)$$

where $v(r)$ is the free-electron-gas correlation energy [34], and α is the static electric dipole polarizability of the atom. The r_c is the first crossing point of the $v(r)$ and $-\alpha/2r^4$ curves [35]. The exchange effects are supposed to be small at the incident energies considered in the present investigation and hence are neglected. The scattering amplitudes for electron scattering on the atom are obtained using the following equation:

$$f(\theta, k) = \frac{1}{2ik} \sum_{l=0}^{l_{\text{max}}} (2l+1)(e^{2i\delta_l} - 1)P_l(\cos\theta) + \pi\alpha k \left(\frac{1}{3} - \frac{1}{2} \sin \frac{\theta}{2} - \sum_{l=1}^{l_{\text{max}}} \frac{P_l(\cos\theta)}{(2l-1)(2l+3)} \right), \quad (7)$$

where $P_l(\cos\theta)$ are Legendre polynomials and the second term in Equation (7) is the Born scattering amplitude for potential of the form (6). In the presented calculations $l_{\text{max}} = 100$. The phaseshifts δ_l are connected with the asymptotic form of the wave function, $u_l(r)$, by:

$$\tan \delta_l = \frac{B_l}{A_l}. \quad (8)$$

The differential cross section for elastic electron scattering from a particular atom was calculated according to:

$$\frac{d\sigma}{d\Omega} = |f(\theta, k)|^2, \quad (9)$$

while atomic integral elastic cross section was derived from the following expression:

$$\sigma = \frac{4\pi}{k^2} \left(\sum_{l=0}^{l_{\text{max}}} (2l+1) \sin^2 \delta_l + \sum_{l=l_{\text{max}}+1}^{\infty} (2l+1) \sin^2 \delta_l^{(B)} \right). \quad (10)$$

Since the above approach has yielded quite encouraging results for molecular targets of tetrahedral symmetry like XY₄ (X=C, Si, Ge; Y=H, F, Cl) and for C₂F₆ [36, 37], it

is expected that the present differential and integral cross sections for electron elastic scattering by C_3F_6 may also be fairly reliable.

3. Results

The present differential cross sections (DCSs) for electron elastic-scattering from C_3F_6 molecule calculated at incident energy ranging from 50 to 1000 eV are shown in Figures 1 and 2.

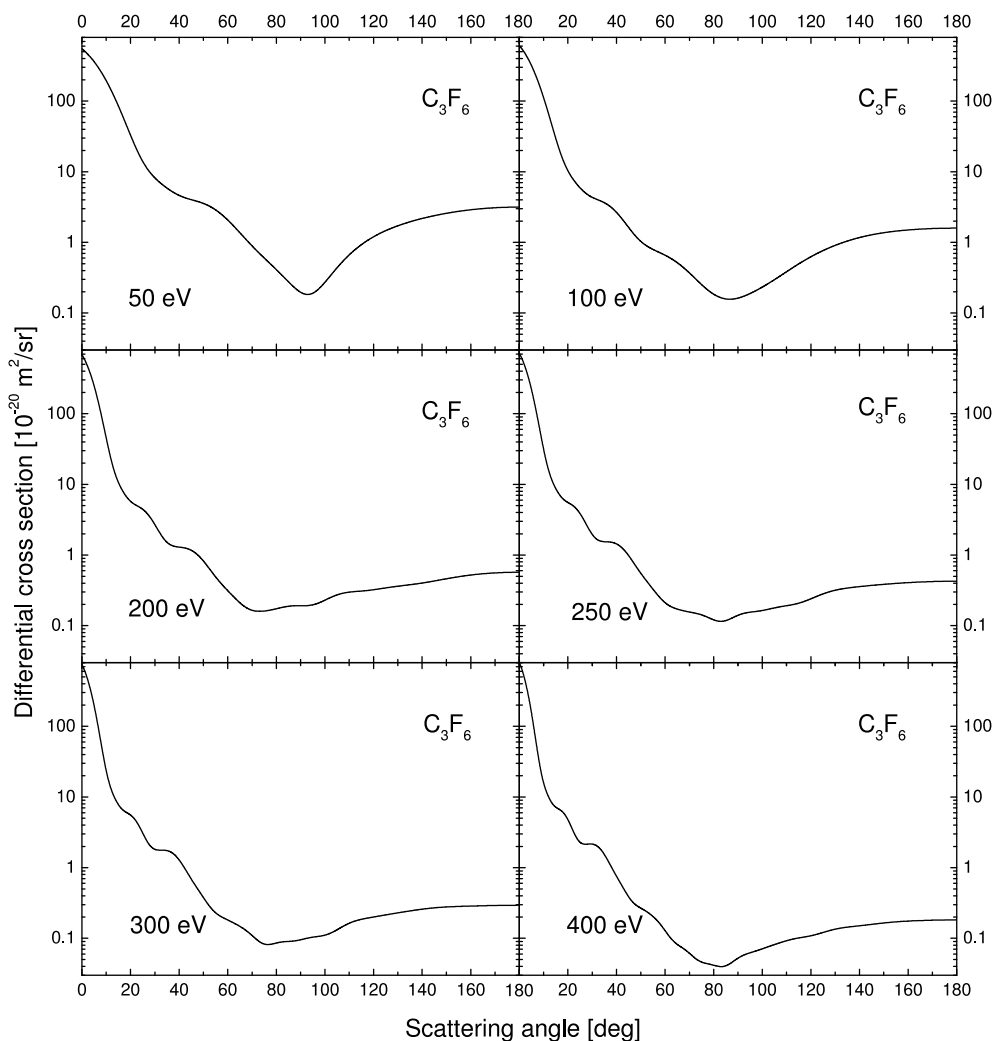


Figure 1. Differential cross section for elastic electron collisions with C_3F_6 molecules at intermediate energies

The values of calculated DCS and integral cross sections (ICSs) at various energies are also presented in Table 1.

There are no elastic measurements and calculations to compare with the present results. It is evident from both figures that, for all investigated energies, the qualitative nature of the calculated DCSs is almost the same. DCS exhibits one distinct minimum

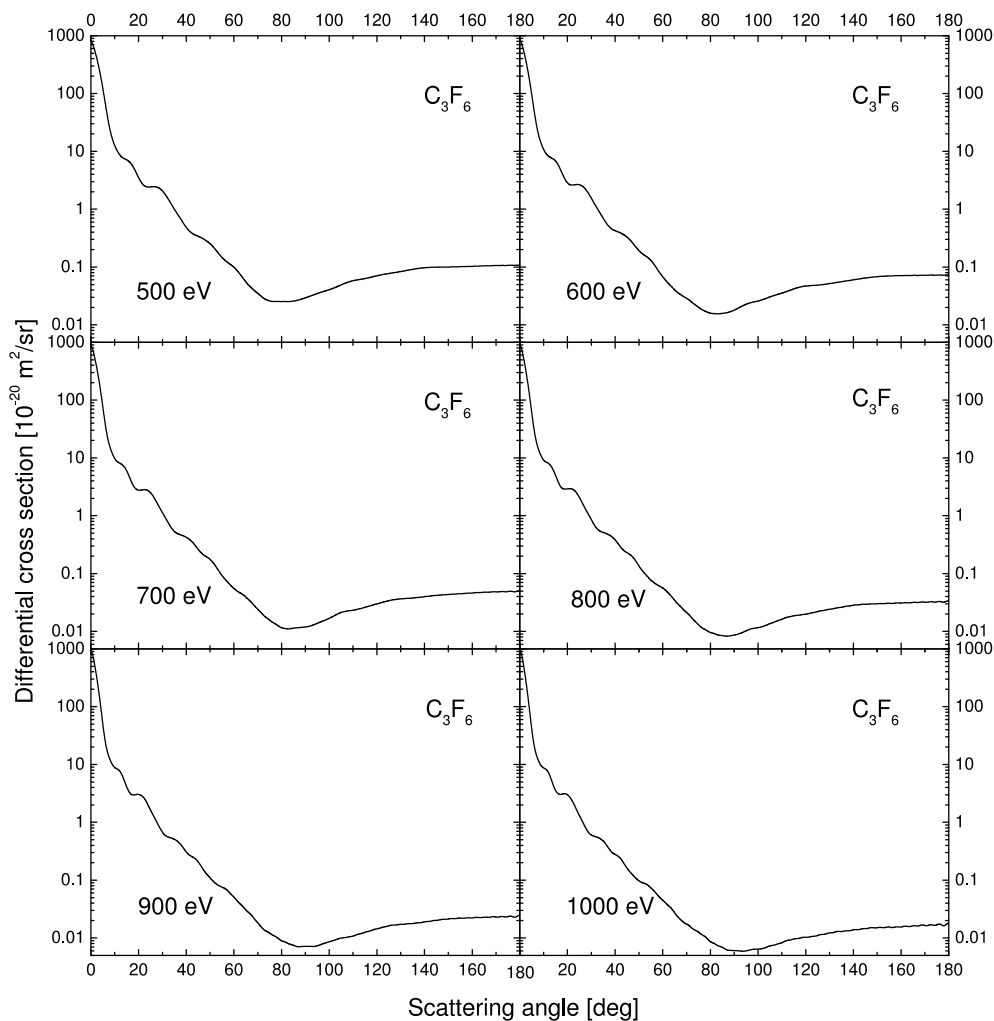


Figure 2. Differential cross section for elastic electron collisions with C_3F_6 molecules at high energies

at the scattering angle located between 80° and 90° . For lower scattering angles, in the $20\text{--}40^\circ$ angular range, some additive weak features are barely discernible. With the increasing incident energy, the DCS increases at near-zero degree scattering angles and decreases for backward scattering. The integral elastic cross section is presented in Figure 3 together with “experimental” elastic cross section estimated as a difference of the absolute *grand* total cross section [14] and the total ionization cross section data [12].

Although, at the investigated energies, inelastic processes are dominated by ionization, the “experimental” elastic cross section evaluated in this way can differ slightly from the true elastic cross section, as we have neglected the contribution from non-ionizing inelastic collisional processes. In the investigated energy range, the elastic cross section is a monotonically decreasing function of the incident electron energy. At the lowest energies studied, *viz.* below 70eV , the applied approximation gives values of

Table 1. Calculated differential (DCS) and integral (ICS) cross sections (in $10^{-20}\text{m}^2/\text{sr}$ and 10^{-20}m^2 , respectively) for $e^- - \text{C}_3\text{F}_6$ elastic collisions

| Angle [deg] | Energy E [eV] | | | | | | | | | | | |
|-------------|-----------------|-------|-------|-------|--------|--------|--------|--------|--------|---------|---------|---------|
| | 50 | 100 | 200 | 250 | 300 | 400 | 500 | 600 | 700 | 800 | 900 | 1000 |
| 0 | 550 | 610 | 682 | 713 | 745 | 796 | 837 | 872 | 906 | 940 | 975 | 1011 |
| 5 | 361 | 317 | 246 | 218 | 194 | 153 | 119 | 93.6 | 74.0 | 59.3 | 48.3 | 40.1 |
| 10 | 194 | 111 | 44.8 | 31.2 | 23.3 | 10.0 | 12.5 | 10.8 | 9.74 | 9.15 | 8.85 | 8.72 |
| 15 | 85.2 | 29.6 | 10.7 | 8.71 | 7.80 | 7.18 | 7.15 | 6.81 | 5.99 | 4.98 | 4.12 | 3.55 |
| 20 | 33.0 | 10.4 | 5.77 | 5.54 | 5.56 | 4.78 | 3.53 | 2.86 | 2.78 | 2.92 | 3.04 | 3.00 |
| 25 | 13.8 | 5.93 | 4.45 | 3.86 | 3.10 | 2.29 | 2.43 | 2.65 | 2.51 | 2.08 | 1.64 | 1.32 |
| 30 | 8.19 | 4.34 | 2.69 | 1.96 | 1.82 | 2.16 | 2.07 | 1.55 | 1.13 | 0.857 | 0.674 | 0.604 |
| 35 | 5.87 | 3.65 | 1.52 | 1.54 | 1.74 | 1.53 | 1.01 | 0.696 | 0.537 | 0.514 | 0.509 | 0.474 |
| 40 | 4.63 | 2.65 | 1.29 | 1.43 | 1.28 | 0.751 | 0.483 | 0.424 | 0.424 | 0.381 | 0.306 | 0.277 |
| 45 | 4.02 | 1.62 | 1.17 | 0.977 | 0.686 | 0.385 | 0.340 | 0.328 | 0.262 | 0.230 | 0.206 | 0.160 |
| 50 | 3.56 | 1.03 | 0.820 | 0.550 | 0.376 | 0.267 | 0.252 | 0.192 | 0.176 | 0.135 | 0.110 | 0.0975 |
| 55 | 2.89 | 0.795 | 0.491 | 0.328 | 0.230 | 0.203 | 0.147 | 0.130 | 0.0926 | 0.0772 | 0.0759 | 0.0735 |
| 60 | 2.08 | 0.658 | 0.313 | 0.211 | 0.182 | 0.128 | 0.0994 | 0.0676 | 0.0554 | 0.0563 | 0.0510 | 0.0450 |
| 65 | 1.37 | 0.515 | 0.210 | 0.171 | 0.147 | 0.0817 | 0.0553 | 0.0400 | 0.0392 | 0.0343 | 0.0307 | 0.0271 |
| 70 | 0.891 | 0.369 | 0.165 | 0.156 | 0.109 | 0.0612 | 0.0351 | 0.0288 | 0.0242 | 0.0212 | 0.0183 | 0.0174 |
| 75 | 0.602 | 0.253 | 0.161 | 0.142 | 0.0828 | 0.0461 | 0.0260 | 0.0202 | 0.0157 | 0.0130 | 0.0122 | 0.0126 |
| 80 | 0.410 | 0.184 | 0.173 | 0.188 | 0.0856 | 0.0414 | 0.0253 | 0.0159 | 0.0116 | 0.00957 | 0.00894 | 0.00861 |
| 85 | 0.276 | 0.158 | 0.188 | 0.118 | 0.0903 | 0.0409 | 0.0258 | 0.0158 | 0.0114 | 0.00846 | 0.00748 | 0.00687 |
| 90 | 0.197 | 0.163 | 0.191 | 0.140 | 0.0952 | 0.0523 | 0.0294 | 0.0180 | 0.0119 | 0.00863 | 0.00717 | 0.00603 |
| 95 | 0.191 | 0.189 | 0.197 | 0.155 | 0.104 | 0.0623 | 0.0348 | 0.0228 | 0.0138 | 0.0103 | 0.00732 | 0.00599 |
| 100 | 0.272 | 0.233 | 0.229 | 0.164 | 0.111 | 0.0709 | 0.0405 | 0.0259 | 0.0168 | 0.0116 | 0.00856 | 0.00641 |
| 105 | 0.439 | 0.297 | 0.273 | 0.178 | 0.132 | 0.0814 | 0.0492 | 0.0305 | 0.0213 | 0.0137 | 0.00998 | 0.00730 |
| 110 | 0.666 | 0.384 | 0.298 | 0.192 | 0.163 | 0.0923 | 0.0580 | 0.0354 | 0.0232 | 0.0163 | 0.0108 | 0.00830 |
| 115 | 0.928 | 0.496 | 0.309 | 0.208 | 0.186 | 0.101 | 0.0633 | 0.0419 | 0.0261 | 0.0186 | 0.0126 | 0.00960 |
| 120 | 1.20 | 0.627 | 0.322 | 0.240 | 0.200 | 0.109 | 0.0706 | 0.0469 | 0.0299 | 0.0199 | 0.0144 | 0.0103 |
| 125 | 1.47 | 0.766 | 0.342 | 0.282 | 0.214 | 0.121 | 0.0773 | 0.0486 | 0.0340 | 0.0220 | 0.0162 | 0.0111 |
| 130 | 1.72 | 0.907 | 0.362 | 0.317 | 0.229 | 0.135 | 0.0836 | 0.0518 | 0.0366 | 0.0242 | 0.0172 | 0.0122 |
| 135 | 1.95 | 1.04 | 0.379 | 0.341 | 0.244 | 0.144 | 0.0912 | 0.0557 | 0.0378 | 0.0262 | 0.0178 | 0.0129 |
| 140 | 2.18 | 1.17 | 0.401 | 0.357 | 0.258 | 0.150 | 0.0972 | 0.0601 | 0.0399 | 0.0284 | 0.0190 | 0.0139 |
| 145 | 2.39 | 1.28 | 0.429 | 0.371 | 0.270 | 0.157 | 0.0995 | 0.0645 | 0.0420 | 0.0295 | 0.0202 | 0.0145 |
| 150 | 2.58 | 1.37 | 0.461 | 0.384 | 0.278 | 0.165 | 0.100 | 0.0680 | 0.0435 | 0.0301 | 0.0213 | 0.0150 |
| 155 | 2.75 | 1.44 | 0.493 | 0.396 | 0.283 | 0.171 | 0.101 | 0.0702 | 0.0450 | 0.0307 | 0.0221 | 0.0155 |
| 160 | 2.90 | 1.50 | 0.521 | 0.406 | 0.286 | 0.175 | 0.103 | 0.0709 | 0.0461 | 0.0309 | 0.0222 | 0.0155 |
| 165 | 3.00 | 1.54 | 0.543 | 0.415 | 0.289 | 0.178 | 0.105 | 0.0718 | 0.0477 | 0.0318 | 0.0230 | 0.0162 |
| 170 | 3.01 | 1.57 | 0.558 | 0.421 | 0.291 | 0.180 | 0.106 | 0.0720 | 0.0483 | 0.0321 | 0.0231 | 0.0165 |
| 175 | 3.15 | 1.59 | 0.567 | 0.424 | 0.293 | 0.182 | 0.107 | 0.0724 | 0.0489 | 0.0325 | 0.0234 | 0.0169 |
| 180 | 3.16 | 1.59 | 0.571 | 0.426 | 0.294 | 0.183 | 0.108 | 0.0738 | 0.0503 | 0.0339 | 0.0247 | 0.0181 |
| $\sigma(E)$ | 38.1 | 22.7 | 14.7 | 12.8 | 11.4 | 9.42 | 8.11 | 7.14 | 6.4 | 5.8 | 5.31 | 4.91 |

integral elastic cross section which distinctly exceed the experimental estimation. This disagreement may be due to neglecting the bond distortion and multiple scattering at impact energies at which electron wavelength becomes comparable to the internuclear distances. Above 70eV, agreement between theory and experiment is satisfactory. Based on our calculations, we have estimated that, for energies ranging from 100 to 370eV, the elastic processes in the C_3F_6 molecules constitute up to 70–60% of the total cross section, which seems to be characteristic for perfluorinated targets [14]. It is well known that total cross sections measured with a transmission technique can

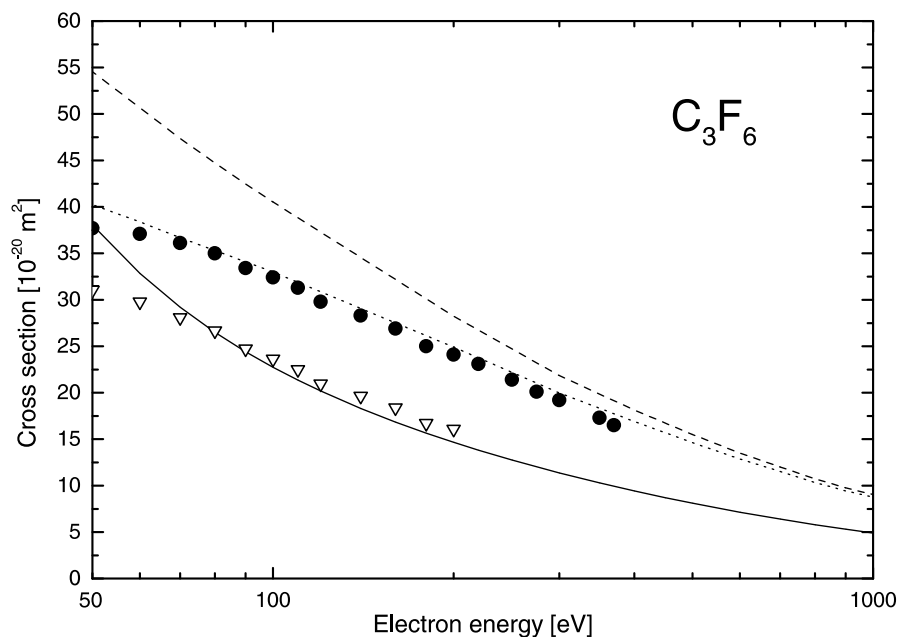


Figure 3. Comparison of integral cross sections: — elastic, present calculations (IAM); ∇ experimental elastic, estimated as a difference of absolute total [14] and ionization [12] cross sections; \bullet experimental absolute total cross section [14]; - - - - theoretical total cross section (EGAR model) [13]; - · - · - theoretical total cross section (AR model) [13]

be lowered by incomplete discrimination of electrons which are scattered throughout small angles in the forward direction [38]. This is mainly due to electrons which undergo elastic scattering, as the electrons which are scattered inelastically in the very-near-forward direction can be discriminated using an electron-energy filter. The uncertainty in the total cross section, related to this effect, increases with the impact energy. The amount, $\Delta\sigma_{\text{tot}}(E)$, by which the total cross section is lowered due to this effect can be roughly estimated as follows:

$$\Delta\sigma_{\text{tot}}(E) = \left. \frac{d\sigma}{d\Omega} \right|_{\theta=0} \Delta\Omega, \quad (11)$$

where $d\sigma/d\Omega|_{\theta=0}$ is the differential cross section for elastic electron scattering at zero degree angle and $\Delta\Omega$ is the angular acceptance of the detector. Having in hand angular distributions of scattered electrons calculated in the present work, we can use them to estimate the $\Delta\sigma_{\text{tot}}(E)$ for absolute experimental C_3F_6 data [14]. Respective $\Delta\sigma_{\text{tot}}(E)$ values do not exceed 2% for 200–250eV, 2.5% at 350eV and 3% at 370eV.

4. Conclusions

In this work the differential and integral cross sections for the elastic scattering of electrons from hexafluoropropene have been calculated, for impact energies from 50 to 1000 eV. The present results, obtained using the independent atom approximation with static-polarization model potential, are in reasonable agreement with elastic cross sections derived from the experimental total and ionization cross sections.

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