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Total cross sections for electron scattering with some C₃ hydrocarbons

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Abstract

Absolute total cross sections (TCSs) for electron scattering from two C₃H₄ isomers (allene and propyne) and from propane (C₃H₈) have been measured in a linear electron-beam transmission experiment for impact energy ranging from 0.5 to 370 eV. Low-energy TCS functions for C₃H₄ are dominated by prominent structures: a resonant-like enhancement of the cross section for allene peaks at around 2.3 and 3.4 eV for propyne, while very broad enhancement is centred at 9.5–10 and 8.0–8.5 eV for allene and propyne respectively. Some supplementary weak features are also discernible. The general shape of cross sections for both C₃H₄ isomers is similar except that the cross section enhancements are at different locations. In addition, the TCS for propyne has been compared with cross sections for other open-chain hydrocarbons with three carbon atoms, i.e. propene (C₃H₆) and propane (C₃H₈), and the effect of the multiplicity of the C–C bond on the low-energy scattering is demonstrated.

1. Introduction

Electron scattering data for hydrocarbons are important for modelling electron-assisted processes ranging from fuel combustion to interstellar clouds (see Kaifu (1990), Morgan (2000) and Tanaka and Inokuti (2000) and references therein). Although numerous experimental investigations of electron interaction with hydrocarbon molecules have appeared in the literature, the available results are still fragmentary and mostly concern the simplest compounds (e.g. CH₄).

A systematic experimental study of the relationship between molecular structure and the shape and magnitude of the electron-scattering total cross section (TCS) was initiated by Brüche (1927, 1930) and Schmieder (1930). More recently, further extensive TCS experimental studies for hydrocarbonic series have been carried out by Floeder *et al* (1985), Sueoka and Mori (1986), Nishimura and Tawara (1991) and Lunt *et al* (1998). To provide new experimental

data, which may give some further insight into the role of the geometrical arrangement of atoms in a target molecule in the electron scattering process, we have measured absolute electron-scattering TCSs for C_3H_4 , the smallest hydrocarbon with geometrical isomers. In the present work the measurements were carried out for two C_3H_4 isomers: allene (1, 2-propadiene) and propyne (methylacetylene). To date, descriptions in the literature of the processes involved in electron collision with allene molecules have been sparse and are limited to low-energy trapped-electron spectra measured by Knoop (1972), the electron impact excitation (Mosher *et al* 1975) and the electron transmission spectrum (Ciommer *et al* 1984). More attention has been dedicated to electron scattering from another C_3H_4 isomer, propyne. Bowman and Miller (1965), Knoop (1972), Dance and Walker (1974) and recently Palmer *et al* (1999) measured the trapped-electron near-threshold excitation spectra at subionization energies. The electronic excitation of a propyne molecule at intermediate electron impact energies was investigated by Stradling *et al* (1976), Flicker *et al* (1978) and Fridh (1978). Giordan (1983) and Ciommer *et al* (1984) recorded the derivative with respect to impact energy of the electron current transmitted through the propyne. The appearance potentials and abundance of positive ions formed in electron-propyne scattering were determined by Franklin and Mogenis (1967) while negative ion formation via dissociative electron attachment was studied by Rutkowsky *et al* (1980). With the exception of the electron attachment data, intensities obtained in the aforementioned experiments are given in relative scale only.

To verify whether a change in the character of the carbon-carbon bond reflects a TCS energy dependence, we used a series of open-chain C_3 hydrocarbon molecules of different C-C bond multiplicities: C_3H_4 , C_3H_6 and C_3H_8 . Even though there is enough e^- - C_3H_8 TCS data (Brüche 1930, Schmieder 1930, Floeder *et al* 1985, Nishimura and Tawara 1991, Tanaka *et al* 1999) to compare with cross sections for propyne and propene, we have measured the TCS for propane to provide consistent data obtained in the same laboratory.

2. Experimental details

The absolute TCSs presented in this paper were determined by the linear electron-beam transmission method under single-collision conditions. In an ideal transmission experiment (cf Bederson and Kieffer 1971), the total scattering cross section $Q(E)$ at given impact energy E can be obtained from the Bouguer-de Beer-Lambert law

$$I(E, n) = I(E, 0) \exp[-nlQ(E)], \quad (1)$$

where $I(E, n)$ and $I(E, 0)$ are the intensities of the projectile beam transmitted through a scattering volume of length l in the presence of a target with density number n or in its absence ($n = 0$), respectively.

The apparatus used for the reported measurements has been intensively exploited in a series of experiments (e.g. Szmytkowski and Kwitniewski 2002) and only a brief outline of the apparatus and the experimental procedure will be given here. A beam of quasi-monoenergetic electrons ($\Delta E \simeq 70$ meV, FWHM) was produced by an electron gun followed by a 127° cylindrical condenser working as an energy dispersing element. An electron-optical lens system controlling the trajectory and the energy of the electron beam directs it into the scattering cell filled with the molecules under study. The electrons which succeeded in leaving the interaction region through the exit orifice of the scattering chamber were discriminated on their route to the detector with a retarding field element which prevents inelastically scattered electrons being collected with a Faraday cup.

Inevitable target effusion throughout the orifices of the scattering cell causes the determination of the effective value of the product nl in expression (1) to be one of the critical

points in measurements of accurate absolute TCSs. Following the effective scattering path-length calculations of Nelson and Colgate (1973), we deduced that for the present scattering cell geometry, the effect of density drop across the orifices of the scattering chamber is nearly compensated for by accompanying lengthening of the target region and that the geometrical distance between orifices ($L = 30.5$ mm) can be, to within 0.5%, adopted as the effective absorption length. The absolute number density, n , was evaluated from the ideal gas law based on absolute measurements of a gas-target pressure in the centre of the reaction volume and the gas temperature, taking into account a thermal transpiration effect according to the Knudsen (1910) formula. The gas effusing through orifices of the scattering cell into the electron optics volume may affect the intensity of the primary electron beam and in consequence the measured cross section. To avoid such variations, the measurements were carried out in such a way that the background pressure in the region of the electron optics was constant, independently of whether the target was present or not in the scattering chamber. The pressure in the electron optics volume was always three orders of magnitude lower than the pressure in the scattering cell (~ 0.1 Pa).

Another serious problem constantly arising in measurements of absolute TCSs, which tends to lower the measured cross sections, is an imperfect discrimination by the detector system against electrons which are scattered through small angles in the forward direction and contribute to the transmitted current. For electron scattering, the error associated with this effect is usually higher for molecules with a permanent electric dipole moment and it increases with impact energy. Estimation of this effect and the correction of the measured cross section would be possible if the angular distributions of electrons scattered on target molecules were known. To date, no such data are available for C₃H₄ isomers. Rough estimation, based on differential cross section data for other targets, shows that the lowering of the measured TCS should not exceed 4% at the highest applied energies for propyne and should be less than 3% for allene.

As a reference for the absolute electron energy scale, the well-known resonant structure in N₂ was chosen; a series of oscillations in the transmission current at around 2.3 eV was visible when some amount of nitrogen was admitted to the gas target under study. The TCSs obtained at the same incident energy appeared to be independent, within the random experimental uncertainties, of the applied pressure and the electron-beam intensity. The final TCS value at each particular energy is a weighted mean of data obtained in independent series (4–12) of individual runs (7–10 in a series). The statistical uncertainty (one standard deviation of the weighted mean value) of the measured TCS is nearly 2% below 1 eV and well below 1% over all other investigated energies. The direct sum of all potential systematic uncertainties of measured individual quantities used for the TCS derivation was estimated to be 7–8% below 2 eV, gradually decreasing to 5% in the range 10–100 eV and increasing again to 8–9% at the highest energies applied.

Commercially available gas samples (Sigma–Aldrich: allene, 97%; propyne, 98%; propane, 98%) were used without any further purification.

3. Results and discussion

3.1. Allene, H₂C=C=CH₂

The present absolute TCSs for electron collision with allene molecule, and the associated errors, are shown in figure 1 as a function of impact energy over the range from 0.5 to 370 eV. Table 1 presents the measured cross section in numerical form.

According to the behaviour of the measured e⁻–allene TCS energy dependence, the whole investigated energy range can be conveniently divided into some distinct regions. In the first, ranging from 0.5 to about 1 eV, the TCS varies very slowly with the energy increase. The next

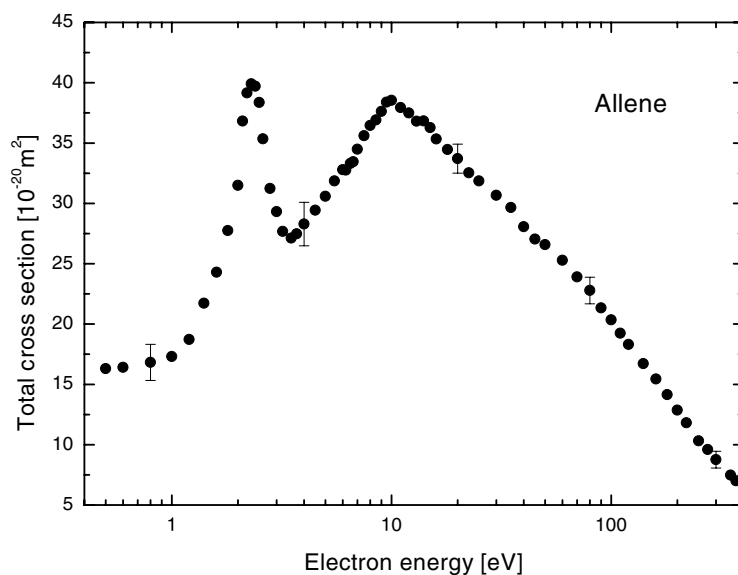


Figure 1. Total electron–C₃H₄ (allene) scattering cross sections. (●) present results; the error bars at selected points represent the overall (systematic plus statistical) experimental uncertainties.

Table 1. Absolute TCSs measured for electron impact on allene (CH₂CCH₂) in units of 10^{−20} m².

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.5	16.3	3.2	27.7	10	38.5	70	23.9
0.6	16.4	3.5	27.1	11	37.9	80	22.8
0.8	16.8	3.7	27.5	12	37.5	90	21.3
1.0	17.3	4.0	28.3	13	36.8	100	20.4
1.2	18.7	4.5	29.4	14	36.8	110	19.2
1.4	21.7	5.0	30.6	15	36.3	120	18.3
1.6	24.3	5.5	31.9	16	35.3	140	16.7
1.8	27.8	6.0	32.8	18	34.5	160	15.4
2.0	31.5	6.2	32.8	20	33.7	180	14.2
2.1	36.8	6.5	33.3	22.5	32.5	200	12.9
2.2	39.2	6.7	33.4	25	31.9	220	11.8
2.3	39.9	7.0	34.5	30	30.7	250	10.3
2.4	39.7	7.5	35.6	35	29.7	275	9.60
2.5	38.4	8.0	36.5	40	28.1	300	8.76
2.6	35.4	8.5	36.9	45	27.0	350	7.47
2.8	31.2	9.0	37.6	50	26.6	370	7.00
3.0	29.3	9.5	38.4	60	25.3		

energy region, spanning 1–3.5 eV, is dominated by a very intense and narrow (~ 0.8 eV) TCS enhancement, peaking near 2.3 eV with a maximum intensity of $40 \times 10^{-20} \text{ m}^2$. The magnitude of the enhancement, with respect to slower rising background, is extraordinary and it amounts to nearly $17 \times 10^{-20} \text{ m}^2$. Such a significant TCS structure is most probably associated with the formation of a shape-resonant state when the projectile electron is temporarily attached to the target molecule. Some indication on the resonant character of the electron scattering around

Table 2. Absolute TCSs measured for electron impact on propyne (CH₃CCH) in units of 10⁻²⁰ m².

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.5	18.9	3.3	41.7	10	37.5	70	23.1
0.6	18.7	3.4	42.1	11	36.4	80	21.9
0.8	19.1	3.5	41.6	12	35.4	90	20.7
1.0	19.6	3.7	40.0	13	35.2	100	19.7
1.1	20.0	4.0	38.0	14	35.0	110	18.8
1.2	20.2	4.5	35.8	15	34.6	120	17.8
1.4	20.8	5.0	35.1	16	34.0	140	16.4
1.6	22.0	5.5	35.2	18	32.8	160	15.0
1.8	22.8	6.0	36.0	20	31.5	180	13.7
2.0	24.2	6.5	36.7	25	30.4	200	12.4
2.2	25.9	7.0	37.7	30	29.1	220	11.4
2.4	28.6	7.5	38.4	35	28.3	250	10.1
2.6	32.0	8.0	38.8	40	27.3	275	9.31
2.8	35.4	8.5	38.9	45	26.4	300	8.54
3.0	39.3	9.0	38.5	50	25.8	350	7.56
3.2	41.2	9.5	38.0	60	24.1	370	7.09

2 eV comes from the excitation spectrum of Knoop (1972) and the derivative transmission spectra of Ciommer *et al* (1984). The excitation data demonstrate that the resultant parent anion, (C₃H₄)^{*}, created in this energy range, decays preferably via autodetachment of the extra electron, leading to vibrational excitation of the allene molecule in its electronic ground state. Following the TCS deep minimum, located at 3.5 eV, the cross section increases again up to a value of $38 \times 10^{-20} \text{ m}^2$ in the vicinity of 9.5–10 eV. The broad TCS enhancement, centred close to 10 eV, resembles those observed for many targets studied so far and which have been accounted for by elastic scattering with some contribution from a number of weak inelastic components allowed at these energies (Knoop 1972), among them also resonant ones (Winstead *et al* 1992, Boesten *et al* 1994). Two weak features superimposed on both sides of the enhancement around 10 eV are also worth noting: a barely visible shoulder near 6 eV on the low-energy slope, and a more pronounced structure located at around 14 eV on the descending high-energy slope of the enhancement. These features are probably associated with the formation of resonances when the extra electron is bonded to excited electronic states of the target molecule. From 15 eV up to the highest energy studied, the TCS decreases steadily along with impact energy; nevertheless, another very broad and weak feature is discernible around 70–80 eV, close to the typical location of the maximum in the total ionization cross section for hydrogen-containing compounds.

3.2. Propyne, H₃C–C≡CH

The results of our absolute measurements for propyne are presented in figure 2, showing the energy dependence of the total electron scattering cross section from 0.5 to 370 eV. Numerical values of our TCS data are provided in table 2.

In the energy range investigated, the total electron-scattering cross section for the propyne molecule has two very distinct enhancements. The first strong feature of $42 \times 10^{-20} \text{ m}^2$ peaks at 3.4 eV. This pronounced structure is superimposed onto the low-energy slope of the very broad hump centred at around 8.0–8.5 eV, with a maximum value of $39 \times 10^{-20} \text{ m}^2$. The increment of the TCS around 3.4 eV is mainly related to the temporary accommodation

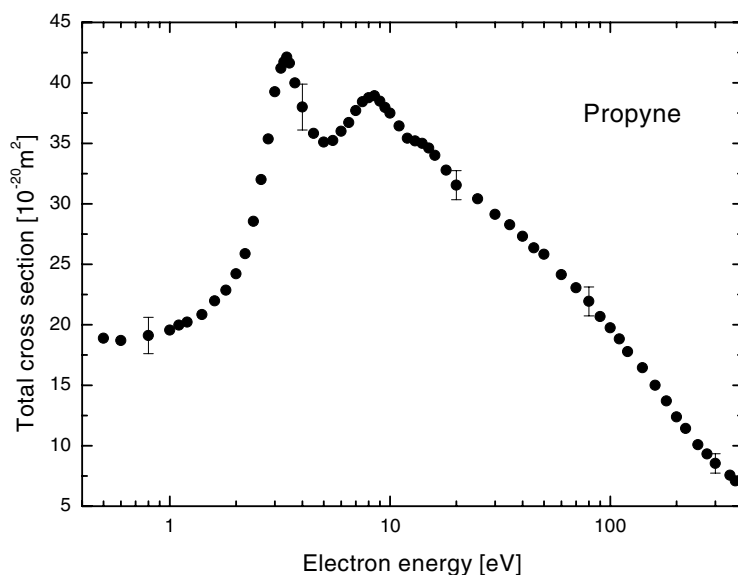


Figure 2. Total electron–C₃H₄ (propyne) scattering cross sections. (●) present results; the error bars at selected points represent the overall experimental uncertainties.

of the incident electron into the lowest unoccupied π^* orbital of the molecule (Palmer *et al* 1999), involving formation of the shape-resonant state which subsequently decays via autodetachment of the extra electron and/or via a dissociative channel. The autodetachment decay channel was evidenced in the low-energy experiments of Bowman and Miller (1965), Knoop (1972), Dance and Walker (1974) and Palmer *et al* (1999). In the near-threshold excitation spectra they observed a weak feature peaking between 2.8–3.0 eV and attributed it to a vibrational excitation of the parent molecule in its electronic ground state. The formation of the negative-ion resonance near 3 eV, decaying via the dissociative channel, was demonstrated by Rutkowsky *et al* (1980), although their absolute data also indicate that the dissociative electron attachment does not play a significant role in the resonant e^- –propyne scattering. A transmission experiment of Giordan (1983) positioned the resonance at 3.14 eV while similar measurements of Ciommer *et al* (1984) localized it close to 2.8 eV. For energies above 8.5 eV, the TCS function decreases but in the vicinity of 14 eV a weak structure can be distinguished. This feature may be associated with the formation of resonances correlated with the excited electronic states of molecules. At the lowest applied energies, below 1 eV, the TCS seems to increase as the incident energy falls; this behaviour may be related to the permanent electric dipole moment ($\mu = 0.78$ D) of propyne.

3.3. Propane, C₃H₈

The absolute TCS for electron–propane scattering measured in this work is shown in figure 3 and compared with recent TCS experimental data of Floeder *et al* (1985), Nishimura and Tawara (1991) and Tanaka *et al* (1999). Pioneering measurements of Brüche (1930) and Schmieder (1930) are also included for completeness. Table 3 presents our TCS data in numerical form.

In the energy range studied, the e^- –C₃H₈ TCS curve has only one very broad enhancement with a maximum of 50×10^{-20} m² located at 8.5 eV. Such behaviour of the TCS energy dependence is typical for open-chain saturated hydrocarbons (Brüche 1930). Some change in

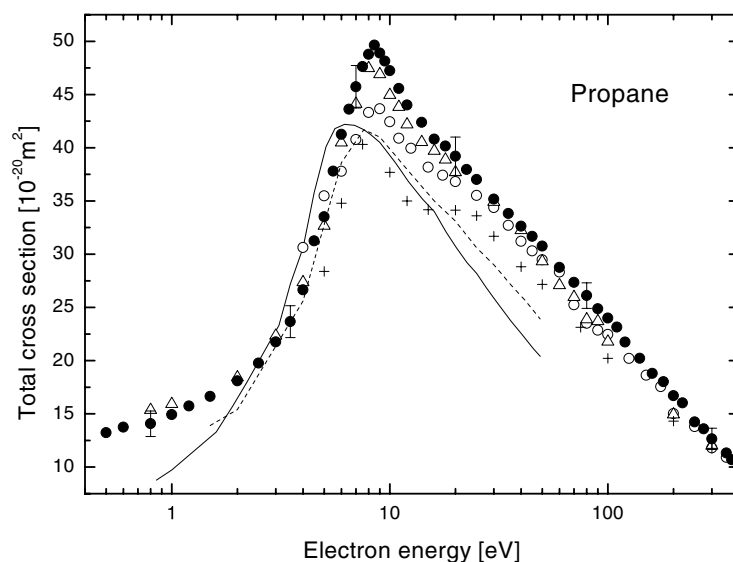


Figure 3. Experimental total electron-C₃H₈ scattering cross sections: (●) present results; the error bars at selected points represent the overall (statistical plus systematic) uncertainties; (Δ) (Tanaka *et al* 1999); (○) Nishimura and Tawara (1991); (+) Floeder *et al* (1985); (—) Brüche (1930); (- - -) Schmieder (1930).

Table 3. Absolute TCS measured for electron impact on C₃H₈ (propane) in units of 10⁻²⁰ m².

Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS	Energy (eV)	TCS
0.5	13.2	6.0	41.3	20	39.2	120	21.7
0.6	13.8	6.5	43.6	22.5	38.0	140	20.2
0.8	14.1	7.0	45.7	25	37.0	160	18.8
1.0	14.9	7.5	47.6	30	35.2	180	18.0
1.2	15.7	8.0	48.8	35	33.8	200	16.7
1.5	16.6	8.5	49.6	40	32.6	220	16.0
2.0	18.1	9.0	48.9	45	31.7	250	14.2
2.5	19.8	9.5	48.1	50	30.8	275	13.6
3.0	21.8	10	47.2	60	29.0	300	12.6
3.5	23.7	11	45.6	70	27.4	350	11.3
4.0	26.6	12	44.0	80	26.1	370	10.7
4.5	31.2	14	42.4	90	24.9		
5.0	33.5	16	40.8	100	24.0		
5.5	37.8	18	40.2	110	23.1		

the slope of the TCS function is visible at around 15 eV. In general, the energy dependence of all experimental TCSs displayed in figure 3 is similar, though some divergencies in absolute cross section values obtained in different laboratories are visible. The present results consistently exceed all previous experimental data and the greatest difference in the magnitude appears in the vicinity of the TCS maximum where disaccord between our results and absolute values of Floeder *et al* (1985) reaches 25% and, with those of Nishimura and Tawara (1991), up to about 15%. The agreement with very recent results of Tanaka *et al* (1999) obtained using the time-of-flight discrimination is very good over the entire common energy range of measurements.

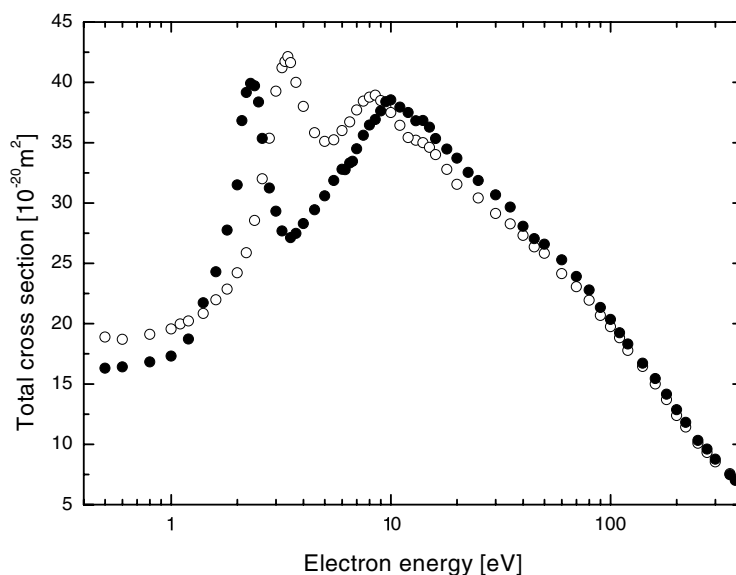


Figure 4. Comparison of total electron scattering cross sections: (●) allene; (○) propyne.

3.4. Isomeric effect

In figure 4 we compare the electron-scattering TCSs for two investigated C_3H_4 isomers, allene and propyne. Allene and propyne contain the same constituent atoms but in each molecule they are arranged in a different way. Whereas the allene is an open-chain molecule with two adjacent carbon-carbon double bonds and with two CH_2 groups twisted with respect to each other by 90° , the propyne has one triple and one single C-C bond with, respectively, one methyl and one CH group attached on the edges of each molecule. In spite of different geometry of both isomers, the shape of their TCSs appears to be very similar. Both compared TCS energy functions exhibit two very distinct features: a narrow resonant-like maximum in the range 2–4 eV superimposed on the low-energy slope of a very broad hump centred between 8 and 10 eV. On the right-hand side of the hump near 14 eV, a shoulder and, at around 70–80 eV, a change of the slope can be easily distinguished in both compared curves.

Examination of the compared cross sections at lower impact energies, where resonant processes become important, also reveals some quite distinct quantitative differences:

- (i) the low-energy TCS enhancement for allene is centred at 2.3 eV while that for propyne is shifted to a higher energy (3.4 eV);
- (ii) the width of the 2.3 eV peak for allene is about 0.8 eV while that for propyne, at 3.4 eV, is nearly twice as broad (~ 1.4 eV);
- (iii) the maximum of the second TCS enhancement for allene is located near 9.5–10 eV, that is about 1.5 eV above the second maximum for propyne. In addition, in the vicinity of 6 eV the TCS for allene has a weak structure which could not be discernible for propyne;
- (iv) at energies below 9.5 eV, the TCS for propyne is generally higher than that for allene, probably due to the permanent electric dipole moment of propyne ($\mu = 0.78$ D), whereas
- (v) from 9.5 up to about 150 eV the TCS for propyne becomes higher.

It is also significant to note that, at intermediate energies, the TCS for $e^- - C_3H_4$ scattering seems to become independent of the geometrical arrangement of atoms in a target molecule and the magnitude of both cross sections coincides.

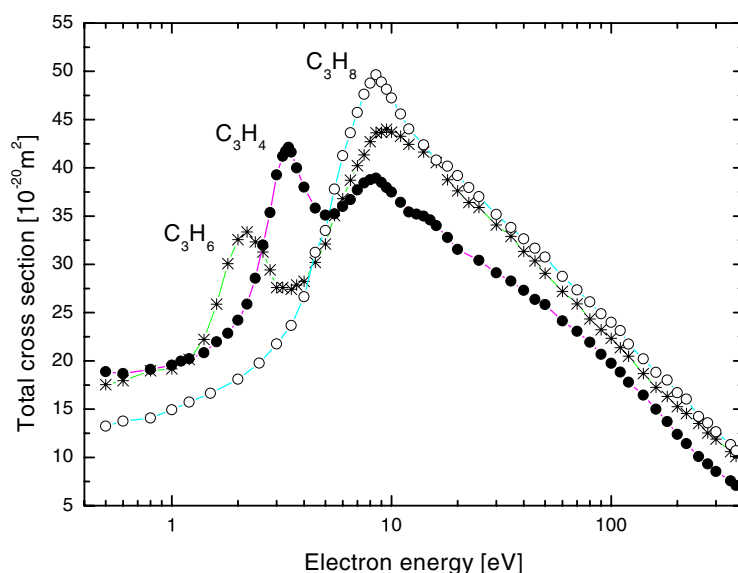


Figure 5. Comparison of electron-scattering TCSs for open-chain C₃ hydrocarbon molecules: (●) propyne in the present work; (*) propene, Szymtkowski and Kwitniewski (2002); (○) propane in the present work.

(This figure is in colour only in the electronic version)

3.5. C₃ open-chain hydrocarbons

In figure 5 the electron-scattering TCSs for propyne, propene and propane are compared. All these targets are small open-chain hydrocarbon molecules with three carbon atoms and a methyl group on one end of molecule but they differ in multiplicity of C–C bonds. One would expect therefore that this difference would be reflected in the low-energy electron scattering TCSs for the compared molecules. Indeed, we observe that the low-energy resonant maximum for propyne (C≡C–C) is located at 3.4 eV, for propene (C=C–C) is centred near 2.3 eV, while propane (C–C–C) has only the very broad enhancement located between 8 and 10 eV, which is also a common feature for other compared targets. It is worth noting that the resonant maximum for molecules with a double bond (e.g. allene in the present work; ethylene, Lunt *et al* (1994)) is usually located at lower energies than for targets with triple bonds (cf acetylene, Sueoka and Mori (1989)).

At impact energies above 7 eV, the compared molecules of larger geometrical size have higher TCSs. However, at lower energies the relation reverses, which may be related to the dipole interaction dominating the collisional dynamics in the low-energy electron scattering by polar molecules; permanent dipole moments for the compared molecules are in inverse relation to their molecular size.

4. Conclusions

The work presented here provides absolute TCSs for electron scattering from two geometrical isomers of C₃H₄ (allene and propyne) and from the C₃H₈ molecule for incident electron energies ranging from 0.5 up to 370 eV. The general shape of the cross sections for the pair of C₃H₄ isomers appears to be quite similar. TCS curves for C₃H₄ have one low-energy resonant-

like feature followed by the broad enhancement on the right-hand side of which a shoulder is discernible. The main difference concerns the location of these features: 2.3, 8.5–9 and 14 eV in the allene TCS energy function and 3.4, 8–8.5 and 14 eV for propyne. There is also some difference in the magnitude of the cross section for both isomers. Below 9 eV the TCS for propyne is generally higher and above 9 eV the relation between the magnitudes of cross sections reverses until about 150 eV where the magnitudes become the same. Comparison of the TCSs for selected open-chain C₃ hydrocarbons (propyne, propene and propane) demonstrates the effect of the multiplicity of carbon–carbon bonds on the location of the low-energy resonant enhancements.

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