

# Total cross sections for electron scattering by CF<sub>4</sub>, CCl<sub>4</sub>, CF<sub>2</sub>Cl<sub>2</sub>, CClF<sub>3</sub> and CFCI<sub>3</sub> at 30–5000 eV: A modified additivity rule approach

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## Abstract

The total cross sections for electron scattering by chlorofluoromethane molecules (CF<sub>4</sub>, CCl<sub>4</sub>, CF<sub>2</sub>Cl<sub>2</sub>, CClF<sub>3</sub> and CFCI<sub>3</sub>) are calculated using a modified additivity rule approach, which takes into consideration the target molecular dimension and the geometric shielding effect in a molecule. The atoms are presented by a spherical complex optical potential which is composed of static, exchange, polarization and absorption terms. The total cross sections are quantitatively compared with those attained by measurements and other theories wherever available at 30–5000 eV. Good agreement is obtained above 100 eV.

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## 1. Introduction

The total cross section (TCS) for electron–molecule scattering has important applications in plasma-assisted processing, gaseous laser, chemical etching, atmospheric science, semiconductor devices and military fields [1]. For electron–atom scattering, a number of successful approximate methods are available over a wide energy range. However, for electron–molecule scattering, few equivalent approaches exist for arbitrary atomic composition over the corresponding energy range, especially at intermediate and high energies. The reason is that the electron–molecule scattering presents a more complex problem than the electron–atom scattering due to the multicenter nature, the lack of a center of symmetry (in the case of polyatomic

and hetero-nuclear molecules) and its nuclear motion. In order to deal with the electron–molecule scattering over a wide energy range, many new approximation methods have been proposed and developed in recent years. In these methods, the additivity rule (AR) model [2] is a fairly simple but practical one, particularly for simpler and smaller molecules at high enough energies. However, for relatively larger molecules or complex polyatomic molecules [3,4], great discrepancies can still be seen even at energy of several hundreds of eV.

In order to extend the AR model validity to lower energies, many investigations have been made in recent years. Bobeldijk et al. [5] presented two geometric AR methods, and obtained encouraging electron impact ionization cross sections of several molecules at energy of several tens of eV, but they did not calculate the TCSs of electron scattering by molecules. Deutsch et al. [6] applied a modified AR (with atomic weighted factors) to molecular ionization cross sections, but they did not calculate the TCSs of electron scattering by molecules, too. Joshipura et al. [7]

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separated the polarization interaction from the optical potential, and obtained atomic cross sections from the remaining interactions. Then, they summed atomic cross sections with the cross section for scattering on the molecular polarization potential. Employing this approach, they calculated the TCS for simple molecules and improved their results in a way. Karwasz et al. [8] proposed a formation of the AR, where a molecular cross section can be approximated by a Born-like two-parameter formula. Though this method can attain good results over a wide energy range, the applications are limited since the two adjustable values are different for each target, and their value determinations require experimental findings. Recently, to find the TCSs of electron scattering by molecules, Joshipura et al. [9] presented a modified AR, named group additivity method, where the cross section arising from each constituent group of a molecule was added together to obtain the TCS. Using this method, Joshipura et al. have calculated the TCSs of electron scattering by several molecules such as  $\text{CF}_4$ ,  $\text{C}_2\text{F}_4$ ,  $\text{C}_2\text{F}_6$ ,  $\text{C}_3\text{F}_8$  and  $\text{CF}_3\text{I}$ , and good agreement with experiments is obtained over a wide energy range. Jiang et al. [10] have presented a modification method named the energy-dependent geometric AR approach, and calculated the TCSs for electron scattering by numbers of molecules. Since the target molecular dimension was not considered, obvious deviations can still be seen at lower energies for relatively larger molecules or complex polyatomic molecules. To improve the agreement between theories and experiments, in this paper, we will take into consideration the shielding effect of atoms in a molecule and discuss another modification method based on the AR model, which depends on the target molecular dimension and the energy of the incident electrons.

## 2. Methods

In the AR model, the molecular orbital is described by the sum of the valence orbitals of all atoms present in the molecule. Thus the AR model can be written as

$$Q_{\text{AR}}(E) = \frac{4\pi}{k} \text{Im} F_m(\theta) \approx \frac{4\pi}{k} \text{Im} \sum_{j=1}^N f_j(\theta=0) = \sum_{j=1}^N q_T^j(E), \quad (1)$$

where  $F_m(\theta=0)$  is the electron-molecule scattering amplitude for forward direction;  $q_T^j(E)$  and  $f_j(\theta=0)$  are the TCS and the complex scattering amplitude for the  $j$ th constituent atom, respectively.

In Eq. (1), one main effect is not considered: a close-packed molecule is not fully transparent for low-energy electrons, and the “inner” atoms are partially shielded by the “outer” atoms and do fewer contributions to the molecular TCSs at lower energies than those at higher energies. The shielding effect, which leads to negative contributions to the TCSs in the AR model, is dependent on the molecular geometry. Accounting for the shielding effect,

Bobeldijk et al. [5] presented a geometric additivity rule (GAR) to calculate the ionization cross section. It is obvious that the GAR model, which incorporates the geometry of a molecule, is a good approach to obtain the TCSs for electron scattering by molecules, particularly for complex polyatomic molecules or relatively larger molecules at low energies [10]. When the GAR model is applied to calculate the TCS, it can be expressed as

$$Q_{\text{GAR}}(E) = \frac{1}{3} Q_{\parallel}(E) + \frac{2}{3} Q_{\perp}(E), \quad (2)$$

where  $Q_{\parallel}(E)$  and  $Q_{\perp}(E)$  are the TCS for electrons approaching the molecule parallel to the  $Z$  axis ( $\theta=0^\circ$ ) and the TCS for an approach perpendicular to the  $Z$  axis ( $\theta=90^\circ$ ), respectively. For instance, in the case of molecule  $\text{CCl}_4$ ,  $Q_{\parallel}(E)$  and  $Q_{\perp}(E)$  are both equal to  $Q_{\text{CCl}_3}$ . And  $Q_{\text{CCl}_3}$  is the TCS of electron scattering by  $\text{CCl}_3$  and is obtained still by Eq. (1).

Calculations have shown that the GAR model is valid at lower energies [5,10] and overestimates the shielding effect in molecules over the high-energy region, resulting in lower TCS results. Whereas the AR model is accurate at higher energies [2–4,11,12] and disregards the shielding effect at lower energies, resulting in higher TCS results compared with experimental findings, especially for relatively larger or complex polyatomic molecules. We know that a close-packed molecule is not fully transparent for low-energy electrons, but the transparency will improve as the energy increases. That is to say, when the incident electron energy is very low, the “inner” atoms are shielded partially by the “outer” atoms, thus the GAR model can give encouraging results, but the AR model not. When the energy is high enough, the molecule is fully transparent and each atom in the molecule can be freely scattered. Thus the interaction effect between atoms can be neglected at sufficiently high energies. Hence, the AR model is valid, but the GAR model not. Taking into account these factors, we incorporate the GAR and AR, and present a semi-empirical formula,

$$Q_{\text{TCS}}(E) = Q_{\text{GAR}}(E) + A[Q_{\text{AR}}(E) - Q_{\text{GAR}}(E)]. \quad (3)$$

Now, the key problem is how to choose the parameter  $A$ . The suitable expression of the parameter  $A$  should correctly reflect that the geometric shielding effect varies as the incident electron energy and the target molecular specifications, such as target molecular geometric dimension, total electron number and atom number in a target molecule. Choosing the parameter  $A$ , three factors should be considered. Firstly, when the incident electron energy is low enough, since the GAR model is valid, the  $A$  must satisfy  $Q_{\text{TCS}}(E) \approx Q_{\text{GAR}}(E)$ . And when the incident electron energy is high enough, because the AR model is accurate, the  $A$  must satisfy  $Q_{\text{TCS}}(E) \approx Q_{\text{AR}}(E)$ . This means that the empirical fraction  $A \rightarrow 0$  if  $E \rightarrow 0$ , and  $A \rightarrow 1$  if  $E \rightarrow \infty$ . That is to say, the higher the energy is, the less the effect on the TCSs should be. Secondly, at the same energy, the larger the size of a molecule is, the greater the

discrepancies between the TCSs calculated according to Eq. (1) and the experimental values are. This implies that the larger the volume of a molecule is, the smaller the empirical fraction  $A$  should be. Figs. 1–5 in this paper are a group of good examples. And the calculations also show that when molecules have the same total number of electrons, the more the total number of atoms in the molecule is, the smaller the empirical fraction  $A$  should be. And when molecules have the same total number of atoms, the more the total number of electrons in the molecule is, the smaller the empirical fraction  $A$  should be, too. And thirdly, calculations show that Eq. (1) cannot give a correct shape of TCS curve. We can clearly see this in log–log curves plotted by using the calculation results given in [3,4,11,12]. The main problem is that these log–log curves are steeper than the ones given by the corresponding experiments. Thus, the empirical fraction  $A$  relating to  $E$  and some molecular parameters should ensure a reasonable

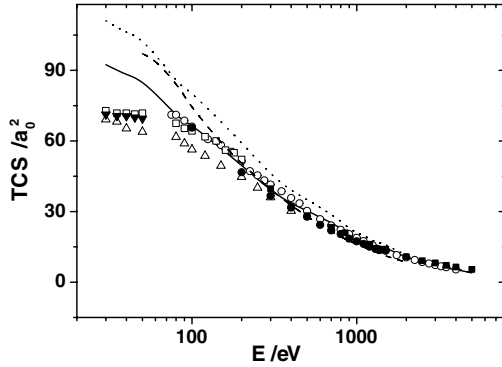


Fig. 1. TCSs for e-CF<sub>4</sub> scattering. Theoretical results: solid line, present modified AR calculations; dotted line, present unmodified AR calculations; dashed line, Jushipura et al. [9]. Experimental results: open circle, Zecca et al. [19]; full circle, Manero et al. [20]; full circle, Ariyasinghe [21]; open square, Szmytkowski et al. [22]; open triangle, Sueoka et al. [23]; full triangle, Jones [24].

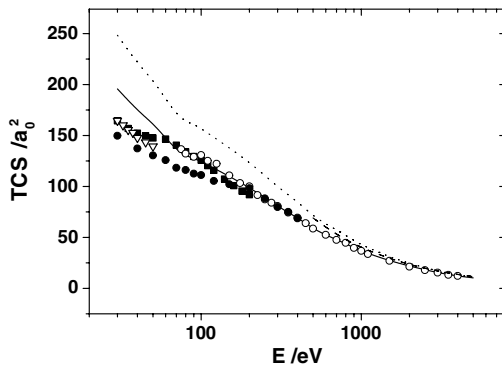


Fig. 2. TCSs for e-CCl<sub>4</sub> scattering. Theoretical results: solid line, present modified AR calculations; dotted line, present unmodified AR calculations; dashed line, García and Blanco [26]. Experimental results: open circle, Zecca et al. [19]; full square, Szmytkowski et al. [22]; open triangle, Jones [24]; full circle, Hamada and Sueoka [25].

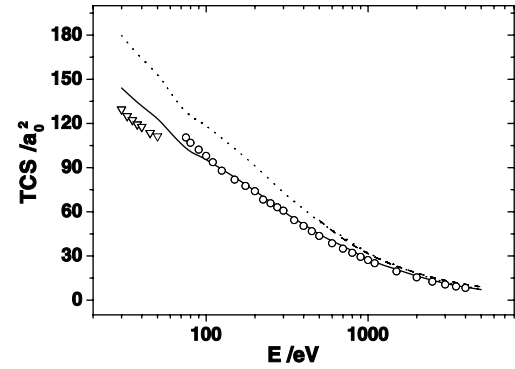


Fig. 3. TCSs for e-CF<sub>2</sub>Cl<sub>2</sub> scattering. Theoretical results: solid line, present modified AR calculations; dotted line, present unmodified AR calculations; dashed line, García and Blanco [26]. Experimental results: circle, Zecca et al. [19]; triangle, Jones [24].

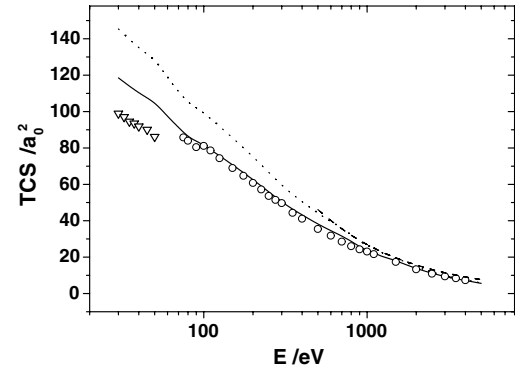


Fig. 4. TCSs for e-CClF<sub>3</sub> scattering. Theoretical results: solid line, present modified AR calculations; dotted line, present unmodified AR calculations; dashed line, García and Blanco [26]. Experimental results: circle, Zecca et al. [19]; triangle, Jones [24].

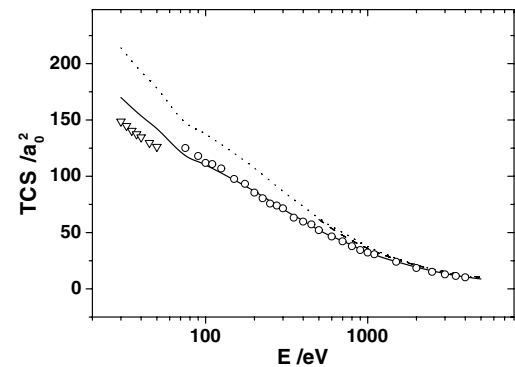


Fig. 5. TCSs for e-CFCl<sub>3</sub> scattering. Theoretical results: solid line, present modified AR calculations; dotted line, present unmodified AR calculations; dashed line, García and Blanco [26]. Experimental results: circle, Zecca et al. [19]; triangle, Jones [24].

shape for TCSs against the experimental data in the whole energy range. Taking into account these factors, we assume that

$$A = \frac{E}{mndC + E}. \quad (4)$$

Here,  $E$  in unit of eV is the incident electron energy.  $m$  and  $n$  are the numbers of electrons and atoms in the molecule, respectively.  $d$  in atomic unit is the target molecular dimension, which can be roughly determined by the bond length and bond angle given in [13]. And  $C$  is a constant considering the reasonable dimension relation, which is equal to  $1 \text{ eV}/a_0$ . According to those proposed by Bobeldijk et al. [5], molecules can be appropriately divided into two groups. One has cylindrical patterns, which can be appropriated by a rod-like shape. The other has circular patterns, which can be appropriated by a torus-like shape. Molecules such as  $\text{CF}_4$ ,  $\text{CCl}_4$ ,  $\text{CF}_2\text{Cl}_2$ ,  $\text{CClF}_3$  and  $\text{CFCl}_3$  have circular patterns and are approximated by a torus-like shape. Thus the parameter  $d$  in Eq. (4) is regarded as the diameter of the largest torus of the target molecule.

$q_T^j(E)$  in Eq. (1) can be obtained by the method of partial-waves

$$q_T^j(E) = q_e^j(E) + q_a^j(E) \\ = \frac{\pi}{k^2} \sum_{l=1}^{l_{\text{MAX}}} (2l+1) \times [|1 - S_l^j|^2 + (1 - |S_l^j|^2)], \quad (5)$$

where  $q_e^j$  and  $q_a^j$  are elastic and absorption cross sections, respectively.  $S_l^j$  is the  $l$ th complex scattering matrix element of the  $j$ th atom, which is related to the partial-wave phase shift as  $S_l^j = \exp(2i\delta_{lj})$ . To obtain  $S_l^j$ , we solve the following radial equation

$$\left( \frac{d^2}{dr^2} + k^2 - V_{\text{opt}} - \frac{l(l+1)}{r^2} \right) u_l(r) = 0, \quad (6)$$

under the boundary condition ( $r \rightarrow \infty$ )

$$u_l(kr) \sim kr[j_l(kr) - in_l(kr)] + S_l kr[j_l(kr) + in_l(kr)], \quad (7)$$

where,  $j_l$  and  $n_l$  are spherical Bessel and Neumann function, respectively. The limit  $l$  is taken large enough to generate the higher partial-wave contributions until again a convergence of less than 0.5% is achieved in the TCSs.  $V_{\text{opt}}$  is the spherical complex optical potential which incorporates all the important physical effects and is given by

$$V_{\text{opt}}(r) = V_s(r) + V_p(r) + V_e(r) + iV_a(r). \quad (8)$$

In detail, the static potential  $V_s(r)$  for electron–atom system is calculated by using the atomic charge density, determined from the well-known Hartree-Fock atomic wavefunction [14]. The exchange potential  $V_e(r)$  is provided by Riley and Truhlar [15]. Polarization potential  $V_p(r)$  is given by Zhang et al. [16]. And the absorption potential  $V_a(r)$  is presented by Staszewska et al. [17].

### 3. Results and discussion

Using the potential  $V_{\text{opt}}$ , we have calculated the TCSs for electron scattering by atoms He, Ne, Ar, Kr and Xe [18] at 30–5000 eV. Compared with the experimental data, the calculated TCS results are encouraging at the involved energies within the experimental errors. It shows that the electron–atom potential used in this paper appears to be

satisfactory, at least for the calculation of the TCSs on electron–atom scattering over the present energy range.

Employing the Eq. (3) together with the potential  $V_{\text{opt}}$ , we have calculated the TCSs for electron scattering by polyatomic molecules  $\text{CF}_4$ ,  $\text{CCl}_4$ ,  $\text{CF}_2\text{Cl}_2$ ,  $\text{CClF}_3$  and  $\text{CFCl}_3$  at 30–5000 eV. Our calculated results given by Eq. (3) (present modified AR calculations) and the results given by Eq. (1) (present unmodified AR calculations) together with the measurements [19–25] and some theories [9,26] are shown in Figs. 1–5, respectively.

Fig. 1 shows the present modified AR calculations, present unmodified AR calculations for electron scattering by  $\text{CF}_4$  together with the measurements obtained by Zecca et al. [19] at 75–4000 eV, Manero et al. [20] at 300–5000 eV, Ariyasinghe [21] at 100–1500 eV, Szmytkowski et al. [22] at 30–200 eV, Sueoka et al. [23] at 30–400 eV and Jones [24] at 30–50 eV and the theories obtained by Joshipura et al. [9] at 50–2000 eV. As a whole, the present modified AR calculations are slightly closer to the experimental findings [19–21] than the ones obtained by Karwasz et al. [8] and Joshipura et al. [27] at intermediate and low energies. However, for the sake of clarity, the theoretical results obtained by Karwasz et al. [8] and Joshipura et al. [27] are not included in Fig. 1. Obviously, the present modified AR calculations are in excellent agreement with the measurements obtained by Zecca et al. [19], Manero et al. [20] and Ariyasinghe [21] at the whole overlapping energies, and also in good accord with the measurements obtained by Szmytkowski et al. [22] above 80 eV and Sueoka et al. [23] above 200 eV. For example, the differences between the present modified AR calculations and the measurements [19] are only 0.3%, 1.0%, 0.2%, 0.2% and 0.4% at 80 eV, 100 eV, 500 eV, 1000 eV and 4000 eV, respectively; and the differences between the present modified AR calculations and the measurements [21] are about 3.9%, 5.7%, 12.2% and 9.4% at 100 eV, 300 eV, 800 eV and 1400 eV, respectively. Whereas the present unmodified AR calculations show great deviations when they are compared with the measurements [19–24] at intermediate and low energies. Even compared with Jones [24] at 30–50 eV, Szmytkowski et al. [22] below 80 eV and Sueoka et al. [23] below 200 eV, one can instantly see that the present modified AR calculations are much closer to them than the present unmodified ones. Thus, for the proposed formula, good agreement with measurements [19–23] is obtained when incident electron energy is above 100 eV or so.

As far as the TCSs of electron scattering by  $\text{CCl}_4$  are concerned, we have found four groups of measurements obtained by Zecca et al. [19] at 75–4000 eV, Szmytkowski et al. [22] at 0.5–200 eV, Jones [24] at 0.6–50 eV, Hamada and Sueoka [25] at 0.7–400 eV, and three groups of theories obtained by Karwasz et al. [8] at 50–3000 eV, García and Blanco [26] at 500–10000 eV and Joshipura et al. [27] at 60–2000 eV in the literatures to the best of our knowledge. In order to avoid congestion, only present modified and unmodified AR calculations together with these measurements above 30 eV and one group of theories obtained



by García and Blanco [26] at 500–5000 eV are shown in Fig. 2. Similarly to electron scattering by  $\text{CF}_4$ , the present modified AR calculations are in slightly better agreement with the measurements [19] than the theories obtained by Karwasz et al. [8] and Joshipura et al. [27] at intermediate and low energies. When comparing the present modified AR calculations with the above-mentioned measurements [19,22,24,25], we have found that there is in good agreement between the present modified ones and the measurements obtained by Zecca et al. [19] at the whole overlapping energies, Szmytkowski et al. [22] above 60 eV and Hamada and Sueoka [25] above 100 eV, but in poor agreement with the measurements obtained by Jones [24] at 30–50 eV, Szmytkowski et al. [22] below 60 eV and Hamada and Sueoka [25] below 100 eV. From these comparisons, we find that good agreement can be obtained between the present modified AR calculations and the available measurements above 100 eV or so.

For the TCSs of electron scattering by  $\text{CF}_2\text{Cl}_2$ ,  $\text{CClF}_3$  and  $\text{CFCl}_3$ , we have only found two groups of measurements obtained by Zecca et al. [19] at 75–4000 eV and Jones [24] at 0.6–50 eV, and two groups of theories given by García and Blanco [26] at 500–10000 eV and Karwasz et al. [8] at 50–3000 eV in the involving energy range. We depict the modified and unmodified AR results, two groups of the experiments [19,24], the theories given by García and Blanco [26] in Figs. 3–5, respectively. In theories, the present modified AR calculations are closer to the experimental findings [19] than the theories obtained by Karwasz et al. [8] (not shown in Figs. 3–5). Take  $\text{CF}_2\text{Cl}_2$  as an example. The  $\text{CF}_2\text{Cl}_2$  calculated values [8] differ from the measurements [19] about by a constant (5%) value at all energies above 500 eV, whereas the present modified AR calculations deviate from the measurements [19] only by 5.9% at 80 eV, 2.3% at 100 eV, 1.8% at 300 eV and 2.2% at 1000 eV, respectively. In experiments, the present modified AR calculations are in excellent accord with the above-mentioned measurements at the whole overlapping energies except those obtained by Jones [24] at 30–50 eV, and the higher the energy is, the smaller the discrepancies between the present modified AR calculations and the measurements [19] are. Obviously, the proposed formula can give encouraging results above 75 eV, as seen in Figs. 3–5.

From Figs. 1–5, we clearly see that the present modified AR model is more effective than the unmodified one over the present energy range. The modified AR calculations are in fairly good agreement with the available measurements for  $\text{CF}_4$  and  $\text{CCl}_4$  above 100 eV and for  $\text{CF}_2\text{Cl}_2$ ,  $\text{CClF}_3$  and  $\text{CFCl}_3$  above 75 eV. However, the present unmodified AR results substantially exceed the experimental findings at intermediate and low energies, especially below 500 eV or so. Comparing the present two theoretical calculations, we find the contributions of the shielding effect to the TCSs are larger in the low and intermediate energy range, particularly for polyatomic molecules which contain higher atomic weight atoms.

At lower energies, as seen in Figs. 1–5, the present modified AR calculations are obviously higher than all the measurements because we do not consider the interference effect originating from each constituent group in a molecule [9]. This effect may be important below 100 eV or so as the de Broglie wavelength (0.123 nm) associated with the incident particle will become comparable to the bond length of the molecule (e.g. the C–Cl bond length in  $\text{CCl}_4$  and the C–F bond length in  $\text{CF}_4$  are of 0.1767 and 0.1323 nm [13], respectively). Thus it might lead to an overestimation of the calculated TCSs [9]. In addition, the multicenter scattering and valence-bond distortion [28,29] are also neglected in the present investigations. It will increase the possibility of multiple scattering when the de Broglie wavelength of the incident electrons is comparable to the internuclear distance of the molecule at energies of several tens of eV. It is expected that the inclusion of multiple scattering effect at lower energies will reduce the TCSs by a significant amount [28]. The likely effect of the valence-bond distortion is also to reduce the cross sections [29]. So, though we have taken into consideration the geometric shielding effect in the molecule, TCS differences still exist between the present modified AR calculations and the experimental findings at lower energies.

With the energy increasing, the wavelength of the incident electrons becomes smaller and smaller compared with the bond length of the molecule, and the interactions among the atoms in the molecule become weaker and weaker, then the above-mentioned effects contribute less and less. Therefore, the higher the energy is, the better the modified AR calculations are. So, the modified AR model can attain excellent calculations, as verified in Figs. 1–5.

Finally, we briefly state the TCS results of electron scattering by  $\text{CH}_4$ ,  $\text{C}_2\text{H}_2$ ,  $\text{C}_2\text{H}_4$ ,  $\text{C}_2\text{H}_6$  and  $\text{C}_2\text{H}_3\text{F}_3$ . From the comparison between present modified AR calculations and the measurements [30–41], as a whole, we find present modified AR calculations are in good accord with the available measurements above 100 eV. Similarly to the molecules shown in Figs. 1–5 in this paper, the higher the energy is, the smaller the discrepancies between present modified AR calculations and the measurements are. Whereas the present unmodified AR calculations show great discrepancies, especially at intermediate and low energies. The detailed calculation results and the comparison with the available measurements will be reported in the near future.

#### 4. Conclusions

The original AR model disregards molecular geometric properties, thus the molecular scattering problem is reduced to the atomic scattering one that is easier to handle. And since the original AR model ignores the geometric shielding effect, the multicenter scattering effect [28], the valence-bond distortion effect [29] and the interference effect originating from each constituent group [9], obvious deviations from the measurements can be seen at intermediate and low energies [11,12]. In this paper, we think that

the geometric shielding effect in a molecule, which leads to smaller TCSs than predicted by the original AR model, is relevant to the target molecular dimension, particularly for complex polyatomic molecules or relatively larger molecules at low energies. And by taking into consideration the contributions of geometric shielding effect in a molecule as the incident electron energy varies, the modified AR, which combines successfully the properties of the GAR and AR approach, is presented. The modified AR approach is simple but more effective for electron scattering by complex molecules  $\text{CF}_4$ ,  $\text{CCl}_4$ ,  $\text{CF}_2\text{Cl}_2$ ,  $\text{CClF}_3$  and  $\text{CFCl}_3$ , which can be approximated by a torus-like shape [5], than the original AR at 30–5000 eV. Great success has been made at lower energies, especially above 100 eV, though we ignore the multiple scattering effect, the valence-bond distortion effect and the interference effect between chemical groups in the present investigations. As further investigations, we will take into consideration these effects so that we can extend the validity of the AR model to energy of several tens of eV for even more complex molecules.

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