

Differential and integral cross-sections of e-O₂, O₃, NO, CO scattering at energies 100–1000 eV

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Abstract. A modified additivity rule is formulated to calculate the differential cross-sections for elastic scattering of electrons from molecules. It improves the results at small angles and at relatively lower incident energies (<1000 eV). Integral cross-sections calculated presently are combined with the known total ionization cross-sections to obtain total (complete) cross-sections. An extension of the present approximation to larger molecules is also suggested.

Keywords. Electron molecule scattering; differential and total cross-section; polarization potential.

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1. Introduction

It is a common knowledge now that the cross-sections of various processes of electron scattering on molecules are required in many applications. Although total cross-sections are more useful from this point of view, the differential cross-sections are also important for comparing the theory with experiments. The present paper examines the elastic scattering of intermediate to high energy electrons by four molecular targets, viz., oxygen, ozone, nitric oxide and carbon monoxide. At impact energies (E_i) above 100 eV or so, the collisions of electrons with atoms and molecules generally result in *elastic* scattering accompanied mainly by *ionization* [1,2]. Almost above 500 eV the independent atom model (IAM) [3,4] becomes valid for lighter molecules, so that the electron–molecule collision can be viewed in terms of an appropriate electron–atom problem. However, the IAM and the consequent additivity rule (AR) for total cross-section become meaningful at high enough energies. Therefore, typically in the range of 100–1000 eV, it is more appropriate to consider the modified additivity rule (MAR) formulated by us [5]. Now, the aim of the present paper is to formulate the MAR for *differential* cross-section (DCS) of electron scattering by simple molecules. The calculated DCS can be integrated over the scattering angles (θ, ϕ) to determine the integral elastic cross-section $\sigma_{el}(E_i)$. To this cross-section we can add the known total *ionization* cross-section $Q_{ion}(E_i)$, to obtain [6] the total (*complete*) collision cross-section $Q_T(E_i)$.

2. Theoretical highlights

At high enough energies where the IAM is valid, one can write [3,5] the electron–molecule scattering amplitude f_M as a superposition over the scattering amplitudes $f_i(\theta, k)$ of the n constituent atoms, with $i = 1, \dots, n$. The average molecular DCS is expressed as

$$\bar{I}(\theta; k; R) = |f_M(\theta; k; \vec{R})|^2. \quad (1)$$

For molecular oxygen the DCS depends parametrically on the bond length $R = 2.29a_0$, while for the triatomic target O_3 , it involves the shorter bond lengths $R_{12} = R_{23} = 2.42a_0$ and the longer bond length $R_{23} = 4.17a_0$. In this approximation the scattering amplitudes $f_i(\theta; k)$ are calculated from the *total* atomic potential consisting of the static, the exchange and the polarization potentials. The approximate DCS derived from the simple AR formulation as above are known to overestimate at energies that are not very high. Typically, at energies below about 500 eV, the small angle AR DCS are found to be larger than the experimental data and accurate theories by about 20% [4,5]. To correct for this behaviour, at least partially, we must incorporate the long range polarization potential at the direct, i.e., molecular level. An improved approach, based on this idea was developed [5] to obtain *total* cross-sections of e-molecule scattering. In order to extend the same to calculate the DCS, we represent the total spherical e-molecule interaction as,

$$V = V_{SR} + V_{LR}, \quad (2)$$

where V_{SR} is the short range part and V_{LR} is the long range part of the total potential. Our molecular targets are either weakly polar or non-polar, viz., O_2 . Hence the only long range potential is the polarization potential here. Equation (2) allows us to express the e-molecule scattering amplitude f_M as a sum of the short range contribution f_{SR} and the long range contribution f_{LR} , i.e. [5],

$$f_M(\theta; k) = f_{SR} + f_{LR}. \quad (3)$$

The amplitude f_{LR} is calculated at the molecular level, by considering the polarization potential [2,5],

$$V_{pol}(r; k) = -\frac{\alpha_0 r^2}{2(r^2 + r_c^2)^3}, \quad (4)$$

where r is the distance of the external electron from the molecular centre of mass and α_0 is the static dipole polarizability of the molecule. Further, r_c is an energy dependent cut-off parameter. We have determined the cut-off parameter by employing the first ionization energy I instead of the average excitation energy Δ , since I is very accurately known for a number of targets including the present ones. The polarization potential as employed through eq. (4) is the only long range potential here, since it falls off like $1/r^4$ at large distances $r \gg r_c$. This potential influences in particular the small angle scattering. In view of eq. (4) the long range contribution f_{LR} in eq. (3) is now denoted as f_{pol} .

Next, to calculate f_{SR} we retain the IAM approximation, and obtain the short range atomic amplitudes corresponding to the *atomic* (short range) potential,

$$V_A = V_{st} + V_{ex}. \quad (5)$$

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The amplitudes f_{SR} and f_{pol} are derived through the partial wave analysis of V_A and V_{pol} respectively. Finally the MAR DCS are obtained from eqs (1) and (3).

The total elastic cross-sections σ_{el} can be calculated by integrating the molecular DCS over all angles, at various energies. It has been demonstrated in [6] that when the incident energy is high, the electron–molecule total (*complete*) cross-section Q_T is given reasonably well by adding total ionization cross-section Q_{ion} to the elastic contribution σ_{el} . Thus,

$$Q_T(E_i) = \sigma_{el}(E_i) + Q_{ion}(E_i). \quad (6)$$

3. Results, discussions and conclusions

The difference between the AR and the MAR methods depends on the polarization effect. Hence the ozone molecule is suitable for the present study in view of its high polarizability, $\alpha_0 = 21.7a_0^3$. Figure 1 shows our calculated DCS for e-O₃ scattering at 100 eV. Our results in these two methods agree in the general shape of the curves, but differ in magnitude. The AR DCS are higher than the MAR DCS, and the difference is more at the small and the intermediate angles. Since no experimental differential cross-sections are available for O₃ at any energy, the present results are compared mutually. At 100 eV, the reduction in the forward DCS observed with the MAR is about 30% of the AR value. The MAR results are expected to be reliable. The theoretical DCS at 100 eV show a dip around 100°, for all the targets examined presently. Figure 2 exhibits our calculated results on O₂ and O₃ targets at 300 eV. Comparisons are also made with the available experimental data on O₂. The calculated MAR DCS on O₂ are in good agreement with the measurements of Daimon *et al* [7] as well as Iga *et al* [4], except for large angles. Our MAR method of calculation treats the polarization potential at the direct molecular level, and this long range potential

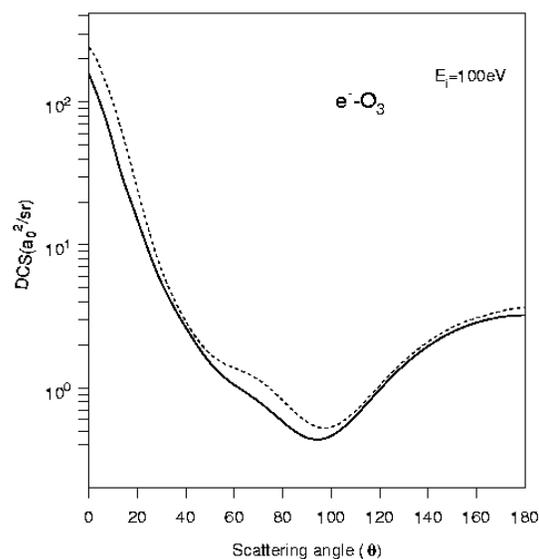


Figure 1. Present DCS for e-O₃ scattering at 100 eV. - - - AR; _____ MAR.

affects the DCS at small angles. We find, therefore, a significant decrease in the cross-sections of ozone in the MAR, as compared to the AR. The difference between the present two methods tends to vanish beyond $\theta = 20^\circ$, at 300 eV. Hence we have shown only the MAR results, which are closer to the measured data at small angles of scattering. The O_2 DCS are in general lower than those of O_3 , as expected. Apart from this, one finds here the diffraction-like maxima in the DCS of triatomic target O_3 as a function of angles. These oscillations are less pronounced in the case of the diatomic O_2 molecule. On account of this, the DCS of the two targets are almost the same at about 10° . Let us turn now to figure 3, which corresponds to 500 eV. Here, the lower curve for the MAR DCS on O_2 are seen

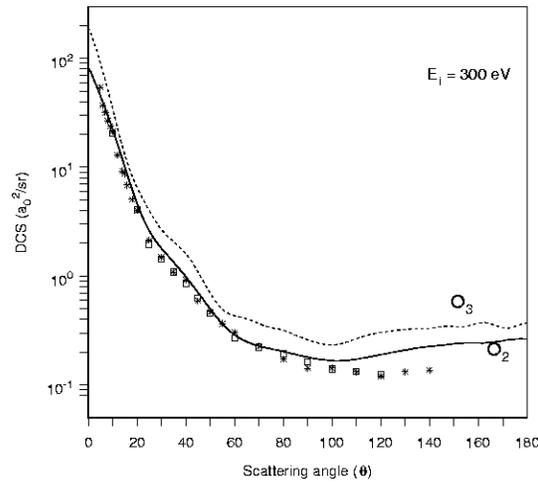


Figure 2. Present DCS for e- O_3 scattering at 300 eV. Experimental data on O_2 also is shown. *** Diamon *et al* [7]; □□ Iga *et al* [4].

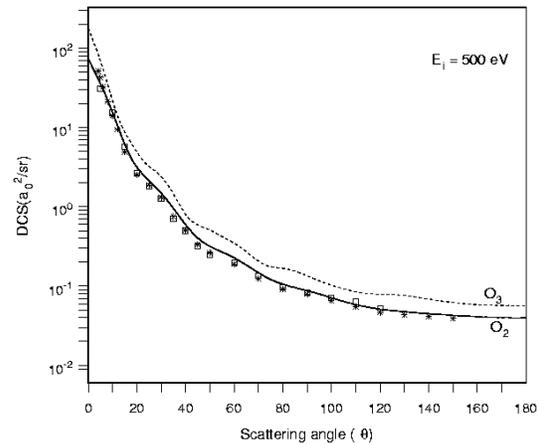


Figure 3. Present DCS for e- O_3 scattering at 500 eV. Experimental data on O_2 also is shown. *** Diamon *et al* [7]; □□ Iga *et al* [4].

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to be in a very good accord with the experimental data [4,7]. The oscillations in the angular distribution are found to increase with energies, as also observed by Pablos *et al* [6]. At about 1000 eV and beyond, the polarization effect becomes insignificant, and the MAR merges with AR.

Consider now the NO and CO molecules, having polarizabilities 11.48 and 13.17 in a_0^3 respectively. Figures 4 and 5 exhibit the e-NO scattering at sample energies of 100 and 500 eV. Experimental data are not available for comparisons at these energies. Despite the importance of NO in atmospheric studies due to its role in the catalytic destruction of ozone [8], there are relatively few e-NO scattering measurements. Elastic DCS for e-NO system are calculated by Lee *et al* [9] in 5–500 eV range using the Born-closure Schwinger-

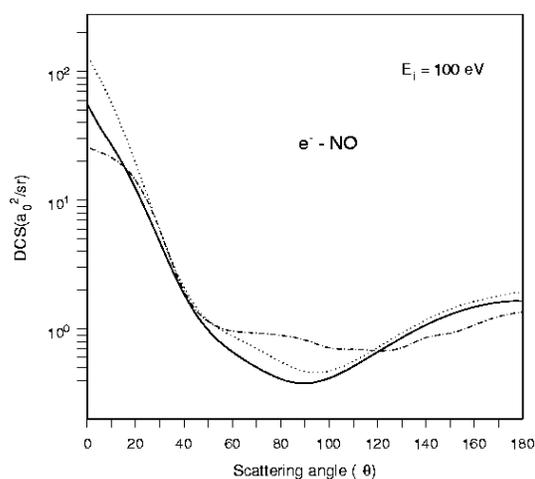


Figure 4. Theoretical DCS for e-NO scattering at 300 eV. - - - - AR; _____ MAR; - · - · - Lee *et al* [9].

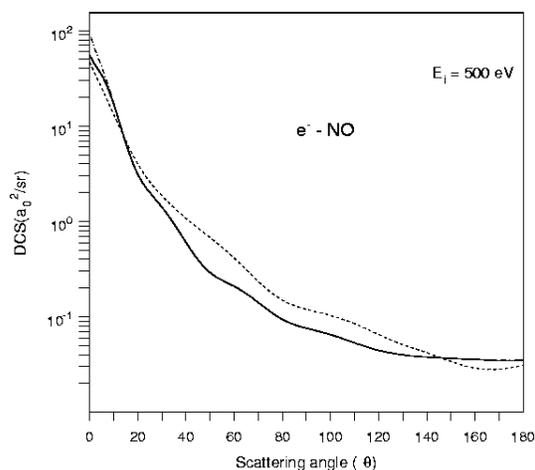


Figure 5. Theoretical DCS for e-NO scattering at 500 eV. - - - - AR; _____ MAR; - · - · - Lee *et al* [9].

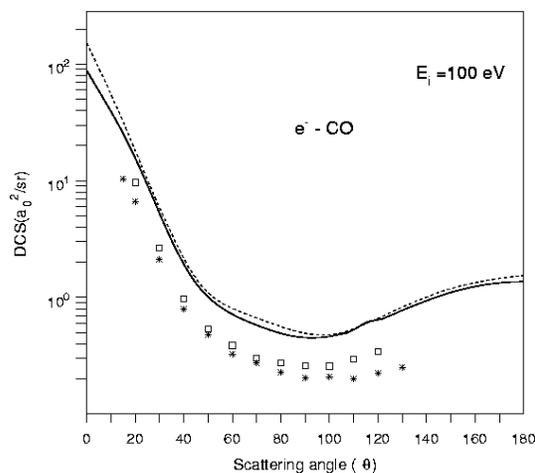


Figure 6. Present DCS for e-CO scattering at 100 eV. Experimental data: *** Tanaka *et al* [10], □□□ Nickel *et al* [11].

variational method. We have compared our MAR results with the calculations of [9] at 100 and 500 eV. In the small angle region up to about 20° the MAR DCS are substantially lower than the AR values. Our MAR approximation and the calculations of Lee *et al* [9] differ from each other. The DCS of [9] are on the lower side at small angles, because they [9] have neglected the polarization effects. At 500 eV (figure 5), the difference between the present and the previous data [9] still exists. The oscillations in the angular distribution are clearly visible at this energy. Finally in figure 6 our theoretical results for e-CO scattering are compared with the experimental work of Tanaka *et al* [10] and Nickel *et al* [11] at 100 eV. We notice that the present curve overestimates the experimental data, but reproduces the shape. The overestimation at a relatively lower energy 100 eV can be understood in terms of the nature of the present approximations.

The total (integrated) elastic cross-sections σ_{el} at various energies are determined by integrating the MAR DCS. The total (*complete*) cross-sections (TCS) Q_T are found using eq. (6). In tables 1 and 2 we have shown these total cross-sections for the present targets. In general the total cross-sections are less affected by the present modification as against the DCS, which are appreciably reduced in the forward directions at lower energies. For example, the MAR formulation applied to e-O₂ collisions at 100 eV reduces the TCS by about 8%, in comparison with the AR result. Now as table 1 shows, the present (i.e. MAR) TCS Q_T of molecular oxygen are in good accord with the two sets of measurements, viz., those of Dalba *et al* [12] and Dababneh *et al* [13]. At the lower end of the present energy range our calculated values are somewhat higher than the compared data. In the case of ozone too we find a good agreement (table 1) between the present calculations and the measured Q_T data determined in the joint Spain–UK experiment [6]. Actually in [6] the experimental results have been compared with our earlier MAR values. No other comparisons are available for O₃. Throughout the present work the small contribution of the dipole potential (of polar targets) has been ignored. Let us make a brief remark here in this regard. We find in the case of ozone (dipole moment 0.21 a.u.) that at 100 eV, the total cross-section Q_T with and without dipole contribution is 13.76 Å² and 13.91 Å²

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Table 1. Integrated elastic cross-sections Q_{el} and total (complete) cross-sections Q_T both in 10^{-16} cm², for O₂ and O₃ molecules.

Energy E_i (eV)	e-O ₂				e-O ₃		
	Present results		Experimental Q_T		Present results		Experimental Q_T
	σ_{el}	Q_T^*	(a)	(b)	σ_{el}	Q_T^*	(c)
100	6.21	9.11	8.59	8.00	9.62	13.72	–
300	2.88	5.21	4.86	4.90	4.46	7.96	7.75
500	2.02	3.73	3.68	3.50	3.13	5.69	5.69
700	1.53	2.88	2.89	–	2.37	4.49	4.44
1000	1.18	2.23	2.08	–	1.95	3.50	3.39

*Including Q_{ion} of refs [18,19].

(a) Dalba *et al* [12]; (b) Dababneh *et al* [13]; (c) Pablos *et al* [6].

Table 2. Total cross-sections as in table 1, but for NO and CO molecules.

Energy E_i (eV)	e-NO			e-CO		
	Present results		Experimental Q_T	Present results		Experimental Q_T
	σ_{el}	Q_T^*	(a)	σ_{el}	Q_T^*	(b)
100	6.16	9.29	8.46	7.39	10.03	9.01
300	2.99	5.43	5.18	3.21	5.19	4.85
500	2.09	3.94	3.60	2.19	3.68	3.56
700	1.57	3.08	2.77	1.62	2.83	2.80 ^(c)
1000	1.21	2.41	2.11	1.02	1.97	2.20 ^(c)

*Including Q_{ion} of ref. [19].

(a) Dalba *et al* [12]; (b) Kwan *et al* [15]; (c) Garcia *et al* [16].

respectively. Thus the rotational excitation cross-section of O₃ calculated in the first Born approximation [14] is about 2% of the Q_T , and is insignificant at higher energies. Table 2 exhibits our Q_T results for NO and CO in the present energy range. The TCS of the NO molecule are nicely comparable to the experimental data of Dalba *et al* [12], except at about 100 eV. At 100 eV our MAR calculations of Q_T for all the present targets are seen to overestimate the experimental data by about 10%. The underlying theoretical model [3] breaks down at lower energies. The present TCS of e-CO scattering are compared in table 2, with the experiments of Kwan *et al* [15] up to 500 eV, and of Garcia *et al* [16] at higher energies. Here again the agreement between the theory and experiment is quite satisfactory.

Thus in conclusion, the present work highlights our MAR method for calculating e-molecule differential cross-sections at energies where the high energy IAM is still not quite valid. The total (complete) cross-sections Q_T obtained presently are also satisfactory, although the effect of target polarization on the TCS is rather small. It is interesting to note that the MAR approximation can be extended to the well-known polyatomic molecules.

Thus, we can consider (chemical) *groups* of bonded atoms rather than individual atoms in a molecule, and treat them as approximately independent scattering centers in the range of high incident energies. This, the so-called 'group additivity' approach [17], would be meaningful since the polarizabilities of many important molecules are available in literature.

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