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Additivity rule for electron-molecule cross section calculation: A geometrical approach

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Abstract

A new formulation of the additivity rule is proposed. Here atomic cross sections are added by taking into account the geometrical screening of the component atoms, as seen by the impinging electron. Atomic cross sections are obtained starting from the experimental TCS of simple molecules (H_2 , N_2 , O_2 , CO) via inversion procedure. The model has been successfully applied to the linear molecules NO, N_2O , CO₂, for energies as low as 50 eV up to 5 keV. A simplified algorithm has been used to evaluate the cross sections of NO₂ (a bent molecule) and of CH₄: the results underestimate the low energy cross sections. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Total Cross Section (TCS) for electron scattering by atoms and molecules plays an important role in many applied sciences [1]. There is, however, a lack of theoretical calculations in the high and intermediate energy range at which almost all molecular inelastic channels are open. Therefore, simpler, phenomenological procedures have been recently developed.

One such methodology develops from the consideration that, at high impact energies, the TCS of a molecule approaches the sum of the atomic TCS of its constituents [2]. Recently, the "additivity rule" (AR) prescription has been used to evaluate both high-energy molecular elastic [3] and total (i.e. elastic + inelastic) cross sections [4–7]. In this range (say from a hundred eV to a few thousand eV) those works were moderately successful, whereas failed completely at lower energies. This is obviously due to the fact that AR neglects the interaction of constituent atoms inside the formed molecule. All these recent works used ab- initio calculations in the optical approximation (see Jain and Baluja [8]) to derive atomic cross sections and summed these contributions to yield molecular TCS. The use of AR at high energies is well established for elastic cross sections in the framework of an independent-atoms model (IAM) [9,10]. This model includes intrinsically the molecular geometry via interference scattering terms. However, the application of the IAM to *inelastic* scattering is not straightforward [11].

In the most recent papers [12] some progress has been made in extending AR validity to low energies (50–100 eV) by including molecular polarization potential. A way to include effects of molecular geometry in TCS was proposed by Jiang et al. [13,14] for linear targets. In the low energy limit they used geometry-averaged cross sections determined as $\sigma_M(E) = 1/3\sigma_{\parallel}(E) + 2/3\sigma_{\perp}(E)$, where $\sigma_{\parallel}(E)$ and $\sigma_{\perp}(E)$ are the cross sections for electrons approaching the molecule parallel and perpendicular to its axis, respectively.

In a recent paper [15] a different, semiempirical approach has been proposed. Experimental TCS in the 100–4000 eV range have been approximated by a two parameters formula

$$\sigma(E) = \frac{\sigma_z b}{b + \sigma_z E} \tag{1}$$

with σ_z defining the cross section in the zero-energy limit, and *b* giving the asymptotic slope of TCS in the high energy (several keV) limit. We have shown that the parameter σ_z is related to molecular polarisability while the molecular *b* parameter turn out to be the sum of the corresponding atomic parameters. The model has been successfully applied to molecules of compact geometry (tetrahedral and octahedral).

In this paper we present a different formulation of the additivity rule which takes into account the geometry of the molecule. In particular, we study the case of linear (NO, N₂O, CO₂) and bent triatomic (NO₂) molecules, and spherical molecules (CH₄). Explicit geometrical calculations of the molecular cross sections through spatial averaging of the different orientations under which the molecule is seen by the impinging electrons in a real experiment is performed. This leads to express the molecular TCS as a linear combination of the atomic TCS, that is:

$$\sigma_M = \sum_{\alpha} k_{\alpha} \, \sigma_{\alpha} \tag{2}$$

where the energy-dependent k-coefficients are linked to the geometric parameters of the molecule. Those coefficients approach unity at high energies, so in this range the present method merges with the simple AR prescription.

Like in our previous paper [15], an inverse additivity procedure has been applied to extract atomic TCS from experimental molecular TCS. In turn, these data are used to predict cross sections for more complex molecules. The predicted cross sections are then compared with measured ones.

2. Model

At energies above 50 eV different inelastic channels are open including multiple ionisation and electronic excitations. Therefore, we will assume that σ_{α} can be described as a "black sphere" (i.e. perfectly absorbing). The atomic TCS σ_{α} is given then by a known quantity [16]

$$\sigma_a = 2\pi r_a^2 \tag{3}$$

where r_a is the black sphere radius.

In collision experiments where the target is in a gas phase, molecules are randomly oriented and the observed cross sections are averaged over the different relative beam-to-molecule orientations. In order to compare the calculated cross section with the experimental cross sections need to take the average value of σ_M over all the possible orientations of the molecules. Considering collision energy well above the roto- vibrational levels of the target, we will perform the average procedure in the fixed-nuclei approximation: the molecule will be seen as a rigid body. We note that in our model the atom-atom interaction within the molecule is accounted for implicitly: the ground state average values of the interactions.

The following part is dedicated to find analytically the average TCS over different orientations for a given diatomic heteronuclear molecule.

A diatomic molecule will be represented by two associated spheres with radii

$$r_{\alpha} = \sqrt{\sigma_{\alpha}/2\pi}, \quad \alpha = 1,2$$
 (4)

and a centre-to-centre distance which equals the molecular bound length d. The projection of d on the xy plane, perpendicular to the direction z of projectile electrons, is indicated by l.

We first evaluate the projected area on the *xy* plane of the associated spheres for a given angle θ of the molecular axis with the *z* axis. The reference system and the projection of the spheres in the three cases described below are schematically reported in Fig. 1(a) and (b)–(d), respectively. We have thus $l = d\sin \theta$. The projected area A_{12} reads as a linear combination

$$A_{12} = C_{1j}A_1 + C_{2j}A_2$$

where $A_1 = \pi r_1^2$ and $A_2 = \pi r_2^2$.

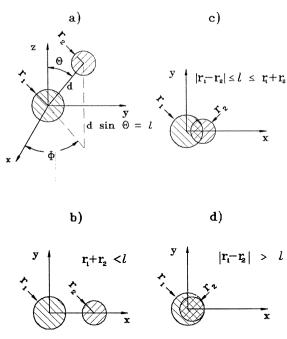


Fig. 1. Reference system for the calculation of the diatomic molecule projection. Schematic representation of three cases described by Eqs. (5)-(7).

The coefficient $C_1 j$, C_{2j} are evaluated in the following three cases.

Ist case (Fig. 1(b), the projection of the two spheres do not overlap:

$$C_{11} = C_{21} = 1 \quad \text{for } r_1 + r_2 < l \tag{5}$$

Ind case (Fig. 1(c), the projection of the two spheres partially overlap:

$$C_{12} = \left(1 - \frac{1}{\pi} \arccos b_1 + \frac{b_1}{\pi} \sqrt{1 - b_1^2}\right)$$

$$C_{22} = \left(1 - \frac{1}{\pi} \arccos b_2 + \frac{b_2}{\pi} \sqrt{1 - b_2^2}\right)$$
for $|r_1 - r_2| \le l \le r_1 + r_2$ (6)

where

$$b_1 = \frac{x}{r_1} = \frac{r_1^2 - r_2^2 + l^2}{2r_1 l}, \quad b_2 = \frac{x}{r_2} = \frac{r_2^2 - r_1^2 + l^2}{2r_2 l}$$
(7)

 b_1 and b_2 are respectively the distances of the centres of the two projected spheres (Fig. 1(c) from

the co-ordinate x of the intersection point of the two projections.

IIIrd case (Fig. 1(d), the projection of the two spheres completely overlap:

$$C_{13} = h(r_1 - r_2), \quad C_{23} = h(r_2 - r_1)$$

for $l < |r_1 - r_2|$ (8)

Where h(x) is a Heaviside function.

We gather the expression referred to the different cases into a unique formula:

$$A_{12}(\theta) = C_{1j}(\theta; r_1, r_2, d) A_1 + C_{2j}(\theta; r_1, r_2, d) A_2$$
(9)

where C_{1j} and C_{2j} are the coefficients of A_1 and A_2 in the three different cases of Eqs. (5), (6) and (8), and *j* equals 1, 2, 3 according to specific cases of overlap, see below Eqs. (15)–(17).

The average of A_{12} over all the possible θ reads as:

$$\langle A_{12}(\theta) \rangle = \langle C_{1j}(\theta; r_1, r_2, d) \rangle A_1$$

+ $\langle C_{2j}(\theta; r_1, r_2, d) \rangle A_2$
= $k_1(r_1, r_2, d) A_1 + k_2(r_1, r_2, d) A_2$
(10)

or, in terms of molecular and atomic cross sections:

$$\sigma_{M} = 2\langle A_{12}(\theta) \rangle = k_{1}\sigma_{1} + k_{2}\sigma_{2}$$
(11)

Making the average over the solid angle, we have thus to solve the integral

$$k_{1,2}(r_1, r_2, d) = \frac{1}{4\pi} \int d\Phi \int C_{1j,2j}(\theta; r_1, r_2, d) \sin\theta d\theta$$
(12)

The geometrical representation is clearly symmetric under rotation about the *z* axis and about the molecular dipole direction. Also, the coefficients $k_{1,2}$ are written with a dependence on three parameters, but they appear only through the ratios r_1/d and r_2/d , because the coefficients cannot be affected by an overall scaling of lengths. Note additionally, that as the atomic cross sections σ_{α} depend on energy, also the coefficients $k_{1,2}$ are energy dependent. We now give the solution for $k_{1,2}(r_1, r_2, d)$, rewriting the Eq. (7) for b_1 and b_2 as a function of the variable $\xi = \cos \theta$

$$F(\xi) = b_1(\xi) = \frac{r_1^2 - r_2^2}{2dr_1\sqrt{1 - \xi^2}} + \frac{d}{2r_1}\sqrt{1 - \xi^2}$$
(13)

We also define the following constants:

$$a_{1,2} = \sqrt{1 - \left(\frac{r_1 \pm r_2}{d}\right)^2}$$
(14)

We have to distinguish three cases:

1. the two spheres do not overlap each other, $r_1 + r_2 < d$,

$$k_{1} = \int_{0}^{a_{1}} C_{11}(\theta; r_{1}, r_{2}, d) dx + \int_{a_{1}}^{a_{2}} C_{12}(\theta; r_{1}, r_{2}, d) dx$$
$$+ \int_{a_{2}}^{1} C_{13}(\theta; r_{1}, r_{2}, d) dx$$
$$= 1 - (1 - a_{2}) h(r_{1} - r_{2}) - \frac{1}{\pi} \int_{a_{1}}^{a_{2}} (\arccos F(\xi))$$
$$- F(\xi) \sqrt{1 - F^{2}(\xi)} dx$$
(15)

2. the two spheres are partially overlapping: $r_1 + r_2 > d$, $|r_1 - r_2| < d$.

$$k_{1} = \int_{0}^{a_{2}} C_{12}(\theta; r_{1}, r_{2}, d) dx + \int_{a_{2}}^{1} C_{13}(\theta; r_{1}, r_{2}, d) dx$$
$$= a_{2} - (1 - a_{2})h(r_{1} - r_{2}) - \frac{1}{\pi} \int_{0}^{a_{2}} (\arccos F(\xi)) - F(\xi) \sqrt{1 - F^{2}(\xi)} dx$$
(16)

3. one sphere is included in the other: $|r_1 - r_2| > d$

$$k_1 = \int_0^1 C_{13}(\theta; r_1, r_2, d) \, dx = h(r_1 - r_2) \tag{17}$$

In all three cases $k_2(r_2, r_1, d) = k_1(r_1, r_2, d)$.

In Fig. 2 calculated values of the $k_{1,2}$ coefficients are plotted for chosen values of non-dimensional variables r_1/d and r_2/d .

The above formalism considers the problem of estimating a molecular TCS by using the cross sections - at a given energy - of its atomic constituents. However, in general TCS of molecules are

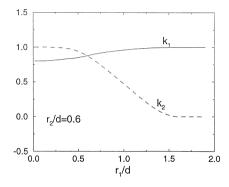


Fig. 2. Variation of the geometrical coefficients k_1 and k_2 with dimensionless radii of atoms. *d* stands for the molecular bond length.

better known than the TCS of atoms. Therefore, it is of interest to study an inversion algorithm, in order to extract atomic cross sections from TCS values of their compounds.

We first consider the simplest case of a homonuclear molecule. The radii of the associated spheres are $r_1 = r_2 = r$, and Eq. (11) reads as:

$$\sigma_M = 4\pi r^2 k_1(r,d) \tag{18}$$

In the present case, σ_M and *d* can be considered as known quantities, while *r* is unknown. If we define the auxiliary function

$$G_1(r) = \sigma_M - r\pi r^2 k_1(r) \tag{19}$$

the searched values of r are given by $G_1(r) = 0$. If G(r) is also a monotonically decreasing function of r, the inversion will be single-valued.

The case $r_1 \neq r_2$ can be treated on the same footing. If r_2 is a known parameter, i.e. if we know the cross section of one of the atoms, we can rewrite the *k*-coefficients as depending on the only variable r_1 :

$$k_{1,2}(r_1, r_2, d) \to k_{1,2}(r_1)$$
(20)

We can thus define the auxiliary function

$$G_2(r_1) = \sigma_M - r\pi r_1^2 k_1(r_1) - 4\pi r_2^2 k_2(r_1), \quad (21)$$

and find the searched value of r_1 by imposing $G_2(r_1) = 0$.

Without increasing the complexity of calculations the above model for diatomic molecules can be, in some cases, easily extended to triatomic and polyatomic targets. In the case of linear or slightly bent triatomic molecules with identical satellite atoms, if the shadowing effect between the two satellite atoms is neglected the σ_M can be written as:

$$\sigma_M = 2\,\sigma_1 k_1 + (2k_2 - 1)\,\sigma_2 \tag{22}$$

where the index 2 refers to the central atom.

This approximation is certainly true for triatomic linear molecules, where the central atom is larger or comparable in size with the others. In the other cases the approximation (Eq. (22)) overestimates the exact theoretical prediction of the geometrical shadowing model. We have applied Eq. (22) to two linear (CO_2 , N_2O) and one bent (NO_2) triatomic molecules.

This approximation can be also adapted for highly symmetric polyatomic molecules with light satellite atoms. As a case study we have calculated σ_M for CH₄ with the formula:

$$\sigma_{\rm CH_4} = 4\sigma_1 k_1 + (4k_2 - 3)\sigma_2 \tag{23}$$

where 1 stays for H atom and 2 for the central C atom. Similarly as in the case of triatomic molecules, we disregard mutual shadowing effects.

3. Choice of the data

We have used the inversion algorithm described in the previous paragraph to determine the atomic cross sections for H, N and O starting from the measured TCS of diatomic homonuclear molecules. Then, with use of the direct algorithm (21) the atomic carbon TCS has been derived from measured data for CO. In the successive steps, these atomic cross sections were used in calculation f other molecular TCS.

Molecular bond lengths were taken from CRC Handbook [17]. As input values of the molecular TCS we have used the data from our laboratory [18–20] in the range from less than 100 eV to a few keV, and from the collaborating Gdansk laboratory for energies from 50 eV to more than 100 eV. These data, at overlapping energies agree well with other recent experiments [21,22]. The lower limit of the energy range for the application of our calculation has been chosen as 50 eV. This reflects the limits of applicability of any AR method. At energies below a few tens of eV, quantum effects in scattering become dominant so it is conceptually impossible to consider the molecular TCS as a superposition of the atomic ones.

As a final remark, note that the input TCS values actually used in our calculation, have been obtained by fitting Eq. (1) to the quoted experimental data. We use the fitted values since the experimental data are subject to statistical spread and are only available at selected energies.

4. Results and discussion

We report in Table 1 the present results for C, H, N and O targets. The results for H and O targets, at selected collision energies, are compared in Fig. 3 with some earlier calculations and available experimental data. The comparison shows a relatively good agreement with the optical potential calculations (Joshipura and Patel for the C, N, O targets [6] and Jiang et al. for the hydrogen atom [4]). When compared with the experimental determinations, the present results for O merge well with the beam-recoil experiment of Sunshine et al. [24], while they give the hydrogen TCS about 20% lower than the one measured recently by Zhou et al. [25]. However, a discrepancy for the hydrogen results is expected measurements for atomic hydrogen are difficult and the data can be subject to large uncertainties. On the other side, the molecular TCS for H₂ [20] are possibly subject to an angular resolution error at high energies. Note, that at 300 eV the present result is closer to the experimental TCS than the optical-model calculation of Jiang et al. [4].

Table 1

TCS for C, N, O, H as obtained by the present inversion procedure method applied to experimental data. TCS for NO, NO₂, N₂O, CO₂, CH₄, as obtained by the present geometrical AR method

<i>E</i> (eV)	С	Ν	0	Н	NO	NO_2	N_2O	CO_2	CH_4
50	6.61	7.51	6.28	2.20	9.70	12.12	13.35	14.72	10.16
100	5.37	5.94	5.09	1.47	8.00	10.26	11.11	12.55	8.23
200	3.84	4.11	3.64	0.86	5.93	7.90	8.38	9.69	6.13
500	1.99	2.05	1.93	0.39	3.39	4.84	4.95	5.51	3.33
1000	1.11	1.13	1.10	0.21	2.07	3.04	3.07	3.21	1.88
2000	0.6	0.60	0.59	0.11	1.15	1.71	1.71	1.75	1.00
5000	0.25	0.25	0.25	0.043	0.49	0.73	0.73	0.74	0.42

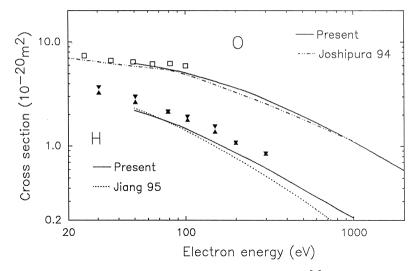


Fig. 3. Present results for H and O atoms compared to other calculations: Joshipura and Patel [6], complex optical potential for AR; the same method by Jiang et al. [4]. Experimental data: upper and lower limits for H from [25]; recoil-beam method for O [24].

In Fig. 4 the present results for NO, are compared with the AR and modified AR models of Joshipura

and Patel [6,12], and with the recent experimental data from our laboratory (unpublished) and previous

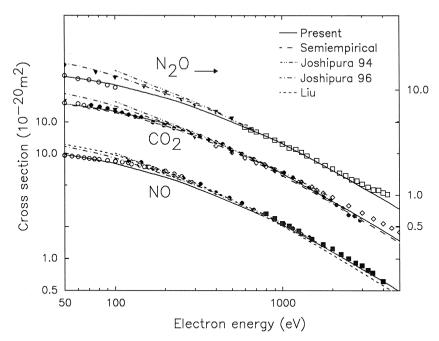


Fig. 4. Present results, solid line, for NO, CO_2 and N_2O molecules compared with other AR models: dashed-dot-dot line, simple AR [6],; dashed-dot line, AR with molecular polarizability [12]. For NO medium broken line, semiempirical model using formula (1) by Liu and Sun [5]; for CO_2 long broken line, semiemperical model [15], AR model of Ref. [12] coincides with geometrical shielding AR of Jiang et al. [14], not shown. Experimental data: open circles, Gdansk laboratory [23,27,30]; full circles, Trento laboratory [26,27]; for NO, squares, Trento laboratory, unpublished; inverted triangles, Detroit laboratory [29,31]; rhombuses [28]; open squares [32].

measurements [23,26]. Different AR formulations agree well at high energies but the present model is much closer to the experimental data than the results of Ref. [6,12]. In the same figure we report results of a semiempirical model of Liu and Sun [5] in which parameters *b* and σ_z in Eq. (1) have been correlated to the molecular bond length. This model overestimates the experimental cross section by 10% at 100 eV and underestimates it by a similar amount at 3000 eV.

Similar discrepancies are visible for the CO_2 molecule, see Fig. 4. The present model agrees well with experimental data [27–29] in the whole 50–5000 eV range. Previous modified ARs, both with geometrical shielding [14] and molecular polarizability [12] overestimate the experiments at low energies, by more than 20% at 50 eV. Fig. 4 shows also the results obtained with our previous semiempirical formulation [15] – the two results practically overlap.

For N_2O molecule, in Fig. 4 some discrepancy is observable between two existing low-energy measurements [30,31]. The present result, based on inversion of the low-energy Gdansk data [23], agrees better with the determination from the same laboratory [30]. The geometrical-shielding AR [14] agrees better with the experiment of Kwan et al. [31]. The simple AR [6] is higher than the experimental data in the whole 100-1000 eV, with a relative difference diminishing with the energy. The present model reproduces well the recent high-energy experiment [32] up to 3000 eV.

For the triatomic bent molecule, NO_2 previous AR models [6,12] overestimate the experimental data at 100 eV by a 30% and 20%, respectively. The present model is lower by 10% at 100 eV and merges with the experiments [33,34] at higher energies, see Fig. 5.

Present results for CH_4 fall between two existing experimental [35,36] determinations at high energies, while below 200 eV are somewhat (up to 20% at 50 eV) lower than the existing measurements [21,35]. The previous semiempirical result [15] performs better for energies lower then 200 eV. This difference can be attributed to the fact that in this paper we have used some simplifications to extend the formalism for triatomic molecules to polyatomic ones. On

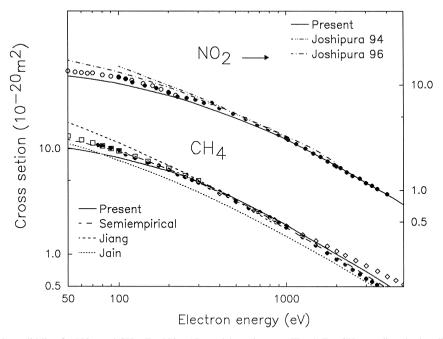


Fig. 5. Present results, solid line for NO_2 and CH_4 . For NO_2 AR model captions see Fig. 4. For CH_4 , medium broken line, simple AR [4]; short broken line, ab initio optical model [8]. Experiment: open circles, Gdansk [33,35]; full circles, Trento [34,35]; rhombuses [36]; open squares [21].

the other side, our previous formulation [15] includes explicitly the relation between TCS and polarizability.

We have to stress now that the accuracy of the present result depends on the experimental errors present in the starting data base of measured molecular cross section. Discrepancies between the experimental data set used in this paper and measurements from other laboratories are as big as 15-20% for some targets (N₂O [29,30] at 100 eV or CO₂ [27,28] at 3000 eV). In the future, a self-consistent application of the additivity rule could help to assess the quality of experimental data.

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