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Electron scattering by N_2 and CO at intermediate energies: 1–10 keV

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Abstract

Semi-empirical total cross sections for electron scattering from N_2 and CO in the energy range 1–10 keV have been obtained by combining transmission-beam measurements for impact energies up to 5 keV with an asymptotic behaviour at high energy according to the Born–Bethe approximation. The influence of the forward electron scattering in our experimental systems has been evaluated by means of a Monte Carlo electron transport simulation. This procedure has proved that electrons scattered in the forward direction may be the main error source in total cross section measurements at high energies, even in experiments with reasonable angular resolution. Serious discrepancies with results obtained with Ramsauer-type devices have been confirmed for this energy range.

1. Introduction

Recently, an experimental work devoted to a comparative study of the total cross sections (σ_T) for electron scattering by N_2 and CO in the energy range 80–4000 eV has been published by Karwasz et al. [1]. Apart from the important applications that the knowledge of these parameters have in many scientific and technological fields, the mentioned work was mainly prompted by a divergence which could exist in the previous measurements of García et al. [2,3], between the σ_T values of these molecules at high energies. The data of Refs. [2,3] were obtained by using two different experimental set-ups based on the same linear transmission-beam technique, while those of Ref. [1] were measured with a Ramsauer-type apparatus [4].

More recent experiments on electron–molecule (CO_2, NH_3) scattering at high energies (above 1000 eV) have shown serious discrepancies between the σ_T values obtained by García and Manero [5,6] in a linear apparatus, and those measured by Szmytkowski et al. [7] and Zecca et al. [8] with a circular Ramsauer-type system. This work has been motivated by the supposition that electron scattering in the forward direction could be the main cause of these discrepancies. In order to evaluate the influence of this effect in total cross section measurements, we have carried out Monte Carlo simulations of our previous electron transmission experiments [2,3]. The results of this calculation have been used to correct the experimental values where the predicted contributions for forward scattering were appreciable.

On the other hand, we present theoretical total cross sections which have been evaluated by means of the Born–Bethe approximation [9–11] in the framework of the independent atom model [12]. A semi-empirical expression for σ as a function of energy has been obtained by fitting the energy dependence of the experimental values to an asymp-

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totic behaviour according to the Born–Bethe calculations. This formula reproduces to good approximation the experimental results in the energy range 1–5 keV, and has been used to extrapolate σ_T values up to 10 keV.

2. Procedure

Transmission-beam experiments are based on the measurement of the attenuation of a linear electron beam through a gas cell containing the molecules of interest. The recorded beam intensity (I) follows the law

$$I = I_0 \exp(-nL\sigma_T), \tag{1}$$

where I_0 is the intensity of the primary beam, L is the interaction region length, n is the molecular density, which is obtained from the measurement of pressure and temperature in the gas cell, and σ_T is the total cross section.

However, Eq. (1) represents the ideal case, in which the beam is infinitely narrow and the solid angle subtended by the detector is zero. In a real experiment, the contribution of small-angle scattering [13] must be incorporated into Eq. (1), giving the expression:

$$I = I_0 \exp\left[-nL\sigma_T + n\int_0^L \mathrm{d}x \int_0^{\Delta\Omega(x)} \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right) \mathrm{d}\Omega\right], \tag{2}$$

where the corrective term represents the effect of the colliding electrons, at a distance X from the entrance of the gas cell, which are scattered into the solid angle subtended by the detector, $\Delta\Omega(x)$, and $\mathrm{d}\sigma/\mathrm{d}\Omega$ represents the sum of the elastic and inelastic differential cross sections.

According to Eq. (2), the apparent total cross section (σ_a) measured in a transmission experiment tends to be less than the true σ_T value:

$$\sigma_a = \sigma_T - \frac{1}{L}\int_0^L \mathrm{d}x \int_0^{\Delta\Omega(x)} \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right) \mathrm{d}\Omega. \tag{3}$$

The magnitude of the corrective term depends on the scattering geometry. A sketch of the scattering geometry used in the experiments of Refs. [2,3] is shown in Fig. 1. In this case, the geometrical characteristics are defined by the size of the apertures limiting the gas cell (d_1, d_2) and the entrance aperture to the analyser (d_3), as well as for the length of the collision chamber (L) and the distance between the end of the chamber and the entrance aperture of the analyser (D). The dimensions of these parameters for the experimental conditions of Refs. [2,3] are given in Table 1. To evaluate an upper limit for the corrective term of Eq. (3), the method commonly used by the different authors is that given by Blaaw et al. [13]. The procedure is based on the fact that the differential cross sections for high-energy electron scattering reach their maximum values for small θ angles, so if the energy resolution of the analyser is high enough to eliminate the inelastic part of the corrective term, it satisfies the inequality

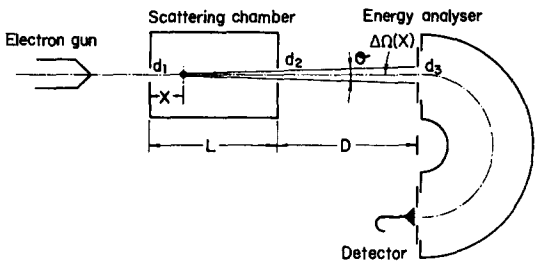

Fig. 1. Sketch of the scattering geometry of the experimental set-ups used in Refs. [2,3].

$$\int_0^L \mathrm{d}x \int_0^{\Delta\Omega(x)} \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\text{elastic}} \mathrm{d}\Omega \leqslant L\Delta\Omega\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\text{elastic},\theta=0}, \tag{4}$$

where $\Delta\Omega$ was defined in Ref. [13] as the solid angle subtended by the detector as seen from the centre of the collision chamber. Differential elastic cross sections ($(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\text{elastic}}$) for N_2 at impact energies between 100 and 3000 eV were measured by Jansen et al. [14]. Extrapolation to zero angle of these measurements indicates that 100 a_0^2/sr can be considered as a good upper limit of $(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\text{elastic},\theta=0}$ for energies above 1.0 keV. The angular acceptance ($\Delta\Omega$), as defined in Ref. [13], is of the order of 10^{-5} sr for both experimental systems used in Refs. [2,3]. This means that the relative error

Table 1
Values of the geometrical parameters which define the scattering geometry used in Refs. [2,3] (see fig. 1)

	Length (mm)		Diameter of apertures (mm)		
	L	D	d_1	d_2	d_3
Ref. [2]	150–300	50	2	2	1
Ref. [3]	70–127	150	1	1	1

contribution of electrons scattered elastically in the forward direction, i.e.

$$\frac{\sigma_a - \sigma_T}{\sigma_a} \leqslant \frac{\Delta\Omega}{\sigma_a}\left(\frac{d\sigma}{d\Omega}\right)_{\text{elastic},\theta=0}, \quad (5)$$

would be less than 0.1% even at 5 keV. However, this criterion is an approximation which does not take into account the dependence of the angular acceptance on the coordinates of each collision event, $\Delta\Omega(x, y)$, which can be relevant in some geometrical configurations. For this reason we have carried out a Monte Carlo simulation of the electron transport through the experimental set-ups used in Refs. [2,3]. The calculations have been made for N_2 at 1.0, 2.0 and 3.0 keV impact energies by using the elastic differential cross sections measured by Jansen et al. [14]. The corresponding total cross section data have been taken from Ref. [2]. Total elastic cross sections have been obtained by integrating an analytical fit to the differential values given in Ref. [14] by means of a non-linear least-squares method. The inelastic part was evaluated by subtracting those values from the experimental total cross sections. As the energy resolution used in both experiments was sufficient to discriminate inelastic processes, the path of electrons colliding inelastically were considered as finished in the simulation.

In accordance with the method used in Refs. [2,3], the simulation has been repeated for different target pressures ranging from 5 to 30 mTorr. Semi-logarithmic plots of the transmitted intensities (both including and not including elastically scattered electrons into the detection angle) were fitted by two straight lines. The relative difference between the simulated apparent and true cross sections $[(\sigma_a - \sigma_T)/\sigma_a)]$ was deduced from the slope of these lines.

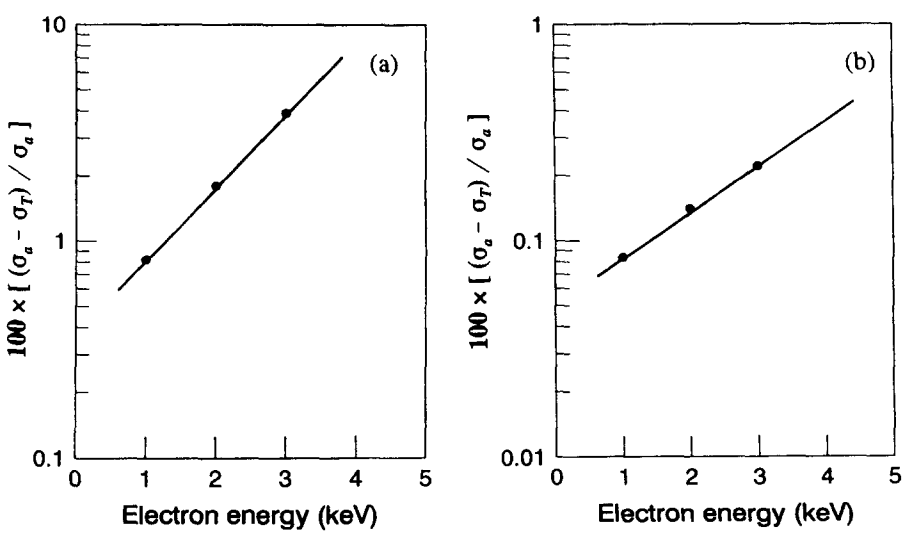

Fig. 2. Relative difference between the total cross sections with (σ_a) and without (σ_T) forward scattering contamination for e–N_2 interactions, deduced from the Monte Carlo simulation at 1, 2 and 3 keV. (a) For the experimental conditions of Ref. [2]. (b) For the experimental conditions of Ref. [3].

3. Results

3.1. Monte Carlo simulation

A semi-logarithmic plot, versus electron energy, of the results obtained from the Monte Carlo simulation is shown in Fig. 2a for the experimental conditions of Ref. [2], and in Fig. 2b for those of Ref. [3]. As shown in this figure, the error contribution of the forward scattering increases exponentially with energy and does not follow the quasi-linear dependence deduced from Eq. (5). Moreover, the correction percentage shows a clear dependence on the beam diameter and on the distance from the end of the collision chamber to the entrance aperture of the analyser. In the experimental conditions of Ref. [2] the correction is about 4% at 3.0 keV, and an extrapolation of these results to 5 keV could give values higher than 10%. However, in the scattering geometry used in Ref. [3] the influence of this efect is about 0.2% at 3 keV. According to this, the total cross sections for e–N_2 scattering given in Ref. [2] must be corrected, while the σ_T values for e–CO scattering given in Ref. [3] up to 5 keV can be considered free of forward scattering contamination, within the experimental errors (3%).

3.2. Born–Bethe calculations

A combined Born–Bethe theory [9–11] gives the formula for the total cross sections:

$$\frac{E_0}{R}\frac{\sigma_T}{a_0^2} = \pi\left[A_{el} + B_{el}\frac{R}{E_0} + C_{el}\left(\frac{R}{E_0}\right)^2 + \ldots\right] + 4\pi\left[M_{TOT}^2 \ln\left(4C_{TOT}\frac{E_0}{R}\right) + \ldots\right], \quad (6)$$

where E_0/R is the incident energy in Rydberg units and a_0 is the Bohr radius. The constants A_{el}, B_{el}, C_{el}, M_{TOT} and C_{TOT} are related to the internal dynamic properties of the targets (Inokuti [9], Inokuti and McDowell [10], Inokuti et al. [11]). In these works, calculations of the mentioned parameters are given for some atoms. In the case of N_2 molecules, direct calculations of these constants from molecular wavefunctions can be found in the work of Liu [15]. To our knowledge, similar data for CO molecules are not available in the literature. However, for energies high enough that the independent atom model of Mott and Massey [12] applies, and by introducing the optical theorem for the forward scattering amplitude, the total cross sections for molecular targets can be expressed, to a good approximation, in terms of those of their constituent atoms (Joshipura and Patel

Table 2
Total cross sections for electron scattering by N_2 and CO in the energy range 1–10 keV

Energy (keV)	Semi-empirical				Born–Bethe			
	present work		Karwasz et al. [1]		present work		Liu [15]	
	N_2	CO	N_2	CO	N_2	CO	N_2 [a]	N_2 [b]
1.0	7.49	7.71	7.33	7.35	10.2	10.6	10.5	9.56
1.5	5.37	5.63	5.14	5.13	7.09	7.34	7.33	6.62
2.0	4.25	4.52	3.96	3.95	5.46	5.66	5.66	5.09
2.5	3.56	3.83	3.21	3.20	4.46	4.62	4.62	4.14
3.0	3.08	3.34	2.71	2.70	3.78	3.91	3.92	3.50
3.5	2.73	2.97	2.34	2.33	3.20	3.40	3.41	3.04
4.0	2.46	2.68	2.06	2.05	2.91	3.01	3.02	2.69
4.5	2.24	2.45	1.84	1.83	2.61	2.70	2.71	2.41
5.0	2.07	2.26	1.66	1.65	2.37	2.45	2.46	2.19
6.0	1.79	1.95	1.39	1.38	2.00	2.08	2.08	1.85
8.0	1.43	1.54	1.05	1.04	1.54	1.59	1.60	1.42
10.0	1.19	1.28	0.84	0.84	1.25	1.30	1.30	1.15

[a] Calculations obtained from Hartree–Fock molecular wavefunctions, omitting electron correlation efects.
[b] Calculations obtained from the dipole oscillator strength distribution given in Ref. [19].

[16], Sun et al. [17], Jiang et al. [18]). Accordingly, the Born–Bethe total cross section (σ_{BB}) for electron scattering by N_2, evaluated by means of the atomic data given by Inokuti and McDowell [10] and Inokuti et al. [11], follows the expression:

$$\frac{\sigma_{BB}}{a_0^2}\frac{E_0}{R} = 441 + 72.9\ln\frac{E_0}{R} - 289\frac{R}{E_0} + \ldots. \quad (7)$$

Similarly, the σ_{BB} values for electron scattering by CO can be obtained from:

$$\frac{\sigma_{BB}}{a_0^2}\frac{E_0}{R} = 459 + 75.1\ln\frac{E_0}{R} - 295\frac{R}{E_0} + \ldots. \quad (8)$$

Data given by Eqs. (7) and (8) are shown in Table 2 for energies ranging from 1 to 10 keV. In this table are also included the Born–Bethe values obtained by Liu [15] for N_2 by two different methods, which differ mainly in the M_{TOT}^2 coefficent of Eq. (6). The first method is based on a Hartree–Fock calculation of the molecular wavefunction, omitting the effect of electron correlations. The second one, considered by Liu [15] as being more accurate, is based on the dipole oscillator strength distribution given by Zeiss et al. [19].

3.3. Semi-empirical fits

Experimental σ_T values given in Ref. [3] for the energy range 1–5 keV and the results of Ref. [2] corrected for forward scattering contamination up to 3 keV have been compared with those predicted by the Born–Bethe approximation. In both cases the calculated data are higher than the experimental ones. However, this discrepancy tends to be less as the energy increases. Hence we have studied their relative difference as a function of energy. Fig. 3 is a semi-logarithmic plot of $(\sigma_{BB} - \sigma_T)/\sigma_{BB}$ versus E_0/R, using the corrected values for N_2 (Fig. 3a) and the experimental data for CO (Fig. 3b) mentioned above. The points for N_2 lie on a straight line which can be fitted by the following exponential function:

$$\frac{\sigma_{BB} - \sigma_T}{\sigma_{BB}} = 0.32\exp\left(-\frac{1}{400}\frac{E_0}{R}\right). \quad (9)$$

Similarly, the data for CO can be fitted by the expression:

$$\frac{\sigma_{BB} - \sigma_T}{\sigma_{BB}} = 0.37\exp\left(-\frac{1}{240}\frac{E_0}{R}\right). \quad (10)$$

Thus semi-empirical total cross sections for N_2 at electron energies above 1 keV can be obtained from

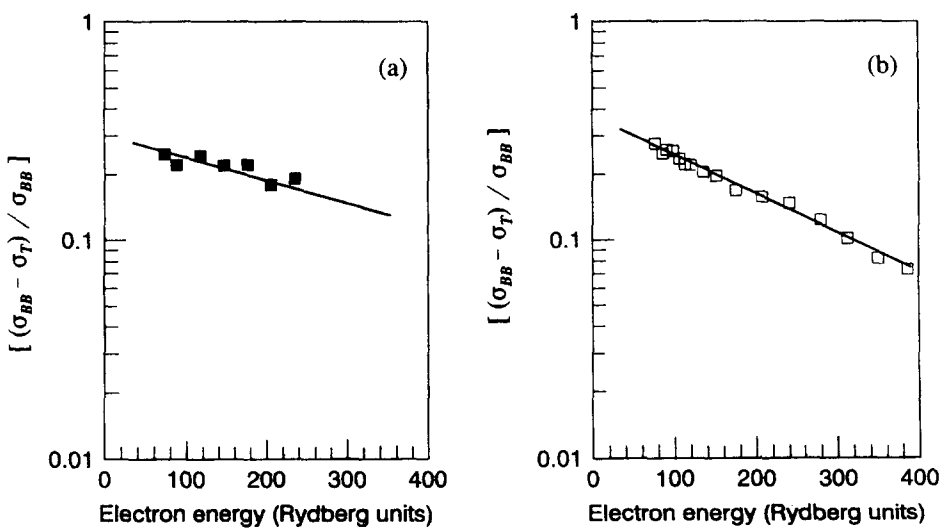

Fig. 3. Relative difference between the experimental total cross sections (σ_T) and those predicted by the Born–Bethe approximation (σ_{BB}) as a function of electron energy. (a) For N_2 using the experimental results of Ref. [2] corrected by the present Monte Carlo simulation. (b) For CO using the experimental data of Ref. [3].

$$\sigma_T = \left[1 - 0.32 \exp\left(-\frac{1}{400}\frac{E_0}{R}\right)\right]\sigma_{BB}, \qquad (11)$$

where σ_{BB} is given by Eq. (5). A similar expression can be written for CO as

$$\sigma_T = \left[1 - 0.37 \exp\left(-\frac{1}{240}\frac{E_0}{R}\right)\right]\sigma_{BB}, \qquad (12)$$

where σ_{BB} is given in this case by Eq. (6).

Semi-empirical total cross sections obtained from formulae (11) and (12), for impact energies ranging from 1 to 10 keV, are shown in Table 2. Data deduced from the semi-empirical fit given in Ref. [1] are also included in Table 2 together with the theoretical values given by the Born–Bethe approximation.

In order to estimate the accuracy of the semi-empirical σ_T values, the contribution of the partial errors introduced in formulae (11) and (12) has been studied. In accordance with Ref. [3], the experimental error for CO has been taken as $\pm 3\%$. For N_2 this error has been increased up to 4% to include the uncertainty introduced by the correction deduced from the Monte Carlo simulation. Concerning the σ_{BB} values, the spread of data for N_2 obtained from wavefunctions of different accuracy (see Table 2) is $\pm 6\%$. Thus we have considered this value as a good indication of the precision of the calculations, independent of the validity of the Born–Bethe approximation. By combining these uncertainties we have estimated an error limit of 7% for the semi-empirical total cross sections for e–CO interactions and up to 9% in the case of N_2. However, the good reproducibility of the experimental values could reduce this error limit by half for energies ranging from 1 to 5 keV.

4. Discussion and conclusions

As can be seen in Table 2, the results of the Born–Bethe calculation in the framework of the independent atom model for N_2 show a general good agreement, within 10% of those obtained by Liu [15] from molecular parameters. Similar calculations for CO show the same shape and trend as for the N_2 data, being 4% higher for the whole energy range (1–10 keV). Concerning semi-empirical values, the present results for CO are also slightly higher (about 8%) than those of N_2. These differences are within the estimated uncertainties, so they should not be considered as significant. However, the values deduced from the experimental results of Karwasz et al. [1] for both molecules are clearly lower than the present ones and the Born–Bethe calculations. This discrepancy increases with energy, reaching values of about 50% at 10 keV.

We can conclude that the apparent difference between the total cross sections for electron scattering by N_2 and CO deduced from Refs. [2,3] can be attributed to a remaining forward scattering contamination in the experimental conditions of Ref. [2]. Discrepancies in the results of Karwasz et al. [1] with the present results and those given for the Born–Bethe approximation persist for these molecules, being larger for increasing energies. This seems to indicate that a forward scattering simulation of the Ramsauer-type apparatus used in Ref. [1] would be required and especially in that case where inelastic scattering was only partially resolved [1].

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