23 February 2001

ELSEVIER

Chemical Physics Letters 335 (2001) 227–233

CHEMICAL PHYSICS LETTERS

www.elsevier.nl/locate/cplett

Cross-sections for electron scattering by O_2 at intermediate and high energies, 0.1–10 keV

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Received 17 November 2000; in final form 18 December 2000

Abstract

Cross-section data, both for elastic and inelastic electron collisions with molecular oxygen, in the energy range 0.1–10 keV, have been obtained in this study by combining theoretical and experimental methods. Accurate total cross-sections (within 3%) have been measured in a transmission beam system for electron energies ranging from 400 to 5500 eV. Differential and integral elastic cross-sections have been calculated by means of a scattering potential method in the framework of the independent atom model at electron energies ranging from 0.1 to 10 keV. The consistency of our theoretical and experimental results has been confirmed by comparison with previous elastic and ionisation experimental cross-sections.

1. Introduction

Important scientific and technological applications based on modelling of secondary electron interactions are demanding cross-section data for electron scattering over a wide energy range. Molecular oxygen plays an important role in atmospheric processes and other studies related with biological effects of radiation. However, collision data for electron scattering by O_2 at energies above 500 eV are scarce. At these energies (E), electrons are preferentially scattered around the forward direction. Therefore, accurate measurements require very high angular and energy (ΔE) resolution: $\Delta E/E$ must be in the order of 10^{-4} and the acceptance angle of the detector about 10^{-5} sr. From the theoretical point of view, large numbers of possible collision processes at these high energies require the use of different approximations (Born–Bethe theory, potential scattering methods ...). These considerations prompted the present Letter. In this work, accurate total cross-section (σ_T), within 3%, for electron scattering by O_2 in the energy range 400–5500 eV have been measured in a transmission beam experiment. As shown in previous papers [1–3], the first Born approximation overestimates cross-section values in this energy range and especially those corresponding to elastic processes. However, this approximation provides a correct asymptotic behaviour for the

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0009-2614/01/$ - see front matter
PII: S0009-2614(01)00051-3

total cross-sections at increasing energies. Therefore, we have used the Born–Bethe theory in the framework of the independent atom model to calculate σ_T values up to 10 keV. More accurate cross-section data, differential and integral, for elastic processes have been also calculated in this study by using a scattering potential involving static, exchange and polarisation terms. The consistency of the present experimental and theoretical results has been verified by comparison with previous data available in the literature.

2. Experiment

Absolute total cross-sections for electron scattering by O_2 have been measured in a transmission beam system for electron energies ranging from 400 to 5500 eV. The experimental set-up has been previously described [1–3] and will be only briefly referred to in this Letter. The electron beam had 1 mm diameter and the current operation was typically 10^{-13} A. The collision chamber was defined by two apertures of 1 mm diameter separated by a distance (L) which was varied from 70 to 127 mm according to the experimental requirements. An absolute capacitance manometer (MKS 127 A) measured gas pressure in the chamber. The energy of the emerging electrons from the gas cell was analysed by an electrostatic hemispherical spectrometer combined with a retarding field. Under these conditions, a constant energy resolution better than 1 eV was obtained for the whole energy range (400–5500 eV). A typical energy spectrum of electrons scattered into the detection angle is shown in Fig. 1. The exit of the collision chamber and the entrance of the analyser were separated by a vacuum length (D) of 150 mm which ensured that the acceptance angle of the analyser was lower than 3.5×10^{-5} sr. A two-stage microchannel plate operating in single pulse mode detected the transmitted electrons. The ultimate pressure in the region of the energy analyser and electron detector was on the order of 10^{-7} Torr.

The experimental method is based upon the attenuation of the electron beam measurement

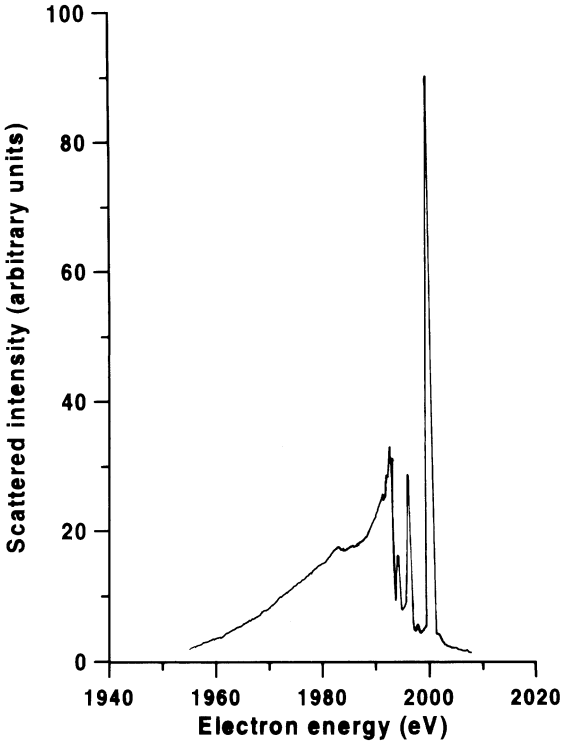

Fig. 1. Energy spectrum of electron scattered into the detection angle when the incident energy was 2000 eV and the O_2 pressure in the collision chamber was 45 mTorr.

when pressure in the gas cell was varied from 2 to 70 mTorr. Decay curves obtained in this way allows the total cross-section to be deduced from the slope of a semilogarithmic plot. Statistical uncertainties of the results were lower than 2%. Uncertainty in the pressure measurement was assumed to be lower than 1% (manufacturer data). Measurements were carried out at electron currents ranging from 10^{-13} to 10^{-15} A. For this current range, no dependence of σ_T on electron intensity was found. The definition of the actual absorption length was checked by repeating the measurement with two different collision chamber lengths, namely 70 and 127 mm. Both results were found to be in agreement within statistical uncertainties.

The electrons scattered in the forward direction cause the main error source at intermediate and high energies. However, in the condition of this experiment, electrons scattered inelastically into

the detection angle are completely discriminated by the energy analyser (see Fig. 1). The contribution of the elastic scattering to this effect was evaluated by means of a Monte Carlo simulation of the electron transport through the gas cell [4]. As shown in [4], the geometrical conditions of this experiment ensure that the error contribution of electrons elastically scattered into the detection angle is lower than 1%.

The experimental results obtained in this study are shown in Table 1 along with previous measurements, for comparison. By combining the above-mentioned error sources, an uncertainty of about 3% can be derived for the present measurements. As shown in Table 1, there is an excellent agreement, within error limits, with previous experimental data [5,6] at energies for which comparison is possible. Above 1.6 keV, the present results are the first measurements of the total cross-section for electron scattering by O_2.

3. Theory

3.1. Born–Bethe calculations

Assuming that the electron energy is high enough to allow the description of the incident and scattered particles in terms of plane waves, integral cross-sections can be calculated by means of the Born–Bethe theory [7–9]. In this approximation, the total scattering cross-section (σ_T^{BB}) can be written as

$$\frac{\sigma_T^{BB}}{a_0^2} = \pi\left[A_{el}\left(\frac{R}{E}\right) + B_{el}\left(\frac{R}{E}\right)^2 + \dots\right] + 4\pi\left[M_{inel}^2\left(\frac{R}{E}\right)\ln\left(4c_{inel}\frac{E}{R}\right) + \dots\right], \quad (1)$$

where E/R is the incident energy in Rydberg units (1 Ryd = 13.605 eV) and a_0 is the Born radius. A_{el}, B_{el}, M_{inel} and c_{inel} are defined by atomic targets

Table 1
Experimental total cross-sections, in atomic units (a_0^2), for electron scattering by O_2 in the energy range 0.4–5.5 keV

Energy (keV)	This work	Dalba et al. [5]	Dababneh et al. [6]
0.400	15.2	15.78	15.4
0.484		13.15	
0.500	12.6		12.5
0.576		11.72	
0.650	10.3		
0.676		10.32	
0.784		9.21	
0.870	8.11		
0.900		8.34	
1.024		7.44	
1.100	6.93		
1.156		6.76	
1.280	6.09		
1.296		6.12	
1.444		5.55	
1.500	5.48		
1.600		5.11	
1.750	4.92		
1.900	4.46		
2.150	4.16		
2.400	3.86		
2.850	3.41		
3.250	3.05		
3.750	2.73		
4.100	2.55		
4.550	2.36		
5.080	2.20		
5.500	2.03		

in [8,9]. For electron–molecule collisions at high energy, where the independent atom model [10] applies, and taking into account the optical theorem for the forward scattering amplitude [10], Eq. (1) can be expressed as a function of the constituent atoms of molecular targets [11]. Accordingly, parameters required in Eq. (1) can be calculated by using the atomic wavefunctions given by a standard Hartree–Fock procedure [12]. In these conditions, the total cross-section for electron scattering by O_2 is given by

$$\frac{\sigma_T^{BB}}{a_0^2} = 453\left(\frac{R}{E}\right) + 64.1\left(\frac{R}{E}\right)\ln\left(\frac{E}{R}\right) - 402\left(\frac{R}{E}\right)^2. \quad (2)$$

As expected, total cross-sections given by the Born–Bethe theory in the framework of the independent atom model overestimate experimental values in the energy range 0.4–5.5 keV. If the results given by (2) are compared with the experimental ones of Table 1, a maximum discrepancy of 45% is found for 0.4 keV. This discrepancy decreases with energy, reaching an excellent agreement for 5.5 keV. Calculated data are compared with the experimental ones in Fig. 4.

3.2. Scattering potential calculations

As shown previously [1–3], the failure of the Born approximation at low energies is mostly due to the elastic part. For this reason, it is necessary to study elastic processes by means of a more elaborated theory. We have recently applied [13] a scattering potential procedure to obtain differential and integral elastic cross-sections for electron scattering by noble gases, which showed a good agreement with the experiment. A similar proce-

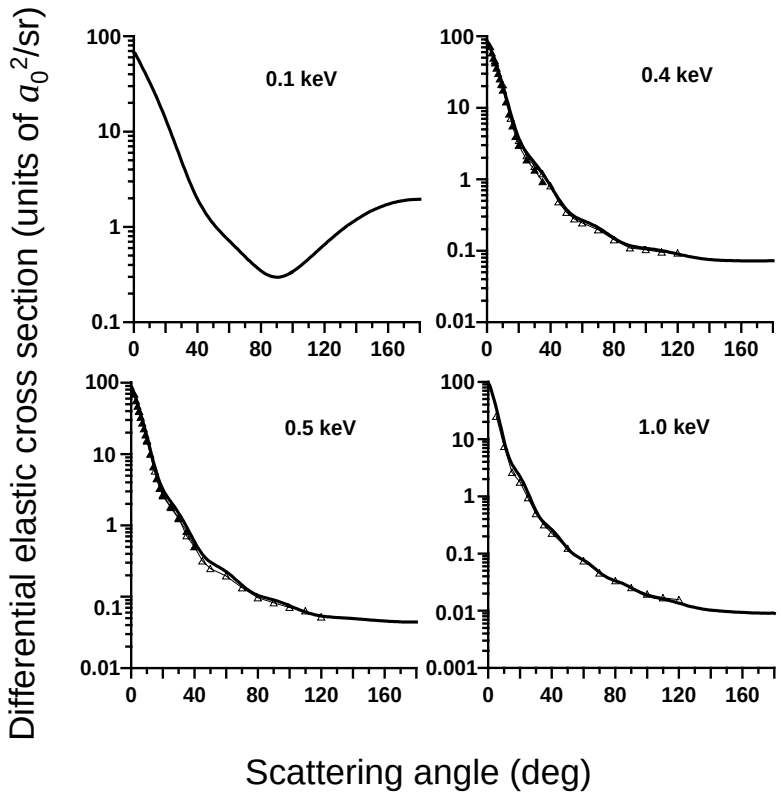

Fig. 2. Differential elastic cross-sections for e–O_2 collisions vs the scattering angle at 0.1, 0.4, 0.5, and 1 keV. Solid line, present calculations. ▲, experimental results of Bromberg [14]. △, experimental results of Iga et al. [15].

dure can be applied to molecular targets by assuming that the incident electron energy is high enough to validate the independent atom model [10]. In these conditions, the differential cross-section averaged over all orientations of the molecular axis is given by

$$\sigma_{\mathrm{M}}(\theta) = \sum_{i,j=1}^{N} f_j(\theta) f_i^*(\theta) \sin qr_{ij}/qr_{ij}, \tag{3}$$

where N represents the number of atoms within the molecule (here, $N = 2$), $f_j(\theta)$ is the scattering amplitude due to the jth atom of the molecule, $q(= 2k \sin\theta/2)$ is the magnitude of the momentum (k) transferred in the collision, and r_{ij} is the distance between the ith and the jth atoms. The atomic scattering amplitude $f_j(\theta)$ for oxygen atoms was obtained in a partial-wave expansion, given by

$$f_i(\theta) = \frac{1}{2ik} \sum_{l} (2l+1)\left(\mathrm{e}^{2i\delta_{l_j}} - 1\right) P_l(\cos\theta), \tag{4}$$

where $P_l(\cos\theta)$ are the Legendre polynomials and δ_{l_j} is the lth partial-wave phase shift for the jth atom of the molecule. To obtain δ_{l_j}, the appropriate scattering equation was solved numerically under proper boundary conditions [13]. The constituent atoms were replaced by a local spherically symmetric potential (V_{sc}). As shown in [13], this potential can be described for elastic processes as

$$V_{\mathrm{sc}} = V_{\mathrm{s}}(r) + V_{\mathrm{e}}(r) + V_{\mathrm{p}}(r). \tag{5}$$

Here V_{s} is the static potential for the electron–atom system, which has been calculated by using the atomic charge density deduced from Hartree–Fock atomic wavefunctions [12], and V_{e} and V_{p} represent exchange and target polarization contributions, respectively, as described in [13].

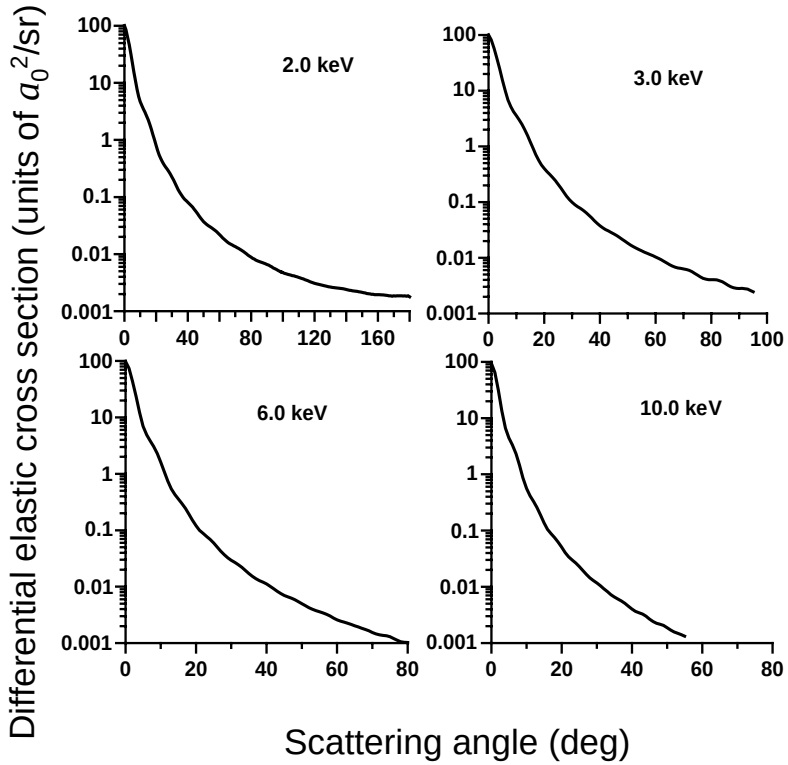

Fig. 3. Differential elastic cross-sections for e–O_2 collisions vs the scattering angle at 2.0, 3.0, 6.0, and 10 keV. Solid line, present calculations.

In Figs. 2 and 3, differential cross-sections for elastic scattering by O_2 calculated by the above procedure are plotted for 0.1, 0.4, 0.5, 1.0, 2.0, 3.0, 6.0 and 10 keV. As these figures show, there is a good agreement between the present calculations, the measurements of Bromberg [14] up to 500 eV and those of Iga et al. [15] up to 1 keV.

The integral elastic cross-sections deduced from the present calculation are shown in Fig. 4. Very recent calculations of Machado et al. [16] derived from the combination of the Schwinger variational iterative method and a distorted-wave approximation (see [16]) are also included for comparison. As this figure shows, there is an excellent agreement between both calculations in the overlapping energy region (100–500 eV).

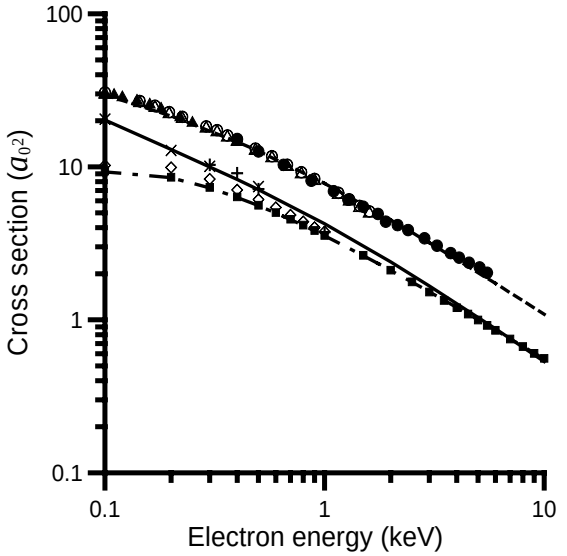

Fig. 4. Integral cross-sections for electron scattering by O_2. Elastic cross-section data: ——, present calculations; ×, calculations of Machado et al. [16]; +, measurements of Iga et al. [15]. Ionisation cross-section data: ■, experimental results of Schram et al. [17]; ◇, measurements of Krishnakumar and Srivastava [18]; –·–, calculations of Hwang et al. [19]. Total scattering cross-section data: ●, present measurements; ○, experimental values of Dalba et al. [5]; Δ, measurements of Dababneh et al. [6]; ▲, experimental values of Szmytkowski and Maciag [22]; – – –, theoretical values obtained by adding the present integral elastic cross-sections with the total ionisation (including neutral dissociation) cross-sections given in [19].

4. Discussion of results

As it has been already mentioned, there is a good agreement between the present total cross-section measurements and data existing in the literature, where the comparison has been possible. The agreement between our calculated elastic cross-sections and those measured by Bromberg [14] and Iga et al. [15] has also been shown in Figs. 2 and 3. In order to check the consistency between our experimental and theoretical results, we can incorporate data of complementary processes given by other authors. In the energy range considered here, ionisation and neutral dissociation are the most important inelastic processes (see Fig. 1). Representative measurements of ionisation cross-section on oxygen for electron energies ranging from 0.6 to 20 keV were given by Schram et al. [17]. Recently, new experimental data have been published by Krishnakumar and Srivastava [18]. Concerning theoretical ionisation cross-section, extensive data can be found in the paper of Hwang et al. [19], which were calculated by means of the binary-encounter-dipole model developed by Kim and Rudd [20]. This method has been demonstrated to be very useful in the study of electron impact ionisation of a large number of molecules over a wide energy range [21]. As shown in Fig. 4, there is a reasonable agreement between theoretical and experimental ionisation cross-sections for the energy range considered here. By adding the ionisation cross-section to the integral elastic one, values close to the total cross-sections must be obtained. In particular, by using data of Hwang et al. [19], which include neutral dissociation, the result of this addition must be practically the total cross-section. As Fig. 4 shows, this result is in concordance with the present experimental total cross-section and with previous measurements at lower energies [5,6].

5. Conclusions

An effective method to obtain reliable cross-section values for electron scattering by O_2 over a wide energy range (0.1–10 keV) has been presented in this paper. Accurate total cross-sections for

electron energies ranging from 0.4 to 5.5 keV have been measured in a transmission-beam experiment. For energies above 1.6 keV, these results are the first values reported in the literature. Differential and integral cross-sections have been calculated by means of a scattering potential method, which gave results in excellent agreement with previous experimental [14,15] and theoretical [16] data, where comparison has been possible. The consistency of the results presented in this paper with previous data of total ionisation cross-sections has been also proved.

Acknowledgements

This work was partially supported by the Spanish Programa Nacional de Promoción General del Conocimiento.

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