

Electron-molecules scattering cross section for use in ESEM: Application to H_2O and N_2

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Abstract

In this work, precise values are presented for elastic and total scattering cross sections as a function of energy for water vapor and N₂. The values we present gives accurate results that are in very good agreement with the experimental results. Hence, these cross sections values can be employed accurately and efficiently in a Monte Carlo simulation in ESEM, without the need to go through rather complicated look-up softwars, thus making the whole process quick, efficient and removing possible computational time and errors that may arise from different cross sections model. **Key-words :** *Optical Potential; Scattering cross section; ESEM; H₂O; N₂*

Résumé

Dans ce travail, des valeurs précises sont présentées pour les sections efficaces de diffusion élastique et totale en fonction de l'énergie pour la vapeur d'eau et N2. Les valeurs que nous présentons donnent des résultats précis et en très bon accord avec les résultats expérimentaux. Par conséquent, ces valeurs peuvent être utilisées avec précision et efficacité dans les simulations de Monte Carlo dans un ESEM, sans passer par des logiciels de recherche plutôt compliqué, rendant ainsi l'ensemble du processus rapide et efficace en éliminant le temps de calcul et les erreurs qui peuvent provenir de différents modèles de sections efficaces.

Mots-clés : Potentiel optique; section efficace de diffusion; ESEM, H₂O ; N₂

1. Introduction

Environmental SEM (ESEM) allows even nonconducting, out-gassing, dirty, oily or wet samples to be examined non-destructively in humidified or gaseous environment. With ESEM, many experiments may be performed. Low voltage ESEM (LVESEM) is a promising new technique for morphological characterization. Operating at low voltage has a particular advantage on the resolution. The main limitation concerning the use of the ESEMs is the scattering of the electrons of the primary beam with the molecules of the gas. To do this, electron scattering cross sections are needed, as a function of the energy of electron beam.

An analytical derivation of the differential cross-section has been presented by Lenz [1] and adapted by Jost and Kessler [2] and Danilatos with multiple scattering approaches [3]. Farley & Shah [4] also examined electron beam scattering in a low vacuum environment for the case of N₂ and H₂O. Theoretical scattering cross sections for N2 were obtained using the same general procedures and assumptions as that of Moncreif et al. [5] witch used single scattering regime. However, the theoretical total scattering cross sections obtained by Farley & Shah were found to be approximately 50% higher in magnitude for the same beam energy. This difference was attributed to the use of different effective atomic radii; however it seems that no definitive values of the cross-sections have been claimed yet. The differences between theoretical and experimental results remain without some convincing argument in favor of one or the other outcome.

2. Physical model

The first step to develop a general scattering calculation framework for electrons, based on independent atom representations [6-7], is to describe interactions with atomic targets as accurately as possible. For this purpose, we represent scattering from atoms by an interacting complex potential (i.e. the optical potential):

$$V(r) = V_{st}(r) + V_{ex}(r) + V_{cp}(r) + iW_{abs}(r)$$
(1)

where Vst(r) is the electrostatic potential, Vex(r) is the local exchange potential, Vcp(r) is the correlation–polarization potential and Wabs(r) is the absorption potential.

This later is obtained from the local-density approximation, using the Born–Ochkur approximation and the Lindhard dielectric function to describe the interactions with a freeelectron gas, and multiplied by an energy-independent empirical strength factor Aabs.

$$W_{abs} = A_{abs} \frac{\hbar}{2} [v_L \rho_e \sigma_{bc} (E_L, \rho_e, \Delta], \qquad (2)$$

$$v_L = (2E_L/m_e)^{1/2}$$
(3)

corresponding to the local kinetic energy:

$$E_{L}(r) = E - V_{st}(r) - V_{ex}(r)$$
(4)

With an appropriate selection of $A_{abs}(3<A_{abs}<4)$, the absorption cross section, obtained from the partial-wave calculation with the aid of the optical theorem, practically equals the inelastic cross section for projectiles with energies from about 100 eV and higher.

The electrostatic potential Vst(r) is calculated from knowledge of the nuclear and electronic charge distributions. The nucleus is assumed to have a finite size. The corresponding positive charge density is described by the Fermi distribution. The electron density is obtained from self-consistent relativistic Dirac-Fock calculations (DF). The local exchange potential, Vex(r), accounts for the indistinguishability of the projectile and the electrons of the target atom. The recommended exchange potential is that of Furness and McCarthy [8]. The correlation-polarization potential combine the empirical Buckingham potential with the correlation potential Vcor(r) obtained from the localdensity approximation (LDA)[9]. The imaginary absorption potential, i.e., the imaginary part of the optical potential is calculated from the local-density approximation proposed by Salvat[9] and It's obtained rom the local-density approximation, using the Born-Ochkur approximation and the Lindhard dielectric function to describe the interactions. Note that the complex optical potential as such does not require any fitting procedure; however, it was possible to vary one parameter (A, the potential strength in the evaluation of absorption potential) in the calculation to bring theory and experiment even closer to each other.

In order to obtain molecular cross sections, the independent atom model has been followed by applying an addition procedure, commonly known as the additivity rule (AR). Thus, the total cross section for a molecule is the incoherent sum of the total cross sections for the constituent atoms. Raj [10] made the first application of the additivity rule to obtain the elastic cross sections for electron scattering from simple molecules. Joshipura and Patel [11] and Sun et al. [12] also employed the additivity rule to obtain the total cross sections (elastic and inelastic) for electron scattering with simple molecules, and proved that the additivity rule is proper for the calculation of the total cross sections for electron scattering from simple and smaller molecules. Assuming that each atom in a molecule scatters independently and that redistribution of atomic electrons due to the molecular binding is unimportant, we can reduce the electron molecule collision into the problem of collision with individual atoms. The main limitation of the AR is that no molecular structure is considered, thus it is really only applicable when the incident electrons are fast enough to effectively "see" the target molecule as a sum of the individual atoms. In the other hand, the atom in the molecule is different from the one in the free stat. thus we must introduce some property of the molecule. This can be any molecular property that accounts for the structure and/or energies of the molecule. So in this paper, the

individual atomic cross sections are calculated after replacing the first excitation potential of the constituent atoms by the first excitation channel of the molecule, and taking in a count the molecular polarizability. For non-polar molecules with induced dipole polarizability α_d^{mol} , the longrange polarization potential is $-\alpha_d^{mol}e_2/(2r4)$. For polar molecules, the effective dipole polarizability αd ,eff(i) of the ith atom of the molecule is proportional to the polarizability of the free atom, $\alpha d(i)$, and that the sum of effective atomic polarizabilities must be equal to the molecular polarizability. Thus, in the calculation of scattering amplitudes from the ith atom, we use the effective polarizability [9]:

$$\alpha_{d,eff}(i) = \alpha_d^{mol} \alpha_d(i) \left[\sum_j \alpha_d(j) \right]^{-1}$$
(5)

where the summation extends over all atoms in the molecule. Consequently the following AR model can be obtained: Thus the total and differential elastic cross sections for a molecule are given by:

$$\sigma_T = \frac{4\pi}{k} Im f_M(\theta = 0) = \frac{4\pi}{k} Im \sum_{j=1}^N f_j(\theta = 0) = \sum_{j=1}^N \sigma_T^j(E)$$
(6)

$$\left(\frac{d\sigma}{d\Omega}\right)_{Incoh} = \sum_{i} \frac{d\sigma_{i}}{d\Omega}$$
(7)

In the other hands, and with the assumptions stated above, the elastic scattering from the molecule can also be approximated as coherent sum of the wave functions scattered from all atoms in the molecule, averaged over all orientations of the molecular axis:

$$\left(\frac{d\sigma}{d\Omega}\right)_{Coh} = \sum_{i,j} \frac{\sin(qr_{ij})}{qr_{ij}} \left[f_i(\theta)f_j^*(\theta) + g_i(\theta)g_j^*(\theta)\right]$$
(8)

where $fi(\theta)$ and $gi(\theta)$ are the direct and spin-flip scattering amplitudes respectively, for the ith atom in the molecule. rij is the separation between the ith atom and the jth atom present in the molecule. q is the magnitude of the momentum transfer in the collisions.

This approximate DCS, overestimate at intermediate and low energies. Typically, at energies below few keV, the small angle DCS are found to be larger than the experimental data. This overestimation in molecular cross section can be attributed to two the fact that mutual overlapping by neighboring atoms is ignored; as a result the inner atoms are partially screened by the outer atoms. The inclusion of the 'screening effect' leads to a decrease in molecular scattering cross section than that predicted by coherent AR. However, incorporation of the screening effect at the molecular level is difficult theoretically.

3. Results and Discussion

The present work proposes, a theoretical calculation of the elastic and total scattering of electrons by water vapor and N2 molecules in the ESEM for a large range of incident

energies, and the good agreement with the existing experimental data verifies the accuracy of the calculated cross sections even in the low-energy domain where fine effects such as polarization and exchange are introduced. Fig.(1,2,3) shows a comparison of our results with the experimental measurements of Danjo and Nishumira [13], Kataz et al. [14], Shyn and Grafe [15], Dubois and Rud [16], Jensen et al. [17], Bromberg et al. [18] and Herrmann et al.



Fig.1. Differential elastic-scattering cross sections, $d\sigma/d\Omega$ at 200eV. (A):N₂ (B):H₂O





Fig.

2. Differential elastic-scattering cross sections, $d\sigma/d\Omega$ at 500 eV. (A):N2 (B):H2O



Differential elastic-scattering cross sections, $d\sigma/d\Omega$ at 1000eV. (A):N₂ (B):H₂O

The elastic DCSs calculated with the complex optical potential are clearly in better agreement with the measurements. The agreement between theory and experiment is observed in the general behavior, i.e., both in the shape and absolute nature of the angular distributions of the incoherent DCSs. Note that coherent scattering reproduce well the experimental differential CS but gives much higher integral values (35% relative error at low energy and 10% at high energy).

By comparison, we have reported in all figures the theoretical singly elastic differential cross sections commonly used in our earlier electron track-structure obtained by using the Lenz formula [1]. Thus present very large discrepancies with our results, especially in the low-energy regime. In fact, this formulation is inappropriate to describe the low energy electron scattering and experimental data are usually introduced in the Monte Carlo simulation. However, for higher energies (5000 eV), the Lenz formula can be applied and gives results similar to ours, except in the small scattered angle domain.

Table 1. Elastic electron and total scattering from H_2O (in units of $10^{-16}\,\mbox{cm}^2).$

Energy (eV)	H ₂ O									
		El	astic (σ_e)	Total (σ _T)						
	Inc	Coh								
200	2.02	3.14	2.11[14]	5.0	4.80[21]					
500	1.00	1.37	1.04[14]	2.5	2.48[21]					
1000	0.58	0.756	0.55[14]	1.41	1.42[21]	2.13[27]				
5000	0.144	0.173	0.146[24]	0.36	0.37[25] 0.38[2	6] 0.55[27]				
10000	0.075	0.09	0.079[24]	0.19	0.19[25] 0.31[2	6] 0.30[27]				

Table 2. Elastic electron and total scattering from N_2 (in units of $10^{\text{-16}}\,\text{cm}^2).$

Energy (eV)	N ₂										
		Ela	astic (σ_e)		Total (_T)						
	Inc	Coh									
200	3.44	4.68	3.50 [29]		6.40	6.43[28]					
500	1.815	2.33	1.82 [18]		3.55	3.58[22]					
1000	1.07	1.32	1.20 [19]	1.0[29]	2.03	2.08[22]		3.0 [27]			
5000	0.26	0.30	0.298[30]		0.56	0.57[26]	0.58[23]	0.778[27]			
10000	0.133	0.158			0.29	0.31[26]	0.33[23]	0.426[27]			

The present results for the total cross sections for electron scattering from N_2 and H_2O are shown in Tables 1 and 2 with the available experimental and theoretical data. From it, we can see that the present results are in good agreement with the experimental data above 200 eV. For example, the larger differences between the present results and the measurements of Zecca et al. [21], and Karwaz et al. [22] are only 5% at all energies for H2O and 3% for N2. The present results are in good accord with the empirical formula of Garcia and Manero [23] above 800 eV for N2 and above 1000 eV for H2O with the complex optical potential of F.Blonco et al. [24]. Above 5000 eV, there are scar of experimental data, so the present results are compared with the data of Scott et al. [25] and Rattenberger et al. [26], the results of the empirical formula of Garcia and Danilatos [27]. Good agreements are obtained.

Conclusion

We have presented values of the elastic and total cross sections for different energy electron impact with water vapor and N₂. A complex optical potential is derived for each system from target wave functions. The present model mainly requires the target charge density, polarizability, excitation potential, etc., of the molecule and especially the best choice of the potential strength value. Our results are in very good agreement with available measurements.

References

- [1] Lenz F. Z Naturforsch 1954;9a:185–204.
- [2] Jost K, Kessler J. Z Phys 1963;176:126–49.
- [3] Danilatos GD. Scan Microsc Int 1990;4:799-823.
- [4] Farley AN, Shah JS. J Microsc 1990;158:379–88.
- [5] Moncrieff DA, Robinson VNE, Parker PR. J Phys D ApplPhys 1976;12:481–8.
- [6] A.C. Yates, Phys. Rev. 176 (1968) 173.
- [7] D.W.Walker, Phys. Rev. Lett. 20 (1968) 827.
- [8] J.B. Furness, I.E. McCarthy, J. Phys. B: At. Mol. Phys. 6 (1973) 2280.
- [9] F.Salvat, Phys. Rev. A 68 (2003) 012708.
- [10] D. Raj, Phys. Lett. A 160, 571 (1991).
- [11] K. N. Joshipura and P. M. Patel, Z. Phys. D 29, 269 (1994).
- [12] J.F. Sun, Y. H. Jiang, and L. D. Wan, Phys. Lett. A 195, 81(1994).
- [13] Danjo A and Nishimura H 1985 J. Phys. Soc. Japan 54 1224–7.
- [14] Katase A, Ishibashi K, Matsumoto Y, Sakae T, Maezono S, Murukami E, Atanabe K and Maki H 1986 J. Phys. B:At. Mol. Phys. 19 2715–34
- [15] Shyn T W and Grafe A 1992 Phys. Rev. A 46 4406
- [16] R D DuBois and M E Rudd. J. Phys. B: Atom. Molec. Phqs., Vol. 9, No. 15, 1976
- [17] Jansen R H J, de Heer F J, Luyken H J, van Wingerden B and Blaauw H J 1976 J. Phys. B: Atom. Molec. Phys. 9 185
- [18] Bromberg J P.1970 J.Chem.Phys.52 1243
- [19] Herrmann D 1974 Verhandlunyen der DPG.Friihjahrstagung, Stuttgart (Weinheim: Physik-Verlag) p 413
- [20] Danilatos, G.D., 1988.Advances in Electronics and Electron Physics 71, 109–250.
- [21] Zecca A., Karwasz G., Oss S., Grisenti R. and Brusa R.S., J. Phys. B, 20 (1987) L133.
- [22] Karwase G., Brusa R. S., Gasparoli A. and Zecca A., Chem. Phys. Lett., 211 (1993)529.
- [23] G. Garcia, M. Roteta, F. Manero. Chemical Physics Letters 264 (1997) 589-595
- [24] F. Blanco, A.Munoz, D.Almeida, F.Ferreira da silva, P.Limao-vieira, M. Fuss, and A.G. Sanz, G. Garcia. eur. phys. j. d (2013) 67: 199
- [25] Wight, S.A., Konicek, A.R., 2012. Micron 43, 985–991.
- [26] J.Rattenberger, J.Wagner, Schröttner, S.Mitsche, A.Zankel, Scanning 31 (2009) 1–7

[27] GD.Danilatos, Micron 45 (2013) 1-16

- [28] Nickel J. C., Kanik I., Trajmar S. and Imre K., J. Phys. B, 21 (1992) 2427.
- [29] Yukikazu Itikawa. J. Phys. Chem. Ref. Data, Vol. 35, No. 1, 2006
- [30] Ashok Jain and K. L. Baluja. Physical Review , volume 45, number 1 1 january 1992