Doubly Differential Cross Section for Electrons Scattered by Nitrogen

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A semiemprical model, as initially proposed by Rudd [Phys. Rev. A, 44, 1644 (1991)], is used to describe the doubly differential cross section for electron-impact ionization of molecular nitrogen. Model parameters for the molecule are determined by fitting to the available experimental data. A considerably good agreement is observed between the experimental results obtained by means of time-of-fliglit technique and the values given by the model.

I. Introduction

Nitrogen is the most abundant atmospheric molecule and been extensively studied in the last decades. A few secondary-electron-production cross sections for electron-impact ionization of molecular nitrogen are available in the literature[1–7]. Such dats is of fundamental interest due to their importance in testing theoretical descriptions of the ionization process. Disagreements in the shape of the angular distributions of ejected electrons are observed among the experimental results on doubly differential cross sections (DDCS) for molecular nitrogen, considering those reported since the 70's[2–7]. We are not aware of any attempt to calculate a rigorous quantum mechanical model to describe the emission processes for the molecule. In 1991, Rudd described a semiempirical model that provided analytical expressions for the singly and doubly differential and total cross sections[8]. It seemed to be successful for helium[8] and molecular hydrogen[8,9] targets. As stated by the author, no attempt was made to apply the model to targets containing more than one shell in order to avoid complexities that could be attributed to inner-shell contributiom[8]. In this work, we use the semiempirical model for molecular nitrogen. As in the helium and molecular hydrogen cases, parameters for the investigated molecule were obtained by fitting to the experimentally available data. A good description seems to be provided by the model for molecular nitrogen electron-impact ionization cross section, differential in energy and angle of ejection. Overall, they seem in good agreement with those resuls presented by Goruganthu et al.[7] who have energy analyzed scattered and ejected electrons by means of the time-of-night technique. Their results are presented for ejection angles from 30° to 150°, in steps of 15°, at 2, 4, 6, 8, 10, 20, and 40 eV ejected energies. Primary energies are of 200, 500, 1000, and 2000 eV, a range wide enough for optimal use in applications related to radiation chemistry, biology, and plasma physics, among other fields. Other results obtained by use of other techniques[2,4,6] are also compared to the semiempirical data obtained in this work.

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II. Theory

The doubly differential cross section (DDCS) can be obtained by measuring the energy and angular distributions of outgoing electrons with only one electron detector. Numerical integration of the DDCS over all angles is usually carried out to obtain the singly differential cross section (SDCS):

$$\sigma(W) = 2\pi \int_0^{\pi} \sigma(W,8) \sin\theta d\theta \ , \qquad (1)$$

where W is the ejected-electron energy and θ is the scattering angle. Integration of the DDCS over both angle and ejection energy yields the total ionization cross section (TICS):

$$\sigma_{\rm ion} = (1/2) \int_0^{T-T_{\rm ion}} \sigma(W) dW \ , \qquad (2)$$

where T is the primary energy and $I_{\rm ion}$ the ionization potential. The factor $(1/2)$ takes into account the secondary and the scattered primary electrons outgoing from each ionizing collision.

Working with the reduced quantities w $= W/I_{\rm ion}$, and t $= T/I_{\rm ion}$, the semi-empirical expression for the DDCS can be written as:

$$\sigma(w,t,\theta) = G_1[f_{BE}(w,t,\theta) + G_2 f_b(w,t,\theta)] \ , \qquad (3)$$

where

$$G_1 = [SF(t)f_1(w,t)/I_{\rm ion}]/[g_{BE}(w,t) + G_2 g_b(w,t)] \ , \qquad (4)$$

$$f_{BE}(w,t,\theta) = 1/\{1 + [(\cos\theta - G_3)/G_4]^2\} \ , \qquad (5)$$

which represents the binary-encounter peak, and

$$f_b(w,t,\theta) = 1/\{1 + [(\cos\theta + 1)/G_5]^2\} \ , \qquad (6)$$

isto describe the possible rise in the cross sections in the backward direction as seen in the H_2 [9,10], He [3,11], N_2 [4,6] and H_2O [3,12,13] cases, with G_5 adjusted for the best fit to the data. In Eq. (4), the numerator gives the SDCS with $S = 4\pi a_0^2 N(R/I_{\rm ion})^2$ (a_0 being the Bohr radius, N the number of eletrons in the target, and R = 13.6 eV), and $F(t)$ being a function that is $(1/t)Int$ dependent at high energies, as verified by Bethe[14] by use of the Born approximation, given by[9]

$$F(t) = A_1 Int/t + A_2/(t + A_4) + A_3/t^2 \ , \qquad (7)$$

where A_1, A_2, A_3, and A_4 are parameters to be adjusted by fitting to the experimental data. The f_1 term in the SDCS expression will be taken as[9]

$$f_1(w,t) = [1/(w+1)^n] + [1/(t-w)^n] - [1/((w+1)(t-w))^{n/2}] \ , \qquad (8)$$

which appears in the Mott cross section for a collision with a bound electron when n $\simeq$ 2 [15]. The second term in the equation above is due to exchange while the third one represent an interference. The other model parameters are

$$g_{BE} = 2\pi G_4\{\tan^{-1}[(1-G_3)/G_4] + \tan^{-1}[(1+G_3)/G_4]\} \qquad (9)$$

$$g_b = 2\pi G_5 \tan^{-1}(2/G_5) \ , \qquad (10)$$

$$G_2 = \gamma\{[1-(w/t)]^3/[t(w+1)] \ , \qquad (11)$$

with

$$G_3 = [(w+1)/t)^{1/2} \ , \qquad (12)$$

as given by Kim[16],

$$G_4 = \beta[(1-G_3^2)/w]^{1/2} \ , \qquad (13)$$

as given by Rudd[8], and with β and γ being adjustable parameters.

III. Results and discussions

In order to start using the semiempirical model to analyze the DDCS for molecular nitrogen (N=14), it was necessary to rely on some initial parameters.

It is well ltnown that molecular nitrogen has five diferent ionization energies (15.6, 16.9, 18.7, 37.3, and 409.5 eV) leading to the formation of N_2^+ [17]. Obviously, the last ionization channel is closed for 200-eV incident electrons. The lowest ionization energy value,

15.6 eV, that leads to the formation of the single positive ion was taken into account as the **I„** value in this work since it was the one used by Goruganthu et al.[7] to determine their ejected energies ancl also to define the Rutherford cross section they employed for carrying Platzman plot analysis (see their Fig. 4) out. So, in taking the lowest ionization energy for the nitrogen molecule we have not considered any contribution from the other various orbitals of the target in the calculation of DDCS values.

The initial values of $G_5 = 0.33$, $\beta = 0.60$, and $\gamma = 10$ were those suggested by Rudd et al.[9] for $e^- + H_2$ collisions. With these values and the results of G_3, which we obtained in this work by use of Eq. (12), the values of $g_b = 2.9$, G_2, G_4, and g_{BE} were then obtained. As emphasized by Rudd[8], tlie parameters G_3 and G_4, and hence f_{BE}, carry information on the position and shape of the binary peak. However, the use of Eqs. (5), (12) and (13) makes them independent of the target being analyzed. Just to mention, while Rudd et al[9] employed Eq. (12) for $e^- + H_2$ collisions, more recently Berakdar and Klar[18] used a slightly different expression for tlie binary peak center, $G_3 = \cos\theta_b = (w/t)^{1/2}$, when investigating structures in triply and doubly differential ionization cross sections of atomic hydrogen.

The value of "n", in the f_1 term in Eq. (8), was initially obtained by fitting integral elastic cross section results from Mu-Tao and Freitas[19] theoretically obtained by use of a renormalized potential model for incident electrom with energies from 50 to 800 eV.

With all the values showed above, a first set of values for the parameters $A_1, ..., A_4$ in Eq. (7) was obtained to describe the experimental DDCS results for molecular nitrogen reported by Goruganthu et al.[7]. The DDCS results then obtained are shown In Fig. 1 for 200-eV incident electrom and ejected energies of 10 and 20 eV. Although quantitatlve agreement between the results seems poor, the model predicted quite well the center of the binary peak, near 70° for both ejected energies. Shallow minimaat higher scattering angles, above 120°, were also predicted by the model.

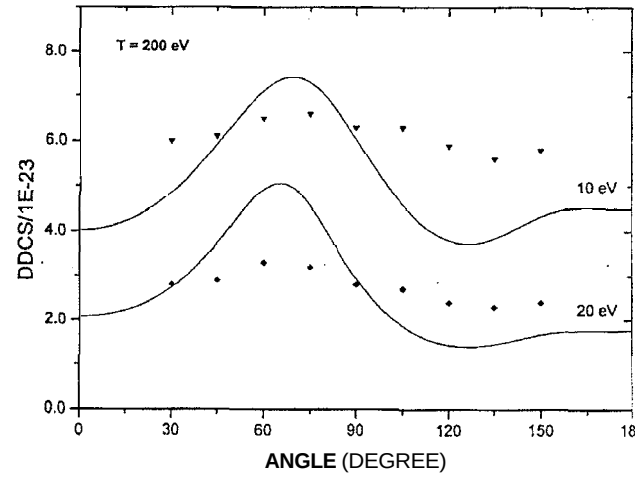

Figure 1: DDCS for 200-eV $e^- + N_2$2 collisions vs. scattering angle for ejected electrons of 10 and 20 eV. The line is obtained using tlie following parameters: $A_1 = 0.275, A_2 = 0.75$, $A_3 = -2.0$, $A_4 = 3.0$, and n = 2.86. The experimental results are from Goruganthu et al.[7].

Further adjustments of the model parameters, including those initially fixed, improved the fitting to the data. Values of parameters used in this work for tlie best fitting to the inolecular nitrogen DDCS, in comparison to the experimental data of Goruganthu et a~.[~], are listed in Table 1. Tlie DDCS results are plotted in Figs. 2 - 5. Overall, as it can be seen, the model provides a good qualitative description of the DDCS within the experimental error bars reported in Ref. 7. It predicts the shifting of the center of the binary peak to higher angles as the primary energy increases. Minima in the bacltwarci direction are also predicted by tlie model for low-energy ejected electrons. They become more prominent at lower primary energies. Better agreement among tlie experimental data from Ref. 7 ancl the theory is observed as the ejection as well as tlie incident energies increase. For 10-eV ejection energy, the discrepancy between the results gets larger as the electron incident energy increases.

When the ejected electron energy is 2 eV, adjustments to the model do not allow reproduction of the experimental results of Goruganthu et al.[7] at primary energies of 200 and 500 eV. The slope of the curve given

by the semieinpirical model employed here suggests the shape of the results reported by Goruganthu et al.[7] for primary energies of 200 and 500 eV. The DDCS values reported by Shyn[6] by rneans of a crossed-beam method exhibited a prominent peak in the forward direction at lower ejected energies. A similar result lias been reported by DuBois and Rudd[4] who have used a static gas cell. Experimental molecular nitrogen DDCS results are also reported by Opal et al.[2] at 200, 500, 1000, and 2000 eV and for ejection energies above 6 eV. Overall, their results qualitatively reproduce those reported by Goruganthu et al.[7]. Results from Opal et al.[2], DuBois and Rudd[4], and Shyn[6] are not shown in Figs. 2 - 5

Table 1 - Values of fitting parameters.

A1	0.3
A2	0.45
A3	-2.0
A4	3.0
n	2.13
G5	0.33
β	1.0
γ	10.0

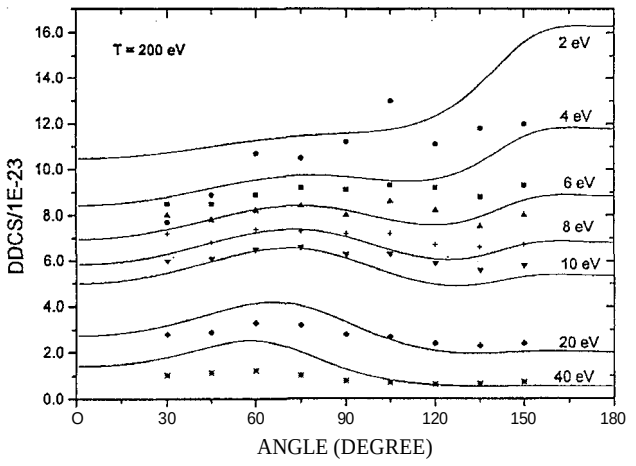

Figure 2: Results of DDCS vs. scattering angle for 200-eV incident electrons. The experimental results are from Goruganthu el al.[7] for ejected electrons of: • 2 eV, ■ 4 eV, A 6eV, + V 8eV, V 10eV, ♦ 20eV and * 40 eV.

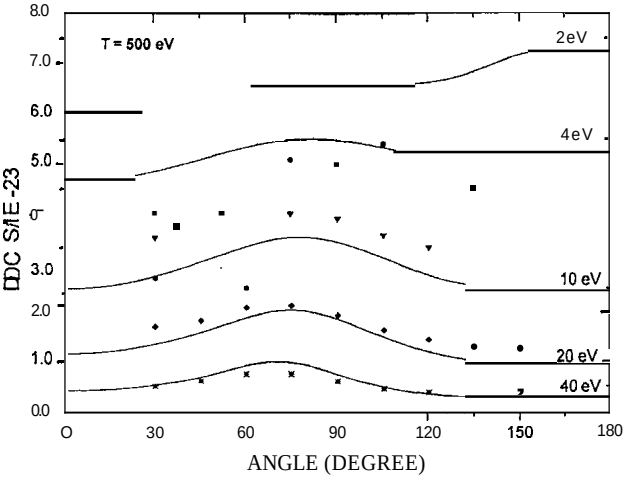

Figure 3: The same as in Fig. 2 for 500-eV incident electrons.

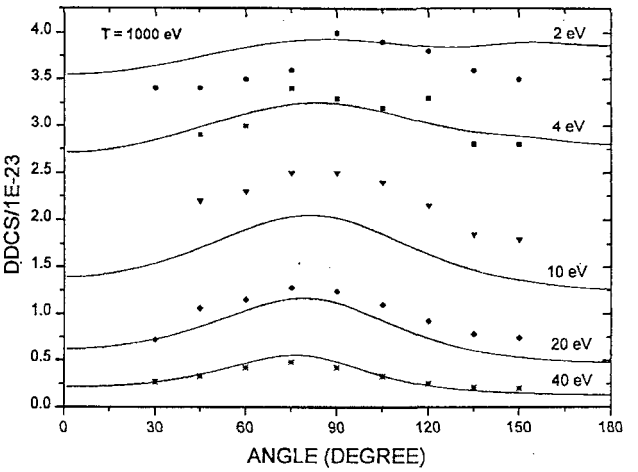

Figure 4: The same as in Fig. 2 for 1000-eV incident electrons.

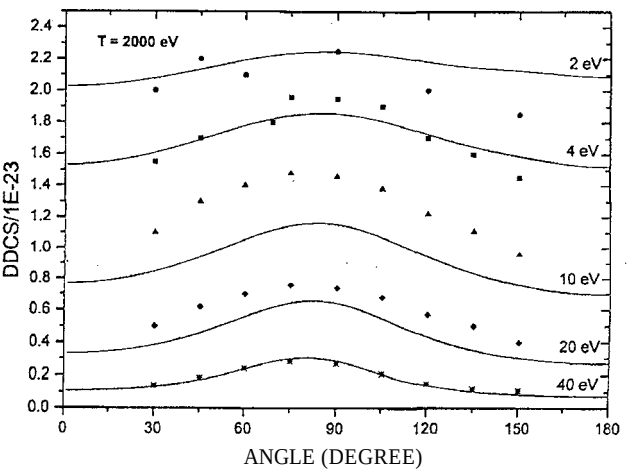

Figure 5: Tlie same as in Fig. 2 for 2000-eV incident electrons.

In Fig. 6, results obtained by use of the semiempirical model are compared to the DDCS experimental data from Opal et al.[2], at 200-eV Incident energies, for ejected energies of 6, 10, and 20 eV. The solid lines results were obtained with the values given in Table 1 for the fitting parameters. The results represented by the dashed lines were calculated using those parameters initially taken into account as discussed in the first paragraph of this section. As it can be seen, the dashed line results seem to provide a better description of the DDCS data of Opal et al.[2]. However, the data from Ref. 2 do not suggest the existence of any minimum in the backward direction in contrast to the measurements by Goruganthu et al.[7]. Both the dashed line and the solid line model results predict the minima structures observed by the latter authors.

In Fig. 7, the DDCS semiempirical results for 1000-eV $e^- + N_2$ collisions are presented as a function of the ejected energy W for scattering angles of 15° and 105°. Similar shapes, as expected for DDCS spectra, have been observed for electron colisions with H_2[9] and H_20 [3,13]. Inner-shell features typical of molecular nitrogen are not W predicted by the semiempirical model. One of the main important features of the K-shell spectra of N_2 is the $^1\sum_g^2 \rightarrow \Pi_{u,g}$, known as the pre ionization line, at 401.1 eV energy loss. Such a transition has been extensively investigated by energy-loss electron impact spectroscopy and photoabsorption techniques[20].

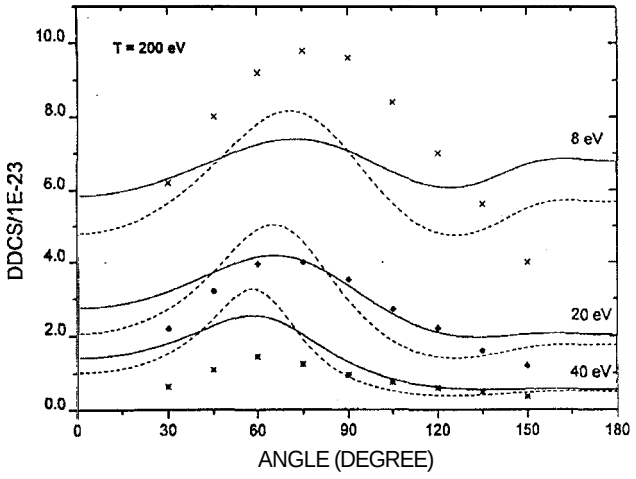

Figure 6: DDCS results for molecular nitrogen at 200-eV incident energy for ejected electrons of 6, 10 and 20 eV. Solid and dashed lines, theory (see text for more details); experimental data are from Opal et al.[2].

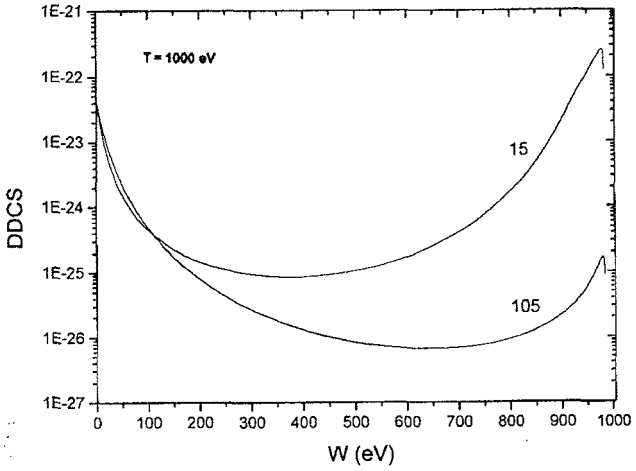

Figure 7: DDCS semiempirical model results for molecular nitrogen at 1000-eV incident energy as a function of the ejected energy, W, for scattering angles of 15° and 105°.

IV. Conclusion

A semiempirical model as proposed by Rudd[8] was used to obtain DDCS data for molecular nitrogen. As far as we línow, this is the first attempt made to employ such a theoretical model in order to describe the DDCS for molecules larger than H_2. The model parameters were adjusted in order to provide the best fit to the experimental results. As discussed previously, the experimental DDCS results of Goruganthu et al.[7] for the nitrogen molecule seem the most reliable ones available in the literature. They have fitted their results to a Legendre polynomial expansion as a function of the angle of ejection. The fitting coefficients, in Ref.7, were adjusted for each incident and ejected energies in the angular region. The model employed in this work has the advantage of reproducing quite well the data by Goruganthu et al.[7] by just employing few parameters. Studies are underway in order to extend the use of the semiempirical model for other molecules, including CO and H_2O, so we could check for any correlation between the model parameters and the masses or total interna1 energies of the molecules.

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