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Energy Spectrum of Electrons Produced by Electron Impact Ionization of Helium, Nitrogen & Oxygen Molecules

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An analytical expression based on Born-Bethe and Møller formulae is proposed for the energy spectrum of secondary electrons produced by the electron impact ionization of atoms and molecules. The results obtained for the ionization of He, N_2 and O_2 are in fair agreement with the experimental data of Opal *et al.* [Opal, C. B., Peterson, W. K. & Beaty, E. C., *J. chem. Phys.*, **55** (1971), 4100] for impact energy $E > 100$ eV and also with the Born calculations performed by employing generalized oscillator strength for $E > 300$ eV.

1. Introduction

DURING an inelastic collision of an energetic electron with an atom or a molecule, ionizing collisions dominate and both slow as well as fast secondary electrons are produced. The fast secondaries produce additional ionization and excitations. The slow secondaries are more effective in producing spin exchange excitations and rotational and vibrational excitations of the molecules. Thus, these secondaries play very important role in many upper atmospheric phenomena and it is of interest to have a knowledge of the energy distribution of the secondary electrons.

Green and Barth[1] proposed a semi-empirical relation for the energy spectrum of the secondary electrons. The investigation was further extended by Green and his associates[2-5] and with the help of the proposed relations a number of upper atmospheric investigations were carried out. In 1969, Khare[6] also proposed two semi-empirical relations for the ionization cross-section per unit energy range; one for the production of slow secondaries and another for the fast secondaries. The above relations were again used to calculate total ionization-cross section[7], fluorescence efficiencies[6,8,9], mean energy loss per ion pair[10,11] and stopping power[11]. In almost all investigations satisfactory agreement with the available experimental data have been obtained. Some other semi-empirical relations for the energy spectrum of the secondary electrons were also proposed by several workers[12-16].

Recently a number of investigators[17-22] have experimentally measured the energy spectrum of the secondary electrons. The most extensive investigation and first of its kind is that of Opal *et al.*[18]. They have reported relative measurement of cross-sections which are differential in energy and angle of the electrons ejected from He, N_2 and O_2 for incident energies between 50 eV and 2 keV. The measurements were integrated over angle and the resulting secondary electron cross-sections were normalized by absolute elastic scattering data to obtain ionization cross-section per unit energy range. If data of Opal *et al.*[18] for 100 and 500 eV in helium are extrapolated towards low energy of ejected electrons, they yield results which are consistent with the trapped electron technique measurements of Grissom *et al.*[19]. Theoretical calculations of Kim and Inokuti[23] also yield about the same values. On the other hand such extrapolation of the data of Crooks and Rudd[21] yields results which are in marked disagreement with the values obtained by Grissom *et al.*[19]. Thus the measurements of Opal *et al.*[18] provide a basis for comparison with theoretical calculations of secondary electron energy distribution. Recently Green and Sawada[24] have fitted the experimental data of Opal *et al.*[18] with an empirical relation having four free parameters. The proposed relation has no resemblance to the various formulae earlier proposed by Green and his associates and, at present, cannot be justified theoretically. Hence, it is of interest to re-examine the theoretical relations proposed by Khare[6] and also combine the two proposed relations into one so that it becomes applicable for all types of secondaries. Furthermore, within the frame work of first Born approximation, the ionization cross-section per unit energy range can be obtained by integrating the generalized oscillator strength. Hence, the experimental data may be compared with such theoretical results. In the present investigation only atmospheric gases namely He, N_2 and O_2 are considered.

2. Theory

In the first Born approximation the double differential cross-section for the ionization is given by

$$\frac{\partial^2\sigma}{\partial W\,\partial K}dK = \frac{4\pi a_0^2\,R^2}{EW}\;\frac{\partial f\,(W,\,K^2)}{\partial W}\;\frac{d\,(K^2)}{K^2}\quad ...(1)$$

where E is the energy of the incident electron which loses an energy W (greater than the ionization potential I of the target) and $\partial f\ (W,K^2)/\partial W$ is the generalized oscillator strength corresponding to a momentum change $\hbar\mathbf{K}$ suffered by the incident electron during the collision, R the Rydberg constant and a_0, the first Bohr radius. On the assumption that due to an energy loss W, a secondary electron of energy $\epsilon = W - I$ is produced, the energy loss cross-section $S\,(E,\,W)$ becomes equal to the ionization cross-section per unit energy range $Q\,(E,\,\epsilon)$ and we have

$$Q(E,\,\epsilon)=S(E,\,W)= \frac{4\pi a_0^2\ R^2}{EW} \times \int_{K^2_{min}}^{K^2_{max}} \frac{1}{K^2}\,\frac{\partial f\,(W,\,K^2)}{\partial W}\,d\,(K^2)\quad ...(2)$$

An examination of the Bethe surface[25] formed by the double differential ionization cross-section shows that two distinct domains are of importance. The first domain, where energy loss W is small or moderate and $(Ka_0)^2$ is small, represents the soft collisions. These collisions largely depend upon the dipole property of the target. It is well known that the dipole property governs the photoabsorption and is sensitive to the electronic structure of the target. As shown by Miller and Platzmann[26], in this domain Eq. (2) reduces to

$$Q_B\,(E,\,\epsilon) = S_B\,(E,\,W) = \frac{4\pi a_0^2\ R^2}{EW}\,\frac{\partial f\,(W,\,O)}{\partial W}\,\ln CE\quad ...(3)$$

where $\partial f\,(W,\,O)/\partial W$ is the optical oscillator strength and C is a constant (see Inokuti[25]). It may be noted that Khare and Padalia[7] have shown that for He, N_2 and O_2, the optical oscillator strength may be represented by the simple formula

$$\frac{\partial f\,(W,\,O)}{\partial W} = A\,\exp\,(-BW)\quad ...(4a)$$

However, for oxygen molecule when W is less than 16 eV, $\partial f\,(W,\,O)/\partial W$ were represented by

$$\frac{\partial f\,(W,\,O)}{\partial W} = mW+k\quad ...(4b)$$

where A, B, m and k are constants and have been obtained by Khare and Padalia[7] fitting theoretical values of $\partial f/\partial W$ for He calculated by Bell and Kingston[27] and extrapolated experimental values of $\partial f\,(W,\,O)/\partial W$ for N_2 and O_2 given by Silvermann and Lassettre[28].

In the second domain where both W and $(K\,a_0)^2$ are large, the essential features of the Bethe surface is common to all atoms and molecules. In this domain W is much larger than the nucleus-electron binding energy and hence the collision may be treated as the collision between two free electrons. Moller[29] has studied quantum mechanically such a problem and obtained for non-relativistic energies.

$$Q_M\,(E,\,\epsilon)=S_M\,(E,\,W) = \frac{4\pi a_0^2\ R^2}{E\,\epsilon^2}\left[1-\frac{\epsilon(E-2\epsilon)}{(E-\epsilon)^2}\right]\quad ...(5)$$

Khare considered only the first term of Eq. (5) and multiplied it by the number of ionizable electrons (S) to obtain

$$Q\,(E,\,\epsilon) = S\,(E,\,W) = \frac{4\pi a_0^2\ R^2}{E}\,\frac{S}{\epsilon^2}\quad ...(6)$$

It may be noted that Eq. (6) with $S=1$ is identical with the leading term of the expression obtained by Omidvar[30] for the production of fast secondaries in the ionizing collision of electrons by hydrogen-like atom. It may further be noted that both Eqs. (3) and (6) are applicable for high values of E and were extended semi-empirically for low values of E by Khare[6].

Before proceeding further we compare Eqs. (3) and (5) with experimental data. We consider the simplest target namely helium. Fig. 1 shows the plots of Q_B and SQ_M for $E=500$ eV along with the experimental data of Opal *et al.*[18] and Gissom *et al.*[19]. S is equal to 2 and Eq. (4) is utilized for evaluating $\partial f\,(W,\,O)/\partial W$ with the values of A and B as given by Khare and Padalia[7]. The value of C is taken to be 0·3165 eV^{-1} (Inokuti and Kim[31]). It is evident from Fig. 1 that Q_B agrees with experimental results

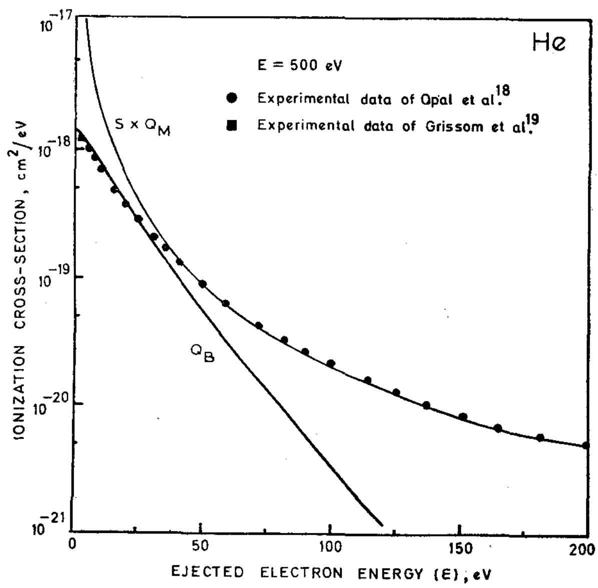

Fig. 1—Plots of cross-section $Q(E,\,\epsilon)$ for He at E=500 eV. [Q_B is obtained from Eq. (3) and SQ_M from Eq. (5) with S=2 ● and ▲ are the experimental data of Opal *et al*[18]. and Girssom *et al.*[19], respectively.]

for low values of ϵ whereas SQ_M is in good agreement with the data for large values of ϵ ($>$ 40 eV). Thus it confirms the assumption made by Khare[6]. However, a more complex quantum mechanical treatment is required to cover the whole energy region of ϵ. No satisfactory theoretical investigation, particularly for molecules, has yet been carried out. Hence, considering the applications of $Q(E, \epsilon)$ to various investigations, it is highly desirable to combine Eqs. (3) and (5) in such a way that they explain the experimental results for all values of ϵ and E. Accordingly we take

$$Q(E, \epsilon) = S(E, W) = f_1(E, \epsilon)\, Q_B(E, \epsilon) + f_2(E, \epsilon)\, S\, Q_M(E, \epsilon) \qquad \ldots(7)$$

with

$$f_1(E, \epsilon) = \frac{1}{(1+I/E)^2}\left[1 + \frac{\epsilon}{E-I}\right] \frac{\ln[1+C(E-I)]}{\ln CE} \qquad \ldots(8)$$

$$f_2(E, \epsilon) = \frac{\epsilon^3}{\epsilon^3+\epsilon_0^3}\left[1 - \frac{I}{E}\right] \qquad \ldots(9)$$

It may be noted that $f_1(E,\epsilon)$ and $f_2(E,\epsilon)$ are arbitrary factors which are introduced to give best visual agreement with the experimental data of Opal *et al.*[18]. Such a consideration yields ϵ_0 equal to 50 eV, a value which is consistent with Fig. 1. It is easy to verify that Eq. (7) reduces to Q_B or $S\,Q_M$ in the required limits and tends to zero as E approaches I.

Eq. (2) may also be employed to obtain $Q(E\ \epsilon)$ provided $\partial f(W, K^2)/\partial W$ is known. Bell and Kingston[32] and Manson[33] have calculated $\partial f(W, K^2)/\partial W$ in the continuum of helium. A more elaborate calculation has been carried out by Oldham[34]. For high values of $(Ka_0)^2$ and W, the following approximate formula for $\partial f(W, K^2)/\partial W$ (Mott and Massay[35]) may be employed

$$\frac{\partial f(W, K^2)}{\partial W} = \frac{2^9 K_w^2 \mu^6 \{K^2 + \frac{1}{3}(\mu^2+k^2)\} \exp\left\{-\frac{2\mu}{k}\tan^{-1}\frac{2\mu K}{K^2+\mu^2-k^2}\right\}}{a_0^2 \{\mu^4 + 2\mu^2(K^2+k^2) + (K^2-k^2)^2\}^3 \{1-\exp(-2\pi\mu/k)\}} \qquad \ldots(10)$$

where

$$W = \hbar^2 K_w^2/2m \qquad \ldots(11)$$

$$\epsilon = \hbar^2 k^2/2m \qquad \ldots(12)$$

and $\mu = Z/a_0$, Z for helium being taken to be (27/16), and m is the mass of the electron.

For N_2 and O_2, no such theoretical calculations are available. However, $\partial f(W, K^2)/\partial W$ as a function of (Ka_0) have been measured by Silvermann and Lassettre[28] and Tisone[36] for some discrete values of the energy loss.

As mentioned earlier Green and Sawada[24] have given analytical expression for calculating $Q(E, \epsilon)$. According to them

$$Q(E, \epsilon) = \sigma_0 \frac{K}{E} \ln\left(\frac{E}{J}\right) \times \frac{\Gamma_s E^2}{(E+I)^2\left(\epsilon - T_s + \frac{1000}{E+2I}\right)^2 + \Gamma_s^2 E^2} \qquad \ldots(13)$$

where σ_0 is equal to 10^{-16} cm^2. The values of the parameters Γ_s, T_s, K and J tabulated by Green and Sawada[24] are different for different targets and are obtained by fitting Eq. (13) with the experimenral results of Opal *et al.*[18]. The values of $Q(E, \epsilon)$ have also been obtained from Eq. (13) for the sake of comparison.

3. Calculations, Results and Discussion

Eq. (7) has been employed to calculate $Q(E, \epsilon)$ for He, N_2 and O_2 in the incident energy range 50-2000 eV. As mentioned earlier Eqs. (3) and (4) are utilized to obtaine Q_B. The constants A and B for helium atom are taken to be same as listed by Khare and Padalia[7]. However, the value of C is taken to be 0·3165 eV^{-1} (Inokuti and Kim[31]). For N_2 and O_2 molecules it may be noted that the recent investigations of Skerbele and Lassettre[37] and Tisone[36] suggest that the values of the generalized oscillator strength (GOS) obtained by Silvermann and Lassettre[28] are too large due to streaming error in McLeod gauge and should be multiplied by 0·754. This fact is further corroborated by El-Sherbine and Van Der Wiel[38] who have measured $\partial f/\partial W$ for N_2 and CO. Their values of the oscillator strengths for ϵ greater than 10 eV are also about 0·754 times the values reported by Silvermann and Lessettre[28]. However, for $\epsilon < 10$ eV they are lower than the corrected values of Silvermann and Lessettre and thus they would further increase the difference between theory and experiment if used in the calculation of $Q(E, \epsilon)$ (see Figs. 2 and 3). Hence, while using Eq. (4) for N_2 and O_2 we have carried out two sets of calculations, one with the original values of A, B, m and k as listed by Khare and Padalia[7] and other with 0·754 times the values of the above constants. The values of B and C are taken to be same. The value of Q_M is obtained from Eq. (5). Finally, $f_1(E, \epsilon)$ and $f_2(E, \epsilon)$ are calculated from Eqs. (8) and (9). For helium atom, the value of S is 2 and for N_2 and O_2 the values of S are taken to be 10 and 12 respectively. The values of $Q(E, \epsilon)$ so obtained are shown by curves B and A in Figs. 2-7.

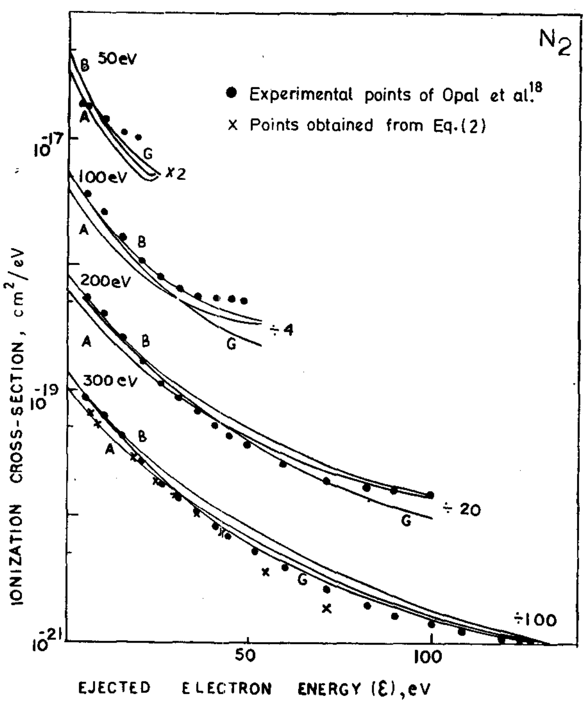

Fig. 2—Plots of cross-section $Q(E, \epsilon)$ for electron impact ionization of N_2 molecules for different values of E between 50 and 300 eV [Curve: B, obtained from Eq. (7) with original $\partial f/\partial W$ values of Silvermann and Lessettre[28]; A, obtained from Eq. (7) with corrected $\partial f/\partial W$ values of Silvermann and Lessettre[28]]

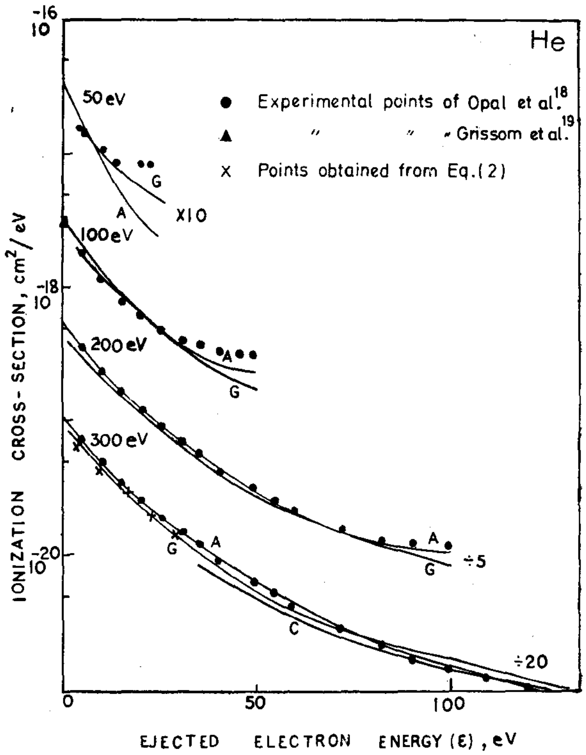

Fig. 4—Cross-section $Q(E, \epsilon)$ for electron impact ionization of He atoms for the values of E lying in the range 50-300 eV [Curve: A, obtained from Eq. (7); G, from Eq. (13); C, from Eq. (2)]

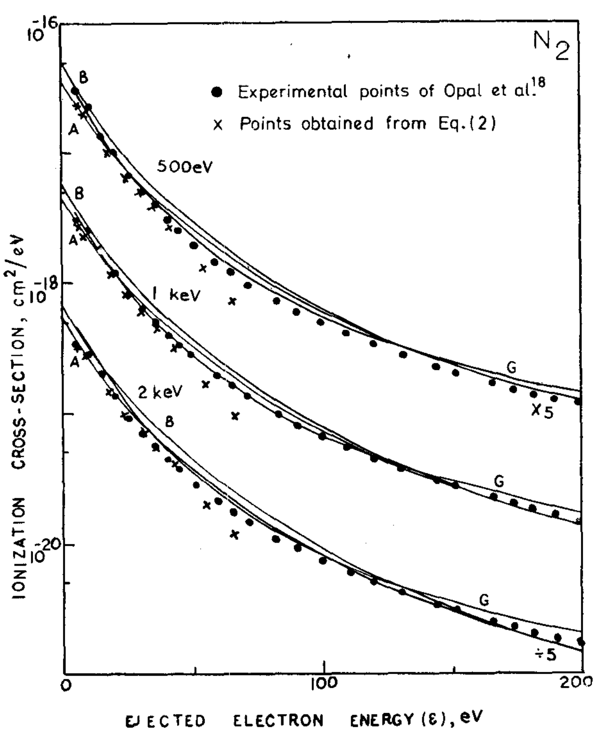

Fig. 3—Same as Fig. 2 but values of E lie in the range 500 eV – 2 keV

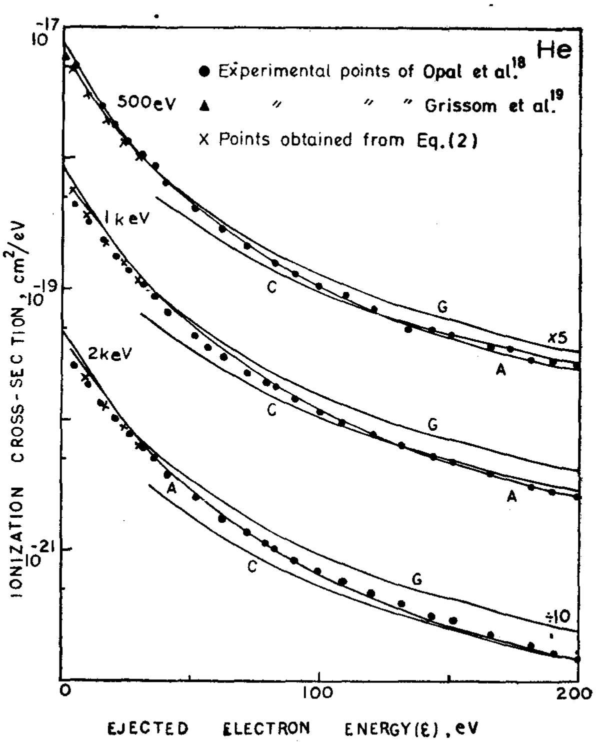

Fig. 5—Same as Fig. 4 but values of E lie in the range 500 eV – 2keV

To perform Born calculations, Eq. (2) is utilized along with the assumption that GOS is independent of incident energy. However, recently Oda and Nishimura[22] have observed that even at $E=500$ eV and 1 keV this is not so. For helium we have employed theoretical values of GOS calculated by Oldham[34] at $W=28{\cdot}5$, 34·5, 41·8, 48·1 and 54·3 eV.

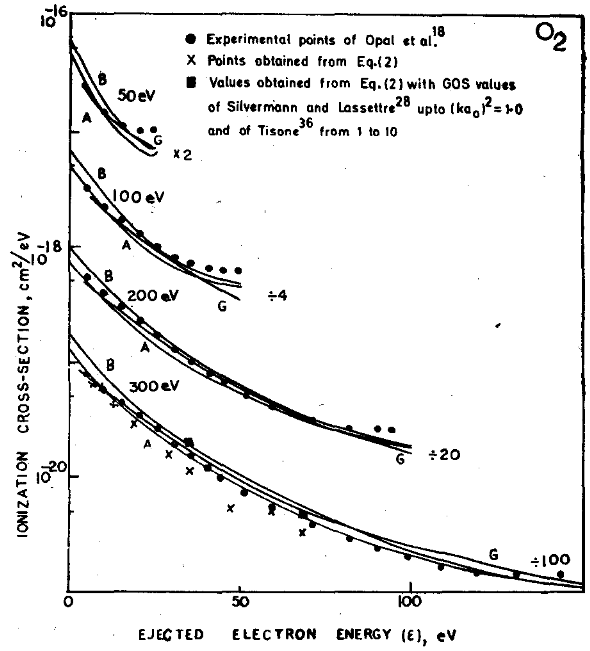

Fig. 6—Cross-section $Q(E, \epsilon)$ for electron impact ionization of O_2 molecules for the values of E lying in the range 50-300 eV [See caption of Fig. 2 for other details]

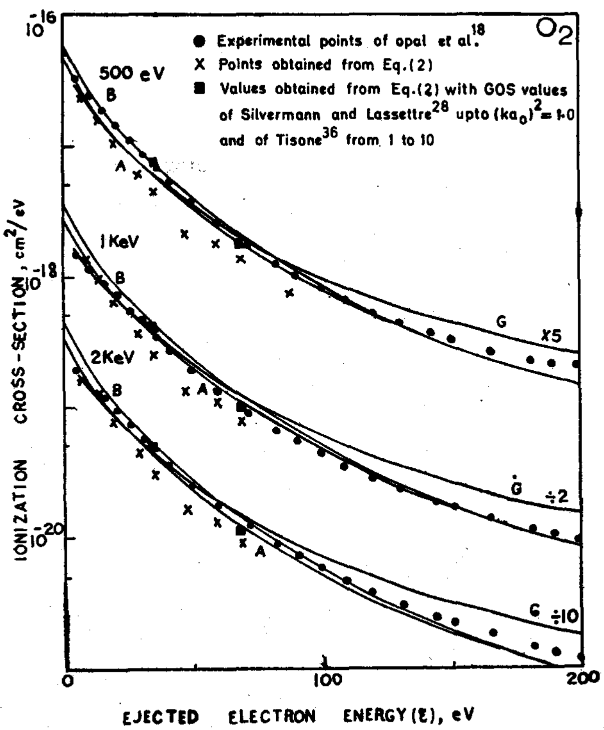

Fig. 7—Same as Fig. 6 but values of E lie in the range 500 eV—2keV

In Oldham's calculations the highest value of $(Ka_0)^2$ is 3·0. Whenever the values of GOS for $(Ka_0)^2 > 3{\cdot}0$ are required, Eq. (10) is used. The values of $Q(E, \epsilon)$ so obtained are shown by the crosses in Figs. 4 and 5. For $W \geqslant 60$ eV, Eq. (10) is employed for all values of $(Ka_0)^2$ and the values of $Q(E, \epsilon)$ are shown by the curves C. For molecular targets, experimental data of GOS obtained by Silvermann and Lassettre[28] and corrected by Skerbele and Lassettre[37] are utilized in Eq. (2) to obtain $Q(E, \epsilon)$. For molecular nitrogen they have measured GOS for $W = 22{\cdot}73$, 24·53, 34·16, 40·63, 46·2, 51·46, 58·23, 70·41 and 82·17 eV upto $(Ka_0)^2=2{\cdot}0$. The results of $Q(E, \epsilon)$ using corrected GOS are shown in Figs. 2 and 3 by crosses. For molecular oxygen, Silvermann and Lassettre[28] have measured GOS for $W=19{\cdot}7$, 21·7, 26·2, 32·2, 41·5, 47·8, 59·7, upto $(Ka_0)^2=1{\cdot}0$ and for 71·6 and 80·8 eV energy losses the maximum value of $(Ka_0)^2$ is 4·0. Results of $Q(E, \epsilon)$ obtained by using corrected values of these GOS are shown by crosses in Figs. 6 and 7. Recently, Tisone[36] has measured GOS for $W=30$, 50, 80 and 100 eV from $(Ka_0)^2=1$ to 10 with $E=500$ eV and utilized them alongwith the corrected GOS of Silvermann and Lassettre[28] to obtain $S(E, W)$. Thus, the upper limit of integration of Eq. (2) has been increased to 10. Similar calculations have been carried out by us at other impact energies, i. e. 300, 1000, 2000 eV for $W=47{\cdot}8$ and 80·8 eV. The results so obtained are shown by rectangles in Figs. 6 and 7. The values obtained by Eq. (13), given by Green and Sawada[24] are represented by the curves G.

Let us consider the ionization of helium atom. Figs. 4 and 5 show that Eq. (7) yields results which are in good agreement with the experimental data for all impact energies except at $E=50$ eV. In case of ionization of nitrogen molecule, Figs. 2 and 3 show that Eq. (7) with corrected values of $\partial f/\partial W$ underestimates $Q(E, \epsilon)$ at low values of ϵ and overestimates by approximately same amount at intermediate values of ϵ for impact energies less than or equal to 500 eV. Here again at $E=50$ eV agreement is not satisfactory but at 1 and 2 keV, the theoretical values are in fair accord with the experimental data. When original values of GOS, as given by Silvermann and Lessettre[28] are used, curve B shows that the agreement for low ϵ improves but the disagreement at intermediate values of ϵ increases. Figs. 6 and 7 show that curve A for oxygen molecule is in satisfactory agreement at all energies. Curve B seems to overestimate the cross-section at low values of ϵ at most of the impact energies considered. However, in most of regions covered, the difference between the experimental data and the theoretical results is about 25%

Table 1—Comparison of the values of $R=Q(E, \epsilon)/Q(E, \epsilon = 13{\cdot}8$ eV) for different values of E and ϵ in the electron impact ionization of He

E (eV) ↓	ϵ (eV)→	25	37·5	87·5	137·5	200
	a	0·514	0·291			
100	b	0·499	0·303			
	c	0·539	0·370			
	a	0·509	0·282	0·0660		
200	b	0·510	0·287	0·0702		
	c	0·506	0·277	0·0644		
	a	0·506	0·279	0·0641	0·0274	
300	b	0·510	0·289	0·0562	0·0259	
	c	0·505	0·281	0·0483	0·0239	
	a	0·506	0·276	0·0625	0·0265	0·01280
500	b	0·509	0·281	0·0533	0·0191	0·00924
	c	0·501	0·288	0·0517	0·0194	0·00920
	a	0·506	0·275	0·0613	0·0259	0·0125
1000	b	0·506	0·275	0·0503	0·0181	0·00801
	c	0·526	0·301	0·0560	0·0212	0·00877
	a	0·506	0·274	0·0606	0·0255	0·0123
2000	b	0·503	0·269	0·0471	0·0170	0·00772
	c	0·545	0·315	0·0621	0·0207	0·0100

a. Obtained from Eq. (13); b, Present results obtained from Eq. (7); c, Exptl. results of Peterson *et al.*[17].

which is about the same as the uncertainty in the experimental data. It may be noted that the secondary electron energy distribution of Opal *et al.*[18] extend down to about 5 eV. Recently, Grissom *et al.*[19] used a trapped electron technique to obtain cross-sections for the production of secondary electrons in the energy range 0-1 eV for the ionization of helium. The present calculations are in good agreement with their measurements performed at $E=100$ and 500 eV (see Figs. 4 and 5).

An examination of the Born calculations shows that for helium when GOS of Oldham alongwith Eq. (10) are utilized the results are in satisfactory agreement with the experimental data. However, the calculations carried out with Eq. (10) for the range of W greater than 60 but less than 100 eV yield lower results. The disagreement increases with incident energy. This indicates that for helium, Eq. (10) is not a useful representation of the GOS for small values of momentum transfer. The defect may lie in the non-orthogonality of the employed wave functions. For molecular targets it may be noted that for large W, the values of $Q(E, \epsilon)$ obtained from Eq. (2), with corrected data of Silvermann and Lassettre[28] for GOS, are lower than the experimental data. The agreement improves when GOS for higher values of $(Ka_0)^2$, given by Tisone[36], are included. This is as expected because for large W (> 40 eV) appreciable contribution comes from the higher values of $(Ka_0)^2$. We also notice that Eq. (13) of Green and Sawada[24] yields results which are in satisfactory agreement with the experimental data for impact energies less than 300 eV. However, for helium atom and oxygen molecule the results obtained from Eq. (13) for $E \geqslant 500$ eV are much greater than the experimental data for the production of fast secondaries. Moreover, it contains 4 free parameters whose values are different for different targets and are determined by fitting the experimental data. Hence, it may be difficult to apply it for new targets. On the other hand in Eq. (7), we require values of optical oscillator strength, which are available for large number of targets from the photoabsorption data. The constant C can either be theoretically calculated or obtained from the total ionization cross-section data (Inokuti[25]).

Peterson *et al.*[17] have measured the ratio $R=Q(E, \epsilon)/Q(E, \epsilon=13{\cdot}8)$ for electron impact ionization of helium. In Table 1, we compare this ratio obtained in the present calculations with the experimental data and also with the values obtained from Eq. (13). The present values are in satisfactory agreement with experimental values at all values of E and ϵ. The maximum difference is about 25% at $\epsilon=87{\cdot}5$ eV and $E=2$ keV. Eq. (13) also yields more or less similar agreement.

Finally, we conclude that the analytical expression (7) for $Q(E\ \epsilon)$ yields satisfactory results and should be useful for upper atmospheric investigations. Our preliminary results for total ionizotion cross-section obtained by integrating Eq. (7) are in good agreement with the experimental data. Further investigations are in progress.

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