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ENERGY AND ANGULAR DISTRIBUTION OF ELECTRONS RESULTING FROM DIRECT IONIZING COLLISIONS OF ELECTRONS WITH ATOMS

BY

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ABSTRACT

The energy and angular distributions of secondary electrons resulting from direct ionizing collisions of electrons with helium at the primary energy $E_0=500$ eV and with krypton at $E_0=1$ keV were measured. The total spectra, consisting of the continuum spectra due to direct ionization and the sharp line spectra due to indirect ionization (autoionization or Auger process), were measured with an energy resolution of about 0.2 eV in FWHM over the angular range from 10° to 130° and the continuum spectra were derived from the total spectra for electron energies from 25 eV to 45 eV for helium and from 24 eV to 52 eV for krypton.

The double differential cross sections for ionizing collisions of electrons with helium and krypton were calculated utilizing the binary encounter theory. The identity between scattered and ejected electrons as well as the exchange effect between incident and atomic electrons were taken into consideration. The results of calculations were compared with several experimental results. The general shapes of double differential cross sections measured were well reproduced in the significantly wide angular and energy ranges by the binary encounter theory, although discrepancies between theory and experiment exist in the forward and backward directions. The forward rise of double differential cross sections observed in most of experiments may be mainly attributed to the exchange term for the ejected electrons.

1. Introduction

In a previous paper,[1] the autoionization processes in helium were investigated by measuring the energy spectra of secondary electrons, that is electrons resulting from ionizing collisions of electrons with helium atoms. While the knowledges on the indirect ionization process, such as autoionization and Auger process, are obtained from the discrete part of the energy spectra, the continuum part gives the information on the direct ionization. The study on the energy and angular distribution of secondary electrons in the direct ionization is important from the following points of view: (1) The measurement of double differential cross sections,* which are differential in both energy and detection angle of secondary electrons, is a far more sensitive means of testing the theory about the ionization processes than the measurement of total ionization cross sections. (2) The *ddcs* about the secondary electrons is useful in understanding the phenomena in many fields of physics, *e.g.* energy deposition of high energy radiation, upperatmospheric phenomena, etc. (3) The line shapes

* Hereafter, abbreviated to "*ddcs*".

of energy spectra due to electrons ejected from autoionization states in helium undergoes a marked change as the detection angle is varied.[1] According to the extended understanding of Fano's theory in reference (1), the angular variation of shape parameter, q, is mainly ascribed to the variation of *ddcs* for the direct ionization with detection angles (see eq. (10) in reference (1)). Therefore, the *ddcs* for the direct ionization in the energy region relating to autoionization is necessary for the understanding of autoionization process.

During the 1930's several groups measured the energy and angular distributions of secondary electrons.[2,3] Since, while some interesting features have been found in these studies, most of them were subject to several defects on the experimental conditions such as the elimination of the stray magnetic field, the purity of target gases, the effect of the condition of metal surface, etc., the renewed measurement of secondary electrons with lower energies, especially less than 50–eV, has been requested for a long time with more refined techniques.

The *ddcs* of the electron ejection from atoms by proton and other heavy charged particle impact has been recently measured by Rudd *et al.*[4] and Toburen,[5] and compared with the theoretical *ddcs* calculated by the Born approximation[6] and the binary enounter theory.[7] The recent investigation on the energy and angular distribution of secondary electrons by electron impact is rather few. The angular distribution of secondary electrons resulting from the direct ionization in helium was measured by Oda, Nishimura and Tahira[8] in the energy regions relating to autoionization. Their measurement was performed with energy resolution good enough to discriminate the electrons ejected indirectly from those ejected or scattered directly. Ehrhardt *et al.*[10] measured the *ddcs* in electron impact ionization of helium and argon, showing that for high incident energy the outgoing electrons belong to one of the two energetically well separated groups; either the fast electrons observed mainly in forward direction or the slow electrons distributed isotropically in all angles. Opal *et al.*[9] also measured the *ddcs* for secondary electrons with energies lower than half the primary energies.

The scattered and ejected electrons can be treated separately from the theoretical point of view. Massey and Mohr[11,12] calculated the energy and angular distribution of electrosn ejected and scattered from hydrogen atoms by electron impact, utilizing the Born approximation. Although the *ddcs* for proton impact has been calcualted by several authors recently and detail comparison with the experimental *ddcs* has shown several interesting features, there has been no calculation for electron impact which can be compared with the recent measurements. Moreover, the exchange effect, which has been often considered in the calculation of total ionization cross sections by electron impact, has been not yet taken into consideration in the calculation of the *ddcs* for electron impact. The angular distribution of electrons ejected from helium atoms and hydrogen molecules by proton impact were calculated by Bonsen and Vriens,[7] utilizing the binary encounter theory, which shows that the experimental results are well described by this theory in significant ranges of ejection angles and energies, although the discrepancies exist in the forward and backward directions. The angular distribution of fast charged particles scattered from hydrogen atoms has been calculated by the binary encounter theory as well as the Born approximation, where the binary encounter theory has given results almost similar to those by the Born approximation for the scattering with the large energy loss and large momentum transfer. These results suggest that the binary encounter theory may give a fairly good description on the energy and angular distribution of the secondary electrons within a certain range of validity. It is important to investigate in more details on the range of validity of the binary encounter theory in several systems, because the calculations by this theory can be performed more easily than those by the Born approximation, especially for atoms and molecules more complicated than hydrogen atom.

2. Experiment

The angular distributions of secondary electrons resulting from the direct ionization were measured in the energy region involving the discrete spectra due to the indirect ionization. The experimental apparatus is the same as that mentioned in a previous paper.[1] Typical examples of energy spectra of electrons resulting from ionizing collisions of electrons with helium and krypton are shown in Figs. 1 and 2, respectively. The sharp line spectra shown in these figures correspond to electrons ejected from autoionization states for helium[1] and to M-NN Auger electrons for krypton. It is noted that a large portion of energy spectra in this energy region is occupied by those due to the indirect processes. Therefore, the energy resolution should be good enough to discriminate line spectra embedded in the ionization continuum from the total spectra, as in the present study (0.2–eV in FWHM). The intensities of continuum parts at several electron energies were normalized taking into consideration the beam current of primary electrons, the target gas pressure, and the collision volume at each angle. The overall uncertainty is estimated to be about 20%.

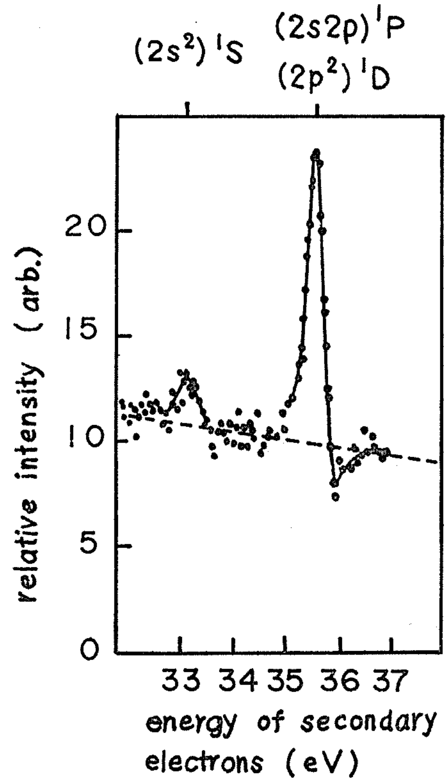
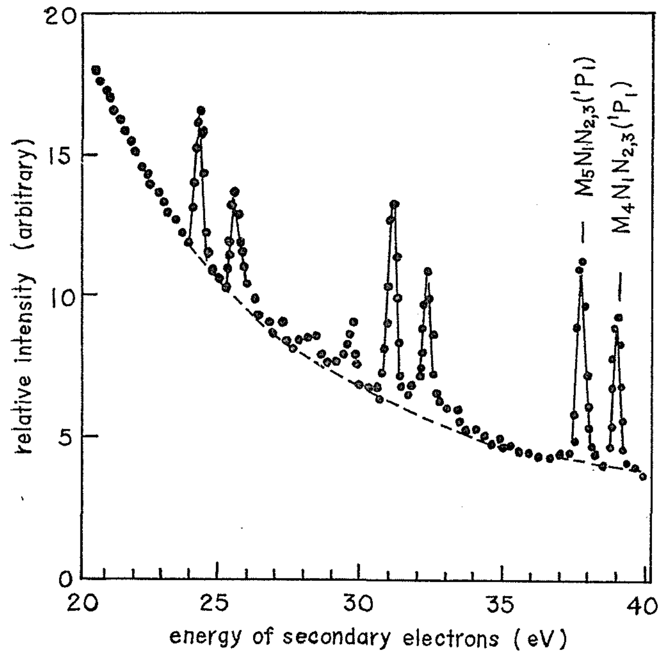


Fig. 1. Energy spectrum of electrons resulting from ionizing collisions of electrons with helium. Primary electron energy is 500–eV and detection angle is 107°. The broken line drawn in this figure shows the continuous part of energy spectrum, while the sharp line structures are due to electrons ejected from autoionization states in helium.

a)



b)

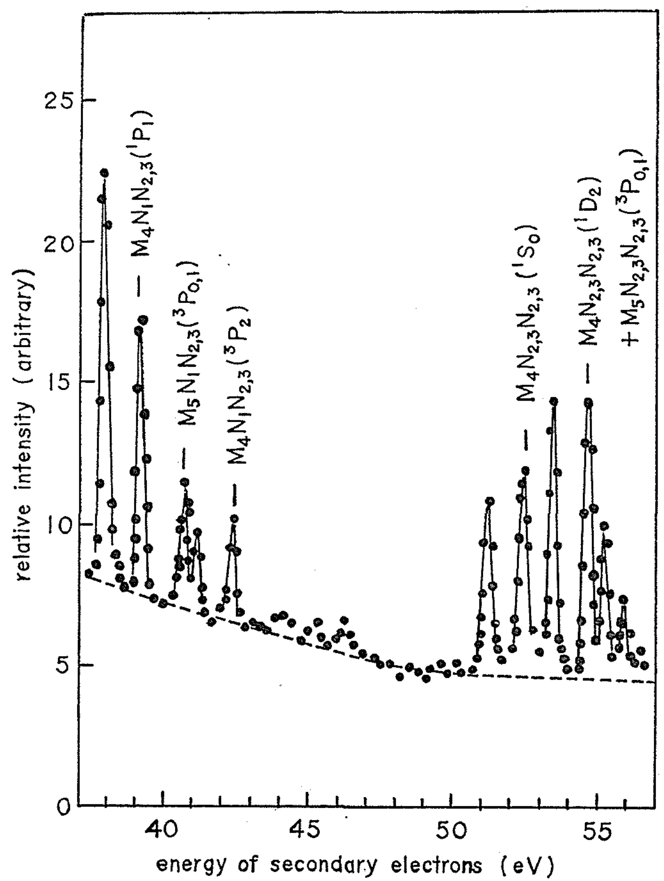


Fig. 2. Energy spectrum of electrons resulting from ionizing collisions of electrons with krypton. Primary electron energy is 1 keV and detection angle is 87°. The broken lines drawn in figures show the continuous part of energy spectrum, while sharp line structures are due to MNN Auger electrons ejected from krypton. The energy ranges of secondary electrons; (a) 20 eV～40 eV and (b) 37 eV～57 eV.

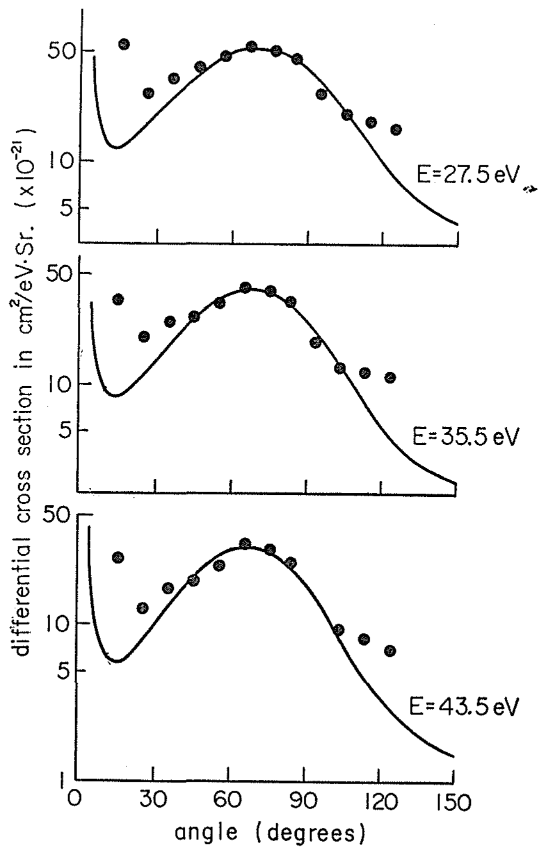


Fig. 3. Angular distribution of secondary electrons resulting from ionizing collisions of electrons with helium. Primary electron energy is 500 eV. The energies of secondary electrons, E, are shown on curves. Closed circles: measured double differential cross sections; Full curves: ones calculated by the binary encounter theory.

In Figs. 3 and 4 are shown the angular distributions of electrons resulting from the ionizing collisions of 500–eV electrons with helium and of 1 keV electrons with krypton respectively, where the measured angular distributions were normalized so that the peak height of the broad maxima measured coincide with those theoretically calculated with the binary encounter theory. All of these measurements show the existence of the broad maxima in the intermediate angular regions. Besides, the steep rises in the forward direction are shown, which have not been observed by Opal, *et al.*[9]

3. Calculations of ddcs by the binary encounter theory

3–1 Formulation

In the binary encounter theory (B.E.T.), an incident electron is supposed to interact with only one of atomic electrons at a time and the cross sections for the electron-atom collisions are obtained by integrating the cross sections for the binary encounter collisions between incident and atomic electrons over the velocity distribution of the atomic electrons. The double differential cross section for observed electrons is the sum of both that for ejected

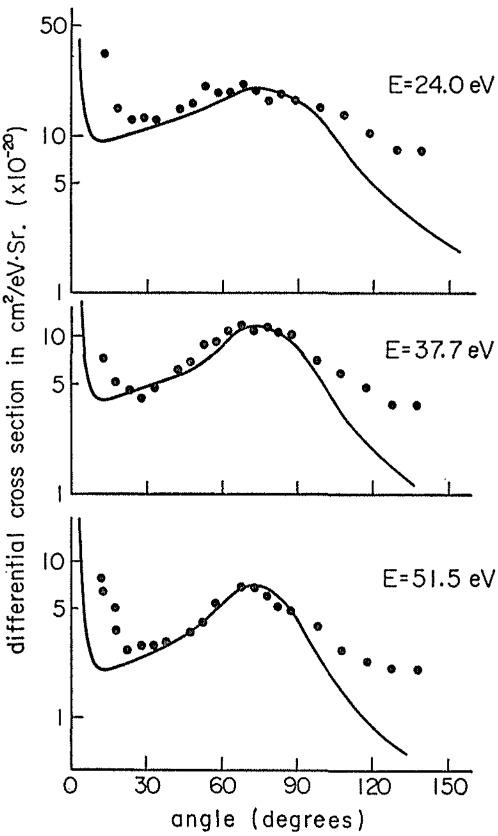


Fig. 4. As in Fig. 3, except that the primary electron energy is 1 keV and the target atoms are krypton.

electrons and that for scattered electrons. The double differential cross section for ejected electrons is given by

$$\frac{d^2\sigma_e}{dE_e d\Omega_e}=N\int_{v_{2min}}^{v_{2max}}\{\sigma_D{}^e(v_2)+\sigma_{EX}{}^e(v_2)-\frac{1}{2}\sigma_I{}^e(v_2)\}f(v_2)dv_2, \tag{1}$$

where $\sigma_D{}^e(v_2)$, $\sigma_{EX}{}^e(v_2)$, and $\sigma_I{}^e(v_2)$ are the direct, exchange, and interference term respectively, E_e is the energy of ejected electrons, $d\Omega_e$ is unit solid angle in the direction of ejected electrons, N is the number of electrons in an atom, $f(v_2)$ is the velocity distribution of atomic electrons, and v_{2min} and v_{2max} are the lower and the upper bounds for the velocity of atomic electrons contributing to the differential cross section, respectively[7]. The expressions for $\sigma_D{}^e(v_2)$, $\sigma_{EX}{}^e(v_2)$, and $\sigma_I{}^e(v_2)$ are given as follows:

$$\sigma_D{}^e(v_2)=\frac{m^2e^4v_1v_2'}{2v_2E^3S^3}\{mv_2'^2\sin^2\theta_e-ES^2/(mv_1)^2\}\ , \tag{2}$$

$$\sigma_{EX}{}^e(v_2)=\frac{2m^2e^4v_2'}{v_1v_2}\frac{1}{S^5}\ , \tag{3}$$

and

$$\sigma_I{}^e(v_2)=\frac{2me^4v_2'}{v_1v_2E}\frac{\Phi}{S^3}\ , \tag{4}$$

where v_1 is the velocity of incident of electrons, v_2 is the velocity of atomic electrons, v_2' is the velocity of ejected electrons, θ_e is the ejection angle with respect to the direction of incident electrons, m and e are the mass and the charge of the electron, respectively, E is the direct energy transfer, S is the magnitude of the exchange momentum transfer, S, and Φ is the phase factor which was taken to be unity in this calculation.

The expression for E, $\mathbf{S}$, and S are given as follows:

$$E=\frac{1}{2}mv_1^2-\frac{1}{2}mv_1'^2=\frac{1}{2}mv_2'^2-\frac{1}{2}mv_2^2\,, \tag{5}$$

$$\mathbf{S}=m(\mathbf{v}_1-\mathbf{v}_2')\,, \tag{6}$$

and

$$S=m(v_1^2+v_2'^2-2v_1v_2'\cos\theta_e)^{1/2}\,, \tag{7}$$

where v_1' is the velocity of scattered electrons. The observed kinetic energy of ejected electrons, E_e, is

$$E_e=E-U\,, \tag{8}$$

where U is the binding energy of electrons to be ejected.

While the expression for $\sigma_D{}^e(v_2)$ can be derived from that given by Bonsen and Vriens[7] for the proton impact, those for $\sigma_{EX}{}^e(v_2)$ and $\sigma_I{}^e(v_2)$ were newly derived (See Appendix).

The double differential cross section for scattered electrons was before derived by vriens[14] as follows:

$$\frac{d^2\sigma_S}{dE_S d\Omega_S}=N\int_{v_{2min}}^{\infty}\{\sigma_D{}^S(v_2)+\sigma_{EX}{}^S(v_2)-\frac{1}{2}\sigma_I{}^S(v_2)\}f(v_2)dv_2\,, \tag{9}$$

$$\sigma_D{}^S(v_2)=\frac{2m^2e^4v_1'}{v_1P^5}\frac{1}{v_2}\,, \tag{10}$$

$$\sigma_{EX}{}^S(v_2)=\frac{2e^4v_1v_1'}{{}^2mP|v_1^2+\frac{2}{m}(U-E)|^3}\;\frac{v_1^2+2v_2^2+\frac{2}{m}U-\frac{P^2}{2m^2}-\frac{2E^2}{P^2}}{(v_1^2+v_2^2+\frac{2}{m}U)v_2}\,, \tag{11}$$

$$\sigma_I{}^S(v_2)=\frac{4e^4v_1v_1'}{P^3|v_1^2+\frac{2}{m}(U-E)|}\;\frac{1}{(v_1^2+v_2^2+\frac{2}{m}U)v_2}\,, \tag{12}$$

and

$$P=m(v_1^2+v_1'^2-2v_1v_1'\cos\theta_S)^{1/2} \tag{13}$$

where $\sigma_D{}^S(v_2)$, $\sigma_{EX}{}^S(v_2)$, and $\sigma_I{}^S(v_2)$ are the direct, exchange, and interference terms respectively, E_S is the energy of scattered electrons, P is the direct momentum transfer, and $d\Omega_S$ is the unit solid angle in the direction of scattered electrons, θ_S. The symmetrical collision model[15] was applied to the derivation of Eqs. (11) and (12). The sum of the differential cross sections $d\sigma_e{}^2/dE_e d\Omega_e$ and $d^2\sigma_S/dE_S d\Omega_S$ was calculated and compared with the experiments.

3–2 Results of Calculation and Discussion

When the formulae developed in 3–1 are applied to the actual collision processes, several points must be taken into consideration. First, the exact form of velocity distribution of atomic electrons, $f(v_2)$, is known only for hydrogen atom and the approximate forms must be used for the other atoms and molecules. Second, the contribution from atomic electrons in each subshell must be added to obtain the overall cross section for multishell atoms, *e.g*, krypton. In this paper all of these points are not necessarily taken into consideration, because the calculation described here is only the first step towards more elaborate calculation.

The hydrogen velocity distributions were assumed for atomic electrons in helium and krypton. The effective nuclear charge in helium was calculated from the average kinetic energy of atomic electrons, 39.49 eV, which was derived using the virial theorem. For krypton, only the electrons in the outermost shells, i.e. 4s and 4p shells, have been taken into consideration and the velocity distributions have been derived from the hydrogenic wave function[16] with the effective nuclear charge calculated according to Slater's rule.[17] The hydrogenic velocity distribution for helium may be a fairly good approximation, because the more elaborate velocity distribution used for the proton impact[7] gave the results almost similar to those obtained utilizing the hydrogenic distribution. The velocity distributions in atomic units used in this calculation are summarized as follows: For the 1s shell of helium,

$$f(v_2)=\frac{32}{\Pi}\frac{Z^5 v_2^2}{(v_2^2+Z^2)^4}, \tag{14}$$

where Z, the effective nuclear charge, is 1.7037.

For the 4p shell of krypton,

$$f(v_2)=\frac{2^{22}}{30\Pi}\frac{Z^7 v_2^4(1280v_2^4-224v_2^2Z^2+5Z^4)^2}{(16v_2^2+Z^2)^{10}}, \tag{15}$$

where Z is 8.25.

For the 4s shell of krypton,

$$f(v_2)=\frac{2^{15}}{\Pi}\frac{Z^5 v_2^2(16v_2^2-Z^2)(256v_2^4-96Z^2v_2^2+Z^4)^2}{(16v_2^2+Z^2)^{10}} \tag{16}$$

where Z is 9.65.

The binding energy, U, used in this calculation is 24.58 eV, 13.996 eV and 27.42 eV for the 1s shell of helium, the 4p shell of krypton, the 4s shell of krypton, respectively.

In Fig. 5 are shown the calculated angular distributions of secondary electrons resulting from the ionizing collisions of 1 keV electrons with helium for several energies of secondary electrons. As shown in Fig. 5, the electrons with lower energies are observed over the wide angular range with broad maxima at the intermediate angle. As the energy increases, these maxima shift towards smaller angle and the shapes of the angular distribution become more sharp. The positions of these maxima correspond to the angles which are derived from the conservation of momentum in the collision between an incident electron and an atomic electron assumed to be at rest before collision. The present calculations were compared with several experimental results. In Figs. 3 and 4 in 2, the calculations for helium and krypton atoms are shown together with experimental results. The experimental results of Opal *et al.*[9] which were normalized using the total ionization cross sections or the elastic scattering cross sections, giving the absolute ordinate scale, are compared with the present calculations in Fig. 6. The angular distributions obtained by these experiments are rather well reproduced by the present calculations in the intermediate angular range, although significant discrepancies exist in the forward and backward directions. The binary encounter cross sections are about 1.5 times as great as the absolute cross sections measured by Opal et al.[9] as shown in Fig. 6, whereas the shapes and positions of broad maxima are well reproduced by the B.E.T. The rise of *ddcs*'s in the forward directions is remakably seen in

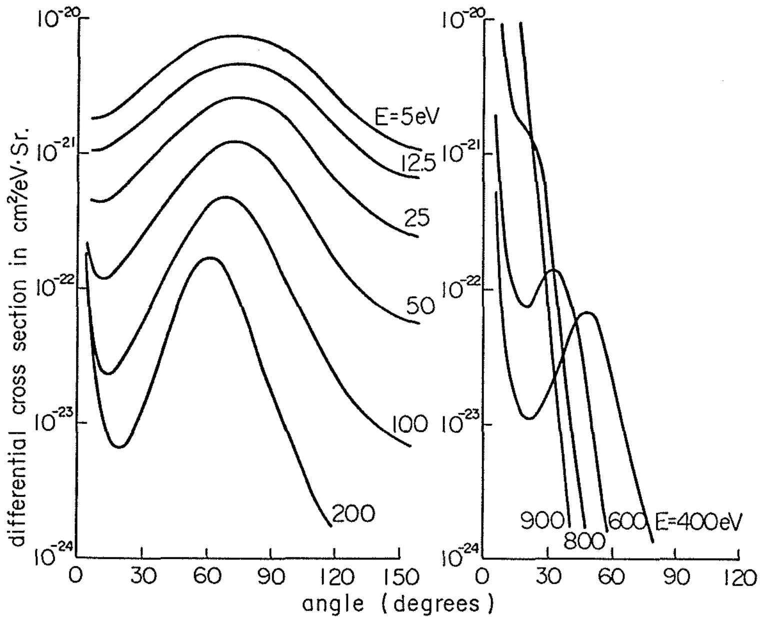


Fig. 5. Angular distribution of secondary electrons resulting from the ionizing collisions of electrons with helium calculated using the binary enounter theory. Primary electron energy is 1 keV. The energies of secondary electrons, E's, are shown on curves.

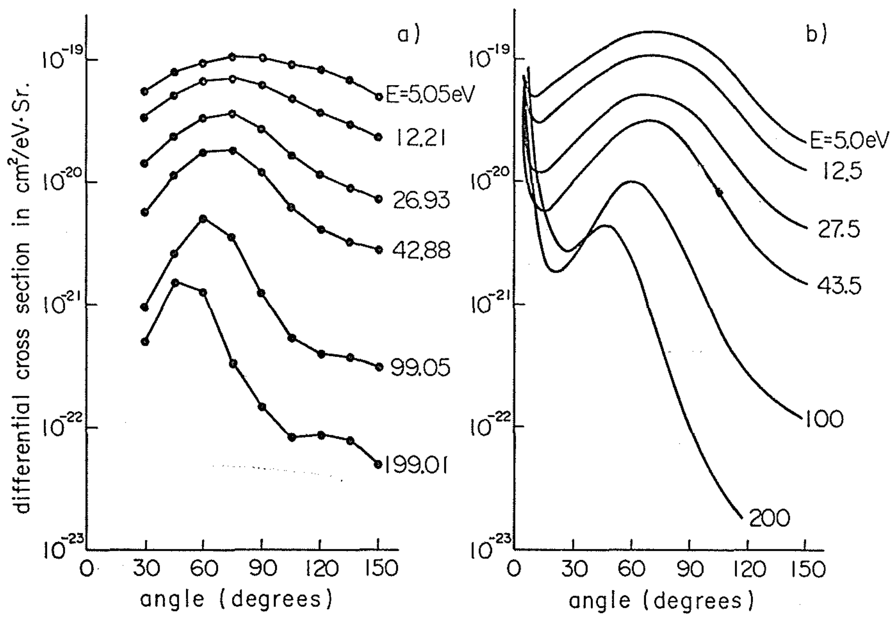


Fig. 6. Angular distribution of secondary electrons resulting from the ionizing collisions of electrons with helium. Primary electron energy is 500 eV. The energies of secondary electrons, E's ,are shown on curves.

a) experiment (Opal *et al.*[9]).

b) double differential cross sections calculated by binary encounter theory.

Figs. 3 and 4.* While the forward rise shown in the angular distributions of electrons ejected by the proton impact[4] was explained by the attractive interaction between the ejected electrons and the scattered protons,[18] the forward rise shown here in the case of electron impact may be regarded to be due to the mechanism quite different from that for the proton impact. As shown in Figs. 3 and 4, the B.E.T. calculations reproduce this forward rise. The cross section calculated theoretically is composed of several terms, *i.e.* direct, exchange

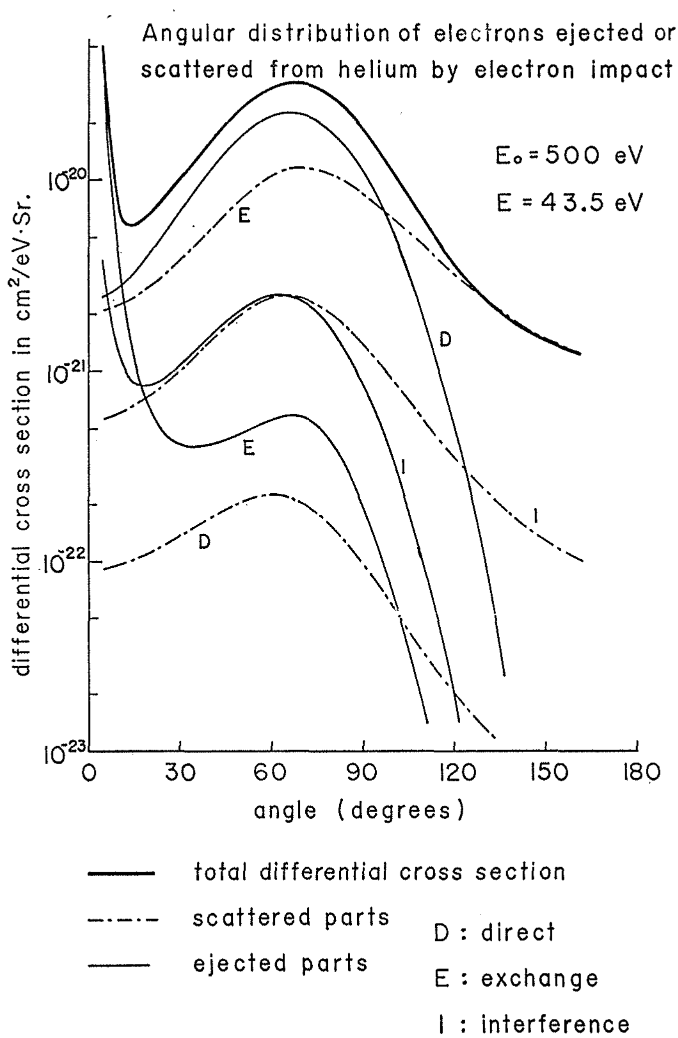


Fig. 7. Six components contributing to the total double differential cross section by the binary encounter theory for helium. Primary energy is 500–eV. The energy of secondary electrons is 43.5–eV.

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* The background counts due to stray electrons ejected from metal surfaces by primary electrons become significant for measurements in the forward directions. The experimental results shown in Figs. 3 and 4 have been obtained by subtracting these background counts from the total counts measured.

and interference term for the ejected electrons as well as for the scattered electrons. Each component contributing to the theoretical total differential cross section is illustrated in Fig. 7 for secondary electrons with energy 43.5 eV. From this figure, it can be seen that the forward rise may be attributed to the exchange term for ejected electrons.

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APPENDIX

The cross section $\sigma_{\pm}(E,P,v_2)$ for transfer of energy between E and $E+dE$ and transfer of momentum between P and $P+dP$ in scattering of electrons with velocity v_1 by atomic electrons with velocity v_2 is given by[14)]

$$\sigma_{\pm}(E,P,v_2)dEdP=\frac{8e^4}{v_1^2v_2X_p^{1/2}}\left[\frac{1}{P^4}\frac{1}{S^4}\pm\frac{2\Phi}{P^2S^2}\right]dEdP\,, \tag{A.1}$$

$$=(\sigma_D+\sigma_E\pm\sigma_I)dEdP\,, \tag{A.2}$$

where σ_+, σ_- are the cross sections for collisions in which the total spin quantum number is respectively 1 or 0, S is the exchange momentum transfer as given in 3, e is the electron charge. In eq. (A.1),

$$X_p=1+2(\hat{v}_1\cdot\hat{v}_2)(\hat{v}_1\cdot\hat{\boldsymbol{P}})(\hat{v}_2\cdot\hat{\boldsymbol{P}})-(\hat{v}_1\hat{v}_2)^2-(\hat{v}_1\cdot\hat{\boldsymbol{P}})^2-(\hat{v}_2\cdot\hat{\boldsymbol{P}})^2\,, \tag{A.3}$$

where $\hat{v}_2$, $\hat{v}_2$ and $\hat{\boldsymbol{P}}$ are the unit vector along v_1, v_2 and momentum transfer vector $\boldsymbol{P}$. The cross section for the unpolarized electrons is derived from σ_+ and σ_- as follows,

$$\sigma(E,P,v_2)=\frac{1}{4}\sigma_+ +\frac{3}{4}\sigma_-\quad,$$

$$=\sigma_D+\sigma_E-\frac{1}{2}\sigma_I\,, \tag{A.4}$$

using the relation between S and θ_e (see eq. (7) in 2).

The double differential cross section for the electrons ejected from atoms is obtained by averaging the cross section $\sigma(E,P,v_2)$ over the direction and magnitude of the velocity v_2. The quantity S must be fixed in this averaging procedure, because S contains the direction of ejected electrons. These procedures were adopted for the derivation of the direct term of the double differential cross section by Bonsen and Vriens.[7)] Hence the derivations for exchange and interference term in eq. 1 in section 2 are given here.

The exchange and interference term of the cross section differential in E and S for the binary collisions are derived from the second and third term of eq. (A.1) as follows,

$$\sigma_{EX}(E,S,v_2)=\frac{8e^4}{v_1^2v_2}\frac{1}{X_S^{1/2}}\frac{1}{S^4}\,, \tag{A.5}$$

$$\sigma_I(E,S,v_2)=\frac{16\Phi e^4}{v_1^2v_2}\frac{1}{X_S^{1/2}}\frac{1}{S^2\{m^2(v_1-v_2)^2-S^2\}}\,. \tag{A.6}$$

The equations (A.5) and (A.6) are obtained using the kinematical relations,

$$S^2+P^2=m^2(v_1-v_2)^2\,, \tag{A.7}$$

$$P^2X_P=S^2X_S\,, \tag{A.8}$$

where X_S is obtained from X_P by replacing $\hat{\boldsymbol{P}}$ in X_P by $\hat{\boldsymbol{S}}$, the unit vector along $\boldsymbol{S}$. When the target electrons have an isotropic velocity distribution, the averaging over the direction of v_2, i.e. the angle χ between v_2 and v_1, can be performed as follows,

$$\sigma_{EX}(E,S,v_2)=\int_{\chi_{min}}^{\chi_{max}}\sigma_{EX}(E,S,v_2)\frac{1}{2}\sin\chi\,d\chi\,,$$

$$=\frac{4\pi e^4}{v_1^2v_2}\frac{1}{S^4}\,, \tag{A.9}$$

$$\sigma_I(E,S,v_2)=\frac{4\pi e^2\Phi}{mv_1^2v_2E}\frac{1}{S^2}\,, \tag{A.10}$$

where the integration limits are determined by the condition $X_S\geqq 0$. The cross sections differential in unit solid angle, *i.e.* eqs. (3) and (4) in 2, are obtained from eqs. (A.9) and (A.10).