Physica **47** (1970) 307–319 © *North-Holland Publishing Co.*

ANGULAR DISTRIBUTION OF ELECTRONS EJECTED BY CHARGED PARTICLES

I. IONIZATION OF He AND H_2 BY PROTONS

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Received 17 October 1969

Synopsis

Cross sections for ejection of electrons from He and H_2 by 100 to 2000 keV protons have been calculated with the binary-encounter theory. Comparisons are made with experimental and with Born cross sections. The binary-encounter cross sections agree very well with the experimental ones for intermediate ejection angles and not too small ejection energies. The discrepancies found at small and at large angles are discussed in some detail.

1. *Introduction.* The binary-encounter theory has been used[1]) previously to study the energy and angular (momentum transfer) distribution of fast charged particles scattered by atomic hydrogen in various initial states. In this theory, the atomic electrons are assumed to be free and independent scattering centers, which assumption was shown to be justified for sufficiently large energy and momentum transfers.

Here we extend the field covered by the binary-encounter theory in three aspects. Firstly, we study the distribution of ejected electrons instead of the distribution of scattered particles. Secondly, we do not confine ourselves anymore to fast incident particles but also consider slow ones. Hence distortion effects, which (among other things) are not included in the binary-encounter theory, will become important and quantitative information on the influence of distortion is obtained from comparisons of binary-encounter and experimental cross sections. Thirdly, in calculating cross sections we do not confine ourselves to hydrogenic target atoms (as in ref. 1), but study the distribution of electrons ejected from He and H_2.

Cross sections for electron ejection from He and H_2 by 50–300 keV protons have been measured by Kuyatt and Jorgensen[2]), who have briefly reviewed the earlier experimental work in this field, Rudd and Jorgensen[3]), and Rudd, Sautter and Bailey[4]). In this paper we make extensive comparisons with Rudd's[4]) experimental and with his Born cross sections. Born calculations have been reported further by Oldham[5]) and binary-encounter calculations have been made by Lee and Hasted[6]), Rudd and Gregoire[7]) and Garcia[8]). Rudd and Gregoire and Garcia independently made the same calculations. They used the cross-section formulae derived by Thomas[9]) to compute the differential cross sections per unit energy transfer for ejection of electrons from He and H_2. In the calculation, a scaled hydrogenic velocity distribution was used for the atomic electrons. Our calculations are more detailed in that we compute double instead of single differential cross sections. The information obtained here on angular distributions for different ejection energies, and thus on the region of validity of the binary-encounter theory, is almost completely lost in the integration over ejection angles[7,8]). Furthermore, we use more accurate wave functions to evaluate the velocity distributions of the atomic (He) and molecular (H_2) electrons. Information about the influence of the ground-state wave function on the cross sections is obtained by using different wave functions in the calculation.

2. *Basic formulae.* We first derive the cross-section formula for ejection of an atomic electron with a particular energy in a certain direction. The starting point for our analysis is the cross section $\sigma(E, P, \boldsymbol{v}_2)\, \mathrm{d}E\, \mathrm{d}P$ for transfer of energy between E and $E + \mathrm{d}E$ and transfer of momentum between P and $P + \mathrm{d}P$ in scattering of particles with velocity $\boldsymbol{v}_1$, mass m_1 and charge z_1e by particles with velocity $\boldsymbol{v}_2$, mass m_2 and charge z_2e. According to eq. (6–3–21) of ref. 10,

$$\sigma(E, P, \boldsymbol{v}_2) = \frac{8z_1^2z_2^2e^4}{v_1^2v_2P^4X_P^{\frac{1}{2}}} \tag{1}$$

for non-identical particles (otherwise exchange and interference terms have to be added[10])). In eq. (1),

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

$\hat{\boldsymbol{v}}_1$, $\hat{\boldsymbol{v}}_2$ and $\hat{\boldsymbol{P}}$ being the unit vectors along $\boldsymbol{v}_1$, $\boldsymbol{v}_2$ and $\boldsymbol{P}$, respectively.

We introduce the "exchange momentum transfer"

$$\boldsymbol{S} = m_1\boldsymbol{v}_1 - m_2\boldsymbol{v}_2' = m_1\boldsymbol{v}_1' - m_2\boldsymbol{v}_2,$$

obtained from $\boldsymbol{P} = m_1\boldsymbol{v}_1 - m_1\boldsymbol{v}_1' = m_2\boldsymbol{v}_2' - m_2\boldsymbol{v}_2$ by interchanging the final momenta $m_1\boldsymbol{v}_1'$ and $m_2\boldsymbol{v}_2'$, as a variable. Then

$$S^2 + P^2 = (m_1\boldsymbol{v}_1 - m_2\boldsymbol{v}_2)^2 - 2(m_1 - m_2)\, E \tag{3}$$

and

$$P^2X_P = S^2X_S, \tag{4}$$

where X_S is obtained from X_P by replacing $\hat{\boldsymbol{P}}$ in X_P by $\hat{\boldsymbol{S}}$, the unit vector along $\boldsymbol{S}$. Eqs. (3) and (4) imply that

$$\mathrm{d}P/X_P^{\frac{1}{2}} = -\mathrm{d}S/X_S^{\frac{1}{2}} \tag{5}$$

and consequently

$$\sigma(E, S, \boldsymbol{v}_2) = |\partial P/\partial S|\,\sigma(E, P, \boldsymbol{v}_2) =$$
$$= \frac{8z_1^2z_2^2e^4}{v_1^2v_2X_S^{\frac{1}{2}}}\,[(m_1v_1')^2 + (m_2v_2')^2 - 2m_1m_2(\boldsymbol{v}_1\cdot\boldsymbol{v}_2) - S^2]^{-2}. \tag{6}$$

If we assume that the target electrons have an isotropic velocity distribution (as in He and H_2), we can integrate over χ, the angle between $\boldsymbol{v}_2$ and $\boldsymbol{v}_1$, and over the azimuthal angle ϕ of $\boldsymbol{v}_2$ with respect to $\boldsymbol{v}_1$. Because ϕ is not a variable in eq. (6), $\sigma(E, S, \boldsymbol{v}_2) = \sigma(E, S, v_2, \chi, \phi) = \sigma(E, S, v_2, \chi)$ and

$$\sigma(E, S, v_2) = \int_{\chi_{\min}}^{\chi_{\max}} \sigma(E, S, v_2, \chi)\, \tfrac{1}{2}\sin\chi\, \mathrm{d}\chi, \tag{7}$$

where the integration limits are determined by the condition $X_S \geq 0$. Eqs. (6) and (7) yield

$$\sigma(E, S, v_2) = [\pi z_1^2z_2^2e^4S/4(m_1m_2)^{\frac{3}{2}}v_1^2v_2E^3] \times$$
$$\times [-S^4 + 2S^2\{(m_1v_1)^2 + (m_2v_2)^2 - (m_1 - m_2)\,E\}$$
$$- \{(m_1v_1')^2 - (m_2v_2)^2\}\{(m_1v_1)^2 - (m_2v_2')^2\}] \times$$
$$\times [S^2 + (m_1 - m_2)(m_2v_2'^2 - m_1v_1^2)]^{-\frac{3}{2}}. \tag{8}$$

Next, we introduce the ejection angle θ, between $\boldsymbol{v}_2'$ and $\boldsymbol{v}_1$. Hence $S^2 = (m_1v_1)^2 + (m_2v_2')^2 - 2m_1m_2v_1v_2'\cos\theta$. Multiplying eq. (8) by $|\partial S/\partial\theta|$ then gives the differential cross section $\sigma(E, \theta, v_2)$ for ejection of the atomic (molecular) electron between θ and $\theta + \mathrm{d}\theta$ and with an energy between $E - U_i$ and $E - U_i + \mathrm{d}E$, where U_i is the binding energy of the electron which is ejected. The following step involved is multiplication by $(2\pi\sin\theta)^{-1}$ which replaces the variable θ by the (cone of) solid angle $\mathrm{d}\Omega = \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi$ in the direction $\hat{\boldsymbol{v}}_2' = \hat{\boldsymbol{v}}_2'(1, \theta, \varphi)$. The resulting expression may be written as

$$\sigma(E, \hat{\boldsymbol{v}}_2', v_2) = \frac{z_1^2z_2^2e^4v_1v_2'}{2m_2v_2E^3} \times$$
$$\times\left[m_2v_2'^2\sin^2\theta - E\left\{1 - \frac{v_2'}{v_1}\left(1 + \frac{m_2}{m_1}\right)\cos\theta + \frac{m_2v_2'^2}{m_1v_1^2}\right\}\right]|v_1 - v_2'|^{-3}, \tag{9}$$

where v_2' stands, as before, for $(v_2^2 + 2E/m_2)^{\frac{1}{2}}$. Straightforward application of the laws of conservation of energy and momentum yields the following conditions:

1) for $\cos\theta > 0$ and $v_1' \geq v_1 \cos\theta$, $v_2 \geq \left|\dfrac{A - B}{C}\right|$,

2) for $\cos\theta > 0$ and $v_1' \leq v_1 \cos\theta$, $\left|\dfrac{A - B}{C}\right| \leq v_2 \leq \dfrac{A + B}{C}$,

3) for $\cos\theta < 0$ and $v_1' \geq v_1 |\cos\theta|$, $v_2 \geq \dfrac{A + B}{-C}$, and

4) for $\cos\theta < 0$ and $v_1' \leq v_1 |\cos\theta|$, no v_2 values allowed.

Here,

$$A = (m_1 + m_2)\, v_1' E,$$

$$B = v_1 |\cos\theta| \{(m_1 - m_2)^2 E^2 + 2m_1^2 m_2 v_1^2 E \sin^2\theta\}^{\frac{1}{2}}$$

and

$$C = m_1 m_2 \{v_1^2 \cos^2\theta - v_1'^2\}.$$

To make comparisons with experiment possible, we have next to integrate over the velocity distribution $f(v_2)$ of the atomic (molecular) electrons, between the limits given by conditions 1) to 4), and we have to multiply by the number N of electrons in the atom (molecule):

$$\sigma(E, \hat{\boldsymbol{v}}_e) = N \int_{v_{2\,\min}}^{v_{2\,\max}} \sigma(E, \hat{\boldsymbol{v}}_e, v_2)\, f(v_2)\, dv_2, \tag{10}$$

where $\hat{\boldsymbol{v}}_e$ is the direction of the ejected electrons. As in ref. 11 we assume that $\hat{\boldsymbol{v}}_e = \hat{\boldsymbol{v}}_2'$ and $v_e^2 = v_2'^2 - v_2^2 - 2U_i/m_2$.

Eq. (10) implies that

$$\int_0^\infty f(v_2)\, dv_2 = 1. \tag{11}$$

The integrations in eq. (10) are carried out numerically in this paper.

3. *Application to* He *and* H_2. Among the various wave functions available in literature for He, we now have to select one which provides us with a sufficiently accurate velocity distribution and thus with sufficiently accurate binary-encounter cross sections. For several ejection energies and angles, we calculated therefore cross sections for ejection of electrons ($z_2^2 = 1$) by protons ($z_1^2 = 1$), using five different wave functions. The simplest two of these are hydrogenic (1s) ones, which yield

$$f(v_2) = \frac{32 v_2^2 v_0^5}{\pi (v_2^2 + v_0^2)^4}, \tag{12}$$

with $\frac{1}{2}mv_0^2 = 24.58$ eV and $\frac{1}{2}mv_0^2 = 39.49$ eV, respectively. Hence in the first case we took the average kinetic energy of the atomic electrons equal to their binding energy which is incorrect, but this choice corresponds to the scaled Born cross sections of Rudd *et al.*[4]) In the second case, the expectation value of $\frac{1}{2}mv_2^2$ is correctly given by eq. (12), as follows directly from the virial theorem. The third wave function used is the two-term Hartree–Fock one of Green *et al.*[12]), which gives

$$f(v_2) = \frac{8v_2^2}{\pi}\left\{\frac{\alpha A}{(v_2^2+\alpha^2)^2} + \frac{2\alpha B}{(v_2^2+4\alpha^2)^2}\right\}^2, \tag{13}$$

where $\alpha = 1.4558$, $A = 0.7421$ and $B = 0.4453$ when v_2 is given in units of $e^2/\hbar$. The fourth wave function used is the three-term Hartree–Fock one of Bagus and Gilbert (unpublished, but given in ref. 13), which yields

$$f(v_2) = \frac{8v_2^2}{\pi}\left\{\frac{\alpha A}{(v_2^2+\alpha^2)^2} + \frac{B(3\beta^2-v_2^2)}{(v_2^2+\beta^2)^3} + \frac{C(3\gamma^2-v_2^2)}{(v_2^2+\gamma^2)^3}\right\}^2, \tag{14}$$

where $A = 4.75657$, $B = -1.40361$, $C = -1.26842$, $\alpha = 1.450$, $\beta = 2.641$ and $\gamma = 1.723$, again with v_2 given in units of $e^2/\hbar$. Finally, the fifth wave function is the three-term Hylleraas one of Hicks[14]). The Fourier transform

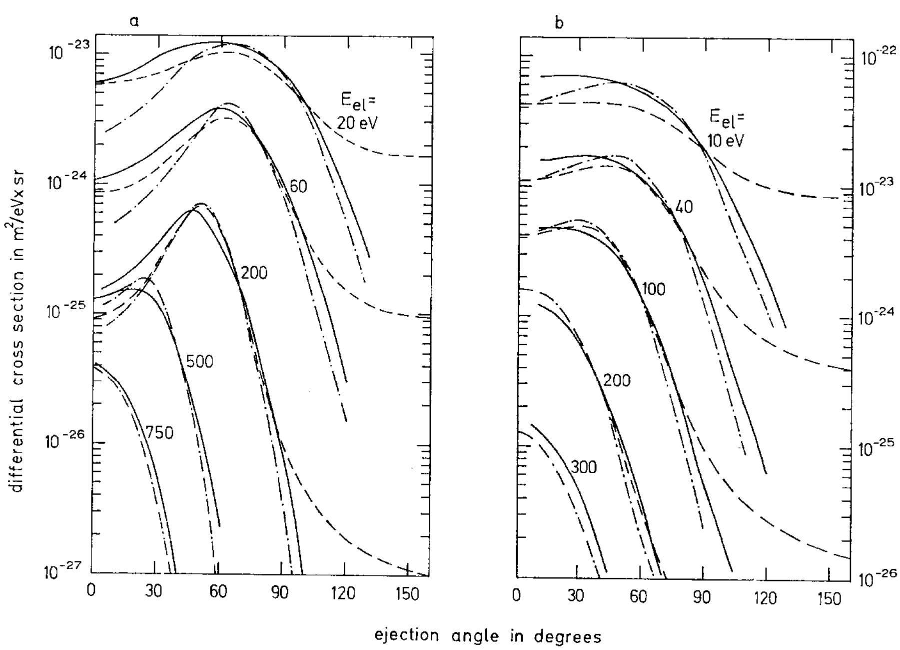

Fig. 1. Cross sections $\sigma(E, \hat{v}_e)$ for ejection of electrons from He by 300 keV (a) and 100 keV (b) protons. Broken curves: scaled Born cross sections; full curves: H.F. binary-encounter cross sections; dot-dashed curves: scaled hydrogenic binary-encounter cross sections. The numbers in the figure give the energy of the ejected electrons. The broken and dot-dashed curves coincide for 500 and 750 eV ejected electrons (a) and for 300 eV electrons (b).

of the latter wave function can be obtained in a straightforward way because it contains only a term r_{12}^2 and no terms with odd powers of r_{12}, the interelectronic distance. Nevertheless, the resulting expression is quite lengthy and because it is probably not superior to eq. (14) it is not reproduced here.

In fig. 1 we compare binary-encounter cross sections, obtained with the three-term H.F. and the hydrogenic ($\frac{1}{2}mv_0^2 = 24.58$ eV) velocity distributions, with the scaled hydrogenic Born cross sections of Rudd *et al.*[4]). The two-term H.F. results almost coincide (within 0.1 to 1%) with the three-term H.F. results, the three-term Hylleraas cross sections differ by no more than 3% from the three-term H.F. ones, and the hydrogenic ($\frac{1}{2}mv_0^2 = 39.49$ eV) cross sections differ only for large angles to a maximum of about 5% from the three-term H.F. ones. Because of these small differences, the latter results are not shown in fig. 1. Fig. 1 shows that the hydrogenic (24.58 eV) binary-encounter and the hydrogenic (24.58 eV) Born[4]) cross sections become almost identical to each other for sufficiently large E, with exception of the very large ejection angles. From fig. 1 it also follows that the hydrogenic ($\frac{1}{2}mv_0^2 = 24.58$ eV) and H.F. binary-encounter cross sections agree with each other only for intermediate angles. In the forward and backward directions discrepancies occur up to a factor of 2. We thus find that scaling of hydrogenic cross sections (binary-encounter *and* Born) on the expectation value of $\frac{1}{2}mv_2^2$ (39.49 eV) leads to cross sections that are in much closer agreement with the more accurate H.F. ones than scaling on the ionization energy U_i (24.58 eV). Scaling on $\langle\frac{1}{2}mv_2^2\rangle$ must therefore be preferred above scaling on U_i. The most striking feature of fig. 1 is that the Born cross sections are orders of magnitude larger than the binary-encounter ones for very large ejection angles. A qualitative explanation for this difference is given in ref. 15. It arises from the interaction of the ejected electron with the rest of the atom, which makes it possible that the electron is ejected in a direction more or less opposite to the one in which it has received a momentum, owing to the (impulsive) interaction with the passing proton.

For molecular hydrogen we used two wave functions for which Fourier transforms could easily be made. The simplest of these is the valence-bond Wang[16]) function which yields[17,18])

$$f(v_2) = \frac{32\alpha^5}{\pi(1+s^2)(v_2^2+\alpha^2)^4}\left\{1 + s\,\frac{\sin(v_2R)}{v_2R}\right\}, \tag{15}$$

where $s = (1 + \alpha R + \alpha^2R^2/3)\exp(-\alpha R)$, $\alpha = 1.166$, $R = 1.406$ and v_2 is given in atomic units ($e^2/\hbar$). The second wave function used is the valence-bond (with ionic term added) function of Weinbaum[19]). The resulting velocity distribution is

$$f(v_2) = \frac{32c_1^2\alpha^5}{\pi(1+s^2)(v_2^2+\alpha^2)^4} \times$$
$$\times \left[1 + c_2^2 + 2c_2 \frac{\sin(v_2R)}{v_2R} + \left\{2c_2 + (1+c_2^2)\frac{\sin(v_2R)}{v_2R}\right\}s\right], \quad (16)$$

where s has the same meaning as in eq. (15), $\alpha = 1.193$, $R = 1.417$, $c_1 = 0.806$ and $c_2 = 0.256$. The binary-encounter cross sections calculated with these two velocity distributions differ by no more than 5%.

4. *Comparison with experiment.* In figs. 2, 3 and 4 we compare the binary-encounter cross sections for ejection of electrons from He (eq. 14) and H_2 (eq. 16) by impact of 100 and 300 keV protons, with the corresponding experimental cross sections of Rudd *et al.*[4]). Good agreement is found for intermediate angles and not too small energy transfers (ejection energies). Owing to the more accurate ground-state wave function used in the binary-encounter theory, better agreement with experiment is obtained than in the Born approximation (see fig. 1) for these intermediate angles and not too small energy transfers.

As has been explained already in section 3, the binary-encounter theory leads to very poor results for large ejection angles, and the experimental cross sections become much larger than the binary-encounter ones.

In the forward direction, the experimental cross sections are much larger than the binary-encounter and Born cross sections for energy transfers smaller than about mv_1^2, m being the electron mass and v_1 the proton velocity. These discrepancies may be caused by the long range interaction of the ejected electron and the proton, which is not correctly accounted for in the theories considered so far. Some qualitative arguments about this distortion effect are given in ref. 15. Detailed theoretical calculations which take account of this effect have been reported very recently by Salin[20]), for ionization of the hydrogen atom by protons.

Energy distributions of electrons ejected at 10° are shown in figs. 5, 6 and 7. These figures illustrate more clearly the large discrepancies found at small θ. The second hump in the cross-section curves is reproduced very well by the theory. In this connection we note that binary-encounter cross sections calculated with the Thomson model, of target particles that are initially at rest, depend in a very simple way on E and θ:

$$\sigma(E, \hat{v}_e) = \frac{z_1^2 e^4}{m_2 v_1^2 E^2}\, \delta\left(\cos\theta - \frac{m_1+m_2}{m_1 v_1}\sqrt{\frac{E}{2m_2}}\right), \quad (17)$$

which reduces to

$$\sigma(E, \hat{v}_e) = \frac{e^4}{m v_1^2 E^2}\, \delta\left(\cos\theta - \sqrt{\frac{E}{2mv_1^2}}\right) \quad (18)$$

for incident protons and target electrons. One can easily verify that the positions of the humps in figs. 5 to 7, as well as the positions of the maxima of the binary-encounter curves in figs. 1 to 4, correspond to

$$E = P^2/2m = 2mv_1^2 \cos^2 \theta$$

and thus to the position of the δ-function in eq. (18). Figs. 5 to 7 and 2 to 4 show that the binary-encounter theory works well for $E > 2mv_1^2 \cos^2 \theta$.

To illustrate more clearly at which velocities of the ejected electrons the maximum discrepancies between theory and experiment occur, we plotted in fig. 8 the ratios of experimental and theoretical cross sections *vs.* $(v_e/v_1)^2$ for ejection angles of 10° and 20°. For all proton energies studied (100, 200 and 300 keV), the maximum discrepancies occur when the velocity of the ejected electron is about equal to that of the proton.

In fig. 9 we plotted binary-encounter cross sections for impact of 2 MeV protons on He. Compared to figs. 1 to 4, we see that the angular distributions are more sharply peaked around the maxima which positions are predicted by eq. (18).

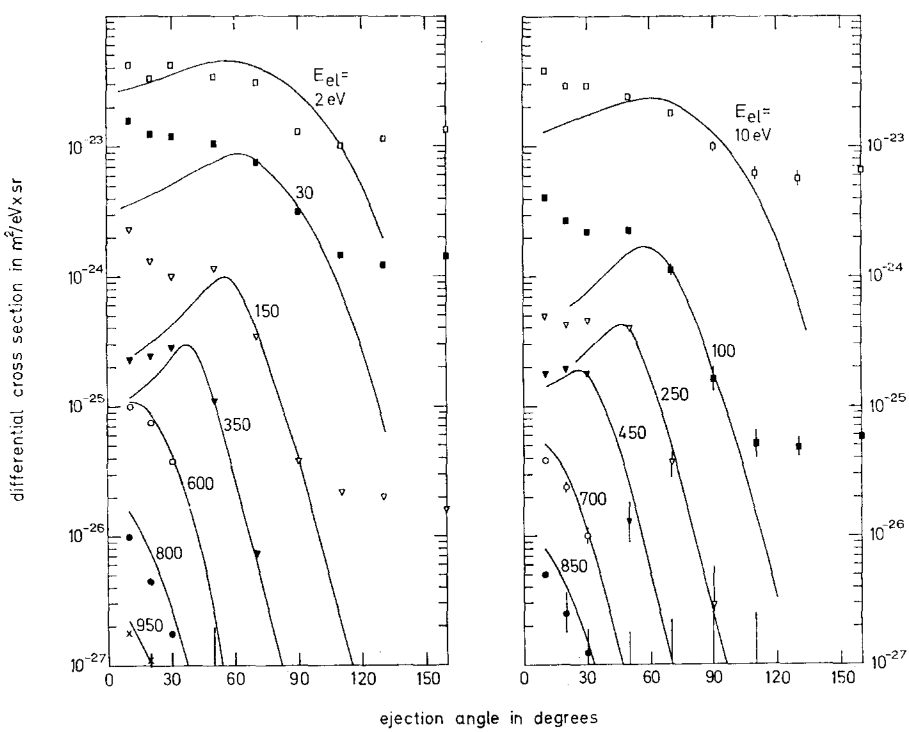

Fig. 2. Angular distribution of electrons ejected from He by 300 keV protons. Full curves: binary-encounter cross sections; squares, triangles and circles: experiment (Rudd *et al.*). Different symbols have been used to distinguish between different ejection energies, given by the numbers in the figure.

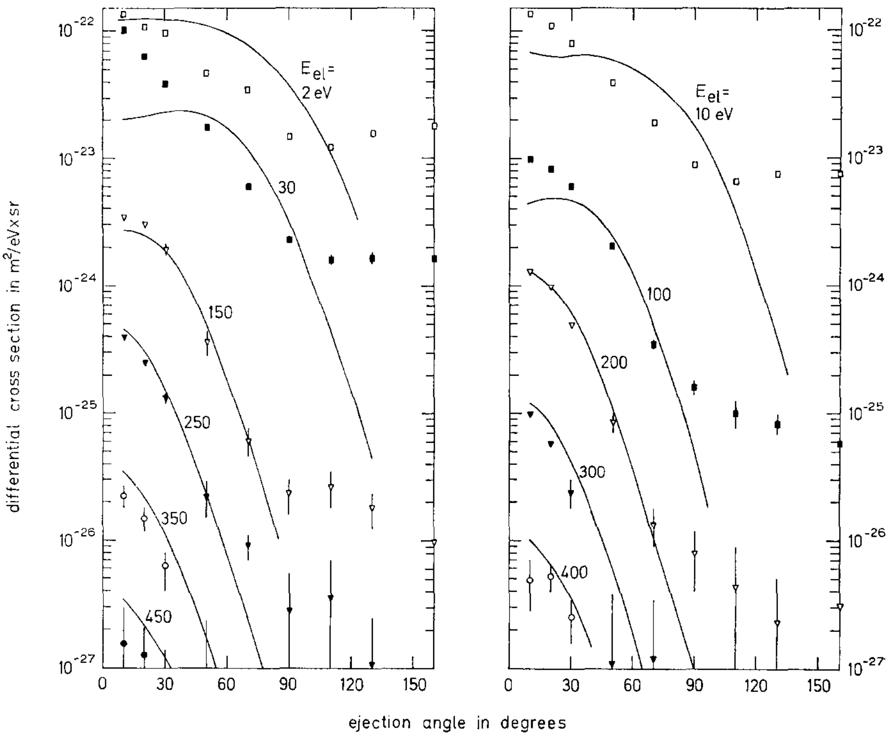

Fig. 3. As in fig. 2 for 100 keV protons.

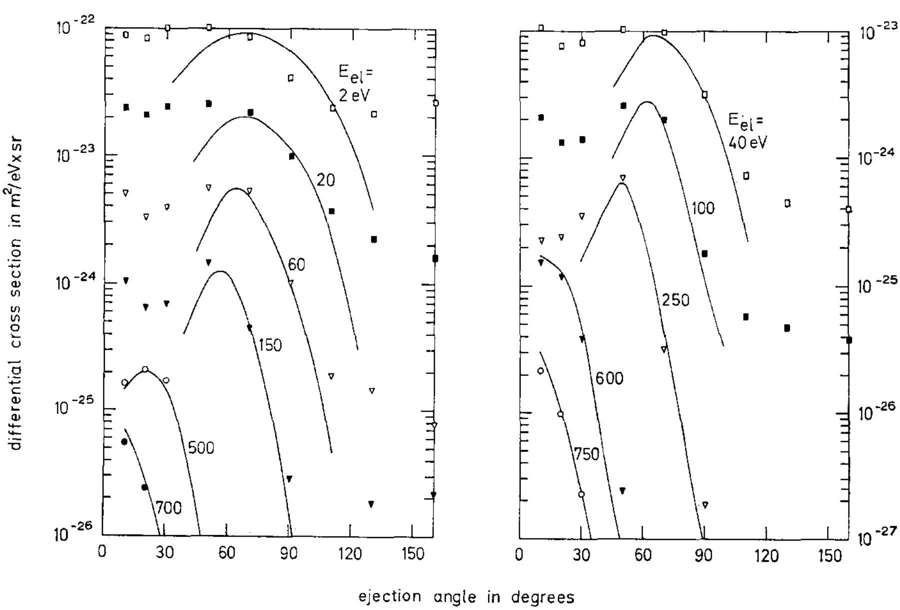

Fig. 4. As in fig. 2 for H_2 and 300 keV protons.

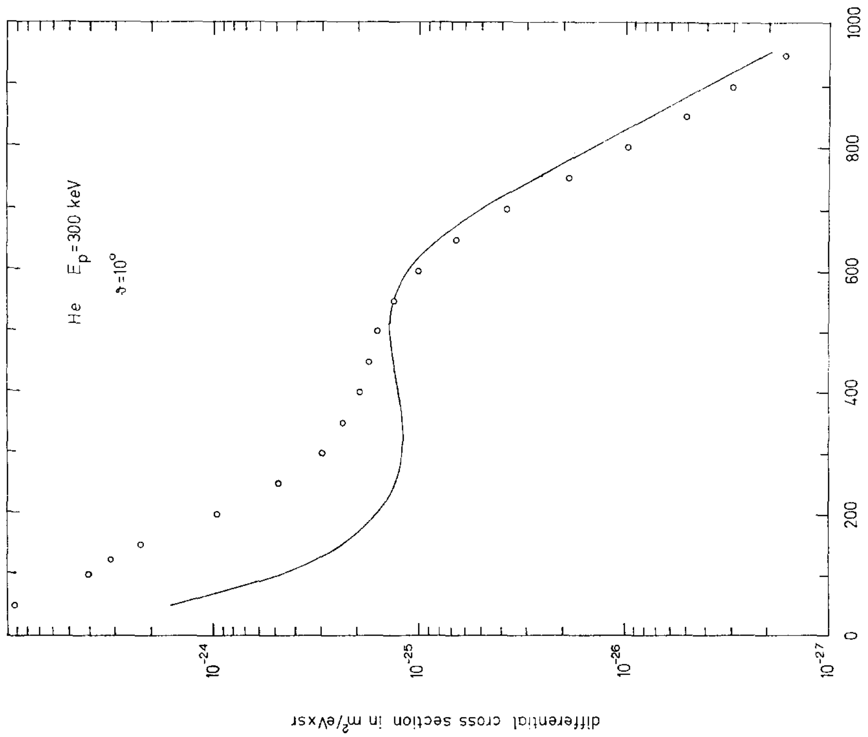

Fig. 5. Energy distribution of electrons ejected from He at 10° with respect to the 300 keV proton beam. Full curve: binary-encounter theory; circles: experiment (Rudd *et al.*).

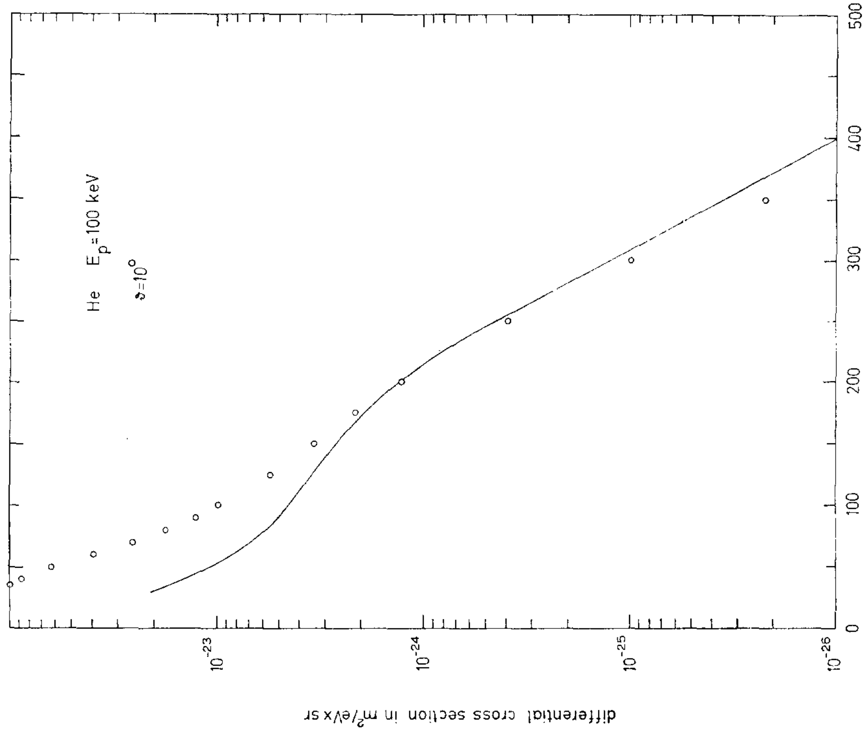

Fig. 6. As in fig. 5 for 100 keV protons.

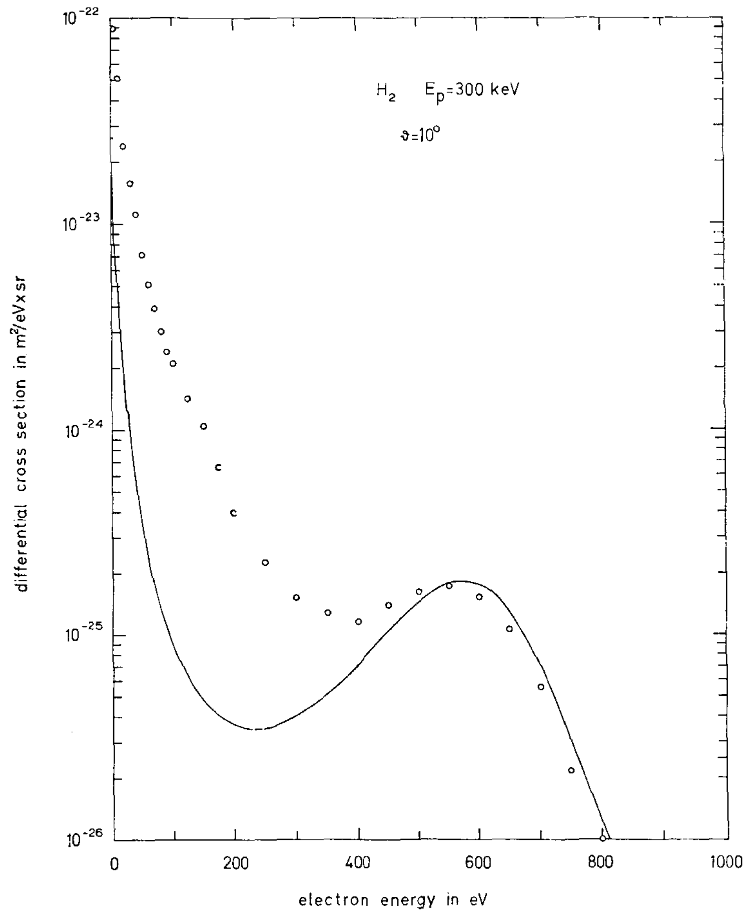

Fig. 7. As in fig. 5 for 300 keV protons incident on H_2.

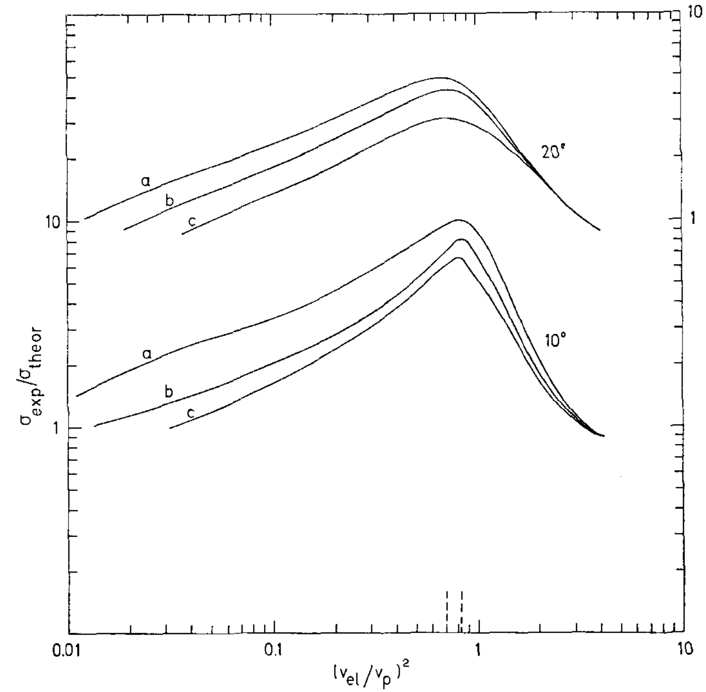

Fig. 8. Ratio of experimental and binary-encounter cross sections for electron ejection from He at 10° and at 20° by 300 keV (a), 200 keV (b) and 100 keV (c) protons. The 10° and 20° data are displaced with respect to each other. The left-hand scale corresponds to the 10° data and the right-hand scale to the 20° data.

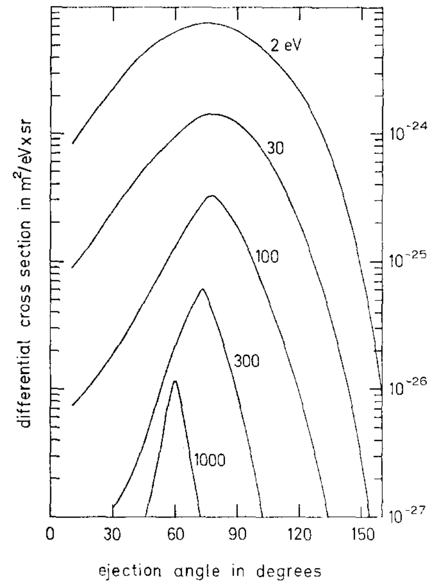

Fig. 9. Binary-encounter cross sections $\sigma(E, \hat{\boldsymbol{v}}_e)$ for electron ejection from He by 2000 keV protons. The numbers in the figure give the energy of the ejected electrons.

5. *Conclusions.* The angular and energy distributions of electrons ejected from He and H_2 by protons are well described by the binary-encounter theory in a significant range of ejection angles and ejection energies. The Born approximation is shown to be superior to the binary-encounter theory only for very large ejection angles ($> 90°$). These results are of practical importance because it is easier to calculate binary-encounter than to calculate Born cross sections, especially for atoms or molecules more complicated than He and H_2. More advanced theories like the one of ref. 20 will be necessary for small ejection angles and not too large ejection energies.

Acknowledgements. We are indebted to Professor M. E. Rudd for generously providing us with very extensive tabular data on his experimental and Born cross sections. This investigation is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie" (F.O.M.) and was made possible by financial support of the "Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek" (Z.W.O.).

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