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CALCULATION ON IONIZING H-ATOM H-ATOM COLLISIONS

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ABSTRACT

The Born approximation is used to calculate the cross sections when an hydrogen atom in an excited state with the principal quantum number n collides with an hydrogen atom in the ground state. Calculation has been carried out when the first atom is ionized while the second atom may have any final state. The cases considered are n = 1, 2, 3, and 4. A closure approximation originally used by Lodge is also used here for high impact energy calculations. We find disagreement with some of the Lodge's numerical results.

At sufficiently high impact energies the cross sections are inversely proportional to the energies. Coefficients of proportionalities for all cases are given. When the incident atom is in a high principal quantum number such that the relation $(m/M)\,K^2\,a_0^2 > n^2 \gg 1$ is satisfied, with K^2 the impact energy in rydberg, m and M the electronic and the atomic masses, and n the principal quantum number considered, the cross section is given as $Q_0 = 4(M/m)\left(\ell n\, 4n^2 - 1/2\right)\left(\pi a_0^2\right)/K^2$.

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I. INTRODUCTION AND FORMULATION

In recent years astrophysicists and plasma physicists have expressed increased interest in the reactions that excited hydrogen atoms undergo when they collide with other hydrogen atoms. In this paper[1] we consider the following three reactions which lead to the ionization of the excited atom:

$$H(n\ell) + H(1s) \rightarrow H^+ + e + H(1s) , \tag{1}$$

$$\rightarrow H^+ + e + H(n'\ell') , \tag{2}$$

$$\rightarrow H^+ + e + H^+ + e , \tag{3}$$

where $n\ell$ are the principal and the azimuthal quantum numbers of the incident atom, and $n'\ell'$ are those of the target atom after collision.

Bates and Griffing[2] have shown that the cross sections for the reactions (1), (2) and (3) in the Born approximation are given by

$$Q(n\ell \rightarrow c,\ 1s \rightarrow n') = \frac{2}{\pi s^2} \sum_m a_m \int dk \int_{q_{min}}^{q_{max}} |T(n\ell m \rightarrow k,\ 1s \rightarrow n')|^2 \, q\, dq . \tag{4}$$

Here, using atomic units, q is the momentum transfer by the excited atom in the center of mass system, m is the absolute value of the magnetic quantum number of the excited atom; $a_m = \epsilon_m/(2\ell+1)$ where $\epsilon_m = 1$ if $m = 0$ and $\epsilon_m = 2$ if $m \neq 0$, and $\mathbf{k}$ is the momentum of the ejected electron. The final state of the target atom is indicated by n. For reaction (2) the cross sections reported here include a summation over all possible bound excited states of the target atom. When the target is also ionized as in reaction (3), $n' = \mathbf{k}'$, and an additional integration with respect to $\mathbf{k}'$ is carried out. The initial relative velocity is s, and finally the absolute values of the transition matrices are given by

$$|T(n\ell m \to \mathbf{k},\ 1s \to 1s)| = \frac{4\pi}{q^2}\left|\left[1 - \frac{16}{(4+q^2)^2}\right]\langle \mathbf{k}\,|e^{i\mathbf{q}\cdot\mathbf{r}}|\,n\ell m\rangle\right| \tag{5}$$

$$|T(n\ell m \to \mathbf{k},\ 1s \to n'\ \ell'\ m')| = \frac{4\pi}{q^2}\left|\langle \mathbf{k}\,|e^{i\mathbf{q}\cdot\mathbf{r}}|\,n\ell m\rangle\langle n'\ \ell'\ m'\,|e^{i\mathbf{q}\cdot\mathbf{r}}|\,1s\rangle\right| \tag{6}$$

$$|T(n\ell m \to \mathbf{k},\ 1s \to \mathbf{k}')| = \frac{4\pi}{q^2}\left|\langle \mathbf{k}\,|e^{i\mathbf{q}\cdot\mathbf{r}}|\,n\ell m\rangle\langle \mathbf{k}'\,|e^{i\mathbf{q}\cdot\mathbf{r}}|\,1s\rangle\right| \tag{7}$$

For evaluation of the matrix elements of $\exp(i\mathbf{q}\cdot\mathbf{r})$ in these equations between $n\ell m$ and $\mathbf{k}$ the general formula given by Omidvar and Sullivan[3] is used, while for the squared modulus of the matrix elements of $\exp(i\mathbf{q}\cdot\mathbf{r})$ between $1s$

and $n'\ \ell'\ m'$, summed over $\ell'\ m'$, the formula given by Bethe[4] is used:

$$\sum_{\ell,\,m} \left| \langle n\ell m \left| e^{i\mathbf{q}\cdot\mathbf{r}} \right| 1s \rangle \right|^2$$

$$= 2^8\, n^7\, q^2 \left[\frac{n^2-1}{3} + (qn)^2 \right] \times \frac{\left[(n-1)^2 + (qn)^2 \right]^{n-3}}{\left[(n+1)^2 + (qn)^2 \right]^{n+3}} \quad . \qquad (8)$$

Concerning the limits on q without loss of accuracy we can at high incident energies let $q_{max} \to \infty$, while to second order in m/M, with m the mass of an electron and M the reduced mass of the system,

$$q_{min} \sim \frac{\Delta E}{2s} \left[1 + \frac{m\,\Delta E}{4M\,s^2} \right] , \qquad \frac{m\,\Delta E}{M\,s^2} \ll 1 , \qquad (9)$$

ΔE being the sum of the excitation energies of the two atoms. For the three reactions (1), (2) and (3) ΔE is given in rydberg units by $k^2 + 1/n^2$, $k^2 + 1/n^2 + 1 - 1/n'^2$, and $k^2 + 1/n^2 + 1 + k'^2$, respectively.

Simplification arises by assuming that q_{min} in (4) is independent of the final state of the target atom. It will be shown below that this assumption is justified when the incident particle has sufficiently high energy. By putting $q_{min} = q_0$ in this case, summing the right hand side of (4) with respect to all the excited states of the target atom, and making use of the closure property of the hydrogen atom

wave functions, we find that

$$Q\left(n\ell \to c,\ 1s \to \sum_{n'}{}'\right) = \frac{128\pi^2}{s^2}\int dk \int_{q_0}^{\infty} \frac{dq}{q}\left[1 - \frac{1}{(1+q^2/4)^4}\right] \times \left|\langle k \,|e^{iq\cdot r}|\, n\ell m\rangle\right|^2 , \qquad (10)$$

where summation with respect to n' includes excitation to the discrete, as well as to the continuum, states of the target atom.

Lodge,[5] in order to determine q_0, has assumed that the largest contribution to the cross section arises from the excited states of the target atom for which $k' \cong k$. This then leads to $\Delta E = 2k^2 + 1/n^2 + 1$, and through (9) q_0 is determined. If, at sufficiently high energies, the contributions from (2) and (3) are added, agreement is found with the value given by (10) using the above value of q_0. This can be seen from Figures 1-3, in the next section, for the C curve is just the sum of (1) and (10). It should be noted, however, that the integrand in (10) for q small behaves as a positive power of q. At sufficiently high incident energies it is therefore immaterial what value for q_0 is used as long as $q_0 \ll 1$.

In contrast to the case of the charged particle-neutral atom collision, the dipole $1/r^2$ interaction potential does not exist for neutral atom-neutral atom collisions. The long range van der Waals potential behaves as $1/r^7$ for distances larger than 137 times the atomic radius, and as $1/r^6$ for smaller distances, and is not important for inelastic collisions. The foregoing discussion implies that

collisions with large inpact parameters are not important. This will have two observable effects. First, few low energy electrons are ejected in neutral-neutral collisions in contrast to the charged particle-neutral collisions; second, the total cross section at high energies is inversely proportional to the incident energy, and the collisional cross section is given by a single parameter. The energy distribution of the ejected electrons and these parameters will be given in the next section.

We now derive an analytic expression at high impact energies for the cross section when the incident atom is in a highly excited state. Since only close collisions are important in atom-atom collisions, when $n^2 >> 1$, with n the principal quantum number of the incident atom, the interaction of the nucleus of the incident atom with the rest of the system can to some extent be neglected, and the problem resembles the scattering of an electron by the ground state of the target atom. This point has been recognized by a number of workers in the field.[6]

However, the presence of the nucleus of the incident atom has the effect of eliminating the $1/r^2$ potential which exists for inelastic electron-atom collisions. We can then treat the problem as the problem of the electron-atom collision provided we eliminate contributions from the small momentum transfers which corresponds to the long range potential.

The inelastic cross section in the Born approximation for scattering of an electron by an atom when the momentum transfer is between q_1 and q_{max} and all

states of the atom are excited is given by[7]

$$Q_0^{in} = \frac{8\pi}{k_1^2} \int_{q_1}^{q_{max}} \frac{dq}{q^3} \left[1 - \left| \langle 0 | e^{iq\cdot r} | 0 \rangle \right|^2 \right] , \qquad (11)$$

with k_1^2 the electron energy in rydberg and $\langle 0|$ the initial state of the atom. In (11) q_1 must be larger than the minimum momentum transfer for the transitions that contribute appreciably to the total cross section. For atomic hydrogen and provided $\Delta\epsilon/k_1^2 \ll 1$, with $\Delta\epsilon$ the excitation energy of the atom, (11) reduces to

$$Q_0^{in} = \frac{4\pi a_0^2}{k_1^2} \left[\ell n \left(\frac{4}{q_1^2 a_0^2} \right) - \frac{13}{12} \right] , \qquad q_1 > \frac{\Delta\epsilon}{2k_1} , \qquad \frac{\Delta\epsilon}{k_1^2} \ll 1 . \qquad (12)$$

The second inequality is also the criterion for the validity of the Born approximation.

We now fix on an expression for q_1. In the impact parameter formulation for atom-atom collisions it would be physically reasonable and accurate to ignore impact radii greater than the radius of the incident, excited atom. For larger radii the interaction between energetic neutral atoms will be negligible. Now q_1 is related to this cut-off radius and an analogous and similarly accurate approximation in the momentum change integral is to assume that $q_1 \simeq 1/na_0$.

To get the total cross section we must add to (12) the contribution from the elastic cross section given by[8] $Q_0^{e\ell} = 7\pi a_0^2/3k_1^2$, $k_1^2 \gg 1$. With these considerations we find for the total cross section

$$Q_0 = Q_0^{e\ell} + Q_0^{in} = \frac{4\pi a_0^2}{k_1^2} \left[\ell n\, 4n^2 - 1/2 \right] . \qquad (13)$$

For transitions that contribute appreciably to the total cross section $\Delta\epsilon$ in (12) lies between 3/4 to 3 rydbergs. We can then combine the condition $n^2 >> 1$, the inequalities in (12), and the criterion for the validity of the Born approximation in the inequalities

$$k_1^2 a_0^2 > n^2 >> 1 \ . \tag{14}$$

In terms of the energy of the incident atom (13) and (14) can be combined into

$$Q_0 = 4(M/m)\left(\ell n\, 4n^2 - 1/2\right)\left(\pi a_0^2\right)/K^2 \ ,$$

$$(m/M) K^2 a_0^2 > n^2 >> 1 \ , \tag{15}$$

with M the mass and K^2 the energy in rydberg of the incident atom, and m the mass of the electron. Equation (15) shows that for sufficiently high incident energies and high principal quantum number of the projectible, n, the ionization cross section increases as $\ell n\, n^2$. Examination of Equation (15) for n = 3 and 4 shows that the results obtained from this equation differ about 2% and 1% respectively from the results of the detailed calculation shown in Table 1.

For low incident energies where the inequalities in (15) are not satisfied but still $n^2 >> 1$, the Born approximation is not valid and the elastic cross section becomes comparable or larger than the inelastic cross section. Derivation of an analytic expression in this case does not seem to be easy. It can however be said that for sufficiently low energies where the inelastic cross section can be

neglected compared to the elastic cross section (cf. Figures 1, 2, 3), the total cross section as n increases should approach an upper bound.

II. RESULTS AND DISCUSSION

Since the azimuthal angles are cyclic in all the integrations, it is evident from (4) that for single ionization a triple, and for double ionization a quadruple, numerical integration should be carried out to find the corresponding cross sections, the integration with respect to the angle of colatitude of $\mathbf{k}'$ being done analytically. These integrations were carried out using the Gaussian quadrature. At impact energies above 100 KeV the sum of reactions (2) and (3) differed by at most a few percent from the results given by (10). This later equation, with the value for q_0 given by Lodge, was therefore used to obtain most of the results at higher energies. This saved much computer time.

In Figure 1 the sum of the cross sections for reactions (1), (2) and (3), when both incident and target atoms are in the ground state, are compared with the measurements of McClure[9] and Wittkower et al.[10] The data of Wittkower et al. contains a contribution from charge exchange

$$H + H \rightarrow H^+ + H^- \ , \tag{16}$$

Between 3.15 KeV and 63 KeV McClure determined the cross section for (16) and subtracted it out. At higher energies he didn't do this but above 100 KeV the contribution from charge exchange is unimportant.[9] Whittkower's incident beam

consisted only of H(1s) atoms while McClure's beam contained some excited hydrogen atoms. However, McClure determined that their presences increased his measurements by at most only a few percent.

The cross sections for (1), (2) and (3) were previously calculated by Bates and Griffing[2] for ground state collisions. Our results for reaction (3) and for the sum of the three reactions are about 2% higher than the graphical results of these authors.

Also shown in Figure 1 is the curve designated by C, which is obtained by using the results of the closure relationship, Equation (10), for the contribution of the excited states of the target atom, and using Lodge's choice for q_0. Above 500 KeV we have estimated reaction (3) by subtracting reaction (2) from the closure relation (10). It should be pointed out that neither in this case, nor in the cases of $n = 2$ and 3, do we find agreement with the numerical values of Lodge. As explained later on, we attribute this to an error in his calculations.

In Figures 2 and 3 similar curves with the same definitions are given for $n = 2$ and 3 cases. It is seen from Figures 1 and 2 that the closure relationship gives the same results as the explicit integrations for energies larger than 100 KeV. Below this energy and for $n = 2$ and 3 it underestimates the total cross sections due to an underestimation of reaction (2) in which the target is excited. Since the experimental results also agree well with those of the Born approximation for energies higher than 100 KeV, it seems that the closure relationship provides a useful way of calculating the total cross sections.

As discussed in the previous section, due to the lack of the $1/r^2$ potential, the cross sections fall off at high energies as the inverse of the energy E. In Figure 4 the product of the energy and cross section, $E \times Q(E)$, is plotted versus E. It clearly is seen that all curves approach their asymptotes which are horizontal lines. Thus at high energies we can write

$$Q(n) = C_n E^{-1} \tag{17}$$

In Table I we give C_n for the various reactions and n = 1, 2, 3, 4 evaluated at 1000 KeV. These C_n and (17) yield usable cross sections for impact energies greater than 500 KeV. For n greater than 4 use Equation (15).

Examination of (4) indicates that for the cross section to take the form (17), the momentum change integral must become a constant, independent of its lower bound q_{min}. We have tested this by putting $q_{min} = 0$ for all energies of the ejected electron and also by use of Lodge's q_J for q_{min} and no noticeable variation in the cross section occurred at high energies. This insensitivity to q_{min} explains the accuracy of the Lodge approximation at high impact energies.

In order to experimentally investigate the dependence of the total cross section on the state n of the projectile, McClure[9] has suggested the formula

$$Q(n) = Q(1s)\, n^{\alpha} . \tag{18}$$

Then by (17)

$$\alpha = \ell n \left(c_n/c_1\right)\Big/\ell n\, n \ . \tag{19}$$

Using this formula, we find $\alpha = 0.917$ for $n = 2$, $\alpha = 0.834$ for $n = 3$, and $\alpha = 0.775$ for $n = 4$. These values are in agreement with the findings of McClure which indicate that α must be less than one, although his measurements are for the comparatively low energy range of 25-100 KeV.

For the excited atom in states $n = 1, 2, 3$ Lodge has previously used reactions (1) and Equation (10) to calculate the curves c shown in Figures 1-3. His results are shown by the dashed lines in Figure 5. The solid lines are our results for the same calculation. We attribute the disagreement of the two sets of curves to an error in Lodges' calculations. He indicates that his reaction (1) calculation for $n = 1$ disagrees with that of Bates and Griffing at low energy. Lodge attributes this to the fact that he has dropped the second order term in q_{min} (cf. Equation (9)). We have checked this and have found that this cannot be the cause of the discrepancy. We can only assume that Lodge has some error in his calculation the effect of which is more pronounced for $n > 1$.

Lodge also repeated the calculation but used a plane wave to represent the ejected electrons. Considering that this approximation is not particularly accurate for impact energies as low as 100 KeV, his results in this case are in reasonable agreement with our curves shown in Figure 5.

In Figures 6 and 7 are shown the energy distribution of ejected electrons when the projectile is a H(2s) atom with energies of 100 KeV and 500 KeV respectively. The contributions of the three reactions are shown and the sum is compared with the electron distribution in an H(2s)-proton collision. In the latter case most of the electrons are ejected with low energies and $dQ/d\epsilon$ for high ϵ behaves as $1/\epsilon^2$, with ϵ the energy of the ejected electron. Note the much reduced size of the low energy peak in the problem of atom-atom collisions. For the 100 KeV projectile, reaction (I), where the target is not excited, dominates the distribution at high ϵ and in this region $dQ/d\epsilon$ also falls off as $1/\epsilon^2$ for atom-atom collisions. The dominance of reaction (1) is of course much more pronounced at lower impact energies. However at higher impact energies reaction (3) becomes most important as Figure 7 indicates. Even in this case reaction (1) is the larger in the very high energy tail, but this region contributes little to the total cross section. Note that in reaction (3) two free electrons are produced, both of these are taken into account in Figures 6 and 7.

In Tables II and III we present numerical values of the cross sections for all three reactions and their sum when the projectile is in the principle quantum states $n = 1$ and $n = 2$ respectively. Note that in Table III the 2s and averaged 2p cross sections are usually about the same size. In fact for all n the cross section seems not to vary greatly with the angular momentum ℓ of the excited electron.

All calculations were done on the IBM 360-91 at the Goddard Space Flight Center.

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Table I

Constants, C_i, for the asymptotic formula $Q_i = C_i/E$, are given for reactions (1), (2), (3) and the sum. Units of C are (KeV πa_0^2). The C's are obtained from cross sections calculated at 1000 KeV and yield cross section accurate to within a few percent at energies greater than 500 KeV.

Reaction	(1)	(2)	(3)	Sum (1 + 2 + 3)
(1s → c, 1s → n')	37.5	17.6	71.5	127
($\bar{2}$ → c, 1s → n')	50.6	59.2	129	239
($\bar{3}$ → c, 1s → n')	54.1	101	161	316
($\bar{4}$ → c, 1s → n')	54.2	136	180	370

Table II

Cross sections for the ionization of H(1s)

when it collides with H(1s) in units of (πa_0^2).

Impact Energy		Reaction			
KeV	Log (E)	(1)	(2)	(3)	(1 + 2 + 3)
1	0	0.010	—	—	0.010
2.5	0.398	0.098	—	—	0.098
5	0.699	0.288	0.000	0.000	0.288
10	1.0	0.499	0.004	0.001	0.504
15	1.176	0.569	0.017	0.007	0.594
25	1.398	0.573	0.056	0.043	0.672
35	1.544	0.531	0.091	0.098	0.721
50.1	1.7	0.460	0.115	0.185	0.760
63.1	1.8	0.408	0.120	0.241	0.769
79.4	1.9	0.355	0.118	0.285	0.758
99.9	2.0	0.301	0.111	0.309	0.721
158.5	2.2	0.211	0.086	0.296	0.594
199.5	2.3	0.172	0.074	0.269	0.515
316.2	2.5	0.113	0.054	0.200	0.364
501	2.7	0.074	0.034	0.137	0.244
1000	3.0	0.038	0.018	0.071	0.127
1,585	3.2	0.024	0.011	0.046	0.081

Table III

Cross sections for the ionization of H(2ℓ) in collisions with H(1s) in units of (πa_0^2).

Impact Energy	Reaction (1) $Q(2\ell \rightarrow c,\ 1s \rightarrow 1s)$			Reaction (2) $Q(2\ell \rightarrow c,\ 1s \rightarrow \sum_B')$			Reaction (3) $Q(2\ell \rightarrow c,\ 1s \rightarrow c)$			Sum (1 + 2 + 3) $Q(2\ell \rightarrow c,\ 1s \rightarrow \Sigma)$		
KeV	2s	$2\bar{p}$	$\bar{2}$	2s	$2\bar{p}$	$\bar{2}$	2s	$2\bar{p}$	$\bar{2}$	2s	$2\bar{p}$	$\bar{2}$
1	0.472	0.820	0.733							0.472	0.820	0.733
2.5	1.42	1.50	1.48							1.42	1.50	1.48
5	1.78	1.73	1.75	0.002	0.006	0.005	0.000	0.000	0.000	1.78	1.74	1.76
10	1.71	1.64	1.66	0.037	0.085	0.073	0.003	0.010	0.008	1.75	1.74	1.74
15	1.52	1.47	1.49	0.262	0.254	0.256	0.015	0.049	0.041	1.80	1.77	1.75
25	1.21	1.19	1.20	0.839	0.645	0.694	0.193	0.221	0.214	2.24	2.06	2.11
35	0.996	.989	0.991	0.971	0.860	0.888	0.503	0.447	0.461	2.47	2.30	2.34
50.1	0.782	0.780	0.780	0.843	0.840	0.841	0.793	0.708	0.729	2.42	2.33	2.35
63.1	0.659	0.657	0.657	0.723	0.752	0.745	0.877	0.812	0.828	2.26	2.22	2.23
79.4	0.547	0.548	0.548	0.610	0.645	0.636	0.882	0.844	0.854	2.04	2.04	2.04
99.9	0.449	0.448	0.448	0.509	0.544	0.535	0.832	0.815	0.819	1.79	1.81	1.80
158.5	0.299	0.300	0.300	0.339	0.362	0.357	0.652	0.651	0.652	1.29	1.31	1.31
199.5	0.241	0.241	0.241	0.281	0.301	0.296	0.553	0.555	0.554	1.08	1.10	1.09
316.	0.156	0.156	0.156	0.177	0.187	0.185	0.379	0.381	0.380	0.712	0.724	0.721
501	0.100	0.100	0.100	0.113	0.119	0.118	0.250	0.251	0.251	0.463	0.470	0.469
1000	0.051	0.051	0.051	0.057	0.060	0.059	0.127	0.131	0.129	0.235	0.242	0.239
1,585	0.032	0.032	0.032	0.036	0.038	0.037	0.082	0.082	0.082	0.150	0.152	0.152

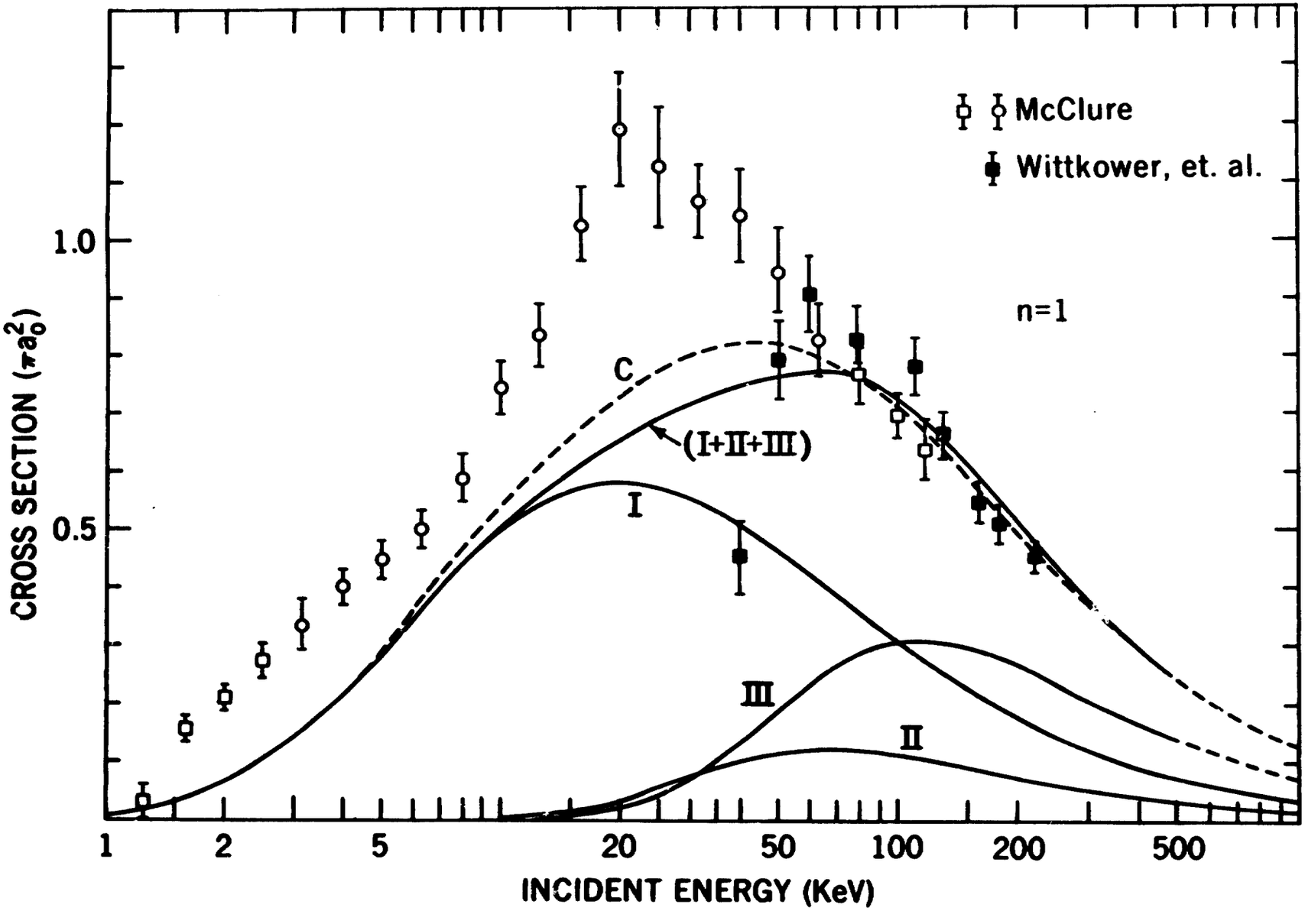

Figure 1. Cross sections for incident H(1s) on H(1s). Curves I, II, and III are due to the reactions (1), (2), and (3). Curve C is the sum of the cross sections using closure relationship, Equation (10), with q_0 given by Lodge. The experimental points of McClure and Wittkower et al. indicated by squares also contain a contribution from charge exchange, (16). The open circles indicate measurements where McClure subtracted out this contribution. Above 100 KeV charge exchange is unimportant. The energy in the abscissa is that of the incident atom in the laboratory system.

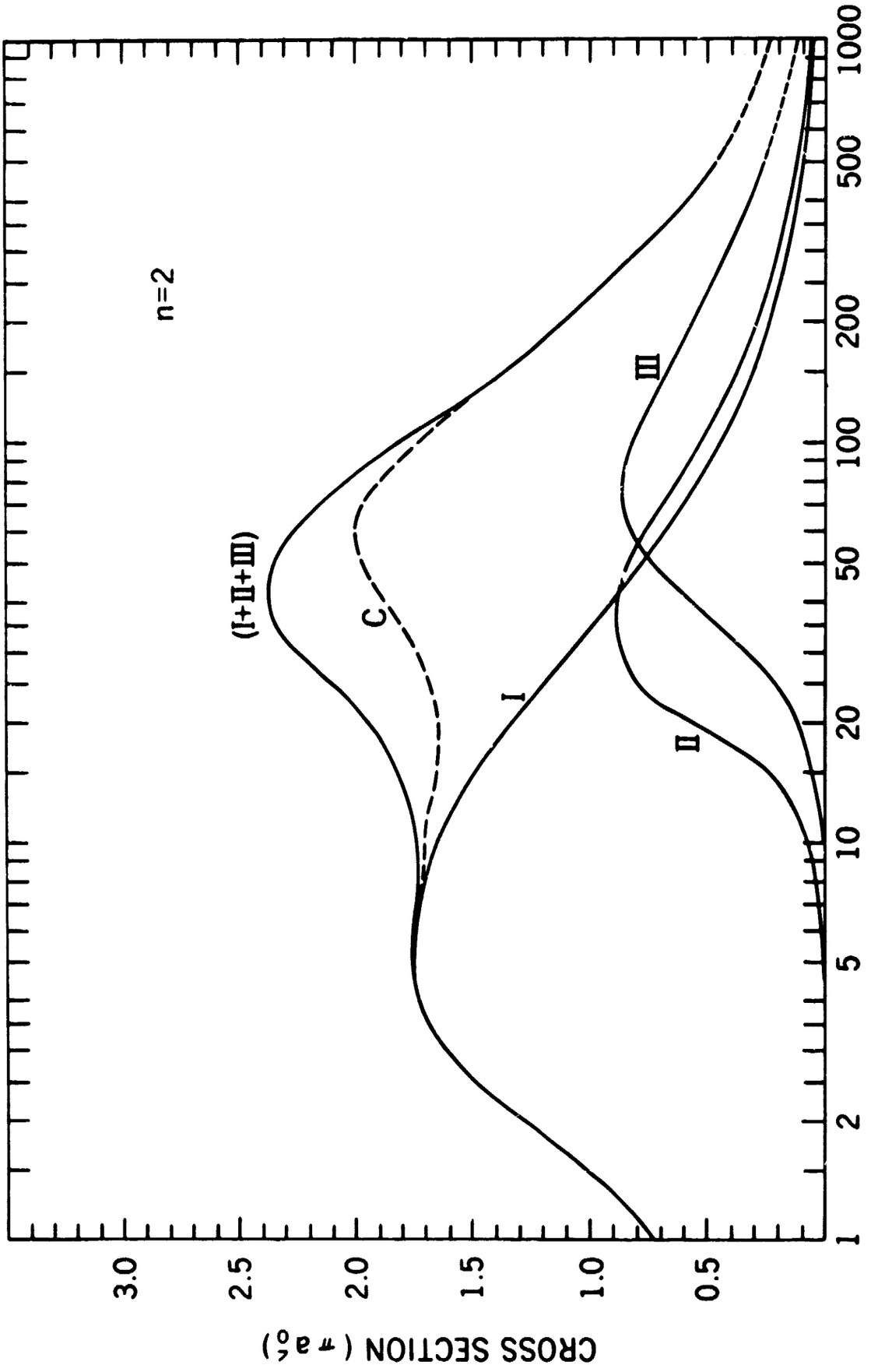

Figure 2. The same as in Figure 1, but with the projectile in n = 2 states.

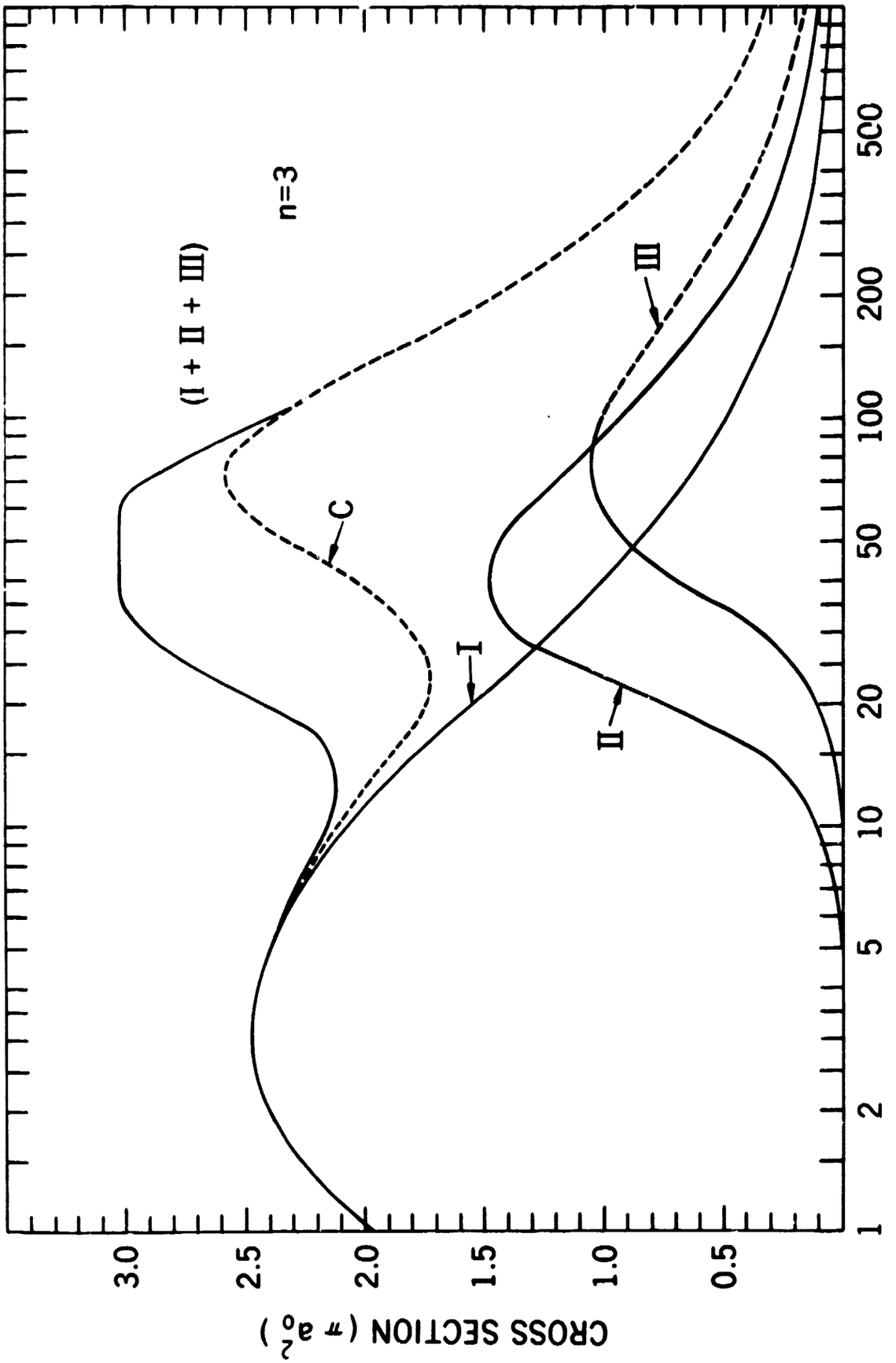

Figure 3. The same as in Figure 1, but with the projectile in n = 3 states.

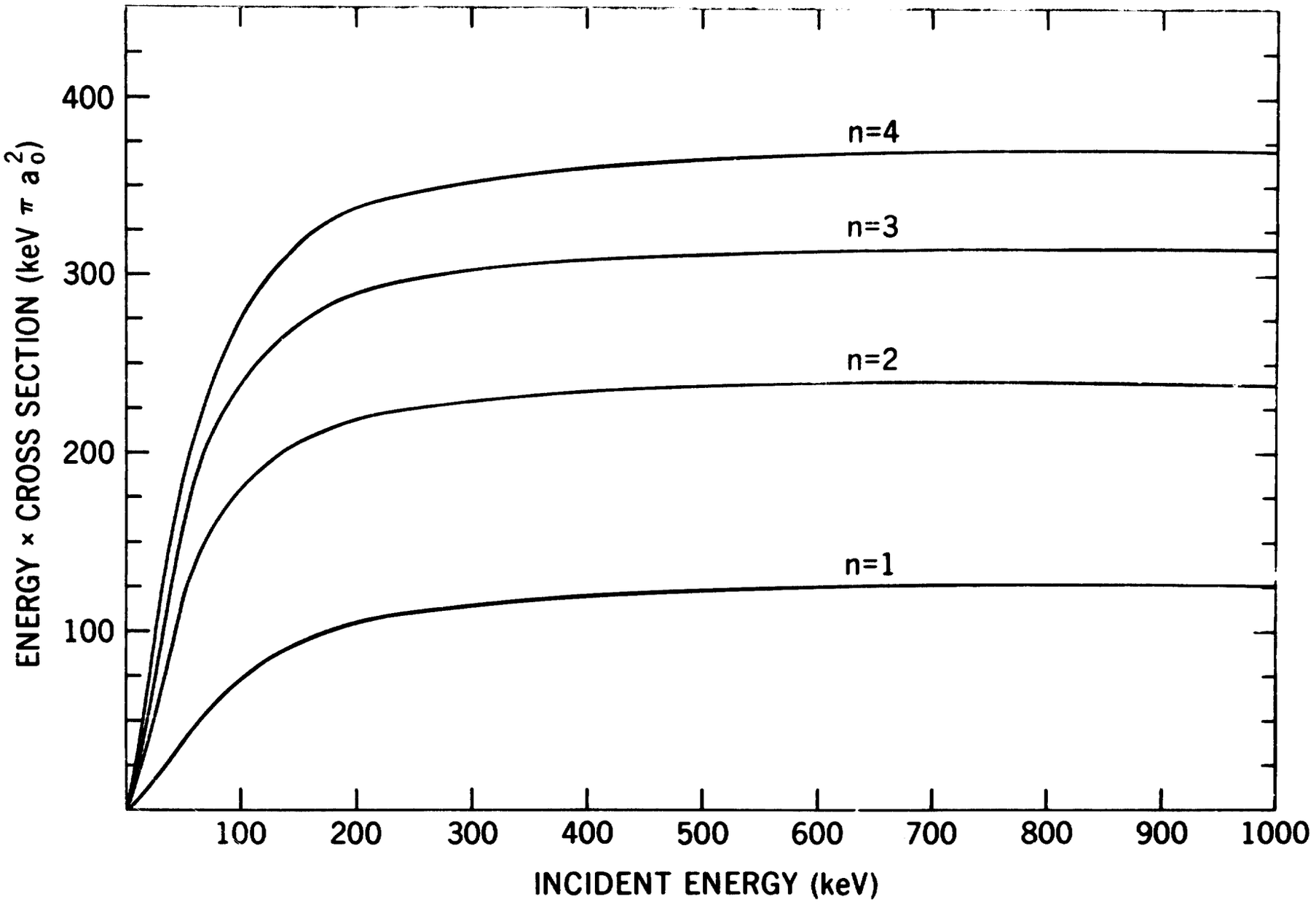

Figure 4. The total first Born ionization cross section multiplied by the energy is plotted against the incident energy for the projectile in principle quantum states n = 1, 2, 3 and 4.

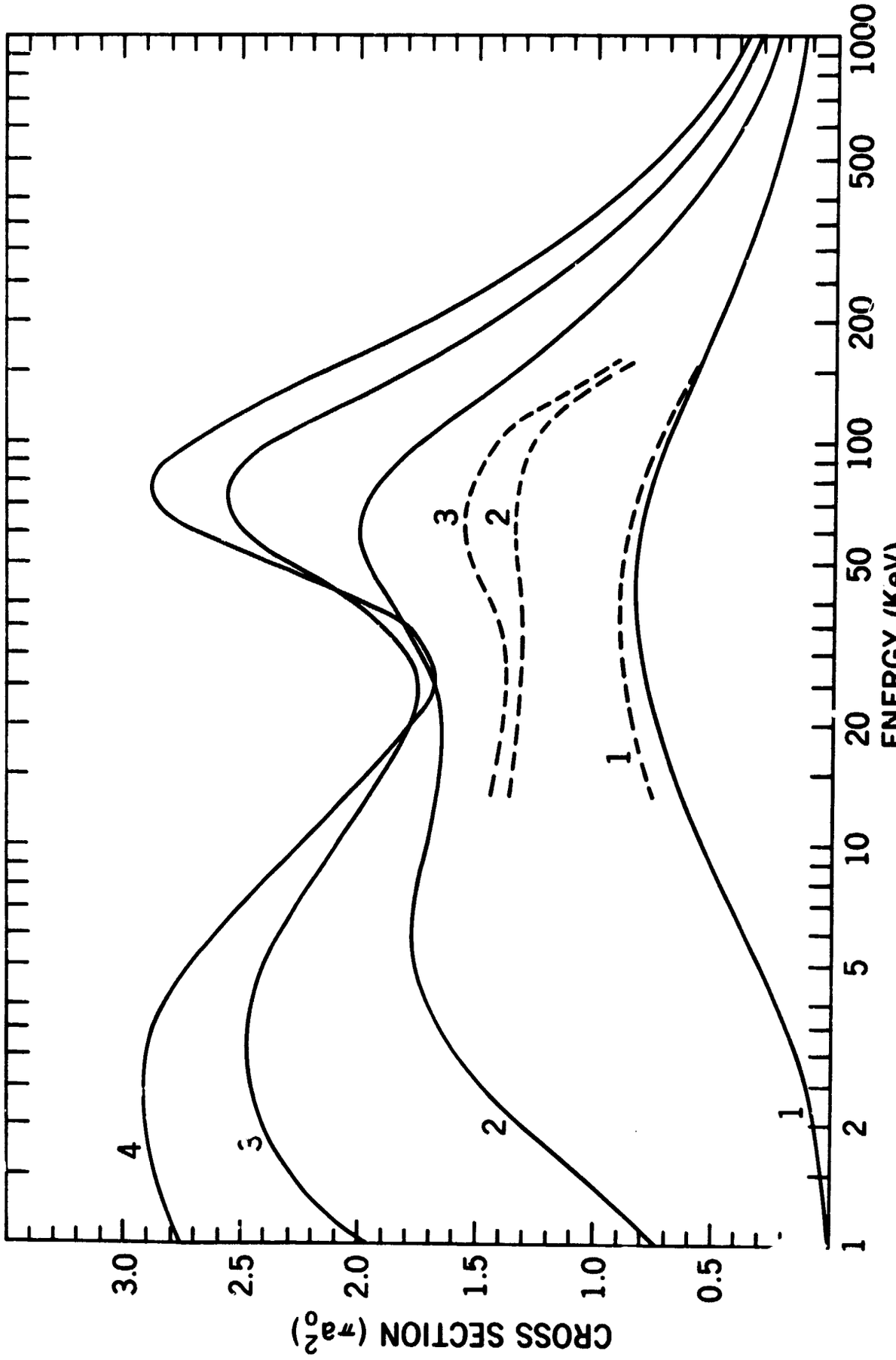

Figure 5. A plot of total cross section Q versus E for n = 1, 2, 3 and 4. All curves are calculated using the closure relation, Equation (10), with q_0 given by Lodge. The solid curves are our calculations, the dashed those of Lodge (Reference 5).

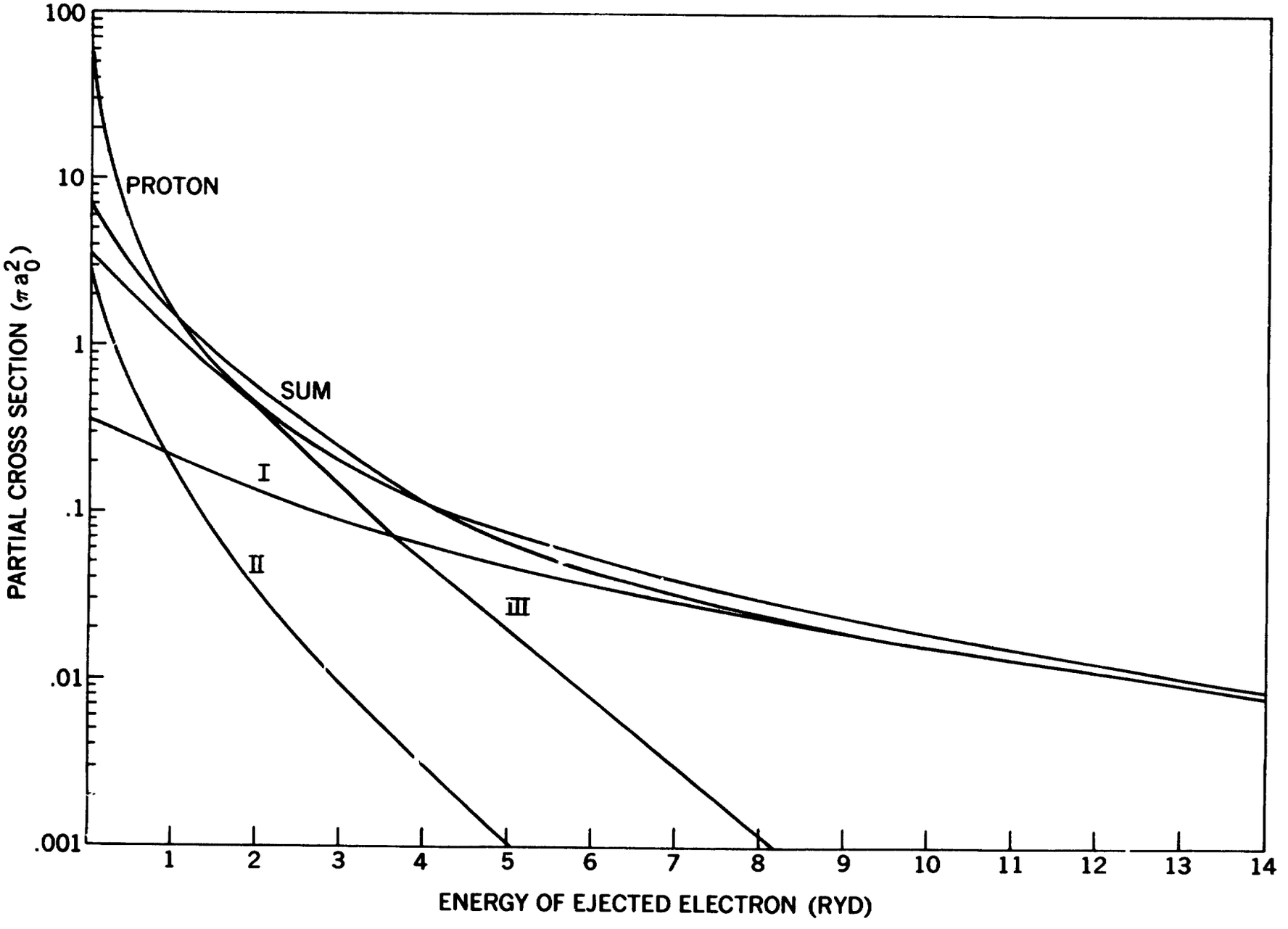

Figure 6. For reactions (1), (2), (3) and the sum, the plot shows the cross section for an electron being ejected with a given energy when H(2s) collides with H(1s) at 100 KeV impact energy. The same probability is also shown for a proton incident on H(2s). Note that in reaction (3) two electrons are produced per reaction.

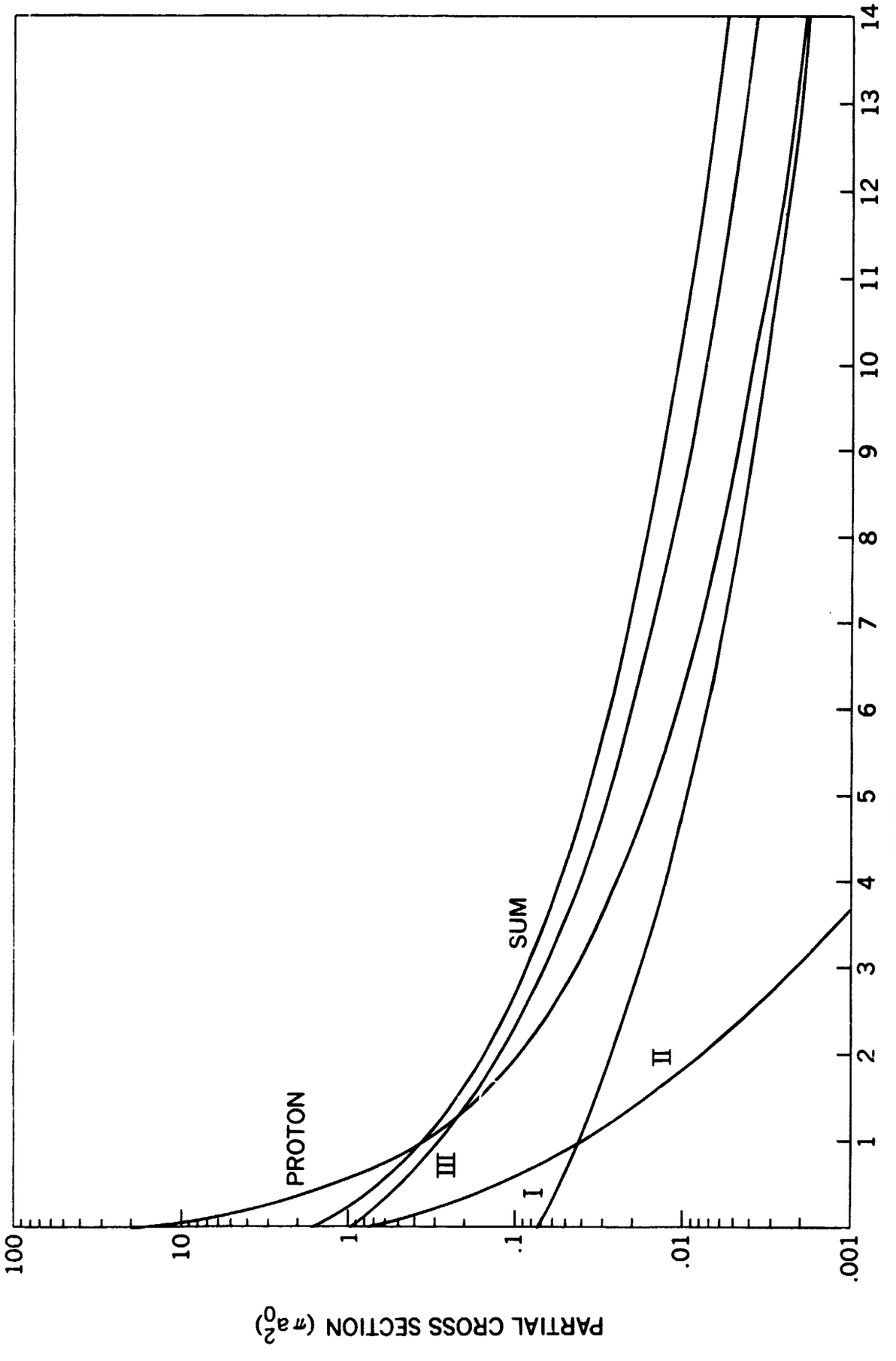

Figure 7. The same as Figure 6 but here the impact energy is 500 KeV.