

EXCITATION AND IONIZATION OF ATOMIC HYDROGEN FROM VARIOUS STATES

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Synopsis

For large energies E_1 of the impinging electrons, the cross sections Q for optically allowed excitation and ionization of atoms are given by $Q = A/E_1 \ln cE_1$, where A and c are constants for one transition. For excitation A is proportional to the optical oscillator strength for the transition. For ionization A is proportional to a quantity M_i^2 which can be derived with one of the sum rules for the oscillator strengths.

For excitation, values of cU (U is excitation energy) are calculated here using known values of Born cross sections for large E_1 or using known values of quantities related to cU . It is indicated how excitation cross sections for small E_1 can be calculated approximately with known values of A and c . It is pointed out that this method probably will be more reliable than "classical" methods.

For ionization, values of M_i^2 and cU_i (U_i is ionization energy) are calculated. Large discrepancies between cU_i values for the same transitions are found, when calculated from (theoretical) cross sections given by different authors. As a possible explanation of these discrepancies, it is indicated that the contribution of the optically disallowed transitions to the total cross sections can be very important, even for large E_1 . Further, a comparison with classical collision theories is made.

Introduction. For electron energies E_1 large enough to ensure the validity of the Bethe-Born approximation¹⁾, the cross sections Q for ionization and for optically allowed excitation of atoms can be written as^{2) 3)}:

$$Q = \frac{A}{E_1} \ln cE_1 \quad (1)$$

where A and c are constants for a given transition. The constant c gives information on the probability for moderate momentum transfers in exciting or ionizing collisions, and is defined in ref. 3. The constant A is equal to $4\pi a_0^2 R^2 f/U$ in the case of excitation, and is equal to $4\pi a_0^2 R M_i^2$ in the case of ionization. Here a_0 is the radius of the first Bohr orbit of hydrogen, R is the Rydberg energy (13.595 eV), f is the oscillator strength, U is the excitation energy and

$$M_i^2 = \int_{\nu_i}^{\infty} \frac{R}{E} \frac{df}{dE} dE \quad (2)$$

where df/dE is the oscillator density for transitions to the continuum²⁾³⁾, and U_i is the ionization energy of the atom.

The relations given above are valid for all atoms, with the restriction (for 1) that E_1 should be large enough. For atomic hydrogen we give some specific relations for f and M_i^2 . We indicate the initial states by quantum numbers n and l , and the final states by quantum numbers n' and l' for excitation and by i for ionization. Then for an optically allowed transition from state nl to state $n'l'$,

$$\frac{Rf}{U}(nl \rightarrow n'l') = \frac{1}{3} \frac{\max(l, l')}{2l+1} (\mathcal{R}_{nl}^{n'l'})^2 \quad (3)$$

where $\mathcal{R}_{nl}^{n'l'}$ is the radial (dipole) matrix element. Relation (3) is given in a slightly different notation by Bethe and Salpeter⁴⁾ and by Green *e.a.*⁵⁾. Further:

$$M_i^2(nl \rightarrow i) = \frac{1}{3} \langle nl | r^2 | nl \rangle - \sum_{n' \neq nl} \frac{Rf}{U}(nl \rightarrow n'l') \quad (4)$$

where r is the position vector of the atomic electron (in units a_0) and $\langle nl | r^2 | nl \rangle$ is the expectation value of r^2 for the nl state of hydrogen. Relation (4) is one of the sum rules for the oscillator strengths⁴⁾⁶⁾.

For transitions from initial states with $n = 1$ to 4, values of the oscillator strengths, values of the squares of the radial matrix elements (for $n' \neq n$ as well as $n' = n$) and values of $\langle nl | r^2 | nl \rangle$ are given by Bethe and Salpeter⁴⁾. More accurate values of the squares of the radial matrix elements are calculated (for $n' \neq n$) by Green *e.a.*⁵⁾ for transitions from initial states up to $n = 20$. Hence for $n' \neq n$ we used the values of $(\mathcal{R}_{nl}^{n'l'})^2$ given by Green *e.a.*, and for $n' = n$ we used those values of Bethe and Salpeter.

With the known values of $(\mathcal{R}_{nl}^{n'l'})^2$ and $\langle nl | r^2 | nl \rangle$, the values of M_i^2 can be found immediately. To obtain the values of c (or cU) for an excitation or ionization process, knowledge of one value of Q for a sufficiently large electron energy E_1 is necessary (see eq. 1). Unfortunately, many calculations of Born cross sections are only carried out for small and moderate electron energies.

1. *Excitation.* 1.1. Calculation of cU values for optically allowed transitions. For transitions from the ground state to the $n' = 2, 3, 4, 5$ and 6 levels, total Born cross sections (summed over l') are calculated by McCarroll⁷⁾. With McCarroll's cross sections for an electron energy*) $E_1 = 72.25 R$, we calculated cU values (eq. 1) which were in excellent agreement with values of $cR/4$ previously given by Bethe (ref. 1 p. 356). However calculation of cU values for optically allowed transitions from the total cross sections will result in cU values that are too large,

*) For all transitions considered in this paper, we used the cross sections corresponding to the largest electron energies for which data exist.

because the contributions of the optically disallowed transitions cannot be neglected. Massey⁸) has given partial Born cross sections (not summed over l'), which seem to be somewhat too small and only for the transition $1s \rightarrow 2p$ a value of Q is given for a sufficiently large electron energy (1000 eV). In our calculation of cU values for the transitions $1s \rightarrow n'p$, we used the total cross sections of McCarroll for $E_1 = 72.25 R$ diminished by 3.45%, which is the relative contribution (according to Massey) of the transitions $1s \rightarrow 2s$ to the total cross section $1s \rightarrow 2(s + p)$ for $E_1 = 1000$ eV. We note that this method is somewhat crude, because the contribution of the optically disallowed transitions to the total cross sections probably will be dependent on n' . For the transitions $1s \rightarrow 2p$, we find $cU = 1.26$ (see also note 3.3), while Bethe gives $cU = 1.49$, McCarroll's value of the total cross section for $E_1 = 72.25 R$ corresponds to $cU = 1.49$, and Massey's partial cross section for $E_1 = 100$ eV corresponds to $cU = 1.04$.

In five papers⁹⁻¹³), Milford and coworkers calculated partial Born cross sections for transitions from initial states with $n = 2, 3, 4, 5$ and 10 for $n' - n = 1$ and 2. For these transitions they also calculated values of the so called "momentum cutoff factor" K_c , where $(K_c a_0)^2 = cU^2/4R$. We simply used this relation for these transitions and assumed (which is important in the following) that the resulting cU values are the correct ones. This is probably justified as Milford and coworkers used Born cross sections for very large electron energies.

For transitions from the $2s$ state to the $n' = 3, 4, 5, 6$ and 7 levels, total (summed over l') Born cross sections are calculated by Boyd¹⁴). Partial Born cross sections and the K_c value for the transition $2s \rightarrow 3p$ are given in ref. 12. Comparison of total and partial cross sections for $E_1 = 9R$ shows that the total cross section ($2s \rightarrow 3$) should be diminished by 33.4% in order to get the correct cU value for $2s \rightarrow 3p$. To obtain the cU values for the transitions $2s \rightarrow n'p$ with $n' = 4, 5, 6$ and 7, the total cross sections for $E_1 = 9R$ were also diminished by 33.4%.

Total Born cross sections for transitions from the $2p$ state to the $n' = 3$ to 7 states are calculated by McCrea and McKirgan¹⁵). Partial Born cross sections and the K_c value for the transition $2p \rightarrow 3d$ are given by Scanlon *e.a.*¹²). By analogous comparison of total and partial cross sections for $E_1 = 9R$, we found that the cross section should be diminished by 7.8% to give the correct cU value for $2p \rightarrow 3d$. This was also done with the cross sections (also for $E_1 = 9R$) for the transitions $2p \rightarrow n'd$ with $n' = 4, 5, 6$ and 7. But in this case the resulting cU values do not seem to be very reliable (e.g. 4.54 for the transition $2p \rightarrow 4d$ and 17.3 for the transition $2p \rightarrow 7d$).

In table I, the resulting values of cU are listed, except for the transitions $2p \rightarrow n'd$ with $n' = 4$ to 7.

From table I it follows for transitions from state nl to state $n'l'$ (where

TABLE I

Values of cU for several optically allowed transitions in atomic hydrogen. The cU values obtained from the empirical eq. (5) are added in parenthesis							
trans.	cU		trans.	cU		trans.	cU
1s → 2p	1.26 *	(1.26)	5p → 6d	0.239	(0.245)	2s → 5p	1.039
2s → 3p	0.510	(0.510)	5d → 6f	0.295	(0.319)	2s → 6p	1.178
2p → 3d	1.096	(1.020)	5f → 6g	0.425	(0.442)	2s → 7p	1.276
3s → 4p	0.328	(0.330)	5g → 6h	0.696	(0.685)	3s → 5p	0.505
3p → 4d	0.503	(0.495)	10s → 11p	0.0974	(0.0979)	3p → 5d	0.938
3d → 4f	0.923	(0.884)	10(9) → 11(10)	0.426	(0.426)	3d → 5f	2.587
4s → 5p	0.244	(0.246)	1s → 3p	2.33 *		4s → 6p	0.383
4p → 5d	0.331	(0.328)	1s → 4p	2.75		4f → 6g	2.774
4d → 5f	0.459	(0.470)	1s → 5p	2.94		2p → 3s	0.566
4f → 5g	0.794	(0.773)	1s → 6p	3.08		3p → 4s	0.334
5s → 6p	0.189	(0.196)	2s → 4p	0.826		3d → 4p	1.394

* See note 3.3.

$l' = l + 1$ for most transitions) that:

- 1° cU increases with increasing l (n and $n' - n$ constant).
- 2° cU increases with increasing $n' - n$ (n and l constant).
- 3° cU decreases with increasing n ($n' - n$ and l constant).

Increasing cU values correspond with increasing probabilities for moderate momentum transfers (see ref. 3).

1.2. Empirical relations for the cU values. Milford¹⁶) has given a method for approximate calculation of Born cross sections for excitation of hydrogen from various states. For these calculations he gives a formula for approximate calculation of K_e values. This formula is somewhat crude because there is no dependence of K_e on the quantum number l . As can be seen from table I, the cU values (and so also the K_e values) depend on l . In ref. 11 a more accurate method to obtain K_e values graphically (by interpolation) is given for $n' - n$ equal to 1 or 2. We found that the cU values for the transitions with $n' - n = 1$ and $l' = l + 1$ are also accurately given by the following expression:

$$cU = 0.984 \left(n + 0.08 - \frac{0.3}{n} \right)^{-1} \left(1 - \frac{l}{n} + 0.18 \frac{l(l-1)}{n^2} \right)^{-1} \quad (5)$$

In table I, the cU values obtained with (5) are added in parenthesis.

1.3. Approximate calculation of excitation cross sections. Recently Kingston¹⁷) compared excitation cross sections for transitions from various initial states of atomic hydrogen calculated with Gryzinski's first (uncorrected¹⁸) formula¹⁹), with the corresponding Born cross sections calculated by Milford and coworkers. To this comparison we add the following example. The "classical" theory predicts that the total cross sections for excitation from the 3s to the $n' = 4$ states, from the 3p to the $n' = 4$

states and from the 3d to the $n' = 4$ states are essentially the same for large E_1 and are not very different for small E_1 . We note that the shape of the velocity distribution function of the atomic electron (which depends on l) has no influence (in "classical" theory) on the cross sections for large E_1 (see ref.18). In reality, for the allowed transitions both the Rf/U and the cU values increase with increasing l . From the calculations of McCoyd *e.a.*⁷⁾ it follows for $E_1 = 1361$ eV that $Q(3s \rightarrow 4) = 3.53\pi a_0^2$, $Q(3p \rightarrow 4) = 4.50\pi a_0^2$ and $Q(3d \rightarrow 4) = 6.615\pi a_0^2$. From the calculations of Kingston it follows for large E_1 , that the "classical" cross sections are much smaller than the Born cross sections. This is to be expected (see refs. 18 and 20), but as the Born cross sections are correct for large E_1 , this means that the "classical" cross sections are not correct. We believe that a (for instance empirical) method in which the cross sections are correlated with the optical oscillator strengths (squares of the radial matrix elements), will give better results (also for small E_1) than "classical" calculations. Such methods are given by Milford¹⁸⁾, Seaton²¹⁾ and the author³⁾.

The semi-empirical formula previously³⁾ given by the author is:

$$Q = AF \frac{(E_1 - U)}{E_1^2} \ln [1 + c(E_1 - U)] \quad (6)$$

where A is given in the introduction, $E_1 \geq U$ and

$$F = 1 + (1 - cU)(0.025 + 1.6/cU)(1 - U/E_1)(U/E_1)^{3/2}$$

In this previous paper³⁾ we restricted ourselves principally to ionization of atoms (then $U = U_i$), and in order to obtain a good agreement with experiment it was useful to introduce the factor F . F diminishes the cross sections for $cU > 1$ and enlarges the cross sections for $cU < 1$ (for $E_1 = U$ and $E_1 \gg U$ is $F = 1$). In ref. 3, excitation cross sections for the transition $H(1s) \rightarrow H(2p)$ calculated with (6) are also given. As for that calculation a cU value 1.04 was used, the influence of F on Q was no more than 1% for each E_1 . We note, as already pointed out in this paper, that a cU value 1.26 is probably more reliable (this gives a maximum change in Q by 8%) for the transition $H(1s) \rightarrow H(2p)$. To check the applicability of (6) for calculation of excitation cross sections, we calculated cross sections for several transitions between excited states of atomic hydrogen. From the comparison of these calculated cross sections with the corresponding Born cross sections, it follows that for excitation it probably will be more reliable (especially for $cU < 1$, as will be explained) to take $F = 1$. Thus we believe that excitation cross sections are more accurately given by.:

$$Q = A \frac{(E_1 - U)}{E_1^2} \ln [1 + c(E_1 - U)] \quad (7)$$

while ionization cross sections can better be calculated with (6). For com-

parison, Born cross sections and cross sections calculated with (7) are listed in table II. The listed Born cross sections are chosen from those given by Milford and coworkers, except for the transition $1s \rightarrow 5p$; for this transition the total cross section calculated by McCarroll is given.

TABLE II

Comparison of excitation cross sections for atomic hydrogen calculated with (7) and with the Born approximation				
trans.	U (eV)	E_1 (eV)	Q eq. (7) (πa_0^2)	Q Born (πa_0^2)
$1s \rightarrow 5p$	13.051	54.38	0.0257	0.0372
$3s \rightarrow 4p$	0.661	2.704	106	116
$3d \rightarrow 4f$	0.661	2.704	429	575
$3p \rightarrow 5d$	0.967	3.193	26.7	36.2
$4p \rightarrow 5d$	0.306	1.200	618	707
$4s \rightarrow 6p$	0.472	1.760	64.3	70
$4f \rightarrow 6g$	0.472	1.760	255	338
$5d \rightarrow 6f$	0.166	0.537	2446	3000
$5g \rightarrow 6h$	0.166	0.537	9002	12740

From table II it follows that (7) gives smaller cross sections than the Born approximation gives. This seems to be correct, as it is known that the Born approximation in general overestimates the cross sections for small E_1 . We note, that when the same calculations are carried out with (6) instead of (7), then for some transitions with small cU values (e.g. $3s \rightarrow 4p$ with $cU = 0.328$) the resulting cross sections are larger than the Born sections.

2. Ionization. 2.1. Calculation of M_i^2 values and cU_i values. For ionization of atomic hydrogen from the $1s$ state a value of $4/cR$ is given by Bethe (ref. 1 p. 359) and Born cross sections are given by Massey¹³). For ionization from the $2s$ and $2p$ states, Born cross sections are calculated by Omidvar and Sullivan²²). They also calculated Born cross sections in parabolic coordinates for initial states with $n' = 3$. These cross sections were not used for calculation of cU_i values, because the cross sections for ionization from initial states which are given in spherical coordinates cannot be evaluated directly from the cross sections for ionization from initial states which are given in parabolic coordinates. Stauffer and McDowell²³) also carried out calculations on ionization of H from various states and found lower cross sections than Omidvar and Sullivan have given. In particular, for ionization from the $2p$ state their cross sections are lower than for ionization from the $2s$ state in contradiction to Omidvar's results. These differences are probably due (as they have pointed out themselves) to the fact that in the calculations of Stauffer and McDowell, only the optically allowed transitions are included.

In table III, the calculated values of M_i^2 (eqs. 3 and 4), $M_i^2 U_i$ and cU_i (eq. 1) are listed. The cross sections used for calculation of the cU_i values are

TABLE III

Values for atomic hydrogen of M_i^2 , $M_i^2 U_i$, cU_i and the cross sections used for calculation of the cU_i value						
trans.	M_i^2	$M_i^2 U_i$ (eV)	cU_i	$Q(E_1)$ (πa_0^2)	E_1 (eV)	ref.
1s \rightarrow i	0.284	3.86	77	0.133*	1000	13
1s \rightarrow i	0.285 ¹⁾	3.87	83	—	—	1
2s \rightarrow i	0.823	2.80	77	3.83*	98.63	22
2s \rightarrow i	—	—	2.7	1.33 [†]	163.14	23
2p \rightarrow i	0.533	1.835	26300	4.1*	98.63	22
2p \rightarrow i	—	—	3.5	0.91 [†]	163.14	23
3s \rightarrow i	1.48	2.24	—	—	—	—
3p \rightarrow i	1.28	1.93	—	—	—	—
3d \rightarrow i	0.645	0.975	—	—	—	—

* total cross sections

† partial cross sections

also given in table III, together with the electron energies E_1 for which these cross sections are calculated. A reference is made to the authors who have calculated these cross sections.

2.2. The influence of optically disallowed transitions. The values of $M_i^2 U_i$ are given in table III for comparison with "classical" (two particle) collision theories. These "classical" theories predict (see ref. 3) that both the $M_i^2 U_i$ values and cU_i values should be the same for ionization from different states, provided that the optically disallowed transitions do not give an important contribution to the total cross sections. For large E_1 , the optically disallowed transitions are not important. For small E_1 , they can be important. To clarify this, we consider the ionization from the 2s and 2p states, for which the $M_i^2 U_i$ values are 2.80 and 1.835 eV respectively. We can conclude from these figures that the partial cross sections of the optically allowed transitions, are larger for ionization from the 2s state than from the 2p state. But the partial cross sections of the optically disallowed transitions are larger for ionization from the 2p state (see table III), such that the total cross sections are not very different for small E_1 (in agreement with "classical" theories). That such a compensation does not always occur, can be illustrated with another example. For ionization of Ar and Ne respectively, the second (uncorrected¹⁸) formula of Gryzinski²⁴) gives excellent agreement with experiment for Ar, while it overestimates the cross sections for Ne by more than a factor 2 for small E_1 (see ref. 25). For Ne, the $M_i^2 U_i$ value is small because the photoionization cross sections are small. So the partial cross sections of the allowed transitions are small. As the total cross sections for Ne are also very small (which follows from the comparison with "classical" theories), the disallowed transitions do not give a compensation for Ne.

The cU_i values 2.7 and 3.5 for the transitions 2s \rightarrow i and 2p \rightarrow i are small

compared to the other values given in table III, but if we compare then with the cU_i values obtained from the measurements of Schram *e.a.*²⁶⁾ for ionization of He, H₂, Ne, Ar, Kr, Xe, N₂ and O₂, they seem to be more reliable than the larger cU_i values. If we assume that all cross sections given in table III are correct, then the contributions of the optically disallowed transitions ($l' - l = 0, 2, 3, 4 \dots$) to the total cross sections are considerable, also for electron energies E_1 large compared to the ionization energies U_i . Recently Peach²⁷⁾ carried out calculations on ionization of H, He, Li and Be. From these calculations it also follows that the contributions of the disallowed transitions to the total cross sections can be very important (e.g. for Li and Be). Thus for some atoms (the rare gases) the disallowed transitions do not seem to be important at all, while for other atoms they seem to be very important.

2.3. Approximate calculation of ionization cross sections. From ref. 3 it follows that (6) gives good agreement with experiment for those ionization processes where the optically disallowed transitions are not very important. Therefore it is to be expected that (6) will give cross sections that are too small for those ionization processes in which the disallowed transitions are important. For instance for ionization of atomic hydrogen from the ground state there are two possibilities of which the second is more likely to be correct. 1°. The cU_i value 77 (or 83) is "correct" (by "correct" we indicate those cU_i values corresponding to the definition given in ref. 3, see also 3.2 and ref. 28), the disallowed transitions are not important and (6) gives good agreement with experiment (see ref. 3). 2°. The "correct" cU_i value is (much?) smaller than 77, the disallowed transitions are important and (6) will give too small cross sections.

3. Notes. 3.1. The problem of the different contributions of the optically disallowed transitions to the total cross sections, will be important only for ionization of atoms. Therefore, a method in which the electron impact ionization and excitation cross sections are correlated with the optical properties of the atom (ref. 3 and this paper), will in principle be more justifiable for (optically allowed) excitation than for ionization of atoms. However, for excitation the electron exchange process can give an important contribution to the excitation cross sections for small E_1 (near threshold).

3.2. The definition of c is given in ref. 3. There and in ref. 2, the dependence of c on the generalized oscillator strength $f(K)$ is explained only for optically allowed excitation of atoms. The definition of K_e , as used by Milford and coworkers, corresponds with this definition of c . Schram pointed out for ionization and excitation of atoms, that the c values derived from the total (optically allowed + disallowed) cross sections are essentially different from (and can be much larger than) the c values derived from the partial

(only optically allowed) cross sections. The cause of this discrepancy will be explained in a subsequent paper²⁸).

3.3. We note that Bates *et al.*²⁹ have given $K_c a_0$ values for the transitions $1s \rightarrow 2p$ and $1s \rightarrow 3p$ in atomic hydrogen. These values correspond with cU values 1.23 and 1.73, while our values are 1.26 and 2.33 respectively. The agreement for $1s \rightarrow 2p$ is reasonable. The disagreement for $1s \rightarrow 3p$ is probably due to the manner in which we have corrected for the disallowed transitions (see 1.1.).

3.4. The method of approximate calculation of excitation cross sections for electric dipole transitions as given in this paper (eq. 7), is similar to Seaton's impact parameter method³⁰ (I.P. method). Both methods require knowledge of the oscillator strength f . Both methods also agree in giving smaller cross sections (for small E_1) than the Born approximation gives. The differences are: *a.* the I.P. method is mathematically more complicated, *b.* it requires knowledge of the so called cutoff radius, while in the present method (eq. 7) the cutoff momentum K_c (cU) should be known, *c.* the I.P. method takes some account of the differences between strong and weak coupling.

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