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Cross-section and rate formulas for electron-impact ionization, excitation, deexcitation, and total depopulation of excited atoms

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For electron-induced ionization, excitation, and deexcitation, mainly from excited atomic states, a detailed analysis is presented of the dependence of the cross sections and rate coefficients on electron energy and temperature, and on atomic parameters. A wide energy range is covered, including sudden as well as adiabatic collisions. By combining the available experimental and theoretical information, a set of simple analytical formulas is constructed for the cross sections and rate coefficients of the processes mentioned, for the total depopulation, and for three-body recombination. The formulas account for large deviations from classical and semiclassical scaling, as found for excitation. They agree with experimental data and with the theories in their respective ranges of validity, but have a wider range of validity than the separate theories. The simple analytical form further facilitates the application in plasma modeling.

I. INTRODUCTION

Multistep excitation and ionization by electrons via higher levels become dominant processes at sufficiently large electron density in various plasmas, e.g., positive column and discharge-sustained excimer laser plasmas.[1-3] The reverse processes of three-body collisional recombination to higher levels and deexcitation between higher levels are dominant in low-temperature afterglow and recombination-laser plasmas.[4,5] In the modeling of these and other nonequilibrium plasmas, using, e.g., collisional-radiative models, it is therefore essential to know the cross sections or rates for these electronic transfer processes. Interest in this subject has recently further increased, because it is now possible to measure the total depopulation as well as the individual transfer rates using time-resolved laser-induced fluorescence spectroscopy.[6,7]

Many cross-section and rate formulas are available in literature for the various processes mentioned above. All formulas, however, have a limited range of validity and there are serious discrepancies among them, some of which have not yet been noted. In the following sections we give a survey and analysis of literature data, combine experimental results and the results of different theories into one set of simple analytical cross-section and rate formulas, and discuss the relation with other experimental and theoretical results. The purpose of the paper is thus to give practical formulas, which are accurate in a wide range of primary energies, including the adiabatic and sudden regimes, and which are applicable for many different transitions, e.g., from the ground state as well as from highly excited states. The paper does not present a new theory. Instead, the available (reliable) experiments and theories are combined in a semiempirical way.

We consider electronic $|p\rangle \rightarrow |n\rangle$ and $|p\rangle \rightarrow |i\rangle$ transitions in an atom, where p and n are the (effective) principal quantum numbers of initial and final states $|p\rangle$ and $|n\rangle$, and $|i\rangle$ denotes the ion ground state. We confine ourselves to states $|p\rangle$ and $|n\rangle$ with only one electron in the (outer) orbit, such as transitions between excited states or transitions from or to the ground state of one of the alkali atoms. The following notation is used: E_{pi} and $E_{pn}=E_n - E_p$ are the ionization, excitation (for $E_n > E_p$), and deexcitation ($E_n < E_p$) energies, E_e and E are the incident electron energy and the energy transfer to the atom, respectively, σ_{pi} and σ_{pn} are the ionization and (de)excitation cross sections, and K_{pi} and K_{pn} are the corresponding rate coefficients obtained by integrating the σ's over a Maxwellian electron energy distribution with temperature T_e.

II. SURVEY AND ANALYSIS OF LITERATURE DATA

A. Ionization

1. Binary-encounter and three-body classical collision theories

We make a distinction between two regimes of the incident electron energy.

i. $E_e > 4E_{pi}$. In this regime of sudden collisions, there is good agreement between the binary-encounter approximation,[8] abbreviated hereafter by BEA, and the Born and Bethe approximations (see Refs. 8–10), and also with the classical three-body theory of Abrines *et al.*[11] (see Figs. 6-6-1 to 6-6-5 in Ref. 8). There is a discrepancy in the high-energy behavior ($E_e \gg E_{pi}$), in that the Born and Bethe cross sections decrease with $E_e^{-1} \times (A \ln E_e$

$+B$), while the BEA and classical three-body cross sections decrease with E_e^{-1}. For the E_e values of interest here (<100 eV), the $A \ln E_e$ term, however, is small with respect to the B term for ionization from the higher excited states.

ii. $E_{pi} < E_e < 4E_{pi}$. In this regime the BEA cross sections are larger (up to about 40%) than those of the more accurate classical three-body theory (cross sections obtained with Monte Carlo trajectory calculations).[11,8] The difference is mainly due to the fact that the BEA does not take the interaction with the core of the atom into account.

In Ref. 8 it was shown that the symmetrical model of the BEA gives the best agreement with the Monte Carlo calculations and with experiment. When this model is applied to excited atoms, for which the kinetic energy of the electron in its orbital is equal to or is in good approximation equal to the binding energy, it yields the differential cross section per unit energy transfer:

$$\frac{d\sigma}{dE} = \frac{\pi e^4}{E_e + \alpha E_{pi}} \left(\frac{1}{E^2} + \frac{4E_{pi}}{3E^3} \right), \qquad (1)$$

where $\alpha = 2$ and $e^2 = 2a_0 \mathcal{R}$ (a_0: Bohr radius, $\mathcal{R}$: Rydberg energy). The total ionization cross section is obtained by integrating from E_{pi} to E_e, the range of possible energy transfers in the continuum. This yields

$$\sigma_{pi} = \frac{\pi e^4}{E_e + \alpha E_{pi}} \left(\frac{5}{3E_{pi}} - \frac{1}{E_e} - \frac{2E_{pi}}{3E_e^2} \right). \qquad (2)$$

The effect of electron exchange (interference) has not been included in Eqs. (1) and (2) because it severely complicates the formulas, while the ionization cross sections are only slightly reduced for excited atoms.[12] In Ref. 13, we replaced $\alpha = 2$ in Eq. (2) by $\alpha = 3.25$. This reduces the cross sections σ_{pi} for small E_e in such a way that the three-body cross sections by Abrines *et al.*[11] are reproduced within their statistical accuracy ($\approx 10\%$), while the correct BEA limit for large E_e remains unaltered. Equation (2) with $\alpha = 3.25$ thus gives accurate cross sections for ionization of highly excited atoms because the classical three-body theory is correct in this limit. Percival and Richards[14] have also suggested that a modification of the BEA formula be used, Eq. (2) with $\alpha = 3$, to represent the ionization cross sections.

Ton-That *et al.*[15] and Hyman[16] recently used more complicated versions of the BEA, including inner-shell ionization, which is important for heavier atoms at higher energies E_e, and including averaging over the momentum distribution of the atomic electrons, to calculate cross sections for ionization of excited atoms. Roy and Rai[17] used a similar more extended version of the BEA to calculate cross sections for ionization of the alkali atoms from the ground state. We have made a comparison between the σ_{pi} calculated using Eq. (2) with $\alpha = 3.25$ and the σ_{pi} from Ref. 15 to 17. The comparison shows very good agreement, typically within 10 to 20%. The symmetrical model of the BEA is also the basis for the calculations in Refs. 16 and 17.

2. Comparison with experiment

Recently, experimental cross sections have become available for ionization from the excited (metastable) states of H (Ref. 18), He (Ref. 19), and Ne and Ar (Ref. 20). When Eq. (2) with $\alpha = 3.25$ is applied to these cases, cross sections are obtained which are 10 to 30% higher than experiment for H$2S$, are 15 to 20% lower for He$\,2^3S$, overlap experiment for Ne*, and are 10 to 15% lower for Ar*. This is in surprisingly good agreement. The more involved BEA calculations of Refs. 15 and 16 are in similar good agreement with experiment. Excited (noble-gas) atoms are further similar to alkali atoms, which have also one loosely bound electron in their outer orbital. From threshold up to about 9 eV (Cs), 12 eV (Rb), and 15 eV (K) above threshold only outer-shell electron ejection occurs, and a comparison can be made between the experimental ionization cross sections for the alkali atoms[21] and those calculated using Eq. (2) with $\alpha = 3.25$. Mutual agreement is found again in a 5 to 30% range. The extended BEA calculation of Ref. 17 yields similar good agreement.

For the atoms considered, the measured and calculated positions and magnitudes of the first maxima of the ionization functions are listed in Table I, to illustrate the good agreement obtained.

In summary, Eq. (2) with $\alpha = 3.25$ is representative of both the classical three-body theory and the BEA in their ranges of validity, it describes the available experimental results fairly accurately, and thus gives in a simple analytic way reliable cross sections for ionization of excited atoms and ground-state atoms with only one loosely bound outer electron (see also Sec. IV).

B. Excitation, deexcitation, and total depopulation

1. Theory

In a review paper, Percival and Richards[14] discussed the different regions of validity of the BEA, classical perturbation theory, three-body Monte Carlo trajectory calculations, Born approximation, and combined theories. They confined themselves mainly to $|p\rangle \rightarrow |n\rangle$ transitions in the hydro-

TABLE I. Experimental (Expt) and calculated [BEA, Eq. (2) with $\alpha = 3.25$] positions and magnitudes of the first maxima of the ionization functions.

	Expt		BEA	
Atom	E_e (eV)	σ_{pi} (10^{-16} cm^2)	$E_e = 2.71 E_{pi}$ (eV)	σ_{pi} (10^{-16} cm^2)
H 2S	13.0 ± 2	9.1 ± 2 [a]	9.2	11.4
He 2^3S	12.6 ± 1	7.1 ± 0.4 [b]	12.9	5.8
Ne*	14.0 ± 2	5.5 ± 0.4 [c]	13.4	5.4
Ar*	12.7 ± 3	8.5 ± 0.4 [c]	11.4	7.4
Li	14.1 ± 1	4.3 ± 0.2 [d]	14.6	4.5
Na	14.0 ± 1	6.8 ± 0.4 [d]	13.9	5.0
K	9.3 ± 0.8	7.8 ± 0.4 [d]	11.8	7.0
Rb	10.3 ± 1	8.2 ± 0.4 [d]	11.3	7.6
Cs	10.1 ± 1	7.1 ± 0.4 [d]	10.6	8.7

[a] Reference 18.
[b] Reference 19.
[c] Reference 20.
[d] Reference 21.

gen atom, with p and $n \geq 5$, and to primary energies $E_e > 4E_{pi}$. For this energy range they presented an empirical formula, fitted to the results of the combined theory.[14,22] Johnson[23] has also given analytic cross-section and rate formulas for transitions in atomic hydrogen. Johnson's cross sections are based on high-energy approximations (impact parameter, Born); he accounts, in an empirical way, for the reduction of the cross sections at low energies, due to adiabatic collisions. Percival and Richards[24] more recently presented a theory for $|pl\rangle \rightarrow |nl'\rangle$ transitions in highly excited nonhydrogenic atoms (also for high primary energies). Mansbach and Keck[25] further performed classical Monte Carlo trajectory calculations for excitation and ionization by thermal electrons. This latter work is complementary to that reported in Refs. 8–11, 14, and 22–24 in that Mansbach and Keck mainly confined themselves to thermal electrons and adiabatic collisions, E_e and $kT_e < E_{pi}$, while the other theories are mainly valid for $E_e > E_{pi}$. Mansbach and Keck did not give cross-section formulas, but presented their results instead in terms of simple analytical formulas for the rate coefficients. Because these formulas will be used in our analysis we reproduce them below [Eqs. (3) to (7)]. For excitation and ionization with energy transfer $\epsilon = E/kT_e$ (>0),

$$\frac{dK}{d\epsilon} = \frac{9.56 \times 10^{-6}}{(kT_e)^{1.5} \epsilon_{pi}^{2.33}} \exp(-\epsilon) , \tag{3}$$

where $\epsilon_{pi} = E_{pi}/kT_e$ and where $dK/d\epsilon$ is given in cm^3 s^{-1}, when kT_e is expressed in eV. For deexcitation ($\epsilon < 0$)

$$\frac{dK}{d\epsilon} = \frac{9.56 \times 10^{-6} \epsilon_{pi}^{2.5}}{(kT_e)^{1.5} (\epsilon_{pi} - \epsilon)^{4.83}} . \tag{4}$$

The total electronic transfer rate coefficient for level p was obtained by integrating over all possible energy transfers[25]:

$$K_p = \int_{-\epsilon_{op}}^{\infty} (dK/d\epsilon) d\epsilon , \tag{5}$$

where $\epsilon_{op} = E_{op}/kT_e$ and $E_{op} = E_p - E_o = E_p$ and $|0\rangle$ is the ground state of the atom. For highly excited states, $E_{oi} \gg E_{pi}$ and

$$K_p = \frac{9.56 \times 10^{-6}}{(kT_e)^{1.5} \epsilon_{pi}^{2.33}} \left(1 + \frac{\epsilon_{pi}}{3.83}\right) \tag{6}$$

in cm^3 s^{-1} with kT_e in eV. Equation (6) contains the contributions of excitation, deexcitation, and ionization. The ionization rate coefficient is obtained separately by integrating Eq. (3) from ϵ_{pi} to ∞. This yields

$$K_{pi} = \frac{9.56 \times 10^{-6}}{(kT_e)^{1.5} \epsilon_{pi}^{2.33}} \exp(-\epsilon_{pi}) . \tag{7}$$

2. *Experiment (excited atoms)*

For excited atoms, the most useful information for the present analysis is available in terms of rate coefficients. In a He afterglow plasma, with temperatures T_e in the range from 300 to 6000 K, Delpech *et al.*[6,7] recently measured total (depopulation) and individual electron-induced transfer rates from triplet levels with $p = 8$–17 to triplet levels $n = 5$–17. They also made a comparison with the rate coefficients K_p and K_{pn} calculated with

(i) the Mansbach and Keck formulas,
(ii) Johnson's formulas, and
(iii) BEA formulas given by Gryzinski.[26]

Gryzinski's rates agree within a factor of 1.5 to 2 with those obtained when using Eq. (1); see also Ref. 8.

The total Mansbach and Keck and the Johnson rate coefficients K_p both overestimate experiment by about 50 to 60%, while Delpech *et al.* claim their experimental data to be accurate within 30%. Gryzinski's K_p overestimate experiment by one ($p = 8$) to more than two ($p \geq 14$) orders of magnitude for $T_e \approx 390$ K. This latter discrepancy is not too surprising because the BEA, Born, and similar higher-energy ($E_e > 4E_{pi}$) approximations are not valid for adiabatic collisions (E_e and $E_{pn} < E_{pi}$). These approximations, and most of the semiempirical formulas fitted to the BEA or Born approximation in the high-energy limit, in general grossly overestimate the cross sections in the adiabatic regime (Refs. 6, 7, 14, and 22 to 25). The mutual agreement between the total electronic transfer rates of Mansbach and Keck and of Johnson is further fortuitous, because the individual transfer rates are very different.[7] Johnson's rates are larger for small energy transfer, $|n-p|=1$, while the Mansbach-Keck rates are larger for $|n-p|\geq 2$ or 3. For $p=10$ and $T_e=2000$ K, the contribution of ionization to K_p^- is, moreover, about 50% according to Mansbach and Keck, while it is negligible according to Johnson.[6]

3. Extension to ground-state atoms

The latter results, in addition to the theoretical work referred to, clearly show that the situation for excitation is considerably more complex than for ionization. For ionization, E_e is always larger than E_{pi}, one theory (the classical three-body theory) is valid for all E_e, and one simple analytic formula, Eq. (2), applies for this whole region. For excitation, E_{pn} is much smaller than E_{pi} for large p and small $|n-p|$. E_e may thus be much larger as well as much smaller than E_{pi}, and still give rise to excitation. Different theories apply in these different regimes and accurate cross sections are available[14,22] only for $E_e > 4E_{pi}$ and $p, n \geq 5$, at least for transitions from the excited states. The most relevant information about the adiabatic regime $E_e < E_{pi}$ is available only in terms of rate coefficients.[6,7,25] Since we wish to construct cross-section and rate formulas that are valid for small and large primary energies and energy transfers, including small as well as large values of p, n, and $s=|n-p|$, more information is needed. For this purpose, the H $1S\rightarrow 2(S+P)$, Li $2S\rightarrow 2P$, Na $3S\rightarrow 3P$, K $4S\rightarrow 4P$, Rb $5S\rightarrow 5P$, and Cs $6S\rightarrow 6P$ transitions are probably most suitable because of the following.

(i) Accurate experimental cross sections are available for these transitions, from threshold to high energies.[27-30]

(ii) There is also one electron in the outer orbital of the atom in the ground state, in which respect these atoms are most similar to excited states.

(iii) The excitation energies for the first resonance transitions in the alkali atoms are also quite small.

(iv) For H the sum of the $1S\rightarrow 2S$ and $1S\rightarrow 2P$ cross sections is available, while the lowest P states of the alkali atoms are well separated from the S and D states. Hence, in both cases we do not have to worry about the separation into final states with different angular momentum.

III. CROSS-SECTION AND RATE COEFFICIENT FORMULAS

In this section, Eqs. (1) to (7), the data referred to[6,7,14,22-25,27-30], and the well-known asymptotic high-energy limit of the excitation cross sections[31] will be used to construct more general cross-section and rate formulas.

A. Ionization

Equation (2) with $\alpha=3.25$ was found already to give accurate cross sections for excited atoms and ground-state alkali atoms. In order to get a rate formula, we numerically integrated Eq. (2) over a Maxwellian electron energy distribution for a wide temperature range. The resulting ionization rate coefficients K_{pi}, plotted in Fig. 1 for the special case $E_{pi}=1$ eV, are thus expected to be accurate too. The corresponding K_{pi} according to the Mansbach and Keck formula, Eq. (7), are also shown in Fig. 1. These coefficients are much too large at large kT_e, which also explains the discrepancy with Johnson's rates (Ref. 6). Consequently the extrapolation made by Mansbach and Keck from the adiabatic regime to that of sudden collisions is incorrect. For small values of kT_e, however, there is rather good agreement between the two sets of rate coefficients in Fig. 1. In constructing an analytic semiempirical formula for the rate coefficients we use Eq. (7) as the small kT_e limit, while the limit for large kT_e follows directly from Eq. (2), by omitting the αE_{pi} term (large E_e) and integrating over a Maxwell distribution. This gives an analytical solution. By dividing Eq. (7) by $[1+(a/\epsilon_{pi})^b+(1.23/\epsilon_{pi})^{1.33}]$, with adjustable a and

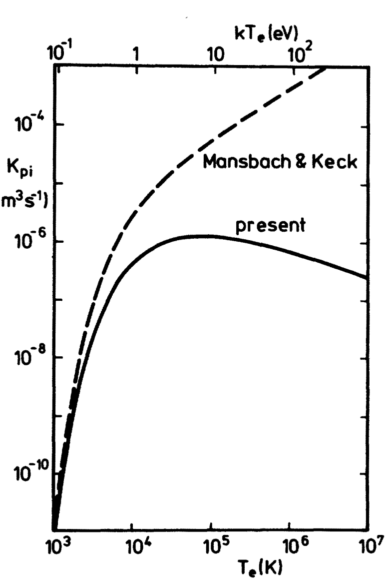

FIG. 1. Ionization rate coefficients K_{pi} for $E_{pi}=1$ eV, according to Mansbach and Keck [Eq. (7) and Ref. 25] and according to the present Eq. (8). The K_{pi} obtained by numerical integration of Eq. (2) are indistinguishable from those obtained from Eq. (8).

b, and $0 < b < 1.33$, the Mansbach and Keck formula is reproduced for $\epsilon_{pi} \gg 1$ ($E_{pi} \gg kT_e$) and the analytical BEA limit is obtained for $\epsilon_{pi} \ll 1$. With $a = 11.3$ and $b = 0.61$, we find that the K_{pi}'s obtained by numerical integration of Eq. (2), with $\alpha = 3.25$, are reproduced within 1% for $kT_e > 0.2E_{pi}$ and within 10% for $kT_e > 0.05E_{pi}$, while a value of K_{pi} only 30% smaller than the numerical value is obtained at $kT_e = 0.01E_{pi}$. These values of a and b thus yield

$$K_{pi} = \frac{9.56 \times 10^{-6}(kT_e)^{-1.5}\exp(-\epsilon_{pi})}{\epsilon_{pi}^{2.33} + 4.38\epsilon_{pi}^{1.72} + 1.32\epsilon_{pi}} . \qquad (8)$$

For ionization from excited states we suggest the use of Eq. (8) for all values of kT_e.

B. Three-body recombination

In equilibrium $n_e^3\alpha_{ip} = n_p n_e K_{pi}$, where n_e and n_p are the electron density and the density of atoms in the $|p\rangle$ state, and α_{ip} is the three-body (ion plus $2e$) recombination coefficient. Using the Saha equation and Eq. (8) we obtain

$$\alpha_{ip} = \frac{3.17 \times 10^{-27}(kT_e)^{-3}(g_p/g_i)}{\epsilon_{pi}^{2.33} + 4.38\epsilon_{pi}^{1.72} + 1.32\epsilon_{pi}} , \qquad (9)$$

where g_p and g_i are the statisical weights of level $|p\rangle$ and of the ion ground state; α_{ip} has the dimension $\mathrm{cm}^6\,\mathrm{s}^{-1}$ when kT_e is expressed in eV. For the larger values of kT_e, α_{ip} is also much smaller than the Mansbach and Keck recombination coefficient. For small values of kT_e these coefficients tend to the same limit.

C. Excitation

1. Cross sections

a. The high-energy limit. The high-energy limit is given by the Bethe formula,[31]

$$\sigma_{pn} = \frac{2\pi a_0^2 \mathcal{R}}{E_e}\left[A_{pn}\ln\frac{E_e}{2\mathcal{R}} + B_{pn}\right], \qquad (10)$$

where

$$A_{pn} = (2\mathcal{R}/E_{pn})f_{pn} , \qquad (11)$$

f_{pn} being the absorption oscillator strength and $\mathcal{R}$ again the Rydberg energy (13.6 eV). B_{pn}/A_{pn} can be expressed in terms of integrals containing $f_{pn}(q)/f_{pn}$, where the variable q is the momentum transfer to the atom and $f_{pn}(q)$ is the generalized oscillator strength.[31] B_{pn} can also be obtained from the measured or calculated (Born, Bethe) high-energy limit of the cross sections.

For the hydrogen atom, Johnson[23] gives an analytical representation for f_{pn} which reproduces the known oscillator strengths for $1 \leq p \leq 19$ and $p < n \leq 20$ (Ref. 32) within 0.5%, and for $20 \leq p \leq 49$ and $p < n \leq 50$ (Ref. 33) within 1%. We therefore use Johnson's formulas for f_{pn} for all p and n. The analytical formulas for B_{pn} given by Johnson[23] are less accurate for the larger p values, while those in Refs. 14, 22, and 34 are inaccurate for large values of $n - p$. Using the B_{pn} values calculated by Matsuzawa,[10] Kingston and Lauer,[35] and Omidvar,[36] and the BEA limit for $n - p \gg p \gg 1$, we therefore construct a more accurate analytical expression valid for all p and n:

$$B_{pn} = \frac{4\mathcal{R}^2}{n^3}\left(\frac{1}{E_{pn}^2} + \frac{4E_{pi}}{3E_{pn}^3} + b_p\frac{E_{pi}^2}{E_{pn}^4}\right) , \qquad (12)$$

where

$$b_p = \frac{1.4\ln p}{p} - \frac{0.7}{p} - \frac{0.51}{p^2} + \frac{1.16}{p^3} - \frac{0.55}{p^4} . \qquad (13)$$

Here, b_p is more accurate than the b_p of Johnson,[23] but Eq. (12) is otherwise the same. In the fitting procedure it was found to be essential to introduce a logarithmic term in Eq. (13). For $n - p \gtrsim p \gg 1$, the last term in Eq. (12) is negligible, $B_{pn} \gg A_{pn}$, and Eqs. (10) and (12) yield cross sections that are identical to those obtained from the BEA, as should be the case. The corresponding BEA cross sections are obtained from Eq. (1) by dividing by the final-state level density per unit energy E, $n^3/2\mathcal{R}$, and by taking the high-energy limit $E_e \gg \alpha E_{pi}$. For all transitions in the hydrogen atom we have now defined the A_{pn} and B_{pn} to be used.

For the first resonance transitions in the alkali atoms, the f_{pn} values are well known (see, e.g., Refs. 28 to 30): 0.75 (Li), 0.98 (Na), 1.00 (K), 1.02 (Rb), and 1.05 (Cs). The B_{pn} values are obtained here from the high-energy limit of the cross sections[28-30] (in Refs. 28–30 Born cross sections and experimental values of f_{pn} were used for normalization).

b. Extrapolation to lower energies. Equations (201) to (215) of Percival *et al.*[14] (also Ref. 22) given an extrapolation of the cross sections for $p \to n$ ($n > p \geq 5$) transitions in the H atom down to $E_e = 4E_{pi}$. Equation (29) of Johnson[23] yields σ_{pn} for all values of E_e, but the semiempirical extrapolation towards small E_e is based on the impact-parameter approximation, which is not valid for adiabatic collisions. The discrepancies between Johnson's σ_{pn} and those of Percival and Gee *et al.* are quite large (by factors of 2 for the $5 \to 6$ and $10 \to 11$ transitions up to a factor of 10 for the $50 \to 60$ transition). Moreover, Johnson's σ_{pn} are negative close to threshold for $n - p = 1$ and $p \gtrsim 20$. For these reasons we make a different extrapolation.

In the present extrapolation Eq. (10) is also used as a starting point. At low energies this formula has two deficiencies.

(i) $A_{pn}\ln(E_e/2\mathcal{R})+B_{pn}$ becomes negative for large p and small $s=|n-p|$. A remedy is to replace $\ln(E_e/2\mathcal{R})$ by $\ln(E_e/2\mathcal{R}+\delta_{pn})$, with $\ln(E_{pn}/2\mathcal{R}+\delta_{pn}) \geqslant -B_{pn}/A_{pn}$.

(ii) At larger E_e, but still in the adiabatic regime, σ_{pn} is much too large and has its maximum at small values of E_e. According to Percival *et al.*, the σ_{pn} instead have their maximum at $E_e > 2E_{pi}$, and for small $s=|n-p|$ and large p even at higher E_e. This energy dependence can be reproduced by replacing E_e in the denominator of Eq. (10) by $E_e+\gamma_{pn}$.

By simultaneously fitting to the experimental σ_{pn} [H$1S\rightarrow 2(S+P)$ and alkali resonance transitions] and to the theoretical σ_{pn} (highly excited H atoms with $p=5$ to 50 and $s=1$ to 10) we obtain a suitable function:

$$\sigma_{pn}=\frac{2\pi a_0^2\mathcal{R}}{E_e+\gamma_{pn}}\left[A_{pn}\ln\left(\frac{E_e}{2\mathcal{R}}+\delta_{pn}\right)+B_{pn}\right], \tag{14}$$

where

$$\delta_{pn}=\exp\left(-\frac{B_{pn}}{A_{pn}}\right)-0.4\,\frac{E_{pn}}{\mathcal{R}} \tag{15}$$

and

$$\gamma_{pn}=\mathcal{R}\left[8+23\left(\frac{s}{p}\right)^2\right]\left(8+1.1ns+\frac{0.8}{s^2}+0.4\,\frac{n^{1.5}}{s^{0.5}}\,|s-1|\right)^{-1}. \tag{16}$$

For the hydrogen atom p and n are the principal quantum numbers. For other atoms p and n are the effective quantum numbers $p=(\mathcal{R}/E_{pi})^{0.5}$ and $n=(\mathcal{R}/E_{ni})^{0.5}$. The complicated structure of Eq. (16) results from the fact that so many transitions are included in the fitting procedure. Note, however, that δ_{pn} and γ_{pn} only depend on atomic parameters and not on E_e. Equation (14) gives cross sections that are finite at threshold. This reflects the fact that many cross sections are indeed finite at threshold (H atom, Ref. 37) and that other σ_{pn} rise very rapidly above E_{pn}.

Figures 2-5 give a comparison between some of the σ_{pn} calculated with Eq. (14) and the experimental and theoretical σ_{pn} used as reference. Good agreement, typically within 5 to 20%, is obtained for all transitions studied, i.e., for the first resonance transitions in the alkali atoms, and for $|p\rangle\rightarrow|n\rangle$ to transitions in the H atom, with $p\geqslant 5$. For some of the resonance transitions in the alkali atoms larger discrepancies are found, but only very close to threshold ($E_e/E_{pn}<1.2$ to 1.5). For a different type of transition between excited states ($|pl\rangle\rightarrow|nl'\rangle$), the $10S\rightarrow 10P$ transition in Na, the σ_{pn} obtained with Eq. (14) agree within a factor of 2 with the theoretical σ_{pn} from Ref. 24 above $E_e=20E_{pn}$ and within a factor of 3 above $5E_{pn}$. For the $11S\rightarrow 12P$ transition in Na there is agreement within a factor of 2 only above $200E_{pn}$, the σ_{pn} according to Ref. 24 decrease much more rapidly with decreasing E_e. When the formula by Gee *et al.*[22] with experimental A_{pn} values is further used for the H$1S\rightarrow 2(S+P)$, Na$3S\rightarrow 3P$, and Cs $6S\rightarrow 6P$ transitions, cross sections are obtained

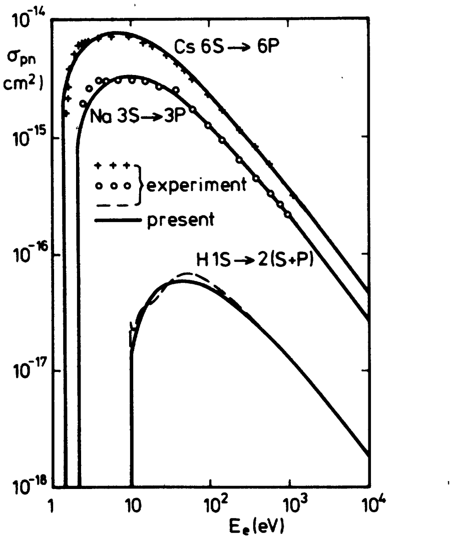

FIG. 2. Experimental (Refs. 27, 28, and 30) and present [Eq. (14)] cross sections σ_{pn} for the H$1S\rightarrow 2(S+P)$, Na$3S\rightarrow 3P$, and Cs $6S\rightarrow 6P$ transitions induced by electron impact.

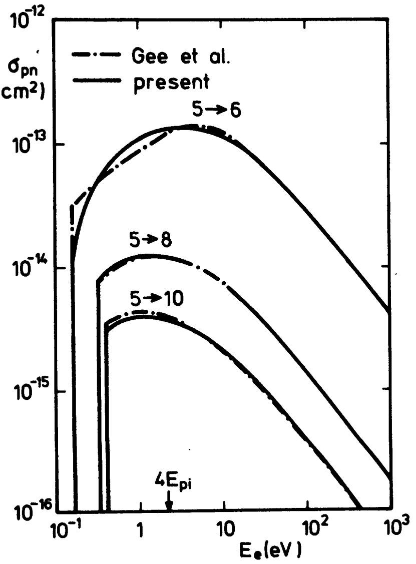

FIG. 3. Electron-impact excitation cross sections for atomic hydrogen ($p=5$) according to Gee *et al.* (Refs. 22 and 14) and according to the present Eq. (14). Gee *et al.* claim their σ_{pn} to be accurate above $E_e=4E_{pi}$.

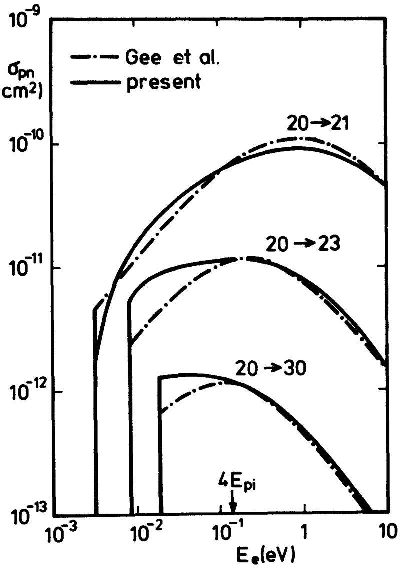

FIG. 4. As in Fig. 3, for the initial-state H $p = 20$.

which are either too large by 50 to 100% (H) or too small by 30 to 60% (Na and Cs) in the energy range up to $E_e = 100$ eV.

2. *Rate coefficients*

Numerical (70-point Laguerre) integration of Eq. (14) over a Maxwell distribution, for temperatures T_e varying in a wide range, yields a set of rate coefficients. It is convenient also to have an analytical formula for these coefficients. An empirical formula which represents the numerical rate coefficients is

$$K_{pn} = \frac{1.6\times 10^{-7}(kT_e)^{0.5}}{kT_e + \Gamma_{pn}} \exp(-\epsilon_{pn}) \times\left[A_{pn}\ln\left(\frac{0.3kT_e}{\mathcal{R}} + \Delta_{pn}\right) + B_{pn}\right], \tag{17}$$

in cm^3 s^{-1}, with kT_e and $\mathcal{R}$ in eV,

$$\Delta_{pn} = \exp\left(-\frac{B_{pn}}{A_{pn}}\right) + \frac{0.06s^2}{np^2}, \tag{18}$$

and

$$\Gamma_{pn} = \mathcal{R}\ln\left(1 + \frac{p^3kT_e}{\mathcal{R}}\right)\left[3 + 11\left(\frac{s}{p}\right)^2\right] \times\left(6 + 1.6ns + \frac{0.3}{s^2} + 0.8\frac{n^{1.5}}{s^{0.5}}\,|s-0.6|\right)^{-1}. \tag{19}$$

Equation (17) gives the correct large kT_e limit. For $kT_e \approx 1$ eV it reproduces the numerical rates within 1 to 10%, and for $kT_e \approx 0.1$ eV it still reproduces these rates within 5 to 20%, except for the alkali atoms where the rates according to Eq. (17) are 30 to 40% lower than the numerical rates at 0.1 eV. In this latter case, however, the numerical rates obtained from Eq. (14) are too large at small kT_e and Eq. (17) gives better agreement with experiment. In a wide temperature range, Eq. (17) is thus representative of the medium- and high-energy theoretical cross sections of excited hydrogen atoms[14,22] as well as for the experimental cross sections for H 1 → 2 and for the first resonance transitions in the alkali atoms.

For several transitions, Figs. 6–8 show a comparison between the present K_{pn}, Eq. (17), the Mansbach and Keck (MK) K_{pn}, and the K_{pn} according to Gee *et al.*[22] (GP). The MK K_{pn}, derived for adiabatic collisions, is obtained[25] from Eq. (3) by replacing ϵ by ϵ_{pn} and by dividing $dK/d\epsilon$ by the final-state level density per unit ϵ: $(R/kT_e)^{0.5}/2\epsilon_{ni}^{1.5}$. This yields

$$K_{pn} = \frac{5.19\times10^{-6}\epsilon_{ni}^{1.5}}{kT_e\epsilon_{pi}^{2.33}} \exp(-\epsilon_{pn}). \tag{20}$$

In the comparison we also apply the GP K_{pn} quite outside the regime of sudden collisions where Gee *et al.* claim their formula to be accurate. The comparison shows the following.

(i) In the regime of sudden collisions, $kT_e \gtrsim 4E_{pi}$, the MK formula leads to much too large rates for large $s = |n-p|$, just as for ionization, whereas the MK rates are much too small for small s and not too small p.

(ii) In the adiabatic regime, towards small values of T_e, the MK rates decrease less rapidly than those obtained from Eq. (17). For the ground-state H and alkali atoms, Eq. (17) represents experiment, and Eq. (20) overestimates the rates for

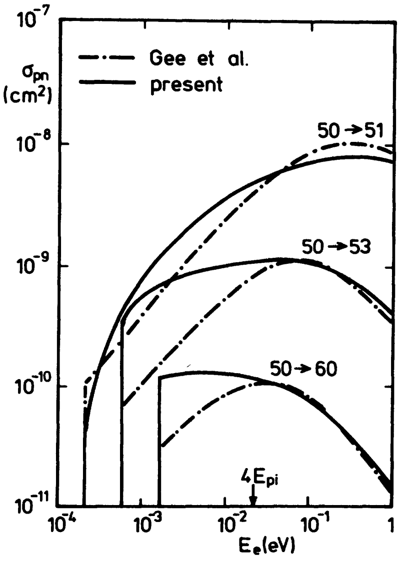

FIG. 5. As in Fig. 3, for the initial-state H $p = 50$.

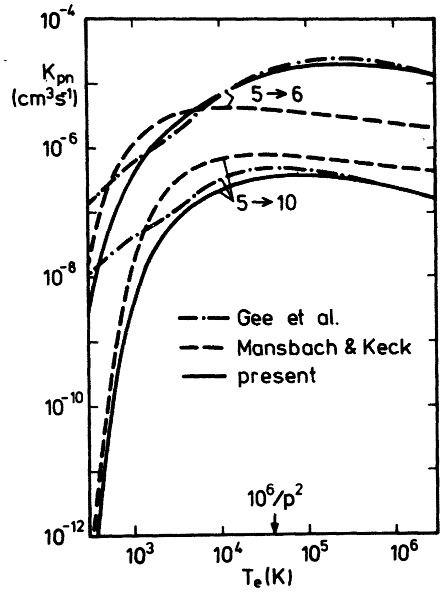

FIG. 6. Rate coefficients K_{pn} for electron-impact excitation of H $p=5$ according to Gee *et al.* (Ref. 22), Mansbach and Keck [Eq. (20) and Ref. 25), and the present Eq. (17). Gee *et al.* claim their K_{pn} to be accurate above $T_e=10^6/p^2$ (in K).

these transitions close to threshold. In this case, however, the level spacing exceeds kT_e and a classical theory should not be expected to yield good results. For the excited atoms the level spacing is small and it is not clear from this comparison which formula is to be preferred in the regime of adiabatic collisions (see also Secs. IIID and IIIE).

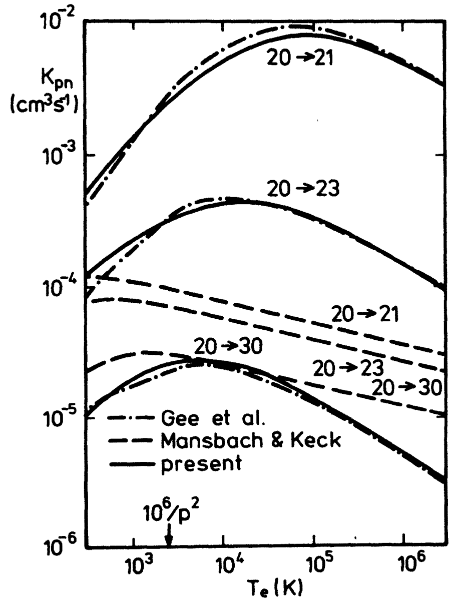

FIG. 7. As in Fig. 6, for the initial-state H $p=20$.

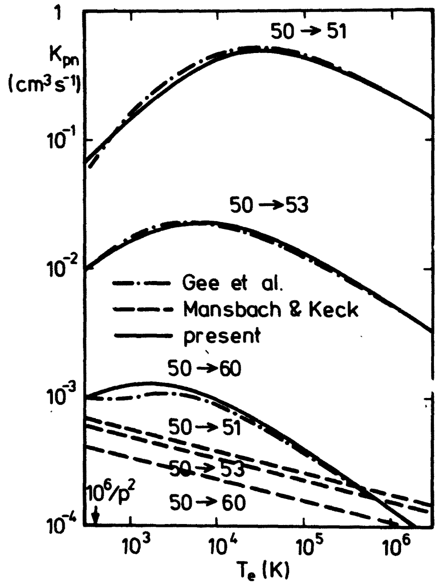

FIG. 8. As in Fig. 6, for the initial-state H $p=50$.

(iii) At large T_e the K_{pn}, according to Gee *et al.* and according to Eq. (17), agree very well, as should be the case since the former K_{pn} were used as a reference in the fitting procedure.

D. Deexcitation

Applying detailed balancing to the cross-section formula for excitation, Eq. (14), yields the cross-section formula for $|p\rangle \rightarrow |n\rangle$ deexcitation,

$$\sigma_{pn}=\frac{2\pi a_0^2\mathcal{R}g_n/g_p}{E_e+\gamma_{np}}\left[A_{np}\ln\left(\frac{E_e}{2\mathcal{R}}+\delta_{np}\right)+B_{np}\right], \quad (21)$$

where g_p and g_n are the statistical weights of levels $|p\rangle$ and $|n\rangle$,

$$\delta_{np}=\exp\left(-\frac{B_{np}}{A_{np}}\right)+0.1\frac{E_{np}}{\mathcal{R}}, \quad (22)$$

and

$$\gamma_{np}=\mathcal{R}\left[8+23\left(\frac{s}{n}\right)^2\right]\left(1+\frac{E_{np}}{E_e}\right)^{-1} \times\left(8+1.1ps+\frac{0.8}{s^2}+0.4\frac{p^{1.5}}{s^{0.5}}|s-1|\right)^{-1}. \quad (23)$$

Applying detailed balancing to Eq. (17) yields the rate coefficient for deexcitating by electrons,

$$K_{pn}=\frac{1.6\times10^{-7}(kT_e)^{0.5}g_n/g_p}{kT_e+\Gamma_{np}} \times\left[A_{np}\ln\left(\frac{0.3kT_e}{\mathcal{R}}+\Delta_{np}\right)+B_{np}\right], \quad (24)$$

where Δ_{np} and Γ_{np} are obtained from Δ_{pn} and Γ_{pn}

by interchanging p and n. Equation (20) similarly leads to the Mansbach and Keck formula

$$K_{pn}=\frac{5.19\times10^{-6}\epsilon_{pi}^{1.5}}{kT_e\epsilon_{ni}^{2.33}}\frac{g_n}{g_p}. \tag{25}$$

Figure 9 shows a comparison between the K_{pn}, for excitation as well as for deexcitation, obtained with

(i) the present formulas (17) and (24),
(ii) the MK formulas (20) and (25),
(iii) the GP formulas (Ref. 22), and
(iv) the K_{pn} measured by Devos *et al.*[7] for He $p=13$ at $T_e\approx300$ K.

The total depopulation rate coefficient K_p [Eq. (27)] is plotted too at $n=p=13$. The present K_p gives the best agreement with experiment, while for the individual K_{pn} better agreement is obtained with the MK formula. Owing to the relatively large error margins in the measured K_{pn}, it is difficult, however, to come to a conclusion about which formula is to be preferred in this regime. The formula of Gee *et al.*[22] is used here at $T_e=300$ K, quite outside the regime $T_e>10^6/[\min(p,n)]^2$; this yields $T_e>5000$ K for excitation, where it is claimed to be accurate. In view of this, the agreement with experiment is very good for excitation. For deexcitation, however, the GP K_{pn} first decrease with decreasing n and next increase by many orders of magnitude, leading to very unrealistic values of K_{pn} and K_p.

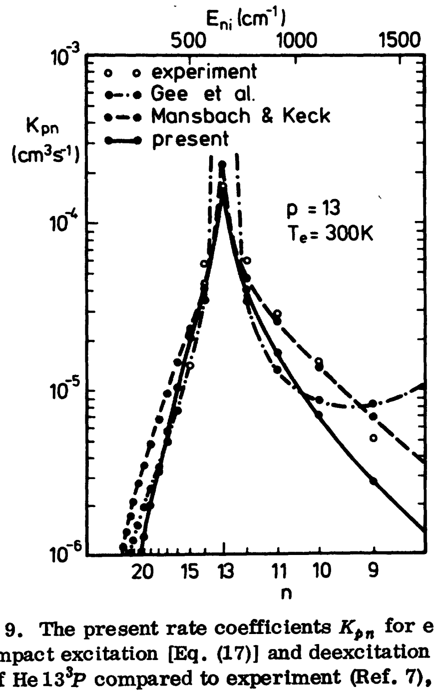

FIG. 9. The present rate coefficients K_{pn} for electron-impact excitation [Eq. (17)] and deexcitation [Eq. (24)] of He 13^3P compared to experiment (Ref. 7), Mansbach and Keck [(Eqs. (20) and (25)], and Gee *et al.* (Ref. 22). The formulas of Gee *et al.* are used here outside the range of T_e values for which these are claimed to be accurate. The total rate coefficients K_p are plotted at $n=p=13$.

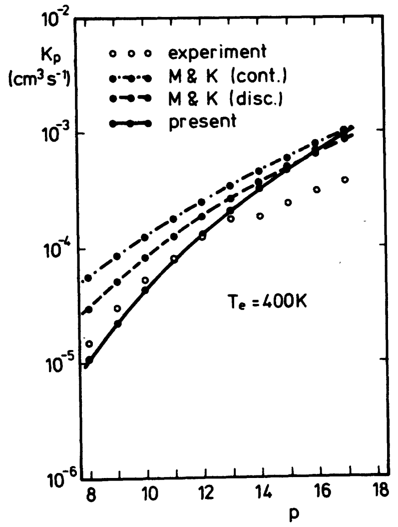

FIG. 10. The total electron-induced depopulation rate coefficients K_p of He 8^3P up to 17^3P according to experiment (Refs. 6 and 7), Mansbach and Keck with the discrete level structure taken into account [Eqs. (7), (20), (25), and (27)], Mansbach and Keck for a continuum of states [Eq. (6)], and according to the present Eqs. (8), (17), (24), and (27).

E. Total depopulation

The cross sections and rate coefficients for total depopulation by electron collisions are given by

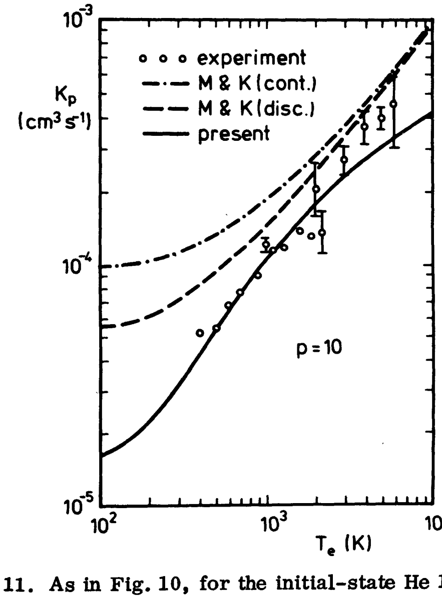

FIG. 11. As in Fig. 10, for the initial-state He 10^3P.

$$\sigma_p = \sigma_{pi} + \sum_{n\neq p} \sigma_{pn} \tag{26}$$

and

$$K_p = K_{pi} + \sum_{n\neq p} K_{pn} \, . \tag{27}$$

Equations (8), (17), and (24) yield the K_p according to the present set of empirical rate coefficients, Eqs. (7), (20), and (25) yield the MK K_p with the discrete nature of the excited states accounted for, and Eq. (6) directly gives the MK K_p for a continuum of states.

Figures 10 and 11 show a comparison between K_p calculated with these three formulas, using the hydrogenic approximation for the energy levels and for the A_{pn} and B_{pn} values, and the experimental K_p for He (Ref. 7). It is to be noted that the experimental K_p values are, of course, more accurate than the experimental K_{pn}. According to Figs. 10 and 11, the present formula gives the best agreement with experiment. Furthermore, the MK rate coefficients, with the discrete nature of the excited states accounted for, agree better with experiment than the MK K_p for a continuum of states. Figure 11 also shows that the present K_p and the discrete MK K_p are in fair agreement with each other only in a limited range of T_e values.

IV. DISCUSSION

So far we have considered $|p\rangle \rightarrow |n\rangle$ and $|p\rangle \rightarrow |i\rangle$ transitions from states $|p\rangle$, with only one electron in the outer orbital. To get formulas of a quite general validity, transitions involving highly excited states (50-51, 50-60), as well as transitions from ground-state alkali atoms, were included in the analysis. Transitions from other ground-state atoms have not been dealt with. The subdivision of $|p\rangle \rightarrow |n\rangle$ into $|pl\rangle \rightarrow |nl'\rangle$ transitions has not been dealt with in detail. We shall comment briefly on these and other subjects.

The empirical and classical ionization cross-section formulas given, for example, by Drawin,[38] Gryzinski,[26] and Lotz,[39] which are also based on classical scaling, give σ_{pi} which are very similar to each other, and which are in good agreement with experiment for ground-state H, He, Ar, Kr, Xe, and various other atoms and molecules. However, these formulas underestimate the cross sections near threshold for the alkali atoms and for the excited noble-gas atoms by about a factor of 2, and disagree furthermore with the classical three-body theory by this same factor. At higher energies there is better agreement. Our formula just applies to the latter transitions. The present formula is thus complementary to these other formulas and is more suitable for ionization from excited states and ground-state alkali atoms.

For excitation, Drawin and Emard[40] have recently given analytical cross-section and rate formulas. These formulas were obtained by fitting to experimental and theoretical cross sections for excitation from the H $1s$ state, and they utilize classical scaling too. Since classical scaling leads to very poor results for transitions between (highly) excited states, formulas like those of Ref. 40 should not be used for these transitions.

This paper deals with $|p\rangle \rightarrow |n\rangle$ transitions and not with the subdivision into $|pl\rangle \rightarrow |nl'\rangle$ transitions (see, e.g., Ref. 41). This need not be a serious restriction for many applications in plasma modeling, because l mixing proceeds rapidly for the higher levels, resulting in a distribution among the various angular-momentum quantum states according to the statistical weights. If one wishes to make such a subdivision, the formulas given here can still be used, in which case the A_{pn} and B_{pn} are the individual $A_{plnl'}$ $(f_{plnl'})$ and $B_{plnl'}$. Individual A and B values have been utilized here for the first resonance transitions in the alkali atoms.

The present σ_{pn} are finite at threshold and the σ_p are finite at the first inelastic threshold. The fact that the MK rate coefficients are larger than the present K_{pn} and K_p at small T_e (Figs. 6, 7, and 11) thus implies that the corresponding σ_{pn} and σ_p would be finite too and would be even larger than our cross sections. In the sudden regime, the MK K_{pn} and K_p decrease more slowly with increasing T_e than the present rate coefficients. This corresponds to cross sections which decrease too slowly with increasing E_e.

For electron-impact ionization, classical scaling (BEA) has been found to work extremely well in a wide range including highly excited Rydberg atoms and ground-state alkali atoms. Similarly, Olson[42] has recently found classical scaling to apply very well too for heavy-particle–heavy-particle ionization in a range including $p = 10$ and 20 Rydberg-atom–Rydberg-atom collisions as well as He $2S$-He $2S$ and H $1S$-H $1S$ collisions.

V. SUMMARY AND RECOMMENDATIONS

For ionization by electron impact, analytical formulas are given for the cross sections and rate coefficients. These formulas have the correct BEA limit at large E_e and kT_e (the small logarithmic term is neglected), they represent the classical three-body theory within statistical accuracy ($\approx 10\%$) at all E_e and kT_e, and they agree within 10 to 30% with the experimental σ_{pi} for excited H, He, Ne, and Ar and ground-state alkali atoms.

An analytical formula is given for the three-body recombination rate coefficients. This formula agrees with the one for the inverse process: electron-impact ionization, and thus has the same accuracy.

For $|p\rangle \rightarrow |n\rangle$ excitation and deexcitation by electron impact, analytical cross-section and rate coefficient formulas are given. The σ_{pn} have the correct large E_e limit (Born, Bethe and also BEA for large $|n-p|$) for all transitions. At all energies, except very close to threshold, the σ_{pn} agree within 5 to 20% with experiment for the first resonance transitions in the alkali atoms and for the H $1 \rightarrow 2$ transition. For the excited states with $p,n \geq 5$, the σ_{pn} agree typically within 5 to 20% with the theoretical σ_{pn} from Gee and Percival *et al.*,[14,22] which are thought to be the most accurate σ_{pn} available. The present K_{pn} and K_p agree rather well with those obtained in Refs. 6 and 7 from laser-induced fluorescence-spectroscopy experiments. The structure of the present formulas is such that cross sections and rate coefficients can also be calculated for the individual $|pl\rangle \rightarrow |nl'\rangle$ transitions, once the individual A and B coefficients are known.

The Mansbach and Keck rate coefficients are shown to be accurate only in a limited (adiabatic) range of T_e values. In the sudden regime the MK K_{pn} leads to very poor results (Figs. 6 through 8), and the applicability for $kT_e < E_{pn}$ is doubtful too.

TABLE II. List of recommended cross-section and rate coefficient formulas for electron-impact ionization (σ_{pi}, K_{pi}), three-body recombination (α_{ip}), excitation and de-excitation (σ_{pn}, K_{pn}), and total depopulation (σ_p, K_p) of excited atoms.

	Equation		
Process	σ	K	α_{ip}
ionization	2 ($\alpha = 3.25$)	8	
recombination			9
excitation	14	17	
deexcitation	21	24	
total depopulation	26	27	

The formulas for σ_{pn} and K_{pn} given by Gee *et al.*,[14,22] which are claimed to be accurate for $p,n \geq 5$, $E_e > 4E_{pi}$, and $T_e > 10^6/p^2$ for excitation, and $T_e > 10^6/n^2$ for deexcitation, are found to work well even outside this regime. However, when two of these restrictions are violated, e.g., for small n and small T_e (Fig. 9), unrealistic results are obtained.

The present cross-section and rate coefficient formulas, which are recommended for future applications, are listed in Table II.

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