

Mutual neutralization in collisions between negative hydrogen ions and singly-charged positive ions: II. He^+ and Li^+ projectiles at low-keV energies

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Abstract. Coupled-channel impact parameter cross sections are reported for mutual neutralization in $\text{He}^+ + \text{H}^-$ and $\text{Li}^+ + \text{H}^-$ collisions for the impact energy range between 0.07 and 25 keV amu⁻¹. The results are obtained using a one-active-electron model and a 22-state two-centre atomic orbital basis. They are compared with available molecular orbital calculations as well as with experimental data. In a wide range of energies considered, good agreement is obtained with recently measured cross sections. Comparison of present results with neutralization cross sections for $\text{H}^+ + \text{H}^-$ collisions reveals some differences in the neutralization curves regarding impact velocity, for different projectiles.

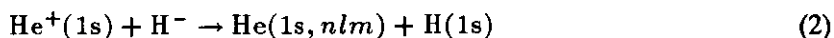
1. Introduction

This work is an extension of the previous calculations (Ermolaev 1988, hereafter referred to as I) of mutual neutralization in collisions between protons and negative hydrogen ions,

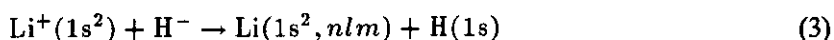


for $0.62 \leq E \leq 80$ keV amu⁻¹ where E is the impact energy of the projectile H^+ in a laboratory frame where the target ion, H^- , is assumed to be at rest.

In the present work, computed cross sections are reported for mutual neutralization in the following two reactions:



for $0.25 \leq E \leq 25$ keV amu⁻¹, and



for $0.07 \leq E \leq 25$ keV amu⁻¹.

Theoretical and experimental studies of reaction (1) have already been reviewed and discussed in I. It may be pointed out here that the theory of mutual neutralization (1) was dominated for a long time by a Landau–Zener picture of transitions (Bates and Lewis 1955). According to this theory, transitions occur in the vicinity of the

pseudocrossing of potential energy curves: neutralization into $H(n=2) + H$ prevails at energies above around 30 eV whereas for lower energies the main contribution comes from neutralization into $H(n=3) + H$. Subsequent close-coupling impact parameter calculations of Sidis *et al* (1983) and Borondo *et al* (1983) using molecular orbital (MO) bases and those in I where an atomic orbital (AO) basis was employed, showed the significance of the $n=3$ transition at E well above 30 eV. Paper I also demonstrated that the $n=4$ transition also contributed within the close-coupling model, despite a large distance to the corresponding pseudocrossing at around 270 au. Comparison of the MO and AO calculations showed good agreement between the two models for impact energies $E \leq 10$ keV. For higher energies, however, the MO data tended to overestimate the experimental cross sections. Recently Shingal *et al* (1985) and Shingal and Bransden (1990) successfully applied a two-active-electron AO theory to reaction (1). Generally, as far as reaction (1) is concerned, a good overall agreement between the theoretical treatments and recent experiments of Szucs *et al* (1984), Peart *et al* (1985) and Schon *et al* (1987) is recorded in a wide energy range.

Reaction (2) was studied experimentally by Gaily and Harrison (1970) and by Peart *et al* (1985, 1987). Reaction (3) was studied by Peart and Foster (1987). On the other hand, the theoretical effort, unlike the case of the H^+ projectile, was rather limited. For reaction (2) there are no theoretical cross sections. The first theoretical treatment of reaction (3) by Bates and Boyd (1956) who obtained the coefficient of ionic recombination between H^- and positive ions of alkali atoms Li^+ , Na^+ and K^+ was based on the Landau-Zener model as well as the earlier work by Bates and Lewis (1955). Recently, a close-coupling treatment of reaction (3) was reported by Mendez *et al* (1990) who developed a molecular theory of neutralization at low impact energies.

In view of the success of the one-active-electron model as applied to reaction (1) in I, the author has decided to carry out a similar study of neutralization reactions (2) and (3).

The plan of the paper is as follows. A brief summary of the theory is given in section 2 which is followed by results in section 3 and concluded by a discussion in section 4.

2. Theory

Calculations were carried out within the impact parameter formalism using a one-active-electron approximation to interactions in the $X^+ + H^-$ system (where X is either He or Li). The corresponding time-dependent Schrödinger equation has the form

$$\left[-\frac{1}{2}\nabla_r^2 + U_A(r_A) + U_B(r_B) - i\frac{\partial}{\partial t} \right] \Psi = 0 \quad (4)$$

the distances r , r_A and r_B being defined in figure 1 where relative motion of A and B is assumed to proceed along a rectilinear trajectory at a constant velocity v . The potential U_A at the H-centre is taken to be a short-range Yukawa-type central potential,

$$U_A(r_A) = -\frac{e^{-\beta' r_A}}{r_A} \quad (5)$$

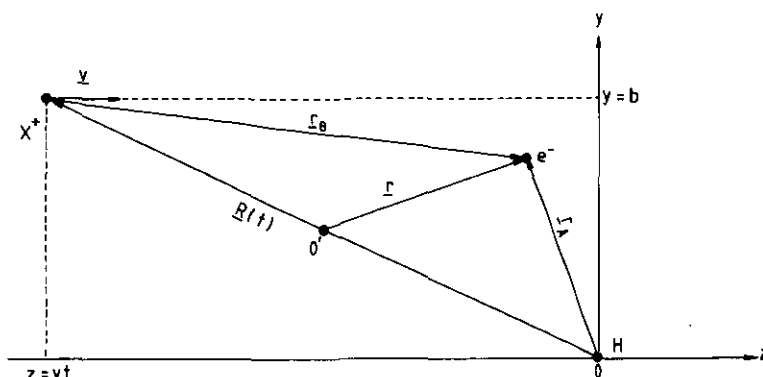


Figure 1. A coordinate system for $X^+ + (H + e^-)$. $O'X^+ = O'O = R(t)/2$.

Table 1. The low-lying eigenvalues $\epsilon(n)$, $l = 0$, of one-electron Hamiltonian H_B with the effective potential U_B (equation (6)) for $Z = 1, 2$ and 3 .

Core ion n	$H^+ (Z = 1)$ $-\epsilon(n)$	$\text{He}^+ (Z = 2)$ $-\epsilon(n)$		$\text{Li}^+ (Z = 3)$ $-\epsilon(n)$	
	Theory	Theory	Experiment ^a	Theory	Experiment ^a
1	0.500 00	0.903 73 ^b	0.903 72		
2	0.125 00	0.157 41	0.145 97	0.198 16 ^b	0.198 16
3	0.055 56	0.064 42	0.061 27	0.074 31	0.074 18
4	0.031 25	0.034 86	0.033 59	0.038 68	0.038 62
5	0.020 00	0.021 81	0.021 17	0.023 67	0.023 64
6	0.013 89	0.014 90	0.014 56	0.015 97	0.015 95

^a Moore (1958).

^b The energy level used to optimize U_B with respect to β .

with $\beta' = 0.8815$, already discussed in I for the $H^+ + H^-$ collisions.

The potential $U_B(r_B)$ at the X-centre is taken to be a sum of a Coulomb potential with the correct asymptotic value of the ionic charge, and a short-range Yukawa-type central potential taking into account the distortion of the Coulomb centre by passive K-shell electron(s):

$$U_B(r_B) = -\frac{1}{r_B} - \frac{(Z_B - 1)e^{-\beta r_B}}{r_B}. \quad (6)$$

If potential U_B is optimized by choosing β to fit experimental values of the ionization potentials for $\text{He}(1s^2)$ and $\text{Li}(1s^2 2s)$ then one obtains

$$\beta = 2.125 \quad \text{for } \text{He}^+ \quad Z = 2 \quad (7a)$$

and

$$\beta = 2.046 \quad \text{for } \text{Li}^+ \quad Z = 3. \quad (7b)$$

Table 1 presents computed eigenenergies of the one-particle Hamiltonian H_A with potential (6), $Z = 1, 2$, and 3 , using the bases of 12 Slater-type orbitals (STO) and

compares them with the corresponding experimental terms. The adopted form of the effective potentials is particularly good for the alkali centre where agreement with the experimental levels is within 0.1%. For the He-centre, only the computed 2s level shows a relatively large deviation of 5% whereas other levels agree with experiment within 0.5%.

The solution Ψ of the time-dependent Schrödinger equation for the active electron is sought as an expansion in terms of a two-centre atomic orbital (TCAO) basis $[u_j^A, u_{j'}^B]$, $j = 1, \dots, N_A$; $j' = 1, \dots, N_B$.

In the usual notations, the expansion takes the form

$$\Psi(b, r, t) = \sum_{j=1}^{N_A} a_j^A(b, t) u_j^A \eta_j^A + \sum_{j'=1}^{N_B} a_{j'}^B(b, t) u_{j'}^B \eta_{j'}^B \quad (8)$$

where b is the impact parameter, $u_j^{A,B}$ and corresponding $\epsilon_j^{A,B}$ are eigenfunctions and eigenenergies of an electron moving in the central field potentials V_A and V_B , respectively, and $\eta_j^{A,B}$ are plane-wave electronic translational factors (PWETF).

Calculations were carried out using a TCAO basis with a single (ground) state $u_1^A(r_A)$ on centre A and a set of 21 orthogonal atomic states $u_j^B(r_B)$ with $l = 0, 1, 2$, centred on the projectile. The atomic states were constructed using sets of STOs specified in table 2 where adjusted values of β in (6) were used to account for a relatively small STO sets employed in the actual calculations of cross sections.

Table 2. The 22-state TCAO bases and STOs used in calculations of neutralization reactions (2) and (3).

nl	ϵ_j	n_j	α_j	nl	ϵ_j	n_j	α_j
A-centre (H) $\beta = 0.8815$				A-centre (H) $\beta = 0.8815$			
1s	-0.027 575	0	0.236	1s	-0.027 575	0	0.236
		1	0.236			1	0.236
		0	0.791			0	3.791
		0	1.146			0	1.146
		0	1.601			0	1.601
B-centre (He ⁺) $\beta = 2.0250$				B-centre (Li ⁺) $\beta = 1.9283$			
2s	-0.158 212	0	1.344	2s	-0.198 175	0	1.259
3s	-0.064 616	1	0.672	3s	-0.074 123	0	0.630
4s	-0.034 947	1	0.500	4s	-0.038 418	1	0.630
<u>5s</u>	0.001 366	2	0.500	<u>5s</u>	0.284 826	2	0.500
		2	0.333			2	0.333
		3	0.250			3	0.250
2p	-0.128 414	1	0.672	2p	-0.133 885	1	0.630
3p	-0.056 661	2	0.500	3p	-0.058 365	2	0.500
4p	-0.031 469	1	0.333	4p	-0.032 313	1	0.333
<u>5p</u>	0.060 117	2	0.333	<u>5p</u>	0.079 433	2	0.333
		3	0.250			3	0.250
3d	-0.055 581	2	0.500	3d	-0.055 621	2	0.500
4d	-0.031 232	2	0.333	4d	-0.031 257	2	0.333
<u>5d</u>	0.088 287	3	0.333	<u>5d</u>	0.086 398	3	0.333
		2	0.250			2	0.250

Numerical procedures for determining the asymptotic values of the expansion coefficients in (8) for $t \rightarrow \infty$ have been described in I. Partial cross sections for mutual neutralization are obtained by integrating the corresponding probabilities $p_j(b) = |a_j(b, -\infty)|^2$ over impact parameter b , and the total cross sections are obtained by summing over all partial cross sections.

3. Results

3.1. Neutralization cross sections

The computed partial $\sigma_m(n)$, $n = 2, 3, 4$, and total σ_m cross sections for mutual neutralization in collisions between H^- ions and singly-ionized positive ions He^+ and Li^+ are presented in tables 3 and 4, respectively. The energy range considered was $0.25 \leq E \leq 25 \text{ keV amu}^{-1}$ for He^+ , and $0.07 \leq E \leq 25 \text{ keV amu}^{-1}$ for Li^+ . The present calculations show that neutralization into $\text{X}(n=2) + \text{H}$ proceeds as a dominating transition at low E . At the same time, cross sections for neutralization into $\text{X}(n=3) + \text{H}$ are also significant, well beyond the low- E end of the energy region presently considered. The situation is qualitatively similar to that found in I for $\text{H}^+ + \text{H}^-$ collisions. It can also be seen that for all projectiles in question, neutralization into $\text{X}(n=4) + \text{H}$ contributes as well.

Experimental total cross sections for He^+ projectiles were measured by Gaily and Harrison (1970) in the energy range between $0.125 \text{ eV amu}^{-1}$ and 5 keV amu^{-1} , and by Peart *et al* (1985) in the energy range between 0.125 and $3.75 \text{ keV amu}^{-1}$. Both sets of data agree with each other within 10% and suggest a possible structure in the cross sections (see also Massey (1976), particularly p 635ff). We note that an earlier observation (Gaily and Harrison 1970) of a structure in neutralization cross sections for reaction (1) has been disproved in the same experiment (Peart *et al* 1985). For Li^+ , the experimental cross sections (Peart and Foster 1987) which exhibit a rather smooth curve, are available in the energy range between 0.038 and $2.74 \text{ keV amu}^{-1}$. Comparison between the reported theoretical results and experiment is presented below in figures 2 and 3.

3.2. Capture into projectile continuum

The STO basis used in present calculations contains six positive energy states $\overline{5s}$, $\overline{5p}$ and $\overline{5d}$ centred on the projectile (see table 2). This allows one to obtain an estimate of capture from H^- into the projectile continuum. The corresponding cross sections σ_m^{CPC} are presented in figure 4. At low energies, these may be related to ionization of the target but as impact energy increases, the main part of the ionization flux gradually shifts to direct ionization. The latter cannot be estimated unless the expansion basis in (4) contains positive-energy states u_j^A centred on target A.

4. Discussion

Generally, theoretical and experimental cross sections for mutual neutralization are in good agreement with each other within the whole energy range considered. The computed neutralization cross sections for He^+ shown in figure 2 display the same

Table 3. Theoretical cross sections for the neutralization reaction $\text{He}^+(1s) + \text{H}^- \rightarrow \text{He}(1s\ n\ell) + \text{H}(1s)$, $n = 2, 3, 4$; $l = 0, 1, 2$ (10^{-16} cm^2).

E (keV amu ⁻¹)	V (10 ⁵ m s ⁻¹)	Total $n = 2$			Total $n = 3$			Total $n = 4$					
		2s	2p		3s	3p	3d	4s	4p	4d	σ_m		
0.250	2.18	6.3	48.0	54.3	9.2	13.7	17.1	40.0	7.6	24.3	7.6	39.5	133.8
0.375	2.67	11.9	55.5	67.4	5.0	7.3	11.4	23.7	4.4	21.1	3.4	28.9	120.0
0.500	3.0	15.6	60.8	76.4	5.9	8.6	14.3	28.8	2.4	11.2	4.0	17.0	122.2
0.750	3.77	14.3	48.7	63.0	6.5	11.0	16.0	33.5	1.1	4.5	2.3	7.9	104.4
1.000	4.36	14.4	46.9	61.3	6.9	12.5	16.6	36.0	0.7	2.8	3.3	6.8	104.1
1.250	4.87	9.7	42.6	52.3	7.6	14.0	17.1	38.7	0.6	2.1	4.6	7.3	98.3
1.500	5.34	7.3	38.3	45.6	8.4	15.2	17.7	41.3	0.5	2.0	5.1	7.6	94.5
1.750	5.77	5.2	34.7	39.9	9.2	16.5	17.2	42.9	0.4	1.9	5.7	8.0	90.8
2.000	6.17	4.7	30.9	35.6	9.4	17.1	17.5	44.0	0.3	1.9	5.6	7.8	87.4
3.000	7.55	3.7	20.3	24.0	10.0	19.0	15.0	44.0	0.2	1.8	4.3	6.3	74.3
5.000	9.75	1.7	11.2	12.9	8.2	12.8	8.8	29.8	0.08	1.2	1.1	2.4	45.1
7.500	11.9	0.9	8.0	8.9	5.5	8.2	4.8	18.5	0.07	0.6	0.2	0.9	28.3
10.00	13.8	1.4	5.7	7.1	3.7	5.0	2.6	11.3	0.07	0.3	0.1	0.5	18.9
12.50	15.4	1.6	3.8	5.4	2.5	3.0	1.4	6.9	0.06	0.1	0.08	0.3	12.5
25.00	21.8	0.6	0.6	1.2	0.4	0.3	0.1	0.8	0.02	0.01	0.02	0.05	2.05

Table 4. Theoretical cross sections for the neutralization reaction $\text{Li}^+(1s^2) + \text{H}^- \rightarrow \text{Li}(1s^2 n\ell) + \text{H}(1s)$, $n = 2, 3, 4$; $l = 0, 1, 2$ (10^{-16} cm^2).

E (keV amu ⁻¹)	V (10 ⁵ m s ⁻¹)	Total $n = 2$				Total $n = 3$				Total $n = 4$	σ_m		
		2s	2p	3s	3p	3d	4s	4p	4d				
0.071	1.16	1.2	52.0	53.2	46.8	10.1	7.8	64.7	3.5	38.0	9.5	51.0	168.9
0.100	1.38	1.5	57.3	58.8	45.4	10.5	8.4	64.3	4.3	33.8	10.2	48.3	171.4
0.143	1.65	0.3	62.0	62.3	39.3	10.0	8.9	58.2	2.2	22.5	11.8	36.5	157.3
0.214	2.02	1.1	63.0	64.1	32.5	10.8	9.8	53.1	2.5	20.3	8.8	31.6	148.8
0.286	2.33	1.3	64.3	65.6	26.6	11.1	10.9	48.6	2.8	17.6	5.9	26.3	140.5
0.429	2.85	2.6	64.4	67.0	16.7	12.9	13.9	43.5	1.5	14.5	2.9	18.9	129.4
0.714	3.68	3.4	62.4	65.8	14.4	14.9	17.0	46.3	0.6	4.9	2.2	7.7	119.8
1.000	4.36	7.2	53.3	60.5	15.0	15.2	17.0	47.2	0.4	2.4	3.2	6.0	113.7
1.428	5.22	10.6	41.0	51.6	16.0	14.9	16.9	47.8	0.6	2.6	4.8	8.0	108.3
2.857	7.37	8.8	23.7	32.5	12.4	14.6	14.5	41.5	0.2	2.3	5.3	7.8	81.8
4.286	9.03	4.6	13.6	18.2	8.4	8.9	9.5	26.8	0.4	1.8	2.4	4.6	49.6
5.714	10.4	4.4	9.7	14.1	6.6	6.5	6.8	19.9	0.5	1.5	1.1	3.1	37.1
7.143	11.6	4.2	8.3	12.5	5.7	5.1	5.2	16.0	0.4	1.3	0.5	2.2	30.7
12.50	15.5	3.9	4.2	8.1	3.3	1.6	1.7	6.6	0.5	0.5	0.1	1.1	15.8
25.00	21.8	2.9	0.7	3.6	0.8	0.12	0.14	1.06	0.2	0.04	0.02	0.26	4.9

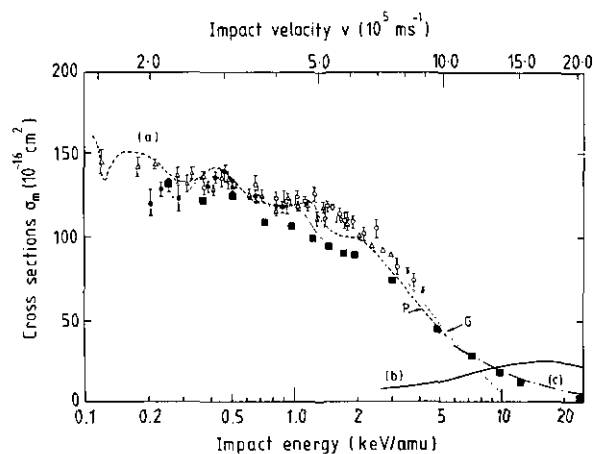


Figure 2. (a) Mutual neutralization $\text{He}^+ + \text{H}^- \rightarrow \text{He} + \text{H}$. Total cross sections σ_m . (i) Theory: ■ present work, 22-state TCAO. (ii) Experiment: Δ , \circ , and \bullet , Peart *et al* (1985); dotted curve G: Gaily and Harrison (1970); broken curve P: Peart *et al* (1976). (b) $\text{He}^+ + \text{H} \rightarrow \text{He} + \text{H}^+$. Total cross sections $\sigma'_c \times 10$: full curve, Olson *et al* (1977). (c) Chain curve: neutralization curve with MTPC, present work.

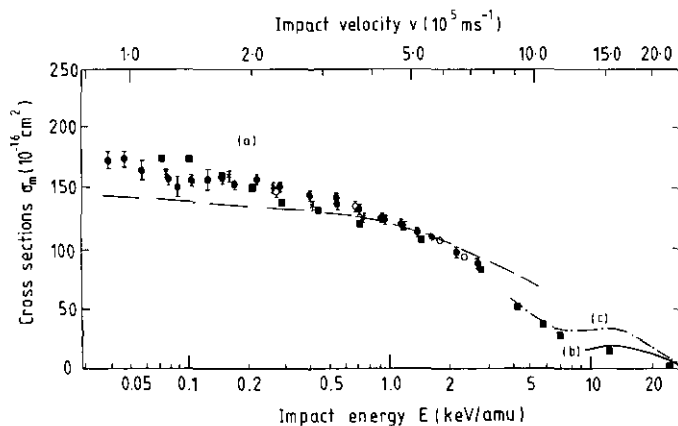


Figure 3. (a) Mutual neutralization $\text{Li}^+ + \text{H}^- \rightarrow \text{Li} + \text{H}$. Total cross sections σ_m . (i) Theory: ■, present work, 22-state TCAO; broken curve, 5-state MO, Mendez *et al* (1990). (ii) Experiment: \times , \circ , and \bullet , Peart and Foster (1987). (b) $\text{Li}^+ + \text{H} \rightarrow \text{Li} + \text{H}^+$. Total cross sections σ'_c : full curve, Shah *et al* (1978). (c) Chain curve: neutralization curve with MPC, present work.

shape including a flat structure between 0.5 and 2 keV amu^{-1} , but they are systematically lower by 10–15% than the experimental data. For the Li^+ projectiles, the corresponding neutralization curve shown in figure 3 is in excellent agreement with the experimental data of Peart and Foster (1987). It is clear that the effective potential (5) is particularly good for alkali atoms. The MO calculation of (3) by Mendez *et al* (1990) is also shown in figure 3. Their curve generally displays good agreement with experiment but tends to underestimate the measured cross sections at lower E

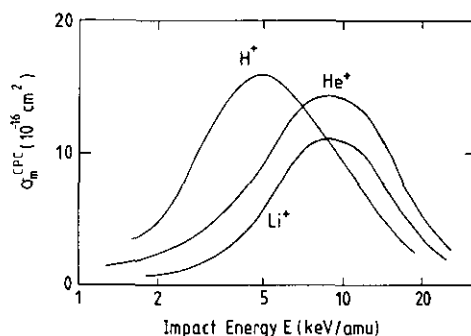


Figure 4. Capture into projectile continuum (CPC). Total cross sections σ_m^{CPC} for H^+ , He^+ and Li^+ projectiles—present work, 22-state TCAO.

and overestimates them at higher E within the interval treated. For higher E , the main deficiency of the MO approach to rearrangement collisions is in the lack of the proper account for momentum transfer as had been pointed out by many authors in the past including Ermolaev (1984) and Newby (1985).

Curves (b) in figures 2 and 3 display total capture cross sections for reactions



and



as reported by Olson *et al* (1977) and Shah *et al* (1978). These can be used to obtain a modified independent-particle correction (MIPC) to the mutual neutralization cross sections at high E in a way similar to that suggested in I for reaction (1). The corrected curves are labelled (c) in figures 2 and 3. The present theory predicts a shoulder at $E \approx 10$ keV amu^{-1} in the Li^+ curve but no such effect is expected for the He^+ projectile in the energy range considered.

Experimental values of partial cross sections $\sigma_m(n)$ are not yet available for either projectile in (1)–(3). However, comparison can be carried out with the existing theoretical data for the Li^+ and H^+ projectiles. The MO calculations of Mendez *et al* (1990) for the $\text{Li}^+ + \text{H}^-$ system used a five-state (four Σ and one Π) molecular basis. This allowed them to obtain partial cross sections for capture into the final $(1s^2)$ 2s, 2p, and 3s states of the lithium atom. These cross sections are compared with the present AO calculations in figure 5.

It can be seen that the partial MO and AO cross sections in figure 5 differ considerably in the whole energy range considered though the *total* MO and AO cross sections are close to each other. A discussion in I suggests that the Yukawa-type potential (5) and the five-STO representation of the ground state H^- function used in the present work, are sufficiently accurate, at least in the intermediate part of the E range. Further calculations with an enlarged TCAO basis and an improved model for the H^- centre may be of particular interest to assess the accuracy of the present computations at low E .

As E increases the MO and AO curves show a tendency to converge but still remain quite different at the highest E where the present AO data are expected to be

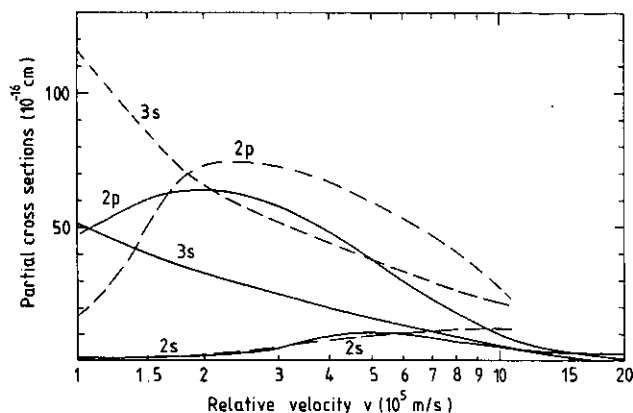


Figure 5. Mutual neutralization in $\text{Li}^+ + \text{H}^-$ collisions. Partial cross sections $\sigma_m(2s)$, $\sigma_m(2p)$, and $\sigma_m(3s)$ for formation of $\text{Li}(nl)$: full curve, present work, 22-state TCAO; broken curve, five-state MO, Mendez *et al* (1990).

more accurate than the five-state MO calculation. Probably, a larger molecular basis ought to be employed to improve on the MO result at high E .

Using the data of tables 3 and 4 the relative distributions of partial cross sections in the final states of X have been obtained as a function of v and displayed in figure 6. For the H^+ projectiles, similar curves were obtained using the two-active-electron cross sections of Shingal and Bransden (1990). Both types of curves show a remarkable consistency with each other, particularly for $n = 2$, thus giving an additional support to the model and calculations reported in the present work. It would be interesting to carry out two-active-electron calculations of reactions (2) and (3) using effective potentials (6) for the He^+ and Li^+ projectiles. This could be an important general test of the one-active-electron model in a wide range of E .

Although all central potentials of form (6) approach the same pure Coulombic potential with $Z = 1$, as $R \rightarrow \infty$, the distribution of *low-lying* excited levels is noticeably different for H, He, and Li as table 1 shows. This has an important physical consequence for collisions of negative hydrogen ions with the singly-charged positive ions H^+ , He^+ , and Li^+ . For low collisional velocities v , where the molecular picture of the collision provides a good guide, transitions are assumed to occur in the vicinity of crossing points, R_{ic} , between the ionic (Coulombic) curve correlated with $X^+ + \text{H}^-$ and the horizontal covalent potential curves associated with $X^*(nl) + \text{H}(1s)$. The values of $R_{ic}(n) = a_0 / [\epsilon_A(1) - \epsilon_B(n)]$ where a_0 is the Bohr radius and ϵ_A and ϵ_B are eigenenergies of the $\text{H} + e^-$ and $X^+ + e^-$ systems, are given in table 5. It shows a considerable spread of $R_{ic}(n)$ for given n , for different projectiles. The table may suggest that in the range of E where this picture is valid, the $n = 3$ and $n = 4$ contributions are particularly significant in the case of the Li^+ projectiles. In the opposite case of higher relative velocities v , the neutralization curves for different projectiles X^+ apparently converge to a common curve, at a surprisingly low $v \approx 0.15\text{--}0.20$ au, in agreement with experiment.

The general conclusion is that the one-active-electron model gives, within a close-coupling AO expansion, a good quantitative description of recombination (1)–(3) in a wide range of impact energies E .

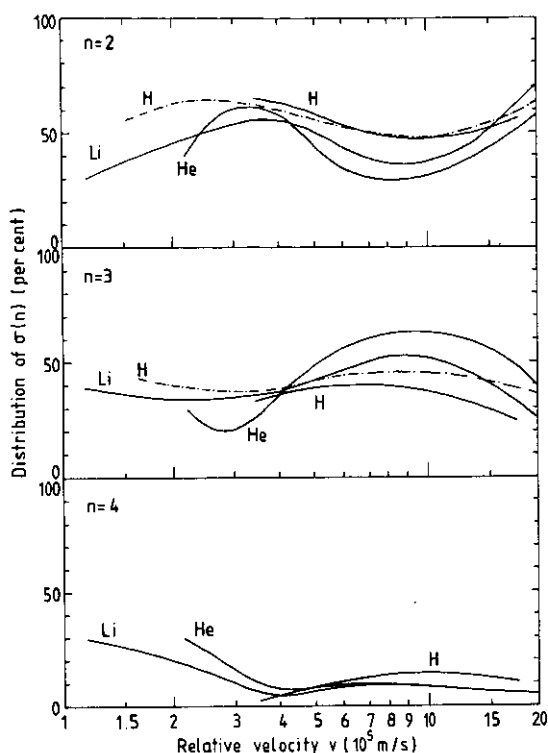


Figure 6. Distribution of $\sigma_m(n)$ for $n = 2, 3$, and 4 in the final $X(n)$ states. Total is taken to be 100. (i) Full curve: Ermolaev (1988), 29-state TCAO, for H; present work, 22-state TCAO for He and Li; (ii) chain curve: 46-state, two-active-electron AO model, Shingal and Bransden (1990), for H.

Table 5. $X^+ \text{H}(1s) - X^+ \text{H}^-$ interactions. The list of $R_{ic}(n)$, with $\varepsilon(n)$ from table 1.

n	H^+	He^+	Li^+
2	10.3	7.7	5.9
3	35.7	27.1	21.4
4	272.0	137.0	90.0

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