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An improved calculation for the ionization–excitation of helium

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Abstract

Cross sections for the ionization–excitation of helium by fast charged-particle impact are calculated using the impact-parameter method and second-order perturbation theory. The inclusion of the two-step 1 mechanism in the calculations considerably improves our previous results. The dependence of the cross sections on the sign of the projectile charge is also investigated.

Experimental data for the ionization–excitation of helium by fast charged-particle impact [1] show that, similarly to double ionization, cross sections for electrons are almost a factor-of-two larger than for the same velocity protons over a wide velocity range (above 5 au). This interesting behaviour has not yet been reproduced theoretically. Most of the theoretical investigations of this transition are only performed for electron projectiles [2–8] and our previous calculations [9,10] lead to a much smaller difference in the cross sections for positively and negatively charged projectiles.

In our previous work [10] we performed a second-order calculation in terms of the projectile–electron interaction. Electron correlation has been taken into account in the initial state by the use of CI wavefunctions, but has been neglected in the final state. In this approximation we have obtained, for the ratio between cross sections for electrons and protons, the value of 1.1 at 5 au velocity, instead of the experimental ratio of 1.6.

Now we include in our calculations the two-step 1 (TS1) mechanism. In this process the electron ejected directly by the projectile interacts with the other electron, causing its transition. In this way electron correlation in the final state is taken into account perturbationally. This mechanism is necessary for the correct description of the two-electron transition, because electron correlation is not included in the wavefunction for the final state. We study the importance of the TS1 mechanism in the calculation of the ionization–excitation cross sections, and compare the obtained cross sections for positive and negative projectiles (protons and antiprotons) at different impact energies.

Our theoretical model has been described in detail previously [11, 12]. We apply in the calculation the impact parameter method, the projectile moving on a classical straight-line

trajectory. The differential cross section for a given energy ϵ_c and angle Ω of the ejected electron can be obtained by an integral of the transition probability over the impact parameter

$$\sigma(\epsilon_c, \Omega) = \int \mathrm{d}^2\boldsymbol{B}\, |a(B)|^2, \tag{1}$$

while the total cross section is obtained by integration over all energies and angles:

$$\sigma = \int \mathrm{d}\Omega \int \mathrm{d}\epsilon_c\, \sigma(\epsilon_c, \Omega). \tag{2}$$

The transition amplitude $a(B)$ can be written as a sum of the amplitudes characteristic for each mechanism. In terms of the many-body perturbation theory through second order these mechanisms are the two-step 2 (TS2—second-order in projectile–electron interaction), shake, ground-state correlation and TS1 (final-state correlation) [11, 13]. As shown below, we take into account ground-state correlation by the use of the correlated wavefunction in the initial state, and in this way it is included to all orders in the generalized shake amplitude $a^{(1)}_{\mathrm{shake}}$. In this model the amplitude reduces to three terms:

$$a = a^{(1)}_{\mathrm{shake}} + a^{(1)}_{\mathrm{TS1}} + a^{(2)}. \tag{3}$$

The shake amplitude expresses the first Born approximation. However, this is not the classical one based on single-configuration wavefunctions and change in the screening after the removal of one electron. $a^{(1)}_{\mathrm{shake}}$ is a generalized shake amplitude, which may also account for electron correlation in the initial and final states, if these are described by correlated wavefunctions

$$a^{(1)}_{\mathrm{shake}} = -\mathrm{i} \int_{-\infty}^{+\infty} \mathrm{d}t\, \exp(\mathrm{i}(E_f - E_i)t) \langle f | [V_1(t) + V_2(t)] | i \rangle. \tag{4}$$

Here E_i and E_f stand for the energies of the initial and final states i and f respectively, while $V_1(t)$ and $V_2(t)$ are the two projectile–electron interaction potentials.

In the present calculations we use correlated CI wavefunctions for the ground state of helium taken from the literature [14]:

$$|i\rangle = \sum_l c_l |i_1^l\rangle |i_2^l\rangle, \tag{5}$$

but the final state is described by uncorrelated, properly symmetrized wavefunctions:

$$|f\rangle = \frac{1}{\sqrt{2}} (|f_c'(1)\rangle |f_e'(2)\rangle + |f_e'(1)\rangle |f_c'(2)\rangle). \tag{6}$$

f_e' stands for the excited state of the He^+ ion, while f_c' is the wavefunction of the ejected electron, calculated numerically in the screened potential of the residual ion. Because f_e' is a 2p orbital, the final state is practically orthogonal to the initial state. To be rigorous, we should mention that the initial state also contains $np n'p$ configurations, and this orthogonality is not exact. However, the $\langle i | f \rangle$ overlap integral for the mean value of the energy of the ejected electron is less than 10^{-2}, and no further orthogonalization is necessary. Through this choice of the wavefunctions $a^{(1)}_{\mathrm{shake}}$ includes the classical shake mechanism and ground-state correlation, but does not include final-state correlation.

Introducing the expressions above for the initial and final states into the formula (4) of the amplitude we get

$$a^{(1)}_{\rm shake} = -{\rm i}\frac{1}{\sqrt{2}}\sum_l c_l\Bigg[\langle f'_e(2)|i^l_2\rangle\int_{-\infty}^{+\infty}{\rm d}t\ \exp({\rm i}(E_f-E_i)t)\langle f'_c(1)|V_1(t)|i^l_1\rangle$$
$$+\langle f'_e(1)|i^l_1\rangle\int_{-\infty}^{+\infty}{\rm d}t\ \exp({\rm i}(E_f-E_i)t)\langle f'_c(2)|V_2(t)|i^l_2\rangle$$
$$+\langle f'_c(2)|i^l_2\rangle\int_{-\infty}^{+\infty}{\rm d}t\ \exp({\rm i}(E_f-E_i)t)\langle f'_e(1)|V_1(t)|i^l_1\rangle$$
$$+\langle f'_c(1)|i^l_1\rangle\int_{-\infty}^{+\infty}{\rm d}t\ \exp({\rm i}(E_f-E_i)t)\langle f'_e(2)|V_2(t)|i^l_2\rangle\Bigg]. \tag{7}$$

Thus, ground-state correlation and classical shake mechanisms are included in the amplitude above to all orders.

The final-state correlation of the two electrons is included in the $a^{(1)}_{\rm TS1}$ amplitude perturbationally. Within second-order many-body perturbation theory the TS1 mechanism means that the $V_1(t') + V_2(t')$ projectile–electron interaction takes the two-electron system to an intermediate state k (with energy E_k), then the v electron–electron interaction leads to the final state. Thus, the TS1 amplitude will read [11]

$$a_{\rm TS1} = -\sum_k\int_{-\infty}^{+\infty}{\rm d}t'\int_{t'}^{+\infty}{\rm d}t\ \exp({\rm i}(E_f-E_k)t)\langle f|v|k\rangle\langle k|V_1(t')$$
$$+V_2(t')|i\rangle\exp({\rm i}(E_k-E_i)t'). \tag{8}$$

In order to obtain a convergent integral over t we introduce in the exponent a dumping factor $+{\rm i}\eta$, and after performing the integration we make $\eta\to 0$

$$a_{\rm TS1} = -{\rm i}\sum_k\int_{-\infty}^{+\infty}{\rm d}t'\,\langle f|v|k\rangle\frac{1}{E_f-E_k+{\rm i}0}\exp({\rm i}(E_f-E_i)t')\langle k|V_1(t')+V_2(t')|i\rangle. \tag{9}$$

Furthermore, we perform the following approximations. For the calculation of this amplitude we consider only the basic $i^1_1 i^1_2 \equiv 1{\rm s}^2$ configuration for the ground state, and also the intermediate state is written as k_1k_2. In these conditions the matrix elements of the projectile–electron interaction become

$$\langle k_1k_2|V_1(t')|i^1_1i^1_2\rangle = \langle k_2|i^1_2\rangle\langle k_1|V_1(t')|i^1_1\rangle \tag{10}$$
$$\langle k_1k_2|V_2(t')|i^1_1i^1_2\rangle = \langle k_1|i^1_1\rangle\langle k_2|V_2(t')|i^1_2\rangle. \tag{11}$$

i^1_1 and k_1 such as i^1_2 and k_2 are not strictly orthogonal, because they are calculated in different potentials (the screening is modified during the collision). However, with good approximation

$$\langle k_2|i^1_2\rangle \approx \langle k_2|i'_2\rangle = \delta_{k_2i'_2} \tag{12}$$
$$\langle k_1|i^1_1\rangle \approx \langle k_1|i'_1\rangle = \delta_{k_1i'_1}, \tag{13}$$

where the primed wavefunctions are calculated with the modified screening. The $\langle i'_2|i^1_2\rangle$ overlap is 0.98, so the change in the screening does not have too much effect on the ground-state wavefunction. Beside $i'_2 \equiv 1{\rm s}$, the largest overlap with the i^1_2 orbital has $k_2 = 2{\rm s}$, where $\langle 2{\rm s}|i^1_2\rangle = 0.15$, other overlap integrals being less than 0.05. In our calculation we neglect the terms containing these small overlap integrals (which squared are less than 0.03), and approximate the $\langle i'_2|i^1_2\rangle$ integral by 1. In these conditions, from the sum over all possible k_1k_2 intermediate states, that over k_2 in the case of the $V_1(t')$ interaction, and that over k_1 for $V_2(t')$ collapse. The energies of the intermediate states will be $E_k = \epsilon_{k_1} + \epsilon'_{i_2}$ or $E_k = \epsilon_{k_2} + \epsilon'_{i_1}$, respectively, where ϵ'_{i_1} and ϵ'_{i_2} stand for the energy of one electron in the 1s state if the other electron has already suffered a transition.

The TS1 amplitude will read

$$a_{\mathrm{TS1}} = -\mathrm{i}\sum_{k_1}\int_{-\infty}^{+\infty}\mathrm{d}t'\,\frac{1}{E_f-\epsilon_{k_1}-\epsilon'_{i_2}+\mathrm{i}0}\exp(\mathrm{i}(E_f-E_i)t')\langle f|v|k_1 i'_2\rangle\langle k_1|V_1(t')|i^1_1\rangle$$
$$-\mathrm{i}\sum_{k_2}\int_{-\infty}^{+\infty}\mathrm{d}t'\,\frac{1}{E_f-\epsilon_{k_2}-\epsilon'_{i_1}+\mathrm{i}0}\exp(\mathrm{i}(E_f-E_i)t')\langle f|v|k_2 i'_1\rangle\langle k_2|V_2(t')|i^1_2\rangle. \tag{14}$$

f is the same symmetrized wavefunction of the excited and continuum electron as in (6). Because of the complex denominator the a_{TS1} amplitude has real and imaginary parts for each parity of the initial and final states. Taking into account that the 'sum over k_1 or k_2 intermediate states' is a sum over the bound states and an integral over the continuum states, the real and imaginary parts can be separated as follows:

$$a_{\mathrm{TS1}} = -\mathrm{i}P\sum_{k_1}\int \mathrm{d}\boldsymbol{k}_1\,\frac{1}{E_f-\epsilon_{k_1}-\epsilon'_{i_2}}\langle f|v|k_1 i'_2\rangle\int_{-\infty}^{+\infty}\mathrm{d}t'\,\exp(\mathrm{i}(E_f-E_i)t')\langle k_1|V_1(t')|i^1_1\rangle$$
$$-\mathrm{i}P\sum_{k_2}\int \mathrm{d}\boldsymbol{k}_2\,\frac{1}{E_f-\epsilon_{k_2}-\epsilon'_{i_1}}\langle f|v|k_2 i'_1\rangle\int_{-\infty}^{+\infty}\mathrm{d}t'\,\exp(\mathrm{i}(E_f-E_i)t')\langle k_2|V_2(t')|i^1_2\rangle$$
$$+\pi\int k_1^c\,\mathrm{d}\hat{\boldsymbol{k}}_1\,\langle f|v|k_1^c i'_2\rangle\int_{-\infty}^{+\infty}\mathrm{d}t'\,\exp(\mathrm{i}(E_f-E_i)t')\langle k_1^c|V_1(t')|i^1_1\rangle$$
$$+\pi\int k_2^c\,\mathrm{d}\hat{\boldsymbol{k}}_2\,\langle f|v|k_2^c i'_1\rangle\int_{-\infty}^{+\infty}\mathrm{d}t'\,\exp(\mathrm{i}(E_f-E_i)t')\langle k_2^c|V_2(t')|i^1_2\rangle. \tag{15}$$

The first two terms, where we have a principal-value integral over the momenta of the continuum intermediate states and a sum over the discrete intermediate states, are the off-shell part of the amplitude, energy is not conserved for these states. The last two terms are the energy-conserving or on-shell part, where $\epsilon_{k_1^c}=E_f-\epsilon'_{i_2}$ and $\epsilon_{k_2^c}=E_f-\epsilon'_{i_1}$. In the calculations we have neglected the bound intermediate states, and have performed only the integral over the continuum, $\boldsymbol{k}_1$ or $\boldsymbol{k}_2$ states, respectively. This approximation is justified because the $1/(E_f-\epsilon_{k_1}-\epsilon'_{i_2})$ fraction has important values only around $\epsilon_{k_1}\approx E_f-\epsilon'_{i_2}$, and this value lies in the continuum, at least 1.5 Hartrees above threshold. In other words, the $\Delta E=E_f-\epsilon_{k_1}-\epsilon'_{i_2}$ deviation from the conservation of the energy in the intermediate state cannot be too large.

As for the second-order amplitude (describing the TS2 mechanism), it is obtained by a double integral over time and a sum over the intermediate states $|k\rangle$ with energies E_k, the infinite number of eigenstates of the two-electron unperturbed Hamiltonian

$$a^{(2)} = -\sum_k\int_{-\infty}^{+\infty}\mathrm{d}t\,\exp(\mathrm{i}(E_f-E_k)t)\langle f|V_1(t)|k\rangle\int_{-\infty}^{t}\mathrm{d}t'\,\exp(\mathrm{i}(E_k-E_i)t')\langle k|V_2(t')|i\rangle$$
$$-\sum_k\int_{-\infty}^{+\infty}\mathrm{d}t\,\exp(\mathrm{i}(E_f-E_k)t)\langle f|V_2(t)|k\rangle\int_{-\infty}^{t}\mathrm{d}t'\,\exp(\mathrm{i}(E_k-E_i)t')\langle k|V_1(t')|i\rangle. \tag{16}$$

In the present calculations we made the same approximations in the calculation of this amplitude as in our previous paper [10]. Only the intermediate states which are reachable from the initial or the final states by a single-electron transition are kept. In these conditions the second-order amplitude becomes

$$
\begin{aligned}
a^{(2)} = &-\frac{1}{\sqrt{2}}\langle f'_e(2)|f_e(2)\rangle\langle i'_1|i^1_1\rangle \int_{-\infty}^{+\infty} \mathrm{d}t\ \exp(\mathrm{i}(E_f - E_{ie})t)\langle f'_c(1)|V_1(t)|i'_1\rangle \\
&\times \int_{-\infty}^{t} \mathrm{d}t'\ \exp(\mathrm{i}(E_{ie} - E_i)t')\langle f_e(2)|V_2(t')|i^1_2\rangle \\
&-\frac{1}{\sqrt{2}}\langle f'_c(2)|f_c(2)\rangle\langle i'_1|i^1_1\rangle \int_{-\infty}^{+\infty} \mathrm{d}t\ \exp(\mathrm{i}(E_f - E_{ic})t)\langle f'_e(1)|V_1(t)|i'_1\rangle \\
&\times \int_{-\infty}^{t} \mathrm{d}t'\ \exp(\mathrm{i}(E_{ic} - E_i)t')\langle f_c(2)|V_2(t')|i^1_2\rangle \\
&-\frac{1}{\sqrt{2}}\langle f'_e(1)|f_e(1)\rangle\langle i'_2|i^1_2\rangle \int_{-\infty}^{+\infty} \mathrm{d}t\ \exp(\mathrm{i}(E_f - E_{ie})t)\langle f'_c(2)|V_2(t)|i'_2\rangle \\
&\times \int_{-\infty}^{t} \mathrm{d}t'\ \exp(\mathrm{i}(E_{ie} - E_i)t')\langle f_e(1)|V_1(t')|i^1_1\rangle \\
&-\frac{1}{\sqrt{2}}\langle f'_c(1)|f_c(1)\rangle\langle i'_2|i^1_2\rangle \int_{-\infty}^{+\infty} \mathrm{d}t\ \exp(\mathrm{i}(E_f - E_{ic})t)\langle f'_e(2)|V_2(t)|i'_2\rangle \\
&\times \int_{-\infty}^{t} \mathrm{d}t'\ \exp(\mathrm{i}(E_{ic} - E_i)t')\langle f_c(1)|V_1(t')|i^1_1\rangle. \qquad (17)
\end{aligned}
$$

Here E_{ie} stands for the energy of the intermediate state when one electron is in the $|i'_1\rangle$ (unscreened) initial state and the other one in the $|f_e(2)\rangle$ excited state, while E_{ic} represents the energy of the intermediate state described by the $|i'_1\rangle|f_c(2)\rangle$ configuration.

We use the model above to perform calculations for the ionization–excitation of helium to the 2p, 3p and 4p states by fast proton and antiproton impact. There are no experimental data for antiprotons, but our result for velocities above 5 au can be compared to the data obtained with the same velocity electrons.

Figure 1 represents our calculated cross sections for the production of the 2p states of the He^+ ion by proton and antiproton impact obtained with the present model along with the previous results without the TS1 mechanism [10] and the experimental data [1]. In spite of the fact that the agreement with the experimental data is not perfect, results are much improved by the inclusion of the TS1 mechanism in the calculations. The results for negative projectiles at 5 au velocity have been doubled, but this value is still below the experimental cross sections for electrons by 50%. The ratio between cross sections for negative and positive projectiles at the same velocity has increased from 1.1 to 1.25, which is still far from the experimental ratio of 1.6.

The experimental data of Bailey *et al* [1] are normalized to the absolute measurements of Forand *et al* [15]. As pointed out by Dogan *et al* [16], although the different absolute measurements lie within their combined errors, there are concerns about the absolute values of the ionization–excitation cross sections derived from undispersed VUV radiation. This possible uncertainty in the experimental data suggests that comparing the ratio of the cross sections obtained with negative and positive projectiles to the experiments would be a more reliable test of the theory than the comparison of absolute values. Furthermore, experiments determine an emission cross section, which includes cascade contributions, while theory determines a $He^+(2p)$ excitation cross section. This suggests that theoretical result should be lower than the experimental data.

It is interesting to remark that the existing theoretical cross sections for electron projectiles do not agree with each other [2–5]. In figure 2 we have plotted the results of our calculations along these theoretical data for electron impact ionization–excitation, in spite of the fact that our model is valid mainly for impact with heavy particles, and direct comparison has sense

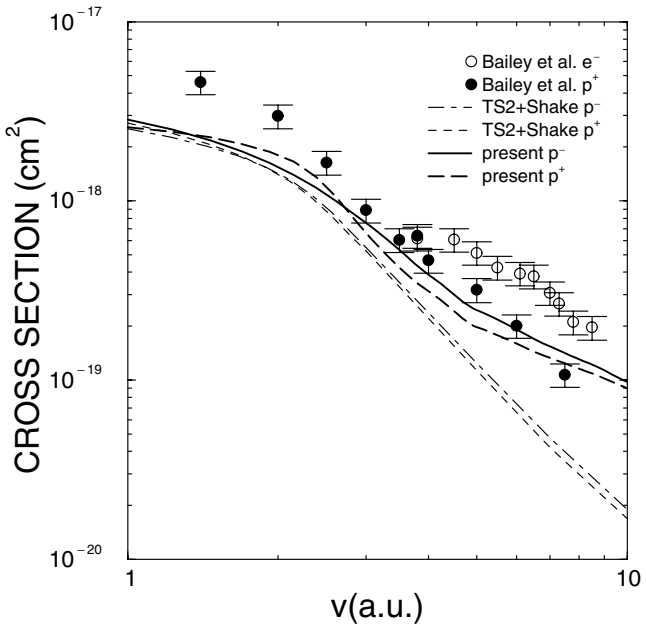

Figure 1. Calculated cross sections for the ionization–excitation of helium to the 2p state by proton and antiproton impact as a function of the projectile velocity along with the experimental data of Bailey *et al* [1]. The present calculations include the TS1 mechanism, while the previous ones account only for shake and TS2.

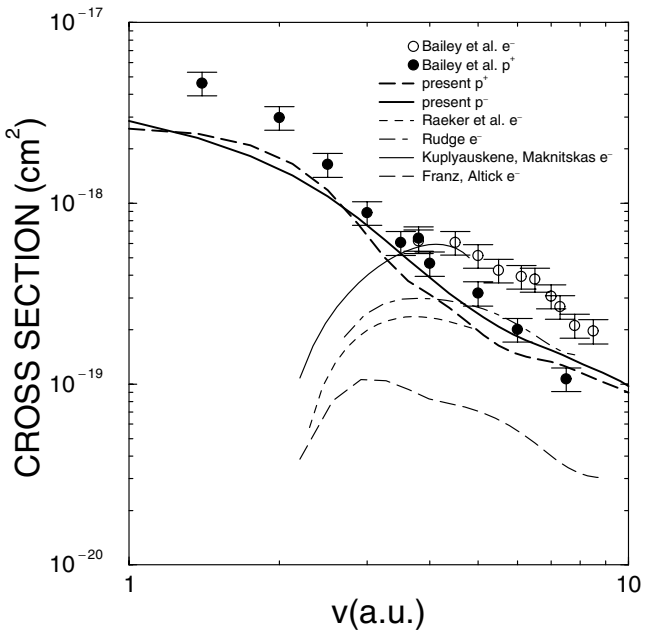

Figure 2. Calculated cross sections for the ionization–excitation of helium to the 2p state by proton and antiproton impact as a function of the projectile velocity along with the experimental data of Bailey *et al* [1] and theoretical results for equi-velocity electron impact [2–5].

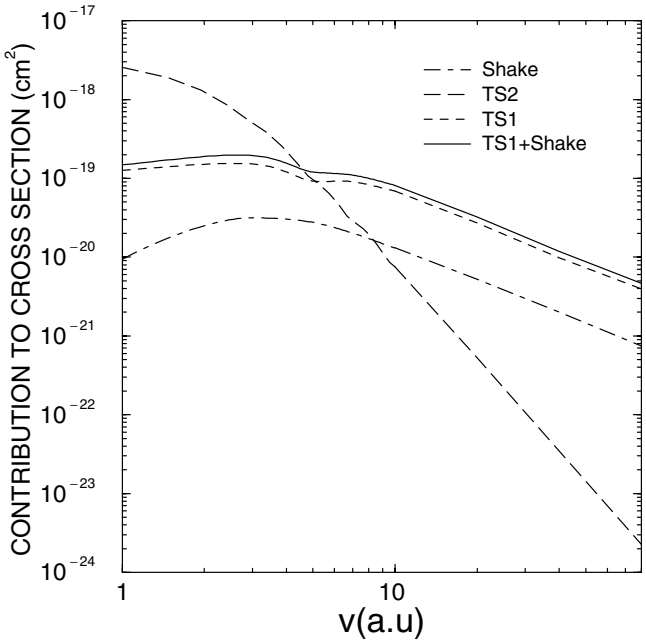

Figure 3. Contributions of different mechanisms to the cross section of the ionization–excitation of helium to the 2p state as a function of the projectile velocity.

only for velocities above 5 au. These theoretical total cross sections for electron impact are all first-order calculations, with a more or less sophisticated description of the initial state and of the ejected electron–residual ion interaction. We have to point out the comments of Dogan *et al* [16] on these results. In the calculations the correct description of both the initial and final states is essential. Raeker *et al* [4] use the R-matrix (close-coupling) expansion for the ejected-electron–residual-ion system. Including more states in the close-coupling expansion reduces the the cross section, leading to worse agreement with the experiment. This is the reason why Kuplyauskene and Maknitskas [3], omitting channel coupling completely, obtain cross sections in good agreement with the experimental data.

Since the publication of these first Born results, some authors have performed calculations also in second order [5–8], obtaining much better agreement with experiment. However, these results are for triple-differential cross sections, only for some certain energies and angles, and are not directly comparable with our total cross sections.

Returning to the discussion for figure 2, the comparison between our and other theoretical cross sections can only be made with some care. We are comparing our second-order results for proton and antiproton impact with first-order results for electron impact.

The relative importance of different mechanisms in the ionization–excitation as a function of the projectile velocity is shown in figure 3. The first-order contribution becomes larger than the second-order one above 5 au velocity. Below this velocity the second-order (TS2) mechanism is dominant. TS1 is very important in the first-order amplitude. Its contribution is by an order of magnitude larger than the shake contribution at all velocities.

Cross sections for the ionization–excitation to the 3p state are represented in figure 4. Here the agreement for negative projectiles with the experimental data and the calculations of Raeker *et al* [4] at sufficiently high velocities is very good, but the ratio between cross sections

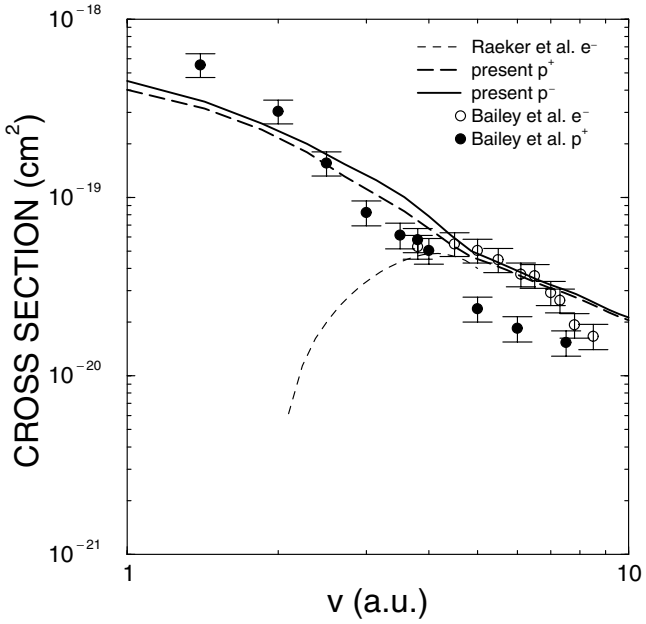

Figure 4. Same as figure 2, but for the 3p state.

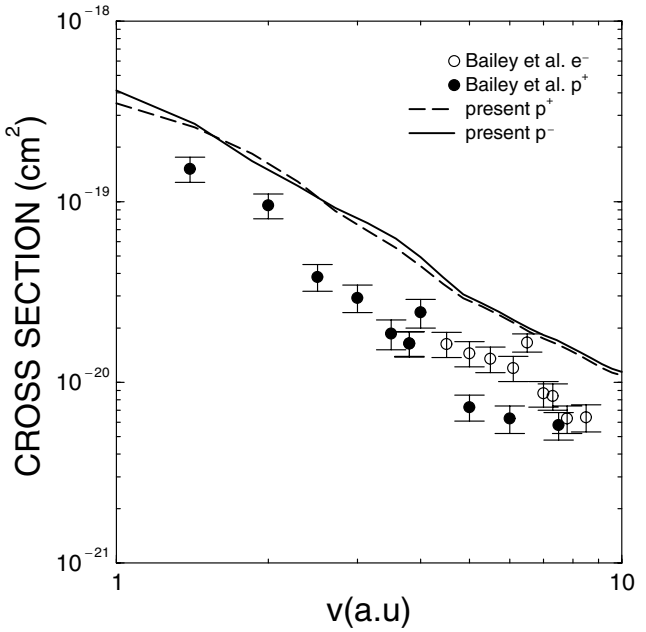

Figure 5. Same as figure 2, but for the 4p state.

for antiprotons and protons is too small compared to the experimental ratio. The situation seems to be similar for the ionization–excitation to the 4p state (figure 5), but in this case the experimental data are much more spread, and no certain conclusion can be formulated.

In conclusion, including the TS1 mechanism in the description of the ionization–excitation process, our previous results [10] are much improved. However, no perfect agreement with experiment is achieved. Possibly, the perturbational description of the final-state correlation is not accurate enough if the ejected electron is very slow. Finally, we should emphasize that our theoretical calculation is the only one for ionization–excitation studying the dependence of the cross section on the sign of the projectile charge.

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