Antiproton stopping power data for radiation therapy simulations

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Abstract

Stopping powers of H, He, H₂, and H₂O targets for antiprotons have been calculated using a convergent close-coupling method. For He and H₂ targets electron-electron correlations are fully accounted for using a multiconfiguration approximation. Two-electron processes are included using an independent-event model. The water molecule is described using a neon-like structure model with a pseudo-spherical potential. Results are tabulated for the purpose of Monte Carlo simulations to model antiproton transport through matter for radiation therapy.

Keywords: stopping power, antiproton, hadron therapy

1. Introduction

Gaining an understanding of how heavy charged particles lose energy as they travel through matter is fundamentally important for a wide range of fields including astrophysics [1], aviation and space exploration [2], and medical radiation therapy [3, 4]. Developments at CERN with the ability of the antiproton decelerator (AD) to produce low-energy antiprotons have significantly increased interest in antiproton physics (see the review of Kirchner and Knudsen [5]). Of particular interest to us is the potential for antiprotons to be used as a new superior modality in radiation therapy.

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It is now well known that hadrons, like protons and carbon ions, provide a significant benefit over photons when used for radiation therapy purposes. However, in terms of antiprotons the picture is not yet complete. Antiprotons were first proposed as a potential radiotherapy tool in 1984 by Gray and Kalogeropoulos [6] due to Monte Carlo simulations showing enhancement of physical dose in the Bragg-peak region. As with other hadrons, antiprotons deposit most of their kinetic energy before coming to rest. Additionally, as they come to rest they annihilate producing π -mesons. These π -mesons will in turn be absorbed by the nucleus on which the antiprotons annihilate resulting in the emission of nuclear fragments with high linear energy transfer (LET). These high-LET secondary particles cause enhancement of the physical dose and relative biological effectiveness (RBE). However, high-LET secondary particles will also be produced by neutrons and the contribution of these particles to the background dose must also be considered.

To fully assess the effectiveness of antiprotons for use in radiation therapy the ACE collaboration has been conducting experiments using the AD facility at CERN. At present they have shown that for the same entrance dose antiprotons give an enhanced biological effect of 4 in the peak region compared with protons [7, 8]. Additionally, due to the limited capabilities of experiment further investigation is carried out using Monte Carlo codes such as FLUKA [9, 10] and SHIELD-HIT [11, 12]. The limiting factor in these Monte Carlo codes is the underlying physics which drive the simulations. Therefore, the theoretical models on which these Monte Carlo codes are based need to be of sufficient accuracy to ensure reliability of their output. To this end, in this paper we present detailed tabulated stopping power data for their implementation into Monte Carlo codes to model antiproton transport through matter.

Specifically, in this paper we present data on antiproton stopping power of H, He, H₂, and H₂O. The data has been generated using a semiclassical time-dependent convergent close-coupling (CCC) method. For He and H₂ we use a multiconfiguration treatment, which fully accounts for electron-electron correlations. We also include ionisation with excitation and double ionisation

processes via an independent event model. In addition, for H_2 we account for all possible orientations of the molecule using an analytic orientation averaging technique and include vibrational excitations. For H_2O we use a neonization method [13], which approximates the water molecule as a Ne-like atom in a pseudo-spherical potential with six p -shell electrons above a frozen Hartree-Fock core. The H_2O data will be particularly useful to assist investigations into antiproton radiation therapy.

The paper is set out as follows: section 2 outlines the method, section 3 presents the results with tabulated data, and finally, section 4 draws conclusions.

2. Method

For the calculation of stopping powers we employ the time-dependent CCC method. The method has been extensively described in the literature for various targets [14–18]. Here we give a brief outline only.

2.1. Time-dependent convergent close-coupling method in impact-parameter representation

The semiclassical impact-parameter approach treats the target electrons fully quantum mechanically, however the motion of the incident antiproton is assumed to follow a straight line and is treated classically. The total electronic scattering wave function is expanded in a complete basis of target pseudostates. Substitution of this expansion into the time-dependent Schrödinger equation leads to a set of coupled-channel differential equations for the time-dependent expansion coefficients A_f . The set is then solved with the condition that the target is initially in the ground state. From these expansion coefficients we can obtain the probability for the target transitioning from the ground state to some final state f at a specific impact parameter b by

$$p_f(b) = |A_f(t = +\infty, b)|^2. \quad (1)$$

This method is used for H, He, and H_2O . The multi-center H_2O problem is reduced to a central one as described in the next subsection. For H_2 the expansion

coefficients A_f will depend on the relative coordinate of the target nuclei $\mathbf{d}$. To account for all possible orientations of the H_2 molecule we take the additional step of factoring out the orientation-dependent parts from our equations and analytically integrating over all orientations of the molecular axis. We then obtain a system of differential equations for the orientation-independent part of the scattering amplitudes $\mathcal{A}_{f\lambda\mu}$. Orientation-averaged transition probabilities at fixed internuclear distance d are then defined by

$$p_f(b) = \sum_{\lambda\mu} \frac{1}{2\lambda+1} |\mathcal{A}_{f\lambda\mu}(t=+\infty, b, d)|^2, \quad (2)$$

where λ and μ are limited by the maximum allowed total orbital angular momentum l_{max} . Integrating Eqs. (1) and (2) over impact parameters yields the scattering cross section:

$$\sigma_f = 2\pi \int_0^\infty p_f(b) b db. \quad (3)$$

To calculate the two-electron processes of ionisation with excitation (IE) and double ionisation (DI) we employ an independent-event model. In this model two-electron processes are considered a two-step process. The first step is single ionisation of the target and the second is excitation or ionisation of the now singly-ionised target. Therefore, the probability of such a process is the product of two probabilities: the total single ionisation probability of the target, p_{ion} , and the probability of the ionised target transitioning from the ground state to some final state k , p_k^+ . The cross section is then

$$\sigma_k^+ = 2\pi \int_0^\infty p_{\text{ion}}(b) p_k^+(b) b db. \quad (4)$$

2.2. Target structure calculations for H , He , H_2 , and H_2O

For hydrogen the pseudostates are obtained by diagonalising the target Hamiltonian H_t for each orbital angular momentum $l \leq l_{\text{max}}$ such that

$$\langle \Phi_\alpha | H_t | \Phi_\beta \rangle = \epsilon_\alpha \delta_{\alpha\beta}. \quad (5)$$

The pseudostates $\Phi(r)$ are linear combinations of the complete Laguerre basis

$$\xi_{kl}(r) = \left(\frac{\lambda_l (k-1)!}{(2l+1+k)!} \right)^{1/2} (\lambda_l r)^{l+1} L_{k-1}^{2l+2}(\lambda_l r) \exp(-\lambda_l r/2), \quad (6)$$

where $L_{k-1}^{2l+2}(\lambda_l r)$ are the associated Laguerre polynomials and index k ranges from 1 to N_l , the maximum number of basis functions. The advantage of this choice of basis is that it allows us to model the whole target spectrum by simply increasing N_l . As N_l increases the negative-energy states converge to the true eigenstates, while the positive-energy states will provide an increasingly dense discretisation of the target continuum.

For helium we obtain two-electron target states using the configuration-interaction (CI) expansion

$$\Phi_n = \sum_k C_k^n \tilde{\Phi}_k. \quad (7)$$

The CI expansion coefficients C_k^n are obtained by diagonalisation of the target Hamiltonian in the basis of configurations $\tilde{\Phi}_k$. The configurations $\tilde{\Phi}_k$ are antisymmetrised two-electron configurations built by orbital angular momentum and spin coupling of one-electron functions. The one-electron basis is constructed from the Laguerre functions ξ_{kl} defined in Eq. (6). For our calculations we use a multiconfiguration approach, meaning we allow several inner electron orbitals in our two-electron configurations. The major advantage of a multiconfiguration approach over a simpler frozen core approach is the high accuracy of the ground state wave function.

For the H_2 target we use the Born-Oppenheimer approximation and fix the internuclear distance at the ground-state equilibrium value of $d = 1.4487$ a.u. Similar to He, target pseudostates are obtained via diagonalisation of the H_2 Hamiltonian in a set of antisymmetrised two-electron configurations constructed from one-electron orbitals. Target pseudostates are first calculated in the body frame using a CI expansion about the midpoint of the internuclear axis, then converted to laboratory frame via a rotation operator. Again, we use a multiconfiguration approach by allowing several inner electron orbitals, whereas the number of one-electron states of the outer electron are as large as required to ensure converged results.

For H_2O we use a neon-like description of the target [13]. The water molecule is described as a dressed pseudo-spherical atom, which reduces a multi-center

problem to a central one. The multi-center nuclei Coulomb potential of H₂O is approximated with

$$V_{\text{H}_2\text{O}} = -\frac{8}{r} - \frac{2(1-\eta)\Theta(R_{\text{H}}-r)}{R_{\text{H}}} - \frac{2(1-\eta e^{1-r/R_{\text{H}}})\Theta(r-R_{\text{H}})}{r}, \quad (8)$$

where R_{H} is the distance between the oxygen atom and either of the two hydrogen atoms and Θ is the Heaviside step function. The parameter η is introduced to account for the deviation of the target potential from spherical symmetry and is varied to match the experimentally measured value for the ground state ionisation energy of H₂O. As it was done for neon [14, 18] we use a model of six p -shell electrons above a frozen Hartree-Fock core with only one-electron excitations from the outer p shell allowed. The appropriate H₂O core wave functions presented in [13] are used.

2.3. Stopping power

The stopping power is the energy loss per unit path length and is defined as

$$-\frac{dE}{dx} = NS(E_0), \quad (9)$$

where $S(E_0)$ is referred to as the stopping cross section and is dependent on the incident energy of the projectile E_0 . The stopping cross section is related to the stopping power by the number of target atoms or molecules per cubic meter, N . The total stopping cross section can be the sum of various contributions.

The electronic stopping cross section is the dominant contribution and is the energy loss associated with the excitation and ionisation of the targets electrons. As discussed previously [14, 15], in the CCC formalism the electronic stopping cross section is given by

$$S_{\text{e}}(E_0) \approx \sum_{f=1}^{N_{\text{T}}} (\epsilon_f - \epsilon_i) \sigma_f + \sum_{k=1}^{N_{\text{T}}^+} (\epsilon_k - \epsilon_i^+) \sigma_k^+. \quad (10)$$

The first summation over the total number of target pseudostates N_{T} represents single excitation and ionisation processes, where the target electron transitions from the ground state of energy ϵ_i to some final state of energy ϵ_f , and σ_f is

the scattering cross section for that transition. For He and H₂, ionisation with excitation and double-ionisation processes are also included, and are represented by the second summation in Eq. (10). Here σ_k^+ is the scattering cross section for one electron being ejected and the other transitioning from the ground state of the ionised target with energy ϵ_i^+ , to some final state of energy ϵ_k . If ϵ_k is positive, this represents double ionisation, while if ϵ_k is negative it corresponds to ionisation with excitation. For H₂, ionisation with excitation processes may be referred to as dissociative ionisation due to the repulsive nature of the H₂⁺ excited states.

Additionally, a H₂ target can be vibrationally excited by the antiproton during the collision process, which will result in further energy loss of the projectile. In this paper we only consider vibrational transitions within the electronic ground state, as these will give the most dominant contribution. The vibrational stopping cross section can be written in the approximate form

$$S_{\text{vib}}(E_0) \approx \sum_{\nu=0}^{N_{\text{vib}}} (\varepsilon_{\nu} - \varepsilon_0) \langle \chi_{\nu}(d) | \sqrt{\sigma_{\text{el}}^{\text{av}}(d)} | \chi_0(d) \rangle, \quad (11)$$

where $\chi_0(d)$ is the ground state vibrational eigenfunction of energy ε_0 , $\chi_{\nu}(d)$ is an excited vibrational eigenfunction of energy ε_{ν} , N_{vib} is the number of included vibrational excitations, and $\sigma_{\text{el}}^{\text{av}}(d)$ is the elastic scattering cross section analytically averaged over all molecular orientations. Approximating the scattering amplitude with $\sqrt{\sigma_{\text{el}}^{\text{av}}(d)}$ is similar to the approach used to calculate kinetic energy release in electron-H₂⁺ collisions [19]. Our current work involving positron-H₂ scattering is showing this to be an accurate approximation.

3. Results

For calculation of the antiproton-hydrogen electronic stopping cross section we require the maximum orbital angular momentum of the target states to be 6 with $N_l = 30 - l$. This leads to a total of 1267 coupled differential equations being solved. In Table 1 we give the electronic stopping cross section data

Table 1: Tabulated data for the antiproton-H stopping cross section, where $a[b]$ represents $a \times 10^b$.

E_0 (keV)	SCS ($10^{-15} \text{eVcm}^2/\text{atom}$)	
	Excitation	Ionisation
1	6.490[−1]	1.981[+0]
2	7.016[−1]	2.346[+0]
4	8.086[−1]	2.731[+0]
10	9.926[−1]	3.317[+0]
15	1.081[+0]	3.575[+0]
20	1.140[+0]	3.720[+0]
30	1.206[+0]	3.868[+0]
50	1.226[+0]	3.857[+0]
70	1.199[+0]	3.693[+0]
100	1.109[+0]	3.378[+0]
250	7.522[−1]	2.148[+0]
500	4.921[−1]	1.274[+0]
700	3.827[−1]	9.577[−1]
1000	2.908[−1]	6.973[−1]
2000	1.738[−1]	3.677[−1]
5000	8.391[−2]	1.576[−1]
7000	6.210[−2]	1.151[−1]
10000	4.439[−2]	8.265[−2]

separated into two components: the stopping cross section for excitation and for ionisation. Results are plotted in Figure 1.

For calculation of the antiproton-helium electronic stopping cross section we require the maximum orbital angular momentum of the target states to be 6 with $N_l = 20 - l$. In our multiconfiguration structure model we also needed to include 5 *s*, 4 *p*, and 3 *d* inner electron orbitals. This leads to a total of 1288 coupled differential equations being solved. With this structure model we obtain a ground state ionisation energy of 24.540 eV, which is very close to the exper-

Table 2: Tabulated data for the antiproton-He stopping cross section, where $a[b]$ represents $a \times 10^b$.

E_0 (keV)	SCS ($10^{-15} \text{eVcm}^2/\text{atom}$)			
	SE	SI	IE	DI
1	2.266[−1]	4.197[−1]	2.716[−2]	1.016[−2]
2	2.417[−1]	7.705[−1]	5.922[−2]	3.016[−2]
4	2.579[−1]	1.210[+0]	7.986[−2]	8.072[−2]
10	2.890[−1]	1.910[+0]	1.049[−1]	1.633[−1]
15	3.078[−1]	2.284[+0]	1.116[−1]	2.055[−1]
20	3.253[−1]	2.565[+0]	1.138[−1]	2.340[−1]
30	3.558[−1]	2.984[+0]	1.131[−1]	2.696[−1]
50	3.960[−1]	3.474[+0]	1.050[−1]	2.972[−1]
70	4.143[−1]	3.693[+0]	9.495[−2]	2.978[−1]
100	4.130[−1]	3.777[+0]	7.999[−2]	2.786[−1]
250	3.377[−1]	3.270[+0]	3.752[−2]	1.695[−1]
500	2.391[−1]	2.346[+0]	1.488[−2]	7.919[−2]
700	1.938[−1]	1.886[+0]	8.685[−3]	4.886[−2]
1000	1.540[−1]	1.451[+0]	4.686[−3]	2.750[−2]
2000	9.285[−2]	8.229[−1]	1.292[−3]	7.936[−3]
5000	4.514[−2]	3.673[−1]	2.142[−4]	1.329[−3]
7000	3.557[−2]	2.599[−1]	1.082[−4]	6.652[−4]
10000	2.675[−2]	1.893[−1]	5.269[−5]	3.255[−4]

imentally measured value of 24.586 eV. As described in section 2.1, to model two-electron processes using the independent-event model we need to perform calculations for He^+ . For these calculations we required the maximum orbital angular momentum to be 4 with $N_l = 20 - l$. This reduction in the required maximum orbital angular momentum is due to the increase in the binding energy of the target electron. In Table 2 we give the electronic stopping cross section separated into four components: the stopping cross section for single excitation, single ionisation, ionisation with excitation, and double ionisation.

Results are plotted in Figure 2.

Figure 1: Stopping cross section associated with excitation and ionisation processes in antiproton-H collisions.

Figure 2: Stopping cross section associated with single excitation, single ionisation, ionisation with excitation, and double ionisation processes in antiproton-He collisions.

Table 3: Tabulated data for the antiproton-H₂ stopping cross section, where $a[b]$ represents $a \times 10^b$.

E ₀ (keV)	SCS (10^{-15} eVcm ² /molecule)				
	vib	SE	SI	IE	DI
1	3.182[−1]	2.268[−1]	3.113[−1]	2.863[−2]	6.221[−3]
2	2.480[−1]	3.834[−1]	6.933[−1]	8.004[−2]	3.159[−2]
4	1.837[−1]	5.412[−1]	1.497[+0]	1.666[−1]	1.174[−1]
10	1.196[−1]	8.745[−1]	3.037[+0]	3.150[−1]	3.267[−1]
15	9.501[−2]	1.067[+0]	3.871[+0]	3.647[−1]	4.386[−1]
20	8.013[−2]	1.214[+0]	4.464[+0]	3.831[−1]	5.096[−1]
30	6.005[−2]	1.391[+0]	5.213[+0]	3.807[−1]	5.790[−1]
50	3.500[−2]	1.520[+0]	5.840[+0]	3.351[−1]	5.998[−1]
70		1.534[+0]	5.929[+0]	2.849[−1]	5.638[−1]
100		1.466[+0]	5.704[+0]	2.225[−1]	4.877[−1]
250		1.072[+0]	3.978[+0]	8.095[−2]	2.217[−1]
500		7.187[−1]	2.456[+0]	2.730[−2]	8.122[−2]
700		5.690[−1]	1.871[+0]	1.506[−2]	4.567[−2]
1000		4.297[−1]	1.381[+0]	7.774[−3]	2.383[−2]
2000		2.511[−1]	7.436[−1]	2.038[−3]	6.235[−3]
5000		1.144[−1]	3.190[−1]	3.298[−4]	1.007[−3]
7000		8.486[−2]	2.318[−1]	1.543[−4]	4.861[−4]
10000		5.959[−2]	1.680[−1]	7.545[−5]	2.387[−4]

For calculation of the antiproton-H₂ electronic stopping cross section we require the maximum orbital angular momentum of the target states to be 5 with $N_l = 20 - l$. For two-electron configurations in our multiconfiguration approach we include the $1s$, $2s$, $2p$, $3s$, $3p$, and $3d$ orbitals for the inner electron. This leads to a total of 843 coupled differential equations being solved. With this model we obtain a two-electron ground state ionisation energy of 31.7007 eV, which is close to the accurate value of 31.9600 eV [20]. Again, to model two-electron processes we perform antiproton-H₂⁺ calculations. For these we also use

$l_{\max} = 5$ and $N_l = 20 - l$. For both H_2 and H_2^+ calculations we use the same internuclear separation of 1.4487 a.u. as required by the independent-event model. In addition to the electronic stopping cross section we calculate the vibration stopping cross section associated with vibrational excitations with the ground electronic state. In Table 3 we give the vibrational stopping cross section as well as electronic stopping cross section separated into four components: the stopping cross section for single excitation, single ionisation, ionisation with excitation, and double ionisation. Results are plotted in Figure 3.

Figure 3: Stopping cross section associated with vibrational excitation, single excitation, single ionisation, ionisation with excitation, and double ionisation processes in antiproton- H_2 collisions.

For calculation of the antiproton- H_2O electronic stopping cross section we require the maximum orbital angular momentum of the target states to be 4 with $N_l = 20 - l$. This leads to a total of 1112 coupled differential equations being solved. With our model the $2p$ -subshell ionisation energy of H_2O is 12.6 eV which is the same as the experimentally measured value since this energy was used to generate the target structure. Using a neon-like model as described in section 2.2 we have only calculated single electron transitions for the outer p

shell. In Table 4 we give the electronic stopping cross section separated into two components: the stopping cross section for excitation and for ionisation. Results are plotted in Figure 4.

Table 4: Tabulated data for the antiproton-H₂O stopping cross section, where $a[b]$ represents $a \times 10^b$.

E ₀ (keV)	SCS (10^{-15} eVcm ² /molecule)	
	Excitation	Ionisation
1	3.789[−1]	1.435[+0]
5	5.973[−1]	2.088[+0]
7	6.685[−1]	2.357[+0]
10	7.703[−1]	2.871[+0]
15	9.142[−1]	3.830[+0]
20	1.045[+0]	4.701[+0]
30	1.224[+0]	6.206[+0]
50	1.390[+0]	8.399[+0]
70	1.388[+0]	9.763[+0]
100	1.320[+0]	1.082[+1]
150	1.147[+0]	1.121[+1]
200	1.008[+0]	1.089[+1]
300	8.260[−1]	9.754[+0]
500	6.029[−1]	7.710[+0]
700	4.714[−1]	6.311[+0]
1000	3.571[−1]	4.966[+0]
3000	1.546[−1]	2.130[+0]
5000	1.008[−1]	1.394[+0]
7000	7.545[−2]	1.050[+0]
10000	5.349[−2]	7.725[−1]

Figure 4: Stopping cross section associated with one-electron excitation and ionisation processes in antiproton-H₂O collisions.

4. Conclusion

In conclusion we have presented data on stopping cross sections of H, He, H₂, and H₂O for antiprotons calculated using the CCC method. For He and H₂ we fully account for electron-electron correlations and include two-electron processes via an independent-event model. Additionally, for H₂ we analytically average over all possible orientations of the molecule as well as including the contribution to the stopping cross section from vibrational excitations. For H₂O we use a neon-like model of six *p*-shell electrons above a frozen Hartree-Fock core with only one-electron excitations from the outer *p* shell allowed. For all these calculations we have presented tabulated data broken down into various different contributions to the total stopping cross section. The data can be implemented into existing Monte Carlo codes to model antiproton transport through matter for radiation therapy simulations, contributing to the assessment of the effectiveness of antiprotons for use in hadron therapy.

Currently we are working towards extending the CCC method to the calcula-

tion of proton stopping powers. Calculations involving protons are significantly more difficult due the need to include rearrangement channels. Additionally, one must take into account the formation of a hydrogen projectile after a rearrangement collision, which further increases the complexity of calculations. The development of a two-center CCC method for proton-hydrogen collisions, which includes charge exchange, is complete. However, stopping power calculations of atomic hydrogen with atomic hydrogen are still under development.

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