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Convergent close-coupling calculations of positron scattering from carbon

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Introduction: Motivation

- Positrons are important in medical industry - positron emission tomography (PET) and positherapy.
- Quantifiable data of scattering processes occurring in these technologies scarce and largely unknown.
- Many modern scattering methods require accurate cross sections for each constituent atom to model the interactions occurring with positrons and biomolecules in these cases.
- Positronium-formation important, in particular, as its decay results in 80 % of detected gamma rays in PET scans.
- With this data can increase accuracy of PET scans and model positherapy.

Introduction: CCC-method

- Have extended atomic single-center convergent close-coupling (CCC) to calculate scattering from multi-electron atoms, without requirement of frozen-core.
- Achieved through utilisation of Multiconfigurational Hartree-Fock (MCHF) method of Fischer *[Comp. Phys. Comm. 64, 369 (1991)]* and MULT codes of Zatsarinny *[Comp. Phys. Comm. 174, 273 (2006))]*
- Have calculated comprehensive scattering data for positron scattering from carbon.
- Addressed deficits in single-center method through use of complex model potential.

Method: Atomic structure calculation

- Target Hamiltonian:
  $H_{\mathrm{T}} = \sum_{i=1}^{N_{\mathrm{e}}} \left(-\frac{1}{2}\nabla_i^2 - \frac{Z}{r_i} \right) + \sum_{i>j=1}^{N_{\mathrm{e}}} \frac{1}{|\boldsymbol{r}_i - \boldsymbol{r}_j|}$
- Configuration-interaction representation of wave function:
  $\Phi_n^N(x_1, \ldots, x_{N_{\mathrm{e}}}) = \sum_{i=1}^{N} C_i^{(n)} \phi_i(x_1, \ldots, x_{N_{\mathrm{e}}})$
- Radial functions obtained via MCHF calculation or Laguerre basis functions:

$$\varphi_{k\ell}(r) = \sqrt{\frac{\alpha_\ell (k-1)!}{(k+\ell)(k+2\ell)!}} (2\alpha_\ell r)^{\ell+1} \times e^{-\alpha_\ell r} L_{k-1}^{2\ell+1}(2\alpha_\ell r), \quad k = 1, \ldots, N_\ell. \tag{1}$$

- Diagonalize target Hamiltonian to obtain CI coefficients.
- Pseudostates satisfy: $\langle \Phi_n^N | H_{\mathrm{T}} | \Phi_m^N \rangle = \epsilon_n^N \delta_{nm}$

Method: Single-Center Convergent Close-Coupling

- Total scattering Hamiltonian: $H = H_{\mathrm{T}} - \frac{1}{2}\nabla_0^2 + V$
- Schrödinger equation for total scattering wave function: $(H - E)|\Psi_i^{(+)}\rangle = 0$
- Solve through expanding wavefunction in set of target pseudostates:

$$\Psi_i^{N(+)} = \sum_{n=1}^{N} F_n^{N(+)}(\boldsymbol{r}_0)\Phi_n^N(\hat{\boldsymbol{r}}_1, \ldots, \hat{\boldsymbol{r}}_N)$$

- Coupled Lippman-Schwinger equations for the T matrix:

$$\langle \boldsymbol{k}_f \Phi_f^N | T | \Phi_i^N \boldsymbol{k}_i \rangle = \langle \boldsymbol{k}_f \Phi_f^N | V | \Phi_i^N \boldsymbol{k}_i \rangle + \sum_{n=1}^{N} \int d\boldsymbol{k} \frac{\langle \boldsymbol{k}_f \Phi_f^N | V | \Phi_n^N \boldsymbol{k} \rangle \langle \boldsymbol{k} \Phi_n^N | T | \Phi_i^N \boldsymbol{k}_i \rangle}{E^{(+)} - \epsilon_n^N - \epsilon_k}$$

- Perform partial-wave expansion of the projectile plane waves and solve the Lippmann-Schwinger equations per total scattering-system angular momentum J.

Method: Single-Center Convergent Close-Coupling Drawbacks

- Approach unstable between positronium-formation threshold and ionization threshold.
- Occurs because boundary conditions with channels corresponding to positive-energy pseudostates are closed while positronium-formation channels are open.
- Positronium-formation included implicitly therefore cannot seperate positronium-formation and direct ionization.

Method: CCC-scaled Complex Model Potential

- Complex potential: $V_{\text{opt}}(r, E_i) = V_{\text{st}}(r) + V_{\text{pol}}(r) + iV_{\text{abs}}(r, E_i)$
- Static potential:
  $V_{\text{st}}(r) = \frac{Z}{r} - 4\pi \left(\frac{1}{r} \int_0^r dr' \rho(r') r'^2 + \int_0^\infty dr' \rho(r') r' \right)$
- Polarization potential: $V_{\text{pol}}(r) = -\frac{\alpha_{\text{D}}}{2(r^2+d^2)^2}$
- Absorption potential:

$$V_{\text{abs}}(r, E_i) = -\rho(r) \left[\sqrt{\frac{T_{loc}}{2}} \left(\frac{8\pi}{10 k_F^3(r) E_i} \right) \times \theta(k_i^2 - k_F^2(r) - 2\Delta)(A_1 + A_2 + A_3) \right]$$

- Staszewska *et al.* *[J. Chem. Phys. 81, 335 (1984)]*.
- Absorption threshold: $\Delta(E) = \Delta_{\text{e}} - (\Delta_{\text{e}} - \Delta_{\text{p}}) e^{-(E_i - \Delta_{\text{p}})/E_m}$
- Form from Chiari *et al.* *[J. Phys. B: At Mol Opt 45, 215206 (2012)]*

Method: CCC-scaled Complex Model Potential

- Solve Lippman-Schwinger equation:

$$\langle \boldsymbol{k}_f | T | \boldsymbol{k}_i \rangle = \langle \boldsymbol{k}_f V | \boldsymbol{k}_i \rangle + \int d\boldsymbol{k} \frac{\langle \boldsymbol{k}_f | V | \ \boldsymbol{k} \rangle \langle \boldsymbol{k} | T | \boldsymbol{k}_i \rangle}{k_i^2/2 - k^2/2 + i0}$$

- Obtain partial-wave Lippman-Schwinger equation:

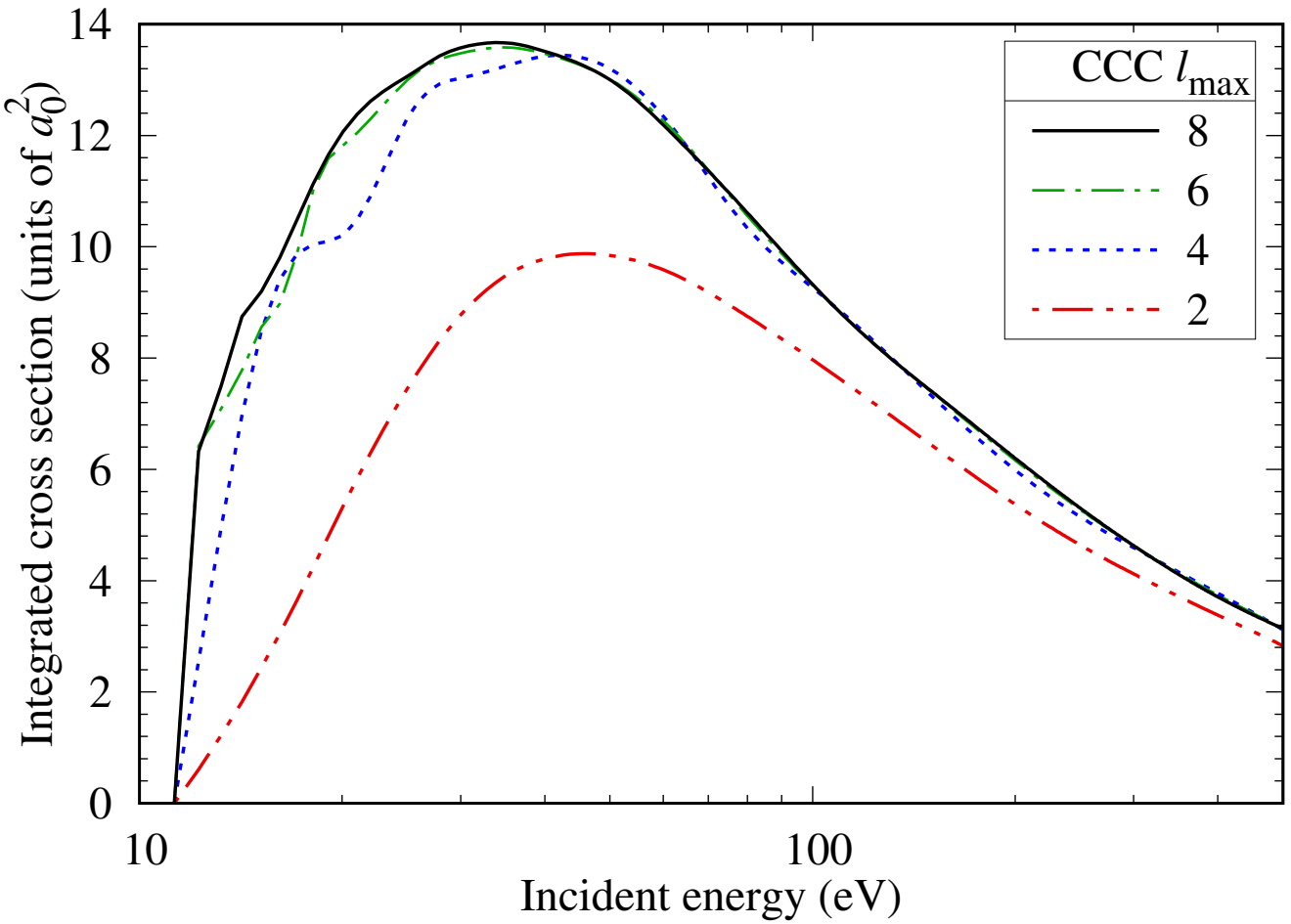
$$T_\ell(k_f, k_i) = V_\ell(k_f, k_i) + \mathcal{P} \int_0^\infty dk \frac{V_\ell(k_f, k) T_\ell(k, k_i)}{k_i^2/2 - k^2/2} - \frac{i\pi}{k_i} V_\ell(k_i, k_i) T_\ell(k_i, k_i)$$

- Total cross section: $\sigma_{\text{tot}} = 4\pi^3 \sum_{l=0}^{\infty} (2\ell + 1) |T_\ell|^2$
- Elastic cross section: $\sigma_{\text{el}} = -\frac{4\pi^2}{\sqrt{2E_i}} \sum_{\ell=0}^{\infty} (2\ell + 1) \text{Im}(T_\ell)$
- Calculate positronium-formation with delta-variational technique.
- Scale direct inelastic component to agree at high energy limit with single-center CCC.
- Scale positronium-formation component to agree at the maximum cross section between ionization threshold and 10 eV above it.

Calculation Details

- A MCHF calculation for C^+ was used to obtain orbitals $1s$ to $5s$.
- All other orbitals obtained by Laguerre basis with $N_\ell = 18 - \ell$, $\ell_{max} = 8$, and $\alpha_\ell = 1.0$.
- Configurations included: $2s^2 2pn\ell$ continuum for $\ell \leq 8$, $2s2p^2 n\ell$ for $\ell \leq 4$, $2p^3 n\ell$ for $\ell \leq 2$, $2s2pn\ell' n\ell$ and $2s2pn\ell^2$ for $n\ell$ and $n'\ell'$ between $3s$ and $5s$.
- Included target states with excitation energies 40 eV above ionization threshold, resulting in 943 target states.
- Calculations completed to $J = 10$ partial waves.
- Extrapolated ionization cross section at high energies with a 4571-state Born calculation.

Convergence study



- Convergence study of total ionization cross section for $\ell_{max} = 2$ to $\ell_{max} = 8$.
- Convergence established for $\ell_{max} > 6$ for energies above 15 eV.

Results: Structure

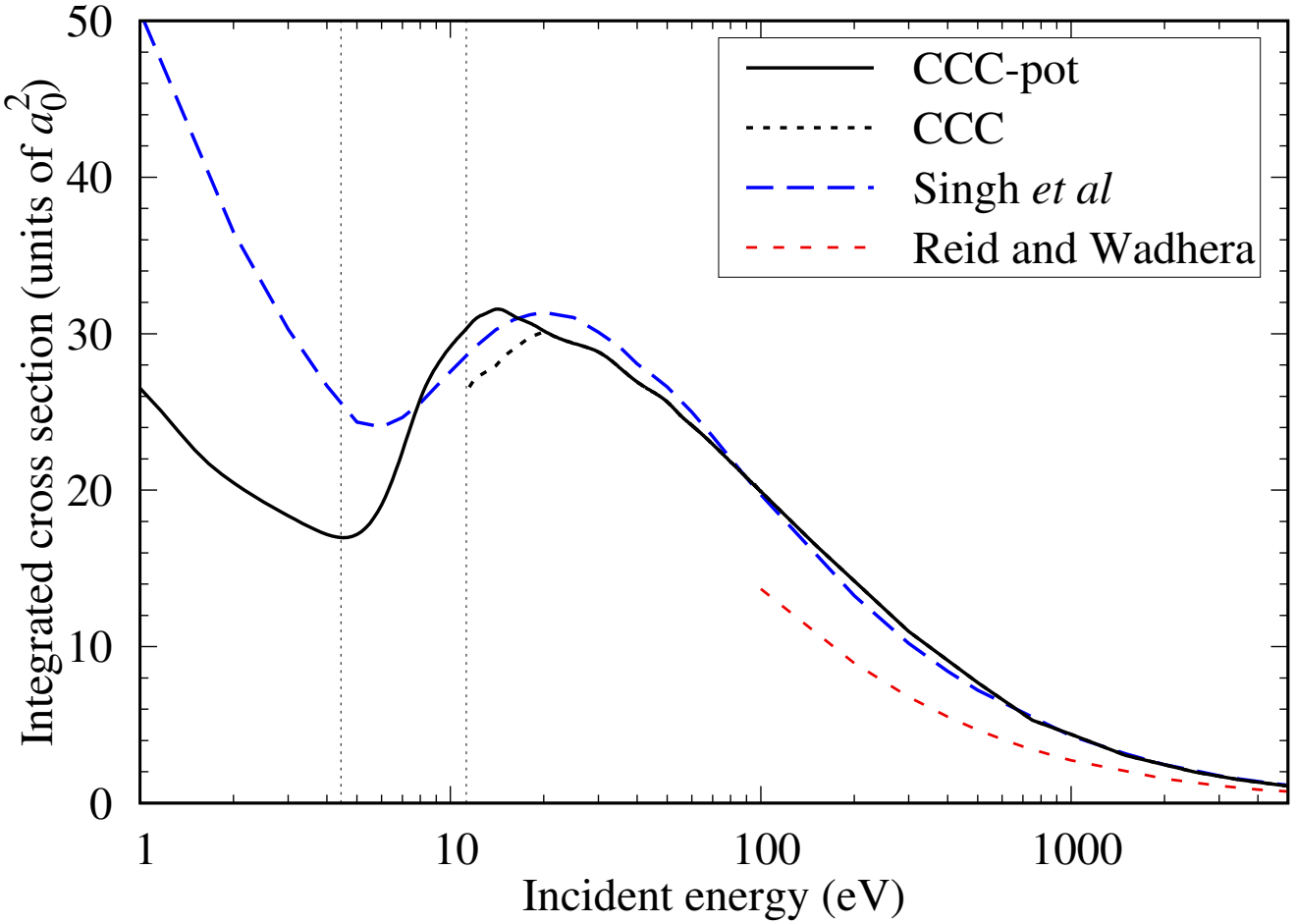
- Excitation energies and oscillator strength for carbon

	State	Term	CCC	Ref. [1]	Ref. [2]	Ref. [3]	NIST. [4]
1	$2s^2 2p^2$	^3P	0.000	0.000	0.000	0.000	0.000
2	$2s^2 2p^2$	^1D	1.372	1.302	1.557	1.545	1.260
3	$2s^2 2p^2$	^1S	2.748	2.629	2.602	2.545	2.680
4	$2s2p^3$	$^5S^o$	3.940	3.963	3.092	3.133	4.179
5	$2s^2 2p3s$	$^3P^o$	7.617	7.527	7.401	8.488	7.481
6	$2s^2 2p3s$	$^1P^o$	7.818	7.750	7.740	8.936	7.680
7	$2s2p^3$	$^3D^o$	7.951	8.004	8.340	8.412	7.942
8	$2s^2 2p3p$	^1P	8.897	8.534	8.451	9.456	8.534
9	$2s^2 2p3p$	^3D	9.032	8.649	8.600	9.589	8.642
10	$2s^2 2p3p$	^3S	9.188	8.775	8.772	9.785	8.767
11	$2s^2 2p3p$	^3P	9.332	8.857	9.309	10.390	8.845
12	$2s2p^3$	$^3P^o$	9.481	9.379	9.517	9.981	9.326
13	$2s^2 2p3p$	^1D	9.553	9.014	9.443	10.757	8.998
14	$2s^2 2p3p$	^1S	9.766	9.172	10.424	11.370	9.168
15	$2s^2 2p3d$	$^1D^o$	10.166	9.614	9.772	10.719	9.627
16	$2s^2 2p4s$	$^3P^o$	10.258	9.673	10.142	10.810	9.683
17	$2s^2 2p3d$	$^3F^o$	10.271	9.687	9.517	10.809	9.695
18	$2s^2 2p3d$	$^3D^o$	10.288	9.705	9.607	10.888	9.705
19	$2s^2 2p4s$	$^1P^o$	10.301	9.685	9.549	10.834	9.709
20	$2s^2 2p3d$	$^1F^o$	10.333	9.716	9.607	10.947	9.732
21	$2s^2 2p3d$	$^1P^o$	10.370	9.748	9.653	10.970	9.758
22	$2s^2 2p3d$	$^3P^o$	10.404	9.840	13.407	11.018	9.830
Ion.	Limit		11.234				11.268
23	$2s2p^3$	$^1D^o$	13.600	12.968	14.470	14.645	
24	$2s2p^3$	$^3S^o$	13.279	13.073	13.407	15.366	13.117
25	$2s2p^3$	$^1P^o$	15.883	15.401	15.927	16.182	

Lower level	Upper level	CCC	Ref. [1]	Ref. [2]	Ref. [3]	NIST. [4]
$2s^2 2p^2\ ^3P$	$2s^2 2p3s\ ^3P^o$	0.146	0.143	0.124	0.154	0.140
	$2s2p^3\ ^3D^o$	0.076	0.073	0.098	0.152	0.072
	$2s2p^3\ ^3P^o$	0.078	0.056	0.028	0.117	0.063
	$2s^2 2p4s\ ^3P^o$	0.037	0.027	0.023	0.010	0.021
	$2s^2 2p3d\ ^3D^o$	0.144	0.096	0.112	0.132	0.094
	$2s^2 2p3d\ ^3P^o$	0.037	0.037	0.340	0.069	0.040
	$2s2p^3\ ^3S^o$	0.143	0.156	0.171	0.269	0.152
$2s^2 2p^2\ ^1D$	$2s^2 2p3s\ ^1P^o$	0.118	0.103	0.128	0.103	0.118
	$2s^2 2p3d\ ^1D^o$	0.017	0.012	0.009	0.007	0.013
	$2s^2 2p4s\ ^1P^o$	0.015	0.007	0.004	0.010	0.011
	$2s^2 2p3d\ ^1F^o$	0.123	0.080	0.061	0.099	0.085
	$2s^2 2p3d\ ^1P^o$	0.011	0.011	0.018	0.014	0.009
	$2s2p^3\ ^1D^o$	0.256	0.224	0.344	0.529	
	$2s2p^3\ ^1P^o$	0.151	0.155	0.351	0.333	
$2s^2 2p^2\ ^1S$	$2s^2 2p3s\ ^1P^o$	0.088	0.090	0.021	0.0076	0.094
	$2s^2 2p4s\ ^1P^o$	0.008	0.011	0.007	0.001	0.005
	$2s^2 2p3d\ ^1P^o$	0.170	0.116	0.050	0.142	0.125
	$2s2p^3\ ^1P^o$	0.148	0.124	0.122	0.633	

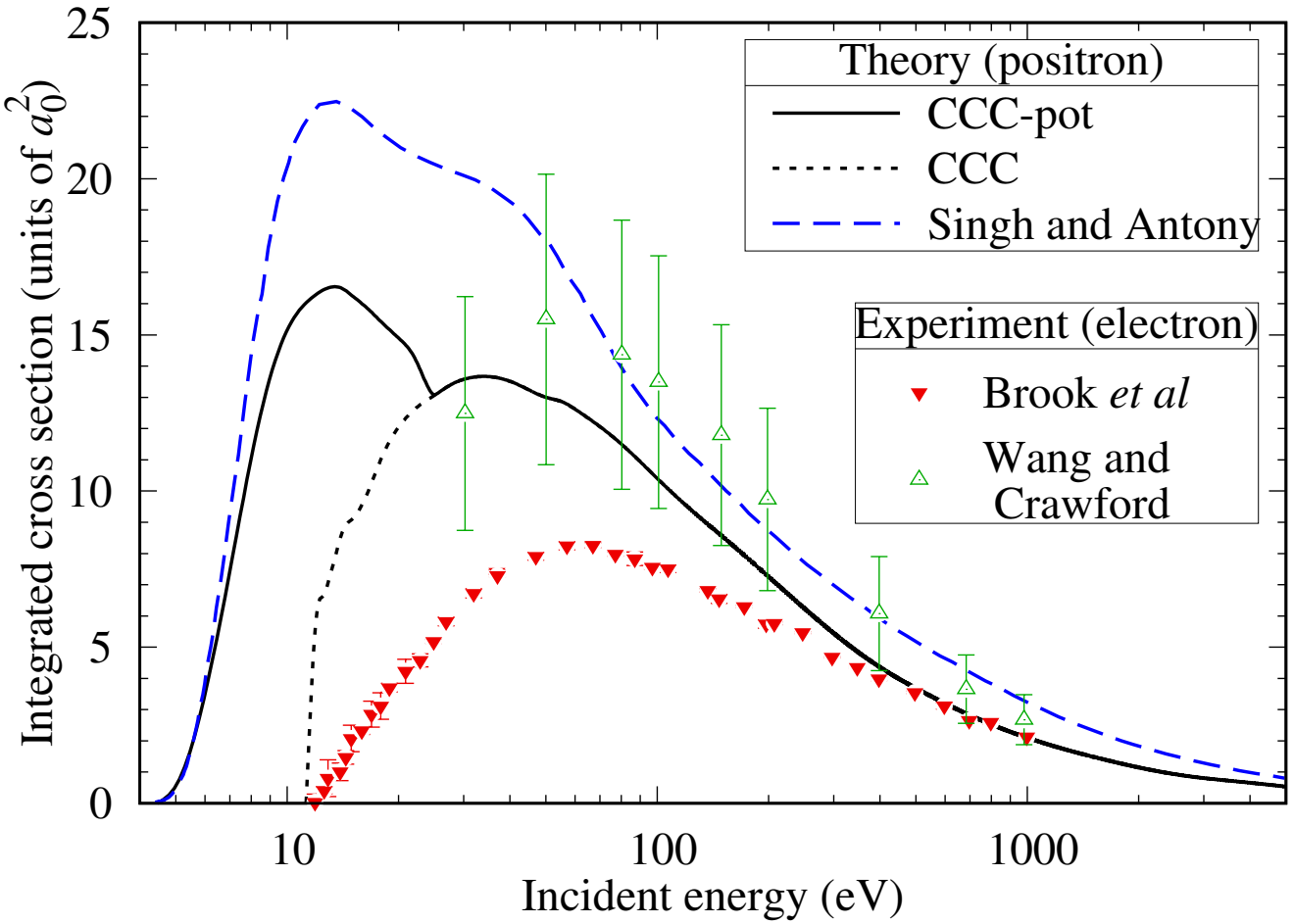
[1] Wang *et al.* *[Phys. Rev. A 87, 012704 (2013)]*
[2] Stancalie *et al.* *[J. Phys. B: At. Mol. Opt. Phys. 576, 01201 0(2015)]*
[3] Dunseath *et al.* *[J. Phys. B: At. Mol. Opt. Phys. 30, 277 (1997)]*
[4] NIST *[https://www.nist.gov/]*

Results: Total Cross Section



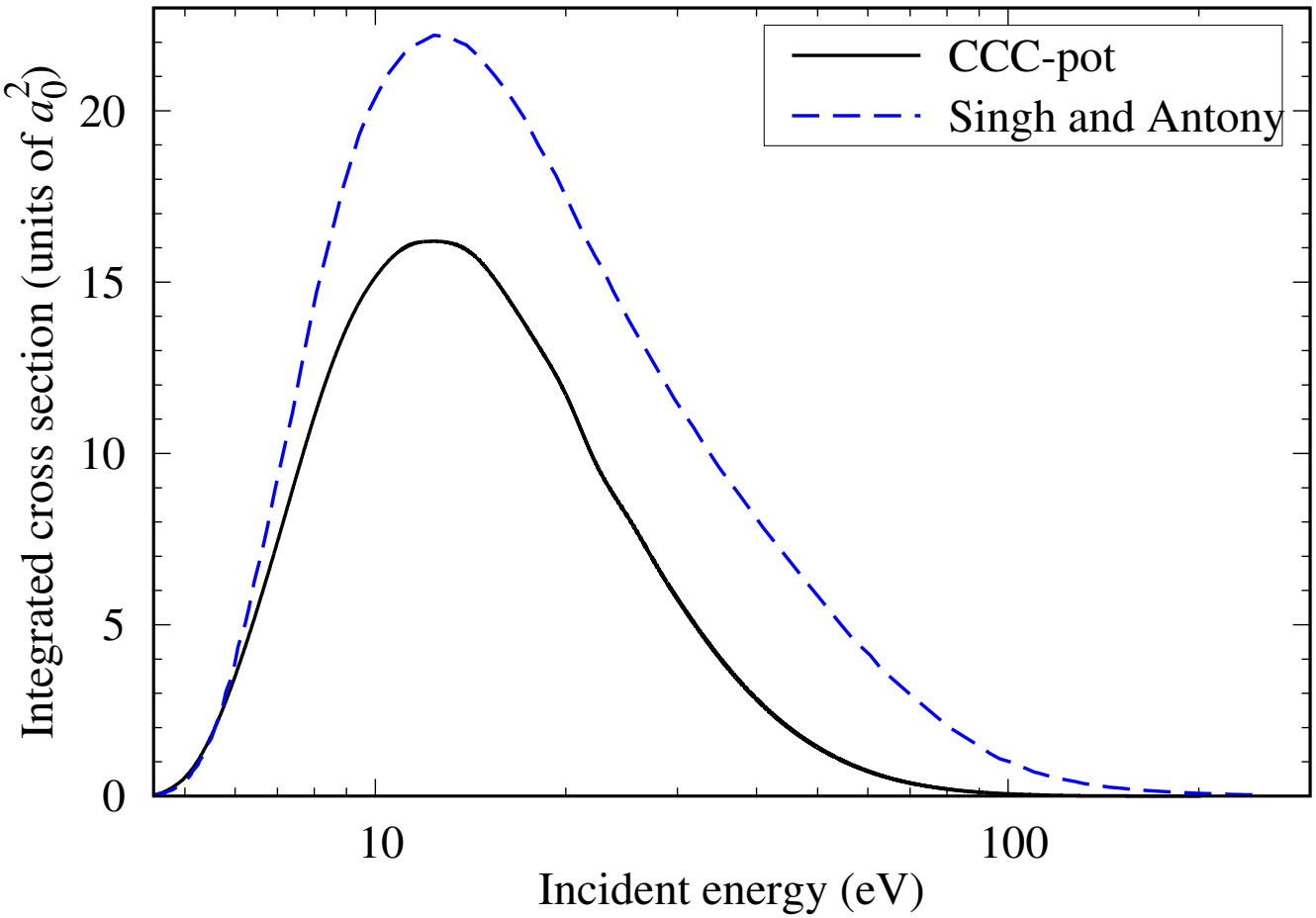
- Agreement with Singh *et al.* *[J. Phys. Chem. A 120, 5685 (2016)]* at high energies, Reid and Wadhera *[J. Phys. B.: At. Mol. Opt. Phys 47, 225211 (2014)]* underestimate both theories.

Results: Total Ionization Cross Section



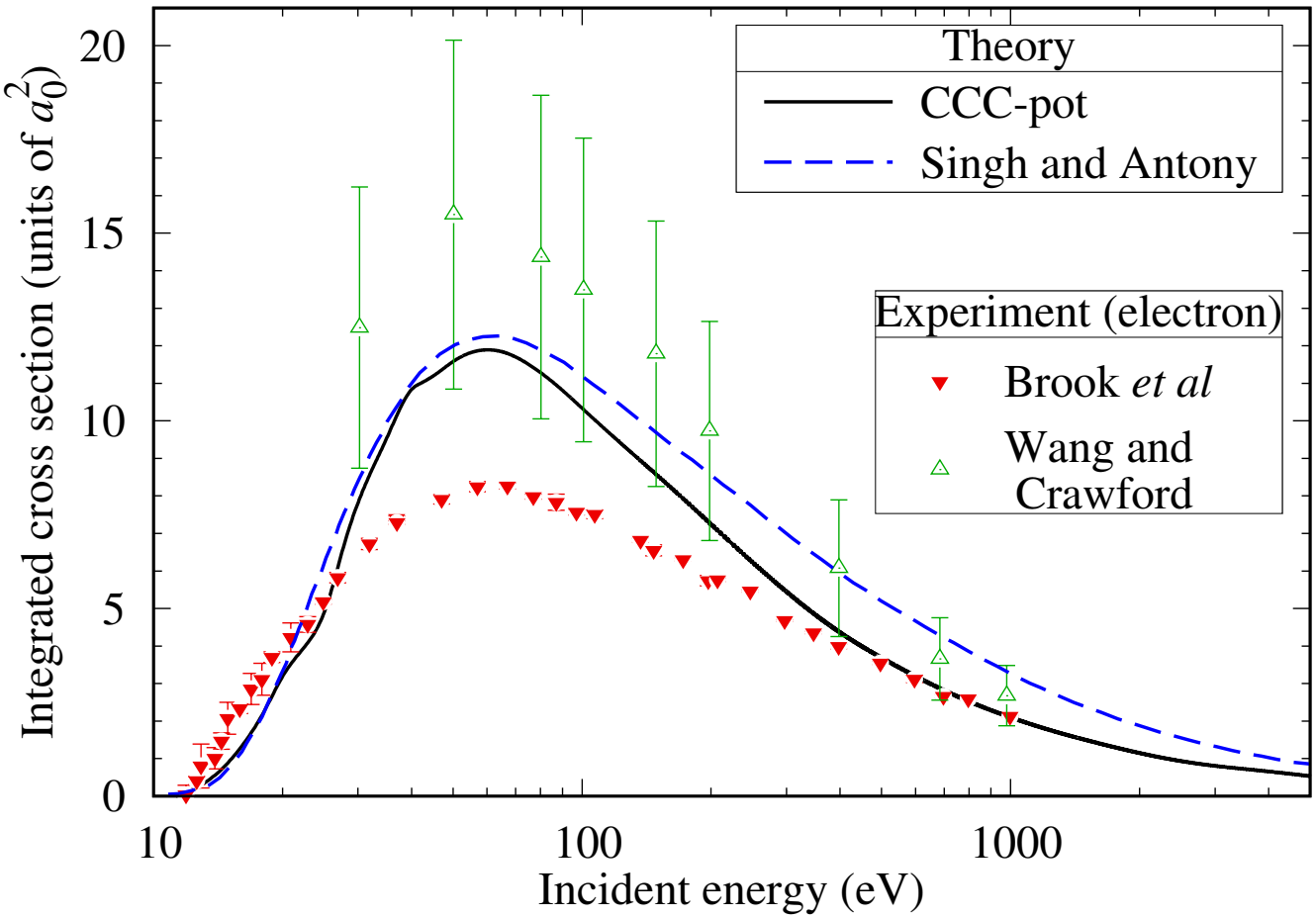
- Results of Singh and Antony *[J. App. Phys. 119, 50006 (2017)]* higher for all energies beside threshold. Agreement viewed with electron experiment of Brook *et al.* *[J. Phys. B.: At. Mol. Opt. Phys. 1, 3115 (1978)]* above 500 eV.

Results: Positronium-formation Cross Section



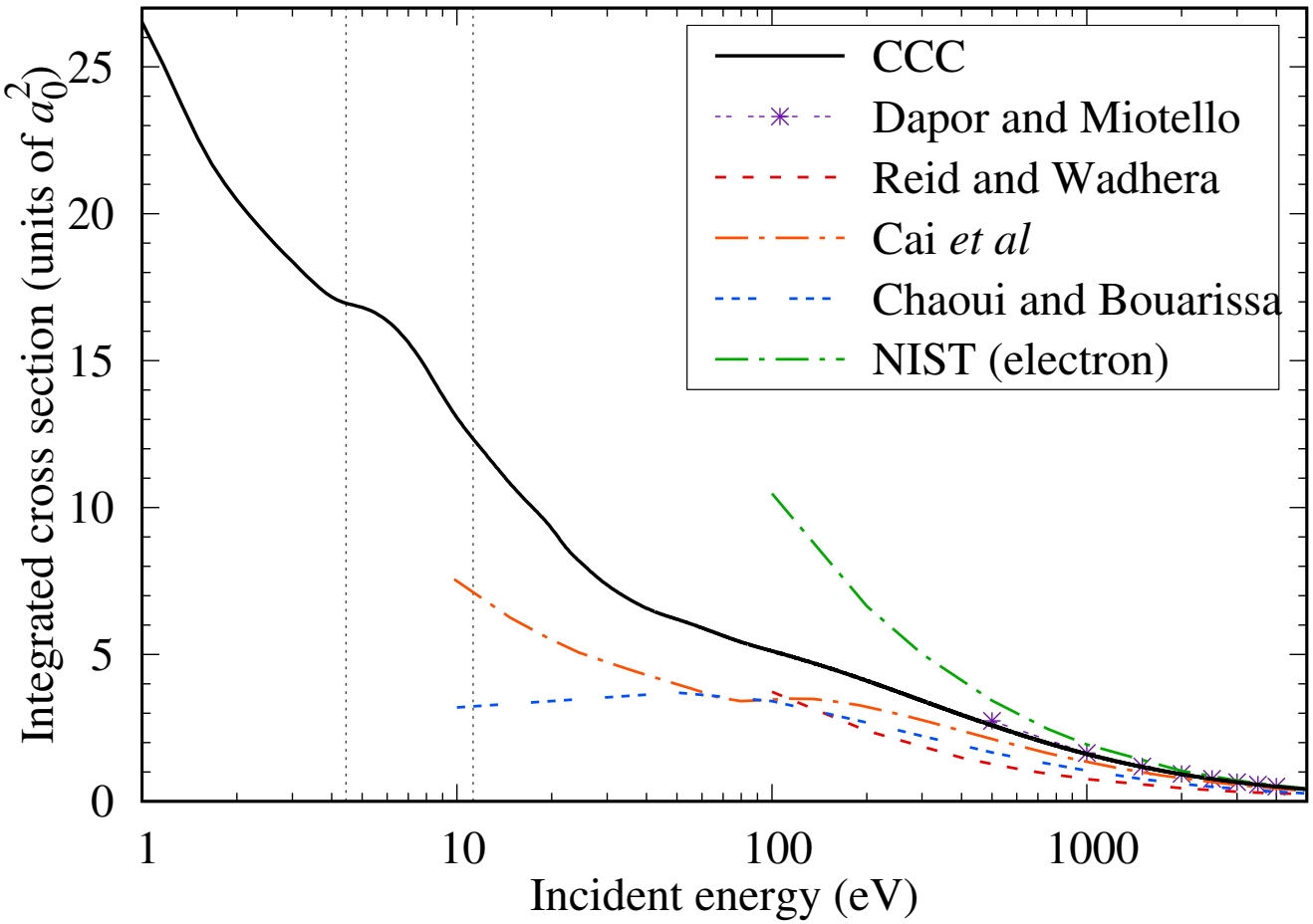
- CCC-scaled model potential lower than Singh and Antony *[J. App. Phys. 119, 50006 (2017)]* for energies above 5 eV.

Results: Direct Ionization Cross Section



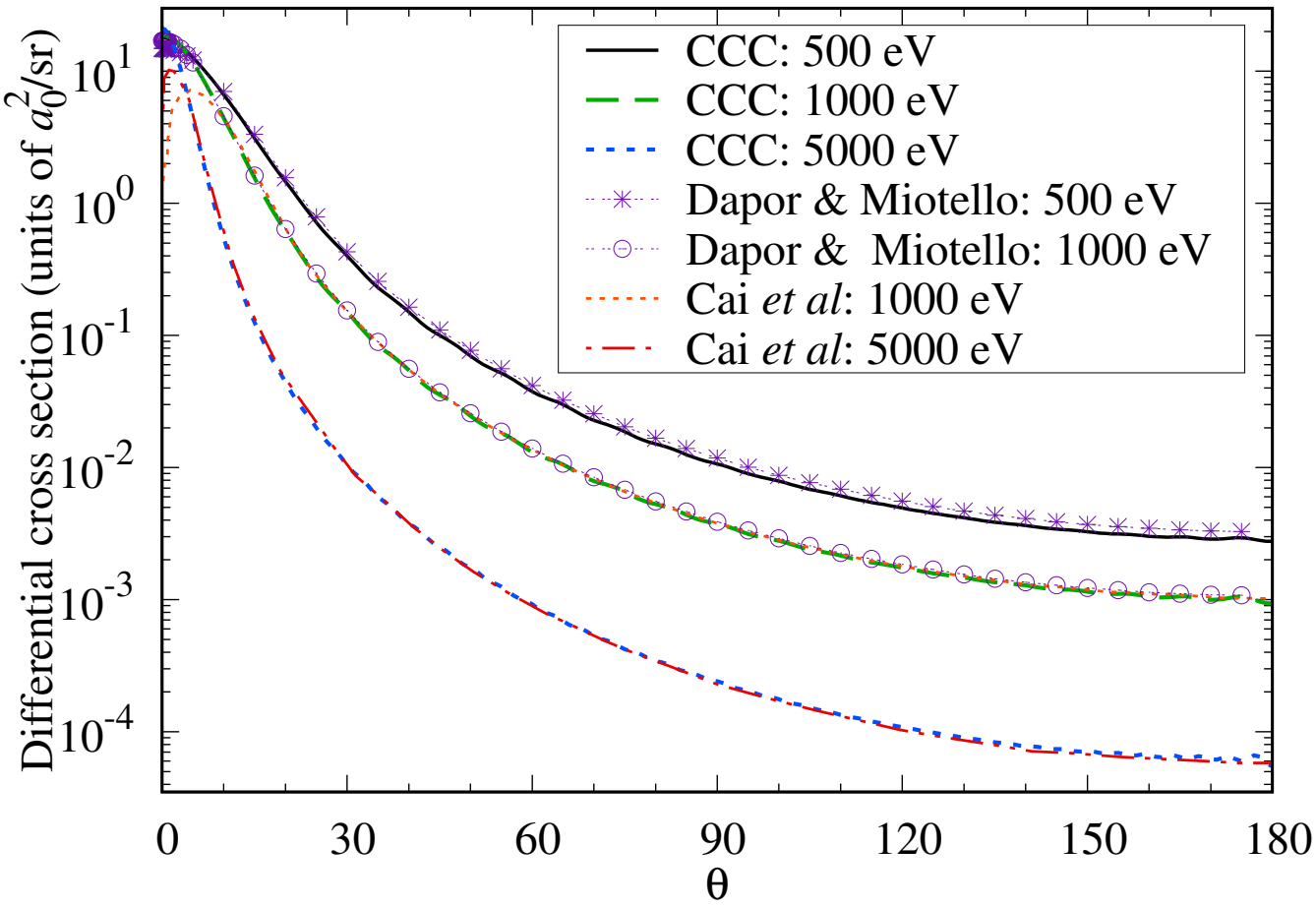
- Singh and Antony *[J. App. Phys. 119, 50006 (2017)]* and CCC in agreement below 40 eV. Agreement between CCC and electron experiment of Brook *et al.* *[J. Phys. B.: At. Mol. Opt. Phys. 1, 3115 (1978)]* above 500 eV.

Results: Elastic Cross Section



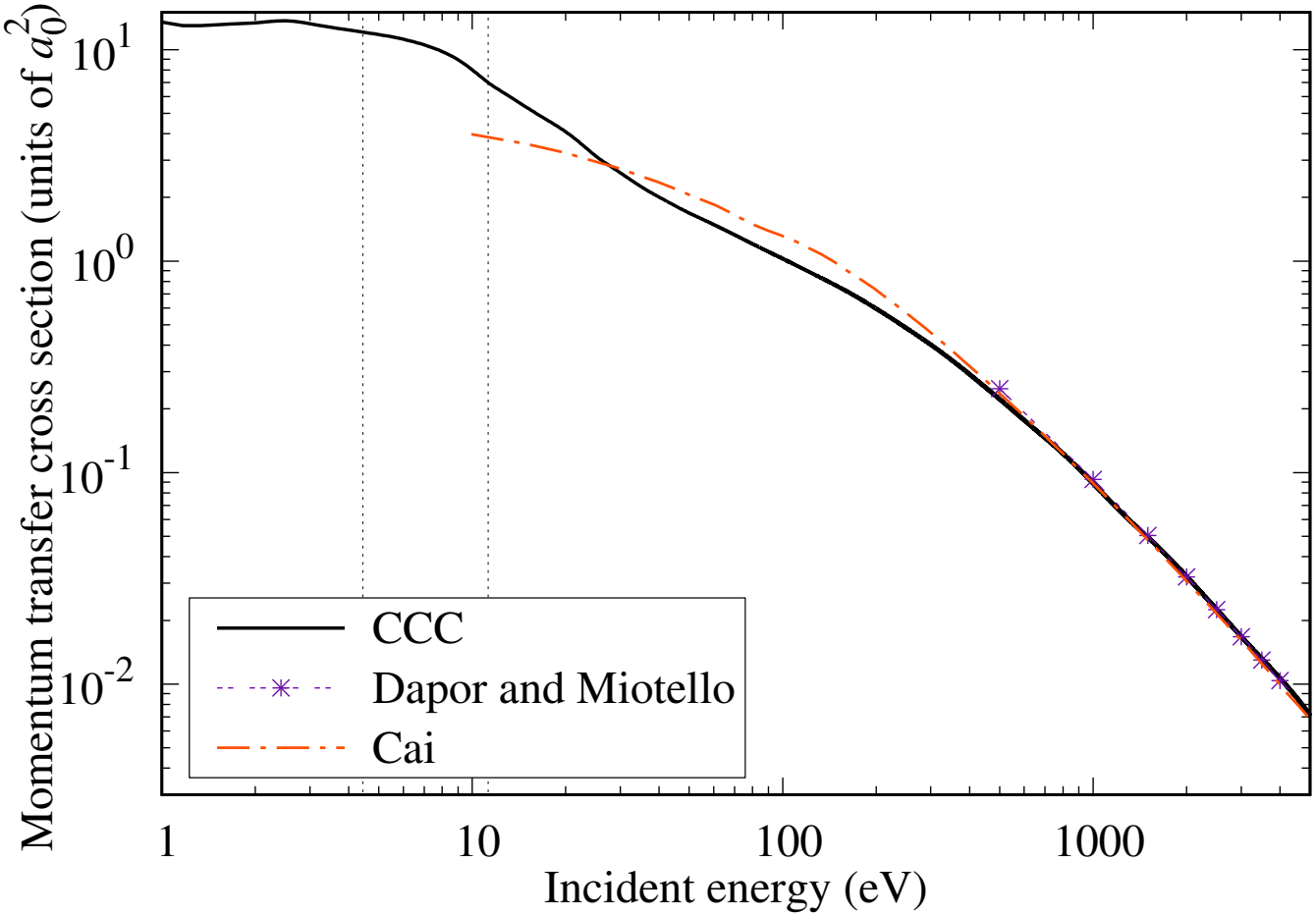
- Close agreement with CCC and calculations of Dapor and Miotello *[At. Data and Nucl. Data Tables 69, 1 (1998)]*.
- Agreement with electron NIST *[https://srdata.nist.gov/srd64/ (2016)]* results above 2500 eV.

Results: Elastic Differential Cross Section



- Close agreement between all theory, except for Cai *et al.* *[J. Phys. Conf. Ser. 262, 012009 (2011).]* at low angles.

Results: Momentum Transfer Cross Section



- Close agreement between all theory above 750 eV.

Results: Low-energy scattering

- From low-energy results can calculate scattering length of -5.03 a_0.
- Can use this to calculate energy of positron-carbon virtual state: 0.537 eV.
- Hidden Ramsauer-Townsend minimum in elastic cross-section, can be observed in *s*-wave component and MTCS.
- Has been demonstrated for noble gases *[Dzuba et al., J. Phys. B: Atom. Mol. Phys. 29, 3151 (1996).)], [Green et al., Phys. Rev. A 90, 032712 (2014).], [F. Arretche, M. V. Barp, W. Tenfen, and E. P. Seidel, Braz. J. Phys 50, 844 (2020).]* .

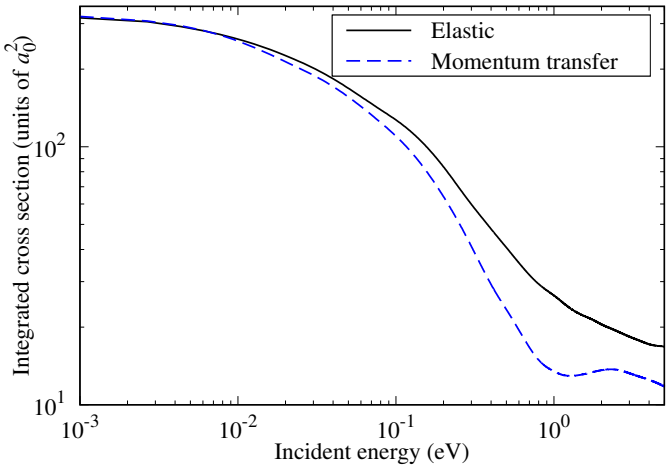


Figure: Low-energy elastic and momentum transfer cross section.

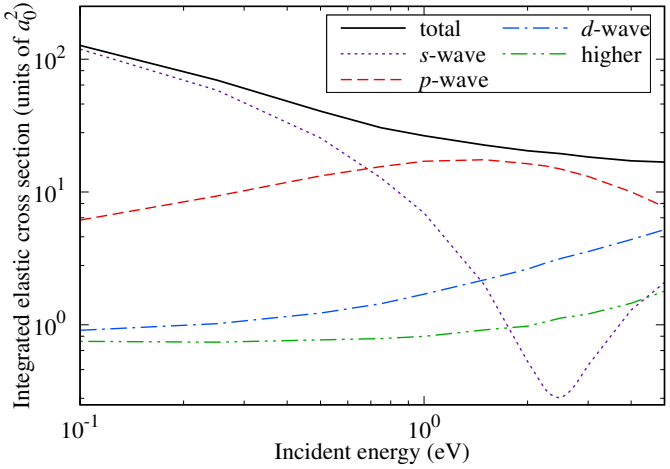
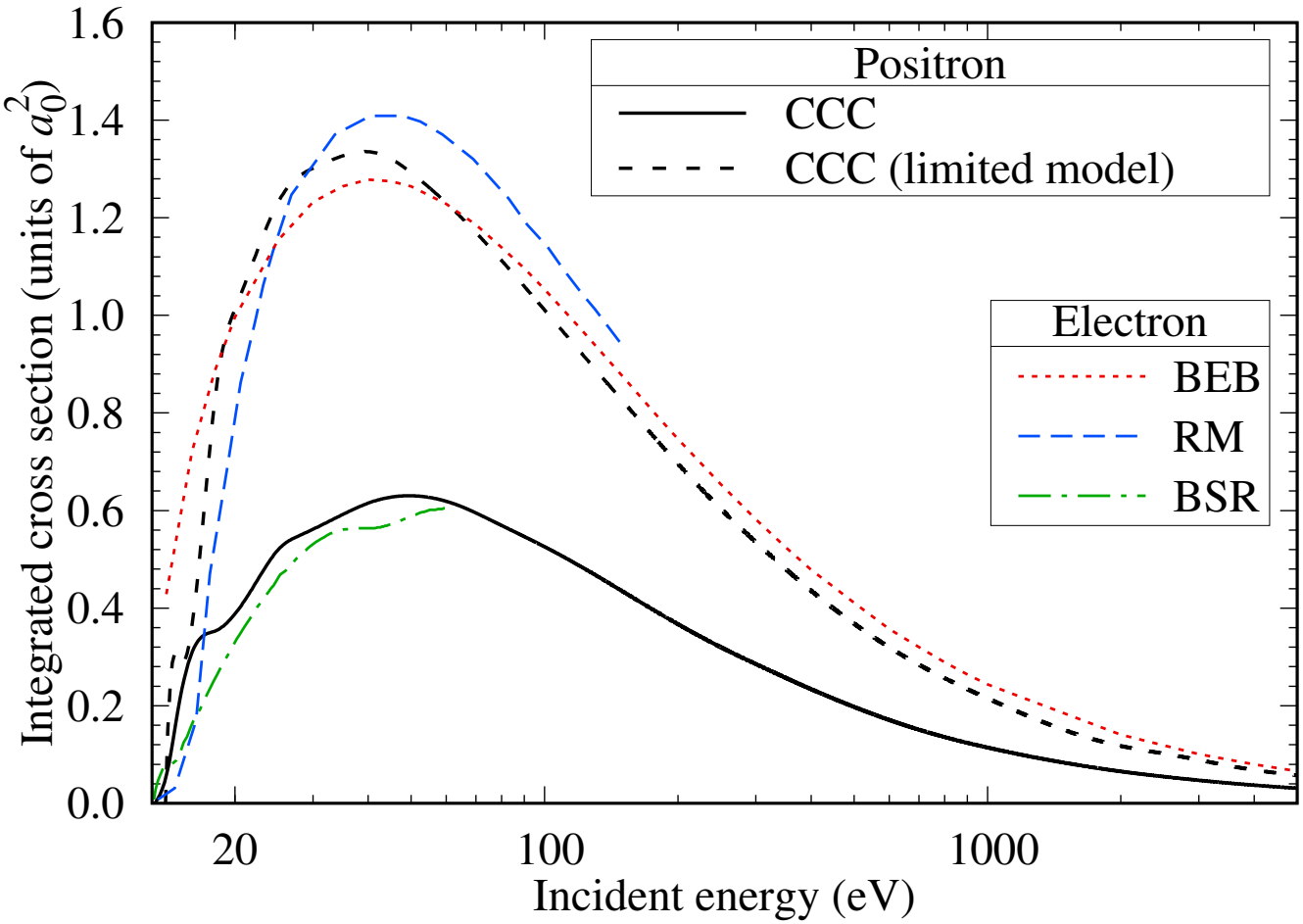


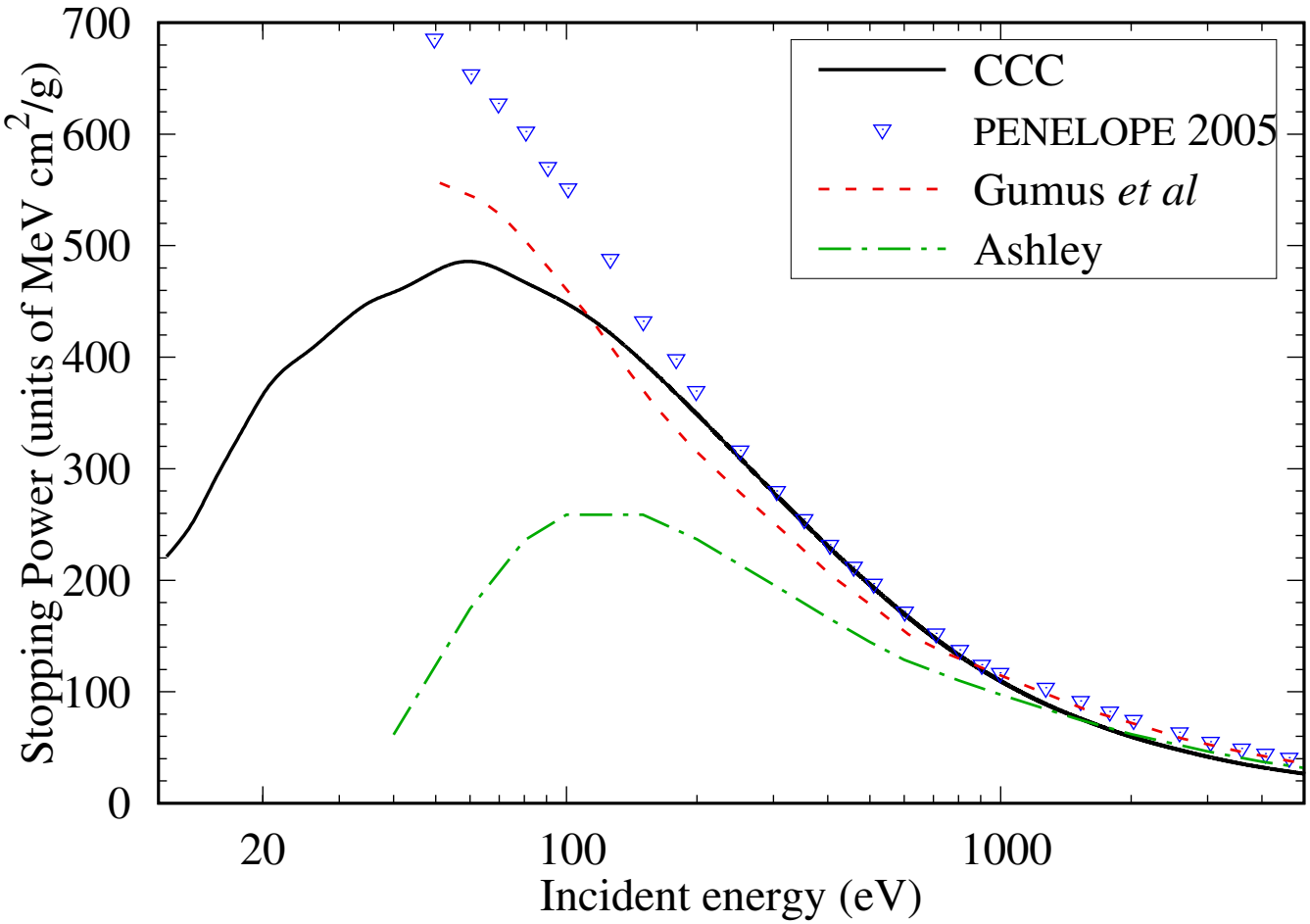
Figure: *s*-, *p*-, *d*- and higher wave components of elastic cross-section.

Results: Excitation to $^3S^o$



- Magnitude in closer agreement with electron-impact BSR *[Phys. Rev. A 87, 012704 (2013)]* results.
- Limited model agreement with other electron-impact calculations suggests inadequate description of $2s2p^2nl$ continuum.

Results: Stopping Power



- PENELOPE *[NEA Databank (2004)]* code in agreement with CCC above 250 eV.
- Ashley *[J. Electron Spectrosc. Relat. Phenom. 50, 323 (1990)]* results in agreement with CCC above 1500 eV.

Conclusion

- Calculated comprehensive cross section data for positron scattering on carbon for various cross sections.
- Also calculated energy of virtual state, scattering length, and stopping power.
- Agreement viewed with carbon experiment and other theory for high energies, with exception for Singh and Antony *[J. App. Phys. 119, 50006 (2017)]* results for ionization.
- Discrepancies present between previous theory at lower energies.

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