CORRELATION EFFECTS IN DIFFERENTIAL ELECTRON-EMISSION SPECTRA OBTAINED FROM DOUBLE IONISATION OF HE BY FAST AU^{53+} IMPACT

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Abstract

We have measured and calculated doubly differential electron-emission cross sections (DDCS's) for well-defined degrees of target ionisation in 3.6 MeV/amu Au^{53+} + He collisions. The calculations are based on the continuum distorted wave with eikonal initial state approximation. We focus on DDCS's obtained in coincidence with double ionisation of the target, and investigate different models for the final two-electron continuum state. Whereas the results of an analysis within the independent particle model overestimate the experimental data badly, we find reduced DDCS's when electron correlation effects are included in a relatively simple fashion. The comparison between theoretical and experimental results indicates that the effective strength of the electron repulsion differs in close and distant collisions.

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1. Introduction

The advent of multi-electron recoil-ion momentum spectrometers (reaction microscopes) has made it possible to map the dynamics of ionising collision events in unprecedented detail and completeness [1,2]. While a considerable part of these data are yet unexplained due to the intricacy of the dynamical many-body Coulomb problem recent theoretical progress in the description of highly-charged ion-atom collisions provided important insights into single and multiple ionisation processes. In particular, it was shown that doubly-differential cross sections (DDCS's) for single-electron emission can be explained in terms of effective single-particle treatments provided that the following requirements are fulfilled: (1) the theoretical description is based on accurate atomic potentials [3]; (2) higher-order contributions to the scattering amplitude are properly taken into account [4]; (3) the results obtained by summation of the contributions from all initially occupied orbitals are compared with experimental data for events that are added according to the degree of ionisation [5,6]. It was found that conditions (1) and two (2) are met when the continuum-distorted-wave with eikonal initial-state (CDW-EIS) approximation [7] is used in conjunction with atomic potentials obtained from the optimised potential method (OPM) [8].

In a recent paper we compared theoretical and experimental DDCS's for 3.6 MeV/amu Au^{53+}-impact on neon and argon atoms in more detail, namely for *given* final charge states q [6]. Such DDCS's are still one-electron spectra, but they are obtained in coincidence with multiple ionisation if $q > 1$. Our calculations were based on a binomial analysis of differential and total CDW-EIS ionisation probabilities. We obtained good agreement with experiment for $q = 1$, and for double ionisation of neon, but observed some discrepancies for double and triple ionisation of argon, whose origin remained somewhat unclear. A possible explanation could be the increasing importance of electron correlation effects at higher multiplicities, which are neglected completely in the calculation.

As the theoretical analysis of this issue is far too complex for true many-electron systems we address the two-electron problem, i.e., collisions from helium targets in the present work. We compare experimental DDCS's for $q = 2$ in 3.6 MeV/amu Au^{53+}-He collisions with results of the binomial analysis, and with results of an improved

model, in which final-state correlation effects are included to some extent. Atomic units ($\hbar = m_e = e = 1$) are used throughout.

2. Theory

A widely used model for the final state of two emitted interacting electrons is the so-called '3C' or BBK wave function [9]. This wave function is the (symmetrised) product of two Coulomb waves, which describe the interactions of both electrons with the target nucleus, and a factor $F_{\rm BBK}$ that accounts for the interaction between the electrons and ensures that the correct asymptotic boundary conditions of the two-electron-one-nucleus system are satisfied. With the momentum difference $k_{12} = |\mathbf{k}_1 - \mathbf{k}_2|$, its inverse $\xi_{12} = 1/k_{12}$, and the distance $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$ between the two electrons $F_{\rm BBK}$ can be written as

$$F_{\rm BBK} = F_{\rm Gam}(k_{12})\ {}_1F_1(i\xi_{12}, 1, -i(k_{12}r_{12} + \mathbf{k}_{12}\cdot\mathbf{r}_{12})), \tag{1}$$

where ${}_1F_1$ is a confluent hypergeometric function, and $F_{\rm Gam}$ is the so-called Gamov (or Sommerfeld) factor

$$F_{\rm Gam}(k_{12}) = \exp(-\pi\xi_{12}/2)\Gamma(1 - i\xi_{12}), \tag{2}$$

which reflects the fact that both electrons cannot be emitted with zero relative momentum due to their Coulomb repulsion.

The authors of Ref. [10] simplified the BBK wave function by replacing the exact value of ${}_1F_1$ at each point $\mathbf{r}_{12}$ by its value at a single point obtained by averaging r_{12} over the ground state of the helium atom. They reported the result

$$F_\alpha(k_{12}) = F_{\rm Gam}(k_{12})\ {}_1F_1(i\xi_{12}, 1, -ik_{12}\alpha) \tag{3}$$

with $\alpha = 0.769$ a.u. In a second model they considered $\alpha = 0$, i.e., they took the confluent hypergeometric function at zero spatial distance $r_{12} = 0$, and attached the Gamov factor only to a two-electron product wave function.

In order to exploit these ideas in the present context we write for the transition amplitude to a two-electron continuum state Ξ_f^-

$$A_{if}(\mathbf{b}) = \langle \Xi_f^- | \Psi(t \to \infty) \rangle. \tag{4}$$

We assume a final state of the form

$$\Xi_f^-(\mathbf{r}_1,\mathbf{r}_2) = \chi_{f_1}^-(\mathbf{r}_1)\chi_{f_2}^-(\mathbf{r}_2)F_\alpha(k_{12}) \tag{5}$$

and a simple product of the solutions of the single-particle equation

$$i\partial_t\psi_{1s}(\mathbf{r},t) = \left(-\frac{1}{2}\Delta + v_{\rm atom}(r) - \frac{Q_P}{|\mathbf{r}-\mathbf{R}|}\right)\psi_{1s}(\mathbf{r},t) \tag{6}$$

as dynamical state vector $\Psi(t\to\infty)$. In Eq. (6), $v_{\rm atom}$ denotes the OPM target potential and Q_P and $\mathbf{R}$ are the charge and the coordinate of the projectile. Our choice for $\Psi(t\to\infty)$ implies that we neglect initial-state and dynamic correlation effects. The transition amplitude (4) can now be written as

$$\begin{aligned} A_{if}(\mathbf{b}) &= \langle\chi_{f1}^-|\psi_{1s}(t\to\infty)\rangle\langle\chi_{f2}^-|\psi_{1s}(t\to\infty)\rangle F_\alpha(k_{12}) \\ &= a_{if_1}(\mathbf{b})a_{if_2}(\mathbf{b})F_\alpha(k_{12}). \end{aligned} \tag{7}$$

We calculate the single-particle transition amplitudes a_{if_i} in the CDW-EIS approximation. With the definition

$$\frac{d^2p_i(b)}{d\varepsilon_{\rm el}d\Omega_{\rm el}} = \frac{1}{2\pi}\int_0^{2\pi} d\varphi_b\,|a_{if}(\boldsymbol{b})|^2, \tag{8}$$

for the differential single-particle ionisation probabilities we obtain for the probability to find the electrons at emission energies $\varepsilon_{\rm el},\varepsilon_{\rm el}^{'}$ and emission angles $\Omega_{\rm el},\Omega_{\rm el}^{'}$

$$\frac{d^4P(b)}{d\varepsilon_{\rm el}d\Omega_{\rm el}d\varepsilon_{\rm el}^{'}d\Omega_{\rm el}^{'}} = |F_\alpha(k_{12})|^2\,\frac{d^2p_{1s}(b)}{d\varepsilon_{\rm el}d\Omega_{\rm el}}\,\frac{d^2p_{1s}(b)}{d\varepsilon_{\rm el}^{'}d\Omega_{\rm el}^{'}}. \tag{9}$$

The DDCS for double ionisation, i.e., the cross section to detect one electron at emission energy $\varepsilon_{\rm el}$ and emission angle $\Omega_{\rm el}$ in coincidence with the recoil-ion charge state $q=2$ is obtained by integrating Eq. (9) over the coordinates of one electron and over the impact parameter b

$$\frac{d^2\sigma_2^\alpha}{d\varepsilon_{\rm el}d\Omega_{\rm el}} = 2\pi\int_0^\infty bdb\frac{d^2p_{1s}(b)}{d\varepsilon_{\rm el}d\Omega_{\rm el}}\int d\varepsilon_{\rm el}^{'}\int d\Omega_{\rm el}^{'}\,\frac{d^2p_{1s}(b)}{d\varepsilon_{\rm el}^{'}d\Omega_{\rm el}^{'}}\,|F_\alpha(k_{12})|^2. \tag{10}$$

We now distinguish the following models.

(1) Final-state correlation is neglected and the result of the binomial analysis of Ref. [6] is obtained if we set $F_\alpha(k_{12})=1$ in Eq. (10). This is the independent particle model (IPM), in which both electrons can be emitted with very similar or even the same momentum $\mathbf{k}$.

(2) If we use the Gamov factor in Eq. (10) ($\alpha = 0$) and note that

$$C_{\rm CDS}(k_{12}) \equiv |F_{\rm Gam}(k_{12})|^2 = \frac{2\pi}{k_{12}[\exp(2\pi/k_{12}) - 1]}, \tag{11}$$

we reproduce a model that was proposed in Ref. [11] for the calculation of *total* double ionisation cross sections. The so-called *coulomb density of states* (CDS) factor has the limiting values

$$\lim_{k_{12}\to 0} C_{\rm CDS}(k_{12}) = 0 \tag{12}$$

$$\lim_{k_{12}\to \infty} C_{\rm CDS}(k_{12}) = 1, \tag{13}$$

i.e., electrons with very different momenta are allowed to move independently, whereas the probability to find both electrons in the same region of momentum space is suppressed exponentially. As the CDS factor is always smaller than one, the corresponding DDCS in Eq. (10) is smaller than its IPM counterpart of model (1) for all relative electron momenta.

(3) For $\alpha > 0$ the correlation factor $|F_\alpha(k_{12})|^2$ has the same limiting values as the CDS factor [Eqs. (12) and (13)], but can become larger than unity at intermediate relative momenta. This is demonstrated in Fig. 1, where we show correlation factors obtained with different values of α. The selection of α-values will be explained in Section 3 along with our cross section results. For all α-values relative momenta $k_{12} < 1$ a.u. are suppressed strongly. The CDS factor ($\alpha = 0$) approaches unity very slowly for large relative momenta. It can be expected that the suppression of double ionisation due to the electron repulsion will be overemphasised in this model, since there is no physical reason to reduce the emission of two electrons with a momentum difference as large as 10–20 a.u. By contrast, the correlation factor approaches unity much more rapidly for $\alpha > 0$. We note, however, that the oscillatory behavior encountered at intermediate relative momenta is counterintuitive and may be regarded as a hint that these final-state correlation models are rather crude.

3. Results and discussion

We have used the models described in Section 2 and compare the corresponding results for the DDCS at $q = 2$ in 3.6 MeV/amu Au^{53+} + He collisions with experimental

data in Fig. 2. The measurements have been put on an absolute scale by using the measured ratio $R_2 = \sigma_2/\sigma_1$ [12] and normalising the DDCS for $q = 1$ to the present CDW-EIS calculations. Details about the operating principle of the reaction microscope and the resolution obtained in this experiment are provided in Ref. [13].

The results of the IPM-based analysis badly overestimate the experimental DDCS for all electron velocities. Inclusion of final-state correlation effects via the correlation factor (3) reduces the DDCS since double ionisation with small momentum differences is suppressed by an amount that depends on the choice of the parameter α in Eq. (3). We have used five different values for α.

(1) The CDS model ($\alpha = 0$) leads to a DDCS, which is significantly smaller than the experimental result. This overestimation of final-state correlation effects was expected from Fig. 1, in which the CDS factor was shown to approach the limit $C_{\rm CDS}(k_{12}) \to 1$ only for very large momentum differences k_{12}.

(2) We followed the prescription of Ref. [10] and replaced the exact value of the confluent hypergeometric function in the BBK final state at each point $\mathbf{r}_{12}$ by its value at a single point obtained by averaging r_{12} over the ground state of the helium atom. We define [cf. Eqs. (1) and (3)]

$$\alpha \equiv \langle r_{12}(1 + \cos(\mathbf{r}_{12}, \mathbf{k}_{12}))\rangle = \langle r_{12}\rangle \tag{14}$$

and obtain

$$\alpha = \langle r_{12}\rangle = \langle \Psi_{gs}^{\rm OPM} | r_{12} | \Psi_{gs}^{\rm OPM}\rangle = 1.362 \text{ a.u.} \tag{15}$$

for the OPM ground-state wave funcion $\Psi_{gs}^{\rm OPM}$ (which is in close agreement with the value for the exact ground state $\langle r_{12}\rangle = 1.422$ a.u. given in Ref. [14]). Why this result differs significantly from the value $\alpha = 0.769$ a.u. of Ref. [10] is unclear to us. Using our value $\alpha = 1.362$ a.u. we find a DDCS for $q = 2$ which is in reasonable agreement with the experimental data for $v_\perp = 0.05$ a.u., but which lies above experiment for the higher transverse velocity cuts.

(3) If we choose $\alpha < 1.362$ a.u. the DDCS's decrease gradually. In particular, we find good agreement with experiment at $v_\perp = 0.45$ a.u. when we use $\alpha = 0.9$ a.u. and at $v_\perp = 0.95$ a.u. with $\alpha = 0.5$ a.u. The value $\alpha = 0.769$ a.u. of Ref. [10] yields a DDCS which lies in between the other ones.

We offer the following remarks as a possible explanation for this behavior. Electron emission at small transverse velocities corresponds mainly to distant collisions, in which the momentum transfer is small. In this region the projectile does not perturb the target atom strongly in the first half of the collision, and it is reasonable to assume that the two electrons have approximately the average distance of the ground state at the time when they are ejected. As a consequence, the calculations with $\alpha = 1.362$ a.u. are in good agreement with experiment at $v_\perp = 0.05$ a.u. For closer collisions the projectile does perturb the atomic ground state while approaching the target. The perturbation leads to a smaller average distance between the electrons, which are both attracted by the high projectile charge. These collisions correspond mainly to higher electron emission velocities, i.e., to higher transverse velocity cuts. In these situations smaller values of α than the one that corresponds to the ground state are required.

It would be interesting to see whether a calculation with the full BBK wave function would lead to good agreement with experiment for all electron velocities. Such a calculation, however, is much more involved as the two-electron transition amplitude (4) does not factorise into two one-electron amplitudes in this case. Future work should be concerned with the question whether α can be determined by some general criteria before using it in the cross section calculations.

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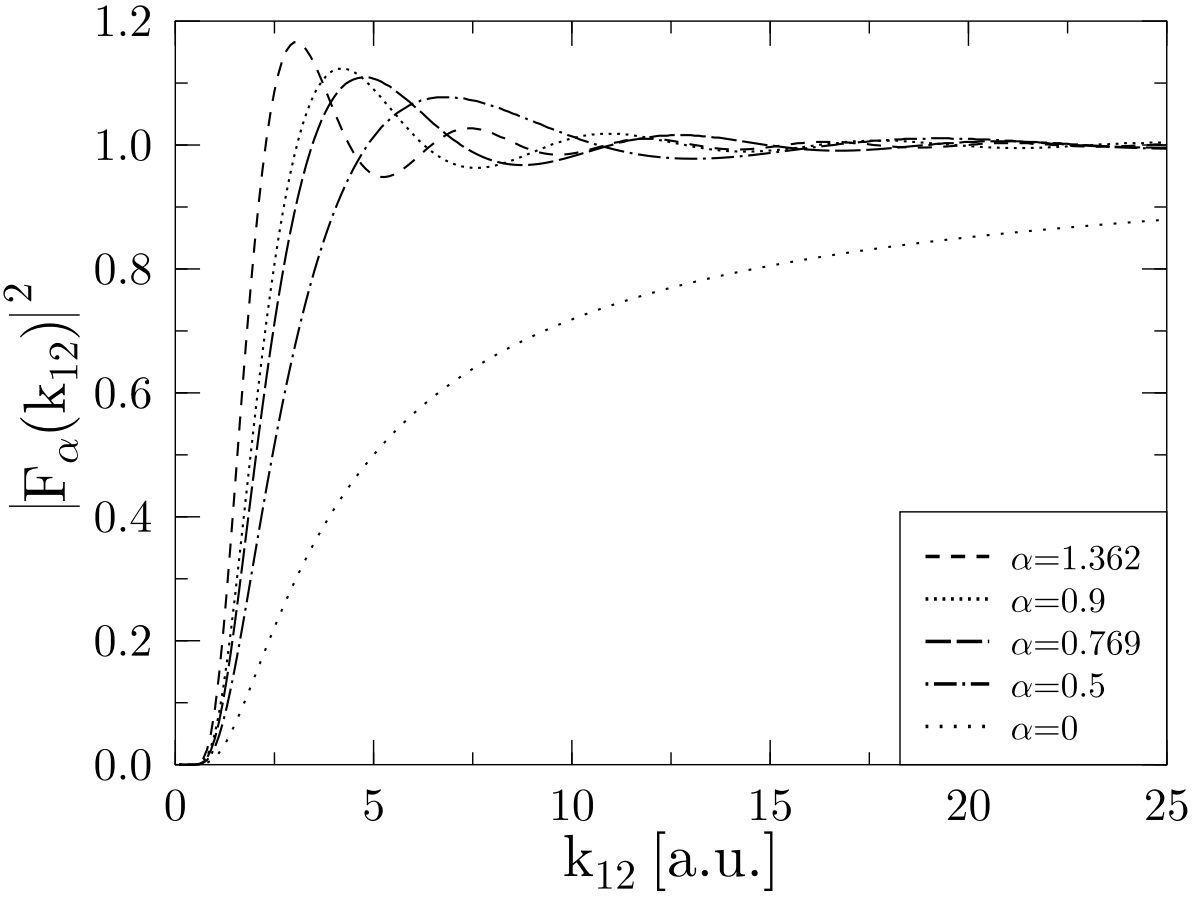
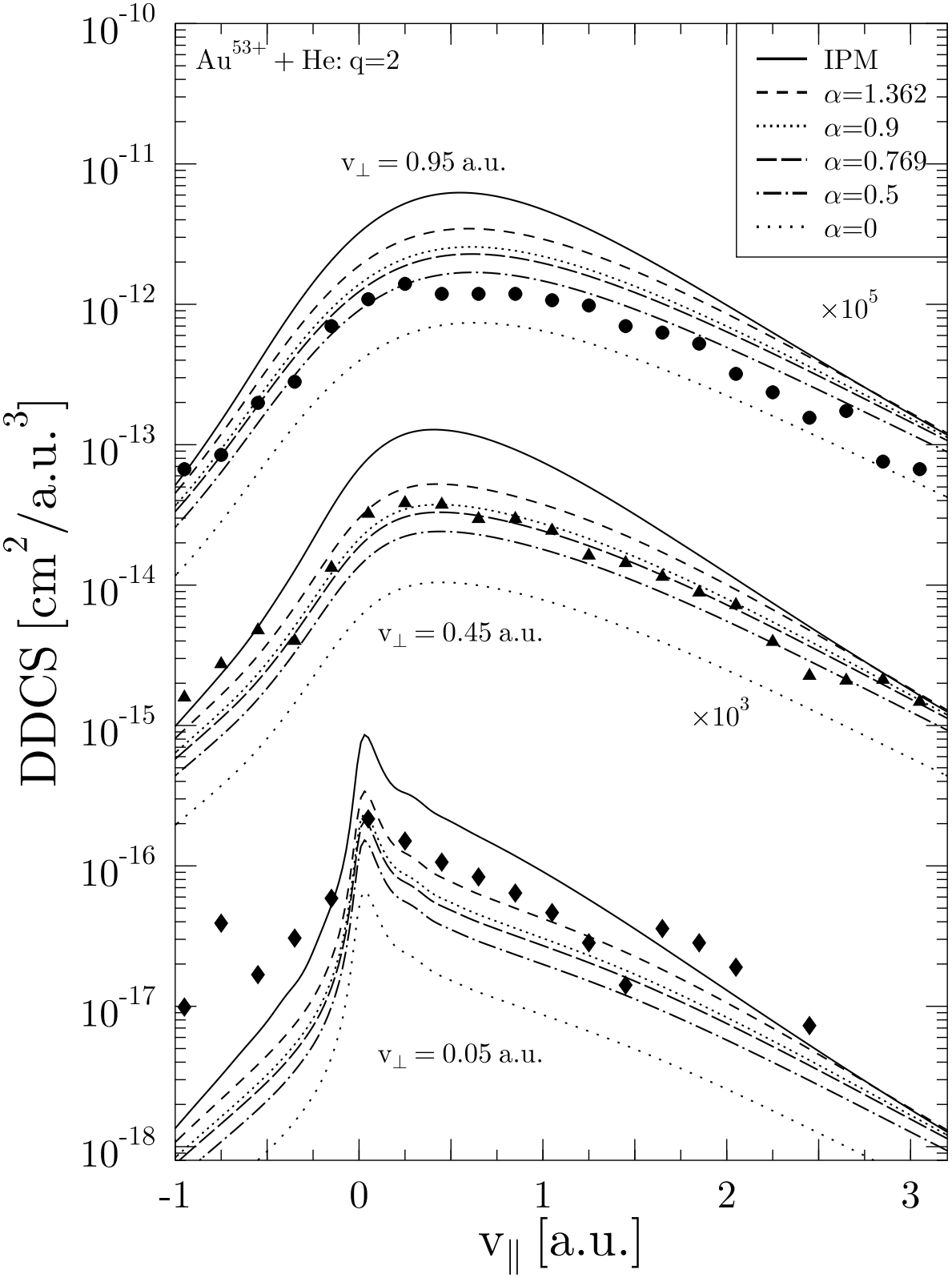
Figures

Fig. 1. Correlation factor $|F_\alpha|^2$ [Eq. (3)] as a function of the momentum difference k_{12} of the electrons.

Fig. 2. DDCS for double ($q = 2$) electron emission in 3.6 MeV/amu Au^{53+}+ He collisions. The DDCS at $v_\perp = 0.45$ and 0.95 a.u. are multiplied by the indicated factors. The calculations refer to the IPM-based analysis and the inclusion of final-state correlation using different values of α in the correlation factor (3). Symbols: present experimental data normalised as described in the text.


1.2
1.0
0.8
0.6
0.4
0.2
0.0
$|F_\alpha(k_{12})|^2$
0
5
10
15
20
25
k_{12} [a.u.]
α=1.362
α=0.9
α=0.769
α=0.5
α=0


Au^{53+} + He: q=2
IPM
α=1.362
α=0.9
α=0.769
α=0.5
α=0
$v_\perp = 0.95$ a.u.
$\times 10^5$
$v_\perp = 0.45$ a.u.
$\times 10^3$
$v_\perp = 0.05$ a.u.
DDCS [cm^2/a.u.^{3}]
$v_\parallel$ [a.u.]
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
10^{-15}
10^{-16}
10^{-17}
10^{-18}
-1
0
1
2
3