Angular Differential Cross-Section for Ionization of Helium in C^{6+} Ion Collision *

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With the help of the density operator, the angular differential cross-section for ionization of helium is calculated within the framework of the one-centre atomic-orbital close-coupling method. We consider a naked C^{6+} ion as projectile with an energy of 2.5 MeV/a.u. Our result agrees well with the experimental data and the other theoretical calculations such as the first Born approximation, various Distorted Wave models and the classical trajectory Monte Carlo simulation.

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Ionization of atoms in collisions with fast ions is a fundamental physical process attracting great experimental and theoretical interest. Single-ionization for very fast fully stripped ions colliding with light atoms is well understood both theoretically [1] and experimentally.[2] For moderate projectile velocities where the processes of excitation, ionization and electron capture compete and interfere strongly, the perturbation models fail or have limited validity.

For slow collisions, where the velocity of the projectile is equal to or smaller than the velocity of the target electron, charge transfer becomes relevant and overwhelms ionization.[3] When the impact parameter is of the magnitude of the target atomic radius, molecular orbitals can be formed. To describe charge transfer mechanisms for slow collisions, one needs two-centre calculations.

Electron–electron correlation may play an important role at low impact energies. The essential role of the wavefunction in describing the ejection of electron has been shown in Ref. [4]. It is necessary therefore to exceed the limitations of the independent-electron model and consider electron–electron correlation.

The experimental setup for measurements has recently gained such high quality and accuracy that the individual momenta of the two participating electrons and of the recoil ion have become measurable,[5] challenging all the models to go beyond total cross-sections and calculate more sensitive observables. On the level of total-cross-sections the models are much too complicated to test and to compare with the others due to the lack of further information. Angular differential cross-sections as measurable quantities makes it possible to analyse the differences and the similarities of different models.

In this work we study the angular distribution of electrons ejected from helium atom under the impact of C^{6+} ion and compare it with the experimental data [6] and various theories. To our knowledge, there has been no angular differential cross-section calculation for atoms in heavy ion projectile impacts using the one-centre atomic-orbital close-coupling (AOCC) method until now. A review article about the semi-classical close-coupling description of atomic collisions can be found in Ref. [7]. In the recent work of Ref. [8], the two- and one-centre AOCC method was employed to calculate the ionization cross-sections.

In this Letter, we extend our one-centre AOCC method to calculate the angular differential cross-section for ionization. Details of our original method can be found in Refs. [9] or [10]. As basis set we use configuration interaction (CI) wavefunctions built up from Slater-like orbitals to describe bound states of helium and regular Coulomb wavepackets to have a finite approximation about the single and double electron continuum. Our method was successfully used to calculate single- and double-ionization total cross-sections of helium in heavy ion collisions,[9,10] and later for photoionization of helium with short intensive XUV laser pulses.[11]

The motion of the projectile is described by a straight-line trajectory with constant speed. For projectile–electron interaction the non-relativistic time-dependent Coulomb potential is used. With the help of the density operator we calculate the angular differential ionization probabilities and a final integration over the impact parameter gives us the angular differential cross-sections which is the new aspect in this work. Atomic units are used throughout the paper unless otherwise mentioned.

For the ionization process we solve the time-dependent Schrödinger equation with time-dependent

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external Coulomb field

$$i\frac{\partial}{\partial t}\,\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)=(\hat{H}_{\rm He}+\hat{V}(t))\,\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t), \qquad (1)$$

where $\hat{H}_{He}$ is the Hamiltonian of the unperturbed helium atom

$$\hat{H}_{\rm He}=\frac{p_1^2}{2}+\frac{p_2^2}{2}-\frac{2}{r_1}-\frac{2}{r_2}+\frac{1}{|\boldsymbol{r}_1-\boldsymbol{r}_2|} \qquad (2)$$

and $\hat{V}(t)$ is the projectile–electron interaction

$$\hat{V}(t)=-Z_p\Big(\frac{1}{R_1(t)}+\frac{1}{R_2(t)}\Big) \qquad (3)$$

with $R_i(t)=((x_i-b)^2+y_i^2+(z_i-v_pt)^2)^{1/2}$, $i=1,2$. To solve (1) we expand $\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2)$ in the basis of eigenfunctions $\{\Phi_j\}$ of the time-independent Schrödinger equation

$$\hat{H}_{\rm He}\,\Phi_j(\boldsymbol{r}_1,\boldsymbol{r}_2)=E_j\,\Phi_j(\boldsymbol{r}_1,\boldsymbol{r}_2) \qquad (4)$$

to yield

$$\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)=\sum_{j=1}^{N}a_j(t)\,\Phi_j(\boldsymbol{r}_1,\boldsymbol{r}_2)e^{-iE_jt}, \qquad (5)$$

where $a_j(t)$ are the time-dependent expansion coefficients for the various channels described by the wavefunctions Φ_j. Inserting this ansatz into (1) leads to a system of first-order differential equations for the expansion coefficients

$$\frac{da_k(t)}{dt}=-i\sum_{j=1}^{N}V_{kj}e^{i(E_k-E_j)t}a_j(t),\quad (k=1,\ldots,N), \qquad (6)$$

where V_{kj} is the coupling matrix $\langle\Phi_k(\boldsymbol{r}_1,\boldsymbol{r}_2)|\hat{V}|\Phi_j(\boldsymbol{r}_1,\boldsymbol{r}_2)\rangle$ including the symmetrized products of the projectile–electron single-particle interaction matrix elements with $\hat{V}(t)$ and electron–electron single-particle overlap matrix elements, respectively.

Denoting the ground state by $k=1$, we use the following initial conditions for solving (6):

$$a_k(t\to-\infty)=\begin{cases}1 & k=1\\ 0 & k\neq 1.\end{cases} \qquad (7)$$

The total cross-section for occupying the helium eigenstate k can be calculated as

$$\sigma_k=2\pi\int_0^\infty bP_k(b,t\to\infty)db \qquad (8)$$

with the probability

$$P_k(b,t\to\infty)=|a_k(t\to\infty)|^2. \qquad (9)$$

The coupled system of (6) has to be solved numerically.

The eigenfunctions Φ_j in (4) are obtained by diagonalizing the Hamiltonian in a basis of orthogonal symmetrized two-particle functions f_μ so that

$$\Phi_j(\boldsymbol{r}_1,\boldsymbol{r}_2)=\sum_\mu b_\mu^{[j]}f_\mu(\boldsymbol{r}_1,\boldsymbol{r}_2)\,. \qquad (10)$$

For the single-particle wavefunctions we use an angular momentum representation with spherical harmonics $Y_{l,m}$, hydrogen-like radial Slater functions and radial regular Coulomb wavepackets. The Slater function reads as

$$S_{n,l,m,\kappa}(\boldsymbol{r})=c(n,\kappa)r^{n-1}e^{-\kappa r}Y_{l,m}(\theta,\varphi), \qquad (11)$$

where $c(n,\kappa)$ is the normalization constant. A regular Coulomb wavepacket

$$C_{k,l,m,Z}(\boldsymbol{r})=q(k,\Delta k)Y_{l,m}(\theta,\varphi)\cdot\int_{E_k-\Delta E_k/2}^{E_k+\Delta E_k/2}F_{k,l,Z}(r)\,dk \qquad (12)$$

with normalization constant $q(k,\Delta k)$ is constructed from the radial Coulomb function

$$F_{k,l,Z}(r)=\sqrt{\frac{2k}{\pi}}e^{\frac{\pi\eta}{2}}\frac{(2\rho)^l}{(2l+1)!}e^{-i\rho}|\Gamma(l+1-i\eta)|\;{}_1F_1(1+l+i\eta,2l+2,2i\rho), \qquad (13)$$

where $\eta=Z/k$, $\rho=kr$.

The wavepackets cover a small energy interval ΔE_k and thereby form a discrete representation of the continuum which can be incorporated into our finite basis set. The normalized Coulomb wavepackets are calculated up to 315 a.u. radial distance or more to achieve a deviation of less then one percent from unity in their norm.

In our approach two different effective charges Z have been used to take into account the difference between the singly- and the doubly-ionized electrons. For singly-ionized states we have used $Z=1.0$, and $Z=2.0$ for the doubly ionized case. A slight deviation from the effective charge gives practically no change in the final spectrum. We cover the single- and double-continuum up to 6 a.u. energy equidistantly.

Out of the single particle states (11, 12) we have used 17 s-functions (9 Slater functions (sf), 4 wavepackets (wp) with $Z=1.0$ and 4 wp with $Z=2.0$), 18 p-functions (6 sf, 6 wp with $Z=1.0$ and 6 wp with $Z=2.0$) and 12 d-functions (4 sf, 4 wp with $Z=1.0$ and 4 wp $Z=2.0$) to construct the symmetrized basis functions $f_\mu^{LM}(\boldsymbol{r}_1,\boldsymbol{r}_2)$. For the $L=0$ configurations we have used ss wavefunctions to obtain a ground state energy of -2.88 a.u. For the $L=1,2$ states we have used sp or sd configurations. The effects of the CI wavefunction can be clearly seen at the values of the bound states. Enhancing the number of the wavefunctions all the bound energy

levels become lower converging to the measured energy values. For $L = 0$ configurations one can use $ss + pp + dd$ angular correlated wavefunctions [11] to obtain a better ground state energy of -2.901 a.u., which is reasonably accurate compared to the 'exact' value of -2.903 a.u.

To test the convergence of our basis we have used 520 basis states first, up to 27 a.u. energy. Our results clearly demonstrate that the channels above 4 a.u contribute very little to the ionization probabilities. The results we present have been calculated with the help of 300 quantum states.

Between the first ionization threshold (-2.0 a.u.) and the lowest auto-ionizing bound state (-0.6931 a.u. for $L = 1$) our basis contains 22 states providing the major contribution for single ionization.

In order to classify the states such as bound, single-ionized or double-ionized states we use a Feshbach projection method described in Refs. [9, 10].

It is well known that with the help of the density operator radial electron density can be calculated in a stationary atom.[12] We use the same idea here, and calculate the azimuthal electron density of the ionized atom with the time-dependent wavefunction (5) after the collision ($t \to \infty$). The density operator of the helium atom reads as

$$\hat{\rho}(\boldsymbol{r}) = \delta(\boldsymbol{r} - \boldsymbol{r}_1) + \delta(\boldsymbol{r} - \boldsymbol{r}_2). \tag{14}$$

Using the mean value of the density operator one can calculate the spatial probability distribution for the emitted electron

$$\begin{aligned}\rho(\boldsymbol{r}) &= \langle \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t)|\rho(\boldsymbol{r})|\, \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t)\rangle \\ &= 2 \int |\, \Psi(\boldsymbol{r}, \boldsymbol{r}_1, t)|^2 d^3 r_1, \end{aligned} \tag{15}$$

where the factor two comes from the symmetry considerations.

Integrating now over the impact parameter and the remaining azimuthal angle we obtain the angular differential cross-section for single-ionized electrons

$$\begin{aligned} P(\theta) =& \frac{1}{2\pi} \int_0^\infty \int_0^{2\pi} \langle \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t)|\hat{\rho}(\boldsymbol{r})| \\ & \cdot \Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t)\rangle r^2 dr d\varphi \\ =& \frac{1}{\pi} \int_0^\infty \int_0^{2\pi} \int_{\boldsymbol{r}_1} |\, \Psi(\boldsymbol{r}_1, \boldsymbol{r}, t)|^2 d^3 r_1 r^2 dr d\varphi \\ =& \frac{1}{\pi} \sum_{j=1}^{N} a_j^*(t) e^{iE_j t} \sum_{k=1}^{N} a_k(t) e^{-iE_k t} \\ & \cdot \int_0^\infty \int_0^{2\pi} \int_{\boldsymbol{r}_1} \Phi_j^\dagger(\boldsymbol{r}_1, \boldsymbol{r}) \Phi_k(\boldsymbol{r}_1, \boldsymbol{r}) r^2 dr d\varphi d^3 r_1, \end{aligned} \tag{16}$$

where $\Phi_j^\dagger(\boldsymbol{r}_1, \boldsymbol{r})$ and $\Phi_k(\boldsymbol{r}_1, \boldsymbol{r})$ are the configuration interaction wavefunctions (10). The angular dependence of $P(\theta)$ is given by the products of spherical harmonics $Y_{l,m}(\theta, \varphi)$ integrated over φ yielding products of associated Legendre polynomials with different angular momentum. A more detailed description of the method can be found in Ref. [13].

For single- and double-ionization many different mechanisms are possible. In slow ion–atom collisions the particles have sufficient time to form a quasi-molecule for a short time. The electrons are in the region between the projectile and target nucleus. When the projectile captures the target electron then it is called the saddle point ionization. At moderate energies the target electron is simply 'kicked out' by the projectile. For double-ionization many different mechanisms can occur. At low impact energies with large perturbation, the ionization is purely sequential, and the electrons are emitted independently one after another. The projectile has enough time to interact with both electrons, this is called the two-step 2 mechanism. In the range of small perturbation when the projectile is quick, two independent projectile–electron interactions become improbable because of the short reaction time. Therefore one can describe the double-ionization by the shake-off mechanism. After a single-ionization event the remaining electron can also be emitted due to the rearrangement of the wavefunction to the new situation of an unscreened target nucleus. In our calculation these two mechanisms interact and cannot be separated.

We have calculated the total cross-sections and compared them with the experimental data.[6] For the single-ionization cross-section we obtain 28.8×10^{-17} cm^2; the experimental value is 31.7×10^{-17} cm^2, which implies a ten-percent discrepancy between theory and experiment. In the case of double-ionization, five percent of the single-ionization total cross-section is considered experimentally, which is 1.5×10^{-17} cm^2. Our calculation gives 1.22×10^{-17} cm^2.

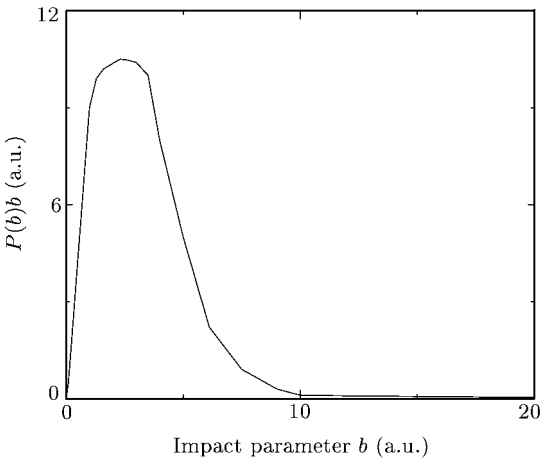

Fig. 1. Impact-parameter dependence of a single-ionized channel.

Figure 1 shows the impact-parameter dependence of a typical single-ionized channel with total angular momentum $L = 1$ and energy of 2.1 a.u. To achieve convergence 14 different impact parameters are calculated up to 50 a.u.

Figure 2 displays our angular differential cross-section results together with the experimental data [6]

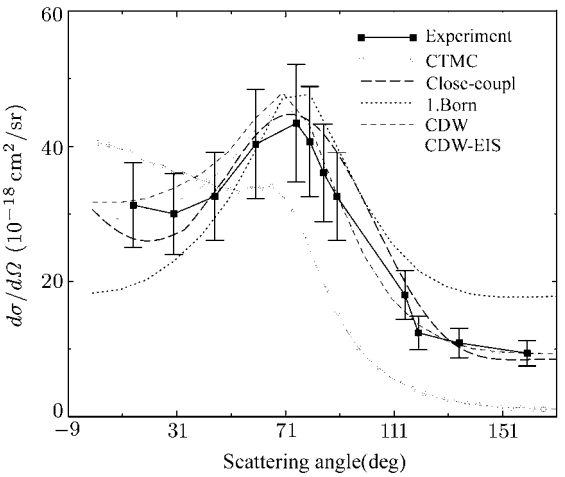

Fig. 2. Angular differential cross-section for ejected electrons emitted in 2.5 MeV/amu C^{6+} helium collisions. Solid squares: experiment.[6] Open connected circles: CTMC.[16] Thick dashed line: Our one-centre AOCC calculation. Dotted line: first Born approximation. Thin dashed line: CDW.[14] Thin dot-dashed line: CDW-EIS.[15].

and various other theories such as the continuum distorted wave (CDW),[14] continuum distorted wave Eikonal initial state (CDW-EIS),[15] first Born approximation and classical trajectory Monte Carlo (CTMC) method.[16] Our calculation is in good agreement with the experiment. At a scattering angle of about 70°, all the quantum mechanical calculations have a maximum. The CDW and CDW-EIS models explain this phenomenon with the binary encounter approach (BEA). Unfortunately, we have not been able to calculate the energy differential cross-sections as yet. Further work is in progress to calculate more sensitive observables from our *ab initio* one-centre AOCC method.

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References

[1] Inokuti M 1978 *Rev. Mod. Phys.* **50** 23
[2] Haugen H K et al 1982 *Phys. Rev.* A **26** 1950
[3] Toshima N 1994 *Phys. Rev.* A **50** 3940
[4] Byron F W et al 1966 *Phys. Rev. Lett.* **16** 1139
[5] Ullrich J et al 1997 *J. Phys. B: At. Mol. Opt. Phys.* **30** 2917
[6] Lokesh C Tribedi et al 1988 *Phys. Rev.* A **58** 3619
[7] Fritsch W et al 1991 *Phys. Rep.* **202** 1
[8] Toshima W 2001 *Phys. Rev.* A **64** 024701
[9] Barna I F 2002 *PhD Dissertation* (University Giessen) http://geb.uni-giessen.de/geb/volltexte/2003/1036
[10] Barna I F et al 2003 *Eur. Phys. J.* D **25** 239
[11] Barna I F et al 2003 *Eur. Phys. J.* D **27** 287
[12] Pfeiffer C et al 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 53
[13] Gagyi-Pállfy A C 2002 *Master Thesis* (University Giessen)
[14] Ɓelkic Dz 1978 *J. Phys. B: At. Mol. Opt. Phys.* **11** 3529
[15] Crothers D S F et al 1983 *J. Phys. B: At. Mol. Opt. Phys.* **16** 3229
[16] Tökési K et al 1994 *Nucl. Instrum. Methods* B **86** 201