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Electron impact ionization of helium: a comprehensive experiment benchmarks theory

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Abstract: Single ionization of helium by 70.6 eV electron impact is studied in a comprehensive experiment covering a major part of the entire collision kinematics and the full 4π solid angle for the emitted electron. The absolutely normalized triple differential experimental cross sections are compared with results from the convergent close-coupling (CCC) and the time dependent close coupling (TDCC) theories. Whereas excellent agreement with the TDCC prediction is only found for equal energy sharing, the CCC calculations are in excellent agreement with essentially all experimentally observed dynamical features, including the absolute magnitude of the cross sections.

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1. Introduction

Collisions of electrons with atoms and molecules that drive chemical and physical reactions are of utmost importance for a broad range of areas from plasma physics to radiation damage in living tissue or the chemistry in planetary atmospheres. Moreover, they are of basic intrinsic interest since they constitute one of the most fundamental realizations of the ubiquitous correlated quantum dynamical few-body problem. Kinematically complete experiments for electron-impact single ionization of atoms, so-called (e, 2e) studies, determine the momentum vectors of all continuum particles for known final-state configuration of the remaining ion thus ultimately benchmarking all facets of quantum few-body Coulomb dynamics.

Electron-impact induced single ionization is a break-up reaction with three free charged particles in the final channel interacting via long range Coulomb forces posing enormous challenges to theory. The complete description of this problem requires covering all possible sets of the kinematical parameters for any initial- and final-state electron energies E_0 , E_1 , E_2 , as well as momenta \vec{k}_0 , \vec{k}_1 , \vec{k}_2 thus calling for approaches that go beyond perturbation theory. So far only the most fundamental three-particle quantum dynamical problem, electron-impact ionization of atomic hydrogen, was claimed having been rigorously solved numerically by ab-initio methods. These are (i) the exterior complex scaling (ECS) [1], (ii) the time-dependent close coupling (TDCC) [2] and (iii) the convergent close coupling (CCC) [3] theories. Just going to helium however, with only one additional bound electron, the full correlated four-particle problem cannot be solved without approximations such that it represents an ideal target to study few-body quantum dynamics beyond three particles. Therefore it has been extensively studied experimentally for impact energies from close to threshold up to 8 keV, see e.g. [4–10]. Theoretically the structure of the helium atom is dominated by one-electron excitations, which allows for a quite accurate description. The non-perturbative CCC and TDCC methods have been successfully applied to describe electron-helium ionizing collisions yielding in general excellent agreement with all available experimental data, see e.g. [5, 11–15]. Thus, recently the question was posed by Bray et al. [13] “*whether the problem of single ionization of helium by electron impact, leaving the ion in the ground state, has also been computationally solved*”. However, puzzling systematic discrepancies observed for some of the coplanar data question this statement calling for comprehensive measurements to benchmark theory. To this end, experimental data are required that are normalized to an absolute scale for any final-state continuum covering a broad range of collision kinematics, coplanar as well as out of plane, and a large part of the 4π solid angle of electron emission.

In this work we comprehensively explore single ionization of helium by 70.6 eV electron impact. Absolute fully differential cross sections are reported covering a major part of the three-particle final-state via three-dimensional (3D) (e, 2e) cross sections ranging from equal to unequal energy

sharing between both outgoing electrons, different sets of projectile scattering angles as well as “in plane” and “out-of-plane” geometries.

2. Experiment

The experiments were performed using an advanced reaction microscope [16] which is especially designed for electron collision experiments. The setup was described before [8, 17], and only a brief outline will be given here. A well focused (1 mm diameter), pulsed electron beam (pulse width ≈ 1.5 ns), produced by a standard thermo-cathode gun, intersects a cold helium gas jet (2 mm diameter) created by supersonic expansion. Using parallel electric and magnetic fields, both the final state electrons and the recoiling ion are projected onto two-dimensional position- and time-sensitive detectors in opposite directions. From the positions of the hits and the times-of-flight, the initial momentum vectors of the detected fragments can be determined. A large part of the entire 4π solid angle is covered for the final state particles, 100 % for the detection of the recoil-ion and about 80 % for electrons. Electrons miss the detector for energies transversal to the spectrometer axis higher than 15 eV and for particular times-of-flight where they arrive close to the spectrometer axis in a bore hole in the electron detector, which is required for the passage of the projectile beam. Experiments for unequal energy sharing ($E_2 \leq 15$ eV) were obtained through triple coincidence detection between two outgoing electrons (e_1 and e_2) and the recoiling ion. The momentum vectors \vec{k}_1 and \vec{k}_2 are measured directly, the recoil-ion momentum (\vec{k}_r) is used as a reference signal for calibration and efficiently subtracting the background as discussed in [17, 18]. For the equal energy sharing case ($E_1 = E_2 = 23$ eV), experimental data were obtained from double coincidence events between one of the outgoing electrons (\vec{k}_1) and the recoil ion (\vec{k}_r). The momentum (\vec{k}_2) of the second electron is then calculated applying momentum conservation as discussed in [5]. The absolute scale of the cross section was obtained by normalizing to the absolute measurements in coplanar geometry by Röder et al. as reported in [19]. All data in the present experiment were recorded simultaneously in a single run. Consequently, once the normalization factor has been fixed for one point, the cross sections for all the other geometries are automatically normalized with respect to each other for all recorded scattering angles and all ejected electron energies.

3. Results and discussions

The details of theoretical approaches to differential ionization have already been extensively discussed in [14, 15] for CCC and in [11, 12] for TDCC. The calculations presented here have the same foundation as described by Bray et al. [13] for CCC and Colgan et al. [12] for TDCC, suitably modified for the present kinematics and geometries. Triple differential cross sections (TDCS) as 3D emission patterns for experiment and CCC calculation are presented in Figure 1 (a) and (b), respectively, for the scattering angle of $\theta_1 = -20^\circ$ of the fast final state electron as function of the emission direction of a slow ejected electron with $E_2 = 5$ eV energy. The projectile is coming in from the bottom (\vec{k}_0) and scattered to the left (\vec{k}_1). [We count positive angles from the projectile forward direction clockwise.] These two vectors define the scattering plane as indicated by the dashed frame in Figure 1(a). The TDCS for a particular direction is given by the distance from the origin of the plot (also corresponding to the collision point) to the point on the surface, which is intersected by the ionized electron's emission direction. The kinematics chosen displays the principal features of the cross section pattern. It is governed by the well-known binary and recoil lobes which are aligned roughly along and opposite the indicated momentum transfer vector \vec{q} , respectively. Both lobes are bent backwards since the emitted electron is repelled by the scattered projectile due to the long-range nature of the Coulomb force. The strength of this post-collision interaction (PCI) effect increases with decreasing relative energy of the two outgoing electrons, and it clearly becomes relevant at the present low impact energy. Another striking feature in the 3D patterns is the significant cross section outside of the scattering plane filling up the angular range in between the binary and recoil lobes. This was first observed for ion-impact single-ionization of helium [20], where it is still not fully understood. Figure 1 exhibits excellent agreement of the experimental and the CCC results. The qualitative features observed in the experiment are very well reproduced by this theoretical method.

For quantitative comparisons of experiment and the CCC and TDCC theories, Figure 2 (a) and (b) present the TDCS in the scattering plane (the dashed frame in Figure 1) and the so-called perpendicular plane which is perpendicular to the scattering plane and includes the incoming

projectile beam (the dotted frame in Figure 1), respectively. Also presented in Figure 2 are the TDCS in the scattering and perpendicular planes for different scattering angles from $\theta_1 = -10^\circ$ to $\theta_1 = -30^\circ$ and the same ejection energy of $E_2 = 5$ eV. The TDCS in the scattering plane (left column) clearly show the basic features of binary (peaked at $\theta_2 \sim 75^\circ$) and recoil lobes (peaked at $\theta_2 \sim 210^\circ$). In the perpendicular plane (right column), three peaks are observed, one strong peak at $\theta_2 = 180^\circ$ (peak I), and two side peaks at $\theta_2 \sim 60^\circ$ and 300° (peaks II). The insets in Figure 2 (c) to (j) are the 3D patterns of TDCS for experiment (left column) and CCC theory (right column). Excellent agreement between experimental data and CCC calculations are found for both in and out of the scattering plane and the 3D patterns. In contrast, discrepancies are observed in comparison with the TDCC theory, where the magnitude of the cross section at the recoil region is significantly higher than in the experiment. From lower ($\theta_1 = -10^\circ$) to higher ($\theta_1 = -30^\circ$) scattering angles the absolute magnitude of the TDCS decreases strongly and the binary to recoil ratio in the scattering plane increases. In the perpendicular plane the peak II to peak I ratio also grows. As we can see from the 3D pattern in Figure 1 (a), the perpendicular plane cuts through the recoil lobe of the 3D pattern in the backward direction. Therefore, the origin of peak I is the well-known nuclear backscattering process leading to the recoil lobe [6]. The peaks II, however, are attributed to the significant contribution of higher order processes filling up the region in between the binary and recoil lobes as discussed in [18, 21]. All the features observed in the experiment are fully reproduced by the CCC theory, and show excellent accuracy in the absolute magnitude.

Further quantitative comparisons between experiment and the theories are shown in Figure 3, where the cross sections for $\theta_1 = -10^\circ$ and -30° , and $E_2 = 3$ eV and 10 eV are presented in the scattering plane (left column) and perpendicular plane (right column). Also included in (a) to (h) are the 3D TDCS patterns for experiment (left column) and CCC theory (right column). The basic features of the TDCS in the scattering, perpendicular planes and 3D images are quite similar to that discussed above. Agreement between experiment and CCC prediction is again excellent, confirming that the CCC calculation is sufficiently accurate to reproduce all the experimental observations. Again the TDCC calculations tend to overestimate the cross section in the recoil region especially for $E_2 = 3$ eV ejection energy.

A comparison at equal energy sharing ($E_1 = E_2 = 23$ eV) is provided in Figure 4, where the cross sections in the scattering and perpendicular planes are shown in the left and right column, respectively, and also the 3D patterns are presented for experiment (left column) and CCC calculations (right column). The results at $\theta_1 = -30^\circ$ scattering angle show again the basic binary and recoil peaks in the scattering plane and three peaks in the perpendicular plane. Moving to $\theta_1 = -60^\circ$ and -120° , the binary to recoil ratio is increasing strongly. The binary and recoil peaks are shifted towards the direction of $\theta_2 = 0^\circ$, and two side peaks in the perpendicular plane merge to one strong maximum at θ_2 around 0° . In comparison with theory this equal energy sharing case is the only one where distinct deviations of the CCC result can be found. These are the binary peak magnitude in Figure 4 (c) as well as the recoil peak heights in (d) and (e). These discrepancies do not seem to follow a systematic trend. On the other hand different from the asymmetric energy sharing cases the TDCC results here are in good agreement and even surpass the accuracy of the CCC result. Examples are the magnitudes of the binary peak in Figure 4 (c) and the recoil peak in Figure 4 (e).

In order to judge the accuracy of the experimental data it should be noted that the statistical errors are given by the error bars of the data points. Possible systematic errors are hard to quantify but one can get an idea of their importance considering that the TDCS in the perpendicular plane must be mirror symmetric with respect to $\theta_2 = 180^\circ$. This symmetry is fulfilled in all cases except in Figure 4 (d) where the FDCS between 30° and 60° shows some deviation from the respective data between 300° and 330° .

Overall the experimental absolutely normalized TDCS are excellently predicted by CCC theory for a large range of kinematics and collision geometries as shown in the Figures 1 – 4. The TDCC theory is very accurate in shape and magnitude of the TDCS at equal energy sharing (Figure 4). Concerning the discrepancies between TDCC and experiments, we note that the equal energy sharing TDCS converge much more rapidly with respect to the number of partial waves included in the calculation, compared to the very asymmetric energy sharing. Also, for the very slow electron TDCS, large radial meshes are often required. Although the TDCC calculations presented here appear well converged with respect to both of these parameters, we plan further convergence

checks for the most asymmetric energy sharing TDCS.

4. Summary

In summary, we have performed a comprehensive study of single ionization of helium by 70.6 eV electron impact. The experimental cross sections are compared to the predictions of the CCC and the TDCC calculations for a large range of kinematics and collision geometries. The TDCS as 3D patterns show excellent qualitative agreement. Comprehensive quantitative comparisons of experiment and theories are presented in the scattering and perpendicular planes. It is found that the TDCC and CCC calculations are in excellent agreement with the experiment for equal energy sharing. Moreover, all the features observed in the experiment, which cover a major part of the entire three-particle final-state configuration, are completely described by the CCC theory with sufficient accuracy including the absolute magnitude of the cross section. Therefore, we conclude that the theoretical description of single ionization of helium by electron impact leaving the ion in the ground state can be sufficiently accurate for many practical purposes.

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Appendix

Table I. Experimental absolute TDCS in the scattering ($//$) and perpendicular (\perp) plane as a function of the emission angle (θ_2) of an electron with $E_2 = 5$ eV. The scattering angle (θ_1) is varied from -10° to -30° . The TDCS are in units of $10^{-21} \text{cm}^2 \text{sr}^{-2} \text{eV}^{-1}$.

θ_2 ($^\circ$)	$\theta_1 = -10^\circ$		$\theta_1 = -15^\circ$		$\theta_1 = -20^\circ$		$\theta_1 = -25^\circ$		$\theta_1 = -30^\circ$	
	$//$	\perp	$//$	\perp	$//$	\perp	$//$	\perp	$//$	\perp
25	101.8	46.1	44.5	21.8	25.5	10.7				
35	178.6	69.1	87.2	33.6	41.5	17.9	25.3	11.2	20.8	13.7
45	287.3	80.5	203.2	55.5	95.7	27.3	50.2	21.8	28.2	14.6
55	381.6	116.6	264.2	68.2	190.2	46.7	97.8	27.1	51.8	19.7
65	461.3	130.0	368.5	93.5	231.5	54.0	135.3	35.8	71.0	24.5
75	435.0	115.0	384.5	84.5	245.8	54.2	154.2	38.8	84.4	27.3
85	309.0	86.0	291.8	81.8	212.6	49.8	137.0	37.3	80.2	23.9
95	217.0	97.0	226.8	95.5	168.8	57.7	123.0	32.7	69.7	21.2
105	99.0	132.0	73.8	71.3	65.9	48.9	47.6	23.5	29.8	15.1
115	68.0	205.6	48.7	114.9	34.2	73.5	21.8	33.0	17.1	16.0
125	95.0	345.0	59.7	170.2	22.8	87.9	12.0	39.7	8.7	20.6
135	177.6	354.0	65.3	193.6	29.8	109.0	15.8	53.4	7.9	27.5
145	235.0	444.0	110.5	261.4	43.0	130.2	20.3	57.8	8.4	30.5
155	335.0	598.9	153.2	266.1	77.5	155.7	43.6	82.6	16.7	37.2
205	796.1	587.3	427.6	294.8	221.0	165.5	119.0	87.3	60.6	41.4
215	779.0	451.0	418.6	306.8	225.9	136.5	116.4	72.5	57.1	36.5
225	652.8	403.0	393.2	232.3	212.5	126.5	110.3	55.1	51.5	29.4
235	592.0	312.0	364.5	156.7	201.8	68.9	92.1	34.9	48.2	23.8
245	504.8	222.4	311.8	122.8	184.2	85.0	77.5	40.4	39.8	24.5
255	374.0	151.0	220.4	90.5	128.2	53.6	57.9	32.9	31.8	17.5
265	259.0	102.0	165.9	102.3	106.0	65.1	51.3	38.0	26.1	22.8
275	141.0	104.0	112.7	88.2	67.4	56.3	42.5	36.0	23.0	27.1
285	89.0	103.0	89.1	93.6	52.1	59.3	30.7	36.8	17.1	23.2
295	66.3	131.3	58.9	114.5	36.3	49.6	21.8	34.5	11.2	27.4
305	32.9	105.8	37.3	69.6	21.7	46.6	18.2	26.5	8.3	17.8
315	19.4	82.9	16.5	53.5	12.6	27.7	7.5	14.9	4.0	9.3
325	28.8	84.5	8.0	32.8	5.8	18.3	3.5	7.7	4.9	12.5
335	26.9	51.8	9.6	27.9	2.7	5.8				

Table II. Experimental absolute TDCS in the scattering ($//$) and perpendicular (\perp) plane as a function of the ejected electron emission angle (θ_2) for two ejected electron energies of $E_2 = 3$ eV and 10 eV and two scattering angles of $\theta_1 = -10^\circ$ and -30° . The TDCS are in units of $10^{-21} \text{cm}^2 \text{sr}^{-2} \text{eV}^{-1}$.

θ_2 ($^\circ$)	$\theta_1 = -10^\circ, E_2 = 3\text{eV}$		$\theta_1 = -10^\circ, E_2 = 10\text{eV}$		$\theta_1 = -30^\circ, E_2 = 3\text{eV}$		$\theta_1 = -30^\circ, E_2 = 10\text{eV}$	
	$//$	\perp	$//$	\perp	$//$	\perp	$//$	\perp
35	157.1	52.4	73.5	25.3			19.3	6.3
45	265.7	77.2	149.9	40.8	22.7	12.9	45.3	11.4
55	450.0	148.0	215.7	56.7	43.0	18.9	65.5	17.5
65	570.9	163.6	252.2	67.3	57.4	19.8	88.6	20.8
75	519.3	137.5	215.5	48.4	69.3	21.8	90.3	26.5
85	463.6	89.1	153.0	55.6	69.9	23.8	77.3	25.0
95	379.6	91.6	83.5	46.4	60.2	17.2	54.3	23.6
105	129.1	125.5	52.8	73.9	32.6	14.6	27.0	20.6
115	82.9	235.6	58.1	121.7	22.9	17.4	10.6	19.2
125	109.1	463.6	85.5	136.8	13.3	23.0	7.6	17.8
135	260.9	551.1	141.0	153.2	9.4	26.1	9.3	27.3
145			196.5	226.3			14.8	33.6
155			243.5	236.1			18.5	35.1
205			362.5	236.9			46.9	31.5
215			335.3	211.7			45.4	31.3
225	971.8	545.9	281.0	156.0	67.2	30.6	40.8	27.6
235	950.9	456.4	235.5	122.0	64.1	25.6	37.5	22.4
245	746.2	276.4	220.1	112.0	48.0	20.7	33.1	25.3
255	392.7	165.5	169.2	78.4	32.6	19.4	30.6	26.3
265	315.8	111.3	96.5	64.4	27.8	20.8	23.6	22.8
275	220.0	127.3	85.0	52.8	22.8	21.6	19.0	29.2
285	154.9	106.9	41.0	57.2	15.1	19.0	11.2	27.0
295	89.1	174.5	38.4	67.6	15.2	20.7	6.2	24.9
305	68.0	148.0	28.6	42.4	9.6	20.6	5.9	17.4
315	28.8	87.7	16.9	38.2	4.5	9.4	4.4	13.1
325	20.9	52.4	13.4	28.2			1.7	7.3

Table III. Experimental absolute TDCS in the scattering (\parallel) and perpendicular (\perp) plane for equal energy sharing ($E_1 = E_2 = 23$ eV) as a function of electron emission angle (θ_2) at different scattering angles $\theta_1 = -30^\circ, -60^\circ$ and -120° . The TDCS are in units of $10^{-21} \text{cm}^2 \text{sr}^{-2} \text{eV}^{-1}$.

θ_2 ($^\circ$)	$\theta_1 = -30^\circ$		$\theta_1 = -60^\circ$		$\theta_1 = -120^\circ$	
	\parallel	\perp	\parallel	\perp	\parallel	\perp
5	4.6	3.7	42.7	32.8	39.0	40.3
15	5.8	3.2	55.1	25.2	32.0	37.0
25	19.7	4.2	54.8	16.5	12.8	21.1
35	22.2	4.2	32.8	10.9	4.0	10.0
45	42.8	6.3	16.6	5.4	2.4	4.5
55	54.2	9.4	9.6	4.9	1.9	2.5
65	52.9	11.8	6.3	4.3	1.8	1.3
75	35.8	13.1	4.2	3.8	2.1	1.1
85	24.0	9.7	2.1	2.8	1.8	0.5
95	13.4	11.5	1.6	2.4	1.0	0.5
105	6.9	10.0	1.2	2.6	1.8	0.4
115	9.8	10.8	1.2	2.4	1.8	0.5
125	9.5	13.7	2.3	2.7	1.4	0.1
135	13.1	15.7	2.7	3.2	1.8	0.5
145	18.2	16.7	4.3	5.4	1.9	0.3
155	15.3	17.7	8.3	6.7	1.3	0.6
165	18.0	23.1	9.2	7.2	1.3	0.6
175	17.9	20.1	8.7	8.4	0.8	0.3
185	21.0	17.9	7.3	7.6	0.1	0.6
195	21.2	17.7	6.7	7.7	0.0	0.5
205	19.8	14.8	3.2	6.2	0.0	0.6
215	15.9	15.4	1.8	5.5	0.0	0.6
225	14.9	12.5	1.1	3.4	0.1	0.6
235	13.5	13.5	0.7	2.8	0.0	0.5
245	8.6	10.5	0.2	2.1	0.0	0.5
255	7.2	9.7	0.1	2.3	0.0	0.6
265	6.0	11.5	0.0	1.9	0.1	0.8
275	6.6	13.8	0.0	3.1	0.0	1.0
285	2.3	14.6	0.0	3.8	0.3	2.0
295	1.4	14.5	0.0	4.8	0.3	2.1
305	0.3	13.4	0.0	6.0	2.1	2.6
315	0.2	10.8	0.1	7.8	3.8	3.9
325	0.0	5.6	0.6	11.6	9.0	6.4
335	0.5	7.1	3.0	16.8	21.1	15.8
345	1.8	4.2	8.2	22.1	32.9	25.1
355	2.3	3.5	22.2	31.4	40.0	40.4

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Fig. 1

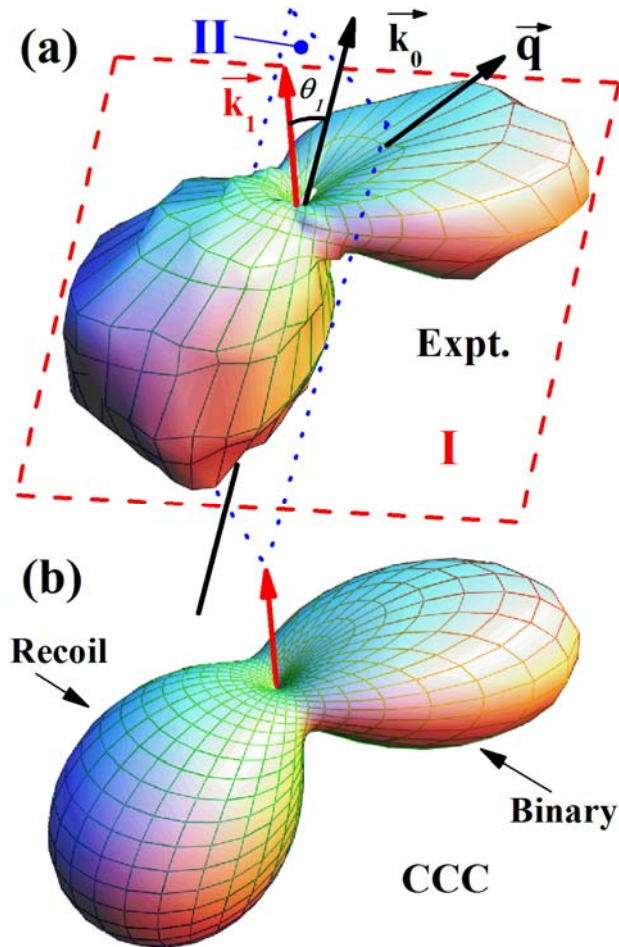


Figure 1. (Color online) Absolute TDCS for $(e, 2e)$ on He as a function of the emission angle of an electron with kinetic energy of $E_2 = 5$ eV. The emission angle of the other electron ($E_1 = 41$ eV) is fixed to $\theta_1 = -20^\circ$. (a) Experimental 3D cross section. (b) CCC calculation. The dashed and dotted frames in (a) indicate the cutting planes through 3D pattern in the scattering (**I**) and perpendicular plane (**II**), respectively.

Fig. 2

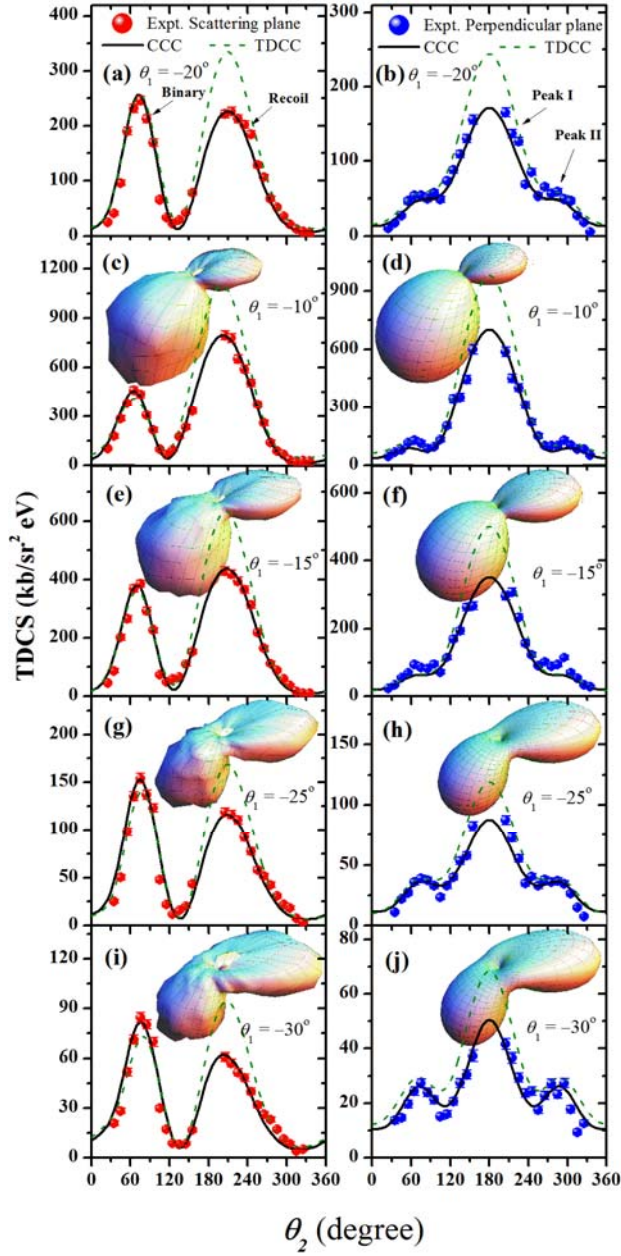


Figure 2. (Color online) Absolute TDCS in the scattering (left column) and perpendicular plane (right column) as a function of the emission angle of an electron with $E_2 = 5$ eV. The other electron's ($E_1 = 41$ eV) emission angle θ_1 is fixed to: (a) and (b) $\theta_1 = -20^\circ$; (c) and (d) $\theta_1 = -10^\circ$; (e) and (f) $\theta_1 = -15^\circ$; (g) and (h) $\theta_1 = -25^\circ$; (i) and (j) $\theta_1 = -30^\circ$. The insets in (c) to (j) are the 3D patterns of the TDCS for experiment (left column) and CCC calculation (right column).

Fig. 3

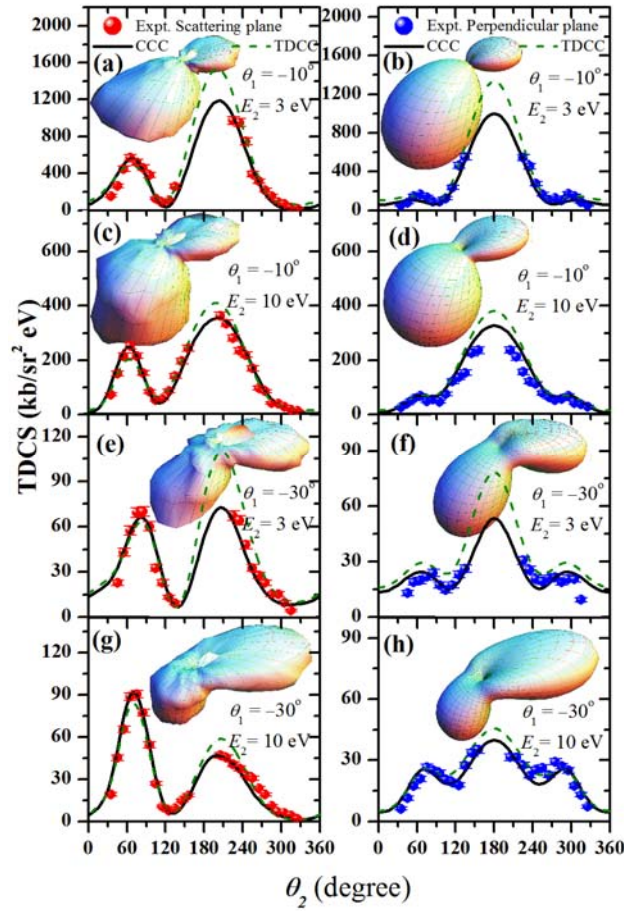


Figure 3. (Color online) Absolute TDCS in the scattering (left column) and perpendicular plane (right column) as a function of the emission angle θ_2 of one electron with energy E_2 . The other electron's emission angle θ_1 is fixed: (a) and (b) $\theta_1 = -10^\circ$, $E_2 = 3$ eV ; (c) and (d) $\theta_1 = -10^\circ$, $E_2 = 10$ eV; (e) and (f) $\theta_1 = -30^\circ$, $E_2 = 3$ eV; (g) and (h) $\theta_1 = -30^\circ$, $E_2 = 10$ eV. The insets in (a) to (h) are the 3D patterns of the TDCS for the experiment (left column) and CCC calculation (right column).

Fig. 4

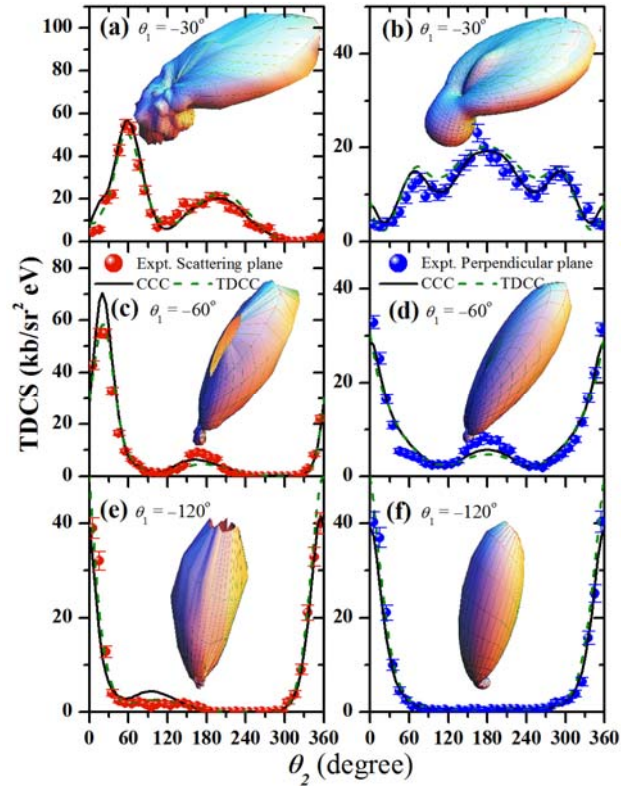


Figure 4. (Color online) Absolute TDCS in the scattering (left column) and perpendicular plane (right column) for equal energy sharing ($E_1 = E_2 = 23$ eV) as a function of the emission angle of one electron (θ_2) with the other electron's emission angle θ_1 being fixed to: (a) and (b) $\theta_1 = -30^\circ$; (c) and (d) $\theta_1 = -60^\circ$; (e) and (f) $\theta_1 = -120^\circ$. The insets in (a) to (f) are the 3D patterns of the TDCS for the experiment (left column) and CCC calculation (right column).