Helicity Dependence of the Photon-Induced Three-Body Coulomb Fragmentation of Helium Investigated by Cold Target Recoil Ion Momentum Spectroscopy

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The angular and energy dependence of circular dichroism in photo-double-ionization of helium at a photon energy of 99 eV is investigated. Using cold target recoil ion momentum spectroscopy the absolute fivefold differential cross section has been obtained by a coincident measurement of the vector momenta of one electron and the recoiling He^{2+} ion covering all relative azimuthal and polar angles. The experimental results are contrasted with numerical calculations using different helium ground state wave functions and forms of the dipole operator. [S0031-9007(98)06337-6]

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Helicity adds a new twist to photo-double-ionization of helium: How is the handedness of a circularly polarized photon, absorbed by the spherical symmetric He^{1S} ground state, transferred to the three-body Coulomb continuum? The handedness leads to a symmetry break in the phase space of the diverging two electrons and the target nucleus. This additional aspect provides a novel tool for the investigation of the photon-induced fragmentation of helium, which is one of the simplest and hence most fundamental manifestations of electron-electron correlation. Helicity dependence can also be used to prove symmetry principles of the three-body Coulomb problem as parity conservation and time reversal invariance.

Helicity dependence in photoionization is termed circular dichroism (CD). CD is well known in magnetic solids and chiral or aligned molecules (for a recent review see [1]). However, Berakdar and Klar [2] have theoretically shown that CD does not require a chiral or aligned initial state. They predicted that CD may also be observable in a coincidence measurement (e.g., of the two electrons) in photo-double-ionization of helium from its ground state. In general an appropriate experimental approach to investigate a many body system is to measure the fully differential cross section for the fragmentation of the system by a coincident determination of the vector momenta of all outgoing particles. In their pioneering coincidence experiment Schwarzkopf *et al.* [3] reported the first fivefold differential cross sections (5DCS) [4] for photo-double-ionization of helium using linearly polarized light. Also the technique of cold target recoil ion momentum spectroscopy (COLTRIMS) has already been used to perform fully differential experiments on photo-double-ionization of helium with linearly polarized light [5]. In 1996 Viefhaus *et al.* [6] reported the first experimental evidence for CD in the helical photo-double-ionization of helium. They obtained the relative 5DCS for five electron energies each at three fixed angles.

In the present experiment we have used COLTRIMS [7–9] to investigate the helical photo-double-ionization of helium at an energy of 20 eV above the double ionization threshold. We have measured the 5DCS for all relative polar and azimuthal angles in the electron energy range of 11.5 through 20 eV. A coincidence solid angle of 10% has been achieved, which is an increase of nearly 3 orders of magnitude compared to the previous experiment [6]. Our experimental technique provides a high statistical accuracy and allows a straightforward normalization, therefore we can put the 5DCS on an absolute scale. In addition, the helicity dependence of the reaction is investigated from different perspectives. We transform the 5DCS in momentum coordinates of both one electron and the recoil ion with respect to the other electron. Our detailed experimental results are compared with numerical calculations in length and velocity form of the dipole operator using different correlated wave functions for helium ground state.

We define CD as

$$\mathrm{CD} = \frac{5\mathrm{DCS}(\sigma^{+}) - 5\mathrm{DCS}(\sigma^{-})}{5\mathrm{DCS}(\sigma^{+}) + 5\mathrm{DCS}(\sigma^{-})}, \qquad (1)$$

where $5\mathrm{DCS}(\sigma^{+/-})$ is the cross section for positive/negative helicity. Appearance of nonzero CD requires that all three dimensions of space are spanned. These constraints on the momentum vectors of the incoming photon ($\mathbf{k}_\gamma$) and two of the three outgoing particles can be derived to [2,10]

$$\mathrm{CD}(\mathbf{k}_1, \mathbf{k}_2) = -i \sum_L \underbrace{f_{LL}(k_1, k_2)}_{\text{(i)}} \underbrace{\mathbf{B}^{LL}_{10}(\hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2)}_{\text{(ii)}} . \quad (2)$$

Here $\mathbf{k}_\gamma$ is taken as quantization axis, $\mathbf{B}$ is a bipolar harmonics as a function of the directions $\hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2$ of the outgoing electrons, and $f(k_1, k_2)$ depends only on the magnitudes k_1, k_2 of the electron momenta. Like all observables, CD must be invariant under exchange of the two electrons, i.e., $\mathrm{CD}(\mathbf{k}_1, \mathbf{k}_2) = \mathrm{CD}(\mathbf{k}_2, \mathbf{k}_1)$. Since $\mathbf{B}(\hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2) = -\mathbf{B}(\hat{\mathbf{k}}_2, \hat{\mathbf{k}}_1)$, it follows from Eq. (2) that $f(k_1, k_2)$ is antisymmetric against exchange of the two electrons $[f(k_1, k_2) = -f(k_2, k_1)]$. The angular distributions are sufficiently illustrated by the bipolar harmonics for $L = 1$

$$\mathbf{B}^{11}_{10}(\hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2) = \frac{i}{\sqrt{2}} (\hat{\mathbf{k}}_1 \times \hat{\mathbf{k}}_2) \cdot \hat{\mathbf{k}}_\gamma . \quad (3)$$

Therefore, nonvanishing CD requires the following: (i) Unequal energy sharing of the two electrons. (ii) Linear independence of $\mathbf{k}_1, \mathbf{k}_2$, and $\mathbf{k}_\gamma$, since CD is a pseudoscalar in the laboratory system. From Eq. (2) it follows that CD is zero when integrating over the direction of one electron (e.g., in a noncoincident experiment), or in the case at least one of the outgoing electrons is in an s state. The latter case is anticipated when one electron is very fast (carrying the angular momentum of the photon) while the other is very slow.

The experiment was performed at the helical undulator beam line BL28A [11] of the Photon Factory in Tsukuba/Japan. The degree of circular polarization (Stokes parameter S_3) was about $S_3 = \pm 0.95$. The angle of the polarization ellipse as well as the degree of linear polarization was monitored during the whole experiment by observing the emission characteristics of the He^{1+} ions produced by single photoionization. The polarization properties are in good agreement with [12]. The effect of the noncircular contributions on the presented 5DCS was carefully estimated and found to be not significant. However, the CD presented later in Fig. 3 is corrected by $1/|S_3|$. We investigated photon energies of 99 and 174 eV corresponding to excess energies $E = 20$ and 95 eV. Here we present only the data for $E = 20$ eV. A precooled supersonic helium gas jet was used as the target. It combines the two most important features for high resolution recoil ion momentum spectroscopy: low internal temperature (<100 mK) and localization of the target ($\phi = 1$ mm). Because of the large coincidence efficiency of our "momentum-microscope" we could close the entrance and exit slits of the beam line to 25 μm, yielding a photon energy resolution of ±0.2 eV. Recoil ions created in the intersection volume of the gas jet and the photon beam are accelerated by a weak electric field (1.4 V/cm) onto a two-dimensional position-sensitive microchannel plate (MCP) detector. The extraction provides a detection solid angle of $\Delta\Omega = 4\pi$ for the recoil ions. An electric field configuration, which focuses the extension of the target region in all three dimensions, was used for the ion extraction. Opposite to the recoil ion detector a second position-sensitive MCP detector with wedge-and-strip readout was placed to detect electrons, yielding a solid angle of $\Delta\Omega/4\pi = 10\%$. A stack of three meshes was mounted in front of the electron detector. The central mesh has been used as retarder in order to detect only the "fast" electrons from 11.5 to 20 eV. With this setup a coincidence count rate of up to 5 s^{-1} was achieved. The COLTRIM spectrometer is shown in Fig. 1.

For each detected double ionization event we recorded five independent observables: the two-dimensional position information of both the recoil ion $(y_{\mathrm{ion}}, z_{\mathrm{ion}})$ and the fast electron $(y_{\mathrm{e1}}, z_{\mathrm{e1}})$ as well as the time-of-flight difference (Δt) between the electron and ion signal. The electron and ion time of flight could not be measured separately due to the use of multibunch operation with a bunch distance of 2 ns in the storage ring. However, using these five observables together with the energy and the three momentum conservation laws one can calculate for each event all nine momentum components of the ion $\mathbf{K}$ and the two electrons $\mathbf{k}_1, \mathbf{k}_2$ uniquely. With this advanced COLTRIMS setup we achieved a momentum resolution for the recoil ions of $\Delta K_{x,z} = \pm 0.075$ a.u. and $\Delta K_y = \pm 0.15$ a.u. For the electrons a momentum resolution of ±0.015 a.u. has been obtained for all components. This corresponds to an electron energy resolution of ±0.5 eV. We used both σ^+ and σ^- helicity to control systematic errors in the experiment, since $5\mathrm{DCS}(\sigma^+)$ must turn into $5\mathrm{DCS}(\sigma^-)$ by inverting the handedness of the three vectors $\hat{\mathbf{k}}_1, \hat{\mathbf{k}}_2, \hat{\mathbf{k}}_\gamma$ as shown by Eq. (3).

Figure 2 shows the recoil ion and electron momentum distribution in Cartesian coordinates in the x-y plane at $K_z = 0$ and $k_{z2} = 0$. The wave vector $\mathbf{k}_\gamma$ points out of the plane of paper. For all four plots we fixed e_1 within $0.9 < k_{x1} < 1.2$ a.u. and with $k_{y1} = k_{z1} = 0$. Thus (a) and (b) show the 5DCS $d^5\sigma/(dk_{x1}dk_{y1}dK_x dK_y dK_z)$ and (c) and (d) $d^5\sigma/(dk_{x1}dk_{y1}dk_{x2}dk_{y2}dk_{z2})$. The circles represent the maximum momentum for the ion and

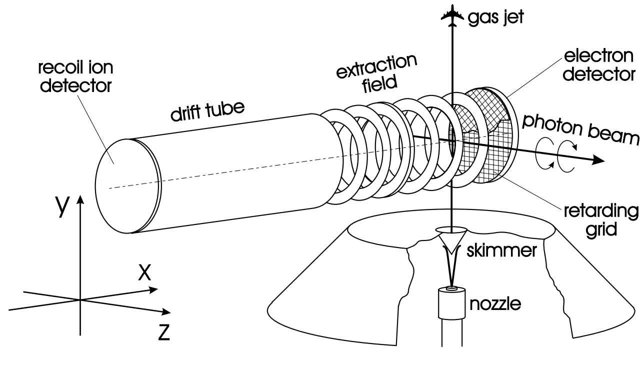

FIG. 1. Cold target recoil ion momentum spectrometer: The photon beam (z direction) is intersected with the supersonic gas jet (y direction) in a weak electric field pointing in the x direction. Electrons and recoil ions are detected by position-sensitive microchannel plate detectors.

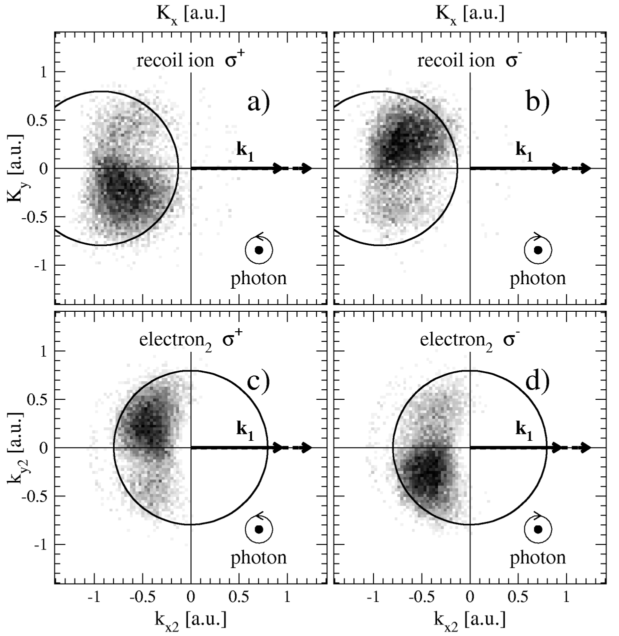

FIG. 2. Recoil ion and electron momentum distribution in the x-y plane for both σ^+ and σ^- as indicated. We fixed e_1 within $0.9 < k_{x1} < 1.2$ a.u. and with $k_{y1} = k_{z1} = 0$. The wave vector $\mathbf{k}_\gamma$ points out of the plane of paper. Circles: maximum magnitude of $\mathbf{K}$ and $\mathbf{k}_2$. The grey scale represents the 5DCS on a linear scale.

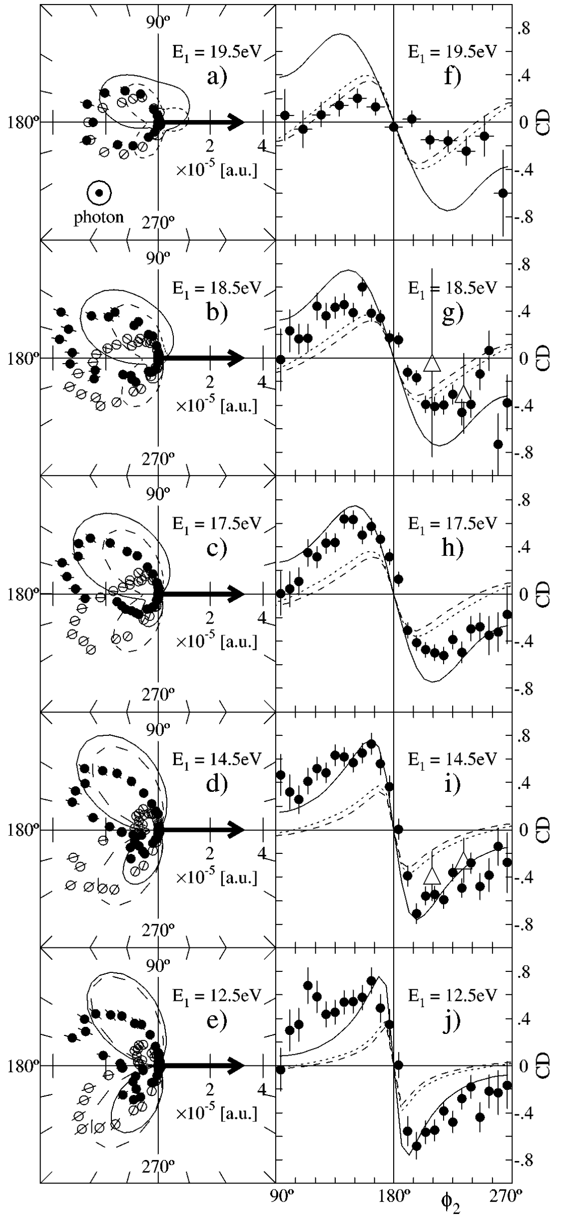

FIG. 3. (a)–(e) Polar plot of the 5DCS $d^5\sigma/(d^2\Omega_1 d^2\Omega_2 dE_1)$ in atomic units. (f)–(j) CD, both as a function of ϕ_2. We chose E_1 as indicated in the figure and $\phi_1 = 0$, $\theta_1 = \theta_2 = 90°$. $\mathbf{k}_\gamma$ points out of the plane of paper. The arrow represents $\hat{\mathbf{k}}_1$. (a)–(e) Experiment for σ^+ (full dots) and σ^- (open dots). Theory for σ^+ using Φ_{H}, calculated in velocity form, multiplied by 4 (full curve) and the same in length form divided by 3 (dashed curve). (f)–(j) Experiment (full dots); theory Φ_{H}, velocity form (full curve); Φ_{H}, length form (dashed curve); and Φ_{S}, length form (dotted curve). Experiment by [6] (open triangles).

e_2, respectively. The 5DCS for the recoil ion peaks around $K_x = -0.7$ and the 5DCS for e_2 around $k_{x2} = -0.4$ due to the fact that the ion and e_2 have to compensate for k_{x1} ($\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{K} = \mathbf{k}_\gamma \approx 0$). The effect of CD is manifested in a shift of the momenta $\mathbf{K}$ and $\mathbf{k}_2$ up and down along the y axis leading to asymmetric distributions with respect to the x axis. The maximum for the ions for σ^+ is at negative K_y and for electrons at positive k_{y2}. The 5DCS for σ^- shows the mirror image of the one for σ^+ with respect to the x axis.

For a quantitative comparison of the experimental and theoretical results, we transform the 5DCS in spherical coordinates. $\theta_{1,2}$ are the polar angles with respect to the z axis (photon direction), $\phi_{1,2}$ the azimuthal angles in the x-y plane, and $E_{1,2}$ the energies of the two electrons. The subscript 1 denotes the fast detected electron. Figures 3(a)–3(e) show the 5DCS $d^5\sigma/(d^2\Omega_1 d^2\Omega_2 dE_1)$ and 3(f)–3(j) the CD, both for coplanar geometry and as a function of ϕ_2. The experimental data are integrated over ±0.5 eV centered around the indicated values for E_1. Note that we cover 360° of ϕ_2 for the 5DCS due to our solid angle of 4π for the recoil ion detection. The measured 5DCS for ϕ_2 between 270° and 90° is too small to calculate the CD in this range.

Our numerical calculations are based on first order pertubation theory using the dipole approximation [2,10]. The final state is described by a 3C-wave function [13]. We use two different correlated helium ground states, a three-parameter Hylleraas-type [14] (Φ_{H}) and a wave function according to Siebbeles *et al.* [15] (Φ_{S}) which fulfills the Kato-cusp condition. We calculated the 5DCS and the CD using Φ_{S} in length and using Φ_{H} in length and velocity form.

Figures 3(a)–3(e) show the absolute 5DCS for σ^+ (full dots) and σ^- (open dots). The full curve shows the 5DCS for σ^+ using Φ_{H} calculated in velocity form and the dashed curve in length form. The results in velocity form are multiplied by 4 while the ones in length form are divided by 3. Three main features are observed in the shape of the 5DCS as follows.

(I) Mirror symmetry with respect to the x axis for exchange of the helicity. (Because of this mirror symmetry we restrict the following discussion to the results for σ^+.)

(II) The maximum of the 5DCS(σ^+) is found for all measured energy sharings at ϕ_2 between 90° and 180°, which means that the momenta of the fast ($\mathbf{k}_1$) and the slow ($\mathbf{k}_2$) electrons together with $\mathbf{k}_\gamma$ form most likely a right-handed system $[(\mathbf{k}_1 \times \mathbf{k}_2) \cdot \mathbf{k}_\gamma > 0]$.

(III) A second maximum between 180° and 270° appears and increases when going from $E_1 = 17.5$ to 12.5 eV.

For the most unequal energy sharing $E_1 = 19.5$ eV the experimental 5DCS is nearly symmetric with respect to the x axis. This may be a consequence of requirement (ii) derived from Eq. (2). The appearance of the second lobe (III) is not surprising since for equal energy sharing a node in the 5DCS at back-to-back emission ($\phi_2 = 180°$) is expected in addition to a vanishing CD. Thus for $E_1 = E_2$ the 5DCS should have at least two equal lobes symmetric to $\phi_2 = 180°$ as in photo-double-ionization with linearly polarized light [3,16]. We observe partial agreement for the relative shape of the experimental 5DCS with the calculation in velocity form, while the shape using length form disagrees. The results using Φ_H or Φ_S as ground state do not differ very much if both are calculated in the same form. All calculated absolute results differ from the experimental data by at least a factor of 3. Similar approaches using a 3C-wave function for the final state were used for linearly polarized light [16], leading to good agreement with the shape of the 5DCS [3,17]. However, Maulbetsch *et al.* [18] have already shown that they generally fail to reproduce the absolute cross section, in some cases by orders of magnitude.

Figures 3(f)–3(j) show the circular dichroism as a function of ϕ_2. Our experimental results are consistent with [6]. Since they used $E = 14.5$ eV we scaled their E_1 by 1.38 in order to compare the same energy sharings E_1/E_2. For all investigated energy sharings the CD shows a maximum for ϕ_2 between 90° and 180°. In accordance with Eq. (3) we observe antisymmetry of the CD with respect to $\phi_2 = 180°$ [CD $(180° + \phi_2) = -$CD $(180° - \phi_2)$]. All three theoretical calculations describe the general trend of the observed CD. However, they lead to a factor of 2 different results if calculated in length or velocity form and fail to predict all details of the energy and angular dependence.

In conclusion, we used cold target recoil ion momentum spectroscopy at the helical undulator beam line 28A of the Photon Factory to image the three momentum components and the vector momenta of the recoil ion and one electron in coincidence. This is equivalent to a $(\gamma, 2e)$ experiment, leading to the fivefold differential cross section. We achieved a coincidence solid angle of $\Delta\Omega/4\pi = 10\%$ at $E = 20$ and 95 eV, mapping a large fraction of the square of the nine-dimensional continuum wave function. We observe discrepancies between experiment and theory indicating a puzzling lack of understanding of a fundamental three-body problem. Our results provide a challenge for future theoretical work and call for more experiments in this field covering the full final state over a solid angle of 4π. Such an experiment is scheduled for 1998 at the Photon Factory.

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