Experimental Evidence for Circular Dichroism in the Double Photoionization of Helium

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Circular dichroism in the double photoionization of He by circularly polarized light has been observed in an angle resolved coincidence experiment at 93.5 eV photon energy. The use of a transmission multilayer, which acts as a quarter-wave plate for the linearly polarized incident light, together with time-of-flight spectrometers, which enable simultaneous detection of all electron pairs, has made the measurements possible. The results show strong dependence of the dichroism on the relative emission angles and the energy sharing between the ejected electrons in good agreement with our numerical calculations. [S0031-9007(96)01618-3]

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Double photoionization of a free atom is one of the most striking manifestations of electron-electron correlations. The more direct way to study double photoionization is an experiment where the two photoelectrons are detected in coincidence, after energy and angle selection. This experiment results in the measurement of the triply differential cross section, $d^3\sigma/d\Omega_a d\Omega_b dE_a$ (TDCS), where Ω_a and Ω_b are the angles of emission of the two electrons and E_a is the energy of one of them. The energy of the other electron is defined by energy conservation $h\nu - I^{2+} = E_a + E_b$, where $h\nu$ is the photon energy and I^{2+} is the double ionization potential. Experimental investigations on the TDCS of the archetypal system for double photoionization, the helium atom, have been possible only since 1993 [1]. Recently Berakdar and Klar [2] have predicted that circular dichroism should be observed in double ionization of helium with circularly polarized radiation. Chirality in a system with a fully symmetric initial state (helium ground state $^1S_0$) might surprise us. Chirality requires distinguishable electrons and indeed Berakdar and Klar [2] and Berakdar *et al.* [3] have shown that the dipole matrix element produces the TDCS with a portion being antisymmetric with respect to the angular and energy exchange of the two electrons. This portion is the source of dichroism. Necessary conditions to observe a nonvanishing dichroism are as follows:

(i) The two electrons unevenly share the excess energy.

(ii) The direction of incident light and the directions of the two ejected electrons are not lying in one plane.

Moreover due to the fact that double photoionization occurs only because of electron correlations, the size and sign of the calculated dichroism depend sensitively on the approximate wave function used in the calculations. In particular the description of the Coulomb interaction in the final state plays a major role in determining shape and magnitude of the TDCS [3]. Only experiments can first prove the existence of the predicted effect and, second, stimulate the development of more sophisticated theoretical models.

In order to illustrate the effect in the angular pattern of the TDCS an example for $h\nu = 93.5$ eV is depicted in Fig. 1 for a fixed direction of one of the emitted photoelectrons ($\mathbf{k}_a$) and a particular unequal energy sharing ($E_a/E_b = 1/9$). Fixing the direction of emission of the second electron to $\mathbf{k}_b$ and changing the helicity

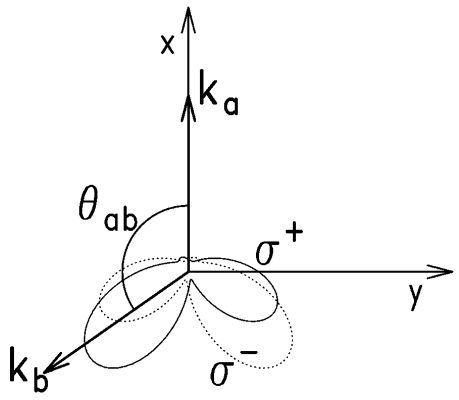

FIG. 1. Schematic drawing of the experimental geometry. The directions of emission of the two electrons given by $\mathbf{k}_a$ and $\mathbf{k}_b$ are lying in a plane perpendicular to the propagation direction of the circularly polarized ionizing radiation which is pointing into the plane of the drawing. The relative angle between the two directions of emission is given by θ_{ab}. A calculated TDCS for $h\nu = 93.5$ eV and $E_a/E_b = 1/9$ is shown for both helicities (σ^+/σ^-).

of the circularly polarized ionizing radiation, circular dichroism would result in a corresponding change in coincidence yield.

In this Letter we report on the observation of a circular dichroism effect in the double ionization of helium measured by angle resolved electron-electron coincidence spectrometry using circularly polarized monochromatic synchrotron radiation. We describe briefly the experimental setup as well as the data analysis. The results are finally compared with *ab initio* calculations.

Until recently, pure circularly polarized radiation was restricted to photon energies below 30 eV [4]. While in the soft x-ray energy region a special insertion device, a helical undulator, is already available to produce circularly polarized radiation directly [5], for extreme ultraviolet (XUV) radiation (double ionization threshold of helium at $h\nu = 79$ eV) we had to develop a quarter-wave plate to phase shift the linearly polarized light [6] of a conventional undulator.

The experiment was performed at the Berliner Elektronenspeicherring-Gesellschaft für Synchrotronstrahlung (BESSY) under single bunch conditions of the electron storage ring. Figure 2 shows the experimental setup. Monochromatized, highly linearly polarized radiation was first converted into circularly polarized light by a Mo/Si transmission multilayer (50 bilayers, $d = 9$ nm, working range 93 ± 5 eV). For an incoming monochromatic photon flux of approximately 10^{13} photons per second per 100 mA ring current (gap setting 38.2 mm, no pinhole, resolving power $E/dE \approx 300$) of the U1-TGM5 beam line [7] a flux higher than 10^{11} photons/s was achieved in the interaction region.

For the quantitative interpretation of the measured TDCS the degree of circular polarization of the ionizing radiation has to be known. The light polarization state was monitored by a Rabinovitch-type [8] linear polarization analyzer consisting of a gold mirror (incident angle 42°) and a GaAs photodiode. The analyzing power

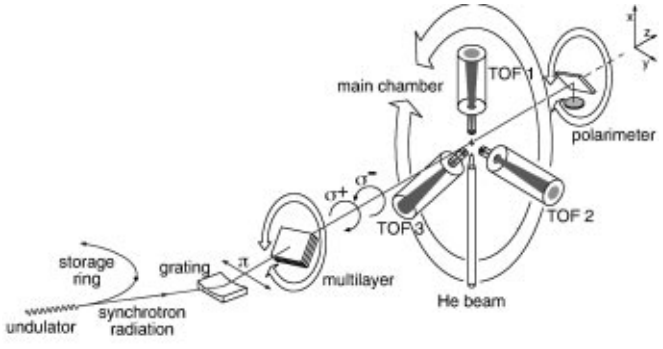

FIG. 2. Schematic drawing of the experimental setup. Three TOF electron analyzers are mounted in a plane perpendicular to the incoming photon beam. The linear polarized radiation is transformed into circular polarized radiation by a transmission multilayer positioned at the entrance of the vacuum chamber. The helicity of the radiation can be switched using appropriate polar angles for the multilayer.

of the polarimeter was higher than 0.97. First the degree of linear polarization was measured without the multilayer ($P_{\text{lin}} = 0.95 \pm 0.02$). Afterwards the optimum incident angle of the multilayer for the given photon energy was determined by minimizing the intensity modulation measured by rotating the polarimeter (<2%). The polarimeter alone cannot distinguish between unpolarized and circularly polarized radiation. However, by successively changing the polar angles of the multilayer and rotating the polarimeter around the light axis, the changing ellipse of light polarization, being circles and lines in the extreme cases, could be observed. At given polar angles the incoming linear polarization was completely converted into circular polarization ($P_{\text{circ}} = P_{\text{lin}}$) [9]. During the experiment the helicity of the radiation was changed every 1000 s by switching between two appropriate polar angles of the multilayer. The absence of linear polarized radiation behind the multilayer was verified after each change.

The circularly polarized radiation intersected at the interaction region an effusive beam of helium atoms. Photoelectrons were analyzed using three time-of-flight (TOF) spectrometers in a plane perpendicular to the incoming radiation. The three analyzers allow simultaneous coincidence measurements at three different relative emission angles θ_{ab} of the two electrons. The angular acceptance of the spectrometers is smaller than $\pm 4°$. Noncoincident as well as coincident TOF spectra were recorded at the same time simultaneously detecting photoelectrons of all possible kinetic energies [10]. An acceleration voltage was applied to the drift tube of the analyzers in order to detect electrons with kinetic energies as low as 0.5 eV. The energy resolution of the analyzers was in the range of 2%–5% of the kinetic energy of the photoelectrons. The photon energy of 93.5 eV was chosen as a compromise between multilayer working range, photon flux of the undulator, and helium double photoionization cross section (maximum ≈10 kb at ≈100 eV [11]). This resulted in a total true coincidence count rate in the order of 10 mHz.

The analysis involves the time-to-energy conversion of both coincident as well as noncoincident TOF spectra [Figs. 3(a) and 3(b)] and subsequently the subtraction of background and random coincidences. The true-to-random ratio, being much higher than one, allows an unambiguous identification of the single accessible final state in the double ionization photoelectron spectrum [Fig. 3(c)]. The TDCS was obtained by adding the coincidence counts over a certain energy interval [≈3 eV, marked as rectangles in Fig. 3(b)] to achieve a sufficient statistical accuracy. For one relative emission angle θ_{ab} and a particular helicity the TDCS is plotted versus kinetic energy E_a in Fig. 3(d). The TDCS has been put on a relative scale using the simultaneously collected noncoincident spectra of the three analyzers because all analyzers lay in a plane perpendicular to the direction of

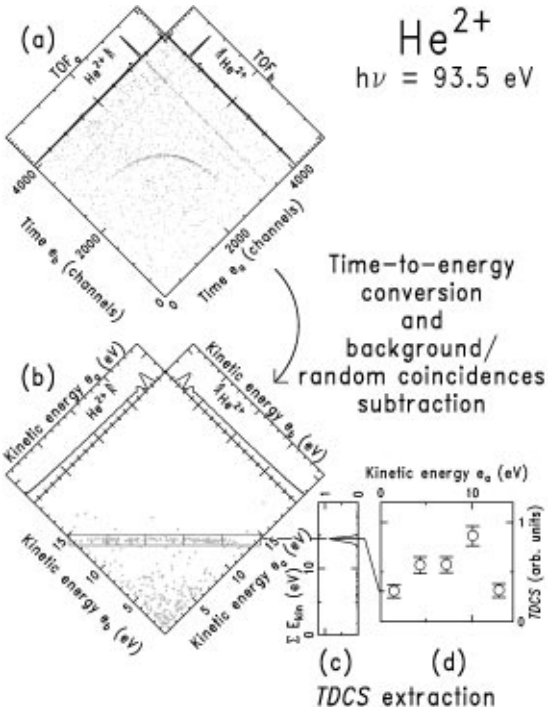

FIG. 3. Overview of the data analysis of the helium double photoionization electron spectra. (a) The noncoincident electron raw spectra of two TOF analyzers as well as the simultaneously recorded two dimensional raw coincidence spectrum of the corresponding analyzer pair. (b) The same spectra on kinetic energy scale and after subtraction of the background and random coincidence events, respectively. (c) The double ionization photoelectron spectrum, generated by plotting all coincidence events versus excess energy. (d) The TDCS intensity versus energy sharing.

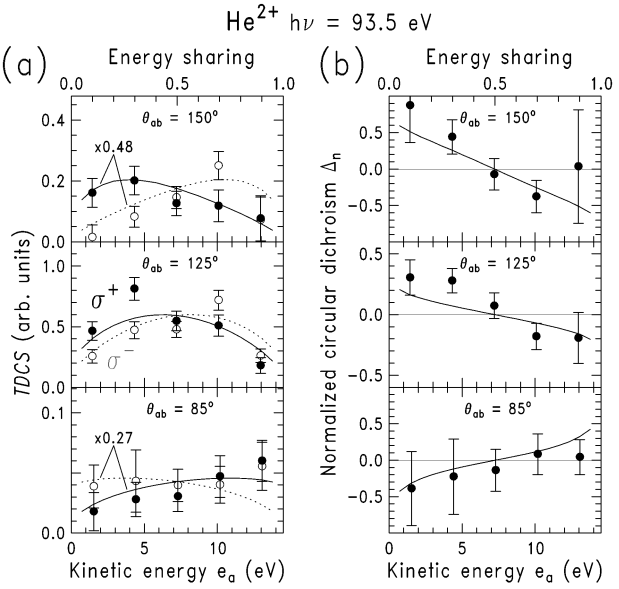

FIG. 4. (a) The helium double photoionization TDCS versus kinetic energy sharing at $h\nu = 93.5$ eV for three different relative emission angles θ_{ab} of the two electrons. The circles correspond to measurements with negative (open) and positive (full) helicity of the circularly polarized radiation. The solid and dotted curves are our theoretical results arbitrarily scaled (as quoted) with respect to the experimental TDCS. (b) Normalized circular dichroism for the three corresponding relative emission angles. The full curves represent our theoretical results. Note the different scaling of the three plots in (a) and (b), respectively.

propagation of the circularly polarized radiation. The low energy Auger spectrum of xenon [12] has been used to determine the relative transmission of the analyzers. The relative transmission did not change while switching the helicity of the radiation.

The resulting TDCS recorded with positive (σ^+) and negative (σ^-) helicity of the circularly polarized radiation for three different relative emission angles θ_{ab} are displayed in Fig. 4(a). In Fig. 4(b) the data were used to calculate the corresponding normalized circular dichroism effect Δ_n given by

$$\Delta_n = \frac{1}{P_{\text{circ}}} \frac{\text{TDCS}(\sigma^+) - \text{TDCS}(\sigma^-)}{\text{TDCS}(\sigma^+) + \text{TDCS}(\sigma^-)}. \qquad (1)$$

Two general features are observed in Fig. 4. First the circular dichroism vanishes for equal and almost equal energy sharing. This is due to the fact that the observer is not able to distinguish between the two electrons under this special condition. The second feature is that the TDCS curves measured with negative and positive helicity of the circularly polarized radiation are mirror images with respect to reflection at the equal energy sharing line. This is a consequence of the fact that changing the helicity of the radiation as well as changing the "labels" ("fast," "slow") of the two electrons leads to the same geometry of the experiment. These findings also show the proper operation of the experimental setup. Furthermore the mirror symmetry also tells that integration over all possible kinetic energy sharings leads to a vanishing difference between the TDCS intensities recorded with negative and positive helicity of the circularly polarized radiation. This stresses the fact that only coincidence measurements with a suitable energy (and angular) resolution enable the observation of circular dichroism.

As far as the results of the different relative angles are concerned it is obvious that the TDCS at $\theta_{ab} = 125°$ has the strongest intensity and clearly shows the circular dichroism. The $\theta_{ab} = 150°$ data show a larger circular dichroism effect despite the large error bars whereas the $\theta_{ab} = 85°$ ones show—if any—an effect with the opposite sign.

Numerical calculations are also shown together with the experimental results. They are obtained along the same lines as Refs. [2,3] using a Hylleraas-type wave function [13] as an approximation for the helium ground state and an approximate three-body scattering wave function [14] for the final state, respectively. The dipole operator was taken in length form. In Fig. 4(a) the calculated TDCS was arbitrarily scaled with respect to our experimental

data showing good qualitative agreement. However, to match the magnitude of the experimental values we had to use individual scaling factors as indicated. Nevertheless there is agreement on the fact that the TDCS magnitude is strongest at $\theta_{ab} = 125°$ and is lowering going towards $\theta_{ab} = 150°$ and being smallest at $\theta_{ab} = 85°$. Comparing the normalized dichroism and thus avoiding any fit of the scales the experimental data points are reproduced very well by the theoretical curves. The sign change as well as the general shape of the theory is consistent with the experiment although at the $\theta_{ab} = 125°$ geometry a slightly larger dichroism is found in the experiment. The present comparison between theory and experiment with respect to the relative size of the TDCS suffers from the statistical uncertainties of the experiment, which hamper a definite judgment of the discrepancies observed. However the differences in the TDCS seem to indicate that further theoretical work beyond the scope of this Letter has to be performed.

In summary, we have reported the observation of circular dichroism effects in the double photoionization of helium. These effects strongly depend on the relative angle and energy sharing of the two emitted electrons. Good agreement is found with the results of our numerical calculations. Even with the large efficiency of the TOF coincidence spectrometers the low value of the TDCS hampers the systematic study needed to distinguish among the predictions based on different wave functions. However, the advent of new undulator beam lines producing circularly polarized XUV radiation will enable the accurate characterization of this newly observed effect, which adds a new facet to the three-body Coulomb system. An additional aspect of circular dichroism in double photoionization will also be the investigation of differences between simultaneous and sequential double photoionization such as Auger decay following photoelectron emission [15] where a deeper understanding would be a step further towards a complete experiment in atomic double photoionization.

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