Energy- and Angle-Resolved Double Photoionization in Helium

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The triple differential cross section for double photoionization in helium has been measured for the first time. Equal energy sharing and a simple geometry for the two emitted electrons were chosen in order to facilitate the comparison with theory. Good agreement between experimental and theoretical data is found for the angular correlation pattern if the mutual Coulomb repulsion between both escaping electrons is taken into account.

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Double photoionization—it might be in one of the two clearly defined limiting forms of a direct process with the simultaneous emission of two photoelectrons or a two-step process with the sequential emission of a photoelectron and an Auger electron, or even a mechanism in between —is due to electron correlations. Helium is the simplest correlated system that shows only direct double ionization and this for all photon energies above the double ionization threshold. Hence, it provides the ideal test case for theoretical treatments of three-particle breakup occurring in double photoionization, if the angle- and energy-resolved triple differential cross section $d^3\sigma/d\Omega_1 d\Omega_2 dE$ (TDCS) is considered. In spite of this importance only three theoretical predictions exist for the TDCS in helium [1-3], and experiments on energy- and angle-resolved photon-induced two-electron emission have hitherto concentrated on other systems (double photoionization in the outer p shell of krypton [4,5], xenon [5,6], and argon [7]; two-step double photoionization in xenon [8]). In this Letter we report the first experimental results for the TDCS of direct double photoionization in helium and compare them with theory.

The experiment has been performed at the electron storage ring BESSY in Berlin at the toroidal grating monochromator TGM4 by applying the method of angle-resolved electron spectrometry and measuring the two ejected electrons in coincidence in two separate spectrometers. The rate of true coincidences was of the order of 20 MHz; accidental coincidences were recorded simultaneously with the true ones and the ratio of true to accidental coincidences typically varied between 1 and 0.2, depending on the actual parameters.

The following parameters which determine the angular correlation pattern have been selected: (i) A photon energy of 99.0 eV (bandwidth 0.35 eV) leads to an excess energy $E_{\rm exc}=10.0$ eV which provides kinetic energies for the ejected electrons large enough to avoid possible disturbances in spectrometer transmission ("cutoff" effect at low kinetic energies) or by scattered electrons. (ii) The pass energies of the electron spectrometers are set to transmit only electrons which share the available excess energy equally ($E_1=E_2=E_{\rm exc}/2$). This selection brings the important advantage that the TDCS becomes insensitive to the circular components (Stokes parameter S_3) in the monochromatized light [9]. (iii) One of the electron spectrometers is placed at a fixed position and aligned to accept electrons which are emitted in the direction of the major axis of the polarization ellipse which defines the x direction. For completely polarized light (Stokes parameter $S_1=1$), this geometry leads to a TDCS pattern which has rotational symmetry around the x axis. In the present case the polarization properties of the incident light are described by $S_1=0.554(10)$, $S_2=0$, $S_3=$unknown, and an angle $\lambda=-14.7(1.0)°$ by which the polarization ellipse is tilted with respect to the plane of the storage ring. Because $S_1<1$ there is no longer axial symmetry but reflection symmetry with respect to the x-z plane remains (z being the direction of the photon beam). (iv) The second electron spectrometer is turned around the z direction. In this way a large portion of the expected angular correlation pattern can be exhausted.

One of the essential first checks for electron-electron coincidences in double photoionization concerns the unambiguous identification of the process. It can be seen from Fig. 1 that—within the range resulting from the finite photon bandwidth and instrumental resolution of

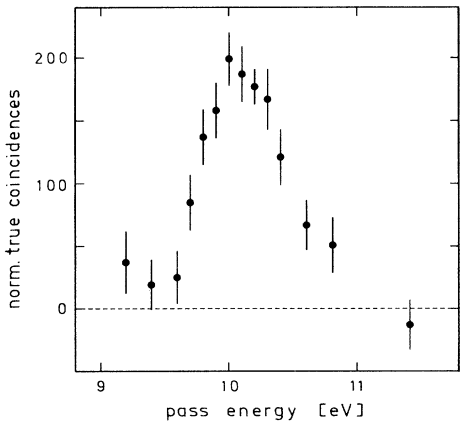

FIG. 1. Coincidence counts as a function of the pass energy E_2 of spectrometer 2 with the pass energy of spectrometer 1 set to $E_1=E_{\rm exc}/2=10$ eV.

both spectrometers—a nonvanishing coincidence signal occurs only if energy conservation is fulfilled.

In Fig. 2 our experimental results (points with error bars) for the TDCS are shown together with some theoretical predictions (solid, dashed, and dotted curves). First one can note several features which are characteristic of the TDCS in helium: (i) Practically no intensity is observed for two-electron emission in opposite directions. This is a consequence of a symmetry property of the two-electron wave function in the continuum (unfavored character). (ii) Practically no intensity is observed for two-electron emission into the same direction. This is the most obvious consequence of the highly correlated motion of both electrons which are subject to their mutual Coulomb repulsion and shows very clearly in the experimental data. (iii) The above-mentioned reflection symmetry with respect to the x-z plane is nicely reproduced in the experimental data within their error bars.

Before a quantitative comparison between experimental data and theoretical predictions can be made, two comments are necessary. First, the experimental data are burdened by the influences of the large solid angles accepted by the electron spectrometers while the theoretical data are valid for pointlike detectors. The solid angle essentially causes a smearing out of otherwise sharp structures. Since within the error bars the zeros in the TDCS are reproduced well, the extremely cumbersome solid-angle correction has been omitted. Second, the experimental data refer to incident light with specific polarization properties. In the present situation the TDCS can be written as the incoherent sum of two contributions which refer to linear polarization along the x and y axis. One has

$$\left(\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE}\right)_{\text{obs}} = \frac{1+S_1}{2}\left(\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE}\right)_{\text{lin.pol.}x} + \frac{1-S_1}{2}\left(\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE}\right)_{\text{lin.pol.}y}.$$

Since S_1 is known and the TDCS can be calculated for the two states of linear polarization, this formula is used for comparing the theoretical predictions with the experimental data. In Fig. 2, the overall intensities of the theoretical and experimental TDCS are adapted to each other by a least-squares fitting procedure for optimum agreement.

Three theoretical curves are included in Fig. 2, and for two of them good agreement with the experiment can be noted (results from Ref. [2] have been omitted because the model leads in the geometry considered to a TDCS whose intensity is concentrated in the half plane where the first electron is emitted, which is in clear contradiction to our experimental data). The solid curve is the result of a direct calculation of the TDCS using correlated wave functions in the initial and final state [3], namely, a three-parameter Hylleraas function [10] for the ground state and for the final state a Garibotti-Miraglia wave function [11] which contains on equal footing all three two-particle Coulomb interactions. The curve shown represents the results for an orthogonalized form of the dipole matrix element in which the length and velocity forms are made to become equal. The dotted and dashed curves in Fig. 2 are taken from the extended Wannier theory [1] where the TDCS is factorized into angular terms and a correlation term $C(1,2)$ describing the Coulomb repulsion between both outgoing electrons. In the standard approach the correlation factor is modeled by a Gaussian function in the relative angle Θ_{12} between both electrons peaked at $\Theta_{12}=180°$ with FWHM $=\Theta_0 \times E_{\text{exc}}^{1/4}$. Three predictions are available for the case considered: $\Theta=103$ deg$\text{eV}^{-1/4}$ [12] and $\Theta_0=91$ deg$\text{eV}^{-1/4}$ [13,14] for the ion charge $Z=2$, and a value $\Theta_0=67$ deg$\text{eV}^{-1/4}$ [15,16] independent of Z which follows from an expansion of the correlation factor (for deviations from the Gaussian shape see Refs. [15–19]). It is an open question whether the extended Wannier theory is valid for the present case of rather high excess energy. Nevertheless the prediction from this model shall be compared also with the experimental data. The dotted curve in Fig. 2 follows from this model using $\Theta_0=91$ deg$\text{eV}^{-1/4}$. It can be seen that the experimental data are not reproduced (the failure of the model for broad Gaussian functions and angles around $\Theta_{12}=0°$ is known [15]). However, if a least-squares fit of the TDCS to the experimental data is performed using Θ_0 in the correlation function and the overall size of the TDCS as free parameters, the dashed curve in Fig. 2 is obtained. It also agrees well with the experimental data, and from the fit one gets $\Theta_0=43(3)$ deg$\text{eV}^{-1/4}$. This value is remarkably smaller than the ones cited above (in this context it should be noted that the first data for double photoioniza-

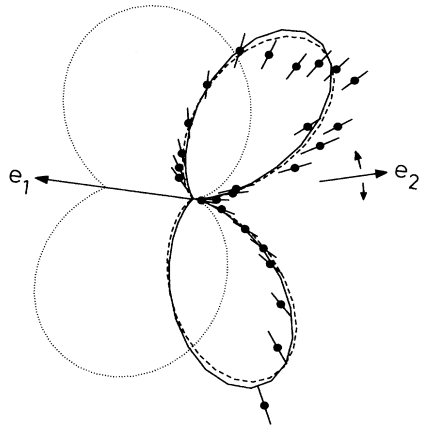

FIG. 2. Relative values of experimental and theoretical TDCS for the double photoionization of helium at $h\nu=99$ eV in the plane perpendicular to the photon beam; direction of electron e_1 fixed; electron e_2 at different angles. For further details see text.

tion in xenon leading to the ^1S final ionic state also show a strong deviation from the standard model [5,6]).

The last result for the correlation factor suggests a separate and more detailed discussion of this significant quantity. For this purpose a factorization of the TDCS into angular terms following from an independent-particle approach and the correlation term $C(1,2)$ is needed. One obtains in the present case

$$\left(\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE}\right)_{\text{lin.pol.}x} \propto (\cos\alpha - 1)^2 C(1,2)$$

and

$$\left(\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE}\right)_{\text{lin.pol.}y} \propto (\sin\alpha)^2 C(1,2)\,,$$

with α being the angle between the acceptance direction of the rotatable electron spectrometer and the x axis. It then becomes possible to evaluate an effective factor $C(1,2)$ also for the experimental and theoretical data where no factorization is given. The result is shown in Fig. 3 in a logarithmic plot which allows a better display of the individual error bars (it should be noted that the experimental data are burdened with the solid-angle effect which is most apparent in the logarithmic plot for $\Theta_{12} \rightarrow 180°$ and $\Theta_{12} \rightarrow 0°$). As expected, the solid and dashed curves (same designation as in Fig. 2) also show good agreement in their correlation factors. The dash-dotted curve is a correlation factor $C(|\mathbf{k}_1 - \mathbf{k}_2|)$, also named the Sommerfeld factor, which depends only on the known wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ and an overall scaling factor from a weighted least-squares fit to the experimental data ($C(|\mathbf{k}_1 - \mathbf{k}_2|) = \pi/\{k_{12}[\exp(\pi/k_{12}) - 1]\}$ with $k_{12} = 0.5|\mathbf{k}_1 - \mathbf{k}_2|$; Refs. [3,20]). It can be seen that the use of such a correlation factor is not capable of reproducing the observed experimental data or the theoretical results which include correlations in the initial and final state. This becomes even more pronounced if all correlation factors in Fig. 3 are normalized to a common maximum value.

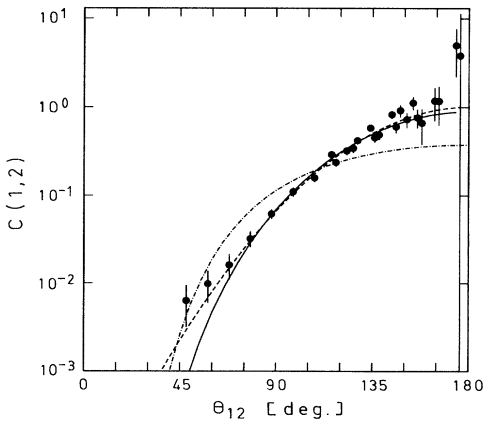

FIG. 3. Relative values of experimental and theoretical correlation factors $C(1,2)$ for the double photoionization of helium at $h\nu = 99$ eV; for details see text.

With the experimental data presented it has been possible for the first time to demonstrate explicitly the effects of correlated motion of two outgoing electrons in the simplest and clearest case which is direct double photoionization in helium. Remarkable agreement has been found with theoretical results which take into account electron correlations in the initial state and all three terms of the mutual Coulomb interactions in the final state and which have been completed long before this experimental study became feasible. Also, good agreement is found if the parameter Θ_0 is adapted in the extended Wannier theory. Since this result comes from just one measured TDCS, nothing can be said about a more general validity. The experiment opens a broad field of further studies on helium to elucidate the general Coulombic three-particle problem, e.g., by changing the angle settings, the excess energy, and/or the energy sharing between both electrons. These results will then provide a sound foundation for the investigation of other systems as, e.g., double photoionization in the outer *np* shells of the other rare gases where different fine-structure components exist for the ion as well as for the two-electron wave function in the continuum and where disturbances by the process of satellite excitation and valence Auger decay might occur.

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