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Energy- and angle-resolved ($\gamma, 2e$) experiments with synchrotron radiation: helium and argon

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Abstract. Typical properties of the triple differential cross section (TDCS) are discussed on the basis of experimental results for helium and argon. The data for helium at excess energies of 20 eV and 10 eV and for equal energy sharing of the two emitted electrons agree well with the theoretical predictions of an *ab initio* calculation. The results for argon, 20 eV excess energy and equal energy sharing, allow a comparison of the TDCS for the $3p^4\ ^1D^e$ and $3p^4\ ^3P^e$ final ionic states. For the $^3P^e$ state, a tentative interpretation of the data is given within the model of the extended Wannier theory for the relative strength of the two contributing electron-pair wave functions in the continuum, $^3P^o$ and $^3D^o$.

1. Introduction

The triple differential cross section (TDCS) obtained in ($\gamma, 2e$) experiments through energy- and angle-resolved detection of the two emitted electrons in coincidence provides – except for the spin projections – complete information in photon-induced double ionisation. Due to the differential nature of the TDCS, inherent dynamical properties of the double photoionisation process are expected to show up most clearly, thereby providing a test case for theoretical treatments of the three-particle-breakup, in particular as a probe for the mutual two-particle Coulomb interactions of the three charged particles.

Contrary to ($e, 2e$) experiments, which have been established since many years with great success, energy- and angle-resolved ($\gamma, 2e$) experiments are still at their beginnings. This can be attributed to the much lower double photoionisation cross section as compared to single ionisation by electron impact as well as to severe time restrictions imposed on experiments performed at a synchrotron radiation facility. Hence, the first study of energy- and angle-resolved double photoionisation was performed using HeII light [1].

In this paper, we present new results for the energy- and angle-resolved double photoionisation in helium and argon which go beyond those published recently [2],[3]. Both experiments were performed at the electron storage ring BESSY/Berlin, at the TGM4 beamline. The method of

angle-resolved electron spectrometry was applied to measure the two ejected electrons in coincidence in two separate spectrometers positioned in a plane perpendicular to the photon beam direction. One spectrometer was mounted at a preselected position, while the other could be turned around the direction of the photon beam defining the z -axis. Equal energy sharing of the excess energy between both electrons ($E_1 = E_2 = E_{exc}/2$) was chosen because then the dependence of the TDCS on the circular polarisation vanishes [4]. For more details on the experimental setup see [2],[3].

Figure 1. Relative values of triple differential cross sections for double photoionisation of helium in the plane perpendicular to the photon beam; experiment: points with errorbars, theory ([5], velocity form): full line. Data are shown for different photon energies and angle settings of the first analyser (data in figure 1 (a) are from ref. [2]). The x -axis indicates the major axis of the polarisation ellipse, the direction of electron e_1 is given by the arrow. The scaling uncertainty in the normalisation factor of the individual measurements is approximately $\pm 20\%$.

2. Helium

The results of the TDCS in helium measured at two different photon energies and for two different positions of the fixed analyser are shown in figure 1 as polar plots. The data are normalised for equal target density and photon flux, and the different analyser transmission functions have been taken into account, thereby allowing a comparison of relative intensities. It should be noted that the data shown in figure 1 are burdened with the large acceptance angles of the spectrometers used which tend to smear out sharp angle-dependent structures in the TDCS. However, this solid-angle effect has been found to be small in the case of helium, at least as far as the shape of the pattern is concerned. This can already be inferred from the pronounced minima for opposite emission where the solid-angle effect is expected to show up most strongly [3]. Therefore, no correction has been applied to the data. Several characteristic features can be noted: (i) Practically no intensity is observed for two-electron emission in opposite directions. This is a consequence of the unfavoured character of the two-electron wave function in the continuum. (ii) Practically no intensity is observed for two-electron emission into the same direction. This reflects directly the highly correlated motion of both electrons which are subject to their mutual Coulomb repulsion.

(iii) The expected reflection symmetry with respect to the plane formed by the major axis of the polarisation ellipse (x -axis) and the photon beam (z -axis) is nicely reproduced in the data of figure 1 (a) within the errorbars. This symmetry no longer holds for the emission of the first electron in a direction different from the x -axis, as shown in figures 1 (b) and (c). This results from the competing influence of two contributions: the electron-photon interaction which refers to the direction of the electric field vector of the incident light, and the final state Coulomb repulsion which refers to the direction of the preselected electron.

Also shown in figure 1 are the theoretical predictions of an *ab initio* calculation [5]. The theoretical data have been adapted to the experimental ones by a least-square fitting procedure. The calculation is based on correlated wave functions in the initial and final states, namely a three-parameter Hylleraas function [6] for the ground state and a Garibotti–Miraglia wave function [7] for the two electrons in the continuum (velocity form results are shown, the length form results are rather similar). The agreement between theoretical and experimental values is remarkable. It is also possible to model the experimental data within the extended Wannier theory [8], using, as in ref. [2], the characteristic quantity Θ_0 (see below) and an overall scaling factor as free parameters. This procedure leads to $\Theta_0 = 43(2)^\circ(\text{eV})^{-1/4}$ for $E_{exc} = 20 \text{ eV}$ and $\Theta_0 = 49(3)^\circ(\text{eV})^{-1/4}$ for $E_{exc} = 10 \text{ eV}$, which might be an indication for an energy dependence of this parameter.

Taking a look at the relative strengths of the different patterns, a comparison of the normalised data of figure 1 leads to the following (note that the experimental relative strength may be affected by the large acceptance angles whereas the theoretical values are computed for a point-like detector): the $\text{TDCS}(E_{exc} = 20 \text{ eV}, \phi_1 = -76^\circ)$ in figure 1 (b) is approximately twice as intense as the $\text{TDCS}(E_{exc} = 20 \text{ eV}, \phi_1 = 180^\circ)$ in figure 1 (a), note the scaling factor 2. This is in good agreement with the factor of about 2 obtained from the ratio of the maxima in the calculations of ref. [5] (velocity and length form results differ only slightly). However, comparing figure 1 (b) to figure 1 (c), one deduces from experiment that $\text{TDCS}(E_{exc} = 20 \text{ eV}, \phi_1 = -76^\circ)$ and $\text{TDCS}(E_{exc} = 10 \text{ eV}, \phi_1 = -76^\circ)$ are of similar magnitude, whilst from the theory ratios of about 5:1 (velocity form) and 4:1 (length form) respectively are expected. This point needs further clarification.

3. Argon

Contrary to Helium, three different final ionic states can be reached in the double photoionisation in the outer p-shell of argon: $^3\text{P}^e$, $^1\text{D}^e$ and $^1\text{S}^e$. Figure 2 shows the normalised TDCS for the $^3\text{P}^e$ and $^1\text{D}^e$ final states (a first attempt to measure also the $^1\text{S}^e$ state failed due to the small intensity and the associated unacceptably large errorbars). Again, several characteristic features can be noted: (i) Within the errorbars, each TDCS exhibits the expected reflection symmetry with respect to the x -axis. (ii) The small intensity of the $^3\text{P}^e$ pattern as compared to the $^1\text{D}^e$ case is striking, because it is just opposite to the behaviour of the total partial cross sections at threshold, where $\sigma_{tot}(^3\text{P}^e)$ is expected to be stronger than $\sigma_{tot}(^1\text{D}^e)$ on grounds of a propensity rule [8] and experimental data [9],[10]. (iii) The pattern for $^3\text{P}^e$ shows a maximum for the emission of both electrons in opposite directions. This reflects the favoured character of the $^3\text{P}^o$ two-electron wave function involved and is in contrast to the $^1\text{D}^e$ case which, like the double photoionisation in helium, has an unfavoured character. (iv) Due to the unfavoured character, the $^1\text{D}^e$ pattern

Figure 2. Relative values of experimental TDCs for the double photoionisation of argon leading to $^3P^e$ (a) and $^1D^e$ (b) final ionic states for $h\nu = 64$ eV.

in argon is expected to have zero intensity for emission of the two electrons in opposite direction (cf. the helium case in section 2). However, the TDCS for elements others than helium has to be regarded as a coherent sum of the transition amplitudes for direct double ionisation and indirect pathways via autoionising intermediate states (resonance affected double ionisation [3]). In the present case, the remaining intensity for emission in opposite directions might be due to such a resonance affected process, but also to the influence of solid-angle effects (for details see ref. [3]).

Figure 3. Photoelectron spectrum of argon at $h\nu = 64$ eV. The spectrum was taken under the quasi-magic angle and has been corrected for the analyser dispersion. The kinetic energy corresponding to $E_{kin} = \frac{1}{2}E_{exc}(^3P^e)$ is marked by the dotted line and labeled S.

Figure 4. Fit of figure 2 (a) using the extended Wannier model [8] with $\Theta_0 = 43^\circ (\text{eV})^{-1/4}$ kept constant (a) and Θ_0 left variable (b). For details see text.

In the following, we shall concentrate on the $^3\text{P}^e$ case, for the $^1\text{D}^e$ case we refer to the detailed discussion in ref. [3]. The photoelectron spectrum shown in figure 3 suggests that for the $^3\text{P}^e$ case at a photon energy of 64 eV the possible contribution of an indirect process should be rather small because no specific structure occurs at $E_{kin} = \frac{1}{2}E_{exc}(^3\text{P}^e)$, i.e. equal energy sharing between the two electrons. Assuming the TDCS to be dominated by direct double photoionisation, an attempt has been made to interpret the data within the model of the extended Wannier theory [8]. Due to dipole selection rules, only two states ($^3\text{P}^o$ and $^3\text{D}^o$) are allowed for the two-electron wave function in the continuum, each having a specific angular structure and amplitude, but a common correlation factor $C(1,2)$. $C(1,2)$ can be approximated by a Gaussian in the relative angle Θ_{12} between both electrons peaked at $\Theta_{12} = 180^\circ$ and with a width $\Theta_{fwhm} = \Theta_0 E_{exc}^{1/4}$. The TDCS can be written as

$$\begin{aligned} \text{TDCS}|_{lin.pol.x} \propto & \sum_{M_i, M_i} \left| \langle 1 M_i 1 M | 10 \rangle a_P A_{1M}(\theta_1, \phi_1, \theta_2, \phi_2) \right. \\ & \left. + \langle 1 M_i 2 M | 10 \rangle a_D A_{2M}(\theta_1, \phi_1, \theta_2, \phi_2) \right|^2 C(1,2). \end{aligned}$$

The angular functions A_{1M} and A_{2M} are given in explicit form in ref. [8]. The index *lin.pol.x* stands for complete linear polarisation of the light in *x*-direction (a similar expression holds for $\text{TDCS}|_{lin.pol.y}$, and the observed TDCS can easily be expressed in terms of $\text{TDCS}|_{lin.pol.x}$ and $\text{TDCS}|_{lin.pol.y}$ [2],[3]). The above formula leads to a parametrisation of the TDCS with four parameters which can be determined by a least-square fitting procedure: an overall scaling factor, the ratio $\rho = |a_D|/|a_P|$, the relative phase $\alpha = \arg(a_D/a_P)$ and Θ_0 . Two possible fits are shown in

figure 4: one (figure 4 (a)) with a fixed value of $\Theta_0 = 43^\circ (\text{eV})^{-1/4}$ which was found for this excess energy in helium, the second (figure 4 (b)) with Θ_0 left variable, leading to $\Theta_0 = 66(10)^\circ (\text{eV})^{-1/4}$. The individual shapes of the two curves differ, however, given the scatter in the experimental data, the fit results are of the same quality. For the parameter ρ one gets in the case of figure 4 (a) $\rho=4.4(5)$ which is twice as much as the value of $\rho=2.2(5)$ obtained for figure 4 (b). This behaviour can be understood because if Θ_0 decreases, so does $C(1,2)$ for small relative angles Θ_{12} . This decrease is then partly compensated by an increase in the ratio ρ (A_{2M} dominates A_{1M} for small relative angles). Surprisingly, both fits lead to very similar results for the parameter α ($124(5)^\circ$ and $115(5)^\circ$ for the fits in figure 4 (a) and 4 (b) respectively), thus indicating a partially destructive interference of the two amplitudes. In summary, figure 4 suggests that for $E_{exc}=20.6$ eV, the major contribution to the TDCS comes from the $^3D^\circ$ state. This result – though obtained for experimental data which still need a considerable improvement in the errorbars – differs from the only available other experiment [1], performed on krypton at an excess energy $E_{exc}=2.26$ eV where an excellent description of the data was obtained considering the $^3P^\circ$ state alone ($\rho=0$). The difference might first of all be attributed to the individual behaviour of the rare gases, but different excess energies and different strengths of resonance affected double ionisation might also play a role.

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