LETTER TO THE EDITOR

A procedure to extract the complex amplitudes of He photodouble ionization from experimental data

P Bolognesi[1], A S Kheifets[2], I Bray[3], L Malegat[4], P Selles[4], A K Kazansky[5] and L Avaldi[1,6]

[1] CNR-IMIP, Area della Ricerca di Roma, CP10, 00016 Monterotondo Scalo, Italy
[2] Research School of Physical Sciences and Engineering, Australian National University, Canberra, ACT 2000, Australia
[3] CAMSP, School of Mathematical and Physical Sciences, Murdoch University, Murdoch, Perth 6150, Australia
[4] LIXAM, UMR 8624 du CNRS, Bâtiment 350, Université Paris-Sud, 91405 Orsay-Cedex, France
[5] Fock Institute of Physics, The University of St Petersburg, St Petersburg 198504, Russia
[6] INFM-TASC, Gas Phase Beamline at Elettra, Area Science Park, I-34012 Trieste, Italy

Abstract
A procedure to extract the two complex amplitudes that govern the He photodouble ionization process from the experimental data is proposed. The results are compared with the predictions of the convergent close coupling and hyperspherical R-matrix with semiclassical outgoing wave theories.

Photodouble ionization (PDI) of He is the archetypal three-body Coulomb problem (two interacting particles in the field of a positive *structureless* residual ion) and as such it has attracted a lot of experimental and theoretical interest. The complete experimental characterization of this process implies experiments in which either the two photoelectrons [1, 2] or one of the photoelectrons and the residual ion [3] are detected in coincidence. In such experiments the triply differential cross section (TDCS) $\mathrm{d}^3\sigma/\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2\,\mathrm{d}E_1$, i.e. a cross section differential in the angles of emission of the two photoelectrons $\Omega_1 = (\vartheta_1, \varphi_1)$ and $\Omega_2 = (\vartheta_2, \varphi_2)$ and in one kinetic energy E_1, is measured. The kinetic energy E_2 of the other electron is determined by energy conservation. Since the first experimental determination of the TDCS in 1993 [1] a large set of data from the near-threshold [2] region up to a photon energy of about 500 eV [4] has been published. At pace with the experimental work extensive theoretical effort has been applied [5–10]. Experimentally, at a fixed incident energy the TDCS can be measured under a variety of kinematic conditions (changing the energy sharing between the electrons and/or their angles of emission). This results in a large body of data and hampers direct comparison between different sets of data and between experiment and theory. In this paper we propose a procedure to overcome this difficulty by extracting from the experimental data the underlying basic quantities which allow one to reconstruct any

particular TDCS, namely the moduli and the relative phase of the gerade and ungerade complex amplitudes of the process.

By considering the $^1P^o$ symmetry of the electron pair continuum wavefunction, the TDCS can indeed be written in a way that allows the full separation of the geometrical factors and the dynamic parameters [11, 12]. In the case of incident radiation that propagates along the z axis and is fully linearly polarized along the x axis the TDCS measured in the (x, y) plane can be written

$$\text{TDCS}(E_1, E_2, \theta_{12}) = |A_g(E_1, E_2, \theta_{12})(\cos\vartheta_1 + \cos\vartheta_2) + A_u(E_1, E_2, \theta_{12})(\cos\vartheta_1 - \cos\vartheta_2)|^2 \tag{1}$$

where ϑ_1 and ϑ_2 are the angles of emission of the two photoelectrons in the (x, y) plane, measured from 0 to 2π with respect to the polarization axis x, and θ_{12} is the relative angle between the directions of emission of the two photoelectrons. The complex amplitudes A_g and A_u are respectively symmetric (gerade) and antisymmetric (ungerade) with respect to the exchange of the two electrons—which amounts to the exchange of E_1 and E_2 since $\theta_{12} = \theta_{21}$. The θ_{12}, E_1 and E_2 dependence of the amplitudes includes all the physical information on the dynamics of the process, i.e. the effects of the electron–electron and electron–residual ion interactions. It is clear from (1) that the TDCS is invariant upon multiplication of the two amplitudes by the same phase factor. Hence the only quantities of physical significance are the moduli of A_g and A_u and their relative phase δ. Since the theoretical models directly calculate these quantities it is of considerable interest to look for a procedure which extracts them from the experimental measurements.

Recently such a procedure, based on internormalized data sets at four *specific* experimental geometries, was proposed by Krässig [13]. Here we propose a different procedure that allows one to extract $|A_g|$, $|A_u|$ and $\cos\delta$ from *any* set of three *absolute* experimental determinations of the TDCS at the same E_1, E_2 and θ_{12} values but at different θ_1. The same procedure allows one to extract the scaled complex amplitudes $|a_g| = C|A_g|$, $|a_u| = C|A_u|$ and $\cos\delta$ from *any* set of three *relative internormalized* experimental determinations of the TDCS, where C is a real positive constant independent of E_1, E_2 and θ_{12}.

Thus, for each experimental determination $i = 1, 2, 3$, let us denote by σ_i the measured quantity, be it the absolute TDCS or the coincidence yield that is proportional to the latter, and by

$$\alpha_i = \cos\vartheta_{i1} + \cos\vartheta_{i2} \tag{2a}$$

$$\beta_i = \cos\vartheta_{i1} - \cos\vartheta_{i2} \tag{2b}$$

the geometric factors. The measured quantities can then be expressed by expanding (1) as

$$\begin{pmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_3 \end{pmatrix} = \begin{pmatrix} \alpha_1^2 & 2\alpha_1\beta_1 & \beta_1^2 \\ \alpha_2^2 & 2\alpha_2\beta_2 & \beta_2^2 \\ \alpha_3^2 & 2\alpha_3\beta_3 & \beta_3^2 \end{pmatrix} \begin{pmatrix} |a_g|^2 \\ |a_g||a_u|\cos\delta \\ |a_u|^2 \end{pmatrix} \tag{3}$$

or in matrix notation $\vec{\sigma} = A\vec{x}$. The solution to this equation reads $\vec{x} = A^{-1}\vec{\sigma}$. The elements of the covariance matrix associated with the calculated quantities are then given by

$$\langle \Delta x_i \Delta x_j \rangle = \sum_{k,l=1}^{3} (A^{-1})_{ik}(A^{-1})_{jl}\langle \Delta\sigma_k \Delta\sigma_l \rangle. \tag{4}$$

Assuming that the uncertainties in the quantities σ_i are statistically independent, i.e. $\langle \Delta\sigma_k \Delta\sigma_l \rangle = \delta_{kl}\langle (\Delta\sigma_k)^2 \rangle$, we get

$$\langle (\Delta x_i)^2 \rangle = \sum_{k=1}^{3} (A^{-1})_{ik}\langle (\Delta\sigma_k)^2 \rangle((A^{-1})^t)_{ki} \tag{5a}$$

$$\langle \Delta x_i \Delta x_j \rangle = \sum_{k=1}^{3} (\boldsymbol{A}^{-1})_{ik} \langle (\Delta \sigma_k)^2 \rangle ((\boldsymbol{A}^{-1})^t)_{kj}. \tag{5b}$$

The uncertainty in $\cos \delta$ is then easily obtained from the elements of the covariance matrix as

$$\langle (\Delta(\cos \delta))^2 \rangle = \left\langle \left(\Delta \frac{x_2}{\sqrt{x_1 x_3}} \right)^2 \right\rangle = \frac{x_2^2}{4x_1^3 x_3} \langle (\Delta x_1)^2 \rangle + \frac{1}{x_1 x_3} \langle (\Delta x_2)^2 \rangle + \frac{x_2^2}{4x_3^3 x_1} \langle (\Delta x_3)^2 \rangle$$
$$- \frac{x_2}{x_1^2 x_3} \langle \Delta x_1 \Delta x_2 \rangle + \frac{x_2^2}{2x_1^2 x_3^2} \langle \Delta x_1 \Delta x_3 \rangle - \frac{x_2}{x_3^2 x_1} \langle \Delta x_3 \Delta x_2 \rangle. \tag{6}$$

The technique outlined above can be used only if certain dynamic and geometric conditions are fulfilled, so that the $\boldsymbol{A}$ matrix can be inverted to yield non-zero values of the components of $\vec{x}$.

It is worthwhile mentioning that the procedure outlined above only enables one to determine the cosine of the relative phase between the two complex amplitudes. Thus the experiment cannot be considered a complete one, because the sign of the phase remains undetermined. Therefore, not all the observables of the double photoionization process can be determined from (3). For example, the sign of the circular dichroism in experiments with radiation including a circularly polarized component [14] cannot be obtained. However, this procedure can easily be turned into a complete one. For instance, one could consider absolute measurements of the TDCS obtained using elliptically polarized light: the absolute magnitudes and relative phase (i.e. the cosine *and* sine of this phase) of the two amplitudes would be obtained from a quadruplet of such measurements performed for the same energies and relative angle of the two electrons. The extraction procedure would then imply solving a linear system of four equations in the four variables $|a_g|^2$, $|a_u|^2$, $|a_g||a_u| \cos \delta$ and $|a_g||a_u| \sin \delta$.

The procedure also assumes that the experimental uncertainty in θ_{12} is negligible compared to the uncertainties $\{\langle (\Delta \sigma_i)^2 \rangle\}$ of the measured coincidence yield. This means that the application of the method to experimental TDCS resulting from an integration over large $\Delta\vartheta$ and $\Delta\varphi$ intervals is not straightforward.

In this respect, our procedure is complementary to the one previously proposed by Krässig [13]. That procedure has the advantage of making use of the full set of coincidence events that can be obtained by a COLTRIMS complete set of data. However, the four specific experimental geometries needed are not easily achieved by most of the experimental apparatuses used in PDI experiments. In contrast, the simpler procedure used here can be applied with no restrictions to any set of experimental data.

The procedure has been applied to the TDCS of He measured on a relative scale at an excess energy $E = E_1 + E_2 = 40$ eV above threshold in unequal energy sharing ($E_1 = 5$ eV and $E_2 = 35$ eV) by Bolognesi *et al* [15]. This data set is particularly suited for the proposed procedure because three angular distributions have been measured simultaneously. In the experiment one electron was detected at $\vartheta_{1i} = 0°$, $30°$ and $60°$ for $i = 1$, 2 and 3, respectively, while the second one was collected at $105° \leqslant \vartheta_{2i} \leqslant 325°$. Moreover, complementary TDCS patterns, obtained in two experiments by the interchange of the electron kinetic energies, $E_1 \leftrightarrow E_2$, have been measured. Due to the symmetry properties of a_g and a_u the only difference between the solutions of (3) based on the two complementary sets should be a difference of π in the respective relative phases of the gerade and ungerade amplitudes. Comparison of these two sets of solutions will therefore illustrate the reliability of the method. To conclude, a final advantage of this set of measurements is that the $\pm 3°$ [15] angular acceptance in the dispersion plane of the spectrometers makes plausible the assumption of a negligible uncertainty in the determination of the angles.

The $|a_g|^2$ and $|a_u|^2$ squared amplitudes, their ratio and the cosine of their relative phase obtained applying the procedure to the experimental data are shown in figures 1–4, respectively.
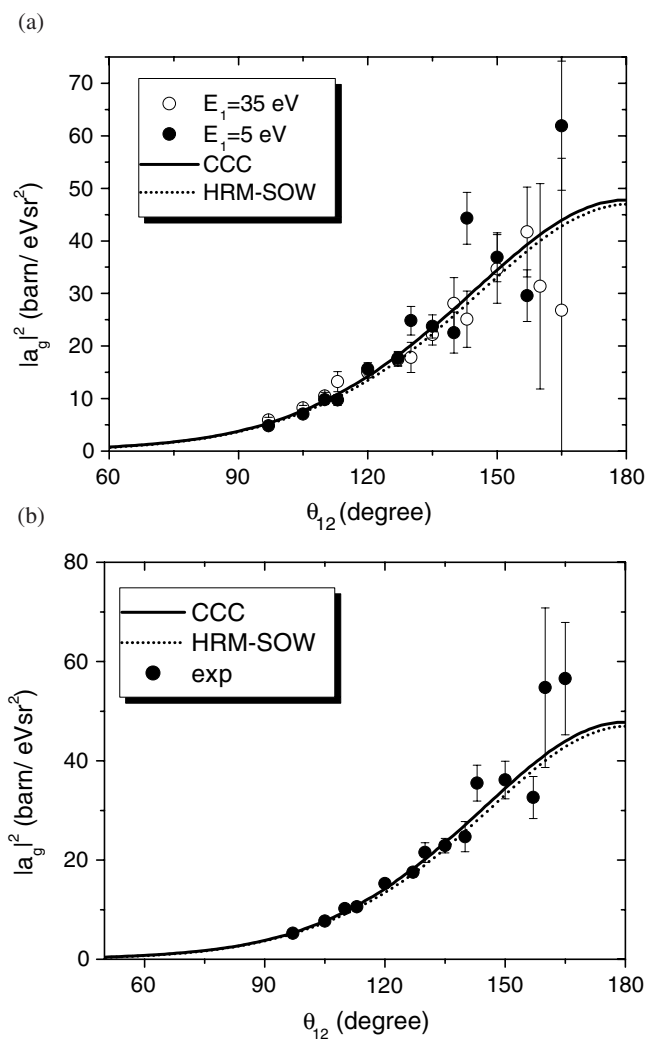
(a)

(b)

Figure 1. $|a_g|^2$ obtained from the two experimental TDCS at $E = 40$ eV, $E_1 = 35$ eV and $E_2 = 5$ eV (open circles), and $E_1 = 5$ eV and $E_2 = 35$ eV (black circles) (a) and from the weighted average of the two experiments (b) versus the relative angle θ_{12}. The experimental results are compared with the predictions of the CCC (full curve) and HRM-SOW (dotted line) theories.

In figure 1(a) $|a_g|^2$ obtained using both sets of measurements in the complementary kinematics are shown. At the θ_{12} values, where multiple experimental determinations exist, a weighted average has been calculated and reported in the figure. The two sets of results are in agreement within the respective uncertainties, thus in figure 1(b) as well as in all the other figures only the weighted average of the two sets is reported. In the same figures the results are compared with the predictions of the convergent close coupling (CCC) [7] and the hyperspherical R-matrix with semiclassical outgoing (HRM-SOW) [8] methods. For a better comparison of the experimental and theoretical shape of the $|a_g|^2$ and $|a_u|^2$ squared amplitudes, the experimental $|a_g|^2$ has been rescaled to the mean value of the two theories at $\theta_{12} = 135°$ in figure 1. The same scaling factor has been used in figure 2 for the $|a_u|^2$ squared amplitude. The absolute
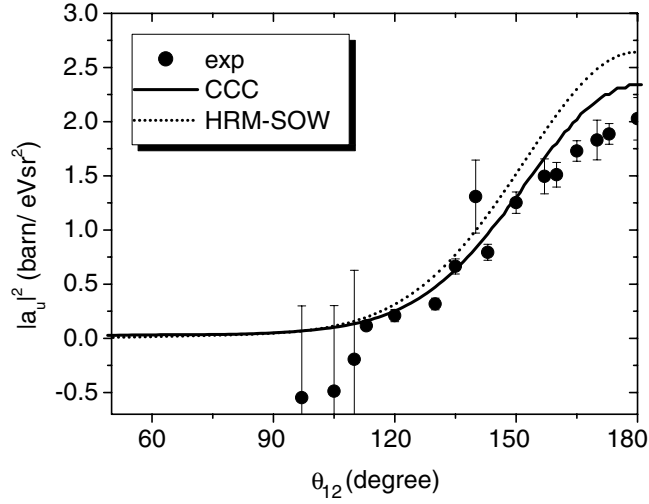
Figure 2. $|a_u|^2$ versus the relative angle θ_{12}. The same symbols as in figure 1(b) are used.

Table 1. Absolute values of $|a_g|^2$, $|a_u|^2$, $\cos\delta$ at $\theta_{12} = 180°$ and the maximum values of $|a_g|^2/|a_u|^2$ predicted by the CCC [7] and HRM-SOW [8] methods.

	CCC	HRM-SOW
$|a_g|^2@\theta_{12} = 180$ (barn eV^{-1}sr^{-2})	47.748	46.972
$|a_u|^2@\theta_{12} = 180$ (barn eV^{-1}sr^{-2})	2.341	2.644
$|a_g|^2/|a_u|^2$ max	78.00	77.98
$\theta_{12}@|a_g|^2/|a_u|^2$ max (deg)	99	92
$\cos\delta@\theta_{12} = 180$	-0.5236	-0.5056

values of the complex amplitudes and $\cos\delta$ predicted by the theories at $\theta_{12} = 180°$, together with the maximum values of $|a_g|^2/|a_u|^2$ are presented in table 1.

Despite the good statistical quality of the experimental data (see figures 1, 2 in [15]) the results for the $|a_g|^2/|a_u|^2$ ratio and $\cos\delta$ display some scattering. Moreover, the error propagation results in large error bars, especially in the region near $\theta_{12} = 180°$ for $|a_g|^2$ and $\theta_{12} = 90°$ for $|a_u|^2$.

As for $|a_g|^2$ the two theories predict the same shape in good agreement with experiment. The values of $|a_u|^2$ near $\theta_{12} = 180°$ predicted by both theories overestimate the experimental results, with the CCC value being closer to experiment than the HRM-SOW one. From figures 1 and 2, the half widths at half maximum of $|a_g|^2$ and $|a_u|^2$ can be estimated roughly as 87° and 66°, respectively. This indicates a steeper decrease of the ungerade squared amplitude from its maximum, compared to the gerade one, resulting in a maximum of the $|a_g|^2/|a_u|^2$ ratio located away from 180° as one can see from figure 3. A small difference in the shape and the position of the maximum of the ratio is observed in the predictions of the two theories. Unfortunately the quality of the data in the region $100° < \theta_{12} < 120°$ and the absence of experimental determinations in the region below $\theta_{12} = 100°$ does not allow one to distinguish between the two predictions. Figure 4 shows that the relative phase is varying with θ_{12}, although the variation in the experiments seems to be less pronounced than in the theoretical predictions.
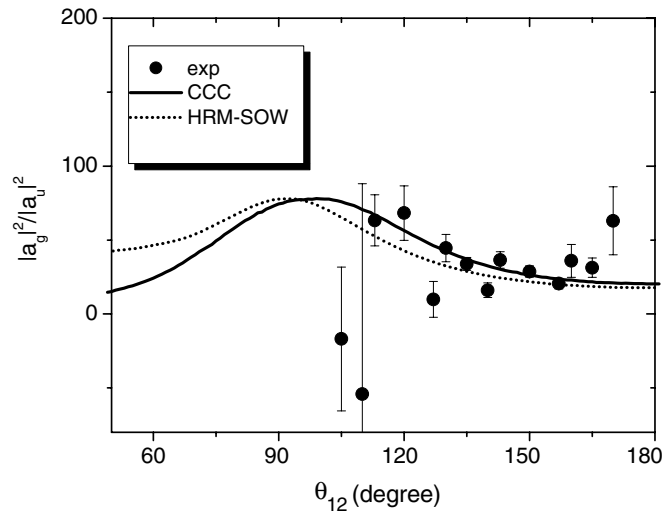
Figure 3. The $|a_g|^2/|a_u|^2$ ratio versus the relative angle θ_{12}. The same symbols are used as presented in figure 1(b).
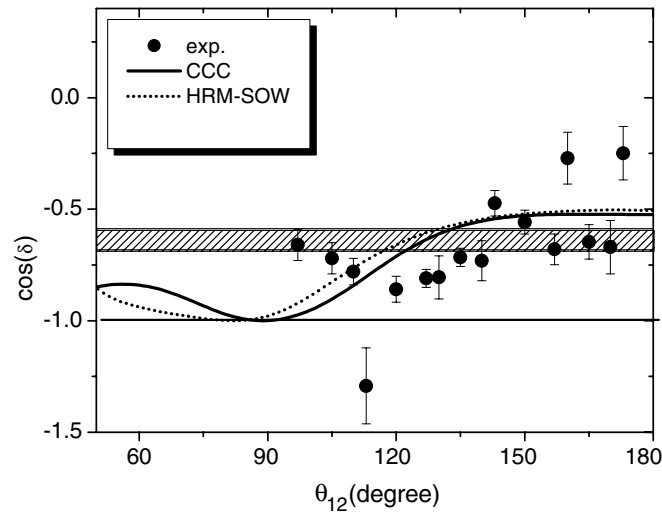

Figure 4. $\cos(\delta)$ versus the relative angle θ. The same symbols as in figure 1(b) are used. The stripe shows the value obtained by Bolognesi *et al* [15] in the framework of the practical parametrization proposed by Cvejanovic and Reddish [16].

Thus the constant phase proposed within the framework of the *practical parametrization* by Cvejanovic and Reddish [16] might be too crude an approximation. The dashed stripe in figure 4 shows the band of uncertainty of the *constant* value obtained in a previous representation of the same data by Bolognesi *et al* [15] using the *practical parametrization* [16]. This value is quite close to the average (-0.68 ± 0.27) of the values obtained in the present analysis. The agreement between the theoretical values and experimental ones is reasonable for $\theta_{12} > 120°$. Theories then predict a phase difference close to $180°$ at $\theta_{12} < 100°$. A smaller value is determined by experiment at $\theta_{12} = 100°$, while no experimental data are available in the region below $100°$ to prove or disprove the predicted trend.

In summary we have proposed a procedure to extract from the experimental data the two complex amplitudes of PDI. This direct procedure does not rely upon any assumption regarding the functional dependence of these amplitudes in E_1, E_2, or θ_{12}. The use of the amplitudes makes the comparison among the various experiments as well as between experiment and theory more compact and direct, and allows one to outline differences that may be hidden in the comparison of the TDCS because of the modulation imposed by the kinematic factors. The results for an experiment performed in complementary kinematics at $E = 40$ eV prove that the method is feasible provided the measured TDCS are characterized by a high statistical accuracy. Moreover, this first application shows that direct comparison of the amplitudes predicted by the different theories may be valuable by itself. For example, some differences have been observed between the predictions of the two theories used for $\theta_{12} < 90°$ for the $|a_{\mathrm{g}}|^2/|a_{\mathrm{u}}|^2$ ratio and $\cos \delta$, thus suggesting that future measurements should not neglect this region.

References

[1] Schwarzkopft O, Krässig B, Elminger J and Schmidt V 1993 *Phys. Rev. Lett.* **70** 3008
[2] Huetz A and Mazeau J 2000 *Phys. Rev. Lett.* **85** 530
[3] Ullrich J, Moshammer R, Dörner R, Jagutzki O, Mergel V, Schmidt-Böcking H and Spielberg L 1997 *J. Phys. B: At. Mol. Opt. Phys.* **30** 2917
[4] Knapp A *et al* 2002 *Phys. Rev. Lett.* **89** 033004
[5] Maulbetsch F and Briggs J S 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 4095
[6] Pont M and Shakeshaft R 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** L571
[7] Kheifets A S and Bray I 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** L447
    Kheifets A S and Bray I 1998 *Phys. Rev. Lett.* **81** 4588
[8] Malegat L, Selles P and Kazansky A K 2000 *Phys. Rev. Lett.* **85** 4450
    Selles P, Malegat L and Kazansky A K 2002 *Phys. Rev.* A **65** 032711
[9] Pindzola M S and Robicheaux F 1998 *Phys. Rev.* A **57** 318
[10] Rescigno T N, Baertschy M, Isaacs W A and McCurdy C W 1999 *Science* **286** 2474
[11] Briggs J S and Schmidt V 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** R1
[12] Huetz A, Selles P, Waymel D and Mazeau J 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 1917
[13] Krässig B 2001 *Correlations, Polarization and Ionization in Atomic Systems (AIP Conf. Proc. vol 604)* ed D H Madison and M Schulz (Rolla, MO: American Institute of Physics) p 12
[14] Soejima K, Danjo A, Okumo K and Yagishita A 1999 *Phys. Rev. Lett.* **83** 1546
[15] Bolognesi P, Camilloni R, Coreno M, Turri G, Berakdar J, Kheifets A S and Avaldi L 2001 *J. Phys. B: At. Mol. Opt. Phys.* **34** 3193
[16] Cvejanovic S and Reddish T J 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 4691