Photodouble ionization of He with circularly polarised synchrotron radiation: complete experiment and dynamic nodes

P. Bolognesi[1], V. Feyer[1,2], A. Kheifets[3], S. Turchini[4], T. Prosperi[4], N. Zema[4] and L. Avaldi[1,5]

[1]CNR-IMIP, Area della Ricerca di Roma1, 00016 Monterotondo Scalo,Italy
[2] Institute of Electron Physics, National Academy of Sciences, Uzhgorod, Ukraine
[3] Research School of Physical Sciences and Engineering, ANU, Canberra, Australia
[4]CNR-ISM, Area della Ricerca di Roma 2, Via Fosso del Cavaliere, Roma,Italy
[5]CNR-INFM-TASC, Gas Phase Photoemission Beamline@Elettra, Trieste, Italy

Abstract

A set of measurements with linearly and circularly polarised radiation has been performed in order to obtain the moduli and relative phase of the two complex amplitudes which determine the triply differential cross section, TDCS, for the photodouble ionization, PDI, of He. Thus a complete PDI experiment has been done. Then the amplitudes have been used to calculate other observables of the process (linear dichroism, asymmetry parameter of the angular distribution of one of the two photoelectrons and the recoil momentum distribution of the He^{2+} ions). In addition the measurements with circularly polarised radiation undoubtedly prove the existence of the dynamic nodes predicted by some theories in the circular dichroism of PDI of He.

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The understanding of the correlated motion of electron pairs plays a key role in the knowledge of the structure and properties of matter and is of fundamental interest in a number of branches of physics [1]. Photodouble ionization, PDI, of He represents the simplest process where the role of the electron correlations can be studied in detail and in the past ten years a lot of experimental and theoretical interest has been paid to this process [2-4].

The most suited way to study PDI is an experiment where either the two photoelectrons or a photoelectron and the recoiling ion are detected in coincidence. In these experiments the triple differential cross section, $d^3\sigma/d\Omega_1 d\Omega_2 dE_1$ (TDCS) is measured. $\Omega_1=(\vartheta_1,\varphi_1)$ and $\Omega_2=(\vartheta_2,\varphi_2)$ are the angles of emission of the two electrons and E_1 is the energy of one of them. The energy of the second electron is determined by energy conservation $h\nu\text{-}IP^{2+}= E_1+E_2$ where $h\nu$ is the photon energy and IP^{2+} is the double ionization potential. By considering the $^1P^o$ symmetry of the electron pair continuum wavefunction and the invariance by rotation around a preferential symmetry axis, the TDCS can be written in a way that allows the full separation of the geometrical factors and the dynamic parameters [2,4]. While the geometrical factors come from the description of the interaction of the photon with the target in the dipole approximation, the dynamic terms include all the physical information on the dynamics of the process, i.e. the effects of the electron-electron and electron-residual ion interactions. This information is provided by two complex amplitudes $a_g(E_1,E_2, \vartheta_{12})$ and $a_u(E_1,E_2, \vartheta_{12})$ [5], which are respectively symmetric (gerade) and antisymmetric (ungerade) with respect to the exchange of the two electrons. These are the basic quantities that can be calculated by the theories and then used to reconstruct any particular TDCS. Thus the extraction of the moduli and relative phase, δ, of the gerade and ungerade complex amplitudes from the experimental data at a fixed incident energy and energy sharing between the electrons is of considerable interest, because it allows a direct comparison among different sets of data and between theory and experiment. After some attempts relying upon parametrization of a_g and a_u with various degree of complexity and approximation [6,7], Bolognesi *et al.* [8] proposed a procedure that i) does not rely on any approximation, ii) needs only three determinations of the TDCS at the same relative angle ϑ_{12} between the photoelectrons and iii) can be applied to any set of experimental data. Up to now the method has been applied only to measurements with linearly polarized incident radiation. In such a case the $|a_g|^2$, $|a_u|^2$ and cos δ are obtained by solving a set of three non-linear equations. However the sign of the phase δ remains undetermined. If two sets of measurements obtained at the same photon energy and energy sharing with both linearly and circularly polarized radiation are combined then also the sign of δ can be determined and the complete description of the PDI process achieved. Indeed when the incident radiation is circularly polarized the He TDCS displays an helicity dependence, i.e. a non vanishing circular dichroism, CD [9,10]. The observation of chirality in a pair of electrons ejected by a target with a spherically symmetric ground state($^1S^e$), like the He atom, might surprise. Indeed in a common wisdom chirality is a property associated to objects which are not identical to their mirror image. However Berakdar and Klar [9,11] have shown that, due to parity conservation, the helicity of the incident photon is transferred to the three body-system, the He atom, and the continuum spectrum of this excited system depends on the helicity of the absorbed photon. In the initial state the system made by the atom and the photon has a certain parity, carried by the photon. In the final state, after the photon absorption, this parity state is carried by the two electrons, which form a “chiral pair”. The conditions to observe a non vanishing CD are : i) the two electrons unevenly share the excess energy; ii) the direction of the incident light and the directions of the two ejected electrons are not lying in one plane. CD is proportional to sin δ. Thus, according the procedure proposed in [8], the combination of the CD measurement and three TDCS measured with linearly polarized radiation provides the full set of information for a complete experiment . Analogous results can be obtained, as shown by Knapp et al. [12], by applying the procedure proposed by Krässig [13]. That procedure makes use of the full set of coincidence events obtained by a COLTRIMS [14] set of data. However the four specific experimental geometries needed are not achieved by most of the experimental apparatuses used in PDI experiments. By contrast, as stated above, the simpler procedure used here can be applied with no restrictions to any set of experimental data.

A few measurements of the CD are available at 9, 20 60, 100 and 450 eV above the IP^{2+}[15-20]. Only the ones at 100 and 450 eV have been coupled with measurements with linearly polarised radiation to obtain the amplitudes of PDI. Moreover a specific aspect predicted by theory of PDI with circular polarised radiation has not yet been investigated in detail. This is the existence of dynamic nodes of the CD, occurring whenever the ungerade and gerade amplitudes of the TDCS have a phase difference δ = 0 or π [10]. They have been predicted to occur at certain values of E_1+E_2, $R=E_1/E_2$ and relative angle ϑ_{12} by calculations based on the 3C theory [10]. This finding has been supported by later calculations in the framework of the Convergent Close Coupling, CCC, model [21] and recently by the lowest-order perturbative approach, LOPT, of Istomin et al [22], while the hyperspherical *R* matrix with semiclassical outgoing waves, H*R*M-SOW, model predicts no such nodes [19]. As for the experiments some evidences have been provided [15,18], but nobody performed the most direct experiment to check the theoretical predictions. This involves a measurement of the CD at fixed E_1 and ϑ_{12} ($\vartheta_{12} \neq 0$ or 180°, which would make CD=0 always) as a function of hv and, of course, of E_2, which has to be scanned simultaneously with hv. In such a measurement the “existence” of a dynamic node is directly verified by the observation of a change in the sign of the measured CD.

This work aims to combine two sets of measurements with linearly and circularly polarised radiation in order to achieve a complete PDI experiment, to use the obtained moduli and relative phase of $a_g(E_1,E_2, \vartheta_{12})$ and $a_u(E_1,E_2, \vartheta_{12})$ to calculate other observables of PDI, and to investigate the existence of the dynamic nodes.

The experiments have been performed using the electron-electron multicoincidence end-station [23] at the Gas Phase Photoemission [24], GAPH, and Circularly Polarised [25], CIPO, beamlines of the Elettra storage ring. Two independently rotatable turntables, which hold seven and three electrostatic hemispherical analyzers, respectively, are housed in the chamber. The larger turntable rotates in the plane perpendicular to the direction, **z**, of propagation of the incident radiation. The three spectrometers of the smaller turntable are mounted at 0°, 30° and 60° with respect to the polarisation vector $\boldsymbol{\varepsilon}=\varepsilon\mathbf{x}$ of the light. This turntable can be rotated from the perpendicular plane to the $(\mathbf{z},\mathbf{x})$ plane, but in these measurements it has been kept in the perpendicular plane. The three fixed analyzers have been set to detect electrons of kinetic energy E_1=3.5 eV, while the other seven detect electrons with a kinetic energy E_2 from 20 to 50 eV depending on the measurement. The energy resolution and the angular acceptance in the dispersion plane of the spectrometers were $\Delta E/E_{1,2}$=0.03 and ±3°, respectively. The relative angular efficiency of the analyzers has been established by measuring photoelectron angular distributions with well known asymmetry parameters.

The light source of the GAPH beamline was an undulator of period 12.5 cm, 4.5 m long. The experiment has been performed at $h\nu$=127 eV, using the first harmonic of the undulator, which is expected to be completely linearly polarized. This was checked by measuring the photoelectron angular distribution of He^+ ($n=2$) at the same photon value. The light source of the CIPO beamline is an electromagnetic elliptical undulator/wiggler [26], which allows to work with variable polarization state using the first harmonic emission between 5 and 150 eV. According to results of the characterisation measurements with a multilayer polarimeter [27,25], the radiation emitted by the electromagnetic undulator in the range of interest is characterized by the Stoke parameters S_1=0.31, S_3=±0.95 and the major axis of the polarization ellipse lays at λ=35° with respect to the orbital plane of the electrons in the ring. Here we adopted the definition that S_3=1 corresponds to a radiation whose polarization vector rotates clockwise for an observer looking towards the source.

In the case of an incident radiation that propagates along the z axis and is linearly polarised along the $\boldsymbol{\varepsilon}=\varepsilon\mathbf{x}$ axis, taking into account the invariance with respect to the rotation around a preferential symmetry axis, the TDCS can be written [5]

$$TDCS(E_1,E_2,\vartheta_{12}) = \left| a_g(E_1,E_2,\vartheta_{12})(\cos\vartheta_1 + \cos\vartheta_2) + a_u(E_1,E_2,\vartheta_{12})(\cos\vartheta_1 - \cos\vartheta_2) \right|^2 =$$
$$= |a_g|^2(\cos\vartheta_1 + \cos\vartheta_2)^2 + |a_u|^2(\cos\vartheta_1 - \cos\vartheta_2)^2 + 2|a_g||a_u|\cos\delta(\cos\vartheta_1^2 - \cos\vartheta_2^2) \qquad (1)$$

where ϑ_1 and ϑ_2 are the angles of emission of the two photoelectrons with respect to $\boldsymbol{\varepsilon}$ and ϑ_{12} is their relative angle. In the general case of an elliptically polarized radiation the TDCS in the plane perpendicular to the direction of the incident radiation can be expressed as

$$\begin{aligned} TDCS(E_1,E_2,\vartheta_{12}) &= \left\{ |a_g|^2(1+\cos\vartheta_{12}) + |a_u|^2(1-\cos\vartheta_{12}) \right\} + \\ &+ S_1\left\{ \cos(2\vartheta_1 + \cos\vartheta_{12})\left[|a_g|^2(1+\cos\vartheta_{12}) - |a_u|^2(1-\cos\vartheta_{12}) \right] + 2\sin(2\vartheta_1 + \cos\vartheta_{12})\sin\vartheta_{12}\,\mathrm{Re}(a_g a_u^*) \right\} + \qquad (2) \\ &- 2S_3 \sin\vartheta_{12}\,\mathrm{Im}(a_g a_u^*) \end{aligned}$$

where ϑ_1 is now referred to the major axis of the polarization ellipse.

In the case of fully circularly polarised radiation ($S_3=\pm1$, $S_1=0$) the TDCS is independent of ϑ_1. Thus TDCS measured at the same ϑ_{12}, but at different ϑ_1 can be added up. The main effect when using elliptically polarised radiation is a variation of the ratio of the two lobes of the TDCS measured at different ϑ_1. This has been observed for instance in the measurements by Collins et al [19], where ϑ_1 was varied over more than 100°. Here ϑ_1 varies ±30° with respect to the direction of the major axis of the polarization ellipse. A simulation of the TDCS in the conditions of our experiment using Eq. (2) and the amplitudes predicted by the CCC model showed variations of TDCS comparable or less than the uncertainty associated with the measurements. Therefore we added up the data at the same ϑ_{12} of the three TDCS, simultaneously measured in the experiment .

The circular dichroism, CD, is obtained by measuring the TDCS with radiation of both helicities

$$CD(E_1,E_2,\vartheta_{12}) = \frac{1}{|S_3|}\left[TDCS_L(E_1,E_2,\vartheta_{12}) - TDCS_R(E_1,E_2,\vartheta_{12}) \right] =$$
$$= +4\sin\vartheta_{12}\,\mathrm{Im}(a_g a_u^*) = -4\sin\vartheta_{12}|a_g||a_u|\sin\delta \qquad (3)$$

where $TDCS_L$ ($TDCS_R$) is the TDCS for PDI by left (right), i.e. $S_3=-1$ (+1), circularly polarized radiation. From the experimental point of view it is also convenient to define the normalized circular dichroism, CD_n, defined as

$$CD_n(E_1,E_2,\vartheta_{12}) = \frac{1}{S_3}\frac{TDCS_L(E_1,E_2,\vartheta_{12}) - TDCS_R(E_1,E_2,\vartheta_{12})}{TDCS_L(E_1,E_2,\vartheta_{12}) + TDCS_R(E_1,E_2,\vartheta_{12})} \qquad (4)$$

Eq.s(1-4) show that a proper set of measurements of the TDCS with linearly and circularly polarized radiation provides the full information on the complex amplitudes, i.e. their moduli and relative phase .

In panels (a-c) of fig. 1 the three TDCS measured simultaneously at 127 eV for E_1=3.5 eV and ϑ_1=0,30 and 60 ° with linearly polarized radiation are shown. In panel (d) the CD_n measured at the same photon energy and energy sharing is displayed. In panels (a-c) also the polar plots of the TDCS, which allow to evaluate the variation in shape versus ϑ_1, are shown. In the panel (d) the TDCS measured with the two helicities of the radiation used to extract the CD_n are also shown. All the experimental data have been compared with the prediction of the CCC model [21]. The CCC method is a fully numerical approach that relies on heavy computations. For the final state, it solves the Schrödinger equation for the system of a photoelectron scattering on a singly charged ion by employing the close coupling expansion of the total two-electron wave function. The initial state is represented by a highly correlated Hylleraas type wave function, the use of which ensures that the results have insignificant dependence on the gauge of the electromagnetic interaction. The He CCC integrated PDI cross sections and TDCS agree with experimental data over a broad energy range. The three TDCS of fig. 1(a-c), being measured simultaneously, are on the same relative scale, therefore in the comparison with the theory they have been arbitrarily rescaled by a common factor. Thus in this case, we observe that the CCC predicts correctly the shape of the TDCS as well as the relative intensity versus ϑ_1. The quality of the experimental TDCS measured with circularly polarized radiation is worse than that of the measurements with linearly polarized radiation. However a reasonable agreement within the experimental uncertainties is observed between theory and experiment in both the TDCS and CD_n. The procedure proposed by Bolognesi et al [8] has been applied to the data of the three TDCS measured with linearly polarized radiation . The method is based on three independent determinations of the TDCS at the same E_1,E_2 and ϑ_{12}. These values are inserted in a non-linear system based on Eq. (1) , whose solution gives a set of $|a_g|^2$, $|a_u|^2$ and cos δ. These quantities when used to reproduce the measured CD_n provide us with the sign of the relative phase δ, too. The results are shown in fig. 2, where they are also compared with the CCC predictions. Due to the geometrical constraints of our set-up we can obtain the amplitudes and the phase only in the region ϑ_{12} >90°, because with the three analyzers fixed at 0,30 and 60° we can not obtain three independent determinations of the TDCS at ϑ_{12} <90°. The moduli of the amplitudes in the region investigated are well represented by Gaussian functions. This is in agreement with the results by Kheifets and Bray [28] who theoretically verified that the Gaussian approximation for the a_g and a_u amplitudes holds up to an excess energy of 60 eV. The full width at half maximum of the a_g amplitude (Γ_g=98.7±2.9°) appears to be bigger of the a_u one (Γ_u =73.7±3.8°). These values agree very well with the analysis of previous measurements at 40 eV [7,29] and 60 eV [30] above the IP^{2+}. The agreement with the theoretical predictions is excellent also in the case of the phase.

The knowledge of a_g and a_u and their relative phase allows to predict other observables of the process. For example the linear dichroism, $\Delta_{lin}=\Delta TDCS(E_1 \leftrightarrow E_2)$, i.e. the difference between two TDCS measured with linearly polarized radiation in which the energies of the two electrons are exchanged, $E_1 \leftrightarrow E_2$, has been calculated using the experimental a_g and a_u for $\vartheta_1 = 0°$. At variance with circular dichroism which depends on Im(a_u), Δ_{lin} depends only on Re(a_u) [2]. The results are shown in fig.3a, where they are also compared with the predictions of CCC. In addition by integrating eq. (1) over all the directions of one of the two photoelectrons the asymmetry parameter, β, of the angular distribution of the fast(slow) photoelectron, $d^2\sigma / dEd\Omega_i \propto 1 + \beta_i P_2(\cos \vartheta_i)$ i=1,2, can be obtained. The calculated β from the experimental amplitudes 0.68±0.14 and -0.12±0.10 for the fast and slow photoelectron respectively, well compare with the predicted values by CCC (0.80 and -0.18, respectively). The angular distribution of one of the photoelectrons of PDI of He has been previously investigated both experimentally [31] and theoretically [32] at 120 eV. By considering the same energy sharing of the present experiment we found a very good agreement between those experimental and theoretical values and the ones of this work for the fast photoelectron. In the case of the β of the slow photoelectron the present experimental and theoretical values agree with the prediction of [32], while the experimental value of [31] appears to be higher (0.5±0.2). Finally also the recoil momentum distribution of the He^{2+} ion, dσ/dK where $\mathbf{K}=\mathbf{k}_1+\mathbf{k}_2$ is the center of mass of the two-electron subsystem and therefore the opposite of the ion recoil momentum, has been calculated from the a_g and a_u. The procedure and the numerical integrations used to obtain dσ/dK as well as β from Eq. (1) are detailed in [33, 34]. The derived dσ/dK is compared with the predictions of CCC in fig.3b. This latter finding shows for the first time that it is possible to reconstruct from an electron-electron experiment with conventional fixed analyzers a typical experimental observation of a COLTRIMS experiment.

It is interesting to note that in fig.1d the CD_n crosses zero and changes sign at about ϑ_{12}=85°. This is due neither to a vanishing value of the amplitudes nor to the geometrical factor in Eq. (3). Thus it must be due to a zero in the sinδ. Indeed in fig. 2 the theory predicts δ=π at this ϑ_{12}. Therefore this corresponds to a clear observation of a dynamic node. In a recent work Istomin et al. [22] discussed the origin of the dynamic nodes within the frame of their LOPT model. Their analysis of the CD versus ϑ_{12} resulted in the predictions that “at all excess energies up to 50 eV” two dynamic nodes should be observed: one in the range $14.6° \leq \vartheta_{12} \leq 40.1°$ and the other in the range $81.3° \leq \vartheta_{12} \leq 88.4°$. The present observation confirms the prediction of a node in the range of the larger ϑ_{12}. No experimental data cover the range of the smaller ϑ_{12}, however the present calculations do not appear to support the prediction of a node in that region.

In order to investigate better the existence of dynamic nodes a second set of measurements has been done at hv between 102.5 and 142.5 eV. The energy E_1 of the slow electron was kept fixed at 3.5 eV, while the energy E_2 of the fast electron was incremented according to the variation of the hv. In fig. 4a the CD_n measured at six different energies are reported versus the relative angle ϑ_{12}. A change in sign of the measured CD_n, from ϑ_{12}=67°, where within the experimental uncertainties the values at all the energies are positive, to ϑ_{12}=127°, where all the measured values are negative, is clearly observed. To emphasize this result the measured CD_n at ϑ_{12}=67, 97 and 127° are reported as a function of E_2 in fig. 4b. Consistently with the predictions of the CCC model the CD_n is changing sign at ϑ_{12}=97° when E_1=3.5 eV and $E_2 \approx 35$ eV. This result proves the existence of the dynamic nodes, whose occurrence depends on the energy sharing and relative angle ϑ_{12} between the two electrons, as was firstly predicted by Berakdar *et al* [10].

The realization of complete experiments, where all the basic quantities, i.e. the amplitudes of a particular process, are measured, is one of the main goal in atomic physics. The knowledge of these elements allows one to predict all the other observables of the process and represents the ultimate test for any theoretical description. Electron-electron and electron-ion coincidence experiments have been proved to be one among the most suited tools for the realization of complete photoionization experiments, as shown by the photoelectron-Auger electron experiments in the case of the atomic photoionization [35,36] and by the photoelectron-ion experiments in the case of the molecular inner shell photoionization. [37]. Here the combination of a series of electron-electron coincidence experiments with linearly and circularly polarized radiation enabled us to obtain a unique determination of the amplitudes needed to achieve a complete quantomechanical description of the PDI in He. The obtained amplitudes have been used to predict other observables of the PDI process. In particular it has been shown that the data measured in the electron-electron coincidence experiments can be used to calculate the recoiling ion momentum distribution, a typical result of COLTRIMS. Another result of this work is the experimental observation of a node in the CD in a condition where the relative phase between the two amplitudes is 0 or π. Model analytical derivations [38] showed that the vanishing of CD at certain emission angles and energy sharings is completely dependent on electron-electron correlations. Thus a better understanding of this aspect of CD may result of prominent importance in one-photon two-electron emission in highly correlated materials [39].

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Figure captions

Figure 1 : He TDCS for E_1=3.5 eV, E_2=44.5 eV and ϑ_1=0° (a), 30° (b) and 60°(c). In the top-left corner of each panel the experimental data are reported also in polar plots. In panel (d) the CD_n versus ϑ_{12} is shown. The TDCS obtained with the two helicities of the radiation are reported in the top-right ($TDCS_L$) and bottom-left ($TDCS_R$) corners of the same panel. The experimental TDCS are compared with the predictions of the CCC model (solid line).

Figure 2 : $|a_g|^2$, $|a_u|^2$ and δ as obtained from the experimental TDCS and the CD_n shown in fig. 1 are compared with the predictions of the CCC (full line).

Figure 3 : (a) Δ_{lin} as calculated from the experimental amplitudes and relative phase at ϑ_1=0° is compared with the predictions of CCC (full line). In the inset the complementary TDCS predicted by CCC are reported in a polar plot : E_1=3.5 eV, E_2=44.5 eV and ϑ_1=0° (full line) , E_1=44.5 eV, E_2=3.5 eV and ϑ_1=0° (dotted line); (b) $d\sigma/dK$ versus the rescaled momentum of recoiling nucleus. The $d\sigma/dK$ calculated from experimental amplitudes (dotted line) and the one predicted by CCC (solid line) have been renormalized to a maximum value of 1.

Figure 4 : CD_n versus ϑ_{12} at E_1=3.5 eV and six different $20.5 \leq E_2 \leq 59.5$ eV (a) and versus E_2 at ϑ_{12}=*67*° (dots), *97*° (triangle) and *127*°(diamond) (b). The solid lines in (b) are the CCC predictions.

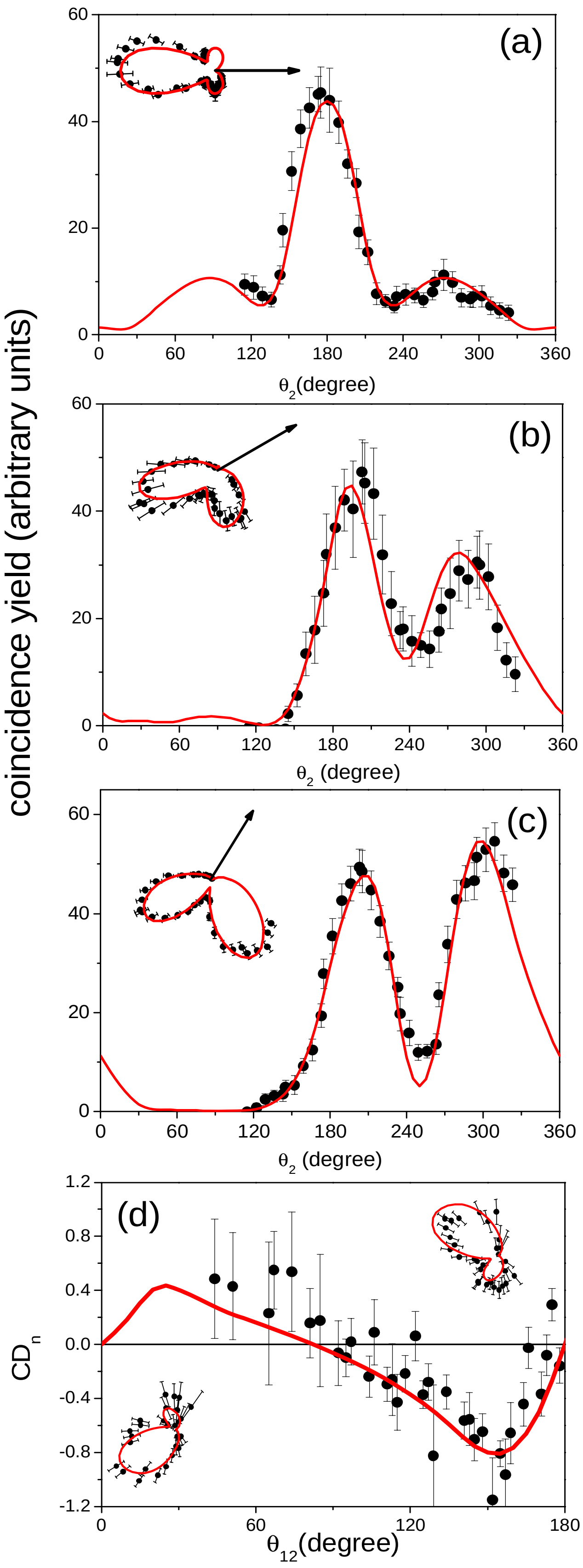

(a)
(b)
(c)
(d)
coincidence yield (arbitrary units)
θ_2(degree)
θ_2 (degree)
θ_2 (degree)
CD_n
θ_{12}(degree)

Figure 1

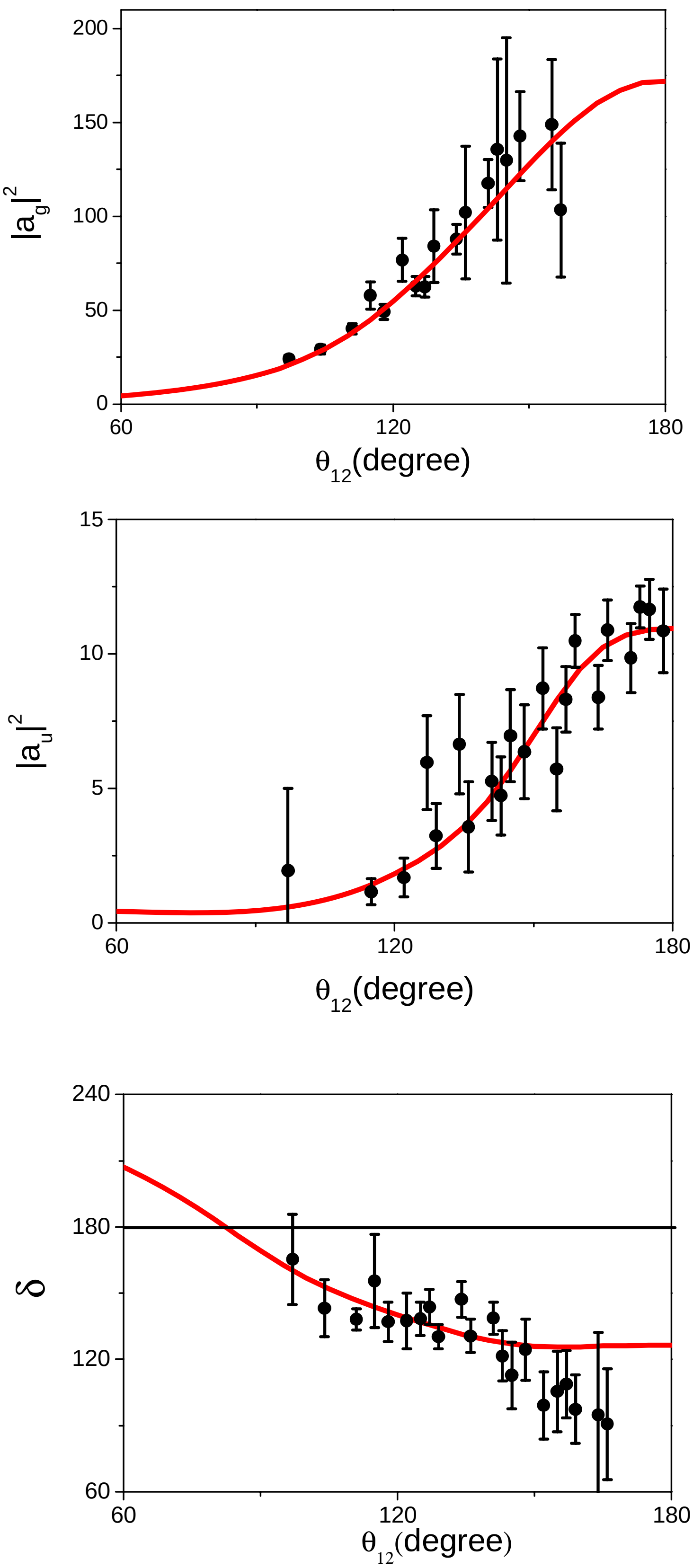

200
150
100
50
0
$|a_g|^2$
60
120
180
θ_{12}(degree)
15
10
5
0
$|a_u|^2$
60
120
180
θ_{12}(degree)
240
180
120
60
δ
60
120
180
θ_{12}(degree)

Figure 2

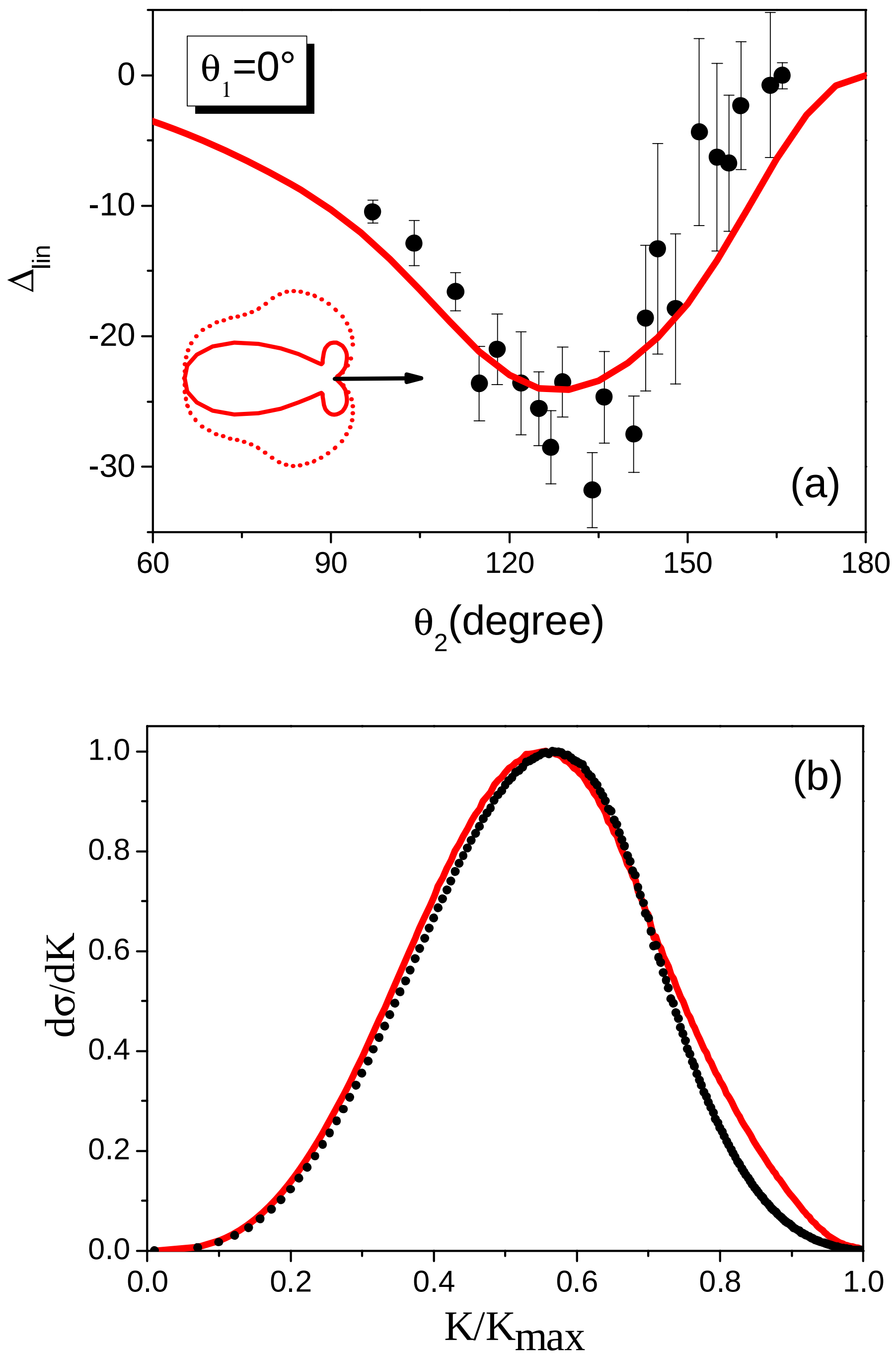
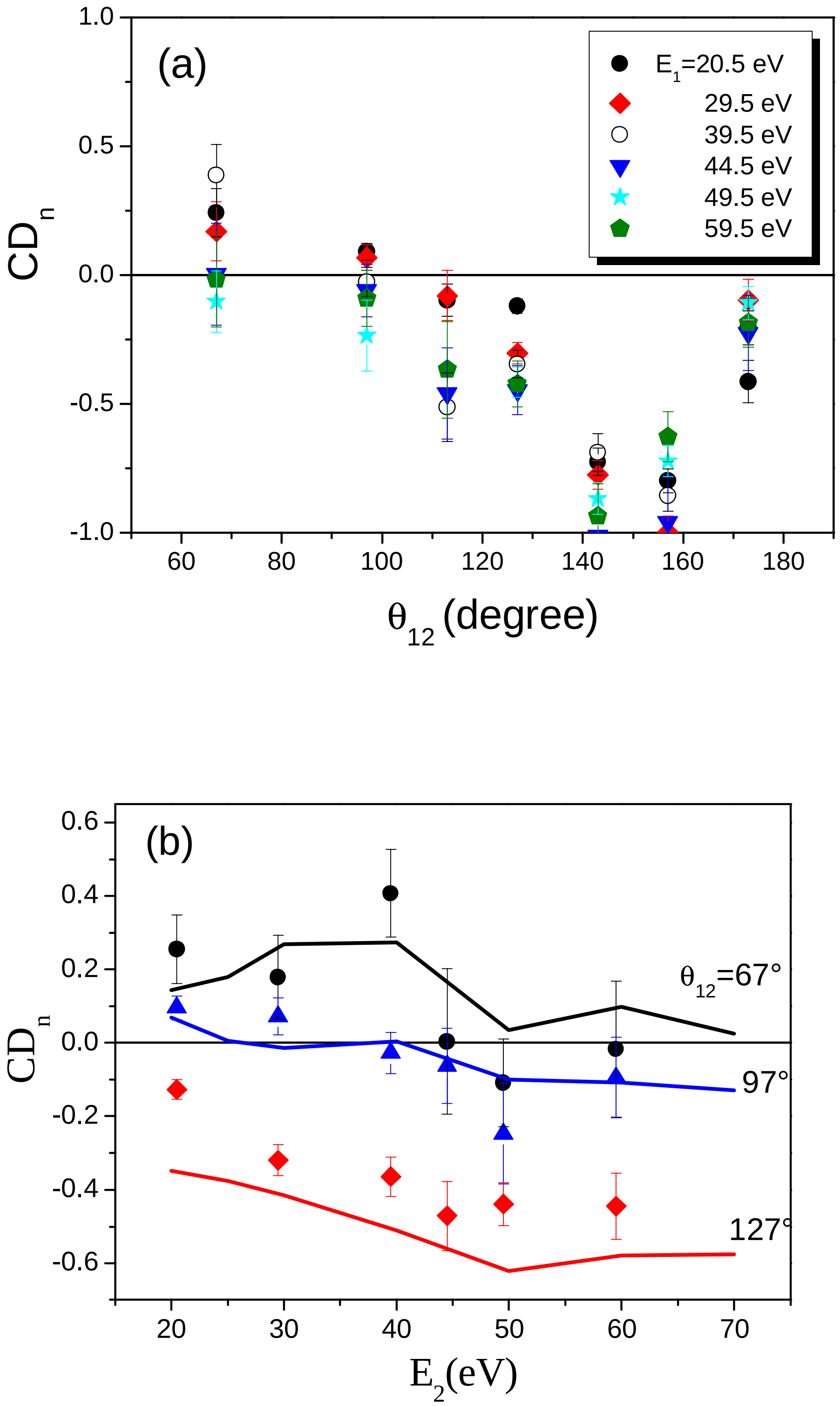

θ1=0°
0
-10
-20
-30
Δlin
60
90
120
150
180
θ2(degree)
(a)
1.0
0.8
0.6
0.4
0.2
0.0
dσ/dK
0.0
0.2
0.4
0.6
0.8
1.0
K/Kmax
(b)

Figure 3



(a)
E1=20.5 eV
29.5 eV
39.5 eV
44.5 eV
49.5 eV
59.5 eV
CDn
θ12 (degree)
(b)
θ12=67°
97°
127°
CDn
E2(eV)

Figure 4

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