

STUDYING COMPTON DOUBLE IONIZATION OF HELIUM ATOM WITH COLTRIMS DETECTOR.

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September 19, 2021

The talk is based on the unique experiments carried out by the Goethe University team (Frankfurt am Main):

M. Kircher, G. Kastirke, M. Weller, L. Ph .H. Schmidt, I. Vela-Pérez, M. Waitz, T. Mletzko, S. Grundmann, D. Kirchner, A. Khan, F. Trinter, T. Jahnke, M. S. Schöffler, and R. Dörner,

as well as on theory and calculations by theorists:

O. Chuluunbaatar (JINR, Russia), S. Houamer (Univ.Setif-1, Algeria), I.P. Volobuev and Yu.V. Popov (SINP MSU, Russia).

A joint paper is under preparation.

Compton single ionization of helium was presented in the paper

M. Kircher, F. Trinter, S. Grundmann, I. Vela-Perez, S. Brennecke, N. Eicke, J. Rist, S. Eckart, S. Houamer, O. Chuluunbaatar, Yu.V. Popov, I.P. Volobuev, Kai Bagschick, M.N. Piancastelli, M. Lein, T. Jahnke, M.S. Schöffler, and R. Dörner. “Kinematically complete experimental study of Compton scattering at helium atoms near the threshold”, Nature Physics **16** (2020), 756 -760

as well as in the talk of M. Kircher at the ISIAC satellite.

Here is the second part of this story ...

In general, Compton scattering is a relativistic process. Its theory for single ionization is well known for a long time and presented in many textbooks. However, Compton double ionization remained unstudied. If the initial photon energy ω_1 is of the order of a few tens of keV, and the energy of the emitted electron is up to a few tens of eV, the non-relativistic description is applicable. The energy and momentum conservation laws are

$$\omega_1 = \omega_2 + I_p + E_1 + E_2 + E_{ion}, \quad (1.1)$$

$$\vec{k}_1 = \vec{k}_2 + \vec{p}_1 + \vec{p}_2 + \vec{K}, \quad (1.2)$$

where I_p is the ionization potential, E_i (\vec{p}_i) is the energy (momentum) of an emitted electron, E_{ion} (\vec{K}) is the energy (momentum) of the residual ion, ω_i (\vec{k}_i) is the energy (momentum) of the initial (final) photon. The momentum transfer is given by $\vec{Q} = \vec{k}_1 - \vec{k}_2$.

In the non-relativistic energy range we can use the atomic units $e = m_e = \hbar = 1$. In these units, $E_i = p_i^2/2$, $k_i = \omega_i/c$, $\omega(a.u.) = \omega(keV)/27.2(eV) \sim c (= 137)$, so that $k_i \sim 10$ in the chosen photon energy range.

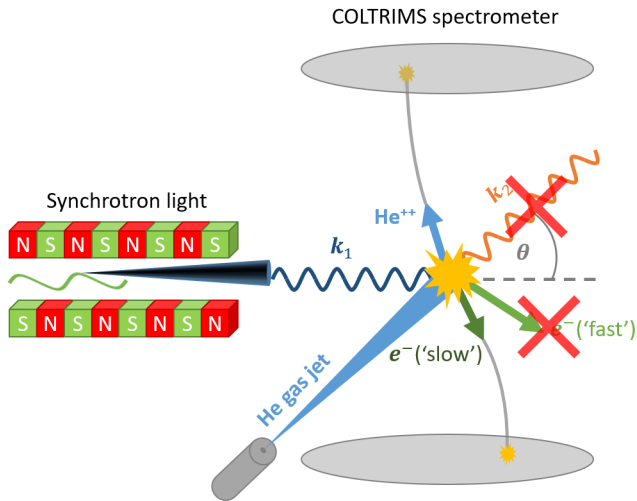
Motivation

The main motivation of this investigation is to establish a new method of studying the correlations of electrons in an atomic target in analogy with the (e,3e) and (e,3-1e) Electron Momentum Spectroscopy (quasielastic scattering).

Here the first step towards this goal is presented, namely, we show the feasibility of such very delicate measurements.

Experiment

Scheme of the experiment



Definitions

In the experiment under consideration, the energy and momentum of the initial photon were $\omega_1 = k_1 c = 40$ keV ($k_1 = 10.7$ a.u.). We choose \vec{k}_1 as the z-axis. The energy (momentum) of the scattered photon is denoted by $\omega_2(\vec{k}_2)$, and $\vec{Q} = \vec{k}_1 - \vec{k}_2$ is the momentum transfer. The linear polarizations of the photons \vec{e}_1 , \vec{e}_2 satisfy the conditions $(\vec{e}_1 \cdot \vec{k}_1) = (\vec{e}_2 \cdot \vec{k}_2) = 0$. As we have already mentioned, the momenta of the emitted electrons are denoted by \vec{p}_1 , \vec{p}_2 , \vec{K} is the momentum of the residual ion He^{++} . In the non-relativistic case $p_j^2/2$ is the electron energy, $K^2/2M$ is the energy of the ion, which we neglect in the calculations, and $I_p = 79$ eV is the helium DI potential. In the atomic units we use, $\alpha = 1/c = 1/137$, $r_0 = \alpha^2$ is the classical radius of the electron: $r_0^2 = 7.94 \cdot 10^{-26}$ cm², $r_0^2/(2\pi)^6 = 1.3 \cdot 10^{-30}$ cm² = $1.3 \cdot 10^{-6}$ barn. = $0.46 \cdot 10^{-13}$ a.u.

The cross section of double Compton ionization is extremely small, about a million times smaller than the typical cross section of photoionization at the corresponding threshold.

Non-relativistic approach

We consider Compton scattering at helium atoms. The non-relativistic TDSE, which describes the atom-light interaction, reads:

$$i\frac{\partial}{\partial t}\Psi(\vec{r}_1, \vec{r}_2, \vec{r}_n, t) = \left[\frac{1}{2} \left(-i\vec{\nabla}_1 - \frac{1}{c}\vec{A}(\vec{r}_1, t) \right)^2 + \frac{1}{2} \left(-i\vec{\nabla}_2 - \frac{1}{c}\vec{A}(\vec{r}_2, t) \right)^2 + \frac{1}{8M} \left(-i\vec{\nabla}_n + \frac{2}{c}\vec{A}(\vec{r}_n, t) \right)^2 - \frac{2}{|\vec{r}_n - \vec{r}_1|} - \frac{2}{|\vec{r}_n - \vec{r}_2|} + \frac{1}{|\vec{r}_1 - \vec{r}_2|} \right] \Psi(\vec{r}_1, \vec{r}_2, \vec{r}_n, t). \quad (2)$$

In Eq. (2) $M = 1836$ a.u. is the proton mass, \vec{r}_n is the coordinate of the helium nucleus, and $\vec{r}_{1,2}$ denote the coordinates of the electrons. The vector potential is defined as follows

$$\frac{1}{c}\vec{A}(\vec{r}, t) = \sqrt{\frac{2\pi}{\omega_1}} \vec{e}_1 e^{i(\vec{k}_1\vec{r} - \omega_1 t)} + \sqrt{\frac{2\pi}{\omega_2}} \vec{e}_2 e^{-i(\vec{k}_2\vec{r} - \omega_2 t)} + (c.c.). \quad (3)$$

Here \vec{e}_1 and \vec{e}_2 are linear polarizations of the initial and final photons. This choice of the vector potential corresponds to a single incident photon and a single outgoing photon per unit volume. We remind, that $(\vec{k}_i \cdot \vec{e}_i) = 0$, so that $\text{div}\vec{A}(\vec{r}, t) = 0$ (the Coulomb gauge).

The interaction term of an electron and the photon is written as

$$\begin{aligned}
 V_{int} = & i \frac{1}{c} (\vec{A}(\vec{r}, t) \cdot \vec{\nabla}_r) + \frac{1}{2c^2} A^2(\vec{r}, t) = \\
 & i \left(\sqrt{\frac{2\pi}{\omega_1}} e^{i(\vec{k}_1 \vec{r} - \omega_1 t)} (\vec{e}_1 \cdot \vec{\nabla}_r) + \sqrt{\frac{2\pi}{\omega_2}} e^{-i(\vec{k}_2 \vec{r} - \omega_2 t)} (\vec{e}_2 \cdot \vec{\nabla}_r) \right) + \\
 & \left(\frac{\pi}{\omega_1} (1 + e^{2i(\vec{k}_1 \vec{r} - \omega_1 t)}) + \frac{\pi}{\omega_2} (1 + e^{-2i(\vec{k}_2 \vec{r} - \omega_2 t)}) \right) + \\
 & \left(\frac{2\pi}{\sqrt{\omega_1 \omega_2}} (\vec{e}_1 \cdot \vec{e}_2) e^{i[(\vec{k}_1 - \vec{k}_2) \vec{r} - (\omega_1 - \omega_2)t]} \right) + (c.c). \quad (4)
 \end{aligned}$$

The term marked in red is the well-known Kramers - Heisenberg - Waller matrix element. It is also called A^2 term, which is analogous to the FBA in agreement with the ionization reactions with electrons and bare ions. We focus our attention on considering this term.

The fully differential cross section (FDCS) of the double Compton ionization process can be written in the form:

$$d^{12}\sigma = \frac{(2\pi)^6 \alpha}{\omega_1 \omega_2} |M|^2 \delta(\omega_1 - I_p - \omega_2 - p_1^2/2 - p_2^2/2) \delta^3(\vec{k}_1 - \vec{k}_2 - \vec{p}_1 - \vec{p}_2 - \vec{K}) \times$$

$$\frac{d^3 k_2}{(2\pi)^3} \frac{d^3 p_1}{(2\pi)^3} \frac{d^3 p_2}{(2\pi)^3} \frac{d^3 K}{(2\pi)^3}. \quad (5)$$

Only a slow electron and the helium ion He^{++} are detected in the experimental setup. Therefore, we have to integrate this expression with respect to \vec{k}_2 and \vec{p}_1 to get 6DCS. We obtain

$$\frac{d^6\sigma}{d^3 p_2 d^3 K} = \frac{\alpha^2}{(2\pi)^6 \omega_1^2} \int \frac{d^3 k_2}{k_2} |M(\vec{k}_1 - \vec{k}_2 - \vec{p}_2 - \vec{K}, \vec{p}_2; \vec{k}_1, \vec{k}_2)|^2 \times$$

$$\delta\left(t - \frac{k_2}{k_1} - \frac{\alpha}{2k_1} (\vec{k}_1 - \vec{k}_2 - \vec{p}_2 - \vec{K})^2\right), \quad t = 1 - \frac{E_2 + I_p}{\omega_1} \approx 1. \quad (6)$$

Here $\vec{k}_1 - \vec{k}_2 - \vec{p}_2 - \vec{K} = \vec{p}_1$ stands for the momentum of electron 1, and the delta function takes into account energy conservation. We define x , $0 \leq x \leq 1$ by the relation $k_2 = k_1 x$ and rewrite Eq. (6) as follows:

$$\frac{d^6 \sigma}{d^3 p_2 d^3 K} = \frac{\alpha^4}{(2\pi)^6} \int_0^1 x dx \int d\Omega_1 |M(\vec{k}_1 - \vec{k}_2 - \vec{p}_2 - \vec{K}, \vec{p}_2, \vec{K}; \vec{k}_1, \vec{k}_2)|^2 \times \delta \left(t - x - \frac{\alpha k_1}{2} \left(\vec{n}_1 - \vec{n}_2 x - \frac{\vec{p}_2 + \vec{K}}{k_1} \right)^2 \right). \quad (7)$$

In Eq. (7) \vec{n}_1 (\vec{n}_2) is the unit vector directed along the vector \vec{k}_1 (\vec{k}_2).

Now we consider the expression in the argument of the delta function. First, we note that $t = 1$ with a high accuracy. The factor $\alpha k_1/2$ can be estimated as $\alpha k_1/2 \approx 0.04$. For the photon backscattering the contribution of the expression in the round brackets in the delta function is about 16%, if $|\vec{p}_2 + \vec{K}| \ll k_1$. An approximate solution of the equation for x to the first order in α is $x_1 \approx 1 - \frac{\alpha k_1}{2} (\vec{n}_1 - \vec{n}_2 - \frac{\vec{p}_2 + \vec{K}}{k_1})^2$. This means that the energy of the final photon is very close to the energy of the initial one. Thus,

$$\frac{d^6\sigma}{d^3p_2 d^3K} = \frac{\alpha^4}{(2\pi)^6} \int d\Omega_1 x_1 |M(\vec{k}_1 - \vec{k}_2 - \vec{p}_2 - \vec{K}, \vec{p}_2, \vec{K}; \vec{k}_1, \vec{k}_2)|^2, \quad (8)$$

where we have put $\vec{k}_2 = k_1 x_1 \vec{n}_2$.

Within the A^2 model we have

$$M(\vec{p}_1, \vec{p}_2; \vec{e}_1, \vec{e}_2) = (\vec{e}_1 \cdot \vec{e}_2) \langle \Phi^- (\vec{p}_1, \vec{p}_2) | e^{i\vec{Q} \cdot \vec{r}_1} + e^{i\vec{Q} \cdot \vec{r}_2} | \Phi_0 \rangle. \quad (9)$$

If the momentum transfer Q is fairly large, the momenta of the electrons are quite different: $\vec{p}_1 = \vec{Q} - \vec{p}_2 - \vec{K}$. This kinematical condition is very close to the above mentioned EMS (e,3e) geometry, where the two final electrons (the incident one and the fast ejected one) approximately share the initial energy, and their scattering angles are about 45° . In this case the momentum transfer is maximal. However, the third ejected electron must be slow, and its production mechanism is mainly shake off. It was shown that this kinematics is the best one to observe the correlations by measuring the angular distributions of the final fragments.

To get the final formulas we choose the final state orthogonal to the initial one:

$$\langle \tilde{\Phi}^-(\vec{p}_1, \vec{p}_2) | = \langle \Phi^-(\vec{p}_1, \vec{p}_2) | - \langle \Phi^-(\vec{p}_1, \vec{p}_2) | \Phi_0 \rangle \langle \Phi_0 |, \quad (10)$$

and take into account the symmetrization by introducing the factor $1/\sqrt{2}$. The summation/averaging over the final and initial photon polarizations gives the factor $[1 + (\vec{n}_1 \cdot \vec{n}_2)^2]$ in integral (8). **Finally we obtain**

$$6DCS \equiv \frac{d^6\sigma}{d^3p_2 d^3K} = \frac{r_0^2}{(2\pi)^6} \int d^2n_2 x_1 [1 + (\vec{n}_1 \cdot \vec{n}_2)^2] \left| \langle \Phi^-(\vec{p}_1, \vec{p}_2) | e^{i\vec{Q} \cdot \vec{r}_1} | \Phi_0 \rangle + \langle \Phi^-(\vec{p}_1, \vec{p}_2) | e^{i\vec{Q} \cdot \vec{r}_2} | \Phi_0 \rangle - 2 \langle \Phi^-(\vec{p}_1, \vec{p}_2) | \Phi_0 \rangle \langle \Phi_0 | e^{i\vec{Q} \cdot \vec{r}} | \Phi_0 \rangle \right|^2. \quad (11)$$

In the integrands the variable p_1 is mated to the variable r_1 . Therefore, the first term in Eq. (11) describes the direct process (T_1), the second one corresponds to the exchange process (T_2), and the third term appears due to the orthogonalization (T_3). We remind that $r_0^2/(2\pi)^6 = 1.3 \cdot 10^{-30} \text{ cm}^2 = 1.3 \cdot 10^{-6} \text{ barn.} = 0.46 \cdot 10^{-13} \text{ a.u.}$

To estimate the ability of this method to study (ee)–correlations in the target atom, we use the correlated final BBK wave function and a highly correlated trial initial state, normalized to 1, which we denote as CF:

$$\Phi_0^{CF}(\vec{r}_1, \vec{r}_2) = \sum_{j=1}^{10} D_j (e^{-a_j r_1 - b_j r_2} + e^{-a_j r_2 - b_j r_1}) e^{-\gamma_j r_{12}}. \quad (12)$$

This function provides the helium DI ionization potential $I_p^{CF} = 2.90371$ a.u., which is practically equal to its experimental value $I_p^{exp} = 2.903724$ a.u.. The final double continuum wave function is given by the well-known BBK (3C) function:

$$\Phi_{3C}^{(-*)}(\vec{r}_1, \vec{r}_2) = e^{i\vec{p}_{12} \cdot \vec{r}_{12}} \phi_1^{-*} \phi_2^{-*} \phi_{12}^{-*}. \quad (13)$$

Here the Coulomb wave function

$$\phi_j^{-*}(\vec{p}_j, \vec{r}) = R(\xi_j) e^{-i\vec{p}_j \cdot \vec{r}} {}_1F_1[-i\xi_j, 1; i(p_j r + \vec{p}_j \cdot \vec{r})],$$

with

$$\vec{p}_{12} = \frac{1}{2}(\vec{p}_1 - \vec{p}_2); \quad \xi_{12} = \frac{1}{2p_{12}}; \quad \xi_j = -\frac{2}{p_j} \quad (j = 1, 2); \quad R(\xi) = e^{-\pi\xi/2} \Gamma(1 + i\xi).$$

Physical considerations

Let us consider now in more detail the physical mechanisms that control the Compton double ionization. The presentation here is based on a simpler model. We describe the "fast" electron by a plane wave and the initial state by the simplest Hylleraas helium wave function ($I_p^{Hy} = 2.85$)

$$\Phi_0^{Hy}(\vec{r}_1, \vec{r}_2) = \frac{Z^3}{\pi} e^{-Z(r_1+r_2)}, \quad Z = 27/16.$$

The "slow" electron is described by the Coulomb wave ($Z = 2$). This simplest Hy + PW + CW model provides analytical expressions for the matrix elements T_j and allows one to give them a simple physical interpretation.

Recall that, after the symmetrization, the matrix elements T_j are presented in such a way that the momentum of the fast electron \vec{p}_1 mates the coordinate \vec{r}_1 . In the matrix element T_1 the momenta \vec{Q} and \vec{p}_1 form the combination $\vec{Q} - \vec{p}_1 = \vec{p}_2 + \vec{K}$ in the exponential, i.e. here the total momentum in the exponential is small. By varying this momentum and measuring it, we probe the momentum distribution of the active electron with coordinate \vec{r}_1 . This term corresponds to the direct interaction of a photon with an electron. This electron both absorbs the photon and emits it. The second ("slow") electron is emitted with momentum \vec{p}_2 due to the shake-off mechanism. The T_1 term gives the most valuable information about the internal structure of the atom, in full analogy to (e,3e). This term does not depend on the photon scattering angle θ , and its integration in Eq. (11) does not change it.

Now we turn to the term T_2 . Here the fast momenta \vec{Q} and \vec{p}_1 no longer meet each other, and enter different integrals over the coordinates, and their contributions quickly vanish with the increase of the momentum transfer \vec{Q} as the photon scattering angle increases. In the matrix element T_2 , the momenta form the combination $\vec{Q} - \vec{p}_2$ in the exponential. At large scattering angles of the photon, the momentum transfer increases rapidly, and this term rapidly tends to zero. Here the physics can be interpreted as follows. Electron 2 absorbs the photon and transfers the absorbed momentum to electron 1 through the internal ee-correlation. Electron 1 escapes with the momentum \vec{p}_1 . This is a typical exchange process, and in the absence of any correlations of electrons in the atom, even through the mean field, this term is equal to zero.

The matrix element T_3 is artificial, because it arises due to the non-orthogonality of the probing functions of the initial and final states of the helium atom, and in the Hy + PW + CW model does not depend at all on the angles of the electron and ion. Usually it is small in the entire range of photon scattering angles. We can also connect this term with the nucleus, which absorbs the photon and transfers the momentum (not energy) to the "fast" electron as well.

Results

What do we calculate?

Despite the ability to measure in coincidence the energy and angles of both the final ion and one ("slow") of the electrons, the extremely small cross section (about 10^6 times smaller than the typical photoionization cross section) does not allow to gain sufficient statistics for a limited time of the experiment, therefore various single differential cross sections (SDCS) have been measured

$$SDCS_K = \frac{d\sigma}{dK} = (2\pi)K^2 \int_0^\pi \sin \theta_K d\theta_K \int d^3 p_2 \text{ 6DCS} \quad (16.1)$$

Here the integration with respect to p_2 is performed over the interval $0 < p_2 < p_{20} = 1.1$ a.u.

$$SDCS_p = \frac{d\sigma}{dE_2} = (2\pi)p_2 \int_0^\pi \sin \chi d\chi \int d^3 K \text{ 6DCS} \quad (16.2)$$

Here the integration with respect to K is performed over the interval $0 < K < K_0 = 2$ a.u.

$$SDCS_\chi = \frac{d\sigma}{d \cos \chi} = (2\pi) \int_0^{p_{20}} p^2 dp \int d^3 K \text{ 6DCS}. \quad (16.3)$$

Here again $0 < K < K_0 = 2$ a.u.

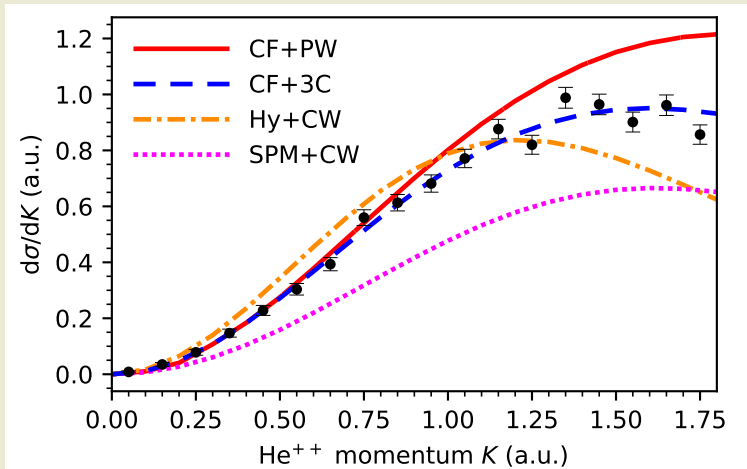


Figure 2: $SDCS_K$ Eq. (16.1) (in atomic units, but the coefficient α^4 in 6DCS is omitted for convenience) versus the ion momentum K in atomic units. In all calculations the "slow" electron is described by the Coulomb wave (CW) with $Z = 2$. Solid red line: CF+PW ("fast" electron). Dashed blue line: CF+3C. Dash-dotted orange line: Hy+CW ("fast" electron, $Z = 1$). Dotted magenta line: SPM+CW ("fast" electron, $Z = 1$). Data is always normalized to the integral of CF+3C (in all figs)

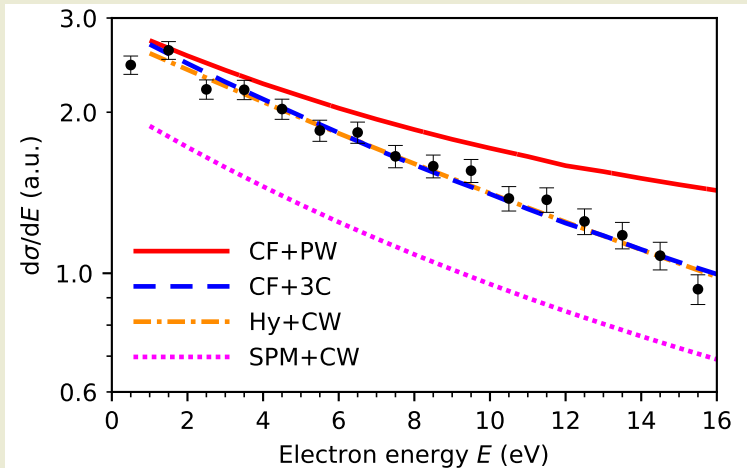


Figure 3: $SDCS_p$ Eq. (16.2) versus the electron energy E_e in eV. All other definitions are the same as in Fig. 2

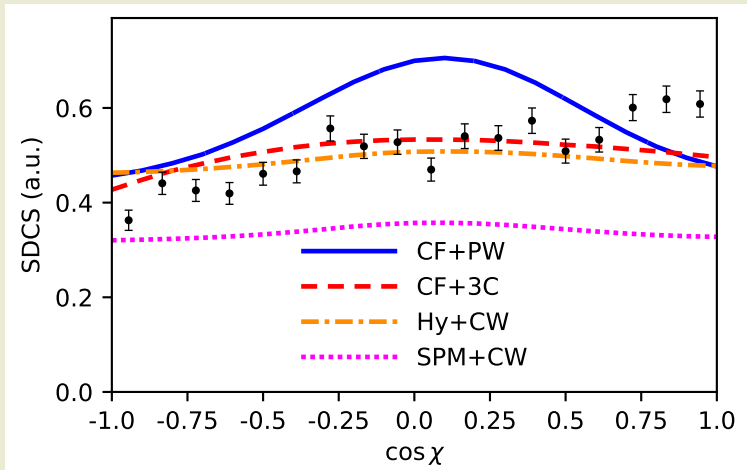


Figure 4: $SDCS_{\chi}$ Eq. (16.3) versus the $\cos \chi$ (the angle of the "slow" electron). All the other definitions are the same as in Fig. 2.

Everything said above about the terms T_j applies to the matrix elements before the integration in formula (11). In the Hy + PW model, the integration in (11) does not affect the term T_1 , because it does not depend on the momentum transfer explicitly. The result of the integration of the other terms is non-trivial, but **the main contribution to integral (11) comes from the integration over the forward scattering cone of the photon, $\theta \lesssim 30^\circ$.**

For example, let us pay attention on the peak at $\chi = 90^\circ$ for the model CF+PW. This peak comes from T_2 , mainly from the small photon scattering angles. In this case vector \vec{Q} is small and practically orthogonal to \vec{k}_1 . The term T_2 reaches its maximum, when $\vec{Q} - \vec{p}_2 \sim 0$, i.e. $\chi \sim 90^\circ$. Correlations in the initial state do not change this conclusion.

Thus, the scattering process considered here is analogous to (e,3e), or (ion, ion 2e) reactions with fast projectiles, but the emitted electrons are slow and have approximately equal energies. Such reactions are called "dipolar". The first (e,3e) experiment in this setting was carried out by A. Lahmam-Bennani.

As the scattering angle increases, the terms T_2 and T_3 decrease rapidly, whereas the matrix element T_1 survives and, as it was explained above, it carries the most valuable spectroscopic information on the distribution of momenta of the pair of electrons in an atom. Thus, the question arises: is it possible to somehow cut off the negative impact of the forward scattering cone of the photon? Say, by integrating in (11) not from zero, but from a cutoff angle θ_0 . This question has a positive answer, but not for the given design of the COLTRIMS.

Concluding remarks and perspectives

- The talk presents the results of the first experiment on measuring a number of single differential cross sections of the Compton double ionization of the helium atom. For this purpose, the COLTRIMS detector and a powerful source of synchrotron radiation have been used. A characteristic feature of this experiment is the measurement in coincidence of the momentum of one (slow) of the emitted electrons and the momentum of the ion residue. The final scattered photon was not detected. The initial photon energy was 40 keV.
- The theoretical description of the experiment is based on the so-called A^2 approximation. The theoretical and numerical treatment of the problem was characterized by multiple integration in deriving various single differential cross sections. For a pair of strongly correlated initial and final states, the number of integrations was 9, for which it was necessary to develop special numerical methods and to carry out a serious analytical analysis of the inner integrals.
- Even the single differential cross sections have shown the selective capabilities of the Compton double ionization as a spectroscopic method in atomic physics along with other similar methods. The best fit of the set of the experimentally measured single differential cross sections was given by the most correlated pair of the trial functions.
- We have to change the design of COLTRIMS so as to be able to exclude from statistics the events with the photon scattered in the forward cone.

Thank you for your attention !