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Compton Ionization of Atoms as a New Method of Spectroscopy of Outer Shells

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Abstract—A method of measuring the full differential cross section (FDCS) of single Compton ionization of a helium atom, which does not need detecting the scattered photon, has been presented in a recent paper published in the journal Nature physics [1]. The experimental data have been described within a theory based on the A^2 approximation. In the case of low (of the order of several keV) photon energies the model has given a good fit of the data. In the present paper, the possibility of studying the momentum distribution of the active electron in the target atoms with the help of such reactions is discussed in more detail.

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INTRODUCTION

Recently, a kinematically complete experimental measurement of Compton ionization cross sections on free helium atoms has been conducted by an international research group at synchrotron Petra III (DESY, Hamburg) with the help of the COLTRIMS detector (COLd Target Recoil Ion Momentum Spectroscopy). The obtained results have also been described theoretically.

The experiment observed the Compton scattering of photons with an energy of 2.1 keV by helium atoms near the ionization threshold, that is, the reactions, where the transferred energy turned out to be close to the potential of single ionization of the helium atom $I_p = 24.6$ eV. As a result, a noticeable difference was found between the cross sections calculated for various initial and final wave functions of the atom and the cross sections measured in various kinematic domains of angles and energies [1].

A theoretical description of Compton scattering at free electrons based on the concept of photon as a relativistic particle was put forward independently by A. Compton [2] and P. Debye [3] almost a hundred years ago, but this description completely ignored the effects of electron binding in atoms. These effects were first taken into account by J. Dumont [4]. Shortly after quantum mechanics arose and the structure of atoms was described in its framework, based on the results of his experiments, he assumed that, with the help of Compton scattering, one can study the structure of the atoms of the scatterer. He connected the momentum distribution of the electrons bound in the material of a scatterer with the broadening of the photon energy spectrum observed at a fixed scattering angle. In particular, he considered several trial momentum distributions for different electronic states and found that the structure of the observed spectrum of photons scattered by beryllium atoms can be well fitted theoretically with the help of quantum mechanical description of bound electrons in atoms.

Since the time of Compton's research, the experiments in this field have been using a coincidence method for simultaneous detection of the electron produced in the process of ionization and the scattered photon. This method was put forward in 1924 by the German physicists W. Bothe and G. Geiger [5] just for investigating the Compton effect. However, the application of the electron-photon coincidence method to precision measurements is impossible due to a number of technical restrictions. The situation changed after the invention of a new method of registration of scattered particles called the cold target recoil ion momentum spectroscopy (COLTRIMS) [6], which opened up a real opportunity to use Compton scattering for measuring the angular and energy spectra of the scattered photons and the electrons produced in the process of a single ionization. The COLTRIMS method makes it possible to carry out measurements in coincidence with high accuracy by simultaneously measuring the momenta of the electron and recoil ion. In particular, this technique allows one to collect the elec-

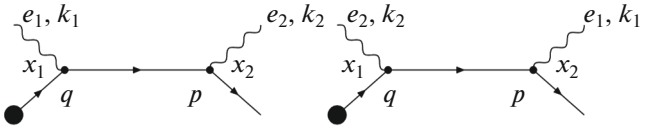

Fig. 1. Feynman diagram of Compton ionization of an atom.

trons and ions from almost the total solid angle $\Omega_{tot} = 4\pi$. In so doing, the momentum of the scattered photon can be found from the law of momentum conservation, as a result of which there is no need to detect the photon. In experiments with atoms, this is often just impossible, since the cross sections are extremely small (about a million times smaller than the typical cross section of photoionization), and their measurement requires a very high synchrotron radiation density.

In quantum electrodynamics, the standard theory of Compton ionization is based on two Feynman diagrams (Fig. 1) [7], but one can also describe this process with the help of nonrelativistic Schrödinger equation [8] for photons with energies of several (and even several tens of) keV. As a result, the matrix element corresponding to these two diagrams can be represented as the sum of two terms. Both these terms are of the second order in the electron charge, but the form of the first term, usually denoted A^2, is similar to the first Born approximation (FBA) for the process of ionization of an atom by a charged particle (proton, electron), whereas the form of the second (integral) term is close to that of the second Born approximation (SBA). For the chosen photon energy, the main contribution is given by FBA, and the second term turns out to be small and plays the role of a correction. A detailed analysis of the contributions of these terms using the hydrogen atom as an example is given in [9].

This theoretical model turned out to be simple enough and allowed us to regard a number of trial functions of initial and final states, to compare the results with the experiment, and to estimate the ability of this new method to perform a precision spectroscopy (angular and energy) of the outer shells of an atom (molecule). Meanwhile, the experiments distinguished between pairs of the trial functions, which demonstrated the possibility of using the Compton ionization along with the well-known spectroscopic methods such as $(e, 2e)$, (p, pe), etc.

Thus, the experimental and theoretical results, recently presented in the journal Nature physics [1], have demonstrated new possibilities of Compton ionization of atoms as an effective method of spectroscopy of the outer shells of atoms and molecules. This has been made possible by precision measurements of very small differential cross-sections using modern devices. Due to this progress, the attempts of the scientists, who pioneered the Compton effect almost a hundred years ago, to use it for the purposes of spectroscopy of quantum objects with the help of their imperfect instruments, received today a new stimulus.

In the paper, the formulas are presented in the atomic units $e = m_e = \hbar = 1$, if it is not specified otherwise.

THEORY

As it is noted in the Introduction, the modern standard theoretical description of Compton scattering at free and bound electron is performed within the framework of QED. However, there is a series of logical pitfalls in the rigorous relativistic approach, namely:

(1) The ion in QED is regarded not as a particle, but as a source of a classical external Coulomb field. Of course, the ion mass $M_{ion} = 4 \times 1836$ a.u. being huge, such a consideration is permissible, but in the COLTRIMS detector, where the ion is registered and its momentum is measured, it moves and behaves like a particle.

(2) The construction of probing wave functions with electron correlations for an atom is extremely problematic.

(3) It is also very difficult to find the Green's function of an electron in the Coulomb field of the ion.

However, traditionally the Compton effect was described by the time-dependent nonrelativistic Schrödinger equation (SH). For photon energies of several tens of keV and low electron energies within a hundred eV this description turns out to be quite acceptable. The ion remains practically at rest during the collision at such energies and acquires a momentum K. The laws of energy-momentum conservation take the form:

$$\omega_1 = \omega_2 + I_p + E_e + E_{ion}, \tag{1.1}$$

$$\mathbf{k}_1 = \mathbf{k}_2 + \mathbf{p} + \mathbf{K}. \tag{1.2}$$

In (1), I_p is the single ionization potential of the atom, $E_e(\mathbf{p})$ is the energy (momentum) of the ejected electron, $E_{ion}(\mathbf{K})$ is the energy (momentum) of the residual ion, $\omega_i(\mathbf{k_i})$ denotes the energy (momentum) of the initial (final) photon. The momentum transfer is denoted by $\mathbf{Q} = \mathbf{k_1} - \mathbf{k_2} = \mathbf{p} + \mathbf{K}$.

The SH for the helium atom with a vector potential of electromagnetic field **A** looks like:

$$\begin{aligned} & i\frac{\partial}{\partial t}\Psi(\mathbf{r_1},\mathbf{r_2},\mathbf{r_n},t) \\ &= \left[\frac{1}{2}\left(-i\nabla_1 - \frac{1}{c}\mathbf{A}(\mathbf{r_1},t)\right)^2 + \frac{1}{2}\left(-i\nabla_2 - \frac{1}{c}\mathbf{A}(\mathbf{r_2},t)\right)^2\right. \\ &\quad + \frac{1}{8M}\left(-i\nabla_n + \frac{1}{c}\mathbf{A}(\mathbf{r_n},t)\right)^2 \\ &\quad \left. - \frac{2}{|\mathbf{r_n}-\mathbf{r_1}|} - \frac{2}{|\mathbf{r_n}-\mathbf{r_2}|} + \frac{1}{|\mathbf{r_1}-\mathbf{r_2}|}\right]\Psi(\mathbf{r_1},\mathbf{r_2},\mathbf{r_n},t). \end{aligned} \quad (2)$$

In (2) $M = 1836$ MeV is the proton mass, $\mathbf{r_n}$ is the coordinate of the nucleus, and $\mathbf{r}_{1,2}$ stand for the coordinates of the electrons. The vector potential is defined as follows:

$$\begin{aligned} \frac{1}{c}\mathbf{A}(\mathbf{r},t) &= \sqrt{\frac{2\pi}{\omega_1}}\mathbf{e_1}e^{i(\mathbf{k_1}\mathbf{r}-\omega_1 t)} \\ &+ \sqrt{\frac{2\pi}{\omega_2}}\mathbf{e_2}e^{-i(\mathbf{k_2}\mathbf{r}-\omega_2 t)} + c.c.. \end{aligned} \quad (3)$$

Here $\mathbf{e_1}(\mathbf{e_2})$ denotes the linear polarization of the initial (final) photon. This choice of the vector potential implies the normalization of the photon wave function to one photon per unit volume, which allows one to describe processes with one absorbed and one emitted photon. We remind that $(\mathbf{k_i}\cdot\mathbf{e_i}) = 0$, and the relation $\mathrm{div}\mathbf{A}(\mathbf{r},t) = 0$ is valid, which imposes the Coulomb gauge on the electromagnetic field.

The interaction term of an electron and the field is written in the form:

$$\begin{aligned} V_{\mathrm{int}} &= i\frac{1}{c}(\mathbf{A}(\mathbf{r},t)\cdot\nabla_{\mathbf{r}}) + \frac{1}{2c^2}A^2(\mathbf{r},t) \\ &= i\left(\sqrt{\frac{2\pi}{\omega_1}}e^{i(\mathbf{k_1}\mathbf{r}-\omega_1 t)}(\mathbf{e_1}\cdot\nabla_{\mathbf{r}}) + \sqrt{\frac{2\pi}{\omega_2}}e^{-i(\mathbf{k_2}\mathbf{r}-\omega_2 t)}(\mathbf{e_2}\cdot\nabla_r)\right) \\ &\quad + \left(\frac{\pi}{\omega_1}(1+e^{2i(\mathbf{k_1}\mathbf{r}-\omega_1 t)}) + \frac{\pi}{\omega_2}(1+e^{-2i(\mathbf{k_2}\mathbf{r}-\omega_2 t)})\right) \\ &\quad + \left(\frac{2\pi}{\sqrt{\omega_1\omega_2}}(\mathbf{e_1}\cdot\mathbf{e_2})e^{i[(\mathbf{k_1}-\mathbf{k_2})\mathbf{r}-(\omega_1-\omega_2)t]}\right) + c.c. \end{aligned} \quad (4)$$

The last term in the big round brackets is the well-known interaction of Kramers–Heisenberg–Waller [10, 11], which defines the so-called A^2 approximation in the theory of Compton scattering. The terms in the first brackets describe a successive absorption and emission of the photon by an electron, which assumes the intermediate Green's function of the atom in the corresponding matrix element. These interaction terms have been studied in more detail in our recent paper [9] and will not be considered here, because they are rather small for a relatively high photon energy.

In the following calculations of the ionization cross section, performing the standard operations and omitting the details:

(1) We disregard the interaction of the nucleus with the electromagnetic field, because it is inversely proportional to the mass of the nucleus (which is also in accordance with the relativistic consideration);

(2) We integrate the matrix element with respect to the time t and the coordinate of the ion r_n, which now, in the absence of the interaction, determine the uniform motion of the atom giving the corresponding delta functions of conservation of energy and momentum;

(3) We perform further integrations, which remove the delta functions.

As a result, we obtain the full differential cross section (FDCS) of the single ionization of a helium atom by Compton scattering

$$\frac{d^3\sigma}{dE_e d\Omega_e d\Omega_1} = \frac{\alpha^4}{(2\pi)^3}p\left(1 - \frac{E_e + I_p}{\omega_1}\right)\frac{1}{2}\sum_{e_1,e_2}|M|^2. \quad (5)$$

The sum in (5) means the averaging over the polarization $\mathbf{e_1}$ of the incoming photon and the summation over the polarization $\mathbf{e_2}$ of the outgoing photon. The matrix element takes the form

$$M(\mathbf{Q},\mathbf{p}) = (\mathbf{e_1}\cdot\mathbf{e_2})\left\langle \Phi_f^-(p)\left|\sum_{j=1}^{2}e^{i\mathbf{Q}\mathbf{r}_j}\right|\Phi_0\right\rangle. \quad (6)$$

It should be noted that the initial $\langle \mathbf{r_1},\mathbf{r_2}|\Phi_0\rangle$ and final $\langle \Phi_f^-(\mathbf{p})|\mathbf{r_1},\mathbf{r_2}\rangle$ wave functions of the atom must be orthogonal, i.e. $\langle \Phi_f^-(\mathbf{p})|\Phi_0\rangle = 0$, or $M(0,\mathbf{p}) = 0$. If these functions are trial functions and not eigenfunctions of the Hamiltonian of the helium atom, they should be orthogonalized.

In (6), the initial state of the helium atom $\Phi_0(\mathbf{r_1},\mathbf{r_2})$ is a symmetric trial function, which may include electronic correlations of various degrees, and the final function is chosen in its asymptotic form

$$\begin{aligned} \Phi_f^{-*}(\mathbf{p};\mathbf{r_1},\mathbf{r_2}) &= \frac{1}{\sqrt{2}}[\varphi^{(*-)}(\mathbf{p},\mathbf{r_1};Z)\varphi_0^{He+}(r_2) \\ &+ \varphi^{(*-)}(\mathbf{p},\mathbf{r_2};Z)\varphi_0^{He+}(r_1)], \end{aligned} \quad (7)$$

where

$$\begin{aligned} \varphi_0^{He+}(r) &= \sqrt{\frac{8}{\pi}}e^{-2r}, \quad \varphi^{(*-)}(\mathbf{p},\mathbf{r};Z) \\ &= e^{-\pi\zeta/2}\Gamma(1+i\zeta)e^{-i\mathbf{p}\cdot\mathbf{r}}{}_1F_1[-i\zeta,1;i(pr+\mathbf{p}\cdot\mathbf{r})]. \end{aligned}$$

The Coulomb function $\varphi^{(*-)}(\mathbf{p},\mathbf{r};Z)$ depends on the effective charge $Z = -p\zeta$ of the ion, which acts on the electron when it leaves the atom. Inside the atom the effective charge depends on the radius r, and asymptotically $Z = 1$.

Let us now turn to the matrix element in (6). It has been obtained as the term of the second order in the A^2

model. However, in the scattering theory of charged particles (electrons, protons) by atoms, this matrix element can be regarded as a plane-wave first Born approximation (PWFBA). When calculating the matrix element in (6) and taking symmetrization into account, we group the terms in (7) in such a way that we link the variable $\mathbf{p}$ to the variable $\mathbf{r_1}$. As a consequence, we get

$$\frac{1}{2}\sum_{e_1,e_s}|M|^2 = (1+\cos^2\theta)|T_1+T_2-2\langle\Phi_0|e^{i\mathbf{Q}\cdot\mathbf{r}}|\Phi_0\rangle T_3|^2, \quad (8)$$

where

$$T_1(\mathbf{p},\mathbf{Q}) = \langle\tilde{\Phi}^-(\mathbf{p})|e^{i\mathbf{Q}\cdot\mathbf{r}_1}|\Phi_0\rangle;$$
$$T_2(\mathbf{p},\mathbf{Q}) = \langle\tilde{\Phi}^-(\mathbf{p})|e^{i\mathbf{Q}\cdot\mathbf{r}_2}|\Phi_0\rangle; \quad (9)$$
$$T_3(\mathbf{p},\mathbf{Q}) = \langle\tilde{\Phi}^-(\mathbf{p})\mid\Phi_0\rangle.$$

The tilde in (9) means that the wave function is no longer symmetric. Since in the atomic units $k=\omega/c\,(c=137)$, $Q=k_1\sqrt{1-2t\cos\theta+t^2}$, and θ denotes the scattering angle photon, i.e. the angle between the vectors $\mathbf{k_1}$ and $\mathbf{k_2}$. The variable

$$t=\left(1-\frac{I_p+E_e}{\omega_1}\right),$$

is, in fact, the ratio of the energy of the final photon and that of the initial photon, $t=\omega_2/\omega_1$.

The first term T_1 in sum (8) corresponds to the process, where electron 1 (active) absorbs and emits a photon, gets a momentum transfer $\mathbf{Q}$ and leaves the atom. The term depends on the momentum $\mathbf{q}=\mathbf{p}-\mathbf{Q}$, because the momentum $\mathbf{p}$ is subtracted from the momentum $\mathbf{Q}$ in the exponential. Since $\mathbf{q}+\mathbf{K}=0$, this momentum is the momentum of the bound active electron in the atom initially at rest. This is the direct process, which plays the most important role in the reactions of scanning the momentum distributions in atoms, such as $(e,2e)$ [12, 13]. The main contribution to the integral defining the term T_1 does not depend on the magnitude of the vectors $\mathbf{p}$ and $\mathbf{Q}$, but only on their difference. Of course, the active electron is described by a Coulomb wave, but it rapidly goes to the plane wave with increasing electron momentum p, which enhances even more the effect of the term T_1 as a spectroscopic tool.

The summand T_2 in sum (8) describes a different physical process. Passive electron 2 absorbs a photon and passes its energy to the electron 1 and remains in the atom, whereas electron 1 leaves the atom. Such a process would be just impossible, if the electrons in the atom were not somehow correlated. Therefore, this matrix element decreases rapidly with the growth of momenta Q and p and its negative effect levels out.

The third term T_3 appears as a result of orthogonalizing the initial and final states and also rapidly decreases.

Thus, the larger the values of Q and p, the better the Compton ionization works as a method for studying mometum distribution of the active electron in an atom. Let us estimate, at what energies of the initial photon and the scattering angles of the final one it is possible. At relatively small energy of the emitted electron $t \lesssim 1$. Let the photon energy be 10 keV. Then $\omega_1 = 10\,000/27.2 = 367.6$ a.u. and $k_1 = 2.68$. We assume the condition $Q \sim p$ (the so-called the Bethe ridge). Then $E_e \sim k_1^2/2(1-2t\cos\theta+t^2)$ and $k_1^2/2 = 73$ eV. The mometum transfer reaches its maximum for backscattering, i.e. $Q_{max} = k_1(1+t)$. Thus, $0 < E_e \lesssim 300$ eV, and the larger energies of the emitted electrons are reached, when the photons are scattered into the backward cone. Note that $t \sim 1$ with a high accuracy.

Hence, we come to the corollary that the larger the cutoff angle θ_0 of the scattered photons, outside of which we collect statistics of events in coincidence, the closer the total amplitude in (8) is to the term T_1, which gives information of spectroscopic value. Some calculations and estimates are presented below.

CALCULATIONS AND DISCUSSION

Although it is possible to obtain valuable information from the measurements in coincidence, cross sections (6) are still very small. For this reason, one has to measure various integral cross sections, from which useful information about the structure of an atomic target can also be extracted. However, here we are interested in studying the role of various terms in expression (8). For this purpose, we calculate the single differential section

$$\mathrm{SDCS}_p = \frac{d\sigma}{dE_e} = 2\pi\int_0^\pi \sin\chi d\chi \int_0^{2\pi} d\phi \int_{\theta_0}^{\pi} \sin\theta\, d\theta\, 3\mathrm{DCS} \quad (10)$$

in the electron energy range $0 < E_e < 150$ eV for four cutoff angles $\theta_0 = 0, 20°, 30°, 90°$. The simplest Hylleraas model of the wave function of helium atom

$$\Phi_0(\mathbf{r_1},\mathbf{r_2}) = \phi(r_1)\phi(r_2), \quad \phi(r) = \sqrt{\frac{Z^3}{\pi}}e^{-Zr}, \quad Z = 27/16$$

is used for the estimates. Each of the four plots in Fig. 2 shows the contribution of only the T_1 term (solid curves) and the sum of all three terms in (8).

As it has been predicted, the dashed curve approaches the solid curve for T_1 with increasing elec-

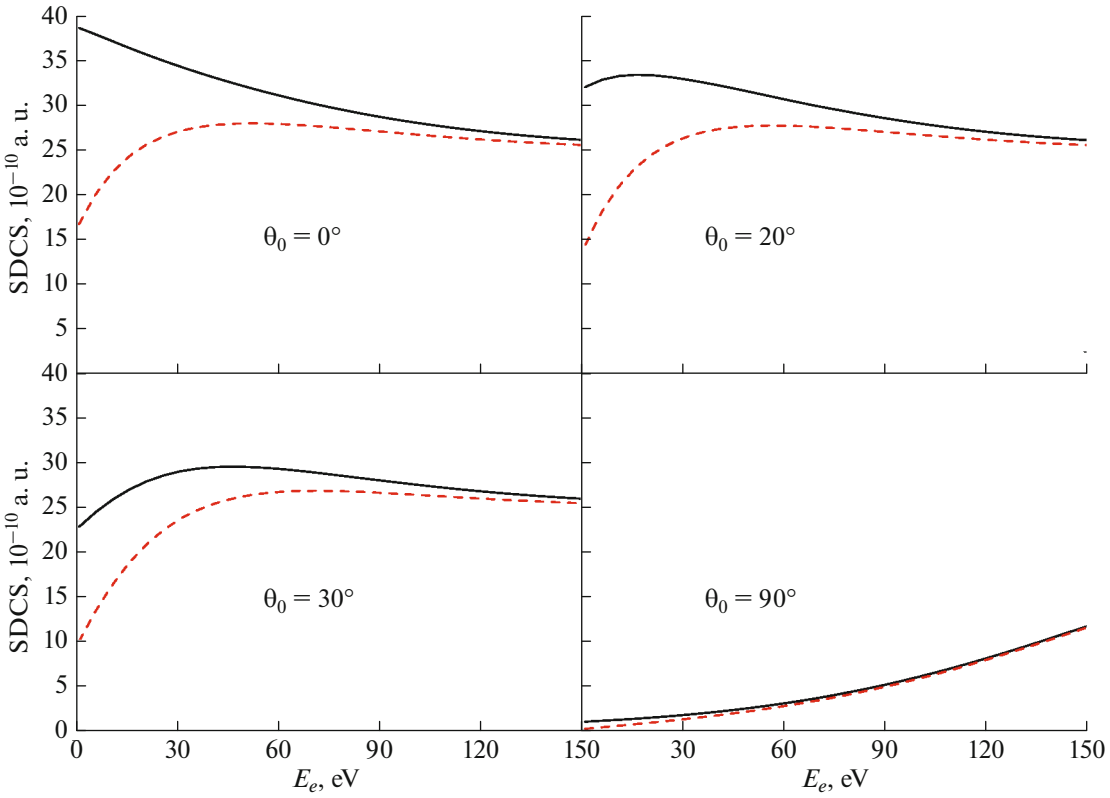

Fig. 2. 3DCS as a function of the energy E_e of the emitted electron. The photon energy is $\omega = 10$ keV. The cutoff angles θ_0 are shown in figures. The solid curve is calculated only with the term T_1, the dashed curve represents the sum of all terms in (8).

tron energy and the growth of the momentum transfer. Surely, a balance is needed between realizing a small contributions of the undesirable terms T_2 and T_3 and the accumulation of statistics. Figure 2 shows that at $\theta_0 = 90°$ the cross section seems to be small, although it is not so. It grows and reaches its maximum in the electron energy range close to $E_e \sim Q^2/2$. In order to make estimating the cross-section value more convenient, here we convert the atomic units to the more usual ones: 1 a.u. $= 10^6$ barn/(eV sr^2).

Finally, let us consider one of the possibilities of using the differential cross section 3DCS for the purposes of a direct study of the momentum distribution of the active electron in a target. If we replace the Coulomb wave by the plane wave in the matrix element T_1 in the domain of energies and angles, where the terms T_2 and T_3 vanish, we obtain the direct Fourier transform of the one-electron wave function $\phi(r)$. To investigate the question of "transformation" of the Coulomb wave into a plane one, we select the kinematics, where the vector $\mathbf{p}$ is collinear to the vector $\mathbf{Q}$. Then $|\mathbf{Q} - \mathbf{p}| = |Q - p| = q$. The momentum q determines just the momentum of the electron "inside" the atom. Let us consider at once the case of the boundary value of the momentum transfer, which can be achieved in this setting, i.e. put $\theta = 180°$, $Q = 2k_1$. The minimum value $q = 0$ is attained for the electron energy $E_e \approx 390$ eV. For comparison, in Fig. 3 the momentum distribution $A/[(Q - p)^2 + Z^2]^4$ is shown, which is normalized to the solid curve at its "tails". It can be seen from the Figure that even the peak of the cross section 3DCS$/p$ does not reach the weakly pronounced peak of the plane-wave momentum distribution at $E_e \approx 390$ eV. The comparison is even worse for smaller fixed scattering angles. This means that even larger values of the momentum transfer are needed, i.e., even larger photon energies.

Fortunately, unlike the (e, 2e) reaction, where the momentum transfer Q^4 is in the denominator of the expression for the cross section, because the interaction is mediated by a virtual photon (Coulomb potential), there is no such thing here, since the interaction occurs with a real photon, and we can increase both Q and E_e. However, these options are hindered by the capabilities of experimental detector devices.

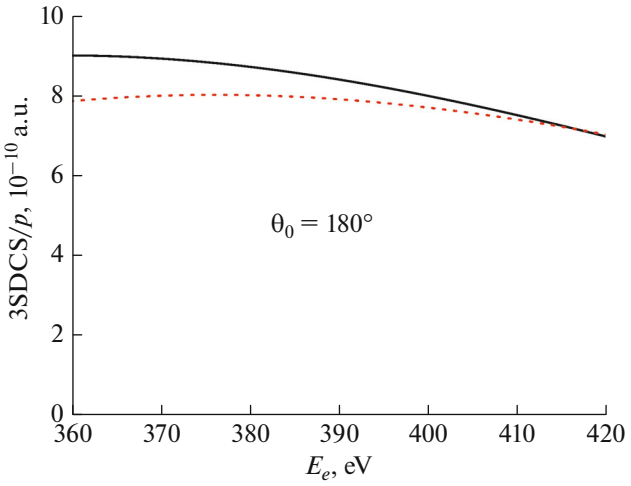

Fig. 3. 3DCS/p as a function of the energy E_e of the emitted electron under the condition of the vectors **p** and **Q** being collinear. The photon energy is $\omega = 10$ keV. The scattering angle is $\theta = 180°$. The solid curve is T_1 only, the dashed curve is the reference function $A/[|Q - p|^4 + Z^2]^4$ normalized to the tails of the energy spectrum.

CONCLUSIONS

In this paper, we have shown that the process of single Compton ionization of an atomic target has a good capability to be used as a tool for studies of the momentum distribution of an active electron in the target. This becomes possible at large momenta of the emitted electron and large momentum transfer Q, when the contribution of the "unwanted" terms T_2 and T_3 becomes small. In this process the variable $q = |\mathbf{Q} - \mathbf{p}|$ is just equivalent to the momentum of the active electron inside the target before its interaction with the photon. Hence, this variable should be small and can be varied in a required range by changing the angles and magnitudes of the vectors Q and p.

With the current level of experimental devices, the measurement of very small cross sections of Compton ionization is not a fantasy, and therefore Compton ionization may well compete with other known methods of atomic spectroscopy, in particular, $(e, 2e)$ in the EMS kinematics.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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