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Back-to-back emission of the electrons in double photoionization of helium

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We calculate the double differential distributions and distributions in recoil momenta for the high energy non-relativistic double photoionization of helium atom. We show that the results of recent experiments provide the pioneering experimental manifestation of the quasifree mechanism for the double photoionization which was predicted long ago in our papers. This mechanism provides a surplus in distribution over the recoil momenta at small values of the latter, corresponding to nearly "back-to-back" emission of the photoelectrons. Also, in agreement with previous analysis we demonstrate that this surplus is due to the quadrupole terms of the photon-electron interaction. We present the characteristic angular distribution for the "back-to-back" electron emission. The confirmation of the quasifree mechanism existence opens a new area for exciting experiments, which are expected to increase our understanding of the electron dynamics and of the bound states structure.

1. Introduction. In this Letter we calculate the recoil momentum distribution of the helium double photoionization cross-section in the high photon energy non-relativistic limit. We calculate also the energy distribution in the "back-to-back" configuration of the emitted electrons. Our results are in qualitative agreement with the data of recent experiments on the helium double ionization by photons with energies 800 and 900 eV [1, 2] that provide information on the cross-section dependence upon recoil momenta q of the nucleus. Although in [1, 2] quantitative results are not presented, the experiments demonstrate definitely that the distribution of outgoing electrons has a prominent enhancement at small q of about 2 arb. units. The kinematics of these experiments enables to separate the non-dipole contributions at small values of q. Thus, the observed enhancement is entirely due to the non-dipole terms. Therefore, we conclude, as did the authors of experiments [1, 2], that it demonstrates for the first time the existence of the *quasifree mechanism* (QFM) of the double photoionization, which was predicted many years ago [3].

By that time only two mechanisms of the process were known. In both of them the electron that directly interacts with the photon obtains almost all the incoming photon energy ω. In the first mechanism, named *shake-off*, the second electron is ionized due to instant change of acting upon it effective field. In the second, called *knock-out* mechanism, the fast photoelectron inelastically collides with the bound one, transferring to it a small part of own energy. The two mechanisms could be clearly separated in the case of high photon energies

$$\omega \gg I, \tag{1}$$

with I denoting the single-particle binding energy, since the interactions between the photoelectron and the bound one can be neglected in the shake-off mechanism. Note that both mechanisms include the single photoionization as the first step.

The key point of the third mechanism, predicted in [3], is that the two electrons can absorb a photon almost without participation of the nucleus. Such electrons leave the atom in opposite directions with the same energies, i.e. "back-to-back". Neither the shake-off nor the knock-out mechanisms contribute to this kinematics region since the single photoionization is not allowed for free electrons and thus in ionization, caused by a photon carrying the energy ω, momentum $q = (2\omega)^{1/2}$ (in atomic system of units $e = \hbar = m$, used in this paper) should be transferred to the nucleus.

We emphasize that the peak of the distribution in recoil momentum of the double photoionization cross-section at $q = 0$ observed in [1, 2] is a very important qualitative result, proving the existence of QFM. Two other mechanisms provide only one peak at large values of q.

Due to the peak of the distribution $d\sigma/d\varepsilon dq^2$ QFM has several specific features. The common view before prediction of QFM and decades after was that the photoelectrons energy spectrum curve has **U** shape with high maxima at the edge regions of the spectra. QFM predicted a local maximum at the center of the energy distribution that leads to **W** shape. Another feature of QFM is that its contribution decreases with photon energy slower than the contributions of two other mechanisms. Thus, the account of QFM leads to qualitative change in the high energy non-relativistic asymptotic of the double-to single photoionization cross sections ratio. It is important that QFM requires going beyond the dipole approximation, since there is no dipole moment of the two-electron system at $\mathbf{q} = 0$ (see [4] for more details).

For a long time it was beyond the experimental abilities to find a manifestation of QFM in experiments. However, some attempts were undertaken to check the QFM existence performing the numerical calculations of the double photoionization cross-section. Unfortunately, they failed to confirm the presence of QFM. Later it was understood [5] that QFM is extremely sensitive to the analytical properties of the initial state wave functions. In particular, it cannot be reproduced using uncorrelated initial state electron wave functions, which were used in the calculations mentioned above.

Some of the calculations lead to the **W** shape of the spectrum (see, e.g. [6]) even in the dipole approximation. It was shown, however, in [7] that the central peak was spurious, being entirely a consequence of oversimplified approximations for the wave functions of either initial or the final states. The consistent approach provided cancelation of spurious terms and restoration of the **U** shape of the spectrum in the dipole approximation.

Several recent calculations beyond the dipole approximation [8, 9] confirmed the increase of the differential cross sections at the center of the energy distribution, thus supporting QFM.

2. The quasifree mechanism. In the single-electron ionization by photons carrying energies, which satisfy the condition $\omega \gg I$, the recoil momentum q essentially exceeds the characteristic binding momentum $\eta = (2I)^{1/2}$ of the atomic electron. Therefore, this process is impossible without participation of the nucleus.

However, in the double photoionization there is a kinematical region, where the process on the free electrons *can take place.* Following the general analysis of Bethe [10], one can expect the increase of the differential cross section of a high energy process in this region. This happens because the recoil momentum can be made small ($q \sim \eta$), and the process takes place at the distances of the order $r_b \sim 1/\eta$, just the very distances from the nucleus, where the bound electrons are mainly localized. Each act of transferring of a larger momentum requires approaching the nucleus at somewhat smaller distances where the electron density is smaller. This leads to a smaller value of the amplitude.

The ideas developed in [10] were employed in investigations of the high energy electron scattering [11] and of other high energy atomic processes [4, 12]. They were used in analysis of the double photoionization in [3].

Let us find the kinematical region where double photoionization on the free electrons is possible. In the rest frame of the initial atom the recoil momentum is

$$\mathbf{q} = \mathbf{k} - \mathbf{p}_1 - \mathbf{p}_2. \tag{2}$$

Here $\mathbf{k}$ is the photon momentum, $\mathbf{p}_i$ are the momenta of the outgoing electrons. The corresponding energies are $\omega = ck$, and $\varepsilon_i = p_i^2/2$ ($p_i = |\mathbf{p}_i|$), $\varepsilon_1 + \varepsilon_2 = \omega - I$; c is the speed of light. Thus, $p_i \gg k$. For the process on the free electrons

$$\mathbf{q} = 0. \tag{3}$$

This condition can be correct only if the large momenta $\mathbf{p}_i$ almost compensate each other. Hence, the photoelectrons are emitted mostly "back-to-back", with $t \equiv (\mathbf{p}_1 \cdot \mathbf{p}_2)/p_1 p_2$ close to -1, while the values $p_1 \approx p_2$, i.e. $|p_1 - p_2| \ll p_{1,2}$. Combining Eqs. (2) and (3) we find $|p_1 - p_2| < k$, i.e.

$$\beta \equiv |\varepsilon_1 - \varepsilon_2|/E \le \sqrt{2\omega/c^2}; \quad E = \varepsilon_1 + \varepsilon_2, \tag{4}$$

with $E = \varepsilon_1 + \varepsilon_2$ being the total energy carried away by the photoelectrons. While we consider the case of the nonrelativistic photoelectrons, we have $\sqrt{\omega/c^2} \ll 1$, and thus, $\beta \ll 1$.

In double photoionization of the atom we find the increase of the amplitude at the recoil momentum $q \ll p_i$. In the beginning of this section we presented qualitative arguments [10] that momenta $q \le \eta$ become important. If $k \ll \eta$ (in the case of atomic helium this means that $\omega \ll 6\,\text{KeV}$) the condition $q \le \eta$, which determines the quasifree kinematics, requires that

$$\beta \le \sqrt{2I/E} \ll 1. \tag{5}$$

As we saw earlier, there is no dipole contribution in exactly free kinematics with $q = 0$. The process on the free electrons is caused by higher multipole terms with the quadrupole term providing the leading contribution. Of course, in the quasifree kinematics there is a non-vanishing dipole term in the amplitude, but it contains a small factor $(\mathbf{e}\cdot\mathbf{q})$. Therefore, it is strongly suppressed [5], and the quadrupole terms dominate for $\omega \ge 800\,\text{eV}$. Note that in experiment described in [1] they detect the recoil ions moving perpendicular to the polarization direction. This completely eliminates the contribution of the dipole terms. Thus, only the QFM contribution provides a strong maximum in distributions over the recoil momenta, corresponding to emission of "back-to-back" electrons.

In QFM, two bound electrons have to exchange large momenta in the initial state. Thus, they have to approach each other at small distances $r_{12} \ll r_b$, while their distances from the nucleus are still of the Bohr radius r_b order. Hence, it is reasonable to attribute the QFM amplitude to the properties of the initial state wave function $\Psi(r_1, r_2, r_{12})$ at $r_{12} = 0$. It was shown in [7] that the amplitude contains the factor $\partial\Psi/\partial r_{12}$ at $r_{12} = 0$, which is connected to the function $\Psi(r, r, 0)$ by the cusp condition [13]:

$$\Psi(r, r, 0) = 2\partial\Psi/\partial r_{12}|_{r_{12}=0}. \tag{6}$$

Now let us illustrate these statements in the framework of a simplified model, in which we neglect all interactions of the outgoing electrons. Such approach is justified since interactions between the outgoing electrons and nucleus are defined by their Sommerfeld parameters $\xi_i = Z/p_i$ [14]. Since both $\varepsilon_i \gg I$, we find $\xi_i \ll 1$, and the amplitude can be expanded in powers of ξ_i with the interaction neglected in the lowest term. The amplitude M can be expressed in terms of the initial wave function Ψ_i in momentum presentation

$$M = (4\pi/c)^{1/2}[(\mathbf{e}\cdot\mathbf{p}_1)\Psi_i(\mathbf{p}_1, \mathbf{q}-\mathbf{p}_1) + \\ + (\mathbf{e}\cdot\mathbf{p}_2)\Psi_i(\mathbf{p}_2, \mathbf{q}-\mathbf{p}_2)]. \tag{7}$$

Applying the Lippman–Schwinger equation to the function $\Psi_i(\mathbf{p}, \mathbf{q}-\mathbf{p})$ at $p \gg \eta$, $q \ll p$ we obtain for the leading term of expansion in powers of $1/p^2$ [12]

$$\Psi_i(\mathbf{p}, \mathbf{q}-\mathbf{p}) = \frac{8\pi c}{p^4} S(q^2), \tag{8}$$

with

$$S(q^2) = \int \frac{d^3 f}{(2\pi)^3} \Psi_i(\mathbf{f}, \mathbf{q}-\mathbf{f}) = \\ = \int d^3\mathbf{r}\Psi(r, r, 0)\exp[-i(\mathbf{q}\cdot\mathbf{r})]. \tag{9}$$

Thus, the amplitude is suppressed at $q \gg r_b^{-1} \sim \eta$. Treating the interaction between the bound electrons in the lowest order of perturbation theory, we must put $\Psi(r, r, 0) = \varphi_C^2(r)$, with $\varphi_C(r)$ being the single-particle Coulomb wave function for the electron in $1s$-state. This leads to $S(q^2) = 16\eta^4/(q^2 + 4\eta^2)^2$. Here $\eta = Z$, while Z is the charge of the nucleus. Thus, indeed the distribution in recoil momentum q at $q \ll p_i$ is enhanced at $q \sim \eta$. As it is demonstrated by estimations, the region of distribution enhancement $q \sim \eta$ remains the same, even when the final state interaction is taken into account.

3. Distribution in recoil momentum. If interaction between photoelectrons is neglected, we obtain

$$\frac{d^2\sigma}{dq^2 d\varepsilon_1} = \frac{128}{15}\frac{\omega c^3}{E^4} S^2(q^2), \tag{10}$$

with the function $S(q^2)$ defined by Eqs. (8) and (9).

In [15, 16] we obtained the analytical expressions which approximate very precise wave functions [17] at $r_{12} = 0$. They approximate the improved wave functions obtained in [18], as well. In the simplest case [15]

$$\Psi(r, r, 0) = \Psi(0, 0, 0)\exp(-2Zr), \tag{11}$$

with Z being the charge of the nucleus. For the functions obtained in [17, 18] $\Psi(0, 0, 0) \approx 1.37$. We find immediately

$$S(q^2) \approx \frac{22\pi\eta}{(q^2 + 4\eta^2)^2}, \tag{12}$$

with $\eta = Z$.

To obtain the distribution $d\sigma/dq^2$, one should integrate the distribution (6)–(10) over ε_1, having in mind that $q \geq |p_1 - p_2|$. In actual calculations we employ the combination of two exponential terms [16], which gives a very accurate approximation of the exact wave function at the electron-electron coalescence line.

For the photon energies of about 1 KeV employed in the experiments [1, 2] parameters $\xi_i \approx 1/3$. Thus, it is desirable to avoid expansions in ξ_i, taking into account interaction with the nucleus. In this case the factor $\exp[-i(\mathbf{q}\cdot\mathbf{r})]$ in the integrand of Eqs. (8) and (9) should be replaced by the product of two continuum Coulomb functions. The integral can be evaluated analytically by employing the technique, developed in [19]. Finally, we obtain

$$\frac{d^2\sigma}{dq^2 d\varepsilon_1} = \frac{128}{15}\frac{\omega c^3}{E^4} S^2(q^2) F(\xi_i, q^2). \tag{13}$$

The function F with $F(\xi_1 = 0, \xi_2 = 0, q^2) = 1$ has a simple analytical form. However, the expression is too cumbersome to be displayed in the Letter. Note that the main q^2 dependence is contained in the factor $S^2(q^2)$ with $S(q^2)$ given by Eqs. (8) and (9).

These equations enable us to obtain the angular distribution at the point of exactly "back-to-back" emission by presenting

$$\frac{d^2\sigma}{dt d\varepsilon_1} = 2p_1 p_2 \frac{d^2\sigma}{dq^2 d\varepsilon_1}. \tag{14}$$

We calculate the distributions $d^2\sigma/dtd\varepsilon_1$ and $d\sigma/dt$ at the point of exactly "back-to-back" emission $t = -1$. In Fig. 1 we provide example of the distribution $d^2\sigma/dtd\varepsilon_1$

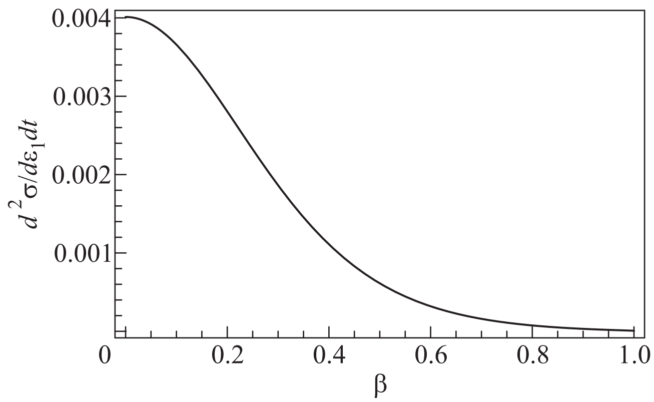

Fig. 1. Energy distribution for the "back-to-back" emission ($t = -1$) presented by Eq. (14) for $\omega = 900$ eV considered in [2]. The value $d^2\sigma/d\varepsilon_1 dt$ is given in barn/eV

for the energy $\omega = 900$ eV in [2]. One can see that the main contribution to $d\sigma/dt$ comes from $\beta \leq 0.3$, in agreement with Eq. (4). In Fig. 2 we present the dependence of the distribution $d\sigma/dt$ upon the photon energy in the region near 1 keV. At $\omega = 900$ eV we find $d\sigma/dt = 0.52$ barn. Since the important interval of t is $I/\omega \approx 0.06$ the contribution to the total cross section is 0.03 barn in agreement with [20].

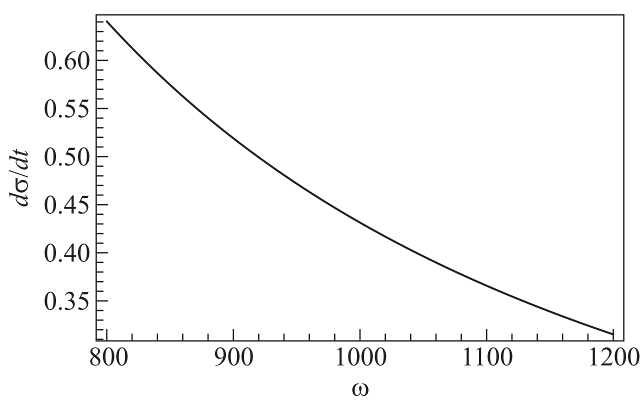

Fig. 2. Dependence of the differential distribution $d\sigma/dt$ at $t = -1$ on the photon energy in keV region. The value of $d\sigma/dt$ is given in barns

4. Summary. The recent experiments [1, 2] demonstrated that the distribution of the double photoionization in recoil momentum has an excess at small q. In this Letter we show that it comes from the quadrupole terms of the electron-photon interaction. Recall that the two other mechanisms, i.e. the shake off and the knock-out contain a single-electron photoionization as the first step. Thus, they require large recoil momentum and do not contribute at small q. Hence, the results of the experiments [1, 2] confirm the existence of quasifree mechanism predicted long ago [3]. At the time of publication of [3] experimental detection of QFM was beyond the experimental abilities. After the latter have been developed (mainly due to invention of the recoil momentum spectroscopy), the interest to the mechanism increased strongly, and its understanding has been improved [4, 5, 16].

In this Letter, we calculated the distributions $d^2\sigma/dq^2d\varepsilon_1$ and $d\sigma/dq^2$ for the non-relativistic high energy double photoionization. In the region $q \ll p_i$, the distributions have local peaks at $q^2 = 0$ with the width of the order η^2. Such data were obtained in the experiments [1, 2]. Thus, our results are consistent with those of [1, 2]. Unfortunately, there are no quantitative data in [1, 2]. Hence, as it stands now, the calculated and measured values cannot be compared directly.

The advent of new powerful light sources provides additional possibilities in experimental studies of photoabsorbtion processes [21]. In particular, more detailed investigation of photoionization by the photons caring the energies ω of about 1 KeV becomes possible. Therefore, we can expect more detailed measurements of the electron distributions of the double photoionization. In its turn, this will require more detailed calculations. In the present Letter, we found the distributions at $q = 0$. As the next step, one should investigate the shape of the peak calculating the distributions at small but finite values of q. It was demonstrated (see [4]) that QFM changes the shape of the spectrum curve at $\omega \sim 1$ keV. This also stimulates further studies.

The investigation of QFM enables to clarify the behavior of the wave function of the helium atom near the singular electron-electron coalescence line $r_{12} = 0$. Besides the purely theoretical interest, this is important for precise computations of various atomic characteristics. Recall that the proper treatment of the $\Psi(r_1, r_2, r_{12})$ at the three-particle coalescence point $r_1 = r_2,\ r_{12} = 0$ enabled to diminish strongly the number of parameters that determine the bound state wave functions .

QFM becomes more important with the photon energy growth. At $\omega \geq c^2$, the outgoing electrons become relativistic, and their contribution to the total cross section is about the same as that of the shake-off [20]. Therefore, the investigation of the relativistic case is also one of the forthcoming problems. We hope that further research will move into the region of such photon energies and will confirm the predicted fine structure of the central peak of the energy distribution. We believe also that contribution of QFM to the total cross section, resulting in a prominent slope of the double-to-single photoionization ratio will be measured. This is increasingly necessary since there is a large disagreement between the experimental and theoretical data in this case [22].

QFM is also interesting from another point of view. One of the problems studied nowadays is the role of nondipole terms in the nonrelativistic photoionization – see, e.g. [8, 9]. From this point of view, inclusion of the quadrupole terms enables to take into account QFM. In relativistic case, one can analyze the relative role of different multipoles that are strongly influenced by QFM.

In addition, the results of this Letter and of the recent experiments open a new field for studies of two-electron ionization not only by photons but by other projectiles, e.g. by fast electrons or heavy ions.

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1. M. S. Schöffler, T. Jahnke, M. Waitz et al., ICPEAC 2011; http://www.qub.ac.uk/icpeac 2011/.
2. Th. Weber, M. S. Schöffler, A. Belkacem et al., Bull. Amer. Phys. Soc. **56**(5), 144(2011).
3. M. Ya. Amusia, E. G. Drukarev, V. G. Gorshkov, and M. P. Kazachkov, J. Phys. B **8**, 1248 (1975).
4. M. Ya. Amusia, E. G. Drukarev, and V. B. Mandelzweig, Phys. Scr. C **72**, 22 (2005).
5. T. Suric, E. G. Drukarev, and R. H. Pratt, Phys. Rev. A **67**, 022709 (2003).
6. M. A. Kornberg and J. E. Miraglia, Phys. Rev. A **48**, 3714 (1993).
7. E. G. Drukarev, N. B. Avdonina, and R. H. Pratt, J. Phys.B **34**, 1 (2001).
8. J. A. Ludlow, J. Colgan, T. G. Lee et al., J. Phys. B. **42**, 225204 (2009).
9. A. G. Galstyan, O. Chuluunbaatar, Yu. V. Popov, and B. Piraux, Phys. Rev. A **85**, 023418 (2012).
10. H. Bethe, in *Handbuch der Physik*, Springer, Berlin, 1933, v. 24/1.
11. M. Inokuti, Rev. Mod. Phys. **43**, 297 (1971).
12. E. G. Drukarev, Physics-Uspekhi, **50**, 835 (2007).
13. T. Kato, Commun. Pure Appl. Math. **10**, 151 (1957).
14. H. A. Bethe and E. E. Salpiter, *Quantum mechanics of one-and two-electron atoms*, N.Y.: Plenum/Rosetta, 1971.
15. E. G. Drukarev, M. Ya. Amusia, E. Z. Liverts et al., JETP **103**, 690 (2006).
16. E. Z. Liverts, M. Ya. Amusia, R. Krivec, and V. B. Mandelzweig, Phys. Rev. A **73**, 012514 (2006).
17. M. I. Haftel, R. Krivec, and V. B. Mandelzweig, J. Comput. Phys. **123**, 49 (1996).
18. E. Z. Liverts and N. Barnea, Comp. Phys. Com. **182**, 1790 (2011).
19. A. Nordsieck, Phys. Rev.**93**, 785 (1954).
20. E. G. Drukarev, Phys. Rev. A **51**, R2648 (1995).
21. S. W. Epp, J. R. Crespo Lopez-Urrutia, G. Brenner et al., Phys. Rev. Lett. **98**, 183001 (2007).
22. J. Hoszowska, J.-Cl. Dousse, W. Cao et al., Phys. Rev. A **82**, 063408 (2010).