

Shape variation of the two-electron photoionization spectrum with photon energy growth

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We trace the evolution of the shape of the two-electron photoionization spectrum with photon energy growth using quite a precise two-electron wave function, obtained by the correlation function hyperspherical harmonic method. We obtain the values of ω_1 and ω_2 at which the spectrum curve changes its shape. At $\omega=\omega_1$ the U shape changes to a W shape. At $\omega=\omega_2$ the central W peak splits into two. We consider ground states of the helium atom and of heliumlike ions with the nuclear charge Z , the negative ion of hydrogen H^- , and the excited n^1S state of helium. The limiting laws for $Z \gg 1$ and $n \gg 1$ are obtained. The analysis is carried out *without* calculations of the particular energy distributions themselves.

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I. INTRODUCTION

In this paper we analyze the evolution of the shape of the photoelectron energy distribution in double photoionization with the growth of the photon energy ω . We consider a number of systems containing two bound electrons. These are the ground states of the helium atom and of light heliumlike ions, the negative ion of hydrogen H^- , and also the excited states of the helium atom n^1S ($n > 1$).

The qualitative picture of evolution of the photoelectron spectrum curve shape with photon energy growth was presented long ago [1,2]. It was shown that at some values of the photon energies ω exceeding strongly the electron binding energies I , i.e., at $\omega \gg I$, the energy distribution becomes a result of interplay of three distinctive mechanisms. In the well-known shake-off (SO) mechanism [3] one of the electrons is ionized by the direct absorption of the incoming photon, while the second is moved into a continuous spectrum as a result of an instant change of the field acting upon this electron due to the removal of the first electron. The SO mechanism dominates at the edge of the spectrum, where the photon transfers most of its energy $\varepsilon \approx \omega$ to the absorbing electron, while the secondary electron obtains a small amount of energy $\varepsilon \sim I$. If both outgoing electrons are fast, i.e., at $\varepsilon_{1,2} \gg I$, the contribution of SO is strongly suppressed, and the energy distribution is determined by the interplay of two other alternative mechanisms. One of them is the final state scattering (FSS), in which one electron absorbs the incoming photon and then ionizes the second electron after colliding with it. Since $\varepsilon_{1,2} \gg I$, both electrons carry large momenta $p_i = (2m\varepsilon_i)^{1/2} \gg \eta$ with η standing for the characteristic momentum of the bound electron, while m denotes the electron mass. In the FSS mechanism the first electron obtains momentum p_1 from the nucleus, transferring momentum p_2 to the secondary electron in the course of the scattering after photon absorption, i.e., in the final state. In the quasifree mechanism (QFM) the electrons exchange large momenta $p_1 \sim p_2$ without the participation of the nucleus, which is possible only when they are close to each other.

The photoelectron spectrum obtains sharp peaks at the edges of the energy interval $\varepsilon_{1,2} \rightarrow 0$, where the process is dominated by the SO mechanism. At $\varepsilon_{1,2} \gg I$ the energy distribution is a result of interplay of the FSS and QFM. Qualitatively the FSS and SO spectra are similar. So, neglecting the QFM we would obtain a smooth U-shaped spectrum curve, with a minimum at the central point $\varepsilon_1 = \varepsilon_2 = E/2$, with $E = \omega - I^{2+}$ being the total energy available to the outgoing electrons. However, the role of QFM increases with the growth of ω . Since the free process is possible only in the vicinity of the center, QFM provides a central peak for ω exceeding some value ω_1 . The spectrum then acquires a W shape until the photon energy ω reaches a certain value ω_2 . At $\omega = \omega_2$ the central peak splits into two, which are shifted symmetrically from the center. There is a local minimum at the central point $\varepsilon_1 = \varepsilon_2$ at $\omega > \omega_2$. This fine structure of the curve at $\omega > \omega_2$ is due to the quadrupole nature of the QFM [1].

The condition $\omega \gg I$ enables one to obtain explicit expressions for the contributions of the FSS and QFM to the two-electron photoionization energy spectrum by using the perturbative description of the final state wave function [4]. These expressions contain the initial state wave function $\Psi(\mathbf{r}_1, \mathbf{r}_2)$. In the FSS one of the electrons should be close to the nucleus. Thus the amplitude contains a parameter that is an integral of the function $\Psi(0, \mathbf{r})$. In the QFM the electrons should be very close to each other. Thus, the amplitude contains an integral of the function $\Psi(\mathbf{r}, \mathbf{r})$. We find the values of ω_1 and ω_2 without calculating a particular energy distribution. Instead of this, we analyze the second derivative of the photoelectron energy distribution $d\sigma^{2+}/d\varepsilon_1$ with respect to ε_1 , at the central point $\varepsilon_1 = E/2$. This analysis enables us to make some more conclusions about the shape of the spectrum curves.

The magnitude of the QFM contribution is very sensitive to the quality of the approximate wave functions used in the calculations. It was shown in [5,6] that the use of oversimplified wave functions of the initial or final states leads to large but spurious QFM contributions in the framework of

TABLE I. The values of ω_1 , ω_1^* , ω_2 , ω_2^* in keV for the ionization of ground states of the heliumlike systems with the nuclear charge Z . The column with $Z=1$ shows the data for the negative ion of hydrogen H^- .

Z	1	2	3	4	5
ω_1	0.549	1.93	3.70	5.89	8.49
ω_1^*	0.544	1.89	3.57	5.61	7.96
ω_2	3.97	8.89	13.7	18.5	23.3
ω_2^*	3.98	8.90	13.8	18.6	23.5

the dipole approximation, where it has to be zero. Even a consistent treatment of the QFM, but without highly accurate wave functions, results in quantitatively different energy distributions for different types of the initial state wave functions, at least in the case of the helium atom [6].

In this paper we employ very accurate and locally correct wave functions [7,8], obtained by a direct solution of the three-body Schrödinger equation. The wave functions are calculated by the correlation function hyperspherical harmonic method (CFHHM). They have been used for the calculations of various characteristics of two-electron systems, both in their bound- [9] and continuous-spectrum states [10], always demonstrating high accuracy.

For the ground state of the helium atom we find

$$\omega_1 = 1.9 \text{ keV.} \quad (1)$$

We consider also the ground states of the heliumlike ions with the nuclear charge Z . The value of Z should not be too large, i.e., $(\alpha Z)^2 \ll 1$ ($\alpha \approx 1/137$ is the fine structure constant). The latter inequality ensures that the relativistic corrections are small. We find a simple equation for the energy ω_1^* , which approximates the value of ω_1 with the relative accuracy αZ . For large values of Z , $Z \gg 1$, we obtain

$$\omega_1^* = 0.65Z^{14/9} \text{ keV.} \quad (2)$$

The behavior $Z^{14/9}$ can be obtained by using the Coulomb (hydrogenlike) functions.

We show that Eq. (2) holds with 1% accuracy for $Z \geq 3$. We find that the central peak splits into two ones at

$$\omega_2 = 8.9 \text{ keV} \quad (3)$$

in the case of the ground state of helium. For ions with $Z \gg 1$ the analysis carried out with the Coulomb functions predicts that ω_2 increases linearly with Z . This is supported by computations with precise wave functions that yield

$$\omega_2^* = 4.7Z \text{ keV} \quad (4)$$

at $Z \gg 1$.

For the ionization of helium excited n^1S states we found that the values of ω_1 and ω_2 depend upon n very weakly. This is consistent with the general properties of the bound-state wave functions in the limit $n \gg 1$ [11].

Results for small values of Z and n are presented in Tables I and II. The tables also contain the results for the hydrogen negative ion H^- .

TABLE II. The values of ω_1 , ω_1^* , ω_2 , and ω_2^* in keV for the ionization of the excited n^1S states of helium atom.

n	2	3	4
ω_1	2.17	2.22	2.23
ω_1^*	2.28	2.34	2.36
ω_2	12.4	12.7	12.8
ω_2^*	12.4	12.7	12.8

In this paper we employ the system of units with $\hbar=c=1$. Therefore the relation between the energy ω and the linear momentum k of the photon is $\omega=k$. The averaged momentum of the bound K -electron can be estimated by the hydrogenlike value $\eta=m\alpha Z=3.73Z$ keV. For the atomic helium the screened value of the nuclear charge $Z_0=27/16$ [11] corresponds to $\eta=6.3$ keV.

We present the general equations in Sec. II. The evolution of the spectrum curve from the U to the W shape is considered in Sec. III. More complicated structures at larger energies are analyzed in Sec. IV. We summarize our results in Sec. V.

II. GENERAL EQUATIONS

The energy distribution of the fast photoelectrons ($\varepsilon_{1,2} \gg I$) from the double photoionization can be presented as the additive sum of the FSS and QFM contributions, represented by the terms $T_1(\omega, \gamma)$ and $T_2(\omega, \gamma)$, respectively,

$$\frac{d\sigma^{2+}(\omega, \varepsilon)}{d\varepsilon} = T_1(\omega, \gamma) + T_2(\omega, \gamma). \quad (5)$$

Here we introduced

$$\gamma = \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \quad (6)$$

and assumed $\varepsilon_1 \geq \varepsilon_2$. The interference terms are strongly suppressed due to the different angular distributions from the FSS and QFM. The momenta of the outgoing electrons are mostly orthogonal in the case of FSS, having the opposite directions in QFM.

The contribution of the FSS can be presented in a simple way if the outgoing electrons are described by the single-particle functions $\psi_s(\mathbf{r})$. If both electrons belong to the same shell, we find [4,12]

$$T_1(\omega, \gamma) = \frac{\langle r^{-2} \rangle}{4\pi} \sigma^+(\omega) \frac{d\sigma_{ee}(\omega, \gamma)}{d\varepsilon}, \quad (7)$$

with $\langle r^{-2} \rangle = \int d^3\mathbf{r} \psi_s^2(\mathbf{r})/r^2$, while σ^+ and σ_{ee} denote the one-electron photoionization cross section of the shell considered and the free electron-electron scattering cross section in the spin-singlet state, respectively.¹ Using the well-known expression

¹To simplify the notations we set $\varepsilon_1 + \varepsilon_2 = \omega - I^{2+} = \omega$, since $\omega \gg I^{2+}$.

$$\frac{d\sigma_{ee}}{d\varepsilon} = \frac{\pi\alpha^2}{\omega} \left(\frac{1}{\varepsilon} + \frac{1}{\omega - \varepsilon} \right)^2, \quad (8)$$

one obtains

$$T_1(\omega, \gamma) = \frac{4\alpha^2 \langle r^{-2} \rangle \sigma^+(\omega)}{\omega^3 (1 - \gamma^2)^2}. \quad (9)$$

The cross section of the *K*-shell photoionization in the single-particle approximation is [11]

$$\sigma^+(\omega) = |\psi_s(0)|^2 \Lambda(\omega),$$

$$\Lambda(\omega) = \frac{2^{11/2} \pi^2 \alpha (\alpha Z)^2}{3m^5} \left(\frac{m}{\omega} \right)^{7/2}. \quad (10)$$

If the bound two-electron system is described by the correlated wave function $\Psi(\mathbf{r}_1, \mathbf{r}_2)$, the FSS contribution can be presented as

$$T_1(\omega, \gamma) = \frac{2\alpha^2 \mu \Lambda(\omega) k_1}{\omega^3 (1 - \gamma^2)^2}, \quad (11)$$

with

$$\mu = \int d^3\mathbf{r} |\Psi(0, \mathbf{r})|^2 \frac{1}{r^2}, \quad (12)$$

where $k_1=2$ for heliumlike ions and $k_1=1$ for H^- and for the excited states of helium. Thus, the FSS contribution is presented via a single parameter of the initial state, expressed by Eq. (12).

The QFM contribution has a more complicated form, containing a function of the momentum $\mathbf{q} = \mathbf{p}_1 + \mathbf{p}_2 - \mathbf{k}$ transferred to the nucleus, integrated over kinematical constraints. The QFM amplitude can be written as [1,4]

$$F_{QFM}(\mathbf{k}, \mathbf{p}_1, \mathbf{q}) = F_0(\mathbf{k}, \mathbf{p}_1) S(q^2), \quad (13)$$

with $F_0(\mathbf{k}, \mathbf{p}_1)$ given by the following expression:

$$F_0(\mathbf{k}, \mathbf{p}_1) = (4\pi\alpha)^{3/2} 4\sqrt{2} \frac{(\mathbf{e}\mathbf{p}_1)(\mathbf{p}_1\mathbf{k})}{m^3 \omega^3}. \quad (14)$$

Here $F_0(\mathbf{k}, \mathbf{p}_1)$ is the amplitude of interaction of a photon with a system of two electrons at rest, with the final state consisting of two free electrons with momenta $\mathbf{p}_1, \mathbf{k} - \mathbf{p}_1$. The factor

$$S(q^2) = \int d^3\mathbf{r} \Psi(\mathbf{r}, \mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}} \quad (15)$$

describes the transfer of momentum \mathbf{q} to the nucleus by the bound electrons. The QFM contribution to the energy spectrum can be presented as

$$\frac{d\sigma^{2+}}{d\varepsilon} = \frac{1}{4} \frac{m^2}{\omega} |F_{QFM}|^2 \frac{dq^2 dt}{(2\pi)^3} \quad (16)$$

with $t = \mathbf{p}_1 \mathbf{k} / p_1 k$, the value $|F_{QFM}|^2$ is averaged over the photon polarizations. Using Eqs. (13)–(16), we can present

$$T_2(\omega, \gamma) = \frac{3\sqrt{2}}{2\pi^2 Z^2} \Lambda(\omega) \left(\frac{\omega}{m} \right)^{1/2} \int_{-1}^1 dt t^2 (1 - t^2) \Phi(\omega, \gamma, t) \quad (17)$$

with $\Lambda(\omega)$ defined by Eq. (10), while

$$\Phi(\omega, \gamma, t) = \int_{q_{\min}^2}^{4p_1^2} dq^2 D(q^2),$$

$$D(q^2) = |S(q^2)|^2. \quad (18)$$

The function $S(q^2)$ is defined by Eq. (15),

$$q_{\min}^2(\gamma, t) = (p\gamma - \omega t)^2. \quad (19)$$

Here $p = (m\omega)^{1/2}$ is the value of the momentum of the outgoing electrons at the central point of the spectrum $\gamma=0$.

In the QFM the nucleus obtains the momentum $q \sim \eta$, which is much smaller than the outgoing electron momenta. The values of $q \sim \eta$ are inside the interval of integration on the right-hand side (rhs) of Eq. (12) if the value of γ is small enough. In any case, the QFM kinematics is available at the central point, i.e., at $\gamma=0$.

The values of ω where the spectrum curve with a minimum at the central point converts into that with a maximum, and vice versa, are determined by the equation

$$T_1''(\omega, 0) + T_2''(\omega, 0) = 0, \quad (20)$$

with $T_i''(\omega, 0)$ being the the second-order derivative with respect to γ at $\gamma=0$.

A straightforward calculation, which involves integration in the angular variable t [Eq. (17)] by parts enables us to present Eq. (20) in the form

$$\frac{8\alpha^2 \mu k_1}{\omega^3} + \frac{6\sqrt{2}}{\pi^2 Z^2} \omega m \left(\frac{\omega}{m} \right)^{1/2} A(\omega) = 0. \quad (21)$$

Here

$$A(\omega) = \int_{-1}^1 dt t^2 (1 - 2t^2) D(\omega^2 t^2), \quad (22)$$

while the function D , defined by Eq. (18), can be presented as

$$D(\omega^2 t^2) = \left| 4\pi \int_0^\infty dr r^2 \frac{\sin \omega tr}{\omega tr} \Psi(r, r) \right|^2. \quad (23)$$

III. FROM U TO W

At small values of ω the first term on the left-hand side (lhs) of Eq. (21) dominates and therefore the sum on the lhs is positive. At some ω value, $\omega = \omega_1$, the second term compensates the first one.

Let us start with the heliumlike ions. We can estimate μ as $\mu \sim \eta^5$, and thus $\omega_1 \sim \eta (\alpha Z)^{5/9} \ll \eta$. On the other hand, the integral on the rhs of Eq. (23) is dominated by the values of r of the order of the size of the $1s$ state, i.e., $r \sim \eta^{-1}$. Thus, we can try the expansion in powers of ωtr in the integrand of Eq. (23). Setting $(\sin \omega tr) / \omega tr = 1$ we obtain

$$A(\omega) = \text{const} = A = -\frac{2}{15}B, \quad (24)$$

with

$$B = \left| \int d^3\mathbf{r} \Psi(\mathbf{r}, \mathbf{r}) \right|^2. \quad (25)$$

Assuming that the function $A(\omega)$ is given by Eq. (24), we find the solution of Eq. (21)

$$\omega_1^* = 5^{2/9} (\pi\alpha Z)^{4/9} \left(\frac{8\mu^2}{B^2 m} \right)^{1/9}. \quad (26)$$

As one can see from Table I, the values of ω_1 and ω_1^* are very close indeed.

The parameters μ and B defined by Eqs. (12) and (25) are determined by the wave functions $\Psi(0, \mathbf{r})$ and $\Psi(\mathbf{r}, \mathbf{r})$ at $r \sim \eta$. At large $Z \gg 1$ one can expect, at these distances the hydrogenlike behavior of the functions [7,8]. Thus, at $Z \gg 1$ the parameter B does not depend on Z , while $\mu \sim Z^5$. Hence, $\omega_1^* \sim Z^{14/9}$. The straightforward computations lead to Eq. (1).

Since the important values of r in the rhs of Eq. (25) are of the order of η^{-1} , we have neglected the terms of the relative order of about αZ by setting $(\sin \omega r)/\omega r = 1$. Thus, the equality $\omega_1 = \omega_1^*$ holds with the relative accuracy αZ .

Turning to the excited n^1S states of helium, let us start from the limiting case $n \gg 1$. In the single-particle picture of the FSS, the cross section $T_1(\omega, \gamma)$ depends on two parameters of the initial state. These are the expectation value $\langle r^{-2} \rangle$ of one of the electrons and the normalization factor $|\psi_s(0)|^2$ of another electron—see Eqs. (9) and (10). In the limit $n \gg 1$ one can expect the product of the Coulomb functions to be a qualitatively good approximation. In this case both $\langle r^{-2} \rangle$ and $|\psi_s(0)|^2$ decrease as n^{-3} with growing n [10]. As to the QFM contribution, the function $\Psi(\mathbf{r}, \mathbf{r})$ describes the configuration in which the external electron comes to the distances about the size of the internal electron orbit. The shape of the r dependence does not depend on n in this case. Thus, all the n dependence is contained in the normalization factor n^{-3} . Hence, both terms on the lhs of Eq. (21) depend on n for $n \gg 1$ in the same way. Therefore the value of ω_1 as well as the approximate solution of Eq. (21)

$$\omega_1^* = 5^{2/9} (\pi\alpha Z)^{4/9} \left(\frac{2\mu^2}{B^2 m} \right)^{1/9} \quad (27)$$

do not depend on n for $n \gg 1$. This equation is true for the negative ion of hydrogen H^- as well.

The values of ω_1 and ω_1^* for $n=2, 3, 4$ are presented in Table II. One can see that the n dependence is very weak even for these values of n .

Note that at the point ω_1 we still have $T_2 \ll T_1$, i.e., the QFM provides a small correction to the contribution of FSS. This happens because the cross section $T_2(\omega, \gamma)$ depends on γ via the parameter $p^2 \gamma^2 / \eta^2$, with $p^2 / \eta^2 = m\omega / \eta^2 \gg 1$. Thus,

$T_2''(\omega, 0) / T_2(\omega, 0) \sim \omega / I \gg 1$. On the other hand, $T_1''(\omega, 0) = 4T_1'(\omega, 0)$. Hence, Eq. (21) leads to $T_2 \ll T_1$. This means that evaluation of the spectrum curve starts with a small QFM surplus on the smooth FSS curve.

IV. FINE STRUCTURE OF THE PEAK

At $\omega > \omega_1$ the lhs of Eq. (20) becomes negative. However, one can show that there is one more root of this equation. Assuming $\omega \gg \eta$ we find that $T_1''(\omega, 0) \ll T_2''(\omega, 0)$, while $T_2''(\omega, 0) > 0$. The latter inequality can be obtained by noting that the integrand on the rhs of Eq. (23) is determined by $r \sim \eta$. Thus, the integral on the rhs of Eq. (22) is dominated by small $|t| \sim 1/\omega r \ll 1$, where the integrand is positive. Hence, we can expect that there exist a root $\omega_2 \sim \eta$ of Eq. (21). At $\omega \sim \eta$ the FSS contribution T_1 is at least $\alpha \approx 1/137$ times smaller than the QFM term T_2 . This means that the solution ω_2^* of the equation

$$A(\omega) = 0 \quad (28)$$

is expected to be close to the solution of Eq. (21).

This is supported by the computations which employ our initial state wave functions. The results are presented in Tables I and II.

We can trace the dependence of ω_2^* on Z for the photoionization of heliumlike ions with the nuclear charge $Z \gg 1$. The integrand on the rhs of Eq. (23) is saturated by $r \sim \eta^{-1}$. To obtain the dependence on Z we can approximate the wave function $\Psi(\mathbf{r}, \mathbf{r})$ by a product of the hydrogenlike $1S$ functions, i.e., by setting $\Psi(\mathbf{r}, \mathbf{r}) \approx e^{-2\eta r}$ (the normalization factor is not important for estimation). This gives

$$D(\omega^2 t^2) = D_h(\omega^2 t^2) = \left[\frac{2^6 \pi^2 \eta^2}{(\omega^2 t^2 + 4\eta^2)^2} \right]^2. \quad (29)$$

Now Eq. (28) can be written as

$$\int_{-1}^1 dt t^2 (1 - 2t^2) \frac{1}{(t^2 + a^2)^4} = 0, \quad (30)$$

with $a = 2\eta/\omega$. The solution is $a = 1.534$ [of course, the left-hand side of Eq. (30) can be evaluated explicitly], and thus $\omega_2^* = 1.30\eta = 4.85Z$ keV. This is very close to Eq. (4), which is true with 1% accuracy even for $Z=5$.

In the case of the ionization of the excited n^1S states of helium, the dependence of the values ω_2 and ω_2^* on n is very weak. The reasons are the same as for the weak dependence of the values ω_1 and ω_1^* .

V. SUMMARY

In this paper we traced the evolution with energy of the photoelectron spectrum of the double photoionization of atomic helium, light heliumlike ions, the negative ion of hydrogen H^- , and of the excited states of atomic helium. We found the values of the photon energies ω_1 , at which the U-shaped curve converts into a W-shaped curve, and ω_2 , at which the central peak splits into two. We found the limiting equations for ω_1 and ω_2 for the ionization of the heliumlike

ions with $Z \gg 1$. We obtained the limiting behavior $\omega_1 = \text{const}$ and $\omega_2 = \text{const}$ at $n \gg 1$ for the ionization of the excited n^1S states.

We carried out the corresponding calculations by using the wave functions developed by using the correlation function hyperspherical harmonic method [7,8]. We show that the limit laws start working at rather small values of Z and n .

We obtained the values of ω_1 and ω_2 without building particular energy distributions. Such distributions will be presented in a separate publication.

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