Pis'ma v ZhETF, vol. 78, iss. 3, pp. 141 – 145

Double K-shell ionization of atoms by a single photon

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Submitted 26 June 2003

We investigate the double K-shell ionization of heliumlike ions by a single photon. A fast convergency of QED perturbation theory with respect to parameter $1/Z$ is demonstrated in entire nonrelativistic domain for moderate nuclear charge numbers $Z \geq 2$. The ratio of double-to-single photoionization cross sections is calculated for light heliumlike ions, taking into account the leading orders of $1/Z$ and αZ expansions. A comparison of our results with available experimental data for a number of neutral atoms is presented.

PACS: 31.25.–v, 32.30.Rj, 32.80.Fb, 33.60.Fy

In studies of electron correlations in atoms, the most attractive processes are those, in which the electron-electron interaction plays the crucial role. One of such a fundamental phenomenon is the double photoionization of an atom caused by the absorption of a single photon, the so-called double photoeffect, which has been investigated for more than 30 years [1 – 3]. Since a photon interacts only with one electron, the simultaneous ejection of two electrons is exclusively caused by the electron-electron interaction. Accordingly, electron correlations show up here most clearly. So far the experiments have been mainly performed with helium, the simplest many-electron atomic system. The majority of investigations concerns the energy dependence of the ratio R of double-to-single photoionization cross sections [4 – 6]. With increasing photon energy ω, the ratio R grows rapidly beyond the ionization threshold. Then, after having a maximum near the threshold, it declines slowly approaching the constant limit of 1.72(12)% [7] in the asymptotic domain of nonrelativistic photon energies much larger than the threshold energy I_{2K} for double ionization from the K-shell, that is, $I_{2K} \ll \omega \ll m$, where m is the electron mass ($\hbar = c = 1$). One of the most frequent problems in theoretical descriptions of the double photoionization is the gauge dependence of numerical results.

Due to recent developments of novel synchrotron radiation sources, experiments with intense collimated beams of tunable monochromatic x-rays in the keV regime became feasible [7 – 9]. This represents a challenge to theoretical investigations of double ionization of atomic inner-shell electrons in the entire nonrelativistic domain, both for photon energies $\omega \ll m$ and for targets with moderate values of nuclear charge numbers Z. In Refs. [10, 11], a Z-scaling law was suggested for the ratio R of double-to-single photoionization cross sections in the asymptotic energy regime, $I_{2K} \ll \omega \ll m$. For the energy domain near the threshold, $\omega \simeq I_{2K}$, *ab initio* calculations are presently not available. Nevertheless, one can mention here a model estimate of the two-electron photoejection cross section σ^{++} obtained in Ref. [12], which however strongly disagrees with the existing experimental data. Another numerical calculation of $Z^4\sigma^{++}$ has been performed within the framework of convergent close-coupling model for He, Li^+, and O^{6+} [13].

In this Letter, we have investigated the double photoionization of the ground state of heliumlike ions. The perturbation theory is developed with respect to the electron-electron interaction. As a zero approximation, Coulomb wave functions and Coulomb Green functions are utilized. The study is performed for photon energies much smaller than the electron rest energy. Accordingly, all electrons involved in the ionization process are considered as being nonrelativistic. This implies the smallness of the Coulomb parameter, that is, $\alpha Z \ll 1$. However, the nuclear charge number Z is supposed to be high enough to utilize $1/Z$ as expansion parameter. A similar approach has been already used in the asymptotic part of the nonrelativistic domain, $I_{2K} \ll \omega \ll m$, where all formulae can be significantly simplified [10]. In contrast to Ref. [10], we consider the entire nonrelativistic domain of incident photon energies with special emphasis on the threshold region. We have analyzed limits of applicability of the approximations employed. Since K-shell electrons are essentially separated from the other electrons in an atom, it turns out that our formulae also describe fairly well the double K-shell ionization in the case of light neutral atoms.

In leading order of nonrelativistic perturbation theory the double photoionization is described by the gauge invariant set of the Feynman diagrams depicted in Fig.1. To ensure gauge invariance, the energy conservation law

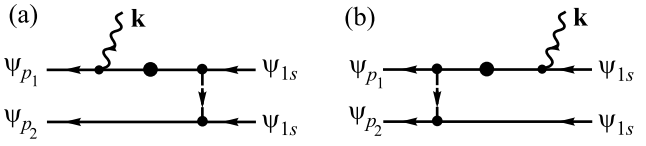

Fig.1. Feynman diagrams for the double ionization of the atomic K-shell by a single photon. Solid lines denote electrons in the Coulomb field of nucleus, dashed line denotes the electron-electron Coulomb interaction, and the wavy line denotes an incident photon. The line with a heavy dot corresponds to the Coulomb Green function. For this line, only the energy is conserved, while the momentum is violated due to the interaction with a nucleus. Diagram (a) takes into account the electron-electron interaction in the initial state, while diagram (b) accounts for it in the final state. In addition to these graphs, one also has to take into account the corresponding exchange diagrams

is defined to zeroth order as follows [14]:

$$E_{p_1} + E_{p_2} = \omega - I_{2K}. \tag{1}$$

Here E_{p_1} and E_{p_2} are the one-electron energies in the continuum final state, $I_{2K} = 2I$ with $I = \eta^2/2m$ being the Coulomb potential for single ionization, and $\eta = m\alpha Z$ is the average momentum of a K-shell electron. Since the photon energy ω is distributed between two electrons, an escaping electron can have any energy within the range between 0 and $\omega - I_{2K}$.

The domain of photon energies near the threshold of double photoionization corresponds to the dipole regime characterized by $\omega \ll \eta$. Under the latter condition, the photon momentum $\mathbf{k}$ can be neglected, while the recoil momentum $\mathbf{q}$ transferred to the nucleus and the momenta $\mathbf{p}_1$ and $\mathbf{p}_2$ of both outgoing electrons are of the order of a characteristic atomic momentum, $q \sim p_1 \sim p_2 \sim \eta$. Correspondingly, the process occurs at atomic distances of the order of the K-shell radius. This implies that the interaction with the Coulomb field of the nucleus has to be included in the initial, intermediate, and final electron states, that is, Coulomb wave and Coulomb Green functions should be used already in zeroth approximation [15 – 17]. In addition, the electron-electron interaction has to be taken into account in both initial and final states. All Feynman graphs depicted in Fig.1 are expected to give comparable contributions to the total cross section for double photoionization.

Except for the parameter αZ, there are further Coulomb parameters involved in the problem, which characterize the interaction of intermediate and both outgoing electrons with the Coulomb field of nucleus. They are given by $\xi = \eta/p$ with p being a momentum of the virtual electron, $\xi_1 = \eta/p_1$, and $\xi_2 = \eta/p_2$. Formally, these ξ parameters are values of the order of 1. Accordingly, dependencies upon them in Coulomb Green and wave functions have been taken into account exactly, but including terms of the order of αZ only. Terms of the order of $(\alpha Z)^2$ have been omitted in the present consideration. In the vicinity of the threshold, it can happen that the momenta of the ejected electrons, p_1 and/or p_2, become extremely small or, equivalently, ξ_1 and/or ξ_2 can reach rather large values compared to 1. This situation corresponds to the infrared catastrophe or to the quasiclassical limit, known also as the Wannier regime [18].

In the following, it is convenient to introduce dimensionless quantities, such as energies and momenta. We shall express all momenta in units of $\eta = m\alpha Z$. For example, the dimensionless momentum of a photon is just $\mathrm{k} = k/\eta$. The energies are calibrated in units of $I = \eta^2/2m$. Then the dimensionless energy of the incoming photon is given by $\varepsilon_\gamma = \omega/I$, while $\varepsilon_i = E_{p_i}/I$, $(i = 1, 2)$, represent the dimensionless energies of the ejected electrons. The energy conservation law reads as $\varepsilon_1 + \varepsilon_2 = \varepsilon_\gamma - 2$. The double-photoionization threshold corresponds to the photon energy $\varepsilon_\gamma^{\mathrm{th}} = 2$. The amplitudes of the process being expressed in terms of dimensionless quantities become dependent on the nuclear charge number Z via the photon momentum $\mathrm{k} = \alpha Z \varepsilon_\gamma/2$.

The total cross section of the double photoionization can be cast into the following form

$$\sigma^{++}(\varepsilon_\gamma; \mathrm{k}) = \sigma_0 \frac{2^{10}\pi}{3Z^4} Q(\varepsilon_\gamma; \mathrm{k}), \tag{2}$$

where $\sigma_0 = \alpha\pi a_0^2$ and $a_0 = 1/m\alpha$ is the Bohr radius. Due to a complicated dependence upon the variables ε_γ and k, the function $Q(\varepsilon_\gamma; \mathrm{k})$ can be obtained only by a numerical integration. The dipole regime corresponds to the approximation $\mathrm{k} = 0$. A smallness of the photon momentum is due to the mutual interplay between two input quantities, ε_γ and Z. If one sets $\mathrm{k} = 0$, the function $Q(\varepsilon_\gamma)$ becomes independent of the value of Z. The function holds the same for the whole helium isoelectronic sequence. Accordingly, the ratio of double-to-single ionization cross sections, which is usually measured experimentally, is given by

$$R(\varepsilon_\gamma) = \frac{\sigma^{++}(\varepsilon_\gamma)}{\sigma^{+}(\varepsilon_\gamma)} = \frac{1}{Z^2}\frac{Q(\varepsilon_\gamma)}{H(\varepsilon_\gamma)}. \tag{3}$$

The energy factor $H(\varepsilon_\gamma)$, which enters in expression for the cross section of the single K-shell photoionization in the leading order of perturbation theory, reads as follows [19]

$$H(\varepsilon_\gamma) = \frac{1}{\varepsilon_\gamma^4}\frac{\exp(-4\tau\cot^{-1}\tau)}{[1-\exp(-2\pi\tau)]}, \tag{4}$$

where $\tau = 1/\sqrt{\varepsilon_\gamma - 1}$.

All our numerical calculations have been performed within the Coulomb gauge, since it easily allows to separate the leading contributions in the nonrelativistic limit. Corrections due to spin-dependent terms turn out to be suppressed by a factor of the order of $(\alpha Z)^2$ in transition amplitudes of the process and, therefore, have been neglected. We have compared the universal quantity $Z^4\sigma^{++}$ calculated in the dipole approximation according to Eq. (2) with that obtained in Ref.[13]. The curves lie quite close to each other, although they are not identical. The contributions to σ^{++} due to two-photon exchange graphs are relatively small. In Fig.2, the universal ratio $Z^2R(\varepsilon_\gamma)$ is depicted, which is valid for moderate Z

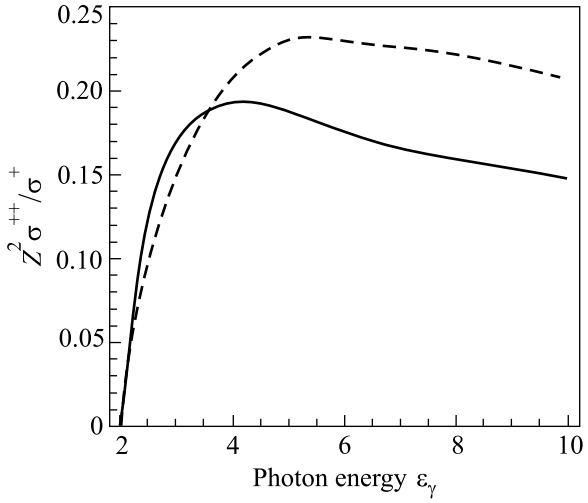

Fig.2. Different contributions to the universal ratio $Z^2R(\varepsilon_\gamma)$ of double-to-single photoionization cross sections calculated in Coulomb gauge for k = 0 according to Eq. (3). Dashed line: contribution due to the electron-electron interaction in the initial state only; solid line: total contribution of all diagrams

values. Accounting for the electron-electron interaction to the final state results in significant corrections to the total cross section of the double photoionization. Note, however, that the individual contributions of each diagram are gauge dependent.

In Fig.3, we compare our numerical results for the ratio σ^{++}/σ^+ with most recent measurements for helium [4–6]. The comparison of theoretical and experimental data is given in dependence on the excess energy, since the experimental threshold energy $I_{2K}^{\rm exp} = 79.0$ eV and the theoretical one $I_{2K} = 108.85$ eV are different. However, this is an apparent problem. In fact, the expansion with respect to $1/Z$ converges by an order of magnitude in whole nonrelativistic domain, starting from helium. The contribution to the ionization potential due to the

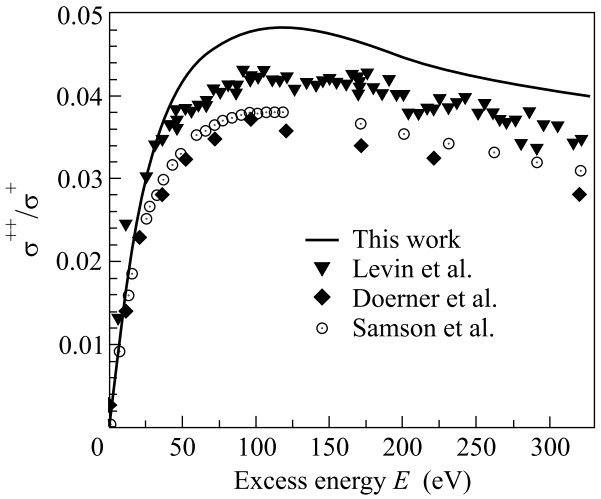

Fig.3. The ratio of double-to-single photoionization cross sections of the ground state of helium is given as a function of the excess energy $E = E_{p_1} + E_{p_2}$. Experimental data taken from Levin et al. [4], Dörner et al. [5], and Samson et al. [6] account also for the contribution from the cross section σ^{+*} of single photoionization with excitation, while the theoretical calculation is performed according to Eq. (3)

electron-electron interaction treated by the exact one-photon exchange leads to the correction $\Delta I_{2K}^{(1)} = -34.02$ eV, that is, the threshold energy becomes equal to 74.83 eV. The correction due to two-photon exchange diagrams, which is known to be $\Delta I_{2K}^{(2)} = 4.29$ eV [20], yields a threshold energy of about 79.12 eV. The contribution, which results from the three-photon exchange, has been calculated in the ladder approximation [21]. It yields $\Delta I_{2K}^{(3)} = -0.12$ eV [22] and brings the theoretical threshold energy in agreement with the experimental value. Independent measurements of the ratio of double-to-single photoionization cross sections performed by Dörner et al. [5] and by Samson et al. [6] seem to be more reliable than the experimental data by Levin et al. [4] (see discussion in Ref. [6]). The disagreement between our results and those obtained in Refs. [5, 6] is due to next-to-leading order terms of perturbation theory in $1/Z$, which have been omitted in the present evaluation of single photoionization cross section.

The fast convergency of the expansion over $1/Z$ in entire nonrelativistic domain, including the threshold area, can be understood by a careful distinction between formal and real characteristic scales. The characteristic spatial distances involved in the problem turn out to be somewhat larger than the K-shell radius. This is confirmed, for example, by calculations of the double-electron ionization in Compton scattering, where perturbation theory with respect to $1/Z$ is proven to be

For various neutral atoms, the nuclear charge numbers Z, the experimental energies ω of an incident photon, the experimental potentials $I^{\rm exp}$ for single K-shell ionization, dimensionless photon energies ε_γ, effective values $Z_{\rm eff}$ for the nuclear charge, and the theoretical and experimental ratios $R(\varepsilon_\gamma)$ of double-to-single K-shell ionization cross sections are tabulated. The photon energies ω are calibrated in units of the experimental ionization potentials $I^{\rm exp}$ taken from Ref. [26]. The theoretical ratios $R(\varepsilon_\gamma)$ are calculated using the effective values $Z_{\rm eff}$ according to Eq. (3)

Neutral atom	Z	ω (keV)	$I^{\rm exp}$ (keV)	ε_γ	$Z_{\rm eff}$	$R(\varepsilon_\gamma)$		
						this work	experiment	
Ne	10	3	0.87	3.45	8.0	$0.29 \cdot 10^{-2}$	$0.3 \cdot 10^{-2}$	[27]
Ti	22	17.4	4.97	3.50	19.11	$0.51 \cdot 10^{-3}$	$0.53 \cdot 10^{-3}$	[28]
Cr	24	17.4	5.99	2.90	20.98	$0.37 \cdot 10^{-3}$	$0.38 \cdot 10^{-3}$	[28]
Fe	26	17.4	7.12	2.44	22.88	$0.23 \cdot 10^{-3}$	$0.24 \cdot 10^{-3}$	[28]
Ni	28	17.4	8.34	2.09	24.76	$0.51 \cdot 10^{-4}$	$1.1 \cdot 10^{-4}$	[28]
Cu	29	20	8.99	2.22	25.70	$1.1 \cdot 10^{-4}$	$1.3(3) \cdot 10^{-4}$	[9]
Mo	42	50	20.01	2.50	38.35	$0.87 \cdot 10^{-4}$	$3.4(6) \cdot 10^{-4}$	[8]

satisfactory for helium in the asymptotic domain of nonrelativistic photon energies [23, 24]. The distinction between formal and real scales also explains the fact that for heliumlike ions with $Z > 2$ the double photoionization from the excited 2^1S state turns out to be more probable than from the K-shell [11, 25]. It should be noted that in the next-to-leading order of perturbation theory in $1/Z$ the vertex contributions due to the crossed diagrams occur, which entangle correlation effects in the initial and final electron states.

Considering double K-shell photoionization in neutral atoms, the wave functions and Green functions possess essentially a non-Coulomb behavior. Accordingly, numerical calculations require the use of the Hartree-Fock method already in zeroth approximation. Formally, the screening effect can be simulated by replacing the true nuclear charge number Z in Eq. (3) by an effective value $Z_{\rm eff}$. The latter can be defined by equating the experimental potential $I^{\rm exp}$ for single K-shell ionization and the effective one, that is, $I^{\rm exp} = m(\alpha Z_{\rm eff})^2/2$. Apart from the nickel and molybdenum systems, a good overall agreement between our predictions for neutral atoms with available experimental data is achieved (see Table). The significant disagreement for the nickel atom seems to be just due to high uncertainties of the results, both theoretical and experimental. The ratio $R(\varepsilon_\gamma)$ here is extremely sensitive to the photon energy, because the latter is very close to the threshold energy. In the case of molybdenum, a possible explanation for the deviation may be connected with relativistic effects, for example, due to the spin-orbit interaction, which have been neglected in the present consideration. Another reason might be higher error bars rather than those quoted in Ref. [8].

In conclusion we demonstrate a fast convergency of QED perturbation theory with respect to parameter $1/Z$ in entire nonrelativistic domain for $Z \geq 2$. The double K-shell photoionization has been investigated for heliumlike ions and neutral atoms with moderate Z values, taking into account the leading orders of $1/Z$ and αZ expansions. Going beyond the leading-order consideration requires rigorous QED description.

We thank E. P. Kanter and R. W. Dunford for providing experimental data to us prior to publication. A.M. is grateful to the Dresden University of Technology for the hospitality and for financial support from DFG. G.S. and G.P. acknowledge financial support from BMBF, DFG, and GSI. A.N. is supported by the Alexander von Humboldt Foundation. A.M. and I.M. acknowledge support from RFBR (Grants # 01-02-17246 and # 00-15-96610).

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