Breakdown of asymptotics of the double photoionization of helium at the high-energy limit

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It is shown that the contribution to the double-to-single ionization ratio $\sigma^{2+}(\omega)/\sigma^{+}(\omega)$ of helium caused by small momentum transfer breaks down the asymptotics of this ratio at the high-energy limit. The addition of up to 10% at $\omega = 12$ keV is expected to cause a slope in the ratio, which can be related to the properties of the ground state. The contribution is expected to be important also in the analysis of the double photoionization of H^- and H_2.

I. INTRODUCTION

Early experiments on the double photoionization of helium,

$$\gamma + \text{He} \rightarrow \text{He}^{2+} + e^- + e^- \tag{1}$$
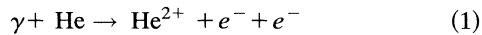
with photons of energies up to hundreds eV showed the ratio of cross sections for double ionization to single ionization

$$R(\omega) = \frac{\sigma^{2+}(\omega)}{\sigma^{+}(\omega)} \tag{2}$$

to turn into constant

$$R = R_0 \approx 1.6 \times 10^{-2} \tag{3}$$

for photon energies much larger than the binding energies J. The result was supported by numerous calculations reviewed in [1,2].

Recent experiments [3] carried out with photons of energies of several keV gave data consistent with Eq. (3). However, it was noted in [4] that the experiments detected the number of ions He^+ and He^{2+} in the final state. At the energies $\omega \gtrsim 6$ keV, the former were produced mostly by Compton scattering. Thus the result of these experiments supports

$$R_t(\omega) \equiv \frac{\sigma_t^{2+}(\omega)}{\sigma_t^{+}(\omega)} = \text{const}, \tag{4}$$

with the total cross sections $\sigma_t = \sigma + \sigma_c$, where $\sigma_c^{+\ (2+)}$ denotes the cross section of the Compton process $\omega + \text{He} \rightarrow \omega' + \text{He}^{2+} + e^- + e^-$ [4].

The results of the first calculations of σ_c^{2+} have been published recently [5]. Since the cross sections σ^{2+} and σ_c^{2+} can be distinguished experimentally [4] it is reasonable to start with the simpler problem of calculating the ratio $R(\omega)$ determined by Eq. (2) at larger energies. In this paper we calculate a contribution that causes a breakdown of the asymptotics described by Eq. (3). It increases the value of R by $5-10$ % at $\omega = 12$ keV.

II. BREAKDOWN OF ASYMPTOTICS

To understand the nature of the contribution, recall the mechanism of single photoionization in the asymptotic region, which requires that large momentum be transferred to the nucleus since the process cannot take place if the electron is free. The energy dependence of the amplitude F_S for the single photoionization is determined by that of the bound-electron wave function Ψ in momentum space. Effectively,

$$F_s = (4\pi\alpha)^{1/2} \frac{(e p_1)}{m} \psi(p_1 - k). \tag{5}$$

Since $\omega \gg J$, $p_1 - k \approx p_1$ is much larger than the characteristic momentum η of the bound electron η. (If the bound electrons are described by Coulomb functions with effective charge Z_e, $\eta = m\alpha Z_e$.) Bound electrons can exchange an infinite number of virtual photons with momentum $f \sim \eta$ but each exchange by momentum $p_1 \gg \eta$ leads to a small factor of the order η^4/p_1^4; see Fig. 1(a). So large momentum p_1 is transferred to the nucleus by a single hard photon and

$$\psi(p_1 - k) = \frac{-8\pi m \alpha Z N_0}{(p_1 - k)^4}. \tag{6}$$

Here N_0 is the wave function at the origin of configuration space. Substituting Eq. (6) into Eq. (5) we find the amplitude
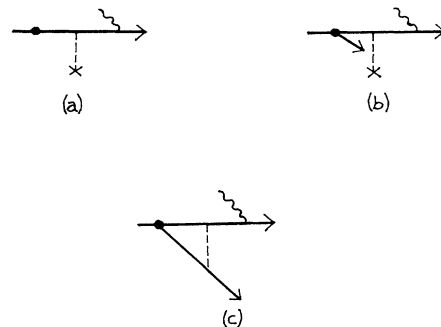

FIG. 1. The Feynman diagrams describing the mechanism of single and double photoionization processes in the asymptotic region [(a) and b)] and the mechanism causing the breakdown of asymptotics (c). The dark blob represents soft photons with momenta $f \sim \eta$ exchanged between electrons and between electrons and the nucleus. The dashed line shows the hard virtual photon with momentum $f \gg \eta$.

F_s. In the general case the two-electron system is described by the function $\Psi(r_1, r_2)$. The amplitude for single ionization is

$$F_s = (4\pi\alpha)^{1/2} \frac{(ep_1)}{m} \frac{8\pi\, m\alpha\, Z}{(p_1 - k)^4} N, \tag{7}$$

with

$$N = \langle \varphi_0(r) | \Psi(r,0) \rangle, \tag{8}$$

where φ_0 describes the electron in the ground state of He^+.

The main contribution to the cross section of double photoionization comes from the part of the spectrum where one of the outgoing electrons carries a small energy $\varepsilon_2 \sim J \ll \omega$. One of the bound electrons again transfers a large momentum $p_1 \gg \eta$ to the nucleus. The amplitude takes the form of Eq. (7), with N being replaced by the matrix element $\langle \varphi_2(r) | \Psi(r,0) \rangle$ and with φ_2 describing the outgoing electron with energy ε_2 in the field of the nucleus [Fig. 1(b)]. The amplitude of the double ionization contains F_s as a factor.

The cross section

$$d\sigma^{2+} = \frac{\pi}{\omega} |F|^2 \delta(\omega - \varepsilon_1 - \varepsilon_2) d\Gamma, \tag{9}$$

with $d\Gamma \equiv [d^3p_1/(2\pi)^3][d^3p_2/(2\pi)^3]$, is dominated by $p_2 \sim \eta$, $\varepsilon_2 \sim J$. This leads to a similar energy dependence of σ^+ and σ^{2+}, i.e. to Eq. (3), and the integral over $p_2 \sim \eta$ determines the value of R_0.

If the energies of both outgoing electrons $\varepsilon_{1,2} \sim \omega \gg J$, the process also yields large momentum that is to be transferred to the nucleus unless the values of the energies are not close to $\omega/2$. In this region the amplitude also contains the amplitude of single photoionization [6]. The contribution of this part of the spectrum to R is of the order $(J/\omega)^2$.

However, if the values $\varepsilon_{1,2}$ are close to $\omega/2$, an alternative mechanism is possible in which large momentum $k - p_1$ is transferred to the second electron but not to the nucleus—see [6] and the more detailed analysis in [7]. The process can take place without the nucleus; the amplitude F_0 for the process when the initial state is that of two free electrons at rest does not vanish. It yields

$$k = p_1 + p_2. \tag{10}$$

Equation (10) leads to

$$\left| \frac{\varepsilon_1 - \varepsilon_2}{\omega} \right| \lesssim \left(\frac{\omega}{m} \right)^{1/2}. \tag{11}$$

One can see from Fig. 1(c) that the structure of the amplitude is similar to that of the single-photoionization process F_s. However, now it is the second electron that accepts the large momentum $k - p_1$ and then leaves the atom with momentum $p_2 = k - p_1$. For that part of the spectrum determined by Eq. (11) the amplitude can be presented as the product of the amplitude for the free process F_0 and a factor that depends on momentum $q = k - p_1 - p_2$ that is transferred to the nucleus,

$$F(\omega, p_1, q) = F_0(\omega, p_1) S(q), \tag{12}$$

where

$$F_0 = (4\pi\alpha)^{3/2} \left(\frac{(ep_1)}{(p_1 - k)^4} + \frac{(ep_2)}{(p_2 - k)^4} \right). \tag{13}$$

The factor $S(q)$ is easily expressed through the two-electron wave function in momentum space

$$S(q) = \int \frac{d^3f}{(2\pi)^3} \Psi(q - f, f) \tag{14}$$

or in configuration space

$$S(q) = \int d^3r \, \Psi(r,r) e^{-iqr}. \tag{15}$$

Now we calculate the cross section using Eq. (9) and presenting $d\Gamma = [d^3p_1/(2\pi)^3][d^3q/(2\pi)^3]$. The integral over q is dominated by $q \sim \eta \ll p$. The amplitude F_0 appears to be $(\omega/m)^{1/2}$ times smaller than that of the single photoeffect. Neglecting the terms $\sim (k/p_1) \sim (\omega/m)^{1/2}$ in Eq. (13), we obtain $F_0 \sim (e, p_1 + p_2) = (ek) = 0$. Thus the part of the spectrum determined by Eq. (11) gives a correction of the order ω/m to the ratio $R(\omega)$. A straightforward calculation gives

$$R(\omega) = R_0 + \frac{8\sqrt{2}}{5Z^2} C \frac{\omega}{m}, \tag{16}$$

with

$$C = \frac{1}{N^2} \int \frac{d^3q}{(2\pi)^3} |S(q)|^2 = \frac{1}{N^2} \int d^3r |\Psi(r,r)|^2. \tag{17}$$

In terms of Hylleraas variables [8] $s = r_1 + r_2$, $t = r_1 - r_2$, $u = |r_{12}|$, the function $\Psi(r,r) = \Psi(s,t,u)$ with $s = 2r$, $u = t = 0$. If $\Psi(r_1, r_2)$ is the product of single-particle functions, i.e., $\Psi(r_1, r_2) = \psi(r_1)\psi(r_2)$, we obtain

$$C = \frac{1}{N^2} \int d^3r |\psi(r)|^4. \tag{18}$$

Note that the contribution can be obtained from the most general form of the equation for double ionization in configuration space:

$$F = \langle \phi(r_1, r_2) | \gamma_1 + \gamma_2 | \Psi(r_1, r_2) \rangle, \tag{19}$$

with $\gamma_i = e^{-i(kr_i)}(e\nabla_i)$ and ϕ the wave function of the final state. Since $\varepsilon_{1,2} \gg J$, the outgoing electrons can be described by plane waves. Thus Eq. (19) consists of terms of the form $\int dr_1 dr_2 \Psi(r_1, r_2) \exp[i(kr_1) - i(p_1 r_1) - i(p_2 r_2)]$. In the general case, the exponential factor limits the phase volume of integration by $|r_{1,2}| \lesssim p_{1,2}^{-1} \ll \eta^{-1}$, while the bound electrons are concentrated mostly at the distances $r \sim \eta^{-1}$. Thus the amplitude is strongly quenched. However, introducing variables $R = (r_1 + r_2)/2$, $\rho = (r_1 - r_2)/2$, we find that the exponential factor becomes $\exp[i(qR) + i(k - p_1 + p_2, \rho)]$. Thus if $q \sim \eta$ both r_1 and r_2 can be made of the order of η while the

distance between the bound electrons r_{12} should be small. We explicitly build the function Ψ at $r_{12} \sim p^{-1}$ by the calculation in momentum space.

III. SUMMARY

We have calculated the contribution that causes the breakdown of the asymptotics of the process described by Eq. (1). It is proportional to the photon energy ω. The leading relativistic corrections to the cross sections σ^+ and σ^{2+} are also of this order. However, the proportionality between the amplitudes is maintained in the relativistic region, as well [9]. Hence the corrections cancel out in the ratio R_0. Thus the second term of Eq. (16) provides the only correction of the order ω/m.

Analysis of the energy distribution carried out in [7] shows the correction to exist at all the energies $\omega \gg J$. If $\omega \gg \eta(\alpha Z)^{1/3} \approx 1$ keV it is the leading correction to Eq. (3). The contribution becomes noticeable at larger values of ω.

We find that $C = 1/8$ if the hydrogenlike functions are used as the functions ψ. Note that the value does not depend on the charge of the nucleus. For the Hartree-Fock functions, $C = 0.11$. The value appears to be smaller for the functions depending on r_{12}. Considering the Hylleraas three- and six-parameter functions, which are polynoms in s, t, and u, we find $C = 7.0 \times 10^{-2}$ and $C = 6.8 \times 10^{-2}$. The more complicated Kinoshita function gives $C = 5.5 \times 10^{-2}$. (The explicit form of the functions and the values of parameters are presented in Table 6 of [10].)

Thus the correction increases the value of R by about 10% for the largest value of $\omega = 12$ keV for which there are experimental data nowadays. The effect increases linearly with ω. Measurements at higher values of ω should show a slope in the function $R(\omega)$. It is determined by certain properties of the ground state—see Eqs. (16) and (17).

The contribution occurs in the double photoionization of any system. In photoionization of heavier atoms it is of minor importance if at least one of the ionized electrons is an external one. In effect the latter interacts with the effective charge $Z_e \approx 1$ and $R \sim Z_e^{-2}$, while the second term of Eq. (16) is proportional to Z^{-2}. However, for the special case of double photoionization of the K shell, the dependence of both terms of right-hand side of Eq. (16) on Z is the same. The contribution is important in the analysis of the double ionization of lighter systems containing two electrons, such as the negative ion H^- and molecular hydrogen H_2 [11,12].

In other ionization processes (i.e., the ionization by electron impact) with large energy transferred to an atom, the double-to-single-ionization ratio also turns into a constant in asymptotics. However, the contribution to the ratio obtained in this paper does not emerge [9]. Indeed, in these processes the energy is transferred to the atom by virtual photons. Now single ionization is allowed for free electrons, and this also applies to the double ionization. Both processes take place with small momentum transfer $q \sim \eta$. The constant double-to-single ratio is determined by the part of the spectrum where one of the electrons is slow, i.e., $\varepsilon_1 \sim \omega$; $\varepsilon_2 \sim J$, while the region $\varepsilon_{1,2} \sim \omega$ provides a small correction $\sim (J/\omega)^2$.

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