Quantum-classical hybrid approach to helium double photoionization

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The photoionization process is divided into (A) the absorption of the photon by one electron and (B) the correlated motion of the electron pair leading to singly or doubly ionized helium. We relate (A) to the total cross section to be calculated analytically in the quasiclassical reflection approximation as known from molecular problems. For (B) the two-electron wave function is propagated with the semiclassical version of Feynman's path integral to separate single and double ionizing events. A probe for (B) is the ratio between double and single ionization. The results for (A) and (B) and for absolute ionization cross sections obtained by combining (A) and (B) are in good agreement with different experiments that cover together a range of photon energy from the double-ionization threshold to several hundred eV.

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Probing the correlated dynamics of two electrons in helium with synchrotron radiation has a long tradition. Recent absorption experiments in the energy range of isolated resonances of the helium atom show good agreement with *ab initio* quantum calculations [1] and can be interpreted in terms of approximate quantum numbers and propensity rules [2,3]. Above the double-ionization threshold $E=0$ the experimentally obtained angular distributions of both continuum electrons [4] are generally in agreement with calculations [5]. Surprisingly, a less detailed observable, the ratio σ^{++}/σ^{+} between double and single photoionization, which has been frequently measured [6–13], is only in fair agreement with theoretical predictions [14–19]. Moreover, the theoretical results do not agree with each other and no calculations exist for the threshold region around ≈ 80 eV photon energy, apart from the well known Wannier predictions for the double-ionization threshold itself [20,21].

In this Rapid Communication we will present a simple theoretical description of the photoabsorption process in helium, which is based on the assumption that double ionization is a two-step process: First, one electron absorbs the photon; in a second step, energy is transferred from one electron to another electron through a collision, so that both electrons can escape the nucleus. The first step is observable through the total cross section $\sigma_D(\omega)$, which is the sum of single and double ionizing events, as far as they are energetically possible, $\sigma_D(\omega)=\sigma^{++}(\omega)+\sigma^{+}(\omega)$. We will calculate $\sigma_D(\omega)$ analytically within a quasiclassical reflection approximation [22].

Process (B), the partition of the cross section between σ^{++} and σ^{+}, is mainly a consequence of the correlated motion of the two electrons *after* the absorption of the photon. The partition is observable through the ratio σ^{++}/σ^{+}. From the moment of the absorption on, the dynamics takes place on the new energy shell, which, for the present work, is located in the double continuum of the helium atom.

The correlated two-electron dynamics is represented by a semiclassical propagator of total angular momentum $L=0$ in a restricted two-dimensional configuration space, spanned by the two electron-nucleus distances r_i with fixed interelectronic angle $\theta=\pi$. These approximations may be justified as follows. It is known that helium resonances that differ only by the quantum number of total angular momentum are close in energy (for instance, $3s^2,{}^1S^e$ and $3s3p,{}^1P^o$). Hence, the corresponding difference between the S- and P-wave cross sections in the double continuum is also small. It will even be smaller for the *ratio* σ^{++}/σ^{+}, which is reasonably approximated by the S-wave cross section.

Total cross sections are dominated by contributions from angles that are fixed points of the classical dynamics. In the case of two electrons the fixed points are $\theta=\pi$ and $\theta=0$. However, only in the high energy limit, when the electron-electron interaction is a sufficiently weak perturbation, will the fixed point $\theta=0$ become important. For energies considered here we will calculate σ^{++}/σ^{+} only at the fixed point $\theta=\pi$.

To simulate the helium ground state classically we take an analytic phase-space distribution that is close to the various planar periodic orbits presently discussed for the helium atom [23]. We put the two electrons in a completely out of phase motion on a quarter circle of fixed hyperradius $\mathcal{R}_0\equiv(r_1^2+r_2^2)^{1/2}=1.4$ (atomic units will be used unless stated otherwise). This value comes from the average hyperradius (4.15 in energy scaled atomic units, see [23]) divided by the appropriate energy for the ground state (which is for the corresponding classical orbit roughly -2.97 a.u. in WKB

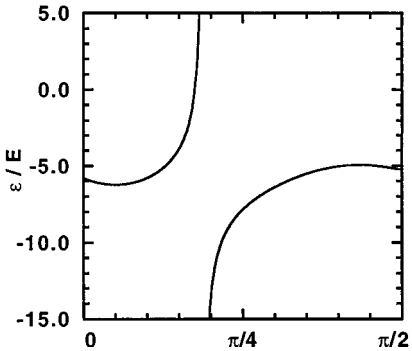

FIG. 1. Classical deflection function (see text) for the helium electron pair after absorption of a photon.

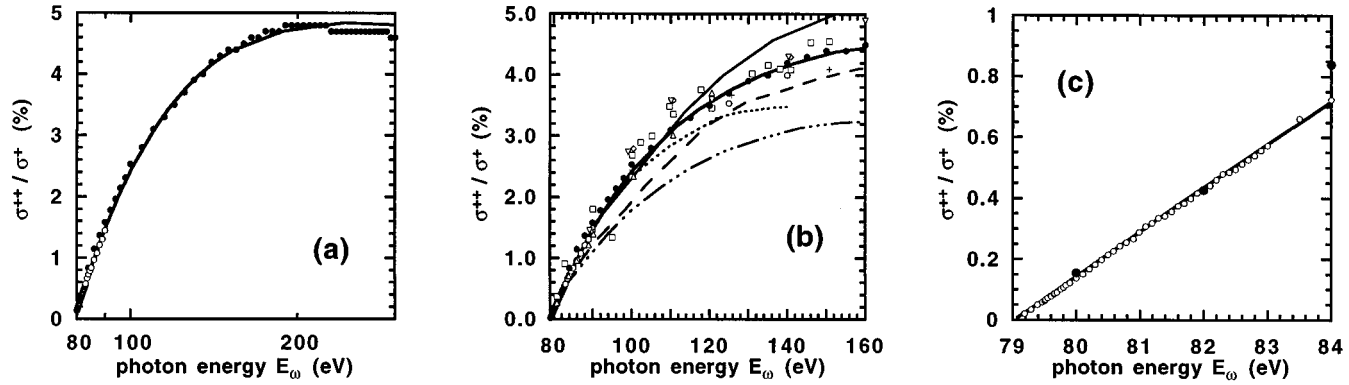

FIG. 2. Ratio of double to single photoionization, part (a) in logarithmic scale, parts (b) and (c) details in linear scale for the region of the maximum of the cross section and the threshold, respectively. Bold solid line, theory according to Eq. (5), other theories (curves have been graphically extracted from the respective publications); solid line, [14]; dotted line, [17]; dashed line, [18] (acceleration gauge, smoothed); dotted-dashed line, [15] (velocity gauge), data from [19] are similar but not shown; experiments: ○, [9]; ●, ''experimentally recommended data'' [13]; □, [12]; ◇, [8]; +, [6]; △, [11]; ▽, [7].

quantization [23]). With the interelectronic angle $\theta=\pi$, the resulting phase-space distribution is a one-parameter manifold dependent on the phase $0\leqslant\eta\leqslant\pi/2$ of the orbit on the circle of radius $\mathscr{R}_0$. However, the phase-space distribution to be propagated must be on the final energy shell E, and this is achieved by boosting the momentum of one electron to the appropriate new energy shell, $p_1'\rightarrow p_1'+2\omega$ where $\omega=E-E'$. (We use unmarked variables for the final state and primes for variables before the propagation.) The dipole amplitude in length form including the relevant semiclassical propagator for the dynamics of the electron pair after the absorption reads

$$d(\epsilon,E',E)=\sum_j \sqrt{\mathscr{D}_j(\epsilon,E',E)}\exp[i\Phi_j-i\nu_j\pi/2], \quad (1)$$

with

$$\mathscr{D}_j(\epsilon,E',E)=\frac{2}{\pi}\left|\frac{d\eta'}{d\epsilon}\right|_j [r_1'(\epsilon)+r_2'(\epsilon)]^2 \quad (2)$$

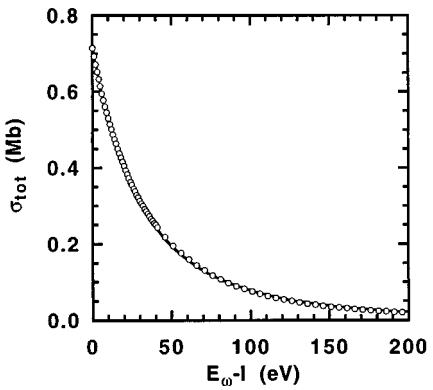

FIG. 3. Total photo cross section: full line, Eq. (7); circles, experiment by Samson *et al.* [27].

the classical probability for the jth orbit leading to a final energy ϵ of one electron following photon impact. Φ_j is the classical action and ν_j the Maslov index of the jth orbit [24], while $r_1'(\epsilon)=\mathscr{R}_0\cos\eta'(\epsilon)$ and $r_2'=\mathscr{R}_0\sin\eta'$ are the positions of the electrons before the absorption.

The essential object in Eq. (1) is the classical deflection function $\epsilon(\eta')$ (Fig. 1). It is monotonic apart from two small intervals $\Delta\eta'$ where artificial extrema are formed due to caustics. We will circumvent the problem that results from the caustics by approximating the total ionization probability in the restricted phase space classically,

$$P_D(E',E)=\int_{-\infty}^{\infty}\sum_j \mathscr{D}_j(\epsilon,E',E)d\epsilon=\mathscr{R}_0^2\left(1+\frac{2}{\pi}\right). \quad (3)$$

For the double ionizing events ($0\leqslant\epsilon\leqslant E$) only *one* trajectory contributes to the sum in Eq. (2) (see Fig. 1). However, we must take into account the Pauli principle for the identical electrons and add to the contribution from the trajectory with final energy ϵ the amplitude from the trajectory where the other electron has energy ϵ. The action is invariant under electron exchange, $\Phi(\epsilon,E',E)=\Phi(E-\epsilon,E',E)$. Hence, the differential probability for finding one electron with energy ϵ in a double ionized state after photon impact reads

$$P_D(\epsilon,E',E)=|\mathscr{D}(\epsilon,E',E)^{1/2}+\mathscr{D}(E-\epsilon,E',E)^{1/2}|^2. \quad (4)$$

From Eq. (4) we easily obtain the probability P_D^{++} for double photoionization

$$P_D^{++}(E',E)=\int_0^{E/2} P_D(\epsilon,E',E)d\epsilon. \quad (5)$$

The desired ratio of σ^{++}/σ^{+} is now given by $P_D^{++}/(P_D-P_D^{++})$ and is shown in Fig. 2 along with experimental and other theoretical results. Good agreement is achieved in the intermediate region with the ''experimentally recommended data'' [13] [parts (a) and (b)] and with the data

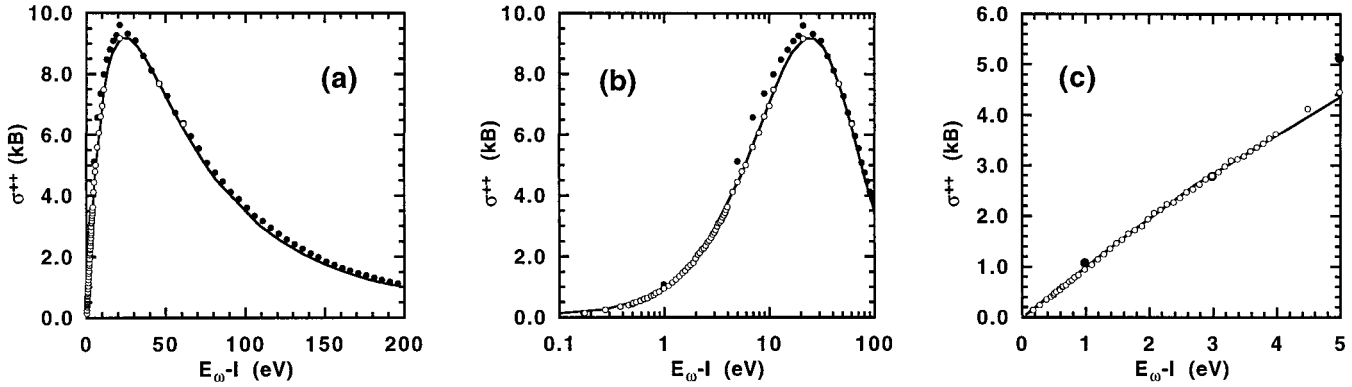

FIG. 4. The absolute double photoionization cross section as a function of the energy above the double-ionization threshold. Coding of the points as in Fig. 2. Note that the absolute experimental data from Kossmann *et al.* [9] have been obtained from $\sigma^{++}=\rho/(\rho+1)\sigma_D$, where $\rho=\sigma^{++}/\sigma^{+}$ and σ_D has been taken from [27]. For a discussion of this procedure see [17,13].

by Kossmann *et al.* [9] near threshold [part (c)]. Note that the present approach reproduces the classical result by Wannier in the limit $E\rightarrow 0$, namely, $\sigma^{++}\propto E^{1.056}$. For a realistic judgment of the agreement with the experiment in Fig. 2 one should keep in mind the approximations of the present approach and the experimental errors of 3% to 5% [13].

To obtain *absolute* cross sections, we calculate first the total photo cross section,

$$\sigma_D(\omega)=\frac{2\pi\alpha}{\omega}\int_{-\infty}^{+\infty}dt\langle\Psi|\vec{D}^{\dagger}e^{-iHt}\vec{D}|\Psi\rangle e^{iEt}, \qquad (6)$$

with the dipole operator $\vec{D}$ and the fine structure constant α. The total final energy is $E=E_i+\omega$, where E_i is the initial energy $E_i=2.848$ a.u. obtained by the ground state wave function $\Psi\propto\exp[-\beta(r_1+r_2)]$ with effective charge $\beta=2-5/16$ [25]. We use the reflection principle that has been developed for photodissociation in molecules [22] to obtain an analytic expression for σ_D. The propagator $\exp(-iHt)$ in Eq. (6) is approximated classically, which reduces the time integral to a δ function linking the electron radius to the photon frequency, $r=\omega^{-1/2}$. The result is [26]

$$\sigma_D(\omega)=\frac{1}{\beta^2}\frac{\alpha\pi^2 2^7}{3\sqrt{8}}x^{-7/2}e^{-\sqrt{8/x}}, \qquad (7)$$

where $x=2\omega/\beta^2$. The cross section from Eq. (7) is compared in the relevant energy range with recent experimental data [27] in Fig. 3.

Having completed the calculation of the two separate processes (A) and (B), we can now calculate the absolute double photoionization cross section from Eqs. (3) and (5),

$$\sigma^{++}=\frac{P_D^{++}}{P_D}\sigma_D. \qquad (8)$$

The result is shown in Fig. 4. Part (a) presents an overview, part (b) the region around the maximum on a logarithmic scale to emphasize smaller energies, and part (c) the threshold region. While the overall agreement is good one sees [as in Fig. 2(a)] an increasing deviation towards higher energies of several hundred eV. As mentioned above, this is to be expected since the calculation of process (B), the propagation of the correlated two electron dynamics after the absorption of the photon, has been carried out in a restricted phase space only.

A full semiclassical propagation of the electron pair would certainly improve the asymptotic behavior for $E\rightarrow\infty$. Another problem which must be solved in this context is the representation of the initial state. Here, it has been modeled by a phase-space distribution on a fixed hyperradius $\mathscr{R}_0$. To describe stationary states of helium classically is a difficult problem since almost all two-electron trajectories autoionize because of the missing lower limit for the negative energy. Conceptual improvement of the present approach will depend mainly upon future ideas of how to model a two-electron ground state classically.

One might find it surprising that the total cross sections for atomic double photoionization far from threshold can be described by an electron motion restricted to the collinear phase space. However, again, this can be understood from a similar behavior below threshold that is reflected by the propensity rules for photoabsorption from the ground state into doubly excited states of helium [2,3]. There, it is found that predominantly two-electron resonant states whose geometry in a body-fixed frame approaches for high excitation energies a collinear configuration are populated.

More importantly from a general perspective, the seemingly oversimplified picture of atomic double photoionization as a two-step process is confirmed by the quantitative comparison with the experiment. Recently, this two-step process has been independently suggested to understand the distribution of the recoil momentum of the He^{2+} ion after double photoionization [28]. The distribution is directly observable in experiments performed by cold target recoil ion momentum spectroscopy [28].

To summarize we have presented a quantum-classical hybrid approach for the calculation of *absolute* single- and double-ionization cross sections following photon impact on helium. The ratio σ^{++}/σ^{+} has been calculated *semiclassi-*

cally by propagation of the two electron dynamics starting from a classical initial phase-space distribution. The absolute total cross section has been calculated analytically with a standard quantum wave function for the helium ground state and a classical reflection approximation for the dynamics.

For collision physics with massive projectiles and in cases without resonances from internal excitation, the present hybrid approach might also prove useful to bridge the gap between threshold behavior and the asymptotic validity of the Born approximation in the calculation of ionization cross sections.

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