Total quadruple photoionization cross section of beryllium

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In a quasiclassical framework, we formulate the quadruple ionization by single-photon absorption of the Coulomb five-body problem. We present the quadruple photoionization total cross section of the ground state of beryllium for energies up to 620 eV. Our results for energies close to threshold are in agreement with the Wannier threshold law for four-electron escape. In addition, the agreement of our results with a shape formula provides support for the overall shape of our total quadruple cross section. Finally, we find that the photon energy where the maximum of the total photoionization cross section occurs for single, double, triple, and quadruple photoionization of H, He, Li, and Be, respectively, seems to follow a linear relation with the threshold energy for complete breakup of the respective element.

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I. INTRODUCTION

Multiple ionization of atoms by single-photon absorption is a process of great interest since it probes the correlated motion of many electrons. Quadruple photoionization of the ground state of beryllium is the most fundamental atomic process involving four bound electrons. No other theoretical or experimental study is, to our knowledge, currently available regarding escape of four initially bound electrons.

The ionization processes involving two-electron escape can now be treated quite accurately and significant progress has been made concerning their understanding [1]. Regarding the three-electron escape by single-photon absorption from the ground state of Li, significant advances have been made over the last few years in obtaining total triple ionization cross sections both experimentally [2,3] and theoretically [4–6]. However, still great challenges remain, particularly concerning differential cross sections [7–10].

Triple ionization of the ground state of Li by electron impact, that is, a four-electron escape process, has been addressed in very recent years. These experimental [11] and theoretical [12] studies address very high energies of the impacting electron. The current theoretical study is the first, to our knowledge, work on four-electron escape by single-photon absorption for up to intermediate excess energies. Our study is based on an extension of the half collision model. The latter is based on Samson's [13] original idea for double photoionization: the ejection of the second electron resembles electron impact ionization by the primary electron that absorbs the photon and leaves the atom. We formulate the extension of the half collision model for the quadruple ionization in a quasiclassical framework. That is, we propagate classical trajectories using the classical trajectory Monte Carlo (CTMC) method [14,15] with a Wigner distribution as our initial state. This simple model has proven very successful in describing the double photoionization from the ground state of He [16] as well as the singly excited states of He [17]. The same simple model has also accurately described the total photoionization cross section [6], as well as single differential cross sections [9] and double energy differential cross sections [10] for the three-electron escape from the ground state of Li for up to intermediate excess energies. The significance of studying quadruple photoionization using a simple model, which has already proven very successful in describing several fragmentation processes, is twofold: First, given the challenges experimental as well as exact quantum mechanical studies will face in the future, our results will serve as a benchmark calculation for these studies of the total quadruple photoionization cross section. Second, given that close to threshold the quadruple ionization is a prohibitively rare process for *ab initio* studies with the current state of the art computational capabilities one can hope to gain physical insight only using simple but accurate models. Our current study is a first step in this direction. In the future our studies will address the shape of the interelectronic angular distributions of the four escaping electrons and whether an unexpected pattern arises for energies close to threshold. That was found to be the case for Li with the three electrons escaping in a surprising T-shape structure for excess energies close to threshold [6].

II. QUASICLASSICAL FORMULATION OF QUADRUPLE IONIZATION OF BERYLLIUM

We formulate the quadruple photoionization process from the Be ground state ($1s^2 2s^2$) as a two step process (see Refs. [4,6,13,16]) in the following way:

$$\sigma^{4+} = \sigma_{\mathrm{abs}} P^{4+}, \tag{1}$$

where σ_{abs} is the total photoabsorption cross section and P^{4+} is the probability for quadruple ionization. For σ_{abs} we use the experimental data from Ref. [19]. Our formulation accounts for the second step (half collision model). Our construction of the initial phase space density $\rho(\gamma)$ for the quadruple photoionization of beryllium is similar to the double photoionization of He [16] and the triple photoionization formulation of Li, which has been detailed in [6]. First, one electron absorbs the photon (photoelectron) at time $t=t_{\mathrm{abs}}=0$. Through electronic correlations, the energy is redistributed, resulting in four electrons escaping to the continuum. We first assume that the photon is absorbed by a $1s$ electron at the nucleus ($\mathbf{r}_1=0$). This latter approximation becomes exact in the limit of high photon energy [20]. For Be,

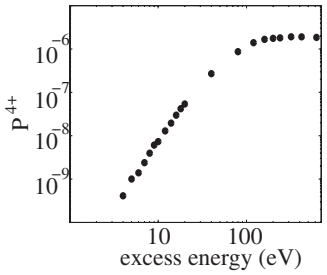

FIG. 1. Quadruple photoionization probability as a function of excess energy.

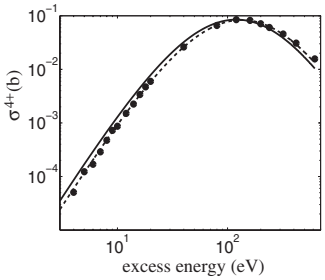

FIG. 2. Quadruple photoionization cross section as a function of excess energy. The black circles are our results. For the solid and dashed lines see the main text.

at the energies we consider, the cross section for photon absorption from a $1s$ orbital is much larger than from a $2s$ orbital [21]. Hence, we can safely assume that the photoelectron is a $1s$ electron, which significantly reduces the initial phase space to be sampled. We also neglect antisymmetrization of the electrons in the initial state. We denote the photoelectron by 1, the other $1s$ electron by 2, and the two $2s$ electrons by 3 and 4, respectively. Following photon absorption, we model the initial phase space distribution of the remaining three electrons, $1s$ and two $2s$, by the Wigner transform [22] of the corresponding initial wave function $\psi(\mathbf{r}_1=0,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)$, where $\mathbf{r}_i$ are the electron vectors starting at the nucleus. We approximate the initial wave function as a simple product of hydrogenic orbitals $\phi_i^{Z_i}(\mathbf{r}_i)$, with effective charges Z_i, to facilitate the Wigner transformation. The Z_i are chosen so that they reproduce the known ionization potentials I_i, namely, for one of the two $2s$ electrons $Z_4=1.656$ ($I_4=0.343$ a.u.), for the other $2s$ electron $Z_3=2.314$ ($I_3=0.669$ a.u.), and for the $1s$ electron $Z_2=3.363$ ($I_2=5.656$ a.u.). (We use atomic units throughout the paper if not stated otherwise.) The excess energy E is given by $E=E_\omega-I$ with E_ω the photon energy and $I=14.67$ a.u. the Be quadruple ionization threshold energy. Given the above considerations, the initial phase space density is given by

$$\rho(\gamma)=\mathcal{N}\delta(\mathbf{r}_1)\delta(E_1+I_1-\omega)\prod_{i=2,3,4}W_{\phi_i^{Z_i}}(\mathbf{r}_i,\mathbf{p}_i)\delta(E_i+I_i), \tag{2}$$

with normalization constant $\mathcal{N}$. We determine the quadruple ionization probability P^{4+} by discretizing the initial phase space. We then propagate the classical equations of motion using the discretized phase space as initial conditions. The full Coulomb five-body Hamiltonian is used for the classical propagation. Regularized coordinates [23,24] are used to avoid problems with electron trajectories starting at the nucleus. We label as quadruply ionizing those trajectories where the energies of all four electrons are positive, $E_i>0$ with $i=1,2,3,4$, asymptotically in time.

III. RESULTS

In Fig. 1, we present our results for P^{4+}. The very small value of the quadruple photoionization probability clearly shows the difficulties involved in obtaining theoretical results or experimental measurements of the five-body breakup process. Using Eq. (1) we present in Fig. 2 the results for the quadruple photoabsorption cross section (black circles). Since there are no other results currently available we cannot compare our results on an absolute scale.

However, a comparison of our results with a shape function applicable to single-photon multiple ionization processes [25] provides some indication of how well our results reproduce the shape of the total cross section. This shape function reproduces, by construction, the correct behavior of the single-photon multiple-ionization cross section for excess energies close to threshold (Wannier law [26]) and for large excess energies. It depends on two parameters, the position E_M and height σ_M of the cross section maximum and is given by

$$\sigma(E)=\sigma_M x^\alpha\left(\frac{\alpha+7/2}{\alpha x+7/2}\right)^{\alpha+7/2}, \tag{3}$$

where α is the characteristic exponent in the Wannier threshold law and x is the excess energy E scaled by E_M. For the breakup of a four-electron atom this characteristic exponent was found to be 3.331 for a plane configuration where the electrons escape on the apexes of a square and 3.288 for a three-dimensional configuration where the electrons escape on the apexes of a tetrahedron [18]. From the above two values for α it is the smallest one, $\alpha=3.288$, that dominates the threshold behavior and thus the value we use in the shape formula in Eq. (3). Using the shape formula without any fitting parameters but substituting for $E_M=120$ eV and $\sigma_M=0.0847$ b, which are the location and height of the maximum cross section from our results, we obtain the solid curve shown in Fig. 2. Given that there are no fitting parameters the agreement between our numerical results and the shape formula is quite good, providing support that the shape of the cross section we obtain is the correct one. The agreement becomes even better if instead we use the E_M and σ_M as fitting parameters to fit our results with the shape formula. Doing so, we obtain the values $E_M=134.5$ eV and $\sigma_M=0.0834$ b and the agreement of the fitted shape formula (dashed curve in Fig. 2) with our results is indeed very good.

Also, for small values of the excess energy we have fitted our results for the quadruple cross section with the formula

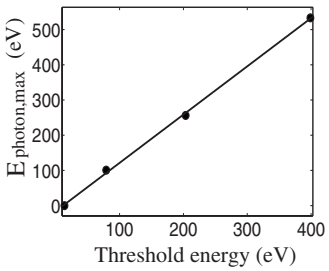

FIG. 3. Photon energy where the maximum of the cross section for single photoionization of H, double photoionization of He, triple photoionization of Li, and quadruple photoionization of Be occurs as a function of the threshold energy of the respective process.

$$\sigma(E_\omega) \propto (E_\omega/I - 1)^\alpha, \tag{4}$$

with I the fragmentation threshold energy of beryllium and E_ω the photon energy. For energies from 4 to 9 eV our results yield a characteristic exponent α of 3.17, which is quite close to the analytical value of 3.288. The difference can probably be attributed to the fact that our current results go only down to 4 eV. It is a well known fact that as the energy increases the value of α decreases [6]. Thus, most probably, if we were able to obtain results for energies below 4 eV α would have been closer to the actual value of 3.288. Currently, however, the numerical difficulties involved in obtaining results below 4 eV are prohibitive.

At this point, a few comments are in order. Our quasiclassical formulation is most accurate close to threshold. It can describe the process accurately up to at most intermediate excess energies while it cannot account for high energies. For high energies the process can only be described quantum mechanically. In the case of triple ionization from the ground state of Li our simple product initial state has proven to be very good in describing not only the total cross section [6] but also single [9] and double energy differential cross sections up to excess energies of 220 eV (see [10]), with $I=203.5$ eV for the triple escape process. In the current paper we consider only a total absorption cross section and not differential ones with the former being much less sensitive than the latter to electron-electron correlation. The success of our product initial state in describing single and double differential cross sections for double [16] and triple photoionization [9,10], which are more sensitive to electron-electron correlation compared to the total cross section, strongly suggests the success of our model in describing accurately the total quadruple photoionization cross section up to intermediate excess energies.

A comparison of the maximum of the single photoionization cross section for H, 6.3×10^6 b, with the maxima for double photoionization of He, 8.76×10^3 b [27], triple photoionization of Li, 7.47 b, and quadruple photoionization of Be, 0.0847 b, clearly shows how rare the quadruple photoionization process is. For the double ionization cross section of He we use the results presented in Ref. [27] while for the triple photoionization of Li we use the results presented in Ref. [6]. Another interesting feature is the relation between the ionization threshold energy and the photon energy $E_{\omega,M}=I+E_M$, where the maximum of the photoionization cross section of the respective process occurs. For the single photoionization of H, $E_M=0$ eV, for the double photoionization of He, $E_M=21.8$ eV [27], while for the triple and quadruple photoionization of Li and Be, E_M, is, respectively, 52.7 and 134.4 eV. The values of 52.7 and 134.4 eV were found by fitting Eq. (3) to our results for triple [6] and quadruple photoionization, respectively. In Fig. 3, we plot $E_{\omega,M}$ as a function of I for the above four processes. Interestingly, we find that for these four processes the maximum of the total photoionization cross section seems to follow a linear relation with the threshold energy for complete breakup of the respective element. Whether there is a physical reason underlying this fact is an interesting question for future research.

A future study of interest for the four-electron escape in Be would be to investigate whether the collision processes the four electrons follow to escape to the continuum can still be described as a sequence of three-body subsystems as is the case for Li [9]. Another interesting aspect to be investigated is the shape of the interelectronic angular distributions for energies close to threshold. In the case of the three-electron escape from the ground state of Li we found that in an energy regime where the characteristic exponent $\alpha=2.16$ [28] is recovered the angular distribution has a surprising T-shape distribution. This T shape is different than what one would expect from the three electrons escaping in the apexes of an equilateral triangle for excess energy zero [28]. At zero excess energy, the four electrons escape in the apexes of a tetrahedron [18]. It remains to be seen whether a surprising pattern will be found for the four-electron escape as was the case for the three electrons.

In conclusion, we have presented results for the total cross section for the quadruple photoionization of Be. With no other theoretical or experimental results currently available we hope that the current results will serve as a benchmark calculation for future studies of the Coulomb five-body problem.

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