

Quadruple Photoionization of Be

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ABSTRACT: A theoretical method is developed to solve the time-dependent Schrodinger equation on a 4D radial lattice and used to calculate the quadruple photoionization cross sections for the Be atom. At a photon energy of 500 eV cross sections are compared with previous quasiclassical simulations.

1. INTRODUCTION

A theoretical method developed to solve the time-dependent Schrodinger equation on a 3D radial lattice (TDSE-3D) has been used to calculate the triple photoionization of atoms. Total cross sections for the triple photoionization of Li using the TDSE-3D method[1, 2] were found to be in reasonable agreement with experimental measurements[3]. Energy differential cross sections for the triple photoionization of Li[4] at an incident energy of 300 eV were found to be bowl shaped. Angular distributions for the triple photoionization of Li[5] yielded evidence for a T-shaped breakup pattern as predicted by quasiclassical simulations [6, 7]. Energy and angle differential cross sections were also compared for the one-photon and two-photon triple ionization of Li using the TDSE-3D method[8].

Recently a theoretical method developed to solve the time-dependent Schrodinger equation on a 4D radial lattice (TDSE-4D) was used to calculate the triple autoionization of atomic ions[9] in support of experimental measurements[10]. In this paper the TDSE-4D method is further developed and used to calculate the quadruple photoionization of atoms. Total cross sections for the quadruple photoionization of Be at an incident energy of 500 eV are compared with quasiclassical simulations[11].

Details of the TDSE-4D method for four electron systems are presented in section 2, quadruple photoionization cross sections for the Be atom are presented in section 3, and a brief summary is given in section 4. Unless otherwise stated, all quantities are given in atomic units.

2. THEORY

For a four-electron target atom, the TDSE-4D equations are given by:

$$\begin{aligned}
 & i \frac{\partial P_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(r_1, r_2, r_3, r_4, t)}{\partial t} \\
 &= T_{l_1 l_2 l_3 l_4}(r_1, r_2, r_3, r_4) P_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(r_1, r_2, r_3, r_4, t) \\
 &+ \sum_{l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4} \sum_{i < j}^6 V_{l_1 l_2 L_{12} l_3 L_{123} l_4, l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^L(r_i, r_j) \\
 &\quad \times P_{l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^L(r_1, r_2, r_3, r_4, t) \\
 &+ \sum_{l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4} \sum_i^4 W_{l_1 l_2 L_{12} l_3 L_{123} l_4, l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^{LL_0}(r_i, t) \\
 &\quad \times \bar{P}_{l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^{L_0}(r_1, r_2, r_3, r_4, \tau \rightarrow \infty), \tag{1}
 \end{aligned}$$

where $P_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(r_1, r_2, r_3, r_4, t)$ is the radial part of the time evolving total wavefunction and $\bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau \rightarrow \infty)$ is the radial part of the initial total wavefunction.

The l_1, l_2, l_3, l_4 are the angular momenta of each of the four electrons, where l_1 coupled to l_2 yields L_{12} , L_{12} coupled to l_3 yields L_{123} , and L_{123} coupled to l_4 yields L for the time evolving total wavefunction and L_0 for the initial total wavefunction. The kinetic and nuclear potential operator, $T_{l_1 l_2 l_3 l_4}(r_1, r_2, r_3, r_4)$, the six electrostatic repulsion operators, $V_{l_1 l_2 L_{12} l_3 L_{123} l_4, l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^L(r_i, r_j)$, and the four dipole radiation field operators, $W_{l_1 l_2 L_{12} l_3 L_{123} l_4, l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^{LL_0}(r_i, t)$, are all given in the Appendix. The atomic number of the atom is Z , the coupling operator expressions use standard $3j$ and $6j$ symbols, in the length gauge $g(r) = r$, and the linearly polarized electric field amplitude $F(t)$ is proportional to $\cos \omega t$, where ω is the radiation field frequency.

The radial wavefunctions, $\bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau)$ are obtained by relaxation of the Schrodinger equation in imaginary time given by:

$$\begin{aligned}
 & - \frac{\partial \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau)}{\partial \tau} \\
 &= T_{l_1 l_2 l_3 l_4}(r_1, r_2, r_3, r_4) \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau) \\
 &+ \sum_{l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4} \sum_{i < j}^6 V_{l_1 l_2 L_{12} l_3 L_{123} l_4, l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^{L_0}(r_i, r_j) \\
 &\quad \times \bar{P}_{l'_1 l'_2 L'_{12} l'_3 L'_{123} l'_4}^{L_0}(r_1, r_2, r_3, r_4, \tau). \tag{2}
 \end{aligned}$$

As an example, for $\text{Be}(1s^2 2s^2)$ the radial wavefunctions at time $\tau = 0$ are given by:

$$\begin{aligned}
 & \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau = 0) \\
 &= P_{1s}(r_1) P_{1s}(r_2) P_{2s}(r_3) P_{2s}(r_4) \delta_{l_1,0} \delta_{l_2,0} \delta_{l_{12},0} \delta_{l_3,0} \delta_{L_{123},0} \delta_{l_4,0} \delta_{L_0,0}, \tag{3}
 \end{aligned}$$

where $P_{1s}(r)$ and $P_{2s}(r)$ are radial wavefunctions for Be^{3+} . To prevent relaxation to any state involving three or four electrons with $1s$ character, a Schmidt orthogonalization is made at every imaginary time step:

$$\begin{aligned}
& \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau) \\
&= \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r_4, \tau) \\
&- \int_0^\infty P_{1s}(r'_3) \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r'_3, r_4, \tau) dr'_3 P_{1s}(r_3) \\
&- \int_0^\infty P_{1s}(r'_4) \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r_3, r'_4, \tau) dr'_4 P_{1s}(r_4) \\
&+ \int_0^\infty \int_0^\infty P_{1s}(r'_3) P_{1s}(r'_4) \bar{P}_{l_1 l_2 L_{12} l_3 L_{123} l_4}^{L_0}(r_1, r_2, r'_3, r'_4, \tau) dr'_3 dr'_4 \\
&\quad \times P_{1s}(r_3) P_{1s}(r_4) .
\end{aligned} \tag{4}$$

The initial condition for the solution of the TDSE-4D equations of Eq.(1) are given by:

$$P_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(r_1, r_2, r_3, r_4, t = 0) = 0 . \tag{5}$$

The quadruple photoionization cross section for $t \rightarrow \infty$ is given by:

$$\sigma = \frac{\omega}{I} \sum_{l_1 l_2 L_{12} l_3 L_{123} l_4} \frac{\partial Q_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(t)}{\partial t} \tag{6}$$

where

$$\begin{aligned}
Q_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(t) &= \sum_{k_1} \sum_{k_2} \sum_{k_3} \sum_{k_4} \int_0^\infty dr_1 \int_0^\infty dr_2 \int_0^\infty dr_3 \int_0^\infty dr_4 \\
&\times P_{k_1 l_1}(r_1) P_{k_2 l_2}(r_2) P_{k_3 l_3}(r_3) P_{k_4 l_4}(r_4) \\
&\times P_{l_1 l_2 L_{12} l_3 L_{123} l_4}^L(r_1, r_2, r_3, r_4, t) .
\end{aligned} \tag{7}$$

Care must be taken in the sums over electron momentum, k_i , to avoid double counting of distinct continuum states. We also restrict the sums over electron momentum so that the conservation of energy:

$$E_{atom} + \omega = \frac{k_1^2}{2} + \frac{k_2^2}{2} + \frac{k_3^2}{2} + \frac{k_4^2}{2} \tag{8}$$

is approximately conserved.

3. RESULTS

We employ a radial grid with a uniform mesh spacing of $\Delta r = 0.10$ with a total number of points $N = 360$. A complete set of bound and continuum orbitals are generated by matrix diagonalization of a one-electron Hamiltonian. For Be^{3+} using a Coulomb potential we obtain $\epsilon_{1s} = 209.6$ eV and $\epsilon_{2s} = 53.9$ eV.

Upon relaxation of the time-dependent close-coupling equations in imaginary time, using a $(360)^4$ point lattice, it was found that a relaxation time of 10.0 au ($\Delta \tau = 0.01$) yielded a ground state energy of -381.0 eV. The relaxation used one coupled channel for $L_0 = 0$ with $l_1 = 0, l_2 = 0, l_3 = 0, l_4 = 0, L_{12} = 0, L_{123} = 0$. The experimental Be ground state energy is -399.0 eV.

Once a fully correlated initial state was obtained, the time-dependent close-coupling equations were propagated in real time for 10 periods ($\Delta t = 0.002$) using the same $(360)^4$ point lattice. We note that the TDSE-4D equations for both imaginary time relaxation and real time propagation used a $(360)^4$ point lattice partitioned over 20,736 cores on a massively parallel computer. The propagation used four coupled channels for $L = 1$ with $l_1 = 1, l_2 = 0, l_3 = 0, l_4 = 0, L_{12} = 1, L_{123} = 1; l_1 = 0, l_2 = 1, l_3 = 0, l_4 = 0, L_{12} = 1, L_{123} = 1; l_1 = 0, l_2 = 0, l_3 = 1, l_4 = 0, L_{12} = 0, L_{123} = 1;$ and $l_1 = 0, l_2 = 0, l_3 = 0, l_4 = 1, L_{12} = 0, L_{123} = 0$. For $\omega = 500.0$ eV, and an excess energy of 119.0 eV, and an intensity $I = 10^{14}$ W/cm², we obtained a cross section of 3.2×10^{-28} cm². The cross section obtained by Emmanouilidou[11] using a quasiclassical simulation at an excess energy of 120.0 eV is 8.4×10^{-26} cm².

4. SUMMARY

In conclusion, we have applied the TDSE-4D method to calculate quadruple photoionization cross sections for the Be atom. Our first calculation with a restricted minimal number of coupled channels at a photon energy of 500 eV yielded a cross section much smaller than that obtained using a quasiclassical simulation method[11]. Larger calculations are necessary before a detailed comparison can be made with previous work[11].

In the future we plan to extend our TDSE-4D calculations for the quadruple photoionization of Be using a smaller uniform mesh spacing Δr , more mesh points N , and more coupled channels $l_1 l_2 L_{12} l_3 L_{123} l_4 L$. It will take extended runs on massively parallel supercomputers to obtain a fully converged cross section.

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5. APPENDIX

$$T(r_1, r_2, r_3, r_4) = \sum_i^4 \left(-\frac{1}{2} \frac{\partial^2}{\partial r_i^2} + \frac{l_i(l_i + 1)}{2r_i^2} - \frac{Z}{r_i} \right) \quad (9)$$

$$\begin{aligned} V^L(r_1, r_2) &= (-1)^{l_1+l'_1+L_{12}} \delta_{l_3, l'_3} \delta_{l_4, l'_4} \delta_{L_{12}, L'_{12}} \delta_{L_{123}, L'_{123}} \\ &\times \sqrt{(2l_1 + 1)(2l'_1 + 1)(2l_2 + 1)(2l'_2 + 1)} \\ &\times \sum_{\lambda} \frac{(r_1, r_2)_{\lambda}^{\lambda}}{(r_1, r_2)_{\lambda}^{\lambda+1}} \begin{pmatrix} l_1 & \lambda & l'_1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_2 & \lambda & l'_2 \\ 0 & 0 & 0 \end{pmatrix} \\ &\times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ l'_2 & l'_1 & \lambda \end{Bmatrix}, \end{aligned} \quad (10)$$

$$V^L(r_1, r_3) = (-1)^{l_2+L_{123}} \delta_{l_2, l'_2} \delta_{l_4, l'_4} \delta_{L_{123}, L'_{123}}$$

$$\begin{aligned} & \times \sqrt{(2l_1 + 1)(2l'_1 + 1)(2l_3 + 1)(2l'_3 + 1)(2L_{12} + 1)(2L'_{12} + 1)} \\ & \times (-1)^\lambda \sum_{\lambda} \frac{(r_1, r_3)_{<}^\lambda}{(r_1, r_3)_{>}^{\lambda+1}} \begin{pmatrix} l_1 & \lambda & l'_1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_3 & \lambda & l'_3 \\ 0 & 0 & 0 \end{pmatrix} \\ & \times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ L'_{12} & \lambda & l'_1 \end{Bmatrix} \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ l'_3 & L'_{12} & \lambda \end{Bmatrix}, \end{aligned} \quad (11)$$

$$\begin{aligned} V^L(r_1, r_4) &= (-1)^{l_2+l_3+L_{12}+L'_{12}+L} \delta_{l_2, l'_2} \delta_{l_3, l'_3} \\ & \times \sqrt{(2l_1 + 1)(2l'_1 + 1)(2l_4 + 1)(2l'_4 + 1)} \\ & \times \sqrt{(2L_{12} + 1)(2L'_{12} + 1)(2L_{123} + 1)(2L'_{123} + 1)} \\ & \times \sum_{\lambda} \frac{(r_1, r_4)_{<}^\lambda}{(r_1, r_4)_{>}^{\lambda+1}} \begin{pmatrix} l_1 & \lambda & l'_1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_4 & \lambda & l'_4 \\ 0 & 0 & 0 \end{pmatrix} \\ & \times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ L'_{12} & \lambda & l'_1 \end{Bmatrix} \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ L'_{123} & \lambda & L'_{12} \end{Bmatrix} \\ & \times \begin{Bmatrix} L_{123} & l_4 & L \\ l'_4 & L'_{123} & \lambda \end{Bmatrix}, \end{aligned} \quad (12)$$

$$\begin{aligned} V^L(r_2, r_3) &= (-1)^{l_1+l_2+L_{12}+L_{123}+l'_2+L'_{12}} \delta_{l_1, l'_1} \delta_{l_4, l'_4} \delta_{L_{123}, L'_{123}} \\ & \times \sqrt{(2l_2 + 1)(2l'_2 + 1)(2l_3 + 1)(2l'_3 + 1)(2L_{12} + 1)(2L'_{12} + 1)} \\ & \times (-1)^\lambda \sum_{\lambda} \frac{(r_2, r_3)_{<}^\lambda}{(r_2, r_3)_{>}^{\lambda+1}} \begin{pmatrix} l_2 & \lambda & l'_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_3 & \lambda & l'_3 \\ 0 & 0 & 0 \end{pmatrix} \\ & \times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ \lambda & L'_{12} & l'_2 \end{Bmatrix} \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ l'_3 & L'_{12} & \lambda \end{Bmatrix}, \end{aligned} \quad (13)$$

$$\begin{aligned} V^L(r_2, r_4) &= (-1)^{l_1+l'_2+l_3+L} \delta_{l_1, l'_1} \delta_{l_3, l'_3} \\ & \times \sqrt{(2l_2 + 1)(2l'_2 + 1)(2l_4 + 1)(2l'_4 + 1)} \\ & \times \sqrt{(2L_{12} + 1)(2L'_{12} + 1)(2L_{123} + 1)(2L'_{123} + 1)} \\ & \times \sum_{\lambda} \frac{(r_2, r_4)_{<}^\lambda}{(r_2, r_4)_{>}^{\lambda+1}} \begin{pmatrix} l_2 & \lambda & l'_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_4 & \lambda & l'_4 \\ 0 & 0 & 0 \end{pmatrix} \\ & \times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ \lambda & L'_{12} & l'_2 \end{Bmatrix} \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ L'_{123} & \lambda & L'_{12} \end{Bmatrix} \\ & \times \begin{Bmatrix} L_{123} & l_4 & L \\ l'_4 & L'_{123} & \lambda \end{Bmatrix}, \end{aligned} \quad (14)$$

$$V^L(r_3, r_4) = (-1)^{l_3+l'_3+L_{12}+L_{123}+L'_{123}+L} \delta_{l_1, l'_1} \delta_{l_2, l'_2} \delta_{L_{12}, L'_{12}}$$

$$\begin{aligned}
 & \times \sqrt{(2l_3 + 1)(2l'_3 + 1)(2l_4 + 1)(2l'_4 + 1)(2L_{123} + 1)(2L'_{123} + 1)} \\
 & \times (-1)^\lambda \sum_{\lambda} \frac{(r_3, r_4)_{\leq}^\lambda}{(r_3, r_4)_{>}^{\lambda+1}} \begin{pmatrix} l_3 & \lambda & l'_3 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_4 & \lambda & l'_4 \\ 0 & 0 & 0 \end{pmatrix} \\
 & \times \left\{ \begin{matrix} L_{12} & l_3 & L_{123} \\ \lambda & L'_{123} & l'_3 \end{matrix} \right\} \left\{ \begin{matrix} L_{123} & l_4 & L \\ l'_4 & L'_{123} & \lambda \end{matrix} \right\}, \quad (15)
 \end{aligned}$$

$$\begin{aligned}
 W(r_1, t) &= (-1)^{l_2+l_3+l_4+L_{12}+L'_{12}+L_{123}+L'_{123}+L+L_0} \delta_{l_2, l'_2} \delta_{l_3, l'_3} \delta_{l_4, l'_4} \\
 & \times \sqrt{(2l_1 + 1)(2l'_1 + 1)(2L_{12} + 1)(2L'_{12} + 1)} \\
 & \times \sqrt{(2L_{123} + 1)(2L'_{123} + 1)(2L + 1)(2L_0 + 1)} \\
 & \times g(r_1) F(t) \begin{pmatrix} l_1 & 1 & l'_1 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L_0 \\ 0 & 0 & 0 \end{pmatrix} \\
 & \times \left\{ \begin{matrix} L_{123} & l_4 & L \\ L_0 & 1 & L'_{123} \end{matrix} \right\} \left\{ \begin{matrix} L_{12} & l_3 & L_{123} \\ L'_{123} & 1 & L'_{12} \end{matrix} \right\} \\
 & \times \left\{ \begin{matrix} l_1 & l_2 & L_{12} \\ L'_{12} & 1 & l'_1 \end{matrix} \right\} \quad (16)
 \end{aligned}$$

$$\begin{aligned}
 W(r_2, t) &= (-1)^{l_1+l_2+l'_2+l_3+l_4+L_{123}+L'_{123}+L+L_0+1} \delta_{l_1, l'_1} \delta_{l_3, l'_3} \delta_{l_4, l'_4} \\
 & \times \sqrt{(2l_2 + 1)(2l'_2 + 1)(2L_{12} + 1)(2L'_{12} + 1)} \\
 & \times \sqrt{(2L_{123} + 1)(2L'_{123} + 1)(2L + 1)(2L_0 + 1)} \\
 & \times g(r_2) F(t) \begin{pmatrix} l_2 & 1 & l'_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L_0 \\ 0 & 0 & 0 \end{pmatrix} \\
 & \times \left\{ \begin{matrix} L_{123} & l_4 & L \\ L_{01} & L'_{123} & \end{matrix} \right\} \left\{ \begin{matrix} L_{12} & l_3 & L_{123} \\ L'_{123} & 1 & L'_{12} \end{matrix} \right\} \\
 & \times \left\{ \begin{matrix} l_1 & l_2 & L_{12} \\ 1 & L'_{12} & l'_2 \end{matrix} \right\} \quad (17)
 \end{aligned}$$

$$\begin{aligned}
 W(r_3, t) &= (-1)^{l_3+l'_3+l_4+L_{12}+L+L_0} \delta_{l_1, l'_1} \delta_{l_2, l'_2} \delta_{l_4, l'_4} \delta_{L_{12}, L'_{12}} \\
 & \times \sqrt{(2l_3 + 1)(2l'_3 + 1)(2L_{123} + 1)(2L'_{123} + 1)(2L + 1)(2L_0 + 1)} \\
 & \times g(r_3) F(t) \begin{pmatrix} l_3 & 1 & l'_3 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L_0 \\ 0 & 0 & 0 \end{pmatrix} \\
 & \times \left\{ \begin{matrix} L_{123} & l_4 & L \\ L_0 & 1 & L'_{123} \end{matrix} \right\} \left\{ \begin{matrix} L_{12} & l_3 & L_{123} \\ 1 & L'_{123} & l'_3 \end{matrix} \right\} \quad (18)
 \end{aligned}$$

$$W(r_4, t) = (-1)^{l_4+l'_4+L_{123}+1} \delta_{l_1, l'_1} \delta_{l_2, l'_2} \delta_{l_3, l'_3} \delta_{L_{123}, L'_{123}}$$

$$\begin{aligned}
 & \times \sqrt{(2l_4 + 1)(2l'_4 + 1)(2L + 1)(2L_0 + 1)} \\
 & \times g(r_4)F(t) \begin{pmatrix} l_4 & 1 & l'_4 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L_0 \\ 0 & 0 & 0 \end{pmatrix} \\
 & \times \left\{ \begin{array}{ccc} L_{123} & l_4 & L \\ 1 & L_0 & l'_4 \end{array} \right\} \tag{19}
 \end{aligned}$$



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