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Electron-Impact Triple Ionization of Li ($1s^22s$)

M. S. PINDZOLA[1] AND J. P. COLGAN[2]

[1]*Department of Physics, Auburn University, Auburn, AL*
[2]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM*

ABSTRACT: A time-dependent close-coupling method for four-electron atomic systems is used to calculate the electron-impact triple ionization of the ground-state Li atom. Cross sections are calculated at three different incident electron energies for the $\mathcal{L} = 0$ partial wave.

1. INTRODUCTION

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 the C^{2+} ($2s^22p^2$) excited atom ion[1] in support of experimental measurements[2]. The TDSE- 4D method was also used to calculate the quadrupole photoionization of the Be ($1s^22s^2$) atom[3] to compare with quasiclassical simulations[4].

In this paper we develop a time-dependent close-coupling method for four-electron atomic systems to calculate the electron-impact triple ionization of atoms. The TDSE-4D method is used to calculate the electron-impact triple ionization of the ground state of the Li atom at three different incident electron energies for the $\mathcal{L} = 0$ partial wave.

Details of the time-dependent close-coupling method for four electron systems are presented in section 2, triple ionization cross sections for the Li ($1s^22s$) ground state 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

The time-dependent Schrodinger equation for a four-electron atom is given by:

$$\frac{i\partial\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,\vec{r}_4,t)}{\partial t} = H_{atom}\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,\vec{r}_4,t) \quad (1)$$

where

$$H_{atom} = \sum_{i=1}^{4}\left(-\frac{1}{2}\nabla_i^2 - \frac{Z}{r_i}\right) + \sum_{i<j=1}^{4}\frac{1}{|\vec{r}_i - \vec{r}_j|}\ , \quad (2)$$

$\vec{r}_i$ are electron coordinates, and Z is the atomic number. The total electronic wavefunction for a given total angular momentum $\mathcal{L}$ may be expanded in coupled spherical harmonics:

$$\Psi(\vec{r}_1,\vec{r}_2,\vec{r}_3,\vec{r}_4,t) = \sum_{l_1,l_2}\sum_{L_{12},l_3}\sum_{L_{123},l_4}\frac{P^{\mathcal{L}}_{l_1l_2L_{12}l_3L_{123}l_4}(r_1,r_2,r_3,r_4,t)}{r_1r_2r_3r_4}$$
$$\times\sum_{M_{123},m_4}C^{L_{123}\ l_4\ \mathcal{L}}_{M_{123}m_40}\sum_{M_{12},m_3}C^{L_{12}\ l_3\ L_{123}}_{M_{12}m_3M_{123}}\sum_{m_1,m_2}C^{l_1\ l_2\ L_{12}}_{m_1m_2M_{12}}$$
$$\times Y_{l_1m_1}(\hat{r}_1)Y_{l_2m_2}(\hat{r}_2)Y_{l_3m_3}(\hat{r}_3)Y_{l_4m_4}(\hat{r}_4)\ . \quad (3)$$

We note as found in the Clebsch-Gordan coefficients that 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 $\mathcal{L}$. Upon substitution of Ψ into the time-dependent Schrodinger equation, we obtain the time-dependent close-coupled partial differential equations for each $\mathcal{L}$ symmetry:

$$\begin{aligned} \frac{i\partial P^{\mathcal{L}}_{l_1 l_2 L_{12} l_3 L_{123} l_4}(r_1,r_2,r_3,r_4,t)}{\partial t} &= T_{l_1 l_2 l_3 l_4}(r_1,r_2,r_3,r_4) \\ &\quad \times P^{\mathcal{L}}_{l_1 l_2 L_{12} l_3 L_{123} l_4}(r_1,r_2,r_3,r_4,t) \\ &+ \sum_{l_1',l_2',L_{12}',l_3',L_{123}',l_4'} \sum_{i<j=1}^{4} V^{\mathcal{L}}(r_i,r_j) \\ &\times P^{\mathcal{L}}_{l_1' l_2' L_{12}' l_3' L_{123}' l_4'}(r_1,r_2,r_3,r_4,t) \end{aligned} \tag{4}$$

where

$$T_{l_1 l_2 l_3 l_4}(r_1,r_2,r_3,r_4) = \sum_{i=1}^{4} \left(-\frac{1}{2}\frac{\partial^2}{\partial r_i^2} + \frac{l_i(l_i+1)}{2r_i^2} - \frac{Z}{r_i} \right) , \tag{5}$$

and the coupling operators are given in terms of 3j and 6j symbols by:

$$\begin{aligned} V^{\mathcal{L}}(r_1,r_2) &= (-1)^{l_1+l_1'+L_{12}} \delta_{l_3,l_3'} \delta_{l_4,l_4'} \delta_{L_{12},L_{12}'} \delta_{L_{123},L_{123}'} \\ &\times \sqrt{(2l_1+1)(2l_1'+1)(2l_2+1)(2l_2'+1)} \\ &\times \sum_{\lambda} \frac{(r_1,r_2)_<^{\lambda}}{(r_1,r_2)_>^{\lambda+1}} \begin{pmatrix} l_1 & \lambda & l_1' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_2 & \lambda & l_2' \\ 0 & 0 & 0 \end{pmatrix} \\ &\times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ l_2' & l_1' & \lambda \end{Bmatrix} , \end{aligned} \tag{6}$$

$$\begin{aligned} V^{\mathcal{L}}(r_1,r_3) &= (-1)^{l_2+L_{123}} \delta_{l_2,l_2'} \delta_{l_4,l_4'} \delta_{L_{123},L_{123}'} \\ &\times \sqrt{(2l_1+1)(2l_1'+1)(2l_3+1)(2l_3'+1)(2L_{12}+1)(2L_{12}'+1)} \\ &\times (-1)^{\lambda} \sum_{\lambda} \frac{(r_1,r_3)_<^{\lambda}}{(r_1,r_3)_>^{\lambda+1}} \begin{pmatrix} l_1 & \lambda & l_1' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_3 & \lambda & l_3' \\ 0 & 0 & 0 \end{pmatrix} \\ &\times \begin{Bmatrix} l_1 & l_2 & L_{123} \\ L_{12}' & \lambda & l_1' \end{Bmatrix} \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ l_3' & L_{12}' & \lambda \end{Bmatrix} , \end{aligned} \tag{7}$$

$$\begin{aligned} V^{\mathcal{L}}(r_1,r_4) &= (-1)^{l_2+l_3+L_{12}+L_{12}'+\mathcal{L}} \delta_{l_2,l_2'} \delta_{l_3,l_3'} \\ &\times \sqrt{(2l_1+1)(2l_1'+1)(2l_4+1)(2l_4'+1)} \\ &\times \sqrt{(2L_{12}+1)(2L_{12}'+1)(2L_{123}+1)(2L_{123}'+1)} \\ &\times \sum_{\lambda} \frac{(r_1,r_4)_<^{\lambda}}{(r_1,r_4)_>^{\lambda+1}} \begin{pmatrix} l_1 & \lambda & l_1' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_4 & \lambda & l_4' \\ 0 & 0 & 0 \end{pmatrix} \\ &\times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ L_{12}' & \lambda & l_1' \end{Bmatrix} \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ L_{123}' & \lambda & L_{12}' \end{Bmatrix} \\ &\times \begin{Bmatrix} L_{123} & l_4 & \mathcal{L} \\ l_4' & L_{123}' & \lambda \end{Bmatrix} , \end{aligned} \tag{8}$$

$$V^{\mathcal{L}}(r_2,r_3) = (-1)^{l_1+l_2+L_{12}+L_{123}+l_2'+L_{12}'}\delta_{l_1,l_1'}\delta_{l_4,l_4'}\delta_{L_{123},L_{123}'} \times \sqrt{(2l_2+1)(2l_2'+1)(2l_3+1)(2l_3'+1)(2L_{12}+1)(2L_{12}'+1)} \times (-1)^{\lambda}\sum_{\lambda}\frac{(r_2,r_3)_<^{\lambda}}{(r_2,r_3)_>^{\lambda+1}}\begin{pmatrix} l_2 & \lambda & l_2' \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} l_3 & \lambda & l_3' \\ 0 & 0 & 0 \end{pmatrix} \times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ \lambda & L_{12}' & l_2' \end{Bmatrix}\begin{Bmatrix} L_{12} & l_3 & L_{123} \\ l_3' & L_{12}' & \lambda \end{Bmatrix}, \quad (9)$$

$$V^{\mathcal{L}}(r_2,r_4) = (-1)^{l_1+l_2'+l_3+\mathcal{L}}\delta_{l_1,l_1'}\delta_{l_3,l_3'} \times \sqrt{(2l_2+1)(2l_2'+1)(2l_4+1)(2l_4'+1)} \times \sqrt{(2L_{12}+1)(2L_{12}'+1)(2L_{123}+1)(2L_{123}'+1)} \times \sum_{\lambda}\frac{(r_2,r_4)_<^{\lambda}}{(r_2,r_4)_>^{\lambda+1}}\begin{pmatrix} l_2 & \lambda & l_2' \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} l_4 & \lambda & l_4' \\ 0 & 0 & 0 \end{pmatrix} \times \begin{Bmatrix} l_1 & l_2 & L_{12} \\ \lambda & L_{12}' & l_2' \end{Bmatrix}\begin{Bmatrix} L_{12} & l_3 & L_{123} \\ L_{123}' & \lambda & L_{12}' \end{Bmatrix} \times \begin{Bmatrix} L_{123} & l_4 & \mathcal{L} \\ l_4' & L_{123}' & \lambda \end{Bmatrix}, \quad (10)$$

$$V^{\mathcal{L}}(r_3,r_4) = (-1)^{l_3+l_3'+L_{12}+L_{123}+L_{123}'+\mathcal{L}}\delta_{l_1,l_1'}\delta_{l_2,l_2'}\delta_{L_{12},L_{12'}} \times \sqrt{(2l_3+1)(2l_3'+1)(2l_4+1)(2l_4'+1)(2L_{123}+1)(2L_{123}'+1)} \times (-1)^{\lambda}\sum_{\lambda}\frac{(r_3,r_4)_<^{\lambda}}{(r_3,r_4)_>^{\lambda+1}}\begin{pmatrix} l_3 & \lambda & l_3' \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} l_4 & \lambda & l_4' \\ 0 & 0 & 0 \end{pmatrix} \times \begin{Bmatrix} L_{12} & l_3 & L_{123} \\ \lambda & L_{123}' & l_3' \end{Bmatrix}\begin{Bmatrix} L_{123} & l_4 & \mathcal{L} \\ l_4' & L_{123}' & \lambda \end{Bmatrix}. \quad (11)$$

We note that the indicies for V^{L} (r_i, r_j) have been skipped for the sake of brevity, that is $l_1l_2L_{12}l_3L_{123}l_4, l_1'l_2'L_{12}'l_3'L_{123}'l_4'$.

The initial condition for the solution of the time-dependent close-coupling equations of Eq.(4) in real time t is given by:

$$P^{\mathcal{L}}_{l_1l_2L_{12}l_3L_{123}l_4}(r_1,r_2,r_3,r_4,t=0) = \sum_{l}\bar{P}_{lll}(r_1,r_2,r_3,\tau\rightarrow\infty) \times G_{k_0l_0}(r_4)\delta_{l_1,l}\delta_{l_2,l}\delta_{l_3,l}\delta_{L_{123},0}\delta_{l_4,l_0}, \quad (12)$$

where the three-electron radial wavefunctions, $\bar{P}_{lll}(r_1,r_2,r_3,\tau)$, are obtained by solution of the TDCC-3D equations for the relaxation of a three-electron atom in imaginary time (τ).

Following the time propagation of the TDCC-4D equations, the triple ionization cross section is given by:

$$\sigma_{triple} = \frac{\pi}{2k_0^2} \int_0^\infty dk_1 \int_0^\infty dk_2 \int_0^\infty dk_3 \int_0^\infty dk_4 \sum_{\mathcal{L}} 2(2\mathcal{L}+1) \times \sum_{L_{12},L_{123}} \sum_{l_1,l_2,l_3,l_4} |P^{\mathcal{L}}_{l_1 l_2 L_{12} l_3 L_{123} l_4}(k_1 l_1, k_2 l_2, k_3 l_3, k_4 l_4)|^2 , \quad (13)$$

where $P^{\mathcal{L}}_{l_1 l_2 L_{12} l_3 L_{123} l_4}(k_1 l_1, k_2 l_2, k_3 l_3, k_4 l_4)$ is a four-electron momentum space wavefunction found by projection of the time evolved coordinate space wavefunctions onto fully anti-symmetric products of four box-normalized continuum orbitals.

3. RESULTS

The time-dependent close-coupling method was used to calculate the $\mathcal{L} = 0$ triple ionization cross section for the ground state of the Li atom. Calculations were carried on a 192×192×192×192 point lattice with a mesh spacing of $\delta r = 0.20$ ranging from $r = 0.00$ to $r = 38.4$ for all four sets of points.

Complete sets of bound, $P_{nl}(r)$, and continuum, $P_{kl}(r)$, single particle states are generated by matrix diagonalization of one-electron Hamiltonians. The completeness is exact for the number of points and mesh spacing of the radial grid.

Upon relaxation of the time-dependent close-coupling equations of Eq.(4) in imaginary time employing a $(192)^4$ point lattice with $\Delta r = 0.20$, the 7 coupled channels of Eq.(12) yield an energy of -186.58 eV for $\mathcal{L} = 0$ after 500 time steps.

After propagation of the time-dependent close-coupling equations of Eq.(4) in real time employing a $(192)^4$ point lattice with $\Delta r = 0.20$, the 13 coupled channels with $\mathcal{L} = 0$ yield a total triple ionization cross section of 6.84 $\times 10^{-9}$ Mb at an incident energy of 200 eV.

4. SUMMARY

In conclusion, we have formulated a time-dependent close-coupling (TDSE-4D) method to calculate electron-impact triple ionization cross sections for atoms. Our first application was the calculation of $\mathcal{L} = 0$ triple ionization cross sections for the ground state of the Li atom.

In the future we plan to extend the present four-electron time-dependent close-coupling (TDSE-4D) method to calculate $\mathcal{L} = 1, 2, 3$ triple ionization cross sections for the ground-state of the Li atom.

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