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Ab-initio studies of few photon ionisation of helium

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Abstract. In this work, an ab-initio method is used to study few-photon ionisation of helium with linearly and circularly polarized light.The photoionization cross sections are estimated from the ionization yields calculated by directly propagating the time-dependent Schrödinger equation (TDSE) and are compared with those obtained from the lowest-order (non-vanishing) perturbation theory (LOPT). The results show a clear deviation of the TDSE yields from those of the LOPT as well as the dominance of the linearly-polarized ionization yields over the circularly polarized ones, towards higher field intensities.

1. Introduction

We have recently witnessed the emergence of the free-electron lasers (FEL) capable to deliver coherent, intense radiation of short duration in the regime of soft- and hard- x-ray radiation sources [1,2]. In terms of the laser-atom interaction, the most prominent characteristic is that FEL radiation can interact *directly* with the inner-most electrons, in contrast to the nature of interaction of long-wavelength lasers (e.g. Ti:Sapphire at 800 nm). To this end, a direct method of solving the two-electron TDSE is used, which has been thoroughly applied successfully to a large variety of atomic systems over the years. It is worthwhile that the particular calculational method guarantees its versatility to include one-electron to few-electron systems, molecular systems and even quantum dot systems, more recently [3,4,5].

2. Ionization yields from TDSE and LOPT

Briefly, the calculational method involves the calculation of the helium's electronic structure by employing a configuration interaction (CI) approach of the helium eigenenergies as well as the corresponding dipole transition matrix elements. Next, these field-free calculated quantities are inserted into the TDSE, which is used to propagate the time-dependent wavefunction in the presence of the external electromagnetic field. Finally, photoionization cross sections are straightforwardly calculated using the calculated ionization yields [6].

TDSE ionization yields. The TDSE is expressed as,

$$\imath\partial_t\psi(\boldsymbol{r_1},\boldsymbol{r_2},t) = [\hat{H} + \hat{D}(t)]\psi(\boldsymbol{r_1},\boldsymbol{r_2},t) \tag{1}$$

where, $\psi(\boldsymbol{r_1},\boldsymbol{r_2};t)$ is the two-electron time-dependent wavefunction, $\hat{H}$ is helium's field-free hamiltonian and $\hat{D}(t)$ is the field-atom interaction operator given by $\hat{D}(t) = -\boldsymbol{A}(t)[\boldsymbol{p_1} + \boldsymbol{p_2}]$, where $\boldsymbol{p_1}$ and $\boldsymbol{p_2}$ are momenta of the two electrons. Within the dipole approximation the

Table 1: Comparison of one photon and three photon cross section values obtained from LOPT and TDSE

(a) One photon cross section values obtained from LOPT ($\sigma_1^{(pt)}$) and TDSE ($\sigma_1^{(td)}$) for a pulse intensity of 10^{13} W/cm^2. The yield $Y(t)$ is evaluated after the end of the pulse. $\Delta = 100 \times (|\sigma_1^{(td)} - \sigma_1^{(pt)}|)/\sigma_1^{(td)}$

ω(eV)	$Y(t)$ (10^{-3})	$\sigma_1^{(td)}$ (Mb)	$\sigma_1^{(pt)}$(Mb)	Δ(%)
42	4.045	2.203	2.186	0.77
46	3.761	1.644	1.636	0.49
50	2.367	1.222	1.208	1.15
54	1.439	0.866	0.834	3.69

(b) Three photon cross section values obtained from LOPT ($\sigma_3^{(pt)} = 6.5 \text{ x } 10^{-85} cm^6 s^2$) and TDSE ($\sigma_3^{(td)}$) for a 10eV and 14.8fs pulse. The yield $Y(t)$ is evaluated after the end of the pulse. $\Delta = 100 \times (|\sigma_3^{(td)} - \sigma_3^{(pt)}|)/\sigma_3^{(td)}$

Intensity (10^{13}W/cm^2)	Y(t) (10^{-5})	$\sigma_3^{(td)}$ (10^{-85}cm^{6}s^2)	Δ(%)
1	0.806	98	93.36
3	4.2276	19.2	66.15
8	25.858	6.195	4.92
10	50.052	6.140	5.86
30	1341.9	6.079	6.92
80	97654	23.39	72.21
100	83640	10.26	36.65

electromagnetic potential is $\boldsymbol{A}_q(t) = -\int_0^t dt' \boldsymbol{E}_q(t')$, with q to denote the field's polarization state. Expanding the wavefunction in the basis of field-free helium eigenstates, $\phi_{nLM_L}(\boldsymbol{r_1}, \boldsymbol{r_2})$, whose eigenenergy is given by E_{nLM_L}, we have $\psi(\boldsymbol{r_1}, \boldsymbol{r_2}; t) = \sum_{nLM_L} C_{nLM_L}(t)\phi_{nLM_L}(\boldsymbol{r_1}, \boldsymbol{r_2})$, where n, L, M_L are the principal, angular momentum and magnetic quantum numbers respectively. Substituting this into Eq. (1) we end up with a system of first-order differential equations for the expansion coefficients, $C_{nLM_L}(t)$ in terms of known quantities:

$$i\dot{C}_{nLM_L}(t) = C_{nLM_L}(t)E_{nLM_L} + \sum_{n'L'=L\pm 1} C_{n'L'M_L\pm q}(t)\hat{D}_{nLM_L;n'L'M_L\pm q}(t) \tag{2}$$

with $\hat{D}_{nLM_L;n'L'M_L\pm q}(t) = \langle \phi_{nLM_L}|\hat{D}_q(t)|\phi_{n'L'M_L\pm q}\rangle$. It is easily derived from the dipole selection rules that ionization proceeds with $\Delta M_L = q$. Therefore, linearly polarized (LP) light ($q = 0$) ionizes helium without changing the magnetic quantum number, while for circularly polarized (CP) light ($q = \pm 1$) changes by 1. The ionization yield can now be calculated from the following relation, for times greater than the pulse duration,

$$Y(t) = 1 - \sum_{nLM_L}^{E_{nLM_L} \leq E_{IP}} |C_{nLM_L}(t)|^2 \tag{3}$$

where E_{IP} represents the ionization potential. We then calculate the cross sections derived from TDSE by using, $\sigma_N^{(td)} = (\frac{\omega^N}{I_0^N \tau_N})Y(t), N = 1, 3$, where $\tau_1 = \frac{3}{8}\tau_p$ and $\tau_3 = \frac{231}{1024}\tau_p$ are the effective interaction times for one-and three-photon process, respectively. Also, I_0 is the peak intensity and τ_p is the pulse duration.

LOPT cross sections The single-photon and three-photon cross section according to LOPT in atomic units are given by, $\sigma_1^{(pt)} = \frac{4\pi^2}{c}\omega|D_{100;n10}|^2$ and $\sigma_3^{(pt)} = 2\pi(\frac{2\pi\omega}{c})^3 \sum_{L=1,3} |D_{100;nLL}^{(3)}|^2$ where $D_{100;nLL}^{(3)}$ is the effective 3-photon transition matrix element and ω is the photon energy [6].

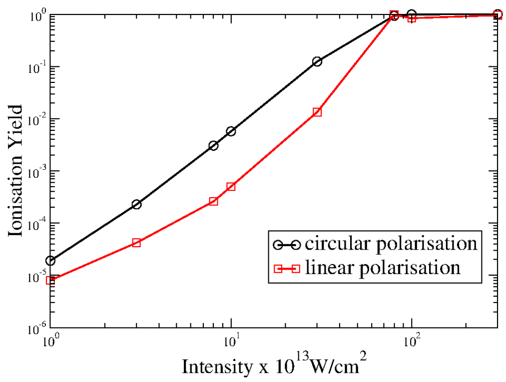

Figure 1: Ionization Yield vs peak intensity for circular and linear polarisations of a 10 eV and 14.8 fs pulse.

3. Results and Discussions

Comparing the photoionization cross sections estimated from the TDSE calculated yields with those obtained from LOPT, it can be seen that for low energies the cross sections are in good agreement while for higher energies, the corresponding values deviate, see table 1(a). This is because LOPT does not take into account excitation and/or ionization channels of He^+, which reside above 40.8 eV from its ground state. Also, resonant ionization through the 1s2s He state results to differences between the TDSE and LOPT approaches even for low intensities, along with expected differences for high intensities; the latter can be seen in table 1(b). The ionization yields for circular and linear polarization are compared with each other in the case of the three-photon ionization Fig. (1). For low intensities, circular polarization ion yields are higher than that of linear polarization. This is due to the strongest two-electron transition matrix element of the circularly polarized light relative to the linearly polarized light [7]. Nevertheless, towards higher intensities (where higher-photon absorption cross sections should be calculated) the situation should be reversed due to the presence of overwhelming number of ionization channels for linearly polarized light; to establish this fact one needs to calculate and compare photoionization absorption cross section higher than three, a task that was not taken here. These observations are in agreement with similar calculations performed in Mg [8].

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