Non-linear photointeractions under FEL intense short-wavelength radiation: case study

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Abstract. We present a theoretical study of a two-photon single-colour and two-colour excitation of ground-state krypton atoms to the final $3d_{5/2}^{-1}4d$ autoionising states.

1. Introduction

Experimental studies of multiphoton excitation or ionisation of matter with short-wavelength radiation have become feasible only recently: Free-electron laser (FEL) sources have opened ways to new experiments in which non-linear processes can be studied with radiation in the extreme ultraviolet and x-ray regime. In order to understand such experimental results on more complex systems, quantitative experimental and theoretical studies on simpler systems, such as noble gases, have been undertaken.

Depending on the intensity and pulse duration of the radiation, and the system under investigation, various theoretical approaches may need to be employed [1, 2, 3, 4]. If the intensity is sufficiently low, so that the ponderomotive energy is much smaller than the photon energy, and the pulse duration, in terms of cycles, is sufficiently long, a description in terms of appropriate cross sections and rate equations may be sufficient. If several ionic species are involved, the relevant ionisation yields can be obtained through the solution of coupled differential (kinetic or rate) equations describing their evolution during the laser pulse [3, 4]. Generalised multiphoton cross sections [1, 2], which enter these differential equations as parameters, can be calculated in the appropriate lowest non-vanishing order perturbation theory. The necessary accuracy in the calculation of such cross sections is dictated by the type and accuracy of the experimental data under consideration. In certain cases that may involve more detailed data, the use of density matrix techniques, within the space of the necessary atomic states, may be in order.

In this paper, we present a brief outline of the theoretical treatment of two-photon, single-colour and two-colour resonant excitation of krypton atoms from the ground state to the $3d_{5/2}^{-1}4d$ autoionising states with the total angular momentum $J = 0$ and $J = 2$. The motivation for this work came from ongoing experiments at the FEL facility FLASH in Hamburg [5]. Our aim here is to set up a simple theoretical framework for the experiment, providing an estimate of the expected ion yields and their dependence on the laser parameters, which can serve the purpose of optimisation of the experimental procedure.

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2. Two-photon excitation of the krypton atom

We describe the atom with a 5-level atom model within the density matrix formalism. The values of the laser parameters employed in the calculation are chosen so as to correspond to the experimental conditions.

We study two different ways of exciting the atom. In the first case, to be referred to as the two-colour case, we consider the krypton atoms exposed simultaneously to the FEL and Ti:sapphire (IR) laser pulses. The energy of the IR photons is fixed to 1.55 eV, and the energy of the FEL photons is chosen so that the sum of the photon energies $\hbar \omega_1 + \hbar \omega_2$ lies close to the energy difference between the ground state and the higher of the two $3d_5/2 4d J = 2$ final states.

In the second case, to be referred to as the single-colour case, we consider the excitation with two FEL photons, each of them having the energy of 46 eV. In both of the above cases, the two-photon excitation proceeds through the $3d_{5/2} 5p J = 1$ intermediate state.

The $3d^{−1}4d$ and $3d^{−1}5p$ states form two groups of states separated in energy by approximately 1.2 eV: the lower-lying group is characterised by the coupling of the $3d$ electrons to the total angular momentum $J_h = 5/2$ and the higher-lying group by $J_h = 3/2$. On the basis of known experiments [6], the widths of $3d^{−1}5p$ states are known to be about 100 meV, stemming predominantly from the Auger decay of the $3d$ hole, and this is also the value we assume for the $3d_{5/2}4d$ and $3d_{5/2}5p$ states. For the two-colour case, the bandwidth of the FEL operating at $\sim 90$ eV is about 0.9 eV, whereas the bandwidth of the IR laser is much smaller [5]. For 46-eV incident photons, the FEL bandwidth is further reduced to approximately one half of the above value. Although, in all generality, the present model should also include the $J_h = 3/2$ states, they have not been taken into account for the purposes of the present preliminary calculations. Their inclusion should not substantially change the results or affect the main ideas. Thus, we focus on the excitation to the final states with $J_h = 5/2$ only.

2.1. Theoretical model

The system under investigation is depicted in Fig. 1. We describe it with the Hamilton operator

$$ H = \sum_i E_i |i⟩⟨i| - \frac{\hbar}{2} \left( \Omega_{21}(t) e^{-i \omega_1 t} |2⟩⟨1| + \sum_{k \geq 3} \Omega_{k2}(t) e^{-i \omega_2 t} |k⟩⟨2| + \text{H.c.} \right), $$

where the first term represents the free atom, and the second the atom-field interaction. The energies of the atomic states $|i⟩$ are denoted by $E_i$. The frequencies $\omega_1$ and $\omega_2$ are associated with the first (FEL) and the second (IR or FEL) photon, respectively. The Rabi frequencies $\Omega_{12} = \Omega_{21}$, $\Omega_{2k} = \Omega_{k2}$, $k = 3, \ldots, 5$, are assumed real. In the semi-classical treatment, in which the field is treated classically, the Rabi frequency connecting the states $|i⟩$ and $|j⟩$ is defined as:

$$ \Omega_{ij}(t) = \hbar^{-1} 2F(t) D_{ij} = \hbar^{-1} 2F_0 \exp \left( -\frac{4t^2 \ln 2}{T^2} \right) D_{ij}, $$

where $2F_0 = 2E_1$ ($2F_0 = 2E_2$) is the electric field amplitude associated with the first (second) radiation field, $T = \tau_1$ ($T = \tau_2$) the duration of the first (second) pulse, and $D_{ij}$ the electric dipole matrix element. Note that the intensity of the radiation is connected to the field strength through the equality $I(t) = 2\epsilon_0 c F^2(t)$ (in SI units, $c$ is the speed of light in vacuo, and $\epsilon_0$ is the vacuum permittivity).

Since under the conditions relevant to the experiment, the rotating-wave approximation (RWA) is valid, the time-evolution of the system is described by the following set of equations:
\[ \dot{\sigma}_{11} = -\frac{i}{2} (\Omega_{12}\sigma_{12} - \Omega_{12}\sigma_{12}^*), \]  

\[ \dot{\sigma}_{22} = -\Gamma_2/\hbar \sigma_{22} - \frac{i}{2} (-\Omega_{12}\sigma_{12} + \Omega_{12}\sigma_{22}^* + \Omega_{23}\sigma_{23} - \Omega_{23}\sigma_{23}^*), \]

\[ \dot{\sigma}_{kk} = -\Gamma_k/\hbar \sigma_{kk} - \frac{i}{2} (-\Omega_{2k}\sigma_{2k} + \Omega_{2k}\sigma_{2k}^*), \quad k = 3, \ldots, 5, \]

\[ \dot{\sigma}_{12} = \hbar^{-1} \left[ -\Gamma_2/2 - \gamma_1 - i(E_1 + S_1 - E_2 - S_2 + \hbar\omega_1) \right] \sigma_{12} \]

\[ \dot{\sigma}_{1k} = \hbar^{-1} \left[ -\Gamma_k/2 - i(E_1 + S_1 - E_k - S_k + \hbar\omega_1 + \hbar\omega_2) \right] \sigma_{1k} \quad k = 3, \ldots, 5, \]

\[ \dot{\sigma}_{23} = \hbar^{-1} \left[ -\Gamma_2/2 - \Gamma_3/2 - \gamma_2 - i(E_2 + S_2 - E_3 - S_3 + \hbar\omega_2) \right] \sigma_{23} \]

\[ \dot{\sigma}_{24} = \hbar^{-1} \left[ -\Gamma_2/2 - \Gamma_4/2 - \gamma_2 - i(E_2 + S_2 - E_4 - S_4 + \hbar\omega_2) \right] \sigma_{24} \]

\[ \dot{\sigma}_{25} = \hbar^{-1} \left[ -\Gamma_2/2 - \Gamma_5/2 - \gamma_2 - i(E_2 + S_2 - E_5 - S_5 + \hbar\omega_2) \right] \sigma_{25} \]

\[ \dot{\sigma}_{3k} = \hbar^{-1} \left[ -\Gamma_3/2 - \Gamma_k/2 - i(E_3 + S_3 - E_k - S_k) \right] \sigma_{3k} \quad k = 4, 5, \]

\[ \dot{\sigma}_{45} = \hbar^{-1} \left[ -\Gamma_4/2 - \Gamma_5/2 - i(E_4 + S_4 - E_5 - S_5) \right] \sigma_{45} \]

Figure 1. Two-photon excitation of the krypton atom.
The elements of the density matrix in the frame rotating with the frequency of the external fields are denoted by $\sigma_{ij}$, the Auger energy widths of the intermediate ($i = 2$) and final states ($i = 3, \ldots, 5$) by $\Gamma_i$, and the bandwidths (half-width at half-maximum) of the first and the second radiation field by $\gamma_1$ and $\gamma_2$. Strictly speaking, the widths $\gamma_1$ and $\gamma_2$ describe phase fluctuations of the driving fields. However, if no bound-bound transition is saturated, the present treatment is also valid in the case of amplitude fluctuations of the fields. The field-induced coupling between the levels of the model results in energy shifts of these levels. In addition, we also introduce shifts $S_i$ due to all other levels not included in the 5-level model. It turns out that the shifts of the levels are dominated by the ponderomotive shift. It is well known that the ponderomotive shift $U_p$ is inversely proportional to the square of the photon frequency. In the two-colour case, the ponderomotive shift due to the IR laser is thus much larger than the shift due to the FEL, and we may write $S_i(t) \approx \frac{e^2 I_2(t)}{(2m_e e_0 c \omega_2^*^2)}$, $i = 2, \ldots, 5$. For the single-colour case, we write $S_i(t) = \frac{e^2 I_1(t)}{(2m_e e_0 c \omega_1^2)}$. The shift of the initial state has been set to zero.

The parameter values used in the calculation are given in Table 1.

|   | $E_i$ [eV] | $D_{2i}$ [a.u.] | $D_{12}$ [a.u.] |
|---|-----------|-----------------|-----------------|
| 1 | 91.245    | 0.857           | -0.027          |
| 2 | 92.033    | 2.234           |                 |
| 3 | 92.049    |                 |                 |
| 4 | 92.111    |                 |                 |
| 5 |           |                 |                 |

2.2. Two-colour excitation

![Two-photon two-colour excitation](image)

Figure 2. Two-photon two-colour excitation. Left: the peak intensity of the IR laser is varied. Right: the peak intensity of the FEL is varied. Thin solid, dashed, and thick solid patterns correspond to the final states $|3\rangle$, $|4\rangle$, and $|5\rangle$, respectively. See text for details.

We have calculated the ion yields resulting from various $3d_{5/2}^{-1}4d$ ($J = 0, 2$) final states. The photon energy of the IR laser is fixed to $\hbar \omega_2 = 1.55$ eV ($\lambda_2 = 800$ nm). The energy of the FEL...
photons $\hbar \omega_1$ is chosen in such a way that the sum of the photon energies $\hbar \omega_1 + \hbar \omega_2$ is equal to the energy of level 4 shifted by $S_1$, i.e., $\hbar \omega_1 = (E_4 + S_4) - (E_1 + S_1) - \hbar \omega_2$, at a selected value of the peak intensity of the IR laser. The pulse durations are $\tau_1 = 30$ fs for the FEL and $\tau_2 = 120$ fs for the IR laser. The bandwidth (half-width at half-maximum) of the FEL is set to $\gamma_1 = 0.45$ eV, whereas the bandwidth of the IR laser has been neglected because it is much smaller than $\gamma_1$ and would not affect the results.

In Figure 2(a) we show the variation of the yields with the peak intensity of the IR laser $(I_{2,\text{max}} = 2\epsilon_0cE_2^2)$. The peak intensity of the FEL is $I_{1,\text{max}} = 2 \times 10^{13}$ W/cm$^2$. Red (dark grey), green (light grey), and black curves correspond to the energy of the first photon $\hbar \omega_1$ chosen to reach the (shifted) level 4 at the IR peak intensities $1 \times 10^{12}$ W/cm$^2$ ($\hbar \omega_1 = 90.559$ eV), $1 \times 10^{13}$ W/cm$^2$ ($\hbar \omega_1 = 91.097$ eV), and $1 \times 10^{14}$ W/cm$^2$ ($\hbar \omega_1 = 96.473$ eV). Especially interesting is the behaviour of the ion yields for intensities above $10^{12}$ W/cm$^2$: it should be pointed out that the shape of the yields is the consequence of a rather intricate interplay between the intensity dependence of the driving ($\Omega_{ij}$) and the induced shifts. In Figure 2(b) we show the dependence of the ion yields on the FEL peak intensity $(I_{1,\text{max}} = 2\epsilon_0cE_1^2)$. Red (dark grey), green (light grey), and black curves correspond to the IR peak intensity set to $2 \times 10^{13}$ W/cm$^2$ ($\hbar \omega_1 = 91.694$ eV), $1 \times 10^{14}$ W/cm$^2$ ($\hbar \omega_1 = 96.473$ eV), and $5 \times 10^{14}$ W/cm$^2$ ($\hbar \omega_1 = 120.37$ eV).

2.3. Single-colour excitation

As in the two-colour case, we have calculated the single-colour ion yields (Figure 3) resulting from the Auger decay of the final states. The photon energy of the FEL has been fixed to $\hbar \omega_1 = \hbar \omega_2 = 46.0$ eV, and the pulse duration set to $\tau_1 = \tau_2 = 30$ fs. The bandwidth of the FEL is set to $\gamma_1 = \gamma_2 = 0.25$ eV.

![Figure 3](image_url)

**Figure 3.** Two-photon single-colour excitation. Thin solid, dashed, and thick solid lines correspond ion yields from the final states $|3\rangle$, $|4\rangle$, and $|5\rangle$, respectively. The dashed line (top) shows a quadratic dependence of the yield on the peak intensity (i.e., $Y \propto I_{1,\text{max}}^2$) to guide the eye.

For low peak intensities of the FEL (below $10^{14}$ W/cm$^2$), the yields exhibit a quadratic behaviour, as can be seen by comparing the slopes of the calculated yields with the (quadratic) power dependence shown in Figure 3, top. At higher intensities, for $I_{1,\text{max}} \gtrsim 10^{14}$ W/cm$^2$, a deviation of the yields from the quadratic behaviour can be observed: although the field-induced shifts may be small for lower intensities, they are no longer negligible above $10^{14}$ W/cm$^2$. The
final $3d^{-1}4d$ states move away from the energy $E_1 + \hbar \omega_1 + \hbar \omega_2$, and the yields do not grow proportionally to $I_{1,\text{max}}^2$ any longer.

3. Conclusion
We have studied two-photon excitation of the ground-state krypton atoms exposed to: (i) two-colour FEL + IR laser pulses, and (ii) single-colour FEL pulses. The present calculations serve as a theoretical framework for the interpretation of the recent experimental data. We have used the density-matrix formalism to calculate population of the $3d^{-1}4d J = 0, 2$ final states and the ion yields resulting from the Auger decay. We have used realistic parameter values to approach the experimental situation. One of the surprises in the course of this work has been the crucial role of the ponderomotive shifts in the single-colour case, in which the photon energy of 46 eV is rather large. Although such shifts are still smaller than the photon energy, they are comparable to the laser bandwidth and they need to be taken into consideration in order to optimise the two-photon resonance condition at the peak of the pulse.

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