Two-color pump-probe dynamics
of transitions between doubly excited states of Helium

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Abstract

We discuss the dynamics of the two-photon resonant ionization of helium involving two autoionizing states in the presence of two-color laser fields. The first source is tuned around the transition from the ground state to the $2s2p \, ^1P^o$ autoionizing state, and the second couples the state $2s2p \, ^1P^o$ to the $2p^2 \, ^1S$ autoionizing state. The laser coupling between the doubly excited states is shown to lead to modifications of the Beutler-Fano profile and the appearance of an Autler-Townes doublet. This double resonance effect between autoionizing states can be observed at moderate laser intensities easily attainable by currently operated sources.
I. INTRODUCTION

The development of lasers of high-intensity and high-frequency made possible the experimental investigations of many atomic systems and revealed new phenomena. For example, the strong electromagnetic coupling of two autoionizing states and their coherent interaction, in some cases, could lead to partial stabilization and population trapping in the ground state. The interaction of atomic autoionizing states with an external electromagnetic field has been considered in a number of papers. An experimental and theoretical investigation of the effects of the strong electromagnetic coupling of two autoionizing states on the photoionization properties of Mg demonstrated the coherent interaction between the autoionizing states.

Many experiments involving autoionizing states, have been performed over the last years and the observed dynamics present very interesting features. In this study, we consider the case of helium atom in which the ground state is coupled to the 2s2p $^1P^0$ autoionizing state, through a laser with frequency $\omega_1$. A second laser, with frequency $\omega_2$, couples the 2s2p $^1P^0$ state to the 2p$^2$ $^1S$ autoionizing state. The latter has not been observed in any photoexcitation experiment and the results of the present study can be used for a new experimental investigation. A similar system involving double autoionization resonance, has been studied theoretically in detail, in connection with laser-induced transitions between triply excited hollow states. Here we present only the essential formulae of our approach, since the complete theoretical treatment has been published elsewhere. The atomic parameters of the states involved are calculated by an ab-initio approach. In Sec. 2 we give a short description of our theoretical methodology and the basic dynamic equations describing the problem. In Sec. 3 we solve the equations and present and analyze our results. Finally, we conclude in Sec. 4 with suggestions for prospective experimental investigations.

II. THEORETICAL APPROACH

We consider Helium atom subject to two linear polarized laser fields with frequencies $\omega_1$ and $\omega_2$, respectively. The relative phase between them is ignored and the total laser field has the form:

$$E(t) = E_1(t) \exp(i\omega_1 t) + E_2(t) \exp(i\omega_2 t) + \text{c.c.}$$  \hspace{1cm} (1)
The frequencies $\omega_1$ and $\omega_2$ are chosen so as to be tunable around the selected resonance transitions, namely $1s^2 1S \rightarrow 2s2p \, 1P^o$ and $2s2p \, 1P^o \rightarrow 2p^2 1S$. For the time-dependent field amplitudes $E_i(t)$, $i = 1, 2$ we choose a convenient form for the pulse envelope, namely $a \sin^2$, avoiding the long tails of a Gaussian which make the numerics more difficult, without significantly affecting the results. The explicit form is:

$$E_i(t) = \mathcal{E}_i^{(0)} \sin^2\left(\frac{\pi t}{\tau_i}\right), \quad \text{with } 0 \leq t \leq \tau_i,$$

(2)

with $\mathcal{E}_i^{(0)}$ the maximum field strength and $\tau_i/2$ is the Full Width at Half Maximum (FWHM). We assume that $\tau_i$ is few picoseconds and the simultaneous action of $E_i(t)$, $i = 1, 2$, i.e. $\tau_1 = \tau_2$.

The next step is to solve the time-dependent Schrödinger equation. In the following sections we use the notation $|g\rangle$ for the ground state $1s^2 1S$ of He, and $|a\rangle$, $|E_a\rangle$ and $|b\rangle$, $|E_b\rangle$ for the discrete and continua parts belonging to the $2s2p \, 1P^o$ and $2p^2 1S$ doubly excited states, respectively. Note that these autoionizing states are single channel Feshbach resonances.

The wave function for this standard model system shown in fig.1, can be expressed as:

$$|\Psi(t)\rangle = C_g(t) |g\rangle + C_a(t) |a\rangle + C_b(t) |b\rangle + \int dE_a C_{E_a}(t) |E_a\rangle + \int dE_b C_{E_b}(t) |E_b\rangle.$$

(3)

The Hamiltonian operator of the system is written as: $H = H_0 + V + V_d$, with $H_0 |\mu\rangle = E_{\mu} |\mu\rangle$, $\mu = a, b, g$, and $V$ being the configuration interaction coupling the discrete parts of the doubly excited states to the continua and $V_d = V_d(t)$ is the field-atom interaction. Projection of the individual states in the expansion of $|\Psi(t)\rangle$ leads to a set of coupled differential equations containing amplitudes for the discrete parts as well as for the continua.

The introduction of (i) the slowly varying amplitudes $c_i(t)$ which are defined by $c_i(t) = C_i(t)e^{i(E_i/h + \Delta\omega)t}$ and $\Delta\omega$ is the sum of the frequencies of the absorbed photons, (ii) the application of the rotating wave approximation (RWA), and (iii) the adiabatic elimination of the continua lead to the following set of equations for the discrete-state amplitudes:

$$i\hbar \frac{\partial}{\partial t} \mathbf{c}(t) = \mathbf{H}(t)\mathbf{c}(t),$$

(4)

where

$$\mathbf{H}(t) = \begin{bmatrix}
S_g - \frac{i}{2} \gamma_g & \tilde{\Omega}_{ga} & S_{gb} - \frac{i}{2} \gamma_{gb} \\
\tilde{\Omega}_{ag} & -\delta_1 - \frac{i}{2}(\Gamma_a + \gamma_a) & \tilde{\Omega}_{ab} \\
S_{bg} - \frac{i}{2} \gamma_{bg} & \tilde{\Omega}_{ba} & -\delta_1 - \delta_2 - \frac{i}{2}(\Gamma_b + \gamma_b)
\end{bmatrix}.$$

(5)
and \( c(t) = [c_g(t), c_a(t), c_b(t)]^T \). In equation (5) \( \delta_1 = \omega_1 - (E_a^{(0)} + S_a - E_g - S_g) \), \( \delta_2 = \omega_2 - (E_b^{(0)} + S_b - E_a^{(0)} - S_a) \) are the detunings, \( E_a^{(0)} \), \( E_b^{(0)} \) and \( \Gamma_a, \Gamma_b \) the resonance energy and width of the doubly excited states, \( S_g, S_a, S_b, S_{gb} \) and \( \gamma_g, \gamma_a, \gamma_b, \gamma_{gb} \) the laser induced shifts and widths. The generalized complex Rabi frequencies \( \tilde{\Omega}_{ga} \) and \( \tilde{\Omega}_{ab} \) are defined as:

\[
\tilde{\Omega}_{ga} = \Omega_{ga} \left( 1 - \frac{i}{q_a} \right) = \frac{1}{2} E_1(t) D_{ga}^{(z)} \left( 1 - \frac{i}{q_a} \right) \tag{6}
\]

and

\[
\tilde{\Omega}_{ab} = \Omega_{ab} \left( 1 - \frac{i}{q_{ab}} \right) = \frac{1}{2} E_2(t) D_{ab}^{(z)} \left( 1 - \frac{i}{q_{ab}} \right) \tag{7}
\]

with \( D^{(z)} \) being the dipole along the polarization direction \( z \), and \( q_a \) and \( q_{ab} \) the Fano line shape parameter for the transition \( |g\rangle \rightarrow |a\rangle \) and its generalization for the transition \( |a\rangle \rightarrow |b\rangle \). Obviously the Hamiltonian of equation (5) is Non-Hermitian. The coefficients \( c_i(t) \) are slowly varying in the sense that the transformation \( c_i(t) = C_i(t)e^{(E_i/h+\Delta\omega)t} \) has removed their rapid variation.

For the derivation of the above equations the laser induced continuum-continuum couplings have been neglected. It has been shown that the line shapes are not affected by

\[
h\omega_1 = 60.15 \text{ eV}
\]

\[
h\omega_2 = 1.94 \text{ eV}
\]
continuum-continuum transitions and the same is true and for the total photoionization rate. Also, preliminary calculations showed that we can ignore the laser-induced couplings between the discrete parts $|a\rangle$ and $|b\rangle$ of the resonance states and the non-resonant part of the continua $|E_b\rangle$ and $|E_a\rangle$ respectively, and the second order effect of the laser induced coupling of the ground state $|g\rangle$ to $|b\rangle$ via the non-resonant part of the continua $|E_a\rangle$. By solving the above equations the ionization yield into each channel and the total ionization probability can be calculated. The total ionization probability is:

$$P(t) = 1 - |c_g(t)|^2 - |c_a(t)|^2 - |c_b(t)|^2$$  \hspace{1cm} (8)$$

\textit{Atomic structure parameters.} We have calculated all of the parameters pertaining to the atomic levels coupled by the process described above, through an \textit{ab initio} approach. In order to take into account the electron correlation, very accurately, a large number of configurations were selected. The MCHF method has been used to perform the present calculations. The MCHF wave-function expansion for the ground state of He was over a set of 15 configuration states coupled to form a $^1S$ term. The radial wave functions for the different orbitals were obtained by the MCHF procedure, varying all the orbitals simultaneously. Minimization of the total energy yielded an energy of -2.9033 a.u. to be compared to the accurate value of -2.903724 a.u. from extensive variational calculations. The autoionizing states 2s2p $^1P^0$ and 2p$^2$ $^1S$ of He are calculated using a partition of the function space within the framework of the Feshbach formalism. Using appropriate $Q$ and $P$ projection operators, we can represent these resonances as quasibound states embedded in a continuum. The localized wavefunctions and of these states are also calculated by a MCHF approach. For 2s2p $^1P^0$ we used 27 configuration states and the energy obtained for this state is: -0.69256 a.u. For the 2p$^2$ $^1S$ state we used 30 two-electron configuration states and the energy obtained for this state is: -0.62218 a.u. The correlation effects are important for an accurate description of these states. This can be seen from their configuration expansion, which for the 2s2p $^1P^0$
is of the form:

\[ \psi(2s2p \, P^o) = 0.953(2s2p) - 0.291(2p3d) - 0.076(3s3p) + \ldots \]

while for the \(2p^2 \, 1S\) state we have:

\[ \psi(2p^2 \, 1S) = 0.787(2p^2) - 0.554(2s^2) + 0.173(3s^2) - 0.139(3p^2) + 0.179(3d^2) + \ldots \]

The autoionization widths of these states are calculated by the well known complex-coordinate method\(^1\). We choose the open channel component of the resonant wavefunctions to be:

\[ u(1s\ell \, 1L^\pi) = \hat{A} \left( \phi_{1s}(r_1) \sum_i c_i \chi_i(\rho_i^*) Y_{\ell m}(\Omega_2) \right) \]  

with \(\chi_i(\rho^*) = (\rho^*)^k e^{-a_i \rho^*}\). The radial function \(\phi_{1s}(r)\) was kept fixed to the hydrogen-like orbital of He\(^+\) whereas, in the variant of the complex-coordinate approximation followed here, for \(\chi_i(\rho^*)\) the radial coordinate takes the form \(\rho_i^* = r_i e^{-i\theta}\). The non-linear parameters \(a_i\) and the expansion coefficients \(c_i\) are subject to a variational optimization for the calculation of the complex energy eigenvalues pertaining to the autoionizing resonant states.

Our results for the energy position, including the energy shift, and the width, of the autoionizing states presented here, can be compared to other more elaborate calculations\(^2\). The wave-functions described above were also used for the calculation of the dipole moments for the various transitions involved. All the parameters that enter in the calculation are shown in table 1. We note here that the use of the complex-coordinate method provides a powerful tool for the calculation of the complex Rabi frequency \(\tilde{\Omega}_{ab}\) for a transition between autoionizing states.

### III. RESULTS AND DISCUSSION

The solution of the system of differential equations given by Eq. (4) can provide us the information about the temporal evolution of the system under consideration. We have chosen to study the response of the system under the simultaneous action of the electric field \(E_1\) with frequency \(\omega_1\) and a pulse duration \(\tau_1=5\) ps and of the electric field \(E_2\) with frequency \(\omega_2\) and of the same duration.

Figures 2 and 3 show the photoionization yield of He\(^+\) as a function of the detunings of the laser sources for a series of intensities and detunings. As it can be seen from Fig. 2 the line
FIG. 2: He$^+$ photoion yield as a function of the detuning $\delta_1$, associated with the transition between the ground state and the 2s2p$^1P^o$ doubly excited state. The laser coupling the 2s2p$^1P^o$ and 2p$^2^1S$ states is on resonance ($\delta_2=0$). We clearly see that the laser coupling induces an Autler-Townes doublet.

Shape changes significantly with the intensity of the laser coupling the 2s2p$^1P^o$ and 2p$^2^1S$ doubly excited states. At low intensities we have a line shape for the lowest $^1P^o$ Feshbach resonance of He which is a typical Beutler-Fano profile$^{13}$. As the intensity is increased a doublet appears due to the ac Stark splitting as a result of the laser induced oscillation between 2s2p$^1P^o$ and 2p$^2^1S$. This structure is known as an Autler-Townes doublet and the separation between the two peaks carries information about the dipole matrix element coupling the doubly excited states. In Fig. 3 the frequency $\omega_1$ is on resonance ($\delta_1=0$) while the laser frequency $\omega_2$ is varied ($\delta_2 \neq 0$). In this case the coupling of the doubly excited states reveals a window resonance on the photoionization cross section.

The number of the electrons emitted in the energy region of each autoionizing state is proportional to the ionization signal given by the formula:

$$S_i = \int_{-\infty}^{+\infty} |c_i(t)|^2 \Gamma_i dt$$

with $i = a, b$. The magnitude of $S_i$ depends on the time evolution of the coefficients $c_i$ and, in a more crucial way, on the values of the autoionizing widths of the resonant states.
FIG. 3: He$^+$ photoion yield as a function of the detuning $\delta_2$, associated with the $2s2p \, ^1P^o \rightarrow 2p^2 \, ^1S$ transition. The laser coupling the ground state with the $2s2p \, ^1P^o$ doubly excited state is on resonance ($\delta_1=0$). We clearly see that the appearance of a window resonance as the laser intensity increases.

have calculated $S_a$ and $S_b$ for various values of $I_2$ ranging from $10^8 W/cm^2$ to $2 \times 10^{11} W/cm^2$ and for ($\delta_1 \neq 0$, $\delta_2 = 0$) and our results are shown in fig. 4a and 4b.

IV. CONCLUSIONS

To our knowledge the autoionizing state $2p^2 \, ^1S$ of He has not been observed in any multiphoton process. There has not been any detection of this state by two-photon processes and the energy position of this resonance is measured only in scattering experiments. In this study we showed that, the coupling of this state via a laser field of moderate intensity to another autoionizing state, has significant and detectable results in the photoionization cross section of He. The prospective pump-probe two color experiments utilizing sources as, for example, the Deutsches Elektronen-Synchrotron Free Electron Laser (DESY FEL) provide an apparatus for the detection of the resonant coupling of autoionizing states as these described here$^{14}$. The laser intensities used in our study are attainable and proposed experiments with Free-Electron Laser (FEL) sources are readily available$^{15,16}$. 
FIG. 4: The ionization signal in the energy range of the autoionizing states (a) $2s2p \, ^1P_0$ and (b) $2p^2 \, ^1S$, for various values of the laser intensity $I_2$. The laser coupling the $2s2p \, ^1P_0$ and $2p^2 \, ^1S$ states is on resonance ($\delta_2=0$).

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