

Ionization and transient absorption control with a resonant attosecond clock

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Abstract. Metastable states are important actors in the ionisation of atoms and molecules. Sub-femtosecond extreme ultraviolet pulses can coherently populate several transiently bound states at once, thus starting the attosecond clocks which are required to monitor and control ultrafast electronic evolution above the ionisation threshold. Here we illustrate, from a theoretical point of view, the effects coherent superpositions of $^1P^o$ doubly excited states in the helium atom have on channel-resolved photoelectron spectra as well as on the transient absorption spectrum of the atom in the extreme ultraviolet region, when they are created by a single-attosecond pulse in the presence of a strong few-cycle near-infrared/visible pulse which acts as a probe. Interference fringes varying rapidly with the pump-probe time delay are visible in both photoelectron and transient absorption spectra. From such fringes, the wave packet itself can conceivably be reconstructed. Conversely, all observables are modulated by the characteristic beating periods of the wave packet, so that control of partial ionisation yields, branching ratios, and light absorption or amplification can be achieved.

1. Introduction

Modern attosecond techniques [1] enable us, in principle, to monitor and control the ionization processes in atoms [2–7] and molecules [8–10]. Since the dynamics triggered by sub-femtosecond pulses [11, 12] generally unfolds in a spectral region above the ionisation threshold of the system, metastable states are bound to play a major role in many coherent control protocols [5, 10, 13]. In this context, the doubly excited states of the helium atom are an ideal subject of investigation because they permit to assess in detail, from both an experimental and a theoretical point of view, the capabilities of attosecond pump-probe techniques. This is because, on the one side, the helium atom is sufficiently simple; with the help of modern computers and well established *ab-initio* methods, we can make virtually exact theoretical predictions on the outcome of the interaction between the atom and almost arbitrary external pulses. On the other side, helium is also sufficiently complex to give rise to much of the many-body phenomena that are observed for larger systems, most prominently: parent-ion polarization, Auger decay, and multi-channel inelastic scattering. Here we examine the role of the doubly excited states which converge to the $N=2$ excitation threshold of the He^+ parent ion in pump-probe experiments where a weak extreme ultraviolet (XUV) isolated attosecond pump pulse, with central energy close to 60 eV, is used in conjunction with a dressing visible (VIS, $\lambda = 730$ nm) or near-infrared (NIR, $\lambda = 800$ nm) Ti-Sapphire compressed probe pulse. We look at two different kind of observables, the photoelectron distribution in separate channels [4, 5], and the variation of the XUV pulse



spectrum as it is transmitted through the sample [3, 14, 15]. In absence of the dressing pulse, the photoelectron and the absorption spectra are in close correspondence to each other, through the optical theorem, as well as to the spectrum that is recorded in stationary conditions using, e.g., monochromatised synchrotron radiation. In the presence of the dressing pulse, however, transition amplitudes involving several VIS/NIR photons, and even non-perturbative processes (Rabi oscillations, tunnelling, recollision of slow photoelectrons), can take place. As a result, several quantum paths can contribute coherently to each final state in the continuum, leading to interferences that reveal the phase relation between different intermediate resonant states. Seen from a different point of view, when dressed by an external strong pulse, the atom is not a time-invariant system anymore. As a consequence, the dipolar response of the dressed atom to the XUV pulse can no longer be expected to be diagonal in frequency; i.e., a frequency component $\mathcal{E}_0(\omega)$ in the perturbing field can induce a coherent response $\delta\mathcal{E}(\omega')$ at a different frequency, which in turn can combine with a pre-existing component of the XUV pulse at the same frequency, $\mathcal{E}_0(\omega') + \delta\mathcal{E}(\omega')$, thus leading, again, to interference fringes or even to light amplification.

2. ab initio simulation

To compute the response of the helium atom in its ground state to external radiation pulses and reproduce with high accuracy either the fully-resolved photoelectron distribution in the single-ionisation region across the N=2 threshold or the transient absorption spectrum in the XUV region, we represent the atomic wave function on a *B*-spline close-coupling basis [16, 17]. The close-coupling basis comprises several partial-wave channels (PWC), defined as the antisymmetrised and angularly coupled product of a bound state of the He⁺ parent ion and of an electron state with well defined orbital angular momentum,

$$\phi_{\alpha E} = \hat{A} \Theta_{S\Sigma} \mathcal{Y}_{L\alpha\ell\alpha}^{LM}(\Omega_1, \Omega_2) R_{N_\alpha L_\alpha}(r_1) \frac{f_{\alpha E}(r_2)}{r_2}, \quad (1)$$

where α is an index that uniquely identifies the PWC, \hat{A} is the antisymmetrizer, $\Theta_{S\Sigma}$ is a two-electron spin function with total spin S and spin projection Σ , $\mathcal{Y}_{L\alpha\ell\alpha}^{LM}$ is a bipolar spherical harmonics with total angular momentum L and projection M , $R_{N_\alpha L_\alpha}$ is the radial part of the frozen He⁺ parent ion state with principal quantum number N_α and angular momentum L_α . Finally, $f_{\alpha E}$ is the radial function of the second electron, which is associated to the orbital angular momentum ℓ_α , but is otherwise unconstrained. We limit the close coupling expansion to the PWCs with $N_\alpha \leq 2$: $1sE_L$, $2sE_L$, $2pE_{L+1}$, and $2pE_{L-1}$ (if $L > 0$). Such truncated close-coupling expansion is not sufficient to fully represent the correlated two-electron single-ionization space of helium in the energy region of interest. We account for the contribution of the single- and double-ionization closed channels not included in the expansion with an additional *full-CI* pseudo-state localized channel (LC) comprising a large number of normalized two-electron functions. All radial functions are expressed in terms of *B*-splines [18, 19] in a quantisation box with a finite size R_{box} . A large box, $R_{\text{box}} = 1200$ a.u., is required to compute the photoelectron distribution in the N=2 channels. A smaller box, $R_{\text{box}} = 400$ a.u., is sufficient to compute the transient absorption spectrum. Starting from the ground state Ψ_g , the state of the atom is propagated under the action of the external fields by integrating the TDSE numerically [5],

$$i\partial_t \Psi(t) = \left[H_0 + \alpha \vec{A}(t) \cdot \vec{P} + V_{\text{abs}} \right] \Psi(t), \quad \lim_{t \rightarrow -\infty} e^{iE_g t} \Psi(t) = \Psi_g, \quad (2)$$

where H_0 is the field-free atomic Hamiltonian, $\vec{A}(t)$ is the total external vector potential, \vec{P} is the total canonical electronic linear momentum, α is the fine-structure constant and V_{abs} is a complex absorption potential that prevents unphysical reflections from the box boundary. To solve (2), we

use a second-order split-exponential propagator [5, 20]. At the end of the simulation, as soon as the external pulses are over, we extract the distribution of the photo-fragments by projecting the wave-packet on a complete set of scattering states of the field-free atomic hamiltonian [5, 21, 22],

$$P_{N_a L_a}^{2S+1L^\pi}(E) = \sum_{\Sigma M \ell_\alpha} |\langle \psi_{N_a L_a, E \ell_\alpha}^{\Gamma(-)} | \Psi(t_{\max}) \rangle|^2, \quad (3)$$

where Γ is a collective index for all good global quantum numbers, N_a and L_a indicate the principal quantum number and angular momentum of a given parent ion (e.g., $N_a L_a = 1s, 2s, 2p$), α indicates all the partial wave channels generated from that parent ion, and the $(-)$ sign means that the scattering states fulfil incoming boundary conditions [23]. The single-ionization scattering states of helium are obtained with the K -matrix method [24], an L^2 realization of configuration interaction in the continuum. For more details, see [16, 17, 21, 22, 24] and references therein. We compute the transient absorption spectrum of the weak attosecond XUV pulse associated to a pump-probe simulation with the formula [20]

$$\sigma_{\text{TAS}}(\omega; t_d) = \frac{4\pi}{\omega} \text{Im} \frac{\tilde{P}(\omega; t_d)}{\tilde{A}_{\text{XUV}}(\omega; t_d)} \quad (4)$$

where \tilde{P} and \tilde{A}_{XUV} are the Fourier Transforms of the total electronic canonical momentum and of the XUV vector potential amplitude, respectively. Both quantities depend parametrically on the time delay between the XUV and the VIS pulse. The latter defines the time origin for the laser-dressed atom.

3. Numerical results

To illustrate the capabilities of the photoelectron and light-transmission interferometric techniques to highlight the dynamics of doubly excited states in the presence of a dressing field, we conducted two separate series of pump-probe simulations. In a first series, we employ an XUV pulse with central energy $\omega_{\text{XUV}} = 60.7$ eV, intensity $I_{\text{XUV}} = 1$ TW/cm², and duration $\text{fwhm}_{\text{XUV}} = 270$ as, in conjunction with an IR pulse with wavelength $\lambda_{\text{IR}} = 800$ nm, intensity $I_{\text{IR}} = 1$ TW/cm² and duration $\text{fwhm}_{\text{IR}} = 3.9$ fs. Please notice that the comparatively high intensity of the XUV pulse serves the only purpose of providing a better contrast in the numerical outcome of the simulation. Indeed, due to the high frequency of the XUV pulse, an intensity of 1 TW/cm² leads to negligible two-photon transition amplitudes; any ionisation process examined in the present work is linear with respect to the XUV intensity. As anticipated above, at the end of each simulation we can extract the asymptotic photoelectron distribution in any channel (state of the parent ion), direction or global symmetry, as a function of the photoelectron energy. In figure 1 we show the $^1P^o$ contribution to the photoelectron spectrum for the $2s$ and the $2p$ channels as a function of the photoelectron energy and of the time delay. In both channels, for time delays such that the two pulses overlap, the spectrum is dominated by the streaking signal [25]. For larger time delays, one can observe several overlapping hyperbolic interference fringes, which are due to the interference between the direct ionisation amplitude in the $N=2$ channel, generated by the XUV pulse alone, with the resonant ionisation amplitude involving first the absorption of an XUV photon by a doubly excited state, followed by the absorption of two or four IR photons. These fringes are analogous to those reported by Mauritsson *et al.* [26] for the ionisation of the helium atom at a much lower energy, in correspondence of the $N=1$ threshold. The contributions of different doubly excited states can be easily disentangled with a Fourier transform of the spectra with respect to the time delay. A second important component of the photoelectron spectrum is the beating between pairs of doubly excited states. If observed experimentally, such beatings would indicate, first, that two corresponding doubly excited states

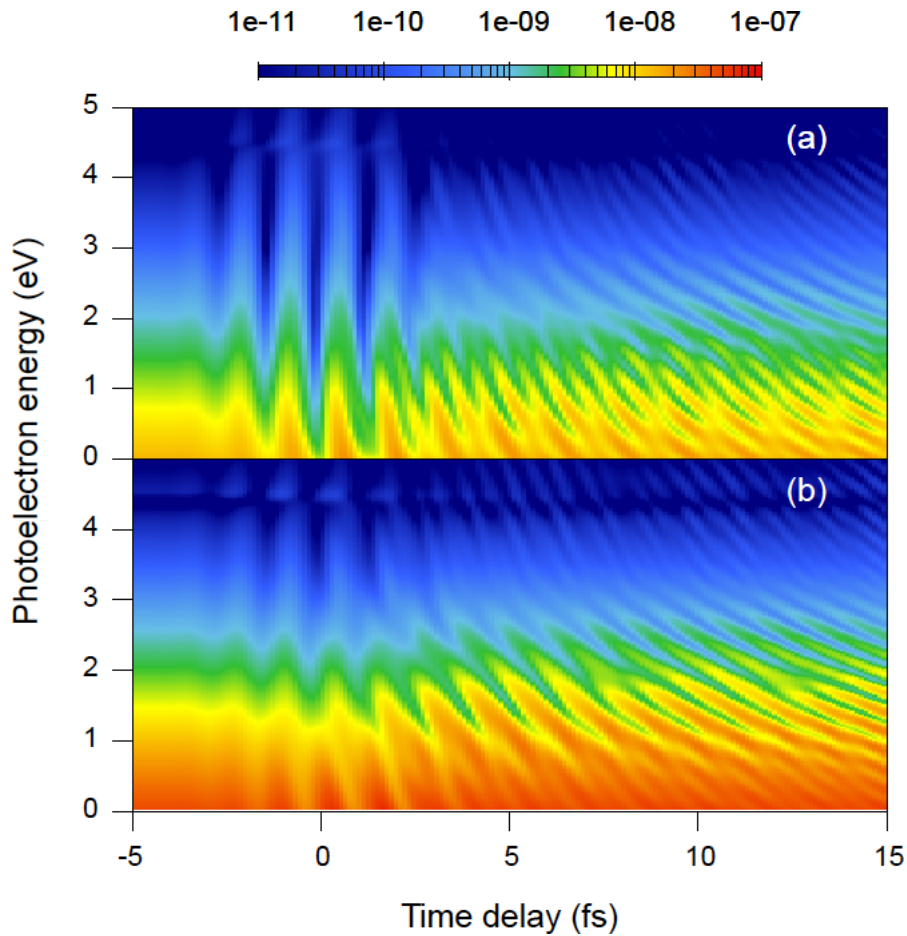


Figure 1. Photoelectron spectrum for the $1P^o$ component of the wave packet in the $2s$ (a) and $2p$ (b) channels, as a function of the time delay.

are coherently populated and, second, that such coherence can be exploited to control the yield and the branching ratios of the ionisation process. In the present case, for example, the partial ionisation yields in the $2s$ and $2p$ channels are both strongly modulated by the beating between the first two doubly excited states in the $sp_{2,n}^+$ series. Our original calculations [5], conducted with a minimal close-coupling basis, indicated that the modulations in the two channels are significantly out of phase. As a consequence, the $sp_{2,2}^+ - sp_{2,3}^+$ beating can be used to control the $2s/2p$ ionisation branching ratio using the pump-probe time-delay as a handle. The present calculation, which is conducted in a larger box and which comprises a fully correlated basis, confirms qualitatively our original prediction and actually indicates that the two oscillations are almost in phase opposition. As a consequence, the $sp_{2,2}^+ - sp_{2,3}^+$ beating remains clearly visible in the $2s/2p$ branching ratio, while it is significantly less visible in the total $N = 2$ ionisation yield. As it turns out, the total ionisation probability to the $N = 2$ channels is still strongly modulated, but the observed oscillations have a much larger period than the one of the $sp_{2,2}^+ - sp_{2,3}^+$.

In a second series of simulations, we compute the transient absorption spectrum of helium in the presence of a visible dressing pulse with duration (fwhm) of 7 fs, carrier wavelength $\lambda = 730$ nm, and peak intensity $I_{\text{VIS}} = 3.5$ TW/cm², as a function of the time delay with respect to a weak 60 eV sub-femtosecond XUV pulse (fwhm=500 as) [20]. These parameters are chosen to reproduce the experimental conditions reported in a recent measurement by Thomas Pfeifer and co-workers [27]. The transient absorption spectrum is shown in figure 2a. For sufficiently

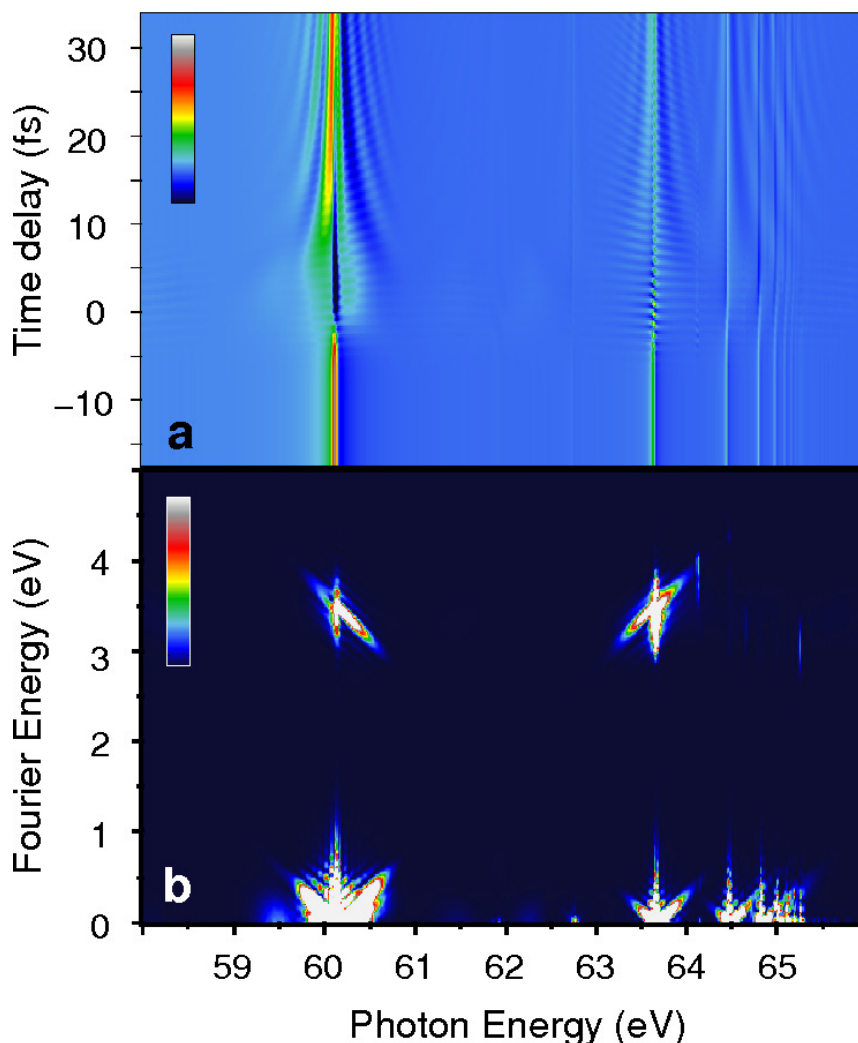


Figure 2. Attosecond transient absorption spectrum of helium in the vicinity of the doubly excited states converging to the $N=2$ threshold, for a dressing pulse with intensity $I = 3.5 \text{ TW/cm}^2$, as a function of: (a) the time delay; (b) the Fourier energy (b), after the Fourier Transform with respect to the time delay is taken. Negative time delays correspond to the dressing pulse coming first. See text for details.

large negative values of the time delay, $t_d \leq 5 \text{ fs}$, when the dressing visible pulse impinges on the atom before the XUV pulse and the two pulses do not overlap, the transient absorption spectrum coincides with the familiar absorption spectrum of the field-free atom. In particular, one can recognise the Fano profiles of the $sp_{2,n}^+$ bright series which accumulate at the $N=2$ threshold (photon energy = 65.4 eV). This is because the visible dressing pulse is too weak to alter the state of the target, due to the large energy required to excite the helium atom. When the two pulses overlap, the lineshapes in the spectrum are severely altered. The most characteristic feature of this time-delay region is the Autler-Townes splitting of the $sp_{2,n}^+$ state, which is the energy counterpart of a Rabi oscillation between this doubly excited states and the $2p^2 \ ^1S$ resonance, whose energy is located midway between those of the $sp_{2,2}^+$ and of the $sp_{2,3}^+$ states. In fact, for larger values of the dressing laser intensity, the $sp_{2,2}^+$ Fano profile splits into three peaks which are indicative of a multi-level multi-photon Rabi oscillation involving the three doubly excited

states. When the two pulses do not overlap anymore, the resonant profiles in the spectrum slowly converge again to the field-free limit. In this latter time-delay region, we can recognise two major kind of interference fringes close to the first two $sp_{2,n}^+$ doubly excited states. The fringes of the first kind converge exponentially towards the resonance positions and arise whenever the visible dressing pulse impinges on the residual localised component of an autoionizing state with a lifetime that is large if compared to the duration of the pulse. As a result of the interaction with the laser, both the phase and the absolute value of the coefficient of the localised component are suddenly altered. It is such rapid change in the population and phase of a state which would otherwise decay exponentially that causes the appearance, in the energy domain, of the observed fringes. This phenomenon is entirely similar to the build-up of a Fano profile described in [22, 28]. The fringes of the second kind are those which vary rapidly with the time delay and which bisect the resonance features. The phenomenon at the basis of this second kind of fringes is the interference between the direct ionisation amplitude induced by the XUV pulse and the indirect one due to the absorption or stimulated emission of two additional photons from the visible dressing pulse. When the pulses are separated, the laser-assisted amplitude receives contributions from the resonant component only. The fringes are thus branches of hyperbola similar to those observed in the photoelectron spectrum. The Fourier transform of the transient absorption spectrum variation $\Delta\sigma_{\text{TAS}}(\omega, t_d) = \sigma_{\text{TAS}}(\omega; t_d) - \sigma_{\text{TAS}}(\omega, -\infty)$ with respect to the time delay is reported in figure 2b. The new angular frequency variable associated to the time delay we call Fourier energy [20, 27]. This representation highlights the slowly varying features as bright spots close to zero Fourier Energy, in correspondence of the position of all the terms in the $sp_{2,n}^+$ series, and the rapidly varying features as bright spots at a Fourier energy that is twice the energy of a VIS photon, in correspondence to the first two doubly excited states.

By keeping the pump-probe time delay fixed and varying the intensity of the dressing pulse, Thomas Pfeifer and co-workers demonstrated experimentally [27] that the asymmetry of the resonant Fano profiles can be inverted. In a recent work [29], the authors showed that such inversion can be interpreted in terms of a mixing between the absorption and dispersion resonant response of the system due to the ac-Stark shift imparted by the dressing field to the localised component of the transiently bound state. As long as other mechanisms that dissipate the population of the autoionizing state do not dominate, such interpretation indicates that the inversion of the Fano profile should be periodic with respect to the laser intensity. In fact, in our simulations several inversions affecting all the terms of the $sp_{2,n}^+$ series are observed in the intensity range up to 10 TW/cm².

4. Conclusions

From the amplitude and phase of the rapidly varying fringes in either the photoelectron spectra or the transient absorption spectrum, one can conceivably reconstruct the coherent metastable wave packet generated by the XUV pulse. Acting on the intensity or duration of an additional dressing field, or on the time delay with respect to the XUV pulse, one can exploit perturbative interference effects, dephasing due to ac-Stark shifts of the doubly excited states, and the transfer of population inherent to Rabi oscillations to modify the state of the wave packet for subsequent manipulation. Conversely, one can also use these handles to control the asymptotic distribution of both photoelectrons and transmitted photons, i.e., to modify the branching ratios between the ionisation probabilities to the N=2 and to the N=1 channel, or between the two N=2 channels, on the photoelectron side, or to damp/amplify specific frequency components in the transmitted XUV spectrum. Finally, multi photon transitions between continuum states promoted by the dressing pulse are a major aspect of the processes taking place in the two cases examined here. Due to parity reasons, this means that the observed interferences depend parametrically on the phases and amplitudes of only those frequency components of the XUV pulse which differ from each other by twice the frequency of the dressing field. As a consequence, to observe the

phenomenology described here, it is not strictly necessary to employ an isolated attosecond pulse; a train of attosecond pulses provides an equivalent mean to study the response of the dressed atom (provided that the central frequency of the harmonics, i.e., the frequency of the dressing field, can be changed continuously so to cover the whole energy spectrum). This is all the more true in the case of the three doubly excited states that we examined more in detail, the $sp_{2,2}^+$, the $2p^2$ and the $sp_{2,3}^+$ states, which are separated from each other by an energy approximately equal to that of a quantum in a compressed Ti-Sapphire pulse. All these considerations suggest that an extensive study of the radiative transitions between these doubly excited states can be conducted with the RABITT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) technique [30].

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