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Strong field quantum control by selective population of dressed states

M Wollenhaupt, A Präkelt, C Sarpe-Tudoran, D Liese and T Baumert

University of Kassel, Institute of Physics, Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), Heinrich-Plett-Straße 40, D-34132 Kassel, Germany

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Abstract

We study the dynamics of potassium atoms in intense laser fields using femtosecond phase-locked pulse pairs in order to extract physical mechanisms of strong field quantum control. The structure of the Autler–Townes (AT) doublet in the photoelectron spectra is measured to analyse transient processes. The analysis shows that the physical mechanism is based on the selective population of dressed states (SPODS). Experimental results of closed loop optimization of SPODS are presented in addition. Applications to decoherence measurements with implications for quantum information are also proposed.

Keywords: coherent control, photoelectron spectroscopy, Autler–Townes, closed loop, multiphoton ionization, intense femtosecond fields, SPODS

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The ability to exert microscopic control on quantum systems opens up undreamed-of prospects for applications in physics, chemistry and biology. Quantum control deals with the design of suitably shaped light fields to selectively steer quantum systems from an initial state to a desired final state with high efficiency. Many quantum control strategies have been proposed and demonstrated experimentally which are summarized in recent reviews [1–5].

The design of specific pulse shapes in open loop schemes relies on the knowledge of the potential surfaces and the physical mechanisms. The analysis of the physical mechanisms is practicable in the weak field regime but rather the exception to the rule in strong laser fields. Examples of strong field open loop mechanisms are the RAP (rapid adiabatic passage) [6, 7], the STIRAP (stimulated Raman adiabatic passage) [8] and the modification of the potential surfaces due to the AC-Stark effect [9–11]. An alternative approach based on the use of the combination of pulse shaping techniques [12] with adaptive feedback learning loops (closed loop) was suggested [13] for the case when the underlying potential surfaces are unknown. Implementations of this technique demonstrate the optimization of almost any conceivable physical quantity [14–26]. However, it is not clear whether this methodology is suitable to extract the underlying physical mechanism from the electrical fields obtained during the optimization process. In order to shine some light into the 'black box' of the optimal control experiments our approach is based on the investigation of quantum control on a simple well defined model system excited by well characterized intense laser pulses.

In this contribution, we present a novel strong field multi-photon quantum control scenario based on selective population of dressed states (SPODS). We use intense phase-locked pulse pairs to coherently excite the K 4p ← 4s transition. The dynamics is probed via simultaneous two-photon ionization from the 4p state. This technique allows us to exert phase control with interferometric accuracy and to extract the physical mechanisms of strong field quantum control since the photoelectron spectra contain information on the transient dynamic. Since photoelectron spectra map the population of the dressed states, we are able to manipulate the dressed state population with the relative phase between the pulses. Strong field control on this system using more complex pulses shapes, such as sinusoidal phase modulation in the frequency domain, is reported in [27]. In addition, we investigate if the efficiency of the mechanism can be further optimized by closed loop experiments. This procedure might be useful to devise more efficient strong field control schemes. Eventually, understanding and purposive exploitation of the mechanisms will allow us to go beyond the limitations of

adaptive control which arise from experimental limitations such as the spectral bandwidth or intensity of the laser source and the complex structure of the multi-dimensional search space. The latter issue was theoretically investigated [28] and experimentally studied on a model system using parametrized pulse shapes [27].

This paper is organized as follows. First, the idea of the experiment is discussed in section 2, then the experimental set-up is briefly described in section 3. The two-pulse sequence experiment presented in section 4 is analysed in some detail. The physical mechanism of strong field control based on SPODS is discussed. In addition to the open loop control of the dressed state population, we report on an experiment to study strong field closed loop optimization of SPODS in section 5.

2. Principle of the experiment

Figure 1 shows the experimental set-up and the excitation scheme used in this experiment. Unlike conventional pump–probe scenarios in which the first pump pulse initiates the dynamics which is probed by the second pulse, in our experiment the same pulse is used to drive the neutral dynamics and to trigger the ionization. Since the neutral-to-ionic transitions are much weaker than the K (4p ← 4s) transition, the photo-ionization is described using perturbation theory. The amplitudes $c(\omega_e)$ for the ionization from the 4p excited state read [29]

$$c(\omega_e) = \int_{-\infty}^{\infty} c_{4p}(t) E^2(t) e^{i(\omega_k - \omega_{4p})t}\, dt \qquad (1)$$

where $c_{4p}(t)$ describes the time dependent excited state amplitude, $\hbar\omega_k$ the energy of the free electrons and $E(t)$ the electrical field. From equation (1) it is seen that the amplitudes $c(\omega_e)$ are the Fourier transform of the excited state amplitude $c_{4p}(t)$ windowed by the square of the electrical field $E^2(t)$. As a consequence, the quantum mechanical phase information of the excited state amplitudes is preserved to some extent in the photoelectron spectrum.

3. Experimental details

The experiments were carried out in a high vacuum chamber where a beam of atomic potassium K (4s) intersects perpendicularly with the femtosecond laser pulses leading to photo-ionization (cf figure 1). The released photoelectrons are detected employing a time-of-flight electron spectrometer with an energy resolution of 25 meV at a kinetic energy of 1 eV. The 785 nm, 30 fs FWHM laser pulses—provided by a 1 kHz Ti:sapphire multipass amplifier—are split into two pulses with the same intensity and identical phase relation of the carrier oscillation to the envelope using a Mach–Zehnder interferometer. The pulses are focused into the atomic beam at intensities of 0.5×10^{12} W cm^{-2}. The time delay between the identical laser pulses was set by the variation of the length of one arm of the interferometer and measured interferometrically with the spectral interference simultaneously recorded by an optical spectrometer. In our experiment, reproducible delays with an accuracy of around 0.2 fs were obtained. In addition, a home built phase modulator [30] was inserted in one arm of the interferometer in order to generate phase modulated pulses for the experiments on the closed loop optimization of SPODS.

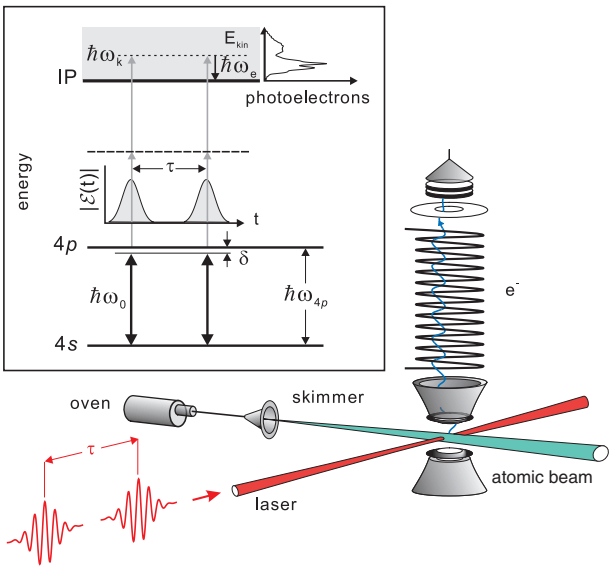

Figure 1. In the experiment femtosecond laser pulses intersect an atomic beam to produce photoelectrons which are measured with energy resolution using a magnetic bottle spectrometer. The inset shows the energy level diagram for excitation of K atoms. A sequence of two time delayed 30 fs FWHM laser pulses with a field envelope $|\mathcal{E}(t)|$ and a carrier frequency ω_0 (corresponding to 785 nm) detuned from the resonance frequency ω_{4p} (768 nm) creates a coherent superposition of the lower 4s and the upper 4p states of K atoms (black arrows). Photoelectron spectra from simultaneous two-photon ionization (grey arrows) of the 4p state to the continuum (ω_k) are measured as a function of the delay τ between the pulses.

4. Pulse sequence experiment

We start with the discussion of a double-pulse experiment. Using quasi-cw optical pulse sequences [31–33] demonstrated the role of the relative phase in these experiments. The potential of phase locked pulse sequences as a novel spectroscopic tool was demonstrated on single-photon molecular transitions [34] and discussed in [35]. Later, shaped weak field pulses were employed in order to control multi-photon transitions in atoms [36–39], molecules [40] and solid state systems [41, 42]. It was pointed out that subtle differences due to the relative optical phase either produced by optical phase shifts or time delay [43, 44] can control the outcome of the experiment. Although the experiments were mostly interpreted in terms of quantum interference, weak field experimental results on coherent systems can be interpreted in terms of (higher order) spectral interference. In our experiment, we study phase effects in strong fields beyond spectral interference.

4.1. Experimental results

Figure 2 shows the measured photoelectron spectra for different delay times 92 fs $+ n/4 \cdot T_B$ ($n = 0$–3) between the two pulses within one Bohr period $T_B = 2\pi/\omega_{4p}$. Since the structure of the photoelectrons is periodically reproduced every Bohr period (2.6 fs) [45] we show the limiting cases at $0T_B$, $0.25T_B$, $0.5T_B$ and $0.75T_B$. In all cases, the photoelectron spectrum is split into the Autler–Townes (AT) doublet [46] due to the high intensity of the laser pulses. Although

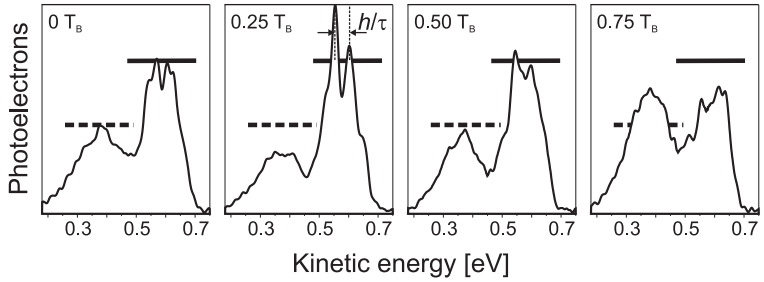

Figure 2. Photoelectron spectra for different delay times 92 fs + $(n/4)T_B$ between the two pulses within one Bohr period $T_B = 2\pi/\omega_{4p}$. The lines indicate the reference intensity for the slow (dashed) and the fast (bold) photoelectrons at $0T_B$ and $0.5T_B$. At $0.25T_B$ the intensity of the fast photoelectrons is enhanced at the expense of the slow photoelectrons. The reverse is observed at $0.75T_B$. The fringes due to the interference of the free electron wavepackets with a spacing of h/τ are most pronounced at $0.25T_B$.

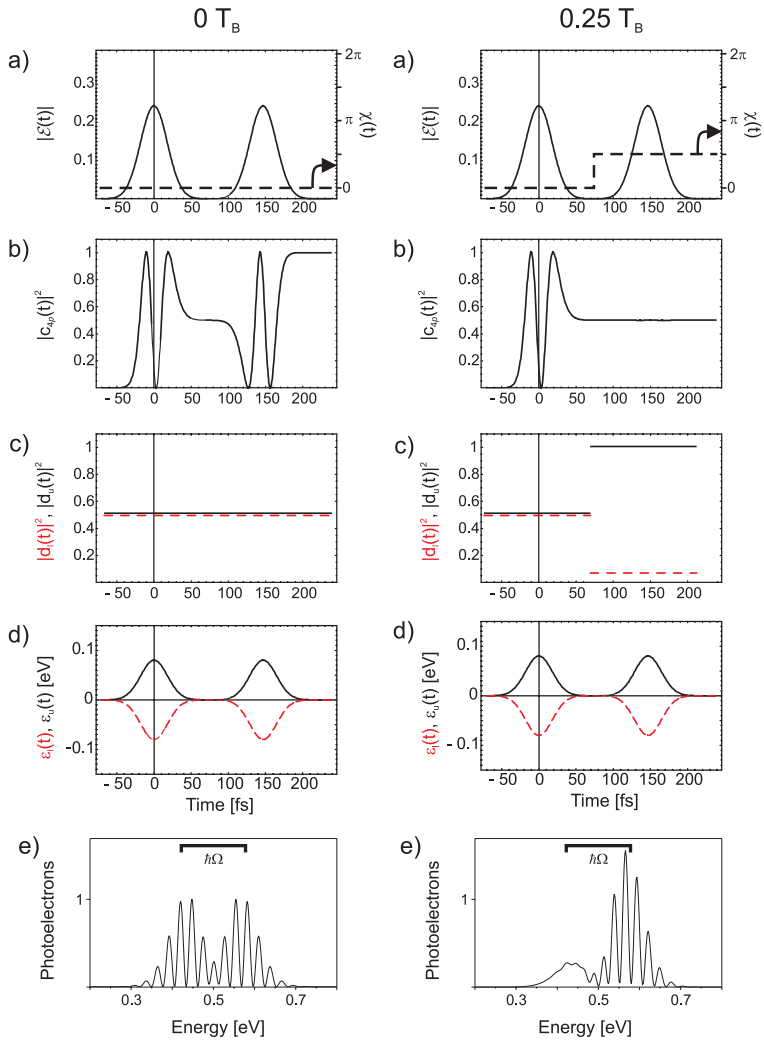

Figure 3. Calculations for resonant excitation and ionization of ground state atoms with a pulse sequence of FWHM = 30 fs pulse duration and $\theta = \frac{7\pi}{2}$ pulse area for $0T_B$ (left) and $0.25T_B$ (right). (a) The envelope of the laser field $|\mathcal{E}(t)|$ and the temporal optical phase $\chi(t)$, (b) the time evolution of the upper bare state population $|c_{4p}(t)|^2$, (c) the population of the upper $|d_u(t)|^2$ (bold) and lower $|d_l(t)|^2$ (dashed) dressed states and (d) the energies of the upper $\varepsilon_u(t)$ (bold) and lower $\varepsilon_l(t)$ (dashed) dressed states. The photoelectron spectra with an Autler–Townes splitting of $\hbar|\Omega|$ is depicted in (e). Comparison of (c) and (e) shows that the photoelectron spectra map the SPODS during the second laser pulse.

the laser pulses are red detuned from the atomic resonance, the fast photoelectrons are more intense than the slow ones. Independent measurements showed that the relative intensity is very sensitive to residual linear chirps of the laser pulse. In this measurement, the intensity of both AT components is controlled by the delay of the second laser pulse. The absolute intensity at $0T_B$ is taken as a reference indicated by the dashed (bold) lines in figure 2 for the slow (fast) photoelectrons.

Besides the higher intensity, the fast photoelectrons show interference fringes. At a delay time of $0.25T_B$ later, the fast photoelectrons are enhanced and show more pronounced interference fringes, whereas the slow photoelectrons are reduced in intensity. The fringe spacing of h/τ corresponding to 45 meV (cf figure 2 at $0.25T_B$) is determined by the delay τ between the pulses [47]. The photoelectron spectrum at $0.5T_B$ is very similar to the reference spectrum. At $0.75T_B$ an opposite structure of the AT doublet is found. Here, the slow photoelectrons are enhanced and the fast ones are reduced. In addition, the interference fringes within the fast AT component as observed at $0.25T_B$ have vanished. Due to the off-resonant excitation and the reduced resolution of the photoelectron spectrometer for very low photoelectron energies, the fringes are not seen in the slow photoelectrons. Since the photoelectron spectrum maps the population of the dressed states [5], the experimental results demonstrate the preferential population of one dressed state during the excitation/ionization process. Apparently, the selectivity is controlled by the relative phase of the second laser pulse via the delay. The relative intensity of the AT doublet could also be controlled with the laser intensity used as control parameter [45].

4.2. Discussion

The experimental observations shown in figure 2 are analysed in the picture of the bare state and the dressed state population where dressed states are eigenstates of the Hamiltonian which includes laser–atom interaction. Since the observed photoelectron spectra are periodically reproduced every Bohr transition period T_B, we discuss the reference case (figure 2, $0T_B$) and the preferential population of the upper dressed state (figure 2, $0.25T_B$). The physical mechanism for $0.5T_B$ is similar to the reference case and the situation of enhanced lower dressed state population ($0.75T_B$) is analogous to $0.25T_B$, where the roles of the upper and lower dressed states are reversed. For clarity, in the theoretical analysis, we present the SPODS mechanism for resonant excitation. For the simulation, the intensity is so chosen that the first pulse prepares the atoms at half population, i.e. $|c_{4s}|^2 = |c_{4p}|^2 = 0.5$. To reproduce the observed AT splitting of about 190 meV, the intensity was adjusted to separate both AT components. For a 30 fs laser pulse, the theoretical AT splitting of $\hbar\Omega$ corresponding to 160 meV exceeds the laser bandwidth of about 60 meV. Figure 3(a) shows the electric field envelope $|\mathcal{E}(t)|$ and the temporal optical phase $\chi(t)$ for two $\theta = \frac{7\pi}{2}$ pulses separated by $\tau = 146.1$ fs ($T_B = 0$). The time evolution of the excited bare state population $|c_{4p}(t)|^2$ is depicted in (b). The first pulse induces Rabi oscillations of the 4p state population. The interaction with the second pulse leads to a continuation of the dynamic. As seen in figure 3(c) both dressed states are equally populated $|d_u(t)|^2 = |d_l(t)|^2 = 0.5$ during the whole pulse sequence. Generally, the excitation of ground state atoms with pulses of constant optical phase does not permit SPODS. The photoelectron spectrum depicted in figure 3(e) shows an AT splitting of $\Delta E = 0.16$ eV in agreement with the maximum splitting of the dressed states ($\varepsilon_u - \varepsilon_l$) shown in figure 3(d). In addition, both AT components show interference fringes with an energy spacing of h/τ which are explained in terms of the final state interference of free electron wavepackets [47]. An alternative interpretation on the basis of the Young double-slit experiment is presented in section 4.3. In the second example the delay is set to 146.7 fs, i.e. a quarter of the Bohr period $0.25T_B$ later. The envelope of the pulse sequence looks very similar to the previous case but the optical phase jumps by $\frac{\pi}{2}$ in between the two pulses. As a consequence, the time evolution of the excited state population is remarkably different (figure 3(b), $0.25T_B$). The bare state population is frozen during the interaction of the atom with the second intense resonant laser pulse. This is the bare state manifestation of SPODS. Indeed, figure 3(c) shows that during the second pulse only the upper dressed state is populated. As seen in figure 3(e) the low energy photoelectrons are reduced in intensity and show no internal structure whereas the high energy photoelectrons are enhanced and display interference fringes similar to the experimental photoelectron spectra shown in figure 2 at $0.25T_B$. The close relation of the interference fringes and the selective population of a single dressed state will be discussed in section 4.3. We note that similar to our analysis in view of ultrafast quantum control, so-called spin-locked states are considered in nuclear magnetic resonance [48] and in the optical regime selective excitation of dressed state was achieved using acousto-optic modulation of continuous wave (cw) lasers [32]. Transient dressed state population in an either cw pump and pulsed probe or vice versa configuration was recently studied theoretically [49]. An application of photon-locking to the control of molecules with complex shaped pulses was reported in [50].

4.3. SPODS: analogy to the Young double-slit experiment

In this section the relation of the dynamics in the neutral states and the photoelectron spectrum is elucidated. The dressed state picture is used to provide an intuitive physical picture of the observed photoelectrons. To this end, we make use of the fundamental complementarity of the 'which way' information and the observation of interference fringes which is the physical essence of the celebrated Young double-slit experiment. We analyse our experiment in terms of a 'double slit in time', i.e. we interpret the interference pattern in the photoelectron spectrum as a manifestation of our ignorance of whether the photoelectrons were produced during the interaction with the first or the second pulse. Figure 4 illustrates how this interpretation works in detail. The strong field interaction of the first pulse with the atom gives rise to the formation of the (equally populated) dressed states. Since the photoelectron spectrum maps the population of the dressed states, an unstructured AT doublet as shown in figure 4(a) is measured in this single-pulse case. If the second laser pulse ionizes the equally populated dressed states similar to the first pulse (figure 4(b)), we no longer know whether the electrons are produced during the first or the second laser pulse. Therefore, both AT components show interference fringes due to the final state interference of evolving free electron wavepackets [47]. In the experimental photoelectron spectrum (figure 2 at $0T_B$) the fringes in the fast photoelectrons are visible, whereas the fringes are not resolved in the low energy part of the photoelectron spectrum. If the phase of the second pulse is such that it creates SPODS, then the second

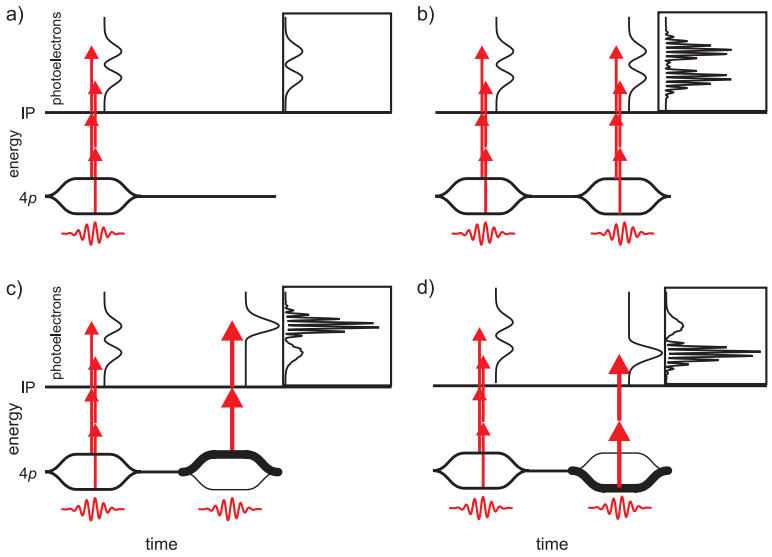

Figure 4. Analogy to the double-slit experiment. (a) Two-photon ionization with a single pulse. Photo-ionization with two pulses (b) if both dressed states are equally populated, (c) if the upper dressed state is selectively populated during the second pulse and (d) if the lower dressed state is selectively populated during the second pulse. The insets show the coherent sum of the photoelectron spectra generated during both pulses, i.e. the measurable spectrum.

pulse produces either exclusively high (c) or low (d) energy photoelectrons. If, for instance, the upper dressed state was selectively populated (figure 4(c)) during the second pulse, the low energy photoelectrons originate from the first pulse. Therefore, the fringes at low kinetic energies have disappeared. In contrast, the ionization pathway for the high energy photoelectrons is unknown and consequently the interference pattern is visible as seen in figure 2 at $0.25T_B$. Obviously, the same principle is operative for the selective population of the lower dressed state as shown in figure 4(d). Here, the fringes in the fast AT component have vanished as also seen in the experimental photoelectron spectra shown in figure 2 ($0.75T_B$).

5. Adaptive feedback control

As stated in the introduction, quantum control by adaptive feedback controlled optimization has been proposed by Judson and Rabitz and today been implemented in numerous experiments ranging from the control of atoms and molecules to the generation of optimized attosecond pulses. In this section, we study the adaptive optimization of one AT component to optimize transient SPODS, i.e. SPODS during the most intense parts of the shaped pulse. In the above two-pulse experiment, SPODS is realized during the second laser pulse with unit efficiency. However, ionization during the first pulse always provides a constant background of symmetric AT signal which cannot be controlled by the phase of the second pulse. Therefore, we investigate if complex pulses exist which optimize transient SPODS beyond the level of control achieved in the two-pulse experiment. A computer controlled phase modulator [30] produces complex shaped femtosecond laser pulses from the initial bandwidth limited 30 fs, 785 nm pulses. We use a linear combination of the area of the fast photoelectrons F and the area of the slow photoelectrons S (see figure 5(a)) as the fitness value ($f = 5F - S$) of each pulse shape to optimize one AT peak against the other and simultaneously obtain the highest possible absolute intensity. The factor of five appears in the fitness function in order to favour the production of fast photoelectrons over the reduction of slow component. The fitness is optimized by the variation of the pulse shape using an evolutionary optimization algorithm. Consequently, the maximum attainable control in this scenario is the complete elimination of one AT component while enhancing the other. Figure 5 shows the experimental photoelectron spectra during the adaptive optimization of the fast photoelectrons. The reference photoelectron spectrum at the beginning of the optimization procedure is depicted in figure 5(a). It is clearly seen that the intensity of the fast photoelectrons rises with increasing number of iterations (b)–(d) beyond the initial intensity indicated by the bold lines. Simultaneously, the slow photoelectrons are significantly reduced in intensity compared to the reference indicated by the dashed line. Normalized to the reference values, the intensity ratio of the AT components—and hence the selectivity—has been increased by a factor of six. This result shows that by use of suitably shaped pulses transient SPODS can be realized with almost 100% efficiency.

6. Conclusion

We studied the light induced dynamics of K atoms excited by a two-pulse sequence in order to analyse a physical mechanism of strong field quantum control based on the selective population of dresses states (SPODS). The intensity of the AT peaks in the photoelectron spectrum was observed as a measure of the dressed state population. By analogy to the Young double-slit experiment an intuitive physical picture for the mapping of the dressed states via the photoelectron spectrum was given. Our results demonstrate our ability to control SPODS. The physical mechanism of strong field control was analysed in some detail. The analysis showed that SPODS was achieved by phase control in intense fields. Using

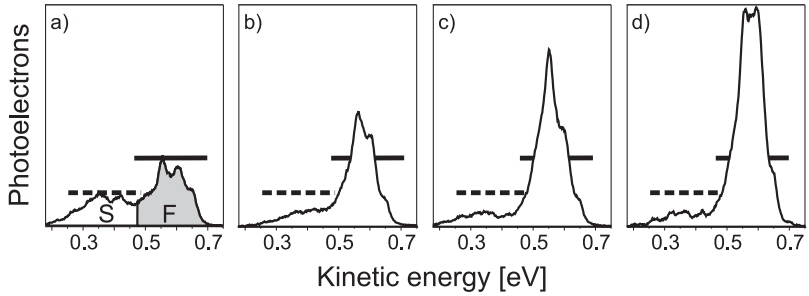

Figure 5. Evolution of the photoelectron spectra during the adaptive optimization of the fast versus the slow photoelectrons. The fitness function is defined as $f = 5F - S$, where F denotes the area of the fast photoelectrons (grey shaded) and S the area of slow photoelectrons. The number of iterations increases from (a) to (d). The lines indicate the reference intensities of the slow (dashed) and fast (bold) photoelectrons at the beginning of the optimization procedure. The optimal pulse (d) realizes the population of the upper dressed state during ionization with very high selectivity.

adaptive optimization, the degree of attainable selectivity could be drastically improved, indicating that SPODS is a general strong field mechanism which is also valid for complex pulse shapes. In the future, we will analyse the dynamics induced by complex pulses obtained from optimization to gain further insights in physical mechanisms of strong field optimal control. Since SPODS is highly sensitive to the coherence of the system it may serve as a tool to study decoherence phenomena relevant to quantum computation. Creation of a superposition state with a first $\frac{\pi}{2}$-pulse and subsequent probe with a phase coherent more intense pulse allows us to measure the system's decoherence. Since the photoelectrons are produced almost exclusively during the second laser pulse, the photoelectron spectrum is determined by the coherence of the system at that time. The gradual loss of the coherence appears as a reduction of the asymmetry of the AT components. This is a direct measurement of the decoherence time and could help to elucidate decoherence mechanisms. Most likely SPODS is at play in molecular systems as well. Corresponding experiments on molecules are currently being prepared in our laboratories.

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