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Photoelectron angular distributions from strong-field coherent electronic excitation

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Abstract Photoelectron Angular Distributions (PADs) resulting from nonperturbative excitation of potassium atoms using shaped femtosecond laser pulses are presented. We study control exerted by (1) the polarization of an unshaped, i.e., a bandwidth-limited light pulse, (2) shaped linearly polarized light, and (3) a combination of both degrees of freedom, i.e., polarization-shaped laser pulses. A theoretical approach to describe PADs from nonperturbative Resonance Enhanced Multi-Photon Ionization (REMPI) with ultrashort polarization-shaped laser pulses is presented and compared to experimental results. Applications of this technique to the generation and observation of atomic ring currents are discussed.

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1 Introduction

The experimental realization of shaped ultrashort laser fields [1, 2] is at the heart of coherent control. Control scenarios ranging from "open-loop" quantum interference on model systems to adaptive control schemes applied to systems with ever increasing complexity [3–13] have been studied theoretically and demonstrated experimentally. In these experiments, highly structured—but in most cases linearly polarized—femtosecond laser pulses have been employed to control light-induced dynamics of quantum systems. Recently, with the advent of pulse shapers that are capable of generating complex shaped time-dependent polarization profiles in addition [14–19], experimental demonstrations of quantum control by making use of polarization-shaping as additional degree of freedom [20–23] have been presented. Another degree of freedom in quantum control is accessible by intense laser fields. In those fields the light–matter interaction is highly nonperturbative and gives rise to level-shifts in the order of hundreds of meV due to AC–Stark shifts. Strong-field coherent control has been demonstrated by dynamic Stark splitting of resonances [24–27] and nonresonant Stark shifts of molecular potentials [28, 29].

In this contribution, we present a coherent control scenario making use of nonperturbative control by intense polarization-shaped laser pulses. In particular, we extend earlier studies on coherent control of electronic excitation by shaped linearly polarized light to nonperturbative light–matter interaction using polarization-shaped laser pulses and detection by Photoelectron Angular Distributions (PAD). Our previous strong-field experiments have been interpreted in terms of Selective Population of Dressed States (SPODS) [30, 31], which provides a unifying framework for established strong-field mechanisms such as Photon Locking (PL) [32–34] by temporal phase discontinuities and Rapid Adiabatic Passage (RAP) [35] by chirped pulses. Realizations of SPODS were demonstrated by using pulse sequences [24, 36], shaped pulses such as sinusoidally phase-modulated pulses [37, 38], chirped pulses [39], and combinations thereof [40, 41]. Physical pictures to describe the mechanism of SPODS involve bare states dynamics [24], dressed states dynamics [30], dynamics of the Bloch-vector [11, 38, 39], and recently a spatio-temporal picture considering the dynamics of coherent electronic wave packets in shaped laser fields [40]. In that contribution, wave packets made of superpositions of $|s, m = 0\rangle$ and $|p, m = 0\rangle$

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states were produced and manipulated by linearly polarized shaped pulses, and the transient dynamics was mapped by simultaneous ionization. An alternative approach to observe coherent transients from shaped femtosecond excitation of atoms by pump–probe techniques was initially reported in [42] and recently applied to photoassociation of ultracold molecules [43]. In this contribution we report on $1+2$ Resonance Enhanced Multi-Photon Ionization (REMPI) experiments employing polarization-shaped laser pulses to (1) control electronic wave packets in the neutral atom as a model system for controlled (neutral) molecular dynamics and (2) exert control on free-electron wave packets by multi-photon ionization. In the first step polarization-shaped pulses are used to excite tailored superpositions of the $|s, m=0\rangle$ and $|p, m=\pm 1\rangle$ electronic states. In the second step multi-photon ionization with photons of specifically tailored polarization allows us to exert control over a whole manifold of electronic states such as the *virtual* $|d, m=0\rangle$ and $|d, m=\pm 2\rangle$ states as well as the final free $|f, m=\pm 1\rangle$ and $|f, m=\pm 3\rangle$ states. In general, as exemplified above, tailoring the state of polarization gives access to controlling angular momenta and—associated with this—the ability to shape the angular distributions of matter waves. In our experiment we shape ultrashort *free electron wave packets* [44] by ultrashort polarization-shaped laser pulses. Detection of PADs [45, 46] is the adequate technique to directly map the spatial facet of coherent strong-field electronic excitation by ultrashort polarization-shaped laser pulses and thereby help us to unravel strong-field dynamics induced by polarization-shaped light. Combining polarization-shaping with detection of PADs has high potential for novel applications such as the observation of controlled dynamics of chiral molecules, the characterization of attosecond dynamics and the generation and detection of atomic ring currents [47]. Initial experimental results relevant to atomic ring currents are also presented in this contribution.

Our contribution is organized as follows. We will start in Sect. 2 with the theoretical description of polarization-shaped laser fields and their interaction with atomic quantum systems. Strong-field neutral excitation and perturbative multi-photon ionization of potassium atoms are discussed consecutively in order to derive a handling of both the radial and the angular distribution of emitted photoelectrons. In Sect. 3, our experimental apparatus is described, including the polarization pulse shaper and the photoelectron imaging spectrometer. Section 4 gives the results obtained from interaction of potassium atoms with the unmodulated pulse, purely phase-shaped pulses and, finally, with complex polarization-shaped laser pulses. We end this article with a brief summary and conclusions.

2 Theory

In this section, we describe $1+2$ REMPI of atoms by ultrashort laser pulses with an arbitrary instantaneous state of polarization detected by PADs loosely following the notation of [48] and [49]. To this end, we introduce the laboratory frame coordinate system in which the (time-dependent) polarization vector $\hat{\boldsymbol{\epsilon}}(t)$ of the laser pulse propagating along the z-axis is located in the x–y-plane (see Fig. 3(e) for a sketch of the coordinate system). PADs are generated by acceleration of the electrons in x-direction. In the following sections we start by introducing polarization-shaping in the frequency domain specifically adapted to describing the interaction of ultrashort polarization-shaped laser pulses with atoms. Subsequently, one-photon strong-field interaction in neutral potassium atoms is looked at by solving the time-dependent Schrödinger equation. Simultaneous multi-photon ionization of the interacting system is treated perturbatively, yielding three-dimensional free-electron wave packets. PADs are obtained by projecting these wave packets onto a two-dimensional detector plane.

2.1 Polarization-shaping

In view of a consistent *quantum mechanical* description of the light–atom interaction given below, the electric field is described in terms of its (negative frequency) analytical signal $\mathcal{E}^-(t)e^{-i\omega_0 t}$ characterized by the complex valued envelope $\mathcal{E}^-(t)$ and oscillating at the central frequency of the laser radiation ω_0. The positive frequency analytical signal customarily used in femtosecond optics [1, 2] is related to our notation by $\mathcal{E}^+(t) = [\mathcal{E}^-(t)]^*$. In order to simplify the notation, the superscript "−" is omitted throughout the remainder of this article. Fourier-transform of the envelope delivers the spectrum $\tilde{\mathcal{E}}(\omega)$. Independent spectral phase-shaping of two orthogonal linearly polarized components of $\tilde{\mathcal{E}}(\omega)$ by the phase functions $\varphi_x(\omega)$ and $\varphi_y(\omega)$

$$\begin{pmatrix} \tilde{\mathcal{E}}_{x,\mathrm{mod}}(\omega) \\ \tilde{\mathcal{E}}_{y,\mathrm{mod}}(\omega) \end{pmatrix} = \tilde{\mathcal{E}}(\omega) \begin{pmatrix} e^{i\varphi_x(\omega)} \\ e^{i\varphi_y(\omega)} \end{pmatrix} \tag{1}$$

yields two linearly polarized modulated spectral light fields $\tilde{\mathcal{E}}_{x,\mathrm{mod}}(\omega)$ and $\tilde{\mathcal{E}}_{y,\mathrm{mod}}(\omega)$ resulting in the shaped temporal fields $\mathcal{E}_{x,\mathrm{mod}}(t)$ and $\mathcal{E}_{y,\mathrm{mod}}(t)$ obtained by inverse Fourier-transform. A more detailed characterization of the experimental implementation of the polarization-shaper is given in Sect. 3.2. In order to describe the light–atom interaction of a laser electric field being in a state of arbitrary polarization, it is advantageous to decompose the field into superpositions of left-handed $\mathcal{E}_L(t)$ and right-handed $\mathcal{E}_R(t)$ circularly polarized shaped light pulses

$$\begin{pmatrix} \mathcal{E}_L(t) \\ \mathcal{E}_R(t) \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} \mathcal{E}_x(t) - i\mathcal{E}_y(t) \\ \mathcal{E}_x(t) + i\mathcal{E}_y(t) \end{pmatrix}, \tag{2}$$