Ultrafast Control of Coherent Electronic Excitation

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With the advent of ultrashort femtosecond laser pulses the temporal aspect of the interplay of light and molecular dynamics came to the fore and became experimentally accessible. The beauty of femtochemistry lies in our ability to observe [1] and to manipulate [2, 3] ultrafast processes as they occur. Shaped femtosecond optical laser pulses [4] are the suitable tools to exert microscopic control on molecular dynamics at the quantum level. The combination of pulse-shaping techniques with closed loop adaptive feedback learning algorithms [5–8] allows to optimize virtually any conceivable observable as reviewed for example in [9, 10]. However, it is not always possible to deduce the underlying physical mechanism from the electrical fields obtained by this procedure. Therefore, the need to bridge the gap between the efficient 'black box' closed loop optimal control methods and detailed understanding of the physical processes especially in strong laser fields is quite evident. To that end we combine femtosecond laser techniques with atomic-/ molecular beam techniques and photoelectron-/ ion detection techniques [11]. So far we have extended weak field methods to free electrons [12]. New techniques making use of polarization control in molecular multi-photon excitation [13] and shaped intense laser pulses for molecular alignment [14] open further dimensions in this field.

An exciting new strong field control scenario is based on ultrafast control of electronic coherent excitation. This approach makes explicit use of the manipulation of the temporal phase of a pulse sequence with attosecond precision [15]. Experimentally we make use of a 1+2 REMPI process on potassium atoms. An intense fs-laser couples coherently the $4s$ – $4p$ level and at the same time ionizes the system in a two photon process

(see Fig.1). The shape of the photoelectron spectra reflects the temporal phase of the excited state amplitude $c_{4p}(t)$ [11].
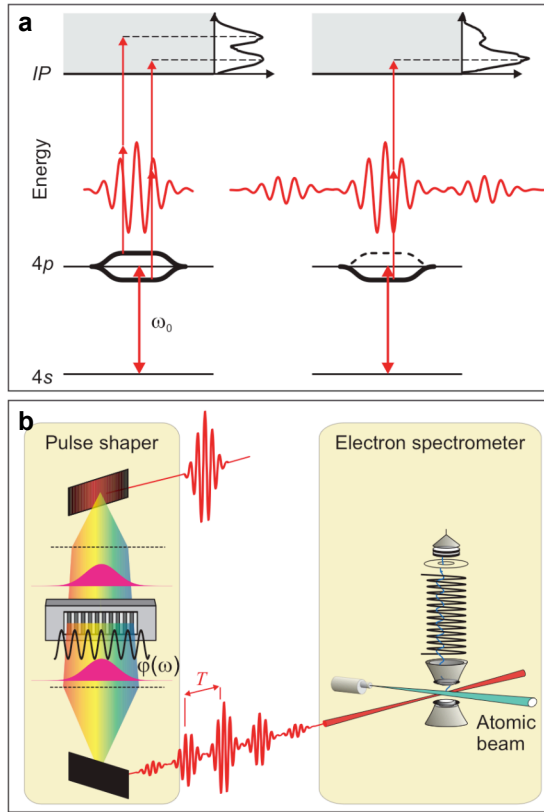

FIG. 1: (a) Schematic of the excitation scheme (potassium-atoms): The bare states are indicated with thin lines. Thick lines illustrate the dressed state splitting during the interaction giving rise to a symmetric Autler Townes splitting (left). Selective population of a dressed state with a tailored pulse train is shown in the right panel, leading to a strongly asymmetric Autler Townes doublet. (b) Schematic of experimental set-up: tailored pulse trains are created via applying a phase mask in the Fourier plane of our pulse shaper [16]. In the case discussed here, the spectrum of our femtosecond laser pulse (785 nm, 30 fs, 0.35–2 µJ) is phase-modulated in frequency domain with a sinusoidal phase function $\varphi(\omega) = A \sin[(\omega - \omega_0) \ T + \phi]$ with $A = 0.2$, $T = 170$ fs and $\omega_0 = 2.40$ fs^{-1} to produce a sequence of pulses in time domain separated by T. The pulses are focused on a potassium atomic beam. The resulting photoelectrons are detected with a magnetic bottle Time of Flight photoelectron spectrometer.
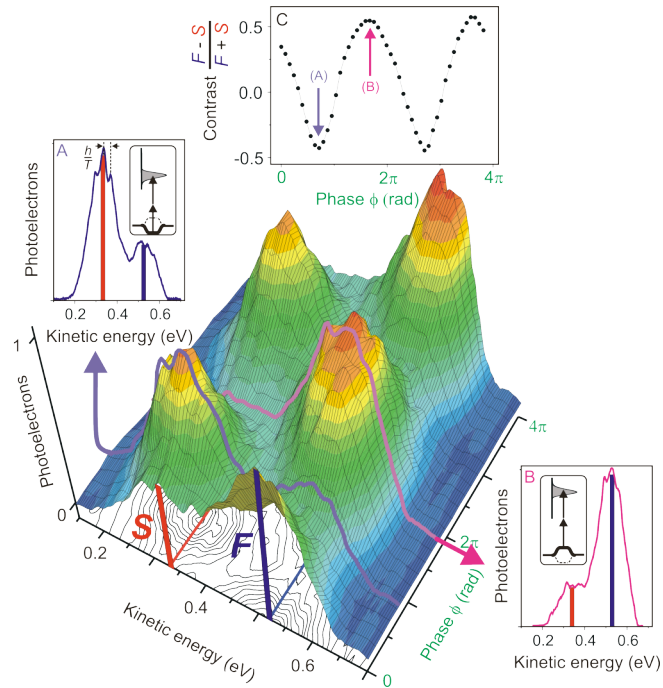
FIG. 2: The Selective Population of Dressed States (SPODS) is directly mapped into the measured photoelectron spectra by variation of the phase ϕ. The maximum of the asymmetric photoelectron distribution alternates between 0.33 eV and 0.52 eV. These results are obtained at a laser energy of $W = 0.5$ µJ. A section through the distribution along the energy axis at $\phi = 0.7\ \pi$ – indicated with a trajectory – yields the photoelectron spectrum (A) where the lower dressed state is selectively populated as depicted in the inset to (A). Fringes in the spectrum with an energy separation of h/T arise from the interference of the free electron wave packets [12] launched during the different pulses. Selective population of the upper dressed state is achieved at $\phi = 1.7\ \pi$ as indicated with a trajectory and plotted in spectrum (B). The signal of the slow photoelectrons at 0.33 eV (S) and the fast photoelectrons at 0.52 eV (F) – as indicated with the bars – is obtained as a function of the phase ϕ by taking a section through the distribution along the phase coordinate. The contrast of F and S, *i.e.* (F–S)/(F+S) as shown in (C) is a measure of the selectivity of dressed state population. The phases corresponding to the highest selectivity for population of the lower dressed state – spectrum (A) – and the upper dressed state – spectrum (B) – are indicated with arrows.

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In particular, the photoelectron spectra map the dressed state population. During the time evolution, the dressed states are characterized by a time-dependent energy splitting giving rise to the observed Autler-Townes (AT) splitting [17] in the photoelectron spectra. Employing two-photon ionization as the non-linear probe step precludes averaging over the intensity distribution within the laser focus since the ionization probability is highest in the spatial region of highest laser intensity. This technique permits us to overcome the common problem of washing out intensity dependent strong field effects. Making use of adaptive feedback learning algorithms we are able to control the dressed state population by more than 90% as seen by the corresponding suppression of one AT component [18]. With the help of tailored pulse trains we demonstrate that this Selective Population of Dressed States (SPODS) is highly selective, tunable (up to 250 meV) and robust [19]. In Figure 2 experimental results – obtained with a pulse train created by applying a sine mask in the Fourier plane of the pulse shaper (see Fig. 1) – are displayed.

Since switching between selective population of either dressed states occurs within a few femtoseconds, this technique is also interesting for applications in the presence of decoherence processes. SPODS can be realized with very different pulse shapes making use of diverse physical mechanisms ranging from Photon Locking [15, 19, 20] to Rapid Adiabatic Passage [21]. Because SPODS combines high selectivity and tunability with efficient population transfer, relevant applications to chemistry – so far investigated theoretically [19, 22] – are within reach.

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