Femtochemistry and Femtobiology
M.M. Martin and J.T. Hynes (editors)

Quantum control beyond spectral interference and population control: Can resonant intense laser pulses freeze the population?

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1. INTRODUCTION

Many different schemes for quantum control have been demonstrated [1]. Prominent examples of which are the Brumer-Shapiro scheme [2], the Tannor-Kosloff-Rice scheme [3] and techniques based on (higher order-) spectral interference. The striking experiments of Silberberg nicely demonstrated the principle of second order spectral interference [4] and recently applications of higher order spectral interference to large organic molecules in solution were reported [5]. However, Rabitz et al. [6] pointed out that "the operating principle for quantum control of any type is the manipulation of constructive and destructive quantum mechanical interferences". Therefore, the ability to measure and to control the quantum mechanical phase is the key step towards a deeper understanding of quantum control. Since the energy resolved photoelectron spectra from simultaneous excitation and ionization are directly related to the temporal evolution of the excited state (population and phase), this technique is most suited to elucidate details of the quantum control dynamics [7]. In particular, the use of pulse sequences has proven a strong tool to study interference effects in atomic and molecular systems in detail [8]. This scheme was extended to the continuum in order to demonstrate the coherence transfer from femtosecond laser pulses to ultrashort free electron wave packets [9]. A variety of important control mechanisms are only accessible when strong laser fields are employed [10]. Examples of coherent control by intense sequential laser pulses are coherent transients such as the photon echo and Ramsey fringes [11] as well as the STIRAP [12].

In this contribution recent results [13] on the control of the quantum mechanical phase of an atomic state in strong laser fields studied using the Autler-Townes (AT) effect [14] in the photoionization of the K ($4p$) state are discussed. We demonstrate quantum control beyond (i) population control and (ii) spectral interference. (i) We show, that for suitable combinations of the laser intensity of the first pulse and the time delay the second resonant intense laser pulse leaves the excited state population unchanged. However, the knowledge of the

temporal evolution of the population is not sufficient since the second laser directly manipulates the quantum mechanical phase which significantly changes the outcome of the experiment. (ii) Control beyond spectral interference is achieved by controlling the quantum mechanical phase without changing the spectrum of the pulse sequence, i.e., since no phase modulation is applied and only the laser intensity is varied, the N-th order spectra are unchanged. Moreover, an uncommon symmetry of the control parameters delay time and laser intensity is observed: with respect to the interferences in the photoelectron spectra the role of time delay and laser intensity is interchangeable for suitable excitation conditions. This new control mechanism combines phase control and high intensity effects through the use of phase locked pulses and intensities that are large enough to cause Rabi cycling. We believe, that this mechanism is at play in many other circumstances as well and that it opens the door to a deeper understanding of quantum control in intense laser fields.

In our experiment a sequence of two intense laser pulses is used to excite K atoms in an atomic beam from the $4s$ to the $4p$ state. Simultaneously, the pulses ionize the excited atoms from the $4p$ state to the continuum via two-photon ionization (Fig. 1). Photoelectron spectra were taken at various delay times between the two laser pulses and at different laser intensities at a fixed delay time.

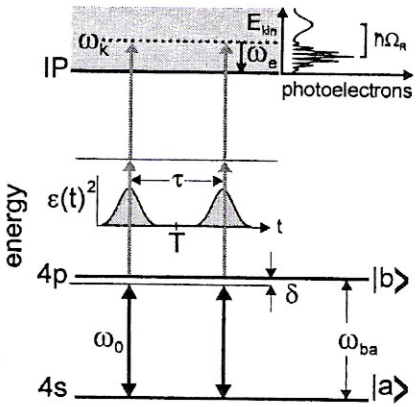

Fig. 1. Energy level diagram for excitation of K atoms.

2. EXPERIMENT

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 photoionization. The released photoelectrons are detected employing a magnetic bottle time-of-flight electron spectrometer. The 785 nm, 30 fs FWHM laser pulses provided by an amplified 1 kHz Ti:sapphire laser system are split into two beams using a Mach-Zehnder interferometer. In the first experiment the time delay τ is varied in a range of 80 to 100 fs with 0.2 fs resolution at a

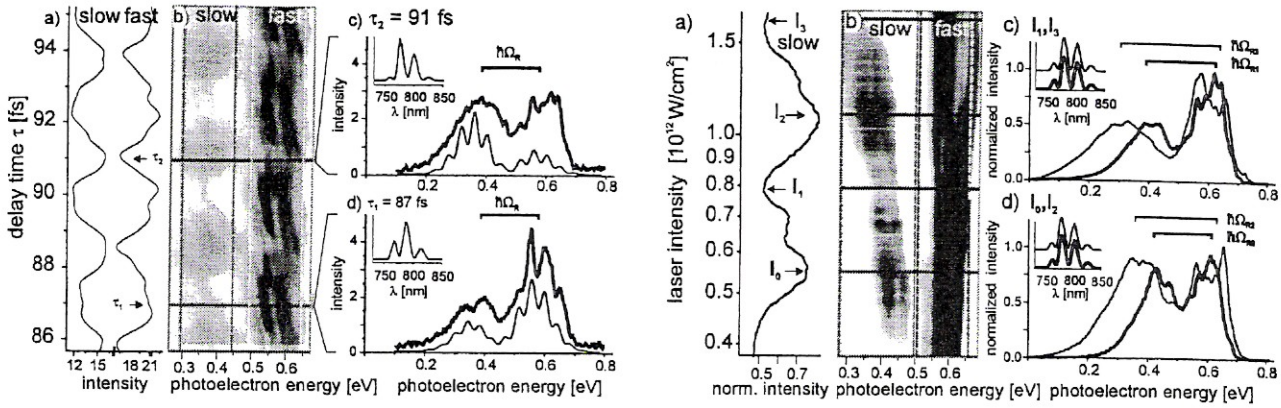

Fig. 2. Left: experimental photoelectron spectra as a function of the delay time τ. Right: Measured photoelectron spectra as a function of the laser intensity at a fixed delay time.

fixed laser intensity of I_0 (0.54×10^{12} W/cm^2). In the second experiment the time delay is kept fixed at 98.6 fs, whereas the energy of both beams is varied from 0.7 I_0 to 3 I_0.

3. RESULTS

At first, we discuss the results of the experiment using a variable time delay at a fixed laser intensity of I_0. Fig. 2(b, left) shows the measured photoelectron spectra as a function of the delay time. Oscillations in the photoelectron signal at the period of the photon frequency of 2.6 fs are observed. The oscillations of slow and fast photoelectrons are out-of-phase. Sections through the photoelectron distribution were taken along the time delay axis for the fast and the slow photoelectrons. In the second experiment the laser intensity was varied but the time delay was fixed Fig. 2(b, right) in order to demonstrate that the control of the interferences is determined by the quantum mechanical phase. The optical spectrum of the pulse sequence remains unchanged for all intensities. A monotonic increase of the splitting of both AT components with increasing laser intensity is observed. The slow electron signal exhibits pronounced oscillations as the intensity is increased.

4. DISCUSSION

The intensity of the first laser is high enough to cause Rabi cycling and, therefore, AT splitting in the photoelectron spectrum. The observed control of interference in the AT doublet arises if the intensity of the first pulse is chosen to yield a population of $|c_b(T)|^2 = 0.5$, i.e. a pulse area of $\Theta = (n+0.5)\pi$. The subsequent time evolution ($t > T$) of c_b depends on the phase $\omega_0\tau$ of the second laser pulse. If the phase $\omega_0\tau$ takes half integer multiples of π, $|c_b(t)|^2$ is unchanged during the second laser pulse and the quantum mechanical phase is altered. With

regard to photo ionization the observations are interpreted in terms of the population of the dressed states. For suitable excitation only one of the dressed states is selectively populated and hence, interference is only seen in one AT component.

5. CONCLUSION

We demonstrate coherent control in strong fields beyond (i) population control and (ii) spectral interference, since (i) control is achieved without altering the population during the second intense laser pulse, i.e., the population during the second laser pulse is frozen, and (ii) the quantum mechanical phase is controlled without changing the spectrum of the pulse sequence. The control mechanism relies on the interplay of the quantum mechanical phase set by the intensity of the first pulse and the phase of the second pulse determined by the time delay.

ACKNOWLEDGEMENT

The support of the Deutsche Forschungsgemeinschaft, the Fonds der Chemischen Industrie and the NRC-Helmholtz program is gratefully acknowledged.

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