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### Authors

Bernhardt, B

Li, X

Beck, AR

et al.

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# Attosecond Transient Absorption Explores Coupling Mechanisms of Autoionizing States

Birgitta Bernhardt<sup>1,2,\*</sup>, Xuan Li<sup>1</sup>, Annelise R. Beck<sup>1,2</sup>, Erika R. Warrick<sup>1,2</sup>, Daniel J. Haxton<sup>1</sup>, C. William McCurdy<sup>1,3</sup>, Daniel M. Neumark<sup>1,2</sup> and Stephen R. Leone<sup>1,2,4</sup>

<sup>1</sup>Ultrafast X-ray Science Laboratory, Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

<sup>2</sup>Department of Chemistry, University of California, Berkeley, California 94720, USA

<sup>3</sup>Department of Chemistry, University of California, Davis, California 95616, USA

<sup>4</sup>Department of Physics, University of California, Berkeley, California

\* E-mail address: BCBernhardt@lbl.gov

**Abstract:** Attosecond transient absorption spectroscopy of highly excited states allows investigating the dynamics of both the decay and the light induced coupling between excited states. We present a rigorous quantum mechanical treatment that provides a generalized discussion of the on- and off-resonant decay dynamics, of resonant and non-resonant couplings to either discrete or continua states and the temporal evolution of the quantum beating of the excited wavepackets. The comparison of the model to experimental results obtained in xenon proves its general applicability.

**OCIS codes:** (320.7150) Ultrafast spectroscopy, (320.7120) Ultrafast phenomena, (320.7100) Ultrafast measurements

Attosecond transient absorption spectroscopy has become the method of choice for time-resolved studies of light-field induced coupling dynamics between highly excited atomic states. In this scheme, an attosecond extreme ultraviolet (XUV) pulse excites several (autoionizing) states and stimulates a transient oscillating dipole moment. This induced time-dependent polarization can be dressed by a time-delayed near infrared (NIR) pulse, resulting in the depletion of the autoionizing states. Due to the relation between polarization and population, the absorbance signal at a resonance decays with twice the state lifetime and hence, state lifetimes can be directly derived from transient absorption measurements. We recently succeeded in confirming lifetime values of the xenon  $5s5p^66p$  and  $5s5p^67p$  states to be  $(21.9 \pm 1.3)$  fs and  $(48.4 \pm 5.0)$  fs that were so far determined only indirectly from linewidth measurements [1].

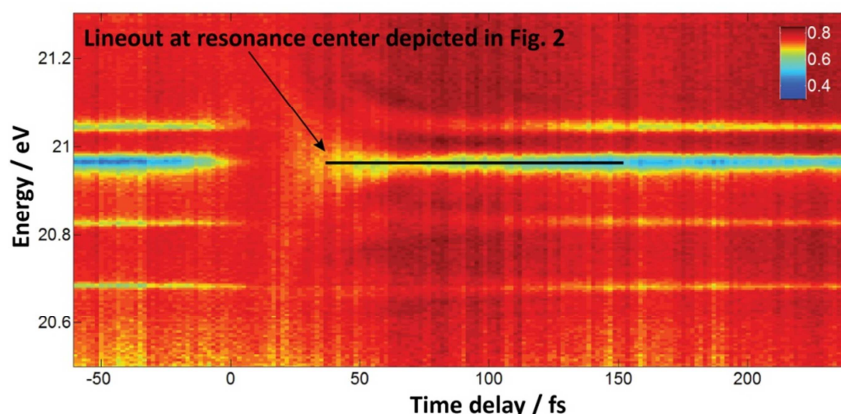


Fig. 1 Transient absorption spectrum in the vicinity of the  $5s5p^66p$  state of xenon at 20.95 eV showing also three neighboring states. Negative delay times correspond to the NIR pulse arriving before the XUV pulse at the sample.

Figure 1 depicts the measured absorbance signal of xenon (expressed as optical density  $OD = -\log_{10}(I_t/I_0)$  with  $I_0$  being the incident XUV intensity and  $I_t$  the intensity transmitted through the gas).

An exponential fit of the observed decay dynamics at the  $6p$  resonance center yields the state lifetime of  $(21.9 \pm 1.3)$  fs (see Ref [2] for details). We have shown that our modelling also predicts the off-resonant absorbance behavior

[2]. The quantum mechanical formalism treats the NIR pulse as a sudden annihilator of the oscillating and decaying dipole and reproduces the experimentally observed energy-dependent absorbance behavior. Here, we have extended the formalism to fully account for i) the direct ionization of the highly excited 6p state into the continuum, ii) the coupling to an individual neighboring state or iii) the resonant coupling of the 6p state to two resonances in the vicinity resulting in an additional oscillation superimposed to the decay signal.

In this conference contribution, we compare experimental data with the theoretical predictions and show that the formalism as outlined in [2] grasps each of the specific coupling mechanisms. Based on a few level simulation we explore the signatures of the transient absorption signal in the i) discrete-continuum and the ii) discrete-discrete coupling case, each time for different NIR intensities, and compare it to our experimental investigations. Specific attention is given to the discrete-discrete coupling case that can result in complex wave packet dynamics which we have observed recently as quantum beats in long lived Rydberg states in neon [3].

Furthermore, as shown in Fig. 2, the signal recorded at the 6p resonance center yields an oscillation with a period of approximately 29 fs. This oscillation can be attributed to the quantum interference with a neighboring resonance. This two-path interference results from one quantum path being the excitation of the 6p state by one XUV photon. The second quantum path describes the absorption of an XUV photon followed by absorption and emission of NIR photons via an intermediate state, resulting in the same final state as created by the attosecond probe alone. The interference between these two paths yields a beating pattern with an oscillation period that is reciprocal to the energy difference between the two resonantly coupled states (case iii) mentioned above). Based on the observed beating frequency we identified a neighbor state 140 meV below the 6p state as predominantly contributing to the transition (resonance at 20.81 eV in fig. 1). Our simulations suggest further that the intermediate state involved in the second quantum path is located at  $E_{6p} + \hbar\omega_{\text{NIR}} = 22.51$  eV. This even-parity intermediate state can be identified as the 5s5p<sup>6</sup>s state, measured previously at 22.563 eV by electron impact [4].

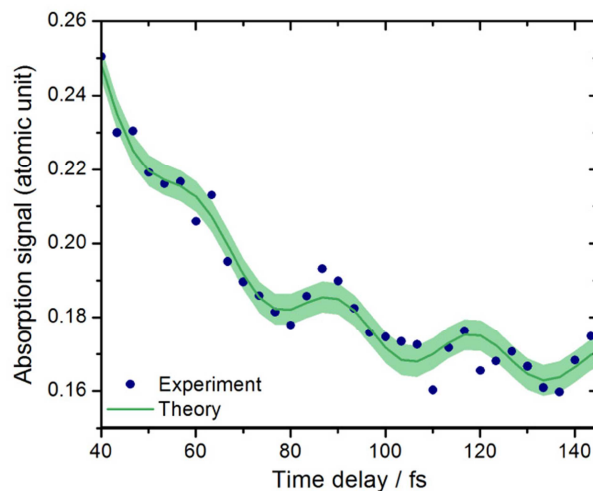


Fig. 2 Signal evolution at the xenon 5s5p<sup>6</sup>6p resonance center with an oscillation superimposed to the exponential decay with a period of about 29 fs, indicative of the quantum beat between the 6p state and the neighbor state located 140 meV below. Blue dots: experimental data, dark green solid line: Theoretical fit with a four-level simulation, light green: corresponding standard deviation.

## References

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