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Autoionizing states in attosecond spectroscopy

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Abstract: Autoionizing states are pervasive features of atomic and molecular ionization processes. We illustrate how the evolution of autoionizing states in polyelectronic atoms can be monitored and controlled with attosecond pump-probe photoelectron and transient-absorption spectroscopy .

1. Introduction

Autoionizing states are a distinct signature of electronic correlation. With the advent of attosecond sources it is now possible to resolve the evolution of these metastable states in real time, and to control them with light [1–4]. Only with the support of new dedicated *ab initio* tools, however, can the promises of attosecond control come to fruition. *Ab initio* time-dependent close coupling has affirmed itself as the leading tool to simulate the ionization of atoms and molecules under the influence of ultrashort pulses, beyond the single-active-electron approximation [5]. In this work I will present recent results obtained with NewStock, a time-dependent program that is able to simulate the photoionization and optical response of small poly-electronic atoms exposed to sequences of light pulses.

2. Theory

NewStock merges the capabilities of the Stock B-spline close-coupling structure code [6], based on the ATSP2K package [7], with those of state-of-the-art time-dependent two-active-electron codes [5, 8]: it builds a close-coupling space in which multi-reference parent ions are augmented with a spherical B-spline basis, and it solves the time-dependent Schrödinger equation with a second-order split-exponential propagator. Channel-specific mask functions periodically extract and analyze the outer part of the state vector, allowing us to simulate the effect of arbitrarily long moderately weak pulses while employing comparatively small quantization boxes. Reflection of the faster components at the box edge is prevented by complex-absorbers. The channel- and energy-resolved photoelectron spectrum is computed by projecting the state function on a complete set of scattering states. The transient-absorption spectrum of the system is obtained from the time-dependent expectation value of the dipole operator, recorded throughout the simulation, until the complete decay of the autoionizing states.

3. Results and discussion

In a first example, we employ attosecond transient-absorption spectroscopy to explore the entangled light-matter character of metastable Autler-Townes multiplets, which makes them autoionizing polaritons, in the Argon atom illuminated by a sequence of extreme ultraviolet harmonics in combination with a mid-IR pulse with a duration of few tens femtoseconds and intensity below 1TW/cm^2. We observe the formation of polariton multiplets between the $3s^{-1}4p$ and several light-induced states, as well as the controllable stabilization against ionization of some polaritons, in excellent agreement with measurements from the group of Arvinder Sandhu, at the University of Arizona (see Fig. 1). Using an extension of the Jaynes-Cummings model to autoionizing states, we show that this stabilization is due to the destructive interference between the Auger decay and the radiative ionization of the polaritonic components.

In a second example, we illustrate how attosecond pump-probe photoelectron spectroscopies can give access to the time-resolved study of the photoelectric effect, a fundamental process that is strongly influenced by electron-electron correlation and in particular by the formation of metastable states. Using the Reconstruction of Attosecond Beating by Interference of Two-Photon Transitions (RABBITT) technique [9, 10], we show how our theoretical predictions for the reconstruction of the ionization phase of the argon atom in proximity of the $3s^{-1}4p$ resonance are in excellent agreement with recent high-resolution measurements by the group of Louis DiMauro, at Ohio State University.

4. Conclusions

The controllable narrowing of autoionizing polaritons' width in argon dressed by mid-IR laser pulses gives new insights into the optical control of electronic structure in the continuum, and unlocks the door to applications of

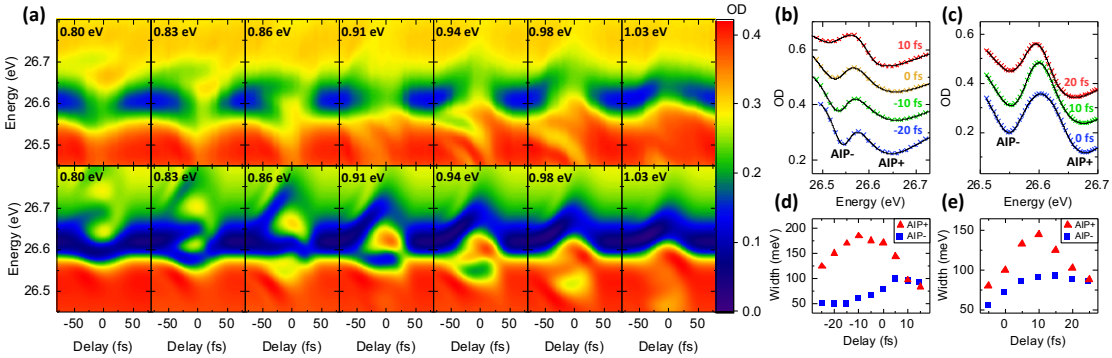

Fig. 1. (a) Exp. (top panels) and Th. (bottom panels) XUV optical density in the vicinity of argon $3s^{-1}4p$ AIS, as a function of the IR pulse delay, for several IR photon energies. Interaction of autoionizing states with light-induced states gives rise to a polariton splitting. Exp. (b) and Th. (c) polaritonic lineshapes for the near resonant case, for several time delays. In absence of interference between radiative and Auger channels, the two polaritons are expected to have comparable widths.

radiative stabilization in metastable poly-electronic systems. Our time-dependent close-coupling simulations of RABBITT resonant photoelectron spectra significantly improves the predictions of earlier models.

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