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Quantifying light-induced linear and nonlinear couplings with tunable transient absorption and four-wave-mixing.

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Light-induced couplings between electronic states can be used to control the optical properties of media. In this context, strong-field transient absorption spectroscopy forms an excellent tool to characterize these couplings, and their effect on electron dynamics, leading to new insights into the light-induced states, autoionization dynamics, light-induced transparency, nonlinear emissions, population transfer etc. We perform such measurements by employing controlled, tunable wavelength infrared pulses to tailor the electronic couplings and study their effect on transient absorption lineshapes. We demonstrate control over Autler-Townes splitting of a bound state, and its evolution to a light-induced structure. We show that non-commensurate extreme-ultraviolet and infrared fields allow background free study of nonlinear four-wave-mixing emissions, which exhibit non-trivial spectral and temporal features. Four-wave-mixing with tunable pulses allows us to quantify the couplings between autoionizing states, study dark state dynamics, and probe the light-induced counterparts of Fano resonances. Systematic studies of light-induced effects help to refine the theoretical models and open up new applications of attosecond transient absorption spectroscopy. This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Award DE-SC0018251.