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Accurate reconstruction of the non-linear response of matter from spectro- scopic absorption data STEFAN DONSA, IVA BREZINOVA, JOACHIM BURGDORFER, Vienna Univ. of Technology, VEIT STOOSS, STEFANO CAVALETTO, ALEXANDER BLATTERMANN, PAUL BIRK, CHRISTOPH KEITEL, CHRISTIAN OTT, THOMAS PFEIFER, Max-Planck-Insitut für Kernphysik — Observing the non-linear electronic response in atoms, molecules, and solids relies on precisely timed interactions with external stimuli. This, typically, requires the time-resolved recording of the response by a probe pulse following the excitation by the pump pulse as a function of the time delay between pump and probe. In this work we report on a new method to obtain time-resolved phase and amplitude information on the non-linear response using just the spec- troscopic data from one single-shot transient absorption measurement. In a proof of principle study we demonstrate the reconstruction of the non-linear dipole moment of doubly excited in Helium driven by an NIR laser pulse. We reconstruct the Rabi-flopping dynamics of the doubly excited states for a wide range of intensities. Comparing the experimental results with ab-initio calculations we are able to identify the states, which participate in the Rabi-flopping process. For stronger NIR intensities we observe that the field-induced ionization dominates over the auto-ionization indicating the break-down of frequently used few-state models.

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