Reconstructing real-time quantum dynamics in strong and short laser fields

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Synopsis: Using ultrashort and phase-locked laser pulses, we present a method that is capable of retrieving the nonlinearly perturbed time-dependent response function of a quantum system driven by electric fields directly from spectroscopic absorption data.

Measurements of time-dependent processes in the quantum world often rely on so-called pump–probe techniques. In these experiments, the dynamics are first triggered by a short light pulse and, after a certain delay, are probed by a second pulse. Time-resolved information is routinely gained by varying the delay between the pump and the probe pulse.

By contrast, for the case of all linear light-matter interaction, the causality of signals that are observed by spectroscopic methods defines the relationship between absorption and dispersion, thus the full temporal response function can be retrieved, with the prominent example of deducing lifetimes from the linewidths of isolated resonances. This concept is widely used in linear response theory to reconstruct time-resolved information directly from measured signals, even without the need for pump-probe-related schemes.

Here, we show how both techniques (pump–probe pulse geometry and linear response theory) can be combined to retrieve real-time information of quantum systems that explicitly include both strong and nonlinear interactions. The first (weak) pulse triggers a linear absorption response which is subsequently influenced by the second pulse, now allowing for nonlinear interactions in the strong-field regime. As we show, the changes introduced with this strong-field interaction can be directly reconstructed in real time from absorption spectra by using the Fourier transform (Fig.1). We demonstrate the validity of this approach with absorption measurements of helium. An attosecond extreme-ultraviolet (XUV) pulse triggers the excitation of the 2s2p doubly excited state, which decays freely until it is further perturbed by a strong femtosecond near-infrared (NIR) laser pulse after a fixed time delay (Fig.1a). This interaction creates nonlinear effects such as Rabi cycling and strong-field ionization, and results in measurable changes of the absorption line shape (Fig.1b,d). From this data alone, it is possible to reconstruct the real-time dipole response of the system (Fig.1c,e). This approach provides direct access to time-resolved dynamics in quantum systems without even varying the temporal delay between the pulses, and can be used to measure and control ultrafast processes in atoms and molecules, as well as to test fundamental time-dependent quantum theories of strong-field light–matter interaction.

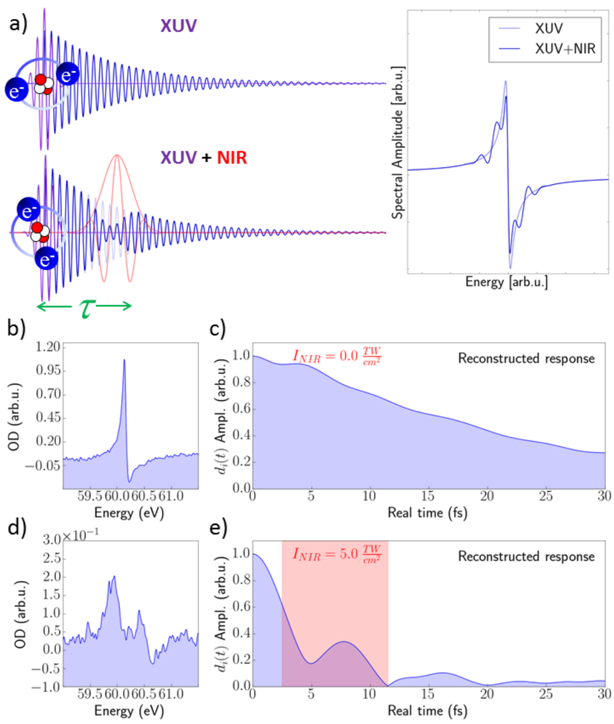

Figure 1: a) Experimental scheme: The NIR pulse modifies the response triggered by the short XUV pulse. The changes in the absorption line shape (right panel) reflect the changes in time-dependent dipole response (left panels). **b),c)** Recorded spectrum of the unperturbed response and its reconstruction. **d),e)** Recorded spectral line after the nonlinear interaction with the NIR pulse (red shaded area) together with its reconstruction. The interaction leads to Rabi-like oscillations of the response and a decrease in amplitude due to the strong field.

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