The time-domain response of autoionization and strong-field ionization

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When matter interacts with light, the lightweight electrons are first to notice. Due to their charge, the electric fields of light pull them out of their structural equilibrium, setting off (to first order) the so-called electric dipole response. The corresponding linear response function is directly accessible by spectroscopy, historically giving rise to the discovery of quantum mechanics, and nowadays a vital tool in almost any branch of science and technology.

Here in this talk, we will consider the generalization of response-function measurements to include strongly interacting, explicitly time-dependent systems such as atoms exposed to strong laser fields.

By a generalization of the Kramers-Kronig principle for pulsed/coherent light fields, it can be shown that a single absorption spectrum encodes the entire temporal response of a system even under explicitly time-dependent external driving.

The idea is experimentally applied to the case of optically coupled short-lived doubly excited states in Helium, driven into a competition between autoionization and strong-field ionization for increasing laser intensity. Agreement with an ab-initio simulation validates the method in Helium.

Finally, the approach is shown to be viable for complex systems, opening fascinating avenues for the exploration, understanding, and the control of larger atoms, (macro-/bio-)molecules in solution or other condensed-phase systems.

**Figure:** Illustration (not to scale) of the correspondence between various ionization spectra (top row), e.g. photoelectron or -absorption spectrum and their time-domain response functions (bottom row). While Photoionization produces a broad spectral feature and an instantaneous response, autoionization produces sharp Fano line shapes corresponding to an exponential temporal response (only amplitude is plotted). In this talk, it will be shown how the spectrum of a strong-field ionizing system looks like and how its temporal response, in amplitude and phase, can be fully retrieved from experimental data.