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Attosecond Transient Absorption Spectroscopy of doubly-excited states in helium

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Synopsis Theoretical calculations of the XUV attosecond transient absorption spectrum (ATAS) of helium in the doubly-excited state region reproduce recent high-precision measurements, reveal novel means of controlling the dynamics of transiently-bound electronic wavepackets in intense laser fields, and indicates a possible extension of 2D-spectroscopies to the XUV range.

Strong-field manipulation of autoionizing states is a crucial aspect of electronic quantum control. Attosecond transient absorption spectroscopy (ATAS) [1] is emerging as a prominent technique complementary to the technologies which detect charged photofragments to monitor and control transiently bound states. Compared to photofragment detection, ATAS provides higher energy resolution and is applicable to condensed phases [2] as well as to the gas phase, thus making it a good candidate for the investigation of ultrafast electron dynamics in chemically relevant samples.

Recent measurements of the ATA spectrum of helium dressed by a few-cycle visible pulse [3] provide evidence of novel ultrafast resonant phenomena, namely, two-photon Rabi oscillations between doubly excited states and the inversion of Fano profiles. Here we present the results of accurate ab-initio calculations that agree with these observations and in addition predict that (i) inversion of Fano profiles is actually periodic in the coupling laser intensity, (ii) the supposedly dark $2p^2$ ¹S state also appears in the spectrum, (iii) the Fourier transform of the ATAS can be construed in terms of an effective nondiagonal electrical susceptibility of the laser-dressed atom. The present results are obtained by direct solution of the time-dependent Schrödinger equation of the helium atom under the influence of external fields with an Arnoldi propagator on a B-spline close-coupling basis. Convergence with respect to close-coupling expansion, angular momentum and radial basis together with analytical continuation to infinite times of the field-free optical response of the system permit to achieve virtually exact results.

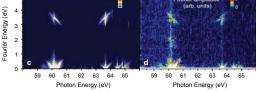


Figure 1. a) Theoretical and b) experimental ATAS as a function of the time delay between an attosecond XUV pump pulse and an intense ($\sim 3 \text{ TW/cm}^2$) VIS probe pulse. c-d): FT with respect to the time delay of the spectra shown in panels a-b).

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