Gauge-invariant absorption by coherent superposition

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Attosecond transient absorption spectroscopy (ATAS) is used to study electron dynamics with the aim of unravelling ultra-fast phenomena in atoms and molecules. There have been many investigations on ATAS between bound states [1], but few works have focused on the coupling of a prepared bound wave packet to both bound and continuum states [2]. A recent experimental investigation of the physical phenomena in this regime is presented in Ref. [3]. In order to disentangle the fundamental processes in this rich transient absorption regime, we establish a gauge invariant formulation of ATAS based on Yang's energy operator [4]. In the present work [5], perturbation theory is used to study the absorption of a hydrogen atom in an initial superposition state interacting with an attosecond pulse. Absorption is studied in both time and energy domains, resolved over the relative phase of the superposition. The model is validated by numerically solving the time-dependent SchrÃ¶dinger equation. The model allows for disentangling the absorption into fundamental processes of resonant and off-resonant nature, being symmetric and anti-symmetric over phase, respectively, see Fig. 1. The off-resonant contribution is found to be significant for states with dipole-allowed transitions to states of lower energy. This yields large regions of emission in the absorption profile of the $2p+3p$ superposition, not present in the $2s+3s$ case. Additionally, agreement with simulations of helium and neon atoms indicates the applicability of our model to more complex atoms. Using the model, we have fully disentangled the ATAS spectra of two-state superpositions in atoms, with coupling to continuum and off-resonant coupling to bound states.

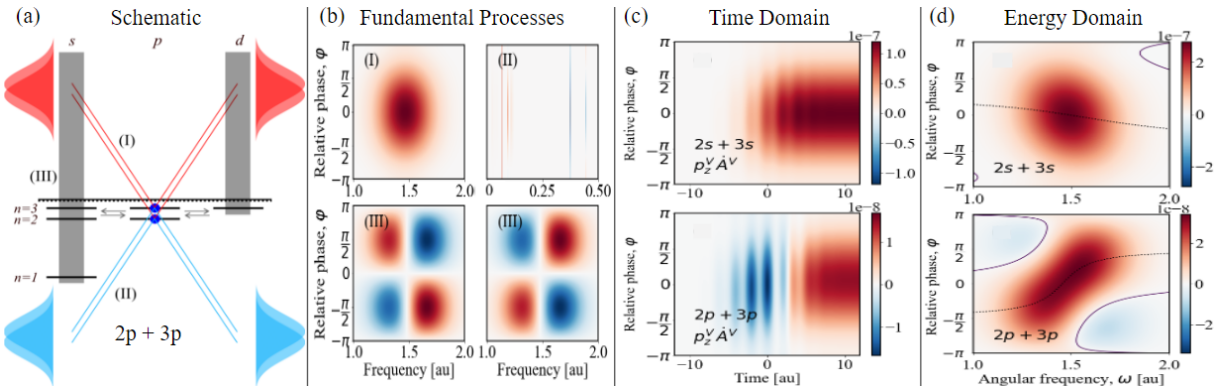

Figure 1: (a) Schematic of ATAS absorption processes: resonant continuum (I), resonant bound (II) and off-resonant (III). The energy-domain absorption of these fundamental processes are presented in (b), resolved over the relative superposition phase and Fourier frequency of the pulse, showing absorption in red and emission in blue. (c) and (d) show the absorption for the prepared superpositions $2s+3s$ and $2p+3p$ in the time and energy domain, respectively.

References

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