Modulation of Attosecond Beating in Resonant Two-Photon Ionization

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Synopsis We present a theoretical study of the photoelectron attosecond beating due to interference of two-photon transitions in the presence of autoionizing states. We show that, as a harmonic traverses a resonance, both the phase and frequency of the sideband beating significantly vary with photon energy. Furthermore, the beating between two resonant paths persists even when the pump and probe pulses do not overlap, thus permitting to reconstruct nonholographically coherent metastable wave packets. We characterize these phenomena with an analytical model that accounts for the effect of both intermediate and final resonances on two-photon processes.

Autoionizing states (AI) are hallmarks of electronic correlation, which shapes the reactivity of all many-body systems. Attosecond light sources provide a new means to investigate the role of such correlated states in atomic transitions, since one can map their evolution on a time scale smaller than their lifetime. The technique of reconstruction of attosecond beating in two-photon transitions (RABITT) [1], which has already been successfully employed to study resonant processes [2], is particularly indicated to this task. So far, however, only transitions through either electronic bound states or autoionizing vibronic states without any appreciable contribution from the intermediate continuum have been considered. The phase of the complex transition amplitude was thus compatible with a change of $\pi$ at the resonance energy.

In general, however, both the localized and continuum components of an intermediate AI contribute to the two-photon amplitude. We found that, when the two contributions are comparable, the phase exhibits a peaked excursion which returns to its original value, instead of a $\pi$ jump. Furthermore, we predict that, for finite pulses, the frequency of the RABITT beating itself is altered. Models normally used to interpret these experiments do not contemplate the presence of resonances, while full \textit{ab initio} calculations are either extremely demanding or outright unfeasible. For this reason, we developed a resonant analytical model which allows us to compute two-photon ionization spectra of attosecond pump-probe interferometric experiments, for arbitrary pulse sequences[3]. The model parameters can be retrieved either from experimental data or from selected \textit{ab initio} simulations. We used this procedure to investigate, with RABITT, the two-photon ionization of helium in the region of the doubly excited states of the atom, obtaining excellent agreement with virtually exact \textit{ab initio} simulations [4]. In particular, we show that both the frequency modulation and phase excursion of the transition through $sp^+_2$ and $sp^+_3$ ($^1P^o$) to $2p^2$ ($^1S^c$) are measurable.

Finally, the beating between the overlapping sidebands of the two intermediate resonances persist even when the pump and probe pulses are separated. From this beating it is possible to reconstruct the metastable wavepacket generated by the pump pulse.

References
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