Disentangling intracycle interferences in the photoelectron spectrum of argon using orthogonally polarized two-colour laser pulses

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Synopsis. Interferences in coincidence-photoelectron spectra from argon are controlled by orthogonally-polarized two-colour laser pulses. Previously unobserved intracycle interferences emitted during non-adjacent quartercycles within a single laser cycle are identified.

In recent years, the widespread availability of multi-dimensional photoelectron spectroscopy has created the opportunity to systematically studying the complex interference patterns inherent to multiphoton ionization of atoms and molecules. The dominant interference mechanism in multiphoton ionization leads to discrete peaks in the photoelectron spectrum (PES) that are spaced by the photon energy of the driving laser field. This mechanism is called above-threshold ionization (ATI) and has its origin in the coherence of ionization events repeating at every oscillation of the optical field [1].

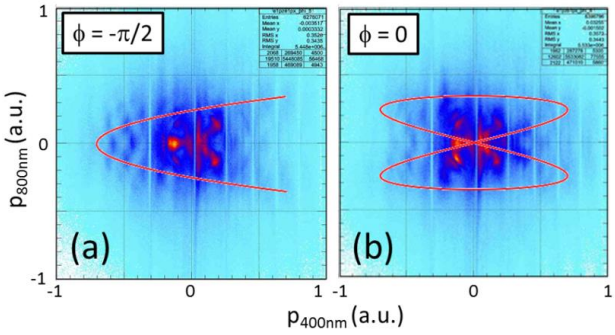

Figure 1. Ar^+ photoelectron spectra from OTC pulses for a phase delay of (a) $\phi=\pi/2$ and (b) $\phi=0\pi$. The vector potential for these OTC pulses is superimposed as a red line.

However, many more interference mechanisms contribute to the observed photoelectron spectrum. Specifically, photoelectrons emitted within one cycle of the optical field can also interfere on the detector. These interferences from photoelectrons originating in adjacent quarter-cycles of the optical field are also known as the temporal double slit (e.g., [1]). In addition, scattering of the photoelectron on the parent ion potential will give rise to holographic interferences [2], and laser induced electron diffraction [3].

Orthogonally-polarized two-colour (OTC) laser pulses [4] have proven to be a helpful tool to disentangle the quantum paths that cause the observed angular modulations in the PES [5,6]. Previously employed OTC pulses were generated by temporally overlapping an intense 800nm pulse with a weaker, perpendicularly polarized, 400nm pulse. Here, we invert the intensity ratio between long and short wave length and use an intensity ratio of approximately 1:20 for 800nm and 400nm, respectively.

We used COLTRIMS to record the three-dimensional photoelectron momentum distribution of argon. Fig. 1 shows examples of the two-dimensional PES in the polarization plane of two OTC pulses including an overlay of the associated vector potential. Using semiclassical modelling we identify a previously unobserved intracycle interference, where the interfering quantum pathways originate in non-adjacent quartercycles within a single cycle of the optical field.

References

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