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Phase Measurement of a Fano Resonance Using Tunable Attosecond Pulses

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Synopsis We study photoionization of argon atoms close to the $3s^23p^6 \rightarrow 3s^13p^64p$ Fano resonance using an attosecond pulse train and a weak infrared probe field. An interferometric technique combined with tunable attosecond pulses allows us to determine the phase of the photoionization amplitude as a function of photon energy. We interpret the experimental results using an analytical two-photon model based on the Fano formalism and obtain quantitative agreement.

Correlated electron dynamics induced by photoabsorption is a process of fundamental importance in nature. The development of table-top attosecond sources in the extreme ultraviolet (XUV) has opened up possibilities of accessing this dynamics directly in the time domain. In particular, correlation is responsible for the Auger decay, a resonant process that dramatically alters atomic photoionization spectra.

When a continuum state belonging to an open ionization channel interacts with a bound state belonging to a closed channel, autoionization may occur. Interference between the different ionization pathways leads to characteristic asymmetric Beutler-Fano profiles in the photoionization cross-section [1]. Studying this interaction in the time domain has been a major goal of attosecond science since the early days [2]. A method to do so is the so-called reconstruction of attosecond beating by interference of two-photon transitions (RABITT) technique [3], in which a train of XUV attosecond pump pulses is combined with a weak infrared (IR) probe with a variable time delay. The train, consisting of odd harmonics of the fundamental IR frequency, initiates the photoionization process. Further absorption or emission of an IR photon gives rise to sidebands. Due to the interference between two quantum paths, the sideband intensities oscillate as a function of the pump-probe delay. From the phase of these oscillations it is possible to obtain the phases of two-photon transition-amplitude and, in turn, of photoemission timing itself.

The RABITT technique has already been used to measure the effect of intermediate bound states on atomic two-photon transitions [4]. In this work, we present a study of the photoionization of argon using an interferometric method based on the RABITT technique, in the proximity of the $3s^23p^6 \rightarrow 3s^13p^64p$ autoionizing resonance, in which a tunable attosecond pulse train is used to initiate the photoionization process. As expected, changing the detuning between the resonance and the harmonic that is closest to it strongly affects the phases of the two adjacent sidebands. However, the phase profile clearly differs from the π jump observed for purely bound states. Our measurements are in quantitative agreement with the predictions of the analytical two-photon model in [5], which we extended to the multichannel case and parametrized with *ab initio* one-photon transition matrix elements obtained with a multi-configuration Hartree-Fock approach [6].

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