

Atto-second time-delay in single and double photoionization of noble gas atoms

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Synopsis We perform a systematic investigation of the photoelectron group delay in valence shell single ionization of Ne, Ar, Kr and Xe from their respective thresholds to photon energy of 200 eV. This allows us to highlight various important aspects of fundamental atomic physics that can be probed by attosecond time delay measurements and to address several controversies in recent experiments. We also use these data as an input to the time delay calculations of single-photon double ionization (PDI) of noble gas atoms. Various PDI channels are examined and estimates are given for associated time delays due to the shake-off and knock-out processes. These estimates are used to interpret recent measurements of PDI of Xe on the atto-second time scale

Time delay in atomic photoionization has become an active and rapidly expanding field of research following pioneering experiments on attosecond streaking [1] and two-photon sideband interference [2]. After correction for Coulomb-laser coupling in the former, or continuum-continuum transitions in the latter experiments, the photoelectron group delay, also known as the Wigner time delay, can be extracted from these measurements. This delay is related to the complex phase of the photoionization amplitude and represents a sensitive probe of ionization dynamics. In valence shell single photoionization of noble gas atoms, the time delay is strongly affected by inter-shell correlation whereupon absorption of the photon in the outer sub-shell results in ejection of the photoelectron from the inner sub-shell [3]. To account for this correlation, we employ the random phase approximation with exchange (RPAE) [4]. We validate our computational technique by making an extensive comparison between the calculated and experimental photoionization cross-sections. Based on this validation, we make specific predictions for the Wigner time delay and perform further comparison with available experimental time delay data. More generally, we demonstrate that the Wigner time delay contains important phase information that enables attosecond time delay measurements to reveal various fundamental aspects of atomic physics. This includes the logarithmic Coulomb singularity and the Levinson theorem which govern the photoelectron scattering phase in the field of the singly charged ion. The third factor is the phase jump of π near the Cooper's minimum which is smoothed by the inter-shell interaction.

Unlike single photoionization, which is only

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partially affected by many-electron correlation, the non-sequential single-photon double ionization (PDI) is driven entirely by this correlation. As such, it has long been regarded as an archetypal reaction to study correlated many-electron dynamics. Up until recently, these studies were limited to cross-section measurements. However, with the latest development of the two-photon sideband interference technique, combined with coincident electron detection, double photoionization of the valence shell of Xe can be resolved on the attosecond time scale [5]. Theoretically, the time delay in PDI can be related to the scattering phases of both photoelectrons and correlation correction. The latter is specific to various PDI mechanisms. We make numerical estimates of the correlated component of the time delay in PDI for the shake-off (SO) and knock-out (KO) mechanisms. In the KO mechanism, the primary photoelectron collides with one of the target electrons and knocks it out. In the SO process, orbital relaxation following the creation of a hole ejects the second electron. We demonstrate that with our estimates of various components of the time delay, the PDI process in Xe seems to be synchronized with the single electron photoionization. This was indeed observed in the recent measurement within the experimental uncertainty [5].

References

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