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

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We perform a systematic mapping of the photoelectron group delay in valence shell single ionization of Ne, Ar, Kr and Xe. This allows us to highlight various important aspects of fundamental atomic physics that can be probed by attosecond time delay measurements. 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 a rapidly expanding field of research following pioneering experiments on attosecond streaking [1] and two-photon sideband interference [2]. The photoelectron group delay, also known as the Wigner time delay, can be extracted from these measurements after some corrections. The group 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.

Unlike single photoionization, which is only partially affected by many-electron correlation, the non-sequential 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 untill recently, these studies were limited to cross-section measurements. However, with the latest development of the two-photon sideband interference technique, combined with conincident electron detection, double photoionization of the velence 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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