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Self-referencing time-domain measurements of femtosecond inner-shell dynamics

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X-ray radiation has been long used to address selectively atoms and to yield structural information with atomic precision. Recently, much effort has been put into extending those measurements to the fourth dimension, time, using ultrashort x-ray pulses to access the dynamics of the systems under study. The advent of X-ray Free Electron Lasers (XFEL) has quickly revolutionized the field of time resolved x-ray techniques. The availability of tunable pulses ranging from the soft to the hard x-ray region, and lasting only few tens of femtoseconds is enabling access to unprecedented temporal resolution. In addition, the huge increase in pulse brightness compared to 3rd generation synchrotron sources has opened the field of intense x-ray interaction with matter, where much is still unknown. Temporal resolution limits arise from the pulse durations of both pump and probe pulses, group velocity mismatch, as well as the timing jitter that rises inevitably between the two independent sources. A significant effort has been put into gaining the ability to measure the timing jitter for every shot, in order to be able to tag the shots depending on the relative delay, and perform post sorting analysis of the data. A precision around 25 fs (FWHM) has been demonstrated, already a considerable improvement over the uncorrected jitter around 400-500 fs (FWHM). However, if one hopes to follow the very fast dynamics of the electronic configuration, often crucial to understand physical and chemical properties, another breakthrough is needed. Importing the laser streaking techniques developed by the attophysics community, one can greatly improve the temporal resolution of pump probe experiments where electrons are collected to follow the processes. Laser streaking has been extensively used to obtain time domain information, using an intense laser field to modify the final energy of a photoelectron created during the x-ray pulse. In certain conditions, there is a one-to-one relationship between the final energy and the time of ionization during a half optical cycle of the intense laser (i.e the streaking ramp), allowing for a direct reconstruction of the temporal profile from the measured energy spectrum. Our scheme consists in measuring simultaneously the streaking of photoelectrons (PE) created by direct photoionization during the x-ray pulse itself and the streaking of other electrons corresponding to the process under study. Efficient collection of the PE streaked spectrum allows for a shot-to-shot positioning of the x-ray pulse against the streaking ramp, so that any streaked electron of interest can then be positioned relative to both the streaking ramp and the x-ray pulse. Demonstration of such a technique was done at LCLS at SLAC, attempting to measure for the first time, in a time resolved fashion, the Auger decay lifetime of light atoms (Ne and Carbon). Using x-ray pulses as short as a few femtoseconds long, and a streaking laser operating in the infrared around 17μm, femtosecond resolution should be possible in the determination of the lifetimes.