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X-ray chronoscopy for investigation of attosecond dynamics in matter

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The X-ray free electron lasers (XFELs) [1] are nowadays used to study the structure and dynamics in matter with unprecedented temporal and spatial resolutions. However, some fundamental electron processes in the matter are too fast to be followed, even with the shortest available XFEL pulses. In this work we report on a new experimental approach to study sub-femtosecond processes in matter. Based on the X-ray chronoscopy concept [2], it explores the time distribution of X-ray pulses before and after interaction with a sample, relaxing the strict requirement of the ultra short pulse duration but needing precise time structure measurement. The pulse time structure can be measured using the state-of-the-art terahertz streaking cameras at XFELs [3] arranged in the camera-sample-camera sequence.

In this presentation, the behavior of X-ray pulse time distribution will be shown for two scenarios: (1) the nonlinear process of saturable absorption at ultra-short time scales [4] and (2) an optical pump-X-ray probe scenario [5], using a simulation based on a rate equation model. In study (1), time distribution of a 7 fs-short (FWHM) pulse of 7130 eV-photons was simulated as it is before and after interaction with 20 μm-thick iron foil. The high photon flux lead to deviation of the transmitted pulse time envelope from that of the incident pulse, including loss of the original Gaussian shape and shift in time phase (i.e. shift of the pulse temporal center of mass). Other effects will be reported as well. The results tend to confirm that X-ray-induced dynamics leading to the target X-ray transparency can be probed through measurement of X-ray pulses time structure. In study (2), ZnSe quantum dots were subjected to interaction with intense optical pump pulses with near-UV photon energy (3.1 eV) and 5 fs (FWHM) pulse duration to excite valence band electrons of the material, and were probed at a delay with an X-ray pulse. The average energy of the X-ray probe pulse was set to 0.1 μJ, the spot on the sample to 100 μm^2 and pulse duration to 20 fs (FWHM). The results showed sensitivity of the transmitted X-ray pulse time distribution to the excited state lifetime and to the pump-probe delay which will be presented in detail.

Part of this work has been published [4] and attracted the EurekAlert!'s attention [6].

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