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Development of Two-Color Sub-Femtosecond Pump/Probe Techniques with X-ray Free-Electron Lasers

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The natural time scale of quantum mechanical motion of electrons in molecules is usually on the order of hundreds of attoseconds. Probing time-dependent dynamics with atomic-site specificity on such timescales requires the generation of soft X-ray attosecond pulses pairs with variable delay and synchronization down to the sub-femtosecond level. We report the generation of GW-level attosecond pump/probe pulse pairs with tunable sub-femtosecond delays at the Linac Coherent Light Source (LCLS). The attosecond 365 eV pump pulse is first generated via the Enhanced Self-Amplified Spontaneous Emission (ESASE) method, then the attosecond 730 eV probe pulse is produced by re-amplifying the electron beam microbunching after the magnetic chicane. Due to the harmonic amplification, the minimal delay between pump-probe pulse pairs (limited by slippage between the light field and the electron bunch) can be shorter than 0.5 femtoseconds. We use the angular streaking technique to measure temporal delays between pump/probe pulse pairs at multiple beamline configurations. When the delay chicane is turned off, the averaged delay is increased by ~ 150 attoseconds by adding one undulator module for probe pulses. Long delays can be set up by turning the delay chicane on. These experimental results agree with start-to-end XFEL simulations. Looking toward future experiments, our sub-femtosecond pump/probe technique can be applied to observe electronic charge dynamics in molecular systems.

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Footnotes

- Zholents, A. A. Physical Review Special Topics-Accelerators and Beams 8.4 (2005): 040701.
- Duris, J., et al. Nature Photonics 14.1 (2020): 30-36.
- Li, S., et al. Optics Express 26.4 (2018): 4531-4547.

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Yes

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