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Spectrotemporal shaping of attosecond x-ray free-electron laser pulses

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The development of attosecond methods at free-electron lasers has led to new possibilities in the probing and control of electronic dynamics in molecules. Beyond simple observation of ultrafast processes, one of the longstanding goals of atomic and molecular physics is control of the electronic wavefunction on attosecond timescales. This implies a need to go beyond impulsive excitation with isolated pulses: more general spectrotemporal pulse shaping is demanded. We present a method to shape the spectrotemporal characteristics of attosecond XFEL pulses in a two-stage scheme. First, an isolated attosecond pulse is generated using now-state of the art methods. That pulse then interacts with a fresh part of the electron beam in a second stage where control of the undulator taper leads to direct shaping of the output pulse. We highlight several example shaping options: pulse pairs with controllable time and color separation, isolated fs-scale pulses with controllable linear chirp, and trains of attosecond pulses with tunable spacing. We highlight one atomic physics application –to use pulse pairs to control and probe the decay of core-excited electronic wavepackets.

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