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Beyond the Frontiers of Time-Resolved Spectroscopy[1]

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Time-resolved spectroscopy experiments typically require the measurement of at least two ("pump" and "probe") interactions of a field (e.g. a laser pulse) with the spectroscopic target system (e.g. atom, molecule) at variable but known temporal delays. It is often assumed that the shortest dynamics measurable with such techniques is on the order of the pulse duration of the pump and probe events. In this talk, it will be shown that attosecond electron wavefunction beating can, in principle, be resolved by employing a nonlinear interferometry concept with phase-stabilized femtosecond pulses that does not require attosecond pulses for pumping nor probing. The perfectly coherent and reproducible electric fields of the pump and probe pulses thus seem the ultimate technical goal to achieve the highest temporal resolution in science. By contrast, however, it will be shown that statistically varying (colored-noise) partially coherent pulses typically produced at free-electron lasers (FELs) can be beneficial in resolving dynamics beyond their average pulse duration. These findings may carry general implications for the future development of time-resolved spectroscopy.

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