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Attosecond Spectral Interferometry and Quantum Control of Electron Dynamics

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The direct time-resolved observation of electron dynamics in atoms and molecules is a key goal of attosecond physics. To this end, several pioneering spectroscopic techniques have been conceived and experimentally demonstrated by a number of research groups in the past, including the optical streak camera in the gas phase and on surfaces of solids, tunneling spectroscopy, as well as recollision tomography and related methods. All these schemes, however, require strong laser fields for probing the dynamics and thus potentially distort the "natural" field-free energy-level structure and dynamic evolution of the system to be studied. In this talk, some novel spectroscopic routes and first experiments are presented that allow the measurement of coherent electron dynamics in field-free environments. The key idea is the usage of interferometric methods, e.g. realized by two interferometrically stable and time-delayed attosecond pulses. Combined with coincidence detection techniques (reaction microscopes, COLTRIMS), these interferometric methods will enable the attosecond time-resolved measurement of multi-electron correlation in atoms and molecules. In addition, experimental results for coherent control of electron dynamics will be presented. Also here, we focus on the low-intensity regime, where we directly control electronic quantum-path interference instead of quasi-classical trajectories that typically prevail at higher intensities. Multi-photon N/N+1 absorption-pathway interference is controlled by varying the carrier–envelope phase (CEP) of few-cycle light fields in a generalized Brumer-Shapiro control scheme. The results yield important conclusions for laser control with shaped ultrashort pulses, proving the necessity of CEP stabilization to exert full control over quantum systems.