Ultrafast spectroscopy and imaging of molecules with classical, quantum, and noisy x-ray pulses

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We demonstrate how stimulated Raman detection of an X-ray probe may be used to monitor the phase and dynamics of the valence electronic state wavepacket created by e.g. photoexcitation, photoionization and Auger processes. Conical intersections (CoIn) dominate the pathways and outcomes of virtually all photophysical and photochemical molecular processes. Short X-ray pulses can directly detect the passage through a CoIn with a high temporal and spectral sensitivity. Applications will also be made for X ray sum frequency generation, and detecting molecular chirality.

Quantum light opens up new avenues for spectroscopy by utilizing parameters of the quantum state of light as novel control knobs and through the variation of photon statistics by coupling to matter. Nonlinear optical signals induced by quantized light fields and entangled photon pairs will be presented. Quantum phase-sensitive diffraction and imaging using entangled photons is presented. A novel quantum diffraction-based far-field imaging technique whereby one photon of an entangled pair is diffracted of a sample and detected in coincidence with its twin is proposed. When a molecule interacts with an external field, the phase information is imprinted in the state of the field in a detectable way. A Schmidt decomposition of the state of the field and the resulting diffraction signal can be used for image enhancement by reweighing the Schmidt modes. Imaging with weak quantum fields is made possible, avoiding damage to delicate biological samples.

Time- and frequency-resolved ultrafast diffraction of noisy X-ray pulses is presented. A frequency-dependent stochastic phase can be used as a frequency marker. Separation of different frequency contributions to the diffraction pattern can be achieved. Fine-tuned correlation functions can control specific charge density contributions. These results are applicable for inherently noisy sources such as SASE free electron lasers, as well as to coherent sources with intentionally added randomness.

References

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