Ultrafast HHG Spectroscopy in Classical and Quantum Light Regimes

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Recently a novel method for monitoring electronic coherences using ultrafast spectroscopy in a high harmonic regime has been developed. This method is based on the time-domain high-order harmonic spectroscopy, where a coherent superposition of the electronic states is first prepared by the strong optical laser pulse using a three-step mechanism introduced by Lewenstein and Corkum. The coherent dynamics can then be probed by the higher-order harmonics generated by the delayed probe pulse. In the previous meeting, we reported various nonlinear optical signals in a high harmonic regime, including multi-harmonic wave-mixing and pump-probe signals. We now extend these methods for the studies of conical intersections in complex molecular systems undergoing nonadiabatic dynamics by providing a high resolution for tracking the diabatic potential surface crossing and studies of geometric phases in the wavepacket dynamics. Furthermore, the nature of multi-wave mixing in a high harmonic regime allows for modifying the statistics of light and gives rise to quantum squeezing between the higher harmonics suitable for higher signal-to-noise ratio measurements of electronic properties in the multi-eV range.

References

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