Overcoming the classical limits of ultrafast spectroscopy with entangled photons
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Optical spectroscopy is a powerful tool to investigate the microscopic mechanisms underlying different physical, chemical and biological processes. Ultrafast spectroscopy has played a pivotal role in unveiling the early snapshots of photophysical events with femtosecond time resolution. In its standard implementations, the system under study is subjected to a series of pulses, whose frequencies and time delays are the control knobs. Such setups follow a semiclassical description of light–matter interaction, in which light pulses are treated as electromagnetic waves neglecting their quantum nature.

Many current research actions, are focussing on the possible exploitation of properties of single quantum objects, like state superposition and entanglement, to achieve the so called “quantum advantage”, like the capability to perform computations with quantum-bits which cannot be done with classical computers.

Recently, a growing body of theoretical studies has proposed to use quantum states of light, such as squeezed states or entangled photon pairs (EPPs), to enhance also ultrafast optical spectroscopy. Quantum light offers several advantages for spectroscopy, such as enhancing signal-to-noise ratio, providing novel control knobs (such as energy, time and polarization entanglement of the EPPs) for designing experiments or even generating completely new signals with respect to classical light. Despite the plethora of theoretical proposals, experimental demonstrations of nonlinear spectroscopy with quantum light are very limited.

This project, that is granted by PRIN2022 (grant number 2022HL9PRP) aims to replace the classical probes in ultrafast spectroscopy with EPPs and demonstrate the "quantum advantage" in terms of sensitivity and novel spectroscopic signals for the study of molecular systems and aggregates. The project has the following scientific objectives:

- to develop the theoretical tools required to model ultrafast spectroscopy with quantum light for realistic molecular systems.
- to use EPPs in order to probe exciton-exciton interactions in molecular dimers/oligomers, both in the presence and in the absence of a background excitonic population.
- to compare numerical simulations with experimental results and finally provide the first demonstration of the quantum advantage in ultrafast spectroscopy with quantum light.