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Time-resolved photon antibunching in single molecules

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Abstract

Single molecule spectroscopy involves measuring objects one at a time, enabling conclusions to be drawn free from ensemble averaging effects or complex interdependencies. Measuring the light emitted by single molecules or nanoscopic objects also allows signatures of quantum behaviour such as photon antibunching to be observed. Conventional antibunching measurements record the intensity-weighted average of the photon statistics over the excited state lifetime, giving a single correlation curve. This allows limited insight into any fast (ps-ns) evolution of the system, thus information on how states form, move and interact is typically lost.

Here I will talk about our recent work using modern photon counting hardware to time-resolve photon statistics, down to times as short as ~50 ps, observing how the antibunching signature evolves during the molecule's excited state lifetime. This allows the fingerprints of state coupling and losses to be identified. The concept will be first explored with DNA origami, where a fixed number of, and position, of emitters is defined. I will then discuss these measurements in more complex nanoscopic aggregates, before applying the techniques to observations in single organic semiconducting polymer chains. Finally, I will talk about our latest research developing two-colour time-resolved photon antibunching measurements, where coupling between energetically different states can be explored. This work has relevance to the photophysics of organic semiconducting materials, but also to the development of quantum optical measurements that can assess coherence and the quantum fidelity of excited states in polyatomic systems within the domain of quantum information science.