Department of Physics
Condensed Matter Physics
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UNIVERSITY OF OXFORD

CONDENSED MATTER SPECIAL SEMINAR

Friday 7th December at 10.30am

"Bright Triplet State, Long Exciton Dephasing and Superfluorescent Emission from Cesium Lead Halide Perovskite Nanocrystals"

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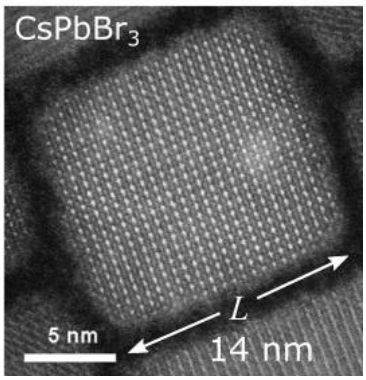
The lowest energy exciton in organic materials is a poorly emitting triplet state according to Hund's rules. For inorganic semiconductors, similar rules predict an analog of this triplet known as the dark exciton. This state releases photons slowly, therefore materials that disobey these rules have been sought. However, despite considerable experimental and theoretical efforts, no inorganic semiconductors have been identified in which the lowest exciton is bright. The emission of fully inorganic cesium lead halide perovskite-type nanocrystals is narrowband and tunable over a wide energy range with photoluminescence quantum yields of up to 90%[1] at room temperature. Experiments on single quantum dots reveal photon emission rates ~20 and ~1000 times higher compared to any other conventional semiconductor nanocrystals at room and cryogenic temperatures, respectively. By temperature dependent quantum yield and intensity-decay time correlation measurements we investigate the nature of of this exceptionally fast and almost blinking free emission[2,3].



To access the excitonic coherence properties, we perform degenerate four-wave mixing on $CsPbBr_2Cl$ nanocrystal films. We obtain a dephasing time of about 27 ps at low temperature which is almost an order of magnitude larger compared to any other colloidal quantum dots with similar oscillator strength. In addition, we observe signatures of a coherent exciton-phonon coupling.

Our experimental findings of extraordinary high oscillator strength together with an exceptionally long dephasing time suggest that lead halide perovskite nanocrystals are a prime candidate for the observation of coherent excitonic phenomena. We use arrays of densly packed nanocrystals, so called superlattices, built by solvent-drying-induced spontaneous self-assembly allowing us to observe superfluorescent (SF) emission. SF is characterized by a dynamical red-shifted emission with more than twenty-fold accelerated radiative decay, extension of first-order coherence time by more than a factor of 4, photon bunching and Burnham-Chiao ringing behaviour at high excitation density[4].

Our comprehensive set of experimental results is the first demonstration of a bright triplet state, collective optical behaviour and extendet coherent states within colloidal nanocrystals opening new avenues towards ultra-bright and muli-photon light sources.

References:
[1] Protesescu *et al.*, Nano Lett. **15**, 3692–3696 (2015)
[2] Becker *et al.*, Nature, **553**, 187-193 (2018)
[3] Rainò *et al.*, ACS Nano **10**, 2485–2490 (2016)
[4] Rainò *et al.*, Nature (2018), just online doi:10.1038/s41586-018-0683-0

Host: Prof Donal Bradley

Audrey Wood Seminar Room, Clarendon Laboratory