Photonic and excitonic coupling in nanocrystal superstructures

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Assembled colloidal quantum dots (QDs) hold great promise as optically active films in novel optoelectronic devices. The wide tunability, low-temperature processability, and high efficiency of QDs make them very beneficial for a wide range of device applications – from LEDs to lasers to photodetectors and photovoltaic devices. The working principles of these devices rely on efficient charge carrier transport and, in the case of photovoltaic devices, on light absorption. Both however are limited for quantum dots and their aggregates.

A recently developed emulsion-templated assembly technique yields QD supercrystals of extraordinary structural quality, expected to feature `artificial solids', with properties such as band-like transport, beneficial for devices. Here, we show that supercrystals of CdSe and CsPbBr perovskite QDs simultaneously enhance the absorption efficiency and inter-dot coupling of QDs (Fig. 1). By comparing optical measurements with Mie theory, we demonstrate that supercrystals can exhibit resonant optical properties resulting in absorption efficiencies greater than 1 in the visible range. By performing ligand exchange, we increase the excitonic coupling, and obtain signatures of band-like states: Ultrafast transient absorption (TA) spectroscopy shows that biexcitons transit from a bound to a free state and the density of states and carrier cooling rates increase due to the increased interdot coupling. These results reveal a new state of matter with properties architecturally designed to feature simultaneously photonically and electronically coupled QDs for future optoelectronic devices.