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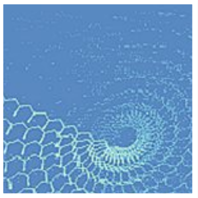

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Superfluorescence from Nanocrystal Superlattices

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Quantum dots have become essential building blocks in various optoelectronic devices. For example, quantum dots are used in television displays as highly efficient, narrowband green and red emitters. Furthermore, they are a useful source of non-classical light, *e.g.* allowing single photon emission, which might play a pivotal role for the development of future quantum technologies. Recently, $APbX_3$ (A = FA, MA, Cs; X = I, Br, Cl) perovskite nanocrystals[1] (NCs) have emerged as a new class of quantum emitters. Their narrow-band, blinking-free and high oscillator strength emission[2] had been assigned to bright triplet exciton states.[3]

Using the facile solvent-evaporation method, perovskite NCs spontaneously form highly-ordered three-dimensional (3D) superlattices,[4] a long-dreamed bottom-up approach to artificial materials with tailor-made properties. In our most recent work,[5] we have found that these cuboidal self-assembled superlattices exhibit superfluorescent (SF) emission at cryogenic temperatures. SF is a cooperative radiation effect resulting from the spontaneous buildup of coherence between initially independent dipoles due to quantum fluctuations, resulting in an effective, single giant emitting dipole (see Fig. 1).

An example of superlattices and a typical photoluminescence (PL) spectrum obtained at cryogenic temperatures are presented in Fig. 2. The spectrum features two emission bands: the higher energy band (peak at 525 nm) represents the emission of uncoupled NCs. The additional red-shifted emission band centered at 535 nm is associated with the cooperative emission of coherently coupled NCs. This assignment is supported by time-resolved experiments (Fig. 2C), which show a much shorter and non-exponential decay for coupled NCs, as originally predicted by Dicke for a superradiant emission.[6] Further evidence is found by exploring the statistics of emitted photons (Fig. 2D). Typical coherent light, as from a laser, shows a random distribution (Poissonian) of photon arrival times on a detector, while a single NC exhibits photon anti-bunching (sub-Poissonian distribution). In contrast, the cooperative emission from coupled NCs leads to coherent multi-photon emission bursts: highly correlated multiple photons are then emitted within a short time interval, giving rise to a bunching peak (Fig. 2D).

These results motivate future use of perovskite NCs in ultrafast light emitting diodes and novel entangled multi-photon quantum light sources for quantum cryptography and quantum sensing.

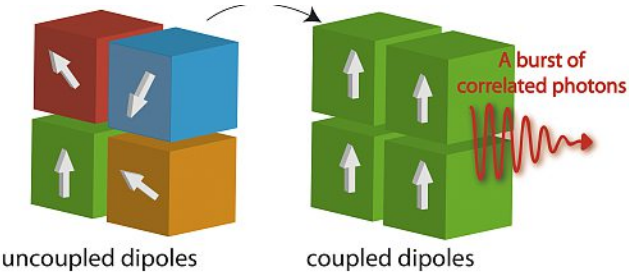

Fig. 1. Schematic of the build-up of superfluorescence in NC superlattices.

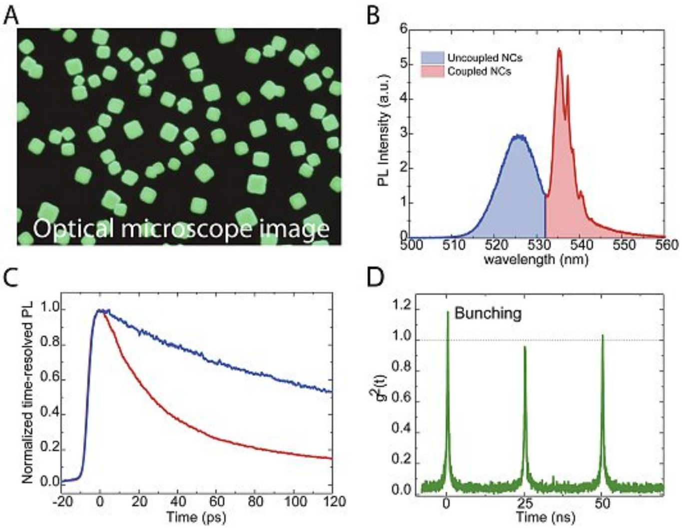

Fig. 2. A) Superlattices under the microscope (UV illumination). B) The low-temperature PL spectrum of a single $CsPbBr_3$ NC superlattice. C) Time-resolved PL traced for uncoupled (blue) and coupled (red) NCs. D) Second order correlation measurements showing bunching behavior (displayed here for pulsed excitation).

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[1] L. Protesescu, S. Yakunin, M. I. Bodnarchuk, F. Krieg, R. Caputo, C. H. Hendon, R. X. Yang, A. Walsh, M. V. Kovalenko, *Nanoletters* **2015**, *15*, 3692.

[2] G. Rainò, G. Nedelcu, L. Protesescu, M. I. Bodnarchuk, M. V. Kovalenko, R. F. Mahrt, T. Stoferle, *ACS Nano* **2016**, *10*, 2485.

[3] M. A. Becker, R. Vaxenburg, G. Nedelcu, P. C. Sercel, A. Shabaev, M. J. Mehl, J. G. Michopoulos, S. G: Lambrakos, N. Bernstein, J. L. Lyons, T. Stöferle, R. F. Marht, M. V. Kovalenko, D. J. Norris, G. Raino, A. L. Efros, *Nature* **2018**, *553*, 189.

[4] M. V. Kovalenko, L. Protesescu, M. I. Bodnarchuk, *Science* **2017**, *358*, 745.

[5] G. Rainò, M. A. Becker, M. I. Bodnarchuk, R. F: Mahrt, M. V. Kovalenko, T. Stöferle, *Nature* **2018**, *563*, 671.

[6] R. H. Dicke, *Phys. Rev.* **1954**, *93*, 99.

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