Superfluorescence from Lead Halide Perovskite Quantum Dot Superlattices

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An ensemble of emitters can behave significantly different from its individual constituents when interacting coherently via a common light field. After excitation, collective coupling gives rise to an intriguing many-body quantum phenomenon, resulting in short, intense bursts of light: so-called superfluorescence[1]. Because it requires a fine balance of interactions between the emitters and their decoupling from the environment, together with close identity of the individual emitters, superfluorescence has thus far been observed only in a limited number of very specific systems, such as certain atomic and molecular gases and a few select solid-state systems[2-7]. With colloidal nanocrystals, however, which are increasingly recognized as bright, practically suited photonic sources for optoelectronics[8,9], the generation of superfluorescent light was precluded by inhomogeneous emission broadening, low oscillator strength, and fast exciton dephasing. Using caesium lead halide (CsPbX$_3$, X = Cl, Br) perovskite nanocrystals[10-13] that are self-organized into highly ordered three-dimensional superlattices allows us to observe key signatures of superfluorescence: dynamically red-shifted emission with more than twenty-fold accelerated radiative decay, extension of the first-order coherence time by more than a factor of four,

photon bunching, and delayed emission pulses with Burnham–Chiao ringing behaviour[14] at high excitation density. These mesoscopically extended coherent states can be employed to boost opto-electronic device performances[15] and enable entangled multi-photon quantum light sources[16,17].

Spontaneous emission (SE) of photons, such as fluorescence commonly used in displays or lighting, occurs due to coupling excited two-level systems (TLS) to the vacuum modes of the electromagnetic field, effectively stimulated by its zero-point fluctuations. In 1954, R. H. Dicke predicted[18] that an ensemble of N identical TLS confined in a volume smaller than $\sim\lambda^3$ (λ is the corresponding emission wavelength of the TLS) can exhibit coherent and cooperative spontaneous emission. This so-called superradiant emission results from the coherent coupling between individual TLS through the common vacuum modes, effectively leading to a single giant emitting dipole from all participating TLS. Superradiant emission has been observed in distinctly different physical systems, such as molecular aggregates and crystals[19], nitrogen vacancy centres in diamond[20] and epitaxially grown quantum dots[21]. In the case when the excited TLS are initially fully uncorrelated, the coherence can be established only through spontaneously triggered correlations due to quantum fluctuations rather than by coherent excitation. When this occurs, a so-called superfluorescent (SF) pulse is emitted[1] (Figure 1, illustrated for the present study). Both superradiant emission and coherent SF bursts are characterized by an accelerated radiative decay time $\tau_{SF}\sim\tau_{SE}/N$, where the exponential decay time τ_{SE} of the uncoupled TLS is shortened by the number of coupled emitters N. In addition, SF exhibits the following fundamental signatures, which magnitudes are also dependent on the excitation density: (i) a delay or build-up time $\tau_D\sim\ln(N)/N$ during which the emitters couple and phase-synchronize to each other, and which corresponds to the time delay between the excitation and onset of the cooperative emission (Figure 1) and (ii) coherent Rabi-type oscillations in the time domain due to the strong light–matter interaction, known as Burnham–Chiao ringing[14,22].

SF was first observed in a dense gas of hydrogen fluoride[2], followed by a limited number of solid-state systems, such as CuCl nanocrystals (NCs) formed in a NaCl matrix[4], KCl crystals doped with peroxide anions (O_2^-) (ref.[3]), and some select semiconductor crystals (ZnTe and InGaAs/GaAs multi-quantum wells)[5,6]. Practical implementation of such an enhanced radiative property is a persistent challenge. Besides stringent requirements for the

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emissive material (e.g., high oscillator strength, small inhomogeneous line-broadening, small exciton dephasing), equally important are structural, optical and device engineerability. Colloidal semiconductor NCs, also known as colloidal quantum dots (QDs), could fill this gap as they are structurally and optically versatile, and highly suited for the entire visible spectral range. While they are actively pursued for photonic applications[8,9,23], they have not been reported to exhibit SF.

Here, we use colloidal NCs of caesium lead halide perovskites ($CsPbX_3$, X = Cl, Br) that can be synthesized with narrow size dispersion and are known to exhibit moderate quantum confinement effects, resulting in narrow-band emission combined with exceptionally large oscillator strength from a bright triplet state[10,11,24]. In order to foster cooperative behaviour, we employ structurally well-defined, long-range ordered, and densely packed arrays of such NCs, known as superlattices, produced by means of solvent-drying-induced spontaneous assembly[25-28]. Similarly, regular arrays of II-VI semiconductor NCs have been used to obtain collective effects in the electronic domain, *i.e.,* band-like transport[15]. Figure 2a outlines the superlattice formation (see also Methods), using a solution of highly monodispersed $CsPbBr_3$ NCs with a mean size of 9.5 nm and standard size-deviation of less than 5% (Extended Data Figure 1). In the self-assembly process, cubic individual superlattice domains are formed (*i.e.*, supercrystals), each consisting of up to several millions of NCs. Optical microscopy (Figure 2c) reveals superlattices with a lateral size of up to 5 μm, randomly distributed in a uniform film on a 5×7 mm sample (Figure 2d). Transmission electron microscopy confirms that highly ordered superlattices consist of well-separated individual NCs (Figure 2e and Extended Data Figure 2). More details of the self-assembly process are reported in the Methods section.

Figure 3a displays the photoluminescence (PL) spectrum of a single $CsPbBr_3$ superlattice (excited at 3.06 eV) exhibiting two emission peaks. This and all other optical measurements were performed at a temperature of 6 K in vacuum or Helium atmosphere (see Methods for details). The high-energy emission peak coincides with the centre energy of the disordered dense film of $CsPbBr_3$ NCs (glassy state) and is therefore assigned to non-coupled QDs. In addition, a narrow, red-shifted emission peak appears in superlattices, which we assign to the emission of coupled QDs, which is best fitted with a Lorentzian (full width at half maximum, $FWHM_{coupled}$ = 11 meV). The peaks of the uncoupled QDs in a superlattice and

95 in the glassy films are best fitted with a Gaussian, as expected for disordered ensembles. The

96 width of the uncoupled QDs ($FWHM_{uncoupled}$ = 55 meV) is slightly broader than the one of the

97 amorphous film ($FWHM_{amorphous}$ = 35 meV), which can be explained assuming that the more

98 "identical QDs" within the superlattice are now forming the peak of coupled QDs while the

99 remaining uncoupled ones appear more disordered than the inhomogeneous energy

100 distribution of the primary QD material. We can exclude that the red-shifted feature, which is

101 at ~70 meV lower energy than the uncoupled QD emission, originates from the emission from

102 trions, bi-excitons or multi-excitons, because their energy shifts reportedly are 10–20 meV

103 (refs. [11,12]), and these would be observable in the disordered ensemble, too. The number and

104 interaction strength of coupled QDs determine the magnitude of the energetic shift. Statistics

105 from 10 superlattices from different samples give an average static red-shift of (64±6) meV,

106 average $FWHM_{coupled}$ = (15±4) meV and average $FWHM_{uncoupled}$ = (49±21) meV. In most

107 superlattices, we observe a sub-structure in this red-shifted emission band, which we attribute

108 to the presence of several, slightly different independent domains within the same individual

109 superlattice.

110   A central feature of the cooperative emission is the modification of the radiative

111 lifetime[18], as demonstrated experimentally with several quantum emitters[6,20,21]. In time-

112 resolved PL decay measurements, at a very low excitation fluence (5 nJ/cm^2), we do not

113 observe a significant modification of the decay of the coupled QD emission compared to the

114 uncoupled QD emission (Figure 3b inset). The absence of accelerated emission at vanishing

115 excitation density and the presence of the red-shifted feature also in PL excitation scans

116 (Extended Data Figure 3) corroborates that the static ~70 meV red-shift originates from

117 incoherent coupling of the QDs in the ground state, similar to various molecular aggregates[19].

118 Already at a slightly higher excitation density, we observe an accelerated PL decay of the

119 coupled QD emission peak in comparison to the PL decay of uncoupled QDs with $1/e$ decay

120 times of $\tau_{SF} = 148\,\mathrm{ps}$ and $\tau_{QD} = 400\,\mathrm{ps}$, respectively, for an excitation density of 500 nJ/cm^2

121 per pulse (Figure 3b). In contrast to the predominantly mono-exponential decay of the

122 uncoupled QDs, this SF emission decay is approximated well by a stretched exponential[29]

123 (see Methods section), because the number of excited coupled emitters, and therefore the

124 emission acceleration, varies during the decay. Furthermore, in contrast to the uncoupled

125 QDs, the SF decay time is strongly dependent on excitation power (inset Figure 3b) because

126 it scales with the coupling strength among the QDs, given by the intensity in the common

127 light-field that effectively corresponds to a change in the number of coherently coupled QDs.

128 When the spectrally and temporally integrated emission is fitted with a power law, we obtain

129 an exponent of 1 (Extended Data Figure 4), indicating that excitation density-dependent non-

130 radiative decay channels (e.g. Auger recombination) are absent. Notably, no threshold

131 behaviour as for amplified spontaneous emission (ASE) is observed.

132 The cooperative emission process strongly influences the coherence of the emitted

133 light. First-order correlation measurements of each of the two emission peaks by means of a

134 Michelson interferometer allow us to monitor the interference pattern and therefore the phase

135 coherence time (Figure 4a). The emission band of the uncoupled QDs exhibits 38 fs

136 coherence time, best fitted with a Gaussian decay (Figure 4a, upper graph), typical of

137 incoherent (thermal) light sources. The emission from the coherently coupled QDs (Figure 4a,

138 lower graph) exhibits a much longer coherence time with an exponential decay of 140 fs. For

139 some superlattices, a Gaussian decay is observed (Extended Data Figure 5a), which might

140 be attributed to number fluctuations within the coherent SF state[30].

141 Second-order coherence of the emitted light is evinced by the statistics of the photon

142 arrival time on a detector[31]. Typical coherent light, as from a laser, shows a random

143 distribution (Poissonian) of photon arrival times, while a single TLS exhibits photon

144 antibunching (sub-Poissonian distribution). In contrast, the cooperative emission from coupled

145 QDs leads to coherent multi-photon emission bursts. Figure 4 reports the second-order

146 correlation function, $g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2}$ for both PL emission bands, where $I(t)$ is the signal

147 intensity at time t. For the uncoupled QD emission (Figure 4b, upper graph), a flat $g^{(2)}(\tau) = 1$

148 is observed because the experimental temporal resolution (40 ps) is insufficient to resolve the

149 expected thermal bunching. The SF emission band, however, shows pronounced photon

150 bunching (Figure 4b, lower graph) because the coherent coupling leads to the correlated

151 emission of multiple photons within a short time interval. Photon bunching is only observable

152 in superlattices with single or a few SF domains, *i.e.*, where no sub-structure is visible in the

153 red-shifted emission band, because spectrally overlapping uncorrelated aggregated domains

154 within the same superlattice reduce the bunching peak's visibility, as predicted by theory[32].

155 Yet, it is a robust effect that is observed with pulsed excitation and for mixed-halide

156   (CsPbBr$_2$Cl, emitting at higher energies) QD superlattices, too (see Extended Data Figure 5b

157   and 6b, respectively). Remarkably, some superlattices with supposedly well-isolated

158   coherently coupled QDs exhibit $g^{(2)}(\tau) > 2$ (inset Figure 4), similar to superthermal

159   emission[31]. The exponential decay time of the second-order correlation is of the order of the

160   radiative decay of the SF emission for low excitation densities ($\tau_{g^{(2)}} = 224\,\text{ps}$).

161   Very distinct characteristics of SF emission concern the time evolution of the emitted

162   light under strong driving conditions. Figure 5a shows a streak camera image acquired at an

163   excitation density of 600 µJ/cm^2, where in addition to a drastically shortened radiative decay,

164   a finite rise time and subsequent oscillations of the emission are observed. Quantitative

165   analysis on spectrally integrated PL decay traces for various excitation power densities is

166   shown in Figure 5b (for details see Methods). As a function of the excitation density, the

167   decay time shortens to 14 ps (Figure 5c, upper panel). From this decay shortening, which is

168   an order of magnitude stronger than reported for the collective emission from other QD

169   systems[4,21], we can estimate an average number of coherently coupled QDs to be $N \sim 28$.

170   This is only an effective value and a conservative estimate, because the energetic disorder of

171   the QD emission energies (FWHM$_{coupled}$ = 11 meV) still exceeds significantly the emission

172   peak width of individual QDs (typically FWHM ~ 1 meV, ref. [11]) and thereby effectively

173   reduces the SF coupling[33]. The SF emission experiences a dynamical red-shift of up to 15

174   meV due to renormalization of the emission energy from the coherent coupling[7], which

175   decreases in the course of the decay as the number of excitations reduces (Extended Data

176   Figure 7). The peak intensity increases super-linearly over three orders of magnitude (Figure

177   5c, middle panel), according to a power-law dependence with an exponent of $\alpha = 1.5 \pm 0.1$,

178   deviating from the theoretically expected value of $\alpha = 2$ (ref. [7]), presumably due to saturation

179   effects[5]. Nevertheless, no significant quenching effects of the emission for high excitation

180   powers were found, verifying that the decay remains essentially radiative (Extended Data

181   Figure 4d). Furthermore, a shortening of the SF delay time (τ_D), after which the photon burst

182   is emitted, is observed (Figure 5c, bottom panel). This characteristic of SF is a consequence

183   of the time it takes for phase-locking the individual dipoles and scales with the number N of

184   excited coupled QDs according to $\tau_D \sim \frac{\log(N)}{N}$ (see Methods section).

As SF crucially depends on low decoherence and low inhomogeneous spread, it should be noted that SF coupling is strongly affected by the environment around the QDs (number of free ligands), the superlattice assembly, and by the quality of the QDs themselves. Thus, while a large fraction of the superlattices displays a red-shifted peak from the coupled QD emission, the amount of photon-bunching and Burnham–Chiao ringing varied from superlattice to superlattice. However, experiments employing different batches of NCs and superlattice assemblies of $CsPbBr_3$ and $CsPbBr_2Cl$ NCs (see Extended Data Figures 6-9) were consistently reproducible, but further optimization of the synthesis and assembly is likely to improve the yield of SF domains. It is important to note that experiments on control samples with diluted, uncoupled QDs under similar excitation conditions do not show any of these signatures of SF (Extended Data Figure 10), proving that it is a genuine multi-particle effect.

Our measurements reveal that coherent SF coupling can be achieved in long-range ordered self-assembled superlattices of fully inorganic $CsPbX_3$ perovskite NCs, resulting in strong emission bursts. Colloidal NCs and their assemblies have proven to be excellent building blocks for a large variety of opto-electronic devices, and these cooperative effects now allow modification of the opto-electronic properties beyond what is possible on the individual QD level with chemical engineering approaches. This opens up new opportunities for high-brightness and multi-photon quantum light sources, and could enable the exploitation of cooperative effects for long-range quantum transport and ultra-narrow tuneable lasers.

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Main Figure Legends

273 **Figure 1 | Schematic of the build-up process of SF.** An initially uncorrelated ensemble of TLS (randomly
274 oriented green arrows) is excited by a light pulse (blue arrow). After time τ_D their phases are synchronized
275 (aligned green arrows) such that they cooperatively emit a SF light pulse (red arrow). Grey cubes represent long-
276 range ordered self-assembled superlattices.

277

278 **Figure 2 | Formation of CsPbX$_3$ (X = Cl, Br) NC superlattices by drying-mediated self-assembly. a**,
279 Illustration of the assembly process. **b**, High-resolution scanning transmission electron microscopy image
280 (HAADF–STEM) of a single CsPbBr$_3$ NC. **c**, Optical microscope image and **d**, photograph (under UV light) of a
281 layer of micron-sized, three-dimensional, cubic-shaped NC superlattices. **e**, HAADF–STEM image of a single
282 superlattice of CsPbBr$_3$ NCs. The cubic shape of the individual perovskite NC building blocks is translated into
283 the symmetry of the superlattice (simple cubic packing). The inset shows a zoom-in where the individual NCs are
284 visible.

285

286 **Figure 3 | Optical properties of CsPbBr$_3$ QD superlattices. a**, PL spectrum of a single CsPbBr$_3$ superlattice
287 (black solid line). The high-energy band is assigned to the emission of uncoupled QDs. The low-energy band is
288 the result of the emission of coupled QDs and is not present in glassy films of NCs (green solid line). The shaded
289 areas are fits to the data (see main text). **b**, Time-resolved PL decay of the two emission bands at 500 nJ/cm^2
290 excitation density after applying suitable spectral filters to separate the two components. With increasing
291 excitation density, the decay from the coherently coupled QDs is significantly faster than from the uncoupled
292 ones. The inset shows the power-dependence of the 1/e-decay times of both components.

293

294 **Figure 4 | First- and second-order coherence properties of CsPbBr$_3$ QD superlattices. a**, First-order
295 correlation of the two emission bands as obtained from the interference fringe visibility using a Michelson
296 interferometer. The high-energy band of the uncoupled QDs has a very short phase coherence time (<40 fs,
297 upper graph), whereas the red-shifted band from the coupled QDs is characterized by much longer phase
298 coherence (140 fs, lower graph). The solid lines are fits to the data (see text). The inset shows an example of the
299 real space interferogram. **b**, Second-order correlation function, $g^{(2)}(\tau)$, obtained with a Hanbury–Brown and
300 Twiss setup in start–stop configuration. For the high-energy band (upper graph), a flat profile with $g^{(2)}(\tau) = 1$ is
301 observed. The red-shifted emission band (lower graph) from the SF emission displays a pronounced bunching
302 peak, characteristic of the correlated emission during a photon burst. The data are fitted to the function $g^{(2)}(\tau) =
303 1 + A \cdot exp(-(|\tau - \tau_0|/\tau_c)$ (solid lines). The inset shows an example of superbunching with $g^{(2)}(0) > 2$ from a
304 single superlattice.

305

306 **Figure 5 | Burnham–Chiao ringing behaviour of CsPbBr$_3$ QD superlattices. a**, Streak camera image of SF
307 dynamics obtained with high excitation density of 600 µJ/cm^2. **b**, Extracted time-resolved emission intensity
308 traces for five different excitation powers. Solid red lines are weighted best-fits to a model that employs a bi-
309 exponential decay function with damped oscillations. **c**, *Top:* Effective SF decay (blue circles) as a function of
310 the excitation power density fitted according to the SF model (solid blue line). *Middle:* Red circles represent the
311 peak SF emission intensity that increases super-linearly with excitation power, corresponding to a power-law
312 dependence with an exponent $\alpha = 1.5 \pm 0.1$ (solid dark-red line). *Bottom:* The extracted delay time τ_D (green

313 circles) decreases at high excitation power due to the increased interaction among the emitters. The green solid
314 line is the best fit according the model described in the Methods section. The error bars represent the
315 parameters' fit uncertainty.

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317

METHODS

Synthesis of CsPbBr₃ nanocrystals. In a 25 ml three-necked flask, $PbBr_2$ (69 mg, 0.188 mmol, Aldrich, 99%) was suspended in octadecene (5 ml), dried at 100°C for 30 min, and mixed with oleic acid (0.5 ml, vacuum-dried at 100°C) and oleylamine (0.5 ml vacuum-dried at 100°C). When $PbBr_2$ was dissolved, the reaction mixture was heated up to 180°C and preheated caesium oleate in octadecene (0.4 ml, 0.125 M) was injected. The reaction mixture was cooled immediately with an ice bath to room temperature.

Synthesis of CsPbBr₂Cl nanocrystals. In a 25 ml three-necked flask, $PbBr_2$ (45 mg, 0.12 mmol, Aldrich, 99%), $PbCl_2$ (18 mg, 0.064 mmol, ABCR) and 1 ml trioctylphosphine (Strem, 97%) was suspended in octadecene (5 ml), dried at 100°C for 30 min, and mixed with oleic acid (0.5 ml, vacuum-dried at 100°C) and oleylamine (0.5 ml vacuum-dried at 100°C). When $PbCl_2$ and $PbBr_2$ were dissolved, the reaction mixture was heated up to 180°C and preheated caesium oleate in octadecene (0.4 ml, 0.125 M) was injected. The reaction mixture was cooled immediately with an ice bath to room temperature.

Purification and size-selection of CsPbX₃ (X = Cl, Br) nanocrystals. A critical factor for self-assembly of cubic-shaped $CsPbX_3$ NCs is to start with an initially high level of monodispersity. The crude solution was centrifuged at 12100 rpm for 5 min, following which the supernatant was discarded, and the precipitate was dissolved in 300 µl hexane. The hexane solution was centrifuged again and the precipitate was discarded. The supernatant was diluted two times and used for further purification. Subsequently, two methods of purification of the NCs were applied: (a) 50 µl hexane, 0.6 µl oleic acid, and 0.6 µl oleylamine were added to 50 µl NCs in hexane. The colloid was destabilized by adding 50 µl acetone, followed by centrifuging and dispersing the NCs in 300 µl toluene. This solution was used further for the preparation of the 3D-superlattices. (b) 50 µl hexane and 100 µl toluene were added to 50 µl NCs in hexane. The colloid was destabilized by adding 50 µl acetonitrile, followed by centrifuging and dispersing the NCs in 300 µl toluene. This solution was used further for the preparation of the 3D-superlattices.

Preparation of 3D-superlattices. $CsPbX_3$ NC superlattices were prepared on glass or on 5×7 mm silicon substrates. Shortly before the self-assembly process, the silicon substrate was dipped into 4% solution of HF in water for 1 min, followed by washing with water. In a typical assembly process, the substrate was placed in a $10 \times 10 \times 10$ mm Teflon well and 10 µl of purified NCs in toluene were spread onto the substrate. The well was covered with a glass

slide and the toluene was then allowed to evaporate slowly. 3D-superlattices of CsPbBr$_3$ NCs were formed upon complete evaporation of the toluene. Typical lateral dimensions of individual superlattices ranged from 1 to 10 μm wherein some of them arrange into clusters of several superlattices and others remain spatially well-isolated so that PL measurements can be performed on an individual superlattice.More intense purification or greater polydispersity of NCs led to disordered or 2D assemblies (glassy films). Furthermore, the formation of NC superlattices can serve to further narrow the size distribution and shape uniformity within the ensemble (with smaller or larger NCs being repelled from the NC domain), especially in the case of simple cubic packing of cubes, which is particularly intolerant to size and shape variations.

Optical spectroscopy. All measurements were performed at cryogenic temperatures (6 K). For PL, time-resolved PL, and second-order photon-correlation measurements on single superlattices, the sample was mounted in an evacuated liquid-helium flow cryostat on *xyz* positioning stages and excited with a fibre-coupled excitation laser at an energy of 3.06 eV, either in continuous wave mode or pulsed mode with 40 MHz repetition rate (pulse duration 50 ps). The excitation laser was filtered with a short-pass filter and directed towards the long-working distance 100× microscope objective (numerical aperture $NA = 0.7$) by a dichroic beam splitter, resulting in a nearly Gaussian-shaped excitation spot with $1/e^2$ radius of 1.4 μm. The emission was collected via the same microscope objective and filtered using a tuneable bandpass filter. For PL measurements, the collected light was then dispersed by a 300 lines/mm grating inside a 750 mm monochromator and detected by an EMCCD camera. For measurements of the PL decay, we filtered the emission with a tuneable band-pass (FWHM = 15 nm) and recorded the decay with an avalanche photo diode single photon detector with a time resolution of $30 \, \mathrm{ps}$ connected to a time-correlated single-photon-counting system. The photon correlation was recorded using a similar setup with two detectors in a Hanbury–Brown–Twiss setup configuration.

To record streak camera images and first-order coherence measurements, we excited the sample, which was mounted in an exchange-gas cryostat at 6 K, with a frequency-doubled regenerative amplifier seeded with a mode-locked Ti:sapphire laser with a pulse duration of 100–200 fs and a repetition rate of 1 kHz at 3.1 eV. For both excitation and detection, we used an 80 mm lens ($NA = 0.013$ after iris), resulting in an excitation spot area of 20×40 μm.

381 The recorded PL was dispersed by a grating with 150 lines/mm in a 300 mm spectrograph

382 and detected with a streak camera with a nominal time resolution of 2 ps and an instrument

383 response function FWHM of 4 ps (see Extended Data Figure 10). First-order coherence

384 measurements were performed using a Michelson interferometer. Here a non-polarizing

385 beam splitter is used to split and recombine the light in the two interferometer arms, with one

386 arm including a retroreflector on a delay stage with 100 nm step resolution. A tuneable band-

387 pass filter is applied to select the emission from either the coupled or the uncoupled QDs. The

388 interferogram was recorded as real-space images of the recombined and focused detection

389 beams on a camera.

390 **Optical properties of superfluorescence, superradiance, and subradiance.** As shown in

391 Figure 3b, we observed that the PL decay of the SF state is initially very fast and cannot be

392 described with a single exponential because the decay rate is dependent on the number of

393 excited TLS, $\Gamma(N) \sim N$, and therefore decreases during the decay. Consequently, the SF

394 decay rate should converge towards the decay rate of the uncoupled nanocrystals. However,

395 we observe that the SF decay trace crosses the bi-exponential PL decay of the uncoupled

396 QDs after 97% of the photons are emitted due to long decay components. These long decay

397 components might originate from coupled QDs where the individual dipoles are out of phase

398 and interfere destructively, known as subradiance (SBR)[29,34]. In ensembles with

399 inhomogeneously broadened PL, SF and subradiant states can coexist, and we find a good

400 agreement of the predicted excited state population with the measured PL decay[35].

401 An out-of-phase coupling amongst the QDs is expected to result in a higher photon energy of

402 the subradiant state compared to the SF state. In Extended Data Figure 7 and Extended Data

403 Figure 9, we provide an analysis of the dynamical energy shift observed at high excitation

404 power density for $CsPbBr_3$ and $CsPbBr_2Cl$ QD superlattices, respectively. Examples of

405 emission spectra at different times are reported in Extended Data Figure 7a and Extended

406 Data Figure 9a for the respective QD halide compositions. In Extended Data Figure 7b and

407 Extended Data Figure 9b, we plot the fitted centre photon energy of time-sliced PL spectra (2

408 ps bin) as a function of the fitted peak area (*i.e.*, the time-dependent emission intensity), as

409 obtained from excitation power-dependent streak camera images, again for both, $CsPbBr_3$

410 and $CsPbBr_2Cl$ QD superlattices. This effectively shows the energetic shift of the SF state as

411 a function of its occupation, with the different curves representing different initial excitation

412 powers. The green arrows indicate the time sequence of the individual analysed spectral

traces. By increasing the excitation power, we observe that the initial dynamical red-shift is the largest for the highest excitation power, as is expected from its relationship to the number of excited coupled QDs. Hence, when the number of excited coupled QDs decreases during the decay process, the emission energy blue-shifts to higher energy, as can be seen in Extended Data Figures 7c and 9c where the fitted centre photon energy is plotted as a function of time. We observe the most pronounced energetic blue-shift for the highest excitation power, resulting in a final emission with a photon energy that has been boosted incrementally more in comparison to the blue-shift for low excitation power, which is another indication of the presence of subradiant states that emit at higher energies. For high excitation power, the SF state becomes depopulated much faster since more QDs are coupled simultaneously. Then, at long timescales after the initial decay, the percentage of subradiant states becomes dominant, resulting in a blue-shift of the PL emission.

Superfluorescence fit model. SF decay traces as in Figure 3b cannot be fitted well with mono- or bi-exponential functions because the decay rate is proportional to the number of excited coupled QDs $\Gamma(N) \sim N$, which also decays over time. Furthermore, the resulting characteristic decay neither exactly follows stretched-exponential nor a power-law dependence[36], whereas the PL decay of the uncoupled QDs is well described by a bi-exponential behaviour, where the initial fast decay $\tau_{QD} = 349.8 \pm 0.4\,\text{ps}$ accounts for over 96% the total emitted photons. Nevertheless, we found that the best approximate fit to the SF decay trace is the Kohlrausch stretched-exponential decay model $\frac{f(t)}{f(0)} = \exp[-(\Gamma_{\text{stretched}} \cdot t)^{\beta}]$, where $\Gamma_{\text{stretched}}$ is the average decay time and $\beta \in [0,1]$ is the stretch parameter, which represents the distribution of decay rates[37]. Using this model to fit the SF decay curve, we obtain an average decay time $\tau_{\text{stretched}} = 40.4 \pm 0.5\,\text{ps}$ and a stretch parameter $\beta = 0.457 \pm 0.002$.

At a high excitation density, as shown in Figure 5b for $CsPbBr_3$ QD superlattices and in Extended Data Figure 8b for $CsPbBr_2Cl$ QD superlattices, we observe oscillations in the decay. To model the SF decay with this characteristic ringing behaviour, we used a decay model consisting of a bi-exponential decay that is multiplied by a damped oscillating term $1 + B \cdot \exp(-\gamma_{\text{Damp}}t) \cdot \cos(\omega t + \phi_0)$. Furthermore, for the rising edge of the emitted pulse we

442 take into account a Gaussian rise term $\sim \exp[-\left(\frac{t-\tau_D}{\tau_{rise}}\right)^2]$, such that the complete fit function is

443 given by (ref.[38]):

$$f(t) = \sum_{n=1,2} A_n \cdot \exp\left(\frac{\tau_{rise}^2}{4\tau_n^2} - \frac{t-\tau_D}{\tau_n}\right)$$

$$\cdot \left[\frac{1}{2}\left(1 + B \cdot \exp\left(-\gamma_{Damp}(t-\tau_D)\right) \cdot \cos(\omega(t-\tau_D) + \phi_0)\right)\right.$$

$$\left. \cdot \left[1 + \text{erf}\left(\frac{t-\tau_D}{\tau_{rise}} - \frac{\tau_{rise}}{2\tau_n}\right)\right]\right]$$

444

445 Here, A_n are the amplitudes of the exponential decay with the corresponding decay time

446 constants, τ_n. Both the fast decay time and the long decay time component (Extended Data

447 Figure 7d for CsPbBr$_3$ QD superlattices and Extended Data Figure 9d for CsPbBr$_2$Cl QD

448 superlattices) decrease upon increasing the excitation density, whereas the rise time,

449 $\tau_{rise} = 2.4 \pm 0.3$ ps for CsPbBr$_3$ QD superlattices ($\tau_{rise} = 3.4 \pm 1.0$ ps for CsPbBr$_2$Cl QD

450 superlattices), stays approximately constant (probably clamped by the time resolution of the

451 setup). In the upper panel of Figure 5c, we plot the power-dependent effective decay time

452 $\tau_{SF} = \frac{A_1\tau_1 + A_2\tau_2}{A_1 + A_2}$ for CsPbBr$_3$ QD superlattices, where τ_1, τ_2 are the decay times of the bi-

453 exponential fit and A_1, A_2 the corresponding amplitudes, which was fitted with $\tau_{SF}(P) = \frac{\tau_{QD}}{\zeta \cdot P + 1} +$

454 y_0, with a fixed value of $\tau_{QD} = 400$ ps, obtained from the time-resolved PL measurements of

455 uncoupled QDs and an additional offset y_0 to account for effects like the finite time resolution.

456 We obtain good agreement with the expected behaviour ($\tau_{SF} \sim \tau_{QD}/N$) for a value $\zeta_{CsPbBr3} =$

457 $0.29 \pm 0.04 \frac{cm^2}{\mu J}$. In the lower panel of Figure 5c, we plot the delay time τ_D for the CsPbBr$_3$ QD

458 superlattices as a function of the excitation power. In our analysis, the delay time is composed

459 of the actual delay time due to the SF build-up and a systematic, constant time-offset because

460 the absolute arrival time of the excitation pulse (which has a different wavelength than the

461 emission) at the sample cannot be measured reliably at the required precision from the streak

462 camera data. We observe a decrease in τ_D of ~6 ps when increasing the excitation density by

463 almost 2 orders of magnitude. We have fitted this behaviour with $\tau_D = y_{offset} + A \cdot \ln(\zeta P_{Exc} +$

464 $1)/(\zeta P_{Exc} + 1)$ because we assume that $\tau_D \sim \ln(N)/N$ and that the number of excited coupled

15

emitters $N \sim \alpha P_{\text{Exc}} + 1$ is proportional to the excitation power. Herein, we use a fixed value $\zeta_{CsPbBr3} = 0.29 \pm 0.04 \frac{cm^2}{\mu J}$, which we obtained from the fit of the effective decay in the upper panel of Figure 5c. The resulting fit agrees very well with the data. To obtain the absolute time delay, we subtracted the constant offset y_{offset} of the time-delay fit from the time-delay data points. SF occurs when $\sqrt{\tau_{\text{SF}}\tau_{\text{D}}} < T_2^*$, where T_2^* is the exciton pure dephasing time, whereas $\sqrt{\tau_{\text{ASE}}\tau_{\text{D}}} > T_2^*$ signifies the amplified spontaneous emission (ASE) regime, when τ_{ASE} is the decay time (ref. [33]). Considering that the coherence time $T_2 < T_2^*$ extracted from the full-width at half-maximum of single QDs[11] is of the order of $T_2 = 6.6 \, \text{ps}$, our measurements reveal a fast decay of ~14 ps and a delay time of < 1 ps which satisfies the criterion for the appearance of SF.

A similar analysis was performed on CsPbBr$_2$Cl QD superlattices as shown in Extended Data Figure 8. In the upper panel of Extended Data Figure 8c we plot power-dependent effective decay time and fit the data with the same model as described above, using $\tau_{QD} = 250 \, \text{ps}$ and we obtain a value $\zeta_{CsPbBr2Cl} = 0.08 \pm 0.01 \frac{cm^2}{\mu J}$. The peak intensity of the decay curves as a function of the excitation density is shown in the middle panel of the Extended Data Figure 8c, which increases super-linearly with a power-law dependence with an exponent $\alpha_{CsPbBr2Cl} = 1.3 \pm 0.1$. Also the delay time decreases as a function of the excitation density as displayed in the lower panel of Extended Data Figure 8c and fit the data. Again, we fit the data using the same formula for τ_D as described above with a fixed value $\zeta_{CsPbBr2Cl} = 0.08 \pm 0.01 \frac{cm^2}{\mu J}$ and obtain good agreement.

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Author contributions

The work originated from ongoing interactions between G.R., M.V.K, R.F.M., and T.S. M.I.B. prepared the samples and performed their structural characterization. G.R., M.A.B., and T.S. performed the optical experiments, interpreted the data with input from R.F.M. G.R. and M.A.B. wrote the manuscript with input from all the co-authors. R.F.M., M.V.K., and T.S. supervised the work.

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Data availability. The data that support the findings of this study are available from the corresponding authors upon reasonable request.

The authors declare no competing interests.

Extended Data Figure Legends

Extended Data Figure 1 | Quantitative analysis of CsPbBr$_3$ NC size distribution. a, Low-resolution TEM image of the NC material which is used to prepare the superlattices. **b**, Histogram of NC sizes (of >100 NCs) as obtained from TEM image analysis. The solid line is a fit with a normal distribution.

Extended Data Figure 2 | HAADF-STEM image of a single superlattice of CsPbBr$_3$ NCs. Individual NCs (bright spots in the image) are well-resolved.

Extended Data Figure 3 | PL excitation measurement of CsPbBr$_3$ QD superlattices. Using a weak, tuneable excitation source, we plot the PL intensity (black circles) obtained at 2.30 eV photon energy as a function of excitation photon energy. The shaded areas are Gaussian peak fits. This shows that the coupled QD feature is also present in the absorption spectrum, in addition to the uncoupled QDs and more energetically higher states.

Extended Data Figure 4 | Power dependent PL properties of CsPbBr$_3$ QD superlattices. a, Colour-coded PL emission in the low-power excitation regime, shown for increasing excitation fluence of 10 nJ/cm^2 (light green), 60 nJ/cm^2 (light blue), 150 nJ/cm^2 (yellow), 310 nJ/cm^2 (dark green) and 600 nJ/cm^2 (dark blue). **b**, PL intensity integrated over the spectral emission range of the uncoupled QDs (blue circles) and coupled QDs (red circles) in a log-log plot and the total emitted intensity (yellow circles). Fits to the data reveal a perfectly linear behaviour, as represented by a fitted power-law exponent m of 1. **c**, Colour-coded PL emission in the high-power excitation regime, shown for increasing excitation fluence of 330 µJ/cm^2 (light green), 1270 µJ/cm^2 (light blue), 2130 µJ/cm^2 (yellow), 3470 µJ/cm^2 (dark green) and 6330 µJ/cm^2 (dark blue). **d**, PL intensity integrated over the spectral emission range of the uncoupled QDs (blue) and coupled QDs (red) in a log-log plot and the total emitted intensity (yellow). Fits to the data reveal a power-law behaviour with a linear increase for the SF emission, a slightly sublinear increase for the uncoupled QDs and a less sublinear increase for the total emitted intensity.

Extended Data Figure 5 | Gaussian first-order coherence decay and photon bunching in pulsed excitation of CsPbBr$_3$ QD superlattices. a, First-order coherence of the coupled QD emission extracted from the fringe contrast of the interferograms as a function of differential delay time between the arms of a Michelson interferometer, revealing a mixture of Gaussian (Kubo) and exponential decay (for some of the superlattices). **b**, Second-order photon correlation measurement of the coupled QD emission from a single superlattice showing photon bunching at zero delay under pulsed excitation with a 40 MHz repetition rate.

Extended Data Figure 6 | SF in CsPbBr$_2$Cl QD superlattices. a, Band-pass filtered PL spectra of uncoupled QDs (blue) and coupled QD emission (red) of CsPbBr$_2$Cl perovskite superlattices. **b**, Second-order photon correlation measurement of uncoupled QDs (top panel, $g^{(2)}(\tau) = 1$) showing flat correlation function, and the coupled QD emission peak band (lower panel, $g^{(2)}(0) = 1.15$) showing photon bunching.

Extended Data Figure 7 | SF decay and dynamic red-shift of CsPbBr$_3$ QD superlattices. a, PL spectra (integrated over 2 ps time window) at different time delays in a semi-log scale. **b**, The PL spectra are fitted to a single Gaussian peak function, and the fitted peak amplitude as a function of the emission energy is plotted for various excitation densities. Green arrows indicate the time evolution of the emission peak. The black dashed line denotes the mean energy at the lowest excitation density, and the grey shaded area is the peak's FWHM. **c**, Fitted peak centre energy as a function of time for various excitation densities. **d**, Fast and slow PL decay time

572   components τ_1 and τ_2 of the SF bi-exponential fit model as a function of excitation density in a semi-log plot. The
573   error bars represent the parameters' fit uncertainty.

574   **Extended Data Figure 8 | Burnham–Chiao ringing behaviour in CsPbBr$_2$Cl QD superlattices. a**, Streak
575   camera image of SF dynamics obtained with high excitation density of 1600 µJ/cm^2. **b**, Extracted time-resolved
576   emission intensity traces for three different excitation powers. Solid lines are best-fits to a model that employs a
577   bi-exponential decay function with damped oscillations. **c**, *Top:* Effective SF decay (blue circles) as a function of
578   the excitation power density fitted according to the SF model (solid blue line). *Middle:* Red circles represent the
579   peak SF emission intensity that increases super-linearly with excitation power, corresponding to a power-law
580   dependence with an exponent $\alpha = 1.3 \pm 0.1$ (solid dark-red line). *Bottom:* The extracted delay time τ_D (green
581   circles) decreases at high excitation power due to the increased interaction among the emitters. The green solid
582   line is the best fit according the model described in the Methods section. The error bars represent the
583   parameters' fit uncertainty.

584

585   **Extended Data Figure 9 | SF decay and dynamic red-shift of CsPbBr$_2$Cl QD superlattices. a**, PL spectra
586   (integrated over 2 ps time window) at different time delays in a semi-log scale. **b**, The PL spectra are fitted to a
587   single Gaussian peak function, and the fitted peak amplitude as a function of the emission energy is plotted for
588   various excitation densities. Green arrows indicate the time evolution of the emission peak. The black dashed
589   line denotes the mean energy at the lowest excitation density, and the grey shaded area is the peak's FWHM.
590   **c**, Fitted peak centre energy as a function of time for various excitation densities. **d**, Fast and slow PL decay time
591   components τ_1 and τ_2 of the SF bi-exponential fit model as a function of excitation density. The error bars
592   represent the parameters' fit uncertainty.

593

594   **Extended Data Figure 10 | PL decay from a control sample of diluted CsPbBr$_3$ QD. a**, Streak camera
595   measurement of a control sample prepared by spin-coating a low concentration of CsPbBr$_3$ QD dispersed in
596   polystyrene such that no QD coupling is realized. The excitation conditions were comparable to Figure 5, using
597   an excitation density of 1600 µJ cm^{-2}. The comparably long decay requires that on the streak camera a longer
598   time range with lower temporal resolution compared to the measurement in Figure 5 has to be used. The
599   absence of dynamic red-shift, accelerated decay and ringing proves that the observed SF features cannot be
600   explained by single QD physics but are a multi-particle effect. **b**, Time-resolved PL trace (blue) obtained from
601   this streak data. For direct comparison, the equivalent data from a superlattice sample showing SF (Figure 5b,
602   1200 µJ cm^{-2}) is shown (red). Inset: Instrument response function of the streak camera. Using the same
603   instrument settings as for the superlattices measurement, the temporal response of the streak camera using
604   scattered light from a 100 fs laser pulse at 400 nm gives a Gaussian-shaped instrument response with a FWHM
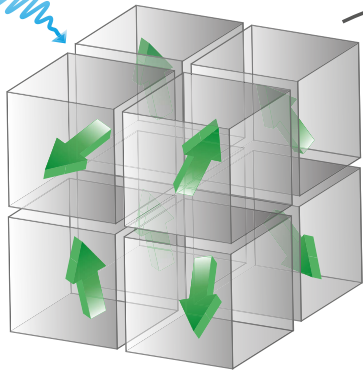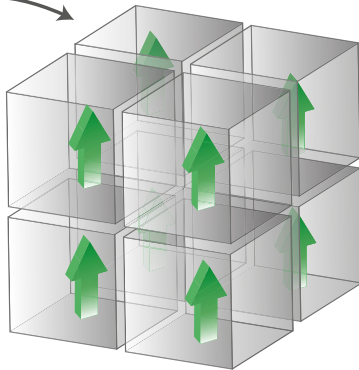605   of 4.3 ps (Gaussian fit is shown as red solid line).

606

Excitation

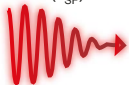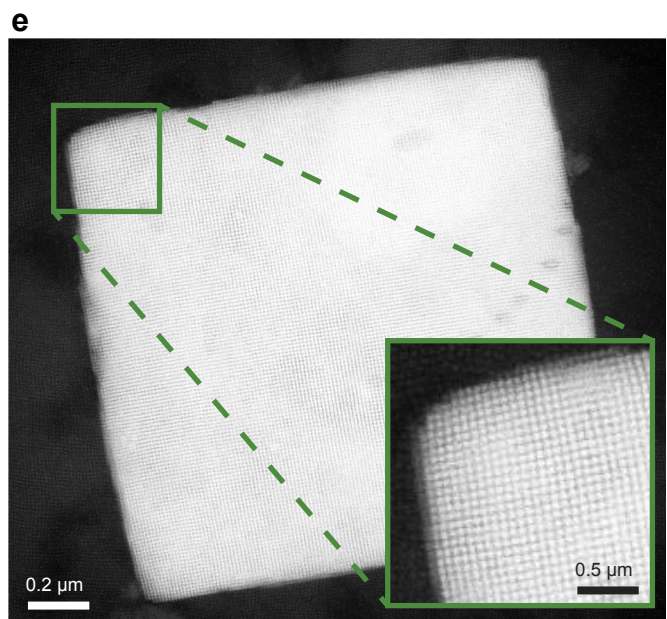Establish coherence via photon field

(τ_D)

Superfluorescence
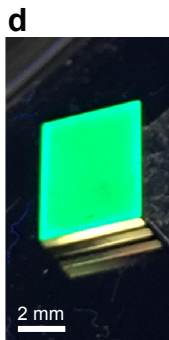
(τ_{SF})

Uncorrelated dipoles

Correlated dipoles

a

b

2 nm

c

20 μm

d

2 mm

e

0.2 μm

0.5 μm

a

Normalized intensity vs Energy (eV)

Legend:
- PL superlattices
- PL QDs in glassy thin film

Coupled QDs

Uncoupled QDs

b

Normalized intensity vs Time (ns)

$\tau_{Uncoupled} = 400$ ps
$\tau_{Coupled} = 148$ ps

Inset: $1/e$ decay time (ps) vs Excitation density ($\mu J/cm^2$)

a ... **b**

a

Time (ps) vs Energy (eV)

Intensity (a. u.)

b

Intensity (a. u.) vs Time (ps)

- 1200 μJ/cm²
- 600 μJ/cm²
- 240 μJ/cm²
- 60 μJ/cm²
- 20 μJ/cm²

c

τ_{SF} (ps)

Peak intensity (a. u.)

τ_D (ps)

Excitation density (μJ/cm²)