Single-Photon Superradiance from a Quantum Dot

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We report on the observation of single-photon superradiance from an exciton in a semiconductor quantum dot. The confinement by the quantum dot is strong enough for it to mimic a two-level atom, yet sufficiently weak to ensure superradiance. The electrostatic interaction between the electron and the hole comprising the exciton gives rise to an anharmonic spectrum, which we exploit to prepare the superradiant quantum state deterministically with a laser pulse. We observe a fivefold enhancement of the oscillator strength compared to conventional quantum dots. The enhancement is limited by the base temperature of our cryostat and may lead to oscillator strengths above 1000 from a single quantum emitter at optical frequencies.

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Enhancing and tailoring light-matter interaction is at the heart of modern quantum physics, partly because it enables studying hitherto unexplored realms of physics and partly to meet the steep requirements for quantum-information science. Photonic nanostructures efficiently tailor the density of optical states and have proven very useful to this end. For example, cavities can reach strong coupling to emitters [1,2] or mechanical objects [3], and photonic waveguides enable efficient photonic switches [4] and single-photon sources [5]. Another approach to enhancing light-matter interaction concerns tailoring the capability of the emitter to be polarized, i.e., the oscillator strength. This can be achieved with collective effects such as superradiance [6], which has been studied in ensembles of atoms [7], ions [8], Bose-Einstein condensates [9], and superconducting circuits [10]. Collective enhancement can occur at the single-photon level if a single quantum of energy is distributed coherently in an ensemble [6]. This single-photon superradiance (SPS) has been studied so far in ensembles of noninteracting emitters such as nuclei [11], and is central to schemes for robust quantum communication [12] and quantum memories [13]. A drawback of noninteracting systems is their harmonic energy structure, which prohibits deterministic preparation of a particular collective state. Here we show that the fundamental optical excitation of a weakly confining quantum dot is a generalization of SPS. We prepare the collective quantum state deterministically with a laser pulse and demonstrate its superradiant character. Our findings underline the extraordinary potential of weakly confining quantum dots for achieving unprecedented light-matter coupling strengths at optical frequencies, which would improve the radiative efficiency, quantum efficiency, quantum nonlinearities, and coherence of single-photon sources in nanophotonic quantum devices [14].

We study quantum dots formed by intentional monolayer fluctuations of a quantum well, which were pioneered by Gammon *et al.* [15], cf. Fig. 1(a). The subwavelength size of the quantum dot is key to achieving a large collective enhancement; in larger ensembles, such as atomic clouds, the enhancement is reduced by destructive interference [6,16]. The fundamental optical excitation of the quantum dot is an electron-hole pair bound by electrostatic attraction and quantum confinement, i.e., an exciton. We demonstrate that the exciton recombines radiatively with a quantum efficiency of $(99 \pm 2)\%$, which is the highest reported on quantum dots so far [17–19]. The resulting single photons inherit the superradiant character in the form of an enhanced emission rate compared to conventional strongly confining quantum dots. We employ a recently developed method exploiting the fine structure of the exciton [19] and measure an oscillator strength of up to 96 ± 2. The corresponding superradiant enhancement of about 5 is limited by the base temperature of our cryostat (7 K) and could potentially be orders of magnitude larger at temperatures below 1 K.

The hallmark of SPS is the symmetric collective quantum state [6],

$$|\Psi\rangle = \frac{1}{\sqrt{N}} \sum_j |g_1 g_2 \ldots e_j \ldots g_N\rangle, \tag{1}$$

where N is the number of emitters, the jth emitter is in the excited state $|e\rangle$, and all others are in the ground state $|g\rangle$. The remarkable property of $|\Psi\rangle$ is that it interacts with light N times stronger than a single emitter. This state describes a noninteracting ensemble, where the excitation is localized in a single emitter at a time as depicted in Fig. 1(b). In a system of interacting particles, such as a semiconductor quantum dot, the wave functions of the underlying atoms

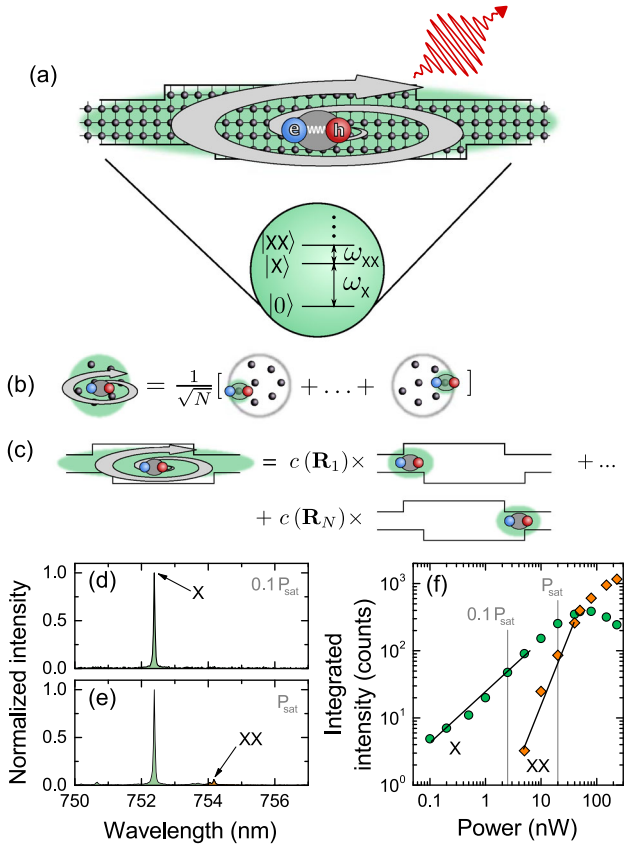

FIG. 1. Superradiant excitons in quantum dots. (a) A quantum dot defined by intentional monolayer fluctuations weakly confines electrons (e) and holes (h), which are mutually bound by electrostatic attraction. Notably, the spectrum is anharmonic due to interactions; i.e., the energy $\hbar\omega_{XX}$ of a biexciton is less than the energy $\hbar\omega_X$ of a single exciton. The swirling arrow indicates superradiantly enhanced light-matter coupling. (b) SPS is defined in an ensemble of noninteracting emitters as a symmetric superposition of different excitations. (c) The excitonic enhancement of light-matter interaction may be regarded as a generalization of SPS: the exciton is a symmetric superposition of excitations. (d) Measured photoluminescence spectrum at 10% of the exciton saturation power $P_{sat} = 20$ nW. Only the exciton is observed. (e) At the saturation power, the biexciton becomes discernible. (f) The excitons and biexcitons are distinguished by their power-law dependence on excitation power P: the fits yield $P^{0.86}$ and $P^{2.01}$, respectively.

overlap, leading to delocalized excitations. This destroys the collective enhancement of light-matter interaction and causes conventional quantum dots to exhibit small oscillator strengths of about 10, despite the fact that they embody tens of thousands of atoms.

The spatial extent of delocalized excitations is a fundamental property of semiconductors and is determined by the size of an exciton. Enhancement of light-matter interaction can therefore be achieved only in quantum dots that are larger than the exciton radius. This regime is known as weak confinement and the enhancement of the light-matter coupling in weakly confining quantum dots was first predicted by Hanamura [20] and dates back to early theoretical studies of impurities in semiconductors [21]. Here we show that this effect is equivalent to SPS, and the exciton state can be written as (see Supplemental Material [22])

$$\Psi_X(\mathbf{R},\mathbf{r}) = \sum_j c(\mathbf{R}_j)\phi_X(\mathbf{R}-\mathbf{R}_j,\mathbf{r}), \tag{2}$$

where $\mathbf{r}$ ($\mathbf{R}$) is the relative (center-of-mass) electron-hole coordinate, and the index j runs over the unit cells constituting the quantum dot. The function ϕ_X describes an exciton with the size and oscillator strength of a conventional quantum dot, while c is responsible for the collective enhancement as illustrated in Fig. 1(c). The light-matter coupling is proportional to the number of atoms comprising the weakly confining quantum dot. This analysis shows that this effect is a generalization of SPS, cf. Figs. 1(b) and 1(c), and the two effects are equivalent if c is constant throughout the quantum dot. The constant phase of c found for ground-state excitons with s-like symmetry ensures constructive interference among the excitations defined by ϕ_X.

An exciton governed by Eq. (2) has been long sought in solid-state quantum optics [42] because it can lead to large oscillator strengths. Realizing weakly confining quantum dots has been a challenge so far because it requires precise control over growth parameters to obtain a homogeneous potential profile over extended length scales. Previous studies on large quantum dots [18,19] revealed small oscillator strengths, which is believed to be caused by inhomogeneous potential profiles within the quantum dots. Previous works [43,44] found fast recombination rates in gallium-arsenide quantum dots but without rigorous information about the impact of nonradiative processes, see Supplemental Material [22] for further discussions. The measured spectra shown in Figs. 1(d) and 1(e) were obtained by exciting in the quasicontinuum energy band of the quantum well as discussed below. An exciton and a biexciton are identified as shown in Fig. 1(f). These quasiparticles radiate at different frequencies, cf. Fig. 1(e), which reflects the spectral anharmonicity of the quantum dot.

To identify proper excitation conditions of the quantum dot, we probe the spectrum of states with photoluminescence-excitation spectroscopy as displayed in Fig. 2(a). The spectrum shows a quasicontinuum band of quantum-dot states hybridized with quantum-well states as well as the exciton manifold in which we identify the $1s$, $2s$, and $3s$ states of two-dimensional excitonic hydrogen [45]. Key features of the spectrum are summarized in Fig. 2(b). We use two excitation conditions to prepare the $1s$ exciton: (i) Pumping in the quasicontinuum band of states (C-type excitation) allows extracting the impact of nonradiative

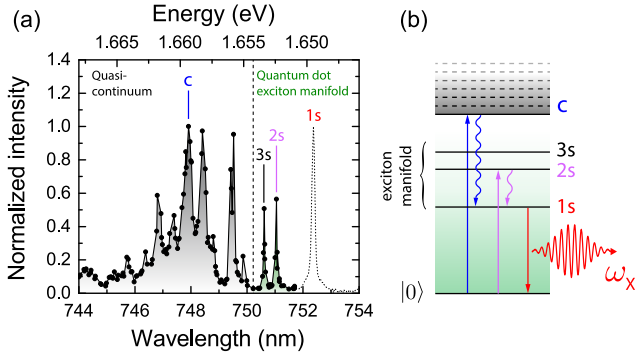

FIG. 2. Deterministic preparation of superradiant excitons. (a) Photoluminescence-excitation spectrum obtained by integrating the emission of the $1s$ transition while scanning the excitation wavelength. It features a quasicontinuum and resolves the lowest-energy states of the exciton manifold, labeled $1s$, $2s$, and $3s$. (b) Two excitation schemes are used in our study. With C-type excitation, an equal bright- and dark-exciton population is prepared, which is important for extracting the impact of nonradiative processes. Deterministic preparation of the bright exciton is achieved by pumping into the $2s$ state.

processes governing the $1s$ exciton decay. Since the quantum dot traps carriers with random spin, equal populations of spin-bright and spin-dark $1s$ excitons are prepared. While only bright excitons emit light, the dark excitons influence the decay dynamics and play a key role in revealing nonradiative effects (see Supplemental Material [22]). (ii) Deterministic preparation of spin-bright superradiant $1s$ excitons is achieved by pumping into the $2s$ exciton state, cf. Fig. 2, with a pulsed laser. This is feasible since the decay cascade from $2s$ to $1s$ is spin conserving [46] and spin-dark states are not populated. Deterministic excitation occurs when applying sufficient optical power (300 nW) to saturate the emission from the $1s$ state.

The figure of merit for collective enhancement of light-matter interaction is the oscillator strength f, which gauges the strength of the interaction with light. The oscillator strength is determined by the radiative spontaneous-emission rate of an emitter placed in a homogenous photonic environment. In an experiment, however, the oscillator strength is masked by nonradiative effects and the nonhomogeneity of the photonic environment, whose contributions are fully addressed in our study, see Supplemental Material [22] for further details. Central to our analysis is a recently developed method exploiting the fine structure of excitons to rigorously separate radiative from nonradiative effects [47]. Figure 3(a) shows raw data of the time-resolved decay of the deterministically prepared $1s$ exciton. We measure an excellent near-unity radiative efficiency of $\eta = (99 \pm 2)\%$, which is the highest ever measured on quantum dots so far. The extracted oscillator strength of $f = 72.0 \pm 0.8$ is enhanced far beyond the upper limit of $f = 17.4$ for conventional quantum dots at this wavelength. By combining structural information about the sample with the measured oscillator strength we can faithfully reconstruct the exciton wave function and find a diameter of 24 nm, which is smaller than the wavelength of light yet sufficiently large to embody ~90 000 atoms in a collective quantum state sharing a single quantum of energy.

Figure 3(b) shows the second-order correlation function obtained in a Hanbury Brown–Twiss (HBT) experiment from which we find a normalized zero-time correlation function of $g^{(2)}(0) = 0.13$. Antibunched emission directly pinpoints to the presence of a single quantum of matter inside the quantum dot, which is an essential property of SPS, cf. Eq. (1). In conjunction with the measured enhanced oscillator strength for a spatially confined

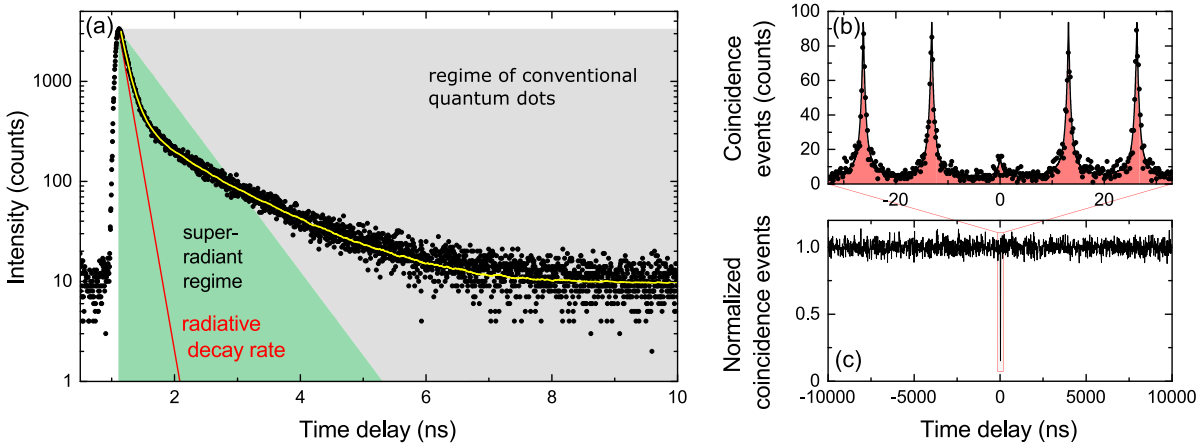

FIG. 3. Experimental demonstration of single-photon superradiance from a quantum dot. (a) Time-resolved decay (black points) of the bright $1s$ exciton obtained under $2s$ excitation. We obtain an excellent fit (yellow line) to the theoretical model when convoluting with the instrument-response function of the detector. After separating nonradiative from radiative effects we extract a radiative decay rate of (8.3 ± 0.1) ns^{-1} (red line), which is deeply in the superradiant regime (green area). (b) HBT measurement of the emitted photons showing $g^{(2)}(0) = 0.13$, which proves single-photon emission. (c) Long-time-scale HBT measurement where each coincidence peak has been numerically integrated. No blinking is observed.

exciton, this is the unequivocal demonstration of SPS in a quantum dot. Solid-state quantum light sources often suffer from charge traps that switch the emitter into an optically dark state, also known as blinking processes, and reduce the preparation efficiency of bright states. This can be quantified from HBT correlations acquired over long time scales as shown in Fig. 3(c). No bunching effects are observed, which shows that this single-photon source is free from blinking on a time scale of at least 10 μs.

Single-photon superradiance is a robust phenomenon in quantum dots due to the anharmonic spectrum. The only experimentally relevant parameter that may be detrimental to SPS is temperature. For a thermal de Broglie wavelength larger than the quantum-dot size, the exciton Pauli blockage is broken and leads to multiphoton emission, thereby destroying SPS. This nontrivial effect is beyond the scope of the current work and will be presented elsewhere.

We have measured the oscillator strength of nine quantum dots and found them all to be superradiant with an average oscillator strength of $f = 76 \pm 11$. Further experimental data are included in the Supplemental Material [22]. Remarkably, we have measured a homogeneous-medium radiative decay rate of up to $\Gamma_{\mathrm{rad}} = (11.1 \pm 0.2)\ \mathrm{ns}^{-1}$, which is the fastest value ever reported for any single-photon source and corresponds to an oscillator strength of $f = 96 \pm 2$. Such a quantum dot can deliver a radiative flux of single photons equivalent to more than five conventional quantum dots.

The superradiant enhancement of the light-matter coupling in quantum dots is proportional to the number of atoms in the collective state, and can potentially be much larger than reported here. The enhancement factor may realistically reach 100× for quantum-dot diameters of ∼100 nm [48] corresponding to an oscillator strength of $f \sim 1500$. Such highly superradiant quantum dots may exist in our sample, but the temperature at which the experiment is carried out ($T = 7$ K) does not allow resolving such large oscillator strengths. This is because in large quantum dots the confinement energy may become smaller than the thermal energy, which results in populating excited states with reduced oscillator strength. The maximum oscillator strength $f_{\mathrm{max,th}}$ that can be resolved at a temperature T is calculated for a quantum dot in which the energy difference between two eigenstates equals $4k_BT$ (a detailed analysis is provided in the Supplemental Material [22])

$$f_{\mathrm{max,th}} = \frac{4\hbar E_P}{M\omega a_0^2}\frac{1}{\xi}\frac{1}{k_BT}, \tag{3}$$

where E_P is the Kane energy, M the exciton mass, a_0 the exciton radius, k_BT the thermal energy, and $1{:}\xi$ the in-plane aspect ratio of the quantum dot ($\xi \geq 1$). At $T = 7$ K we find that oscillator strengths larger than $f_{\mathrm{max,th}} = 170$ cannot be resolved for in-plane symmetric quantum dots, and $f_{\mathrm{max,th}}$ decreases even further for more realistic asymmetric shapes. Oscillator strengths of ∼1500 require temperatures below ∼0.8 K. The light-matter coupling may be further enhanced $> 10\times$ beyond the homogeneous-medium value by the Purcell effect [14]. This could allow studying fascinating non-energy-conserving effects such as the ultrastrong regime of light-matter coupling [49]. The repetition rates of single-photon sources would approach the terahertz regime yielding radiating powers of hundreds of nanowatts from a single quantum emitter. The single-photon emission would potentially be highly coherent, partly due to an intrinsically weaker coupling to nuclear spin noise [50] and phonon dephasing [51] for large excitons, partly because the dephasing mechanisms present in solid-state environments would be largely negligible compared to a radiative decay at subpicosecond time scales. Even larger decay rates could become possible in materials with small Bohr radii [52,53], see Eq. (3). In particular, fast decays have recently been reported in CdSe nanoplatelets [54]. Another intriguing aspect of the SPS regime is that the collective Lamb shift is predicted to be finite [55] without the renormalization schemes required in the quantum electrodynamics of conventional emitters.

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