A Quantum Dot Single Photon Source

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Abstract. We demonstrate heralded single photon emission from a self-assembled InAs quantum dot (QD). Pulsed optical excitation (82 MHz) together with Coulomb renormalization effects allows for the realization of regular single photon emission at the excitonic transiton (1X) with nearly 100 % efficiency. By temperature tuning, we are able to shift the 1X transition into resonance with a whispering gallery mode of a microdisk (Q $\sim$ 6500) and achieve turnstile operation of the coupled QD-cavity system. On resonance, the Purcell effect causes a reduction of the 1X transition lifetime leading to a reduced time jitter of the photon emission event and ensuring that photons are primarily emitted into a cavity mode.

1 Introduction

A single photon source, which is able to generate photons on demand, has been a major challenge for many years. Such a source allows the ultimate quantum control of the photon generation process, i.e., single photons can be generated within short time intervals and a deterministic dwell time between successive photon generation events. This makes it possible to encode information on a single photon level. Such a source is of interest for future applications in quantum computing [1] and quantum cryptography [2].

One might suspect that single photons could be produced with high probability by adjusting the average number of photons in a light pulse. This is not possible since photons from classical light sources follow Poisson statistics or super-Poisson statistics [3]. For example, if one adjusts the average number of photons in a laser pulse as low as 0.1, 0.5 % of the pulses will contain two or more photons, 9 % will have one photon, and 90.5 % will contain no photon at all.

The basic requirement of a single photon source is that the active emitter possess a high quantum efficiency $\eta \sim 1$ and is able to emit one photon after each other. Such a photon antibunching behavior has been reported for different single quantum emitters, e.g., an atom [4], a stored ion [5], a molecule [6], a

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semiconductor quantum dot [7,8], and a single nitrogen-vacancy center in diamond [9,10]. Photon antibunching is a necessary but not sufficient condition for a single photon turnstile device; an additional mechanism for regulating the excitation process is required to realize single photon pulses.

A single photon turnstile device based on a mesoscopic double barrier p-n heterojunction was proposed in 1994 [11]. An extension of this proposal was recently demonstrated [12] where single as well as multiple photon emission events with a repetition rate of 10 MHz at 50 mK has been reported. This device utilizes Coulomb blockade of tunneling for electrons and holes in a mesoscopic p-n diode structure to regulate the photon generation process. In this scheme, single electron and hole charging energies must be large compared to the thermal background energy to ensure single photon emission. Therefore, this device can only be operated at ultra-low temperatures ($T \leq 1$ K). Triggered single-photon sources based on a single molecule have been demonstrated [13,14] whereby regulation of the photon emission process is achieved either by adiabatic passage techniques or non-resonant pumping with pulsed optical excitation. More recently, triggered single photons have been generated by pulsed optical excitation of a single quantum dot [15,16]. The photon correlation measurements reported in Refs. [13,14,16] showed background emission leading to a significant probability ($P = 0.12$ - 0.26) for two-photon pulse generation, while vanishing two-photon generation is reached in our results [15]. In addition, the QD has also been resonantly coupled to a high-Q cavity microdisk mode in our work. We demonstrate that the Purcell effect significantly reduces the jitter in the photon emission time leading to an estimated possible repetition rate of ~ 1 GHz for such a device.

To ensure single photon generation at the fundamental QD exciton transition (1X), we adjust the pump power so that two or more electron-hole pairs are captured by the QD during each excitation pulse. The energy of the photons emitted during relaxation depends significantly on the number of multiexcitons that exist in the QD, due to Coulomb interactions enhanced by strong carrier confinement [18]. If the total recombination time of the multiexciton QD state is longer than the recombination time of the free electron-hole pairs, each excitation pulse can lead to at most one photon emission event at the 1X-transition. Therefore, regulation of photon emission process can be achieved due to a combination of Coulomb interactions creating an anharmonic multiexciton spectrum and slow relaxation of highly-excited QDs leading to vanishing re-excitation probability following the photon emission event at the 1X-transition [18]. If the QD exciton recombination is predominantly radiative, every excitation pulse from the mode-locked laser will generate a single photon pulse.

2 Experimental

Our samples were grown by molecular beam epitaxy (MBE) on a semi-insulating GaAs substrate. Figure 1 shows the microdisk structure which consists of a 5µm diameter disk and a 0.5µm $\mathrm{Al}_{0.65}\mathrm{Ga}_{0.35}\mathrm{As}$ post. The disk area consists of 100 nm GaAs, an InAs QD layer, and 100 nm GaAs. Details of the microdisk processing can be found in Ref. [19]. The QDs were grown using the partially covered island technique [20] with a gradient in the QD density reaching from $\leq 10^8\ cm^{-2}$ to $\sim 10^{10}\,\mathrm{cm}^{-2}$ across the sample wafer. The QDs possess a diameter of ∼ 40–50 nm and a height of ∼ 3 nm, emitting in the wavelength range from 920 to 975 nm.

Our experimental setup combines a low-temperature diffraction-limited scanning optical microscope for spatially resolved photoluminescence (PL) spectroscopy and an ordinary HBT setup for photon correlation measurements. The system provides spectral resolution of 70 μeV, spatial resolution of 1.7µm , and temporal resolution of 420 ps. The microdisks are mounted in a He gas flow cryostat. Optical pumping is performed with a mode-locked femtosecond (∼ 250 fs) Ti:sapphire laser, operating at 750 nm. The electron-hole pairs were mainly generated in the GaAs barriers and subsequently captured by the QDs within a short timescale ($<$ 35 ps [21]). A microscope objective (with numerical aperture NA = 0.55) was used to focus the excitation laser onto the sample and to collect the emitted PL from the QDs. The collected light was spectrally filtered by a monochromator and then split with a 50/50 beamsplitter. The resulting two light beams were focused onto two single-photon-counting avalanche photodiodes (SPAD). The pulses from the two SPADs were used to start and stop a time-to-amplitude converter (TAC) whose output is stored in a multichannel analyzer (MCA). The resulting histograms yield the number of photon pairs $n(\tau)$ with arrival time separation of

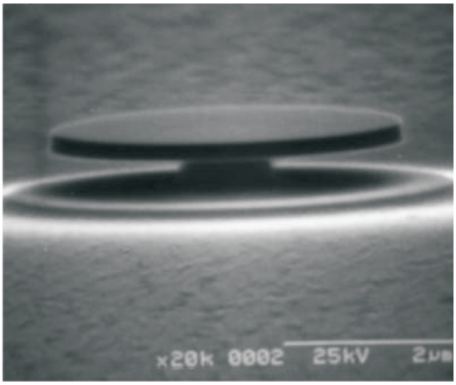

Fig. 1. The microdisk structure that consists of a 5µm diameter disk and a 0.5µm post. The GaAs disk area that supports high-quality factor whispering gallery modes is 200 nm thick and contains InAs quantum dots

$\tau = t_{start} - t_{stop}$. The measured distribution $n(\tau)$ is equivalent to the unnormalized correlation function $G^{(2)}(\tau)$ in the limit where the reciprocal of the average counting rate is much longer than the measured time separation τ between photon pairs [22], which was always the case for our measurements.

3 Results

Figure 2 shows power dependent PL spectra for a 5µm diameter disk in the range between 1.310 and 1.348 eV. For this measurement the sample was excited with a continuous-wave Ti:sapphire laser at 760 nm. At low excitation power (1 W/ cm^2), a single sharp line (1.3222 eV) due to single exciton recombination (1X) is observed. With increasing excitation power two lines at 1.3208 and at 1.3196 eV appear below the single exciton line. The line at 1.3196 eV shows a superlinear increase with excitation intensity and originates from a biexciton decay (2X) whereas the line at 1.3208 eV (M) is due to background emission which is coupled into a whispering gallery mode (WGM). The correlation of the M line to a WGM will become clear in the discussion below. The inset of Fig. 2 shows the measured cw correlation function $g^{(2)}(\tau)$ for the 1X transition of the single QD in the microdisk at the

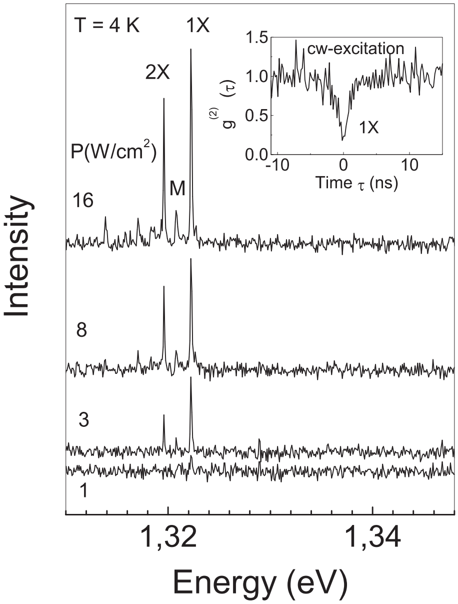

Fig. 2. Power dependent PL spectra from a single InAs QD embedded in a 5µm diameter microdisk. Contributions from the excitonic ground transition (1X), higher excited states (e.g. biexciton (2X)), and a whispering gallery mode (M) are visible. Inset: Measured cw correlation function $g^{(2)}(\tau)$ for the single QD 1X transition. The time bin is 195 ps and the excitation power is 160 W/ cm^2

onset of saturation. Saturation is defined here as the pump intensity where the 1X line reaches its maximum intensity. The dip at $\tau = 0$ arises from photon antibunching [4] and the fact that $g^{(2)}(\tau) < 0.5$ proves that the emitted light from the 1X transition stems from a single, anharmonic quantum emitter.

Photon correlation measurements carried out under pulsed excitation yield signatures for turnstile operation, by discriminating between one and two-photon (Fock-state) pulses as well as coherent-state pulses [13]. For a pulsed periodic coherent source which emits Poissonian light, the peak at $\tau = 0$ would be identical to the peaks at integer multiples of the repetition rate T_{rep}, for all values of the mean photon number. In contrast, for an ideal turnstile device the peak at $\tau = 0$ is absent [23].

Figure 3 shows the measured unnormalized correlation function $G^{(2)}(\tau)$ for (A) the pulsed Ti:sapphire laser, and (B) the 1X transition of a QD that is far detuned from all WGMs (T= 4 K). The pump intensity in this experiment corresponds to an excitation of the QD where the 1X emission is well into the saturation regime [24]. As expected, the measured $G^{(2)}(\tau)$ of the pulsed

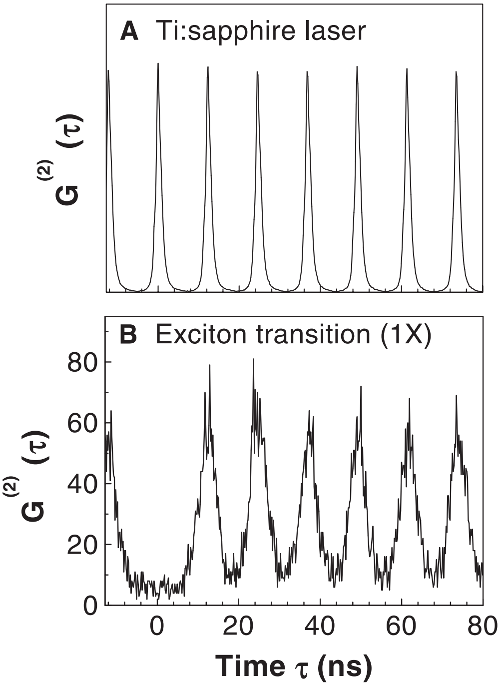

Fig. 3. Measured unnormalized correlation function $G^{(2)}(\tau)$ of (A) a mode-locked Ti:sapphire laser (FWHM = 250 fs), and (B) a single QD excitonic ground state (1X) emission under pulsed excitation conditions (82 MHz). The QD 1X transition was out of resonance with the microdisk modes

Ti:sapphire laser exhibits peaks at integer multiples of $T_{rep} = 12.27\,\mathrm{ns}$ with negligible signal in between the peaks. The measured $G^{(2)}(\tau)$ of the QD 1X emission at $T = 4$ K (Fig. 3B) also shows peaks at integer multiples of T_{rep}, indicating the locking of the photon emission to the pulsed excitation. But in contrast to the mode-locked laser, the peak at $\tau = 0$ is no longer present, i.e., the probability of finding a second photon following the detection of the first photon at $\tau = 0$ vanishes. This is the principal result of our work: absence of the peak at $\tau = 0$ provides strong evidence for an ideal single photon turnstile operation.

The lifetime of the single exciton ground-state transition (1X) was determined from cw antibunching experiments to be 2.2 ns, which is the shortest possible total recombination time for a multiply-excited QD. As the recombination time in the GaAs barrier and the wetting layer is considerable faster (100–200 ps), no free carriers are available to re-excite the QD after the 1X recombination process. As discussed earlier, only one 1X recombination process can occur per excitation pulse under these conditions. To ensure that a single photon is indeed emitted for each excitation pulse, the pump power of the excitation laser should be adjusted so that the probability of having no injected electron-hole pair in the QD is negligible. The fact that the photon correlation measurement depicted in Fig. 3B was obtained well in the saturation regime ensures that QD is multiply-excited in our experiments. In addition, the quantum efficiency of the QD has to be high $\eta \sim 1$ to avoid nonradiative recombination processes. Recent experiments reported in Ref. [8] have shown that for our samples the dominant recombination mechanism is radiative. These facts allow us to conclude that the generated light at the excitonic ground-state transition energy 1X is a stream of single photons with a repetition rate of 82 MHz.

By temperature tuning we are able to shift the 1X transition shown in Fig. 2 into resonance with the cavity mode M (Q ∼ 6500). The crossing between the WGM and the QD 1X-transitions is shown in Fig. 4a where we plot the energies of the two lines vs. temperature. The WGM appears at an energy of 1320.7 meV at 4 K and shifts only slightly to an energy of 1319.6 meV at 54 K. On the other hand, the QD 1X-transition shifts strongly with temperature, over 3 meV within 50 K temperature difference. The different energy shifts of the 1X-transition and the WGM with temperature give rise to a crossing of the two resonances. The temperature dependence of the energy of the WGM can be attributed to the change in the refractive index of GaAs with temperature. On the other hand, the temperature dependence of the energy of the 1X-transition is caused by the changes in the bandgaps of InAs and GaAs with temperature.

Figure 4b shows the change in the intensity of the WGM emission as function of the 1X-WGM detuning. At a temperature of 44 K (zero detuning) the intensity of the WGM luminescence increases by a factor of 29 compared to its value at 4 K, strongly indicating a resonance between the QD 1X-transition

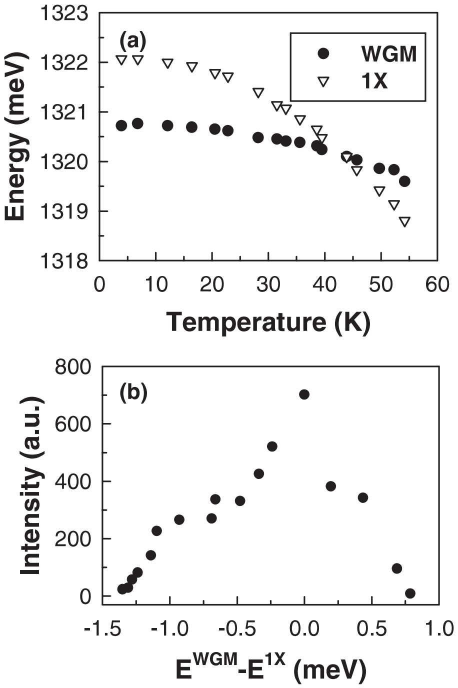

Fig. 4. (a) Change of the WGM and the 1X-transition emission energy with temperature (excitation power = 13 W/ cm^2). (b) Change in the intensity of the WGM luminescence with detuning (excitation power = 13 W/ cm^2)

and the WGM. The observed crossing together with resonant enhancement of luminescence are evidence for the weak coupling between the WGM and the single QD.

In the weak coupling regime enhancement of the spontaneous emission rate of the QD 1X-transition due to the Purcell effect [25] is expected. To quantify the magnitude of the Purcell effect we have carried out pump-power dependent cw photon correlation measurements; this method has been previously shown to be a reliable alternative to standard time-resolved measurements for determining recombination times [8]. Moreover, this method is able to discriminate between single QD emission ($g^2(0) < 0.5$) and emission from several QDs ($g^2(0) > 0.5$). Fig. 5 shows photon correlation measurements performed at 4 K (out of resonance) and 44 K (in resonance) with excitation powers of 36 W/ cm^2 and 5 W/ cm^2 respectively. After normalization, the measured correlation functions show clear dips at zero time delay ($g^{(2)}(0) = 0.08$ at 4 K, $g^{(2)}(0) = 0.38$ at 44 K) indicating strong photon antibunching. Since $g^{(2)}(0) < 0.5$ in our measurements, we can state that the observed emission lines stem from the 1X-transition of a single QD [8]. The

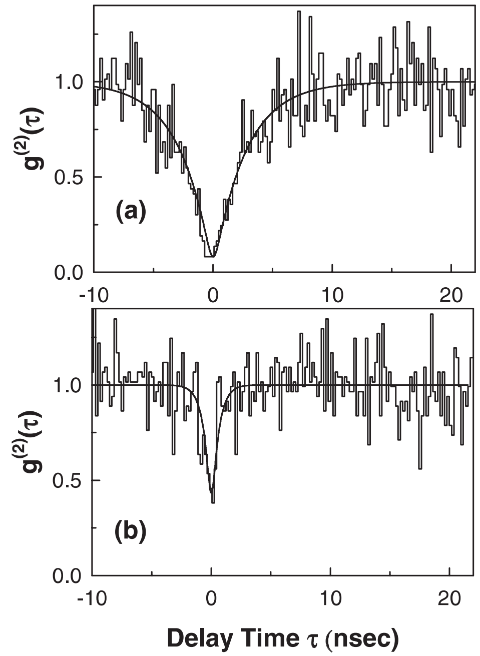

Fig. 5. Measured photon correlation function of the 1X-transition of the single QD: out of resonance with the WGM, at 4 K, under an excitation power of 36 W/ cm^2 (trace a), and in resonance with the WGM, at 44 K, under an excitation power of 5 W/ cm^2 (trace b)

observation of $g^{(2)}(0) < 0.5$ at 44 K also supports that, in resonance, the QD 1X-transition is the main emission feeding the WGM luminescence.

From photon correlation measurements at two different pump powers, we deduced the lifetime of the 1X-transition at 4 K. In these measurements decay times of 2.7 ns and 1.5 ns were observed at excitation levels below saturation (36 W/ cm^2, Fig. 4a) and at the onset of saturation (92 W/ cm^2) of the 1X-transition, respectively. From a three-level rate-equation model that includes 1X and biexcitonic (2X) transitions and omits any higher multiexcitonic recombinations or any other population decay channels (e.g. Auger processes) [8], a lifetime of 3.4 ns is determined for the 1X-transition at 4 K. A conventional time-correlated single photon counting (TCSPC) measurement on this QD exhibits a decay time of 2.8 ns, showing reasonable agreement with the lifetime deduced from the photon correation measurements. These values are larger than previously reported lifetimes for the 1X-transition of a single InAs QD (∼ 1 ns) [24]. The lifetime difference can be explained by the different photonic environment created by the microdisk that partially inhibits spontaneous emission [8].

Our photon correlation measurements at resonance, 44 K, revealed decay times of 560 psec and 370 psec at pump powers of $5\,\mathrm{W/\,cm^2}$ (Fig. 4b), and $45\,\mathrm{W/\,cm^2}$ respecticely, corresponding to excitation levels below the saturation of the 1X-transition. By using the pump power dependent method described in the previous paragraph, a lifetime of 590 psec is determined for the 1X-transition at 44 K. This provides a strong indication of lifetime reduction caused by the Purcell effect. A more detailed temperature dependent TCSPC study that demonstrates the Purcell effect will be published elsewhere [26].

If a quantum emitter is spectrally matched with a single cavity mode, located at a maximum of the electric field, and its dipole is aligned with the local electric field, the Purcell factor is given by $F_p = 3Q\lambda_c^3/4\pi^2 V$ [25], where Q is the quality factor of the cavity, λ_c is the wavelength of the emission in the cavity, and V is the effective mode volume. For a microdisk, the ideal spontaneous emission enhancement can be estimated by factor of $((2/2)F_p + 1)$, where the various terms account, respectively for WGM degeneracy (2), the random dipole orientation in the plane of the QD (1/2), and the contribution of the emission into leaky modes (1) [18]. Taking the parameters of our microdisk an enhancement in the spontaneous emission rate of 17 is estimated. The fact that the measured value ($\sim 5-6$) is much smaller than the predicted value 17 for ideal coupling can be attributed to the non-ideal spatial overlap of the QD and the WGM. Finally, we want to stress that a thorough theoretical analysis of the Purcell factor in our microdisk would have to account for the special photonic environment of the microdisk which is beyond the scope of this contribution.

Figure 6 shows the measured unnormalized correlation function $G^{(2)}(\tau)$ for the 1X transition of Fig. 2 (A) out of resonance (T= 4 K), and (B) in resonance (T= 36 K) with the WGM. The resonance condition is achieved at slightly lower temperature due to the higher pump power used in the pulsed experiment ($22\ \mathrm{W/\,cm^2}$). We emphasize that the photon correlation signals shown in Figs. 3B and 6 are obtained for different QDs; the 1X recombination time for the QD analyzed in Fig. 6 is 3.4 nsec, which explains the appearance of broader peaks. When the QD is in resonance with the WGM, the FWHM of the photon correlation peaks are narrower (factor 3.4) than the out of resonance case, i.e., the time jitter between successive photon generation events is reduced. This is a direct consequence of the Purcell effect which causes a reduction of the ground state transition lifetime τ_{1X} and ensures that photons are primarily emitted into the cavity mode.

A small peak at $\tau = 0$ is observed in the resonance case (see Fig. 6B). The intensity ratio of this peak to the peaks at iT_{rep} is directly related to the fraction of pulses having two or more photons [13]. An experimental ratio R = 0.29 is deduced from Fig. 6B. The fact that R is larger than the ideal value of zero could be due to the Purcell effect, which increases the probability of capturing a second electron-hole pair from the wetting layer after

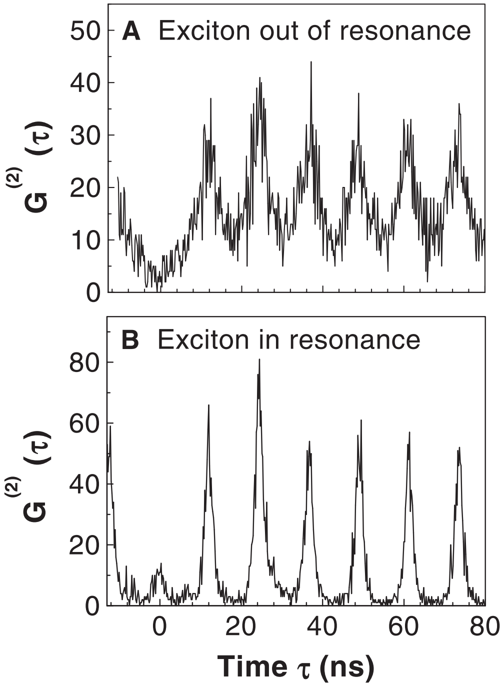

Fig. 6. Measured unnormalized correlation function $G^{(2)}(\tau)$ of a single QD excitonic ground state emission (A) out of resonance, and (B) at resonance with a cavity mode ($Q \sim 6500$), under pulsed excitation conditions (82 MHz). The average pump intensity in both cases was $\sim 22\ \mathrm{W/cm^2}$

the 1X recombination process has occurred. Another possible explanation is the contribution from the background light generated by the wetting layer or by the excited states of other QDs. There are two experimental observations that support the latter explanation: first, even when the ground-state transition of the QD (1X) is off resonance the mode emission is still visible, indicating the influence of the background (see Fig. 2). Second, using higher average pump powers P in the resonant case increases R (R = 0.36 (0.55) for P = 56 (303) $\mathrm{W/cm^2}$).

4 Summary

In conclusion, we have demonstrated heralded single photon emission from a self-assembled single InAs QD embedded in a semiconductor microdisk structure. Without coupling to a cavity mode nearly 100 % of the excitation pulses lead to the emission of a single photon with a repetition of 82 MHz. If the excitonic transition (1X) is coupled to a high-Q whispering gallery mode ($Q \sim 6500$) up to 70 % of the excitation pulses give rise to single photon emission. Due to the Purcell effect in the cavity, the time jitter of the photon emission is reduced by a factor of 3.4, thus allowing repetition rates of 1 GHz. We envision that the operating temperature of the single-photon source can be easily extended to T= 77 K, which would be very significant for practical applications. Room temperature operation could in principle be achieved by using QDs with higher confinement potentials to suppress non-radiative carrier losses into the barriers.

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