Short Communication

Bright and pure single-photons from quantum dots in micropillar cavities under up-converted excitation

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Self-assembled semiconductor quantum dots (QDs) are potentially scalable candidates for solid-state single-photon emitters which deterministically generate one single-photon per excitation. Such on-demand single-photon sources serve as one of the key elements in modern photonic quantum technologies. Until very recently, near-optimal QD single-photon sources with simultaneously high degree of brightness, single-photon purity and indistinguishability have been achieved in QD-micropillar systems, showing superior performance in photonic quantum computation/simulation, e.g., Boson sampling, thanks to the large Purcell effect and the resonant excitation scheme. In particular, advances in resonant excitation have shown that the excitonic states in single QDs can be coherently driven and manipulated, so as to minimize the undesirable dephasing processes via the reduction in fluctuations of the electrostatic environments and elimination of incoherent phonon assisted carrier relaxation.

However, direct implementations of resonant excitation on QDs in other nanostructures for efficient single-photon generation, such as nanowire, micro-lens, circular Bragg grating, and photonic crystal nanocavity, are still a challenge because it is technically nontrivial to differentiate the fluorescence from the same frequency laser scattered by the photonic nanostructures. Up-converted (UC) photoluminescence (PL) in semiconductors, describing emissions at energies higher than that of the excitation energy, has attracted growing interest in the last few years, e.g., UC excitation can be used to excite the QDs via the two-photon absorption (TPA) or Auger processes. A common drawback is that the UC process, especially a direct TPA process, is always associated with a significantly lower efficiency as compared to its down-converted counterpart. A typical QD TPA experiment requires an ensemble of QDs as well as pulsed laser to provide enough excitation power.

In this short communication, we present the first realization of UC excitation, to the best of our knowledge, on single QDs for efficient single-photon generations. The experimental setup and micropillar cavity modes are shown in Supplementary data. We investigate a QD that was deterministically embedded in a micropillar via the fluorescence imaging technique. The energy levels of the QD system are schematically shown in Fig. 1a, describing the aboveband (blue line) and UC (red line) excitation processes. In the aboveband excitation, the carriers are directly generated in the valence band and conduction band by using a laser with the energy higher than the bandgap of GaAs material. The generated carriers then relax to the wetting layer and further down to the first excited state of QD via a phonon mediated process before the radiative single-photon emission. On the other hand, the carriers are directly generated in the QD excited states in the UC excitation scenario that we will explain in details. Fig. 1b and c show the PL spectra of the QD continuously excited via aboveband (780 nm) and UC (940 nm) excitations at the saturation power, respectively. Spectral resonance between fundamental mode (FM) and the QD is introduced at the measurement temperature of 53 K. The detected fluorescent intensity in silicon charged couple device (CCD) from the QD via the UC excitation scheme is ~22.8 counts/s which is very close to the number (24.6 counts/s) achieved in the aboveband excitation. Furthermore, the wetting layer (WL) emission at ~865 nm shown in the aboveband excitation is strongly suppressed in the UC excitation scheme, indicating the reduction of carrier re-capture process that we shall present in the photon correlation measurements. Fig. 1d demonstrates the signal intensities as a function of the excitation power for both aboveband excitation and UC excitation in the logarithm scale. The quadratic dependency (\(P_x = 1.73 \pm 0.11\) in UC excitation indicates that the...
signal is generated from a two-photon nonlinear process as compared to the slope of $0.69 \pm 0.04$ in the aboveband excitation. The sideband ($\sim 940$ nm) of the distributed Bragg reflector (DBR) in the micropillar is clearly seen via a high power aboveband excitation spectrum, shown in the Fig. 1e. To reveal the origin of this efficient UC process, we perform temperature dependent photoluminescence measurements.

Fig. 1. Sketch of the excitation mechanism, PL spectra and PLE measurements. (a) Schematic energy-level diagram illustrating aboveband and UC excitation schemes. The blue line connecting the lowest quantum-dot states indicates spontaneous emission under the aboveband excitation, while the red line indicates spontaneous emission under the UC excitation. (b), (c) PL spectra of a single QD in a micropillar under 780 and 940 nm CW excitation at saturated power. (d) Excitation power dependence of the fluorescence intensity under aboveband (blue squares) and UC (red circles) excitation. The slope of each power density dependence is indicated. (e) A typical PL from QD-in-micropillar excited under 780 nm CW excitation at very high excitation power. (f), (g) The integrated intensity of the emission with varied CW excitation wavelength with half-saturated excitation power at 53 and 10 K, respectively.

Fig. 2. Single-photon emission from aboveband and UC excitation. (a) and (b) Detected fluorescent counts of the same QD as a function of 780 nm (black triangles) and 940 nm (black circles) pulsed excitation power. $P$ and $P_{sat}$ represent the excitation and saturation power. The inset shows a spectrum after a 920 nm narrow band filter. (c) and (d) Hanbury Brown and Twiss (HBT) measurement of single-photon purity under 780 nm (c) and 940 nm (d) pulsed excitation.
minescence excitation (PLE) measurements for the exciton line at 915 nm. When the QD is coupled to the fundamental cavity mode at T = 53 K, the UC-excited PL intensity increases remarkably as the tunable laser lies within the sideband of the DBRs, shown in Fig. 1f. The peaks match well with the aboveband spectrum shown in Fig. 1e. On the other hand, the intensity remains almost static when the QD is off resonance with the cavity mode at T = 10 K, shown in Fig. 1g.

The brightness and single-photon purity of the photons emitted by the QD excited via aboveband and UC excitation schemes are quantitatively compared by photon counting and correlation measurements. Excited by 120 fs optical pulses with a repetition rate of 79.3 MHz, the photon count rates in the avalanche photodiode (APD) as a function of the excitation power under pulsed aboveband and UC excitations are plotted in Fig. 2a and b, achieving 786,000 and 640,700 counts/s with excitation wavelengths of 780 and 940 nm, respectively. Each inset shows a spectrum after an 810 nm long pass filter and a 920 nm narrow band filter with a bandwidth of (1.0 ± 0.2) nm. By carefully calibrating the loss of the setup, a single-photon collection efficiency up to (77 ± 6)% is obtained for the UC excitation, that is, the percentage of generated single photons collected into the first objective lens (NA = 0.65) (see Note 3 in the Supplementary data). The second order correlation of the emitted photons under aboveband and UC excitation schemes are shown in Fig. 2c and d, in which g(2)(0) of 0.294 ± 0.031 and 0.044 ± 0.003 are obtained, respectively. For the similar brightness, the UC excitation scheme provides more than 6 times enhancements of the single-photon purity. We now provide insights into the nonlinear transfer dynamics of the QD states couple to the microcavity. As shown in Fig. 1a, we illuminated the conduction band (E_g) and valence band (E_V) along the growth direction for the InAs/GaAs QDs structure. We consider the InAs QD states coupled to the cavity mode own a bandgap of E_g = 1.355 eV. And the minimum of a continuum of GaAs states and the InGaAs wetting layer states lay hundreds of meV above the InAs QD states. When using aboveband excitation, carriers are generated in the GaAs surrounding the quantum dots, captured first by the wetting layer and then fall into the excited states of quantum dots before the spontaneous emission process. While in UC excitation, the laser is tuned to a higher transition within the quantum dot. Moreover, Fig. 1b and c depict that the carriers excited in the WL region via UC excitation is less than that from aboveband excitation. To explain this behavior, we speculate that the carriers excited by TPA process are more inclined to be captured by QDs due to low-dimensionality effect [17–19]. These carriers are subsequently available for radiative recombination resulting in PL at a higher energy than the input photon. Consequently, this low-dimensionality effect combined with the microcavity structure allow us to employ enhanced TPA to coherently excite QDs and enable generation of pure single photons, which verify the striking reduction of the g(2)(0) shown in Fig. 2d under the UC excitation.

In conclusion, we have demonstrated the successful implementations of UC excitation on single QDs for efficient generations of single-photons. By tuning the excitation laser frequency into the high-energy sidebands of the DBR in the micropillar while keeping the QD spectrally resonant with the cavity mode, we achieved bright on-demand single-photon emissions with a collection efficiency of (77 ± 6)% and a g(2)(0) of 0.044 ± 0.003 that is six times better than the value under aboveband excitations. Our results suggest that UC excitation technique could be serving as a powerful tool for efficiently generating high-quality single-photons for QDs embedded in photonic nanostructures in quantum computation/simulation experiments.

Conflict of interest
The authors declare that they have no conflict of interest.

Appendix A. Supplementary data
Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.scib.2018.05.024.

References
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