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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 (ODs) 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 [1]. Until very recently, near-optimal QD single-photon sources with simultaneous high degree of brightness, single-photon purity and indistinguishability [2] have been achieved in QD-micropillar systes, showing superior performance in photonic quantum computation/simulation, e.g., Boson sampling [3], 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 [4,5].

However, direct implementations of resonant excitation on QDs in other nanostructures for efficient single-photon generation [6], such as nanowire [7], micro-lens [8], circular Bragg grating [9,10], and photonic crystal nanocavity [11] etc., is 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) [12] or Auger processes [13]. 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

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experiment requires an ensemble of QDs as well as pulsed laser to provide enough excitation power [14].

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 (Note 1 and 2 online). We investigate a QD that was deterministically embedded in a micropillar via the fluorescence imaging technique [15,16]. 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 singlephoton 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 OD 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

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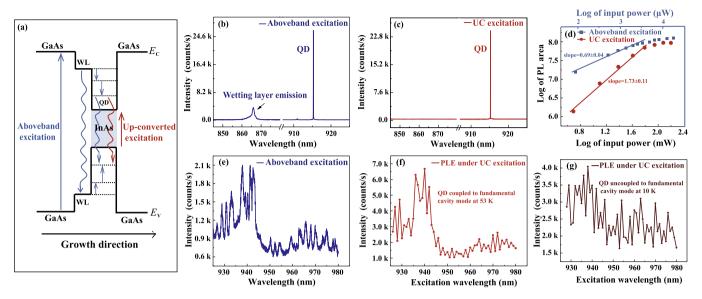


Fig. 1. Sketch of the excitation mechanism, PL spectra and PLE measurements. (a) Schematic energy-level diagram illustrating aboveband and UC excitation schemes. The blue line connecting the lowest quantum-dot states indicates spontaneous emission under the aboveband excitation, while the red line indicates 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) excitations. 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.

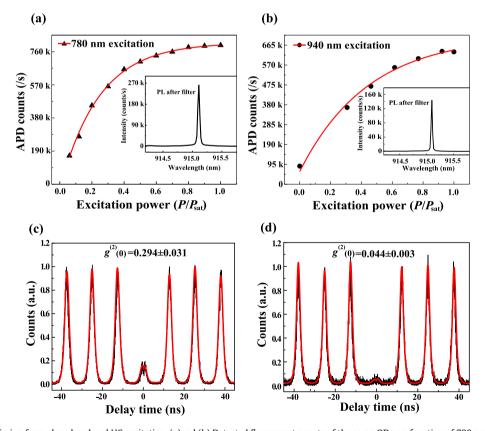


Fig. 2. Single-photon emission from aboveband and UC excitation. (a) and (b) Detected fluorescent counts of the same QD as a function of 780 nm (black triangles) and 940 nm (black circles) pulsed excitation power, P and P_{sat} represent to the excitation and saturation power. The inset shows a spectrum 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.

signal is generated from a two-photon nonlinear process as compared to the slope of 0.69 ± 0.04 in the aboveband excitation. The sideband (~ 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 photolu-

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