Strongly suppressed multi-photon generation from a single quantum dot in metal-embedded structure

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We introduce novel metal-embedded semiconductor pillar structure including quantum dots as highly efficient single-photon sources for quantum information applications. We have developed related processes for fabricating the proposed structure, and have demonstrated strongly suppressed multi-photon generation and improved photon extraction efficiency. As a result, under non-resonant excitation, which is highly preferable for the practical use, single-photon purity with $g^{(2)}(0)$ as low as 0.015 has been measured. Extremely small semiconductor volume in the metal embedded structure is a crucial factor for this observation. Moreover, single-photon generation rate from a single quantum dot up to 6.4 million per second has been achieved.

1 Introduction

For quantum information applications, realization of efficient single-photon sources is one of the key issues. Semiconductor quantum dots (QDs) are a good candidate for realizing single-photon sources, entangled-photon-pair sources [1-3], and quantum gates [4] etc. Especially, in view of the single-photon sources based on QDs, high-bit-rate on-demand generation is expected.

However, for the practical use, two main issues are remaining. First, multi-photon generation evaluated by a second-order photon correlation function at zero time delay $g^{(2)}(0)$ should be minimized. For long-distance quantum key distribution, lower $g^{(2)}(0)$ is crucial for practical applications with high bit rate operation [5]. So far, many groups have reported single photon emission from QDs [6-9], and relatively low $g^{(2)}(0)$ values of 0.02-0.03 were reported in ref. 6, 7. However, these values have been obtained under p-shell resonant excitation which requires strict energy tuning. For practical devices driven by electrical pumping, further suppression of multi-photon generation without strict energy tuning is much preferable. Another issue is to approach the on-demand operation. Generally, photon extraction efficiency from QDs is less than 1% in the case of a planar surface due to large difference of refractive indices between semiconductors and air. In order to overcome these limitations, we propose metal-embedded semiconductor pillar structure with QDs inside. In this work, we demonstrate the fabrication of the proposed structure. The fundamental properties based on high-purity single-photon generation and photon extraction efficiency under the non-resonant excitation are evaluated.

2 Sample preparation

A schematic of the metal-embedded structure is shown in Fig. 1. A GaAs pillar structure including InAs QDs is embedded in a metal film. The typical pillar height is 250 nm, and the InAs QDs are located 50 nm above the metal-semiconductor interface. Due to the photon reflection from the metal surface, improved photon extraction efficiency to air is expected. Moreover, suppression of multi-photon generation is expected because of an extremely small semiconductor volume, which can be a source of background emission.

The sample fabrication process is illustrated in Fig. 2. The GaAs/InAs QD/GaAs heterostructures were grown by metalorganic molecular-beam epitaxy (MOMBE) on a GaAs substrate (dot density of 5×10^{9} cm^{-2}). After the pil-

lar structure fabrication with electron-beam (EB) lithography and dry etching processes, a removal layer composed of SiO_2/Au was deposited (a). Then, the whole structure was embedded in metal (b). Turned upside down, the metal surface was bonded on a supporting substrate (c). Finally, the GaAs substrate was removed (d). Thanks to the presence of the removal layer, the GaAs substrate is easily peeled off and the pillar structure is transferred to the supporting substrate.

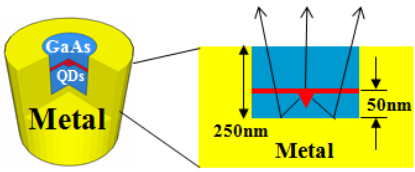

Fig. 1 Schematic of metal-embedded structure

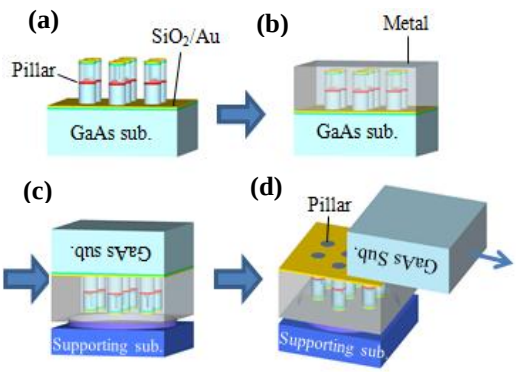

Fig. 2 Metal-embedded structure fabrication process

3 Experimental setup

As an excitation source, a mode-lock Ti:sapphire laser (energy: 1.5498eV, frequency: 82MHz) was used. The laser beam was focused the on a single pillar structure with an objective lens with a numerical aperture (NA) of 0.42. The PL spectrum was detected with a Si charge-coupled-device (CCD) camera. The second-order photon correlation function $g^{(2)}(\tau)$ was measured using a Hanbury-Brown and Twiss (HBT) setup [10] detected by single-photon counting modules (SPCM). All measurements were performed at 20K.

4 Observation of luminescence enhancement in the embedded structure

The number of QDs contributing to the luminescence varies depending on the pillar diameter. Therefore integrated photoluminescence (PL) intensity which is proportional to the total QD number in the pillar was examined. The square-root of the integrated PL intensity of two types of pillar structures is compared for various pillar diameters (Fig. 3). The red dashed line is the least-square fit to the data measured from conventional un-embedded pillar structures, while the blue dashed line is the one from metal-embedded pillar structures. Linear increase of the square-root of the integrated PL intensity is clearly observed for both the structures, however, exhibiting very different slopes. 6.4-fold increase in the slope is observed for the metal-embedded pillar structure. The PL intensity is written as follows:

$$I_{PL} = S \cdot n_{dot} \cdot \eta_{\text{int}} \cdot G \cdot C \cdot \eta_{trans}, \qquad (1)$$

where S is the pillar cross-sectional area, n_{dot} is the dot density, η_{int} is the internal quantum efficiency, G is the effective excitation rate, C is the photon extraction efficiency, and η_{trans} is the optical setup transmission efficiency. Since the square root of S is proportional to the pillar diameter, 6.4^2 ~40 times enhancement of the excitation × extraction efficiencies (GC product) assuming the identical internal quantum efficiency in the two cases.

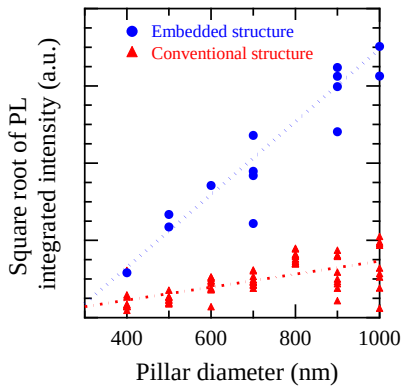

Fig. 3 Pillar diameter dependence of the square-root of integrated PL intensity. Conventional pillar structures (red) and metal-embedded pillar structures (blue) are compared.

5 Photon correlation measurements

The single photon purity and photon extraction efficiency are estimated from the photon correlation and photon counting measurements. A part of the PL spectrum measured from a single pillar in the metal-embedded structure under non-resonant excitation is shown in Fig. 4. The pillar diameter is 500nm.

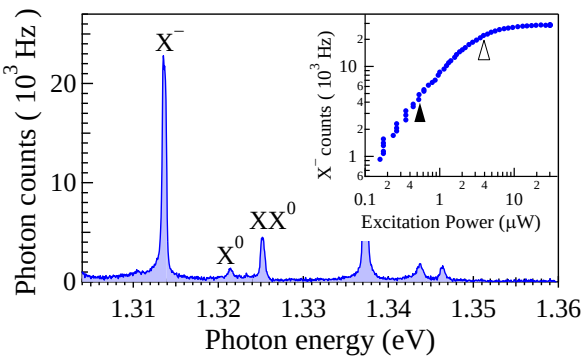

Fig. 4 PL spectrum from a single metal-embedded pillar structure with a diameter of 500nm. (inset) Excitation power dependence of the X^- emission line.

The three emission lines (1.31-1.33eV) originate from a single QD assigned as a neutral exciton (X^0), neutral biex-

citon (XX^0), and negatively charged exciton (X^-). The assignment is supported with the photon cross-correlation measurements between X^0 and XX^0 emission lines indicating bunching and the one between X^0 and X^- emission lines indicating anti-bunching. Since the X^- emission line dominates the PL spectrum, we focus on the X^- emission line hereafter for the further optical characterization. Other huge benefits of X^- emission are no fine structure splitting and dark state.

5.1 Strongly suppressed multi-photon generation

The photon auto-correlation of the X^- emission line was measured under the weak excitation power of 0.5 μW corresponding to the closed triangle in the excitation power dependence in the inset of Fig. 4, which is about 14% of the saturation level (I_{ex}/I_{max} ~ 0.14). The measured photon correlation function is displayed in Fig. 5. Very clear anti-bunching at zero time delay is observed.

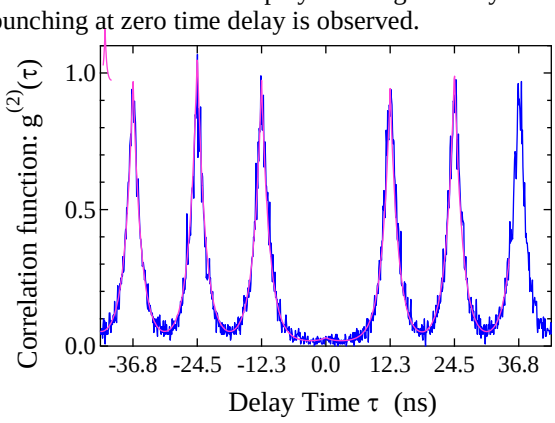

Fig. 5 2nd-order photon auto-correlation function of X^- emission line under weak excitation power (I_{ex}/I_{max} ~ 0.14). The blue line is measured data, and the pink line is the fitted curve using Eq. (2).

The slope of the correlation function is limited by the recombination lifetime, which is determined to be 1.7 ns by a time-resolved PL measurement (not shown). Background is not subtracted in Fig. 5. The measurement is very well fitted with the following equation:

$$B+\sum_n a_n \exp\left\{-\left|\tau-n/f\right|/\tau_{lifetime}\right\}, \qquad (2)$$

where B is the background, f is the excitation frequency, and $\tau_{lifetime}$ is the recombination lifetime of X^-. As a result, low $g^{(2)}(0)$ of 0.015 ± 0.005 is obtained under non-resonant excitation. This is attributed to extremely limited semiconductor volume (~0.049 μm^3 in this case of the 500-nm pillar diameter).

5.2 Photon extraction efficiency

The photon extraction efficiency from a single QD coupled to an objective lens is estimated under the higher excitation power of 4.0 μW, which is denoted by the open triangle in the inset of Fig. 4. In this case, the photon number counted by one of the SPCMs was 25,000 count per second (cps), and the obtained $g^{(2)}(0)$ increased to 0.25. Thus, the net single photon detection rate after compensating a contribution of multi-photon emission corresponds to $25{,}000\times(1-0.25)^{1/2}$ [11]. All optical losses (LPF, HPF, and BPF filter transmittance, coupling to objective lens, SPCM detection efficiency, and transmission loss) were measured or given. Thus, considering the total optical loss (24.7 dB) and the laser repetition frequency (82 MHz), the extraction efficiency was estimated to be ~7.8%. Compared with the case of an unprocessed sample (< 1%), the improvement of the photon extraction efficiency by the present metal-embedded structure is evident. This value indicates single-photon generation from a single QD at a rate of 6.4 million per second.

6 Summary

We have proposed metal-embedded semiconductor pillar structure for efficient and highly pure single-photon sources and report the realization of this structure. As a result, 40-times enhancement of the excitation and extraction efficiency product was confirmed. Single photon purity expressed by $g^{(2)}(0)$ is estimated to be as low as 0.015, even under non-resonant excitation. Single-photon generation from a single QD at a rate of 6.4 million per second is achieved with the improvement of the photon extraction efficiency up to ~7.8%.

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