

Solid-state photon generator of silica-coated quantum dot combined with metamaterial fabricated by the scanning probe microscope lithography

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Abstract

We prototyped the novel solid-state photon generator composed of silica-coated QD and metamaterial element using the scanning probe microscope lithography. From the temperature dependence of micro photoluminescence spectra characteristics, the sharp emission from the device was confirmed as the result of the resonance between QD light emission and induced current in a metamaterial element.

1. Introduction

The realization of quantum information processing devices is expected to increase the capacity and reliability of telecommunications for the development of a highly information-oriented society. One of the promising materials for the device is a quantum dot (QD). We have succeeded in changing the apparent size of QD with silica coating while maintaining the emission wavelength [1]. Research on micro-size metal structures which realize non-natural electromagnetic properties, i.e., metamaterials, has been actively done. We have proposed the novel photon generation device using silica-coated QD combined with a metamaterial element drawn by the electron beam lithography [2]. Based on the Purcell effect, due to the resonance of induced current oscillation in the metamaterial and light emission of QD, the photon generation of the device is expected to have high emission efficiency, high responsiveness, and strong directivity. In this study, the photon generating device was prototyped using the scanning probe microscope (SPM) lithography (Fig. 1). SPM usage is advantageous for position control of QD at the device fabrication [3]. We report the primal optical performance of the device.

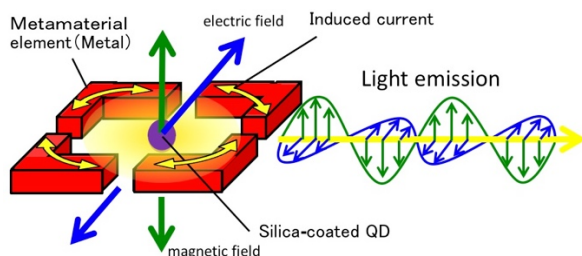


Fig. 1 Schematic of our photon generating device.

2. Experimental

The metamaterial structure for 1300-nm resonance was designed by the simulation using FDTD (finite-difference time domain) method. Silica-coated QDs were prepared by the same procedure in our previous research [1]. First, PbS QDs were synthesized with oleylamine as a ligand. A reverse micelle solution was prepared with surfactant, cyclohexane and aqueous ammonia. Then, these two were mixed and stirred while dropping TEOS (tetraethyl orthosilicate) to obtain silica-coated QDs.

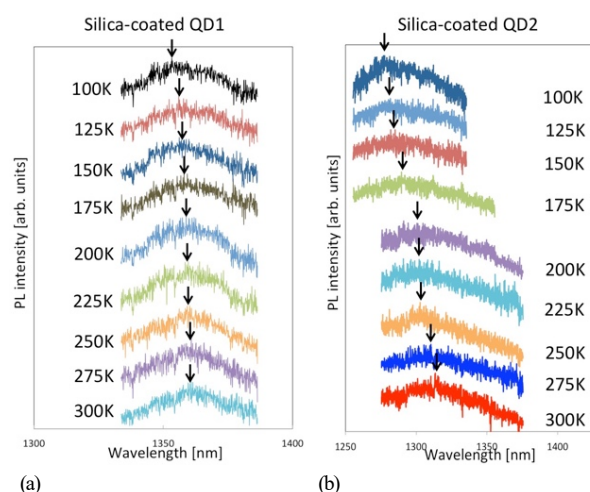


Fig. 2 Temperature dependence of PL spectra. Results of two types of QDs are compared. Peak positions are indicated by the arrows.

Figure 2 shows the temperature dependence of photoluminescence (PL) spectra of two types of silica-coated QDs. For PbS QDs, the tendency of the emission wavelength to change with temperature has been reported to be size dependent [4]. Below 4 nm, dE_g/dT is negative, and above 4 nm, dE_g/dT is positive. Here, E_g is a band gap and T is a temperature. QD1 has small dE_g/dT of -27.5 [$\mu\text{eV/K}$], and QD size is considered to be about 4 nm. QD2 has large dE_g/dT of -148 [$\mu\text{eV/K}$], and QD size is considered to be smaller than 4 nm.

The target device was manufactured. First, a mask pattern of nanohole was formed on a Si substrate by anodic oxidation using SPM, and KOH wet etching was done. Then,

after the substrate surface was covered with a self-assembled monolayer (SAM) of hexamethyldisilazane (HMDS), a silicon oxide pattern of metamaterial element was drawn by SPM and plated by nickel. Since the SAM portion is hydrophobic and the silicon oxide is hydrophilic, the oxide pattern was selectively plated. Finally, the silica-coated QDs were trapped in the nanohole.

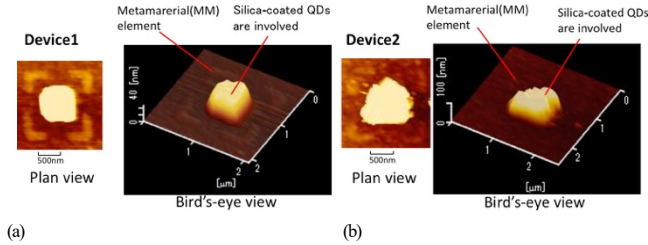


Fig. 3 SPM image of fabricated devices using (a) QD1 and (b) QD2.

3. Results and discussion

SPM images of devices fabricated using silica-coated QD1 and QD2 respectively are shown in Fig. 3. The convex portion at the center is the nanohole formation area where QDs were trapped. Around the QD area, a 4-division split ring metamaterial element was prepared. By the detailed SPM image analysis, we estimated that about 100 pieces of silica-coated QDs were included in the device. Though the final target number of QD is one, we intended in this work to confirm the resonance characteristics of QD emission.

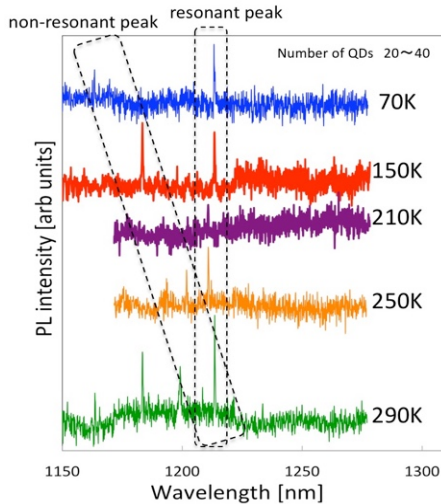


Fig. 4 Temperature dependence of μ -PL spectra of fabricated device.

The temperature changes of micro PL (μ -PL) spectra were evaluated. As a result of device 1 including QD1, a stable peak was confirmed at the wavelength of about 1140 nm at each temperature, though the emission wavelength of QD1 slightly changed with temperature (see Fig. 2(a)). Since the resonant wavelength of the metamaterial does not change with temperature, the stable emission wavelength proved the resonance in the device.

Figure 4 shows the result of μ -PL measurements of the device 2 using QD2. At each temperature, multiple sharp emission peaks were observed, which may be derived from a single QD. There were two trends of the peak position in the temperature change: one having a tendency of the emission wavelength to shift to the short wavelength side when the temperature is lowered, and one having a constant wavelength around 1210 nm regardless of the temperature. The trends of peak wavelength with temperature variation can be seen clearly in Fig. 5. The case when shifting with temperature is considered that the emission may be from the same QD at every temperature since the amount of shift is the same as in QD ensemble. On the other hand, in the case of temperature-independent peaks, the emission came from different QDs at each temperature and were enhanced by the resonance with metamaterial since the resonant wavelength of the metamaterial does not change with temperature.

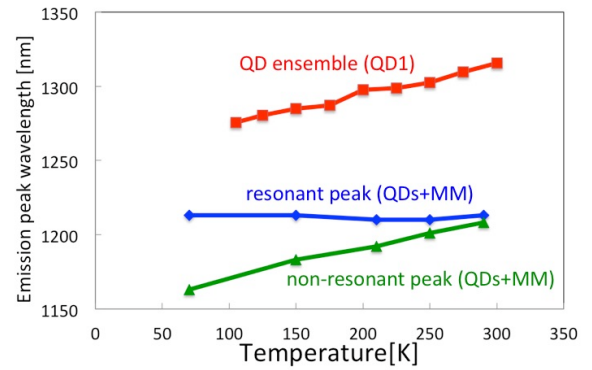


Fig. 5 Temperature dependence of peak position of the device. Characteristic of QD ensemble is compared.

4. Conclusions

Photon generating device using silica-coated PbS QDs combined with metamaterial element was fabricated using SPM lithography. Temperature dependence of μ -PL spectra suggested the device emission from a small number of QDs in resonance with the metamaterial.

Acknowledgements

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References

- [1] K. Mukai, I. Okumura, Y. Nishizaki, S. Yamashita, and K. Niwa, *Jpn. J. Appl. Phys.* **57**, 04FH01 (2018).
- [2] K. Mukai, S. Watanabe, I. Okumura, and T. Sugimoto, *Jpn. J. Appl. Phys.* **58**, SBBI06 (2019).
- [3] K. Mukai, A. Hirota, and S. Nakashima, *Jpn. J. Appl. Phys.* **54**, 04DJ02 (2015).
- [4] A. Olkhovets, R.-C. Hsu, A. Lipovskii, and F. W. Wise, *Phys. Rev. Lett.* **81**, 3539 (1998).