optica

Deterministic coupling of epitaxial semiconductor quantum dots to hyperbolic metamaterial: supplementary materials

Y. D. JANG,[1] J. S. BAEK,[1] V. DEVARAJ,[1] M. D. KIM,[1] J. D. SONG,[2] Y. WANG,[3] X. ZHANG,[3] AND D. LEE[1,*]

[1]Department of physics, Chungnam National University, Daejeon 34134, South Korea
[2]Center of Opto-Electronic Material and Devices, KIST, Seoul 02792, South Korea
[3]Nanoscale Science and Engineering Center, University of California, Berkeley, California 94720, USA
*Corresponding author: dleee@cnu.ac.kr

Published 12 July 2018

This document provides supplementary information to "Deterministic coupling of epitaxial semiconductor quantum dots to hyperbolic metamaterial, https://doi.org/10.1364/OPTICA.5.000832. In this document we provide detailed information about the optimal HMM-quantum dot distance, PL spectra from various HMM structures with a fixed distance, dispersion curves of the HMM structures, and chemical etching.

1. The optimal HMM-quantum dot (QD) distance

In order to find the optimal distance between the QDs and HMM, we have prepared QD samples of different capping layer thickness by chemical wet etching: 7.0, 16.4, 27.7, 36.1 nm, measured by atomic force microscope (AFM). After the etching, the 7.0 nm-distance QD sample shows a ~25 nm red-shift of the photoluminescence (PL) peak with significant PL intensity decrease, indicating strain relaxation around the QDs (Fig. S1); strain relaxation causes a red-shift of the PL peak in a compressively strained InAs QD. However, the 16.4, 27.7, and 36.1 nm-distance QD samples show comparable and strong PL intensity without PL peak shift, as compared to a 50 nm (no etching) distance sample, attesting no strain relaxation. The PL spectrum is measured from the top with top-side excitation.

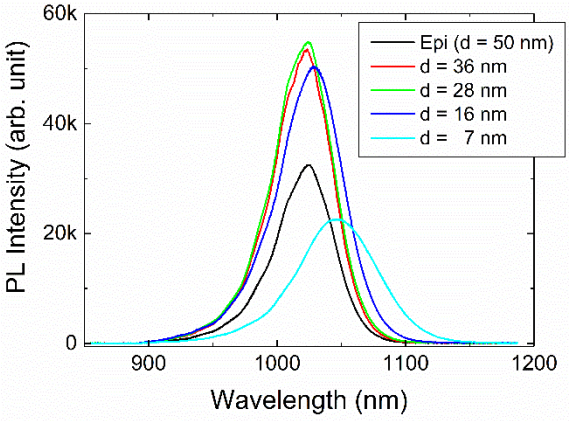

Fig. S1. PL spectra at 12 K from QD samples of various QD–HMM distance before HMM deposition.

For HMM-coupled QD structures, 15 nm-thick Ag and 10 nm-thick Ge layers are alternatively deposited for 5 periods on the etched QD sample. The PL spectrum is obtained from the GaAs substrate side with substrate-side excitation at 850 nm, since high-*k* PL modes cannot escape the HMM. In this case, the 16.4 nm sample shows PL intensity decrease, compared to the 27.7 and 36.1 nm samples, but the peak position is the same as before, suggesting no strain relaxation (Fig. S2). The absence of peak shift indicates no strain relaxation for the 18.4 nm distance sample, even after HMM deposition. The reduction in PL intensity is caused by the stronger HMM coupling of QD emission: the forward propagating signal into the HMM increases due to the stronger QD–HMM coupling, resulting in reduced uncoupled (to the HMM) QD emission to the GaAs substrate.

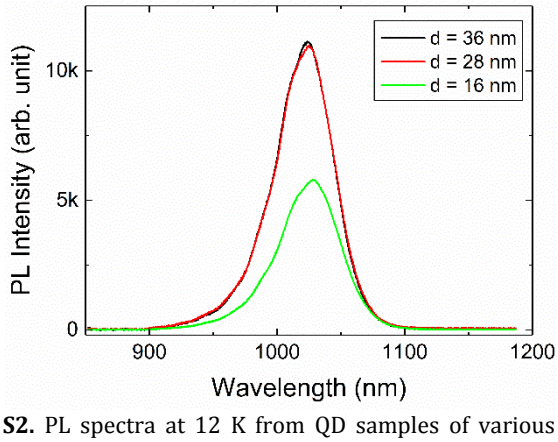

Fig. S2. PL spectra at 12 K from QD samples of various QD–HMM distance after HMM deposition. All the samples have the same HMM structure: five pairs of 15 nm Ag / 10 nm Ge layers.

Consequently, the distance of 18.4 nm, including the 2 nm Ge layer, between QDs and HMM is the right choice for this work since it is thick enough to not induce strain relaxation but thin enough to provide for strong coupling. Note the same PL spectrum for the 29.7 and 38.1 nm samples, confirming that 29.7 nm is already too far from the HMM to have significant coupling.

2. PL spectra from various HMM structures with a fixed distance

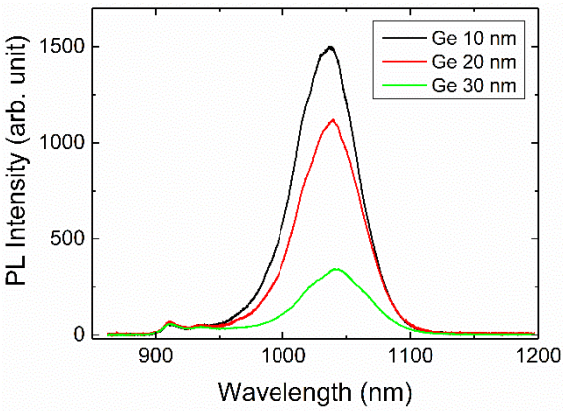

Fig. S3. PL spectra measured after HMM deposition, plotted as a function of Ge thickness.

The distance between QDs and HMM is fixed at 18.4 nm. Figure S3 shows the weaker PL intensity for the 30 nm Ge sample, measured from the substrate side. This is due to the stronger coupling of QD emission to the HMM than those for the other samples. This interpretation is confirmed by PL decay measurement (Fig. 3).

3. FDTD simulation of QD–HMM coupling

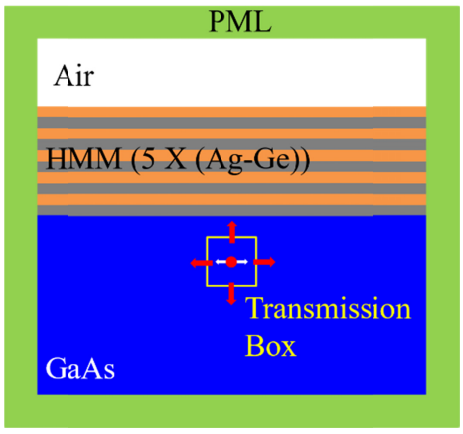

Fig. S4. Schematic for FDTD simulation.

To calculate the enhancement of the radiative recombination rate of a dipole emitter (Purcell enhancement), we have used a commercial simulation package, FDTD Solutions (Lumerical Inc.). The schematic for FDTD simulation is outlined in Fig. S4. Perfectly matched layers (PMLs) are employed to simulate the rate enhancement with open boundaries.

We have calculated the total transmitted power through a small transmission box (10 nm X 10 nm X 10 nm) containing a dipole emitter near the HMM. The transmitted power enhanced by the large density of states of the HMM is divided by that through the same size box containing a dipole in vacuum to get the Purcell enhancement.

As the QDs are located at the same distance from the HMM, we do not need the summation of different decaying components, as was done in previous reports for dye molecules in PMMA [1,2]. In the plane, although there are many QDs, wavefunction overlap between QDs is quite negligible, even for those with high dot density [3]. Thus, the QDs in this work are independent of each other, such that the Purcell enhancement of ensemble QDs is the same as that of individual QDs in the same plane.

4. Dispersion curves of the HMM structures

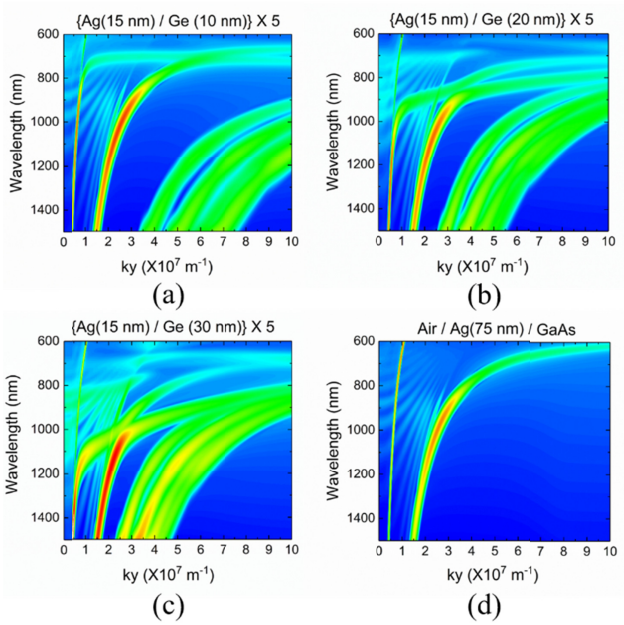

Fig. S5. Simulated dispersion curves of (a–c) HMM structures with Ge thicknesses of 10, 20, and 30 nm, respectively. Ag thickness is fixed to 15 nm. (d) Dispersion curve of surface plasmon-coupled structure with GaAs/Ag (75 nm)/air.

Dispersion curves of the HMMs are obtained by FDTD simulation and displayed in Fig. S5. High-k modes, as expected for HMMs, appear for all HMM structures (a–c), indicating the formation of HMM for all three structures. In contrast, the dispersion curve of the structure with a 75 nm-thick Ag layer only (the total Ag thickness in each HMM, Fig. S4d) shows only a surface plasmon polariton dispersion curve and, as a result, provides a limited photonic density of states at 1030 nm.

Emission rate enhancement depends largely on three factors: the number of available modes, $dk/d\omega$ for each mode, and available k depending on the distance between an emitter and HMM. Since the emitter distance of 18.4 nm is already significant, available k values should not be large, limiting ky values. The 10 nm Ge HMM sample has a large minimum ky due to corresponding hyperboloidal dispersion; consequently, contributing modes are reduced within the ky limit. In addition, $dk/d\omega$ is smaller for the 10

nm Ge HMM sample than for the 30 nm one at 1030 nm. The 30 nm Ge sample has more modes within the *ky* limit and larger $dk/d\omega$, compared to the other two samples, providing the highest rate enhancement (PF) in accordance with the calculation and experimental result (Fig. 4).

5. Chemical etching.

Wet chemical etching of the GaAs capping layer is used to control the distance between the HMM and QDs. For precise distance control, we have used a slow etchant (etch rate: 0.4 nm/s) mixed with H_3PO_4: H_2O_2: H_2O = 1 : 4 : 490. The temperature of the etchant is controlled in a water bath set at 14 °C. Etching is performed 30 minutes after mixing the chemicals with stirring. After etching, the sample is rinsed sufficiently with de-ionized water. The etch depth is determined using AFM on a photoresist patterned sample.

References

1. C. L Cortes, W. Newman, S. Molesky, and Z. Jacob, "Quantum nanophotonics using hyperbolic metamaterials", J. Opt. 14, 063001 (2012)

2. D. Lu, J. J. Kan, E. E. Fullerton, and Z. Liu, "Enhancing spontaneous emission rates of molecules using nanopatterned multilayer hyperbolic metamaterials," Nat. Nanotech. **9**, 48 (2014).

3. Y. D. Jang, E. G. Lee, J. S. Yim, D. Lee, W. G. Jeong, S. H. Pyun, and J. W. Jang, "Unambiguous observation of electronic couplings between InGaAs/InGaAsP quantum dots emitting at 1.5 μm," Appl. Phys. Lett. **88**, 091920 (2006).