Dynamical optical near-field of energy transfers among quantum dots for a nanometric optical buffering

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Abstract: We observed energy transfers among quantum dots dynamically. The energy transfer occurs from smaller to larger quantum dots in 150 ps and is applicable to the nanophotonic buffering device.

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Energy transfers among quantum dots enable several unique nanophotonic devices, such as an AND-gate [1], NOT-gate [2], an optical nanofountain [3], an optical nano content addressable memory (CAM) [4], and an optical digital to analog converter [5] and so on. Investigations of the energy transfer are essential to construct and optimize these nanophotonic devices. In this paper, we show the experimental results of time-resolved near-field spectroscopy and discuss the energy transfers among quantum dots. The experimental results have indicated the delay time coming from the energy transfer is applicable to the novel optical buffer.

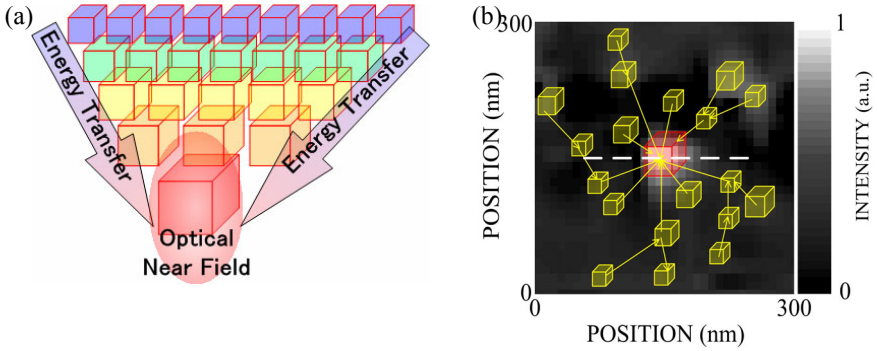

Fig.1. (a) Explanation of the optical nanofountain composing many quantum dots. (b) the two-dimensional near-field scanning image of optical nanofountain.

We have reported energy transfers among quantum dots via an optical near field [6]. The energy transfer occurs from a higher energy level to a lower energy level, *i.e.*, from smaller to larger quantum dots. This unidirectional energy transfer and successive relaxation are applicable to the optical energy concentration. We have called this device an *optical nanofountain*, because its mechanism looks like a fountain, which concentrate and spurt water caught at high potentials. Figure 1 (a) shows a schematic explanation of the optical nanofountain, which is composed of many different-sized quantum dots. When these quantum dots have resonant energy sublevels of carriers, the energy transfer occurs via the optical near field as illustrated by the arrows in Fig. 1(a). Optical energy incident to the optical nanofountain is ultimately concentrated at the largest quantum dot whose size corresponds to the focal spot size of this device. In the experiments, we used CuCl cubic quantum dots embedded in a NaCl matrix. Since this has an inhomogeneous size distribution and a random arrangement of quantum dots, in some areas of the sample, the quantum dots are arranged to act as an optical nanofountain. We maintained the sample temperature at 45 K, as at too low a temperature the resonant condition becomes tight, due to narrowing of the homogeneous linewidth, while at too high a temperature unidirectional energy transfer is obstructed by thermal activation of excitons in the quantum dots. Figure 1 (b) shows the spatial distribution of the output intensity from the optical nanofountain found from the sample with an optical near-field microscope in the collection mode operation. Here, the collected luminescence photon energy was from 3.297 eV to 3.215 eV, which corresponded to the luminescence from 3.1- to 10-nm quantum dots, respectively. The bright spot at the center in the figure

corresponds to the spurt from the optical nanofountain.

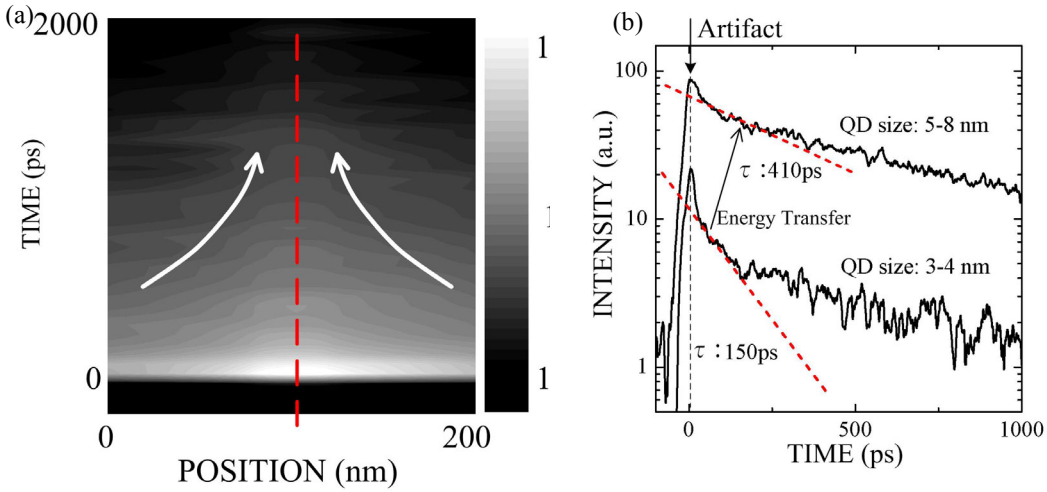

Fig.2. (a)Time evolution of output intensity from the optical nanofountain observed along with the white broken line in Fig.1 (b). (b) Time evolution of output intensity from the optical nanofountain at the spouting spot, *i.e.*, the red broken line.

Figure 2 (a) shows the time evolution of output intensities from the optical nanofountain observed at each position along with the white broken line in Fig.1 (b). Here we used the SHG of mode-locked Ti: sapphire laser (λ=365 nm: pulse duration=2ps: repetition rate=80MHz) as a light source and a micro-channel plate as a photo-detector of the time correlation single photon counting method with a 20-ps time resolution. The signal decay time at the center position (the red broken line) was slower than that of the other positions. This experimental result shows the time-evolution of the energy transfer to the spouting spot of the optical nanofountain, as shown by the two arrows.

Figure 2 (b) shows the time evolution of output intensity from the optical nanofountain at the spouting spot. AT the time origin, the small artifact of the pump pulse was observed and the signal quickly decayed. The radiative life time of quantum dots was proportional to their volume (for example τ=2ns for 3.5-nm quantum dots)[7]. However, the intensity of the smaller quantum dots (QD size: 3-4 nm) decayed faster than the larger quantum dots (QD size: 5-8 nm) and their decay time was 150 ps and 410 ps, respectively, because the excitation energy in the smaller quantum dot transfers to the larger neighboring quantum dot. After 300 ps, both signal decayed with the relaxation time of 700 ps. This relaxation time agrees with the radiative lifetime of the 7-nm CuCl quantum dot [7], which was larger than mean quantum-dot size of the used sample (mean size: 4-nm). Namely, the energy transfers to larger quantum dots accelerate radiative recombination.

These time-resolved experimental results are reasonable to previous works [1-6]. Additionally, we can propose a new nanophotonic device, which is the nanometric optical buffer by using the long recombination lifetime of smaller quantum dot. For example, when 2-nm CuCl quantum dots are used as excitation energy buffers, we preserve the signal for 10 ns. Such long time optical buffering function in nanometric region is only realized by the nanophotonic device using energy transfer and is very important for the nanophotonic system.

[1] T. Kawazoe, K. Kobayashi, S. Sangu, and M. Ohtsu, Appl. Phys. Lett., **82**, 2957 (2003).
[2] T. Kawazoe, K. Kobayashi, and M. Ohtsu, CLEO 2005, CTuF4, May 24 - 26, 2005 in Baltimore, USA.
[3] T. Kawazoe, K. Kobayashi, and M. Ohtsu, Appl. Phys. Lett., **86**(10), 103102 (2005).
[4] M. Naruse, T. Miyazaki, F. Kubota, T. Kawazoe, K. Kobayashi, S. Sangu, M. Ohtsu, Optics Letters, **30**, 201 (2005).
[5] M. Naruse, T. Miyazaki, T. Kawazoe, K. Kobayashi, S. Sangu, F. Kubota, M. Ohtsu, IEICE Trans. on Electron., Vol. E88-C, 1817 (2005).
[6] T. Kawazoe, K. Kobayashi, J. Lim, Y. Narita, and M. Ohtsu, Phys. Rev. Lett. **88**, 067404 (2002).
[7] T. Itoh, T. Ikehara, and Y. Iwabuchi, J. Lumin. **45**, 29 (1990).